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Quantifying the Airshed for Fine Particles: Using Geographic Information System (GIS), Remote Sensing Data, and *In Situ* Monitoring

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ABSTRACT *The advances in remote sensing and the need for air pollution data in remote areas highlighted the need to examine the current capabilities to fill gaps in ambient air monitoring. This paper describes delineating the boundaries of fine particulate matter ($PM_{2.5}$) nonattainment areas by integrating PM ambient data and satellite aerosol optical depth (AOD) with geostatistical kriging. Correlation between the ground and satellites for the three year average was adequate in the upper Midwest and northeast (greater than r = .57) and typically very poor in the rest of the country (less than r = .20). To improve the geospatial prediction of the extent of aerosols, satellite AOD data from the MODerate-resolution Imaging Spectroradiometer (MODIS) sensor was used. This addition of daily air quality data aids prediction of air pollution extent, making the approximation of fine particle airsheds possible.*

Introduction

According to the United States Environmental Protection Agency (US EPA), six million tons of fine particles (PM2.5), 15 million tons of sulfur dioxide (SO_2), and 21 million tons of nitrogen oxides (NOx) are directly emitted into the atmosphere each year in the United States (US EPA, 2008). These particles and gases can be generated locally and transported for days over a vast distance (Prospero & Lamb, 2003; Grousset *et al.,* 2003; Toon, 2003). These emissions have resulted in levels of air pollutants in many parts of the country that exceed national ambient air quality standards (NAAQS) (US EPA, 2005a).

There is abundant epidemiological evidence that air pollution is significantly associated with increased mortality and morbidity (Cohen *et al.,* 2005; Ostro *et al.,* 2008). The associations are especially strong for fine particles $PM_{2.5}$ (less than or equal to 2.5 μm) (Peng *et al.,* 2008; Lippmann & Ito, 2002; Kuenzli *et al.,* 2004; Schwartz, Dockery, & Neas, 1996; Schwartz, 2003; Wilson & Suh, 1997). The association between an increase in mortality as a consequence of severe pollution events has been known since the 1950s (Bell *et al.,* 2004; Ciocco & Thompson, 1961; Logan & Glas, 1952). The EPA first set an

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annual particulate (less than 40 μm) standard of 75 μg/m^3 in 1971 and subsequently tightened the standard in 1987 to smaller (less than or equal to 10 μm) PM_{10} at 50 μg/m^3, and a 24-hour standard was set to 150 μg/m^3. Results of long-term studies had been published (Pope *et al.*, 2002, 2004; Krewski *et al.*, 2000; US EPA, 2006), and in 1997 the EPA adopted a fine PM standard (US EPA, 1997). Regulations prescribe measurements of the total mass of $PM_{2.5}$ (per cubic metre) taken at three-day intervals at 1100 permanent ground-based monitoring stations in the US (Figure 1). Exceedances of the EPA annual $PM_{2.5}$ standard (15.0μg/m^3) are assessed from the data at each monitoring station on a rolling average over three years. Nonattainment designations are defined by the Clean Air Act (CAA) as an 'area that does not meet (or contributes to ambient air quality in a nearby area that does not meet) the national primary[2] or secondary[3] ambient air quality standard for that pollutant' (CAA, 1990).

Satellite sensors such as the Moderate Resolution Imaging Spectroradiometer (MODIS), have been used to estimate aerosol optical depth (AOD) offering the advantage of giving meaningful air quality values almost daily, in heretofore unmonitored areas. Studies have shown a relationship between AOD and fine particulate mass on a local level during the high PM season, which varies geographically in the United States: summer in the southeast and winter in the north and west (Hutchison, 2003; Wang & Christopher, 2003). Rather than identifying separate urban centres, a multi-state airshed approach is needed to identify the spatial distribution of $PM_{2.5}$. The 48 US states were divided into seven geographic regions (Samet *et al.*, 2000).[4]

The use of Geographic Information System (GIS) mapping with MODIS data has been used for years to evaluate the spatial and temporal extent of environmental problems (McCarthy *et al.*, 2007; Sohrabinia & Khorshiddoust, 2007; Mesev, 1999; Luscombe & Hardy, 1988). National Atmospheric and Space Administration (NASA) now offers daily georeferenced images that have been used by land managers in recent disasters such as Hurricane Katrina and western wildfires (NASA, 2008). The methodology developed in this study takes advantage of local accuracy of the Federal Reference Method (FRM) monitors and the wide spatial availability from the satellite data.

