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Examining the representativeness of home outdoor $PM_{2.5}$, EC, and OC estimates for daily personal exposures in Southern California

Regina E. Ducret-Stich · Ralph J. Delfino · Thomas Tjoa · Armin Gemperli · Alex Ineichen · Jun Wu · Harish C. Phuleria · L.-J. Sally Liu

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Abstract Recent studies have linked acute respiratory and cardiovascular outcomes to measurements or estimates of traffic-related air pollutants at homes or schools. However, few studies have evaluated these outdoor measurements and estimates against personal exposure measurements. We compared measured and modeled home outdoor concentrations with personal measurements of traffic-related air pollutants in the Los Angeles air basin (Whittier and Riverside). Personal exposure of 63 children with asthma and 15 homes were assessed for particulate matter with an aerodynamic diameter less than 2.5 μm ($PM_{2.5}$), elemental carbon (EC), and organic carbon (OC) during sixteen 10-day monitoring runs. Regression models to predict daily home outdoor $PM_{2.5}$, EC, and OC were constructed using home outdoor measurements, geographical and meteorological parameters, as well as CALINE4 estimates at outdoor home sites, which represent the concentrations from local traffic sources. These home outdoor models showed the variance explained (R^2) was 0.97 and 0.94 for $PM_{2.5}$, 0.91 and 0.83 for OC, and 0.76 and 0.87 for EC in Riverside and Whittier, respectively. The $PM_{2.5}$ outdoor estimates correlated well with the personal measurements (Riverside R^2=0.65 and Whittier R^2=0.69). However, excluding potentially inaccurate samples from Riverside, the correlation between personal exposure to carbonaceous species and home outdoor estimates in Whittier was moderate for EC (R^2=0.37) and poor for OC (R^2=0.08). The CALINE4 estimates alone were not correlated with personal measurements of EC or other pollutants. While home outdoor estimates provide good approximations for daily personal $PM_{2.5}$ exposure, they may not be adequate for estimating daily personal exposure to EC and OC.

Keywords Air pollution · Elemental carbon · Organic carbon · Exposure modeling · Traffic exhaust

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R. E. Ducret-Stich (✉) · A. Ineichen · H. C. Phuleria · L.-J. S. Liu
Department of Epidemiology and Public Health, Environmental Exposure Sciences, Swiss Tropical and Public Health Institute, P.O. Box 4002, Basel, Switzerland
e-mail: regina.ducret@unibas.ch

R. E. Ducret-Stich · A. Gemperli · A. Ineichen · H. C. Phuleria · L.-J. S. Liu
University of Basel,
Basel, Switzerland

R. J. Delfino · T. Tjoa · J. Wu
Department of Epidemiology, School of Medicine,
University of California,
Irvine, CA, USA

L.-J. S. Liu
Department of Environmental and Occupational Health Sciences,
University of Washington,
Seattle, WA, USA

Introduction

Numerous epidemiological studies have found associations between outdoor air pollution and adverse respiratory outcomes (Brunekreef and Holgate 2002; Pope and Dockery 2006). In particular, traffic-related air pollution has been found to affect respiratory health (Jansen et al. 2005; McCreanor et al. 2007; Meng et al. 2007) especially in children (Delfino et al. 2009; Koenig et al. 2005; Ryan et al. 2005; Ryan and LeMasters 2007; Sarnat and Holguin 2007; Trasande and Thurston 2005). In the Los Angeles (LA) area, many houses and schools are close to major roads and freeways, increasing children's exposure to air pollution from traffic (Künzli et al. 2003). The Southern California Children's Health Study showed positive associations between exposure to long-term traffic-related air pollution and asthma prevalence in a pediatric cohort (Gauderman et al. 2005; McConnell et al. 2006) and negative effects on lung growth independent of background air pollution levels (Gauderman et al. 2007). A limited number of studies also linked acute respiratory effects to personal $PM_{2.5}$ exposure in children and adults (Delfino et al. 2004, 2006, 2008; Ebelt et al. 2005; Koenig et al. 2005; Strand et al. 2006; Trenga et al. 2006).

Due to the significant intra-urban spatial variation of traffic air pollution (Goswami et al. 2002; Liu et al. 2007; Zhu et al. 2002), exposure proxies or land-use regression models have been used to estimate long-term personal exposure to traffic pollutants in urban areas (Hoek et al. 2008; Jerrett et al. 2005). Despite broad applications of these methods, only Van Roosbroeck et al. (2006, 2007, 2008) have evaluated certain exposure proxies against personal exposure to soot. The land-use regression models have yet to be validated against personal exposure measurements. Additionally, it remains unclear whether these models are useful for estimating short-term (daily) personal exposure to traffic pollutants.

The present study aimed to address this issue by evaluating the ability of daily home outdoor air pollutant estimates from land-use regression models to represent daily personal exposure to air pollutants, including $PM_{2.5}$, EC, and OC. To our knowledge, this is the first study using personal OC measurements to validate model estimates for personal exposure to OC. We constructed land-use regression models for traffic pollutants outside homes using geographical parameters and outdoor pollutant measurements. CALINE4 model estimates for traffic pollutants were also incorporated and tested. These model predictions were compared against personal measurements in two cities with different ambient pollution source characteristics.

Method

Study design

This work was part of a panel study evaluating acute health outcomes of 63 children with asthma living in the cities of Riverside and Whittier in the LA air basin (Delfino et al. 2006, 2008; Fig. 1). Riverside is a smog receptor site downwind from urban LA (Kim et al. 2002; Na et al. 2004). There, 31 subjects were followed periodically from August through mid-December 2003. Whittier is a site immediately downwind of vehicular emission sources. There, 32 subjects were followed periodically from July through November 2004. In each city, eight 10-day exposure monitoring periods (runs) were conducted, consisting of four subjects with concurrent personal monitoring of $PM_{2.5}$ mass, EC, and OC. In each run, one subject's residence and a central site were monitored and modeled for concurrent $PM_{2.5}$ mass, EC, and OC (Fig. 1).

Exposure measurements

Personal exposure

The following measurements were made in each subject over the 10-day run. We measured 1-min average $PM_{2.5}$ using the personal DataRAM (MIE pDR-1200; Thermo Electron Corp., Franklin, MA, USA). The pDR is an integrated nephelometer with a 2.5-μm sharp-cut cyclone (BGI model GK 2.05, KTL cyclone, GI Inc., Waltham, MA, USA) operated at 4 L/min. It was carried by each subject in a specially designed soundproof backpack with separate compartments for the subject's school books. $PM_{2.5}$ mass was also collected on a 37-mm (back-up) quartz filter (Whatman Inc, Florham Park, NJ, USA), which was placed downstream of the pDR and collected particles over each of ten 24-h sampling periods. These filters were pre-baked prior to sampling to remove any carbon. Analysis for EC and OC was done using the thermal manganese dioxide oxidation protocol (Fung et al. 2002). A HOBO logger (Onset Computer Corp., Pocasset, MA, USA) was used to record 1-min relative humidity (RH) and temperature. All pDR data were adjusted for the effect of RH (Wu et al. 2005a). Continuous (1-min) and gravimetric (24-h) measurements of the personal sampler were validated by comparing them against each other and against reference methods (Chakrabarti et al. 2004). Continuous $PM_{2.5}$ measurements from the pDR were compared with collocated measurements from a Beta Attenuation Monitor (R^2=0.75, corrected for RH). Gravimetric measurements from the back-up filter of the pDR were compared with gravimetric measurements from a Partisol sampler (R^2=0.93) and with the 24-h average from the Beta Attenuation Monitor (R^2=0.71). The R^2 between the filter-based personal $PM_{2.5}$ and the continuous personal $PM_{2.5}$ (corrected for RH) was 0.56.

