

A spatial-temporal regression model to predict daily outdoor residential PAH concentrations in an epidemiologic study in Fresno, CA

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ABSTRACT

Background: Polycyclic aromatic hydrocarbons (PAHs) are generated as a byproduct of combustion, and are associated with respiratory symptoms and increased risk of asthma attacks.

Objectives: To assign daily, outdoor exposures to participants in the Fresno Asthmatic Children's Environment Study (FACES) using land use regression models for the sum of 4-, 5- and 6-ring PAHs (PAH456). **Methods:** PAH data were collected daily at the EPA Supersite in Fresno, CA from 10/2000 through 2/2007. From 2/2002 to 2/2003, intensive air pollution sampling was conducted at 83 homes of participants in the FACES study. These measurement data were combined with meteorological data, source data, and other spatial variables to form a land use regression model to assign daily exposure at all FACES homes for all years of the study (2001–2008).

Results: The model for daily, outdoor residential PAH456 concentrations accounted for 80% of the between-home variability and 18% of the within-home variability. Both temporal and spatial variables were significant in the model. Traffic characteristics and home heating fuel were the main spatial explanatory variables.

Conclusions: Because spatial and temporal distributions of PAHs vary on an intra-urban scale, the location of the child's home within the urban setting plays an important role in the level of exposure that each child has to PAHs.

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1. Introduction

The majority of health effect research focused on daily exposure to ambient pollutants has used a single central monitor to assign exposure to participants (Peel et al., 2005; Penttinen et al., 2001), relying on the assumption that temporal variability overshadows spatial variability such that exposure misclassification is negligible. When incorrect, this assumption could lead to exposure misclassification of spatially heterogeneous pollutants and result in significant differences in the correlation to the health outcome (Sarnat et al., 2010; Wilson et al., 2005, 2007). The degree of

Abbreviations: DEP, Diesel exhaust particles; EPA, United States Environmental Protection Agency; FACES, Fresno Asthmatic Children's Environment Study; FL, Fuel loading factor for agricultural burning; HI, Home intensive; IARC, International Agency for Research on Cancer; LUR, Land use regression; PAH, Polycyclic aromatic hydrocarbons; PAH456, Sum of the PAHs with 4-, 5-, or 6-rings; SJVUAPCD, San Joaquin Valley Unified Air Pollution Control District.

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heterogeneity of a pollutant's spatial distribution can be tested by examining the absolute concentrations, correlation coefficients, and the coefficient of divergence between measured data at different sites (Wilson et al., 2005). Using these techniques, researchers have found many air toxics, including PAHs, with enough heterogeneity that using a single monitor would cause some degree of exposure misclassification (Lehndorff and Schwark, 2004; Levy et al., 2001). If the distribution of PAHs is heterogeneous within the study area and some spatially resolved measurement data are available, modeling the spatial distribution may be preferable to using a central monitor value directly (Ito et al., 2004). However, in a moderate to large urban area for a cohort epidemiology study collecting personal PAH samples or even a large number of cross-sectional samples is not feasible. The intent of this paper is to model the daily individual exposures to outdoor residential PAHs, over 8 years of follow-up, through land use regression (LUR) modeling.

PAHs are a class of compounds characterized by fused aromatic rings that form when organic matter undergoes incomplete combustion. PAHs generally exist in complex mixtures of combustion products

such as diesel exhaust, soot, and wood and tobacco smoke. They exist in ambient air as gases (gas-phase) and adsorbed to particulate matter (particle-phase or particle-bound). PAHs are produced by both diesel and gasoline fuel combustion (Cadle et al., 1999; Marr et al., 1999; Riddle et al., 2007; Rogge et al., 1993), as well as biomass burning (Jenkins et al., 1996; Schauer and Cass, 2000). However, PAHs are not merely another proxy for traffic exhaust, they are well-known as carcinogens (International Agency for Research on Cancer, 1989) and toxic air contaminants (Office of Environmental Health Hazard Assessment, 2001). PAHs have most recently been implicated in short-term respiratory health outcomes (Delfino, 2002; Diaz-Sanchez et al., 1994) and immunological functioning related to mechanisms involved with asthma and atopy (Finkelman et al., 2004; Nadeau et al., 2010; Perera et al., 2009). Despite this increasing interest in health effects related to exposure to ambient concentrations of PAHs, to our knowledge, no data exist on daily intra-urban spatial distributions or individual exposure estimates in the context of an epidemiologic cohort study.

LUR models are spatial regression models that relate location-specific data on pollutant concentrations to location-specific source and environment data using regression (Briggs et al., 1997). Unlike interpolation methods, such as kriging, LUR models are able to exploit measurement data to build a smooth pollutant surface even when there are significant local sources and intra-urban variability (Jerrett et al., 2005). The majority of LUR models and spatial models for air pollution exposures related to health effects has focused on modeling annual average exposure to NO₂, CO, or particulate matter (Hoek et al., 2008). While PAHs share some emission sources with these pollutants, the spatial distributions of these three pollutants are not identical (Fischer et al., 2000; Levy et al., 2001; Sarnat et al., 2010).

2. Methods

2.1. Study background and population

The combination of Fresno's geographic location and meteorology contributes to very poor air quality in both the summer and

the winter (Blanchard et al., 1999). Because of the Southern Sierra Nevada in the West, the Tehachapi Mountains on the south, and the Coastal Range Mountains in the East, the San Joaquin air basin (in which Fresno is located) does not have an outlet for air pollution. Additionally, during the winter months, inversion layers from lowered mixing heights cause stagnation in the valley air. Main sources of PAHs in Fresno are the freeways, major arterials, agricultural burns, residential fireplace and woodburning stove use, and heating (Schauer and Cass, 2000). Fresno has three major freeways, but US Route 99 on the western edge of the city, is heavily used by truck traffic through the region whereas centrally-located State Route 41 is primarily local, light-duty vehicles (Margolis et al., 2009).