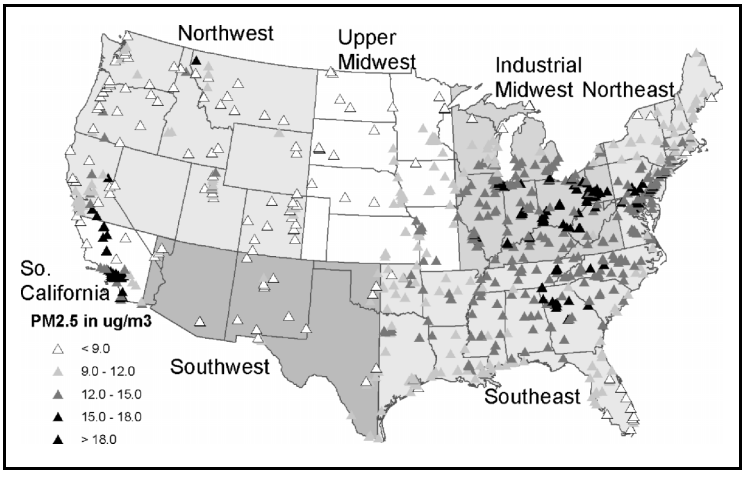

Figure 1. $PM_{2.5}$ monitor values for average of years 2000–03 with PM regions.

Methods

Analysis began with two sets of measurements from the years 2001–03; $PM_{2.5}$ mass concentration from the EPA and MODIS-derived aerosol optical depth (AOD) from NASA. This three-year period was chosen because at the time of this writing, it was the most current quality assured data and was being used for non-attainment purposes for the latest $PM_{2.5}$ standard. The processing of the data is described in detail elsewhere (US EPA, 2005c; NASA, 2005; Remer *et al*., 2001). The PM data for each monitor was averaged by season and annually. This approach differs from the data handling requirements of the Clean Air Act (CAA) Part 50 Appendix N (US EPA, 2002b), requiring quarterly means. The following method also differs from a strict interpretation of the Clean Air Act requirements for designating nonattainment areas (CAA, 1990), which allow for rare exceedances based on exceptional events such as dust storms or wildfires (US EPA, 2005b). In this study, to evaluate the seasonal air quality patterns, all data values were included in the calculations. The Aerosol Optical Depth (AOD) is processed by NASA into the five Atmosphere Products. The one evaluated in this study is the Level 3 Aerosol Product derived at .47 and .66μm (averaged wavelength of .55μm) at 10km resolution. NASA's dark target method uses reflectance measured at 2.1μm to estimate surface reflectance in the visible range, since most aerosols (especially fine-mode dominated) are transparent at that wavelength (Chu *et al.*, 2003). An 'urban/industrial' algorithm is used in the US east of 100° longitude, and a 'smoke' algorithm in the west, based on the differences in aerosol scattering properties (Kaufman & Tanre, 1998; Chu *et al.*, 2002). This global product retrieves values at 10×10 km^2 grid ($1° \times 1°$) which yields 815 evenly spaced points across the US. There are obstacles to overcome with this nascent technology; cloud cover will mask an aerosol measurement, as do highly reflective surfaces such as deserts and ocean sunglint. Therefore, on cloudy days, complete coverage of AOD values were not obtained. Spatial averaging partially overcomes these data gaps, as does seasonal and annual averages.

Spatial interpolation is the estimation of a value at a location that falls between stations with observed values. Using this method, it is possible to take air samples and create a continuous surface of values throughout the study area where monitors are not available. The output includes the uncertainty of these estimations. An increasingly common method in the field of spatial statistics, or geostatistics, is the use of kriging (Goovaerts, 1997) which forms weights from surrounding measured values to predict values at unmeasured locations from a semivariogram that incorporates the spatial structure of the data (Johnson *et al*., 2001). These methods have been explored to enhance exposure assessment (Wong *et al*., 2004) and to evaluate spatial variability (Cressie *et al.*, 1999). Ordinary kriging assumes that the phenomenon influencing the known values are statistically homogenous throughout the region being mapped and assumes an unknown constant mean, (μ); the equation is

$$Z(s) = \mu + \varepsilon(s),$$

where (s) is a location and Z(s) is the value at that location.

