The DOE/NREL Environmental Science Program

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ABSTRACT

This paper summarizes the several of the studies in the Environmental Science Program being sponsored by DOE's Office of Heavy Vehicle Technologies (OHVT) through the National Renewable Energy Laboratory (NREL). The goal of the Environmental Science Program is to understand atmospheric impacts and potential health effects that may be caused by the use of petroleum-based fuels and alternative transportation fuels from mobile sources. The Program is regulatory-driven, and focuses on ozone, airborne particles, visibility and regional haze, air toxics, and health effects of air pollutants. Each project in the Program is designed to address policy-relevant objectives. Current projects in the Environmental Science Program have four areas of focus: improving technology for emissions measurements; vehicle emissions measurements; emission inventory development/improvement; ambient impacts, including health effects.

INTRODUCTION

Increasingly stringent air pollution regulations regarding new tailpipe certification and fuel standards from the U.S. Environmental Protection Agency (EPA) provide major challenges for light- and heavy-duty vehicle manufacturers and fuel producers. In addition, the EPA recently introduced more stringent regulations regarding ambient ozone and PM$_{2.5}$ concentrations. In 1999, EPA also enacted its Regional Haze Rule, designed to improve visual air quality in national parks and wilderness areas. This regulation calls for states and federal agencies to improve visibility in 156 different areas, including the Grand Canyon, Yosemite, and the Great Smoky Mountains. States are required to provide a regulatory pathway that would take the national parks and wilderness areas (called "Class I" areas) from current conditions to "natural" or background conditions by 2064. Class I areas include large national parks and wilderness areas in existence in August 1977.

As a result of the 1990 Amendments to the Federal Clean Air Act, EPA is also required to regulate the emissions of 188 pollutants called air toxics. Currently, EPA is completing its National Scale Air Toxics Assessment that will focus on 33 air toxics that are thought to be of most importance in urban areas. The Assessment will also include diesel particulate matter, which is a component of diesel exhaust. In addition, the California Air Resources Board (CARB) recently declared diesel particulate matter a toxic air contaminant.

RESEARCH EFFORTS

DIESEL AEROSOL SAMPLING METHODOLOGY STUDY - One of the largest DOE/OHVT research programs currently underway is the $3 million CRC Project E-43, titled "Diesel Aerosol Sampling Methodology." The program started in September 1998. Funding has been provided by OHVT through NREL, CRC, Alliance of Automobile Manufacturers, Engine Manufacturers Association, the South Coast Air Quality Management District, and the California Air Resources Board. Sixty percent of the project support has been provided by the OHVT through NREL. In addition, $435,000 of in-kind contributions are being made by Cummins and Caterpillar. The project investigators are the University of Minnesota, West Virginia University, Carnegie Mellon University, UC Davis, Desert Research Institute, Tampere University (Finland), and the Paul Scherrer Institute (Switzerland).

The project objectives are:
- Determine actual particle size distributions and particle number concentrations in
exhaust plumes from on-road heavy-duty vehicles
• Compare on-road data with data generated in dynamometer testing facilities
• Determine the zone of influence of ultrafine particle emissions from a roadway
• Characterize chemical composition and surface properties of bulk PM emissions

This project is to be completed in the fall of 2001. The motivation for this study is to develop sampling methodologies to measure and examine nanoparticle emissions (particles with an aerodynamic diameter less than 50 nanometers). State-of-the-art particle sizing instruments have been employed during the on-road and dynamometer studies. Included among the samplers are the Multi-Orifice Uniform Deposit Impactor (MOUDI), nano-MOUDI, Electrical Low Pressure Impactor (ELPI), Electrical Aerosol Analyzer (EAA), Scanning Mobile Particle Sampler (SMPS), and nano-SMPS. The first three samplers perform particle sizing using aerodynamic properties of aerosols; the latter three samplers size particles using electrical mobility properties of sampled particles. Particles having diameters less than 10 nanometers have been measured in this project. In addition to characterizing particle size distributions, particle number distributions have been measured.

GASOLINE VEHICLE EXHAUST PARTICLE STUDY - Using particle sampling techniques developed during the Diesel Aerosol Sampling Methodology Study above, a team has been assembled by the DOE/OHVT Environmental Science Program to measure particles from a variety of gasoline-powered vehicles under a variety of conditions. This project has three, and possibly, four objectives:
• to measure representative, on-road particle size distributions and particle number concentrations from motor vehicles, distinguishing between gasoline and diesel exhaust. This objective will sample an on-road fleet to report differences in particle size and number distributions of exhaust from gasoline and diesel vehicles;
• to compare that information with data acquired from a small set of light-duty gasoline vehicles tested on a chassis dynamometer with a dilution tunnel using the Unified Cycle, at both room temperature and 30 °F;
• to characterize the bulk and size-segregated chemical composition of the PM emitted in exhaust from gasoline vehicles; and
• if results from the first two objectives do not agree with regard to PM size and number distributions, utilize the latest information on proper test cell dilution procedures to see if the agreement can be improved.

