



A wide area of air pollutant impact downwind of a freeway during pre-sunrise hours

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ARTICLE INFO

Article history:

Received 10 October 2008

Received in revised form

13 February 2009

Accepted 13 February 2009

Keywords:

Vehicle-related air pollutants

Ultrafine particles

Mobile platform

Roadway

Exposure assessment

ABSTRACT

We have observed a wide area of air pollutant impact downwind of a freeway during pre-sunrise hours in both winter and summer seasons. In contrast, previous studies have shown much sharper air pollutant gradients downwind of freeways, with levels above background concentrations extending only 300 m downwind of roadways during the day and up to 500 m at night. In this study, real-time air pollutant concentrations were measured along a 3600 m transect normal to an elevated freeway 1–2 h before sunrise using an electric vehicle mobile platform equipped with fast-response instruments. In winter pre-sunrise hours, the peak ultrafine particle (UFP) concentration ($\sim 95\,000\text{ cm}^{-3}$) occurred immediately downwind of the freeway. However, downwind UFP concentrations as high as $\sim 40\,000\text{ cm}^{-3}$ extended at least 1200 m from the freeway, and did not reach background levels ($\sim 15\,000\text{ cm}^{-3}$) until a distance of about 2600 m. UFP concentrations were also elevated over background levels up to 600 m upwind of the freeway. Other pollutants, such as NO and particle-bound polycyclic aromatic hydrocarbons, exhibited similar long-distance downwind concentration gradients. In contrast, air pollutant concentrations measured on the same route after sunrise, in the morning and afternoon, exhibited the typical daytime downwind decrease to background levels within $\sim 300\text{ m}$ as found in earlier studies. Although pre-sunrise traffic volumes on the freeway were much lower than daytime congestion peaks, downwind UFP concentrations were significantly higher during pre-sunrise hours than during the daytime. UFP and NO concentrations were also strongly correlated with traffic counts on the freeway. We associate these elevated pre-sunrise concentrations over a wide area with a nocturnal surface temperature inversion, low wind speeds, and high relative humidity. Observation of such wide air pollutant impact area downwind of a major roadway prior to sunrise has important exposure assessment implications since it demonstrates extensive roadway impacts on residential areas during pre-sunrise hours, when most people are at home.

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1. Introduction

Air quality in the vicinity of roadways can be seriously impacted by emissions from heavy traffic flows. As a result, high concentrations of air pollutants are frequently present in the vicinity of roadways and may result in adverse health effects. These include increased risk of reduced lung function (Brunekreef et al., 1997), cancer (Knox and Gilman, 1997; Pearson et al., 2000), adverse respiratory symptoms (Van Vliet et al., 1997; Venn et al., 2001;

Janssen et al., 2003), asthma (Lin et al., 2002; McConnell et al., 2006), and mortality (Hoek et al., 2002).

Previous studies have shown elevated vehicle-related air pollutant concentrations and gradients downwind of roadways during daytime. Hitchins et al. (2000) measured concentrations of fine and ultrafine particles (UFP) at a distance of 15–375 m from a major roadway during the daytime. They found concentrations decayed to about half of the peak value (at the closest point to the roadway) at approximately 100–150 m from the roadway on the normal downwind side. Particle concentrations were not affected by the roadway at a distance farther than 15 m on the normal upwind side, indicating a sharp gradient of fine and ultrafine particles. Similar studies were conducted by Zhu et al. (2002a,b), who measured ultrafine particles, CO, and black carbon (BC) on the

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upwind (200 m) and downwind (300 m) sides of a freeway in Los Angeles during the daytime. Peak concentrations were observed immediately adjacent to the freeway, with concentrations of air pollutants returning to upwind background levels about 300 m downwind of the freeway.

The few near-roadway studies conducted at night indicated larger areas of impact than during daytime. UFP concentrations at night were reported by Zhu et al. (2006), who conducted measurements upwind (300 m) and downwind (500 m) of a freeway from 22:30 to 04:00. Although traffic volumes were much lower at night (about 25% of peak) particle number concentrations were about 80% of the daytime peak 30 m downwind of the freeway, with UFP concentrations of $\sim 50\,000\text{ cm}^{-3}$ about 500 m downwind of I-405, a major Los Angeles freeway during the night. Fruin and Isakov (2006) measured UFP concentrations in Sacramento, California, near the I-50 freeway between 23:00 and 01:00 and found 30–80% of maximum centerline concentrations (measured on a freeway overpass) 800 m downwind.

In the present study, the use of a full-size, motorized mobile platform (MP) allowed more pollutants to be measured than previous nighttime studies and with improved spatial and temporal resolution. While traveling at normal vehicle speeds, an instrumented mobile platform allows measurements over greater distances and in shorter times (Bukowiecki et al., 2002a,b, 2003; Canagaratna et al., 2004; Kittelson et al., 2004a,b; Khlystov and Ma, 2006; Kolb et al., 2004; Pirjola et al., 2004, 2006; Unal et al., 2004; Weijers et al., 2004; Westerdahl et al., 2005; Yao et al., 2005; Isakov et al., 2007; Baldauf et al., 2008; Fruin et al., 2008). However, to date, such studies have focused almost entirely on daytime and evening periods.

In the present study, air pollutant concentrations were measured over a wide area on the south and north sides of the I-10 freeway in west Los Angeles, California, 1–2 h before sunrise in the winter and summer seasons of 2008 using an electric vehicle mobile platform equipped with fast-response instruments. We observed a much wider area of impact downwind of the freeway than reported in previous daytime and evening studies, consistent with low wind speed, absence of turbulent mixing, and nocturnal radiation inversions. Our pre-sunrise results were also strikingly different from those we observed for the same route during the daytime. Our observation of a wide area of impact during pre-sunrise hours, up to about 600 m upwind and 2000 m downwind, has significant implications for exposures in residential neighborhoods adjacent to major roadways.

2. Methods

2.1. Mobile platform and data collection

A Toyota RAV4 sub-SUV electric vehicle served as the mobile platform, with self-pollution eliminated by the non-polluting nature of the vehicle. Table 1 shows a complete list of sampling instruments and equipment installed on the mobile platform. The time resolution for most instruments ranged from 5 to 10 s except the Aethalometer, which had 1 min time resolution. The instrument power supply and sampling manifold were similar to that described by Westerdahl et al. (2005).

