Published in final edited form as:
Environ Res. 2012 October ; 118: 8–15. doi:10.1016/j.envres.2012.06.011.

Use of satellite-based aerosol optical depth and spatial clustering to predict ambient $PM_{2.5}$ concentrations

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Abstract

Satellite-based $PM_{2.5}$ monitoring has the potential to complement ground $PM_{2.5}$ monitoring networks, especially for regions with sparsely distributed monitors. Satellite remote sensing provides data on aerosol optical depth (AOD), which reflects particle abundance in the atmospheric column. Thus AOD has been used in statistical models to predict ground-level $PM_{2.5}$ concentrations. However, previous studies have shown that AOD may not be a strong predictor of $PM_{2.5}$ ground levels. Another shortcoming of remote sensing is the large number of non-retrieval days (i.e., days without satellite data available) due to clouds and snow- and ice-cover.

In this paper we propose statistical approaches to overcome these two shortcomings, thereby making satellite imagery a viable method to estimate $PM_{2.5}$ concentrations. First, we render AOD a robust predictor of $PM_{2.5}$ mass concentration by introducing an AOD daily calibration approach through the use of mixed effects model. Second, we develop models that combine AOD and ground monitoring data to predict $PM_{2.5}$ concentrations during non-retrieval days. A key feature of this approach is that we develop these prediction models separately for groups of days defined by the observed amount of spatial heterogeneity in concentrations across the study region. Subsequently, these methodologies were applied to examine the spatial and temporal patterns of daily $PM_{2.5}$ concentrations for both retrieval days (i.e., days with satellite data available) and non-retrieval days in the New England region of the U.S. during the period 2000-2008. Overall, for the years 2000-2008, our statistical models predicted surface $PM_{2.5}$ concentrations with reasonably high R^2 (0.83) and low percent mean relative error (3.5%). Also the spatial distribution of the estimated $PM_{2.5}$ levels in the study domain clearly exhibited densely populated and high traffic areas. The method we have developed demonstrates that remote sensing can have a tremendous impact on the fields of environmental monitoring and human exposure assessment.

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Conflict of Interest: The authors declare that they have no actual or potential conflict of interest.

Keywords

MODIS; aerosol optical depth; AOD; $PM_{2.5}$; mixed effects model; cluster analysis; $PM_{2.5}$ prediction model

1. Introduction

Particle pollution has been recognized as a significant concern related to human health and global climate change in many parts of the world (Brunekreef and Holgate, 2002; Ramanathan et al. 2001). Airborne particulate matter with aerodynamic diameter ≤2.5 μm ($PM_{2.5}$) is a mixture of pollutants including sulfate, nitrate, ammonium, organic compounds, elemental carbon, metal oxides, and dust or soil particles (U.S. EPA 2004). Numerous studies have shown that ambient $PM_{2.5}$ concentrations are associated with adverse health effects such as increased mortality and morbidity, aggravated respiratory and cardiovascular symptoms, and lower birth weight (Bell et al. 2007, 2010a; Franklin et al. 2007; Gent et al. 2003, 2009). In most epidemiological studies, subject-specific $PM_{2.5}$ exposures are generally assessed by measuring ambient $PM_{2.5}$ concentrations at one or more outdoor monitoring sites. However, sparse $PM_{2.5}$ monitoring spatial networks may limit our ability to accurately assess human exposures to $PM_{2.5}$, since concentrations measured at an outdoor site may be less representative of the subjects' exposures as the distance from the monitor increases (Bell et al. 2010b; Lee et al. 2011a). In time-series analyses $PM_{2.5}$ exposures should be highly correlated with ambient $PM_{2.5}$ concentrations. In cross-sectional studies long-term $PM_{2.5}$ exposures should be assessed with great accuracy (Bell et al. 2010b; Ito et al. 2004; Pinto et al. 2004). Furthermore, in interest of reducing cost, $PM_{2.5}$ monitoring sites operate only a few days per week at varying frequencies such as every day, every third day, and every sixth day. Thus epidemiological studies are often compromised due to the lack of continuous measurements. In conclusion, due to their spatial and temporal limitations, current $PM_{2.5}$ monitoring networks cannot provide sufficient data to fully assess $PM_{2.5}$ human exposures for health effect studies and are hindered in their ability to help answer some key scientific questions, such as the effect of cumulative exposures over several days.

Satellite remote sensing provides data on aerosol optical depth (AOD), a measure of light extinction by atmospheric aerosols (i.e., light scattering and absorption). AOD values reflect particle abundance in the atmospheric column, and thus they have been used in statistical models to predict ground-level $PM_{2.5}$ concentrations (Engel-Cox et al. 2004; Liu et al. 2005, 2007a, 2007b, 2007c, 2009; Schaap et al. 2009). Satellite-based $PM_{2.5}$ monitoring has been considered to complement ground $PM_{2.5}$ monitoring networks, especially for regions with a limited number of $PM_{2.5}$ monitors. However, most of previous studies have reported that AOD has a low to moderate $PM_{2.5}$ predictive ability (i.e., coefficient of determination $R^2 <$ 0.60) (Hoff and Christopher, 2009), which may not be sufficient for health effect studies. In a recent paper we introduced a new daily calibration technique for Moderate Resolution Imaging Spectroradiometer (MODIS) AOD to accurately predict ground $PM_{2.5}$ concentrations (Lee et al. 2011b).

AOD values cannot be retrieved on days with clouds, high surface reflectance due to snow- and ice-cover, or retrieval errors. As a result, AOD data are not available for a large fraction of days (non-retrieval days) and thus $PM_{2.5}$ concentration predictions are not always possible.

Due to the low to moderate $PM_{2.5}$ predictability of AOD measurements and the large number of non-retrieval days, satellite remote sensing has played a limited role in the field of particle exposure assessment. As mentioned above, we have already addressed the low

predictability issue by introducing the daily AOD calibration approach. The challenge of the infrequent satellite measurements will be addressed in this paper. Specifically, we have developed a statistical model to predict daily $PM_{2.5}$ concentrations for both retrieval and non-retrieval days in the region of New England, U.S. for the years 2000-2008. This is of paramount importance in our efforts to enhance spatial and temporal coverage of $PM_{2.5}$ concentration estimates, leading to more reliable environmental impact assessment, exposure assessment and health effect studies. Together these efforts render satellite remote sensing a powerful tool in the fields of environmental monitoring and human exposure assessment.

