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TECHNICAL PAPER

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Estimating Fine Particulate Matter Component Concentrations and Size Distributions Using Satellite-Retrieved Fractional Aerosol Optical Depth: Part 1—Method Development

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ABSTRACT

We develop a method that uses both the total column aerosol optical depth (AOD) and the fractional AOD values for different aerosol types, derived from Multiangle Imaging SpectroRadiometer (MISR) aerosol data, to estimate ground-level concentrations of fine particulate matter ($PM_{2.5}$) mass and its major constituents in eastern and western United States. Compared with previous research on linking column AOD with ground-level $PM_{2.5}$, this method treats various MISR aerosol components as individual predictor variables. Therefore, the contributions of different particle types to $PM_{2.5}$ concentrations can be estimated. When AOD is greater than 0.15, MISR is able to distinguish dust from non-dust particles with an uncertainty level of approximately 4%, and light-absorbing from non-light-absorbing particles with an uncertainty level of approximately 20%. Further analysis shows that MISR Version 17 aerosol microphysical properties have good sensitivity and internal consistency among different mixture classes. The retrieval uncertainty of individual fractional AODs ranges between 5 and 11% in the eastern United States, and between 11 and 31% in the west for non-dust aerosol components. These results provide confidence that the fractional AOD models with their inherent flexibility can make more accurate predictions of the concentrations of $PM_{2.5}$ and its constituents.

IMPLICATIONS

Studying the health effects of specific $PM_{2.5}$ constituents is very important in shaping air quality standards and control policies for $PM_{2.5}$. However, such studies are difficult to conduct due to the lack of reliable $PM_{2.5}$ speciation exposure estimates. This article presents a method that uses satellite-retrieved column fractional AOD values for different aerosol types, along with aerosol transport model results, to estimate ground-level concentrations and size distributions of $PM_{2.5}$ and its major constituents. This method can provide valuable information on the spatial characteristics of $PM_{2.5}$ constituents over large areas, and hence can help in designing more efficient emission control policies.

INTRODUCTION

Epidemiologic studies have demonstrated a consistent positive association between ambient fine particle ($PM_{2.5}$, particulate matter ≤ 2.5 μm in aerodynamic diameter) pollution levels and adverse health effects such as increased mortality and morbidity, particularly among those with chronic respiratory and cardiovascular diseases.[1] This association has been demonstrated for a wide range of concentration levels in various regions of the world, without an apparent safety threshold.[2] Urban aerosol is a complex mixture of particles primarily composed of sulfate (SO_4^{2-}), nitrates (NO_3^-), ammonium (NH_4^+), elemental carbon (EC), organic compounds (OC), and various metals.[3] The relative contributions of these species vary substantially by location and season. Although health effects have been related to mass concentrations, it is not likely that mass is the only factor regulating particle toxicity. Limited animal studies indicate that hematologic changes and lung inflammation may be related to certain chemical components of $PM_{2.5}$.[4,5] Since early 2000, the U.S. Environmental Protection Agency (EPA) has established the $PM_{2.5}$ Speciation and Trends Network (STN) to provide data on $PM_{2.5}$ components, to identify sources, develop emission control implementation plans, and support ongoing health-effects studies.[6] Although currently there are approximately 200 $P.M_{2.5}$ speciation monitoring sites throughout the United States, STN is still too sparse to provide full data support for large-scale population exposure studies.

In December 1999, the National Aeronautics and Space Administration (NASA) launched Terra, the first of its Earth Observing System (EOS) satellites. The main mission of Terra is to help us better understand the state of the Earth and its atmosphere as well as the environmental impacts of human activities.[7] One of the instruments aboard, the Multiangle Imaging SpectroRadiometer (MISR), was designed to retrieve column aerosol amounts and properties, even over complex land surfaces. In addition to making global measurements on the Earth's environment and climate, it provides the

possibility for large-scale particle pollution monitoring from space. MISR aerosol optical depth (AOD), a measure of column aerosol abundance, is sensitive to particles with diameters ranging from approximately 0.05 to 2.5 μm, which roughly corresponds to the size range of $PM_{2.5}$ and includes accumulation-mode dust and pollution particles.[8] In various modeling analyses, MISR AOD has shown a significant association with ground level $PM_{2.5}$ concentrations.[9–11]

