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Measurements of Ultrafine Particles and Other Vehicular Pollutants inside a Mobile Exposure System on Los Angeles Freeways

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ABSTRACT

A mobile exposure and air pollution measurement system was developed and used for on-freeway ultrafine particle health effects studies. A nine-passenger van was modified with a high-efficiency particulate air (HEPA) filtration system that can deliver filtered or unfiltered air to an exposure chamber inside the van. State-of-the-art instruments were used to measure concentration and size distribution of fine and ultrafine particles and the concentration of carbon monoxide (CO), black carbon (BC), particle-bound polycyclic aromatic hydrocarbons (PAHs), fine particulate matter ($PM_{2.5}$) mass, and oxides of nitrogen (NO_x) inside the exposure chamber. This paper presents the construction and technical details of the van and air pollutant concentrations collected in 32 2-hr runs on two major Los Angeles freeways, Interstate 405 (I-405; mostly gasoline traffic) and Interstate 710 (I-710; large proportion of heavy-duty diesel traffic). More than 97% of particles were removed when the flow through the filter box was switched from bypass mode to filter mode while the vehicle was driving on both freeways. The filtration system thus provides a great particulate matter exposure contrast while keeping gas-phase pollutant concentrations the same. Under bypass mode, average total particle number concentration observed inside the exposure chamber was around 8.4×10^4 and 1.3×10^5 particles cm^{-3} on the I-405 and the I-710 freeways, respectively. Bimodal size distributions were consistent and similar for both freeways with the first mode around 16–20 nm and the second mode around 50–55 nm. BC and particle-bound PAH concentrations were more than two times greater on the I-710 than on the I-405 freeway. Very weak correlations were observed between total particle number concentrations and other vehicular pollutants on the freeways.

IMPLICATIONS

Ample evidence has been published that supports the hypothesis that ultrafine particles are associated with cardiovascular effects. The highest exposure to ultrafine particles occurs during freeway travel. The present study, conducted on I-405 (mostly gasoline traffic) and I-710 (large proportion of heavy-duty diesel traffic) freeways, shows bimodal size distributions. BC and particle-bound PAH concentrations are more than two times greater on I-710 than on the I-405 freeway. Very weak correlations are observed between total particle number concentrations and $PM_{2.5}$, BC, and gaseous pollutants on both freeways.

INTRODUCTION

Epidemiological studies from all over the world have consistently linked increases in particulate matter (PM) exposure to increases in cardiovascular mortality and morbidity.[1,2] Ample evidence has been published recently that supports the hypothesis that ultrafine particles (UFPs, diameter <100 nm) are associated with cardiovascular effects because of their high deposition efficiency in pulmonary regions and their propensity to penetrate the epithelium and reach the blood and other organs.[3] The cardiovascular system is currently considered a target for UFPs, which have been shown to translocate from the lung into the systemic circulation in animal models[3–8] and in human subjects.[9,10] The presence of UFPs in the circulation has been shown to affect hemostasis.[11] This could lead to accumulation and potentially adverse reactions in critical organs such as the liver, heart, and even the brain, consistent with the hypothesis that UFPs play a role in the onset of cardiovascular disease.[6]

With improvements in particle measurement technology, it is feasible to investigate the adverse health effects of smaller particle sizes.[12] A daily mortality study in Ehrfurt, Germany was the first epidemiology study that examined and found significant associations between exposure to UFPs and mortality from respiratory and cardiovascular disease.[12] Recently epidemiological studies, dealing with short-term effects in adults and children with asthma and daily mortality, have addressed the role of UFPs.[12–15] These studies concluded that health effects were more closely associated with the number of UFPs

than the mass of the fine particles. A recent study found that time spent in traffic 1 or 2 hr earlier increased the relative risk of subsequent myocardial infarction onset two- to threefold compared with control periods.[16] Alteration of heart rate variability has been associated with particle number concentration in the general environment[17] and roadway exposure to PM mass.[18]

Very high number concentrations of UFPs (>100,000 particle/cm^3) were found near major freeways in the Los Angeles Basin.[19–22] Studies conducted in the vicinity of Interstates 405 (I-405) and 710 (I-710) in southern California found that the number concentration of UFPs was approximately 25 times greater adjacent to the freeways than upwind background levels and that pollution levels gradually decreased to near upwind (background) levels at approximately 300 m (1000 ft), downwind from the freeway during daytime. Similar decay profiles were later observed by researchers from cities around the world.[23,24] Because approximately 40% of Californians spend 1–3 hr commuting between home and work each workday, it was estimated that in-vehicle time contributes 10–50% of commuters' daily UFP exposure.[25]

Previous in-vehicle exposure studies focused primarily on coarse PM (PM_{10}) and fine PM ($PM_{2.5}$), metals, and gas-phase pollutants such as carbon monoxide (CO), ozone, oxides of nitrogen (NO_x), and volatile organic compounds.[26–29] Recently a few mobile laboratories have been developed worldwide to study UFP on-road. These studies focus on diesel emissions,[30–32] UFP formation,[33] UFP spatial and temporal variations,[34,35] and on-freeway UFP concentrations.[36] There are no systematic measurements of UFP inside vehicles and associated human health effects during commuting.

The objective of this study was to develop a mobile laboratory to facilitate concurrent measurements of near real time, in-vehicle concentration and size distribution of UFP, other traffic-related air pollutants, and cardiovascular health indicators in human subjects. This paper presents the construction and technical details of the mobile exposure laboratory and summarizes exposure levels achieved under filtered and unfiltered mode on two major Los Angeles freeways, I-405 (mostly gasoline traffic) and I-710 (large proportion of heavy-duty diesel traffic). The innovative design of the mobile exposure system and the selection of the above two freeways resulted in good exposure contrast, which is required for successful health effect studies.

