II. ADVANCED COMBUSTION AND EMISSION CONTROL RESEARCH FOR HIGH-EFFICIENCY ENGINES

D. Health Impacts

II.D.1 Health Effects from Advanced Combustion and Fuel Technologies

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Objectives

- Understand potential impact of developing fuel, combustion, and aftertreatment technologies on air quality and, thereby, human health.
- Quantify mobile source air toxic emissions from advanced technologies.
- Link emissions measured in laboratory setting to air quality impact, and thereby, health impact.

Accomplishments

- Measured mobile source air toxics (MSATs) from European Saab Biopower vehicle that is optimized for performance with E85 fuel to provide information on the health impact of ethanol fuel introduction.
  - E85 increases cold-start ethanol emissions to levels of 48 mg/mile on the Federal Test Procedure (FTP) driving cycle.
  - E85 reduces gasoline-derived MSAT emissions by a factor of 10 as compared with gasoline fuel over the FTP and US06 driving cycles.
- Characterized mobile source air toxics from high efficiency clean combustion (HECC) on a diesel engine to determine any health impacts of advanced diesel combustion modes.
  - Formaldehyde emissions from HECC were a factor of two higher than the traditional diesel combustion emissions.
  - Acetaldehyde and acrolein emissions were also observed to increase significantly during HECC operation.

Future Directions

- Evaluate the impact of butanol fuel addition to ethanol-gasoline fuel mixtures on emissions.
- Determine the efficiency of catalytic aftertreatment for reduction of MSATs from HECC diesel operation.

Introduction

Scientists and engineers at national laboratories, universities, and industrial companies are developing energy efficient technologies for transportation, and the U.S. Department of Energy (DOE) is actively involved in this innovative research. However, care must be taken to insure that any new technology developed for transportation must not adversely directly impact human health or impact the health of the environment, which subsequently may impact human health. To address this need, DOE sponsors research studies on the potential health impacts of advanced technologies for transportation including advanced fuels, combustion techniques, and emissions controls (also known as “aftertreatment” or, more commonly, “catalytic converters”). DOE sponsored health impact research activities at Oak Ridge National Laboratory (ORNL) are presented in this report.

ORNL conducts research on advanced fuel, combustion and emissions control technologies at the National Transportation Research Center (a joint ORNL and University of Tennessee research center) in Knoxville, TN. As the research is being conducted on engine and chassis dynamometers, specific studies of the potential health impacts of the technologies are conducted by the ORNL team for DOE’s health impacts program. Results from two ORNL studies in FY 2007 are presented here. Both studies focused on measuring MSATs from advanced technologies; the technologies were (1) an ethanol fuel optimized vehicle and (2) an advanced diesel combustion technique that results in simultaneous reduction of NOx and particulate emissions called HECC.

Approach

MSATs from E85 Fueled Vehicle

A 2007 Saab 9-5 Biopower 2.0t vehicle was acquired by ORNL and DOE and evaluated for fuel efficiency performance on E85 and gasoline fuels [1]. The engine is optimized for performance with E85 fuel and in fact accelerates faster to 60 mph with E85 vs. gasoline fuel. CO emissions are lower with E85 fuel and the emissions overall are below U.S. EPA Tier 2, Bin 5 levels. MSAT

emissions were sampled and analyzed with a variety of techniques including Fourier transform infrared (FTIR) and gas and liquid chromatography via collection with 2,4-dinitorophenylhydorazine (DNPH) cartridges. The measurements were cycle averages for the FTP and US06 driving cycles; a photo of the vehicle on the chassis dynamometer is shown in Figure 1. In addition to MSAT emissions, ethanol, an unregulated species, was quantified.

MSATs from HECC Diesel Mode

A Mercedes 1.7-liter 4-cylinder diesel engine was operated at 1,500 rpm and 2.6 brake mean effective pressure (BMEP) on an engine dynamometer under production calibration conditions that are typical of traditional diesel combustion. The engine is shown in Figure 2. The load and speed chosen are representative of a universal moderate load condition for a light-duty vehicle. The engine was also operated at the same speed and load with the engine operating in HECC mode. The MSAT emissions from both the traditional and advanced HECC combustion modes were measured with FTIR and gas and liquid chromatography via collection with DNPH cartridges.

FIGURE 1. Saab 9-5 Biopower Vehicle on a Chassis Dynamometer

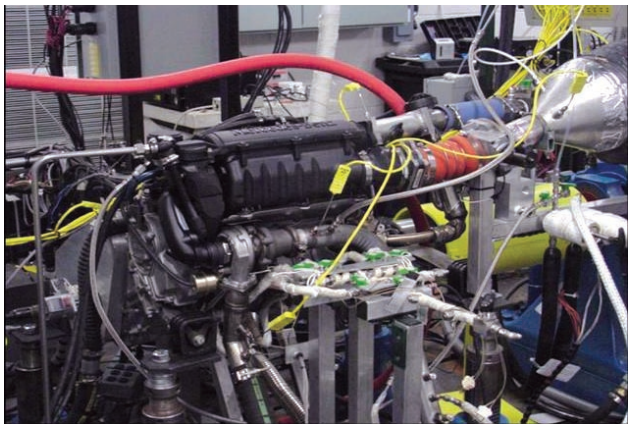

FIGURE 2. Diesel Engine Operated in HECC in ORNL's Engine Dynamometer Laboratory

Results

MSATs from E85 Fueled Vehicle

Ethanol emissions with E85 fuel operation were found only during the FTP driving cycle, but aldehyde emissions, typically associated with alcohol motor fuels, were higher than with gasoline operation, as shown in Figure 3. All of the FTP emissions essentially occurred in Bag 1 of the driving cycle, which indicates that the ethanol emissions were primarily due to cold operating conditions. Qualitative observation of the FTIR data indicated that the first thirty seconds of the Bag 1 portion was the source of both the ethanol and aldehyde emissions. The very low levels of aldehyde emissions during the US06 indicate that they are primarily associated with cold operation.

Although an increase was observed in ethanol emissions from E85 for cold conditions, E85 fuel demonstrated beneficial reduction of some MSAT species. Figure 4 shows select MSAT species emissions that are derived from gasoline fuel. The summation of these MSAT emissions decreased substantially for the E85 fuel relative to gasoline. The MSAT reduction occurred for both FTP and US06 driving cycles. Note that the US06 gasoline fuel results show higher levels of MSATs than the FTP gasoline fuel data due to the more aggressive driving cycle of the US06 test which results in more enrichment operation of the engine; enrichment

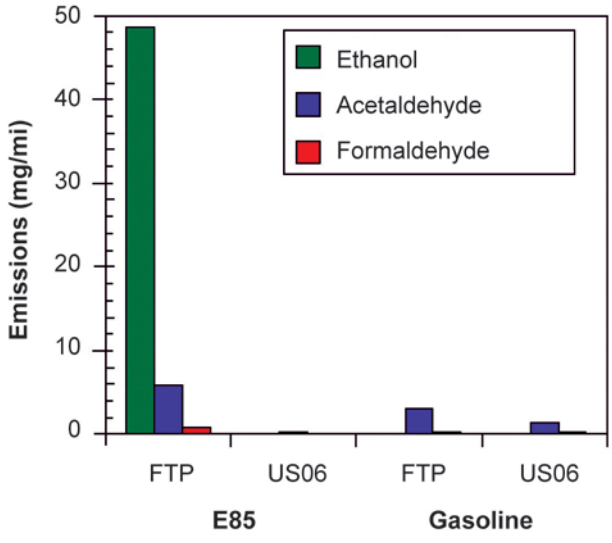

FIGURE 3. Ethanol and Aldehdye Emissions as a Function of Driving Cycle

leads to higher levels of unburned hydrocarbon emissions from the engine.

