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Notes from the 2003 MIT Symposium on Particulate Matter

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MARAMA was among the symposium's co-sponsors, which also included NESCAUM, EPA, DOE, DOT, CARB, the Health Effects Institute, the National Commission on Energy Policy, the Diesel Technology Forum, API, EPRI, Cummins, Exxon, Ford, PSEG, and Sunoco. Bob Slott of the MIT Earth, Atmospheric, and Planetary Sciences Department, and Praveen Amar of NESCAUM were the primary organizers of the symposium.

Presentations on the following major topics are summarized below: PM characteristics, PM standards, public outreach information, PM monitoring, health effects, analysis of PM, control strategies, and control policy.

PM Characteristics

Nobel laureate Mario Molina of MIT was the first speaker, discussing atmospheric chemistry and PM. (He shared the 1995 prize in chemistry for his work in the early 1970s explaining the atmospheric chemistry of the ozone layer.)

Molina noted the complexity of particulate matter, citing the heterogeneity found even in a single particle of soot. He identified a need for more measurement of very short-time variation in the chemical composition of particles and particle counts.

He and Luisa Molina have studied air quality in Mexico City, and he explained that the PM_{10} and ozone improvements there were due to reduced sulfur in fuel, catalytic converters on some cars, and meteorological variation, as well as (somewhat) to car inspections. He commented that the ambient standards are goals, not the ultimate goal (i.e., a healthy environment).

Jim Vickery (EPA, ORD) provided an excellent summary of the NARSTO PM Assessment. He stated that in general about two thirds of PM is anthropogenic in origin. Sources and properties of PM vary by region and season, so control strategies will also need to vary. Organic carbon is the most poorly understood constituent of PM, and on average it constitutes about one fourth of the mass. Analytical tools are still being developed, and there is low confidence in organic carbon modeling. Analysis requires the use of a combination of several tools. The effect of reducing NO_x and VOC emissions may be either positive or negative for PM. Further research is needed in carbon, long-term monitoring, modeling, emissions, and analytical protocols.

PM Standards

John Bachmann (EPA, OAQPS) covered the air quality standards and regulatory framework. Due the complexity of PM, it is difficult to determine what aspect of PM is causing health impacts. Bachmann predicted that researchers will not come up with a "silver bullet," and that the current approach to regulating PM mass (as opposed to particle count or some specific constituents of PM) will continue for the foreseeable future. He commented on the controversy over health effects of sulfates and stated that most studies show that sulfates do have health effects.

He stressed the high priority EPA places on implementing the current PM standard and predicted that the EPA Staff Paper (summarizing the Criteria Document and making recommendations concerning revisions to the PM NAAQS) would recommend a tighter $PM_{2.5}$ standard as well as

a new PM-coarse standard. He expects EPA to propose a revised PM standard in spring 2005. PM-coarse will be much more complex to analyze than $PM_{2.5}$.

Given the regional nature of PM, Bachman said EPA had concluded that it was better to start with a regional strategy and then add local controls, rather than the other way around. He identified three options for addressing transport:

- The Clear Skies Act (for utilities)
- An EPA Transport Rule (for utilities and other sources)
- A combination of a new law for utilities and a new regulation for other sources.

EPA is working to adopt a transport rule as soon as practicable, but it will be effective in 2010 at the earliest. Pittsburgh is one of the areas with remaining $PM_{2.5}$ problems in 2020 after the full implementation of the Clear Skies proposal. He emphasized the importance of local controls.

Bachmann sees 2010 as an important crossroads. The first phase of the Clear Skies or transport rule will go into effect, but there will also be new information about health effects. There may be a need to have a shorter-term standard, or to look at components of PM in addition to $PM_{2.5}$ mass.

Public Outreach Information

Tom Curren (EPA, OAQPS) talked about EPA's trends report and the initiative to start forecasting $PM_{2.5}$ October 1, 2003. He noted that PM mapping and forecasting were more complex than ozone mapping and forecasting. He suggested that with an annual standard one might want to track year-to-date values versus normal values in order to give the public a better idea of whether the current year was cleaner than average or not. He noted that coarse particles will be a greater problem in the west than the east. He also showed that reductions in SO_2 required by the acid rain program have resulted in reductions in sulfates and have brought $PM_{2.5}$ down.

PM Monitoring

Judy Chow (Desert Research Institute) noted that the nature of a monitoring network strategy depends on the purposes of the monitoring. She noted that spatial interpolation requires a dense network, and that assessing the impacts of nearby sources requires short-term monitoring.

Chow noted that PM monitoring is subject to various errors introduced by the equipment, the filters, the procedures for handling samples, etc. For example, nitrate is volatile and can evaporate in hot weather when a filter is left in the field. Organic carbon measurements are especially sensitive to the type of filters used.

