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Using aerosol optical thickness to predict ground-level $PM_{2.5}$ concentrations in the St. Louis area: A comparison between MISR and MODIS

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Abstract

Using two general linear regression models, we compared the ability of the aerosol optical thickness (AOT) retrieved by the Multiangle Imaging SpectroRadiometer (MISR) and the Moderate Resolution Imaging Spectroradiometer (MODIS) to predict ground-level $PM_{2.5}$ concentrations in St. Louis, MO and its surrounding areas . The models included meteorological parameters obtained from the National Oceanic and Atmospheric Administration (NOAA)'s Rapid Update Cycle (RUC20) model as covariates. Both MISR and MODIS AOT values were highly significant predictors of $PM_{2.5}$ concentrations. The MISR and MODIS models have overall comparable predictability of ground-level $PM_{2.5}$ concentrations. The MISR model explained a slightly greater percentage (62%) of the variability in $PM_{2.5}$ concentrations than the MODIS model (51%), and thus was a better fit. Over the entire data range, the MISR model underpredicts $PM_{2.5}$ concentrations by approximately 12%, whereas the MODIS model underpredicts $PM_{2.5}$ concentrations by approximately 18%. This underestimation occurred primarily at higher $PM_{2.5}$ concentrations in both models. The regression coefficients from two models were highly comparable, suggesting that combining MISR and MODIS AOT data might benefit from the higher predicting accuracy of MISR and the better spatial coverage of MODIS. The newly developed particle size/shape indicators in MISR and MODIS aerosol product did not significantly improve our ability to predict $PM_{2.5}$ concentrations using AOT measurements. Finally, using hourly $PM_{2.5}$ concentrations did not seem to improve its association with AOT for the current study region.

Keywords: MISR; MODIS; Aerosol optical thickness; AOT; $PM_{2.5}$; Particulate matter; Statistical analysis; General linear model; Terra satellite; St. Louis; Particle health effects

1. Introduction

Exposure to fine particulate matter ($PM_{2.5}$, particles smaller than 2.5 μm in aerodynamic diameter) has been associated with adverse health effects such as mortality, asthma, and chronic obstructive pulmonary disease (COPD) (Pope & Dockery, 1999; Rojas-Bracho et al., 2000; Schwartz et al., 1996). Air pollution epidemiologic studies, such as the highly influential Six Cities study (Dockery et al., 1993) and American Cancer Society (ACS) study (Pope et al., 1995) linked study participants with existing pollution level measurements from ground-level monitoring stations in order to examine the association between particle air pollution and its effect on public health. A major limitation faced in these and similar studies is the ability to reliably capture particle exposure for the study population. The current $PM_{2.5}$ monitoring network, which is overseen by the United States Environmental Protection Agency (USEPA) and operated by the state, local, and tribal air pollution control agencies, includes approximately 1000 Federal Reference Method (FRM) and 200 continuous $PM_{2.5}$ monitors as well as 200 $PM_{2.5}$ speciation monitors. Even with this large number of monitors, many rural and suburban regions as well as some smaller urban areas are not covered. In these studies, an average $PM_{2.5}$ pollution level often had to be assigned to the entire community within a certain distance of each monitor. The lack of spatial coverage and misclassification of $PM_{2.5}$ exposure can severely reduce the statistical strength of these studies and bias the results towards the null hypothesis of no significant association between pollution and adverse health outcomes. Statistical interpolation based solely on $PM_{2.5}$

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concentrations measured at central stations may extend the spatial coverage but can also introduce substantial uncertainty.

Since 1999, the National Aeronautics and Space Administration (NASA) has launched a series of satellite sensors, including the Multiangle Imaging SpectroRadiometer (MISR) (Diner et al., 2002) and the Moderate Resolution Imaging Spectroradiometer (MODIS) (Remer et al., 2005). Both aboard the Terra satellite, these two sensors can measure particle abundance and composition with nearly global coverage at moderate spatial resolutions. Particle information retrieved by satellite sensors may be suitable for monitoring $PM_{2.5}$ concentration spatial and temporal trends over large geographical areas. As a rapidly developing new technology, the capabilities and products of satellite remote sensing are generally unfamiliar to the PM monitoring and public health research communities. To date, there are a limited number of published studies that apply satellite retrieved particle properties to quantitatively indicate ambient $PM_{2.5}$ concentration levels. One study found a linear relationship (correlation coefficient $r=0.7$) between MODIS AOT and $PM_{2.5}$ concentrations measured by tapered-element oscillating microbalances (TEOM) in Jefferson County, Alabama (Wang & Christopher, 2003). Another study showed mixed results when comparing MODIS AOT with 24-h PM_{10} (particles smaller than 10 µm in aerodynamic diameter) concentrations, where a linear relationship was found ($r=0.82$) at one site in Italy but not at two other sites in Los Angeles and Beijing (Chu et al., 2003). MODIS data has also been examined as an indicator of air quality on a regional scale (Engel-Cox et al., 2004). It was found that the correlation between MODIS AOT and 24-h $PM_{2.5}$ concentration was stronger in the eastern U.S. than in the western U.S.

Most studies examining the association between particle concentrations and AOT applied simple statistical tools such as linear correlation and simple linear regression with AOT as the only $PM_{2.5}$ concentration predictor. These techniques are not sufficient to describe the complex relationship between AOT and $PM_{2.5}$. In a previous study (Liu et al., 2005), we showed that the association between $PM_{2.5}$ concentration and MISR AOT can be strongly influenced by meteorological, geographical, and seasonal conditions. Of particular importance is the aerosol vertical distribution, which provides a link between ground-level $PM_{2.5}$ concentrations, of major interest for air quality, and the total-column AOT measured by MISR. Since these factors show such importance, they should be considered when relating satellite AOT measurements to $PM_{2.5}$ concentration.

MISR and MODIS have different instrument designs and retrieve aerosol optical properties using different algorithms. MISR uses the systematic change in atmosphere and surface contributions to top-of-atmosphere reflection signals observed by its nine view angles at four wavelength bands to retrieve aerosol information over land. MISR AOT and other aerosol information are calculated based on a rich set of aerosol mixture models and reported at 17.6 km resolution. It has a global

Fig. 1. The study region and the distribution of EPA $PM_{2.5}$ monitors, urban areas, and interstate highways. The geographic location of the domain is shown in the upper left corner. The domain is marked as the thick grey square. The St. Louis $PM_{2.5}$ non-attainment area designated by EPA is the area with grey hatch.

coverage between two and nine days depending on latitude. MODIS uses prescribed spectral relationships between its blue (470 nm), red (660 nm), and shortwave infrared (2.13 μm) wavelength bands to retrieve aerosol information over land. MODIS AOT and other aerosol information are calculated based on simplified assumptions of particle composition, and reported at 10 km resolution. MODIS has a global coverage of 1–2 days. Both sensors pass over the northern hemisphere between 10:00 am and noon. Since they are both aboard the NASA Terra satellite, data generated by these two sensors may complement each other with regard to measurement accuracy and spatial coverage. We are not aware of any study that compares the capability of MISR and MODIS AOT to predict $PM_{2.5}$ concentrations in a similar modeling setting. In the current analysis, we develop two general linear regression models using MISR and MODIS AOT, respectively as the main predictor of $PM_{2.5}$ concentration, and a few assimilated meteorological parameters as covariates. Subsequently, we compare the performance of the two models and examine their similarities. We evaluate the impact of newly developed MISR and MODIS particle size parameters on model performance as well as the influence of sample size on predictor stability. Finally, we examine whether the temporal resolution of $PM_{2.5}$ concentration has any impact on model performance.

