Comparison of Daytime and Nighttime Concentration Profiles and Size Distributions of Ultrafine Particles near a Major Highway

YIFANG ZHU,* THOMAS KUHN,
PAUL MAYO, AND WILLIAM C. HINDS

*Department of Environmental Health Sciences,
University of California Los Angeles, 650 Charles E. Young
Drive South, Los Angeles, California 90095*

Previously we have conducted systematic measurements of the concentration and size distribution of ultrafine particles in the vicinity of major highways during daytime in Los Angeles. The present study compares these with similar measurements made at night. Particle number concentration was measured by a condensation particle counter (CPC) and size distributions in the size range from 7 to 300 nm were measured by a scanning mobility particle sizer (SMPS). Measurements were taken at 30, 60, 90, 150, and 300 m upwind and downwind from the center of the I-405 freeway. Average traffic flow at night was about 25% of that observed during the day. Particle number concentration measured at 30 m downwind from the freeway was 80% of previous daytime measurements. This discrepancy between changes in traffic counts and particle number concentrations is apparently due to the decreased temperature, increased relative humidity, and lower wind speed at night. Particle size distributions do not change as dramatically as they did during the daytime. Particle number concentration decays exponentially downwind from the freeway similarly to what was observed during the day, but at a slower rate. No particle number concentration gradient has been observed for the upwind side of the freeway. No $PM_{2.5}$ and very weak PM_{10} concentration gradients were observed downwind of the freeway at night. Ultrafine particle number concentration measured at 300 m downwind from the freeway was still distinguishably higher than upwind background concentration at night. These data may be used to help estimate exposure to ultrafine particles in the vicinity of major highways for epidemiology studies.

Introduction

Although there is a clear correlation between particulate pollution episodes and increased morbidity and mortality, the causal mechanism of these health effects is not yet clear (*1, 2*). Health effects appear to correlate better, or equally well, with ultrafine particle (Dp < 100 nm) number concentration than with, for example, $PM_{2.5}$ mass concentration (*3, 4*). It has been suggested that ultrafine particles are more toxic and biologically active than larger particles (*5−7*).

Vehicular emission is a major source of ultrafine particles in urban areas with dense roadway systems (*8, 9*). New engine

technology and fuel reformulation have led to decreased particle mass concentrations emitted from vehicles, but ultrafine particle number concentrations have remained unchanged or even increased (*10*). Formation of ultrafine particles depends on the dilution condition of the engine exhaust after leaving the tailpipe (*11, 12*).

Previously we conducted measurements of particle number concentration and size distribution near two major freeways (Interstates 405 and 710) in Los Angeles, California (*13, 14*). Summertime measurements at both freeways showed rapid decrease in particle number concentration and significant changes in particle size distribution with increasing distance downwind from the freeway for particles in the size range of 6−220 nm. During the sampling periods the average temperature and humidity in the Los Angeles basin were moderately high: 34 °C and 66%, respectively. In winter 2001, we repeated our measurements at freeways I-405 and I-710 (*15*). Particle number concentration in the size range of 6−12 nm was significantly higher in winter than in summer. The concentration decreased at a slower rate in winter than in summer indicating a weaker atmospheric dispersion effect in winter.

The general meteorological conditions of the Los Angeles basin change with the season and with the time of day. The conditions at night influence the dynamic processes that change the ultrafine size distribution and concentration in the vicinity of a freeway. Reduced temperatures are also likely to increase emission from vehicles (*11, 16*), while lower traffic flow may reduce emissions. In this study we measured the number concentration and size distribution of ultrafine particles in the vicinity of Interstate 405 at nighttime. We compared the results of the present study with previous studies of this freeway during daytime in summer (*13*) and winter (*15*) to investigate the degree to which differences in atmospheric conditions, such as temperature, humidity, and atmospheric mixing, affect the characteristics of ultrafine particles.