METHODS

An on-freeway exposure system was designed to expose senior (60 or older) subjects to either freeway air or filtered freeway air. Each participant underwent four 2-hr freeway exposures—one on I-405 and one on I-710, and one each on the same freeways, but with filtered air. The I-710 freeway is the major truck-shipping route connecting Long Beach port to downtown Los Angeles. It has nine to ten lanes both ways and up to 25% of vehicles are heavy-duty diesel trucks.[19] The I-405 freeway is one of the busiest freeways in the United States. It has nine to ten lanes both ways and approximately 5% diesel traffic.[20]

On-Freeway Exposure Measurement System

The on-freeway exposure system was installed in a 2002 Chevrolet Express nine-passenger van. Originally the van was configured to sit nine people with seats arranged in four rows of two, two, two, and three seats each. In the first row the passenger seat and its base were removed to accommodate the high-efficiency particulate air (HEPA) filter box, air delivery fan, and airflow control valve. The HEPA filter was used to provide particle-free air for filtered runs. Health responses observed during filter air runs can be used as background/control to evaluate the actual the health effects associated with exposure to UFPs on freeways. The second row of seats occupied their original position, but the seats were installed in the human subject exposure enclosure. All seats in the third row and the two leftmost seats in the fourth row were removed to accommodate the batteries, power supply, and instrument platform. The floor of the van was flat with some stiffening corrugations.

The final layout is shown in Figure 1. The instrument platform height was 0.81 m (32"), which was 0.13 m (5") below the mouth height of the subjects when seated in the human subject exposure enclosure. This permitted short, straight sample lines from the subjects' breathing zone to all instruments, as described below. The instrument platform was constructed of 25-mm (1-in.)-thick plywood and was 1.12 m wide by 1.58 m long. It was attached with neoprene vibration absorbing mounts to a table frame. The table frame was attached to the van floor. The battery box, inverter, and charger were mounted together, as a separate unit, to the van floor beneath the instrument platform (Figure 1). The instrument operator sat in the rear seat and had access to all instruments and the power supply. The operator could communicate with the driver and subjects by a wired intercom system.

The exposure enclosure was designed to accommodate one or two human subjects for environmental health studies. It was constructed of 6-mm (0.25-in.)-thick static-dissipative acrylic sheets attached to an aluminum angle frame. This static-dissipative material was used to prevent any buildup of charge on the enclosure walls that might lead to a loss of particles. In addition, the ductings to the enclosure and the sample lines from the enclosure to the instruments were either metal, plastic with conductive paint, or conductive rubber tubing. The gross dimensions of the enclosure were 1.05 × 1.25 × 1.23 m high. The enclosure had a flow distribution manifold on top with 20 25-mm holes and three layers of window screen at the outlet to provide an evenly distributed gentle flow of air over the subjects. Air exited the exposure enclosure through two 76-mm (3-in.) diameter ducts, located on the back wall beneath each seat. Sampling lines came from a 150- × 200-mm (6- × 8-in.) sampling panel located on the back wall of the enclosure, between the two subjects approximately 0.4 m from their mouths. Subject access was provided by a removable one-piece door, which sealed to a steel frame around the enclosure doorway by magnetic seals on the enclosure door.

Measured Environmental Parameters

Table 1 summarizes environmental parameters and associated measurement methods used in the on-freeway exposure system. A panel located at the height of subjects'

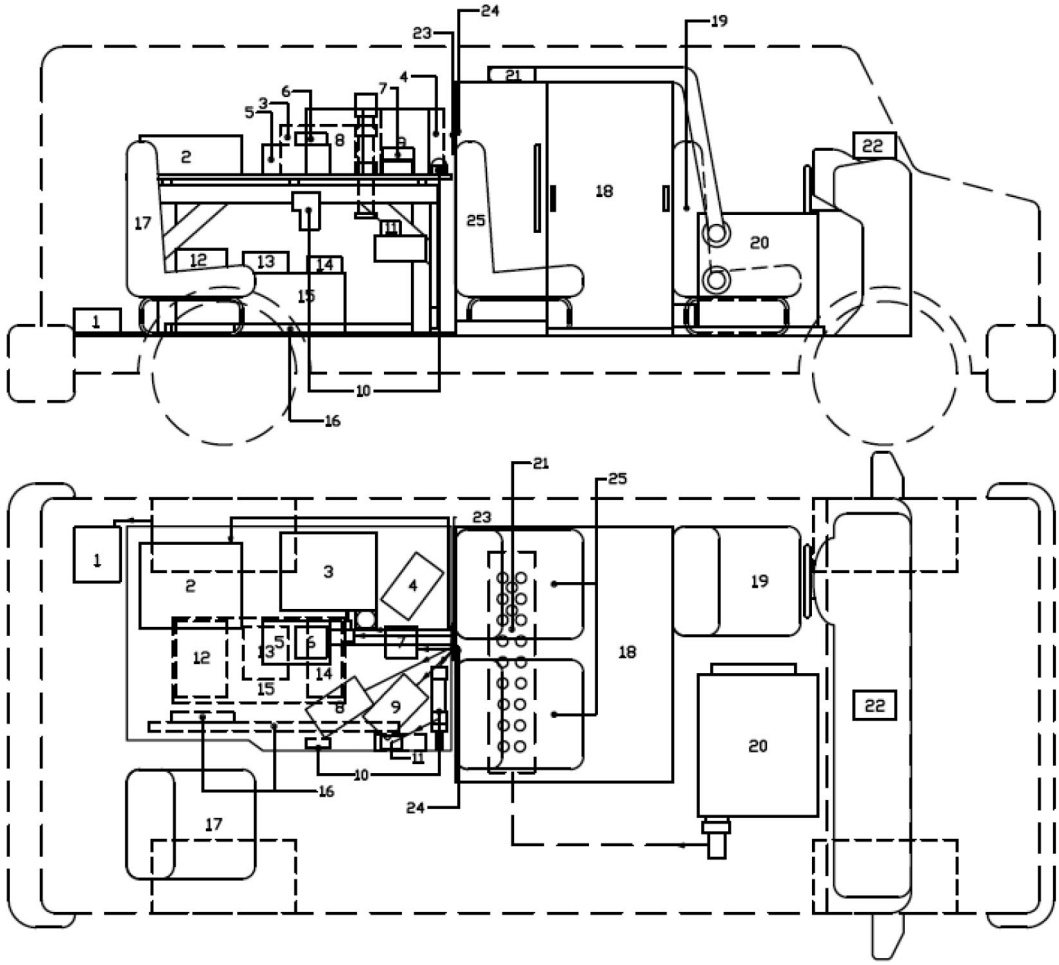

Figure 1. Schematic diagram of the exposure and measurement system (upper panel: side view, lower panel: top view). (1) NO_x pump, (2) NO_x analyzer, (3) SMPS (electrostatic classifier), (4) SMPS (CPC), (5) PAS 2000, (6) DustTrak ($PM_{2.5}$), (7) DustTrak (PM_{10}), (8) aethalometer, (9) CPC 10, (Q-Trak 11), BGI pump for PEM filter, (12) inverter, (13) switch box, (14) battery charger, (15) battery box, (16) power strips, (17) operator seat, (18) exposure chamber, (19) driver seat, (20) filter box and fans, (21) chamber inlet, (22) camcorder, (23) chamber sampling panel, (24) PEM 2.5 filter, and (25) subject seats.

