Abstracts

SESSION 10 – Environmental Science and Health Impacts

Chair: **James Eberhardt, U.S. Department of Energy**

History of the FreedomCAR Environmental Science & Health Impacts R&D Activity

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The Alternative Motor Fuels Act (AMFA) sought to lessen the dependence of the U.S. transport sector on imported oil by encouraging the use of other fuels including gaseous fuels, especially those that can be produced domestically, such as natural gas and ethanol from biomass. Less is known about the emission characteristics of alternative fueled vehicles compared to those from gasoline- and Diesel-fueled vehicles. In order to ensure that large scale utilization of these alternative fuels would not cause negative environmental and health impacts, the Department of Energy, Office of Transportation Technologies initiated shortly after the passage of AMFA a research program to investigate the "Atmospheric Reactions" which emissions from these fuels would undergo. In 1996 with the emphasis on "Dieselization" and the creation of the Office of Heavy Vehicle Technologies, work was directed at the investigation of the health impacts of Diesel particulate emissions. This work has sought to put the health impacts of Diesel emissions into context with emissions from other sources, especially automotive gasoline. Emissions from the latter often have been assumed to be benign due to the installation of the automotive catalytic converter in 1975.

Presently, the FreedomCAR and Vehicle Technologies (FCVT) Program conducts an Environmental Science and Health Impacts (ES&HI) activity that seeks to provide a rational basis for determining and comparing vehicle emissions, the relative contribution to the ecosystem of both petroleum and alternative fuel vehicle emissions, and their impact on human health. The aim is to provide a basis for minimizing the environmental and health impacts of vehicle emissions, especially emissions from new technologies being developed by FCVT so as to avoid unanticipated negative impacts from large-scale use of these fuels and vehicles.

A brief history of the DOE/FCVT ES&HI activity will be presented from the years following the passage of AMFA in 1988 to the present. The discussion will address the evolving nature of Diesel emissions, the focus of the DEER Conferences, in the context of motor vehicle emissions in general.

DOE's Gasoline/Diesel PM Split Study

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The Department of Energy's (DOE) Gasoline/Diesel particulate matter (PM) Split Study was conducted to quantify the relative contributions of tailpipe emissions from gasoline-powered motor vehicles and Diesel-powered motor vehicles to the ambient concentrations of fine particulate matter ($PM_{2.5}$) in the urbanized region of Southern California using an organic compound-based chemical mass balance model (CMB). This study involved several groups working cooperatively on sample collection and quality assurance aspects of the study, but working independently, at least initially, on chemical analysis and data analysis. Groups participating in the Study include California's Bureau of Automotive Repair, South Coast Air Quality Management District, U.S. Environmental Protection Agency, Ralph's Groceries, Clean Air Vehicle Technology Center, West Virginia University, the University of Wisconsin at Madison (UWM), and the Desert Research Institute (DRI). Source testing of 59 light-duty vehicles (including 2 Diesel vehicles) was completed in June 2001; ambient measurements were performed in July 2001, and the testing of 34 heavy-duty vehicles was completed in September 2001. DRI used sample collection and chemical analysis methods consistent with those employed during the 1996/97 Northern Front Range Air Quality Study. Parallel samples were collected by UWM using methods consistent with previous PM studies in Los Angeles. This paper describes the chemical compositions and apportionments of gasoline- and Diesel-powered vehicles that were obtained by DRI.

Twenty-four hour ambient samples were collected on Teflon and quartz filters and Teflon-impregnated glass fiber (TIGF) filters followed by polyurethane foam (PUF) plugs and XAD-4 resin cartridges for twenty-eight consecutive days at air monitoring stations in downtown Los Angeles and Azusa. Teflon filters were analyzed for gravimetric mass, elements, and ions, and quartz filters were analyzed for organic and elemental carbon by Thermal Optical Reflectance (TOR) and Thermal Optical Transmittance (TOT) using both IMPROVE and NIOSH protocols. The TIGF/PUF/XAD samples were combined and extracted together by day of week, and analyzed for polycyclic aromatic hydrocarbons, hopanes, steranes, alkanes, methoxyphenols, lactones, sterols, and polar organic compounds. A third set of ambient samples was collected from a mobile sampling van at several regionally representative sites and at locations with expected higher proportions of PM emissions from Diesel trucks (e.g., Terminal Island, truck stop, highway truck routes) and from gasoline vehicles (e.g., congested freeway during commuter rush hour, surface streets during weekends, a parking lot at major sporting events). Black carbon and total particulate matter were monitored continuously by photoacoustic and DustTrak instruments. As a prelude to the chemical mass balance analysis, we examined the variations in relative abundances of key marker compounds in the source-dominated ambient samples relative to corresponding variations in the samples from regional air quality monitoring sites by day of week. The ambient source apportionments are placed in context with previous ambient source apportionment studies and emission inventory trends. This work was supported by the DOE's Office of FreedomCAR & Vehicle Technologies through the National Renewable Energy Laboratory.

