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Ambient air quality measurements from a continuously moving mobile platform: Estimation of area-wide, fuel-based, mobile source emission factors using absolute principal component scores

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HIGHLIGHTS

- Heavy and light duty fuel-based emission factors estimated for NOx, CO, BC, and PN.
- Factors are consistent with remote sensing, vehicle chase, and dynamometer studies.
- Method relies solely on observed on-road concentration measurements.

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ABSTRACT

We have applied the absolute principal component scores (APCS) receptor model to on-road, background-adjusted measurements of NOx, CO, CO_2, black carbon (BC), and particle number (PN) obtained from a continuously moving platform deployed over nine afternoon sampling periods in Seattle, WA. Two Varimax-rotated principal component features described 75% of the overall variance of the observations. A heavy-duty vehicle feature was correlated with black carbon and particle number, whereas a light-duty feature was correlated with CO and CO_2. NO_x had moderate correlation with both features. The bootstrapped APCS model predictions were used to estimate area-wide, average fuel-based emission factors and their respective 95% confidence limits. The average emission factors for NOx, CO, BC and PN (14.8, 18.9, 0.40 g/kg, and 4.3×10^{15} particles/kg for heavy duty vehicles, and 3.2, 22.4, 0.016 g/kg, and 0.19×10^{15} particles/kg for light-duty vehicles, respectively) are consistent with previous estimates based on remote sensing, vehicle chase studies, and recent dynamometer tests. Information on the spatial distribution of the concentrations contributed by these two vehicle categories relative to background during the sampling period was also obtained.

1. Introduction

In recent years, a number of investigators have deployed continuously moving vehicle platforms to characterize on-road pollutant concentrations and near road pollution gradients (Westerdahl et al., 2005; Pirjola et al., 2006; Rogers et al., 2006; Clements et al., 2009; Johnson et al., 2009; Wang et al., 2009; Durant et al., 2010; Hagler et al., 2010; Thornhill et al., 2010; Barzyk et al., 2012; Kozawa et al., 2012; Massoli et al., 2012; Padro-Martinez et al., 2012; Pirjola et al., 2012; Choi et al., 2013; Quiros et al., 2013; Brantley et al., 2014; Lahde et al., 2014; Patton et al., 2014; Riley et al., 2014) as well as larger scale urban gradients (Bukowiecki et al., 2003; Weijers et al., 2004; Isakov et al., 2007; Larson et al., 2009; Mohr et al., 2011; Aggarwal et al., 2012; Hu et al., 2012; Buzcu-Guven et al., 2013; Choi et al., 2013; Levy et al., 2014; Patton et al., 2015; Wu et al., 2015). In this sampling protocol, the mobile platform moves with traffic; for comparison, other mobile sampling protocols involve sampling at one fixed site for a defined period before moving to the next site. Continuously

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moving platforms provide broad spatial coverage, allowing for characterization of spatial variation in ambient pollution levels.

Continuously moving platforms have also been used to assess emissions from mobile sources in real world environments. "Vehicle chase" studies of exhaust plumes from individual vehicles have provided *in situ* estimates of emission factors for a variety of traffic-related pollutants as an alternative to traditional laboratory dynamometer testing, on-board vehicle emissions monitoring, and fixed-site sensing in tunnels and at roadsides. These include measurements made closely behind the vehicle tailpipe under relatively controlled driving conditions either on a test track or with a towed platform (Vogt et al., 2003; Cocker et al., 2004a, 2004b; Shah et al., 2004; Giechaskiel et al., 2005; Morawska et al., 2005; Carpentieri & kumar, 2011, Carpentieri et al., 2011; Kwak et al., 2014; Lee et al., 2015), and measurements made from chase vehicle platforms moving through urban traffic (Canagaratna et al., 2004; Kittelson et al., 2004; Kolb et al., 2004; Herndon et al., 2005; Jiang et al., 2005; Johnson et al., 2005; Shorter et al., 2005; Yli-Tuomi et al., 2005; Kittelson et al., 2006a, 2006b; Zavala et al., 2006; Durbin et al., 2008; Westerdahl et al., 2009; Zavala et al., 2009; Park et al., 2011; Kam et al., 2012; Liggio et al., 2012; Ning et al., 2012; Hudda et al., 2013; Jezek et al., 2015; Lau et al., 2015).

The studies cited above primarily address each pollutant separately. However, several investigators have further examined the multivariate correlations between simultaneously measured pollutants using principal components analysis (Bukowiecki et al., 2003; Riley et al., 2014). Multivariate receptor models traditionally applied to fixed site data have also been applied to mobile measurements of traffic-related pollutants, including absolute principal component scores (APCS) (Bruno et al., 2001), and positive matrix factorization (Thornhill et al., 2010; Buzcu-Guven et al., 2013; von der Weiden-Reinmuller et al., 2014). The impacts of vehicle emission regulations have also been assessed by combining mobile monitoring results with prior information on the traffic mix during sampling (Johnson et al., 2009; Liggio et al., 2012; Hudda et al., 2013; Kozawa et al., 2014).

Here we explore estimating area-wide average vehicle emission factors by applying the APCS receptor model to measurements obtained from a continuously moving platform. The model predictions are used to estimate average fuel-based emission factors by source-related feature within the study area. The pollutants we measured (NO_x, CO, CO_2, black carbon and particle number concentration) were chosen based on 1) the important contribution of traffic sources to these pollutant's emissions and resulting concentrations; 2) the relative importance of these species in distinguishing emissions from light duty versus heavy duty vehicles (Park et al., 2011; Dallmann et al., 2012, 2013; Pachon et al., 2012; Dallmann et al., 2013); 3) the required sensitivity and response time available from relatively low cost pollutant-specific monitors easily deployable on a mobile platform (Riley et al., 2014); and 4) the recognized health effects of pollutant exposure to ultrafine particle number (Devlin et al., 2014; Peters et al., 2015), black carbon (Janssen et al., 2012), CO(EPA, 2010), and NO_x (EPA, 2016).

