

Yang Liu^{*}
Harvard University, Boston, MA

1. Introduction

After numerous epidemiological studies started as early as in the 1970s, scientists have quantitatively established that exposure to ambient particulate matters (PM) is associated with morbidity and mortality (Pope 2000; Wallace 2000). In particular, exposure to PM_{2.5} (particles with diameters less than 2.5 μm) is associated with illness and premature death independent of the effects of other, gaseous pollutants in the atmosphere (Schwartz et al. 1999). The importance of long-term PM_{2.5} monitoring has recently been emphasized since studies have relationship between ambient PM_{2.5} concentrations and health with no clear threshold (Smith and Jantunen 2002; Schwartz et al. 2002).

Evaluation of chronic population exposures over a large geographical region relies on long-term monitoring data from a comprehensive network such as the United States Environmental Protection Agency's (EPA) compliance network. However, operating and maintaining such networks are costly. Air quality models can be used to estimate PM_{2.5} concentrations where ground monitoring is absent. Due to various reasons such as the lack of accurate emissions inventory, limited computing resources, and model parameterization, daily PM_{2.5} concentrations predicted by these models may be biased.

With the rapid development of satellite remote sensing technology in recent years, aerosol optical properties retrieved by spaceborne sensors have emerged as another potential method of monitoring ground level air quality since these data products provide nearly global coverage at moderate spatial resolution for multiple years. This paper reports the findings from two studies using the aerosol optical thickness (AOT) retrieved by the Multi-angle Imaging SpectroRadiometer (MISR) as an indicator of the abundance of ambient PM_{2.5}.

2. Study I: the Development of a Regression Model to Estimate 24-hr PM_{2.5} Concentrations in Eastern U.S.

In this study, we develop an empirical regression model using MISR AOT as the major predictor to estimate ambient 24-hr PM_{2.5} concentration. Our objective is to explore the possibility of MISR AOT as an effective indicator of ground level particle pollution.

2.1 Description of Data and Method

Three major data sources are used in this study. A total of 2,505 24-hr PM_{2.5} measurements are collected from 346 sites within the EPA's compliance network in the eastern United States are collected from the year 2001 (Figure 1). This dataset is divided into a model dataset and a validation dataset according to site ID numbers randomly assigned by EPA. The study region is divided into three sub-regions, i.e., the New England, the Mid Atlantic, and South Atlantic region in order to examine geographic variability among the observed results.

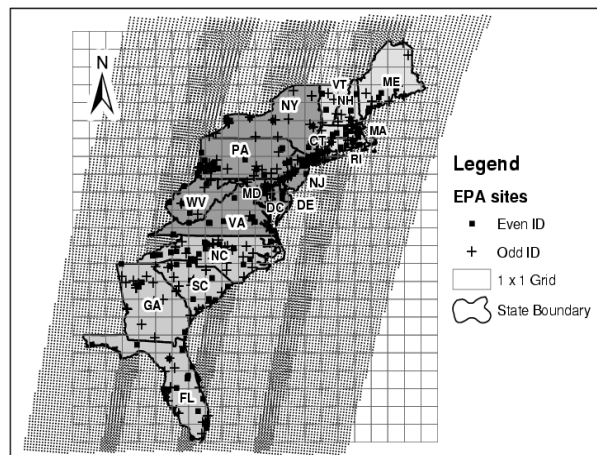


Figure 1. MISR spatial coverage of the study region. The ground tracks of MISR paths are shown as dotted strips in the map. Only the ground tracks of MISR path 11, 14, 17, and 20 are shown for clarity of the map. The three sub-regions are marked in different grey colors. The study area is also divided into 1°×1° grids.

^{*} Corresponding author address: Yang Liu; email: yliu@deas.harvard.edu

MISR AOT data (Diner et al. 1998) covering the east coast is downloaded from Atmospheric Sciences Data Center at NASA Langley Research Center (<http://edg.larc.nasa.gov/>). The spatial resolution of MISR AOT is 17.6 km. To reduce the impact of potential outliers in AOT data due to MISR retrieval errors, valid AOT values are averaged within each 50 x 50 km region (3 x 3 MISR pixels).

To evaluate the impact of aerosol vertical profile and particle growth effect under high relative humidity conditions, the mean relative humidity (RH, in %) within the lower troposphere and planetary boundary layer height (PBL, in km) are extracted from Goddard Earth Observing System (GEOS-3).

2.2 Model Development

The empirical regression model used in the current analysis can be expressed as:

$$\ln(PM_{2.5}) = \beta_0 + \beta_1(\text{Variable}_1) + \dots + \beta_n(\text{Variable}_n) + \beta_{RH}(RH) + \beta_{AOT}\ln(AOT) + \beta_{PBL}\ln(PBL)$$

The dependent variable on the left hand side, $PM_{2.5}$, is the 24-hour average ground level $PM_{2.5}$ concentrations measured at various EPA monitoring sites in 2001. The independent variables on the right hand side include relative humidity (RH), aerosol optical thickness (AOT), and height of the planetary boundary layer (PBL) that are geographically matched to each $PM_{2.5}$ measurement, as well as various categorical variables (Variable_1 through Variable_n) listed in Table 1. β_0 through β_n are regression coefficients for Variable_1 through Variable_n . Likewise, β_{RH} , β_{AOT} , and β_{PBL} are regression coefficients for RH, AOT, and PBL, respectively. An exponential function of RH is used to account for the super-linear growth of particle size with increasing relative humidity (Malm et al. 2000; Chin et al. 2002). This model form provides stronger physical background, more flexibility and predicting power as compared to a previous study, where a simple linear regression model is fit between AOT and $PM_{2.5}$ concentrations (Engel-Cox et al. 2004).

