Atmos. Chem. Phys. Discuss., 10, 5599–5626, 2010
www.atmos-chem-phys-discuss.net/10/5599/2010/

Atmospheric Chemistry and Physics Discussions

This discussion paper is/has been under review for the journal Atmospheric Chemistry and Physics (ACP). Please refer to the corresponding final paper in ACP if available.

Short-term variation in near-highway air pollutant gradients on a winter morning

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Received: 8 January 2010 – Accepted: 23 January 2010 – Published: 25 February 2010

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Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

Quantification of exposure to traffic-related air pollutants near highways is hampered by incomplete knowledge of the scales of temporal variation of pollutant gradients. The goal of this study was to characterize short-term temporal variation of vehicular pollutant gradients within 200–400 m of a major highway (>150 000 vehicles/d). Monitoring was done near Interstate 93 in Somerville (Massachusetts) from 06:00 to 11:00 on 16 January 2008 using a mobile monitoring platform equipped with instruments that measured ultrafine and fine particles (6–1000 nm, particle number concentration (PNC)); particle-phase (>30 nm) NO_3^-, SO_4^{2-}, and organic compounds; volatile organic compounds (VOCs); and CO_2, NO, NO_2, and O_3. We observed rapid changes in pollutant gradients due to variations in highway traffic flow rate, wind speed, and surface boundary layer height. Before sunrise and peak traffic flow rates, downwind concentrations of particles, CO_2, NO, and NO_2 were highest within 100–250 m of the highway. After sunrise pollutant levels declined sharply (e.g., PNC and NO were more than halved) and the gradients became less pronounced as wind speed increased and the surface boundary layer rose allowing mixing with cleaner air aloft. The levels of aromatic VOCs and NO_3^-, SO_4^{2-} and organic aerosols were generally low throughout the morning, and their spatial and temporal variations were less pronounced compared to PNC and NO. O_3 levels increased throughout the morning due to mixing with O_3-enriched air aloft and were generally lowest near the highway reflecting reaction with NO. There was little if any evolution in the size distribution of 6–225 nm particles with distance from the highway. These results suggest that to improve the accuracy of exposure estimates to near-highway pollutants, short-term (e.g., hourly) temporal variations in pollutant gradients must be measured to reflect changes in traffic patterns and local meteorology.

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1 Introduction

Exposure to traffic-related air pollutants near highways is associated with adverse health effects including cardiopulmonary disease, asthma and reduced lung function (Brugge et al., 2007; Brunekreef et al., 1997; Gauderman et al., 2007; Hwang et al., 2005; McConnell et al., 2006; Nicolai et al., 2003; Van Vliet et al., 1997; Venn et al., 2001). These findings have motivated research to better understand the kinds and amounts of pollutants in the near-highway environment as well as the factors governing the temporal and spatial variations in pollutant concentrations. Much attention has been focused on ultrafine particles (UFP; diameter <100 nm) because they are more toxic per unit mass than particles with larger diameters (Dockery et al., 2007; Oberdörster et al., 1995). Studies have shown that concentrations of UFP as well as other primary vehicular emissions are elevated near highways but then decrease to background within several hundred meters primarily as a result of dilution (see Table 1). The factors that impact the magnitude and extent of these gradients include traffic conditions, temperature, relative humidity, topography, wind direction and speed, atmospheric stability, and mixing height.

The effects of wind direction and speed on near-highway pollutant gradients have been demonstrated in several studies. For example, Beckerman et al. (2008) reported that UFP and NO_2 levels decreased to background within 300–500 m on the downwind side of Highway 401 in Toronto, while on the upwind side UFP and NO_2 levels decreased to background within 100–200 m of the highway. Similar upwind-downwind differences have been reported for other highways by Hitchens et al. (2000); Zhu et al. (2002a, b, 2006), and Hagler et al. (2009). Of these, the findings of Zhu et al. (2006) are particularly noteworthy. Zhu et al. showed that upwind-downwind UFP gradients near I-405 in Los Angeles were reversed following a 180-degree change in prevailing wind direction. Also, they showed that UFP levels varied inversely with wind speed: UFP levels decreased by a factor of 2.5 following a 2.4-fold increase in wind speed.

Atmospheric stability and mixing height also strongly influence pollution gradients

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near highways. Jänhall et al. (2006) showed that peak concentrations of UFP, CO, NO and NO_2 measured during the morning rush hour at a fixed site in Göteborg, Sweden, were between 2- and 6-fold higher on days with surface temperature inversions compared to days without inversions. Furthermore, they found that once the surface boundary layer lifted by late morning and surface air layers were able to mix with those aloft, traffic-related pollutant concentrations decreased to the same levels observed on days without inversions. In a year-long study of polycyclic aromatic hydrocarbons (PAH) and other traffic-related pollutants in Quito, Ecuador, Brachtl et al. (2009) found that pollutant levels increased sharply after 05:00, peaked at around 06:00–07:00, and then decreased sharply thereafter despite the relative constancy of traffic flow. This pattern was attributed to near-daily surface temperature inversions that are most pronounced just before sunrise.

