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Land use regression models for ultrafine particles, fine particles, and black carbon in Southern California

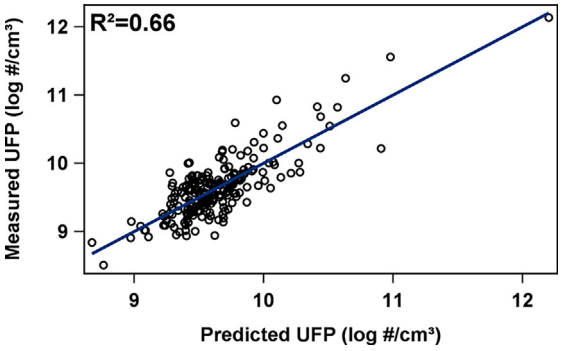
Rena R. Jones [a,*], Gerard Hoek [b], Jared A. Fisher [a], Sina Hasheminassab [c], Dongbin Wang [c], Mary H. Ward [a], Constantinos Sioutas [c], Roel Vermeulen [b,d], Debra T. Silverman [a]

[a] Occupational and Environmental Epidemiology Branch, Division of Cancer Epidemiology and Genetics, National Cancer Institute, Rockville, MD, United States
[b] Institute for Risk Assessment Sciences, Division of Environmental Epidemiology, Utrecht University, Utrecht, Netherlands
[c] Department of Civil and Environmental Engineering, University of Southern California, Los Angeles, CA, United States
[d] University Medical Center, Utrecht University, Utrecht, Netherlands

HIGHLIGHTS

- Exposure models are needed to disentangle the association between UFP and $PM_{2.5}$ on health risks.
- Short-term campaign in Los Angeles and surrounding counties measured UFP, $PM_{2.5}$, and BC.
- Land use regression models developed and exposures estimated for Southern California cohort.
- UFP and $PM_{2.5}$ measurements and predictions uncorrelated; independent health risks discernable

GRAPHICAL ABSTRACT



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ABSTRACT

Exposure models are needed to evaluate health effects of long-term exposure to ambient ultrafine particles (UFP; <0.1 μm) and to disentangle their association from other pollutants, particularly $PM_{2.5}$ (<2.5 μm). We developed land use regression (LUR) models to support UFP exposure assessment in the Los Angeles Ultrafines Study, a cohort in Southern California. We conducted a short-term measurement campaign in Los Angeles and parts of Riverside and Orange counties to measure UFP, $PM_{2.5}$, and black carbon (BC), collecting three 30-minute average measurements at 215 sites across three seasons. We averaged concentrations for each site and evaluated geographic predictors including traffic intensity, distance to airports, land use, and population and building density by supervised stepwise selection to develop models. UFP and $PM_{2.5}$ measurements ($r = 0.001$) and predictions ($r = 0.05$) were uncorrelated at the sites. UFP model explained variance was robust ($R^2 = 0.66$) and 10-fold cross-validation indicated good performance ($R^2 = 0.59$). Explained variation was moderate for $PM_{2.5}$ ($R^2 = 0.47$) and BC ($R^2 = 0.38$). In the cohort, we predicted a 2.3-fold exposure contrast from the 5th to 95th percentiles for all three pollutants. The correlation between modeled UFP and $PM_{2.5}$ at cohort residences was weak ($r = 0.28$), although higher than between measured levels. LUR models, particularly for UFP, were successfully developed and predicted reasonable exposure contrasts.

* Corresponding author at: Occupational and Environmental Epidemiology Branch, Division of Cancer Epidemiology and Genetics, National Cancer Institute, 9609 Medical Center Drive, Room 6E606, Rockville, MD 20850, United States.
E-mail address: rena.jones@nih.gov (R.R. Jones).

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1. Introduction

Numerous epidemiologic studies have shown associations of short- and long-term exposure to particulate matter air pollution characterized as particles <10 μm in aerodynamic diameter (PM_{10}) or <2.5 μm ($PM_{2.5}$) and adverse health effects (Kim et al., 2015). Evidence of health effects of ultrafine particles (UFP; <0.1 μm) is still accumulating (Ohlwein et al., 2019); however, UFP may be more toxic due to unique physiochemical properties that increase their potential for adsorption and interaction with tissues and cellular targets (Health Effects Institute, 2013). Importantly, although UFP dominate the number-based concentration of airborne PM, they comprise a small fraction of particle mass and thus are not well represented by measurements of other PM size fractions, including PM_{10} and $PM_{2.5}$.

Outdoor UFP are not included in the U.S. National Ambient Air Quality Standards, therefore no routine monitoring data exist and the spatial distribution of UFP in urban areas of the U.S. is not well characterized. UFP are formed by direct emissions from anthropogenic sources such as traffic (mobile sources) or industrial sources, or by nucleation and condensation of volatile and semi-volatile vapors (Brines et al., 2015; Kulmala et al., 2004; Sowlat et al., 2016). Directly emitted and nucleated sources tend to be localized and dissipate quickly after emission, but UFP are also influenced by meteorology and can form via further atmospheric transformation and may be regionally dispersed as a result. The agglomeration of UFP into larger particles (e.g., $PM_{2.5}$) results from atmospheric condensation of low-volatility organic species (Sioutas et al., 2005) under rates and conditions that vary depending on season and region. UFP are chemically complex, but the main constituents from mobile source combustion are agglomerated organic and black carbon (BC; a marker for diesel exhaust), ions such as sulfate, and trace amounts of metals (Daher et al., 2013; Mathis et al., 2004; Robert et al., 2007). The relative contribution of other sources to outdoor UFP, such as combustion byproducts of indoor cooking and heating, are region-dependent (Denier van der Gon, 2010). In large cities, the major UFP exposure source is traffic, especially emissions from heavy-duty diesel trucks and accelerating vehicles (Sowlat et al., 2016; Hasheminassab et al., 2013; Kaur et al., 2005; Morawska et al., 2008). Studies in the U.S. have found that traffic contributions to outdoor UFP vary dramatically by distance to roadways, and that most UFP exposure for individuals living in a major city likely arises from outdoor sources rather than from other microenvironments (Charron and Harrison, 2003; Wahlina et al., 2001; Zhu et al., 2005).

