# An LUR/BME Framework to Estimate PM$_{2.5}$ Explained by on Road Mobile and Stationary Sources

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Ⓢ *Supporting Information*
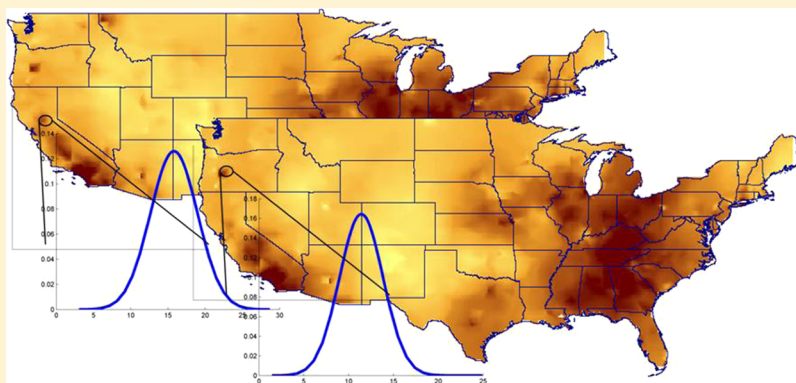
ABSTRACT: Knowledge of particulate matter concentrations <2.5 μm in diameter (PM$_{2.5}$) across the United States is limited due to sparse monitoring across space and time. Epidemiological studies need accurate exposure estimates in order to properly investigate potential morbidity and mortality. Previous works have used geostatistics and land use regression (LUR) separately to quantify exposure. This work combines both methods by incorporating a large area variability LUR model that accounts for on road mobile emissions and stationary source emissions along with data that take into account incompleteness of PM$_{2.5}$ monitors into the modern geostatistical Bayesian Maximum Entropy (BME) framework to estimate PM$_{2.5}$ across the United States from 1999 to 2009. A cross-validation was done to determine the improvement of the estimate due to the LUR incorporation into BME. These results were applied to known diseases to determine predicted mortality coming from total PM$_{2.5}$ as well as PM$_{2.5}$ explained by major contributing sources. This method showed a mean squared error reduction of over 21.89% oversimple kriging. PM$_{2.5}$ explained by on road mobile emissions and stationary emissions contributed to nearly 568 090 and 306 316 deaths, respectively, across the United States from 1999 to 2007.

■ INTRODUCTION

Chronic exposure to ambient PM$_{2.5}$ is linked to increased morbidity and mortality in many epidemiological studies[1,2] and results in high population burden,[3,4] making it a large public health concern. Hence quantifying accurate air pollution exposure has become paramount and has prompted different approaches to estimate chronic PM$_{2.5}$ levels across space and time.

As our awareness of the impact of air pollution has increased, so has the interdisciplinary nature of exposure assessment. Researchers from these disciplines range from air pollution scientists to epidemiologists to risk assessors who are all involved in better understanding air pollution processes and its health effects. Disciplines also extend to cost-benefit analysts, policy makers and regulators whose goals are air pollution abatement through policy to efficiently diminish its burden on the population. Because of the wide range of groups involved there is a critical need for methods that are accurate in estimating chronic levels of PM$_{2.5}$ and are both accessible and interpretable by a wide audience. It is this wide audience which

we are keeping in mind in advancing methods used to estimate chronic PM$_{2.5}$ levels.

Existing methods used to estimate PM$_{2.5}$ levels fall in several classes that include (1) chemical transport models (CTM), (2) land use regression (LUR), (3) satellite data, and (4) different geostatistical approaches. LUR is a regression model which estimates air pollution as a function of explanatory variables. LUR takes characteristics from the study area (traffic count, road length, distance to nearest road, elevation, land cover, household density, wind, etc.) and develops a multiple linear regression model which aims at describing a pollutant of interest.[5−7] Most LUR models are geared toward a model that explains the most variability of the dependent variable (i.e., the model with the highest possible r^2) on a relatively small scale.[8] LUR has been widely used for exposure estimation.[9] Each of

these methods has its distinct characteristics and corresponding utility. They range from process-based prediction methods to data-driven statistical estimation methods. The first two classes of methods are defined by their ability to predict levels based on a model representation of the processes that lead to air pollution. This is useful in estimating contributions from various emission sources. The latter two classes are driven by observations, such as satellite readings or measurement from ground monitoring stations. These are useful for obtaining estimates grounded to physical measurements of $PM_{2.5}$. Although no categorization is without exception or entirely distinctive, these classes demonstrate possible methodological procedures. These four classes also differ widely in terms of accuracy, complexity, numerical cost and accessibility (see Supporting Information (SI)).

Geostatistical approaches provide, like satellite data, observationally driven estimates of $PM_{2.5}$. They usually consist of linear kriging estimators of $PM_{2.5}$ calculated from measurements at ground monitoring stations. These methods provide accurate estimates in the vicinity of monitoring stations and are simple to use, thereby providing a widely utilized approach. However, like any observationally driven estimation method, geostatistical methods alone cannot be used to explain contributions from major contributing sources.

While work has been done to develop methods individually within the four classes mentioned above, there is also interest in combining approaches across classes to create an estimation framework that combines the strengths of the respective groups. The goal of this work is to combine a process based method and an observationally based estimation method to create a combined estimation method that can be used by a wide audience to accurately estimate the distribution the annual $PM_{2.5}$ concentration across the continental United States (U.S.) from 1999 to 2009, and to quantify how much of the estimated annual $PM_{2.5}$ concentration can be explained by the major contributing sources of on road mobile emissions and stationary emissions.

We will achieve our goal by using the Bayesian Maximum Entropy (BME) knowledge synthesis framework[10,11] to combine LUR with geostatistical estimation. BME utilizes Bayesian epistemic knowledge blending to combine data from multiple sources. For our process-based method we select LUR over CTMs because of its ability to use readily available information about on road mobile emissions and stationary emissions to predict annual $PM_{2.5}$. For our observationally based method we rely on a geostatistical analysis of ground observations of $PM_{2.5}$ concentrations because of the relatively large number of monitoring stations providing accurate measurements across the U.S. By combining methods like LUR and BME we can create a model that is numerically efficient, applicable and interpretable over a large domain size.

