SPECIAL REPORT

HEALTH EFFECTS INSTITUTE

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# Research Directions to Improve Estimates of Human Exposure and Risk from Diesel Exhaust

A Special Report of the Institute's Diesel Epidemiology Working Group

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The Health Effects Institute, established in 1980, is an independent and unbiased source of information on the health effects of motor vehicle emissions. HEI supports research on all major pollutants, including regulated pollutants (such as carbon monoxide, ozone, nitrogen dioxide, and particulate matter) and unregulated pollutants (such as diesel engine exhaust, methanol, and aldehydes). To date, HEI has supported more than 200 projects at institutions in North America and Europe and has published over 100 research reports. Consistent with its mission to serve as an independent source of information on the health effects of motor vehicle pollutants, the Institute also engages in special review and evaluation activities.

Typically, HEI receives half its funds from the US Environmental Protection Agency and half from 28 manufacturers and marketers of motor vehicles and engines in the United States. Additional funds for the diesel studies reported here were provided by the California Air Resources Board, Engine Manufacturers Association, and American Petroleum Institute. Regardless of funding sources, HEI exercises complete autonomy in setting its research priorities and in reaching its conclusions. An independent Board of Directors governs HEI. The Institute's Health Research and Review Committees serve complementary scientific purposes and draw distinguished scientists as members. The results of HEI-funded research and evaluations have been used in public and private decision making.

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# EXECUTIVE SUMMARY

## INTRODUCTION

Diesel engines are used extensively in transportation, especially in heavy-duty applications due to their power, durability, and efficiency. They are more efficient than gasoline engines and emit less carbon dioxide (a greenhouse gas) per unit of work, thus having advantages over gasoline engines in addressing global warming, an issue with increasing importance as the number of vehicle-miles-traveled increases rapidly around the world. Engine manufacturers have significantly lowered the emissions of particles in diesel exhaust over the last decade by improving engine and emission-control technologies. In spite of the many positive attributes of diesel engines, concerns remain about two emissions: nitrogen oxides, which contribute to ozone formation, and particulate matter, which is associated with adverse acute and chronic health effects.

Several state, national, and international agencies have concluded that diesel exhaust is a probable lung carcinogen (see Table 1 in the Diesel Epidemiology Working Group Report). This conclusion implies important public health consequences for the many urban areas (and possibly some rural areas) where diesel vehicles are prevalent and for workers exposed to diesel exhaust in their jobs. Animal models have proven inadequate for obtaining quantitative estimates of the cancer risk diesel emissions pose to the general population. As a result, epidemiologic data from studies of diesel-exposed workers have been the starting point for developing risk models. Numerous studies of diverse design have examined the association between lung cancer risk and diesel exhaust exposure. The difficulty of accurately estimating occupational exposure has limited interpretation of the resulting evidence and its utility for quantitative risk assessment.

In 1998, HEI initiated its Diesel Epidemiology Project, a multifaceted research and assessment effort to (1) develop new research initiatives, including feasibility studies to identify potential new cohorts or to improve exposure assessment methods, and (2) evaluate the strengths and limitations of the published epidemiologic studies available for quantitative risk assessment. As a part of the project, HEI appointed the Diesel Epidemiology Expert Panel to conduct a systematic review of these studies. The Expert Panel reported its findings and recommendations in the 1999 HEI Special Report *Diesel Emissions and Lung Cancer: Epidemiology and Quantitative Risk Assessment*. They concluded that enhanced exposure and epidemiologic data were needed for quantitative risk assessment and that these data might come from further exploration of existing studies or from new studies. The Expert Panel recommended that HEI consider undertaking a new epidemiologic study after HEI's feasibility studies were completed and evaluated and after attempts to retrofit improved exposure assessments to existing studies were evaluated for their ability to support quantitative risk assessment.

The Diesel Epidemiology Working Group was formed in the fall of 2000 to

- review reports from 6 diesel feasibility studies funded by HEI to provide information on potential study populations and on exposure assessment methods; and
- consider the results of the feasibility studies and other ongoing research in order to develop a new research agenda to seek better information for quantitative risk assessment of lung cancer and other chronic diseases that may result from exposure to diesel exhaust.

In considering the expectations for a new study, the Working Group recognized that policy makers need risk estimates that are more precise and more certain than those that can be generated from the data now available. Issues related to exposure assessment arise both in designing an epidemiologic study to characterize health effects associated with exposure and in estimating the magnitude of the risk for various populations. In both contexts, characterization of exposure to

diesel exhaust must be sufficiently specific to distinguish its contributions to health effects from those of other sources.

Not only is diesel exhaust a complex mixture of gases and particles, but the particles themselves are a complex mixture; the characteristics depend on many factors including the diesel fuel formulation, combustion conditions in the engine, emission-control technologies, and conditions in the ambient environment into which the exhaust is emitted. Approaches to exposure assessment need to discriminate diesel particles from the many other sources of particles in the complex mixture of particles in air. Current approaches are somewhat nonspecific, using measurements both of particle mass and of marker compounds, such as elemental carbon, that have sources other than diesel emissions. In an epidemiologic study, exposures of participants need to be estimated with as little misclassification as possible. For population exposure assessment, information is needed on typical or average exposures and on the upper tail of the exposure distribution, which carries the greatest risks.

## TECHNICAL EVALUATION OF DIESEL FEASIBILITY STUDIES

The 6 feasibility studies described in this report were funded by HEI to provide insight about, first, whether a new retrospective or prospective epidemiologic study could provide data to improve estimates of cancer risk from exposure to diesel exhaust and, second, about whether new methods of exposure analysis would allow us to reevaluate older epidemiologic studies.

The Working Group evaluated the following reports on possible new cohorts for investigation:

- *Cancer Risk from Diesel Emissions Exposure in Central and Eastern Europe: A Feasibility Study*, Paolo Boffeta and colleagues, International Agency for Research on Cancer, Lyon, France
- *Cancer Risk from Diesel Exhaust Exposure in the Canadian Railroad Industry: A Feasibility Study*, Murray Finkelstein and Dave Verma, McMaster University, Hamilton, Ontario, Canada
- *Quantitative Assessment of Lung Cancer Risk from Diesel Exhaust Exposure in the US Trucking Industry: A Feasibility Study*, Eric Garshick and colleagues, Channing Laboratory, Brigham and Women's Hospital, Boston MA, USA

These well-executed studies evaluated the following cohorts: (1) a diverse set of historical cohorts of workers exposed to diesel emissions, including miners and bus or railroad transport company workers, in Russia and several Eastern European countries; (2) Canadian railroad workers; and (3) US truckers. The results of the studies illustrated the difficulties epidemiologic researchers face in finding suitable populations for study—populations with (a) adequate exposure data available, (b) exposures in a range likely to be informative and relevant to ambient levels, (c) data available on potential confounding and modifying factors, and (d) offering sufficiently large sample sizes. Each of the cohort studies had strengths and limitations, but for all of them accuracy of exposure assessment was a problem. The Working Group believes that full studies of these cohorts would probably add to the evidence regarding the carcinogenicity of diesel exhaust. However, it concluded that such studies could not be justified for the purpose of generating substantially accurate exposure-response relations, which the Working Group believes should be the goal of future epidemiologic studies of diesel exhaust.

The Working Group evaluated the following exposure assessment reports:

- *Measurement of Diesel Aerosol Exposure: A Feasibility Study,* David Kittelson and colleagues, University of Minnesota, Minneapolis, Minnesota
- *Measuring Diesel Emissions Exposure in Underground Mines: A Feasibility Study*, Barbara Zielinska and colleagues, Desert Research Institute, Reno, Nevada
- *Real-World Particulate Matter and Gaseous Emissions from Motor Vehicles in a Highway Tunnel*, Alan Gertler and colleagues, Desert Research Institute, Reno, Nevada

The feasibility studies addressing methods for exposure assessment and characterization of diesel particles are complementary to the proposals of new populations to be investigated

because improved exposure assessment methods are needed to reduce misclassification of exposure and enhance precision in epidemiologic studies. The findings confirm that we still lack sufficiently specific methods for characterizing exposures to diesel exhaust. The Working Group agrees with the investigators that elemental carbon may be a useful indicator of occupational exposure to diesel exhaust in settings where diesel exhaust is the dominant source of particulate emissions. However, additional surrogates should be explored because elemental carbon as the only marker lacks the sensitivity and specificity necessary as a signature for ambient exposure, which includes elemental carbon from other combustion sources. Improved surrogates for diesel exhaust exposure might be used to enhance exposure assessment for past studies, to strengthen future epidemiologic studies, and to assess population exposure.

The Working Group also found that considerations of statistical power in the feasibility studies did not address quantification of risk to the degree of precision necessary for quantitative risk assessment. The calculations also did not acknowledge the uncertainty that arises from using estimated exposure values.

## RESEARCH RECOMMENDATIONS

Because many agencies have concluded that diesel particles are a probable human carcinogen, the emphasis in risk assessment has shifted to quantifying the burden of lung cancer and other diseases associated with exposure to diesel emissions. This shift requires quantitative estimates of the relation between disease risk and the level and conditions of exposure. Epidemiologic studies of diesel-exposed populations can provide the risk estimates with the needed precision only if (a) populations with exposures relevant to the general population are studied, (b) exposures are accurately estimated, (c) data are available on potential confounding factors, and (d) sufficient data are accrued during follow-up. Thus, there are serious problems in improving knowledge regarding both exposure and response to diesel emissions. Nonetheless, the insights from these feasibility studies suggest areas for future investigation.

The Working Group does not recommend initiating a new cohort study of any of the proposed populations at this time; the high probable cost and continuing methodologic and data challenges make it difficult to predict that such a study would substantially improve quantitative estimates of risk. To meet these challenges, the Working Group has identified activities that could enhance risk assessments over the next 1 to 2 years (short-term activities), 3 to 10 years (medium-term activities), and 10 to 20 years (long-term activities).

### SHORT-TERM ACTIVITIES

Proposed short-term activities largely relate to improving exposure assessment, including the possibility of defining a signature that is characteristic of diesel emissions. Additional recommendations address certain design issues that would enhance any new epidemiologic studies. The Working Group proposes the following specific activities related to exposure assessment:

- Convening a workshop to plan approaches for exploring emissions and monitoring data to identify signatures of diesel exhaust that would enable measurement in ambient exposures. Contemporary techniques for data exploration could be applied to the substantial exposure data sets in existence or under development by HEI and other organizations. An anticipated product would be a list of available data and potentially informative approaches for analysis. HEI intends to follow the workshop with a research initiative to conduct analyses of existing and emerging data sets to identify a diesel signature.
- Characterizing diesel particles in HEI studies in a standard way in both laboratory and population contexts.
- Taking advantage of the US Environmental Protection Agency's Supersites and speciation sites by additional targeted data collection and analyses to develop a regional characterization of organic compounds associated with particulate matter.
- With full consideration of uncertainty, calculating sample sizes needed for future epidemiologic studies to characterize exposure-risk relations. The Working Group suggests that HEI take the lead in developing these calculations, a task to be accomplished using simulation methods or other approaches.

### MEDIUM-TERM ACTIVITIES

The Working Group's recommendations for activities that would produce results in 3 to 10 years are based in part on the likely existence of new epidemiologic studies and anticipated advances in understanding the molecular and cellular biology of lung cancer. They also build on the results anticipated from the proposed short-term activities. The Working Group recommends:

- Continuing to evaluate opportunities to enhance exposure assessment approaches, including, for example, analysis of filters from special monitoring initiatives (eg, Supersites and the speciation network) and single-particle analysis based on mass spectroscopy.
- Evaluating opportunities to apply new approaches for assessing diesel signatures in epidemiologic studies in progress.
- Assessing possible biomarkers for lung cancers related to diesel emissions exposure.
- Conducting further research directed at effects of diesel exhaust exposure other than cancer.

### LONG-TERM ACTIVITIES

In making recommendations for activities that will produce results beyond 10 years, the Working Group assumes that quantitative risk assessment will remain central in managing potential risks of diesel emissions. The Working Group recommends:

- Designing and implementing population surveillance instruments for exposure to diesel exhaust using validated source signatures. Surveillance strategies might also include modeling ambient air data and biomarkers.
- Periodically performing source-signature characterization to track changes in emissions as engine and emission-control technologies and fuels evolve. This might be accomplished with periodic tunnel studies or laboratory studies.
- Undertaking further laboratory and population-based studies on risk of lung cancer or other diseases, possibly using biomarkers of dose and exploring gene-environment interactions.

Although the Working Group does not recommend undertaking a major new epidemiologic study at present, it may be appropriate to implement a new study as research methods evolve for both exposure and outcome assessment. The Working Group does not foresee a waning of interest in the possible adverse effects of diesel exhaust and their quantitative relations with exposure. Methodologic challenges to conducting such a study include obtaining complete and accurate exposure assessment, identifying an exposure marker or set of markers that is a specific signature for diesel exhaust (and likely to reflect diesel emissions in the future), maintaining sufficiently complete follow-up over a 25-year period, and measuring other critical exposures and confounding factors. Diesel emissions are being reduced and modified as a result of changes in engine and emission-control technologies; consequently, it is not clear that the exposures and outcomes measured in a prospective study started today would have relevance for the exposures and health effects present 25 years from today. Over time, we anticipate improved methods for exposure assessment and for use of biomarkers, a focus of our proposed short-term and medium-term research agendas. With better research tools, it may become feasible to carry out more informative epidemiologic studies in the future, although future researchers may still face the challenges of continuously changing technology and emissions.

## SUMMARY

As emphasis has shifted toward quantifying the potential burden of lung cancer associated with diesel exhaust, HEI has evaluated the available epidemiologic evidence and sought ways to enhance these data. It has convened expert panels and carried out workshops for this purpose. The feasibility studies discussed in this report were one outcome of this process. The findings of these studies contribute to our understanding of the potential for epidemiologic research to quantify disease risks associated with diesel emissions. On the basis of reviewing these studies, the Working Group has recommended a series of short-term, medium-term, and long-term research activities. The Working Group does not recommend proceeding with full studies of the populations considered in the feasibility studies, largely because of concern about the ability of investigators to estimate past exposures with sufficient certainty.

Because concern for the public health consequences of diesel exposure is likely to continue, HEI might consider establishing populations for prospective observation so that exposure assessment with sufficient validity could be implemented. The activities recommended by the Working Group should enhance epidemiologic studies in progress and planning for future research.

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# PART I

# Report of the Diesel Epidemiology Working Group

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# Section 1: Scientific, Technological, and Regulatory Background

## INTRODUCTION

### BACKGROUND

Diesel engines are used extensively in transportation and industry, especially in heavy-duty applications such as trucks, buses, construction and farm equipment, locomotives, and ships, because of their power and durability. Diesel engines have many advantages over gasoline engines, including better fuel efficiency and lower emissions of some air pollutants (carbon monoxide [CO*] and hydrocarbons). Because they are more efficient than gasoline engines, diesel engines emit less carbon dioxide ($CO_2$, a greenhouse gas) per unit of work, and thus have advantages over gasoline engines for addressing global warming, an issue with increasing importance as the number of vehicle miles traveled increases rapidly around the world. In Europe, more than 50% of the new light-duty passenger vehicles are powered by diesel engines. In the United States, the percentage is much lower, but increased use of light-duty diesel engines has been proposed as part of the nation's energy conservation and climate change strategies. However, in addition to the many positive attributes of diesel engines, there are concerns about their emission of high levels of oxides of nitrogen ($NO_x$), which contribute to ozone formation, and of particulate matter (PM), which is associated with adverse acute and chronic health effects. Engine manufacturers have worked to reduce these emissions with improved engine technology and emission-control technology, and have succeeded in lowering the emissions of diesel particles significantly over the last decade. Nonetheless, with the growing use of diesel engines, concerns remain about health effects of exposure to diesel particles.

*A list of abbreviations and other terms appears on page 54.

Part I of this report is one portion of Health Effects Institute Special Report *Research Directions to Improve Estimates of Human Exposure and Risk from Diesel Exhaust*, which also includes five Investigators' Reports and an Executive Summary.

Although this document was produced with partial funding by the United States Environmental Protection Agency under Assistance Award R82811201 to the Health Effects Institute, it has not been subjected to the Agency's peer and administrative review and therefore may not necessarily reflect the views of the Agency, and no official endorsement by it should be inferred. The contents of this document also have not been reviewed by private party institutions, including those that support the Health Effects Institute; therefore, it may not reflect the views or policies of these parties, and no endorsement by them should be inferred.

The relationship between diesel exhaust exposure and risk of lung cancer has been a public health concern for several decades. Initially, the focus was on occupational exposures among transportation workers, underground miners, and others. However, with the energy crisis of the early 1970s use of diesel-powered vehicles for transport increased, and the focus broadened to include potential risks to the general population in urban areas with heavy vehicle traffic. More recently, there has been concern that exposure to diesel exhaust also may enhance allergic sensitization and cause or exacerbate asthma.

Recent evidence that current levels of particulate air pollution are associated with excess morbidity and mortality has furthered concern about the risk of diesel exhaust. In urban areas, direct emissions of PM from diesel engines represent about 10% of the mass of ambient particles; secondary particles (primarily nitrates and sulfates) from diesel engines can contribute another 5%. At roadside locations near high diesel traffic, diesel contributions to ambient PM can be even greater (see Lloyd and Cackette 2001).

Diesel exhaust has several characteristics that support the plausibility of the hypothesis that it increases lung cancer risk. The particles are small enough to penetrate deeply into the lung; they contain carcinogens, including polynuclear aromatic hydrocarbons (PAHs); and extracts from particles have proved mutagenic in short-term assays. The carcinogenicity of diesel exhaust has been investigated over the last several decades using both epidemiologic and toxicologic approaches. Inhaled diesel exhaust has been shown to induce lung tumors in rats (reviewed in HEI 1995; Mauderly 2000), but the relevance of these findings to respiratory carcinogenesis in humans is not clear, especially since similar carcinogenicity has not been found in mice and Syrian hamsters (HEI 1995; Mauderly 2000). Furthermore, the doses used in the rat studies were at levels associated with the "lung overload" phenomenon, in which high doses of diesel PM overwhelm the clearance mechanisms of the lung and particles accumulate. At these high doses, some types of particles that are considered to be noncarcinogenic also seem to cause lung cancer in rats (HEI 1995; Mauderly 2000), with equal potency to diesel exhaust, so the carcinogenicity may be a consequence of the

non-specific phenomenon of lung overload. Because of these uncertainties in the animal evidence, epidemiologic studies of occupationally exposed groups have been the principal source of data on the lung cancer risk to humans posed by diesel exhaust. Numerous studies of diverse worker populations have been conducted, using either case-control or cohort approaches (reviewed in HEI 1995; Mauderly 2000). Although these studies have, in general, found a 20% to 40% increase in lung cancer risk for exposed workers, questions continue about each of these studies, and few of them provide detailed characterization of the diesel exposures (HEI 1995).

Environmental policy discussions around diesel use continue to be driven largely by concern about lung cancer risk. Several international and national health agencies have reviewed relevant epidemiologic and experimental studies of diesel engine exhaust and lung cancer, and have classified (or proposed to classify) the exhaust mixture (or the particulate component of the mixture) as a potential or probable human carcinogen. Their conclusions about the carcinogenicity of diesel exhaust are summarized in Table 1. The current consensus is that diesel exhaust is probably carcinogenic in humans.

Risk models based on epidemiologic data from exposed workers in the limited number of studies that have exposure data have been used to estimate the population burden of cancer risk from diesel exhaust. However, numerous limitations and associated uncertainties are inherent in the epidemiologic findings, including (1) the difficulty of accurately estimating exposures across the full working experiences of the study participants, (2) short or incomplete follow-up, (3) inability to measure and control for possible confounding factors, and (4) insufficient sample sizes (HEI 1999a). In addition, only a few studies have attempted to provide the exposure data needed for characterization of the dose-response relationship between occupational exposures to diesel exhaust and lung cancer risk, and none of these provide adequate information to give a high degree of confidence (HEI 1999a).

Given the widespread use of diesel engines for transportation and in many different industries, designating diesel exhaust as a probable lung carcinogen has important public health implications for the many urban areas (and possibly some rural areas) where diesel vehicles are prevalent, and for workers exposed to diesel exhaust in their jobs. Within cities, ambient diesel exhaust concentrations can be assumed to be heterogeneous, with elevated concentrations following patterns of heavy road traffic and sources such as bus or truck depots. This variation in ambient concentrations has led to concern that some populations, particularly disadvantaged and minority communities, may be exposed to diesel exhaust at higher levels and suffer health inequities as a result. Projections that use of diesel vehicles will increase because they are more fuel-efficient than gasoline-powered vehicles, with overall lower emissions of $CO_2$, have increased concern.

Recently, a new public health concern has been raised by experimental studies, both in vitro and in vivo, showing that diesel exhaust particles can increase allergic responses and inflammation. Cases of asthma and other allergic diseases have risen in prevalence around the world, especially among inner-city children. Scientists have advanced the hypothesis that diesel exhaust particles exacerbate asthma in this population of asthmatic individuals. The Health Effects Institute issued Request for Applications (RFA) 00-01, *Effects of Diesel Exhaust and Other Particles on the Exacerbation of Asthma and Other Allergic Diseases* (HEI 2000), to explore this hypothesis by funding relevant research, and began funding this research in early 2002.

## HEI INITIATIVES

The Health Effects Institute has had a sustained interest in research on diesel exhaust almost since its founding. Its initial diesel research program, begun in 1983, focused on the potential carcinogenicity of mutagenic nitropyrene compounds adsorbed on diesel particles (Beland 1986, 1991, 1995; Bond et al 1986; King 1988; Maher et al 1988; Wolff et al 1988; Jeffrey et al 1990; Moon et al 1990; Maher et al 1993; Grosovsky et al 1999). Other research included several efforts to identify biomarkers of diesel exposure (Groopman 1987; Beland 1989; Schenker et al 1990; Giese and Vouros 1993; El-Bayoumy et al 1994). Research carried out by HEI-funded investigators has also addressed how the chemical characteristics and mutagenicity of diesel particles were affected by emission controls and by different fuels (Bagley et al 1987, 1993). In the mid-1980s, HEI funded an important bioassay to try to determine whether particles or adsorbed organics were responsible for diesel exhaust's carcinogenic effects in rats (Mauderly et al 1994; Belinsky et al 1995; Randerath et al 1995).

In 1995, HEI published a report that reviewed research on the health effects of exposure to diesel emissions, and what was known, not known, and uncertain about the risks of such exposure (HEI 1995). In 1998, HEI initiated its Diesel Epidemiology Project, with the following objectives: (1) development of new research initiatives, including feasibility studies to identify potential new cohorts or improved exposure assessment methods; (2) evaluation by an expert panel of the strengths and limitations of the published epidemiologic studies available for

**Table 1.** Agency Findings on the Carcinogenicity of Diesel Exhaust

| Reference | Findings |
|---|---|
| National Institute for Occupational Safety and Health 1988 | Animal evidence "confirmatory" for carcinogenesis<br>Human evidence "limited"<br>Diesel exhaust classified as "potential occupational carcinogen"<br>No quantitative risk assessment |
| International Agency for Research on Cancer 1989 | Rat data "sufficient" for carcinogenicity<br>Human epidemiologic data "limited"<br>Diesel exhaust considered a "probable" human carcinogen<br>No quantitative risk assessment |
| World Health Organization 1996 | Rat data support carcinogenicity<br>Human epidemiologic data suggest "probably carcinogenic"<br>Epidemiologic studies considered "inadequate for a quantitative estimate of human risk"<br>Rat data used for quantitative risk assessment |
| California Environmental Protection Agency 1998 | Rat data "have demonstrated" carcinogenicity of diesel exhaust<br>Causal association of diesel exhaust and lung cancer in epidemiologic studies is a "reasonable and likely explanation"<br>Human epidemiologic data preferred for quantitative risk assessment because of uncertainties in rat data<br>California Air Resources Board designated diesel particulate matter as a "toxic air contaminant" (development of unit risk based on Garshick et al [1988]) |
| National Toxicology Program 2000 | Diesel exhaust particulates listed as "reasonably anticipated to be a human carcinogen" on the basis of elevated lung cancer occurrences in occupational groups exposed to diesel exhaust and supporting animal and mechanistic studies<br>No quantitative risk assessment |
| Environmental Protection Agency 2000 | Rat experiments provide "extensive evidence" for carcinogenicity<br>Other supporting evidence<br>Human epidemiologic studies strong, but less than sufficient for definitive conclusion<br>Diesel emissions considered "likely to be carcinogenic to humans"<br>Range of cancer risk estimates developed (on the basis of animal, epidemiologic, and comparative potency data)<br>No quantitative risk assessment |

quantitative risk assessment (QRA) and discrepancies in their reported exposure-response findings (HEI 1999a); and (3) convening a workshop (held in Atlanta, April 1999) to develop a research strategy to improve risk assessment (HEI 1999b).

To address the first objective, HEI issued RFA 98-3, *Epidemiologic Investigations of Human Populations Exposed to Diesel Engine Emissions: Feasibility Studies,* in May 1998 (HEI 1998). This RFA solicited applications for short-term feasibility studies designed to provide information on potential study populations and on exposure assessment methods. The results of the RFA were intended to guide HEI in considering whether to undertake a major epidemiologic study, particularly if a promising approach and population were identified. Investigators were asked to respond to one of the following objectives in the RFA:

- Identify study populations exposed to diesel emissions that could provide reasonably precise estimates of small to moderate excesses of lung cancer or chronic nonmalignant cardiorespiratory disease.
- Develop an exposure assessment strategy that includes (a) validation of measurement techniques and/or (b) development of approaches for using data generated by these techniques to make quantitative estimates of retrospective exposure to diesel engine emissions.

After evaluation by an expert panel and discussion by the HEI Research Committee, HEI funded 6 feasibility studies in response to this RFA (Table 2). These studies were completed in the fall of 2000, and the Diesel Epidemiology Working Group was convened to review the findings of these studies and to make recommendations with

**Table 2.** HEI Diesel Feasibility Studies Funded Under RFA 98-3

| | |
|---|---|
| **Cohort Studies** | |
| *Cancer Risk from Diesel Emissions Exposure in Central and Eastern Europe: A Feasibility Study* | Paolo Boffetta, John Cherrie, Graeme Hughson, and Alexandre Pitard<br>International Agency for Research on Cancer, Lyon, France |
| *Cancer Risk from Diesel Exhaust Exposure in the Canadian Railroad Industry: A Feasibility Study* | Murray M Finkelstein and Dave K Verma<br>McMaster University, Hamilton, Ontario, Canada |
| *Quantitative Assessment of Lung Cancer Risk from Diesel Exhaust Exposure in the US Trucking Industry: A Feasibility Study* | Eric Garshick, Thomas J Smith, and Francine Laden<br>Channing Laboratory, Brigham and Women's Hospital, Boston MA |
| **Exposure Studies** | |
| *Measurement of Diesel Aerosol Exposure: A Feasibility Study* | David B Kittelson, Winthrop Watts, Gurumurthy Ramachandran, Dwane Paulsen, and Charles Kreager<br>University of Minnesota, Minneapolis MN |
| *Measuring Diesel Emissions Exposure in Underground Mines: A Feasibility Study* | Barbara Zielinska, John C Sagebiel, Jake McDonald, C Fred Rogers, Eric Fujita, Pierre Mousset-Jones, and James E Woodrow<br>Desert Research Institute, Reno NV |
| *Real-World Particulate Matter and Gaseous Emissions from Motor Vehicles in a Highway Tunnel* | Alan J Gertler, John A Gillies, William R Pierson, C Fred Rogers, John C Sagebiel, Mahmoud Abu-Allaban, William Coulombe, Leland Tarnay, and Thomas A Cahill<br>Desert Research Institute, Reno NV |

regard to future research to improve information for QRA, including the possibility of a new epidemiologic study of groups exposed to diesel exhaust. This report provides the Working Group's evaluation of these studies, 3 of which were designed to evaluate potential study cohorts (Boffetta et al, Finkelstein and Verma, and Garshick et al, this volume), and 3 of which presented exposure assessment strategies (Kittelson et al, this volume; Zielinska et al, this volume; Gertler et al 2002).†

This evaluation addresses not only the scientific merits of the 6 feasibility studies, but also the high-priority areas for future contributions to our understanding of the risks of inhaling diesel exhaust. We suggest further methodologic work that is needed to guide the development of any additional studies. The scope of our recommendations extends beyond epidemiologic issues to cover approaches to exposure assessment that would also advance understanding of potential risks to populations from diesel exhaust exposure.

† Five of the six feasibility studies are published in this volume. The sixth feasibility study, by Dr Gertler and colleagues, *Real-World Particulate Matter and Gaseous Emissions from Motor Vehicles in a Highway Tunnel*, is published in HEI Research Report 107.

The Working Group reviewed the feasibility studies in the context of current evaluations by state, national, and international health agencies of the carcinogenicity of diesel exhaust. The converging conclusions of these evaluations—that diesel exhaust is a probable cause of human lung cancer—compelled the Working Group to consider studies quantifying this risk as having the highest priority and to make recommendations with regard to further research accordingly.

## QUANTITATIVE RISK ASSESSMENT FOR DIESEL EMISSIONS

### MAJOR ISSUES

Risk assessment has been integral to efforts to determine the threat diesel exhaust poses to public health, and it is almost certain to remain a key tool for guiding regulatory decisions. What is needed now is QRA that is sufficiently precise to serve as a basis for public health action. Several broad issues need to be addressed to estimate the nature and extent of health consequences of exposure to diesel exhaust as a basis for supporting risk management.

One set of issues in risk assessment concerns the nature of the relevant outcome or outcomes, which outcomes to evaluate, and the physical and biological processes that underlie the exposure-risk relationship. Each health outcome raises its own set of scientific and technical issues in risk assessment. For diesel exhaust, toxicologic and epidemiologic research indicates that lung cancer as well as respiratory and allergic diseases are of concern.

A second set of issues concerns how to organize and deal with the data bearing on estimation of the risk. Often such data are extensive, highly complex (originating from multiple scientific disciplines), of imperfect quality, and not necessarily directly relevant to the issue of concern (Bailar 1989).

A third set of issues concerns the measurement of exposure: what to measure, how to measure it in terms of individual exposures, the time period of concern, reconstructing exposures that were not measured at the time, adjusting for confounders and coexposures, and determining synergies among pollutants.

A fourth set of issues concerns assessing the sources of uncertainty in the estimates that may be derived from existing data and estimating their magnitude. These may be conveniently classified as true uncertainties (differences between the risk model that is used and the unknown, true state of nature), bias from random and nonrandom errors that are likely to affect exposure estimates (the differences between the data used in the model and the true data), and unmodeled variation among individuals in their sensitivity to diesel exhaust.

These 4 sets of issues commonly arise in risk assessment, leading to a rich mix of complexity and uncertainty, the characterization of which requires substantial multidisciplinary expertise. Uncertainty amplifies controversy, and careful attention is needed to all details of risk assessment to ensure that methods and results are transparent. This transparency is especially important in the case of evaluating risks posed from exposure to diesel exhaust, a widespread environmental exposure with multiple interested stakeholders.

## FOUR-ELEMENT PARADIGM

QRA for carcinogens is generally described as a 4-element paradigm, following on a National Research Council report on risk assessment; these 4 elements are hazard identification, dose-response assessment (including sensitive subpopulations), exposure assessment, and risk characterization (National Research Council 1983). The Diesel Epidemiology Working Group has adopted the National Research Council paradigm as the best way to assemble what is known, identify areas of uncertainty and other topics for further study, and estimate the size and nature of the risk to humans as well as the uncertainty in that estimate. Each of the 4 assessment steps has associated limitations and uncertainties, as discussed below.

### Hazard Identification

Epidemiologic and toxicologic studies have clearly implicated diesel exhaust as a hazard (HEI 1995; Mauderly 2000); however, diesel exhaust is a complex mixture of many chemical species, each having its own potential toxicities. Moreover, the characteristics of diesel exhaust have changed and will continue to change over time as a result of changes in engine technology, emission-control technology, and fuel composition. Although diesel exhaust has been characterized as a probable or likely human carcinogen, it will be necessary to continue evaluation of emissions for changes in toxicity over time. Without better understanding of the toxicity of the components of diesel exhaust and their interactions with other ambient pollutants, we are forced to treat diesel exhaust as though it were a single pollutant that is stable over time, while it is actually a complex mixture of PM (itself a complex mixture) and gaseous compounds that changes in the ambient environment (Kalish et al 1991).

### Exposure Assessment

The difficulties in identifying the components of diesel exhaust that cause adverse health effects lead to serious uncertainties in estimating the exposures important to the public. For the purpose of risk assessment, present knowledge and technologies point us toward measurement of a set of surrogate pollutants, rather than direct measurements of diesel exhaust or particles, for use as a "signature" of the presence of diesel exhaust. For example, general indicators of vehicle emissions such CO, nitrogen dioxide ($NO_2$), or elemental carbon (EC) might be measured and estimates made of the contribution of diesel sources to their concentrations. On this assumption, the population exposures and disease risks from diesel sources could be evaluated under different risk management strategies. Taking this a step further, one might assume that a specified change in the set of measured pollutants that make up the signature for diesel exhaust will be matched by a proportional change in diesel exhaust, and hence in health risk.

When feasible, the validity of using ambient pollutant concentrations (eg, from area monitors) to estimate exposure should be assessed by comparison with more targeted data (eg, from personal monitors). Investigators should strive to describe the actual distribution of exposures, not

only the mean (or mean and variance) of exposure values. One might expect such distributions to be highly skewed, so that a small proportion of the target population may receive very high exposures. The few highest exposures may account for most or all of the adverse outcomes of exposure. Such patterns are of concern to urban communities that may be at the high end of exposure distributions.

Most published epidemiologic studies on the health risks of diesel exhaust have examined occupationally exposed populations, using limited actual measurements or groups of individuals exposed to different conditions, to estimate the exposure levels and identify exposed as compared with nonexposed groups. These results are often extrapolated to the general population exposed to lower concentrations in the ambient air. However, methods of estimating the public's exposure to diesel exhaust are subject to substantial limitations. It is common and necessary to circumvent these limitations by making certain assumptions about the relationship between the measurements in hand and actual exposures: for example, that all persons in a specified city or segment of a city are exposed to the levels of pollutants measured by a central monitor; or that data from a personal monitor carried by a single person for a few days are representative of what the general population is actually breathing over an extended time period. These assumptions may have limited supporting data, and there may be little opportunity to carry out validation studies. Nevertheless, such assumptions may be accurate enough to be useful for estimating the risk to public health, assessing the consequences of risk management strategies, and tracking improvements in public health. Although estimating the general population's exposure to diesel exhaust is difficult and perfect validity is not attainable, the critical question is whether the estimates are accurate enough for the purpose of QRA. Questions about necessary degrees of accuracy and precision in critical inputs need to be addressed at the time a new research study, or a new risk assessment strategy, is being designed.

**Dose-Response Assessment**

Understanding the relationship between exposure and health outcomes requires an assessment of the relationship between the level of exposure and the rate of occurrence of the biological response of interest. These responses can be quantal (that is, either present or absent), such as cancer or loss of a fetus, or they may occur on a graded, even continuous scale, such as degree of lung damage.

For cancer, while the risk for each person is, in an ultimate sense, either 0 or 1, we cannot ordinarily determine risk with that precision. Rather, we express our knowledge as a probability that cancer will occur, on a scale from 0 (no risk) to 1 (certainty of cancer). Such estimates for a particular individual are mathematically equivalent to estimates of the expected number of cancers in a defined comparable population. Then, risks can be expressed conveniently as a graph (of exposure vs risk) or as a mathematical function that describes the probability of the outcome in relation to exposure (or dose). These probabilities commonly vary within a population, with some subpopulations at higher risks than others, perhaps because of genetic factors, higher exposures, or exposure to other, synergistic agents.

As an example of heightened susceptibility, smokers are substantially more susceptible than nonsmokers to the lung cancer risks associated with exposure to asbestos or radiation. Such potentiating factors may operate for diesel exposures as well, although none have been identified. Smokers, who already have a high risk of developing lung cancer, might have greater susceptibility to additional diesel-induced cancer than nonsmokers. Attention may be focused on investigating susceptibility of special groups whom society seeks to protect: children, fetuses (who are susceptible through developmental effects), and persons who are old or already ill with other serious conditions (Bailar and Louis 1988; American Thoracic Society 2000).

An understanding of susceptibility is critical in risk assessment to ensure that risks are managed appropriately for all segments of the population. In the presence of heterogeneity of susceptibility in a population, an overall risk characterization may obscure substantial differences among the risks to different members of the population. Finding previously unknown modifiers of risk may change risk assessments and their implications for risk management. Although we anticipate that the rapid advances in molecular and cellular biology will provide genetic markers for susceptibility, these advances have not reached the point of identifying persons who are unusually susceptible to or resistant to the effects of diesel exhaust.

The dose-response relationship is customarily expressed as a mathematical function or as a graph (that expresses the underlying mathematical function). The shape of this function has critical implications for the findings of a risk assessment. The generally assumed functions show 0 risk at 0 dose, high risk at very high doses, and a monotonically increasing curve between those extremes. There may be a lack of monotonicity if a different biological mechanism begins to operate at higher doses; for example, in exposure of breast cells to radiation, low doses are carcinogenic, but higher doses reduce carcinogenic responses by sterilizing or killing the cells.

Describing dose-response relationships is often challenging, as data may be available for only a few points

along the dose-response curve. These few data points may be subject to substantial bias or random error, and in epidemiologic studies they summarize effects over broad ranges of exposure rather than at fixed points. Even though exposure can be tightly controlled in the laboratory, animal studies may be subject to considerable degrees of randomness because of their small samples. There is no consensus about the general shape of dose-response curves for many exposures and biological outcomes. For numerous toxicity endpoints, and particularly for cancer, many experts believe that the actual risk often lies on or below an exponential curve (or straight line) fitted to a set of observed data points, while it is clear that some exposures may carry substantially higher risks (Bailar et al 1988).

A further complication is the time interval of exposure that is biologically relevant. For effects that are expressed in a short time period, there may be little difficulty in looking at the whole period of concern. For long-delayed effects, however, and for those that may have a considerable latency period before they become detectable, the matter is more complex. For example, assessing the lifetime probability of developing some specific form of cancer in an exposed group is complicated by the fact that human cancers have a long latency period. Often at least 10 years pass before even the first exposure-related cancers begin to appear, while the excess risk then extends over several decades. Other questions include whether we should adjust for expected deaths from other causes of death ("intercurrent mortality"), or estimate the cumulative risk on the assumption that the exposed persons would otherwise live beyond the whole period of follow-up. And, should we focus on the effects on total mortality, or on mortality from a specific cause of death that has been linked to the exposure?

Most cancer risk assessments assume that the dose-response relationship is linear. If the true shape is nonlinear but a linear relationship is assumed, there may be serious errors in estimating both total population risk and risks in susceptible populations. This problem is of special concern when the dose-response relationship is sublinear, perhaps with a threshold, so that the average person is at little or no risk. The problem is also of concern when the dose-response relation is supralinear, so that persons with low exposure are at greater risk than has been estimated. Having the full distribution of exposures can permit reasoned judgments about (1) whether some persons are exposed at levels far higher than others, and (2) whether individual susceptibility is or is not likely to be critical in assessing individual risk.

### Risk Characterization

Risk characterization provides a summary to be used by risk managers. A risk characterization should address the population's burden of disease that is attributable to the exposure of concern and the extent to which the risk varies across the population with differences in level of exposure and susceptibility. Although risk assessors and risk managers often focus on some single, central estimate of risk, a proper risk characterization also must describe the sources of uncertainty, listing all the assumptions, often numerous, that have been used in estimating the risk and the potential magnitudes of their consequences.

For diesel emissions, key sources of uncertainty are evident. We can estimate population exposure only by using exposure models or surrogate pollutants. With present data, we are unable to adequately explore the heterogeneity of exposures owing to the poor definition of exposure. The shape of the dose-response relationship is also uncertain, particularly at the levels at which most population exposure occurs. Typically, a linear dose-response relationship is assumed, but neither epidemiologic data nor toxicologic data provide strong evidence regarding whether this choice is appropriate. Improved understanding of mechanisms of action, including the uptake, pharmacodynamics, pharmacokinetics, and generation or degradation of toxic metabolites, as well as cellular effects, may lead to more biologically based risk models and greater certainty in estimates, but even when the data are available, their use is limited by other uncertainties (particularly with regard to exposures).

## DIESEL TECHNOLOGY AND EMISSIONS

### USE OF DIESEL ENGINES

Diesel engines have widespread use in transportation and industry because of operating characteristics that are favorable in comparison with gasoline engines. Diesels are more efficient than gasoline engines of comparable power because they operate at higher compression ratios and their efficiency is not severely compromised by operating under light-load conditions. When used in motor vehicles, diesel engines have a 20% advantage on an energy basis and a 30% advantage on a fuel volume (per-gallon) basis—the greater advantage on a volume basis reflects the 10% greater energy content per gallon of diesel fuel (Waters 1992). In some light-load applications, such as taxicabs, the fuel economy advantage is even greater. In Europe, a diesel passenger car with a fuel

economy of more than 80 miles per gallon (3 L per 100 km) is commercially available.

In the United States, diesels are the engine of choice for heavy-duty locomotives, trucks, construction and farm equipment, and marine vessels. In the largest heavy-duty trucks (class 8, 18-wheelers), the transition to diesel engines occurred between about 1950 and 1980, and they are now used nearly exclusively. The transition of locomotives from coal to diesel engines occurred between 1950 and 1960. In these applications the higher costs of diesel engines are justified by higher fuel efficiency and a resulting lower overall operating expense in comparison with engines of other types.

The use of diesel engines in medium- and light-duty trucks is increasing. The increased use in both commercial and personal light-duty trucks has been driven by manufacturers' difficulties in meeting the corporate average fuel economy standards with gasoline engines as the size and power of these vehicles increases. Light-duty trucks in the United States include trucks, vans, and sport utility vehicles. Use of diesel engines in passenger cars is limited in the United States but widespread in Europe, where diesel cars constitute more than 50% of new passenger car sales in some countries. The popularity of diesel vehicles in Europe is driven by higher motor fuel costs and, in some countries, by differential taxation policies. The European auto industry has pledged to further reduce $CO_2$, largely through increased use of diesel fuel.

## DIESEL ENGINE CHARACTERISTICS AND TYPES

In a diesel engine, air is compressed and fuel is injected near or before the point where the piston reaches maximum compression (ie, top dead center). The compression of the air increases its temperature to a level at which the fuel ignites (compression ignition). The amount of air in the cylinder exceeds that needed to completely burn the fuel; hence, these engines are in the category of "lean burn," in contrast to most modern gasoline engines that operate with fuel-air mixtures that are nearly stoichiometric (ie, the number of molecules of fuel matches the number of molecules of oxygen needed for complete combustion).

Several types of diesel engines are manufactured. Light-duty diesels historically have used indirect injection, in which fuel is injected into a small prechamber, ignited, and then mixed and burned with air in the main cylinder. A new generation of light-duty engines, introduced because of efficiency advantages, have direct injection, in which fuel is injected directly into the main cylinder. Light-duty engines typically have high compression ratios, on the order of 18:1 or higher, for better starting. Heavy-duty engines are generally direct-injected and have somewhat lower compression ratios, about 16:1, for optimum efficiency. Both 4-stroke engines (power produced every 2 revolutions) and 2-stroke engines (power produced every revolution) are used in heavy-duty applications. Two-stroke engines have higher emissions but deliver greater power (for the same size engines) than 4-stroke engines. Two-stroke engines are disappearing from heavy-duty road applications because of their poorer fuel economy and higher emissions but continue to dominate locomotive and marine applications. Most heavy-duty engines, and some light-duty engines, are turbocharged to increase power and efficiency.

A new type of diesel engine, the homogeneous charge compression ignition engine, is under intense development, and early versions are already in use. In these engines the fuel and air are premixed, still with an excess of air, before compression ignition. They offer the possibility of greatly reduced emissions of particles and $NO_x$ without loss of efficiency. Control of ignition timing and operation in the homogeneous mode over the entire range of engine speed and load remains a problem. These engines may have the additional advantage of being able to work with a broader range of fuels than conventional diesels.

## DIESEL FUEL

Refining of diesel fuel is less complex and requires less energy than refining of gasoline. Refineries in the United States produce more gasoline than diesel fuel, while in most other countries of the world the production profile is reversed. One key property of diesel fuel is the ease with which it ignites, quantified by the cetane number, with higher numbers indicating greater ignitability. For gasoline, quality is quantified by the octane number, with higher values indicating less susceptibility to knocking, a form of compression ignition. Hence good-quality diesel engine fuels are poor-quality gasoline engine fuels, and vice versa. Other properties that affect engine performance and emissions include aromatic hydrocarbon content (high levels increase particulate emissions), sulfur levels (high levels increase particulate emissions and interfere with some emission controls), and distillation characteristics (heavier fuels have more high molecular weight components and are more difficult to burn cleanly). Large, slower speed engines, as used in locomotive and marine applications, are more tolerant of poor-quality fuels.

Current commercial fuels include, in order of increasing density and viscosity, reformulated diesel, diesel #1, diesel #2, diesel #4, and bunker (Goodger 1975; US Environmental Protection Agency [EPA] 2000). These fuels can differ in cetane number, aromatic content, sulfur level, levels of contaminants (eg, metals), and viscosity.

Differences among fuels, historically and in different applications, may limit the applicability of exposures and risks determined in one setting (place and time) to those in different settings.

Future diesel fuels will contain much lower levels of sulfur and may also include a broad variety of types: water emulsions, in which water is suspended in the fuel to improve combustion and reduce emissions; biodiesel, derived from plant oils such as rapeseed oil; oxygenates, either as additives to petroleum-derived diesel fuels, such as methyl *tert*-butyl ether, or as alternative diesel fuels, such as dimethyl ether; and gas-derived fuels or blending components, such as Fischer-Tropsch diesel fuel, which can be synthesized from natural gas and used as a diesel fuel or blended with petroleum-based diesel fuel. These fuels may be introduced for emission advantages or as alternatives to petroleum-derived diesel fuel. Some Fischer-Tropsch diesel fuel is now used as a blending agent, and the California Air Resources Board has approved use of water emulsion fuel as a strategy to reduce emissions of $NO_x$ and particles. Some natural gas is being used in heavy-duty engines, primarily in bus and other transit fleets. These engines generally are spark-ignited, rather than compression-ignited, and replace engines in an application that is traditionally filled by diesels.

## CHARACTERISTICS OF DIESEL EMISSIONS

Uncontrolled diesel engines emit high concentrations of particles, $NO_x$, and aldehydes and low concentrations of CO and hydrocarbons. Particle concentrations are high because of fuel-rich high-temperature combustion; as fuel and air are not premixed, a locally fuel-rich region always surrounds the fuel injection jet. The high temperature, which occurs near locally stoichiometric conditions, favors nitric oxide (NO) formation. Because of the high compression ratio, peak temperatures are higher than in gasoline engines, and NO formation is somewhat greater. The $NO_x$ in diesel exhaust contain a higher fraction of $NO_2$ than exhaust from gasoline engines because the excess air intake of diesel engines allows greater conversion of NO to $NO_2$. Aldehydes are formed as fuel that is hot but unburned mixes with the excess, cooler, surrounding air. Emissions of CO and hydrocarbons are low compared with those from stoichiometric gasoline engines because the excess air in diesel engines favors more complete oxidation of the fuel.

Diesel emissions contain hundreds of different organic compounds. Those found in the gas phase under atmospheric conditions are termed volatile organic compounds (VOCs), and those found in either the gas or condensed phases are termed semivolatile organic compounds (SVOCs). Many of the SVOCs are primarily gaseous in the hot exhaust but condense on the PM as the exhaust dilutes and cools.

There is a tendency to focus on PM in diesel exhaust because it is often a visible component, but diesel exhaust includes much more than the visible PM. The nonparticulate fraction of diesel exhaust may also have important health effects, especially the SVOCs that can condense onto the diesel PM. In particular, gaseous diesel exhaust contains many compounds that are considered toxic or carcinogenic, including aldehydes, aromatics (eg, benzene), 1,3-butadiene, and PAHs and nitro-PAHs. Diesel particles also contain PAHs, nitro-PAHs, and other forms of organic carbon (OC), as well as EC, sulfate, and metals.

### Particle-Phase Emissions

Considerations of the risks of diesel exhaust have often emphasized the particle-phase component, which has carcinogenic constituents and penetrates into the lung. Diesel particles are complex, covering a range of sizes and morphologies, and having myriad chemical components that vary with engine characteristics, operations, and fuels. Because of this complexity, the physical and chemical characteristics of diesel particles have not been fully elucidated and only a relatively small fraction of the mass of organic emissions has been identified by chemical species.

The size distribution of exhaust particles is relevant to potential health risks, as size is one of the key determinants of penetration into the lung and the site of deposition in the lung. The size distribution of diesel particles differs from engine to engine, among fuels, and with operating conditions in the same engine, such as engine load and exhaust dilution. Like most combustion-generated aerosols, diesel particles are relatively fine; the typical particle has a small aerodynamic diameter, less than about 1 µm (Sidebar 1). The size distribution shows a significant fraction of the total mass at about 0.3 µm in the accumulation mode, accounting for about 80% to 95% of the mass. The mass in the accumulation mode leads to the visible exhaust since particles of this size are efficient at scattering and absorbing light. Morphologically, particles of this size are nonspherical, consisting of much smaller particles that have agglomerated. They may contain condensed material such as SVOCs and sulfuric acid (Figure 1).

Some studies suggest that there can also be a large number of nanoparticles (particle diameter less than 0.05 µm or 50 nm) in the nuclei mode, though the nanoparticles still account for only a small fraction of the total mass. However, there is a general concern among health scientists that ultrafine particles (diameters of less than 0.1 µm or 100 nm), which largely overlap with nanoparticles, may

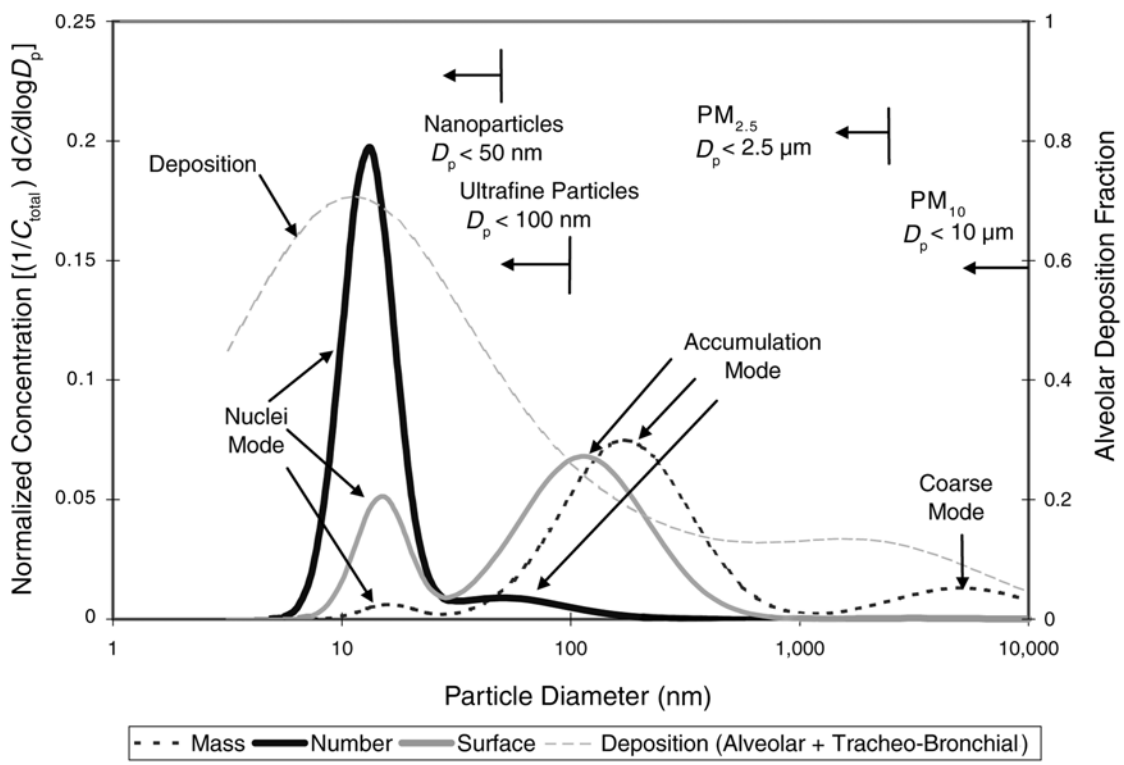
## SIDEBAR 1. EXAMPLE SIZE DISTRIBUTION AND ALVEOLAR DEPOSITION FRACTION FOR DIESEL PARTICULATE MATTER

The diameter is often used as a measure of particle size. The most common size descriptor is the aerodynamic diameter, which depends on the particle density and is defined as the diameter of a spherical particle (assumed to have a density of 1 g/m$^3$) with the same settling velocity as the particle being measured.

Particles tend to fall into 3 size classifications referred to as modes. The Figure illustrates typical mass-weighted, number-weighted, and surface-weighted size distributions for diesel particles. It also illustrates the alveolar deposition fraction for particles of different diameters. Whereas most of the mass is due to the larger particles in the accumulation mode, most of the number is due to particles in the nuclei mode. The size distribution varies depending upon engine design, fuel characteristics, and operating conditions. There is some suggestion that as the mass of emissions goes down, a greater fraction may be emitted in the ultrafine mode.

**Aerosol Particle Sizes (all in aerodynamic diameter)**

| | |
|---|---|
| $PM_{10}$ | 10 µm or smaller |
| $PM_{2.5}$ | 2.5 µm or smaller |
| Ultrafine particles | 0.1 µm (100 nm) or smaller |
| Nanoparticles | 0.05 µm (50 nm) or smaller |
| Coarse mode | 1.5 µm and larger |
| Accumulation mode | 0.05 µm (50 nm)–1 µm |
| Nuclei mode | 0.005 µm (5nm)–0.050 µm (50 nm) |




**Sidebar 1 Figure. Normalized mass-weighted and number-weighted particle-size distribution curve from typical diesel exhaust.** *C* denotes the number or mass concentration as indicated; *d* denotes differential; and $D_p$ is the mean particle diameter. (Adapted from Kittelson et al, this volume.)

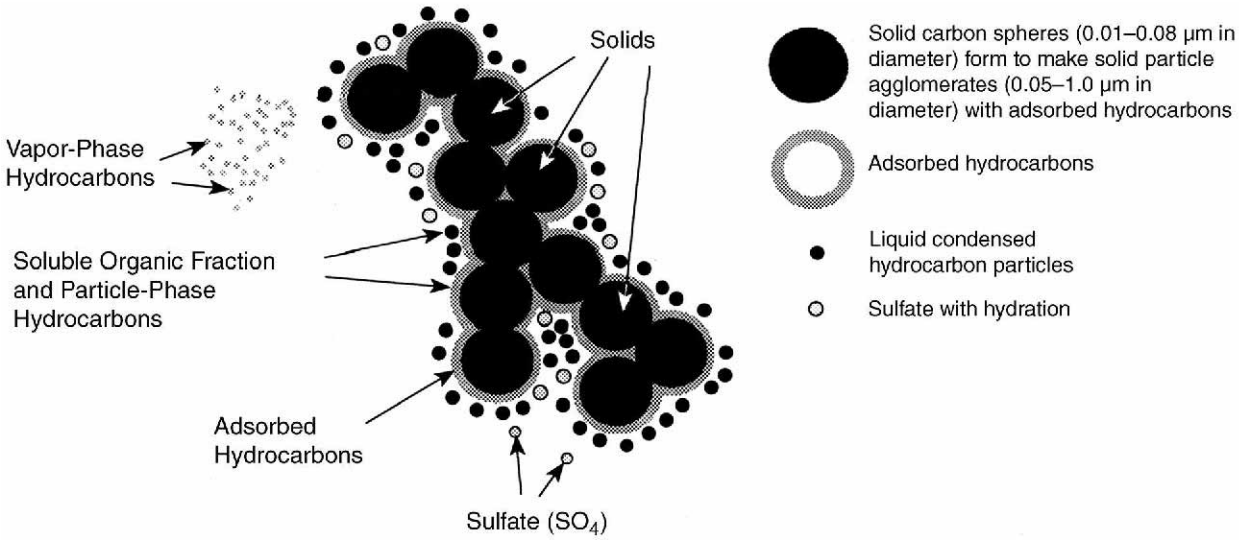



**Figure 1. Schematic representation of diesel exhaust particles and vapor-phase compounds.** (Adapted with permission from SAE Paper 940233, © 1994, Society of Automotive Engineers Inc.)

carry greater risk per unit weight than particles of other sizes. The formation of the accumulation mode diesel particles as aggregates of very small carbonaceous particles and ash gives these particles a high surface area; consequently, they are very efficient at adsorbing SVOCs, PAHs and PAH derivatives (nitro-PAHs and oxidized PAHs), and inorganic compounds (including sulfate).

Because of concern about possible health risks of exposure to very small particles, a number of studies have focused on ultrafine diesel PM. One of the HEI feasibility studies, Gertler and colleagues (2002), studied particles in a freeway tunnel and found an increase in 0.02-µm particles with an increasing proportion of diesel vehicles. This positive association suggests that under real-life conditions diesel exhaust contains ultrafine particles or provides a substrate for their formation. This finding contrasts with some laboratory measurements. For example, Kleeman and associates (2000), using a different sampling strategy—a dilution tunnel—did not find a peak in this size range. This finding suggests that when source sampling is done under conditions of clean air in the laboratory, surfaces for condensation are not available.

Ultrafine particle formation is expected to be sensitive to emission control technologies, engine design, and fuel changes. Kittelson and colleagues (this volume) found that using an oxidizing catalyst to remove organic compounds reduced ultrafine particle formation. Some studies also have suggested that even though the total mass of particle emissions has been reduced with newer diesel engines, they may emit a larger number of ultrafine particles than older engines (Bagley et al 1996; Baumgard and Johnson 1996; Kreso et al 1998; Abdul-Khalek et al 1999). The rise in ultrafine particles may reflect a decrease in accumulation mode particles, thereby reducing the surface area for condensation of SVOCs and sulfuric acid, and a consequent tendency to increased nucleation.

Fuel composition also affects ultrafine particle formation. Some investigators have concluded that decreasing sulfur content in fuel will decrease the nucleation and condensation of fine particle sulfate (Opris et al 1993; Baumgard and Johnson 1996; Abdul-Khalek et al 1999). Other factors, such as fuel quality, additives, and injection pressure, also affect the formation of ultrafine diesel particles.

Chemically, diesel particles are composed primarily of EC, OC, and sulfate, although their composition is highly variable. For example, the EC component can range from about 30% to 90% of the mass. In general, about 30% of the mass is composed of unburned oil and fuel and partially combusted or pyrolysis products, with the remainder as condensed inorganic oxides (eg, sulfate), water, and ash, which can contain trace metals (Figure 2).

The range of component organic compounds in diesel particles is large, reflecting the complex composition of diesel fuel and oil, the formation of other organic compounds during combustion, and the condensation of SVOCs onto the particles. The chemical species present in the organic fraction of diesel particles have been characterized by extracting the particles with organic solvents and then analyzing the species in the extract. This component

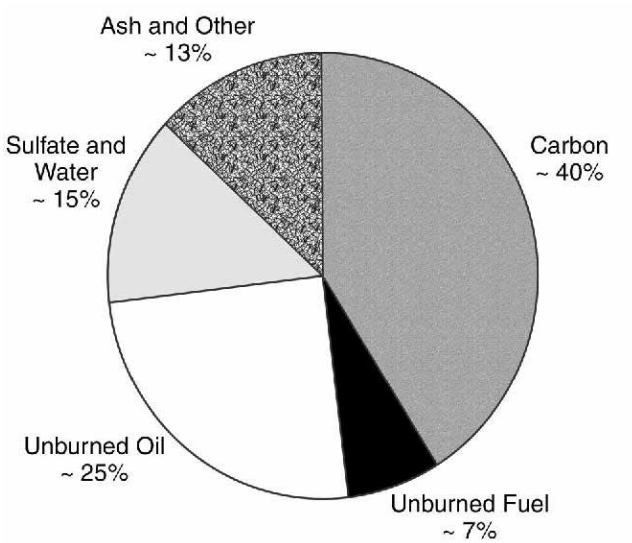



**Figure 2. Chemical composition of emissions from a heavy-duty diesel engine.** Dominant components are carbon and unburned oil and fuel. The composition varies significantly with engine design, operation, and fuel. (Adapted from Kittelson 1998.)

of the particles, referred to as the soluble organic fraction (SOF), is largely unburned fuel and oil (Kittelson 1998), with a small fraction of combustion products. Some PAHs are included in this fraction although most of the diesel PAHs are in the gas phase.

Schuetzle (1983) used different solutes to split the SOF of particulate matter into 3 fractions: nonpolar, moderately polar, and polar. Most of the mass (57%) was found in the nonpolar fraction, being primarily (90%) large aliphatic hydrocarbons ($C_{14}$ and above). The next largest fraction was polar (32%), containing organic (alkanoic) acids and PAHs and their derivatives. The remaining mass (11%) was found in the moderately polar fraction, in which species were primarily oxygenated and nitrated PAHs. Rogge and colleagues (1993) also used organic fractionation, coupled with gas chromatography–mass spectrometry analysis, to investigate diesel particle composition. About 90% of the mass was not identified specifically, but was primarily large hydrocarbons. Much of the resolved mass was large alkanes and alkanoic acids. The PAHs and their derivatives accounted for about 7% of the identified mass.

Zielinska and coworkers (this volume) found similar results in a mine where diesel engines were in use. The engines used in mining operations are generally not the same as those in highway vehicles, and the operating mode distribution is also different; however, the results from the mine were similar to those from other environments. Zielinska and colleagues provide extensive data on speciation of both the particle-phase and gas-phase organic composition, as well as on the size distribution of mass and of OC and EC. Elemental carbon was, on average, about 40% of the total carbon, lower than is typical for on-road diesel vehicles. Much of the carbon was found in the 0.148-µm size fraction and below. Sulfate mass varied widely, from 1% to 24% of the total mass, and was found largely in the form of smaller particles (approximately 50 nm). From this, it appears that particles less than 50 nm in aerodynamic diameter (nanoparticles) are rich in sulfate and, in at least some cases, carbon.

The presence of PAHs and their derivatives in diesel emissions is of particular concern because some are suspected to be carcinogenic. Both PAH and PAH derivatives come largely from the diesel fuel. Owing to their low combustion rate, a large portion of the PAHs in fuel remains uncombusted or partially oxidized. For example, Zielinska and colleagues (this volume) identified individual PAHs and PAH derivatives in their study of underground mines. Whether emissions of PAHs have decreased over time is uncertain. While Zielinska did find a reduction in the amount (mass) of PAHs from the 1980s, the trend was scattered and moderate. The reduction may have been due, in part, to a decrease in the PAH content of fuel after 1993. Reducing the aromatic content of diesel fuel, which is currently capped at 35%, reduces emissions of PAHs, particularly those with lower molecular weights. Thus, PAH emissions should continue to drop in the future as total particle emissions decrease. The amount of SOF emissions (ie, on a per-engine basis) appears to be declining with time, both absolutely and as a fraction of total diesel particles (Dietzmann et al 1981; Warner-Selph and Dietzmann 1984; Rogge et al 1993; Graboski et al 1998).

Sulfate comprises from 1% to 10% of diesel particle mass, depending on fuel composition. Sulfur in the fuel oxidizes to form mostly sulfur dioxide ($SO_2$) with small amounts of sulfur trioxide ($SO_3$). The $SO_3$ picks up water to form sulfuric acid, which has a very low vapor pressure, and condenses either as nuclei mode particles or onto the preexisting PM. As noted above, a large fraction of the ultrafine particles may be sulfate, formed when there is insufficient aerosol surface area to adsorb the rapidly condensing sulfuric acid. The sulfur fraction of diesel particles decreases as the amount of sulfur in the fuel declines. The $SO_2$ can transform into secondary sulfate particles, adding to the ambient PM burden from diesel emissions.

The metals present in diesel particles come from trace amounts in the fuel and oil (eg, as additives). After combustion, they are exhausted as ash. Studies have found varying levels of metals, from less than 1% to 5%, with higher levels coming from engines with higher emissions of lubricating oil (Kittelson 1998). Some of the metals

found in diesel particles are iron, silicon, chromium, mercury, and barium (Kittelson 1998).

### Gas-Phase Emissions

A substantial proportion of the mass of diesel exhaust is not in the particle phase but in the gas phase. The high air-to-fuel ratio in diesel engine combustion leads to relatively low, though not negligible, emissions of CO and VOCs, low levels of gaseous SVOCs and $SO_2$, and high levels of $NO_x$. It is difficult, at present, to reduce the downstream levels of $NO_x$, as in gasoline-fueled cars, owing to the excess oxygen in the exhaust. Diesel trucks and other diesel engine applications are becoming proportionally greater contributors to the $NO_x$ emissions in the United States, as $NO_x$ emissions from gasoline-fueled vehicles and electric utility sources decline.

Organics in diesel exhaust include both lighter VOCs and heavier SVOCs in addition to the organic fraction of the diesel PM discussed above. Diesels emit large amounts of gas-phase aldehydes; these are largely formaldehyde but include acetaldehyde and higher molecular weight aldehydes. Only a small fraction of the aldehydes condense because they are generally smaller molecules with higher vapor pressures. Lighter PAHs are also found in the gas phase and are dominant, by mass, over the condensed-phase PAHs (Zielinska et al, this volume). Naphthalenes dominate the gas-phase PAH emissions.

Chlorinated dioxins and furans, which are considered air toxics and possibly carcinogenic, may also be emitted as a result of incomplete combustion. The chlorine component can come from additives in the fuel or oil, or from road salts. In recent tunnel measurements, Gertler and colleagues (1998, 2002) reported an emission rate of 172 pg/km of toxicity equivalency quotient (TEQ). (A TEQ is referenced to the equivalent toxicity of 2,3,7,8-tetrachlorodibenzo-*p*-dioxin [TCDD]). The measurements of Gertler and colleagues for TCDD fall between measurements of others, which range from about 2 pg TEQ/km (Hagenmaier et al 1990) to 5100 pg TEQ/km (Oehme et al 1991) for a light-duty diesel vehicle. These compounds are found primarily in the gas phase at higher exhaust temperatures, with a fraction condensing as the exhaust cools.

## REGULATION AND CONTROL OF DIESEL EMISSIONS

In the past, diesel engine emissions have been less regulated than gasoline engine emissions, but on-road diesel engines now face severe emission reduction requirements (Table 3). Emissions from diesel engines in off-road applications have been regulated only recently, and much less stringently than those used in on-road applications (Table 4).

**Table 3.** United States On-Road Heavy-Duty Diesel Emission Standards

| Year | $NO_x$ (g/bhp-hr) Trucks and Buses | Particulate Matter (g/bhp-hr) Trucks | Particulate Matter (g/bhp-hr) Buses |
|---|---|---|---|
| 1985 | 10.7 | No standard | No standard |
| 1988 | 10.7 | 0.6 | 0.6 |
| 1990 | 6.0 | 0.6 | 0.6 |
| 1991 | 5.0 | 0.25 | 0.25 |
| 1993 | 5.0 | 0.25 | 0.1 |
| 1994 | 5.0 | 0.1 | 0.07 |
| 1996 | 5.0 | 0.1 | 0.05 (0.07 in use) |
| 1998 | 4.0 | 0.1 | 0.05 (0.07 in use) |
| 2004 | 2.4[a] | 0.1 | 0.05 |
| 2007–2010[b] | 0.2 | 0.01 | 0.01 |

[a] Hydrocarbons and $NO_x$.

[b] Proposed by EPA (Federal Register 2001b), to be phased in. Includes 15 ppm sulfur fuel. Nonmethane hydrocarbon standard = 0.014 g/bhp-hr.

**Table 4.** United States Off-Road Diesel Emission Standards

| Year | Source | $NO_x$ (g/bhp-hr) | Particulate Matter (g/bhp-hr) |
|---|---|---|---|
| 2002–2004 | Locomotives, line haul | 7.4 | 0.45 |
| 2005 | Locomotives, line haul | 5.5 | 0.20 |
| 2002–2004 | Locomotives, switch | 11.0 | 0.34 |
| 2005 | Locomotives, switch | 8.1 | 0.24 |
| 2004–2007 | Marine, < 5 L/cylinder | 7.2 to 7.5[a] | 0.20 to 0.40 |
| 2007 | Marine, > 5 L/cylinder | 7.8 to 11.0[a] | 0.50 |
| 1996 | Nonroad, 175 to 750 hp | 6.9 | 0.40 |
| 1997–1998 | Nonroad, 50 to 175 hp | 6.9 | — |
| 2000 | Nonroad, > 750 hp | 6.9 | 0.40 |
| 2005–2008[b] | Nonroad, < 100 hp | 3.5 to 5.6[a] | 0.30 to 0.60 |
| 2005–2008[b] | Nonroad, > 100 hp | 4.8 to 4.9[a] | 0.15 |

[a] Hydrocarbons and $NO_x$.

[b] Proposed by EPA (Federal Register 2001a).

Currently, most diesel engines are able to meet emission standards through engine modifications such as high-pressure injection, computer-controlled injector timing, and, in some cases, recirculation of exhaust gas. Meeting the stringent 2004 Tier 2 light-duty standards (set by the EPA) that apply to both gasoline and diesel engines and the 2007 heavy-duty standards will require exhaust after-treatment. Technologies under development include catalytic oxidation, continuously regenerated traps, actively regenerated traps, $NO_x$ storage catalysts, selective catalytic reduction with urea, individual electronically controlled valve actuation, and catalytic fuel additives. Using these controls, under certain operating conditions it has been possible to reduce both particles and $NO_x$ by more than 90%. The most effective control methods for particle mass probably also effectively reduce particle number (the nanoparticles), aldehydes, and hydrocarbons (Manufacturers of Emission Controls Association 1999).

Despite the more stringent regulations for $NO_x$, Yanowitz and associates (2000) found relatively little decrease in $NO_x$ emissions from in-use heavy-duty diesels (in grams per mile) between 1975 and 1995. In contrast, engine certification data showed a substantial decrease in particles and $NO_x$ (Figure 3). An engine operating strategy that optimizes in-use fuel economy is believed responsible. Over the same period, however, Yanowitz and colleagues found significant decreases in PM in both diesel engine certification data and measurements of on-road emissions. Tunnel studies have also found decreases in diesel PM emissions (eg, Pierson and Brachaczek 1983; Gertler et al 1995a,b, 2002; Kirchstetter et al 1999). Chassis dynamometer and tunnel measurements supply strong evidence that particulate emissions from the in-use on-road diesel truck fleet have decreased substantially in the past 25 years (Figure 4). This drop is consistent with the increasingly stringent emission standards.

Recent measurements of 10 heavy-duty trucks from a precontrol period (1952 to 1975), under transient operating conditions (more demanding than the steady tunnel driving), showed particle emissions of 1.0 to 5.6 g/km (Fritz et al 2001). These data suggest that heavy-duty truck emissions of particles (per mile) have decreased substantially as a result of emission regulation. This improvement in individual vehicle emissions has occurred while the number of diesel vehicle miles traveled and the amount of diesel fuel consumed have increased greatly. The tunnel data also showed that particulate emissions per mile from light-duty gasoline vehicles were roughly one tenth those from heavy-duty vehicles (Fritz et al 2001). This suggests that in urban settings where the number of miles traveled by gasoline vehicles is much greater than the number traveled by diesel vehicles, the 2 types of vehicles may be comparable contributors of particles. However, the relative contributions of particles by gasoline and diesel vehicles remain controversial, with conflicting data preventing a clear resolution.

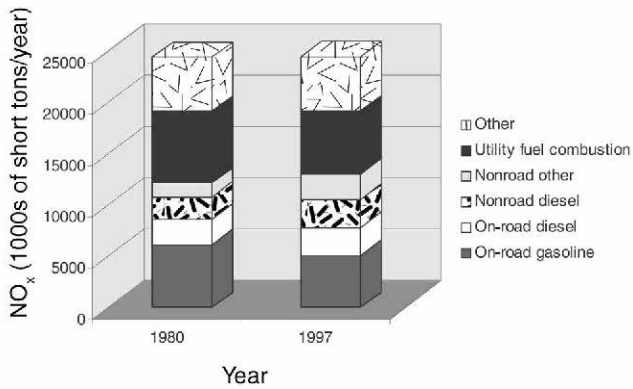



**Figure 3. Nitrogen oxide emissions in the United States, 1980 and 1997.** As shown, the total emissions remained relatively constant between 1980 and 1997, though the fraction from diesel engines (on-road and nonroad) increased. (Adapted from US EPA data [EPA 1999].)

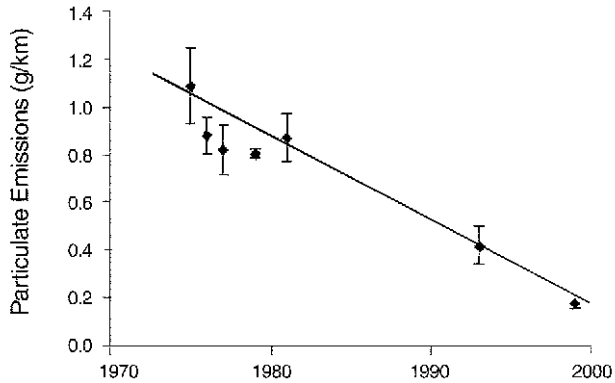



**Figure 4. Measurements of heavy-duty diesel particulate emissions at the Tuscarora Tunnel of the Pennsylvania Turnpike.** Error bars represent the uncertainty estimates in the emission rates. (Adapted with permission from Gertler et al 2001.)

Tunnel and chassis dynamometer measurements of $NO_x$ emitted by heavy-duty diesels showed little decrease following regulation, which is not consistent with emission standards (Yanowitz et al 1998; Gertler et al 2002). An engine operating strategy that optimizes in-use fuel economy is considered responsible.

Diesel particles are regulated by the mass emitted, but the size distribution of particles has garnered increased interest with the observation that engine modifications to reduce mass can increase the number of the smallest particles (nuclei mode or nanoparticles). Measurements of both particle mass and particle number are sensitive to exhaust dilution ratios, temperature, humidity, and fuel sulfur levels, leading to some controversy over correct particulate characterization. Tunnel tests, which involve real-world dilution conditions, have confirmed the existence of large numbers of nuclei-mode particles in the range of 8 to 20 nm (Figure 5). These particles may have existed historically and it is possible that at least some of them are not a result of new technology.

As regulations have tightened over time, diesel fleet emissions have dropped, but with a substantial lag. Diesel engines last much longer than gasoline-fueled engines and are often rebuilt, so that turnover is slow. Further, the use of diesel trucks for hauling a growing volume of goods continues to increase and may not yet be near its maximum (Figure 6). In addition, nonroad diesel emissions have increased significantly. While total emissions of diesel PM in the United States have decreased since 1990 (EPA 2000) and on-road diesel PM levels are expected to continue to decline (Figure 7), nonroad diesel PM is expected to increase.

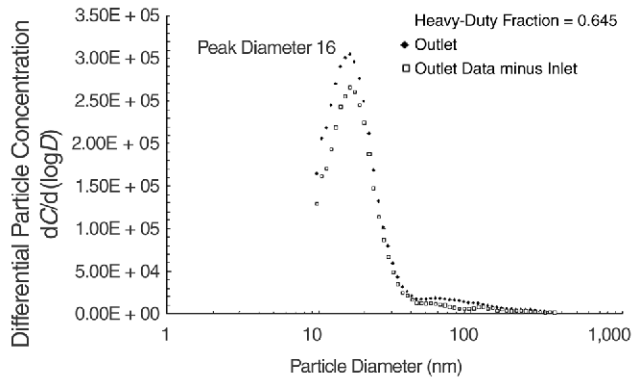



**Figure 5. Tuscarora Tunnel measurements from a scanning mobility particle sizer (5/18/1999): Outlet site data with and without subtraction of inlet data.** *C* = concentration and *D* = particle diameter. (Adapted from Gertler et al 2002.)

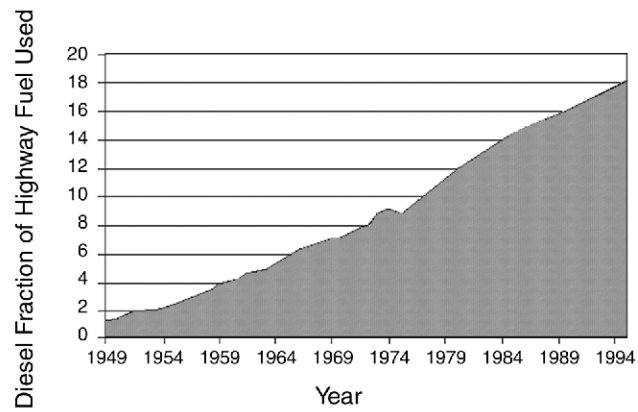



**Figure 6. Diesel fuel use as a fraction of total automotive fuel use.** (Adapted from HEI 1999a.)

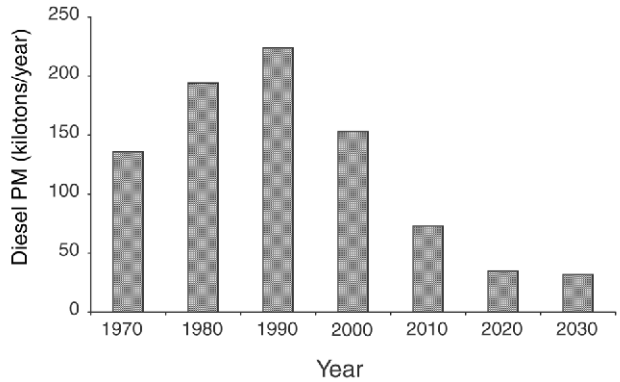



**Figure 7. On-road heavy-duty diesel PM emissions in the United States.**

## TOXICOLOGIC STUDIES OF DIESEL EXHAUST

The toxicity of diesel exhaust has been studied for decades in a variety of assay systems, ranging from in vitro studies, to chronic (2-year) inhalation studies in several animal species, to short-term exposures of human volunteers (Rudell et al 1996, 1999; Salvi et al 1999). The extensive literature has been comprehensively reviewed in several publications (Mauderly 1992, 2000; HEI 1995; Cohen and Nikula 1999; EPA 2000). We touch on selected studies relevant to the risk assessment issues that are the focus of this report. These studies include in vitro assays relevant to carcinogenesis, chronic inhalation assays, primarily for carcinogenicity, and investigations relevant to noncancer effects of diesel exhaust exposure.

### IN VITRO TESTS

In vitro tests of the toxicity of diesel exhaust have focused on its genotoxicity. Carcinogens in the gas phase of diesel exhaust include ethylene, benzene, 1,3-butadiene, acrolein, formaldehyde and other aldehydes, and a variety of PAHs. Particle-associated carcinogens include PAHs and nitro-PAHs. Extracts of diesel soot evaluated in the

*Salmonella typhimurium* (Ames) assay were mutagenic (Huisingh et al 1978; Bagley et al 1987). The most active mutagenic fractions of the extracts were the nitro-PAHs (International Agency for Research on Cancer 1989). Mutagenic activity was also observed in the mouse lymphoma mutation assay (Mitchell et al 1981) and in the embryonic cells of Syrian hamsters (Morimoto et al 1986). Diesel soot extracts also cause structural chromosomal aberrations and sister chromatid exchanges in different systems (International Agency for Research on Cancer 1989). Thus, the in vitro data indicate that diesel soot extracts contain genotoxic chemicals.

The bioavailability of these mutagenic compounds in vivo, a key determinant of their likely toxicity, has been studied with a variety of techniques. Higher amounts of DNA adducts can be found in the lungs of rats exposed to diesel exhaust (reviewed in Shirnamé-Moré 1995). Other investigators have supplemented diesel particles with radiolabeled PAH and observed the transfer of one or more compounds in vivo (Bevan and Ruggio 1991; Gerde et al 2001), or have observed the ability of microsomes to enhance transfer of PAH from the particles (Leung et al 1988). In all these cases there was evidence that only a small fraction of the PAH was bioavailable in vivo.

## CHRONIC INHALATION STUDIES

Inhalation studies in rats have indicated that it is the particle phase of diesel exhaust that causes pulmonary toxicity after chronic exposures (Heinrich et al 1986). Two-year inhalation studies with diesel exhaust emissions have been conducted in rats, mice, and Syrian hamsters (reviewed in Mauderly 2000). Although high lung burdens of diesel soot caused an inflammatory response in all 3 species (Henderson et al 1988), only rats had a dose-related increase in the incidence of lung tumors. Rats are known to develop lung tumors in response to high lung burdens of several types of poorly soluble particles of low toxicity, including titanium dioxide (Lee et al 1985) and coal dust (Martin et al 1977). The lungs of rats are prone to develop lung tumors when the lung burden of poorly soluble particles reaches a volume greater than 60 to 600 $\mu m^3$ per alveolar macrophage, as has been reported (Morrow 1988). This mechanism is not relevant to most scenarios of human exposure. Chronic inhalation of coal dust leads to lung tumors in rats, while coal miners are at risk for pneumoconiosis and chronic airflow obstruction, but not for lung cancer (Miller and Jacobsen 1985).

The induction of tumors in rat lungs following chronic inhalation of diesel exhaust could be caused by the same factors that lead to rat lung tumors following inhalation of relatively inert particles. Alternatively, the tumors could be caused by PAHs or other organic material adsorbed on the surface of the diesel soot particles. In an effort to clarify the mechanisms of tumor induction in rat lungs, rats were exposed to carbon black containing only traces of organic material at levels similar to those used in the diesel studies (Mauderly et al 1994; Nikula et al 1995; Mauderly 2000). The numbers and types of lung tumors induced by diesel soot exposure in the rats were the same as those induced by carbon-black exposure. These findings suggested that the tumors in the rats exposed to diesel exhaust resulted nonspecifically from the high lung burden of poorly soluble particles and not from specific diesel carcinogens. Thus, the animal model results may not be relevant to humans, who are exposed to low doses of diesel exhaust.

The consensus report of a workshop held by the International Life Sciences Institute (2000) concluded that organic compounds adsorbed to the particles in diesel exhaust are involved in effects in humans. The EPA, in its draft health assessment document for diesel exhaust (EPA 2000), stated that while rat lung cancer response to diesel exhaust is not suitable for dose-response extrapolation to humans, the positive lung cancer response does imply a hazard for humans.

## NONCANCER TOXICITY

Recent research indicates that airborne PM, including diesel soot, can enhance allergic responses. Studies on the ability of diesel exhaust particles to act as immunologic adjuvants have been reviewed (Casillas et al 1999; Chambellan et al 2000). In comparisons of responses to nasal instillations of allergens with or without diesel particles, responses to those with diesel particles were characterized by an increased release of histamine from activated mast cells (Diaz-Sanchez et al 2000) and an increase in IgE production (Saxon and Diaz-Sanchez 2000). Studies by Ormstad (2000) indicated that diesel particles can absorb allergens as well as act as adjuvants for responses to the allergens. Fahy and colleagues (1999) used peripheral blood monocytes from allergic patients to demonstrate that the combined exposure to diesel soot and an allergen resulted in increased release of chemokines via activation of mitogen-activated protein kinase pathways.

Steerenberg and coworkers (1999) have suggested a model for testing the adjuvant activity of particles in the Brown Norway rat. The test involves intranasal and intratracheal challenges with pollen grains with and without diesel particles. In a mouse model reported by Granum and associates (2001), polystyrene particles injected intranasally or intratracheally in combination with an allergen were found to enhance both allergen-specific and total IgE responses when compared with allergen

alone. Also in a mouse model, van Zijverden and colleagues (2000) showed that various particles, including diesel PM, carbon black, and silica particles, stimulated immune responses mediated by T helper 2 cells. All the particle types acted as adjuvants, but in different ways. The mice treated with diesel particles (by intranasal instillation) had high levels of $IgG_1$ and IgE in serum and high levels of cells that produce $IgG_1$ antibody and IgE antibody in lymph nodes, bone marrow, and spleen, suggesting that the diesel particles skewed the immune response toward the T helper 2 cell side. The other particles showed a mixed response with stimulation of both T helper 1 and T helper 2 cell responses. These studies, characterized by direct introduction of diesel PM by intranasal or intratracheal instillation, represent unusually high localized doses of diesel PM.

Overall, both human and animal data indicate that diesel particles are capable of enhancing, but not inducing, an allergic response; however, the experimental models do not reflect the usual circumstances of exposure of the general population (Gavett and Koren 2001). The potential for airborne particles, including diesel soot, to enhance allergic responses has been cited in regard to the rise of asthma and other allergic diseases. If diesel particles have a significant role in causing or exacerbating asthma and allergic diseases, the public health burden could be substantial, given the prevalence of these disorders.

Finally, developmental effects on fetuses have been reported when pregnant rats were exposed from day 7 through day 20 of pregnancy to either diluted diesel engine exhaust containing 5.6 $mg/m^3$ particles or filtered diesel exhaust (Watanabe and Kurita 2001). Both types of diesel exposure resulted in an increase in the anogenital distance and delayed and disturbed development of the testes, ovaries, and thymuses in the pups, an increase in maternal testosterone, and a decrease in maternal progesterone compared with control animals. The authors referred to the symptoms as "masculinization" of the fetus and attributed the cause to the gaseous phase of diesel exhaust. Further work is necessary to confirm and elucidate these findings.

In summary, toxicologic studies indicate that high lung burdens of diesel soot result in lung tumors in the rat, but not in other species. The combination of animal studies and human clinical studies suggests that diesel soot may act as an adjuvant to increase responses to allergens in experimental models. The implications of this for general environmental exposure conditions are being investigated.

## EPIDEMIOLOGIC STUDIES OF DIESEL EXHAUST

### BACKGROUND

As animal models are inadequate for obtaining quantitative estimates of the cancer risk posed to the general population by diesel emissions, epidemiologic data have been the starting point for developing risk models. Numerous studies of diverse design have examined the association between lung cancer risk and diesel exhaust exposure. The difficulty of accurately estimating occupational exposure across the full work experience has limited interpretation of the resulting evidence and its utility for QRA. Nonetheless, reviewing agencies have used data from several studies to derive risk models; among the principal studies considered for this purpose are the study of US railroad workers carried out by Garshick and colleagues (1987, 1988) at Harvard and the study of US teamsters in the trucking industry carried out by Steenland and others (1990, 1998) at the National Institute for Occupational Safety and Health.

The epidemiologic literature has been reviewed extensively in previous HEI reports and elsewhere, and the Working Group did not repeat these reviews (HEI 1995, 1999a; California Environmental Protection Agency 1998; EPA 2000; Mauderly 2000). Most recently, HEI's Diesel Epidemiology Expert Panel (HEI 1999a) reviewed in detail the 2 epidemiologic studies that have been the principal basis for risk assessment—the Harvard study of US railroad workers (Garshick et al 1987, 1988; Hammond et al 1988; Woskie et al 1988a,b; Crump et al 1991; California Environmental Protection Agency 1998; Crump 1999) and the study of US teamsters carried out by the National Institute for Occupational Safety and Health (Steenland et al 1990, 1992, 1998; Zaebst et al 1991). The utility of these studies for QRA was reviewed, with an overall finding that enhanced exposure and epidemiologic analyses are needed for QRA. The Expert Panel thus recommended that an additional prospective cohort study, possibly with retrospective components, be considered after (1) HEI's feasibility studies were completed and evaluated, and (2) attempts to retrofit improved exposure assessments to existing studies were evaluated with respect to the adequacy of the exposure data to support QRA (HEI 1999a).

The EPA also concluded that there is uncertainty about the exposure-response relationship in the epidemiologic data (Garshick et al 1988; Hammond et al 1988; Woskie et al 1988a,b), and therefore did not calculate a unit risk estimate. However, it did provide a possible range of lung cancer risk from environmental exposure to diesel exhaust ($10^{-3}$ to $10^{-5}$ deaths per lifetime exposure in the general

population) as an indication of the significance of the potential hazard (EPA 2000). These studies are limited, however, in the scope of their exposure data and in the extent of information available on smoking, a critical confounding factor (HEI 1999a). Consequently, estimates from risk models based on these studies are subject to so much uncertainty that most knowledgeable observers consider their results to be unacceptable for use in policy decisions.

The California Environmental Protection Agency's Office of Environmental Health Hazard Assessment (1998) analyzed more than 30 studies of people who worked around diesel equipment, including truck drivers, railroad workers, and equipment operators. In contrast to the EPA, they concluded that these studies provide strong evidence that long-term exposure to diesel emissions causes lung cancer, and used the railroad worker data (Garshick et al 1987, 1988; Hammond et al 1988; Woskie et al 1988a,b; Crump et al 1991) to calculate a unit risk value.

In considering the expectations for any new study, the Working Group recognized that policy makers need risk estimates that are both more precise and more certain than those that can now be generated. Although the needed gain in precision and reduction in uncertainty cannot be specified directly, a dialogue with potential users of the risk models might give researchers some direction. In addition, exploratory work and simulation can be used to assess the information gains from studies with different sample sizes and degrees of exposure misclassification.

## MODELING LUNG CANCER RISK

The Working Group considered other respiratory carcinogens to anticipate the form of a useful exposure-risk model and the data needed to develop such a model. It selected 2 example carcinogens: cigarette smoke and radon. Cigarette smoking, the cause of most lung cancers in developed countries, has been investigated extensively. Abundant epidemiologic data show that risk increases with the number of years of smoking and with the average number of cigarettes smoked over time. In analyses of several large databases, risk has been shown to vary in distinct patterns with duration and quantity of smoking, so that summary, cumulative measures are not appropriate (Burns et al 1997). For exposure to diesel exhaust, which contains some of the same carcinogens as tobacco smoke, it may also be inappropriate to use measures that aggregate duration and intensity of exposure. For smokers who quit successfully, the risk, relative to that in continuing smokers, declines with increasing duration of maintained cessation. If there is a similar decline in risk after exposure to diesel exhaust ends, then a simple constant relative risk model, as has been widely used in the past, may be inappropriate.

The Working Group also considered the example of occupational exposure to radon and its progeny. Numerous groups of underground miners have been investigated, and data have been pooled for analysis from 11 cohort studies having quantitative exposure estimates (Lubin et al 1994; National Research Council 1999). The Biological Effects of Ionizing Radiation (BEIR) VI Committee used these data to develop a model for describing lung cancer risk in relation to estimated radon exposure (National Research Council 1999). The BEIR VI Committee had 2 preferred dose-response models for the relative risk, as shown in Sidebar 2. These models describe how the excess relative risk for lung cancer resulting from exposure to radon progeny in underground mines varied with the temporal profile of exposure and age.

The relative risk models have multiple time-dependent exposure variables including time since exposure, exposure rate, and attained age. Extensive exploratory analyses had shown that the data were best described by including time-dependent exposure terms, rather than modeling that effect of exposure as constant over time. As would be anticipated, projections based on the time-dependent model for risk of lung cancer from radon exposure (TSE/AGE/DUR-CAT model) differed from projections based on the constant relative risk model (TSE/AGE/WL-CAT model). The BEIR VI Report also explored the complex consequences of time-dependent variation in the degree of error in exposure estimates, a problem inherent to the study of lung cancer risk in workers exposed to diesel emissions. It also offered extensive discussion and analyses of the combined effect of smoking and radon. The BEIR VI Committee's analysis makes clear the extensive data requirements for characterizing this joint effect with sufficient precision for policy-making purposes.

To be useful for guiding public policy, any lung cancer risk model for diesel exhaust must have scenarios that include profiles of general population exposure that extend across the full lifetime. It must also reflect the possibility of synergism with smoking, on the reasonable assumption that the population will include a large pool of smokers for the foreseeable future. The Working Group proposes that the ideal model would incorporate terms to represent duration and intensity of exposure, and allow for risk to vary with time since exposure. Separate models might be appropriate if smokers and nonsmokers were found to have substantially different risks. These considerations suggest that models estimating relative risk in relation to cumulative exposure may not suffice.

**SIDEBAR 2. TWO DOSE-RESPONSE MODELS FOR RELATIVE RISK OF LUNG CANCER DEATH FROM RADON EXPOSURE IN MINERS**

These 2 models (National Research Council 1998) describe the relative risk (RR) of lung cancer death associated with exposure to the radioactive decay products of radon; this exposure is estimated by units of working level (WL) in months. Exposures during the 5-year period immediately preceding lung cancer death are not counted as having a biological action; that is, a 5-year lag period is assumed. Each miner's exposure to radon is then summarized as $w_{5-14} + \theta_2 w_{15-24} + \theta_3 w_{25+}$, where the $w$ terms describe the exposures received in time windows 5 to 14 years before the development of lung cancer ($w_{5-14}$), 15 to 24 years before the development of lung cancer ($w_{15-24}$), and 25 or more years before the development of lung cancer ($w_{25+}$). The $\theta$ terms allow the effect of exposure to vary for each of the 3 time windows. The term $\theta_1$, not shown explicitly, is assumed to be 1. The term $\beta$ describes the increment in risk per unit of this time-weighted exposure. The increment in RR is also allowed to vary by the attained age through the term $\phi_{age}$, which describes modification of the effect of radon progeny exposure by the attained age. The values for $\phi_{age}$ show a decline in RR with increasing age. Finally, the model incorporates variation in the RR with the rate at which exposure is received. The 2 models differ in this respect: the Time Since Exposure / AGE / Working Level-CATegory model incorporates the average exposure concentration; the TSE/AGE/DUR-CAT model uses the time over which exposure is received (DURation) as an indicator of exposure rate. In both models, the term $\gamma$ indicates the exposure rate effect, showing that the effect of exposure is increased by receiving the exposure at a lower exposure rate.

TSE/AGE/WL-CAT Model

$RR = 1 + \beta \times (w_{5-14} + \theta_2 w_{15-24} + \theta_3 w_{25+}) \times \phi_{age} \times \gamma_{WL}$,

where $\beta = 0.0611$; $\theta_2 = 0.81$; $\theta_3 = 0.40$;

$\phi_{age}$ = 1.00 for age < 55 years,

0.65 for age ≥ 55 but < 65 years,

0.38 for age ≥ 65 but < 75 years, and

0.22 for age ≥ 75 years; and

$\gamma_{WL}$ = 1.00 for WL < 0.5,

0.51 for WL ≥ 0.5 but < 1.0,

0.32 for WL ≥ 1.0 but < 3.0,

0.27 for WL ≥ 3.0 but < 5.0,

0.13 for WL ≥ 5.0 but < 15.0, and

0.10 for WL ≥ 15.0.

TSE/AGE/DUR-CAT Model

$RR = 1 + \beta \times (w_{5-14} + \theta_2 w_{15-24} + \theta_3 w_{25+}) \times \phi_{age} \times \gamma_{DUR}$

where $\beta = 0.0039$; $\theta_2 = 0.76$; $\theta_3 = 0.31$;

$\phi_{age}$ = 1.00 for age < 55 years,

0.57 for age ≥ 55 but < 65 years,

0.34 for age ≥ 65 but < 75 years, and

0.28 for age ≥ 75 years; and

$\gamma_{DUR}$ = 1.00 for DUR < 5 years,

3.17 for DUR ≥ 5 but < 15 years,

5.27 for DUR ≥ 15 but < 24 years,

9.08 for DUR ≥ 25 but < 35 years, and

13.6 for DUR ≥ 35 years.

The Working Group also considered the requisite certainty of lung cancer estimates that might come from use of an epidemiologically based risk model. Uncertainty reflects not only the statistical precision of estimates, but also the gaps in knowledge that require important assumptions in model development. Some of these sources of

uncertainty are listed in Table 5 for a model that yields an estimate of risk (or excess lung cancer deaths) and a characterization of the precision of the estimate, usually in the form of (95%) confidence intervals. These confidence bounds must be widened to reflect the many uncertainties beyond the statistical considerations inherent in confidence intervals; true uncertainty bounds will invariably be much larger than confidence intervals. The BEIR VI Committee offered new statistical approaches for characterizing uncertainty; it found that uncertainty bounds widened quickly as the number of sources of uncertainty that were quantitatively considered increased.

In the Working Group's view, the degree of uncertainty to be tolerated in guiding policy development with epidemiologic evidence is inherent to the policy-making process itself. In the case of diesel exhaust, uncertainty is inevitable; it can be reduced through careful selection of study populations, rigorous exposure assessment strategies, sufficiently large sample sizes, and prospective data collection to ensure high-quality information on exposure. Studies can be designed with a view toward the degree of uncertainty that will be acceptable when the data are used for risk assessment. However, judgments as to the level of uncertainty to be tolerated are not scientific but rather reflective of the policy-making process. Here, there should be substantial, continuing dialogue between scientists and policy makers.

**Table 5.** Sources of Uncertainty in Lung Cancer Risk Estimates from a Risk Model Based on Epidemiology

**Arising from the Model**

- Exposure measurement error
- Error in ascertaining outcome
- Failure to account for confounding
- Inadequate consideration of effect modification
- Model misspecification
- Lack of precision in estimates

**Arising from Use of the Model for Quantitative Risk Assessment**

- Uncertain applicability to population of interest
- Extrapolation of exposure-time–risk relations beyond the period of observation
- Uncertainty of exposure distribution
- Differing distributions of potential modifying factors in original population and population used for estimating risks

## EXPOSURE ASSESSMENT ISSUES

### BACKGROUND

Issues related to exposure assessment arise both in designing an epidemiologic study to characterize health effects associated with diesel exhaust exposure and in assessing and estimating the risks associated with exposure to the general population and relevant subpopulations. In both contexts, characterization of exposure to diesel exhaust must be sufficiently specific to distinguish between the contributions of diesel exhaust to personal or population health effects and those of other sources. Current approaches for assessing exposure to diesel emissions are somewhat nonspecific, using measurements of both particle mass and marker compounds that have sources other than diesel emissions, such as EC. In an epidemiologic study, there is also the need to estimate exposures of participants with as little misclassification of exposure as possible. For population exposure assessment, information is needed on typical or average exposures and on the upper tail of the exposure distribution, which carries the greatest risks.