2. Methods

2.1. $PM_{2.5}$ measurements

$PM_{2.5}$ ambient air samples were collected at 69 U.S. Environmental Protection Agency (EPA) $PM_{2.5}$ monitoring sites in Connecticut (CT), Massachusetts (MA), Rhode Island (RI), Southern Maine (ME), New Hampshire (NH), and Vermont (VT) for the years 2000-2008 (**Fig. 1**). At the 69 monitoring sites, 24-hr integrated $PM_{2.5}$ filter samples were collected with varying frequencies including every day, every third day, and every sixth day as per EPA's monitoring program design. Not all monitoring sites operated for the entire nine years thus the number of monitoring sites varied by year.

2.2. Satellite data

We obtained MODIS AOD data (Collection 5; Level 2 aerosol product) from the National Aeronautics and Space Administration (NASA)'s Earth Observing System (EOS) satellites, Terra and Aqua, over the New England region for the years 2000-2008. The MODIS AOD measurements have a relatively fine spatial (10×10 km^2 grid) and temporal (every one to two days) resolutions, which makes them appropriate for daily air quality monitoring (Al-Saadi et al. 2005). The over-land retrieval algorithm of Collection 5 primarily uses three wavelength channels of 0.47, 0.66, and 2.12 μm (Levy et al. 2007), finally reporting AOD values at the wavelength of 0.55 μm, with the expected uncertainty of $\Delta AOD = \pm 0.05 \pm 0.15 \times AOD$ (Levy et al. 2010; Remer et al. 2008). The Terra and Aqua satellites cross the equator at two different times, approximately 10:30 am (descending orbit) and 1:30 pm (ascending orbit) local sun times, respectively, providing aerosol information at two different times per day with a scanning swath of 2,330 km (cross-track) by 10 km (along-track at nadir). Details about the MODIS AOD retrieval algorithm can be found in Levy et al. (2007, 2009). The daily averages of Terra and Aqua AOD values are most likely to reflect the aerosol loading on a given day, but both AOD values are not always retrieved each day. Due to the diurnal variations (Green et al. 2009) and potential calibration differences between two satellite sensors, averaging the Terra and Aqua AOD measurements would not be appropriate depending on the data availability on a given day. Moreover, the Terra satellite was launched in December, 1999, while the Aqua satellite was launched in May 2002, thus only Terra measurements are available for the years 2000-2002. Consequently, we primarily used the Terra AOD measurements. The missing Terra AOD values were estimated from the Aqua AOD ones, if they were available, using an adjustment factor. This factor was equal to the ratio of the average Terra AOD to Aqua AOD for those days when both the Terra and Aqua measurements were available. We created 582 grid cells (10×10 km^2) covering the New England region in ArcGIS (Version 9.3; ESRI), and all the subsequent analyses were based on the grid cells.

2.3. Statistical model

$PM_{2.5}$ prediction for retrieval days—We have previously introduced a daily AOD calibration method that renders AOD a robust predictor of surface-level $PM_{2.5}$

concentrations. This calibration method assumes minimal spatial variability of time-varying parameters influencing $PM_{2.5}$-AOD relationships on a given day (Lee et al. 2011b). The proposed calibration approach relied on daily ground $PM_{2.5}$ concentrations measured at multiple sites. Daily $PM_{2.5}$-AOD relationships were derived using a mixed effects model with random intercepts and slopes (Fitzmaurice et al. 2004) as follows:

$$PM_{ij} = (\alpha + u_j) + (\beta + v_j) \times AOD_{ij} + s_i + \varepsilon_{ij}$$
$$(u_j \quad v_j) \sim N[(0 \quad 0), \Sigma] \tag{1}$$

where PM_{ij} is the $PM_{2.5}$ concentration at a spatial site i on a day j; AOD_{ij} is the AOD value in the grid cell corresponding to site i on a day j; α and u_j are the fixed and random intercepts, respectively; β and v_j are the fixed and random slopes, respectively; $s_i \sim N(0, \sigma_s^2)$ is the random intercept of site i; $\varepsilon_{ij} \sim N(0, \sigma^2)$ is the error term at site i on a day j; and Σ is the variance-covariance matrix for the day-specific random effects. The fixed effects represent the average intercept and $PM_{2.5}$-AOD slope, and the random effects explain the daily-varying relationships. Due to the large number of sampling days it was not possible to run the mixed effects model for the entire study period. Therefore, we split the whole dataset (Years 2000-2008) into 5 subsets (Years 2000-2001, 2002-2003, 2004-2005, 2006-2007, and 2008). To build the mixed effects model, we matched the $PM_{2.5}$ concentrations measured at a monitoring site and AOD values obtained for the corresponding grid cells. We removed the sampling days with only one $PM_{2.5}$-AOD pair on a given day before running the model, since a slope cannot be determined with only one pair. In addition, we excluded those days when the root mean squared error (RMSE) between the measured and predicted $PM_{2.5}$ concentrations was greater than 5 $\mu g/m^3$ or when the estimated AOD slope was negative. These days were not considered reliable for calibrating AOD data due to $PM_{2.5}$ instrumental measurement errors, cloud contaminated AOD, or any other potential errors. The number of $PM_{2.5}$-AOD pairs for each data subset was 1,299 (2000-2001), 1,801 (2002-2003), 1,680 (2004-2005), 1,745 (2006-2007), and 972 (2008). We assessed the model performance using the coefficient of determination (R^2) and percent mean relative errors (% MRE) between the measured and predicted $PM_{2.5}$ concentrations. We obtained the measured $PM_{2.5}$ concentrations from 69 EPA monitoring sites and the predicted values from our model estimations in the grid cells corresponding to the respective monitoring sites. The % MRE is calculated as [|mean predicted $PM_{2.5}$ – mean measured $PM_{2.5}$| / (mean measured $PM_{2.5}$)] × 100. The R^2 values show how well the measured and predicted $PM_{2.5}$ concentrations are correlated, while the % MRE values present systematic differences between those concentration levels. Together, the values of R^2 and % MRE are indicative of the ability of our modeling approach to produce reliable $PM_{2.5}$ estimates for both time-series and cross-sectional health effect studies.