MSATs from HECC Diesel Mode

Diesel operation with the advanced HECC technique leads to a simultaneous reduction in NOx and particulate emissions without sacrificing fuel efficiency. At the 1,500 rpm, 2.6 BMEP operating condition used in this study, HECC operation resulted in a 90% reduction in NOx and a 70% reduction in smoke as compared with traditional diesel combustion. However, MSAT emissions for HECC were greater than with traditional diesel combustion. Figure 5 shows three common diesel MSAT emissions (formaldehyde, acetaldehyde, and acrolein) expressed in mass per time units for HECC and baseline (traditional combustion) conditions. Increases in the concentration of these species were even more significant due to the fact that lower exhaust flow results from HECC. Since emission regulations are mass-based as opposed to concentration-based, the emissions data here is reported as mass. The formaldehyde emissions were the highest and were almost double for HECC vs. the baseline. Although acetaldehyde and acrolein emissions were lower than formaldehyde, the increase in acetaldehyde and acrolein emissions relative to the baseline case were more dramatic. All the data reported here was obtained without emission control in the engine exhaust. Catalytic aftertreatment may be able to substantially reduce the increased MSAT emissions measured and will be the focus of future work.

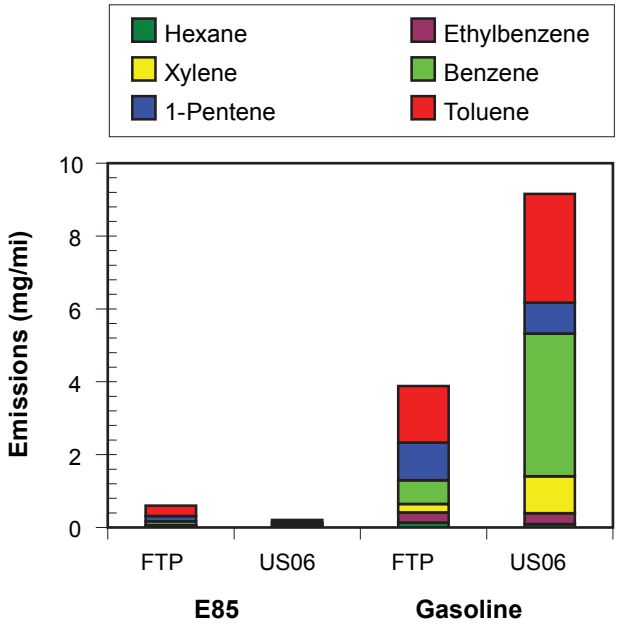

FIGURE 4. Gasoline-Derived MSAT Emissions as a Function of Driving Cycle and Fuel

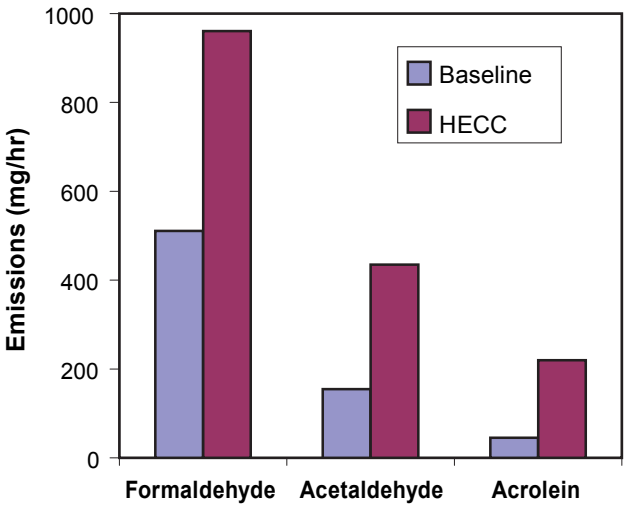

FIGURE 5. Formaldehyde, Acetaldehyde, and Acrolein emissions as a Function of Combustion Mode

Conclusions

- E85 Fueled Saab Biopower Vehicle:
  - E85 fuel decreases the amount of gasoline-derived MSATs by a factor of approximately 10 in comparison to gasoline fuel during FTP and US06 driving cycles.
  - Ethanol emissions increase dramatically for E85 in comparison to gasoline for the FTP driving cycle due to cold start emissions, but no increase in ethanol emissions were observed for the higher temperature US06 driving cycle.
- High Efficiency Clean Combustion:
  - In comparison to traditional diesel combustion, HECC operation results in lower NOx and particulate matter emissions; however, MSAT emissions from HECC were higher.
    - Formaldehyde emissions from HECC were almost a factor of two greater than from traditional combustion.
    - Acetaldehyde and acrolein emissions, while not as great as formaldehyde emissions, were observed at significantly higher quantities for HECC vs. traditional combustion.

References

1. Brian West, Alberto Lopez, Timothy Theiss, Ronald Graves, John Storey, Sam Lewis, "Fuel Economy and Emissions of the Ethanol-Optimized Saab 9-5 Biopower", *Society of Automotive Engineers Technical Paper Series* 2007-01-3994 (2007).

FY 2007 Publications/Presentations

1. Jim Parks, Sam Lewis, John Storey, Terry Miller, Joshua Fu, Boris Hromis, and Ralph McGill, "Mobile Source Air Toxics Measured at a Hot Spot of Diesel Truck Idling Emissions", *Presentation at the CRC Mobile Source Air Toxics Workshop* in Phoenix, AZ on October 24–25, 2006.

2. Terry Miller, Josh Fu, Boris Hromis, John Storey, and James Parks, "Diesel Truck Idling Emissions – Measurement at a PM2.5 Hot Spot", *Transportation Research Board,* Paper No. 07-3157 (2007).

3. James E. Parks, John M. E. Storey, Terry L. Miller, Joshua S. Fu, and Boris Hromis, "Diesel Truck Idling Emissions – Mobile Source Air Toxics Measured at a Hot Spot", *Transportation Research Board,* Paper No. 07-3158 (2007).