MISR measures the reflected sunlight from Earth in four spectral bands (446, 558, 672, and 866 nm) at each of the nine cameras pointed in the forward, nadir, and aft directions along the flight path, with spatial resolution of up to 275 m/pixel. This unique multiangle design allows MISR to observe the atmosphere through effective path lengths ranging from one to three, and scattering angles from approximately 60° to 160° in mid latitudes, which provides information on aerosol microphysical properties such as particle size distribution and shape in addition to AOD. The MISR Standard Aerosol Product reports total-column aerosol properties at a spatial resolution of 17.6 km, about once per week in mid latitudes. In developing the aerosol retrieval approach, MISR pre-selects a set of climatologically probable aerosol mixtures to represent aerosol types globally. These mixtures consist of several individual aerosol components that are defined by a size distribution, shape, complex index of refraction, and scale height. The description of these components and the mixtures they form is documented in the MISR Aerosol Physical and Optical Properties (APOP) file and the Aerosol Mixture file. As the MISR instrument and retrieval approach are relatively new, the integration of new knowledge about real-world aerosol properties and MISR algorithm performance leads to modifications of retrieval algorithm and updates of the APOP and Aerosol Mixture files. MISR data are reprocessed periodically to implement these changes, and a new version of data is released. The various parameters included in the MISR aerosol data product can have different maturity levels. Some parameters can be validated more easily, such as AOD, therefore they are more robust and have reached a higher maturity level. Other parameters, such as the retrieved aerosol microphysical properties, are more difficult to validate, and are as yet at a lower maturity level (see MISR aerosol data version information at *http://www.eosweb.larc.nasa.gov/PRODOCS/misr/Version/pge9.html*).

As a research instrument, MISR does not have near real-time data processing capability, therefore is not suitable for regulatory air quality monitoring. However, its unique multiangle technique provides a richer aerosol dataset to study long-term spatial and temporal trends of particle mass, composition, and other information, compared with other spaceborne aerosol sensors. The total AOD (called regional mean or best estimate AOD in the MISR data product) used in previous MISR aerosol related studies are in fact the average optical depth of all successful mixtures.[10–12] The AOD value of each mixture as well as the corresponding success flag (indicator of whether a mixture passes all the selection criteria and is considered a good fit to the observations) are all stored in the MISR data product. To date, no work has been published on how to utilize MISR-retrieved aerosol microphysical properties to help identify ground-level $PM_{2.5}$ size distribution and speciation. We developed a novel method that uses MISR-retrieved aerosol microphysical properties to estimate ground level mass concentrations and size distributions of $PM_{2.5}$ and its key constituents. This effort represents the cutting edge of what is possible with current versions of the data products. We first describe the latest MISR aerosol product (Version 17) for which over a year of data was available at the time of this analysis. These data include mid-visible wavelength (558 nm) AOD and particle property information. In the *Methods* section, we describe our four-step approach of estimating $PM_{2.5}$ mass concentrations and size distributions using fractional AODs, that is, the contribution of each MISR aerosol component to total AOD. Given the lower maturity level of MISR particle property information, we briefly assessed the sensitivity of MISR data in distinguishing different aerosol components in this paper. We defined a prevalence ratio of conflicting aerosol mixtures to measure the consistency of MISR-retrieved aerosol mixtures.

MISR-RETRIEVED AEROSOL MICROPHYSICAL PROPERTIES

All the MISR data products are distributed through the Atmospheric Science Data Center (ASDC) at NASA Langley Research Center (*http://www.eosweb.larc.nasa.gov/PRODOCS/misr/data.html*). MISR retrieves aerosol properties by first assuming a set of aerosol mixtures in the atmosphere. For each MISR observation at a given location and time, top-of-atmosphere radiances for all the mixtures are computed and compared with the MISR observations to determine those mixtures that provide good fits to the data, that is, the "successful mixtures."[13] The theoretical foundation of aerosol microphysical properties for earlier MISR aerosol product versions is described in detail elsewhere.[14] Here we provide the details related to the Version 17 data used in the current paper and the case study presented in the companion paper. Different versions of APOP files, which have been used in various versions of MISR data products, can be obtained from ASDC. Eight aerosol components from the (Version 19) APOP file are used for the Version 17 MISR Aerosol Product (MIL2ASAE) (Table 1). The naming convention of these components is generally particle shape (i.e., spheres, grains, or ellipsoids), followed by a qualitative scattering property (i.e., non-light-absorbing or light-absorbing), and followed by the effective radius for a number-weighted log-normal distribution (e.g., 0.06 μm or 2.80 μm). Single scattering albedo at 558 nm wavelength is added if necessary to distinguish components. Components 19 and 21 are accumulation and coarse-mode dust analogs, having different particle size distributions and single scattering albedos.[15] These components are selected on the basis of the analysis of multiple global atmospheric transport model results, field observations, and the expected

Table 1. Aerosol components assumed in MISR Version 17 retrievals.

