



## APPENDIX AVAILABLE ON THE HEI WEB SITE

### Research Report 165

#### Allergic Inflammation in the Human Lower Respiratory Tract Affected by Exposure to Diesel Exhaust

Marc A. Riedl et al.

#### Appendix E

#### Generation and Characterization of Diesel Exhaust in a Facility for Controlled Human Exposures

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**Appendix E was originally Appendix D**

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# Generation and Characterization of Diesel Exhaust in a Facility for Controlled Human Exposures

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## ABSTRACT

An idling medium-duty diesel truck operated on ultralow sulfur diesel fuel was used as an emission source to generate diesel exhaust for controlled human exposure. Repeat tests were conducted on the Federal Test Procedure using a chassis dynamometer to demonstrate the reproducibility of this vehicle as a source of diesel emissions. Exhaust was supplied to a specially constructed exposure chamber at a target concentration of  $100 \mu\text{g} \cdot \text{m}^{-3}$  diesel particulate matter (DPM). Spatial variability within the chamber was negligible, whereas emission concentrations were stable, reproducible, and similar to concentrations observed on the dynamometer. Measurements of nitric oxide, nitrogen dioxide, carbon monoxide, particulate matter (PM), elemental and organic carbon, carbonyls, trace elements, and polycyclic aromatic hydrocarbons were made during exposures of both healthy and asthmatic volunteers to DPM and control conditions. The effect of the so-called "personal cloud" on total PM mass concentrations was also observed and accounted for. Conventional lung function tests in 11 volunteer subjects (7 stable asthmatic) did not demonstrate a significant

change after 2-hr exposures to diesel exhaust. In summary, we demonstrated that this facility can be effectively and safely used to evaluate acute responses to diesel exhaust exposure in human volunteers.

## INTRODUCTION

Advances in diesel engine design and related fields such as fuels and emission control technologies have led to substantial decreases in emissions from newer engine systems.<sup>1–3</sup> Several papers on diesel exhaust (DE) and its effects on human health have been published in recent years.<sup>4,5</sup> Studies on human bronchial cells show the impact of exposure to ambient particulate matter (PM)<sup>6,7</sup> as well as diesel particulate matter (DPM),<sup>8</sup> with at least one study suggesting that ambient PM and DPM differ only in magnitude of response.<sup>9</sup> Furthermore, evidence exists for the carcinogenicity of DE in rats, although the use of rat studies to estimate human cancer risk is debatable.<sup>10</sup> Therefore there is a strong need to investigate both acute and chronic effects of DE in humans.

Several studies exist on the exposure of both humans and animals to DE under various conditions of exposure source and subject health. Kittelson et al.<sup>11</sup> conducted on-road measurements of exposure of rats to DE, characterizing size distribution, elemental carbon (EC), and the gas-phase species carbon monoxide (CO), carbon dioxide (CO<sub>2</sub>), and nitric oxide (NO). The use of a thermal denuder suggested that most of the particles consisted largely of volatile material. In the field of controlled exposure studies, McDonald et al.<sup>12</sup> reported on the characterization of a DE exposure facility using a single-cylinder diesel engine for whole-body exposure of animals to DE.

## IMPLICATIONS

A facility has been developed for exposure of human subjects to accurate, representative, low-level concentrations of diesel exhaust. DPM within the facility is found to be representative of the chemical and physical characteristics of diesel exhaust from current on-road diesel fleets with stable concentrations achieved with respect to time and concentration during exposure of 11 human subjects.

Rudell et al.<sup>13</sup> were among the first to use an idling truck as an emission source for exposure of healthy volunteers to DE within an enclosed chamber. They found that the symptoms reported by these volunteers were in agreement with previous exposure studies to DE, and that lung function did not seem to be affected as a result of the exposure. In a later study using the same facility, Nordenhäll et al.<sup>14</sup> exposed healthy subjects to DE at concentrations of 10 and 300  $\mu\text{g} \cdot \text{m}^{-3}$  from an idling four-cylinder diesel engine. Subjects were asked to alternate between periods of rest and moderate exercise on a stationary bicycle. Exposure to DE induced an inflammatory response in their airways.

This work modified an existing exposure facility<sup>15</sup> for further investigation of the acute effects of human exposure to DE. The modified facility needed to fulfill the following objectives: (1) the exposure must be stable with respect to time for a desired concentration for accurate dose–response characterization, and (2) the exposure concentrations must be reproducible for purposes of statistical accuracy. This paper describes the facility's design, construction, and evaluation and presents the characterization of DE during exposures of 11 humans–4 healthy subjects and 7 asthmatic volunteers.

## EXPERIMENTAL METHODS

The initial objective was to deliver well-characterized DE from an in-use nonmodified diesel engine to the test facility for human exposure. We initially procured a medium-duty truck and characterized its emissions using a federally certified operating cycle (the Federal Test Procedure [FTP]<sup>16</sup>). DE was characterized at the facility at a DPM mass concentration of 100  $\mu\text{g} \cdot \text{m}^{-3}$ .

### Vehicle Selection

DE was generated using a 1999 medium-duty Ford pickup truck with a 250-hp International T 444E 7.27L turbocharged V-8 engine. The vehicle mileage was approximately 60,000 miles, which was deemed sufficient to generate stable emissions.<sup>17</sup> The truck was equipped with a factory-installed oxidation catalyst installed when it was purchased for the study. After characterizing the truck's emissions in the presence of the catalyst, the catalyst was removed. The rationale was that the study should simulate emissions from the large majority of diesel vehicles in use (such as heavy-duty trucks and buses) rather than the small number or light-duty trucks equipped with the catalyst. Although the majority of light-duty trucks sold after 2000 have an oxidation catalyst, heavy-duty vehicles use primarily exhaust gas recirculation.

