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New algorithms and their application for satellite remote sensing of surface PM2.5 and aerosol absorption

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ABSTRACT

Estimation of Particulate Matter (PM) concentration and aerosol absorption is very important in air quality and climate studies. To date, smoke, mineral dust and anthropogenic pollutants are the most uncertain aerosol components in their optical and microphysical properties. In this study, we retrieve the PM2.5 and Absorbing Aerosol Optical Depth (AAOD) from the Total Ozone Mapping Spectrometer (TOMS), the Moderate Resolution Imaging SpectroRadiometer (MODIS) and the Multiangle Imaging SpectroRadiameter (MISR) measurements. A global chemical transport model (GEOS-CHEM) is used to simulate the vertical profiles of PM2.5 and AAOD. We find that the 2003 heat wave has strong impact on PM2.5 across Europe and increased the average PM2.5 concentration by 18%. The aerosol species with the largest concentration increase are ammonium nitrate, black carbon and mineral dust. The Aerosol Robotic Network (AERONET) measurements have been used to validate our retrieval of AAOD. We find that there is a significant agreement between AERONET measurements and our retrievals with the correlation coefficient, slope and intercept of 0.91, 0.99 and 0.001, respectively. The absorbing aerosols can exert negative health effect, increase positive aerosol radiative forcing and contribute positive aerosol-climate feedbacks.

1. Introduction

Atmospheric aerosols have a significant effect on human health (Dockery et al., 1993; Pope et al., 2002) and on regional and global climate changes (Hansen et al., 1998; Ramanathan et al., 2007). To date, the information on aerosol size distribution and chemical composition is still limited. In order to improve our understanding of the causes, impacts and control of Particulate Matter (PM) pollution, which affects many regions of the world, detailed knowledge of aerosol properties is essential for the emission reduction and air quality monitoring.

Epidemiological studies reveal that fine PM with diameter smaller than 2.5 μm (PM2.5) has adverse health effects including asthmatic symptoms, pulmonary inflammation, cardiopulmonary mortality, and lung cancers (Anderson, Martello, & Rohar, 2002; Atkinson, 2001; Pope et al., 2002). Particle chemical compositions have also been associated with high risks to human health. Increasing data on air quality monitoring shows that many toxic aerosol species accumulate in the fine particle size fraction. This fraction of particles has higher oxidative stress potential and can penetrate and destroy mitochondria with epithelial cells (Li, Hao, Phaden, Hinds, & Nel, 2003). As aerosols have a direct relevance to air quality and health effect, it is important to quantify the optical and microphysical properties of fine and ultra fine particles.

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Remote sensing measurements have provided a good opportunity to detect the source and monitor the transport and evolution of aerosols released from biomass burning and dust storms. The Moderate Resolution Imaging Spectrometer (MODIS) and the Multiangle Imaging SpectroRadiometer (MISR) instruments have been operated on board the Terra satellite since December 1999. Aerosol Optical Depth (AOD) can be determined from MODIS and MISR measurements over oceans and most land surfaces (Diner et al., 1998; Kahn et al., 2005; Kaufman et al., 1997; King et al., 1992; Tanre, Kaufman, Herman, & Matoo, 1997) through observations at visible and infrared wavelengths. The Total Ozone Mapping Spectrometer (TOMS) aerosol index at the ultraviolet wavelength is sensitive to absorbing aerosols so that it can be used to detect cabonaceous and dust particles (Herman et al., 1997; Torres, Bhartia, Herman, Ahmad, & Gleason, 1998). The relationship between column AOD and surface PM2.5 has been explored over the United States using satellite AOD data (Liu, Sarnat, Kilaru, Jacob, & Koutrakis, 2005; Wang & Christopher, 2003). They find that MODIS or MISR AOD has good correlations with hourly or monthly PM2.5 measurements. However, the relationship is simply empirical due to the complicated dependence on several factors such as aerosol chemical composition, size distribution, aerosol profile and atmospheric conditions. The application of satellite observations in assessing the PM for mega cities such as Mexico City, Delhi, Hong Kong and New York has also been shown to be promising (Gupta, Christopher, & Wang, 2006; Massie, Gille, Edwards, & Nandi, 2006). High correlation between the daily mean AOD from satellite and ground measurements indicates that satellite observations can be used to detect heavy pollution episodes, such as mega city haze, forest fires and dust storms. Koelemeijer, Homan, and Matthijse (2006) detected an increase in AOD over Europe using MODIS data during the summer of 2003. The question remains: How much does the average PM2.5 concentration increase due to the anomalous heat wave of 2003? Which kinds of aerosol species make a large contribution to the increase of PM2.5? What kind of interaction exists between air quality and climate change? Here, we use AOD and size information from satellite data and aerosol profiles from the GEOS-CHEM model to derive the surface PM2.5 concentrations. Furthermore, we use our recently developed retrieval algorithm for aerosol single scattering albedo (Hu, Martin, & Fairlie, 2007) to calculate the Absorbing Aerosol Optical Depth (AAOD).