No.	Name	Min d[a] (μm)	Max d[a] (μm)	Characteristic Diameter (μm)[b]	Distribution Width[b]	Volume-Weighted Mode Diameter (μm)[3]	Single Scattering Albedo (558 nm)
1	Spherical_nonabsorbing_0.06	0.002	0.8	0.06	1.65	0.10	1
2	Spherical_nonabsorbing_0.12	0.002	1.5	0.12	1.70	0.21	1
3	Spherical_nonabsorbing_0.26	0.02	3.0	0.24	1.75	0.45	1
6	Spherical_nonabsorbing_2.80	0.2	100.00	1.00	1.90	4.55	1
8	Spherical_absorbing 0.12_ssa_green_0.9	0.002	1.5	0.12	1.70	0.21	0.90
14	Spherical_absorbing 0.12_ssa_green_0.8	0.002	1.5	0.12	1.70	0.21	0.80
19	Grains_mode1_h1 (dust)	0.2	2.0	1.00	1.50	1.40	0.98
21	Spheroidal_mode2_h1 (dust)	0.2	12.0	2.00	2.00	5.22	0.90

Notes: [a]Min d and Max d are the minimum and maximum particle diameters for a given aerosol component. [b]The number-weighted lognormal size distribution is described as

$$\frac{dN(d)}{dd} = \frac{1}{d \times \ln(\sigma) \times \sqrt{2\pi}} \exp\left[\frac{(\ln(d) - \ln(d_c))^2}{2(\ln(\sigma))^2}\right],$$

where σ is the distribution width and d_c is the characteristic diameter of the distribution. [c]The volume-weighted mode diameter is calculated assuming a spherical particle shape.

MISR sensitivity to particle size, shape, and optical properties.[14] All the components are assumed to have lognormal size distributions, which are widely used in both the aerosol optics and aerosol chemistry communities. They represent reasonably well the typical peak in particle concentration, the large-size-regime tail, and the usual, fairly steep cutoff for small sizes. Assuming a constant particle density, the relative mass distribution by particle size can be calculated, and is shown in Figure 1. Because components 2, 8, and 14 are all assumed to have identical size distributions, they are represented in Figure 1 by a single curve. The mass distributions of components 6 and 21 are very similar, with 21 slightly flatter than 6. As indicated in Figure 1, particles larger than 2.5 μm may contribute substantially to particle mass for these two components.

The radiative properties of each component such as the path radiances at different wavelengths and satellite viewing geometries are calculated and archived in a look-up table. During the retrieval process, the path radiance of each aerosol mixture is calculated, and compared with the observed radiances.[13,14] A set of statistical tests are performed to determine which of the aerosol mixtures fit the observed radiance well. For computational efficiency, an aerosol mixture is limited to consist of up to three externally mixed aerosol components in the MISR operational retrieval algorithm. The contribution of an aerosol component to total column AOD may range from 0 to 100% in increments no smaller than 5%, and contributions from all components sum to 100%. Even with only three components in each mixture, over 12,000 possible mixtures can be theoretically made out of eight aerosol components, but most of them are not common in the natural environment. In addition, sensitivity studies and available validation measurements indicate that if MISR can distinguish two aerosol components externally mixed in the atmospheric column, each component's contribution to AOD can be retrieved to an accuracy of approximately 20%. This result is expected for reasonable aerosol optical depth, surface properties, and relatively cloud-free conditions, and it may be exceeded for better conditions.[15–17] It implies that MISR is only able to distinguish a limited number of aerosol mixtures with confidence. Given all these constraints, 74 aerosol mixtures are considered in the Version 17 MISR aerosol retrieval. These mixtures can be organized into eight mixture classes consisting of all mixtures having the same components, but in different proportions (Table 2). The naming convention of the mixture classes is generally particle shape, followed by the description of each of the components (e.g., effective radius and single scattering albedo).

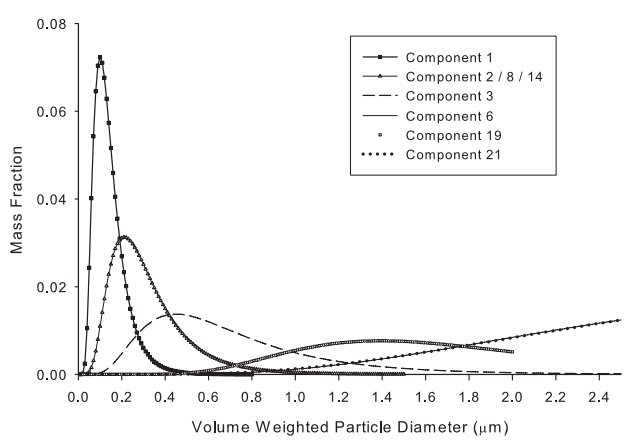

Figure 1. Mass size distribution for the MISR aerosol components. Note that Components 8 and 14 have the same mass distributions by size as Component 2, and the distributions of Components 6 and 21 are nearly identical in the plotted size range. The truncated lognormal distributions reflect the maximum particle sizes listed in Table 1. The mode diameters are also listed in Table 1.

METHODS

Calculation of Fractional AOD in the Lower Atmosphere

We can calculate the fractional AOD contribution of each aerosol component as its average contribution to total AOD across all the successful mixtures (eq 1). The result is up to eight component-specific AOD values for any given MISR aerosol observation, each of which

Table 2. MISR aerosol mixtures used in Version 17 retrievals and the contributions of aerosol components to each mixture.

