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Source proximity and residential outdoor concentrations of $PM_{2.5}$, OC, EC, and PAHs

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Abstract

We examined the effect of proximity to specific mobile, area, and point sources on the residential outdoor concentrations of fine particulate matter PM ($PM_{2.5}$) and several of its particle components. Integrated (48-h) $PM_{2.5}$ samples were collected outside non-smoking residences in Elizabeth, NJ, between summer 1999 and spring 2001. Samples were analyzed for $PM_{2.5}$ mass, organic and elemental carbon (OC and EC, respectively), trace elements, particle-phase polycyclic aromatic hydrocarbons (p-PAHs), and other important particle species. Information about the proximity of the study homes to potential mobile and area sources of OC, EC, p-PAHs, sulfur (S), and selenium (Se) (including urban interstate highways, local roadways, the Newark International Airport, the Elizabeth seaport, and a nearby refinery in Linden, NJ) were retrieved from a database that included detailed emissions, meteorological, and geographical data for the study area. The dependence of residential outdoor concentrations on source proximity and on various meteorological parameters was then examined for each species by multiple linear regression analysis. As expected, the predicted ambient air concentrations of all particle species (except S, Se) decreased with increasing distance from the sources. Although the enhancement in $PM_{2.5}$ and OC levels outside the study homes closest to primary PM sources was modest (e.g., 1.6 and 2.5 times the background levels 37 m from interstate highways), the elevation of EC and p-PAH concentrations was substantial outside the closest study homes (i.e., about 20 times for p-PAHs 37 m from interstate highways and about 14 times for EC 192 m from the refinery in Linden, NJ). The predicted EC concentrations 192 and 500 m from the oil refinery were 22.8 and 3.0 $\mu gC/m^3$,

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Conflict of interest

The authors declare no conflict of interest.

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compared with an urban background of 1 μgC/m^3. Thus, emissions from this source might dramatically affect EC exposure for residents living in its close proximity.

Keywords

source proximity; particulate matter exposure; organic carbon; elemental carbon; particle-phase polycyclic aromatic hydrocarbons; RIOPA

Introduction

Living near major roadways has been identified as a risk factor for respiratory and cardiovascular problems (Van Wijnen and Van der Zee, 1998; Kaiser, 2005; Pope and Dockery, 2006). Among the various size fractions of particulate matter (PM) components, fine and ultrafine PM (particles with an aerodynamic diameters less than 2.5 and 0.1 μm, respectively) are hypothesized to be responsible for these observed adverse health effects (Xia et al., 2004; Nel et al., 2006).

Although fine and ultrafine PM are emitted from a variety of primary (e.g., roadways, airports, seaport, refinery, industries) and secondary (e.g., atmospheric photochemistry) sources, the majority of "source proximity" studies conducted to date have focused on the influence of proximity to roadways on air pollution concentrations and exposures. For example, in work conducted in the Netherlands, Roorda-Knape et al. (1998) found that unlike fine PM ($PM_{2.5}$), outdoor and indoor black carbon (BC) and NO_2 concentrations were higher close to roadways (50 m) and declined exponentially with distance. Similarly, Zhu et al. (2002) observed that the CO and BC concentrations and the ultrafine particle number concentration were highest in the vicinity (17 m) of a freeway (located in the Los Angeles area) highly influenced by heavy-duty diesel trucks, and decreased exponentially with increasing distance from the source, tracking each other well. In a study conducted in the New York metropolitan area, Lena et al. (2002) investigated the impact of large truck traffic intensity on residential outdoor concentrations of BC and predicted an increase of 1.69 μg/m^3 BC per 100 large truck/h ($P = 0.001$; $R^2 = 0.84$) at the examined sidewalk locations. Unlike BC, $PM_{2.5}$ concentrations were only weakly associated with local traffic.

This study (conducted within the Relationship of Indoor, Outdoor, and Personal Air, RIOPA, study) examines the effect of proximity to specific mobile, area, and point sources on residential outdoor concentrations of $PM_{2.5}$, organic carbon (OC), elemental carbon (EC), two selected particle-phase polycyclic aromatic hydrocarbons (p-PAHs) (Benzo-[*ghi*]-Pyrene and Coronene), sulfur (S), and selenium (Se) in Elizabeth, NJ. To our knowledge, this is the first time that the effect of proximity to specific mobile, area, and point sources on the residential outdoor concentrations of these PM components has been studied.

Potential associations are examined by means of multiple linear regression analysis (Netter et al., 1996; Kwon et al., 2006) using outdoor air concentrations as dependent variables and meteorology and proximity information as independent variables. The urban concentrations of S and Se are expected to be dictated by multi-day transport and transformation of coal combustion emissions (Lee et al., 2002). Thus, we do not expect to see enhanced

concentrations of these $PM_{2.5}$ species in close proximity to local sources. In contrast, Coronene, Benzo-[*ghi*]-Pyrene and EC are locally emitted directly from combustion sources. They are frequently used as tracers of primary combustion emissions from motor vehicle engines. For these species, we expect concentrations to be higher outside residences closer to roadways. OC and $PM_{2.5}$ are both emitted from local primary sources and formed in the atmosphere across the northeastern United States.

It should be noted that residential outdoor air pollutants are the major contributors to indoor concentrations of several particulate species (e.g., $PM_{2.5}$, EC, PAHs; Naumova et al., 2002, 2003; Meng et al., 2005; Polidori et al., 2006a, 2007). A better understanding of the impact of neighborhood-scale sources on residential outdoor concentrations will aid the design of effective air quality control strategies and regulatory decision making for public health protection.

Methods

Sampling and Analyses

As part of the RIOPA study, 48-h integrated $PM_{2.5}$ samples were collected outside non-smoking residences in Elizabeth, NJ, between summer 1999 and spring 2001 (Figure 1). The city of Elizabeth, NJ, is located in Union County, about 12 miles from New York City. It has a population of about 120,000 habitants, one of the highest population densities in New Jersey (approximately 10,000/mi^2), and several area and point sources of air pollutants (e.g., truck loading and unloading areas, petrochemical industrial facilities). Several roadway classes (e.g., interstate, major, and minor arterials) intersect the city.

During RIOPA study, one to two Elizabeth homes were sampled concurrently, and ∼80% of the homes were sampled a second time approximately 3 months later. The outdoor samplers were placed in secure locations in the front or back yard of each home. The home selection criteria for Elizabeth included oversampling residences located within 500 m of heavily trafficked roadways. A detailed description of the RIOPA study design (Weisel et al., 2005) and sampling details (Meng et al., 2005; Weisel et al., 2005; Polidori et al., 2006a) are provided elsewhere.

Fine PM samples were collected at approximately half of the study homes, and analyzed for PM_{25} mass (Meng et al., 2005), OC and EC (Polidori et al., 2006a), functional groups (Reff et al., 2005), elements (Meng et al., 2005), gas and p-PAHs (Naumova et al., 2002, 2003), and Chlordanes (Offenberg et al., 2004). Gas-phase aldehydes (Liu et al., 2006) and volatile organic compounds (VOCs; Weisel et al., 2005) were also measured. OC was corrected for the adsorption of organic vapors on the quartz fiber filter (QFF; positive artifact) by subtracting the backup filter OC on a sample-by-sample basis as described by Polidori et al. (2006a). The number of samples analyzed for each species considered in this study (i.e., PM_2.5, OC, EC, particle-phase Benzo-[*ghi*]-Pyrene and Coronene, S, and Se) is reported in Table 1.