Calibration checks and flow checks were conducted on a bi-monthly and daily basis, respectively, as described in Kozawa et al. (2009). For calibrations, a standard gas containing a mix of NO and CO was diluted using an EnviroNics 9100 Multi-Gas Calibrator and Teledyne API Zero Air System (Model 701) to calibrate the CO and NO/NO_x analyzers. CO₂ was calibrated with zero air and span gas cylinders from Thermo Systems Inc. A DryCal DC-lite flow

Table 1
Monitoring instruments on the mobile platform.

Instrument	Measurement Parameter	Time Resolution
TSI Portable CPC, Model 3007	UFP Count (10 nm–1 μm)	10 s
TSI FMPS, Model 3091	UFP Size (5.6–560 nm)	10 s
TSI DustTrak, Model 8520	PM _{2.5} Mass	5 s
Magee Scientific Aethalometer	Black Carbon	1 min
EcoChem PAS 2000	Particle Bound PAH	5 s
Teledyne API Model 300E	CO	20 s
LI-COR, Model LI-820	CO ₂	10 s
Teledyne-API Model 200E	NO _x , NO, NO ₂	20 s
Vaisala Sonic Anemometer and Temperature/RH Sensor	Local Wind Speed and Direction, Temperature, Relative Humidity (RH)	1 s
Stalker LIDAR and Vision Digital System	Traffic Documentation, Distance and Relative Speed	1 s

meter, with a flow range of 100 ml min⁻¹ to 7 L min⁻¹ and an accuracy of $\pm 1\%$, was used to check the flows of each instrument.

2.2. Route

For pre-sunrise measurements, the mobile platform was driven on a fixed route over three days in the winter season and two days in the summer season of 2008. The route covered a total length of about 3600 m approximately perpendicular to the I-10 freeway in Santa Monica, California (Fig. 1). The solid line in Fig. 1 shows the section of the route over which the mobile platform traveled about 8–10 times during each monitoring period, reaching about 1200 m south of the freeway. The dashed line shows the extended section of the route, over which the mobile platform traveled 2–4 times during each monitoring period, reaching about 2600 m south of the freeway. The pre-sunrise route crossed a number of local surface streets; these are shown in Fig. 1 together with their normal distances to the freeway as measured from Google Map. The route was selected because it passed under the I-10 freeway, and because there was little traffic flow on the route itself or on the perpendicular surface streets (e.g. Olympic Blvd., Pico Blvd. etc.) during pre-sunrise hours. Hence, the majority of measurements were not significantly affected by local surface street traffic. The route also passed through a dense residential neighborhood where the elevated air pollutant concentrations have significant exposure implications.

During sampling, the mobile platform was intentionally stopped to avoid localized impacts from individual vehicles whenever necessary. During data reduction, pollutant concentration spikes, if verified from videotape to be caused by a nearby vehicle, were excluded from the analysis.

2.3. Real-time traffic flow

Traffic flows were collected or measured on the I-10 freeway, the pre-sunrise route itself, and the major surface streets transecting the pre-sunrise route. Real-time traffic flow on the freeway was obtained from the Freeway Performance Measurement System (PeMS) provided by the UC Berkeley Institute of Transportation. Sensors were located at the Dorchester Station, about 300 m from the intersection of the pre-sunrise route and the freeway. Since there were no on-ramps or exits between the Dorchester Station and our route, the PeMS data accurately represented the traffic flow on the I-10 freeway where our route passed under the freeway. Traffic flow on the pre-sunrise route itself was monitored and recorded by a Stalker Vision Digital System on the mobile platform. The recorded videos were replayed and vehicles on the pre-sunrise



Fig. 1. Pre-sunrise route. The solid line indicates the route 1000 m and 1200 m north and south of the I-10 freeway, respectively. The dashed line indicates the route extended to 2600 m south of the I-10 freeway. Mobile platform measurements on this route were continuous; circles indicate location of cross streets, not fixed site monitoring.

route were manually counted. Traffic flows on the major cross streets (e.g. Olympic Blvd., Pico Blvd., and Ocean Park Blvd.) were manually counted during the winter season on a weekday at times similar to when the pre-sunrise measurements were conducted.

2.4. Data analysis and selection of key pollutants

Data were adjusted for the varying response times of the instruments on the mobile platform to synchronize the measurements. NO_x , CO, CO_2 , and particulate data (UFP, BC, and $\text{PM}_{2.5}$ mass) were synchronized with particle-bound polycyclic aromatic hydrocarbon (PB-PAH) data measured by the PAS instrument, which had the fastest response time. NO, UFP, and PB-PAH were selected in the present study for detailed spatial analysis because of their rapid and large variation on and near roadways. The overall response time for the PAS instrument was determined by comparing the time of signal peaks in the PB-PAH time-series to the corresponding time of acceleration of a vehicle in front of our mobile platform (as recorded on videotape). This time difference was less than 10–15 s and includes the transport time (typically a few seconds) for the plume from the emitting vehicle to reach the inlet of the sampling duct of the mobile platform. Given the short response times of our instruments and our driving speeds of 5–15 MPH, the spatial resolution of our mobile platform measurements was typically in the range of 25–75 m, with the finer spatial

resolution (~ 25 m) near the edges of the freeway where we drove more slowly.

Measurements were made continuously over the entire route, not at fixed stationary sites. The measured real-time concentrations of UFP, PB-PAH, and NO along the pre-sunrise route were averaged for each intersection using a few data points measured at and immediately adjacent to the intersection. Although the peak air pollutant concentration always occurred downwind of the I-10 freeway, its value changed with time due to changing traffic volumes on the I-10 freeway and varying meteorological conditions, so peak pollutant concentrations were used to calculate normalized relative pollutant concentrations. For example, in the winter season, the measured averaged peak UFP concentration was about $95\,000\text{ cm}^{-3}$, but the instantaneous peak values varied in the range of $62\,000$ – $135\,000\text{ cm}^{-3}$ (four to nine times the background concentrations).