2.3 Regression Analysis

The empirical model described in previous section is fitted using the model dataset. Overall, the model results are highly significant ($p < 0.0001$) explaining 43% of the variability in corresponding ground level $PM_{2.5}$ concentrations. AOT, PBL, RH, as well as all the categorical

variables listed in Table 1 are found to be highly significant predictors of $PM_{2.5}$ ($p < 0.0001$). Concentration impact factors (CI factors) for the categorical variables are calculated as the exponentials of the parameter estimates. A CI factor can be interpreted as the impact of a categorical variable at certain level to the association between MISR AOT and $PM_{2.5}$ as compared to the reference level of this factor.

Table 1. Definition of all categorical variables used in estimating $PM_{2.5}$ concentration with MISR AOT and mixing height.

Variable	Level
Region	New England
	Mid Atlantic
	South Atlantic
Season	Winter (Dec - Feb)
	Spring (Mar - May)
	Summer (Jun - Aug)
	Fall (Sep - Nov)
Site location	Rural
	Suburban
	Urban
Distance from Coast	Within 100 km from coast
	Beyond 100 km from coast

Table 2. Estimated regression coefficients. N = 1315. $R^2 = 0.43$.

Model Variables	Estimates	Std Err	P value	CI factor
Intercept	3.891	0.102	< 0.0001	48.97
Season				
Winter	0.048	0.031	0.12	1.05
Spring	-0.296	0.036	< 0.0001	0.74
Summer	0.009	0.038	0.80	1.01
Fall	0.000			1.00
Region				
New England	-0.157	0.038	< 0.0001	0.85
Mid Atlantic	0.005	0.027	0.84	1.01
South Atlantic	0.000			1.00
Distance from coast				
≤ 100 km	-0.193	0.028	< 0.0001	0.82
> 100 km	0.000			1.00
Site location				
Rural	-0.296	0.047	< 0.0001	0.74
Suburban	-0.083	0.025	0.001	0.92
Urban	0.000			1.00
RH	-0.634	0.115	< 0.0001	
Ln(AOT)	0.447	0.022	< 0.0001	
Ln(PBL)	-0.361	0.023	< 0.0001	

* Reference level of a given categorical variable.

The estimated power of AOT (0.447 ± 0.022) is less than one, indicating $PM_{2.5}$ concentrations varied sub-linearly with AOT measurements. The

greater variability of the observed AOT values is likely due to the fact that AOT measures particle abundance within the entire atmospheric column. Both photochemical reactions, which occur mainly within the boundary layer, and the long-range transport of particles, which occurs in the free troposphere, can have substantial impacts on AOT values. As a result, AOT measurements exhibits a greater variability as compared to ground level $PM_{2.5}$ concentrations, which is less influenced by long-range transport of particles.

The significance of PBL in predicting surface $PM_{2.5}$ concentrations reflects the difference in particle vertical profile within and above the boundary layer. Fine particles tend to be more homogeneous within the boundary layer due to convective mixing as compared to particles in the free troposphere. Fine particles emitted from the surface are diluted within the boundary layer as PBL height increases resulting a lower $PM_{2.5}$ concentration. This may explain the negative sign of the estimated power on boundary layer height (-0.361 ± 0.023). In addition, the observation that the magnitude of the parameter estimate is substantially smaller than one indicates that surface $PM_{2.5}$ concentrations vary at a slower rate as compared to boundary layer height.

The negative parameter estimate of the exponential of RH (-0.634 ± 0.115) indicated that same AOT values correspond to lower $PM_{2.5}$ concentrations as RH increases. This result shows that the model provides a correction for the humidification effect on particle light extinction. MISR measures AOT at ambient meteorological conditions. Under high relative humidity (RH > 70%), which is observed frequently in this study, hygroscopic particles such as ammonium sulfate and ammonium nitrate can grow 2 – 10 times in size resulting dramatic increase of their light extinction efficiencies (Chin et al. 2002). In contrast, $PM_{2.5}$ concentrations are measured under controlled lower RH conditions (at 40% RH). As a result, same AOT values at high RH levels will correspond to less particle dry mass as compared to low RH conditions. It should be noted that since actual particle size does not grow strictly exponentially and depends on particle composition. Therefore, the exponential form of RH is a simplified representation of particle growth effect based on the regression statistics.

The impact of particle size, composition and vertical distribution on the association between MISR AOT and $PM_{2.5}$ concentrations is also reflected by the categorical variables. Possible interpretations to the impact of these variables are

given below. The effect of season is highly significant ($p < 0.0001$), with the association between $PM_{2.5}$ and AOT found to be significantly weaker in the spring as compared other seasons. The CI factor of 0.74 for the spring indicates that predicted $PM_{2.5}$ concentration is 26% lower in the spring than in the fall with all other parameters being equal. This could be because particle vertical distribution is generally different in the spring from other seasons. Asian dust events in the spring can significantly increase the particle concentrations in the free troposphere (Thulasiraman et al. 2002), resulting a larger proportion of particles above the boundary layer as compared to other seasons. Because the particle mass loading below the boundary layer determines surface $PM_{2.5}$ concentration, similar AOT levels will predict lower surface $PM_{2.5}$ concentration in the spring as compared to other seasons.