2. Data collection and integration

The study region for the current analysis is an approximately 300 km by 300 km, relatively flat region covering southwestern Illinois and eastern Missouri (latitude range [37.2°N, 40.0°N], longitude range [89.5°W, 93.0°W]) (Fig. 1). The region contains the city of St. Louis and the surrounding counties, which EPA designated as a $PM_{2.5}$ non-attainment area in April 2005. According to the 2000 Census, there are 29 communities with a population over 20,000 in this region, and the St. Louis metropolitan area is a major industrial center with a population of 2.5 million. We chose this study domain to represent various $PM_{2.5}$ pollution levels found in large metropolitan areas as well as surrounding suburban and rural areas. There are more than 20 $PM_{2.5}$ EPA monitors as well as the EPA St. Louis PM Supersite in the region, measuring both general population exposure in urban and suburban settings and environmental backgrounds. Our study region is climatologically affected by a wide range of synoptic weather patterns and is free of localized influences from the Great Lakes, the oceans, the Gulf of Mexico, and mountains. The relatively flat inland terrain is likely to simplify the atmospheric boundary layer structure and improve the quality of assimilated meteorological parameters used in the analysis.

2.1. MISR and MODIS aerosol data

We downloaded MISR aerosol data (version 15) covering our study region for 2003 from the Atmospheric Sciences Data Center at NASA Langley Research Center (http://eosweb.larc.nasa.gov). We extracted AOT at 558 nm (MISR parameter name: RegBestEstimateSpectralOptDepth) and the AOT fractions by particle shape (MISR parameter name: RegBestEstimateSpectralOptDepthFraction, fourth and fifth components). The AOT fractions by particle shape give the percent contribution of spherical particles such as particles of anthropogenic origin and sea salt particles to the total MISR AOT, and the percentage contribution of nonspherical particles such as mineral dust and possibly thin cirrus to the total MISR AOT (these two percentages add up to 100%). Because there are no major local dust emission sources in our study region, transported dust is probably the major contributor to the nonspherical fraction of MISR AOT in the St. Louis area. Since transported dust plumes tend to move above the boundary layer, they are likely irrelevant to ground level $PM_{2.5}$ concentrations in our study region. Therefore, the spherical fraction of MISR AOT, called MISR partial AOT hereinafter, might be a better predictor of ground-level concentrations as compared to the total MISR AOT.

We downloaded 2003 MODIS aerosol data (collection 4) from the Earth Observing System Data Gateway at the Goddard Space Flight Center (http://delenn.gsfc.nasa.gov/~imswww/pub/imswelcome). We extracted AOT at 550 nm (MODIS parameter name: Optical_Depth_Land_And_Ocean). Over land, the MODIS AOT fraction of fine mode particles (MODIS parameter name: Optical_Depth_Ratio_Small_Land) is a rough estimate based on the ratio of MODIS path radiances at 660 nm and 470 nm (Remer et al., 2005). We calculated the MODIS partial AOT by including only the AOT fraction due to fine mode particles.

In the analysis described later in this paper, we evaluated whether the MISR and MODIS partial AOT values could better predict ground level $PM_{2.5}$ than the total AOT values. The quality control flags in both the MISR and MODIS products were used to prevent low-quality data from entering the data processing (Wedad Abdou and Lorraine Remer, personal communication).

2.2. EPA $PM_{2.5}$ measurements

The $PM_{2.5}$ data in our study region for 2003 were downloaded from the EPA's Air Quality System Technology Transfer Network (http://www.epa.gov/ttn/airs/airsaqs). We collected 24-h average $PM_{2.5}$ mass concentrations from 22 FRM monitors. We chose 24-h average $PM_{2.5}$ concentration as the dependant variable of our models because it is the national ambient air quality standard for $PM_{2.5}$ and there are five times more FRM monitors nationwide than the continuous monitors. Establishing the association between AOT and 24-h $PM_{2.5}$ concentration is more appropriate for pollution monitoring and health effect studies. Nonetheless, hourly $PM_{2.5}$ concentrations were collected from two continuous monitors in order to study the impact of $PM_{2.5}$ temporal variability on model performance. The spatial distribution of these monitors is shown in Fig. 1.

2.3. Meteorological data

The meteorological fields used in the current analysis are generated by the Rapid Update Cycle (RUC) model. The RUC model is a high-frequency operational weather forecast and data assimilation system developed by the Earth System Research Laboratory, RUC development group at NOAA (Benjamin et al., 2004a,b). RUC data are archived at the Atmospheric Radiation Measurement (ARM) Program Climate Research Facility Data Archive (http://www.archive.arm.gov/). The third generation of the RUC model (RUC20), which covers the year 2003, integrates observations from surface reporting stations and buoys, rawinsondes and special dropwinsondes, wind profilers, commercial aircraft, and satellites into a numerical forecast system that makes short-term weather forecasts at 1-h frequency. The 1-h frequency of the RUC model enabled us to compute meteorological parameters including the boundary layer mixing height (PBL), surface wind speed and wind direction at 10 m above the ground, mean lower-troposphere relative humidity (RH) (from surface to approximately 820 hPa), and surface air temperature at 2 m above the ground between 10 am and noon corresponding to the MISR/MODIS overpass time. The original spatial resolution of RUC20 outputs is 20 km. For the convenience of data storage and computational expediency, we downloaded the 40 km resolution RUC20 meteorological parameters averaged from the original 20 km resolution outputs. Given the flat terrain in our study region, we do not expect any substantial loss of information due to this spatial averaging.