2. Materials and methods

2.1. Study area

Sampling took place over a four to five hour interval on five afternoons in late September (20–21st & 23–25th) and four afternoons in early December (5,8,10,11) of 2012 in the Georgetown and South Park neighborhoods of the city of Seattle, Washington, USA (Fig. 1a). The sampling route encompassed approximately 30 km^2 and consisted of 38% major highways and 62% surface

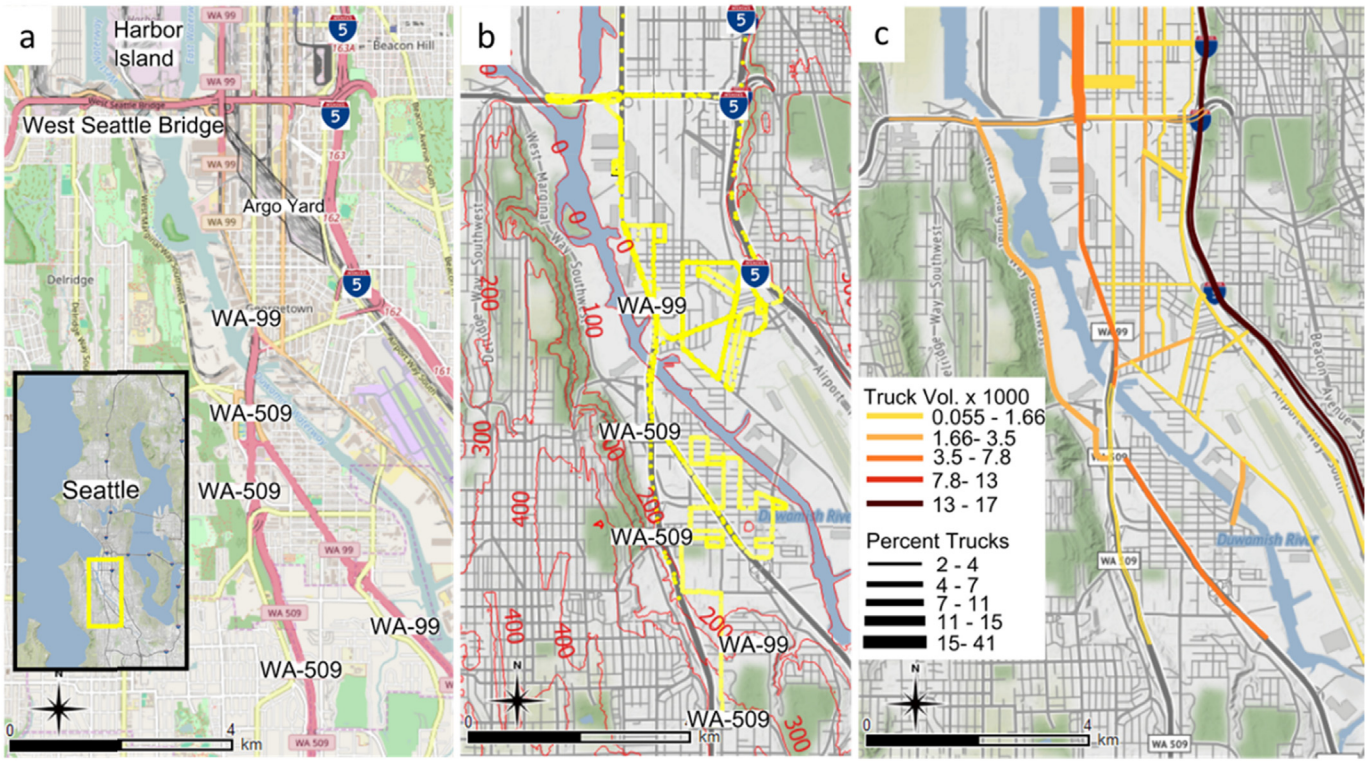

Fig. 1. Maps of the study area. (a) Location of the study area in Seattle (inset), major roadways, the Harbor Island docks and the Argo rail yard; (b) Mobile platform sampling route (yellow) and elevation contours in feet (red); (c) Average daily truck volumes and percent trucks along major truck routes.

streets (Fig. 1b). The sampling route passed under the West Seattle Bridge (never on the bridge itself), and along the local port access roads at this location in both the eastbound and westbound directions. The study area is mostly flat except that the southern end rises up approximately 110 m vertically over a horizontal distance of 5.2 km, or an approximate 2% slope (see Fig. 1b). The mobile monitoring route climbed up this hill on southbound SR 509 (divided, limited-access highway) and descended on surface streets.

The surrounding neighborhoods are a mixture of residential, commercial, and industrial areas with major transportation hubs for freight, major freeways (Interstate-5, Washington highways 99 and 509), rail spurs, and the Port of Seattle. The study area had the highest ranking among ten representative Seattle ZIP codes for air pollution and potential exposure to highly contaminated sites (Gould and Cummings, 2013). The study area is a major freight hub with significant truck traffic. Fig. 1c shows the average daily truck volumes and percent truck traffic on the major truck routes (Transportation 2016).

2.2. Mobile monitoring measurements

Ten second averages were collected simultaneously from five different instruments. The sampling route was driven two to three times on each sampling day, such that multiple visits were made to the same locations. The platform sampled at the speed of surrounding traffic on different types of roadways with a median speed of 26 km/h (25th percentile: 9 km/h, 75th percentile 44 km/h). Variability in vehicle speed resulted in 10-s measurements spaced at variable distances from each other (25th percentile: 48 m, 75th percentile: 179 m, mean: 119 m). Twelve percent of the time there was no distance between ten second intervals.

