

MEASUREMENT OF DIESEL EXHAUST AEROSOL

by

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ABSTRACT

Extensive use of diesels in the mining industry has given rise to questions concerning the exposure of miners to respirable diesel-exhaust aerosol (RDEA). Quantifying this exposure depends on the ability to sample and measure the respirable diesel component of mine aerosol. In coal mines, such measurement is complicated by the carbonaceous nature of both the diesel and coal dust particulate that make up the respirable aerosol. Hence, special sampling and analytical techniques must be used to distinguish between these two aerosol components.

As part of a continuing study of diesel-exhaust aerosol characteristics, the Bureau of Mines has applied size-selective sampling together with gravimetric analysis to determine the sepa-

rate RDEA and coal dust aerosol concentrations in diesel equipped underground coal mines. This technique, using a cascade inertial impactor for size-selective sampling, is based on analysis of the mine aerosol size distribution and the premise that the RDEA portion of the aerosol is predominantly submicron and the mineral dust portion is mostly greater than one micron in size. In the study, chemical mass balance modeling, based on the distribution of chemical elements in aerosol and aerosol source material samples, is used to referee the analysis of diesel exhaust and mineral dust aerosol concentrations from the size-selective sampling results.

Based on the results of the study, the Bureau, in cooperation with the University of Minnesota under sponsorship by the Generic Mineral Technology Center for Respirable Dust (GMTC-RD)

and the Mine Safety and Health Administration (MSHA), are developing a prototype personal diesel-exhaust aerosol sampler for underground coal mines. The sampler has three stages, a pre-classifier stage to select for respirable aerosol, an inertial impaction stage with a size cut at $0.8 \mu\text{m}$ aerodynamic diameter to separate diesel aerosol from mineral dust and collect the mineral dust, and a final filtration stage to collect the diesel fraction of the sample. Sample fractions are gravimetrically analyzed to determine concentrations of the diesel and mineral dust portions of respirable coal mine aerosol. The sampler is currently being evaluated in both laboratory and field experiments conducted by the Bureau, MSHA, and the GMTC-RD.

INTRODUCTION

A miner working in an underground coal mine with diesel equipment is exposed to a wide array of pollutants from the diesel exhaust. These include CO , CO_2 , NO , NO_2 , SO_2 , diesel-exhaust aerosol, and a variety of hydrocarbon compounds. A quantitative definition of the health risk resulting from these exposures remains elusive, but during the past several years progress has been made in defining the problem.

Results from epidemiological studies¹, animal inhalation studies²⁻⁴, and in vitro studies⁵ have provided sufficient data for the National Institute of Occupational Safety and Health (NIOSH) to recommend that "whole diesel exhaust be regarded as a potential occupational carcinogen, as defined in the Cancer Policy of the Occupational Safety and Health Administration."⁶ NIOSH further stated that "though the excess risk of cancer in diesel-exhaust-exposed workers has not been quantitatively estimated, it is logical to assume that reduction in exposure to diesel exhaust in the work-place would reduce the excess risk."

The Mine Safety and Health Administration (MSHA) has been directed by an advisory committee to establish a respirable diesel-exhaust aerosol

(RDEA) standard and regulations to minimize exposure to diesel pollutants in underground coal mines.⁷ Both NIOSH and MSHA reports point out the need for development of measurement techniques for RDEA.

In an ongoing research program, the Bureau of Mines^{8,9} and the University of Minnesota's Particle Technology Laboratory¹⁰, operating under a grant from the Generic Minerals Technology Center for Respirable Dust (GMTC-RD)¹¹, have studied the use of size-selective sampling techniques to measure the mass concentration of diesel aerosol in underground coal mines. The study was based on the premise that size-selective sampling techniques can be used to separate diesel-exhaust aerosol, which is predominantly submicron in size, from coal dust aerosol, which is mostly greater than a micron in size. Results from the study have confirmed this premise and have been used to design a personal diesel-exhaust aerosol sampler for use in underground coal mines.¹² This paper reviews general mine aerosol characteristics, the data used in development of the personal sampler, its design, and its evaluation in both laboratory and field tests.¹³

