Emission Rates of Particulate Matter and Elemental and Organic Carbon from In-Use Diesel Engines

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Elemental carbon (EC), organic carbon (OC), and particulate matter (PM) emission rates are reported for a number of heavy heavy-duty diesel trucks (HHDDTs) and back-up generators (BUGs) operating under real-world conditions. Emission rates were determined using a unique mobile emissions laboratory (MEL) equipped with a total capture fullscale dilution tunnel connected directly to the diesel engine via a snorkel. This paper shows that PM, EC, and OC emission rates are strongly dependent on the mode of vehicle operation; highway, arterial, congested, and idling conditions were simulated by following the speed trace from the California Air Resources Board HHDDT cycle. Emission rates for BUGs are reported as a function of engine load at constant speed using the ISO 8178B Cycle D2. The EC, OC, and PM emission rates were determined to be highly variable for the HHDDTs. It was determined that the per mile emission rate of OC from a HHDDT in congested traffic is 8.1 times higher than that of an HHDDT in cruise or highway speed conditions and 1.9 times higher for EC. EC/OC ratios for BUGs (which generally operate at steady states) and HHDDTs show marked differences, indicating that the transient nature of engine operation dictates the EC/OC ratio. Overall, this research shows that the EC/OC ratio varies widely for diesel engines in trucks and BUGs and depends strongly on the operating cycle. The findings reported here have significant implications in the application of chemical mass balance modeling, diesel risk assessment, and control strategies such as the Diesel Risk Reduction Program.

Introduction

Particulate emissions from diesel engines are composed predominantly of elemental carbon (EC) and organic carbon (OC) (1). The elemental fraction stems from fuel droplet pyrolysis, while the organic fraction originates from unburned fuel, lubricating oil, and combustion byproducts (2, 3). Many carcinogenic and mutagenic compounds have been measured in the organic fraction of diesel particulate matter (PM) (4–9). Additionally, workplace exposure to diesel exhaust has resulted in elevated levels of DNA adducts in chronically exposed personnel (10).

TABLE 1. CARB HHDDT Cycle Properties

	distance (mi)	duration (s)	avg speed (mph)	max speed (mph)	max acceleration (mph s ⁻¹)
cold start/idle	0	600	0	0	0
creep	0.124	253	1.77	8.24	2.3
transient	2.85	668	15.4	47.5	3.0
cruise	23.1	2083	39.9	59.3	2.3

Diesel engines are believed to be the major source of EC in the atmosphere (11). This has led to the use of EC as a marker for assessing human exposure to diesel exhaust, for determining the contribution of diesel engines to ambient particulate concentrations, and for a surrogate for diesel PM (12-15). The use of integrated EC mass in these calculations requires that ambient EC be dominated by diesel sources and be released at fixed ratios of EC to total PM mass. However, Hildemann et al. (5) reported EC and OC emission rates for various combustion sources and determined that gasoline automobiles and oil-fired burners release significant quantities of EC. Other researchers have noted that the emission rate of EC and OC from diesel engines varies with engine load and is not constant over the entire operating range of the engine (4, 16). Thus, the use of EC as a marker for diesel exhaust or as a surrogate for total diesel PM is only valid when other sources are insignificant (17) and engine operating conditions are well-defined (18).

Most research to-date has tested diesel engines on chassis and engine dynamometer facilities with either steady-state cycles or transient cycles (4–7). These researchers report that heavy-duty diesel engines following steady-state cycles release PM with 20–39% OC and 33–51% EC (4, 5), while heavy-duty engines following transient cycles such as the Central Business District Cycle and hot-start Federal Test Procedure produce PM with 30–35% OC and 30–43% EC (6, 7).