A complete description and diagram of the mobile platform is given in Riley et al., (2014). Briefly, the mobile monitoring platform consisted of a 2012 Ford Escape hybrid-electric vehicle. A GPS was mounted on the roof of the vehicle to record position and speed. Tables S−1 provides a full list of instrument measurements used in this analysis. Two sampling inlets were mounted on the roof rack on the driver's side of the vehicle in forward position leading to gaseous and particle measurement instrumentation, respectively. The sampling inlets were positioned above the vehicle boundary layer, and connecting tubes entered the vehicle through the otherwise sealed left rear window where they were connected to the instruments.

Particle loss was minimized by using stainless steel, copper, and conductive flexible tubing for the particle sampling inlet and tubing which connected to the assorted analyzers. We used a set of diffusion screens upstream of the PN counter that excluded particles that are nominally <50 nm. The cylindrical inlet tube geometry and sample flow rate corresponds to a Reynolds number of approximately 140. There is negligible particle loss under these conditions. Based on theoretical estimates of diffusion loss in a tube under laminar flow conditions, about 1.3% of 50 nm spherical particles would be lost during passage through the sample inlet, with lower losses for larger particles (Gormley and Kennedy, 1948−1950). Our screened instrument with 50 nm lower detection limit differs from other studies which used instruments that can detect smaller particles that have the potential for somewhat larger losses in the sampling inlet tubing.

The exhaust pipe from the vehicle's gasoline engine discharged on the right side low to the ground, away from the elevated, left-side air monitoring inlets. To further minimize the potential for self-pollution, the vehicle's gasoline engine would typically shut off when stopped at red traffic lights. In addition, we used our estimates of the contributions from light duty vehicles as presented in the Results section to further estimate the potential for self-pollution. Specifically, we first classified our vehicle's speed into two categories: < 2.3 km/h and ≥2.3 km/h to separate the idle or near idle conditions from those while moving along in traffic. The former category has the greatest self-pollution potential. Based on the PCA results derived for all days, we then asked whether there was a statistically significant difference between the idle or near-idle contributions from light duty vehicles (the CO-rich feature factor scores) versus the contributions at higher vehicle speeds. We found no significant difference between the two groups, with a mean score difference (idle − moving) of 0.031 [95% confidence interval = −0.021 to 0.083; p = 0.24].

The CO_2 meter exhibited a zero offset that could not be directly adjusted on the instrument itself. Therefore zero gas checks were made periodically before and after the study field measurements using a certified zero gas additionally passed through soda lime and drierite. The average offset for zero air determined from six comparisons between January and July 2013 was 122.6 ppm. We subtracted this zero offset from all field readings. However, the one minute average departures from a ten minute rolling background value relevant to our PCA analysis (discussed in section 3.1) are not affected by this zero offset adjustment. This is because the same CO_2 zero offset quantity is subtracted from both the measured concentration and the corresponding estimated background level. Further, the variability of the zero offset values was low. During the zero air checks, the one-minute average CO_2 concentrations were logged at rolling 10 s increments and these smoothed values exhibited a coefficient of variation during different comparisons of 0.54% for 4.17 min, 0.36% over 4.33 min, 0.72% over 2.5 min, 0.35% over 3.67 min, and 0.05% for a half minute interval.

Instrument quality control objectives and evaluation methods are summarized in Tables S−2. Automated flagging was implemented to censor data corresponding to instrument error codes, instrument operation out of specified parameters, or data otherwise missing (e.g., because the instrument rebooted itself, lost power, etc.). The time series for each pollutant was then manually reviewed and cross-checked with field technician notes. Details of the quality control procedures can be found in (Riley et al., 2016).

3. Theory/calculation

3.1. Smoothing and background adjustment

A number of approaches have been used to analyze continuously moving mobile platform data (Brantley et al., 2014). In principle, the mobile platform measurements include: 1) contributions from local on-road traffic emissions from individual vehicle plumes; 2) neighborhood scale background contributions from nearby traffic; and 3) urban scale background contributions affecting multiple neighborhoods within the urban area. The first two contributions are similar in spatial scale to the microscale (10−100 m), middle scale (100−500 m) and neighborhood scale (500 m to 4 km) representative monitoring domains as discussed by Watson (1997) (Watson et al., 1997). In contrast, the urban scale contributions can include variations across neighborhoods (4−100 km) and even a major contribution from global background levels in the case of CO_2.

We first smooth the time series each day by taking a moving block average of consecutive observations in a seventy second interval centered on a given 10-s observation. We estimate the background concentration ($B_{i,t,q}$, described below) associated with each 10 s observation period, and subtract this background value from the 70s moving block average concentration. We then combine all smoothed, background-adjusted observations across all valid 10-s observation periods over all days.

Specifically, we compute the background adjusted concentration of the ith species separately for each 10 s observation for each day as follows.

$$C^*_{i,t,q} = C_{i,t,q} - B_{i,t,q} \quad (1)$$

Where

$C_{i,t,q}$ is the 70-s moving average concentration between t-30s and t+30s centered on the 10-s period of interest t, ($t = 1,T_q$) on day q,

and

$B_{i,t,q}$ is a rolling minimum of $C_{i,t,q}$, centered on the 10-s period of interest on day q.

Specifically

$$B_{i,t,q} = \min\{C_{i,t-\tau}\ldots C_{i,t+\tau}\}_q \quad (2)$$

where $\tau = 300$ s.