Proximity and Meteorological Data

Information about the proximity of the RIOPA homes to potential sources of OC, EC, p-PAHs, S, and Se was retrieved from a comprehensive database developed by Kwon et al. (2006) to study the effect of source proximity on residential outdoor VOC concentrations in Elizabeth, NJ. This database includes detailed emissions, meteorological, and geographical data for the study area. ArcView GIS (version 3.1, ESRI, Redlands, CA, USA) was used to measure the distances between geographical locations and to build geographical inputs for statistical data analysis.

National emission inventory of 1999—Emission data for $PM_{2.5}$ mass, OC, EC, p-PAHs, and S for counties containing or adjacent to the Elizabeth area (i.e., Union, Essex, and Hudson Counties in New Jersey, and Richmond County in New York) were obtained from the 1999 National Emission Inventory (NEI, version 3.0 for the hazardous air pollutants, released in December 2003, and version 3.0 for the criteria pollutants, released in February 2004; http://www.epa.gov/ttn/chief/net/1999 inventory.html).

In addition to roadways, the most important sources of primary $PM_{2.5}$ in the Elizabeth area are a number of non-point sources for diesel emissions in and near Elizabeth, NJ (Figure 1). These included a truck depot (South Broad Street), a bus depot (first Street), the Port Authority-Marine Terminal, the Newark Liberty International Airport, and a truck loading and unloading area (located on Division Street, in very close proximity to one of the biggest commerce/ business centers in New Jersey). All of these sources are located in or near the North to Northeast section of Elizabeth with the exception of the oil refinery to the South. The truck and bus depots were quite close to a subset of RIOPA homes. The total number of non-road diesel sources (i.e., lawn and garden equipment, construction equipment, snow blowers, and so on) in the Elizabeth area was considerably lower than the on-road emissions and were not considered in the RIOPA database (the study area is highly urbanized and non-road mobile activities are limited). Secondary formation from precursors emitted regionally is also a substantial contributor to urban concentrations of $PM_{2.5}$, OC, and S (Chuersuwan et al., 2000; Lee et al., 2002, 2004; Polidori et al., 2006b).

Meteorological data for New Jersey—Meteorological data for Newark International Airport (EWR, WBAN 14734), NJ, on the northern boundary of Elizabeth, were obtained from the National Climatic Data Center, National Oceanic and Atmospheric Administration (NCDC/NOAA) database (www.ncdc.noaa.gov/oa/climate/rcsg/datasets.html). The hourly information was averaged over 48-h periods to match exactly the sampling time of the RIOPA samples. The meteorological data extracted included the following: dry bulb temperature (°K), relative humidity (RH%), precipitation (mm), atmospheric pressure (mm Hg), wind speed (*U*, m/s), wind direction (tens of degrees from true north), and mixing height (m).

In addition, the atmospheric Pasquill stability classes were obtained every 3 h during sampling periods from the NOAA AIR Resources laboratory archive (http://www.arl.noaa.gov/ready.html). As the atmospheric stability is a categorical variable, it could not be used directly in the regression analysis. Rather, the stability class was assigned

a code of “1” when “stable” or “neutral” (stability classes D, E, F, and G), and “0” when “unstable” (stability classes A, B, and C). A probability variable was then generated to represent the percent of the sampling period when the Pasquill stability class was “stable and neutral”. This probability variable was used in the analysis.

As reported by Kwon et al. (2006), micrometeorological variations in the wind direction near the ground (caused by urban structures around the sampling site) could not be accurately represented by the hourly wind direction information collected at the Newark International Airport station. Also, as 48-h average pollutant concentrations were used in our regression models and no dominant wind direction existed for the vast majority of 48-h sampling periods (all but a few), a wind direction variable was not included in the analyses that follow. It is quite possible that stronger associations with more sources would be seen if a similar analysis were performed with 1-h concentration and meteorological data.

Census 2000 TIGER/Line data sets—Spatial information regarding the locations of roads, railroads, and municipal boundaries (i.e., county, township, and city borderlines) used to calculate distances from sampled residences to major roadways were obtained from the Census 2000 TIGER (Topologically Integrated Geographic Encoding and Referencing system) data sets (http://www.census.gov/geo/www/tiger).

To improve the accuracy of the proximity data, TIGER data were compared with those obtained from Digital orthoquarter quadrangles (DOQQs), which are diagrams combining the image of a photograph with the geometric qualities of a map. The 1997 DOQQs were downloaded from the New Jersey Image Warehouse site of the NJ DEP, Bureau of GIS (http://njgin.nj.gov/OIT_IW/index.jsp).

The addresses of area and point sources in the Elizabeth area were obtained from web site listings, from the yellow pages (http://www.yellowbook.com), and from the Emergency Response/HAZMATof Union County (Division of Environmental Health and Emergency Management). Locations were verified by visual observation. The actual coordinates of each point line and area source were integrated with Census 2000 TIGER/Line data set to determine the distances to the study homes through ArcView GIS. The closest boundary of the facility was used to calculate distances for all area sources, except that the point of release was used for the refinery. The most important area and point sources considered in this study are as follows: a truck depot (truck warehouse; located on South Board Street), a truck loading and unloading area (on Division Street), a school bus depot (Summer Street), a transportation bus depot (first Street), the Newark International Airport (EWR), the Elizabeth seaport, and a nearby refinery (situated in Linden, NJ) (see Figure 1 for further details).

Road network—The functional classifications of roadways in Elizabeth were from the New Jersey Department of Transportation, Bureau of Transportation Data and Development (http://www.state.nj.us/transportation/refdata), and were organized through ArcView GIS (Kwon et al., 2006). The resulting diagrams provided a graphical representation of state, toll, county roads, intersecting streets, and other types of geographical data. In addition, these diagrams included the width and the centerline of all roadways. The distance between each

home and the nearest roadway was calculated using geoprocessed and verified locations of residential outdoor samplers and the centerline of the roadways (Figure 1).

Residential outdoor species concentrations (i.e., $PM_{2.5}$, OC, EC, particle-phase Coronene, particle-phase Benzo-[*ghi*]-Pyrene, S, and Se), meteorological and source emission inventory data, and proximity data used for all statistical data analysis presented here are reported in the Supplementary Information (Supplementary Table SI 1 to SI 4; see Supplementary Information for further details).

Statistical Data Analysis

In this study, the dependence of residential outdoor concentrations on source proximity was examined for each species by multiple linear regression. SAS for Windows (Version 8.2, SAS Institute, Cary, NC, 2003) was used for all statistical data analyses. Ln-transformed outdoor concentrations were used because their distributions were closer to normal with more constant variance than the non-transformed concentrations.

First, bivariate Pearson's correlations between the ln-transformed outdoor concentrations and predictor variables (i.e., proximity and meteorological variables) were used to identify potentially important predictors. Variables with statistically significant correlations at $\alpha = 0.05$ ($P < 0.05$) were included in preliminary regression analyses.

Preliminary multiple linear regression analyses between the residential outdoor concentration of each particulate species (Y_i) and a group of predictors (i.e., proximity to roadways, proximity to point and area sources, or meteorological variables, X_i) were then performed by stepwise selection to narrow the selection of variables. Predictors with $P < 0.15$ were selected for further evaluation.