MINE AEROSOL CHARACTERISTICS

Mine aerosols arise from a variety of sources and, as illustrated in fig. 1, the aerosol mass size distribution is influenced by these sources.¹⁴ Fig. 1 also displays the physical mechanisms such as condensation and coagulation that transfer aerosol mass from one size to another. There are three distinct size ranges that can be identified from features in measured size distributions. The smallest of these, 0.001 to $0.08 \mu\text{m}$, is the Aitken nuclei range which contains primary aerosol from combustion sources such as diesel engines and secondary aerosol formed from coagulation of primary aerosols to form chain aggregates. The next size range, 0.08 to approximately $1.0 \mu\text{m}$, termed the accumulation range, contains emissions in this size range plus aerosol accumulated by mass transfer through coagulation and

condensation processes from the nuclei range. The last range, 1.0 μm to approximately 40 μm , is termed the coarse aerosol size range. Aerosols within this range generally originate from mechanical processes such as rock fracture and bulk material handling. Mineral dust aerosol reentrained by mine haulage vehicles during the load-haul-dump cycle is an example of an

in-mine source that will contribute aerosol to this size range. A division is usually made between the Aitkin nuclei and the accumulation range aerosol and the remaining coarse aerosol at 1.0 μm . This distinction is possible because sources of aerosol in the two ranges are usually different and the coarse aerosol range contains very little mass transferred from the accumulation range by coagulation.

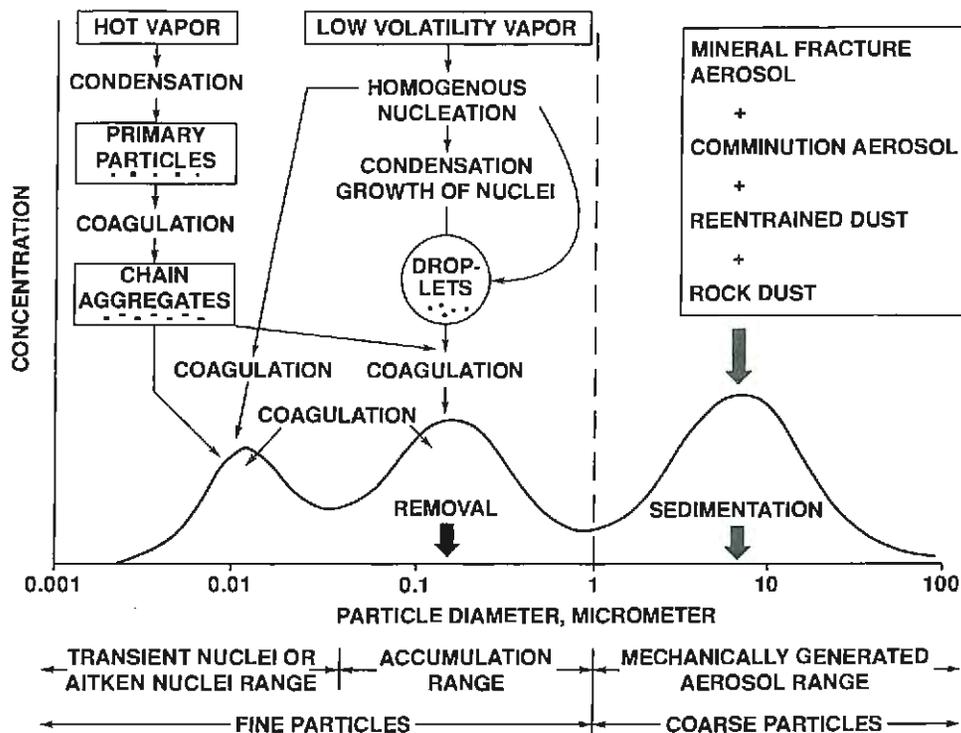


Figure 1: In-mine aerosol characteristics exhibiting the modal nature of the size distribution and conventional terminology for description.