4. Douglas R. Lawson, John M. E. Storey, and James E. Parks II, "The OFCVT Health Impacts Program", *Poster Presentation at the 13th Diesel Engine-Efficiency and Emissions Research (DEER) Conference* in Detroit, MI on August 13-16, 2007.

5. Brian West, Alberto Lopez, Timothy Theiss, Ronald Graves, John Storey, Sam Lewis, "Fuel Economy and Emissions of the Ethanol-Optimized Saab 9-5 Biopower", *Society of Automotive Engineers Technical Paper Series* 2007-01-3994 (2007).

II.D.2 Collaborative Lubricating Oil Study on Emissions (CLOSE) Program

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Objective

The objective of this project is to quantify the role of engine lubricating oil on particulate matter (PM) emissions from in-use motor vehicles fueled with gasoline, 10% ethanol (E10), diesel, biodiesel, and compressed natural gas (CNG) while operating on fresh and used crankcase lubricants.

Accomplishments

- Established a Cooperative Research and Development Agreement (CRADA) between NREL and the South Coast Air Quality Management District (SCAQMD) and the California Air Resources Board (CARB) for program funding.
- Obtained funding from the Coordinating Research Council (sponsored by the automotive and petroleum industries) for program support.
- Obtained support from the American Chemistry Council Petroleum Additives Product Approval Protocol Task Group to provide new and aged engine lubricating oils for all vehicles that will be tested in the program.
- Increased scope of project to include medium-duty vehicles and E10 and biodiesel fuel testing as part of the overall project.
- Began testing of first vehicle in program in May 2007.

Future Directions

A variety of light-, medium-, and heavy-duty (LD, MD, HD) vehicles will be tested over different driving test cycles at room temperature (72°F) and cold temperature (20°F) on chassis dynamometers. The test matrix depicting the vehicles and test conditions is shown in Table 1.

TABLE 1. CLOSE Program Test Matrix

Test Temperature	72°F (nominal)				20°F			
Test Lubricant	Fresh		Aged		Fresh		Aged	
Vehicle / Sample Number	1	2	1	2	1	2	1	2
LD gasoline (normal PM emitter)	√	√	√	√	√	√	√	√
LD gasoline (high PM emitter)	√	√	√	√	√	√	√	√
LD E10 (normal PM emitter)	√	√	√	√	√	√	√	√
LD E10 (high PM emitter)	√	√	√	√	√	√	√	√
MD diesel (normal PM emitter)	√	√	√	√	√	√	√	√
MD diesel (high PM emitter)	√	√	√	√	√	√	√	√
MD biodiesel (normal PM emitter)	√	√	√	√	√	√	√	√
MD biodiesel (high PM emitter)	√	√	√	√	√	√	√	√
HD CNG (normal PM emitter)	√	√	√	√				
HD CNG (high PM emitter)	√	√	√	√				
HD diesel (normal PM emitter)	√	√	√	√				
HD diesel (high PM emitter)	√	√	√	√				

The engine lubricating oil used in the program is labeled with deuterated hexatriacontane ($C_{36}D_{74}$). This tracer, along with other naturally-occurring compounds found in lubricating oil, such as hopanes and steranes, will be used to quantify the relative contributions of PM formed from the fuels and the lubricants used in the vehicles that will be tested.

Normal and high-emitting vehicles representing gasoline, diesel, and CNG-powered vehicles will be tested. Lubricants used in each technology will be representative of those currently on the market, with both new and aged lubricants being tested in the program. The fuels used in the vehicles will be gasoline containing no ethanol, E10, CARB diesel, biodiesel, and CNG. Room temperature and cold temperature testing will be performed on all of the light- and medium-duty

vehicles. Cold temperature testing will not be conducted on the heavy-duty vehicles due to funding limitations.

The data collected throughout the study will be chemically analyzed with detailed speciation to quantify the relative importance of the fuel and lubricant to PM emissions from these vehicles under the variety of testing conditions specified in the study design.

Introduction

Recent air quality studies conducted in Denver, Phoenix, Washington D.C., and Office of FreedomCAR and Vehicle Technologies' (OFCVT) Gasoline/Diesel PM Split Study in Los Angeles have shown that PM from gasoline engines is a more significant contributor to ambient air quality than PM from diesel engines [1,2]. For example, data collected in Washington, D.C., over a ten-year period suggest that PM from gasoline exhaust is ten times more important to the emission inventory than diesel exhaust, as shown in Figure 1 [3].

OFCVT's Comparative Toxicity Studies have also shown that the toxicity from gasoline exhaust on a per-unit-mass basis is at least as toxic as that from diesel exhaust, and that high emitters' toxicity is even greater than that from normal emitters [4].

Because PM from both gasoline and diesel exhaust is so important to human health and ambient air quality, it is important to understand its source – whether it derives from the fuel, the lubricant, or both, and to understand the engine operating conditions that are responsible for PM emissions. That is the objective of the CLOSE Program.

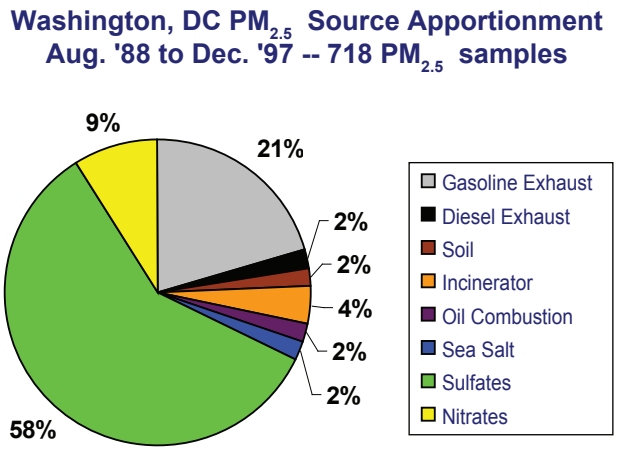

FIGURE 1. Source apportionment of $PM_{2.5}$ in the Washington, D.C. area. Samples were collected between 1988 and 1997.

Approach

The CLOSE Program will conduct extensive chemical and physical characterizations of PM emissions from vehicles fueled with gasoline, E10, diesel, biodiesel, and natural gas while operating on fresh and used crankcase lubricants in an effort to improve our current understanding of the impact of crankcase lubricant formulations on vehicle PM emissions. In-use light- and heavy-duty vehicles are being recruited, including both normal and high-PM emitters, and operated on chassis dynamometers at room temperature (72°F nominal) and 20°F. Gaseous (total hydrocarbons [THC], non-methane hydrocarbons [NMHC], carbon monoxide [CO], nitrogen oxides [NOx]) and real-time particle emissions are being measured, and PM and semivolatile organic compound (SVOC) samples are being collected for subsequent chemical analyses. Physical PM measurements will be conducted to obtain data on particle size, which will be investigated over the various driving conditions carried out on the dynamometers.