The selected meteorological and distance variables were then used together as predictors of each particulate species in a series of multiple linear regression analyses (i.e., forward selection, backward elimination, and stepwise selection) to verify that the regression results were consistent and robust. The parameter selection criteria used for forward selection, backward elimination, and stepwise selection were $P < 0.50$, $P < 0.10$, and $P < 0.15$, respectively. Owing in part to different parameter selection criteria, the numbers of predictors chosen by the models varied somewhat. The models selected by the forward, backward, and stepwise selection methods, and the corresponding statistical results were compared and evaluated. For each examined particulate species the "best-fitting" multiple linear regression model was considered to be the one (a) satisfying the three major assumptions of linear regression (described in detail below), (b) with the highest overall and partial R^2, and (c) with a parameter coefficient (Cp) approximately equal to the number of predictors selected.

The major assumptions of linear regression analysis are (a) normality (the errors should be normally distributed), (b) homogeneity of variance (the error variance should be constant), and (c) linearity (the relationships between the predictors and the outcome variable should be linear). To verify normality, the studentized residuals were obtained for all regression models. Severe outliers were considered to be those points that were either 3 interquartile

ranges below the first quartile or 3 interquartile -ranges above the third-quartile. The presence of any severe outliers would be sufficient to reject normality at the 5% significance level. No severe outliers were found in any of the "best-fitting" models. The heteroscedasticity (e.g., variability of error variance) of model parameters (Netter et al., 1996) and the multi-collinearity of predictor variables were visually examined on the appropriate diagnostic plots (i.e., residual *vs* predicted plots, and probability–probability plots).

Finally, the best-fitting regression equations were used to estimate the effects of source proximity and meteorological parameters one at a time on residential outdoor concentrations of $PM_{2.5}$, OC, EC, Coronene, and Benzo-[*ghi*]-Pyrene. Specifically, concentrations of each PM species were calculated by varying the value of one predictor over the range of observed predictor variable values, while holding all other predictors constant. For the proximity variables, the "constants" were given as the maximum distances between the study homes and emission sources. The median values of meteorological variables were used. When this approach is applied to proximity data, the concentration of the PM species is described as the sum of its "urban background" level and the enhancement because of source proximity. It should be noted that urban point and area sources also contribute to the "urban background concentration" of $PM_{2.5}$.

Results

Preliminary Selection of Predictors

Bivariate Pearson's correlations and preliminary regression analyses between ln-transformed outdoor $PM_{2.5}$ mass and species concentrations and the predictor variables (Table 2) show that the proximity and meteorological variables were correlated with outdoor concentrations in the expected direction. In particular, proximity to roadways or other PM sources and atmospheric stability (stab) were positively correlated with one or more of the particulate species, while wind speed (*U*) and mixing height (mixH) were inversely correlated. Meteorological variables showed a stronger relationship with particulate species concentrations than distances to sources. Atmospheric stability was associated ($P < 0.01$) with all examined species. In addition, significant correlations ($P < 0.05$) between distances to one or more types of roadways were found for all particulate species except S and Se. As S and Se are dominated by regional transport, this gives confidence to the results obtained from these preliminary analyses.

Selection of the "Best-fitting" Models

A summary of the best-fitting models for all studied species is reported in Table 3. All best-fitting models met the assumptions of linear regression: absence of outliers, normality, homogeneity of variance, and absence of multicollinearity between predictors. In all cases, except OC, the best-fitting models were obtained through stepwise selection. As the significance level for forward selection (0.5) is higher than that of stepwise selection (0.15), in almost every case forward selection resulted in models that were identical to those obtained by stepwise selection. In the case of S, the forward selection model was overspecified as determined by *C*p. In the case of OC, forward selection was considered the

best-fitting model because stepwise selection did not satisfy all three assumptions of linear regression. Even so, coefficients obtained by forward and stepwise selection were nearly identical (within 6%) for the variables in the best-fitting OC model. The effect of season is not included in the regression models because of the limited number of samples and because the inclusion of season as an independent variable over-specified all of the regression models and caused severe multicollinearity issues.

The *F*-statistics for all best-fitting models and the probability values (*P*-values) for all parameter estimates were always statistically significant ($P_{\text{model}} < F$). The partial R^2 for the meteorological variables was typically larger than that for the proximity variables, implying that a greater percentage of the explanatory power was because of the changes in the meteorological conditions rather than in the distance to emission sources. As the multicollinearity was not significant for any model, no considerable interaction between the predictor variables was present in the "best-fitting models."

"Best-fitting" model for $LnPM_{2.5}$—The "best-fitting" (six-parameters) regression equation for the ln-transformed outdoor concentration of $PM_{2.5}$ ($LnPM_{2.5}$) was obtained by "stepwise" selection:

$$\mathrm{LnPM}_{2.5}=1.00+20.08\times\mathrm{F11\_inv}+4.27\times\mathrm{F19\_inv}+51.17\times\mathrm{truck\_inv}-0.13\times U+0.44\times\mathrm{srability} \quad (1)$$

where F11_inv is the inverse distance to interstate highways, F19_inv is the inverse distance to local roadways, truck_inv is the inverse distance to a major truck loading and unloading area in Elizabeth (Division Street), *U* is the wind speed, and stability is the atmospheric stability. Atmospheric stability is the most significant factor in the regression analysis (partial $R^2 = 0.32$) and has a positive coefficient, consistent with expectations and observations for $PM_{2.5}$ (Marcazzan et al., 2002; Celis et al., 2003). Atmospheric stability was included as a statistically significant predictor in the best-fitting models of all particulate species analyzed in this study. Wind speed has a negative coefficient, implying that high-wind speeds reduce outdoor $PM_{2.5}$ concentration (Roorda-Knape et al., 1998; Zhu et al., 2002). The inverse distance to interstate roadways (F11_inv), local roadways (F19_inv), and to the truck loading and unloading area (truck_inv) were also selected as significant predictor variables in the model; they all have positive coefficients.

The overall model is statistically significant ($P < 0.0001$), has an R^2 of 0.47, and a Cp of 6 (the same as the predicted number of parameters) indicating that the regression equation has an appropriate number of predictors. This means that 47% of the variance in the ln-transformed outdoor concentration of $PM_{2.5}$ can be explained by the variance of this six-parameter model. The good explanatory power of this model is somewhat surprising given the diversity of sources and formation mechanisms contributing to $PM_{2.5}$ mass concentrations. All parameter estimates have met a 0.15 significant level for entry into the model ($P < 0.15$).

"Best-fitting" model for LnOC—The "best-fitting" (four-parameters) regression equation for LnOC was obtained by "forward" selection:

$$\mathrm{LnOC} = -2.18 + 39.02 \times \mathrm{F11\_inv} - 0.01 \times \mathrm{precipitation} + 0.66 \times \mathrm{stability} \quad (2)$$

The two meteorological variables selected were atmospheric stability (stability) and precipitation (precipitation). The inverse distance to interstate roadways (F11_inv) was the last significant predictor variable to be selected. The overall model is statistically significant ($P < 0.0001$), has an R^2 of 0.33, and a Cp of 4, the same as the predicted number of parameters. All predictor estimates have met a 0.20 significant level for entry into the model ($P < 0.20$). The coefficient of determination for LnOC ($R^2 = 0.33$) was the lowest of the entire study, indicating that less of the variance in LnOC was explained by primary source proximity and meteorology than for any other examined PM_{25} component. One likely reason is that OC has major contributions both from the local primary sources and regional secondary sources, whereas the other examined PM_{25} species are dominated by either local primary sources (e.g., EC, Coronene, and Benzo-[*ghi*]-Pyrene) or regional sources (e.g., Se and S).