SIZE DISTRIBUTION MEASUREMENT OF DIESEL AND COAL DUST AEROSOL

From the aerosol characteristics summarized in fig. 1, two general observations apply to diesel-exhaust aerosol in underground mines. The first is that diesel exhaust should not be expected to contribute to mine aerosol greater than 1.0 μm in size. It originates as a submicron aerosol and coagulation processes are not sufficient to transfer much mass to the coarse size range. Secondly, mechanically generated aerosols such as coal and mineral, do not have significant mass with size less than 1.0 μm . These two observations gave rise to the

premise on which size-selective sampling was originally proposed for measurement of diesel-exhaust aerosol. To test this hypothesis, a series of laboratory and field experiments were conducted.

Laboratory Study

Laboratory experiments were conducted to investigate the feasibility of using the Micro-Orifice, Uniform Deposit Impactor (MOUDI) *, to measure the size distribution of aerosols containing various mixtures of coal dust and diesel-exhaust aerosols.^{10,11} The objective of the work was to determine the mass

concentration of diesel-exhaust aerosol in an airborne mixture of coal dust and diesel-exhaust aerosol from the size distribution of the mixture. A prototype seven-stage MOUDI with cut sizes ranging from 0.1 to 10 μm was used.¹⁵

The experiment successfully demonstrated that coal dust and diesel-exhaust aerosol can be separated and measured on the basis of size. Data from the tests, typified in fig. 2, show that the overall diesel exhaust/coal aerosol size distribution is bimodal, i.e., it has two maxima or modes. The diesel exhaust, or sub-micron mode aerosol, has a mass median aerodynamic diameter (MMD) of approximately 0.15 μm . The coal dust super-micron mode has a MMD in the 3 to 10 μm size range. A clear separation between the two modes exists in the 0.7 to 1.0 μm size range with the minimum near 0.8 μm . Analysis of the combined size distribution permits a quantitative determination of the diesel aerosol contribution to within 15 percent.

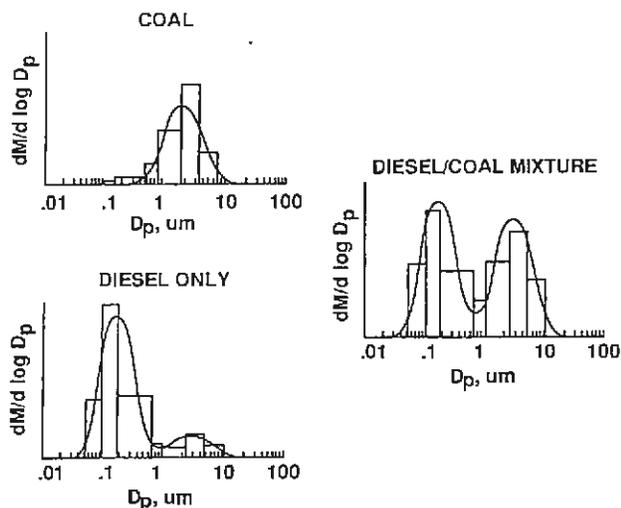


Figure 2: Laboratory diesel/coal aerosol size distributions vs. relative concentration.

Field Studies

Five mines were visited during field studies designed to confirm the laboratory results and develop a data base for use in designing a diesel aerosol sampler.¹⁶ Three of the mines were equipped with diesel haulage equipment and two had all-electric equipment. The electric equipped coal

mines were used to generate comparison samples representative of a mine environment without diesel aerosol.

Measurements consisted of collecting size differentiated aerosol samples at four locations in longwall development sections employing a continuous miner; in the air intake entry; conveyor/beltway entry, air return entry, and haulageway. A ten-stage MOUDI (MSP, model 100)*, operated at a flow rate of 30 L/min, was used for the field size distribution measurements.

Samples used to develop design and performance criteria were those collected at the conveyor belt or in the haulageway, since they are from areas where workers are exposed. These were collected only when the breaker was on and diesel haulage equipment was in use. As a result, the measurements were not representative of worker exposure for the work shift.

Trace element profiles of mine aerosol and related sources were also measured. This included analysis of Indium added to the fuel as a unique diesel source tracer. Source apportionment analysis using chemical mass balance (CMB) modeling were performed on these samples and used to referee results of the size distribution measurements.⁸ The trace element analysis used was instrumental neutron activation analysis (INAA).¹⁷

Field Measurement Results

Average aerosol size distributions measured in the haulageway of the all-electric and diesel equipped coal mines are shown in figs. 3 and 4. The MOUDI collects the sampled aerosol in size intervals by aerodynamic diameter. The number of size intervals provides enough differential size resolution to model the measured aerosol size distributions with empirical functions represented by the continuous curves in the figures. This analysis, termed modal analysis, uses a sum of two log-normal functions to fit the data.¹⁸ Each function represents one of the maxima or modes evident in the data.