Fixed-site measurements

Concurrent with the personal measurements, simultaneous indoor and outdoor monitoring was conducted at one home and at a central site station during each of the sixteen 10-day runs. The central site in Riverside was the South Coast Air Quality Management District (SCAQMD) monitoring site, while in Whittier it was set up by us at one of the subjects' residences (Fig. 1). At all of these indoor, outdoor,

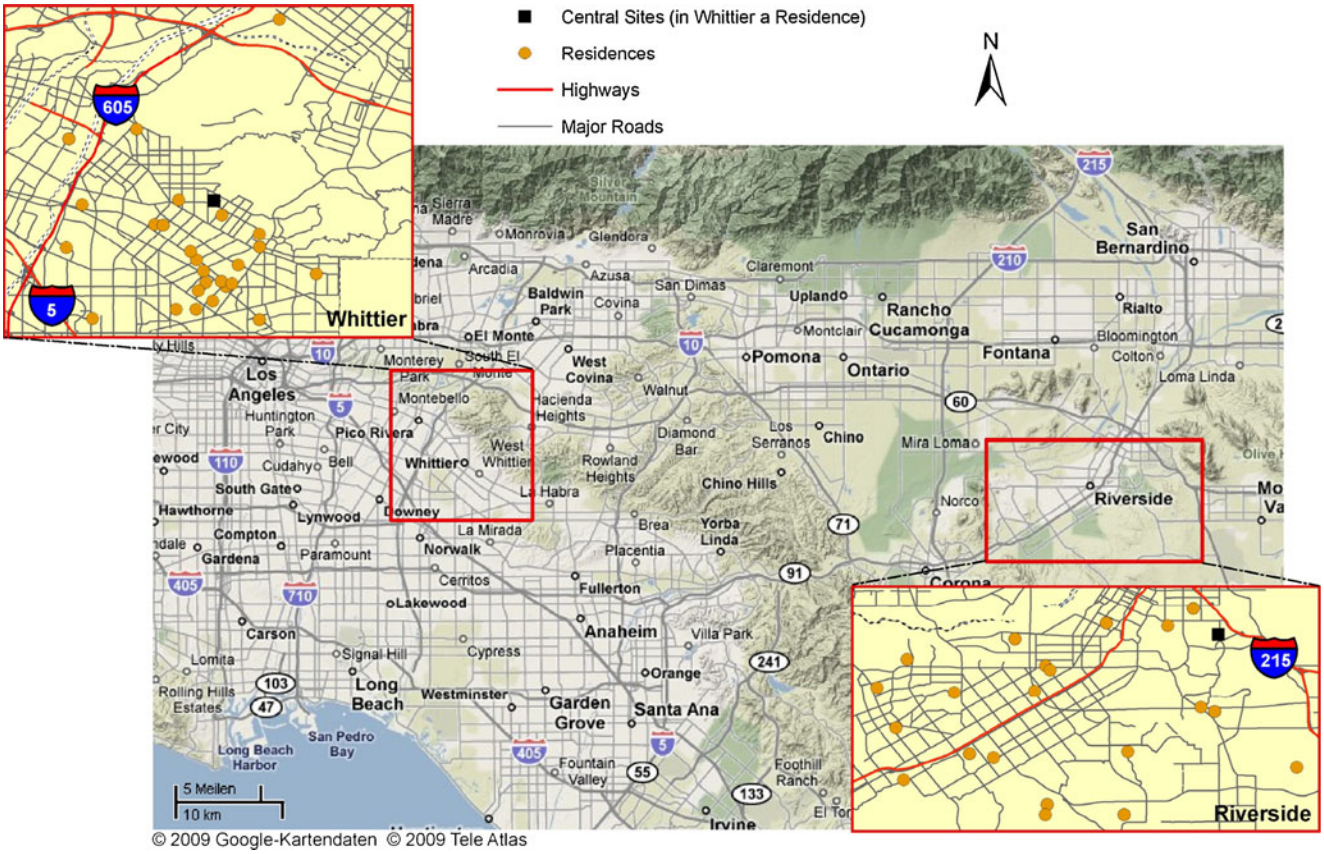

Fig. 1 Study area: Whittier and Riverside, in the Los Angeles air basin of Southern California. Detailed maps are not to scale

and central sites, 24-h $PM_{2.5}$ measurements were collected on Teflon and quartz filters using Harvard Impactors (Air Diagnostics and Engineering, Inc., Naples, ME, USA; Liu et al. 2003). Mass measurements were conducted with the Teflon filters using standard gravimetric methods. All quartz filters were analyzed for EC and OC using the thermal manganese dioxide oxidation protocol as with the personal filters (Fung et al. 2002).

Geocoding and traffic variables

Residences and schools were geocoded using the TeleAtlas Eagle Geocoding service (TeleAtlas, Redwood City, CA, USA). Annual average daily traffic count data in 2000 were obtained from the California Department of Transportation (Caltrans), assigned to TeleAtlas roadway links and adjusted to represent the years of 2003 and 2004 based on a statewide vehicle-miles-traveled growth, i.e., 2.4% per year from 2000 to 2004 (Wu et al. 2005b). Distance to different types of roadways (freeway, arterial, and collector roads) were calculated in ArcGIS 8.3 (ESRI, Redlands, CA, USA) based on TeleAtlas MultiNet™ USA roadway network. Traffic densities were calculated using the density plotting feature of ESRI Spatial Analyst software (ESRI, Redlands, CA, USA).

Dispersion model estimates

The CALINE4 dispersion model was used to predict traffic-specific pollutant concentrations ($PM_{2.5}$, EC, OC) for receptors given the source strength using emission factors, meteorology, and site geometry (Benson 1992). The uncertainties in EC and OC emission factors are discussed elsewhere (Wu et al. 2009). The original CALINE4 model was further modified to incorporate contributions from road segments within 5 km to a receptor (Wu et al. 2005c). Meteorological predictor variables were hourly wind speed, wind direction, and temperature which were taken from the Rubidoux SCAQMD site for Riverside and at the Pico Rivera SCAQMD site for Whittier, respectively. Also included in the predictions were average hourly mixing heights by season (cool and warm) which were obtained from the 1997 Southern California Ozone Study at the Los Angeles and Ontario International Airports for assignments to Whittier and Riverside, respectively (Croes and Fujita 2003).

Analysis

Summary characteristics and correlations were calculated for personal, home indoor, home outdoor, and central site measurements by city. Separate and pooled mixed linear regression models with a random household effect were constructed for Riverside and Whittier to predict 24-h average home outdoor concentrations of $PM_{2.5}$, EC, and OC, respectively. The full model for each pollutant had the form:

$$C_{ij}^{\text{out}} = \beta_0 + \beta_1 \times C_j^{\text{Cn}} + \beta_2 \times C_{ij}^{\text{CAL}} + \beta_3 \times \text{city} + \sum_m \alpha_m \times \mathit{traffic}_{im} + \sum_n \delta_n \times G_{in} + \sum_p \gamma_p \times \mathit{Met}_{jp} + \sum_q \lambda_q \times \text{time}_{jq} + \varepsilon_{ij} \quad (1)$$

where C_{ij}^{out} and C_{ij}^{CAL} were the measured and CALINE4 modeled home outdoor pollutant concentrations, respectively, at home i on day j, and C_j^{Cn} represents central site measurements. City was an indicator variable in the pooled model. Three traffic variables (traffic_{im}, m=1–3) were used, including distance weighted traffic counts at the residence for heavy-duty vehicles, light-duty vehicles, and total traffic. The four geographical variables (G_{in}, n=1–4) included population density and minimal distance from the residence to roads of three different classes (highway (including freeways and other highways), arterial roads, and collector roads). Twelve meteorological variables (Met_{jp}, p=1–12) were tested including 24-h averages of temperature, relative humidity, season, wind speed, wind vectors, and wind direction frequencies. Wind vectors were calculated as the vector sum of hourly wind speeds and directions over a day with the resulting average wind directions categorized into four quadrants (N–E, E–S, S–W, and W–N). Wind direction frequencies were defined as hours per day from each of the four quadrants. Three time variables (time_{jq}, q=1–3) were included to account for daily or weekly cycles, including date_j, sin(t), and cos(t), where $t=2\pi \times \text{date}_j/7$. ε_{ij} described the model error.

In a first step, predictors were entered in the models using forward, backward, or stepwise selection procedures. Stepwise linear regression with a 0.1 significance level chosen for a covariate entering or staying in the model led to the best models. For the pooled models, a "city" effect was forced in if it was not retained during the selection process. In a second step, the important predictors that were determined from the above models were used in a mixed model with a random effect to account for data clustering within homes. The random effect fitted best when a compound symmetry correlation structure with heterogeneous variances between repeated measures was applied. Final models were selected based on model fit using the Akaike's information corrected criterion (AICC). We used the same modeling approach to predict the ratios of home outdoor to central site measurements that was used as a method to predict spatial variation. All models were examined for reliability using the "leave-one-out" cross-validation approach, where each observation was removed from the dataset and evaluated against the model prediction.