The study investigators are the University of Minnesota, University of Wisconsin-Madison, Ricardo Engineering, and Mercedes-Benz. This two-year project began in December 2000.

WEEKEND OZONE STUDY - Since the mid-1970s it has been shown that ozone ($O_3$) levels in California are higher on weekends than on weekdays, in spite of the fact that ozone pollutant precursors are lower on weekends than on weekdays (Levitt and Chock, 1976; Altshuler et al. 1995). Figure 1 shows the number of monitoring station hours in the South Coast (Los Angeles) Air Basin (SoCAB) exceeding the one-hour standard on weekends and weekdays between 1993 and 1996. The “weekend effect” has also been observed in the northeastern cities of Washington D.C., Philadelphia, and New York (SAIC, 1997).

Under subcontract to NREL, the Desert Research Institute (DRI) and Sonoma Technology, Inc. (STI) are conducting a study of the causes of elevated ozone levels on weekends in the South Coast (Los Angeles) Air Basin (SoCAB). The work is being conducted in three phases over a period of 30 months, and is to be completed in February 2002. In Phase I, a retrospective analysis of existing ambient air quality and meteorological data for the South Coast Air Basin was performed to improve the current conceptual understanding of the physical and chemical processes that drive the “weekend effect” and to refine and provide the hypotheses that will be addressed by this study (Fujita et al. 2000). Phase II, a field measurement program, was conducted in late summer-early fall 2000 to collect and assemble air quality and emission activity databases to test the hypotheses. Data analyses during Phase III will include temporal and spatial variations in ozone, ozone precursors, and emission source indicators [e.g., carbon monoxide (CO), nitric oxide (NO), “elemental” and “organic” carbon (EC and OC), total and speciated non-methane hydrocarbons (NMHC)], time-resolved hydrocarbon source apportionment, analysis of emissions activity
data, evaluation of the weekend effect by semi-empirical methods and review of air quality model simulations of Southern California Ozone Study (SCOS97-NARSTO) episodes. The results of these analyses will be reconciled with the current understanding of the weekend ozone effect in the SoCAB.

Phase I of the study was coordinated by NREL with DOE/OHVT support. Study Phases II and III are supported by NREL with some funding from the Coordinating Research Council. Phase I was being conducted in coordination with the Weekend Effect Workgroup sponsored by the California Air Resources Board (ARB). Information on the overall project and individual study results are available at http://www.arb.ca.gov/aqd/weekendeffect/weekendeffect.htm.

AIR TOXICS MODELING STUDY - A variety of approaches have been used, or are planned, for air toxic modeling to support a number of emerging air toxic programs focusing on various emission sources with various spatial scales ranging from the local scale up to regional and national scales. Challenges in addressing the needs of regulators from local through national scales combined with limited resources and lack of a cohesive plan have resulted in a default situation wherein various existing modeling tools are borrowed and utilized to address the issue, often in combination with one another and often containing ad hoc modifications. Examples of this approach are contained in EPA’s draft guidance on modeling urban air toxics, in recent EPA releases on modeling for National Air Toxics Assessment (NATA), in modeling for the mobile source assessment under the Clean Air ACT 202(l), and in local agency efforts like MATES-II in the South Coast Air Basin.

Both uncertainties and sensitivities to model formulations and model input assumptions, have essentially been overlooked in most cases. Some newer concepts like micro-environment models have been developed to improve realism, however these improvements are still in their infancy and need further development. Default values in some instances lack appropriate justification and documentation, thus realism in model results is even more uncertain.

Selective culling of limited monitoring data may be creatively called upon to imply consistency with modeling results, at times even going so far as to be considered “validation”. Clearly, before such circumstantial information can be of any scientific use a protocol is needed to address numerous issues including monitoring methods, calibration, detection limits, statistical use of non-detects, duration of data record, consistency among monitors, etc.

