ITEM NO: 3
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PROPOSAL NO.: 2452-214

STAFF EVALUATION OF AN INTERAGENCY RESEARCH PROPOSAL

TITLE:	Determination of the Contributions of Light Duty and Heavy Duty Vehicle Emissions to Ambient Particles in California
CONTRACTOR:	University of California, Riverside
PRINCIPAL INVESTIGATOR:	Kimberly Prather
AMOUNT:	$200,653
DURATION:	24 Months

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I. SUMMARY

To attain the National Ambient Air Quality Standards for particulate matter (PM), California needs to develop controls to reduce ambient concentrations of aerosols, including aerosols generated by internal combustion engines. Particles emitted by diesel engines are of special interest since, in addition to being a significant contributor to ambient PM, diesel particles have also been determined to be a Toxic Air Contaminant. Developing a scientific foundation for future PM controls requires research on three fronts: 1) to characterize directly emitted "primary" aerosols in sufficient detail to link them to particular sources; 2) to study of the dynamics of particle aging in ambient air; and 3) to investigate "secondary" particle formation from gaseous precursors in direct gas-to-particle, droplet, and condensation processes. The tasks in this proposal would address certain open issues regarding the characterization of motor vehicle aerosols in both controlled (dyno testing) and ambient settings, and would then apply a refined understanding of motor vehicle aerosols to the analysis of data collected in brief field studies and archival data from previous field studies.

Dr. Prather's previous work with Aerosol Time of Flight Mass Spectrometry (ATOFMS) has demonstrated that ATOFMS is uniquely well suited to these problems since it can overcome many of the limitations of conventional aerosol analysis. ATOFMS provides real-time data, allowing much better linkage between sample data and source activity. It eliminates long time lags between sample collection and data availability, thus facilitating adaptability in experimental work. The ATOFMS analyzes particles virtually instantaneously, thus eliminating positive and negative chemical artifacts that develop in filter and impactor samples. Most importantly, ATOFMS provides specificity regarding individual particles' sizes and compositions, allowing direct observation of a heterogeneous mix of particles in ambient air.

II. TECHNICAL SUMMARY

Objective

The objective of this project is to obtain real-time direct measurements of motor vehicle-related ambient aerosols, segregated by general vehicle class — light duty gasoline (LDG) *versus* heavy-duty diesel (HDD). These measurements would use ATOFMS to pair individual particles collected from ambient air with particles collected directly from motor vehicles. Since source test methods for motor vehicle particle emissions do not perfectly replicate "real world" conditions, actual field measurements would be preceded by a set of smog chamber experiments designed to refine the linkages between source-test and ambient "fresh" motor vehicle particles and to clarify the effects of "aging" in ambient air on motor vehicle particles.

Background

This project builds on three studies, performed by Dr. Prather and others that are rooted in the SCOS 97-NARSTO Aerosol Program (SCOSAero). As an adjunct to the SCOSAero field sampling program, the U.S. Department of Energy's National Renewable Energy Laboratory (NREL) asked Dr. Prather to attempt to identify the partial contributions from light duty gasoline (LDG) and heavy duty diesel (HDD) vehicles in the SCOSAero ambient aerosols. Previous ARB-funded research had

identified the Caldecott Tunnel (California Highway 24 between Oakland and Walnut Creek) as an excellent place to study diesel vs. gasoline emissions, because virtually all its HDD traffic is funneled through one bore of the multi-bore tunnel. This situation allows direct comparison of emissions from a mixed population of LDG and HDD vehicles with that of a "pure" LDG vehicle population. In an effort to get in-use vehicle emissions contemporary with the ambient sampling of SCOSAero, an ATOFMS and accompanying samplers (to duplicate field setups for SCOSAero) were deployed to the Caldecott Tunnel in the fall of 1997. Unfortunately, the operating conditions in the tunnel (low ambient temperature) reduced the ATOFMS' desorption laser power, rendering the tunnel data not directly comparable with that from the SCOSAero field sampling. Serendipitously, the low power made detection of high-carbon number compounds easier and facilitated identification of marker PAHs. Preliminary results from the tunnel experiment, published early this year (Figures 1 & 2 and Gross, *et al*., 2000; referenced in proposal), show that the ATOFMS did detect significant size distribution, chemical composition, and particle number differences between emissions from LDG and HDD vehicles.