We validated the mixed effects model using a cross-validation (CV) method which checked for potential over-fitting. First, we randomly separated the entire dataset into 10 different subsets, each of them encompassing approximately 10% of the data. Each 10% subset of data was retained from the dataset, and the rest 90% of the data was used to fit the model. The fitted model was applied to predict $PM_{2.5}$ concentrations for the 10% of the retained days. This process was repeated for each of the 10 subsets, and the predicted $PM_{2.5}$ concentrations were compared to the measured $PM_{2.5}$ concentrations using R^2 and % MRE values.

$PM_{2.5}$ prediction for non-retrieval days—The spatial patterns of the observed daily $PM_{2.5}$ concentrations vary due to changes in meteorology and source emissions which influence the impact of local and regional sources in the study region. A cluster analysis

using K-means was performed to identify groups of days with similar $PM_{2.5}$ concentration spatial patterns in the R software. This analysis was based on the $PM_{2.5}$ concentrations measured at the sampling sites within the study region and included the entire dataset (i.e., retrieval and non-retrieval days for the 9 year period). The K-means method is used to partition observations into K different subsets (i.e., clusters), yielding a solution that minimizes the within-cluster variance and maximizes the between-cluster variance. That is, the cluster analysis determines groups of days exhibiting similar spatial concentration patterns.

As mentioned above the number of available $PM_{2.5}$ monitoring sites varied by day due to differences in sampling frequencies, site operating period, and missing data. Due to sampling design considerations, a large number of sites (>35 out of 69) operated every third day. The $PM_{2.5}$ measurements in the sampling frequency of every third day were always on the same day for all sites. Thus the cluster analysis was applied to every third day data, which made it possible to obtain reliable day-specific $PM_{2.5}$ spatial patterns. Furthermore, the cluster analysis was performed on $PM_{2.5}$ concentration differences, obtained by subtracting the daily regional $PM_{2.5}$ concentrations from the respective $PM_{2.5}$ concentrations. On a given day the regional $PM_{2.5}$ concentration was calculated by averaging the daily $PM_{2.5}$ concentrations measured at all available monitoring sites. Since cluster assignment was done for every third day, the same cluster classification was applied to the adjacent two days, assuming an identical spatial pattern for three consecutive days. Finally, an important feature of the proposed approach is that the cluster analysis is independent of satellite data retrieval because it was based on $PM_{2.5}$ ground measurements.

Following the mixed effects model and the cluster analysis, $PM_{2.5}$ concentrations in each of the grid cells were predicted for days when no AOD values were available. Toward this end, a cluster-specific $PM_{2.5}$ prediction model was developed which used a generalized additive model (GAM) (Hastie and Tibshirani, 1990) as follows:

$$PM_{predicted\ \ ij}=\beta_0+\beta_1\times PM_{regional\ \ j}+s(latitude,longitude)_i+\varepsilon_{ij} \qquad (2)$$

where $PM_{predicted\ ij}$ is the AOD-derived $PM_{2.5}$ concentration at a spatial site i on a day j (from equation (1)); $PM_{regional\ j}$ is the regional $PM_{2.5}$ concentration on a day j; $s(latitude, longitude)_i$ is a smooth function of location (latitude and longitude) for site i, and ε_{ij} is the error term at site i on a day j. We modeled this smooth function as a thin plate spline as implemented in the R software package. As shown by equation (2), the predicted $PM_{2.5}$ concentrations from the mixed effects model were used as a dependent variable. For each cluster, these $PM_{2.5}$ concentration values were regressed on the regional $PM_{2.5}$ levels and the spatial smooth function of latitude and longitude. As a result, a predicted spatial surface of $PM_{2.5}$ concentrations could be generated for each group of days defined by the clustering algorithm. For each of the non-retrieval days $PM_{2.5}$ concentrations in each grid cell were estimated by assuming that each cluster had a single $PM_{2.5}$ spatial surface and the regional $PM_{2.5}$ concentrations reflected the temporal variability of $PM_{2.5}$ over the study domain. As mentioned above, each cluster includes both retrieval and non-retrieval days because the cluster analysis was independent of AOD data retrieval. The model performance for non-retrieval days was also examined by comparing the measured and predicted $PM_{2.5}$ concentrations using R^2 and % MRE estimates. In order to compare the GAM predicted $PM_{2.5}$ concentrations to the measured ones at the monitoring sites we adjusted for site bias using the random site estimates from the mixed effects model. The statistical approach to predict $PM_{2.5}$ concentrations within the study domain for non-retrieval days is summarized in **Fig. 2**.

2.4. Spatial variability of $PM_{2.5}$ levels

We predicted concentrations for all retrieval and non-retrieval days during the 9-year period. Subsequently, we estimated the 9-year average $PM_{2.5}$ concentrations for each of the grid cells in the study domain. We also present the $PM_{2.5}$ concentration maps for each of the identified clusters. For the concentration maps, we split the distributions into 6 equally-sized bins due to the log-normally distributed $PM_{2.5}$ levels.

3. Results and Discussion

3.1. Descriptive statistics

The mean (SE) $PM_{2.5}$ concentrations measured at the 69 EPA monitoring sites varied from 7.96 (0.32) $\mu g/m^3$ in Lebanon, NH (Site ID: 33-09-0010) to 16.38 (0.22) $\mu g/m^3$ in New Haven, CT (Site ID: 09-09-0018). The overall mean $PM_{2.5}$ concentration across the monitoring sites was 11.07 $\mu g/m^3$ (SD=1.62 $\mu g/m^3$). The $PM_{2.5}$ concentrations measured at the spatial sites were not based on the same number of sampling days due to differences in sampling frequencies, site operation periods, or missing data, thus, the reported $PM_{2.5}$ levels may not be directly comparable. Mean (SE) daily AOD values for the grid cells covering the New England region varied from 0.06 (0.01) to 0.30 (0.01). On average 627 AOD values per grid cell were retrieved which corresponds to 19.1% of the study period.