### Fuel Selection

California Air Resources Board ultra low sulfur diesel (ULSD) was selected as the fuel to minimize interference from formation of oxides of sulfur ( $\text{SO}_x$ ) during the exposure testing, because  $\text{SO}_2$  is a known trigger of asthma symptoms and airway constriction.<sup>18</sup> Table 1 presents the properties of the ULSD fuel, which is representative of future diesel fuels as dictated by the state of California

**Table 1.** Properties of ULSD fuel.<sup>29</sup>

Property	Test Method	Limit
Ash, wt %, max	D-482	0.01
Carbon residual, 10% Btms, wt %, max	D-524	0.35
Cetane index, typical	D-4737	55
Cetane number, typical	D-613	53.5
Cu strip corr, 3 hr at 122 °F, max	D-130	3
Distillation	D-86	
Temperature 90%, °F		540–640
Final boiling point, °F, max		698
Flash point, F, min	D-56	125
Gravity, API <sup>a</sup> , typical	D-287	38
Lubricity, g, typical	D-6078	3100
Stability, mg/100 mL, max	D-2274	1.0
Sulfur, ppm	D-5453	15
Viscosity at 40 °C, centiStokes	D-482	1.9–4.1

Notes: API = American Petroleum Institute; corr = corrosion; Btms = bottoms.

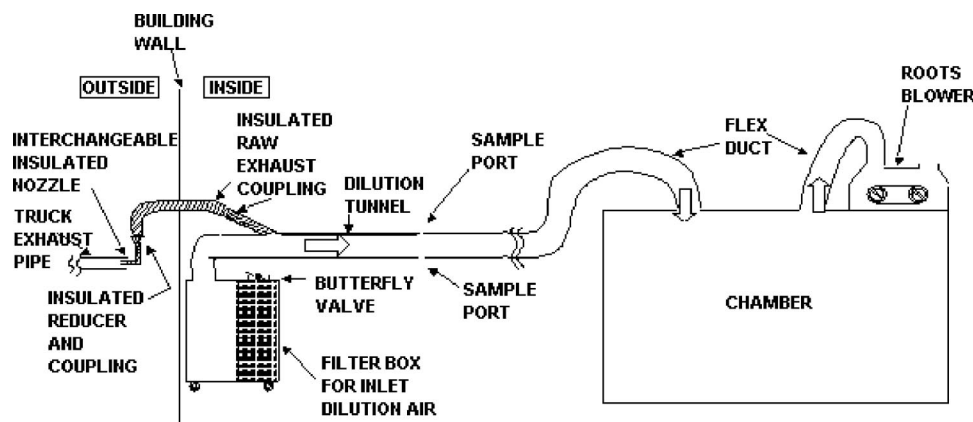
and federal government regulations effective 2006 and beyond.<sup>19</sup>

### Exhaust Characterization: FTP-75 Test Cycle

Characterization of the test vehicle was carried out in the Vehicle Emissions Research Laboratory (VERL) at the Bourns College of Engineering-Center for Environmental Research and Technology (CE-CERT) in Riverside, CA, as described in detail by Durbin et al.<sup>2</sup> Briefly, VERL uses a 48-in Burke E. Porter 3900-3595 two-wheel drive single-roll electric chassis dynamometer (Burke E. Porter Machinery); a dedicated 12-in. inner diameter, 132-in. long dilution tunnel for DE; and a Pierburg positive displacement pump-constant volume sampler (PDP-CVS) to sample exhaust for gaseous and particle emissions (Kolbenschmidt Pierburg). A Pierburg AMA-2000 exhaust emissions monitoring system was used for measurement of regulated gas-phase emissions, i.e., total hydrocarbons (THCs), non-methane hydrocarbons (NMHCs), methane ( $\text{CH}_4$ ), CO,  $\text{CO}_2$ , and oxides of nitrogen ( $\text{NO}_x$ ). PM mass was measured using polytetrafluoroethylene (PTFE) filters weighed on a Cahn C-35 (Thermo) microbalance. The VERL facility is termed “certification-quality” on the basis of available facilities and comparison tests with other certified laboratories. The vehicle was tested on the FTP (FTP-75) driving cycle. This is the cycle used for emission certification of light-duty vehicles in the United States and consists of three phases: cold start, transient, and hot start.<sup>16</sup>

### Exposure Chamber and Air Monitoring System

Figure 1 shows a schematic of the exposure chamber facility at the Los Amigos Research and Education Institute (LAREI) in Downey, CA. The following procedures were followed for exposure testing. Before the start of each test day, the truck was warmed up to stabilize DE emissions by driving a prescribed 10-mi loop in the area surrounding the laboratory. The truck was then driven



**Figure 1.** Schematic of exposure chamber at LAREI.

back to the laboratory and parked with the engine shut off in a location physically just on the outside of the room containing the exposure facility. A nozzle inserted into the tailpipe collected undiluted exhaust, which was transferred to a dilution tunnel connected to the exposure chamber. The building heating/ventilation/air conditioning (HVAC) system was used as the source of dilution air to better ensure minimal fluctuation in dilution air properties over time. The exposure facility was equipped with a variable-speed Roots blower downstream, with a Mitsubishi FR-S520-1.5K-NA three-phase 240 V 50/60 Hz frequency controller (flow rate range: 0–600 ft<sup>3</sup>/min). The Roots blower served to draw out the ambient air from the facility into which the diluted DE from the dilution tunnel was drawn.