No research has reported EC and OC emission rates of heavy-duty diesel trucks collected while the vehicle transits on the road. In this paper, a unique laboratory is used to sample diesel exhaust and collect PM samples from trucks on the road as they are driven over a prescribed four-phase driving cycle. On-road measurements were necessary as electronically controlled engines have multiple operating modes that change the emissions characteristics. PM mass, elemental carbon, and organic carbon emission rates are reported for 11 trucks over the four distinct phases of the California Air Resources Board (CARB) Heavy Heavy-Duty Diesel Truck (HHDDT, gross vehicle weight >33 000 lb) cycle (*19*). Additionally, a comparison of EC and OC emissions of these trucks and those of similarly sized back-up generators (BUGs) is presented.

Experimental Section

Driving Cycle and Test Site. The CARB HHDDT cycle (19) was selected for emission rate testing under different vehicle operating conditions. The driving cycle simulates four basic HHDDT operational modes: cold start/idle, creep, transient, and cruise. Cold start/idle consists of a cold start followed by a 10-min idle; creep phase simulates slow driving such as stop and go in heavily congested traffic; transient phase simulates light to medium traffic arterial road driving; cruise phase simulates highway driving. Table 1 provides characteristics of the HHDDT cycle with a velocity profile provided in Figure 1.

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FIGURE 1. CARB 4-phase HHDDT test cycle consists of a cold start/idle (I), creep (II), transient (III), and cruise (IV) phase.

vehicle	test date	truck model	odometer (mi)	engine year	engine model	rated power (hp)	speed (rpm)
1	07/20	Freightliner D120	545 700	1996	Detroit Diesel Series 60	360/400	1800
2	07/18	International 9800 SBA	442 674	1997	Cummins M11	330	1800
3	07/24	Freightliner D120	512 786	1997	Cummins N14	370/435	1800
4	07/29	Freightliner C-120	353 953	1997	Cummins N14	370/435	1800
5	08/14	Freightliner C-120	449 404	1997	Detroit Diesel Series 60	370/430	1800
6	08/17	Freightliner C-120	489 310	1998	Detroit Diesel Series 60	470	2100
7	09/06	Freightliner C-120	469 801	1998	Detroit Diesel Series 60	360	1800
8	12/18	Freightliner C-120	163 349	1998	Detroit Diesel Series 60	370/430	1800
9	01/24	Freightliner C-120	521 048	1998	Caterpillar C-12	355/410	2100
10	01/10	Freightliner C-120	382 246	1999	Caterpillar C-12	355/410	1800
11	09/17	Freightliner C-120	9 000	2000	Caterpillar C-15	475	2100

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The test site used for this study, located in Coachella, CA, was chosen to minimize grade effects (*20*) while providing a safe area for on-road testing. Low-speed and high-speed test cycles were performed on area local roads and Interstate 10, respectively.

CE-CERT Mobile Emissions Laboratory. All sample media were collected using the CE-CERT Mobile Emissions Laboratory (MEL); details of the MEL are provided elsewhere (21, 22). Briefly, the MEL is a 53-ft trailer with a full-scale dilution tunnel (up to 4000 cfm full flow) designed to meet Code of Federal Regulations (23) requirements. The primary dilution tunnel is a full-flow constant-volume sampling system utilizing smooth approach venturis and a dynamic flow controller to control tunnel flow. A double-wall insulated stainless steel snorkel connects the MEL directly to a diesel engine's exhaust system providing total capture of the exhaust stream. The exhaust stream is diluted with ambient air. Prior to entering the dilution tunnel, coarse and fine particles are removed from dilution air via filtering, and hydrocarbons are removed via an activated charcoal filter. Analyzers for NO_x, CO, CO₂, total hydrocarbons, and methane sampling are withdrawn at a distance 10 tunnel diameters from the initial point of mixing of dilution air and the exhaust. The weight of the MEL (20 400 kg) provides engine load during on-road testing.

