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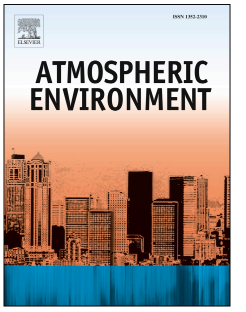

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PII: S1352-2310(17)30584-8

DOI: 10.1016/j.atmosenv.2017.09.003

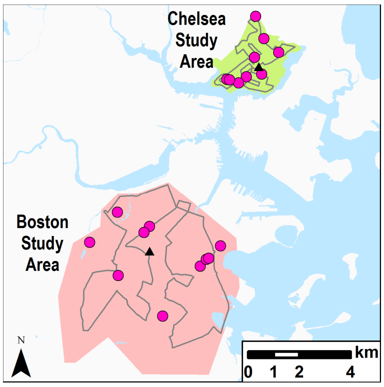
Reference: AEA 15541

To appear in: *Atmospheric Environment*

Received Date: 10 May 2017

Revised Date: 31 August 2017

Accepted Date: 1 September 2017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Comparisons of Traffic-Related Ultrafine Particle Number Concentrations Measured in Two Urban Areas by Central, Residential, and Mobile Monitoring

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Abstract

Traffic-related ultrafine particles (UFP; <100 nanometers diameter) are ubiquitous in urban air. While studies have shown that UFP are toxic, epidemiological evidence of health effects, which is needed to inform risk assessment at the population scale, is limited due to challenges of accurately estimating UFP exposures. Epidemiologic studies often use empirical models to estimate UFP exposures; however, the monitoring strategies upon which the models are based have varied between studies. Our study compares particle number concentrations (PNC; a proxy for UFP) measured by three different monitoring approaches (central-site, short-term residential-site, and mobile on-road monitoring) in two study areas in metropolitan Boston (MA, USA). Our objectives were to quantify ambient PNC differences between the three monitoring platforms, compare the temporal patterns and the spatial heterogeneity of PNC between the monitoring platforms, and identify factors that affect correlations across the platforms. We collected >12,000 hours of measurements at the central sites, 1,000 hours of measurements at each of 20 residential sites in the two study areas, and >120 hours of mobile measurements over the course of ~1 year in each study area. Our results show differences between the monitoring strategies: mean one-minute PNC on-roads were higher (64,000 and 32,000 particles/cm^3 in Boston and Chelsea, respectively) compared to central-site measurements (23,000 and 19,000 particles/cm^3) and both were higher than at residences (14,000 and 15,000 particles/cm^3). Temporal correlations and spatial heterogeneity also differed between the platforms. Temporal correlations were generally highest between central and residential sites, and lowest between central-site and on-road measurements. We observed the greatest spatial heterogeneity across monitoring platforms during the morning rush hours (06:00-09:00) and the lowest during the overnight hours (18:00-06:00). Longer averaging times (days and hours vs. minutes) increased temporal correlations (Pearson correlations were 0.69 and 0.60 vs. 0.39 in Boston; 0.71 and 0.61 vs. 0.45 in Chelsea) and reduced spatial heterogeneity (coefficients of divergence were 0.24 and 0.29 vs. 0.33 in Boston; 0.20 and 0.27 vs. 0.31 in Chelsea). Our results suggest that combining stationary and mobile

monitoring may lead to improved characterization of UFP in urban areas and thereby lead to improved exposure assignment for epidemiology studies.

Keywords: particle number concentration, ultrafine particles, mobile monitoring, stationary monitoring, residential monitoring, exposure

1. Introduction

Traffic-related air pollution (TRAP) is a complex mixture of particles and gases. Although exposure to TRAP is associated with increased morbidity and mortality (HEI Panel of the Health Effects of Traffic-Related Air Pollution, 2010; World Health Organization, 2013) there remains a lack of causal evidence to link health impacts to specific pollutants. One pollutant that may play a role in causing adverse health effects is ultrafine particles (UFP; <100 nanometers in aerodynamic diameter), which are ubiquitous in the urban environment. UFP originate mainly from combustion sources with some of the highest concentrations occurring near highways and major roadways (Karner et al., 2010; Patton et al., 2014b). UFP are of particular concern due to their small size, which allows them to penetrate deeper into the lungs, cross biological barriers, and be translocated to other organs where they can cause adverse health effects (Geiser et al., 2005; HEI Review Panel on Ultrafine Particulates, 2013; Oberdörster et al., 2005). Since the 2013 HEI report new studies have reported associations between traffic-generated UFP and markers of cardiovascular disease risk and mortality (Lane et al., 2016; Ostro et al., 2015; Viehmann et al., 2015).

UFP concentrations can vary significantly over short time and distance scales (Karner et al., 2010; Levy et al., 2014; Riley et al., 2014). For example, Pattinson et al. (2014) observed that UFP increased >2-fold at a near-roadway site within a three-hour window after the start of the morning rush hour but concurrent concentrations were ~40% lower at a site 130 m downwind from the road. The considerable fine spatial-scale and temporal variability of UFP poses a challenge for exposure assessment; therefore, care must be taken in designing UFP monitoring networks in order to adequately capture the variation and minimize exposure error (HEI Review Panel on Ultrafine Particulates, 2013; Pekkanen and Kulmala, 2004).

In epidemiological studies of UFP, models based on local meteorology and traffic conditions have been developed to estimate UFP concentrations across urban areas (Aguilera et al., 2016; Lane et al., 2016). Widely-differing monitoring networks have been used to model UFP, and characterize UFP in general, including long-term stationary monitoring (Aalto et al., 2005; Cyrys et al., 2008; Moore et al., 2009), mobile monitoring (Aggarwal et al., 2012; Li et al., 2013; Padró-Martínez et al., 2012; Patton et al., 2015; Steffens et al., 2017; Weichenthal et al., 2016; Zwack et al., 2011), monitoring at central sites and multiple short-term stationary sites (Abernethy et al., 2013; Eeftens et al., 2015; Fuller et al., 2012; Hofman et al., 2016; Klompmaker et al., 2015; Meier et al., 2015; Puustinen et al., 2007; Rivera et al., 2012; Wolf et al., 2017), or a combination of mobile and stationary monitoring (Hankey and Marshall, 2015; Kerckhoffs et al., 2016; Riley et al., 2016; Sabaliauskas et al., 2015) (Table S1). While Kerckhoffs et al. (2016) observed modest correlations between on-road and nearby short-term stationary-site PNC, it remains unclear if these results can be generalized to other study areas and other platform comparisons or if use of a particular platform measures systematically different concentrations. Knowledge of the similarities and differences between monitoring platforms and the predominant factors that drive temporal and spatial heterogeneity could improve monitoring-network designs, and thereby reduce exposure error in epidemiological studies of UFP.

In this study, we examined ambient particle number concentration (PNC; a proxy for UFP) from three different monitoring platforms – centrally-located sites, multiple short-term residential sites, and a mobile air-monitoring laboratory – in two study areas within the Boston, MA (USA), metropolitan region. Our objectives were to (1) quantify measurement differences from one monitoring platform to another, (2) estimate the consistency of temporal patterns and the heterogeneity of PNC across monitoring platforms, and (3) identify the factors that affect PNC correlations in both study areas. This effort was undertaken as a step toward assigning exposure to participants in the Boston Puerto Rican Health Study (BPRHS) cohort which is examining associations with cardiovascular health outcomes (Tucker et al., 2010).

2. Materials and Methods

2.1 Study Areas

PNC monitoring was conducted in Boston and Chelsea, the cities in which the BPRHS cohort is primarily located (Fig. 1). The Boston study area was 40 km^2 of which 40% is classified as residential (total study-area population: 318,000), while 13% and 4% are classified as commercial and industrial, respectively (MassGIS, 2005). The two largest roadways in Boston, Interstate Highways 90 (I-90) and 93 (I-93), transect the outer northern and eastern edges of the study area, respectively; average weekday daily traffic on these highways in 2010 was 110,000 and 195,000 vehicles/day (vpd), respectively (Boston Region Metropolitan Planning Organization, Central Transportation Planning Staff, 2011).

The Chelsea study area was 6 km^2. Approximately 27% of the land in Chelsea is classified as residential (total study-area population: 36,000), 12% as commercial, and 11% as industrial (MassGIS, 2005). U.S. Route 1 (US-1; 83,000 vpd) (Boston Region Metropolitan Planning Organization, Central Transportation Planning Staff, 2011) transects the city north to south; Massachusetts Route 16 (MA-16; 40,000 vpd) (Boston Region Metropolitan Planning Organization, Central Transportation Planning Staff) runs west to east along the northern outskirts of the study area. Heavy-duty diesel trucks and ocean-going ships are common in the southern parts of Chelsea where storage and distribution facilities are located on the Mystic River and Chelsea Creek. Also, Boston Logan International Airport, the busiest airport in New England (~1,000 flight operations/day), is 4.5 km southeast of the geographic center of the Chelsea study area and 7.5 km northeast of the geographic center of the Boston study area.

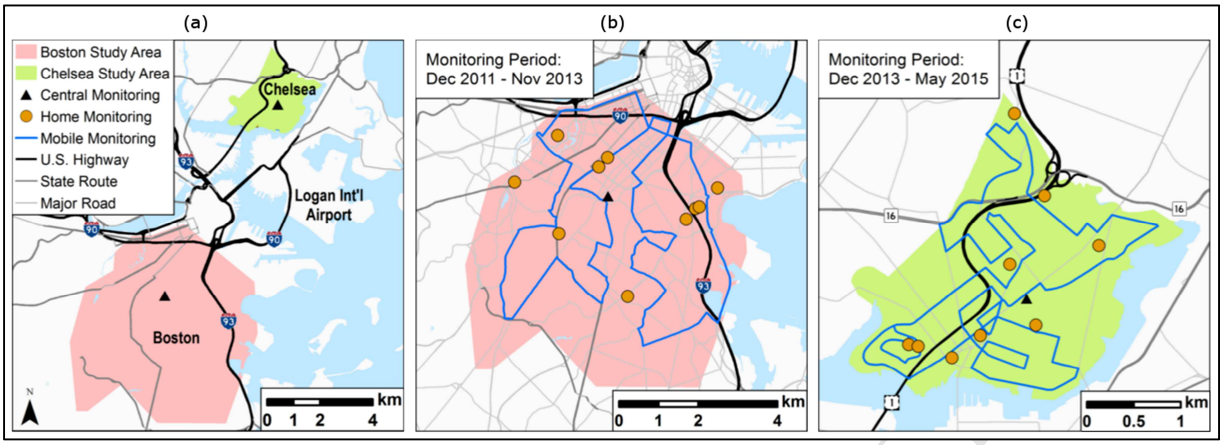


Figure 1: (a) Location of the Boston and Chelsea study areas. (b) Boston study area; central site, 11 residences, and mobile monitoring route are shown. (c) Chelsea study area; central site, 9 residences, and mobile monitoring route are shown.

2.2 Monitoring Network

Ambient PNC measurements were collected in each study area at centrally-located stationary sites, residential stationary sites, and on roads with a mobile laboratory that was driven along fixed routes. In the Boston study area, the central site was collocated at the U.S. Environmental Protection Agency Speciation Trends Network site (EPA-STN, ID: 25-025-0042), which was 1 km from the geographic center of the study area. Monitoring was performed there from December 2011 to November 2013. Residential monitoring was conducted at 11 homes of BPRHS participants (0.28 sites/km^2 of the study area) for six weeks each between May 2012 and November 2013. Residential sites were selected based on their proximity to highways and major roads (the latter defined as >20,000 vpd): three sites were <100 m, four between 100-200 m, and four >200 m from highways or major roads (Table S2). Mobile monitoring was conducted along a 40-km route in the study area (Fig. 1b) between December 2011 and November 2013 on 42 days representing all four seasons, all days of the week, and most times of day (Fig. S1). The 11 residential sites were 15-1,100 m from the mobile-monitoring route.