Results

At the time of this report, vehicle testing has begun on the light-duty normal emitter, which is a 2006 Chevy Impala having 37,000 miles on the odometer. Figure 2 shows the vehicle on the dynamometer, and Figure 3 shows the sampling ports and equipment used in the sampling tunnel.

FIGURE 2. Light-Duty Normal Emitter Tested in the CLOSE Program, a 2006 Chevrolet Impala

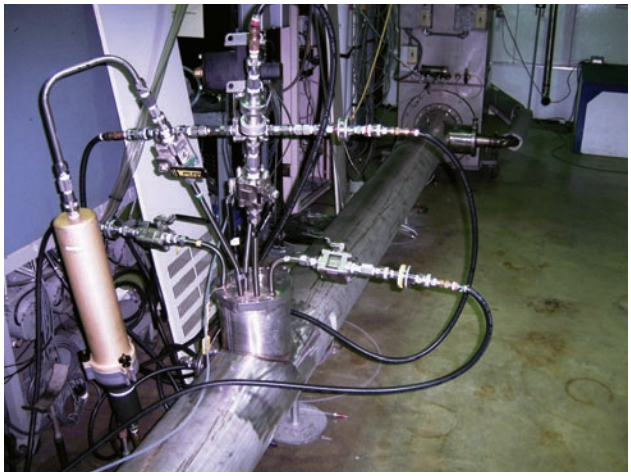

FIGURE 3. Sampling Probes Used to Collect Exhaust Emissions Samples from the Dilution Tunnel

Conclusions

Because this project has just started, there are no conclusions at the time of this report. The CLOSE Program will be completed by December 2008, so results will be available at that time.

References

1. Fujita, E.M., D.E. Campbell, W.P. Arnott, B. Zielinska, J.C. Chow. Evaluations of Source Apportionment Methods for Determining Contributions of Gasoline and Diesel Exhaust to Ambient Carbonaceous Aerosols, *J. Air & Waste Manage. Assoc.*, Vol. 57, pp. 721-740 (2007).

2. Lough, G.C. and J.J. Schauer. Sensitivity of Source Apportionment of Urban Particulate Matter to Uncertainty in Motor Vehicle Emissions Profiles, G.C. Lough and J.J. Schauer, *J. Air & Waste Manage. Assoc.*, Vol. 57, pp. 1200-1213 (2007).

3. Kim, E. and P. K. Hopke. Source Apportionment of Fine Particles in Washington, D.C., Utilizing Temperature-Resolved Carbon Fractions, *J. Air & Waste Manage. Assoc.*, Vol. 54, pp. 773-785 (2004).

4. McDonald, J.D., I. Eide, J.C. Seagrave, B. Zielinska, K. Whitney, D.R. Lawson, J.L. Mauderly. Relationship between Composition and Toxicity of Motor Vehicle Emission Samples, Environ. Health Persp., Vol. 112, pp. 1527-1538 (2004).

FY 2007 Publications/Presentations

1. "The Collaborative Lubricating Oil Study on Emissions (CLOSE) Project," presented at DEER 2007 Conference, Detroit, MI; August 15, 2007.

II.D.3 Health Impacts: Respiratory Response

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Objectives

- Compare health hazards of competing power technologies
- Determine the most biologically important components of emissions
- Evaluate health benefits of emission reduction technologies
- Evaluate emerging technologies for unanticipated health hazards

Approach

- Use animal and cell tests to characterize and quantify adverse health effects.
- Adapt, optimize, and validate biological test systems for emissions testing.
- Test whole and fractionated emissions from engines operated in the laboratory and vehicles operated elsewhere.
- Conduct detailed physical-chemical analysis of emissions and samples.
- Use physical-chemical fractionation and exposures to specific compounds to identify and confirm toxic components.
- Examine emerging technologies prior to commercialization.

Accomplishments

- Evaluated health effects of inhaled nanoparticles from new and used diesel crankcase oil and sulfate.
- Discovered that oil and sulfate nanoparticles had little lung toxicity, but altered responses of immune cells elsewhere in the body.
- Completed analysis of results from a comprehensive study of the effects of repeated inhalation exposure to laboratory-generated gasoline emissions.
- Discovered that inhaled nitrogen oxide and carbon monoxide can duplicate some effects of gasoline emissions on blood vessels outside the lung.

Future Directions

- This subproject of the Health Impacts activity will be completed in early FY 2008.
- Wrap-up will include examining effects of nanoparticles and emission gases at lower concentrations, and publishing results.
- Future effort in this area will focus on evaluating emerging technologies.

Introduction

This project is the biological evaluation subproject of the Health Impacts activity, and supports meeting DOE technical targets by: 1) placing in proper context the health hazards of engine emissions relative to other air quality hazards; 2) comparing the relative hazards of emissions from different fuel, engine, and emission reduction technologies; 3) determining the key toxic components among the hundreds of components of vehicle emissions; 4) demonstrating that reductions in emissions are paralleled by reductions in health hazards; and 5) evaluating emissions from emerging technologies to avoid unintended health consequences prior to commercialization. This project addresses potentially technology-limiting issues that are not addressed in other DOE or non-DOE programs. This project complements other Health Impacts subprojects that are characterizing emissions, determining the impacts of emissions on air quality, and conducting long-term health studies of 2007-2010-compliant diesel systems.

Approach

This project employs a four-tactic strategy to placing the health hazards of vehicle emissions in proper context, identifying the key toxic components, and evaluating new technologies. The first tactic involves laboratory evaluations of emission samples collected from vehicles and environments elsewhere. The second tactic involves animal studies of inhaled whole emissions, separated fractions of emissions, and specialized aerosols generated in the laboratory. Tissue oxidative stress, lung inflammation, cardiovascular effects, and immune responses are evaluated. The third tactic employs multivariate and univariate statistical analyses of data on composition vs. biological response

to identify components that cause adverse effects. The fourth tactic involves working with DOE managers and industry partners to identify advanced technologies having the greatest near-term commercialization potential, and using the above approaches to evaluate those technologies for unintended health consequences.

Work during the past year focused primarily on two issues pertaining to current technologies in order to clarify issues that must be addressed for emerging technologies: 1) the health importance of emission nanoparticles; and 2) the health effects of gases that comprise the non-particulate components of gasoline and diesel engine emissions. There has been considerable speculation about the health effects of so-called "nanoparticles" (under 50 nm diameter) from combustion and other sources. Questions have been raised about the importance of nanoparticles formed by condensation of vapors downstream of exhaust after-treatment devices. Such particles have always existed in engine emissions, but no technology has been available for separating them from other emissions for biological studies. A vaporization-condensation method was developed by this project last year to generate aerosols of nanoparticles from crankcase oil and sulfate, which are the primary sources of this material in contemporary emissions. This year, mice were exposed by inhalation to aerosols of new and used diesel engine lubrication oil and sulfate at a single high concentration to determine if measurable effects occur.

