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Ultrafine particle exposures while walking, cycling, and driving along an urban residential roadway

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HIGHLIGHTS

- UFP was lower in morning (~60%) and when driving with windows closed (~70%).
- On-roadway $PM_{2.5}$ was substantially lower than the annual NAAQS of 15 μg m^{-3}.
- UFP and $PM_{2.5}$ were similar while walking, cycling, and driving with windows open.
- Respiratory UFP exposure > 7× higher for cycling and walking than driving modes.
- A model showed policy scenarios could reduce background-subtracted UFP by 83%.

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ABSTRACT

Elevated concentrations of ultrafine particles (UFPs, <0.1 μm), which have been linked to adverse health effects, are commonly found along roadways. This study reports UFP and $PM_{2.5}$ concentrations and respiratory exposures among four transportation modes on an urban residential street in Santa Monica, California while walking, cycling, and driving with windows open and windows closed (with air recirculation on). Repeated measurements were made for nine days during morning (7:30–9:30), afternoon (12:30–14:30), and evening (17:00–19:00) periods. Median UFP concentrations ranged $1–3 \times 10^4$ particles cm^{-3}, were 70% lower in afternoon or evening periods compared to the morning, and were 60% lower when driving with windows closed than open. Median $PM_{2.5}$ ranged 2–15 μg m^{-3}, well below the annual National Ambient Air Quality standard of 15 μg m^{-3}. Respiratory UFP exposure (particles inhaled $trip^{-1}$) was ~2 times higher while driving with windows open, ~15 times higher when cycling, and ~30 times higher walking, than driving with windows closed. During one evening session with perpendicular rather than parallel wind conditions, absolute UFP concentration was 80% higher, suggesting influence of off-roadway sources. Under parallel wind conditions, a parameter called emissions-weighted traffic volume, used to account for higher and lower emitting vehicles, was correlated with beach-site-subtracted UFP using second-order polynomial model ($R^2 = 0.61$). Based on this model, an 83% on-roadway UFP reduction could be achieved by (1) requiring all trucks to meet California 2007 model-year engine standards, (2) reducing light-duty vehicle flows by 25%, and (3) replacing high-emitting light-duty vehicles (pre 1978) with newer 2010 fleet-average vehicles.

1. Introduction

Ultrafine particles (UFPs, <0.1 μm) are found in elevated concentrations near roadways with motorized vehicular traffic (Morawska et al., 1999; Westerdahl et al., 2005; Zhu et al., 2002). The average employed Los Angeles County citizen in 2010 spent ~54 min commuting each day (US Census, 2010). Depending on UFP exposure while not commuting, such as occupational and lifestyle exposures, suburban or urban roadway travel could account for 17–50% of daily UFP exposure (Fruin et al., 2008; Wallace and Ott, 2011; Zhu et al., 2007). Exposure to traffic-related UFPs has been shown to be harmful to human health by toxicological and epidemiological studies (Brown et al., 2001;

Abbreviations: PNC, particle number concentration; WCPC, water-based CPC; EUs, emissions units.

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Strak et al., 2010; Vinzents et al., 2005; Wichmann and Peters, 2000).

Previous studies have quantified UFP concentrations and exposures of street users during active transportation (walking and/or cycling) and motorized transportation. A study in Montreal, Canada found 2.0 times higher particle number concentration (PNC) for motorists than pedestrians, but on separate routes for each mode (Weichenthal et al., 2008). Another similar study in London, England found the average motorist-to-pedestrian UFP ratio, which also used separate routes for the two modes, was 0.7 (Briggs et al., 2008). A study in Copenhagen compared the same urban routes for motorists and pedestrians, and found total particulate matter (PM, total mass with no upper size limit) ratio of motorists to pedestrians was ~2 (Rank et al., 2001). Int Panis et al. (2010) found that for various routes across three Netherlands cities, no differences in UFP, $PM_{2.5,}$ or PM_{10} concentrations between driver and cyclist, but reported four times higher respiratory exposure to cyclists from increased particle deposition as a result of elevated breathing rates during exercise (Int Panis et al., 2010). A roadway design study in Portland, Oregon found significantly lower UFP concentrations on the right than left side of parked cars separating motor vehicles from a cycling lane (Kendrick et al., 2011).

These previous studies established ranges of UFP and PM concentrations to which humans are exposed during transportation. They are limited, however, in that they did not control for vehicle ventilation settings that can reduce in-cabin UFP concentrations by up to 85% when recirculation modes were used (Zhu et al., 2007). They are further limited in that transportation modal UFP concentrations were compared across separate routes, or different times of the day, or without comparison of meteorological conditions as in Kendrick et al. (2011). Finally, to the best of our knowledge, no studies have compared air pollutants among transportation modes in the United States.

This study included concurrent measurements of UFP and $PM_{2.5}$ while walking, cycling, and driving. There were nine sampling days between March 22 and April 21, 2011, consisting of measurements in the morning (7:30–9:30), afternoon (12:30–14:30), and evening (17:00–19:00) along a 1-km urban residential roadway, Ocean Park Boulevard, in Santa Monica, CA. The objective of this study is to measure relative concentration and estimate respiratory exposures of UFP and $PM_{2.5}$ among transportation modes. These data can be used to assess the effects of meteorology, roadway design, and mobile source reduction programs on the concentration and respiratory exposure to traffic-related UFPs.

2. Material and methods

2.1. Site location and descriptions

The selected study site was located on Ocean Park Boulevard (Ocean Park) between Neilson Way (Neilson, west boundary) and Lincoln Avenue (Lincoln, east boundary) in Santa Monica, California (Fig. 1). The studied sector of the roadway had a complex terrain with two hills (maximum elevation difference of 20 m) in a length of 1 km. Ocean Park is an arterial residential road, with traffic volume ranging from 900 to 1200 vehicles h^{-1} during the study. The roadway included one lane in each direction, a cycle lane, and an elevated sidewalk for the complete length. The surrounding area consisted mainly of residential houses on both the north and south sides of the roadway; other than emissions from roadways adjacent to Ocean Park, there appear to be no substantial direct PM sources.

The 1-km roadway segment selected for this study enabled repeated measurements to more accurately detect pollutant differences among street users and transportation modes. The authors acknowledge, despite this key advantage, that the segment may not represent a typical street user's daily total on-roadway exposure to UFP and $PM_{2.5}$.

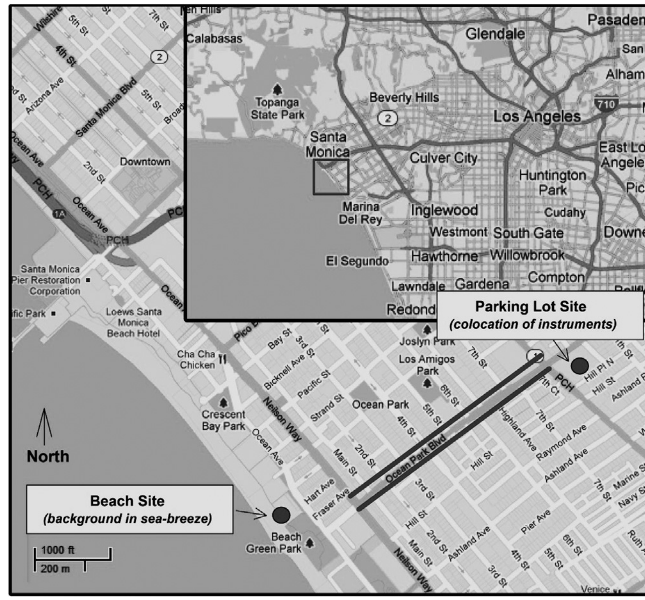

Fig. 1. Area map displaying location of 1-km study site (parallel lines), beach site, and parking lot site.