To examine the variation in personal exposure explained by the modeled home outdoor concentrations, adjusted R^2 from the linear regression and the bias (the differences between the measured and modeled values) are reported. Linear regression was also used to compare personal $PM_{2.5}$, EC, and OC measurements with the corresponding CALINE4 estimates at home. All statistical analyses were performed with SAS 9.1 (SAS Institute Inc., Cary, NC, USA).

Quality control

The data collection rate, defined as the number of valid samples divided by the total number of expected samples, for personal measurements of $PM_{2.5}$, EC, and OC ranged between 76% to 89% in Riverside and 94% to 95% in Whittier. For personal measurements in the 63 subjects, data from four Riverside subjects were excluded from analysis as the residences of two subjects (one with home monitoring) were outside the geographical area for the CALINE4 model and the other two subjects were not geocoded due to inadequate TeleAtlas data. Outdoor measurements from seven Riverside homes (excluding one above) and eight Whittier homes were pooled, totaling 131 $PM_{2.5}$ and 129 EC and OC measurements with matched central site measurements for modeling.

In Riverside, personal EC and OC data were excluded from analysis for the following reasons. We found poor correlations of personal EC and OC with indoor EC and OC (non-significant r values, 0.08 and 0.22, respectively). Despite the moderate correlations between personal and measured outdoor EC and OC in Riverside (r=0.35 and 0.45, respectively), the predicted outdoor concentrations from the home outdoor models did not describe the variation of the personal measurements for EC and OC (R^2=0.01 and 0.03, respectively). We attribute these results to a possible leakage problem in the filter cassettes at Riverside (cassettes were hand clamped not vise clamped). In addition, other unmeasured factors could have influenced these results, including those related to the community, differences in organic aerosol composition (described below) and thus OC sampling artifacts, or to between-subject differences in time-activity in Riverside vs. Whittier.

We also learned later that the pDRs used for Whittier subjects were calibrated by the manufacturer with different reference aerosols from those used for the Riverside pDRs, even though both were called "Arizona road dust". Furthermore, we expected a different aerosol composition in Whittier (more of a source site with higher primary combustion aerosols) vs. Riverside (more of a receptor site with higher secondary photochemical aerosols). To adjust for this calibration difference, the personal pDR measurements were compared with the indoor Harvard Impactor measurements during the days when the subjects spent more than 98% of the time at home (additional Data given in Online Resource 1). While home indoor and personal measurements were about the same in Riverside (slope= 0.75), personal $PM_{2.5}$ measurements in Whittier had to be corrected according to following equation:

$$PM_{2.5,corr} = 0.317 \times PM_{2.5,meas} + 4.61$$

$$(N = 13, R^2 = 0.97) \quad (2)$$

The precision of the pDR was 5 μg/m³ (Liu et al. 2002). The limit of detection (LOD), defined as three times the standard deviation of the field blanks, was 0.15 and 0.63 μg/m³ for personal EC and OC in Whittier, respectively. The LOD for indoor, outdoor, and central site EC and OC measurements using the Harvard Impactors was 0.06 and 0.30 μg/m³, respectively.

Results

Summary statistics

Outdoor $PM_{2.5}$ averaged 28.3 and 16.7 μg/m³ in Riverside and Whittier, respectively. Personal $PM_{2.5}$ and outdoor EC and OC concentrations were also higher in Riverside than Whittier (Fig. 2). In Riverside, central site, personal, and home outdoor $PM_{2.5}$ concentrations were similar and about two times higher than the indoor concentrations. The low indoor concentrations might be explained by the more frequent use of air conditioning in Riverside compared to Whittier (42% vs. 34%). In Riverside, we also observed a difference between the homes with and without monitoring. Average pDR measurements when children were inside at home were lower in the group of children with home monitoring (20 μg/m³ vs. 26 μg/m³). For EC and OC in Riverside, central site levels were higher than home indoor and outdoor concentrations. In Whittier, $PM_{2.5}$ and EC levels were similar across all microenvironments, respectively, while OC levels were higher for personal and home indoor environments. The CALINE4 model estimates for $PM_{2.5}$, EC and OC from local mobile sources were expectedly lower than the actual measurements, which include all sources. Assuming CALINE4 estimates were accurate, then about 30% of outdoor $PM_{2.5}$ and 60% of EC and OC would have come from local traffic in Whittier, while in Riverside the local traffic contribution would only be 20% for $PM_{2.5}$ and 45% for EC and OC. This is in accordance with the GIS data (Table 1) showing that subjects in Whittier lived closer to major roads and were exposed to more traffic exhaust than subjects in Riverside.

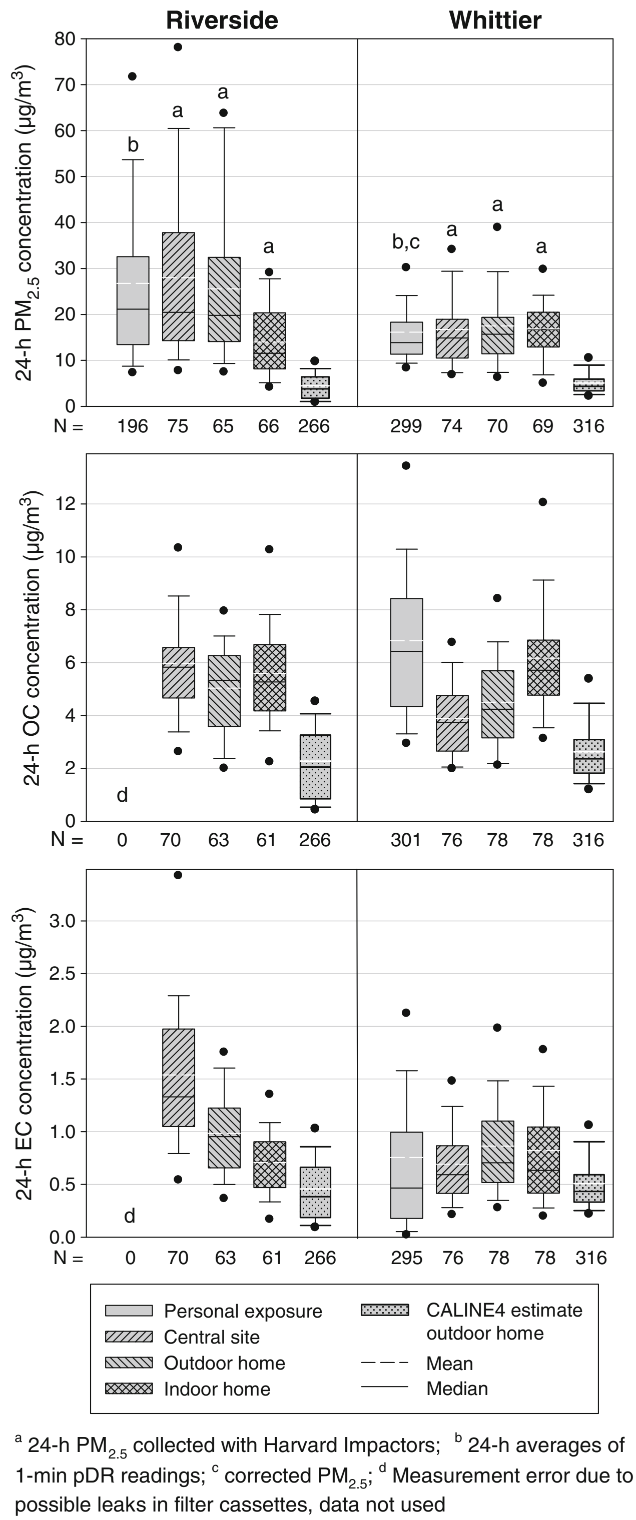

Fig. 2 Daily averages of air pollution measurements by location

Correlation between personal, central site, and home measurements

Home outdoor concentrations of $PM_{2.5}$ were strongly correlated with those at the central sites (r=0.96–0.97)