The Fresno Asthmatic Children's Environment Study (FACES) focused on quantifying the relationship between air pollution and the natural history of asthma symptoms and lung function growth in 315 young children, ages 6–11 years at entry, with asthma who reside in Fresno, California. Further details on the design of the study and cohort characteristics have been published elsewhere (Mann et al., 2010; Margolis et al., 2009; Nadeau et al., 2010). The participants in FACES were contacted every 3 months to collect health data and confirm residential address. From 2/2002 through 2/2003, a substudy of 83 homes of FACES participants, the home intensive (HI) substudy, was conducted to collect more detailed pollution and activity data from participants (Fig. 1). These participants were selected based on residential location in relation to anticipated traffic exposure (low or high) and smoking status of parents (non-smoking).

2.2. Field sampling

Two field sampling methods were used to characterize airborne concentrations of ambient PAHs for modeling daily exposures. First, a real-time monitor of particle-bound PAHs, the PAS2000 (EcoChem Analytics, League City, TX) collected data from October 2000 through September 2008 at the EPA Supersite in Fresno (Fig. 1). The PAS2000 uses a photoelectric aerosol sensor to measure the levels of particle-bound ambient PAH with three rings or greater.

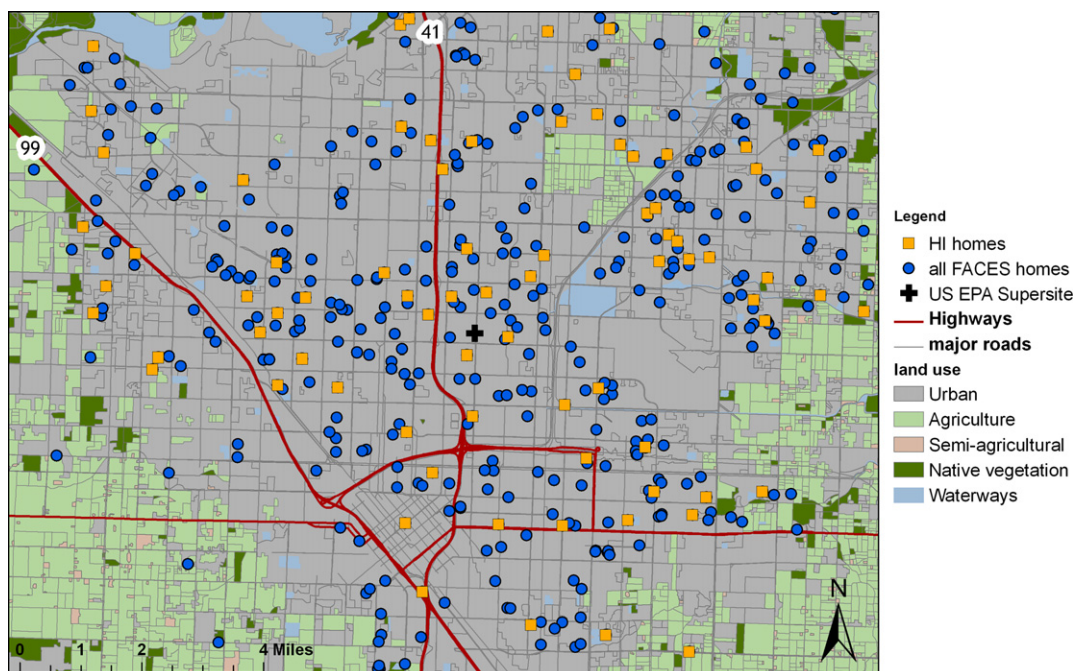


Fig. 1. Locations of FACES participants (blue circles and orange squares), HI sub-study participants (orange squares), and the US EPA Supersite.

The second sampling method collected PAHs on pre-baked quartz fiber filters (PallFlex Tissue Quartz) that were impregnated with XAD4 resin. Two filters in series per sample were collected using a Harvard-type impactor with a PM10 inlet and airflow of 10 l min^{-1} . These samples were collected during the HI substudy, 2/2002–2/2003, outside the 83 selected homes. The PAH samplers at the homes were placed in the backyard of the residence away from porches or overhangs and preferentially in the middle of the yard. Samples were collected for 24 h (8–8pm) on five days within each two-week panel study. There were 23 panels during the substudy, and 28 of the 83 homes participated in more than one panel. Field blank filters were collected at the same time.

The PAS2000 and 2 integrated filter samplers were co-located to validate the inter-comparability of the two methods. There were 27 days of sampling split between the warm and cool season, resulting in 44 paired sets of filter-PAS2000 samples. These samples were also used to select the best and most inclusive grouping of PAHs. As discussed in Section 2.3, the filter analysis provides concentrations for 14 PAHs. We wanted to select the group of PAHs that included the maximum number of PAHs and had a good correlation coefficient when compared to the PAS2000 concentration.

2.3. Laboratory and data analysis

The PAS2000 values were collected at 1-min intervals. These 1-min intervals were integrated into 24-h average concentrations to match the sampling period of the HI filter samples.

Filter samples were extracted by sonication in three aliquots of dichloromethane followed by vacuum filtration. The extracts were concentrated for analysis under nitrogen. Analyses were performed on a Hewlett Packard model 6890 Gas Chromatograph equipped with a 30 m (50%-Phenyl)-methylpolysiloxane fused silica capillary column and a 5972 Mass Selective detector operating in the selected ion-monitoring mode for enhanced sensitivity. The filters were analyzed and blank-corrected for 14 PAHs – acenaphthylene, acenaphthene, fluorene, anthracene, phenanthrene, fluoranthene, benz[a]anthracene, chrysene, benzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[ghi]perylene, indeno[1,2,3-cd]pyrene, and dibenz[a,h]anthracene.

2.4. Temporal information

The PAS2000 measurement data collected at the EPA Supersite was used as the primary independent variable for measuring temporal variability. To refine the estimates of temporal variability, meteorological variables describing hourly wind speed, hourly wind direction, daily wind trajectory, daily mean temperature, daily mean precipitation, and daily mean relative humidity (%) were also included. Hourly wind speed and direction were measured at the EPA Supersite. From the wind speed and direction data, wind trajectory and wind recirculation factors were calculated. Wind recirculation varied from 0 to 1 and calculated as: $1 - (\text{Net/Total transport distance})$ (Allwine and Whiteman, 1994). The net transport distance is the resultant distance that a given wind parcel traveled during the time of observation. The total transport distance is the total distance that the wind parcel traveled during the time of observation. A low value indicates well-developed airflow and transport. The daily mean precipitation was measured at California State University, Fresno. Temperature, relative humidity (%), and select hourly wind data were measured at both the EPA Supersite and at two other fixed sites within the Fresno/Clovis Metropolitan area (Clovis and Sierra Sky Park).