The central site in Chelsea was located on the third-story roof of The Neighborhood Developers building (6 Garrish Road) near the geographic center of the city. Monitoring was conducted there from January 2014 to May 2015. Residential monitoring was conducted at 9 homes of BPRHS participants (1.5 sites/km^2 of the study area) for six weeks each between February and December 2014. One site was <100 m, six between 100-200 m, and three >200 m from highways or major roads (Table S2). Mobile monitoring was conducted along a 20-km route in the study area (Fig. 1c) between December 2013 and May 2015 on 46 days representing all four seasons, all days of the week, and most times of day (Fig. S2). All 9 residential sites were 5-150 m from the mobile-monitoring route.

2.3 Instruments

Water-based condensation particle counters (CPC; TSI, Model 3873; 7-3,000 nm) were used to measure ambient PNC at the central and residential sites. The central-site CPCs were housed in locked, weatherproof, and temperature- and humidity-controlled boxes. Conductive silicon tubing (50 cm) was used to draw air from outside the box to the CPC inlet. Mean PNC measurements were recorded every 30 s (except at the Boston central and residential sites prior to May 2013 when mean PNC was recorded every minute). During weekly site visits, the CPCs underwent routine maintenance as needed (i.e., wick changes, flow checks), data were downloaded, and the instrument time was reset as necessary (CPC time drifted <1 min per week) to the National Institute of Standards and Technology official time (time.gov).

Residential monitoring was conducted at homes of BPRHS cohort participants continuously for six consecutive weeks, with up to two homes in the same study area undergoing monitoring concurrently. We monitored both outdoor and indoor air at the residential sites via two separate conductive inlet lines of equal length (100 cm; one outdoors and one indoors; CPCs were positioned indoors) that were connected to a solenoid valve that switched between the two every 15 min (indoor results are not presented in this manuscript). Residential sites were visited weekly to conduct routine equipment maintenance, download data, and reset instrument clocks.

Mobile monitoring was performed with the Tufts Air Pollution Monitoring Laboratory (TAPL), which has been described in detail elsewhere (Padró-Martínez et al., 2012). Briefly, the TAPL is a gasoline-powered Class-C recreational vehicle (2002) that contained a butanol-based CPC (TSI, Model 3775; 4-3,000 nm). The CPC measured PNC at one-second intervals to capture the rapid changes in on-road concentrations. The CPC inlet was mounted on the roof at the front of the vehicle, 9 m upwind from the exhaust tailpipe. Each monitoring session lasted 3-6 hours between 05:00 and 21:00. Due to the large size of the Boston study area, monitoring was randomly assigned to commence at the beginning or middle of the route at the start of each monitoring session. A single loop along the Boston route took 1.5-3 hours, while a single loop along the Chelsea route took approximately one hour. A GPS receiver (Garmin eTrex) recorded latitude and longitude every second.

2.4 Data Quality Assurance and Processing

Data were reviewed for very low concentrations (<500 particles/cm^3) and measurements automatically flagged by the instrument (e.g., due to nozzle clogs and low pulse heights). Data marked with these flags and/or concentrations <500 particles/cm^3 were removed (<1% of the data). We did not correct for particle losses in the sampling lines; the sampling lines were relatively short and losses have been observed to be small for exhaust particles >20 nm diameter (especially for short sampling lines) (Kumar et al., 2008). Data from monitoring at the residential sites required additional processing to minimize the possibility of mixed indoor and outdoor air downstream of the solenoid valve (7-13%), i.e., we removed at least the first 60 s of data each time the solenoid switched between outdoor and indoor air and vice versa. At two residential sites (Home 3 in Boston and Home 15 in Chelsea), mixing of indoor and outdoor air could not be ruled out completely; however, rather than removing these residential sites from the analyses we conducted a sensitivity analysis both with and without these sites. PNC measurements from the TAPL were adjusted for a three-second lag (travel time in the sample tubing between the inlet and the CPC). To minimize bias in the on-road data set due to self-sampling of TAPL exhaust, data were removed when speeds were <5 km/h for >10 s (which typically occurred at intersections). Data were removed for an additional 10 s after the TAPL's speed increased above 5 km/h to ensure that exhaust was flushed from the sampling line (15-30% of data removed, mostly during times when the TAPL was idling at traffic lights). Additionally, we inspected the data set for potential outliers by checking if any data point increased more than a factor of 10 from the preceding data point (no outliers were identified). We also examined on-road data for impacts due to emissions from nearby vehicles that resulted in PNC spikes. Spikes were identified as one-second on-road measurements more than two standard deviations above the daily mean on-road PNC (Patton et al., 2014a). Using this definition, 3.4% of data in the Boston data set and 2.5% of data in the Chelsea data set were identified as spikes. Table 1 summarizes the different monitoring-platform comparisons and the amount of data used in the statistical analyses.

Platform Comparison	**Averaging Period**	**Median Number of Data Points Used to Generate Statistics (range of n)**	
		Boston	*Chelsea*
Central-Site to Homes[a]	1 minute	21,872 (5,291-29,388)	26,542 (19,762-31,876)
Central-Site to Homes	1 hour	753 (221-1,074)	919 (778-1,006)
Central-Site to Homes	1 day	30 (8-44)	37 (31-42)
Central-Site to On-Road[b]	1 minute	47 (30-98)	187 (72-610)
Homes to On-Road[c]	1 minute	45	247

[a] Central-site to home PNC comparisons were grouped by individual home. [b] Central-site to on-road PNC comparisons were grouped by 200-m grid cells. [c] Homes to on-road PNC comparisons were pooled into single data sets, one for each study area.

Table 1: Summary of monitoring-platform comparisons.

Water- and butanol-based CPCs were collocated in the laboratory for side-by-side analysis (i.e., using one-second mean PNC over several hours with background and elevated PNC using a candle). Water-based CPCs measured PNC to within ±10% of one another, consistent with manufacturer-stated error. Comparisons between the butanol-based CPC and water-based CPCs showed good agreement (r^2 = 0.94), but the butanol-based CPC consistently measured 14% higher PNC across the entire concentration range tested due to its lower cutpoint (d_{50} = 4 nm compared to 7 nm for the water-based CPCs). To account for this difference, PNC measurements from the butanol-based CPC were adjusted downward by 14%. Temperature, humidity, wind speed and wind direction data were acquired at one-minute time resolution from the National Weather Service station at Boston Logan International Airport (KBOS) (NOAA National Centers for Environmental Information).

2.5 Statistical Analyses

Boxplots and heat maps were used to assess the temporal patterns of PNC measured by the three monitoring platforms. Temporal PNC trends were investigated by plotting data by month and year, hour of the day, and wind speed and direction. Additionally, we examined the differences between weekdays and weekends as well as between rush hours (i.e., 06:00-09:00 and 15:00-18:00) and other hours (i.e., 09:00-15:00 and 18:00-06:00). We also used mapping tools to investigate spatial changes in PNC. To visualize differences between two platforms, we used Bland-Altman plots to determine whether mean differences in PNC measurements between different platforms significantly deviated from zero across the entire measurement spectrum (Martin Bland and Altman, 1986). The calculated differences between the three monitoring strategies were to quantify general heterogeneity and potential systematic shifts between the platforms due to factors such as the location of the monitors relative to sources or the composition and volume of traffic on nearby streets, as opposed to errors in the measurements themselves.

To compare PNC measurements from the different platforms (i.e., central to residential sites, central sites to on-road, and residential sites to on-road), Pearson linear correlation coefficients (r) and coefficients of divergence (COD) were calculated (Moore et al., 2009; Wongphatarakul et al., 1998). Pearson correlations were used to explore the consistency in the temporal patterns between the different platforms while COD values were used to explore spatial variability. COD is defined by Eq. (1):

$$\mathrm{COD}_{jk} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left(\frac{x_{ij}-x_{ik}}{x_{ij}+x_{ik}}\right)^2} \tag{1}$$

where x_i is the i^{th} PNC observation at either site j or k, and n is the number of observations. COD values range from 0 to 1, with 0 denoting identical measurements and 1 denoting completely heterogeneous measurements; a value of 0.2 was used to distinguish homogeneous (COD <0.2) from heterogeneous

(COD >0.2) data sets consistent with previous studies (Moore et al., 2009; Wilson et al., 2005). To examine the possible effect of outliers on the Pearson correlation coefficients (i.e., additive error driven by local sources near the different monitors), we also calculated Pearson correlations on log-transformed PNC and Spearman correlations on non-transformed PNC for each of the platform comparisons. Pearson correlations, COD values, and Bland-Altman plots were used to understand how the three monitoring platforms compared to each other: Pearson correlations to measure the synchronicity in temporal trends, COD values to determine spatial heterogeneity, and Bland-Altman plots to visualize systematic differences in measurements. Only concurrent data were used for comparisons across platforms (i.e., paired one-minute, hourly, or daily PNC depending on the time-averaging comparison being made). Comparisons were made to both on-road measurements and an on-road data set from which spikes were excluded.

For central-site-to-residential-site comparisons, mean concentrations over one minute, one hour, and one day were calculated for central and residential sites and paired by timestamp if data coverage per averaging period exceeded 50%. For the comparisons between central-site and on-road monitoring, one-minute mean central-site data was compared to one-minute mean as well as median on-road PNC within 200-m grid cells that were constructed across the study areas. If at least 10 s of on-road data were available per minute per grid cell, then one-minute means and medians were calculated for on-road data and paired to the central site data by timestamp. Furthermore, only grid cells with >30 paired data points were used in the analyses (i.e., the mobile laboratory was in the grid cell for >10 s on at least 30 separate loops of the mobile monitoring route). Lastly, for comparisons between residential and on-road PNC, 500-m buffers were constructed around the homes, and for on-road data within each buffer one-minute means and medians were calculated and paired to the residential-site data by timestamp. R (version 3.3), MATLAB (version 8.0), and ArcGIS Desktop (Release 10.4) were used for all analyses and the generation of figures.

3. Results & Discussion

3.1 Temporal and Spatial PNC Trends

In the Boston study area, PNC was highest during winter (December-February) and lowest during summer (June-August) with median winter concentrations up to a factor of two higher than median summer concentrations (Fig. 2a). The seasonal differences were consistent across the three monitoring platforms (Table 2). PNC was also higher during weekday morning and evening rush hour periods (Fig. 2b), particularly during west-to-northwest and to a lesser extent northeast winds (17% and 7% of the study period, respectively; Fig. 2c and S3a), but this pattern was generally absent on weekends (Fig. S3b). All three monitoring platforms observed the same general trends. PNC was substantially lower during overnight hours on all days of the week and across all wind directions compared to daytime hours (Table 2). On-road PNC near I-90 and I-93 were elevated relative to other road segments in all seasons (Fig. S4); median PNC within 300 m was 29,000 particles/cm^3 versus 23,000 particles/cm^3 throughout the rest of the study area. Similarly, PNC was also elevated on other highly-trafficked roads. Our findings of seasonal and diurnal differences in PNC were consistent with other studies (Aalto et al., 2005; Cyrys et al., 2008; Meier et al., 2015; Sabaliauskas et al., 2015; Wang et al., 2011), including those from metropolitan Boston (Fuller et al., 2012; Padró-Martínez et al., 2012; Patton et al., 2014b).