Mixture Class	Mixture Class Name	1st Component and Its Fractional Contribution to Mixture	2nd Component and Its Fractional Contribution to Mixture	3rd Component and Its Fractional Contribution to Mixture
1 (Mixtures 1–10)	Spherical_Reff_0.06_Reff_2.80_ Nonabsorbing	1, Fractions: 0.9, 0.85, 0.8, 0.75, 0.7, 0.6, 0.5, 0.4, 0.3, 0.2	6, Fractions: 0.1, 0.15, 0.2, 0.25, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8	None
2 (Mixtures 11–20)	Spherical_Reff_0.12_Reff_2.80_ Nonabsorbing	2, Fractions: 0.9, 0.85, 0.8, 0.75, 0.7, 0.6, 0.5, 0.4, 0.3, 0.2	6, Fractions: 0.1, 0.15, 0.2, 0.25, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8	None
3 (Mixtures 21–30)	Spherical_Reff_0.26_Reff_2.80_ Nonabsorbing	3, Fractions: 0.9, 0.85, 0.8, 0.75, 0.7, 0.6, 0.5, 0.4, 0.3, 0.2	6, Fractions: 0.1, 0.15, 0.2, 0.25, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8	None
4 (Mixtures 31–40)	Spherical_Reff_0.12_SSA_green_ 0.9_Reff_2.80_SSA_green_ 1.0_Absorbing	8, Fractions: 0.9, 0.85, 0.8, 0.75, 0.7, 0.6, 0.5, 0.4, 0.3, 0.2	6, Fractions: 0.1, 0.15, 0.2, 0.25, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8	None
5 (Mixtures 41–50)	Spherical_Reff_0.12_SSA_green_ 0.9_Reff_2.80_SSA_green_1.0_ Absorbing	14, Fractions: 0.9, 0.85, 0.8, 0.75, 0.7, 0.6, 0.5, 0.4, 0.3, 0.2	6, Fractions: 0.1, 0.15, 0.2, 0.25, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8	None
6 (Mixtures 51–62)	Spherical_Reff_0.12_Reff_2.80_ Med_Dust	2, Fractions: 0.72, 0.48, 0.16, 0.54, 0.36, 0.12, 0.36, 0.24, 0.08, 0.18, 0.12, 0.04	6, Fractions: 0.08, 032, 0.64, 0.06, 0.24, 0.48, 0.04, 0.16, 0.32, 0.02, 0.08, 0.16	19, Fractions: 0.2, 0.2, 0.2, 0.4, 0.4, 0.4, 0.6, 0.6, 0.6, 0.8, 0.8, 0.8
7 (Mixtures 63–70)	Spherical_Reff_0.12_Med_Dust_ Coarse_Dust	2, Fractions: 0.4, 0.4, 0.4, 0.4, 0.2, 0.2, 0.2, 0.2	19, Fractions: 0.48, 0.36, 0.24, 0.12, 0.64, 0.48, 0.32, 0.16	21, Fractions: 0.12, 0.24, 0.36, 0.48, 0.16, 0.32, 0.48, 0.64
8 (Mixtures 71–74)	Med_Dust_Coarse_Dust	None	19, Fractions: 0.8, 0.6, 0.4, 0.2	21, Fractions: 0.2, 0.4, 0.6, 0.8

represents the aggregated contribution of one aerosol component.

$$AOD_{i,k} = \frac{\sum_{j=1}^{74} \alpha AOD_{\text{mixture j}} \times Fraction_{\text{component i in mixture j}}}{No.\ of\ successful\ mixtures} \quad (1)$$

where $AOD_{i,k}$ is the fractional component i in the air column for a MISR aerosol observation k; $AOD_{\text{mixture j}}$ is the total AOD (558 nm) of mixture j; $Fraction_{\text{component i in mixture j}}$ is the contribution of component i to the total mid-visible AOD for mixture j; and $\alpha = 1$ if mixture j is a successful mixture; otherwise $\alpha = 0$.

To compare MISR fractional AOD, which is a column measure, with EPA $PM_{2.5}$ constituent concentrations measured at ground level, the vertical distribution of particles must be considered. Because measurements of aerosol vertical profiles over large regions of the United States are not currently available, we used the simulation results from chemical transport models such as GEOS-Chem or air quality models such as CMAQ[18,19] to fill in this information. There are few publications regarding the validation of model-simulated aerosol vertical profiles. One study reports that the uncertainty of GEOS-Chem vertical profiles of aerosol extinction is reported within 25% of lidar observations at a few locations.[20] The lower-air proportion of fractional AODs are calculated by scaling the MISR column AODs with model-simulated particle profiles as shown in eq 2.[9]

$$\text{MISR lower-air AOD} = \frac{\text{Model lower-air AOD}}{\text{Model column AOD}} \times \text{MISR total AOD} \quad (2)$$

It should be noted that the MISR retrieval reports column average aerosol shape, single-scattering albedo, and size distribution, as there is no height-resolved aerosol information in the MISR data, except for stereo-derived elevations for discrete aerosol plumes. In addition, the MISR data alone cannot determine whether different sized particles are from different sources, or are simply more or less hydrated, unless detectable differences in other particle properties, such as shape, make these distinctions possible. The uncertainties caused by these assumptions can not be quantified without additional data, hence are not discussed further. From this point on, we refer to MISR lower air fractional AODs simply as MISR fractional AODs.