Not only is diesel exhaust a complex mixture of gases and particles, but the particulate fraction itself is also a complex mixture, with characteristics depending on many factors including the diesel fuel formulation, combustion conditions in the engine, and conditions in the ambient environment into which the exhaust is emitted. In assessing exposure to diesel exhaust, whether for individuals or for populations, the particle fraction of the aerosol is the target for measurement. However, the researcher needs to use approaches that discriminate diesel particles from the many other sources of particles in the complex and heterogeneous particle mixture in air. This discussion begins with approaches for monitoring diesel PM followed by an examination of the applicability of these data for assessing the risks of diesel exposure.

### ASSESSING EXPOSURE TO PARTICLES

Exposures to any pollutant may be assessed either indirectly using measures of ambient or microenvironmental concentrations, or directly using measures of personal exposure. Methods for measuring ambient or microenvironmental concentrations can involve complex instrumentation that gives highly time-resolved particle number or particle mass data. In addition, impaction or filter-based technologies can be used to collect integrated mass concentration data. Methods for evaluating personal exposure

are, in general, less complicated, less detailed, and less accurate. They also tend to be less expensive, so that they can be used for large samples.

The options for assessing particles in ambient air or in microenvironments include direct-reading instruments and particle collection methods. Direct-reading instruments can give time-resolved information about particle mass, number concentration, and particle numbers across different size ranges. The instruments include the tapered element oscillating microbalance, the aerodynamic particle sizer, the condensation nuclei counter, the differential mobility analyzer, the electrical aerosol analyzer, and optical particle counters such as the Climet sampling systems and MIE's Data Ram. These instruments use continuous mass analysis, light scattering, electric mobility, diffusion, or aerodynamic sizing to detect particles, and therefore do not differentiate among particles by chemical composition. Two direct-reading instruments that do give some chemical composition information are the aethalometer and an instrument that measures total particle-adsorbed PAHs. The aethalometer measures changes in absorbance at 880 nm, a wavelength found to absorb for EC with few interferences in the ambient air. The direct-reading PAH monitor measures total particle-bound PAH concentration by the photoionization of the adsorbed PAHs. Both of these instruments give time trend information on specific constituents of PM.

Particle collection methods are impaction-based or filter-based, and include single-stage and multiple-stage impactors that have a range of inlets designed to exclude particles above specific aerodynamic diameters. Particles are generally collected on substrates that can be analyzed for a range of chemical constituents. Different collection media are available, and choices among them depend on the type of analysis to be conducted. These methods include, but are not exclusive to, high-volume, cascade, low-pressure, and microorifice impactors, which collect integrated mass information over a range of particle sizes, and single-size-selective inlet impactors, which give integrated mass information for one size range. Personal monitors for particles generally use a single-size-selective inlet followed by a filter for particle collection.

## ISSUES IN EXPOSURE ASSESSMENT FOR EPIDEMIOLOGY

Risk assessment requires an estimate of the risk of lung cancer (or other health outcome) in relation to diesel exhaust exposure in one or more relevant groups of persons. Epidemiologic studies carried out to derive such estimates need a reasonably accurate assessment of exposure at the individual level. Accuracy implies that the exposure metric be both unbiased and precise, so that neither systematic nor random and nondifferential errors in exposure assessment may seriously bias an observed exposure-response relationship (Thomas et al 1993). Despite efforts to minimize such errors, they inevitably occur during an epidemiologic study, and they should be characterized and quantified as part of the study. With adequate quantitative characterization, observed exposure-response relationships may be adjusted for measurement error, or, at the very least, the results may be interpreted in light of the likely effects of the errors.

Ideally, exposure of each study subject would be measured continuously over the full, biologically relevant period of exposure. With the possible exception of studies of ionizing radiation, this ideal cannot be practicably met in studies of environmental agents and disease. An alternative is to develop a model of exposure, then link the predicted values of the model to each subject's history using exposure predictors common to both exposure information and subject history (Seixas and Checkoway 1995). (Case-control studies may ascertain exposures in a similar fashion using a job exposure matrix, or more directly by interview of subjects or proxies.) In many occupational studies, the exposure model is as simple as number of years with a specific job title, linked to the subject through the work history. In such cases, the model parameters can be simply thought of as exposure groups. Models of exposure determinants may also be much more complex, including dimensions for time, time spent in specific locations or conducting specific tasks, process parameters such as ambient temperatures, and so forth (Burstyn and Teschke 1999). In either simple or complex models, the exposure estimates generated will include both systematic and random errors.

Systematic errors in exposure data occur often because the sampling effort was designed for some objective other than that of the epidemiologic research (Ulfvarson 1983). Most commonly, exposure data are collected to ensure compliance with regulatory limits, or to evaluate the risks associated with the upper end of the risk distribution. As such, historical data can be used to estimate exposure to all study subjects only if both the sampling design and potential biases can be fully characterized (Seixas et al 1990), which is rarely possible.

Another common limitation to exposure information is the lack of sufficient information over the etiologic period of interest. Quantitative industrial hygiene measurements preceding the 1960s are rare, and since the mid-1970s routine measurement of exposure has occurred only in selected industries. As a result, quantitative information concerning exposure is frequently available only for recent

periods, and historical extrapolation of exposure must generally rely on models based on very limited data for selected segments of the population, engineering reports, or subjective professional judgments. None of these models can be validated or verified, and applying sensitivity analyses to the model assumptions is frequently the only recourse for model validation.

Some insight into the degree of random error in exposure assessment using models based on simple grouping schemes has come from random effects analysis of variance, examining the within-subject and between-subject variability in exposure data, and by examining variance components using alternative grouping (ie, modeling) strategies. In one analysis of variance with components in 165 exposure groups (defined by job and factory from numerous previous studies), the median between-subject variance (geometric standard deviation) was 1.43, indicating greater than a 4-fold range between the upper 97.5th and lower 2.5th percentiles of the group members (Kromhout et al 1993). Ten percent of the groups had over a 50-fold range in their subjects' exposure levels, indicating a large degree of between-subject variability in these exposure models based on the commonly used determinant of job title. It is likely that the short-term extremes would be moderated by averaging exposures over longer periods, which may be more relevant for chronic diseases such as lung cancer.

In addition to precision of the model-based exposure estimate, the distribution of individual exposures among cohort members is a key component of accurate exposure-response analysis. Broadening the random effects model to consider the distribution of exposure groups in a cohort study, within-subject, within-group (between-subject), and between-group variance components, and their potential effects on measurement error biases, have been explored. Kromhout and colleagues (1995) have suggested that measurement error bias in this context may be minimized by using an exposure model that maximizes the contrast or resolution of the modeled exposures, while also maximizing the precision of the group average mean.

## IS THERE A DIESEL PARTICLE SIGNATURE?

An ideal method to monitor diesel PM in the ambient air or workplace environment would capture a specific quantitative signal for diesel emissions. To date, however, no highly specific signature for diesel particles has been identified, because many of the compounds in the complex and variable mixture in diesel particles have other sources in the environments. As discussed above, diesel particles consist of EC, sulfates, a range of adsorbed hydrocarbons including PAHs and nitroarenes, and condensed liquid hydrocarbons. If a specific diesel signature could be identified, it could be used to increase the accuracy of exposure estimates made either for epidemiologic research or for exposure assessment.

Several studies have identified diesel fuel combustion as an important source of EC in outdoor air and a significant contributor to the ambient air burden of the remaining associated constituents (Gray et al 1986; Schauer et al 1999, Kleeman et al 2000). However, EC and OC are also components of emissions from gasoline combustion, wood burning, and food preparation. In a study of fuel use and particle emissions in southern California, Gray and associates (1986) estimated that sources of EC other than diesel contributed as much as 50% of the total ambient EC concentrations. Other sources for some of the PAHs found in diesel emissions include gasoline combustion, wood burning, refuse burning, asphalt, commercial solvents, tobacco smoke, and charcoal-broiled food preparation (International Agency for Research on Cancer 1973). Sulfates are an end product of coal and petroleum combustion as well as diesel exhaust and other sources (Finlayson-Pitts and Pitts 1986).

Because of the complex nature of diesel particle composition and because EC can account for as much as 90% of the total mass of diesel exhaust PM, many researchers use the EC signal as a tracer for diesel exhaust. This approach works well in occupational settings, such as underground mines (excluding coal mines), where the principal or only source of combustion particles is combusted diesel fuel and the combustion is in a well-defined environment. However, EC is less useful as a tracer for diesel exhaust in environments affected by other sources, in particular the ambient environment. This point is underscored by the results of 2 of the current HEI diesel feasibility studies (Kittelson et al, this volume; Gertler et al 2002), which report that EC is emitted from light-duty vehicles as well.

Therefore, by the most conservative estimation, the use of EC alone as the signature for diesel particles leads to a substantial misclassification of the sources of particles that will ultimately bias any subsequent risk assessment. A key issue for accurately assessing exposure to diesel exhaust for the population in general is the development of an alternative approach to capturing the diesel signature. This may involve combination of the concentrations of particular constituents of diesel exhaust (for example, EC plus specific PAHs) that can serve as a signature for diesel emissions.

## ISSUES IN EXPOSURE ASSESSMENT FOR QUANTITATIVE RISK ASSESSMENT

Accurate risk assessment requires that estimates of etiologically relevant exposures for the population or subgroups of interest be coherent with exposure estimates from which risk relationships have been derived. For diesel exhaust, the challenge is to extend risk estimates made from occupationally exposed workers, typically at higher exposure levels, to the general population. For the workers, estimates of exposure will generally be based on some combination of monitoring and modeling, while for the general population exposures will be variably derived from monitoring and partitioning of concentrations of particles, gaseous pollutants, or signature compounds. Population exposure models might also be used. While occupational exposure estimates will generally cover the etiologic window to the extent possible, population estimates will generally reflect current exposures. These differences in exposure assessment strategies for occupational groups and the general population are an unavoidable source of uncertainty.

The implications of this problem can be illustrated by considering the likely distribution of exposure to diesel exhaust for the general population, in which the majority have exposures near the low end of the diesel particle concentration distribution (Figure 8). Risk estimates based on observations made at the high end of the exposure distribution in occupational and animal studies and extrapolated down to the general population carry their own uncertainties. Basing the exposures on a poor metric such as EC further limits the reliability of these risk estimations.

In assessing whether new retrospective studies of diesel exhaust and lung cancer or other health effects can provide informative risk estimates, several key components of exposure assessment must be addressed. First, are there sufficient exposure data available over the etiologic period to provide long-term quantification of exposure? If such data are available, can potential systematic biases in these data be reasonably well understood and quantified? If there are gaps in the available data, can some model be used to fill these gaps, and can that model be validated? If historical data, or a combination of data and models, are available, can their results be directly linked to the study subjects? Finally, if a valid model can be used to estimate exposure to the study subjects, do the results of this model provide sufficient precision, and sufficient contrast (ie, range of exposures), to allow for accurate estimation of the exposure-response relationship? Populations for which these questions can be answered affirmatively may be appropriate for new, retrospective studies of the risks of exposure to diesel exhaust. If adequate responses to these questions are absent for a particular population, then it may be that only a prospective study can be accurately informative about the health risks of diesel exhaust.

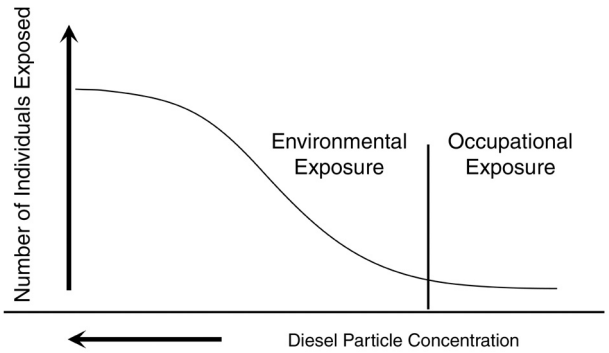



**Figure 8. Schematic of likely exposure distributions by number of individuals in the general population with environmental exposure and number of workers with occupational exposure.**

# Section 2: Technical Evaluation of the Diesel Feasibility Studies

## INTRODUCTION

Epidemiologic studies can make unique contributions to understanding environmental risks because they evaluate the effects of pollutants in humans under relevant exposure concentrations. However, researchers face methodologic challenges in carrying out such studies, including some that arise in long-term epidemiologic studies generally and others that are particular to diesel exhaust studies. Diesel emissions are a complex, temporally changing mixture that cannot easily be tracked over time. Further, cigarette smoking, a major cause of lung cancer and chronic obstructive pulmonary disease, may be a potential confounder, but smoking status of participants in a study may not be available, or substantial efforts may be needed for proper ascertainment.

The 6 feasibility studies described in this report were funded to provide insight into whether a new retrospective or prospective epidemiologic study could provide data that would improve our ability to estimate cancer risk from exposure to diesel exhaust, and whether new methods in exposure analysis would allow us to reevaluate older epidemiologic studies (HEI 1999c). Three of the feasibility studies (see Table 2) addressed possible new cohorts for investigation, and 3 studies addressed new exposure assessments.

## STUDIES TO IDENTIFY NEW COHORTS

### BOFFETTA AND COLLEAGUES

The study by Dr Paolo Boffetta and colleagues (*Cancer Risk from Diesel Emissions Exposure in Central and Eastern Europe: A Feasibility Study*) assessed the feasibility of assembling a multicenter set of historical cohorts of workers exposed to diesel emissions in the Czech Republic, Estonia, Hungary, Latvia, Lithuania, Poland, Russia, Slovakia, and Slovenia. It further assessed whether past exposure could be reconstructed for these cohorts. The occupations and industries included were workers in nonmetal mines, workers in bus transport companies, and workers in railway transport companies.

This successful feasibility study was well designed and conducted, using appropriate methods. The study documented the variations in exposure data from one country and industry to another and tested new modeling techniques for exposure reconstruction. The results of the feasibility study clearly showed that because of wide variation in the types of measurements from which historical diesel exhaust exposures could be reconstructed, the proposed study would not be a large, cohesive cohort study, but rather a collection of small studies.

The Working Group does have specific concerns about this study. There are inherent limitations in the available data. The exposure data are seriously incomplete, and there is substantial variation in data quality and availability across the cohorts. There is considerable heterogeneity among the study sites with respect to numbers of exposed workers, years since diesel exposure began, years of follow-up available, and the measured concentrations of dust, OC, and EC. Experience gained with US studies has demonstrated the difficulties of using exposure reconstruction methods; the task would be even more complicated in a multinational study with different types of data available. In Hungary, for example, current exposure measures are available only for CO and $NO_x$, with sporadic measures of total dust. Estimates of past exposures would be based on these current incomplete measures. The authors propose further monitoring to compensate for the limited data in the Slovakia magnesite mining industry; however, this may not improve estimates of past exposure.

Changes in exposure over time cannot be adequately documented for these groups. The nature and timing of changes in technology, fuel, and equipment vary from country to country. For example, in Estonia Hungarian buses were used exclusively until 1991, at which time used buses, with quite different emission characteristics, were imported from Sweden. Such dramatic changes further complicate the reconstruction of past exposures, which would be based on estimates from current exposures to emissions from the newer equipment. In addition, Eastern European diesel engines are older than those used in Western Europe or the United States, and exposures to emissions from this older equipment may not be relevant to current risks in areas using newer equipment. The authors do not fully describe the completeness and

accuracy of historical records from which additional data are needed for reconstruction of past exposure, and it is not clear whether such data are sufficiently similar from one country or industry to another. Some information about the historical background concentrations of diesel exhaust would be obtained from interviews with "knowledgeable local people." It is not clear how such information could be translated into quantitative exposure estimates. No data on active and passive smoking are available.

The investigators' exposure reconstruction model did not work well for current data. The modeling methods were accurate for only 2 of 5 job groups, and the modeling inaccuracies resulted in considerable overestimation of exposure. The model is likely to be less precise for historical data, and might be even less accurate in a larger study.

Uncertainties in exposures and in estimates are not reported. Without some knowledge of exposure measurement error, it is impossible to adjust for possible attenuation and compare estimates with those from other studies. Without knowledge of the uncertainty in an estimate, it is also not possible to provide a range of values supported by the data (eg, a confidence interval) or to give the study its appropriate weight when it is combined with other studies. In summary, while the proposed retrospective cohort study of workers exposed to diesel emissions is technically feasible, it would be of limited value for QRA.

**FINKELSTEIN AND VERMA**

The study by Drs Murray Finkelstein and Dave Verma (*Cancer Risk from Diesel Exhaust Exposure in the Canadian Railroad Industry: A Feasibility Study*) examined the possibility of determining the relationship between diesel exhaust exposure and lung cancer mortality in Canadian railroad workers. Their proposed cohort is approximately 150,000 workers who were employed by the Canadian National Railway and the Canadian Pacific Railway between 1983 and 2000, with some exposure data available since 1960, the date of complete dieselization of locomotives in Canada. In this cohort 1,080 lung cancer deaths are expected.

The investigators examined the suitability of data for about 74,000 Canadian National workers. Demographic information came from railway records; medical records provided information on health status during employment, including smoking history. Job histories allowed division into 5 groupings of jobs with similar exposures (no exposure, exposure not classifiable, low exposure, moderate exposure, and high exposure). The high-exposure job group included machinists, mechanical laborers, engineers, conductors, brakemen, flagmen, and yard helpers. Inclusion of the Canadian Pacific workers would double the cohort. The data available for the Canadian Pacific workers are expected to be about the same as those for the Canadian National workers.

In a full study, historical reconstruction of exposure back to 1960 would be necessary because industrial hygiene measurements did not begin until the mid-1990s. The investigators made 155 new EC measurements to provide insight into exposures by job category. Those with the highest exposures were engineers and conductors in trailing locomotives, followed by shop workers, followed by workers in leading locomotives. Substantial changes in work practices and job categories would, however, make re-creation of exposures difficult. Elemental carbon levels were generally low (0 to 25 $\mu g/m^3$ in the 155 samples) and were nondetectable in a large fraction. Exposures were close to ambient background levels.

Drs Finkelstein and Verma's well-conducted feasibility study addresses many of the key design issues in exposure analyses. Their industrial hygiene measurements extend understanding of exposure of railway workers to diesel particles, at least as represented by EC. The methods and study design were appropriate to evaluate feasibility. However, the potential success of the proposed full-scale study is limited by several difficulties.

Historical data on exposure levels are not available, necessitating reconstruction of earlier exposures. The numerous assumptions inevitably required to reconstruct historical exposures makes them uncertain. Even a modest misclassification of exposure greatly reduces statistical power, which is therefore probably overestimated in this report. Changes in job categories and changes in exposures within a job category will likely contribute to the substantial misclassification problem. In addition, the investigators did not address uncertainty and its consequences for bias in risk estimates.

The current EC concentrations found by the authors are close to and overlap with those in the urban ambient environment, making results relevant to the general population. However, the current on-the-job levels of EC are so low that off-the-job exposures may be important confounders, and the range of exposure may not be sufficient to be informative. Exposures by job title are not very different, and within-category exposures are not available.

Potential confounding from active tobacco smoking could be assessed from historical smoking data. However, potential confounding from asbestos exposure was not considered and might be difficult to control. Exposure of mechanical trade workers, a large subset of this cohort, to other potential carcinogens such as silica, asbestos, and welding fumes may prevent inclusion of this group. Workers hired before 1960, when dieselization was com-

pleted, will have been exposed to emissions from coal- or oil-burning steam locomotives. The difficulty of correcting for this exposure may also lead to exclusion of these workers from the cohort.

Other difficulties with this study include the question of applicability of past exposures in assessing future risk, owing to changes in diesel engine technologies and fuels. In addition, since the diesel engines and fuels used in locomotives are different from those used in on-road vehicles, emissions and health effects observed from railway studies may not be applicable to the problem of exposure to highway diesel emissions.

A strength of this study was the opportunity to measure emissions from older locomotives, yielding data that could be valuable in understanding historical exposures and their relevance to modern engines. However, information on historical fuel characteristics and the replication of a historical fuel would be necessary to accurately extrapolate to historical exposures.

Canadian railway workers, because they are a large population for whom demographic and medical records are available, might be a valuable cohort for a study of the relationship between exposure to diesel PM and lung cancer. However, because of the difficulty of re-creating historical exposures and questions regarding the relevance of historical exposures to assessing current and future risks, the results of the proposed study may not be useful for QRA. The authors claim that the study would be able to detect relative risks of 1.15 to 1.20 with sufficient power, but we question whether this level of sensitivity could be attained. The potential for the study to contribute to a QRA was not addressed. At a minimum, a more detailed demonstration that the quantitative relationship (slope) between lung cancer and diesel particulate exposure can be extracted from the proposed cohort study is required, including the uncertainties in the slope resulting from the uncertainties in exposure.

Finally, given the differences in engines, fuels, and operating conditions, it is not clear whether exposures of railroad workers can be directly extrapolated to exposures of the general population to diesel emissions from the light-duty and heavy-duty fleet.

## GARSHICK AND COLLEAGUES

This study by Dr Eric Garshick and colleagues (*Quantitative Assessment of Lung Cancer Risk from Diesel Exhaust Exposure in the US Trucking Industry: A Feasibility Study*) explored the most relevant dimensions of exposure analysis in regard to the potential for carrying out an informative study of diesel-exposed workers. The investigators focused on the US trucking industry and were successful in obtaining the cooperation of 4 large trucking companies as well as the Teamsters Union. Their evaluation of the employment records suggested that the structure and depth of information was sufficient to construct a cohort of workers with high potential for exposure to diesel exhaust. The pension records of the Teamsters Union were shown to be useful to document aspects of employment, particularly duration. The cooperation of the companies and the union raises the likelihood that they would cooperate in a larger study.

Diesel equipment and use patterns have varied over time, and no companies have systematically monitored how the changes have affected exposures. The investigators propose a model-based exposure estimation approach, as no reasonable alternative is available. They set out strategies that could be used along with general exposure models. The model that would be used to support any cohort study that involves past exposures would need to address exposures across a matrix defined by job type, terminal size, location, and characteristics, as well as calendar interval. A model of this sort might be developed, though it would require multiple assumptions. Model results would be subject to substantial uncertainty, and estimates for individual workers would not be fully independent, reducing statistical power. The Working Group noted some specific weaknesses in the details of the proposed approach, but understands the need for the type of proposed general exposure model.

The report provides the results of fewer than 100 separate concentration and exposure measurements made in the feasibility study. The levels, measured at the Atlanta and New England trucking terminals, were in general quite low (for example, mean $PM_{10}$ concentrations ranged from 51 to 278 $\mu g/m^3$ in the Atlanta terminal, as compared with a mean concentration of 36 $\mu g/m^3$ at an upwind site). If, in fact, levels were this low for other terminals and workers, the likelihood of detecting an excess risk seems small. It might, of course, be useful to know that no risk can be detected at certain low exposure levels.

The investigators' overall conclusion from this study is that an investigation of lung cancer risk in the trucking industry is feasible. They propose 3 alternative study designs and offer a power calculation for a nested case-control study, conducted within a cohort. The investigators calculate that 400 lung cancer cases, each having 10 controls, could detect a relative risk of 1.4 with 80% power, assuming that 25% of the controls are in a diesel-exposed job. However, other studies (reviewed in HEI 1995 and Mauderly 2000) have suggested that the actual relative risk is likely to be less than 1.4. Furthermore, this power calculation

appears to refer to the ability to identify a lung cancer hazard attributable to diesel exhaust, rather than the precision with which lung cancer risk could be estimated in relation to exposure. The calculation also does not acknowledge the uncertainty that arises from imputation. Finally, the use of 10 controls for each case is excessive; 2 or 3 controls per case is more standard. Using a large number of controls in an attempt to obtain more data can divert effort and attention and ultimately reduce data quality. Further, the use of 10 controls rather than 3 reduces the variance of the difference in rates by only 17%.

This feasibility study provides valuable insight into the possibility of carrying out a new epidemiologic study of diesel-exposed workers in the trucking industry. Garshick and colleagues demonstrate the feasibility of carrying out such a study; they have shown that the trucking industry and the Teamsters Union would cooperate and that strategies and databases are available to estimate exposures. As a result, the Working Group was pleased to learn of the recent funding received by the investigators based on this pilot work, which will likely be a useful addition to the identification of the hazard of exposure to diesel exhaust. However, their work, including the pilot measurements of exposures, suggests that the inherent exposure measurement error in a retrospective cohort study or a case-control study would make it unlikely to produce precise enough risk estimates for current regulatory needs.

## EXPOSURE STUDIES

### KITTELSON AND COLLEAGUES

This study by Dr David Kittelson and colleagues (*Measurement of Diesel Aerosol Exposure: A Feasibility Study*) evaluated the feasibility of measuring EC, mass of black carbon, and surface area, number concentration, and size distribution of airborne diesel PM in 3 populations occupationally exposed to relatively low levels of diesel exhaust. The populations were bus drivers, parking garage attendants, and mechanics. The investigators used several methods to measure diesel PM. The different instruments and methods gave results that were consistent among the methods, detecting diesel PM at the low end of typical occupational exposures (which are, in turn, at the high end of typical ambient exposures). The 3 occupational groups had similar exposures to airborne diesel PM when analyzed by mass, but different exposures when analyzed by particle surface area and particle number distributions.

Through their considerable expertise in measurement instrumentation, the investigators analyzed diesel aerosols by a variety of methods that capture different components of the mixture. However, this report falls short of identifying a diesel signature to differentiate exposure to diesel exhaust from exposure to gasoline exhaust. Such identification is central to evaluating exposure-response from field studies, as is determining whether the signature changes as the aerosol ages.

The issue of EC as a specific marker of diesel PM is not resolved. Elemental carbon accounts for a significant fraction of diesel PM emissions, but only a small percentage of the automobile fleet is diesel-powered. In most nonoccupational settings, gasoline-powered vehicles are likely to dominate the vehicle fleet contributing to the total particle burden. So, even if the EC fraction of the particles emitted from the tailpipes of gasoline-powered vehicles is small, the fraction of gasoline vehicles present is so large that the relative contribution to total EC by these vehicles may be large as well. Therefore, EC by itself is not a reliable specific marker for diesel emissions. The same situation exists for use of individual PAHs and other components.

Using an aethalometer, the investigators found variability in levels of black carbon over time in the parking garage, and they concluded that this reflected variability in traffic during the day. However, since most of the traffic in the garage was automobile traffic, and most automobiles are not diesel-fueled, this suggests the aethalometer was capturing more than only diesel contributions to black carbon. There was a close correspondence between the averaged black carbon concentration and the integrated EC concentration in this location, which implies that the EC measurement is not specific to diesel.

Kittelson and colleagues used the Scanning Mobility Particle Sizer and the Ultrafine Condensation Particle Counter. These instruments both count particles in the ultrafine size range, but their actual ranges are slightly different. Although these instruments appear to capture the variability of particle number concentration, the investigators note that the measurements they obtain reflect the presence of particles from all vehicles, both diesel and gasoline-powered, as would be expected.

A photoelectric aerosol sensor and a diffusion charger were used in tandem to measure PAH adsorbed to the surface of PM, which the investigators used as a surrogate for diesel PM. However, there are many sources of PAH in the environment including, but not limited to, gasoline-powered vehicles and coal, wood, and tobacco combustion.

The investigators state that the reported techniques were useful for measuring (generic) particle surface area concentration, which may be important in a health effects context. However, there does not appear to be a diesel focus to this measurement. The investigators also state that instruments

could detect tobacco-related smoking events in the parking garage on a day when the parking attendants reported smoking (illustrated in Figure 14 of their report). The effect of tobacco smoke on measurements seems to argue against their use as specific indicators of diesel PM.

A strength of the study is the amassing of data obtained from different instruments, providing the potential for a simple and meaningful index of exposure. For example, multivariate analyses could be developed to nominate one or more diesel signatures. Measurements aligned reasonably well in time, and a principal components decomposition (after taking logs and setting aside outliers) could be done on cross-sectional time slices so that the principal component decompositions could be related over time. Such an approach has the potential to summarize the ensemble of exposure measurements, to elucidate how the relationships among measurements change over time, and, possibly, to suggest a candidate health-effect signature for diesel exhaust.

These investigators have considerable expertise in making real-time and integrated measurements of PM. Because of the strong design and careful execution of this study, use of information in this report, or advice from these investigators, would benefit design of other epidemiologic studies.

## ZIELINSKA AND COLLEAGUES

The main aims of this study by Dr Barbara Zielinska and colleagues (*Measuring Diesel Emissions Exposure in Underground Mines: A Feasibility Study*) were to improve the characterization of current diesel exhaust emissions, and to use that characterization for source apportionment. The investigators were able to assess source exposure in underground ore mine workers with reasonable accuracy. To achieve this they (1) characterized ambient air in the mines with respect to PM composition and organics in both the gas and condensed phases (eg, VOC, SVOC, and particle-bound organic compounds); (2) characterized the composition of emissions from sources that contribute to PM in mines; and (3) conducted source apportionment modeling to identify the source of the PM in the mine.

The proposed use of miners as a study population has some important strengths. The closed nature of the mines limits workers' exposures to relatively few sources, and the PM exposures in the mines are dominated by diesel sources. Therefore, there is likely to be little variability in the sources contributing to the exposures. The exposures are well above background, and thus the impact of variations in the background is minimal. Also, there is not likely to have been significant variation in exposure over time, since the same diesel engines are maintained over a long period of time, and estimation of changes over time may be more feasible than in other settings.

Use of miners as a study population, however, also has drawbacks. The high concentrations of these occupational exposures to diesel emissions (ranging from 500 to 2700 µg/$m^3$ for diesel PM) introduce potential problems in extrapolating to ambient concentrations, particularly if there is any nonlinearity in the relationship between exposure and health effects. Further complicating the extrapolation is the question of whether diesel emissions in mines are similar in their composition and aging to diesel emissions from trucks, buses, and other motor vehicles in situations more typical of general population exposure (for example, on the open road and in areas of above-ground construction). Fuels and engines, as well as conditions of mixing and particle growth, which could alter the composition of PM, could continue to change substantially over time, as they have changed over past decades. Concurrent high levels of ore dust in the mines also complicate interpretation. Further receptor modeling techniques to complement the chemical mass balance (CMB) approach used in this study would strengthen it, as would extending results to a source-oriented approach.

As with the other feasibility studies, the treatment of uncertainty is important. The CMB method used integrates uncertainty into the results. The study would be improved with the diagnosis of causes of uncertainty so that they can be addressed if a new study is undertaken. Uncertainty estimates for all modeling and measurement results would improve the study. For example, the investigators found that as they increased the number of compounds in their CMB calculations (eg, included EC), the predicted results of the model got worse. Generally, this should not be the case, but it can occur if there are significant uncertainties (and errors) in the profiles. A thorough uncertainty analysis might identify and quantify the source of the major uncertainties. Knowledge of those uncertainties could then be used to improve future studies. Several other issues need to be addressed: What level of increase in lung cancer could be detected with a high probability by a study in this miner population? What uncertainties are introduced in conducting a retrospective exposure analysis? Could the results from this mine be extended to other mines, and if so, what sorts of additional measurements would be needed?

The approach the investigators took to start the task is appropriate. They used both ambient samplers and personal monitors. Further, they sampled the primary sources that might contribute to PM levels in the mine: diesel engines, ore dust, and drill oil. From this, they developed

source profiles not only for diesel particles, but also for SVOC and VOC. Next, they used CMB modeling to assess the fraction of PM attributable to each source. The investigators found that diesels contributed a majority of the PM in the mines; ore dust was the second most common source, and oil was the third. The source characterization appears to be excellent, and the investigators have provided a unique set of data for the mine emissions. The SVOC components may be of particular interest to other investigators, although detailed analysis of all the emission components will be of great interest. Many other exposure assessment projects lack such measurements.

The current report, however, cannot be viewed as conclusive. Some aspects of the CMB modeling raise cautionary flags. For example, the addition of oil to the model caused it to give erroneous predictions. Similarly, using concentrations of EC and OC in the CMB model resulted in poorer performance, even though EC and OC are a large fraction of the diesel emissions and might be expected to help improve allocations to diesel sources.

The investigators found a poor correlation between the results of a photoelectric aerosol sampler and actual measurements ($r = 0.07$). Nevertheless, they described a relationship for use of the photoelectric aerosol sampler and suggested its data be used as a surrogate for diesel PM. Although the results obtained with this sampler suggest the presence of diesel exhaust, they do not seem to be a reliable indicator for total diesel PM.

In summary, the study accomplished the major objective of better characterizing diesel emissions, including nonparticulate components, and assessing exposure of workers in the mine to those emissions. If the approach were expanded to other mines to obtain more measurements, and a good source characterization were developed, then an expanded exposure analysis would be feasible, though not directly usable for estimating retrospective exposures. A substantial measurement program would be needed. The CMB application provides a reasonable route for exposure apportionment, but the results (particularly the uncertainties and sensitivities) need more explanation.

## GERTLER AND COLLEAGUES

This study by Dr Alan Gertler and colleagues (*Real-World Particulate Matter and Gaseous Emissions from Motor Vehicles in a Highway Tunnel*) determined the contribution of diesel and gasoline engine emissions to the ambient mixture of particulate and gaseous emissions in air samples from a busy highway tunnel (Gertler et al 2002). The authors also determined particle number and chemically speciated, size-segregated particle distributions in heavy-duty and light-duty vehicles traveling through the tunnel and compared the chemical profiles obtained in their study with the results of earlier tunnel studies to assess how diesel emissions have changed with improved engine design. A total of 10,061 vehicles were observed (6,888 automobiles, 290 medium-duty trucks, and 2,883 large trucks). For each run, the investigators recorded the number of light-duty and heavy-duty vehicles traversing the tunnel, their ages, and their average speed. They counted the numbers of vehicles at days and times when the distribution of the 2 classes varied. For example, heavy-duty vehicles (large trucks) dominated late-night periods, while light-duty vehicles (automobiles and medium-duty trucks) dominated at midday and during weekends. This allowed the investigators to estimate the average contributions to the ambient air for each class.

Two of the aims of the study were source apportionment of light-duty and heavy-duty vehicle emissions and improved understanding of ultrafine particle emissions. The investigators calculated emission levels of a broad range of air pollutants by measuring airflow into and out of the tunnel with extensive and precise instrumentation. They measured $PM_{2.5}$, $PM_{10}$, size-fractionated particle mass, concentrations of selected elements, PAHs, $C_8$ to $C_{20}$ hydrocarbons, CO, $CO_2$, total hydrocarbons, NO, and $NO_x$. They calculated the emission rates for several pollutants as a function of the concentrations measured at the tunnel inlet and outlet, the volumetric airflow in the tunnel, and the number of vehicles traversing the tunnel.

Heavy-duty vehicle particle emission rates were significantly higher than those from light-duty vehicles. The ultrafine fraction of PM, which contained sulfur as a major component, dominated particle number concentrations for both heavy-duty and light-duty vehicles. Comparison of data in the current study with those from a 1993 study in the same tunnel indicated that heavy-duty vehicle emission rates for particle mass, $C_8$ through $C_{20}$ hydrocarbons, CO, and $CO_2$ decreased substantially. Light-duty $PM_{2.5}$ mass emission rates were 8 to 10 times lower than heavy-duty $PM_{2.5}$ mass emission rates. However, because light-duty vehicles dominate the overall driving fleet, the authors suggest that the PM contribution from those vehicles may exceed that of the heavy-duty fleet. However, this conclusion is uncertain because the method used to estimate the light-duty vehicles' particulate emissions from the tunnel measurements does not allow a precise determination of their magnitude. The investigators compared the levels of EC and OC to determine $PM_{2.5}$ profiles in the tunnel air. They concluded that EC and OC levels might not accurately determine the contribution of gasoline and

diesel vehicles to PM emissions. In agreement with Kittelson and colleagues (described earlier), Gertler and colleagues question the use of EC levels as a surrogate for diesel-related particles in many investigations.

The authors suggest that $NO_x$ emissions should have decreased with new regulations on diesel emissions, implemented after 1992. Heavy-duty vehicle $NO_x$ emissions in the current study were unchanged from results in a 1992 tunnel study. Because $CO_2$ emissions decreased (an indicator of improved fuel economy), the authors suggest that the newer diesel engines are being operated in a manner designed to improve fuel economy at the cost of $NO_x$ emissions, which are precursors of ground-level ozone.

The study by Gertler and colleagues substantially adds to our knowledge of the emission rates, particle size distribution, and chemical composition of diesel and gasoline engine PM under on-road driving conditions and presents new data on gaseous emissions. For comparison, the investigators reported data for light-duty gasoline engines. They used state-of-the-art instrumentation and minimized extraneous influences on emissions by performing the study in a highway tunnel.

## SUMMARY

A major challenge of these, or any, studies specifically focused on diesel PM is that diesel PM is a subset of the larger category of ambient PM. Like PM from many other sources, diesel PM is a complex mixture of unburned fuel and combustion products, and its physical configurations and chemical compositions vary substantially with combustion and atmospheric conditions. The size distribution of diesel PM falls within the size range for PM from other combustion processes. Finding a specific tracer for diesel PM is therefore difficult and cannot depend on particle size alone.

A similar challenge lies in the choice of a metric to assess health effects. Epidemiologic evidence generally points to the smaller particles as being the agents of injury, and diesel particles tend to be in the smaller, respirable size range. However, it is unknown which aspect of small particles is responsible for their deleterious effects: Is it the size, chemical composition, surface area, shape, or some combination of these attributes? In order to understand the key determinants of toxicity, it is necessary to investigate all of these areas and related matters.

The use of EC as a marker for diesel exhaust is a central issue for most diesel exposure assessments. Although diesel sources are often the major contributor of EC in the ambient air, there is usually a contribution by products of combustion from other sources, including gasoline engines, wood burning, etc, most of which produce some EC. The contribution of diesel exhaust needs to be confirmed in each case, perhaps by the use of other tracers. The uncertainty introduced by the contribution of non-diesel sources to ambient EC requires assessment.

The Diesel Epidemiology Working Group believes that an important task at this time is to develop improved exposure-response data for risk assessment, rather than to test the hypothesis that diesel exhaust causes lung cancer. It is not clear that the proposed cohort studies whose feasibility is examined here would improve the basis for QRA. These studies would, on the whole, probably add to the evidence regarding the carcinogenicity of diesel exhaust, but do little to quantify such a relationship.

# Section 3: Conclusions and Research Recommendations

## INTRODUCTION

This report begins by setting out a framework that anticipates future needs for quantitative assessments of the risk of lung cancer and possibly other diseases from long-term exposure to diesel emissions. Epidemiologic studies can contribute to risk assessments by providing information on the hazards of agents of concern, on the quantitative relationship between exposure and disease risk, and on population patterns of exposure. Expert panels have concluded that diesel particles are a probable human carcinogen, so the emphasis in risk assessment has shifted to quantifying the burden of lung cancer and other diseases associated with exposure to diesel emissions. This shift requires quantitative estimates of the relationship between disease risk and the level and conditions of exposure. Epidemiologic studies of diesel-exposed populations can provide the needed risk estimates, but only if populations with exposures relevant to the general population are studied, exposures are accurately estimated, data on potential confounding factors are available, and sufficient data are accrued during follow-up for estimation of risks with the needed precision.

In recognition of the need for improved QRA of diesel emissions, the Health Effects Institute requested applications for feasibility studies to identify new, potential study populations and to develop new or improved methods for exposure assessment, which would require advances in both measurement of exposures and measurement of health outcomes in exposed populations. These studies of the feasibility of new epidemiologic investigations were well executed. They illustrated the difficulties faced by epidemiologic researchers in finding suitable populations for study—populations with adequate exposure data available, having exposures in a range likely to be informative and relevant to ambient levels, having data available on potential confounding and modifying factors, and offering sufficiently large sample sizes. Although populations were identified that might contribute to our understanding of occupational exposure and diesel exhaust, the Working Group concluded that full studies of the proposed groups could not be justified for the purpose of generating sufficiently accurate exposure-response relationships.

Three of the feasibility studies addressed methods for exposure assessment and characterization of diesel particles. These studies are complementary to the proposals for new populations to be investigated, as improved exposure assessment methods are needed to reduce misclassification of exposure in epidemiologic studies and enhance precision. The findings confirm that we still lack sufficiently specific methods for characterizing exposures to diesel exhaust. Some specific markers for diesel exposure have been proposed, eg, EC, but the sensitivity and specificity of these markers appear to vary from environment to environment. Thus, there are serious problems in improving knowledge regarding both exposure and response to diesel emission. Nonetheless, the insights from these feasibility studies suggest areas for future investigation.

## RECOMMENDATIONS

The Working Group does not recommend initiation of a new cohort study of any of the proposed populations at this time. The Working Group concluded that available methods and populations would not result in a sufficient gain in certainty of risk estimates over those already in use, and the funding commitment would be substantial for a prospective cohort study. To meet these challenges, the Working Group has identified activities that could enhance risk assessments over the next 1 to 2 years (short-term activities), 3 to 10 years (medium-term activities), and 10 to 20 years (long-term activities).

### SHORT-TERM ACTIVITIES

Substantial collection of data on levels and characteristics of PM is now in progress across the United States and elsewhere. Studies funded by HEI, the Supersites program, and projects of the EPA PM centers are generating large quantities of data that could be informative regarding diesel emissions and ambient air pollution.

Proposed short-term activities largely relate to exposure assessment and the possibility of defining a signature that is characteristic of diesel emissions. Recommendations are also offered to further understanding of certain design issues for epidemiologic studies and for enhancing any new studies funded by the Health Effects Institute.

The Working Group agrees with the study investigators that EC may be a useful indicator of *occupational* exposure to diesel exhaust when diesel is the dominant source of PM emissions. However, additional surrogates should also be explored because EC as the only marker lacks the sensitivity and specificity necessary as a signature for *ambient* exposure. Improved surrogates might be used to enhance exposure assessment for past studies, depending on data and sample availability. Validated surrogates could strengthen future epidemiologic studies, and surrogates would also have application in population exposure assessments. Signature characteristics of diesel particles need to be identified and applied in exposure assessments for epidemiologic studies and also in populations to which risk estimates are to be transferred. Though the Working Group cautions against immediately initiating epidemiologic studies to characterize the exposure-response relationship, it proposes the following specific activities around exposure assessment:

- Convening a workshop to plan approaches for exploring emissions and monitoring data to identify signatures of diesel PM. Substantial exposure data sets have already been developed and continue to be developed by HEI and other organizations. These might be explored with contemporary techniques for data exploration (eg, data mining). This workshop would bring together the investigators who have collected data on diesel particles and experts in instrumentation, health effects, data analysis, and risk assessment. An anticipated product would be a listing of available data and potentially informative approaches for analysis. HEI intends to follow the workshop with a research initiative to conduct analyses of such data sets to identify a signature. The specific aims of the workshop would include (1) identifying possible signatures, eg, EC or other chemical components of diesel exhaust; (2) identifying previously collected data and current data collection that would be used for this purpose; (3) specifying additional measurements that might be useful; and (4) addressing approaches for data analysis. Possible resulting initiatives might include plans for assembling databases and archiving and sharing filters with diesel PM. Funding initiatives might support additional data collection, specimen analysis, and data analysis on existing sample collection efforts. The workshop might also identify new high-priority feasibility studies to complement those reported here.
- Standard characterization of diesel particles. The Working Group suggests that diesel particles collected under investigations funded by HEI (see HEI 2000), whether in laboratory or population contexts, undergo a standard characterization.
- Regional PM characterization. The Supersites initiative of the EPA may present an opportunity to learn more about diesel particles with targeted, additional data collection. The Working Group recommends a coordinated characterization of the organic compounds in PM using a similar (traceable) approach for each, such that a regional organic PM characterization is possible. This should be integrated into an analysis of the filters taken by all of the states at the speciation sites, not only the Supersites.
- Calculation of sample size needs for future epidemiologic studies to characterize exposure-risk relationships. Power considerations included in the feasibility study reports do not address quantification of risk to the degree of precision that would be necessary for QRA. The calculations also do not acknowledge the uncertainty that arises from using estimated exposure values. The Working Group suggests that HEI take the lead in developing these calculations, a task to be accomplished using simulation methods or other approaches.

## MEDIUM-TERM ACTIVITIES

The Working Group's recommendations for medium-term activities are based in part on likely new epidemiologic studies as well as anticipated advances in understanding of the molecular and cellular biology of lung cancer. They also build on the results anticipated from the short-term activities. The Working Group recommends:

- Continued evaluation of opportunities to enhance exposure assessment approaches, including, for example, analysis of filters from special monitoring initiatives (eg, Supersites and the speciation network) and single particle analysis based on mass spectroscopy.
- Evaluation of opportunities to apply new approaches for assessing signatures in epidemiologic studies in progress.
- Assessment of possible biomarkers for lung cancers associated with exposure to diesel emissions.
- Further research directed at effects other than cancer, building on findings of studies funded under HEI's RFA 00-1 or other studies.

### LONG-TERM ACTIVITIES

In making recommendations for activities extending beyond 10 years, the Working Group assumes that QRA will remain central in managing the potential risks of diesel emissions. The Working Group recommends:

- Designing and implementing population surveillance instruments for exposure to diesel exhaust, using validated source signatures. Strategies for surveillance might also include modeling of ambient air data and biomarkers.
- Periodically performing source signature characterization to track changes in diesel emissions as technology and fuels evolve. This might be accomplished with periodic tunnel studies or laboratory studies.
- Undertaking further laboratory and population-based studies on the risk of lung cancer or other diseases. These studies might use biomarkers of dose and explore gene-environment interactions.

Although the Working Group does not recommend a new epidemiologic study at present, it may be appropriate to implement a new study as research methods evolve for exposure and outcome assessment. We discuss some of the needed elements for such a study in Appendix A. We do not foresee a waning of interest in the possible adverse effects of diesel exhaust and their quantitative relations with exposure.

There are obvious methodologic challenges to conducting such a study. These include obtaining complete and accurate exposure assessment, identifying an exposure marker that is a signature for diesel exhaust and for no other airborne exposures (and is likely to continue to reflect diesel emissions in the future), maintaining sufficiently complete follow-up over a 25-year period, and measuring other critical exposures. Diesel engine technology is changing, and emissions from diesel exhaust are being reduced; consequently, it is not clear that the exposures and outcomes measured in a prospective cohort study started today would have relevance for the exposures and health effects present 25 years from today. Over time, we anticipate improved methods for exposure assessment and for use of biomarkers, a focus of our proposed short-term and medium-term agendas. With better research tools, it may become feasible to carry out more informative epidemiologic studies in the future, although future researchers may still face the challenges of continuously changing technology and emissions.

## SUMMARY

Since its founding, HEI has addressed the risk of lung cancer associated with exposure to diesel exhaust. With the shift in emphasis toward quantifying the potential burden of lung cancer associated with diesel exhaust, HEI has evaluated the available epidemiologic evidence and sought ways to enhance these data. It has convened expert panels and carried out workshops for this purpose. The feasibility studies discussed in this report, funded under RFA 98-3, were one outcome of this process. The findings of these studies contribute to our understanding of the potential for epidemiologic research to quantify disease risks associated with diesel emissions. The Working Group does not recommend proceeding with full studies of the populations considered here, largely because of concern about the lack of available data from which one could estimate past exposures. As concern for the public health consequences of diesel exposure is likely to be maintained, consideration might be given to establishing populations for prospective observation so that exposure assessment with sufficient validity could be implemented. The activities recommended by the Working Group should enhance epidemiologic studies in progress and planning for future research.

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# Appendix A. Protocol for Prospective Cohort Study of Diesel Exhaust Exposure and Chronic Disease Outcomes

The Diesel Epidemiology Working Group has determined that for the purpose of QRA of the chronic health effects of diesel exhaust, the best strategy may be to design and conduct a new prospective occupational cohort study based on recent workers. Though such a study would require a lengthy follow-up period, it should begin to yield important information during that period. The Working Group also recognizes that no single study can meet all the ideal standards for such a study. Therefore, the protocol will need to be specific about which problems will drive the study design. Here, we comment on some of the critical issues.

The Working Group has observed that the most serious difficulty with existing diesel exhaust studies has been exposure assessment; this is often the case in other areas of QRA of health effects. Difficulty in obtaining appropriate and useful exposure data will continue to be a problem for any new study of diesel exhaust, especially because of the changing nature of diesel emissions with changes in engine design, fuels, and control technologies. Therefore, this component of any new study must be given special attention.

A prospective occupational cohort study designed to produce a QRA of lung cancer and other chronic diseases resulting from exposure to diesel exhaust must include a large, stable population of workers with known exposure to diesel exhaust. Occupational cohorts most appropriate for such a study would be truckers, railroad workers, agricultural workers, miners, drivers of other diesel-powered vehicles, and mechanics working with diesel engines. Ideally, extensive baseline exposure data, as well as complete information on the cohort members, including demographics and work history, should be available for the occupational cohort selected. For retrospective studies of diesel exhaust exposure conducted to date, such as studies of US truckers or US railroad workers, exposure data are known to be incomplete, thus limiting the quality and quantity of baseline data available. Since a major purpose of obtaining a QRA is to specify the risk to the general population from exposure to diesel exhaust, any occupational cohort selected for such a study should have exposure similar to that of the general population. In the United States, truckers may be the most appropriate occupational cohort from which to estimate general population risk.

Smoking history is critical to risk assessment, and the ideal prospective study should measure tobacco use and exposure to environmental tobacco exposure. It will be important to consider also measuring exposure to other occupational lung carcinogens, particularly radon and asbestos. Dietary practices over time should be documented, as well as nonoccupational exposure to lung carcinogens such as pesticides and cleaning compounds.

The protocol should delineate data available from existing records, as well as data to be obtained from questionnaires, serum samples, and personal and environmental exposure measurements. These measurements should be made periodically, over the span of the prospective study. Ideally, the study should be designed to continue for at least 25 years, although critically important results should be available much sooner, perhaps within 5 years. Methods to be used to determine dose, duration, and timing of exposure should be clearly stated.

The ideal study should define each industry and occupational group to be studied, their sources of exposure to diesel exhaust, and their projected levels of exposure. The records available to characterize the overall work group and each individual worker (ie, demographics and employment history by industry and occupation) should be described completely. The study also should describe the information that will have to be obtained from interviews (such as previous work history and demographics not available from the workplace) in order to define the industry and occupational group and its individual members prior to the initiation of the study. A rationale should be given for inclusion of each group and for exclusion of any occupational group in the same industry.

The study should define one or more populations and subpopulations to be included, particularly the most susceptible populations; provide available data about the populations, including any data available from current or past studies; provide a thorough description of the stability or mobility of the populations and the potential for following

the study subjects through the duration of the study; and demonstrate that the investigation will have access to the populations throughout the period of follow-up. There should be reason to expect that the exposures to be studied will continue for much of the study group through much of the study period. There should be means for continuing lifetime follow-up of subjects whose status changes, for example, by changing jobs or industry, or by retirement. Expected completion of follow-up should be addressed in some detail.

The ideal study should define the sources of diesel exposure to be included in the study and measures of diesel exposure to be utilized. The study should include a combination of direct measurements via personal monitors and environmental measurements via area monitors, and the methods and timing of obtaining exposure measurements should be specified. Personal exposure measurement is less complicated, consisting primarily of samples captured on various filter materials. Ambient and environmental monitoring can be done using direct-reading instruments or particle collection methods.

While the ideal study would measure exposure continuously for each subject over the full period of the study, such an approach would be prohibitively intrusive and expensive. Substitutes and compromises should be critically appraised in terms of how well they can take the place of continuous personal monitoring. For particulate matter, direct-reading instruments can provide time trend information about specific constituents of particle mass, physical characteristics of particle mass, particle number concentration, and particle numbers across a range of particle sizes. Direct-reading instruments that can measure aspects of chemical composition should be considered. Particle collection methods collect particles on filters that can be analyzed for a range of chemical constituents. The ideal study should describe the integration of data derived over time by all 3 approaches: personal monitoring and ambient environmental monitoring using both direct-reading instruments and particle collection instruments.

Exposure indicators should be specified (eg, EC, complex organics, and heavy metals), as well as the particle size range considered to have a biological effect. Diesel particulate matter contains EC, partially combusted organics, unburned fuel and oil, sulfate, and ash. Particles in the ultrafine size range (0.05 µm or smaller) are suspected of having health effects, and must be measured. Gas-phase emissions should also be measured, particularly aldehydes, PAHs, chlorinated dioxins, and difurans, since these components may be valuable in determining exposure to diesel exhaust.

The protocol should include provisions for changes in exposure assessment as new equipment becomes available and new concerns about diesel emissions arise. The manner in which new measures of exposure will be integrated into the study as they become available should be specified. As diesel engine technology changes, the ideal study should identify changes in diesel engine use within the cohort and related exposure. Measurements should be taken at times of peak, average, and low exposure, and the investigators should have access to records of diesel engine purchase, maintenance, and repair.

Further, measurement should be made of changes in diesel exposure due to changes in diesel engines, diesel fuels, and exhaust or emission controls. Measures of exposures by dose, dose intensity, duration, and frequency should be made periodically throughout the study. The source of each agent measured and characterization of the chemical and physical characteristics of the main sources of diesel exposure should be specified. The type of personal or environmental monitor used also must be specified.

Outcomes measured should include both incidence of and mortality from lung cancer and any other chronic diseases encompassed by the study, which at a minimum should include other chronic respiratory diseases and cardiovascular diseases. Ideally, the source of incidence data should be physician and hospital records, while the source of mortality data should be death certificates. Because of the long latency period between first exposure to a carcinogen and first occurrence of a solid tumor, and because of the need for enough time to accommodate enough health outcomes for the study, the ideal study should be carried out for a minimum of 25 years.

Measurements of uncertainties and bias will be important, and should include missing data, incomplete data—particularly for exposure data—and the details of dose and duration of each exposure variable. We know that a large sample size will be needed if risks are to be characterized precisely in the presence of some degree of error in classifying exposures. We propose sample size calculation as one of the short-term follow-up activities in this report.

A prospective occupational cohort study that attempts to meet the ideals described here is likely to be costly, given the need to have sufficient participants and sufficiently accurate characterization of exposure and outcome, as well as confounding and modifying factors. However, the large cohort and the long-term follow-up will provide opportunities to study many topics beyond the relationship between diesel exhaust and health, and some support may be available for such secondary studies.

# Abbreviations, Chemical Formulas, and Other Terms

| | |
|---|---|
| BEIR | Biological Effects of Ionizing Radiation [Committee] |
| CMB | chemical mass balance |
| CO | carbon monoxide |
| $CO_2$ | carbon dioxide |
| EC | elemental carbon |
| EPA | Environmental Protection Agency (US) |
| NO | nitric oxide |
| $NO_2$ | nitrogen dioxide |
| $NO_x$ | oxides of nitrogen |
| OC | organic carbon |
| PAHs | polynuclear aromatic hydrocarbons |
| PM | particulate matter |
| $PM_{10}$ | PM with an aerodynamic diameter of 10 µm or less |
| $PM_{2.5}$ | PM with an aerodynamic diameter of 2.5 µm or less |
| QRA | quantitative risk assessment |
| RFA | Request for Applications |
| SOF | soluble organic fraction |
| $SO_2$ | sulfur dioxide |
| $SO_3$ | sulfur trioxide |
| SVOCs | semivolatile organic compounds |
| TEQ | toxicity equivalency quotient |
| TCDD | 2,3,7,8-tetrachlorodibenzo-*p*-dioxin |
| VOCs | volatile organic compounds |

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# Investigators' Report

## Cancer Risk from Diesel Emissions Exposure in Central and Eastern Europe: A Feasibility Study

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# Cancer Risk from Diesel Emissions Exposure in Central and Eastern Europe: A Feasibility Study

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## ABSTRACT

The objectives of this study were to assess the feasibility of enrolling a multicenter historical cohort of workers exposed to diesel emissions in central and eastern Europe (the Czech Republic, Estonia, Hungary, Latvia, Lithuania, Poland, Russia, Slovakia, and Slovenia) and of reconstructing past exposures for the cohort. We developed a company questionnaire aimed at collecting standardized information to determine the number of workers who might be included in the cohort study and their exposures. Our collaborators contacted companies that might participate in the study, provided a contact person with questionnaires to complete, and checked the completed questionnaires. Data from company questionnaires were combined to estimate the size of the expected cohort in terms of exposure to diesel emissions and relevant agents. We determined that it would be possible to enroll 3 multicenter cohorts of workers exposed to diesel exhaust in nonmetal mining, bus transport, and railway transport. A total of 16 companies from 8 countries agreed in principle to participate in the proposed study, yielding an estimated total of 46,500 exposed workers with 295 deaths from lung cancer expected among them. The study would have statistical power to assess a 30% increase in lung cancer risk in each industry-specific cohort and a 15% increase overall. Groups of workers without exposure to diesel exhaust, from the same companies, would be available for internal comparisons. The cohorts would be primarily composed of male workers.

Limited exposure data were collected from 3 companies, 1 in Russia and 2 in Estonia. We measured exposure to diesel particulate matter, expressed as elemental carbon (EC*), and determined the mean levels of exposure for 5 job groups: drivers of suburban goods trains and drivers of shunting locomotives, 15.5 $\mu g/m^3$; bus drivers, 9.3 $\mu g/m^3$; bus mechanics, 37.6 $\mu g/m^3$; and oil-shale miners, 202 $\mu g/m^3$. These measured exposure levels were similar to those of comparable workers in western Europe and North America.

Finally, we devised a theoretical model to describe the exposure process and used the measured exposures to carry out a limited validation of our approach. The retrospective exposure assessments accurately predicted exposure levels (in comparison with the measured levels) in 2 of the 5 job groups examined. Exposure predictions for 3 job groups were outside the measured ranges, probably owing to a lack of detailed information about ventilation rates, work practices, and background levels of diesel particulate matter in the ambient atmosphere.

We conclude that a retrospective cohort study of miners and of workers in bus and railway transport companies is feasible and worthwhile in central and eastern Europe and Russia. These workers are currently exposed to diesel exhaust at levels similar to those experienced by workers in western Europe and North America. Occupational exposure data are available from industrial hygienists but need to be complemented by additional measurements. A model to reconstruct past exposure has been developed and could be used, in combination with the measured exposures, to reconstruct exposure to diesel particulate matter in a retrospective epidemiologic study.

## INTRODUCTION

Exposure to diesel emissions was classified in 1989 as probably carcinogenic to humans by the International Agency for Research on Cancer (IARC, Lyon, France) on the basis of sufficient evidence of carcinogenicity in laboratory animals and limited evidence of carcinogenicity in

This Investigators' Report is one section of Health Effects Institute Special Report *Research Directions to Improve Estimates of Human Exposure and Risk from Diesel Exhaust*, which also includes a report by the HEI Diesel Epidemiology Working Group, four other Investigators' Reports, and an Executive Summary. Correspondence concerning this Investigators' Report should be addressed to Dr P Boffetta, IARC, 150 Cours Albert-Thomas, 69008 Lyon, France.

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* A list of abbreviations and other terms appears at the end of this Investigators' Report.

humans (IARC 1989). Since then, reports of epidemiologic studies have provided information on cancer risk in occupational groups exposed to diesel emissions. A recent meta-analysis identified an overall relative risk of lung cancer of 1.33 (95% confidence interval, 1.21 to 1.46) (Lipsett and Campleman 1999). The available studies involved railroad workers, heavy equipment operators, garage workers, truck and bus drivers, and dockworkers. In a few instances, several occupational groups were classified according to probability or intensity of exposure to diesel emissions on the basis of a job exposure matrix or an expert assessment of occupational histories (Coggon et al 1984; Lerchen et al 1987; Magnani et al 1988; Boffetta et al 1990; Siemiatycki 1991). Several studies have reported an excess risk of cancers other than lung cancer. In particular, an elevated risk of bladder cancer has been reported among truck drivers (Silverman et al 1983, 1986; Jensen et al 1987) and other occupational groups exposed to diesel emissions (Howe et al 1980; Coggon et al 1984; Schoenberg et al 1984; Risch et al 1988).

Despite the relatively large number of epidemiologic investigations on carcinogenicity of diesel emissions (for reviews, see Cohen and Higgins 1995; Boffetta et al 1997; Bhatia et al 1998), interpretation of this evidence is limited by concomitant exposures of the populations to emissions from nondiesel engines, notably gasoline engines. In addition, data in the available studies are not adequate to serve as the basis for a quantitative assessment of the risk of lung cancer. Therefore, further large-scale retrospective epidemiologic studies, coupled with quantitative assessments of past exposures, are needed.

For several reasons workers in central and eastern Europe are valuable populations in which to investigate the long-term health effects of suspected carcinogens. Exposure levels in most occupational settings have been higher than in western countries (Bulbulyan and Boffetta 1999; Fabianova et al 1999). Women have been employed in industries with exposure to carcinogens on a larger scale than in western European countries (Bulbulyan et al 1992), thus providing the opportunity to study differences in occupational cancer risk by gender.

During the time of centrally planned economies, many sectors of the manufacturing and service industries in central and eastern Europe were organized in relatively large companies. Typically owned by public institutions, these companies had a low workforce turnover. They have since been privatized and reduced their workforces, but records of previous employees are accessible. Exposure to possibly noxious agents has in many cases been documented; however, the records are not all computerized and thus might not be directly accessible.

Furthermore, a network of industrial hygienists existed in all these countries. These experts were in charge of monitoring and controlling hazardous exposures (although with limited practical efficacy) either in a given geographical region or, more often, within a given industry. These experts currently represent a valuable resource for occupational epidemiologic studies.

Follow-up for mortality in cohorts from these countries is feasible with a success rate comparable to that obtained in Western countries (for examples, see Table 1). Reference mortality rates for most neoplasms are available for the central and eastern European countries. In addition, high-quality cancer registries have operated for several decades in some regions (Table 2), which allows additional follow-up for cancer incidence.

This report considers 3 different aspects of this work in turn: the epidemiologic study, collection of occupational exposure data, and retrospective exposure assessment.

## SPECIFIC AIMS

The overall objectives of the project were (1) to investigate the feasibility of a multicenter historical cohort study of workers exposed to diesel emissions in central and eastern Europe, and (2) to assess whether we could reconstruct diesel exhaust exposures for the cohort members.

**Table 1.** Examples of Central and Eastern European Cohort Studies with Completeness of Follow-Up

| Country | Occupational Exposure | Number of Workers | Follow-Up Period | Completeness of Follow-Up (%) | Reference |
|---|---|---|---|---|---|
| Poland | Asbestos | 670 | 1945–1985 | 99.6 | Szeszenia-Dabrowska et al 1988 |
| Czech Republic | Uranium | 4,320 | 1975–1990 | 99.0 | Tomasek et al 1994 |
| Russia | Fertilizer | 4,996 | 1965–1990 | 93.4 | Bulbulyan et al 1996 |

**Table 2.** Regions of Collaborating Countries Covered by Population-Based Cancer Registries Until 1992

| Country | Region | Good-Quality Data Available |
|---|---|---|
| Czech Republic | Whole country | 1983–1992 |
| Estonia | Whole country | 1983–1992 |
| Hungary | Szabolcs-Szatmar and Vas counties | 1962–1987 |
| Latvia | Whole country | 1983–1992 |
| Poland | Various regions | 1965–1992[a] |
| Romania | Cluj county | 1974–1987 |
| Russia | St Petersburg | 1983–1987 |
| Slovakia | Whole country | 1978–1992 |
| Slovenia | Whole country | 1956–1992 |

[a] Data are available for different periods in different regions; only Krakow and Warsaw data are available for the whole period 1965–1992.

## METHODS

### FEASIBILITY OF EPIDEMIOLOGIC STUDY

#### Development of Company Questionnaire

A company questionnaire was developed with the aim of collecting standardized information on the size and exposure of the groups of workers that might be included in the cohort study (Appendix A). Special emphasis was put on the availability of information on exposure to agents other than diesel emissions. The questionnaire was prepared at IARC, translated into the national language in each collaborating center, and back-translated into English at IARC to ensure the integrity of the original translation.

#### Contact with Possible Participating Companies

The feasibility study was conducted in the Czech Republic, Estonia, Hungary, Latvia, Lithuania, Poland, Russia, Slovakia, and Slovenia. Study collaborators are listed in the Acknowledgments section. A similar study, not reported here, was conducted in Romania and Israel. The feasibility study focused on 2 broadly defined groups of workers with potentially high exposure to diesel emissions: drivers (ISCO 985), mechanics (ISCO 843), and railway workers (ISIC 71) in land transport; and miners (ISCO 71) in nonmetal mining. (The codes in parentheses are from the International Standard Classification of Occupations [ISCO; International Labour Office 1968] and the International Standard Industrial Classification [ISIC; United Nations 1971])

Each collaborating center identified and contacted companies in its study area that could include a substantial number of workers in the 2 categories defined above. The initial contact aimed at identifying companies available to participate in the study, the approximate size of an exposed cohort and of another cohort not exposed to diesel emissions, the expected completeness of available employment records, and the available information about changes in sources of diesel emissions and the environment where the emissions occurred.

#### Completion of Questionnaires

The collaborators visited each company and presented the questionnaire to the contact persons. The contact persons, who typically were production managers, technical engineers, or industrial hygienists, were in charge of completing the questionnaire. They also provided additional information such as copies of sample pages of employment rosters and exposure records. The completed questionnaires were returned to the collaborators, who checked the completeness and quality of the information collected. Typically, the collaborators visited the company on several occasions to complete or clarify some aspects of the information collected. The questionnaires were then translated into English, and copies were transferred to IARC and to the Institute of Occupational Medicine (IOM, Edinburgh, UK).

During the feasibility study and after completion of the company questionnaires, IARC epidemiologists visited each collaborating center; they also visited some of the companies and discussed details about all companies with the contact persons. In order to carry out personal exposure monitoring for selected occupations, chosen to represent a diverse range of exposure scenarios, IOM industrial hygienists visited four centers. In addition, the hygienists assessed current work practices and conditions and verified or clarified the scope and quality of preexisting exposure-related information that would be required for the reconstruction of exposure histories.

#### Estimate of Statistical Power of Cohort Study

We derived from company questionnaires the approximate distribution by gender, age, and calendar period of first employment of workers in the companies retained for a full-scale study. We estimated the person-years of observation specific to gender, age, and calendar year, and applied the relevant incidence of lung cancer (Parkin et al 1997) in order to estimate the number of cases of lung cancer expected in each cohort.

Approximately 80 expected cases of lung cancer among exposed subjects would be necessary to obtain an 80%

power to detect as statistically significant (at α level of 0.05) a 30% increase in mortality based on external comparison (Breslow and Day 1987).

## OCCUPATIONAL EXPOSURE DATA

### Availability of Historical Exposure Data

The availability and quality of historical exposure data was assessed from information gathered through company questionnaires and site visits by IARC epidemiologists and IOM industrial hygienists. Copies of raw results of measurements were obtained whenever available. Information about historical conditions and work practices was also obtained from the company representatives, who had many years of experience in the industry.

### Measurement of Current Exposures

At the time of the survey, information about current work practices and conditions was obtained by observation and by discussion with company representatives.

Measurements of personal exposure to respirable dust and diesel exhaust were made using IOM respirable dust samplers fitted with foam inserts size-selective for particulate matter 10 µm or smaller in aerodynamic diameter ($PM_{10}$). The IOM sampler cassettes were loaded with 25-mm quartz fiber filters, and the cassettes were weighed before the size-selective foam inserts were added. Each sampler was positioned in the breathing zone by clipping it to the worker's lapel. The sampler was connected to a battery-operated pump by means of polyvinyl chloride tubing, and the sampling flow rate was set to 2.0 L/min. This flow rate was checked at the beginning and end of sampling and, when possible, at regular periods between these times. The sampler was worn for a full work shift to ensure that the measured concentrations represented daily average exposures.

We anticipated that some workers might consider personal sampling an unacceptable intrusion, in which cases the sampler could be placed at a fixed location within the normal working environment. This was not necessary, however, and we were able to monitor all tasks in the conventional manner.

In railway transport, it was not possible to directly observe the work practices of the train drivers owing to the distances involved and limited space available on board. Sampling was restricted to short-haul locomotive drivers—that is, drivers of shunting locomotives and those operating suburban goods trains in a circular route around the city.

At the end of sampling, the filter cassette was removed from the sampling head and the size-selective foam insert was removed and discarded. The cassettes were then analyzed gravimetrically to determine the respirable dust concentrations.

Each filter sample was analyzed for organic carbon (OC) and EC using evolved gas analysis with a thermal optical sensor in accordance with National Institute for Occupational Safety and Health (NIOSH) method 5040 (NIOSH 1996). This work was carried out by David Dabill of the Health and Safety Laboratory, Sheffield, England. After analysis of the filter samples, the respirable dust concentrations and EC concentrations were calculated. The geometric mean (GM) and geometric standard deviation (GSD) of concentrations were calculated for each set of measurements from similarly exposed groups.

Airborne concentrations of EC are taken to be a reliable marker of diesel exhaust concentrations, and Verma and colleagues (1999) have shown this to be a suitable method for monitoring diesel exhaust exposures in the railroad industry. The American Conference of Governmental Industrial Hygienists (ACGIH) has now proposed a threshold limit value of 0.05 $mg/m^3$ for diesel exhaust (ACGIH 2000), and until further clarification, this value is assumed to apply to the EC content of submicron dust particles.

Use of the respirable dust samplers was not practical in the oil-shale mine, so cyclone-type respirable dust samplers (SIMPEDS samplers, Casella, London, UK) were used instead. These samplers were prepared for gravimetric determination, but losses from the filter media due to the brittle nature of the quartz fiber filters prevented it. The samples were analyzed for EC and OC, however, as described previously in this report.

Ideally, we would have preferred to use a sampler incorporating a preselector with a submicron-sized cutoff for the oil-shale mine, but this type of equipment was not available at the time of survey. The measurements were therefore intended to be indicative only.

# RESULTS

## FEASIBILITY OF EPIDEMIOLGIC STUDY

The company questionnaire was translated into the national languages. Selected sections were back-translated into English at IARC, and the back-translations were compared with the original English version. No important discrepancies were identified.

A total of 16 companies were retained for analysis at the end of the feasibility study (Table 3): 7 companies were involved in nonmetal mining, 2 in railway transport, and 7

in bus transport. Slovenia was the only country in which no suitable companies were identified.

Table 3 presents also the estimated total number of workers and the number of workers exposed to diesel emissions who could be included in a historical cohort study in each company. Most workers exposed to diesel emissions were men. The total estimated size of the cohort exposed to diesel emissions was 46,500 male workers, of whom 55% were miners.

The period of available follow-up and the year of first use of diesel engines were determined by country and type of industry (Table 4). We found that all groups of workers exposed to diesel exhaust could be followed for at least 25 years.

The expected numbers of person-years and cases of lung cancer among male workers exposed to diesel emissions were determined (Table 5). The combined cohort of exposed workers would contribute more than 800,000 person-years of observation, yielding an estimated 295 cases of lung cancer. The cohorts of miners and bus workers would meet the target statistical power, while the cohort of railway workers would have a power of 70% to detect a 30% increased risk.

**Table 3.** Companies Retained in Feasibility Study, Total Number of Male Workers, and Number of Workers Exposed to Diesel Emissions

| | Nonmetal Mining[a] | | | Railway Transport | | | Bus Transport | | |
|---|---|---|---|---|---|---|---|---|---|
| Country | Company | All Workers | Exposed Workers | Company | All Workers | Exposed Workers | Company | All Workers | Exposed Workers[b] |
| Czech Republic | 1C | 5,000 | 2,000 | 0 | | | 0 | | |
| Estonia | 1O | 14,000 | 3,000 | 0 | | | 1 | 18,000 | 6,000 |
| Hungary | 1B | 20,000 | 12,000 | 0 | | | 0 | | |
| Latvia | 0 | | | 1 | 5,000 | 1,500 | 3 | 20,000 | 3,000 |
| Lithuania | 0 | | | 0 | | | 1 | 14,000 | 2,000 |
| Poland | 0 | | | 0 | | | 2 | 8,000 | 2,500 |
| Russia | 0 | | | 1 | 9,000 | 6,000 | 0 | | |
| Slovakia | 3C,1M | 71,000 | 8,500 | 0 | | | 0 | | |
| Slovenia | 0 | | | 0 | | | 0 | | |
| Total | 7 | 110,000 | 25,500 | 2 | 14,000 | 7,500 | 7 | 60,000 | 13,500 |

[a] C indicates coal; O, oil shale; M, magnesite; B, bauxite.

[b] Excluding drivers.

**Table 4.** Year of First Use of Diesel Engines and Year of Beginning of Possible Follow-Up by Industry and Country[a]

| | Nonmetal Mining | | Railway Transport | | Bus Transport | |
|---|---|---|---|---|---|---|
| Country | Diesel | Follow-Up | Diesel | Follow-Up | Diesel | Follow-Up |
| Czech Republic | 1960 | 1975 | — | — | — | — |
| Estonia | 1972 | 1972 | — | — | 1945 | 1956 |
| Hungary | 1974 | 1957 | — | — | — | — |
| Latvia | — | — | 1967 | 1950 | 1960 | 1946 |
| Lithuania | — | — | — | — | 1947 | 1947 |
| Poland | 1970 | 1973 | — | — | 1960 | 1960 |
| Russia | — | — | 1950 | 1950 | — | — |
| Slovakia | 1960 | 1962 | — | — | — | — |
| Slovenia | — | — | — | — | — | — |

[a] If there were several companies in the same industry and country, then the earliest years are given here.

**Table 5.** Estimated Person-Years and Cases of Lung Cancer Among Male Workers Exposed to Diesel Emissions by Industry and Country

| | Nonmetal Mining | | Railway Transport | | Bus Transport | |
|---|---|---|---|---|---|---|
| Country | Person-Years | Lung Cancer[a] | Person-Years | Lung Cancer[a] | Person-Years | Lung Cancer[a] |
| Czech Republic | 24,000 | 10 | — | — | — | — |
| Estonia | 36,000 | 12 | — | — | 120,000 | 40 |
| Hungary | 120,000 | 55 | — | — | — | — |
| Latvia | — | — | 37,000 | 12 | 75,000 | 24 |
| Lithuania | — | — | — | — | 50,000 | 17 |
| Poland | — | — | — | — | 50,000 | 16 |
| Russia | — | — | 144,000 | 49 | — | — |
| Slovakia | 153,000 | 60 | — | — | — | — |
| Slovenia | — | — | — | — | — | — |
| Total | 333,000 | 137 | 181,000 | 61 | 295,000 | 97 |

[a] Cases of lung cancer.

The 7 mines included in the study were involved in mining coal (1 in the Czech Republic and 3 in Slovakia), oil shale (1 in Estonia), bauxite (1 in Poland), and magnesite (1 in Slovakia). Exposure to silica, radon, arsenic, or other lung carcinogens was minimal or absent in all these mines.

A number of companies in addition to those listed in Table 3 were contacted during the feasibility study, but they were excluded for different reasons. In particular, a Polish copper mine and a Slovakian iron mine were excluded because of possible coexposure to radon or arsenic. Two small truck transport companies in Latvia were excluded because they would have been the only companies from this industry. One railway transport company and 2 bus transport companies in Lithuania, as well as a bus transport company in Slovakia, qualified for the feasibility study, but the size of the cohorts and the number of expected lung cancer cases were small (fewer than 5 in each company).

Drivers constituted a large proportion of the workers in bus transport companies, but we considered only garage mechanics as definitely exposed to diesel exhaust. Drivers were therefore excluded from calculation of the expected size of the exposed cohort in bus transport companies. Information on past smoking habits was available only for workers in the cohorts from the Czech Republic and Poland.

Tables 3 and 5 report the results for male workers only. Although the companies included in the feasibility study employed many women, they were mainly in jobs not exposed to diesel exhaust. The only job with potential exposure to diesel exhaust in which a sizable number of women were employed was bus driver. Among bus mechanics, railway conductors and mechanics, and underground miners (the jobs with the highest potential for exposure in our study), the number of women was very small.

In parallel to the field work in the 9 countries included in the feasibility study, contacts have been established with a salt mine in Romania (600 workers, of whom 200 currently are exposed to diesel exhaust) and a railway company in Israel (2,000 workers, of whom 500 currently are exposed to diesel exhaust). Although we have not reviewed the employment and exposure records in these companies with the same detail as for the other companies, these data might become available for a full-scale epidemiologic study.

## AVAILABILITY OF HISTORICAL EXPOSURE DATA

The availability and quality of historical exposure data were assessed for all 16 companies that agreed in principle to participate in a diesel emissions exposure study. The results are presented here by industry.

### Nonmetal Mining

***Czech Republic: Coal Mining*** A great deal of exposure data for the Czech coal mining company was included in the feasibility study. Benzene, polycyclic aromatic hydrocarbons, and airborne dust were regularly monitored. The benzene concentration can be considered to be a specific marker of diesel exhaust exposure because there were no other sources of exposure, and these measurements may be used to understand the historical pattern of exposures. Measurements of EC concentrations from at least one of the coal mines will be necessary in order to provide a baseline measurement of current exposure.

***Estonia: Oil-Shale Mining*** The available exposure data for the Estonian oil-shale mining company included measurements of carbon monoxide and nitrogen dioxide made in 1986 and repeated in 1998. A limited number of benzene measurements were also available. Measurements of respirable dust, being obtained by regular static monitoring at the mine entrance since 1970, were also available. Although the measurements were quoted as respirable dust, the sampling method was for total respirable dust. The local custom is to assume, on the basis of microscopic evaluation of the particle sizes, that all of the airborne dust is respirable.

The diesel-powered front loaders were mainly of Ukraine origin although some vehicles from Finland had been introduced. These vehicles were well maintained, and their diesel exhaust emissions were not considered to be excessive. The mine had strict rules for monitoring ventilation rates, and a preliminary inspection at the time of our visit indicated that the mine was efficiently ventilated. The ventilation conditions appeared to have been relatively stable since 1970.

Work practices in the mine were such that diesel-exposed workers (loader drivers and support operators) were segregated by time from nonexposed groups such as drillers and shot-firers. It may therefore be possible to include the latter occupations in a control cohort.

Before 1995 the number of mine workers exposed to diesel exhaust was greater because approximately 12 manual workers were usually deployed in the vicinity of the diesel-powered loaders.

***Hungary: Bauxite Mining*** Regular (monthly) measurements were available only for carbon monoxide and nitrogen oxides in the Hungarian bauxite mine; sporadic measurements of total dust were also available. Although the carbon monoxide results might provide some information on exposure to diesel exhaust, an accurate assessment of past and current exposure would require further monitoring.

***Slovakia: Coal Mining*** In the 3 Slovakian coal mines, the available data for carbon monoxide and oxides of nitrogen were considerable. These measurements were obtained by area sampling (rather than by personal monitoring). These measures probably will not be useful in assessing exposure levels because only the concentrations within a certain range of a specified limit value were usually recorded. Therefore, the measured carbon monoxide levels will provide a positive bias in any exposure assessment. Further monitoring for EC concentrations would be required to establish current exposure levels.

***Slovakia: Magnesite Mining*** The sample analyses available from the Slovakian magnesite mining company were limited to 8 polycyclic aromatic hydrocarbon samples collected in 1999, inside cabins of diesel-powered loaders used within the mine workings. Because of their limited nature, it is not clear whether these can be compared with any historical measurements. An accurate assessment of exposure for these workers will require additional monitoring.

**Bus Transport**

***Estonia*** The Estonian bus company had very little occupational exposure data available for bus mechanics and none for bus drivers. None of the data were relevant in estimating past exposures to diesel exhaust particulate matter.

In 1991 the bus company introduced used buses imported from Sweden (Scania, Volvo). Prior to 1991 only Hungarian buses (Icarus) were used. Although the Hungarian buses were quite old at the time of this study, they were still highly regarded and had been considered to be reliable vehicles when new. It may be concluded that exposures have been quite stable for the last 30 years or so with only modest changes recently.

In any case, the exposures of bus drivers appeared to be quite low and mainly influenced by the numbers of diesel-powered trucks and buses on the road rather than by the emissions from the bus that they were driving. There are significant differences in environmental pollution in Estonia due to burning of shale oil in power stations, and the relative health effects may need to be evaluated.

Special measures have been in place since 1993 to control exposure to asbestos during work on brake parts.

***Latvia*** The working conditions and usage of diesel engines in the 3 Latvian bus companies appeared to follow the same pattern as in the Estonian company. The majority of buses were of Hungarian production with some Western vehicles being introduced. Since the early 1980s, the maintenance program has included active tuning of the engines to reduce exhaust emissions. If necessary, the diesel fuel can be cleaned using a centrifugal filtering device. This means that low-grade diesel fuel can be improved on site, preventing high emission levels.

Again, buses were stored outdoors, thereby eliminating high concentrations of airborne diesel exhaust within the garages when the vehicles are started up in the morning. Smoking is prohibited for both drivers and passengers on buses in Latvia and has always been so.

The bus depots included separate departments that covered general maintenance, body repairs, hydraulics, engineering, and so forth. Consequently, not all of the workers

were exposed to diesel exhaust. However, the employment records are sufficiently detailed to identify those workers who were exposed. The exposure data available for the bus companies were limited. Annual checks were carried out on airborne dust, welding exhaust, etc. These data will have no direct relevance in estimating exposure levels for EC but may be useful in further analyzing how conditions have changed over time. There are currently no specific measures for controlling asbestos exposure during maintenance of brakes, and this would have to be evaluated in terms of the potential to cause lung cancer.

***Lithuania*** Very few measurements of carbon monoxide had been made at the Lithuanian bus company we studied, and these related only to the year 1998. No other exposure measurements relevant to diesel particulate matter exposure were available. Any future evaluation of exposure in this company would require additional monitoring.

***Poland*** Detailed exposure data, including regular monitoring of benzene, benzo[*a*]pyrene, carbon monoxide, and nitrogen oxides, were available since 1987 for 1 of the 2 Polish bus companies. In the other company, regular monitoring has taken place only for benzene and total dust. These measurements will be useful to assess past exposure patterns. It will be necessary to measure EC concentrations to assess current exposure to diesel exhaust.

**Railway Transport**

***Latvia*** The Latvian railway depot we studied included a large repair and maintenance workshop. Locomotives (for shunting) were of Czech origin, while the passenger trains were of Latvian manufacture. The locomotives were highly regarded and the company's standard of maintenance was high. By observation, there was no evidence of black smoke emissions from the exhausts, and even though shunting locomotives would maneuver rolling stock into the maintenance buildings, there was no noticeable exhaust in the indoor atmosphere. Smoking is allowed for train drivers.

The conditions also appeared to have remained stable for at least 30 years, so exposures would seem to have been generally low. Because the working conditions and work practices adopted by the Latvian railway drivers were very similar to those of the Russian cohort, the exposures to diesel exhaust particles can reasonably be assumed to be similar in magnitude.

***Russia*** Comprehensive information about work practices, historical uses of diesel engines, and exposure data in the Russian railway industry is available from the company. These data include 18 years' worth of "soot" measurements from 1976 to 1994 obtained by static sampling in locomotive cabins. The sampling and analytic methods used to obtain these data are unclear, and the measured values seem too low to be simple measurements of total respirable dust. However, the data will be useful in assessing how conditions have changed over time if a year-by-year analysis can be provided.

Suvorov and Shterengarz (1986) found the soot in diesel emissions in Russian railways, at percentage of engine power, to be 83 ± 38, 79 ± 35, 90 ± 36, and 51 ± 14 mg/m$^3$, respectively, for an engine that was idle, at 25% power, at 50% power, and at maximum power. On the basis of 200 to 300 measurements obtained over a 10-year period (1970 to 1980), they found the mean level of soot in the locomotive cabin of trains to be 0.109 (range, 0.00 to 0.169) mg/m$^3$ and the mean level in cabins of shunting locomotives to be 0.133 (range, 0.00 to 0.363) mg/m$^3$. Anecdotal information suggested that operational conditions have been stable from 1970 to present, but little information was available about conditions and work practices prior to this time.

A great many related measurements were available covering the period 1976 to 1994. These included measurements of benzene, hydrocarbons, polycyclic aromatic hydrocarbons, carbon monoxide, oxides of nitrogen, and oxides of sulfur. However, these measurements will be only marginally relevant in estimating exposures in terms of EC concentrations.

There were no indications that drivers or mechanics would be exposed to other agents that may cause lung cancer. Mechanics were exposed to chrysotile asbestos during some maintenance activities, but in attempts to measure the asbestos exposure levels, fiber concentrations were below the limit of detection (R Shterengarz, personal communication, 2000).

## MEASUREMENT OF CURRENT EXPOSURES

Current diesel exhaust exposure levels were measured for train drivers in Russia and for bus mechanics, bus drivers, and oil-shale mine workers in Estonia. Approximately 50 samples were collected in total from 3 companies. The sampling was carried out in close cooperation with local collaborators and host companies, and the occupational groups were selected after consultation with all parties. The sampling program provided a consistent set of diesel exhaust exposure data of similarly exposed occupational groups. The data were used in the exposure reconstruction model, which might be applied to a full-scale cohort study. Tables 6 through 10 report the results of measurements in each industry, and Table 11 summarizes the results across industries. We have compared our results with those of other studies based on EC determination (Table 12).

**Railway Workers**

Personal air monitoring of respirable dust ($PM_{10}$), OC, and EC was carried out for drivers of short-haul suburban goods trains (Table 6) and for drivers of locomotives carrying out shunting maneuvers (Table 7) in Russia. The local industrial hygienist reported that shunter drivers may have higher exposures than main line train drivers. This higher exposure was anticipated in part because drivers must lean out of the cabin window to see the rail track during coupling and uncoupling operations and when maneuvering rolling stock into sidings or sheds. We therefore distinguished between these shunter drivers and the main line drivers operating short-haul suburban goods trains in a continuous circular route around the city.

**Table 6.** Diesel Exhaust Exposures of Short-Haul Drivers and Assistant Drivers of Suburban Goods Trains in Russia

| Sample Code | Locomotive Driver/Assistant[a] | Time On | Time Off | Sample Duration (min) | Volume (L) | $PM_{10}$ (mg/m$^3$) | Organic Carbon (μg/m$^3$) | Elemental Carbon (μg/m$^3$) |
|---|---|---|---|---|---|---|---|---|
| OM01 | Driver* | 0748 | 2055 | 787 | 1,495 | 0.85 | 111.2 | 16.9 |
| OM04 | Assistant* | 0757 | 2054 | 777 | 1,476 | 0.73 | 78.5 | 10.6 |
| OM06 | Driver | 0822 | 2108 | 766 | 1,379 | 0.82 | 65.5 | 16.9 |
| OM11 | Driver† | 0917 | 2131 | 734 | 1,358 | 1.60 | 348.4 | 69.0 |
| OM09 | Assistant† | 0915 | 2112 | 717 | 1,291 | 1.45 | 351.4 | 71.8 |
| OM12 | Driver | 0731 | 2015 | 764 | 1,452 | 0.79 | 68.1 | 17.8 |
| OM13 | Driver | 0732 | 2015 | 763 | 1,450 | 0.81 | 63.9 | 12.2 |
| OM17 | Driver‡ | 0802 | 2112 | 790 | 1,541 | 0.34 | 57.1 | 10.7 |
| OM19 | Assistant‡ | 0802 | 2116 | 794 | 1,588 | 0.51 | 53.6 | 13.5 |
| OM21 | Driver§ | 0833 | 2134 | 781 | 1,562 | NA[b] | 75.2 | 13.8 |
| OM22 | Assistant§ | 0833 | 2135 | 782 | 1,564 | NA[b] | 81.3 | 12.0 |
| OM23 | Driver | 0900 | — | — | — | Rejected | Rejected | Rejected |
| Geometric mean | | | | | | 0.8 | 95.0 | 18.3 |
| Geometric standard deviation | | | | | | 1.6 | 2.0 | 2.0 |

[a] The symbols *, †, ‡ and § indicate paired driver and assistant driver teams sharing same locomotive (type 2M62).

[b] NA indicates sample was not analyzed gravimetrically owing to losses from filter media.

**Table 7.** Diesel Exhaust Exposures of Shunting Locomotive Drivers and Assistant Drivers in Russia

| Sample Code | Locomotive Driver/Assistant[a] | Time On | Time Off | Sample Duration (min) | Volume (L) | $PM_{10}$ (mg/m$^3$) | Organic Carbon (μg/m$^3$) | Elemental Carbon (μg/m$^3$) |
|---|---|---|---|---|---|---|---|---|
| OM02 | Shunter driver | 0752 | 1820 | 628 | 1,225 | 0.85 | 128.8 | 12.3 |
| OM03 | Shunter driver | 0754 | 2037 | 763 | 1,488 | 1.54 | 255.5 | 30.8 |
| OM05 | Shunter driver | 0816 | 2023 | 727 | 1,381 | 0.82 | 39.0 | 5.3 |
| OM08 | Shunter driver* | 0850 | 2042 | 712 | 1,353 | 0.93 | 90.4 | 8.9 |
| OM07 | Assistant driver* | 0840 | 2045 | 725 | 1,378 | 0.62 | 99.8 | 19.7 |
| OM14 | Shunter driver† | 0735 | 1835 | 660 | 1,320 | 0.72 | 81.1 | 6.4 |
| OM15 | Assistant driver† | 0736 | 1835 | 659 | 1,318 | 0.51 | 90.5 | 10.1 |
| OM16 | Shunter driver | 0740 | 2034 | 774 | 1,548 | 0.71 | 174.5 | 22.9 |
| Geometric mean | | | | | | 0.8 | 105.1 | 12.3 |
| Geometric standard deviation | | | | | | 1.4 | 1.8 | 1.9 |

[a] The symbols * and † indicate paired driver and assistant driver teams sharing same locomotive (type T3M2).

The measured range of $PM_{10}$ concentrations for the train drivers was 0.34 to 1.60 mg/m$^3$ with a GM of 0.8 mg/m$^3$; the EC concentrations ranged from 10.6 to 71.8 µg/m$^3$ with a GM of 18.3 µg/m$^3$ (Table 6). Two of the train drivers had much higher exposures than the rest of the group (samples OM09 and OM11). The ratios of OC to EC were similar for all samples, so it is unlikely that these higher exposures were due to contamination. Since these high-exposure samples were obtained from 2 drivers working together, their work presumably differed in some way from that of the others.

The shunter drivers had exposures similar to those of the local train drivers. Their $PM_{10}$ concentrations ranged from 0.51 to 1.54 mg/m$^3$ with a GM of 0.80 mg/m$^3$. EC concentrations ranged from 5.3 to 30.8 µg/m$^3$ with a GM of 12.3 µg/m$^3$ (Table 7). Several of the sample EC concentrations for the shunter drivers were very low at 5.3, 6.4, and 8.9 µg/m$^3$. The nature of the work that these drivers were performing is unclear, but the exposures indicate general background levels for an urban environment (Zaebst et al 1991).

The quoted concentrations were obtained over 12-hour work shifts and as such are a measure of concentrations across the full work shift. The measured concentrations compare well with the EC concentrations quoted by Verma and colleagues (1999) for Canadian railroad drivers (see Table 12). This suggests that diesel exhaust exposures of railway workers in Russia currently are the same as exposures in most Western countries.

**Bus Mechanics and Drivers**

Personal air monitoring of respirable dust ($PM_{10}$), OC, and EC was carried out for bus mechanics involved with maintenance and repair work (Table 8) and for bus drivers (Table 9) in a major urban bus company in Estonia. The vehicle mechanics were expected to have higher diesel exhaust exposures than the bus drivers. Although local exhaust ventilation systems were provided for static testing of engines, vehicles were regularly being moved around, resulting in a gradual buildup of diesel exhaust inside the workshops. The garages were used only for repair and maintenance. Buses were stored outdoors, a practice that avoids the production of high indoor concentrations of exhaust when buses are started up before being used.

In these garages the mechanics were a separate group from welders and therefore not exposed to welding fumes or other airborne contaminants that would interfere with the exposure assessment. The measured range of $PM_{10}$ concentrations for the mechanics was 0.45 to 2.21 mg/m$^3$ with a GM of 1.02 mg/m$^3$; the EC concentrations were in the range of 24.8 to 52.1 µg/m$^3$ with a GM of 37.6 µg/m$^3$ (Table 8). The bus drivers had lower exposures than the

**Table 8.** Diesel Exhaust Exposures of Bus Mechanics in Estonia

| Sample Code | Location of Sample[a] | Time On | Time Off | Sample Duration (min) | Volume (L) | $PM_{10}$ (mg/m$^3$) | Organic Carbon (µg/m$^3$) | Elemental Carbon (µg/m$^3$) |
|---|---|---|---|---|---|---|---|---|
| OM24 | TH-2 Liin | 0820 | 1601 | 461 | 922 | 1.59 | 109.9 | 33.3 |
| OM25 | TH-2 Liin | 0820 | 1558 | 458 | 916 | 1.25 | 86.4 | 25.9 |
| OM26 | TH-2 Liin | 0820 | 1555 | 455 | 910 | 2.21 | 140.1 | 29.3 |
| OM27 | TH-2 Liin | 0820 | 1552 | 452 | 904 | 1.34 | 132.2 | 43.3 |
| OM28 | TH-2 Liin | 0820 | 1552 | 452 | 904 | 0.67 | 164.7 | 51.0 |
| OM29 | JR-Pomivahetus | 0824 | 1617 | 473 | 946 | 0.73 | 129.9 | 37.1 |
| OM31 | JR-Pomivahetus | 0824 | 1559 | 455 | 910 | 0.56 | 121.2 | 43.0 |
| OM32 | JR-Pomivahetus | 0824 | 1605 | 461 | 922 | 0.45 | 93.4 | 24.8 |
| OM33 | JR-Pomivahetus | 0824 | 1623 | 479 | 958 | 1.81 | 168.4 | 39.6 |
| OM34 | JR-Pomivahetus | 0824 | 1623 | 479 | 958 | 0.97 | 181.7 | 41.0 |
| OM35 | TH-2 Konveier | 0832 | 1600 | 448 | 896 | 1.22 | 271.5 | 52.1 |
| OM36 | TH-2 Konveier | 0832 | 1600 | 448 | 896 | 1.72 | 130.2 | 32.5 |
| OM37 | TH-2 Konveier | 0832 | 1600 | 448 | 896 | 0.57 | 178.7 | 38.7 |
| OM38 | TH-2 Konveier | 0832 | 1600 | 448 | 896 | 1.15 | 228.2 | 50.6 |
| OM39 | TH-2 Konveier | 0832 | 1550 | 438 | 880 | 0.71 | 108.3 | 35.1 |
| Geometric mean | | | | | | 1.02 | 142.5 | 37.6 |
| Geometric standard deviation | | | | | | 1.6 | 1.4 | 1.3 |

[a] TH-2 Liin is the department carrying out maintenance of Icarus buses; JR-Pomivahetus, maintenance of non-Icarus buses; TH-2 Konveier, conveyor line maintenance (mixture of Icarus and other buses).

mechanics. Their $PM_{10}$ dust concentrations ranged from 0.40 to 1.01 mg/m$^3$ with a GM of 0.58 mg/m$^3$, and their EC concentrations were 5.7 to 10.5 µg/m$^3$ with a GM of 9.3 µg/m$^3$ (Table 9).

All exposure measurements were obtained over 8-hour work shifts and as such are a measure of concentrations across the full work shift.

A comparison of these exposures with those of truck drivers and mechanics in the United States as quoted by Zaebst et al (1991) indicated that the exposures for Estonian bus drivers were similar to those of US truck drivers (see Table 12). This suggests that the main source of diesel exhaust exposure was the general urban environment.

The mean EC concentration of 37.6 µg/m$^3$ measured for bus mechanics in Estonia is higher than the mean value of 12.1 µg/m$^3$ quoted for US truck mechanics (Zaebst et al 1991). This is most likely due to the differences in working environments between truck and bus repair depots rather than differences between Estonia and US work practices.

### Oil-Shale Miners

We visited a large oil-shale mine in Estonia intending to carry out personal sampling, but production had temporarily ceased owing to technical problems. We toured the production areas, however, and the mine safety engineer explained the work practices and production methods. The Estonian collaborators carried out the sampling at a later date under normal production conditions.

Oil shale is deposited in bands separated by layers of limestone. The limestone contains some crystalline silica (quartz). Owing to the nature of the work, current dust and silica exposures appear to be rather high.

The mining method in this case was the pillar-and-room technique. The oil-shale seam is drilled and blasted, and the resultant material is transferred to a conveyor by means of diesel-powered front-end loaders.

The diesel-exposed workers (loader drivers and support operators) were segregated by time from nonexposed groups such as drillers and shot-firers (workers preparing the explosive charges). Therefore, the loader drivers were not exposed to airborne dust from any other operation; however, this in itself appears to be a significant source of dust exposure. The mine utilized water sprays on conveyor transfer points, but it was not clear whether any dust suppression was applied to the loose material as it was loaded by the diesel-powered vehicles. The mine workers were supplied with disposable face-piece respirators that filter dust, but anecdotal evidence suggested that they only wore these respirators for the very dustiest of tasks.

The technical problems with sampling for EC as an indicator for diesel exhaust in coal mines are well known. Diesel exhaust particles are largely submicron in size, and larger carbon-based compounds (such as those present in coal dust) interfere with the analysis. Usually, an impactor or a similar preseparator with a submicron cutoff is necessary to separate out the larger particles. This option was not practical at the time of survey, so cyclone-type respirable dust samplers were used to obtain an estimate of exposure levels. Errors are assumed to be inherent in this approach.

Estimates of $PM_{10}$ were possible from only 2 of the 5 samples; other filters were rejected because of losses from the filter media (Table 10). For the 2 samples for which estimates were possible, the concentrations were 4.07 and 2.73 mg/m$^3$. These measures are likely to underestimate the true concentrations, however, because we suspect losses from these samples as well.

Analyses for EC were possible, however, and the measured concentrations were in the range of 96.5 to 377.2 µg/m$^3$ with a GM of 202.4 µg/m$^3$ (Table 10). This compares well with previous measurements of EC in zinc and potash mines carried out by others (Haney and Fields 1996; Dahmann and Bauer 1997; Stanevich et al 1997).

