SESSION II

Diesel Engine Issues and Challenges

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DIESEL AND GASOLINE LIGHT TRUCK EMISSIONS: "THE REST OF THE STORY"

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ABSTRACT

What is the impact on ambient air quality in major metro areas if a significant share of light truck gasoline engines are displaced by diesels? This is a systems question that requires a systems analysis. Such an analysis is presented for two LDT4 light trucks; one powered by a gasoline engine and the other by a diesel. Vehicle plus upstream emissions are accounted for and a total environmental cost per 1,000 miles is estimated for each truck using damage values per ton for each pollutant. Tailpipe, evaporative, fuel processing upstream, atmospheric secondary reactions, road dust and greenhouse gases, are included.

REFERENCES

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2. Methods of Valuing Air Pollution and Estimated Monetary Values of Air Pollutants in Various U.S. Regions, M.Q. Wang,, D.J. Santini, S.A. Warinner.

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L.D. TRUCKS' FUTURE POWER PLANT

- 50% MORE MPG
- BETTER DRIVEABILITY
- MORE TORQUE, PULLING POWER
- HIGHER RESALE VALUE
- LONGER LIFE

(BUT, ITS A DIESEL)

BIG IMAGE PROBLEM

- CUSTOMER PERCEPTION
- ENV./HEALTH CONCERNS

IS THIS IMAGE ACCURATE?

HOW DOES DIESEL IMPACT:

- **HUMAN HEALTH COST**
- **LOCAL AND REGIONAL ENVIRONMENT**
- **GLOBAL CLIMATE CHANGE**

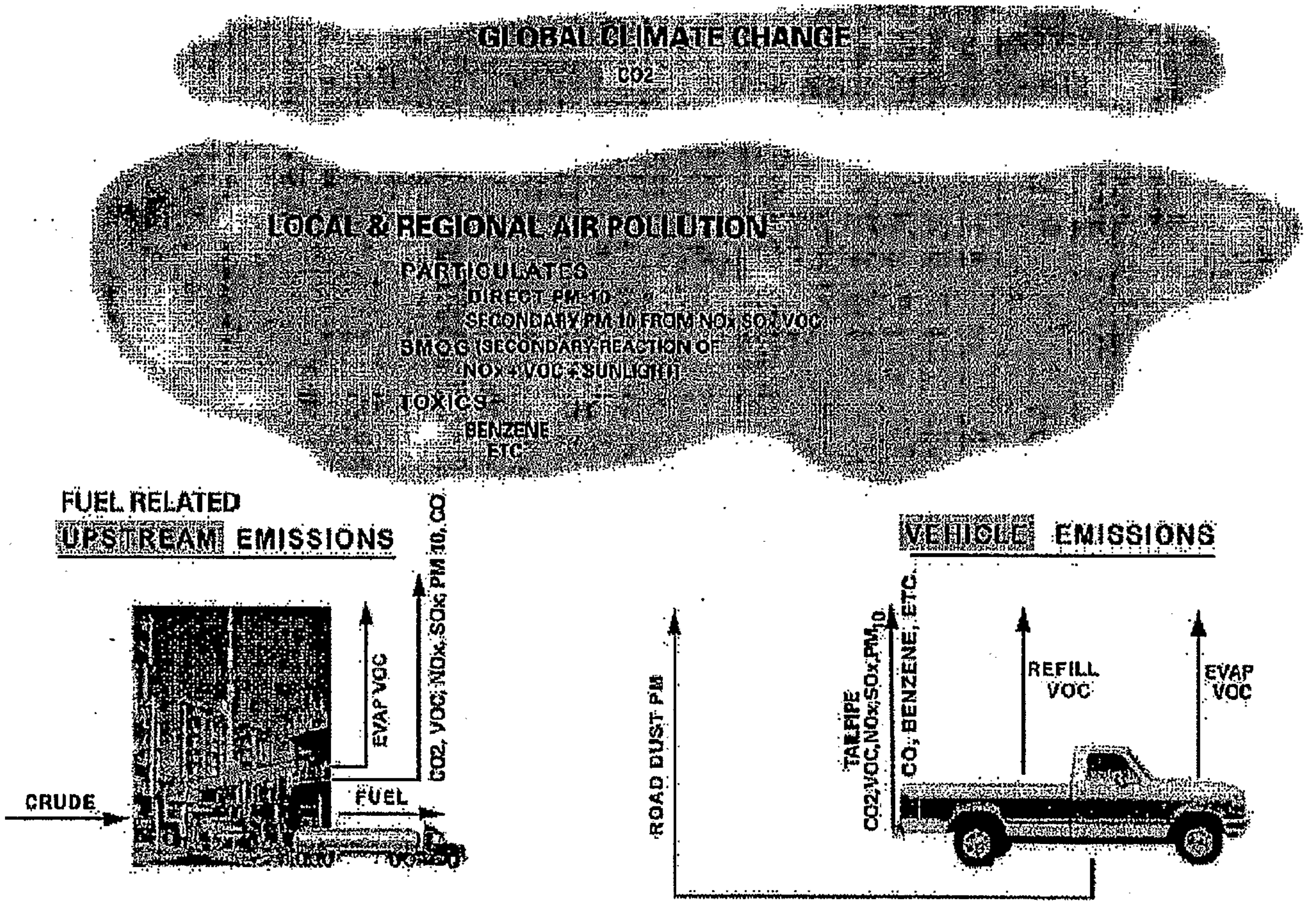
HOW CAN WE EVALUATE AND COMPARE ?

HOW CAN WE MAKE BEST USE OF THE EXISTING STUDIES AND DATA ?

NEED:

<u>SYSTEMS LOOK</u> INCLUDING:

- TAILPIPE EMISSIONS
- VEHICLE EMISSIONS
- ATMOSPHERIC REACTIONS
- UPSTREAM EMISSIONS



VEHICLE & UPSTREAM EMISSIONS, PLUS SECONDARY REACTIONS CONTRIBUTE TO LOCAL AND REGIONAL AIR POLLUTION AND GLOBAL CLIMATE CHANGE

THE SOCIAL COST OF THE HEALTH EFFECTS OF MOTOR-VEHICLE AIR POLLUTION

Report # 11 in the Series:

The Annualized Social Cost of Motor-Vehicle Use in the United States, based on 1990-1991 Data