The knowledge base considered in the BME method consists of general knowledge describing generalizable characteristics of the space/time $PM_{2.5}$ field (such as its space/time trends and dependencies, its relationship with respect to various emissions, etc), and site specific knowledge that include hard data (data without measurement error) and soft data (data with measurement errors which can be non-Gaussian). The strategy we will use in this work is to employ LUR to describe the general trends of annual $PM_{2.5}$ concentrations over the entire U.S. and model the $PM_{2.5}$ residuals (obtained by removing the LUR offset) using BME. This will allow us to rigorously account for the non-Gaussian uncertainty associated with

annual $PM_{2.5}$ concentration calculated from daily concentrations where some daily concentrations may be missing.[12]

One outcome of our work is the development of an LUR for the prediction of annual $PM_{2.5}$ concentrations across the continental U.S., which is a geographical domain of a fairly large size. While many previous studies have developed LUR models over small geographical domains where high predictability can be achieved,[13] each specific LUR model is usually only valid for the small region for which it was developed.[6] In other words high predictability is achieved by sacrificing generalizability[14,15] (see SI). There have been comparatively fewer studies that developed an LUR with lower predictability but higher generalizability. The LUR we present fills that knowledge gap, with a specific focus on using annual $PM_{2.5}$ explained by on road mobile emissions and stationary emissions as its predictors.

Another outcome of our work is the sequential integration of two classes of methods (LUR and geostatistical) to create a combined LUR/BME estimation method that borrows strengths from each of its constituent. Combining methods is a growing research area and our work contributes to that field. While very few works have looked at combining LUR and BME approaches[16,17] or LUR and kriging approaches,[18] more studies are needed in order to explore the various ways by which to combine these methods. We focus specifically on using LUR to provide general knowledge about $PM_{2.5}$, using BME to account for the incompleteness of daily samples, and making the combined method accessible to a wide audience. Other strategies and focus will undoubtedly have to be investigated in future works, for example creating more elaborate LUR models[17] including those which use meteorological data.[19]

Finally we use our LUR/BME model to perform a risk assessment that differentiates the number of annual $PM_{2.5}$ predicted deaths that can be explained by on road mobile emissions and stationary emissions. The dichotomous assignment of $PM_{2.5}$ to these two sources allows for straightforward abatement strategies. This assessment is useful on its own to generate research questions that can improve methods used to calculate death reductions achieved under various scenarios of source reductions.

■ MATERIALS AND METHODS

$PM_{2.5}$ Data. Raw daily federally referenced method (FRM) $PM_{2.5}$ monitoring data collected from 1999 to 2009 were obtained from the EPA's Air Quality Systems (AQS) database across the contiguous United States.[20] Whenever a daily $PM_{2.5}$ monitoring value reported below the detection limit of its monitor, it was replaced by the mean of a log-normal distribution that was fit to all reported below-detect values. Daily values were averaged whenever two or more daily $PM_{2.5}$ monitoring values were reported by collocated monitors on a given day/site.

Annual $PM_{2.5}$ were calculated from daily $PM_{2.5}$ monitoring values as follows: every day for which a station reported a daily $PM_{2.5}$ monitoring value, a corresponding annual $PM_{2.5}$ was calculated by taking the arithmetic average of all the daily monitoring values reported at that station over the previous year (i.e., 365 days) including that day. Note that this one year period could include time before January 1, 1999 (i.e., the first day for which daily monitoring data were available).

The intended sampling frequency of a given daily monitoring station was used to calculate how many daily monitoring values should have been reported in a given year period. Comparing

this number to the actual number of reported monitoring values informs us about the incompleteness of intended sampling over that given year. We use this to assess the uncertainty associated with the corresponding annual $PM_{2.5}$.

LUR Data. The LUR model predicts annual $PM_{2.5}$ given a series of predictive LUR independent variables that characterize the effect of (a) elevation, (b) on road mobile emissions, and (c) stationary emissions. A detailed explanation of all data sources for each LUR independent variable is described in the SI.

We focus on on road mobile emissions and stationary emissions because they are two major contributors to anthropogenic pollution. For stationary emissions, we used data from the EPA's National Emissions Inventory[21] (NEI), which provides inventories of stationary emissions (in tons/ year) of the main constituents of $PM_{2.5}$ (i.e., SO_2, NH_3, $PM_{2.5}$-primary and NO_X). These inventories are reported in a manner that is consistent across the U.S. We assume that at space/time location $p = (s, t)$, the effects of stationary source emissions decrease exponentially with distance between the source and the location p, as given by the equation $V_{i,p} = \sum_{n=1}^{N} em_i(e_n,t) \cdot p((-3\|e_n - p\|)/(d_{r_i}))$, where, $i = SO_2$, NH_3, $PM_{2.5}$, NO_X, $em_i(e_n,t)$ is the emissions in tons/year of constituent i at stationary source emissions location e_n and time t and d_{r_i} is the exponential decay range in km. It would be difficult to consistently and accurately measure on road mobile emissions across the entire U.S. Thus for on road mobile emissions we use data estimating vehicular traffic (annual average daily traffic counts for each major highway road segment in the U.S. as estimated through linear referencing[22]) and population density (people/km²) to construct variables that estimate total traffic (TT), average congestion (AC), and emission efficiency (EE) based on population density. Emission efficiency is added to correct for the assumption that every mile driven produces the same amount of emissions regardless of vehicle type by hypothesizing that areas with high population density tend to have vehicles better suited for urbanized environments, which (in general) are more fuel efficient. These traffic and emission efficiency variables are then combined to provide an estimate of on road mobile emission, thereby bypassing the laborious task of obtaining on road mobile emission data directly for a nationally sized domain.