Cross validation then removes one value at a time from the kriging—and predicts back to that measured location—giving a predicted and measured value and their difference, as a prediction error (Krivoruchko & Gotway, 2004). Kriging was used to bring the satellite data together spatially and temporally with the ground monitor data (Kyriakidis, 2004) using the ArcGIS® product *Geostatistical Analyst* from ESRI shown in (Figure 2).

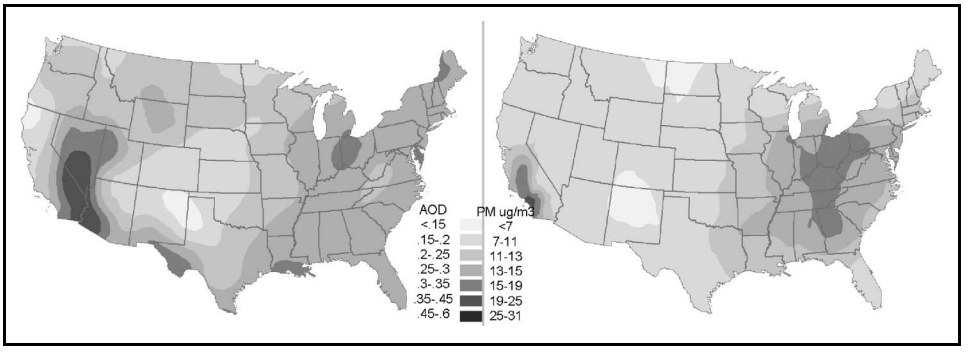

Figure 2. Kriged 2001 annual AOD and fine PM.

Since ground-based $PM_{2.5}$ data is not collected daily, the data was averaged from both satellite and ground-based monitors by month, season, and annually to analyse for their correlation. The AOD surface was then interpolated to the approximately 900 FRM sites that had complete data to obtain the co-located values for the correlation analysis (see Figure 3). Eastern and western regions were separated using 100° longitude as the boundary, and the US was further split into the PM analysis zones, as defined by the NMMAPS study and illustrated in (Figure 1). A correlation analysis was performed for each monthly pair. These kriged AOD surfaces for each region were then matched to FRM monitoring sites by kriging the PM data. Figure 2 shows the differences of kriging AOD (r = 0.45) and kriging $PM_{2.5}$ (r = 0.34) as the surface upon which the other variable was estimated.

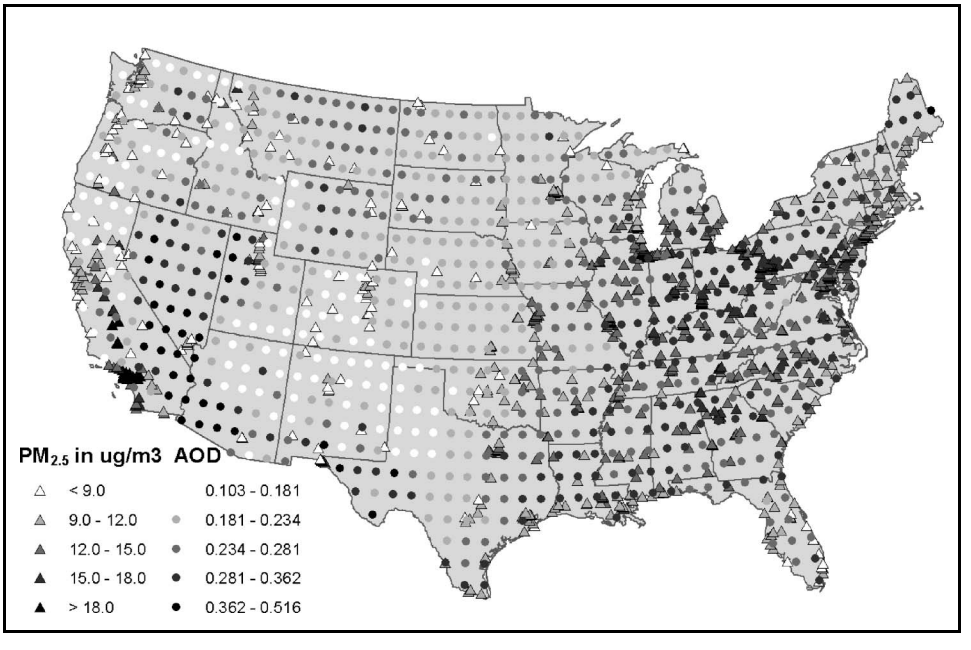

Figure 3. Cokriging variables: AOD with PM.