The choice of $2\tau = 600$ s for the rolling minimum is motivated in part by the fact that over a 10 min time period the mobile platform on average traverses a spatial extent of ~4 km, the upper bound of the neighborhood monitoring scale. We apply this rolling minimum separately to each day's observations to adjust each day based on its conditions, and we then pool $C^*_{i,t,q}$ across all times ($t = 1,T_q$) and days ($q = 1, \ldots Q_d$) to create $C^*_{i,k}$ such that $k = 1, \ldots N$ where $N = T_q{}^*Q_d$. The $C^*_{i,k}$ are considered time-independent observations because: 1) they are adjusted for daily background values; and 2) the local on-road, vehicle-induced turbulence is the major dilution mechanism of the underlying exhaust plumes, independent of temporal changes in the less intense turbulence mixing associated with larger scale meteorology. The latter assumption might not be true if there are major temporal changes in the larger scale meteorology, but we purposely sampled in the afternoon during periods without frontal passages or other major changes in hourly wind speeds. The use of a rolling 5th percentile rather than a rolling minimum gave essentially the same results as the rolling minimum (results not shown). This is not surprising given that the rolling minimum we used is based on the one minute smoothed values reported every ten seconds in the moving window and that this smoothed minimum value represents the 2nd percentile in that moving window. Finally, we form a set of adjusted concentrations, $C^{adj}_{i,k}$, by removing entire samples from the initial set of $C^*_{i,k}$ using the following criteria: 1) the value of $C^*_{CO_2,k}$ is less than 5 ppmv, the instrument precision limit; 2) value of $C^*_{i,k}$ is above the 95th daily percentile value for a given pollutant The first criterion is applied to more confidently ensure the presence of individual combustion exhaust plumes at concentrations above the local CO_2 background and is less stringent than the 20 ppmv criterion previously used in vehicle chase studies[56]. We also analyzed the data excluding step 2, and it did not make an appreciable difference to the results.

3.2. Absolute principal component score (APCS) model

We chose to use the APCS receptor model. Other multivariate receptor models such as PMF could also be used. However, PMF requires specification of the species measurement uncertainties, which in this case are more complex than usually encountered with raw concentration data. We are analyzing the departures from baseline, which is not only a difference between two measured values, but also a value whose precision varies in a complex way with the raw species concentration. Therefore, although PMF is another potentially viable option, we feel that including it here is beyond the scope of this paper. We also prefer the fact that the species weights in PCA are based on their overall variance rather than their measurement uncertainty, potentially reducing the influence of meteorologically driven day to day variation in the species concentration in these on-road plumes.

The APCS model was first described by Thurston (1985) (Thurston and Spengler, 1985) for fine particle mass apportionment and subsequently used to apportion individual VOC species (Miller et al., 2002; Guo et al., 2004). Principal components analysis is first applied to the standardized, adjusted concentrations for the *m*-species across all days, specifically

$$Z_{i,k} = \frac{C^{adj}_{i,k} - \overline{C}^{adj}_i}{\sigma_i} \quad (i = 1, \ldots m; k = 1, \ldots N) \quad (3)$$

where $C^{adj}_{i,k}$ has mean $\overline{C}^{adj}_i$ and standard deviation σ_i. We chose to use the standardized concentrations rather than the raw, adjusted concentrations to more equally weight each species in the final solution. We retain p ($p \leq m$) principal components based upon their having eigenvalues > 0.9 and then apply a Varimax rotation to these components.

Absolute principal component scores (APCS) for the Varimax rotated components are calculated from the scores, $S_{j,k}$, for the kth observation of the jth component as follows:

$$APCS_{j,k} = S_{j,k} - (S_0)_j \quad (i = 1, \ldots p) \quad (4)$$

where $S_{j,k}$ are the scores derived from the $Z_{i,k}$ and $(S_o)_j$ is the predicted value of the zero vector using the rotated PCA model. The $APCS_{j,k}$ are then regressed against the $C^{adj}_{i,k}$.

$$C^{adj}_{i,k} = (b_0)_i + \sum_{j=1}^{p} b_{i,j}\left(APCS_{j,k}\right) + \varepsilon_{i,k} \quad (5)$$

The intercept in Equation (5) is the contribution to the adjusted values from sources unaccounted for in the PCA[73]. The predicted concentration of pollutant i ($\widehat{Y}_i$) contributed by feature j to the kth sample is then defined by equation (6).

$$\widehat{Y}_{i,j,k} = b_{i,j}\left(APCS_{j,k}\right) \quad (6)$$

3.3. Fuel-based emission factors

The average emission factor (EF) values were computed as follows:

$$EF_{i,j} = \frac{\alpha(W_c)_j}{N}\sum_{k=1}^{N}\left\{\frac{\widehat{Y}_{i,j,k}}{\left(\widehat{Y}_{CO_2}\right)_{j,k} + \left(\widehat{Y}_{CO}\right)_{j,k}}\right\} \quad (7)$$

where

$EF_{i,j}$ is the average fuel-based emission factor in grams of pollutant i per kg of fuel burned for source-related feature j;
N is the total number of samples (=9541)
$(W_c)_j$ is the carbon weight fraction of the fuel corresponding to the jth source-related feature;

$(W_c)_j$ is a units conversion factor (=1 for CO, black carbon and NO_x concentrations reported in units of $\mu g/m^3$ and = 10^{12} for

particle number concentration reported in units of number/cm^3).

We applied a blocked bootstrap to the above model (equations (3)–(7)) in order to estimate the uncertainties in EF_{ij}. The blocked bootstrap was chosen to minimize potential autocorrelations in the $C_{i,k}^{adj}$ possibly due to correlated background values not accounted for in our model. We randomly sampled with replacement from non-overlapping blocks with a fixed block size of 1780 consecutive observations, repeating the bootstrap procedure 10,000 times. Optimal univariate block sizes were determined using the "b.star" function within the "np" package in R (Politis and White, 2004; Patton et al., 2009). The bootstrap block size was chosen to be the maximum of the set of five univariate block sizes, one estimate for each of the five species. Matching of the bootstrapped features to their respective base case features was done by using the corresponding EF_{ij} for NOx, BC and CO. Our reported 95% confidence limits of EF_{ij} were then taken from the distribution of average EF_{ij} estimated from each of the 10,000 bootstrapped values.