PM was collected using the secondary dilution system (SDS) installed in the MEL and designed to meet 2007 CFR specifications (*24*). The SDS includes an impactor (*25*) installed at the inlet of the SDS for removal of particles with

an aerodynamic diameter greater than 2.5 μ m (24); a number of parallel sample channels for simultaneous collection of multiple filter media; and a temperature control to maintain filter face temperatures at 47 ± 5 °C (24). Diluted exhaust is isokinetically withdrawn from the primary dilution tunnel and further diluted with clean, dry dilution air. This diluted sample is then split into several channels for the collection of various filter media and gas-phase samples. Further details of the SDS are described elsewhere (22). A schematic of the SDS is given in Figure S1 of the Supporting Information.

Test Fleet and Fuel. Emissions from 11 HHDDTs are reported in this paper; the vehicles are summarized in Table 2. Each vehicle was powered by a 4-stroke, turbocharged diesel engine. Ultra-low sulfur diesel, sulfur content <15 ppm, was used in this research. Fuel properties are listed in Table 3.

Filter Media and Analysis. Pall Gelman (Ann Arbor, MI) 47 mm PTFE Teflo filters were used to collect PM mass. Filter preparation and handling met the requirements of the CFR (*23, 24*). Filter weights were measured with a Cahn (Madison, WI) C-35 microbalance.

Pall Gelman (Ann Arbor, MI) 47 mm Tissuquartz fiber filters were used to collect PM for EC and OC analysis. Each quartz filter was pretreated in a furnace at 600 °C for 5 h. Each filter was stored in a separate sealed Petri dish at 10 °C prior to and after sampling. A Sunset Labs (Forest Grove, OR) thermal/optical carbon aerosol analyzer analyzed a 1.5 cm² quartz filter punch following the temperature program outlined in the NIOSH 5040 method (*13*). A correction of

TABLE 3. Typical Properties of Ultra-Low Sulfur Diesel Fuel Used in This Work

property	test method	limit
Ash, wt %, max carbon residue, 10% btms, wt %, max cetane index, typical cetane no., typical Cu Strip Corr., 3 h at 122 F, max distillation	D-482 D-524 D-4737 D-613 D-130	0.01 0.35 55 53.5 3
<i>T</i> 90%, °F final boiling pt, °F, max flash point, °F, min gravity, API, typical lubricity, g, typical stability, mg/100 mL, max sulfur, ppm viscosity, cSt at 40 °C	D-56 D-287 D-6078 D-2274 D-5453 D-482	540-640 698 125 38 3100 1.0 15 1.9-4.1

35% for OC gas-adsorption artifact was applied to the final OC. The correction factor was based on a comparison of OC captured on parallel quartz filters with one train containing an XAD-4-coated annular denuder for gas-phase semivolatiles upstream of the filter. Correction factors were determined for all engine operating modes. These factors ranged from 23 to 38%. A weighted average, based on the total number of samples taken, of 35% was calculated as the OC gas-adsorption artifact.

Results and Discussion

PM Emission Rates. The PM mass emission rates for each vehicle (g min⁻¹ for cold start/idle, g mi⁻¹ for other phases of the HHDDT cycle) are summarized in Table 4. Generally, the grams per mile emission rates for each vehicle were highest for the creep phase and lowest for the cruise phase. On average, a vehicle would have to travel 3.6 mi in cruise mode to release the equivalent PM mass as it would traveling 1 mi in the creep phase. Similarly, the same vehicle would have to travel 2.3 mi in cruise mode to release the equivalent PM mass as traveling 1 mi in the transient phase. The decrease

in PM mass emission rates from model year 1997 to model year 1998 during the creep phase was attributed to a reduction in OC (see Elemental and Organic Carbon Emission Rates for Individual Vehicles section).