Two studies from the Los Angeles Basin further demonstrate the importance of atmospheric stability. Zhu et al. (2006) found that UFP concentrations at 30 m from I-405 were only 20% lower at night (22:30–04:00) than during the day despite a 75% decrease in nighttime traffic flow. This apparent discrepancy was attributed to a combination of higher vehicle speeds, lighter winds, decreased air temperature, and increased relative humidity during the nighttime. Hu et al. (2009) found that pre-sunrise UFP levels were elevated above background as far as 600-m upwind and 2600-m downwind from I-10 despite relatively low traffic flow. Also, pre-sunrise pollutant concentrations were much higher during the winter than summer. These observations were attributed to surface temperature inversions and low wind speeds that are characteristic of the LA Basin in pre-sunrise hours. Seasonal differences in pollutant concentrations may be due in part to differences in boundary layer heights. Hu et al. reported that temperature inversions occurred at a lower elevation in the atmosphere in winter compared to summer, and that this, combined with relatively low nighttime winds in winter, resulted in shallower surface mixed layers during winter monitoring.

Knowledge of the causes of variations in the magnitude and extent of traffic-related pollutant gradients near highways has relevance for minimizing exposure assessment

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errors in health effects studies. Because pollutant levels at a given location can vary significantly throughout the day, repeated measurements must be made to properly characterize this variation. With the exception of a few recent studies (e.g., Jänhall et al., 2006; Hu et al., 2009), relatively little work has been done to measure the rates of change of near highway air pollution gradients. Therefore, the objective of our study was to measure hourly variations in near-highway air pollutant gradients. We did this using a mobile monitoring platform equipped with rapid response instruments. The pollutants studied included ultrafine and fine particles, gas-phase volatile organic compounds, CO_2, NO, NO_2, O_3, and particle-phase NO_3^-, SO_4^{2-}, and organic compounds. The study was conducted on a weekday morning in winter to capture near-highway air pollutant gradients under heavy traffic conditions during the transition from relatively light pre-sunrise winds to stronger winds following sunrise.

2 Methods

2.1 Study area

The study was conducted near Interstate 93 (I-93) in the eastern part of Somerville, Massachusetts (Fig. 1). I-93 has four north-bound and four south-bound lanes, is 40-m-wide, and carries an average of ~150 000 vehicles per day (MA Highway Dept., 2008). The highway is elevated 4.5–6 m above street level in the study area and is filled underneath except at crossing points for local roads. The northbound lane is flanked on its east side by a 3-m-high concrete sound barrier. Sound barriers are of significance as they tend to impact pollution levels near highways (Baldauf et al., 2008). Massachusetts Route 38, a four-lane highway carrying ~20 000 vehicles/day (MA Highway Dept., 2008), runs parallel to I-93 through the study area; Massachusetts Route 28, a six-lane highway that carries ~50 000 vehicles/day (MA Highway Dept., 2008), runs at a 60° angle to I-93 southeast of the study area (Fig. 1). Neither Routes 38 nor 28 are elevated above grade. The study area is bordered by the Mystic River to the north,

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Massachusetts Route 28 to the south and east, and Winter Hill (elevation=40 m) to the west. The area is relatively flat with the exception of a small hill (elevation=18 m) located near the intersection of Putnam Road and Temple Road. The study area is thickly settled with many single- and two-family houses and two- and three-story apartment buildings within 25–400 m of the highway, and other than highway and street traffic (particularly on the west side of I-93) the study area contains no other known significant sources of air pollution. Wednesday, 16 January 2008 was chosen because relatively stable weather was forecast for that day. Monitoring was done between 06:00 and 11:00, before, during, and after morning rush hour.

2.2 Data collection

Real-time measurements of particle size distribution and particle number concentration (PNC), CO_2, NO, NO_2, O_3, aromatic volatile organic compounds (AVOC; the sum of benzene, toluene, xylene isomers, ethyl benzene and C_3-benzene isomers), and particle-bound nitrate, sulfate, and organics were made using the Aerodyne Research Inc. (ARI) mobile laboratory (AML) (Kolb et al., 2004). The AML is a walk-in panel truck equipped with a suite of air-monitoring instruments, a GPS receiver, computers, a power generator, batteries, and a video camera. NO_2 was measured with a quantum cascade tunable infrared laser differential absorption spectrometer (QC-TILDAS; ARI, Billerica, MA) operating at 1606 cm^{-1}. CO_2 was measured with a non-dispersive infrared sensor (Model 6262, LI-COR, Lincoln, NB), NO was measured with a chemiluminescence analyzer (Model 42i, Thermo Fischer, Waltham, MA), O_3 was measured with an ultra-violet absorption monitor (Model 205, 2B-Tech, Boulder, CO), and AVOCs were measured with a proton-transfer mass spectrometer (PTR-MS, Ionicon Analytik, Austria). Size distributions of airborne particles (6–225 nm) were measured using a scanning mobility particle sizer (SMPS; Model 3076, TSI, Shoreview, MN), and total particle number concentration (7–1000 nm) was measured using a condensation particle counter (CPC; Model 3022A, TSI).

An Aerodyne aerosol mass spectrometer (AMS) (Jayne et al., 2000; Canagaratna

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et al., 2007) with a C-ToF spectrometer (Drewnick et al., 2005) was used to measure chemical composition of size-resolved submicron particles. The AMS measures chemically-speciated mass loadings of aerosol particles in the 50–1000 nm size range. A collection efficiency of 0.5 (Canagaratna et al., 2007 and references therein) was used in the calculation of the reported mass concentrations. Contributions from inorganic species were identified according to the method published by Allan et al. (2004). Mass spectra were analyzed using positive matrix factorization (Lanz et al., 2007; Ulbrich et al., 2009). As in other urban areas (Zhang et al., 2007), two dominant organic components – HOA (hydrocarbon-like organic aerosol) and OOA (oxidized organic aerosol) – accounted for 98% of the observed organic aerosol mass. The HOA component generally correlates well with tracers of vehicle emission (i.e., BC, CO, NO_x) indicating it has a dominant contribution from fresh vehicle emissions; the oxidized nature of the OOA component reflects photochemically-aged urban aerosol (Cangaratna et al., 2007; Zhang et al., 2007).