Cokriging is the process of modelling two or more variables simultaneously and is especially useful when the data have a cross-correlation but are not spatially co-located. An empirical variogram is constructed for both variables, and a cross-variogram model is used to describe the co-variation between the pairs of variables (Johnson *et al.*, 2001). It was assumed that this would generate more optimal spatial predictions by minimizing kriging errors of both AOD and $PM_{2.5}$. The three-year averaged AOD and $PM_{2.5}$ values were used.

Results

Initially, the averaged data from the 2001 FRM monitoring network were compared to that of the MODIS data. This comparison was done to assess the use of the aerosol product quantitatively to predict fine PM. For 2001, there were data from slightly fewer than 900 monitors, and 815 data points for the AOD, depending on daily data availability. The network of FRM monitors were used because it is the data used by the EPA in trend analysis, attainment demonstration, and designation policy. Although continuous monitors are used by states for reporting the Air Quality Index (AQI) (US EPA, 2002a), they are not recognized by the EPA for nonattainment purposes (US EPA, 1998a, 2000).

Table 1 lists Pearson correlation coefficients (r) for both $PM_{2.5}$ values predicted to the AOD points and the AOD values predicted to the FRM monitor sites (see Figure 4). All values had a P value less than .05, and were considered statistically significant, except the April PM to AOD correlation. Spring is typically a season with poor correlations between PM and AOD, and April was exceptionally uncorrelated. Melting snow/ice is believed to impair AOD retrievals and consequently produce such artifacts.

Table 1. Pearson correlation coefficients of $PM_{2.5}$ FRM and MODIS AOD

2001	Kriged PM to Measured AOD	Kriged AOD to Measured PM
Averages for the entire contiguous US		
Annual	.34	.457
January	.23	.33
February	.14	.14
March	.12	.18
April	.002*[+]	.06
May	.34	.50
June	.44	.64
July	.62	.68
August	.68	.80
September	.41	.44
October	.28	.28
November	.29	.14
December	.35	.38
Fall	.37	.30
Winter	.23	.26
Spring	.03	.13
Summer	.61	.77

Notes: * Indicates the P value is > .05 (little evidence of correlation).
[+] P > .05.

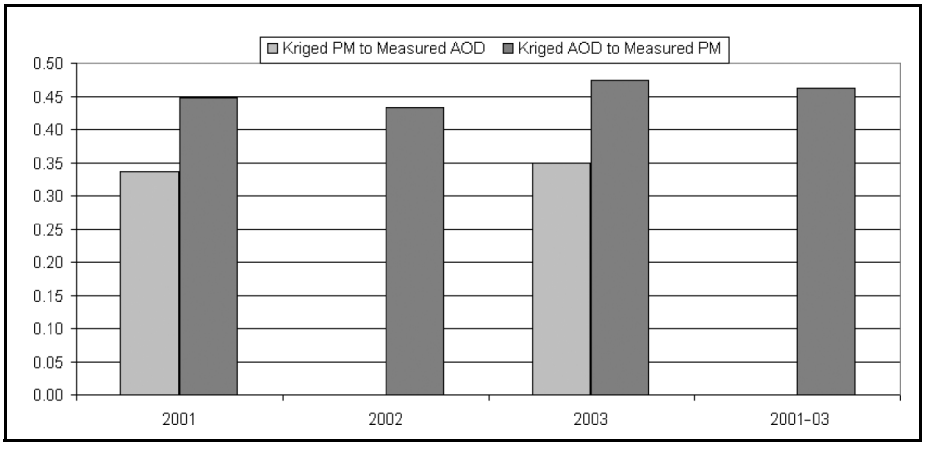

Figure 4. Correlations for 2001, 2002, 2003, and 2001–03.

