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ABSTRACT

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

INTRODUCTION

Increasingly stringent air pollution regulations regarding new tailpipe certification standards and National Ambient Air Quality Standards (NAAQS) provide serious challenges for light- and heavy-duty vehicle manufacturers. For example, EPA recently introduced more stringent regulations regarding ambient ozone and $PM_{2.5}$ concentrations. However, these regulations have recently been questioned in court decisions. EPA also recently enacted its Regional Haze Rule, whose goal is to have air quality approaching background conditions in Class I areas in the U.S. by 2064. These Class I areas include large national parks and wilderness areas in existence in August 1977.

At the same time, recent work completed as part of the Northern Front Range Air Quality Study (NFRAQS, http://nfraqs.cira.colostate.edu) suggests that there are major discrepancies between source apportionments derived from ambient observations and current government emission inventories. Emission inventories often provide a frame of reference for development of air quality management strategies because they estimate emissions from different sources. Thus, the inventory must be accurate so policy makers can plan effective programs to reduce emissions.

The NFRAQS was designed to provide information to policy makers in Colorado who are responsible for managing air quality. 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 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 $PM_{2.5}$, as shown in Figure 1, were:

- Gasoline vehicle exhaust, 28%
- Diesel exhaust (from all diesels), 10%
- Dust and debris, 16%
- Wood smoke, 5%
- Meat cooking, 4%
- 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-fueled vehicles and engines was three times the direct $PM_{2.5}$ contribution from diesel-fueled vehicles and engines. In contrast, in current emission inventories diesel vehicles are projected to produce more $PM_{2.5}$ emissions than gasoline-powered vehicles. High emitting or smoking gasoline-fueled vehicles, which comprise a small fraction of the in-use vehicle fleet, produced nearly half of the gasoline exhaust particles. The diesel

exhaust particles come from trucks, locomotives, construction equipment and other sources. $PM_{2.5}$ directly emitted from diesel vehicles and engines was one-third of that from gasoline vehicles and engines, even though diesel-powered vehicles comprise five percent of the regional vehicle miles traveled. 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 (Figure 1).

The OHVT/NREL ES&HE program is working to verify the NFRAQS results and to continue policy-relevant research regarding mobile source emissions.

COOPERATIVE RESEARCH EFFORTS

DOE's Office of Heavy Vehicle Technologies is aware of the research experience available within other organizations, such as the U.S. Environmental Protection Agency (EPA) and the Coordinating Research Council (CRC). The OHVT/NREL ES&HE program has been designed in cooperation with EPA and CRC. The CRC is funded equally by the American Petroleum Institute and the Alliance of Automobile Manufacturers and has been sponsoring vehicle emissions research since 1957. CRC-sponsored research is publicly available and is used by industry to ensure optimum compatibility and customer satisfaction, and by industry, government, and the public to enhance joint achievement of clean air and other applicable goals. Regular meetings and communication also are taking place between EPA's Office of Mobile Sources and NREL so that areas of mutual research needs can be identified and duplication of effort avoided.

One of the most significant cooperative research programs currently underway is the $1.7 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, South Coast Air Quality Management District, and 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 principal investigator is the University of Minnesota, with subcontracts with West Virginia University, Carnegie Mellon University, Desert Research Institute, Paul Scherrer Institute (Switzerland), and Tampere University (Finland).

The CRC E-43 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

The E-43 project is to be completed in the first part of 2001. The motivation for this study is to develop sampling methods to measure and examine nanoparticle emissions (particles with an aerodynamic diameter less than 50 nanometers). State-of-the-art particle sizing instruments will be used 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 sizing; the latter three samplers size particles using electrical mobility properties of sampled particles. Particles having diameters less than 10 nanometers will be measured in this project. In addition to characterizing particle size distributions, particle number distributions will also be measured.

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 wintertime, NREL plans to augment funding to the CRC E-43 project to study particle size emissions from spark ignition vehicles. This study will begin during the first part of 2000.

A second collaborative program between the ES&HE Program and CRC and possibly other sponsors is a study designed to investigate and understand the causes for higher ozone observations on weekends than on weekdays in the South Coast (Los Angeles) Air Basin. On average, ground-level ozone is about 15 percent higher on weekends than on weekdays in Los Angeles, despite lower hydrocarbon and nitrogen oxide precursor emissions. Also, heavy-vehicle activity on weekends is about half that observed during weekdays in Los Angeles. This phenomenon is observed in other California locations and in other locations in the U.S. Understanding why the atmosphere responds to this change in emissions has important implications for air quality management strategies. NREL issued a Request for Proposals for this project in May 1999, and after selection of contractors, it is anticipated that the project will begin in late 1999 or early 2000.