The CI factor of 0.85 for geographical region suggests that MISR predicts lower $PM_{2.5}$ levels in the New England region as compared to the other regions. $PM_{2.5}$ concentrations in New England region are heavily impacted by transported pollutants from distant industrial and urban sources in the Mid Atlantic and mid-western U.S. as well as southern Canada (Slater et al. 2002). As a result, more sulfate particles from transported precursors such as SO_2 and less carbonaceous particles may be found in particle mixtures throughout New England. Sulfate particles generally have higher light extinction efficiencies than carbonaceous particles, especially under high relative humidity conditions (Chin et al. 2002). Consequently, under the same meteorological conditions, lower particle concentrations in New England region will be needed to achieve the same AOT level in the other two regions.

Other variables being equal, MISR predicts lower $PM_{2.5}$ concentrations for rural sites (CI factor = 0.74) as compared to those at suburban (CI factor = 0.92) and urban sites (reference state, CI factor = 1.0). Urban sites are generally characterized by greater anthropogenic $PM_{2.5}$ emission sources than suburban or rural sites. Therefore, a larger proportion of particle mass at urban sites is nitrate and carbonaceous particles, generated from mobile source emissions as compared to rural sites (Kleeman et al. 2000). As previously mentioned, sulfate particles have higher light extinction efficiencies than carbonaceous particles. Consequently, a larger amount of particle mass is needed to achieve the same AOT level in urban areas as in rural areas. Particle

composition in suburban sites may be influenced by both long-range transport and local emissions. Therefore, a slight correction effect is noted.

Other variables being equal, MISR also predicts lower $PM_{2.5}$ concentrations at coastal sites (CI factor = 0.82) as compared to inland sites. Previous research has shown that the warm conveyor belts (i.e., moist air streams that rise ahead of surface cold fronts), which can lift ground level pollutants to upper troposphere and then transport them over the continents, most frequently originate in the boundary layer of the eastern seaboard of North America and Asia, close to the heavy anthropogenic emissions (Stohl et al. 2001). Therefore, it is possible that a larger proportion of particles reside at higher altitude in the coastal region as compared to the inland region in this study. Similar levels of AOT will correspond to lower surface $PM_{2.5}$ concentrations at coastal sites, therefore, than at inland sites given the fact that the particle mass loading below the PBL determines surface $PM_{2.5}$ concentration.

2.4 Model Validation

In order to evaluate model performance, the regression model developed in the previous section is applied to the validation dataset to generate predicted $PM_{2.5}$ concentrations. On average, predicted $PM_{2.5}$ concentrations are $1.2 \mu\text{g}/\text{m}^3$ lower than observations. Differences decreased to $0.4 \mu\text{g}/\text{m}^3$ when $PM_{2.5}$ concentrations greater than $40 \mu\text{g}/\text{m}^3$ are removed. A linear regression between predicted and observed $PM_{2.5}$ concentrations yielded an R^2 of 0.48 (Figure 2). The model Root Mean Square Error (RMSE) is $\pm 6.2 \mu\text{g}/\text{m}^3$ for a mean $PM_{2.5}$ concentration of $13.8 \mu\text{g}/\text{m}^3$. Model substantially underestimates $PM_{2.5}$ concentrations at higher concentrations ($> 40 \mu\text{g}/\text{m}^3$). This could be because over 98% of $PM_{2.5}$ concentrations are below $40 \mu\text{g}/\text{m}^3$ in the modeling dataset. Therefore, current parameter estimates do not sufficiently represent the association between $PM_{2.5}$ and the independent variables at higher $PM_{2.5}$ concentrations. In addition, higher daily-average $PM_{2.5}$ concentrations are often strongly influenced by pollution episodes that occur during a short period of the day. The impact of these episodes may not be captured within the MISR measurement time window (10 – 11 a.m. local time). As a result, MISR AOT measurements cannot sufficiently represent the daily-average $PM_{2.5}$ concentrations under such circumstances. The predicted vs. observed regression slope

approached 1.00 and intercepts are insignificant when those high observations are excluded (again, less than 2% of total data). Additionally, the model RMSE is reduced to $5.3 \mu\text{g}/\text{m}^3$ for a mean $PM_{2.5}$ concentration of $13.2 \mu\text{g}/\text{m}^3$ when these high $PM_{2.5}$ concentrations are excluded. Since current air quality models, including Eulerian box models (Pun et al. 2001), Lagrangian plume models (Hudischewskyj et al. 1989), and 3-D Eulerian models (Jacobson et al. 1997; Seigneur et al. 1999; Seigneur et al. 2000), have been shown to agree within 17 – 46% of ground based measurements, the results from our regression model are comparable with these models.