By

Donald R. McCubbin

Mark A. Delucchi

August 1996

METHODS OF VALUING AIR POLLUTION AND ESTIMATED MONETARY VALUES OF AIR POLLUTANTS IN VARIOUS U.S. REGIONS

by

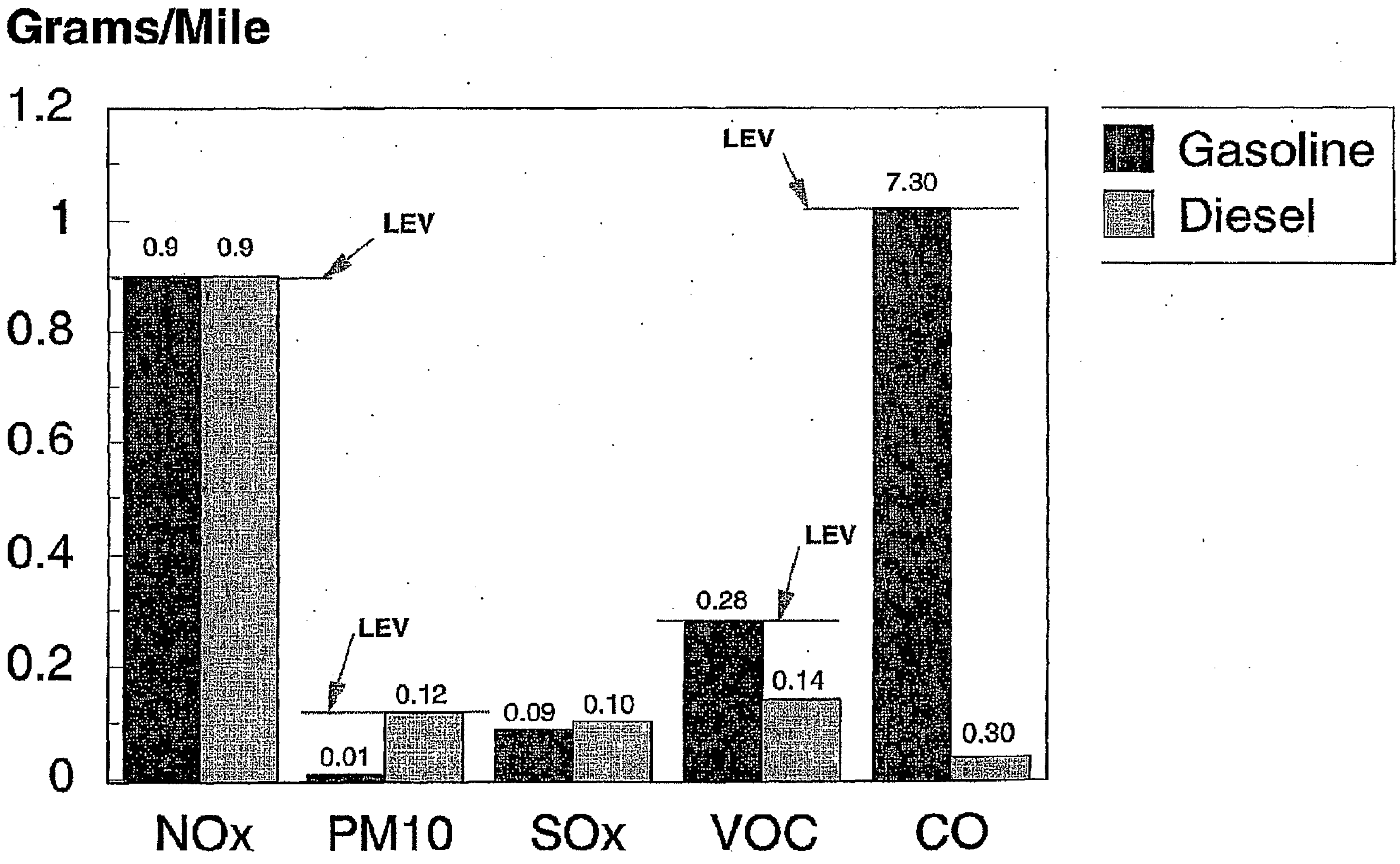
M.W. Wang, D.J. Santini, and S.A. Warinner

THE CLIMATE CHANGE ACTION PLAN

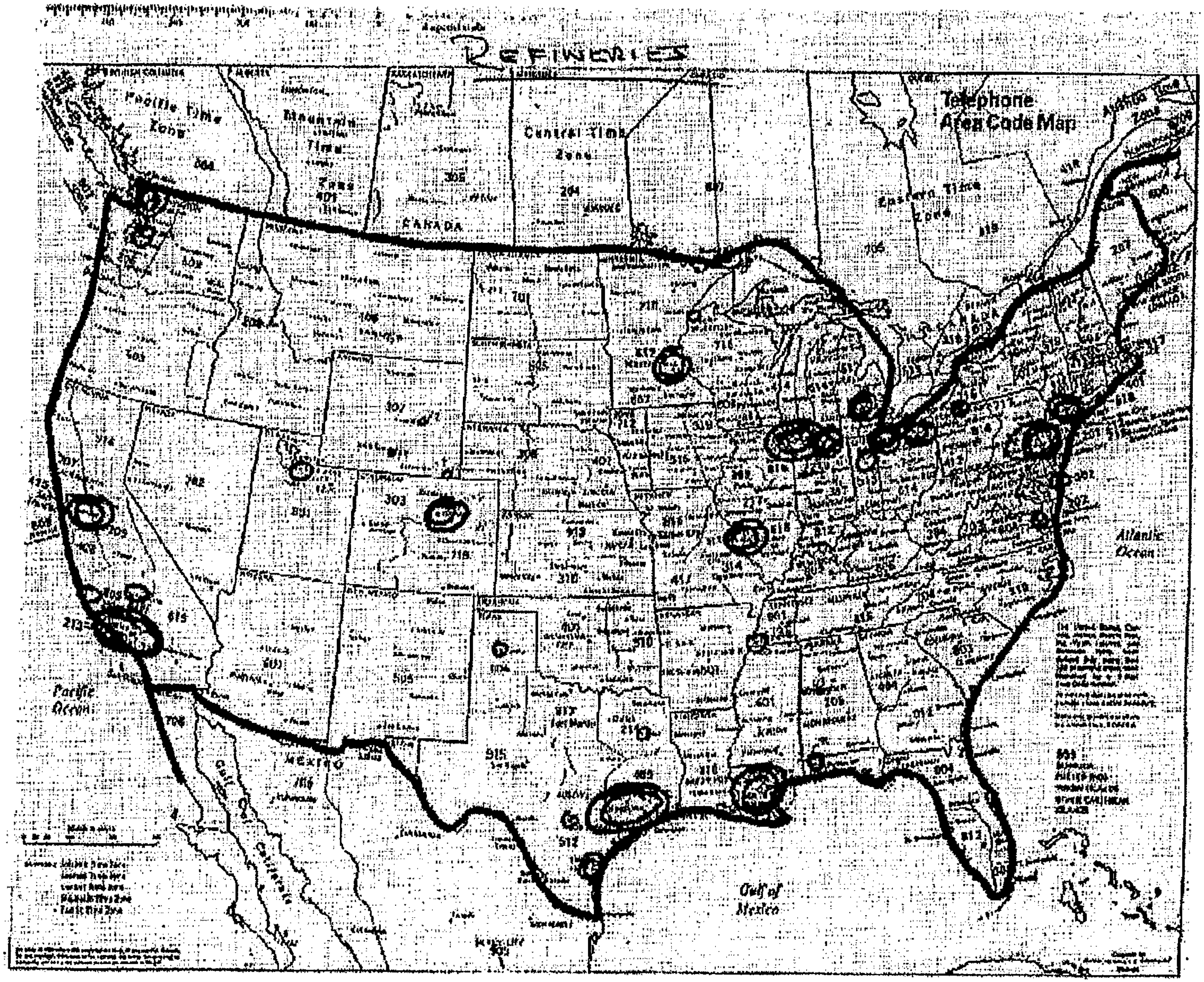
President William J. Clinton
Vice President Albert Gore, Jr.

October 1993

ASSUMED LDT4 TAILPIPE EMISSIONS



REFINERIES


Telephone Area Code Map
Pacific Time Zone
Mountain Time Zone
Central Time Zone
Eastern Time Zone
Atlantic Time Zone
CANADA
MEXICO
Pacific Ocean
Atlantic Ocean
Gulf of Mexico
Gulf of California

$$\underset{\substack{\uparrow\\ \text{vehicle +}\\ \text{upstream}}}{\mathrm{g/mi}} \times \underset{\substack{\nwarrow\\ \text{damage based}\\ \text{cost for each}\\ \text{emission (direct}\\ \text{+ secondary)}}}{\mathrm{\$/kg}} \times \mathrm{1kg/1000g} = \underset{\substack{\uparrow\\ \text{env. / health effects}\\ \text{cost per 1000 mi}}}{\mathrm{\$/1000\ mi}}$$

EMISSIONS VALUE SUMMARY, $ / KG

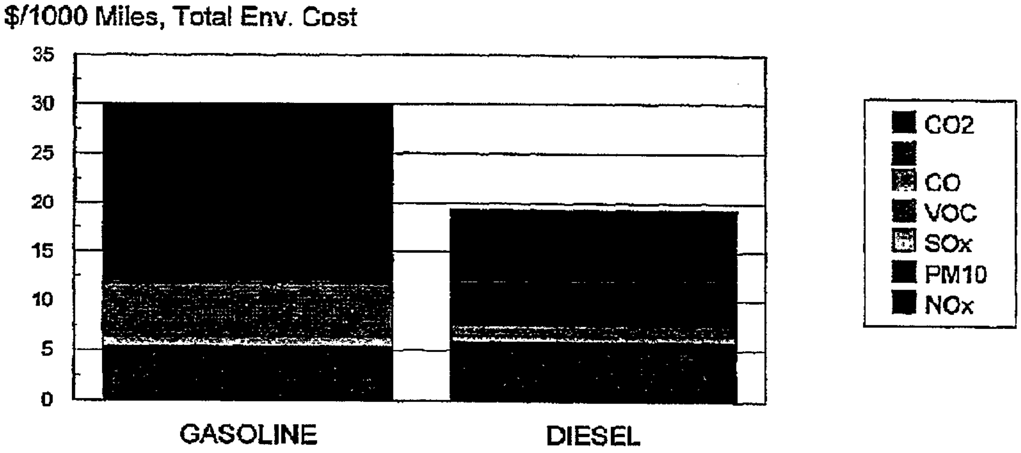
STUDY> EMISSION	ARGONNE AVERAGE	UC DAVIS LOWER BOUND	UC DAVIS UPPER BOUND
NOx	5.31	1.50	22.08
TAILPIPE PM (100% PM 2.5)	7.16	14.81	225.36
SOx	3.19	4.40	35.28
VOC	2.66	0.13	1.25
CO		0.01	0.01
NOx+VOC		0.02	0.12
UPSTREAM PM (69% PM2.5)	7.16	13.14	162.69
PVD ROAD DUST (25% PM2.5)		1.05	37.13
TIRE WEAR (25% PM2.5)		1.05	37.13
BRAKE WEAR (42% PM2.5)		1.15	54.20
CO2	0.017	0.0034	0.034