Table 1 Daily averages of GIS parameters at subjects' homes

GIS variables	Riverside panel (27 subjects)				Whittier panel (32 subjects)			
	N	Mean (standard deviation)	Median	Min/Max	*N*	Mean (standard deviation)	Median	Min/Max
Population density (1/km^2)	266	1,712 (1,057)	1,696	141/3,782	316	3,568 (1,528)	2,952	978/6,575
Minimal distance to highways (m)	266	2,653 (1,933)	2,175	460/6,938	316	1,371 (929)	1,084	121/3,339
Minimal distance to arterial roads (m)	266	488 (537)	310	6/2,593	316	336 (317)	248	20/1,130
Minimal distance to collector roads (m)	266	492 (338)	401	26/1,275	316	294 (324)	144	4/1,238
Total traffic count (distance weighted)	266	404 (567)	228	68/3,149	316	617 (479)	545	125/2,207
Heavy-duty vehicle traffic count (distance weighted)	266	21 (18)	12	5/75	316	23 (18)	16	5/89
Light-duty vehicle traffic count (distance weighted)	266	384 (562)	216	63/3,074	316	594 (464)	521	116/2,147

and less so with the home indoor measurements (r=0.48–0.79) (Table 2). Likewise, personal $PM_{2.5}$ measurements showed good correlations with those at the central (r=0.81–0.84) and home outdoor (r=0.77–0.88) sites and less so with those at home indoor sites (r=0.65–0.85). Compared to $PM_{2.5}$, slightly weaker correlations were found between home outdoor and central site measurements of OC (r=0.78–0.86) and EC (r=0.68–0.89). Correlations for OC and EC between indoor and outdoor measurements were weaker in Riverside (r=0.41 and 0.49, respectively) than in Whittier (r=0.72 and 0.63). In Whittier, correlations between personal and central site OC and EC were low (r=0.22 and 0.29, respectively) and correlations between personal and outdoor site OC and EC were moderate (r=0.55 and 0.57, respectively). Personal EC concentrations showed a strong correlation with home indoor EC (r=0.90) and personal OC showed a moderate correlation with indoor OC (0.54).

As the CALINE4 model estimated air pollution exposures driven by local traffic exhaust alone, evaluation of these estimates is not straightforward because our ambient measurements include both local and regional pollution. Thus, we compared CALINE4 estimates to home outdoor and personal EC measurements, which were assumed to better represent local traffic sources than OC or $PM_{2.5}$. In Riverside, the correlation between measured EC and estimated CALINE4 EC for the home outdoor environment was not significant. In Whittier, CALINE4 home outdoor EC estimates showed a moderate correlation to home outdoor EC measurements (r=0.51), while little correlation was found with personal exposure to EC, even after excluding subjects who reported indoor sources (r=0.18, without outliers).

Home outdoor models

The best models from the stepwise regression for home outdoor $PM_{2.5}$, OC, and EC for individual cities and pooled data are shown in Table 3. The central site measurement was the predominant predictor in all models, accounting for more than 93%, 61%, and 46% of the variability in home outdoor $PM_{2.5}$, OC, and EC concentrations, respectively. For $PM_{2.5}$, the adjusted R^2 was over 0.94 in models for Riverside, Whittier, and the pooled data. For OC models, the adjusted R^2 was 0.91 for Riverside, 0.83 for Whittier, and 0.80 for the pooled model. The second most important predictors in the OC models included minimal distance to collector roads for Riverside (partial R^2=0.14) and temperature for Whittier (partial R^2=0.16). For EC models, the adjusted R^2 was 0.76 for Riverside, 0.87 for Whittier, and 0.75 for the pooled model. The second most important predictors in the EC models included minimum distance to

Table 2 Pearson correlations between the concentrations at different locations by pollutant

		Riverside			Whittier			Pooled		
		Central Site	Home outdoor	Home indoor	Central Site	Home outdoor	Home indoor	Central Site	Home outdoor	Home indoor
$PM_{2.5}$	Home outdoor	0.97		0.79	0.96		0.48	0.97		0.57
	Personal	0.81	0.88	0.85	0.83	0.77	0.74	0.84	0.86	0.65
OC	Home outdoor	0.86		0.41	0.78		0.72	0.79		0.56
	Personal	N/A	N/A	N/A	0.22	0.55	0.54	N/A	N/A	N/A
EC	Home outdoor	0.68		0.49	0.89		0.63	0.70		0.55
	Personal	N/A	N/A	N/A	0.29	0.57	0.90	N/A	N/A	N/A

All correlations were significant with p<0.01

highway for Riverside (partial R^2=0.19) and wind direction for Whittier (partial R^2=0.06). Both OC and EC models with pooled data identified population density as the second most important predictor. While wind variables were significant in all EC and OC models, CALINE4 estimates only entered the EC model for Whittier.

Since the central site measurements accounted for most of the temporal variation, we also tested models for the prediction of ratios of home outdoor to central site measurements by the same variables tested above. This was intended to reduce the temporal variation across raw measurements that were taken at different times (different 10-day runs) and to examine predictors of spatial variation. Results in Table 4 suggest that spatial variation was only a fraction of the total variation in the measurements because all ratio models had lower R^2 values than the concentration models (Table 3). The EC ratio model for Whittier had the lowest R^2 suggesting lower spatial variability in EC (Table 4). Given the higher R^2 values, we used the concentration models in the following section for evaluation of the representativeness of predicted outdoor home to personal exposure measurements.

Figures 3 and 4 provide a visual display of model performance. These figures plot 10-day averaged ratios of home outdoor to central site OC as well as EC, using actual measurements (Fig. 3a, b for Whittier and Fig. 4a, b for Riverside) as well as model predictions using equations developed from Table 3 models (Fig. 3c–f for Whittier and Fig. 4c–f for Riverside). We used two types of models, one was specific to the city (Figs. 3c, d and 4c, d), the other was the model using data from both cities (pooled model) and applied for predictions in the specific city (Figs. 3e, f and 4e, f). In Whittier, air pollution concentrations were lower at the central site than those at outdoor home sites, resulting in ratios mostly above 1. In contrast, the higher concentrations at the Riverside central site, located 600 m from the 215 freeway, resulted in lower ratios. In Whittier, no spatial patterns could be observed for either measured (Fig. 3a) or estimated OC ratios (Fig. 3c and e), whereas in Riverside, both measured (Fig. 4a) and estimated OC ratios (Fig. 4c and e) were higher along freeways and in areas with a denser street network. For EC, plots based on actual measurements showed higher ratios along freeways in both cities (Figs. 3b and 4b), which were captured by the city-specific models (Figs. 3d and 4d). In Whittier, however, the freeway effect disappeared in the pooled model predictions (Figs. 3f).

Comparisons between personal and predicted outdoor exposures

Estimates from the home outdoor $PM_{2.5}$ models explained 65%, 69%, and 69% of the variation in personal $PM_{2.5}$ measurements in Riverside, in Whittier, and both cities pooled, respectively (Fig. 5a). The prediction bias, expressed as the difference between measured and predicted values was below 1 μg/m^3 for all models. There was no difference in the performance between the city-specific and the pooled model predictions (Fig. 5a and b, respectively). Estimated outdoor $PM_{2.5}$ explained more variation in personal $PM_{2.5}$ exposure among individuals with monitored homes than those without monitored homes (Fig. 5c).

Comparisons between personal and estimated outdoor OC and EC data were performed for Whittier only, as the personal EC and OC data in Riverside were removed after quality control. Predictions from the home outdoor OC model explained little of the variation in personal OC exposure (R^2=0.05). Exclusion of three unexplained high OC measurements (>3 SD from the mean and identified with arrows in Fig. 6) increased the model fit slightly to 0.08, with a prediction bias of 2.3 μg/m^3, about 35% of the mean (6.6 μg/m^3). Outdoor OC estimates at monitored homes explained only a slightly higher percentage of the variability in measured personal OC as compared with those at the non-monitored homes (Fig. 6).

Similarly, home outdoor EC estimates explained a small percentage of the variation in personal EC measurements (R^2=0.1). However, exclusion of four outliers (>3 SD from the mean) due to candle burning and cooking increased the R^2 to 0.37 (Fig. 7). The prediction bias was −0.2 μg/m^3, which is 33% of the mean (0.6 μg/m^3). The EC model predicted slightly better for the subjects with home measurements.

Effects of PM sources

We further examined these outdoor predictions by removing measurements with self-reported indoor sources (near smoking or cooking), defined when there was at least one 15-min entry of any indoor pollution event in the time-activity diary during each run day. The percentage of pollution events was similar in both cities, 42 of 266 in Riverside and 62 of 316 subject-days in Whittier. No significant differences in the performance of model predictions were found for all $PM_{2.5}$, OC, and EC models between the groups with or without reported indoor sources.