breathing zone in the back wall of the exposure chamber was used to hold sampling lines. No consideration was given to isokinetic sampling because wind speed was minimal inside the exposure chamber. Stainless steel and flexible conductive tubing (Part 3001940, TSI Inc.) were used for sample inlet lines for all particle-measuring instruments to avoid particle losses due to electrostatic forces. To minimize the residence time and particle loses, all of the sampling lines were kept horizontal, straight, and at minimum length. Sampling line losses due to diffusion were calculated to be less than 5% for 10-nm particles for all instruments.[37]

A scanning mobility particle sizer (SMPS; 3936L85, TSI Inc.) was used to measure fine and UFP size distribution in the size range of 7.6 to 289 nm. This instrument consists of two components, a Model 3080 Electrostatic Classifier (with a Model 3081 LDMA) to select particles of a given size and a water-based condensation particle counter (CPC; 3785, TSI Inc.) to count the particles. The newly developed water-based CPC[38] is essential for in-vehicle exposure assessment of particle number concentrations. Butanol-based CPCs are well known to emit butanol vapors and are not suitable for human health studies in confined environments. A second water-based CPC (3785, TSI Inc.) was used to measure total particle number concentration in a size range of 5–6 nm to a few micrometers at 1-sec intervals. The high-time-resolution CPC data captured the rapid change in particle number concentration during freeway travel and provided a measure of total particle number concentrations.

A TSI DustTrak photometer (Model 8520 TSI, Inc., St. Paul, MN) with a $PM_{2.5}$ inlet impactor was used to continuously monitor particle mass concentration. Powered by an internal battery, the DustTrak samples air at a constant flow rate of 2 L/min by means of a built-in diaphragm pump. The sampled airstream passes through a light scattering optical sensing component. The detected signal is processed by lock-in circuitry followed by high-resolution digitization. The DustTrak covers a concentration range of 1 $\mu g/m^3$ to 400 mg/m^3 and provides an auto-ranging digital display for both real-time and time-averaged concentrations. DustTraks data were calibrated against simultaneous gravimetric measurements using a personal environmental monitor (PEM) that collects $PM_{2.5}$ mass.

Table 1. Environmental measurement parameters and methods.

	Species/Parameter	Instrument	Detection Limit	Flow Rate (L/min)	Amps (110 V)	Weight (kg)	Dimension (L × W × H; cm)	Response Time (sec)
1	UFP size distribution	TSI SMPS	Single particle	1.0	2	23.2	46 × 41 × 41	100 up, 20 down scan
2	Particle number concentration	TSI CPC	Single particle	1.0	1	5.5	28 × 16 × 31	1
3	$PM_{2.5}$ real-time mass	TSI DustTrak 8520	1 μg/m^3	2	Battery	3	41 × 33 × 19	60
4	BC	Portable aethelometer AE-42	1 μg/m^3	5	Battery	6.5	30 × 21 × 32	60
5	CO, CO_2 real-time, temperature, relative humidity	TSI Q-Trak	1 ppm, 0.1 °C, 1%	n/a	Battery	0.55	18 × 11 × 4	60
6	Particle-bound PAHs	EcoChem PAS 2000	ng/m^3	2	0.5	9	32 × 24 × 13	30
7	NO, NO_2, NO_x	API 200AU with pump	~1 ppb	1	2.5	29.5	61 × 43 × 18	1
8	Location and speed	Camcorder, Garmin GPS18	10 m	n/a	Battery	0.1	6 × 6 × 2	1
9	Two laptops	Compaq	n/a	n/a	Battery	5.4	33 × 23 × 3	n/a

Notes: NO = nitric oxide; NO_2 = nitrogen dioxide; n/a = not applicable.

A Magee scientific aethelometer model AE-42 was used to measure the elemental carbon concentration in near-real time. This device measures the attenuation of a beam of light transmitted through a filter while the filter is continuously collecting an aerosol sample. The instrument measures particulate black carbon (BC) in the near infrared (λ = 880 nm) using a solid-state source. The rate of accumulation of BC is proportional to both the BC concentration in the airstream and the flow rate. A sample cycle of 1 min was used. Data were continuously logged into an internal data logger. The factory calibration was used for this instrument.

Particle-bound polycyclic aromatic hydrocarbons (PAHs) were measured in near-real time with an EcoChem Analytics PAS 2000. The instrument measures total, nonspecific PAHs (with three rings or more) primarily adsorbed to the particle's surface. It is sensitive in the ng/m^3 range. It has built-in data logging storage for 8000 data points (date, time, value). Manufacture calibrations were used for this equipment. Measured PAH concentrations were found to be comparable with previously reported levels on Los Angeles freeways.[36]

NO_x were measured in near-real time with a Teledyne API model 200AU chemiluminescent trace analyzer with an external pump. The instrument covers a range from 5 ppb to 2 ppm. It was calibrated with span gas in the laboratory.

The concentration of CO and carbon dioxide (CO_2) were determined by a Q-Trak indoor air quality monitor (Model 8550, TSI Inc.). An electrochemical cell is used for detecting these species. The detection limit for CO and carbon dioxide (CO_2) is 1 ppm. Data were continuously logged into an internal data logger and then downloaded to a personal computer. A time resolution of 1 min was used. The instrument was calibrated by sampling known concentrations of CO and CO_2. The Q-track was also used to monitor in-vehicle temperature and relative humidity. Data reduction and analysis of the Q-Trak output was conducted by the TrakPro software (version 3.33, TSI Inc.).