During the past few years, this project collaborated with other federal and non-federal partners (including several major large engine manufacturers and fuel providers) to conduct detailed studies directly comparing health effects of engine emissions to those of other pollution sources. Results finalized last year yielded the unexpected finding that blood vessels outside the lung were adversely affected by non-particulate components of gasoline emissions [3]. We hypothesized that nitrogen oxides and carbon monoxide might produce these effects through certain cellular chemical reactions, and might thus explain a portion of the known linkage between close proximity to traffic and cardiovascular effects in humans. This year, we exposed mice to these gases in pure form to determine whether these mechanisms were plausible.

Results

The detailed results of this project are communicated in numerous technical presentations and peer-reviewed scientific publications (FY 2007 products listed in Publications/Presentations, full listing available on request). Key recent results are summarized here.

For an initial exploration of the plausibility of hazard from emission nanoparticles, mice were exposed 6 hours/day for 7 days to aerosols of 20 nm oil and sulfate nanoparticles at 10^6 particles/cubic centimeter. Aerosols were formed by heating oil or dilute sulfuric acid, followed by carefully controlled condensation and dilution. We used new Shell Rotella-T® 15W-40 crankcase oil and the same oil from a normal change interval of a 2000 model Cummins 5.9L ISB engine using pre-2007 certification fuel and operated on repeated Environmental Protection Agency (EPA) heavy-duty certification cycles. Lung irritation was evaluated by analysis of lung surface fluid and histopathology. Earlier studies had revealed that the function of immune cells of the spleen was diminished by inhaled diesel emissions and wood smoke [1,2]. Cells (T and B lymphocytes) were therefore removed from spleens of nanoparticle-exposed mice and their ability to divide and produce antibody were measured.

The exposures to nanoparticles did not cause evidence of lung inflammation, either by histopathology or cell and biochemical markers in airway fluid. It is notable that exposure to this high concentration of the three aerosols did not cause inflammatory or irritant responses of the types considered sensitive indicators of the effects of whole emissions in human subjects. The only evidence that the lungs were exposed was an increase in tissue levels of hemeoxygenase-1, a general marker that is increased by many inhaled pollutants.

The exposure caused a mild suppression of the systemic (outside the lung) immune system. In a systemic immune response, lymphocytes in the spleen and other lymphoid tissues must divide to produce more cells, and must also produce antibodies to foreign materials. Two types of lymphocytes, T and B cells, were removed from the spleen and treated with materials that stimulate cell division and antibody production. In condensate-exposed animals, the cell division responses of both cell types were similarly reduced (Figure 1, showing only T cells). New and used oil caused similar modest reductions (approximately 20%), and sulfate caused a greater (nearly 40%) reduction. The three exposures also reduced the ability of cells to produce antibodies (Figure 2, showing only T cells). New and used oil caused reduced antibody formation approximately 60%, but sulfate reduced antibody production by over 70%.

These initial findings provide important new information. First, nanoparticles may present little, if any, hazard to the lung, but may have effects on the systemic immune system. Second, there was no difference between the responses to new and used diesel crankcase oil, suggesting that contaminants accumulating normally in diesel crankcase oil may not present an increasing hazard as they accumulate. Third, pure sulfate nanoparticles having quite different chemistry than oil caused the same effects, and actually caused greater immune suppression than the oil aerosols. Together with the concurrent finding in

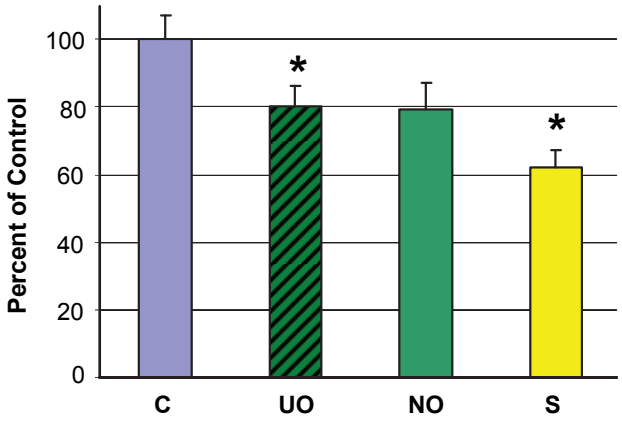

FIGURE 1. Response of Splenic T Lymphocytes to Stimulation to Divide (with Concanvalin-A) (Bars represent means ± standard error expressed as percentages of responses of sham-exposed mice. Asterisks indicate statistically significant responses. C = sham-exposed controls, UO = used oil, NO = new oil, S = sulfate.)

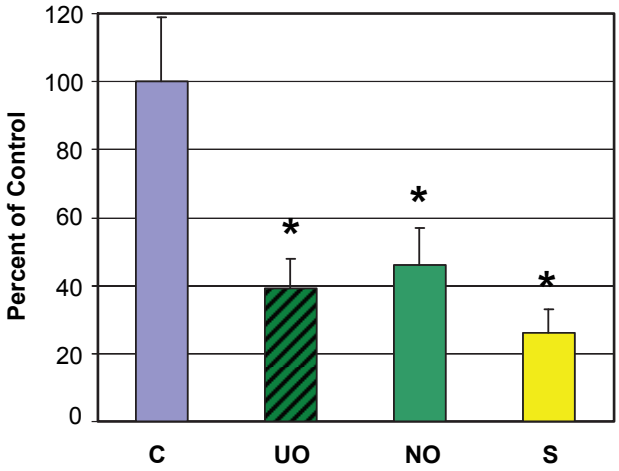

FIGURE 2. Response of Splenic T Lymphocytes to Stimulation to Produce Antibody to Foreign Protein (Sheep Red Blood Cells) (Bars represent means ± standard error expressed as percentages of responses of sham-exposed mice. Asterisks indicate statistically significant responses. C = sham-exposed controls, UO = used oil, NO = new oil, S = sulfate.)

another project that solid carbon nanoparticles cause very similar effects [4] and the earlier finding that whole diesel emissions and wood smoke exert similar systemic immune effects [1,2], these results suggest a general response to nanoparticles and pollution mixtures rather than a specific response to engine emissions. It will be important to follow up these first results with exposures to lower concentrations of emissions nanoparticles, to determine whether more broadly-relevant exposures cause measurable effects.

In the second study, mice were exposed 6 hours/day for 7 days to 17 ppm nitrogen oxide (NO), 0.2 or 2.0 ppm nitrogen dioxide (NO_2), 8 or 80 ppm carbon monoxide (CO), or the combination of 8 ppm NO_2 and 80 ppm CO. This study was an initial exploration of the plausibility that these gases might have caused the vascular effects observed in mice exposed to filtered gasoline emissions. The higher concentrations in this study mimicked those at the highest level of whole gasoline emissions in the previous work. Several indicators of oxidative stress and early stages of adverse tissue responses were measured in blood vessels near the heart.