2.4. Data integration

In order to combine the RUC and satellite data, we averaged hourly values of the RUC20 meteorological parameters including PBL, surface wind speed and direction, surface air temperature, and relative humidity between 10 am and noon local time. The EPA $PM_{2.5}$ concentrations were then matched with the averaged RUC meteorological parameters from the RUC grid cell within which the EPA monitor falls. The global validation of MISR AOT against observations from the Aerosol Robotic Network (AERONET) used the average of 3×3 17.6 km-MISR pixels centered at each AERONET site (~26–35 km radius), along with the central pixel alone, to account for the different sampling methods of MISR AOT (instantaneous measurements) and AERONET AOT (average of a 2-h window centered on the MISR overpass time) (Kahn et al., 2005). For the same reason, the global validation of MODIS AOT over land against AERONET took the average of 5×5 10 km-MODIS pixels centered at each AERONET site (~25–30 km radius) (Chu et al., 2002). Since the current analysis is similar to these validation studies in that regional measurements are matched with point measurements, we applied a 30 km search radius around each EPA monitor for averaging both MISR and MODIS AOT pixels. Depending on where an EPA monitor is located, this search radius covers the center coordinates of 8 to 12 MISR pixels or 27 to 32 MODIS pixels. Applying identical search radii to both MISR and MODIS aerosol data facilitates the comparison of data from the two sensors. All MISR or MODIS AOT pixels that fall within the search radius around the EPA monitor were collected. For each day with a $PM_{2.5}$ concentration measurement, we calculated the mean and standard deviation of MISR and MODIS AOT. To avoid determining the AOT value from a single pixel, we required that there be at least two valid MISR or MODIS AOT measurements per day within each search radius. In addition, to reduce the likelihood of including spurious AOT pixels in the analysis, we removed the MISR and MODIS AOT data records whose coefficient of variation (standard deviation divided by mean AOT) are greater than 0.5. On average, there are 6 MISR AOT measurements and 13 valid MODIS AOT measurements in each search radius on each day. Finally, we matched both the MISR and MODIS AOT data with the EPA $PM_{2.5}$ concentrations and the time-averaged RUC20 meteorological parameters by date. We refer to these two final datasets as the MISR dataset (sample size = 359) and the MODIS dataset (sample size = 1779) hereinafter.

3. Methods

Since the MISR spherical particle fractions have not been fully validated and the MODIS fine mode fractions are currently qualitative, we used the original MISR and MODIS AOT values to develop our general linear models and later examined the impact of using the partial AOT values in a subsequent regression analysis. The average temporal spacing between two consecutive matched EPA-MISR observations is 10 days or longer. The average temporal spacing between two consecutive matched EPA-MODIS observations is three days at the monitors with a daily sampling schedule and longer at the monitors implementing every three or six day sampling schedules. Autocorrelation coefficients calculated at each of the six monitors with daily sampling schedules drop below 0.2 for a lag of three days or longer (Fig. 2). Therefore, we conclude that the autocorrelation among observations at each monitor is negligible, and the persistence time scale for the aerosol events in the St. Louis area is likely no greater than two days.

We built a general linear regression model (GLM) using matched RUC meteorological parameters and either MISR or MODIS AOT values as predictors of daily $PM_{2.5}$ concentration:

$$[PM_{2.5}] = e^{\beta_0+\beta_1(\mathrm{TEMP})+\beta_2(\mathrm{DIR})+\beta_3(\mathrm{NONSPRING})} \times \mathrm{WIND}^{\beta_4} \times \mathrm{PBL}^{\beta_5} \times \mathrm{AOT}^{\beta_6+\beta_7\times\mathrm{NONSPRING}} \quad (1)$$

The dependent variable, $[PM_{2.5}]$, represents the 24-h $PM_{2.5}$ concentration measured at the EPA monitors. The predictor variables include surface air temperature (TEMP), surface wind speed (WIND), mixing height (PBL), either MISR or MODIS

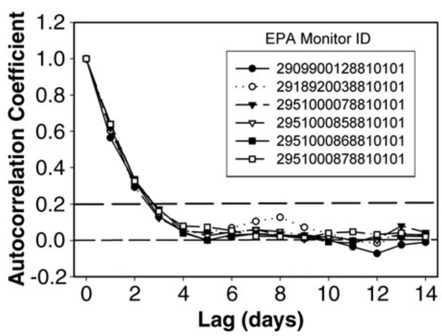

Fig. 2. Correlogram of 24-h $PM_{2.5}$ concentrations measured at six monitors with daily sampling schedules.

AOT (AOT), and two categorical variables: wind direction indicator (DIR), and seasonal indicator (NONSPRING). Based on our preliminary analysis and due to the relatively small sample size of the MISR dataset, seasons are clustered into a dichotomous variable, i.e., NONSPRING = 0 for spring season, and NONSPRING = 1 otherwise. MODIS data exhibits a similar seasonal pattern so we define the seasonal indicator in the MODIS dataset as we do in the MISR dataset, which also assures consistency for model comparison. We also allow the association of MISR or MODIS AOT with $PM_{2.5}$ concentration to be modified by the seasonal indicator NONSPRING. In Eq. (1), AOT, PBL, and WIND are assumed to have power law functional forms to account for their nonlinear relationships with $PM_{2.5}$ concentration. TEMP takes an exponential form so that the impact of temperature fluctuation (both positive and negative) on the association between $PM_{2.5}$ concentration and AOT could be modeled. For easier interpretation of the regression coefficients, we center TEMP by subtracting its annual mean. Since WIND and PBL must be positive, we center these two variables by dividing them by their respective annual means. The difference in the annual mean values between the MISR dataset and the MODIS dataset is less than 5% for TEMP and WIND, and 8% for PBL. The description of these predictor variables is summarized in Table 1. To account for the particle size growth with increased RH, we derived nonlinear light scattering growth curves (results not shown) using data from Day and Malm (2001) and Day et al. (2000) to modify MISR and MODIS AOT values. However, this process did not improve model performance or change regression coefficients significantly. As a result, RH was not included in the final model.

Taking log on both sides gives the linear form of Eq. (1):

$$\begin{aligned}\ln[PM_{2.5}] = {} & \beta_0 + \beta_1(\mathrm{TEMP}) + \beta_2(\mathrm{DIR}) \\ & + \beta_3(\mathrm{NONSPRING}) + \beta_4 \ln(\mathrm{WIND}) \\ & + \beta_5 \ln(\mathrm{PBL}) + \beta_6 \ln(\mathrm{AOT}) + \beta_7 \\ & \times \mathrm{NONSPRING} \times \ln(\mathrm{AOT}) \qquad (2)\end{aligned}$$

The log-transformations reduce the skewness in the data distribution of $PM_{2.5}$, AOT, PBL, and WIND, consequently

Table 1
Definitions of predictor variables used in Eqs. (1) and (2)

Name	Property	Unit	Description
AOT	Continuous	Dimensionless	MISR or MODIS total AOT
TEMP	Continuous	Dimensionless	Surface level air temperature measured at 2 m above the ground centered by removing the sample mean.
WIND	Continuous	m/s	Surface wind speed measured at 10 m above the ground centered by subtracting the sample mean.
PBL	Continuous	km	Mixing height centered by being divided by the sample mean.
DIR	Categorical		Wind direction (blowing from). Level = East, South, West, and North
NONSPRING	Categorical		Level = 1 if sampling date is between March and May, = 0 otherwise