EMISSIONS VALUE SUMMARY $/KG

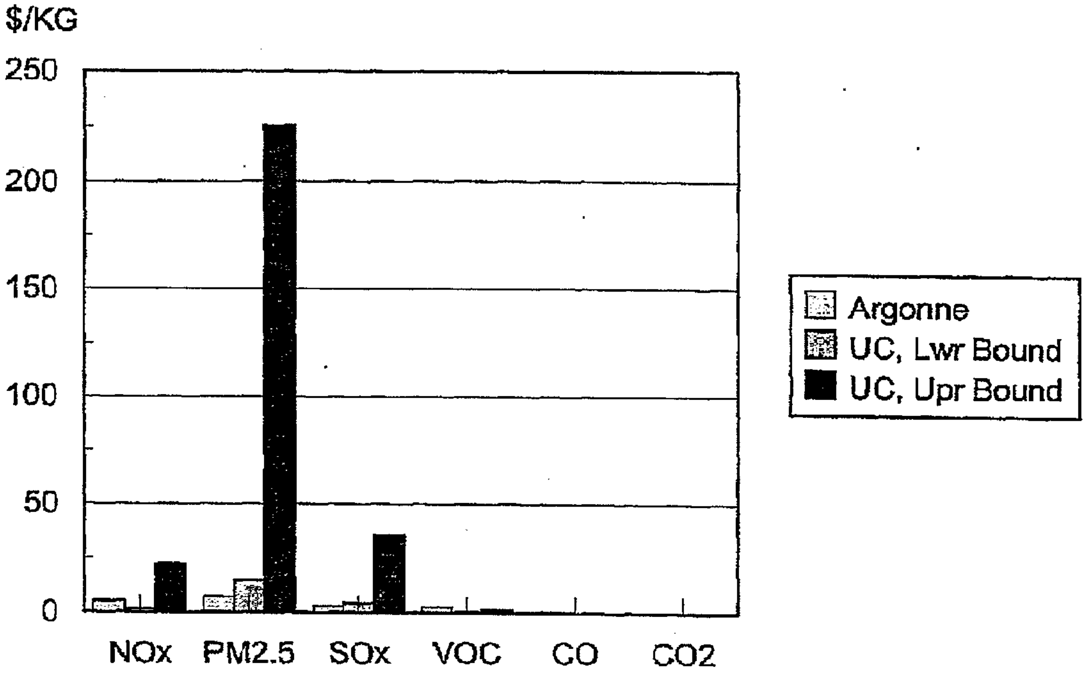
STUDY> EMISSION	**ARGONNE S UMMARY AVER GE**
NOx	5.3
PM 10	7.1
SOx	3.1
VOC	2.6
CO	0

RESULTS WITH ARGONNE STUDY AVERAGE EMISSION VALUES

Tailpipe Emissions: Nominal LEV, LDT4
Upstream Emissions: Refinery, Storage, Transport, Nat'l Averages

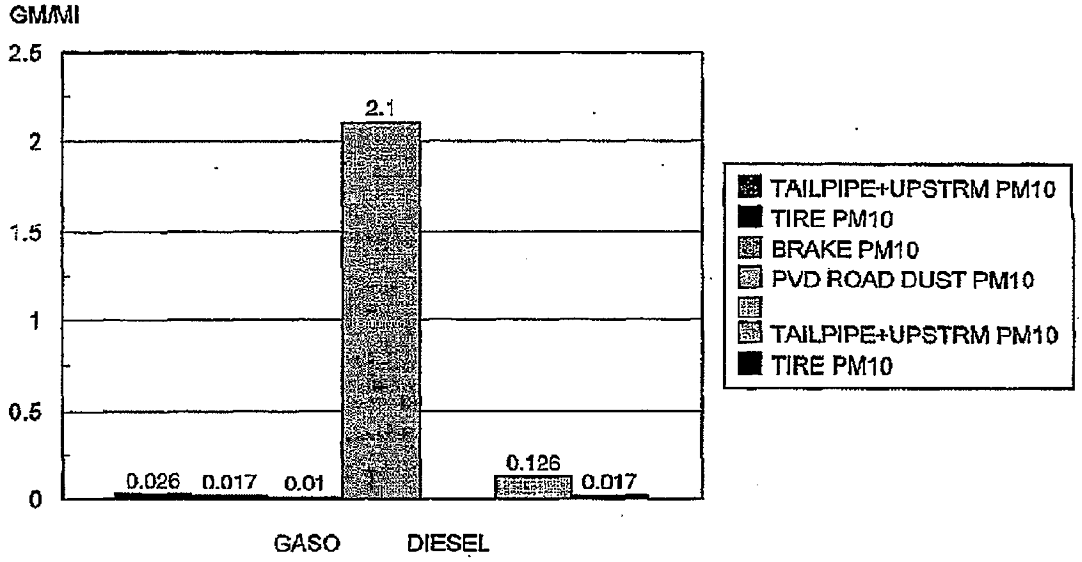


ENV. / HEALTH EFFECTS COST, $/KG



TAILPIPE, TIRE & BRAKE, UPSTREAM PM10

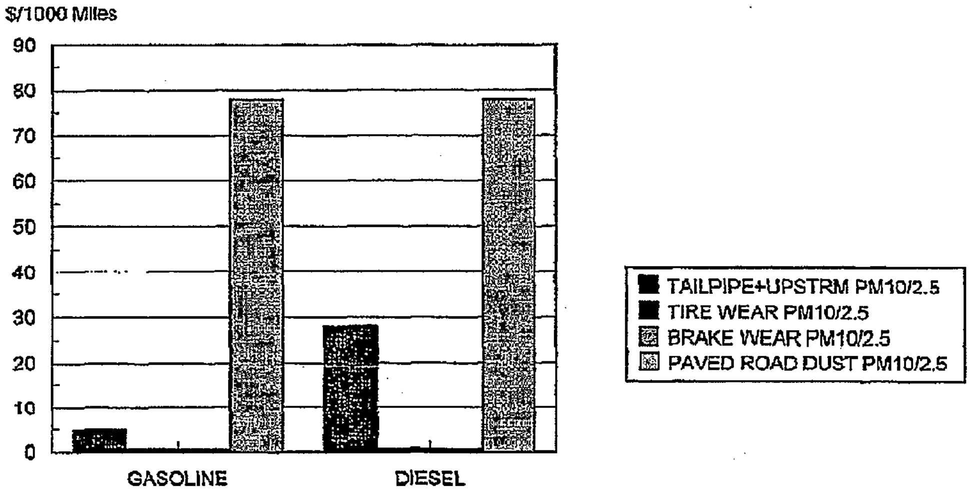
EACH HAS DIFFERENT SIZE DISTRIBUTION AND COMPOSITION



RESULTS W/ U OF CA, DAVIS STUDY EMISSION VALUES (UPPER BOUND)