The chemiluminescence sensor and the PTR-MS were both calibrated by successive dilution of air from cylinders containing concentration standards as described elsewhere (Wood et al., 2008; Rogers et al., 2006), leading to uncertainties (1σ) of 10% (NO) and 25% (AVOC). The NO_2 measurements were calibrated with a similarly diluted standard (NO_2 produced via ozonolysis of 30 ppm NO in nitrogen) with uncertainties of 8%. The CO_2 sensor response was checked with a two-point calibration at 0 and 400 ppm CO_2 in air (Scott Specialty Gases), with an estimated uncertainty of 5%. The manufacturer-stated accuracy of the O_3 measurements was 2%.

The particle inlet manifold was made from stainless steel and copper tubing – to minimize particle loss due to electrostatic deposition – and equipped with a cyclone separator to remove coarse (>2.5 µm) particles. The gas inlet manifold was made from perfluoro-alkoxy Teflon™ tubing and contained a 0.45-µm Teflon filter. All instruments were operated at a frequency of 1 Hz with the exception of the SMPS and the AMS, which reported measurements every 110 and 15 s, respectively.

Continuous measurements were recorded throughout the monitoring period as the

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AML was repeatedly driven along roadways perpendicular and parallel to I-93 (Fig. 1), respectively. The AML was driven as slowly as traffic allowed (2.2–11 m/s) to capture local-scale changes in pollutant levels. The wind primarily blew from the northwest throughout the monitoring period (Fig. 2); therefore, Memorial Road, Connors Road, and Mystic Avenue (Route 38) on the west side of I-93 were selected as the upwind transects, while Shore Drive, Bailey Road, Putnam Road, and Temple Road on the east side of I-93 were used for the downwind transects (Fig. 1). Upwind pollutant levels were measured from 40 to 190 m from I-93, while downwind levels were measured from 34 to 395 m from the highway. Wind speed, wind direction and air temperature data were recorded at the Hormel Stadium light tower (height=40 m) in the city of Medford, ~0.5 km north of the study site. Hourly vehicle counts for I-93 were provided by the Massachusetts Highway Department.

2.3 Data reduction

Unusually high concentrations of AVOC – defined as values higher than the upper quartile +1.5-times the interquartile range based on box plot analysis (Devore, 2004) – were considered indicative of self-sampling or encounters with nearby vehicle exhaust plumes; therefore, they were removed from the dataset along with corresponding measurements from the other instruments. GPS data was analyzed using ArcGIS 9.2 (ESRI, Redlands, CA). Data from the SMPS was adjusted following a laboratory calibration of the instrument after sampling. Due to the frequency of data collection and the slow speed at which the AML was traveling, several measurements of each pollutant were often taken at approximately the same location during a given run. These measurements were averaged prior to data analysis. The averaged data was not significantly different than non-averaged data according to the Wilcoxon Rank Sum Test with $P < 0.05$.

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3 Results and discussion

3.1 Meteorological and traffic conditions

On 16 January 2008 it was partly cloudy from 06:00 to 09:00 and mostly clear thereafter. There was no precipitation during the monitoring period (06:00–11:00); the 24-h mean barometric pressure and relative humidity were 30.20 inches of Hg and 53%, respectively (http://www.wunderground.com, last access: 29 June 2008). The air temperature was relatively constant (−6.1 °C) between the start of monitoring and sunrise, and rose steadily thereafter to −1.7 °C at 11:00 when monitoring ended (Fig. 2a). The wind speed behaved similarly: winds were light (2–5 m/s) before sunrise, and then increased to between 5–9 m/s by 11:00. The wind direction (WNW) was relatively constant over the 5-h monitoring period (Fig. 2b). Traffic flow rate changed over the monitoring period as shown in Fig. 2c. Traffic flow rate was about 6000 (vehicles/h) at the start of monitoring at 06:00, peaked at 8800 vehicles/h at 08:00, and decreased gradually to about 7800 vehicles/h by 11:00. The fleet composition and average vehicle speed were not measured.

3.2 Spatial and temporal variation of CPC and SMPS measurements

Changes in highway traffic flow rate, wind speed, and surface boundary layer height greatly impacted pollutant gradients near I-93. Particle number concentrations (PNC) were highest early in the morning but then decreased rapidly after sunrise as the surface boundary layer lifted and wind speed increased (Fig. 3). Between 06:00 and 08:00 downwind PNC was highest (7×10^4–9×10^4 particles/cm^3) at 34 m from I-93, the nearest point at which measurements were made, but decreased ~2-fold within 100–250 m from the highway. Beyond 250 m downwind, PNC values were relatively constant at ~3×10^4 particles/cm^3. After 09:00 the highest downwind PNC values again occurred nearest to I-93, but were 2–3-fold lower (~3×10^4 particles/cm^3) compared to measurements made between 06:00 and 08:00. Upwind of I-93 the highest PNC values, mea-

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sured nearest to the highway (40 m), were about 40% lower than the highest downwind concentrations, and dropped off sharply at 60–70 m. Beyond this distance, the profiles were relatively flat, indicative of well-mixed conditions. It is likely that the PNC spikes immediately upwind of the highway were caused by traffic on Rt. 38. As was observed in the downwind profiles, PNC levels in the upwind profiles were also generally higher early in the morning (06:37) compared to later in the morning (08:15 and 09:47). Our observations are in agreement with those of Hu et al. (2009) who found that above-background UFP levels extended much further from highway I-10 in LA (both upwind and downwind) before sunrise compared to after sunrise.