Seasonal effects

As the measurements were taken in two different seasons in each city, we looked for differences in model performance by season. For $PM_{2.5}$ the correlations between measured personal exposure and predicted home outdoor concentrations showed no significant difference between summer

Table 3 Results of linear regression modeling for home outdoor measurements of $PM_{2.5}$, OC, and EC

Dependent variable *Model*	N	Predictor Variable	Estimate	SE	Partial R^2	Adj. Model R^2
Home $PM_{2.5}$ (μg/m^3)						
Riverside	62	Central site $PM_{2.5}$ measurement (μg/m^3)	0.77**	0.02	0.96	0.97
		Relative humidity at central site (%)	0.07**	0.03	0.01	
		Heavy-duty vehicle traffic count (distance weighted)	0.10**	0.03	0.01	
		Weekly time term (sin)	1.09*	0.54	2.0E-03	
Whittier	69	Central site $PM_{2.5}$ measurement (μg/m^3)	0.97**	0.03	0.93	0.94
		Average wind speed (miles/h)	−0.89	0.62	4.9E-03	
		Weekly time term (sin)	0.95*	0.45	4.0E-03	
		Frequency of wind direction from N to E	0.12	0.08	3.3E-03	
Pooled	131	Central site $PM_{2.5}$ measurement (μg/m^3)	0.83**	0.02	0.94	0.96
		Minimal distance to highway (m)	−7.4E-04**	1.8E-04	0.01	
		City	−0.14	0.67	0.01	
		Total traffic count (distance weighted)	2.1E-03*	8.0E-04	2.6E-03	
		Weekly time term (sin)	1.00**	0.37	2.5E-03	
		Average temperature at central site (°F)	−0.09**	0.03	2.2E-03	
		Average wind speed (miles/h)	−0.65	0.34	1.0E-03	
Home OC (μg/m^3)						
Riverside	55	Central site OC measurement (μg/m^3)	0.65**	0.04	0.74	0.91
		Minimal distance to collector road (m)	1.5E-03**	2.0E-04	0.14	
		Weekly time term (sin)	0.40**	0.12	0.02	
		Average wind direction from N to E	0.85**	0.30	0.01	
		Relative humidity at central site (%)	0.02**	4.7E-03	0.01	
Whittier	75	Central site OC measurement (μg/m^3)	1.13**	0.07	0.61	0.83
		Average temperature at central site (°F)	−0.11**	0.02	0.16	
		Average wind direction from W to N	1.27**	0.40	0.03	
		Average wind speed (miles/h)	−0.59**	0.18	0.02	
		Weekly time term (cos)	−0.40**	0.13	0.02	
		Minimal distance to highway (m)	−3.7E-04	2.2E-04	3.4E-04	
Pooled	130	Central site OC measurement (μg/m^3)	0.77**	0.05	0.62	0.80
		Population density (per km^2)	2.7E-04**	7.6E-05	0.12	
		Average wind direction from W to N	0.59*	0.27	0.02	
		Relative humidity at central site (%)	9.9E-03	5.9E-03	0.02	
		Minimal distance to highway (m)	−1.7E-04**	5.6E-05	0.01	
		City	0.37	0.44	0.01	
		Frequency of wind direction from E to S	−0.10**	0.03	0.01	
		Frequency of wind direction from S to W	−0.04	0.02	0.01	
		Average wind speed (miles/h)	−0.28*	0.12	0.01	
		Average wind direction from E to S	0.63	0.33	1.3E-03	
Home EC (μg/m^3)						
Riverside	55	Central site EC measurement (μg/m^3)	0.49**	0.05	0.46	0.76
		Minimal distance to highway (m)	−6.9E-05**	1.5E-05	0.19	
		Average wind direction from N to E	−0.48**	0.11	0.05	
		Average wind direction from E to S	−0.40*	0.16	0.03	
		Average wind speed (miles/h)	0.10*	0.04	0.02	
		Daily time term (sin)	−0.09*	0.04	0.02	
Whittier	75	Central site EC measurement (μg/m^3)	0.93**	0.06	0.79	0.87
		Frequency of wind direction from N to E	0.03**	4.9E-03	0.06	
		EC CALINE4 home estimates	0.38**	0.12	0.02	

Table 3 (continued)

Dependent variable *Model*	*N*	Predictor Variable	Estimate	SE	Partial R^2	Adj. Model R^2
Pooled	130	Central site EC measurement (μg/m^3)	0.66**	0.04	0.48	0.75
		Population density (per km^2)	6.6E-05**	2.1E-05	0.16	
		Minimal distance to highway (m)	−6.1E-05**	1.6E-05	0.04	
		Relative humidity at central site (%)	3.8E-03*	1.8E-03	0.03	
		Frequency of wind direction from E to S	−0.04**	8.2E-03	0.02	
		Frequency of wind direction from S to W	−0.02**	6.7E-03	0.02	
		Average wind direction from N to E	−0.24*	0.10	0.01	
		City	0.17	0.13	1.2E-04	

SE standard error

*$p<0.05$; **$p<0.01$

and winter (R^2=0.69 and R^2=0.62, respectively). However, in Whittier home outdoor models for EC and OC explained more of the personal exposure variance in winter than in summer (EC: R^2=0.56 and 0.30, OC: R^2=0.22 and 0.09, respectively).

Discussion

In contrast to a previous validation study (Nethery et al. 2008), we found that predictions for daily concentrations of $PM_{2.5}$ in the outdoor home model were good surrogates for personal exposure to $PM_{2.5}$. Since local traffic accounted for less than 30% of the $PM_{2.5}$ measurements in our study cities, the remarkable performance of the $PM_{2.5}$ models likely reflected the common sources of regional transported $PM_{2.5}$ contributing to both the personal and outdoor $PM_{2.5}$. Home outdoor prediction models for specific components of $PM_{2.5}$ (EC and OC) were poorer indicators of personal exposure probably because these exposures are more affected by local sources such as traffic. Therefore, home outdoor models may not be adequate for predicting personal short-term exposure to specific sources that are relevant to studies of acute health outcomes. This conclusion most likely does not apply to the prediction of long-term exposures in studies of chronic health outcomes because it is expected that a smoothing of daily exposure variation would lead to less error in the prediction. This issue could not be addressed in the present study because we only collected ten consecutive days of sampling per subject.

We found strong correlations between personal and indoor EC but weaker correlations with outdoor and central site EC. Similar correlations were reported for EC during the summer in Boston, MA, USA (Brown et al. 2008) and for black smoke in Gothenburg, Sweden (Johannesson et al. 2007). The EC models showed comparable spatial patterns and predictors between our study cities. Clougherty et al. (2008) reported similar predictor variables which were important for personal EC and NO_2 models during the summer in Boston. Ryan et al. (2008) showed an improvement by 0.02 for the model R^2 of an outdoor model for traffic-related EC when adding wind parameters to the model in addition to traffic parameters. In our models, wind parameters showed similar effects with partial R^2 between 0.01 and 0.06. The lack of predictive power of the home outdoor EC models for personal EC exposure in the present study could be explained by the moderate correlations between personal and actual outdoor EC measurements (r=0.57 in Whittier). A better approach to predicting personal exposure to EC would entail the combination of a better model to predict home outdoor EC and knowledge of other sources of personal EC exposure linked to personal activities. Measurement errors in personal EC exposure might also contribute to part of the poor prediction.

This was the first study that examined the spatial variation of OC and predictors of personal OC exposure. Major predictors for the spatial variation of outdoor home OC varied depending on the study area (based on our ratio models for home outdoor to central site OC measurements). In Riverside, the major predictor was the heavy-duty vehicle counts, while in Whittier it was temperature. When data were pooled, the minimal distance to collector roads was the most important predictor, likely because this variable provided a local source contrast between these two cities. Wind variables played a minor role in all ratio models, likely accounting for some of the upwind/downwind influences of OC sources.