A scanning mobility particle sizer (SMPS)<sup>20</sup> comprising a TSI 3081L cylindrical differential mobility analyzer (DMA; TSI), a TSI 3760A condensation particle counter (CPC), and a TSI 3077 <sup>85</sup>Kr neutralizer was used to measure the desired mass concentration in this case. This instrument is similar to one described in Cocker et al.<sup>21</sup> For the test, the truck engine was turned on and allowed to generate raw exhaust by regular idling (900 rpm [revolutions per minute]). The nominal dilution ratio used is 1500:1 to achieve the DPM test mass concentration of 100 μg · m<sup>-3</sup>. Coupled with high dilution ratios, such a setup would be expected to provide a steady level of diluted DE to the exposure chamber. A flow of 2900 L/min through the chamber—a Plexiglas room<sup>15</sup> with volume near 9.7 m<sup>3</sup>—gave an average DE residence time of approximately 200 sec.

### Chemical Analysis

NO and NO<sub>2</sub> were measured with an API model 200A chemiluminescent detector (Teledyne API). CO was measured with an API model 300 gas filter correlation CO analyzer. Gas-phase carbonyl compounds were collected on 2,4-dinitrophenylhydrazine (DNPH) cartridges (Waters). These cartridges were eluted with 5 mL of acetonitrile and analyzed using a Shimadzu high-performance liquid chromatograph (HPLC) with ultraviolet-visible detection following the Society of Automotive Engineers (SAE) 930142 method.<sup>22</sup>

Total particulate mass was measured using 47-mm diameter, 2-μm pore size Teflo PTFE filters (Pall). All filters

were conditioned in a constant temperature/constant relative humidity (RH) chamber maintained at 25 °C and 40% RH for a period of 24 hr, both before and after sampling. Each conditioned filter was weighed three times on a Cahn C-35 (Cahn) microbalance, and the average weight was noted. For the test run dated April 13, 2005 (to investigate the impact of human presence inside the chamber), samples for PM mass were also taken on 47-mm aluminum substrates placed in an MSP model 110 10-stage Micro-Orifice Uniform Deposit Impactor (MOUDI) and weighed in the same manner as the PTFE substrates. An SMPS was also used in parallel with the filter to obtain particle size distributions over the 33–730 nm size range. Particulate organic carbon (OC) and EC was collected on 47-mm diameter Tissuquartz quartz-fiber filters (Pall) and analyzed using a thermal/optical carbon aerosol analyzer (Sunset Laboratory) following the National Institute for Occupational Safety and Health 5040 method.<sup>23</sup> Polycyclic aromatic hydrocarbons (PAHs) were analyzed on a combination of quartz-fiber filters and sampling tubes filled with Amberlite XAD-4 (Supelco) resin bounded by polyurethane foam (PUF; University Research Glassware) plugs, and analyzed using a modified Environmental Protection Agency (EPA) TO-13A method.<sup>24</sup> Briefly, PAH samples were extracted using a Dionex (Sunnyvale) ASE-200 accelerated solvent extractor, rotary evaporated using a Buchi (Flawil) rotavapor, and injected on to an Agilent 6890/5973N gas chromatograph-mass spectrometry (GC-MS) system with a programmable temperature vaporization (PTV) inlet. Additional information on the method may be found elsewhere.<sup>25</sup>

### Chamber Characterization

Multiple tests were performed at a DPM mass exposure level of 100 μg · m<sup>-3</sup> for 2 hr each. Before the start of each testing session, the truck was warmed up to stabilize DE emissions as described previously. Air was monitored first with the chamber empty, next with investigators as subjects, and finally with volunteers for the health-effects studies. The following sections discuss the detailed results for these experiments.

### Human Exposure Testing

Clinically healthy volunteers and volunteers with a history of asthma were recruited as subjects for the exposure



**Table 2.** FTP-75 emissions results for test vehicle ( $\text{g} \cdot \text{mi}^{-1}$ ).

	FTP Run 1	FTP Run 2	FTP Run 3 <sup>a</sup>	FTP Run 4 <sup>a</sup>	Average
<b>THC</b>					
Phase 1	0.33	0.27	0.29	0.25	
Phase 2	0.33	0.35	0.38	0.38	
Phase 3	0.29	0.29	0.29	0.27	
Weighted	0.32	0.31	0.34	0.32	0.32
<b>NMHC</b>					
Phase 1	0.32	0.26	0.29	0.25	
Phase 2	0.33	0.34	0.37	0.37	
Phase 3	0.28	0.29	0.29	0.27	
Weighted	0.31	0.31	0.33	0.32	0.32
<b>CO</b>					
Phase 1	1.41	1.37	1.47	1.44	
Phase 2	1.65	1.67	1.68	1.73	
Phase 3	1.07	1.05	1.09	1.10	
Weighted	1.44	1.44	1.48	1.50	1.46
<b>NO<sub>x</sub></b>					
Phase 1	5.74	5.41	5.43	5.63	
Phase 2	3.90	3.82	3.78	3.80	
Phase 3	3.73	3.71	3.66	3.69	
Weighted	4.23	4.12	4.09	4.15	4.15
<b>CO<sub>2</sub></b>					
Phase 1	721	700	675	681	
Phase 2	681	677	653	650	
Phase 3	594	592	572	571	
Weighted	665	658	635	634	648
<b>PM mass</b>					
Phase 1	0.19	0.26	0.33	0.20	
Phase 2	0.20	0.14	0.31	0.31	
Phase 3	0.27	0.21	0.25	0.24	
Weighted	0.21	0.19	0.30	0.27	0.24

