

## Characterization and Control of Fine Particles: Overview of NRMRL Research Activities

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### Abstract

Research at the U.S. Environmental Protection Agency's (EPA's) National Risk Management Research Laboratory (NRMRL) in the area of particulate matter (PM) is designed to provide critical information regarding emission rates, characteristics, and control approaches for PM 2.5  $\mu\text{m}$  in aerodynamic diameter and smaller (PM<sub>2.5</sub>). NRMRL researchers are also studying penetration of outdoor particles into indoor environments as well as indoor sources of particles.

PM<sub>2.5</sub> source profiles are being developed using a dilution sampling system to capture both primary particles and particles formed by condensation of organic compounds. This research will provide information needed by source receptor models to identify the contribution of different sources to the ambient particle loading. Emissions characterization research is being conducted on particle size and composition for residential wood combustion systems such as woodstoves and fireplaces, for industrial combustion sources burning coal and heavy fuel oil, for diesel truck engines during on-road operation, and for open burning of biomass. Emission inventory research on ammonia from animal feeding operations and fugitive dust from construction activities will improve understanding of PM<sub>2.5</sub> sources and their impact on atmospheric chemistry and ambient PM<sub>2.5</sub> concentrations. NRMRL researchers are studying innovative control technologies for stationary sources. The technologies being evaluated include improved primary particle collection systems and multipollutant controls such as fluid bed absorber systems. Research related to indoor PM<sub>2.5</sub> includes a study of the penetration rate of outdoor particles into indoor environments, as well as research into methods of measuring the contribution of indoor particle sources to total indoor PM<sub>2.5</sub> concentration.

NRMRL researchers are also collaborating with health effects scientists to help link particle characteristics to health effects, with atmospheric scientists to better understand the fate of particles in the atmosphere, and with other organizations to create a base of broad scientific expertise and interaction.

### Introduction

Over the last several years, increasing numbers of studies have shown an association between increases in ambient concentrations of particulate matter (PM) smaller than 2.5  $\mu\text{m}$  in aerodynamic diameter (PM<sub>2.5</sub>) and increases in adverse health effects, including increased mortality rates.<sup>1,2,3</sup> These studies led to the revision of the National Ambient Air Quality Standard (NAAQS) for PM. The NAAQS revisions added two new primary PM<sub>2.5</sub> standards: a 65  $\mu\text{g}/\text{m}^3$  24-hour average, and a 15  $\mu\text{g}/\text{m}^3$  annual mean. At the same time, the current 24-hour and annual standards of 150 and 50  $\mu\text{g}/\text{m}^3$ , respectively, for PM smaller than 10  $\mu\text{m}$  in aerodynamic diameter (PM<sub>10</sub>) were retained.<sup>4</sup>

The U.S. Environmental Protection Agency's (EPA) Office of Research and Development (ORD) has designed and is implementing a comprehensive research program to address the scientific questions associated with PM<sub>2.5</sub>, including additional epidemiological studies to evaluate potential subpopulations who may be more susceptible to increased levels of ambient PM<sub>2.5</sub>, investigations into the possible causes of the health effects associated with higher ambient PM<sub>2.5</sub> concentrations, enhanced ambient monitoring, improved atmospheric chemistry models, studies characterizing emissions from different sources, including sources found indoors, and evaluations of air pollution

control technologies for new and retrofit applications. Emissions characterization studies and control technology development research are being conducted by ORD's National Risk Management Research Laboratory (NRMRL), and are the subject of this paper.

### **NRMRL Research Activities**

In order to accurately model ambient concentrations of PM<sub>2.5</sub>, accurate inventories of source emissions of both primary particles and secondary particle precursors must be available as input to those models. Mass emission rates as a function of industrial or personal activities, species of compounds, and size distribution of particles are all important to understanding the atmospheric formation of secondary particles and the deposition rates of particles. Such information is also important to health scientists who are seeking to identify specific ambient particle characteristics (such as size distribution and chemical composition) that may be linked to adverse health impacts. Implementation of the standards will require not only an accurate characterization of emissions, but also an understanding of what technologies can cost-effectively reduce such emissions. NRMRL PM<sub>2.5</sub> research is characterizing particle emissions and improved methods for reducing those emissions to support the Agency's goal of reducing risk associated with exposure to ambient concentrations of PM<sub>2.5</sub>.

### Source Sampling and Source Profiles

Conventional PM source sampling methods intentionally prevent the condensation of many organic compounds. However, ambient PM<sub>2.5</sub> samples indicate relatively large amounts of organic carbon (OC), or carbon-containing organic compounds. It is important to understand the contribution these compounds make to the total ambient concentration for two reasons. First, urban ambient air samples contain 20-30% or more OC, a significant fraction of the total ambient mass. Second, specific organic compounds may act as "marker" compounds that can be used to link specific source categories to their contribution to ambient samples. Therefore, it is important to conduct sampling operations in such a way that as much as possible of the condensible organic fraction of the exhaust gases is collected. NRMRL has constructed a modified version of the dilution sampling system pioneered by the California Institute of Technology to allow collection of these organic compounds.<sup>5</sup> This sampling system is shown in Figure 1.

