Chapter 10

POLLUTANT LEVELS IN UNDERGROUND COAL MINES USING DIESEL EQUIPMENT

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Permissible exposure limits (PELs) have been established for gaseous pollutants, carbon monoxide (CO), carbon dioxide (CO_2), nitric oxide (NO), nitrogen dioxide (NO_2), and some gas-phase hydrocarbons emitted in diesel exhaust. There is, as yet, no PEL recommended for diesel exhaust aerosol (DEA), nor is there a standard method for sampling this aerosol.

The University of Minnesota and the U.S. Bureau of Mines have collaborated to develop a personal diesel exhaust aerosol sampler (PDEAS) which utilizes size-selective inertial impaction and gravimetric analysis. During the field tests of this sampler, numerous air quality measurements were made in underground coal mines that use diesel equipment.

The mine mean DEA concentrations for the five mines surveyed, determined with the PDEAS in the haulageway, was 0.89 mg/m^3, with a standard deviation of 0.44 mg/m^3. DEA contributed 52 % of the respirable aerosol at this location.

In three of the mines filter samples were collected for DEA-associated polynuclear aromatic hydrocarbons (PAHs) and biological activity determinations. Two of the mines were also monitored for the major gaseous constituents found in diesel exhaust. In general, the PAH and biological activity levels were similar for all three mines, and indicate that up to 25 % of the haulageway concentrations may be contributed by outby diesel vehicles. Measured concentrations of CO, CO_2, NO, NO_2, and SO_2, were well below regulated levels.

INTRODUCTION

Diesel exhaust contains pollutant gases, such as carbon monoxide, carbon dioxide, nitric oxide, nitrogen dioxide, and gas-phase hydrocarbons, as well as DEA. Much of the health-related concern focuses on DEA and associated organic compounds (Watts, 1992a). A wide variety of these PAHs have been identified and some are known carcinogens and/or mutagens. The U.S. Mine Safety and Health Administration (MSHA) has proposed new PELs for these and other contaminants (MSHA, 1989). MSHA has also published an advance notice of proposed rulemaking to establish a separate PEL for diesel particulate (MSHA, 1992).

The U.S. Bureau of Mines has collaborated with the University of Minnesota to develop and field test a PDEAS. The PDEAS is a three stage sampler based on the MSA[1] personal respirable dust sampler. It utilizes a respirable cyclone preclassifier followed by a 0.8 μm cut point impactor and afterfilter operating at a flow rate of 2 L/min. Respirable aerosol greater than 0.8 μm in size is collected by the impactor while DEA, less than 0.8 μm in size, is collected by the afterfilter. Hence, gravimetric analysis of the afterfilter permits measurement of DEA concentrations. This development and laboratory evaluation of the PDEAS were described previously by Cantrell (1990) and Rubow (1990). During field tests of the sampler, numerous air quality measurements were made in continuous miner sections of five underground coal mines that use diesel haulage equipment. These air quality measurements included levels of selected PAH and biological activity associated with DEA collected in the intake and haulageway areas of three of the five underground mines, and CO, CO_2, NO, and NO_2 in two of the mines. The objectives of this paper are to present the DEA and associated pollutant concentrations measured in these mines and to assess the impact of diesel face-haulage equipment on underground mine air quality.

MINE DESCRIPTIONS

The mines used for the PDEAS evaluation were designated J, K, L, N, and O. Mines K, N, and O are located in the Western United States, while mines J and L are located in the East. Each mine produces high volatile, bituminous coal with shift production levels varying from 500 to 2000 tons/section. Seam heights varied from 1.5 to 3.0 m. Mines K and N use continuous mining to develop longwall panels. The others are strictly room-and-pillar operations using continuous miners.

The number and types of diesel-powered vehicles used at these mines were described by Watts (1992b). Mines J, K, N, and O use diesel power to assist in a wide range of activities in addition to coal haulage. These included road maintenance, personnel and materials transport, lubrication, and welding. Mine L used only three diesel-powered shuttle cars to haul coal.