Notes: <sup>a</sup>Test vehicle with DOC removed.

testing via word-of-mouth and advertisements. Each subject gave signed informed consent to participate. Each subject was exposed on separate days to DE, HEPA-filtered air, and filtered air (FA) with NO<sub>2</sub> added at a concentration of 0.35 parts per million (ppm; similar to that in DE), for periods of 2 hr, including four 15-min bouts of moderate exercise on a stationary bicycle. Exposures of a given subject occurred in randomized order, at intervals of 4 weeks or longer. Subjects were not informed about the specific exposure sequence to minimize any placebo effects. For safety, a continuous readout of electrocardiogram and heart rate was monitored during exposure, and blood pressure was recorded periodically. A conventional measurement of lung function (forced expired volume in 1 sec [FEV<sub>1</sub>]) and a brief clinical examination were performed just before and after exposure.

## RESULTS AND DISCUSSION

### FTP-75 Tests: Gas-Phase Emissions

Table 2 lists gas-phase and PM emissions measured in VERL for the test vehicle running on the FTP-75 driving cycle. The original equipment diesel oxidation catalyst (DOC) on the test vehicle tended to preferentially reduce the tailpipe emissions for PM mass and lower the PM/NO<sub>x</sub> ratio. To have the exhaust closely resemble typical on-road diesel emissions, it was decided to remove the DOC

**Table 3.** Comparison of average weighted FTP-75 results for test vehicle with other vehicles of similar makes and models ( $\text{g} \cdot \text{mi}^{-1}$ ; weighted FTP emissions).

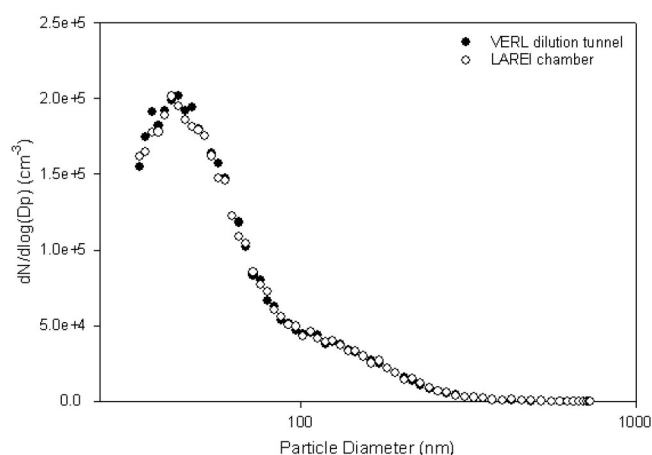
	This Study	Vehicle No. 1		Vehicle No. 2
		ULSD	CARB	ECD-1
THC	0.32	0.52	0.47	0.14
NMHC	0.32	0.55	0.50	0.19
CO	1.46	1.59	1.43	0.28
NO <sub>x</sub>	4.15	4.59	4.64	4.75
PM	0.24	0.34	0.30	0.03

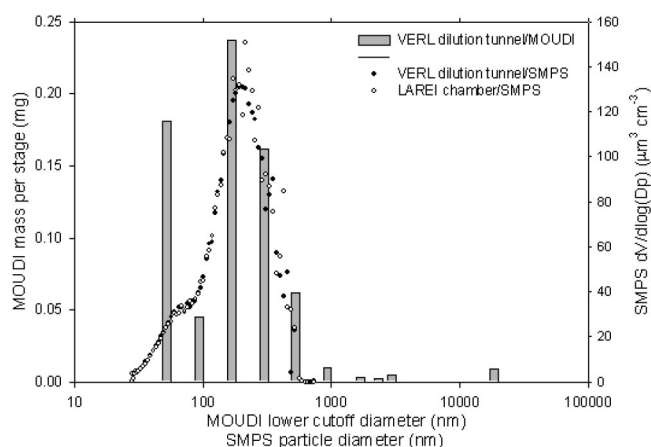
for the chamber characterization tests and exposure studies. Average weighted emission factors for THC, NMHC, CO, NO<sub>x</sub>, and CO<sub>2</sub> were found to be 0.32, 0.32, 1.46, 4.15, and 648  $\text{g} \cdot \text{mi}^{-1}$ , respectively. These compare favorably with other vehicles of similar make and model equipped with the same engine (Table 3; Durbin et al.<sup>2</sup>) and are typical of modern diesel vehicles.

### FTP-75 Tests: PM Emissions

Particulate mass concentration was measured simultaneously with an SMPS and with Teflo filters. However, only Teflo filter weights are presented in Table 2 under the heading "PM Mass." This is because the FTP-75 cycle is a transient cycle, making it difficult to compare SMPS data with cumulative filter data. As an illustration, PM fluctuations in raw exhaust are on the order of once per second, whereas the instrument acquires a scan every 60 sec. The values obtained are typical of those obtained for other diesel vehicles of similar size and age tested at VERL.