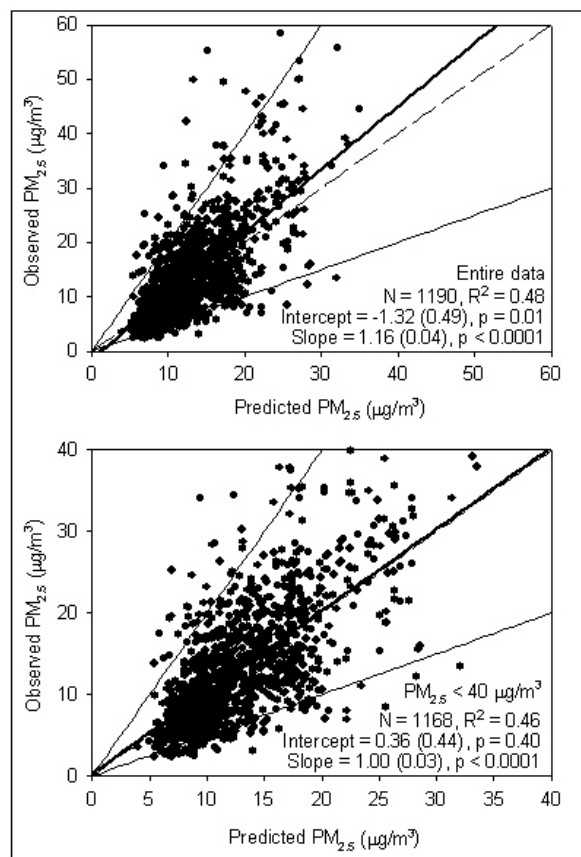


Figure 2. Scatter plots of predicted vs. observed $PM_{2.5}$ mass concentration for the entire validation dataset (upper panel) and for observations less than $40 \text{ mg}/\text{m}^3$. The adjusted R^2 , parameter estimates and p-values of the estimates are shown in each plot. The 1:1 line (dashed) is shown as reference. The regression line is shown as the thick solid line and the upper and lower bounds (factor of 2) are shown as thin solid lines.

3. Study II: Mapping Annual Mean Ground Level PM_{2.5} Concentrations Using MISR AOT Over Contiguous U.S.

In this study, we develop a simple physical model using MISR AOT and aerosol simulation results from a global atmospheric chemistry model (GEOS-CHEM) to estimate annual average PM_{2.5} concentration over the contiguous U.S.. Our objective is to evaluate MISR's capability to serve as a cost-effective extension of ground PM monitoring network.

3.1 Description of Data and Method

Three major data sources are used in this study. As discussed in Section 2, aerosol vertical profile has a significant impact on AOT's predicting power on ambient PM_{2.5}. To treat the spatial and temporal variation of aerosol vertical profile more effectively, aerosol simulation results from the GEOS-CHEM model is used in this study. The fully coupled oxidants-aerosol simulation by GEOS-CHEM provides sulfate (SO₄²⁻), nitrate (NO₃⁻), ammonium (NH₄⁺), elemental carbon (EC), and organic carbon (OC) aerosol concentrations for the period of 2001 at 3-hour temporal resolution, 2° latitude x 2.5° longitude horizontal resolution, and 30 sigma vertical layers. When calculating AOT using aerosol dry mass concentrations, particle growth with increased relative humidity is taken into account by applying different hygroscopic growth factors to all hydrophilic species using local relative humidity conditions. Detailed descriptions of GEOS-CHEM as well as its aerosol simulations can be found elsewhere (Bey et al. 2001; Park et al. 2003). Monthly mean dust and sea salt concentrations for 2001 from the Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) model are used to complement GEOS-CHEM aerosol fields. The 3-hour simulation results are first interpolated to 10 a.m. local time values, sampled on the dates when MISR had valid AOT retrievals, then integrated into annual averages in order to be compared with annual mean MISR AOT values.

Daily average PM_{2.5} concentrations measured by gravimetric methods in 2001 from 1,137 sites of EPA's compliance network, primarily located in urban areas and surrounding suburbs, are collected and integrated into annual averages in each GEOS-CHEM model grid cell (Figure 3). Validated daily average mass concentrations of SO₄²⁻, EC, OC, and mineral dust were collected from 131 sites of EPA's PM_{2.5} speciation trends

network (STN) and integrated into annual averages in model grid in order to analyze the difference between simulated PM_{2.5} concentrations and EPA PM_{2.5} measurements for individual aerosol components.

MISR AOT data that covered the contiguous United States for 2001 were obtained from the NASA LARC Data Center, and then integrated into annual averages in 2°x2.5° model grid cells. The data covering North Dakota, South Dakota and Minnesota in the winter and spring is excluded due to potential cloud contamination. The temporal and spatial variability of MISR AOT errors is partly corrected in this analysis by applying linear regressions between MISR and AERONET AOT values presented in (Liu et al. 2004).

The final dataset consists of totally 159 annual data records, each containing the seasonal average EPA PM_{2.5} measurement, simulated PM_{2.5} concentration and AOT, MISR AOT, and MISR PM_{2.5} concentrations in each GEOS-CHEM model grid cell.

3.2 Coupling of the Global Models with MISR

Although we previously show in Section 2 that our empirical regression model can predict surface PM_{2.5} concentrations with a relative error of approximately 45%, half of the variability in PM_{2.5} concentrations cannot be explained probably due to the lack of information on aerosol vertical profile and long-range aerosol transport events. In addition, empirical models must be calibrated before transferring to other regions. We here use simulated AOT and PM_{2.5} concentrations from GEOS-CHEM to define a physically consistent relationship between AOT and surface level PM_{2.5} concentration:

$$\text{MISR PM}_{2.5} \text{ Concentration} = \frac{\text{Simulated Surface PM}_{2.5} \text{ Concentration}}{\text{Simulated Column AOT}} \times \text{MISR AOT}$$

This relationship is then applied to MISR AOT to infer PM_{2.5} distributions. We refer to the PM_{2.5} concentrations derived from this simple model as MISR PM_{2.5} concentrations hereinafter.