SAMPLING AND ANALYSIS METHODS

Aerosol Measurements

Aerosol samples were collected in the mine portal area, the clean air intake to the continuous miner section, the haulageway one crosscut inby from the feeder breaker and belt, in the return airway, and on selected personnel. The haulageway sampling site was located near the point where the diesel-powered shuttle cars turn around to dump their loads. Additional respirable and DEA samples were collected and have been reported by Haney (1990).

PDEAS Samples. Twelve samplers were deployed at five different locations in continuous miner sections utilizing diesel-powered shuttle cars. These locations were the intake entry, haulageway, return entry, each shuttle car, and, in a few instances, on one of the research personnel. Collected samples were analyzed gravimetrically in a humidity controlled weighing facility. Mine Research Establishment (MRE) equivalent respirable dust concentrations were determined for the samples as (1.38 x (mass deposited on the impactor plate) + filter mass) divided by the volume of sampled air (Treaftis, 1984). DEA concentrations were determined by dividing the filter mass by the volume of air sampled. No correction was made for submicrometer mineral dust aerosol or the background aerosol entering the section through the intake airway.

High-volume (HI-VOL) Samples. HI-VOL samplers operating at a flow rate of 1.13 m^3/min were used to collect submicrometer particles in three of the mines: K, L, and O. The samplers incorporated slotted impactors with cut sizes of 3.5, 2.0, and 0.95 μm to remove coal and rock dust. Particles less than 0.95 μm in size collected by the afterfilter were considered primarily diesel in origin (Cantrell, 1987). These samplers were placed in the same locations in the haulageway and section intake as the PDEAS. Haulageway sample times varied from 8 to 60 min. It should be noted that the haulageway samples were collected only when the haulage vehicles were present at the sampling sites. Hence, concentrations reported in this paper represent worst case levels attainable in the haulageways of the mines surveyed. The intake samplers were essentially operated continuously during a working shift, up to 6 h. Data from these samples represent potential background contributions from outby diesel vehicles to diesel emissions in the haulageway areas.

After sample collection, the impaction substrates and backup filters containing the less than 0.95 μm size particles were wrapped in aluminum foil, placed in paper folders, and stored at -20° C until shipment on dry ice to Michigan Technological University for analysis (Bagley, 1992). There, the filters were first analyzed gravimetrically after equilibration in a humidity controlled chamber. The filters were then restored at -20° C until they were extracted.

The soluble organic fraction (SOF) was removed by Soxhlet extraction (24 h) with dichloromethane. A small aliquot of the total extract (of known volume) was allowed to evaporate to constant mass on a pre-weighed glass-fiber filter disk (Bagley, 1991b). The mass of the SOF was calculated from the mass of extractables found on the disk. For PAH and biological activity determinations, the individual extracts from all haulageway or intake filters from the same day from each mine were combined to reflect mean levels over each day's entire sampling period.

PAH Measurement. PAH measurements were done using SOF extracts from mines K, L, and O. The PAHs chosen for quantification were fluoranthene, benz[a]anthracene, chrysene, and benzo[a]pyrene. These compounds were selected on the basis of their activity as mutagens or carcinogens or, in some cases, their reactivities as precursors to biologically active species. These PAHs were also known to be components of diesel exhaust and pure reference standards are commercially available for accurate quantification. Each combined SOF sample was first separated into several fractions using column chromatography. The fraction containing the PAH was then analyzed by high-performance liquid chromatography for individual components, followed by quantification using a fluorescence detector and comparison to known standards (Gratz, 1991) (McClure, 1992).

Biological Activity. This was evaluated by assaying the SOF using a modification of the microsuspension version of the *Salmonella typhimurium*/microsome mutagenicity bioassay, or Ames assay. Specific details on the conduct of this assay have been presented elsewhere by Bagley (1991b). Assay response is reported in terms of revertants, bacteria having the original mutation corrected by interaction with one or more mutagenic chemicals. If assay-detectable mutagenic organic compounds are present, there will be a dose-related increase in bacterial response. The dose-response data are used to calculate an activity value, revertants per microgram of sample; the higher the activity value, the more mutagenic or biologically active the test material is considered to be.