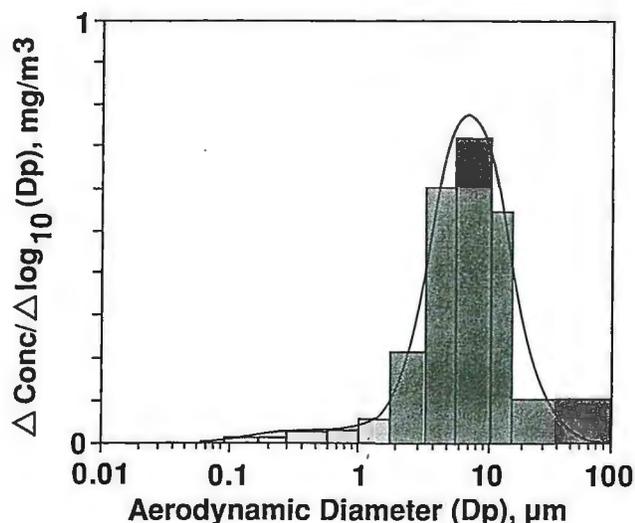


Figure 3: Average mass size distribution measured in haulageways of the all-electric mines.

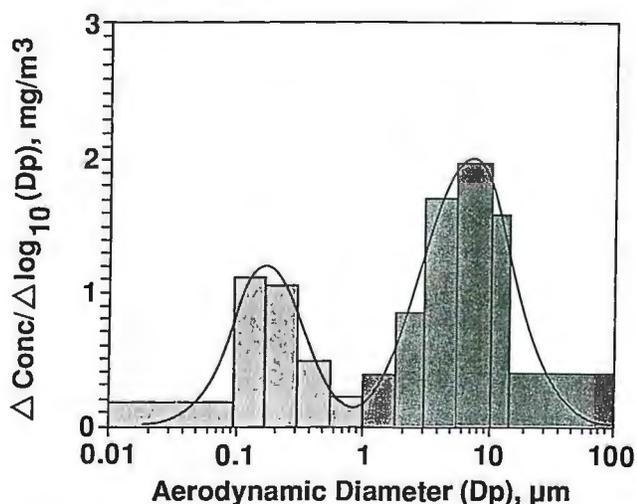


Figure 4: Average mass size distribution measured in haulageways of the diesel equipped mines.

Each mode is identified with the aerosol contributed by a primary aerosol source: diesel-exhaust aerosol for the submicron mode and mineral dust for the coarse particle mode. Under this assumption, the separate contribution from these sources to the total aerosol concentration can be determined using the modal analysis results. Treating each mode as a source-connected entity also permits the determination of the amount of coarse particle mode aerosol that encroaches on a sample of sub-

micron mode aerosol as it might be collected by an ideal single-stage size-selective sampler; i.e., one with a step collection efficiency. This is done by integrating the distribution function for the coarse particle mode over the range of sizes for which the submicron mode aerosol is collected. An illustration of such modal overlap for the average diesel/coal size distribution of fig. 4 is given in fig. 5. The range of integration is 0.001 to 0.8 μm . Shaded areas indicate the portion of the submicron and coarse aerosol that will contribute to an ideal sub-0.8 μm sample.

Design Criteria

Optimum particle size for separation of diesel from mineral dust aerosol was determined by integrating the log-normal functions that describe the size-distribution modes from zero up to a given separation size; as illustrated in fig. 5. In this way, the aerosol mass collected by an ideal sampler with this separation size was determined. Using these results, the percent gravimetric error made by assigning the aerosol mass collected by such a sampler to diesel aerosol alone was determined by its difference from the total submicron mode mass as a function of separation size. The optimum separation size for the personal diesel sampler thus determined is $0.8 \pm 0.1 \mu\text{m}$.

