Mapping annual mean ground-level $PM_{2.5}$ concentrations using Multiangle Imaging Spectroradiometer aerosol optical thickness over the contiguous United States

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[1] We present a simple approach to estimating ground-level fine particulate matter ($PM_{2.5}$, particles smaller than 2.5 μm in diameter) concentrations by applying local scaling factors from a global atmospheric chemistry model (GEOS-CHEM with GOCART dust and sea salt data) to aerosol optical thickness (AOT) retrieved by the Multiangle Imaging Spectroradiometer (MISR). The resulting MISR $PM_{2.5}$ concentrations are compared with measurements from the U.S. Environmental Protection Agency's (EPA) $PM_{2.5}$ compliance network for the year 2001. Regression analyses show that the annual mean MISR $PM_{2.5}$ concentration is strongly correlated with EPA $PM_{2.5}$ concentration (correlation coefficient $r = 0.81$), with an estimated slope of 1.00 and an insignificant intercept, when three potential outliers from Southern California are excluded. The MISR $PM_{2.5}$ concentrations have a root mean square error (RMSE) of 2.20 μg/m^3, which corresponds to a relative error (RMSE over mean EPA $PM_{2.5}$ concentration) of approximately 20%. Using simulated aerosol vertical profiles generated by the global models helps to reduce the uncertainty in estimated $PM_{2.5}$ concentrations due to the changing correlation between lower and upper tropospheric aerosols and therefore to improve the capability of MISR AOT in estimating surface-level $PM_{2.5}$ concentrations. The estimated seasonal mean $PM_{2.5}$ concentrations exhibited substantial uncertainty, particularly in the west. With improved MISR cloud screening algorithms and the dust simulation of global models, as well as a higher model spatial resolution, we expect that this approach will be able to make reliable estimation of seasonal average surface-level $PM_{2.5}$ concentration at higher temporal and spatial resolution. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0394 Atmospheric Composition and Structure: Instruments and techniques; *KEYWORDS:* MISR AOT, GEOS-CHEM, $PM_{2.5}$

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1. Introduction

[2] Epidemiological studies around the world have shown strong and consistent correlations between adverse health effects and outdoor fine particle matter ($PM_{2.5}$, particles with diameters less than 2.5 μm) [*Pope*, 2000]. Some of the mortality studies as reviewed by *Wallace* [2000] have consistently shown an increase of 1–8% in deaths per 50 μg/m^3 increase in outdoor air particle concentrations without apparent threshold. The importance of long-term $PM_{2.5}$ monitoring has recently been emphasized in order to evaluate the health effects of low or moderate exposure as well as repeated exposure to elevated pollution levels [*Samet et al.*, 2000; *Schwartz et al.*, 1996]. However, for many epidemiology studies, particle measurements from stationary ambient monitoring (SAM) sites have been used as surrogates of exposures for individuals living substantial distances (20–100 miles) from the sites [*Ito et al.*, 2001].

[3] Since aerosol optical properties such as aerosol optical thickness (AOT, a dimensionless measure of aerosol abundance and its light extinction capability in the entire air column) derived from satellite observations is directly related to particle mass loading [*Chow et al.*, 2002; *Malm et al.*, 1994], studying its association with surface-level $PM_{2.5}$ mass concentration may provide a cost effective

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way for $PM_{2.5}$ pollution monitoring. To date, satellite remote sensing has been applied to monitor long-range transport of Asian and Saharan dust [*Chiapello and Moulin*, 2002; *Husar et al.*, 2001; *Wang et al.*, 2003] and characterize ambient particulate pollution [*Falke et al.*, 2001; *Liu et al.*, 2002; *Sifakis and Deschamps*, 1992].

[4] The launch of NASA EOS satellite Terra in December 1999 provided a new opportunity for monitoring particle pollution from space. Two instruments aboard Terra, MISR and the Moderate-Resolution Imaging Spectroradiometer (MODIS), were specially designed to retrieve aerosol optical properties including AOT over most of the land surface and the oceans [*Diner et al.*, 1998; *Kaufman et al.*, 1998, 2002; *Martonchik et al.*, 1998]. Preliminary studies have shown that MODIS AOT data can be used to detect and track the transport of pollutants and extreme pollution episodes [*Engel-Cox et al.*, 2004; *Hutchison*, 2003]. In addition, a strong linear relationship (correlation coefficient $r = 0.70$) was found between MODIS AOT measurements and 24-hour $PM_{2.5}$ concentrations from seven sites in Alabama, indicating a good potential for satellite derived aerosol optical properties to be used in air quality studies [*Wang and Christopher*, 2003].

[5] We showed in a previous study that an empirical regression model using MISR AOT and a few geographical and meteorological parameters is able to estimate surface-level 24-hour average $PM_{2.5}$ concentrations within approximately 45%, with a correlation coefficient of approximately 0.7 between observed and predicted $PM_{2.5}$ concentrations (Y. Liu et al., Estimating ground level $PM_{2.5}$ over the eastern United States using satellite remote sensing, submitted to *Environmental Science and Technology*, 2004, hereinafter referred to as Liu et al., submitted manuscript, 2004). This paper extends our previous study by developing a simple approach that establishes a predictive relationship between surface $PM_{2.5}$ concentrations and AOT. This simple approach relies on a global chemistry and transport model (CTM) to provide a better physical basis for relating satellite AOT measurements to the spatial and temporal pattern of surface $PM_{2.5}$ concentrations.

[6] In section 2, we describe this approach, which involves analysis of data from the CTMs, $PM_{2.5}$ data from the U.S. Environmental Protection Agency's (EPA) $PM_{2.5}$ compliance network and speciation and trend network (STN), and AOT retrievals from MISR for the year 2001. In section 3, the summary statistics of the different data sets are presented. The agreement in geographical patterns and seasonal variations between model and observations is discussed. $PM_{2.5}$ concentrations derived from the MISR AOT are compared with EPA $PM_{2.5}$ data and sources of uncertainties are discussed in detail. Finally, major findings and potential future improvements to the current analysis are summarized in section 4.

2. Description of Data and Method

2.1. Simulated Aerosol Data by GEOS-CHEM and GOCART

[7] The GEOS-CHEM model is a global 3-D tropospheric chemistry and transport model driven by assimilated meteorological observations from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office. The fully coupled oxidants-aerosol simulation by GEOS-CHEM provides sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), elemental carbon (EC), and organic carbon (OC) aerosol concentrations for the period of 2001 at 3-hour temporal resolution, 2° latitude × 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 hydroscopic growth factors to all hydrophilic species using local relative humidity conditions [*Martin et al.*, 2003]. Detailed descriptions of GEOS-CHEM as well as its aerosol simulations can be found elsewhere [*Bey et al.*, 2001; *Park et al.*, 2003, 2004]. The lowest model levels are centered at approximately 10, 50, 100, 200, and 400 m above the surface.