3. Results and discussion

3.1. Meteorological data

Meteorological conditions, including atmospheric stability, temperature, relative humidity, wind speed and wind direction, play an important role in determining air pollutant concentrations and gradients along and downwind of roadways. During each run, the mobile platform was periodically stopped at locations along the pre-sunrise route to obtain wind data from on-board instruments (Table 2). These data were compared with the measurements from the Santa Monica Airport (SMA) located about 1500 m downwind of the I-10 freeway and in the immediate vicinity of the route. Both the averaged wind speeds measured by the mobile platform and by the SMA were quite low during pre-sunrise hours, in a range of 0 – 1.0 m s^{-1} and the averaged difference between the two measurements was about 0.3 m s^{-1} . Temperature and relative humidity were obtained from SMA data.

Fig. 2 shows the wind roses and vector-averaged wind orientation for five days, March 7, 12, 18, June 30, and July 2, from data collected by instruments on the mobile platform. Wind speeds were low during the pre-sunrise hours, with monitoring period averages ranging from 0.0 to 1.0 m s^{-1} . The averaged wind directions measured by the mobile platform indicated a predominant direction of N/NE/NW during the pre-sunrise runs, which agreed reasonably well with airport wind direction data. For this predominant wind direction, the north side of the I-10 freeway was upwind; the south side downwind. Although having a predominant direction from north, the wind was not completely perpendicular to the I-10 freeway. Hence, the distances pollutants traveled from the freeway to various locations along the route, including the major cross-surface streets, were generally longer than indicated by distances shown in Fig. 1. For example, the straight perpendicular distance of Ocean Park Blvd. to the I-10 freeway is ~ 950 m, whereas for the averaged wind direction of 25° for the pre-sunrise run, the distance pollutants traveled was ~ 1050 m. However, due to the variability of meteorological conditions, the perpendicular distances were used to indicate impact distances in the present study.

While detailed thermal structure data for the lowest layers of the atmosphere in the area of our pre-sunrise route were not available, the available data indicate the days sampled had stable (i.e., vertical) temperature profiles or strong nocturnal radiation inversions in the hours before sunrise. Data recorded at the Santa Monica Airport indicated the nights on which sampling took place were clear up to at least 3000 m, and had either offshore flow or a weak land breeze, also consistent with clear skies; clear skies are conducive to the formation of nocturnal surface inversions due to enhanced radiative heat loss in the infrared. Data collected by the

Table 2
 Meteorological conditions during pre-sunrise runs (2008).

Date	Measurement period	Sunrise	Atmospheric Stability from LAX Profiler data	Wind Speed ^a (m s ⁻¹)		Wind Direction ^a (°)		Temperature (°C)	Relative Humidity (%)
				MP	SMA	MP	SMA	SMA	SMA
March 7	6:20–7:50 ^b	7:14 ^b	N.D. ^c	0.9	1.0	13	5	11	79
March 12	6:00–7:30	7:07	Surface inversion to 250–300 m	1.0	1.0	53	20	13	66
March 18	6:10–7:20	6:59	Surface inversion to 190 m	0.8	1.0	6	45	9	61
June 30	4:00–6:30	5:45	Stable to 190 m, inversion above	0.7	0.0	288	0	17	87
July 2	4:30–6:45	5:45	Stable to 260 m, inversion above	0.7	1.0	315	340	17	84

^a Averaged values for the measured period.

^b Time corrected to Pacific Day Light Time (PDT); change from PST to PDT occurred on March, 9, 2008.

^c Profiler came online the following evening. The following night (3/8) experienced a surface inversion for the entire night.

South Coast Air Quality Management District (SCAQMD) at the Los Angeles Airport (LAX), ~8 km south of pre-sunrise route, were also consistent with an inversion or stable conditions at the surface. On 3/10 and 3/18, the data showed temperature inversions from the

lower edge of the measurements at 130 m up to 190 m or more, respectively. On 6/30 and 7/2, the profiles were stable from 130 to 190 or 260 m, respectively, with capping inversion layers above. Wind speeds during the pre-sunrise hours were too low to create