3.4. Comparing feature scores with truck traffic

For visual comparisons of the feature scores to truck traffic data obtained from Seattle Department of Transportation (Fig. 1c), the scores for each feature were spatially binned into 200 × 200 m grid cells. The median values were then computed for each cell. We chose the median to minimize the influence of a few relatively high concentration plumes within a given cell. The spatial differences in the factor scores reflect the on-road spatial differences in these median concentrations in the heavy duty or light duty vehicle plumes across grid cells over the study period. The spatial grid represents a much higher data density than the traffic count data depicted by the roadway segments, and can be used to identify hotspots that might not be evident in the truck volume data. Spatial binning was performed using the 'raster' and 'OpenStreetMap' packages and R version 3.2.0.

To evaluate the distribution of scores for each Seattle Department of Transportation (SDOT) roadway segment with available traffic count data, we first generated a 50 m buffer around the SDOT roadway segments shown in Fig. 1c and then spatially joined our individual roadway measurements with the SDOT features. This appended the truck volume and the percent truck information in the SDOT layers to our dataset (see Fig. 1b and c). In our case, these roadways buffers did not overlap substantially with other roadway buffers. We then examined the distribution of scores associated with the SDOT data, specifically the daily truck volumes and percent trucks. We calculated the linear least squares regression coefficients and pearson's r correlation coefficients between the traffic variables and the median scores on the roadway segments. GIS analysis was performed using QGIS version 2.16.3, regression analysis performed in R (3.2.0).

4. Results

4.1. Observed and background-adjusted concentrations

Table 1 summarizes the values of $C_{i,t,q}$, $B_{i,t,q}$ and $C_{i,k}^{adj}$. The 70-s smoothed concentrations of NO_x, BC, CO, PN have greater variation about their means than CO_2. The estimated background values as a percentage of the average observed values are lower for NOx, BC and PN than they are for CO and CO_2.

4.2. Varimax rotated components

Varimax rotated principal component analysis of the adjusted concentration data ($C_{i,k}^{adj}$) resulted in two features as shown in Table 2. The Varimax rotated factor loadings for these two features are also shown in Table 2 along with the initial eigenvalues and the corresponding percent variance for each of the initial principal component features prior to rotation. Both BC and PN are heavily loaded on the first feature and therefore we initially refer to this as the "BC-rich" feature. In contrast, CO and CO_2 are the main species associated with the second feature. We refer to this feature as "CO-rich".

The Varimax-rotated absolute principal component scores (overall and by season) are plotted by feature on a map of the study area in Fig. 2 (the map was generated using OpenStreetMap, R Package version 0.3.1).

4.3. Estimated fuel-based emission factors

Table 3 summarizes the estimated average fuel-based emission factors predicted by equation (7) for both features as well as the corresponding 95% confidence limits estimated from the blocked bootstrap. Also shown are the average light and heavy duty vehicle emission factors reported by other U.S. field studies using either mobile monitoring of general traffic, chase vehicles, or near-road fixed site measurements.

In addition, we have compared our estimated emission factors

Table 2
Varimax-rotated Principal Component Loadings based on $C_{i,k}^{adj}$.

	BC-rich feature	CO-rich feature
NO_x	0.62	0.69
BC	0.88	0.09
CO	0.15	0.74
PN	0.87	0.26
CO_2	0.19	0.76
Initial Eigenvalues	2.78	0.94
% Variance After Varimax Rotation	39.3	35.1

Table 1
Summary of mean concentrations.

Species	One-minute smoothed concentrations ($C_{i,t,q}$)	Background concentrations from Eqn. (2) ($B_{i,t,q}$)	Background adjusted & trimmed concentrations[b] ($C_{i,k}^{adj}$)
NO_x (ppb)	56.3	11.5	36.4
	(7.0, 164)[a]	(0.1, 56.8)	(5.7, 96.3)
BC (ng/m^3)	2093	603	1110
	(431.5, 6053)	(39, 1974)	(271, 3348)
CO (ppm)	1.57	1.18	0.30
	(0.65, 2.58)	(0.40,1.71)	(0.05,0.77)
PN (#/cm^3)	2.02×10^4	5.19×10^3	1.13×10^4
	(3.99×10^3, 5.79×10^4)	(5.42×10^2, 1.29×10^4)	(1.74×10^3, 3.18×10^4)
CO_2 (ppm)[c]	557.1	524.0	27.8
	(494.1, 644.0)	(484.0, 592.6)	(6.2, 74.6)

[a] 5th and 95th percentile values.
[b] Background adjusted values above 95th %tile were removed.
[c] Adjusted for zero offset based on quality checks with zero-gas (see section 2.2).