A few months later, in order to get a "matching" set of data under full laser desorption power conditions, the same instrumentation suite was deployed to sample exhaust from 11 in-use motor vehicles running the Federal Test Procedure (FTP) in ARB's dyno bays at El Monte. This experiment also showed a clear distinction between LDG and HDD vehicle emissions (Figures 3 -7 in proposal). As expected, these data were consistent with, but not identical to the tunnel observations.

Finally, Dr. Prather's group ran a short sampling study alongside a freeway in Riverside to characterize "ambient" motor vehicle particles. This study produced a third set of distinguishing characteristics (Figures 8 & 9 in proposal) which were consistent with the dynamometer test data, but also showed evidence of atmospheric transformation ("aging") of particles.

Taken together, these studies show that there are clear markers for distinguishing LDG particle emissions from those of HDDs, and that these markers can be found in ambient air. Closing the circle to get from quantitative estimates of the LDG and HDD contributions to ambient particle concentrations, however, requires resolving the differences seen in these three experiments. The issues to be addressed include tying "fresh" in-use emissions (tunnel) to dyno tests; linking dyno test data to ambient measurements; and characterizing the aging processes so that not only "fresh" particles, but also "old" motor vehicle particles in ambient air can be linked to their sources.

A. Project Summary

The proposed work involves three sets of experimental observations and a data analysis program. These are broken down into a core project and optional tasks.

Core Project: The core of this project is a set of experiments designed to resolve questions arising from the variation between dyno test emissions and "real world" aerosols observed in the roadside study. Experimentation would address two sources of variation. The experimental results would then be applied in analysis of existing ambient aerosol data.

Experiment 1. Quantify the effects of dilution tunnel operations on sampled exhaust particle characteristics in controlled sampling (dynamometer tests). Using both gas and diesel vehicles as particle sources, and sampling with a variable dilution tunnel, Dr. Prather's group would perform a set of experiments to quantify the impact of dilution tunnel operations on LDG and HDD exhaust samples. Secondarily, these experiments would explore how changing "real world" conditions (temperature, speed, etc.) may alter in-use motor vehicle particle emissions. These experiments should provide:

- Tight linkage between dynamometer test particle data and "fresh" real-world motor vehicle particles as observed in the Freeway Study
- Improved LDG and HDD emission "profiles" suitable for recognizing each vehicle

type's emissions in ambient samples (such as the SCOSAero data)

- Discussion of how to minimize dilution-tunnel-induced artifacts in future LDG and HDD dynamometer tests.

Experiment 2. Quantify the differences between test emissions and "aged" ambient particles due to the effects of atmospheric transformations on particles. The effects of residence time, ultraviolet light, and vapor-phase oxidants on particle size and chemistry would be explored by putting LDG and HDD exhaust streams into a smog chamber where each of these variables can be separately manipulated. The goal of this work would be to explain the modified particles observed in the Freeway Study.

Ambient Data Analysis. Demonstrate determination of the contributions of gasoline and diesel vehicles in ATOFMS ambient particle data. Archived ambient ATOFMS measurements would be analyzed for their LDG and HDD mass fractions, using the size- and chemically-resolved emission profiles (developed in Experiments 1 and 2) for "fresh" and "aged" LDG and HDD particles and neural net classification methods (developed under a previous ARB contract). The data sets to be evaluated were obtained in various field experiments, including the 1997 Caldecott Tunnel Study, the El Monte dynamometer tests, the Freeway Study, and the multi-day data sets from the SCOSAero program in Southern California. The results of this effort would be a demonstration of highly time-resolved LDG/HDD source apportionment from ATOFMS sampling.