**Table 9.** Diesel Exhaust Exposures of Bus Drivers in Estonia

| Sample Code | Vehicle Type | Time On | Time Off | Sample Duration (min) | Volume (L) | $PM_{10}$ (mg/m$^3$) | Organic Carbon (µg/m$^3$) | Elemental Carbon (µg/m$^3$) |
|---|---|---|---|---|---|---|---|---|
| OM41 | Scania 2729 | 1030 | 1758 | 448 | 874 | 0.50 | 91.2 | 10.4 |
| OM42 | Scania 2536 | 1325 | 2030 | 425 | 829 | 1.01 | 65.5 | 10.5 |
| OM43 | Volvo 2223 | 1332 | 1958 | 386 | 772 | 0.40 | 68.5 | 10.5 |
| OM44 | Scania 2724 | 1345 | 1925 | 340 | 714 | 0.71 | 46.1 | 5.7 |
| OM45 | Icarus 2237 | 1357 | 2008 | 371 | 742 | 0.46 | 68.9 | 10.4 |
| Geometric mean | | | | | | 0.58 | 66.5 | 9.3 |
| Geometric standard deviation | | | | | | 1.5 | 1.3 | 1.3 |

Diesel exhaust exposures by industry are summarized in Table 11 and compared with previous studies in Table 12.

## RETROSPECTIVE EXPOSURE ASSESSMENT

### Development of Exposure Assessment

Information collected via the company questionnaire and site visits was summarized, and a common strategy for retrospective assessment of exposures was devised. The method is based on information about the work tasks and the work environment. The structure of the method reflects the process of concomitant generation at source, subsequent dispersal into the work environment, and finally, interaction of the worker with the pollutant. For each task, possible sources of the hazardous substance are identified and categorized in relation to the worker as either near-field or far-field sources. Near-field sources are those within 1 m of the worker in any horizontal direction; far-field sources are the others. For each category of source, the active emission is estimated from 3 key factors: the intrinsic emissions, the handling or processing associated with the source, and the effect of any localized engineering controls that were present at the source. These parameters define the concentration of substance emitted to the near field or far field.

The method has been developed and its use validated for different substances known to cause or suspected to cause lung cancer. Diesel particulate matter is not validated, however, although further consideration was given to how the method could be adapted to assess such exposure.

**Table 10.** Diesel Exhaust Exposures of Oil-Shale Mine Loader Drivers in Estonia

| Sample Code | Vehicle Type | Time On | Time Off | Sample Duration (min) | Volume (L) | $PM_{10}$ (mg/m$^3$) | Organic Carbon (µg/m$^3$) | Elemental Carbon[a] (µg/m$^3$) |
|---|---|---|---|---|---|---|---|---|
| OM47 | PD-8 M | 1225 | 1800 | 335 | 662 | NA[b] | 213.7 | 96.5 |
| OM48 | Toro 301-D | 1200 | 1935 | 455 | 1,001 | NA | 232.3 | 223.3 |
| OM49 | PD-8 M | 1215 | 1700 | 285 | 627 | 4.07 | 520.7 | 377.2 |
| OM51 | PD-8 M | 1210 | 1950 | 460 | 1,012 | 2.73 | 317.1 | 206.2 |
| OM52 | PD-8 M | 1228 | 1520 | 172 | — | Rejected | Rejected | Rejected |
| Geometric mean | | | | | | 3.33 | 300.9 | 202.4 |
| Geometric standard deviation | | | | | | 1.33 | 1.5 | 1.8 |

[a] Elemental carbon concentrations are likely to overestimate diesel fume exposures owing to the carbon content of oil-shale dust.

[b] NA indicates sample was not analyzed owing to losses from filter media.

**Table 11.** Summary of Diesel Exhaust Exposures by Job

| | Diesel Particles as $PM_{10}$ (mg/m$^3$) | | | Elemental Carbon (µg/m$^3$) | | |
|---|---|---|---|---|---|---|
| Industry/Job | GM | GSD | Range | GM | GSD | Range |
| **Railway transport** | | | | | | |
| Train driver | 0.80 | 1.6 | 0.34–1.60 | 18.3 | 2.0 | 10.6–71.8 |
| Shunter driver | 0.80 | 1.4 | 0.51–1.54 | 12.3 | 1.9 | 5.3–30.8 |
| Combined | 0.80 | 1.5 | 0.34–1.60 | 15.5 | 2.0 | 5.3–71.8 |
| **Bus transport** | | | | | | |
| Bus driver | 0.58 | 1.5 | 0.40–1.01 | 9.3 | 1.3 | 5.7–10.5 |
| Mechanic | 1.02 | 1.6 | 0.45–2.21 | 37.6 | 1.3 | 24.8–52.1 |
| **Oil-shale mining** | | | | | | |
| Loader driver | NA[a] | NA | NA | 202 | 1.8 | 97.0–377.0 |

[a] NA indicates samples were not available.

**Table 12.** Comparison of Concentrations of Elemental Carbon with Those Obtained in Previous Investigations

| Job/Industry | Country | Number of Samples | Type of Sample | Mean Elemental Carbon (μg/m$^3$) | Source |
|---|---|---|---|---|---|
| **Railroad train drivers** | | | | | |
| | Russia | 19 | Personal | 15.5 | This study |
| | Canada | 9 | Area | 17.8 | Verma et al 1999 |
| **Road drivers** | | | | | |
| Bus | Estonia | 5 | Personal | 9.3 | This study |
| Truck | USA | 56 | Personal | 4.0 | Zaebst et al 1991 |
| **Mechanics** | | | | | |
| Bus | Estonia | 15 | Personal | 37.6 | This study |
| Truck | USA | 80 | Personal | 12.1 | Zaebst et al 1991 |
| **Loader drivers** | | | | | |
| Oil-shale mine | Estonia | 4 | Personal | 202 | This study |
| **Ramcar driver** | | | | | |
| Potash mine | USA | 6 | Personal | 345 | Stanevich et al 1997 |
| **Winning workers** | | | | | |
| Salt/potash mines | Germany | 210 | Personal | 152 | Dahmann & Bauer 1997 |
| **Face haulage drivers** | | | | | |
| Zinc mine | USA | 19 | Area | 140,260 | Haney & Fields 1996 |
| Potash mine | Mexico | 30 | Area | 150,450,420 | Haney & Fields 1996 |
| **Typical background levels** | | | | | |
| | Russia | — | — | 5–7 (estimated) | This study |
| | Estonia | — | — | 5–7 (estimated) | This study |
| Highway | USA | 21 | Area | 3.4 | Zaebst et al 1991 |
| Residential | USA | 22 | Area | 2.0 | Zaebst et al 1991 |

The questionnaire information was evaluated for likely availability of exposure measurements and other relevant information. In particular, we aimed at assessing the type of diesel engine used, engine maintenance schedules, fuel used, and workload demanded of the engine. We also assessed the proximity of workers to the engine exhaust, the size of rooms occupied by the workers, the likely general ventilation conditions, the use of any control measures such as local ventilation or enclosed cabs, the duration of engine running, and whether the engine exhaust vented inside a building.

Quantitative exposure assessments were considered practical in companies with good contextual exposure information, preferably with exposure measurements, although additional measurements of current exposure could be collected. A ranked categorical assessment of exposure level was feasible when the information was of poorer quality. Plants with little or no information about the type of exposure were excluded.

### Evaluation of Proposed Exposure Reconstruction Method

The validity of the proposed exposure reconstruction method was evaluated using the exposure data obtained during the visits. The reconstruction method, developed in other studies (Cherrie et al 1996), was adapted by us to deal with the special circumstances of workplace diesel exhaust exposure. Further guidelines will be produced to support the assessments for diesel particulate matter. An experienced industrial hygienist was presented with descriptive information for the sampled occupations and asked to estimate exposure levels using the method. These data were then compared with measured values. Provided the correlation was good (that is, $r > 0.7$) between the measured and

estimated exposure levels, we considered the method suitable for a subsequent study.

**Theoretical Model of Diesel Exposure**

We have developed a theoretical description of the exposure process from which we plan to reconstruct past exposures in a full-scale epidemiologic study. This builds on work for an earlier epidemiologic study investigating the risks of lung cancer from man-made vitreous fibers and for other research currently underway (Cherrie et al 1996; Cherrie 1999; Cherrie and Schneider 1999).

In this scheme we use EC as a marker for diesel particulate matter. The exposure level is defined as the average concentration of EC inhaled by a worker during a task or some other defined period of time. The exposure level ($C$) is the sum of contributions from a multiplicity of background sources ($C_B$); from other more local sources, that is, in the far field ($C_{FF}$); and from sources in close proximity to the workers, that is, in his or her near field ($C_{NF}$):

$$C = C_B + C_{FF} + C_{NF} \tag{1}$$

An arbitrary distinction is made between the worker's near field, which is defined as a cubic space with dimensions 2 × 2 × 2 m centered on the subject's head, and the far field, which is the remainder of the immediate environment. For indoor spaces the far field is defined by the boundary walls of the space, while outdoors the far field is defined as being within 10 m of the subject. In modeling exposure we have normally assumed that emissions are dispersed uniformly from the source in all directions. However, this is not the case with diesel emissions because of the high initial velocity of the exhaust gases.

The background concentration of EC, which arises from diesel engines and other sources in the general urban environment, depends on many factors, including the number of vehicles actively emitting, the types of engine, and meteorologic conditions. The background concentrations probably are well represented by fixed location monitors situated some distance away from localized sources (such as busy roads) and are relatively constant at any one time across a town or city.

The far-field contribution to diesel exhaust exposure may come from the exhaust plume of the vehicle being driven by the individual or from other vehicles in the general vicinity.

The intensity of any source is determined by 3 factors: the intrinsic emission ($\varepsilon_i$), the handling or processing ($h$), and the effectiveness of any localized controls ($\eta_{lc}$). In the case of diesel particulate matter, the intrinsic emission is determined by the age and type of diesel engine being used and corresponds to the concentration measured about 1 m from the exhaust outlet, downstream, and out of the exhaust plume. The handling would be described by the way in which the engine was being driven, and the effectiveness of local controls would be determined by the presence of any particulate traps on the tailpipe. We have assumed a further reduction of exposure in a vehicle with an enclosed cab.

The 3 parameters, that is, $\varepsilon_i$, $h$, and $(1 - \eta_{lc})$, are multiplied together to provide the active emission of the source ($\varepsilon_a$). Note that the efficiency of local controls is expressed as a fraction, and that the multiplier used to obtain the reduction in active emission due to the controls is expressed as 1 minus the efficiency of the controls. Three further parameters are incorporated into the basic model: the fractional time the source is active ($t_a$), the efficiency of any personal (respiratory) protection worn by the subject ($\eta_{ppe}$), and the gravimetric dispersion of the emissions from the source ($d_{gv}$). So, for a single source close to a worker, the near-field exposure level ($C_{NF}$) would be as follows:

$$C_{NF} = [\varepsilon_{i,NH} \bullet h_{NF} \bullet (1-\eta_{lc,NF}) \bullet t_{a,NF} + \varepsilon_{p,NF}] \bullet (1-\eta_{ppe}) \bullet d_{gv,NF} \tag{2}$$

The passive or fugitive emission ($\varepsilon_p$) generally represents emission from resuspension of settled dust or evaporation of spilled volatile liquids. In the case of diesel exhaust, $\varepsilon_p$ would correspond to fugitive emissions from the engine exhaust system that may diffuse into the environment where the worker was located. For example, if the engine were mounted under or in front of the driver's cab in a truck. In addition, although workers can wear some form of respiratory protection against diesel particulate matter, in practice this is not done.

For a source in the far field of the worker, similar considerations apply, and so the following equation gives the far-field contribution to the exposure level:

$$C_{FF} = [\varepsilon_{i,FF} \bullet h_{FF} \bullet (1-\eta_{lc,FF}) \bullet t_{a,FF} + \varepsilon_{p,FF}] \bullet (1-\eta_{ppe}) \bullet d_{gv,FF} \tag{3}$$

Dispersion of the pollutant depends on the proximity of the source to the person exposed (that is, whether the source is in the person's near or far field) and on the directional nature of the initial dispersion. In addition, if the source is inside a building, then the volume of the enclosed space and the quantity of general ventilation will also determine exposure level.

**Table 13.** Recommended General Ventilation Multipliers for Reconstructing Exposure (Cherrie 1999)

| Scenario | Near-field General Ventilation Multiplier ($d_{gv,NF}$) | Far-field General Ventilation Multiplier ($d_{gv,FF}$) | Conditions |
|---|---|---|---|
| Large room and good general ventilation | 1 | 0.05 | ≥ 1,000 $m^3$ and ≥ 10 air changes per hour |
| Large workroom and poor ventilation | 1.5 | 0.5 | ≥ 1,000 $m^3$ and ≤ 1 air change per hour |
| Small workroom with good ventilation | 3 | 1.3 | ≤ 100 $m^3$ and ≥ 10 air changes per hour |
| Small workroom with poor ventilation | 15 | 14 | ≤ 100 $m^3$ and ≤ 1 air change per hour |

Cherrie (1999) has presented information about dispersion from the source, which is reproduced in Table 13. For sources within the worker's near field in a large, poorly ventilated room, the general ventilation multiplier would be 1.5. Assuming no sources are in the far field, then there would be no far-field component of exposure level (that is, $\varepsilon_{i,NF} = 0$ ). If the source were in his far field, then the ventilation multiplier would be 0.5. Also, for a small, poorly ventilated workroom, the general ventilation multiplier would be similar regardless of whether the source was in the near or far field (that is, 15 vs 14).

For outdoor diesel sources in the far field, we have assumed a Gaussian dispersion pattern. This allows the reduction in concentration to be estimated along with an adjustment of the $t_a$ to allow for variation in the plume trajectory because of changes in wind direction. For example, moving away from the source is assumed to reduce the particulate concentration approximately according to the inverse square of the distance from the source. In addition, we assume that the direction of the exhaust plume may vary according to the wind direction. The probability that it will blow from the source toward the worker is reflected in the model in $t_a$. If there is no preferred wind direction in relation to the line between the source and worker, then it is assumed that $t_{a,FF}$ would be best estimated as the angle of expansion.

### Using Theoretical Model to Predict Exposure

We have used the model to estimate average exposure level for the 5 groups of workers for whom measurements of exposure were undertaken. Estimates were made as EC with no adjustment made to estimate 8-hour time-weighted average level. The exposure assessments were carried out by one of us (JWC), who was unaware of the measurement data at the time of the assessments. Information about the exposure circumstances was obtained partly by observation of work activities (train drivers in Russia) or by descriptions from another investigator who had visited the sites, and partly from the questionnaire information or other descriptive information acquired during the study.

Little objective information was available to determine the magnitude of parameters that should be input to the model, and so most assignments were based on the judgment of the assessor. Many of the samples were obtained for drivers who, because of the nature of their job, were away from the investigators for most of the sampling duration; thus it was not possible to obtain detailed contextual information about the circumstances of the exposure. For this reason the exposure assessments were completed for all of the broad job categories identified, rather than for each individual measurement.

### Comparison of Model Predictions with Measured Exposures

The estimated EC exposures ranged from 5 μg/$m^3$ for the bus drivers to 500 μg/$m^3$ for the oil-shale miners (Figure 1). For local train drivers, the main determinant of exposure was judged to be the time that the driver spent in the station while the train was being loaded and unloaded. This activity, although it was judged to take only approximately 25% of the work time, accounted for approximately 70% of the estimated exposure for these drivers. The remainder of their exposure was considered to come from background sources within the city (that is, $C_B$). It was assumed that while the train was in motion, the driver was not exposed because the exhaust plume would be directed away from his cab.

The mean measured exposure level for the local train drivers was 18 µg/m$^3$, but excluding 2 outlying points reduced the average to 14 µg/m$^3$. These data compare well with our estimated level of 16 µg/m$^3$. We are unsure of the reason for the 2 outliers, but it is likely that these drivers worked part of their shift in an enclosed or semi-enclosed space such as an engine shed or tunnel.

The estimated exposure level for the shunter drivers (61 µg/m$^3$) was higher than the measured level (15 µg/m$^3$). The estimated level for these workers was mostly determined by circumstances: when the train was moving so slowly that the exhaust plume might blow toward the driver's cab and when the driver had his head out of the cab window. From our observations we estimated that the train was either stationary or moving backward about half of the work shift and that the driver had his head out of the window for about a quarter of this time. If he had spent only 25% of his time moving slowly and 12% with his head out of the window, then the estimated exposure would have been 21 µg/m$^3$.

The estimated exposure level for the bus mechanics was 43 µg/m$^3$, and the mean measured exposure level was 39 µg/m$^3$. The main source of exposure for this group was judged to be exhaust from buses moving around the garage (about 8% of the work shift). Drivers were assumed to be exposed for a longer period of time, but their exposure was lower because we assumed that the cab reduces the exhaust entering the driver's near field. The estimated exposure level for bus drivers was 5 µg/m$^3$, and the average measured exposure level was 9.5 µg/m$^3$.

For the oil-shale miners, the estimated exposure level was 500 µg/m$^3$ and the average measured exposure was 220 µg/m$^3$. The main reason for this group having the highest exposures was their relatively continuous work in a confined space. Part of the reason for this overestimation may have been the difficulty in judging the impact of general ventilation on controlling exposure. We used the available data for large, well-ventilated spaces, shown in Table 13, although the actual ventilation rates were much higher than this.

The association between the average measured and estimated exposure levels for all jobs combined (Figure 2) was similar to that seen in another comparison exercise that we had undertaken (Cherrie and Schneider 1999). Although the association between measurements and estimates was fairly good, there was a tendency to overestimate rather than underestimate exposure.

### Use of Model to Reconstruct Exposure

The model provides a scheme for describing exposure in a way that can help in estimating past exposure. The estimates clearly depend on the quality of information about the work and the work environment. Information about the work processes can be obtained from records and by interview with long-service employees or retired workers. This information may then provide the basis for reconstructing exposure levels.

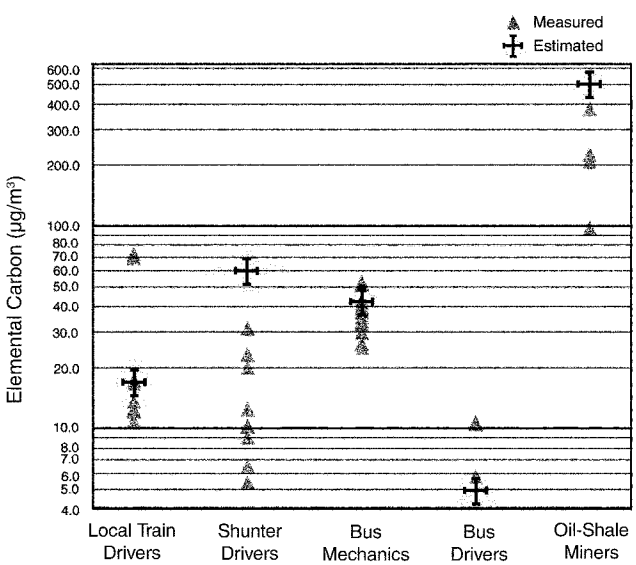


**Figure 1. Comparison of estimated and measured EC exposure levels by job.**

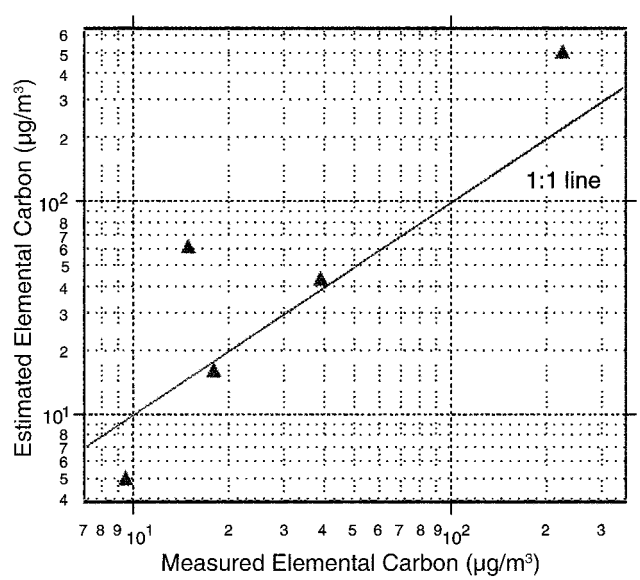


**Figure 2. Comparison of average measured and estimated EC exposure levels for all jobs combined.**

Combining measurements of current exposure levels with the exposure reconstructions provides an opportunity to minimize bias in the estimates and to refine the magnitude of component factors in the theoretical model. Further experimental investigations may result in refinement of the model parameters. Historical background concentrations of diesel particulate matter could be estimated from information about diesel traffic density obtained either from historical records or from interviews with knowledgeable local people.

## DISCUSSION

The feasibility study showed that it is possible to enroll in the Czech Republic, Estonia, Hungary, Latvia, Lithuania, Poland, Russia, and Slovakia 3 multicenter cohorts of workers exposed to diesel exhaust in nonmetal mining, bus transport, and railway transport. A total of 16 companies have agreed in principle to participate in the study, yielding an estimated 46,500 exposed workers, among whom 295 deaths from lung cancer are expected. The statistical power of the study would be adequate to assess a relatively small increase in lung cancer incidence. Groups of workers from the same companies, without exposure to diesel exhaust, are available for internal comparison. The cohorts will be primarily composed of male workers. In the main study it may be possible to obtain participation of additional companies from the countries involved as well as from Romania and Israel.

On the basis of limited environmental sampling, we conclude that current central and eastern European exposure levels are similar to those encountered in comparable workplaces in western Europe and North America.

Occupational exposure data for a possible cohort study are available in all of the companies that agreed to participate in a study. In most of the companies, however, the data would need to be complemented by additional measurements. Industrial hygiene expertise is available in these companies. Follow-up of cancer mortality would be possible in all the countries included in the feasibility study. Additional follow-up for cancer incidence is feasible in the Czech Republic, Estonia, and Slovakia, and all mining companies retained in the study are in these 3 countries.

A limitation of a possible future epidemiologic study is the lack of information on tobacco smoking history for most of the workers. Ad hoc collection of information on current workers would therefore be required to control for tobacco smoking on the basis of individual data.

One reason for conducting the feasibility study in countries of central and eastern Europe was the interest in exploring the possibility of enrolling a cohort of women with substantial exposure to diesel exhaust. The results of the feasibility study show, however, that few women were employed in jobs with high exposure. A model to reconstruct past exposure has been developed and compared with limited current exposure measurements. The prediction was accurate in 2 of the 5 job groups examined. In the 3 job groups in which the exposure predictions were outside the measured ranges, predictions were hampered by inadequate detailed information about ventilation rates, work practices, and background levels of diesel particulate matter in the ambient atmosphere. After collection of appropriate information on the companies, the model could be used in a retrospective epidemiologic study.

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## APPENDIX AVAILABLE ON REQUEST

The following appendix is available by contacting the Health Effects Institute. Please give the author name, full title and number of the report, and the title of the appendix.

APPENDIX A. Company Questionnaire

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**Graeme Hughson** is an occupational hygienist within the Research Division of the IOM, in Edinburgh, Scotland. Born and educated in Edinburgh, he has remained with the IOM for 20 years, during which time he has been involved in a range of occupational hygiene research and consultancy projects. Originally employed as a scientific technician, he obtained a distance learning degree with the Open University and later a diploma in occupational safety and health. He is now a full member of NIOSH and is actively involved in the preparation and delivery of presentations, lectures, and training courses, including a presentation to a government select committee and various specialist occupational health and hygiene courses run by IOM. His main research interests are in exposure assessment methods. Recent work includes an independent validation of the EASE model as well as a thorough review of industrial dermal zinc exposures presented at the annual conference of the British Occupational Hygiene Society in April 2000.

**Alexandre Pitard** has recently terminated his research work on the present feasibility study in the Unit of Environmental Cancer Epidemiology, IARC, in Lyon, France, funded by an IARC Special Training Award. He is now a researcher at the Regional Health Observatory in Rouen, France. He obtained a PhD in biostatistics from Besançon University in 1997 and worked in the Unit of Biostatistics and Epidemiology of the University Hospital of Besançon to develop methods to address colinearity in time series analysis. He spent one year in the Cancer and Public Heath Unit of the London School of Hygiene and Tropical Medicine as a visiting research fellow funded by the French Association against Cancer, to develop a method to assess trends in cancer survival. His current research interests include the causes of bladder cancer, the effects of air pollution on drug sales, and methodology in cancer epidemiology, in particular, estimation of cancer cases in small geographical areas.

## ABBREVIATIONS AND OTHER TERMS

| | |
|---|---|
| ACGIH | American Conference of Governmental Industrial Hygienists |
| EC | elemental carbon |
| GM | geometric mean |
| GSD | geometric standard deviation |
| IARC | International Agency for Research on Cancer |
| ILO | International Labour Office |
| IOM | Institute of Occupational Medicine |
| ISCO | International Standard Classification of Occupations |
| ISIC | International Standard Industrial Classification |
| NIOSH | National Institute for Occupational Safety and Health |
| OC | organic carbon |
| $PM_{10}$ | particulate matter 10 µm or less in aerodynamic diameter |

HEALTH EFFECTS INSTITUTE

# Investigators' Report

## Cancer Risk from Diesel Exhaust Exposure in the Canadian Railroad Industry: A Feasibility Study

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# Cancer Risk from Diesel Exhaust Exposure in the Canadian Railroad Industry: A Feasibility Study

Murray M Finkelstein and Dave K Verma

## ABSTRACT

We investigated the feasibility of studying the quantitative relation between exposure to diesel fume and lung cancer in the Canadian railroad industry. Canada's 2 major railways, Canadian National Railways (CN*) and Canadian Pacific Railways (CP), participated in the feasibility study. The investigation focused on gathering employee data, historical information, and measures of workplace exposures to diesel exhaust.

We obtained from CN a list of the approximately 74,000 employees on the payroll on or after 1983. We concluded that the populations contributed to a full study by CP would be roughly similar. We compiled a job dictionary (available on request from authors) and combined jobs with similar exposure profiles into job groups. For CN, we organized 4,451 job titles into 37 homogeneous job groups. These homogeneous job groups were further amalgamated into 5 groups of jobs with similar levels of exposure: no exposure; no exposure and not classifiable; low exposure and not classifiable; moderate exposure; and high exposure. Industrial hygiene reports pertaining to diesel exhaust provided reliable quantitative exposure data useful for a full study back only to 1990. Other information used for reconstructing historical exposures included reports for CN containing general information about past exposures going back to 1956, data from locomotive inventories, shop diagrams showing the ventilation layout, and interviews with employees. Other information exists in libraries and archives. We determined that equipment 10 to 40 years old was available for testing. Records with respect to other toxic substances were also available.

Personal and area measurements for diesel exhaust included our 155 elemental carbon (EC) samples, 166 previously obtained EC results, and data from historical industrial hygiene reports. Those reports had measurements of oxides of nitrogen ($NO_x$), carbon monoxide (CO), sulfur dioxide ($SO_2$), coefficient of haze, benzene solubles, polycyclic aromatic hydrocarbons (PAHs), aldehydes, total dust, and respirable dust. None of these historical measures correlated well with the newer measure, EC. We suspect that, owing to differences in dust exposure levels between major trade groups, respirable dust may not be an appropriate measure of exhaust.

The sampling data could generally be described with lognormal distributions. In mechanical services, we found no significant differences in the exposure profiles of different work areas or occupations. In transportation services, exposures were significantly higher in the trailing locomotives than in the lead locomotives. Exposures were ranked as follows: lead locomotives < shops < trailing locomotives. Exposures in cabooses, in the absence of travel through tunnels, were low.

A few of the major shops at CN and CP remained in use. Local and general exhaust ventilation at these shops had not changed significantly since the 1960s (the period we were interested in), but work practices and volume of work had changed. We believe that it would be possible to model historical exposures by taking account of documented changes in ventilation and work volume.

Many pieces of equipment no longer used by CP or CN were acquired by Canadian shortline railroads. Several of these small railroads permitted us to conduct exposure reconstruction tests on their equipment, and samples were thus obtained on equipment no longer used by the study railroads. These data could be used to estimate historical exposures to CN and CP employees.

We conclude that an epidemiologic study of exposure-response relations for lung cancer in the Canadian railroad industry is feasible. Canada's 2 major railroads have indicated their willingness to participate and have graciously

* A list of abbreviations and other terms appears at the end of this Investigators' Report.

This Investigators' Report is one section of Health Effects Institute Special Report *Research Directions to Improve Estimates of Human Exposure and Risk from Diesel Exhaust*, which also includes a report by the HEI Diesel Epidemiology Working Group, four other Investigators' Reports, and an Executive Summary. Correspondence concerning this Investigators' Report may be addressed to Dr Murray M Finkelstein, Program in Occupational Health and Environmental Medicine, McMaster University, 1200 Main St West, Room 3H50, Hamilton, Ontario L8N 3Z5, Canada.

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cooperated with this feasibility study. Reasonable estimates can be made of current and historical exposure conditions and employee-specific estimates of cumulative exposure can be computed. The population size would be adequate to detect relative risks increased by 15% to 20%. The exposure levels we measured were quite low and of the same order of magnitude as exposures in the general environment. Study of this population should thus inform the debate about acceptable levels of exposure for the general population.

## INTRODUCTION

The widespread and increasing use of diesel powered equipment has generated concern about the health effects of both occupational and environmental exposures (HEI 1995; Birch and Cary 1996a). Diesel exhaust has been classified as a probable human carcinogen by the International Agency for Research on Cancer (IARC 1989), a potential carcinogen by the National Institute for Occupational Safety and Health (NIOSH 1988), and a suspected human carcinogen (with an A2 classification) by the American Conference of Governmental Industrial Hygienists (ACGIH 1999). The number of workers possibly exposed in a variety of industries is considerable because diesel use is widespread (Gonzalez and Agudo 1999). Miners, bridge and tunnel workers, loading dock workers, truck drivers, bus drivers, forklift drivers, farm workers, railroad workers, and auto, truck, and bus maintenance garage workers are all exposed to diesel exhaust.

In response to concerns about the carcinogenicity of diesel exhaust, we explored the feasibility of determining dose-response relations between exposure to diesel exhaust fume and development of lung cancer in the Canadian railroad industry.

### CARCINOGENICITY OF DIESEL EXHAUST

Evidence of diesel exhaust carcinogenicity is fairly strong in animal studies (NIOSH 1988; ACGIH 1999). In 1995, the Health Effects Institute concluded that epidemiologic data were consistent in showing weak associations between exposure to diesel exhaust and lung cancer. The available evidence suggests that long-term exposure of workers to diesel exhaust in a variety of occupational circumstances is associated with an increase of 1.2 to 1.5 fold in the risk of lung cancer compared with workers classified as unexposed. In widely quoted epidemiologic studies of railroad workers, Garshick and colleagues (1987a, 1988) found a significantly increased odds ratio for lung cancer of 1.4 (95% confidence interval, 1.06 to 1.88) although these findings have been criticized (Crump 1999). More recently, long-haul and short-haul diesel truck drivers have been shown to have an excess risk of lung cancer with an odds ratio of 1.3 (95% confidence interval, 0.81 to 2.11) (Steenland et al 1998). A meta-analysis of 29 published cohort and case-control studies also supported a causal association between lung cancer and exposure to diesel exhaust with relative risks of 1.22 to 1.43 (Bhatia et al 1998). A commentary on the study suggested the need for investigations of large cohorts of heavily exposed workers with an adequate period for the development of lung cancer, with historical quantitative measurements, and with information on cigarette smoking (Silverman 1998). Another meta-analysis of 47 studies concluded that the pooled risk estimate for lung cancer from diesel exhaust is 1.47 (Lipsett and Campleman 1999).

### DIESEL EXHAUST AND ITS MEASUREMENT

Diesel exhaust is a complex mixture of compounds containing both gaseous and particulate fractions. The gaseous constituents include CO, $NO_x$, aldehydes, and PAHs. The particles have solid carbon cores that are produced during combustion and, when released to the atmosphere, tend to form chains or aggregates. More than 95% of particles are reported to be less than 1 µm in diameter (ACGIH 1999). An estimated 1,800 different substances from the combustion process can be adsorbed onto diesel exhaust particles and constitute 15% to 65% of the total particulate mass (ACGIH 1999). A widely quoted Japanese study described the discovery of a highly mutagenic and suspected human carcinogen, 3-nitrobenzanthrone (a nitro-PAH), in diesel exhaust (Enya et al 1997). Pearce (1977) has postulated that this carcinogen may be responsible for an increase in lung cancer incidence in areas with congested vehicle traffic.

Historically, a variety of contaminants have been measured and used as indexes of exposure to diesel exhaust, including simple gravimetric determinations (ie, total dust and respirable dust), CO, $NO_x$, carbon black, and specific PAHs. In the Canadian underground mining environment, gravimetric methods have been widely used to measure respirable combustible dust. In 1999, the ACGIH proposed, in its list of intended changes, a threshold limit value calculated as a time-weighted average (TLV-TWA) exposure for diesel exhaust of 0.05 $mg/m^3$ and gave it the carcinogenicity classification of A2, a suspected human carcinogen (ACGIH 1999). In documentation of the TLV for diesel exhaust, the methods of assessing diesel exposure were specified:

- Measure the mass concentration (chemically) of EC or total carbon in the submicrometer portion of the

respirable particulate, in the respirable particulate, or in the total particulate matter (EC method).

- Measure the mass concentration (gravimetrically) of the respirable fraction of the particulate matter (respirable combustible dust method).
- Measure the mass concentration (gravimetrically) of the submicrometer portion of the respirable particles (cyclone and impactor methods).

Diesel exhaust particles are primarily carbon, so the measurement of carbon is the logical surrogate. However, the many nondiesel sources of organic carbon (OC) in workplaces make a method based on total carbon measurement prone to interference. Since most of the previously used methods for measuring occupational exposure to diesel exhaust lack adequate sensitivity and specificity, a new approach, referred to as the EC or thermo-optical method (NIOSH method 5040) was developed (Birch 1998). This method measures EC, OC, and carbonate, which can be summed to obtain total carbon. A 37-mm open-face total dust assembly mode is suggested for sampling except in coal mines, where a submicron sampler must be used to minimize the collection of coal dust (NIOSH 1988).

A limited number of exposure measurement studies using one or more of the techniques suggested by ACGIH have been carried out in coal and noncoal mining environments and in trucking and railroad environments (Woskie et al 1988a,b; Zaebst et al 1991; Gangal and Dainty 1993; Haney and Fields 1996a,b; Haney et al 1997; Stanevich et al 1997). The studies conducted in US mines have included measurements of diesel particulate matter, using an impactor designed by the US Bureau of Mines. In Canadian mines, a 10-mm cyclone at 1.7 L/min has been used to collect respirable dust, which, when ashed, gives respirable combustible dust. The study in the railroad industry, conducted as an adjunct to an epidemiologic study, determined exposure as respirable particulate matter. The concentration of respirable particulate matter, adjusted to remove the fraction of cigarette smoke, termed *adjusted respirable particulate*, was selected as the marker of diesel exposure (Woskie et al 1988a).

## RETROSPECTIVE EXPOSURE ASSESSMENT

Retrospective exposure assessment is necessary to provide perspective on results of prospective epidemiologic cohort studies. Suggested methods include expert assessments, employee questionnaires, job exposure matrices, and exposure reconstruction (Esmen 1979; Bond et al 1991; Gerin and Siemiatycki 1991; Orlowski et al 1993; Hornung et al 1994; Seixas and Checkoway 1995; Armstrong et al 1996; Rybicki et al 1997). A more recent method classifies employee exposures as a function of time using Bayesian probabilistic reasoning and subjective expert assessments (Ramachandran and Vincent 1999). Another method involves structured subjective exposure assessments (Cherrie and Schneider 1999). Esmen (1979) has suggested that to improve quantitative estimation, exposure could be assessed by compiling a list of uniform tasks for each job, obtaining measurements for each of those tasks, and applying those measurements to each job. This type of analysis has been successfully performed (Dement et al 1983; Verma et al 1989). Collection and review of other types of qualitative exposure information (such as ventilation records, equipment lists, and production records) can be applied to current data to estimate earlier exposure (Verma et al 1989; Stewart et al 1991; Seixas and Checkoway 1995; Burstyn and Teschke 1999).

Questionnaires have been used when exposure information was nonexistent or rare (Gerin and Siemiatycki 1991; Birdsong et al 1992; Belletti et al 1993; Orlowski et al 1993; Teschke et al 1994). Questionnaires can be used to verify work histories, or to gain knowledge of workplace factors. Although both open-ended and closed-ended formats have been tested (Teschke et al 1994), it has been recommended that in order to maximize validity, questionnaires should be administered and analyzed by industrial hygienists and administered in person (Gerin and Siemiatycki 1991; Stewart et al 1991).

Another method to improve estimation of historical exposure is reconstruction or simulation on the basis of interviews or corporate documentation (Ayer et al 1973; Verma et al 1989; Chiazze et al 1993). In the 1989 study of Canadian hardrock miners, Verma and colleagues reconstructed working conditions for an experimental stope dry-drilling operation in order to obtain high dust readings of both konimeter and gravimetric samples.

## RAILROAD WORKERS AS STUDY POPULATION

Railroad workers are a large population repeatedly exposed to diesel emissions. Previous studies have included one of 55,000 retired American railroad workers by Garshick and colleagues (Garshick et al 1987a,b, 1988; Woskie et al 1988a,b) and another by Howe and colleagues (1983) of 44,000 CN pensioners.

### Mechanical Services

Mechanical services employees are responsible for maintenance and repair of rolling stock. They work in diesel shops, where locomotives are repaired, and car shops, where freight cars are repaired. These employees also maintain the facilities in which they work (although

engineering services employees are responsible for construction of the facilities). Job titles found in the mechanical services division include pipefitters, machinists, welders, mechanics, electricians, boilermakers, carpenters, laborers, car men, hostlers, and engine attendants. Hostlers are responsible for moving trains in and out of the repair shops. These workers can operate moving trains within the confines of the diesel shop and its associated shop and fueling tracks. Supervisors and mechanical planners are also in the repair shops at times. Individuals who work in this division are usually assigned to a particular location and work fixed hours on a specific shift. Because most of the occupations are skilled trades, new employees start out as laborers. As they gain seniority, knowledge, and skills, they graduate to a particular trade. Employees with significant experience may be promoted to supervisory positions and management.

We identified areas of diesel exhaust exposure at each shop, in order of highest to lowest risk of exposure: the turnaround area; the outdoor tracks where locomotives await repair or assignment; the outdoor fueling and sanding areas; the heavy repair area; and the rebuild or overhaul area identified as the back shop. In general, locomotives for inspection and tuning are brought into the turnaround area, which generally has large service doors at both ends of tracks 160 to 180 feet long with the capacity for 3 to 4 engines. Each track may be equipped with an overhead exhaust canopy. The locomotive engines have to be run briefly during testing, but in some shops this is done outdoors. Diesel exhaust is released into the shop during work in the turnaround area and also when the locomotives are brought into the shop. Therefore, this area of the shop has the highest potential for diesel exposure. Functions performed here include sanding and refueling (if not already done outside), brake adjustments and replacement, cleaning, stocking, and fluid level checks. Depending on workload, 2 to 5 groups of locomotives may be processed per track in an 8-hour shift. At many facilities, locomotives used to enter the shop at the end of each trip, but currently, locomotives tend to "gas and go," especially if fuel and sand facilities are located outdoors.

More extensive repairs are done in the heavy repair area, where activities include scheduled and unscheduled maintenance, steam cleaning, prime-mover repair and maintenance, power assembly replacement, parts replacement, load testing, wheel turning and replacement, welding, electrical system repair, and traction motor repair and replacement. This area is sometimes partially separated from the turnaround area. The heavy repair area is considered to have the next highest potential for exposure to diesel exhaust because the locomotives may be running continuously while in this area. The heavy repair shop has local and general ventilation for the control of fumes. Although back shops are becoming rare, this is the mechanical services facility expected to have the lowest exposure. Locomotives may receive repair lasting several weeks while in the back shop. At one time both CN and CP had several back shops, but currently the only active back shop facilities are at Calgary (CP) and Winnipeg (CN). Activities performed at these facilities have included locomotive construction and assembly, wreck repair, wheel and traction motor replacement, mid service-life refurbishment and renewal, and frame and body overhaul. These facilities are very large (on the order of 400 to 600 feet long with ceilings as high as 80 feet). Some of these facilities were built in the early 1900s and therefore have only natural ventilation. Heavy-duty mechanics, electricians, welders, pipefitters, and sheet metal workers work in these shops.

### Transportation Services

The transportation services division is responsible for generating revenue by transporting commodities to customers. Employees include individuals who operate the trains and those who are responsible for traffic control and transportation planning. Occupations include engineers, conductors, firemen, trainmen, and helpers. Supervisory staff are involved in quality control, planning, and training. Clerical occupations in this division include traffic controllers, stationmasters, and yardmasters although computerized control is replacing the last occupation. The major facilities where these people work are on the main lines and in the yards, traffic control centers, and stations. Shift work is typical; main line employees work only when work is available. When they complete a trip, they are assigned to the bottom of a list of employees waiting for the next assignment. Yard employees can work regular shifts but generally earn a lower income than main line employees. Yard crews generally include an engineer, a foreman, and a helper.

Historically, jobs on the trains included brakemen, trainmen, and firemen (coal stokers). In the past 10 years the complement has been reduced to only 2 employees, a conductor and an engineer. Occasionally, supervisory personnel such as trainmasters will ride trains. Currently, the career progression is usually from apprentice, to conductor, to engineer. Those that progress to management can become stationmasters, trainmasters, or network directors.

### Engineering Services

The engineering services division builds, maintains, and repairs property, buildings, bridges, and track using specialized equipment and other construction vehicles. Employees in this division are responsible for ensuring the

property is open in order to permit safe passage of all train traffic. Engineering services is also responsible for constructing nonrailway buildings such as storage sheds and bunkhouses. When a new facility, such as a diesel shop, is constructed, engineering services employees design and oversee the development. Occupations include field workers, gang laborers, equipment operators, signal maintainers, and track maintainers. Supervisory staff include track maintenance foremen, production supervisors, and construction engineers. Gang work, which involves track maintenance and construction, is seasonal in Canada. Many employees who work on maintenance-of-way gangs do so during the months of April to October. Although some of these employees have seniority of more than 20 years, they are often classed as temporary employees. Senior employees, especially foremen, have assignments that last throughout the year.

In general, most engineering workers are assigned to specialized crews in the peak months. These crews include rail gangs, tie gangs, welding gangs, and bridge and structures gangs and can have anywhere from 4 to 30 workers using 1 to 12 pieces of equipment. The smaller section crews or signal maintenance crews may have as few as 2 to 4 employees and a high-rail truck.

### Unionization

Most of the railroad jobs with diesel exposure are unionized. Workers are subject to union rules; collective agreements govern promotion, compensation, and job duties. Over the years, many jobs have been combined or eliminated because of major corporate restructuring. Many of these changes are outlined in the union collective agreements, which also outline cutoff dates and affected occupations. Furthermore, Statistics Canada has tracked employee statistics in the Canadian railway industry, including occupation listings and job definitions. Therefore historical job occupations (including total employment numbers) can be related to current ones.

## DIESEL EQUIPMENT USED BY CANADIAN RAILROADS

The first recorded use of diesel engines in Canada was in late 1929. Diesel locomotives became a greater part of the fleets in the 1940s and 1950s, and as a result, steam engines were gradually phased out of service. The last scheduled runs of steam engines on CN and CP were in 1960 although some units were kept in yard service for a few more years. Therefore, 1960 is taken as the cutoff date for the transition between steam and full dieselization at both companies. We obtained the annual locomotive inventories at both companies from 1955 to 1999 (Appendix B).

Before 1977, CP and CN operated their own passenger train networks. Pressure from the railways to eliminate the money-losing operations and rising public interest led to the idea of a passenger train network owned and operated by the state. Following the lead of the United States in the implementation of its Amtrak network, the Canadian government formed VIA Rail that year (Canadian Railroad Historical Association 1977). As a result, CN and CP phased out high-speed passenger trains from their inventories. According to reports from former workers, the typical exposure of a passenger train employee tends to be less than that of a freight train employee. The reason for this is that diesel exhaust cannot drift forward as easily into the locomotive cab. Furthermore, CN and CP conductors traditionally worked in the passenger compartments, which were isolated from the diesel exhaust. According to several retired employees, when VIA Rail was formed, some employees at both CN and CP transferred to the government-owned corporation to work exclusively on passenger services.

Another major change at the railways was the elimination of cabooses. Before 1989, Transportation Canada guidelines required that cabooses be attached to all trains. The conductor and tail-end brakeman used the caboose as an office where they prepared paperwork, monitored brake pressure, and sat in an elevated area, the cupola, which permitted them to monitor the train. The caboose was furnished with a washroom, cooking facilities, and a small generator (originally powered by fuel oil, later by diesel) to provide electrical power and heat. Crews away from home for extended periods used the caboose as mobile housing in order to spend more time on the road. With gradual reduction in the size of train crews and emergence of new technologies (in particular tail-end devices known as *SBUs* that control braking and monitor train functions), cabooses became obsolete. Elimination of cabooses affected employees' diesel exposures in 2 ways: conductors were moved to the head end of the train, where they could be exposed to greater volumes of diesel exhaust. Smaller crews meant that conductors had to take on more tasks that involved work in the engines and outside on the ground. This increase in exposure to engine exhaust was balanced by the decrease in exposure to caboose generator exhaust and drift-down smoke from the engines.

The last major change has been gradual restructuring of railway operations in Canada. In a move to become more efficient, both CN and CP have cut the number of jobs since 1994. At one time, each railway had more than 100,000 employees. But changes to union collective agreements, changes in regulations allowing the railways to abandon or sell unprofitable lines, and the above-noted changes in technology reduced the need for labor. Other changes included

centralized repair facilities, outsourcing of maintenance and repair, increased mechanization of engineering operations, and cuts to management overhead. Currently, the combined workforce is less than 40,000 employees.

The most significant changes occurred in transportation services. Train crews in the 1960s and 1970s consisted of 5 people: engineer, fireman (or helper), head-end brakeman (trainman), tail-end brakeman, and conductor. In the late 1970s to early 1980s, the fireman's job was eliminated, followed a few years later by the tail-end brakeman's job. By the late 1980s, the head-end brakeman's job had been merged with the conductor's job and the conductor had moved to the engine.

The other main change that accompanied this consolidation was elimination of regional mechanical service facilities. Both CP and CN abandoned many unprofitable lines and, along with them, many local diesel repair shops. From the 1960s through the 1980s, both companies had large and small diesel repair facilities. As they began to focus on main line operations, however, it became necessary to centralize repair facilities. As a result, both companies now operate only 5 or 6 main centers compared with the 13 to 15 smaller centers they previously operated. They operate modern locomotive equipment and are in the process of retiring inefficient and nonproductive rolling stock. Along with the abandonment of many eastern lines, historical diesel repair shops such as those in Moncton, New Brunswick, no longer exist. In response to these changes, regional short line railroads have emerged to take over many smaller lines and some of the facilities.

Fortunately, many of these smaller railroads still operate equipment that was used in the past by both CN and CP. It therefore would be possible to model historical operating conditions and to obtain representative air samples. Since train operation on a branch line is different from operation on a main line (slower speed and shorter trains), the average exposure to the train employee may have also changed. To control for this minor change, branch lines still used by CN and CP could be used for comparison.

## METHODS

### DATA COLLECTION

The goal of the present study was to investigate the feasibility of determining the relations between quantitative measures of exposure to diesel exhaust and lung cancer incidence (and possibly other outcomes including bladder cancer and nonmalignant respiratory disease) among workers in the Canadian railroad industry. Canadian National and Canadian Pacific Railways accepted our invitation to participate in the study. The employees of principal concern for this study, those with diesel fume exposure, worked in 3 main divisions at the railway companies: mechanical services, transportation services, and engineering services.

#### Employee Records

Contacts with staff at CN and CP facilitated collection of records pertinent to establishment of the study cohort, employee classifications, and evaluation of current and historical exposures and facilities. Computer files containing a chronological record of jobs for each employee were obtained from the Human Resources Departments. For CN, the file included all employees on the payroll in 1983 or later. Corporate restructuring at CP during this feasibility study put intense pressure on their Information Technology Department. As a result of corporate resource limitations, we were able to obtain records only for current employees by the time of this writing. Efforts are being made to obtain data for terminated and pensioned employees, and we anticipate that these data would be available for a full study.

#### Historical Records of Facilities and Industrial Hygiene

After speaking with archive specialists at both companies, we learned that CN places all of its archives at the National Archives in Ottawa, but CP maintains its own files of historical information at its archives department in Montreal. Managers and retirees suggested to us that company employees could provide the best information about historical facilities and work practices. Books and manuals were also a good source for historical information. After consulting with several railway enthusiasts and reviewing books (Bown et al 1987, 1991, 1999), we directed inquiries to current and retired employees. Walk-through surveys of different railroad facilities were arranged in order to gather information. Long-term employees were approached about the availability of shop manuals, diagrams, and equipment specifications. A short information-gathering questionnaire was developed (Appendix A) and tested on 6 retired employees. The intent of this questionnaire was to gain qualitative data on past working conditions and practices.

### EXPOSURE ASSESSMENT

#### Mechanical Services

We were given access to the maintenance shops at both CN and CP. Sampling sites for the study were selected for representativeness and convenience. Both personal and area samples for a variety of contaminants were collected

at these locations. Most of the summer sampling was conducted at CP for comparison with samples taken previously at CN as part of an earlier study (Verma et al 1999). Samples were also taken during winter to compare the effects of weather and seasonal work practices on diesel exhaust exposure.

Personal samples were collected from machinists, electricians, hostlers, and laborers directly involved in the turnaround and heavy repair areas and occasionally from the supervisor. Area samples were collected from turnaround and heavy repair areas. Personal measurements of hostlers represented the highest outdoor exposures.

### Transportation Services

***Sampling of Current Exposures*** A number of area samples were taken in the cabs of lead locomotives or trailing units of locomotives during several trips. These samples measured the engineer's and conductor's exposures while on board. The results of these tests were then combined with the results of earlier EC tests conducted by both railways into a spreadsheet for statistical analysis. We felt that area samples were truly representative of the exposures because the engineers stayed at their stations for 99% of their shifts. Some personal measurements were taken with conductors and brakemen to assess whether their exposures differed from those of engineers.

***Historical Exposures*** We wanted to test a number of locomotives representative of CP and CN fleets in the 1960s, 1970s, and 1980s to form an impression of historical onboard conditions. In order to obtain measurements for locomotives no longer in the inventory at CN and CP, we approached a number of regional short line railways about the prospect of testing their locomotives for exhaust levels. Although not a complete account of the historical inventory, this was our attempt to demonstrate the feasibility of (1) reconstructing historical exposures on old locomotives, (2) obtaining representative EC measurements for smaller regional railway lines that the large companies abandoned in recent years and that tended to use older, less-efficient locomotive power, and (3) obtaining some EC measurement in cabooses, which railway regulations no longer require on trains.

We investigated construction of a job exposure matrix based on a variety of techniques, including worker and pensioner self-assessments, published environmental data, expert assessments by industrial hygiene staff, job task evaluations, current and historical industrial hygiene monitoring, and reconstruction of past conditions.

### Engineering Services

Managers at both CN and CP considered their employees to be exposed, but with uncharacterizable exposures. Further complicating assessment of this occupational group was the fact that most work is seasonal (from April through October). In the past, exposure monitoring for these employees focused on short-term samples of enclosed areas such as stations, tunnels, and rock cuts. Since some crews use large diesel-powered equipment such as spikers, rail heaters, and boom trucks, we decided to take a few EC measurements for comparison with those made on locomotives. Assessment of exposure using historical equipment was not possible because the life span of engineering equipment is 10 to 15 years at the most. Personal and area measurements were taken.

## SAMPLING AND ANALYTICAL METHODS

The sampling duration ranged from 4.5 to 10 hours with the majority of personal and area samples 7 to 8 hours. Some shorter-term samples of less than 2 hours were also taken in shops and on trains. Spot samples were taken in tunnels to assess tunnel-specific exposures. In addition, some EC and respirable dust measurements were taken for comparison with a DustTrak (TSI, St Paul MN) continuous dust-monitoring device.

In total, 155 EC samples, 51 respirable dust samples, and 23 total dust samples were collected during this study. All samples obtained during the study were collected between April and December, representing both summer and winter conditions. Attempts were made to sample on cold days when doors were closed as well as on warm days when doors were open. When a side-by-side comparison was made between respirable dust and EC, a nylon cyclone-type sampler and an open-face total dust sampler were used. The EC method (Birch 1998) specifies the use of a total dust sampler with an open-face configuration. On 3 locomotive trips to obtain samples (one sample was spoiled by a problem in sampling rate) EC measurements were taken by an open-face sampler alongside a cyclone sampler. One manager at a short-line railway suggested that oil film and smoke levels increase when a locomotive is due for repair; therefore, we decided to take side-by-side measurements in 2 locomotives (lead locomotive and trailing locomotive) for engine oil mist, total dust, and EC.

Finally, 10 pairs of duplicate EC samples were obtained in order to conduct a small interlaboratory comparison. Two open-face EC samplers were set side-by-side, at the same flow rate of 2 L/min, and were started and stopped at the same time. One sample was sent to CANMET (Natural Resources Canada) in Sudbury, Ontario, while the other

was sent to DataChem Laboratories in Salt Lake City, Utah. As reported in the Results section, we found good comparability between the 2 laboratories.

The analytical results from this study were combined with the results of an earlier study in which 100 EC samples, 62 $NO/NO_2$ samples, and 61 respirable dust samples were taken (Verma et al 1999). Also included were the results of 66 EC samples taken by CN and CP themselves. All samples were obtained within the last 5 years and therefore were considered to represent current conditions. Both short-term and long-term samples were collected and analyzed. Short-term samples were used to identify the effect of peak exposures on daily average exposures and to measure the effects of differences in ventilation and work practices on exposure.

Air monitoring was performed using constant flow samplers (model P2500A, DuPont) and high-flow and low-flow adapted air sampling pumps (models 224-43XR, 224-PCXR3, 224-PCXR4, 224-PCXR7, SKC, and Airchek 224-52) connected via tubing to appropriate collection media. In accordance with proper industrial hygiene practices, all pumps were calibrated before and after use with a flow calibrator (DryCal DC-1, BIOS) and primary flow meters (DC-Lite) to obtain average flow rates. Deviations were noted. Quartz filter preparation prior to sampling was carried out in our laboratory in accordance with appropriate practices. Finally, field blanks were submitted with each sample in accordance with NIOSH methods.

The bulk of EC samples were analyzed by CANMET, which uses NIOSH method 5040 (Birch 1998). This method measures both EC and OC.

Respirable dust was collected by a cyclone sampler attached to a pump running at 1.7 L/min and was measured by the weight difference of a preweighed polyvinyl chloride filter in accordance with NIOSH method 0600. Dust was collected on a preweighed closed-face polyvinyl chloride filter, without cyclone, running at 2 L/min and analyzed by NIOSH method 0600. This method of sampling was used in an earlier study (Woskie et al 1988b). The $NO_2$ and NO samples were analyzed by NIOSH method 6014. The respirable dust, total dust, oil mist, aldehyde, and $NO_x$ samples were analyzed by our own laboratory, which is accredited by the American Industrial Hygiene Association (AIHA).

## QUALITY CONTROL

Quality control procedures were followed during field sampling as well as during the analysis of samples. Field sampling quality control included precalibration and post-calibration of sampling devices, use of field blanks, and monitoring of the sampling train during the sampling period. Analyses in our laboratory, as well as at CANMET and DataChem Laboratories, included the use of blanks and other quality control procedures. CANMET participates in a round-robin of EC measurements with other laboratories and provided proof of their proficiency.

## STATISTICAL ANALYSIS

### Industrial Hygiene Samples

In addition to the sampling and analytical methods, every sample was classified according to type (personal or area), location (turnaround area, heavy repair area, or on board the locomotive), and ventilation situation (doors open or doors closed). We hypothesized that the sampling results would follow a logarithmic normal distribution. Histograms, box plots, and cumulative probability distributions were produced and inspected for distributional shape and assessment of outliers.

Many of the samples contained concentrations of EC below the detection limit (BDL) of the analytical method. We used maximum likelihood estimation (Finkelstein and Verma 2001), as implemented in an Excel spreadsheet, to compute the geometric means and standard deviations in the presence of BDL values.

Multivariate analyses were performed to assess the influence of explanatory factors on the concentrations of EC. Because the data were left-censored at the detection limit, we used left-censored parametric regression modeling, as implemented in the SPlus statistical software module censorReg (Mathsoft 1999). We used the lognormal distribution as the parametric model for the data.

### Epidemiologic Sample Size

In order to assess statistical power for an epidemiologic study, we computed the number of deaths from lung cancer that would be expected in the CN cohort if mortality rates were the same as those of the population of the province of Ontario. We used the MANYRS computer program (Coleman et al 1986) to calculate expected mortality from all causes and mortality from lung cancer in 5-year age and calendar intervals. Because we had no information about actual deaths in the cohort, we adjusted the population size for intercurrent mortality by subtracting expected all-cause mortality in prior calendar intervals at entry to each subsequent 5-year calendar interval.

## RESULTS

### STUDY POPULATION

We obtained a demographic and job history file from each of the 2 participating companies. Industrial hygiene professionals and managers with long service proposed a list of jobs that had exposures to diesel exhaust that were both consistent and quantifiable. Of the *mechanical services trades*, pipefitters, electricians, machinists, and laborers had exposures that were deemed significant. Boilermakers and welders had duties equivalent to those of a pipefitter. In the *transportation services trades*, conductors, engineers, and helpers/brakemen were considered to have exposures that were significant and readily quantifiable. Other jobs reviewed and considered exposed but not quantifiable included supervisors and front line managers in the mechanical services and transportation services trades, and all hourly employees in the engineering services trades (trades responsible for right-of-way maintenance and all construction activities.) For this study we paid particular attention to the employees in the divisions with quantifiable exposures, namely transportation services and mechanical services.

#### Canadian National Data

From CN we received a data file containing a list of all employees on the payroll on or after 1983. The file included a complete list of all jobs for each employee. The numbers of employees, by division, were determined (Table 1). From an epidemiologic point of view, the total numbers of employees in each division contribute to the person-years at risk, but only the terminated employees are at risk of death from lung cancer; therefore, we cross-tabulated length of employment by year terminated for employees in transportation services and for employees in mechanical services (Table 2).

Figure 1 shows the ages at termination among the transportation services and mechanical services employees at CN. Figure 2 shows the distributions of length of service at termination for transportation services and mechanical services employees at CN.

**Table 1.** Number of Employees on CN Cohort List by Major Division

| Division | Cohort Members | Current Employees |
|---|---|---|
| Transportation services | 12,402 | 5,715 |
| Mechanical services | 20,302 | 5,481 |
| Engineering services | 13,647 | 3,972 |
| All other | 27,808 | 8,535 |

**Table 2.** Length of CN Employment by Year Terminated for Transportation and Mechanical Services Employees

| | Years of Employment | | | | | |
|---|---|---|---|---|---|---|
| Year Terminated | Fewer than 10 | 10–19 | 20–29 | 30–39 | 40 or More | Total |
| **Transportation Services Employees** | | | | | | |
| Before 1985 | 111 | 46 | 31 | 681 | 132 | 1,001 |
| 1985–1989 | 216 | 298 | 228 | 1,577 | 424 | 2,743 |
| 1990–1994 | 212 | 316 | 266 | 598 | 165 | 1,557 |
| 1995–1999 | 128 | 184 | 482 | 506 | 86 | 1,386 |
| Total | 667 | 844 | 1,007 | 3,362 | 807 | 6,687 |
| **Mechanical Services Employees** | | | | | | |
| Before 1985 | 459 | 141 | 153 | 636 | 246 | 1,635 |
| 1985–1989 | 2,375 | 1,345 | 602 | 1,538 | 506 | 6,366 |
| 1990–1994 | 439 | 943 | 771 | 733 | 238 | 3,124 |
| 1995–1999 | 372 | 1,133 | 1,264 | 870 | 57 | 3,696 |
| Total | 3,645 | 3,562 | 2,790 | 3,777 | 1,047 | 14,821 |

### Canadian Pacific Data

Because of resource difficulties caused by corporate restructuring during the feasibility study, we were able to obtain only a roster of current employees and their job histories in time for the preparation of this report. At CP 4,318 employees were in mechanical services and 5,366 were in transportation services. At CN, 5,481 current employees were in mechanical services and 5,715 current employees were in transportation services. We thus anticipate that the populations contributed by the 2 companies to a full study would be roughly similar.

## INDUSTRIAL HYGIENE INFORMATION

### Facilities and Work Practices from Walk-Through Surveys

Several walk-through surveys were carried out at diesel shops, roundhouses, and railway lines. Diesel shops varied in size and function. Operations included heavy repair, engine repair and replacement, traction motor repair and replacement, a body shop, wheel turning and repair, welding areas, parts shop, machining area, sanding, refueling, inspection, cleaning, and general maintenance. Most work was done close to the locomotive tracks, which varied in number depending on the size of the facility. Most tracks included pits that gave mechanics access to

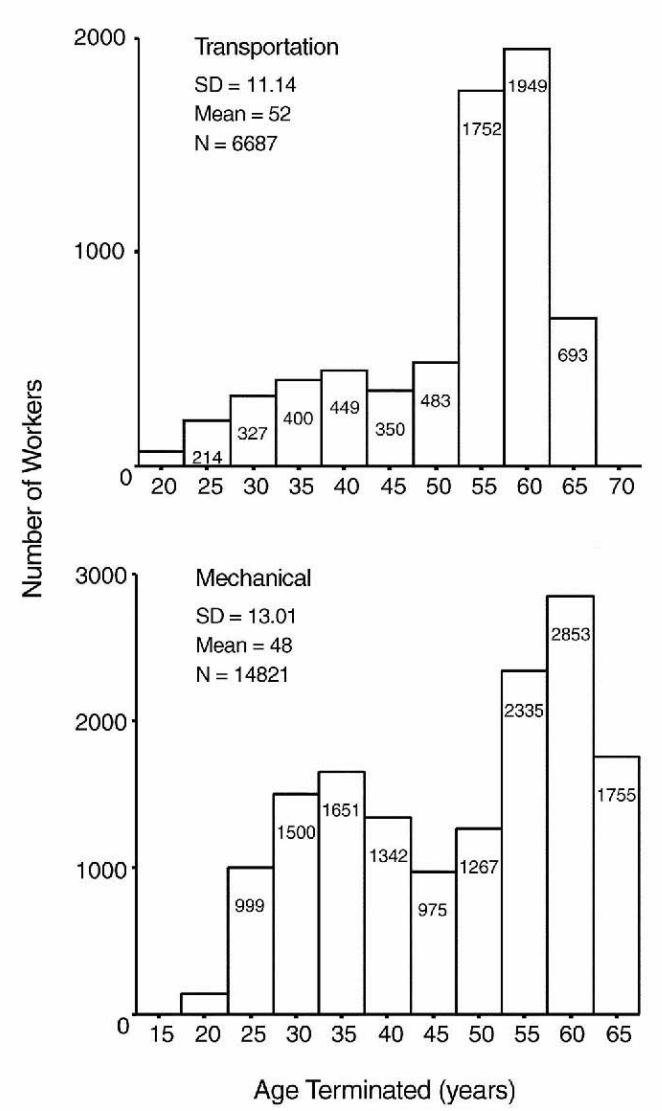


**Figure 1. Distribution of CN employees in transportation services and in mechanical services by age terminated.**

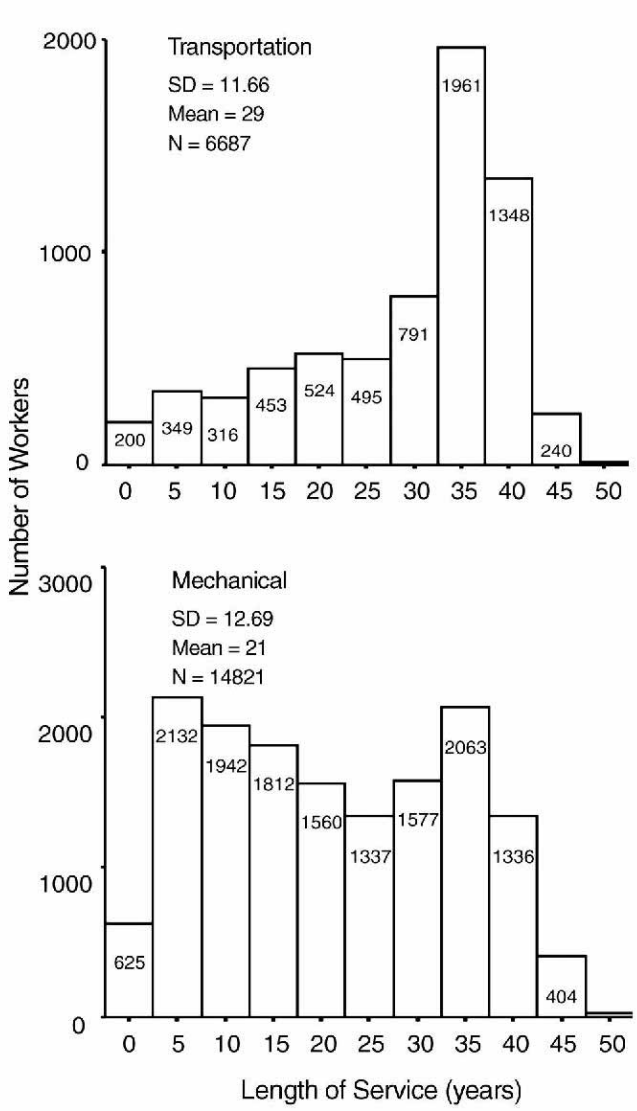


**Figure 2. Distribution of CN employees in transportation services and in mechanical services by years of service at termination.**

the underside of the locomotive chassis. Roughly half of all work in the main shops was carried out in the pits. The exceptions to this were the areas where overhead cranes were used and the back shops. In these areas, pits were not usually present. Scaffolding was present in some areas of the diesel shop.

Most work was carried out on locomotives when the engines were turned off, but some running work, in particular brake tests, was carried out indoors. The type of running work carried out indoors was governed by local rules and ventilation quality. The type of ventilation seemed to be determined by age of the facility. A mix of general and local exhaust ventilation was found in the shops.

Daily inspection, as well as trip preparation that required less than 4 hours of work, was done in the running repair, turnaround, or trip centers. The tracks in these areas usually had scaffold structures running the length of the track along with drop pits. This gave mechanics access to the cabin and engine doors and to parts for brake tests and maintenance. Depending on the age of the building, the routine maintenance track would have a combination of local and general exhaust ventilation. In the case of diesel shops, local ventilation was defined as a canopy exhaust system directly over the exhaust stack of a locomotive. Ceiling height varied but, in general, was 25 to 35 feet. Canopies were made of canvas, plastic, or steel.

### Determinants of Exposures in Mechanical Services Areas

Each day, locomotives undergoing repair, or scheduled for repair, were brought indoors. Other locomotives, waiting for refueling or assignment, were usually stored outside the shop on tracks adjacent to the facility. Although some locomotives were equipped with automatic-shutoff mechanisms (for summer), most (which could be as many as 200 locomotives) would idle outside continuously, especially during winter to prevent engine water from freezing.

The local and general ventilation systems were usually turned off except when locomotives were being tested or moved. At the end of each track was a large overhead door. Beside each door was the heating, ventilation, and air conditioning systems. During the summer, the doors were generally kept open to moderate indoor temperature and control exhaust. In winter, the doors were kept closed except when locomotives were moved, at which time the rules stipulate that doors must be left open. Some facilities permitted locomotives to be run indoors, while at others, sanding and refueling took place outside in the yard. Outdoor sanding and refueling has a dramatic effect on indoor exhaust exposure by eliminating the need to bring locomotives into the shop for cleaning and inspection. According to some employees, the ventilation system was sometimes left off while locomotives were running in the shop.

When not required to idle for a prolonged time, a locomotive would be run indoors for brake pressurization and testing, postmaintenance testing, cold weather start-ups, load testing, drying, and train movements. Cold weather start-ups would also be performed outdoors if the rules dictated this, as would full-power tests. Maintenance duties performed outdoors while the locomotive was running included sanding and refueling.

The walk-through surveys of the diesel shops suggested that diesel exhaust exposure levels have the potential to vary between shops according to work practices (indoor running of locomotives), type and age of locomotives assigned to each shop, ventilation setup of each shop, size, and geographic location. Besides diesel exhaust, other potential exposures in these facilities included grease, engine oil, solvents, silica, asbestos, welding fumes, dust, and cigarette smoke.

During the early years when diesel locomotives were used, roundhouses were used for maintenance, repair, and storage of locomotives. A walk-through survey of 2 roundhouses showed that they were constructed of wood and brick and had layout similar to those of currently used shops with respect to scaffolding and drop pits. Structural differences included construction of wood, brick, or both, low ceilings (25 to 30 feet) at the ends of each track, with a higher mid-roof section (40 to 50 feet); general ventilation or no ventilation with the exception of the doors and sliding windows (approximately one eighth to one fourth the number of fans found in current facilities); and no heating, ventilation, and air conditioning system.

### Historical Modifiers of Diesel Exhaust Exposure

The major changes and suspected modifiers of exposure in the industry are summarized in Table 3 along with their suspected effects on diesel exhaust exposure and relative importance.

***Mechanical Services*** The division with the greatest time-dependent modifiers of exposure was mechanical services. Canadian railroad companies have used different types of facilities for the maintenance, repair, and storage of diesel locomotives since their introduction 70 years ago. In his retrospective reassessment of Garshick and Woskie's 1988 epidemiologic study, Crump (1999) discussed the use of different facilities such as roundhouses and diesel repair shops. The Canadian railroad industry has used both diesel-specific facilities and adapted steam facilities. Roundhouses were initially constructed for steam locomotives. These facilities first relied upon natural ventilation for

exhaust control. As diesel locomotives were introduced, the roundhouses were not properly equipped and therefore had to be adapted with general exhaust vents installed in the ceiling.

As diesels replaced steam engines, both CN and CP constructed diesel-specific facilities. Initially, these facilities were constructed with general exhaust ventilation, but some were constructed with dedicated local ventilation. This ventilation generally consisted of several axial fans, 4 feet in diameter, installed in the ceiling of the buildings in which locomotives were stored and maintained. Doors were generally left open during summer months to improve ventilation, whereas they were closed in winter. Several employees described conditions in the shop as being better in summer than in winter. However, these descriptions were based solely on their observations regarding indoor haze and odor.

The last roundhouse being used for diesel repair in the Canadian railroad industry was closed in the late 1980s. Both CN and CP maintain several large diesel facilities. The diesel shops can be broken into 3 distinct areas: (1) The back shop or heavy repair shop is where scheduled heavy maintenance occurs. Tasks in this area include traction motor replacement, engine removal and replacement, or any other complex maintenance. (2) The running repair or turnaround area is where locomotives typically receive routine preventative maintenance including restocking, cleaning, refueling, brake testing, and brake replacement. Some diesel shops are equipped for indoor sanding and refueling, whereas others are equipped only for outdoor sanding and refueling. (3) Finally, diesel storage is where locomotives are kept for assignment and scheduled maintenance.

In recent years, both CN and CP have abandoned or sold their smaller facilities and some of the largest back shops. Very recently, CP laid off employees at Ogden Shops in Calgary in a move that shadows another recent trend in heavy industry, outsourcing. Table 4, adapted from *The Canadian Trackside Guide* (Bown et al 1987, 1991, 1999),

**Table 3.** Effect of Work Practices and Engineering Changes on Exposure

| Work Practice/Engineering Change | Time of Change | Effect on Exposure | Significance of Change |
|---|---|---|---|
| **Transportation services** | | | |
| Longer trains | 1980s–1990s | Increase | Minimal |
| Elimination of caboose | 1989 | Increase | Moderate |
| Low sulfur fuel | 1970s | Decrease | Minimal |
| Cab seals, positive pressurization of electrical cabinets | 1980s | Decrease | Significant |
| Run locomotives short nose forward | 1970s–1980s | Decrease | Significant |
| Train handling | 1980s–1990s | Decrease | Minimal |
| Increased weight per car | 1990s | Unknown | |
| Longer hours, extended runs | 1980s–1990s | Increase | Moderate |
| Tunnel ventilation, purge requirements | 1980s–1990s | Decrease | Significant |
| **Mechanical services** | | | |
| Centralized repair | 1990s | Increase | Moderate |
| General ventilation | 1960s | Decrease | Significant |
| Local ventilation | 1960s–1990s | Decrease | Significant |
| Low idle & automatic start/stop systems | 1980s–1990s | Decrease | Moderate |
| Consolidation of job tasks (hostler and laborer) | 1990s | Increase | Moderate |
| Indoor shut-down policy (depends on region) | 1980s–1990s | Decrease | Significant |
| **Engineering services** | | | |
| Use of modified road vehicles as opposed to trackmobiles for transportation | 1980s–1990s | Decrease | Minimal |
| Mechanized road building | 1970s–1990s | Increase | Moderate |
| **Other significant changes** | | | |
| Contracting out of asbestos control and remediation for locomotives | 1980s–1990s | Decrease | Significant |
| Nonsmoking policy (in diesel shops only) | 1990 | Decrease | Significant |

shows a list of mechanical services facilities compiled for 1987, 1991, and 1999. Before 1991, almost all of CP's locomotive power east of Ontario consisted of Alco or Montreal Locomotive Works (MLW) locomotives. At the same time, power assigned to Ontario shops consisted mostly of older remanufactured GM engines with a lot of shop switchers and low-power road switchers. Power assigned to shops from Manitoba west to British Columbia consisted of remanufactured road switchers, shop switchers, and 1970s-era GM locomotives. Many of these larger GM locomotives have since been moved into Ontario and Quebec as the MLW locomotives were retired and sold.

**Table 4.** Major Diesel Repair Facilities and Number of Locomotives Serviced in 1987, 1991, and 1999

| | | Number of Units Serviced[c] | | |
|---|---|---|---|---|
| Shop Name, Location[a] | Type of Shop[b] | 1987 | 1991 | 1999 |
| **Canadian National** | | | | |
| Prince George BC | RR | 69 | 87 | 9 |
| Prince Rupert BC | RR | 8 | — | — |
| Thornton Yard Vancouver BC | HR, RR | 79 | 40 | 60 |
| Walker Yard (Calder) Edmonton AB | HR, RR | 300 | 360 | 105, GM maintains 329 |
| Saskatoon SK | RR | 63 | 28 | 20 |
| The Pas MB | ? | 16 | 10 | — |
| Symington, Winnipeg MB | HR, RR, back shop | 246 | 239 | 131, GE maintains 158 |
| Neebing Yard, Thunder Bay ON | RR | 43 | 27 | — |
| Capreol ON | Roundhouse | 11 | — | — |
| Windsor ON | ? | 10 | — | — |
| Sarnia ON | RR | 19 | 28 | — |
| Fort Erie ON | ? | 51 | — | — |
| MacMillan Yard, Toronto ON | HR, RR | 259 | 188 | 308, GM maintains 223 |
| Taschereau Yard, Montreal PQ | HR, RR | 390 | 440 | 119 |
| Montreal PQ | ? | — | 30 | — |
| Point St Charles, Montreal PQ | Back shop | 30 | — | — |
| Senneterre PQ | RR | 54 | 33 | — |
| Gordon Yard, Moncton NB | RR | 236 | 253 | — |
| Rockingham Yard, Halifax NS | Roundhouse | 29 | — | — |
| St John's NF | ? | 36 | — | — |
| **Canadian Pacific** | | | | |
| Coquitlam BC | ? | 82 | 74 | 58, GE maintains 184 |
| Alyth Yard, Calgary AB | HR, RR | 303 | 326 | 272 |
| Ogden Shops, Calgary AB | Back shop | 1 | 1 | 1 |
| Moose Jaw SK | HR, RR | 10 | 21 | 58 |
| Sutherland Yard, Saskatoon SK | ? | 15 | — | — |
| Winnipeg (Weston Yard) MB | HR, RR | 416 | 384 | 44, GM maintains 229 |
| Thunder Bay ON | ? | 26 | 33 | 39 |
| Sudbury ON | ? | 11 | 11 | — |
| Toronto (Agincourt Yard) ON | HR, RR | 142 | 166 | 277 |
| St Luc Yard, Montreal PQ | HR, RR | 234 | 243 | 18 |
| Bayshore Yard, St John NB | ? | 10 | 14 | — |
| Kentville NS | ? | 5 | 4 | — |

[a] List does not account for smaller service centers that do not have official locomotive assignments. BC indicates British Columbia; AB, Alberta; SK, Saskatchewan; MB, Manitoba; ON, Ontario; PQ, Province of Quebec; NB, New Brunswick; NS, Nova Scotia; NF, Newfoundland.

[b] RR indicates routine repair; HR, heavy repair.

[c] Numbers include nonpowered locomotives used for traction motors only.

New GE and GM locomotives have been replacing the 1970s GM locomotives in western Canada.

In contrast, CN maintained a mixture of MLW and GM diesels (high-horsepower road switchers and shop switchers from the 1950s to the 1970s) from Ontario eastward, with all MLWs located in Montreal and eastern Canada. In western Canada, CN had a blend of locomotives built from the 1950s to the 1980s. By 1991, MLWs were concentrated in the Maritime Provinces east of Quebec as CN began to replace its fleet with new GM and GE locomotives throughout Canada. Currently, all early 1990s GE locomotives are concentrated in western Canada, and new GM locomotives have been introduced in both eastern and western Canada.

In recent years mechanical operations have become increasingly centralized as small shops closed and rail lines were abandoned. Many of these shops serviced smaller road switchers and shop switchers from the 1950s and 1960s. Exposure estimates would require the investigation and modeling of these facilities. The roster of current facilities indicates that the Prince George or Saskatoon facilities for CN and the Thunder Bay facilities for CP would be appropriate locations for modeling these conditions.

We searched for historical ventilation drawings of diesel repair facilities. Canadian Pacific provided us with blueprints of several old facilities including roundhouses and diesel shops. When CP introduced diesel locomotives, it apparently had engineering processes in place to create proper ventilation in the shops. Although cruder and less efficient than dedicated local exhaust systems, all of the facilities reviewed had general ventilation in the form of ceiling fans. Some of the drawings included fan specifications and designed throughput. (A sample index of blueprints available for one CP shop is included in Appendix C.) CN was not able to provide us with blueprints because a search for these would have entailed manual review of all blueprints organized by subdivision, a process estimated to involve 2 or 3 months of work. CN did indicate that blueprints were stored both in Montreal and in Edmonton, but the staff were unable to estimate the quality of these documents in terms of age and content. The hours of work have not changed significantly in the mechanical services divisions. Employees work standard 8-hour shifts on rotating schedules as they always have. Division of labor has not changed much, although in some shops tasks have been merged. For instance, at CN in Winnipeg, duties formerly divided among pipefitters, boilermakers, welders, and machinists are now combined under the general heading of heavy-duty mechanic. Other duties, particularly heavy repairs, have been contracted to other companies leaving only scheduled maintenance and trip preparation in the hands of CN and CP.

***Transportation Services*** In the transportation trades, the 2 greatest changes in work practices that have affected exposure are the elimination of cabooses on trains and the running of trains with the short nose forward. According to some retired employees, the original convention of running a locomotive with the engine compartment in front of the cab was a carryover from the steam era. The main reason for running this way was that the long nose of the engine provided greater protection for the crew in the event of a collision. However, the drawback was reduced visibility and greater exposure to exhaust drifting in through the windows. In the early 1970s, the railroads introduced locomotives with controls oriented the opposite way: short nose forward. Although single-locomotive trains on the few remaining branch lines sometimes run long nose forward and switcher-type locomotives are oriented this way, the practice is no longer common. Other than these 2 changes, the job of operating a train from the perspective of brakeman (now conductor) and engineer is currently much the same as it was in the 1950s.

Both CN and CP have been using diesel-powered equipment since the 1920s (Canadian Railroad Historical Association 1988), but dieselization was not complete until 1960. Since the 1960s, Canadian railway companies have used locomotives built by a variety of manufacturers, predominantly GM, MLW, and GE (Bown et al 1999). The principal difference between the GM locomotives and the Alco or MLW locomotives is the engine. The GM locomotives run on a 2-stroke cycle. Alco and MLW locomotives use 4-stroke engines. Other differences among the manufactured locomotives included engine size, displacement, induction method, and number of cylinders. To determine which locomotives to select for testing, we prepared a database of annual locomotive inventories at the railway companies (Appendix B).

A number of trends were apparent. According to employees, locomotives tend to migrate from West to East. New locomotives are usually introduced into service in western Canada, where freight revenues are more profitable. As they age and become less reliable for heavy freight service, they are moved to eastern Canada. They also tend to be moved from main line service to branch line or yard service.

Another apparent trend was the increased use of high-horsepower locomotives. In the 1950s and 1960s, many locomotives were purchased with lower power specifications (1,000 to 2,000 hp). Since the 1970s, most locomotives have been purchased with higher power specifications (3,000 to 4,000 hp). Newer ones are being built with the

capability of generating 6,000 hp. As a result, current trains are longer and require fewer engines. For example, grain trains in western Canada can run with 3 new GE locomotives as opposed to 5 of the older GM locomotives. CP has studied the effect of this change on exhaust emissions in tunnels (CP Rail, internal report 1994). Its locomotive roster is slightly older than CN's. CN made greater purchasing efforts in the mid 1970s and from the late 1980s to the present. CP had more evenly distributed purchasing between 1970 and 1990 with many locomotives purchased recently. When the American subsidiaries were excluded, CN's Canadian fleet was 300 units larger than CP's. Further analysis showed that CN had marginally more road freight units than CP.

The third trend was the gradual elimination of MLW and Alco locomotives from the Canadian fleets. According to employees, these locomotives were considerably smokier than GM units. Further, limited parts availability and longer maintenance downtimes, combined with the move toward efficiency in the mechanical division, resulted in their elimination from railroad fleets. In order to collect exposure data for these locomotives, we approached some of the short line railroads that had purchased them. Six yard and freight locomotives were selected for testing in order to obtain representative numbers for EC exposure for comparison with samples of current locomotives.

Locomotive motors consist of 3 components: the prime mover or engine block, the power assembly or cylinder package, and the generator/alternator. The prime mover is the housing for the power-generating components, the power assemblies. They are complete packages of injectors, valves, cylinders, and sleeves, and when worn out, they are replaced in their entirety. The generator/alternator is the electrical device that converts combustion power to electrical power and drives the wheels.

Alco and MLW locomotives used different variations of the 251B power assembly. The difference between a low-power locomotive and a high-power locomotive was the number of cylinders. In other words, a 1,000-hp switcher might have only 6 cylinders, whereas a freight engine with 3,000 hp might have 16 cylinders. Early versions of this locomotive used a completely different power assembly and were difficult to find. General Motors locomotives have been equipped with variants of the 567, 645, and 710 power assemblies since the 1950s. General Electric locomotives use the 7FDL16 power assemblies and are considered the evolution of the Alco and MLW engines.

Unlike automobile engines, locomotive power assemblies are changed as entire units every 5 to 7 years. The technology of the assembly may be improved when the new one is installed. The power assembly can incorporate improved cylinder design, better fuel injectors, or a lighter shaft, thus improving engine efficiency. One study carried out for the Association of American Railroads (AAR) (Markworth et al 1994) evaluated the effect of upgraded components on exhaust emissions. We concluded that a 35-year-old locomotive could have better particulate emissions characteristics than a 2-year-old locomotive.

A more radical modification of locomotive engine is to replace or rebore the prime mover to a larger power assembly. This is common in GM locomotives. As components for the 567 power assemblies found in 1950s and 1960s locomotives become more difficult to obtain, many of the locomotives are being converted to 645 power assemblies. The AAR carried out studies to test the implications of these changes and concluded that locomotives with a larger bore emitted fewer particles on a gram-per-horsepower-hour basis (Conlon 1988; Fritz et al 1995).

Environment Canada compiles statistics of locomotive emissions (Transportation Systems Division, Environment Canada, Railway Association of Canada 1999). Its inventory of particulate emissions from 1975 to 1999 suggests that locomotive particulate emissions have decreased continuously over time. However, the data were based on emissions characteristics of locomotives tested in the AAR reports by Southwest Research Institute, which did not test particulate emissions by MLW or Alco engines specifically. Tests made on a GE engine with a 4-stroke cycle similar to that of the Alco engine revealed emissions similar to those of a GM engine (Fritz et al 1989). According to a company engineer, his team concluded that MLW locomotives were similar to GM locomotives in both fuel consumption and particulate output. This assessment seems to conflict with ad hoc employee reports that MLW locomotives were smokier than GM locomotives during start-up, but smoke often results from incomplete combustion due to turbo lag, poorly functioning governor, turbo damage, or improper fuel injection.

Turbo lag occurs when throttle is increased. During start-up, increased quantities of fuel are injected into the combustion chamber as an aerosol. This low initial air-fuel mixture results in incomplete combustion. To control the amount of fuel injected at any power setting, a governor is used. Once the engine is up to speed and sufficient air is forced into the chamber, a greater air-fuel mixture can be used. Turbos on all 4-stroke locomotives such as Alcos and GEs have always been exhaust-driven only. As the engine responds to increased fuel injection, the initial low level of exhaust gases at low revolutions per minute is unable to power the turbo, which is supposed to force air into the combustion chamber. In contrast, GM engines feature a turbo that is crankshaft-driven at low engine revolutions per

minute. A clutch disengages the turbo at high revolutions per minute, which then can operate on exhaust drive alone. The turbo assist is required because the 2-stroke piston is unable to create the negative pressure inherent in 4-stroke pistons that draws air into the combustion chamber. Old GM 2-stroke engines used a Roots Blower supercharger.

The old injectors were mechanical, whereas the new ones are electronic. Electronic injection is better able to control the amount of fuel entering the combustion chamber. Newer designs enable engineers to customize the timing and amount of fuel to optimize economy, emissions, or power characteristics. With both new and old injectors, however, fouling of the injector tip can cause the improper mixture or a poor aerosol to be injected, resulting in improper combustion. Likewise, a damaged turbo charger can cause the wrong quantity of air to be injected into the engine, resulting in elevated smoke levels. Researchers have studied opacity and particulate emissions (Markworth et al 1990; Fritz et al 1995) but have been unable to define specifically the relation between visible smoke (opacity) and particulate emissions. Visible smoke cannot, therefore, be considered a reliable indicator of exposure.

***Engineering Services*** The current engineering services tasks are similar to those in the 1950s and 1960s. Historically, this type of work involved heavy physical labor; however, many of the tasks that were performed by hand during the steam era are now performed by machine. According to current and former employees, diesel-powered equipment was already in use when diesel locomotives were introduced. Other work practices, such as the use of personal protective equipment, have also been noted, although protective equipment for diesel exhaust, with the exception of an occasional dust mask, is not used. When asked whether significant changes in work practices had occurred over the years, many workers agreed that only improved ventilation in enclosed areas (such as tunnels) and regulations concerning general safety have made a difference. The employees made more comments about road dust and creosote wood preservatives than about diesel exhaust. Also, better door seals and cab heaters have improved the working conditions in these trades. Finally, this trade seems to be following the same outsourcing trend found in the mechanical trades. Although most work is still performed by CN and CP employees, some tasks such as rail inspection and grinding are increasingly being contracted to specialized companies. This outsourcing is still a contentious issue between the unions and the companies.

Engineering gangs use a variety of specialized diesel-powered equipment including cranes, spikers, rail aligners, ballast regulators, rail heaters, and motor cars. The maintenance equipment is assigned to a particular region and used for many years. Although the average age of equipment is not available, it generally is used for 12 to 15 years until no longer serviceable or useful. This is significantly less than the serviceable age of most locomotives (30 to 40 years). Over the last 50 years, many tasks such as rail laying or tie laying have been mechanized. In the opinion of some workers, however, the current equipment is not significantly different from that used in the late 1970s. Workers did note that mobile lodging cars and motor cars have been replaced by hotels and high-rail trucks (pickup trucks with pneumatically lowered rail bogie wheels).

**Exposures Other than Diesel Fume**

Mancuso (1983) reported the results of a mortality study suggesting that diesel shop workers were at risk of occupational cancer due to asbestos exposure. Garshick and colleagues (1987a,b; 1988) reported interviews with steam shop workers which suggested that they received considerable exposure to asbestos during their careers. This was especially true of boilermakers, pipefitters, and machinists, who used asbestos for boiler and pipe insulation as a fire retardant. We performed a preliminary search for information about asbestos use, control, and exposure at CN and CP during the diesel era.

Limited documentation was found at CN with respect to asbestos exposure in diesel shop workers, car shop workers, and transportation trades and in areas of locomotive insulation. Furthermore, asbestos was used as insulation in freight cars and as a fire retardant in diesel shops, especially roundhouses where Transite board was used to prevent fires from fire-box sparks on steam engines. The information revealed that CN had performed exposure monitoring as far back as the 1980s and had a control program and biological monitoring program in place by the early 1990s. CN has had a program in place since 1991 to gradually replace all asbestos insulation used on locomotives. In general, asbestos was found on first-generation and second-generation locomotives built between the 1950s and 1970s. Thus, until recently, some trades, especially pipefitters, would have been exposed to asbestos. The other major exposed group was car men, who would strip and repair rail cars. Internal industrial hygiene investigations of asbestos fiber exposures of CN car men were reported to be below established TLVs.

No documentation on asbestos exposure monitoring or affected trades is available from CP. One discussion with a diesel shop employee revealed that as late as the mid 1990s, CP had unknowingly purchased asbestos insulation to be installed on locomotives even though an asbestos control

program was in place. Asbestos removal at CP has been contracted out to service firms rather than being performed by CP employees. During one walk-through survey of a diesel shop, a stripped-down locomotive appeared to have asbestos insulation at the nose of the body and around the sanding compartment. This observation should be regarded with caution, however, because no sample of the suspected substance was obtained for testing. After reviewing the documentation and speaking with industrial hygiene specialists, we decided that pipefitters, sheet metal workers, boilermakers, car men, and scrap burners have been exposed to asbestos since the 1960s. In addition, machinists and electricians have been moderately exposed. In the running trades, brakemen and conductors who worked in cabooses before 1990 may have been exposed to asbestos brake-pad dust. However, no exposure information exists to support or refute this claim. In summary, quantitative estimation of lifetime asbestos exposure at CN and CP is not possible owing to the scarcity of exposure data, and only ad hoc reports may be used to judge exposure of this important confounder.

Silica exposure is also a factor to consider. Silica was formerly used in sandblasting and is still used for traction on locomotives. An internal CN document showed that as early as 1983, CN was using steel shot and glass beads for sanding in enclosed areas. This document also lists exposed occupations as car men, machinists, and helpers. Fine silica sand is still used for traction control. Often when locomotives are pulling heavy trains or operating in wet conditions, the wheels slip. When sensors indicate a loss of traction, locomotives are equipped with injectors that spray sand under the wheels to control grip. When sand is applied, a fine cloud of dust may develop around the undercarriage of the locomotive and drift into the cab of trailing locomotives or cabooses. According to former employees, conditions in the cabooses of trains were usually dusty, and sometimes these workers could tell when sand was applied. Sand is not applied constantly, however, so actual exposure to silica may only account for 1 to 2 minutes per shift.

In contrast to the running trades, some of the diesel shop workers are exposed to silica sand from these machines for a longer time. This exposure generally occurs during turnaround and trip preparation when laborers fill the locomotives with sand from an overhead hopper. This operation may be performed indoors or outdoors. Protective respiratory equipment is provided, but some workers have been observed not using their equipment. Most often, this procedure is performed by any of the laborers in the diesel shop. At one time though, some workers such as laborers and hostlers were assigned the sole duty of sanding and refueling. How long protective equipment has been issued to workers is not known, but it has been assumed that exposure control at CN has been conducted since the early 1980s. As with asbestos, quantitative estimation of silica exposure is not possible although internal documentation at both companies showed that exposures ranged from 0.05 to 0.228 mg/m$^3$. The highest exposures exceeded the current limits of exposure to respirable crystalline silica.

The final factor for consideration is welding fumes. Although not performed extensively in the average diesel shop, welding does occur in the diesel heavy repair area or back shop, in the car shop, and in the maintenance-of-way gangs, especially welding gangs. In 1998, Human Resources Development Canada–Labour Branch carried out a survey of welding fume exposures (mainly in the railway industry although not limited to that industry). This report concluded that welding fumes were not a significant hazard although some samples at a CP back shop exceeded the permissible limit when local or general exhaust ventilation was not provided.

### Industrial Hygiene Surveys and Records

All industrial hygiene records and reports pertaining to diesel exhaust were obtained from the head offices of CN and CP. Since the 1950s, CN has carried out hazard analysis, with area and personal monitoring. Contaminants studied have included particulate matter, carbon black, NO, $NO_2$, CO, volatile organic compounds, silica, welding fumes, $SO_2$, total dust, asbestos, and respirable dust. Measurements for CO, $SO_2$, $NO_2$, and NO involved short-term spot samples and long-term samples. Measurements for smoke and particulate matter included coefficient of haze and high-volume filtration samples, coal tar pitch volatile compounds, and total dust. Some reports resulted from research projects carried out by the federal government or the research department at CN, while others resulted from investigations into accidents or employee complaints of working conditions. Although only a few records can be found pertaining to surveys carried out before 1980, the available records permit tracking and comparison of results using $NO_x$ or CO as the benchmark indicators.

Several surveys carried out in the 1980s and early 1990s showed that the analytical method used for diesel particulate matter was measurement of total dust. Total dust and analysis of the gas constituents can be used to generate relative comparisons between current conditions and historical conditions. However, $NO_x$ correlates poorly with EC measurements, and respirable dust correlates weakly (Verma et al 1999).

Canadian Pacific had no record of industrial hygiene monitoring before 1990. Moreover, surveys in the early 1990s focused on special areas such as the tunnels in western Canada, where $NO_2$, NO, CO, formaldehyde, and $SO_2$ were

sampled. Only in the last few years has the company carried out industrial hygiene surveys in the diesel shops or on trains. EC sampling carried out at both railway companies since the mid 1990s included studies both of locomotives and mechanical facilities.

One of the problems complicating use of prior EC data was the lack of uniform industrial hygiene practices. Inconsistencies existed in the provision of blank samples as specified by NIOSH method 5040, treatment of detection limits, and reporting of data. Standard practice was to submit one blank, as a minimum, with each set of samples. Analysis of company hygiene data revealed that some sets of samples were submitted for analysis without the addition of blanks. Since a blank would be used to correct any measured value by actually adjusting downward the final result, some EC values may be biased high. In addition, the laboratory practice at McMaster University (AIHA accredited) is to subtract the detection limit per filter from the reported measured value. Therefore, any blank-corrected values that are less than 1.7 µg/filter (as reported to be the detection error by the laboratory) are reported as less than 1.7 µg. Some of the earlier industrial hygiene reports either failed to report the detection limits or provided a detection limit of 3 standard errors (common practice). As a result, some values may have been reported as below detection when the measured results were in fact valid according to the McMaster University method. Finally, some reports contained only time-weighted values for concentration and not the actual filter weight. We took these concentrations as valid without having the capability of verifying the concentration on the basis of sample volume and filter weight.

### Important Historical Surveys

One important record of industrial hygiene monitoring located at CN was an extensive inventory of occupations and related hazards in the mechanical services and engineering services trades. We extracted the information to create a database that could be used to compare similar tasks among jobs.

In 1991, CN had carried out an industrial hygiene analysis of tasks. This involved assessing a number of mechanical services and engineering services occupations in central and eastern Canada for relevant tasks, exposures, and use of personal protective devices. Although many tasks were described, we decided to examine the jobs for key tasks and activities such as maintenance and repair. We transferred the written information to a database. This task analysis did not involve the transportation services trades. Although CN has undergone reorganization, these trades have not changed significantly. A summary of the most common tasks is given in Table 5.

### Sampling Results for Mechanical Services

We combined the results of sampling obtained during the feasibility study with those obtained previously in surveys by McMaster University and the 2 companies, providing 195 samples for analysis. Of these, 65 (33%) contained EC concentrations below the detection limit of the analytical method.

We were interested in determining the mean concentration of EC, the statistical distribution of concentrations among the samples, how the concentrations related to job title, area sampled, season, and door position, and whether they were personal or area samples. In the results presented here, we proceed from simple descriptions to more complex multivariate analyses, taking account of those samples whose results were below the detection limit of the analytical method.

***Statistical Distribution of Sample Results*** In a lognormal quantile plot of the EC sampling results for mechanical services (Figure 3), apart from a few overly large results, the overall distribution can be seen to be basically lognormal.

***Area and Personal Sampling*** In a dot plot of the EC results by area samples versus personal samples (Figure 4), with results below the detection level displayed as 0, the distributions can be seen to be similar. Formal testing of the null hypothesis (no difference between these distributions) presented a statistical challenge because 33% of the observations were below the detection level and because the detection limits varied by sampling times and volumes. To analyze these data, we used left-censored parametric regression modeling as implemented in the SPlus statistical software module censorReg. We used lognormal as the parametric model for the data. This model did not reject the null hypothesis of equal distributions ($P = 0.69$).

***Season*** There were 120 summer and 75 winter samples. Season might influence worker exposure because the shop doors would be closed during the winter, influencing ventilation, and because the temperature might influence the emissions of diesel engines at cold start-up. Surprisingly, in a dot plot of the EC sampling results by season (Figure 5), the summer results were significantly higher than those obtained during the winter ($P < 0.001$).

***Indoor Versus Outdoor Samples*** We compared the results of sampling indoors versus outdoors, controlling for season. The EC concentrations of indoor samples were 1.5-fold higher, but the difference was not statistically significant ($P = 0.4$).