Chow emphasized the need for more work to understand the differences between Federal Reference Method monitors for $PM_{2.5}$ and continuous monitors. She noted the difficulty in measuring certain metals, and commented on the differences between morning and afternoon particles. She noted that PMcoarse is very different in composition than $PM_{2.5}$, and so monitoring strategies may differ.

Chuck Kolb of Aerodyne Research, Inc. also discussed monitoring methods. His bottom line was that the FRM is not the best we can do now, and that continuous monitors should be tested with calibration gasses rather than compared to FRMs. He stated that continuous sampling provides greater time and spatial resolution and can do more to help us understand PM.

Phil Hopke of Clarkson University, who is Chair of EPA's Clean Air Science Advisory Committee (CASAC), presented a critique of EPA's national monitoring strategy. He complimented EPA on being willing to make major changes in current PM monitoring. He emphasized the need to obtain rural background concentrations as well as

population exposure. He encouraged EPA to work on harmonizing the speciation trends network (STN) and IMPROVE monitoring methods to facilitate urban vs. rural comparisons. CASAC's review of the monitoring strategy is due by the end of September, and Hopke expects the strategy to be implemented between 2003 and 2007. He sees it as a good base to build on. He would like to see half of the FRM monitors replaced with continuous monitors.

David Kittelson (U of Minnesota) commented that very small "nano" particles from diesel engines are very important. They represent 90% of the number of particles, and more than 30% of the particle mass. Modest changes in dilution conditions substantially change measurements of emissions of various particle sizes. He advocated more research and measurement of these particles.

Health Effects

Michael Brauer of the U of British Columbia discussed factors influencing exposure to PM. He said 40-70% of a person's exposure to PM is linked to outdoor concentrations. He divided people into three classes: 1) those exposed to tobacco smoke, 2) the elderly, whose biggest exposure comes from ambient PM that has come indoors, and 3) the general population. Factors which influence exposure include the city you live in, your proximity to roads or other sources, local topography and meteorology, variation within your city in particle size and composition, and the time of day/day of year. In-vehicle exposure can exceed ambient levels and is a significant part of a person's total exposure. Sulfate exposure is much more highly correlated to ambient exposure than is total PM exposure. The increase in air conditioning has led to a lower correlation between ambient PM levels and hospital admission for heart problems.

Nancy Brown (Lawrence Berkeley National Labs) discussed studies of indoor PM exposure. She noted that personal exposure to PM is much more linked to ambient PM than ozone exposure is to ambient ozone.

During the Q&A period, advice was given to stay indoors during peak ozone periods but otherwise to open windows or go outdoors in order to lower PM exposure.

Ron Wyzga of EPRI described a five-year study in the Atlanta area looking at air pollution levels and mortality and morbidity. The only significant association between air pollution and daily mortality was found in daily CO concentrations. Wyzga said it's not clear whether CO is an indicator of some other problem or a direct cause. The spatial homogeneity of ozone was quite high within the region, and $PM_{2.5}$ exhibited moderate to high homogeneity. George Allen (NESCAUM) noted that Atlanta is largely air conditioned. Wyzga noted that most cardiovascular effects occurred in the winter when there was a lot of woodburning.

Mark Utell (University of Rochester) explained a variety of heart problems related to PM exposure. He noted there has been major progress in the last five years in studying why PM causes cardiopulmonary effects. He commented that ultrafine PM reduces the ability of the lung to transfer oxygen to the blood, and that this may help explain the CO effect in EPRI's Atlanta study.

Doug Dockery (Harvard) explained there is a lag of a few days after a high PM event before health effects are manifested. Mortality effects are seen three days to two weeks after a pollution event. Respiratory effects may occur three weeks after the episode. He discussed the Harvard 6 Cities study (1974-1989) as compared to the American Cancer Society study of 1982. He believes the ACS study underestimated risks, and that the Harvard study did a better job of quantifying the effects of changes in PM.

Tom Grahme (DOE) criticized the design of the 6 Cities Study and concluded that a large

part of the mortality shown in studies of PM may be due to exposure to high levels close to highways. He noted that in the 6 Cities Study, Steubenville's improvements in PM and health were due to reductions in coke oven emissions, which are very toxic. Similarly, in St. Louis, a coke company and a lead smelter shut down. He cited a review in the March 2003 "Inhalation Toxicology" journal that assessed sulfate and nitrate toxicology studies and found no adverse effects of secondary sulfates and nitrates. He suggested that exposure to urban traffic could explain PM risks.