The objectives of this project, co-funded by DOE/OHVT and CRC, are:

- to establish the current level of scientific rigor that regulatory agencies are considering for air toxics modeling
- to identify key issues still to be addressed
- to identify near-term opportunities for improvements in modeling primary and relatively stable emissions of two suspected risk-driving chemicals, namely benzene and diesel particulate matter, including modeling of micro-environments
- to identify longer-term opportunities for improvements in modeling highly reactive emissions like 1,3-butadiene and formaldehyde, and for secondary air toxic chemicals like formaldehyde and acetaldehyde; consider opportunities for creating a more seamless approach for modeling the spectrum of air toxic chemicals; consider the implications of using monitoring data to complement modeling (i.e., when is it appropriate to use and how to use it)
- to demonstrate alternative approaches to the current modeling for air toxics (i.e., multiple approaches because of multiple programs worked by multiple agencies)
- to provide examples to immediately improve modeling for two risk-driving chemicals, namely benzene and diesel particulate matter.

Two subcontractors, AER and ENVIRON, are conducting this study for NREL and CRC. The project will be completed during the first half of 2002. This project will advance state-of-the-science modeling for air toxics.

GASOLINE/DIESEL PM SPLIT STUDY - The 1996-1997 Northern Front Range Air Quality Study, NFRAQS http://www.nfraqs.colostate.edu suggests that there are major discrepancies between source apportionments derived from ambient observations and current government emission inventories. Emission inventories provide a frame of reference for development of air quality management strategies, because they
estimate emissions from different sources. Thus, the inventories must be accurate so policy makers can design air quality management strategies that reduce air pollution in the most cost-effective manner.

One of the NFRAQS policy-relevant objectives was to identify the sources of directly emitted PM$_{2.5}$ (airborne particles less than 2.5 micrometers in diameter). The NFRAQS was designed to provide information to policy makers in Colorado who are responsible for managing air quality.

The NFRAQS results suggest that during the winter episodes of highest PM$_{2.5}$ concentrations in the metro Denver area, receptor modeling estimated that the most important sources or contributors to PM$_{2.5}$ were:

- Gasoline exhaust, 30%
- Diesel exhaust, 10%
- Dust and debris, 15%
- Wood smoke, 5%
- Meat cooking, 5%
- Particulate ammonium nitrate (formed in the atmosphere from a variety of sources), 25%
- Particulate ammonium sulfate (formed in the atmosphere from a variety of sources), 10%

During the episodes studied, the direct PM$_{2.5}$ contribution from gasoline-powered vehicles and engines was three times the direct PM$_{2.5}$ contribution from diesel-powered vehicles and engines. In contrast, in current emission inventories suggest that diesel vehicles produce more PM$_{2.5}$ emissions than gasoline-powered vehicles. The diesel exhaust particles come from trucks, locomotives, construction equipment and other sources. High-emitting or smoking gasoline-powered vehicles, which comprise a small fraction of the in-use vehicle fleet, produced nearly one-half of the gasoline exhaust particles. Fine particles from road debris and dust, construction activities, and wind-blown sand contributed 16% of the total PM$_{2.5}$, an amount much lower than current emission estimates.

Particulate ammonium nitrate and ammonium sulfate are formed in the atmosphere from gas-phase emissions of ammonia, nitrogen oxides, and sulfur dioxide. These are called secondary particles because they are not emitted directly. Because the NFRAQS program results suggest that PM$_{2.5}$ emissions from spark ignition vehicles are more important than PM$_{2.5}$ emissions from diesel vehicles in Denver in the winter, the OHVT/NREL Environmental Science Program will support a field study to confirm the NFRAQS results in the South Coast Air Basin during the summer of 2001. Participants include the University of West Virginia, University of Wisconsin-Madison, Desert Research Institute, EPA, and the California Bureau of Automotive Repair.

COMPARATIVE TOXICITY STUDY - In this study, the relative toxicities of exhaust from spark-ignition and diesel vehicles will be evaluated. In 1999, the California Air Resources Board declared diesel exhaust as a toxic air contaminant, but there has been no work by the regulatory agencies to assess the relative toxicity of exhaust from these two engine technologies.

The study subcontractors are Southwest Research Institute (SwRI), Desert Research Institute (DRI), University of Dayton Research Institute, National Institute of Occupational Health and Safety (NIOSH), Oak Ridge National Laboratory, and Lovelace Respiratory Research Institute (LRRI). There are three separate phases in this program. The collection phases are dynamometer-based collection of up to four grams of PM$_{2.5}$ and associated semi-volatile organic compound (SVOC) exhaust samples from a series of different spark-ignition and diesel vehicles (performed at SwRI) and collection of PM$_{2.5}$ and SVOC samples at the Fort McHenry (Baltimore) tunnel (performed by DRI). Morphological analyses of the collected PM$_{2.5}$ samples have been performed by Oak Ridge National Laboratories.