Sampled flue gas passes through a cyclone to remove particles larger than 10 μm in aerodynamic diameter, then through a heated sampling line into a U-shaped dilution chamber. The flue gases (collected at a rate of 15-50 Lpm) are diluted with up to 1500 Lpm of clean dry air to achieve dilution ratios of up to 100:1 (air to flue gas). Cooled and diluted particle samples can be collected from a filter at the bottom of the dilution chamber, or a portion of the sample can pass into a residence time chamber that provides a longer residence time for additional condensation and chemical reaction to occur prior to sampling. Sampling ports at the bottom of the residence time chamber allow several measurement systems (scanning mobility particle sizing system, organic compound sampling system, or others) to collect samples simultaneously.

Sources were chosen to be tested based on several criteria. The source category's national ranking as measured by total mass emissions, the lack of one or more known elemental markers, the presence of organic compounds, and the lack of previous source profile information were all taken into account when determining which source categories would be most strongly considered. Sources that have already been tested include open burning of biomass, residential wood combustion, and diesel trucks during on-the-highway operation. Among the sources that are being considered or planned for future tests are wood-fired industrial boilers, pulp and paper plant operations, and commercial jet engines.

### PM from Stationary Combustion Sources

Work at NRMRL has identified some fundamental characteristics of particles generated by the combustion of heavy fuel oil and pulverized coal.<sup>6,7</sup> There is a large body of literature describing

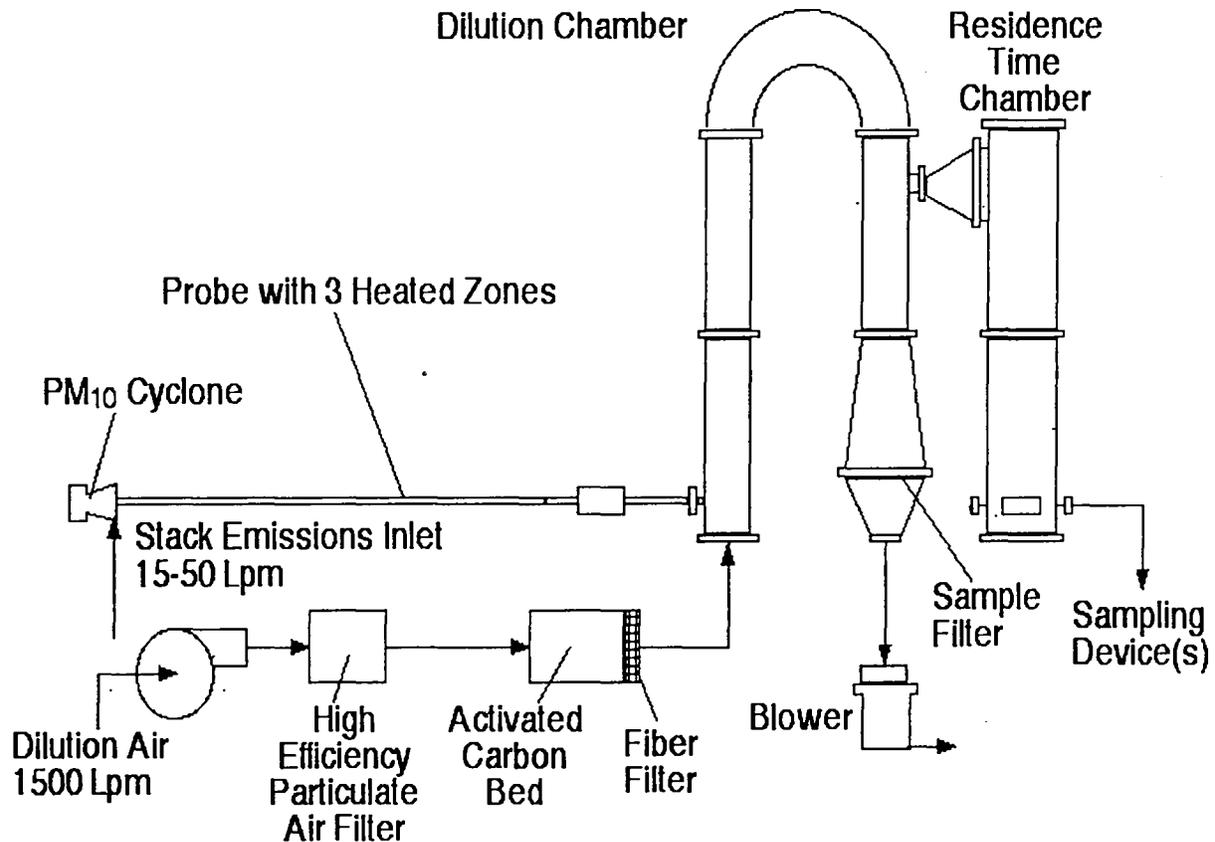


Figure 1. Schematic of NRMRL's dilution source sampling system for measuring  $PM_{2.5}$ .

the characteristics of PM generated by the combustion of pulverized coal. However, much of this work is relatively old and did not have access to modern measurement methods and equipment.