Dan Greenbaum (Health Effects Institute) commented that it is clear that traffic emissions are of concern, but that it's too early to conclude that there is no risk from sulfates. He noted that EPA is putting substantial mobile source emissions controls in place. He reiterated John Bachmann's point that it is unlikely that there is a "silver bullet" responsible for PM health effects. We know more than we did five years ago, and we will continue to learn more.

Analysis of PM

Ted Russell (Georgia Tech) talked about PM modeling. Simulation models for PM have a similar scientific foundation as ozone models, and the trend is toward "one atmosphere" models, but PM modeling is about 10 years behind ozone modeling. One needs to look at a longer period of time, more episodes to get a more complete picture of PM. Use of the models outside California is exposing problems in models that worked well in CA.

Russell said that sulfate modeling is pretty good, though cloud processes are difficult. The models are not working as well for nitrates, organics, and soil.

Emissions are the single biggest uncertainty in the models. Ammonia, organic carbon, elemental carbon, VOC (both anthropogenic and biogenic), and NO_x emissions need improvement. CO and SO2 inventories seem to be in pretty good shape.

More monitoring data is needed to help evaluate and improve PM modeling. He believes using specific organic carbons as tracers may help.

Russell's work has shown that the "bounce back" in nitrate concentrations when sulfates are reduced (due to the freeing up of ammonia which then combines to form nitrates) is relatively insignificant. Wintertime NO_x reductions will help reduce PM. In-state and neighboring-state SO2 reductions will have the greatest impacts on PM. He said some research suggests that reducing black carbon can have a big impact on climate.

John Watson (Desert Research Institute) commented that receptor models (source apportionment using air quality data) and source models (calculating concentrations based on emissions) are complementary, and both should be based on physical reality.

He explained some of the ideas in using receptor models. The first step is to review the constituents measured and get a general idea of the types of sources that may cause these concentrations. Obtain available source emissions profiles. Look at trends in concentrations and emissions. Use more than one model.

He advocated more testing of ambient air and source emissions using comparable methods for organic tracers. Greater time resolution helps show precise source impacts. He advocated changing the method for calculating source emissions to handle condensable emissions better by using a dilution method.

Spyros Pandis (Carnegie Mellon U) talked about what has been learned from the analysis of the Pittsburgh supersite data and related modeling. He explained that using some simple models with ambient data can teach us a lot.

In summary, Pandis concluded there is a large amount of water included in $PM_{2.5}$ as measured by the FRM. There is a large regional contribution to both sulfates and organic PM. Nitrate substitution does exist. Sources of organics include primary biogenic OC, transport, and biomass burning. Frequent nucleation events were observed, when a very large number of particles are formed. Semi-continuous monitoring methods can be used to "fingerprint" sources. Models are improving.

Pandis noted that there are many days with low levels of PM. On high days in the summer, sulfates dominate; in the winter, sulfates are still present, but nitrates also increase. In the summer, the FRM-measured $PM_{2.5}$ is greater than the sum of measured constituents due to the hygroscopicity of sulfate. In winter, the FRM-measured $PM_{2.5}$ is less than the sum of measured constituents due to the evaporation of nitrate off the filter.

Sulfate can spike very high for short periods of time as weather fronts pass through—going from 20 μg/m3 to 60 μg/m3 in 2 hours, e.g.

The formation of nitrate requires nitric acid and free ammonia. The reaction is favored with low temperatures and relatively high humidity. It would help to measure nitric acid vapor and ammonia gas to improve modeling.

If sulfate is reduced, nitrate will increase, but the overall total $PM_{2.5}$ will decrease. Controlling ammonia can reduce the nitrate increase. NO_x controls will help reduce nitrate in winter, but won't have much effect in summer.

Pandis was able to partition OC into primary and secondary using the OC/EC ratio and some indicator species. He calculates that 30-40% of summertime sulfate is secondary, 20% in the fall, and 10% in the winter. He believes 80 to 90% of OC was imported—the particles had been in the air for a day or two. EC was about half local and half transport.

His models worked fairly well for total $PM_{2.5}$ and for sulfate, and reasonably well for nitrate. It was more difficult to model periods of high secondary PM.

Control Strategies

Axel Friedrich (Germany) discussed PM controls in Europe. He stressed the importance of retrofitting filters on diesel vehicles, saying that this could save tens of thousands of lives per year. After years of effort, Germany has instituted a high tax for high emitting vehicles. Each European country has a NO_x cap, and meeting these caps will require vehicle NO_x control. He stated that a PM filter and SCR combination is feasible for trucks and will reduce NO_x. He noted that easing congestion just increases VMT.