**Table 5.** Most Common Mechanical Services and Engineering Services Tasks at CN and Associated Jobs

| Task | Associated Jobs |
|---|---|
| **Mechanical services (diesel shop only)** | |
| Locomotive inspection and repair | Boilermaker, electrician, machinist, pipefitter, sheet metal worker |
| Shop maintenance | Laborer, machinist, electrician, pipefitter |
| Sanding and refueling | Laborer, machinist |
| Moving | Laborer, hostler's helper |
| Cleaning and restocking | Laborer |
| Locomotive maintenance | Electrician, pipefitter, car man, mechanic, sheet metal worker |
| Brake repair | Machinist, car man |
| Road repair | Electrician, machinist |
| Rebuilding | Machinist |
| Retooling | Machinist |
| Fabrication | Boilermaker, machinist |
| Welding, grinding | Boilermaker, car man, pipefitter |
| Lab analysis | Machinist |
| Locomotive troubleshooting | Machinist |
| Supervision, assisting | Mech planner supervisor, program supervisor, car supervisor |
| **Engineering services** | |
| Inspection, supervision, assisting | Asst track supervisor, asst track maintenance foreman, extra gang foreman, production supervisor, track maintenance foreman |
| Building, maintenance, repairs | Bridgeman/building and structures |
| Loading | Extra gang laborer |
| Cutting, drilling, strengthening rail | Extra gang laborer |
| Undercutters, ballast, tie injectors, cranes | Machine operator |
| Inspections, replacements, switch lubrication, ditch work, snow removal | Track maintainers trackman |

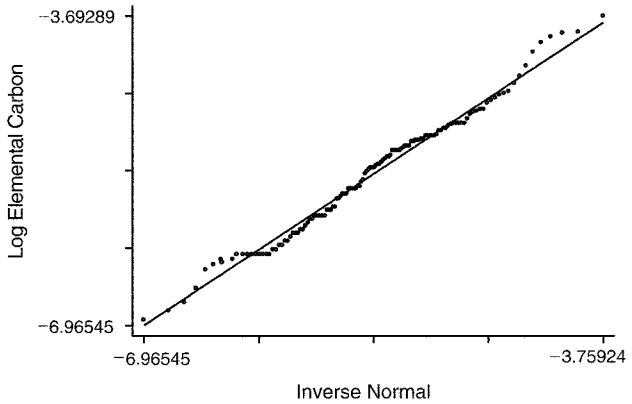


**Figure 3. Quantile distribution of logarithm of results from mechanical services EC sampling.**

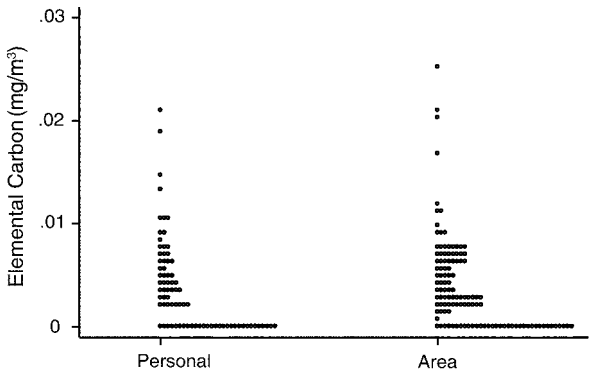


**Figure 4. EC sampling for mechanical services by sampling method (personal and area).** The samples that were below the detection limit are shown as 0.

***Area and Job*** Both personal samples and area samples were obtained. We determined the number of samples obtained by area as well as the number of samples below the detection limit (Table 6). Figure 6 shows a dot plot of the results of sampling for EC by area. Figure 7 shows a lognormal quantile plot labeled by area for the 3 areas with more than 10 samples each: diesel shop, heavy repair area, and turnaround area. We did a formal comparison of the distributions of samples for these 3 areas. The means were diesel shop, 0.0045 mg/m$^3$; heavy repair area, 0.0041 mg/m$^3$; and turnaround area, 0.006 mg/m$^3$. Although the mean result for the turnaround area was higher than the results for other areas, the differences were not statistically significant ($P = 0.6$).

The personal samples were classified by work area and by job. When personal samples were obtained, the job title of the worker wearing the sampler was identified, and we determined the distribution of jobs by area (Table 7). Figure 8 shows the distribution of EC sampling results by job title, and Figure 9 shows the distribution by job title and area sampled. There were no significant differences by job.

We compared the exposures of the laborers, hostlers, and machinists with the average of all other mechanical services jobs. After controlling for season, the average exposure of the laborers was significantly (50%) lower than that of all other jobs combined ($P = 0.02$). This was largely determined by the large proportion of samples below the detection limit for laborers, 7 of 13 (54%), compared with 23 of 65 (35%) for all other jobs. Neither the hostlers' nor the machinists' results differed from the average of all the other jobs combined.

**Table 6.** Number of Mechanical Services Samples by Area

| Area | Total Samples | BDL[a] |
|---|---|---|
| Back shop | 3 | 2 |
| Diesel shop | 33 | 13 |
| Fuel plant | 2 | 0 |
| Heavy repair | 41 | 14 |
| Office | 2 | 1 |
| Outside | 7 | 3 |
| Stores | 1 | 1 |
| Turnaround | 106 | 31 |

[a] Number of samples below the detection limit.

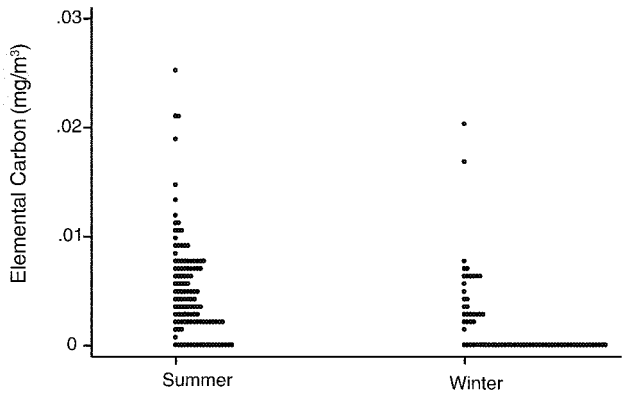


**Figure 5. EC sampling for mechanical services by season.**

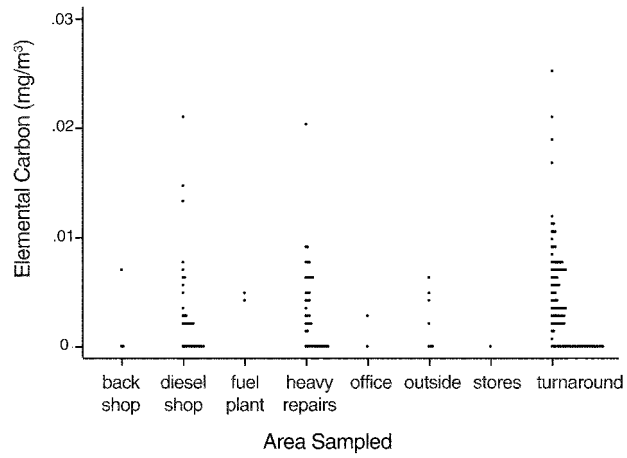


**Figure 6. EC sampling for mechanical services by area.**

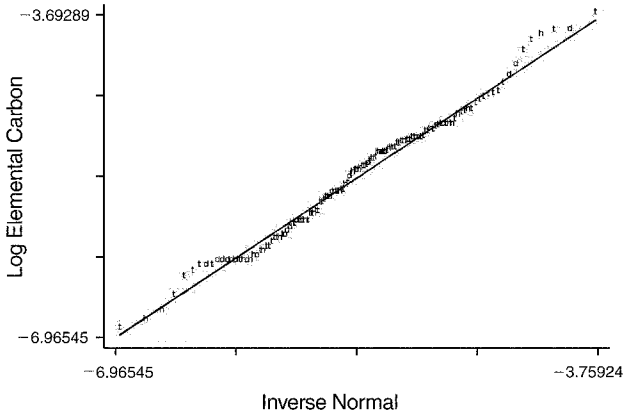


**Figure 7. Quantile distribution plot of logarithm of EC sampling for mechanical services labeled by area: d, diesel shop; h, heavy repair area; t, turnaround area.**

**Table 7.** Distribution of Mechanical Services Jobs by Area

| Area | Electrician | Hostler | Laborer | Machinist | Mechanic | Other | Pipefitter | Stores | Supervisor |
|---|---|---|---|---|---|---|---|---|---|
| Diesel shop | 2 | 3 | 0 | 3 | 0 | 5 | 1 | 0 | 1 |
| Heavy repair | 2 | 0 | 0 | 8 | 1 | 2 | 1 | 0 | 1 |
| Outside | 1 | 3 | 0 | 3 | 0 | 0 | 0 | 0 | 0 |
| Stores | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 1 | 0 |
| Turnaround | 1 | 3 | 13 | 15 | 0 | 2 | 0 | 0 | 6 |
| Total | 6 | 9 | 13 | 29 | 1 | 9 | 2 | 1 | 8 |

***Door Position*** Door position was noted to be closed ($N$ = 49; 25%), half-open ($N$ = 25; 13%), or open ($N$ = 51; 27%) at the time of sampling. There was no information about the door position for 68 (35%) of the samples. Door position was, as expected, strongly related to season. In the summer, the doors were closed during 25% of the sampling. In the winter, the doors were closed during 96% of the sampling. In a univariate analysis, the mean EC concentrations did not differ in relation to door position. When season was included in a multivariate model, concentrations were found to be 50% higher ($P < 0.001$) when the doors were closed.

***Multivariate Models*** The determinants of individual exposure might be expected to include such factors as season, door position, job, and area. The results of analysis of these factors taken one at a time were presented above.

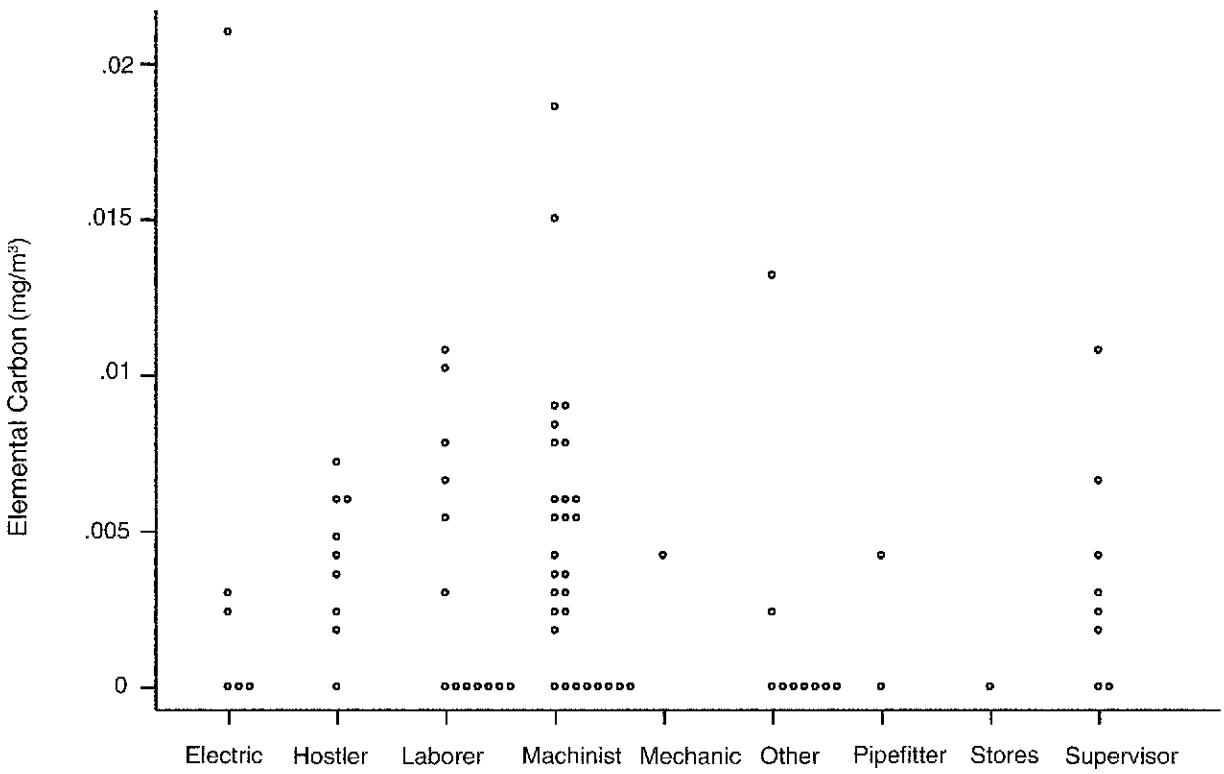


**Figure 8. Results of EC sampling for mechanical services by job title.**

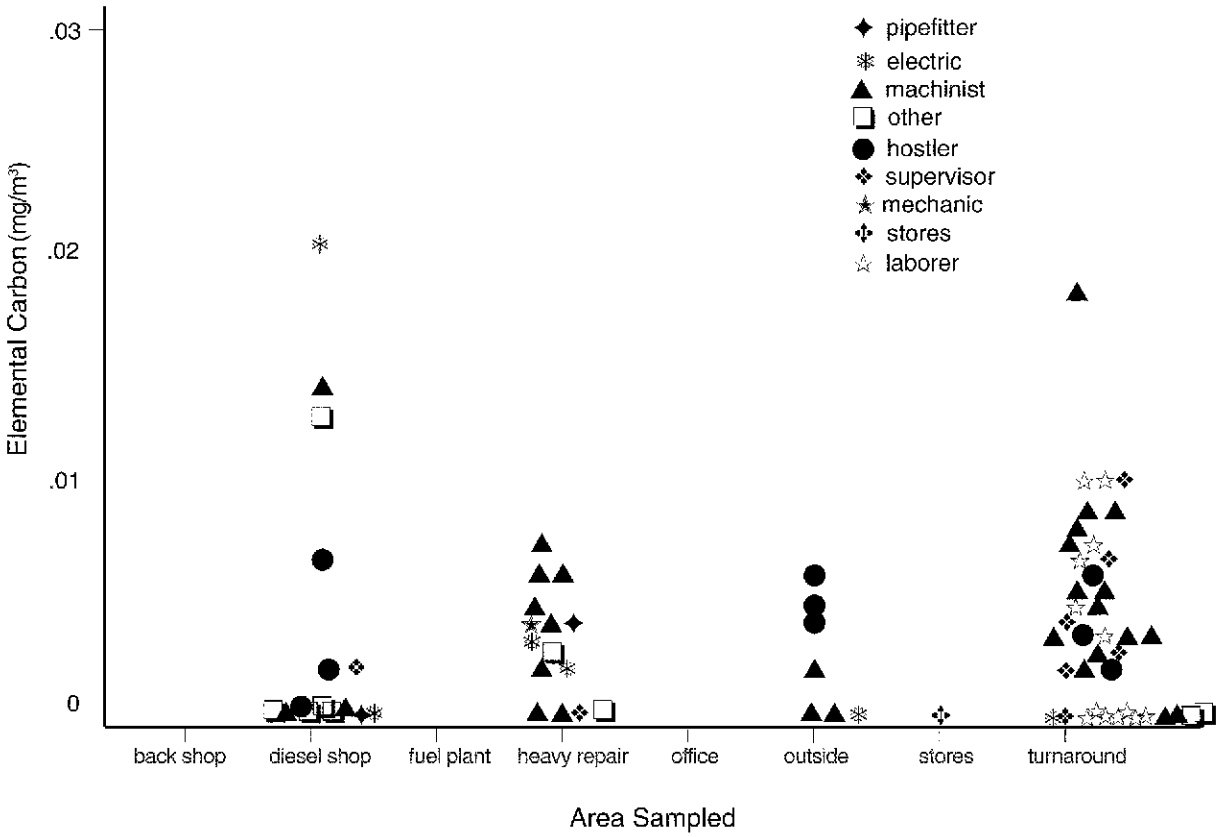


**Figure 9. Results of EC sampling for mechanical services by area sampled and job title.**

We used censored parametric analysis, with a lognormal distribution, to investigate multivariate models (Table 8).

***Conclusions About Exposures in Mechanical Services*** Pooling data we had collected and data collected by the companies resulted in a moderate amount of data for analysis. When looking at the individual job level and taking account of door position, however, we found the numbers available in the various cross-tabulations were often quite small.

We drew the following conclusions from the feasibility study. Season and door position in the shops appeared to be important in determining mean exposure level. No significant differences were found between area sampling and personal sampling or between sampling by work areas or by jobs. In assigning exposures for exposure-response calculations, we suggest that the exposure distributions can safely be presumed to be lognormal in mechanical services. Because there were no important differences between sampling by work areas or by jobs, one could assign a mean exposure to all workers with variability determined by the standard deviation of the lognormal distribution.

**Sampling Results for Transportation Services**

We combined the results of samples obtained during the feasibility study with those obtained previously in surveys by McMaster University and the companies, resulting in 70 transportation services samples for analysis. Of the 70 samples, 26 (37%) were below the detection limit of the analytical method.

***Statistical Distribution of Sample Results*** In a lognormal quantile plot of the EC sampling results for transportation services (Figure 10), exposures were highest in the trailing units. In a censored parametric analysis, with a lognormal probability distribution (Figure 11), the exposures in the trailing units were significantly higher than those in the lead units or in samples representing exposures in both positions. The distribution of residuals from the model showed good model specification (Figure 12). We determined the maximum likelihood estimates of the EC exposure concentrations by position on the train (Table 9) and the distribution of exposures in the lead locomotive units (Figure 13).

**Table 8.** Multivariate Models for EC Exposure in Mechanical Services

| Model | Result | Comment |
|---|---|---|
| 1. Season | Significant ($P < 0.0001$) | EC was lower in winter |
| 2. Season + indoors | Indoors/outdoors not significant ($P = 0.4$) | |
| 3. Season + door position | Door position significant ($P < 0.01$) | EC was higher with doors closed |
| 4. Season + door position + indoors/outdoors | Door position remains significant; indoors/outdoors not significant | |
| 5. Season + door position + work area | Season and door position significant; work area not significant | Exposures did not vary by work area |
| 6a. Season + door position + laborer | Season and door position significant; laborer not significantly different from other jobs | Exposures did not vary by job title |
| 6b. Season + door position + machinist | Season and door position significant; machinist not significantly different from other jobs | Exposures did not vary by job title |
| 6c. Season + door position + hostler | Season and door position significant; hostler not significantly different from other jobs | Exposures did not vary by job title |

In a censored parametric analysis, with a lognormal distribution, exposure did not differ significantly by season. In a censored parametric analysis with lognormal distribution designed to compare current and historical locomotives, exposure did not differ significantly in relation to locomotive model. Other analyses showed that exposures were lower with the windows closed, but the difference was not statistically significant ($P = 0.5$).

Train crews worked in cabooses until about 1990. In order to allow us to collect some information about exposures in a caboose, CP, in July 1999, attached a caboose to a freight train traveling between Calgary in Alberta and Revelstoke in British Columbia, on the far side of the Rogers Pass. The outbound route thus included a major climb through the Rocky Mountains as well as a long climb through the 11-km Connaught Tunnel. We obtained samples on both the outbound and return trips. On the outbound trip, exposures to EC in the caboose exceeded those in the lead locomotive

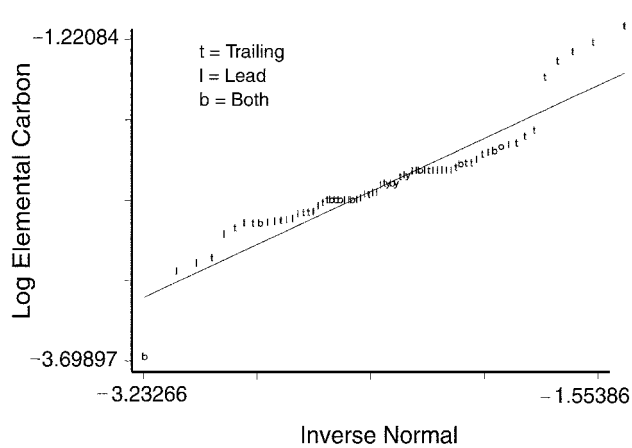


**Figure 10. Quantile distribution of logarithm of EC sampling from transportation services labeled by position on locomotive.**

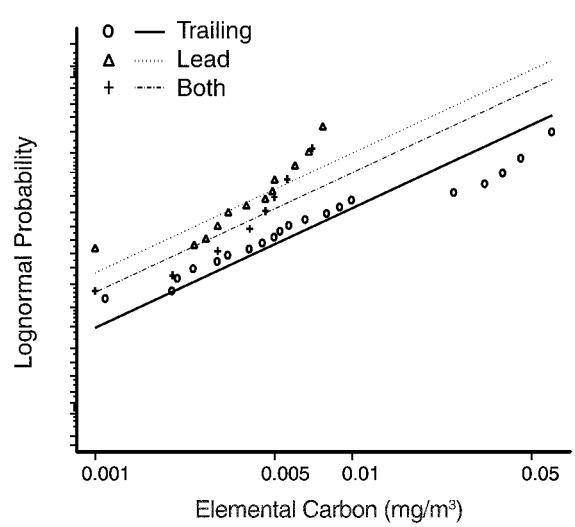


**Figure 11. Lognormal probability plot showing results of censored parametric model with maximum likelihood estimates for EC exposure versus position on train.**

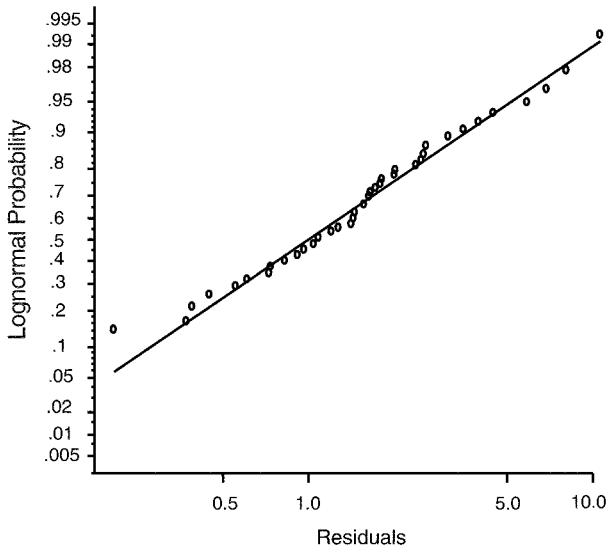


**Figure 12. Lognormal probability plot showing distribution of maximum likelihood estimate residuals from censored parametric model in Figure 11.**

**Table 9.** Maximum Likelihood Estimation of EC Concentrations in Transportation Services Locomotives

| Position | Samples (#) | Geometric Mean | Geometric SD | Arithmetic Mean |
|---|---|---|---|---|
| Both lead and trailing | 10 | – 5.87 | 0.68 | 0.0035 |
| Lead locomotives | 31 | – 6.12 | 0.79 | 0.0031 |
| Trailing locomotives | 24 | – 5.26 | 1.29 | 0.012 |

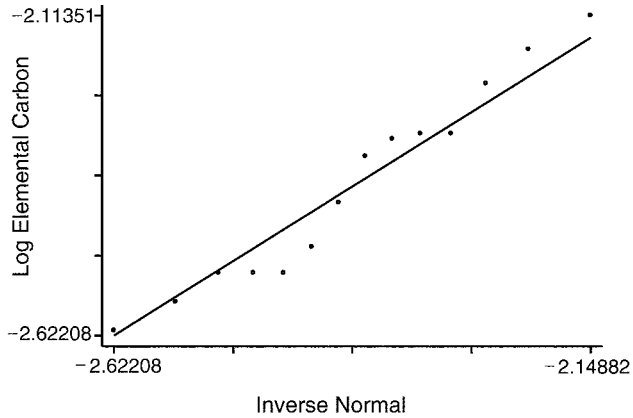


**Figure 13. Quantile distribution of the logarithm of EC sampling results in lead locomotives.**

and were comparable to those in the trailing unit (Figure 14A). This was largely attributable to a rise of at least 50-fold in EC exposure during the slow uphill climb in the Connaught Tunnel. On the return trip back to Calgary (Figure 14B), exposures in the caboose were much lower; however, it rained during the return trip, and some fumes may have been washed out before reaching the caboose.

***Conclusions About Exposures in Transportation Services*** By pooling data we had collected and data collected by the companies, we obtained a moderate amount of data for analysis. We drew the following conclusions from the feasibility study. The only significant determinant of exposure in the locomotives was position. In the current cab-forward configuration, the operators sit in front of the exhaust stacks. In earlier decades, exposures were almost certainly higher because the exhaust stacks were in front of the cab. We were unable to detect a significant influence of locomotive model, season, or window position on exposure. In assigning exposures for exposure-response calculations, we suggest that after accounting for

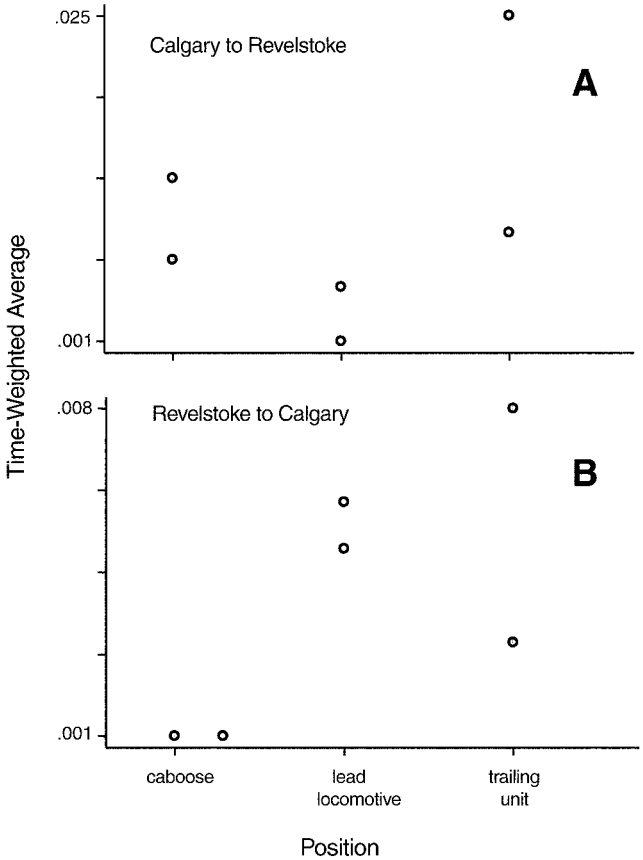


**Figure 14. Time-weighted average EC exposures from Calgary to Revelstoke and return from Revelstoke to Calgary, by position on train.**

locomotive position, the EC exposure distributions can safely be assumed to be lognormal. One could thus assign a mean exposure to train crews, irrespective of locomotive model, with variability determined by the standard deviation of the lognormal distribution. Past exposures in cabooses were probably low during routine operating conditions. This could be confirmed by further sampling in nonmountainous regions.

### Comparison of Mechanical Services and Transportation Services Exposures

In a censored parametric analysis, with a lognormal distribution, exposures in mechanical services were significantly higher ($P = 0.049$) than exposures in the lead units of locomotives and significantly lower than exposures in the trailing units of locomotives ($P = 0.004$; see Figures 15 and 16 and Table 10).

### Interlaboratory Comparisons

Measurements of the paired samples by CANMET and Datachem Laboratories were compared (Table 11). The filters contained very small weights of EC that were close to the analytical limit of detection; however, in general, the weights reported by the laboratories were quite similar.

### Duplicate Analyses

We performed duplicate analyses on 7 samples by analyzing 2 different wedges from the same filter. The results were similar (Table 12); however, the filters contained very small weights of EC that were close to the analytical limit of detection.

## EPIDEMIOLOGY SAMPLE SIZE

In order to assess statistical power for an epidemiologic study, we computed the number of lung cancer deaths that would be expected in the CN cohort if mortality rates were the same as those of the population of the province of

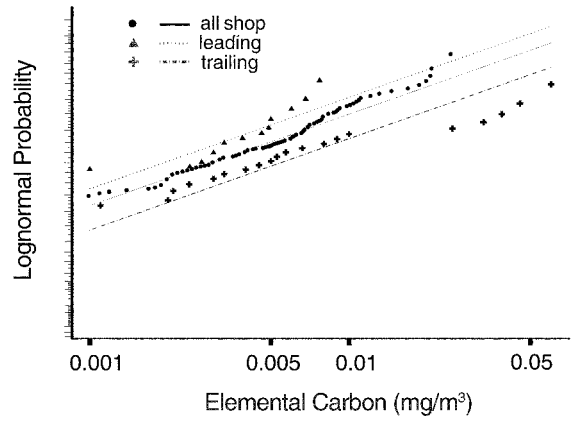


**Figure 15. Lognormal probability plot with maximum likelihood estimates of EC exposures in mechanical services (all shops) and in transportation services by position on train (lead or trailing locomotive).**

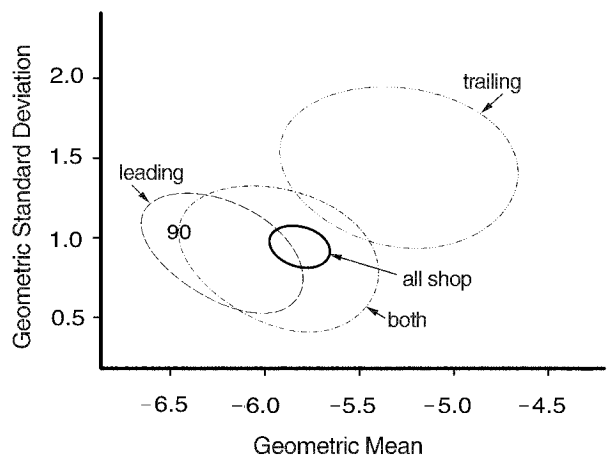


**Figure 16. The 90% confidence regions for means and standard deviations of EC exposure concentrations in transportation services by position on train (lead locomotive, trailing unit, or both) and in mechanical services (all shops).**

**Table 10.** EC Exposures in Transportation Services and Mechanical Services

| Location Sampled | Samples (#) | Geometric Mean | Geometric SD | Arithmetic Mean |
|---|---|---|---|---|
| **Transportation Services** | | | | |
| Lead locomotives | 31 | − 6.12 | 0.79 | 0.003 |
| Trailing locomotives | 24 | − 5.26 | 1.29 | 0.012 |
| **Mechanical Services** | | | | |
| Diesel shop, area samples | 84 | − 5.76 | 0.87 | 0.0046 |
| Diesel shop, personal samples | 30 | − 5.97 | 1.17 | 0.005 |
| Turnaround area | 105 | − 5.87 | 1.23 | 0.006 |
| Diesel shop and heavy repair area | 73 | − 5.96 | 1 | 0.0043 |

**Table 11.** EC Weights Reported by CANMET and Datachem Laboratories for 10 Paired Samples

| EC Weight (mg) CANMET | EC Weight (mg) DataChem Laboratories |
|---|---|
| 0.0018[a] | 0.0013[a] |
| 0.0018[a] | 0.0022 |
| 0.0028 | 0.0033 |
| 0.0015[a] | 0.0013[a] |
| 0.0036 | 0.0027 |
| 0.0044 | 0.0045 |
| 0.0022 | 0.0017 |
| 0.0025 | 0.0013[a] |
| 0.0003[a] | 0.0017 |
| 0.0014[a] | 0.0013[a] |

[a] Number given here is the detection limit; sample reported was below the detection limit.

**Table 12.** EC Weights Reported for Two Wedges from the Same Filter

| EC Weight (mg) Wedge 1 | EC Weight (mg) Wedge 2 |
|---|---|
| 0.0022 | 0.0013[a] |
| 0.0028 | 0.0033 |
| 0.0017 | 0.0014 |
| 0.001[a] | 0.0037 |
| 0.0065 | 0.0053 |
| 0.001[a] | 0.0045 |
| 0.0093 | 0.0082 |

[a] Number given here is the detection limit; sample reported was below the detection limit.

Ontario. We used the MANYRS computer program (Coleman et al 1986) to calculate expected mortality from all causes and from lung cancer in 5-year age and calendar intervals (Table 13). Since no employees were known to have died, the program provided expected mortality with the full cohort entering each 5-year calendar interval. We thus adjusted the population size for intercurrent mortality by subtracting expected all-cause mortality in prior calendar intervals at entry to each subsequent 5-year calendar interval. There were 415 expected lung cancer deaths among mechanical services and transportation services employees. Previous studies of diesel-exposed employees

**Table 13.** Mortality Predicted in CN Cohort from 1983 to 1999 Based on Ontario Mortality Rates

| Division | Total Deaths | Lung Cancer Deaths |
|---|---|---|
| Engineering services | 1,100 | 135 |
| Mechanical services | 2,100 | 255 |
| Transportation services | 1,300 | 160 |

had found relative risks of about 1.3. We might thus expect to see 540 lung cancer deaths, rather than the 415 estimated. In that event, for the standardized mortality ratio of 1.3, the 95% confidence interval would be 1.19 to 1.41.

We thus concluded that a full study would have adequate power to detect a standard mortality ratio of 1.3 in the CN cohort alone. Since we expect the CP cohort to be similar in size and demographics, we anticipate the study would have adequate statistical power to detect a lung cancer risk increased by 15% to 20%.

## DISCUSSION

Canada's 2 major railways, CN and CP, agreed to participate in the feasibility study. The focus of the study was to gather employee data, historical information, and information on workplace diesel exhaust exposures. The results of the study, in terms of its specific objectives, are summarized below.

### COHORT COMPOSITION AND DEMOGRAPHICS

We planned to evaluate the availability of records to determine the numbers of current, retired, and separated employees; the distributions of age, hire date, termination date, and length of service; and the number of employees known to be deceased (useful information for calculations of statistical power).

We obtained from CN a data file containing a list of some 74,000 employees on the payroll on or after 1983. The file included a chronological list of all jobs held at the company. Information about the vital status of former employees was not available in these files. Because of corporate restructuring, CP could provide only a roster of current employees, and their job histories, in time for preparation of this report. By comparing the rosters of current employees, and taking account of the corporate histories, we concluded

that the populations contributed to a full study by each company would be roughly similar.

## SMOKING INFORMATION

Cigarette smoking is a major confounder in the assessment of occupational lung cancer risk. We determined that smoking data were recorded in the employee medical files. These could be accessed in the case-control phase of a full study.

## JOB HISTORIES, JOB DICTIONARY, AND JOB CLASSIFICATIONS

In order to assess health risk in relation to exposure, it is necessary to estimate the exposures of individual workers. This requires job information for individuals. We obtained a chronological job listing for each employee and compiled a job dictionary. In collaboration with knowledgeable personnel, we combined jobs with similar exposure profiles into job groups. For CN, we organized 4,451 job titles into 37 homogeneous job groups. These homogeneous job groups were further amalgamated into 5 groups of jobs with similar exposures. (Appendix D, available on request, shows the classification scheme.)

## ESTIMATION OF NUMBERS OF EXPOSED EMPLOYEES

Combining information about exposure categories with the sampling information from the individual work records, we estimated the numbers of diesel-exposed employees in the various job categories. For CN, we estimated 13,647 exposed employees in engineering services, 20,302 in mechanical services, and 12,402 in transportation services. We anticipate that the numbers of exposed employees in these divisions would be similar in the CP cohort.

## IDENTIFICATION OF INFORMATION ABOUT HISTORICAL EXPOSURES

In order to allow for the prolonged period (latency) between exposure and disease, a full study of diesel exhaust exposure and lung cancer risk would be a retrospective cohort study. It would thus be necessary to estimate the magnitude of previous exposures. We collected and examined copies of industrial hygiene reports pertaining to diesel exhaust. We determined that reliable quantitative exposure data useful for a full study go back only to 1990. Other reports are available (for CN only) going back to 1956. These reports provide general information about historical conditions. We obtained other information useful for reconstructing historical exposures from locomotive inventories, shop diagrams with ventilation layout, and interviews with employees. Other information exists in libraries and archives. We determined that historical equipment with ages varying from 10 to 40 years was available for testing and that records with respect to other toxic substances were also available.

## EXPOSURE ASSESSMENT PROGRAM

One of the aims of the exposure assessment program was to identify current exposure levels and to assess variability across job classifications. A preliminary survey already completed at CN (Verma et al 1999) provided information about current exposure to diesel emissions for a few occupations in terms of EC, respirable dust, and respirable combustible dust. We planned to extend the survey at CN to include a wider representation of jobs and, in addition, to conduct a survey at CP, the other participant in the study.

In the previous hygiene survey at CN (Verma et al 1999), we used the Sunset Laboratory in Oregon to analyze the EC samples. The Government of Canada's CANMET laboratory in Sudbury, Ontario, also developed the capability of EC analysis and is a participant in the EC analysis round-robin quality assurance program. We planned to send 10 duplicate samples to each laboratory for a comparison of analytic results.

We obtained personal and area measurements for diesel exhaust, including 155 EC samples (71 for trains, 76 for diesel repair, and 8 for track repair) under different work practice and ventilation conditions. These samples were combined with 166 previously obtained EC results and compared with other types of samples. These other samples included data from historical industrial hygiene reports. Those reports had measurements of $NO_x$, CO, $SO_2$, coefficient of haze, benzene solubles, hydrocarbons, aldehydes, total dust, and respirable dust. We found that none of these historical measures correlated well with the newer method of measuring EC. We determined that due to different dust exposure levels between major trade groups, respirable dust might not be an appropriate measure of exhaust. Ten EC samples were taken in duplicate and sent to CANMET in Ontario, and DataChem Laboratories in Utah, for comparison. We found that these analyses produced comparable results.

The data could generally be described by lognormal distributions. In mechanical services, there were no significant differences in the exposure profiles of different work areas or occupations. In transportation services, exposures were significantly higher in the trailing locomotives than in the lead locomotives.

The exposures could be ranked as follows: lead locomotives were less than shops, which were in turn less than

trailing locomotives. Exposures in cabooses, in the absence of travel through tunnels, were found to be low.

### EXPOSURE RECONSTRUCTION

In the absence of adequate historical sampling results, it is desirable to attempt to re-create historic exposure conditions by, for example, running old equipment or reconstructing previous ventilation conditions in maintenance shops. We wanted to explore the availability of historic equipment and records of workplace design and work practices.

Many of the major shops at CN and CP are no longer used, but some of the old shops remain in use. Local and general exhaust ventilation at these shops has not changed significantly since the 1960s (the period we are interested in), and exposure monitoring of current conditions provides a reasonable starting point for the estimation of historical conditions. Work practices have changed and so has the volume of work done in the shops. We believe that it will be possible to model historic exposures by taking account of documented ventilation and work volume changes.

We learned that Canadian short line railroads acquired many pieces of equipment no longer used by CP or CN. We obtained the cooperation of several small railroads to carry out exposure reconstruction tests on their equipment. Samples were thus obtained on equipment no longer used by the study railroads. These data can be used to estimate historical exposures of CN and CP employees.

## CONCLUSIONS

We conclude that an epidemiologic study of exposure-response relations for lung cancer in the Canadian railroad industry is feasible. Canada's 2 major railroads indicated their willingness to participate and graciously cooperated with this feasibility study. Reasonable estimates can be made of current and historical exposure conditions and employee-specific estimates of cumulative exposure can be computed. The population size would be adequate to detect relative risks increased by 15% to 20%.

It is noteworthy that the exposure levels we measured were quite low, of the same order of magnitude as exposures in the general environment. Study of this population should thus inform the debate about acceptable levels of exposure for the general population.

## ACKNOWLEDGMENTS

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## APPENDICES AVAILABLE ON REQUEST

The following appendices are available by contacting the Health Effects Institute. Please give the author name, full title and number of the report, and the titles of appendices you request. The appendices available on request are:

Appendix A, Questionnaire

Appendix B, Annual Locomotive Inventories

Appendix C, Sample Index of Diesel Shop Blueprints

Appendix D, Job and Exposure Classification Scheme

Appendix E, Photographs of the Railroad Work Environment

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## OTHER PUBLICATIONS RESULTING FROM THIS RESEARCH

Finkelstein MM, Verma D. 2001. Exposure estimation in the presence of nondetectable values: Another look. Am Ind Hyg Assoc J 62:195–198.

## ABBREVIATIONS AND OTHER TERMS

| | |
|---|---|
| AAR | Association of American Railroads |
| ACGIH | American Conference of Governmental Industrial Hygienists |
| AIHA | American Industrial Hygiene Association |
| BDL | below the detection limit |
| CN | Canadian National Railways |
| CO | carbon monoxide |
| CP | Canadian Pacific Railways |
| EC | elemental carbon |
| GE | General Electric |
| GM | General Motors |
| IARC | International Agency for Research on Cancer |
| MLW | Montreal Locomotive Works |
| NIOSH | National Institute for Occupational Safety and Health |
| NO | nitric oxide |
| $NO_2$ | nitrogen dioxide |
| $NO_x$ | oxides of nitrogen |
| OC | organic carbon |
| PAH | polycyclic aromatic hydrocarbon |
| $SO_2$ | sulfur dioxide |
| TLV | threshold limit value |
| TWA | time-weighted average |

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# Investigators' Report

# Quantitative Assessment of Lung Cancer Risk from Diesel Exhaust Exposure in the US Trucking Industry: A Feasibility Study

Eric Garshick, Thomas J Smith, and Francine Laden

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# Quantitative Assessment of Lung Cancer Risk from Diesel Exhaust Exposure in the US Trucking Industry: A Feasibility Study

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## ABSTRACT

The objectives of this study were to test the feasibility of identifying a population exposed to diesel exhaust in which small to moderate excesses in lung cancer could be estimated with reasonable precision and to develop a strategy to provide quantitative estimates of current and past exposures. We chose to assess the feasibility of designing an epidemiologic study based in the US trucking industry.

With cooperation of the Motor Freight Carriers Association (the trucking industry trade association) and the International Brotherhood of Teamsters (Teamsters union), 4 large unionized national trucking companies agreed to participate in the feasibility study. We obtained samples of personnel, payroll, and truck inventory records and interviewed long-term employees, record managers, and senior management. The types of retirement records available from 2 large Teamsters union pension funds were determined. A pilot questionnaire was mailed to 526 employees at one terminal to obtain information on smoking behavior and job history. Short-term variations in exposure were assessed by measurement of air quality in truck cabs, loading docks, and yards in 2 large urban terminals and 4 small rural terminals. Measurements included elemental carbon (EC*) and organic carbon (OC) particles 2.5 µm or smaller in diameter, and respirable particulate clusters 2.5 µm or smaller in aerodynamic diameter ($PM_{2.5}$). The OC collected in high-volume area samples was further analyzed to assess the extent to which particles collected in the loading dock area came from diesel vehicles. Past studies and outside exposure databases were reviewed.

Major determinants of exposure included an individual's job title, terminal size, and terminal location. A gradient of exposure was identified. Smoking behavior did not differ between long-haul drivers and other workers. In 1985, the number of male union workers at the 4 companies whose job history could be characterized was 55,750, and in 1999 it was 72,666. A retrospective cohort study of workers from the cooperating trucking companies and the Teamsters union alive in 1985 with mortality assessed through 2000 would have a greater than an 80% power to detect a relative risk of lung cancer of 1.25 to 1.29 attributable to diesel exposure. Thus, epidemiologic studies can be designed to study the occurrence of lung cancer and to estimate past exposures to diesel exhaust among employees of the trucking industry.

## INTRODUCTION

Diesel exhaust contains fine particles with mutagenic and carcinogenic organic compounds, including polycyclic aromatic hydrocarbons coated on an EC core. These particles aggregate into respirable clusters (2.5 µm or smaller in diameter) and are readily inhaled deep into the lung. In 1989, the International Agency for Research on Cancer (IARC) classified diesel exhaust as a probable human lung carcinogen (IARC 1989). Epidemiologic evidence for carcinogenicity in humans was considered to be limited. Although diesel exhaust causes lung cancer in rats, this response has been found to be species-specific, attributable to a nonspecific effect of inhaled fine particles in rats, and is not considered relevant to assessment of diesel exhaust as a human carcinogen. Although more than 30 occupational epidemiologic studies have been conducted (Cohen and Higgins 1995; Bhatia et al 1998), inadequacies in study design, especially the exposure assessment, have prevented the conclusion that exposure to diesel exhaust definitely results in lung cancer.

Until recently, it has not been possible to find a large cohort with more than 20 years of exposure to diesel

* A list of abbreviations and other terms appears at the end of this Investigators' Report.

This Investigators' Report is one part of Health Effects Institute Special Report *Research Directions to Improve Estimates of Human Exposure and Risk from Diesel Exhaust*, which also includes a report by the HEI Diesel Epidemiology Working Group, four other Investigators' Reports, and an Executive Summary. Correspondence concerning this Investigators' Report should be addressed to Dr Eric Garshick, VA Boston Healthcare System, VAMC 1400, West Roxbury MA 02132.

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exhaust, and sufficient duration to allow for the latency associated with the development of lung cancer. Studies conducted in the 1980s, particularly in US railroad and trucking industry workers, did not include large numbers of subjects meeting this criterion because diesel engines had not been widely introduced 20 years before the studies were conducted. Another weakness of past studies is the lack of exposure measurements and meaningful quantitative estimates of previous exposure for workers. These measurements are necessary to establish that workers identified by general job title were actually exposed to diesel particles, to validate exposure classifications, and to estimate the quantitative relationship between exposure and lung cancer. There have not been lung cancer mortality studies of workers exposed to diesel exhaust in which personal exposure estimates were based on profiles drawn from records detailing each subject's job title and work location and linked to the results of representative sampling across the relevant industry and to historical reconstruction of exposure.

With the support of the 4 largest unionized trucking companies in the United States, which currently employ more than 72,000 unionized workers, the Motor Freight Carriers Association, and the Teamsters union, we assessed the feasibility of designing an epidemiologic and exposure assessment study to address the deficiencies of earlier studies of lung cancer risk in the trucking industry. Through visits to the 4 companies and communication with the 2 largest Teamsters pension funds, the Central States Southeast and Southwest Areas Pension Fund and the Western Conference of Teamsters Pension Trust, we assessed the quality of records of personnel, equipment, payroll, operations, and retirement in order to reconstruct job and exposure histories. Exposure was assessed at 2 large terminals (more than 500 workers each) in an urban area (Atlanta GA) and at 4 small terminals (fewer than 30 workers each) in rural New England. Our survey indicated that the range of exposure in these workers included the levels commonly observed in the general population; therefore, the workers' lung cancer risk attributable to diesel exhaust exposure would be relevant to the general population risk from diesel emissions and other fine particulate air pollution.

In the trucking industry, 3 primary work sites are associated with exposure to diesel exhaust: the vehicle cab, the terminal dock area, and the shop (Figure 1). In all 3 areas, both emission sources within the area and emissions that infiltrate from the outside are important contributors to exposure. Local air pollution and traffic emissions may contribute to exposures in the mobile vehicle cab and in the fixed work sites. Emissions from trucks bringing freight into and out of the terminal yard may contribute to the indoor levels of the dock area and the shop. The degree to which each set of sources contributes to indoor exposures depends on the size and location of the terminal and shop. A small terminal in a rural or semirural area will have low air pollution and limited vehicle traffic near and within its yard. A large terminal in an urban area can have high background air pollution and heavy vehicle traffic near and within its yard. The review of terminal size and location showed that there are both large and small terminals in urban and rural areas. There is also some evidence that emissions from a truck's engine may enter its cab (Zaebst 1989a,b,c,d,e,f; Zaebst et al 1990). Thus the vehicle operator could be exposed both to traffic emissions and to leaks from the engine compartment. All of the job category and work location factors must be considered when assessing current and past exposures.

The overall goal of this study was to determine the feasibility of designing an epidemiologic study to assess lung cancer risk from long-term exposure to diesel exhaust for

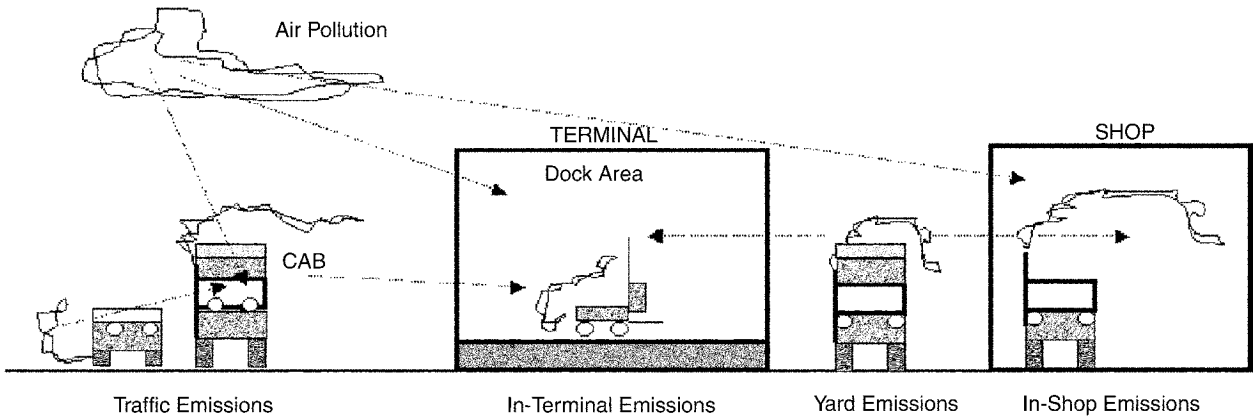


**Figure 1. Emission sources of worker exposures in the trucking industry.** (The depictions of emissions as smoke in the diagram are exaggerated.)

the purposes of hazard identification and risk assessment based on exposure estimates. In this case, feasibility is determined by availability of a sufficiently large cohort with long-term exposures to diesel exhaust in a range of intensities and by the likelihood that these exposures can be quantified with suitable accuracy and precision. To allow for the latency of lung cancer, the cohort must have exposures of more than 20 years' duration.

## SPECIFIC AIMS

The specific aims of this feasibility study were to

- identify a study population exposed to diesel emissions suitable for making a reasonably precise estimate of a small to moderate excess risk of lung cancer attributable to exposure; and
- develop an exposure assessment strategy to provide quantitative estimates of current and past exposure to diesel exhaust for trucking industry workers and validate these measurement techniques.

## METHODS

### PERSONNEL DATABASE ASSESSMENT

#### Company-Held Work History Records

Through visits for 1 to 2 days with senior management, personnel managers, and record managers at the 4 trucking companies, the extent of computerized records and the type and quality of information available were assessed. Samples of computerized personnel records were obtained, and the availability of paper records was determined. The availability of payroll records also was investigated. Company officials and long-term employees were interviewed both by telephone and in person to gain an understanding of the relationship between job title and job duties, and of historical changes in job-specific tasks and how they varied by company, terminal location, and terminal size. After each visit, study staff maintained regular communication with company personnel to obtain the information reviewed in this report.

#### Teamsters Union Employment Records

Although smaller pension funds serve Teamsters in the eastern part of the United States, the Midwest and West are served by 2 large plans. The Central States Southeast and Southwest Areas Pension Fund, headquartered in Chicago, Illinois, serves 34 states, and the Western Conference of Teamsters Pension Trust, headquartered in Edmonds, Washington, serves 13 states. We visited the headquarters of the Central States Pension Fund, met with its administrator and data management personnel, and assessed the availability and quality of work history information. Samples and descriptions of retirement records were obtained and reviewed. Discussions regarding the availability of information from the Western Conference Pension Trust were conducted by telephone, mail, and e-mail.

### COMPANY DATA ON TRUCK FLEETS

The availability of computerized and paper-based truck inventory and maintenance records at each company was ascertained from data managers and truck operations personnel, and samples of these records were reviewed. Company officials were asked to provide dates when each company first used diesel vehicles, when diesel pick-up and delivery (P&D) trucks were introduced, and when diesel forklifts were used. An overview of current and historical vehicle purchasing and retirement policies was also obtained to gauge the differences in fleet ages and characteristics between companies. The vehicle-use policy of each company was used to assess the likelihood of older vehicles staying in service, the vehicle mix, and the location of older trucks for possible emission assessment. The following books were reviewed for information on historical fleet characteristics and vehicle-use practices, and for confirmation of dates of diesel vehicle use obtained from interviews: *The Roadway Story* (Cantelon and Durr 1996); *Yellow in Motion: A History of Yellow Freight System, Incorporated,* 2nd edition (Filgas 1971), revised edition (Filgas and Waters 1987); and *Never Stand Still: The History of Consolidated Freightways, Inc and CNF Transportation, Inc 1929-2000* (Durr and Cantelon 1999).

### COMPANY DATA ON TERMINALS

Current addresses of terminals and information about terminal design and operations, including information on diesel fuel use and on freight loading and unloading in the dock area, were obtained from all companies. The size of each terminal was determined from the terminal codes in the personnel files, and terminals were grouped according to the number of workers assigned to them (fewer than 30, 30 to 99, 100 to 199, and 200 or more workers). ArcView (Environmental Systems Research Institute 1996), a geography-based information system software, was used to overlay the terminal addresses on a US Census Bureau map with locations designated as urban or rural. This information was then used to determine the current distribution of terminals and personnel in urban and rural areas.

## SMOKING AND WORK HISTORY INFORMATION AND FEASIBILITY OF COLLECTION BY QUESTIONNAIRE

The availability of information regarding cigarette smoking history in trucking company workers was investigated, as well as the feasibility of conducting a survey of current and former workers by mail. A pilot mail survey was sent to all 526 workers at a large trucking terminal in Atlanta, Georgia. Employees were initially sent a cover letter and a 4-page detailed questionnaire including questions on date of birth, height and weight, educational status, job title, smoking history, duration of work in the trucking industry and as a Teamster, and a job matrix covering lifetime employment history. Nonrespondents received a second copy of this questionnaire with a cover letter. The third mailing to nonrespondents consisted of a cover letter and an abbreviated questionnaire ("1-page questionnaire") with less detailed smoking questions and no job matrix. (The questionnaires are available on request as Appendix B.)

## HISTORICAL DATABASES ON TRUCK FLEETS, TRAFFIC, AND AIR POLLUTION

The availability of current and historical databases containing information on factors that might contribute to diesel exhaust exposure was investigated. The feasibility and limitations of use were determined for each database to identify those that could aid in the development of exposure models.

### Truck Registration and Use Data

Commercial and governmental databases containing information on vehicle registration and truck use were identified and reviewed. We looked specifically for data to characterize each company's vehicle fleet, the distribution of trucks nationally, and factors that might influence emissions and exposure variables.

### Traffic Volume and Road Use

Databases maintained by the Federal Highway Administration and the Bureau of Transportation Statistics, both in the US Department of Transportation (DOT), were identified and reviewed to determine the nature of current and historical information on highway traffic counts and road use by different types of vehicles.

### Truck Engine Specifications and Emission Factors

The availability of engine certification data was assessed by contacting engine manufacturers through the Engine Manufacturers Association. The US Environmental Protection Agency (EPA) National Vehicle Fuel Emissions Laboratory also was contacted. The type and quality of vehicle and engine design specification information available from manufacturers was evaluated by review of independent publications.

### Background Air Pollution

The EPA and the individual states maintain an extensive monitoring network for air pollutants across the country, and these data are available through the Aerometric Information Retrieval System (AIRS), a public-access database (http://www.epa.gov/airs/). The availability of this database and its suitability to provide historical information and variation in background particulate air pollution were reviewed. Since climatic factors such as temperature, precipitation, and wind direction can strongly influence exposure, the availability of current and historical meteorologic information from the National Oceanic and Atmospheric Administration (NOAA) National Climate Data Center (http://www.ncdc.noaa.gov/) also was assessed.

## EXPOSURE ASSESSMENT

One of the key challenges to exposure assessment is the choice of exposure markers. In this study our goal was to identify a marker that not only represents exposure to diesel exhaust, but also serves as an index of carcinogens in diesel exhaust, (ie, quantitatively related to the cancer risk). This second dimension is important because all compounds in diesel emissions are probably not carcinogenic and the carcinogenicity of the total particle may correlate poorly with the carcinogenic fraction.

A chemical mass balance method has been used to determine the relative contributions of specific emission sources to the total mass in atmospheric particulate samples (Cass et al 1984; Waalkes and Ward 1994; Schauer et al 1996; Watson et al 1998). In these tests, ambient airborne EC appeared to be most closely related to diesel emissions (Cass et al 1984; Cass and Gray 1995; Birch and Cary 1996). However, ambient air exposures are complex, with many sources of air contaminants, including EC. Source apportionment based on details from the chemical mass balance method makes it possible to determine how much EC is coming from diesel engines. For example, diesel vehicles emit large amounts of EC, only small amounts of cholestane and related compounds, and no levoglucosan (a plant sugar), whereas gasoline vehicles emit relatively large amounts of cholestane compounds, little EC, and no levoglucosan; softwood combustion releases relatively large amounts of levoglucosan and moderate amounts of EC and cholestane. An air sample showing large amounts of EC, small amounts of cholestane and related compounds, and small amounts of levoglucosan would implicate diesel

emissions as the main contributors with small amounts of gasoline vehicle emissions and softwood smoke included.

On the basis of source apportionment findings and exposure data from other investigators, we chose EC as our primary marker of diesel emissions, and OC and $PM_{2.5}$ as markers to track exposures to other general air contaminants. The quality of EC as a diesel marker in occupational settings varies because proximity to the source is an important determinant of exposure intensity. For workers whose jobs bring them close to operating diesels, diesel exhaust is a major component of the air contaminants so EC would be a good marker of exposure. For workers with no nearby diesel sources, EC may be a poor marker because more than half of the EC in the ambient air may come from other combustion sources such as home heating or wood fires (Cass et al 1984; Waalkes and Ward 1994; Watson et al 1998). Combining source apportionment with use of EC as a marker compensates for the lack of a unique marker of diesel emissions.

Independent of its application as a marker for the quantity of diesel emissions in the air, the suitability of EC as a marker for human carcinogens in diesel exhaust is difficult to assess. It is not clear whether EC itself is a human carcinogen, or the organic compounds in diesel exhaust are the primary carcinogens, or EC and OC together are the agents of cancer risk. No carcinogenic pollutant is unique to diesel emissions; rather, all of the common combustion sources overlap with diesel emissions. The relative concentrations vary, but virtually all are present.

Some of the goals of the feasibility study were to determine the applicability of potential sampling methods, to verify that personal exposure monitoring is feasible, and to obtain limited data on current exposures. We used real-time and integrative sampling to obtain personal, area, and source data for $PM_{2.5}$. Particulate samples were analyzed to measure concentrations of EC and OC.

### Exposure Assessment Methods

Personal exposure to $PM_{2.5}$ was measured by filter sampling using the Personal Environmental Monitor (PEM) (SKC, Eighty Four PA), which has a small impactor and 37-mm Teflon filter with a pore diameter of 0.2 µm. Mass collected on filters was determined by gravimetric analysis using an analytic balance (Micro-Gravimetric M5, Mettler Instruments Corp, Hightstown NJ) and Pallflex Teflon 37-mm fiber filters (Pall Corp, East Hills NY), and TissueQuartz 25-mm fiber filters (URG, Chapel Hill NC). The filters were weighed after humidity equilibrium was attained (after at least 24 hours) in a chamber. At the end of sampling, the filter was taken back to the laboratory. After humidity equilibrium was attained, the filter was reweighed to determine weight gain. A second set of $PM_{2.5}$ samples was collected using a similar sampler with a TissueQuartz filter. These samples were analyzed for EC and OC using the National Institute for Occupational Safety and Health (NIOSH) 5040 thermo-optical method (Birch and Cary 1996; Cassinelli and O'Connor 1998) at the laboratory of Dr James Schauer, University of Wisconsin, Madison.

Real-time measurements of $PM_{2.5}$ were made with the DustTrak (TSI, Shoreview MN) using laser light scattering to detect airborne particles after passage through an impactor that removes particles larger than 2.5 µm in diameter. Concurrently with the DustTrak monitor, a Q-Trak (TSI) was used to obtain real-time data on carbon monoxide (CO), carbon dioxide ($CO_2$), temperature, and relative humidity. The Q-Trak uses a set of physicochemical sensors. The sampler inlet was placed on the back of the seat, at shoulder level, next to the driver, so the sample represented the air quality in the center of the cab.

Large-volume integrative samples for detailed chemical analysis were collected using a 47-mm TissueQuartz filter sampler with a large cyclone separator (less than 2.5-µm-diameter cutoff) operated at 16.7 L/min for 8 to 12 hours. Gas chromatography/mass spectroscopy (GC/MS) was used by Dr Schauer's laboratory to analyze these samples for a wide range of specific organic compounds (Schauer et al 1996). This analysis involved extensive sample preparation, including spiking the filters with 7 deuterated internal recovery standards, and 5 extraction steps. Extracts were combined and reduced in volume, and finally half of the combined extract was derivatized with diazomethane to esterify organic acids. Both the derivatized and underivatized extracts were analyzed on a GC/MS (GC model 5890 and MS model 5972, Hewlett-Packard), using a capillary column 30 m × 0.25 mm in diameter (HP-1701). More than 100 compounds are quantified with a relative error of 20%. The following chemical compound groups are evaluated: *n*-alkanes, polycyclic aromatic hydrocarbons, substituted phenols, guaiacol and related compounds, syringol and related compounds, *n*-alkanoic acids, *n*-alkenoic acids, alkane dicarboxylic acids, aromatic carboxylic acids, resin acids, levoglucosan and other sugars, and other organic compounds. The cost and complexity of the analysis limit its application to composited samples from work locations where the types of sources are likely to be approximately constant.

### Exposure Measurement Strategy

Testing was conducted at 2 large terminals in the Atlanta area during October 1999. In total, 45 $PM_{2.5}$ personal samples (with EC and OC measurements) and

14 DustTrak personal samples were collected for forklift operators, mechanics, hostlers, long-haul drivers, local (P&D) drivers, and dockworkers directly involved in daily terminal activity. For area samples, 90 $PM_{2.5}$ (with EC and OC), 7 source apportionment (later composited for analysis), and 58 DustTrak samples were collected from the loading dock, shop, fueling lane, and clerk's office. An upwind background sample, called the "yard sample," was also collected at each site. Sampling methods were compared by hanging several different samplers side-by-side in different areas. Concurrent with the gravimetric monitoring, real-time monitoring of airborne particles was done with a DustTrak to obtain 1-minute averages of light-scattering respirable $PM_{2.5}$. One-minute averages of CO, $CO_2$, temperature, and relative humidity were also measured with the Q-Trak. Exposures also were assessed at 4 small rural terminals in New England by sampling in the dock area and upwind in the yard in April to May 2000 with the collection of 18 $PM_{2.5}$, 14 EC and OC, and 16 DustTrak area samples.

The vast majority of personal samples and area samples were collected over 8 to 12 hours. A limited number of side-by-side personal samples also were collected by equipping a worker with 2 personal samplers to assess the relationship between URG personal measurements of EC and OC and PEM gravimetric measurements for personal sampling. The area samples were taken at similar locations in the terminal area, 5 feet from the deck level, with 4 different collecting devices (size-selective samplers): DustTrak, Q-Trak, personal sampler, and PEM.

### Previous NIOSH Studies

In 1988 and 1989 NIOSH sponsored Health Hazard Evaluation studies by Zaebst and coworkers to characterize exposures to diesel emissions in the trucking industry (summarized in Zaebst et al 1990). A total of 327 personal and area samples were collected in 7 large truck freight terminals. These data represent a critical historical database on conditions in the late 1980s. The individual data points given in the report appendices are an important resource. We also used the NIOSH studies to help assess other sources of exposure variability so that we could identify current and historical factors that will define differences in exposure across the industry.

## STATISTICAL ANALYSIS

### Smoking and Work History Questionnaire

Chi-square tests were performed to compare questionnaire respondents and nonrespondents and various parameters describing smoking behavior by job title. Means were compared using *t* tests. Logistic regression models were used to identify predictors of current-smoking or ever-smoking status. Linear regression methods were used to assess predictors of average cigarette consumption.

### Exposure Assessment

Results of the environmental sampling surveys were expressed as geometric mean (GM) and geometric standard deviation (GSD) because the distribution of sampling results is approximately log-normal. The results of real-time sampling from the DustTrak (plots of $PM_{2.5}$ level versus time) are also presented for various locations. Analysis of variance was used to compare between-terminal variations. Although some of the limited data collected in this study was evaluated statistically, the focus was on the feasibility of monitoring exposure. The scope of the project was limited by the sponsor. As a result, the analysis of measurement error is based primarily on published data and NIOSH Health Hazard Evaluation Reports.

# RESULTS

## PERSONNEL

### Company Databases

***Job Activities*** The companies invited to participate in the feasibility study are the 4 largest national, unionized, less-than-truckload freight carriers in the United States. These companies have provided shipping services for large packages and freight to industrial, commercial, and residential customers since the 1920s and 1930s. They send a P&D driver to pick up the freight, which is often on pallets. The freight is returned to the local terminal, where it is consolidated onto a trailer with other loads bound for similar parts of the country. Next, one or more long-haul drivers drive the trailer to the regional hub terminal closest to the delivery point. The trailer load is broken down into smaller loads that are either delivered to their destinations or sent to smaller local terminals from which P&D drivers deliver them to their destinations.

The major duties in the trucking industry that involve possible exposure to diesel exhaust are grouped into job categories (Table 1). Because these companies are all unionized, job duties were the same across the 4 companies with only minor differences for each job title. Our discussions indicated that major job-specific duties had not changed significantly over time for most jobs. Union rules permit workers to bid on terminal-based jobs, which are

**Table 1.** Major Job Categories, Duties, and Locations in Unionized Trucking Companies

| Job Category | Job Duties | Job Location |
|---|---|---|
| Long-haul driver | Operate heavy-duty tractor-trailer trucks between cities | Highway truck cab |
| P&D driver | Drive tractors and smaller single-bodied trucks either within cities or in rural areas; pick up and deliver cargo between terminal docks and consumers | In and out of truck cab |
| Dockworker | Load and unload cargo; may operate forklifts | Loading dock |
| P&D/dockworker | Combination job: each day performs activities of either P&D driver or dockworker; combination job is more likely at smaller terminals | In and out of truck cab; loading dock |
| Mechanic | Repair and maintain tractors; job may include fueling | Truck repair shop |
| Hostler | Drive tractor or "yard tug" (small, specialized tractor units that do not comply with emissions standards), moving trailers to and from the freight dock | Terminal yard |
| Clerk | Category includes cashiers, dock clerks, dispatchers, customer service representatives, and others not regularly near diesel vehicles | Offices, occasionally on dock |

awarded on the basis of years of seniority working at a terminal. Therefore, movement between terminals is limited once workers are hired, and job categories tend to remain stable throughout each worker's employment tenure.

Mechanics experienced the greatest change in job duties over time. Whereas in the past they performed most major engine and body repair on company trucks in house, currently they are responsible primarily for preventive maintenance, such as oil and tire changes. The division between dockworkers and P&D drivers is not always sharp. Some workers perform both duties on a day-to-day basis, as needed, and have the job title P&D/dockworker or combination worker. These workers are usually stationed at smaller terminals, and their actual job duties depend on the requirements at the terminal on a given day.

***Work History*** Each company has a computerized personnel record system that contains, for each worker, name, social security number, race, sex, date of birth, date of hire, union membership status, current and previous job titles, terminal code (which can be linked to an address), and a record of when the person was actively working or on extended leave due to layoff or illness. The companies differ regarding the dates that records were computerized and the extent of the information available. Between 1979 and 1993, three of the companies computerized the complete job histories for all active employees. The company that computerized in 1993 also included the work histories for all workers employed on or after 1980 even if they were not employed when the database was established. The fourth company started its computerized database in 1972 and included all jobs held beginning in 1971. If an employee worked at another company acquired by one of these 4 companies, then the date of hire reflects that person's start date at the original company.

Additional information is available from each company in the form of paper or microfiche records. All 4 companies maintain noncomputerized personnel records for current employees and for other employees within 3 years of termination. The original job applications, which include previous job history outside the company, are also available for these employees. Detailed paper records and job applications are available only after 1995 for one of the companies because its warehouse was destroyed in a storm. However, employee identification cards with basic identity information, job title, terminal location, union seniority date, date hired, last day worked, and reason for termination were saved and can be used to fill in some missing information.

The availability of payroll records listing hours worked for each job assignment was also investigated. These data may be useful for determining distribution of time spent between dock work and P&D for combination jobs. Archived computerized payroll records are not available for 2 of the companies. One company computerized its payroll records starting in 1972, and the other has noncomputerized payroll records from 1979 to 1995 and computerized payroll information since 1995.

The existence of information regarding cigarette smoking was also explored. Although the DOT requires drivers to pass a physical examination every 2 years, information on cigarette smoking is not collected. Furthermore, the companies do not maintain medical or other health-related records that might contain smoking data.

Although the yearly listings of job and terminal location were available in the company personnel databases, the

companies did not have the in-house computer support to extract these data. Instead, 3 of the 4 companies provided computerized cross-sectional snapshots of the data for either December 31 or November 30 of 1985, 1990, 1994 or 1995 and for 1998 or 1999. Two of these companies also provided data for 1980, and data for additional dates were acquired from some companies. The fourth company provided a copy of its computerized personnel database of all employees on the payrolls in 1980 and every year subsequently. Only current (1999) or last job title (before retirement) and terminal location were included. Therefore, for this company the cross-sectional data were extracted and it was assumed that job title was stable throughout a worker's tenure. All identifiers were removed before we obtained these data.

***Computer Records*** Results of the computerized personnel record review were summarized for the years 1985, 1990, 1995, and 1999 (Table 2). Assuming conversion from gasoline-powered trucks to diesel-powered vehicles by 1985 (see Table 5), long-haul drivers with long-term employment in these companies would have had at least 20 years' experience of driving diesel trucks by the time of this study. Most workers in the industry are male and white although the proportion of nonwhite employees has been increasing gradually over time. In 1985, most of the union women (89.2%) had clerical jobs, thereby limiting our ability to include them in a meaningful fashion in an epidemiologic study of the health effects of diesel exhaust. The missing indicators for union status in Table 2 are due to omissions in the data provided by one of the companies.

**Table 2.** Summary of Computerized Personnel Records from Four Trucking Companies: Sample Size and Percentage of Total

| | 1985 | | 1990 | | 1995 | | 1999 | |
|---|---|---|---|---|---|---|---|---|
| Characteristic | *n* | % | *n* | % | *n* | % | *n* | % |
| **Total sample** | 65,391 | | 86,500 | | 97,532 | | 86,949 | |
| **Sex** | | | | | | | | |
| Male | 60,632 | 92.9 | 79,779 | 92.3 | 88,621 | 90.9 | 79,540 | 91.6 |
| Female | 4,655 | 7.1 | 6,618 | 7.7 | 8,822 | 9.1 | 7,304 | 8.4 |
| Missing | 104 | 0.2 | 103 | 0.1 | 89 | 0.1 | 105 | 0.1 |
| **Race** | | | | | | | | |
| African American | 4,154 | 6.4 | 7,586 | 8.8 | 10,483 | 10.8 | 10,054 | 11.6 |
| Asian | 55 | 0.1 | 237 | 0.3 | 457 | 0.5 | 428 | 0.5 |
| White | 52,991 | 81.2 | 68,012 | 78.8 | 80,480 | 82.6 | 70,738 | 81.4 |
| Hispanic | 2,208 | 3.4 | 2,963 | 3.4 | 5,285 | 5.4 | 5,004 | 5.8 |
| Native American | 168 | 0.3 | 236 | 0.3 | 360 | 0.4 | 290 | 0.3 |
| Unknown/Other | 5,709 | 8.7 | 7,284 | 8.5 | 348 | 0.4 | 306 | 0.3 |
| Missing | 106 | 0.2 | 182 | 0.2 | 119 | 0.1 | 129 | 0.1 |
| **Union status** | | | | | | | | |
| Union | 42,204 | 87.9 | 58,874 | 86.7 | 78,123 | 84.6 | 72,842 | 89.4 |
| Nonunion | 5,828 | 12.1 | 8,989 | 13.2 | 14,280 | 15.5 | 8,647 | 10.6 |
| Missing | 17,359 | 26.5 | 18,637 | 21.5 | 5,129 | 5.3 | 5,460 | 6.3 |
| **Age in years** | | | | | | | | |
| Mean ± SD[a] | 43.1 ± 10.1 | | 43.1 ± 10.5 | | 43.8 ± 10.6 | | 45.4 ± 10.2 | |
| Sample size | 60,227 | | 80,005 | | 96,952 | | 86,720 | |
| **Years worked** | | | | | | | | |
| Mean ± SD[b] | 10.0 ± 8.5 | | 8.8 ± 8.2 | | 9.5 ± 7.9 | | 10.6 ± 8.2 | |
| 75th percentile | 15.2 | | 13.7 | | 13.6 | | 14.8 | |
| Sample size | 60,634 | | 79,416 | | 88,185 | | 84,367 | |

[a] Age was calculated for workers with date-of-birth information.

[b] Years worked was calculated for workers with date-of-hire information.

The nonunion workers include management and casual labor, and most of the missing date-of-birth and date-of-hire information in the databases reflects the inclusion of casual labor in the data extraction. One of the companies was missing job titles for 70% of its employees in the 1985 data extraction, whereas in 1990 this information was missing for only 6% of its workforce. Because job titles are generally stable in the industry, the 1990 data were used to fill in the missing information for 1985. In summary, extensive computerized records are available from each company, including for workers with longtime employment in the trucking industry during the time diesel trucks were primarily used.

The numbers of male unionized workers were determined for the 4 companies and for specific job titles in 1985, 1990, 1995, and 1999 (Table 3). For the company missing union status indicators, all employees with these job titles were considered to be union members and thus were included in the totals. In 1985 the number of male workers categorized as likely belonging to the Teamsters union was 55,750, and this workforce increased in later years as the industry consolidated through acquisitions. The percentage of long-haul drivers remained relatively constant (between 24% and 27% of the workforce) between 1985 and 1999. The proportion of workers classified solely as P&D drivers fell slightly as the proportion of combination workers (P&D/dockworkers) increased. The proportion of mechanics also decreased, which was consistent with the practice of contracting the heavy repair work to outside vendors. Most of the workers listed under the job titles of management and trainee were from the company for which the union status indicator was missing and therefore may not have been union members. In other companies, some of the managers of smaller terminals and persons whose job title was listed as trainee were union members; for completeness, therefore, all workers with these job titles from this company were included in the totals. In any study using these data, the union membership of these workers could be verified by a more detailed search of each company's personnel records.

The age distribution, mean years worked, and the third quartile of the distribution of years worked were determined for all union male workers for whom date of birth was available in the 4 companies in 1985 and 1999 (Table 4). More than half of the entire unionized workforce and of the long-haul drivers were between the ages of 35 and 54 in both years. The age distribution indicates, however, that the midpoint was slightly older in 1999 than in 1985 and that the long-haul drivers tended to be older than the rest of the population. In both 1985 and 1999, nearly 50% of

**Table 3.** Male Union Workers Employed in Four Trucking Companies by Year and Job Title

| | 1985 | | 1990 | | 1995 | | 1999 | |
|---|---|---|---|---|---|---|---|---|
| Job Title/Company | *n* | % | *n* | % | *n* | % | *n* | % |
| **Total sample** | 55,750 | | 72,274 | | 77,009 | | 72,666 | |
| **Job title** | | | | | | | | |
| Long-haul driver | 15,190 | 27.3 | 18,447 | 25.5 | 20,840 | 27.1 | 17,409 | 24.0 |
| P&D driver | 9,595 | 17.2 | 13,924 | 19.3 | 11,458 | 14.9 | 9,876 | 13.6 |
| Dockworker | 8,887 | 15.9 | 15,662 | 21.7 | 16,482 | 21.4 | 16,699 | 23.0 |
| P&D/dockworker | 8,621 | 15.5 | 13,029 | 18.0 | 17,993 | 23.4 | 18,750 | 25.8 |
| Mechanic | 3,283 | 5.9 | 3,292 | 4.6 | 2,955 | 3.8 | 2,824 | 3.9 |
| Hostler | 1,415 | 2.5 | 2,029 | 2.8 | 2,224 | 2.9 | 2,361 | 3.3 |
| Clerk | 2,105 | 3.8 | 2,608 | 3.6 | 2,275 | 3.0 | 1,837 | 2.5 |
| Janitor | 200 | 0.4 | 243 | 0.3 | 200 | 0.3 | 191 | 0.3 |
| Management | 2,443 | 4.4 | 2,557 | 3.5 | 2,453 | 3.2 | 2,511 | 3.5 |
| Trainee | 177 | 0.3 | 222 | 0.3 | 111 | 0.1 | 203 | 0.3 |
| Others/Unknown | 3,834 | 6.9 | 261 | 0.4 | 18 | 0.02 | 5 | 0.01 |
| **Company** | | | | | | | | |
| A | 4,837 | 8.7 | 4,708 | 6.5 | 12,726 | 16.5 | 11,983 | 16.5 |
| B | 18,506 | 33.2 | 21,431 | 29.7 | 22,377 | 29.1 | 23,693 | 36.7 |
| C | 17,004 | 30.5 | 24,331 | 33.7 | 21,118 | 27.4 | 17,645 | 24.3 |
| D | 15,403 | 27.6 | 21,804 | 30.2 | 20,788 | 27.0 | 19,345 | 26.6 |

**Table 4.** Age Distribution of All Male Union Workers and Long-Haul Drivers Employed in 1985 and 1999 at Four Trucking Companies

| | All Male Union Workers | | | | Long-Haul Drivers | | | |
|---|---|---|---|---|---|---|---|---|
| | | | Years Worked | | | | Years Worked | |
| Age Group (yr) | *n* | % | Mean | 3rd Quartile[a] | *n* | % | Mean | 3rd Quartile[a] |
| **1985** | | | | | | | | |
| < 35 | 11,019 | 21.5 | 4.4 | 7.3 | 1,051 | 7.2 | 4.1 | 7.3 |
| 35–44 | 16,533 | 32.4 | 8.6 | 12.8 | 4,339 | 29.8 | 7.6 | 11.4 |
| 45–54 | 15,467 | 30.2 | 12.2 | 17.9 | 5,640 | 38.7 | 10.9 | 16.3 |
| 55–64 | 7,932 | 15.5 | 18.2 | 26.9 | 3,447 | 23.6 | 16.5 | 24.8 |
| ≥ 65 | 149 | 0.3 | 21.7 | 30.8 | 81 | 0.6 | 20.9 | 30.7 |
| Total[b] | 51,100 | | | | 14,558 | | | |
| **1999** | | | | | | | | |
| < 35 | 11,204 | 15.4 | 4.0 | 6.4 | 913 | 5.3 | 2.9 | 4.2 |
| 35–44 | 20,727 | 28.6 | 8.5 | 12.3 | 3,194 | 18.5 | 6.7 | 10.9 |
| 45–54 | 25,296 | 34.9 | 13.2 | 20.2 | 6,781 | 39.0 | 11.5 | 15.8 |
| 55–64 | 14,780 | 20.4 | 15.5 | 22.3 | 6,083 | 35.1 | 14.5 | 21.8 |
| ≥ 65 | 555 | 0.8 | 18.6 | 25.3 | 329 | 2.0 | 18.5 | 25.3 |
| Total[b] | 72,562 | | | | 17,300 | | | |

[a] Twenty-five percent of employees worked at least this many years.

[b] Total number of union employees for whom date of birth was available.

the workers (those 65 years of age of older), including the long-haul drivers, had more than 20 years of employment in their current company. The distributions of age and years worked in 1990 and 1995 were similar to the distributions in 1999 (data not shown).

### Teamsters Union Employment Records

Information on the numbers and vital status of relevant members of the Central States Pension Fund and the Western Conference Pension Trust was obtained. In January 1999, 183,661 retired Teamsters were receiving benefits from the Central States Pension Fund, and in November 1999 approximately 188,000 active members were contributing to the pension funds. The 4 companies had 36,010 employees contributing to this fund at this time (19.2% of fund membership), and the 21,889 former workers from these companies made up 11.9% of the fund retirees receiving benefits. In 1999, there were 7,113 deaths among all Central States Pension Fund members receiving benefits.

In 1999, employers were contributing to the Western Conference Pension Trust for 270,744 employees, with 14,228 members (5.3%) coming from the 4 large unionized carriers. Overall, 169,667 Teamster retirees were receiving benefits from the Western Conference Pension Trust, but the proportion of retirees from the 4 large unionized carriers was not obtained for this report. Therefore, of the 72,666 active Teamsters union members in the 4 companies in 1999, 49.6% belonged to Central States Pension Fund and 19.6% belonged to Western Conference Pension Trust, for a total of 69.2% of the workers currently contributing to one of the two large Teamsters union retirement plans. Approximately 25 small funds accounted for the other 30.9% of the workers' retirement benefits.

The 2 pension funds we studied maintain computerized records of the contributions made by each unionized trucking company on behalf of a worker, including dates and company name. In the Central States Pension Fund, the only specific job histories available are the self-reports provided by each worker when he or she completes an application at the time of retirement. The applicant lists job title and dates of service with each employer; this information is not verified against company records. In the Central States Pension Fund, starting in 1992, these records are available from an image retrieval system, and prior to 1992, on microfiche. No job history information is collected by the Western Conference Pension Trust. No other specific details about job title and location are maintained by either

fund, and medical information and information on personal habits, such as cigarette smoking, are not collected. Thus, although the pension funds cannot provide many details about a participant's job history or risk factors, they can provide information on total years of creditable service as a Teamster, beyond what is available from the current or last employer, and they provide an independent means of verifying data on work in the unionized trucking industry.

## COMPANY RECORDS ON TRUCK FLEETS

### Vehicle and Equipment Records

Each company has 2 computerized systems containing vehicle and equipment information: asset registers and maintenance records. The extent and availability of these records vary by company. Asset registers record the following information about every truck and forklift purchased or sold by the company: vehicle serial number, make, model, year, and engine type as well as location assignment for forklifts and P&D vehicles. The asset registers are computerized and available for all vehicles ever owned by one of the companies, from the 1980s for 2 companies, and for the last 5 years from the other. Additional information may be retrievable from paper records starting as early as 1959 in one company. Furthermore, long-time personnel in the asset and maintenance departments often are able to reconstruct much of this information from memory, because companies typically purchased only one or two types of trucks per year. The maintenance departments keep information on vehicle component changes, such as engine overhauls and air-conditioning removals. These data supplement those provided by the asset registers, and although some are computerized for some companies, they are available mainly in paper form.

An historical listing of the number and design of diesel P&D vehicles assigned to each terminal and the composition of the long-haul fleet can be constructed for each company. Long-haul vehicles are not assigned to a terminal and are used throughout a company's system. However, the long-haul drivers are assigned to a specific home terminal, which indicates the region where they drive.

From the preliminary information provided, we constructed a timeline for the conversion to diesel long-haul vehicles, P&D trucks, and forklifts (Table 5). Each company started using long-haul diesel trucks in the 1950s, and all 4 companies had converted from gasoline long-haul vehicles by 1965. Diesel vehicles were introduced into the P&D fleets beginning in 1972. Depending on the company, the year that the P&D fleet was 100% diesel varied considerably (1980 to 1992), but the majority of combined fleets were diesel by the late 1980s. Diesel forklifts were in general use by the mid 1980s.They were then phased out between 1990 and 1996 and were replaced with propane units as part of contract negotiation with the Teamsters union in the early 1990s. According to information from one company, diesel forklifts were most likely to be used in the large distribution centers but not in the smaller local terminals. Company

**Table 5.** Years of Introduction of Diesel Long-Haul Trucks, Pick-up and Delivery Trucks, and Forklifts at Four Trucking Companies

| | Year by Company | | | |
|---|---|---|---|---|
| Diesel Use | A | B | C | D |
| **Long-haul trucks** | | | | |
| First used | 1957 | 1952 | 1951 | 1955 |
| 100% use[a] | 1962 | 1954 | 1961 | 1965 |
| **P&D trucks** | | | | |
| First used | 1978 | 1977 | 1974 | 1972 |
| 100% use | 1987 | 1992 | 1980 | 1983 |
| **Forklifts** | | | | |
| First used | None used | 1979 | 1986 | 1982 |
| 100% use | | 1986 | 1986 | 1984 |
| Phase-out | | 1990 | 1991 | 1992 |
| Last used | | 1996 | 1994 | 1996 |

[a] Year by which all of the vehicles in the fleet were diesel.

records can be used to identify where diesel forklifts were used.

### Historical Changes in Vehicle Use

Vehicle replacement and utilization practices differ among the 4 companies. Three of the 4 companies have historically converted long-haul vehicles to P&D vehicles after 10 to 15 years of service and then used them for an additional 10 to 15 years. Each company keeps its vehicles in service for different lengths of time, and within each company this policy has changed historically. The trend is toward keeping the vehicles on the road longer. Because of these policies, the age of the fleets varies substantially between companies. For example, data from one company showed that several hundred trucks purchased in 1975 to 1979 are still in service, whereas another company keeps its vehicles for only 5 years.

Maintenance policies for all 4 companies generally call for restoring vehicles to original operating conditions rather than replacing the engines (although different engines may be installed in some of the older vehicles). Any replacement of engines is recorded and archived. The long-haul vehicles have been air conditioned for many years (for example, new vehicles purchased by one company have been air conditioned since approximately 1980). Since P&D vehicles are not air conditioned, the air conditioning is removed when a long-haul vehicle is converted to local use.

## TERMINAL CHARACTERISTICS

Through analysis of annual reports, personnel records, terminal operation records, and other company sources (such as maps and company histories), the total number of terminals and the size of each terminal at each of the 4 companies starting in 1985 were determined. Current terminal addresses were obtained from all companies and historical terminal addresses from one company. Samples of terminal blueprints illustrating design features were obtained from some of the companies. We have determined that it is feasible to obtain this information on all relevant terminals.

Terminal codes and the number of employees assigned to each terminal are available from the personnel databases. In 1985 there were 2,200 work locations in the personnel databases; the number increased to 2,427 by 1990. Since 1990, the trend has been toward a reduction in the number of smaller terminals, and the total number of work locations decreased to 1,770 in 1995 and 1,337 in 1999. However, because some of the work locations specified in the personnel files are at the same physical address or terminal (in other words, office areas and repair shops may have unique codes), the number of total terminals is overestimated. For example, there were only 1,267 unique terminals in the address files in 1999. Therefore, historical address information (available from all companies) should be used to determine unique terminal addresses in a larger study. Of the 1,267 unique addresses in 1999, most were terminals with fewer than 30 employees (Table 6). Because background air pollution (and respirable particle levels) vary with urban or rural location, the addresses of the 1999 terminals were overlaid on a map of the United States (Figure 2) and classified as urban or rural as defined by the US Census Bureau. Of the 1999 employees, 58.5% worked in rural locations, mainly at large terminals with 100 or more employees (Table 7). Examination of air pollution data, local truck traffic volume, and number of trucks assigned may result in finer gradations of exposure potential, which will permit better resolution of any exposure effects.

**Table 6.** Distribution in 1999 of 1,267 Total Terminals at Four Trucking Companies by Size and Number

| Employees per Terminal | Number of Terminals | Percentage of Total |
|---|---|---|
| <30 | 849 | 67.0 |
| 30–99 | 280 | 22.1 |
| 100–199 | 50 | 3.9 |
| 200+ | 88 | 6.9 |

An alternative source of terminal address data is the RJ Polk Company Truck Group (Southfield MI), which provides data on the number of trucks assigned to each terminal address and detailed information on each truck. These data indicated that terminals with few trucks also had small workforces in the company personnel databases. Of 1,232 registration addresses identified in September 1999 by RJ Polk Company, 626 terminals (50.8%) had 1 to 4 trucks assigned and 247 terminals (20.1%) had 5 to 9 trucks assigned.

Current records of diesel fuel use and of freight volume by terminal are available, but historical records are limited. Companywide summaries of fuel used are available for the last 3 to 10 years, depending on company policy, and are mostly in paper form. Terminal-specific information on the amount of freight moved across a dock is available for the last 3 to 12 years, and companywide summary data are available for the last 10 to 15 years, also in paper form.

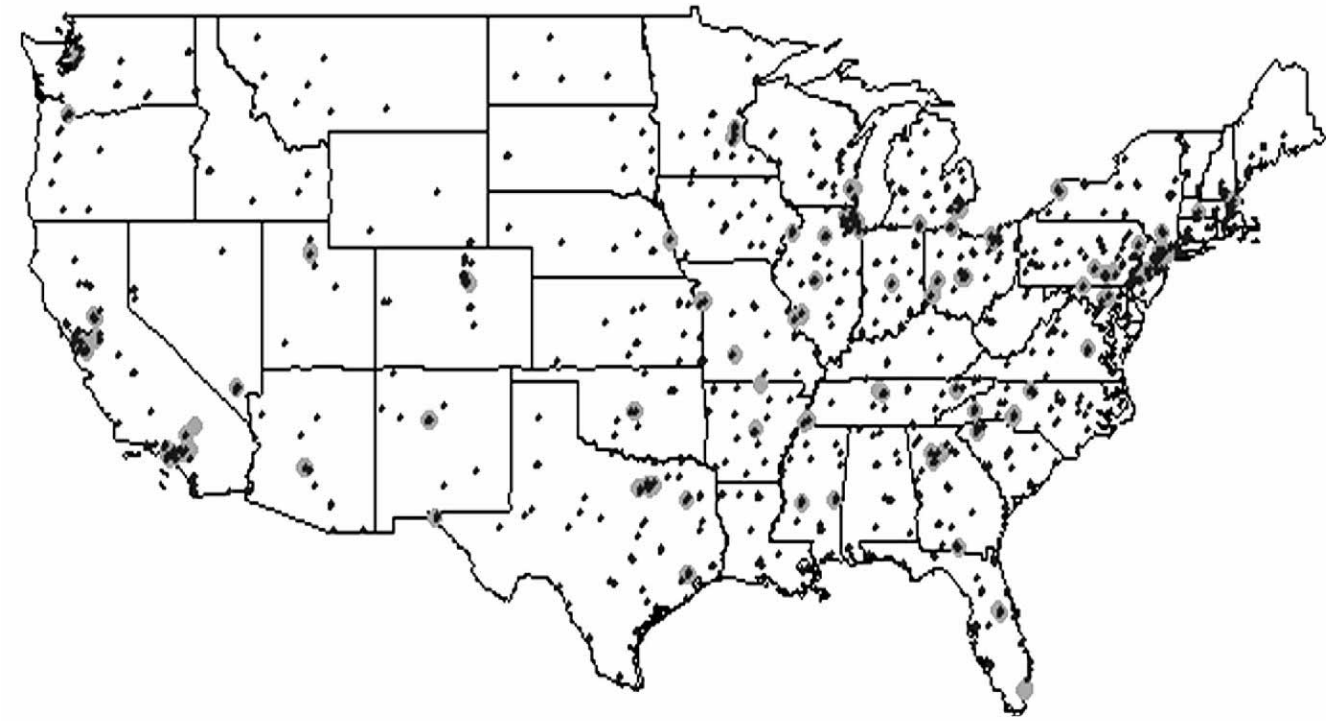

**Figure 2. Map of the United States showing size distribution and locations of the 1,267 unique terminal addresses identified for the 4 trucking companies in 1999.** Large dots are terminals with 100 or more workers and small dots are terminals with fewer than 100 workers.

## SMOKING AND WORK HISTORY QUESTIONNAIRE

### Response Rates

The smoking and work history questionnaire and the mailing protocol are described in the Methods section. The original sample comprised 526 subjects who worked at the same terminal. However, 2 workers had been terminated, and 14 had either missing or incorrect address information. Of the remaining 510 subjects (489 male, 21 female), 247 (48.4 %) responded after 3 mailings. In total, 175 of the 4-page questionnaires and 72 of the 1-page questionnaires were returned. The response rate among the 294 male white unionized workers was 49.7% (146 respondents). The 37.9% response rate among 145 unionized workers of other races was significantly less ($P = 0.02$). Based on the job title provided by the company, the response rate among 213 long-haul drivers, regardless of race, was 49.8% (Appendix A, Table A1). This rate was slightly higher than the rate among the other male unionized workers combined (42.0%) but not significantly so ($P = 0.10$). This slight difference in response rates for long-haul drivers persisted even when the rates were stratified by race. Subsequent analyses were based on the total population of 201 male white and nonwhite union workers who responded to the questionnaire. Data are presented in detail in the tables of Appendix A.

### Respondent Population Description

#### *Self-Reported Job Title Versus Company Job Title*

To assess the extent that the job title listed in company personnel records can be used to determine a worker's actual job title and duties, company job titles for these workers were compared with the job titles obtained by self-report on the mail survey (Table A2). Agreement between self-report job title and company job title for the long-haul drivers, mechanics, and hostlers was nearly perfect. The distinctions between the P&D drivers, the dockworkers, and the P&D/dockworkers were less clear. This company classifies nearly all of its P&D drivers as P&D/dockworkers. However, a P&D/dockworker might drive a P&D truck most of the time and thus call himself a P&D driver, or he might work predominantly on the dock and call himself a dockworker. If the exact job titles available for all workers are compared, then 79.6% of the self-reports agreed with the job title in the company records. If P&D driver, dockworker, and P&D/dockworkers are considered the same job, then there was 94% agreement.

**Table 7.** Distribution in 1999 of Workers at Four Trucking Companies by Terminal Location and Size

| | Terminals <100 Workers | | Terminals >100 Workers | | Total | |
|---|---|---|---|---|---|---|
| Location | *n* | % | *n* | % | *n* | % |
| Urban | 8,911 | 12.3 | 21,274 | 29.3 | 30,185 | 41.5 |
| Rural | 15,438 | 21.3 | 27,043 | 37.2 | 42,481 | 58.5 |
| Total | 24,349 | 33.5 | 48,317 | 66.5 | 72,666 | |

***Personal Characteristics*** The mean age of the male union respondents was 51.8 ± 9.6 years. Characteristics of the long-haul drivers were compared with those of other workers combined (Table A3). The long-haul drivers were significantly older than the other workers ($P < 0.0001$), which is consistent with the pattern observed in the entire workforce. Fewer long-haul drivers had educational training beyond high school ($P = 0.07$). Body mass index, a possible indicator of dietary and exercise habits, was similar across job titles in this population.

### Smoking History

Smoking behavior was examined by self-reported job title (Table A3). Of all respondents, 33 (16.4%) were current smokers, 93 (46.3%) were former smokers, and 75 (37.3%) were never smokers. The prevalence of smoking (current or ever) and the average number of cigarettes smoked per day over time were generally similar across union job categories although long-haul drivers tended to smoke more (Table A3). The numbers of workers in the job categories other than long-haul drivers were too small to allow further analyses. When the job categories were grouped into long-haul drivers versus other jobs, these variables were not significantly different (Table A4). The long-haul drivers had also smoked longer than had other workers ($P = 0.0004$). However, they were also the oldest and thus had had more opportunity to smoke. This difference was no longer statistically significant after duration of smoking was adjusted for age (adjusted mean was 24.2 years of smoking for long-haul drivers and 19.8 years for all other workers, $P = 0.13$). In multivariate logistic regression models that included age, educational status, and body mass index, the self-reported response to "long-haul driver (yes/no)" was not a significant predictor of ever or current smoking ($P = 0.59$ and 0.86, respectively). Age, educational status, and body mass index for all workers were also not significant predictors of smoking behavior in the multivariate model ($P > 0.20$) and were similar between smokers and nonsmokers when examined independently (Table A6). Overall, these pilot data suggest that smoking behavior is similar between the long-haul drivers and other workers, and among the other workers, and smoking behavior is independent of job title.

### Work History

Information regarding job history was obtained in 2 different ways on the questionnaire: simple direct questions and a detailed job matrix. Workers were asked what year they started working in the trucking industry, what year they joined the Teamsters union, and their current job title. Calculation of years working for the current company was based on the date of hire provided by the company. Respondents also completed a job matrix listing their past job titles, whether the job was full-time or part-time, dates of service, company name, and major job duties. In analyzing the job matrix data, we assumed that a part-time job occupied 50% of a worker's time on the job during the indicated time period.

Comparisons between number of years worked in the trucking industry, years as a Teamster, and years worked for the current company were based on the 125 male union workers for whom complete information was available. On average, workers started in the trucking industry in their mid to late 20s (Table A5), so at the time of the survey these workers had worked in the industry for more than 20 years. Approximately 4 to 5 fewer years were spent as a member of the Teamsters union, and even fewer years were spent working in their current company (Table A5). This difference in number of years worked as a Teamster and for the current company is probably a function of the terminal and company that was selected for this survey. The company had greatly expanded in recent years, and before 1995 the terminal belonged to an acquired unionized company. Almost half of the workers at this terminal had worked for the previous company, and we were able to account for this in our analyses. In this terminal, 41.8% of the workers had been employed for less than 10 years and only 15.9% had been employed for more than 20 years. This is a younger age distribution than that of the rest of the company. This company had only recently started to hire large numbers of new workers as it expanded.

Only 43 (34.4%) of these 125 workers completed the job matrix in sufficient detail to allow for meaningful analysis. Among the 20 long-haul drivers in this subgroup, 25.4 years (± 8.9 SD) of their entire job history in trucking

was spent as a long-haul driver; among 16 P&D/dockworkers, 22.1 years (± 8.0 SD) of their entire job history was in similar jobs in the trucking industry; and among 6 mechanics, 27.7 years (± 6.9 SD) of their entire work history was spent as a mechanic. Therefore, when considered together, these workers had spent many years employed in the trucking industry, mostly as a Teamster in the same or a similar job.

## HISTORICAL DATABASES ON TRUCK FLEETS, TRAFFIC, AND AIR POLLUTION

Many government departments and agencies maintain current and historical databases on factors that might contribute to the development of exposure models. Information that could be utilized for this purpose is also available from commercial sources. The scope and quality of information in these databases vary widely, however, and many of them are inappropriate for use in epidemiologic studies. We have included information on selected large databases as well as their level of appropriateness for use in epidemiologic studies.

### Truck Registration and Use Data

The most comprehensive external truck registration database was the RJ Polk Company Truck Group, which offers the list for purchase. This database provides a detailed national listing of new truck registrations by company and address starting in the early 1990s. For several states, registration address cannot be provided because of state regulations. Truck model, cab specifications (eg, cab over engine or engine in front of cab), engine type and model, and detailed engine specifications are available. The RJ Polk Company also maintains a database updated quarterly starting in the early 1990s that provides the same information on all trucks in service based on registration address and company name. Several other companies and organizations, including the American Trucking Association, Transportation Technical Services (Fredericksburg VA), and Trinc Transportation Consultants (Washington DC), publish yearly aggregated data for all trucking companies, including total number of trucks.

The Truck Inventory and Use Survey database was considered to be a potential general source of information to profile trucks in service in the United States. Others have used this database to profile the operational characteristics of the US truck fleet. First developed in 1963 by the Department of Commerce, the database has been updated every 5 years. A survey mailed to a random sample of private and commercial truck owners is used to collect data on a single truck. Owners are asked to estimate the number of miles traveled by the vehicle, the numbers of trailers usually hauled, and the type and weight of a load. This database contains company-based or terminal-specific truck data, but it does not have information about the types of trucks used by each company because information is collected only on one truck in a fleet. Therefore, the data reported do not reflect the distribution of trucks operated by each company, and how accurately these data represent long-haul and P&D trucks driven by unionized trucking company workers is uncertain.

### Traffic Volume and Road Use

The DOT maintains information on traffic counts on all of the federal and statewide highways starting in 1970. Summary information is also available for some urban and rural main roads. This database includes traffic volume (in vehicle miles traveled) by vehicle type and class (car, motorcycle, truck, or bus), classification of interstate (urban or rural), vehicle weight, and average daily traffic count. These variables are available on a monthly or yearly basis. Information on traffic volume on smaller highways and local roads is available only at the state or county level, and availability and amount of information varies by county and state.