2007 Diesel Particle Measurement Research Project

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In 2007, on-highway heavy-duty Diesel engines will be required to meet a particulate matter (PM) emission level of 0.01 g/hp-hr, a 90 percent reduction from the current emission level. In addition, exhaust PM composition is expected to change because of new catalyzed exhaust traps/filters implemented to help meet the new standards. These traps/filters will stop most PM except a small mass of particles, mainly consisting of volatile and semi-volatile hydrocarbon and sulfate species derived from unburned and partially burned fuel and lubricating oil. This low level of remaining volatile PM mass poses a technical challenge for its accurate measurement using the 2007 prescribed filter-based sampling protocol. Not only is there concern for sampling a very low quantity of PM mass to be deposited on the sample filter, but also for several factors including filter handling, filter media, negative or positive artifact, sampling system conditioning, and control of the dilution parameters that can significantly affect quantifying PM mass emission.

Most concerns relative to PM measurement from future Diesel engines are to be addressed in Project E-66, sponsored by the Coordinating Research Council, Department of Energy/National Renewable Energy Laboratory, Engine Manufactures Association, Environmental Protection Agency, and the California Air Resource Board. The effect of filter media, filter face velocity and temperature on PM measurement will be investigated in Phase 1 of the project. This phase also includes comparing the performance of alternative real time particle measuring instruments to that of the filter-based method. Other phases will examine the effect of primary and secondary dilution parameters on PM measurement and the potential use of partial flow sampling systems as an alternative to constant volume sampling under transient engine testing.

The proposed presentation will cover the progress made during Phase 1 of Project E-66 with information on other phases of the project.

On-Road Exposure and Emission Measurements

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The University of Minnesota's mobile emission laboratory (MEL) has been used to characterize on-road particle exposures and to determine engine emission factors under real world conditions. Particles emitted by modern Diesel and spark ignition engines do not all form during combustion. Many particles, particularly those in the 3 to 30 nm diameter (nuclei mode) range, form from volatile materials as the exhaust dilutes and cools in the atmosphere. These particles may constitute 90 percent or more of the nanoparticle (< 50 nm) number emissions. Unfortunately, the formation of these particles is a nonlinear, gas-to-particle nucleation process that is extremely dependent upon dilution conditions. These conditions are difficult to simulate in the laboratory, especially in animal exposure facilities. Using the MEL as an exposure platform avoids this difficulty by using the actual on-road atmosphere to provide dilution. Two types of experiments have been performed. In the first, the MEL was driven on an urban and rural route and, to the extent possible, on-road plumes from Diesel powered heavy-duty trucks were used to provide the exposures. In the second, a new sampling system was installed on the MEL that made it possible to sample the MEL's diluted exhaust plume. A 2000 model year engine that provides a particle signature, which is characteristic of modern engines, powers the MEL. We describe the characteristics of the exposures achieved by the two test conditions along with the instrument array, calibration, particle losses, sampling artifacts, ambient conditions, traffic conditions, and sampling locations or routes. As an added benefit of this study, real world particle emission factors for heavy-duty Diesel engines have been determined. The instrument suite includes an Scanning Mobile Particle Sizer to size particles in 9 to 300 nm size range; a Ultrafine Condensation Particle Counter with a lower limit 50 percent counting efficiency at 3 nm; instruments to measure total submicron particle surface area; a Photoelectric Aerosol Sensor to measure aerosol photoemission response; carbon dioxide, carbon monoxide, and nitrogen oxides analyzers; and a thermal denuder to distinguish between solid and volatile particles.

Relationship Between Toxicity and Composition of Inhaled Diesel Exhaust

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Diesel engine emissions change in composition with variations in fuel, engine configuration, engine operation, and emission reduction technologies such as catalyzed particle traps. While previous studies have shown that Diesel exhaust can cause inflammation in rodents and humans and decreased resistance to respiratory infection in mice, there is little information on the impact of changes in exhaust composition (resulting from changes in fuel, engine operation, and after-treatment technology) on these effects. To address this, identical inflammation and respiratory infection assessments were conducted after exposure of mice to Diesel exhaust generated from a single cylinder Diesel engine generator under four steady-state conditions: 1) high-load operation, number 2 certification fuel (371 ppm sulfur, 29 percent aromatics), emissions diluted to 200 $\mu g/m^3$ particulate matter (PM); 2) high-load operation, certification fuel, non-catalyzed ceramic trap, same dilution rate as condition one; 3) low-load operation, certification fuel, diluted to 200 $\mu g/m^3$ PM; and 4) high-load operation, 15 ppm sulfur fuel, catalyzed ceramic trap, same dilution rate as condition one. Changing the operation (load) of the engine resulted in an approximately 4 times change in particle organic content. Lower load operation yielded increased vapor phase organics and PM primarily composed of condensates that were smaller in size than produced during high load. Resistance to infection was decreased and lung inflammation was increased when the non-volatile (black carbon) portion of the exhaust was highest (high load condition). All health effects were decreased with higher organic carbon (low load) and with the use of a catalyzed emissions trap/low sulfur fuel combination.