2.2. Study design

Each of the nine sampling days consisted of three sessions of two-hour continuous sampling in the following periods: morning (7:30 to 9:30), afternoon (12:30 to 14:30), and evening (17:00 to 19:00). In each session, two researchers were sampling in roundtrips along the Ocean Park Blvd (2 km total), one for walking and cycling and the other for driving and beach measurements. Each researcher was carrying one set of instruments to sample UFP, $PM_{2.5}$, and CO_2, as described in Section 2.3 Instrumentation. In each session, two walking trips (one time in each direction), four cycling trips, and six driving trips with windows open, and four driving trips with windows closed were completed. For each driving trip with windows closed, air conditioning recirculation was applied, and two consecutive round-trips were made before opening windows. Measurements while returning the vehicle to the starting location were excluded from analyses. Under typical traffic conditions, driving, cycling, and walking required ~4, ~7 and ~25 min per roundtrip, respectively. Two to three times during each session, ~5 min of measurements took place at the beach site shown in Fig. 1. During sea-breeze conditions (all afternoon and evening sessions except for Thursday evening April 14), this location served as a background monitoring site where no upwind combustion sources were present.

2.3. Instrumentation

UFP, $PM_{2.5}$, and CO_2 sampling instruments were carried in a backpack and by hand for walking modes. For cycling modes, instruments were carried in a backpack and mounted to a seat post-mounted clamp-on rack to a mountain bicycle with low tire pressure (~1.5 bar). A 2002 MY 2-door coupe was used for the driving modes with instruments placed on the passenger seat. Before and after each session, all instruments were synchronized in time, set to record data at 1-s intervals, and collocated for at least five minutes for subsequent data validation at the parking lot site shown in Fig. 1.

We measured PNC from 0.01 to 1.0 μm with a portable condensation particle counter (CPC, TSI model 3007, TSI, Inc., Shoreview, MN, USA) for walking and cycling. We measured PNC >5 nm with a water-based Condensation Particle Counter WCPC (TSI Model 3785, TSI, Inc., Shoreview, MN, USA) and PNC from 0.02 to 1.0 μm with a P-Trak (TSI model 8525, TSI Inc., Shoreview, MN, USA) for both driving and beach-site measurements. Although UFP refers explicitly to PNC for particles < 100 nm, this term will be applied hereafter in place of PNC for consistent use of terminology. Since near-roadway distributions of PNC are typically >90% within the UFP size range, this terminology is most appropriate for the context of this study.

$PM_{2.5}$ concentrations were measured by two DustTrak units (TSI model 8520, TSI Inc., Shoreview, MN, USA) for all modes. All DustTrak $PM_{2.5}$ data were divided by a factor of 2.4 corresponding to U.S. EPA Federal Reference Method gravimetric values determined by Zhang and Zhu (2010). This conversion factor is within 10% of that reported by a previous study (Yanosky et al., 2002).

CO_2 was measured using two Q-Trak units (TSI model 8554, TSI Inc., Shoreview, USA) for all modes. Elevation, latitude, longitude, speed, and bearing direction were measured by two GPS units (Qstarz model BT-Q1000XT, Qstarz Ltd., Taipei, Taiwan) for all modes. For the first three days of the study (March 22, 24, and 26), the WCPC was not used; instead the CPC 3007 instrument was used for driving and walking modes, and the P-Trak was used for cycling and beach-site measurements. The appropriate corrections were applied to account for instrumental differences, described in the Section 2.5.1.

2.4. Traffic and meteorological data

Video footage was recorded for 5 min each 15 min for the first 90 min of each session. The camera was located on the southeast corner of Ocean Park and Lincoln. Traffic entering and exiting the eastern boundary of the Ocean Park study location was counted and classified by the number of cyclists, pedestrians, and total motorized traffic. Motorized traffic was further classified into the following categories: light-duty vehicles 1978 and after, light-duty vehicles 1977 and before, solid waste collection trucks, class 5 and 6 diesel light-duty trucks, school buses, heavy-duty diesel trucks (class 7 and 8), and public city buses. These classifications were used to calculate emissions-weighted traffic volume (EUs h^{-1}) for each session as described in Section 2.6.

Meteorological data were obtained from a weather station located 7.3 km to the northeast of the study site, operated by the University of California Los Angeles, Department of Atmospheric and Oceanic Sciences. Data presented in this study reflect only those during active measurement periods.

2.5. Analyses

Before statistical analysis, data were plotted in time series and manually analyzed. Due to the nature of performing field measurements, instruments were intermittently malfunctioning (from bumps while cycling from uneven pavement) and recorded data inconsistent with on-roadway distributions. These erroneous data were removed from the dataset.

2.5.1. Instrument validation

Collocation periods, lasting five minutes before and after each sampling session, served to make measurements to relate instrumental readings of UFP, $PM_{2.5}$ and CO_2. Comparative measurements were taken at the parking lot site, shown in Fig. 1, near Lincoln and Ocean Park. All instruments were placed on passenger seat of the vehicle with the doors open for the duration of this five-minute period. The fluctuation of local sources and meteorology provided a wide range of UFP and $PM_{2.5}$ and enabled development of more accurate correlation functions between instruments for all measured ranges.

From the collocation data, all measurements were corrected to readings of instruments used for drive-mode measurements. The UFP concentration from the P-Trak was corrected to WCPC UFP concentration data using a single linear regression from all collocation periods ($\text{P-Trak}_{corrected} = 1.497{*}\text{P-Trak}_{original} + 4546$, $R^2 = 0.70$). UFP concentration from the CPC 3007 was corrected to WCPC UFP concentration data using separate regression equations for each period (average $R^2 = 0.48$). It should be noted that measurement bias has been observed not only between handheld and standard-size models (Hämeri et al., 2002), but also among standard-size instruments of the same model (Lee et al., 2013). Stronger correlations were found for $PM_{2.5}$ ($\text{DustTrak}_{corrected} = 0.91{*}\text{DustTrak}_{original} + 1.61$, $R^2 = 0.91$) and CO_2 ($\text{Q-Trak}_{corrected} = 1.45{*}\text{Q-Trak}_{original} - 137$, $R^2 = 0.66$) measurements.

2.5.2. Normality tests

UFP data were log-transformed and tested for normality using a Kolmogorov–Smirnov test for large ($n > 2000$) samples (p-value < 0.01). The results for UFP data are therefore reported as geometric mean with a geometric standard deviation, and resemble a lognormal distribution.