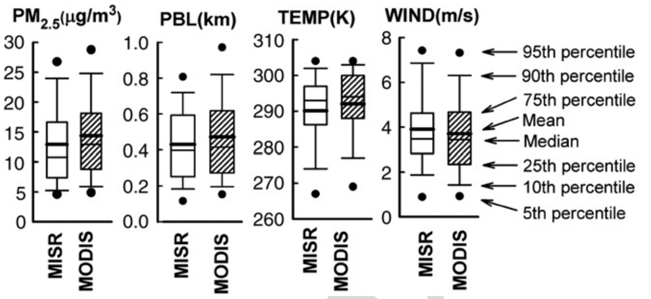

Fig. 3. Boxplots showing the distribution (5th, 10th, 25th, median, 75th, 90th, and 95th percentiles) of $PM_{2.5}$ concentrations, mixing height (PBL), surface wind speed (WIND), and surface air temperature (TEMP). The means values are shown as the thick solid line in the plots.

increasing the accuracy of the estimated regression coefficients and their standard errors. β_1 through β_6 are regression coefficients for predictor variables TEMP, DIR, NONSPRING, ln(WIND), ln(PBL) and ln(AOT), respectively. Since we include an interaction term between ln(AOT) and NONSPRING in order to model the changing association between $PM_{2.5}$ concentration and AOT by season, model intercept is β_0 is in springtime, and ($\beta_0+\beta_3$) during the rest of the year. In addition, the regression coefficient for ln(AOT) is β_6 in springtime, and ($\beta_6+\beta_7$) during the rest of the year. For Eq. (1) to be valid, we assume that the air within the boundary layer is well mixed, and the vertical distribution of particles above the boundary layer is relatively smooth. This assumption is commonly satisfied over land during daytime when there is sufficient surface heating (Holton, 2004). Since there are no AOT retrievals during very cloudy days and our study region is not close any large water bodies, the conditions encountered in the current analysis when this assumption tends to break down is expected to be rare. The various meteorological parameters and the seasonal variable included in the models are all considered as indicators of the change of particle vertical profile and composition. When there are discontinuous particle intrusion layers above the boundary layer, the association between ground-level $PM_{2.5}$ concentration and satellite-derived, total-column AOT will be weakened and a measured or model-simulated particle vertical profile is required to separate the AOT proportion in the boundary layer (Liu et al., 2004a).

We evaluated model performance by comparing predicted $PM_{2.5}$ concentrations with EPA observations before and after reversing the log transformation. All statistical analyses were conducted using the SAS system (SAS Institute Inc., Cary, NC).

4. Results and discussion

4.1. Summary statistics of data

Summary statistics of $PM_{2.5}$ concentrations as well as the RUC20 meteorological parameters in their original values (i.e., before centering) in the MISR and MODIS datasets are presented in Fig. 3. Overall, the mean and median values of $PM_{2.5}$, PBL, and TEMP happen to be slightly higher in the MODIS dataset than in the MISR dataset. The mean and median

Table 2
Summary statistics of MISR and MODIS total AOT and partial AOT data

	Total AOT		Partial AOT	
	MISR	MODIS	MISR	MODIS
N	359	1779	359	1779
Mean	0.14	0.31	0.13	0.30
Min	0.03	0.02	0.02	0.01
5th percentile	0.03	0.08	0.03	0.08
10th percentile	0.04	0.11	0.03	0.10
25th percentile	0.06	0.18	0.06	0.17
Median	0.10	0.30	0.10	0.28
75th percentile	0.15	0.42	0.15	0.41
90th percentile	0.29	0.55	0.26	0.55
95th percentile	0.35	0.61	0.35	0.61
Max	1.02	1.02	1.00	1.02

value of WIND in the two datasets are very similar. These variables also have comparable ranges in both datasets. However, MODIS AOT is consistently higher than MISR AOT throughout the year (Table 2). Remer et al. (2005) reported a 53–54% positive bias in MODIS AOT over the U.S. when compared with AERONET AOT measurements which are generally considered to be very accurate (Smirnov, 2000). Abdou et al. (2005) observed a similar discrepancy between MODIS AOT over land, and MISR and AERONET AOT. This bias could be manifested in our dataset and may explain the observed difference between MISR and MODIS AOT. As shown in Table 2, the partial AOT values (spherical fraction for MISR and fine-mode fraction for MODIS) have distributions very similar to their respective total AOT values. This indicates that in our dataset coarse mode particles contribute little to the total AOT retrieved by either MISR or MODIS.

4.2. Comparison of the overall model performance

The MISR and MODIS datasets are fit to Eq. (2) separately. Hereinafter, we call the model expressed in Eq. (2) separately for each satellite sensor: the MISR model when MISR AOT is a predictor and the MODIS model when MODIS AOT is a predictor. Overall, the MISR model explains more variability in daily $PM_{2.5}$ concentrations ($R^2=0.62$) than the MODIS model ($R^2=0.51$) (Table 3). All the predictors in the MODIS model are highly significant at the $\alpha=0.001$ level, whereas ln(PBL) in the MISR model is not significant at the $\alpha=0.05$ level. Standard diagnostics for linear regression models, such as the jackknife residuals and the Cook's Distances do not indicate any outliers or substantial fluctuation of variances in either model. The MISR model predicts an annual mean $PM_{2.5}$ concentration of 12.3 μg/m^3, approximately 5% lower than the annual mean of EPA observations (12.9 μg/m^3). The MODIS model predicts an annual mean $PM_{2.5}$ concentration of 13.3 μg/m^3, approximately 7% lower than the annual mean of EPA observations (14.2 μg/m^3). After we reverse the log transformation by taking the exponential of the fitted ln[$PM_{2.5}$] values and compared with EPA observations, the root-mean-square error (RMSE) is 4.5 μg/m^3 for the MISR model, and 5.6 μg/m^3 for the MODIS model. Given the mean EPA $PM_{2.5}$ concentrations, we calculate the relative error (RMSE divided by mean $PM_{2.5}$ concentration) to be 35% for the MISR model and 39% for the MODIS model.

Regression analysis using the fitted and the observed log-transformed $PM_{2.5}$ concentrations (ln[$PM_{2.5}$] in Eq. (2)) shows an approximately 2% underestimation for both the MISR model and the MODIS model, i.e., fitted ln[$PM_{2.5}$]$=0.98\times$observed ln[$PM_{2.5}$]. After reversing the log transformation, we observe that the MISR model underpredicts $PM_{2.5}$ concentrations by approximately 12% and the MODIS model underpredicts $PM_{2.5}$ concentrations by approximately 18% over the data range (Fig. 4). Underestimation occurs primarily at higher $PM_{2.5}$ concentrations in both models. This phenomenon may be attributed to the flattening effect of log transforming both the dependent ($PM_{2.5}$) and some of the predictor variables. Although log-transformation corrects the non-normality of

Table 3
Estimated regression coefficients of the MISR model (sample size $N=359$) and MODIS model (sample size $N=1779$) using total AOT

