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Study of ultrafine particles near a major highway with heavy-duty diesel traffic

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Abstract

Motor vehicle emissions usually constitute the most significant source of ultrafine particles (diameter $<0.1\,\mu m$) in an urban environment. Zhu et al. (J. Air Waste Manage. Assoc., 2002, accepted for publication) conducted systematic measurements of the concentration and size distribution of ultrafine particles in the vicinity of a highway dominated by gasoline vehicle. The present study compares these previous measurements with those made on Interstate 710 freeway in Los Angeles. The 710 freeway was selected because more than 25% of the vehicles are heavy-duty diesel trucks. Particle number concentration and size distribution in the size range from 6 to 220 nm were measured by a condensation particle counter and a scanning mobility particle sizer, respectively. Measurements were taken at 17, 20, 30, 90, 150, and 300 m downwind and 200 m upwind from the center of the freeway. At each sampling location, concentrations of carbon monoxide (CO) and black carbon (BC) were also measured by a Dasibi CO monitor and an Aethalometer, respectively. The range of average concentration of CO, BC and total particle number concentration at 17 m was 1.9–2.6 ppm, 20.3–24.8 $\mu g/m^3$, 1.8×10^5–$3.5\times10^5/cm^3$, respectively. Relative concentration of CO, BC and particle number decreased exponentially and tracked each other well as one moves away from the freeway. Both atmospheric dispersion and coagulation appears to contribute to the rapid decrease in particle number concentration and change in particle size distribution with increasing distance from the freeway. Average traffic flow during the sampling periods was 12,180 vehicles/h with more than 25% of vehicles being heavy-duty diesel trucks. Ultrafine particle number concentration measured at 300 m downwind from the freeway was indistinguishable from upwind background concentration. These data may be used to estimate exposure to ultrafine particles in the vicinity of major highways.

Keywords: Ultrafine particles; Freeways; Diesel; Carbon monoxide; Black carbon

1. Introduction

Epidemiological data from air pollution studies have shown a consistent relationship between increases in particulate matter (PM) exposure and contemporary increases in mortality and morbidity (Schwartz, 1991; Dockery et al., 1993; Pope et al., 1995; Vedal, 1997). However, the underlying biological causes of the health effects of PM exposure and the correct measurement metric are unclear. For example, it is not clear whether the mass concentration (Osunsanya et al., 2001) or the number concentration (Peters et al., 1997; Penttinen et al., 2001) is most important in causing these adverse

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PM health effects. Currently, there are several hypotheses used to explain the association of PM and observed adverse health effect. One argues that particle surface contaminants, such as transition metals, contribute towards ill health (Fubini et al., 1995; Gilmour et al., 1996), wherein the ultrafine particles are thought to act as vehicles for those contaminants, initiating local lung damage when the particles deposit on the epithelial surfaces. Another hypothesis is that the physical characteristics (e.g. number, size, shape, aggregation properties) are important in producing health effects (Bérubé et al., 1999). Particle shape and size are critical factor controlling where the inhaled particles deposit in the various regions of human respiratory system by the complex action of aerosol deposition mechanisms (Hinds, 1999).

Recent toxicological studies have concluded that ultrafine particles (diameter <100 nm) are more toxic than larger particles with the same chemical composition and at the same mass concentration (Ferin et al., 1990; Oberdörster, 1996, 2001; Donaldson et al., 1998, 2001; Churg et al., 1999; Brown et al., 2000). Currently, however, only the mass concentration of PM <10 μm in aerodynamic diameter (PM_{10}) and <2.5 μm ($PM_{2.5}$) are regulated. Information about ultrafine particles is usually not available. In fact, even though ultrafine particles represent over 80% of particles in terms of number concentration in an urban environment (Morawska et al., 1998a, b), the less numerous but much heavier particles of the accumulation (0.1–2 μm) and coarse (2.5–10 μm) modes dominate mass concentration measurements. Thus, number concentration, together with the size distribution of ultrafine particles, is needed to better assess ambient air quality and its potential health effects.

Emission inventories suggest that motor vehicles are the primary direct emission sources of fine and ultrafine particles to the atmosphere in urban areas (Schauer et al., 1996; Shi et al., 1999; Hitchins et al., 2000). Although traffic-related air pollution in urban environments has been of increasing concern, most studies have focused on gaseous pollutants, total mass concentration, or chemical composition of particulate pollutants (Kuhler et al., 1994; Clairborn et al., 1995; Williams and McCrae, 1995; Janssen et al., 1997; Roorda-Knape et al., 1998a, b; Wrobel et al., 2000). Booker (1997) found that particle number concentration was strongly correlated with vehicle traffic while PM_{10} was essentially uncorrelated with traffic. Since the majority of particle number from vehicle exhaust are in the size range 20–130 nm for diesel engines (Morawska et al., 1998a, b) and 20–60 nm for gasoline engines (Ristovski et al., 1998), it is important and necessary to quantify ultrafine particle emission levels, and to determine ultrafine particle behavior after emission as they are transported away from the emission source—busy roads and freeways.