The speed, direction, and location of the vehicle were monitored every a few seconds with a Garmin GPS-18 global positioning system linked directly to the instrument laptop computer. City select North American map data were loaded onto the laptop to provide a trace of the vehicle's path during each run. An 8-mm, high-resolution video recorder (camcorder) was mounted on the dashboard of the test vehicle to record traffic conditions during all measurement periods. It has been shown that a vehicle's occupants are primarily exposed to the exhaust of neighboring vehicles, particularly those directly ahead of the occupant's vehicle.[36,39] The camcorder was set to a wide angle to view as much of the freeway traffic as possible.

All instruments were powered by a battery-operated power supply system. The four batteries were sealed 97 A · hr, 12-V, lead acid, gel-type marine batteries (West Marine, Sea-Gel Deep Cycle 97) supplying power to a 1 kW pure sine wave inverter (120 V output) (Xantrex model 1000) with an associated charging system (Xantrex model TrueCharge 40+). The batteries were housed in a battery box secured to the vehicle deck behind the subject

enclosure and under the instrument platform. The inverter, switch box, and charger were mounted on top of the battery box (Figure 1). The unit weighs approximately 160 kg (350 lbs) so Teflon skids were mounted on the bottom to facilitate installing and removing it from the van.

RESULTS AND DISCUSSION

The following sections present the overall performance of the exposure system and summarize exposure levels achieved during the first 32 exposure runs (8 subjects). Associated health data and their relationship to environmental measurements will be presented in future publications.

Exposure System Performance

The fan box was designed to draw air from the inside of the van and deliver it into the exposure chamber. The air change rate (ACR) of the exposure chamber was calculated by measuring the airflow rate out of the chamber (1840 L/min for filtered air and 2134 L/min for unfiltered air) and the volume of the chamber (1455 L) and was found to be 84 hr^{-1} for filtered air and 88 hr^{-1} for unfiltered air. These ACRs are in agreement with our recent observations inside passenger vehicles driving on Los Angeles freeways.[25] The HEPA filter system was designed to allow air to either pass through the filter or bypass it and flow directly to the exposure chamber. The particle loss through the fan box under bypass mode (air not filtered) was determined by measuring the ambient concentration and size distribution of particles and those at the outlet of the filter box. Penetration ratio was calculated for each size range and also for the entire size range. Five to ten percent of losses were observed in the nanometer size range because of the greater diffusion effect. No losses were observed for particles in the size range of 175–220 nm. Overall, there was a less than 5% loss of total particles from ambient to the exposure chamber. When the HEPA filter system was set to filter mode, more than 97% of total particles were removed as discussed in the next section. Under normal operation the power supply system can power all the instruments for about 3 hr.

Summary of Measured Pollutant Concentrations

Figure 2 presents a 1-hr typical time-series plot for measured pollutants inside the exposure chamber while driving on the I-710 freeway under bypass filter mode. Scanswere averaged over 1-min intervals. There was a 10-min break in BC concentration (Figure 2c) because of an aethalometer automatic tape-advancing event. In general, two to three tape-advancing events were usually observed on the diesel-truck-dominated I-710 freeway whereas such events usually occurred only once on the gasoline-vehicle-dominated I-405 freeway during typical 2-hr exposure runs. This is likely due to the much greater BC concentrations from diesel emissions on the I-710 freeway. Strong concentration variations were observed for all measured pollutants. There is usually a factor greater than 5 between the highest and the lowest concentrations.

Figure 3 shows the histograms of major air pollutant concentrations based on 1-min averaged observations. The black bars and the gray bars represent the I-405 and I-710 freeway, respectively. For each figure, the *x*-axis is

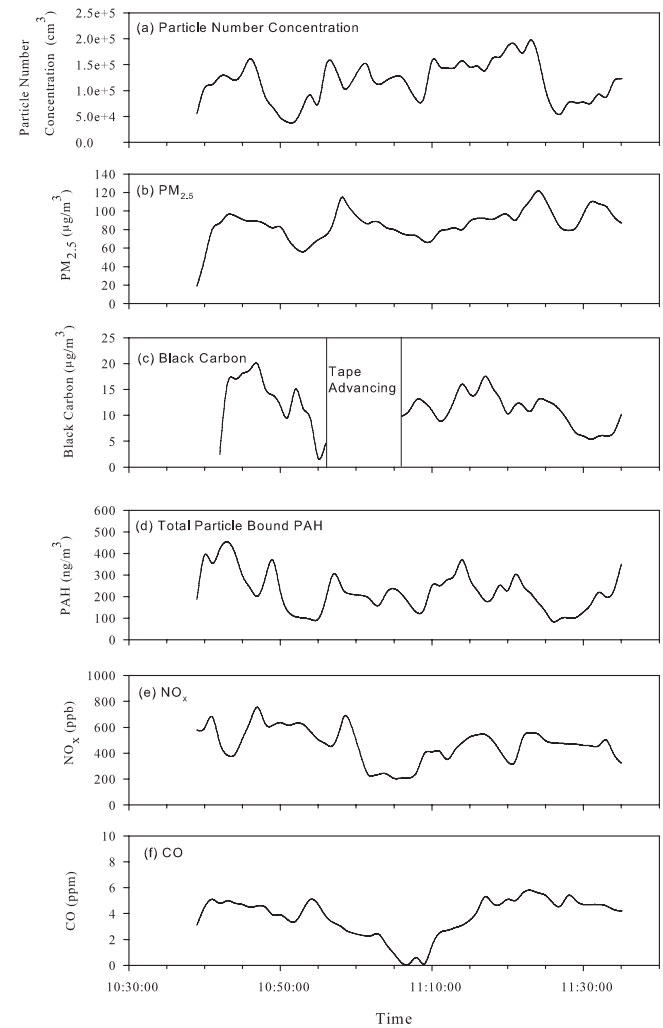

Figure 2. Typical 1-hr time-series plots for pollutant concentrations measured inside the exposure chamber on the I-710 freeway. (a) Particle number concentration, (b) $PM_{2.5}$, (c) BC, (d) total particle-bound PAH, (e) NO_x, and (f) CO.

the measured pollutant concentrations in linear scale and the *y*-axis is observation frequencies in percentage. In general, all pollutant concentrations show a dominant mode with right-skewness. The modes of pollutant concentrations measured on the I-405 freeway were usually to the left of those measured on the I-710 freeway except for CO. This suggests exposure levels on the I-405 freeway were generally lower than those on I-710. This is particularly true for total particle number concentrations (Figure 3a), BC concentrations (Figure 3c), particle-bound PAH concentrations (Figure 3d), and NO_x concentrations (Figure 3e).