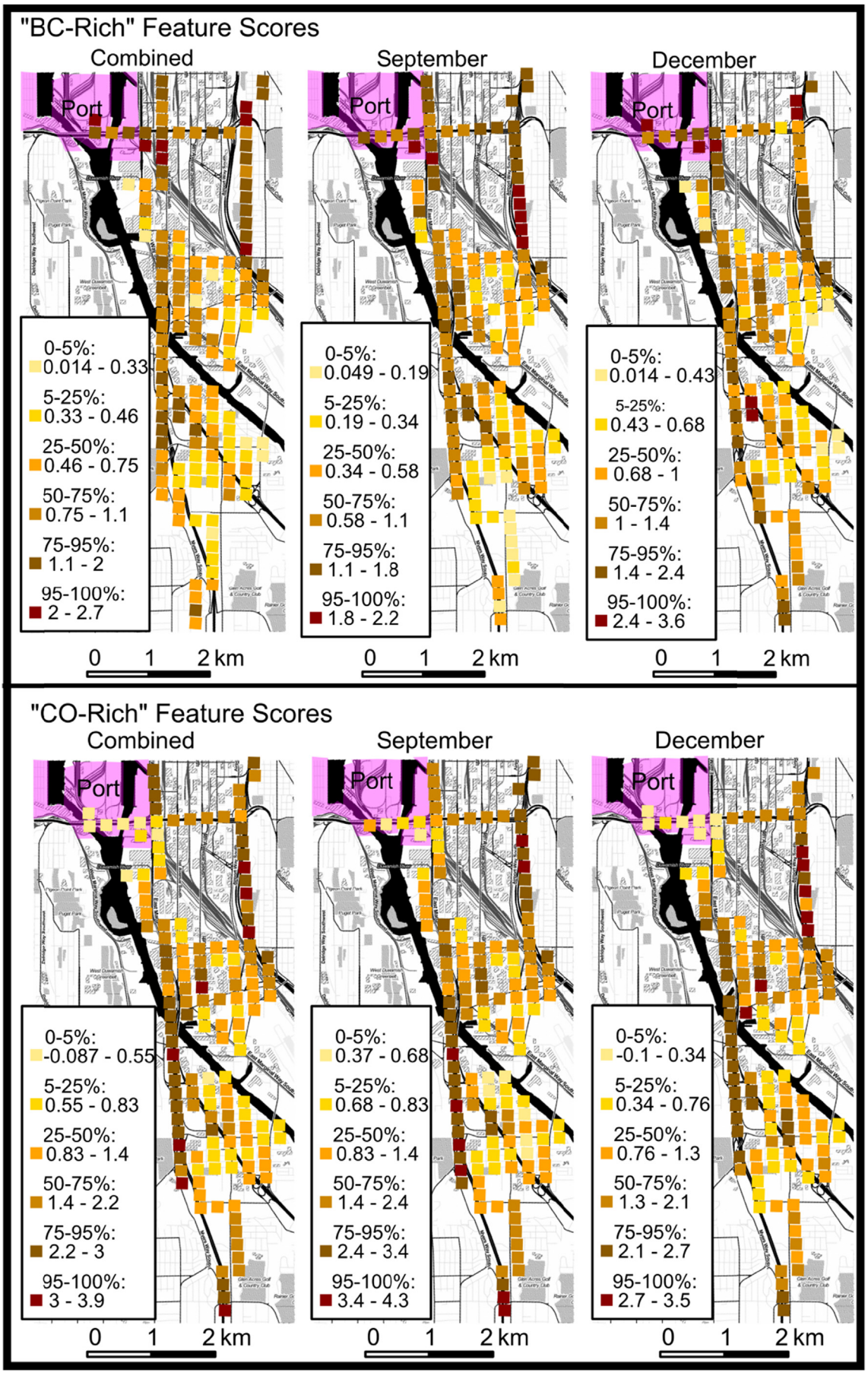

Fig. 2. Map of varimax-rotated absolute principal component scores (see also equation (4) in section 3.2). The color scale represents quantiles of the resultant cell medians. The black area is water. The pink area in the upper left-hand corner is the southern tip of Harbor Island, part of the Port of Seattle. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 3
Average derived fuel-based emission factors compared to recent U.S. Field studies.[a]

Study	Year	Sampling type	NOx (g/kg)	CO (g/kg)	BC (g/kg)	PN (10^{15}/kg)
Heavy Duty Vehicles						
BC-rich feature (this work)[b]	2012	Moving in traffic	14.8 [9.9–21.9]	18.9 [8.0–35.3]	0.40 [0.29–0.58]	4.3 [2.9–6.2]
Bishop et al., 2015,[c,d]	2013	Remote sensing across road	20.7, 20.3 [19.1–22.3], [18.9–22.1]	2.3, 5.1 [1.5–3.1], [4.7–5.5]	0.02, 0.23 [0.014–0.026], [0.17–0.29]	–
Preble et al., 2015,[e]	2013	Fixed site near road	15.4 [14.5–16.3]	–	0.28 [0.23–0.33]	2.5 [2.0–3.0]
Hudda et al., 2013,[f]	2011	Moving in traffic	15,16 <9.2, 10>	–	0.41, 1.33 <0.21, 0.33>	4.2, 5.2 <3.4, 3.1>
Liggio et al., 2012	2010	Moving in traffic	–	–	0.51 <3.1>	–
Dallmann et al., 2012	2010	Fixed site in tunnel	28 [26.5–29.5]	–	0.54 [0.47–0.61]	–
Bishop et al., 2012a,[c,d]	2010	Remote sensing across road	47.8, 29.2 [46.6–49.0], [27.6–30.8]	–	–	–
Park et al., 2011	2007	Vehicle chase	34	36	0.5	4.5
Johnson et al., 2009,[g]	2007	Moving in traffic	14.0 <5.5>	–	–	3.2 <2.8>
Ban-Weiss et al., 2010,[h]	2006	Fixed site in tunnel	–	–	–	3.3 [2.0–4.6]
Ban-Weiss et al., 2008	2006	Fixed site in tunnel	40.0 [37–43]	–	0.92 [0.85–0.99]	–
Light Duty Vehicles						
CO-rich Feature (this work)[b]	2012	Moving in traffic	3.2 [2.8–3.6]	22.4 [19.7–25.0]	0.016 [0.011–0.021]	0.19 [0.13–0.25]
Hudda et al., 2013	2011	Moving in traffic	3.8 <1.4>	–	0.07 <0.05>	0.43 <0.26>
Kozawa et al., 2014,[j]	2009–2011	Moving in traffic	2.7, 4.0 <0.4, 0.3>	24, 27 <1.6, 3.1>	0.015, 0.067 <0.011, 0.031>	0.28, 0.58 <0.31, 0.30>
Dallmann et al., 2013	2010	Fixed site in tunnel	1.90 [1.82–1.98]	14.3 [13.6–15.0]	0.010 [0.008–0.012]	–
Bishop et al., 2012b,[d]	2010	Remote sensing across road	4.1 [3.9–4.3]	19.4 [16.8–22.0]	–	–
Liggio et al., 2012	2010	Moving in traffic	–	–	0.12 <0.08>	–
Bishop et al., 2010,[d,k]	2008	Remote sensing across road	4.0, 4.6, 5.9 [3.8–4.2], [4.4–4.8], [5.1–6.7]	16.6, 20.0, 21,4 [14.6–18.6], [18.0–22.0], [20.4–22.4]	–	–
Park et al., 2011	2007	Vehicle chase	9.4	47	0.06	0.60
Ban-Weiss et al., 2008	2006	Fixed site in tunnel	3.0 [2.8–3.2]	–	0.026 [0.022–0.030]	–
Ban-Weiss et al., 2010,[h]	2006	Fixed site in tunnel	–	–	–	0.39 [0.25–0.53]