Period	Median 1-min PNC in Boston (particles/cm^3)			Median 1-min PNC in Chelsea (particles/cm^3)		
	Central Site	*On Road*[c]	*Residential*	*Central Site*	*On Road*[c]	*Residential*
Winter[a]	28,000	33,000	21,000	20,000	26,000	16,000
Summer[a]	14,000	18,000	8,500	11,000	14,000	9,100
Overnight[b]	16,000	n/a	9,500	13,000	n/a	10,000
Daytime[b]	21,000		12,000	15,000		12,000
All Data	18,000	27,000	11,000	14,000	18,000	11,000

[a] Dec., Jan., and Feb. represent winter months; Jun., Jul., and Aug. represent summer months. [b] 18:00-06:00 represent overnight hours; 06:00-18:00 represent daytime hours. [c] On-road data was largely from the daytime, thus no comparison was made to overnight hours (n/a = not applicable).

Table 2: Summary of median one-minute PNC by monitoring platform.

Temporal trends in the Chelsea study area were similar to Boston. PNC was highest during winter and lowest during summer (Table 2 and Fig. 3a) across all monitoring platforms. Overnight PNC was substantially lower compared to daytime concentrations (Table 2). As in Boston, PNC was higher during weekday mornings (Fig. 3b and Fig. S3c) irrespective of wind direction; an increase in PNC was observed during the evening rush hour period, but especially during south-southeast (SSE) winds (6% of the study period; Fig. S3c). Weekend trends were largely absent in Chelsea except for elevated PNC during SSE winds (Fig. S3d; average PNC was approximately twice the average for all other wind directions). This is likely due to aviation-related emissions from Logan Airport, which is ~4 km southeast of the stationary monitor (Hudda et al., 2016). Higher PNC was observed along the US-1 and MA-16 corridors, while concentrations were generally lower in residential areas with less traffic (Fig. S5). Tables S3-S5 in the Supporting Information summarize the data obtained from all three monitoring platforms from Boston and Chelsea.

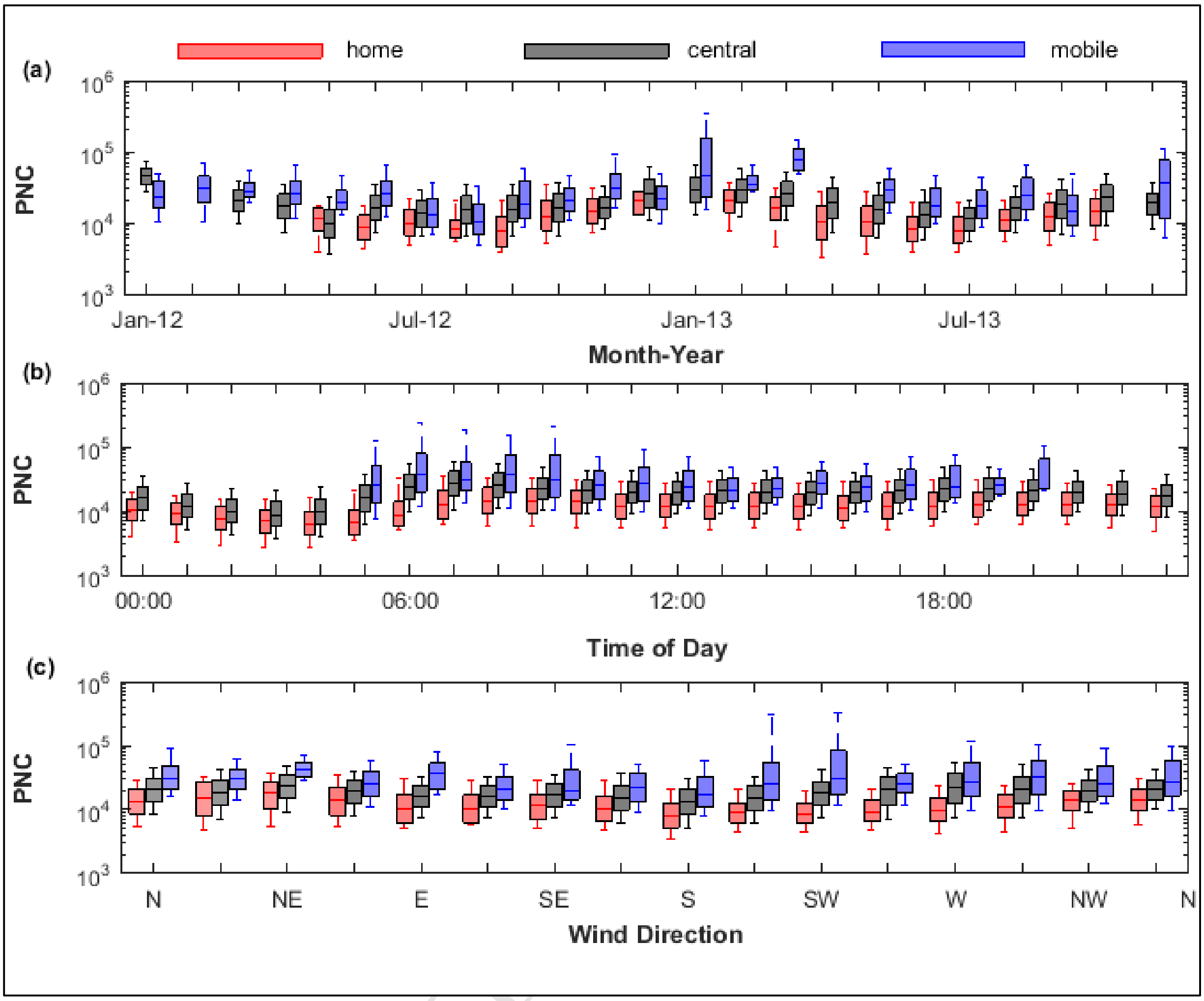


Figure 2: Boxplots of PNC by (a) month, (b) time of day, and (c) wind direction measured at central sites (black), homes (blue), and with a mobile laboratory (red) in Boston. Mobile monitoring occurred between 05:00 and 21:00.

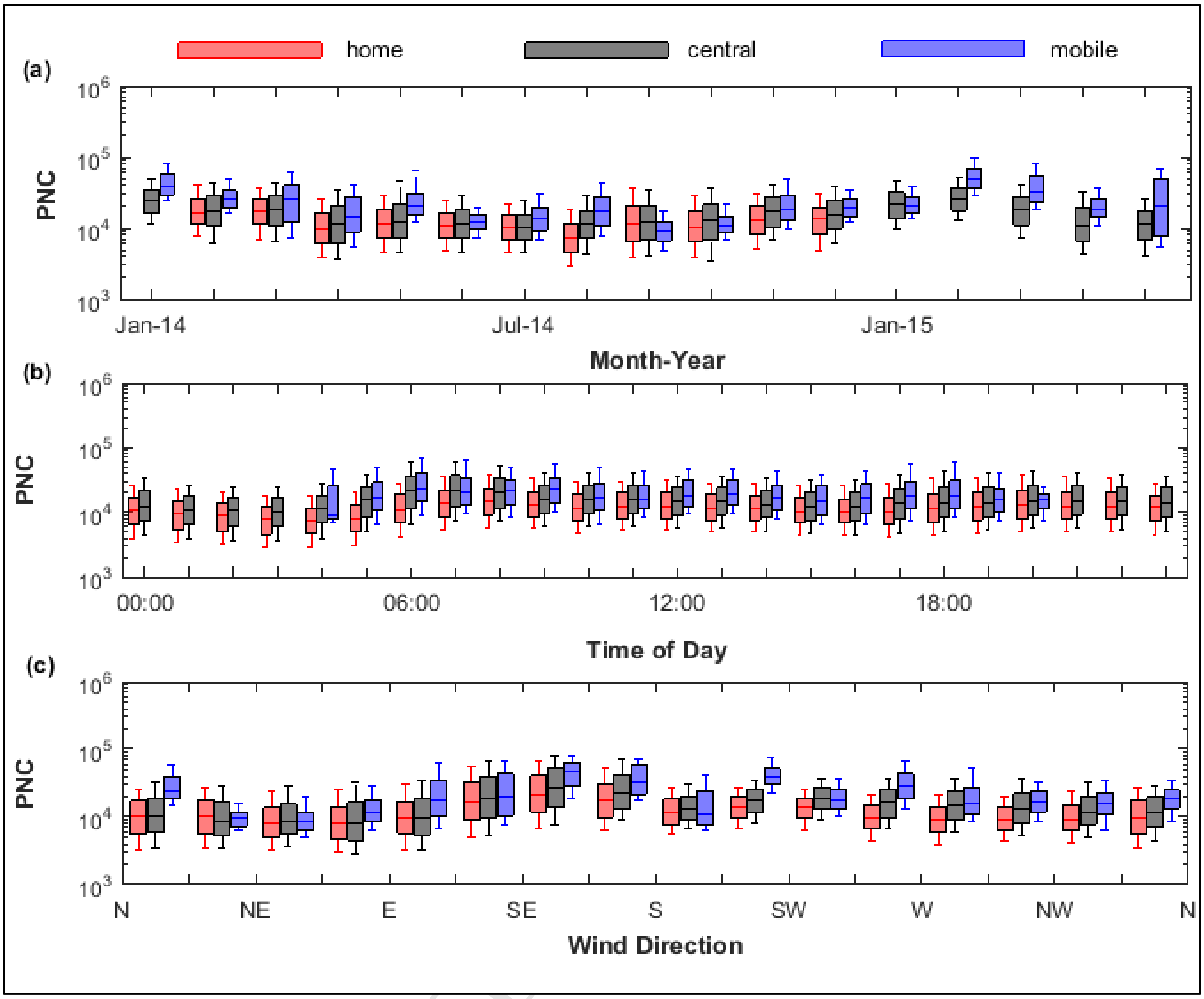


Figure 3: Boxplots of PNC by (a) month, (b) time of day, and (c) wind direction measured at central sites (black), homes (blue), and with a mobile laboratory (red) in Chelsea. Mobile monitoring occurred between 05:00 and 21:00.

3.2 Systematic Differences Between Monitoring Platforms

PNC measurements from the three different monitoring platforms were significantly ($p<0.05$) different. One-minute-average PNC at the central sites in Boston and Chelsea were higher (6,200 particles/cm^3 and 3,700 particles/cm^3, respectively) than concurrent measurements at the residential sites (Fig. 4a,b). These differences did not attenuate as a result of averaging over longer periods (i.e., one hour or one day) (Fig. S6). On-road PNC measurements were significantly higher than central-site measurements; the systematic measurement difference was >5-fold higher in Boston than in Chelsea (35,000 particles/cm^3 vs. 6,700 particles/cm^3, respectively) (Fig. 4c,d). Likewise, on-road PNC measurements near residential sites were significantly higher than the residential-site measurements (19,000 particles/cm^3 on average in Boston and 5,300 particles/cm^3 on average in Chelsea) (Fig. 4e,f). Spikes in PNC from vehicles near the mobile laboratory strongly influenced the on-road measurements. Removing these spikes from the data resulted in significant ($p<0.05$) reductions (46-95%) in the systematic differences in central-site-to-on-road comparisons and non-significant reductions (26-30%) for residential-site-to-on-road comparisons (Fig. S7 and S8).