3.2. Model prediction

The mixed effects model generated 994 daily $PM_{2.5}$-AOD relationships for the years 2000-2008. The number of the determined relationships did not vary much by year, ranging from 94 in 2000 to 131 in 2007 and 2008. However, the number of the relationships varied considerably by season, with summer being the highest (N=329) followed by fall (N=324), spring (N=253), and winter (N=88). The low number of retrieval days in winter was due to the larger number of days with clouds or snow in this season. Note that the number of $PM_{2.5}$ ground measurements is generally constant throughout the year. The fixed effects of intercepts and slopes (AOD) for each of the 5 data subsets 2000-2001, 2002-2003, 2004-2005, 2006-2007, and 2008 were statistically significant ($p<0.05$), and the random effects of intercepts and slopes varied substantially by day. The daily intercepts and slopes (the mean of fixed plus random effect estimates) varied by season: 8.43 (SD=3.98), 7.98 (SD=3.86), 11.02 (SD=5.52), and 8.99 (SD=4.50) for intercepts; 8.18 (SD=4.12), 7.22 (SD=4.18), 9.25 (SD=5.31), and 8.49 (SD=4.63) for slopes in winter, spring, summer, and fall, respectively. The random site estimates for densely populated and high traffic areas were positive, indicating the necessity to include the site term in the mixed effects model to adjust for site bias. The cluster analysis of the entire dataset for the years 2000-2008 yielded 9 different clusters. Each of the clusters consisted of 1,404 (42.7%), 678 (20.6%), 441 (13.4%), 189 (5.8%), 162 (4.9%), 132 (4.0%), 105 (3.2%), 96 (2.9%), and 81 (2.5%) days, respectively. For all the cluster-specific performed GAMs, corresponding regional $PM_{2.5}$ concentrations and spatial smooth function of coordinates were statistically significant ($p<0.05$).

The results of mixed effects model used to estimate $PM_{2.5}$ concentrations for retrieval days are shown in **Fig. 3**. The model explained 93% of the variability in the measured $PM_{2.5}$ concentrations obtained at the 69 monitoring sites ($R^2=0.93$). There was a good agreement between the measured and predicted $PM_{2.5}$ concentrations [slope=1.02 (SE=0.003) and intercept=-0.20 (SE=0.043)]. In addition, the cross-validation (CV) mixed effects model explained 88% of the variability in the observed $PM_{2.5}$ concentrations ($R^2=0.88$) with a slope of 1.00 (SE=0.004) and an intercept of 0.02 (SE=0.054). This suggests that AOD can be a robust predictor of $PM_{2.5}$ in the mixed effects model. Also the model performance using these two simple linear regression models between the measured and predicted $PM_{2.5}$

concentrations suggested that excessive over-fitting did not occur and the mixed effects model can be reliably applied to any grid cell in the study region.

The site-specific $PM_{2.5}$ predictability was examined by estimating the R^2 and % MRE values for each of the 69 monitoring sites for both retrieval and non-retrieval days (**Fig. 4**). Each monitoring site had the measured and predicted $PM_{2.5}$ concentrations, and the comparison between the measured and predicted $PM_{2.5}$ levels showed the R^2 and % MRE for each site. These 69 R^2 values and 69 % MRE values were represented in each box plot of **Fig. 4**. For retrieval days, the average R^2 and % MRE for 69 monitoring sites were 0.90 (SD=0.06) and 1.5 (SD=1.8) %, respectively. For the non-retrieval days, the average R^2 and % MRE values were 0.80 (SD=0.10) and 6.1 (SD=4.4) %, respectively. Therefore, models for both the retrieval and non-retrieval days predicted $PM_{2.5}$ concentrations accurately with reasonably high R^2 and low % MRE values for most spatial sites. As expected, model performance was slightly better for retrieval days. In the past, low predictive power and a large fraction of non-retrieval days have limited the application of satellite-based $PM_{2.5}$ exposure assessment to epidemiological studies.

Model performance tests were conducted by year and season (**Table 1**). Overall, our statistical models predicted surface $PM_{2.5}$ concentrations with high R^2 (0.83) and low % MRE (3.5%) values. Yearly analysis showed constantly high R^2 values ranging from 0.73 in 2000 to 0.87 in 2007 and low % MRE varying values from 2.3% in 2003 to 4.8% in 2002. Moreover, seasonal comparisons show that daily ground $PM_{2.5}$ concentrations can be reliably estimated for all four seasons: R^2 ranged from 0.75 in winter to 0.87 in summer and % MRE varied from 2.1% in spring to 5.0% in winter. The $PM_{2.5}$ predictive ability in winter was lower than for other seasons, although the model performance was still reasonable. This may be explained by the higher proportion of non-retrieval days during the winter.

Our study suggests that when satellite data are appropriately modeled they can be used to predict $PM_{2.5}$ exposures for epidemiological studies based on temporal (e.g., time-series and case-crossover studies) and spatial (e.g., cross-sectional studies) variation in pollutant concentrations. Time-series and case-crossover studies investigate associations between day-to-day variations in exposures and health outcomes. Therefore, the high correlations between the measured and predicted $PM_{2.5}$ concentrations (assessed by Pearson correlations or R^2) indicate that the satellite-based $PM_{2.5}$ predictions can provide reliable exposure estimates for longitudinal health effect studies. Furthermore, cross-sectional studies examine associations between health effects and long-term $PM_{2.5}$ exposures across communities. The very good agreement between the site measured and predicted mean $PM_{2.5}$ concentrations, as assessed by % MRE, suggests that satellite remote sensing can enhance our ability to assess $PM_{2.5}$ exposures for the cross-sectional studies. Often $PM_{2.5}$ concentrations vary spatially (Hoek et al. 2002; Kim et al. 2005). Therefore, using one or a limited number of outdoor monitors may not be sufficient to produce accurate human exposure assessments. This exposure error can result in the underestimation of health risks associated with $PM_{2.5}$ exposures (Jerrett et al. 2005; Thomas et al. 1993; Zeger et al. 2000). Use of MODIS AOD data helps to capture some or most of the spatial heterogeneity thus reducing exposure error.