As illustrated by the response of the indicator endothelin-1 in Figure 3, neither of the concentrations of NO_2 nor the lower concentration of CO caused changes in blood vessels. However, exposures to the high concentrations of CO and NO did cause significant responses. The lack of importance of NO_2 was reinforced by the finding that addition of NO_2 to the high concentration of CO did not increase the response. This same pattern was observed in the indicators

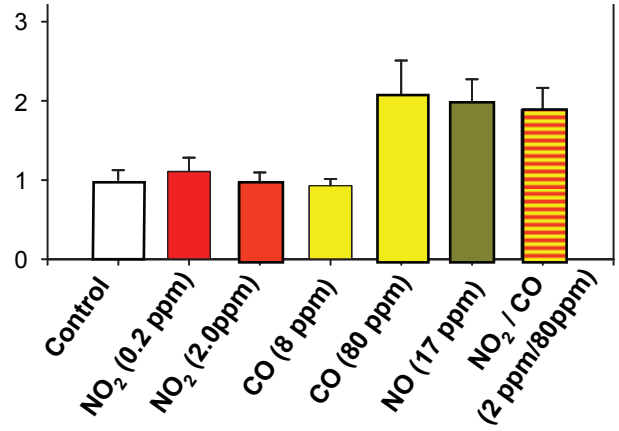

FIGURE 3. Levels of the Damage Indicator Endothelin-1 in Aorta near the Hearts of Mice Exposed to Gases (Bars represent means ± standard error of responses in comparison to sham-exposed controls [control response = 1]. Asterisks indicate statistically significant responses.)

hemeoxygenase-1 and matrix metalloproteinase-9. However, some other indicators of vascular response that were altered in the gasoline emission study were not altered by any of the single-gas exposures.

Although only a preliminary exploration, this study provided useful information. First, neither NO_2 nor CO at levels meeting current environmental standards caused measurable effects. The lack of effect of NO_2 is not surprising, because the reactivity of the gas would be expected to prevent any direct effect on tissues outside the lung. Second, the fact that high concentrations of both NO and CO did cause measurable effects suggests

that our hypotheses about the mechanisms of response to high levels of gasoline emissions are at least partially correct. Third, the fact that only some, but not all, of the markers of blood vessel effects were increased suggests that other non-particulate emissions, such as volatile organic compounds, also contributed to vascular responses at high concentrations of filtered gasoline emissions. Experimental logistics limited the number of exposures that could be included in this series; exposures to a lower concentration of NO and to a combination of NO and CO should be conducted. It will be important to determine whether more realistic exposures to NO present a hazard, and whether the effects of NO and CO are additive.

Conclusions

Emissions from all internal combustion motive power technologies evaluated to date are attended by potential adverse health impacts of some nature and at some exposure level. These effects need to be considered in the evaluation of emerging technologies. The new evidence indicates that attention to crankcase oil emissions is warranted, and emissions of organic and sulfate condensate nanoparticles should be minimized. The evidence also suggests that the non-particle components of emissions warrant continued attention. Fortunately, with few exceptions, significant effects are only observed at the highest exposure concentrations that might be encountered, and such exposures in a few occupational settings or traffic "hotspots" should be of shorter duration than the experimental exposures. Also fortunately, the fuel-trap-catalyst technologies required to meet 2010 on-road diesel emission standards should markedly reduce not only gases, but also vapors that could condense to form nanoparticles.

FY 2007 is the last year of funding for this project in its present form. Many, but not all, issues pertaining to the purpose of this project have been addressed. As noted above, a few additional exposures would be required to complete the recent work by determining whether effects occur at more widespread exposure levels. This will be accomplished as funding is available.

References

1. Burchiel, S.W., F.T. Lauer, J.D. McDonald, and M.D. Reed: Systemic Immunotoxicity in AJ Mice following 6-Month Whole Body Inhalation Exposure to Diesel Exhaust. *Toxicol. Appl. Pharmacol.* 196: 337-345, 2004.

2. Burchiel, S.W., F.T. Lauer, S.L. Dunaway, J. Zawadzki, J.D. McDonald, and M.D. Reed: Hardwood Smoke Alters Murine Splenic T Cell Responses to Mitogens Following a Six Month Whole Body Inhalation Exposure. *Toxicol. Appl. Pharmacol.* 202 (3): 229-236, 2005.

3. Lund, A.K., T.L. Knuckles, C.O. Akata, R. Shohet, J.D. McDonald, A. Gigliotti, JC. Seagrave, and M.J. Campen: Gasoline Exhaust Emissions Induce Vascular Remodeling Pathways Involved in Atherosclerosis. *Toxicol. Sci*. 95: 485-494, 2007.

4. Mitchell, L.A., J. Gao, R.V. Wal, A. Gigliotti, S.W. Burchiel, and J.D. McDonald: Pulmonary and Systemic Immune Response to Inhaled Multiwalled Carbon nanotubes. *Toxicol. Sci.* 100: 203-214, 2007.

FY 2007 Publications/Presentations

Publications

1. Braun, A., F.E. Huggins, A. Kubatova, S. Wirick, M.M. Maricq, B.S. Mun, J.D. McDonald, K.E. Kelly, N. Shah, and G.P. Huffman: Towards Distinguishing Wood Smoke and Diesel Exhaust Particulates in Ambient Particulate Matter. *Atmos. Environ*. (submitted).

2. Mauderly, J.L. and J.C. Chow: Health Effects of Organic Aerosols. *Inhal Toxicol*. (provisionally accepted – in revision).

3. Mauderly, J.L. and E. Garshick: Diesel Exhaust. Chapter 17 in: *Environmental Toxicants*, Lippmann, Ed., Wiley, New York (accepted and in press).

4. McDonald, J.D., M.D. Reed, M.J. Campen, E.G. Barrett, JC. Seagrave, and J.L. Mauderly: Health Effects of Inhaled Gasoline Engine Emissions. *Inhal. Toxicol.* 19(Suppl. 1): 107-116, 2007.

5. Lund, A.K., T.L. Knuckles, C.O. Akata, R. Shohet, J.D. McDonald, A. Gigliotti, JC. Seagrave, and M.J. Campen: Gasoline Exhaust Emissions Induce Vascular Remodeling Pathways Involved in Atherosclerosis. *Toxicol. Sci.* 95: 485-494, 2007.

6. Seagrave, JC., S. Dunaway, P. Hayden, J.D. McDonald, S. Stidley, and J.L. Mauderly: Responses of Differentiated Primary Human Lung Epithelial Cells to Exposure to Diesel Exhaust at an Air-Liquid Interface. *Exper. Lung Res.* 33:27-51, 2007.

Presentations

1. Mauderly, J.L.: Health Effects of Organic Aerosols, Introduction and Summary, Workshop on Health Effects of Organic Aerosols, EPRI, Palo Alto, CA, October 24–25, 2006.