Parameter	Total MISR AOT ($R^2=0.62$)			Total MODIS AOT ($R^2=0.51$)		
	Estimate	Standard error	CI factor	Estimate	Standard error	CI factor
Intercept						
Spring	3.12***	0.12	22.8	2.48***	0.06	12.1
Non-Spring	3.71***	0.19	40.9	2.90***	0.08	17.6
ln(AOT)						
Spring	0.19***	0.05	$AOT^{0.19}$	0.20***	0.04	$AOT^{0.20}$
Non-Spring	0.44***	0.08	$AOT^{0.44}$	0.45***	0.06	$AOT^{0.45}$
ln(PBL)	0.01	0.04	$PBL^{0.01}$	−0.14***	0.02	$PBL^{-0.14}$
ln(WIND)	−0.33***	0.04	$WIND^{-0.33}$	−0.17***	0.02	$WIND^{-0.17}$
TEMP	−0.005*	0.002	$e^{-0.005\times TEMP}$	0.005***	0.001	$e^{0.005\times TEMP}$
Wind direction						
From East	−0.04	0.06	0.94	0.14***	0.04	1.15
From North	−0.02	0.05	0.98	0.11***	0.03	1.12
From South	0.44***	0.05	1.55	0.44***	0.02	1.55
From West[a]	0.00		1.00	0.00		1.00

* Significant at the $\alpha=0.05$ level.
** Significant at the $\alpha=0.01$ level.
*** Significant at the $\alpha=0.001$ level.
[a] Reference level in the categorical predictor.

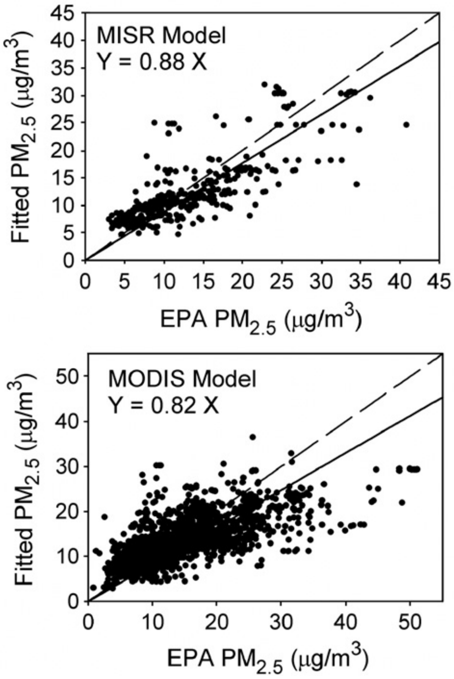

Fig. 4. Scatterplots of fitted $PM_{2.5}$ concentrations obtained from the MISR and MODIS general linear models vs. EPA observations. Solid line represents simple linear regression with intercept excluded. Dashed line represents 1:1 reference.

$PM_{2.5}$ data and gives more accurate standard errors of the regression coefficients, it constrains the relation between $PM_{2.5}$ concentrations and each of these predictors to an exponential form. Reversing the log-transformation tends to yield a flatter regression slope, resulting underestimation at higher concentrations.

The overall model performance in the current analysis is higher than similar previous studies. Stepwise regression analysis indicates that MISR AOT explains approximately 35% of the variability in 24-h $PM_{2.5}$ concentrations, as compared to 18% in Liu et al. (2005). Factors that might contribute to this improvement include better MISR data quality (version 15 used in the current analysis vs. version 12 in Liu et al. (2005)), and the potentially simpler particle vertical profiles encountered in our study region as compared to the coastal regions in Liu et al. (2005). MODIS AOT explains approximately 27% of the variability in 24-h $PM_{2.5}$ concentrations, which is comparable with the correlation coefficients between 0.45 and 0.56 previously reported in St. Louis (Engel-Cox et al., 2004). The correlation coefficients between the fitted and the observed $PM_{2.5}$ concentrations are 0.78 and 0.69 for the MISR and the MODIS datasets, respectively, which make the current models better tools in quantitatively estimating $PM_{2.5}$ exposure as compared to simple linear models using AOT as the only predictor.

It should be noted that EPA measurements of 24-h $PM_{2.5}$ concentrations are assumed to be accurate when we analyze the overall model performance. In fact, the absorption or desorption of semi-volatile compounds such as nitrates and carbonaceous species can sometimes introduce substantial positive or negative biases to filter based $PM_{2.5}$ mass measurements. This uncertainty may account for part of the unexplained variability. However, it is impossible to estimate or correct these potential biases caused by the semi-volatile species in our $PM_{2.5}$ concentration data so we cannot explore this matter further.

4.3. Comparison of the regression coefficients of predictor variables

We interpret the relationship between $PM_{2.5}$ concentration and AOT as well as the influences of the other predictors in the models on this relationship primarily using concentration impact factors (CI factors) (Table 3). The CI factor of a log-transformed continuous variable is this variable to the power of its regression coefficient. For example, PBL has a regression coefficient of −0.14 in the MODIS model. Its corresponding CI factor is $PBL^{-0.14}$, which represents the impact of centered mixing height on the association between $PM_{2.5}$ concentration and MODIS AOT. The CI factor of the non log-transformed continuous variable, i.e. TEMP, is the exponential of the regression coefficient of TEMP multiplied by the variable. For example, TEMP has a regression coefficient of −0.005 in the MISR model. Its corresponding CI factor is $e^{-0.005\times \mathrm{TEMP}}$, which represents the impact of surface temperature variation (around the annual mean) on the association between $PM_{2.5}$ concentration and MISR AOT. Since the categorical variable DIR has four levels, each of the four levels has a CI factor, calculated as the exponential of the regression coefficient for each level. Since westerly wind was chosen as the reference direction, it always has a regression coefficient of zero and a CI factor of one. The other three CI factors can be interpreted as the impact of different wind directions on the fitted $PM_{2.5}$ concentrations as compared to the reference level.

4.3.1. Model intercepts

The CI factor of the model intercept is calculated as the exponential of the estimated intercept. As mentioned previously, we allow the association between $PM_{2.5}$ concentration and AOT to vary by season. As a result, the CI factor of the MISR model intercept is 22.8 $\mu g/m^3$ in springtime and 40.9 $\mu g/m^3$ during the rest of the year. The CI factor of the MODIS model intercept is significantly smaller (12.1 $\mu g/m^3$ in springtime and 17.6 $\mu g/m^3$ during the rest of the year). Since the meteorological predictors PBL, WIND and TEMP are all centered, these CI factors have clear physical interpretations. For example, the intercept CI factor of 22.8 $\mu g/m^3$ in the MISR model means that in springtime, when mixing height, wind speed, and surface temperature are at their annual mean levels and wind blows from the west, a MISR AOT value of 1.0 corresponds to the ground-level $PM_{2.5}$ concentration of 22.8 $\mu g/m^3$. Similarly, the intercept CI factor of 12.1 $\mu g/m^3$ in the MODIS model means that in springtime, when mixing height, wind speed, and surface temperature are at their annual mean levels and wind blows from the west, a MODIS AOT value of 1.0 corresponds to ground $PM_{2.5}$ concentrations of 12.1 $\mu g/m^3$. This finding suggests that under similar conditions, MISR AOT values predict significantly higher $PM_{2.5}$ concentrations than MODIS AOT values in our study region.