Morawska et al. (1999) measured the horizontal and vertical profiles of submicrometer particulates (16–626 nm) near a major arterial route in the urban area of Brisbane, Australia. They found, with the exception of measurements in close proximity to the road (about 15 m), that the horizontal ground-level profile measurements did not show statistically significant differences in fine particle number concentration for up to 200 m distances away from the road. Hitchins et al. (2000) examined the particle size distribution and concentration in the size range from 15 nm to 20 μm at distances from a road ranging from 15 to 375 m at two sites in Australia. They conducted measurements under different wind conditions and found that when the wind is blowing directly from the road, the concentration of the fine and ultrafine particles decayed to about half of their maximum at a distance of 100–150 m from the road. Shi et al. (1999) measured ultrafine particle number concentrations and size distributions at a busy roadside and at nearby urban background sites in Birmingham, United Kingdom. They observed a faster decline of particle number concentration than mass concentration. In a recent study, Shi et al. (2001) reported that the fraction of particles <10 nm represents more than about 40% of the total particle number concentrations at 4 and 25 m from the roadside curb.

While there have been recent studies of ultrafine particles from traffic in other countries, except for Zhu et al. (2002), no comparable work has been done in the Los Angeles basin, a home to more than 15 million individuals and 10 million vehicles contributing to daily traffic. Previous studies have shown that meteorological conditions may affect substantially the characteristics of PM emitted from vehicles. Kittelson et al. (2001) found in their on-road PM measurements that the concentration of particles in the nuclei mode increases by nearly a factor of 10 as the (air) temperature is reduced from 25°C to 15°C. This observation suggests that there could be significant differences in the tendency to form semi-volatile nanoparticles between, for example, northern Europe and Southern California.

Zhu et al. (2002) conducted a systematic ultrafine particle study near one of the busiest freeways in the Los Angeles basin, Interstate 405. Traffic on that freeway was dominated by gasoline-powered cars and light trucks, with <5% of vehicles being heavy-duty diesel trucks. In the US, spark ignition vehicles usually account for most of the vehicles operating on highways. However, since diesel vehicles emit more PM on a fleet averaged, gram-per-vehicle mile mass basis (Kittelson et al., 2001), and that diesel engine exhaust has been proposed as carcinogen in animals and probably carcinogenic for humans (IARC, 1989), it is necessary and timely to conduct a comprehensive study of ultrafine particles in the vicinity of a diesel vehicle dominated freeway. Thus, the aim of the present paper is

to systematically evaluate ultrafine particles in the vicinity of the 710 freeway in the Los Angeles basin, a freeway where more than 25% of vehicles are heavy-duty diesel trucks. Particle number concentration and size distribution in the size range from 6 to 220 nm are measured along with CO and black carbon (BC) as a function of distances upwind and downwind the 710 freeway. The results from the current study are compared to these by Zhu et al. (2002) which were obtained near the 405 freeway.

2. Experimental

2.1. Description of sampling site

This study was conducted in the City of Downey along Southern Avenue between 30 August and 27 October 2001. The location was chosen for its proximity to the freeway and the lack of other nearby ultrafine particle emission sources. Southern Avenue is located perpendicular to Interstate 710 Freeway and Garfield Avenue near the Los Amigos Country Club. Freeway 710 runs generally north and south near the sampling site and parallels the Los Angeles River.

This location is ideal for this study for several reasons. First, there are no other major roadways near the sampling sites along Southern Avenue. Second, businesses along Southern Avenue generally have large open land areas with little activities during the day. Thus, there is minimal local traffic influence at the sampling locations. Third, the freeway is at the same elevation as Southern Avenue. The only separation between the freeway and Southern Avenue is a metal chain link fence along the freeway. This allowed measurements as close as 3 m from the edge of the freeway. Fourth, a nearby residential area approximately 200 m upwind from the freeway was easily accessible for sampling.

During the sampling period, a fairly consistent eastward wind developed each day starting at approximately 11:00 AM. This wind carried the freeway vehicular emissions directly to the sampling location. The 710 freeway has eight lanes, four north bound and four south bound. It is approximately 26 m wide including a 1-m-wide median strip. Measurement site locations for this study were designated by their distance from the center of the median strip. Thus, the distance from each sampling location to the nearest traffic lane is 13 m less than the indicated distance.

Freeway 710 is a major truck route in Southern California with a large percent of the traffic consisting of heavy-duty diesel trucks. During the sampling period, traffic density ranged from 180 to 230 vehicles/min passing the sampling site, total for both directions, with approximately 25% of the vehicles being heavy diesel trucks.

2.2. Sampling and instrumentation

Wind speed and direction were measured at a fixed site 6 m above the ground level 20 m downwind of 710 freeway, which also served as a particle number concentration control site. Wind data were averaged over 1 min intervals and logged into a computerized weather station (Wizard III, Weather Systems Company, San Jose, CA). Throughout each measurement period, the traffic strength on the freeway, defined as number of vehicles passing per minute, was continuously monitored by a video recorder (camcorder), which captures all eight lanes of the freeway. After each sampling session, the videotapes were replayed and traffic density counted manually. Three 1-min samples were randomly selected from each 10-min interval. Cars, light trucks, and heavy-duty trucks were counted separately to estimate the traffic density by type of vehicle.

Particle number concentration and size distribution in the size range from 6 to 220 nm were measured by a condensation particle counter (CPC 3022A; TSI Inc., St. Paul, MN) and a scanning mobility particle sizer (SMPS 3936, TSI Inc., St. Paul, MN). The sampling flow rate of the SMPS was adjusted to 1.5 lpm in order to measure particles as low as 6 nm as well as to minimize the diffusion losses of ultrafine particles during sampling. Flexible, conductive tubing (Part 3001940, TSI Inc., St. Paul, MN) was used for sampling to avoid particle losses due to electrostatic forces. The sizing accuracy of the SMPS was verified in the laboratory by means of monodisperse polystyrene latex spheres (PSL, Polysciences Inc., Warrington, PA). Data reduction and analysis of the SMPS output was done by the Aerosol Instrument Manager software (version 4.0, TSI Inc., St. Paul, MN). Measurements were taken at 17, 20, 30, 90, 150, and 300 m downwind and 200 m upwind from the center of the freeway 710. At each location, three size distribution samples were taken in sequence with the SMPS. Scanning time for each was 180 s.