2.5.3. Particle respiratory exposure calculation

UFP and $PM_{2.5}$ respiratory exposures (number or μg inhaled $trip^{-1}$) are functions of arithmetic concentration (concentration $volume^{-1}$) to which the individual is exposed, the length of each trip (time $trip^{-1}$), and gender-average breathing rates available from Hinds (1999): 130, 390, and 580 mL s^{-1} for driving, walking, and cycling modes, respectively. Note that particle respiratory exposure in this study differs from average particle concentration because it includes breathing rates for the average human. Respiratory deposition is not reported because no particle size distribution measurements were included in this study. More specific classifications of breathing rates, accounting for gender, age, and physical condition are important for exposure science and epidemiological studies; however, inclusion of these parameters is beyond the scope of this work.

2.5.4. Beach-site UFP and $PM_{2.5}$

Each five-min beach-site measurement served as a reference for on-roadway UFP and $PM_{2.5}$ measurements. Instances of interference, such as cigarette smoking on the beach were noted in the field log and omitted from the dataset to maintain an accurate reference location. The term beach-site-subtracted UFP refers to on-roadway measurements after subtraction of session-resolved geometric UFP means at the beach site. The rationale for reporting beach-site-subtracted UFP is to differentiate between particles of natural background and anthropogenic (here, motor-vehicle) origin. The "incremental" beach-site-subtracted UFP values are reported throughout this paper.

2.5.5. Spatial UFP profiles

Using longitudinal data from the GPS instruments, data collected while cycling and walking were combined, but segregated by period, and assigned to a location on Ocean Park. Regions near major thoroughfare roadways (Neilson, Main, and Lincoln) were classified as "boundary zones," to ostensibly capture the effects of vehicle idling, acceleration, and pollutants originating from traffic signals. The remaining observations were grouped into 64 evenly-spaced segments of ~13 m each, approximately the width of an intersecting cross street. The influence of roadway grade, the presence of underpasses, and within-route intersections were tested using a multivariable linear regression in the form UFP = β_1*Grade + β_2*Underpass + β_3*Intersection + Intercept. Roadway grade was calculated as an absolute decimal value (e.g. 0.03 for a 3% grade) and used indicator variables for the presence (=1) or absence (=0) of the underpass beneath 4th Street and intersections. The results of this analysis are reported in Fig. 7 and discussed in Section 3.3.

2.6. Emissions-weighted traffic volume

A novel approach is used to predict pollutant reductions from changes in traffic fleet by accounting for the effects of observed higher and lower-emitting vehicles. Using this approach, 1 emission unit (EU) is defined as the emissions from one fleet average vehicle. Small and Kazimi (1995) estimated the US gasoline fleet average PM_{10} emission factor for 2000 at 10 mg mile^{-1}, but since the on-roadway UFP has decreased by approximately 50% over the past decade (Quiros et al., 2013), the 2011 fleet average is assumed ~5 mg mi^{-1} (8 mg km^{-1}). This value is reasonable considering the maximum PM emissions limit is 10 mg mi^{-1} for most new vehicles, however the exact value is less important than the relative ratios to PM emissions of higher-emitting to fleet-average vehicles. After making total traffic volume counts from recorded video footage, the video was replayed for classification by vehicle type. Literature-derived PM emission factors were applied and the number of EUs was calculated accordingly: light-duty vehicles 1977 and before (50 EUs), solid waste collection trucks (9 EUs), class 5 and 6 light-duty diesel trucks (24 EUs), school buses (26 EUs), class 7 and 8 heavy-duty diesel trucks (39 EUs), and public city buses (6 EUs) (California Air Resources Board, 2002, 2006, 2011; Small and Kazimi, 1995). Categorized video classified vehicle flows were multiplied by the respective EUs to obtain the emissions-weighted traffic volume (EUs h^{-1}). For sessions with a parallel sea breeze (230 ± 15°), emissions-weighted traffic volume was a significant predictor of beach-site-subtracted UFP for the walking mode. Regression results and rationale for methodological selection are found in results and discussion Section 3.4.

3. Results and Discussion

3.1. UFP and $PM_{2.5}$

Fig. 2(a–b) show box and whisker plots of UFP and $PM_{2.5}$ measured for each of the four modes and at the beach site during morning, afternoon, and evening periods. The boxes represent the interquartile range (IQR, 25th to 75th percentile); whiskers indicate one and a half times the IQR; and the “X’s” are the 95th percentile extent of the distribution. UFP distributions appear lognormal confirming the result of the normality test (Section 2.5.2). Both UFP and $PM_{2.5}$ were substantially elevated in the morning relative to afternoon and evening periods. This pattern is consistent with previous measurements across the Los Angeles region where higher particle concentrations were observed before the development of the daily sea-breeze under static ground-level mixing conditions (Kim et al., 2002). These concentration data are tabulated in Table 1 alongside respiratory exposure for UFP and $PM_{2.5}$.

As shown in Fig. 2(a) and Table 1, no relevant differences of geometric mean, geometric standard deviation, and 95th percentile extent were observed among cycling, walking, and driving-with-window-open modes. Cycling, walking, and driving-with-windows-open measurements were taken within three lateral meters along the roadway, and all had direct contact to the outside air. Collectively, UFP differed across period and the geometric means for these modes were 3.1, 1.2, and 1.1 × 10^4 particles cm^{-3} for morning, afternoon, and evening respectively. The corresponding

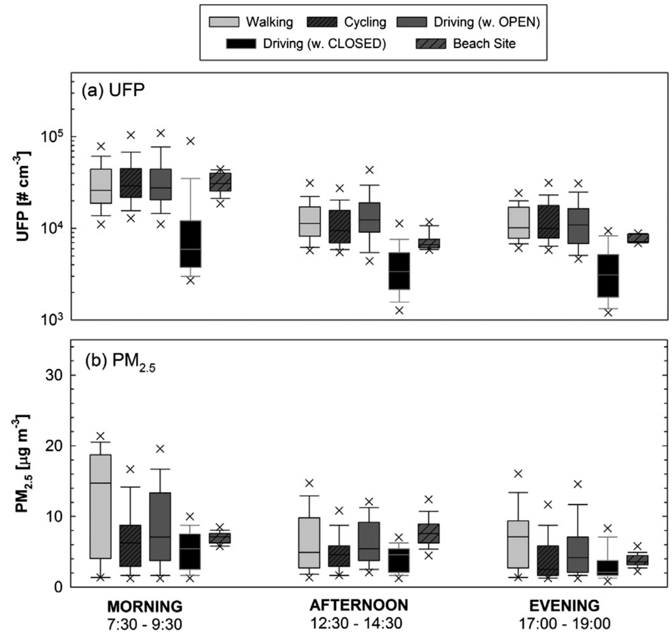

Fig. 2. Boxplots displaying distributions for (a) UFP and (b) $PM_{2.5}$ among each mode and period.

beach-site geometric means were 3.2, 0.8, and 0.9 × 10^4 particles cm^{-3} for these respective periods.