Although the R^2 values of our home outdoor OC models were above 0.8, these models provided poor estimates for personal OC exposure. The weak personal–central site correlations of OC measurements also indicated sources other than regional PM contributing to personal OC exposure. Nevertheless, OC measurement error might also

Table 4 Results of linear regression modeling for ratios of home outdoor to central site measurements of $PM_{2.5}$, OC, and EC

Dependent variable *Model*	N	Predictor variable	Estimate	SE	Partial R^2	Adj. model R^2
$PM_{2.5}$ ratio						
Riverside	62	Population density (per km^2)	1.7E-04**	2.7E-05	0.36	0.43
		Relative humidity at central site (%)	4.3E-03**	1.1E-03	0.07	
		Average wind direction from N to E	0.12	0.07	0.03	
Whittier	69	Frequency of wind direction from N to E	0.02**	0.01	0.24	0.31
		Weekly time term (cos)	−0.09**	0.03	0.08	
		Average wind direction from W to N	0.20	0.11	0.02	
Pooled	131	Minimal distance to highway (m)	−5.3E-05**	1.1E-05	0.24	0.38
		Frequency of wind direction from N to E	0.02**	4.8E-03	0.06	
		Average temperature at central site (°F)	−0.01*	2.1E-03	0.05	
		Weekly time term (cos)	−0.08**	0.03	0.05	
		city	0.04	0.07	2.0E-03	
		Average wind direction from W to N	0.10	0.05	5.4E-05	
OC ratio						
Riverside	55	Heavy-duty vehicle traffic count (distance weighted)	0.01**	1.2E-03	0.47	0.70
		Relative humidity at central site (%)	3.3E-03**	9.0E-04	0.09	
		Average wind speed (miles/h)	0.09**	0.02	0.09	
		Weekly time term (sin)	0.05*	0.02	0.03	
		Frequency of wind direction from N to E	0.02*	0.01	0.02	
		Average wind direction from N to E	−0.26**	0.07	0.02	
Whittier	75	Average temperature at central site (°F)	−0.02**	4.1E-03	0.45	0.58
		Average wind direction from W to N	0.35**	0.11	0.07	
		Weekly time term (cos)	−0.12**	0.03	0.06	
		Average wind speed (miles/h)	−0.10*	0.04	0.02	
		Heavy-duty vehicle traffic count (distance weighted)	3.3E-03	1.7E-03	0.01	
Pooled	75	Minimal distance to collector road (m)	−4.2E-04**	6.4E-05	0.24	0.63
		city	−0.03	0.05	0.17	
		Frequency of wind direction from N to E	0.04**	0.01	0.08	
		Average temperature at central site (°F)	−0.01*	3.4E-03	0.06	
		Relative humidity at central site (%)	4.9E-03*	1.9E-03	0.04	
		Weekly time term (cos)	−0.08**	0.02	0.03	
		Daily time term (sin)	−0.05*	0.02	0.01	
		Average wind direction from N to E	−0.34**	0.08	0.01	
EC ratio						
Riverside	55	Relative humidity at central site (%)	0.01**	1.5E-03	0.34	0.66
		Minimal distance to highway (m)	−5.6E-05**	9.5E-06	0.24	
		Weekly time term (sin)	0.07*	0.03	0.04	
		Average temperature at central site (°F)	0.01	0.03	0.03	
		Average wind speed (miles/h)	0.07*	0.03	0.03	
		Average wind direction from S to W	0.14*	0.03	0.02	
Whittier	75	Ratio of EC CALINE4 home/central site estimates	0.24**	0.07	0.14	0.31
		Average temperature at central site (°F)	−0.03**	0.01	0.14	
		Average wind direction from N to E	−0.61*	0.27	0.04	
		Weekly time term (cos)	−0.12	0.07	0.03	
Pooled	130	Ratio of EC CALINE4 home/central site estimates	0.22**	0.08	0.41	0.53
		Population density (per km^2)	−4.1E-06	4.3E-05	0.04	
		Frequency of wind direction from N to E	0.03*	0.01	0.04	
		Weekly time term (sin)	0.11*	0.05	0.02	
		Daily time term (sin)	−0.08	0.04	0.02	
		Frequency of wind direction from S to W	−0.02	0.01	0.01	
		Average wind speed (miles/h)	0.12*	0.05	0.01	
		Average wind direction from N to E	−0.52**	0.16	0.01	
		City	0.32*	0.15	4.1E-03	

SE standard error

$*p<0.05$; $**p<0.01$

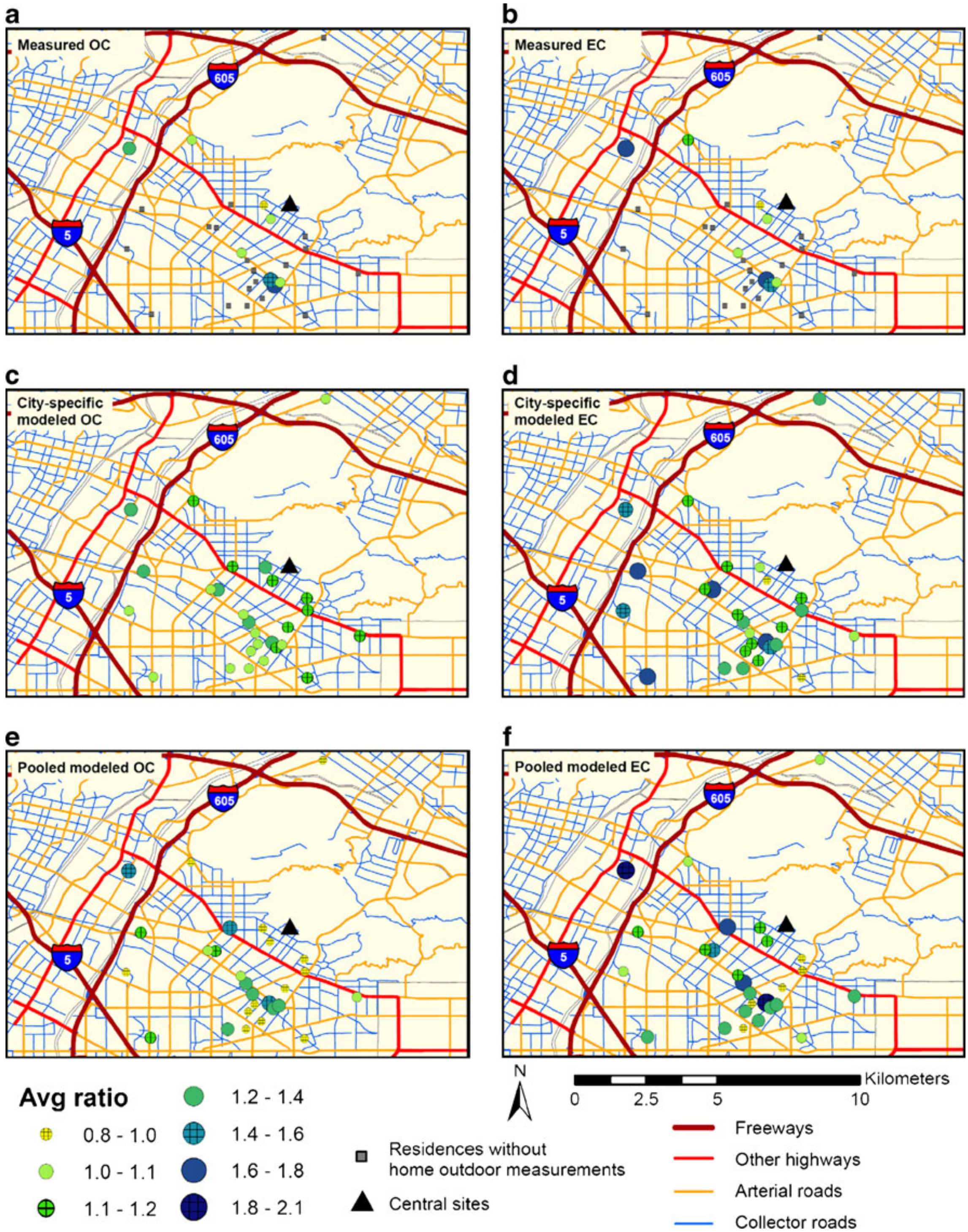

Fig. 3 Plots of 10-day average ratios of home outdoor to central site OC (*left panel*) and EC (*right panel*) levels in Whittier, using measurements (**a** and **b**), city-specific model predictions (**c** and **d**), and pooled model predictions (**e** and **f**)

contribute to part of the poor prediction of personal exposure (which was possible for the Riverside data we omitted). Previous studies have reported OC sampling artifacts due to the OC adsorption onto quartz filters, especially for OCs with lower molecular weights (Kirchstetter et al. 2001; Olson and Norris 2005; Turpin et al. 1994, 2000). This positive artifact could become profound for indoor measurements due to more abundant OC sources indoors than outdoors (Landis et al. 2001; Long et al. 2000; Pang et al. 2002). As our study did not implement back-up quartz filters to correct for the sampling artifact, personal OC measurements could be overestimated (Fig. 6).