There is relatively less information available characterizing PM from residual fuel oil combustion. For both heavy fuel oil and pulverized coal, NRMRL has been working collaboratively with EPA's National Health and Environmental Effects Research Laboratory (NHEERL). This collaboration has provided a significantly different perspective on what characteristics and constituents are of interest. Previous work<sup>6</sup> focused on measurement of trace metals that are considered to be carcinogenic or toxic, while first-row transition metals (copper [Cu], iron [Fe], nickel [Ni], vanadium [V], and zinc [Zn]) are of interest in the current NRMRL/NHEERL work. In addition, factors such as ash melting temperature and resistivity, studied to improve operation and pollution control system design, are less important from the perspective of understanding health effects. The NRMRL/NHEERL studies have emphasized a loosely defined "bioavailability" that indicates the propensity of a substance to be absorbed into the body or captured by the body's defense mechanisms. A relatively simple surrogate for bioavailability is the solubility of a sample in deionized water.

Recent tests at NRMRL measured the particle size distributions and the elemental compositions of PM generated by the combustion of residual fuel oil and pulverized coal.<sup>6</sup> These tests show that the water solubility of transition metals in oil fly ash is substantially higher than for the bulk of pulverized coal fly ash, as shown in Figure 2. This difference in solubility indicates that oil fly ash is potentially more toxic than a coal fly ash containing a similar amount of transition metal due to a higher bioavailability. However, these tests have not yet been able to measure the solubility of the

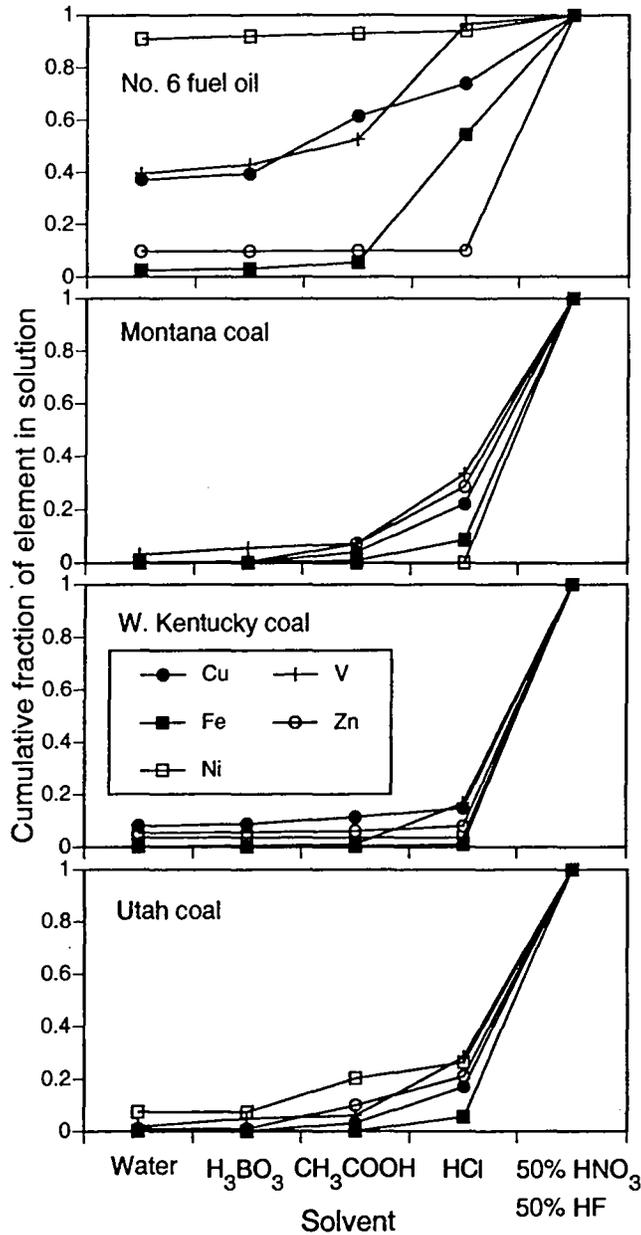


Figure 2. Cumulative fraction of elements in solution for particles generated by the combustion of No. 6 fuel oil and three pulverized coals.

ultrafine ( $< 0.1 \mu\text{m}$ ) fraction of the pulverized coal fly ash.