[8] Since EPA's compliance network measures 24-hour average $PM_{2.5}$ concentration according to a fixed sampling schedule (every third or sixth day) regardless of weather conditions, seasonal mean GEOS-CHEM surface $PM_{2.5}$ concentrations were calculated using all eight 3-hour outputs per day for each species. The calculation of columnar AOT values followed the methodology given by *Chin et al.* [2002]. The 3-hour values were first interpolated to 10 a.m. local time values (MISR measurement time window), sampled on the dates when MISR had valid AOT retrievals, then integrated into seasonal averages in order to be compared with seasonal mean MISR AOT values.

[9] Monthly mean dust and sea salt concentrations for 2001 from the Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) model were used to complement GEOS-CHEM aerosol fields. General descriptions of the GOCART model simulation of dust and sea salt are provided elsewhere [*Chin et al.*, 2002; *Ginoux et al.*, 2001]. Particle concentrations and the associated AOT values for fine sea salt (effective radius of 0.80 μm), and fine dust (sum of effective radius ranges 0.1–0.18 μm, 0.18–0.30 μm, 0.3–0.6 μm, 0.6–1 μm, and part of 1–1.8 μm) provided by the GOCART model were combined with the particle concentrations for SO_4^{2-}, NO_3^-, NH_4^+, EC, and OC and their associated AOT values provided by GEOS-CHEM. From this point on, we refer to the total aerosol AOT and total surface aerosol concentration predicted by combining GEOS-CHEM and GOCART outputs as simulated AOT and simulated $PM_{2.5}$ concentration.

2.2. EPA 24-Hour Average $PM_{2.5}$ Mass Concentration and Speciation Data

[10] The EPA's $PM_{2.5}$ compliance network was initiated in 1997 and designed to measure compliance of both the annual and 24-hour $PM_{2.5}$ National Ambient Air Quality Standard (NAAQS). Daily average $PM_{2.5}$ concentrations measured by gravimetric methods (Federal Register 40 CFR part 50, 5 Feb. 1998) in 2001 from 1137 sites of EPA's compliance network, primarily located in urban areas and surrounding suburbs, were collected and integrated into seasonal averages in each GEOS-CHEM model grid cell (Figure 1). Validated daily average mass concentrations of SO_4^{2-}, EC, OC, and mineral dust were collected from 131 sites of EPA's $PM_{2.5}$ speciation trends network (STN) and integrated into seasonal averages in 2° × 2.5° model grid in order to analyze the difference between simulated $PM_{2.5}$ concentrations and EPA

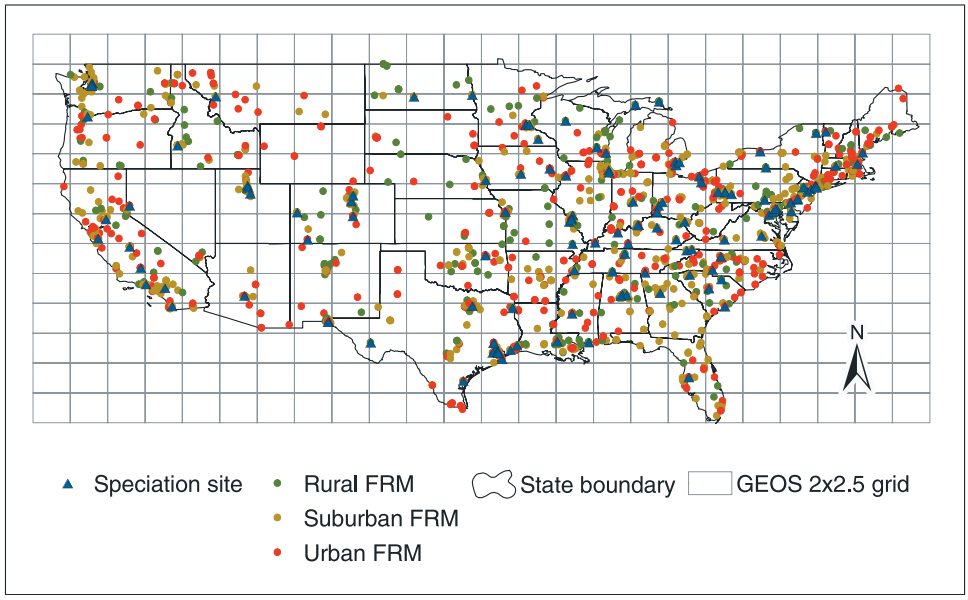

Figure 1. Spatial distribution of the EPA $PM_{2.5}$ compliance monitoring sites (FRM sites, circles) and speciation and trend sites (STN sites, triangles) in the contiguous United States. Data in this study are collected from 1137 FRM sites and 131 STN sites for the year 2001.

$PM_{2.5}$ measurements for individual aerosol components [*Rao et al.*, 2002] (Figure 1). To reduce the influence of potential outliers, only the grid cells with more than 30 EPA measurements per season were included in the analysis.

2.3. MISR AOT Retrievals

[11] MISR AOT data have a spatial resolution of 17.6 km and achieve global coverage in nine days [*Diner et al.*, 1998; *Martonchik et al.*, 2002]. It is most sensitive to particles in the diameter range from 0.05 to 2.0 μm [*Kahn et al.*, 1998], corresponding to the size range of $PM_{2.5}$. All MISR AOT data (mostly version 12) that covered the contiguous United States for 2001 were obtained from the NASA Langley Research Center Atmospheric Sciences Data Center (http://edg.larc.nasa.gov/~imswww/imswelcome/index.html), and then integrated into seasonal averages in 2° × 2.5° model grid cells. We excluded AOT values greater than 1.5 from our analysis because they are probably contaminated by clouds (D. Diner, personal communication, 2003). In addition, the data from North Dakota, South Dakota and Minnesota in the winter and spring is excluded because of a potential cloud contamination in MISR AOT. This issue will be further discussed in the following analysis. We previously showed that the overall retrieval error of MISR AOT is $\Delta AOT = \pm 0.04 \pm 0.18 \times AOT$ over the contiguous United States [*Liu et al.*, 2004]. The seasonal and geographical variability of MISR AOT errors was partly corrected in this analysis by applying linear regressions between MISR and AERONET AOT values presented by *Liu et al.* [2004]. Finally, only those grid cells with more than 30 AOT measurements per season were included in the analysis in order to reduce the influence of potential outliers.