2. Methodology

Suspended PM is complicated by virtue of its chemical composition, size distribution and particle shape. Air quality standards have been expressed in terms of particles mass concentration as PM2.5 or PM10 which represents particles of aerodynamic size less than of 2.5 and 10 μm, respectively. Two common approaches have been currently used to represent the particles size: the modal and sectional approaches (Zhang et al., 2004). For the modal approach, the particle size distribution is assumed to be composed of several lognormal modes

$$n_j(r) = \frac{N_j}{\sqrt{2\pi}\ln\sigma_{g,j}}\frac{1}{r}\exp\left[-\frac{(\ln r - \ln r_{g,j})^2}{2(\ln\sigma_{g,j})^2}\right] \tag{1}$$

where N_j is a normalizing constant (in cm^{-3}); r is the radius; $\log\sigma_{g,j}$ is the geometric standard deviation. $r_{g,j}$ is the geometric mean radius.

For each aerosol type, the AOD can be computed at wavelength λ as follows:

$$\tau_j(\lambda) = \int_{z_1}^{z_2}\int_{r_{min}}^{r_{max}} Q_e(x,m)\pi r^2 n_j(r)\,dr\,dz \tag{2}$$

where Q_e is the extinction efficiency with size parameter x and refractive index m. z is the altitude. For non-spherical particles, the size parameter $x_{eff} = 2\pi r_{eff}/\lambda$. The effective radius r_{eff} is defined as the ratio of the third to second moment of the aerosol size distribution (Hansen & Travis, 1974)

$$r_{eff,j} = \int_{r_{min}}^{r_{max}} n_j(r)r^3\,dr \Big/ \int_{r_{min}}^{r_{max}} n_j(r)r^2\,dr \tag{3}$$

Then the mass concentration between the ground and altitude z can be expressed as

$$M_j(r_l, z) = 4\rho r_{eff,j}\tau_j f/(3Q_j^{ext}z) \tag{4}$$

where r_l is the selected radius upper limit for the aerosol size distribution, such as 1.25 μm for PM2.5 or 5 μm for PM10. ρ is the aerosol density and Q_j^{ext} is the average column extinction efficiency. f is the fraction of optical depth below altitude z. The values of f, $r_{eff,j}$ and z are selected from the GEOS-CHEM model with validation from ground-based and air-based measurements. The effective radius is related to aerosol size, shape, mixing state and hygroscopy. We use the calculated relationships to interpret the satellite retrievals of $r_{eff,j}$ and alternatively employ in situ measurements of aerosol optical and microphysical properties to calibrate the effective radius for specific uses. We assume uniform aerosol properties between the surface and altitude z. The total aerosol mass concentration for the external mixture can be calculated using the sum of the component aerosols, such as sulfate, nitrate, ammonium, black carbon, organics, dust and sea salt. The extinction efficiency is calculated by Mie theory for spherical particles. For non-spherical particles, we use the T-Matrix method (Mishchenko, Lacis, Charlson, & Tavis, 1995) to calculate the extinction efficiency and effective radius. We use AOD data from MODIS over the ocean and from MISR over the land. The detailed microphysical and optical properties of major aerosol components used in this study are given in Table 1.