### Information on Air Quality

The EPA and the individual states maintain an extensive monitoring network for various air pollutants across the country, and these data are available through AIRS, a public-access database (http://www.epa.gov/airs). The pollutants monitored include total suspended particles, particulate matter less than or equal to 10 µm diameter ($PM_{10}$), and $PM_{2.5}$, depending on the site. The national database was started in 1983, but measurements of $PM_{2.5}$ have only recently been added. Some states and cities had monitoring data as early as the late 1960s, and this information was included in AIRS. These data can be used to assess exposure to ambient air pollution and to understand the changes in magnitude of pollution over time. The location of each monitoring station is given by latitude and longitude and can be linked to other geography-based information such as terminal address. The limited quality of information for earlier periods must be considered in any study.

Climatic factors such as temperature can strongly influence exposure. Information on climate can be obtained from the NOAA National Climate Data Center. This database includes hourly, daily, monthly, and yearly information on temperature and barometric pressure. Available in computerized form from 1961 forward, this resource can be linked to the trucking terminals using the latitude and longitude of the monitoring stations.

### Truck Engine Certification Data and Emission Factors

The EPA certification program for heavy-duty engines tests diesel engines by using an engine dynamometer. Engine testing started in 1971, but particulate matter emissions were not regularly assessed until 1988, the year that a particulate matter standard went into effect for heavy-duty diesel engines. Manufacturers perform these tests on their engines and submit the data to EPA, which may independently conduct further testing. Diesel engine manufacturers were contacted via the Engine Manufacturer's Association and asked to provide engine certification data for selected vehicles used by several of the companies in the past. Beginning in 1988, the emissions clearly met the required standard for each year. The data available before 1988 were minimal. The extent of these data was not specifically ascertained because the exposure models developed for this proposal were not based on engine certification data (see later section on exposure model development). The relevance of engine certification data for predicting in-use emissions has not been established because the test process may not represent actual operating conditions.

Chassis dynamometer testing may also be used to obtain data on emissions directly from heavy-duty diesel vehicles using simulated driving conditions. Limited testing has been done in the past because truck emission standards have not been based on these test results. Chassis dynamometer particulate matter test results for heavy-duty vehicles (trucks and buses) have been summarized by Yanowitz and coworkers (2000) using data available starting in 1975. This review showed a general reduction in particulate matter emissions since 1975. For emissions in a given year, variability among engines was considerable. Because many preregulation vehicles are in operation in the participating companies, a statistical model can be developed to test whether truck model year can be used to estimate personal exposure to diesel exhaust.

The Diesel Truck Index (Truck Index, Santa Ana CA) database was used to assess the availability of data on design specifications for heavy-duty truck and diesel engines. This paper-based yearly manual also provides detailed information on truck model, cab design, engine model, and various engineering specifications for trucks from many manufacturers starting in 1980. Current data are also available from the engine and truck manufacturers. Engine manufacturers are also able to provide model and engineering specifications for diesel engines manufactured before 1980.

Historical specification data on vehicles and engines are available from the library of the American Truck Historical Society (2000) in Birmingham, Alabama. This library contains over 1,000 books, 25,000 pieces of vintage truck sales literature, over 30,000 vintage truck magazines, and 5,000 owner, repair, and sales manuals and is available to members of the society. This society, with over 21,000 members, is dedicated to preserving the history of companies and individuals involved in the trucking industry. It sponsors yearly meetings and truck shows at which members display trucks that are more than 25 years old. These gatherings of several hundred old vehicles could also provide an opportunity to obtain chassis dynamometer emissions data on older vehicles to compare with current vehicles.

## EXPOSURE ASSESSMENT

### Current Exposure Assessment

Field tests were conducted to evaluate various sampling methods, to verify that personal exposure monitoring was feasible, and to obtain some limited data on current exposures to diesel exhaust. Real-time and integrative sampling were used for personal and area monitoring of $PM_{2.5}$. Particulate samples were analyzed to measure concentrations of EC and OC. Trucking industry and terminal operations were reviewed to identify the possible sources of diesel emissions and sources of EC, OC, and $PM_{2.5}$, the markers selected as indices of exposure. See the Methods section for further discussion of these markers.

Field tests, including both personal and fixed location sampling, were conducted for 1 week in Atlanta in 2 large urban terminals operated by 2 of the participating companies. Fixed location sampling was also conducted for several days in 4 small rural terminals in New England. Cooperation by local management, union representatives, and workers was excellent. The GM and GSD exposures to $PM_{2.5}$, OC, and EC by job and work site were determined (summarized in Table 8). The GM ratio of EC to OC was also determined for each job and work location. Similar measurements were made at fixed locations in the 4 small, rural New England terminals to compare the background and within-terminal EC and OC levels relative to the large urban terminals in Atlanta.

***Exposure Findings*** The background levels were represented by yard samples that were taken upwind of the terminals at the property line (Table 9). These samples represent the quality of the ambient air entering the properties. This was especially important because a number of other truck freight terminals are in the area, the terminals are located on one of the runway approaches to the international airport, and a major 8-lane interstate highway is within a half mile. In general, levels of the individual

**Table 8.** Exposure Assessment Results in Two Atlanta Terminals[a]

| | $PM_{2.5}$ (μg/m$^3$) | | OC (μg/m$^3$) | | EC (μg/m$^3$) | EC/OC |
|---|---|---|---|---|---|---|
| Samples | *n* | GM (GSD) | *n* | GM (GSD) | GM (GSD) | GM % |
| **Personal samples** | | | | | | |
| Long-haul driver | 4 | 55.1 (1.20) | 5 | 27.3 (1.95) | 3.6 (1.95) | 15.5 |
| P&D driver | 5 | 119.5 (1.47) | 5 | 48.0 (1.34) | 6.0 (1.62) | 12.4 |
| Dockworker | 11 | 276.8 (2.06)[b] | 12 | 87.1 (1.93) | 7.4 (2.00) | 11.8 |
| Mechanic | 10 | 152.4 (2.13)[b] | 10 | 56.4 (2.39) | 3.6 (1.62) | 6.3 |
| **Area samples** | | | | | | |
| Terminal dock | 10 | 53.3 (1.55) | 16 | 28.9 (1.37) | 4.2 (1.82) | 9.2 |
| Shop | 18 | 51.0 (1.79) | 6 | 19.2 (1.61) | 3.2 (1.45) | 16.4 |
| Yard (upwind) | 8 | 35.9 (1.22) | 6 | 18.8 (1.43) | 2.2 (1.62) | 11.6 |

[a] Measurements of $PM_{2.5}$, OC, and EC were made on different types of filters.

[b] Only the URG type filter samples were available for mass estimates, which were found to be biased low relative to the PEM samples.

**Table 9.** Area Levels of EC and OC (μg/m$^3$) in Large Terminals Compared with Small Terminals

| | Atlanta (2 large urban terminals) | | | New England (4 small rural terminals) | | |
|---|---|---|---|---|---|---|
| Area | *n* | OC GM (GSD) | EC GM (GSD) | *n* | OC GM (GSD) | EC GM (GSD) |
| Terminal dock | 16 | 28.9 (1.37) | 4.2 (1.82) | 8 | 23.0 (1.75) | 0.6 (2.66) |
| Yard (upwind)[a] | 6 | 18.8 (1.43) | 2.2 (1.62) | 7 | 11.9 (1.41) | 0.3 (2.14) |

[a] Yard (upwind) level is equal to background levels.

markers were lower in yard samples than in corresponding indoor samples from the same terminal.

***Exposures by Job Title*** The jobs differed in exposure levels (Table 8 and Figure 3): long-haul drivers had the lowest and dockworkers had the highest. The differences in exposure to EC, the diesel exhaust marker, were relatively small, but the occupational $PM_{2.5}$ exposures were substantially higher than outdoor levels. The 3 exposure measures were related but not well correlated, except for EC and OC, which had an $R^2 = 0.81$ for log–log correlation. The level of $PM_{2.5}$ was weakly negatively correlated with levels of both EC (–0.49, $R^2 = 0.24$) and OC (–0.367, $R^2 = 0.135$) for log–log correlation. Assuming that EC represents diesel exposure in these settings, these results show that $PM_{2.5}$ would be a poor marker of diesel exposure and that OC would add little information (although the sampling numbers were small).

The driver samples represent different traffic situations. The long-haul drivers were driving in the evening on suburban and rural highways, whereas the P&D drivers were driving into Atlanta during the day and were frequently in heavy traffic. These differences were reflected in the personal sampling results for all 3 contaminants measured. The long-haul drivers had higher EC exposure levels than the yard levels. The P&D drivers had much more exposure to EC, OC, and $PM_{2.5}$. The EC values suggest that drivers

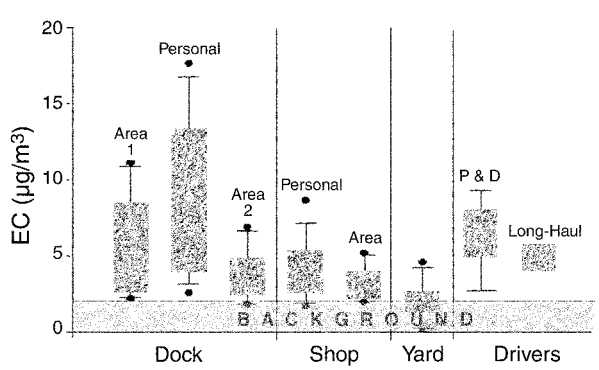


**Figure 3. Elemental carbon exposure by job (long-haul and P&D drivers) and work location (determined using personal and area monitors) for 2 large urban freight terminals.**

had exposures to diesel exhaust from traffic and possibly from small engine leaks into the cab, although the extent of exposure from possible leaks was not assessed. The EC comparison between the long-haul drivers and P&D drivers gives an indication of the expected rural-to-urban gradient in driver exposures to diesel emissions, which was approximately 2-fold.

Dockworkers and mechanics had higher personal exposures to $PM_{2.5}$ than did either type of driver. Dockworkers were the most highly exposed of all workers. Probably the heavy vehicle traffic in the dock area produced considerable diesel exhaust, propane forklift truck exhaust, and surface dust particles from tires. The source apportionment analysis of the composited high-volume filters from the dock area showed a chemical composition associated with idling diesel trucks (JJ Schauer, personal communication, 2000). This demonstrated that source apportionment analysis was feasible, but the data were too limited to draw detailed conclusions. Personal exposures of the dockworkers were generally higher than the dock area measurements presented in Table 8. This is typical of jobs in which the source of exposure is localized.

Mechanics' exposure is associated with the sporadic operation of truck engines when the tractors are brought in and out of the shop area plus any exhaust in the area from refueling, which was done in a part of the shop building. While the $PM_{2.5}$ and OC exposures for mechanics were higher than for long-haul drivers, the EC exposure for mechanics was similar to that for the long-haul drivers. This suggests a difference in sources for the particulate matter and organic compounds between these jobs.

Area samples were also collected at 4 small terminals in rural New England. The size and location (urban versus rural) of a freight terminal should affect the level of exposure because fewer diesel emission sources operate around smaller terminals in rural areas. This is clearly shown in Table 9, which compares the EC and OC levels measured in the large terminals in Atlanta with those in the 4 small New England terminals. A difference of 6 to 7 fold was seen between the GM exposures to EC for the Atlanta and New England terminals in both the dock and yard samples. The background EC level measured upwind was substantially higher in the urban location. The large terminals showed a GM 2.0-$\mu g/m^3$ increase from yard traffic, whereas the small terminals showed a 0.3-$\mu g/m^3$ increase. The 2-fold difference in EC levels between the yard and dock locations for both Atlanta and New England terminals was not statistically significant ($P > 0.05$). These data show that even in rural locations the existence of diesel sources near the terminal can increase exposure levels.

The differences in OC were much smaller than the differences in EC between the urban and rural measurements. In the urban areas, the background levels of OC were 58% higher and the dock levels were 26% higher. The relative differences between the terminal and yard were larger for the rural terminals, indicating a larger impact from the terminal emission sources. The increase above background for both EC and OC is likely to be proportional to the number of vehicles assigned to the terminal and to the volume of freight handled.

Interpretation of the measurements made during the feasibility study is tentative because both the total number of observations and the number of sites visited were small. The limited data did not permit a formal statistical analysis. These results strongly suggest, however, that this cohort experienced a wide range of exposures to diesel exhaust and that the source-receptor approach is useful for analyzing the exposure. The observed variability and mean EC levels showed little overlap between the large urban and small rural terminals. This indicates that the exposure contrast will be sharp, avoiding the need to rely on statistical differences. The differences were also large relative to measurement error of the monitoring techniques. If a large number of samples are collected under a wide variety of conditions, there are likely to be other subtler differences associated with characteristics of the exposure situations.

Mean estimates of current ambient exposure to EC from diesel exhaust ranged from 1.9 to 5.6 $\mu g/m^3$ in southern California (Cass and Gray 1995); historical peaks of 15 to 30 $\mu g/m^3$ have been reported in the same area (Cass et al 1984). Concentrations of EC on the sidewalk, collected in 1996 in Harlem NY, ranged from 1.5 to 6.0 $\mu g/m^3$, and variations were directly related to diesel vehicular traffic (Kinney et al 2000). Therefore, our results in trucking company workers occupationally exposed to diesel exhaust are relevant to selected general population exposures.

An important feature of the exposures observed was that they cover and extend above the range of normal urban population exposures. As a result, the study findings will be able to address the critical question of risk derived from low-level population exposures. In addition, inclusion of source apportionment methods in the study protocol makes it feasible to characterize the source of the EC and to partition the mix of pollutants associated with the range of common combustion sources: cars, trucks, home heating, power generation, and secondary pollutant formation.

***Temporal Variations in Exposure*** Direct reading instruments, the DustTrak for $PM_{2.5}$ and the Q Trak for CO, $CO_2$, temperature, and relative humidity, were used to measure

changes in particulate concentration over several hours and up to a full shift in the terminal areas of the dock and shop and in the cabs of operating vehicles. The objectives were to determine variability of conditions over time and to detect specific sources of potentially confounding materials such as cigarette smoke. $CO_2$, temperature, and relative humidity are indicators of room occupancy and ventilation. Human breath is the main source of $CO_2$ in an occupied room. When doors or windows are opened, temperature and humidity will change toward outdoor conditions, which rarely equal those indoors. As a result, the time profiles of temperature reflect changes in ventilation with outdoor air. This was seen in the analyses of data within the operating truck cab, in which it was clearly evident when the windows were opened or closed.

Figure 4 shows a typical time profile of variation in $PM_{2.5}$ levels in the dock area of one of the Atlanta terminals compared with those in the yard upwind. The spikes of particulate matter in the dock area relative to the yard represent the diesel emissions entering the work area from trucks outdoors. A similar pattern was seen at the dock area of a rural New England terminal (Figure 5). Note that the dock area levels were much lower than those in urban Atlanta, confirming the findings shown in Table 9 for the integrated samples.

Real-time monitoring in the truck cabs was used to observe how traffic and driving location affected the $PM_{2.5}$ level. Figure 6 shows the $PM_{2.5}$ levels during a single delivery run. None of the exposure differences during the trip was dramatic, including the difference between stop-and-go traffic and urban highway driving. Apparently, the turbulent mixing is sufficient to smooth out short-term variations. Real-time monitoring was also used to observe the effects of cigarette smoking (Figure 7). Each relatively

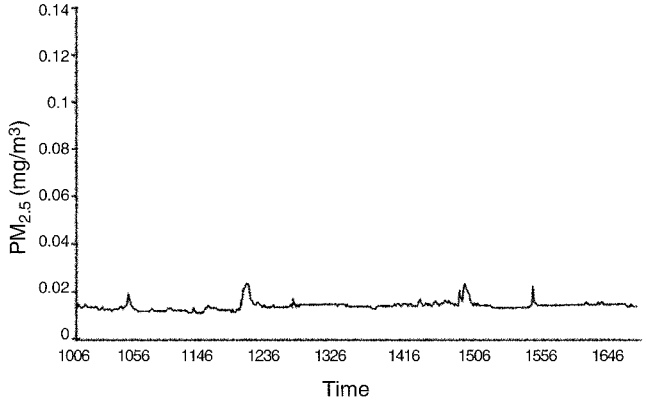


**Figure 5. Variation in real-time $PM_{2.5}$ levels in the dock area of a rural New England terminal.**

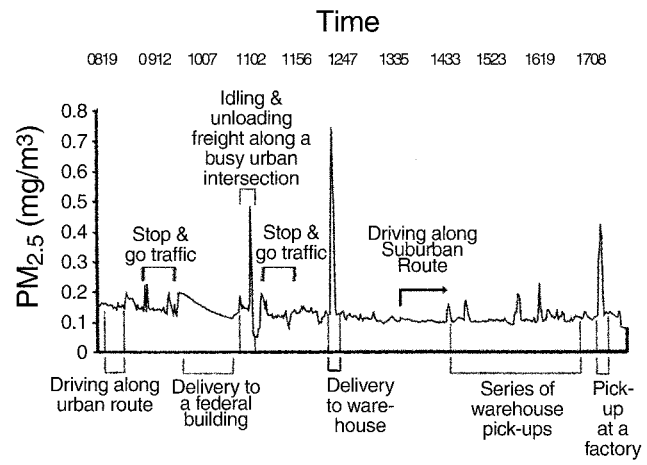


**Figure 6. Real-time profile of $PM_{2.5}$ levels in a truck cab in Atlanta with driver's activities identified by time period.**

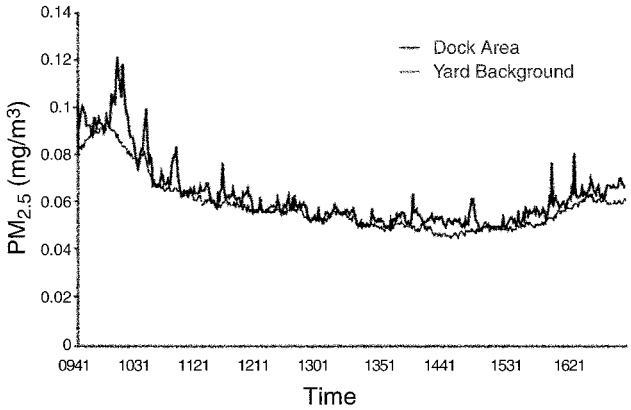


**Figure 4. Variation in real-time $PM_{2.5}$ levels in the dock area and in the yard (background level) upwind of an urban Atlanta terminal.**

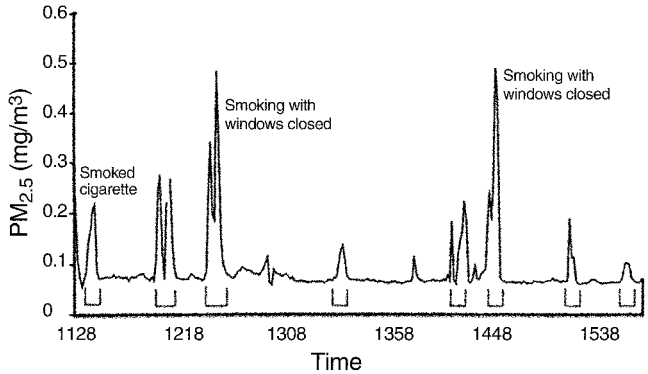


**Figure 7. Real-time profile of $PM_{2.5}$ levels in a truck cab in Atlanta with a driver smoking cigarettes.** ⊔ = smoking.

brief spike represents one cigarette smoked with the windows open based on recorded time of cigarette smoking (a study team member accompanied the driver and recorded conditions during the trip). There was little tendency for the smoke to accumulate during these warm-weather tests, which would not be the case if the windows were closed.

Comparisons were made between the $PM_{2.5}$ levels measured with the DustTrak and the standard PEM filter sampler. The weak correlation ($R^2 = 0.07$) is consistent with the sensitivity of the DustTrak's response to particle size distribution, which is likely to vary across terminal work sites and from day to day in the truck cab. The plan was to obtain PEM measurements of average $PM_{2.5}$ concurrently with the DustTrak measurements and to use the PEM value as a 1-point calibration to set the scale for the responses. The calibration factor is the ratio of PEM measured value to mean DustTrak value for the same time period. This calibration was performed for each time profile. We determined that the DustTrak is more useful as an indicator of changes over time than as an indicator of the absolute $PM_{2.5}$ levels in this setting.

***Comparison to Zaebst NIOSH Data*** A rich source of historical data was found in the NIOSH Health Hazard Evaluation reports by Zaebst (1989a,b,c,d,e,f ) and coworkers (1990). These data provided not only a time point for comparison with the current sampling data set, but an opportunity to identify variability across terminals because 6 larger terminals were evaluated.

An overview comparison was made for EC between the Atlanta data and the Zaebst data, as shown in Table 10. Generally, our observations were comparable to those obtained by Zaebst for warm weather, but there were some differences. Exposures of long-haul drivers were much lower in our Atlanta data than in the Zaebst data, which may reflect the effect of new diesel technology because the majority of the Atlanta vehicles were less than 5 years old. Why the Atlanta measurements for dockworkers using propane forklift trucks were so much higher was not clear, although the number of samples obtained in our data set was small.

The Zaebst data permitted analysis of the variability across terminals compared with workers within terminals using a 1-way analysis of variance. In general, the majority of variation was observed between terminals. For example, for dockworkers at terminals where diesel forklift trucks were used, the variation in EC exposure (GSD) between terminals was 1.21, and the variation in GM exposure across workers was 1.17. The variability across terminals was generally small (ie, between-terminal GSD < 1.4), as shown in Table 10. Values of GSD are multipliers and are not additive like the SD. For example, 1 GSD above the GM is given by GM × GSD. The larger GSD values indicate increasingly skewed distributions, whereas values below

**Table 10.** Variation in Elemental Carbon Exposure by Job and Work Site in Large Terminals: Comparison with Zaebst Data

| | Atlanta Data (1999) | | | Zaebst Data (1988–1989) | | | |
|---|---|---|---|---|---|---|---|
| Job and Weather/Vehicle | *n* | GM ($\mu g/m^3$) | GSD | *n* | GM ($\mu g/m^3$) | GSD (overall) | GSD (between)[a] |
| Long-haul driver | | | | | | | |
| Warm weather | 5 | 3.6 | 1.95 | 38 | 7.0 | 1.51 | 1.20 |
| Cold weather | — | — | — | 34 | 2.0 | 1.94 | 1.32 |
| P&D driver | | | | | | | |
| Warm weather | 5 | 6.0 | 1.62 | 25 | 6.3 | 1.93 | 1.35 |
| Cold weather | — | — | — | 31 | 2.8 | 1.63 | 1.32 |
| Dockworker, forklift type | | | | | | | |
| Propane/gas | 12 | 7.4 | 2.00 | 21 | 2.4 | 2.53 | 2.26 |
| Diesel | — | — | — | 42 | 23.2 | 1.51 | 1.21 |
| Mechanic | | | | | | | |
| Warm weather | 10 | 3.6 | 1.62 | 38 | 4.8 | 2.39 | 1.27 |
| Cold weather | — | — | — | 42 | 28.0 | 2.79 | 2.41 |
| Hostler[b] | 3 | 5.8 | 1.27 | 8 | 7.2 | 1.38 | 1.21 |

[a] GSD (between) is the variability between terminals in Zaebst data (Zaebst 1989a,b,c,d,e,f; Zaebst et al 1990).

[b] Referred to as *driver-yard hostler* in Zaebst data.

1.4 are nearly symmetric and are indistinguishable from normal distributions. Two groups had large GSDs, the mechanics in cold weather and the dockworkers using propane or gasoline forklifts. In both cases, the numbers of observations were small and the groups were heterogeneous. In the Zaebst data, 2 of the terminals did not use diesel-powered forklifts: one used propane and the other used gasoline. For the mechanics, one of the terminals had very high exposures in cold weather, reportedly because the shop was "very closed up," which would trap any exhaust emissions. Weather had little effect on the terminal docks sampled by Zaebst because there were no doors or they were never closed. As a result of the low variability from terminal to terminal, it may be feasible to sample a small number of representative terminal types and obtain reasonable exposure estimates for the workers at those terminal types.

### Development of Models to Estimate Current Exposures

One goal of this feasibility study was to determine whether it is possible to extrapolate past exposures. The proposed strategy for this extrapolation was development of a statistical model of current exposures using defining factors that determined the exposure conditions. These factors could then be used to determine past exposure conditions by modeling the effects of changes in the factors. The original plan included additional sampling to characterize the statistical variability, but that plan was dropped when the budget was reduced by the sponsors. As a result, this analysis is largely descriptive rather than quantitative. A full study would be necessary for sufficient characterization of this statistical variability.

***Rationale*** Previous studies of diesel exposure have not included statistically representative samples of exposure for subjects in the epidemiologic cohort, which has limited the accuracy of exposure assignments. Data from a statistical survey of exposures across a stratified random sample of work environments in the trucking industry will make it possible to develop quantitative statistical models. These will describe variations in exposure intensity, such as EC, as a function of each work site's characteristics (exposure factors). The predictive capabilities of this model and its error structure can be assessed with calibration and validation data collected at the same time. Such studies were beyond the reduced scope of the feasibility study but would be included in a larger exposure assessment.

A source-receptor model can be used to describe exposure to diesel exhaust in a particular occupational environment or work site. Such a model defines the relationship between the concentration of contaminants in an individual's breathing zone, the emission of contaminants by sources, the transport and losses of the emissions, and the atmospheric processes that modify and dilute the emissions. The source-receptor model can be a regression model with terms for each of the model processes and factors that modify exposure intensity. This is a common approach for modeling air-pollution exposure (Kauppinen et al 1994), and Smith and colleagues (1993) have used it to describe occupational exposure of gasoline transportation workers.

Separate source-receptor models would be developed for 3 major trucking industry work sites: vehicle cab, terminal dock area, and shop (see Figure 1). In these models, the diesel engine is considered the source and the exposed worker is the receptor. The model has a term for each of the 3 sources: background air pollution, local outdoor emissions that infiltrate the work area, and indoor sources within the work area. The local outdoor and indoor source terms are defined by types and intensities of emission sources and modifiers. For example, the local outdoor source term can be defined by the number and types of trucks in the yard area of a terminal. These elements are multiplied by factors that quantify infiltration (such as the distance from the source or degree of enclosure of the indoor workplace). These factors are empirically determined by regression analysis of measured exposures as a function of workplace covariates. For this approach to be applicable to the estimation of historical exposures, the covariates must be factors that are available from historical records. Although current indoor exposures could be characterized on the basis of tracer gases and truck emissions, the tracer studies could not be done for all of the terminals where subjects have worked, and some terminals no longer exist. Alternatively, the more practical empirical factors in the model might be outdoor source, defined by number of trucks assigned to a terminal, and infiltration, defined by number of doors in the building; both factors are available from historical records held by the trucking companies.

Examples of data-derived exposure factors (Table 11) were developed from area levels (Table 9), using a simple concept: observed levels in the dock areas inside the terminals are the sum of the background level plus the emissions infiltrating from the truck activity in the yard, as shown below:

$$\text{Dock Area Exposure} = (\text{Background}) + (\text{Local Emissions by Trucks in Yard})$$

Because the background levels were measured upwind in the yard, the exposures due to truck activity in the yard could be estimated by subtracting the background from the

**Table 11.** Examples of Factor Modeling Relationships to Estimate Elemental Carbon Exposures (μg/m$^3$)

| | EC by Terminal Size Factor[a] | |
|---|---|---|
| Location Factor[b] | Large 2.0[c] | Small 0.3[c] |
| Urban 2.2 | 4.2 | 2.5 |
| Rural 0.3 | 2.3 | 0.6 |

[a] Elemental carbon exposures calculated by summing the appropriate terminal size and location factors.

[b] Estimated factors for location are the levels from the observed background yard measurements (Table 9).

[c] Estimated terminal size factors are equal to emissions infiltrating from the truck activity in the yard, calculated by subtracting background yard measurements from measurements in the terminal dock (see Table 9): for large terminals (Atlanta), 4.2 − 2.2 = 2.0; for small terminals (New England), 0.6 − 0.3 = 0.3.

observed dock area levels as shown. Large terminals have much more truck activity in their yards than do small terminals: hence more EC emissions. The volume of truck traffic can be estimated from the number of trucks assigned to a particular terminal at each point in time, which is available historically from truck company records and outside sources. A modified relationship would be developed as shown below for location $i$ at time $t$, where β is the observed regression coefficient:

$$\text{Dock Area Exposure}[i,t] = [\text{Background for Location } i,t] + \beta\,[\text{Number of Trucks Assigned to Terminal } i,t]$$

If the EC data collected during the feasibility study represent the current time period, then the data can be recombined to estimate the dock area exposures for various combinations of terminal size and location. Further, the average exposures at large and small terminals in urban and rural locations can also be calculated. This simple example (Table 11) illustrates the principles involved and shows that there is nearly an order of magnitude range in exposures expected across these combinations.

***Data Sources*** In addition to data collected during a detailed survey of current exposure by monitoring, data can be obtained directly from a company's records and from external databases. These can be used to form exposure prediction equations and to extrapolate historical exposures. The companies' computerized and paper records on fleet characteristics, along with the RJ Polk Company Truck Group database, can provide both the age of the fleet and how many trucks of different types were assigned to a given terminal at a given time. The DOT information on traffic counts on all federal and statewide highways could be used to create an index of pollution exposure from traffic sources linked to terminal addresses.

General outdoor background air pollution, including but not limited to $PM_{2.5}$, can be quantified on the basis of data from the AIRS database. The AIRS data have limitations because of changes in methods used to obtain them over time and because less precise methods were used to collect the older data. The utility of these data will have to be assessed. Results from scientific studies contemporaneous with the older AIRS data may be useful in deriving factors to calibrate between older air pollution measurements and current measurements of EC. It was beyond the scope of this study to determine the feasibility of such work. The latitude and longitude of each monitor are provided and can be linked to the terminal addresses; interpolation techniques can be used to extrapolate information for terminals without a nearby monitor to obtain measurements directly. Information from NOAA National Climate Data Center can be used to account for climatic factors. These data will be useful for air pollution modeling to estimate background exposures in areas distant from monitoring stations. Some interpolation will be needed to estimate background levels at all of the terminals.

***Source-Receptor Exposure Factor Models*** Development of these models was beyond the scope of the feasibility study but would be part of a larger study. Random effects models can be developed for exposure factors for each type of work site and for EC, OC, and $PM_{2.5}$ using the general form shown below:

$$\begin{aligned}\text{Exposure Intensity} = {} & (\text{Background}) \\ & + \text{sum (Indoor Emissions)} \\ & + \text{sum (Outdoor Emissions)}\end{aligned}$$

Each of the terms in parentheses will be associated with specific combinations of factors derived from historical data on the terminals, trucks, and characteristics of the surrounding areas. This general model can be applied to each of the specific job and work-site combinations and can be used to estimate exposures in jobs, work sites, and terminals that were not measured. A term for weather or climate can also be considered for each model: The Zaebst data showed large differences in exposure measurements for warm and cold weather. A seasonally weighted average exposure can be estimated for each terminal by the weather parameters of its area. In addition, the models can be constructed so as to use historically available data to estimate past exposures and to validate the exposure extrapolation against the older data collected by Zaebst. Historical changes in factors may produce corresponding

changes in both the composition and the intensity of exposure. Historical exposures can be extrapolated from the current exposure models by substituting values with values taken from historical written and electronic documents. The same exposure factors used to predict EC could be evaluated to determine whether they can also be used to predict OC and $PM_{2.5}$ as alternative markers of exposure to diesel or confounding materials.

***Model for Vehicle Cabs*** The general model can be modified to reflect the specific sources of emissions experienced by drivers. Model factors are somewhat different for long-haul drivers than for P&D drivers.

$$\text{Exposure Intensity (EC } \mu g/m^3) = \beta_0 + \beta_1 \text{ ( Background)} + \beta_2 \text{ (Truck Factor)} + \beta_3 \text{ (Outdoor Emissions Entering Cab)}$$

Background levels can be determined by assessment of terminal location (urban versus rural and region of the country); current background EC, OC, and $PM_{2.5}$; and EPA and state environmental sampling data in locations that are not measured. It is important to demonstrate that values in EPA and state data are comparable to those measured. Long-haul drivers drive predominantly on highways outside cities and towns, and the observed levels in the feasibility study and the Zaebst data were consistent with this. Therefore, their background exposure can be assumed to be low and equivalent to rural levels measured by region of the country. The background for the P&D driver is the local air pollution in the location where deliveries are made (such as in the areas surrounding the terminals). Outside cities, deliveries in rural areas and small towns usually have low background air pollution as indicated by rural sampling done in these tests, by the EPA, and by some of the states. The value for background pollution can be determined by reevaluating the measured current background levels in urban and rural terminals or by consulting historical local air pollution data, when available, in the AIRS database.

The truck factor is an addition, which is a function of the trucking company, terminal location, and truck model, year, and cab design. The company-specific diesel timeline (Table 2) can be used to determine the probability that a given long-haul driver or P&D driver was driving a diesel truck during a given period of time. For both types of drivers, engine emissions may also enter the cab through leaks at the floor level. The amount of emissions entering the cab depends on the quantity of emissions entering the engine compartment, any leaks in the exhaust system, and the size of holes or gaps into the cab. Such gaps may be breaks in the gear-shift rubber boot or the seal between the cab and engine compartment, both of which depend on the type of cab, company repair and maintenance practices, and age of the vehicle. These will affect the distribution of in-cab exposures, with higher exposure more likely to be associated with older vehicles. As it will not be possible to specifically identify vehicles with exhaust leaks into the cabs, one goal of a detailed exposure assessment could be to determine if cab design is a significant predictor of exposure, and how exposure is broadly affected by model year and maintenance practices. These variables, specific to the company and to the terminal location, can be determined from company databases.

The outdoor emissions entering the cab are a function of the number and type of vehicles in traffic. Dilution of on-the-road emissions entering the cab depends on the distance between vehicles and their average speeds, both of which affect the amount of turbulent mixing. Open or closed windows and the use of "fresh air" vents that draw in contaminated air also contribute to exposure, but practices vary among drivers and cannot be assessed for individuals in the cohort. At highway speeds, emissions are rapidly diluted by turbulent mixing, whereas in stop-and-go traffic (more likely to be experienced by P&D drivers in urban locations), emissions may accumulate around the slow-moving vehicles. Our real-time monitoring of $PM_{2.5}$ found no evidence of this, however. A potential index of outdoor emissions can be based on heavy truck counts per mile collected by individual states or DOT.

***Model for Terminal Dock and Shop Operations*** The general model can be modified as shown below to reflect the specific sources of emissions experienced by dock and shop workers:

$$\text{Exposure Intensity (EC } \mu g/m^3) = \beta_0 + \beta_1 \text{(Background)} + \beta_2 \text{ (Indoor Emissions in Terminal Dock or Shop)} + \beta_3 \text{ (Yard Emissions)}$$

The background for a terminal is the local air pollution in its location: city versus small town or rural area. The AIRS data can be used to estimate the background levels. Outside cities, the terminals in rural areas and small towns are expected to have background air pollution levels consistent with rural measurements. Comparison between measured background levels and local air pollution data can determine how well the available pollution data correlate with measurements of terminal-area background exposures.

Indoor emissions in the dock area are defined by the number of forklifts within the dock area. The forklift emissions are released into the volume of air in the terminal,

which is proportional to the floor space or the number of doors. This factor can be examined as a function of company data on each terminal's loading dock area, number of loading dock doors, and number of forklifts assigned. Historically, diesel forklifts were used in larger terminals in 3 of the 4 companies (see Table 2). Therefore, for the reconstruction of historical exposure, a scaling factor based on data from Zaebst et al (1991) can be developed to account for this exposure for the years diesel forklifts were used.

Indoor emissions in shop are defined by number and type of tractors or trucks being repaired. The emissions are released into the air volume of the shop so the size affects the exposure intensity. The size of the shop can be obtained from company records, and the number of vehicles can be estimated from the number of mechanics assigned to that location. These factors can be examined in the exposure assessment as potential predictors of exposures.

The quantity of outdoor yard emissions entering the building is affected by the number and type of vehicles operating in the yard and by the amount of dilution or mixing in the yard before the emissions enter the terminal or the shop. The average number of diesel vehicles and terminal size can be examined as predictors of exposure at each location.

For P&D/dockworkers who may work on a loading dock or drive a P&D truck depending on terminal size and location, exposure assessment would require weighting time spent on the dock versus time driving. This proportion would be applied to the exposure model developed for each separate work location. Company payroll records can be used to identify hours spent working on a loading dock or driving a truck and to determine an exposure profile (mean and variation) based on terminal size and location for a typical P&D/dockworker.

**Estimation of Historical Exposures**

Two definitions of long-term exposure to diesel exhaust can be assigned to each individual. First, exposure can be defined categorically on the basis of yearly job title, terminal size, and terminal location (that is, region of the country and urban versus rural area). Second, exposure can be defined quantitatively as cumulative dose of EC using the statistical model derived from the exposure assessment discussed in the previous section. The company personnel files provide chronological job title and terminal information. Teamsters union pension fund records of contributions made by each unionized trucking company on behalf of a worker supplement this information for retired workers.

Previous epidemiologic studies have used job title alone to categorize exposure to diesel exhaust (Steenland et al 1990; Cohen and Higgins 1995; Bhatia et al 1998). For this cohort, the results showed that job alone might give highly misclassified exposure assignments. This definition should be refined by including terminal characteristics and formulating job-terminal exposure categories for historical periods. The different duties encompassed by the job title determine the potential contact that an individual has with diesel vehicles. However, size and location of the trucking terminal also appear to strongly influence exposure and the actual definition of job duties covered by the job title. Large terminals have greater activity and number of vehicles, which leads to higher exposures. At smaller terminals there is also less distinction between dockworkers and P&D drivers because a given worker often performs both sets of duties. Therefore, a dockworker at a small terminal does not have the same exposures as one at a larger terminal. In addition, the urban or rural location of the terminal influences the exposures experienced by all employees, including the drivers. Terminals in urban areas have higher levels of background air pollution owing to the other sources around them. Drivers based at these terminals are more likely to drive congested city routes than drivers from rural terminals. Long-haul drivers are assigned to a home terminal, indicating the region of the country in which they are most likely to drive. Intercity highways and those in the heavily populated East Coast and West Coast areas are more congested than those in less populated parts of the country. All of these factors influence exposure to diesel exhaust and therefore should be taken into account when grouping trucking employees into exposure categories. Dockworkers and clerks at small rural terminals appeared to have the lowest exposure, with exposures at or near background levels.

Several semiquantitative exposure indices can be developed from the duration an individual has worked in the above exposure categories. These permit evaluation of important exposure contrasts. Because there is little crossover among the categories, risk associated with years of work at small terminals can be compared with risk associated with years of work in large terminals to determine whether the cumulative risk differs by terminal size. Since P&D trucks are assigned to specific terminals and terminals vary in the year when they first used diesel trucks, years driving a diesel P&D truck can be used to index driver exposure. Similarly, diesel exposure for mechanics and dockworkers can be calculated on the basis of an individual's assignment to a specific terminal. Years of use of diesel-powered forklifts can also be determined. Years of driving a P&D truck for city delivery can be compared with

years of rural delivery to determine whether driving in traffic increases the risk beyond driving by itself. These categorical and semiquantitative exposure categories have a major advantage in their historical accuracy of potential exposure because they are based on company records and do not depend on the accuracy or precision of retrospective exposure estimates.

Quantitative historical exposures can be backward extrapolated from a statistical evaluation of current exposures. The same statistical model can be used based on historical changes in the exposure factors. In developing this approach, important elements are (1) a set of factors for which historical data are available over the whole time period of interest, and (2) data that document the qualitative and quantitative effects of changes that can be used for model validation. Our feasibility study demonstrated that this is generally possible, but all historical factors that might be used to extrapolate exposure in the models developed may not be available. For example, air pollution data for the 1970s are limited, and specific highway and regional traffic count data are not generally available before the 1970s. The effect of these incomplete data on the extrapolation of historical exposure will depend on the importance of the individual factors in the exposure model. Possibly surrogates for these incomplete data can be determined. However, these limitations would affect calculation of only a small fraction of cumulative exposure if diesel exposure were to be assessed through 2000 and beyond in the design of a cohort study.

Other exposure-modifying factors cannot be modeled explicitly but may be important in evaluating the measurement data and extrapolating past exposures. For example, local variations in the mix of vehicles and traffic patterns will affect the exposure relationship. Cities and urban areas can have substantial variation in local industries and geography. The effects of these limitations can be minimized with a sufficiently large statistical sample that cuts across the whole industry. Such a sample would permit reasonably precise historical estimates of average exposure intensity within categories.

In the ideal case, we would want extensive measurement data to validate our extrapolation model. However, occupational and environmental measurements of EC are limited, especially data collected with the best methods, and are not available prior to the mid 1980s. Several studies have shown that reflectance from deposits on filter paper and the old method for measuring air pollution by light transmission through deposits on filter paper (coefficient of haze) are highly correlated with EC measurements (Cass et al 1984; Kinney et al 2000). As a result, it may be possible to obtain estimates of EC for comparison with our model in the large cities where coefficient of haze was routinely measured.

Changes in emission sources are one key area of concern. Truck, car, and industrial emissions have all changed over time because of the ongoing efforts to reduce air pollution. These changes have been documented to different degrees, but generally the data include only relatively coarse assessments of total particulate matter and toxic gases. The source-receptor model implies that changes in sources will be reflected in changes in exposure at the receptor. Reductions in total emissions must be assumed to reduce component emissions, which may only be approximately true but difficult to verify.

**Estimation of Measurement Error**

Exposure measurement error will likely affect our assessment of the true relative risk of lung cancer attributable to diesel exhaust exposure. Errors depend on the quality and detail of the data on the sampling sites and historical operations. This error is most likely to be nondifferential (ie, unrelated to disease status). Thus, our observed relative risks for a dichotomous exposure will be underestimates of the true relative risk (Rothman 1981). However, the effect of error on the assessment of dose response may go in either direction (Dosemeci et al 1990; Birkett 1992). The magnitude of these effects on the observed point estimate of the relative risks can be estimated. A correction factor, $\gamma$, can be derived from the variability of EC levels between jobs, between workers in the same job, and within an individual working at the same job over time (Rosner 2000). The true logistic regression coefficient, $\beta^*$, would then be estimated as the observed $\beta$ divided by $\gamma$, and the confidence intervals of the corrected relative risk would be wider than those of the observed risk.

The data from this feasibility study were too limited to allow direct estimation of measurement error between jobs, urban and rural locations, and large and small terminals. However, we used the GSD between all jobs measured during this feasibility study and estimates of the GSD between and within workers obtained from published data and NIOSH Health Hazard Evaluation Reports to illustrate a few possible examples if lung cancer risk were compared between 2 jobs (Table 12). In our extreme example, if we assume a GSD of 1.4 between jobs, and a combined GSD of 2 between and within workers, the $\beta$ would be underestimated by 5-fold. The preliminary exposure data showed a range of 6 to 7 fold in exposures between large urban truck terminals and small rural terminals. That broad range will allow some imprecision in the numerical estimates of exposure and maintain the ability to detect differences in risk associated with exposure if the GSDs used

**Table 12.** Estimated Measurement Error Correction Term

| Observed Between Jobs[a] | | Estimated Between Workers + Within Workers[a] | | |
|---|---|---|---|---|
| GSD | $\sigma^2$ | GSD | $\sigma^2$ | Estimated $\gamma$[b] |
| 1.415022 | 0.12051 | 1.3 | 0.068835 | 0.64 |
| | | 1.6 | 0.220903 | 0.35 |
| | | 2.0 | 0.480453 | 0.20 |

[a] $\sigma^2 = [\ln(GSD)]^2$.

[b] $\gamma = \sigma^2(\text{between jobs})/[\sigma^2(\text{between jobs}) + \sigma^2(\text{between workers}) + \sigma^2(\text{within workers})]$.

in these categories are similar to those obtained for job-exposure categories in a larger study. A full study would include more extensive assessment of measurement error.

## DISCUSSION

Occupational groups likely to be exposed to diesel exhaust, such as trucking industry workers and professional drivers, have had a consistently elevated risk of lung cancer mortality of 1.2 to 1.4 (Cohen and Pope 1995; Bhatia et al 1998). Many occupational epidemiologic studies have attributed this excess mortality to diesel exhaust exposure without validating the relationship between job title and the potential for actual exposure. With the exception of previous studies conducted in US railroad workers by our research group (Garshick et al 1987, 1988; Woskie et al 1988a,b), efforts to relate job title to exposure intensity have been limited. In previous trucking industry studies, including a large study of Teamster retirees from the Central States Pension Fund (Steenland et al 1990), the assumption had been made that job title alone was an adequate surrogate for exposure to diesel exhaust. Previous studies were limited in their ability to include individuals with a duration of follow-up and documented exposure longer than 20 years because of the dates that diesel engines had been introduced. These limitations prevented firm conclusions that diesel exhaust is a definite human lung carcinogen, particularly at low levels of exposure.

In this feasibility study, we have located a large cohort and identified a broad range of source materials that can be used to construct a unique epidemiologic database: extensive trucking company personnel records and Teamsters union pension fund records. Our preliminary exposure measurements, together with a review of trucking company operations, indicate that terminal size (number of assigned trucks), location (urban or rural), and job title are important determinants of exposure. Consequently, the use of job title alone to categorize exposure would result in considerable misclassification of exposure. This would result in difficulty detecting a true effect of exposure, assigning exposure levels, and determining a dose-response relationship. This proposed database will be a considerable improvement over databases used in past studies. It also appears feasible to conduct a comprehensive assessment of current exposure. These measurements, along with historically available well-characterized exposure factors, can be used to develop a statistical model to extrapolate past exposures.

### SUMMARY OF TRUCKING INDUSTRY COHORT

To investigate the feasibility of designing an epidemiologic study, cooperation was sought and obtained from 4 large, unionized trucking companies. Starting in the mid 1980s, computerized records are available from the 4 companies that include not only yearly job history and dates of illness and layoffs but also terminal size and location. There were 55,750 male unionized workers employed in 1985, and there were 72,666 workers employed in 1999. All 4 companies have complete job and terminal assignment histories available for each worker starting in 1971. Because job and terminal assignments are stable in the industry, it would be reasonable to base pre-1971 job history on the 1971 job titles for one company missing earlier data. Because this company had converted from gasoline to diesel-powered long-haul trucks by 1965, relatively few years of important work history would be based on this assumption. As the other companies had also converted to diesel long-haul trucks by 1965, the long-haul drivers employed in 1985 could have been driving a diesel vehicle for 20 years or more, depending on employer and the duration of employment before 1985. Drivers of P&D trucks have had less exposure because diesel P&D trucks were introduced starting in the mid- to late-1970s and made up 100% of the fleet in three of the four companies by 1987. Diesel forklifts were used in the dock areas of the larger terminals for 5 to 10 years in the mid-1980s and were a major source of dockworker exposures. In an epidemiologic study using these data, detailed company and external records (eg, from RJ Polk Company) identified in this feasibility study would be reviewed to assign more specific dates of diesel use for each terminal in most cases. Therefore, by 2000, trucking workers exposed to diesel exhaust would have had sufficient duration of exposure and latency for lung cancer to develop if it is attributable to exposure.

Two large Teamsters union pension funds, which track employee contribution history and the names of the trucking company contributing on a worker's behalf, have expressed interest in this work, and one has agreed to participate in a study. Unlike company work records, these records do not include terminal size and location. The utility of the information obtained from Teamsters union pension funds would be to assess the extent of difference between years worked as a Teamster and years worked as based on company hire dates. Some workers may have changed union employers, as suggested by the work history questionnaire data.

## SUMMARY OF EXPOSURE ASSESSMENT

It appears feasible to evaluate exposure to diesel exhaust using 2 exposure methods. The first is to categorize exposure on the basis of terminal location and size, job title, and dates of the assignment of diesel vehicles and forklifts (determined from company records and RJ Polk Company databases). Exposure categories could be developed from the duration of an individual's work in different exposure situations. These categorical and semiquantitative exposure categories have the major advantage over job title alone that their historical accuracy is based on existing records and does not depend on the accuracy or precision of quantitative retrospective exposure estimates.

The second method is to define exposure in terms of estimated quantitative intensities of EC. The choice of EC as the primary marker for diesel exposure was based on source emission studies that clearly linked EC to diesels. An industrywide survey of current exposure is needed to evaluate the potential sources of exposure variation in the industry and to guide development of a statistical model. Given the uniformity of jobs and work practices in the unionized industry, representative sampling and development of a statistical exposure model are expected to estimate current exposures of workers in the cohort without major misclassification. Exposure factors would be identified, including specific terminal location and size, EPA data on local air pollution, the number and type of trucks assigned to a terminal, truck cab design, forklift type, and measurements of regional traffic composition and volume. In a large epidemiologic study, it is not possible to measure the exposure of each subject, so reasonably homogeneous groups are formed based on factors that define average exposure using a statistical model.

Changes in exposure factors, such as changes in emissions rates or composition due to changes in truck engine design, will produce changes in exposure. Historical exposures can be extrapolated from the current statistical exposure models by substituting historical values for current significant exposure factors. Assessment of historical exposure is more complex than defining a statistical model for current exposure because it is dependent on identifying historical information describing factors that influence exposure over time. Although extensive historical records are available, the assignment of exposures may be less certain for earlier exposures because of the limited information on certain exposure factors. In an epidemiologic study designed to assess exposure through 2000, these earlier exposures would represent only a small fraction of the total cumulative duration of exposure of a cohort of workers alive in 1985.

Although EC is clearly a marker for diesel emissions, it may not be a marker for an agent contributing to the carcinogenicity of diesel exhaust. Cancer risk may also be related to OC or to total $PM_{2.5}$. This limitation can be addressed by developing exposure models based on organic compounds, OC, and $PM_{2.5}$ and testing them in risk models. The same exposure factors used to predict EC can be evaluated to determine whether they can also be used to predict OC and $PM_{2.5}$ as alternative markers of exposure to diesel exhaust. In addition, EC and its associated organic compounds may be a causal agent for human lung cancer independent of the source of the EC. Our research tools cannot easily distinguish the risk associated with a pollutant source that does not have unique emissions. Because pollution interventions focus on sources, epidemiologic findings linking particular types of airborne particulate components to risk will implicate all of the sources of those materials. For example, if EC is associated with risk, then trucks, buses, cars, and home heating may all be contributors although diesel exhaust would be the largest source of EC in this occupational setting. Alternatively, if the hypothesis is that polycyclic aromatic hydrocarbon compounds influence cancer risk, then all sources of those compounds are contributors. A source apportionment strategy similar to that used in the feasibility study could identify where particular compounds are coming from currently (although historical sources of EC may have varied). According to test data, reduction in sulfur content of diesel fuel (after 1993) or changes in the aromatic content of diesel fuel are unlikely to have influenced the rate of EC and OC emissions (Norbeck et al 1998).

## SMOKING AND WORK HISTORY QUESTIONNAIRE

The data from our pilot questionnaire suggest that the prevalence of cigarette smoking in long-haul drivers is similar to that in other workers. Although the number of workers in each job category was small, smoking behavior also appeared to be similar among other job categories. The prevalence of ever smoking was approximately 62%, similar

to prevalences obtained from National Health Interview Surveys: 64.8% among male transportation workers in 1990 (Nelson et al 1994) and 68.4% among truck drivers in 1978 to 1980 (Brackbill et al 1988). Overall, the prevalence of current smokers was lower than expected. Roughly 18% of the respondents said that they were smokers, while in the National Health Interview Survey reports the prevalence was 38.6% in 1990 (Nelson et al 1994) and 53.1% in 1978 to 1980 (Brackbill et al 1988). Although it is possible that current smokers chose not to respond to the survey, it is also possible that fewer trucking industry workers currently smoke, a trend consistent with national statistics.

Cigarette smoking, although a clear cause of lung cancer, will not confound the relationship between diesel exhaust exposure and lung cancer unless it is also differentially related to exposure. Because the cohort was selected to include only blue-collar workers, a correlate of smoking habits, it is unlikely that job title and diesel exhaust exposure category would be systematically related to cigarette smoking. The preliminary data presented in this feasibility report suggest that job title is not associated with smoking behavior in the trucking industry, as would be expected in a cohort of blue-collar union workers in a single industry. Factors other than exposure that might influence smoking rates in an epidemiologic study include race, region of residence, and birth cohort. These factors can be taken into account when assessing potential lung cancer risk in a retrospective cohort study to compensate for the lack of direct knowledge of smoking habits. As demonstrated in our previous studies in railroad workers, another blue-collar cohort, small, unmeasured differences in smoking habits between exposed and unexposed workers are not likely to substantially reduce an observed relative risk attributed to diesel exhaust exposure (Larkin et al 2000).

After 3 mailings, the response rate to the questionnaire was 48.3%. The response rate to the 4-page questionnaire of the second mailing was 34%, with an additional 14% obtained after the third mailing in which the questionnaire was reduced to 1 page and greatly simplified by eliminating the job matrix. Several factors in addition to current smoking status may have precluded response to the questionnaire. Although workers did answer direct questions about their job, only a minority provided useful information on the job history matrix, a more complex and time-consuming task. Therefore, the response rate probably would have been greater if the shorter questionnaire were used for the first 2 mailings. Race was also a determinant of response. Workers in this terminal were 28.4% minority, mostly African Americans, who had a lower response rate than white workers. The tendency of African Americans to be less likely to participate as research subjects is well described (Shavers-Hornaday et al 1997; Corbie-Smith et al 1999).

Given the resources available to conduct this investigation, we were not able to pilot test several versions of a smoking and work history questionnaire. In a larger study, a variety of methods for increasing the response rate to key questions and among different groups of workers could be explored. These methods would include the use of employee focus groups to design and test a shorter, more focused questionnaire; Internet-based or personal computer–based questionnaires available for completion at selected terminals; use of a research assistant to administer the questionnaire at terminals selected for study; and telephone calls to obtain information from the nonrespondents.

## POSSIBLE STUDY DESIGNS

We conclude that it is feasible to design an epidemiologic study of exposure to diesel exhaust and the risk of lung cancer using records and environmental exposure data from the 4 trucking companies we studied. The goal of this feasibility study was identification of a cohort and proposal of study designs, as opposed to the full design of an epidemiologic study. Thus, we describe here 3 potential study designs, their strengths and limitations, and sample size calculations, and we estimate and discuss the impact of measurement error on the observed relative risk.

### Retrospective Cohort Study

***Study Population and Exposure Assessment*** In the retrospective cohort study that we propose, the population would consist of the 55,750 male Teamsters union members on the payroll of the 4 companies in 1985 for whom nearly complete job history information, including terminal assignment, is available. Together with an assessment of current exposure, these data would be used to create semiquantitative and quantitative estimates of exposure for each worker over time. Additional data are available from Teamsters union pension funds to refine the assessment of length of employment in the trucking industry for each worker. By 1985 the industry would have been using diesel long-haul trucks for 20 years or more, diesel P&D trucks for 5 to 10 years or more, and diesel forklifts on the docks for about 5 years. Therefore, the exposure experienced by this population would allow assessment of the effects of latency and cumulative exposure.

***Case Ascertainment and Follow-Up*** In this proposed study design, case-specific mortality (including lung cancer) would be assessed using the National Death Index from 1985 through the present time. Person-time of

follow-up would be calculated from the date of first hire in the 4 companies through date of death or end of follow-up, whichever comes earlier.

***Power Calculations*** Power calculations are presented for three exposure scenarios (1, 2 and 3) based on workers employed in 1985 with mortality follow-up through 2000 (16 years). The age distribution of the cohort in 1985 (Table 4) was compared with age-specific and year-specific lung cancer and overall mortality rates for white males to estimate overall mortality and mortality due to lung cancer. The formulae presented in Rosner (2000; Equation 14.33) were used to calculate the sample size for comparing 2 different incidence rates and for constructing a confidence interval around an assumed relative risk. A $P$ value of 0.05 and a baseline lung cancer rate among the reference group equivalent to age-specific and year-specific rates for white males were assumed (Table 13). The percentages of workers in urban and rural terminals with fewer than 100 workers and with 100 or more workers were based on the 1999 distribution of workers (Table 7). The job distribution of workers whose 1985 job titles were not known ($n$ = 3,834) was assumed to be the same as that of workers with known jobs.

In scenario 1, exposure assignment was based on job title and terminal assignment. The diesel exhaust exposure group included all long-haul drivers, mechanics, hostlers, as well as the dockworkers and P&D drivers only at large terminals ($n$ = 37,910). The unexposed group included all other workers (14,006). In scenario 2, the calculation was repeated for the long-haul drivers ($n$ = 15,190) and the unexposed workers from scenario 1. For scenario 3, exposure assignment was based on both terminal location (urban or rural) and terminal size (large or small). Dockworkers and P&D drivers at small rural terminals and clerks, janitors, union management, and trainees at all terminals were considered unexposed ($n$ = 10,690); and all other workers were considered exposed ($n$ = 41,181). With power of 80%, the detectable relative risk ranged from 1.27 to 1.33 (Table 13).

In an additional scenario (not shown), dose response across exposure groups was assessed assuming quartiles of EC exposure based on industrial hygiene measurements or 4 semiquantitative exposure categories. This included workers at large, urban terminals as those with the most exposure and workers at small, rural terminals as those with the least exposure. If 850 cases of lung cancer occurred during the study period, then there would be 80% power to detect a significant trend across exposure groups, with a relative risk of 1.23 for subjects in the highest group (Chapman and Nam 1968).

**Table 13.** Power of Study to Detect Various Relative Risks[a]

| | Scenario 1 | Scenario 2 | Scenario 3 |
|---|---|---|---|
| Relative risk at 80% power | 1.27 | 1.33 | 1.30 |
| 95% confidence interval | 1.08, 1.49 | 1.11, 1.60 | 1.09, 1.55 |
| Deaths | 6,644 | 3716 | 6,644 |
| Lung cancer cases | 850 | 465 | 879 |
| Relative Risk | | Power | |
| 1.35 | 0.94 | 0.83 | 0.89 |
| 1.40 | 0.97 | 0.90 | 0.95 |
| 1.45 | 0.99 | 0.95 | 0.98 |
| 1.50 | 0.99 | 0.97 | 0.99 |

[a] See text for description of the exposure scenarios. Relative risk and 95% confidence interval was calculated when power was 80% for various exposure scenarios.

***Adjustment for Smoking and Other Possible Confounders*** A limitation of a retrospective cohort study is the lack of specific information on potential confounders such as cigarette smoking. No specific individual information on smoking history is available. The results of the mail survey on smoking behavior performed for this feasibility study suggest that cigarette smoking would not confound the relationship between diesel exposure and lung cancer because smoking behavior was not related to job category. However, the indirect method described by Schlesselman (1978) and Axelson (1978) to adjust for confounding using group-level information could be used to assess the impact of cigarette smoking on lung cancer risk. A sample of the survivors of the cohort in 2001, the majority of the population (estimated $n$ = 49,170), can be surveyed to inquire about smoking habits. Each company maintains the addresses of employees who are still working and who have recently retired (generally within 1 to 5 years), and the addresses of most of the older retirees will be available through the Teamsters union pension funds. Thus, a detailed assessment of smoking habits based on job title and work location among these workers could be performed and used to calculate an overall adjustment factor for the effect of smoking. This method assumes that the relative distribution of smoking habits based on the various exposure categories (job, terminal, and other categories) has remained stable over time and that there is no interaction between exposure and cigarette smoking in lung cancer etiology. Several different definitions of smoking

behavior could be explored, such as duration, amount smoked, time since quitting, and age at start.

### Case-Control Study

In this section we describe two possible case-control study designs: one nested in the retrospective cohort described in a previous section and the other identifying the population from the Teamsters union pension funds.

***Nested Case-Control Study*** Of the 850 to 879 lung cancer deaths expected in the retrospective cohort starting in 1985 (Table 13), approximately 400 cases are expected to occur between 1995 and 2000. In a nested case-control design, the next of kin can be contacted and asked to provide information on cigarette smoking. Steenland and coworkers have demonstrated the feasibility of obtaining next-of-kin smoking information several years after death of a Teamster (Steenland et al 1990). If a ratio of 1 case to 10 controls for the estimated 400 lung cancer cases is used, then the power to detect a relative risk of 1.4 would be a greater than 80%, assuming that 25% of the controls worked in a diesel-exposed job.

***Teamster Pension Fund Population*** It is also feasible to identify recent deaths using the databases maintained by the Central States Pension Fund and Western Conference Pension Trust and to obtain smoking information from next of kin. However, these funds include only retired workers and do not provide the detailed job history information available from company records. Furthermore, most of the workers in the pension funds who die each year would not have been employed by the 4 large unionized carriers; currently only 12% of workers in the Central States Pension Fund and 5 % of workers in the Western Conference Pension Trust are from these companies. Thus, only a small proportion of deaths would be accompanied by detailed work history information. We estimate that 500 cases and 10 controls per case would be required to detect a relative risk of 1.35 with an $\alpha$ of 0.05 and 80% power. In the previous study by Steenland and colleagues (1990) of the Central States Pension Fund, it took 2 years to collect approximately 1,000 cases of lung cancer with next of kin available for contact. If we restricted our study population to the employees of the 4 companies, case ascertainment in this study would take even longer.

Another limitation of this study design is that these pension funds no longer routinely collect death certificates as they did during the time of Steenland's study. Therefore, lung cancer cases would have to be identified either by contact with next of kin for all reported deaths or by searching the National Death Index. This could be both costly and time-consuming.

### Prospective Cohort Study

The third possible study design is a prospective study in which current workers and recent retirees would be identified from the company rosters and the Teamsters union pension funds, or follow-up of a cohort identified during a retrospective cohort study would be continued. It would be necessary to recruit participants by mail questionnaire to obtain information about cigarette smoking, as well as information on job duties, work experience beyond the current job, and other potential sources of diesel exhaust exposure. Information on other potential lung cancer risk factors such as diet could be obtained. Exposure would be determined from a combination of information from company records, from current and future measurements at the terminals and in the trucks, and directly from the workers via questionnaire. Disease outcome would be assessed prospectively, so the assessment of lung cancer occurrence would require long-term follow-up. One advantage of this study design is that lung cancer incidence, as well as outcomes not related to cancer such as respiratory symptoms and illnesses could be identified by follow-up questionnaires. Lung cancer mortality would still be assessed using the National Death Index, as well as from return of the questionnaire by next of kin.

### Summary of Study Designs

The advantages of the retrospective cohort and case-control designs for such a study are that exposures and deaths have already occurred, and detailed job history records can be used to help define exposure. However, a retrospective reconstruction of exposure would be less precise than current measurements and would not assess the effects of more recent changes in diesel technology necessary to meet emission standards instituted in the 1990s. A retrospective cohort study considered alone would also contain limited information about potential confounders. A case-control study using lung cancer cases obtained solely through the large Teamsters union pension funds is feasible but would be inefficient because lung cancer deaths from the largest 4 carriers would need to be identified over years to take advantage of the company work history records.

The prospective study is the most expensive and the most time-consuming of the study designs described, but these limitations would be mitigated by continuing follow-up of a retrospective cohort. Also, repeated documentation of further changes in diesel technology and exposure would be required with the long-term follow-up. At the

same time, this study design has many advantages. Information on potential confounders, including but not limited to smoking history, would be collected directly from the study subjects. Furthermore, the prospective design allows for the most accurate assessment of exposure and for assessment of effects of more recent engine designs. Finally, the assessment of cancer incidence and noncancer outcomes not related to mortality would be possible.

In conclusion, it is important to determine the relationship between diesel exhaust exposure and lung cancer because exposures are widespread and large numbers of workers are regularly exposed at levels similar to that experienced by a substantial proportion of the general population. A wealth of data is available on long-term exposure to diesel exhaust and on a gradient of exposure within the unionized trucking industry. Therefore, this is a unique population in which to assess the association between diesel exhaust and lung cancer, and such an epidemiologic study is feasible.

## ACKNOWLEDGMENTS

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## APPENDIX A. Results of Pilot Study Questionnaire

**Table A1.** Questionnaire Response Rate by Company Job Title Among 439 Male Union Employees (201 Responses)

| Job Title[a] | Total Employed | Respondents *n* | Respondents % |
|---|---|---|---|
| Long-haul driver | 213 | 106 | 49.8 |
| Dockworker | 132 | 52 | 39.4 |
| P&D/dockworker | 31 | 15 | 48.3 |
| Mechanic | 34 | 15 | 44.1 |
| Hostler | 26 | 12 | 46.2 |
| Clerk | 2 | 1 | 50.0 |
| Janitor | 1 | 0 | 0.0 |

[a] Job titles were obtained from the company employee roster.

**Table A2.** Comparison of Self-Report of Current Union Job with Company Personnel Records: 201 Male Union Workers[a]

| Company Job Title[b] | *n* | Self-Reported Job Title[c] | *n* | % |
|---|---|---|---|---|
| Long-haul driver | 106 | Long-haul driver | 106 | 100 |
| Dockworker | 52 | Dockworker | 25 | 48.1 |
| | | P&D/dockworker | 19 | 36.5 |
| | | P&D driver | 1 | 2.9 |
| | | Hostler | 7 | 13.5 |
| P&D/dockworker | 15 | P&D/dockworker | 4 | 26.7 |
| | | P&D driver | 9 | 60.0 |
| | | Long-haul driver | 1 | 6.7 |
| | | Hostler | 1 | 6.7 |
| Mechanic | 15 | Mechanic | 15 | 100 |
| Hostler | 12 | Hostler | 9 | 75.0 |
| | | Dockworker | 2 | 16.7 |
| | | P&D/dockworker | 1 | 8.3 |
| Clerk | 1 | Clerk | 1 | 100 |

[a] If only exact job titles are compared, there are 160 matches (79.6% correct); if P&D driver, P&D/dockworker, and dockworker are considered the same job, there are 189 matches (94% correct).

[b] Job title obtained from company employee roster.

[c] Job title obtained from questionnaire.

**Table A3.** Mean Age, Smoking Prevalence, Years Smoked, and Average Lifetime Cigarette Use Among 201 Male Union Workers

| Self-Reported Job Title[a] | | Age | Ever Smokers | | Current Smokers | | Years Smoked | Cigarettes Daily |
|---|---|---|---|---|---|---|---|---|
| | *n* | Mean ± SD | *n* | % | *n* | % | Mean ± SD (*n*) | Mean ± SD (*n*) |
| Long-haul driver | 107 | 53.7 ± 7.6 | 67 | 62.6 | 16 | 15.0 | 27.1 ± 12.8 (45) | 25.3 ± 14.1 (64) |
| P&D driver | 11 | 49.0 ± 5.2 | 6 | 54.6 | 2 | 18.2 | 6.3 ± 8.4 (3) | 25.5 ± 14.6 (6) |
| Dockworker | 27 | 48.3 ± 8.1 | 17 | 63.0 | 5 | 18.5 | 19.2 ± 10.0 (11) | 20.3 ± 11.7 (13) |
| P&D/dockworker | 23 | 47.0 ± 8.5 | 14 | 60.9 | 4 | 17.4 | 13.5 ± 8.8 (10) | 19.1 ± 10.1 (11) |
| Mechanic | 15 | 48.6 ± 8.1 | 11 | 73.3 | 3 | 20.0 | 17.8 ± 15.8 (9) | 23.6 ± 9.1 (9) |
| Hostler | 17 | 49.4 ± 7.4 | 11 | 64.7 | 3 | 17.6 | 22.5 ± 14.4 (7) | 22.3 ± 15.9 (11) |
| Clerk | 1 | 56.5 | 0 | 0 | 0 | 0 | | |

[a] Job title obtained from self-report on questionnaire.

**Table A4.** Differences Between Long-Haul Drivers and Others Among 201 Male Union Workers

| | Long-Haul Drivers | Others[a] | *P* Value[b] |
|---|---|---|---|
| Total number | 107 | 94 | |
| Age (mean ± SD) | 53.7 ± 7.6 | 48.7 ± 7.7 | 0.0001 |
| Ever smoker (%) | 62.6 | 62.8 | 0.98 |
| Current smoker (%) | 15.0 | 18.1 | 0.55 |
| Average cigarettes per day (mean ± SD)[c] | 25.3 ± 14.1 | 21.7 ± 12.1 | 0.16 |
| Years smoked (mean ± SD)[d] | 27.1 ± 12.8 | 17.0 ± 12.8 | 0.0004 |
| Education beyond high school (%)[e] | 29.6 | 44.8 | 0.07 |
| Body mass index (mean kg/m$^2$ ± SD)[f] | 29.3 ± 5.8 | 29.2 ± 4.6 | 0.94 |

[a] Others includes clerks, dockworkers, hostlers, mechanics, P&D drivers, and P&D/dockworkers.

[b] *P* value from *t* tests for age, average cigarettes per day, years smoked, and body mass index; from $\chi^2$ test for ever smoker, current smoker, and education beyond high school.

[c] Average number of cigarettes per day over all-time smoking for ever smokers (64 long-haul drivers, 50 others).

[d] Number of years smoked for current smokers and ever smokers (45 long-haul drivers, 40 others).

[e] Education status available for 71 long-haul drivers and 67 others.

[f] Height and weight information available for 69 long-haul drivers and 64 others.

**Table A5.** Comparison of Age, Years in Industry, Years as a Teamster, and Years at Current Company Among 125 Male Union Workers[a]

| Job Title | *n* | Current Age (Mean ± SD) | Age Started in Trucking (Mean ± SD) | Years in Trucking (Mean ± SD) | Years as Teamster (Mean ± SD) | Years at Current Company (Mean ± SD) |
|---|---|---|---|---|---|---|
| Long-haul driver | 63 | 54.1 ± 7.4 | 24.9 ± 6.5 | 29.7 ± 8.4 | 23.0 ± 10.8 | 11.1 ± 7.7 |
| P&D/dockworker[b] | 38 | 48.5 ± 7.8 | 27.5 ± 7.1 | 21.5 ± 8.3 | 19.3 ± 7.9 | 15.1 ± 5.7 |
| Mechanic | 13 | 47.7 ± 8.3 | 22.2 ± 4.2 | 25.9 ± 8.5 | 19.9 ± 9.0 | 13.8 ± 8.8 |
| Hostler | 11 | 50.2 ± 7.1 | 26.7 ± 9.1 | 24.0 ± 9.3 | 20.4 ± 7.0 | 15.7 ± 7.0 |

[a] Workers who completed the job matrix questions on the 4-page questionnaire.

[b] Because of small sample sizes, P&D/dockworker includes P&D drivers, P&D/dockworkers, and dockworkers.

**Table A6.** Differences Between Ever Smokers and Never Smokers in 201 Male Union Workers

| Smoking Status | Age ± SD | > High School | Body Mass Index ± SD ($kg/m^2$) | White |
|---|---|---|---|---|
| Ever smoker | 52.0 ± 8.0 (*n* = 126) | 33/87 (37.9%) | 27.6 ± 5.3 (*n* = 18) | 91/146 (72.2%) |
| Never smoker | 50.0 ± 8.0 (*n* = 75) | 18/51 (35.3%) | 29.5 ± 5.2 (*n* = 115) | 55/246 (73.3%) |
| *P* value[a] | 0.09 | 0.76 | 0.18 | 0.86 |

[a] *P* values are from *t* test comparing means.

## APPENDIX AVAILABLE ON REQUEST

Appendix B, Health and Work History Questionnaires, may be obtained by contacting the Health Effects Institute. Please give the full title of the Research Report, the first author's name, and the title of the appendix you wish to request.

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## ABBREVIATIONS AND OTHER TERMS

| | |
|---|---|
| AIRS | Aerometric Information Retrieval System |
| CO | carbon monoxide |
| $CO_2$ | carbon dioxide |
| DOT | US Department of Transportation |
| EC | elemental carbon |
| EPA | US Environmental Protection Agency |
| GC/MS | gas chromatography/mass spectroscopy |
| GM | geometric mean |
| GSD | geometric standard deviation |
| IARC | International Agency for Research on Cancer |
| NIOSH | National Institute for Occupational Safety and Health |
| NOAA | National Oceanic and Atmospheric Administration |
| OC | organic carbon |
| P&D | pick-up and delivery |
| PEM | Personal Environmental Monitor |
| $PM_{2.5}$ | particulate matter 2.5 µm or less in aerodynamic diameter |
| $PM_{10}$ | particulate matter 10 µm or less in aerodynamic diameter |
| $R^2$ | multivariate coefficient of determination |

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# Investigators' Report

## Measurement of Diesel Aerosol Exposure: A Feasibility Study

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# Measurement of Diesel Aerosol Exposure: A Feasibility Study

David Kittelson, Winthrop Watts, Gurumurthy Ramachandran, Dwane Paulsen, and Charles Kreager

## ABSTRACT

Traditionally, assessment of exposure to diesel exhaust aerosol (when not based on job titles or other surrogate measures) has been based on mass concentration, but this may not accurately reflect the full complexity of the exposure. Recent studies have suggested that particle number and surface area may be more health-relevant indices of diesel exhaust exposure than mass concentration. Since the most appropriate metric is still unclear, a better method of assessing diesel aerosol exposure is required, one that allows conversion from one measurement metric to another.

This project evaluated the feasibility of measuring mass, surface area, and particle number of the diesel exhaust aerosol in an occupational study of workers with low-level exposures. We evaluated the exposures of 3 occupational groups using the mass concentration of elemental carbon (by method 5040 of the US National Institute for Occupational Safety and Health [NIOSH*]) as well as other measurements obtained in near real time with an array of aerosol instruments: the mass of black carbon (BC) by aethalometer; surface area by diffusion charger (DC) and photoelectric aerosol sensor (PAS); particle number concentration by ultrafine condensation particle counter [UCPC]; and particle size distribution by scanning mobility particle sizer (SMPS). Sampling focused on the Twin Cities (Minneapolis–St Paul) and University of Minnesota transportation systems. Each type of personal sample was collected on (or in the breathing zone) of surrogate bus drivers, parking ramp attendants, and garage mechanics. Area samples were collected in buses, parking garages, and vehicle maintenance garages. These sampling locations were chosen to represent environments where vehicles powered by diesel fuel or gasoline operate on a regular basis in different ratios. The relationship between different exposure metrics (based on mass, surface area, and number) was studied, and differences in the exposures of the 3 occupational groups could be identified by these exposure metrics.

The measurements made by each instrument were consistent with each other, thus providing a useful validation of these methods. The DC and the UCPC data compared well with the more detailed information provided by SMPS. Therefore, the DC and UCPC might be used in future epidemiologic studies as they are more rugged than SMPS and thus more suitable for field use. Together they gave a meaningful representation of particle size, surface area, and particle number concentration. The near real-time aethalometer measurements of BC compared well with the elemental carbon concentrations obtained using a filter sample, while providing much lower limits of detection. The 3 occupational groups in this study had exposures between 1 and 10 $\mu g/m^3$ (ie, at the low end of occupational exposures and near the high end of ambient exposures). The results of this study show that existing instruments can accurately measure low diesel exhaust exposures.

Mass exposures to elemental carbon for the 3 occupational groups did not differ significantly statistically but were lowest for ramp booth attendants and highest for garage mechanics. In terms of surface area concentration, garage mechanics had an exposure distribution that was different ($P < 0.05$) from those of surrogate bus drivers and parking ramp attendants (whose exposure distributions were similar to each other). In terms of particle number concentration, the parking ramp attendants had much lower exposures than the garage mechanics ($P < 0.05$). Thus, depending on the exposure metric chosen, the 3 occupational groups had similar or different exposures.

* A list of abbreviations and other terms appears at the end of this Investigators' Report.

This Investigators' Report is one section of Health Effects Institute Special Report *Research Directions to Improve Estimates of Human Exposure and Risk from Diesel Exhaust*, which also includes a report by the HEI Diesel Epidemiology Working Group, four other Investigators' Reports, and an Executive Summary. Correspondence concerning this Investigators' Report may be addressed to Dr David Kittelson, Department of Mechanical Engineering, Center for Diesel Research, 111 Church Street SE, University of Minnesota, Minneapolis MN 55455-0111.

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## INTRODUCTION

Studies worldwide have found an elevated risk of lung cancer associated with occupational exposure to diesel emissions (HEI 1995; California Environmental Protection Agency 1998; US Environmental Protection Agency [EPA] 2000) in a variety of cohorts such as truck drivers, equipment operators, railroad workers, and bus workers. However, the absence of quantitative exposure data from which to estimate the dose-response relationship has limited use of these data for quantitative risk assessment (HEI 1995). These studies qualitatively estimated exposure by job title, duration of employment, and other means. Mauderly (1992), in his review of the epidemiologic evidence for adverse health effects from diesel exposures, underscored this point: "The greatest single problem in these studies is the inaccuracy of the exposure assessment...."

The precise risk of lung cancer from low-level exposure to diesel emissions is unknown. Animal studies have indicated that the particulate matter (PM) component of diesel exhaust was responsible for lung tumor development and that the mutagenic compounds adsorbed on the particles were not as important as the carbonaceous core (HEI 1995). More recent studies have linked adverse health effects to environmental exposure to fine particles less than 2.5 µm in median aerodynamic diameter ($PM_{2.5}$) (EPA 1999; Vedal 1997). Given that diesel exhaust particles are primarily less than 1 µm in diameter, it is important to assess diesel exposures within the context of all PM exposures. Although nearly all of the mass emitted by engines is in the fine particle range, nearly all of the number is in the nanoparticle range (Kittelson 1998). Emission inventories suggest that engines and vehicles are the principal contributors of fine particles to the atmosphere in urban areas. Bagley and colleagues (1996) showed that a low-emission diesel engine emitted much higher concentrations of nanoparticles than an older design. Other studies have suggested that at similar mass concentrations, nanometer-size particles are more harmful than micron-size particles (Seaton et al 1995; Donaldson et al 1996, 1998, 2001).

Diesel PM is small in size and composed of organic carbon and elemental carbon, adsorbed and condensed hydrocarbons, and sulfate. The proportion of organic carbon to inorganic carbon varies depending on fuel, engine type, duty cycle, engine maintenance, operator habits, emission control devices, lubricating oil consumption, and other factors. In general, nonextractable elemental carbon accounts for a greater fraction of diesel PM mass than extractable organic compounds (Perez and Williams 1989). By number count, most diesel aerosol is found in the nuclei mode with a particle diameter ($D_p$) less than 50 nm and a number median diameter between 15 and 20 nm when measured under atmospheric conditions. These particles are termed *nanoparticles*. However, most of the aerosol mass lies in the accumulation mode with particle size ranging from 50 to 1,000 nm and the median aerodynamic diameter between 100 and 200 nm (Kittelson 1998). The proposed threshold limit value of 0.15 mg/m$^3$ for diesel PM less than 1 µm (American Conference of Governmental Industrial Hygienists [ACGIH] 1998) as well as EPA regulations limiting diesel exhaust emissions and air quality standards for PM, are all based on mass measurements. As discussed by McCawley (1990), mass measurements of total diesel PM as the sole indicator of exposure may not accurately estimate the diesel PM deposited in lung tissue because the mass median diameter of diesel aerosol is near the maximum size for deposition in lung alveoli (Task Group on Lung Dynamics 1966). If detailed particle size data (including number, volume, surface area, and mass) are available, then a better estimate of dose and possible health effects can be derived.

Figure 1 illustrates relationships between idealized diesel PM number-weighted and mass-weighted particle size distributions and the alveolar deposition curve (Task Group on Lung Dynamics 1966; Raabe 1982; Kittelson 1998). On a number basis, nanoparticle (nuclei mode) deposition is much more efficient than accumulation mode deposition although the total mass of nanoparticles deposited is smaller. Nanoparticles are typically hydrocarbons or sulfate formed by nucleation during dilution and cooling of the exhaust, while accumulation mode particles are mainly carbonaceous soot agglomerates formed directly by combustion.

Emission standards on diesel engines have led to dramatic reductions in particle mass emitted. In a study by Bagley and colleagues (1996), however, a low-emission diesel engine emitted a much higher number of nanoparticles than an older design. This finding has raised questions about whether number-based emission standards should be considered. Unlike mass, number is not conserved, however, and may change dramatically during dilution and sampling, making it difficult to design a number standard.

Idealized diesel aerosol number-weighted and mass-weighted particle size distributions (Kittelson 1998) are trimodal and lognormal in form (Figure 1). The concentration of particles in any size range is proportional to the area under the corresponding curve in that range. Most of the particle mass is in the accumulation mode with particle diameters of 0.05 to 1.0 µm. This is where the carbonaceous agglomerates and associated adsorbed materials reside. Particles in the nuclei mode typically have diameters of 0.005

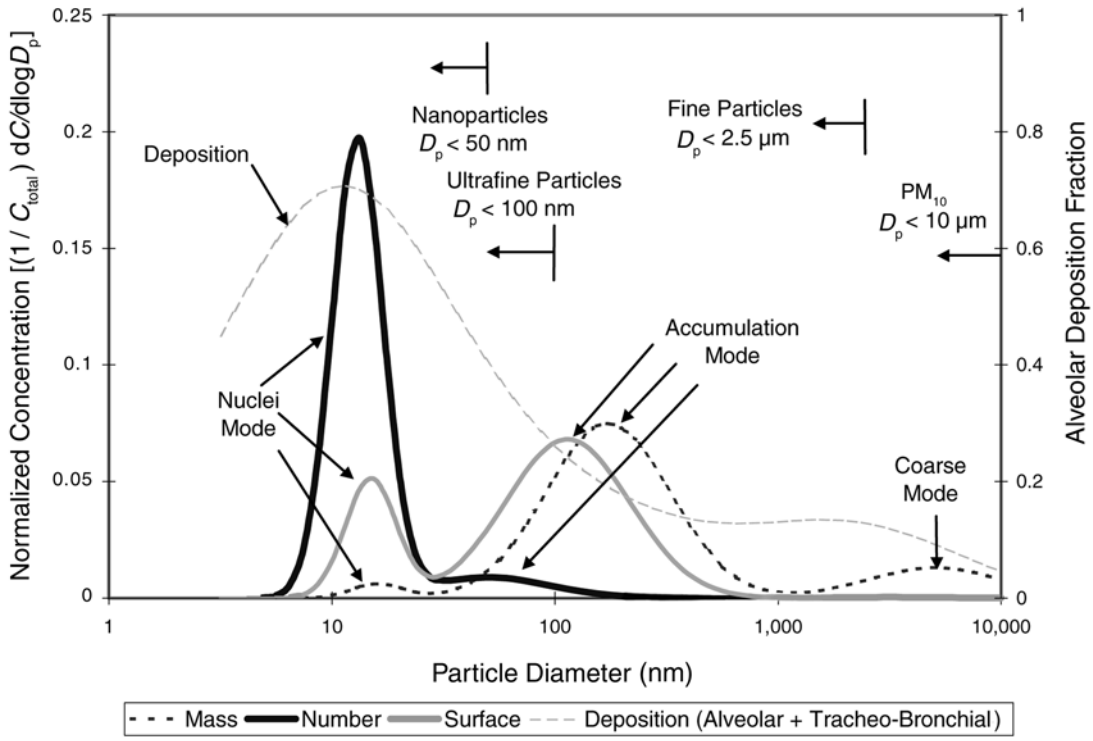


**Figure 1. Normalized mass-weighted and number-weighted particle size distributions and alveolar deposition curve from typical diesel exhaust.** *C* denotes number or mass concentration as indicated; d denotes differential; and $D_p$ is mean particle diameter. (Adapted from ICRP 1994.)

to 0.05 µm and consist of volatile organic and sulfur compounds (formed during exhaust dilution and cooling) along with solid carbon and metal compounds (from the combustion process). The nuclei mode typically comprises 1% to 20% of the particle mass and more than 90% of the particle number. The coarse mode, 5% to 20% of the particle mass, consists of accumulation mode particles deposited on cylinder and exhaust system surfaces and later reentrained. The definitions used to categorize atmospheric particles in this Investigators' Report are $PM_{10}$, $D_p < 10$ µm; fine particles, $D_p < 2.5$ µm; ultrafine particles, $D_p < 0.10$ µm; and nanoparticles, $D_p < 0.05$ µm (50 nm), but the definitions of ultrafine particles and nanoparticles are not universally set. Note that by number, the majority of the particles emitted by a diesel engine (or a spark-ignition engine) are nanoparticles. Routine industrial hygiene measurements (such as respirable or total dust) do not provide information on the aerosol size distribution and rarely, if ever, provide information on nanoparticle exposure. Typically, these measurements are based on total mass with no detailed size fractionation or chemical speciation; therefore, these measurements are nonspecific in nature.

Figure 2 depicts the trimodal surface area that arises from different mechanisms of aerosol generation (Whitby and Cantrell 1976). Primary combustion aerosols (including diesel exhaust aerosol) are formed as very small particles (nuclei mode, 0.001 to 0.08 µm), but physical mechanisms such as condensation and coagulation quickly transfer the aerosol mass to the larger particles of the accumulation mode. These processes result in a mass median diameter of approximately 0.2 µm for diesel aerosol. Mechanically generated aerosols, on the other hand, typically contain particles larger than 1 µm in diameter (with a small portion of the mass under 1 µm).

Note that the particle size distributions shown in Figures 1 and 2 are weighted by mass, number, and surface area. These distributions were determined from different instruments to cover the complete range of particle size distributions. The mass, surface area, and volume size distributions were then calculated. Whitby and coworkers (1975) conducted roadside measurements using a previous University of Minnesota mobile laboratory and confirmed the presence of aerosol in multiple modes, including a substantial nuclei mode, and demonstrated the value of determining the ratio of particle number to volume. An example of the number-to-volume conversion is discussed below.

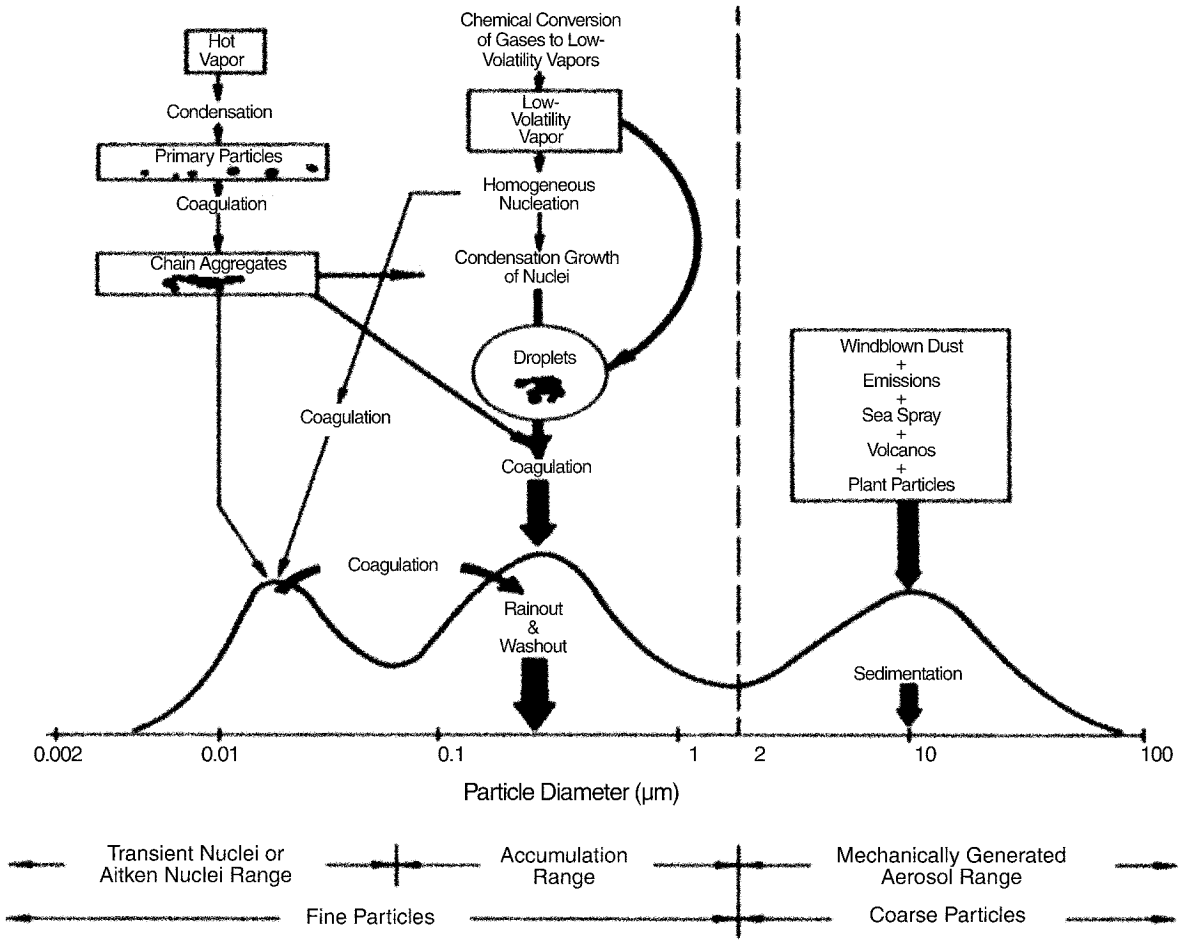


**Figure 2. Atmospheric distribution of particulate matter by surface area showing principal modes, sources of mass, and processes involved in mass transfer and removal (Whitby and Cantrell 1975).**

The fine particle standards proposed by the EPA were based on studies that linked fine particles with adverse health effects. Many of these studies showed a higher correlation between health effects and fine particle concentration rather than the concentration of $PM_{10}$ (Dockery et al 1993; Pope et al 1995; EPA 1999). Thus, adverse health effects seemed to be associated with smaller particles. Several factors may be involved in the explanation for this observation. The number of particles and the particle surface area per unit mass increase with decreasing particle size. The efficiency of deposition in the human respiratory tract depends on particle size. In particular, pulmonary deposition increases with decreasing particle size; thus, the received dose measured by particle number or surface area will increase as the particle size decreases. However, current PM pollution standards are based on either mass emission rates or time-weighted-average mass concentrations.

The specific surface area of diesel exhaust particles is typically in the range of 100 $m^2/g$ (Jakab et al 1992) for a 0.03-μm carbon sphere. Nearly all of the surface area of individual nuclei that comprise agglomerates is available for adsorption. Thus, the surface area of diesel particles is probably more a function of the size of the individual nuclei in the agglomerate than of the total agglomerate size. Various atmospheric constituents will compete with exhaust constituents for this surface area. Oberdörster and colleagues (1998) showed that for titanium dioxide ($TiO_2$), particle surface area had a higher correlation with respiratory response than either mass or number.

Special concerns have been raised about ultrafine particles and nanoparticles (Donaldson et al 1998). Particles that are nontoxic at the micron size may be toxic in the nanometer size. Among rats exposed to the same mass of $TiO_2$ as particles either 0.25 or 0.02 µm in diameter, those receiving nanoparticles retained more particles in the interstitial tissue of the lung and developed marked inflammatory responses (Seaton et al 1995). Comparison of surface free radical activities of ambient $PM_{10}$ particles and of $TiO_2$ particles that were 0.5 and 0.025 µm in diameter showed considerable activity for $PM_{10}$ and much more activity for 0.025-µm $TiO_2$ particles than for 0.5-µm $TiO_2$ particles (Donaldson et al 1996). It is striking that $TiO_2$, usually regarded as biologically inert, produced strong responses when in the form of nanoparticles. Another normally inert substance, Teflon, showed acute pulmonary toxicity in rats when administered as modest concentrations of 0.03-µm Teflon fume particles (Warheit et al 1990).

These and similar studies have raised concern about particles in the fine, ultrafine, and nanoparticle size ranges, but the decision to base the proposed EPA standard on particles less than 2.5 µm in diameter was somewhat arbitrary. This decision was made with insufficient data on the health impact of even smaller particles and nearly no data on human exposure to these very small particles. In view of the strong adverse health effects shown by nanoparticles in animal studies, future standards might be imposed on ultrafine particles or nanoparticles.

Nearly all of the aerosol mass emitted by engines is in the fine particle range and nearly all the number is in the nanoparticle range. Emission inventories suggest that engines and vehicles are the principal contributors of fine particles to the atmosphere in urban areas. For example, Schauer and associates (1996) estimated that 51% to 69% of the fine particles in downtown Los Angeles ambient air in 1982 were vehicle related. Roughly one quarter of these fine particles were fugitive dust and not directly engine related. Harrison (1996) estimated that approximately 90% of the $PM_{10}$ particles emitted in greater London in 1990 were related to road transport. Emission inventories for this area are not available for nanoparticles, but our measurements made near roadways suggest that vehicle exhaust is an important source (Kittleson et al 2001), perhaps even more than for $PM_{10}$. For example, Harrison (1996) states that particle number concentrations (mainly nanoparticles) are much more elevated near roadways than are $PM_{10}$ mass concentrations. A study by Booker (1997) performed in urban and rural Oxfordshire showed that total particle number concentration correlated strongly with vehicle traffic while $PM_{10}$ was essentially uncorrelated with traffic.

Kittelson (1998) illustrated the relative importance of nanoparticles as follows. Current on-highway engines are required to emit no more than one sixth the particle mass allowed for 1988 engines and probably less than one tenth that of an uncontrolled engine. Current emission standards do not address particle size. However, concern is growing that perhaps they should, or that a standard should address particle number. To illustrate the importance of particle number, the following is given.

Let $n$ be the number of particles and $N$ be the number concentration. Then

$$N = \frac{\text{number of particles}}{\text{volume of gas}} = \frac{n}{\text{volume of gas}}\left(\frac{\text{particles}}{\text{cm}^3}\right). \tag{1}$$

Let $v$ be the volume of PM and $V$ be the volume concentration. Then

$$V = \frac{\text{volume of particulate matter}}{\text{volume of gas}} = \frac{v}{\text{volume of gas}}\left(\frac{\mu\text{m}^3}{\text{cm}^3}\right). \tag{2}$$

Hence,

$$\frac{N}{V} = \frac{\text{number of particles}}{\text{volume of particulate matter}} = \frac{n}{v}\left(\frac{\text{particles}}{\mu\text{m}^3}\right). \tag{3}$$

Current air quality and emission standards are based on total particulate mass, $m$, but recently concerns have been raised about the number of particles, $n$, where

$$n = v\left(\frac{n}{v}\right) = \frac{m}{\rho}\left(\frac{n}{v}\right) = \frac{m}{\rho}\left(\frac{N}{V}\right). \tag{4}$$

Here $\rho$ is the average density of the PM. For spherical particles,

$$v = \sum_i n_i \pi \frac{D_i^3}{6} = \pi \frac{D_{\text{av}}^3}{6} \sum_i n_i = \pi \frac{D_{\text{av}}^3}{6} n, \tag{5}$$

where $n_i$ and $D_i$ are, respectively, the number and diameter of particles of size class $i$ and $D_{\text{av}}$ is the diameter of average volume. Then

$$\frac{n}{v} = \frac{N}{V} = \frac{6}{\pi D_{\text{av}}^3} \text{ and } D_{\text{av}} = \left(\frac{6}{\pi\left(\frac{N}{V}\right)}\right)^{1/3}. \tag{6}$$

An example is useful to illustrate the scale of these quantities. For particles with a diameter of average volume of 50 nm, or 0.05 µm, the number of particles per volume of PM would be

$$\frac{N}{V} = \frac{6}{\pi(0.05)^3} = 15{,}000\left(\frac{\text{particles}}{\mu\text{m}^3}\right).$$

The number of spherical particles in 1 g of this PM, assuming an average density of 1 g/cm$^3$, would be

$$n = \frac{m}{\rho}\left(\frac{N}{V}\right) = \frac{1\,\text{g}}{1\frac{\text{g}}{\text{cm}^3}10^{-12}\frac{\text{cm}^3}{\mu\text{m}^3}}15{,}000\ \frac{\text{particles}}{\mu\text{m}^3}$$
$$= 15 \times 10^{15}\ \text{particles!}$$

The report by Bagley and colleagues (1996) raised concern that the new diesel technology would lead to increased emissions of nanoparticles. The relative emissions of tiny particles are well represented by *N/V* ratios. Kittelson (1998) compared *N/V* ratios found in the literature from different sources. The results suggested that *N/V* ratios in the range of 50,000 to 60,000 particles/µm$^3$ (or, assuming spherical, unit density particles, $5 \times 10^{16}$ to $6 \times 10^{16}$ particles/g of emitted particle mass) are probably common for older diesel and spark ignition engines operating under highway cruise conditions. This result is consistent with the *N/V* ratio of 26,000 observed over a rural freeway under mixed traffic conditions (Kittelson et al 1988). Kittelson (1998) gave an overall range of 1,000 to 300,000 particles/µm$^3$ ($1 \times 10^{15}$ to $3 \times 10^{17}$ particles/g of emitted particle mass), and the low-emission engine tested by Bagley and colleagues (1996) produced from 100,000 to 300,000 particles/µm$^3$.

In view of the above, future epidemiologic studies clearly should be based on exposure assessment methods with health-relevant exposure metrics. Since the most appropriate metric is still unclear, a better assessment of diesel aerosol exposure is required. The new method should allow conversion from one measurement to another, such as number or surface area concentration to mass concentration.

## SPECIFIC AIMS

Sampling focused on transportation systems in the Twin Cities and the University of Minnesota. Personal samples were collected from the breathing zone of surrogate bus drivers, parking ramp attendants, and garage mechanics to obtain exposure distributions for these similarly exposed groups. Area samples were collected in buses, parking garages, and vehicle maintenance and repair garages. These sampling locations were chosen to represent environments where vehicles powered by diesel fuel or gasoline operate on a regular basis in different ratios.

The objective of this study was to validate diesel aerosol measurement techniques and to provide a more comprehensive evaluation of diesel exposure. To assess exposure, measurements from instruments capable of determining the number, volume, surface area, and mass size distributions as well as particle-associated total polycyclic aromatic hydrocarbon (PAH) content, in near-real time, were combined with time-weighted-average personal exposure measurements of elemental carbon and organic carbon. Thus the aerosol footprints of the different locations could be determined using sampling methods that enabled us to measure different aerosol characteristics in near-real time as well as the full-shift exposure. The relationship between different exposure metrics (based on mass, surface area, and number) was studied, and differences in exposures of the 3 occupational groups were identifiable when measured by these exposure metrics.