We did not observe significant particle evolution with distance from the highway. The nearly linear relationship between PNC and CO_2 (Fig. 4a) suggests that PNC attenuation with distance was largely due to dilution. The slight concave curvature in the 06:27 and 07:20 data suggests some net particle formation (i.e., from nucleation and condensation into the >7-nm window of the CPC), which would be expected given the cold air temperature (−6.1−−1.7 °C), but the overall trend of these two plots is linear (R^2 was >0.91 for both datasets), indicating that if evolution was occurring it was indistinguishable from noise in the dataset. This is consistent with Zhang et al. (2004) who compared the effects of particle dynamics (i.e., condensation and evaporation) and dilution on PNC near highways in LA and found that the effects of particle dynamics were generally much lower in winter than summer.

Particles <50 nm in diameter dominated the particle size distribution measurements throughout the morning; nearly 80% of particles counted in the 6–225 nm size range were <50 nm (Fig. 4b). Between 06:00 and 09:00 there were roughly equal number concentrations of particles in the 6–25 and 25–50 nm size ranges at each distance from I-93; however, after 09:00 there was an apparent shift toward relatively more particles occurring in the smallest size range. The relative number concentrations of >50 nm particles did not substantially change either with distance from the highway – which is consistent with other near-highway studies (Zhu et al., 2006; Hu et al., 2009) – or with time.

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3.3 Spatial and temporal variation of gaseous pollutant measurements

Profiles of CO_2 and NO_x measured downwind of I-93 showed the same general spatial and temporal differences as were observed for PNC (Fig. 3). Between 06:00 and 08:00, CO_2 and NO_x were highest near I-93 and then decreased to background within 200–300 m downwind of the highway. After 09:00 near-highway concentrations were much lower compared to earlier times and the profiles were generally much flatter. Upwind of the highway, spatial and temporal variations in CO_2 and NO_x levels were generally consistent with upwind variations in UFP. O_3 levels were nearly three-fold higher (>25 ppb) after 09:00 compared to pre-sunrise levels (<10 ppb), both upwind and downwind of the highway (Fig. 5). This is evidence that mixing with air from aloft was the dominant source of O_3. Overlying air layers generally contain low levels of PNC, CO_2 and NO_x, so concentrations of these pollutants are expected to decrease at ground level after the surface boundary layer mixes. However, the opposite is true for O_3: air layers aloft generally contain elevated levels of O_3 (Trainer et al., 1987); thus, O_3 levels were expected to increase at ground level after sunrise. The relatively low O_3 levels measured near the highway – particularly in the 06:27, 07:20, and 08:07 downwind profiles – are likely attributable to reaction of O_3 with NO. NO_2 photolysis, which regenerates O_3, was too slow to overcome O_3 losses (Pinto et al., 2007). Similar O_3 concentrations and spatial trends were reported by Beckerman et al. (2008) in their study of Highway 401 in Toronto.

The downwind profiles for methanol show similar spatial and temporal variations to those exhibited by the other vehicle exhaust components (Fig. 3). Methanol is present in both the vehicle exhaust and is a major component in windshield wiper fluid (Rogers et al., 2006). These emissions rapidly mix within the ambient air and appear as a single source downwind from the vehicle. The methanol concentration was highest early in the morning 07:20 when the winds were the lightest. The presence of methanol in windshield wiper fluids makes the methanol measurements sensitive and highly variable due to the presence of vehicles in the direct vicinity of the AML. The interference

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from local traffic is evident in Fig. 3 by the large spikes in the upwind profile.

The AVOC concentrations (the sum of the signals from benzene, toluene, xylene isomers, ethyl benzene and C_3-benzene isomers) remained relatively constant throughout the morning and exhibit only weak spatial and temporal variations (Fig. 3). The high temporal response (100 ms dwell per mass) limited the precision of these measurements. Similar to the methanol, the highest downwind AVOC concentrations were observed during the low wind 07:20 time period. The AVOC concentrations are generally consistent with previous measurements made in the Boston area by Baker et al. (2008), which suggest an ambient AVOC concentration of 0.4 ppb with an enrichment of 4 ppt AVOC/ppb excess CO. Our measurements indicate that CO levels were enhanced by ~200 ppb above background (results not shown), suggesting an AVOC concentration of ~1.2 ppb, which is in good agreement with that shown in Fig. 3.