Vehicles 6 and 10 appear to be outliers in Table 4. The 1998 engine in vehicle 6 had significantly higher PM emissions than the two other engines of the same model and year (vehicles 7 and 8) during transient and cruise operation. The increase is attributed to increased elemental carbon (see Fleet Averaged Elemental and Organic Carbon Emission Rates section). The 1999 CAT C-12 engine in vehicle 10 had significantly higher PM emissions as well. This vehicle was unable to meet the required accelerations and higher speeds of the cycle and exhibited significant deviations from the speed trace. We did not further pursue the reasons for poor engine performance.

Fleet Averaged Elemental and Organic Carbon Emission Rates. A Freightliner truck equipped with a model year 2000 Caterpillar C-15 engine was tested on three nonconsecutive days to estimate variability in EC and OC mass emission rates. Table 5 summarizes the results of the tests. The cruise phase exhibited the best reproducibility of EC and OC while cold start/idle exhibited the greatest variation. Overall, the coefficient of variation for EC was lower than OC for all HHDDT cycles.

Fleet average EC and OC emission rates (g mi⁻¹, g min⁻¹, and g (kg of CO_2)⁻¹) are summarized in Table 6. Average EC and OC emission rates (g mi⁻¹) are largest for creep phase and transient phase, respectively. EC and OC emission rates per time (g min⁻¹) were largest for transient operation. The high standard deviations in measured EC and OC indicates a large variability in the EC and OC emission rates between vehicles with engine type, model year, manufacturer, and size. Averages calculated based on engine manufacturer, model year, and/or size show smaller standard deviations. The EC and OC emission rates are similar to those reported by others (4–7). Table 7 summarizes EC and OC emissions as a percent of total PM mass during this study compared with other research groups.

	TABLE	4. Average	of Two	Measurements	of PM	Mass	Emission	Rates	of 11	Vehicles	over	the	CARB	HHDDT	Cycle
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vehicle	engine	cold start/idle (mg min ⁻¹)	creep (mg mi ⁻¹)	transient (mg mi ⁻¹)	cruise (mg mi ⁻¹)
1	1996 DDC-60	19.5 ± 8.1	822 ± 12	490 ± 16	106 ± 3
2	1997 Cummins M11	15.1 ± 1.3	1110 ± 190	724 ± 67	221 ± 6
3	1997 Cummins N14	17.9 ± 1.4	1551 ± 181	864 ± 100	513 ± 39
4	1997 Cummins N14	21.6 ± 3.9	1234 ± 37	596 ± 25	206 ± 27
5	1997 DDC-60	22.4 ± 6.6	1734 ± 632	825 ± 1	69 ^b
6	1998 DDC-60	46.8 ± 3.1	612 ± 25	818 ± 31	184 ± 13
7	1998 DDC-60	10.4 ± 1.9	479 ± 75	571 ± 48	88.6 ± 4
8	1998 DDC-60	72.1 ± 0.5	625 ± 102	578 ± 10	61.3 ± 1
9	1998 CAT C-12	25.4 ± 10.1	643 ± 102	503 ± 3	102 ± 3
10	1999 CAT C-12	31.4 ± 0.9	1427 ± 461	803 ± 12	686 ± 17
11	2000 CAT C15	67.3 ± 15.0	940 ± 185	449 ± 70	128 ± 5
	fleet average	30.8 ± 21.0	1016 ± 426	656 ± 153	215 ± 201

^a Uncertainty is the range of the two measurements. ^b Only one measurement was available for this data point.

TABLE 5. Results of Repeatability Tests

	idle		creep		transient		cruise	
	(mg min ⁻¹)		(mg mi ⁻¹)		(mg mi ⁻¹)		(mg mi⁻¹)	
	EC	00	EC	00	EC	00	EC	00
no. of tests	8	8	18	18	16	16	8	8
average	1.88	40.98	520.28	419.88	295.35	174.42	80.55	41.11
SD ^a	0.40	7.78	59.64	93.52	20.70	18.44	2.67	2.21
SD ^a	0.40	7.78	59.64	93.52	20.70	18.44	2.67	