Table 2. 2001 seasonal and regional correlation coefficients

2001	Kriged PM to Measured AOD	Kriged AOD to Measured PM
Seasonal divided at 100° W longitude		
East Fall	.51	.21
East Winter	.17	.40
East Spring	–.013*	–.04*
East Summer	.81	.68
West Fall	.24	.35
West Winter	.27	.11
West Spring	.16	.16
West Summer	.27	.42
Seasonal by PM regions		
IndMW Fall	.46	.21
IndMW Winter	.25	.55
IndMW Spring	.25	.06*
IndMW Summer	.88	.74
NE Fall	.59	.45
NE Winter	–.24*	–.14*
NE Spring	–.42	–.59
NE Summer	.84	.80
SE Fall	–.36	–.32
SE Winter	–.45	–.37
SE Spring	–.05*	–.09*
SE Summer	.69	.63

Notes: * Indicates the P value is >.05 (little evidence of correlation).

Analysis of the tabular data indicates that there are months and seasons of varying correlations (Table 2), with the summer showing correlations nearly double the value than that of any other season and the spring especially uncorrelated (see Figure 5). This exceptional seasonal

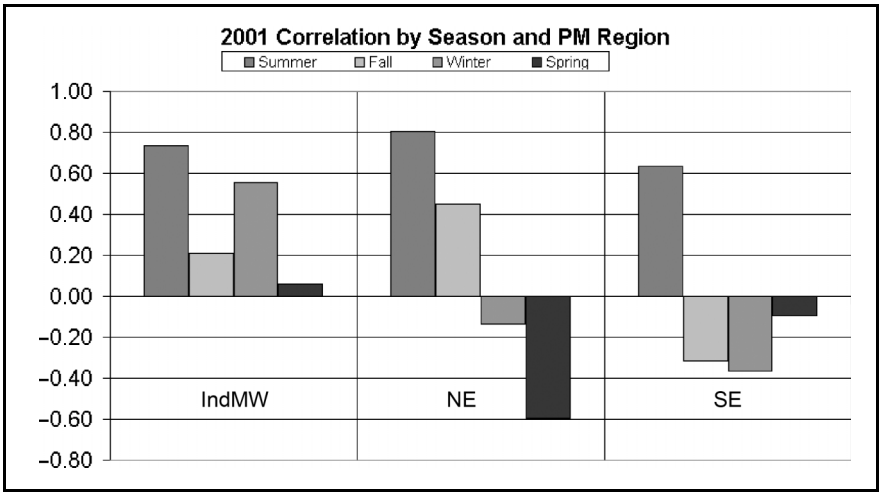

Figure 5. 2001 seasonal correlations of AOD to PM by region.

difference casts doubt on the significance of the annual means. These results reflect the analysis of the entire contiguous 48 United States and a further analysis divided this area into smaller regions (see Figure 6). The eastern half of the US has relatively better monitor coverage and the errors are smaller. Better correlations have been noted in the eastern as opposed to the western US and smaller kriging errors from this analysis suggest that underlying monitor proximity could explain the lower correlation.

The PM and AOD datasets were combined by cokriging to optimize the prediction in the study areas. Given that the AOD surface was the most structured in space and the prediction errors were minimal, Goovaerts recommended the use of AOD as the primary

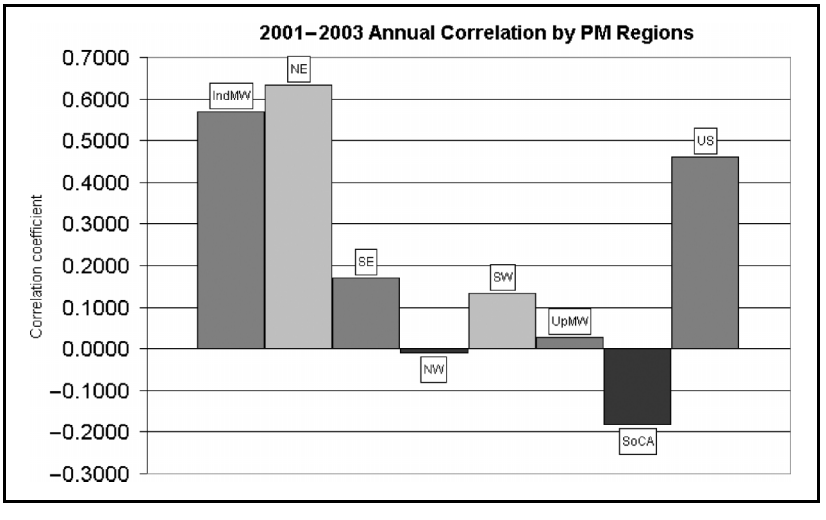

Figure 6. Annual correlations of AOD to PM by region (2001–03).