2.4. Coupling of the Global Models With MISR

[12] We previously showed that an empirical regression model is able to predict surface $PM_{2.5}$ concentrations by using MISR AOT data and simple meteorological and geographical predictors with a relative error of approximately 45% ($r \approx 0.7$) and with no significant biases compared to observations (Liu et al., submitted manuscript, 2004). However, half of the variability in $PM_{2.5}$ concentrations cannot be explained probably because of 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 the simulated AOT and $PM_{2.5}$ concentrations from GEOS-CHEM and GOCART model to define a physically consistent relationship between AOT and surface-level $PM_{2.5}$ concentration:

$$\text{MISR } PM_{2.5} \text{ Concentration} = \frac{\text{Simulated Surface Level } PM_{2.5} \text{ Concentration}}{\text{Simulated Column AOT}} \times \text{MISR AOT} \quad (1)$$

This relationship, as defined in equation (1), is then applied to MISR AOTs 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 terms of particle mass concentrations and optical properties used in this analysis is summarized in Table 1.

[13] 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. For example, if both simulated $PM_{2.5}$ concentrations and AOT have consistent biases (i.e., both high or both low), the uncertainty of MISR AOT measurements will be more influential in determining the uncertainty in MISR $PM_{2.5}$ than either simulated $PM_{2.5}$ concentration or simu-

Table 1. Definitions of Terms Used in This Analysis

Term	Unit	Definition
Simulated $PM_{2.5}$ concentration	$\mu g/m^3$	sea salt and dust mass concentrations are derived from GOCART, and mass concentrations for SO_4^{2-}, NO_3^-, NH_4^+, EC, and OC are derived from GEOS-CHEM; these two sets of concentration data are combined to form the total simulated $PM_{2.5}$ concentrations
Simulated AOT	unitless	AOT values for sea salt and dust are from GOCART, and AOT values for the remaining particulate species are from GEOS-CHEM; these two sets of AOT data are combined to form the total simulated AOT estimates
EPA $PM_{2.5}$ concentration	$\mu g/m^3$	daily average $PM_{2.5}$ mass concentrations measured by gravimetric methods collected from the EPA $PM_{2.5}$ Monitoring and Compliance Network
MISR AOT	unitless	total column AOT retrieved by MISR instrument aboard the Terra satellite
MISR $PM_{2.5}$ concentration	$\mu g/m^3$	daily average $PM_{2.5}$ concentrations estimated by equation (1)

lated AOT. Therefore the MISR $PM_{2.5}$ is likely to have less discrepancy in spatial and seasonal patterns than simulated $PM_{2.5}$ concentrations when compared with EPA measurements. 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 subgrid 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; *Turpin and Lim*, 2001]. 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 equation (1) 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 nonlinear relationship. Under such circumstances, MISR $PM_{2.5}$ derived from equation (1) would only be a first-order approximation of EPA $PM_{2.5}$ measurements.

3. Results and Discussion

[14] The final data set consists of totally 577 seasonal 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, and 159 annual data records after averaging all seasonal records. The relationships among three $PM_{2.5}$ concentration parameters and two AOT parameters are studied using scatterplots, and Spearman's correlation coefficients. In addition, 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 capability of equation (1) in estimating individual $PM_{2.5}$ concentration over a given grid cell.

3.1. Comparison Between Simulated $PM_{2.5}$ and EPA $PM_{2.5}$ Measurements

[15] The annual average simulated $PM_{2.5}$ concentration (8.36 (mean) ± 3.28 (standard deviation) $\mu g/m^3$) is approximately 20% lower than EPA $PM_{2.5}$ concentrations (10.76 ± 3.14 $\mu g/m^3$) with a similar dynamic range (Table 2). Figure 2 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° × 0.5° grid. The annual mean 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 scatterplot 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 $\mu g/m^3$ (Figure 3). The three data points that apparently deviate from the general pattern of the data set 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.

[16] Seasonally, the difference between simulated $PM_{2.5}$ concentrations and EPA measurement is the largest in the winter (12.17 $\mu g/m^3$ versus 8.26 $\mu g/m^3$, 32% difference)

Table 2. Summary Statistics of Annual and Seasonal Average MISR $PM_{2.5}$ Concentrations, EPA $PM_{2.5}$ Concentrations, Model Columnar AOT, MISR AOT, and MISR $PM_{2.5}$ Concentrations in the United States for the Year 2001

Season	Variable	Unit	N	Mean	SD[a]	Min	Max
Annual	EPA $PM_{2.5}$	$\mu g/m^3$	159	10.76	3.14	4.72	20.51
	simulated $PM_{2.5}$	$\mu g/m^3$	159	8.36	3.28	3.70	17.17
	MISR $PM_{2.5}$	$\mu g/m^3$	159	9.68	3.68	3.22	18.24
	MISR AOT	unitless	159	0.13	0.03	0.05	0.22
	simulated AOT	unitless	159	0.10	0.03	0.06	0.22
Winter	EPA $PM_{2.5}$		129	12.17	3.62	5.21	24.77
	simulated $PM_{2.5}$		129	8.27	3.93	2.32	19.69
	MISR $PM_{2.5}$		129	8.56	3.20	2.43	17.99
	MISR AOT		129	0.08	0.02	0.03	0.11
	simulated AOT		129	0.07	0.03	0.03	0.16
Spring	EPA $PM_{2.5}$		135	10.23	3.34	3.85	20.48
	simulated $PM_{2.5}$		135	8.50	3.46	2.90	17.29
	MISR $PM_{2.5}$		135	11.51	5.00	4.29	26.92
	MISR AOT		135	0.16	0.04	0.08	0.32
	simulated AOT		135	0.11	0.03	0.07	0.25
Summer	EPA $PM_{2.5}$		155	11.32	4.91	4.15	22.98
	simulated $PM_{2.5}$		155	8.39	3.34	2.74	17.39
	MISR $PM_{2.5}$		155	11.68	5.81	4.08	29.23
	MISR AOT		155	0.18	0.08	0.05	0.38
	simulated AOT		155	0.13	0.05	0.06	0.29
Fall	EPA $PM_{2.5}$		158	9.98	3.18	3.28	24.05
	simulated $PM_{2.5}$		158	8.32	3.18	3.79	15.05
	MISR $PM_{2.5}$		158	7.71	3.37	2.07	19.46
	MISR AOT		158	0.09	0.03	0.04	0.19
	simulated AOT		158	0.10	0.03	0.04	0.20

[a]SD, standard deviation.