In addition to size distribution and the total number concentration, the concentrations of BC and carbon monoxide (CO), were monitored simultaneously at each sampling location. Before each measurement session, all instruments were time synchronized. Data were averaged after collection over the time periods corresponding to the scanning intervals of the SMPS. A Dual Beam Aethalometer (Model AE-20, Andersen Model RTAA-900, Andersen Instruments Inc., Smyrna, GA) was used to measure the BC concentrations every 5 min. Concentrations of CO were measured by a near-continuous CO monitor (Dasibi Model 3008, Environmental Corp., Glendale, CA) every minute. The CO monitor was calibrated by means of standard CO gas (RAE systems Inc., Sunnyvale, CA) in the laboratory and automatically zeroed each time the power was turned on.

Electric power for the control site CPC and Weather Station was obtained by an extension cord to a nearby office. Electric power for other sampling instruments at the sampling locations was supplied by a 1.2 kW gasoline-powered portable power generator (Model EU 1000i, Honda Motor Co., LTD., Tokyo, Japan). The generator was placed approximately 50 m downwind of each sampling location. Both total particle number and CO concentrations were measured at the control site with the generator turned on and with it turned off. No detectable difference was observed.

Table 1 gives the sampling dates and times and summarizes the instruments that were used on each date. The weather station and control CPC were placed at the 20 m downwind control site and sampled throughout the sampling period each day. All other applicable instruments were moved together and sampled simultaneously at each sampling location. It takes about 10 min to complete sampling at each location and 120 min to complete a set, all six locations. Three to four sets were performed on each sampling date.

3. Results and discussion

The results presented below include measurements of total particle number concentrations by a control CPC, wind velocity by a Weather Wizard III, both positioned at a fixed location 20 m downwind of the freeway; and CO, BC concentration, and ultrafine particles size distributions upwind and at six downwind distances from freeway 710.

3.1. Wind effects

Changes in wind conditions have been reported to modify dramatically the pattern of total particle number concentration versus distances from a major road (Hitchins et al., 2000). Consistency in wind speed and direction allows data from different days to be averaged together (Zhu et al., 2002). Wind speed and direction were measured, averaged and logged over every 1-min interval throughout each sampling period. One hundred wind data points were randomly selected out of more than 5000 observations from all the sampling dates and plotted in Fig. 1. The orientation of freeway 710 and the sampling road, Southern Avenue, are also shown in the Fig. 1. The Weather Wizard III instrument recorded wind direction at a 22.5° interval (e.g. 11.25° on either side of N, NNE, etc.) and wind speed at 0.4 or 0.5 m/s intervals. In the figure, duplicate observations were spread out slightly in both directions to better illustrate how strong the wind was and how often the wind came from certain directions. Based on all 5000 observations, the percent of sampling time that the wind came from each 22.5° segment is also shown in Fig. 1. As shown in Fig. 1, about 80% of the time, the wind was coming directly from the freeway towards the sampling road with a speed <3 m/s. The consistency of observed wind direction and speed is a result of a generally low synoptic wind velocities and a consistent sea breeze in the sampling area.

In this study, we found that not only wind direction, but also wind speed, played an important role in determining the characteristics of ultrafine particles near the 710 freeway, similar to the observations made by Zhu et al. (2002) near the 405 freeway. However the pattern of total particle number concentrations as a function of wind speed is somewhat different for the two studies. Fig. 2 shows total particle number concentrations measured by the control CPC, located 20 m downwind of the 710 freeway versus wind speed. Averaged data for the 405 freeway from Zhu et al. (2002) are also plotted for comparison. The CPC was programmed to archive averaged total particle number concentrations at 1-min interval in synchronization with the averaging time of the meteorological data. Only wind data within $\pm 22.5°$ of normal to the freeway was used in this figure which accounts for more than 60% of the total observations. The difference between the absolute value of total particle number concentration is due in part to the difference in the sampling distance. The control CPC was located 20 m downwind from the 710 freeway but 30 m from the 405 freeway. Assuming the fitted exponential decay characteristics of ultrafine particles holds right to the edge of the freeway, it is thus

Table 1
Sampling dates, time and instruments used

Date	Time	Weather Wizard III	Control CPC	SMPS	CO monitor	Aethalometer
08/30/01	10:00–15:30	×	×	×	×	×
09/05/01	10:30–16:00	×	×	×		
09/21/01	10:00–15:00	×	×	×	×	×
09/25/01	10:30–16:00	×	×	×	×	×
10/05/01	10:30–16:00	×	×	×	×	×
10/24/01	10:00–15:30	×	×		×	×
10/30/01	10:00–15:30	×	×	×		