Fig. 2(a) also shows when driving with closed windows and recirculation on, a 75% UFP reduction was observed in the morning, ~40% reductions in the afternoon and evening (geometric means 7.8, 7.2, and 7.7 × 10^3 particles cm^{-3} for morning through evening, respectively). These in-cabin reductions are less substantial than the 85–90% reductions reported in previous Los Angeles vehicle cabin studies with similar vehicle configurations (Zhu et al., 2007). The difference may have been due to the chosen routes, speeds, vehicles tested, and duration of each continuous run with closed windows. Nevertheless, driving with closed windows with recirculation on appears to be an effective technique to mitigate street user exposure on Ocean Park. The build-up of in-cabin CO_2 remained a concern (Zhu et al., 2007) and concentrations > 2000 ppm were found during this study. The National Institute for Occupational Safety and Health designates 1000 ppm as an indoor air quality guideline to limit complaints related to conditions such as headache and fatigue

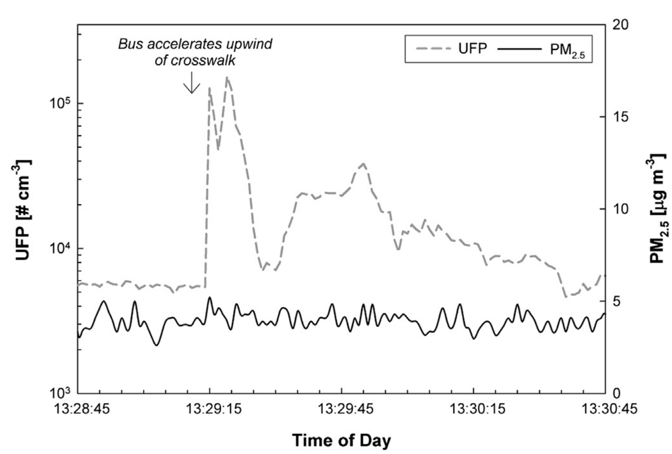

Fig. 3. Time series displaying UFP and $PM_{2.5}$ measurements downwind of an accelerating public bus on Thursday April 14 afternoon.

(NIOSH, 1987). Accordingly, the tradeoff between elevated exposures to in-cabin CO_2 versus UFPs remains a salient issue, but is beyond the scope of this paper.

Fig. 2(b) shows $PM_{2.5}$ concentration while walking had the broadest distribution in terms of IQR and 95th percentile extent of all modes. In addition, walking median $PM_{2.5}$ was higher than other modes in the morning (~15 versus ~6 μg m^{-3}) and evening (~8 versus ~4 μg m^{-3}), but not afternoon periods (all modes ~5 μg m^{-3}). When driving with windows closed and recirculation on rather than open windows, $PM_{2.5}$ decreased by ~25%. Beach-site $PM_{2.5}$ during morning and afternoon sessions was ~8 μg m^{-3}, twice the evening value of ~4 μg m^{-3}. Median and mean $PM_{2.5}$ levels for all modes attain the annual National Ambient Air Quality Standards of 15 μg m^{-3}, at the time of this study. However, measurement for regulation requires standardized methods not employed here.

The two right columns of Table 1 show particle number and $PM_{2.5}$ respiratory exposure for a roundtrip (total distance of 2 km) by period and mode. Highest particle number and $PM_{2.5}$ respiratory exposures were for the walking mode (8–21 × 10^9 particles and 7.7–15.1 μg roundtrip^{-1}), nearly twice the second-highest mode, cycling (4–11 × 10^9 particles and 3.6–7.0 μg roundtrip^{-1}), fifteen times higher than driving with windows open (4.7–14 × 10^8 particles and 0.4–0.8 μg roundtrip^{-1}), and thirty times higher than driving with windows closed and recirculation on (1.7–9.0 × 10^8 particles and 0.2–0.4 μg roundtrip^{-1}). Active transportation modes (cycling and walking) are associated with substantially elevated respiratory exposures to on-roadway pollutant levels than passive modes (driving). Street users while driving can cut in half their particulate respiratory exposure by keeping the windows closed and recirculation on. All street users can reduce particle number and $PM_{2.5}$ respiratory exposure by 25–50% by substituting a morning trip with an afternoon or evening trip.

Fig. 3 illustrates a difference between UFP and $PM_{2.5}$ from a plume of a public city bus fueled by Compressed Natural Gas. The data were measured while walking when the bus accelerated from a complete stop onto the inclined ramp from Ocean Park to the 4th Street. During this isolated incident no other immediate local traffic was present, and the bus was directly upwind (by several meters) of the instruments. There was a dramatic increase in UFP but not $PM_{2.5}$ concentration. The influence of vehicle emissions on $PM_{2.5}$ may be more regional as a result of photochemical formation pathways (Zheng et al., 2002). The influence of secondary formation of PM from mobile-source-emitted constituents is an important, but independent area. The remainder of this paper will characterize on-roadway pollutants with respect to UFP data.

3.2. Meteorology and UFP

This study included a variety of environmental conditions relative to the weather extremes in southern California. The session-average temperature and humidity, defined as the average over each two-hour measurement period, ranged between 10 and 25 °C, and 15 and 95%, respectively. Relative humidity usually was inversely related to temperature. Typically, daily temperature maxima occurred during afternoon and minima during morning and evening sessions. One exception was Thursday April 14, where the highest daily temperature was measured in the evening.

3.2.1. Wind direction and UFP

Fig. 4 is a radial plot of wind velocity for each study session. The location of the points represents the direction from which the wind originated, consistent with standard meteorological practices. The morning wind speed was low (<3 m s^{-1}) and the direction ranged between 60 and 220°. The afternoon wind speed ranged 3–7 m s^{-1} and wind direction was near-parallel ranging between 190 and

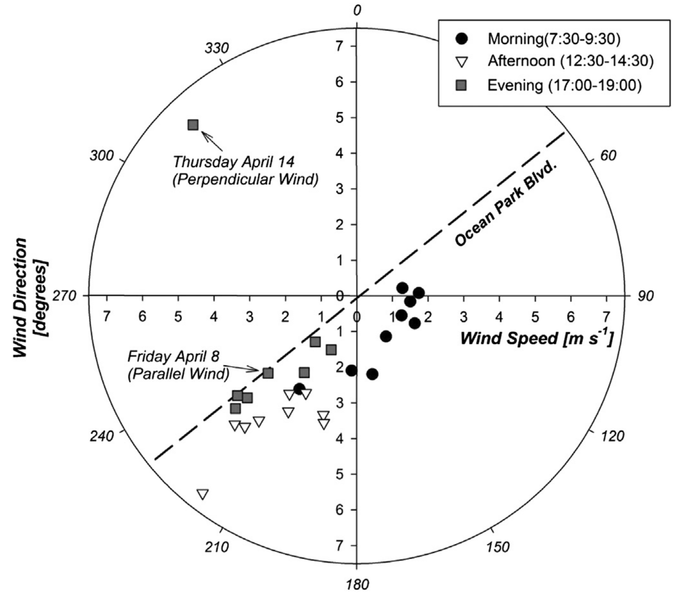

Fig. 4. Average wind velocities for each session grouped by period.