Large Area Variability LUR Model. Our large area variability LUR expresses the annual $PM_{2.5}$ at space/time location $p = (s,t)$, where $s = (s_1,s_2)$ is the spatial coordinate and t is time, as a linear combination of the corresponding LUR independent variables at p. The first independent variable consists of the elevation $V_{Elev,p}$ at p. The next three independent variables characterize the effect of on road mobile emissions. They are denoted as the column vector $V_{mobile,p} = [V_{TT,p} \ V_{AC,p} \ V_{EE,p}]^T$, where the subscript T denotes the transpose, and $V_{TT,p}$, $V_{AC,p}$ and $V_{EE,p}$ are variables characterizing total traffic, average congestion, and emission efficiency, respectively, at p. The last four independent variables characterize the effect of stationary emissions. They are denoted as $V_{stationary,p} = [V_{SO2,p} \ V_{NH3,p} \ V_{PM2.5,p} \ V_{NOX,p}]$, where $V_{SO2,p}$, $V_{NH3,p}$, $V_{PM2.5,p}$ and $V_{NOX,p}$ are variables charactering the concentrations of SO_2, NH_3, $PM_{2.5}$, and NO_X, respectively at space/time location p.

We consider models that include the elevation variable, at least 1 out of the 3 on road mobile emission variables, and at least 1 out of the 4 stationary emission variables, which results

in a total of $1 \times \sum_{i=1}^{3} \binom{3}{i} \times \sum_{i=1}^{4} \binom{4}{i} = 1 \times 7 \times 15 = 105$ candidate models. These models are expressed by the following equation

$$Z_p = \beta_0 + \beta_{Elev} V_{Elev,p} + (I_{mobile} \cdot \times \beta_{mobile}) V_{mobile,p}$$
$$+ (I_{stationary} \cdot \times \beta_{point}) V_{point,p} + \varepsilon_p \quad (1)$$

where Z_p is annual $PM_{2.5}$ at p, β_0 is the equation intercept, β_{Elev}, $\beta_{mobile} = [\beta_{TT} \ \beta_{AC} \ \beta_{EE}]$ and $\beta_{stationary} = [\beta_{SO2} \ \beta_{NH3} \ \beta_{PM2.5} \ \beta_{NOX}]$ are linear coefficients for the independent variables $V_{Elev,p}$, $V_{mobile,p}$ and $V_{stationary,p}$, respectively, $I_{mobile} = [I_{TT} \ I_{AC} \ I_{EE}]$ and $I_{stationary} = [I_{SO2} \ I_{NH3} \ I_{PM2.5} \ I_{NOX}]$ are vectors of indicator values (0 or 1) such that at least one element in both I_{mobile} and I_{point} must be 1, the "$\cdot\times$" operator denotes the element-by-element multiplication between same-sized vectors and ε_p is a homoscedastic error term.

Due to the large overlap in annual $PM_{2.5}$, only a subset of annual $PM_{2.5}$ was used to construct the LUR model to avoid collinearity. Namely, only the last annual $PM_{2.5}$ in a calendar year was used from each station (approximately 11 000 data values), encompassing all daily values.

Each of the 105 candidate LUR models were optimized by selecting hyperparameter values that maximized the LUR r^2. A hyperparameter is a physical parameter within each variable that is allowed to adjust based on predictability of annual $PM_{2.5}$. Hyperparameters for annual $PM_{2.5}$ include the radii a_{r_1}, a_{r_2} and a_{r_3} for the buffers used to calculate total traffic, average congestion, and emission efficiency, respectively, and the exponential decay ranges for stationary source variables (i.e., d_{ri} described in the SI). The *fminsearch* function of MATLAB was used to search for hyperparameter values that maximized the LUR r^2. The search was started given an initial selection of hyperparameters described in SI.

The Akaike Information Criteria (AIC) and all variance inflation factor (VIF) values were found for each of the 105 optimized candidate LUR models. AIC is a measure of parsimony of a model and VIF is a measure of collinearity of a model. Out of the 105 optimized models, our final model has the lowest AIC value among models with VIF values <10 and with physically plausible βs. The βs have to be positive in order to be plausible, with the exception of negative βs for emission efficiency and elevation.

BME Methodology. BME is a mathematically rigorous geostatistical space/time framework developed by Christakos.[10,23] BME can incorporate information from many different sources and BME is implemented using the *BMElib* suite of functions in MATLAB.[11] The buttress of BME has been detailed in other works,[11,23,24] and can be summarized as performing the following steps: (1) gathering the general knowledge base (G-KB) and site-specific knowledge base (S-KB) about the mapping situation, (2) using the *Maximum Entropy* principle of information theory to process the G-KB in the form of a prior probability distribution function (PDF) f_G, (3) integrating S-KB using an epistemic *Bayesian* conditionalization rule on data f_S with and without measurement error to create a posterior PDF f_K, and (4) creating space/time estimates based on the analysis. We use a space/time random field (S/TRF) to describe the variability of annual $PM_{2.5}$ across the U.S. Our notation a for S/TRF will consist of denoting a single random variable Z in capital letters, its realization, z, in lower case; and vectors and matrices in bold faces (e.g., $Z =$

$[Z_1,...,Z_n]^T$ and $z = [z_1,...,z_n]^T)$. Let $Z(\boldsymbol{p}) = Z(\boldsymbol{s},t)$ be a space/time random field (S/TRF) representing annual $PM_{2.5}$.

We define the transformation of the $PM_{2.5}$ data $\boldsymbol{z}_h$ observed at locations $\boldsymbol{p}_h$ as

$$\mathbf{x}_h = \mathbf{z}_h - o_Z(p_h) \tag{2}$$

where $o_Z(\boldsymbol{p})$ may be any deterministic offset that can be calculated without error as a function of the space/time coordinate $\boldsymbol{p}$. We then define $X(\boldsymbol{p})$ as the S/TRF representing the variability and uncertainty associated with the transformed data $\boldsymbol{x}_h$, and we let $Z(\boldsymbol{p}) = X(\boldsymbol{p}) + o_Z(\boldsymbol{p})$ be the S/TRF representing $PM_{2.5}$.