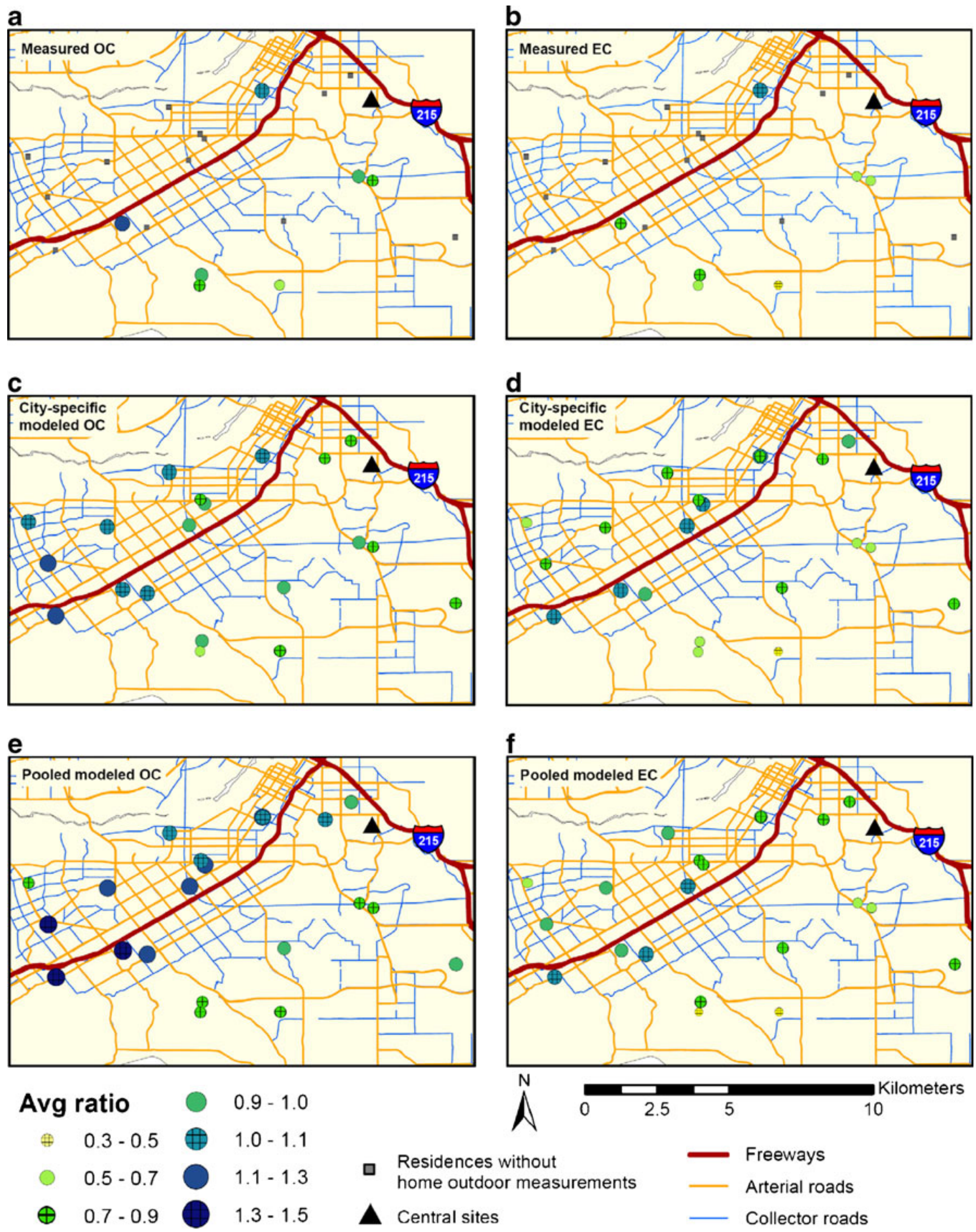

Fig. 4 Plots of 10-day average ratios of home outdoor to central site OC (*left panel*) and EC (*right panel*) levels in Riverside, using measurements (**a** and **b**), city-specific model predictions (**c** and **d**), and pooled model predictions (**e** and **f**)

The CALINE4 model, which takes into account source strengths and atmospheric convection processes, has been used by previous studies to estimate traffic-specific outdoor exposure in chronic health effect assessment (Gauderman et al. 2005; Molitor et al. 2006, 2007). In our stepwise regression modeling, the CALINE4 estimates only entered the models in Whittier for the prediction of home outdoor EC. It is likely that seasonal averages of meteorological parameters and annual traffic counts that were used in the CALINE4 model predictions could not capture the finer temporal variation in our daily measurements. The CALINE4 estimates also suffered from missing heavy-duty truck counts for some

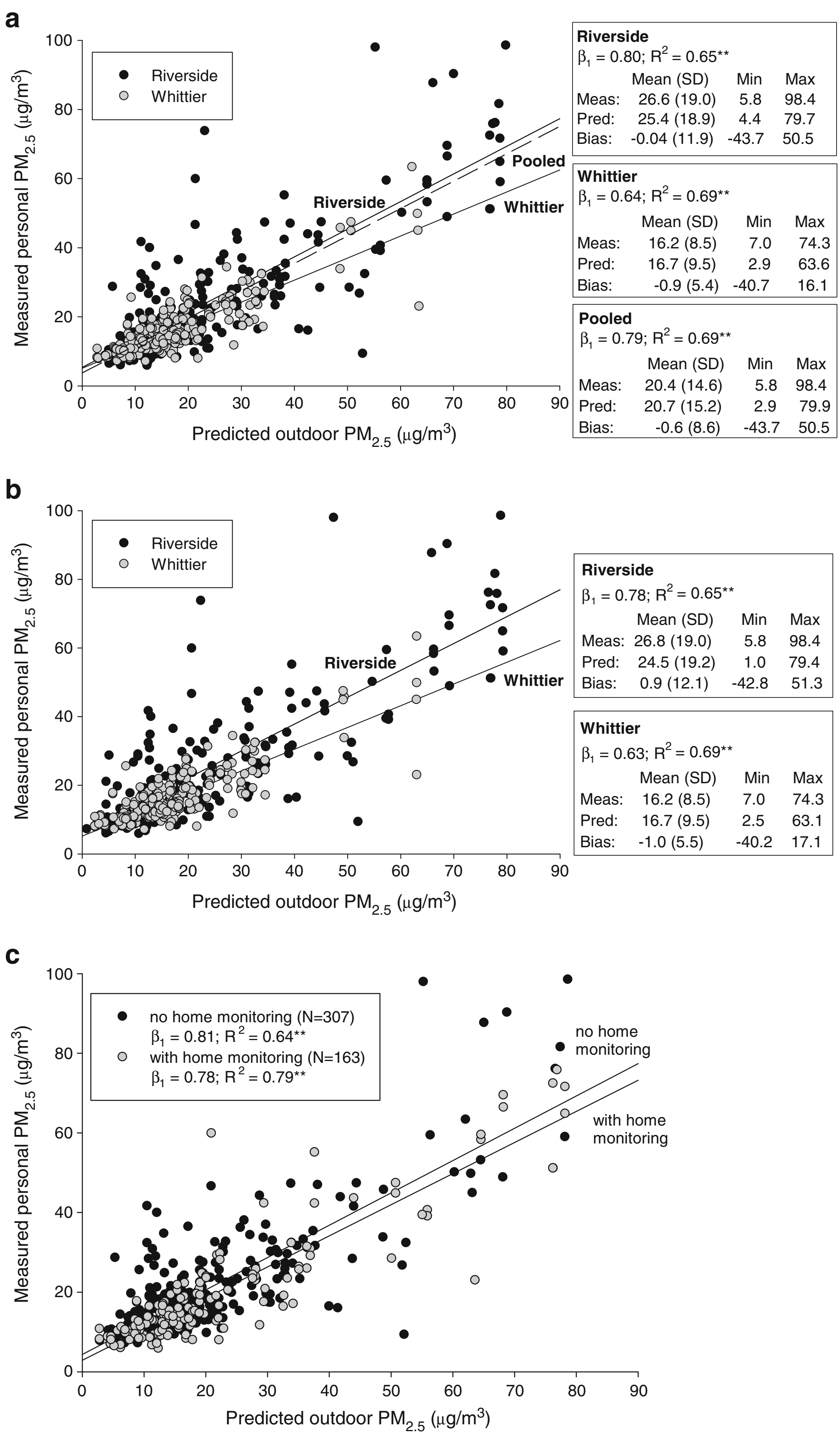

Fig. 5 Relationship between measured personal $PM_{2.5}$ and predicted home outdoor $PM_{2.5}$ from concentration models with a random "home" effect using **a** a pooled model, **b** city-specific models, and **c** grouped by monitored homes from the pooled model (β_1=estimate of regression slope; $^{**}p<0.01$). Statistics for measured (*Meas*) and predicted (*Pred*) $PM_{2.5}$ and the *Bias* are provided in *text boxes*