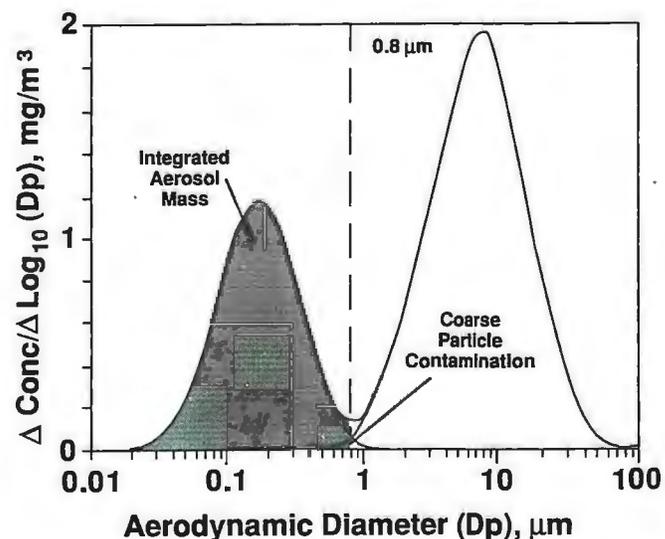


Figure 5: Integration of aerosol mode functions to calculate aerosol mass $< 0.8 \mu\text{m}$.

PERSONAL DIESEL-AEROSOL SAMPLER

A prototype personal diesel-exhaust aerosol sampler has been designed for underground coal mines.^{1,3} The sampler, depicted in fig. 6, has three stages and employs inertial impaction for separation and collection of the diesel and mineral dust fractions of the sampled respirable aerosol. The first stage is an inertial preclassifier that separates and collects the larger, non-respirable aerosol. The preclassifier used in this design is a 10-mm Dorr-Oliver cyclone. Its second stage is a multiple-nozzle impactor with a sharp separation or cut size that only passes aerosol with less than $0.8 \mu\text{m}$ aerodynamic diameter. Here, aerosol larger than $0.8 \mu\text{m}$ is deposited on an impaction plate. The third stage, which is a filter, collects all aerosol less than $0.8 \mu\text{m}$ aerodynamic diameter. The sampler operates at a sampling flow rate of 2 L/min and is designed to be compatible with commercial personal sampler pumps.

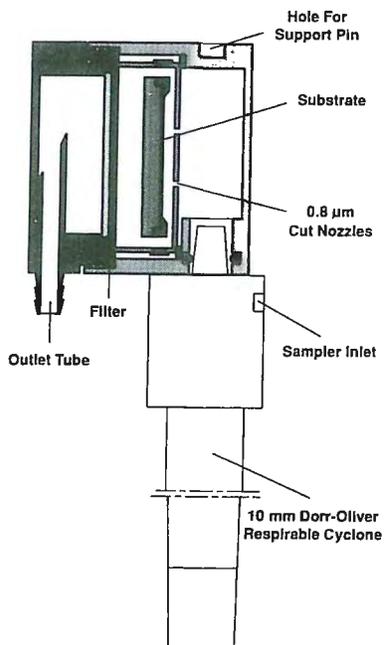


Figure 6: Schematic diagram of the personal diesel-exhaust aerosol sampler.

The $0.8 \mu\text{m}$ cut size impactor was designed using accepted impactor theory and design guidelines.^{1,9,20} An impactor with four nozzles was found to exhibit the best tradeoff between pressure drop, overall particle mass loading, and diameter of the particle deposits on the impaction plate. In this development, impactors with 1, 4, 7, and 22 nozzles were tested with no degradation in collection efficiency or precision of the cut size.

Primary limitations on in-mine personal diesel-exhaust aerosol sampler performance are diesel aerosol loss from the sample, variable contamination by coarse particle mode aerosol, and accuracy of the gravimetric analysis performed on the sample. Of secondary importance is the presence of background aerosol in the sample. The latter is due to external, or atmospheric, and non-diesel out-by sources of submicron aerosol such as welding fumes. The sub- $0.8 \mu\text{m}$ sample contains most of the diesel particulate material present in the mine air, plus a small amount of mineral dust contamination, usually less than 10 pct. If a sample flow rate of 2 L/min is used and gravimetric analysis is to within 0.1 mg, a sub- $0.8 \mu\text{m}$ aerosol concentration is estimated to have an a-priori limit of detection of 0.3 mg/m^3 .