3.3. Spatial patterns of $PM_{2.5}$ concentrations

The spatial distribution of the predicted 9-year average $PM_{2.5}$ concentrations is shown in **Fig. 5**. In the figure, each grid cell value represents the average of daily predicted $PM_{2.5}$ concentrations for 9 years. Our prediction models estimated daily $PM_{2.5}$ concentrations for the entire study period (2000-2008) including both retrieval and non-retrieval days. Thus the number of the predicted $PM_{2.5}$ concentrations in each grid cell is identical, and the reported grid cell concentrations can be compared directly. The estimated average $PM_{2.5}$ concentrations during the period of 2000-2008 ranged from 10.25 to 11.44 $\mu g/m^3$. Note that

on a given day concentration ranges were larger. Higher concentrations were predicted for densely populated and high traffic areas (e.g., Bridgeport, Hartford, and New Haven, CT, Boston and Springfield, MA, and Providence, RI) and high point emission source areas (e.g., power plants located in the coastal cities, Somerset and Salem, MA) (U.S. EPA, 2008). Previous studies have shown that the Northeastern cities of the U.S. are mostly impacted by the regionally transported $PM_{2.5}$ pollution (Lee et al. 2011a; Liu et al. 2003). As a result, average $PM_{2.5}$ concentrations tend to exhibit low spatial variability throughout the study region (Burton et al. 1996; Suh et al. 1997).

$PM_{2.5}$ concentration spatial patterns may vary daily depending upon the prevailing meteorological conditions and the location and characteristics of the impacting PM and gaseous pollutant sources (Seinfeld and Pandis 2006). The New England area is the receptor of pollution mostly transported by northwestern, western, and southwestern winds. In addition, this region is impacted by emissions produced within the metropolitan New York area. Therefore, the spatial patterns and composition of particles in the New England depend on many time-varying parameters. However, it is possible to distinguish discrete spatial patterns that may reflect certain synoptic conditions. Using cluster analysis we were able to identify 9 distinct spatial patterns (clusters) for the entire 9 year study. The 9 concentration maps are shown by **Fig. A1**. In the figure, each concentration map characterized different spatial patterns of $PM_{2.5}$ in terms of spatial gradients and $PM_{2.5}$ levels. The spatial gradients clearly displayed a group of days (i.e., clusters) influenced by transported pollution from the metropolitan New York area and Canada. The average $PM_{2.5}$ concentration levels and the range of the $PM_{2.5}$ levels (i.e., the highest minus lowest $PM_{2.5}$ concentrations) varied by cluster. This may be due to $PM_{2.5}$ source locations/emission rates, local meteorology (i.e., prevailing winds and stability), and the relative contributions of transported and local pollution over the region. The 9 distinct cluster-specific concentration maps, as shown in **Fig. A1**, provide evidence that the cluster analysis successfully captured and represented the heterogeneous $PM_{2.5}$ spatial patterns in the study region. Moreover, these spatial patterns help us qualitatively examine the characteristics of particle pollution (e.g., days that are strongly influenced by transported pollution).

4. Conclusions

We have introduced a new approach that uses satellite AOD data to predict the spatial and temporal patterns of $PM_{2.5}$ levels in New England. Our method is based on the daily calibration of AOD measurements using ground-level $PM_{2.5}$ concentrations, which was accomplished using a mixed effects model. These calibrations are necessary, since the relationship between AOD and $PM_{2.5}$ concentrations depends on many time-varying parameters such as particle concentration vertical profile, particle composition, and relative humidity among others. Daily calibration renders AOD a better predictor of $PM_{2.5}$, and it represents a significant improvement over previous studies, which assume a constant relationship between the two parameters.

Furthermore, we have proposed a new method to predict $PM_{2.5}$ concentrations during non-retrieval days, which are quite frequent in New England due to clouds and snow. The cluster analysis, which was based on the analysis of ground $PM_{2.5}$ measurements obtained at a large number of sites in New England, identified 9 distinct spatial patterns of $PM_{2.5}$. Each of the spatial patterns determined a single $PM_{2.5}$ spatial surface, generating 9 cluster surfaces of $PM_{2.5}$ over the study period. The cluster-specific analysis allowed accurate prediction of all the missing $PM_{2.5}$ concentrations in each grid cell. The cluster analysis is crucial in predicting ground $PM_{2.5}$ concentrations for non-retrieval days, while overcoming the limitation of satellite data availability.

Overall, we have demonstrated the tremendous potential of satellite AOD data to accurately predict exposures to $PM_{2.5}$. These data are necessary for both short- and long-term $PM_{2.5}$ epidemiological studies. As satellite remote sensing improves, data with finer spatial and temporal resolutions will become available in the future, leading to more accurate $PM_{2.5}$ exposure estimates. With regard to the MODIS, AOD data with the spatial resolution of 3 km is expected in the near future. This improvement will enhance our ability to assess daily subject-specific $PM_{2.5}$ exposures, since data with finer spatial resolution may further reduce exposure measurement errors.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

The authors thank the Harvard-EPA Clean Air Research Center and the Yale Center for Perinatal, Pediatric and Environmental Epidemiology. This publication was made possible by USEPA grant RD 83479801. Its contents are solely the responsibility of the grantee and do not necessarily represent the official views of the USEPA. Further, USEPA does not endorse the purchase of any commercial products or services mentioned in the publication. This publication was also supported by NIEHS grants R01ES016317 and R01ES019587.

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Highlights

- Satellite-based $PM_{2.5}$ prediction has the potential to monitor $PM_{2.5}$ air quality.
- We use an AOD daily calibration approach to predict $PM_{2.5}$ for retrieval days.
- The amount of $PM_{2.5}$ spatial heterogeneity can be observed.
- These enable us to develop $PM_{2.5}$ prediction models for non-retrieval days.