In addition to measurement data, seasonal variables were assigned based on date of sample collection and average weekly concentrations of PAH at the EPA Supersite. A binary variable was

used to indicate if the sample was collected on a weekday (Monday through Friday) or on the weekend (Saturday or Sunday).

2.5. Geographic information

Geographic information was collected, compiled and processed in ArcGIS 9.2 (ESRI, Redlands, CA) and, when necessary, further data calculations were completed in SAS 9.1 (SAS, Cary, NC).

2.5.1. Traffic

Two datasets were used to define traffic-related spatial variables: roadway locations from the TeleAtlas MultiNet™ USA (TAMN) roadway database and vehicle activity data from the California Department of Transportation (Caltrans). The vehicle activity data were GIS-based and contained estimates of annual average daily traffic (AADT) volumes traveling both directions on select road segments, and truck-traffic-volumes for freeways and state freeways (Margolis et al., 2009).

Five categories of traffic variables were defined: roadway proximity, roadway density, traffic intensity, and home location in relation to school bus exposures. Roadway proximity measured the distance from the homes to the nearest of each of five major road types (freeway, major arterial, minor arterial, major collector, and minor collector). Freeways are high-capacity, high speed multi-lane roadways with limited access to other roadways. Arterial roads are large-capacity city roads, connecting collector and local roads to freeways. Collector roads are smaller and slower than arterials, in general, and provide connection between local roads and arterial roads, in addition to access to residential properties. Roadway density was defined as the sum of total roadway length of each of the five major road types within each of 5 circular buffers (100 m, 200 m, 300 m, 400 m, and 500 m). Traffic intensity at each participant home was calculated based on AADT counts from Caltrans, assuming Gaussian decay of exhaust emissions with distance to the roadway (Wilhelm and Ritz, 2003). Last, the impact of diesel school buses was represented using the distance to the nearest elementary school as a proxy for exposure to bus exhaust. Elementary school proximity was used to capture this source in Fresno, because elementary schools are used as primary bus stops for the school district, not children's homes. School buses do not have established routes, but vary their route depending on the driver and traffic conditions (Fresno Unified School District, personal communication, 2005).

2.5.2. Land use

Land use data were obtained from the California Department of Water Resources county-wide California Land & Water Use surveys from Fresno County (2000) and Madera County (2001). Land use types used were urban, vacant urban (including parking lots), landscaped urban, residential urban, commercial, industrial, agricultural, semi-agricultural, native vegetation, and native riparian. To describe the participant's neighborhood, each participant home was assigned three sets of values: land use type on which the participant's home was located; land use types within 5 circular buffers (radii 100 m, 200 m, 300 m, 400 m, and 500 m) near the home; and total land use area, by type, within the same 5 buffers around the home.

2.5.3. Agricultural burning

We obtained agricultural burning data for Fresno and Madera counties from the San Joaquin Valley Unified Air Pollution Control District for 2000 through 2007. The records of approved agricultural burn permits contain information on date, location, and type and amount of materials burned. Permits without location information, 9% of total, were excluded from our analyses. Two types of variables

were formed for 4 circular buffers of radii 5 miles, 10 miles, 15 miles and 20 miles (8 km, 16.1 km, 24.1 km, 32.2 km): daily count of agricultural burns and total tons burned within each buffer.

2.5.4. Neighborhood variables

Data from the United States 2000 Census SF3 dataset was selected for transportation, home fuel use, or socioeconomic characteristics. All blockgroup and census tract variables were assigned using location of residence. Additionally, neighborhood data describing house characteristics from the FACES datasets and spatial location are included as part of this subset.

2.6. Exposure modeling

2.6.1. Model form and selection

To assign daily PAH exposure for all participants in FACES for all days during the FACES study, LUR modeling was used. The residential outdoor PAH model used the filter measurement data from the HI substudy as the dependent variable and the PAS2000 measurement data at the EPA Supersite and other temporal and geographic information as the independent variables. Because the filter collection in the HI substudy contains repeated sampling during each two-week panel (up to 5 measurements/home), a mixed-effects regression model was used as the final form of the model. Potential determinants of residential outdoor PAH concentration were treated as the fixed effects, and sample location was treated as a random effect. The form of the model was:

$$Y_i = u + B1 + B2 + \dots + Bk + X_i + e_i$$

where Y_i is the concentration of residential outdoor PAH measured at the i th location, u is the true underlying mean of residential outdoor PAH averaged overall strata, $B1, B2, \dots, Bk$, are fixed effects of k determinants, X_i is the random effect of the i th location, and e_i is the random error.

Because of the large number of variables, prior to fitting the model manually we used two methods to improve model fit. First, we removed variables with little or no variability from the pool of candidate variables. Binary variables were required to have at least 10% of the observations in one category. Continuous variables were required to show a histogram with at least two stratifications where at least 10% of the observations were in the smaller strata. Second, the forward stepwise selection algorithm in PROC REG in SAS 9.1 (Cary, NC) was used to reduce the dimensionality of the model. Candidate independent variables were run in 11 subject matter groupings (PAH measurement at the central site, wind speed, wind direction at the EPA Supersite, wind direction at other fixed sampling sites, other meteorological characteristics, traffic, land use, agricultural burning, neighborhood and housing characteristics, day of the week of sampling and season) with the residential outdoor PAH concentration as the dependent variable. The entry alpha value was set to 0.10 and the alpha to stay value to 0.15. Variables selected from each of these groups were run manually in PROC MIXED. Mixed modeling does not allow for R^2 calculation, instead we used the statistical significance of each parameter estimate and the change in the variance component estimates to guide inclusion and exclusion from the model. Additionally, we confirmed that there was no collinearity between the final selected covariates. The temporal autocorrelation of the residuals, spatial autocorrelation of the residuals using Moran's I , examination of standardized residuals (using the Cholesky decomposition), examination of the index of agreement (IOA) and the root mean squared error (RMSE) from leave-one-out cross-validation were used to evaluate model fit and performance.