Materials and Methods

This study is a follow-up to daytime measurements (published previously in refs *13, 15*) near the Interstate 405 freeway. The same sampling site, similar instruments, and same sampling protocol were used to ensure comparability of nighttime and daytime ultrafine particle size distributions and number concentrations. Seven night measurements (11 p.m. to 4 a.m.) were conducted from January 31 to February 8, 2005.

Description of Sampling Site. Freeway I-405 runs generally north−south (actual orientation 330°) along the western boundary of the Los Angeles National Cemetery and the eastern boundary of the Veterans Affairs (VA) facility. Measurements were taken along Constitution Avenue, which runs perpendicular to the freeway. Constitution Avenue passes through a tunnel under the freeway thereby providing easy access to both east (LA Cemetery) and west (VA) sides of the freeway. A detailed description of the sampling site was given previously (*13*).

The site lies 6.4 km east of Santa Monica Bay and the Pacific Ocean. In this area, a consistent sea breeze (eastward from the ocean) usually starts in the mid-morning, reaches its maximum early to mid-afternoon, and dies out in the early evening. Once the land cools at night, the wind reverses its direction and usually forms a weak offshore sea breeze (westward toward the ocean) at night. Thus, in our previous studies during the daytime, the Los Angeles National Cemetery on the eastern side of the I-405 freeway was downwind. In the present study at night, the Los Angeles

TABLE 1. Sampling Dates and Instruments Used

date	time	Weather Wizard III	CPC	SMPS	Q-Trak (CO)	Dusttrak (PM$_{2.5}$)	Dusttrak (PM$_{10}$)
1/31/05−2/1/05	23:30−5:00	×	×	×	×	×	
2/1/05−2/2/05	22:30−4:00	×	×	×	×	×	
2/2/05−2/3/05	22:30−4:00	×	×	×	×	×	
2/3/05−2/4/05	22:30−4:00	×	×	×	×	×	
2/4/05−2/5/05	22:30−4:00	×	×	×	×	×	
2/7/05−2/8/05	22:30−4:00	×	×	×	×		×
2/8/05−2/9/05	22:30−4:00	×	×	×	×		×

National Cemetery was upwind and the VA facility was downwind of the freeway. There are no significant ultrafine particle emission sources on either side of the freeway. During the sampling period, consistent traffic patterns were observed. Traffic density usually started from about 70 vehicles/min passing the sampling site, total in both directions at 11 p.m., gradually dropping to 20 vehicles/min at 2 a.m., and increasing to 50 vehicles/min at 4 a.m. each night. Within each measurement cycle (∼50 min), traffic density stayed fairly stable. Constant vehicle speeds were observed throughout all sampling nights with an average of 28.8 m/s (64 mi/h).

Sampling and Instrumentation. Meteorological data were measured at a fixed site near the freeway. Carbon monoxide (CO), PM$_{10}$, PM$_{2.5}$, total particle number concentration, and ultrafine particle size distribution were measured at increasing distances on both eastern and western sides of the I-405 freeway. Table 1 gives the sampling dates and times, and summarizes the instruments that were used on each date. All instruments, except the weather station, were placed on an electric vehicle and were driven together to sample simultaneously at each location. Two sealed 12 V gel batteries (97.6 Ah) and a 1.0 kW sine wave inverter were used to provide power for the other instruments. It took about 6 min to complete sampling in one location and about 50 min to finish a set of five sampling points on one side of the freeway. Three to four sets were performed at the VA (downwind site) and one or two sets were performed at the cemetery (upwind site) each night.

Wind speed and directions were measured 6 m above the ground level by a computerized weather station (Wizard III, Weather Systems Company, San Jose, CA). Due to the nature of the weak onshore sea breeze at night, the wind speed is constantly below 1 m/s or sometimes below the detection limit of Wizard III of 0.4 m/s. To correct for low wind speed, meteorological data collected at the University of California Los Angeles (UCLA) atmospheric science building meteorological station, which is about a mile east of the sampling site, were also retrieved. The UCLA meteorological station measures wind speed down to 0.1 m/s and wind direction to 1°. Good agreement was found between the two methods for wind speeds greater than 0.4 m/s. Wind data from the UCLA meteorological station were used for all subsequent data analysis.