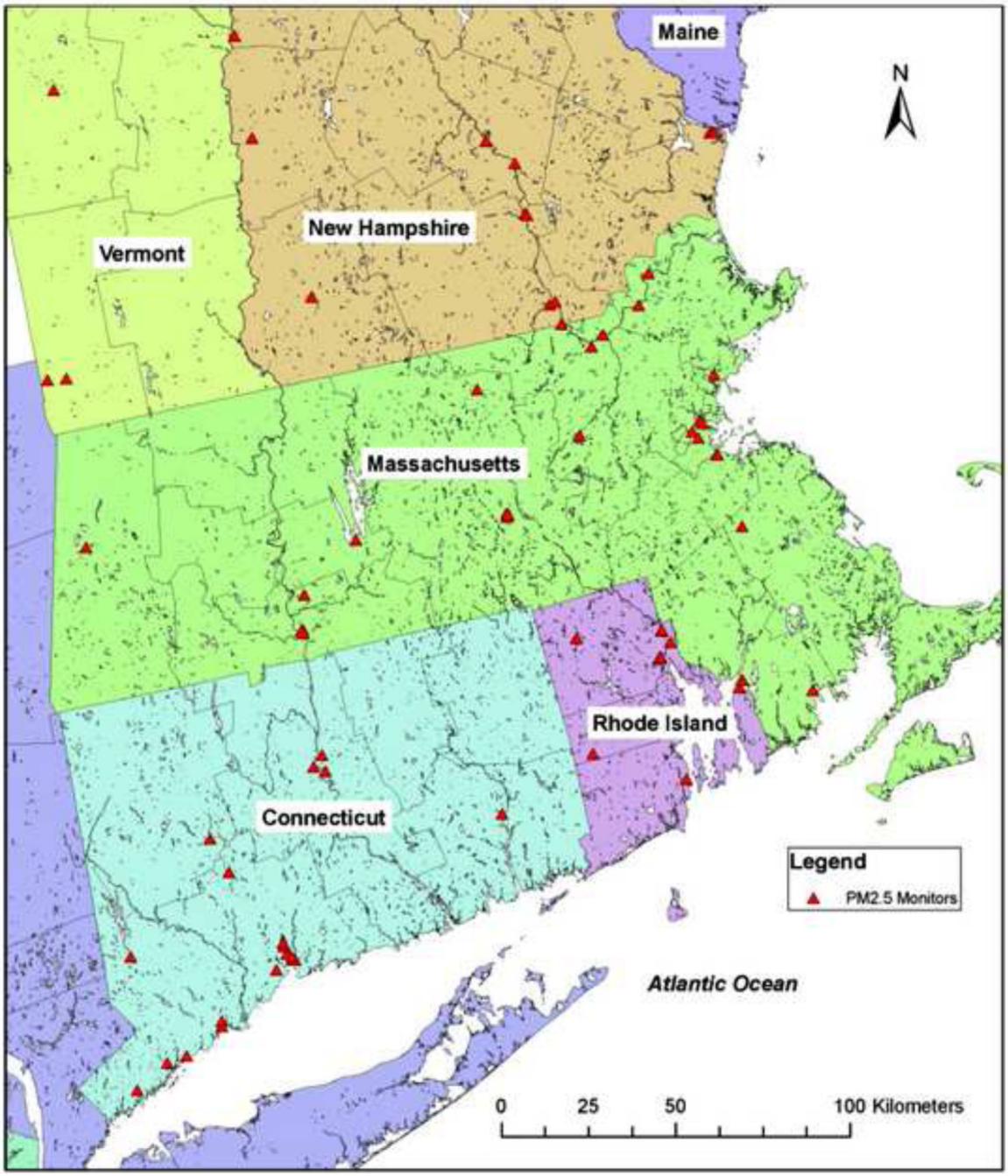

Fig. 1.
Location of 69 EPA $PM_{2.5}$ monitoring sites in the study region. This study region is covered by 582 grid cells (10×10 km^2).

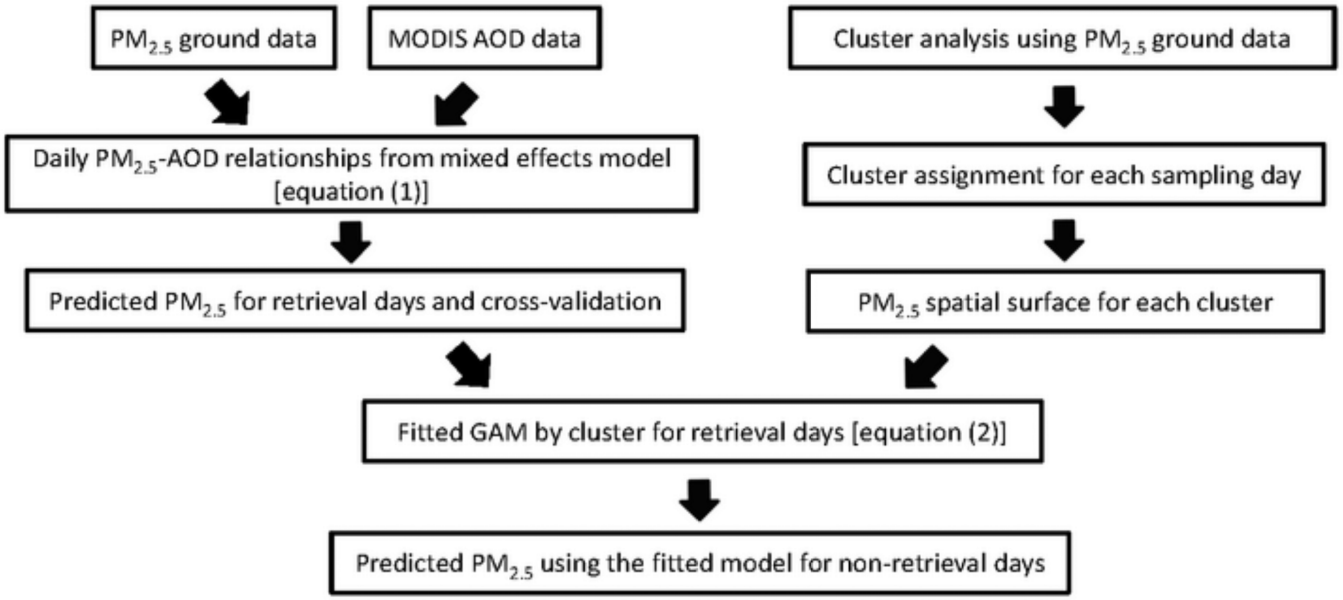

Fig. 2.
Flowchart summarizing $PM_{2.5}$ prediction for non-retrieval days.