Linking EPA $PM_{2.5}$ and Component Concentrations with MISR Fractional AODs

We can conduct regression analysis to link the concentrations of $PM_{2.5}$ mass and its major constituents (e.g., SO_4^{2-}, NO_3^-, OC, EC, and possibly silicon concentration as a surrogate for mineral dusts) measured by the STN network, with MISR fractional AODs. The general model form is given in eq 3. On the left hand side, the dependant variable, EPA $PM_{2.5}$ constituent concentration, may include $PM_{2.5}$ mass concentration, concentrations of the individual $PM_{2.5}$ constituents, or silicon concentration. On the right hand side, MISR fractional AODs serve as the main predictor variables. The change of relative humidity (RH) is expected to influence the association between AOD and particle mass concentrations.[21,22] In principle, the form of eq 3 allows the effect of RH on each MISR aerosol component to be assigned individually. For example, because RH changes have a very limited impact on mineral dust particles, no correction factors are necessary for these components. If total AOD is used as the single predictor, applying a RH correction factor to it would introduce uncertainty to predicted $PM_{2.5}$ concentrations when dust makes up a

large proportion of particle mass. Assigning RH correction factors to non-dust MISR aerosol components requires careful consideration because each of them can be a mixture of several species with identical size distributions. Seasonal and regional indicators can be included in the models if detailed information on particle hygroscopicity is not available. Apparently, knowledge about the spatial and temporal variation of $PM_{2.5}$ pollution plays an important role in determining the specific formats of seasonal and geographical variables as well as the RH correction factors. Interested readers can find a few examples in our previous publications.[10,11]

$$\text{EPA } PM_{2.5} \text{ Constituent Concentration} = \beta_0 + \sum_{i=1}^{8} \beta_i \times \text{MISR fractional } AOD_i \times \text{RH correction factor}_i + \beta_9 \times \text{Seasonal Indicator} + \beta_{10} \times \text{Geographical Indicator} \quad (3)$$

The adjusted R^2 is defined as $1 - [(n-1) \times (1-R^2)]/(n-k)$, where R^2 is the regular model R^2, defined as the ratio of the sum of squares explained by a regression model and the total sum of squares around the mean. n is the number of non-missing observations, and k is the number of variables in the model. When the sample size is small, the adjusted R^2 can effectively account for the lost degrees of freedom caused by adding more variables into the model.[23] Because MISR data are relatively sparse, the adjusted R^2 value can serve as the primary measure of model performance and selection criterion. Physical interpretation of the parameter estimates should also serve as an important model selection criterion. At comparable adjusted R^2 levels, the model with positive regression coefficients for all fractional AOD variables should be selected as the final model because it is physically meaningful. The regression coefficients of the seasonal and geographical indicators can be either positive or negative.

Regression diagnostics such as the Cook's Distance (*Cook's D*) and the Studentized Residual can be applied to identify data points that have significant impacts on the estimated regression coefficients, or deviate significantly from the overall trend of the dataset. To determine whether a *Cook's D* value of a given data point is large (i.e., this data point may have a large impact on the regression coefficients), Kutner et al.[24] recommend comparing it to an F distribution with $p + 1$ degree of freedom in the numerator and $N - p - 1$ degree of freedom in the denominator. p is the number of predictor variables in the regression model, and N is the total number of non-missing data. If the *Cook's D* value of a data point is smaller than the 10th or 20th percentile value of this F distribution, the data point has little influence on the estimated regression coefficients. The absolute value of the Studentized Residual = 3 may also be used as an initial screening criterion, which identifies individual residuals beyond 99.7% of the data population.

Estimating the Size Distributions of $PM_{2.5}$ Constituents Using Regression Results

We can calculate the size distribution of a specific $PM_{2.5}$ constituent using the regression model linking it with fractional AODs. For each observation, the product of a significant fractional AOD value and its regression coefficient in a given model has the unit of a concentration. It represents the estimated contribution of this aerosol component to the concentration of the $PM_{2.5}$ constituent, which is the dependent variable of this model. When averaged over all the observations, we can calculate the percentage contribution of each significant MISR aerosol component to a $PM_{2.5}$ constituent. Because each MISR aerosol component has its specific particle size distribution mathematically expressed as a probability density function (PDF), the estimated size distribution of a $PM_{2.5}$ constituent (also expressed as a PDF) can be calculated by superimposing the PDFs of all significant MISR components weighted by their percentage contributions (eq 4).

$$\text{PDF of a } PM_{2.5} \text{ constituent} = \sum_{i=1}^{8} f_i \times \text{PDF of MISR component } i \quad (4)$$

The factor f_i is the percentage contributions of MISR component i to the mass concentration of a specific $PM_{2.5}$ constituent. If component i is not statistically significant in the model, f_i should be set to zero. A significant model intercept represents the proportion of $PM_{2.5}$ constituent concentration, which cannot be explained by the model, and therefore should be excluded when estimating the size distribution. Doing so will not affect the shape of the estimated size distribution, but we should be aware that such an estimated size distribution represents only the proportion of a specific $PM_{2.5}$ constituent explained by the model. This calculation can be conducted for each season and geographical region or for the combined dataset.