Laboratory Calibration

Collection efficiency of the $0.8 \mu\text{m}$ cut size impactor as a function of aerosol size was determined using monodisperse calibration aerosol. These test aerosols were uniform size polystyrene latex (PSL) particles ranging from $0.5-$ to $1.05-\mu\text{m}$ diameter. Fig. 7 shows the results of these measurements. Here, data for the aerosol collection efficiency of the impactor are presented as a function of particle aerodynamic diameter. The impactor cut size, defined as the particle size corresponding to the 50 percent particle collection efficiency, was found to be $0.76 \pm 0.05 \mu\text{m}$. The sharpness of cut is measured by the geometric standard deviation of the collection efficiency curve. This was

found to be 1.15, indicating a sharp cut for the impactor. The geometric standard deviation is calculated as the square root of the ratio of the particle diameter corresponding to the 84.1 percent collection efficiency to the diameter at an efficiency of 15.9 percent.

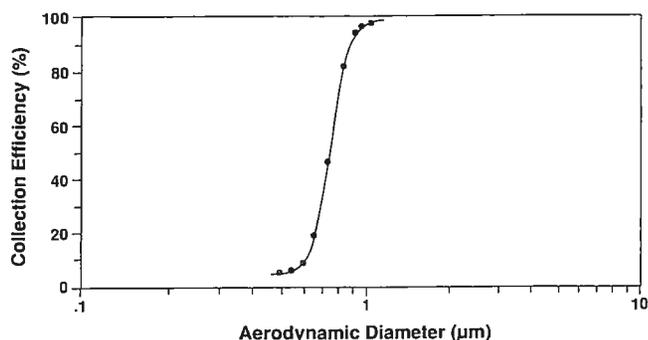


Figure 7: Particle collection efficiency of the 0.8 μm impactor.

Field Evaluation

The personal sampler has been evaluated in comprehensive sampling studies performed in three underground coal mines. In each mine, 12 samplers were deployed in 6 different locations of a continuous miner section utilizing diesel-powered shuttle cars. These locations were the intake entry, haulageway, return entry, continuous miner, each shuttle car, and, in a few instances, on one of the research staff.

A comparative study was made of data obtained with the personal sampler and that computed from an analysis of detailed size distributions obtained using a MOUDI. The average fraction of respirable material less than 0.8 μm was 30 percent at the continuous miner and in the return entry, 65 percent in the haulageway, and 50 percent for the research staff member who spent time at each test site.

ONGOING AND FUTURE RESEARCH

Evaluation of the personal sampler is continuing. Detailed tests are

planned for at least two more mines. In addition to comparison with the MOUDI, several referee methods, including CMB model analysis of aerosol elemental composition, evolved gas analysis of aerosol carbon and nitrogen, and Raman spectroscopy analysis of sample carbon are being used in these tests. MSHA is also using this sampler, plus others, in additional field tests that include some metal and nonmetal mines.^{2,1}

CONCLUSIONS

The primary results of this study are:

- (1) Based on the premise that diesel-exhaust aerosol is predominantly submicron in size and coal dust is largely greater than a micron in size, size-selective sampling can be effective as a technique for measuring diesel-aerosol concentrations in underground coal mines.
- (2) In-mine measurement of mine aerosol size distributions and CMB analysis confirms the premise of using aerosol size to separate diesel exhaust and mineral dust aerosol during sampling.
- (3) A personal diesel-aerosol sampler has been developed for measuring the diesel-aerosol concentrations in underground coal mines. This device consists of three sequential stages. The first stage preclassifier limits the sample to respirable aerosol. The second stage separates the resulting respirable aerosol into two parts by aerodynamic diameter using a single-stage impactor with a cut size of 0.8 μm . The third stage collects the < 0.8- μm -diameter aerosol on a media that is suitable for gravimetric analysis.
- (4) The sampler is currently being evaluated in both laboratory and field experiments conducted by the Bureau, MSHA, and the GMT-C-RD.

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