The MISR PM_{2.5} concentrations differ from the simulated PM_{2.5} concentrations in three ways. First, the MISR PM_{2.5} concentrations are less likely to be affected by possible biases in the aerosol vertical distribution estimated by the global models because the biases are attenuated by the ratio of

simulated $PM_{2.5}$ concentrations over simulated AOTs. Second, because MISR AOT has a much higher spatial resolution than the global model simulations, MISR $PM_{2.5}$ concentrations would be able to reflect the impact of sub-grid variation of particle properties. Finally, it has been shown that the discrepancy between gravimetric $PM_{2.5}$ concentrations and the sum of all measured particle components can be as large as 28 – 42% (Andrews et al. 2000). This discrepancy is likely due to uncertainties in organic carbon and dust measurements. These differences between global model predictions and EPA $PM_{2.5}$ measurements are likely to be reduced with the calibration of MISR AOT. It should be noted that this equation assumes that the atmospheric column is dominated by one aerosol component. When two or more important aerosol components with different optical properties and vertical distributions are present, AOT and $PM_{2.5}$ concentrations are likely to have a non-linear relationship. Under such circumstances, MISR $PM_{2.5}$ derived in this study would only be a first-order approximation of EPA $PM_{2.5}$ measurements.

3.3 Results and Discussion

Reduced major axis lines are used to characterize the overall relationship between simulated $PM_{2.5}$ concentrations and EPA $PM_{2.5}$ measurements, as well as the agreement between simulated AOT and MISR AOT retrievals (Hirsch

and Gilroy, 1984). When comparing the MISR $PM_{2.5}$ concentrations with EPA $PM_{2.5}$ concentrations, simple linear regression is used because we are interested in examining the model's capability of estimating individual $PM_{2.5}$ concentration over a given grid cell.

3.3.1 Comparison of Simulated $PM_{2.5}$ with EPA $PM_{2.5}$ Measurements

Figure 3 compares annual average simulated $PM_{2.5}$ and observed $PM_{2.5}$ concentrations in the contiguous United States. The EPA $PM_{2.5}$ measurements are plotted on a $0.5^\circ \times 0.5^\circ$ grid. The annual average simulated $PM_{2.5}$ concentrations capture the geographic characteristics of EPA $PM_{2.5}$ measurements very well nationwide with the exception of the San Joaquin Valley and southern California where the models substantially underestimate $PM_{2.5}$ concentrations. A scatter plot shows that annual average simulated $PM_{2.5}$ concentrations have a good linear relationship with EPA measurements ($r = 0.74$, reduced major axis line slope = 1.04) but with a negative offset of $2.88 \mu\text{g}/\text{m}^3$ (Figure 4). The three data points that apparently deviate from the general pattern of the dataset are all from Southern California. Excluding the three potential outliers does not have a significant impact on the parameter estimates of the reduced major axis line.

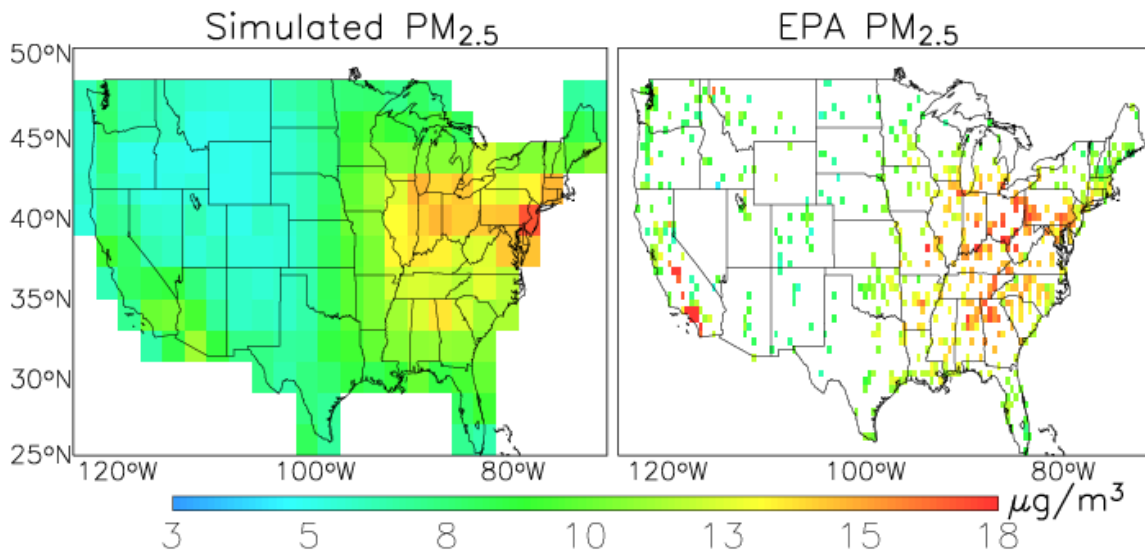


Figure 3. 2-D plot of annual simulated $PM_{2.5}$ concentrations integrated in $2^\circ \times 2.5^\circ$ grid cells (left) vs. EPA $PM_{2.5}$ measurements integrated in $0.5^\circ \times 0.5^\circ$ grid cells (right). The scale saturates at $18 \mu\text{g}/\text{m}^3$ to best display the color contrast in the plot (99th percentile of EPA $PM_{2.5}$ measurement = $17.05 \mu\text{g}/\text{m}^3$).

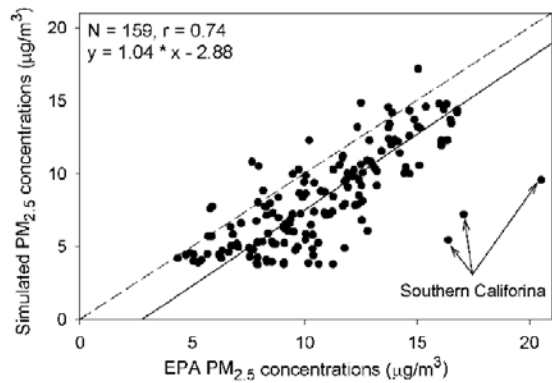


Figure 4. Scatter plot of annual average simulated $PM_{2.5}$ concentration vs. EPA $PM_{2.5}$ measurements. The reduced major axis line is shown as the solid line in the plot. The 1:1 line is shown as the short dashed line for reference. Three potential outliers pointed by arrows are all Southern California grid cells.