Optional Task 1: Upwind-downwind freeway sampling. This task entails field sampling for roughly one week. Quantification of vehicle emission rates, using modeled dilution, would require meteorological support and, possibly, a cooperating investigator to perform detailed organic compound speciation. The goal of this project would be to confirm, in ambient sampling, the fresh emission characteristics and particle aging dynamics interpreted from the Core Project.

Optional Task 2: Repeat the Caldecott Tunnel Study with updated ATOFMS instruments operating in both low- and high-power modes. As before, sampling would take place in the HDD and LDG bores of the tunnel for a period of one week. This would directly link the low-power results of 1997 (and the PAHs thus observed) with the "normal" ATOFMS data obtained in ambient studies. This study would permit comparison of LDG and HDD particle emission profiles obtained from dynamometer testing of limited numbers of vehicles with those developed from in-use fleet emissions.

A secondary goal of this sampling program would be the comparison of low- and high-power ATOFMS data to explore methods to estimate high-carbon number compound (PAHs) concentrations in real-time ATOFMS ambient aerosol measurements.

Optional Task 3: Add nanoparticle measurements to Core Project experiments and Optional Tasks 1 and 2 above.

This involves installing a size-selective tunable inlet system on existing ATOFMS units. This modification would extend the size range of the ATOFMS data from its "normal" detection minimum of 0.3 micrometers down to about 10 nanometers. This latter size range is currently the subject of considerable interest as a possible source of much of the adverse health effects attributed to PM exposure. Experiments incorporating these measurements could help resolve the current controversy about the "real world" emission rates and atmospheric lifetimes of nanoparticles from internal combustion engines. Conventional sampling methods are unable to provide the size- and time-resolved chemical data necessary to study these dynamics.

B. Project Management

The project would proceed in stages. The controlled experiments with the dilution tunnel and smog chamber would be completed and the results reviewed by ARB staff prior to commencing fieldwork for either the Freeway or Caldecott Tunnel Optional

Tasks. Detailed sampling plans for these latter tasks would be prepared in consultation with ARB staff.

ARB would closely monitor progress of the research and work with the investigators to assure that the experimental program focuses on questions relevant to improving motor vehicle particle emission data, facilitates comparison of these projects' results with other recent on-road vehicle studies, and addresses issues relevant to designing control strategies for PM.

The staging of the project and the seasonal limitations on fieldwork require that this project be conducted over two years.

III. STAFF COMMENTS

This project represents a fortuitous opportunity for ARB to resolve several significant questions regarding aerosol pollution from motor vehicles. The previous work done by these investigators gives them immediate knowledge of the existing data and the sampling and data analysis technologies to be used. Continuation of a line of research previously developed in close cooperation with ARB is also reflected in the focused nature of the proposal – the ATOFMS technology and the previous work are well known to ARB staff, since we were actively involved in it. This situation allows the proposal to focus on specific research questions without the necessity of re-presenting the "nuts and bolts" of the ATOFMS or the studies on which this research would build. Overall, this project is much more highly integrated than would be the case if the previous work were only available to the investigators second hand (*i.e*. through published literature).

Technologically, the experimental work in the Core Project has two important strengths. First, by using real time data collection, it is significantly more cost effective than would be the case if conventional "off line" chemical analyses were required. Second, real time data permit adaptive experimentation since unexpected results are available for immediate analysis and response. This provides benefits both through experimental setup cost savings and in the flexibility to follow up on novel findings.

Finally, this research benefits from ongoing collaboration with NREL through cost participation from that agency to cover travel expenses for fieldwork.

IV. STAFF RECOMMENDATION

Staff recommends the Research Screening Committee approve this proposal, including the Core Project and all three Optional Tasks, for a total amount not to exceed $200,653, subject to inclusion of appropriate additions and revisions in response to the staff comments, and any changes and additions specified by the Committee.