Table 2 summarizes run-averaged concentrations of major environmental parameters measured on the I-405 and I-710 freeways under both filtered and unfiltered conditions. Standard deviations are given in the parentheses. Concentrations of all measured pollutants were higher on the diesel freeway than on the gasoline freeway. The highest 1-min averaged particle number concentration, 730,000 particles per cm^3, was observed on the I-710 freeway. The highest 1-min averaged $PM_{2.5}$ concentration, 150 $\mu g/m^3$, was also observed on the I-710 freeway

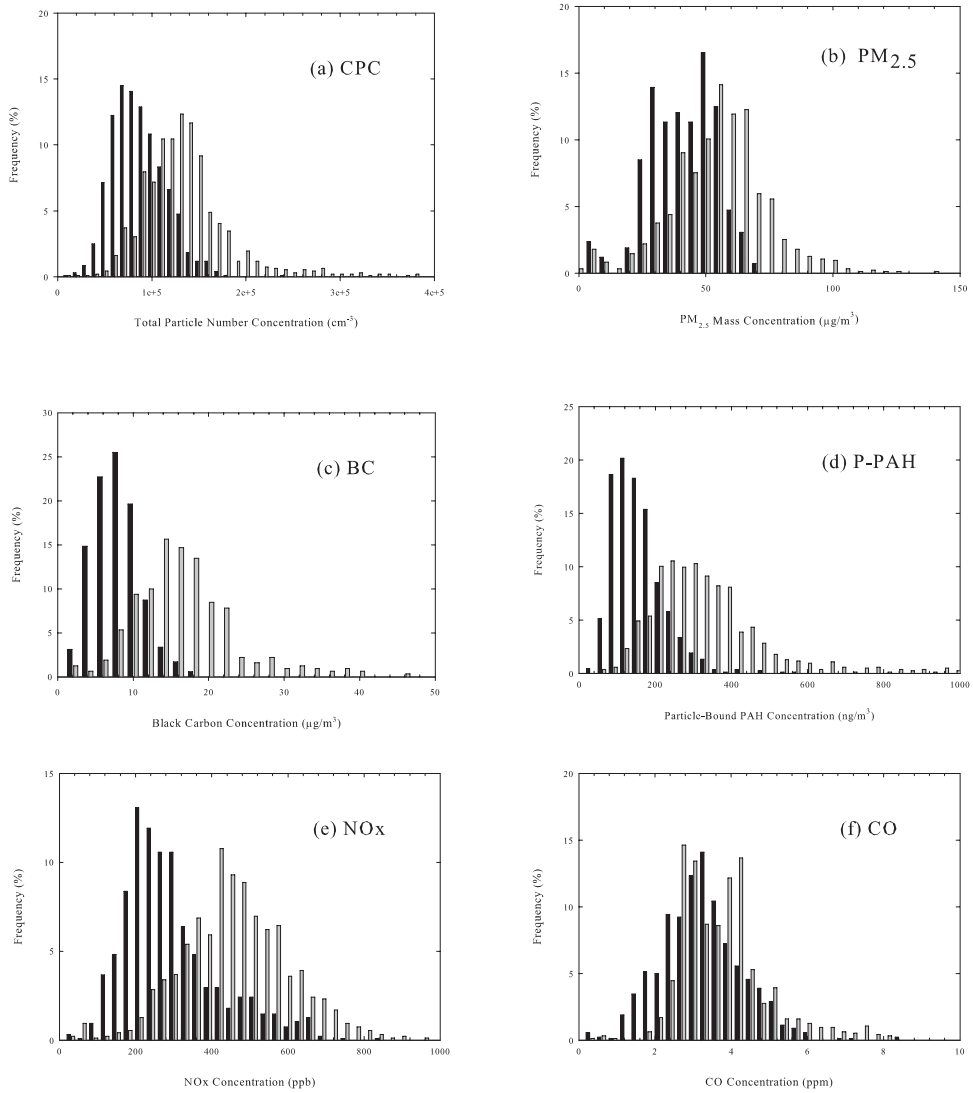

Figure 3. Histogram of measured (a) total particle number, (b) $PM_{2.5}$, (c) BC, (d) particle-bound PAH, (e) NO_x, and (f) CO concentrations inside the exposure enclosure while driving on the I-405 and I-710 freeways under unfiltered mode. Black bars represent I-405. Gray bars represent I-710.

on the same day but at a different time. For $PM_{2.5}$, BC, particle-bound PAH, and NO_x, concentrations on the I-710 freeway were at least two times greater than those on the I-405 freeway. Under filtered conditions, for both freeways, significant decreases were observed for particle-phase pollutants but not for gas-phase pollutants.

UFP Size Distributions on Freeways

Typical time-resolved UFP size distributions measured inside the exposure chamber on the two freeways are shown as contour plots in Figure 4, where the *x*-axis presents the time at which data were collected, the *y*-axis is the particle size in log scale, and the color intensity indicates normalized particle number concentration (dN/dLogDp) for a given size at a given time. The same concentration scale was used for both freeways. Data were collected when the vehicle ventilation system was set to air conditioning on and recirculation off. Previously we found vehicle ventilation settings affect the penetration characteristics of UFPs into vehicles.[25] The current ventilation settings were chosen to ensure subjects' comfort inside the exposure chamber. The same ventilation settings were used for all runs.

In general, particle number concentrations were lower on the I-405 freeway than on the I-710 freeway, as indicated by more green color in Figure 4b than in Figure 4a. For both freeways, hot spots usually occurred around 10–30 nm corresponding to a primary nuclei mode. There were periods between hot spots when particle number concentrations were lower on both freeways. This happened because each 2-hr exposure run typically consisted of two round-trip loops on the freeway. The lower concentration periods correspond to the turning points at which we drove off the freeways to use local streets to get back on the freeway in the opposite direction. In Figure 4a, a second mode around 60–100 nm usually occurred suggesting bimodal size distributions on the I-710 freeway. This is in agreement with what we previously observed inside passenger vehicles on the I-710 freeway.[25]

Table 2. Summary of major environmental parameters collected under different exposure conditions.