Land use regression (LUR) is a modeling approach used to characterize long-term average air pollutant concentrations at a fine spatial scale, providing high-resolution exposure estimates for epidemiologic studies. LUR models for UFP have been developed primarily in Europe and Canada (Abernethy et al., 2013; Sabaliauskas et al., 2015; Eeftens et al., 2012; Eeftens et al., 2016; Hoek et al., 2011; Montagne et al., 2015; van Nunen et al., 2017; Rivera et al., 2012; Cattani et al., 2017; Wolf et al., 2017; Weichenthal et al., 2016a). To fully capture the spatial variability of UFP, many studies have applied short-term and mobile measurements collected in real time at a variety of sites to represent the range of sources and concentrations in the area of interest. Previously published long-term LUR UFP models have differed in model structure and performance, likely due to differences in monitoring area characteristics, number of sites, and the duration and frequency of monitoring. Few previous studies have developed multiple pollutant models derived from the same monitoring effort, and similarly, there are few U.S.-based LUR models for UFP (Fuller et al., 2012; Li et al., 2013; Hankey and Marshall, 2015; Hankey et al., 2019; Patton et al., 2014; Zwack et al., 2011). Several LUR models have been applied in recent years in epidemiologic evaluations of health effects of long-term UFP exposure (Bai et al., 2019; Downward et al., 2018; Weichenthal et al., 2017a; Weichenthal et al., 2017b; Ostro et al., 2015). These studies have either focused on UFP alone or included measurements of $PM_{2.5}$ from another monitoring effort, raising issues of the comparability of exposure assessment for ultrafine and fine particles.

The objective of the current effort was to develop a LUR to provide high spatial resolution UFP exposure estimates for the Los Angeles Ultrafines Study, a subcohort of NIH-AARP Diet and Health Study participants residing in Los Angeles, Orange, and Riverside counties of California. We additionally aimed to develop LUR models for BC and $PM_{2.5}$ from the same short-term monitoring effort. The study area differs from most European and Canadian study areas in that freeways are the most prevalent transportation routes in the metropolitan area of Los Angeles, and thus are the most dominant source of traffic emissions that include UFP (Sowlat et al., 2016).

2. Materials and methods

2.1. Sampling design

The study catchment area included the South Coast Air Basin (hereafter, LA Basin) covered by Los Angeles County and parts of Orange and Riverside counties (Fig. 1). The short-term monitoring campaign was based on protocols in the EXPOsOMICS study (van Nunen et al., 2017) and modified to accommodate the layout of the LA Basin and its major UFP emissions source, freeways. Potential monitoring sites were identified within 12 freeway-centered clusters covering the area, including near I-405, I-10, CA-110, I-710, I-5, CA-210 east, and CA-60 highways (Fig. 1). Because Los Angeles International Airport (LAX) is also an important source of UFP in the Los Angeles area (Shirmohammadi et al., 2017; Hudda et al., 2014), we defined a LAX cluster and included sites near this source. Within each cluster, sites were placed in four categories relative to the freeway: upwind of this source, or downwind at minimum fixed distances of 50–150 m, >150–300 m, and >300 m (Fig. S1). Initially, we selected 238 candidate locations across the catchment area using a Geographic Information System (GIS) and/or other mapping tools to cover locations with varying air pollution concentrations, traffic intensities and composition, and different land uses in order to maximize exposure and predictor contrasts. Sites were also selected to avoid other local emission sources (e.g., gas stations, fast food restaurants) within a 100 m radius. Potential sites were evaluated in 360° view in Google Maps® and visited by field staff to confirm suitability for monitoring, which required that the sampling vehicle could safely remain stationary for the 30-minute measurement period and the site had limited local emission sources nearby. Characteristics of each site were noted by field staff and reviewed by the full study team. After excluding unsuitable sites, the 215 final sampling sites included a minimum of 20 sites in each cluster, with the exception of Orange and Riverside Counties (12 sites each).

2.2. Short-term monitoring campaign

Pollution measurements were collected using a hybrid vehicle as a measurement platform, installed with battery-operated instruments to measure three key air pollutants at each sampling site: UFP particle number concentrations (#/cm^3), and BC (ng/m^3) and $PM_{2.5}$ (μg/m^3) mass concentrations. UFP measurements were collected with a DiSCmini (miniature diffusion size classifier, Matter Aerosol, Wohlen, Switzerland) portable particle counter, which measures UFP with diameters of 10–700 nm in a concentration range of 10^3 to 10^6 cm^{-3} within a sampling interval of 1 s (Fierz et al., 2011; Ragettli et al., 2014). Concentrations of BC were