In this work, we consider two choices for $o_Z(\boldsymbol{p})$: (1) a constant value and (2) the LUR estimate $\hat{z}_{\text{LUR},P}$ given by

$$\hat{z}_{\text{LUR},p} = \hat{\beta}_0 + \hat{\beta}_{\text{Elev}} V_{\text{Elev},p} + (\hat{\mathbf{I}}_{\mathbf{mobile}}.\, {}^*\hat{\boldsymbol{\beta}}_{\text{mobile}}) \mathbf{V}_{\text{mobile},p}$$
$$+ (\hat{\mathbf{I}}_{\mathbf{stationary}}.\, {}^*\hat{\boldsymbol{\beta}}_{\text{stationary}}) \mathbf{V}_{\text{stationary},p} \tag{3}$$

where the estimated $\hat{I}$s indicators and $\hat{\beta}$s coefficients are those derived in our final annual $PM_{2.5}$ LUR model. We can then calculate $\hat{z}_k$, the estimated annual $PM_{2.5}$ at unmonitored location $\boldsymbol{p}_k$ by obtaining the BME estimate $\hat{x}_k$ for the transformed S/TRF $X(\boldsymbol{p})$ at the estimation point $\boldsymbol{p}_k$, and adding back $o_z(\boldsymbol{p}_k)$, the offset calculated at $\boldsymbol{p}_k$.

The G-KB for the transformed S/TRF $X(\boldsymbol{p})$ consists of its expected value $m_x(\boldsymbol{p})$ and covariance function $c_X(\boldsymbol{p},\boldsymbol{p}')$ (see SI).

The S-KB for $X(\boldsymbol{p})$ consists of hard and soft data. The hard data $\boldsymbol{x}_h = \boldsymbol{z}_h - o_Z(\boldsymbol{p}_h)$ are obtained based on annual $PM_{2.5}$ values $\boldsymbol{z}_h$ calculated at hard data points $\boldsymbol{p}_h$ where at least 75% of intended samples were collected, in line with EPA regulations pertaining to valid design values.[25] Data points not meeting this completeness criterion are classified as the soft data points $\boldsymbol{p}_s$, with an uncertainty attributed to the incompleteness of intended sampling. Following Akita et al,[12] the uncertainty associated with the annual $PM_{2.5}$, z_s for station i and date t is described by a Gaussian PDF truncated below zero, with mean $\mu_{i,t}$ and standard deviation $\sigma_{i,t}$. The mean $\mu_{i,t}$ is simply the sample mean of the $n_{i,t}$ daily concentrations ($z_{i,j}$, $j = 1,...,n_{i,t}$) recorded at station i over 1 year preceding date t. The epistemic uncertainty associated with the incompleteness of intended sampling is characterized by the difference between $n_{i,t}$ and the intended number of samples $n_i^* \geq n_{i,t}$ that would have been collected if the station worked as intended in accordance with the monitor's sampling frequency. Therefore a reasonable choice for the standard deviation quantifying that uncertainty is

$$\sigma_{i,t} = \sqrt{\frac{\sum_{j=1}^{n_{i,t}} (z_{i,j} - \mu_{i,t})^2 / (n_{i,t} - 1)}{n_{i,t}}} \times \sqrt{\frac{n_i^* - n_{i,t}}{n_i^*}} \tag{4}$$

where the first factor is the standard deviation of the sample mean and the second factor is a population correction factor that accounts for the incompleteness of intended sampling from a population of size n_i^*. The PDF for x_s is then derived from the PDF for z_s by simply using the transformation $x_s = z_s - o_z(\boldsymbol{p}_s)$.

The G-KB and S-KB for the S/TRF $X(\boldsymbol{p})$ can overall be written as $G = \{m_X(\boldsymbol{p}), c_X(\boldsymbol{p},\boldsymbol{p}')\}$ and $S = \{x_h, f_S(x_S)\}$, and in this case the BME posterior PDF for $X(\boldsymbol{p}_k)$ at estimation point $\boldsymbol{p}_k$ is given by $f_K(x_k) = A^{-1} \int d\mathbf{x}\, f_S(\mathbf{x}_S) f_G(\mathbf{x})$ where $\mathbf{x} = (x_k, \mathbf{x}_h, \mathbf{x}_s)$ is a realization of X at points $\boldsymbol{p} = (\boldsymbol{p}_k, \boldsymbol{p}_h, \boldsymbol{p}_s)$ and A is a normalization constant.[10,25] Finally the PDF for z_k is obtained by simply using the back-transformation $z_k = x_k + o_z(\boldsymbol{p}_k)$.

Comparison of Methods Using Cross-Validation Analysis. In order to test the estimation improvement of LUR and BME, a cross-validation was performed to compare three different methods used in this study: (a-*constant/hard*) setting the deterministic global (i.e., covering a substantial domain where variability within the domain can be largely diverse) offset $o_Z(\boldsymbol{p})$ to a constant value and considering all data as hard, (b-*LUR/hard*) setting the global offset to the LUR model and considering all data as hard and (c-*LUR/hard and soft*) setting the global offset to the LUR predicted value and considering data as hard and soft as defined in the previous section. For each of these methods, the cross validation procedure consists of randomly selecting 20 000 hard data points, removing each one at a time, and re-estimating it from the remaining annual $PM_{2.5}$. The cross-validation statistics investigated include mean squared estimation error (MSE), root mean squared estimation error (RMSE), mean absolute estimation error (MAE), mean of the root variance of the posterior PDFs (MR), the square of Pearson's correlation coefficient, and the square of Spearman's correlation coefficient. Equations for each measure are defined in the SI. Along with the leave-one-out cross validation (LOOCV) of 20,000 hard data point, a 10-fold spatial cross-validation was also performed.