TABLE 6. Fleet Average EC and OC Emission Rates (in mg min⁻¹, mg mi⁻¹, and mg (kg of CO_2)⁻¹) Emitted over the CARB HHDDT Cycle

	idle	creep	transient	cruise
EC (mg min ⁻¹) OC (mg min ⁻¹) EC (mg mi ⁻¹) OC (mg mi ⁻¹) EC (mg (kg of $CO_2)^{-1}$)	$\begin{array}{c} 4.10 \pm 2.38 \\ 20.9 \pm 11.6 \end{array}$ $23.1 \pm 14.7 \end{array}$	$\begin{array}{c} 10.4 \pm 4.8 \\ 17.0 \pm 6.4 \\ 340. \pm 140. \\ 607. \pm 329. \\ 63.5 \pm 27.5 \end{array}$	$\begin{array}{c} 110.7\pm27.0\\ 45.5\pm13.2\\ 446.\pm115.\\ 182.9\pm51.2\\ 154.3\pm43.9\end{array}$	$\begin{array}{c} 93.0\pm 68.3\\ 42.3\pm 26.8\\ 175.\pm 172.\\ 74.7\pm 56.3\\ 87.1\pm 74.1\end{array}$
OC (mg (kg of $CO_2)^{-1}$)	113.5 ± 55.7	105.1 ± 39.3	63.2 ± 19.2	38.0 ± 25.6

TABLE 7. Comparison of EC and OC Percent of Total PM Mass with Other Studies of HDD Engines

source	cycle	fuel	no. of vehicles	EC	OC
6	hot start FTP	CA RFD ^a	2	30.8 ± 2.6	19.7 ± 1.6
7	Central Business District	no. 2 diesel	4	43.3 ± 20.1	35.4 ± 17.8^{b}
4	1600 rpm and 100% load		1	33.87 ± 3.85	24.53 ± 11.87
	1600 rpm and 50% load		1	35.88 ± 3.20	44.67 ± 8.40
	1600 rpm and 25% load		1	24.58 ± 3.47	57.66 ± 8.28
	2600 rpm and 100% load		1	51.52 ± 1.58	24.46 ± 10.16
	2600 rpm and 50% load		1	47.30 ± 2.62	28.87 ± 2.80
	2600 rpm and 25% load		1	35.16 ± 1.48	49.89 ± 8.85
5	4 mode ^c		2	41	39
this study	cold start/idle	$ULSD^d$	11	17.3 ± 12.3	72.7 ± 14.8
<u>,</u>	ARB creep	ULSD	11	36.8 ± 13.6	60.1 ± 14.1
	ARB transient	ULSD	11	67.8 ± 7.2	28.8 ± 9.1
	ARB cruise	ULSD	11	61.0 ± 7.3	33.0 ± 15.0

^a CA RFD, California reformulated diesel. ^b OC was reported as raw OC/total PM mass. No corrections were applied to account for hydrogen or oxygen associated with the organic carbon. ^c 4-mode steady-state cycle with accelerations and decelerations. ^d ULSD: ultra-low sulfur diesel.





Elemental and Organic Carbon Emission Rates for Individual Vehicles. EC and OC emission rates for each tractor are displayed in Figures 2 and 3. OC is the major species during cold start/idle and creep operation, and EC is the major species during transient and cruise operation. As previously mentioned, the decrease in PM mass emissions from engine model year 1997 to engine model year 1998 during the creep phase is attributable to a decrease in OC emissions as seen in Figure 3. This decreased OC emission rate is consistent with oil control measures enacted by engine manufacturers between 1997 and 1998 where, in general, cylinder liner surface finishes, piston ring oil control performance, and valve guide oil sealing were improved (*26*). During the transient and cruise phases, the engine emits substantially less OC so the net PM reduction is not as pronounced.