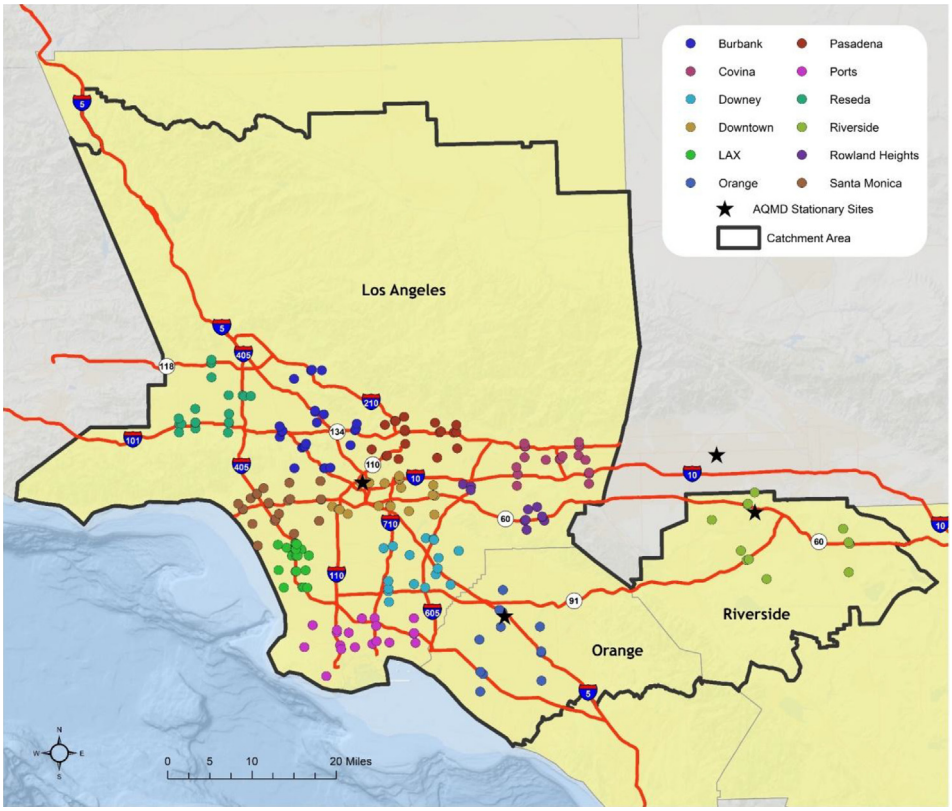

Fig. 1. Map of 215 UFP monitoring sites within 12 sampling clusters and four South Coast Air Quality Management District (AQMD) stationary monitoring sites.

measured using a micro-Aethalometer in 10 second intervals (model AE-51 microAeth, Magee Scientific, Berkeley, CA). DiSCmini and micro-Aethalometer AE-51 units were calibrated by the manufacturer prior to the study. Since two units of each device type were used during this study, we collocated them on a monthly basis to ensure consistency between their measurements. Past studies have evaluated particle number concentrations measured by the DiSCmini by comparing to widely used scanning mobility particle sizers and condensation particle counters and reported agreement within 10–20% (Fierz et al., 2011; Habre et al., 2018; Meier et al., 2013; Mills et al., 2013). Several studies have also evaluated the performance of the AE-51 against other BC monitors (e.g. multi-angle absorption photometer and rack mount aethalometer) and reported agreement within 7–12% (Cai et al., 2014; Cheng and Lin, 2013; Viana et al., 2015). A DustTrak (Model 8520, TSI Inc., Shoreview, MN) was also deployed to measure continuous $PM_{2.5}$ mass concentrations (1 s) at the sites. Agreement within 10–15% of gravimetric $PM_{2.5}$ measurements has been previously demonstrated for this device (Kam et al., 2011).

Sampling was conducted in three separate time periods over the course of 9 months in 2016, during a cool phase (Jan–March), spring phase (April–June) and warm phase (July–August). During each phase of sampling, short-term measurements (i.e., 30 min/site) were collected during non-rush hours (9:30–16:00) in order to represent the long-term traffic mean and site-specific UFP concentrations. Monitoring took place on different non-rainy weekdays and at various times of day; measurements were taken at the sites during different time intervals in each season.

2.3. Geographic predictors

Spatial predictor variables were generated for each of the sites in ArcGIS® using the site coordinates (derived first from Google maps and confirmed or adjusted during site visits from GPS) and digital datasets on land use, traffic, proximity to airports and ports, and population and housing characteristics. Predictors and buffer sizes were similar to those used in studies in Europe and Canada (Hoek et al., 2011; Montagne et al., 2015; van Nunen et al., 2017; Weichenthal et al., 2016b). We generated traffic predictors in circular buffers at radii of 50, 100, 300, 500, 1000 and 5000 m using road network data (TIGER/Line Shapefiles, 2000), including the sum of all road lengths within the buffer, and the sums of different road types separately, including A1 (major highway), A2 (major highway with restricted access), A3 (secondary roads), and A4 (neighborhood roads). For each buffer, we generated weekday vehicle miles traveled (VMT) for trucks and passenger vehicles from a local traffic demand model (Southern California Association of Governments, 2012), and computed the inverse distance and inverse distance squared to roadways and to major local sources (airports, ports). To account for average wind direction at the sites, we used an approach similar to Abernethy et al. to also create wedge-shaped buffers for roadway predictors (Abernethy et al., 2013; NOAA Automated Surface Observing System, n.d.). Land use variables reflected the percent of area within the buffers of each land use type (USGS, 2011), and population and housing unit density estimates included counts per square km within the buffers (American Community Survey, 2010). NO_2 at the Census block level from a national spatiotemporal model was used to represent background concentrations of traffic-related air pollutants (Bechle et al., 2015). A complete list of predictors and variable names is shown in Table S1.