2. Mauderly, J.L.: Disentangling Components Responsible for Health Effects of Complex Air Pollution Mixtures, Desert Research Institute, Reno, NV, November 21, 2006.

3. Mauderly, J.L.: Health Research Needs for Multi-Pollutant Air Quality Management. NARSTO Workshop on Multi-Pollutant Air Quality Management. Durham, NC, January 10, 2007.

4. Mauderly, J.L. et al.: Disentangling the Health Hazards of Complex Mixtures of Air Pollutants. Division of Occupational and Environmental Health Sciences, University of Texas School of Public Health, Houston, TX, February 16, 2007.

5. Seagrave, JC., M.J. Campen, S. Dunaway, G. Herbert, J.L. Mauderly, and J.D. McDonald. Exposure to Gasoline Engine Exhaust Causes Oxidative Stress in Rats. Society of Toxicology Annual Meeting, Charlotte, NC, March 2007.

6. McDonald, J.D., JC. Seagrave, L. Mitchell, A. Gigliotti, and J.L. Mauderly. Pulmonary Inflammatory and Systemic Immune Responses to Inhaled Oil Nanocondensates. Society of Toxicology Annual Meeting, Charlotte, NC, March 2007.

7. Lund, A.K., T. Knuckles, JC. Seagrave, C.O. Akata, J.D. McDonald, M.J. Campen: Exposure to Whole Gasoline Engine Emissions Results in Alterations of Molecular Pathways Involved in Progression of Atherosclerosis. Society of Toxicology Annual Meeting, Charlotte, NC, March 2007.

8. Reed, M.D., A. P. Gigliotti, and J.A. Berger: Gasoline Emissions Affect Clearance of Intratracheally Instilled *Pseudomonas aeruginosa*. Society of Toxicology Annual Meeting, Charlotte, NC, March 2007.

9. Mauderly. J.L.: Health Impacts of Diesel Emissions: An Update. Environment Canada, Ottawa, Canada, April 2007.

10. Mauderly, J.L.: ACES Phase 3: Chronic Inhalation Bioassay. Annual Meeting of the Health Effects Institute, Chicago, IL, April, 2007.

11. Barrett, E.G., M.D. Reed, J.D. McDonald, S. Shinnik, and M. Anaya: Effects of Gasoline Engine Emissions on Pre-Existing Allergic Airway Responses. Annual Conference of the American Thoracic Society International Conference, San Francisco, CA, May 2007.

12. Mauderly, J.L.: Health Impacts of Diesel Emissions: An Update. Committee on 21st Century Truck Program, National Research Council, National Academy of Sciences, Washington, D.C., May 2007.

13. Mauderly, J.L., M.J. Campen, J.D. McDonald, and JC. Seagrave: Components Responsible for the Health Effects of Engine Emissions. Diesel Engine Emissions Reduction Conference (DEER 2007), Detroit, MI, August 2007.

II.D.4 The Advanced Collaborative Emissions Study (ACES)

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Objectives

- **Phase 1**: Extensive emissions characterization (at an existing emissions characterization facility) of four production-intent heavy duty diesel engine and control systems designed to meet 2007 standards for particulate matter (PM) and nitrogen oxides (NOx). One engine/aftertreatment system will be selected for health testing.
- **Phase 2**: Extensive emissions characterization of a group of production-intent engine and control systems meeting the 2010 standards (including more advanced NOx controls to meet the more stringent 2010 NOx standards).
- **Phase 3**: One selected 2007-compliant engine will be installed in a specially-designed emissions generation and animal exposure facility (Phase 3A) and used in chronic and shorter-term health effects studies to form the basis of the ACES safety assessment (Phases 3B and 3C). This will include periodic emissions characterization during both a core 24-month chronic bioassay of cancer endpoints in rats and shorter term biological screening assays in both rats and mice (Phase 3B) as well as emission characterization during a set of shorter animal exposures and biological screening using accepted toxicological tests after the end of the chronic bioassay (Phase 3C). (NOTE: Only the emissions characterization and shorter-term biological screening activities at the beginning and during Phase 3 are components of the DOE ACES contract.)

Accomplishments

General Oversight

- Held meetings with the ACES Oversight, Advisory and Steering Committees regarding finalization of emissions generation and characterization, health effects assessment, and cost savings options for Phase 3, and to finalize financial commitments (Fall 2006).
- Held a meeting with the ACES Oversight and Advisory Committees to discuss progress and technical issues (April 2007).
- Held meetings of the CRC ACES Panel at Southwest Research Institute (SwRI) to discuss technical issues pertaining to emissions characterization (Spring 2007).
- Communicated at meetings and conference calls with engine manufacturers, ACES Oversight, Advisory and Steering Committees, and selected members of the CRC ACES Panel to develop a plan for selection of one engine for use in Phase 3 (Spring and Summer 2007).
- Held a Panel meeting to discuss and review competitive proposals for biological screening studies submitted in response to Request for Applications (RFA) 06-2 (August 2007).

Phase 1

- Selected fuel and lubricant suppliers and arranged for delivery to both SwRI and Lovelace Respiratory Research Institute (LRRI) (October 2006).
- Finalized the contract with SwRI for Phase 1 emissions characterization (December 2006).
- Completed preparation of the engine testing facility at SwRI (February 2007).
- Received four test engines from the participating engine manufacturers at SwRI and agreed upon the order in which they would be tested (April 2007).
- Finalized and approved the 16-hour test day cycle developed by West Virginia University based on a combination of Federal Test Procedure (FTP) and California Air Resources Board (CARB) cycles for use in Phase 3 (June 2007).
- Finalized the emissions characterization protocol with the CRC ACES Panel and the investigators' team at SwRI and approved a revised program plan from SwRI (June 2007).
- Finalized the last remaining technical details concerning the emissions characterization protocol for Phase 1 and started emissions characterization

of the first 2007-compliant engine (engine A) at SwRI (July 2007).
- Completed emissions characterization of the first and second 2007-compliant engines (engines A and B) and finalized procedures for trap regeneration (September 2007).

Phase 3

- Selected LRRI to conduct Phase 3A and B, the core emissions characterization and chronic bioassay (Fall 2006).
- Finalized details of facility development, emissions characterization, and health effects assessment for Phase 3 with the LRRI principal investigator and the ACES HEI Oversight Committee, specifically regarding cost savings options (Fall 2006).
- Conducted a site visit to LRRI to discuss cost-saving options related to engine facility, dynamometer, and dilution systems with key experts on the HEI ACES Oversight and Advisory Committees and additional experts identified by CRC (December 2006).
- Extended the deadline for submission of proposals for biological screening under RFA 06-2 to May 24, 2007 to increase the potential of a large number of high quality applications for additional biological screening during Phase 3 and encouraged prospective applicants to contact the LRRI team to discuss technical and logistical issues (Spring 2007).
- Finalized remaining technical details of the chronic exposure bioassay and related biological screening studies in Phase 3 (May 2007).
- Agreed upon a revised budget with LRRI, completed the first round of contract negotiations, and began facility implementation (August 2007).
- Received five applications for shorter term biological screening studies during Phase 3B submitted under RFA 06-2 (May and June 2007).
- Reviewed the five applications to RFA 06-2 with an expert Review Panel and solicited revised applications (August 2007).
- Discussed with the four engine manufacturers and CRC the specifications of the facility implementation at LRRI in order to allow any of the 2007-compliant engines to be accommodated, if selected (August, September 2007).