Real-time traffic flow, defined as number of vehicles passing per minute, and speed data were extracted from the Freeway Performance Measurement System (PeMS) maintained by the UC Berkeley Institute of Transportation. The monitor locations are between Wilshire and Sunset Boulevard very close to the sampling site. Traffic volume on the freeway was also monitored periodically by a video recorder (camcorder) on all sampling nights. After sampling, the videotapes were replayed and traffic density was counted manually. Good agreement was found between PeMS data and video records. PeMS data, which are continuous, were then used for all subsequent data analysis.

Particle number concentration was measured by a condensation particle counter (CPC 3022A; TSI Inc., St. Paul,

MN). Ultrafine particle size distributions in the size range from 7.4 to 290 nm were measured by a scanning mobility particle sizer (SMPS 3936, TSI Inc.). The SMPS system includes a long DMA and a water-based CPC (CPC 3785; TSI Inc.) that samples at a flow rate of 1.0 L/min. Due to high diffusion losses, the SMPS may have a lower detection limit for particles less than 10 nm. To keep diffusion losses to a minimum, short, flexible, and conductive tubing (Part 3001940, TSI Inc.) was used. Measurements were taken 30, 60, 90, 150, and 300 m from the center on both sides of the freeway. At each location, three SMPS samples were taken, with a scanning time of 120 s each. Aerosol Instrument Manager software (version 5.3.1.0, TSI Inc.) was used to export CPC and SMPS output.

In addition to size distribution and total number concentration, the concentrations of CO, PM$_{10}$, or PM$_{2.5}$ mass, were monitored simultaneously at each sampling location. Before each measurement, all instruments were synchronized. Concentrations of CO were measured by a Q-Trak IAQ monitor (model 8550, TSI Inc.). All collected data are logged into 1-min intervals in a memory. Calibrations were performed before and after the sampling campaign using standard CO gas (RAE systems Inc., Sunnyvale, CA) in the laboratory. A DustTrak aerosol monitor (model 8520, TSI Inc.) was used as a continuous particle mass monitor for either PM$_{10}$ or PM$_{2.5}$ with corresponding size selective inlets. The DustTrak was calibrated by the manufacturer against the Federal Reference Method prior to the study. All data were averaged later over the time periods corresponding to the scanning intervals of the SMPS using Statistical Analysis System (SAS version 8.01).