The overall underestimation of $PM_{2.5}$ concentrations might be attributed to the discrepancy between chemical and gravimetric measurements found in surface level monitoring campaigns, with the sum of all component concentrations often smaller than the gravimetric measurements of $PM_{2.5}$ concentrations, as previously mentioned.

3.3.2 Comparison Between Simulated and MISR AOT

Annual average simulated AOT generally captures the spatial pattern of MISR AOT measurements, with higher values in the east and lower values in the west (Figure 5). A scatter plot shows that simulated AOT has a good linear relationship ($r = 0.80$) with MISR AOT with a small offset (intercept = - 0.007) although simulated AOT shows a low bias of 17% (reduced major axis slope = 0.83, figure is not shown here). Current MISR AOT data includes the aerosol extinction effect in the entire atmospheric column in both the troposphere and the stratosphere (Charlie Welch, personal communication). Although stratospheric AOT is usually at an order of magnitude smaller than tropospheric AOT (Kent et al. 1994), it likely contributes to the difference of approximately 0.03 between the means of MISR AOT and simulated AOT. In addition, as previously mentioned, the sum of the known particle species concentrations can be significantly smaller than $PM_{2.5}$ concentrations measured by gravimetric methods. This deficit is also likely reflected in the underestimation of AOT by GEOS-CHEM and GOCART.

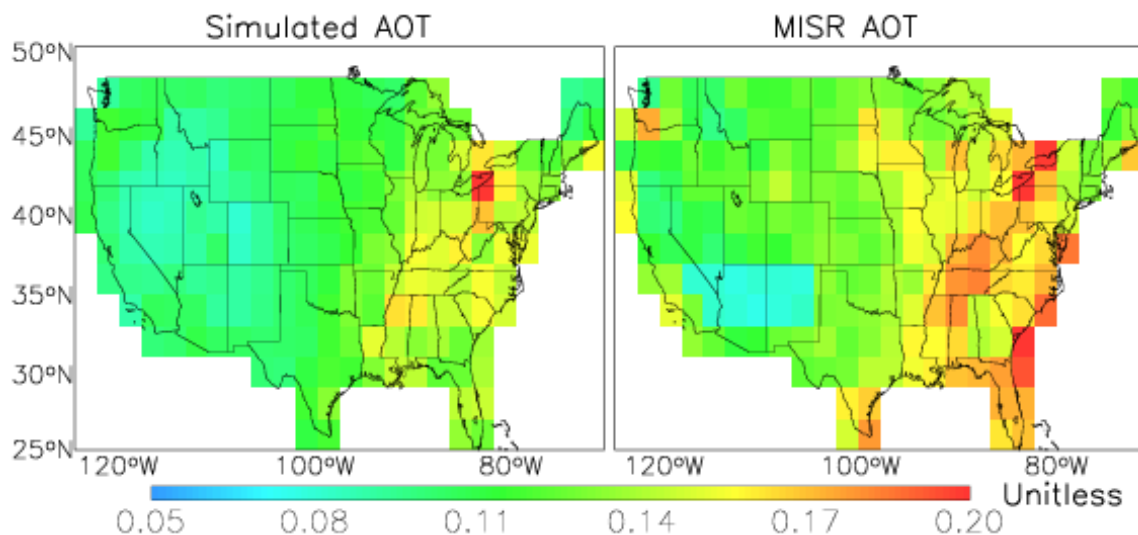


Figure 5. 2-D plot of annual average simulated AOT integrated in $2^\circ \times 2.5^\circ$ grid cells (left) vs. MISR AOT measurements (right) integrated in $2^\circ \times 2.5^\circ$ grid cells.

3.3.3 Comparison of MISR PM_{2.5} Concentrations With EPA Measurements

As shown in Figure 6, the annual MISR PM_{2.5} concentrations exhibit an improved agreement with EPA measurements in spatial pattern as compared to simulated PM_{2.5} concentrations, with more comparable concentrations in eastern central United States, DC-Maryland region. The ratio of MISR PM_{2.5} concentrations over EPA measurements is on average 0.90 with a standard deviation of 0.23. The MISR PM_{2.5} concentrations are generally lower than the EPA measurements in the northwest and higher in the east. Regression analysis shows that annual average MISR PM_{2.5} concentrations have a good linear relationship with EPA measurements ($r = 0.78$, linear regression slope = 0.91) and the estimated intercept is insignificant ($p = 0.84$). The RMSE of MISR PM_{2.5} is $2.32 \mu\text{g}/\text{m}^3$. Although PM_{2.5} concentrations in Southern California are underestimated, the MISR PM_{2.5} concentrations

are approximately 30 – 50% higher than the simulated PM_{2.5} concentrations in this region. When these three points are excluded, the relationship is further improved ($r = 0.81$) with an estimated slope of 1.00 and insignificant intercept. The RMSE is also improved to $2.20 \mu\text{g}/\text{m}^3$.