Tailpipe Emissions: Nominal LEV, LDT4

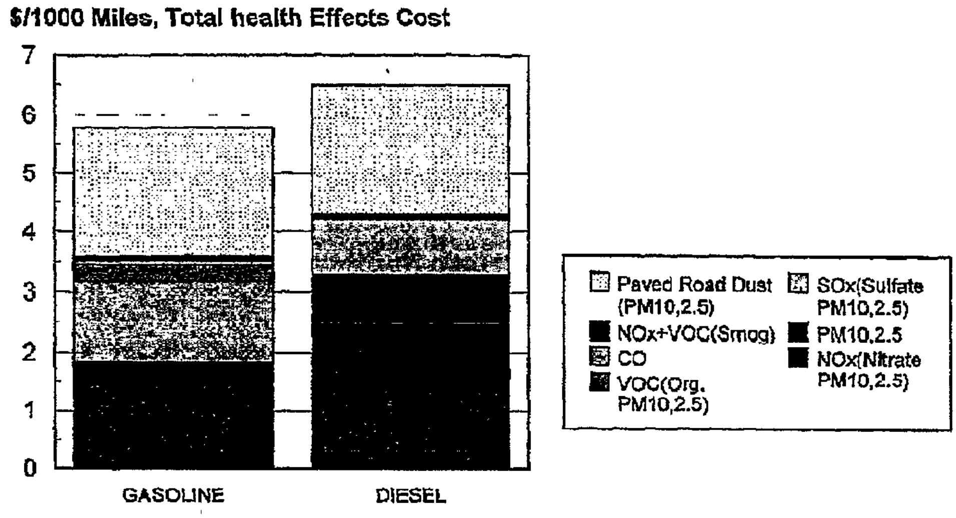
Upstream Emis: Refinery, Storage & Transport in Major Metro Areas



RESULTS W/ U OF CA, DAVIS STUDY EMISSION VALUES (LOWER BOUND)

Tailpipe Emissions: Nominal LEV, LDT4

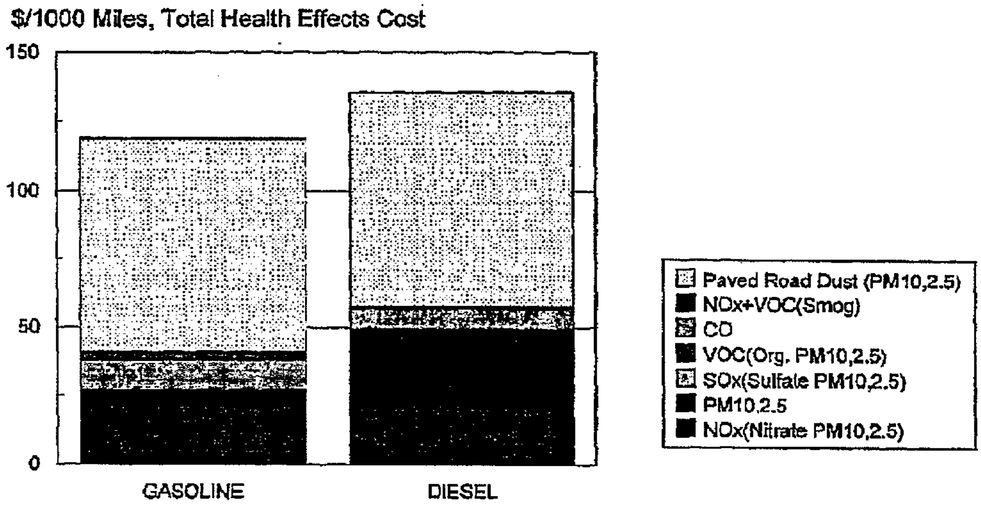
Upstream Emis: Refinery, Storage & Transport in Major Metro Areas



RESULTS W/ U OF CA, DAVIS STUDY EMISSION VALUES (UPPER BOUND)

Tailpipe Emissions: Nominal LEV, LDT4

Upstream Emis: Refinery, Storage & Transport in Major Metro Areas



SUMMARY:

WITH A SYSTEMS LOOK AT MAJOR METRO AREAS,

THE NEXT GENERATION OF DIESEL ENGINES
APPEARS TO BE AS ENVIRONMENTALLY FRIENDLY AS
THE NEXT GENERATION OF GASOLINE ENGINES,

AND YET PROVIDE GREAT ADVANTAGES IN
GREENHOUSE GAS EMISSIONS AND FUEL USE.

ADDENDUM

Since the 1998 DEER workshop at Castine we have obtained better vehicle emission level estimates than the LEV vehicle level estimates used in the foregoing Castine presentation.
For a simpler, more realistic and more accurate analysis of the vehicle emissions only, for current Tier 1 Light Trucks, we can use the following output of EPA's MOBILE5. MOBILE5 estimates the "average over in-use life", meaning the overall average gm/mi for the overall average vehicle life using EPA's assumptions on vehicle life distribution, inspection and maintenance, fuel variations, aftertreatment degradation, seasonal effects, etc. This was done for light trucks in the GVW range of 6000 to 8500 LB (LDT3&4), for both a diesel powered light truck and a comparable gasoline engine powered one (i.e. a current GM 6.5 l turbo diesel in a Chevrolet 1/2 ton 4X4 pickup and the same truck powered by a Chevrolet 350 gasoline engine).

Current Technology LDT (6,000 - 8,500 GVW) Emission Factors

Average Over In-use Life (g/mi)

	Gasoline			Diesel		
	Summer	Winter	Annual	Summer	Winter	Annual
Exhaust THC	1.93	3.32	2.62	0.71	0.71	0.71
Hot Soak & Diurnal TH	0.20	0.01	0.10	0.00	0.00	0.00
Resting Loss THC	0.02	0.00	0.01	0.00	0.00	0.00
Running Loss THC	0.41	0.00	0.21	0.00	0.00	0.00
Refueling THC	0.02	0.03	0.03	0.00	0.00	0.00
Total THC	2.58	3.35	2.97	0.71	0.71	0.71
Exhaust CO	24.49	40.94	32.72	1.61	1.61	1.61
Exhaust NOx	1.89	2.36	2.12	1.24	1.24	1.24
Exhaust SO2	0.086	0.086	0.086	0.119	0.119	0.119
Exhaust PM10	0.015	0.015	0.015	0.109	0.109	0.109
Tire Wear PM10	0.008	0.008	0.008	0.008	0.008	0.008
Brake Wear PM10	0.013	0.013	0.013	0.013	0.013	0.013

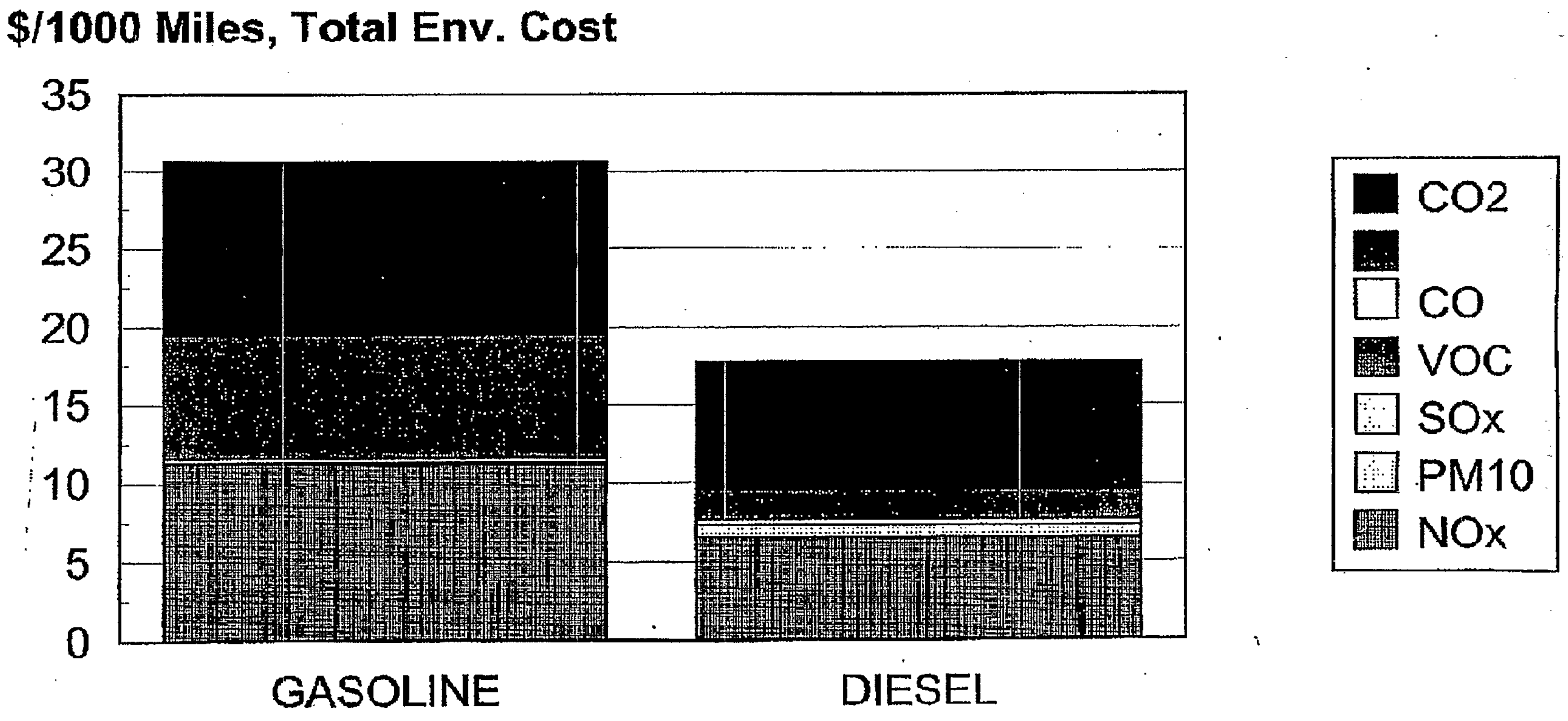
Notes:

1. Assumed national average of 28% enhanced I/M and reformulated gasoline; 14% basic I/M and non-reformulated gasoline; and 58% no-I/M and non-reformulated gasoline.
2. HC (expressed as total HC), CO and NOx from MOBILE5a; SO2 and PM from PARTS.

Combining the above overall average gm/mi for each vehicle emission with the environmental/health effects damage value for each emission, yields the following barcharts. Note that when these more realistic vehicle emission levels are used, diesel has lower impact than gasoline even without consideration of upstream emissions. Furthermore, diesel still shows lower impact even with use of UC Davis' extreme particulate values and without consideration of CO2.

RESULTS WITH ARGONNE STUDY AVERAGE EMISSION VALUES, & EPA MOBILE 5 EMISSION LEVELS

Vehicle Emissions w/o Tire & Brake Wear
No Upstream Emissions



CATERPILLAR'S LIGHT TRUCK CLEAN DIESEL PROGRAM – TECHNOLOGIES TO MEET FUTURE HSDI EMISSIONS REGULATIONS

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Caterpillar, Inc.

INTRODUCTION

Caterpillar Inc.'s Engine Research department is involved in a program to develop technologically advanced prototype engines for the Sport Utility Vehicle (SUV) and Light Truck (LT) vehicle markets. Meeting the objectives of this program, which is co-funded by the Department of Energy's Office of Heavy Vehicle Technologies (DOE- OHVT) office, will result in vehicular powerplants capable of delivering significant fuel savings in this fast-growing consumer market.

Light Trucks (pickups and sport utility vehicles < 8500# GVW) now comprise over 40% of new vehicle sales in the U.S. and well over half of the new vehicle fuel consumption levels. At the same time, higher technology diesel engines are making dramatic gains in the higher priced fuel markets such as Europe. Automakers and the DOE feel that there is a demand waiting to be satisfied with an advanced diesel engine for LT's and SUV's (Fig. 1) in the U.S. market. Converting these vehicles to advanced, ultra low emitting diesel engines would improve their tank-miles-per-gallon fuel economy by at least 50%. This could easily translate into a fuel savings of 0.7 MBPD (million barrels per day), or roughly 20% of our nation's total oil import levels.

The challenge in DOE's Light Truck program is to maintain the inherent fuel economy benefits of the diesel engine powerplant while meeting the stringent gaseous emissions requirements as defined in EPA legislation. Caterpillar's Light Truck Clean Diesel (LTCD) program will deliver prototype vehicles in 2002 demonstrating a 50% fuel economy improvement over 1997 gasoline powered vehicles while complying with EPA's Tier II emissions regulations.

CATERPILLAR'S LTCD PROGRAM

Caterpillar is teamed with Ford Motor Company on this 5 year program to develop prototype vehicles demonstrating the target fuel economy and emissions levels (Fig. 2) through the use of significantly advanced compression ignition, direct injection (CIDI) technologies. In 2002, the development team will deliver prototype vehicles with CIDI powerplants exhibiting performance specifications approximating those shown in Fig. 3. These powerplants will meet the requirements of the program and provide excellent driveability due to the excellent low-end torque characteristics inherent in CIDI engines.

The program (Fig. 4) consists of five primary tasks. Current activities focus on development tasks 1.1 – 1.3 which include defining specifications for the technologically advanced vehicle and powertrain, developing parallel technologies necessary to meet the program goals and demonstrating these technologies on mule HSDI engines. Prior to their incorporation into the LTCD demonstration engines, each technology will pass through a rigorous downselection process involving assessments against performance, cost and feasibility goals.

Due to the anticipated significant challenge of meeting the Tier II emissions goals, the scope of Caterpillar's LTCD program was modified in early 1998 to augment development efforts in many of the technology areas. One of the primary augmented focus areas was emissions and aftertreatment. Inability to meet the requirements of Tier II emissions legislation would be unacceptable, even if the vehicles were to meet the 50% fuel economy improvement goal.

As the program scope was modified, the development team established a second phase of emissions development (Phase 2) and pulled-forward the date for demonstration of the original emissions goals (Phase 1.) Phase I emissions will be demonstrated in-vehicle and phase 2 emissions will be demonstrated on an engine transient emissions test bed. Both phases will target the 50% fuel economy goal.

EMISSIONS PROGRAM

Several drivers are mandating a more focused effort on emissions reductions technology development at Caterpillar. These include the general long-term emissions and aftertreatment research activities currently underway, the recent off-cycle emissions issue and the desire to leverage successful technology across the entire Caterpillar product line. In particular on the LTCD Program, the renewed technology emphasis coupled with aggressive transient emissions targets have led to a two-phase emissions program.

In Phase 1, emissions targets are 0.5 g/mile NOx and 0.05 g/mile particulate matter (PM) over the US FTP-75 transient emissions cycle. These targets are for the contractually obligated F-150 vehicle demonstration at the end of the LTCD Program. In addition, a Phase 2 program is planned with emissions targets of 0.05 g/mile NOx and 0.01 g/mile PM (LEV II). Achievement of these emissions levels will only be attempted over the FTP-75 transient emissions cycle in an engine dynamometer demonstration.

The difficulty of achieving these goals in a vehicle with a ~5000 lb. test weight is recognized from the outset. However, the stringent targets will drive emissions and aftertreatment technology development as applied to HSDI Diesel engines to new levels.

A schematic showing the major general elements of the Phase 1 and 2 programs is presented in Fig. 5 with the more detailed paths given in Fig. 6. The Phase 1 program is underway in Q4 '98, and the majority of the Phase 2 program begins in Q3 '99, although several activities have already been started. The baseline engine is a Perkins 3.0L V6 with the Bosch VP-44 fuel system, electronic EGR system and an oxidation catalyst. FTP-75 emissions data were obtained in a test vehicle at Light Truck/SUV road load conditions. The cycle NOx and particulate emissions were 0.65 g/mile and 0.1 g/mile, respectively as indicated in Fig. 6 and are the starting emissions levels for the Caterpillar LTCD Program.

The first major emissions reduction technology to be developed is in the area of fuel injection systems. The Caterpillar HEUI-B fuel system will be implemented in the LTCD engine at an early stage. This fuel system offers cam-independent flexible injection control (small pilot quantities, split injections, adjustable front-end rates, etc.) that will provide approximately 20% lower particulate emissions levels than the existing VP-44 fuel system. Air systems technology development is also a strong emphasis of the Caterpillar LTCD Program. Variable geometry turbocharging along with a fast acting electronically controlled EGR system capable of tight AFR control are major program components. Careful control of these components will be accomplished using model-based mapping with feedback control. The tighter transient AFR control will allow the EGR system to reduce particulates by an additional 25% with a further 5-15% reduction in NOx emissions levels. Combustion optimization techniques (bowl/swirl/fuel spray matching) using CFD to guide an experimental test program should lead to further 10% reductions in NOx and PM.

Depending upon which Phase 1 path is chosen (see Fig. 6a), a more conventional aftertreatment technology will then be required to achieve the emissions targets. The first option is an active lean NOx catalyst with supplemental HC injection. NOx conversion efficiencies of 30% with this type of system have been advertised by several major catalyst OEMs. However, an FTP-75 cycle conversion efficiency of 18% should be adequate to achieve Phase 1 emissions targets, assuming the other systems achieve reductions as indicated. Alternatively, with a different injection timing and EGR tune for NOx control, an advanced Diesel oxidation catalyst capable of 20% solid particulate reduction could be used. Either of these paths requires overall NOx and PM reductions of 23% and 46%, respectively, compared with the existing baseline engine. This level of reduction will require significant technological advances over the next two years, but is feasible based on existing information.