The fanning effect observed in the Bland-Altman plots in Fig. 4 indicates the presence of additive error structure in the PNC measurements, i.e., as the mean PNC between any two platforms increased, the difference in PNC measurements by the two platforms also increased. This can potentially lead to overestimating the reported differences between the platforms and inflate Pearson correlations. We also generated Bland-Altman plots based on log-transformed PNC (Fig. S9-S11); log-transformation mitigated the impact of outliers. The fanning effect in these plots was dramatically reduced and mean differences were closer to zero, nonetheless the differences between platforms were still statistically significant: on-road concentrations were higher than central-site concentrations and both were higher than concentrations at residences. Systematically lower concentrations at residences has important implications for exposure assessment in epidemiology studies because most studies to date use stationary, central sites and/or mobile monitoring as the basis for exposure assessment which could lead to overestimated exposures.

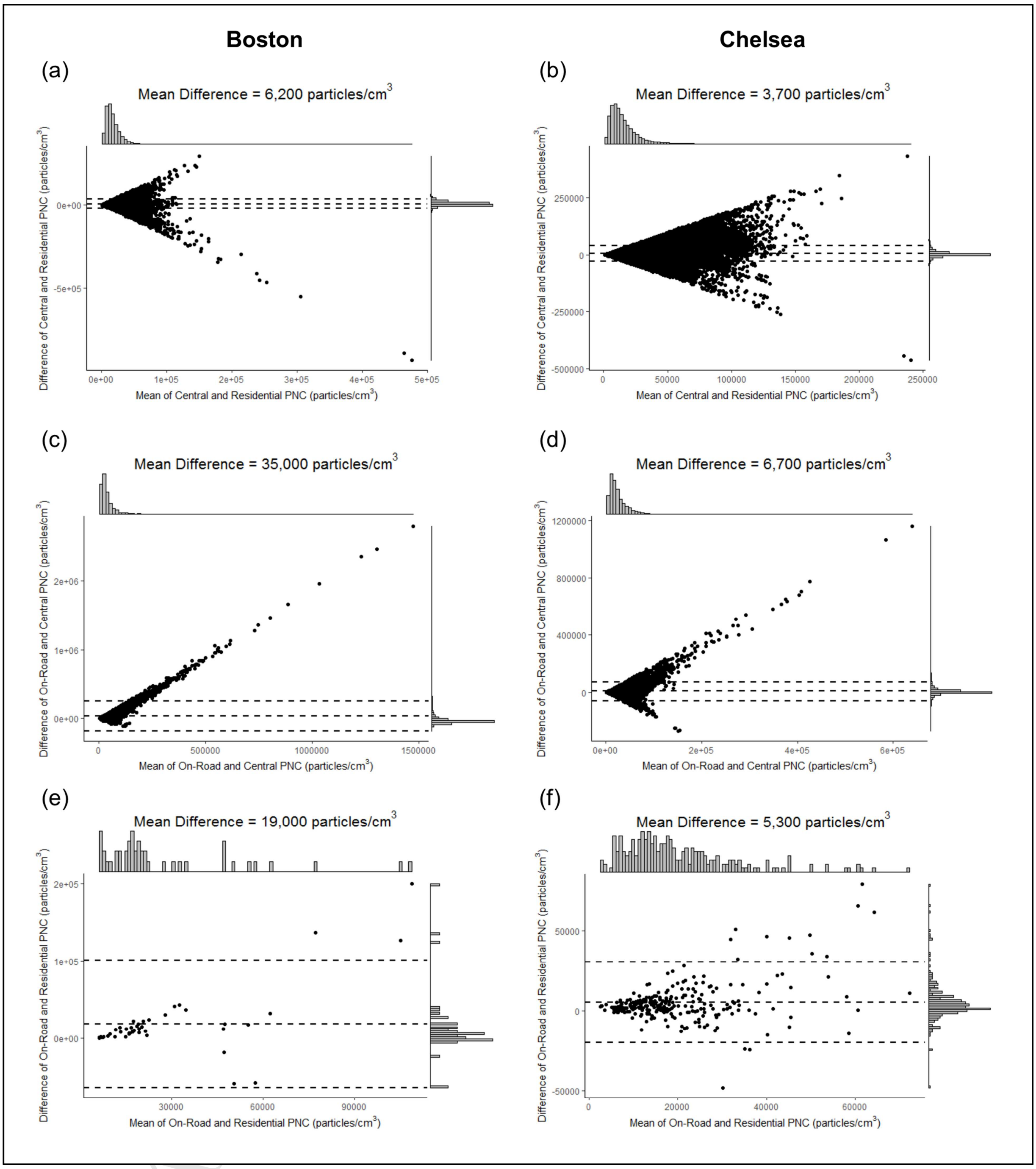


Figure 4: Bland-Altman plots of the mean PNC measured by the two platforms being compared (x-axis) versus the difference in measured PNC (y-axis). Differences from zero indicate positive or negative differences between the platform listed first in the axis label relative to the second. Trending tendencies above zero indicate systematic positive differences. The center dashed line represents the mean

difference; the outer dashed lines represent ± two standard deviations from the mean difference. The distribution of data can be determined by the histograms along the x2 and y2 axes. (a,b) Comparisons between central-site and residential-site PNC; (c,d) comparisons between central-site and on-road PNC; (e,f) comparisons between residential and on-road PNC.

3.3 Correlations Between PNC Monitoring Platforms

Pearson correlation coefficients between the different platforms were generally similar in both study areas (Table 3 and Fig. 5a,b). Median central-to-residential-site and central-site-to-on-road Pearson correlations were not significantly different in either Boston or Chelsea. Only when the entire data set was used to calculate a single correlation coefficient for each of the platform comparisons were correlations significantly different (see call-out plots in Fig. 5a,b). COD values for each of the platform comparisons were significantly different in both study areas, but only when comparing on-road-to-residential COD to the median central-to-on-road COD (Table 3 and Fig. 5c,d). Results did not change when we removed Homes 3 and 15 in the sensitivity analysis (Table S6). The correlation of on-road and central-site measurements with residential-site PNC suggests that exposure assessment based on on-road or central-site PNC should reflect temporal trends at homes.

	Central-Site:Homes		**Central-Site:On-Road**		**Homes:On-Road**[a, b]	
	Boston (*n*=11)[c]	Chelsea (*n*=9)[c]	Boston (*n*=178)[c]	Chelsea (*n*=90)[c]	Boston (*n*=1)[c]	Chelsea (*n*=1)[c]
r	0.39 (0.26-0.47)	0.45 (0.33-0.62)	0.45 (0.43-0.47)	0.43 (0.39-0.44)	0.18	0.62
COD	0.33 (0.31-0.36)	0.31 (0.26-0.33)	0.37 (0.36-0.38)	0.30 (0.29-0.31)	0.41	0.26

[a] Only six out of 11 homes were included in the Boston analysis. Of the other five home sites, two were not within 500 m of the TAPL route, three others were not monitored outdoors when the TAPL passed by. [b] The 95% confidence interval for the single Pearson correlation coefficient for the homes-to-on-road comparison in Boston and Chelsea was -0.12 to 0.45 and 0.53 to 0.69, respectively. [c] *n* represents the number of Pearson correlations or COD values in each summary statistic and not the number of data points used to calculate a Pearson correlation or COD value, which are presented in Table 1.

Table 3: Median summary statistics with 95% confidence intervals for each monitoring platform comparison based on one-minute PNC.

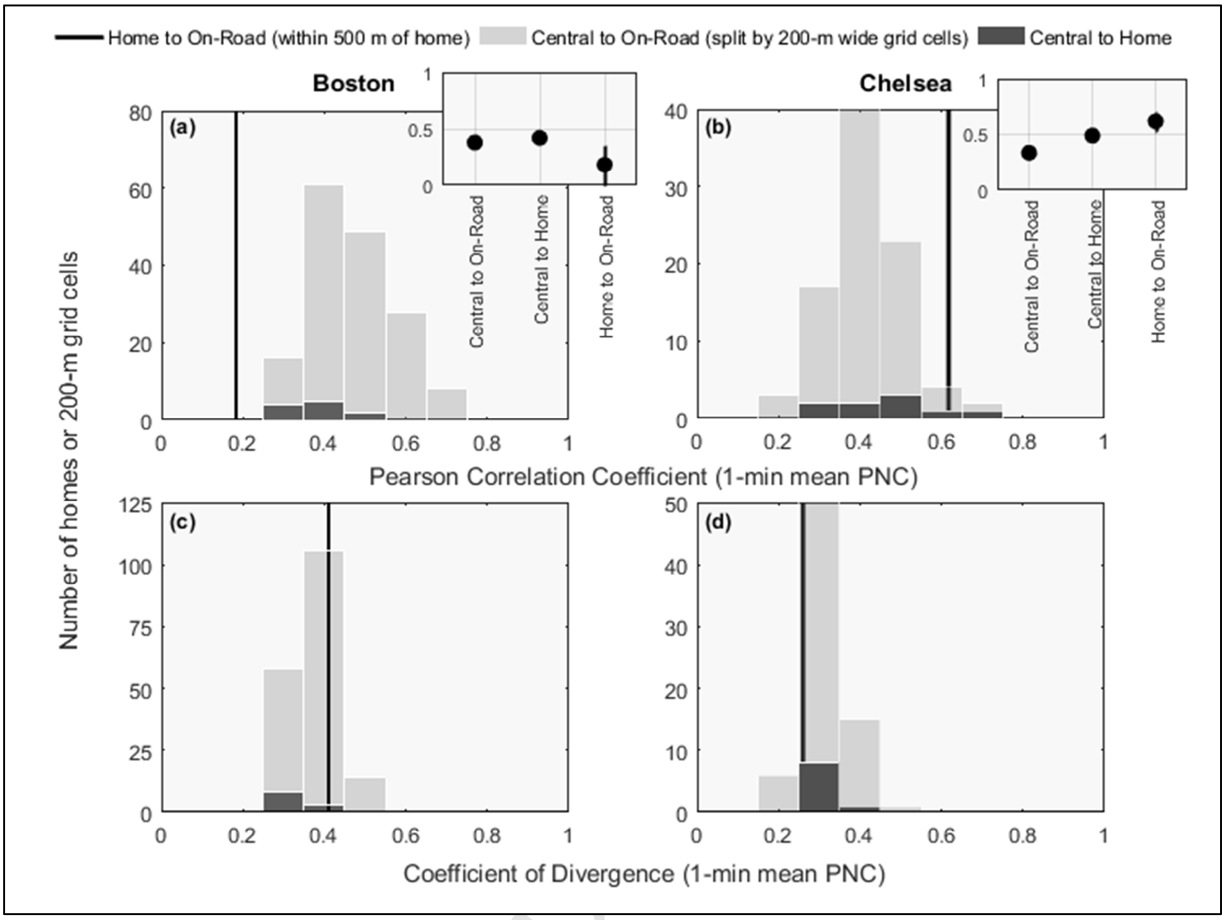
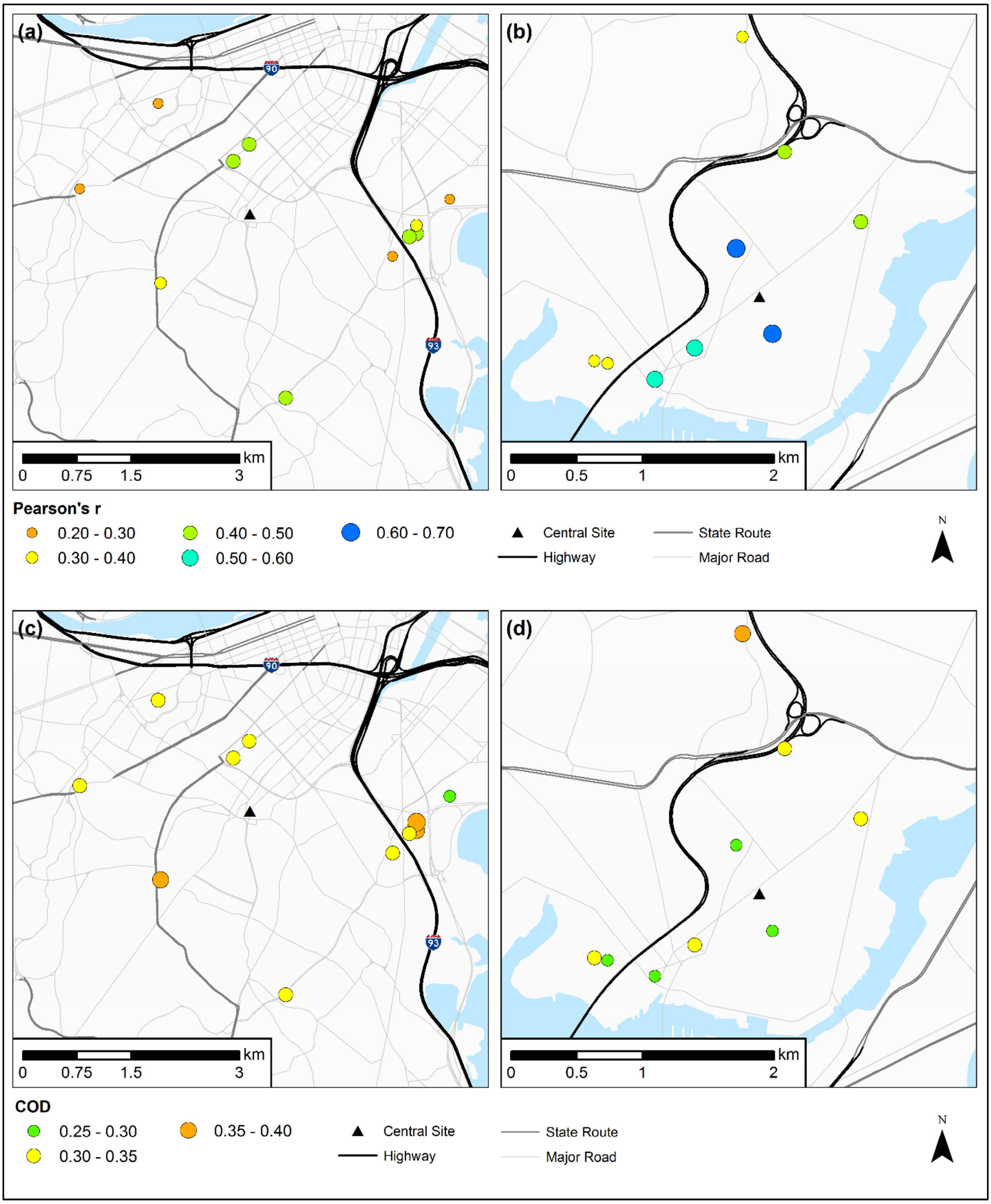