## METHODS

### INSTRUMENTATION

In order to determine the particle mass and number distribution of diesel aerosol over the entire size range requires more than one instrument. Depending on their principle of operation, instruments can determine the size distribution in a span of time ranging from seconds to hours. Kittelson and colleagues (1998) previously reviewed diesel PM sampling methods. The instrument suite used in this project was able to characterize aerosol from smaller than 10 nm to 10 µm in size and was uniquely capable of characterizing exposure (Table 1).

#### Elemental Carbon and Organic Carbon Sampling

Elemental carbon accounts for a major fraction of diesel PM emissions, acts as a carrier of suspected mutagens and carcinogens, and is readily detectable by thermo-optical analysis (Watts 1995).

Fowler (1985) concluded that elemental carbon is the most reliable overall measure of exposure to diesel exhaust because of the difficulty of selecting an extractable organic compound or class of compounds as a surrogate of exposure. The soluble organic fraction associated with diesel exhaust aerosol is highly variable in composition and chemically complex, and uncertainty exists about the compounds responsible for mutagenic and carcinogenic activity. Also, low concentrations and the presence of interfering chemical compounds make analysis difficult.

NIOSH has adopted the temperature-based thermo-optical method for speciation of organic (volatile and non-

**Table 1.** Summary of Instrumentation Used to Determine Metrics of Exposure to Diesel Exhaust Aerosol

| Instrument or Method | Operating Principle | Metric | Resolution (ranges) | Response Time (sec) | Flow Rate (L/min) | Portable Version Available |
|---|---|---|---|---|---|---|
| EC/OC analysis using thermo-optical method | Light transmittance through pyrolytically generated EC and OC | EC and OC | 0.01 μg/m$^3$ (> 0.4) | Not applicable | 3 | Yes |
| Aethalometer | Light attenuation | BC | 30 ng/m$^3$ (0–500+) | 60 | 3 | No |
| DC | Unipolar diffusion charging | Particle surface area | 10 μm$^2$/cm$^3$ (0–2,500) | < 2 | 1.5 | Yes |
| PAS 2000 | Photoelectric ionization | Surface-bound PAH | 1 ng/m$^3$ (0–1,000) | < 10 (adjustable) | 2 | Yes |
| SMPS | Electrical mobility | Particle size (number, surface area, volume) | 32 channels/decade (0.010–0.30 μm) | 60–90 | 1 | No |
| UCPC | Optical particle counter | Particle number | 0.01 *n*/cm$^3$ (0–100,000) | < 2 | 1.5 | Yes (not ultrafine) |

volatile) and inorganic (carbonate) compounds, and elemental carbon as a standard method for diesel exhaust. This method does not rely on gravimetric analysis. As noted previously, particles less than 1 μm in size are predominantly from combustion sources. Thermo-optical analysis is used to detect and quantify different types of carbon. According to NIOSH, the working range is 4.4 to 312 μg/m$^3$ with a limit of detection of about 1.3 μg/m$^3$ for a 960-L air sample collected on a 37-mm filter with a 1.54-cm$^2$ punch from the sample filter. If a lower limit of detection is desired, then a larger sample volume and a 25-mm filter may be used. Other sources of combustion aerosol such as automotive exhaust contribute organic carbon and elemental carbon to samples although the amount of aerosol in automotive exhaust is far less than the amount in diesel exhaust. This analytical method is described in detail in the *Manual of Analytical Methods* (Birch and Cary 1996; Department of Health and Human Services 1996).

Zaebst and associates (1991) made the following arguments for the use of elemental carbon as a surrogate measure of diesel exhaust exposure and for the thermo-optical method of analysis: First, most diesel exhaust aerosol mutagenicity appears to be associated with the particulate phase, and lung tumors induced in laboratory rats have been associated primarily with the particulate phase. Second, clearance in animals is adversely affected by particulate deposition, and chemically inert substances deposited in the lung alone may induce lung cancer. Third, thermo-optical analysis has 100-fold greater sensitivity to submicron elemental carbon detection than gravimetric analysis, and the diesel PM fraction is mostly carbon. Finally, nearly all elemental carbon from vehicular traffic is attributable to diesel exhaust. Further evidence to support Zaebst and associates' arguments was reported by Cass and Gray (1995). They reported that although diesel engines accounted for only 6% of the fuel use in Los Angeles, they contributed nearly two thirds of elemental carbon particle emissions during 1982. Cass and Gray focused on fine particles (< 2.0 μm) as opposed to submicron particles as suggested by Zaebst and associates.

The percentage of elemental carbon in total diesel PM varies. Major factors that contribute to this variation are engine type, duty cycle, fuel, lubricating oil consumption, engine maintenance, and the presence or absence of emission control devices. Fluctuation in the ratio of elemental carbon to organic carbon can cause an inaccurate estimate of the total diesel PM present if only elemental carbon is measured. The estimate will improve if both organic carbon and elemental carbon are measured and an adjustment is made for the presence of nondiesel carbon sources such as spark-ignition engine exhaust. (Note that NIOSH method 5040 gives both elemental carbon and organic carbon.) More detailed information on the analysis of carbonaceous compounds is available elsewhere (Hering et al 1990).

The personal sampling train for elemental carbon and organic carbon samples in this study consisted of a 10-mm cyclone (MSA, Pittsburgh PA), a 37-mm prefired, ultrapure,

quartz-fiber filter (Pallflex Products Company, Putnam CT) held in a 3-piece 37-mm cassette, and an MSA ELF pump. The MSA ELF pump had an internal laminar flow element and required calibration every 200 hours. Pumps were calibrated with an electronic bubble meter (Gilibrator, Clearwater FL) for 2.5 L/min. Two samples were collected and immediately sent to Sunset Laboratory (Forest OR) for elemental carbon and organic carbon analysis using a thermo-optical technique (NIOSH method 5040). Because the mass collected in those samples was near the detection limit for elemental carbon, we increased our sampling flow rate to 3 L/min to collect more material on the filter. The minimum sampling time was 6 hours. Increasing the flow rate decreased the cut size of the cyclone (at 1.7 L/min the 10-mm cyclone had a 50% cutoff point of 3.5 µm and at 3.5 L/min the cutoff point was 2.85 µm), but since diesel and spark-ignition aerosols are nearly all less than 1 µm in diameter, the impact on the exposure estimates was negligible.

Quartz-fiber filters are known to adsorb organic vapors during sampling. A correction factor was estimated for this adsorption by using dynamic blanks. A Teflon filter (Zefluor, Pall Life Sciences, Ann Arbor MI) was placed on top of the Pallflex filter in the 3-piece cassette, and samples were collected as discussed previously in this report. Teflon filters do not adsorb organic vapor but do remove PM, allowing the organic vapor to pass through and adsorb on the Pallflex filter, which we then analyzed by NIOSH method 5040 for elemental carbon and organic carbon. Pallflex filters are believed to reach equilibrium with the adsorbed organic material, thus allowing estimation of a correction factor. In addition to dynamic blanks, a limited number of field blanks were submitted for analysis. The field blanks were identical to normal samples except that they were not subjected to the sampling process. However, they were exposed to the same environment as the sample cassettes.

### Aethalometer

An aethalometer (Hansen et al 1984) continuously measures BC by attenuation of a light beam transmitted through a filter while the aerosol is being sampled through the filter. For a constant sampling flow rate, the rate of increase of optical absorption by the aerosol deposit on a continuously collected filter is proportional to the aerosol BC concentration. According to Hansen and coworkers (1984), BC can be measured with a time resolution of 3 seconds, approximately 10% accuracy, and a measurement resolution of about 30 ng/m$^3$ over a 3-second span at a flow rate of 60 L/min.

### Diffusion Charger

The DC measures the total surface area of PM. Positively charged ions are produced by a glow discharge forming in the neighborhood of a very thin wire. These ions attach themselves to the sampled aerosol stream with a certain probability. The charged aerosol particles are then collected on a filter. The electric current flowing from the filter to ground is measured and is proportional to the number of ions attached to the particles. For particles in the free molecular range, the attachment is proportional to the surface area of the particles but independent of the composition of the particles (Adachi et al 1985). Hence, for particles below 100 nm in diameter, the DC measures the total surface area of the particles without being dependent on the chemistry of the particles (Siegmann et al 1999).

### Photoelectric Aerosol Sensor

PASs have been used to quantify total surface-bound PAH content associated with diesel PM (Burtscher and Siegmann 1993; Burtscher et al 1993; Hart et al 1993). The PAS gives an indication of elemental carbon because it measures surface-PHA-like behavior. In a typical PAS configuration, the aerosol passes through an electrical condenser that removes any charged particles or ions. The remaining neutral particles pass through a photoemission chamber where they are irradiated with a UV laser or flash lamp. Particles with photoelectric work functions below the photon energy of the UV source emit electrons and become positive ions (Weiss 1997). The photoemission chamber has a small electric field that precipitates out the high-mobility electrons and negative ions. The positive particles are collected on a filter, and an electrometer measures the resulting current. Intensity of the particle photoemission is linearly related to the amount of particle-bound PAHs and the particle surface area (Burtscher and Siegmann 1993).

The PAS measures photoemission from aerosols, which is related to particle surface and gives a surface area concentration. The studies cited in this section all reported their results as photoemission intensity over time. More details on the relation of photoemission to PAH concentration, determined by chemical analysis, are given elsewhere (Hart et al 1993).

Only submicron particles may be irradiated and charged. Particles larger than 1 µm cannot be charged efficiently and are removed from the aerosol before it enters the photoemission chamber. Lower particle size limits and concentration limits for PAS instruments have not been reported. This method does not quantify individual PAH compounds. The detection limit is about 1 ng/m$^3$ with a time resolution of about 1 second (Burtscher and Siegmann 1993). The PAS operates in near-real time, is small in size, and is battery

operated. In this study, we used the PAS in conjunction with the personal elemental carbon/organic carbon samplers.

### Scanning Mobility Particle Sizer

Size distributions were determined using the SMPS (model 3934, TSI, St Paul MN), which has been described in detail elsewhere (Wang and Flagan 1990). This classifier can size particles by their electrical mobility equivalent diameter in the range from 7 to 700 nm. For this project it was set to scan particles with diameters from 8 to 300 nm, a range in which it works reliably. Below 0.01 µm (10 nm), efficiency of the classifier decreases as a result of diffusional broadening, reduced charging efficiency, and particle losses within the analyzer. The SMPS, considered the laboratory standard for diesel aerosol measurement, is commonly used to measure diesel exhaust aerosol size distributions in both laboratory engine tests and environmental studies (Kittelson et al 1998). In this study, the SMPS was placed in the general area of the worker and monitored at all times by project personnel. This approach enabled us to associate size distributions and concentrations with specific events during the test period.

The SMPS uses continuous voltage scans to analyze electrical mobility rather than the voltage stepping used by other instruments such as the electrical aerosol analyzer. After classification by the SMPS classifier, the particles are counted in each size category by an ultrafine condensation particle counter (UCPC). The UCPC has low particle residence time allowing rapid, high-resolution determination of the size distribution of submicron particles. After allowances are made for particle residence times, the entire size distribution can be derived from a single scan. Scans of 100 particle-size intervals can be completed in 60 seconds.

### Ultrafine Condensation Particle Counter

A second UCPC (model 3025A, TSI Inc) was used to count the total number of submicron particles. The UCPC counts particles in the range of 3 to 1,000 nm and operates by saturating the carrier airstream with a vapor, which produces supersaturation that causes vapor condensation on the particles, enlarging them to sizes that can be more readily measured by light scattering. The model 3025A UCPC measures the number concentration of small particles with high efficiency. Because of a vapor sheath that confines the aerosol flow to a path near the centerline of the condenser, all particles experience nearly identical supersaturation conditions. The result is a sharply defined lower limit of size detection. This unique flow design minimizes response time and diffusion losses of ultrafine particles. Therefore, rapid changes in aerosol concentration can be measured with accuracy. The UCPC's counting efficiency is optimized by a special flow design, which allows detection of ultrafine particles in concentrations up to $10^5$ particles/cm$^3$. If concentrations are higher than $10^5$ particles/cm$^3$, then the aerosol concentration must be diluted before it reaches the UCPC. In addition, the UCPC's response time is 1 second and it measures rapidly changing aerosol concentrations, characteristics that make it ideal for measuring transient aerosols such as those created by hydrocarbon combustion.

## SAMPLING STRATEGY

### Sampling Locations and Occupational Categories

In the workplace, variability in exposures tends to be high. Transportation workers are exposed to roadway aerosol (primarily from diesel and spark-ignition engines) in concentrations ranging from 1 to 100 µg/m$^3$. (This exposure is low compared with exposures in underground mining, for example, where aerosol concentrations are primarily diesel in origin and range from 100 to 1,000+ µg/m$^3$ [Watts 1995].) Roadway aerosol is described as either *fresh* or *aged*. As an exhaust plume dilutes and cools, nucleation, condensation, evaporation, coagulation, and numerous other events take place. These processes occur rapidly, typically within 0.25 seconds, while the exhaust plume is in the critical phase of dilution at 5:1 to 50:1. Particle growth also occurs, and the mass shifts from nuclei mode to accumulation mode. In addition, the environment in which the aerosol is generated affects the combustion aerosol both chemically and physically. Events occur more slowly as the aerosol continues to age and is transformed by chemical and physical reactions. Eventually it is difficult to differentiate between what is and what is not diesel aerosol.

The Twin Cities and University of Minnesota transportation systems together provided an opportunity to measure different exposure settings. In these environments vehicles powered by diesel fuel or gasoline operated on a regular basis in different combinations and from almost all gasoline vehicles to nearly all diesel-fueled buses. Both area and personal samples were collected depending on the specific location and situation. Area samples differ from personal samples in that they are not collected on individual workers, are not necessarily collected over a full work shift, and generally require bulky instrumentation that is not battery powered or portable. Although area samples provide more information on aerosol characteristics, personal time-weighted samples are closer to the actual exposure of the worker. Several studies have found that results from personal samples were higher than results from area samples. This points to the need for data other than area measurements of concentration to estimate actual worker exposures. Ideally, time-weighted-average personal samples should be

collected in the breathing zone of the worker over a full work shift using a battery-powered or passive sampler. During this study, however, personal exposure measurements were not always accessible because either management or union personnel raised objections, suggesting that the samplers might interfere with job performance. In many cases we had to depend on the samplers being placed as close to the breathing zone of the worker as possible, generally within 91 cm (3 ft). In the occupational cohorts studied, a full work shift was 6 to 7 hours depending on the job.

Three occupational categories were monitored: bus drivers (personal samples), parking ramp attendants (personal and area samples), and garage mechanics (personal and area samples). These were selected because they provided a mix of exposure scenarios, which differed with the mix of gasoline and diesel exhaust as well as the age of the aerosol, and because they were representative of workers employed in the transportation industry.

Although the University of Minnesota pledged cooperation in this project, the transportation company that operated the University bus fleet refused to allow samples to be collected on its bus drivers or on its buses. This forced the investigators, through the University, to negotiate with the Metropolitan Council that operates the Twin Cities bus fleet to allow project personnel, acting as surrogate bus drivers, to obtain samples while riding metropolitan buses.

For the parking ramp attendants cohort, personal and area samples were obtained in 2 University of Minnesota parking ramps after an agreement was negotiated with the University's Parking and Transportation Services. Although personal samples were not collected in the breathing zone of the parking ramp attendants, they were collected very close to their breathing zone.

For the garage mechanics cohort, an agreement negotiated with the Minnesota Department of Transportation allowed us to sample at 2 garages that housed diesel vehicles used for various road work: the Camden garage in Minneapolis and a relatively new garage off Cedar Avenue in Richfield MN.

### Number of Personal Samples for Each Cohort

Personal samples were collected for surrogate bus drivers, parking ramp attendants, and garage mechanics to estimate exposure to elemental carbon less than 0.8 µm in size. Sunset Laboratory analyzed samples for elemental carbon/organic carbon content by NIOSH method 5040. The number of samples to be collected to estimate the mean concentration was determined by the expected standard deviation of the measurement method, the range about the sample mean for the population, and the confidence level which defined that range (Devore and Peck 1993). For a confidence level of 95%, the corresponding sample size was calculated by the following equation:

$$n = \left(\frac{1.96\sigma}{B}\right)^2, \tag{7}$$

where $\sigma$ is the expected standard deviation of the method, and $B$ is the value such that the population mean lies within a range $(x - B, x + B)$ with 95% confidence (where $x$ is the sample mean). The expected standard deviation of the elemental carbon method is 8% of the measurement (Birch and Cary 1996). If $B$ were chosen to be 2.5% of the sample mean, then a sample size of 40 would be sufficient to determine exposure for each population within 2.5% with 95% confidence.

### Surrogate Bus Drivers

Two members of the project team served as surrogates for bus drivers and rode the metropolitan buses for 6 to 8 hours wearing personal samplers. On 2 occasions, the surrogate bus drivers wore multiple samplers so that the intersampler variability could be studied. We assumed that surrogate bus driver exposure would be similar to the actual bus driver exposure. Minnesota prohibits smoking on buses although some bus drivers may smoke when they are not driving. Neither surrogate bus driver smoked. The specific routes and stops of the bus ridden defined the surrogate bus driver's activity. The amount of time spent on the bus was recorded each day.

**Table 2.** Summary of Bus Measurements

| Bus Route | Description of Route | Number of Measurements | | |
|---|---|---|---|---|
| | | EC/OC | PAS | DC |
| 16 | Downtown Minneapolis to downtown St Paul | 9 | 4 | 3 |
| 27 | Downtown Minneapolis to Anoka | 8 | 3 | 2 |
| 75 | Downtown Minneapolis to Wayzata | 11 | 1 | 1 |
| 80 | Downtown Minneapolis to Mall of America | 11 | 0 | 0 |
| | Total | 39 | 8 | 6 |

Personal air monitoring of the bus routes comprised 39 air samples divided, roughly equally, among 4 different bus routes in the Twin Cities metropolitan area (Table 2). These routes were selected, prior to sampling, on the basis of their traffic patterns. Route 16 traveled between downtown Minneapolis and downtown St Paul, where the bulk of the traffic was a mixture of spark-ignition commuter automobiles with some diesel truck traffic. Route 27 traveled between downtown Minneapolis and suburban Anoka approximately 20 miles away. Traffic comprised spark-ignition autos and diesel trucks that followed this route to different industrial sites. Route 75 was also a suburban corridor to downtown Minneapolis but had somewhat lighter diesel truck traffic. Route 80 went from downtown Minneapolis to the Mall of America and was composed mostly of freeway traffic along Interstate 35W going south to Bloomington. The bus stop at the Mall of America was an underground terminus shared by many other bus routes and therefore had more diesel bus traffic.

The surrogate bus driver sat behind the actual bus driver and wore the cyclone sampler on a vest in the breathing zone. Project personnel entered the bus at the terminal bus stop and sat as close to the driver as possible near the window. The rider then got off the bus and waited for the next bus to take them back to their point of origin. The same bus typically would not run the same route all day long. In fact, different buses with different drivers would travel each bus route throughout the day.

The surrogate bus drivers also carried 2 real-time, battery-operated, portable instruments that were small versions of the DC and PAS detectors previously described (EcoChem Analytics, League City TX). They held the instruments in their laps with a common sampling tube for the 2 instruments extending into the breathing zone next to the cyclone sampler. Both instruments gave a running 10-second average of the surface area and PAH concentrations. At the end of the day, data were downloaded onto a computer. These instruments had a battery life of approximately 5 hours. In contrast, the personal sampling pumps would run for the entire time that the surrogate bus driver rode the bus. Thus, meaningful comparisons between the time-integrated PAS and DC measurements and the elemental carbon/organic carbon concentrations were not always possible.

Elemental carbon/organic carbon samples were taken for all 39 bus rides. The elemental carbon/organic carbon filters were collected at the end of the day and stored in a freezer until shipped to Sunset Laboratory for analysis. Because only one person could take the PAS and DC measurements, PAS data were obtained for only 8 of the 39 bus ride days and DC data were obtained for only 6 of the 39 bus ride days. On the days these samples were taken, the field personnel also maintained a log of the traffic pattern encountered by the bus on a minute-by-minute basis. The breakdown of the bus-phase sampling method is in Table 1.

**Parking Ramp Attendants**

Parking ramp attendants at the University were exposed to a mixed aerosol with the major contribution coming from automobiles. The 2 locations used were the 4th Street ramp and the Washington Avenue ramp on the University of Minnesota East Bank Campus. The Washington Avenue ramp was one of the busiest campus parking facilities with constant in-and-out traffic. The 4th Street ramp had periods of heavy commuter traffic in the morning and afternoon. Attendants were located at both the entrance and exits of the Washington Avenue ramp on opposite sides of the building, while the entrance and exits to the 4th Street ramp were located on the same side of the building. An effort was made to exclude smokers from the sample population although we were not always successful. While the NIOSH method of analysis for elemental carbon can separate diesel aerosol from cigarette smoke, it becomes more difficult to determine the fraction of organic carbon attributable to diesel aerosol when cigarette smoke is present.

Attendants at these parking ramps worked in booths with positive pressure ventilation and windows. Ventilation in the booths at both facilities was assessed and found to provide positive pressure relative to the ambient atmosphere (ie, air flowed from inside the booths to the outside, not vice versa). However, the intake vents for both parking ramps were subject to exhaust aerosol or other contamination.

The cyclone sampler was placed near the breathing zone of the parking ramp attendants close to the open window. Several different attendants worked in the parking ramp booths throughout the day. To ensure sufficient mass for chemical analysis of the elemental carbon/organic carbon, we measured a modified area sample, keeping the cyclone around the breathing zone for several attendants rather than limit the sampling to one specific individual during the day. Typically, sampling began in the morning and ended in the late evening. Video cameras recorded activity 24 hours a day at these parking ramps, and summary traffic count data were available to the investigators.

The battery life of the portable PAS and DC instruments was not a problem in the parking ramps because AC power was available inside the booths. This allowed us to place the instruments next to the cyclone by the window and maintain power throughout the sampling period. The elemental carbon/organic carbon filters were collected at the end of the day and stored in a freezer until being shipped to Sunset Laboratory for analysis.

In total, 11 elemental carbon/organic carbon samples were collected at the 4th Street ramp. Four additional samples were collected as dynamic blanks. Results from these dynamic blanks were also used to correct for adsorbed organic compounds on samples collected during the bus sampling. In addition to elemental carbon/organic carbon samples, we also obtained measurements using the portable PAS and DC instruments at the 4th Street ramp.

Sampling at the Washington Avenue ramp utilized a suite of real-time instruments (Coordinating Research Council [CRC] Project; see Kittelson et al 1998). This suite included an SMPS, full-size PAS and DC instruments, aethalometer, and UCPC. Instruments were set up in a storage room adjoining the attendant's main office area and booth. A sampling line was run from the attendant's window through the false ceiling and into the storage area (not shown) where the flow was routed to each of the instruments (Figures 3A and 3B). The instrument suite ran for approximately 2 weeks at this location. The only difference in the elemental carbon/organic carbon sampling was that some of the samples were taken from outside the booth in an effort to compare the outside aerosol with the aerosol collected inside the booth under positive pressure. In total, 23 elemental carbon/organic carbon samples were obtained from the Washington Avenue ramp. Four dynamic blanks were also taken at this location. Twelve of the 23 elemental carbon/organic carbon samples were taken alongside the fixed, real-time instruments. Table 3 outlines the instrumentation used during the parking ramp phase of this study.

### Garage Mechanics

Of the 3 occupational cohorts in this study, the garage mechanics represented a more traditional occupational exposure group, with exposures that were more easily defined. We believe that the PM exposure of the garage mechanics was primarily due to diesel aerosol, but cigarette smoke and other submicron aerosol (such as welding fumes) may also have been present in their environment. The garage phase of this study was designed to provide an environment dominated by diesel aerosol for characterization. The locations were 2 Minnesota Department of Transportation maintenance garages that housed diesel vehicles used for a variety of roadwork activities.

The first maintenance garage investigated was the Camden garage in Minneapolis with approximately 15 workers on any given workday. The instruments used were cyclone samplers to obtain elemental carbon/organic carbon samples, the aethalometer, the UCPC, and the portable PAS and DC. The instruments were assembled

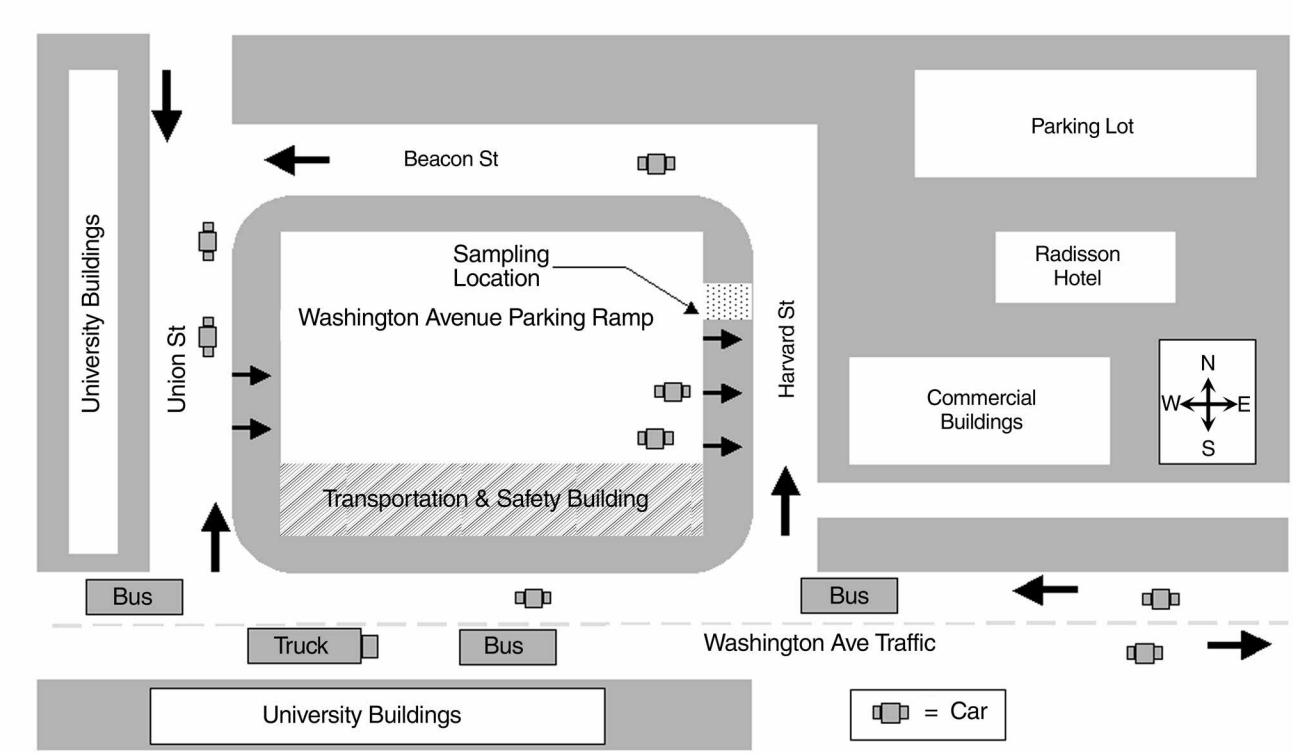


**Figure 3. Washington Avenue parking ramp. (*A*)** Sampling locations.

**Table 3.** Summary of Parking Ramp Measurements

| Location | Number of Measurements | | |
|---|---|---|---|
| | EC/OC | Portable PAS and DC | Real-time Instrument Suite[a] |
| 4th St ramp | 11 | 11 | 0 |
| Washington Ave ramp | 23 | 8 | 10 |
| Total | 34 | 19 | 10 |

[a] This suite included an SMPS, full-size PAS and DC, aethalometer, and UCPC.

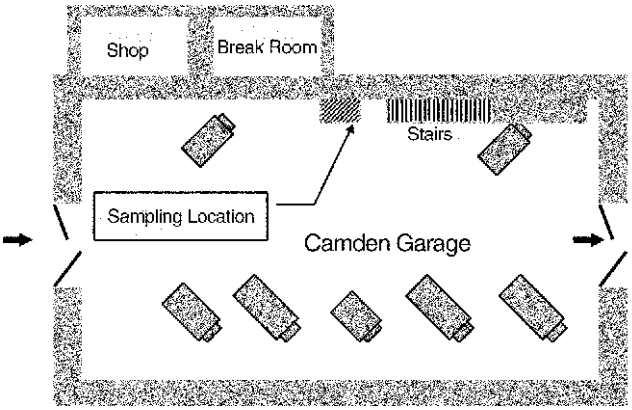


**Figure 4. Camden garage sampling location.**

against the wall nearest the workers' break room on a set of storage shelves (Figure 4). The location, midway between the large doors at opposite ends of the garage, was chosen for its convenience for supplying power and support for all instruments as well as its exposure to the aerosol most frequently encountered by garage workers.

Nineteen elemental carbon/organic carbon samples were taken at the Camden garage at 3 different locations. Seven were taken alongside the fixed, real-time instruments, in the main garage area where most of the vehicles were run. The other twelve were split equally between a maintenance bay separate from the main garage area (maintenance workers)

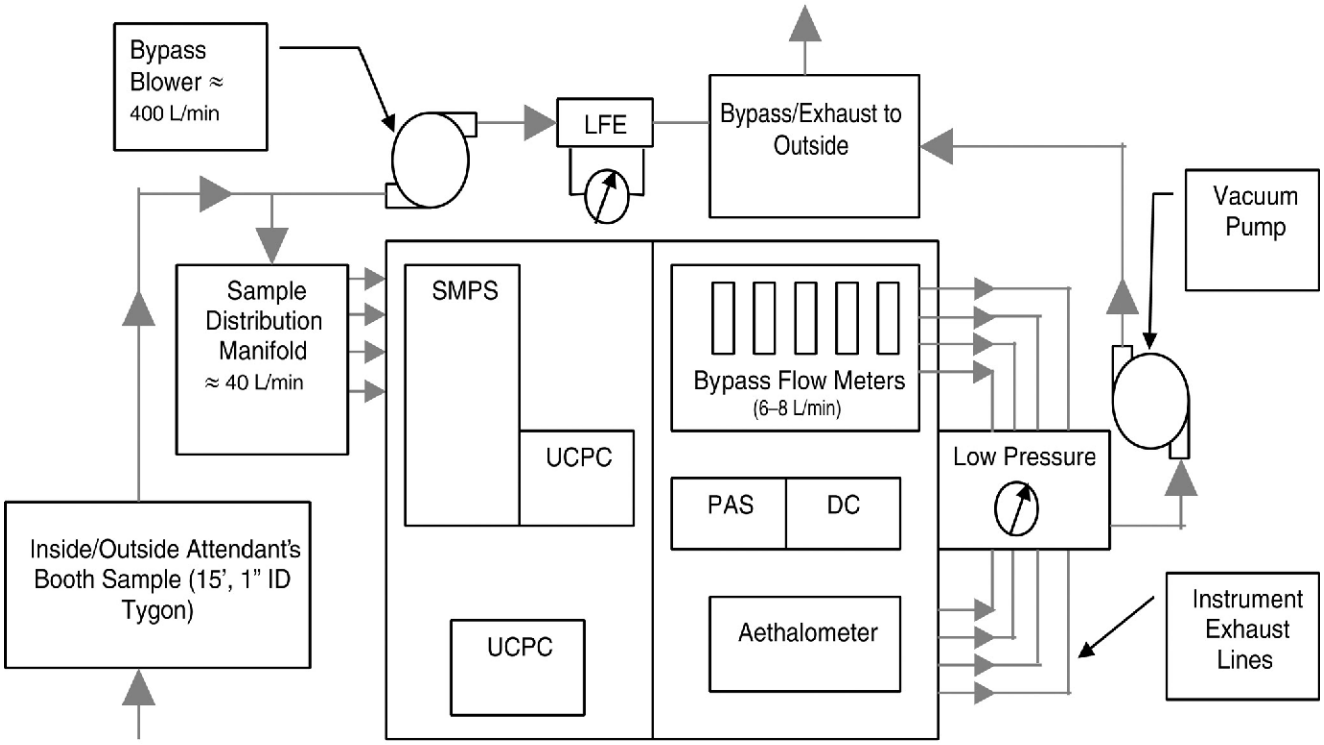


**Figure 3. Washington Avenue parking ramp (*continued*). (*B*)** Sampling system. Personal samplers and the portable PAS/DC were placed at the system-sampling inlet. The Tygon tubing inlet (1-inch inside diameter) was placed at head level and ran a length of 15 feet to the sampling system. A primary transfer blower provided the transport momentum to the sampling system and decreased the residence time to less than 1 second. Before the blower, the instruments draw a sample from a secondary transport line. After the instruments draw the sample from the secondary transport line, the excess or bypass flow is dumped to a low-pressure vessel. In addition, the instruments are exhausted to the same low-pressure vessel to complete the sampling system.

**Table 4.** Summary of Garage Measurements

| Location | Number of Measurements | |
|---|---|---|
| | EC/OC | Real-time Instrument Suite[a] |
| Camden garage | 19 | 7 |
| Cedar Ave garage | 15 | 6 |
| Total | 33 | 13 |

[a] This suite included an SMPS, full-size PAS and DC, aethalometer, and UCPC.

and a loft location one story above the instruments (clerical workers). Two maintenance workers were employed at the Camden garage during the workday. All samples were area samples. Roadworkers would typically start a vehicle at the garage and then drive off to the work site.

The second location was a relatively new garage off Cedar Avenue in Richfield. The same instrumentation was used to sample at the Cedar Avenue garage as at the Camden garage. The instruments were placed along a wall at the midpoint between the ends of the garage. Fifteen elemental carbon/organic carbon samples were obtained from the Cedar Avenue garage. Six were taken alongside the real-time instruments. Two other locations within the Cedar Avenue garage were also sampled: one was the separate maintenance bay, and the other was at a support beam in the middle of the main garage between 2 rows of parked diesel vehicles. Table 4 summarizes measurements from the garage sampling locations.

## RESULTS

This section is divided into 3 parts, each part describing the exposure assessment results based on a different exposure metric–mass concentration, surface area concentration, and number concentration.

### MASS CONCENTRATION MEASUREMENTS

The average elemental carbon concentrations were determined for all 3 occupational categories—surrogate bus drivers, parking ramp attendants, and garage mechanics (Figure 5). The specific locations where measurements were made (ie, 4 bus routes, 2 parking ramps, and 2 maintenance garages with 3 sampling locations in each garage), further divide these categories. Each measurement is the average elemental carbon concentration over a work shift (typically 6 to 7 hours). A number of measurements taken in the parking ramps and buses were close to or below the limit of detection (approximately 1 μg/m$^3$). Clearly, there were no statistically significant differences between the groups. The highest levels of elemental carbon were found in the maintenance garages, which were dominated by diesel exhaust, and the lowest levels were found in the parking ramps, which were dominated by spark-ignition (gasoline) exhaust aerosol. The parking ramps, with the lowest diesel aerosol traffic, showed the lowest elemental carbon concentrations. The 4 bus routes constituted hybrids of diesel and spark-ignition exposure, and these samples had intermediate levels of elemental carbon. The elemental carbon levels ranged from the method detection limit (MDL) of 1 μg/m$^3$ to 5.6 μg/m$^3$ for the 4 bus routes, to 2.15 μg/m$^3$ for the parking ramps, and to 17.7 μg/m$^3$ for the maintenance garages. On 2 occasions, the surrogate bus driver wore 3 samplers simultaneously so that intersampler variability could be determined. On the first occasion, all 3 samplers measured elemental carbon levels below the MDL. On the second occasion, the elemental carbon levels were 3.12, 2.68, and 3.07 μg/m$^3$, respectively (0.24 μg/m$^3$ SD).

The very low elemental carbon levels in the parking ramps reflected little diesel traffic going in and out. The measurements were made inside the booths (near the breathing zone of the workers), and because the booths were under positive pressure, outside air was prevented from entering them.

Of the 2 garages, the Camden garage was older and had less ventilation, and its elemental carbon levels were substantially (but not significantly) higher than those of the Cedar Avenue garage. We expected, on the basis of a preliminary walk-through survey, that the amount of diesel

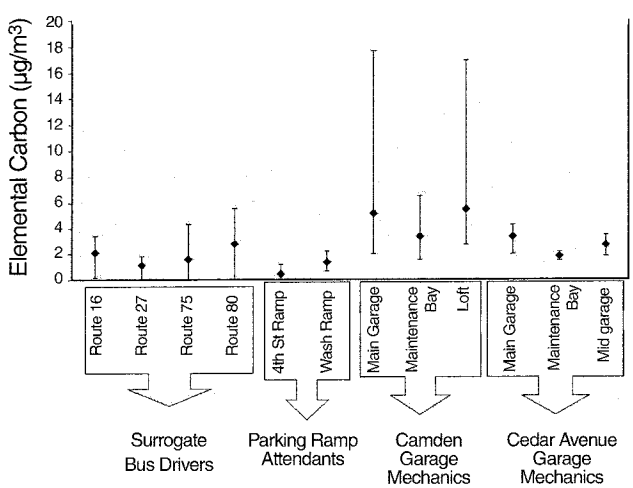


**Figure 5. Exposures to elemental carbon for 3 occupational groups: surrogate bus drivers, parking ramp attendants, and garage mechanics.** The groups are further divided into the different locations within each category where measurements were obtained. The error bars represent the range between the maximum and minimum values of the measurements.

exhaust present in the maintenance bays would be light compared with the diesel exhaust encountered in the main garages where most of the vehicles were operated. As Figure 5 shows, however, the average levels were about the same in both locations although the highest levels were seen in the main garages. Even more surprisingly, the elemental carbon levels in the Camden garage loft, where the clerical workers were located, were as high as those in the main garage area.

The relationship between elemental carbon levels and diesel sources was also illustrated by comparing the average number of diesel vehicles encountered by each bus along its route and the levels of elemental carbon in samples from that route (Figure 6). The highest elemental carbon concentration came from Route 80, which also encountered the highest diesel traffic, mainly at its terminus in an underground parking lot with high diesel bus traffic at the Mall of America. Indeed, the conditions at the Mall of America bus terminus closely resembled those at the maintenance garages.

Results from the maintenance garages were consistent with, but somewhat lower than, those of a previous study conducted by NIOSH of trucking industry workers (Zaebst et al 1991). In that study the geometric mean elemental carbon exposure was 3.8 µg/m$^3$ for highway truck drivers, 4.0 µg/m$^3$ for local truck drivers, 12.1 µg/m$^3$ for bus mechanics, and 27.2 µg/m$^3$ for dockworkers using diesel-powered forklift trucks. In the same study, background levels measured in a residential area and near the highway ranged from 1 to 3 µg/m$^3$, similar to the levels we measured in the buses and parking ramps.

The average organic carbon exposure levels also were determined for all 3 occupational categories (Figure 7). There were no clear trends in the variation of organic carbon levels across the different categories. This was expected because whereas only diesel exhaust is a source of elemental carbon, there are multiple sources of organic carbon. The organic carbon component of aerosol also varies significantly with engine duty cycle and other parameters.

In addition to the integrated elemental carbon/organic carbon measurements, we obtained real-time BC levels using an aethalometer. In the 2 maintenance garages, we placed the aethalometer alongside the filter elemental carbon/organic carbon samplers and ran them simultaneously over an entire work shift. In a plot of the elemental carbon levels versus the time-integrated aethalometer BC levels over each shift (Figure 8), the correlation between the 2 types of measurements, with an $R^2_{\text{adjusted}} = 0.69$, is

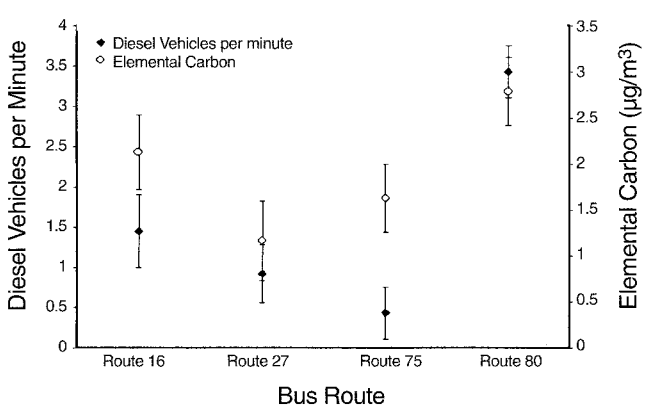


**Figure 6. Average elemental carbon exposures and average number of diesel vehicles per minute encountered along each bus route.**

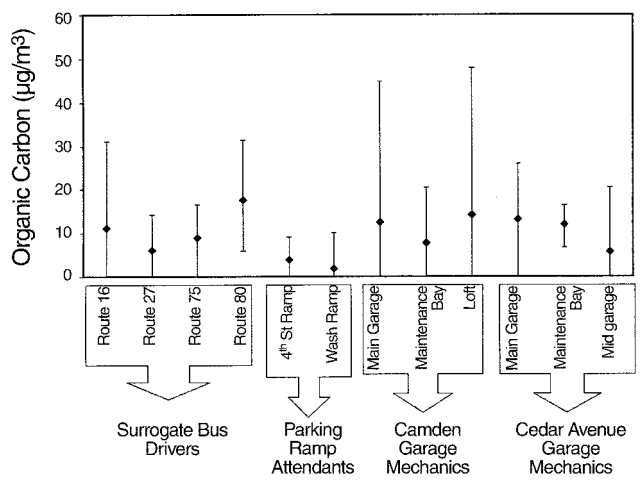


**Figure 7. Exposures to organic carbon for 3 occupational groups: surrogate bus drivers, parking ramp attendants, and garage mechanics.** The groups are divided into the different locations within each category where measurements were obtained. The error bars represent the range between the maximum and minimum values of the measurements.

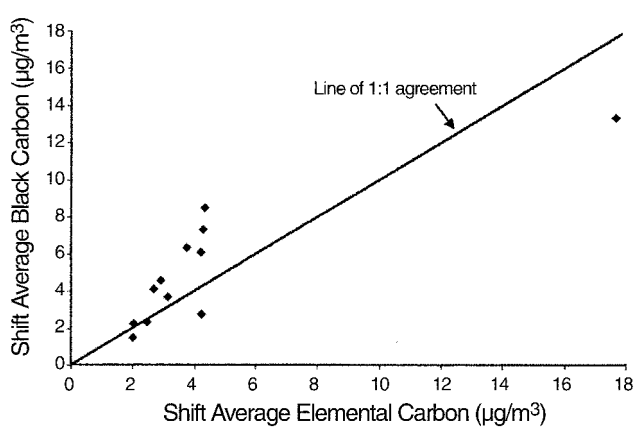


**Figure 8. Plot of average elemental carbon exposure concentrations versus time-integrated aethalometer black carbon exposure concentrations for garage mechanics during work shifts.** Each work shift ranged between 6 and 7 hours.

reasonably high. The slope of the linear regression fit is 0.67 with an intercept of 2.26.

The 10-minute average BC concentrations over 1 shift period (Figure 9) varied substantially within 1 day, reflecting the level of automobile traffic in the garage. For instance, the low levels in the early afternoon (approximately 1230 hours) may have been due to the decreased traffic during the lunch break. Figure 9 also shows that the BC concentration integrated over the entire shift is very close in value to the elemental carbon filter. Figures 8 and 9 provide useful validation of the comparability of aethalometer measurements to the more traditional filter measurements. Figure 9 also illustrates an important advantage of the real-time aethalometer: it can be used to measure concentrations much lower than 1 $\mu g/m^3$ (the MDL for elemental carbon measurements). In fact, concentrations as low as a few tens of nanograms per cubic meter can be routinely measured.

This capability is also nicely illustrated in Figure 10, which shows the cumulative frequency distribution of BC levels inside and outside a parking ramp booth. The median BC levels inside the booth were approximately 400 $ng/m^3$, a result consistent with the low elemental carbon levels in the parking ramps (see Figure 5). The maximum concentration inside the Washington Avenue booth was approximately 2,000 $ng/m^3$, while the maximum level outside the booth was approximately 6,000 $ng/m^3$, or 3-fold higher. The relatively higher level outside the booth suggests a source of elemental carbon, perhaps high-emitting gasoline engines.

## SURFACE AREA CONCENTRATION MEASUREMENTS

Two instruments, the DC and PAS, were used for measuring the surface area concentrations of aerosols in the 3 environments. Both portable and full-size DC and PAS units were used in the study. As mentioned earlier, the DC measures the total surface area of the particles irrespective of their chemistry, while the PAS measures the surface-adsorbed PAHs, a surrogate for the surface area of diesel exhaust aerosol specifically. Both these instruments were set to record a measurement every 10 seconds. The clocks of these instruments were synchronized so that they could record measurements simultaneously.

In a bar chart comparison of DC measurements in buses, parking ramps, and maintenance garages (Figure 11), the distributions of exposures in buses and parking ramps are not very different. The median exposure in buses was 40 $cm^2/m^3$ with a geometric standard deviation of 2.5, while the median exposure in the parking ramps was 30 $cm^2/m^3$, with a geometric standard deviation of 3.3.

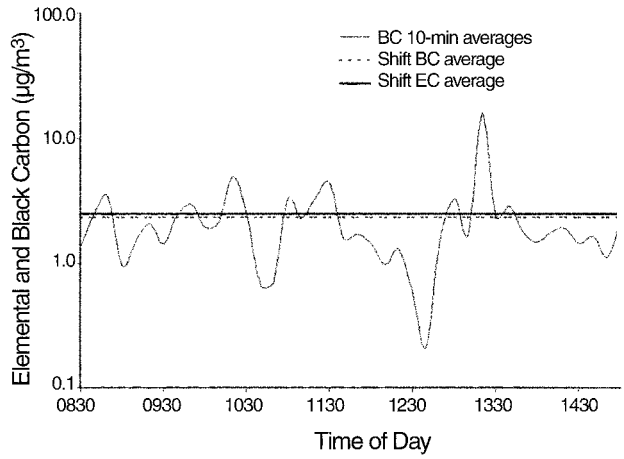


**Figure 9. Variation of 10-minute average black carbon concentrations measured by aethalometer over 1 work shift in the Camden maintenance garage.** Also shown are the time-weighted average EC concentration obtained by the cyclone filter sampler and the time-integrated average BC levels over the entire work shift.

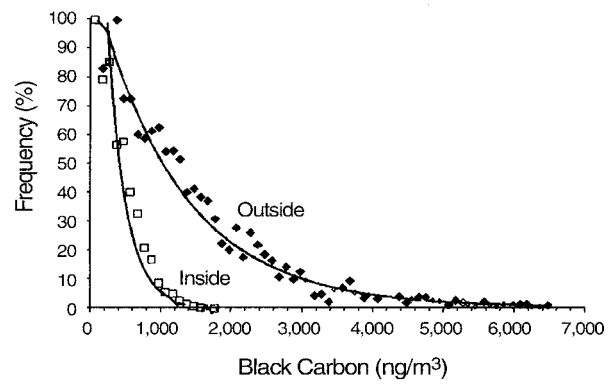


**Figure 10. Cumulative frequency distribution of black carbon levels inside and outside a Washington Avenue parking ramp booth as measured by aethalometer.**

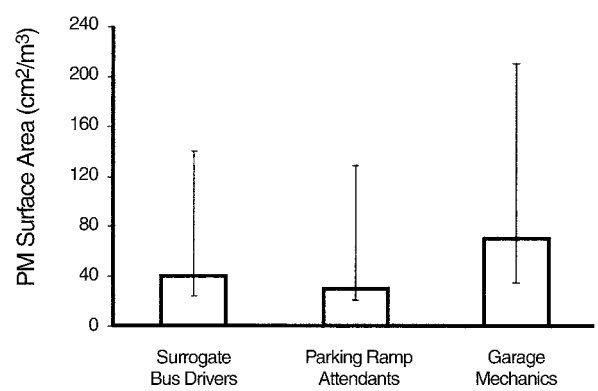


**Figure 11. Diffusion charger measurements of total surface area of particulate matter in buses, parking ramps, and maintenance garages.** A DC measurement was obtained every 10 seconds, and the cumulative distribution includes all data from every sampling day for each worker category.

However, the distribution of exposures in the maintenance garages was very different from the distributions of exposures in the buses and parking ramps, with a higher median of 70 $cm^2/m^3$ and a geometric standard deviation of 2.0. Although the median exposure in the parking ramps was the lowest, the large standard deviation resulted from a few occasions of very high exposure concentrations.

In a bar chart comparison of the PAS measurements in the buses, parking ramps, and maintenance garages (Figure 12), the parking ramps had the lowest median PAH exposure of 6 $ng/m^3$ with a geometric standard deviation of 3.3. The buses had a median PAH exposure of 15 $ng/m^3$ with a geometric standard deviation of 2.6. As with the DC measurements, the maintenance garages had the highest surface area exposures with a median PAH value of 40 $ng/m^3$ and a geometric standard deviation of 2.5. Thus, when surface area concentration was used as the exposure metric, the 3 work environments (and the workers in those environments) had distinct exposure profiles.

The PAS is sensitive to particles with surface-adsorbed PAHs and to the polycyclic nature of the soot component of diesel exhaust. Further, the DC provides the surface area of all aerosol particles (less than 100 nm). For these reasons, some have hypothesized that the ratio of measurements from the 2 instruments (PAS/DC) should give a measure of the diesel exhaust contribution to total aerosol in a mixed vehicular environment if gasoline exhaust and diesel exhaust are the only types of aerosol present (Siegmann et al 1999). Thus, the maintenance garage would have higher PAS/DC ratios than the bus and parking ramp environments. However, when we plotted the frequency distributions of the PAS/DC ratios in the buses, parking ramp booths, and maintenance garages, they were quite similar (Figure 13), with a mode of around 0.2. These findings show that the PAS/DC ratio was not a useful indicator of diesel exhaust aerosol in this study. It is possible that the concentrations of diesel aerosol were too low and not sufficiently different from background concentrations to make a clear footprint of diesel aerosol. Alternatively, the assumption that elemental carbon comes mainly from diesel engines may be flawed and off-design gasoline engines may be another source. Alone, however, each instrument is useful to the extent that it provides an easy way to measure surface area concentration rapidly and reliably. Furthermore, if particle surface area is linked to adverse health effects, then these instruments could provide important information on exposures regardless of the source.

Another interesting finding came from plotting all the 10-second average PAS and DC measurements obtained in the parking ramp booths against each other (Figure 14). A

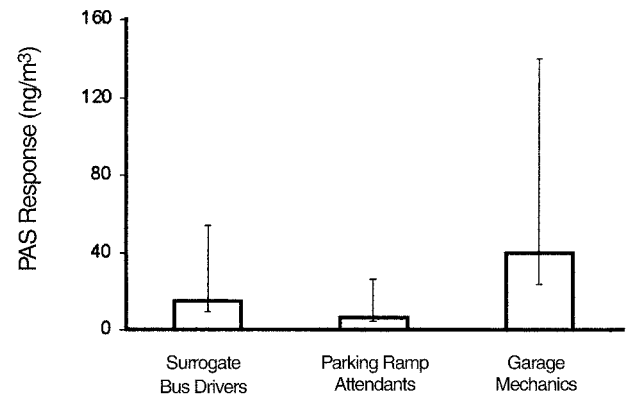


**Figure 12. Comparison of photoelectric aerosol sensor measurements of polycyclic aromatic hydrocarbons in buses, parking ramps, and maintenance garages.** A PAS measurement was obtained every 10 seconds, and the cumulative distribution includes all data from every sampling day for each worker category.

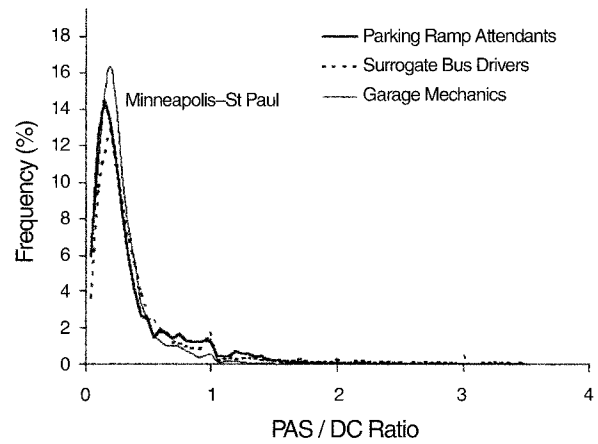


**Figure 13. Frequency distribution of ratio of simultaneous photoelectric aerosol sensor measurements and diffusion charger measurements in buses, parking ramps, and maintenance garages.** PAS and DC measurements were obtained every 10 seconds, and the frequency distribution includes all data points from every sampling day for each worker category.

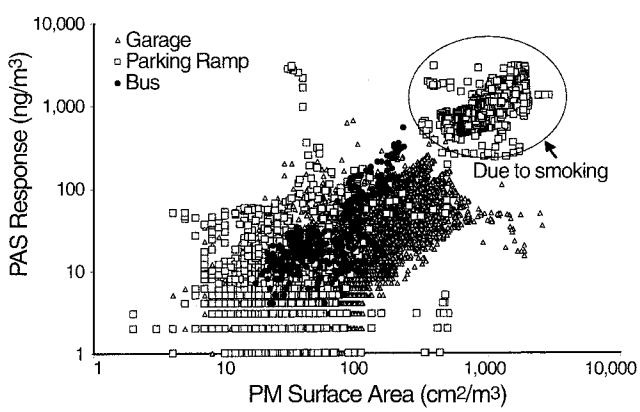


**Figure 14. Real-time simultaneous photoelectric aerosol sensor measurements of PAHs versus diffusion charger measurements yielding total surface area of particulate matter in parking ramp booths.** Note the effect of smoking events on the measurements.

cluster of data points with much higher PAS and DC measurements (shown within the circle in the top right-hand corner) resulted from 1 sampling day when the parking ramp attendants were smoking. Thus, these instruments are sensitive enough to detect the effects of isolated events, such as smoking, that have obvious epidemiologic use.

## NUMBER CONCENTRATION MEASUREMENTS

At selected sampling locations, particle number concentrations were measured using a UCPC (model 3025A, TSI) that provided total counts and an SMPS (model 3934, TSI) that provided size-segregated particle counts. The UCPC is quite rugged and does not require a lot of working knowledge to operate; however, the SMPS requires quite a bit of knowledge and experience to understand and operate.

An integrated number concentration was obtained from the SMPS size distribution by summing the number of particles counted in each scan channel. We compared the real-time UCPC number concentration and the integrated SMPS number concentration for each day of sampling in the Washington Avenue parking ramp (Figure 15). The UCPC data were averaged on a 1-minute time base to match the time base of the SMPS data and to reduce the number of data points. The measurements from the 2 instruments followed the same trend for each day of sampling even though the SMPS requires a sampling time of 60 seconds and the UCPC is a real-time instrument.

The samples from the last 2 periods shown in Figure 15 were taken outside the parking ramp booth. These concentrations were, on average, 4 to 5 times greater than concentrations inside the booth. A comparison of the steadier inside concentrations with the outside samples shows that variation in local particle concentrations was reduced by the integrating volume effect of the attendant's booth.

The UCPC concentrations were consistently higher than the SMPS concentrations and approximately 3.3 times higher (for example, SD = 1.1) for Feburary 28, 2000. Part of our routine calibration of the SMPS and the UCPC was to compare their responses to a polydisperse aerosol with a mean diameter of 50 to 60 nm and very few particles smaller than 10 nm. The instruments agreed very well with these calibration aerosols, so the difference in their results cannot have been due to a calibration error. Rather, the difference in the measurement ranges of the instruments explains the difference in their results. The UCPC was designed to detect particles as small as 3 nm, while the lower sizing limit of the SMPS in the operating mode used in this study was 8 nm. Therefore, particles between 3 and 8 nm in diameter would account for the difference in number count between the UCPC and the SMPS. The particle sources that were near the parking ramp, mainly spark-ignition engines, are associated with large numbers of extremely small particles.

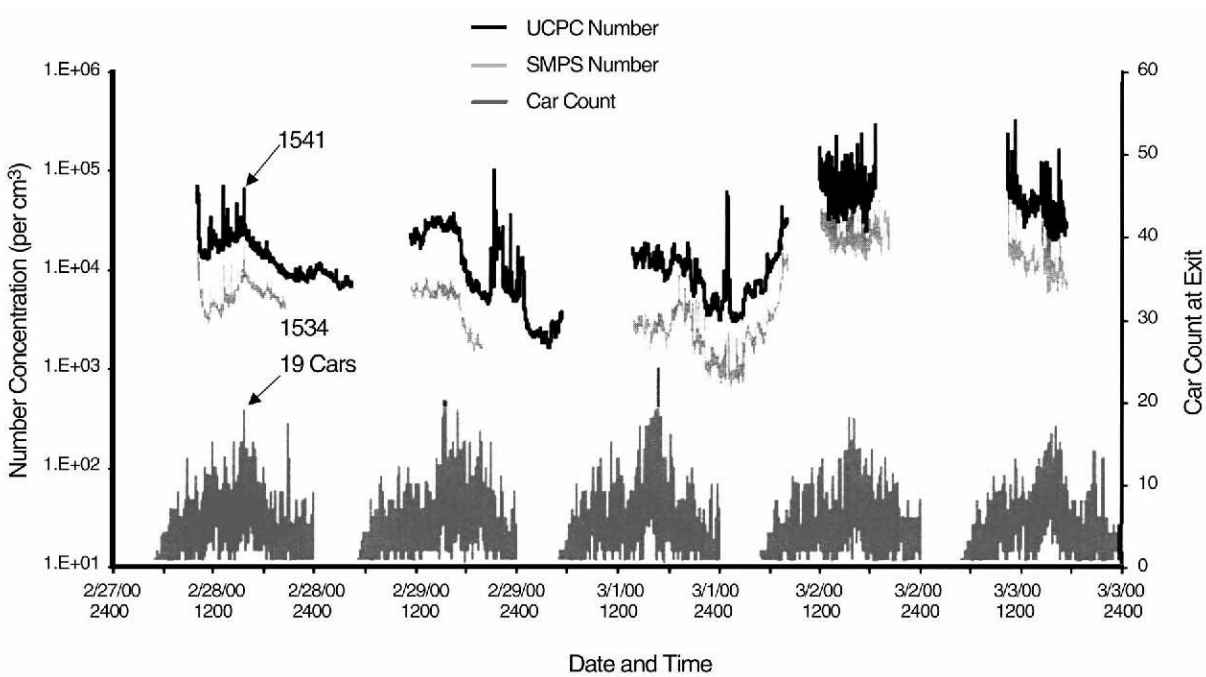


**Figure 15. Real-time number concentrations from ultrafine condensation particle counter and integrated number concentrations from scanning mobility particle sizer for each sampling day at Washington Avenue parking ramp.**

Figure 15 also shows the car counts for each sampling day. The car counts were determined from parking ramp transaction data, which recorded the time of each transaction at the 4 exit booths. The data for February 28 show a positive relationship between the car count peak and the peak particle count of the SMPS and UCPC. This shows that the change in characteristics of the aerosol samples was primarily a function of the traffic through the parking ramp.

In a plot of the integrated SMPS surface area concentration and the corresponding 1-minute average DC readings at the Washington Avenue parking ramp (Figure 16), all days show a close match between the measurements of the 2 instruments with the exception of the first half of February 28. This was most likely due to a system malfunction on the first day of sampling. Specifically, from 1200 hours on February 29 to 0800 hours on March 2, the measurements from the SMPS and DC tracked very well even with distinct variations in aerosol concentrations throughout this period. This is evidence that the DC could be used in place of the more expensive SMPS to obtain surface area particle concentrations in studies of ambient air quality. Like the UCPC, the DC has a response time on the order of 1 second. It is simple to use, requires little maintenance, and costs a fraction of an SMPS system, which makes it more likely to be used in exposure assessment studies.

Figure 17 shows a comparison between the SMPS-calculated geometric mean aerosol particle diameter, $D_g$, and data from the UCPC and DC. The $D_g$ was determined from an SMPS particle size distribution using the following relationship:

$$D_g = \exp\left(\frac{\sum n_i \ln D_i}{N}\right), \tag{8}$$

where $D_i$ and $n_i$ are the particle diameter and the number concentration of channel $i$, respectively, and $N$ is the total number concentration.

$$D_{as} = \sqrt{\frac{S}{\pi N}}, \tag{9}$$

where $S$ is the total surface area ($\mu m^2/cm^3$) measured by the DC and $N$ is the total number concentration (particles/$cm^3$) measured by the UCPC. Figure 17 shows how closely the calculated $D_{as}$ tracked with the $D_g$ of the SMPS. Along with Figure 17, Figure 18 is a plot of the average $D_{as}$ and $D_g$ for each sampling day at the Washington Avenue ramp. The average $D_{as}$ and $D_g$ for all sampling days were 44.4 nm (SD, 10) and 34.0 nm (SD, 5.3), respectively. These sizes are not expected to be the same. The $D_{as}$ should be larger because it is surface weighted and most of the surface

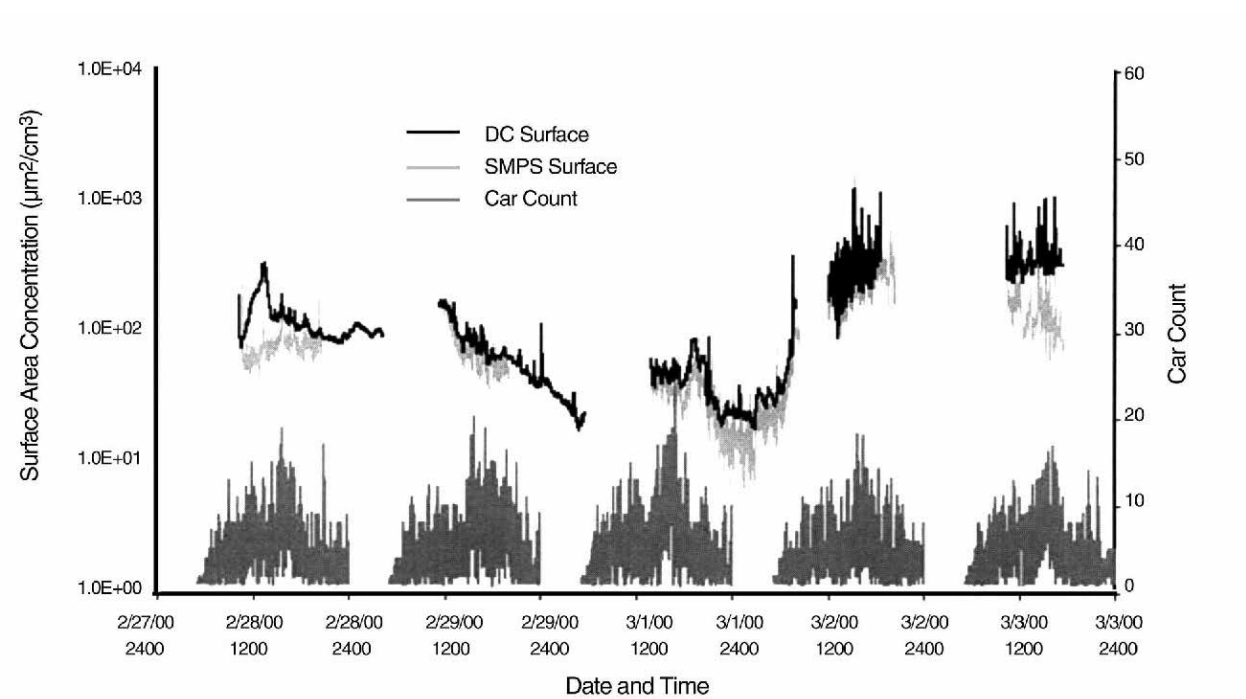


**Figure 16. Integrated scanning mobility particle sizer surface area concentrations and corresponding 1-minute average diffusion charger readings at Washington Avenue parking ramp.**

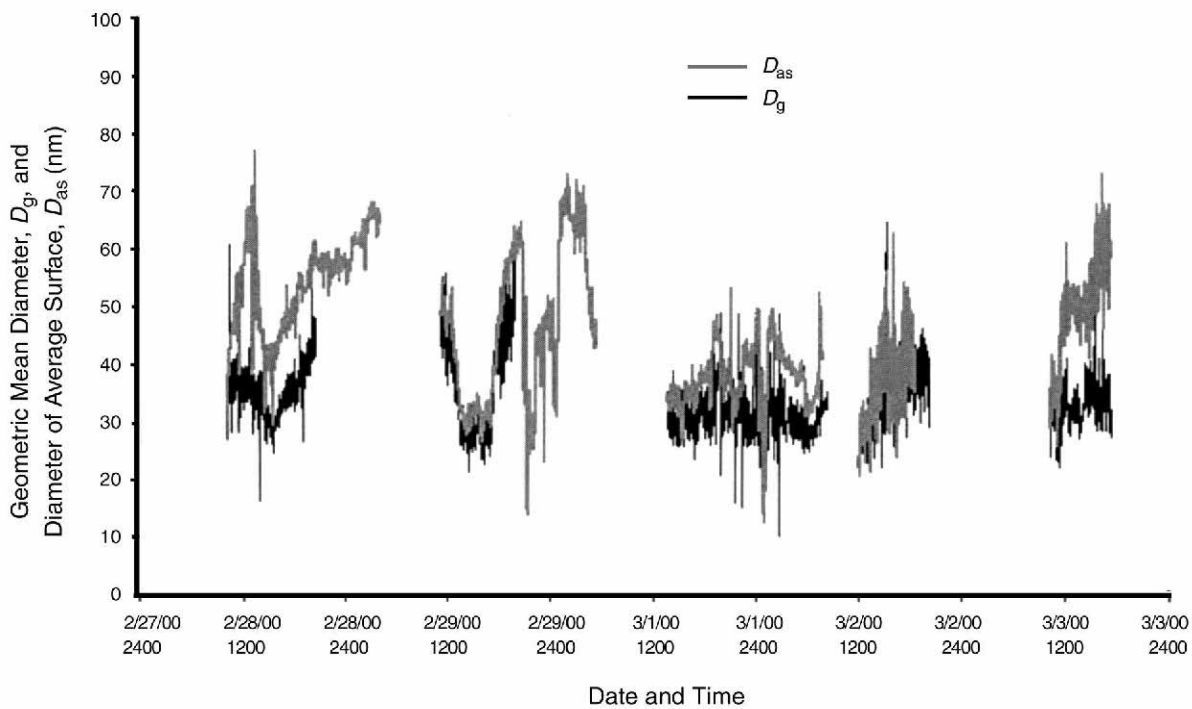


**Figure 17. Geometric mean particle diameter ($D_g$) calculated from scanning mobility particle sizer measurements, and average surface diameter ($D_{as}$) obtained from ultrafine condensation particle counter and diffusion charger measurements.**

area is found in the smaller particles. These results show that 2 completely different methods could be used to resolve particle size. Furthermore, not only do the DC and UCPC give real-time concentrations, but when combined, they also give a meaningful representation of the particle size in real time.

In a plot of the integrated SMPS number concentration for February 28 (Figure 19), the peak particle concentration occurred around 1600 hours, as did the peak car count

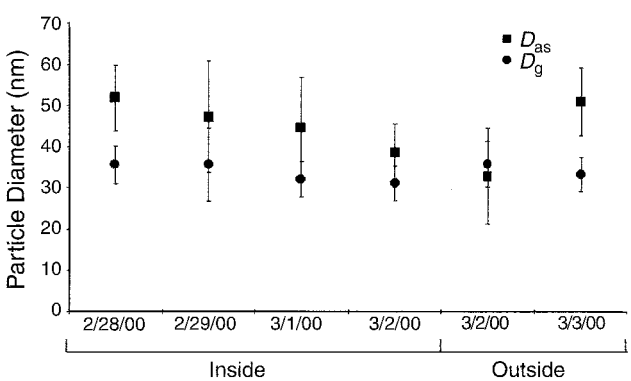


**Figure 18. Geometric mean particle diameter ($D_g$) and average surface diameter ($D_{as}$) for each sampling day at Washington Avenue parking ramp.**

(see Figure 15). In addition, the $D_g$ (plotted on the secondary axis) was observed to be 27.5 nm at peak particle concentration. Spikes in number concentration were generally associated with a decrease in particle size–most likely related to fresh combustion-generated particles, which are extremely small.

In a set of continuous data for March 2 with the PAS and DC concentrations plotted on the primary axis and the UCPC and aethalometer concentrations on the secondary axis (Figure 20), the instruments' responses did not seem strikingly similar. However, a 1-hour window of sampling on March 2 (Figure 21) showed more closely the similarity in instrument responses. Both plots show that number concentration (UCPC) has a much wider dynamic range than other measures of aerosol concentration. This is because number concentration is dominated by particles in the smallest size range. These particles have the shortest atmospheric residence times (Harrison 1996) Thus their concentration depends strongly upon the proximity of a local source.

Figure 22 represents a collection of continuous SMPS scans (570 scans) for 1018 to 2029 hours on February 28. Each window represents approximately 1 hour of scans, and each distribution is a 10-minute average of those scans. This plot shows the life cycle of an aerosol distribution inside the

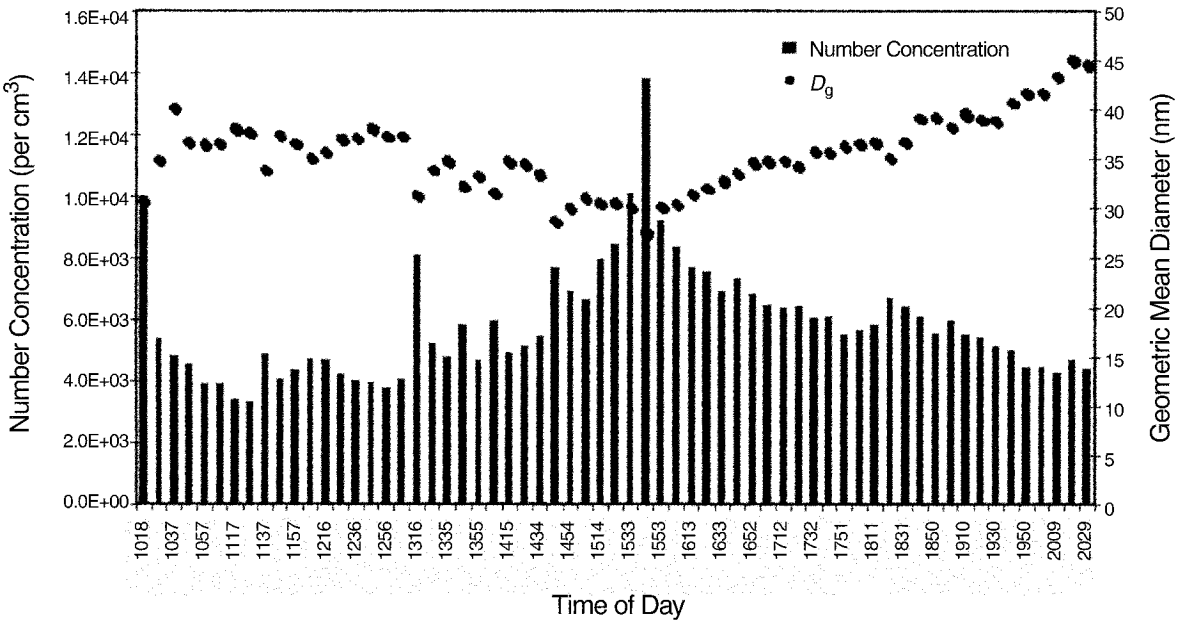


**Figure 19. Integrated scanning mobility particle sizer number concentration and geometric mean particle diameter ($D_g$) for 1 sampling day (February 28, 2000) at Washington Avenue parking ramp.**

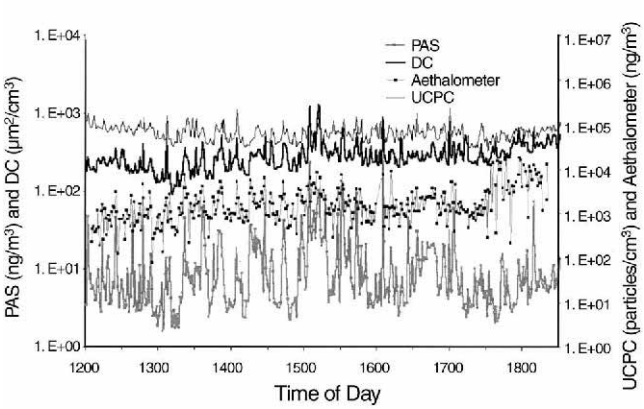


**Figure 20. Continuous measurements made by photoelectric aerosol sensor, diffusion charger, ultrafine condensation particle counter, and aethalometer over 1 sampling day (March 2, 2000) at the Washington Avenue ramp.**

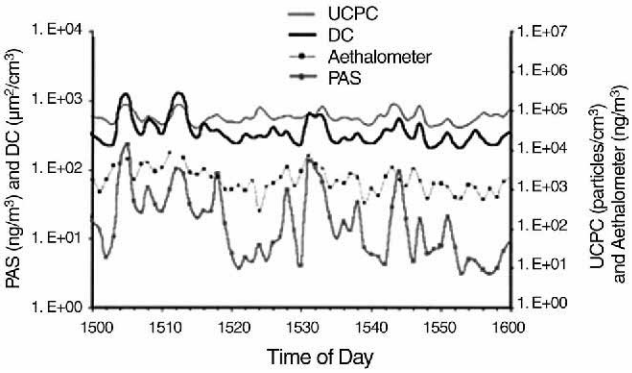


**Figure 21. Continuous measurements made by photoelectric aerosol sensor, diffusion charger, ultrafine condensation particle counter, and aethalometer over 1 hour on March 2, 2000.**

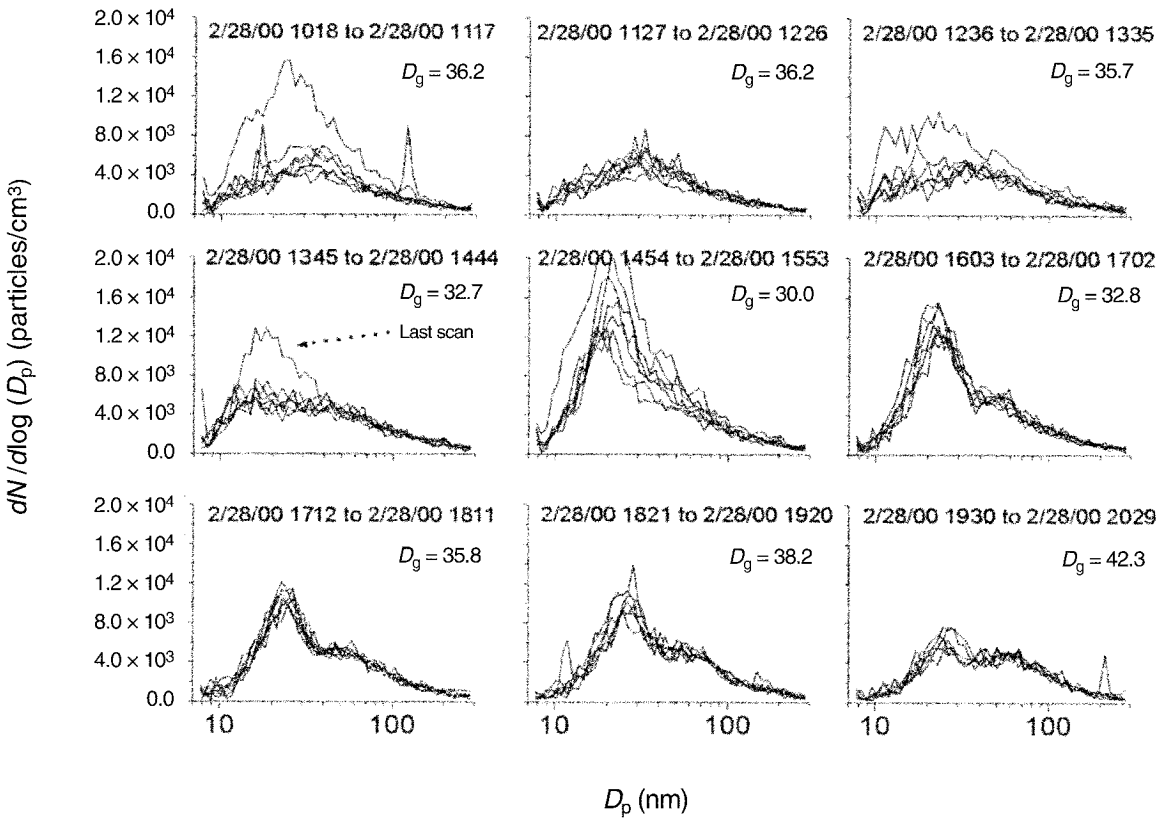


**Figure 22. Number-weighted particle size distribution functions measured by scanning mobility particle sizer over 1 day (February 28, 2000) at Washington Avenue parking ramp.** Each distribution is a 10-minute average of scans. (*d* denotes differential; *D*g is geometric mean particle diameter; *D*p is particle diameter.)

parking ramp attendant's booth. The top 3 windows show distributions representative of near-background distributions for that day. The next 3 windows show the beginnings of a bimodal distribution as the day progresses. As discussed earlier, bimodal distributions are typical of ambient aerosol containing combustion particles. The smaller-diameter mode, having a larger concentration, represent particles in the nuclei mode (approximately 15 to 25 nm), and the larger-diameter mode represents particles in the accumulation mode (approximately 40 to 100 nm or larger).

Figure 23 shows the average aerosol number concentrations inside and outside the attendant's booth at the Washington Avenue parking ramp and in the 2 maintenance garages. As expected, the number concentrations inside the booth (where the workers would be exposed) were much lower than those outside because the booth was under positive air pressure. The concentrations inside the booth ranged between $1 \times 10^4$ and $2.5 \times 10^4$ particles/cm$^3$. The concentrations outside the booth were similar to those measured in the 2 maintenance garages and ranged between $2.5 \times 10^4$ and $2.25 \times 10^5$ particles/cm$^3$. Thus, the exposures of the 2 corresponding cohorts (parking ramp attendants and garage mechanics) were different when measured in terms of distribution of particle number. No significant differences were noted between the exposure levels in the 2 maintenance garages.

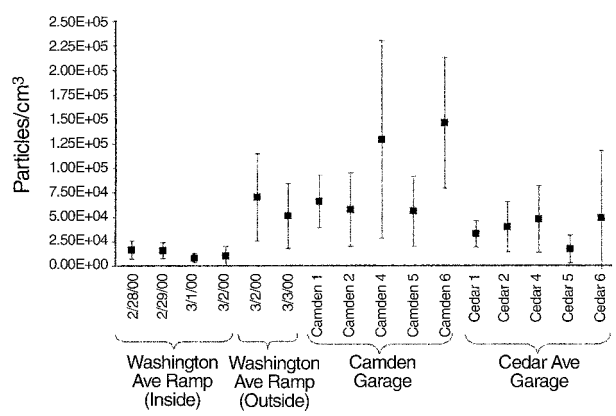


**Figure 23. Average aerosol number concentration inside and outside attendant's booth at Washington Avenue parking ramp and in Camden and Cedar Avenue maintenance garages.**

## DISCUSSION

Assessment of exposure to diesel exhaust aerosol (when not based on job titles or other surrogate measures) usually has been based on particulate mass concentration. The proposed threshold limit value of 0.15 mg/m$^3$ for diesel exhaust particles less than 1 µm in diameter (ACGIH 1998), as well as EPA regulations limiting diesel exhaust emissions and air quality standards for PM, were all based on mass measurements. However, this metric may not accurately reflect the full complexity of diesel exhaust exposure. Recent animal studies have suggested that particle number and surface area concentrations may be more relevant to health. Therefore, if size distribution data (number, volume, surface area, and mass) can be obtained, then lung deposition can be modeled in detail, and a better estimate of the received dose and potential health effects can be derived.

In this study, we evaluated the exposures of 3 occupational groups using both the traditional mass concentration of elemental carbon and an array of instruments designed to measure mass, surface area, and number concentration in real time. The measurements made by the different methods were consistent with each other, providing a useful validation of these methods. For instance, the time-integrated values of real-time aethalometer measurements compared well with the elemental carbon concentrations obtained using filter samples. Likewise, the surface area obtained directly using the DC compared well with surface area calculated using the detailed size distribution data from the SMPS. This finding is useful because it means the DC, which is a much less expensive and more rugged instrument than the SMPS, may be more appropriate for future epidemiologic studies. We also found that the DC and UCPC together gave both real-time concentrations and a meaningful representation of particle size in real time that were comparable to measurements made by the SMPS. This is another useful finding because the UCPC is also a rugged instrument that can be used routinely in field studies. The SMPS, in contrast, requires constant attention and highly trained field personnel. A comparison showed that number concentrations measured using the UCPC were generally higher than those measured using the SMPS because the detection threshold is 3 nm for the UCPC and 8 nm for the SMPS. The difference indicates that combustion sources, in this case spark-ignition engines, emitted large concentrations of extremely tiny particles that are not counted by the SMPS.

Animal studies and previous occupational exposure studies have typically been at high exposure levels. Rodent exposures that caused lung tumors have ranged from 1,000 to 10,000 µg/m$^3$, while occupational exposures have ranged between 10 and 1,000 µg/m$^3$ (HEI 1995). Therefore, these rodent studies have limited use in estimating human cancer risk due to ambient exposure. The 3 occupational groups in this study had exposures between 1 and 10 µg/m$^3$ (ie, at the low end of occupational exposures and near the high end of ambient exposures). This study shows that existing instruments can accurately measure these low exposures. For instance, the aethalometer can accurately measure mass concentrations that are much lower than the detection limit of more traditional elemental carbon measurements.

The current study also illustrates the advantages of using real-time instruments to measure exposure. The real-time measurements in this study helped quantify the effect of individual events on total time-integrated exposure. For example, exposures in the parking ramps closely tracked the traffic levels, and the instruments had sufficiently fine time resolution to pick out a smoking event during 1 sampling day. These capabilities would allow task-based exposure assessment in future occupational epidemiologic studies.

The 3 occupational groups in this study did not differ significantly in terms of their mass exposures to elemental carbon although the mean exposures were lowest for the parking ramp attendants, intermediate for the surrogate bus drivers, and highest for the garage mechanics. In terms of surface area concentrations, exposure for the garage mechanics was distinctly different from those of the surrogate bus drivers and the parking ramp attendants (whose exposure distributions were similar to each other). In terms of number concentrations, the parking ramp attendants had much lower exposures than the garage mechanics. Thus, depending on the exposure metric chosen, the 3 occupational groups had similar or different exposures. At this point, which exposure metric is the most relevant to human health is still unclear; however, it is clear that in future epidemiologic studies the choice of exposure metric will affect the classification of workers into similarly or differently exposed groups. These measurements did not reveal clear signatures that could be used to distinguish between diesel and gasoline engines. Both engine types operate on hydrocarbon fuels and may produce sufficiently similar exhaust aerosol signatures under some conditions to make unique source identification difficult.

## ACKNOWLEDGMENTS

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We thank the primary sponsors of the E-43 project, CRC and the Department of Energy's National Renewable Energy laboratory for their support. We also thank the cosponsors of the E-43 project, which include the Engine Manufacturers Association, Southcoast Air Quality Management District, the California Air Resources Board, Cummins Engine Company, Caterpillar Inc, Volvo Truck Corporation, and National Institute of Occupational Safety and Health (NIOSH).

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## ABOUT THE PRIMARY AUTHORS

**David B Kittelson** is a professor of mechanical engineering and director of the Center for Diesel Research at the University of Minnesota. He has BS and MS degrees in mechanical engineering from the University of Minnesota and received his PhD in chemical engineering from the University of Cambridge, England. Professor Kittelson has 30 years of experience conducting diesel exhaust aerosol studies and over 100 publications in the field. Most recently he led a team investigating diesel aerosol measurement methods for the EPA. Currently, he is leading an international group of investigators evaluating diesel aerosol sampling and measurement methods for CRC. The fundamental goal of the CRC research program is to develop methods to measure diesel aerosol in the laboratory that result in measurements that are similar to those obtained under roadway conditions. This entails a fundamental understanding of aerosol formation, transformation, and measurement as well as the processes that have a major effect on aerosol size distribution (such as dilution, nucleation, condensation, adsorption, and coagulation).

**Winthrop F Watts Jr** is a research associate at the Center for Diesel Research, University of Minnesota. He earned a BA in biology from Salem State College, an MS in public health from the University of Massachusetts, and a PhD in environmental health from the University of Minnesota. Dr Watts has 20 years of experience in industrial hygiene and, especially, diesel exposure measurements. Recently, he completed an investigation of the impact of a blended biodiesel fuel on diesel PM concentrations in an underground metal mine in Sudbury, Ontario. In 1996, he prepared the Diesel Toolbox for the Mine Safety and Health Administration. He was a member of the Health Effects Institute's Diesel Working Group and prepared the chapter on occupational exposure in this Report. During his tenure with the US Bureau of Mines, Dr Watts was involved in many diesel-related studies evaluating the effectiveness of ceramic filters, alternative fuels, and oxidation catalysts. He and Dr Kittelson are the principal investigators on the CRC-sponsored diesel aerosol measurement research program. He has been actively involved in all phases of that research program that includes on-highway and test cell aerosol measurement.

**Gurumurthy Ramachandran** is an associate professor in the Division of Environmental and Occupational Health in the School of Public Health at the University of Minnesota. He earned a bachelor's degree in electrical engineering from the Indian Institute of Technology and an MS in environmental engineering from Virginia Polytechnic Institute and State University. He received his PhD in environmental sciences and engineering from the University of North Carolina, Chapel Hill. Professor Ramachandran's research interests are in exposure assessment methodology, especially for aerosol exposures, as well as aerosol sampling issues. He recently led a NIOSH-funded study for developing retrospective exposure assessment methods for nickel aerosols in a primary nickel smelting facility. He also has extensive experience in developing algorithms for obtaining aerosol size distributions from aerosol spectrometers. He is a coinvestigator for an EPA-funded project on measurement and source apportionment of human exposures to hazardous air pollutants (volatile organic compounds) and $PM_{2.5}$ in the Minneapolis–St Paul metropolitan area. One of the objectives of this study is to measure various subfractions of fine particles such as $PM_{2.5}$ (personal exposures and area concentrations), ultrafine particle count (using a condensation nuclei counter), size distributions using a micro-orifice uniform deposit impactor (MOUDI), and real-time $PM_{2.5}$ mass concentrations using a DustTrak. Currently, Professor Ramachandran is a member of the Exposure Assessment Strategies Committee of the American Industrial Hygiene Association.

## ABBREVIATIONS AND OTHER TERMS

| | |
|---|---|
| ACGIH | American Conference of Governmental Industrial Hygienists |
| BC | black carbon |
| CRC | Coordinating Research Council |

| | |
|---|---|
| $D_{as}$ | diameter of average surface area |
| $D_g$ | geometric mean diameter |
| $D_p$ | particle diameter |
| DC | diffusion charger |
| EC | elemental carbon |
| EPA | US Environmental Protection Agency |
| LFE | laminar flow element |
| $N/V$ | ratio of particle number concentration to PM volume concentration |
| NIOSH | US National Institute for Occupational Safety and Health |
| OC | organic carbon |
| PAH | polycyclic aromatic hydrocarbon |
| PAS | photoelectric aerosol sensor |
| PM | particulate matter |
| $PM_{2.5}$ | particulate matter with an aerodynamic diameter of 2.5 µm or less |
| $PM_{10}$ | particulate matter with an aerodynamic diameter of 10 µm or less |
| SMPS | scanning mobility particle sizer |
| $TiO_2$ | titanium dioxide |
| UCPC | ultrafine condensation particle counter |

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# Investigators' Report

## Measuring Diesel Emissions Exposure in Underground Mines: A Feasibility Study

Barbara Zielinska, John C Sagebiel, Jake McDonald, C Fred Rogers, Eric Fujita, Pierre Mousset-Jones, and James E Woodrow

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# Measuring Diesel Emissions Exposure in Underground Mines: A Feasibility Study

Barbara Zielinska, John C Sagebiel, Jake McDonald, C Fred Rogers, Eric Fujita, Pierre Mousset-Jones, and James E Woodrow

## ABSTRACT

The overall objective of this project was to develop a method to accurately quantify the exposure of underground miners to particulate matter (PM*) in diesel exhaust, respirable dust, and oil mist. To establish mine-specific chemical profiles, detailed chemical analyses were performed on collected samples of diesel exhaust, drilling and lubricating oil, and 2 ores from an underground gold mine in Nevada as possible aerosol sources of exposure. Next, underground ambient air samples were collected for 3 consecutive shifts from June 4 to June 6, 1999. These samples consisted of PM less than 2.5 µm in aerodynamic diameter ($PM_{2.5}$) for mass and inorganic species as well as particles and semivolatile organic compounds obtained by a filter backed up with an adsorbent trap. In addition, 2 micro-orifice uniform deposit impactor (MOUDI) samplers were deployed at one site to determine the chemistry of size-fractionated samples. At the same time that ambient samples were being obtained, battery-powered personal samplers with $PM_{2.5}$ inlets were sent with 6 miners per 12-hour shift.

The source samples of emissions from diesel vehicles were dominated by carbon with an average of approximately 40% elemental carbon (EC) and 60% organic carbon (OC), as determined by the thermo-optical reflectance (TOR) method. The MOUDI distributions of emissions samples from the diesel vehicles showed large variability but generally had the greatest mass of particles in the nuclei mode (< 50 nm) and in the accumulation mode (approximately 150 nm).

The ambient samples in the mine showed $PM_{2.5}$ concentrations of 400 to 1,100 µg/m$^3$ with higher concentrations near the mining operations. Ambient samples were composed of 72% carbon (75% EC and 25% OC). Ambient MOUDI distributions were bimodal with peaks for the largest particles and the accumulation mode particles and low levels of particle mass in the nuclei mode. Chemical analysis of the MOUDI samples showed the geologic signature elements (silicon, iron, and aluminum) in large particles and carbon in accumulation and nuclei mode particles.

The personal samples had $PM_{2.5}$ concentrations of 550 to 2,700 µg/m$^3$ with higher concentrations associated with jobs in active mining areas. Carbon accounted for approximately 68% of the mass in personal samplers.

Chemical mass balance (CMB) modeling was used to apportion the sources. In the ambient mine samples, the diesel emissions source signature accounted for 72% to 94% of the mass, while ore accounted for 2.2% to 14%. These values suggest that the miners were exposed to concentrations of diesel PM significantly higher than the current proposed standard for exposure.

## INTRODUCTION

Atmospheric fine particulate matter ($PM_{2.5}$) has been associated with human health effects. Studies on the epidemiology of this relationship (Dockery et al 1993; Heath et al 1995; Pope et al 1995 a,b; Schwartz et al 1996) and possible causal mechanisms (Seaton et al 1995) have been reported, and the controversy that surrounds the debate about PM and health effects has been discussed (Vedal 1997). Important sources of fine particles include mobile sources (especially diesel vehicles, with emissions dominated by particles smaller than $PM_{2.5}$), power generation, oil combustion, and geologic material.

Diesel exhaust is a complex mixture containing thousands of organic and inorganic compounds, in both particulate and gaseous phases, that are readily inhaled. Because some of these compounds, such as benzene and some polycyclic aromatic hydrocarbons (PAHs) and nitro-PAHs, are known

* A list of abbreviations and other terms appears at the end of this Investigators' Report.

This Investigators' Report is one section of Health Effects Institute Special Report *Research Directions to Improve Estimates of Human Exposure and Risk from Diesel Exhaust*, which also includes a report by the HEI Diesel Epidemiology Working Group, four other Investigators' Reports, and an Executive Summary. Correspondence concerning this Investigators' Report may be addressed to Dr Barbara Zielinska, Desert Research Institute, 2215 Raggio Parkway, Reno NV 89512-1095.

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human carcinogens, widespread use of diesel engines has raised concerns about the potential health effects of their emissions. The major concern has been the potential for producing lung cancer although diesel exhausts may also contribute to other health problems. Diesel exhaust has been classified as a possible human carcinogen by the US National Institute for Occupational Safety and Health (NIOSH 1988), as a probable carcinogen by the International Agency for Research on Cancer (IARC 1989), and as a toxic air contaminant by the California Air Resource Board (California Environmental Protection Agency 1998).

Outdoor exposure to diesel PM less than 10 µm in aerodynamic diameter ($PM_{10}$) has been measured at 0.2 µg/m$^3$ in relatively pristine remote locations to 3.6 µg/m$^3$ in smog-impacted airsheds such as California's South Coast Air Basin (California Environmental Protection Agency 1998). Near roadways, concentrations of diesel $PM_{10}$ have been as high as 8 µg/m$^3$. Industrial surveys of miners, forklift operators, truck drivers, and railroad workers have found daily exposures to diesel PM of 4 to 1,700 µg/m$^3$ with mine workers in enclosed spaces at the highest concentrations (NIOSH 1988).

Although authorities agree that exposure to diesel exhaust is hazardous, their estimates of the potency or unit risk vary. In part, this variation results from the lack of clearcut cause-and-effect epidemiology with measures of exposure and health outcomes; from inconsistencies as to whether confounding factors (such as cigarette smoking) were clearly addressed; and from the paucity of quantitative exposure measurements needed to clarify the dose-response relationship (California Environmental Protection Agency 1998; Zaebst et al 1991). On the basis of human data, the California Environmental Protection Agency (1998) proposed a range of lifetime cancer unit risk (95% upper confidence limit) from $1.3 \times 10^{-4}$ to $1.5 \times 10^{-3}$ per µg/m$^3$ with a mean of $4 \times 10^{-4}$ per µg/m$^3$. This upper mean estimate corresponds to an increase of 4,000 cancers per million people exposed over a lifetime to 10 µg/m$^3$ of diesel exhaust particles in the air that they breathe. Such estimates have also been made for noncancer health effects from breathing diesel exhaust: respiratory effects such as asthma as well as reproductive, developmental, and immunologic effects (California Environmental Protection Agency 1998). Although cancer end points drive the human health risk assessments, the noncancer effects can be very important, particularly in high-exposure situations (such as may exist in mines) and in sensitive subpopulations. California's Office of Environmental Health Hazard Assessment (OEHHA) has supported 5 µg/m$^3$ as the chronic inhalation reference exposure level (California Environmental Protection Agency 1998).

On October 29, 1998, the US Department of Labor's Mine Safety and Health Administration (MSHA) proposed new health standards for underground metal and nonmetal (or noncoal) mines that use diesel equipment (MSHA 1998). The rule established a concentration limit that would be phased in over a 5-year period. An interim limit of 400 µg/m$^3$ of total carbon (TC) would go into effect after 18 months of MSHA education and technical assistance to mine operators. A final limit of 160 µg/m$^3$ would become effective in 5 years. TC would be determined using NIOSH method 5040 (NIOSH 1999). Surveys of some mines showed that the workers' exposures to diesel PM significantly exceeded (on the order of a few hundred percent) proposed health standards. However, the contributions of potential sources to worker exposure are not clear. Reliable methods for measuring human exposure to diesel emissions and attributing source contributions are needed in order to develop effective approaches to attain the new standards.

Currently, 3 methods of sampling and analysis of diesel PM in metal and nonmetal mines are available: respirable combustible dust sampling and analysis, size-selective sampling with EC or gravimetric analysis, and respirable dust sampling with EC analysis. Although each provides information concerning diesel PM, none measures diesel PM directly (Cantrell and Watts 1997). The respirable combustible dust method (used in Canadian noncoal mines) estimates diesel PM concentrations from the amount of combustible material (at 500°C) in a respirable dust sample. To account for unburned fuel, evaporated lubricating oil, and other carbonaceous material, the amount of measured combustible dust is multiplied by an empirical correction factor of 0.67. This method is not precise and is not suitable for coal mines or other environments where nondiesel carbonaceous aerosol is present.

Size-selective sampling takes advantage of the difference in aerodynamic diameter between combustion-generated and mechanically generated aerosol (the mass median diameter for diesel aerosol is approximately 0.2 µm, with 90% of the particles smaller than 1.0 µm). Usually, the inertial impaction method is used (McCartney and Cantrell 1992) to collect respirable dust. This sampler collects particles in 2 stages: those larger than 0.8 µm (mostly of nondiesel origin) and those 0.8 µm or smaller (predominately diesel). Gravimetric analysis is used to determine the mass concentration in each size fraction. This method does not address the presence of oil mist and is not very precise for lower concentrations of diesel PM or for submicron aerosol from multiple sources (Cantrell and Watts 1997).

The combination of size-selective sampling with other analytic techniques, such as EC measurements, can provide

more accurate chemical information for suspended diesel PM. EC generally accounts for 40% to 60% of diesel PM mass and is detectable at low levels using size-selective sampling and TOR analysis (Chow et al 1993) or the thermo-optical transmittance or EC method (method 5040, NIOSH 1999). The EC/OC ratio in diesel PM depends on a number of factors (eg, fuel, engine type, duty cycle, engine maintenance, lubricating oil consumption, and use of emission control devices). Variations in EC/OC ratios can result in inaccurate estimates of the total diesel PM present (Cantrell and Watts 1997). Thus, in a proposed health standard for underground metal and nonmetal mines, MSHA (1998) used TC as a measure of exposure to diesel emissions, but other sources of OC may be present in mines. Therefore, a method is needed to evaluate TC analysis as a technique for estimating diesel PM exposure. This method also should allow for quantification of the contribution to PM from oil mist and from other sources of mine aerosol.

## SPECIFIC AIMS

The overall objective of this project was to develop a method to precisely apportion diesel PM, respirable dust, oil mist concentrations, and other contaminants to which workers were exposed in an underground gold mine. The specific aims were as follows:

1. characterize chemical composition and particle size of organic and inorganic contaminants from specific sources and in ambient air in an underground gold mine;
2. apportion the contribution of specific sources (diesel equipment, mechanical particle generation, oil mist, cigarette smoke, etc) to the total airborne contaminant load in the mine; and
3. estimate exposures for mine workers from these sources.

In ambient air studies, receptor modeling is used to identify the sources of airborne pollutants and to determine their contributions to ambient aerosol mass concentrations, gaseous and particulate species concentrations, and light extinction. The CMB model is the most widely used receptor model for apportioning sources of ambient $PM_{10}$ (Watson et al 1990a,b). The CMB model compares the chemical compositions of source emissions, known as *source profiles*, with those of ambient aerosol. Inorganic and organic constituents, including trace elements, sulfate ($SO_4^{2-}$), nitrate ($NO_3^-$), ammonium, and total particulate OC and EC, are typically measured in $PM_{10}$ source apportionment studies. The source profile species and the receptor concentrations, each with uncertainty estimates, serve as input data to the CMB model. The model's output data consist of the contributions for each source type to the total aerosol mass as well as to individual species in the source profile and the ambient aerosol profile. The model calculates values for contributions from each source and the uncertainties of those values. Input data uncertainties are used both to weight the relative importance of the input data to the model solution and to estimate uncertainties of the source contributions.