The Phase 2 emissions targets mandate substantially improved performance from the fuel and air systems (engine out reductions of 44% NOx and 61% PM). In addition, advanced aftertreatment solutions capable of further 70-85% reductions in NOx and PM will be required. Overall reductions from the baseline engine of approximately 90% for both NOx and PM will be required to meet the stringent Phase 2 targets. Collaborative programs between Caterpillar and catalyst OEMs and consulting firms are underway to develop these advanced aftertreatment systems.

Three primary paths have been identified as indicated in Fig. 6b. The first uses diesel fuel with a regular (~300 ppm) sulfur level. The technology with the most potential here is a system using a urea-SCR system combined with a catalyzed diesel particulate filter. The second path requires low sulfur (<10 ppm) fuel but allows for NOx trap and continuously regenerating trap (CRT) technology. Finally, a third path using alternative technologies has been identified. Here, advanced systems such as a partial oxidation fuel reformer, non-thermal plasma (with catalyst) and homogeneous charge compression ignition combustion are being investigated as enabling technologies.

Clearly there are numerous inhibitors to implementation of each of these advanced diesel emissions reduction technologies in light truck and sport utility vehicle applications in the future. Foremost are the time, cost, and technology hurdles associated with their development. Lower temperature SCR catalysts and better NOx trap regeneration schemes are being investigated, but must be further developed. Durable/advanced sensors for urea and NOx must be developed for use in feedback control and anti-defeat applications.

The highly interactive nature between the fuel injection, turbocharger, EGR and aftertreatment systems will mandate improvements in microprocessor speed and controls algorithms. The increasing stringency of the legislated emissions levels makes the task more difficult to achieve. In addition, for several of the aftertreatment technologies, lower fuel sulfur levels will be required to improve catalyst performance and prevent poisoning. Finally, for SCR aftertreatment systems, infrastructure improvements will be needed to support the distribution of urea.

CONCLUSIONS

DOE's light truck program offers significant challenges but also has the potential to bring tremendous value to this country through large reductions in fuel usage in the light truck vehicle market and through significant reductions in one of the primary greenhouse gases CO2. The diesel engine powerplant offers the greatest potential for meeting the fuel economy goals in this program in the shortest amount of time. Meeting Tier 2 emissions requirements, while still maintaining a significant fuel economy advantage, will be the significant challenge. Caterpillar and other U.S. diesel engine manufacturers have continuously improved the emissions and performance of their products over the past 20 years, reducing regulated NOx emissions by 70 percent and lowering particulate emissions by 90 percent. During that same period, diesel engine fuel efficiency has improved over 25 percent. These same, and even more advanced, technologies when incorporated into light duty platforms will result in products that meet customer expectations and provide significant value to the country.

POLARIZED LIGHT SCATTERING FOR DIESEL EXHAUST PARTICULATE CHARACTERIZATION

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ABSTRACT

The polarization properties of scattered light are being exploited to determine the size distribution and composition of the exhaust particulate from diesel engines. The feasibility of the approach was established through preliminary measurements using a polarization-modulated angle-scanning nephelometer (POLNEPH) to completely characterize the intensity and polarization of the scattered light. The size distribution and complex index of refraction of the particles were determined from the data by simultaneously fitting measured the angle-dependence of three or four scattering transformation matrix elements with Mie scattering calculations. Based on the results of the preliminary measurements a new instrument (Scatterometer) was designed specifically for rapid *in-situ* determination of particle size and optical properties of modern diesel engines.

INTRODUCTION

This research is directed towards developing a new method and instrument to characterize diesel exhaust particles by the measurement of the intensity and polarization of light scattered from the exhaust stream. In particular, we are developing a *real-time* light scattering instrument to measure the size and optical properties of diesel particulates. This instrument development program will enable the study of the exhaust particle characteristics as a function of engine running conditions, fuel composition, and exhaust after treatment processes. It will also allow the study of various post-combustion factors such as aftertreatment technology, and dilution conditions (dilution volume ratio, injection techniques, time, and relative humidity) on the particle size distribution and content.

We are undertaking this research because of the increased importance of small particles released into the environment. Accurate, real-time measurements of particulate matter (PM) emissions are essential for engine development and monitoring evaluation of after-treatment technologies and emissions compliance. Airborne particulates are known to constitute a major human health risk[1]. Recent epidemiological studies report that particles with diameters of less than 2.5 μm are most dangerous - a fact of particular relevance to emissions from modern diesel engines that are in this range. Further, these particles are known to have a profound effect on visibility in Class I areas and constitute a major source of carbonaceous particulates in populated areas.

Diesel particulate pollutants consist primarily of soot resulting from incomplete combustion of the fuel[2,3]. Concern about the health effects of airborne particulate matter led the federal government, in 1971, to regulate such pollutants and in 1987, by basing the standard on the mass of particles with diameters $d < 10$ μm, further tightened the regulation. Recent epidemiological studies[1,4] have indicated that the most dangerous particles have $d < 2.5$ μm and a more stringent standard has been promulgated by EPA. These health effects are of special pertinence to diesel engine emissions. Particulate emissions in diesel exhausts that range in size from small 10-30 nm spheres to clusters (agglomerates) of these spherules with diameters up to 10 μm, are a major source of the most hazardous aerosols[5]. Furthermore, transportation-generated particles have several special problems[2]. They are emitted close to ground level in areas of high population density and, due to their small size, have long lifetimes. They also can transport heavy metals and other pollutants, which may be absorbed on the surface of the particles, and deposited deep in the lungs.

Current measurement methods are inadequate to fully characterize these particulates. Conventional methods that rely on the exhaust opacity (smoke meters) do not work with newer direct-injection diesel engines because the total mass and particle size in the emissions do not cause sufficient opacity to provide reliable measurements. Filter methods are slow and cumbersome and give only total mass information. Multistage impactors require time-consuming analysis methods and most do not accurately measure the smallest particles in the exhaust. Advanced particle characterization instruments such as the Scanning Mobility

Particle Sizer generally require long sampling times to obtain size distribution information and therefore do not characterize transient changes in particle characteristics. In addition, these methods provide little or no information about particle composition or its carbon content.

THEORETICAL BACKGROUND

To better understand the approach used in this work a brief description of light scattering formalization is given here. The intensity and polarization of any beam of light can be described by a 4-element Stokes vector, *I*, the components of which measure the following:

I = total intensity of light,
Q = ±90° polarization,
U = ±45° polarization,
V = circular polarization.

Light scattering may be described theoretically by the transformation of the 1 x 4 Stokes vector by a 4 × 4 Mueller matrix, $I' = M I$, where the M is the Mueller scattering matrix and I' the Stokes vector of the *scattered* light. The complete *angle dependence* of the matrix elements represents all the information available from light scattered without wavelength shift[6]. The approach used in this work relies on matching measurements and theory of the angle dependence of several elements of the Mueller matrix. The matrix is determined by the characteristics of the scatterers, including their size, structure, symmetry, orientation, and complex refractive index m ($m = n - i\kappa$), where n is the real (refractive) and κ the imaginary (absorptive) part of the refractive index. The Mueller matrix associated with a suspension of aerosol particles can be used to describe and quantify the effects of particles on polarized light. In general, eight elements of the Mueller matrix (normalized by the total intensity; designated as $S_{xy} = S_{xy}/S_{11}$) are non-zero for aerosols: S_{11}, $S_{12} = S_{21}$, S_{22}, S_{33}, S_{44}, and $S_{34} = - S_{43}$.

Determination of Size and Refractive Index for Spherically-Symmetric Particles

The size, refractive index and shape information for a distribution of particles from the observed scattering cannot in general be obtained by direct inversion of the Mueller matrix. It is necessary, therefore, to calculate the scattering from assumed ensembles of particles using a variety of models based on the scattering system and compare the results with the experimental data. If the particles are spherical or nearly so, then the scattering of such an ensemble can be calculated rigorously using Mie scattering models[6,7]. It is only necessary to calculate 4 elements of the scattering matrix for spherical particles - all others being zero, equal to, or the negative of the four calculated, and S_{22} is unity. Particles may be considered spherical when S_{22} is greater than 0.9 at all angles[8]. If the particles are spherical, agreement between observation and calculations based on the Mie model is excellent[7,9].