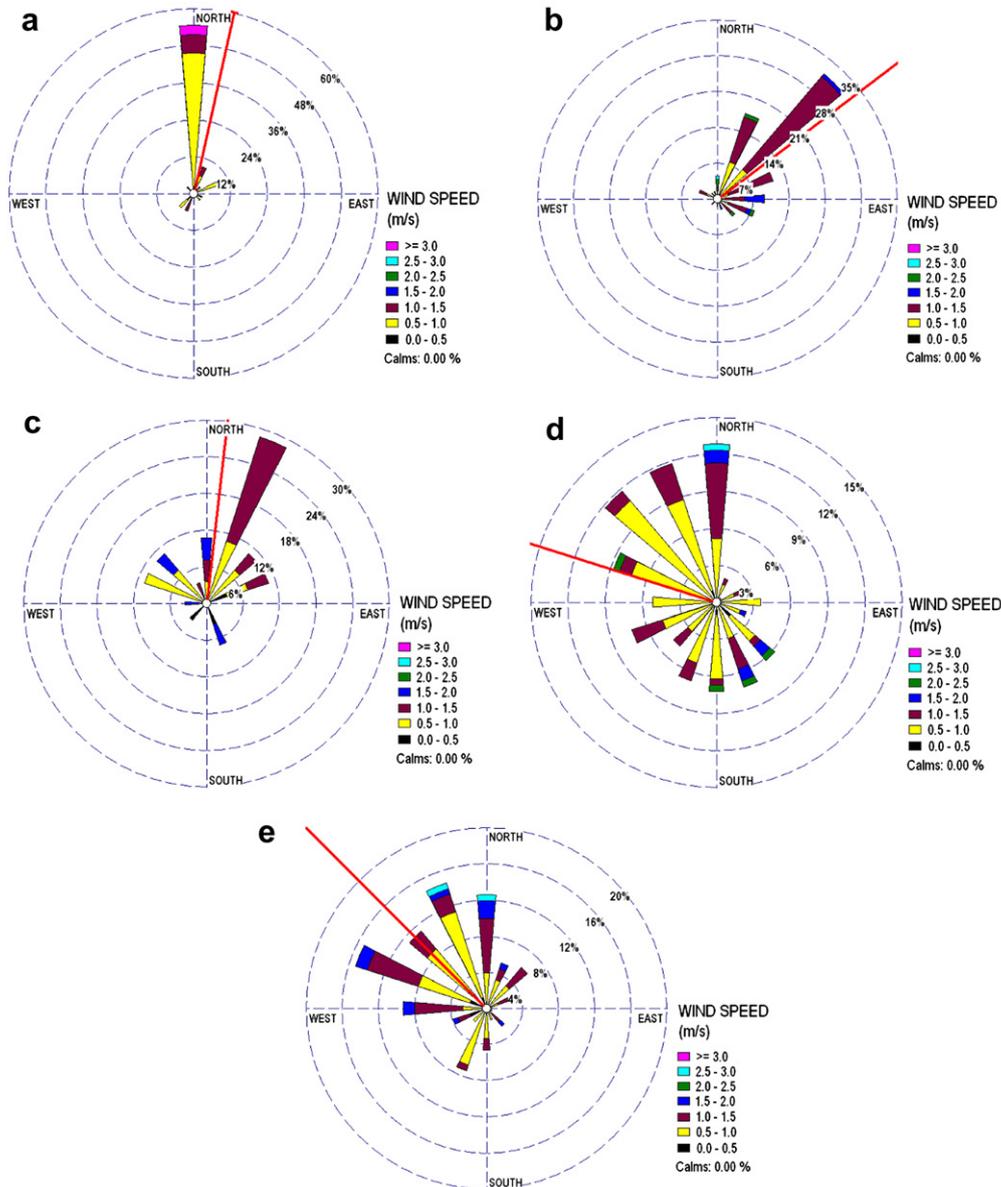


Fig. 2. Wind roses for pre-sunrise sampling hours. (a) March 7; (b) March 12; (c) March 18; (d) June 30; (e) July 2. The thin line in each wind rose indicates vector-averaged wind orientation.

appreciable vertical mixing in the presence of these temperature profiles, and the shallow mixed layer was likely thinner in March than in June/July.

3.2. Observation of a wide impact area downwind of the freeway during pre-sunrise hours

As shown in Fig. 3, a wide impact area of elevated UFP concentrations, up to 2000 m downwind and 600 m upwind of the I-10 freeway, was observed during the pre-sunrise hours on the monitoring days in the two seasons. In this wide impact area, elevated UFP concentration extended beyond Donald Douglas Loop N located on the south side and 1200 m downwind of the freeway (Fig. 3). Here, 1200 m downwind, the average UFP concentrations during the winter sampling hours, typically 06:00–07:30, were as high as $\sim 40\,000\text{ cm}^{-3}$. Only at a downwind distance of about 2600 m (Palms Blvd.), did the UFP concentration drop to $\sim 15\,000\text{ cm}^{-3}$, comparable to the upwind background level.

In the winter season, the peak UFP concentration was approximately $95\,000\text{ cm}^{-3}$ a few tens of meters downwind of the freeway. Upwind, the concentration dropped sharply to around $40\,000\text{ cm}^{-3}$ 30 m upwind (Virginia Avenue) and returned to background levels of $\sim 15\,000\text{ cm}^{-3}$ at $\sim 800\text{ m}$ on the upwind side, creating a moderate upwind gradient north of the I-10 freeway (Fig. 3). Interestingly, the upwind impact distance during the pre-sunrise hours, $\sim 600\text{ m}$, was far greater than that of $\sim 15\text{ m}$ observed during the day by Hitchins et al. (2000) and also greater than that measured by Zhu et al. (2002b). This may be caused by the occasionally variable wind direction during the pre-sunrise hours for which the nominal upwind side of the I-10 freeway could temporarily become downwind. These occasional impacts on the nominal upwind side of the freeway appear to have had substantial influence on the averaged upwind UFP concentrations due to their otherwise low levels.

As seen in Fig. 3, the UFP concentration also decreased on the downwind side, but much more slowly than on the upwind side. At a downwind distance of about 600 m from the freeway, UFP concentrations during winter were about twice those on the upwind side ($50\,000\text{ cm}^{-3}$ vs. $22\,000\text{ cm}^{-3}$). Even 950 m downwind, at the intersection of Ocean Park Blvd., the UFP concentration remained as high as $45\,000\text{ cm}^{-3}$, higher than at 30 m upwind. These pronounced differences in gradients of UFP concentrations resulted in strong contrasts between the upwind and downwind sides of the I-10 freeway during pre-sunrise hours (Fig. 3).

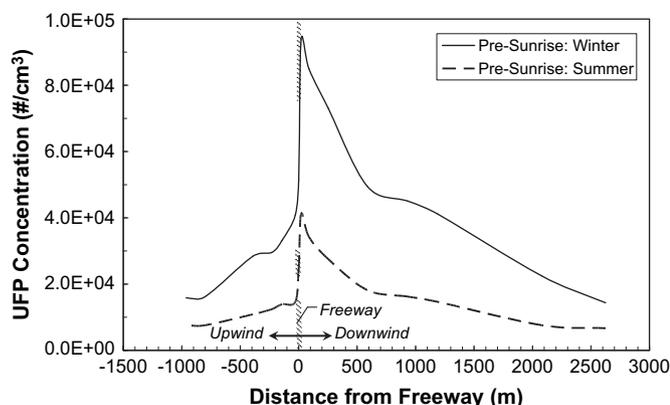


Fig. 3. Ultrafine particle concentrations and gradients along the pre-sunrise route. Positive distances are downwind and negative distances upwind from the I-10 freeway. Data were acquired continuously, up to the edges of the freeway.