To check the residuals in the model, they were transformed to have a constant variance and zero correlation based on the Cholesky decomposition. This transformation was done with SAS PROC MIXED with the VCIRY option. The transformed residuals were tested for normality through examination of the histogram and the normal quantile–quantile plot. The scaled residuals also were used to test that the model covariance assumptions were adequate. This was done informally by examination of a scatterplot of the absolute value of the transformed residuals versus the predicted filter PAH values. As with simple regression model, the residuals should show no systematic pattern. More formally, the overall fit of the covariance was tested by calculation of a smoothed plot of the empirical semi-variogram with time as the unit of lag.

Leave-one-out cross-validation was used to evaluate the final mixed model iteratively, leaving out one HI location at a time, since there were multiple observations at each HI location. Parameter estimates from each model were used to calculate the omitted location values, and normality of the distribution of the parameter estimates was evaluated. From the leave-one-out resulting data, the index of agreement (IOA) and the root mean square errors were calculated to evaluate the model goodness of fit (Willmott, 1982; Wilson et al., 2005). The IOA values span 0–1, with values over 0.5 considered to show a good model fit. The total, systematic, and unsystematic root mean squared errors (RMSE, RMSEs, RMSEu) were calculated to look at the error in the model. The RMSE is a measure of the total average difference between the observed and predicted. The RMSEs represents the error that is intrinsic to the model; i.e. that error which can be predicted mathematically. The RMSEu represents the potential accuracy of the model. The lower the total, systematic, and unsystematic RMSE, the better the model fit. All three metrics are in the units of the observed variable.

Moran's I statistic is a measure of spatial autocorrelation which reflects the level of spatial dependence in second order effects, i.e. deviations of the variable from its mean. It takes point locations as input and assumes these points are centroids representing underlying continuous area data. This is an appropriate assumption for air pollution distribution modeling since each sampling point is assumed to represent the true value of the underlying continuous surface of air pollutant concentration at that location. Moran's I varies from -1 to 1 , with -1 as uniformly distributed, $+1$ as highly clustered, and 0 as randomly distributed. If the Moran's I coefficient is significant, then the pattern of the values cannot be explained by random chance. The Moran's I coefficient was calculated for the residuals from the residential outdoor PAH model to ensure no residual spatial autocorrelation.

2.6.2. Individual exposure estimates

To estimate outdoor individual exposures to PAHs for each day of the FACES study for all participants, the models were applied to each day and each location in the study. The estimates generated with this method are daily outdoor concentrations of PAHs at each child's home. To apply the models, we used parameter estimates generated by the model in combination with the temporal and geographic data in our database to calculate the outdoor concentrations for each participant on each day.

3. Results

3.1. Study background and population

During the time from 1/1/2001 to 9/30/2008, there were 315 FACES participants who lived at a total of 465 residences, a consequence of the fact that some participants moved during the study (Fig. 1). These residences were geocoded using the TAMN roadway database. More than 97% of the residences matched an exact street

address in the database, and the remaining were geocoded with online mapping services or GPS coordinates from home visits.

3.2. Field sampling

The 24-h average concentrations of particle-bound PAHs at the EPA Supersite showed a strong seasonal oscillation during the eight years of available data (Fig. 2). Additionally, there is a statistically significant decreasing trend ($p = 0.007$) in winter-time particle-bound PAHs measured at the EPA Supersite over these eight years.

The 497 24-h PAH filter samples were collected on 126 days outside the 83 participants' homes during the HI study. There were 55 homes sampled in one season and 28 homes sampled in two seasons; up to 5 daily samples collected during each 2-week home visit. The range of homes concurrently sampled on each day of sampling was 1–6 homes.

The comparability between the co-located filter-based PAH samples and the PAS2000 was generally high (Supplemental section, Table 1). The most inclusive group correlation between the two methods was PAHs with 4-, 5-, or 6-rings (fluoranthene, benz[a]anthracene, chrysene, benzo[a]pyrene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[ghi]perylene, indeno[1,2,3-cd]pyrene, and dibenz[a,h]anthracene) with a Pearson correlation coefficient of 0.86. This summary variable, called PAH456, was used to build a model of outdoor residential PAH exposure.

3.3. Temporal information

Because of the topography of the San Joaquin Valley, there is no single prevailing wind direction, but instead a wind direction pattern throughout the day. The dominant 24-h pattern shows that wind measured at the EPA Supersite blows, on most days, from the northwest quadrant in the late afternoon to very early morning hours. During the daylight hours, the wind direction will often reverse and blow from the SW quadrant, but the direction is more variable from day-to-day than in the evening. Wind out of the NW would likely spread the vehicle emissions from U.S. Route 99 across the city. Wind out of the SW comes from a primarily agricultural landscape, with a possibility of spreading larger particles from dirt and dust. Wind speed is variable throughout the year, with summer months showing a higher average speed than winter months.

The weekly average 24-h PAH concentrations at the EPA Supersite were evaluated to find where the concentration rose to winter-time levels from the lower non-winter levels. This rise was defined as a sustained increase. A seasonal variable was defined using that week as the cutpoint: winter (11/22/2002 through 2/8/2003) and non-winter (2/2002 through 11/21/2002). When used in modeling exposures, this variable was generalized to November through February to reflect the winter season. There were 171 samples (34%) collected in the winter season, and 326 (66%) in the non-winter season.

Concentrations of the HI sub-study home samples collected on weekdays were not significantly higher than on weekends. However, given the study design this sample contains considerable spatial heterogeneity that may mask temporal differences. The PAS2000 concentrations from the US EPA Supersite show a statistically significant difference of 1.40 ng m^{-3} between weekdays and weekends. This likely reflects higher traffic intensity due to higher commuter traffic on weekdays.