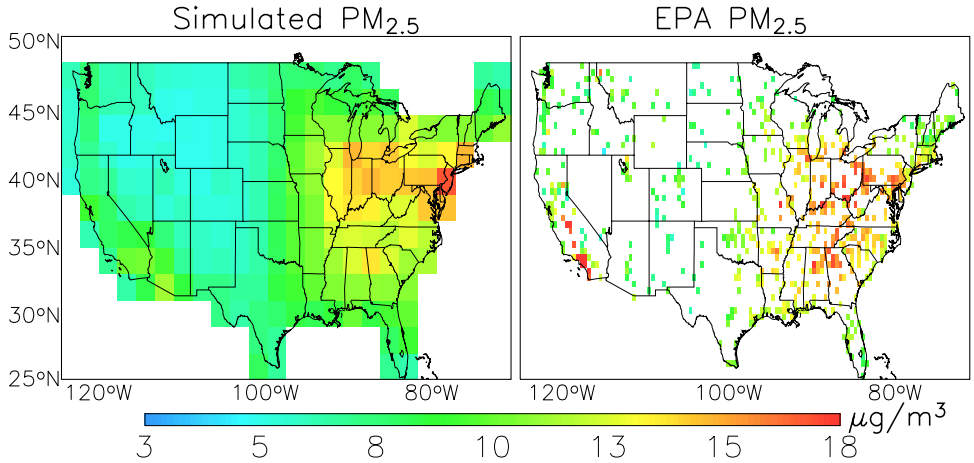

Figure 2. Two-dimensional plot of annual (left) simulated $PM_{2.5}$ concentrations integrated in 2° × 2.5° grid cells versus (right) EPA $PM_{2.5}$ measurements integrated in 0.5° × 0.5° grid cells. The scale saturates at 18 μg/m^3 to best display the color contrast in the plot (99th percentile of EPA $PM_{2.5}$ measurement = 17.05 μg/m^3).

and smallest in the fall (9.98 μg/m^3 versus 8.32 μg/m^3, 17% difference). The global models underestimate $PM_{2.5}$ concentrations by approximately 30% during the summer. Simulated $PM_{2.5}$ concentrations are significantly correlated with EPA measurements in all seasons with the exception of the western United States where correlation coefficients are insignificant in the winter and summer ($p > 0.05$) (Table 3). EPA measurements are substantially higher than simulated $PM_{2.5}$ concentrations in California during the winter and the fall. In addition, during the winter, EPA measurements show strong spatial variation in the northwest region with a number of stations observing much higher $PM_{2.5}$ concentrations than other stations in the same region. Scatterplots of simulated $PM_{2.5}$ concentrations versus EPA $PM_{2.5}$ measurements show that simulated $PM_{2.5}$ concentrations agree with observations better in the east than in the west (Figure 4). Larger scatter is found in the west especially in the winter and summer as compared to in the east.

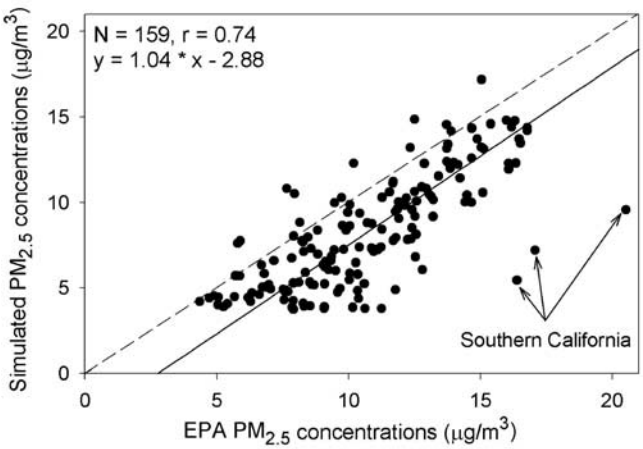

Figure 3. Scatterplot of annual average simulated $PM_{2.5}$ concentration versus EPA $PM_{2.5}$ measurements. The reduced major axis line is shown as the solid line in the plot. The 1:1 line is shown as the short-dashed line for reference. Three potential outliers pointed out by arrows are all Southern California grid cells.

[17] 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. The possible reason for the summer bias in the east is described by *Park et al.* [2004]. The weak correlation found in the west in the summer might be due to the high bias in GOCART monthly mean dust concentrations. Along the coast from Washington State to central California, GOCART dust concentrations are approximately 5–7 μg/m^3. However, dust concentrations measured by STN sites are generally below 1.5 μg/m^3. In addition, in the inland northwest region, GOCART also overestimates dust concentrations by a factor of 2–3. This high bias and disagreement in spatial pattern between simulated and observed dust concentrations likely cause the insignificant correlation. Since in the eastern United States, $PM_{2.5}$ concentrations are

Table 3. Spearman's Correlation Coefficients Between EPA $PM_{2.5}$ and MISR AOT, Simulated $PM_{2.5}$ and MISR $PM_{2.5}$ Concentrations in Each Season in the Eastern and Western United States

		Correlation Coefficient (p Value)[a]		
Season	Region	Simulated $PM_{2.5}$	MISR AOT	MISR $PM_{2.5}$
Winter	east[b]	0.73 (<0.0001)	−0.03 (0.78)	0.49 (<0.0001)
	west[c]	−0.15 (0.23)	0.26 (0.04)	0.09 (0.51)
Spring	east	0.58 (<0.0001)	−0.04 (0.77)	0.19 (0.10)
	west	0.58 (<0.0001)	0.09 (0.51)	0.55 (<0.0001)
Summer	east	0.82 (<0.0001)	0.50 (<0.0001)	0.67 (<0.0001)
	west	−0.02 (0.88)	0.55 (<0.0001)	0.39 (0.0007)
Fall	east	0.68 (<0.0001)	0.09 (0.40)	0.38 (0.0004)
	west	0.65 (<0.0001)	0.43 (<0.0001)	0.64 (<0.0001)

[a]The p values reflect the significance level of the correlation coefficients. The correlation is not significant if p is greater than 0.05.
[b]Refers to regions to the east of 95° longitude.
[c]Refers to regions to the west of 95° longitude.