Risk Assessment Application. The incorporation of the LUR model into the BME methodology has many potential applications including determining the mortality of various diseases attributable to $PM_{2.5}$. Excess mortality was calculated using the methodology presented by Li,[26] assuming linearity, in order to quantify total mortality, mortality from ischemic heart disease (IHD) and mortality from lung cancer (LC). Relative risks for these diseases were obtained from Krewski et al.[27] Deaths at the county level were obtained from the CDC.[28] Excess deaths were calculated for (1) annual $PM_{2.5}$, (2) annual $PM_{2.5}$ explained by on road mobile emissions, and (3) annual $PM_{2.5}$ explained by stationary emissions.

Let $\hat{z}_l(\boldsymbol{p})$ denote our estimate of annual concentrations, where l = total for total $PM_{2.5}$, l = mobile for $PM_{2.5}$ explained by on road mobile emissions, and l = stationary for $PM_{2.5}$ explained by stationary emissions. For l = total we simply use $\hat{z}_{\text{total}}(\boldsymbol{p}) = \hat{z}_{\text{LUR/BME},p}$, where $\hat{z}_{\text{LUR/BME}}(\boldsymbol{p})$ is the LUR/BME estimate of annual $PM_{2.5}$ described earlier. For l = mobile we use the LUR in a relative manner to estimate the ratio $\alpha_{\text{LUR mobile}}(\boldsymbol{p}) = ((\hat{I}_{\text{mobile}} \cdot\times \hat{\beta}_{\text{mobile}}) V_{\text{mobile},\boldsymbol{p}})/(\hat{z}_{\text{LUR},\boldsymbol{p}})$ corresponding to the proportion of $PM_{2.5}$ that the LUR model explains from on road mobile emissions. We then multiply that ratio with the LUR/BME estimate of annual $PM_{2.5}$, so that $\hat{z}_{\text{mobile}}(\boldsymbol{p}) = \hat{z}_{\text{LUR/BME},p} \alpha_{\text{LUR mobile}}(\boldsymbol{p})$. Likewise we use $\hat{z}_{\text{stationary}}(\boldsymbol{p}) = \hat{z}_{\text{LUR/BME},p} \alpha_{\text{LUR stationary}}(\boldsymbol{p}$, with $\alpha_{\text{LUR stationary}}(\boldsymbol{p}) = ((\hat{I}_{\text{stationary}} \cdot\times \hat{\beta}_{\text{stationary}}) V_{\text{stationary},\boldsymbol{p}})/(\hat{z}_{\text{LUR},\boldsymbol{p}}))$. The mortality for a specific cause of death (e.g., total mortality, IHD, LC) attributed to an annual concentration $\hat{z}_l(\boldsymbol{p})$ is given by Li et al.[26]

$$\Delta \boldsymbol{y}_1 = I_0 \times P \times (1 - e^{-\beta(\hat{z}_l(\mathbf{p}) - z_b)}) \tag{5}$$

where I_0 is the baseline incidence rate for the cause of death of interest, β is the corresponding concentration response coefficient, $\mathbf{P}$ is the population at the county level, and z_b is the background concentration. Sources have suggested a background level in the U.S. for $PM_{2.5}$ of 3−5 $\mu g/m^3$.[29] We use $z_b = 5\ \mu g/m^3$.

■ RESULTS

Annual PM$_{2.5}$. There were 1 478 149 annual PM$_{2.5}$ data points from 1999 to 2009 coming from 1576 monitoring stations. These include 406 962 (27.53%) soft data points. The mean of the annual PM$_{2.5}$ is 12.44 μg/m^3, the variance is 11.57(μg/m^3)^2, the skewness is 0.56 and the kurtosis is 5.57. The minimum annual value is 1.63 μg/m^3 and the maximum annual value is 75.40 μg/m^3.

Large Area Variability LUR Model. The final LUR model had six independent variables: elevation, three on road mobile emission variables (total traffic, average congestion, emission efficiency), and two stationary emission variables (NH$_3$ and SO$_2$) (Table 1). Table 1 describes the optimal hyperparameters

Table 1. Hyperparameters and Corresponding β for the Final LUR Model

	final LUR model	
variable	range (km)	$\hat{\beta}$ (μg/m^3 per variable unit)
intercept	NA	7.54×10^{00}
elevation[a]	0	-8.87×10^{-04}
total traffic[b]	694	3.04×10^{-03}
average congestion[c]	33	2.54×10^{-05}
emission efficieny[d]	730	-1.76×10^{-02}
SO$_2$[e]	210	1.10×10^{-04}
NH$_3$[e]	11.5	1.49×10^{-06}

[a]Meters. [b]km driven/km^2. [c]km driven/km. [d]People/km^2. [e]Thousand tons/year.

for each variable along with their corresponding $\hat{\beta}$ values. This LUR model has an $r^2 = 0.53$, providing generalizable predictability of annual PM$_{2.5}$ over the entire U.S. from 1999 to 2009.

LUR/BME Model. The combination of the LUR and BME methods through methods (a) to (c) led to a refined estimation of annual PM$_{2.5}$ as seen in Figure 1 showing estimated levels across the U.S. for May 1, 1999. Method (a-*constant/hard*) using a constant offset and using all data as hard does not differentiate well the annual PM$_{2.5}$ across southern California and estimates fairly benign levels for several states west of the Mississippi river. By incorporating the LUR offset, method (b-*LUR/hard*) provides estimates of annual PM$_{2.5}$ that are more refined and localized. By further incorporating the soft data to the hard data and LUR offset, method (c-*LUR/hard and soft*) further refines the description of hot spots across the country. Method (c) is able to pick up finer scale variation in concentrations compared to methods (a) and (b). This finer scale variation can also be seen in subsequent months (SI Figure S5).