3.4. Geographic information

Nearly half the HI sub-study homes were located on a major collector road (45%). However, the type of roadway with the greatest density near homes, for all buffer sizes, was the minor collector, with 89% of homes within 100 m of a minor collector. In contrast, major arterials were the least common roadway type for both density and proximity. Only 10% of HI sub-study homes were within 500 m of a major arterial.

Most of the participant homes were situated on urban land (95%). Only 23% lived within 500 m of agricultural land, despite the region's high agricultural presence. Additionally, despite the large number of agricultural burns reported in Fresno and Madera counties during the HI sub-study (6390 burns) only 42% of homes had any burning within 20 miles on the day of sampling.

3.5. Exposure modeling

3.5.1. Model selection

The natural logarithm of PAH456 concentrations best described the distribution of concentrations in Fresno and was used in model selection and the final model as the dependent variable. Bivariate

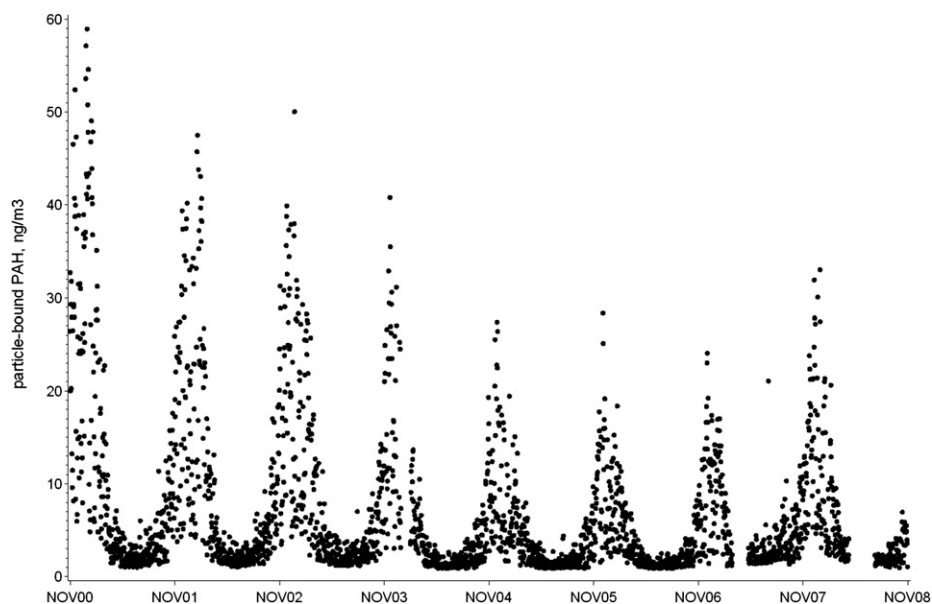


Fig. 2. Time series plots of observed particle-bound PAH concentrations (ng m^{-3}) as measured by the PAS2000 at the US EPA Supersite, 10/31/00–9/30/08.

Table 1Parameters and estimates for the outdoor residential PAH456 concentrations, units in $\log \text{ng m}^{-3}$.

Variable	Estimate (β)	Standard error	Pr > t	Variable range
Intercept	1.29	0.26	<0.0001	–
24-h particle-bound PAH @ Fresno Supersite (ng m^{-3})	0.048	0.006	<0.0001	1.1–50.0
Winter season	0.92	0.15	<0.0001	Binary (0/1)
9 am wind direction from between NW and N	0.56	0.11	<0.0001	Binary (0/1)
24-h relative humidity (%) @ Clovis	–0.017	0.003	<0.0001	28.9–97.0
Closest road to residence is smallest road type	–0.36	0.09	0.0001	Binary (0/1)
Fraction of homes in blockgroup that use gas heating fuel	0.90	0.31	0.0036	0.13–0.84
24-hour average wind recirculation factor	–0.49	0.17	0.0036	0.01–0.93
Total length (meters) of highway within 500 m	0.000082	0.000047	0.08	0–6209

relationships showed that the outdoor residential PAH456 concentrations were not strongly correlated with any single independent variable. The most explanatory independent variable was the 24-h EPA Supersite PAH measurement with $R^2 = 0.19$. This reflects a high degree of variability in the dataset, possibly due to spatial variability.

Using the multi-step approach outlined in Section 2.6 a model was selected for PAH456 with eight fixed effects – five temporal, three spatial – and one random effect, residential location (Table 1). The final full mixed-effects model accounted for 81% of the between-location variance and 18% of the within-location variance (Table 2). The five temporal variables accounted for almost the full change in the between-location variance. However, the addition of the spatial factors affected a 16% overall reduction in the between-location variance. Table 3 shows predicted PAH456 for some combinations of the fixed effects in the model.

The overall fit of the model, from the leave-one-out cross-validation, was good, with the IOA = 0.67. The total RMSE was 2.25 ng m^{-3} , the RMSEs was 1.91 ng m^{-3} , and the RMSEu was 1.62 ng m^{-3} . The model fit did not vary significantly by season. Fig. 3 shows a plot of the observed and the predicted outdoor residential daily PAH456 concentrations. Residual analysis shows normal distributions, with no systemic bias or errors. Temporal autocorrelation was not found in the scaled residuals, nor was there spatial autocorrelation as assessed by the Moran's I statistic ($I = -0.01$, $p = 0.83$).