220°. During all but one evening session, wind speed ranged 1–4 m s^{-1}, and wind direction was parallel to Ocean Park ranging between 220 and 230°. On the evening of Thursday April 14, the typical southwesterly parallel wind became a strong northwesterly (~6 m s^{-1} from 315°). This likely reflects a "Santa Ana Wind" episode, characterized by a warm and dry "land breeze" formed by adiabatic compression of air moving from higher elevations to lower elevations near the coast.

Fig. 5 shows UFP versus time for (a) a typical evening on Friday April 8 with parallel wind (southwesterly) and (b) the evening of Thursday April 14 during the observed perpendicular wind (northwesterly). UFP when driving with windows closed was substantially lower than all other modes and at the beach site. Furthermore, the UFP was appreciably more constant without as many rapid fluctuations observed for modes with direct contact to on-roadway air (driving with windows open, cycling, and walking). These data suggest infiltration of on-roadway UFP is substantially diminished when driving with windows closed and recirculation on, and are consistent with results from Xu and Zhu (2009), which reported delayed and reduced infiltration of UFP into vehicle cabins.

UFP peaks under parallel southwesterly wind conditions (Fig. 5(a)) prevail longer than under perpendicular northwesterly wind conditions (Fig. 5(b)) for all modes with direct contact to outside air. A previous study found the shorter distance along the wind trajectory from the road to the receptor during perpendicular versus parallel winds contributed to the lower UFP concentrations (Molnár et al., 2002). Fig. 5(a) shows during parallel southwesterly wind, incremental values between background and on-roadway minima (defined as the low points between transient UFP spikes) are small and range 1–2 × 10^3 particles cm^{-3}. This is in contrast to the data measured during perpendicular northwesterly wind as shown in Fig. 5(b), where the same incremental values range 6–9 × 10^3 particles cm^{-3}. Furthermore beach-site-subtracted UFP and absolute UFP were 25 and 80% higher than parallel wind evenings, respectively.

3.2.2. UFP concentration frequency distributions

Fig. 6 compares frequency distributions of UFP concentrations for cycling and walking modes within each period under consistent wind velocities. Data were sorted into bins of one thousand

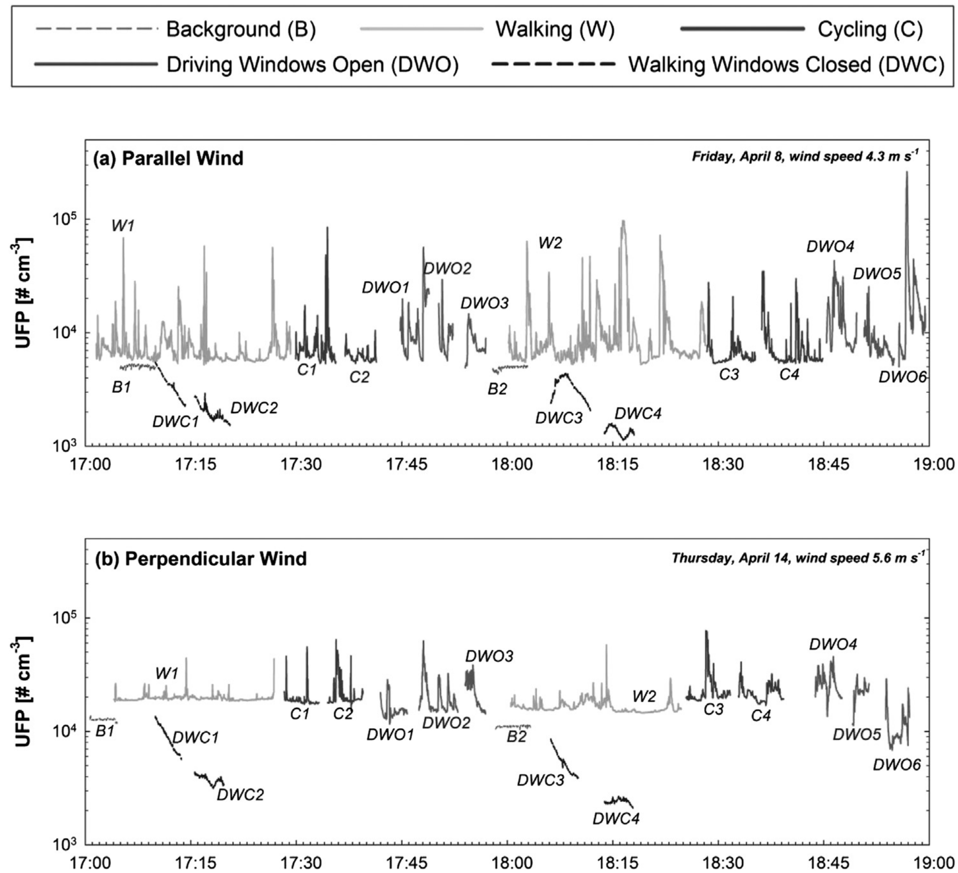

Fig. 5. UFP versus time among for an evening session with (a) a parallel wind direction on Friday April 8, and (b) perpendicular wind direction on Thursday April 14. Continuous line segments on the figure represent one round-trip. Each trip is labeled with a mode abbreviation followed by a trial number. Mode abbreviation *B* stands for beach site, *W* for walking, *C* for cycling, *DWC* for driving with windows closed (and recirculation on), and *DWO* for driving with windows open (e.g. *DWO3* represents the third run of the session for driving with windows open).

particles cm^{-3} (i.e. 2–3 $\times$ 10^3, 3–4 $\times$ 10^3 particles cm^{-3}, etc.) and are plotted on the *x*-axis as a function of beach-site-subtracted UFP. This figure compares relative distributions of sessions in order to better identify the sources of observed UFPs on Ocean Park.

The morning UFP distribution, shown by the black solid line, had the most constant frequency distribution for beach-site-subtracted-UFP bins between -5×10^3 and 1.5×10^4 particles cm^{-3}. During the breakup of the nocturnal surface inversion layer, wind speeds were low (Fig. 4), which may have subjected the beach site to inland combustion sources and roadway emissions were likely diluted less quickly. The afternoon and evening parallel-wind UFP distributions, both had beach-site-subtracted-UFP modes between -1 and 2×10^3 particles cm^{-3}. The parallel-wind evening mode was greater compared to the afternoon mode ($\sim$15% versus

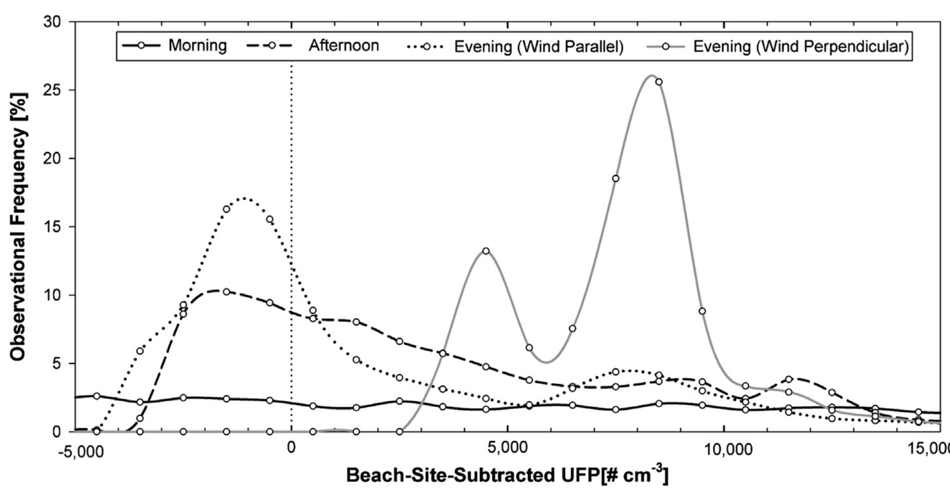

Fig. 6. UFP frequency distributions as a function of beach-site-subtracted UFP during morning, afternoon, evening (wind parallel), and evening (wind perpendicular) sessions. White circles denote the midpoint of beach-site-subtracted UFP bins of one thousand particles cm^{-3}.