Future Directions

Phase 1

- Complete emissions characterization of the third and fourth 2007-compliant engines (engines C and D) at SwRI (Fall 2007).
- Finalize the engine selection criteria described in the Initial Plan for Engine Selection with the ACES Oversight and Steering Committees (Fall 2007).
- Continue to work with LRRI, CRC, and the engine manufacturers to determine appropriate specifications for the LRRI engine facility (Fall 2007).
- Receive detailed the emissions characterization data for all four engines from SwRI and collaborating researchers at Desert Research Institute (Winter 2007).
- Adjust the engine selection criteria if warranted by the findings, as described in the Final Draft Initial Plan; agree upon a Final Plan for Engine Selection (Fall 2007).
- Select one engine for use in Phase 3; receive a duplicate engine and characterize its emissions at SwRI (Spring 2008).
- Ship the selected engine and its duplicate to LRRI for installation in their facility (Spring 2008).
- Receive a Phase 1 Final Report from SwRI (Summer 2008).

Phase 3

- Finalize the second stage of contract negotiations for conduct of the chronic bioassay and biological screening at LRRI (Winter 2007).
- Select contractors to conduct the biological screening study for Phase 3B (Fall 2007).
- Finalize protocols for biological screening with those contractors and the investigators' team at LRRI (Winter/Spring 2008).
- Complete facility implementation at LRRI (Spring 2008).
- Install the selected engine and store the duplicate engine (Spring/Summer 2008).
- Perform initial emissions characterization of the selected and duplicate engines at LRRI to ensure both engines perform in a similar fashion as when they were characterized at SwRI (Summer 2008).
- Start animal exposures for the chronic bioassay and biological screening studies (Fall 2008).

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Introduction

ACES is a cooperative, multi-party effort to characterize the emissions and assess the safety of advanced heavy-heavy duty diesel engine and aftertreatment systems and fuels designed to meet the 2007 and 2010 emissions standards for PM and NOx. The ACES program is being carried out by HEI and CRC. It is utilizing established emissions

characterization and toxicological test methods to assess the possible health effects of production-intent engine and control technology combinations that will be introduced into the market during the 2007-2010 time period. This is in direct response to calls in the U.S. Environmental Protection Agency Health Assessment Document for Diesel Engine Exhaust [1] for assessment and reconsideration of diesel emissions and health risk with the advent of new cleaner technologies.

The characterization of emissions from representative, production-intent advanced compression ignition (CI) engine systems will include comprehensive analyses of the gaseous and particulate material, especially those species that have been identified as having potential health significance. The core toxicological study will include detailed emissions characterization at its inception, and periodically throughout a two-year chronic inhalation bioassay in rats similar to the standard National Toxicology Program (NTP) bioassay. Other specific biological screening studies also will be undertaken in both rats and mice, to evaluate these engine systems with respect to carefully selected respiratory, immunologic, and genotoxic effects for which there are accepted toxicologic tests. It is anticipated that these emissions characterization and health effects studies will assess the safety of these advanced CI engine systems, will identify and assess any unforeseen changes in the emissions as a result of the technology changes, and will contribute to the development of a database to inform future assessments of these advanced engine and control systems.

Approach

Experimental work under ACES will be performed in three phases, as outlined in the Objectives. Detailed emissions characterization (Phases 1 and 2) will be performed by an existing engine laboratory (SwRI) that meets the U.S. Environmental Protection Agency specifications for 2007 and 2010 engine testing. In Phase 1, emissions from four 2007-compliant engine/control systems will be characterized. One engine will be selected for health testing in Phase 3. In Phase 2, emissions from four 2010-compliant engine/control systems will be characterized. In Phase 3, one selected 2007-compliant engine/control system will be installed in a specially designed emission generation facility connected to a health testing facility at LRRI to conduct a chronic inhalation bioassay in rats and shorter term biological screening in rats and mice. During the 2-year bioassay, emissions will be characterized at regular intervals throughout the testing.

The emissions characterization work will be overseen by CRC and its ACES Panel. The health effects assessment will be overseen by HEI and its ACES Oversight Committee. Set-up of the emission generation facility at the health effects testing facility (for Phase 3) and finalizing the protocols for periodic emission characterization throughout Phase 3 will be done with input from the team of investigators selected to conduct emissions characterization in Phase 1 and CRC.

Results

The emissions characterization in Phase 1 is well underway; at this time characterization of two of the four engine/aftertreatment combinations has been completed. Successful meetings were conducted with multiple stakeholders of ACES to discuss and agree upon technical issues for both Phases 1 and 3. Much progress was made this year in terms of developing and finalizing the test cycles and other technical procedures, including how to handle trap regeneration and crankcase emissions. Initial contract negotiations with the emissions characterization and health testing facility for conduct of Phase 3 were completed; facility implementation was started after various cost saving options were identified and agreed upon. A final draft of the Initial Plan for Engine Selection has been written and circulated and is expected to be finalized soon.

Conclusions

We have made substantial progress towards the implementation of the project: (1) emissions characterization in Phase 1 is well underway, and (2) facility implementation for Phase 3 has started and detailed experimental protocols will be finalized in the coming months. Furthermore, five additional teams of investigators have submitted proposals for supplementary biological screening studies to address important health endpoints; revised proposals will be reviewed and funding decisions made in the Fall of 2007.

References

1. U.S. Environmental Protection Agency, 2002. Health Assessment Document for Diesel Engine Exhaust. EPA/600/8-90/057F. U.S. Environmental Protection Agency, National Center for Environmental Assessment, Office of Research and Development, Washington, D.C.

FY 2007 Publications/Presentations

1. Poster Presentation at the CRC ON-Road Vehicle Emissions Workshop in San Diego, March 26-28 2007, "Status of the Advanced Collaborative Emissions Study (ACES)."

2. Poster Presentation at the HEI Annual Conference in Chicago IL, April 15-17 2007, "Status of the Advanced Collaborative Emissions Study (ACES)" (HEI Annual Conference 2007 Program Book, page 68).

3. Poster Presentation "Status of the Advanced Collaborative Emissions Study (ACES)" at the DOE Semi-Mega Merit Review Meeting, Crystal City, VA, June 18, 2007.

4. Poster Presentation at the Diesel Engine Emissions Reduction (DEER) meeting in Detroit MI, August 13–16 2007, P-23: "Status of the Advanced Collaborative Emissions Study (ACES)."