This comparison shows that the capability of MISR AOT to predict surface level PM_{2.5} concentrations can be substantially enhanced by including simulated aerosol vertical profiles. The annual MISR PM_{2.5} concentration is an unbiased predictor of EPA PM_{2.5} measurements with an approximately 20% relative error. The MISR PM_{2.5} concentration is not strongly influenced by the underestimation of both simulated PM_{2.5} concentration and AOT, which agrees with previous discussion. As a result, the difference between annual mean MISR PM_{2.5} concentrations and EPA measurements is smaller as compared to that between simulated PM_{2.5} concentrations and EPA measurements.

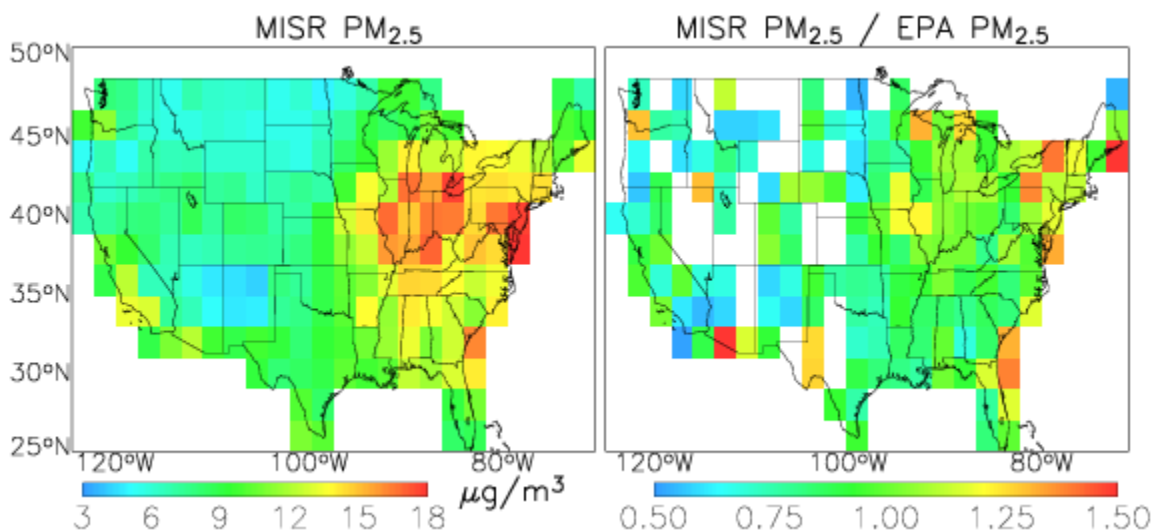


Figure 6. 2-D plot of annual average MISR PM_{2.5} concentrations integrated in $2^\circ \times 2.5^\circ$ grid cells (left) and the ratio of MISR PM_{2.5} concentrations over EPA PM_{2.5} concentrations integrated in $2^\circ \times 2.5^\circ$ grid cells (right). The scale of ratios saturates at 0.5 and 1.5 to best display the color contrast in the plot (1st percentile of ratio = 0.49, 99th percentile of ratio = 1.53).

4. Concluding Remarks

The two studies presented in this paper demonstrate the great potential of spaceborne aerosol sensors such as MISR as a cost-effective extension to ground monitoring networks. The empirical regression model developed in the first study shows that MISR AOT can serve as an important input to air quality models. With the help

of a few simple meteorological parameters, MISR AOT shows a strong association with ground level PM_{2.5} concentrations. The results of the second study illustrate the power of coupling global models and satellite retrieved aerosol optical properties in ambient air quality monitoring. Since the residence time of fine particles in lower troposphere ranges from 24 hours to a few days, surface level PM_{2.5} pollution likely exhibits a

regional nature except near large point sources. Therefore, the current spatial resolution ($2^{\circ} \times 2.5^{\circ}$) would be sufficient to evaluate the population exposure to $PM_{2.5}$ at national scale. Because of the global coverage of the satellite measurements and global model simulation results, this fully predictive approach can be easily transferred to other regions of the world without calibration using ground measurements.

Future research may be conducted to improve the approach presented in these studies. For example, seasonal average $PM_{2.5}$ concentrations may be estimated using the physical model in Section 3 as the launch of more advanced sensors further reduce the noise in AOT and increase the sampling frequency. In addition, a meso-scale model nested in the global CTMs is likely to preserve the impact of global scale aerosol events over the geographical region of interest, such as long-range transport of dust, and meanwhile provide higher spatial resolution. Furthermore, the relationship between MISR AOT, simulated AOT and $PM_{2.5}$ concentration presented in Section 3 may be established for each aerosol component separately. Therefore, how to utilize the aerosol composition and Angstrom exponent information provided in the latest MISR aerosol data product will be an interesting topic for future research.

5. Acknowledgement

This research is supported by Harvard University Center for the Environment (HUCE) Research Project Award, Harvard-EPA Center on Particle Health Effects (R827353-01-0), and the NASA Atmospheric Chemistry Modeling and Analysis Program. The authors appreciate the insightful thoughts from Drs. David Diner and Ralph Kahn and the technical support provided by the staff of the Atmospheric Sciences Data Center at NASA Langley Research Center. Drs. Mian Chin and Paul Ginoux are acknowledged for their help with GOCART dust AOT data. Finally, the authors thank the AERONET PIs for collecting the aerosol data over the United States.

References

Andrews, E., P. Saxena, S. Musarra, L.M. Hildemann, P. Koutrakis, P.H. McMurry, I. Olmez, and W.H. White, Concentration and Composition of Atmospheric Aerosols from the 1995 SEAVS Experiment and a Review of the Closure between

Chemical and Gravimetric Measurements, *J. Air & Waste Manage. Assoc.*, 50 (5), 648-664, 2000

Bey, I., D.J. Jacob, R.M. Yantosca, J.A. Logan, B. Field, A.M. Fiore, Q. Li, H. Liu, L.J. Mickley, and M. Schultz, Global modeling of tropospheric chemistry with assimilated meteorology: Model description and evaluation, *J. Geophys. Res.*, 106, 23,073-23,096, 2001.