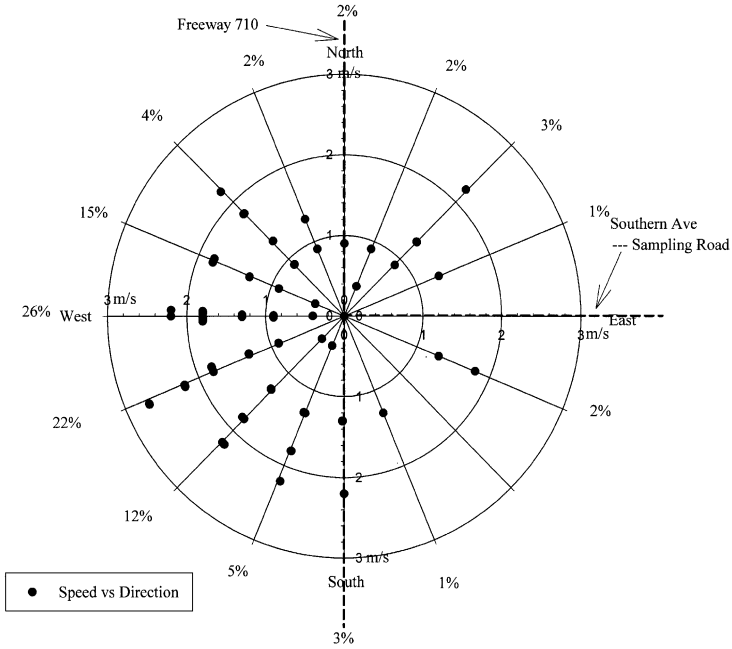


Fig. 1. Wind direction and speed at sampling site.

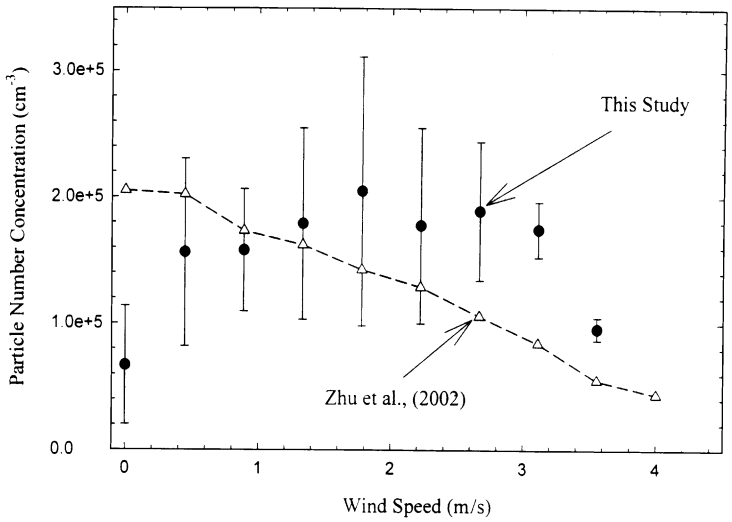


Fig. 2. Total particle number concentration measured by CPC located at 20 m downwind from freeway 710 versus wind speed. Bars indicate one standard deviation.

not surprising, as discussed below, that the CPC will read a greater total particle number concentration at 20 m in the present study than at 30 m in that by Zhu et al. (2002), given similar traffic load on both freeways. However, the relative particle number concentration as function of wind speed are somewhat different in these two studies. The relative particle number concentration decreased as the wind speed increased near the 405 freeway. In contrast, particle number concentration in the 710 freeway first increases, reaches a maximum around 1.5 m/s, and then decreases. There is no obvious explanation for the observed difference. In both studies, data showed large error bars, and the data of low wind speed (<1 m/s) were very limited. In addition, the 405

freeway is elevated approximately 4.5 m above the surrounding terrain, while, the 710 freeway is at ground level, the same as the sampling location. Lower speed wind would be expected to cause less atmospheric dilution, and thus lead to greater particle number concentrations, as Zhu et al. (2002) reported. However, at extremely low wind speeds, it would take a considerably longer time for the wind to carry particles to the sampling port of the CPC, which gives ultrafine particles more time to coagulate with either themselves or with larger particles, a phenomenon that would decrease the total particle number concentration. This may partially explain the observed "n" shape curve in the current study.

3.2. Traffic effects

The portion of freeway 710 passing through the City of Downey is a major truck shipping route. The average traffic volume per hour during the measurement period was: 8730 cars, 870 light trucks, 2580 heavy trucks, and 12180 total vehicles. It is apparent from these numbers that diesel emission vehicles on the 710 freeway represent about 30% of vehicles while on the 405 freeway they represent $<5\%$ (Zhu et al., 2002). Fig. 3 compares the traffic volume on both the 405 and the 710 freeways. Error bars represent one standard deviation. It is seen that the 710 freeway has about 7 times as many diesel vehicles and 70% of gasoline vehicles as the 405 freeway. The total vehicle numbers on both freeways are quite similar 12,180 versus 13,900/h for the 405 freeway.

Zhu et al. (2002) reported that a traffic slowdown on freeway 405 was associated with a drop in total particle number concentration indicating that fewer ultrafine particles are emitted during such events. In this study, the traffic speed on the 710 freeway stayed constant through out the sampling period. No traffic slow down was observed. The difference in the variability of traffic volume on both freeways is indicated by the error bars in Fig. 3.

Zhu et al. (2002) reported that both wind speed and traffic density affected the characteristics of ultrafine particles near the 405 freeway, and the control CPC responded to these effects reasonably well. Thus, subsequent data for ultrafine particle analysis at increasing distances from the freeway were all normalized to the control CPC's reading. An average CPC reading, $\overline{C_N}$, was obtained based on all the measurements. In Figs. 4–6, number concentration and size distribution data were scaled to $\overline{C_N}$ by dividing each measurement by the ratio of CPC reading for the period of measurement to $\overline{C_N}$.