"Best-fitting" model for LnEC—The "best-fitting" (five-parameters) regression equation for the ln-transformed outdoor concentration of EC (LnEC) was obtained by "stepwise" selection:

$$\mathrm{LnEC} = -2.48 + 634.99 \times \mathrm{refinery\_inv} + 78.04 \times \mathrm{truck\_inv} + 0.01 \times \mathrm{RH\%} + 0.32 \times \mathrm{stability} \quad (3)$$

The meteorological variables selected in the "best-fitting" model are the relative humidity (RH%) and the atmospheric stability (stability). RH% is the most significant factor affecting LnEC (partial R^2 =0.24) and similar to atmospheric stability it has a positive coefficient. Although the relationship between EC and RH% was statistically significant, the reason for the positive association with relative humidity is not well understood. Interestingly, precipitation was negatively associated with LnOC but not associated with LnEC. This is consistent with the findings of others that indicate a longer atmospheric lifetime for EC than OC, presumably because wet scavenging removes OC more effectively from the atmosphere (Lim et al., 2003). The inverse distances to a major oil refinery (refinery_inv) located in Linden (NJ) and to the truck loading and unloading area (truck_inv) on Division Street (Elizabeth, NJ) were also selected in the model and have positive coefficients.

This is consistent with the knowledge that EC is a good tracer of diesel exhaust and combustion processes in general (Schauer et al., 1999a, b; Schauer, 2003). The overall model is statistically significant ($P < 0.0001$), has an R^2 of 0.40, and a *C*p of 5, the same as the predicted number of parameters. All parameter estimates have met a 0.15 significant level for entry into the model ($P < 0.15$). It is quite possible that associations with proximity to roadways would be seen given a larger database or if the analyses were conducted with hourly data enabling the consideration of wind direction and diurnal variations in emissions.

"Best-fitting" models for LnBenzo-[ghi]-Pyrene and LnCoronene—The "best-fitting" regression equations for the ln-transformed outdoor concentrations of particle-phase Benzo-[*ghi*]-Pyrene and Coronene (LnB-ghi-P and LnCOR, respectively) were obtained by "stepwise" selection:

$$\mathrm{LnB-ghi-P}=21.93+122.83\times F11\_\ \mathrm{inv}-0.38\times U-0.09\times \mathrm{K}+0.71\times \mathrm{stability} \quad (4)$$

$$\mathrm{LnCOR}=24.61+133.36\times \mathrm{F11\_\ inv}-0.41\times U-0.10\times \mathrm{K}+0.81\times \mathrm{stability} \quad (5)$$

The ln-transformed concentrations of these two p-PAHs (both emitted from combustion sources and used as mobile source tracers) were positively associated with inverse distance to interstate roadways (F11_inv). The same meteorological variables were selected in both models, with regression coefficients of both models having the same sign and a similar magnitude. This is expected as Benzo-[*ghi*]-Pyrene and Coronene have similar chemical/physical properties and atmospheric behavior. The order of selection of the predictor variables was also the same for both p-PAHs: temperature (*K*), wind speed (*U*), inverse distance to interstate roadways, and atmospheric stability (stability). The coefficients of determination (R^2) for LnCOR and LnB-ghi-P (0.67 and 0.66, respectively) were considerably larger than those obtained for PM_{25}, OC, and EC, suggesting that these p-PAHs have less diversity of sources and larger (percentage) source contributions from mobile sources. The two models are statistically significant ($P < 0.0001$) and have a *C*p of 5. All parameter estimates have met a 0.15 significant level for entry into the model ($P < 0.15$).

"Best-fitting" models for LnS and LnSe—The "best-fitting" regression equations for the ln-transformed outdoor concentrations of elemental S and Se (LnS and LnSe, respectively) were obtained by "stepwise" selection:

$$\mathrm{LnS}=4.33-0.11\times 0.11\times U-0.02\times K+0.02\times \mathrm{RH\%}+0.45\times \mathrm{stability} \quad (6)$$

$$\mathrm{LnSe}=1.1594-0.15\times U-0.05\times K+0.02\times \mathrm{RH\%}+0.93\times \mathrm{stability} \quad (7)$$

S and Se were not correlated with any major or minor roadways or other examined local sources. This is expected as the major contributor to S and Se in New Jersey air is coal combustion in upwind states (Lee et al., 2002). This anticipated result gives further confidence to the quality of the other regression models obtained in this study. LnS and LnSe were associated with temperature (*K*), wind velocity (*U*), atmospheric stability (stability), and relative humidity (RH%). The two models are statistically significant ($P < 0.0001$), have an R^2 of 0.52 and 0.44 (for LnS and LnSe, respectively), and a *C*p of 5. All parameter estimates have met a 0.15 significant level for entry into the model ($P < 0.15$).

Effect of Proximity to Sources

When the predictor variables are not covariant, the coefficients in a multiple regression model estimate the change in the response variable per unit increase in the predictor variable when all other predictors are held constant (Netter et al., 1996; Kwon et al., 2006). As the multicollinearity for all models obtained in this study was not significant, it was possible to evaluate the effect of the individual predictors (distance to sources and meteorological variables) on the concentrations of $PM_{2.5}$, OC, EC, Benzo-[*ghi*]-Pyrene, and Coronene.

The relationships between the predicted residential outdoor concentrations of PM_2.5, OC, EC, Coronene, and Benzo-[*ghi*]-pyrene, and the distance to the mobile and area sources derived from the best-fit regression equations are illustrated in Figures 2–5. The predicted ambient air concentrations of all particulate species decreased with increasing distance from the sources. Atmospheric dispersion is probably the dominant process contributing to this rapid decrease in concentration with increasing distance from the emission sources (Zhu et al., 2002). The ranges for the meteorological conditions were 265–305 °K for temperature (*K*), 2.0–8.0 m/s for wind speed (*U*), 40–95% for RH%, and 0–84.1mm for precipitation. The relationships between the predicted concentrations and each meteorological variable are illustrated in Supplementary Figures SI 1 to SI 4 (see Supplementary Information for further details).

For urban interstate highways (F11), the decrease in predicted concentrations of $PM_{2.5}$, OC, Coronene, and Benzo-[*ghi*]-Pyrene (association of EC with F11 was not found) was rapid during the first 250 m, with little change beyond that distance. These findings are consistent with those obtained by Zhu et al. (2002) who found that the ultrafine particle number concentration decreased exponentially with increasing distance from a Los Angeles freeway (17, 20, 30, 90, 150m) and was indistinguishable from the background concentration 300 m downwind. For local urban roadways (F19), the decrease in predicted $PM_{2.5}$ concentrations (the only species where an association was found) was rapid in the first 20 m, with little change beyond 50 m. Many homes in the city of Elizabeth are particularly close to minor roadways. All RIOPA homes in Elizabeth were within 100m of an F19 roadway. The elevation of $PM_{2.5}$ concentrations in close proximity to F19 roadways is likely to reflect a combination of combustion-generated particles and fugitive $PM_{2.5}$ emissions, such as resuspended road dust and tire abrasion. An association with $PM_{2.5}$ and not with combustion tracers would be expected if road dust and other fugitive motor vehicle emissions are important contributors to elevated concentrations within 20 m of F19 roads.