We postulated that CMB modeling could be used in underground mines to provide precise estimates of diesel PM, oil mist concentrations, and other sources of contaminants. To prove this assumption, we tested the following hypotheses:

Distinct chemical profiles can be established for emissions from diesel equipment used in the mines; for drilling and lubricating oil mist; for PM mechanically generated from raw mining material; and for other sources of aerosol that may be present in the mines (including cigarette smoke).

1. These profiles can be based on specific organic compounds (particularly PAHs, hopanes, and steranes), trace elements, OC and EC, particle sizes, and inorganic ions emitted by each source type.
2. These profiles are sufficiently different from one another that the CMB model can be used to apportion aerosols present in mines to their respective sources.
3. Similar chemical information can be obtained with medium-volume, fixed-site samplers and with low-volume, portable, personal samplers used for personal exposure measurements.
4. This report describes the experiments performed in order to test these hypotheses and provides the results of CMB modeling for one underground gold mine.

## METHODS

### SAMPLING STRATEGY AND SAMPLING METHODS

Sampling at the Newmont Mining Corporation's Carlin East gold mine was performed from June 1 to June 6, 1999. First, the representative samples of emissions from aerosol sources present in the mine were collected (including exhaust from diesel equipment and samples from drilling and lubricating oil, diesel fuel, and mineral or rock dust). Next, to apportion aerosol in the mine to its respective sources, the mine atmosphere was assessed using different size-selective samplers. Concomitant with ambient sampling, exposure of miners was measured using personal samplers.

**Source Sampling**

Exhaust from diesel equipment used in the mine was sampled in the aboveground equipment maintenance shop. The vehicles used for source testing were chosen to span a range of engine and vehicle types commonly used in underground mines. Table 1 lists the vehicles tested along with a brief description of the vehicles and some information on the engines. Replicate tests were run on 2 vehicles, ULD008 and ULD024. Power ratings are the manufacturer's specifications, typically at 1,800 or 2,000 rpm. The engine list covers 3 manufacturers and sizes from 1.5-L up to 12.7-L displacement. The engines were turbocharged except for the Kubota. All engines except the Caterpillar and Kubota had electronic controls that allowed us to collect engine performance data during the test runs. Among the vehicles tested were 2 examples of Detroit 50 Series engines, 3 different Detroit 60 Series engines, and 3 different Detroit 4-71TI engines. The opportunity to examine different individual engines allowed us to assess the variability within an engine class as well.

Each vehicle was run through a test cycle consisting of an idle period followed by a period with load to near 100% of the engine rating. This was accomplished by using the vehicle's onboard power takeoff system because use of a dynamometer was not an option for these vehicles. The cycle consisted of 26 minutes divided as 5 minutes idle, 2 minutes load, 5 minutes idle, 2 minutes load, 5 minutes idle, 2 minutes load, and 5 minutes idle.

This cycle provided approximately equal amounts of fuel consumption during the 20 minutes of idle phases as during the 6 minutes of load phases. The test cycle was outlined specifically for this project and was intended not to exactly simulate the driving conditions inside the mine, but rather to give a reasonable distribution of emissions. During the load phases of the test, the engines were loaded and not simply on high idle.

The electronic engine controls store considerable amounts of information, and we were able to obtain this information for most of the vehicles tested. The most critical information was the engine load and the turbocharger boost. Some of the other information collected included rated horsepower (hp), rated revolutions per minute (rpm), engine hours, fuel usage, battery volts, engine oil temperature (°C), ambient air temperature (°C), coolant temperature (°C), idle set (rpm), accelerator (%), engine (rpm), boost pounds per square inch (psi), barometric pressure (psi), torque limit (%), engine load (%), injector control (psi), fuel (gal/hr), transshaft (rpm), cool level, and engine oil pressure (psi).

At the temperatures found in the exhaust of diesel vehicles, many organic compounds that would be present in the aerosol phase under ambient conditions are still in the gas phase; hence, they are not captured by methods that

**Table 1.** Diesel Vehicles Tested, Engine Types, and Power Ratings

| Vehicle ID[a] | Description | Engine[b] | Power (hp) | Displacement (L) | Used in Apportionment |
|---|---|---|---|---|---|
| ULD004 | Loader[c], 6 yard | Detroit 50 Series DDEC | 250 | 8.5 | N |
| ULD005 | Loader, 6 yard | Detroit 50 Series DDEC | 250 | 8.5 | N |
| ULD008 | Loader, 3½ yard | Detroit 4-71TI DDEC | 180 | 4.65 | N |
| ULD008R | Loader, 3½ yard | Detroit 4-71TI DDEC | 180 | 4.65 | Y |
| ULD024 | Loader, 6 yard | Caterpillar 3306 DITA | 275 | 10.5 | Y |
| ULD024R | Loader, 6 yard | Caterpillar 3306 DITA | 275 | 10.5 | Y |
| UHT002 | Truck, 16 ton | Detroit 4-71TI DDEC | 200 | 7.6 | Y |
| UHT005 | Truck, 16 ton | Detroit 4-71TI DDEC | 200 | 8.7 | Y |
| UHT006 | Truck, 26 ton | Detroit 60 Series DDEC | 375 | 12.7 | Y |
| UHT022 | Truck, 26 ton | Detroit 60 Series DDEC | 335 | 12.7 | Y |
| UHT024 | Truck, 26 ton | Detroit 60 Series DDEC | 335 | 12.7 | Y |
| UTR009 | Boss Buggy[d] | Kubota D1503-L-A | 32 | 1.5 | Y |

[a] An "R" after the identification indicates the test run was a replicate.

[b] All engines were turbocharged except for the Kubota D1503-L-A.

[c] This loader is similar to front-end loader: scrapes dirt from ground into trucks.

[d] Boss Buggy is a compact 4-wheel drive tractor.

directly filter particles from the hot exhaust phases (Hildemann et al 1989). In order to simulate the cooling and dilution processes that occur in a plume downwind of a combustion source, Desert Research Institute (DRI) designed a dilution stack sampler to collect fine particulate organic aerosol from combustion sources. The dilution stack sampler is a slightly modified version of the model built and described by Hildemann and associates (1989). Modifications include improvements in portability and a longer mixing chamber (17 effective diameters compared with 10), which provides more mixing time to ensure uniform sample distribution. Emissions were withdrawn from the exhaust through a copper sample line (0.5 in × 8 ft) that was heated to a temperature (150°C) appropriate for avoiding any condensation of stack gas. In the tunnel, the exhaust was mixed with dilution air under turbulent flow to cool and dilute it to near-ambient conditions. Dilution air consisted of ambient air that had been filtered through a high-efficiency particulate air (HEPA) filter (to remove PM) and an activated carbon bed (to remove gas-phase organics). The dilution rate was calculated as the ratio of dilution air to sample line air in the dilution system. The amount of dilution air was calculated directly from the measured differential pressure of air flowing through a high-volume pump. Sample line flow was calculated using a Venturi (a calibrated orifice used to measure flow; Lambda Square, Bay Shore NY) connected to a magnahelic gauge (0–5 inches). Lambda Square conducted calibrations for the sample line temperatures that were used during the testing because the Venturi flow depends on temperature.

After passing through a tunnel length equal to approximately 17 tunnel diameters, a fraction of the diluted exhaust entered a large chamber, where it resided until the samples were collected. Residence time for the operating parameters used was calculated at approximately 80 seconds. At the bottom of this chamber, the samples were drawn through Bendix 240 cyclones located inside the dilution sampler at 113 L/min to remove particles larger than 2.5 µm.

Two DRI fine particle/semivolatile organic compound (FP/SVOC) samplers were used to collect all samples to be analyzed for organic compounds. To account for the total ambient concentrations of semivolatile organic compounds (such as PAHs, which are distributed between gas phase and particle phase), use of a filter followed by a backup solid adsorbent was necessary. For collection of semivolatile PAHs (Zielinska et al 1998) in this study, we used Teflon-impregnated glass-fiber (TIGF) filters of 10-cm diameter, followed by cartridges of polystyrene-divinylbenzene resins (XAD-4) between polyurethane foam (PUF) plugs (referred to as *PUF/XAD-4/PUF cartridges*). The flow was set to 113 L/min using a calibrated rotameter and checked after the sampling was concluded.

Before sampling, XAD-4 resin was Soxhlet-extracted with methanol and then with dichloromethane ($CH_2Cl_2$), each for 8 hours. The cleaned resin was dried in a vacuum oven heated to 40°C and stored in sealed glass containers in a clean freezer. The PUF plugs were Soxhlet-extracted with 10% diethyl ether in hexane, followed by acetone. The TIGF filters were cleaned by sonification in dichloromethane for 30 minutes followed by another 30-minute sonification in methanol. Then they were dried, placed in aluminum foil, and labeled. Each batch of precleaned XAD-4 and approximately 10% of precleaned TIGF filters and PUF plugs were checked for purity by solvent extraction and gas chromatography/mass spectrometry (GC/MS) analysis of the extracts. The PUF plugs and XAD-4 were assembled into glass cartridges (10 g of XAD-4 between 2 PUF plugs), wrapped in aluminum foil, and stored in a clean freezer before shipment to the field.

Inorganic species were collected from the dilution sampler using a Teflon filter, a quartz filter, and a medium-volume DRI fine particulate sampler. The air sample was drawn from the DRI dilution tunnel through a cyclone separator with a cutoff diameter of 2.5 µm operating at 113 L/min. Downstream of the cyclone, a half-inch copper manifold led to a momentum diffuser chamber. At the bottom of the chamber, 2 cassette cartridge holders, one each for the Teflon filter and the quartz filter, were connected to a vacuum pump through a solenoid valve, a ball valve, and a flow controller. An independent elapsed-time meter recorded the sampling time. The flow was set at 20 L/min for each filter, and makeup flow was set at 73 L/min using a calibrated rotameter on the inlet side of each filter holder line. Thus, the total flow was maintained at 113 L/min during sampling.

In addition, we used 2 MOUDI samplers (MSP Corporation, Minneapolis MN), which provided size-resolved gravimetric and chemical data: one sampler was fitted with Teflon media for mass and elemental analyses; the other had aluminum media for TOR carbon analyses. The MOUDI is an inertial cascade impactor with multiple nozzles. At each stage, jets of particle-laden air impinge on an impaction plate, and particles larger than the cut size of each stage are collected on the impaction plates. Smaller particles with less inertia follow the air streamlines and proceed to the next stage. The nozzles of each succeeding stage are smaller than those of the previous stage, giving a higher velocity through the nozzles and a smaller particle size cut. The airflow continues through a series of 8 impactor stages until the smallest particles are removed by the afterfilter. In the configuration we used, the top stage

(which would nominally be 5.6 µm) was greased with high-vacuum grease to collect any larger particles. The stages at 3.16, 1.78, 1.00, 0.54, 0.37, 0.148, 0.105, and 0.054 µm were used in the normal manner, with the afterfilter collecting particles smaller than 0.054 µm. Each MOUDI operated at a flow rate of approximately 30 L/min.

Carbon dioxide was monitored in the diluted exhaust by a nondispersive infrared gas analyzer (model VIA-510, Horiba Ltd, Irvine CA). Other source samples included 4 different types of oil (rock drill oil, 2 types of lubricating oil used for diesel engines, and hydraulic oil), pulverized samples of ore and a waste rock, and a sample of diesel fuel used in the mine.

### Ambient Sampling in the Mine

***Ambient Sampling Sites*** Two ambient sampling sites were chosen in different locations in the mine. There were some limitations. They had to be places where the sampling equipment could be protected from vehicles in the mine, which included the trucks and graders used to maintain the roads. In addition, the samplers required a considerable amount of electrical power. One site had 4 pumps, each drawing 8 A of power at 110 V, in addition to some smaller equipment; the other site had 2 such pumps. (Appendix E gives a complete description of the study site.)

One site was on the main haul road, known as the old decline, at approximately 5,970 feet above mean sea level. This site is called the *upper site,* because the elevation is higher than the other ambient site, or *CE1* for Carlin East site 1. We planned to use this site, which was near the exit of ventilation air, to collect somewhat more aged aerosol mass. This site was farther away from the main active mining areas and was exposed to intermittent fresh emissions as vehicles drove along the haul road. The upper site was also near the collection point for essentially all air in the mine, which has numerous air inlets; thus, essentially all emissions from the mine had to pass by this site.

The second site, termed the *lower site* or *CE2* for Carlin East site 2, was located at approximately 5,600 feet, much closer to the active mining areas. We planned to use this site to determine the concentrations and chemical composition of emissions closer to their sources. After the first sample had been taken, there were changes in the mining activity, and we moved this site to a similar location nearby that was more in line with airflow from the active mining areas. Thus, data from the first sampling period were from a location different from data for the second and third sampling periods.

***Ambient Sampling Methods*** Sampling methods were similar at the 2 sites, but some additional samples were collected at the upper site that were not at the lower site. At each site there was an FP/SVOC sampler. In these locations the $PM_{2.5}$ cyclones were mounted in Teflon-coated stainless-steel containers to protect them from falling material. Flows were measured at the beginning and end of each sampling period and averaged to determine the total volume sampled. The flows were set at 113 L/min. Each site also contained a medium-volume DRI fine particulate sampler for inorganic species. This sampler uses a $PM_{2.5}$ cyclone, identical to that for the FP/SVOC sampler, which leads to a momentum diffuser chamber. At the bottom of the chamber were 2 cassette cartridge holders, one each for the Teflon filter and the quartz filter. Flows for each filter were approximately 20 L/min, with a makeup port flowing at 73 L/min to reach the total flow necessary for proper operation of the cyclone.

The CE1 site also contained 2 MOUDI samplers, one loaded with Teflon media for mass and elemental analyses and one loaded with aluminum media for carbon and ion analyses. The MOUDI inlets were inverted (to prevent falling material and settling particles from entering the inlet) but were otherwise operated exactly as they were in the source sampling.

***Photoelectric Aerosol Sampler*** Another experimental technique tested under ground as part of this project was the photoelectric aerosol sampler (PAS), which has been described previously (Matter et al 1999). The self-contained sampler was installed at CE1 only. The internal sample pump drew air through an inlet, and the internal memory stored the data. This device operates on the principle that higher molecular weight PAHs (those that would be associated with the particle phase) can be ionized by a fairly weak light source and the resulting ion stream can be measured. Thus, the signal strength (in this case, current) is correlated with the total amount of ionizable material present. Exactly what this signal correlates to in terms of concentrations of PAHs in the air is not certain, but the signal should be useful for indicating relative amounts of combustion-related emissions. The instrument was run during the first sampling period and then removed to download the data. We reinstalled it for the third sampling period and for the intercomparison study. Because of trouble with the computer interface, we did not get data for the first 3 periods, but we did get one good dataset for the intercomparison study (last time period).

### Personal Sampling

***Personal Samplers*** Measurements from the Carlin East gold mine included analysis of samples from personal samplers carried by mine personnel. The personal samplers were arranged to include 2 flow branches so that

Teflon and quartz, with backup PUF/XAD-4/PUF media, could be exposed. These samplers had to be loaded with fresh medium and turned on before they were given to the individual workers who carried them during the shifts. Flows were checked both before and after sampling, and a mean value was used to determine total volume sampled. Two batteries were available for each sampler so one was charging while the other was in use.

Two types of inlets were used for these personal sampler systems. An existing US Bureau of Mines impactor was modified and used to provide a 0.8-µm aerodynamic diameter inlet cutoff for one of the personal samplers. In addition, a cutoff of 2.5 µm, which is conventional for fine particles, was used for most of the personal samplers for comparison with the other sampling systems used in this study.

McCartney and Cantrell (1992), on behalf of the US Bureau of Mines, reported the design of a simple impactor inlet for personal samplers. The impactor design was based on earlier work at the University of Minnesota. The newer design utilized 4 orifices, each with a diameter of 0.0235 inches, and accommodated a flow rate of 2.0 L/min. McCartney and Cantrell (1992) presented the test data obtained with the newer design and comparisons with data obtained using the older University of Minnesota design. The newer design was characterized by a 50% aerodynamic diameter cutoff of 0.79 ± 0.01 µm.

In DRI's experimental plan for the Carlin East gold mine, flow rates greater than 2 L/min were desired in order to collect more gas and PM for analysis. After considering the trade-offs between greater flow rates on the one hand, and the battery and pump limitations of personal samplers on the other, a flow rate of 5 L/min was identified as the goal. The US Bureau of Mines impactor, in principle, would meet this goal if the number of orifices were increased from 4 to 10 and all other design features (eg, orifice diameter and separation between orifice plate and impaction plate) were retained.

Subsequently, DRI consulted Dr Cantrell, who replied that the 10-orifice design might involve greater particle losses than the 4-orifice version but otherwise would be acceptable (BK Cantrell, personal communication, February 1999). The maximum loss in the original US Bureau of Mines impactor was estimated at 3%. Somewhat greater losses might be expected to result from the DRI modification because the increase in number of orifices would increase turbulence in the zone between the orifice plate and the impaction plate. Because the losses due to the increased number of orifices would not change the cutoff point, but only the amount of material passing to the collection media, we decided to proceed with the DRI modification to the impactor.

The DRI experiment plan called for most of the personal sampler data to be collected using a 2.5-µm inlet instead of the 0.8-µm impactor. Again, a flow rate of approximately 5 L/min was desired. A commercially manufactured personal sampler cyclone, designated as the KTL cyclone (Koistinen et al 1999), operating at 4 L/min with a 2.5-µm cutoff point, was adopted for this purpose. The KTL cyclone was recommended by BGI (Waltham MA); its design is based on a detailed characterization study (Kenny and Gussman 1997). This choice was also cost effective because the KTL cyclone readily connected to the personal sampling systems with a simple adapter.

***Personal Sampling Methods*** To permit initial assessment of personal exposure to airborne vapors and particles, 6 miners per 12-hour shift were asked to carry personal samplers. Each sampler was fitted to a quartz filter backed with a PUF/XAD-4/PUF cartridge and a Teflon filter. The miners were selected at random from among operators of load, haul, and dump diesel equipment, bolters, and truck drivers. The only stipulation in their selection was that they agreed to participate after they had been given a summary of the goals of the project and assurance that no individual-specific information would be requested or otherwise obtained. Each of the participating miners signed a consent form containing information about the project, its goals, and participants' rights (Appendix D). Four control workers were similarly selected and informed: they were from the mine workforce that did not engage in below-ground activity or in other work that required them to be close to diesel equipment. At the end of each shift, the air samples were retrieved, along with a record of where each of the miners and control subjects worked, the type of equipment used, and how long each person worked at each location. These activities were conducted for 3 consecutive shifts (day, night, and day), resulting in 20 valid samples. Table 2 lists the participants according to their job description, the time period they carried the samplers, the 50% cutoff point for a given sample, as well as the total volumes collected for the quartz filter with PUF/XAD-4/PUF cartridge and for the Teflon filter.

### Intercomparison of Samples for Quality Assurance

In order to compare the personal samplers with the stationary area samplers, 3 personal samplers (2 with a 2.5-µm inlet and 1 with a 1-µm inlet) were operated in parallel with the FP/SVOC sampler and the inorganic sampler, collecting mine air during one night shift on June 5 to 6, 1999. At the same time, the PAS was collecting continuous PAH data.

**Table 2.** Personal Samples Collected in Carlin East Gold Mine

| Participant by Job Title[a] | Date | Start Time | Stop Time | Duration (min) | Sample Volume ($m^3$) Quartz Filter | Sample Volume ($m^3$) Teflon Filter | 50% Cutoff (µm) |
|---|---|---|---|---|---|---|---|
| Office 1 | 6/3/99 | 0811 | 1538 | 447 | 1.29 | 0.404 | 2.5 |
| Office 2 | 6/3/99 | 0814 | 1538 | 444 | 1.30 | 0.371 | 2.5 |
| Office 3 | 6/3/99 | 0809 | 1538 | 449 | 1.30 | 0.373 | 2.5 |
| Office 4 | 6/3/99 | 0819 | 1538 | 439 | 1.30 | 0.338 | 2.5 |
| Bolter | 6/4/99 | 0900 | 1720 | 500 | 1.31 | 0.430 | 2.5 |
| Bolter | 6/4/99 | 0900 | 1720 | 500 | 1.45 | 0.420 | 1.0 |
| Bolter | 6/5/99 | 0724 | 1720 | 596 | 1.68 | 0.548 | 2.5 |
| Jammer | 6/4/99 | 1830 | 0423 | 593 | 1.73 | 0.430 | 2.5 |
| Jammer | 6/4/99 | 0906 | 1720 | 494 | 1.33 | 0.534 | 2.5 |
| Jammer | 6/4/99 | 1830 | 0423 | 593 | 1.60 | 0.492 | 1.0 |
| Jammer | 6/5/99 | 0724 | 1515 | 471 | 1.38 | 0.410 | 1.0 |
| Shifter | 6/4/99 | 0907 | 1616 | 429 | 1.20 | 0.365 | 2.5 |
| Shifter | 6/5/99 | 0724 | 1640 | 556 | 1.59 | 0.350 | 2.5 |
| Truck driver | 6/4/99 | 1830 | 0430 | 600 | 1.19 | 0.425 | 2.5 |
| Truck driver | 6/4/99 | 0905 | 1637 | 452 | 1.44 | 0.504 | 2.5 |
| Truck driver | 6/4/99 | 1830 | 0430 | 600 | 1.63 | 0.569 | 2.5 |
| Truck driver | 6/4/99 | 0904 | 1704 | 480 | 1.11 | 0.624 | 2.5 |
| Truck driver | 6/4/99 | 1830 | 0423 | 593 | 1.44 | 0.510 | 2.5 |
| Truck driver | 6/5/99 | 0725 | 1711 | 586 | 1.71 | 0.522 | 2.5 |
| Truck driver | 6/5/99 | 0725 | 1711 | 586 | 1.61 | 0.727 | 2.5 |

[a] Bolters drill holes and place bolts to support rock walls; jammers place backfill in previously mined areas; and shifters are shift supervisors.

Only the upper site was used for this experiment. The sampling started at 1853 hours and was stopped at 0335 hours.

## CHEMICAL ANALYSIS

### Inorganic Analyses

Samples collected on Teflon-membrane filters (for gravimetric and trace element analyses) and quartz-fiber filters (for OC, EC, and ion analyses) were analyzed by the methods described below. The Teflon filters from personal samplers were analyzed for mass, trace elements, and ions; quartz filters, after a sample was taken for OC/EC analysis, were used for organic analyses.

***Gravimetric Analysis*** Unexposed and exposed Teflon-membrane filters were equilibrated at 20º ± 5°C with 30% ± 5% relative humidity for a minimum of 24 hours before being weighed. Weighing was performed on a Cahn 31 electromicrobalance with sensitivity within 0.001 mg. The charge on each filter was neutralized by exposure to a polonium source for 30 seconds before the filter was placed on the balance pan.

The balance was calibrated with a 20-mg class M weight, and the tare was set before weighing each batch of filters. After every 10 filters were weighed, the calibration and tare were rechecked. If the results of these performance tests deviated from specifications by more than 5 µg, then the balance was recalibrated. If the difference exceeded 15 µg, then the balance was recalibrated and the previous 10 samples were reweighed. At least 30% of the weights were checked by an independent technician, and samples were reweighed if these check weights did not agree within 15 µg of the original weights. Preweights and postweights, check weights, and reweights (if required) were recorded on data sheets as well as being directly entered into a database via a computer serial port (RS232) connection. All $PM_{2.5}$ and $PM_{10}$ Teflon filters were analyzed for mass.

***X-ray Fluorescence*** X-ray fluorescence (XRF) analysis was performed on Teflon-membrane filters using an energy-dispersive XRF analyzer for the following elements: aluminum, silicon, phosphorus, sulfur, chlorine, potassium, calcium, titanium, vanadium, chromium, manganese, iron, cobalt, nickel, copper, zinc, gallium, arsenic,

selenium, bromine, rubidium, strontium, yttrium, zirconium, molybdenum, palladium, silver, cadmium, indium, tin, antimony, barium, lanthanum, gold, mercury, thallium, lead, and uranium.

X-ray fluorescence analyses were performed on an energy-dispersive XRF analyzer (model 700/8000, Kevex Corporation, Scotts Valley CA) using a side-window, liquid-cooled, 60-keV, 3.3-mA rhodium anode x-ray tube and secondary fluorescers. The x-ray output stability was within 0.25% for any 8-hour period within a 24-hour duration. The silicon detector had an active area of 30 $mm^2$, with a system resolution better than 165 eV. The analysis was controlled, spectra were acquired, and elemental concentrations were calculated by software implemented on an LSI 11/23 microcomputer interfaced to the analyzer. Five separate XRF analyses were conducted on each sample to optimize the detection limits for the specified elements.

Three types of XRF standards were used for calibration, performance testing, and auditing: vacuum-deposited thin-film elements and compounds (Micromatter, Deer Harbor WA); polymer films; and US National Institute of Standards and Technology (NIST) thin-glass films. The vacuum-deposited films covered the largest number of elements and were used as calibration standards. The polymer films and NIST thin-glass films were used as quality control standards. The NIST standards are the definitive standard reference material (SRM), but these were only available for aluminum, calcium, cobalt, copper, manganese, and silicon (SRM 1832) and iron, lead, potassium, silicon, titanium, and zinc (SRM 1833). A separate Micromatter thin-film standard was used to calibrate the system for each element.

During XRF analysis, filters were removed from their petri slides and placed with their deposit sides down into polycarbonate filter cassettes. A polycarbonate retainer ring kept the filter flat against the bottom of the cassette. These cassettes were loaded into a carousel in the x-ray chamber that contained 16 openings. The filter identifications were recorded on a data sheet to correspond to numbered positions in the carousel. The sample chamber was evacuated to $10^{-3}$ mm Hg, and a computer program controlled the positioning of samples and the excitation conditions. Complete analysis of 16 samples under 5 excitation conditions required approximately 6 hours. The vacuum in the x-ray chamber and the heat induced by the absorption of x-rays can cause certain materials to volatilize. For this reason, labile species such as $NO_3^-$ and OC were measured on a quartz-fiber filter, rather than on the Teflon-membrane filter that was subjected to XRF analysis.

A quality control standard and a replicate from a previous batch were analyzed with each set of 14 samples. When a quality control value differed from specifications by more than 5%, or when a replicate concentration differed from the original value (when values exceeded 10 times the detection limits) by more than 10%, the samples were reanalyzed. If further tests of standards showed that the system calibration had changed by more than 2%, then the instrument was recalibrated as described above. All XRF results were directly entered into the DRI databases.

***Thermo-optical Reflectance Carbon Analysis*** The TOR method was used to measure OC and EC. This method is based on the principle that different types of carbon-containing particles are converted to gases under different temperature and oxidation conditions. The different carbon fractions from TOR analysis were useful for comparison with results from other methods that are specific to a single definition for OC and EC. These specific carbon fractions also helped distinguish among 7 carbon fractions reported by TOR: The OC evolved in a helium atmosphere at temperatures between ambient and 120°C (OC1), between 120°C and 250°C (OC2), between 250°C and 450°C (OC3), and between 450°C and 550°C (OC4). The EC evolved in an oxidizing atmosphere at 550°C (EC1), between 550°C and 700°C (EC2), and between 700°C and 800°C (EC3).

The TOR carbon analyzer consisted of a thermal system and an optical system. The thermal system consisted of a quartz tube placed inside a coiled heater. Current through the heater was controlled to attain and maintain preset temperatures for given time periods. A punch sample of 0.5 $cm^2$ from a quartz filter was placed in the heating zone and heated to different temperatures under nonoxidizing and oxidizing atmospheres. The optical system consisted of a helium-neon laser, a fiberoptic transmitter and receiver, and a photocell. The filter deposit faced a quartz light tube so that the intensity of the reflected laser beam could be monitored throughout the analysis.

As the temperature increased from ambient (approximately 25°C) to 550°C, organic compounds were volatilized from the filter in a nonoxidizing (helium) atmosphere, but EC was not oxidized. When oxygen was added to the helium at temperatures above 550°C, the EC burned and entered the sample stream. The evolved gases passed through an oxidizing bed of heated manganese dioxide where they were oxidized to carbon dioxide, then across a heated nickel catalyst, which reduced the carbon dioxide to methane ($CH_4$). The methane was then quantified with a flame ionization detector.

The reflected laser light was continuously monitored throughout the analysis cycle. The negative change in reflectance was proportional to the degree of pyrolytic conversion from OC to EC that took place during OC analysis. After

oxygen was introduced, the reflectance increased rapidly as the light-absorbing carbon was burned off the filter. The carbon measured after the reflectance that had attained the value it had at the beginning of the analysis cycle was classified as EC. This adjustment for pyrolysis in the analysis was as high as 25% of OC or EC, and it should not be ignored.

The system was calibrated by analyzing samples of known amounts of methane, carbon dioxide, and potassium hydrogen phthalate. The ratio of the flame ionization detector response to a reference level of methane injected at the end of each sample analysis was determined. Performance tests of the instrument calibration were conducted at the beginning and end of each day's operation. Intervening samples were reanalyzed when calibration changes of more than 10% were found.

Known amounts of American Chemical Society (ACS) certified reagent-grade crystal sucrose and potassium hydrogen phthalate were committed to TOR as a verification of the OC fractions. Fifteen different standards were used for each calibration. Widely accepted primary standards for EC and OC are still lacking. Results of the TOR analysis of each filter were entered into the DRI database.

***Filter Sectioning and Extraction*** Water-soluble chloride, nitrate, and sulfate were obtained by extracting half of the quartz-fiber particle filter in 15 mL of deionized distilled water (DDW). For personal samples, this analysis was performed from Teflon filters because the remainder of each quartz filter after a punch was taken for TOR analysis was used for organic analysis. Each filter was cut in half with a precision positioning jig attached to a paper cutter. The blade was cleaned between filter cuttings. The extraction vials were capped and sonicated for 60 minutes, shaken for 60 minutes, and then aged overnight to ensure complete extraction of the deposited material in the solvent. The ultrasonic bath water was monitored to prevent temperature increases from dissipation of ultrasonic energy in the water. After extraction, these solutions were stored under refrigeration until analysis. The unanalyzed filter half was placed back in the original petri slide and archived.

***Ion Chromatography for Chloride, Nitrate, and Sulfate*** Water-soluble $Cl^-$, $NO_3^-$, and $SO_4^{2-}$ were measured with an ion chromatograph (model 2020i, Dionex, Sunnyvale CA). In ion chromatography, an ion-exchange column separates the sample ions in time for individual quantification by a conductivity detector. Prior to detection, the column effluent enters a suppressor column where the chemical composition of the component is altered, resulting in a matrix of low conductivity. The ions are identified by their elution or retention times and quantified by the conductivity peak area.

Approximately 2 mL of the filter extract was injected into the ion chromatograph. The resulting peaks were integrated, and the peak integrals were converted to concentrations using calibration curves derived from solution standards. The Dionex system for analysis of $Cl^-$, $NO_3^-$, and $SO_4^{2-}$ contained a guard column (AG4a column, catalog number 37042) and an anion separator column (AS4a column, catalog number 37041) with a strong basic anion exchange resin, and an anion micromembrane suppressor column (250-mm × 6-mm internal diameter) with a strong acid ion exchange resin. The anion eluent consisted of sodium carbonate ($Na_2CO_3$) and sodium bicarbonate ($NaHCO_3$) prepared in DDW. The DDW was verified to have a conductivity of less than $1.8 \times 10^{-5}$ Ω/cm prior to preparation of the eluent. For quantitative determinations, the ion chromatograph was operated at a flow rate of 2.0 mL/min.

The primary standard solution containing sodium chloride (NaCl), sodium nitrate ($NaNO_3$), and sodium sulfate ($Na_2SO_4$) were prepared with reagent-grade salts that were dried in an oven at 105°C for 1 hour and then brought to room temperature in a desiccator. These anhydrous salts were weighed to the nearest 0.10 mg on a routinely calibrated analytic balance under controlled conditions of temperature (approximately 20°C) and relative humidity (approximately 30%). These salts were diluted in precise volumes of DDW. Calibration standards were prepared at least once within each month by diluting the primary standard solution to concentrations covering the range expected in the filter extracts and storing them in a refrigerator. The calibration concentrations were 0.1, 0.2, 0.5, 1.0, and 2.0 µg/mL for each of the analysis species.

Calibration curves were obtained weekly. Chemical compounds were identified by matching the retention time of each peak in the unknown sample with the retention times of peaks in the chromatograms of the standards. As quality control checks, a DDW blank was analyzed after every 20 samples to verify the baseline, and a calibration standard was analyzed after every 10 samples. Standards from Environmental Research Associates (ERA, Arvada CO) were used daily as an independent quality assurance check. These standards (ERA wastewater nutrient and ERA mineral wastewater) are traceable to NIST-simulated rainwater standards. If the values obtained for these standards did not fall within a prespecified uncertainty level (typically within 3 SD of the baseline level or 5%), then the samples between that standard and the previous calibration standards were reanalyzed.

After analysis, the printout for each sample in the batch was reviewed for proper operational settings, correct peak

shapes and integration windows, peak overlaps, correct background subtraction, and quality control sample comparisons. When values for replicates differed by more than 10%, or values for standards differed by more than 5%, samples before and after these quality control checks were designated for reanalysis in a subsequent batch. Individual samples with unusual peak shapes, background subtractions, or deviations from standard operating parameters were also designated for reanalysis.

***Resuspension Analysis*** Pulverized ore and waste rock samples obtained in the mine were suspended in the DRI multiple-port resuspension chamber and sampled onto filters through size-selective inlets, which were submitted to chemical analyses. Chow and colleagues (1994b) have described this procedure in detail. Briefly, a cellulose-fiber resuspension chamber was placed over the impactors with cutoff points of PM less than 1 µm in aerodynamic diameter ($PM_1$), $PM_{2.5}$, and $PM_{10}$. The bottom of the chamber was sealed to the platform surface. A 20-cm × 13-cm quartz-fiber filter was mounted across an opening on top of the resuspension chamber to provide filtered makeup air at a flow rate of 80 L/min for concurrent sampling with 8 ports. Individual samples were dried at 40°C and 20% relative humidity, then sieved through a Tyler 400 mesh screen (less than 38-µm geometric diameter, which corresponds to approximately 64-µm aerodynamic diameter for 3 g/cm$^3$ density and spherical particles) prior to resuspension. Approximately 0.1 g of the sieved material was placed in a 250-mL side-arm vacuum flask sealed with a rubber stopper. A hollow glass tube extended through the stopper and into the flask with its outlet 1 cm above the bulk material. As filtered air was pumped through the resuspension chamber and the sampling ports, the vacuum flask was positioned with its side arm at the inlet of the resuspension chamber. A pulsed air jet (45 to 50 L/min), introduced through the glass tube into the vacuum flask, suspended the sieved dust and carried it into the resuspension chamber, where it was turbulently mixed with filtered air.

Aerosolized particles were introduced into the chamber for 5 seconds every 4 minutes until net filter deposits of 0.5 to 5 mg were obtained in each size fraction. Small particles followed the airflow through a tubular aluminum diffuser, 32 cm long by 5 cm in internal diameter, and were collected on a filter located downstream. The impaction plate was coated with a thin layer of grease (type M, Apiezon, London, England) to eliminate particle bounce and reentrainment. The diffuser tube provided a uniform flow and homogeneous sample deposits. Downstream of each filter, a critical orifice maintained a constant flow rate of 10 L/min through each sampling port. The spacing between the sampling ports allowed simultaneous sampling of different particle sizes while minimizing interference with the surrounding airstream. This configuration can accommodate simultaneous sampling for $PM_1$, $PM_{2.5}$, $PM_{10}$, and total suspended particle size fractions onto 2 filter media, any 2 of these size fractions onto 4 different filter media, or any other combination of particle size and filter media. For this study Teflon-membrane and quartz-fiber filters were used and the $PM_{2.5}$ size fraction was subjected to chemical analyses including gravimetric analysis, OC/EC analysis by TOR, ion analysis by chromatography, and element analysis by XRF.

## Organic Analyses

***Extraction of Filters and PUF/XAD-4/PUF Cartridges*** The extraction was performed the same way for personal and FP/SVOC samples. Prior to extraction, the following deuterated internal standards were added to each filter-sorbent pair: naphthalene-$d_8$, acenaphthylene-$d_8$, phenanthrene-$d_{10}$, anthracene-$d_{10}$, chrysene-$d_{12}$, pyrene-$d_{10}$, benz[*a*]anthracene-$d_{12}$, benzo[*a*]pyrene-$d_{12}$, benzo[*e*]pyrene-$d_{12}$, benzo[*k*]fluoranthene-$d_{12}$, benzo[*ghi*]perylene-$d_{12}$, coronene-$d_{12}$, and 1-nitropyrene-$d_9$. Since PUF should not be extracted with $CH_2Cl_2$, the PUF plugs are Soxhlet-extracted separately with hexane/diethyl ether (9:1), and the filter–XAD-4 pairs are microwave-extracted with $CH_2Cl_2$; these methods of extraction have been reported to yield high recovery of PAH (Chuang et al 1990; Zielinska et al 1998a).

The extracts were then concentrated by rotary evaporation at 20°C under gentle vacuum to approximately 1 mL and filtered through 0.45-mm Acrodiscs (Gelman Scientific, now part of Pall Sciences, Ann Arbor MI), and the sample flask was rinsed twice with 1 mL of $CH_2Cl_2$ each time. Approximately 100 µL of acetonitrile was added to the sample, and $CH_2Cl_2$ was evaporated under a gentle stream of nitrogen. This procedure has been tested by Atkinson and coworkers (1988). For personal samples the final volume was adjusted to 100 µL, and for source and ambient samples it was 1 mL.

For hopanes, steranes, and nitro-PAHs, the source and ambient samples had to be cleaned prior to GC/MS analysis using a solid-phase extraction (SPE) technique described by Wang and colleagues (1994a,b). Cleanup was conducted on a 6-mL Supelco SPE cartridge packed with 0.5 g of silicon hydroxide (SiOH). Samples were spiked onto an SPE cartridge along with 10 µL of *n*-tetrocosane-$d_{50}$ (internal standard) and the PAH internal standards described above. Elution and fractionation were conducted with 1 mL of hexane followed by 1.25 mL of benzene/hexane (1:1). Hopanes and steranes were eluted along with *n*-tetrocosane-$d_{50}$ in the hexane fraction, while the nitro-PAHs were eluted in the hexane/benzene fraction.

***Semivolatile Organic Compound Analysis*** The samples were analyzed by electron impact GC/MS for PAHs, hopanes, steranes, and nitro-PAHs. A gas chromatograph (Varian Star 3400CX) equipped with an 8200CX automatic sampler and interfaced to an ion trap (Varian Saturn 2000) was used for these analyses. Injections (1 µL) were made in the splitless mode onto a 30-m 5% phenylmethylsilicone fused-silica capillary column (DB-5ms, J&W Scientific, Folsom CA). The individual compounds were quantified by the selective ion storage technique, and the molecular ion of each compound of interest and the corresponding deuterated internal standard, added prior to extraction, were monitored.

Calibration curves for the GC/MS quantification were made for the most abundant and characteristic ion peaks of the hopanes, steranes, and PAH and nitro-PAH compounds using the deuterated species most closely matched in volatility and retention characteristics as internal standards. Authentic PAH and nitro-PAH standards (purchased from Aldrich, Milwaukee WI) plus NIST SRM 1647 (certified PAH) with the addition of deuterated internal standards were used to make calibration solutions. For quantifying hopanes and steranes the following authentic standards were used: C27 20R-5α,14α,17α-cholestane (purchased from Aldrich), and 17β(H),21β (H)-hopane (purchased from Chiron AS, Norway). The remaining hopanes and steranes were identified by comparison of their mass spectra and retention times with published data (Rogge et al 1993; Wang and Fingas 1995). For quantification of the hopanes and steranes for which authentic standards were not available, the response factors of standards most closely matched in volatility and retention characteristics were used. For quantification of nitro-PAHs, the following authentic standards were used: 1-nitronaphthalene, 1-methyl-2-nitronaphthalene, 2- and 4-nitrobiphenyl, 3-nitrofluoranthene, and 1-nitropyrene.

A 3-level calibration was performed for each compound of interest, and the calibration was checked (using median calibration standards) every 10 samples for accuracy of analyses. If the relative accuracy of measurement (defined as a percentage difference from the standard value) was less than 20%, then the instrument was recalibrated.

***Special Analyses*** Rock drill oil, hydraulic fluid, 15/40-weight lubricating oil, and 40-weight lubricating oil were analyzed for PAHs, hopanes, and steranes. The oils were cleaned and fractionated prior to analysis using the method described by Wang and colleagues (1994a,b). Cleanup was conducted on a 12-mL Supelco SPE cartridge packed with 2 g of SiOH. Cartridges were placed on a vacuum manifold and conditioned prior to cleanup with 14 mL of hexane. Prior to cleanup, oils were diluted in hexane (60 µL/1 mL). Diluted oil (300 µL) was spiked onto an SPE cartridge along with 10 µL of tetrocosane-$d_{50}$ (internal standard) and the PAH internal standard. Samples were eluted and fractionated with 8 mL of hexane followed by 10 mL of benzene/hexane (1:1). The benzene/hexane fraction was analyzed for PAHs, and the hexane fraction was analyzed for hopanes and steranes.

In order to calculate the mass of oil spiked onto the SPE cartridge, oil densities were measured. Density was measured by making 3 consecutive mass measurements of 1-mL oil aliquots at 20°C. Oil densities ranged from 0.88 to 0.91 g/mL.

The rock drill oil, hydraulic fluid, 15/40-weight lubricating oil, and 40-weight lubricating oil also were analyzed for elements by XRF and for OC/EC by the TOR method. For TOR analysis, a 0.56-$cm^2$ punch from a prefired quartz-fiber filter was spiked with approximately 20 µL of oil solution in hexane (1 mg/mL), and the solvent was allowed to evaporate, resulting in approximately 20 µg of oil deposited on the punch. The punches were then subjected to OC/EC analysis as described previously in the section Thermo-optical Reflectance Carbon Analysis. For elements, a 47-mm Teflon-membrane filter was first submerged in a solution of oil in hexane, then the filter was removed, and hexane was allowed to evaporate. The mass of oil deposited on the filter was determined from the differences in filter mass before and after submerging it in the oil solution. The filters were then analyzed by XRF as described previously in the section X-ray Fluorescence.

## PROPAGATION OF ANALYTIC UNCERTAINTY (ERROR)

All analytic results were evaluated in terms of their associated measurement errors according to the following equation:

Uncertainty = Square Root ([Analyte Concentration × Replicate Precision]$^2$ + [Analyte Detection Limit]$^2$).

Replicate precision for each analyte was determined by multiple injections (replicates) of at least 10% of all of the analyzed samples. Precision was then determined by the equation

$$(C1 - C2)/[(C1 + C2)/2] \times 100.$$

By this equation the analytic detection limit (ADL) will determine the analyte uncertainty when sample concentrations (C) approach zero. Similarly, the ADL will have little impact on the uncertainty of a sample in which the concen-

tration is many times the detection limit. In addition to this, the uncertainty in the volume flow is incorporated into the final uncertainty by a similar root-mean-square method. In this way the uncertainty most accurately represents the true uncertainty of the sample. Also, all samples are corrected for blank values in lot-specific sampling media prior to final concentration calculations. Software programs have been developed by DRI to automate the data processing and reporting functions.

## DATA ANALYSIS BY CHEMICAL MASS BALANCE

The CMB is used to estimate the contributions of different sources to carbon and $PM_{2.5}$ concentrations. This section explains how CMB operates, its assumptions, how it was applied to the current study, tests of deviations from CMB assumptions, and sensitivities to uncertainties and inaccuracies in the input data.

### Chemical Mass Balance Formulation

The CMB is one of several receptor-oriented data analysis methods applicable to air resources management. It uses the chemical and physical characteristics of gases and particles measured at the source and at the receptor to both identify the presence of and quantify source contributions to receptor concentrations.

Receptor-oriented simulations are generally contrasted with source-oriented atmospheric dispersion simulations that use pollutant emissions rate estimates, meteorologic transport information, and chemical transformation mechanisms to estimate the contribution of each source to receptor concentrations. The two types of simulation are complementary, with each type having strengths that compensate for the weaknesses of the other. Further information about CMB and other receptor models is available in several review articles, books, and conference proceedings (Watson 1979; Gordon 1980, 1988; Hopke and Dattner 1982; Stevens and Pace 1984; Watson et al 1984, 1990a,b,c,d,e, 1991; Chow and Watson 1985; Hopke 1985a,b; Pace 1986, 1991; Chow et al 1993).

The CMB model consists of a solution to linear equations that express each receptor chemical concentration as a linear sum of products of source profile abundances and source contributions (Friedlander 1973a,b; Cooper and Watson 1980; Gordon 1980, 1988; Watson et al 1984, 1990e, 1991; Hidy and Venkataraman 1996). The source profile abundances (ie, the mass fraction of a chemical or other property in the emissions from each source type) and the receptor concentrations, with appropriate uncertainty estimates, serve as inputs to the CMB. The CMB calculates values for the contributions from each source and the uncertainties of those values.

The CMB is implicit in all factor analysis and multiple linear regression models that intend to quantitatively estimate source contributions (Watson 1979). These models attempt to derive source profiles from the covariation in space and time of many different samples of atmospheric constituents that originate in different sources. These profiles are then used in a CMB to quantify source contributions to each ambient sample.

The CMB is applicable to multispecies datasets, the most common of which are chemically characterized $PM_{10}$, $PM_{2.5}$, and volatile organic compounds. The contribution of important sources of particulate air pollution in the Carlin East gold mine were quantified using semivolatile and particulate organic compounds along with carbon and other selected elements.

The CMB procedure requires: (1) identification of the contributing sources types; (2) selection of chemical species or other properties to be included in the calculation; (3) estimation of the fraction of each of the chemical species that is contained in each source type (source profile); (4) estimation of the uncertainty in both ambient concentrations and source profiles; and (5) solution of the CMB equations.

Several methods have been proposed for solution of the CMB equations: single unique species to represent each source (tracer solution); linear programming solutions; ordinary weighted least squares, weighting only by the precision of ambient measurements (Friedlander 1973a; Gartrell and Friedlander 1975); ridge regression weighted least squares (Williamson and DuBose 1983); partial least squares (Larson and Vong 1989; Vong et al 1988); neural networks (Song and Hopke 1996); and effective variance weighted least squares (Watson et al 1984).

The effective variance weighted solution is almost universally applied because it theoretically yields the most likely solutions to the CMB equations, providing model assumptions are met; it uses all available chemical measurements, not only the so-called tracer species; it analytically bases estimates of the uncertainty of the source contributions on the precision of both the ambient concentrations and the source profiles; and it gives greater influence to chemical species with higher precision in both the source and receptor measurements than to species with lower precision. The effective variance is a simplification of a more exact, but less practical, generalized least squares solution proposed by Britt and Luecke (1973).

The CMB model assumptions are as follows:

- compositions of source emissions are constant over the period of ambient and source sampling;

- chemical species do not react with one another (ie, they add linearly);
- all sources with a potential for contributing to the receptor have been identified and their emissions have been characterized;
- the number of sources or source categories is less than or equal to the number of species;
- the source profiles are linearly independent of one another; and
- measurement uncertainties are random, uncorrelated, and normally distributed.

The degree to which these assumptions are met in applications depends to a large extent on the particle and gas properties measured at source and receptor. Performance of the CMB model is examined generically, by applying analytic and randomized testing methods, and specifically for each application, by following an applications and validation protocol (Pace and Watson 1987). The assumptions are fairly restrictive, and total compliance with them may never be achieved in actual practice. Fortunately, the CMB model can tolerate reasonable deviations from these assumptions although deviations increase the stated uncertainties of the source contribution estimates (SCEs) (Watson 1979; Gordon et al 1981; Henry 1982, 1992; Currie et al 1984; Dzubay et al 1984; deCesar et al 1985, 1986; Javitz and Watson 1986; Lowenthal et al 1987, 1988, 1992; Lowenthal and Rahn 1988a,b; Javitz et al 1988a,b; Cheng and Hopke 1989; Kim and Henry 1989).

The CMB model is intended to complement rather than replace other data analysis and modeling methods. It explains observations that have already been made, but it does not predict the future. When source contributions are proportional to emissions, as they often are for PM, volatile organic compounds, and the sum of gas and particle phases for certain semivolatile components, then a source-specific proportional rollback (Barth 1970; Cass and McCrae 1981; Chang and Weinstock 1975; deNevers and Morris 1975) is used to estimate the effects of emissions reductions. Similarly, when a secondary compound apportioned by CMB is known to be limited by a certain precursor, a proportional rollback is used on the controlling precursor.

The CMB model does not explicitly treat profiles that change between source and receptor. Most applications use source profiles measured at the source; at most, there is dilution to ambient temperatures and less than 1 minute of aging prior to collection to allow for condensation and rapid transformation. Profiles have been aged prior to submission to the CMB, by use of aerosol and gas chemistry models to simulate changes between source and receptor (Friedlander 1981; Lin and Milford 1994; Venkatraman and Friedlander 1994, Watson et al 1996). These models are often overly simplified and require additional assumptions regarding chemical mechanisms, relative transformation and deposition rates, mixing volumes, and transport times.

The CMB model quantifies contributions from chemically distinct source types rather than contributions from individual emitters. It cannot distinguish between sources with similar chemical and physical properties. During the current study, exhausts from different underground mining vehicles showed similar compositions and were grouped into one composite diesel profile. These were distinguished from ore and drilling oil profiles.

### Chemical Mass Balance Input Data

Source profiles and ambient measurements of semivolatile and particle-bound chemical constituents were used as CMB input data. Both source profiles and ambient data included elements, inorganic ionic species, carbon (OC and EC), and speciated organic compounds. The speciated organics were not quantified in the ore because this material is of geologic origin and consists of very low amounts of OC. Table 3 lists the individual samples that were used to create 2 composite vehicle source profiles (DIESEL and DIESEL2) for use in the apportionment. The individual source tests for the vehicles are described in the Source Sampling section and Table 1. The source characterizations of the suspended ore and oil profiles are described in the Resuspension Analysis and Special Analyses sections, respectively. (Chemical abundances of source profiles are presented in Appendix B.) The profiles are expressed as weight percentages of total reconstructed $PM_{2.5}$ mass (ie, geologic sources [1.89 (aluminum) + 2.14 (silicon) + 1.4 (calcium) + 1.43 (iron)] + organics [1.2 (OC)] + EC + sulfate + nitrate + ammonium) with 1 SD analytic errors for individual measurements.

The CMB implements measurement and propagated uncertainties to weight the impact of each fitting species in the outcome of the source apportionment. A high uncertainty estimate for an input species will have little to no

**Table 3.** Individual Diesel Vehicle Exhaust Profiles Included in Composite Profiles

| DIESEL | DIESEL2 |
|---|---|
| ULD008R | ULD004 |
| UHT024 | ULD008 |
| ULD024R | ULD006 |
| ULD006 | |
| ULD022 | |
| UHT024 | |
| UTR009 | |

impact on the apportionment results. For this reason, input species with ratios of concentration to uncertainty less than 1 were removed from the ambient and source composition databases because they would not influence the apportionment.

Uncertainty estimates for each individual profile are the analytic uncertainties explained above under Propagation of Analytic Uncertainty (Error). These measurement errors are assumed to be random, uncorrelated, and normally distributed. When multiple profiles are combined into one input profile, uncertainties are defined as the larger of either the 1 SD variations in fractional abundances among profiles or the root mean square of all analytic uncertainties for each input species. (Uncertainty assessments for each source profile are presented in Appendix B.)

### Chemical Mass Balance Simulation and Measurement Evaluation

The formal protocol for CMB model application and validation (Pace and Watson 1987; Watson et al 1991, 1998) is applicable to the apportionment of particles and gases (Fujita et al 1994). This 7-step protocol:

- determines model applicability;
- selects a variety of profiles to represent identified contributors;
- evaluates model outputs and performance measures;
- identifies and evaluates deviations from model assumptions;
- identifies and corrects model input deficiencies;
- verifies the consistency and stability of SCEs; and
- evaluates CMB results with respect to other data analysis and source assessment methods.

The following subsections evaluate the CMB simulation of source contributions according to these steps.

***Model Applicability*** The requirements for CMB model applicability are that the number of receptor samples taken with an accepted method is sufficient to evaluate temporal and spatial variations in marker species concentration, that samples are analyzed for chemical species which are also present in source emissions, that potential source contributors have been identified and chemically characterized, and that the number of noncolinear source types is less than the number of measured species.

Aerosol samples were taken by well-characterized methods, and measurements were fully evaluated (Chow et al 1994a; Zielinska et al 1998b). Contributors to ambient fine PM (less than 2.5 µm in diameter) in the mine were identified as heavy-duty diesel-mining utility and transport vehicles; ore that is suspended during mining operations (drilling, loading, etc); and drilling oil that is suspended during drilling operations. Representative chemical profiles were acquired from the composites (individual sample profiles with their associated analytic uncertainties are presented in Appendix B).

The number of fitting species used in the CMB (about 60 to 70 organic species and 20 to 25 elements, ions, and carbon species) exceeded the number of source types (up to 3 source types). Thus, the CMB was applicable to source apportionment for the ambient data from the Carlin East gold mine.

***Evaluation and Determination of Final Input Criteria***

Initial CMB tests were performed to evaluate the sensitivity of input profiles to alternative conditions and to select a default set of source profiles and fitting species. Each individual diesel profile was applied along with an ore sample (ORE1) to one of the ambient samples (CE1#3) using a default set of fitting chemical species. The default fitting species included OC and EC, elements, and speciated organics. The impacts of oils, including rock drill oil, hydraulic oil, 15/40-weight lubricating oil, and 40-weight lubricating oil, were evaluated by including them in apportionment sensitivity tests using default ore (ORE1) and diesel vehicle profiles (DIESEL2). No attempt was made to manually improve the apportionment by altering the chemical species for these tests. These tests were used to select a default set of fitting species and to examine the sensitivity of the apportionment to alternative source profiles. Table 4 shows the results of the sensitivity tests, including the effects of using alternative fitting species on apportionment results. Results for each source type and the assessment of alternative fitting species are discussed below.

***Evaluation of Vehicle Profile Sensitivity*** The range of fine particulate mass explained by individual and composite diesel emissions profiles was 80% to 94% (Table 4). With the exception of profiles for vehicles ULD004, ULD005, and ULD008, the individual diesel profiles generally yielded acceptable CMB performance parameters ($r^2 > 0.8$; percentage of mass, 80% to 120%; $\chi^2 < 3$). When multiple diesel profiles were apportioned at the same time, the overall contribution to $PM_{2.5}$ mass for this source was similar to the result when a single profile was used. Because of the similarities in overall chemical composition among exhausts from the vehicles characterized in this study, and the consistency in the apportionment results for each individual profile, a composite profile was created to represent this source. This profile, entitled *DIESEL*, is a composite of all of the vehicles tested with the exception of ULD004, ULD005, and ULD008, which were excluded because of poor CMB performance parameters.

**Table 4.** Sensitivity of Chemical Mass Balance (CMB) Apportionment to Use of Alternative Source Profiles and Fitting Species[a]

| $r^2$ | $\chi^2$ | $PM_{2.5}$ Mass (%) | DIESEL | DIESEL2 | ULD 005 | UHT 006 | ULD 008 | ULD 008R | ULD 004 | UHT 022 | ULD 024 | ULD 024R | UHT 024 | UHT 002 | UHT 005 | UTR 009 | Rock Drill Oil | Lube Oil 15/40 | Lube Oil 40 | Hydraulic Oil | ORE1 | ORE2 | % Diesel | % Ore | % Other |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 0.85 | 0.62 | 98.8 | 493.0 | | | | | | | | | | | | | | | | | | 55.0 | | 90.0 | 10.0 | |
| 0.90 | 1.52 | 92.9 | 458.0[b] | | | | | | | | | | | | | | | | | | 56.9 | | 88.9 | 11.1 | |
| 0.85 | 0.41 | 88.5 | 434.4 | | | | | | | | | | | | | | 5.67[e] | | | | 50.5 | | 88.5 | 10.3 | 1.2 |
| 0.80 | 0.40 | 114.2 | 582.5[c] | | | | | | | | | | | | | | | | | | 50.5 | | 92.0 | 8.0 | |
| 0.87 | 0.52 | 93.3 | 460.7[d] | | | | | | | | | | | | | | | | | | 56.8 | | 89.0 | 11.0 | |
| 0.81 | 0.40 | 90.4 | | 467.2 | | | | | | | | | | | | | | | | | 34.3 | | 93.2 | 6.8 | |
| 0.77 | 6.81 | 43.6 | | | 199.0 | | | | | | | | | | | | | | | | 42.6 | | 82.4 | 17.6 | |
| 0.82 | 4.88 | 91.7 | | | | 453.7 | | | | | | | | | | | | | | | 54.8 | | 89.2 | 10.8 | |
| 0.81 | 5.29 | 79.0 | | | | | 394.6 | | | | | | | | | | | | | | 43.6 | | 90.1 | 9.9 | |
| 0.87 | 2.85 | 80.5 | | | | | | 357.6 | | | | | | | | | | | | | 88.6 | | 80.1 | 19.9 | |
| 0.82 | 4.41 | 84.8 | | | | | | | 436.6 | | | | | | | | | | | | 33.4 | | 92.9 | 7.1 | |
| 0.94 | 1.30 | 84.2 | | | | | | | | 374.2 | | | | | | | | | | | 92.6 | | 80.2 | 19.8 | |
| 0.86 | 3.19 | 78.2 | | | | | | | | | 344.5 | | | | | | | | | | 88.9 | | 79.5 | 20.5 | |
| 0.92 | 1.40 | 103.2 | | | | | | | | | | 462.7 | | | | | | | | | 109.7 | | 80.8 | 19.2 | |
| 0.87 | 2.93 | 92.1 | | | | | | | | | | | 421.4 | | | | | | | | 89.1 | | 82.5 | 17.5 | |
| 0.95 | 0.89 | 92.1 | | | | | | | | | | | | 406.2 | | | | | | | 104.8 | | 79.5 | 20.5 | |
| 0.88 | 2.38 | 104.3 | | | | | | | | | | | | | 472.9 | | | | | | 105.3 | | 81.8 | 18.2 | |
| 0.91 | 1.87 | 98.7 | | | | | | | | | | | | | | 446.8 | | | | | 100.3 | | 81.7 | 18.3 | |
| 0.90 | 4.34 | 101.9 | | | | 239.4 | | 260.4 | | | | | | | | | | | | | 65.2 | | 88.5 | 11.5 | |
| 0.82 | 0.45 | 108.6 | | 472.9 | | | | | | | | | | | | | 94.5[e] | | | | 34.9 | | 78.5 | 5.8 | 15.7 |
| 0.86 | 0.23 | 381.4 | | 456.0 | | | | | | | | | | | | | | 1.8E+308 | | | 54.6 | | 21.6 | 2.6 | |
| 0.83 | 0.34 | 140.9 | | 469.2 | | | | | | | | | | | | | | | 273.6 | | 38.6 | | 60.0 | 4.9 | 35.0 |
| 0.85 | 0.22 | 596.7 | | 452.4 | | | | | | | | | | | | | | | | 1.8E+308 | 57.1 | | 13.7 | 1.7 | |
| 0.86 | 0.31 | 90.4 | | 453.3 | | | | | | | | | | | | | | | | | | 47.6 | 90.5 | 9.5 | |

[a] The reported species is $PM_{2.5}$ and the concentration is 554.5 µg/m$^3$. The mass reported here is the reconstructed mass that was used in the source profiles.

[b] No organic compounds were utilized as fitting species for this test.

[c] No carbon measured by thermo-optical reflectance was used as fitting species for this test.

[d] No semivolatile organic compounds were used as fitting species for this test.

[e] Uncertainty for this number is higher than the apportioned value (error = 105.8 for apportionment with DIESEL and error = 141.9 with DIESEL2).

Descriptions of the vehicles that were included in the apportionment are listed in Table 1. A composite diesel profile does not give information about the contribution of individual vehicles or types of vehicles to ambient mine PM. Rather, this profile helps quantify the ambient contributions of the vehicles collectively. Since the influence of input species is weighted depending on the level of uncertainty, composite samples help to minimize the contribution of individual chemical species that may be highly variable between vehicles.

***Evaluation of the Suspended Ore Profile Sensitivity*** ORE1 was used as the default ore profile for all CMB sensitivity tests. The contribution of suspended ore to fine particle mass ranged from 6% to 20%, depending on the individual vehicle profile used for the sensitivity tests. Larger contributions were observed for ore when the CMB estimated vehicle contribution was less, indicating that the apportionments for these 2 sources were related (colinear). The ore profiles were less than 10% carbon, and their apportionment heavily depended on the crustal elements such as silica, iron, calcium, magnesium, aluminum, lead, and nickel. Many of these metals were also observed in emissions from the diesel vehicles. The MPN matrix (Watson et al 1990e), a function in the CMB software that displays the effect of individual species on the apportionment, showed that many of these metals have some influence on diesel apportionment when individual diesel profiles are used. As mentioned previously, the effect of input species is minimized when their uncertainty is higher. The composite diesel profile used for the final apportionment contained high uncertainty (based on sample variability) for the

metals and minimized the colinearity between the ore and diesel emissions.

The second suspended ore profile, ORE2, was similar in composition and yielded the same result as ORE1 when apportioned with diesel (Table 4). The ORE1 profile was used as the default profile for the final apportionment.

***Evaluation of Suspended Oils Profile Sensitivity*** As a result of including oil source profiles in the source apportionment, either the percentage of mass was grossly overestimated, or the uncertainty in the oil apportionment was higher than the amount of material attributed to ambient mine air. The rock drill oil was tested both with the vehicle exhaust composite profile DIESEL and with the default composite profile DIESEL2. For both of these tests the standard error, an indicator of the precision or certainty of the SCE, was significantly higher than the apportioned amount of rock drill oil. Use of oils in the source apportionment affected the apportionment of diesel vehicles and ore by less than 5%. The results of these sensitivity tests indicated that oils were not an important source of PM in the mine, and they were not included in the final source apportionment.

***Evaluating Effects of Alternative Fitting Species on Source Contribution Estimates*** The default fitting species included inorganic species, carbon, and particle-phase organic species (particulate PAHs, hopanes, and steranes) with values of the ratio of residual (difference between calculated and measured concentrations) to its uncertainty (R/U; see elsewhere under Model Outputs and Performance Measures) between −2 and +2. Organic and elemental carbon were 2 of the fitting species that were not used with the default set because of poor CMB performance for the calculated amounts versus the measured amounts. In previous CMB studies, the split between OC and EC has been important to distinguish diesel vehicles from other carbonaceous sources. Because of the poor CMB fit for OC and EC, TC was used as a fitting species in their place. The effect of not using OC and EC was not important in this study because there are believed to be no other major sources (contributing more than 50%) of carbon in the mine. When OC and EC were used, the source contributions to the explained particulate mass were not affected by more than 5% to 10%. However, the CMB performance parameters and the amount of mass explained by the apportionment were significantly affected by the inclusion of OC and EC. (The rationale for the poor CMB performance of OC and EC here is explained more fully in the section Source Contributions to Particulate Matter.)

The sensitivity tests included the following changes to the default set of fitting species: no speciated organics; no semivolatile speciated organics (defined operationally as PAHs that eluted on a chromatographic column before phenanthrene); and no carbon measured using the TOR method. The sensitivity tests (Table 4) showed that altering the fitting species did not significantly change the source apportionment results for ore and diesel exhaust. The estimated contributions (as a percentage of total explained mass) for each test stayed within 5% of the attribution achieved with the default species. Removal of any of these fitting species resulted in a decrease in CMB performance. The absence of semivolatile or total speciated organics led to an underestimation of the total mass. Excluding carbon measured by TOR yielded an overestimation of the mass. On the basis of these results, the default fitting species were retained in the final source apportionment. During the final source apportionment, performance of the CMB was optimized by excluding individual chemical species that did not fit well (ie, R/U greater than 2). Alternatively, chemical species not included in the default list that fit the CMB estimates well were included to improve model performance. Consistent with the sensitivity test results, exclusion or inclusion of individual species did not change the apportionment results.

## RESULTS

This section presents the summary results in figures and tables. Analytic uncertainties based on the precision of replicate analyses and of air volume measurements were calculated for each individual species. (The full dataset, together with the uncertainty for each measurement, is available in Appendices B and C.)

### SOURCE SAMPLES

#### Diesel Vehicle Emissions

***Estimated Emission Rates*** Emissions from vehicles tested in this program were collected as described in the Methods section under Source Sampling. The sampling system consisted of a heated sample line that was inserted up the exhaust pipe and would draw a sample from that point into the dilution sampler. Table 5 presents some results from the emissions testing program. The dilution ratio multiplied by the concentration found in the diluted stream gives the concentration in the exhaust. Total sample volume is the amount of sample pulled through the heated sample line. The flow in the sample line was determined by a heated Venturi. The exhaust volume is the volume of flow that came out the exhaust pipe. This was

**Table 5.** Emissions Data from Source Testing

| Vehicle ID[a] | Engine | Dilution Ratio | Volume ($m^3$) Sample | Volume ($m^3$) Exhaust | Fraction Sampled | Fuel Used (gal) | Emissions (mg/gal) |
|---|---|---|---|---|---|---|---|
| ULD004 | Detroit 50 Series DDEC | 19 | 2 | 181 | 0.011 | 1.6 | 0.55 |
| ULD005 | Detroit 50 Series DDEC | 21 | 1.4 | 181 | 0.008 | 1.6 | 1.20 |
| ULD008 | Detroit 4-71TI DDEC | 22 | 1.6 | 98 | 0.017 | 1.3 | 0.96 |
| ULD008R | Detroit 4-71TI DDEC | 21 | 1.6 | 95 | 0.017 | 1.3 | 0.56 |
| ULD024 | Caterpillar 3306 DITA | 22 | 1.6 | 250 | 0.006 | 1.6 | 0.62 |
| ULD024R | Caterpillar 3306 DITA | 22 | 1.6 | 250 | 0.006 | 1.6 | 0.70 |
| UHT002 | Detroit 4-71TI DDEC | 20 | 1.7 | 162 | 0.010 | 1.4 | 0.17 |
| UHT005 | Detroit 4-71TI DDEC | 21 | 1.6 | 235 | 0.007 | 1.6 | 0.99 |
| UHT006 | Detroit 60 Series DDEC | 22 | 1.6 | 281 | 0.006 | 2.5 | 1.60 |
| UHT022 | Detroit 60 Series DDEC | 21 | 1.7 | 319 | 0.005 | 1.6 | 1.40 |
| UHT024 | Detroit 60 Series DDEC | 19 | 1.8 | 276 | 0.006 | 2.0 | 0.43 |
| UTR009 | Kubota D1503-L-A | 19 | 1.9 | 17 | 0.113 | 0.2 | 0.17 |

[a] An "R" after the identification indicates the run was a replicate.

determined by the revolutions of the motor and the pressure delivered by the turbocharger. These data were collected by the electronic control systems for those vehicles that had them and were determined manually for the other 2 vehicles. This approach collected only approximately 1% to 2% of the total exhaust stream, except in the case of the much smaller Kubota engine, for which approximately 11% of the exhaust was collected. This is given as the fraction sampled, which is necessary for back-calculating to the total amount emitted. Fuel consumption was determined by the electronic engine data systems, which showed fuel flow at the various load settings, and confirmed by carbon dioxide. For the 2 vehicles that did not have electronic data systems on the engines, values for fuel consumption were estimated from similar-sized engines.

The mass emissions rates were determined by taking the mass determined in the diluted exhaust and multiplying by the dilution ratio to get the concentration in the exhaust stream. This was then multiplied by the exhaust volume to give the total mass emitted. To normalize the exhaust emissions, we divided by the fuel consumed to produce an emission rate in terms of mass emitted per gallon of fuel consumed. In on-road motor vehicle emissions work, emissions are often quantified in terms of mass of pollutant per unit distance driven; however, this is not a useful metric in the underground mine environment. The expression of emissions in terms of mass per gallon of fuel consumed is more useful because a gallon of fuel used is a more precise metric. This value of mass emitted per gallon of fuel is, however, only an estimate based on measured fuel consumption reported by the vehicles' onboard computers and was not externally verified.

Two replicate tests were conducted in this program. Replicates were the same vehicle and the same setup, run as closely as possible a second time with fresh sampling media. The 2 vehicles tested twice were ULD008 and ULD024. In terms of mass emissions rate, ULD024 was reproducible with emissions of 0.62 and 0.70 mg/gal for the replicate tests. ULD008 was less reproducible with emissions of 0.96 and 0.56 mg/gal for the 2 tests. The significant difference in emissions between the first and second ULD008 tests may be related to the engine not having been warmed up for the first test. Many variables can affect emission rates, including how warm the engine is and how precisely the sequence of load and idle periods is reproduced. Finally, we recognize that the engines themselves can have significant variability even if all external parameters are constant.

Within this dataset are different vehicles that have the same engine type. These vehicles allowed us to consider the variability of emissions from different individual engines. Unfortunately, we do not know the complete history of these engines although we do know the age of the engines (represented by total engine hours). For the 3 Detroit 60 Series DDEC engines, the emissions ranged from 0.43 to 1.6 mg/gal and were not directly correlated with engine hours (although all 3 vehicles had similar hours ranging from 6,750 to 9,650). The 2 Detroit 50 Series DDEC engines showed a larger variability with emissions of 0.55 and 1.2 mg/gal, but the older vehicle (12,800 hours) had

lower emissions than the newer vehicle (1,648 hours). Of the 3 Detroit 4-71TI engines, 2 were very new (assuming the data were accurate) at 1.4 and 100 hours, while the last had 1,680 hours. Emissions were lowest for the newest at 0.17 mg/gal, but the other 2 were nearly the same at 0.99 and 0.96 (replicate of 0.56) mg/gal. Thus, we cannot say that engine age was directly associated with emissions, and we acknowledge that individual examples of the same engine can vary appreciably.

The majority of emissions from the vehicles tested were from carbon although several vehicles emitted considerable sulfate. Table 6 presents the emissions in terms of concentration in raw exhaust for mass, carbon, and sulfate, along with ratios of various emissions. The mass is simply gravimetrically determined mass on Teflon filters. Sum of species is the total of all chemically determined species, including carbon, ions, and other elements. TC is the carbon determined by TOR. As can be seen, in most cases the majority of the species determined were carbon. The ratio of sum of species to mass shows a few deviations from 1.0 which suggest that either the mass or chemical determination was in error. Mass errors may have occurred if the filter was not adequately equilibrated either prior to the preexposure weighing or after sampling. Because the majority of the chemical species determined came from carbon, carbon was often viewed as the possible source of error. The quartz filters used to determine carbon can adsorb hydrocarbon vapors from the airstream, thus leading to higher values in the carbon analysis. This process does not occur with the Teflon filters used to determine gravimetric mass. The other possible source of error is that the Teflon and quartz filters have individual flow controls, and nonrandom errors in one or the other flow system could bias the result. This error is unlikely, however, because we used the same rotameter for both channels. Reconstructed mass (RCM) was used in the generation of profiles to help eliminate any variability in mass determination. The objective of source testing was not to obtain emission rates but rather to obtain source profiles that were less variable.

Sulfate emissions in this study ranged from 0.07 to 3.18 mg/m$^3$ in the raw exhaust. This was 1% to 24% of the total mass emitted. Another parameter of interest is the ratio of EC to TC or EC to total mass. In this study, EC was on average 40% of the TC, which is somewhat low for typical on-road diesel vehicles. The EC-to-mass ratio is affected by the issues discussed previously with respect to mass and thus can be considerably different from the EC/TC ratio.

The emission rates and OC/EC ratios determined in this manner were based on a uniform set of operating conditions that may or may not accurately represent the operating environment in the underground mine. As described in Source Sampling, we used a test cycle consisting of a 5-minute idle (4 times) followed by a 2-minute load (3 times) to near 100% engine rating. Thus, for a 26-minute total test cycle, 77% of the time the engine was idle and only 23% of the time it was loaded. The diesel equipment in the underground mine was subjected to heavy load most of the time; therefore, it is unlikely that our test cycle represented driving conditions inside the mine. Without a chassis dynamometer, however, the diesel engine could

**Table 6.** Carbon and Sulfate Emissions (in mg/m$^3$) from Source Tests Calculated to Undiluted Exhaust Concentration

| Vehicle ID[a] | Mass | Sum of Species | Total Carbon | Sum/Mass | Sulfate | Organic Carbon | Elemental Carbon | EC/ TC | EC/ Mass |
|---|---|---|---|---|---|---|---|---|---|
| ULD004 | 4.9 | 6.7 | 5.8 | 1.38 | 0.23 | 3.7 | 2.1 | 0.37 | 0.44 |
| ULD005 | 10.4 | 10.1 | 7.7 | 0.97 | 0.32 | 5.6 | 2.1 | 0.28 | 0.21 |
| ULD008 | 12.4 | 14.0 | 12.6 | 1.12 | 0.14 | 6.8 | 5.8 | 0.46 | 0.46 |
| ULD008R | 7.4 | 11.2 | 10.9 | 1.51 | 0.07 | 6.7 | 4.2 | 0.39 | 0.57 |
| ULD024 | 4.0 | 8.3 | 7.8 | 2.09 | 0.34 | 4.8 | 3.0 | 0.39 | 0.76 |
| ULD024R | 4.5 | 7.0 | 5.8 | 1.55 | 1.09 | 3.3 | 2.4 | 0.42 | 0.54 |
| UHT002 | 1.4 | 6.1 | 5.9 | 4.27 | 0.09 | 4.7 | 1.2 | 0.20 | 0.82 |
| UHT005 | 6.8 | 9.1 | 8.1 | 1.34 | 0.87 | 3.5 | 4.5 | 0.56 | 0.67 |
| UHT006 | 14.3 | 13.4 | 9.2 | 0.94 | 3.18 | 4.4 | 4.7 | 0.51 | 0.33 |
| UHT022 | 7.1 | 10.2 | 9.8 | 1.46 | 0.10 | 4.9 | 5.0 | 0.51 | 0.71 |
| UHT024 | 3.1 | 6.2 | 5.8 | 1.99 | 0.11 | 3.3 | 2.5 | 0.43 | 0.80 |
| UTR009 | 2.3 | 8.3 | 8.0 | 4.20 | 0.11 | 5.4 | 2.7 | 0.33 | 1.35 |

[a] An "R" after the identification indicates the run was a replicate.

not be subjected to a load for longer than 2 minutes without risking damage to the engine. As reported in the literature (Shi et al 2000), the OC/EC distribution in diesel emissions will shift depending on operating conditions and load on the vehicle. A vehicle operating under heavy load will have an OC/EC distribution shifted toward EC, whereas a vehicle at idle or low load can have much more OC present. For this reason, the emission rates and OC/EC ratios presented in this section are only reasonable estimates and not necessarily exactly what would be found in the mine.

***MOUDI Size Distribution of Particles*** The MOUDI samples, described in Source Sampling, were collected to determine the size distribution of emitted particles. Figure 1 shows the diluted exhaust distributions of mass, sulfate, and TC as a fraction of $PM_{2.5}$ mass. These 3 are the main species of interest in the exhaust. No organic species measurements were made on the MOUDI samples. The distribution is shown at each size fraction that was collected. In all cases, stage 7 (0.105 µm) mass is missing because a leak occurred at that stage when the medium was installed. Running without medium did not affect the other stages.

The mass distributions were approximately bimodal with a peak at the 0.148-µm stage and another at the 0.054-µm stage. The exceptions were ULD005 and ULD024, which showed a reasonably large 1.0-µm mass. In the case of ULD005, this result was somewhat suspect because the chemical analysis did not show a similar mass (that is, the sum of chemical species showed only a fraction of the gravimetrically determined mass). The afterfilter was labeled as the stage below 0.054 µm because it collected all material less than the lowest stage. Note that this stage was lost in the UHT006 sample. Some high values were obtained for this stage, especially in the case of UHT022. Such high values are fairly typical in source sampling of motor vehicles and may have resulted, in part, from insufficient time for coagulation of and condensation on these particles prior to sampling.

Sulfate concentration was very low in nearly all the samples with the exception of UHT006, which showed

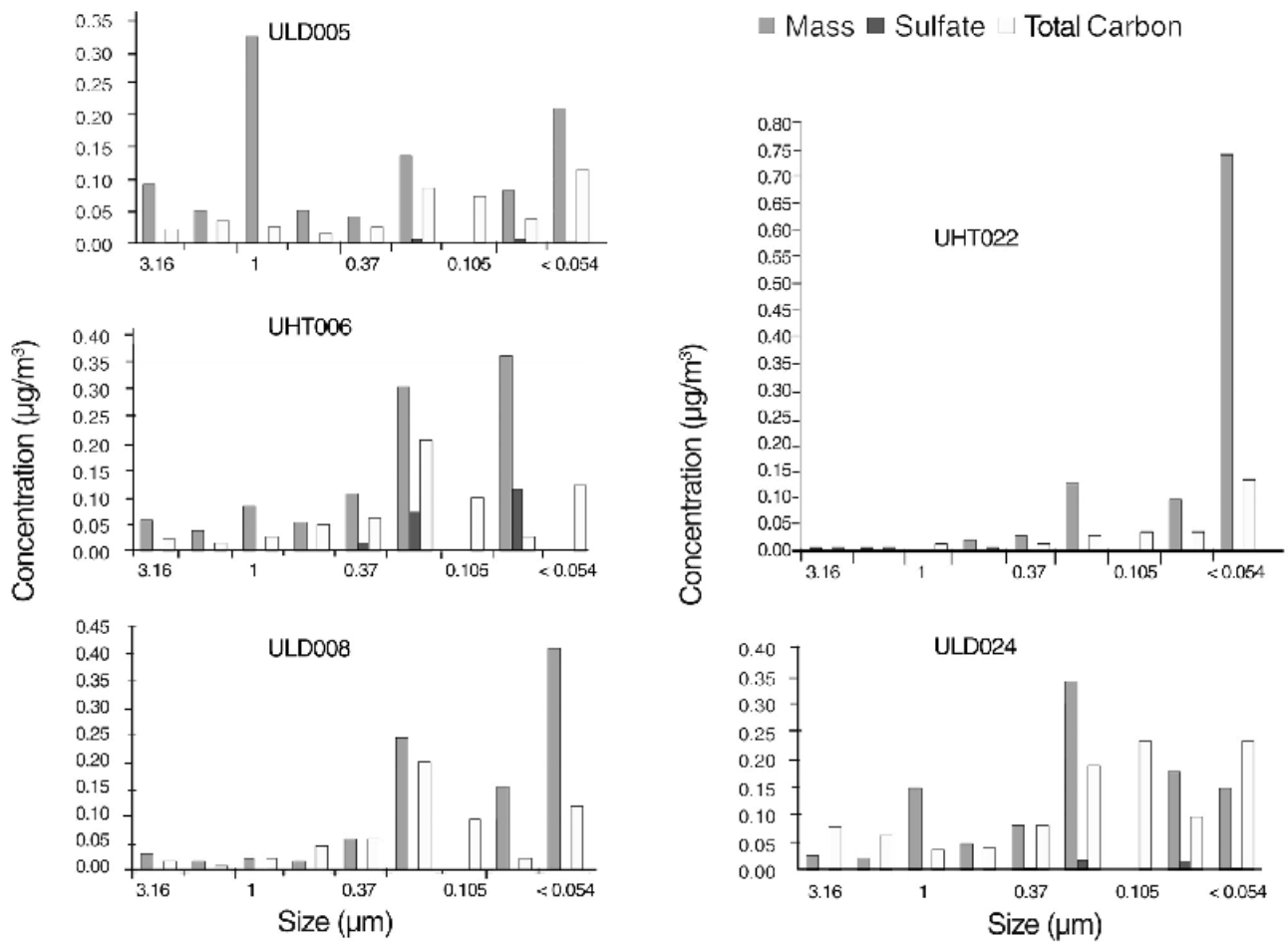


**Figure 1. Micro-orifice uniform deposit impactor (MOUDI) size distributions for emissions from some vehicles in the source testing.**

some sulfate in the smaller size fractions. Carbon was typically a large fraction of the lower mass sizes and generally made up most of the observed mass.

The interesting feature of the MOUDI data was the large mass fraction in the afterfilter. This filter collected particles that were not trapped in the higher stages, including the ultrafine fraction, which in a few cases was a significant fraction of the total mass. To evaluate the importance of this phenomenon, however, we must consider the sample collection method. For the source samples, the dilution sampler diluted the exhaust with nearly particle-free air so that the ultrafine particles had less material to collide with and thus might not grow to larger sizes by coagulation or impaction. In the underground mine, however, these emissions were released into an environment with a large number of existing particles, so coagulation with existing accumulation mode particles might occur, changing the particle size distribution.

***Organic Compounds*** Figure 2 shows the concentrations of gas-phase PAHs (A), semivolatile PAHs (B and C), and particle-phase PAHs (D and E) in the raw exhaust (corrected for the dilution rate in the dilution tunnel) from the heavy-duty diesel equipment tested in the mine (see Table 1 for the description of the equipment). ULD008R and ULD024R represent 2 replicate tests. Although naphthalene was usually the most abundant PAH in the exhaust, it is not presented in Figure 2 because comparison with the diffusion denuder data (B Zielinska, unpublished results) showed that naphthalene was not collected quantitatively on the PUF/XAD-4/PUF cartridge under these conditions. Some breakthrough was also observed for 1- and 2-methylnaphthalene. For clarity of presentation, methyl, dimethyl, and trimethyl isomers of naphthalene, fluorene, phenanthrene, and fluoranthene/pyrene, although quantified as individual isomers, are summed into corresponding methyl, dimethyl, and trimethyl groups of isomers. Also, PAHs, present in low concentration within each group, are not shown in Figure 2 (the full dataset is available as source profiles in Appendix B).

The gas-phase PAHs were the most abundant in diesel equipment emissions (Figure 2). Among the semivolatile PAHs, phenanthrene was usually the most abundant, followed by fluorene, methylphenanthrenes, and dimethylphenanthrenes. Exceptionally high emissions of phenanthrene and anthracene were observed for ULD005. This vehicle (6-cubic-yard load capacity) also showed high emissions of particle-phase PAHs, especially benzo[*a*]anthracene, chrysene, benzo[*b*+*j*+*k*]fluoranthene, and benzo[*e*]pyrene, which is in contrast to all other vehicles tested.

Figure 3 shows concentrations of the most abundant steranes (A) and hopanes (B) in the raw diesel exhaust. The concentrations of these compounds were not high, and the patterns were similar for all vehicles. In addition, the concentrations of hopanes and steranes in the shop blank sample were comparable to those observed in vehicle emissions. (The names and mnemonics of the 19 hopanes and 17 steranes for which all source and ambient samples were analyzed are available in Table 7. A complete list is in Appendix B.)

The concentrations of several nitro-PAHs (1-nitronaphthalene, 1-methyl-2-nitronaphthalene, 2- and 4-nitrobiphenyl, 3-nitrofluoranthene, and 1-nitropyrene) also were measured in the diluted diesel exhaust (Figure 4). Compounds were measured in diluted exhaust, but concentrations were calculated to raw exhaust concentrations based on dilution ratio. The concentrations of more volatile nitro-PAHs, 1-nitronaphthalene and 1-methyl-2-nitronaphthalene (other isomers of methylnitronaphthalene and nitronaphthalene were not measured), were highest in the exhaust of all vehicles, in the range of a few micrograms per cubic meter. Also, 2-nitrobiphenyl was always more abundant than 4-nitrobiphenyl. The concentrations of higher molecular weight nitro-PAHs, 3-nitrofluoranthene and 1-nitropyrene, were much lower, in the range of 0.1 to 0.2 $\mu g/m^3$ of the raw exhaust.

### Other Sources of Fine PM

Figure 5 shows the profiles of steranes and hopanes for the rock drill oil used in the mine, 15/40-weight and 40-weight lubricating oil used for the diesel equipment, and hydraulic oil. Figure 6 shows the PAH profiles for the same oils. For clarity, PAHs are combined into 3 groups, gas phase PAHs (A), semivolatile PAHs (B and C), and particle-phase PAHs, (D and E) corresponding to the descriptions for Figure 2. The drill oil was enriched in particle-phase PAHs in comparison with the other types of oil. It also had much higher contents of hopanes and steranes than the other oils used in the mine. (The organic and inorganic profiles for the oils are available in Appendix B.)

Figure 7 shows the compositions of 2 types of rock found in the Carlin East gold mine, gold ore and waste rock. The data given are for 2.5-µm particles only and were obtained by the resuspension method. The main constituents were silica, aluminum, and iron (corrected for their corresponding oxides) with a small amount (less than 10%) of OC and EC. Possibly a large fraction of the measured OC was $CO_3$ (terrestrial carbon). No special effort was made to account for carbonate relative to OC.

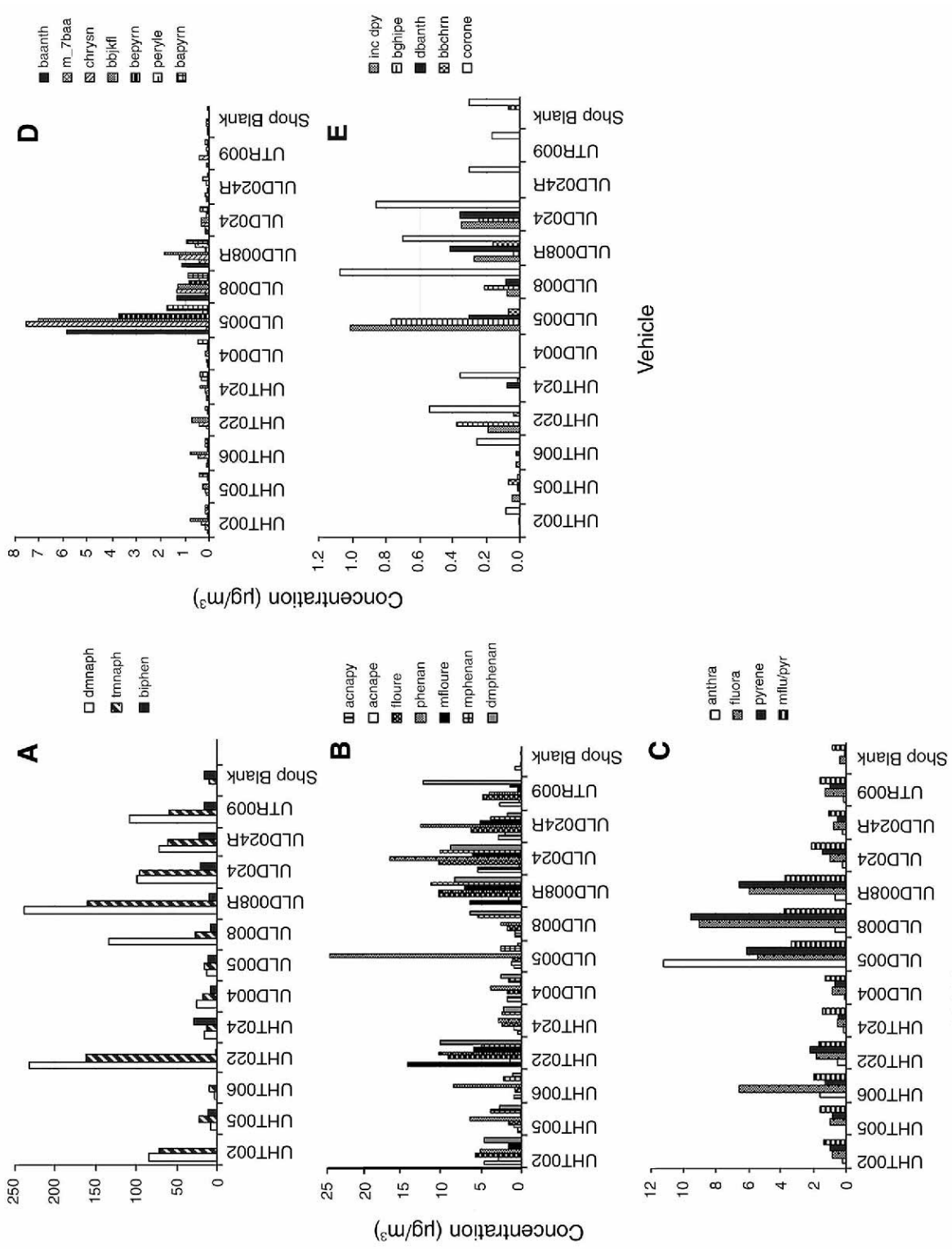


**Figure 2. Concentrations of PAHs in raw exhaust from diesel equipment tested in the mine.** ***A.*** Gas-phase PAHs: dimethylnaphthalenes, trimethylnaphthalenes, and biphenyl. ***B.*** Semivolatile PAHs: acenaphthylene, acenaphthene, fluorene, phenanthrene, methylfluorene, methylphenanthrene, and dimethylphenanthrene. ***C.*** Semivolatile PAHs: anthracene, fluoranthene, pyrene, and methylfluoranthenes/pyrenes. ***D.*** Particle-phase PAHs: benz[*a*]anthracene, 7-methylbenz[*a*]anthracene, chrysene, benzo[*b+j+k*]fluoranthene, benzo[*e*]pyrene, perylene, and benzo[*a*]pyrene. ***E.*** Particle-phase PAHs: indeno[*cd*]pyrene, benzo[*ghi*]perylene, dibenzo[*a*]anthracene, benzo[*b*]chrysene, and coronene.

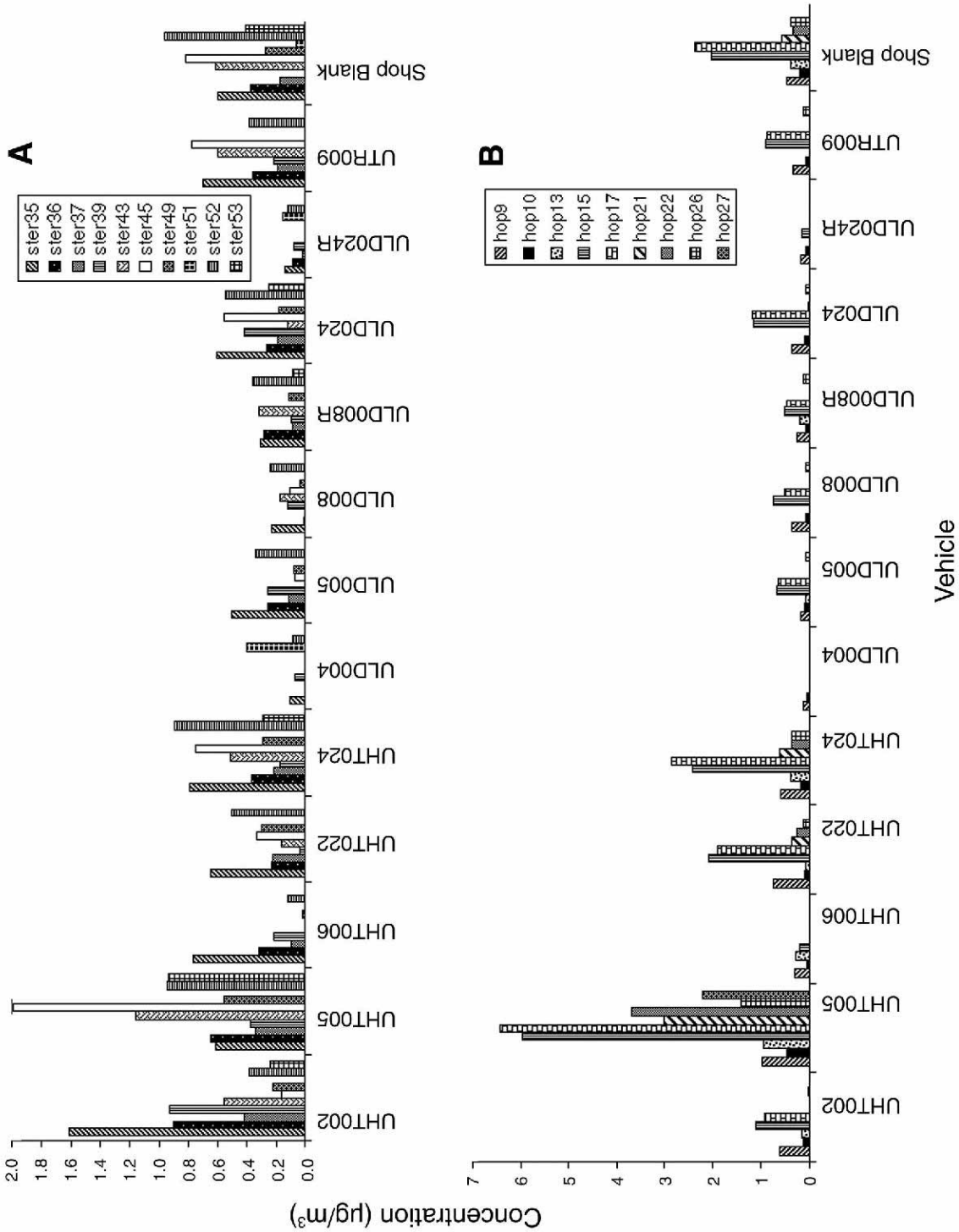


**Figure 3. Concentrations of particle-phase steranes (*A*) and hopanes (*B*) in raw exhaust from diesel equipment tested in the mine.**

**Table 7.** Short Forms and Names of Hopanes and Steranes Tested in all Source and Ambient Samples

| | |
|---|---|
| ster35 | C27-20S-13β(H),17a(H)-Diasterane |
| ster36 | C27-20R-13β(H),17a(H)-Diasterane |
| ster37 | C27-20S-13a(H),17β(H)-Diasterane |
| ster38 | C27-20R-13a(H),17β(H)-Diasterane |
| ster39 | C28-20S-13β(H),17a(H)-Diasterane |
| ster42 | C27-20S5a(H),14a(H)-Cholestane |
| ster43 | C27-20R5a(H),14β(H)-Cholestane |
| ster44 | C27-20S5a(H),14β(H), 17β(H)-Cholestane |
| ster45 | C27-20R5a(H),14a(H),17a(H)-Cholestane |
| ster46 | C28-20S5a(H),14a(H),17a(H)-Ergostane |
| ster47 | C28-20R5a(H),14β(H), 17β(H)-Ergostane |
| ster41 | C29-20R-13a(H),17ß(H)-Diasterane |
| ster49 | C28-20R5a(H),14a(H),17a(H)-Ergostane |
| ster50 | C29-20S5a(H),14a(H),17a(H)-Stigmastane |
| ster51 | C29-20R5a(H),14β(H), 17β(H)-Stigmastane |
| ster52 | C29-20S5a(H),14β(H), 17β(H)-Stigmastane |
| ster53 | C29-20R5a(H),14a(H),17a(H)-Stigmastane |
| hop9 | C27-Tetracyclic terpane |
| hop10 | C27-Tetracyclic terpane |
| hop11 | C28-Tetracyclic terpane |
| hop12 | C28-Tetracyclic terpane |
| hop13 | 18a(H),21β(H)-22,29,30-Trisnorhopane |
| hop14 | 17a(H),18a(H),21β(H)-25,28,30-Trisnorhopane |
| hop15 | 17a(H),21β(H)-22,29,30-Trisnorhopane |
| hop16 | 17a(H),18a(H),21β(H)-28,30-Bisnorhopane |
| hop17 | 17a(H),21β(H)-30-Norhopane |
| hop18 | 18a(H),21β(H)-30-Norneohopane |
| hop19 | 17a(H),21β(H)-Hopane |
| hop20 | 17β(H),21a(H)-Hopane |
| hop21 | 22S-17a(H),21β(H)-30-Homohopane |
| hop22 | 22R-17a(H),21β(H)-30-Homohopane |
| hop23 | 17β(H),21β(H)-Hopane |
| hop24 | 22S-17a(H),21β(H)-30,31-Bishomohopane |
| hop25 | 22R-17a(H),21β(H)-30,31-Bishomohopane |
| hop26 | 22S-17a(H),21β(H)-30,31,32-Trisomohopane |
| hop27 | 22R-17a(H),21β(H)-30,31,32-Trishomohopane |

## AMBIENT SAMPLES

### Inorganic Composition

In addition to the intercomparison samples, ambient samples were collected at 2 different sites for 3 consecutive shifts. (The complete set of data is available in Appendix C.) In this analysis we present the RCM, not the gravimetric mass. Figure 8 compares the mass determined gravimetrically and the RCM, determined by summing the components analyzed and adding the associated oxygen and other elements for mass correction when appropriate. This required multiplying OC by 1.2, aluminum by 1.89, silicon by 2.14, and iron by 1.43. As can be seen in Figure 8, the RCM underestimated the gravimetric mass by approximately 22%. The difference may have been due to water. The underground environment was warm, approximately 30°C, and very humid. Although we did not measure relative humidity underground, the air appeared to be nearly saturated with water. Water will hydrate many inorganic and organic compounds and was weighed, but not measured, in the analysis. The filters were equilibrated in a room with constant 30% relative humidity before and after sampling; however, this procedure may not truly equilibrate some species in which the waters of hydration are held very tightly or particles are layered such that the lower layers are not exposed to the air. This problem has been addressed in detail (Andrews et al 2002) with the conclusion that water may be a significant contributor to the gap between chemical species and gravimetric mass.