4.3.2. MISR/MODIS AOT and seasonal variation

Considering the standard errors of the regression coefficients, the CI factors of MODIS AOT in springtime ($AOT^{0.20}$) and non-spring seasons ($AOT^{0.45}$) are surprisingly comparable with those of MISR AOT ($AOT^{0.19}$ in springtime, $AOT^{0.44}$ in non-spring seasons). This suggests that both MISR and MODIS AOT values have a similar relationship with $PM_{2.5}$ concentrations in the St. Louis area. However, the lower intercept of the MODIS model suggests that MODIS AOT predicts significantly lower $PM_{2.5}$ concentrations than the same level of MISR AOT. It has been previously shown that MISR and MODIS AOT are well correlated despite MODIS AOT being consistently higher over land (Abdou et al., 2005). Comparison studies with AERONET observations indicate that the uncertainties in both MISR and MODIS AOT are proportional to the magnitude of AOT values (Kahn et al., 2005; Liu et al., 2004b; Remer et al., 2005). It is likely that MODIS overestimates AOT by a consistent proportion in our study region. If a true AOT value represents a certain $PM_{2.5}$ concentration level, the same AOT value retrieved by MODIS should represent a smaller $PM_{2.5}$ concentration due to its positive bias. Given the multiplicative form of our model, the consistent positive bias in MODIS AOT could be manifested by the lower model intercepts of the MODIS model. It should be noted that despite the possible bias in MODIS AOT, the MISR and MODIS models have overall comparable predictability of ground-level $PM_{2.5}$ concentrations with the MISR model having a slightly higher R^2 and thus a better fit. As we mentioned in the introduction, MISR and MODIS complement each other due to their different instrument designs and aerosol retrieval algorithms. MODIS aerosol retrieval over land is able to achieve repeat coverage at mid-latitude in 1–2 days owing to its wide swath. Although MISR AOT appeared more accurate, it is only able to achieve repeat coverage at mid latitudes in 5–7 days. The smaller MISR data sample size, which probably causes the instability of some predicators in the MISR model shown later in this paper, is a direct reflection of this difference in spatial coverage. The comparable regression coefficients give hope that the integration of MISR and MODIS AOT data will benefit from the better spatial coverage of MODIS and higher sensitivity of MISR.

Although AOT is positively associated with $PM_{2.5}$ concentration throughout the year, the AOT CI factors are significantly greater during non-spring seasons than in springtime. This means that for the same AOT values, MISR and MODIS both predict larger $PM_{2.5}$ concentrations during non-spring seasons than in springtime. In our previous analysis, we used season as a 4-level categorical variable and found the CI factor for springtime is significantly lower than for the rest of the year over eastern U.S. states (Liu et al., 2005), similar to the finding in this analysis. It has been shown by both modeling analyses and observations that there is substantial long-range transport of dust and pollution aerosols in the free troposphere from east and southeast Asia in springtime (Prospero et al., 2002; VanCuren & Cahill, 2002). In addition, deep convective systems in the spring over the central and south western U.S. can also increase vertical mixing and transport boundary layer pollutants to the free troposphere (Talbot et al., 1998). Both phenomena tend to increase the proportion of particles in the free troposphere. Consequently, the same AOT values would correspond to lower ground level $PM_{2.5}$ concentrations in springtime than during the rest of the year.

4.3.3. Meteorological conditions

Surface level wind speed (WIND) is a highly significant predictor in both models, and the negative sign of its regression coefficient in both models shows that MISR or MODIS AOT predicts lower $PM_{2.5}$ concentrations at higher wind speed (Table 3). Preliminary analysis of the RUC20 data showed that surface level wind speed is strongly correlated with the average wind speed in the lower troposphere. Therefore, the negative sign of its regression coefficient may be attributed to the fact that greater turbulence caused by higher wind speed enhances vertical mixing. Surface wind direction (DIR) is also highly significant in both models. Each wind direction has a different impact on the MODIS model intercept from the reference wind direction (westerly winds) whereas only one wind direction (southerly winds) has a significantly different impact on the MISR model intercept from the reference direction. In both models, the corresponding $PM_{2.5}$ concentrations are approximately 30%–50% higher when wind blows from the south than when it blows from other directions. In 2002, the EPA St. Louis PM Supersite observed dramatically increased SO_2 concentrations when the winds were out of the southwest (Douglas Dockery, personal communication). Because SO_2 is the precursor of sulfate particles and an important indicator of large combustion source emissions, their observations together with our analysis suggest that perhaps a larger proportion of particles in the air stay near the surface when the wind blows from the south. Since there are no major point sources of $PM_{2.5}$ or large communities in the southern part of our domain, the elevated $PM_{2.5}$ level related to southerly winds might be caused by the low-level jet in lower atmosphere that carries polluted air from Texas towards the Great Lakes (Court, 1974).

Surface air temperature (TEMP) is a significant predictor in both models although its impact on the predicted $PM_{2.5}$ concentrations is relatively small. Using the maximum and minimum TEMP values, we calculate the CI factors to be between 0.92 and 1.13 for the MISR model, and between 0.88 and 1.07 for the MODIS model. The signs of the regression coefficients differ in the two models (Table 3). Higher air temperature accelerates the generation of secondary particles near the surface, causing a higher proportion of particle mass in the mixed layer, which would explain the positive regression coefficient of TEMP in the MODIS model. Analysis of the $PM_{2.5}$ speciation data collected in the study region (results not shown) suggests that there is a positive correlation between the ratio of sulfate proportion of $PM_{2.5}$ mass to organic carbon (OC) proportion of $PM_{2.5}$ mass and air temperature. Since sulfate particles tend to have larger extinction coefficients than OC particles (Chin et al., 2002), the same AOT values would correspond to less total $PM_{2.5}$ mass when there are more sulfate particles in the air. This correlation might to some extent be linked to the negative regression coefficient of TEMP in the MISR model. More importantly, model stability analysis, which will be presented later in this paper, shows that TEMP becomes very unstable at smaller sample sizes. This suggests that we could have observed a negative regression coefficient of TEMP in the MISR model by chance.

Mixing height (PBL) has a significant impact on the association between $PM_{2.5}$ concentration and AOT in the MODIS model. Its CI factor ($PBL^{-0.14}$) means that for the same number of particles trapped in the boundary layer, a lower mixing height causes a higher ground level $PM_{2.5}$ concentration, which is what we expected to see. The regression coefficient of ln(PBL) is not statistically significant in the MISR model. This result differs from our previous analysis using a larger dataset covering the eastern U.S. (Liu et al., 2005), where we found a strong and significant impact of PBL on the association between $PM_{2.5}$ concentration and MISR AOT. It might be related to the issue of predictor stability, which we will discuss in Section 4.4.2.