Particle number distributions and volume distributions measured in the VERL dilution tunnel for the study vehicle are represented by the black diamonds in Figures 2 and 3, respectively. Note that the volume, and therefore mass, is dominated by particles with aerodynamic diameters of approximately 200 nm whereas the number mode is dominated by particles with diameters of 80 nm; these distributions are typical for DPM. Also noted is the strong correlation in particle size/mass

**Figure 2.** Particle size distribution of DE from test vehicle measured at VERL and inside LAREI chamber.



**Figure 3.** Particle volume distribution of DE from test vehicle measured at VERL (black diamonds and gray columns) and inside LAREI chamber (white diamonds). Stages are those of MOUDI (in  $\mu\text{m}$ ): stage 1 ( $>10$ ), stage 2 ( $10\text{--}5.6$ ); stage 3 ( $5.6\text{--}3.2$ ), stage 4 ( $3.2\text{--}2.5$ ), stage 5 ( $2.5\text{--}1.8$ ), stage 6 ( $1.8\text{--}1$ ), stage 7 ( $1\text{--}0.56$ ), stage 8 ( $0.56\text{--}0.32$ ), stage 9 ( $0.32\text{--}0.18$ ), stage 10 ( $0.18\text{--}0.10$ ), stage 11 ( $0.10\text{--}0.056$ ), and after filter ( $<0.056$ ).

distributions measured in both systems. The aerosol size distribution obtained from the SMPS was verified against a MOUDI. Data on particle size distribution from the MOUDI are presented as columns in Figure 3. The significant mass observed on the after-filter is attributed to gas-phase adsorption artifact.

### Spatial Distribution within Chamber

During the initial characterization phase, an SMPS was used to ensure that there were no significant spatial nonuniformities within the chamber. The SMPS was set to a fixed voltage of 1000 V, which allowed for a detection speed of 1 Hz and corresponded to a particle diameter ( $d_p$ ) of approximately 180 nm. A probe was used to detect particle counts at nine locations throughout the chamber (near the eight corners and the centroid of the cube), over a period of approximately 40 min. A total of 100 readings were recorded at each position each time. It was determined that nonuniformity in spatial distribution in the chamber was minimal when an oscillating fan was operated within (coefficient of variance 3.7%). The concentration gradients left-to-right, top-to-bottom, and front-to-rear were all found to be nonsignificant ( $P > 0.1$ ) by regression analysis. Variation over time was also nonsignificant, indicating a high degree of stability in the exhaust output and Roots blower operation. Thus, with the internal fan operational, any nonuniformity in spatial distribution within the chamber is trivial from the standpoint of its effect on subjects' exposure levels.

### Gas-Phase Measurements: NO, NO<sub>2</sub>, CO, and Carbonyls

Average NO and NO<sub>2</sub> of  $1.06 \pm 0.08$  ppm and  $0.335 \pm 0.018$  ppm, respectively, were obtained. The NO levels were considered sufficiently low to avoid significant physiological

**Table 4.** CO, NO, NO<sub>2</sub>, PM, EC, and OC concentrations inside the chamber for healthy and asthmatic subjects.

Run Date	Subject	CO (ppm)	NO (ppb)	NO <sub>2</sub> (ppb)	SMPS Set Point ( $\mu\text{g} \cdot \text{m}^{-3}$ )	PM Mass ( $\mu\text{g} \cdot \text{m}^{-3}$ )	OC ( $\mu\text{g} \cdot \text{m}^{-3}$ )	EC ( $\mu\text{g} \cdot \text{m}^{-3}$ )	(OC + EC)/Average PM
3/16/2004	H	3.28	1103	324	96.2	140	77.4	32.9	0.81
3/30/2004	H	2.36	1036	344	ND	132	66.4	30.6	0.71
						92.9	49.4	28.8	0.75
4/6/2004	H	1.49	1123	348	107.6	134	69.4	30.2	0.74
5/5/2004	H	1.64	1046	340	112.1	137	67.2	34.0	0.75
						136	61.4	36.2	0.76
9/2/2004	A	1.7	1048	338.7	110.2	122	68.1	31.1	0.77
						ND	65.6	32.5	0.80
9/20/2004	A	1.4	918.3	300.2	104.9	122	53.6	33.6	0.72
						119	56.3	35.6	0.76
1/25/2005	A	0.50	1034	319.9	ND	142	57.9	31.3	0.63
						ND	59.1	30.8	0.63 <sup>a</sup>
2/28/2005	A	2.3	1041	344.7	104.2	173	65.1	29.9	0.55
						171	62.6	30.2	0.54
4/13/2005	None	ND	ND	ND	102.3	102	66.4	38.4	0.97
4/13/2005	A	ND	ND	ND	90.3	113	73.5	33.9	1.00
						148	74.3	27.5	0.70
4/18/2005	A	1.6	1107	327.0	103.8	144	79.1	26.8	0.73
						165	ND	ND	ND
4/20/2005	A	1.7	1240	365.5	96.8	161	ND	ND	ND
						119	64.4	24.0	0.77
5/18/2005	A	0.82	1013	ND	101.5	112	62.0	27.6	0.78
						137	61.9	31.5	0.70
						129	61.4	32.1	0.70

Notes: H = normal healthy subject; A = asthmatic subject; ND = no data available. <sup>a</sup>Single available PM mass used in lieu of average PM mass.

**Table 5a.** Concentrations of 11 aldehydes and 2 ketones inside the chamber, corresponding to 100  $\mu\text{g} \cdot \text{m}^{-3}$  DEP concentration ( $\mu\text{g} \cdot \text{m}^{-3}$ ).