As shown in Fig. 4, NO and PB-PAH exhibited concentration gradients similar to UFP along the route during the pre-sunrise hours. Peak concentrations of NO and PB-PAH (on the downwind side) were about 165 ppb and 55 ng m^{-3} , respectively, in the winter season. Upwind, NO and PB-PAH concentrations dropped rapidly to 70 ppb and 30 ng m^{-3} , respectively, at a distance of about 150 m. In contrast, on the downwind side, NO and PB-PAH concentrations of 70 ppb and 30 ng m^{-3} , respectively, extended to a distance of about 1200 m from the freeway (NO and PB-PAH data were unavailable for summer measurement due to instrument problems during the pre-sunrise runs).

Fig. 5 shows normalized UFP concentrations on the two sides of I-10 freeway during the pre-sunrise hours in the winter and summer seasons. UFP concentrations were normalized for each complete run traveled on our route, and then averaged together for all the runs for each season. While there was little or no traffic on our route during the pre-sunrise hours, vehicle counts on the same route during the day were much higher and emissions from these vehicles significantly and frequently affected measurements by the mobile platform. Moreover, the pre-sunrise route was only driven once in the morning after sunrise and once in the afternoon, in contrast to multiple times in the pre-sunrise period. For both of these reasons, comparison between pre-sunrise and morning/afternoon measurements on the pre-sunrise route are not meaningful. Instead, we show normalized data from Zhu et al. (2002b), which were not affected by local traffic, to compare with our pre-sunrise measurements.

As Fig. 5 illustrates, pre-sunrise UFP concentration gradients in the present study exhibited very different behavior than the typical narrow daytime UFP gradients measured by Zhu et al. (2002a,b). In our pre-sunrise measurements, UFP concentrations remained elevated above the background level up to $\sim 600\text{ m}$ upwind of the freeway versus only $\sim 17\text{ m}$ upwind for the Zhu et al. (2002b) daytime measurements. On the downwind side in the Zhu et al. (2002b) measurements, UFP concentrations dropped to about 25% of the peak concentration 300 m downwind of the freeway during the day, but in the present study, in strong contrast, the UFP concentrations remained about 40% of the peak as much as 1200 m downwind of the freeway, and was above background levels out to $\sim 2000\text{ m}$ during the pre-sunrise hours.

To quantify these differences in UFP concentrations an equation of the form $C = a + e^{-bx}$ was used to fit our observed relative UFP concentrations downwind of the I-10 freeway during pre-sunrise hours, as well as the daytime data reported by Zhu et al. (2002b). As seen in Fig. 6, the decay constant is a factor of five higher for the

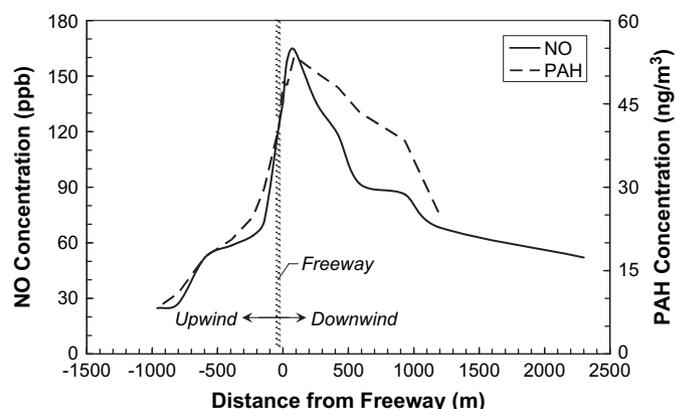


Fig. 4. Average NO and PB-PAH concentrations and gradients, along the pre-sunrise route in the winter season. Positive distances are downwind and negative distances upwind from the I-10 freeway.

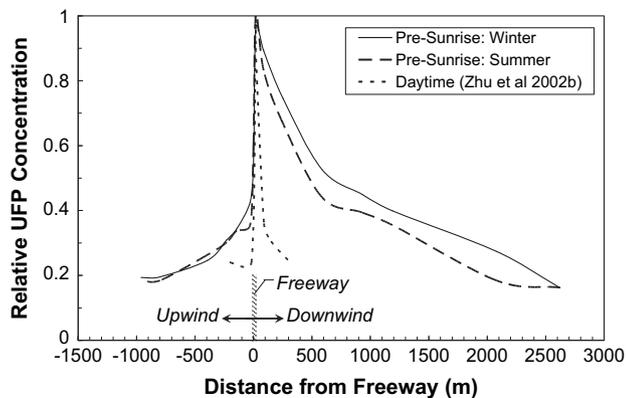


Fig. 5. Relative averaged UFP concentrations and gradients along the pre-sunrise route by season and compared with Zhu et al. (2002b). Positive distances are downwind and negative upwind from the I-10 freeway. Data were acquired continuously for pre-sunrise measurements, up to the edges of the freeway.

daytime vs. the pre-sunrise period, with values of *b* of 0.0098 and 0.0018, respectively.

Pre-sunrise relative UFP concentrations exhibited similar trends in both winter and summer (Fig. 5). Although UFP concentrations in the summer were about 40% those in the winter (due to lower traffic flows on the I-10 freeway, as discussed below), the similar trends in relative UFP concentration imply similar UFP propagation during the pre-sunrise hours in the two seasons although meteorological conditions were somewhat different.

3.3. Correlation of pollutant concentrations with traffic counts on I-10 freeway

PeMS data showed a similar diurnal traffic pattern on the I-10 freeway on different weekdays during the pre-sunrise hours in both winter and summer (Fig. 7b). Traffic counts on the freeway exhibited an approximately linear increase with the time. However, during 04:00–05:30 (when summer measurements were conducted) traffic counts were lower in summer than in winter. We attribute part of the lower traffic counts in summer to most schools being closed and vacation season in summer, as well as the dramatic increase in gasoline prices between March and July 2008, resulting in a significant overall reduction in vehicle miles traveled. Also, sunrise was about one hour and fifteen minutes earlier in

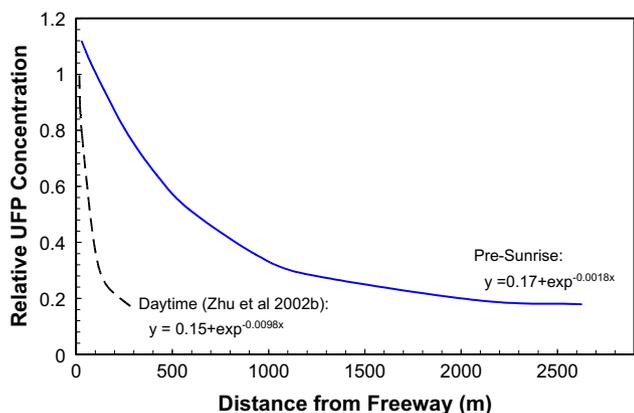


Fig. 6. Exponential fits to the downwind relative UFP concentrations with distance from the I-10 freeway during pre-sunrise hours, compared with fit to daytime data downwind of the I-405 freeway by Zhu et al. (2002b). Data were acquired continuously for pre-sunrise measurements, up to the edges of the freeway.