The carbonaceous fraction (CF) of total PM released was estimated by

$$CF = (EC + 1.2 \times OC)/PM$$
(1)

where the 1.2 accounts for hydrogen and oxygen present in



FIGURE 3. Elemental and organic carbon emission rates of 11 trucks during the creep (II), transient (III), and cruise (IV) phases of the CARB HHDDT cycle.



FIGURE 4. EC/OC ratio of 11 trucks over the CARB HHDDT.

the OC; EC, OC, and PM refer to the mass of EC, OC, and PM collected, respectively. CF was estimated to be approximately 0.90, 0.96, 0.96, and 0.94 for idle phase, creep phase, transient, and cruise phases, respectively. The remainder of PM mass may be attributable to trace elements and ions, although further analysis of the filters is required to confirm this hypothesis.

Comparison of EC/OC for HHDDTs and BUGs. Three BUGs were tested: a 350-kW generator equipped with a 1999 Cummins N14 engine; a 350-kW generator equipped with a 2000 Caterpillar 3406C engine; and a 300-kW generator equipped with a 1985 Detroit Diesel V92 engine. The Cummins and Caterpillar engines are both 4-stroke diesel engines with age, type, and engine block, similar to the tested vehicles. The Detroit Diesel (DDC) engine used older earlier 2-stroke technologies. The BUGs were tested following the ISO 8178B Cycle D2 with No. 2 diesel fuel (*27*). The ISO 8178B Cycle D2 is a constant-speed cycle with the engines tested at five distinct loads (10, 25, 50, 75, and 100% of full load). A load bank designed to dissipate generated electricity as heat provided the proper engine loading.

EC/OC ratios for the 11 tested tractors are given in Figure 4. EC/OC ratio is below unity for cold start/idle and creep operation; it exceeds unity for transient and cruise operation. Figure 5 displays the EC/OC ratios for three BUGs. The EC/OC ratios for the Cummins and Caterpillar BUG are above unity for all modes of operation. The difference in EC/OC ratios between vehicle engines and the Cummins and Caterpillar BUGs are due to a greater organic fraction of PM for the vehicles and a greater elemental carbon fraction for the BUGs. These differences are likely due to the transient nature of vehicle engine operation compared with the steady-



FIGURE 5. EC/OC ratio of three back-up generators.

state operation of the BUGs. Even when trucks are running at a constant vehicle speed, the operating parameters of the engine (boost pressure, fuel rate, rate of heat release, cylinder pressure, etc.) are dynamic and thus lead to higher OC production. The driver needs to occasionally throttle the engine to maintain vehicle speed; the BUG has a constant power draw resulting in steady-state engine operating parameters. Overall, the BUGs emitted 1.2–7 times more PM mass (in terms of g min⁻¹) than the vehicles, dependent on engine and load.

Figure 5 includes an older BUG with a 2-stroke engine. This engine was expected to have significantly different emission characteristics than the BUGs using 4-stroke engines. Higher OC emissions are expected based on the age of the engine, type of engine, and higher emissions standards for PM. Newer engines consume less lubricating oil based on advances in piston sealing and the use of 4-stroke technology. The low EC/OC ratios (much less than unity) reflect the high OC emissions for the older engine.

Discussion. Our data demonstrate that EC and OC emissions differ from engine to engine and are largely dependent on the mode of vehicle operation. This is the first reported study of this dependence from on-road measurements and is important to be recognized when determining source contributions of PM. Variations in emission rates within the same model and model year indicate that engine operating conditions and possibly maintenance are important factors that contribute to EC and OC emission rates. The EC/OC and EC/PM mass ratios exhibit strong dependence on the driving mode. In general, the total PM, EC, and OC emissions rates (g min⁻¹ and g mi⁻¹) are higher at low vehicle speeds compared with high vehicle speeds. The majority of PM emissions are OC at low vehicle speeds versus EC at higher sustained speeds. EC/OC ratio for BUG tests at constant speed and load vary considerably when compared with the transient vehicle engines, suggesting that EC/OC ratios from modern 4-stroke heavy-duty diesel engines are highly dependent on the transient nature of engine operation.