HI-VOL Data. All HI-VOL DEA, SOF, PAH, and biological activity data were converted to a volumetric concentration basis using the total volume flow (cubic meters) for each sample. A mean of daily means value was calculated from the DEA and SOF data from each mine and sampling location. A mean for each mine and sampling location was also determined for PAH concentrations and biological activity.

Gaseous Pollutants

To measure CO and CO_2, filtered mine air was pumped into a 22 L, 5-layer bag at approximately 50 cm^3/min using a Dupont P-125 pump. The CO and CO_2 concentrations were analyzed at the end of the sampling period using an Ecolyzer 2600 CO instrument and a Fuji ZFP5 CO_2 instrument. NO_2 and NO were sampled using Palmes (1976) passive samplers or diffusion tubes (Cantrell, 1992).

RESULTS

Diesel Exhaust Aerosols

Table 1 summarizes the MRE equivalent respirable and DEA results obtained from the PDEAS. Little respirable aerosol is expected to enter the mine environment from outside sources (Watts, 1992b). Each of the mines was located in a sparsely populated region where air pollution is not a problem. Typical sources of respirable aerosols outside the mines are: fugitive dusts generated by vehicular traffic, storage piles, and belt conveyors.

Concentrations of respirable aerosol and DEA at the section intake were dependent upon vehicular traffic in that area. The other sampling locations, i.e., haulageway, shuttle car, and scientist, were located near mining and diesel activity. The return sampling site

Table 1.— Summary Statistics for Respirable and DEA Samples Obtained Using the PDEAS

Location	Number of samples	Respirable aerosol concentration, mg/m^3				DEA concentration, mg/m^3			
		Mean	SD	Median	Range	Mean	SD	Median	Range
Intake	16	0.21	0.15	0.18	0.04 - 0.61	0.13	0.12	0.09	0.01 - 0.48
Personnel	15	1.42	0.75	1.23	0.57 - 3.71	0.38	0.13	0.35	0.24 - 0.70
Haulageway	42	1.72	1.38	1.21	0.59 - 6.15	0.89	0.44	0.75	0.31 - 2.23
Shuttle car	51	1.40	0.54	1.44	0.35 - 2.70	0.67	0.23	0.65	0.19 - 1.22
Return	100	4.63	3.97	2.38	0.91 -15.99	1.43	0.97	1.0	6.46 - 4.03

SD - Standard deviation.

was located at a point where a well-mixed aerosol sample could be obtained, and sufficiently far from the continuous miner to allow settling of large particles. At these locations, the mean concentrations of respirable aerosol and DEA were much higher than at the intake.

Figure 1 is a lognormal probability plot of the cumulative frequency distribution of respirable aerosol (A) and DEA (B) concentrations from PDEAS samples collected in mines J through O at haulageway, shuttle car, continuous miner, and return locations.