Cross validation statistical measures indicated a consistent improvement in mapping accuracy from method (a) to (c) (Table 2). Measures of estimations errors (MSE, RMSE, MAE, MR) decreased from method (a) to (b) and from method (b) to (c), while measures of correlation (Square Pearson's Corr. Coeff. and Square Spearman Corr. Coeff.) increased from method (a) to (b) and from method (b) to (c). Incorporating the LUR offset while using only hard data (i.e., going from method (a) to (b)) resulted in a reduction of 21.89% in MSE. Further incorporating soft data (i.e., going from method (b) to (c)) resulted in an additional reduction of 4.87% in MSE. The reduction in MSE from method (b) to (c) is more pronounced when performing cross-validation on points that contain a

higher percentage of soft data (SI Table S3). This reduction is more pronounced still when estimation neighborhoods around cross-validation locations are forced to have soft data points (SI Table S4).

The r^2 correlation (Square Pearson's Corr. Coeff.) changes from 0.88 for the LOOCV to 0.78 for the 10-fold cross validation. This corresponds to 12.8% shrinkage in r^2, which is reasonable since the training set for the 10-fold cross validation is substantially smaller than that of the LOOCV.

Risk Assessment. Using eq 5 with $\hat{z}_{total}(\boldsymbol{p})$ we find that the number of deaths from 1999 to 2007 predicted from annual PM$_{2.5}$ exposure in excess of background levels is 905 560. These results were validated using the EPA's BenMAP program[30] and are consistent with other estimates.[31]

We then used eq 5 with $\hat{z}_{mobile}(\boldsymbol{p})$ (PM$_{2.5}$ explained by on road mobile emissions) and $\hat{z}_{stationary}(\boldsymbol{p})$ (PM$_{2.5}$ explained by stationary emissions). The mean of the $\hat{z}_{mobile}(\boldsymbol{p})$ across all the space/time data points is 3.4 μg/m^3, while the mean of $\hat{z}_{stationary}(\boldsymbol{p})$ across the same points is only 1.15 μg/m^3. Accordingly the number of deaths attributed to PM$_{2.5}$ explained by on road mobile emissions is greater than the number of deaths attributed to PM$_{2.5}$ explained by stationary emissions (Table 3). For instance, the number of deaths attributed to PM2.5 explained by on road mobile emissions is 568 090 from 1999 to 2007, which is 1.85 times more than the 306 316 deaths attributed to PM$_{2.5}$ explained by stationary emissions. Similarly, on road mobile emissions explained 1.86 times the number of IHD deaths and 1.98 times the number of LC deaths compared to deaths explained by stationary emissions. The number of deaths assumes that the relative risk used in eq 5 can be applied to the entire population and that estimated ambient concentration is a surrogate for exposure. This risk assessment does not incorporate the varying toxicity of PM$_{2.5}$.

This finding is interesting because, according to the NEI, primary PM$_{2.5}$, NO$_2$, SO$_2$, and NH$_3$ coming from on road mobile emissions sum up to 70 834 thousand tons from 1999 to 2007 while primary PM$_{2.5}$, NO$_2$, SO$_2$, and NH$_3$ coming from stationary emissions sum up to 293 446 thousand tons for the same time period (SI Table S2). Hence, even though on road mobile emissions emit only about a quarter of the mass emitted by stationary emissions, the number of deaths predicted from PM$_{2.5}$ explained by on road mobile emissions is almost twice that predicted from PM$_{2.5}$ explained by stationary emissions.

■ DISCUSSION

The first major outcome of our work is the creation of a global LUR model that predicts large area variability of PM$_{2.5}$ across the entire contiguous United States from 1999 to 2009. Only a handful of studies have developed LUR models that can be classified as "general" in that they produced results generalizable to domain sizes as large as ours (SI Figure S1). Although the LUR may perform better in some areas than others, the model is "generalizable" in a relative fashion when compare to LUR models developed over a smaller domain. To the best of our knowledge, the closest LUR models developed over such a large domain size are Hart et al.[15] and Beelen et al.[14] for annual PM$_{10}$, and Beckerman et al.[17] for monthly PM$_{2.5}$.