Figure 5: (a,b) Distribution of Pearson correlation coefficients and (c,d) coefficients of divergence by comparison. A solid vertical line is shown for the home-to-on-road comparison since there was only a single calculated correlation value (Pearson correlation in the Boston home-to-on-road comparison was not significant). Call-out plots in upper right show Pearson correlations for the complete data set by platform comparison (vertical lines represent 95% confidence interval; dots are larger than confidence intervals for some of the platform comparisons).

3.3.1 Central-Site Versus Residential-Site

Pearson correlations between central- and residential-site one-minute-mean PNC in Boston ranged from 0.25 to 0.48 while in Chelsea they ranged from 0.33 to 0.66. Residential sites with the highest Pearson correlations in Boston were typically downwind of high-traffic sources or in high-traffic areas (Fig. 6a). In Chelsea, the highest Pearson correlations were at residential sites east of US-1 (including the central site) with the two highest-correlation sites both within 500 m of the central site (Fig. 6b). COD values based on one-minute-mean PNC were between 0.28 and 0.37 in Boston and between 0.26 and 0.37 in Chelsea, indicating a moderate degree of spatial heterogeneity in both study areas. Residential sites with the lowest COD values were scattered throughout the study area with no apparent pattern (Fig. 6c,d). This suggests that the assumption that residential proximity to monitoring sites will better reflect PNC levels may not be generally applicable.

Averaging PNC data over hours and days resulted in higher temporal correlations (as compared to one minute) in both study areas (Table S7, Fig. S12a,b and S13a,b); however, the results were not significant, likely because of the smaller sample sizes. At longer averaging periods, the effects of transient PNC spikes from local sources (e.g., vehicles) were smoothed out, and the results were more representative of longer trends (e.g., hourly and daily changes in traffic activity and meteorology) across the study area. Pearson correlations based on daily-averaged PNC in Boston and Chelsea (0.69 and 0.71, median values, respectively; Table S7) were consistent with Puustinen et al. (2007), who reported that Pearson correlations between daily-averaged PNC at central and residential sites in four European cities ranged from 0.67 to 0.76 (median values). Comparing central-site and residential-site PNC using Spearman correlation coefficients and Pearson correlation coefficients with log-transformed PNC did not change our results: median correlations increased over longer averaging times in both study areas, but the differences were not significantly different (Tables S7 and S8). Similarly, COD changed by averaging data over longer time periods: COD calculated from daily-averaged PNC were significantly lower than COD based on one-minute-averaged PNC in both study areas (Table S7, Fig. S12c,d and S13c,d).



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393 Figure 6: (a,b) Maps of Pearson correlation coefficients and (c,d) coefficients of divergence between
394 central-site and residential-site PNC (one-minute mean PNC) in Boston (a,c) and Chelsea (b,d).

The generally lower Pearson correlation coefficients and higher COD values in Boston compared to Chelsea (differences were not significant at $p<0.05$) could be due to the location of the Boston central-site monitor in a highly-trafficked area (i.e., at grade and 75 m from the Dudley Square bus station) compared to most of the Boston residential sites (Table 3). We used the EPA-STN site, a secure, centrally-located site >1,500 m from I-93, but it was likely influenced by bus emissions when winds were from the 225° to 315° wind sector (26% of measurements, which excludes hours when buses were not operating). In contrast, the Chelsea central-site monitor was elevated 10 m above grade and set back 45 m from the nearest road as were many of the Chelsea residential sites, with the exception of a diesel rail line 50 m north of the site (<1% of the measurements were impacted by trains). PNC at the Boston central site during the morning rush hour period were generally much higher than at the residential sites. In contrast, in Chelsea we did not observe substantial differences in PNC between the central and residential sites during these hours. Overnight differences in both study areas were minimal and resulted in higher Pearson correlations and lower spatial heterogeneity as expected (Fig. 7).

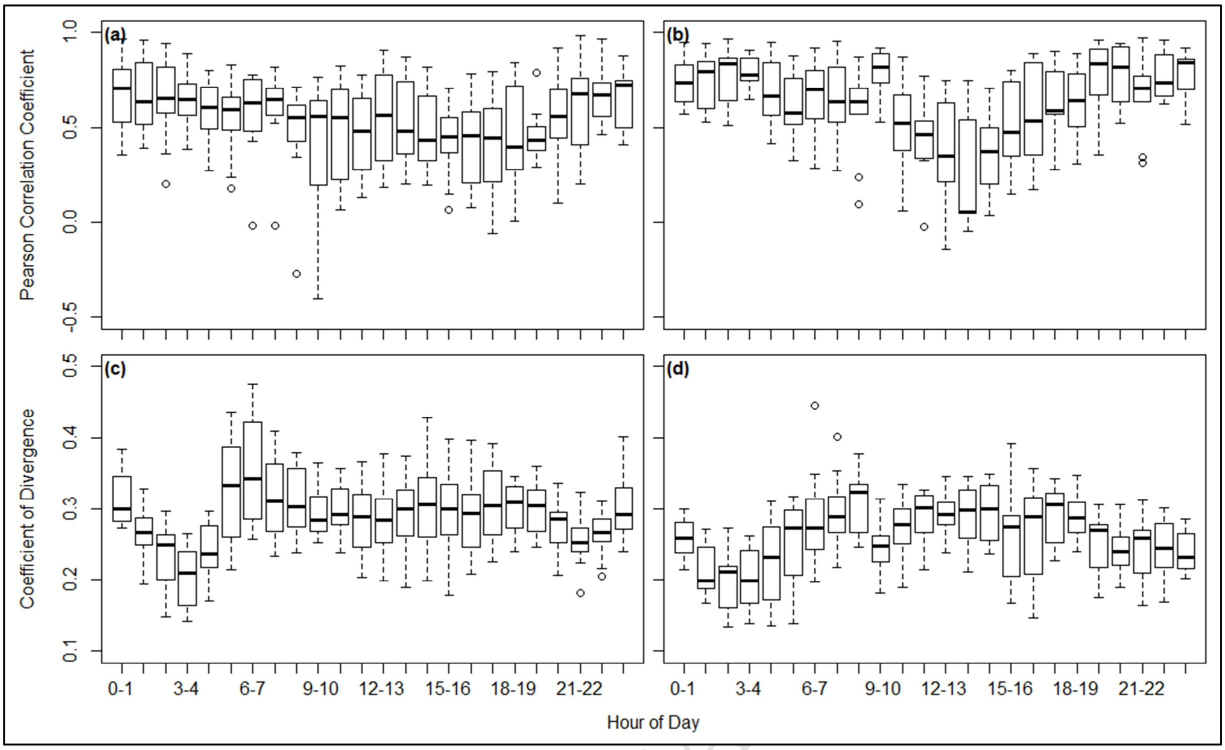
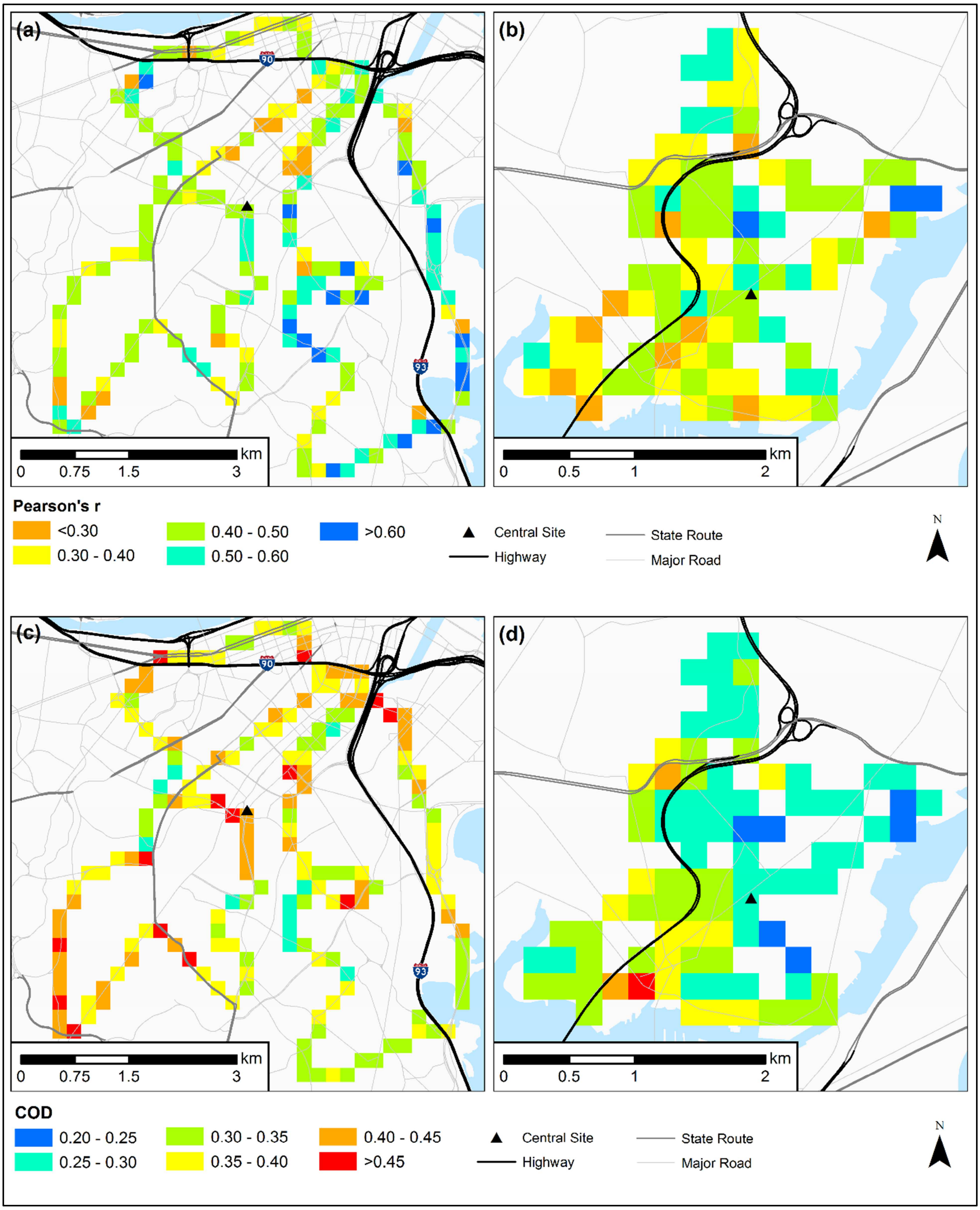