Surface level wind speed and direction are important indicators of horizontal movement and mixing of air masses. Surface air temperature and boundary layer height are closely related to the convective mixing in lower atmosphere and the generation of secondary particles. Their significance as predictors in the current models reflects their ability to modify and enhance the association between AOT and $PM_{2.5}$ concentrations. Stepwise regression analysis indicates that together the meteorological parameters explain approximately 23% of the variability in $PM_{2.5}$ concentrations, as compared to slightly over 15% reported in Liu et al. (2005). Factors causing the better predictability might include the choice of a flat inland domain and the higher spatial resolution of the RUC20 data.

4.4. Other factors affecting model performance

In our previous work, we suggested that the performance of a statistical model linking AOT and $PM_{2.5}$ concentration could be improved in the following ways: 1) information about particle size distribution and composition, 2) information about particle vertical distribution, and 3) the accuracy, resolution and coverage of input data (Liu et al., 2005). In the following sections, we discuss the potential influence of particle size distribution, data sample size, and the temporal resolution of $PM_{2.5}$ concentration.

4.4.1. Regression analysis using partial AOT

Although $PM_{2.5}$ concentration measures fine-mode particles, AOT represents the light scattering capability of all particles in the atmospheric column. When the contribution of coarse particles to AOT is large, the association between AOT and $PM_{2.5}$ concentrations could be weakened. To evaluate the newly developed particle mode indicators in the MISR and MODIS aerosol data, we replace total MISR and MODIS AOT with their respective partial AOT values and fit the models again (Table 4). The model RMSE and relative errors remain the same, indicating no improvement in model performance. The regression coefficients of the intercepts and ln(AOT) increase slightly in both models likely because the partial AOT values are slightly smaller than the total AOT values. None of the regression coefficients are significantly different from those in the original models except TEMP, which is only marginally significant at the $\alpha=0.05$ level in both models. It has been shown that MISR total AOT is sensitive to particles with diameters ranging from about 0.05 to 2.0 μm including accumulation-mode dust particles (Kahn et al., 1998), which roughly corresponds to the size range of $PM_{2.5}$ Examining the MISR and MODIS datasets reveals that the difference between the total and partial AOT is less than 5% for approximately 80% of the data in each dataset. For only 5% of the data, the difference is greater than 25%. This small difference between total and partial AOT seen in our study region explains why there is little impact of using partial AOT for predicting $PM_{2.5}$ concentrations.

4.4.2. Stability of predictor variables

The number of predictors that can be included in a regression model and their robustness depend on the data sample size.

Table 4
Estimated regression coefficients of the MISR model (sample size $N=359$) and MODIS model (sample size $N=1779$) using partial AOT values

Parameter	MISR partial AOT ($R^2=0.61$)			MODIS partial AOT ($R^2=0.52$)		
	Estimate	Standard error	CI factor	Estimate	Standard error	CI factor
Intercept						
Spring	3.16*	0.12	23.6	2.48*	0.05	12.2
Non-Spring	3.74*	0.19	42.1	2.94*	0.08	18.5
ln(AOT)						
Spring	0.20*	0.05	$AOT^{0.20}$	0.20*	0.04	$AOT^{0.20}$
Non-Spring	0.44*	0.08	$AOT^{0.44}$	0.47*	0.06	$AOT^{0.47}$
ln(PBL)	0.006	0.03	$PBL^{0.006}$	−0.14*	0.02	$PBL^{-0.14}$
ln(WIND)	−0.33*	0.04	$WIND^{-0.33}$	−0.16*	0.02	$WIND^{-0.16}$
TEMP	−0.004+	0.002	$e^{-0.004\times TEMP}$	0.002++	0.001	$e^{0.002\times TEMP}$
Wind direction						
From East	−0.060	0.07	0.94	0.12*	0.04	1.13
From North	−0.005	0.05	0.99	0.10*	0.03	1.11
From South	0.42*	0.05	1.52	0.43*	0.02	1.54
From West[a]	0.00		1.00	0.00		1.00

* Significant at the $\alpha=0.001$ level.
+p value=0.068.
++p value=0.061.
[a] Reference level in the categorical predictor.

Table 5
Percentages of 1000 bootstrap sample runs in which each predictor is significant at the $\alpha = 0.05$ level at various sample sizes

Predictor	% significant			
	$N = 360$	$N = 500$	$N = 800$	$N = 1400$
ln(AOT)	99.9	100.0	100.0	100
NONSPRING × ln(AOT)	77.0	86.7	97.5	99.9
ln(PBL)	92.6	98.2	99.9	100.0
ln(WIND)	98.6	99.8	100.0	100.0
Wind direction	100.0	100.0	100.0	100.0
TEMP	44.3	57.5	71.7	91.5
NONSPRING	91.2	97.7	99.8	100.0

Certain predictors can become unstable as the sample size gets smaller. Our concern is that some regression coefficients in the MISR model are unstable due to the relatively small size of the MISR dataset. We apply bootstrap techniques on the larger MODIS dataset with various bootstrap sample sizes to test the stability of the predictor variables used in Eq. (2). This analysis also serves as a performance test of the MODIS model when different datasets are used in the regression. The MODIS dataset is randomly sampled 1000 times at each bootstrap sample size and each random sample is used to fit the MODIS model. The percentages of these 1000 runs in which a predictor is significant are shown in Table 5. The bootstrap model runs provide strong evidence that, except for surface air temperature (TEMP), all the predictors and thus the overall model structure, are stable (greater than 95% chance of being significant) when the data sample has at least 800 data points. When the sample size is reduced to 360 (the size of the MISR dataset), TEMP only has a 44.5% chance of being a significant predictor. Given the general similarity between the MISR and the MODIS datasets, the bootstrap results suggest that the significance of TEMP in the MISR model with total AOT may not be robust.

Table 5 also suggests that ln(PBL) has a high probability (92.6%) of being a significant predictor at a sample size of 360. Analysis of RUC20 data shows an unusually strong correlation between log-transformed wind speed and log-transformed PBL in the MISR dataset in springtime (approximately 30% of the MISR dataset) as compared to the modest correlation in the MODIS dataset. This suggests that the PBL data in the MISR dataset might behave similarly to that 7.4% of the 1000 bootstrap samples of the MODIS dataset in which ln(PBL) is not a significant predictor. A larger dataset with multiple years of MISR data might be needed to provide the statistical power to make ln(PBL) a significant predictor.

