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Study on influence of different mixing rules on the aerosol components retrieval from ground-based remote sensing measurements

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ABSTRACT

Mixing states of aerosol components significantly influence the optical, physical and radiative properties of ambient aerosols. The five-component aerosol composition model, including black carbon (BC), brown carbon (BrC), mineral dust (DU), ammonia sulfate (AS) and aerosol water (AW), is improved with considering different mixing rules in this paper. Then we retrieve the volume fractions and column mass concentrations of these aerosol components at Beijing from ground-based AERONET remote sensing measurements, such as refractive index, size distribution, and single scattering albedo. A residual minimization method is used to derive aerosol composition difference under dust, haze and clean conditions at Beijing in 2011. Three mixing rules including Maxwell–Garnett (MG), Bruggeman (BR) and Volume Average (VA) are demonstrated to have significant influences on the aerosol component retrievals. We find that over 50% difference of volume fraction of DU occurs by switching between MG and BR rules. Therefore, applicability of each mixing rule is also investigated. We propose that BR is more suitable for the dust case, MG is better than other two rules for the haze case, and VA is the best choice for the clean case. We also discuss the application scopes of different mixing rules by comparing the recovered aerosol optical parameters with AERONET observations.

1. Introduction

Aerosol is considered to have significant impacts to global climate and atmosphere environment. It affects climate in both direct and indirect ways (Haywood and Boucher, 2000). Although aerosol scattering at short wavelength is believed to have cooling effects on atmosphere, absorption of aerosols cannot be ignored because the cooling effect of aerosol radiative forcing at the top of atmosphere may change to warming with highly absorbing aerosols (Schuster et al., 2005; Haywood and Shine, 1995; Charlock and Sellers, 1980). Atmospheric models are widely used in estimating global aerosol radiative forcing for their capability of providing a wide spatial and fine temporal estimate (Wang et al., 2013a). However, inevitable large uncertainties have limited their application (Bond and Bergstrom, 2006; Park et al, 2003). Sato et al. (2003) found that models might need 2–4 times increase for BC emission inventories to match single scattering albedo (SSA) measurements by AERONET (AErosol RObotic NETwork) (Holben et al., 1998). Aerosol is a mixture of solid and liquid components suspended in the atmosphere and radiative impacts of aerosol components are widely varying (Alam et al., 2011), making it difficult to accurately estimate the total aerosol radiative impact. Therefore studies on aerosol component and composition become more and more important for the estimation of aerosol radiative forcing. For example, Srivastava et al. (2012a) derived aerosol optical parameters that are important to radiative forcing

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such as aerosol optical depth (AOD) and SSA by aerosol chemical composition measurements and ambient meteorological parameters, and then used them to estimate aerosol direct radiative forcing.

Satellite remote sensing technology provides global aerosol products like AOD and size distribution (Kaufman et al., 2002), but so far it cannot effectively acquire aerosol composition (Ganguly et al., 2009). In situ sampling and laboratory analysis can provide accurate aerosol composition but it is difficult to maintain the natural status of ambient aerosols. Besides, the high time and money cost due to its complexity have limited the application of this technology. In the field of identifying aerosol types and inferring aerosol composition, ground-based remote sensing measurements are becoming promising in recent years for their global coverage and continuous automatic observation, for example, the AERONET. Lee et al. (2010) employed SSA at 440 nm and fine mode fraction (FMF) at 550 nm derived by AERONET to classify global aerosol into several classes. Srivastava et al. (2012b) used the same method to discriminate aerosol types according to their size and radiation absorptivity, and investigated diurnal and spatial variation of aerosol types. Srivastava et al. (2014) also quantified the possible radiative implications of different aerosol types. Aerosol optical and physical properties such as AOD, SSA, refractive indices and size distribution are also useful for aerosol composition retrieval. Schuster et al. (2005) used a three-component aerosol model including black carbon (BC), ammonia sulfate (AS) and water to infer the content and specific absorption of BC. He also investigated the sensitivity of BC content to optical properties. Dey et al. (2006) supplemented partly absorbing components (organic carbon (OC) or mineral dust (DU), depending on observation period) to the model and inferred BC and its specific absorption. Arola et al. (2011) extended the model analogously to acquire absorbing organic carbon (also called as brown carbon, BrC) content. These studies usually ignore DU considering that the spectra of imaginary refractive index of DU and BrC are quite similar. Li Z Q et al. (2013) and Wang et al. (2013a) simulated SSA of BrC and DU and found that differences of SSA spectra in 670–1020 nm can be used to distinguish these two absorbing components. They added SSA information to the inversion scheme and established a five-component model, inferring BrC and DU simultaneously. They further applied this model to the serious haze episodes in Beijing in January 2013, and acquired aerosol composition in haze episodes (Wang et al., 2013b).