The mechanisms governing particle formation suggest that the ultrafine fraction is formed by nucleation of vaporized material, largely metals, and the chemistry of the combustion gases at the nucleation temperatures is such that these metals have the potential to be in relatively soluble forms based on their likely chemistry. Earlier work found that the ultrafine fraction of residual fuel oil fly ash was largely composed of sulfate and metals,<sup>7</sup> and similar processes governing coal fly ash particle formation would suggest that the ultrafine fraction of particles from pulverized coal combustion may have characteristics similar to oil fly ash. The presence of a distinct ultrafine mode generated by nucleation processes has been identified in coal particles, in addition to a mode near  $1 \mu\text{m}$  and a larger mode typical of mechanical processes such as grinding (see Figure 3). Further work is being conducted at NRMRL to investigate the behavior of coal fly ash in more detail.

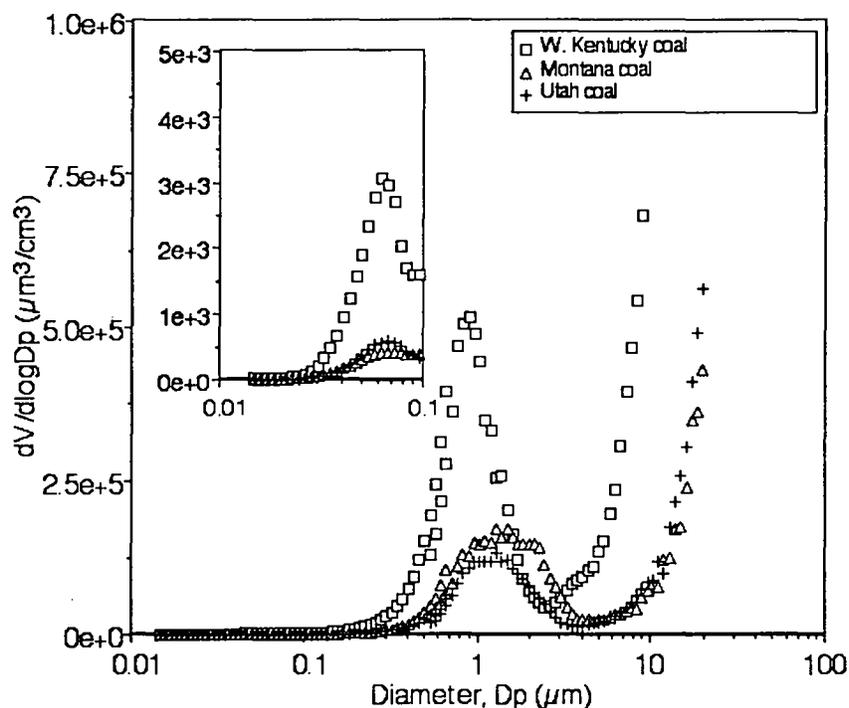


Figure 3. Particle size distributions measured for particles from the combustion of three different pulverized coals.

Source emissions tests have also been conducted to determine mass, size, and composition of particles from residential wood combustion and open burning of biomass. For both of these source types, samples have been collected to determine the types and amounts of organic compounds that are emitted to identify potential marker compounds.<sup>8</sup>

#### PM and PM Precursors from Mobile Sources During On-the-Highway Operation

NRMRL researchers have developed and employed several methods to measure pollutant emissions from mobile sources during operation on the road. The majority of data for mobile sources is collected using dynamometers with operating cycles designed to simulate actual operation. Because these tests are simulations, they do not capture the full range of operating conditions that are experienced during on-road operation. NRMRL's work has focused on determining emissions while vehicles are operating on the highway under real-world conditions.

#### *PM from Diesel Trucks*

PM emissions from diesel engines has also been of concern, since they represent 77% of the primary PM<sub>2.5</sub> emissions from on-road sources and 11% of primary PM<sub>2.5</sub> from non-fugitive sources. These emissions also have characteristics believed to play significant roles in causing adverse health effects, such as small particle size (almost entirely < 1 µm in aerodynamic diameter) and high levels of organic and elemental carbon.

Among the most visible diesel sources are heavy-duty trucks. Conventional emissions sampling from these sources relies upon measurements taken while the engine is mounted to a stationary load that can be varied to simulate actual driving conditions. A more realistic approach is to develop a method that will measure actual emissions during on-highway operation. The on-highway approach can then take measurements for a truck pulling varying loads, at varying road grades, speeds, and acceleration rates. Ideally, one would sample directly from the exhaust plume, after the pollutants had mixed with ambient air and cooled, allowing the natural nucleation and condensation

of gas-phase organic compounds. With this approach, source sampling would capture particles with essentially the same characteristics as those in the ambient atmosphere. However, because the plume moves and because the ambient air, especially along highways, contains pollutants from other sources, the next best approach is to sample in the exhaust stack and use a dilution chamber to simulate the plume.