2.4. Data preparation and analysis

We developed LUR models for log-transformed UFP concentrations using linear regression approaches similar to previous studies (Eeftens et al., 2012; van Nunen et al., 2017), first implementing univariate regressions followed by a supervised selection procedure

that evaluated all potential predictor variables in a stepwise fashion as to correlation and an a priori anticipated direction of effect. We applied standard model diagnostics, including assessments of normality and influential observations using Cook's distance. Collinearity between variables was assessed by variance inflation factor (VIF) (Eeftens et al., 2012; van Nunen et al., 2017). In the stepwise regression, the first predictor chosen was the variable with the highest adjusted explained variance (adjusted R^2) and the pre-specified direction of effect. Remaining predictors were evaluated iteratively and added to the model one-by-one if they contributed the largest improvement in adjusted R^2 and also had the pre-specified direction of effect. Subsequent variables were excluded if: a) the direction of effect changed for previously added predictors; b) the newly added variable was collinear with existing predictors (defined as VIF $\geq$3); or c) upon inclusion, *p*-values for the previously added variables exceeded $\alpha = 0.1$. This selection procedure was repeated until no remaining variable contributed an improvement to the adjusted R^2. We applied a 10-fold cross-validation approach by first randomly distributing monitoring sites into 10 groups, with each group acting as a validation set for one of the 10 models. HV R^2 and RMSE were obtained by regressing the predictions of all 10 validation sets against measured values. We also implemented strategies to improve model stability by running model selection procedures again after excluding predictors with >10% zero values. All statistical analyses were conducted in SAS version 9.3.

We initially developed spatio-temporal models based on the 644 individual 30-minute concentrations at each site and incorporating the wedge-shaped buffers for roadway predictors to account for wind direction in addition to other predictors in circular buffers. To account for temporal variation in pollutant concentrations both within and between days, we corrected raw UFP measurements using background measurements from four South Coast Air Quality Management District stationary monitoring sites (AQMD, 2014) using both ratio and absolute difference approaches as in the ESCAPE study (Eeftens et al., 2012). These modeling approaches did not result in meaningful models, and hence we developed spatial-only models including the raw UFP measurements without background correction. Most sites ($n = 214$) had three individual measurements and one site had two measurements. Variability of these individual 30-minute observations was high, therefore we implemented models after averaging the observations at each monitoring site, as was done in most other studies (Eeftens et al., 2016; Montagne et al., 2015; van Nunen et al., 2017).

2.5. Predictions at cohort residences

The final LUR models were applied to participant addresses in the Los Angeles Ultrafines Study. Briefly, the cohort is comprised of 53,833 NIH-AARP Diet and Health Study (Schatzkin et al., 2001) participants who resided in Los Angeles and parts of Orange and Riverside counties at study enrollment in 1995. The study population was aged over 50 years in 1995 and has been followed prospectively for ascertainment of cancer and other health outcomes for over 20 years. Participants were limited to those with well-geocoded (i.e., point or street address matches) addresses at study enrollment (97%; $n = 52{,}164$). After restricting values of geographic predictors to the bounds observed at monitoring sites, we applied the LUR models to cohort residences and generated predictions for UFP, $PM_{2.5}$, and BC.

3. Results

We found a high degree of spatial variability in the averaged 30-minute UFP means across the 215 sites, with a four-fold difference

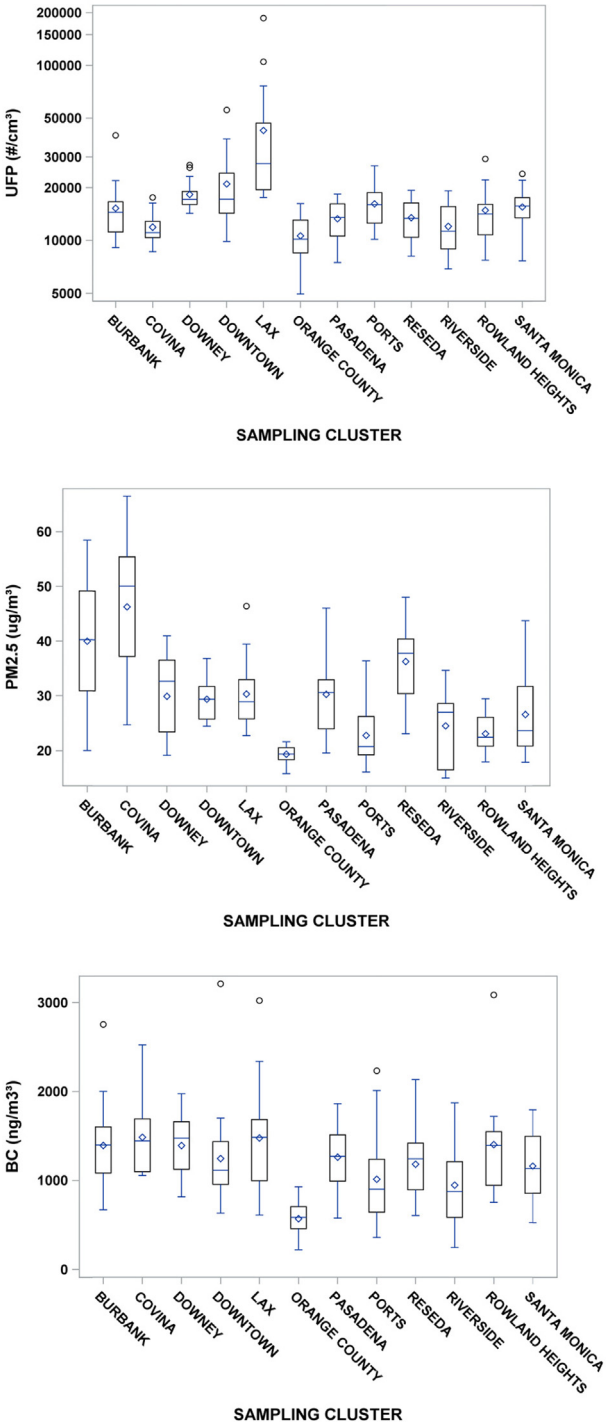

Fig. 2. Distributions of site-averaged 30-minute mean concentrations of UFP (#/cm^3), $PM_{2.5}$ (μg/m^3), and BC (ng/m^3) across 12 sampling clusters.

from the 5th to 95th percentiles. As expected, UFP concentrations were highest at the downwind sites closest to the roadway (50–150 m); however, measured levels at upwind sites were higher than downwind sites >300 m away (medians = 15,068 vs. 13,192 #/cm^3, respectively). There was also variability in average UFP concentrations across the 12 clusters (Fig. 2); concentrations were highest at the sites in downtown Los Angeles (median = 17,194 #/cm^3) and near LAX (median = 27,490 #/cm^3), where two extreme observations were noted (104,569 and 186,198 #/cm^3). The pattern of $PM_{2.5}$ and BC concentrations across these clusters differed

Table 1
Land use regression models for log-transformed UFP (#/cm^3), $PM_{2.5}$ (μg/m^3) and BC (ng/m^3).