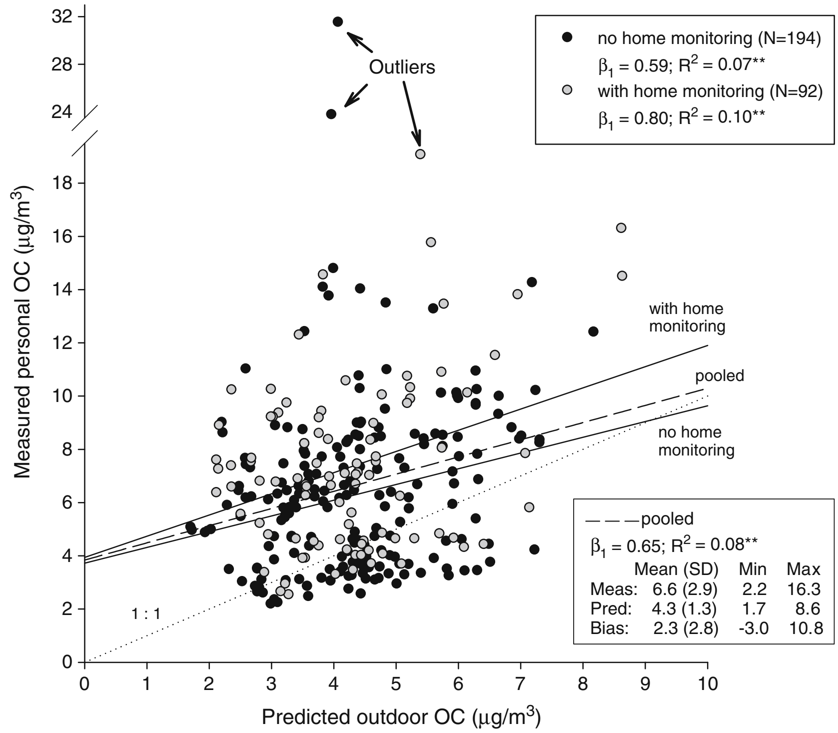

Fig. 6 Relationship between measured personal OC vs. predicted home outdoor OC from the city-specific concentration model for Whittier with a random "household" effect. All statistics were calculated without the three outliers (**$p<0.01$)

roadways in our study areas. Furthermore, no validation study has been conducted to our knowledge to verify the emission factors we used for EC and OC species in the CALINE models. Additionally, home outdoor EC estimates from CALINE4 had a moderate correlation with home outdoor EC measurements and a low correlation with personal EC measurements, which consisted of exposures in various other microenvironments. Therefore, it remains unclear whether studies of health responses to acute exposures should use CALINE4 predictions of outdoor home EC to represent personal exposure to traffic. Studies with more specific traffic markers (e.g., 1-nitropyrene for diesel exhaust) or source apportioned traffic estimates are needed to further evaluate the CALINE4 estimates.

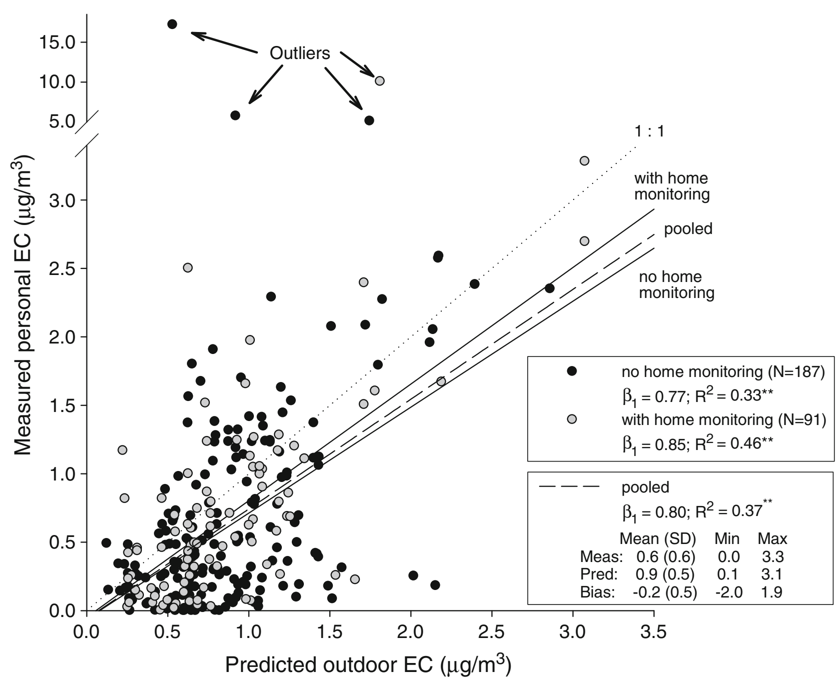

Fig. 7 Relationship between measured personal EC vs. predicted home outdoor EC from the city-specific concentration model for Whittier with a random "household" effect. All statistics were calculated without the four outliers (**$p<0.01$)

The ratio plots (Figs. 3 and 4) demonstrated spatial variation of EC and OC. While measured and predicted (city-specific) EC showed clear freeway effects, OC was more homogeneously distributed with higher ratios in areas with a denser street network. The lack of a freeway influence on OC was likely due to the minor influence of traffic sources, which is in accordance with a study in Mira Loma, close to Riverside (Na et al. 2004). In addition, the limited number of monitored homes might have not adequately covered the entire geographic range. The pooled models resulted in smoothed spatial variation near freeways, especially for EC. In summary, city-specific models captured more spatial variation than the pooled models.

In our study, personal–outdoor correlations for $PM_{2.5}$ were higher than personal–indoor correlations, and home outdoor concentrations were highly related to the central site measurements. Although most studies have found higher personal–indoor correlations than personal–outdoor correlations (Crist et al. 2008; Delfino et al. 2004; Liu et al. 2003; Meng et al. 2005), Brown et al. (2008) reported results similar to ours. The predominant predictor for all home outdoor models was the central site measurements, and these modeled outdoor levels, in turn, predicted short-term personal $PM_{2.5}$ exposure well. The ability to predict personal $PM_{2.5}$ did not differ by models (city-specific vs. pooled). Our results reinforced the earlier findings about the spatial homogeneity of outdoor $PM_{2.5}$ in an air shed (Krudysz et al. 2008) due to the major contribution from regional sources.

Although season was never retained as a significant predictor variable in the home outdoor models, we found seasonal differences in the ability to predict personal EC and OC but not for $PM_{2.5}$. In winter, air stagnation episodes with lower mixing heights lead to increased concentrations of traffic-related carbonaceous aerosols at ground level. This is expected to lead to increased indoor infiltration. On the other hand, $PM_{2.5}$ has a variable mixture of components across seasons, with more secondary aerosols in the summer, including nitrates.

As subjects spent most of their time indoors, we expected that the influence of indoor sources on personal exposure would diminish the predictability of the home outdoor models. However, this was not true for any of the pollutants due in part to the small number of reported incidences of indoor source exposures (mostly over brief periods). Similar results were found by Van Roosbroeck et al. (2008) who showed no effect of indoor sources on personal soot exposures.

Conclusions

We demonstrated that home outdoor models could be constructed with excellent predictions of daily $PM_{2.5}$, OC, and EC concentrations and using a limited number of monitoring sites within a city. Due to different predictive parameters of the EC and OC spatial pattern between Riverside and Whittier, city-specific models performed better than the pooled models. This suggests that future studies should take subregional differences into account for predicting outdoor spatial variation of EC and OC. We found that daily personal $PM_{2.5}$ exposure correlated well with the predicted home outdoor $PM_{2.5}$ concentrations. However, daily personal EC or OC exposure were poorly approximated by home outdoor EC or OC estimates. Results of our personal exposure analysis may not be generalized to other population groups, e.g., adults, as children with asthma probably have different activity patterns. Future work to predict short-term exposure to traffic-related particulate air pollution should focus on building personal exposure models that incorporate information on personal activities, locations, and highly specific measurements of traffic markers.

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