3.3. Change in ultrafine particle size distribution with increasing distance

Fig. 4 depicts ultrafine particle size distributions at 17, 20, 30, 90, 150 and 300 m downwind and 300 m upwind of freeway 710. The size distributions are smoothed and shown together with common scales for both axes. The horizontal axis represents particle size on a logarithmic scale, while the vertical axis represents normalized particle number concentration in the size range of 6–220 nm as measured by the SMPS. Data were averaged for all applicable sampling dates for each distance from the freeway. As shown in Fig. 4, ultrafine particle size

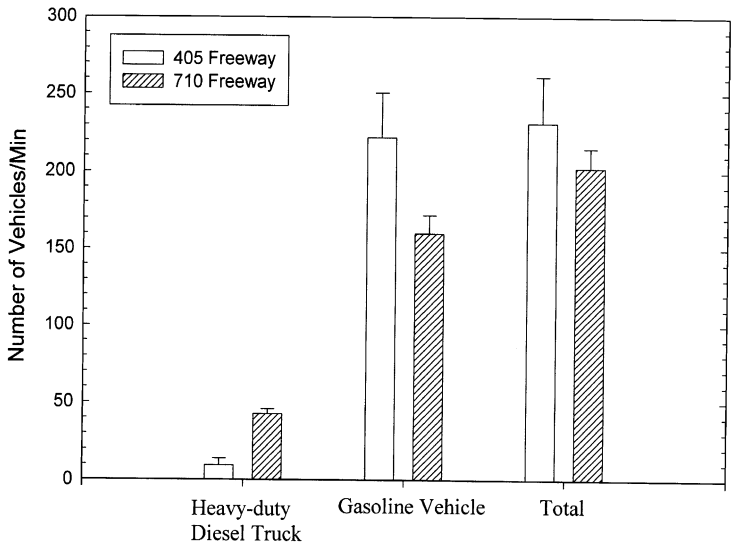


Fig. 3. Traffic volume comparison for the 405 and 710 freeway. Bars indicate one standard deviation.

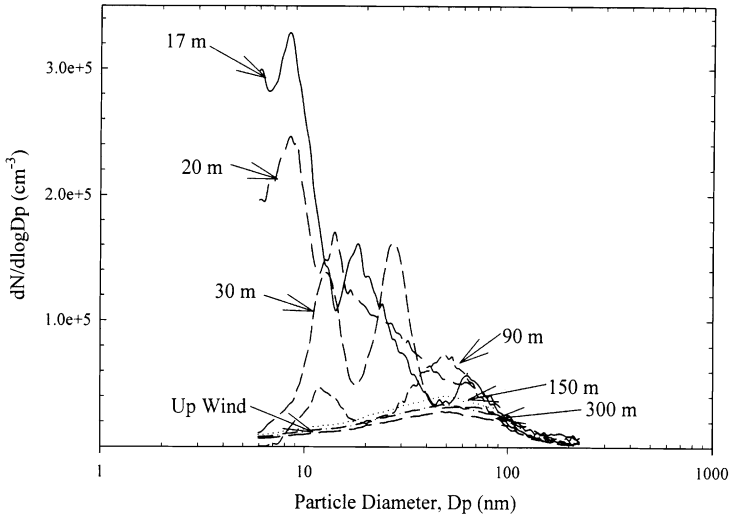


Fig. 4. Ultrafine particle size distribution at different sampling locations near the 710 freeway.

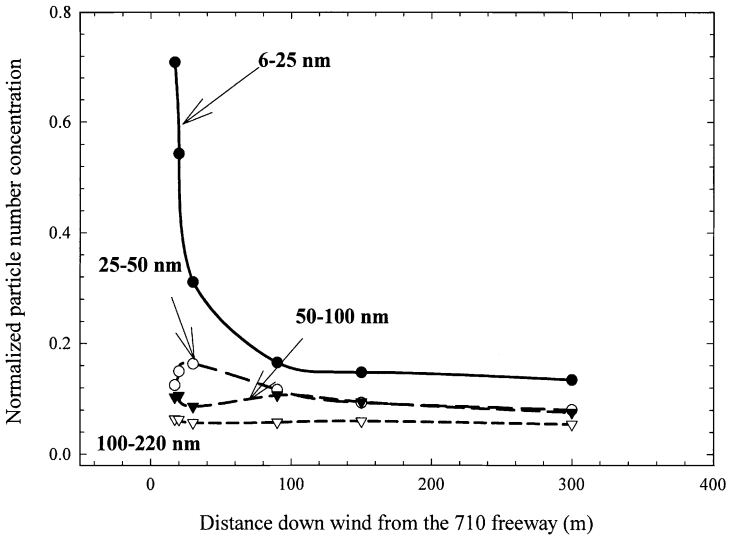


Fig. 5. Normalized particle number concentration for different size ranges as a function of distance from the 710 freeway.

distribution changed markedly and its number concentration dropped dramatically with increasing distance. At the nearest sampling location, 17 m downwind from the center of the freeway, the dominant mode was around 10 nm with a modal concentration of more than $3.2 \times 10^5/cm^3$. This mode remained at 10 nm for the second sampling location, 20 m downwind from the freeway, but its concentration dropped to $2.4 \times 10^5/cm^3$. It shifted to larger size range and its concentration kept decreasing for farther sampling locations. This mode was not observed at distance >150 m downwind from the freeway. The dramatic decrease of particle number concentration in the size range around 10 nm was likely due to atmospheric dilution and several atmospheric aerosol particle loss mechanisms that favor small particles, diffusion to surfaces, evaporation, and coagulation. The smaller the particle, the greater its diffusion coefficient and its Brownian motion. Particles of 10 nm diffuse about 80 times faster than particles of 100 nm (Hinds, 1999). As particle size gets smaller, the Kelvin effect becomes more important, making it easier for molecules to leave the particle's surface by evaporation. In addition, when two small particles collide due to their Brownian motion (coagulate), they form a bigger particle. Thus, coagulation reduces number concentrations and shifts the size distribution to larger sizes.