Table 3. Correlation coefficients for three-year average of kriged AOD

2001–2003	PM to AOD	AOD to PM
US Annual	.34	.46

dataset for interpolation and $PM_{2.5}$ as the covariate (Personal Communication, 20 July 2004). The correlations for the combined 2001–03 averaged data are listed in Table 3. With each surface kriged separately, a closer correlation between AOD and $PM_{2.5}$ occurred with the AOD as a base surface onto which the PM was added.

Discussion

Previous studies of aerosol correlation (Wang & Christopher, 2003; Hutchison, 2003) have depended on the use of proximity measures. In these analyses, only the AOD data collected within a short time and distance of a ground PM measurement were used to analyse possible correlations. This limits the implications of their study to geographical areas around the monitors and does not address areas at a distance from monitors.

Although not remarkable, nor consistent throughout the study period or the entire study area, correlations did exist in the analysis; however, as seen in Figures 7 and 8, it was not found that correlations were achieved throughout the year there is a distinct seasonal trend with the highest correlations found during the summer (Figure 7).

The summer season is unique in several ways that may affect AOD and $PM_{2.5}$ correlation. First, the summer has—on average—the highest aerosol concentrations for both measures (Figure 7); and second, summer typically has the highest mixing height (Hobbs, 2000; Lutgens & Tarbuck, 2004). The higher mixing height, or the altitude which defines the boundary allowing the turbulent mixing of atmospheric constituents, allows the aerosols to fully mix, giving similar values on the ground as throughout boundary layer. The AOD is a total column measurement from over 700km in space. Events occur that push the aerosols above the level that the FRM monitors are able to detect. Others have observed similar seasonal trends (Engel-Cox *et al.*, 2004; Wang & Christopher, 2003).

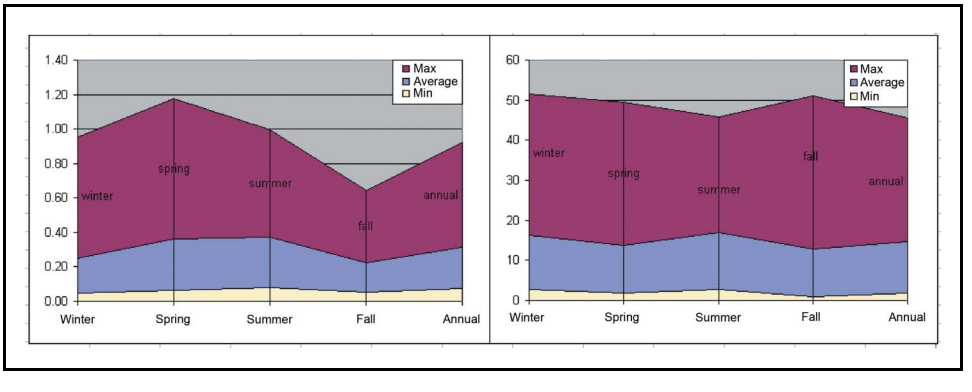

Figure 7. AOD values for 2001 (left) and PM values in µg/m3 (right).

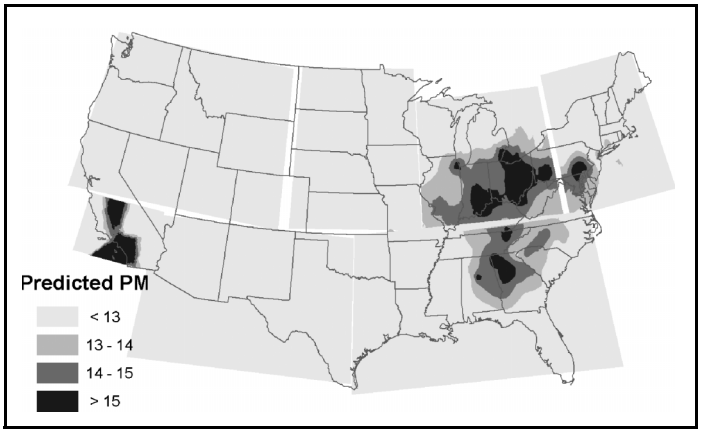

Figure 8. Cokriging 'prediction map' of PM with AOD (2001–03) by PM region, and here placed as a mosaic (the gaps indicate those regional boundaries).