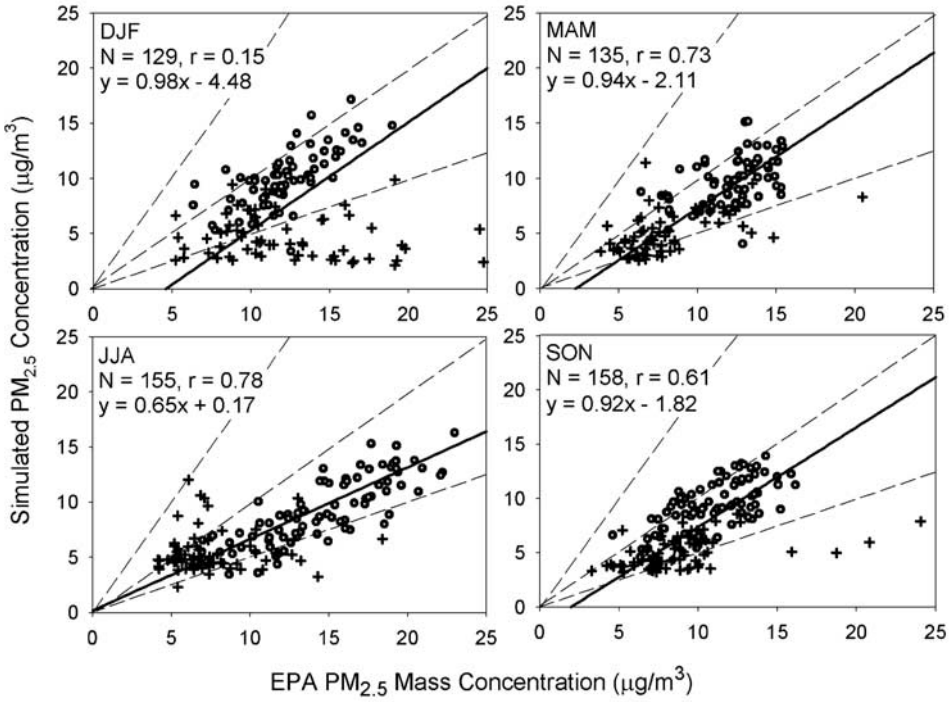

Figure 4. Scatterplots of seasonal average (left) simulated $PM_{2.5}$ concentrations with (right) EPA $PM_{2.5}$ measurements over the United States in 2001. Data from the eastern United States are shown as circles, and data from the western United States are shown as crosses. The total number of data points in each season *N*, the correlation coefficient *r*, and the reduced major axis equation are presented in each plot. The reduced major axis line is shown as the thick solid line in each plot. The 2:1, 1:1, and 1:2 lines are shown as the thin dashed lines for reference.

generally dominated by sulfate that are transported from regional sources of SO_2, the impact of urban excess concentrations is relatively small [*West et al.*, 1999]. Therefore good agreement between model and observations is noted.

[18] The large discrepancy and weak correlation in the northwest region in the winter may be because there is only a limited number of EPA monitoring sites in this region. The EPA $PM_{2.5}$ concentrations from some of the urban sites are heavily influenced by strong carbonaceous aerosols emitted from local sources such as automobiles and wood fires and can be substantially higher than surrounding suburban and rural areas [*Rao et al.*, 2002]. Since EPA sites are relatively sparse in the northwest, a small number of urban sites can greatly influence the mean $PM_{2.5}$ concentration in a $2° \times 2.5°$ grid cell. Therefore it is not surprising to see that global models substantially underestimate $PM_{2.5}$ concentrations at current spatial resolution.

3.2. Comparison Between Simulated and MISR AOTs

[19] Annual average simulated AOT (0.10 ± 0.03) is approximately 20% lower than MISR AOT (0.13 ± 0.06) with a similar dynamic range (Table 2). The largest seasonal difference is found in the spring when simulated AOT (0.11 ± 0.03) is approximately 30% lower than MISR retrievals (0.16 ± 0.04). 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 scatterplot 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 line slope = 0.83) (Figure 6).

[20] A seasonal comparison shows that although significant in all four seasons, the correlations between simulated and MISR AOT values are substantially stronger during the summer (r = 0.78) and the fall (r = 0.62) than during the winter (r = 0.39) and the spring (r = 0.42) (Figure 7). The overall agreement between simulated AOT and MISR AOT retrievals does not vary significantly by geographical region except that MISR AOT is substantially higher than simulated AOT over coastal Washington State, North Dakota, South Dakota and Minnesota during the spring.

[21] Current MISR AOT data includes the aerosol extinction effect in the entire atmospheric column in both the troposphere and the stratosphere (C. Welch, personal communication, 2003). Although stratospheric AOT is usually at least 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.

[22] The discrepancies in spatial and seasonal patterns between simulated AOT and MISR AOT may be attributed to the uncertainties associated with both the global models

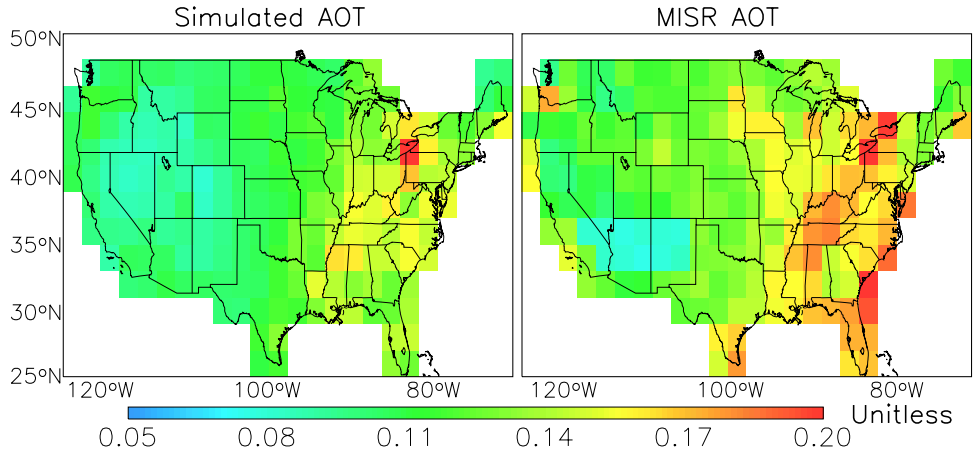

Figure 5. Two-dimensional plot of annual average (left) simulated AOT integrated in 2° × 2.5° grid cells versus (right) MISR AOT measurements integrated in 2° × 2.5° grid cells.

and MISR AOT retrievals. In terms of model uncertainty, the overall weak seasonal correlation between MISR AOT and simulated AOT might partly be due to the use of monthly mean GOCART dust concentrations (sea salt particles only comprise a very small fraction in AOT values over land). Analysis of GEOS-CHEM and GOCART aerosol vertical profiles indicates that on average, dust particles account for 25% of the total columnar AOT in the east and 36% in the west. Additionally, a factor of 2 difference between MODIS AOT, which has a similar accuracy level, and GOCART model results was previously reported in the coastal region of Washington State for the spring of 2001 [*Yu et al.*, 2003]. It was attributed to the active long-range transport of Asian dust in the spring of 2001. Therefore underestimated aerosol events might also contribute to the differences between simulated AOT and MISR AOT.

[23] In terms of the uncertainties in MISR AOT, we previously showed that the retrieval errors in MISR AOT tend to be larger when a large amount of coarse particles is present [*Liu et al.*, 2004]. Air quality in the United States is influenced by transported African dust in the summer [*Prospero*, 1999] and transpacific dust transport from Asia in the spring [*Jaffe et al.*, 2003]. Consequently, the higher noise level in MISR AOT due to higher fraction of dust in the total AOT are likely deteriorate its correlation with simulated AOT. In addition, the region of high AOT values in the winter and spring in North Dakota, South Dakota and Minnesota might be caused by insufficient cloud masking in the MISR retrieval algorithm (R. Kahn, personal communication, 2003). Finally, considering the narrow dynamic range and low values of AOT during the winter, the correlation between simulated AOT and MISR AOT could be significantly weakened by the retrieval errors in MISR AOT.