Current work at NRMRL is comparing results from a dilution sampling system with simultaneous direct plume sampling during on-the-highway operation of a heavy-duty diesel truck. Sampling systems are placed in the truck cab and in the trailer (see Figure 4), and measurements are taken as the truck covers a predetermined route along different grades. Engine and operational variables such as speed, acceleration, engine revolutions per minute (RPM), and exhaust temperature are also measured. Particle samples are collected with an electronic low-pressure impactor (ELPI) that quantifies PM sizes in 12 size bins from 0.032 to 10  $\mu\text{m}$  in aerodynamic diameter at the rate of one measurement per second.

Dilution of the exhaust plume is determined by measurements of carbon dioxide ( $\text{CO}_2$ ) in the stack and in the plume. The exhaust plume was sampled at different locations along the length of the trailer to evaluate plume behavior. These measurements were used to determine the dilution ratio and residence time for the diluted stack sample, so that the diluted stack samples would most closely simulate the actual exhaust plume behavior. The stack sample is initially diluted with dry filtered air at a ratio of 7:1 (air to exhaust), with a residence time of approximately 3 ms to simulate the initial rapid mixing of the exhaust with turbulent air. A second stage further dilutes this mixture with a 10:1 ratio of dry filtered air to diluted exhaust, over a residence time of approximately 500 ms, representative of the slower dilution along the length of the trailer. A final stage of dilution is used to represent the rapid mixing of the plume and air at the end of the trailer. In this stage, the diluted plume is mixed with additional dry filtered air at a ratio of 7:1 (air to diluted exhaust) in approximately 2 ms.

Results of these tests to date indicate that stack sampling with the dilution system measures higher levels of PM than are measured in the naturally diluted exhaust plume (see Figure 4). The size distributions are also slightly larger for the diluted stack sample than for the plume measurements. For several different operational test cases, the results consistently demonstrated a similarly shaped particle size distribution, with the peak of the mode near 0.2  $\mu\text{m}$  in aerodynamic diameter. Although additional work is required to more accurately represent the natural dilution of the exhaust plume, the results to date have shed light on the mechanisms governing particle formation through nucleation and condensation, which may in turn have significant implications regarding the composition of the different particle sizes.<sup>9</sup>

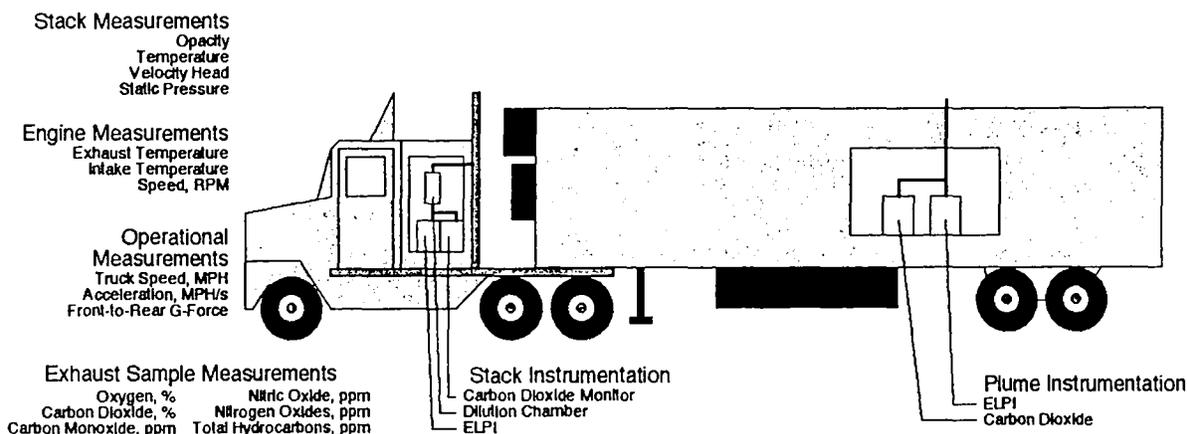


Figure 4. Schematic of diesel truck instrumented for on-the-highway measurement of  $\text{PM}_{2.5}$ .

