

Table 2. Descriptive statistics for observed air sampling temporally-adjusted pollutant concentrations and meteorology

	Summer 2011	Winter 2012	Summer 2012	Winter 2013
PM _{2.5} (mean, SD, min-max $\mu\text{g}/\text{m}^3$)	54.25 (20.9, 0.0)	11.76 (20.1, 0.0)	13.94 (20.2, 0.0)	11.26 (20.1, 0.0)
BC (mean, SD, min-max $\mu\text{g}/\text{m}^3$)	0.53 (0.2, 0.0)	1.34 (0.2, 0.0)	1.26 (0.2, 0.0)	0.92 (0.2, 0.0)
Temperature (mean, min-max $^{\circ}\text{F}$)	66.26 (67.9–76.7)	59.27 (59.9–69.7)	76.99 (76.9–82.9)	59.25 (59.9–69.7)
Relative humidity (mean, min-max %)	65.60 (75.2–88.7)	76.22 (88.2–97.7)	67.39 (88.2–97.7)	72.76 (88.2–97.7)
Wind speed (mean, min-max m/h)	1.79 (1.2–2.2)	1.29 (1.2–2.9)	2.66 (2.2–2.7)	4.27 (2.2–2.9)
Wind direction (percentage of direction)	37% W 33% SW 17% S 13% E	33% W 33% SW 17% S 17% E	33% W 33% SW 17% S 17% E	33% W 33% SW 17% S 17% E
Inversion (percent percentage of direction)	33% 2 inversions 33% 2 inversions 17% 1 inversion	33% 2 inversions 33% 2 inversions 17% 2 inversions	33% 2 inversions 33% 2 inversions 17% 2 inversions	33% 2 inversions 33% 2 inversions 17% 2 inversions

hourly level (i.e. and the 24-h integrated hourly level (i.e. $24 \times$ hourly mean level) was created in which position (percent) on each sampling day was determined. Hourly meteorological data (e.g., wind speed, wind direction, temperature, precipitation, cooling/heating) were downloaded from the National Climate Data Center (i.e. 1950–2000) — full archival format. Backward layer air data was collected at the Pittsburgh National Weather Service station located in Moon Township, PA, approximately 12 miles west (upwind) for predominantly wind direction of Pittsburgh and east (downwind) from the National Oceanic and Atmospheric Administration.

Temporal Adjustment

PM_{2.5} and BC concentrations at distributed monitoring sites were temporally adjusted using the background reference concentration, which was consistently with all sampling events. Because all monitoring sites could not be sampled in the same week, temporal adjustment was performed using a reference site, to allow for comparison between sites. This adjustment also allowed us to assess the temporal contribution in the LUR models. Temporal adjustment methods and variability terms are detailed elsewhere¹⁴ PM_{2.5} concentration at a particular monitoring location was divided by the specific weekly PM_{2.5} concentration from the background site then multiplied by the relevant average PM_{2.5} concentration from the background site.

Quality Assurance/Quality Control

A quality monitoring program was performed each season to evaluate monitors at four randomly-selected sites. Field blanks were deployed during each season, and all concentrations were blank-checked. Hourly PM_{2.5} concentrations were observed from Allegheny County Health Department (ACHD) for the Liberty, Lawrenceville, and Ardara EPA Air Quality System (AQIS) monitoring locations. ACHD data were aggregated to match sampling hours each season, and used as additional validation sites, for consistent temporal trends observed at our background and urban reference sites. BC data was not available from ACHD monitors.

Statistical Analysis

Descriptive statistics, scatterplots, and histograms were used to characterize distributions of PM_{2.5} concentrations and BC distributions, as well as spatial covariates (e.g., traffic density, elevation, industrial emissions) and temporal covariates (temperature, relative humidity, wind speed, wind direction, frequency of inversions) (Table 2). Before modeling, bivariate source-pollutant correlation analyses were performed. Data profiles and model building were performed separately for PM_{2.5} and BC and for summer and winter seasons. Statistical analysis was conducted using the Proc GLM command in SAS version 9.3 (SAS Institute, Cary, NC, USA).

LUR models were implemented using manual forward stepwise linear regression to assess how PM_{2.5} and BC concentrations for the summer and winter seasons, using an adjusted version of the modeling approach described in Chughtai et al.¹⁴ The full set of source indicator covariates (Table 1) were tested individually for each pollutant by year and by season.

First, bivariate correlation coefficients (Spearman) that were examined, and the two covariates with the highest correlations from each source category were individually incorporated, entered by strength of the bivariate correlation. Temporal trends in PM_{2.5} and BC were first incorporated into LUR models using the sampling season-specific background concentration source term with the strongest univariate correlation with the temporally-adjusted pollutants were then incorporated individually, in descending order by strength of the bivariate correlation. Regression models were sequentially fit to assess model model improvement at each stage, using the coefficient of determination (R^2) and removing non-significant covariates to the order of descending R^2 until all terms were significant ($P < 0.05$). Covariates were removed at any stage if covariate selection factor became > 2.0 . Finally, we tested modification of each significant source term affect the bivariate frequency, direction, and wind speed (model adjustments).

LUR model results were mapped to identify systematic spatial variation and bivariate source profiles predicted by LUR, highlighting the variation of additional covariates (i.e., those not added to the LUR model). Scattergrams of residuals were created in QGIS and residuals were mapped against latitude and longitude coordinates (relative degrees) of monitoring locations to explore residual patterns. Spatial autocorrelation in residuals was tested using Moran's I statistic. For each final model, a spatial R^2 (roughly, the proportion of spatial variance explained by LUR terms) was derived, using final LUR covariates to predict temporally-adjusted concentrations.

Predicted PM_{2.5} concentrations and BC distributions were mapped across a regular $100 \times 100 \text{ m}^2$ grid, smoothed using inverse distance weighting (IDW), allowing spatial influence from nearest 100 grid cell centroids. Surface were calculated for contour points of equal concentration across the 100 surfaces, for visualization. Contour intervals of $2 \mu\text{g}/\text{m}^3$ for PM_{2.5} and 0.5 $\mu\text{g}/\text{m}^3$ for BC were selected for display.

Sensitivity Analysis

Covariate selection was sensitivity-tested using cross-validation to assess fit between each significant predictor and new pollutant concentrations, to ensure that candidate covariates selected captured variability across the range of the data, not reflect on outliers or influential points. Five structures and Random Forest automated methods were performed to characterize covariate selection. A summary of each returned term was tested against the residual of the prior model in the sequential model-building process to check for outliers. Model results were examined to ensure normally spatial autocorrelation across the residuals of the distributed sites was determined using Moran's I, and spatial correlations were determined using generalized additive models (GAM) to determine whether residual smoothing was required.

Sensitivity of the model building procedure to temporal adjustment was assessed by comparing two methods (background only vs. background and urban reference site adjustment) and by modeling covariates against temporally-adjusted pollutant concentrations to determine the percentage of spatial variation across the eight LUR models. Background elimination from multivariate linear models including all covariates with significant bivariate correlations with pollutant concentrations further corroborated model structure and covariate selection. For validation of LUR predictions, a random 20% of sites ($n = 7$) were removed from the analysis, and the LUR

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