Preliminary S_{22} measurements of the exhaust from modern direct injection diesel engines indicate that they generally may be treated as spheres (although this is not necessarily true for older diesel engines). Therefore Mie calculations were used to predict the resulting angle dependence of the Mueller matrix. The measured scattering is compared with that predicted by Mie calculations and the input parameters to the model are adjusted until the agreement is optimized. There is no exact solution to this problem; the fitting procedure we employed matches the four relevant matrix elements simultaneously. It is performed iteratively with a computer code developed at LBNL based on the Levenburg-Marquardt optimization technique[10].

The code uses the type of size distribution, mean, minimum, and maximum radii and the complex refractive index as input parameters and iterates to the optimal solution based on the minimization of χ^2, the "goodness of fit" parameter. In the calculations the size probability distribution (generally log normal produced the best fits) and minimum and maximum particle radii were fixed and other inputs were treated as free parameters. These fitting programs run rapidly on modern PCs and can done at a near real-time basis. Thus the procedure is measure the light scattering for several matrix elements at fixed number of angles and apply the optimization technique to rapidly determine the size distribution and complex (refractive and absorptive) indices of refraction. These numbers can be used to determine an effective size, amount of soot present and degree of compactness of the particles.

Non-spherically Symmetric Particles

Theoretical analysis of the more general case of non-spherical particles[11] is more difficult but was considered important in certain cases. Namely older diesel engines produce larger particles that deviate more significantly from spherical symmetry especially as measured in fractions of the wavelength of light. In cases when S_{22} was measured to be significantly less than unity an alternative approach to modeling light scattering from non-spherical diesel soot particles was developed. In this case the particle model represents the particle as an agglomeration of primary soot nanoparticles. These particles or clusters are generated using a random-walk procedure to build up a particle that is characterized by a fractal dimension and a radius of gyration. The number of primary soot particles that are added in the random walk procedure determines the size of the cluster (radius of gyration). The fractal dimension (degree of compactness) is calculated from the coordinates of the computer-generated particle.

Once a particle is generated using the random-walk model, the scattering is calculated from an ensemble average of these particles. Figure 1 illustrates the type of particle model generated by this procedure. It can be seen that the particle model resembles electron micrographs of soot particles. In this case the calculation is based on the coupled-dipole approximation,[12,13] that is well suited to the agglomerate particle model. The calculation is more computationally intensive and therefore required a larger computer than that needed for the Mie calculations. The calculations are carried out in parallel mode with a Cray T3E (with up to 640 processing elements available). In this case the particle size will be determined using the Levenburg-Marquardt optimization technique with look-up tables for the results.

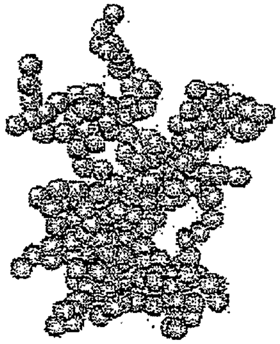

Figure 1. A model of a diesel soot particle generated by a random walk calculation.

EXPERIMENTAL APPROACH

To determine which of the Mueller scattering matrix elements are most diagnostic for characterizing diesel particle size and the most sensitive measurement configuration, extensive measurements were performed using our existing angle-scanning polarization-modulated nephelometer[7,8,9] (POLNEPH).

This instrument uses a linear polarizer and photoelastic modulator operating at 50kHz to modulate the polarization of light from a 532 nm cw Nd YAG laser. The light is passed into the sample region that contains the exhaust plume. The exhaust is directed though the sensing region and drawn off with a vacuum line to confine the stream. The scattered light is collected with a photo-multiplier tube that is mounted on the rotating arm. Before reaching the photomultiplier tube the light passes through a linear polarizer. An angle scan using POLNEPH takes about one minute. By manually selecting the modulator orientation, polarization filters, retarders, and synchronously detecting the 50 and 100 kHz component of the scattered light, all sixteen elements of the Mueller matrix from 10-170° were measured. Agreement between repeated measurements of the same matrix element was excellent. Measurements were routinely made of four elements of the Mueller matrix for comparison with results from analytical models.

RESULTS

The results of scattering measurements using POLNEPH for two matrix elements are presented in Figure 2. The total intensity (S_{11}) and the linear to circular polarization transformation (S_{34}) are shown for two engines and load conditions. The engines are a Cummins B5.9 175 MAN 6 cylinder turbo-diesel connected to an eddy current dynamometer and an Acme Motori - ADX 300 one cylinder direct injection diesel engine used in a motor-generator configuration to provide a load. The total intensity (S_{11}) is seen to vary well over an order of magnitude from the front to back scattering directions. The small diesel shows a larger change with angle for full load vs. idle conditions indicating a larger particle size for full load. The S_{34} matrix element for the small engine demonstrates considerable variation with load. This variation is one of the keys in providing unambiguous particle size information from the multi-curve fitting procedure. Calculations based on the assumption that the particles are very much smaller than the wavelength of light are also shown in the figure (Rayleigh scattering). The Rayleigh equations are often used to describe scattering from diesel particles. It can be seen that it provides a very poor fit to the data, clearly indicating that Rayleigh theory would be inappropriate for analyzing even these small particles.

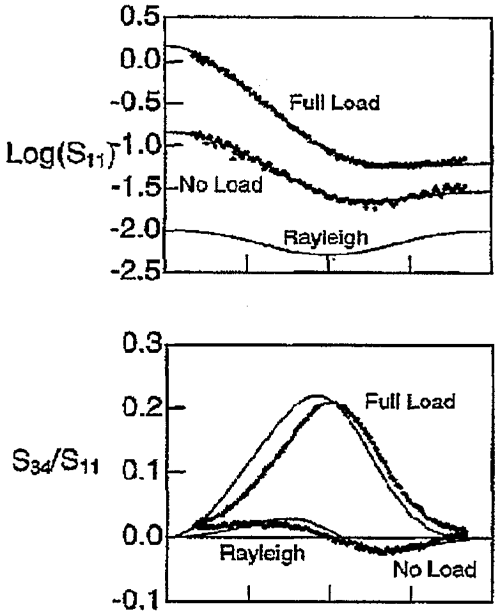

Figure 2. Above: Scattered intensity vs. angle for two diesel engines with varying loads. Below: polarization transformation matrix element S_{34} for the same load (raw exhaust, no dilution).

To illustrate the use of the data to provide size and optical properties, measurements were performed on the Cummins diesel engine using a crude dilution tunnel. The experimental and theoretical results are compared in Figure 3. In this case three matrix elements were measured and matched simultaneously by the curve fitting procedure outlined above. The solid diamonds represent the experimental data. (The curves were fit every degree, the diamonds are used to distinguish the two curves.) Figure 4 illustrates the particle size distributions determined from the data in Figure 3 displayed two ways: as mass vs. size and number of particles vs. size. It can be seen that the medians for these two curves derived from identical data are dramatically different, indicating the prevalence of large numbers of very small particles.

SCATTEROMETER DESIGN

The foregoing experiments demonstrated that three matrix elements were sufficient to unambiguously characterize the diesel exhaust particles from the engines that were measured. This information provided the basis for the design of the *Scatterometer*, an instrument that is under construction for routine diesel exhaust characterization.