Assessment of Health Hazards of Repeated Inhalation of Diesel Emissions, with Comparisons to Other Source Emissions

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It continues to be difficult to place the potential health hazards of Diesel emissions into proper context in regard to hazards from competing technologies of other mobile and stationary source emissions and the potential health benefits of emission reductions. Among the factors contributing to the difficulties is the lack of information on: 1) exposure-response relationships for exposures down to environmental levels and 2) comparative hazards of Diesel and other source emissions. Identically-designed studies of the health effects of subchronic, repeated inhalation exposures to several source emissions are being conducted under the auspices of the National Environmental Respiratory Center program, with support from multiple federal, state, and corporate sponsors, including the Department of Energy and several engine manufacturers. Not only will the study of each source emission provide contemporary assessments of hazard, but the results also allow direct comparisons of hazard among sources and multivariate analyses of the combined data from all studies to determine the physical-chemical components associated with different health effects. Exposures to Diesel emissions and hardwood smoke are completed and most data have been analyzed. An exposure to gasoline emissions is underway, and exposures to urban street dust and coal emissions are planned.

Rodents of different species, strains, genders, and ages, specific to the different health outcomes, were exposed 6 hr/day, 7 days/wk for up to 6 months to clean air or emissions from 2000 Model Cummins ISB 5.9L engines burning certification fuel (350 ppm sulfur, 30 percent aromatics), using Shell Rotella-T 15-40w crankcase oil, connected to eddy current dynamometers, and operated on engine stands on continuously repeating heavy-duty certification test cycles. Whole exhaust was diluted to particle mass concentrations of 1000, 300, 100, and 30 $\mu g/m^3$. The physical-chemical composition of the exposure atmospheres were characterized in detail. Health evaluations included general indicators of toxicity, serum chemistry, hematology, bronchoalveolar lavage, electrocardiograms, development and exacerbation of respiratory allergic responses, resistance to respiratory infection with bacteria and virus, and indicators of cancer hazard including micronuclei in circulating cells, lung adenomas in A/J mice, and lung DNA methylation and oxidative injury. In an identically-designed study, rodents were exposed to smoke generated from burning oak in a non-certified heating stove over a 3-phase, 6-hr daily cycle. Results from the Diesel study will be reviewed, and the health effects of exposure to Diesel emissions and hardwood smoke will be compared.

The California Demonstration Program for Control of PM from Diesel Backup Generators (BUGs)

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Diesel backup generators (BUGs) are an important source of electricity in many critical areas, such as hospitals, municipal services, and bank clearing houses. Emission factors from uncontrolled BUGs are listed in the Environmental protection Agency's (EPA) AP-42 factors but actual in-use measurements indicate that the emissions are much less than the values in the EPA tables. Furthermore, no emission factors existed for controlled Diesel engines used in BUGs.

The report will present emission results from a number of BUGs ranging in size from 300 kW to 2,000 kW and for units built between 1985 and 2002. There were some surprising results. For example, emission factors did not always vary with size; some of the largest units had the lowest emission factors. For controlled BUGs, results show that the particulate matter (PM) emissions can be controlled to low levels with technology borrowed from the mobile source applications. Taken in total, the results indicate that the emissions from BUGs are lower than reported in AP-42 and that PM emissions can be controlled with various technologies so as to reduce the personal exposure to a toxic air contaminant.

Diesel Exhaust: Phsical, Chemical and Celluar Response Characterization

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Diesel exhaust particulates vary in their detailed composition and morphology depending upon engine conditions, oil additives, and other factors. The potential health effects should also depend on these parameters, but few approaches have the potential to link specific cause and effects in sufficient detail and speedily enough to guide optimization of engine technology. Insensitive detailed inline and offline analytical methods are used to probe how engine running conditions change the output particulates. Using well controlled cellular systems permitting rapid throughput, the biological responses to air-delivered particulates are identified be profiling the global genomic responses. Particle characterization was via Single Particle Laser Ablation Time of Flight Mass Spectrometer (SPLAT-ms), the Aerodyne Aerosol mass spectrometer, and automated particle scanning electron microscopy/elemental analysis. Preliminary results are presented, along with updates on cellular response mechanisms, on-cell nanoparticle imaging, and new instruments (SPLAT II).