3.3. Comparison Between MISR $PM_{2.5}$ Concentrations and EPA Measurements

[24] The annual average MISR $PM_{2.5}$ concentration over the United States (9.68 ± 3.68 μg/m^3) is approximately 10% lower than the EPA measurements with a similar dynamic range (Table 2). As shown in Figure 8, 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 the eastern central United States, District of Columbia-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. A scatterplot 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) (Figure 9). The root mean square error (RMSE) of MISR $PM_{2.5}$ concentrations is 2.32 μg/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 μg/m^3.

[25] Seasonally, the difference between MISR $PM_{2.5}$ and EPA $PM_{2.5}$ measurements is the largest in the winter (8.27 μg/m^3 versus 12.17 μg/m^3, 33% difference) and small-

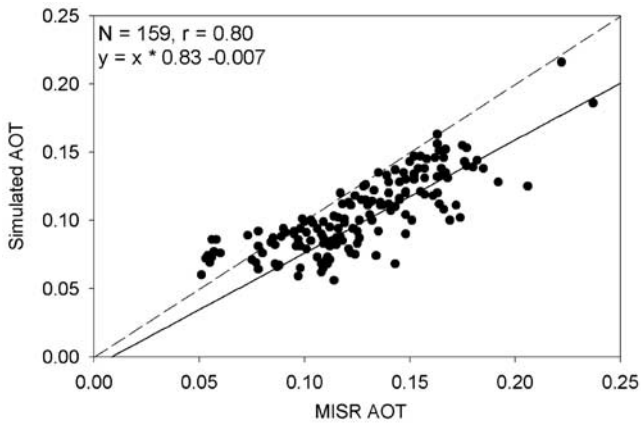

Figure 6. Scatterplot of annual average simulated AOT versus MISR AOT. The reduced major axis line is shown as the solid line in the plot. The 1:1 line is shown as the short-dashed line for reference.

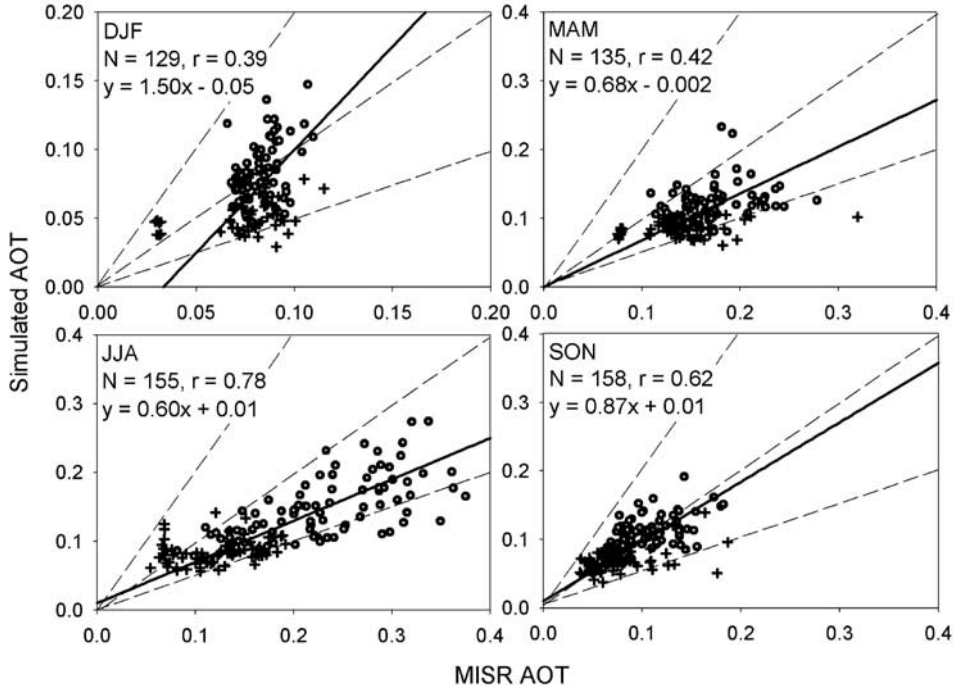

Figure 7. Scatterplots of seasonal average (left) simulated AOT and (right) MISR AOT retrievals in 2001 in the United States. Data from the eastern United States are shown as circles, and data from the western United States are shown as crosses. The total number of data points in each season *N*, the correlation coefficient *r*, and the reduced major axis equation are presented in each plot. The reduced major axis line is shown as the thick solid line in each plot. The 2:1, 1:1, and 1:2 lines are shown as the thin dashed lines for reference.

est in the summer (11.68 μg/m^3 versus 11.32 μg/m^3, −3% difference). The MISR $PM_{2.5}$ concentrations are generally higher than EPA measurements except during the fall (7% lower). Spearman's correlation coefficients between MISR $PM_{2.5}$ and EPA measurements are significant in all seasons with the exception of the winter in the west and spring in the east, which do not indicate an overall improvement over the simulated $PM_{2.5}$ concentrations (Table 3).

[26] We showed in a previous study that MISR AOT and a few meteorological parameters are able to predict surface $PM_{2.5}$ concentrations with a 45% relative error (Liu et al., submitted manuscript, 2004). In this analysis, we show 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 regression results indicate that the annual MISR $PM_{2.5}$ concentration is an

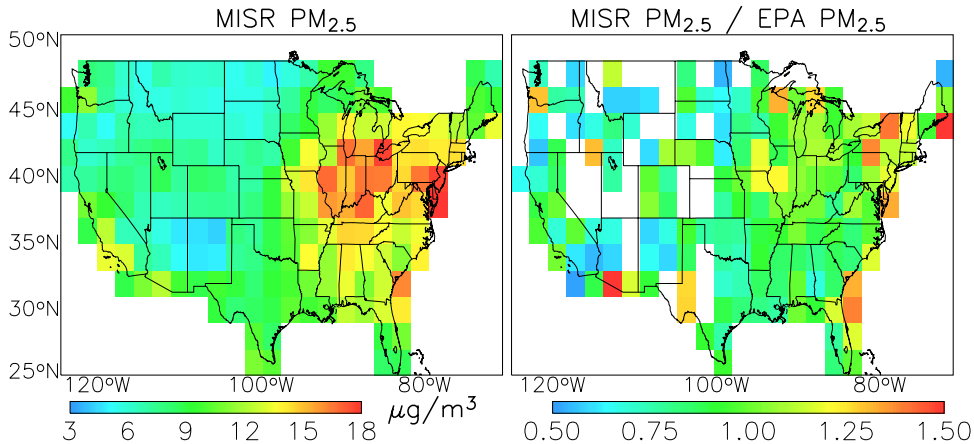

Figure 8. Two-dimensional plot of annual average (left) MISR $PM_{2.5}$ concentrations integrated in 2° × 2.5° grid cells and (right) ratio of MISR $PM_{2.5}$ concentrations over EPA $PM_{2.5}$ concentrations integrated in 2° × 2.5° grid cells. The scale of ratios saturates at 0.5 and 1.5 to best display the color contrast in the plot (first percentile of ratio = 0.49, 99th percentile of ratio = 1.53).