Results and Discussion

Previously, we demonstrated the importance of wind speed and direction in determining particle number concentrations near freeways (*13, 14*). In this study, averaged wind data corresponding to each SMPS scan from all the sampling nights were plotted in Figure 1. The orientation of freeway I-405 and the sampling road, Constitution Avenue, are also shown in the same figure. For most of the sampling time, the wind was coming from the eastern side of the freeway with a speed below 1 m/s. There was one night (2/4/05 to 2/5/05) when wind speeds were greater than 1 m/s. The consistency of the observed wind direction makes the western side of the freeway (VA) a downwind side and the eastern side of the freeway (LA cemetery) an upwind side for all the sampling nights. On a few occasions the wind direction was close to being parallel to the freeway, and corresponding data have not been used for the data analysis.
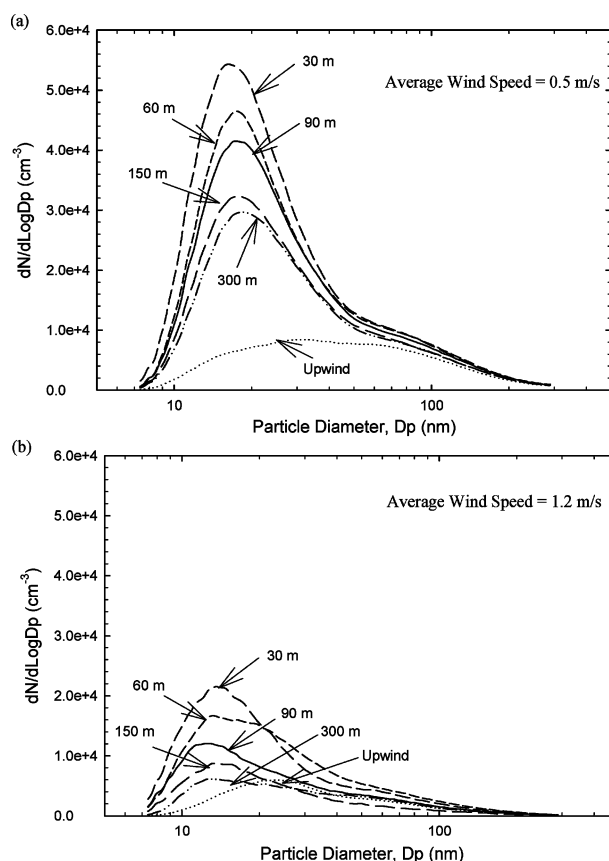
Change in Ultrafine Particle Size Distribution with Increasing Distance. Figure 2a and b show the nighttime ultrafine particle size distribution downwind from the freeway at 5 distances under low and high wind conditions, respectively. All size distributions measured at a given distance were averaged and smoothed by running average of 3 adjacent SMPS channels. For both wind conditions, the distribution showed a relatively stable mode whose location
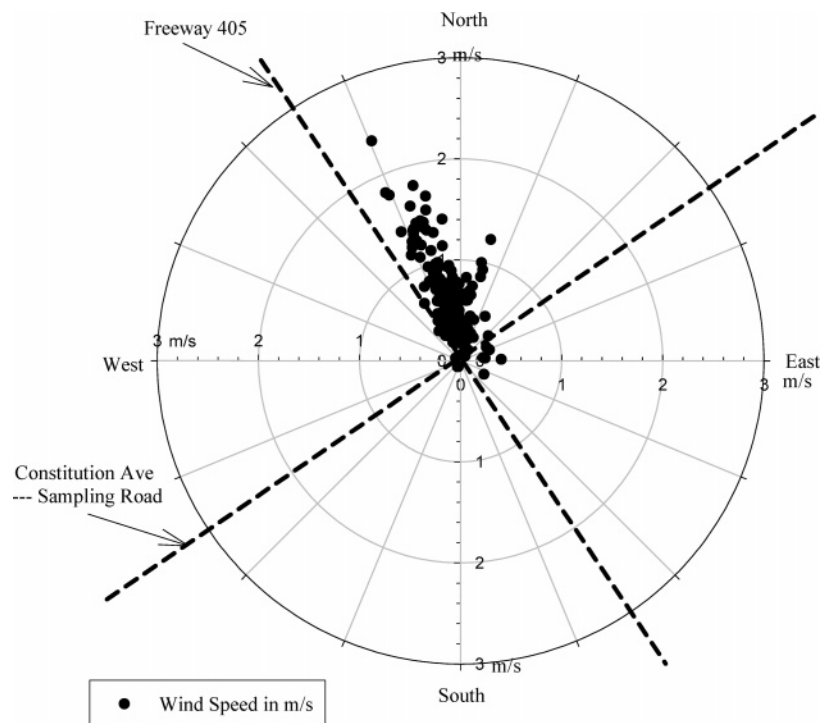
**FIGURE 1. Wind direction and speed at sampling site.**