[a] [ ] = 5th-95th % confidence limits, < > = reported standard deviation.
[b] Confidence limits estimated via blocked bootstrap (see section 3.3 for details).
[c] Values for separate measurements at the Port of Los Angeles and at a Northern California I-5 weigh station.
[d] Confidence limits based on reported standard error.
[e] Values for 2013 drayage trucks at the Port of Oakland.
[f] Separate values for (I-710), (other freeways).
[g] Values for 2006/2007.
[h] For particle diameters > 3 nm.
[j] Lowest and highest mean values and their corresponding standard deviations for multiple campaigns between Sept., 2009 and Sept., 2011.
[k] Values for San Jose, Fresno and West Los Angeles.

with the recent dynamometer tests reported by May et al. (2014) Their study included 51 light duty vehicles and 5 heavy duty vehicles representative of a typical vehicle fleet age distribution. The comparison is shown in Table 4.

As discussed in section 3.4, we have also compared the spatial differences across the study area for each feature score with relevant traffic information, namely daily truck volumes and percent trucks. The results are summarized in Table 5. Additional details about results of the linear regression model are included in the Supplemental Information.

5. Discussion

There have been several studies employing multivariate analysis of air pollution measurements obtained from a continuously moving mobile platform (Bruno et al., 2001; Bukowiecki et al., 2003; Thornhill et al., 2010; Buzcu-Guven et al., 2013; Riley et al., 2014; von der Weiden-Reinmuller et al., 2014). To our knowledge, Thornhill and co-workers[8] conducted the only such study to estimate fuel-based emission factors from motor vehicles. Their study used the PMF model and was limited to a 3 h drive in Mexico City, making it difficult to compare their derived factors with other U.S. studies. As such, we did not compare their results to those from this work.

The previous studies summarized in Table 3 all used univariate approaches to estimating emission factors. These studies include near-road, fixed-site measurements (Ban-Weiss et al., 2008; 2010; Bishop et al., 2010, Bishop et al. 2012a,b; Dallmann et al., 2012, 2013; Bishop et al., 2015; Preble et al., 2015), and on-road, chase vehicle measurements (Park et al., 2011) and have the inherent strength of directly matching their measurements with specific vehicles and driving conditions. However, they do not necessarily capture the variability across a broader set of driving conditions or, in the case of vehicle chase studies, a broader population of vehicles. Several previous on-road mobile monitoring studies have attempted to address this issue by including all on-road observations across a given region after adjustments for background levels (Johnson et al., 2009; Liggio et al., 2012; Hudda et al., 2013; Kozawa et al., 2014). To estimate emission factors, they have combined their background-adjusted measurements post-hoc with on-road fuel

Table 4
Comparison of our estimated emission factors with those from the dynamometer study of May et al. (2014).

Vehicle type	Study	Average emission factor [95% c.i.]		
		CO (g/kg)	NOx (g/kg)	BC (g/kg)[a]
Light duty	This study	22.4 [19.7–25.0]	3.2 [2.8–3.6]	0.016 [0.011–0.021]
	May et al. (51 LEV vehicles)[b]	26.4 [19.1.-33.7]	2.8 [1.8–3.9]	0.017 [0.013–0.022]
Heavy duty	This study	14.8 [9.9–21.9]	18.9 [8.0–35.3]	0.40 [0.29–0.58]
	May et al. (2 vehicles with a DPF)[c]	0.26	8.2	0.001
	May et al. (3 vehicles without a DPF)[b]	8.8 [5.6–12.0]	23.6 [17.2–30.1]	1.18 [0.13–0.24]

[a] May et al. report EC values.
[b] Confidence intervals estimated from computed standard errors.
[c] Confidence intervals were not estimated due to the small number of samples.

Table 5
Associations between truck traffic and median feature scores across the study area.

	Adjusted R^2 [pearson r]	
	Daily truck volumes	Percent trucks on roadway
BC-rich feature	0.41 [0.66]	0.05 [-0.035]
CO-rich feature	0.08 [0.36]	0.02 [-0.27]

use or traffic information in order to separate the contributions of light duty from heavy duty vehicles. Our study is different in that it makes no such assumptions about on-road traffic or fuel use.