The dynamometer sampling collected up to four-gram PM$_{2.5}$ and SVOC samples from the following at ~72 °F: average gasoline emitters, high PM$_{2.5}$ gasoline emitters, smoking gasoline vehicles, current technology diesel vehicles, and high PM$_{2.5}$ diesel emitters. Dynamometer testing at ~30 °F was carried out to obtain PM$_{2.5}$ and SVOC exhaust emission samples at cold temperature (Table 1).

The tunnel vehicle exhaust study, performed at the Fort McHenry (Baltimore) tunnel in September 1999 by DRI, collected from both the light-duty and heavy-duty vehicle bores of the tunnel. The advantage of tunnel study testing is
that exhaust emissions from many vehicles can be collected in a real-world setting. The goal of the tunnel sampling was to collect four-gram samples for toxicity testing.

Once the dynamometer and tunnel samples were collected, they were sent to DRI for detailed chemical analysis. Composited fuel and lube oil samples from the vehicles tested on the dynamometer were analyzed by SwRI and DRI using routine and detailed chemical analysis. Detailed multi-dimensional gas chromatography/mass spectrometry analyses were performed on a limited number of samples at the University of Dayton Research Institute. The collected PM$_{2.5}$ and SVOC samples were extracted at DRI and sent blind to NIOSH and LRRI for subsequent comparative toxicity testing.

The comparative toxicity testing at LRRI will consist of three components: in vitro (cell culture) testing, in vivo (animal) testing; and Ames testing. The in vitro testing will use the A549 human lung epithelial cell line. Responses to these cells to exhaust samples in vitro may reflect responses of lung cells of humans. For the in vivo testing, rats will be instilled intratracheally with exhaust sample. Groups of five rats per does will be sacrificed at several times ranging from immediately to four weeks after instillation; lung lavage fluid, cells, and lung tissue will be analyzed for a variety of parameters such as cell counts and proteins. The standard mutagenicity assay using the Ames test will be performed by NIOSH. Each of these three testing protocols will be followed for the spark-ignition and diesel exhaust PM$_{2.5}$ and SVOC samples, and for the first time, comparative toxicity results will be made available. Results from this project should be available during the latter part of 2001.

CONCLUSION

The Environmental Science Program at the National Renewable Energy Laboratory is funded through DOE’s Office of Heavy Vehicle Technologies. The Program focuses on policy-relevant research issues that can be used in the regulatory setting. Results obtained from these programs will provide a sound scientific basis for regulation as is found necessary.

REFERENCES


CONTACTS

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Figure 1. South Coast Air Basin station-hours on weekends and weekdays above the one-hour ozone standard, 1993-1996.

Table 1. Chassis dynamometer matrix of vehicle tests

<table>
<thead>
<tr>
<th>Sample Description</th>
<th>Temperature</th>
<th>Number of Vehicles</th>
<th>Samples Taken</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gasoline average PM emitter</td>
<td>Room</td>
<td>5</td>
<td>PM &amp; SVOC</td>
</tr>
<tr>
<td>Gasoline black smoker</td>
<td>Room</td>
<td>1</td>
<td>PM &amp; SVOC</td>
</tr>
<tr>
<td>Gasoline white smoker</td>
<td>Room</td>
<td>1</td>
<td>PM &amp; SVOC</td>
</tr>
<tr>
<td>Diesel average PM emitter</td>
<td>Room</td>
<td>3</td>
<td>PM &amp; SVOC</td>
</tr>
<tr>
<td>Diesel high PM emitter</td>
<td>Room</td>
<td>1</td>
<td>PM &amp; SVOC</td>
</tr>
<tr>
<td>Gasoline average PM emitter at 30°F</td>
<td>30°F</td>
<td>5</td>
<td>PM &amp; SVOC</td>
</tr>
<tr>
<td>Diesel average PM emitter at 30°F</td>
<td>30°F</td>
<td>3</td>
<td>PM &amp; SVOC</td>
</tr>
<tr>
<td>Gasoline average PM emitter</td>
<td>Room</td>
<td>5</td>
<td>SVOC (PM)</td>
</tr>
<tr>
<td>Diesel average PM emitter</td>
<td>Room</td>
<td>3</td>
<td>SVOC (PM)</td>
</tr>
<tr>
<td>Gasoline ULEV (1999 Honda Accord)</td>
<td>Room</td>
<td>1</td>
<td>PM &amp; SVOC</td>
</tr>
<tr>
<td>Gasoline NLEV (1999 Ford Windstar)</td>
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<td>European light-duty diesel (1999 Mercedes C220)</td>
<td>Room</td>
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