3.5.2. Individual exposure estimates

The PAH456 model was used to calculate 654,170 individual daily estimates of PAH concentrations outside homes, encompassing all FACES participant-days from 1/1/2001 through 9/30/2008. Table 4 shows the distribution of the estimates compared to the observed (a figure showing the time series of the daily individual estimates for all participants and all days is available in the Supplemental section, Fig. 1). The distribution of the estimates shows that in the years of the HI substudy the model estimates were similar to those of the observed. The average daily ratio of the maximum to minimum concentrations, a measure of the range of the spatial variability in PAH456 in Fresno, compared well between the estimates (ratio = 3.30) and the observed values (ratio = 3.65). Across the Fresno metropolitan area, the average daily ratio of maximum to minimum concentrations is 5.2. Fig. 4 shows the spatial distribution

of PAH456 for a single day, 2/18/03. This day was selected because it is part of the higher winter season (the daily mean is in the highest 5% of days estimated), showing the distribution when it is likely to be both clinically significant and noticeably variable. The map illustrates the spatial variability of the PAH456 estimates across Fresno during the winter – higher values occur in the southern central portion of Fresno, particularly in the downtown region where the freeways intersect. There is also a band of high values in the central northern portion of Fresno, which matches the location of a major arterial, Herndon Avenue, which has high truck traffic across the city. In contrast, Fig. 5 shows the spatial distribution of PAH456 for a single day, 8/14/07, during the non-winter months. This map shows that the majority of the urban area has an estimated PAH456 concentration that is lower than the lowest concentration on the high winter day. The areas of higher relative PAH456 concentrations remain, but the absolute concentrations are lower. Both maps show the locations of the FACES residences, illustrating why the maximum to minimum ratio for the residences is smaller than that of the full city. The density of FACES residences, and Fresno residential neighborhoods generally, is not evenly distributed overall levels of outdoor PAH456 concentrations. The location of residences is biased toward lower concentrations.

4. Discussion

The goal of this research was to specify and implement a model for daily, outdoor, residential PAH concentrations for each FACES participant for use in further epidemiological investigations of acute and longitudinal effects of air pollution on asthmatic children (Mann et al., 2010; Margolis et al., 2009; Nadeau et al., 2010). This goal was achieved with a mixed-effects model based on measurement data combined with longitudinal data collected over more than seven years. The single most predictive covariate in the models presented, assessed by calculating the maximum possible contribution of each covariate given the coefficient and the range of the covariate, was the 24-h particle-bound PAH concentration at the US EPA Supersite. A second highly influential variable in our data was the season during which the sample was collected. The winter season (November through February) is significantly higher in particle-bound PAHs as measured at the EPA Supersite, and the model confirmed that the same seasonality can be observed in the residential data when the spatial distribution is taken into account.

Table 2

Variance component estimates for one-way random effects model and two mixed-effects models. (Number of samples = 497, number of homes = 83)

Variance	Random effects model, est.(CI)	Mixed effect model, temporal fixed effects, est. (CI)	Full mixed effect model, temporal and spatial fixed effects, est.(CI)
Between house variance	0.27 (0.18–0.46)	0.09 (0.05–0.23)	0.05 (0.02–0.21)
Within house variance	0.71 (0.62–0.82)	0.58 (0.51–0.68)	0.58 (0.51–0.67)
% of between house variance explained:	–	65.1%	81.2%
% of within house variance explained:	–	17.7%	18.3%

Table 3
Predicted outdoor residential PAH456 concentrations, using parameter estimates from the full mixed-effects model and values derived from the full dataset of possible days (1/1/01–9/30/08).

	Median values	Winter	High PAH in winter	Living on smallest road	Living near highways	Worst Case	Best Case
24-h particle-bound PAH @ Fresno Supersite (ng m^{-3})	3.1	3.1	21.7	3.1	3.1	54.6	0.9
Winter season	No	Yes	Yes	No	No	Yes	No
9 am wind direction from between NW and N	No	No	No	No	No	Yes	No
Fraction of homes in blockgroup that use gas heating fuel	0.61	0.61	0.61	0.61	0.61	0.90	0.03
24-h average wind recirculation factor	0.26	0.26	0.26	0.26	0.26	0.00	1.00
Closest road to residence is smallest type	No	No	No	Yes	No	No	Yes
Length (meters) of highway within 500 m	0	0	0	0	1819	6208.64	0
24-h relative humidity (%) @ Clovis	51.8	51.8	51.8	51.8	51.8	16.6	99.8
Estimated PAH456 (ng m^{-3})	2.72	6.80	16.49	1.90	3.16	612.20	0.32

This seasonality, lower concentrations during the summer months and higher in the winter months, is typical of $\text{PM}_{2.5}$, CO, elemental carbon, black carbon, NO_2 , and NO_x in Fresno (Blanchard et al., 1999; Schauer and Cass, 2000). A main cause of higher concentrations in the winter months is the altered meteorological conditions, including lower mixing height. As mixing height data were not available during the study, using season as an indicator was useful. A second significant contributor to increased PAH concentrations during the winter in Fresno was increased biomass burning due to woodstove and fireplace use (Launhardt et al., 1998). Both wind direction and wind pattern were important in the model. On days when the wind was blowing from between the NW and N directions, residential outdoor PAH456 was higher. This is likely because Hwy 99 is to the west of the majority of Fresno, meaning that this wind pattern would blow traffic emissions from Hwy 99 across the city. The three spatial variables (i.e. percent of gas heating fuel use, residential road type, and length of freeway within 500 m of residence) each made moderate contributions to the model overall, but significant contributions to the change in between-home variability (16%) once temporal trends were accounted for. The percent of houses using gas as the primary heating fuel is a neighborhood-level variable, whereas the residential road type and the length of freeway within 500 m are individual-level variables. This indicates that the PAH spatial distribution is variable on both a neighborhood- and individual-level when examined as a daily concentration. The temporal variables (i.e. PAH measurement at the EPA

Supersite, wind characteristics, and seasonality) made a larger reduction in the between-home variance (65%), but temporal variability in the data was larger than spatial variability. In addition to explaining a large amount of the variability in the dataset, the model also had good model fit, with IOA sufficiently high at 0.67. The RMSE, total and component parts, were reasonably low given the distribution of the PAH456.

As we reported in Section 3.2, there is a statistically significant decrease in the particle-bound PAH as measured at the EPA Supersite over the 8 years of the study. This may be due to diesel emissions regulations adopted and made effective by the California Air Resources Board (CARB) (Union of Concerned Scientists, 2009). Specifically, effective August 2003, CARB established a new Air Toxics Control Measure limiting school bus idling to 5 min, or if within 100 feet of a school, the bus must be immediately turned off and then restarted less than 30 s before departure. The same regulation applies to transit or delivery vehicles within 100 feet of a school. Similar regulation limiting heavy-duty truck idling was put into effect in January 2005. Additionally, changes to reduce emissions from motor vehicle diesel fuel were put into effect in 2003 and January 2005.