Under the median atmospheric conditions observed in this study (i.e., median values of the meteorological variables in the model), the predicted concentrations of $PM_{2.5}$, OC, Coronene, and Benzo-[*ghi*]-Pyrene for samples collected closest (37m) to the interstate highways (F11) were 1.6, 2.5, 21.4, and 17.0 times higher, respectively, than the corresponding concentrations predicted 250 m away. For $PM_{2.5}$, the predicted concentration for samples 9 m from an urban local roadway (F19) was 1.5 times higher than that 100 m from the same source. Enhancements were quite small for homes 100m from F11 (1.1, 1.3, 2.2, and 2.1 times higher for $PM_{2.5}$, OC, Coronene, and Benzo-[*ghi*]-Pyrene, respectively) and 25m from a local urban roadway (1.1 times higher for $PM_{2.5}$). Surprisingly, no association was found between the distance from the major urban arterials (Route 1 and 9; here categorized as F14) and the concentrations of any of the examined particulate species. A significant association was expected at least for EC, Coronene, and Benzo-[*ghi*]-Pyrene because F14 has heavy diesel truck traffic. Also, the lack of a significant association between distance to interstate roadways (F11) and EC (as seen for $PM_{2.5}$, OC, Coronene, and Benzo-[*ghi*]-Pyrene) is somewhat puzzling. However, a lack of statistically significant association does not imply that a relationship between a particulate species (e.g., EC) and distance to sources (e.g., F11 and F14) does not exist. It is quite possible that EC

associations with proximity to roadways would be seen in a larger database (i.e., more homes adjacent to Routes 1 and 9) or in an analysis using hourly data.

The large truck loading and unloading area on Division Street enhanced predicted $PM_{2.5}$ and EC concentrations as far away as 3000 and 1000 m, respectively. An association over such a long distance was not expected, and might occur because a single location was used to describe emissions distributed over a large surface, and/or because of truck traffic into and out of the considered area. Thus, the "distance" between this area source and the RIOPA homes has a large uncertainty. The predicted concentrations of $PM_{2.5}$ and EC for samples closest to the truck loading and unloading area on Division Street (192 m) were 1.3 and 1.4 times higher, respectively, than those predicted 3000 and 1000m from the same source.

Predicted concentrations of EC were also associated with the distance from a major oil refinery (refinery) located in Linden (NJ). In this case, a rapid decrease in the predicted concentration of EC occurred within the first 1000m. The predicted EC concentration 192 m from the refinery (22.8 $\mu gC/m^3$; distance of the closest study home) was 14.2 times higher than the corresponding concentration 1000 m from the same source. EC was still a factor of 2 higher than the "urban background" 500m from the refinery. This source appears to have a dramatic effect on EC, a minor component of the $PM_{2.5}$ mass but an important tracer for combustion-generated organic PM. The distance from the refinery in Linden (NJ) was not associated with the predicted concentrations of any other examined species.

Proximity and meteorological variables explained 47, 33, 40, 66, 67, 52, and 44% of the ln-transformed residential outdoor $PM_{2.5}$ mass, OC, EC, Benzo-[*ghi*]-Pyrene, Coronene, S, and Se concentration variability, respectively. This explanatory power is reasonable given the 48-h sample collection time. Coefficients of determination generally increase with improved time resolution (e.g., 1 h; Shi and Harrison, 1997). *P*-values and partial R^2 for parameter estimates were more significant for the meteorological variables (in particular atmospheric stability) than for the proximity variables, reflecting the large role that meteorology has in air quality. Meteorological variables accounted for at least 80% of the total explained variance for all examined particulate species.

Limitations

The important limitations of this study are as follows. The variables considered in the regression models are subjected to differing amounts of measurement error. In particular, we expect that the averaging (or use of median values) of meteorological variables over 48 h introduces far greater measurement error than that contained in the distance measures (although hourly variations in emissions levels will also introduce measurement error). Despite this, the selected meteorological variables still tend to explain more of the variance in PM species concentrations than the distance variables. Second, although the relationships between PM species concentrations, source proximity, and meteorology might be expected to vary seasonally, we were not able to examine this with these techniques in this data set. Finally, this analysis made use of 48-h average concentrations. We expect that the measurement of these PM species with 1-h resolution would be more insightful.

Discussion and conclusions

In this and virtually all similar studies, the outdoor concentrations of primary pollutants (e.g., PAHs, CO, BC or EC, and ultrafine particles) are more strongly correlated (inversely) with distance from emission sources than the outdoor concentrations of species formed in the atmosphere (e.g., fine PM, S, and OC). The elevation of $PM_{2.5}$ and OC mass concentrations outside the study homes closest (9–192 m) to primary PM sources was modest (1.6–2.5 times the background levels). The elevation of EC and p-PAH concentrations outside the closest study homes was substantial for some sources (i.e., about 20 times for p-PAHs with F11, and about 14 times for EC with refinery). This is consistent with the knowledge that EC and PAHs are emitted from primary combustion sources, whereas $PM_{2.5}$ mass and OC have both local primary sources and more regional secondary sources. For homes 250m or more from F11 roadways, concentrations were indistinguishable from the urban pollution mix, while the effect of the refinery on EC persisted to about 1000 m.

It should be noted that urban-industrial cities, such as Elizabeth, have many homes in quite close proximity to industrial facilities, transportation hubs, and congested roadways. Approximately 15 and 40% of the Elizabeth population lives within 100 and 250 m, respectively, of interstate roadways (F11). This suggests that a meaningful fraction of the Elizabeth population is exposed to the levels of $PM_{2.5}$ and p-PAHs that are elevated above the urban mix because of residential proximity to interstate roadways. The model results presented here indicate that the oil refinery in Linden (NJ) might dramatically increase EC exposure for Elizabeth residents living in close proximity of the refinery (the predicted EC concentrations 192 and 500 m from this source were 22.8 and 3.02 $\mu gC/m^3$, compared with an urban background of 1 $\mu gC/m^3$). Approximately 5% of the Elizabeth population lives within 200 m of the refinery. In addition, 3% and 7% of Linden residents live within 1000 and 1500m of this source.

It should be noted that the refinery and other point and area sources also contribute to the urban pollution mix (urban background), enhancing urban pollution concentrations above the upwind, regional pollution signal. For Newark, a larger nearby city, urban sources account for roughly 30% of total $PM_{2.5}$ (Chuersuwan, 2001).