shifted only slightly toward larger particle diameters at increasing downwind distance. Under low wind conditions, the mode moved from 16 to 19 nm between 30 and 300 m distance. The total particle number decreased with increasing distances downwind of the freeway. At the eastern (upwind) side no significant concentration gradient was observed and a much broader size distribution was found at all sampling points as indicated in Figure 2. At 300 m downwind from the freeway, the size distribution was still higher than the upwind distribution for the low wind condition, whereas under the high wind condition the 300 m downwind distribution was more similar in concentration to the average upwind distribution. There was a ratio of 2.4 between the averaged wind speed at high and low wind conditions. A similar ratio (~2.5) was observed for the modal concentration at 30 m downwind from the freeway between Figure 2a and b. This is in agreement with atmospheric dispersion theory in which downwind concentrations are inversely related to wind speed.

These results are in contrast with measurements performed in summer at the same site (Figure 5 in ref *13*) and are more similar to measurements in winter (Figure 2 in ref *15*). In summer the total number concentration dropped dramatically with increased distance from the freeway and the size distributions changed significantly. In winter the number concentration did not drop as fast as it did in summer and the shape of the size distribution changed only slightly with one mode persisting at around 7 nm for all distances from 30 to 300 m from the freeway. During nighttime the number concentration decreased with increasing distance but dropped more slowly, even slower than in winter. The changes in the size distributions were limited to the slight shift of the modal diameter and were more comparable to winter measurements. In summer the size distributions changed dynamically. The first mode at 30 m distance was observed at 13 nm and grew to 16 nm at 60 m distance and then disappeared farther from the freeway. The second mode at 30 m distance was at 27 nm and grew to 40 nm at 60 m and then remained constant for increasing distances while its modal concentration decreased. These changes could be explained and modeled with ambient dilution (*17, 18*), coagulation (*19*), and either growth by condensation or

FIGURE 2. Smoothed nighttime ultrafine particle size distribution at different sampling locations near freeway I-405 under (a) low wind and (b) high wind conditions.

shrinkage by evaporation (*20*). Such changes were not observed in winter or nighttime. It should be noted that wind direction at night was not perpendicular to the freeway as we observed during daytime. Thus, the downwind distances
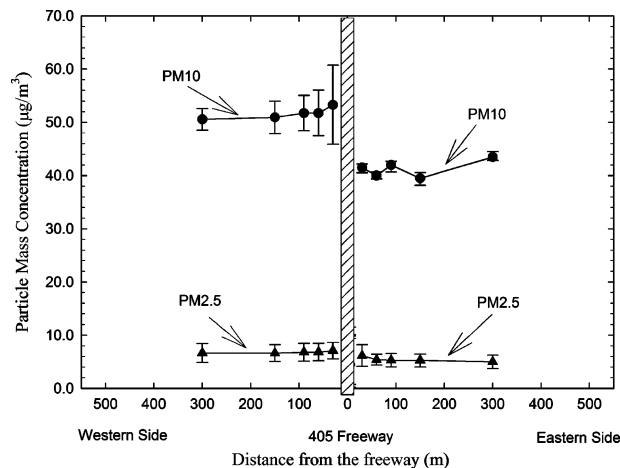
(a)