3.4 Spatial and temporal variation of particle composition measurements

Nitrate and sulfate aerosol concentrations (Fig. 6) were relatively low (<1 μg/m^3) throughout the morning, and the profiles showed little spatial variation with distance from the highway. This was expected because vehicles are not significant direct emitters of nitrate aerosol, and the mandated use of ultra low sulfur diesel fuel (maximum sulfur content 15 ppm$_w$) leads to very low emissions of aerosol sulfate. The overall decrease in the levels of sulfate aerosol throughout the morning is attributable to the increase in surface boundary layer height. The increase in nitrate levels over the same period – except for the 10:41 downwind profile – likely reflects photooxidation of background NO_x. Because the time scale for photooxidation of NO_x to nitrate is relatively long compared to downwind transport (Sander et al., 2006), it is likely that photooxidation of freshly-emitted NO_x from I-93 does not explain the increase in nitrate aerosol levels. Although the AMS is not sensitive to <30 nm particles, these particles would not contribute significantly to the measured aerosol mass concentrations, and therefore we do not believe this significantly impacts our conclusions.

The concentrations of organic aerosol were relatively low (<1.0 μg/m^3) throughout

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the morning (Fig. 6). The downwind organic aerosol profiles <100 m from the highway showed clear gradients in organic mass concentrations, particularly for the 06:27 and 07:20 profiles, similar to that observed for PNC and NO (Fig. 3). The downwind gradients observed at 06:27 and 07:20 also correlate with increased hydrocarbon-like organic aerosol (HOA) fractions, likely from fresh highway emissions. At >100 m upwind there appeared to be local contributions of organic aerosol, particularly in the 08:15 and 09:47 profiles.

3.5 Significance

The results of this study have significance for near-highway air pollution characterization and exposure assessment. The results show that pollutant levels change rapidly as a function of atmospheric mixing conditions and chemical reactions over short distances near highways. We observed that the levels of primary pollutants (UFP and NO_x) were highest under light winds during pre-sunrise hours and that following sunrise pollutant levels decreased rapidly both near the highway and downwind as the mixing height rose and wind speeds increased (Fig. 7). We also observed that the levels of reactive pollutants, such as O_3 and NO, change rapidly over short distances in the near highway zone (Fig. 5). These rapid temporal and fine-grain spatial changes in pollutant levels highlight the need for rapid response instruments housed in mobile monitoring platforms to characterize near-highway air pollution gradients.

The high variability of near-highway pollution levels – particularly UFP – poses a challenge for exposure assessment. Assignment of air pollution exposure generally involves some degree of exposure misclassification; however, for UFP this problem is expected to be elevated compared to pollutants that demonstrate less geographic variation (e.g., $PM_{2.5}$ and black carbon). This may partly explain the paucity of epidemiologic studies of UFP. Indeed, most studies of human health effects and PM have focused on $PM_{2.5}$, PM_{10}, black carbon or elemental carbon (see reviews in Hoek et al., 2009; Knol et al., 2009). These studies typically assign annual average exposure at residential addresses using measured levels at nearby fixed monitors or interpolation

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between multiple fixed monitors. For $PM_{2.5}$, which varies relatively gradually across time and space, the error in exposure assessment introduced by movement of people (e.g., from their homes to where they work or go to school), results in only a limited amount of exposure misclassification. However, for pollutants like UFP that vary more substantially, the error is expected to be larger, perhaps large enough to compromise tests of association with health if exposure assessment approaches similar to $PM_{2.5}$ studies are applied. This suggests that knowledge of short-term temporal and fine-grain spatial resolution of UFP is necessary in studies testing associations between UFP exposure and health outcomes. Further, high resolution pollutant data will need to be weighted by time-activity information in order to assign reasonably accurate exposures to individuals. Given our results here and those of others (e.g., studies cited in Table 1), geographic scaling on the order of tens of meters and time resolution on the order of hourly are needed to capture the rapid changes in near-highway pollutant levels with distance. Also, because meteorological and highway traffic conditions change on multiple time scales, it is necessary to perform monitoring throughout the day on different days and in different seasons to characterize the full range of variability and thereby allow more complete exposure assessments.

Acknowledgements. Traffic data was provided by the Massachusetts Highway Department. Funding for this work was provided by the Mystic View Task Force, Aerodyne Research, Inc. IR&D, and NIH grant ES015462.

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Table 1. Summary of near-highway air pollution monitoring studies.

Study	Location; highway	Distance from highway (m)	Time and season of measurement	Vehicles counts[a]	Pollutants measured[b]
Zhu et al., 2002a	Los Angeles; I-710	17–300 [c]	10:00–16:00; summer, fall	12 180 h^{-1}	UFP+FP (6–220 nm), BC, CO
Zhu et al., 2002b	Los Angeles; I-405	30–300 [c]	10:30–16:00; spring, summer	13 900 h^{-1}	UFP+FP (6–220 nm), BC, CO
Zhu et al., 2004	Los Angeles; I-405+I-710	30–300 (I-405); 17–300 (I-710)	10:00–16:00; winter	200–270 min^{-1} (I-405); 280–230 min^{-1} (I-710)	UFP+FP (6–220 nm), BC, CO
Zhu et al., 2006	Los Angeles; I-405	30–300 [c]	23:30–05:00; winter	20–70 min^{-1}	UFP+FP (6–220 nm), BC, CO
Hu et al., 2009	Santa Monica, CA; I-10	30–2600[c]	1–2 h before sunrise; winter, summer	715 5-min^{-1} (winter); 340 5-min^{-1} (summer)	UFP, $PM_{2.5}$, BC, CO, NO_x
Hagler et al., 2009	Raleigh, NC; I-440	20–300 [c]	00:00–23:59; summer	125 000 d^{-1}	UFP+FP (20–1000 nm), $PM_{2.5}$, PM_{10}, BC, NO_x, CO
Beckerman et al., 2008	Toronto; Highway 401	0–1000	Continuous and peak daily traffic periods; summer	~400 000 d^{-1}	UFP+FP (10–2500 nm), $PM_{2.5}$, NO_x, NO_2, O_3, BC, VOCs
Kittleson et al., 2004	Minneapolis – St. Paul, MN; I-494, Hwy 62	0, 10, 700	13:00-17:00; fall	NR	UFP + FP (3–1000 nm), NO_x, CO_2, CO
Birmili et al., 2008	Berlin; Highway A100	4, 80, 400	Continuous; summer	180 000 d^{-1}	UFP+FP (10–500 nm),