As residential outdoor air penetrates indoors, an increase in the outdoor concentration of these PM species at homes near emission sources is expected to result in an increase in personal exposure for individuals living in those homes (Meng et al., 2005, 2007; Weisel et al., 2005). The majority of $PM_{2.5}$ inside RIOPA study homes was found to be of outdoor origin (Meng et al., 2005), and residential indoor PAHs and EC were almost entirely of outdoor origin (Naumova et al., 2002; Polidori et al., 2006a). Because the local emission sources examined in this study also contribute to the “urban background” aerosol (USEPA, 2004), steps taken to decrease source emissions will also lower urban background levels of $PM_{2.5}$.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgements

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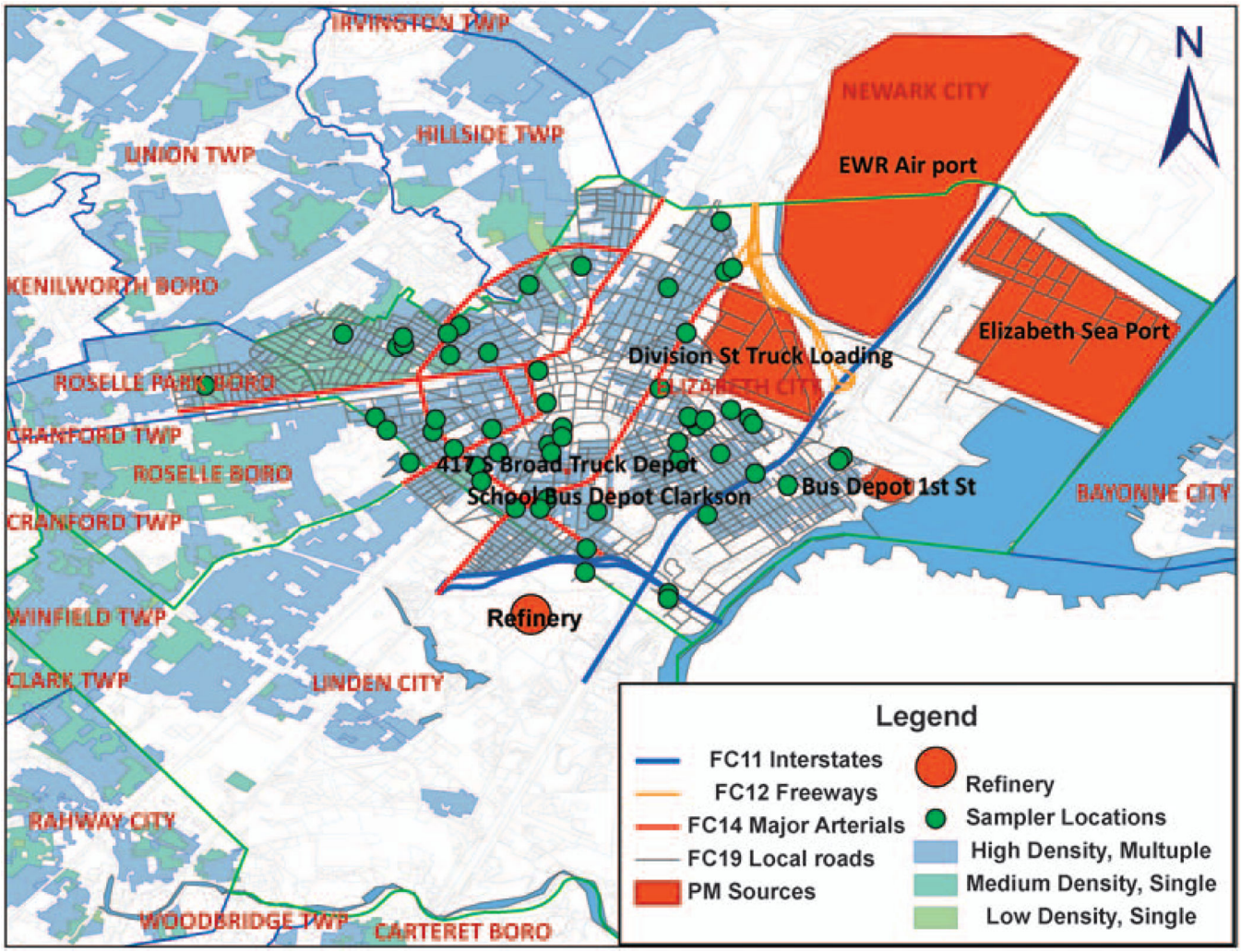

Figure 1.

Identified $PM_{2.5}$ point and area sources in Elizabeth (NJ). Outdoor residential sampler locations have been reported as a green circle to protect the confidentiality of participants.

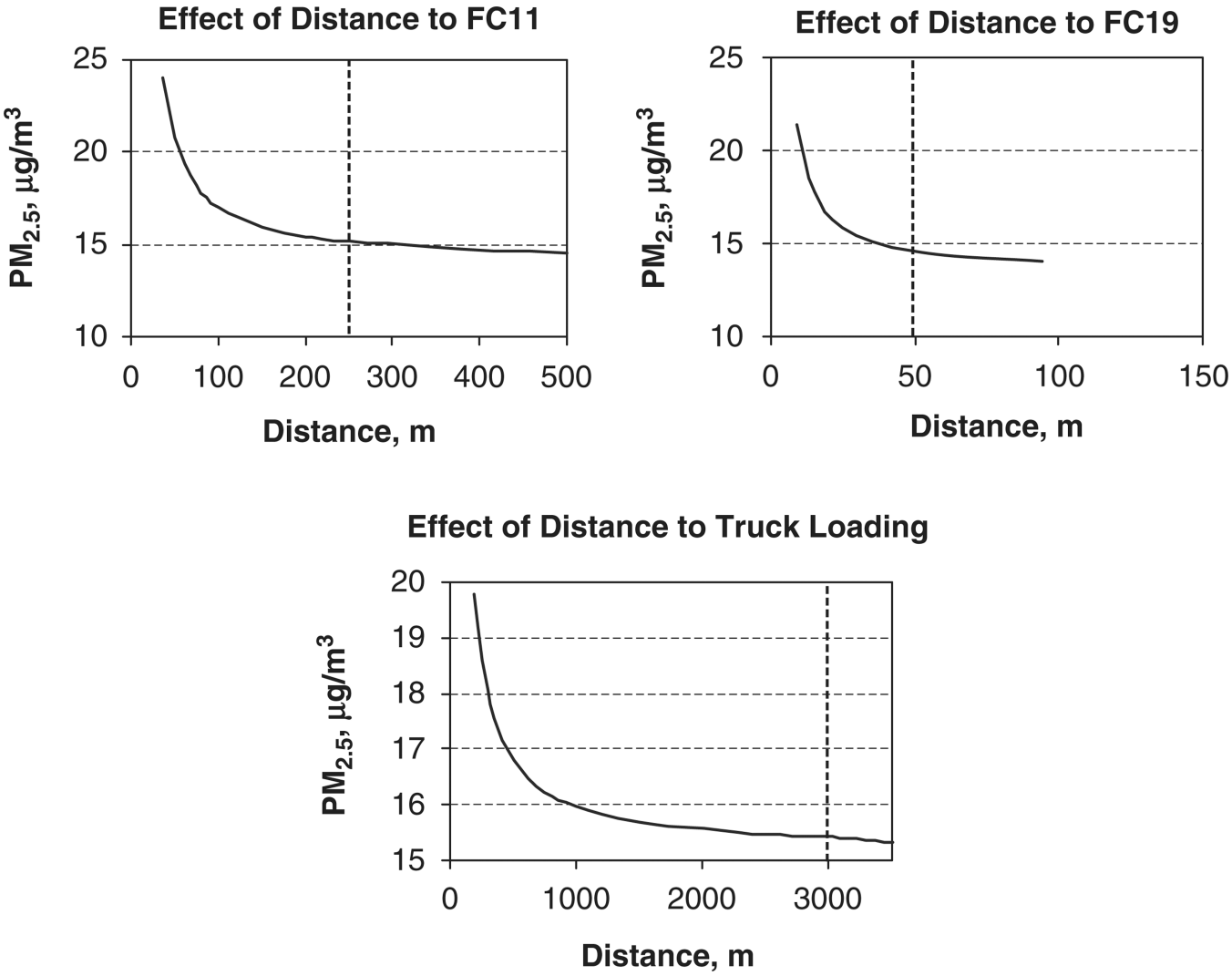

Figure 2.

