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## An improved algorithm for retrieving the fine-mode fraction of aerosol optical thickness, part 1: Algorithm development



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#### ABSTRACT

The fine-mode fraction (FMF) can be a useful tool to separate natural aerosols from man-made aerosols and to assist in estimating surface concentrations of particulate matter with a diameter  $<2.5~\mu m.$  A LookUp Table-based Spectral Deconvolution Algorithm (LUT-SDA) was developed here for satellite-based applications using data such as MODerate resolution Imaging Spectroradiometer (MODIS) measurements. This method was validated against ground-based FMF retrievals from the Aerosol Robotic Network (AERONET). The LUT-SDA was then applied to two MODIS-retrieved aerosol optical thickness (AOT) products for the period of December 2013 to July 2015: the MODIS Collection 6 (C6) Dark Target (DT) AOT product and the simplified high-resolution MODIS Aerosol Retrieval Algorithm (SARA) AOT product. In comparison with the MODIS C6 FMF product in three study areas (Beijing, Hong Kong, and Osaka), FMFs estimated by the LUT-SDA agreed more closely with those retrieved from the AERONET with a very low bias. Eighty percent of the FMF values fell within the expected error range of  $\pm$  0.4. The root mean square error (RMSE) was 0.168 with few anomalous values, whereas the RMSE for the MODIS FMF was 0.340 with more anomalous values. The LUT-SDA FMF estimated using SARA AOT data conveys more detailed information on urban pollution than that from MODIS C6 DT AOT data. As a demonstration, the seasonally-averaged spatial distribution of the FMF in Beijing was obtained from the LUT-SDA applied to SARA AOT data and compared with that of the AERONET-retrieved FMF. Their seasonal trends agreed well.

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#### 1. Introduction

Atmospheric aerosols play a very important role in Earth's energy balance and on climate change (Kaufman et al., 2002; Ramanathan et al., 2007). They can also have an adverse effect on human health (Pope et al., 2002). At present, our understanding of aerosol size distributions from a spatial perspective is still limited.

The fine-mode fraction (FMF) is an important physical property of aerosols that can be used to separate natural aerosols from man-made aerosols (Bellouin et al., 2005). Like AOT, the aerosol FMF can be estimated from either space-borne AOT observations such as those from the MODerate resolution Imaging Spectroradiometer (MODIS) or from the ground-based AOT measurements from the AERONET. Levy et al.

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(2010) reported that the MODIS-retrieved FMF over land is still experimental and highly uncertain. This is because satellite AOT retrievals have to differentiate signals from aerosols and the land surface whose reflectance can be substantial (Diner et al., 2005; Hauser et al., 2005; Mishchenko and Geogdzhayev, 2007; Li et al., 2009; Kokhanovsky et al., 2010; Lee and Chung, 2013). Because of this, the FMF over land is much less accurate than over oceans. However, the AERONET FMF is based on the Spectral Deconvolution Algorithm (SDA), which uses solar extinction data obtained directly from solar measurements. As such, it is only slightly influenced by surface reflectance and is almost as accurate over oceans as over land (O'Neill et al., 2001, 2003). Gasso and O'Neill (2006) showed a good correlation between the fine-mode AOT from a sunphotometer and airborne in situ measurements using the SDA. However, the AERONET has a much smaller spatial coverage than does the MODIS.

The FMF may help estimate surface concentrations of particulate matter (PM) with a diameter < 2.5  $\mu$ m (PM<sub>2.5</sub>), as it pertains to the contributions of smaller and larger particles to the AOT. Many studies have

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attempted to develop statistical models to estimate PM<sub>2.5</sub> from AOT, Di Nicolantonio et al. (2007) reported a sound correlation between the fine-mode AOT and PM<sub>2.5</sub> with a correlation coefficient reaching 0.74. Zhang and Li (2013) found that the relationship between fine-mode AOT and PM<sub>2.5</sub> was stronger than that between AOT and PM<sub>2.5</sub> under hazy weather conditions in winter. Zhang and Li (2015) derived a relation involving the FMF and the columnar volume-to-extinction ratio of fine particulates based on eight AERONET sites, and applied it to estimate PM<sub>2.5</sub> from MODIS measurements. However, Levy et al. (2007) revealed that the MODIS FMF had little correlation with the AERONET FMF. This may be because AERONET and MODIS FMFs were derived using different methods. AERONET assumes a fine mode and coarse mode with no overlap, whereas MODIS uses bimodal lognormal models that may be overlapped to a certain extent (Kampe, 2008). Also, the MODIS FMF is not determined as a continuous variable, but as 11 discrete values from 0 to 1. Jethva et al. (2010) compared the MODIS FMF with the AERONET FMF and found that the root mean square difference between the two was 0.61 (number of samples, N = 651).