Environmental Parameters	Exposure Conditions			
	I-710 Unfiltered	I-710 Filtered	I-405 Unfiltered	I-405 Filtered
Particle number concentration (cm^{-3})	134,000 (27700)	3800 (1000)	83,800 (14700)	2100 (319)
$PM_{2.5}$ ($\mu g/m^3$)	52.3 (11.6)	5.0 (3.6)	43.4 (16.9)	2.6 (3.6)
BC ($\mu g/m^3$)	14.7(2.5)	0.4 (0.09)	7.1 (1.1)	0.2 (0.09)
Particle-bound total PAH (ng/m^3)	319 (54.8)	5.3 (1.3)	141 (16.7)	2.8 (0.4)
NO_x (ppb)	432 (66.3)	385 (112)	267 (114)	272 (96)
CO (ppm)	3.7 (0.9)	2.6 (1.0)	3.1 (0.6)	2.9 (1.9)
Temperature (°C)	21.0 (4.0)	22.1 (4.8)	20.1 (4.3)	20.2 (5.1)
Relative humidity (%)	32.7 (6.9)	29.3 (9.2)	32.9 (8.0)	35.0 (9.0)

Notes: Standard deviations are given in parentheses.

On the I-405 freeway, the primary mode was also around 10–30 nm, but the second mode seems broader and less obvious. These plots will be used later with time-resolved health indicators to study the acute effect of UFPs in different size ranges.

Exposure Contrast

Large exposure contrasts are critical in the success of health effects studies. In this study, the innovative design of the mobile exposure system and the selection between a gasoline- and a diesel-dominated freeway led to good exposure contrasts as discussed in the following sections.

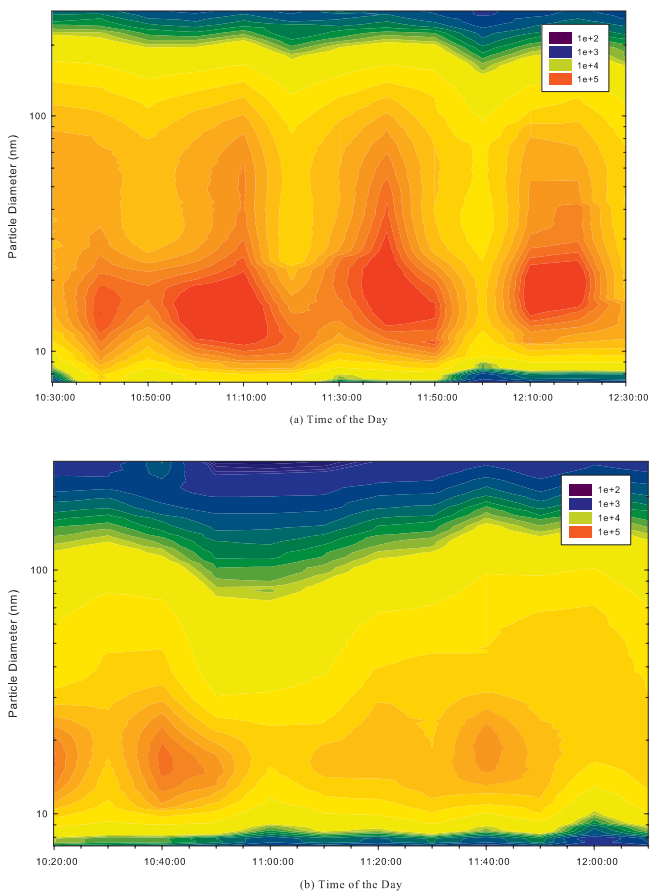

Figure 4. Typical contour plot of UFP number-based size distribution during 2-hr exposure runs under unfiltered mode on the (a) I-710 and (b) I-405 freeways.

I-405 Versus I-710 Freeways. As shown in Table 2 and Figures 3 and 4, higher pollutant concentrations were observed on the I-710 freeway, which has much more diesel traffic. This is in agreement with a previous study that reported a linear relationship between the number of diesel trucks and particle number concentrations on freeways.[36] The UFP size distribution characteristics on the two freeways became even clearer when averaged data were plotted and fitted to bimodal lognormal size distributions in Figure 5, a and b, for unfiltered and filtered conditions, respectively. The data for each freeway were fitted to the sum of two lognormal distributions using a modified SigmaPlot 2000 lognormal six parameter fitting procedure.[40] For each mode, the geometric mean diameter, μ_g, and the geometrical standard deviation, σ_g, are also shown in Figure 5, a and b. Error bars in Figure 5a indicate 1 standard deviation. Error bars in Figure 5b were not included because they were relatively large compared with the mean, thus making it difficult to distinguish between the two freeways. For both freeways under both filtered and unfiltered conditions, a primary mode around 17–20 nm and a secondary mode around 50–55 nm were observed. The geometrical standard deviations for each mode were also comparable. These size distribution characteristics were very consistent as indicated by the smooth error bars in Figure 5a. They were also in good agreement with previously reported in-cabin particle size distributions observed with passenger vehicles on the same freeways.[25] Thus, subjects that underwent exposure runs with unfiltered air were exposed to similar levels of UFPs found inside passenger vehicles under normal driving conditions.

The real difference between the two freeways is the particle concentration. In fact, if we multiply the I-405 freeway data with a factor to match up the total particle number within the measured size range on the two freeways, we obtain almost identical size distribution on both freeways. These results are in contrast to our previous studies conducted on roadsides near freeways, which found that UFP number concentrations and size distributions were not significantly different between I-405 and I-710.[19] This difference may be because near-roadway, as compared with on-roadway, measurements reflect an integrated effect of all the passing vehicles. Although I-710 has a greater fraction of diesel traffic, with each heavy-duty diesel truck accounting for the space of two to three