Figure 7: (a,b) Pearson correlation coefficients and (c,d) coefficients of divergence between central-site and residential PNC by hour of day (mean hourly PNC) in Boston (a,c) and Chelsea (b,d).

3.3.2 Central-Site Versus On-Road

Pearson correlations between PNC measurements from the central-site and on-road monitoring varied widely within the study areas. In Boston correlations ranged from 0.05 to 0.75 and in Chelsea they ranged from 0.23 to 0.69. The wide range of correlations in both study areas likely reflects differences in traffic conditions (and possibly other PNC sources) between the central sites and grid cells. For example, grid cells east of I-93 in the Boston study area were generally more correlated with the central site than the most western portion of the mobile monitoring route (Fig. 8a). This was likely because these grid cells were often downwind of I-93, a significant PNC source, while the Boston central site was at the same time downwind of Dudley Station. In Chelsea, residential areas east of US-1 were more highly correlated with the central site (Fig. 8b), again, likely because of the similarities between the traffic conditions in these particular grid cells and near the Chelsea central site. Using Spearman correlations and Pearson correlations with log-transformed PNC increased the correlation values and showed correlations in Boston and Chelsea were significantly different (Table S8). The COD values ranged from 0.27 to 0.51 in Boston and from 0.23 to 0.45 in Chelsea. In Boston, high COD values were observed throughout much of the study area (Fig. 8c), especially for the grid cells where the mobile laboratory was often in heavy traffic. COD values were generally lower in Chelsea, with the lowest values observed in the residential areas with light traffic (Fig. 8d). Removing on-road spikes from the analyses resulted in a non-significant increase in the median Pearson correlation in the Boston study area (coefficients increased from 0.45 to 0.48) and a significantly higher median Pearson correlation in the Chelsea study area (coefficients increased from 0.43 to 0.50). Median COD values decreased in both study areas (from 0.37 to 0.34 in Boston and from 0.30 to 0.28 in Chelsea). Using on-road median PNC instead of the mean did not significantly change Pearson correlations or COD values in either study area (Table S9 and Fig. S14).


(a)
(b)
km
0 0.75 1.5 3
km
0 0.5 1 2
Pearson's r
<0.30
0.30 - 0.40
0.40 - 0.50
0.50 - 0.60
>0.60
Central Site
Highway
State Route
Major Road
N
(c)
(d)
km
0 0.75 1.5 3
km
0 0.5 1 2
COD
0.20 - 0.25
0.25 - 0.30
0.30 - 0.35
0.35 - 0.40
0.40 - 0.45
>0.45
Central Site
Highway
State Route
Major Road
N

435

Figure 8: (a,b) Maps of Pearson correlation coefficients and (c,d) coefficients of divergence for concurrent one-minute mean PNC from central-site and on-road measurements in Boston (a,c) and Chelsea (b,d).

3.3.3 Residential-Site Versus On-Road

Due to the limited amount of on-road PNC data available when the mobile laboratory was <500 m from residential sites (i.e., 2-8 one-minute-average data points per home in Boston and 7-82 one-minute-average data points per home in Chelsea), the statistics reported here are based on pooled measurements from all residential sites within each study area with all on-road PNC data <500 m of the homes. The Pearson correlation coefficient between residential-site and on-road PNC was 0.18 (not significant) in Boston and 0.62 in Chelsea. The low correlation in Boston is likely because of higher-trafficked roads near the residential sites and the low number of data points (n=45) with a wide confidence interval (95% CI: -0.12-0.45) used in the calculation. Conversely, the higher correlation in Chelsea is likely because both the residential sites and 500-m buffers around these sites were mostly in residential areas, and most of the sections of the mobile monitoring route in commercial and industrial areas fell outside the 500-m buffers around each home. The Chelsea data set also had substantially more data (n=247). Our Pearson correlation of 0.62 in Chelsea is similar to the Pearson correlation between on-road and short-term stationary sites in Amsterdam and Rotterdam where the coefficient was reported to be 0.67 for urban background areas (Kerckhoffs et al., 2016). Our results did not change by using Spearman correlations and Pearson correlations with log-transformed PNC, although correlation values were higher. Spatial differences were greater in Boston (COD = 0.41) than in Chelsea (COD = 0.26) likely because the mobile laboratory traveled on more high-PNC roads within 0-500 m of the residential sites in Boston as compared to Chelsea. Removing short-term on-road spikes increased Pearson correlations in both study areas, but not significantly. The median Pearson correlation was 0.36 in the Boston study area and 0.69 in the Chelsea study area. COD values decreased by 0.02 in both study areas.

3.4 Factors Affecting the Correlations between Monitoring Platforms

We found that the two factors that affected Pearson correlation and COD values the most (of those that we tested) were hour of day and wind direction. Other meteorological factors (e.g., wind speed, temperature, humidity, pressure, and atmospheric boundary layer) influenced the correlations, but to a lesser degree. Since adjusted-R^2 values were lower for other meteorological variables such as temperature and boundary layer, time of day may have served as a proxy for traffic (Tables S10 and S11). Spatial factors such as land use category of the sites and the proximity of monitors to each other did not significantly impact the correlations. The low adjusted-R^2 values are an indication that either unaccounted for factors influence the Pearson correlation and COD values between the measurement platforms or that localized effects (e.g., sources near the monitors) masked the actual meteorological effects.

In general, overnight hours had higher hourly Pearson correlations and lower hourly COD values compared to daytime hours (Fig. 7). This is likely because nighttime vehicle traffic was light, buses were not running between 01:00 and 05:00, and flight operations at Logan Airport were substantially reduced (mean landings and take-offs were 5.0 h^{-1} between 00:00 and 06:00 compared to 46.2 h^{-1} during all other hours (Hudda et al., 2016)). After 05:00 traffic increased throughout the two study areas; however, traffic volume was not uniformly distributed, and thus some areas received much higher increases in PNC than did others. During the daytime COD values in both study areas remained relatively high and then decreased after the evening rush hour period ended at ~19:00. Similar Pearson correlation and COD trends were also observed when one-minute PNC was used, albeit less discernable, indicating the strong influence of traffic. Since participants in epidemiology studies will most often be at home during the night, attention to nighttime exposures may be particularly important.

In the Boston study area, Pearson correlations were highest when winds were from the 45° to 90° (ENE) wind sector (which occurred during 13% of the study period). The highest correlations in Chelsea were observed when winds were from the 180° to 225° wind sector (19% of the study period), followed closely by both the 135° to 180° (SSE) and 225° to 270° wind sectors (6% and 12% of the study period, respectively). Hudda et al. (2016) observed elevated PNC in Boston during ENE winds and in Chelsea during SSE winds and attributed the increases to aviation emissions. Both Fuller et al. (2012) and Patton et al. (2014b) also observed elevated PNC in Boston neighborhoods during winds from the airport. It can be hypothesized that under these wind conditions, aircraft emissions at Logan Airport could have a widespread impact on the entire monitoring domain leading to higher correlations between platforms. Wind conditions also impacted COD values in both study areas. In Boston, higher COD values between central- and residential-site PNC were observed during winds from the 225° to 315° wind sector (32% of the study period), when the central-site monitor was downwind from a major bus station 75 m to the west and other local sources. In contrast, higher COD values between PNC at the central and residential sites in Chelsea were observed when winds were from the 45° to 90° wind sector (10% of the study period) possibly due to upwind sources (e.g., trains traveling along the stretch of rail just northeast of the central site and oil tankers on Chelsea Creek).

3.5 Limitations

Our study had several limitations. First, to minimize the potential for self-sampling we excluded on-road measurements from intersections when the TAPL slowed to <5 km/h for >10 s. Nonetheless, we were able to drive through >65% of intersections without slowing below 5 km/h for >10 s. Therefore, our data set for on-road measurements does not significantly underrepresent the near-intersection environment. Second, we had limited simultaneous deployments at residences with which to calculate Pearson correlations and COD values between different residential sites. This would have allowed us to develop a better understanding of the spatial PNC variability within the study areas; however, we were able to compare each home to the central site and mobile monitoring, which was the main goal of the study. Third, the density of residential monitoring sites was 5-fold higher in the Chelsea study area (1.5 sites/km^2) compared to Boston (0.28 sites/km^2). This may help to explain why we observed generally higher Pearson correlations and lower COD values in Chelsea compared to Boston (Table 3). In comparison to other studies, the densities of residential sites in our two study areas were at the higher end of the range (range = 0.03 to 16.7 sites/km^2, median = 0.15 sites/km^2) (Abernethy et al., 2013; Fuller et al., 2012; Klompmaker et al., 2015; Meier et al., 2015; Moore et al., 2009; Puustinen et al., 2007; Rivera et al., 2012; Sabaliauskas et al., 2015; Wolf et al., 2017). Fourth, in order to have enough data to compare PNC measured at residential sites to on-road measurements we pooled all on-road data within 500-m buffers around all homes rather than calculate correlations for each home separately. While this removed seasonality effects from the data, we found seasonality did not significantly affect the platform correlations (Tables S10 and S11). Fifth, the location of the central site near Dudley Station may not have led to a representative characterization of urban background pollutant levels in the Boston study area. However, the impacts from bus emissions were typically short-lived and were most apparent in the one-minute-averaged PNC data. In contrast, the relatively low impact of local emissions at the central site in Chelsea likely contributed to the higher Pearson correlations and lower COD values in Chelsea compared to Boston. Lastly, while the main objective of this study was to investigate traffic-related UFP we also, unexpectedly, observed impacts from Logan Airport. These impacts were limited to periods when winds were from the direction of the airport (i.e., 13% of the time in the Boston study area and 6% of the time in the Chelsea study area). We conducted a sensitivity analysis to determine whether Pearson correlations and COD values differ when winds from the direction of Logan Airport were excluded from the calculations for both study areas. When winds from Logan were excluded COD values were unchanged, and Pearson correlations were not statistically significantly different except in the Chelsea central-to-residential-site comparison where the correlation was 12% lower. Therefore, aviation impacts from Logan appear to only have had a limited effect on our findings.