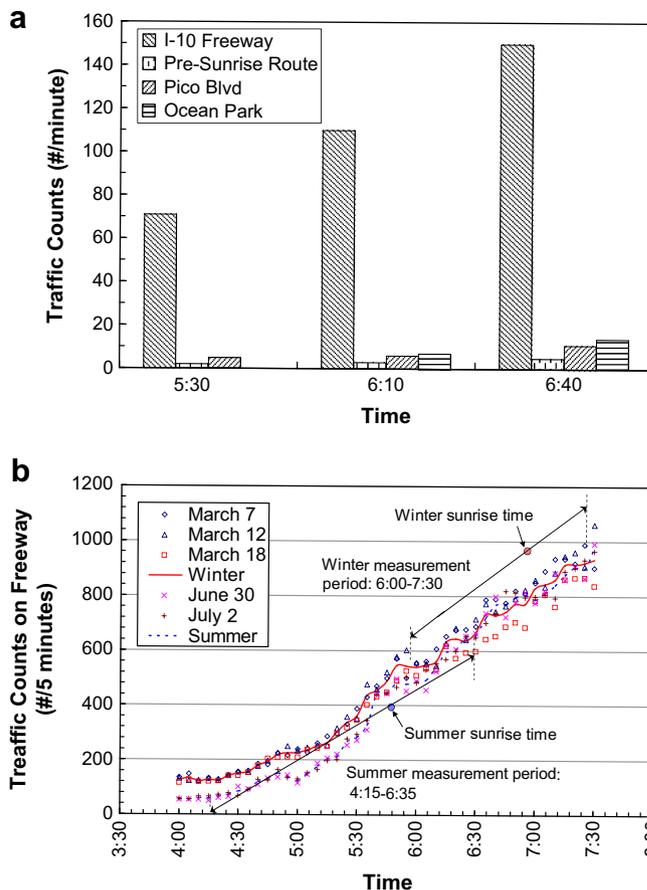


Fig. 7. (a) Comparison of traffic volumes on the I-10 freeway, pre-sunrise route, Pico Blvd., and Ocean Park Blvd. during pre-sunrise hours on a typical weekday; (b) Traffic counts on the I-10 freeway during pre-sunrise measurements; solid line represents averaged count of the three days in the winter season, and dashed line for two days in the summer season. Sunrise times shown here were averaged for each season.

summer (~05:45) than in winter (~07:00), which required an earlier measurement period in summer (~04:15–06:30) compared to winter (~06:00–07:30), and corresponds to much lower overall traffic counts during the pre-sunrise measurement periods in summer.

During the measurement period in winter, traffic counts on the freeway increased from ~530 to ~900 vehicles per 5 min, while in summer counts increased from ~60 to ~620 vehicles per 5 min. Assuming a linear increase of traffic counts with time, the average traffic counts during the pre-sunrise measurements periods, winter versus summer, were ~715 vs. 340 vehicles per 5 min, resulting a ratio of ~2.1. This ratio of seasonal traffic counts compares well with the ratio of the UFP concentrations measured in the winter vs. summer of ~2.2–3.0, depending on distance from the freeway (Fig. 3). It should be noted that the sunrise times during the winter (March) measurements, because they occurred just after the switch to Pacific daylight time (PDT), were close to the latest annual (local) sunrise times, and thus may represent roughly the upper limit for the freeway impact throughout the year.

We attribute the relatively high pollutant concentrations we observed downwind of the I-10 freeway during pre-sunrise hours to emissions of vehicles traveling on the I-10 freeway, combined with strong inhibition of vertical mixing due to stable or inverted temperature profiles near the surface. Fig. 8 shows the UFP and NO concentrations measured at Ocean Park Blvd., ~950 m downwind, vs. the traffic counts on the freeway during the pre-sunrise hours on