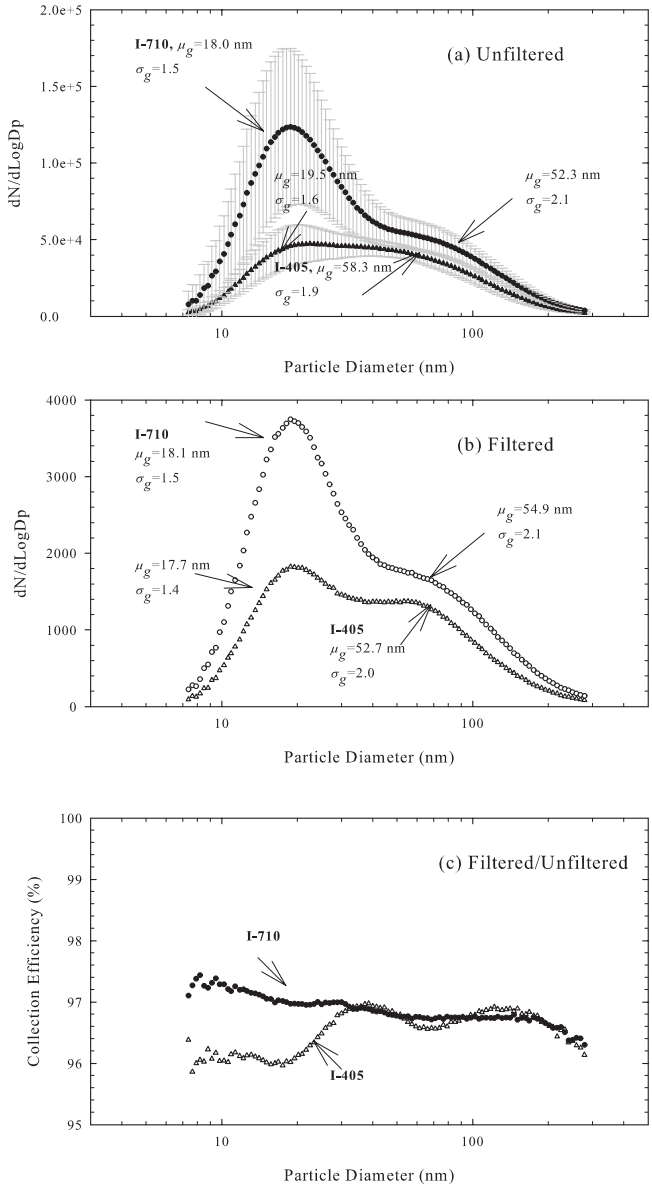

Figure 5. Fitted bimodal particle size distribution inside the exposure chamber under (a) unfiltered and (b) filtered conditions while driving on the I-710 and I-405 freeways, and (c) size-specified collection efficiencies of the filter box. Data were averaged for all applicable runs. Error bars indicate 1 standard deviation.

gasoline vehicles and emitting only about two times more UFPs than gasoline engines,[41] when both freeways were functioning at their full capacity little difference in particle concentrations was observed. However, the data presented in this paper are more likely to be affected by the vehicle that the van is following. Although diesel trucks only comprise approximately 25% of the traffic on the I-710 freeway, they occupied approximately 50% of freeway space. Thus, on the I-710 freeway, the van had a much greater chance to be within the emission plume of a heavy-duty diesel truck than on the I-405 freeways, which translated into a much stronger signature of diesel emissions. A similar scenario has been observed with passenger vehicles on the two freeways.[25]

Filtered Versus Unfiltered Runs. Data presented in Table 2 indicate that more than 97% of the particles were removed on a total number basis under filtered conditions for both freeways. Figure 5c presents the averaged size-specified filter box collection efficiency on both the I-710 and I-405 freeways. In general, the filter box provided approximately 97% collection efficiency for all measured particle sizes. The collection efficiency is slightly lower on the I-405 and slightly higher on the I-710 for particles less than 20 nm in size; however, the differences are within 2%. For particles larger than 30 nm, the collection efficiencies are very similar for both freeways.

As shown in Table 2, BC concentrations were also reduced to 2–3%. More than 98% of total particle bound PAH was removed. The particle-phase pollutant concentrations were all statistically significantly lower under filtered conditions than unfiltered conditions for each freeway. No significant decreases were observed for gas-phase pollutants on the I-405 freeway. Slight decreases of gas-phase pollutant concentrations were observed on the I-710 freeway, but were not statistically significant. In general, NO_x and CO concentrations were comparable with and without the filter. In conclusion, the HEPA filter box works the way we expected in providing excellent PM concentration contrast between exposure and control runs.

UFPs Versus Gas-Phase Pollutants. The matrix of Pearson correlation coefficients among all measured pollutants measured on both the I-710 and I-405 freeways are given in Table 3. Each coefficient is based on more than 900 1-min observations. Associated P values are also given in parentheses below each coefficient. In generally, weak to poor correlations were observed among measured pollutants inside the chamber. The strongest correlations occurred between BC and particle-bound total PAHs for both freeways. This is not surprising because the operating principle for the two instruments used, aethelometer and Echochem PAS, are similar, being based on adsorption of incident light. Very weak or no correlations were observed between particle number concentrations and any other pollutants similar to previously reported data from chasing studies.[42] This is in contrast to previously reported strong correlation between normalized particle number concentrations and BC and CO concentrations near freeways.[19,20] The strong correlations reported previously were spatial correlations between traffic pollutants undergoing similar atmospheric dispersion processes. Once normalized to their maximum concentration, the main factors determining their correlations were the wind speed and direction, which were the same for all pollutants. Correlations presented in Table 3 are temporal correlations. They reflect the source strength of surrounding vehicles on measured pollutants under various driving conditions. The poor correlations suggest that vehicle and driving conditions affect particle-phase and gas-phase pollutant emissions differently. It also indicates that no simple surrogate can be used in place of particle number concentrations for estimating commuters' exposure to UFPs.

Table 3a. Pearson correlation coefficients among measured pollutants inside the exposure chamber under unfiltered mode on the I-710 freeway.

	Particle Number Concentration	$PM_{2.5}$	BC	Particle-Bound PAH	NO_x	CO
Particle number concentration	1.00	–	–	–	–	–
	–	–	–	–	–	–
$PM_{2.5}$	−0.066	1.00	–	–	–	–
	(0.044)	–	–	–	–	–
BC	0.14	0.325	1.00	–	–	–
	(<0.001)	(<0.001)	–	–	–	–
Particle-bound PAH	0.156	0.118	0.612	1.00	–	–
	(<0.001)	(<0.001)	(<0.001)	–	–	–
NO_x	−0.334	−0.181	0.336	0.325	1.00	–
	(<0.001)	(<0.001)	(<0.001)	(<0.001)	–	–
CO	−0.16429	0.305	0.122	0.152	0.072	1.00
	(<0.001)	(<0.001)	(0.001)	(<0.001)	(0.026)	–

Notes: P values are given in parentheses.