Our results agree well with the NOx, PN, CO and BC light duty emission factors reported in the field studies summarized in Table 3 as well as with the dynamometer-based laboratory study reported by May and colleagues (May et al., 2014). They are also consistent with the heavy duty emission factors reported in other studies after accounting for the degree of exhaust emission controls. Based on the estimated confidence intervals, our average heavy-duty vehicle NOx emission factor of 14.8 g/kg is no different than that reported by the most recent studies done in 2011 and later (Hudda et al., 2013; Bishop et al., 2015; Preble et al., 2015), but slightly lower than that from studies done between 2006 and 2010 (Ban-Weiss et al., 2008; Park et al., 2011; Bishop et al., 2012a; Dallmann et al., 2012). This is consistent with the fact that heavy-duty NOx emissions have decreased over this time period due to improved emission control technologies (McDonald et al., 2012; Xing et al., 2013; Lu et al., 2015). Our average heavy-duty emission factors for PN and BC (4.3 and 0.4 g/kg, respectively) are generally consistent with those reported by previous studies.

Our estimates of the heavy duty CO and BC factors are higher than those derived from remote sensing by Bishop and co-workers (Bishop et al., 2015) (Preble et al., 2015) for drayage trucks equipped with diesel particle filters (DPF) in the Port of Los Angeles (see Table 3), and also higher than those derived from dynamometer-testing of DPF-equipped trucks by both Tsai and colleagues (Tsai et al., 2011) and by May and colleagues (May et al., 2014).

The higher BC emission factor we observed compared to some of these other studies is consistent with the absence of diesel particle filters on the majority of the truck fleet at the time in our study area. At the time of our study only 24 percent of trucks entering the Port of Seattle were equipped with either diesel oxidation catalysts (DOCs) or diesel particle filters (DPFs) (Alliance, 2015). This is similar to the estimated percentage of all trucks newer than 2007 across the entire U.S. at the time of our study-30% for diesel trucks (IHS Automotive, 2014), the Port of Los Angeles being a notable exception due to stricter emission regulations. Further, our estimates are in reasonable agreement with the dynamometer-based heavy duty emission factors reported by May and colleagues (May et al., 2014) for vehicles that were not equipped with a diesel particle filter (see Table 4). Our CO emission factor is reasonably consistent with those from May et al. (2014) for trucks without a DPF considering the fact that two of these three trucks were also not equipped with a DOC (see Table 4).

Our exclusion of the largest departures from background (>daily 95th percentile) only modestly affected our emission factor estimates, with the possible exception of the slightly larger estimate for PN for the light duty vehicles using the untrimmed data (see Tables S–3). As a general rule, we prefer exclusion of values exceeding the daily 95th percentile because multivariate methods can potentially be biased by relatively few outliers that may not generally represent the entire vehicle fleet, although in this case it does not appear to make much difference to our overall estimates.

Although the above results strongly suggest that we have derived separate heavy-duty and light-duty vehicle profiles, i.e., fuel-based emission factors, we have also explored the spatial differences in the derived feature scores as an additional check on these attributions. It was not clear *a priori* whether truck volumes or the percentage of trucks was a better predictor of the median heavy-duty feature scores in each cell shown in Fig. 2. However, based on the correlations we report in Table 5 it appears that truck volumes are a better predictor. More importantly, the fact that the spatially varying daily truck volumes in our study area are correlated with the heavy-duty vehicle feature and not the light-duty vehicle feature provides additional evidence that we have correctly labeled these features. Other factors such as driving mode (idle, acceleration, cruise) obviously also affect these scores, but these factors were not assessed because specific vehicles were not intentionally followed by our platform. Traffic congestion and the accompanying truck traffic acceleration from stop would contribute higher levels of emissions to the heavy-duty feature found at key intersections than would a measure such as truck volume which does not take into account the driving mode. However, congestion metrics were not available at the required spatial resolution and were therefore not included in this analysis.

It is interesting to note the location of persistently elevated "BC-rich" scores in Fig. 2, specifically along Interstate 5, SW Spokane Street below the West Seattle Bridge, and WA-99 both near the port and at the intersection with WA-509, including the 1st Avenue South Bridge. This latter location was also identified as being a 'hot spot' for diesel exhaust as identified by measurement of 1-nitropyrene by Schulte et al., (2015). The relatively high values of both feature scores along Interstate 5 are consistent with the high overall traffic volumes along this roadway. The relatively high "CO-rich" scores along Highway 509 are consistent with the higher vehicle specific power associated with ascending this road grade (Zhang and Frey, 2006; Frey et al., 2010).

However, the development of a spatial model to represent the spatial variation of these features is not the focus of this paper. Rather, we have developed an alternative approach to estimating area-wide average emission factors. The map shown in Fig. 2 should be interpreted with caution as it represents only one realization (the base case) of the underlying spatial distribution of the derived feature scores over a limited time period. Given that we exclude the estimated background concentrations from this analysis, our model should not be considered a general representation of the observed, unadjusted concentrations. Clearly other sources can also contribute to the estimated background values such as, for example, the distinctly different emissions from relatively few high emitting light duty vehicles as reported by Tan and colleagues (Tan et al., 2016). The one possible exception is NOx whose average estimated background concentration in this study is a relatively small proportion of its average on-road departures from background. Our approach could provide an independent check on the sub-grid scale spatial allocation of fleet-wide NOx emissions used in urban air quality models (McDonald et al., 2012).

6. Conclusions

We have derived separate estimates of fuel-based emission factors for light-duty and heavy-duty vehicles from measurements taken over space using a continuously moving mobile platform. Our estimates are in good agreement with recent studies using a variety of methods, including stationary remote sensing and vehicle chase platforms, continuously moving mobile platforms, and recent dynamometer results. Our use of a multivariate model, traditionally applied for source apportionment, allows separate estimates of both light-duty and heavy-duty vehicle emission factors based solely on the observed concentrations without reliance on independent traffic information. Information on the spatial distribution of the pollutant concentration of on-road plumes from these two vehicle categories during the sampling period is also obtained.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2016.12.037.

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