Pollutant	Model[a]	R^2	RMSE	HV R^2	HV RMSE
UFP	7.74338 + 2.761089 * DIST_INV_LAX + 0.01834 * NO2 + 0.03491 * AIRPORTPCTA_1KM + 0.00877 * A1ROADLENGTH_CIRCLE_50 + 0.004705449 * DEV_HIGHINT_5000M + 0.10298 * VMT_PASS_1KM_LN − 3.3755 * DECID_FOREST_5000M − 0.3454 * CULTCROPS_1000M − 0.0801485 * MIXED_FOREST_5000M + 0.001980555 * DEV_MEDINT_50M + 0.00588122 * DEV_OPENSP_100M + 0.00307249 * DEV_HIGHINT_50M	0.66	0.27	0.59	0.29
$PM_{2.5}$	−1.714 + 0.37251 * VMT_PASS_5KM_LN + 0.00024061 * ACSHUDENS_5000 + 0.018289032 * DEV_LOWINT_5000M + 0.02109 * NO2 − 0.1615035 * MIXED_FOREST_5000M + 0.00012780 * A2ROADLENGTH_CIRCLE_1000 + 0.00122303 * DEV_HIGHINT_50M	0.47	0.21	0.44	0.24
BC	4.70754 + 0.05269 * NO2 + 0.09068 * VMT_PASS_1KM_LN + 1.55328726 * DIST_INV_LAX + 0.010466185 * DEV_LOWINT_5000M + 0.00907 * A1ROADLENGTH_CIRCLE_50 − 0.1911475 * CULTCROPS_500M − 3.5168 * DECID_FOREST_5000M + 0.01483 * AIRPORTPCTA_1KM + 0.00089133 * A3ROADLENGTH_CIRCLE_100	0.38	0.35	0.32	0.36

[a] VARIABLE (label; unit): DIST_INV_LAX (inverse distance to LAX airport; dist/KM); NO2 (NO_2 estimate for 2010 at year 2000 census block-level; PPB); AIRPORTPCTA_1KM (percent of 1KM buffer that is airport; % area); A1ROADLENGTH_CIRCLE_50 (sum of A1 road length within 50M buffer; M); DEV_HIGHINT_5000M (percent of 5000M buffer classified as highly developed; % area); VMT_PASS_1KM_LN (traffic intensity from passenger vehicles in 1KM buffer; log VMT/yr); DECID_FOREST_5000M (percent of 5000M buffer classified as deciduous forest; % area); CULTCROPS_1000M (percent of 1000M buffer classified as cultivated crops; % area); MIXED_FOREST_5000M (percent of 5000M buffer classified as mixed forest; % area); DEV_MEDINT_50M (percent of 50M buffer classified as developed, medium intensity; % area); DEV_OPENSP_100M (percent of 100M buffer classified as developed, open space; % area); DEV_HIGHINT_50M (percent of 50M buffer classified as highly developed; % area); VMT_PASS_5KM_LN (traffic intensity from passenger vehicles in 5KM buffer; log VMT/yr); ACSHUDENS_5000 (housing unit density within 5000M buffer; housing-units/km^2); DEV_LOWINT_5000M (percent of 5000M buffer classified as developed, low intensity; % area); A2ROADLENGTH_CIRCLE_1000 (sum of A2 road length within 1000M buffer; M); CULTCROPS_500M (percent of 500M buffer classified as cultivated crops; % area); A3ROADLENGTH_CIRCLE_100 (sum of A3 road length within 100M buffer; M).

from UFP; BC concentrations were less variable across clusters than either UFP or $PM_{2.5}$. $PM_{2.5}$ concentrations were highest in the Covina cluster (median = 50.1 μg/m^3) and the average across all sites was 30.9 μg/m^3. Concentrations of all three pollutants were lowest at the Orange County sites (medians = 10,183 #/cm^3, 19.4 μg/m^3, and 585.6 ng/m^3 for UFP, $PM_{2.5}$, and BC, respectively).

Final UFP models including the proximity to LAX and to freeways, housing unit density and highly developed land use explained 66% of the spatial variability in UFP concentrations (Table 1). Model R^2 indicated less good fit for $PM_{2.5}$ and BC models (R^2 = 0.47 and 0.38, respectively). Cross-validation indicated good model performance for all three pollutants; HV R^2 were <7% lower than the model R^2 and RMSE increased by about 7% for UFP, 14% for $PM_{2.5}$, and 3% for BC (Table 1). When we forced a constraint that predictors have <10% of observations as zero values, fewer predictors were retained in models (mostly land use variables were excluded) and we saw no gain in model stability as reflected by differences between model R^2 and cross-validated R^2 (data not shown). In sensitivity analyses, we added sampling cluster to final models to assess whether the developed LUR models accounted for differences in background concentrations across clusters. Inclusion of terms for cluster altered the significance of and degree of collinearity between several predictors but increased the percent of variability explained for the UFP models only by 1%, as assessed by the percent change in the adjusted R^2 compared to the final model. Inclusion of cluster increased the percent variability explained in $PM_{2.5}$ and BC models by 15% and 7%, respectively (Table S2).