10% observation frequencies, respectively). The negative mode values suggest that on-roadway minima were lower than the geometric mean of the beach-site UFP, possibly due to external influences such as cigarette smoke at the beach site not flagged in the field log. Afternoon and evening UFP frequency distributions both appear lognormal, with the majority of the positive values between 0 and 15,000 particles cm^{-3}. The morning UFP frequency distribution is also lognormal, as shown in Fig. 2, but not easily deduced from Fig. 6 because the *x*-axis only extends to 15,000 particles cm^{-3}.

The evening beach-site-subtracted-UFP distributions measured under perpendicular-wind show two distinct modes at $4–5 \times 10^3$ and $8–9 \times 10^3$ particles cm^{-3}. These peaks account for 86% of the total observed UFP frequency, with 14% of observations $>10^4$ particles cm^{-3}. The UFP time series in Fig. 5(b) shows baseline minima values are notably more frequent than transient UFP concentrations. Accordingly, the two distinct UFP frequency distribution peaks consisting of 86% UFP distributions likely correspond to off-roadway rather than transient UFP peaks from on-roadway sources.

These off-roadway sources also appear to affect beach-site UFP, which was also elevated in the perpendicular-wind evening (11,500 particles cm^{-3}) relative to the evening-study average (6100 particles cm^{-3}). Major upwind sources during this wind condition include Pico Boulevard (850 m) and the I-10 Freeway (1230 m). Previous studies of major traffic emissions sources in this region have shown little influence beyond 300 m downwind during daytime conditions (Zhu et al., 2002). Off-roadway emissions transported perpendicular to Ocean Park from the major cross street (Main) and at boundary intersections (Neilson and Lincoln) is another possibility; however Fig. 5(b) does not show patterns of UFP increases among runs (e.g. W1-W2 or C1-C4) that could reflect off-roadway contributions measured at intersections of main cross streets.

Overall, the evening with a dominant northwesterly perpendicular wind was the only instance where the UFP frequency distribution completely greater than the beach-site UFP. These results indicate perpendicular wind, depending on upwind sources, can contribute to greater beach-site-subtracted and absolute UFP. In addition, this discussion highlights the challenge and importance of identifying a suitable background monitoring location for air quality studies.

3.3. On-roadway UFP by location

Fig. 7 displays UFP as a function of location within the study site. Spatial heterogeneity was greatest during morning; the range of segment-average UFP concentration was larger in the morning (~15,000 particles cm^{-3}) than during the afternoon or evenings (~6000 particles cm^{-3}). Higher wind speeds during afternoons and evenings (Fig. 4) may have transported the pollutants more rapidly from emission locations. Alternatively, a higher proportion of recently cold-started vehicles may have been on the roadways in the morning relative to the afternoon or evening periods. Moreover, UFP peaks were measured during all three periods near the underpass at 4th Street (between 0.3 and 0.4 km) and on either side of the hill centered at Highland Avenue (~0.65 and ~0.8 km). The lowest concentrations for all periods were observed between 2nd and 4th streets (~0.2 km).

Roadway grade was significant for daily average ($\beta_1 = 134$ particles cm^{-3} grade-percent^{-1}, *p*-value = 0.007) and morning ($\beta_1 = 430$ particles cm^{-3} grade-percent^{-1}, *p*-value 0.001) datasets. These observations are consistent with previous studies that found an increase in hydrocarbon emissions with increased roadway grade (Cicero-Fernândez et al., 1997; Pierson et al., 1996). There were no statistically significant UFP concentration increases in the underpass or at intersections including at the traffic signal located at 6th Street. Boundary zones were generally lower than within-roadway UFP, except for evening period when UFP was ~3000 particles cm^{-3} higher within proximity to Lincoln. Although roadway grade predictor coefficients were significant, they don't fully explain UFP concentrations (multivariate $R^2 \approx 0.2$), and are not very practical to change for mitigating on-roadway UFP exposures.

3.4. Traffic volume and UFP

Fig. 8 shows traffic volume by period, grouped by weekends and weekdays. Fig. 8(a) shows total motorized traffic volume (including public city buses) and emissions-weighted traffic volume (Section 2.6). Fig. 8(b) shows traffic volumes for pedestrians, cyclists, and public city buses on a different *y*-axis scale. Upward gray error bars indicate one standard deviation of observed variance of five-minute observations. Downward error bars were omitted for visual clarity.

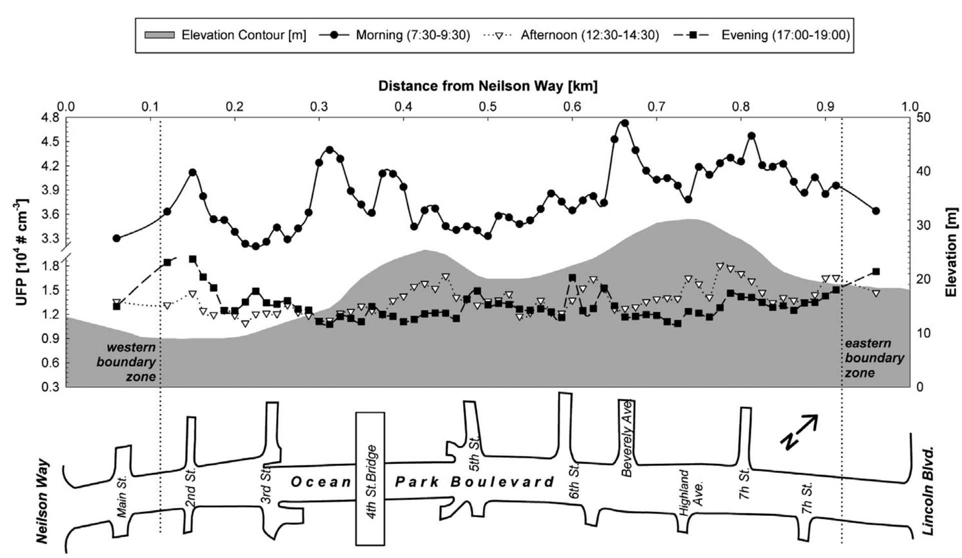

Fig. 7. UFP as a function of location on Ocean Park Boulevard. Data used were from walking and cycling modes combined, from all sessions available for each period in the legend.