Praveen Amar (NESCAUM) provided an overview of emissions controls. He noted that it will be relatively inexpensive to extend summertime NO_x controls to be year-round. He commented that the cost of acid rain controls has been far less than anticipated, and that more SO_2 control is certainly feasible. He recommended a book entitled Coal—A Human History. He explained that mercury controls on power plants involve injecting carbon and then controlling emissions with an ESP or baghouse. He believes controlling SO_2 and NO_x may have an indirect effect of helping to reduce the formation of secondary organic carbon, because the acidic aerosols may speed the formation of secondary organics.

Tom Grahame (DOE) commented that we have to stop using natural gas for power plants. The increased demand is driving up natural gas prices, which is costing jobs in businesses that use natural gas. He commented that marginal coal plants may go out of business if they have to put on controls. John Bachmann responded that EPA's

analysis does not predict shutting down plants at the levels required in the Clear Skies Initiative. He noted that if CO_2 reductions were required, then that would be different.

Steve Cadle (GM) asked us to keep in mind that non-road mobile sources are an important part of the picture for PM. This class of sources is very complex, including many types of vehicles with many emissions standards, and there is little data. In the national inventory, non-road sources emit 4% of $PM_{2.5}$ and 19% of NO_x. Percentages are greater in California.

Doug Lawson (National Renewable Energy Laboratory) discussed a project underway to combine source and ambient testing to determine whether it is possible to attribute ambient concentrations to gasoline vs. diesel engines. If the project is successful in California, it will be expanded to other parts of the country.

In response to a comment, Bob Slott (MIT) mentioned Virginia's remote sensing study of on-road vehicle emissions to determine whether vehicles in I/M areas were cleaner than other vehicles. He noted that the results did show benefits from I/M, though perhaps less than EPA's models would predict.

Wayne Miller (UC Riverside) described efforts to use a mobile lab to develop information to calculate emissions from heavy-duty diesel vehicles. (In informal discussion, he indicated that once his initial work is finished, he would like MARAMA's help in asking EPA to use his data to update their emissions factors.) An audience member commented that on-road testing cannot replace dynamometer testing.

Tim Johnson (Corning) explained that technology keeps evolving to avoid the need for filters on engines, though Corning has done a lot of work to design new filters. He commented that we are not going to make a real dent in diesel emissions without retrofits.

Panel on PM Control Policy

In the final session, seven people addressed the question of what they would do to control PM "if they were King."

Alan Lloyd (CARB) suggested

- Get sulfur in fuel as low as possible
- Focus on off- as well as on-road vehicles
- Continue research, but don't delay controls
- Reduce tailpipe emissions
- Address marine emissions.
- Reduce truck idling and accelerate computer re-programming
- Look at hydrogen options and other technology options

Tom Grahame (DOT)

- Control wood smoke and other sources of black carbon, including uncontrolled coal burning.
- Do a better emissions inventory.
- Conduct toxicological testing to determine what part of PM is harmful.
- Establish central city vehicle-free zones.

Alan Schaefer (Diesel Technology Forum

- Reduce diesel emissions through incentive grants using CMAQ funds.
- Establish in-use testing for diesel vehicles—create a mindset of shared responsibility for cleaner air.
- Reduce vehicle idling.
- Figure out what it will take to get old, dirty diesels off the road.

Vickie Patton (Environmental Defense)

- Tighten off-road standards.
- Control locomotives, commercial marine vessels, and ocean-going vessels.

- Regulate stationary diesel engines.
- Expand diesel retrofit programs.
- Adopt a transport rule that will be effective before the 2007 SIPs are due that will call for year-round NO_x control in the SIP Call region.
- Conduct research on $PM_{2.5}$ health effects, sulfate-nitrate replacement, light duty diesel risks.
- Conduct long-term monitoring and assessment.

Ken Colburn (NESCAUM)

- Get a better understanding of health effects.
- Revise the annual and daily $PM_{2.5}$ standards, and add a 3-hr standard.
- Consider regional emissions standards.
- Continue to emphasize supersites, continuous monitoring, ultrafines, etc.
- Retrofit diesels.
- Lock in low S fuels.
- Baghouses & flue gas desulfurization systems on all coal fired power plants.
- Improve ammonia controls at CAFOs and other major sources.
- Develop alternative energy sources and strengthen CAFÉ standards.

John Shanahan (Republican staff, U.S. Senate Committee on Environment and Public Works)

- Do more cost benefit analysis.
- Compare risks and consider interactions
- Improve the emissions inventory.
- Support Clear Skies.

Chris Miller (Democratic staff, U.S. Senate Committee on Environment and Public Works)

- Adopt a transport rule for SO_x.
- Find the most cost-effective way to reach the standards.
- Establish a comprehensive public health tracking system.
- Reduce reliance on conventional fossil fuel combustion.
- Expedite implementation of MACT standards.
- Seek opportunities to reduce PM through energy and transportation legislation.