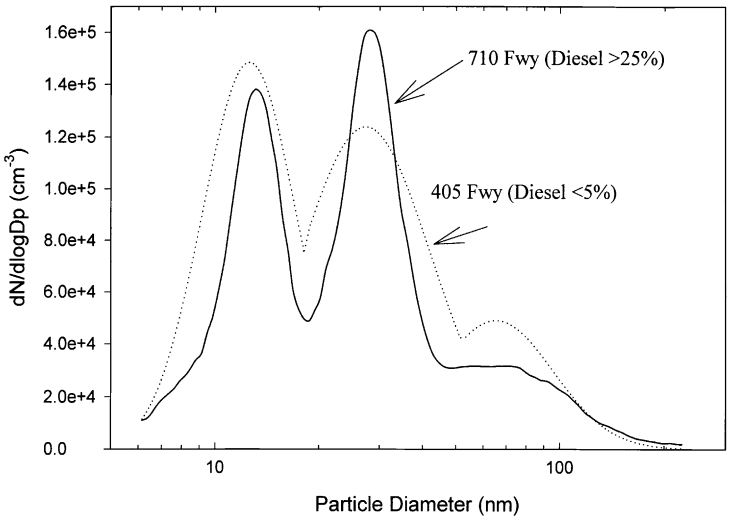
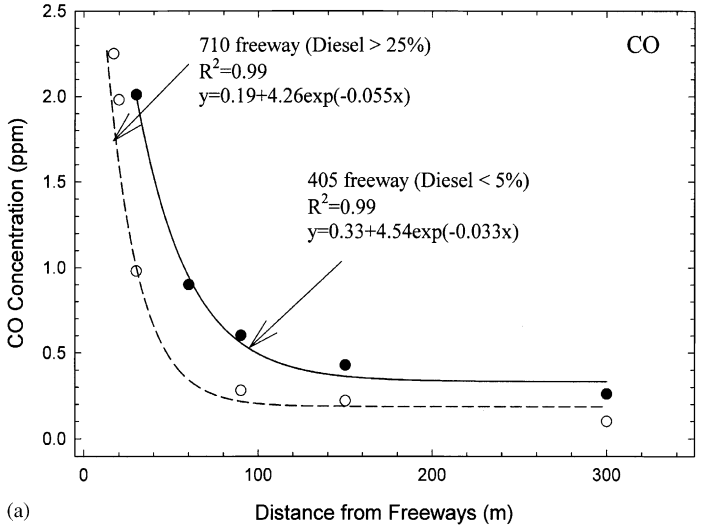


Fig. 6. Comparison of ultrafine particle number concentration at 30 m downwind from 405 and 710 freeway.

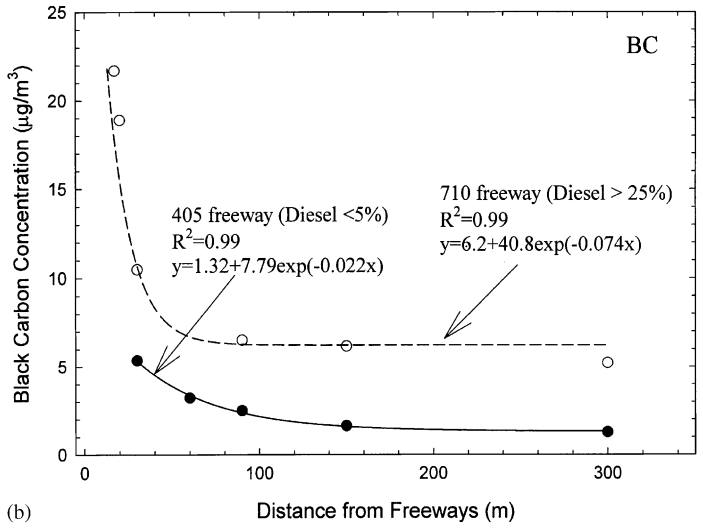
In Fig. 4, the second mode at 17 m downwind from the freeway was around 20 nm with a concentration of $1.5 \times 10^5/\text{cm}^3$. This mode remained at similar size range and concentration for the next sampling location, 20 m, but shifted to 30 nm at 30 m downwind from the freeway. It is of particular note that, while the concentration for the primary mode, 10 nm mode, decreased about 60% of its maximum value from 17 to 30 m with a slight shift in its mode, the 20 nm mode concentration did not change significantly but the modal size shifted noticeably. This second mode continued to shift to larger sizes with increasing distance from the freeway. In general number concentrations for smaller particles, $d_p < 50$ nm, dropped significantly with increasing distances from the freeway, but for larger ones, $d_p > 100$ nm, number concentrations decreased only slightly. These results are in excellent agreement with what Zhu et al. (2002) reported for freeways impacted mostly by gasoline vehicles, which suggests that coagulation is more important than atmospheric dilution for the smallest ultrafine particles and vice versa for large particles. Ultrafine particle concentrations measured at 150 and 300 m downwind of the 710 freeway were statistically within the variation of the 300 m upwind background concentration. The maximum number concentration that was observed next to the freeway was about 30 times greater than that for the background location. This suggests that people who live or work within 100 m downwind of major traffic sources, or spend a substantial amount of time commuting on such highways, will have a much higher ultrafine particle exposure than those who do not. This result can be used in epidemiological studies to estimate exposure to ultrafine particles.