In summary, the fractional AOD approach has four steps. First, we calculate column fractional AOD for each component (eq 1). Second, we estimate lower-air fractional AOD by scaling the column fractional AOD with the simulated aerosol vertical profiles from a CTM or air quality model (eq 2). Third, we build the regression models with fractional AOD values as the major predictors of concentrations of $PM_{2.5}$ constituents such as total mass or various chemical species. RH correction factors and seasonal and geographical indicators of $PM_{2.5}$ variations can also be included in the models to improve model performance (eq 3). These models can be used to produce $PM_{2.5}$ exposure estimates when ground measurements are not available. The final step is to derive particle size distribution information using the regression coefficients (eq 4).

In a case study using 2005 EPA STN data in the continental United States, we estimated ground-level concentrations of $PM_{2.5}$ mass and its major constituents such as sulfates, nitrates, and OC using the approach described in this paper. GEOS-Chem simulated

aerosol vertical profiles provided the scaling factors to calculate the lower-air MISR fractional AOD values. Our results show that regression models with fractional AODs as predictor variables have substantially higher predictive powers when compared with similar models using only total-column AOD as a predictor. The improvements of adjusted R^2 values range from 19 to 44% in the east, and even greater in the west. Fine particle size distributions estimated by these models compared reasonably well with results reported in the literature. Details of this case study are presented in a companion paper, "Estimating Fine Particulate Matter Component Concentrations and Size Distributions Using Satellite-Retrieved Fractional Aerosol Optical Depth: Part 2—A Case Study."[25]

SENSITIVITY ASSESSMENT OF MISR RETRIEVED AEROSOL

Microphysical Properties

As mentioned in the *Introduction*, a set of statistical tests are used in the MISR aerosol retrieval algorithm to determine which of the 74 mixtures are successful. This means that more than one mixture may be considered a good fit to the observations. In general, fewer successful mixtures mean better constrained retrieval. Therefore, the validity and performance of the method described in the previous sections is directly related to MISR's ability to distinguish aerosol components. Note that a more accurate MISR aerosol retrieval (i.e., less successful mixtures) is not equivalent to the observed atmospheric aerosol consisting of only the few components in the successful aerosol mixtures. More realistically, it means that the observed aerosol is better captured by MISR-assumed aerosol optical properties. Fully testing MISR aerosol microphysical property retrievals, especially when multiple particle modes are present, is very difficult. In situ measurements of aerosol optical properties such as those acquired during dedicated field campaigns are required. Given the low maturity level of Version 17 particle size and single-scattering albedo information, we conduct a brief assessment of the consistency of those aerosol mixtures considered successful by the MISR retrieval algorithm. When MISR is not able to distinguish among different aerosol mixtures, it becomes less meaningful to use fractional AODs to predict the concentrations of ground-level $PM_{2.5}$ constituents. This assessment serves partially as a quality assurance for the method developed in this paper.

We conduct this assessment from two different aspects. First, as seen in Table 2, MISR aerosol mixtures can be divided into two large clusters, non-dust (mixtures 1–50) and dust-containing (mixtures 51–74). The non-dust mixtures can be further divided into two clusters, that is, nonabsorbing (mixtures 1–30) and absorbing (mixtures 31–50). The sensitivity of MISR retrievals can be evaluated by examining whether MISR can distinguish different clusters of mixtures. For example, high sensitivity to mineral dust would mean that for a dusty observation, few, if any, non-dust mixtures are considered successful by MISR retrieval algorithm.[15] MISR retrieval sensitivity to particle type is indicated, for example, by whether mixtures from both the non-dust and the corresponding dust-containing mixtures are simultaneously considered successful or are distinguished by the algorithm. Second, we examine whether mixtures within each mixture class having the most different proportions are simultaneously considered successful. For example, if the first mixture (90% component 1 and 10% component 6) and the last mixture (20% component 1 and 80% component 6) in the first mixture class are simultaneously considered good fits to a MISR observation, it indicates that MISR can not effectively distinguish among these aerosol components. In this case, we count that conflicting retrievals occur in the first mixture class in this observation. Inconsistent retrievals in other mixture classes are counted similarly.