Predictors common to both the UFP and $PM_{2.5}$ models included only NO_2 concentration and highly developed land use; predictions were uncorrelated at the sites (Pearson r = 0.05; Fig. 3). In contrast, UFP and BC models shared predictors reflecting traffic and nearby roads, NO_2, and airports, and predictions were moderately correlated at the sites (r = 0.62; p<0.001; Fig. 3). $PM_{2.5}$ and BC predictions were also moderately correlated at the sites (r = 0.51; p<0.001). Measurements of UFP were not correlated with $PM_{2.5}$ (r = <0.001) and weakly correlated with BC (r = 0.41; p < 0.001); correlations between $PM_{2.5}$ and BC measurements were moderate (r = 0.59; p < 0.001; Fig. 3).

At cohort residences, average predicted exposure to UFP was 13,159 #/cm^3, ranging from 3,160 to 106,359 #/cm^3. An approximate 2.3-fold exposure contrast was observed between the 5th and 95th percentiles and was 1.8-fold between the 10th and 90th percentiles (Table 2). The ratios of the 95th to 5th percentiles for $PM_{2.5}$ and BC were both 2.3-fold, respectively. UFP predictions at residences were weakly correlated with $PM_{2.5}$ (r = 0.28) and moderately so with BC (r = 0.64; Fig. 3). BC and $PM_{2.5}$ predictions were also moderately correlated (r = 0.58) at cohort addresses.

4. Discussion

We developed LUR models for ambient UFP, $PM_{2.5}$, and BC in Southern California to characterize the spatial variability in these pollutants across an area of the U.S. well known for traffic congestion and high outdoor air pollution. Few previous studies implementing LUR approaches have included both UFP and $PM_{2.5}$ models derived from the same monitoring effort such as we undertook in this study. The simultaneous collection of measurements and modeling efforts derived from these data allowed us to assess correlations between UFP, $PM_{2.5}$, and BC, both in terms of measured and modeled concentrations.

Our UFP model explained a greater proportion of the spatial variability in ambient UFP compared to models based on short-term monitoring in Vancouver (R^2 = 0.48) (Abernethy et al., 2013), the Netherlands (R^2 = 0.33–0.42) (Montagne et al., 2015) and a recent multi-site European effort (R^2 = 0.50) (van Nunen et al., 2017), and is comparable to a model in Toronto (R^2 = 0.67) (Weichenthal et al., 2016a). These R^2s reflect how well these models predict the average of short-term measurements, which still exhibit some temporal variability. We developed a spatial predictor model for UFP that likely will explain longer term averages better than shorter term average measurements, as the former have less temporal variation (Montagne et al., 2015; van Nunen et al., 2017; Kerckhoffs et al., 2016). Several Dutch studies have indeed documented that spatial models explained external longer-term measurements better than the short-term measurements from which the models were developed (Montagne et al., 2015; van Nunen et al., 2017). Our measurement of pollutants at a relatively large number of sites compared to other short-term monitoring studies was also important given the large size of our study area. Taken together, our results suggest that our UFP model is suitable