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Table 1. Continued.

Study	Location; highway	Distance from highway (m)	Time and season of measurement	Vehicles counts[a]	Pollutants measured[b]
Hitchins et al., 2000	Brisbane; Gateway Motorway+ Wynnum Rd	15–375[c]	NR	2130–3400 h^{-1}	UFP, FP, +CP (15–20 000 nm), $PM_{2.5}$
Morawska et al., 1999	Brisbane; South-east Freeway	10–210[c]	06:00–17:15; spring, summer	NR	UFP+FP (16–626 nm)
Gramotnev+ Ristovski, 2004	Brisbane; Gateway Motorway+Wynnum Rd	25–307	11:00–15:00; spring	3700–5000 h^{-1}	UFP (4–163 nm)
Shi et al., 1999	Birmingham, UK; A38, A441	2–100[c]	12:02–18:24; winter	30 000 d^{-1}	UFP, FP, +CP (10–10 000 nm)
Roorda-Knape et al., 1998	South Holland, Netherlands; A4, A12, A13, A20	15–330[c]	NR	80 000–152 000 d^{-1}	$PM_{2.5}$, PM_{10}, BC, VOCs, NO_2
Kerminen et al., 2007	Helsinki; Highway Itäväylä	9.65	00:00–12:00; winter	300–400 h^{-1} (night); ~3000 h^{-1} (morning)	UFP+FP (7–1020 nm)
Pirjola et al., 2006	Helsinki; Highway Itäväylä	0–140[c]	07:00–09:30, 15:00–18:30; winter, summer	40–70 min^{-1} (winter); 60–90 min^{-1} (summer)	UFP, FP, +CP (3–10 000 nm), NO_x, CO
This study	Somerville, MA; I-93	27–395[c]	06:00–11:00, winter	6020–8770 h^{-1}	UFP+FP (6–1000 nm), NO_x, CO_2, O_3, NO_3^-, SO_4^{2-}, organics

[a]As reported in the article cited.

[b]UFP=ultrafine particles (<100 nm); FP=fine particles (100–2500 nm); CP=coarse particles (>2500 nm); $PM_{2.5}$=mass of particles with aerodynamic diameter ≤2.5 µm; PM_{10}=mass of particles with aerodynamic diameter ≤10 µm; BC=black carbon; [c]Pollutant measurements were made along a transect(s) measured perpendicular to highway. NR=not reported.

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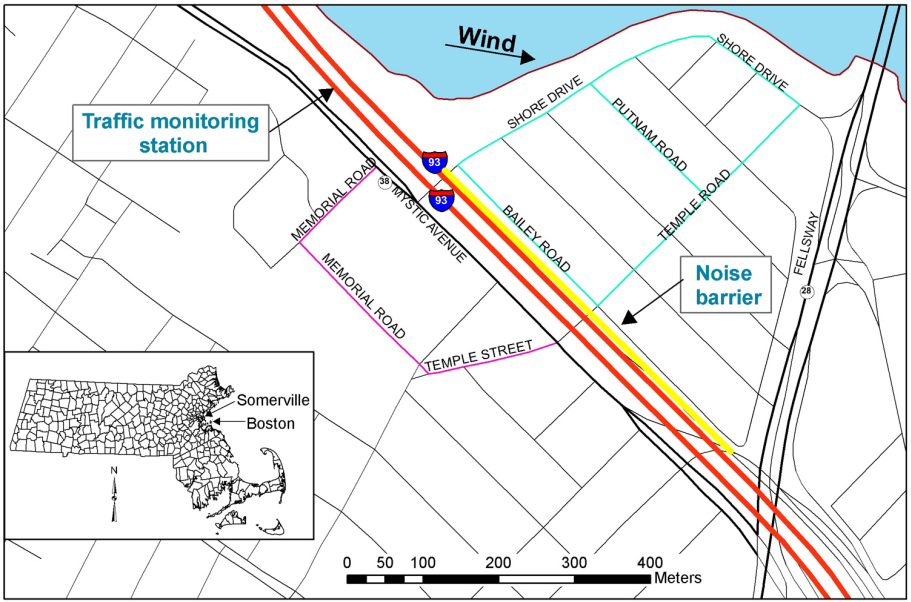

Fig. 1. Monitoring site map.