(b)
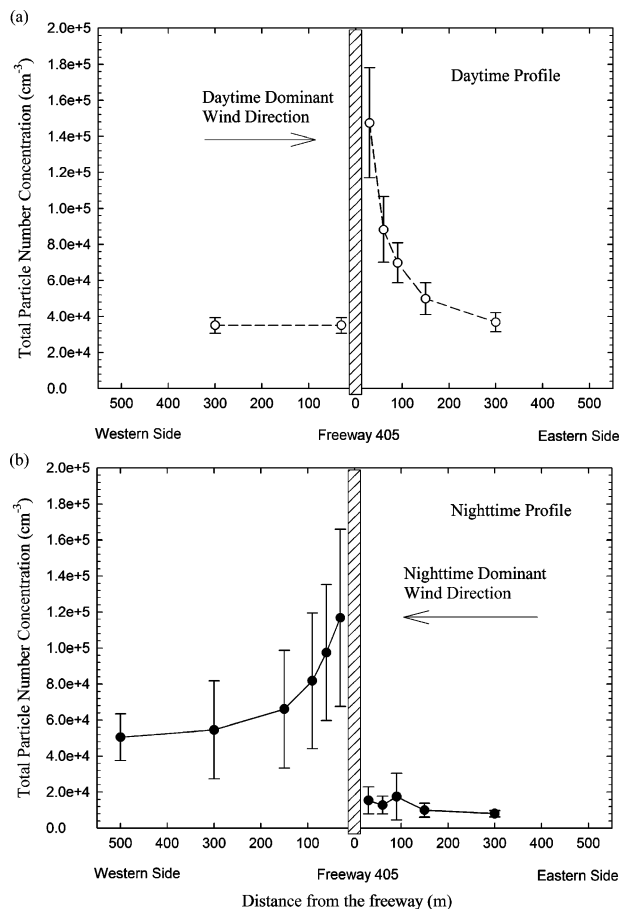
FIGURE 3. Spatial variations of total particle number concentration near freeway I-405 (a) during the day and (b) at night.

FIGURE 4. Spatial variations of PM10 and PM2.5 mass concentrations near freeway I-405 at night.

along wind trajectory at night were about two times the sampled perpendicular distances in Figure 2.

Spatial Profiles of Particle Mass and Number Concentrations. Figure 3 shows the spatial profile for total particle number concentration measured by the CPC (a) during the day and (b) at night. Circles represent mean particle number concentrations measured at different distances from the I-405 freeway. Error bars represent one standard deviation of measured particle number concentration. The daytime and nighttime dominant wind directions are also shown in the figure.

No significant concentration gradient was observed for particle number concentrations on the eastern (upwind) side of the I-405 freeway at night. Particle number concentrations decreased at a slower rate downwind of the freeway at night than during the day. This is mainly due to the lower wind speed and less atmospheric dispersion and constrained dilution effect at night. The number concentration at 300 m dropped only to approximately 40% of its original value (at 30 m). To see how far it takes for the downwind particle number concentration to drop to upwind background levels, we extended our downwind measurements to 500 m on 2 nights. In contrast to what happened during the daytime, where particle number concentration at 300 m downwind of the freeway was comparable to what was measured at 300 m upwind of the freeway, nighttime downwind particle number concentrations at 500 m were still significantly greater than those measured at the upwind site. This implies that freeway emissions have a much broader effect on local air quality at night due to the contribution from nearby vehicle emissions on local streets during the daytime. Although traffic volume on the I-405 freeway at night was only about 25% of the daytime traffic density, the measured particle number
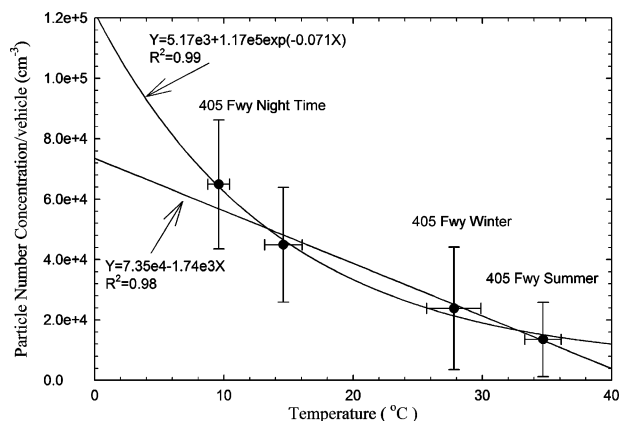
concentration at 30 m downwind of the freeway at night was about 80% of the daytime value. Detailed discussions on this topic are given in the next section.