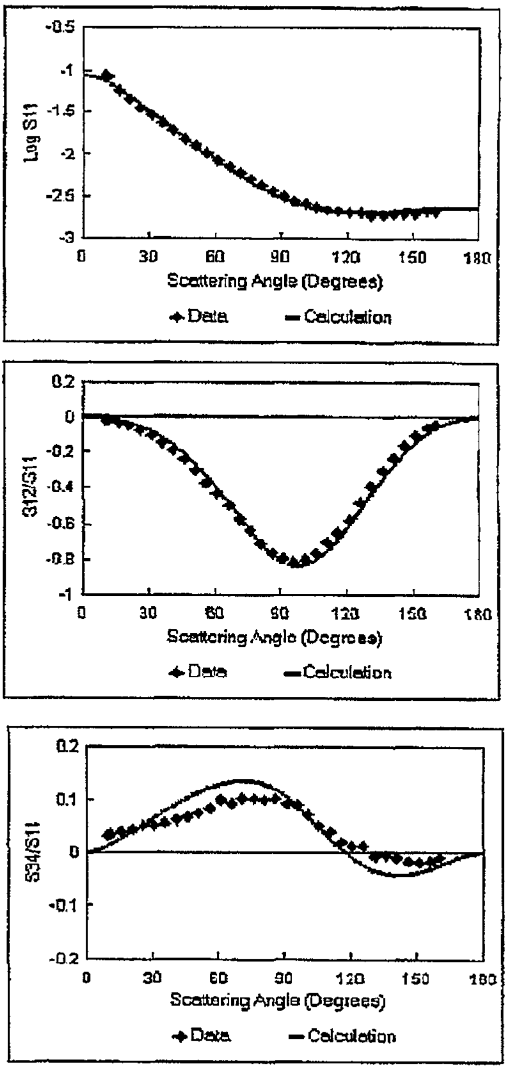

Figure 3. Experimental and calculated angle dependent matrix elements. Based on the iterative fit the size distribution gave an effective mean diameter of 32 nm for a log normal distribution of particles with standard deviation of 0.9. The effective refractive index for the particles was 1.36+0.08*i*.

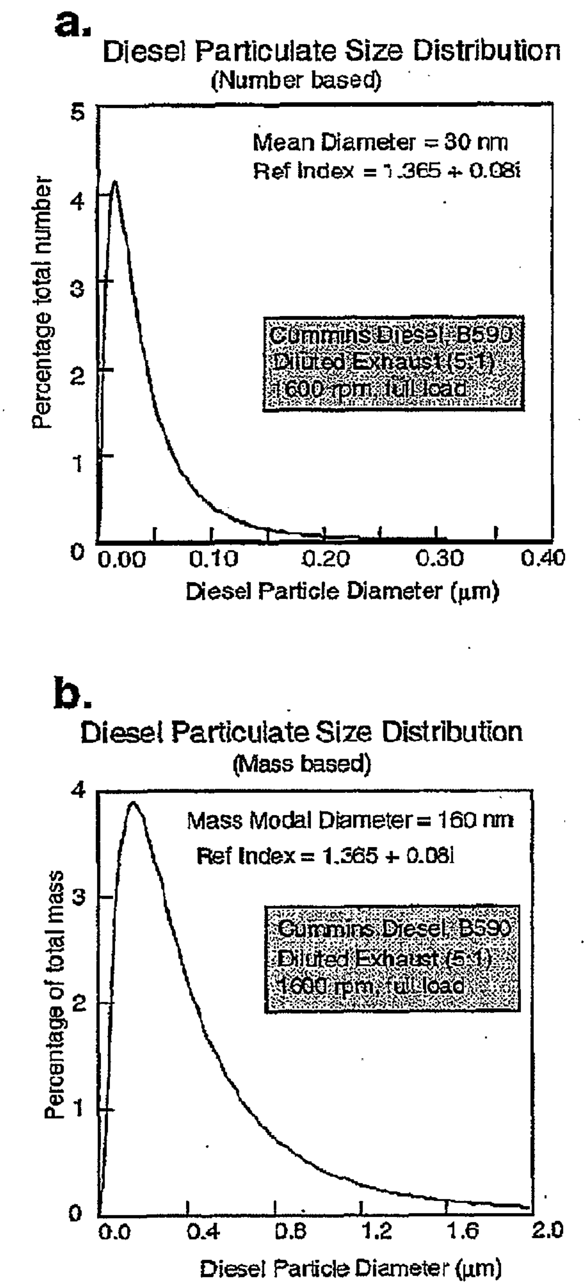

Figure 4. Particle size distributions derived from the data in Figure 3. a) Particle number vs. size b) particle mass vs. size. Please note that the scale of the x-axis changes dramatically. The entirety of figure a fits into the first division of figure b.

The Scatterometer uses the same polarization modulation and phase sensitive detection used in POLNEPH but is specialized for rapid in situ measurements of diesel exhaust stream. In the Scatterometer, instead of scanning with time to measure the angle dependence, the angle dependence will be detected nearly simultaneously using multiple detectors. A new type of compact programmable photomultiplier will be used that allows rapid sensitive measurement of the matrix elements. Examination of the data with the optimization technique showed that twelve detectors would be sufficient for the fitting.

The data from the photomultiplier tubes are multiplexed into a 16-bit A/D converter that is connected to a dedicated PC. The phase sensitive lock-in detection is implemented in the computer software instead of having twelve lock-in amplifiers. The estimated time response of the scatterometer will be in the 1 to 10 Hz range, depending on the A/D configuration. Figure 5 illustrates the final design of the two new scatterometers under construction.

FUTURE PLANS

We plan to complete the construction of two Scatterometers for particle size measurements in early FY 1999. The instruments are designed to operate at a time resolution in the 1-10 Hz rate. At faster rates the data will be taken at a rapid rate and the analysis performed later to analyze rapid transient events. One instrument will be used at ORNL and the other by LBNL with various engines. The Scatterometer results will be compared at ORNL with simultaneous measurement with a Scanning Mobility Particle Sizer and Electrical Aerosol Analyzer. In addition the Scatterometer will be used to characterize exhaust from several fuels in OAAT fuel matrix. The use of shorter wavelengths will be explored for enhanced sensitivity to the smallest particles in modem diesel exhaust. The modeling effort will continue development of the coupled-dipole calculation for sizing non-spherical diesel particles with the Scatterometer.

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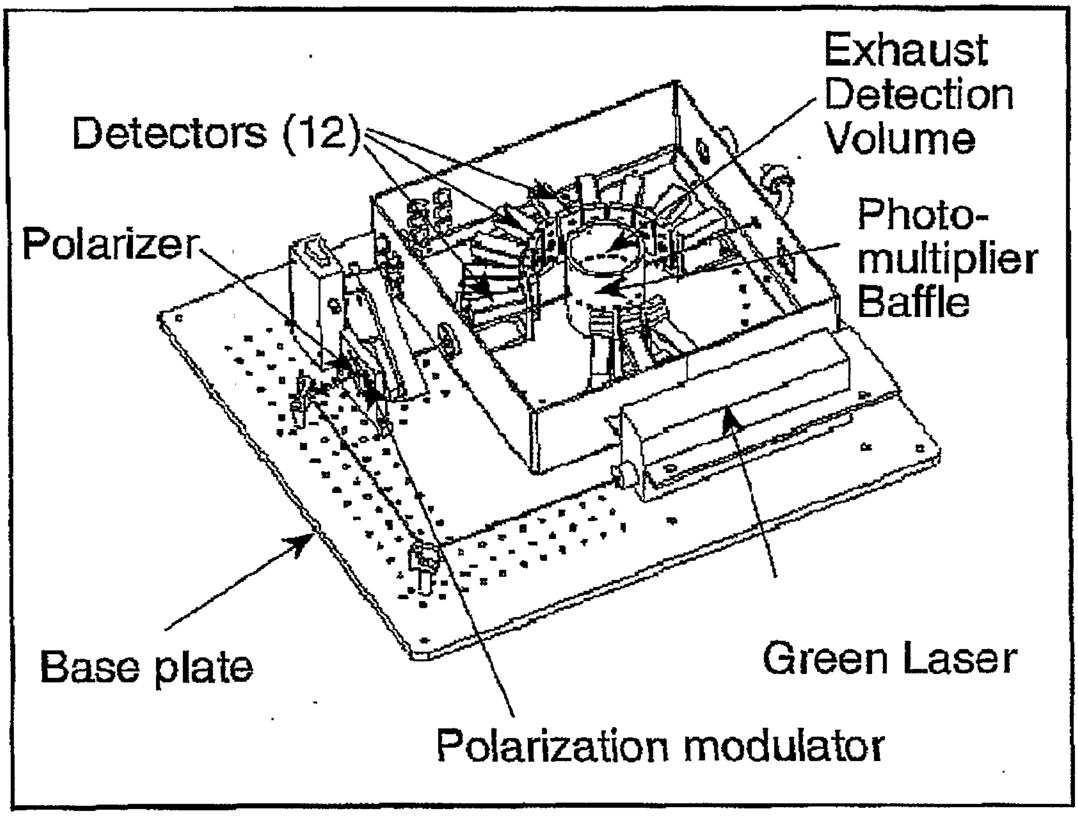

Figure 5. Design of the diesel exhaust scatteromete.

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