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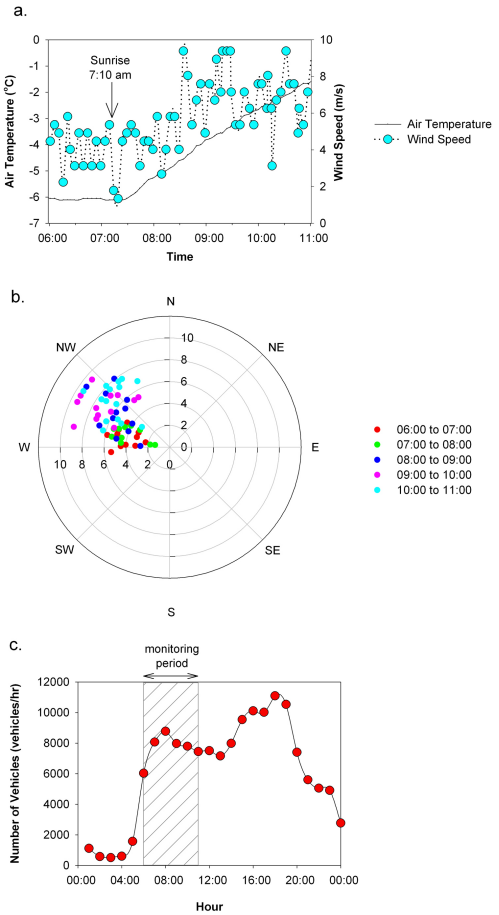

Fig. 2. Weather and traffic data from 16 January 2008. **(a)** Time-series of air temperature and wind speed; **(b)** 5-min average of wind speed and wind direction data collected between 06:00 and 11:00 at the Hormel Stadium light tower in Medford, ~0.5 km from monitoring area; **(c)** time-series of traffic flow on I-93 in both the north and south lanes measured at Somerville-Medford line.

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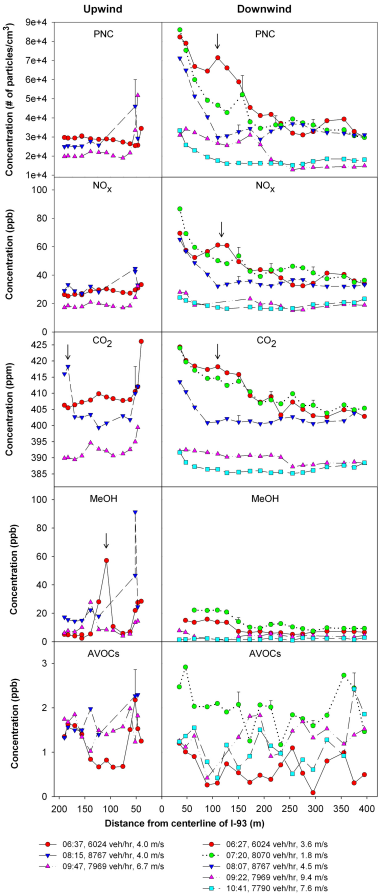

Fig. 3. Spatial and temporal variation of particle number concentration (7–1000 nm), CO_2, NO_x, and AVOC along transects perpendicular to I-93. Legend shows vehicles per hour on I-93 (both directions) and average hourly wind speed. Pollutant spikes indicated with arrows likely represent the plumes from vehicles passing nearby the AML; spikes immediately upwind of the highway were likely due to traffic on Rt. 38. Error bars (1-SD) are shown at locations where the AML stopped briefly and multiple measurements were made.

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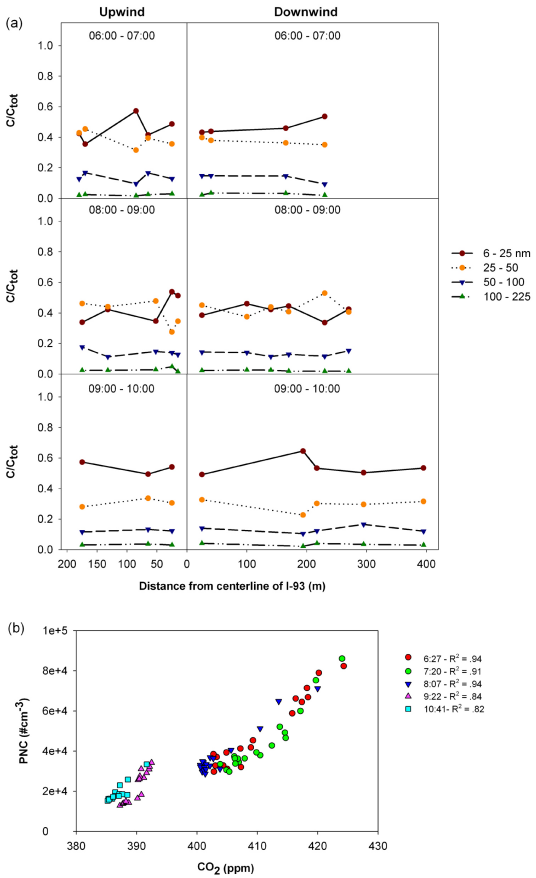

Fig. 4. (a) spatial and temporal variation of particle size distribution along upwind and downwind transects perpendicular to I-93, and **(b)** Particle number concentration (7–1000 nm) vs. CO_2 along downwind transects perpendicular to I-93.