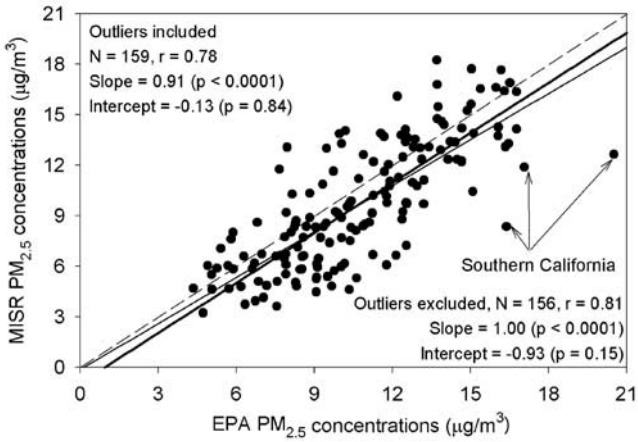

Figure 9. Scatterplot of annual average MISR $PM_{2.5}$ concentrations from equation (1) versus EPA measurements. The parameter estimates of the simple linear regression lines (N, r, and estimated slope and intercept as well as their p values), with and without three outliers from Southern California, are presented. The linear regression line with the outliers included is shown as the thin solid line. The linear regression line with the outliers excluded is shown as the thick solid line. The 1:1 line is shown as the dashed line for reference.

unbiased predictor of EPA $PM_{2.5}$ measurements with an approximately 20% relative error (RMSE over mean EPA $PM_{2.5}$ concentration). The MISR $PM_{2.5}$ concentration is not strongly influenced by the underestimation of both simulated $PM_{2.5}$ concentration and AOT in the winter in northwest, which agrees with previous analysis of equation (1). 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, as shown in the summary statistics and the regression results.

[27] The uncertainty associated with seasonal MISR $PM_{2.5}$ concentrations is significantly greater as indicated by the changing correlation coefficients between seasonal average MISR $PM_{2.5}$ concentrations and EPA measurements (Table 3). The generally weaker correlation in the west as compared to the east may be partially attributed to the heterogeneous spatial distribution of EPA network and the uncertainties in MISR AOT data, as discussed in the previous sections. Higher uncertainties associated with aerosol estimations by global CTMs under complex terrain and climate conditions, such as in Southern California, can also deteriorate the predictability of equation (1).

4. Conclusion

[28] A simple approach to estimating surface-level $PM_{2.5}$ concentration using simulated aerosol data by GEOS-CHEM and MISR AOT retrievals is presented using data in 2001 in the contiguous United States. Simple linear regression shows that the annual MISR $PM_{2.5}$ concentration is strongly correlated with EPA $PM_{2.5}$ concentration (r = 0.78), with an estimated slope of 0.91 and an insignificant intercept. When three potential outliers covering Southern California are excluded, the regression line has a stronger correlation efficient (r = 0.81), an estimated slope of 1.00, and an insignificant intercept. The MISR $PM_{2.5}$ concentrations have a relative error of approximately 20%, indicating a significant improvement over using MISR AOT as the only indicator of ground $PM_{2.5}$ concentrations.

[29] The results of this analysis present a promising application of 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 ($2° \times 2.5°$) 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. Therefore current results are valuable for many parts of the United States without extensive surface-level monitoring networks, and throughout the developing countries where research on particle pollution and related public health issues is severely limited by available $PM_{2.5}$ data. Naturally, results with a finer resolution will improve the characterization of $PM_{2.5}$ spatial variability and will, subsequently, reduce the potential for exposure misclassification in $PM_{2.5}$ epidemiologic studies. For example, simulated particle vertical profiles at a higher spatial resolution (e.g., 10–20 km) will be needed for $PM_{2.5}$ epidemiology studies in large metropolitan statistical areas (MSAs).

[30] Future research may be conducted to improve the approach presented in this analysis. For example, seasonal average $PM_{2.5}$ concentrations estimated by equation (1) contain substantial uncertainties due to the noise in MISR AOT and the difficulties that global CTMs encounter under complex terrain and weather conditions. A mesoscale 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. Together with the continuous improvement of MISR data quality, we can expect that this approach will be able to estimate ground $PM_{2.5}$ concentrations with a higher level of confidence at seasonal level or higher time resolutions. Furthermore, as discussed previously, MISR $PM_{2.5}$ might have substantial uncertainty when more than one major aerosol species with different optical properties and vertical distributions are present in the atmospheric column. The relationship between MISR AOT, simulated AOT and $PM_{2.5}$ concentration similar to what has been presented in equation (1) 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.

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References

Andrews, E., P. Saxena, S. Musarra, L. M. Hildemann, P. Koutrakis, P. H. McMurry, I. Olmez, and W. H. White (2000), 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.

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 (2001), Global modeling of tropospheric chemistry with assimilated meteorology: Model description and evaluation, *J. Geophys. Res.*, *106*(D19), 23,073–23,096.

Chiapello, I., and C. Moulin (2002), TOMS and Meteosat satellite records of the variability of Saharan dust transport over the Atlantic during the last two decades (1979–1997), *Geophys. Res. Lett.*, *29*(8), 1176, doi:10.1029/2001GL013767.

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 (2002), Tropospheric aerosol optical thickness from the GOCART model and comparisons with satellite and Sun photometer measurements, *J. Atmos. Sci.*, *59*, 461–483.

Chow, J., J. Watson, D. Lowenthal, and L. Richards (2002), Comparability between $PM_{2.5}$ and particle light scattering measurements, *Environ. Monit. Assess.*, *79*(1), 29–45.

Diner, D., et al. (1998), Multi-angle Imaging Spectroradiometer (MISR) instrument description and experiment overview, *IEEE Trans. Geosci. Remote Sens.*, *36*(4), 1072–1087.

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

Falke, S., R. Husar, and B. Schichtel (2001), Fusion of SeaWiFS and TOMS satellite data with surface observations and topographic data during extreme aerosol events, *J. Air Waste Manage. Assoc.*, *51*(11), 1579–1585.

Ginoux, P., M. Chin, I. Tegen, J. Prospero, B. Holben, O. Dubovik, and S. Lin (2001), Sources and distributions of dust aerosols simulated with the GOCART model, *J. Geophys. Res.*, *106*(D17), 20,255–20,273.