The Hart et al.[15] and Beelen et al.[14] studies developed regression models to predict annual PM$_{10}$ concentrations across the United States from 1985 to 2000 and across 15 European countries for 2001, respectively. Even though their models differed (i.e., the Hart et al.[15] model used traffic related variables while the Beelen et al.[14] model used meteorology and
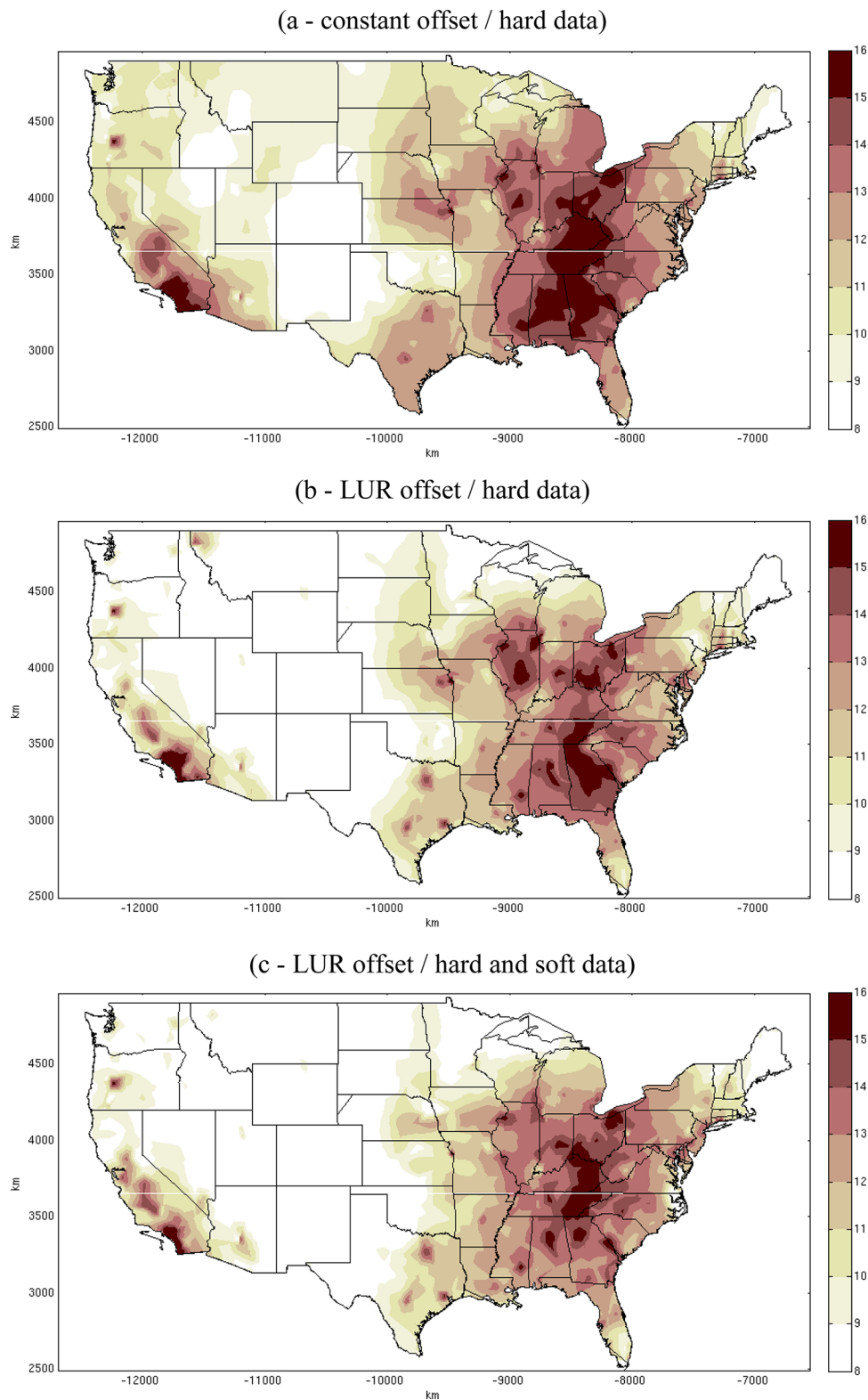
1740

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(a - constant offset / hard data)

(b - LUR offset / hard data)

(c - LUR offset / hard and soft data)

Figure 1. BME predicted annual $PM_{2.5}$ ($\mu g/m^3$) concentration estimation map across the contiguous U.S. on May 1, 1999 for the following methods: (a) constant offset/hard data; (b) LUR offset/hard data; and (c) LUR offset/hard and soft data.

land use), they produced similar r^2 of 49% and 41%, respectively. These studies provided substantial contribution to the literature on annual PM_{10}. However, there is a lack of comparable global models for $PM_{2.5}$. Our study is successful in helping to fill that knowledge gap by providing a general LUR

for $PM_{2.5}$ that achieves an r^2 of 53% that is comparable or better than that for annual PM_{10}.

Of the limited general LUR models developed for the long-term average concentration of $PM_{2.5}$, the LUR-without-remote-sensing model developed by Beckerman et al.[17] is the most comparable to ours. The explanatory variables of that model are

Table 2. Cross Validation Statistical Measures and Percent Change for Three Estimation Methods

method	LUR only	(a) constant/ hard	(b) LUR/ hard	(c) LUR/ hard and soft	% change from (a) to (b)	% change from (b) to (c)
MSE[a]	7.04	1.69	1.32	1.26	−21.89	−4.87
RMSE[b]	2.65	1.30	1.15	1.12	−11.62	−2.46
MAE[b]	1.97	0.79	0.63	0.63	−20.73	−0.45
MR[b]	1.86	1.87	1.12	1.07	−40.25	−4.08
Square Pearson's Corr.[c]	0.50	0.68	0.87	0.88	28.94	0.78
Square Spearman's Corr.[c]	0.55	0.67	0.89	0.89	32.13	0.32

$^a[\mu g/m^3]^2$. $^b\mu g/m^3$. ^{c}Unitless.

Table 3. Death Counts Predicted from Annual PM$_{2.5}$ Explained by on Road Mobile and Stationary Emissions

	predicted from on road mobile emissions	predicted from stationary emissions
1999−2007 all cause mortality	568 090	306 316
1999−2007 ischemic heart disease deaths	415 163	223 341
1999−2007 lung cancer deaths	85 044	43 035

traffic within 1km and green space within 0.1km. The r^2 of that model was 3% for their training data set and 5% for their validation data set. This provides a substantial contribution to the literature because it describes the small area variability at a scale of 0.1−1km which is attributable to primary PM$_{2.5}$ emitted as ultrafine particles and specific metals that are responsible for a large portion of observed health impacts.[17] Hence their relatively small r^2 of 3−5% makes sense since on road emission of primary PM$_{2.5}$ makes up only a small fraction of annual PM$_{2.5}$. By contrast, our explanatory variables capture the large area variability of PM$_{2.5}$, as demonstrated by our hyperparameter values ranging from ten to hundreds of km (Table 1). These values are characteristic of the transport of secondary PM$_{2.5}$ over long distances. Hence our model describes the large area variability of secondary PM$_{2.5}$. Since the majority of PM$_{2.5}$ comes from secondary formation, it makes sense that our model achieves a higher r^2 (53%) than that of Beckerman et al.[17] (3−5%). Therefore their model describes short-area variability due to the local effect of traffic coming from primary PM$_{2.5}$, while our model describes the large area variability of PM$_{2.5}$ due to the long-range effect of secondary PM$_{2.5}$ created from on road mobile emissions and stationary emissions. To the best of our knowledge no other LUR models have predicted the large area variability of secondary PM$_{2.5}$ and our model is the first to address this important knowledge gap. Our LUR model estimated PM$_{2.5}$ coming from on road mobile emissions using TT, AC, and EE. However, due to the complex nature of PM$_{2.5}$, these variables may capture other sources outside of on road mobile emissions. We hypothesized that EE using population density corrects for over prediction of on road mobile emissions coming from TT and AC. Indeed, out of the