As shown by O'Neill et al. (2001), the SDA was developed as a simple and efficient method for determining the fractions of fine and coarse modes from spectral AOTs. They showed that the Ångström exponent (AE) of fine-mode aerosols can be extracted directly from the first and second order spectral derivatives of AOT, which means that values of AOT at a minimum of three wavelengths are needed. However, most of the current satellite aerosol retrieval products provide AOT at only the blue and red bands (Zha et al., 2011; Luo et al., 2015), e.g., the retrieval products derived from the DT method (Levy et al., 2007) and the minimum reflectance technique (Wong et al., 2011). If AOT is available at only two wavelengths, the AE can be calculated using the Volz method (Soni et al., 2011), but the AE derivative cannot be obtained from a second-order polynomial fit. Thus, an FMF retrieval method based on AOT at two bands needs to be developed.

The main objective of this study is to improve the spectral decomposition method so that it can estimate the FMF based on AOT at two

wavelengths following a LookUp Table-based SDA (LUT-SDA) method for satellite applications. The method is demonstrated using ground-based AERONET measurements made in Beijing, Hong Kong, and Osaka. We will then use the derived FMF to estimate PM<sub>2.5</sub>, the subject covered in Part II of this study.

#### 2. Data and methods

#### 2.1. Study area

For the sake of demonstration on an urban scale, we chose Beijing, Hong Kong, Osaka, and their surrounding areas as our test area because of their distinctly different environments and differing meteorological conditions (Fig. 1). Beijing is the most polluted and driest among the three sites. Hong Kong is a coastal city with low to moderate levels of pollution and a marine climate. Osaka is in the centre of Japan and is the largest commercial and industrial city after Tokyo (Sasaki and Sakamoto, 2005). Because AOT retrievals depend critically on surface albedo and aerosol single scattering albedo, the retrievals from each of these regions are different, offering an opportunity to test the spatial adaptability of the LUT-SDA method.

#### 2.2. The LUT-Spectral Decomposition Algorithm (SDA)

The LUT-SDA proposed here uses an LUT for retrieving the FMF from satellite-derived AOT and AE. To build an LUT, a set of hypothetical FMF  $(\eta)$  and AE derivative  $(\alpha')$  values are imported to the SDA calculation along with the satellite-determined AE  $(\alpha)$  for deriving the AE of fine-mode aerosols  $(\alpha_f)$ . The incremental interval for  $\eta$  is 0.01 with a range from 0 to 1. The incremental interval for  $\alpha'$  is 0.001 and its range for a given satellite image will be discussed in section 3.1. The FMF can then be estimated by minimizing  $\chi^2$  in this LUT. The LUT based on the SDA is described as follows:

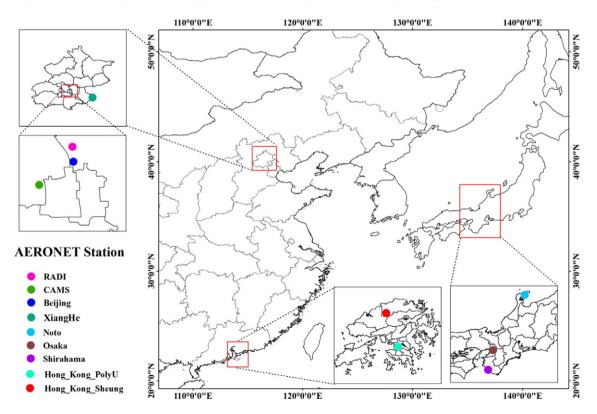


Fig. 1. The Beijing, Hong Kong, and Osaka study areas (outlined in red). The locations of the nine AERONET sites used in the study are: Beijing (39.98°N, 116.38°E), CAMS (39.93°N, 116.32°E), RADI (40.01°N, 116.38°E), XiangHe (39.75°N, 116.96°E), Noto (37.33°N, 137.14°E), Osaka (34.65°N, 135.59°E), Shirahama (33.69°N, 135.36°E), Hong\_Kong\_Polyu (22.30°N, 114.18°E), and Hong\_Kong\_Sheung (22.48°N, 114.12°E).

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