INDEPENDENT ES&HE PROJECTS

The ES&HE Program is currently funding a number of separate studies, three of which are described in this section.

The first project is titled "Review of Air Quality Impacts from Diesel and Gasoline-Powered Vehicles" by Battelle Memorial Institute. The objective of this program has been to provide a review of fine particle source apportionment studies conducted in the South Coast Air Basin and in Denver (the NFRAQS). Both areas have been the subject of major $PM_{2.5}$ studies in recent years, but the study results from the areas have given different results. The Denver results suggest that spark-ignition $PM_{2.5}$ emissions are a more important contributor to the area's $PM_{2.5}$ than $PM_{2.5}$ emissions from diesel vehicles, while the Los Angeles source apportionment studies have shown the opposite. The approach taken by Battelle has been to review all published reports from the two regions, along with interviews with key researchers from those studies and air pollution control agency staff members from Colorado and California. Results from this project will be available in late 1999.

A second project, conducted at the University of Dayton Research Institute (UDRI), has the objective of identifying volatile organic compound and PM emissions from alternative fuels and reformulated fuels. The UDRI study is a fundamental research project using a flow tube reactor where various fuels are oxidized and the partial oxidation products are identified in an on-line analytical system. The most recent research by UDRI is focusing on PM emissions produced by various fuel types.

The third program is a comparative toxicity study, where the relative toxicities of exhaust from spark-ignition and diesel vehicles will be evaluated. In 1999 the California Air Resources Board declared diesel exhaust 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 types.

DOE's OHVT is providing overall direction to the program, with day-to-day activities being directed by NREL. The study subcontractors are Southwest Research Institute (SwRI), Desert Research Institute (DRI), 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).

The dynamometer samples will collect 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 will be carried out to obtain $PM_{2.5}$ and SVOC exhaust emission samples at low temperature.

The tunnel samples, to be collected at the Fort McHenry tunnel in September 1999 by DRI, will be 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 is to collect four-gram samples for toxicity testing.

Once the dynamometer and tunnel samples have been collected, they will be sent to DRI for detailed chemical analysis. Composited fuel and lube oil samples from the vehicles tested on the dynamometer will be analyzed by SwRI and DRI using routine and detailed chemical analysis. The collected $PM_{2.5}$ and SVOC samples will be extracted at DRI and sent blind to 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 dose 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 sent to an outside laboratory for analysis. 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 2000.

SUMMARY

The Environmental Science and Health Effects Program at the National Renewable Energy Laboratory is funded through DOE's Office of Heavy Vehicle Technologies. The ES&HE Program is focusing on policy-relevant research issues that can be used in the regulatory setting. In addition to independent research studies, NREL continues to seek avenues for collaborative research with other interested groups. It is hoped that results obtained from these programs can provide a good scientific basis for additional regulation as necessary.

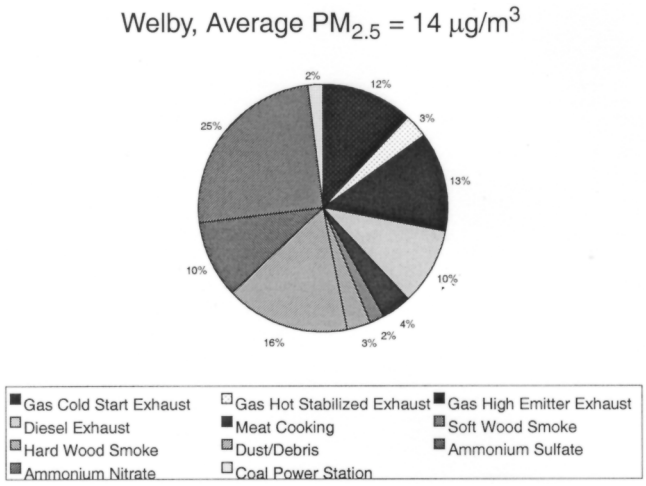

Figure 1. Average source and chemical contributions to the 24-hour average $PM_{2.5}$ concentration at Welby during the Winter 1997 NFRAQS episode periods, using receptor modeling with detailed speciation. Sources of ammonium nitrate and ammonium sulfate were not identified. Average concentrations during the entire winter season are lower than those shown. The day-to-day variability in apportionments is 15-30% with the exception of wood burning, which has greater variation due to burning restrictions. The uncertainty in the apportionments for any single sampling period is about 15-30%.