4.4.3. Temporal resolution of $PM_{2.5}$ data

Both MISR and MODIS AOT are instantaneous measurements of particle abundance whereas EPA 24-h $PM_{2.5}$ concentrations are integrated measures over a day. Diurnal variation of $PM_{2.5}$ concentrations is likely to have a negative impact on the association between the two variables. We averaged $PM_{2.5}$ concentrations between 10 am and 12 am collected from two non-filter based continuous monitors (one in Missouri, the other in Illinois) to represent the mean hourly $PM_{2.5}$ concentrations in the satellite time window. To avoid data correlation between the two monitors, we matched the hourly $PM_{2.5}$ concentrations from each monitor separately with MODIS AOT. MISR data were not used because of its small size after matching. Based on the variable stability analysis, we developed a simplified MODIS model using either hourly or 24-h $PM_{2.5}$ concentrations as the dependent variable, MODIS AOT as the primary predictor, and the most stable meteorological parameters at this sample size (PBL, WIND, and DIR) as secondary predictors (Eq. (3)).

$$\ln[PM_{2.5}] = \beta_0 + \beta_1(\mathrm{DIR}) + \beta_2\ln(\mathrm{WIND}) + \beta_3\ln(\mathrm{PBL}) + \beta_4\ln(\mathrm{AOT_{MODIS}}) \quad (3)$$

Regression coefficients of $\ln(\mathrm{AOT_{MODIS}})$ and ln(WIND) are highly significant while only one level of DIR (from south) is significantly different from the reference wind direction (from west) (Table 6). The intercept, regression coefficients of ln (WIND) and DIR in the four models are not significantly different from each other due to wide confidence intervals. The regression coefficient of $\ln(\mathrm{AOT_{MODIS}})$ is larger when hourly $PM_{2.5}$ concentration is the dependent variable. Because Eq. (3) assumes a power law functional form between $PM_{2.5}$ concentration and AOT and the maximum AOT values observed at these two sites are less than 1.0, this larger regression coefficient can be explained by the fact that hourly $PM_{2.5}$ concentrations measured between 10 am and 12 pm are lower than daily

Table 6
Regression coefficients of simplified MODIS model at two continuous monitors

Parameter	Monitor in MO ($N = 151$)				Monitor in IL ($N = 147$)			
	Hourly $PM_{2.5}$ ($R^2 = 0.41$)		Daily $PM_{2.5}$ ($R^2 = 0.45$)		Hourly $PM_{2.5}$ ($R^2 = 0.50$)		Daily $PM_{2.5}$ ($R^2 = 0.55$)	
	Estimate	Standard error	Estimate	Standard error	Estimate	Standard error	Estimate	Standard error
Intercept	3.25***	0.12	3.20***	0.09	3.24***	0.12	3.07***	0.08
$\ln(\mathrm{AOT_{MODIS}})$	0.56***	0.07	0.42***	0.05	0.69***	0.07	0.45***	0.05
ln(WIND)	−0.30***	0.07	−0.25***	0.05	−0.24***	0.06	−0.24***	0.04
Wind direction								
From East	−0.15	0.15	−0.051	0.11	0.034	0.15	0.16	0.10
From North	−0.10	0.11	−0.036	0.08	−0.018	0.11	0.058	0.08
From South	0.31***	0.10	0.31***	0.08	0.45***	0.10	0.45***	0.07
From West [a]	0.00		0.00		0.00		0.00	

*** Significant at the $\alpha = 0.001$ level.
[a] Reference level in the categorical predictor.

averages at these two sites. The slightly lower R^2 values in the models where hourly $PM_{2.5}$ concentration is the dependent variable is probably due to the higher measurement errors of continuous monitors. Overall, improving the temporal resolution of $PM_{2.5}$ concentration do not seem to improve the model performance in the current analysis.

5. Conclusions

We developed two general linear regression models to compare the ability of MISR and MODIS AOT to predict ground-level $PM_{2.5}$ concentrations in the St. Louis area in 2003. Using a few meteorological parameters including wind speed, wind direction, mixing height (PBL), and surface air temperature as covariates in the models, the MISR and MODIS models explained 62% and 51% of the variation in daily $PM_{2.5}$ concentrations, respectively. Overall, the MISR model underpredicts $PM_{2.5}$ concentrations by approximately 12%, whereas the MODIS model underpredicts $PM_{2.5}$ concentrations by approximately 18%. In both models, the underestimation occurs at higher $PM_{2.5}$ concentrations. Analysis of the predictor CI factors showed that AOT would predict higher $PM_{2.5}$ concentrations during non-spring seasons than in springtime. Both AOT and meteorological parameters explain greater variability in $PM_{2.5}$ concentrations, indicating overall better model performance, compared to our previous results in the coastal regions of eastern US. Despite the reported positive bias in MODIS AOT, the predictability and the regression coefficients of the two models are comparable, suggesting the possibility of integrating the two datasets to take advantage of the better accuracy of MISR and broader spatial coverage of MODIS. We also evaluated a few factors that may influence model performance. The newly developed particle size/shape indicators in the MISR and MODIS aerosol data do not seem to improve model performance because they only marginally modified the total AOT values in the study region. Regression analysis using bootstrapped samples of the MODIS dataset indicates that AOT, wind speed, wind direction and mixing height remain stable predictors of $PM_{2.5}$ concentrations even when only 20% of original MODIS data are used in the model fitting. Although hourly $PM_{2.5}$ concentration measured in the satellite time window corresponds more directly to AOT, using it as the dependant variable does not seem to improve the model performance. 24-h $PM_{2.5}$ concentration would be a more appropriate dependant variable given its higher accuracy and broader spatial coverage.

The empirical models developed in this analysis have the potential weakness of being region-specific, and they require support of ground measurements. In a previous study, we utilized simulated particle vertical profiles to develop a simple physical model which reliably predicts annual mean $PM_{2.5}$ concentrations ($r \sim 0.8$) (Liu et al., 2004a). Although this approach is transferable and does not need the support of EPA measurements, the issue of data accuracy at higher temporal and spatial resolution is likely to limit its application in the epidemiologic studies mentioned in the introduction. On one hand, the spatial resolution of global chemistry transport models (~150 km in Liu et al., 2004a) are too coarse to show the urban-scale pollution level contrasts which can be linked to the health outcomes in the population at risk. On the other hand, fine resolution (below 20 km) simulation of daily particle vertical distributions still needs extensive validation and the current results are less reliable. In the context of the long-term particle pollution epidemiologic studies which usually have limited ground measurements, empirical models such as the ones presented in this analysis can still be very valuable in filling the gaps between the monitors.

Since we only collected a year's worth of data, inter-annual variation of the association was not considered in our models. As a result, it is possible that the model behavior and regression coefficients may change when multiple years of data are used in the model fitting process. Although the multivariate model presented in the current analysis showed significant advantages as compared to the simple linear models seen in the literature, this model form tends to underestimate at high concentrations. To overcome this flattening effect of log-transformation, advanced data driven modeling techniques, such as spline regression, need to be applied. Finally, it has been shown that the variability of lower tropospheric aerosols over ocean increases significantly over scales from 20 km to 200 km (Anderson et al., 2003). The spatial scale of aerosol variability over land is likely to be substantially smaller due to proximity to emission sources. Therefore, operational satellite AOT data with a resolution of a few kilometers, rather than the more coarsely aggregated standard products currently available, would better capture the spatial variability of fine particles while controlling for the uncertainty associated with AOT retrieval.

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