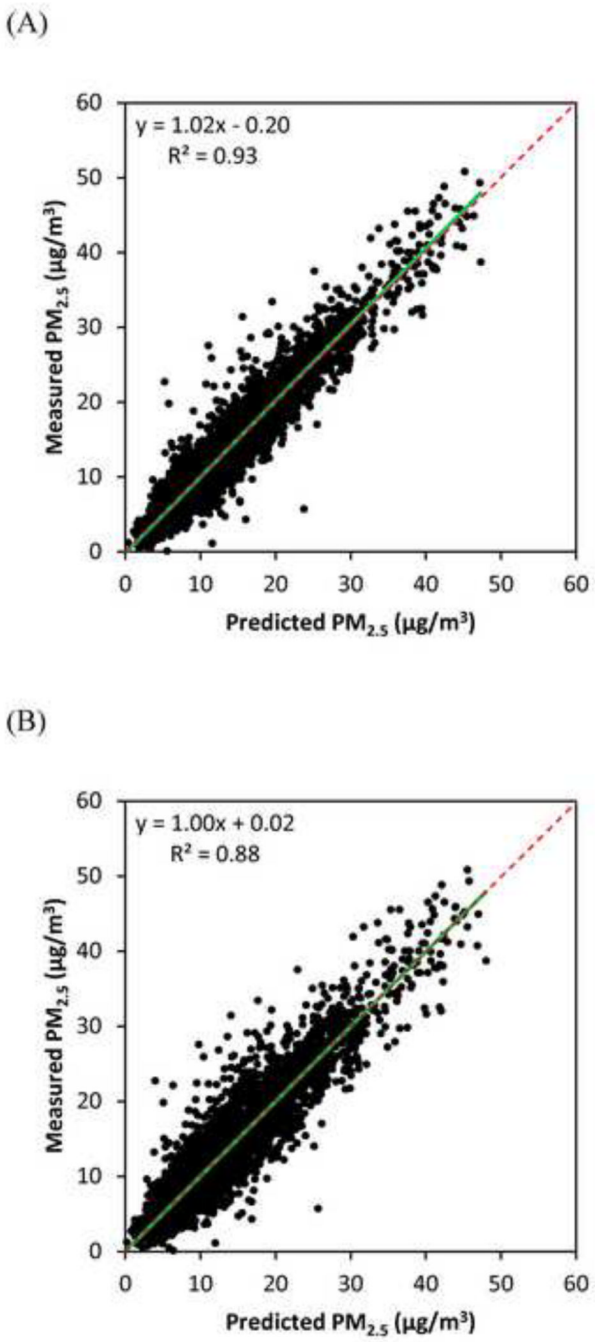

Fig. 3.

Model performance for retrieval days for the years 2000-2008 (Unit: $\mu g/m^3$): (A) Mixed effects model and (B) CV mixed effects model. The green solid line presents the regression line, and the red dashed line displays the 1:1 line indicating perfect agreement.

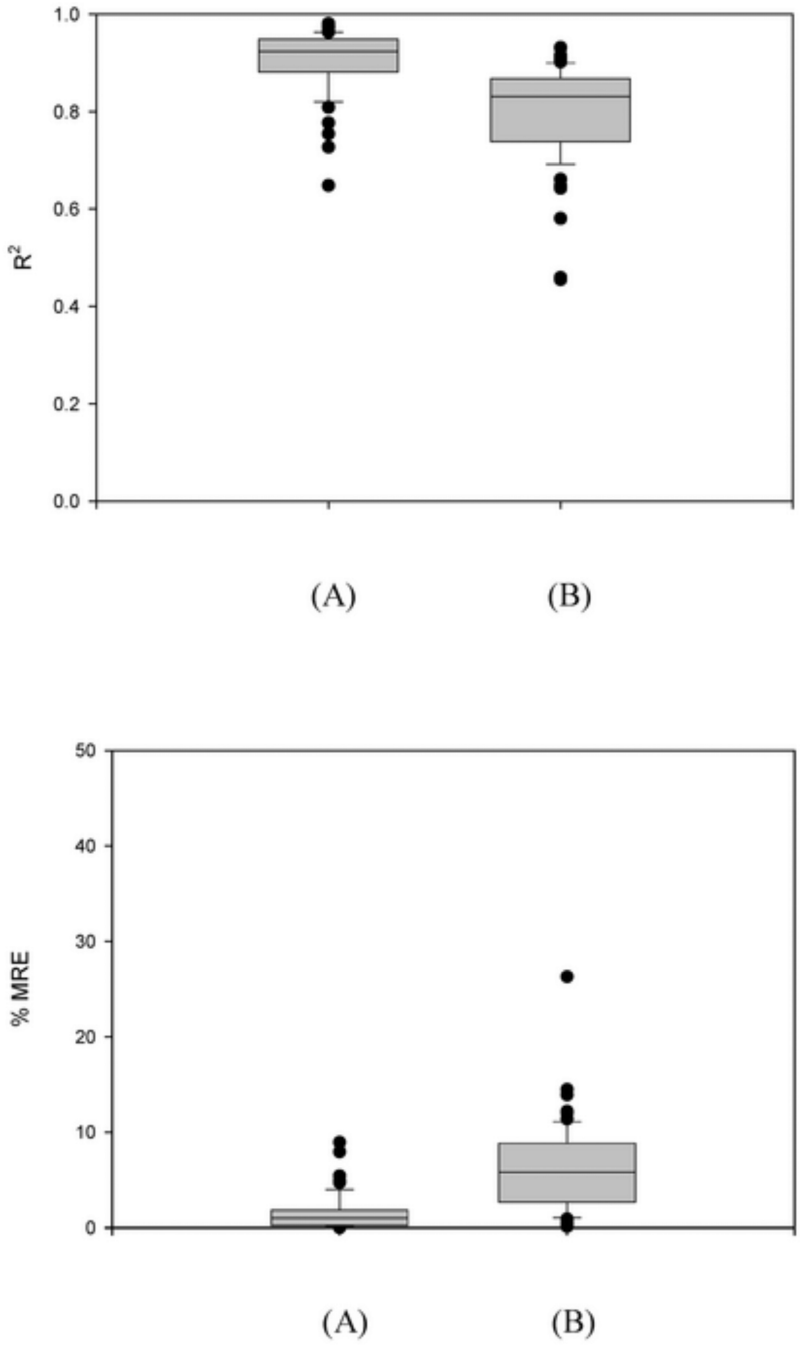

Fig. 4.