To complement the case study presented in the companion article, the sensitivity assessment is conducted using the entire set of MISR aerosol observations spatially matched to approximately 200 EPA STN sites in 2005. There are 4537 matched data records in total. Kahn et al.[16] noted that when AOD at 558 nm falls below approximately 0.1 or 0.2 over ocean, information in the satellite observations about particle properties decreases and the number of successful mixtures often grows. Consequently, we limit our data to those with column AOD value greater than 0.15 and less than or equal to 1.5. Setting the lower limit of 0.15 reduces the statistical power of our analysis, as it excludes over 60% of the raw data and reduces the dynamic range of the data. However, this is necessary so that the quality of MISR data can be maintained. This lower limit may be relaxed to include less polluted days as MISR data become more mature. The upper limit is set to reduce the possibility of including erroneous observations caused by inadequate cloud screening. Although AOD values greater than 1.5 have been frequently observed in the megacities of developing countries such as Beijing,[26] our previous study suggests that it is very rare in the United States.[27] In our case, setting the upper limit of 1.5 only excludes less than 0.3% of the raw data. Because it is widely known that $PM_{2.5}$ composition in the east is different from the west, analyses are conducted for the two regions separately. There are 1269 MISR observations spatially matched to STN sites in the east, and 373 in the west.

The median number of successful aerosol mixtures in each observation is 7 in the east and 17 in the west. The number of successful mixtures decreases as AOD increases, indicating higher sensitivity of MISR to aerosol microphysical properties at higher AOD values, as expected. MISR identified a single successful mixture in 86 observations in the east and 5 observations in the west. For the rest of the data, the average coefficient of variation of AOD (the AOD standard deviation of all successful mixtures divided by the mean AOD) is 15% in the east and 22% in the west. Figure 2 shows the percentages of (a) dust-containing mixtures and (b) non-absorbing mixtures (or bright mixtures) in a MISR

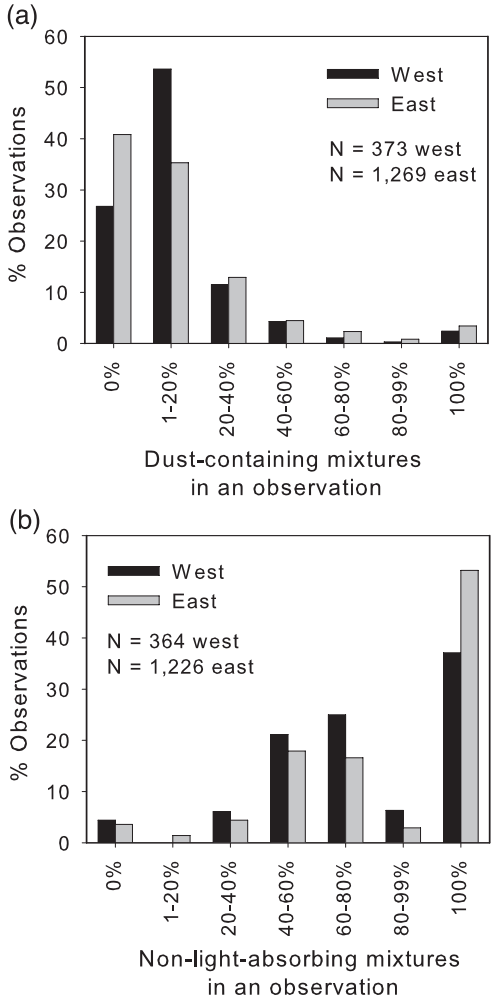

Figure 2. Distributions of (a) dust-containing and (b) non-light-absorbing mixtures. The y-axis represents the percentage of observations in the west or east. The x-axis of plot (a) represents the percentage of dust-containing aerosol mixtures (vs. non-dust mixtures) in an observation. The x-axis of plot (b) represents the percentage of non-light-absorbing aerosol mixtures (vs. the light-absorbing mixtures) in an observation.

observation, separated into east and west. The percentages of non-dust and dust-containing mixtures sum up to one, and the percentages of nonabsorbing and absorbing mixtures sum to 1. A highly skewed distribution indicates higher retrieval sensitivity, therefore lower error. A flat or centered distribution, on the other hand, means inability to distinguish different mixtures, therefore higher retrieval error. MISR aerosol data display similar distributions of the percentage of dust-containing mixtures in an observation in the east and west. Approximately 40% of the observations in the east have no dust-containing mixtures, as compared with 27% in the west. Over 80% of the data have either few dust-containing mixtures (≤20%) or many (≥80%) in one observation, for both east and west. Only 4% of the data have nearly equal numbers of non-dust and dust-containing mixtures (i.e., those having 40–60% of dust-containing mixtures). This 4% retrieval error shows that MISR has a relatively high sensitivity to distinguishing spherical from nonspherical particles. The percentage distributions of non-light-absorbing mixtures are also similar in the east and west, except that over half of the data in the east contain 100% non-light-absorbing mixtures. The particles in the east are brighter possibly because of the greater contribution of bright sulfate particles to the overall aerosol abundance. However, 18–21% of the data have nearly the same number of non-light-absorbing and absorbing mixtures in both the east and west, suggesting that Version 17 data has a larger retrieval error when differentiating particles with different single-scattering albedos. The MISR retrieval assumes that mineral dust has either grain-like (for accumulation mode) or spheroidal (for coarse mode) shapes; all other components are assumed to be spherical.[15] The difference in particle shape, together with scattering properties and size distributions, help MISR distinguish dust particles from other particles more easily than spherical particles having different absorption characteristics. These trends agree with expectations based on MISR pre-launch sensitivity studies.[8,14]