### *Ammonia from Passenger Automobiles*

Ammonia emissions from automobiles, like emissions of carbon monoxide (CO) and nitrogen oxides (NO<sub>x</sub>), varies as the driving conditions (such as acceleration and load) vary. Ammonia (NH<sub>3</sub>) emissions are generated by high levels of nitrogen and low levels of oxygen in the exhaust, which results in the formation of NH<sub>3</sub> in three-way catalysts. Measurement of ammonia emissions during on-the-highway operation requires ammonia monitors that respond quickly to changes in emissions, since the conditions that lead to NH<sub>3</sub> production occur rapidly. NRMRL researchers have been examining different methods to quickly measure NH<sub>3</sub> emissions by simultaneously measuring NO<sub>x</sub> and total nitrogen, and determining NH<sub>3</sub> by calculating the difference. This work has led to a better understanding of the engine and driving conditions that lead to NH<sub>3</sub> emission peaks, information that can be used to generate improved emission inventories for mobile sources.<sup>10</sup>

### Area Sources

The disperse nature of area sources such as construction activities, agricultural operations, and fugitive emissions makes it difficult to quantify those emissions. Typical conditions can be difficult to quantify, and inconsistent sampling conditions and the need to use unique sampling approaches complicate the task. NRMRL has conducted several projects, in addition to the open biomass burning project noted above, to quantify and characterize emissions from area sources.

### *Ammonia from Animal Feeding Operations*

Animal feeding operations are relatively large sources of NH<sub>3</sub> due to the generation of waste from the animals. In some cases, such as for hogs raised in some locations of the U.S., the waste is collected in lagoons and may then be sprayed onto fields to provide a source of nitrogen. Measuring NH<sub>3</sub> emissions from such large sources can be difficult because of the lack of a single exhaust point and the variation in emissions as a function of meteorological variation. One approach being tested at NRMRL is the use of open-path Fourier transform infrared (OP-FTIR) sensing, which allows one to aim the instrument across the plume and determine the concentrations of specific pollutants. NRMRL researchers have measured NH<sub>3</sub> emissions from both a waste lagoon and an animal feeding barn (the waste is generated in the barn and transported to the lagoon) to determine the relative strength of emissions from both facilities.

By measuring during a range of meteorological conditions and with varying numbers of animals present, the governing parameters can be examined to determine the most important factors influencing emissions. Results to date indicate that the OP-FTIR method can be used to measure NH<sub>3</sub> emissions from animal feeding operations, and that there is less variability with respect to the number and size of animals present than was previously expected.<sup>11</sup>

### *Construction Dust*

NRMRL has conducted several tests to measure the dust generated by certain construction activities, in particular earth-moving operations using large construction equipment.<sup>12</sup> A series of tests were conducted using ambient PM samplers placed downwind of diesel scraper-loaders operating over a range of conditions (loading, unloading, transiting). Soil moisture was also measured to evaluate how dust generation was affected by changes in soil conditions. The data collected from these tests are still being analyzed.

### Indoor Air Quality

Particle emissions do not only occur outdoors, but are also generated by indoor activities such as space heating and cooking. Particles can also penetrate into buildings from the ambient environment through open doors and windows and through heating and cooling systems. Research at NRMRL is characterizing both indoor source emissions and penetration of ambient particles into indoor environments.

### Indoor Sources

Using both a test chamber and a residential house dedicated to testing purposes, NRMRL researchers are measuring the particle concentrations generated by indoor activities such as cooking, space heating, and use of candles and incense. Under certain conditions, some of these activities can generate significant amounts of  $PM_{2.5}$  in relation to the volume of a room or a house, and under the appropriate conditions, can result in  $PM_{2.5}$  concentrations higher than the  $PM_{2.5}$  NAAQS. Particles generated by the use of candles also tend to be smaller than  $1\ \mu m$  in aerodynamic diameter. A particle size distribution measured using an electronic low pressure impactor (ELPI) for PM from a scented paraffin candle with three wicks is shown in Figure 5. Particle sizes during normal candle burning and during smoldering are presented, and both demonstrate that a majority of particles from such activities are submicron in size. While smoldering tends to increase the particle sizes, the general shape of the distribution is similar to that for normal candle burning.<sup>13</sup>

In order to accurately measure the particles generated by specific activities, indoor air quality researchers at NRMRL have also evaluated measurement methods using tracer compounds such as sulfur hexafluoride ( $SF_6$ ) and the potential for these compounds to break down as they pass through high temperature regions in heating and cooking flames.<sup>14</sup>

### Penetration of Outdoor Particles into Indoor Environments

Although the epidemiological studies noted in the introduction relate ambient particle concentrations