Spring correlations are especially poor, followed by winter. These seasons are characterized by the lack of leaf coverage, highly reflective snow, and areas of melting snow. MODIS AOD algorithms are especially well-suited for dark targets such as forested areas; and the same algorithms have an especially hard time distinguishing melting snow from elevated aerosol optical depth (Remer *et al*., 2001).

East–west differences in the properties of the fine particles were explored (the sulfate fraction is predominant in the east, and the carbon fraction in the west) (Rao *et al.,* 2002; Pinto *et al*., 2004), but the effects on correlation were inconclusive. The fact that $PM_{2.5}$ data are collected once every three days and AOD is masked on cloudy days due to invalid estimates add an additional hindrance to evaluating a large amount of potentially crucial data values. Two-thirds of the PM mass data are essentially missing from the study by design and an ever-changing number of days of AOD data by the weather.

From ground-based sun photometers, MODIS AOD errors were estimated by Kaufman and Tanre (1998). Random errors may include: instrument calibration (2% to 5%); uncertainty in the surface reflectance (5% to 20%); screening, including removal of pixels using the filter (0% to 10%); and from uncertainty in the use of the models (10% to 20%). Errors due to increased reflectance from the surface will affect the AOD data differently in different seasons of the year, leading to greater error and lower correlation to $PM_{2.5}$ in the winter and spring. The $PM_{2.5}$ FRM values are also subject to calibration error from instruments (US EPA, 1998b).

Given the present analysis, neither an annual average AOD, nor a three-year average, would allow for the adequate annual prediction of fine PM mass over the complete study area. The analysis of three years of monthly data may be sufficient to make assumptions with regard to seasonal or geographical anomalies, but not to completely address the correlation anomalies.

Figure 8 illustrates a prediction map using ordinary cokriging showing the predicted area exceeding the current 15 $\mu g/m^3$ annual standard. This is compared to the official EPA $PM_{2.5}$ nonattainment designations map (Figure 9) (US EPA, 2004). Both maps are using

Figure 9. Nonattainment boundaries (2001–03) by the EPA (US EPA, 2005a).

identical years (2001–03) and illustrate similarity in 'hot spots'. The map in Figure 9 identifies only the counties that have elevated monitored $PM_{2.5}$ data and gaps will be apparent between those monitored counties. The shaded areas in Figure 8 indicate that the pollutant concentration within those shaded boundaries are estimated to be above the labeled concentration. The darkest shading predicts the area that would average at least 15 $\mu g/m^3$ annually, and the other lighter shades indicate the area of potential nonattainment under a more conservative 14 and 13 $\mu g/m^3$ annual standard.

The improved spatial resolution of the aerosol satellite data could be helpful to fill gaps in the ground-based monitoring of rural areas. The boundaries of airsheds were shown to be highly variable in this study because of the seasonally changing locations of the fine particle air masses. But, as shown in Figure 8, specific regions of the country consistently have elevated aerosols. Adding the data from the *Aqua* satellite would provide an afternoon measurement of AOD; together with the *Terra* data, these twice-daily collections would give a more complete coverage to the analysis (Ichoku *et al.*, 2005). Finally, to address the limitation of the disparity of measurement height, another remote sensing instrument would be beneficial: the ground-based and satellite mounted LIDAR (light detection and ranging). The LIDAR emits a laser signal and retrieves the scattering caused by the column of aerosol to give a vertical profile of aerosols. Recent studies have shown promise in capturing this disparity in particle measures by visualizing aerosols aloft (Landulfo *et al.*, 2005; Engel-Cox *et al.*, 2006).

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Notes

1. The views expressed in the article are those of the author and do not necessarily represent the views of the EPA or of the United States.
2. Primary standards protect human health.
3. Secondary standards protect 'welfare', including ecological effects and materials damage.
4. Some analysis results are summarized at a broad regional level using the geographic regions specified below. The regional definitions correspond to the regions identified by the Health Effects Institute (HEI) in a recent PM study. (See Figure 1, page 8, in Samet *et al.*, 'The National Morbidity, Mortality, and Air Pollution Study Part II: Morbidity, Mortality, and Air Pollution in the United States', Health Effects Institute, Research Report Number 94, Part II, June 2000.) The origin of the HEI region definitions can be traced back to Figure 6-30 of the EPA's 1996 PM Criteria Document, which identified regions on the basis of 'uniqueness in aerosol trends, seasonality, size distribution, or chemical composition'.

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