Run Date	Human Exposure	Formaldehyde	Acetaldehyde	Acetone	Acrolein	Propionaldehyde	Crotonaldehyde	Methyl Ethyl Ketone
3/16/2004	H	64	25	55	2.8	5.2	0.8	8.2
3/30/2004	H	73	25	44	4.3	5.2	1.6	6.7
4/6/2004	H	59	28	23	4.3	4.2	0.93	4.2
5/5/2004	H	67	24	31	1.9	4.2	BDL	3.6
9/2/2004	A	58	20	34	3.8	3.4	2	BDL
9/20/2004	A	55	20	26	3	3.5	1.5	3
1/25/2005	A	53	20	29	BDL	3.2	0.64	4.8
2/28/2005	A	62	24	26	0.9	4.8	1	4.9
4/18/2005	A	59	22	20	2.4	4.9	2.3	3.8
4/20/2005	A	63	24	33	3.4	5	2.6	4.1
5/18/2005	A	74	24	26	4.3	5.8	2.7	4.8

Notes: H = Normal, healthy subject; A = Asthmatic subject; BDL = below detection limit.

**Table 5b.** Concentrations of 11 aldehydes and 2 ketones inside the chamber, corresponding to 100  $\mu\text{g} \cdot \text{m}^{-3}$  DEP concentration ( $\mu\text{g} \cdot \text{m}^{-3}$ ).

Run Date	Human Exposure	Methacrolein	Butyraldehyde	Benzaldehyde	Valeraldehyde	Tolualdehyde	Hexanaldehyde
3/16/2004	H	0.58	3.2	7.4	2.2	1.5	7
3/30/2004	H	0.87	3.5	7.8	1.8	1.1	5.8
4/6/2004	H	0.77	2.6	5.6	1.5	0.25	5.3
5/5/2004	H	BDL	4.3	6	BDL	BDL	6.3
9/2/2004	A	1.9	1.5	8.1	5.2	BDL	BDL
9/20/2004	A	BDL	3.2	3.4	BDL	BDL	5.7
1/25/2005	A	0.37	3.1	0.65	1.5	0.57	2.2
2/28/2005	A	0.49	3.2	2.5	1.7	0.91	2.9
4/18/2005	A	0.9	2.3	6.4	1.8	0.64	2.6
4/20/2005	A	0.92	2.2	7.8	1.6	0.72	1.5
5/18/2005	A	2.5	11	7.5	1.8	0.85	3

Notes: H = Normal, healthy subject; A = Asthmatic subject; BDL = below detection limit.

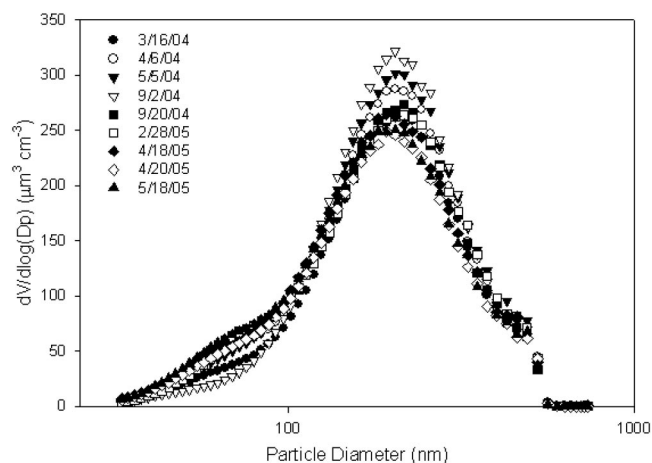
responses. However, the  $\text{NO}_2$  levels (0.335 ppm) were within the range in which health effects have been reported in some studies,<sup>18</sup> making it necessary to include exposures to  $\text{NO}_2$  alone, as well as clean-air control exposures, in the experimental design. Average CO levels were  $1.7 \pm 0.8$  ppm, considered sufficiently low to avoid significant exposure effects. Table 4 summarizes the gaseous measurements for each repeat experiment. Concentrations of carbonyls obtained are presented in Table 5, a and b; repeatability of carbonyl species is generally better than 20%.

### PM Measurements

Mass concentrations and set points for the 11 exposures are reported in Table 4, whereas particle volume distributions for 9 tests (for which data was available) are shown in Figure 4. To obtain mass concentration values, a correction factor of 1.37 is applied to the volume concentrations obtained from electrical mobility measurements to account for DPM density and nonsphericity. Note that the SMPS is used to set the appropriate dilution flow in the chamber by varying the Roots blower speed. The filter-based mass concentrations are used to verify mass loadings in the chamber. The background PM for an unoccupied chamber is less

than  $1 \mu\text{g} \cdot \text{m}^{-3}$  measured when the vehicle engine is not running.

The SMPS also reported number concentrations and size distributions for the experiments performed. The



**Figure 4.** SMPS particle volume distribution of DE from test vehicle measured inside LAREI chamber during human exposure runs for healthy subjects and asthmatics.



white diamonds in Figures 2 and 3 display the particle number and PM mass concentrations in the chamber, respectively. It is seen that these distributions are similar to those obtained on the FTP-75 characterization for the test vehicle. Typical number concentrations measured between 33 and 730 nm are approximately  $113,000 \text{ particles} \cdot \text{cm}^{-3}$ .