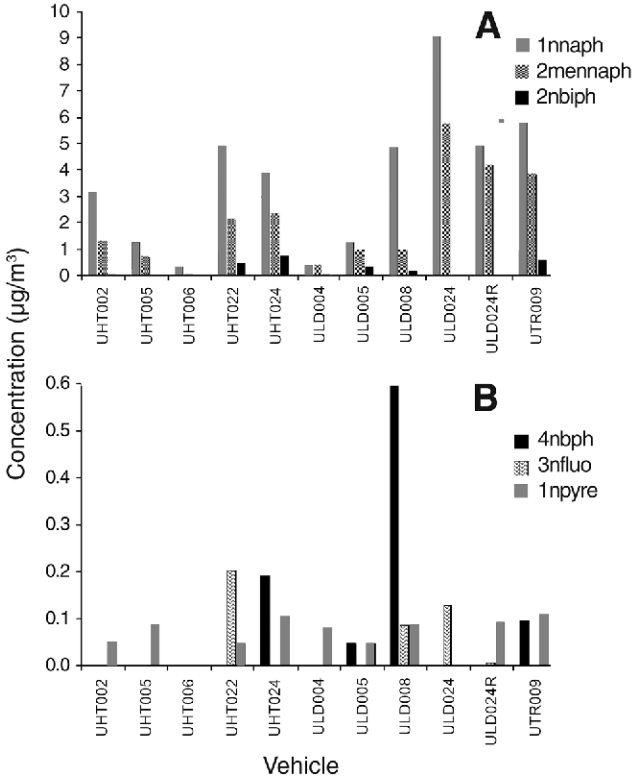


**Figure 4. Distribution of nitro-PAHs in diesel equipment exhaust. *A.*** 1-Nitronaphthalene, 1-methyl-2-nitronaphthalene, and 2-nitrobiphenyl. ***B.*** 4-Nitrobiphenyl, 3-nitrofluoranthene, and 1-nitropyrene.

Figure 9 shows the RCM, TC, and EC concentrations in samples at site CE1. The EC averaged about 75% of the TC, and the proportions were nearly the same at both sampling

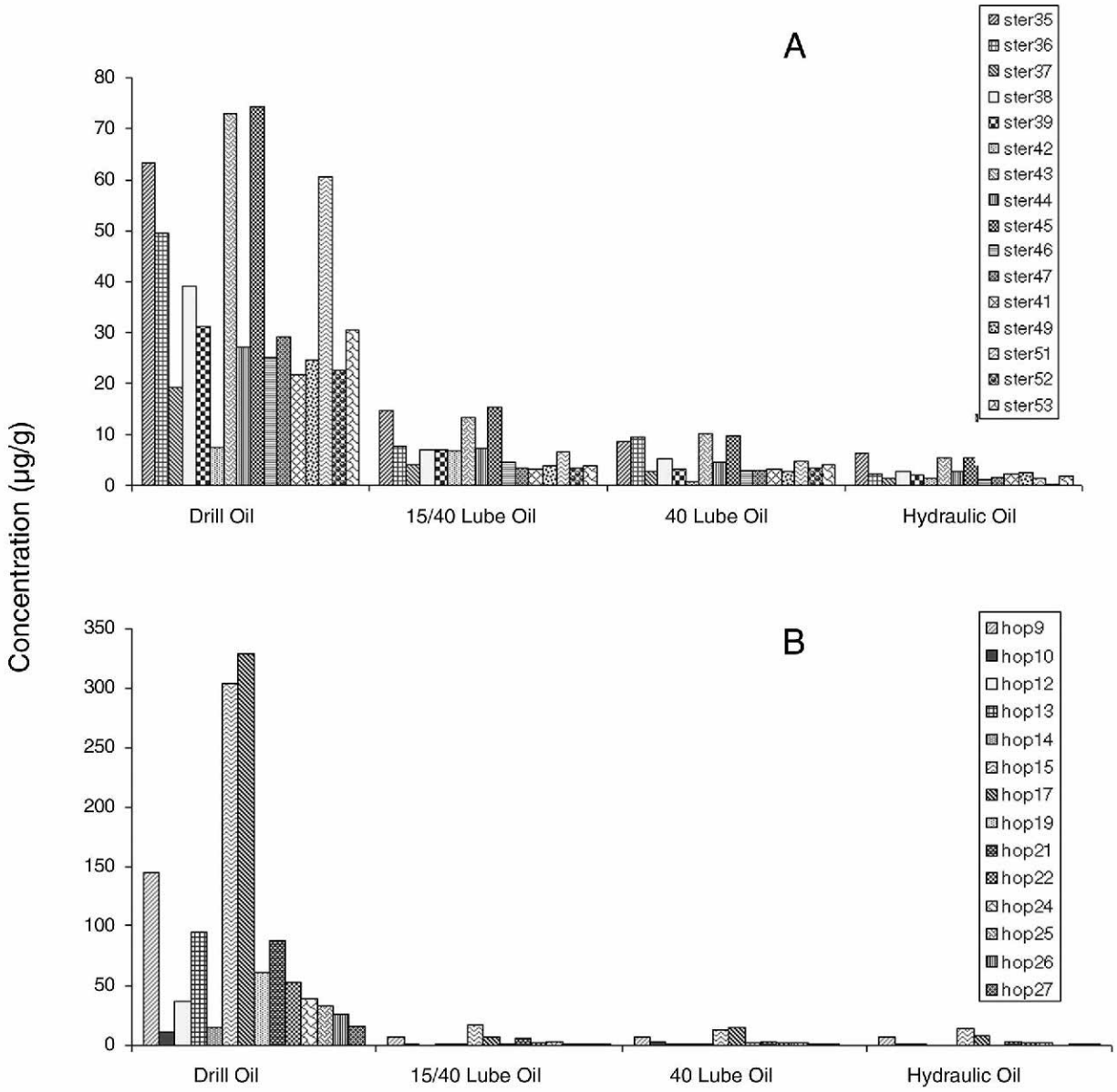


**Figure 5. Composition of steranes (*A*) and hopanes (*B*) in oils used in the mine: rock drill oil, 15/40-weight lubricating oil, 40-weight lubricating oil, and hydraulic oil.**

sites. The TC was approximately 72% of the mass; again, the proportions were nearly the same at both sites. On average, sulfate was approximately 10% of RCM. For elements associated with geologic material, silicon was approximately 6% of mass and very consistent in all samples. Aluminum was approximately 1% in all samples.

Gold was one of the elements measured, and because the samples were taken in a gold mine, we were curious whether much would be detected. No sample showed gold concentrations above the uncertainty of the XRF measurement. The practical detection limit for gold is about 50 ng/filter in the XRF, which for a 10-$m^3$ sample is approximately 0.005 µg/$m^3$. The mined ore contains approximately 0.4 per ton of ore, which is approximately 14 µg of gold per gram of ore. From the source apportionment we know that at most 10% of the $PM_{2.5}$ is geologic in origin; thus, gold would be only 14 ng on a filter sample of 10 $m^3$, with a concentration of 1 mg/$m^3$. This would be below the limit of detection.

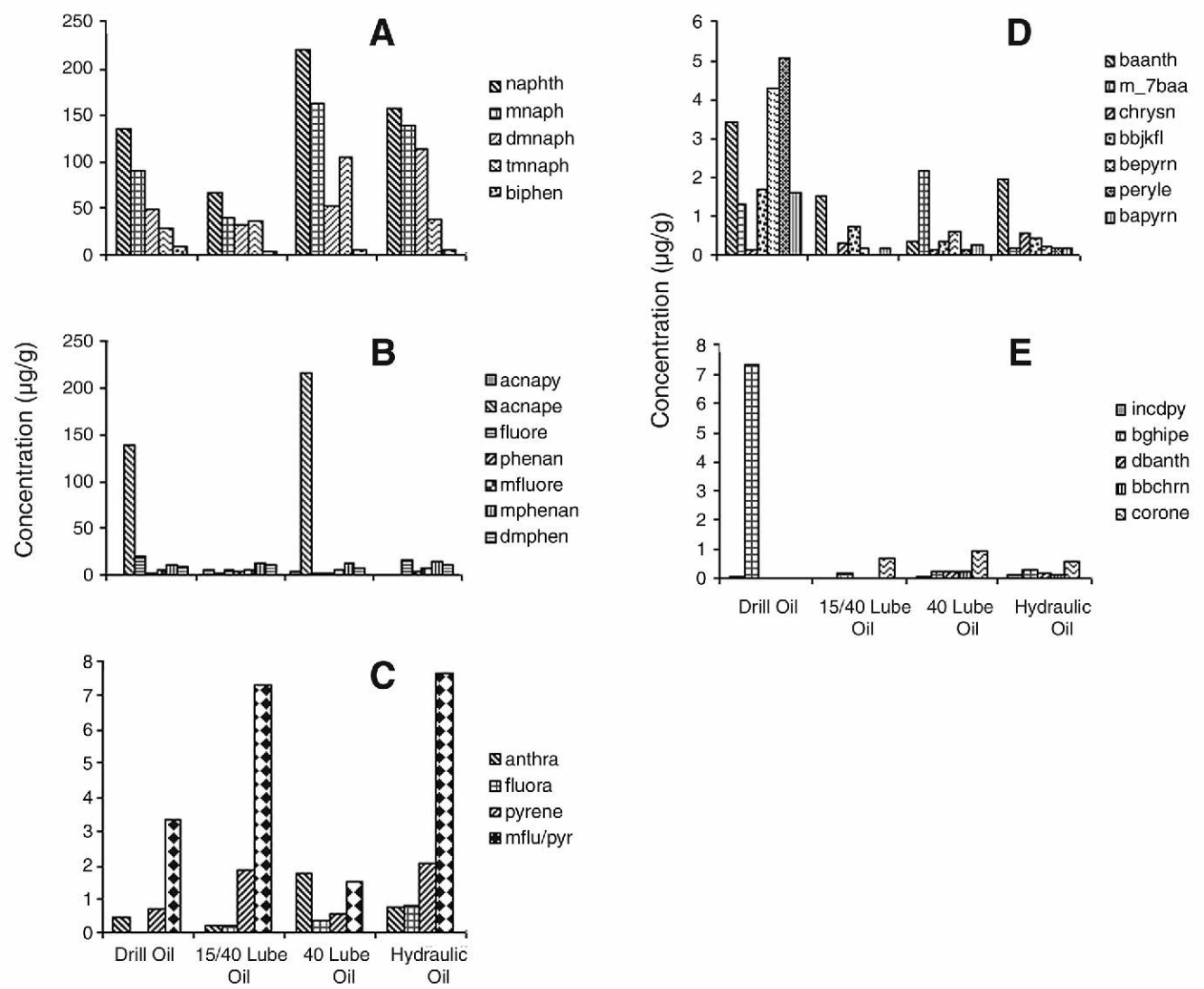


**Figure 6. Compositions of PAHs observed in oils used in the mine: rock drill oil, 15/40-weight lubricating oil, 40-weight lubricating oil, and hydraulic oil. *A.*** Naphthalene, methylnaphthalenes, dimethylnaphthalenes, trimethylnaphthalenes, and biphenyl. ***B.*** Acenaphthylene, acenaphthene, fluorene, phenanthrene, methylfluorene, methylphenanthrene, and dimethylphenanthrene. ***C.*** Anthracene, fluoranthene, pyrene, and methylfluoranthenes/pyrenes. ***D.*** Benz[*a*]anthracene, 7-methylbenz[*a*]anthracene, chrysene, benzo[*b*+*j*+*k*]fluoranthene, benzo[*e*]pyrene, perylene, and benzo[*a*]pyrene. ***E.*** Indeno[*cd*]pyrene, benzo[*ghi*]perylene, dibenzo[*a*]anthracene, benzo[*b*]chrysene, and coronene.

The concentrations of RCM, TC, and EC for the lower site, CE2, are shown in Figure 10. As discussed in the Methods section, this sampling site was moved after the first sample owing to changes in mining activity. There was nearly 2.5 times as much mass at the second location as at the first location; however, the composition stayed relatively constant. The largest difference in composition was in the night shift sample on June 4, 1999, with TC making up only 69% of mass, but the proportion of EC was still similar to the other samples at 79% of TC. For the other 2 sampling periods the concentrations were 76% and 87% TC as a percentage of mass. As with the upper site, the geologic elements constituted a relatively small fraction of the samples, and all samples were similar in composition. Aluminum was approximately 1% of RCM and silicon was less than 6%.

Two MOUDI samplers were also run at the CE1 upper site during the 3 shifts. However, failure of the sampling equipment resulted in loss of the Teflon media sample from the night shift starting on June 4, 1999. (The complete set of MOUDI data is available in Appendix C.) Distributions of the mass and carbon are shown in Figure 11 for the 3 shifts during which samples were taken. For the night

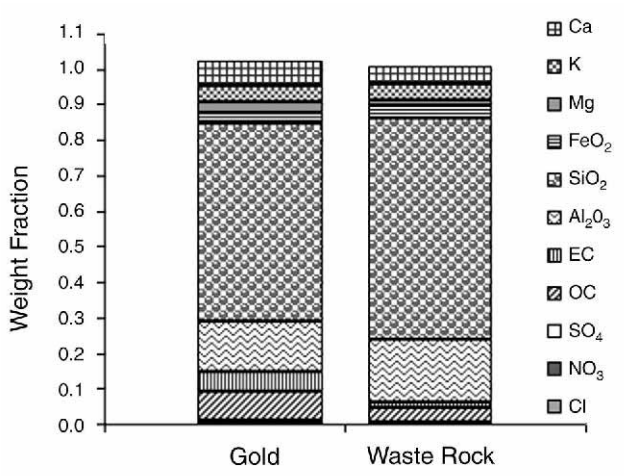


Figure 7. Composition of 2 ore samples: gold ore and waste rock.

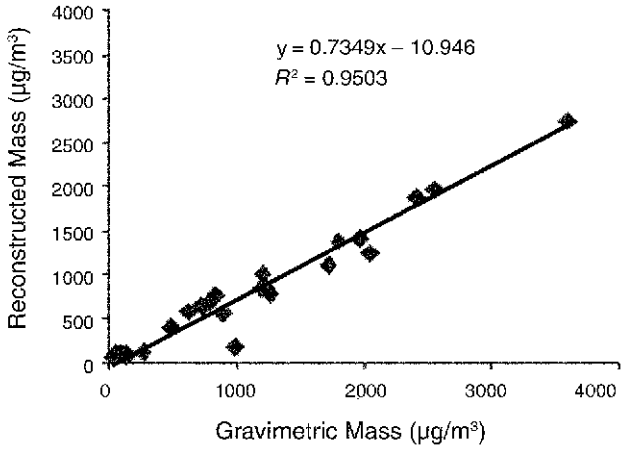


Figure 8. Gravimetric mass versus reconstructed mass for the ambient samples.

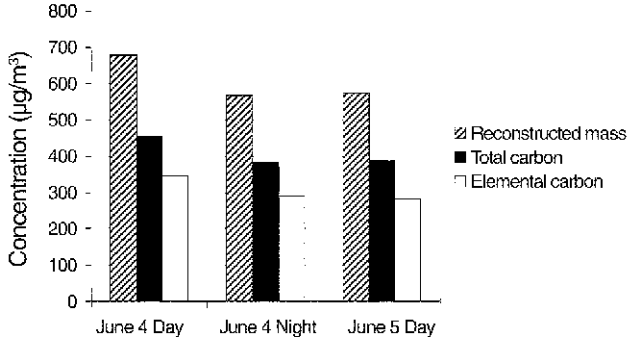


Figure 9. Concentrations at CE1 (upper site) for 3 sampling periods.

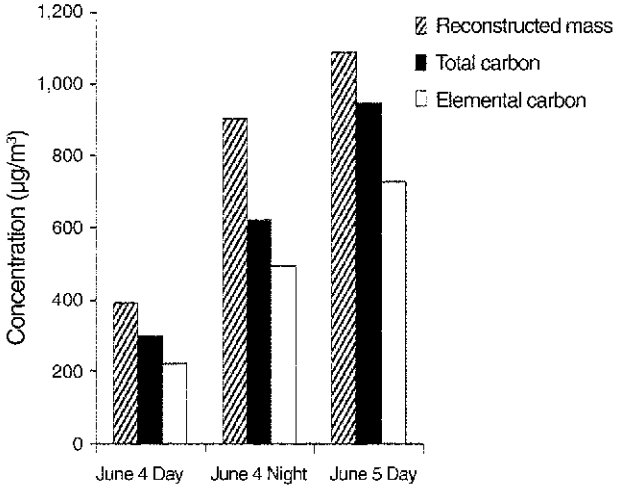


Figure 10. Concentrations at CE2 (lower site) for 3 sampling periods.

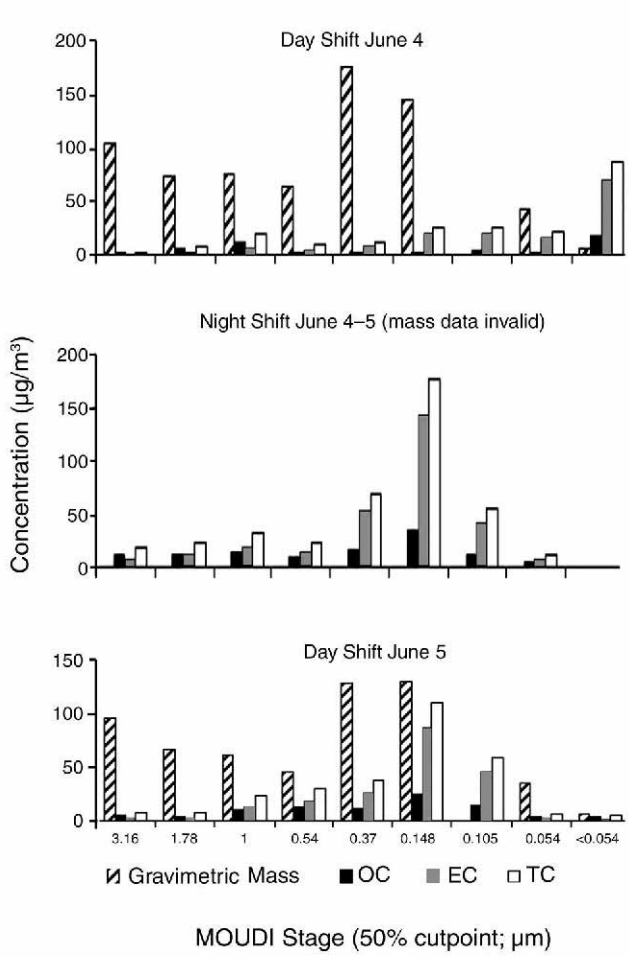


Figure 11. Distribution of mass, OC, EC, and TC in MOUDI samples of ambient air in the upper mine (CE1) for 3 shifts.

shift starting on June 4, 1999, there are no mass data. For presentation of the MOUDI data, we used gravimetric mass because there were several analytic challenges with this dataset, and we were interested in the size distribution for the in situ environment where these were collected. The MOUDI data were not used in the source apportionment. The distributions showed that a considerable amount of the mass was in the larger size fraction (3.16 µm), and that this fraction was composed of material other than carbon (carbon was only 2% to 10% of the mass in this size). This is reasonable since we expected the larger size fraction of PM to have mostly geologic material.

Carbon dominated the accumulation mode (0.148 to 0.105 µm) in the sample for the day shift on June 5, 1999. The distributions appear skewed for the day shift on June 4, 1999. In the very small fractions the carbon was several times the mass. Since mass and carbon were collected on separate media and in separate samplers, some operational difference may have occurred between the 2 samplers. Also notable in Figure 11 is the relative lack of mass at the very lowest fraction. This is in contrast to the distributions seen in the source samples, where a considerable fraction of the mass was in the ultrafine category. The difference may have resulted from the difference in environments into which the emissions were being released. In the source testing, the emissions were released into clean, filtered, and nearly particle-free air. In the mine, the emissions were released into a mixture of fresh outdoor air, other diesel emissions, and dusts from the rock and other material. The additional particles in the mine very likely provided the necessary conditions for coagulation and growth of the ultrafine particles into the accumulation mode or larger. This may account for the difference in percentage of emissions in the ultrafine fraction.

The ratio of EC to TC changed throughout the distribution. For the day shift on June 4, 1999, the EC was 20% of TC in the 3.16-µm fraction and rose to 80% for all fractions less than 0.148 µm. Similar ratio changes were seen in the other 2 samples except that the lowest ratio was approximately 35% for the 3.16-µm fraction.

The size fractions of some common geologic materials are presented in Figure 12 for the 2 samples that had valid data. The elements aluminum, silicon, and iron are presented as their concentrations in the mine air, adjusted for the expected oxygen species present. Note that the scale is much less than that for mass and carbon. The size distributions show that the geologic materials were most prevalent in the largest fractions and nearly nonexistent in the smaller sizes. This is consistent with the hypothesis that the smallest particles were generally created by combustion processes only.

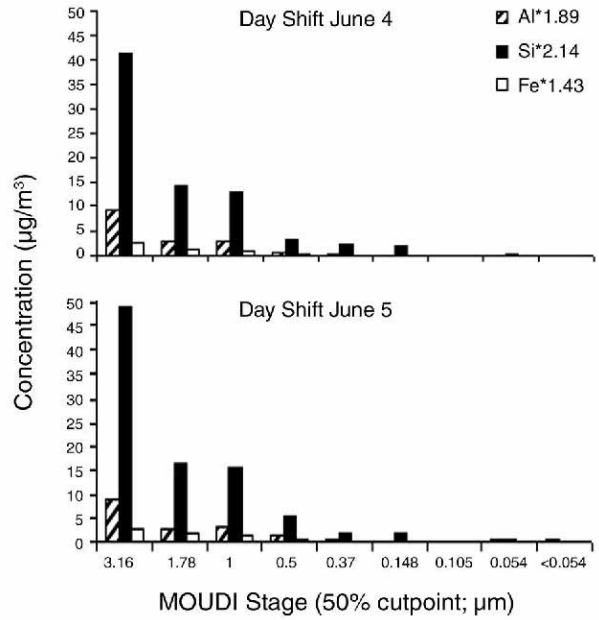


**Figure 12. Distribution of silicon, aluminum, and iron in 2 valid MOUDI samples.**

## Organic Composition

Figure 13 shows the concentrations of gas-phase PAHs (A), semivolatile PAHs (B and C), and particle-phase PAHs (D and E) in the mine air measured during 3 consecutive shifts in 2 locations, CE1 (upper site) and CE2 (lower site). As in the source samples the gas-phase PAHs were the most abundant in the ambient samples, followed by semivolatile PAHs, phenanthrene, methylfluorene, methylphenanthrene, and dimethylphenanthrene. Particle-phase PAHs were present in much lower concentrations, consistent with heavy-duty diesel equipment emissions as the main source of the PAHs.

Figure 14 shows the concentrations of hopanes and steranes measured in the ambient air in the mine. Although the absolute concentrations were different for different shifts, the relative concentrations (profiles) were similar, especially for hopanes.

Figure 15 shows concentrations of the same nitro-PAHs as shown in Figure 4 in the mine air measured during 3 consecutive shifts in 2 locations, CE1 (upper site) and CE2 (lower site). During the fourth shift only the upper location was sampled (CE1#4). As in the source samples of diesel exhaust, the lower molecular weight nitro-PAHs, 1-nitronaphthalene and 1-methyl-2-nitronaphthalene, were the most abundant in the ambient samples. Higher molecular weight nitro-PAHs were present in much lower concentrations, usually

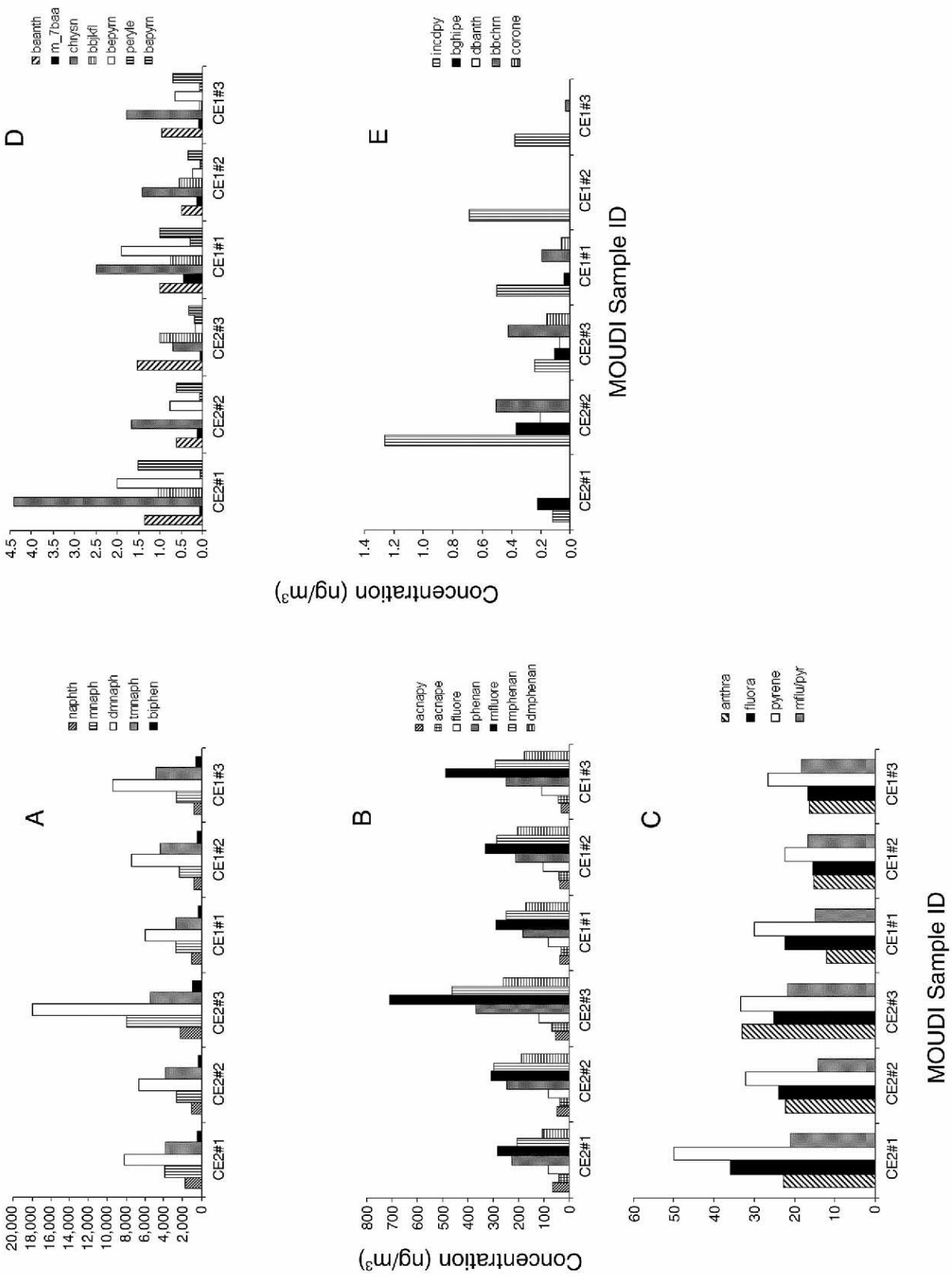


**Figure 13. Concentrations of PAHs in ambient air in the upper (CE1) and lower (CE2) mine sites.** ***A.*** Gas-phase PAHs: naphthalene, methylnaphthalenes, dimethylnaphthalenes, trimethylnaphthalenes, and biphenyl. ***B.*** Semivolatile PAHs: acenaphthylene, acenaphthene, fluorene, phenanthrene, methylfluorene, methylphenanthrene, and dimethylphenanthrene. ***C.*** Semivolatile PAHs: anthracene, fluoranthene, pyrene, and methylfluoranthenes/pyrenes. ***D.*** Particle-phase PAHs: benz[*a*]anthracene, 7-methylbenz[*a*]anthracene, chrysene, benzo[*b*+*j*+*k*]fluoranthene, benzo[*e*]pyrene, perylene, and benzo[*a*]pyrene. ***E.*** Particle-phase PAHs: indeno[*cd*]pyrene, benzo[*ghi*]perylene, dibenzo[*a*]anthracene, benzo[*b*]chrysene, and coronene.

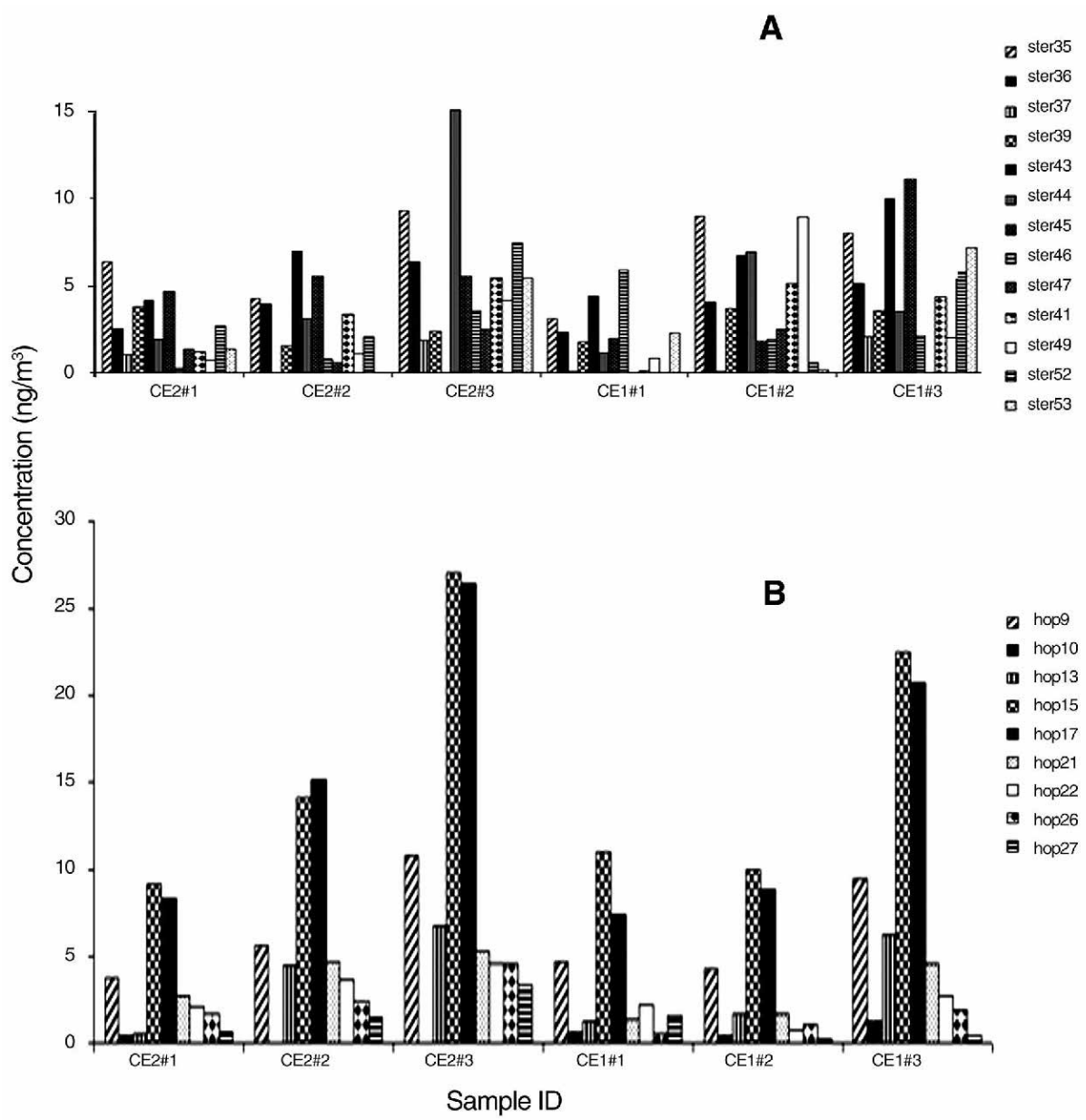


**Figure 14. Concentrations of particle-phase steranes (*A*) and hopanes (*B*) in ambient air in the mine.**

below 1 ng/m$^3$, consistent with heavy-duty diesel equipment emissions as the main source of these nitro-PAHs.

### Photoelectric Aerosol Sampler

The PAS was deployed at the upper site for several shifts as an experiment. This was not in the original program plan, but the opportunity arose to use this instrument at no cost to the project, and we decided to test it for applicability to future projects. Owing to difficulty in the computer interface, we were not able to obtain all of the data from the instrument; however, one complete dataset was obtained from approximately 1600 hours on June 5 to 0400 hours on June 6, 1999 (Figure 16). Each point on the graph represents the average of only 8 seconds of signal, so the fine detail can be seen. During the shift change that took place between approximately 1700 and 1900 hours on June 5, all the miners and diesel equipment were out of the mine, yet

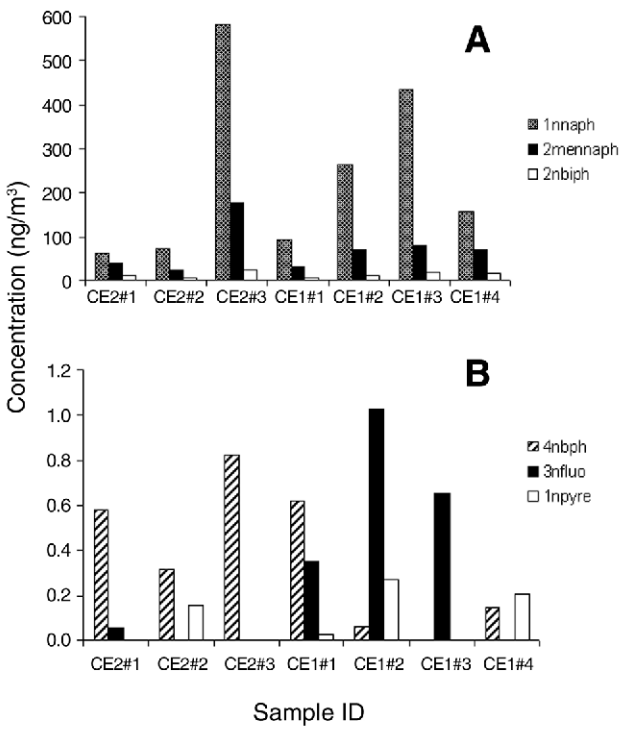


**Figure 15. Distribution of nitro-PAHs in ambient air in the mine.** ***A***. 1-Nitronaphthalene, 1-methyl-2-nitronaphthalene, and 2-nitrobiphenyl. ***B***: 4-Nitrobiphenyl, 3-nitrofluoranthene, and 1-nitropyrene.

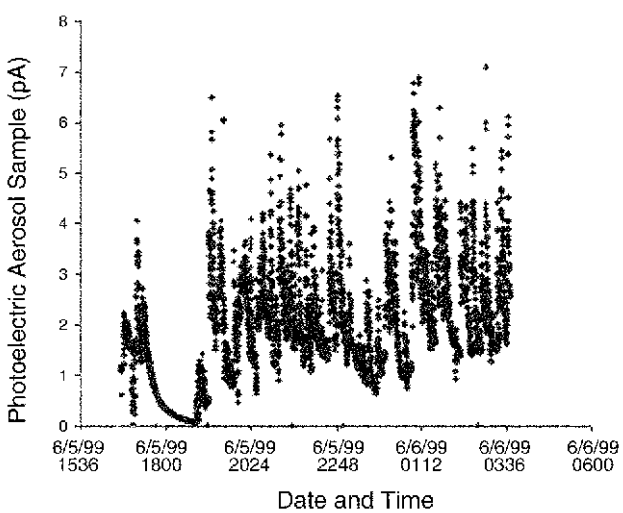


**Figure 16. Photoelectric aerosol sampler signal at CE1 (upper site) during overlap of day and night shifts from June 4 to June 5, 1999.**

the ventilation system was still on. This can be seen in Figure 16 as the decay of the signal with time, very nearly following an exponential decay as the pollutants producing the signal slowly diluted to near 0. As the diesel equipment reentered the mine, the signal quickly rose again. The fine structure of the graph most likely resulted from the signal rising due to the proximity of diesel vehicles driving by the sampling site. This promising result suggests that further research should be undertaken to determine if the PAS signal is truly correlated with diesel exhaust and if this instrument could be used to monitor underground environments.

## PERSONAL SAMPLES

### Inorganic Composition

Personal samples were collected during each shift by 6 workers per shift. In a few cases samples were invalid because of problems with samplers, including leaks. (The entire dataset is available in Appendix C.) As for the ambient data, the personal data were compared with the RCM. Figure 17 shows the scatter plot of the gravimetric mass versus the RCM, and the slope is nearly identical to that for the ambient data. This suggests that whatever phenomenon was affecting the ambient samples was also affecting the personal samples. One data point was eliminated from Figure 17 because the validity of the gravimetric mass result was suspect.

The concentrations of RCM, EC, and TC were determined (Figure 18). The mass concentrations in the personal samples were on average much higher than those in the ambient samples. The single highest value was over 2.7 mg/m$^3$ for a jammer during the night shift. The jammer was responsible for packing the backfill into mined longhole stopes and therefore was driving back and forth in a relatively small area (typically a dead end) with much less ventilation than in other areas of the mine. On the other shifts, the jammers' samples typically had the highest concentrations of RCM, EC, and TC.

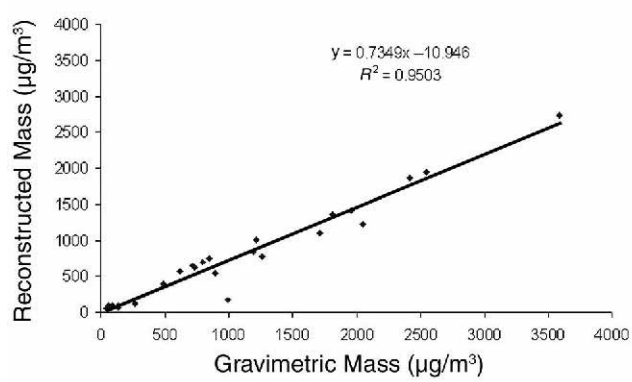


**Figure 17. Gravimetric mass versus reconstructed mass for personal samples.**

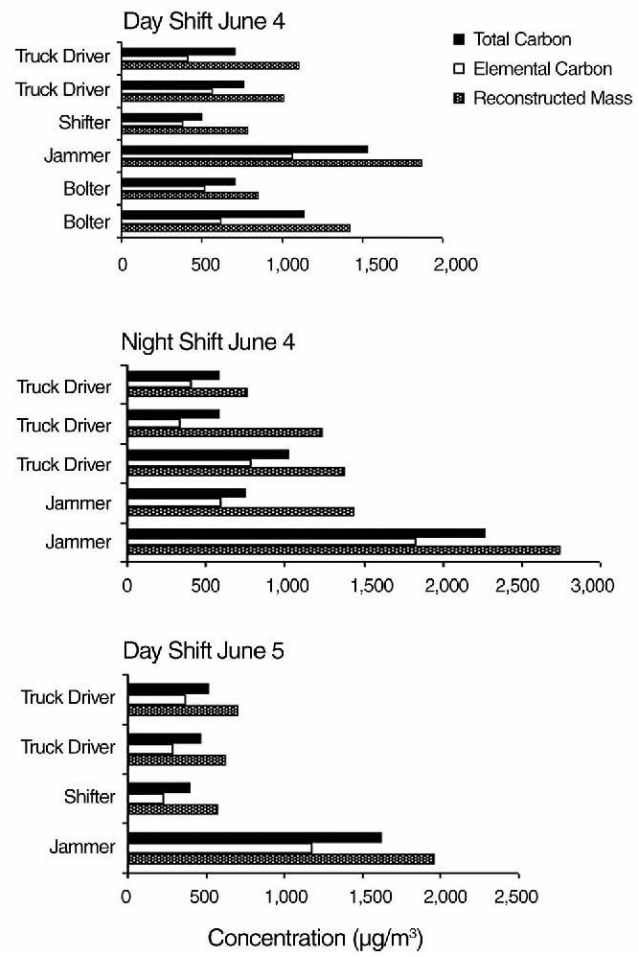


**Figure 18. Concentrations in personal samples for 3 shifts.**

The TC represented on average about 68% of the total mass in the personal samples. Elemental carbon was on average 70% of TC and was reasonably constant through the samples. Similar to the ambient samples, sulfate in the personal samples represented only about 5.5% of the total mass. The geologic elements were a relatively small fraction; only 14% of the RCM was silicon, on average.

### Organic Composition

The concentrations of gas-phase, semivolatile, and particle-phase PAHs were determined for personal samples collected in the mine. In Figure 19, the data obtained from the samplers carried by individual miners were combined into 4 groups according to the job description: truck drivers (7 samples), jammers (4 samples), bolters (3 samples), and shifters (2 samples). Four office workers served as a control group. Although the differences between individual samples were large, the highest concentrations of the most volatile PAHs were observed for jammers and truck drivers, followed by bolters. The heavier PAHs (Figure 19, C–E) were more abundant in the samples collected by bolters. This is consistent with the observation that in comparison with the other types of oil used in the mine, the rock drill oil was rich in heavier PAHs. Bolters, who operated diesel vehicles that drilled and reinforced surfaces in the mine, may have been exposed to mist from the rock drill oil more than the other workers. Jammers, who drove heavy-duty diesel vehicles that packed processed ore back into cavities in the mine, were probably more exposed to diesel fumes than the truck drivers.

As shown in Figure 19 (D and E), higher molecular weight PAHs, especially benzo[*a*]pyrene, 7-methylbenz[*a*]anthracene, benzo[*b*+*j*+*k*]fluoranthene, indeno[123-*cd*]pyrene, and benzo[*b*]chrysene, were found in the samples from the control group, although in much lower concentrations. There are two possible explanations for this observation. First, the higher concentrations for these compounds were observed mostly for samples office 1 and office 2. Although there was no smoking in the offices, smoking was permitted in the building where these offices are located. Thus, cigarette smoke might have penetrated these 2 rooms.

Second, we approached the limit of detection for these PAHs, and higher variability was observed. The measured concentrations of heavy PAHs were in the low range (ng/m$^3$), and the total average volume for personal samples was only approximately 1.4 m$^3$. The instrumental limit of detection for the Varian Saturn ion trap mass spectrometer was approximately 10 pg/µL for heavy PAHs. Since the final volume after extraction of personal samples was 100 µL, if the concentration of given PAHs was 10 ng/m$^3$, then we were measuring approximately 14 pg/µL, very close to the instrumental detection limit. In reality, the minimum detection limits for these PAHs are higher than 10 pg/m$^3$. For these low concentrations, the variability of the media blanks, and not the instrumental limit of detection, determines the minimum detection limit. The uncertainties of these measurements were high, sometimes close to (or higher than) the measured values.

The concentrations of hopanes and steranes in personal samples were averaged according to the job descriptions of the miners (Figure 20). Only a subset of the samples was analyzed for these compounds, including those of 1 office worker, 1 shifter, 2 truck drivers, 3 jammers, and 3 bolters. Bolters' samples were expected to show higher concentrations of hopanes and steranes because of their exposure to drill oil. But the sample collected by the shifter showed the higher concentrations for these compounds, followed by the bolters' samples. Because only 1 shifter's sample

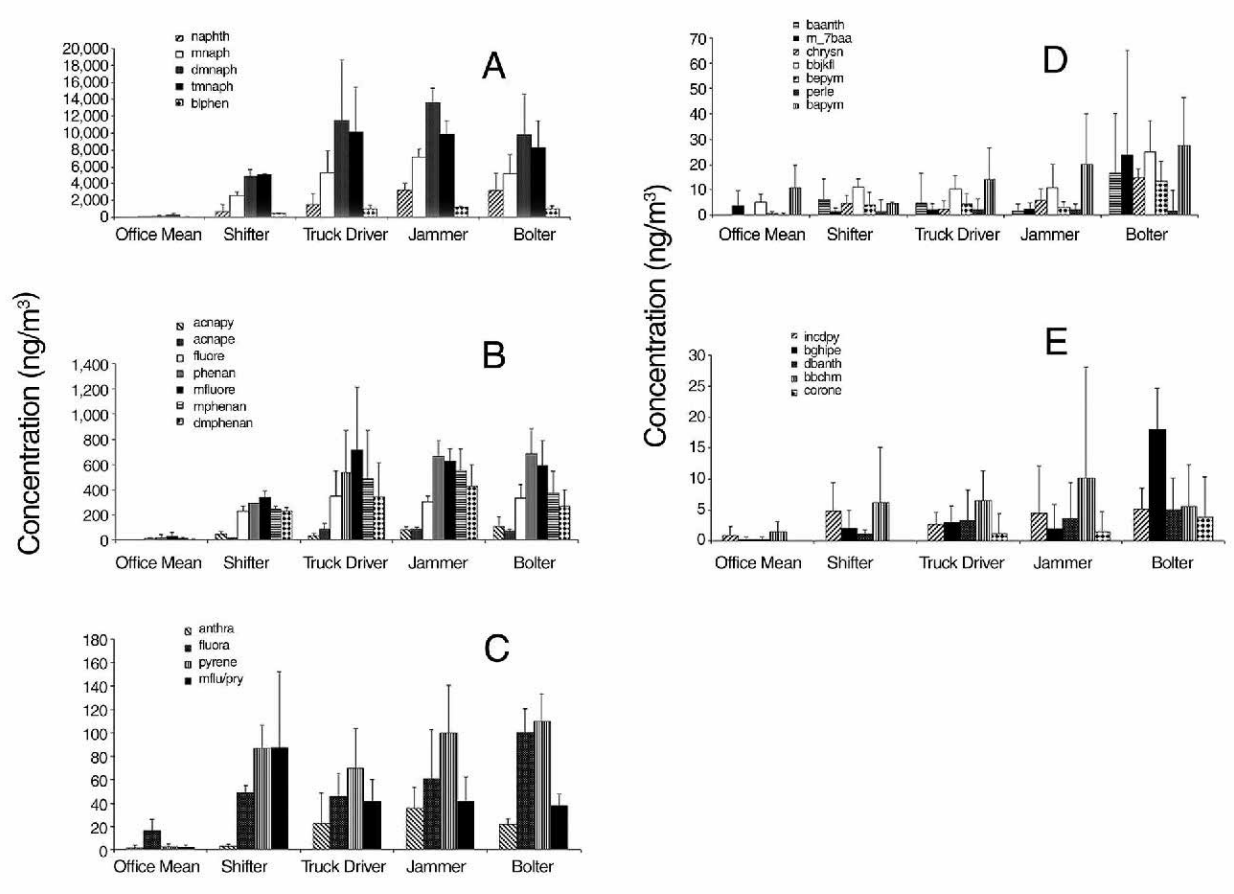


**Figure 19. Concentrations of PAHs in personal samples from the mines.** ***A.*** Gas-phase PAHs: naphthalene, methylnaphthalenes, dimethylnaphthalenes, trimethylnaphthalenes, and biphenyl. ***B.*** Semivolatile PAHs: acenaphthylene, acenaphthene, fluorene, phenanthrene, methylfluorene, methylphenanthrene, and dimethylphenanthrene. ***C.*** Semivolatile PAHs: anthracene, fluoranthene, pyrene, and methylfluoranthenes/pyrenes. ***D.*** Particle-phase PAHs: benz[*a*]anthracene, 7-methylbenz[*a*]anthracene, chrysene, benzo[*b*+*j*+*k*]fluoranthene, benzo[*e*]pyrene, perylene, and benzo[*a*]pyrene. ***E.*** Particle-phase PAHs: Indeno[*cd*]pyrene, benzo[*ghi*]perylene, dibenzo[*a*]anthracene, benzo[*b*]chrysene, and coronene.

was analyzed, however, it is difficult to generalize; the higher shifter value could reflect an artifact. Media and field blanks showed relatively high values for this class of compounds, which makes these measurements more uncertain (uncertainties are available in Appendix C).

## INTERCOMPARISON RESULTS

Samplers were measured side-by-side during one sampling period (the night shift on June 5 to 6, 1999) to compare results from the stationary samplers at the upper site (CE1#4) and 2 personal samplers (P1 and P2) equipped with commercial 2.5-µm-cutoff inlets. In addition, the personal sampler P3, equipped with the experimental 1-µm inlet, was used. Only data from P1 and P2 were used for comparison with CE1#4, however, because of the different cutoff point used for P3. Table 8 shows results of the comparison for an abbreviated list of compounds including the most abundant species measured and some species that were close to the ADLs. Percentage differences are presented in Table 8 for comparisons between P1 and P2, CE1#4 and P1, and CE1#4 and P2. For the more abundant chemical species, the agreement between both personal and stationary samplers ranged between 1% and 30%. Agreement was poor for chemical species present in amounts near the ADLs. This is illustrated by the last 4 organic compounds listed in Table 8, which were all present in very low amounts.

Watson and associates (2002) have described the propagation of error associated with measurement of atmospheric contaminants. In air quality studies, error has been associated with both sampling and analytic techniques. In this study, the colocated personal and ambient samples

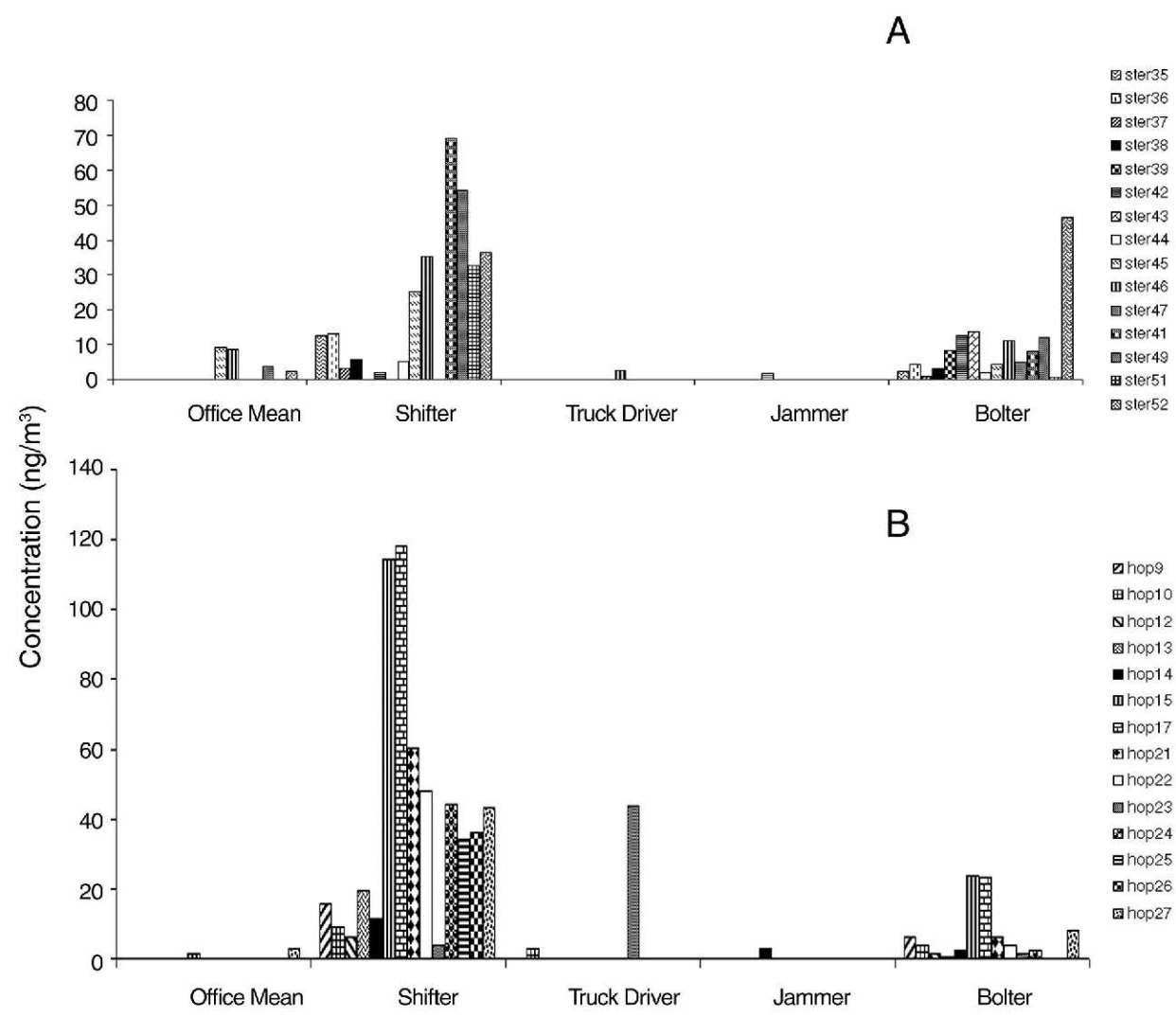


**Figure 20. Concentrations of particle-phase steranes (*A*) and hopanes (*B*) in the personal samples in the mine.**

differed in flow conditions and in the amount of material collected. The area and personal samplers were operated at much different face velocities. Face velocity is affected by the flow rate, the filter diameter, and the amount of material that accumulates on the filters during sampling. Changes in face velocity due to material collected during sampling in concentrated areas such as the mine increase uncertainty in volume measurements. During this study, flow was measured before and after sampling. In general, flow rates dropped during the sampling period, and an average of the 2 flow measurements was used. Using this method, however, it was not clear whether the flow rate dropped linearly throughout the sampling period or was steady for some duration and then dropped. The use of a flow-controlling device that would ensure constant flow during sampling should improve the accuracy of flow measurements.

Analytic accuracy is affected by both the measurement technique and the amount of material that is present for analysis. The total air volume collected for the stationary speciated organic sampler averaged 62 $m^3$, as compared with approximately 1.4 $m^3$ for the personal samplers. The larger volume of air collected with the stationary sampler provided more material and thus increased analytic accuracy. For example, if the given PAH concentration is 10 $ng/m^3$, then we are measuring 620 pg/µL for the ambient sample (since the final volume of ambient and source sample extracts was 1 mL), as opposed to 14 pg/µL for personal samples (final extract volume of 100 µL).

**Table 8.** Comparison of Results Obtained from Side-by-Side Samples Collected in Mine with Stationary Samplers (CE1#4), Two Personal Samplers (P1, P2), and Sampler Equipped with Experimental 1-µm Inlet (P3)

| | | Bulk Composition (µg/m$^3$) | | | | % Difference | | |
|---|---|---|---|---|---|---|---|---|
| Species Measured | Filter Type | CE1#4 | P1 (2.5 µm) | P2 (2.5 µm) | P3 (1 µm) | P1, P2 | CE1#4, P1 | CE1#4, P2 |
| Mass | Teflon | 612.82 | 893.82 | 718.18 | 486.49 | 21.79 | −18.65 | − 7.92 |
| Sulfate[a] | Quartz | 44.16 | 23.99 | 21.86 | 16.84 | 9.32 | 29.59 | 33.79 |
| Organic Carbon | Quartz | 117.78 | 109.36 | 145.64 | 93.59 | 28.45 | 3.70 | −10.58 |
| Elemental Carbon | Quartz | 258.69 | 272.50 | 355.60 | 250.20 | 26.46 | − 2.60 | −15.78 |
| Total Carbon | | 376.47 | 381.86 | 501.25 | 343.79 | 27.04 | − 0.71 | −14.22 |
| Sum of species | | 470.75 | 477.94 | 579.94 | 368.89 | 19.28 | − 0.76 | −10.39 |
| **Select elements**[b] | | | | | | | | |
| Magnesium | Teflon | 1.48 | 2.98 | 1.57 | 0.67 | 62.05 | − 33.78 | − 3.08 |
| Aluminum | Teflon | 6.17 | 9.24 | 7.64 | 0.83 | 18.90 | −19.91 | −10.66 |
| Silicon | Teflon | 21.01 | 30.09 | 24.60 | 3.81 | 20.08 | −17.78 | − 7.88 |
| Sulfate | Teflon | 20.60 | 25.21 | 27.09 | 21.71 | 7.19 | −10.05 | −13.59 |
| Potassium | Teflon | 4.25 | 6.16 | 4.79 | 0.39 | 24.96 | −18.28 | − 5.93 |
| Calcium | Teflon | 12.15 | 17.74 | 13.31 | 1.27 | 28.53 | −18.70 | − 4.56 |
| Iron | Teflon | 2.60 | 4.99 | 2.60 | 0.50 | 62.86 | − 31.55 | − 0.13 |
| **Select organics** | | | | | | | | |
| 2-Methylnaphthalene | TIGF[c] | 876.46 | 993.51 | 996.27 | 1,090.07 | 0.28 | − 6.26 | − 6.40 |
| Biphenyl | TIGF | 414.37 | 517.53 | 527.95 | 513.25 | 1.99 | −11.07 | −12.05 |
| Dimethylnaphthalenes | TIGF | 6,271.66 | 4,706.49 | 4,371.43 | 4,837.75 | 7.38 | 14.26 | 17.85 |
| Trimethylnaphthalenes | TIGF | 3,252.74 | 5,075.32 | 5,225.47 | 4,788.74 | 2.92 | − 21.88 | − 23.27 |
| Methylfluorenes | TIGF | 410.69 | 278.57 | 375.15 | 266.89 | 29.55 | 19.17 | 4.52 |
| Methylphenanthrene | TIGF | 194.61 | 180.52 | 189.44 | 130.46 | 4.82 | 3.76 | 1.35 |
| Dimethylphenanthrenes | TIGF | 111.03 | 79.87 | 81.99 | 71.52 | 2.62 | 16.32 | 15.04 |
| Anthracene | TIGF | 14.21 | 5.84 | 6.21 | 1.99 | 6.09 | 41.71 | 39.16 |
| Fluoranthene | TIGF | 7.74 | 12.34 | 26.09 | 7.95 | 71.56 | − 22.93 | − 54.26 |
| Pyrene | TIGF | 12.93 | 18.83 | 24.22 | 10.60 | 25.05 | −18.58 | − 30.40 |
| Benz[*a*]anthracene | TIGF | 1.04 | 0 | 0 | 0.00 | 0.00 | 100.00 | 100.00 |
| Chrysene | TIGF | 0.8 | 0 | 0 | 0.00 | 0.00 | 100.00 | 100.00 |
| Benzo[*e*]pyrene | TIGF | 0.8 | 0 | 1.24 | 0.00 | 200.00 | 100.00 | − 21.71 |
| Benzo[*a*]pyrene | TIGF | 0.75 | 1.95 | 13.67 | 19.21 | 150.09 | − 44.35 | − 89.58 |

[a] Measured by ion chromatography.

[b] Measured by x-ray fluorescence.

[c] Teflon-impregnated glass-fiber filter followed by PUF/XAD-4/PUF cartridges.

Because of their low concentrations, organic compounds that were present in amounts close to the ADL in the personal samples (10 ng/m$^3$) contained high uncertainty.

Interestingly, the P3 sampler (with 1-µm inlet) showed much lower concentrations of mass and crustal elements such as silicon, aluminum, calcium, magnesium, and iron, than the P1, P2, and CE1#4 samplers with 2.5-µm inlets. Although some differences were observed for OC and EC, they were not pronounced. The difference between P3 and CE1#4 samplers was 22% for OC and 3% for EC concentrations. The 22% difference may indicate that some small portion of the 2.5-µm OC could be due to volatile inorganic carbon in the $CO_3$ form, originating from crustal material. However, the EC was probably due mostly to small particles, less than 1 µm. Thus, use of a 2.5-µm or 1-µm inlet does not make an important difference in the TC measurements (below 10%), but it does for the mass measurements.

## SOURCE APPORTIONMENT

### Model Output and Performance Measures

Pace and Watson (1987) defined several performance measures, which we examined with each CMB. Appendix A shows a typical CMB output report and its associated performance measures. The output contains all information needed to reproduce the SCEs. The upper part of the display shows the source profiles included in each model application. The lower part of the display shows the species that were measured at the receptor and the species included in the CMB calculation. Table 9 shows the results of the CMB apportionment for all samples. Each CMB result includes values for the performance measures that were used to evaluate the goodness of the solution, following the regulatory guidance of Pace and Watson (1987). For these source apportionments, $r^2$ typically exceeded 0.9 and $\chi^2$ values typically ranged from 0.1 to 0.3. Percentage of mass values were between 91% and 105% for all apportionments.

***Deviations from Model Assumptions*** Assumptions 1 and 2 of the CMB model (see Chemical Mass Balance Formulation) specify that compositions of source emissions are constant over the period of ambient and source sampling. As mentioned previously, there was poor agreement between the calculated and measured amounts of OC and EC during this study. The major source of OC and EC in the mine was diesel vehicle emissions. The source tests showed that diesel vehicle emissions were typically composed of 50% to 75% OC and 20% to 40% EC. Alternatively, the ambient samples were composed of 20% to 25% OC and 50% to 70% EC. This poor agreement between the OC and EC compositions of source and ambient samples resulted in poor CMB results for these species. Because the ore contained small amounts of carbon, and diesel exhaust was assumed to be the major source of carbon in the mine, there apparently was a difference in the behavior of OC in the ambient air of the mine compared with OC in the source emissions. However, our test procedure for operating diesel vehicles to collect emissions may not be representative of the modes of operation for these vehicles in the mine.

As described in Source Sampling, we used a test cycle consisting of 5 minutes of idle (4 times), followed by a 2-minute load (3 times) to near 100% engine rating. Thus, for a 26-minute total test cycle, the engine was idling for 77% and was loaded for only 23% of the time. In the underground mine, diesel equipment was subjected to heavy load most of the time, and therefore our test cycle probably did not represent driving conditions inside the mine. These differences may have altered the OC/EC ratio in the vehicle's exhaust. Diesel engines working under no-load or light-load

**Table 9.** Contributions to Reconstructed Fine Particle ($PM_{2.5}$) Mass in an Underground Gold Mine from Chemical Mass Balance Source Apportionment

| Sample ID[a] | Concentration ($\mu g/m^3$)[b] | $r^2$ | $\chi^2$ | % of $PM_{2.5}$ Mass | Ore ($\mu g/m^3$) | Diesel ($\mu g/m^3$) | Sulfate ($\mu g/m^3$) | % of Mass[c] | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | | Ore | Diesel | Sulfate |
| CE1#1 | 657.9 ± 39.1 | 0.962 | 0.105 | 91.830 | 30.8 ± 16.6 | 506.1 ± 63.1 | 67.2 ± 24.5 | 4.68 | 76.93 | 10.21 |
| CE1#2 | 550.9 ± 32.9 | 0.964 | 0.176 | 98.144 | 38.0 ± 9.4 | 446.6 ± 60.9 | 56.1 ± 21.6 | 6.90 | 81.07 | 10.18 |
| CE1#3 | 554.5 ± 32.6 | 0.964 | 0.168 | 104.616 | 74.9 ± 14.7 | 478.0 ± 66.6 | 27.1 ± 23.2 | 13.51 | 86.20 | 4.89 |
| CE1#4 | 525.3 ± 30.7 | 0.943 | 0.302 | 101.597 | 56.5 ± 9.5 | 446.1 ± 58.3 | 31.1 ± 21.6 | 10.76 | 84.92 | 5.92 |
| CE2#1 | 378.9 ± 25.5 | 0.980 | 0.078 | 105.693 | 20.5 ± 7.7 | 371.6 ± 47.9 | 8.4 ± 17.6 | 5.41 | 98.07 | 2.22 |
| CE2#2 | 880.0 ± 55.1 | 0.969 | 0.137 | 97.351 | 98.5 ± 21.3 | 697.3 ± 100.4 | 60.9 ± 33.8 | 11.19 | 79.24 | 6.92 |
| CE2#3 | 1044.5 ± 81.6 | 0.948 | 0.133 | 95.992 | 10.2 ± 21.0 | 1,025.5 ± 129.6 | 23.9 ± 47.7 | 0.98 | 98.18 | 2.29 |

[a] CE1 samples were collected downwind from mining operations to represent aged emissions in the mine. CE2 samples represent samples collected near mining operations. CE2#1 was collected at a different location than CE2#2 and CE2#3.

[b] The $PM_{2.5}$ mass reported here is the reconstructed mass that was used in the source profiles.

[c] Values presented are the percentage of the reconstructed mass.

conditions emit more OC than the same engines working under high-load conditions (Shi et al 2000).

For the reasons described previously, TC (combined OC and EC) was used as a fitting species rather than OC and EC separately. Omitting the use of separated OC and EC did not change the results of the apportionment because the elements and speciated organics provided the information necessary to distinguish diesel exhaust from other sources in the mine. The large number of individual measured species made the apportionment robust enough to omit individual species or classes of some materials without affecting the overall results.

With respect to assumption 3, that all source types are identified and characterized, the percentage of mass performance measures suggest that all of the significant contributors were included in the CMBs. The mass was underestimated by as much as 20% in some of the initial SCEs. In 2 of these 3 samples, the difference between observed and calculated mass may be attributed to an underprediction of sulfate. The sulfate in diesel vehicle emissions was highly variable, ranging from 0.9% to 21% of the fine particulate mass emissions. The composite sample utilized in the final source apportionment had an average sulfate composition of 5.4% ± 7.4%. The initial vehicle apportionment did not completely account for sulfate, which most likely originates from diesel exhaust, because of the uncertainty associated with measurement of this species during this study. The off-road diesel fuel used in these vehicles contained 1,300 ppm of sulfur, considerably more than the current and proposed on-road standards for fuel sulfur content.

The large uncertainty in the average sulfate composition (5.4% ± 7.4%) limited the effect of vehicular sulfate in the source apportionment. For this reason, a sulfate profile was created to apportion any sulfate that was present in the mine and not apportioned from the diesel vehicle profile. The sulfate profile assumed that sulfate was present in the mine in the acidic form. The profile was created by defining the atomic contribution of each element (sulfur, oxygen, and hydrogen) to the mass of sulfuric acid and by setting uncertainties at 10%.

With respect to assumption 4 that the number of species exceed the number of sources, at least 65 species and up to 3 source profiles were used in each calculation. The number of chemical species always exceeded the number of source types. Assumption 5, that the source profiles are linearly independent, was largely eliminated by the inclusion of speciated organic species in the extended datasets. The effects of deviations from assumption 6, that measurement errors are random and normally distributed, remain to be studied. For this study, all of the CMB assumptions were met to the extent that the SCEs can be considered valid.

***Identification and Correction of Model Input Errors*** Any level 3 validation deficiencies in the processing, formatting, compositing, and reporting of ambient concentration and source profile measurements were identified and corrected or flagged as a result of CMB source apportionment. Corrections and flags were incorporated into the database, and the results presented reflect these changes. Some chemical species concentrations were physically unreasonable, as indicated by large $\chi^2$ values with a large R/U value for the related species. As discussed previously, sulfate, OC, and EC consistently showed large R/U values. In these cases the suspect species was removed from the fit. In general, the CMB modeling was robust enough that, when performance measures were within acceptable ranges around target values, suspect concentrations had little effect on the SCEs.

### Consistency and Stability of Source Contributions

The SCEs and the statistical and diagnostic information were reviewed to determine the validity of initial model results. The analysis was repeated by eliminating the oil source profiles that gave negative SCEs, or standard errors that exceeded the SCEs, and by including a sulfate profile to apportion vehicular sulfate emissions that were not accounted for. The good agreement between the calculated source contributions and the measured ambient concentrations indicates that all major source types were included in the calculations, that ambient and source profile measurements were reasonably accurate, and that the source profiles were reasonably representative of actual emissions.

There were 2 sample collection sites in the mine, CE1 and CE2. The CE1 samples were collected downwind from the majority of the mining activities, and the CE2 samples were collected in close proximity to many of the mining activities. The contributions of each source to $PM_{2.5}$ were similar at the 2 sites (Table 9), with diesel emissions contributing 76% to 98% and suspended ore contributing 1% to 13% of the mass. The majority of the remaining mass was sulfate, which most likely arose from vehicle emissions.

Prior to this study the potential sources of fine PM in the mine had been identified as diesel mining vehicles, suspended ore, and suspended oils. The suspended oils were not important contributors to ambient PM in the mine and were excluded from the apportionment; however, their contributions were also evaluated when the composition of the mine air was investigated. To apportion the differences

**Table 10.** Ratio of Sum of Gas-Phase and Particle-Phase Polycyclic Aromatic Hydrocarbons to Sum of Hopanes and Steranes Quantified from Mine Air, Diesel Exhaust, and Oil During This Study

| Chemical Class Ratio[a] | Ambient Air | Diesel Exhaust | Oil |
|---|---|---|---|
| Gas PAH/HOPSTER | 212 | 175 | 0.2–4 |
| Particle PAH/HOPSTER | 16 | 17 | 0.04–0.9 |

[a] PAH indicates polycyclic aromatic hydrocarbon; HOPSTER, hopanes and steranes.

**Table 11.** Pearson Correlation Coefficient for Ambient Air, Diesel Exhaust, and Suspended Oils Based on Their Organic Chemical Contents

| Variable | Ambient Air | Diesel Exhaust | Oils |
|---|---|---|---|
| Ambient air | 1 | 0.9864 | 0.096 |
| Diesel exhaust | 0.9864 | 1 | 0.085 |
| Oils | 0.096 | 0.085 | 1 |

between vehicle emissions and the oils, PAHs and hopanes and steranes were measured. These classes of compounds were observed in both diesel emissions and the oils measured during this study. The proportions of these chemical classes differed between the oils and the diesel emissions. Table 10 shows the ratio of gas-phase PAHs to hopanes plus steranes and the ratio of particle-phase PAHs to hopanes plus steranes in the mine air, compared with the ratios in diesel exhaust and suspended oils. These ratios, along with the correlation matrix shown in Table 11, suggest that the bulk of the organic material observed in the ambient samples during this study arose from diesel exhaust. The ratios were based on a separation of gas-phase PAHs and particle-phase PAHs, operationally defined as the compounds that eluted on a chromatographic column before and after phenanthrene, respectively. The ratios were similar for the mine air and diesel exhaust, but very different for the oils. Table 11 shows the Pearson correlation coefficients for diesel exhaust, oils, and mine air based on their organic chemical compositions. The mine ambient air and diesel exhaust were highly correlated in this study. Of course, these measurements were based on ambient samples collected in this study mine, and this relationship may vary with the study mine, mining activity, and placement of samplers. During the period when these samples were collected, no jackleg drills, which are a major source of drilling oil mists in mine air, were operating in the mine.

Another potential source of fine PM in the mine was cigarette smoking. The isoalkanes and antiisoalkanes are long-chain hydrocarbon compounds that arise from burning tobacco (Rogge et al 1991). These compounds can be used to help distinguish the contribution of tobacco smoke from other sources of particulate air pollution and were investigated in the mine. The isoalkanes and antiisoalkanes were not observed at any concentration in the mine above the ADLs. If present, cigarette smoke was a minor contributor to the ambient PM in this mine. Recent studies showed that indoor particulate material resulting from heavy smoking in an occupational setting yields concentrations of approximately 30 to 40 $\mu g/m^3$ in an unventilated bar (Jenkins and Counts 1999). This concentration represented an enclosed environment that had poor ventilation. The mine air was continually being ventilated, and the volume of air in the mine provided for much more dilution. The total ventilation flow through the mine was approximately 8,500 $m^3$/min (although in some drifts it may have been much lower). To calculate the potential of cigarette smoke to impact mine air, we assumed that in a worst-case scenario each crew member (12 per shift) would smoke a pack of cigarettes (20 cigarettes) over a 10-hour shift. If the emission rate is approximately 20 mg per cigarette (Hildemann et al 1991), then at the ventilation conditions in the mine, the concentration of cigarette smoke would be 0.43 $\mu g/m^3$. At this concentration, the speciated emissions from cigarette smoke would be below the ADL, as observed by our measurements. For these reasons, cigarette smoke was not considered as a fitting source in the source apportionment. Similar to the suspended oils, however, cigarettes may be important for personal exposure to either smokers or persons who come into contact with secondhand smoke in the mine.

## EXPOSURE ASSESSMENT

Fine particles ($PM_{2.5}$) have been implicated as an important component of lung injury. This is likely due to several factors including their relatively large surface area (which causes airway inflammation), the chemical nature of the particulates (such as organic matter [eg, PAH]), and the oxidant generation catalyzed by transition metals associated with the particles. In an underground hard rock mine, PM will occur as fine ($PM_{2.5}$) and coarse ($PM_{10-2.5}$) particles that differ in formation mechanisms, chemical composition, sources, and exposure relationships. Fine PM derives primarily from combustion material that has volatilized and then condensed to form primary PM or from precursor gases reacting in the atmosphere to form

secondary PM. Coarse PM is formed by crushing, grinding, and abrasion of surfaces, which breaks large pieces of material into smaller pieces. These particles are then suspended by the mining activities and by substantial movement of ventilation air through the mine. Energy considerations limit the breakup of large particles and small particle aggregates, generally to a minimum size of about 1 µm in diameter. So, fine PM may contain not only the ultrafine material, but also some of the smaller coarse particles between about 1 and 2.5 µm.

In the monitored mine, the RCM of $PM_{2.5}$ fell in the range of 400 to 2,700 µg/m$^3$, depending on the location and job function (eg, bolter, jammer, shifter, or truck driver). The bolter was exposed to levels of $PM_{2.5}$ in the range of 800 to 1,700 µg/m$^3$; the jammer, 1,400 to 2,700 µg/m$^3$; the shifter, 500 to 600 µg/m$^3$; and the truck driver, 600 to 1,000 µg/m$^3$. The EPA-promulgated (nonoccupational) standards for exposure to $PM_{2.5}$ are 65 µg/m$^3$ for 24-hour average exposure and 15 µg/m$^3$ for annual average exposure (OEHHA, personal communication, 2000). There is no shorter-term (ie, 1 hour) standard. Typical workshifts in the mine were about 10 hours in duration. Obviously, exposure to $PM_{2.5}$ in the mine was at least an order of magnitude greater than the EPA standard. The MSHA standard for metal and nonmetal mines is 400 µg/m$^3$ of TC.

Table 12 lists the metals and sulfate content of the $PM_{2.5}$ recovered from mine air. The studies discussed later in this report have shown that soluble metals, especially metal sulfates, can cause airway inflammation leading to lung damage. Although the exact mechanism of action leading to airway inflammation is not known, it is suspected that the transition metals catalyze a 1-electron reduction of molecular oxygen leading to generation of reactive oxygen species, which are the agents suspected to damage the lungs. Inhibition or reduction of PM-induced lung injury by metal-chelating agents has been used to infer the involvement of metals, and inhibition of damage by hydroxyl radical scavengers has indicated the involvement of free radicals (Gilmour et al 1996; Donaldson et al 1997; Li et al 1997; Goldsmith et al 1998).

The relatively high sulfate content of the PM may imply that some sulfuric acid aerosol was present in the mine air. There have been extensive studies of the effects of controlled exposures to aqueous acid aerosols on human lung function (Folinsbee et al 1997). Even at levels as high as 1,000 to 2,000 µg/m$^3$, however, aqueous acid aerosols had minimal effect on lung function (Culp et al 1995; Frampton et al 1995; Zelikoff et al 1997). The weight of evidence indicates that, especially at concentrations below 200 µg/m$^3$, acid aerosols typically do not tend to change airway responsiveness.

Thus, the focus with regard to exposure of mine workers to PM remains on the metals content of the inhaled particles. Interestingly, the majority of the research referenced in this report was concerned not with the PAH content of PM, but rather with the metal content of PM. Of the metals listed in Table 12, iron was in greatest abundance, followed by zinc, titanium, copper, vanadium, and nickel. This is in line with the fact that iron is the most abundant of the metals that are capable of catalyzing oxidant generation. In tests with human subjects, instillation of iron (III) oxide via the intrapulmonary route led to an inflammatory response in the alveolar fraction of lavage fluid, with an accompanying increase in neutrophils in the bronchial fraction (Lay et al 1998). Furthermore, of the 3 metals commonly found in fly ash—iron, vanadium, and nickel—iron has been associated with mild lung lesions and early cytokine gene expression, while nickel has been associated, in some cases, with more severe lesions such as alveolar edema and hemorrhage (Kodavanti et al 1997).

**Table 12.** Concentration Ranges of Metals and Sulfate in Personal Samples of Mine Air

| | Range of Concentrations in Air (µg/m$^3$) | | | | |
|---|---|---|---|---|---|
| Constituent | Bolter | Jammer | Shifter | Truck Driver | Background[a] |
| Copper | 0.06–0.24 | 0.03–0.36 | 0.12–0.14 | 0.02–0.13 | 0.01–0.06 |
| Zinc | 1.1–1.8 | 1.6–3.8 | 0.77–0.80 | 0.59–1.4 | 0.02–0.08 |
| Vanadium | 0–0.02 | 0–0.37 | 0.01–0.14 | 0–0.34 | 0–0.12 |
| Iron | 2.5–4.9 | 3.0–7.4 | 7.0–8.9 | 3.0–18 | 0.62–0.97 |
| Nickel | 0.02–0.05 | 0.01–0.06 | 0.03–0.04 | 0–0.07 | 0.02–0.03 |
| Titanium | 0.14–0.33 | 0–0.66 | 0.04–0.70 | 0.08–2.0 | 0–0.21 |
| Sulfate | 37–60 | 20–195 | 2.2–19 | 9.0–86 | 0 |

[a] Office workers.

The relevance of the studies by Lay et al (1998) and others to exposure of mine workers to high levels of PM is not clear. The amount of iron oxide (approximately 5 mg) delivered acutely to the human subjects by instillation was orders of magnitude greater than the amount of iron that might be inhaled during a typical workshift in the mine (worst case [truck driver]: approximately 250 µg of iron over 10 hours, derived from a respiration rate of about 1.44 $m^3$/hr). If the inhaled particles are not cleared efficiently from the airway, however, then buildup over several days may approach the level in the instillation study. Even so, studies based on intratracheal instillation are problematic in terms of extrapolating the exposure data to inhalation in humans.

This study, and others, exposed the test subjects to artificially high levels of PM in order to induce measurable responses in a reasonable period of time. Because of this, and the use of laboratory animals in particular, toxicologic data from high-exposure studies have limited use due to difficulties in quantitative extrapolation to humans under typical exposure scenarios. Nonetheless, results from these studies indicate what might happen to humans exposed at lower particulate levels over more prolonged periods of time (eg, in the mine: 10 hours/day, 5 days/week, for 20+ years). Even at PM levels more comparable to those observed in the mine (110 to 733 µg/$m^3$), damage to the lungs of the test animals was measurable within 1 to 3 days during inhalation studies (Gordon et al 1998; Clarke et al 1999). Furthermore, epidemiologic studies of exposure to ambient levels of $PM_{2.5}$ indicated a statistically significant decrease in human longevity, which was attributed primarily to exposure to metals in the PM. Finally, exposure to PM can adversely impact the immune and cardiovascular systems.

Taken together, toxicologic and epidemiologic results from the studies discussed in this report strongly suggest that hard rock mine workers are an at-risk group with regard to compromised health of the airways, lungs, cardiovascular system, and immune system, all of which can reduce longevity.

## DISCUSSION

### OBJECTIVES AND RESULTS

The objectives of this exploratory study were to characterize chemical composition and size distribution of PM in an underground gold mine; apportion the contribution of specific sources to suspended fine PM in the mine; and estimate the exposure of miners to the measured chemical species.

The analyses of fine PM samples collected in ambient air in this mine showed that the concentrations of fine particulate TC exceeded the MSHA interim standard of 400 µg/$m^3$ during 2 shifts at the lower site situated next to active mining area (CE2) and for 1 shift at the upper site (CE1) near the mine exit. TC accounted on average for 72% of the reconstructed $PM_{2.5}$ mass, and EC averaged about 75% of TC. Importantly, the OC/EC measurements were obtained by the TOR method for this study, which usually provides somewhat higher EC values than NIOSH method 5040, which uses thermo-optical transmittance (Chow et al 2001). In some extreme cases, measurement of EC by NIOSH method 5040 may result in values less than half of the values measured by the TOR method. However, the TC values measured by these 2 methods are in good agreement (Chow et al 2001).

The analyses of personal samples showed that the miners were exposed to much higher concentrations of fine particulate TC than those indicated by area samples. Workers in some job categories, such as jammers or bolters, were exposed to TC concentrations exceeding 1 mg/$m^3$. On average, TC represented about 68% of the RCM in the personal samples, and EC accounted for 70% of TC. The gravimetrically determined mass was greater than the sum of the analyzed chemical species. Since the chemical species that were analyzed for this study should have accounted for a majority of the mass of suspended fine PM in the mine, this underestimation of mass was most likely due to water that adsorbed to the filters and was weighed but not analyzed. This same phenomenon has been observed and described in some previous studies (eg, van Loy et al 2000).

The results in this study are comparable to those in an underground exposure study of 5 coal mines by Cantrell and associates (1993). In this study they observed an average mass of fine PM of 0.89 mg/$m^3$ (SD, 0.44 mg/$m^3$) for samples collected with personal sampling devices equipped with impactors with an 0.8-µm cutoff point. Our study utilized a 2.5-µm cutoff point, which would result in collection of a greater fraction of ambient PM relative to the study of Cantrell and associates. For this study, the average concentration of RCM for personal samples was about 1.1 mg/$m^3$ (SD, 0.58 mg/$m^3$). The single highest value was over 2.5 mg/$m^3$ for a jammer (Figure 18).

The mine samples were analyzed for PAHs, hopanes, steranes, and some nitro-PAHs. The gas-phase and semivolatile PAHs, such as phenanthrene, methylfluorenes, methylphenanthrenes and dimethylphenanthrenes, were the most abundant in the mine air, with the particle-phase PAHs present in much lower concentrations. This PAH distribution was similar to the composition of diesel exhaust, the main source of PAH in the mine. The PAH distribution for personal samples generally followed the same pattern, but PAHs were found in much higher concentrations depending on the particular job performed by the miner. Volatile and

semivolatile PAHs were generally more abundant for truckers and jammers, whereas heavy particle-phase PAHs were relatively more abundant for bolters. This is consistent with the observations that (1) the rock drill oil was enriched in heavier PAH than other types of oil used in the mine; and (2) diesel equipment emissions were enriched in volatile and semivolatile PAHs, in comparison with heavy, particle-associated PAHs. Bolters, who operated diesel vehicles that drilled and reinforced surfaces in the mine, may have been more exposed to the drilling oil mist than the other group of workers. Thus, the fine PM concentrations and chemical compositions may differ between samples collected by area samplers and those collected by personal samplers.

Although data on the PAH composition in mine atmospheres are sparse, Cantrell and associates (1993) published the concentrations of 4 PAHs, fluoranthene, benz[*a*]anthracene, chrysene, and benzo[*a*]pyrene, in ambient samples collected in 3 coal mines. Concentrations reported for these PAHs were generally higher than those in the current study. Similarly, the concentrations of fluoranthene, pyrene, and benzo[*a*]pyrene reported for 2 other coal mines (Bagley et al 1991) were higher than those measured during this study. To our knowledge, there are no published data on PAH concentrations from personal samples collected in mines.

Size-resolved gravimetric and chemical data on ambient PM were obtained from MOUDI samples collected at the upper sampling site in the mine. The stages at 3.16, 1.78, 1.00, 0.54, 0.37, 0.148, 0.105, and 0.054 µm were used, with the afterfilter collecting particles smaller than 0.054 µm. The distribution showed that a considerable amount of mass was in the larger size fraction (3.16 µm) and that this fraction was made up of material other than carbon (since the carbon was only 2% to 10% of the mass in this size). Carbon dominated the accumulation mode (0.148 to 0.105 µm). The chemical data show that the geologic materials were most prevalent in the largest fractions and nearly nonexistent in the smaller sizes. This is consistent with previously published data (Cantrell 1987). The ratio of EC to TC changed throughout the size distribution. The EC accounted for 20% to 35% of TC in the 3.16-µm fraction and for up to 80% in all fractions less than 0.148 µm. These size data are consistent with data obtained by Marple and colleagues (1986) in underground metal and nonmetal mines: diesel carbon was found primarily in the submicron size fraction, and the larger size fractions were made up of resuspended ore and waste rock.

An interesting feature of the size distribution data is the difference in distribution of mass observed in the ambient mine samples versus the vehicle source samples for particles with aerodynamic diameter lower than 0.054 µm. This fraction was collected on the afterfilter. In the source samples a considerable fraction of the mass and carbon was on the afterfilter, while very little material was on the afterfilter in the ambient samples. The difference may have been due to the environment into which the emissions were being released. In the source testing, emissions were released into clean, filtered, and nearly particle-free air during sampling. In the mine, the emissions were released into a mixture of fresh outdoor air, other diesel emissions, and dusts from the rock and other material. The additional particles in the mine likely provided conditions suitable for coagulation and growth of the ultrafine particles into the accumulation mode or larger. In addition, the residence time of the material in the source samples was less than 2 minutes, while the residence time of material in the mine prior to sampling was approximately 10 minutes. These differences in condensation nuclei and residence time would explain differences in the observed size distribution between the source and ambient samples.

The CMB was used to estimate the contributions from different sources to carbon and $PM_{2.5}$ concentrations in the mine's air. Prior to this study the potential sources of fine PM in the mine had been identified as diesel mining equipment, suspended ore or waste rock, and suspended oil mists. The CMB analyses showed that diesel emissions were the predominant source of $PM_{2.5}$ in the mine, accounting for 76% to 98% of the RCM. The suspended ore makes up 1% to 13% of the mass, with sulfate accounting for the balance left from ore and diesel emissions. In the ambient samples, sulfate accounted for as much as 10% of the mass. This sulfate likely originated from vehicle emissions. All of the sulfate was not accounted for by the vehicle apportionment: some of the vehicles studied emitted high amounts of sulfate (21% of their fine particle emissions) and were not selectively represented in the source apportionment (a composite of multiple vehicles was used). Possibly these high sulfate emitters contributed the bulk of sulfate emissions to the mine air.

The results of CMB showed that the suspended oils were not an important contributor to ambient PM in the mine, and they were excluded from the final apportionment. The use of the oils in the source apportionment had less than 5% effect on the apportionment of diesel vehicles and ore. The composition of organic material observed in the mine air, compared with the vehicle emissions and oils, also suggests that the contribution of suspended oils in this study mine was negligible. There was a strong correlation ($r^2$ = 0.98) between the composition of diesel exhaust and the ambient air, and a very weak correlation between oils and mine air ($r^2$ = 0.08). This relationship may vary with the study mine, mining activity, and placement of sampling

equipment. During the period when these samples were collected, no jackleg drills were operating in the mine.

Another potential source of fine PM in the mine was cigarette smoking. The isoalkanes and antiisoalkanes proposed as tracers for tobacco smoke (Rogge et al 1991) were measured in this mine. However, they were not observed at any concentration in the mine above the ADLs. If present, cigarette smoke would be a minor contributor to the ambient PM in the mine. Similar to the suspended oil, however, cigarettes may be important for personal exposure to either smokers or persons that come into contact with secondhand smoke in the mine.

Despite the fact that the major source of OC and EC in the mine was diesel vehicle emissions, there was poor agreement between the calculated amounts and the measured amounts of OC and EC in this study. The source tests showed that the diesel vehicle emissions were typically composed of 50% to 75% OC and 20% to 40% EC. In contrast, the ambient samples were composed of 20% to 25% OC and 50% to 70% EC. This poor agreement resulted in poor CMB results for OC and EC.

Since the ore contained small amounts of carbon, and diesel emissions were the major source of fine carbon in the ambient air in the mine, there was an apparent difference in the OC/EC ratio in the ambient samples and that in the source emissions. The procedure used to test diesel vehicle emissions during this study may not have been representative of the mode of operation of these vehicles in the mine and may have resulted in significant differences in the OC/EC ratio in the vehicles' exhaust. The poor agreement with the source and ambient composition of OC and EC also resulted in poor CMB results for these species. Therefore, OC and EC ultimately were not included as fitting species during this study. TC was used as an input species along with speciated organic and elemental species. The CMB sensitivity tests showed that omitting OC and EC did not affect the results of the apportionment. When they were included, performance of the model decreased, but overall results were not significantly changed.

## IMPLICATIONS OF FINDINGS AND RECOMMENDATIONS

The study reported here was designed as a feasibility study and as such was limited in terms of duration (1 year only), scope (1 mine, 3 consecutive shifts), and available resources. The study did not intend to answer the numerous questions concerning level of exposure of the broad population of miners to diesel emissions, or to provide quantitative, subject-specific risk assessment, or to address the issue of sampling methods. Rather, this study assessed the ability of source sampling and CMB modeling to apportion the sources contributing to ambient area samples taken in an underground mine. The conclusion is that the CMB successfully apportioned the material in the mine between diesel exhaust and suspended ore particles. Other possible sources of PM (such as drill oil mist and cigarette smoke) were not detected in the CMB analysis because their contributions to the samples in this study were too low. This does not preclude the possibility that under other environmental conditions—ie, different mines, different sampling locations within the mine, different mining equipment, or longer study period—the final conclusions would be the same. The technologies and methods used in this study have been successfully used in numerous studies of ambient air pollution but had never been tested in the underground environment. Despite the simplicity of the source-receptor relationship in the underground environment, the resolving power of many chemical species used in the CMB was necessary to successfully apportion the sources.

Source apportionment by CMB showed that an average of 86% of the airborne fine particle mass ($PM_{2.5}$) in this mine was composed of diesel exhaust, with ore averaging about 7.5%. The balance of the apportioned airborne material was vehicle-related sulfate that was not accounted for by the CMB vehicle profile. Because the sulfate emission rates from individual vehicles varied from 0.4% to 21% of the fine particle emissions, it was not feasible to account for the entire sulfate load in the mine with an average CMB vehicle profile.

An important result of this study was that CMB apportionment results and mine air composition tests did not reveal significant amounts of suspended oil mist in the ambient mine samples. This may be different in different mines or during periods of mine operation when different types of equipment are in operation.

The other implication from this work is that underground miners in this study mine were exposed to diesel PM at levels above the current proposed interim TC level of 400 $\mu g/m^3$, and certainly above the proposed January 2006 TC limit of 160 $\mu g/m^3$, measured by both area samplers and personal samplers. Personal samples and area samples taken in the mine showed differences in composition and concentration depending on the job function of the person carrying the sampler. For the first day shift, June 4, 1999, the average personal sample showed nearly 75% more mass than the highest area sample. For the night shift starting on June 4, 1999, the average personal sample mass was 67% greater than the highest area sample mass, and the highest personal sample was 3 times the highest area sample. This shift had the single highest concentration from a personal sample in the study. For the last shift monitored,

the day shift starting June 5, 1999, the mean personal sample was slightly less than the highest area sample, but all personal samples were higher than the lower area sample. Although the side-by-side comparison of area and personal samplers showed variability of up to 30% for mass, OC, EC, and TC (see Intercomparison Results), the differences observed for the actual personal and area samples were much higher than 30%. These data clearly show that the area samples taken in this study did not accurately represent the exposure of the miners who carried the personal samplers. The personal samples collected during this study were analyzed for a broad range of organic and inorganic species, and they provide the first dataset relating miners' exposure to speciated PAHs and metals.

Several recommendations emerge from this work:

- The duty cycle used for collecting emission samples from mine equipment must be reconciled with the actual mode of operation of this equipment in the mines. This is particularly important if EC is used as a single tracer for diesel emissions. The differences between the test procedure and the mode of operation of diesel vehicles in the mine may significantly alter the OC/EC ratio in the vehicle's exhaust, resulting in erroneous apportionment of diesel emissions by the CMB model. In this study, TC was used as an input species along with speciated organic compounds and inorganic species. The CMB sensitivity tests showed that, in this case, omitting the OC/EC ratio did not affect the results of the apportionment.
- In order to investigate the feasibility of apportioning oil mists from diesel carbon, future work should utilize more ambient monitoring stations including those adjacent to activities that produce large amounts of oil mist. In addition, to evaluate cigarette smoking as a potential source of fine PM in the mine, some monitoring stations should be located in the areas where miners gather for breaks.
- To create a profile for drilling oil mist in the mine, the oil source samples should be collected as mists directly from the equipment that emits them. This would provide a more accurate assessment of the composition of drilling oil as it exists in the mine air.
- Further research should be conducted specifically toward resolving the issue of gravimetric mass versus reconstructed mass in the underground environment.
- In order to establish proper locations for area samples that will accurately represent miner exposure, a more detailed comparison of area samples versus personal samples should be undertaken. The proposed MSHA rules call for area sampling, but the results of this study indicate that personal samples are often higher than area samples. Also, if mine operators were to install filtered-air cabs on underground diesel equipment, the area samples would be much less indicative of exposure.

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## APPENDICES AVAILABLE ON REQUEST

The following appendices are available by contacting the Health Effects Institute. Please give the full title of report and the titles of the appendices.

Appendix A. Example of a Typical CMB Output During the $PM_{2.5}$ Source Apportionment of the Carlin East Mine

Appendix B. Source Profiles

Appendix C. Ambient and Personal Sampler Data

Appendix D. Information Sheet for Participants

Appendix E. Description of Study Site

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**Barbara Zielinska** is a research professor and director of the Organic Analytical Laboratory at the Desert Research Institute, Reno, Nevada. She received her PhD in chemistry from the Polish Academy of Sciences. Her primary areas of interest include collection and analysis of trace atmospheric organic species present in both gas and particle phases; development of analytical methods for measuring biologically active compounds in primary and secondary particulate organic matter; atmospheric transformations of organic compounds; and particle-associated and volatile organic compound emissions from various sources including diesel- and gasoline-powered vehicles, wood combustion, and meat cooking.

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## ABBREVIATIONS AND OTHER TERMS

| | |
|---|---|
| ACS | American Chemical Society |
| ADL | analytic detection limit |
| CE1 | Carlin East upper mine |
| CE2 | Carlin East lower mine |
| $CH_4$ | methane |
| $CH_2Cl_2$ | dichloromethane |
| $Cl^-$ | chloride |

| | |
|---|---|
| CMB | chemical mass balance |
| $CO_3$ | terrestrial carbon |
| DDW | deionized distilled water |
| DRI | Desert Research Institute |
| EC | elemental carbon |
| ERA | Environmental Research Associates |
| FP/SVOC | fine particle/semivolatile organic compound |
| GC/MS | gas chromatography/mass spectrometry |
| HEPA | high-efficiency particulate air (filter) |
| IARC | International Agency for Research on Cancer |
| MOUDI | micro-orifice uniform deposit impactor |
| MSHA | US Mine Safety and Health Administration |
| NaCl | sodium chloride |
| $Na_2CO_3$ | sodium carbonate |
| $NaHCO_3$ | sodium bicarbonate |
| $NaNO_3$ | sodium nitrate |
| $Na_2SO_4$ | sodium sulfate |
| NIOSH | US National Institute for Occupational Safety and Health |
| NIST | US National Institute of Standards and Technology |
| $NO_3^-$ | nitrate |
| OC | organic carbon |
| OEHHA | US Office of Environmental Health Hazard Assessment |
| PAH | polycyclic aromatic hydrocarbon |
| PAS | photoelectric aerosol sampler |
| PM | particulate matter |
| $PM_{10}$ | PM < 10 µm in aerodynamic diameter |
| $PM_{2.5}$ | PM < 2.5 µm in aerodynamic diameter |
| PUF | polyurethane foam |
| C/M | ratio of calculated to measured species |
| R/U | ratio of residual to its uncertainty (standard error) |
| $r^2$ | bivariate coefficient of determination |
| RCM | reconstructed mass |
| SCE | source contribution estimate |
| $SiO_2$ | silicon dioxide |
| SiOH | silicon hydroxide |
| $SO_4^{2-}$ | sulfate |
| SPE | solid-phase extraction |
| SRM | standard reference material |
| TC | total carbon |
| TIGF | Teflon-impregnated glass-fiber filter |
| TOR | thermo-optical reflectance |
| $\chi^2$ | chi-square (test) |
| XAD-4 | polystyrene-divinylbenzene resins |
| XRF | x-ray fluorescence |

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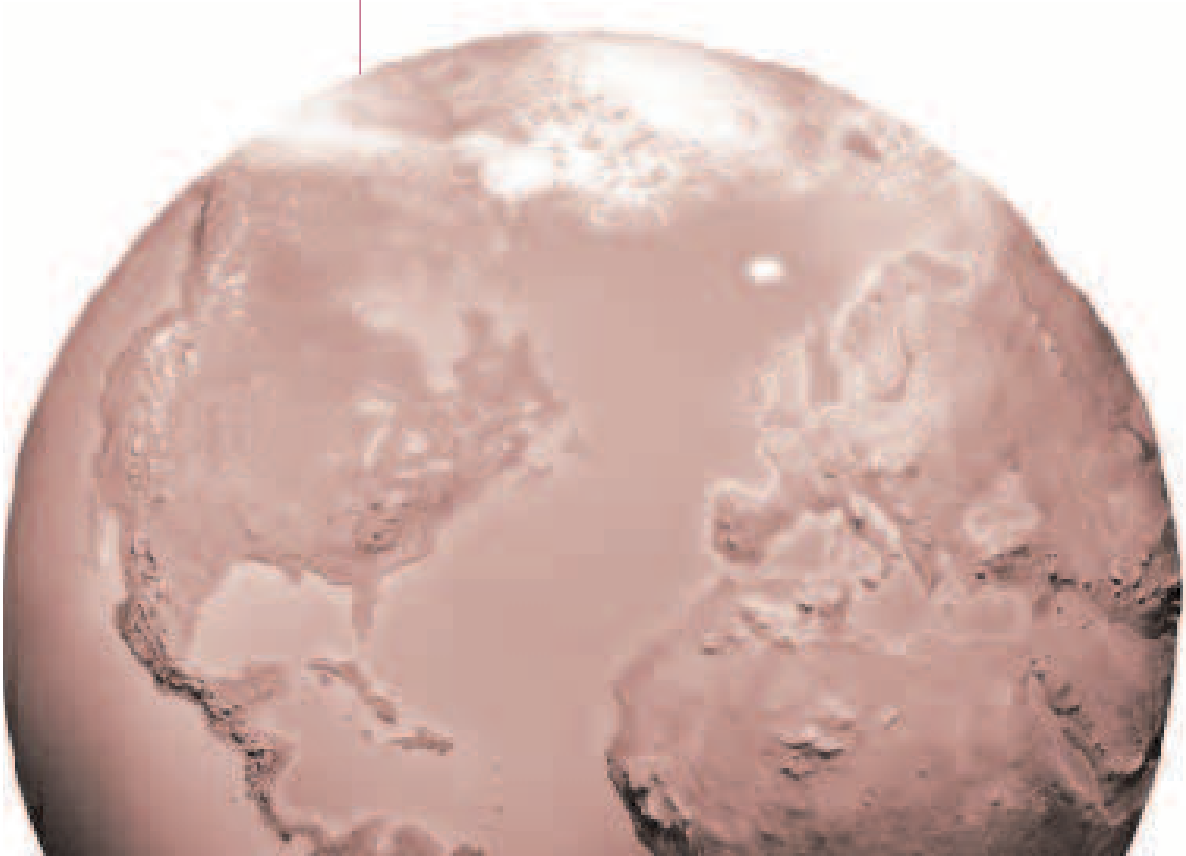
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