Chin, M., P. Ginoux, S. Kinne, O. Torres, B.N. Holben, B.N. Duncan, R.V. Martin, J.A. Logan, A. Higurashi, and T. Nakajima, Tropospheric Aerosol Optical Thickness from the GOCART Model and Comparisons with Satellite and Sun Photometer Measurements, *J. Atmos. Sci.*, 59, 461-483, 2002.

Diner, D., J. Beckert, T.H. Reilly, C. Bruegge, J.E. Conel, R.A. Kahn, J. Martonchik, T.P. Ackerman, R. Davies, S.A.W. Gerstl, H. Gordon, J.-P. Muller, R.B. Myneni, P.J. Sellers, B. Pinty, and M.M. Verstraete, Multi-angle Imaging SpectroRadiometer (MISR) Instrument Description and experiment overview, *IEEE Trans. Geosci. Remote Sens.*, 36 (4), 1072-1087, 1998.

Engel-Cox, J.A., C.H. Holloman, B.W. Coutant, and R.M. Hoff, Qualitative and quantitative evaluation of MODIS satellite sensor data for regional and urban scale air quality, *Atmos. Environ.*, 38 (16), 2495-2509, 2004.

Hirsch, R., and E. Gilroy, Methods of fitting a straight line to data: examples in water resources, *Water Resour. Bull.*, 20 (5), 705 - 711, 1984.

Hudischewskyj, A. B.; Seigneur, C. Mathematical Modeling of the Chemistry and Physics of Aerosols in Plumes. *Environ. Sci. Technol.* 1989, 23, 413-421.

Jacobson, M. Z. Development and application of a new air pollution modeling system — Part III. Aerosol-phase simulations. *Atmos. Environ.* 1997, 31, 587-608.

Kent, G.S., M.P. McCormick, and P.H. Wang, Validation of stratospheric aerosol and gas experiment I and II satellite aerosol optical depth measurements using surface radiometer data, *J. Geophys. Res.*, 99 (D5), 10333-10339, 1994.

Kleeman, M.; Schauer, J.; Cass, G. Size and composition distribution of fine particulate matter emitted from motor vehicles. *Environ. Sci. Technol.* 2000, 34, 1132-1142.

Liu, Y., J. Sarnat, B.A. Coull, P. Koutrakis, and D.J. Jacob, Validation of Multiangle Imaging Spectroradiometer (MISR) aerosol optical thickness measurements using Aerosol Robotic Network (AERONET) observations over the contiguous United States, *J. Geophys. Res.*, 109 (D6), doi:10.1029/2003JD003981, 2004.

Malm, W. C.; Day, D. E.; SM, K. Light scattering characteristics for aerosols as a function of relative humidity: part I: A comparison of measured scattering and aerosol concentrations using the

- theoretical models. *J. Air & Waste Manage. Assoc.* 2000, 50, 686-700.
- Park, R.J., D.J. Jacob, M. Chin, and R.V. Martin, Sources of carbonaceous aerosols over the United States and implications for natural visibility, *J. Geophys. Res.*, 108 (D12), Art. No. 4355, 2003.
- Pope III, C.A., Review: epidemiological basis for particulate air pollution health standards, *Aerosol. Sci. Technol.*, 32, 4-14, 2000.
- Pun, B.; Seigneur, C. Sensitivity of particulate matter nitrate formation to precursor emissions in the California San Joaquin Valley. *Environ. Sci. Technol.* 2001, 35, 2979-2987.
- Schwartz J, Laden F, Zanobetti A. The concentration-response relation between PM_{2.5} and daily deaths. *Environ. Health Perspect.* 110 (10): 1025-1029 OCT 2002.
- Schwartz J, Norris G, Larson T, Sheppard L, Claiborne C, and Koenig J, Episodes of high coarse particle concentrations are not associated with increased mortality. *Environ. Health Perspect.* 107 (5): 339-342 MAY 1999.
- Seigneur, C. Current status of air quality models for particulate matter. *J. Air & Waste Manage. Assoc.* 2000, 51, 1508-1521.
- Seigneur, C.; Pai, P.; Hopke, P.; Grosjean, D. Modeling atmospheric particulate matter. *Environ. Sci. Technol.* 1999, 33, 80A-86A.
- Slater, J.; Dibb, J.; Keim, B.; Talbot, R. Light extinction by fine atmospheric particles in the White Mountains region of New Hampshire and its relationship to air mass transport. *Sci. Total Environ.* 2002, 287, 221-239.
- Smith, K. R. and Jantunen, M. Why particles? *Chemosphere* 2002, 49, 867-871.
- Stohl, A. A 1-year Lagrangian "climatology" of airstreams in the Northern Hemisphere troposphere and lowermost stratosphere. *J. Geophys. Res.* 2001, 106, 7263-7279.
- Thulasiraman, S.; O'Neill, N. T.; Royer, A.; Holben, B.; Westphal, D.; McArthur, L. J. B. Sunphotometric observations of the 2001 Asian dust storm over Canada and the US. *Geophys. Res. Lett.* 2002, 29, art no. 1255.
- Wallace, L., Correlations of personal exposure to particles with outdoor air measurements: A review of recent studies, *Aerosol. Sci. Technol.*, 32 (1), 15-25, 2000.