Table 3 lists the prevalence of conflicting retrievals made by MISR in the first six mixture classes in the east and west, respectively. Conflicting retrievals in the last two mixture classes are not discussed here because they are difficult to define. The prevalence rate is the ratio of the occurrences of conflicting retrievals over the number of successful mixtures in each mixture class. For example, 455 MISR observations in the east report successful mixtures in mixture class 1 and conflicting mixtures are found in 36 of these samples. As a result, the prevalence rate of conflicting retrievals in mixture class 1 in the east is 36/455 = 8%. A higher prevalence rate suggests that MISR cannot accurately determine the relative contribution of each component to total AOD. Therefore, it is an indicator of the retrieval uncertainty of individual fractional AOD values. This ratio ranges

Table 3. The prevalence rates of conflicting MISR retrievals in different mixture classes in the east and west.

Mixture Class	1	2	3	4	5	6
Presence count of successful mixtures in each mixture class in the east	455	1098	343	561	228	743
Prevalence of conflicting retrievals	8%	5%	11%	7%	11%	8%
Presence count of successful mixtures in each mixture class in the west	264	317	191	222	122	271
Prevalence of conflicting retrievals	31%	19%	22%	21%	21%	11%

between 5% (mixture class 2) and 11% (mixture classes 3 and 5) in the east, and between 11% (mixture class 6) and 31% (mixture class 1) in the west. The uncertainties of fractional AOD retrievals are likely to increase the model errors when estimating surface $PM_{2.5}$ concentrations, which will be discussed in the companion paper. When assessed together, the above results show that MISR Version 17 aerosol microphysical properties have good sensitivity and internal consistency in the eastern United States, which gives confidence to our fractional AOD models. The sensitivity is not as good in the west, partially because of a lower overall AOD level (0.24, vs. 0.30 in the east).

In summary, MISR Version 17 data can effectively distinguish between dust particles and non-dust particles, less so between bright and darker particles. In addition, MISR data are more consistent internally in the east than in the west. Significant upgrades to the MISR particle property retrievals are expected in subsequent versions of the product, as the results of recent field campaigns and other analyses are incorporated into the algorithm.

SUMMARY

We take a four-step approach in linking MISR retrieved aerosol microphysical information with ground level $PM_{2.5}$ mass concentrations. We first calculate the fractional AOD values for each MISR aerosol component using the mixtures selected in the MISR Version 17 aerosol product. We estimate the lower atmospheric proportions of the fractional AOD values using aerosol-transport-model-simulated aerosol vertical profiles as scaling factors. Regression models are then developed, with the derived lower-air fractional AODs as major predictor variables to estimate ground-level concentrations of total $PM_{2.5}$ mass and major $PM_{2.5}$ constituents. Finally, particle size distributions can be estimated as well, using the regression coefficients obtained in the third step. The fractional AOD models are able to adjust for differences in fine particle composition between the eastern and western United States. They are also flexible enough to allow better RH correction based on the hygroscopicity of individual aerosol components. Because the fractional AOD approach uses particle type data produced by MISR, in addition to the total AOD, the regression models contain measurement-based information about the concentrations and size distributions of different $PM_{2.5}$ constituents. Our assessment of MISR's sensitivity shows that Version 17 aerosol microphysical properties have good internal consistency; the uncertainty levels of fractional AODs range between 5 and 11% in the east, and between 11 and 31% in the west. Further analysis indicates that MISR is able to distinguish dust particles from non-dust particles with an error of approximately 4%, and to distinguish light-absorbing particles from non-light-absorbing particles with an error of approximately 20%.

Although the fractional AODs can be calculated using MISR data alone, the development of the regression models needs the support of ground-level $PM_{2.5}$ measurements. Like any data-driven statistical model, the regression coefficients of the predictor variables may change if the model is fitted with another dataset. $PM_{2.5}$ speciation data from a few sites are needed to calibrate the regression models before they can be expanded temporally and spatially. In the case where there is no $PM_{2.5}$ speciation data available, directly applying these regression models developed in the United States in another region could result in predicted $PM_{2.5}$ concentrations with higher uncertainties. However, when compared with the most previous model studies that treat total AOD as a single predictor of $PM_{2.5}$, our approach can have significantly improved predicting power, as shown in the companion paper. This approach may be used to extend the STN network to provide better estimates of $PM_{2.5}$ and its major constituents, therefore to help optimize the design of regional or national emission control policies.

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