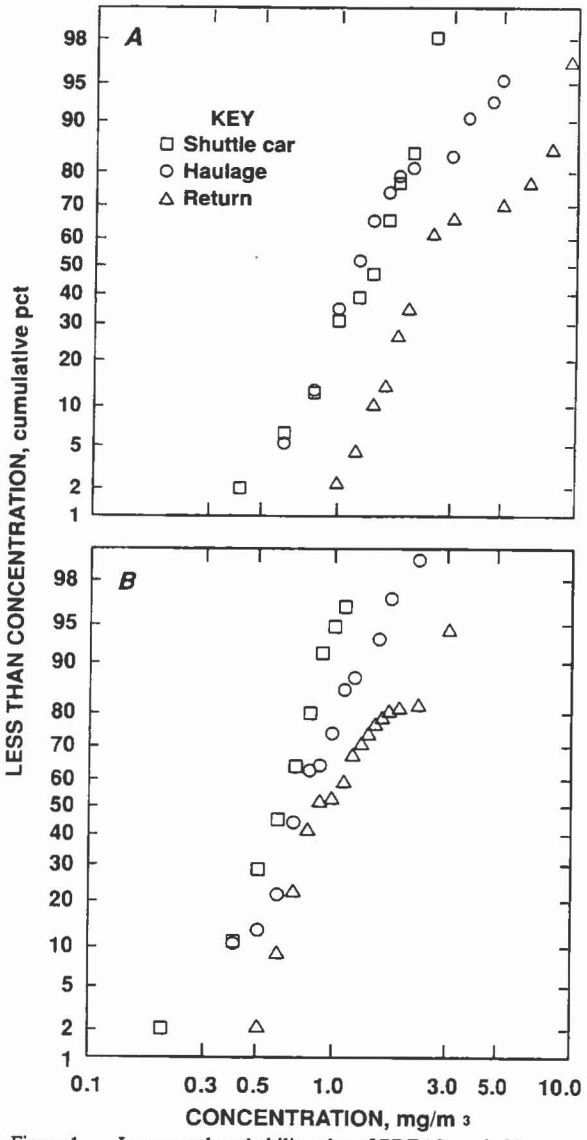

Figure 1. Lognormal probability plot of PDEAS respirable aerosol (A) and DEA (B) distributions.

The haulageway, shuttle car, and return locations have similar distributions for DEA. This suggests that exposure to diesel exhaust is the same throughout the section. Respirable aerosol concentrations are different depending on where the samples are taken: the highest is in the return entry and the lowest is in the haulageway. The difference between the DEA and respirable concentrations is due to the basic dispersion and settling behavior of DEA and respirable coal dust.

Diesel Aerosol Associated SOF. Levels (mg/m³) of DEA and SOF collected by HI-VOL samplers in the intake and haulageway areas of three underground coal mines are presented in Table 2. These data are mean values presented with coefficient of variation (CV) for samples collected over several days at each mine. Although the mean DEA values, from 0.90 to 1.4 mg/m³, represent potentially worst case haulageway levels, they are not excessively greater than mean near-full-shift DEA levels reported in Table 1 that were obtained in the same mines at the same sampling locations using personal samplers (Watts, 1992b). Several other studies have reported full-shift, personal sampler DEA levels of close to 1 mg/m³ (Watts, 1989) (Haney, 1990) (McCauley, 1986).

Slightly greater variation was found in mean haulageway SOF levels with values ranging from 0.08 to 0.19 mg/m³. Differences between mines can again be attributed to differences in vehicle operation, engine type, and fuel composition, as well as possible differences in vehicle maintenance.

The intake DEA and SOF values vary considerably both between and within mines. This would be expected since these samples were collected over a longer period than the haulageway samples, and the DEA collected probably originated from the random operation of diesel support vehicles outby the intake high-volume samplers. At the four mines, the intake air could have contributed from 5 to 27 % of the collected DEA in the haulageway and from 6 to 23 % of the SOF. As previously mentioned, the intake levels vary depending on the activity of the outby diesel vehicles. Therefore, the intake values were not subtracted from the haulageway values, and only the mean and range of intake values are presented.

PAH Levels. Table 3 presents measurements for four biologically important PAHs associated with underground DEA: fluoranthene, benz[a]anthracene, chrysene, and benzo[a]pyrene. As with the DEA and SOF data, the values are presented as mine means (with CV) for both the intake and haulageway sampling locations. The relative ratios of the four PAHs in the haulageways are typical of values found in laboratory-generated exhaust samples from engines similar to those used in these mines (McClure, 1992) (Bagley, 1991a).

Studies have shown that engines produce the highest levels of fluoranthene, for example, when the engine is under low load or is idling (Wall, 1984). In-mine observations indicated that there was some idling for haulage vehicles in all the mines except for mine L, which had the lowest levels of SOF, Table 2, and the lowest PAH levels, Table 3. In general, mine L's PAH levels are much lower than those at mines K and O, based on both mass, μg/g SOF or DEA, and concentration, μg/m³, data. Mine L also had almost three times the coal production of the other two mines, which may translate into heavier loads on the diesel engines, higher exhaust temperatures, minimal amount of engine idling, and proportionately lower amounts of SOF and some types of PAHs. Other factors that may affect PAH emissions are fuel and engine types. Different types of engines were used in these mines, however, the composition of the fuels used during the studies did not differ greatly.

With the exception of most values from mine L, these PAH levels are generally greater than those reported by Westaway (1987) for diesel-equipped nickel and salt mines near Sudbury, Ontario, Canada. However, the noncoal mine samples may have been collected over longer time periods, including periods when no diesel vehicles were operating near the sampling locations.