The model presented above generated the daily spatial distribution of outdoor residential PAH456 concentrations in Fresno, as well as individual exposure estimates for each FACES participant. Our data are representative of outdoor residential exposure, which may be less extreme in concentrations than if samples had been

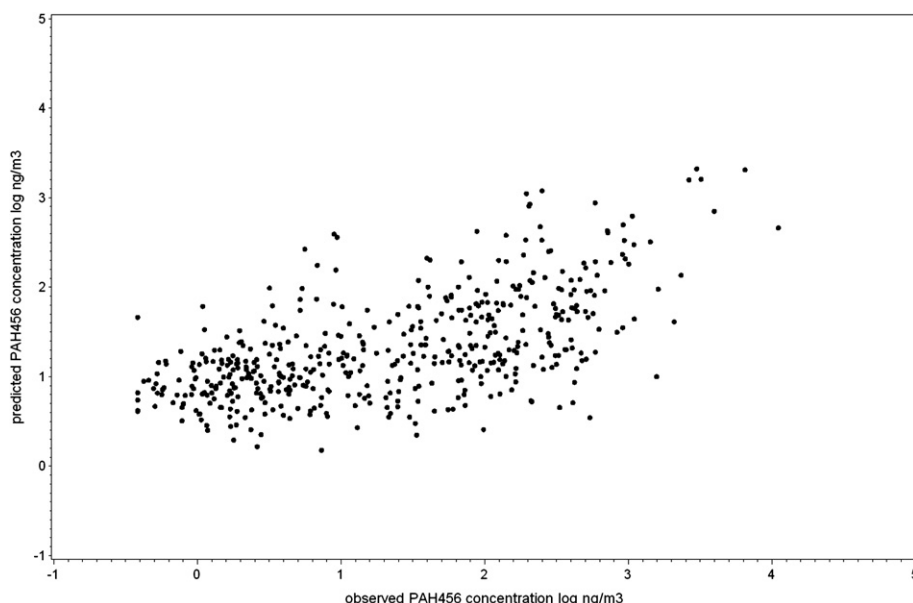


Fig. 3. Plot of the outdoor residential PAH456 concentrations (ng m^{-3}) observed versus the predicted from the model.

Table 4Summary statistics for observed and model-derived estimated daily outdoor residential PAH456 concentrations. All units are in ng m^{-3} .

Variable	Number values	Number homes	Mean (SD)	25th %	Median	75th %	Range
All observed in HI substudy	497	83	6.0 (6.4)	1.6	3.8	8.4	0.7–57.1
Observed in HI substudy in winter only	171	39	9.0 (8.5)	3.1	6.3	12.4	0.7–57.1
All model-derived estimated during HI sub-study timeframe (2/2002–2/2003)	78,119	253	4.4 (3.4)	2.3	3.3	5.2	0.8–52.5
Model-derived estimated during HI sub-study timeframe in winter only	30,941	252	7.0 (4.0)	4.1	5.9	9.0	1.3–37.9

collected directly adjacent to roadways or other sources (Baxter et al., 2007). Previous work describing the spatial distribution of PAHs has been limited primarily to source-to-receptor studies (Li et al., 2003; Vardoulakis et al., 2005), small neighborhood-sized areas (Levy et al., 2001) or to large-scale regional or national distributions (Guo et al., 2003; Lang et al., 2007; Manoli et al., 2004; Prevedouros et al., 2004; Thornhill et al., 2008). Levy et al. (2001) conducted mobile monitoring for PAHs in Roxbury, MA, to determine the impact of a bus terminal on ambient concentrations of PAHs. They were able to observe increased concentrations of PAHs close to the bus terminal and determine, through mixed modeling, that the bus terminal was the major contributor of ambient PAHs. While Levy et al., had a much smaller zone of interest (approximately 2 square miles), a different modeled outcome (1-min average PAH), and limited their sample to a single season, their model findings are consistent with our models, i.e. that traffic is a major contributor to ambient PAH concentrations. Large-scale PAH spatial distribution models presented in the literature use either emissions inventories (Lang et al., 2007; Prevedouros et al., 2004) or a low number of air monitors (Guo et al., 2003; Manoli et al., 2004; Thornhill et al., 2008) in order to describe the distributions of PAHs. In both cases, the uncertainty regarding the spatial distribution is high compared to our models. Urban or national scale descriptions of PAH distributions tend to represent annual or seasonal averages. While these can be useful for policy recommendations or long-term exposure studies, they cannot address the impact of short-term exposure on acute health outcomes.

LUR models have become a popular method for integrating air pollutant measurement data and environmental characteristics. However, there have been no published studies to date of LUR models for ambient PAH concentrations on an annual, seasonal or daily basis. Additionally, there has been no presentation of LUR models of similar pollutants within California's San Joaquin Valley, the region in which Fresno is located and one of those most polluted regions in the United States (EPA, 2001). However, as with other LUR models for traffic pollutants (Hoek et al., 2008), we found that both proximity to freeways (i.e. high volume of traffic) and the size of the road on which the sample was collected were significant factors in estimating the ambient concentration. However, PAHs are significant because they are not merely another marker for traffic exposure, but are implicated in negative health impacts as discussed in Section 1. As such, it is important to evaluate and model PAHs specifically as the spatial distributions of all traffic pollutants are not identical.