The equivalent miles that a vehicle must travel to emit similar masses of EC and OC provide insight into the dependence of vehicle operation on total mass emissions. Based on the results of this study, a HHDDT must travel in cruise mode for an average of 8.1 mi to match the OC output from 1 mi in creep operation. Similarly, an average HHDDT must travel 1.9 mi in cruise mode to match the EC output from 1 mi in creep operation. Put another way, the single average vehicle transiting 50 mi in heavily congested conditions from Riverside County to Los Angeles County would be expected to release 30.3 g of OC trip⁻¹ and 17.0 g of EC trip⁻¹ compared with 3.73 g of OC trip⁻¹ and 8.73 g of EC trip⁻¹ for transit under open highway conditions. The implications for commuters and personal exposure are significant. This finding also suggests that air quality, especially near freeways, can be vastly improved by reducing truck travel during heavily congested conditions.

Fluctuations in EC and OC emission rates and EC/PM fractions have direct implications on chemical mass balance (CMB) models using EC and OC in their chemical profiles. CMB models operate on the assumption that the fractional contribution of each species to total PM can be assumed fixed; given the variability in EC/PM and EC/OC ratios for this study, the efficacy of the model to distinguish between types of combustion sources (e.g., gasoline, diesel, wood combustion) while using EC and OC in the source profiles may lead to significant error. Other researchers have observed this variation in EC/PM and EC/OC and its potential to lead to errors in modeling (*4, 16, 28*).

Additionally, the ARB has adopted a unit cancer risk for diesel PM. However, this risk factor does not account for variations in PM composition due to the type of engine and mode of operation. Therefore, it might be expected that the toxicity and carcinogenicity of the diesel PM would be highly dependent on engine application and vehicle operational mode. If one assumes that the organic fraction is predominately responsible for the carcinogenic and toxic effects of the diesel PM, the human health risk increases almost an order of magnitude in highly congested traffic compared with free-flowing traffic.

In 2000, the California ARB approved the Diesel Risk Reduction Program (29), in which various regulatory and nonregulatory strategies were recommended to reduce the contribution of both mobile and stationary diesel engines to the state's PM inventory. These measures included lower emission standards for new engines, retrofitting existing engines with diesel particulate filters, and new fuel standards. Nonregulatory strategies included the voluntary use of diesel particulate filters for certain industry-related vehicles and idling restrictions. Full implementation of the plan is expected to reduce the diesel PM inventory by 75% by 2010. This reduction is gradual and dependent on the full implementation of the plan. Additional and immediately tangible reductions can be achieved through the use of vehicle flow controls or nonregulatory incentive programs that encourage owners/operators to avoid travel during congested conditions.

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Supporting Information Available

Schematic of the secondary dilution system. This material is available free of charge via the Internet at http://pubs.acs.org.

Literature Cited

- Kleeman, M. J.; Schauer, J. J.; Cass, G. R. Environ. Sci. Technol. 2000, 34, 1132–1142.
- (2) Abdul-Khalek, I. S.; Kittelson, D. B.; Graskow, B. R.; Wei, Q.; Brear, F. SAE Tech. Pap. Ser. 1998, No. 980525.
- (3) Abdul-Khalek, I. S.; Kittelson, D. B.; Brear, F. SAE Tech. Pap. Ser. 1998, No. 1999-01-1142.
- (4) Shi, J. P.; Mark, D.; Harrison, R. M. Environ. Sci. Technol. 2000, 34, 748–755.
- (5) Hildemann, L. M.; Markowski, G. R.; Cass, G. R. Environ. Sci. Technol. 1991, 25, 744–759.
- (6) Schauer, J. J.; Kleeman, M. J.; Cass, G. R.; Simoneit, B. R. T. Environ. Sci. Technol. 1999, 33, 1578–1587.
- (7) Lowenthal, D. H.; Zielinska, B.; Chow, J. C.; Watson, J. G.; Gautam, M.; Ferguson, D. H.; Neuroth, G. R.; Stevens, K. D. Atmos. Environ. 1994, 28, 731–743.
- (8) Rogge, W. F.; Hildemann, L. M.; Mazurek, M. A.; Cass, G. R.; Simoneit, B. R. T. *Environ. Sci. Technol.* **1993**, *27*, 636–651.
- (9) Health Effects Institute. Diesel Exhaust: A Critical Analysis of Emissions, Exposure, and Health Effects, 1995.
- (10) Lloyd, A. C.; Cackette, T. A. J. Air Waste Manage. 2001, 51, 809– 847.