Atmospheric particles are a mixture of different components during complex atmospheric chemical and physical processes such as nucleation and condensation. Mixing states of aerosols have significant impacts on light extinction and thus radiative forcing (Lesins et al., 2002; Li L et al., 2013). For example mass absorption efficiency of BC aerosol varies with its mixing state, geometrical morphology and density (Fuller et al., 1999; Schuster et al., 2005; Dey et al., 2006). There are mainly two assumptions of aerosol mixing. One is external mixing in which each aerosol component is physically separated with the others, i.e., each individual particle contains only one type of component. The other is internal mixing, assuming at least two types of aerosol component included in one particle, and the mixture comprehensively reflects physical and chemical properties of all components. Pure external mixing is rare in realistic aerosol. Field measurements show that most aerosol components are internally mixed with others (Lesins et al., 2002) and telescope data illustrates that over 50% of soot and ammonia sulfate aerosols are internally mixed (Schuster et al., 2005). Sometimes initial aerosols are single particles at the source and then develop to internal mixtures during atmospheric chemical reactions. For example, BC, OC and sulfate could be originated from the same combustion process (Yang et al., 2009); however, they might aggregate into a new internal mixture later. Therefore natural aerosol is generally believed to be internally mixed, and varies with its composition, morphology of particles and relative humidity of atmosphere (Schuster et al., 2009; Lesins et al., 2002; Xue et al., 2011).

Internal mixing states of aerosols (here we mainly focus on the mixing rules) have significant impacts on optical and physical properties of aerosol but have not been given adequate attention. Some studies employed one mixing rule to infer aerosol composition. For example, Arola et al. (2011) applied Maxwell–Garnett (MG) effective medium approximation to the retrieval of four-component aerosol model, Wang et al. (2013a) used Volume Average (VA) mixing rule to calculate refractive indices of aerosol mixture in their five-component model. Some studies investigate the differences between mixing rules. For example, Dey et al. (2006) compared Bruggeman (BR) effective medium approximation and MG in composition retrieval but BR was only applied to a two-component model (BC and AS). Schuster et al. (2005) compared external mixing, MG and concentric sphere models and chose MG to infer BC content. Lesins et al. (2002) systematically analyzed the impacts of aerosol mixing states on radiative forcing and compared refractive index simulation under MG, BR and VA mixing rules, but did not applied their results to actual aerosol composition retrieval.

Based on the assumption of internal mixing aerosol, this paper improves the five-component model described in previous work (Li Z Q et al., 2013; Wang et al., 2013a,b) by applying different mixing rules (MG, BR and VA). We then utilize AERONET observation of Beijing site in 2011 to retrieve aerosol composition under selected typical cases (i.e. dust, haze and clean day). We focus on the comparison of retrievals between these mixing rules, and then use simulated refractive index spectra and aerosol absorption optical depth to investigate the applicability of these mixing rules.

2. Data

2.1. AERONET products

AERONET is composed of over 500 observation sites of sun-sky photometer instruments all over the world (until the end of 2012), providing high quality aerosol products (Holben et al., 1998). Sun-sky photometer is capable of automatic observation of direct solar irradiation and also sky radiation with ALMucantar (ALM) and Solar Principle Plane (SPP) geometries. Retrieval results such as refractive indices and size distribution are acquired by ALM geometry, which supplies sky radiation measurements at 28 relative azimuth angles but a fixed solar zenith, almost covering the entire almucantar. Symmetry criteria of ALM might screen some invalid measurements but at least 21 azimuth angles are needed for the retrieval accuracy. AERONET inversion method