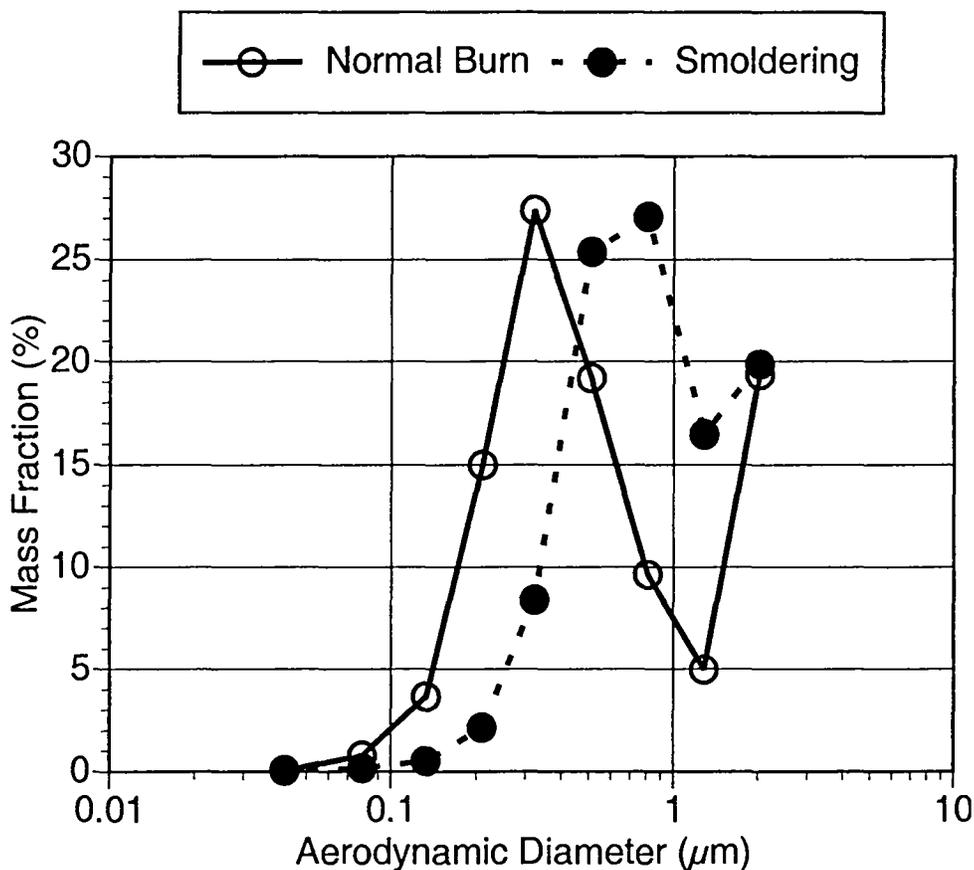


Figure 5. Distribution of particle sizes from a normally burning (open symbols) and smoldering (solid symbols) scented paraffin candle.

to adverse health effects, most people spend the majority, in many cases over 90%, of their time indoors. This is especially true for those who may be particularly susceptible to the effects of air pollution, such as those with asthma or other pulmonary disease. Even those indoors can be exposed to ambient particles, however, since air passing through doors, windows, heating and air-conditioning systems, or cracks can carry ambient particles. NRMRL is conducting research to better understand the mechanisms governing the penetration of ambient particles into indoor environments. Studies of the impact of particle size, differential pressure between outdoors and indoors, and characteristics of cracks have led to the development of a model to predict the particle penetration rate as a function of particle size for different conditions.<sup>15</sup>

#### Control Technology Development

In addition to emissions characterization research, NRMRL has also been evaluating improvements to existing PM control technologies and development of new PM control technologies. New technology development and testing has focused on multipollutant control capability to allow users to minimize total cost and tailor the control system design to meet a number of pollution control needs.

#### *Electrostatic Fabric Filter Testing*

In conjunction with Southern Research Institute and the Southern Company, NRMRL researchers are conducting tests on an electrostatic fabric filter (ESFF) that improves collection of PM<sub>2.5</sub> without significant increases in fan power requirements. This approach allows the installation of a set of filters inside an existing electrostatic precipitator (ESP) housing so that existing particle control systems can be upgraded without requiring additional space. The ESFF can improve performance of either a conventional fabric filter or an existing ESP, particularly with respect to removal of PM<sub>2.5</sub>.

The pilot-scale ESFF is a 0.3 MWe scale pulse-jet cleaned fabric filter fitted with an array of high-voltage electrodes between the bags, as shown in Figure 6. The pilot-scale design has sixteen bags, with a high-voltage array composed of nine corona electrodes suspended from the tubesheet with insulators connected to the high-voltage source on the clean side of the tubesheet.

#### *Circulating Fluidized Bed Absorber*

Use of a circulating fluidized bed absorber (CFBA) system for control of PM and PM precursors such as sulfur dioxide (SO<sub>2</sub>) can reduce the total cost of pollution control for a plant, and has the potential to fit within existing space in retrofit applications. NRMRL is conducting research to develop a computational model of a CFBA system to allow designers to determine size and operating requirements for specific pollution control scenarios. For instance, a high sulfur, low ash coal may require one set of operating parameters, while a low sulfur, high ash coal may require another. Plants that need the flexibility to switch fuels may wish to have a system that will allow adequate capture of both PM and SO<sub>2</sub>, and the potential need to control other pollutants such as mercury can also influence operating parameters. With the development of a computational model, NRMRL researchers will be able to identify design and operational parameters that are optimized for these and other instances.