For the intake PAH levels, there is about the same amount of variation between and within mine-mean values as for the haulageway samples. The potential contributions of these PAHs in the intake air to the haulageway levels are generally within the same

Table 2. Mine Haulageway and Intake DEA, SOF, and Associated Biological Activity[a]

Mine	Site[b]	n[c]	Concentrations DEA (mg/m³)	Concentrations SOF (mg/m³)	% SOF	Revertants/m³	Biological Activity[d] DEA (Rev/μg)	Biological Activity[d] SOF (Rev/μg)
K	In	3	0.21	0.040	19	159[e]	0.762	3.85[e]
		(1-2)	(4.8)	(12)	(16)	(2.2)	(0.28)	(14)
	Ha	3	0.903	0.171	19	444	0.497	2.59
		(3)	(5.2)	(12)	(5.2)	(24)	(29)	(25)
L	In	5	0.07	0.005	5.5	NT[f]	NT	NT
		(1)	(42)	(66)	(73)			
	Ha	5	1.41	0.078	5.7	NT	NT	NT
		(3-4)	(17)	(24)	(13)			
O	In	4	0.39	0.022	5.8	117	0.344	5.83
		(1)	(27)	(32)	(38)	(52)	(51)	(64)
	Ha	4	1.44	0.185	13	691	0.477	3.63
		(2-6)	(14)	(20)	(22)	(44)	(48)	(27)

[a]As determined from HI-VOL samplers: presented as the mean of daily means (CV,%).
[b]Ha = Haulageway; In = Section Intake
[c]Number of sampling days (number of samples per date).
[d]Test strain TA98 without S9 metabolic activation.
[e]Data for only two days.
[f]NT - not tested due to very low levels of SOF.

Table 3. PAH Detected with In-Mine Samples[a]

Mine	Site[b]	n[c]	Parameter	FLU	BaA	CHR	BaP
K	In	3	ng/m³	36 (25)	13 (29)	14 (29)	9.6 (18)
			μg/g SOF	910 (32)	330 (21)	340 (20)	240 (21)
			μg/g DEA	180 (22)	68 (31)	70 (31)	49 (23)
	Ha	3	ng/m³	140 (16)	25 (16)	23 (11)	16 (6.7)
			μg/g SOF	780 (14)	150 (14)	130 (9.6)	94 (5.8)
			μg/g DEA	160 (10)	30 (14)	28 (8.4)	20 (2.3)
L	In	5	ng/m³	4.3 (83)[d]	0.26 (88)	0.38 (60)[d]	0.48 (68)
			μg/g SOF	610 (65)[d]	46 (60)	67 (26)[d]	150 (120)
			μg/g DEA	46 (76)[d]	2.2 (110)	4.4 (49)[d]	4.0 (54)
	Ha	5	ng/m³	48 (30)	1.6 (140)	<0.035[e,f,g]	<0.046[e,f,g]
			μg/g SOF	640 (27)	23 (140)	<0.48[e,f,g]	<0.64[e,f,g]
			μg/g DEA	42 (39)	1.4 (140)	<0.029[e,f,g]	<0.039[e,f,g]
O	In	4	ng/m³	6.9 (36)	5.9 (34)	5.7 (48)	5.4 (80)
			μg/g SOF	340 (45)	320 (60)	260 (33)	320 (93)
			μg/g DEA	19 (49)	17 (50)	15 (61)	16 (98)
	Ha	3	ng/m³	160 (16)	22 (26)	17 (50)	18 (22)
			μg/g SOF	1,110 (12)	150 (26)	120 (62)	120 (16)
			μg/g DEA	120 (11)	16 (12)	13 (54)	14 (22)

[a]Presented as mean (CV,%) for fluoranthene (FLU), benz[a]anthracene (BaA), chrysene (CHR), and benzo[a]pyrene (BaP).
[b]In = Section intake; Ha = Haulageway.
[c]Number of sampling dates.
[d]Four dates.
[e]Three dates.
[f]< indicates minimum detection level (MDL) for the analysis divided by two, Gratz (1991).
[g]All values less than this MDL value.

range as DEA and SOF contributions, about 5 to 28 %. This seems true in mine O for all four PAHs, in mine K for fluoranthene, and in mine L for fluoranthene and benz[a]anthracene. Much greater contributions for the remaining PAH's were found for mines K, 50 to 60 %, and L, over 100 %.