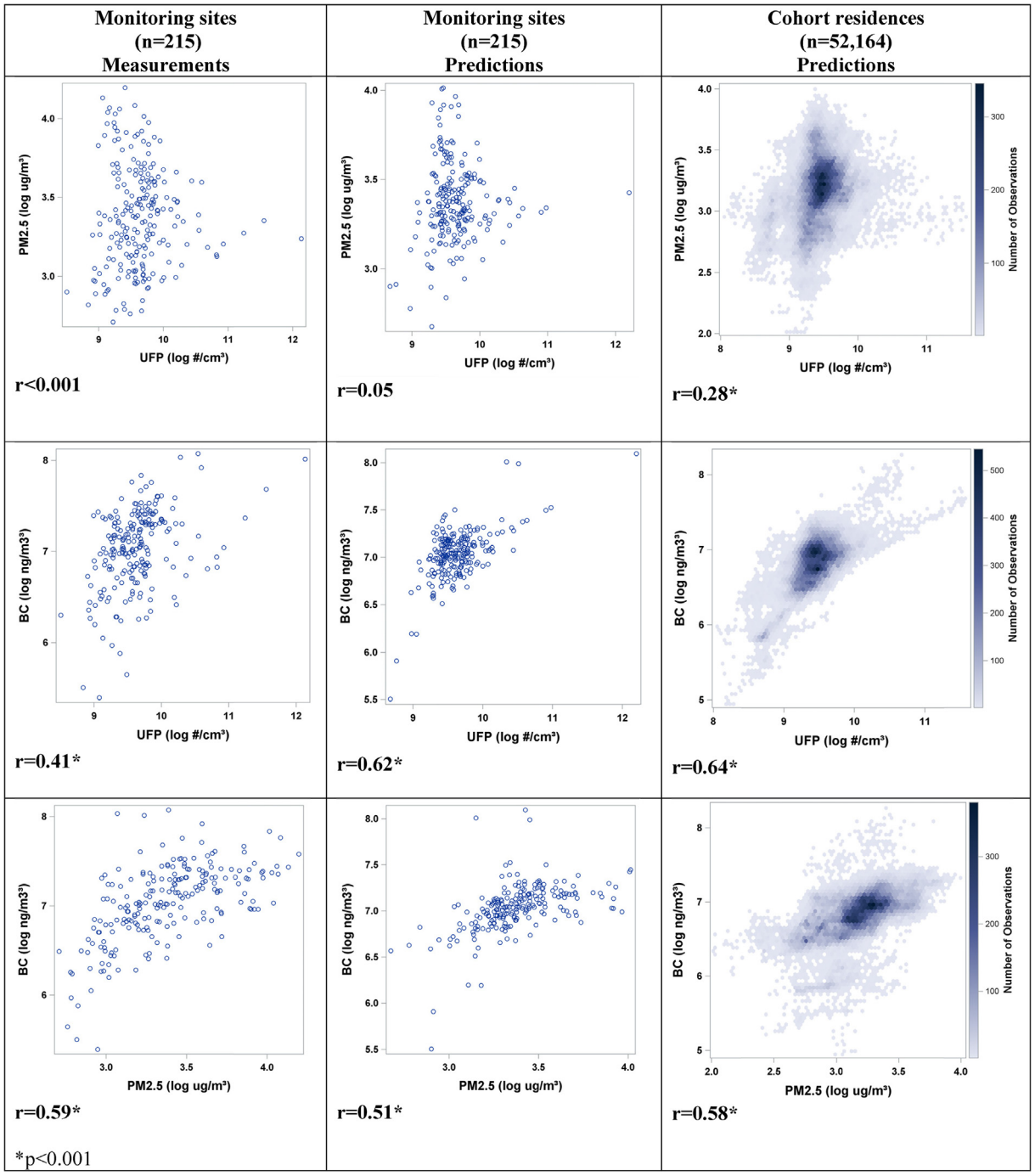

Fig. 3. Pearson correlations between log-transformed UFP (#/cm^3), $PM_{2.5}$ (μg/m^3) and BC (ng/m^3) measurements and predictions at monitoring sites and predictions at cohort residences in 1995.

Table 2
Distribution of predicted concentrations of UFP, $PM_{2.5}$, and BC at cohort residences in 1995.

Pollutant	Min	5th	10th	25th	Mean	Median	75th	90th	95th	Max
UFP (#/cm^3)	3160	8316	9614	11,046	13,159	12,647	14,628	17,171	19,002	106,359
$PM_{2.5}$ (μg/m^3)	7.5	15.5	16.9	20.6	24.5	24.2	27.8	32.1	35.8	54.4
BC (ng/m^3)	141	593	664	796	959	963	1109	1254	1360	3871

for application to epidemiologic analyses of long-term exposure (Abernethy et al., 2013; Montagne et al., 2015; Rivera et al., 2012; Ragettli et al., 2014). In contrast, our $PM_{2.5}$ and BC models performed comparatively less well than the UFP model. Several LURs for $PM_{2.5}$ exist. In Hong Kong, the R^2 for a $PM_{2.5}$ model was 0.63 (Shi et al., 2016). A modified LUR in Southern California that included a machine learning approach to model selection yielded a stronger $PM_{2.5}$ prediction (R^2 = 0.65) (Beckerman et al., 2013).

In the ESCAPE study, median model explained variance for $PM_{2.5}$ was 71%, although models predicted as little as 31% of the variation in $PM_{2.5}$ concentrations in some areas (Eeftens et al., 2012). One explanation for our lower prediction for $PM_{2.5}$ was limited availability of traffic intensity data, represented in our models via roadway metrics reflecting road type and estimates from a traffic demand model. A BC model in Toronto derived from bicycle-based mobile monitoring measurements included similar near-roadway predictors to our model and yielded only a modestly higher R^2 (0.43) (Minet et al., 2018). In contrast, the BC model developed by Hankey et al. in rural Virginia, U.S., was comparatively more predictive of mean BC ($R^2 = 0.67$) (Hankey et al., 2019), as was a model in Vancouver ($R^2 = 0.51$) (Larson et al., 2009), and in the ESCAPE study, where $PM_{2.5}$ absorbance was used as a marker of BC (median $R^2 = 89$) (Eeftens et al., 2012).

The key predictors of UFP in our model, density of major roadways and traffic intensity, are similar to important predictors in many other published LUR models to date (Abernethy et al., 2013; Eeftens et al., 2016; Hoek et al., 2011; Montagne et al., 2015; van Nunen et al., 2017; Rivera et al., 2012; Cattani et al., 2017; Wolf et al., 2017; Weichenthal et al., 2016a). Our sampling campaign was designed around the major UFP source in the area, freeways, and both major roadway density in a 50 m buffer and traffic demand (passenger vehicle miles traveled) were indeed predictive of UFP measurements. In the more compact European cities where these other models have been developed, traffic on major urban roads (versus the freeways in our study) is a common predictor of UFP. Among a limited number of U.S. LUR models based on mobile monitoring, traffic was similarly consistently a strong predictor of UFP, both in urban (Li et al., 2013; Patton et al., 2014; Zwack et al., 2011; Hankey and Marshall, 2015) and rural (Hankey et al., 2019) settings. Our data also demonstrated the influence of airports on UFP concentrations in the study catchment area. Airport predictors were included in both UFP and BC models and suggest that airports, specifically LAX, contribute to concentrations of these pollutants even when accounting for traffic sources. A recent emission rate study by Shirmohammadi et al. reported that within the impact zone of the LAX airport, which is roughly similar to the LAX cluster in our study, the LAX daily contribution to UFP, BC, and $PM_{2.5}$ were approximately 11, 2.5, and 1.4 times greater than the emissions from the surrounding freeways (Shirmohammadi et al., 2017). This finding further corroborates the significance of the proximity to LAX as a predictor of UFP and BC in our study. Few of the previous UFP LURs have evaluated airport predictors (Eeftens et al., 2016; Weichenthal et al., 2016a) although airports have been recognized as an important UFP emissions source, especially in the Los Angeles area (Shirmohammadi et al., 2017; Hudda et al., 2014; Hudda et al., 2018). In contrast, in a Swiss study, percent of airport land cover in a buffer was not an important predictor of UFP (Eeftens et al., 2016). Two Canadian studies found mixed results; in Montreal, airport proximity was positively associated with ambient UFP in single pollutant models but not in a multivariable model (Weichenthal et al., 2016b), and in Toronto the distance to the local international airport was a significant UFP determinant (Weichenthal et al., 2016a).