Inspection of Table 4 suggests a positive bias (median 18.4%, range 8–39%) in  $\text{PM}_{2.5}$  mass measured using PTFE filters relative to that calculated from SMPS measurements using the assumed shape-density correction factor. Because the DE is the only source of PM in the chamber, and because the SMPS has an upper size detection limit of 730 nm (adequate for DE but not for sources such as mechanical abrasion), the presence of another source of larger particles was hypothesized. Studies have shown the presence of a so-called “personal cloud” around human beings, which can add to the ambient PM concentration around the person.<sup>26,27</sup> To test whether the personal cloud was an issue, a run was conducted on April 13, 2005, both with and without a human subject. As seen for those rows in Table 4, the filter weight without the subject was 5% higher relative to the SMPS set point, compared with 62% higher for the filter weight with the subject. A MOUDI was run during both runs to identify the size-weighted concentrations. As seen in Figure 5, the difference in masses is almost entirely accounted for by large particles in the 10–18- $\mu\text{m}$  aerodynamic diameter range—a size characteristic not of combustion aerosols but rather of mechanically generated aerosols such as those observed in “personal cloud” studies listed above. Furthermore, inspection of Table 4 for the run on April 13, 2005 without the human subject shows that the relative ratio of combined OC and EC to total PM mass concentration inside the chamber is 0.97–1.00, a characteristic of DPM for this engine. In contrast, this figure is 0.70–0.73 for the run with the test subject, consistent with

other test runs in Table 4. Several experiments have also been performed with human subjects in the facility in the absence of DE; for these studies typical backgrounds ranged from 20–40  $\mu\text{g} \cdot \text{m}^{-3}$  (gravimetric analysis) whereas SMPS indicated PM loadings less than 1  $\mu\text{g} \cdot \text{m}^{-3}$ . Experiments conducted without human subjects or DE had PM loadings less than 1  $\mu\text{g} \cdot \text{m}^{-3}$  (gravimetric analysis). Thus, the presence of the subject during the exposure can have a significant impact on PM mass loading and composition and can lead to misinterpretation of results if not properly corrected or if simple gravimetric analyses are performed.

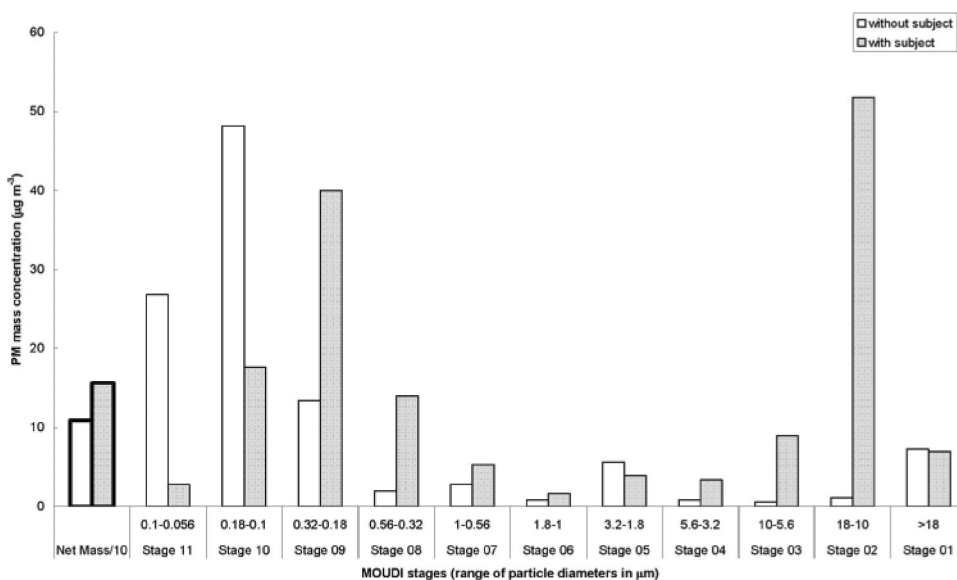
### Trace Element Measurements

Trace elements (41 in all) were sampled on 47 mm Teflo filters identical to those used for PM mass collection. Filters were then analyzed by X-ray fluorescence (XRF) at the analytical laboratories at the South Coast Air Quality Management District (SCAQMD; Diamond Bar, CA). The results are summarized in Table 6.

### Detailed Organic Composition of DPM

The detailed organic analysis of DPM was preceded by an OC-EC analysis to understand the OC/EC ratios in the DPM drawn into the chamber. Table 4 reports the OC and EC concentrations and OC/EC ratios for 12 exposure runs and 1 blank run. The ratios obtained are typical of an idling truck.<sup>1</sup>

Table 7 shows the detailed speciation of the semi-volatile and particle-phase organic compounds (n-alkanes and PAHs) found in the chamber during a typical 2-hr run at a nominal PM mass loading of 100  $\mu\text{g} \cdot \text{m}^{-3}$ . It is observed that dodecane and hexacosane are the predominant n-alkanes, whereas naphthalene and phenanthrene are the predominant PAHs. Relative PAH and n-alkane concentrations are typical of those from an idling truck.<sup>25</sup>



**Figure 5.** MOUDI particle size distribution for LAREI exposure chamber with and without human subject (run date April 13, 2005). Note high PM mass loading on stage 02 ( $d_p = 10\text{--}18 \mu\text{m}$ ).