Biological Activity. Comparison of intake and haulageway mean biological activity values, with CV, for mines K and O are also presented in Table 2. No assays were performed on the samples from mine L, since the small amount of SOF obtained was used solely for PAH quantification.

As with the DEA data, the haulageway biological activity values are similar between the two mines, based on both mass (revertants/μg SOF or DEA) and concentration (revertants/m³) data. Comparison of these activity levels to those obtained in other in-mine studies is difficult because of differences in the specific type of Ames assay used. For comparison purposes, these mean haulageway activity levels are at least 5 to 10 times higher than mean ambient air samples that also were collected using HI-VOL samplers and assayed using similar techniques (Kado, 1986) (Agurell, 1990).

The biological activity levels associated with the intake air were more variable between mines than those measured in the haulageway, with up to threefold differences in levels. The intake activity levels were typically greater than those of the haulageway on a mass basis (revertants/μg SOF or DEA) but were lower on a concentration basis (revertants/m^3) due to the lower DEA levels at the intake. The contribution of DEA-associated activity in the intake air to the observed haulageway levels was within the range of DEA or SOF contributions for mine O, but slightly higher, about 35 %, for mine K.

Gaseous Pollutants

Measurements for CO, CO_2, NO, NO_2, and SO_2 were made at mines J and O at the intake, shuttle car, haulageway, and return locations. These results are summarized in Table 4. All reported concentrations are well below proposed regulated levels.

Table 4. Summary of Gaseous Pollutant Data

	CO	CO_2	NO	NO_2
MINE J				
INTAKE				
Mean, ppm	1.6	580	0.04	0.15
SD, ppm	1.8	290	0.08	0.09
CV, pct	110	50	207	60
SHUTTLE CAR				
Mean, ppm	3.1	1040	4.6	0.48
SD, ppm	0.6	38	1.4	0.11
CV, pct	20	37	30	23
HAULAGE				
Mean, ppm	2.6	910	2.8	0.39
SD, ppm	1.0	35	0.6	0.05
CV,pct	38	38	22	12
RETURN				
Mean, ppm	2.1	850	3.3	0.32
SD, ppm	0.9	24	0.4	0.03
CV, pct	40	28	11	9
MINE O				
INTAKE				
Mean, ppm	2.0	680	0.1	0.04
SD, ppm	1.4	420	0.0	0.03
CV, pct	71	62	32	73
SHUTTLE CAR				
Mean, ppm	4.0	990	3.4	0.28
SD, ppm	0.8	150	2.0	0.17
CV, pct	20	16	60	62
HAULAGE				
Mean, ppm	3.3	900	2.7	0.22
SD, ppm	0.5	120	1.4	0.19
CV, pct	15	13	53	87
RETURN				
Mean, ppm	3.0	930	3.9	0.30
SD, ppm	2.2	380	2.0	0.21
CV, pct	72	41	50	72

SD = Standard deviation.
CV = Coefficient of variation.

DISCUSSION

Other investigators have used size-selective sampling methods to quantify submicrometer aerosol concentrations in diesel-equipped coal mines. McCawley (1986) reported concentrations of < 1 μm aerosol from two underground coal mines using diesel face-haulage equipment. These measurements were made using the Anderson Model 298 cascade impactor. Mean submicrometer aerosol concentrations ranged from 0.1 mg/m^3 at the intake to 0.77 mg/m^3 at the continuous miner for the two mines. Mean NO and NO_2 concentrations were also reported for the two mines and these ranged from 0.11 ppm NO at the intake, to 4.38 ppm at the continuous miner; and 0.14 ppm NO_2 at the intake, to 0.53 ppm at the transfer point. The reported concentrations for NO and NO_2 are well below regulated levels.