Table 1
UFP and $PM_{2.5}$ concentration and respiratory exposures categorized by mode and period. The units for each category as specified under each column heading. GSD denotes geometric standard deviation; SD denotes one arithmetic standard deviation.

	UFP		$PM_{2.5}$	
	Average concentration geometric mean (GSD) [# cm^{-3}]	Respiratory exposure arithmetic mean ± SD [# inhaled $trip^{-1}$]	Average concentration arithmetic mean ± SD [μg m^{-3}]	Respiratory exposure arithmetic mean ± SD [μg inhaled $trip^{-1}$]
Walking, 25 min $trip^{-1}$				
Morning	2.84×10^4 (1.83)	$2.15 \pm 1.04 \times 10^{10}$	11.0 ± 6.6	15.1 ± 9.0
Afternoon	1.27×10^4 (1.73)	$8.60 \pm 2.72 \times 10^9$	6.58 ± 4.27	9.27 + 5.59
Evening	1.16×10^4 (1.64)	$7.83 \pm 2.60 \times 10^9$	5.57 ± 4.98	7.67 + 6.12
Cycling, 7 min $trip^{-1}$				
Morning	3.18×10^4 (1.85)	$1.08 \pm 0.54 \times 10^{10}$	10.5 ± 7.3	6.95 ± 3.79
Afternoon	1.06×10^4 (1.77)	$4.13 \pm 1.93 \times 10^9$	7.11 ± 4.31	4.56 ± 3.27
Evening	1.32×10^4 (1.98)	$4.59 \pm 2.13 \times 10^9$	5.24 ± 4.00	3.61 ± 2.64
Driving with windows OPEN, 4 min $trip^{-1}$				
Morning	3.15×10^4 (1.95)	$1.39 \pm 1.00 \times 10^9$	8.65 ± 8.00	0.74 ± 0.50
Afternoon	1.32×10^4 (1.98)	$5.98 \pm 3.34 \times 10^8$	6.47 ± 3.38	0.56 ± 0.35
Evening	1.11×10^4 (1.85)	$4.73 \pm 2.30 \times 10^8$	5.20 ± 4.28	0.44 ± 0.35
Driving with windows CLOSED, 4 min $trip^{-1}$				
Morning	7.83×10^3 (2.73)	$8.99 \pm 1.18 \times 10^8$	5.23 ± 2.96	0.44 ± 0.23
Afternoon	3.50×10^3 (1.97)	$2.18 \pm 2.04 \times 10^8$	4.15 ± 1.82	0.35 ± 0.13
Evening	3.12×10^3 (1.94)	$1.70 \pm 0.92 \times 10^8$	2.86 ± 2.29	0.25 ± 0.21

Motorized traffic was the lowest in the morning both on weekdays and weekends (~1110 and ~430 vehicles h^{-1}, respectively), peaking evenings on weekdays (~1400 vehicles h^{-1}) and afternoons on weekends (~1210 vehicles h^{-1}). Pedestrian and cyclist activity closely followed these trends; weekday and weekend flows ranged 50–60 and 20–50 pedestrians h^{-1}, and 10–20 h^{-1} and 10–30 cyclists h^{-1}, respectively. Public city bus flows were the highest in the morning at ~13 and ~6 buses h^{-1} weekdays and weekends respectively, and gradually decreased respectively by one half by the evening periods. Pedestrians, cyclists, and public city accounted for 3%, 2%, and 1%, of motorized traffic volume.

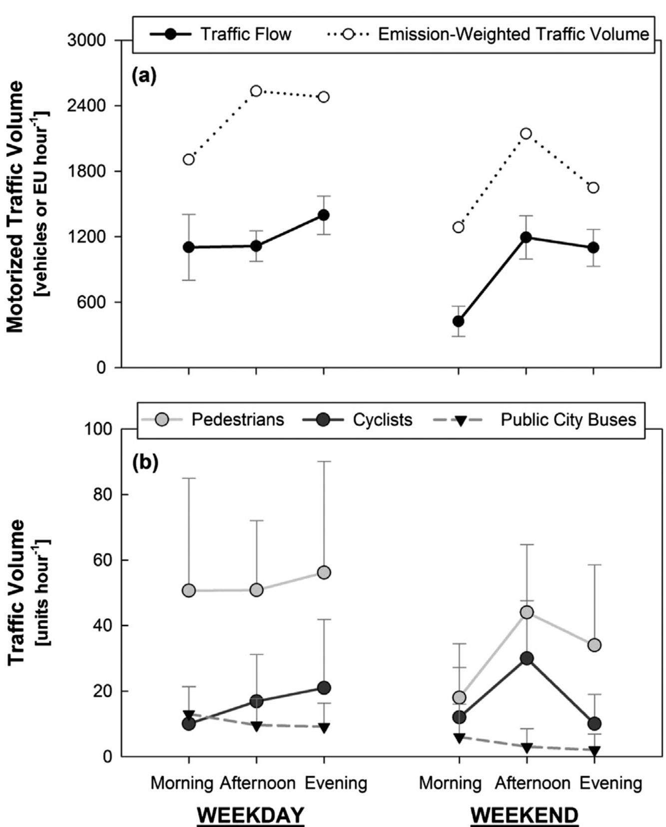

Fig. 8. (a) Total motorized traffic and emissions-weighted traffic volumes, and (b) pedestrian, cyclist, and public city bus traffic volumes, segregated by weekday versus weekend, and sampling period. Total motorized traffic includes public city buses. For measured data only (all data shown except emissions-weighted traffic volume) error bars are displayed, and show one standard deviation of observed variance of five-minute observations.

Fig. 8(a) also shows morning through evening average emissions-weighted traffic volumes were 1,910, 2,540, and 2480 EU h^{-1} on weekdays and 1,290, 2,150, and 1650 EU h^{-1} on weekend days, respectively. The ratios of emissions-weighted traffic volume to motorized traffic volume were 1.6, 2.1, and 2.1 EUs $vehicle^{-1}$ on weekdays and 2.8, 1.7, and 1.4 EUs $vehicle^{-1}$ on weekends. These ratios indicate motorized traffic fleet emissions differed quite significantly among these time periods.

3.4.1. *Emissions-weighted traffic volume*

Fig. 9 shows emissions-weighted traffic volume versus beach-site-subtracted UFP measured walking for parallel sea-breeze conditions (230 ± 15°). These criteria were met during six evening and five afternoon periods, each represented by a data point in the figure. A second-order polynomial relationship was found between beach-site-subtracted UFP while walking and emissions-weighted traffic volume ($R^2 = 0.61$). The curve is forced through the origin (point shown is not a data point) since on-roadway minima are typically equal or even less than beach-site UFP during parallel wind conditions (Fig. 6).