Based on Fig. 4, it is clear that vehicle-emitted ultrafine particles of different size ranges behave quite differently in the atmosphere. Zhu et al. (2002) showed the decay of ultrafine particle number concentrations in four size ranges 6–25, 25–50, 50–100 and 100–220 nm. They found coagulation played a significant role in modifying the particle size distribution of vehicle-emitted ultrafine particle downwind of a freeway. Fig. 5 was prepared in the same ways as Zhu et al. (2002). The measured particle number concentrations in each SMPS size bin were combined in the corresponding size range, and the result was normalize to averaged wind speed. The general trends of sub-grouped ultrafine particle decay curves are quite comparable to those given by Zhu et al. (2002), Figs. 7a and b. Total particle number concentration in the size range of 6 to 25 nm accounted for about 70% of total ultrafine particle number concentration and dropped sharply, by about 80%, at 100 m, and leveled off after 150 m. Overall, it decayed exponentially through out the whole measured distance. Number concentrations in the next two size ranges 25–50 and 50–100 nm, all experienced a shoulder between 17 and 150 m. These results are in excellent agreement with what Zhu et al. (2002) observed and can be explained by particles, in smaller size ranges, coagulating with these particles to increase their size.

Fig. 6 compares the ultrafine particle size distributions at 30 m downwind from the 710 and the 405 freeways. Three-mode lognormal fitting was used for 405 freeway. Raw data were smoothed by averaging for 710 freeway. Heavy-duty diesel trucks on the 710 freeway represent more than 25% of traffic while on the 405 freeway they represent $< 5\%$ (Zhu et al., 2002). Average PM emission rate for heavy-duty diesel trucks is about 0.4 g/mi (California ARB, 2000) while for passenger cars is



(a)



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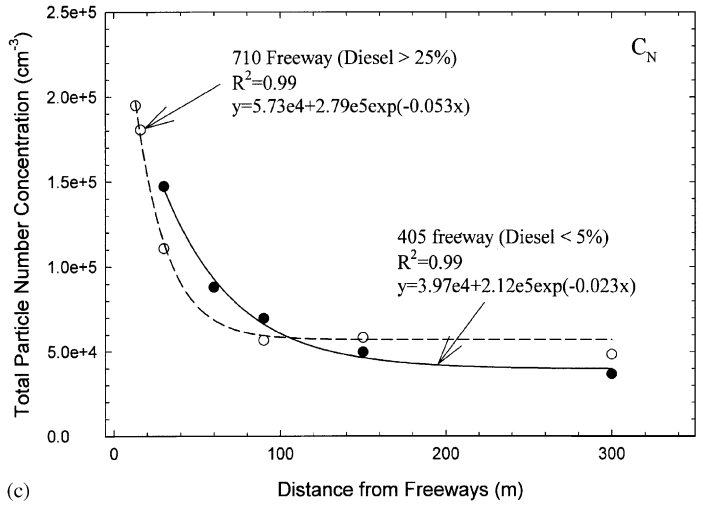

(c)

Fig. 7. Decay curves of: (a) CO, (b) BC and (c) particle number concentration near the 405 and 710 freeway.

about 0.08 g/mi (EPA, 2000). Thus, on the 710 freeway, about 60% of PM emission is due to heavy-duty diesel trucks $((0.25 \times 0.4)/(0.25 \times 0.4 + 0.75 \times 0.08) = 62.5\%)$. In Fig. 6, both size distributions have three distinct modes. The concentration for the first mode, between 10 to 20 nm, is slightly higher near the 405 freeway. This mode is likely to arise from homogeneous nucleation of semi-volatile materials and is similar to that previously reported for direct laboratory measurement of gasoline vehicle emissions (Ristovski et al., 1998). The concentration for the second mode, around 30 nm, is about 30% higher near the 710 freeway than that near the 405 freeway. This mode probably comprises mainly of BC and is likely due to the much higher diesel emissions on the 710 freeway. The last mode, around 70 nm, represents an insignificant contribution to number concentrations for these two freeways and in both cases are comparable to the background concentrations.

3.4. Decay of carbon monoxide, black carbon and particle number concentration

To make this freeway study more comprehensive, the concentrations of CO, BC, and particle number were also measured at increasing distance from the freeway on selected dates, as shown in Table 1. CO and BC were intentionally selected because their ambient concentrations are closely related to vehicular emissions. Averaged concentration and range of values at different distances from the freeway of each measured property are summarized in Table 2. CO and BC concentrations decreased noticeably when moving away from the traffic sources, similar to the findings of the study by Zhu et al. (2002).