- (11) U.S. Environmental Protection Agency. *Health Assessment Document for Diesel Exhaust*. Prepared by the National Center for Environmental Assessment, Washington, DC, for the Office of Transportation and Air Quality; 2002; EPA/600/8-90/057F.
- (12) Birch, M. E.; Cary, R. A. Aerosol Sci. Technol. 1996, 25, 221-241.
- (13) NIOSH. *NIOSH Manual of Analytical Methods*; National Institute of Ocupational Safety and Health: Cincinnati, OH, 1996.
- (14) Groves, J.; Cain, J. R. Ann. Occup. Hyg. 2000, 44 (6), 435-447.
- (15) McDonald, J. D.; Zielinska, B.; Sagebiel, J. C.; McDaniel, M. R. J. Air Waste Manage. 2003, 53, 386-395.
- (16) Kweon, C. B.; Foster, D. E.; Schauer, J. J.; Pkada, S. SAE Tech. Pap. Ser. 2002, No. 2002-01-2670.
- (17) Gillies, J. A.; Gertler, A. W. *J. Air Waste Manage.* **2000**, *50*, 1459–1480.
- (18) Fraser, M. P.; Lakshmanan, K.; Fritz, S. G.; Ubanwa, B. J. Geophys. Res. 2002, 107 (D21), 8346.
- (19) Maldonado, H. Development of Heavy-Duty Truck Chassis Dynamometer Driving Cycles for Source Testing for Emissions Modeling. Presented at the 11th CRC On-Road Vehicle Emissions Workshop, San Diego, 2001.
- (20) Bishop, G. A.; Morris. J. A.; Stedman, D. H.; Cohen, L. H.; Countess, R. J.; Countess, S. J.; Maly, P.; Scherer, S. *Environ. Sci. Technol.* 2001, *35*, 1574–1578.
- (21) Cocker, D. R.; Johnson, K. J.; Shah, S. D.; Miller, J. W.; Norbeck, J. M. Environ. Sci. Technol. 2004, 38, 2182–2189.
- (22) Cocker, D. R.; Shah, S. D.; Johnson, K. J.; Zhu, X.; Miller, J. W.; Norbeck, J. M. *Environ. Sci. Technol.* Submitted for publication.
- (23) Protection of the Environment. *Code of Federal Regulations*, Part 86.1310, Title 40.
- (24) Protection of the Environment. *Code of Federal Regulations*, Part 86.1312, Title 40.
- (25) Biswas, P.; Flagan, R. C. J. Aerosol Sci. 1988, 19, 113-131.
- (26) Keski-Hynnila, D. E. Detroit Diesel, personal correspondence, 2003.
- (27) International Organization for Standardization. ISO 8178-4, 1st ed.; 1996.
- (28) McDonald, J. D.; Zielinska, B.; Sagebiel, J. C.; McDaniel, M. R. Aerosol Sci. Technol. 2002, 36, 1033–1044.
- (29) Stationary Source Division, Mobile Source Control Division. Risk Reduction Plan to Reduce Particulate Matter Emissions from Diesel-Fueled Engines and Vehicles, California Environmental Protection Agency: Sacramento, CA, October 2000.

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