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

Husar, R., et al. (2001), Asian dust events of April 1998, *J. Geophys. Res.*, *106*(D16), 18,317–18,330.

Hutchison, K. D. (2003), Applications of MODIS satellite data and products for monitoring air quality in the state of Texas, *Atmos. Environ.*, *37*(17), 2403–2412.

Ito, K., G. Thurston, A. Nádas, and M. Lippmann (2001), Monitor-to-monitor temporal correlation of air pollution and weather variables in the north-central U.S., *J. Exposure Anal. Environ. Epid.*, *11*(1), 21–32.

Jaffe, D., I. McKendry, T. Anderson, and H. Price (2003), Six 'new' episodes of trans-Pacific transport of air pollutants, *Atmos. Environ.*, *37*(3), 391–404.

Kahn, R., P. Banerjee, D. McDonald, and D. Diner (1998), Sensitivity of multiangle imaging to aerosol optical depth and to pure-particle size distribution and composition over ocean, *J. Geophys. Res.*, *103*(D24), 32,195–32,213.

Kaufman, Y. J., D. Herring, K. Ranson, and G. Collatz (1998), Earth Observing System AM 1 mission to Earth, *IEEE Trans. Geosci. Remote Sens.*, *36*(4), 1045–1055.

Kaufman, Y., D. Tanre, and O. Boucher (2002), A satellite view of aerosols in the climate system, *Nature*, *409*(6903), 215–223.

Kent, G. S., M. P. McCormick, and P. H. Wang (1994), Validation of Stratospheric Aerosol and Gas Experiment I and II satellite aerosol optical depth measurements using surface radiometer data, *J. Geophys. Res.*, *99*(D5), 10,333–10,339.

Liu, G.-R., A. Chen, T.-H. Lin, and T.-H. Kuo (2002), Applying SPOT data to estimate the aerosol optical depth and air quality, *Environ. Model. Software*, *17*(1), 3–9.

Liu, Y., J. A. Sarnat, B. A. Coull, P. Koutrakis, and D. J. Jacob (2004), 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*, D06205, doi:10.1029/2003JD003981.

Malm, W., J. Sisler, D. Huffman, R. Eldred, and T. Cahill (1994), Spatial and seasonal trends in particle concentration and optical extinction in the United States, *J. Geophys. Res.*, *99*(D1), 1347–1370.

Martin, R. V., D. J. Jacob, R. M. Yantosca, M. Chin, and P. Ginoux (2003), Global and regional decreases in tropospheric oxidants from photochemical effects of aerosols, *J. Geophys. Res.*, *108*(D3), 4097, doi:10.1029/2002JD002622.

Martonchik, J. V., D. J. Diner, R. A. Kahn, T. P. Ackerman, M. M. Verstraete, B. Pinty, and H. Gordon (1998), Techniques for the retrieval of aerosol properties over land and ocean using multiangle imaging, *IEEE Trans. Geosci. Remote Sens.*, *36*(4), 1212–1227.

Martonchik, J., D. J. Diner, K. A. Crean, and M. A. Bull (2002), Regional aerosol retrieval results from MISR, *IEEE Trans. Geosci. Remote Sens.*, *40*(7), 1520–1531.

Park, R. J., D. J. Jacob, M. Chin, and R. V. Martin (2003), Sources of carbonaceous aerosols over the United States and implications for natural visibility, *J. Geophys. Res.*, *108*(D12), 4355, doi:10.1029/2002JD003190.

Park, R. J., D. J. Jacob, B. D. Field, R. M. Yantosca, and M. Chin (2004), Natural and transboundary pollution influences on sulfate-nitrate-ammonium aerosols in the United States: Implications for policy, *J. Geophys. Res.*, *109*, D15204, doi:10.1029/2003JD004473.

Pope, C. A., III (2000), Review: Epidemiological basis for particulate air pollution health standards, *Aerosol. Sci. Technol.*, *32*(1), 4–14.

Prospero, J. (1999), Long-term measurements of the transport of African mineral dust to the southeastern United States: Implications for regional air quality, *J. Geophys. Res.*, *104*(D13), 15,917–15,927.

Rao, V., N. Frank, A. Rush, and F. Dimmick (2002), Chemical speciation of $PM_{2.5}$ in urban and rural areas, paper presented at Symposium on Air Quality Measurement Methods and Technology, Air Waste Manage. Assoc., San Francisco, Calif.

Samet, J. M., F. Dominici, F. C. Curriero, I. Coursac, and S. L. Zeger (2000), Fine particulate air pollution and mortality in 20 U.S. cities, 1987–1994, *N. Engl. J. Med.*, *343*(24), 1742–1749.

Schwartz, J., D. Dockery, and L. Neas (1996), Is daily mortality associated specifically with fine particles?, *J. Air Waste Manage. Assoc.*, *46*(10), 927–939.

Sifakis, N., and P.-Y. Deschamps (1992), Mapping of air-pollution using SPOT satellite data, *Photogramm. Eng. Remote Sens.*, *58*(10), 1433–1437.

Turpin, B., and H.-J. Lim (2001), Species contributions to $PM_{2.5}$ mass concentrations: Revisiting common assumptions for estimating organic mass, *Aerosol Sci. Technol.*, *35*(1), 602–610.

Wallace, L. (2000), Correlations of personal exposure to particles with outdoor air measurements: A review of recent studies, *Aerosol. Sci. Technol.*, *32*(1), 15–25.

Wang, J., and S. A. Christopher (2003), Intercomparison between satellite-derived aerosol optical thickness and $PM_{2.5}$ mass: Implications for air quality studies, *Geophys. Res. Lett.*, *30*(21), 2095, doi:10.1029/2003GL018174.

Wang, J., S. A. Christopher, J. S. Reid, H. Maring, D. Savoie, B. N. Holben, J. M. Livingston, P. B. Russell, and S.-K. Yang (2003), GOES 8 retrieval of dust aerosol optical thickness over the Atlantic Ocean during PRIDE, *J. Geophys. Res.*, *108*(D19), 8595, doi:10.1029/2002JD002494.

West, J., A. Ansari, and S. Pandis (1999), Marginal $PM_{2.5}$: Nonlinear aerosol mass response to sulfate reductions in the eastern United States, *J. Air Waste Manage. Assoc.*, *49*(12), 1415–1424.

Yu, H., R. E. Dickinson, M. Chin, Y. J. Kaufman, B. N. Holben, I. V. Geogdzhayev, and M. I. Mishchenko (2003), Annual cycle of global distributions of aerosol optical depth from integration of MODIS retrievals and GOCART model simulations, *J. Geophys. Res.*, *108*(D3), 4128, doi:10.1029/2002JD002717.

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