Figs. 7a–c were prepared by comparing the decay characteristic of CO, BC and particle number concentrations near the 405, gasoline vehicle dominated, and the 710, diesel vehicle dominated, freeways. Exponential decay was found to be a good estimator for predicting total particle number concentrations at different locations (Zhu et al., 2002). Each data point in the figure represents an averaged value for all measurements with similar wind directions. The solid line was the best fitting exponential decay curve, determined using SigmaPlot 2000 nonlinear curve fitting procedure. The best fitting exponential decay equations and R^2 values are also given in the figure. It can be seen, in general, all three pollutants decay at a similar rate near both freeways. This implies that atmospheric dilution plays a comparable role in both studies. As discussed previously, the average wind speed for these two studies are all close to 1.5 m/s. The discrepancies of the curves were mainly due to the different traffic fleet compositions on these two freeways. The 710 freeway has more than 25% heavy diesel trucks while the 405 freeway has $<5\%$. It is well known that diesel engines emit less CO and more BC comparing to spark ignition engines (Kittelson et al., 2001). Fig. 7a shows that the concentration of CO near the 710 freeway is generally half of that near the 405 freeway. By comparison, Fig. 7b shows the BC concentration near a diesel vehicle dominated freeway is more than three times greater than that near a gasoline vehicle dominated freeway. As shown in Fig. 7c, the total particle number concentration close to the 405 freeway is somewhat higher than that near the 710 freeway, but drops faster with downwind distance. Since the rate of coagulation increases with decreasing particle size down to 20 nm (Hinds, 1999), the observed result suggests more of the smallest ultrafine particles, mostly in nanosize range, were emitted from the 405 freeway. This may be explained by a total of 20% more vehicles on the 405 freeway. It was previously reported that number emission rates from the spark-ignition vehicles were much lower than from the diesel vehicles under most operating conditions, but were similar under high-speed highway cruise conditions (Rickeard et al., 1996; Kittelson, 1998). It should also be noted that the exponential decay characteristic appears to extend to about 3 m downwind from the edge of the freeway for all three pollutants. Based on our results we conclude that atmospheric dilution is so rapid that average concentration decays continuously after leaving the tailpipe.

Table 2
Measured averaged concentrations at increasing distances from the freeway[a]

Measurement	Upwind (m)	Downwind distance (m)					
	200	17	20	30	90	150	300
CO (ppm)	0.1 (0.0–0.2)	2.3 (1.9–2.6)	2.0 (1.5–2.4)	1.7 (1.1–1.9)	0.5 (0.2–0.7)	0.4 (0.1–0.5)	0.2 (0.1–0.3)
Black carbon ($\mu g/m^3$)	4.6 (3.1–5.9)	21.7 (20.3–24.8)	19.4 (16.5–21.6)	17.1 (12.6–19.3)	7.8 (4.5–9.3)	6.5 (3.9–9.2)	5.5 (3.5–7.7)
Number concentration ($\times 10^{-5}/cm^3$)	0.48 (0.36–0.57)	2.0 (1.8–2.5)	1.8 (1.5–2.5)	1.6 (1.2–1.9)	0.72 (0.42–1.1)	0.61 (0.35–0.98)	0.49 (0.30–0.59)

[a] Range given in parenthesis.

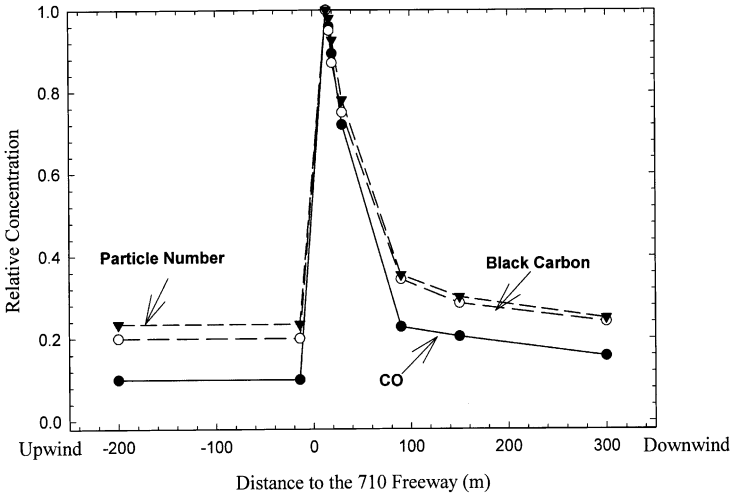

Fig. 8. Relative particle number, BC, CO concentrations versus distance from the 710 freeway.

Fig. 8 shows the decay curves for relative concentrations of CO, BC and total particle number. The curves are normalized and extended to reach 1.0 at the downwind edge of the 710 freeway. Background concentrations are also shown in the figure. It is seen that CO, BC and particle number concentration decreased about 60–80% in the first 100 m and then leveled off somewhat after 150 m, similar to what Zhu et al. (2002) reported. Background CO has a much lower relative concentration while background BC and particle number concentrations are comparable. Thus, CO was diluted more quickly and significantly than BC and particle number concentration. In general, CO, BC and particle number concentrations tracked each other very well. These results confirm the common assumption that vehicular exhaust is the major source for CO, BC and ultrafine particles near a busy freeway. They also support the conclusion made by Zhu et al. (2002) that for the conditions of these measurements the decreasing characteristics of any of these three pollutants could be used interchangeably to estimate the relative concentration of the other two pollutants near freeways.

4. Conclusions and summary

Wind speed and direction are important in determining the characteristic of ultrafine particles near freeways. The average concentrations of CO, BC and particle number concentration at 17 m was 1.9–2.6 ppm, 20.3–24.8 $\mu g/m^3$, 1.8×10^5–$3.5 \times 10^5/cm^3$, respectively. Relative concentration of CO, BC and particle number tracked each other well as one moves away from the freeway. Exponential decay was found to be a good estimator for the decrease of these three pollutants' concentration with distance along the wind direction starting from the edge of the freeway. Measurements show that both atmospheric dilution and coagulation play important roles in the rapid decrease of particle number concentration and the change in particle size distribution with distance away from a freeway.

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