In this study we used Pearson correlation coefficients, COD values, and Bland-Altman plots to describe the similarities and differences in PNC measured by the three platforms. These metrics have limitations that should be discussed in the context of this study. First, Pearson correlations are not robust estimators for severely skewed data. We addressed this in part by calculating both Pearson correlations on ln(PNC) and Spearman rank correlations (a nonparametric test) on PNC, and both sets of estimates showed similar associations between measurement platforms. While we used a natural-log transformation to reduce the left skewness of our data set, we did not explore whether the selected transformation provides the best possible fit. Future studies should consider the sensitivity analysis in choosing the transformational form. Second, while COD values provide a measure of spatial heterogeneity between data sets, the values can be influenced by certain data-set characteristics, such as the units of analysis. Calculating COD values based on ln(PNC), for example, would have generated lower COD values than those we calculated using non-transformed PNC since the concentrations are on two completely different scales. We chose to present non-transformed results of COD to be comparable to literature, but due to the skewed nature of the data we may have overestimated the heterogeneity between platforms. Third, while Bland-Altman plots are useful for visualizing absolute differences between measurements, the results are also influenced by extreme values. To mitigate against this we calculated mean differences using both PNC and ln(PNC), both of which showed there were systematic differences between the platform measurements. Although the natural-log transformation worked well for this study, a more rigorous selection and justification of the transformations would be desirable. It should also be noted that our results for systematic platform differences are based on our specific study design; a different study design – for example, one where we measured on-road PNC only in residential areas – may have generated different measures of systematic differences.

3.6 Implications for Urban Air Quality Monitoring

We designed our monitoring strategy to support the development of finely spatially- (<20 m) and temporally-resolved (hourly) ambient PNC exposure models for BPRHS participants. Central sites were selected to measure long-term temporal trends within the study areas, mobile monitoring was designed to characterize spatial contrasts, and residential sites were meant to be representative of participant exposures at homes. We found that while absolute PN concentrations differed significantly between central-site, on-road, and residential-site monitoring, temporal patterns were similar across the three different monitoring platforms in both study areas. While each monitoring platform has benefits, the decision to use short-term residential monitoring at many sites versus using a small number of longerterm central sites supplemented with mobile monitoring may be better informed by considering the characteristics of the study area. For example, the latter approach may be more effective in areas where higher spatial contrasts are expected – i.e., in areas containing multiple busy roadways – and long-term trends are of interest. New mobile monitoring strategies, such as measuring NO_2 with Google Street View vehicles (Apte et al., 2017), could aid in this approach and may increase the ability to characterize the high spatial variability of UFP. In contrast, the former approach may be useful in more residential areas with fewer busy roadways. Simultaneous application of all three monitoring platforms may be useful for developing models, where mobile monitoring and central-site monitoring can serve to characterize PNC in the study area and residential monitoring can be used for model validation and/or calibration. To our knowledge, only two studies have conducted concurrent long-term/central-site stationary, (multiple) short-term stationary, and mobile monitoring of PNC, both of which were for PNC modeling applications. (Kerckhoffs et al., 2016; Sabaliauskas et al., 2015). In a study in Toronto, Ontario (Canada), Sabaliauskas et al. (2015) conducted continuous central-site monitoring (3 months), short-term monitoring at six sites (1-3 weeks per site), and mobile monitoring between 12:00 and 15:00 on 15 weekdays in the summer. In a study in Amsterdam and Rotterdam in The Netherlands, Kerckhoffs et al. (2016) conducted short-term monitoring at 80 sites per city (three 30-minute visits per site), mobile monitoring on 42 days between 09:00 and 16:00 in winter and spring per city, and continuous long-term monitoring (6 months) at a reference site 30-50 km away. Consistent with our observations, these studies reported generally similar temporal trends between platforms, but significantly higher PNC on roads with the mobile monitor. Our study adds to this body of literature by comparing these three monitoring strategies across longer sampling windows and in all four seasons.

Acknowledgements

We are grateful to Alex Bob, Jessica Perkins, Dana Harada, Amy Hunter, Joanna Stowell, Ruhui Zhao, Madeline Wrable, Hanaa Rohman, Andrew Shapero, Meg Keegan, Wilfred Mbah, and Thomas Heath II for assistance with data collection, to Wig Zamore for advice on study design, and to Allison Patton for advice on mobile monitoring, data quality control, and manuscript preparation. Alexis Soto and Nancy Figueroa recruited participants for home monitoring. We are grateful to Massachusetts Department of Environmental Protection (Roxbury, MA) and The Neighborhood Developers (Chelsea, MA) for providing space and electricity for our monitoring equipment, and to the Chelsea Police Department for allowing us to conduct mobile monitoring in the city. This work was funded by NIH-NHLBI grant CA148612 to the University of Massachusetts Lowell, NIH-NIEHS grant ES015462 to Tufts University, and a Tufts University Water: Systems, Science and Society Fellowship (Tufts Office of the Provost) to MCS.

Conflict of Interest Disclosure

The authors declare no competing financial interest.

References

Aalto, P., Hämeri, K., Paatero, P., Kulmala, M., Bellander, T., Berglind, N., Bouso, L., Castaño-Vinyals, G., Sunyer, J., Cattani, G., Marconi, A., Cyrys, J., Klot, S. von, Peters, A., Zetzsche, K., Lanki, T., Pekkanen, J., Nyberg, F., Sjövall, B., Forastiere, F., 2005. Aerosol Particle Number Concentration Measurements in Five European Cities Using TSI-3022 Condensation Particle Counter over a Three-Year Period during Health Effects of Air Pollution on Susceptible Subpopulations. J. Air Waste Manag. Assoc. 55, 1064–1076. doi:10.1080/10473289.2005.10464702

Abernethy, R.C., Allen, R.W., McKendry, I.G., Brauer, M., 2013. A Land Use Regression Model for Ultrafine Particles in Vancouver, Canada - Environmental Science & Technology. Environ. Sci. Technol. 47, 5217–5225. doi:10.1021/es304495s

Aggarwal, S., Jain, R., Marshall, J.D., 2012. Real-Time Prediction of Size-Resolved Ultrafine Particulate Matter on Freeways. Environ. Sci. Technol. 46, 2234–2241. doi:10.1021/es203290p

Aguilera, I., Dratva, J., Caviezel, S., Burdet, L., de Groot, E., Ducret-Stich, R.E., Eeftens, M., Keidel, D., Meier, R., Perez, L., Rothe, T., Schaffner, E., Schmit-Trucksäss, A., Tsai, M.-Y., Schindler, C., Künzli, N., Probst-Hensch, N., 2016. Particulate Matter and Subclinical Atherosclerosis: Associations between Different Particle Sizes and Sources with Carotid Intima-Media Thickness in the SAPALDIA Study. Environ. Health Perspect. 124. doi:10.1289/EHP161

Apte, J.S., Messier, K.P., Gani, S., Brauer, M., Kirchstetter, T.W., Lunden, M.M., Marshall, J.D., Portier, C.J., Vermeulen, R.C.H., Hamburg, S.P., 2017. High-Resolution Air Pollution Mapping with

Google Street View Cars: Exploiting Big Data. Environ. Sci. Technol. 51, 6999–7008. doi:10.1021/acs.est.7b00891
Boston Region Metropolitan Planning Organization, Central Transportation Planning Staff, 2011. Express Highway Volumes.
Boston Region Metropolitan Planning Organization, Central Transportation Planning Staff, n.d. ADT Data Browser Application [WWW Document]. URL www.ctps.org/map/www/apps/adtApp/index.html (accessed 12.14.16).
Cyrys, J., Pitz, M., Heinrich, J., Wichmann, H.-E., Peters, A., 2008. Spatial and temporal variation of particle number concentration in Augsburg, Germany. Sci. Total Environ. 401, 168–175. doi:10.1016/j.scitotenv.2008.03.043
Eeftens, M., Phuleria, H.C., Meier, R., Aguilera, I., Corradi, E., Davey, M., Ducret-Stich, R., Fierz, M., Gehrig, R., Ineichen, A., Keidel, D., Probst-Hensch, N., Ragettli, M.S., Schindler, C., Künzli, N., Tsai, M.-Y., 2015. Spatial and temporal variability of ultrafine particles, NO2, PM2.5, PM2.5 absorbance, PM10 and PMcoarse in Swiss study areas. Atmos. Environ. 111, 60–70. doi:10.1016/j.atmosenv.2015.03.031
Fuller, C.H., Brugge, D., Williams, P.L., Mittleman, M.A., Durant, J.L., Spengler, J.D., 2012. Estimation of ultrafine particle concentrations at near-highway residences using data from local and central monitors. Atmos. Environ. 57, 257–265. doi:10.1016/j.atmosenv.2012.04.004
Geiser, M., Rothen-Rutishauser, B., Kapp, N., Schürch, S., Kreyling, W., Schulz, H., Semmler, M., Hof, V.I., Heyder, J., Gehr, P., 2005. Ultrafine Particles Cross Cellular Membranes by Nonphagocytic Mechanisms in Lungs and in Cultured Cells. Environ. Health Perspect. 113, 1555–1560. doi:10.1289/ehp.8006
Hankey, S., Marshall, J.D., 2015. On-bicycle exposure to particulate air pollution: Particle number, black carbon, PM2.5, and particle size. Atmos. Environ. 122, 65–73. doi:10.1016/j.atmosenv.2015.09.025
HEI Panel of the Health Effects of Traffic-Related Air Pollution, 2010. Traffic-Related Air Pollution: A Critical Review of the Literature on Emissions, Exposure, and Health Effects (Review No. 17). Health Effects Institute.
HEI Review Panel on Ultrafine Particulates, 2013. Understanding the Health Effects of Ambient Ultrafine Particles (Perspectives No. 3). Health Effects Institute.
Hofman, J., Staelens, J., Cordell, R., Stroobants, C., Zikova, N., Hama, S.M.L., Wyche, K.P., Kos, G.P.A., Van Der Zee, S., Smallbone, K.L., Weijers, E.P., Monks, P.S., Roekens, E., 2016. Ultrafine particles in four European urban environments: Results from a new continuous long-term monitoring network. Atmos. Environ. 136, 68–81. doi:10.1016/j.atmosenv.2016.04.010
Hudda, N., Simon, M.C., Zamore, W., Brugge, D., Durant, J.L., 2016. Aviation Emissions Impact Ambient Ultrafine Particle Concentrations in the Greater Boston Area. Environ. Sci. Technol. 50, 8514–8521. doi:10.1021/acs.est.6b01815
Karner, A.A., Eisinger, D.S., Niemeier, D.A., 2010. Near-Roadway Air Quality: Synthesizing the Findings from Real-World Data. Environ. Sci. Technol. 44, 5334–5344. doi:10.1021/es100008x
Kerckhoffs, J., Hoek, G., Messier, K.P., Brunekreef, B., Meliefste, K., Klompmaker, J.O., Vermeulen, R., 2016. Comparison of Ultrafine Particle and Black Carbon Concentration Predictions from a Mobile and Short-Term Stationary Land-Use Regression Model. Environ. Sci. Technol. 50, 12894–12902. doi:10.1021/acs.est.6b03476
Klompmaker, J.O., Montagne, D.R., Meliefste, K., Hoek, G., Brunekreef, B., 2015. Spatial variation of ultrafine particles and black carbon in two cities: Results from a short-term measurement campaign. Sci. Total Environ. 508, 266–275. doi:10.1016/j.scitotenv.2014.11.088
Kumar, P., Fennell, P., Symonds, J., Britter, R., 2008. Treatment of losses of ultrafine aerosol particles in long sampling tubes during ambient measurements. Atmos. Environ. 42, 8819–8826. doi:10.1016/j.atmosenv.2008.09.003
Lane, K.J., Levy, J.I., Scammell, M.K., Peters, J.L., Patton, A.P., Reisner, E., Lowe, L., Zamore, W., Durant, J.L., Brugge, D., 2016. Association of modeled long-term personal exposure to ultrafine