In our previous study, particle mass concentration measured by a DataRam was found to decrease by only a few percent throughout the measured range. Since no particle size selective inlet was attached to the DataRam in our previous study, those values were viewed as indicators of general trends in total particle mass concentrations and overall changes with distance from the freeway. In this study, we used a DustTrak with either a PM_{10} or a $PM_{2.5}$ inlet to measure particle mass concentrations as indicated in Table 1. Figure 4 depicts the spatial profile of PM_{10} and $PM_{2.5}$ near the I-405 freeway at night. As expected, no concentration gradients were observed for upwind PM_{10} and $PM_{2.5}$ concentrations. Downwind PM_{10} concentrations were higher than those upwind with a slight concentration gradient away from the freeway. This was similar to what we reported previously for total particle mass during the daytime. The higher PM_{10} concentration downwind than upwind is probably due to resuspended dust and tire wear from the traffic. In addition, these data seemed to suggest that vehicle-caused PM coarse ($PM_{2.5-10}$) value is relatively smaller than that in the ambient. No significant difference from upwind and almost no concentration gradient were observed for $PM_{2.5}$ concentrations downwind of the freeway. These results confirm our previous conclusion that although vehicular exhaust on a major freeway is the primary source of nearby ambient ultrafine particles, they contribute relatively little in terms of direct emissions to particulate mass concentrations near freeways. Thus, regulating vehicular emissions in terms of PM_{10} and $PM_{2.5}$ may have little effect on number-based ambient particulate concentrations in urban areas.

Many epidemiology studies show an association between proximity to traffic and health effects, including increased prevalence of asthma, wheezing, recurrent respiratory illnesses in children, and hospital admissions for asthma (*21, 22*). These health effects are likely due to ultrafine particles or unique gas-phase pollutants or both from vehicular exhaust that have decay characteristics similar to ultrafine particles near freeways (*13, 14*). More importantly, many epidemiology studies have relied on distance from major roads or self-reported traffic density as a proxy for exposures. It is clear from Figure 3 that none of these is a good measure of exposure to traffic exhaust. Pollutant levels near roadways depend strongly on the direction of the wind and other meteorological parameters. Thus, diurnal and seasonal variations in meteorology have to be taken into consideration to accurately estimate near-roadway exposures.
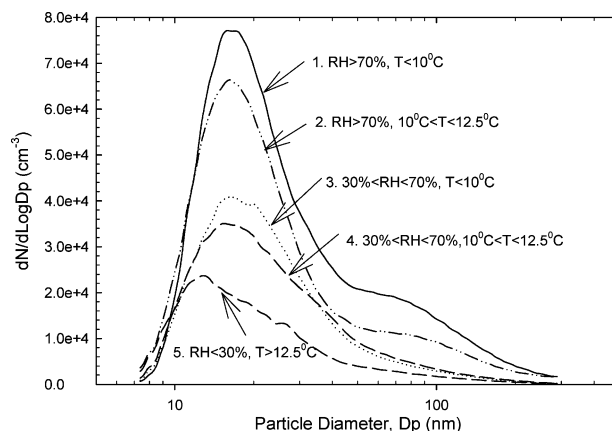
FIGURE 5. Traffic and wind corrected particle number concentrations at 30 m downwind from freeway I-405 at different temperatures.

FIGURE 6. Nighttime ultrafine particle size distributions at 30 m downwind from freeway I-405 at different temperatures and relative humidities.

Temperature and Relative Humidity Effects. It was noted in the discussion of Figure 3 that nighttime traffic volume was 25% of daytime volume, but it generated about 80% of the daytime particle number concentration. Lower wind speed and weaker atmospheric dilution alone could not explain this observed discrepancy because carbon monoxide (CO) concentrations at 30 m downwind of the freeway at night (0.5 ppm) were about 25% of what we observed during the daytime (2.0 ppm). CO concentrations at further downwind and upwind locations are constantly below the instrument detection limit of 0.1 ppm. Besides the dilution effect, the lower temperature at night may also contribute to this observed discrepancy. Colder ambient temperatures contribute to significantly increased nuclei mode particle formation in vehicle exhaust sampled on the freeway and in dilution tunnel experiments (*11, 23*). This observation suggests that there could be significant differences in the formation of ultrafine particles between day and night. In our previous study, higher particle number concentrations were also observed in winter than in summer near the I-405 freeway (*15*).