Measurements of UFP were not correlated with $PM_{2.5}$ and were weakly correlated with BC at our monitoring sites. These observations agree with the findings of previous dynamometer and ambient measurement studies conducted in Los Angeles. Biswas et al. showed that the advanced PM and NOx emissions control technologies on diesel trucks resulted in substantial reduction of PM from these sources but increased UFP emissions, mainly due to the nucleation of semivolatile organic vapors (Biswas et al., 2008). Moreover, using historical ambient speciation data, Hasheminassab et al. showed substantial and concurrent reductions in $PM_{2.5}$ and elemental carbon (a surrogate for BC) over the past decade in the LA Basin (Hasheminassab et al., 2014), while during the same period of time the ambient levels of UFP remained almost unchanged (Sowlat et al., 2016). These findings that PM mass and BC emissions from traffic went down over time while UFP emissions remained unaffected or increased may explain at least some of the low correlations between these pollutants in our data.

The correlations in concentrations of UFP with $PM_{2.5}$ and BC were higher for modeled than for measured concentrations in our data. The correlation between $PM_{2.5}$ and BC was similar for measured and modeled values. This difference in correlations may have been the result of residual temporal variation even after averaging individual measurements per site, and the temporal correlations may differ from spatial correlations. These results could also be an artifact of offering a limited number of spatial predictors into models, or due to the different performance of the models (Montagne et al., 2015). Another explanation for the somewhat stronger correlations between modeled UFP and $PM_{2.5}$ compared to their measurements is due to differences in the variability of predictor values at cohort residences compared to the monitoring sites, which were purposely selected to capture the full distribution of these determinants. The difference in correlation could also be due to insufficient accounting of sampling cluster effects reflecting background concentrations that clearly differed in the measurements of these pollutants. We added cluster to final models to assess whether these terms accounted for differences in background concentrations, and the explained variability in UFP concentrations was largely unchanged in models additionally adjusting for cluster. However, for $PM_{2.5}$ and BC, adding cluster to final models did increase model R^2s, suggesting that background concentrations of these pollutants were not fully explained by their respective LUR models. Given that the pattern of pollutant concentrations for BC and $PM_{2.5}$ were less variable across clusters than UFP, excluding the cluster from final models may explain the differences in correlations between UFP predictions and those of $PM_{2.5}$ and BC. As discrete clusters were defined for the purpose of sampling, application of models including clusters to the cohort residences spread over the study area is not feasible. LUR models generally have limited ability to account for differences in background pollution at scales larger than 5–10 km. Few other studies have assessed spatial correlations between both measured and modeled concentrations of UFP, $PM_{2.5}$ and BC. The correlations between measurements in our data were markedly lower than those in a study in Amsterdam, where the correlation between measured UFP and $PM_{2.5}$ was 0.66 (Hoek et al., 2011). However, the pattern of higher correlations between modeled and measured UFP and $PM_{2.5}$ concentrations was similar to that observed in our study.

One goal of our effort was to develop models reflective of long-term average exposures, and we developed models that used measurements collected across all hours of the day as one strategy to achieve this objective. Although we collected data only January–August, comparison of these data to measurements collected at the background monitoring sites for the full calendar year show that average levels of all three pollutants during our monitoring period were reflective of their respective annual averages (<10% absolute difference for all pollutants; data not shown). Similarly, we avoided peak exposure periods for sampling to better represent the long-term mean exposure experienced by the cohort members at their residence. Inclusion of peak exposures may have value in identifying areas with high levels of these pollutants but may be less ideal for estimates of chronic exposure, as peaks are less stable than average values.

Our study had a number of advantages, including repeated short-term measurements of multiple important traffic-related pollutants at a large number of monitoring sites, and covering a

broad geographic area. The stable weather in Southern California and our 9-month monitoring campaign covering the major seasonal changes indicate our measurements are reasonably reflective of concentrations over the full year. We used modeled traffic intensity estimates from 2012 in lieu of counts collected at the monitoring sites, which may have more accurately reflected this predictor. Like most prior studies, the temporal coverage of land use and road predictors also pre-dated our measurement campaign, but these estimates would be expected to be relatively stable over time (Yang et al., 2018). Given our objective to derive long-term exposure estimates, the design of our sampling campaign also focused on key exposure sources (e.g., airports, traffic) anticipated to be more stable over time. However, our choice to avoid other local sources, such as restaurants and gas stations, is another potential limitation of this effort. Our final models ultimately did not include background correction to account for temporal variability, as we observed minimal changes to our models with this adjustment. A study in the Netherlands similarly observed a lack of improvement to a spatial UFP model based on short-term measurements after reference site adjustment (Montagne et al., 2015), indicating that valid models for UFP may be obtained without this adjustment. Our sensitivity analyses suggested somewhat limited representation of background concentrations in models of $PM_{2.5}$ and BC across clusters. We also recognize the potential importance of meteorological factors to these predictions and attempted to address this source of variation in our modeling. Similar to our study, others have also shown a lack of improvement in model fit with inclusion of meteorological variables (Abernethy et al., 2013; Hankey et al., 2019), while another study found that adjustment for meteorological variables led to more predictive models than did correction for background concentrations (Minet et al., 2018).

5. Conclusions

We developed LUR models for ambient UFP, $PM_{2.5}$, and BC in three counties in Southern California to support UFP exposure assessment in the Los Angeles Ultrafines Study. Simultaneous measurement of all three pollutants allowed comparison of their correlations in measured concentrations as well as their predictions. The majority of spatial variability in mean UFP was explained in a model comprised primarily of traffic- and airport-related predictors, and moderate levels of variability in $PM_{2.5}$ and BC were explained in separate models for these pollutants. These models will be used to evaluate health effects of UFP in epidemiologic studies in Los Angeles, although use of the $PM_{2.5}$ and BC models may require incorporation of additional data to provide more robust exposure estimates.

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Declaration of competing interest

The authors declare no competing financial interests.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.134234.

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