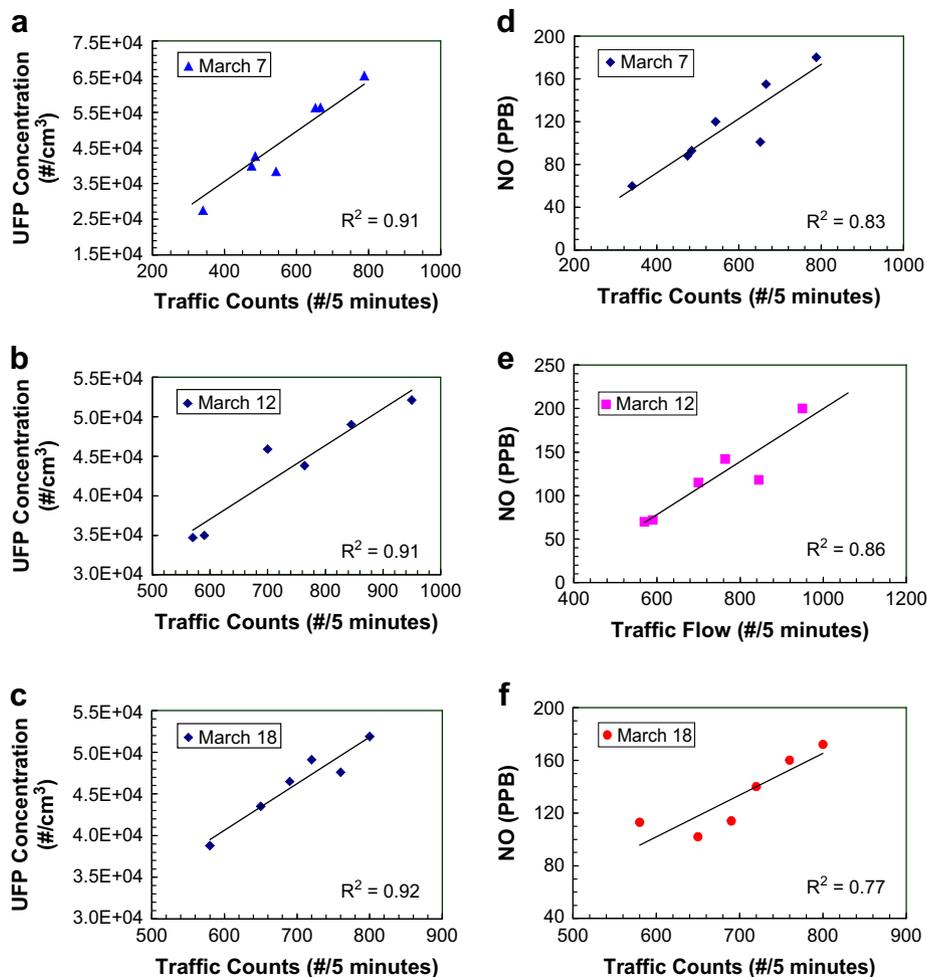


Fig. 8. Linear regressions between UFP and NO concentrations at Ocean Park Blvd. (950 m downwind of I-10 freeway), and traffic counts on the freeway during the pre-sunrise hours in the winter season.

three mornings of the pre-sunrise runs in the winter season. Both the freeway traffic counts (Fig. 7b) and pollutant concentrations increased rapidly during the pre-sunrise hours, and exhibited a strong correlation with each other. For UFP, the values of squared Pearson correlation coefficients (r^2) were above 0.90 and for NO, above 0.77 (nitric oxide data were unavailable for summer measurements due to instrument problems during the pre-sunrise runs). Strong correlations at other distances from the freeway were also found between UFP concentrations and traffic counts on the freeway. For example, the correlation coefficients, r^2 for UFP measured at Pearl St. for three winter sampling days, were above 0.85.

Based on our videotape observations and the traffic counts we conducted on surface streets, as well as the strong correlations presented in Fig. 8, we believe the measured concentrations of air pollutants during the pre-sunrise hours were predominantly determined by the traffic counts on the I-10 freeway, and that the impact of local surface street traffic was minor. Traffic volumes on the pre-sunrise route itself were only about 2% of those on the I-10 freeway at corresponding times. Traffic volumes on the three major surface streets crossing the pre-sunrise route, Ocean Park Blvd., Pico Blvd., (downwind of the freeway), and Olympic Blvd. (upwind of the freeway) were also low, only about 8%, 6%, and 6%, respectively, of those on the freeway. Most of this early-morning cross traffic for our measurement route encountered green lights. If the emissions of the occasional vehicles on these surface streets were significant, the pollutant concentrations measured downwind of

the streets should have been higher than upwind, but this was not the case; no significant gradients in concentration were observed between the two sides of these streets. Hence, the contribution of emissions from vehicles on the surface streets to our pre-sunrise measurements ranged from minor to insignificant compared to emissions from freeway traffic.

One case in which we find evidence of a minor contribution from non-freeway emissions involves the shallow shoulder in UFP concentrations on Ocean Park Blvd. (~950 m downwind) and shown in Fig. 3. Traffic counts on this major surface street were ~8% of the freeway counts (Fig. 7a), which may have resulted in a small local UFP, NO, and PB-PAH contribution to the measured concentration. A local contribution of ~6% traffic count on Pico Blvd. is not apparent in the measured UFP concentration in Fig. 3, probably due to the closer proximity of Pico Blvd. to the I-10 freeway (~250 m downwind).

Although the mobile platform measurements could be affected by emissions from vehicles occasionally encountered on the pre-sunrise route or cross-surface streets, these encounters typically exhibited only a short, transient spike of elevated concentrations. Furthermore, the overall pre-sunrise concentrations and gradients presented were averaged from 18 to 24 runs in winter and 12–16 runs in summer and for all these reasons were generally not significantly affected by emissions from occasionally encountered nearby vehicles. The Santa Monica Airport (SMA), a small local airport, located south of the pre-sunrise route, had no impact on

any of our pre-sunrise measurements since it has severely restricted hours to minimize noise pollution, and was closed during all of our pre-sunrise experiments.

3.4. Size distribution of UFP along pre-sunrise route

The use of a fast mobility particle sizer (FMPS), with its 10 s scans, allowed accurate monitoring of the changing particle size distribution as a function of distance away from the freeway. Fig. 9 shows average UFP size distributions for five downwind and two upwind intersections during the pre-sunrise hours in the winter season, with decreasing particle numbers and increasing sizes as distance downwind increases, until the upwind size distribution was roughly matched at 2600 m. At the downwind intersections up to 1200 m from the freeway, two to four times higher concentrations of ultrafine particles less than 40 nm were observed compared with upwind locations (Fig. 9).

For the intersections nearest the freeway (e.g. Kansas, 100 m downwind, and Pico, 250 m downwind), bi-modal peaks in the size ranges of ~9–12 nm and 16–20 nm were observed. For downwind intersections farther away and for the upwind intersections, UFP peaks observed were typically ~9–12 nm and ~16–20 nm, and 28–35 nm, corresponding to freshly generated UFP and aged particles, respectively. UFP size distributions at a distance of 2600 m downwind (Palms Blvd.) and 1000 m upwind (Harvard St), considered “background” locations, were similar with a dominant mode at 30–60 nm.

In summer, downwind UFP size distributions also had a small mode of 9–12 nm. The persistence of the 9–12 nm peak in UFP concentrations during pre-sunrise hours over a wide area can be attributed to increased condensation of organic vapors and slower rates of conversion to larger particles for the cooler, stable air conditions prior to sunrise during our winter and summer campaigns. These conditions would also promote the more elevated UFP concentrations observed in our pre-sunrise runs compared with daytime runs.