No relationship with beach-site-subtracted $PM_{2.5}$ was found with emissions-weighted traffic volume, consistent with the relationship presented in Fig. 3. However, gravimetric emissions factors were used to calculate emissions-weighted traffic volume for the three following reasons. First, tailpipe emission factors for particle numbers from individual vehicles have not been established. On-roadway tunnel studies (Geller et al., 2005; Kirchstetter et al., 1999) have defined number-based emission factors for broad vehicle classifications (i.e. gasoline and diesel). Moreover, these factors were estimated by fuel-efficiency generalizations and source apportionment assumptions. Second, dynamometer studies measuring tailpipe emission number vary greatly by protocol, have cycle-to-cycle variability, and are confined to laboratory settings with filtered dilution air (Dwyer et al., 2010). Third, a zero-emissions mobile platform following high-emitting light-duty and heavy-duty vehicles in the Los Angeles region found a moderate Spearman rank correlation ($r = 0.41$) between UFP number and $PM_{2.5}$ mass concentrations (Park et al., 2011).

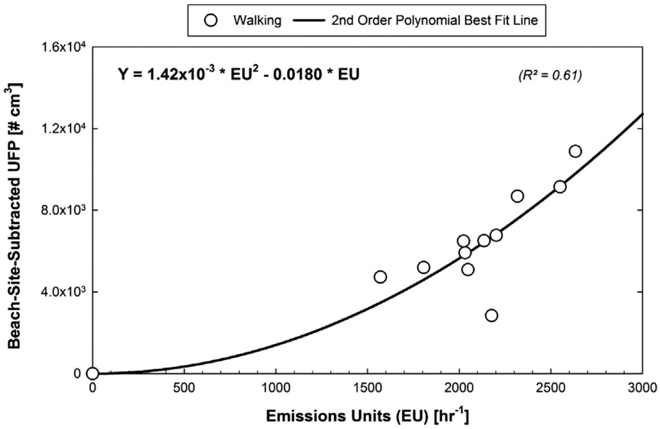

Fig. 9. Second order polynomial regression between beach-site-subtracted UFP while walking versus emissions-weighted traffic volume (EUs h^{-1}) for sessions with parallel wind.

3.4.2. Modeled emission reduction scenarios

For the *ad hoc* purpose of selecting emissions-weighted traffic volume reduction scenarios, we assume light-duty vehicles are for "personal" use (i.e. work, leisure, etc.) and have some degree of trip optionality and mode flexibility. This flexibility could mean greater use of public transit, cycling or walking, or carpooling options. This is in contrast to "service vehicles" (trucks and public buses), which are assumed more necessary to serve the fundamental role of public transportation and goods movement within the community.

If personal vehicle flow were to be reduced by 25%, where personal vehicles are all motorized light-duty vehicles (including high-emitting light-duty) but not trucks or buses, then weekday average emissions-weighted traffic volume would decrease from 2160 to 1730 EUs h^{-1}, and beach-site-subtracted UFP by 36% (CI: 11–61%). This one-quarter reduction of personal vehicles is plausible through increased ridership of public transportation, or adoption of alternative transportation modes including cycling and walking. If the remaining 75% of the high-emitting light-duty fleet were substituted with fleet average light-duty vehicles (5 mg mi^{-1}, 1 EU), weekday average emissions-weighted traffic volume would decrease further to 1230 EUs h^{-1} and beach-site subtracted UFP in total would be reduced by 68% (CI: >36%). This scenario will likely result as the older vehicles in the fleet age and newer low-emitting vehicles are registered. If we assume no change to personal vehicles, but all service vehicles (i.e. light- and heavy-duty trucks, school buses, public buses, etc.) met the California MY 2007 emission standards (10 mg brake-horsepower-h^{-1}, ~2 EUs), weekday average emissions-weighted traffic volume would decrease to 1790 EU and beach-site-subtracted UFP would decrease by 31% (CI: 9–53%). Combining both described reduction scenarios for "personal" and "service" vehicles, predicted emissions would equal 890 EUs h^{-1}, translating into a 83% decrease (CI: >53%) in beach-site-subtracted UFP.

This approach to estimate UFP reductions is novel but approximate and is limited in the following ways. First, a meaningful relationship was only found from walking data. This may be attributed to the greater number of data points in each session (Table 1), thereby sampling emissions from a greater proportion of vehicles classified by video at the intersection of Ocean Park and Lincoln. Second, an element of bias was introduced when classifying vehicles. Whereas some classifications are simple (e.g. public city buses and refuse trucks), accurate classification was not always possible among other vehicles (e.g. pre-1978 and post-1977 light-duty vehicles). Third, the relationship was only established under parallel wind conditions, hypothetically from isolation of emissions within the roadway environment, which is consistent with previous discussion of wind direction (Section 3.2.1). Finally, the regression does not have constant variance; the majority of the emissions-weighted traffic volume values fall between 1230 and 2500 EUs h^{-1}. Since the reduction scenarios calculate emissions-weighted traffic volume to values < 1230 EUs h^{-1}, the beach-site-subtracted UFP estimates should be carefully interpreted within the context of the presented 95% CIs.

4. Conclusions

This study reports UFP and $PM_{2.5}$ concentration and respiratory exposure among four transportation modes on an urban residential street in Santa Monica, CA. Median UFP concentrations ranged 1–3 × 10^4 particles cm^{-3}, were 70% lower in afternoon or evening periods compared to the morning, and were 60% lower when driving with windows closed. For each session, median $PM_{2.5}$ ranged 2–15 μg m^{-3}, which is well below the annual National Ambient Air Quality standard of 15 μg m^{-3}. UFP respiratory exposure per roundtrip, relative to driving with windows closed, was ~2 times higher while driving with windows open, ~15 times higher when cycling, and ~30 times higher walking.

During the morning period, wind speed was low with no predominant direction. These low mixing conditions may have explained the elevated pollutant concentrations reported. During afternoon and evening periods, the wind speed was higher and was predominantly southwesterly – parallel to Ocean Park. Higher UFP levels were observed on the roadway under parallel wind when vehicular emissions were isolated within the roadway environment. As a result of a Santa Ana wind episode, the wind direction for one evening session was northwesterly perpendicular, and on-roadway influences were more transient. However, likely due to off-roadway sources not on Ocean Park, there was an absolute 80% UFP increase. The only significant roadway design factor was roadway grade, but explained only ~20% of the observed UFP variability.

In order to account for emissions from higher and lower emitting vehicles, a parameter called emissions-weighted traffic volume was calculated using gravimetric PM emission factors of six classes of high-emitting vehicles. A second-order polynomial relationship was found from beach-site-subtracted UFP versus emissions-weighted traffic volume for walking mode data under parallel wind conditions ($R^2 = 0.61$). The model predicted on-roadway UFP reductions as a result of three plausible policies reducing vehicle emissions. First, a 31% reduction is predicted if all diesel trucks meet California 2007 MY PM standards. Second, a 36% reduction is predicted if one-quarter of the personal traffic volume is removed (e.g. increased usage of public transit, cycling, or walking or trip consolidation). Third, a total 68% reduction is predicted if the remaining three-quarters of the high-emitting light-duty vehicles (pre-1978) were substituted with fleet-average vehicles. With simultaneous adoption of these three plausible policies, the model predicts an 83% reduction of on-roadway UFPs.

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