To illustrate the temperature effect on particle emission, Figure 5 summarizes all CPC data we obtained at 30 m downwind from the I-405 freeway in summer (*13*), in winter (*15*), and at night (this study). CPC data were corrected for traffic volume, and wind direction and speed. Error bars indicate one standard deviation for both temperature and particle number concentration. Data from the current study were dichotomized into temperature groups while previous data were presented as a single temperature group for summer and winter, respectively. There is a clear trend in particle number concentration per vehicle with respect to ambient temperature. The lower the ambient temperature, the more particles produced by each vehicle. Both linear and exponential curves fit the data very well within our measured temperature range as shown in Figure 5. Large differences between the two curves occur at temperatures below 10 °C. Future measurements at very low ambient temperature are needed to confirm which curve better relates particle number concentrations to temperature. It should be noted that there are other factors that may affect particle number concentrations at 30 m downwind of the freeway besides traffic volume, wind, and temperature. Atmospheric stability, turbulence intensity, and mixing height all change from day to night and may contribute to an even weaker atmospheric dispersion at nighttime. Another factor is vehicle speed. Much higher particle number concentrations have been reported from fast driving vehicles (*24*). The current study was done at around midnight where traffic speed was normally at 28.6 m/s (64 mi/h), considerably higher than what has been observed during the daytime when high traffic

volume caused the traffic sometimes to slow (~5 mi/h). In addition, a higher percentage of diesel trucks (10%) were observed at night than during the day (<5%), which may also contribute to higher particle number concentrations.

Temperature and relative humidity were closely related with a negative correlation (coefficient of $R^2 = 0.78$). It is therefore not immediately clear whether humidity alone, without a change in ambient temperature, had an effect on particle number concentration and size distribution. To investigate this question, size distributions at the site 30 m downwind of the freeway collected at night (VA) were averaged into groups with similar temperature and humidity conditions. The size distributions were very similar to each other when the meteorological conditions were comparable. Figure 6 clearly illustrates that number concentration increased with both decreasing temperature and increasing humidity. The grouping into different temperature and humidity conditions seems to indicate that humidity changes encountered during our study had a greater effect on particle number concentration than temperature changes. In particular, between the two cases 1 and 3 there is a large difference in relative humidity while the temperature is similar (on average 9.4 and 8.5 °C for cases 1 and 3, respectively). The large difference in the mode concentration associated with the two cases (a factor of about two) can therefore be most likely attributed directly to the increased relative humidity. This is supported by the similar trend between cases 2 and 4. This corroborates findings from dynamometer tests that studied separately the effects of temperature and humidity changes in the dilution air and showed that increasing humidity increased the number concentration of the nucleation mode (*25*).

The size distribution was affected by temperature and humidity changes in two ways: first by increased number concentrations as described above, and second by an apparent growth of the aerosol. As can be seen in Figure 6, the modal diameter increased from 13 to 16 nm from the case of relative humidity below 30% with temperature of 15.3 °C to the case of humidity between 30 and 70% with temperatures around 10 °C. For further increased humidity the mode remained at about 16 nm.

In summary, similar to what we observed during the daytime, ultrafine particle size distributions changed and particle number concentration decreased with downwind distance from the freeway at night, but at a slower rate. No $PM_{2.5}$ and very weak PM_{10} concentration gradients were observed downwind of the freeway at night. Ultrafine particle number concentration measured at 300 m downwind from the freeway was still distinguishably higher than upwind background concentration on low-wind nights. This implies

that freeway emissions have a much broader spatial effect on local air quality at night.

Acknowledgments

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