#### **Conclusions**

Development of emission inventories, emissions characterization data for use in atmospheric chemistry models, and performance of improved and new technologies are critical to being able to gain a clear understanding of the sources, ambient concentrations, and effects of PM<sub>2.5</sub>. NRMRL's research into source characterization, indoor air quality, and control technology development will assist the successful and cost-effective implementation of EPA's PM<sub>2.5</sub> ambient air quality standard.

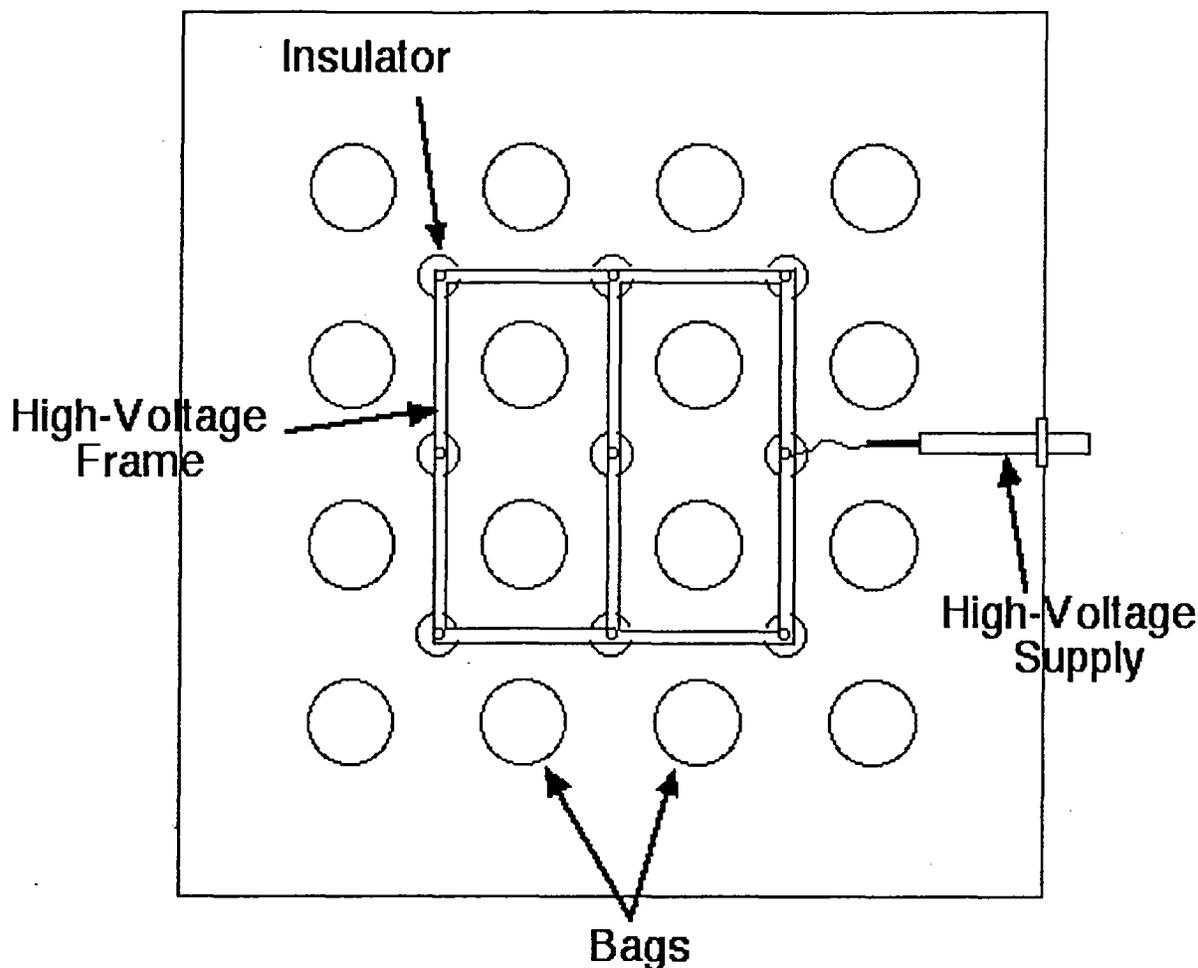


Figure 6. Schematic of electrostatic fabric filter arrangement (plan view).

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17. KEY WORDS AND DOCUMENT ANALYSIS			
a. DESCRIPTORS		b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Pollution	Coal	Pollution Control	13B 21D
Particles	Fuel Oil	Stationary Sources	14G
Emission	Biomass	Indoor Air	08A
Organic Compounds	Ammonia	Feed Lots	07C 07B
Combustion	Construction Materials	Fugitive Dust	21B 13C
Wood			11L
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