Future individual exposure estimates of PAH456 for the FACES cohort will incorporate participants' time spent at school and indoors. Indoor PAH exposures are important because of the high percentage of time that children spend indoors. Indoor PAH concentrations cannot be predicted exclusively through infiltration from outdoors because there are potentially significant indoor PAH sources in residences (Naumova et al., 2002). In order to refine the individual estimates for indoor and outdoor exposures, a model of indoor PAH exposures is needed; one could be developed based on the paired indoor/outdoor HI sub-study filter data (only 82 of the 497

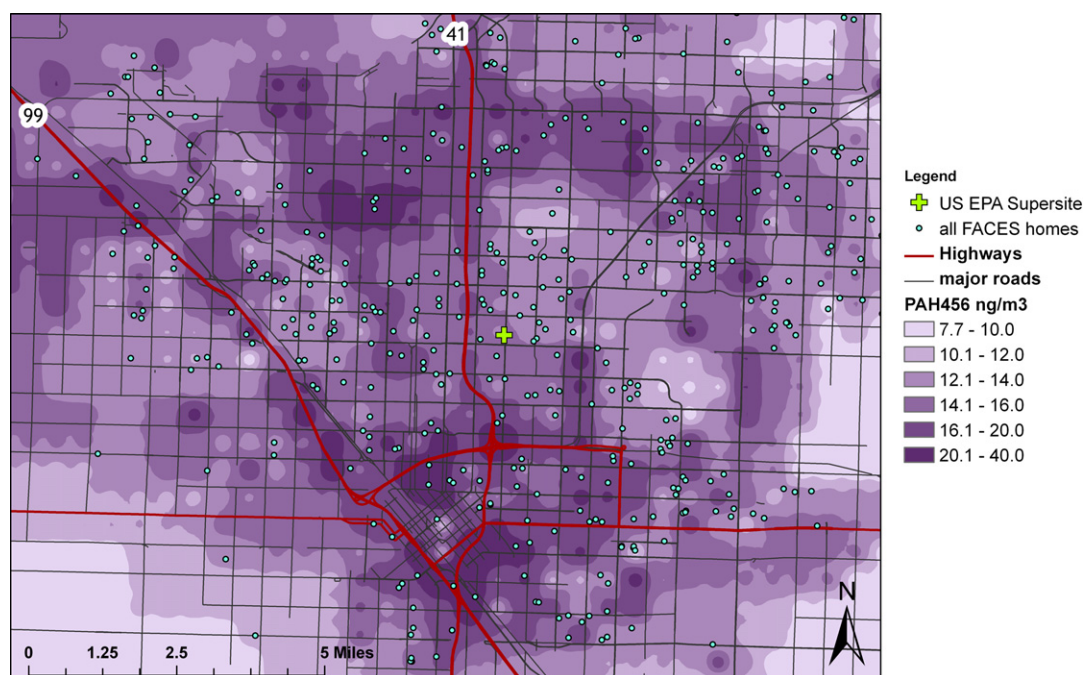


Fig. 4. Spatial distribution for PAH456 concentrations (ng m^{-3}) on 2/18/03, with FACES residential location marked for the 226 participants enrolled on that date. Surface is interpolated from a 750-foot grid across the city, at which points the LUR model was used to calculate concentrations.

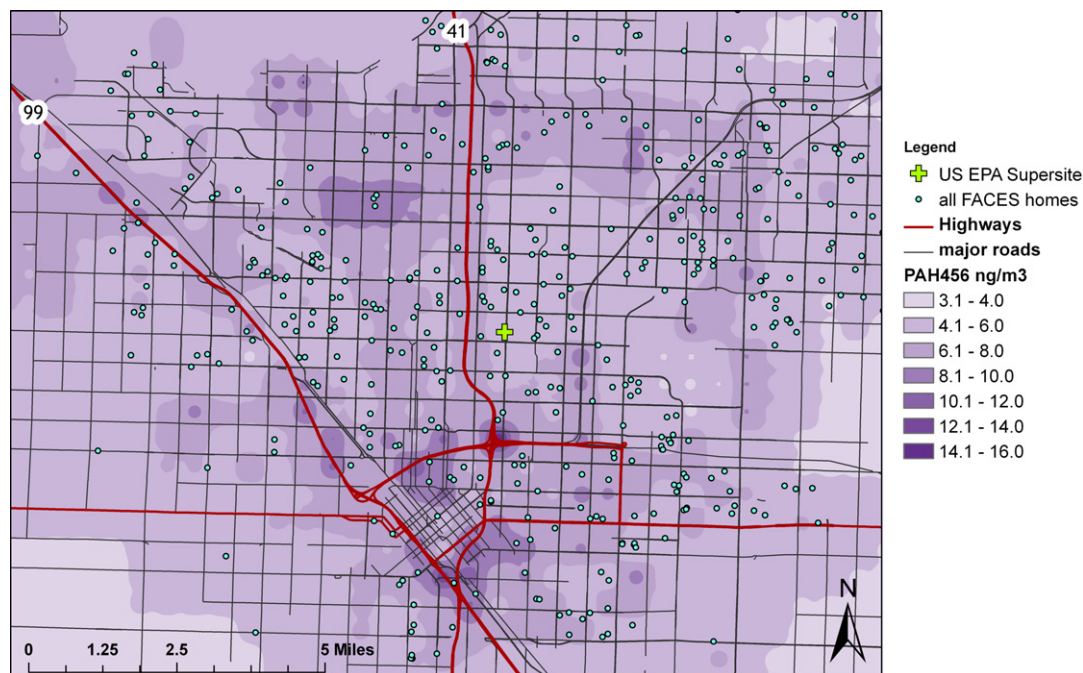


Fig. 5. Spatial distribution for PAH456 concentrations (ng m^{-3}) on 8/14/07, with FACES residential location marked for the 302 participants enrolled on that date. Surface is interpolated from a 750-foot grid across the city, at which points the LUR model was used to calculate concentrations.

matched indoor samples have been analyzed). Using time-location data for each child, a new individual exposure estimate accounting for time in school and indoors at home could be then calculated.

5. Conclusion

In conclusion, we have estimated individual daily outdoor PAH exposure for the 315 participants in the FACES study for over seven years using LUR modeling with mixed-effects regression. We found that traffic characteristics, home heating, season, and meteorology each play an important role in characterizing PAH exposure in Fresno, CA. While temporal variables accounted for more of the total variability within the model, the estimates were significantly improved by the addition of spatial variables. Unlike toxic air pollutants with regional-level variability, PAHs vary widely within a single city or urban area and neighborhood-level effects are important. The location of the child's home within the urban setting plays an important role in the level of exposure to PAHs that each child experiences.

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Appendix. Supplementary material

Supplementary material related to this article can be found online at [doi:10.1016/j.atmosenv.2011.02.014](https://doi.org/10.1016/j.atmosenv.2011.02.014).

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