3.5. Pre-sunrise vs. daytime concentrations in present study: exposure implications

Although traffic volumes on the freeway during the pre-sunrise hours were markedly lower than during the daytime (~30–80% of peak congestion traffic volumes), air pollutant concentrations measured prior to sunrise were significantly higher than in the

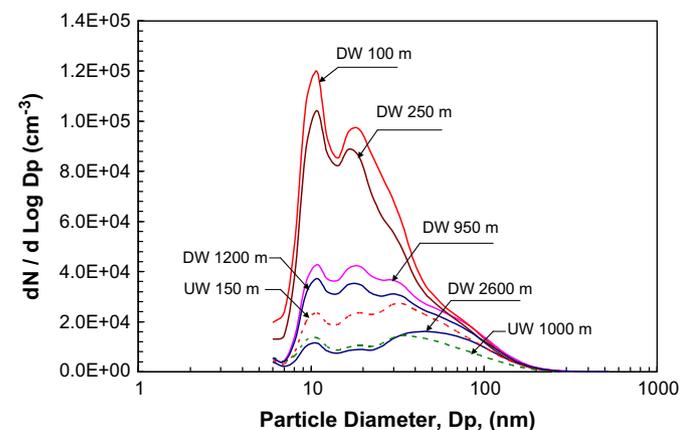


Fig. 9. Size distributions of ultrafine particles measured by a TSI Model 3091 FMPS at upwind (UW) and downwind (DW) intersections during the pre-sunrise hours in the winter season.

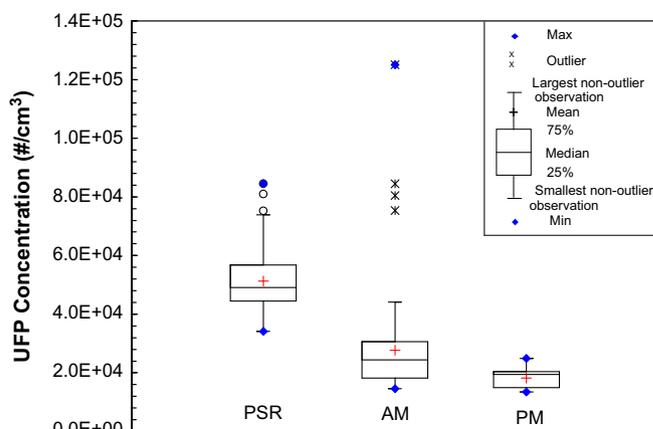


Fig. 10. Comparison of UFP concentrations on Pearl St. (600 m south of I-10 freeway) at different times in winter: pre-sunrise (PSR), morning (AM), and afternoon (PM).

morning or afternoon runs. Fig. 10 shows the UFP concentrations measured at Pearl St., ~600 m south of the freeway, during the pre-sunrise and daytime hours in winter. The median UFP concentrations were $49\,000\text{ cm}^{-3}$, $24\,000\text{ cm}^{-3}$, and $19\,000\text{ cm}^{-3}$ for the pre-sunrise, morning, and afternoon, respectively. Clearly, there was sufficient traffic flow on the I-10 freeway combined with the meteorological conditions during pre-sunrise hours to result in elevated concentrations of UFP, NO, and PB-PAH over a wide area of the downwind (up to ~2000 m) and upwind (up to ~600 m) residential neighborhoods. Since the pre-sunrise hours are at a time when most people are in their homes, our observations imply the potential for elevated exposures for many more residents in these neighborhoods, adjacent to freeways; far above the numbers of people that live within the ~300–500 m range reported in earlier daytime and evening studies. Additional measurements in the pre-sunrise period downwind of other major roadways should be conducted to confirm our novel findings.

4. Conclusions

A wide impact area of elevated pollutant concentrations on the downwind (up to ~2000 m) and upwind (up to ~600 m) sides of a freeway was measured during the pre-sunrise hours under typical meteorological conditions characterized by weak winds and a strong radiation inversion. To make these measurements, a mobile platform, equipped with fast-response monitoring instruments, drove along a transect crossing under the I-10 freeway and passing through a large residential neighborhood. On the upwind side of the freeway, air pollutant concentrations dropped quickly, but remained elevated up to ~600 m. On the downwind side, air pollutant concentrations (UFP, PAH, NO) dropped much more slowly and extended far beyond the typical ~300 m distance associated with the return to background pollutant levels observed in previous studies conducted during daytime. For example, elevated ultrafine particle concentration of about $40\,000\text{ cm}^{-3}$ extended to ~1200 m downwind of the freeway in the winter season, which was about 40% of the peak UFP concentration adjacent to the freeway.

Although traffic volumes during the pre-sunrise hours were lower than during the day, the UFP concentrations were significantly higher in the pre-sunrise period. We attribute this pre-sunrise phenomenon to strong atmospheric stability, low wind speeds ($\sim 0\text{--}1\text{ m s}^{-1}$), low temperatures ($\sim 9\text{--}13\text{ }^{\circ}\text{C}$), and high humidities ($\sim 61\text{--}79\%$), facilitating longer lifetimes and slower transport of UFP before dilution and dispersion to background levels. Nocturnal inversions are

a widespread phenomenon particularly on clear nights, and our results suggest broad areas of elevated pollutants around major roadways are expected to be common in the early-morning hours. The implications of these observations for exposures to vehicle-related pollutants should be further explored.

Acknowledgements

The authors acknowledge support for this study by the California Air Resources Board, Contract No. 04-348. The authors wish to express their appreciation to Drs. Jorn Herner, Ying-Kuang Hsu, and Dane Westerdahl of ARB for their assistance, and Joe Cassmassi and Kelvin Durkee of the SCAQMD for their technical support. We also wish to thank Dr. Richard Turco at UCLA and Dr. Jun Wu at UC, Irvine, as well as Douglas Houston, Albert Chung, Hwajin Kim, Daniel Curtis and Shahir Masri at UCLA, for their contributions to the study. Helpful discussions with James Murakami at UCLA regarding the sources of meteorological data and their interpretation are greatly appreciated.

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