UFPs Versus $PM_{2.5}$. The poor correlations between total particle number concentration and $PM_{2.5}$ mass concentration became even clearer in Figure 6. Paired 1-min $PM_{2.5}$ and total particle number concentrations were plotted for each freeway with respect to different sampling date. R^2 for each run are also given in the figure next to the date when the data were collected. Although particle number concentrations tend to be centered around 1.3×10^5 particles/cm^3 for I-710 and 8.4×10^4 particles/cm^3 for I-405, corresponding $PM_{2.5}$ concentrations range from approximately 20 to 120 μg/m^3. The R^2 between particle number concentration and $PM_{2.5}$ mass concentrations varies from 0.001 to 0.115 on different freeways and on different sampling days. These poor correlations suggest that there is no simple linear relationship between particle number emission factors and particle mass emission factors from vehicles. It also suggests that particles of different size range behave differently when they penetrate into the in-cabin environment. In addition, the after-treatment systems may also play a role. For example, if a diesel truck is equipped with a particle filter, the soot mode will be removed and nucleation mode particles will dominate. These particles contribute to higher number concentrations but not mass. On the other hand, if the soot mode is present, it may at least prevent the formation of nucleation mode particles and result in lower number but higher $PM_{2.5}$ mass. The poor correlation between particle number concentration and $PM_{2.5}$ mass may provide an opportunity to study their health effects separately.

CONCLUSIONS

A nine-passenger van was modified with a HEPA filter unit to facilitate an on-freeway PM health effects study. More than 97% of particles were removed when the flow through the filter box was switched from bypass mode to filter mode while the vehicle was traveling on the freeways. Thus, the filter box provides a great PM exposure contrast while keeping gas-phase pollutant concentrations about the same. Pollutant concentrations were monitored in human subjects' breathing zone, which includes concentration and size distribution of fine and UFPs and the concentration of CO, BC, particle-bound PAHs, $PM_{2.5}$ mass, and NO_x. Average total particle number concentration observed inside the enclosure when the filters unit was in bypass mode was around 8.4×10^4 and 1.3×10^5 particles $\cdot$ cm^{-3} on the I-405 and the I-710 freeway, respectively. The highest particle number concentration observed was over 4×10^5 particles $\cdot$ cm^{-3}. Bimodal size distributions were typical for both freeways with the first mode around 12–20 nm and the second mode around 50–100 nm. BC and particle-bound PAH

Table 3b. Pearson correlation coefficients among measured pollutants inside the exposure chamber under unfiltered mode on the I-405 freeway.

	Particle Number Concentration	$PM_{2.5}$	Black Carbon	Particle-Bound PAH	NO_x	CO
Particle Number Concentration	1.00	–	–	–	–	–
	–	–	–	–	–	
$PM_{2.5}$	0.041	1.00	–	–	–	–
	(0.246)	–	–	–	–	–
BC	0.264	0.240	1.00	–	–	–
	(<0.001)	(<0.001)	–	–	–	–
Particle-bound PAH	0.184	−0.050	0.420	1.00	–	–
	(<0.001)	(0.167)	(<0.001)	–	–	–
NO_x	0.155	−0.049	0.204	0.186	1.00	–
	(<0.001)	(0.158)	(<0.001)	(<0.001)	–	–
CO	0.010	0.345	0.0308	−0.148	0.095	1.00
	(0.771)	(<0.001)	(0.398)	(<0.001)	(0.006)	–

Notes: P values are given in parentheses.

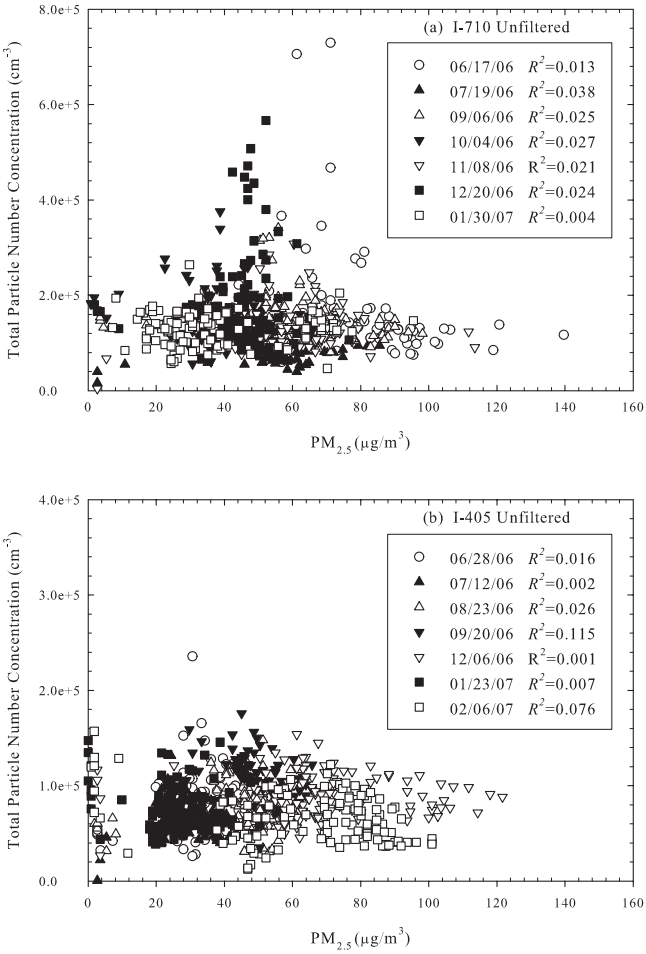

Figure 6. Correlations between $PM_{2.5}$ and total particle number concentrations measured inside the exposure chamber on the (a) I-710 and (b) I-405 freeways under unfiltered mode. Dates and coefficient of determination (R^2) are also shown.

concentrations were more than two times greater on I-710 than on the I-405 freeway because of greater diesel emissions. Very weak correlations were observed between total particle number concentration and other vehicular pollutants on freeways. This system can be used for clinical, environmental studies that investigate the relationships between measures of gas- and particle-phase exposure and short-term health response associated with freeway travel.

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