particles with inflammatory and coagulation biomarkers. Environ. Int. 92–93, 173–182. doi:10.1016/j.envint.2016.03.013
Levy, I., Mihele, C., Lu, G., Narayan, J., Hilker, N., Brook, J.R., 2014. Elucidating multipollutant exposure across a complex metropolitan area by systematic deployment of a mobile laboratory. Atmospheric Chem. Phys. Katlenburg-Lindau 14, 7173. doi:http://dx.doi.org/10.5194/acp-14-7173-2014
Li, L., Wu, J., Hudda, N., Sioutas, C., Fruin, S.A., Delfino, R.J., 2013. Modeling the Concentrations of On-Road Air Pollutants in Southern California. Environ. Sci. Technol. 47, 9291–9299. doi:10.1021/es401281r
Martin Bland, J., Altman, D., 1986. Statistical methods for assessing agreement between two methods of clinical measurement. The Lancet, Originally published as Volume 1, Issue 8476 327, 307–310. doi:10.1016/S0140-6736(86)90837-8
Meier, R., Eeftens, M., Aguilera, I., Phuleria, H.C., Ineichen, A., Davey, M., Ragettli, M.S., Fierz, M., Schindler, C., Probst-Hensch, N., Tsai, M.-Y., Künzli, N., 2015. Ambient Ultrafine Particle Levels at Residential and Reference Sites in Urban and Rural Switzerland. Environ. Sci. Technol. 49, 2709–2715. doi:10.1021/es505246m
Moore, K., Krudysz, M., Pakbin, P., Hudda, N., Sioutas, C., 2009. Intra-Community Variability in Total Particle Number Concentrations in the San Pedro Harbor Area (Los Angeles, California). Aerosol Sci. Technol. 43, 587–603. doi:10.1080/02786820902800900
NOAA National Centers for Environmental Information, n.d. URL https://www.ncdc.noaa.gov/data-access (accessed 6.17.15).
Oberdörster, G., Oberdörster, E., Oberdörster, J., 2005. Nanotoxicology: An Emerging Discipline Evolving from Studies of Ultrafine Particles. Environ. Health Perspect. 113, 823–839. doi:10.1289/ehp.7339
Office of Geographic Information (MassGIS), Commonwealth of Massachusetts, MassIT, 2005. MassGIS Data - Land Use [WWW Document]. URL http://www.mass.gov/anf/research-and-tech/it-serv-and-support/application-serv/office-of-geographic-information-massgis/datalayers/layerlist.html (accessed 3.18.16).
Ostro, B., Hu, J., Goldberg, D., Reynolds, P., Hertz, A., Bernstein, L., Kleeman, M.J., 2015. Associations of Mortality with Long-Term Exposures to Fine and Ultrafine Particles, Species and Sources: Results from the California Teachers Study Cohort. Environ. Health Perspect. doi:10.1289/ehp.1408565
Padró-Martínez, L.T., Patton, A.P., Trull, J.B., Zamore, W., Brugge, D., Durant, J.L., 2012. Mobile monitoring of particle number concentration and other traffic-related air pollutants in a near-highway neighborhood over the course of a year. Atmos. Environ. 61, 253–264. doi:10.1016/j.atmosenv.2012.06.088
Pattinson, W., Longley, I., Kingham, S., 2014. Using mobile monitoring to visualise diurnal variation of traffic pollutants across two near-highway neighbourhoods. Atmos. Environ. 94, 782–792. doi:10.1016/j.atmosenv.2014.06.007
Patton, A.P., Collins, C., Naumova, E.N., Zamore, W., Brugge, D., Durant, J.L., 2014a. An Hourly Regression Model for Ultrafine Particles in a Near-Highway Urban Area. Environ. Sci. Technol. 48, 3272–3280. doi:10.1021/es404838k
Patton, A.P., Perkins, J., Zamore, W., Levy, J.I., Brugge, D., Durant, J.L., 2014b. Spatial and temporal differences in traffic-related air pollution in three urban neighborhoods near an interstate highway. Atmos. Environ. 99, 309–321. doi:10.1016/j.atmosenv.2014.09.072
Patton, A.P., Zamore, W., Naumova, E.N., Levy, J.I., Brugge, D., Durant, J.L., 2015. Transferability and Generalizability of Regression Models of Ultrafine Particles in Urban Neighborhoods in the Boston Area. Environ. Sci. Technol. 49, 6051–6060. doi:10.1021/es5061676
Pekkanen, J., Kulmala, M., 2004. Exposure assessment of ultrafine particles in epidemiologic time-series studies. Scand. J. Work. Environ. Health 30 Suppl 2, 9–18.

Puustinen, A., Hämeri, K., Pekkanen, J., Kulmala, M., de Hartog, J., Meliefste, K., ten Brink, H., Kos, G., Katsouyanni, K., Karakatsani, A., Kotronarou, A., Kavouras, I., Meddings, C., Thomas, S., Harrison, R., Ayres, J.G., van der Zee, S., Hoek, G., 2007. Spatial variation of particle number and mass over four European cities. Atmos. Environ. 41, 6622–6636. doi:10.1016/j.atmosenv.2007.04.020

Riley, E.A., Banks, L., Fintzi, J., Gould, T.R., Hartin, K., Schaal, L., Davey, M., Sheppard, L., Larson, T., Yost, M.G., Simpson, C.D., 2014. Multi-pollutant mobile platform measurements of air pollutants adjacent to a major roadway. Atmos. Environ. 98, 492–499. doi:10.1016/j.atmosenv.2014.09.018

Riley, E.A., Schaal, L., Sasakura, M., Crampton, R., Gould, T.R., Hartin, K., Sheppard, L., Larson, T., Simpson, C.D., Yost, M.G., 2016. Correlations between short-term mobile monitoring and long-term passive sampler measurements of traffic-related air pollution. Atmos. Environ. 132, 229–239. doi:10.1016/j.atmosenv.2016.03.001

Rivera, M., Basagaña, X., Aguilera, I., Agis, D., Bouso, L., Foraster, M., Medina-Ramón, M., Pey, J., Künzli, N., Hoek, G., 2012. Spatial distribution of ultrafine particles in urban settings: A land use regression model. Atmos. Environ. 54, 657–666. doi:10.1016/j.atmosenv.2012.01.058

Sabaliauskas, K., Jeong, C.-H., Yao, X., Reali, C., Sun, T., Evans, G.J., 2015. Development of a land-use regression model for ultrafine particles in Toronto, Canada. Atmos. Environ. 110, 84–92. doi:10.1016/j.atmosenv.2015.02.018

Steffens, J., Kimbrough, S., Baldauf, R., Isakov, V., Brown, R., Powell, A., Deshmukh, P., 2017. Near-port air quality assessment utilizing a mobile measurement approach. Atmospheric Pollut. Res. doi:10.1016/j.apr.2017.04.003

Tucker, K.L., Mattei, J., Noel, S.E., Collado, B.M., Mendez, J., Nelson, J., Griffith, J., Ordovas, J.M., Falcon, L.M., 2010. The Boston Puerto Rican Health Study, a longitudinal cohort study on health disparities in Puerto Rican adults: challenges and opportunities. BMC Public Health 10, 107. doi:10.1186/1471-2458-10-107

Viehmann, A., Hertel, S., Fuks, K., Eisele, L., Moebus, S., Möhlenkamp, S., Nonnemacher, M., Jakobs, H., Erbel, R., Jöckel, K.-H., Hoffmann, B., 2015. Long-term residential exposure to urban air pollution, and repeated measures of systemic blood markers of inflammation and coagulation. Occup. Environ. Med. 72, 656–663. doi:10.1136/oemed-2014-102800

Wang, Y., Hopke, P.K., Chalupa, D.C., Utell, M.J., 2011. Long-term study of urban ultrafine particles and other pollutants. Atmos. Environ., Air Pollution and Health: Bridging the Gap from Sources-to-Health Outcomes 45, 7672–7680. doi:10.1016/j.atmosenv.2010.08.022

Weichenthal, S., Van Ryswyk, K., Goldstein, A., Shekarrizfard, M., Hatzopoulou, M., 2016. Characterizing the spatial distribution of ambient ultrafine particles in Toronto, Canada: A land use regression model. Environ. Pollut., Special Issue: Urban Health and Wellbeing 208, Part A, 241–248. doi:10.1016/j.envpol.2015.04.011

Wilson, J.G., Kingham, S., Pearce, J., Sturman, A.P., 2005. A review of intraurban variations in particulate air pollution: Implications for epidemiological research. Atmos. Environ. 39, 6444–6462. doi:10.1016/j.atmosenv.2005.07.030

Wolf, K., Cyrys, J., Harciníková, T., Gu, J., Kusch, T., Hampel, R., Schneider, A., Peters, A., 2017. Land use regression modeling of ultrafine particles, ozone, nitrogen oxides and markers of particulate matter pollution in Augsburg, Germany. Sci. Total Environ. 579, 1531–1540. doi:10.1016/j.scitotenv.2016.11.160

Wongphatarakul, V., Friedlander, S.K., Pinto, J.P., 1998. A Comparative Study of PM2.5 Ambient Aerosol Chemical Databases. Environ. Sci. Technol. 32, 3926–3934. doi:10.1021/es9800582

World Health Organization, 2013. Review of evidence on health aspects of air pollution – REVIHAAP project: final technical report.

Zwack, L.M., Paciorek, C.J., Spengler, J.D., Levy, J.I., 2011. Modeling Spatial Patterns of Traffic-Related Air Pollutants in Complex Urban Terrain. Environ. Health Perspect. 119, 852–859. doi:10.1289/ehp.1002519

Highlights:

- Ultrafine concentrations were monitored at central-sites, residences, and on-road.
- Time of day and wind direction affected correlations between the three platforms.
- Hourly and daily trends were similar at central sites, residences, and on roads.
- Particle concentrations on roads were significantly higher than other platforms.