Simulated residential ambient air concentrations of fine particulate matter ($PM_{2.5}$) ($\mu g/m^3$) with distance to interstate and local roadways (F11 and F19, respectively), and to truck loading and unloading areas estimated from the best-fitting regression model.

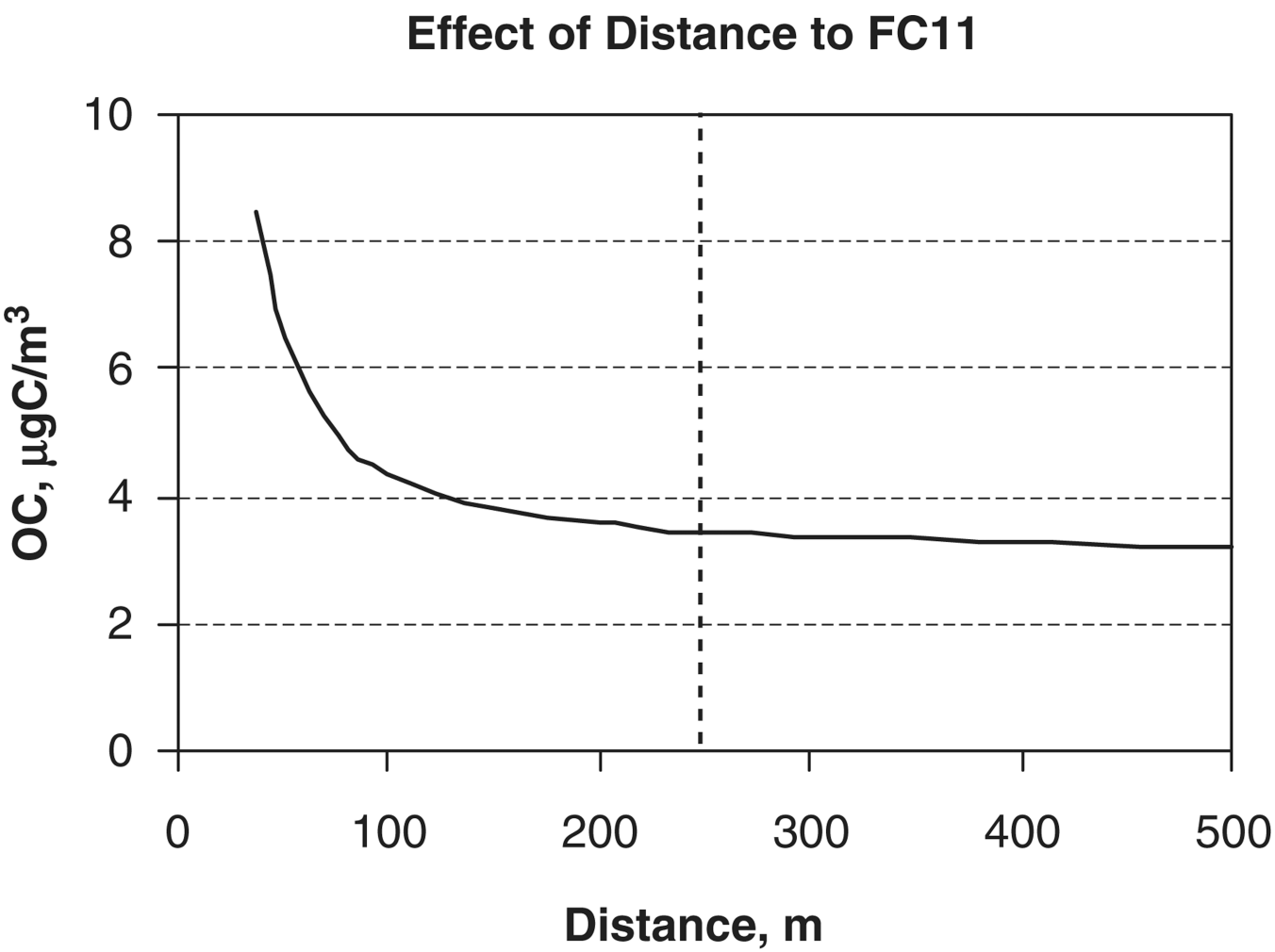

Figure 3.
Simulated residential ambient air concentrations of organic carbon (OC) (µgC/m^3) with distance to interstate roadways (F11) estimated from the best-fitting model.

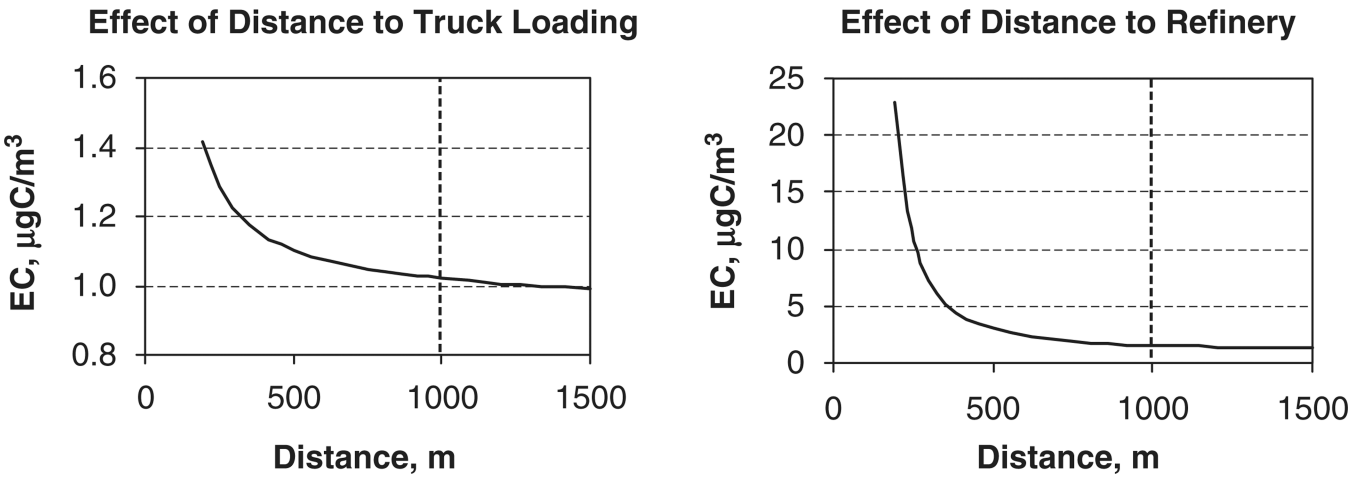

Figure 4.

Simulated residential ambient air concentrations of elemental carbon (EC) (µgC/m^3) with distance to truck loading and unloading areas, and to a major refinery estimated from the best-fitting regression model.