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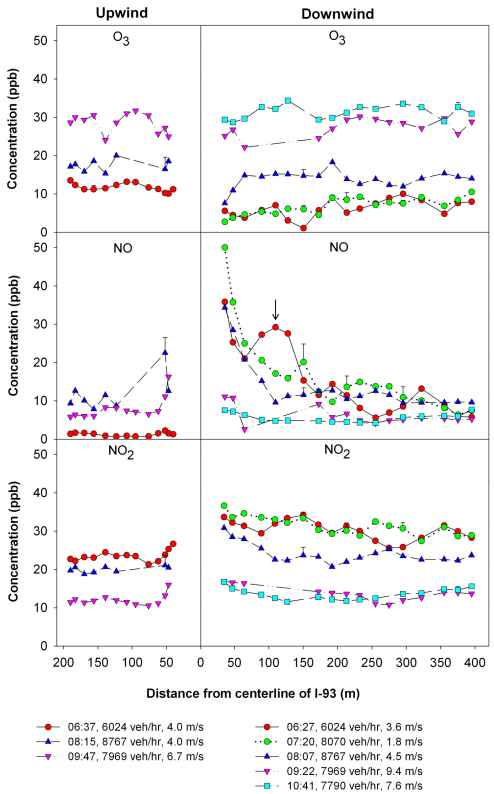

Fig. 5. Spatial and temporal variation of O_3, NO, and NO_2, along transects perpendicular to I-93. Legend shows vehicles per hour on I-93 (both directions) and average hourly wind speed. Error bars (1-SD) are shown at locations where the AML stopped briefly and multiple measurements were made. Spike in 06:27 NO profile (indicated with arrow) likely represents the plume from a vehicle passing nearby the AML; spikes immediately upwind of the highway were likely due to traffic on Rt. 38.

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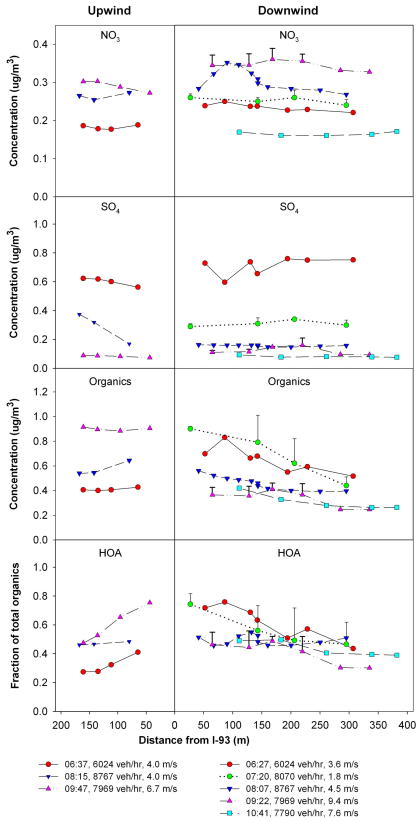

Fig. 6. Spatial and temporal variation of NO_3^-, SO_4^{2-}, organic aerosol and % hydrocarbon-like organic aerosol (HOA) along transects perpendicular to I-93. Legend shows vehicles per hour on I-93 (both directions) and average hourly wind speed. Error bars (1-SD) are shown at locations where the AML stopped briefly and multiple measurements were made. The spike in the 07:20 and 09:47 organic aerosol profiles likely represent the plumes from vehicles passing nearby the AML.

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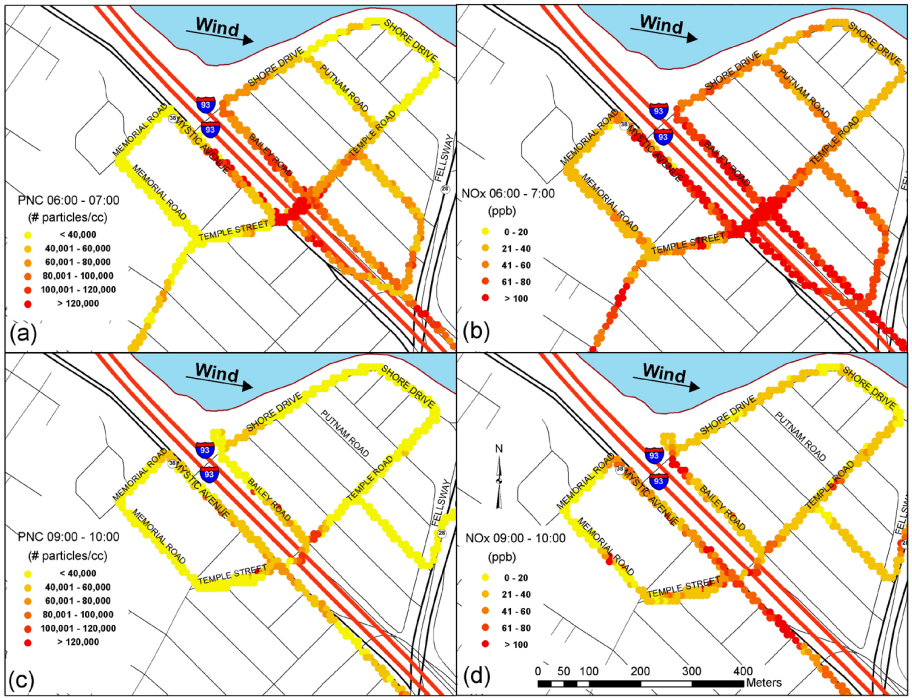

Fig. 7. Spatial distribution of particle number concentration (7–1000 nm) **(a** and **c)** and NO_x concentration **(b** and **d)** measured between 06:00–07:00 and between 09:00–10:00.

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