Haney (1990) conducted tests of a single-jet impactor designed and built by MSHA for DEA measurement at five underground coal mines that use diesel equipment. Miner exposure to DEA ranged from 0.18 to 1.00 mg/m^3 and area samples collected in haulageways agreed within 0.12 mg/m^3 of section worker exposure. At these mines, the MSHA single-jet impactor and PDEAS were used together at a sampling location. More than 60 paired data points were obtained. Respirable aerosol measurements agreed within 25 %, and DEA measurements agreed within 0.06 mg/m^3 of each other.

The mine haulageway atmosphere levels of DEA, SOF, PAH, and biological activity were generally similar in the three mines for which these HI-VOL determinations were made, thus providing a range of values that might be expected to occur when diesel vehicles are operating without emission control devices. The observed differences in levels between the mines can be attributed to differences in parameters such as vehicle type, engine operation and maintenance, ventilation efficiencies, and fuel composition. The value of operating at higher loads with well-maintained vehicles is demonstrated by the consistently low levels of SOF and biologically active PAH at mine L. Potential contributions of intake air to haulageway DEA, SOF, PAH, and activity levels vary considerably, both between and within mines, with the maximum potential contribution to the observed haulageway values typically no more than 25 %.

SUMMARY

Five underground coal mines that use diesel haulage equipment were surveyed, using size-selective sampling by inertial impaction with gravimetric analysis, to determine the concentrations of DEA. The arithmetic mean DEA concentrations determined using PDEAS at the haulageway location for the five mines surveyed was 0.89 mg/m^3, with an average standard deviation of 0.44 mg/m^3. DEA contributed 52 % of the respirable aerosol at this location. It is clear from these data that DEA can contribute significantly to respirable coal mine dust aerosol concentrations in mines that use diesel haulage.

At two of the mines, measurements were made for the gaseous diesel pollutants CO, CO_2, NO, NO_2, and SO_2, and the concentrations were well below regulated levels. At three of these mines samples were collected for analysis of specific PAHs, SOF, and biological activity. In general, the PAH and biological activity levels were similar for all three mines; these data provide a range of values for the mines visited. The associated SOF, PAH and mutagenic activity measurements indicate that up to 25 % may be contributed by outby diesel vehicles.

ACKNOWLEDGMENTS

This research was partially supported by the U.S. Department of Interior's Mineral Institute program, administered by the Bureau through the Generic Minerals Center for Respirable Dust, and under NIOSH Grant No. R010H02611 to Michigan Technological University. The content of this paper is solely the responsibility of the authors and does not necessarily represent the official views of NIOSH.

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Proceedings of the 6th US MINE VENTILATION SYMPOSIUM

June 21-23, 1993
Salt Lake City, Utah

Ragula Bhaskar
Symposium Chairman and Proceedings Editor
Department of Mining Engineering
The University of Utah

Sponsored by
Underground Ventilation Committee of SME
joint committee of the Coal Division
and Mining & Exploration Division
and
The University of Utah

Published by
Society for Mining, Metallurgy, and Exploration, Inc. (SME)
Littleton, Colorado • 1993

Published Proceedings of the Previous Mine Ventilation Symposia

1st Symposium:	1982 — University of Alabama, University (Tuscaloosa), AL, published by the Society of Mining Engineers, P.O. Box 625002, Littleton, CO 80162-5002. LP $10.
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3rd Symposium:	1987 — The Pennsylvania State University, University Park, PA, published by the Society of Mining Engineers, P.O. Box 625002, Littleton, CO 80162-5002. Out-of-print.
4th Symposium:	1989 — University of California-Berkeley, Berkeley, CA, published by the Society for Mining, Metallurgy, and Exploration, Inc., P.O. Box 625002, Littleton, CO 80162-5002. LP $35.
5th Symposium:	1991 — West Virginia University, Morgantown, WV, published by the Society for Mining, Metallurgy, and Exploration, Inc., P.O. Box 625002, Littleton, CO 80162-5002. LP $68.50.

Printed in the United States of America by
Cushing-Malloy, Inc., Ann Arbor, MI

Library of Congress Catalog Card Number 93-83949
ISBN 0-87335-118-5