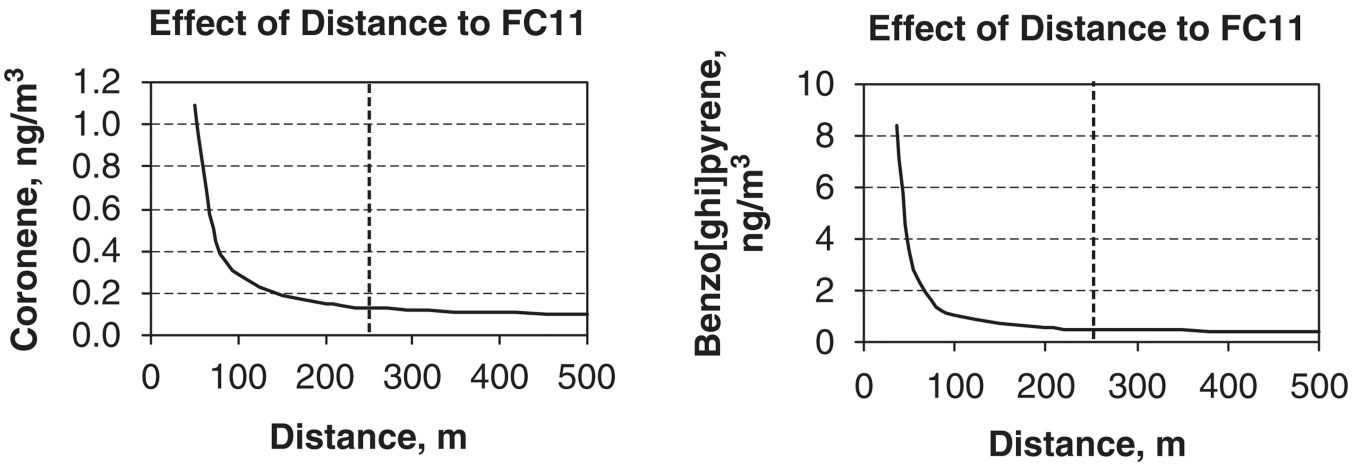

Figure 5.

Simulated residential ambient air concentrations of Coronene and Benzo-[*ghi*]-pyrene (ng/m^3) with distance to interstate roadways (F11) estimated from the best-fitting regression models.

Table 1

Number of samples (*N*) analyzed for each species considered in this study.

$PM_{2.5}$			OC, EC			COR and B-*ghi*-P			S and Se		
N_1	N_2	N^a	N_1	N_2	N^a	N_1	N_2	N^a	N_1	N_2	N^a
25	39	103	28	16	60	30	10	50	30	23	76

[a] $N = 2 \times N_2 + N_1$ (N_1 and N_2 are the number of homes that were sampled once and twice, respectively, during RIOPA).

Table 2

Bivariate Pearson's correlations between ln-transformed outdoor concentrations and predictor variables.

Predictor	CC	*P*	*N*
LnPM$_{2.5}$			
U	−0.50	<0.0001	103
RH	0.39	<0.0001	103
Stab	0.56	<0.0001	103
MixH	−0.26	0.01	103
F19	0.23	0.02	103
F16	0.22	0.03	103
F11	0.21	0.03	103
LnOC			
Stab	0.51	0.001	60
U	−0.34	0.01	60
F16	0.29	0.04	60
LnEC			
RH	0.49	0.001	60
Stab	0.43	0.001	60
MixH	−0.34	0.01	60
U	−0.33	0.02	60
F16	0.29	0.03	60
Precip	0.29	0.03	60
F11	−0.28	0.04	60
LnS			
RH	0.50	<0.0001	76
Stab	0.42	0.001	76
U	−0.34	0.001	76
Precip	0.29	0.01	76
LnSe			
Stab	0.54	<0.0001	76
MixH	−0.28	0.02	76
RH	0.28	0.02	76
U	−0.26	0.03	76
LnB-ghi-P			
F17	−0.44	0.001	50
K	−0.42	0.001	50
Stab	0.40	0.01	50
MixH	−0.29	0.05	50
LnCOR			
Stab	0.44	0.001	50
F17	−0.42	0.001	50
K	−0.42	0.001	50

Predictor	CC	*P*	*N*
MixH	−0.31	0.04	50

CC = Pearson's coefficient of correlation; *P* =*P*-value; *N* = sample size; *U* = wind speed (m/s); Stab = stability (stability class); RH = relative humidity (%); *K* = temperature (°K); Precip = precipitation; MixH = mixing height (Km); F11 = distance to urban interstate highways; F12 = distance to urban freeways and expressways; F14 = distance to urban major arterials; F16 = distance to urban minor arterial; F17 = distance urban collector; F19 = distance to urban local roadway.

Table 3

Best-fit regression models for all particulate species examined in this study.

Pollutant	Row heading	Model intercept	Source				Meteorological variable		
$PM_{2.5}$ (model R^2 = 0.47)[a]	X_i		FC11 $^{-1}$	FC19 $^{-1}$	Truck^{-1}	Stab	*U*		
	β_i (SE)	1.0 (0.6)	20 (11)	4.2 (1.7)	51 (30)	0.43 (0.09)	−0.13 (0.04)		
	P-R^2		0.016	0.052	0.016	0.32	0.066		
[a]EC (model R^2 = 0.40)	X_i		Rer^{-1}		Truck^{-1}	Stab	RH		
	β_i (SE)	−2.5 (0.6)	630 (26)		78 (36)	0.32 (0.13)	0.01 (0.004)		
	P-R^2		0.033		0.050	0.078	0.24		
[b]OC (model R^2 = 0.33)	X_i		FC11^{-1}			Stab	Precip		
	β_i (SE)	−2.2 (0.7)	39 (21)			0.66 (0.14)	−0.01 (0.006)		
	P-R^2		0.043			0.25	0.04		
[a]Coronene (model R^2 = 0.67)	X_i		FC11^{-1}			Stab	*K*	*U*	
	β_i (SE)	24 (4)	133 (42)			0.81 (0.28)	−0.10 (0.01)	−0.41 (0.10)	
	P-R^2		0.091			0.06	0.28	0.24	
[a]Benzo-[*ghi*]-pyrene	X_i		FC11^{-1}			Stab	*K*	*U*	
(model R^2 = 0.66)	β_i (SE)	22 (4)	123 (38)			0.71 (0.26)	−0.09 (0.01)	−0.38 (0.10)	
	P-R^2		0.094			0.06	0.26	0.25	
[a]Sulfur (model R^2 = 0.52)	X_i					Stab	*K*	*U*	RH%
	β_i (SE)	4.3 (2.2)				0.45 (0.11)	−0.02 (0.008)	−0.11 (0.05)	0.02
	P-R^2					0.14	0.23	0.04	0.22
[a]Selenium (model R^2 = 0.41)	X_i					Stab	*K*	*U*	RH%
	β_i (SE)	1.2 (3.5)				0.93 (0.19)	−0.15 (0.08)	−0.02 (0.007)	0.02
	P-R^2					0.29	0.03	0.03	0.03

X_i=*i*th predictor variable; β_i= parameter estimate of *i*th predictor; SE = standard error of parameter estimates; P-R^2= partial R^2 of the variable; FC11^{-1}= inverse distance to urban interstate highways; FC19^{-1}= inverse distance to urban local roadway; Ref^{-1}= inverse distance a major refinery in Linden (NJ); Truck^{-1}= inverse distance to a major truck loading/unloading area in Elizabeth; Stab = atmospheric stability; *K*= temperature; *U* = wind speed; Precip = precipitation; RH% = relative humidity; $PM_{2.5}$= fine particulate matter; OC = organic carbon; EC = elemental carbon.

[a]Model determined by using a stepwise selection (P<0.15).

[b]Model determined by using a forward selection (P<0.50).