$$1 \times (1 + 2 + 1) \times \sum_{i=1}^{4} \binom{4}{i} = 1 \times 4 \times 15 = 60 \quad models$$

that had the EE variable, β_{EE} was positive for the $1 \times (1 + 1 + 0) \times 15 = 30$ models where EE appears without TT and it

consistently switched to being negative for the $1 \times (0 + 1 + 1) \times 15 = 30$ models that contain both the EE and the TT variable. This suggests that EE alone is a surrogate for on road mobile emissions. However, when paired with the TT traffic variable, EE corrects the overestimation of these variables. This finding is in agreement of our hypothesis and therefore supports the conclusion that population density can be used as a surrogate for increased EE of the vehicle fleet. Obtaining accurate estimates of on road mobile emissions along all roads is a difficult task. By using population data to calculate EE, we facilitate this task and as a result we ensure the accessibility of our model to a wider audience.

While previous LUR models represent important contributions to the field, our model differs in several important ways: (1) our model describes large area variability of PM$_{2.5}$, which characterizes the secondary component of this pollutant, (2) the explanatory variables are constructed from data that are easily obtainable by a wide audience and (3) our model allows to distinguish between PM$_{2.5}$ explained by on road mobile emissions and PM$_{2.5}$ explained by stationary emissions. To our knowledge this is one of the first LUR models to capture secondary PM$_{2.5}$ using easily obtainable explanatory variables describing on road mobile emissions and stationary emissions.

The second major outcome of this work is the combination of our LUR model with BME to create a combined LUR/BME hybrid estimation method for annual PM$_{2.5}$. In this hybrid approach, LUR is used as a first step to characterize global trends in PM$_{2.5}$ and BME is used to extract unexplained variability in the residuals. Our results (Table 2) demonstrate that LUR/BME is successful at combining the strengths of each of its component methods. Indeed, LUR/BME results in a 21.89% reduction in MSE and a 28.94% increase in r^2 over BME alone, which is itself more accurate than LUR alone. The population correction factor presented in the soft data variance in eq 4 does not account for the fact that annual PM$_{2.5}$ averages are correlated in time. As well, the number of daily values within a year $n_{i,t}$ does not account for the seasonality of missing values.

Others have combined LUR/BME such as Beckerman et al. Their work saw an r^2 of 0.79 using a validation data set comprised of about 10% of the data. By comparison we achieved an r^2 of 0.78 using a 10-fold cross validation, where each of validation points had similar distance-to-closest-monitor as those of Beckerman et al. A key difference between our works is that we extended their work by incorporating non-Gaussian soft data that rigorously accounted for the uncertainty associated with the incompleteness of daily samples. Our r^2 indicates that our model was successful in this novel incorporation of non-Gaussian soft data in the LUR/BME framework, which resulted in one of the most accurate LUR/BME estimations to date of annual PM$_{2.5}$ as supported by the fact that our r^2 is similar to that of Beckerman et al. A unique strength of our model is that these highly accurate LUR/BME estimates of annual PM$_{2.5}$ can be separated into the portions explained by on road mobile emissions and stationary emissions, which to our knowledge had not been done before to a similar level of precision.

Building on the novel contributions of the first two outcomes of our work, an important third outcome of this work is a risk assessment of annual PM$_{2.5}$ exposure explained from major contributing sources. Estimating annual PM$_{2.5}$ is useful for assessing long-term exposure needed to investigate chronic diseases. Others have already used LUR estimates in

epidemiological studies.[32−35] From 1999 to 2007 there were 568 090 deaths attributed to $PM_{2.5}$ explained by 70 834 thousand tons of primary $PM_{2.5}$, NO_2, SO_2, and NH3 emitted by on road mobile emissions, which correspond to a ratio of 8.02 deaths/thousand tons for on road mobile emissions. By contrast there were 306 316 deaths attributed $PM_{2.5}$ explained by 293 446 thousand tons of primary $PM_{2.5}$, NO_2, SO_2, and NH_3 emitted by stationary emissions, which correspond to a ratio of 1.04 deaths/thousand tons for stationary emissions. These results are informative because they imply that mechanisms involved in the creation and long-range transport of secondary $PM_{2.5}$ lead to substantially differing health impacts depending on whether emissions originate from on road mobile emissions or stationary emissions.

Other works have also examined excess mortality due to current emissions levels. When investigating Massachusetts power plants Levy and Spengler[36] found that current power plant emissions in the surrounding area that emitted above the best available control technology (BACT) resulted in approximately 70 deaths per year in a ∼ 600 km by 600 km region which includes areas of Massachusetts and New York where the power plants were located. According to the BACT of 3 lb/MWh of SO_2 and 1.5 lb/MWh of NO_X, there would be a reduction of 43 951 tons of SO_2 and 4376 tons of NO_X from the two power plants mentioned in the study. This would result in 1.34 deaths/thousand tons of SO_2 and 2.51 deaths/thousand tons of NO_X due to power plants emissions in the area being above the BACT. That work used the CTM CALPUFF in which emission levels can be zeroed out while our work uses an LUR model which measures annual predicted PM2.5. Levy only investigated power plants while our work looked at major contributing sources. Even though LUR cannot be directly compared to CTMs, our LUR results are useful in a relative manner as they allow us to contrast on road mobile emissions and stationary emissions which have not been done before.

In order to reduce the number of deaths due to $PM_{2.5}$ exposure, our results indicate a reduction in one ton of on road mobile emissions would be eight times more beneficial than a one ton reduction in stationary emissions. This may be accomplished though any number of actions such as increased accessibility and reliance on public transportation in areas of high population density to more stringent emission standards that would further promote fuel efficiency.

ASSOCIATED CONTENT

Ⓢ Supporting Information

Further explanation of estimation methods, LUR domain sizes, independent variables, covariance models, BME equations, cross-validation statistics and cross-validation results. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes
The authors declare no competing financial interest.

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