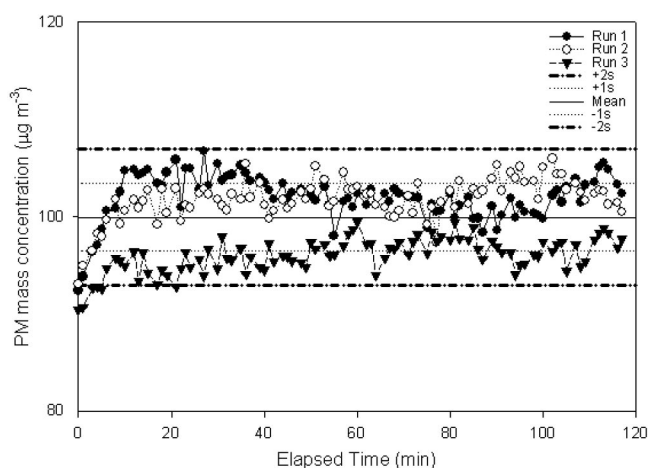
**Table 6.** Average concentrations of 41 trace elements found in the LAREI chamber ( $\mu\text{g} \cdot \text{m}^{-3}$ ).

Species	Average	SD	Species	Average	SD
S	0.385	0.075	V	<0.0038	—
Zn	0.340	0.016	Cr	<0.0095	—
P	0.248	0.020	Co	<0.0038	—
Al	0.221	0.131	Ni	<0.0038	—
Ca	0.194	0.049	Ge	<0.0094	—
Cl	0.131	0.063	As	<0.0038	—
Si	0.120	0.241	Se	<0.0038	—
Fe	0.112	0.037	Rb	<0.0095	—
Mg	0.084	0.028	Sr	<0.0038	—
Ti	0.019	0.004	Y	<0.0095	—
Cu	0.018	0.008	Nb	<0.0095	—
Cd	0.012	0.003	Pd	<0.0095	—
Mo	0.010	0.001	Ag	<0.0095	—
In	0.010	0.001	Sn	<0.0095	—
Pt	0.009	0.000	Sb	<0.0095	—
Pb	0.006	0.002	Cs	<0.0095	—
Ga	0.006	0.004	Ba	<0.0095	—
Mn	0.005	0.002	La	<0.0095	—
Au	0.005	0.003	Bi	<0.0076	—
K	<0.019	—	U	<0.0038	—
Sc	<0.019	—			

**Table 7.** Detailed speciation of semi-volatile and particle-phase organic compounds in chamber for human exposure run dated March 16, 2004.

Species (in elution order)	Concentration ( $\text{ng} \cdot \text{m}^{-3}$ )
PAHs	
Naphthalene	1048
Acenaphthylene	24
Acenaphthene	BDL
Fluorene	BDL
Phenanthrene	80
Anthracene	BDL
Fluoranthene	22
Pyrene	30
Benz(a)anthracene	BDL
Chrysene	BDL
Benzo(b)fluoranthene	BDL
Benzo(k)fluoranthene	BDL
Benzo(a)pyrene	BDL
Indeno[1,2,3-cd]pyrene	BDL
Dibenz[a,h]anthracene	BDL
Benzo[ghi]perylene	BDL
n-Alkanes	
Dodecane	6899
Tetradecane	3183
Hexadecane	3469
Octadecane	2251
Nonadecane	1519
Eicosane	901
Docosane	223
Tetracosane	107
Hexacosane	1545
Octacosane	40
Triacosane	BDL
Hexatriacontane	BDL

Notes: BDL = below detection limit.

**Figure 6.** PM mass concentrations for a typical 1-hr exposure (run date October 31, 2003). The lines marked "Mean" and "±ns" represent the average and nth standard deviation, respectively, of the three runs combined.

### Stability of Chamber Exposure

Figure 6 displays the SMPS-measured PM mass concentration for three different 2-hr mock exposure runs (with no subject in the chamber) at the  $100\text{-}\mu\text{g} \cdot \text{m}^{-3}$  exposure level. It is seen from the figure that the PM concentration is stable to within 2 standard deviations ( $\sim 7\%$ ). Plots for CO and  $\text{NO}_2$  (not shown) also indicate similar trends for gases. The large extent of overlap among the three different  $100\text{-}\mu\text{g} \cdot \text{m}^{-3}$  runs is an indicator of the run-to-run repeatability of the facility.

### Subjects' Responses to Exposure

Table 8 summarizes the responses of the 11 subjects (4 healthy, 7 asthmatic) in terms of lung function ( $\text{FEV}_1$ ) change pre- to postexposure. Mean changes were consistently small (1–2% of baseline values, clinically negligible) and were not significantly different among FA,  $\text{NO}_2$ , and DE exposures, or between healthy and asthmatic groups ( $P > 0.1$  by repeated-measures analysis of variance). Although some subjects reported increased symptoms during DE exposure compared with other conditions, no subject showed a markedly unfavorable clinical response to DE. These initial results appear consistent with findings from a European diesel exposure facility, where subjects' lung function did not change (although

**Table 8.** Mean of change in lung function ( $\text{FEV}_1$ , expressed in mL) during exposure, by subject group and atmosphere.<sup>a</sup>

Subjects	Atmosphere		
	FA	$\text{NO}_2$	DE
Healthy ( $n = 4$ )	+38 (71)	−20 (43)	+30 (107)
Asthmatic ( $n = 7$ )	−24 (302)	+59 (136)	+17 (186)
All ( $n = 11$ )	−2 (239)	+30 (115)	+22 (155)

Notes: Atmosphere differences were nonsignificant ( $P > 0.1$ ) by repeated-measures analysis of variance. Standard deviations are given in parentheses.

other evidence of unfavorable effects was found) in exposures of shorter duration but higher DE concentration.<sup>13,28</sup>

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