Site-specific $PM_{2.5}$ predictability using R^2 and % MRE for the years 2000-2008: (A) Retrieval days and (B) Non-retrieval days. The R^2 and % MRE values were based on the comparison between the measured and predicted $PM_{2.5}$ concentrations by site. Each box plot represents 69 monitoring sites.

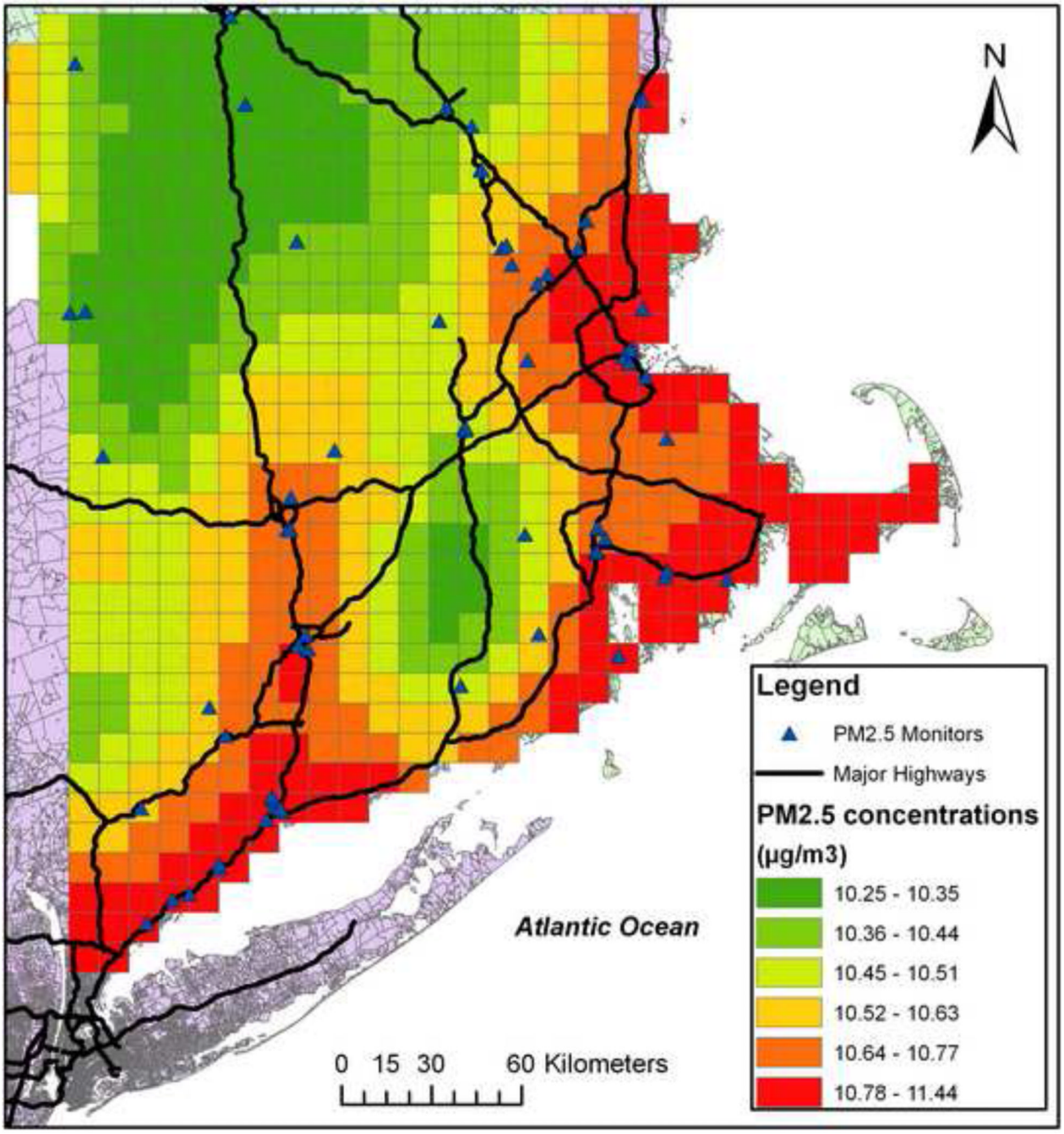

Fig. 5.
Spatial distribution of the 9-year average predicted $PM_{2.5}$ concentrations.

Table 1

Overall, yearly, and seasonal comparisons between the measured and predicted $PM_{2.5}$ concentrations. The measured and predicted $PM_{2.5}$ concentrations and bias are in the unit of $\mu g/m^3$.

	N	$PM_{2.5\ measured}$	$PM_{2.5\ predicted}$	Bias	%MRE	R^2
Overall						
2000-2008	53,035	11.24	10.85	0.39	3.5	0.83
Year						
2000	5,567	11.81	11.41	0.40	3.4	0.73
2001	5,876	12.42	11.97	0.45	3.6	0.81
2002	6,606	11.86	11.29	0.57	4.8	0.85
2003	5,878	11.70	11.43	0.27	2.3	0.82
2004	5,977	11.17	10.80	0.37	3.3	0.82
2005	5,153	11.60	11.14	0.45	3.9	0.81
2006	5,491	10.22	9.88	0.34	3.3	0.87
2007	6,142	10.50	10.16	0.33	3.2	0.87
2008	6,345	9.96	9.65	0.31	3.1	0.81
Season						
Winter	12,478	12.18	11.57	0.61	5.0	0.75
Spring	13,349	9.33	9.13	0.20	2.1	0.78
Summer	13,648	13.69	13.21	0.48	3.5	0.87
Fall	13,560	9.80	9.52	0.28	2.9	0.81

Environ Res. Author manuscript; available in PMC 2013 October 01.