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Estimation of atmospheric columnar organic matter (OM) mass concentration from remote sensing measurements of aerosol spectral refractive indices

Ying Zhang[a], Zhengqiang Li[a,*], Yele Sun[b], Yang Lv[a], Yisong Xie[a]

[a] State Environmental Protection Key Laboratory of Satellite Remote Sensing, Institute of Remote Sensing and Digital Earth, Chinese Academy of Sciences, Beijing 100101, China

[b] Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, China

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ABSTRACT

Aerosols have adverse effects on human health and air quality, changing Earth's energy balance and lead to climate change. The components of aerosol are important because of the different spectral characteristics. Based on the low hygroscopic and high scattering properties of organic matter (OM) in fine modal atmospheric aerosols, we develop an inversion algorithm using remote sensing to obtain aerosol components including black carbon (BC), organic matter (OM), ammonium nitrate-like (AN), dust-like (DU) components and aerosol water content (AW). In the algorithm, the microphysical characteristics (i.e. volume distribution and complex refractive index) of particulates are preliminarily separated to fine and coarse modes, and then aerosol components are retrieved using bimodal parameters. We execute the algorithm using remote sensing measurements of sun-sky radiometer at AERONET site (Beijing RADI) in a period from October of 2014 to January of 2015. The results show a reasonable distribution of aerosol components and a good fit for spectral feature calculations. The mean OM mass concentration in atmospheric column is account for 14.93% of the total and 56.34% of dry and fine-mode aerosol, being a fairly good correlation (R = 0.56) with the in situ observations near the surface layer.

1. Introduction

Aerosol, pending particulate matter in the atmosphere, can not only influence human health causing air pollution, but also reduce the solar radiation and energy budget leading to climate changes (Boucher et al., 2013). Because the different aerosol component may present different spectral characteristics, the aerosol components can be retrieved using optical remote sensing technology. Based on this principle, Satheesh et al. (1999, 2002); Satheesh and Srinivasan (2002); Satheesh and Krishna (2005) developed a remote sensing inversion algorithm of aerosol components which assumed aerosols to be external mixed. However, the method using the fixed size distribution of each aerosol component, which should vary in the real atmosphere. The internal mixing hypotheses are also widely used because its merit on the weak dependence of size distribution and relatively accurate calculation of the aerosol scattering and absorbing characteristics (Schuster et al., 2005, 2009, 2015; Arola et al., 2011; Li et al., 2013; Wang et al., 2013). For absorbing component (e.g. black carbon) coexisting with non-absorbing components (e.g. water content, sulfate, nitrate and ammonium) in aerosols, the absorbing component can be separated from the spectral differences based on remote sensing measurements (Schuster et al., 2005). Schuster et al. (2009) also estimated the dust component which is typically coarse modal particles and with relatively weak light absorbing characteristic (Yang et al., 2009). Similarly, Arola et al. (2011) estimated the brown carbon which presents the optical absorbing characteristic mainly from 200 to 550 nm (Andreae and Gelencsér, 2006), but assuming the conditions with ignoring dust component. In order to distinguish spectrally absorbing components (e.g. brown carbon and dust), the slope of SSA spectrum between 440 and 675 nm is introduced into retrieval processing (Wang et al., 2013). Li et al. (2013) separate several optical absorbing components (i.e. BC, brown carbon and dust) from the optical scattering components using internal mixing hypotheses. They extended the species of remotely sensed aerosol components to five kinds. Moreover, aerosol bimodal characteristics are used to estimate the aerosol absorbing components including BC, brown carbon and hematite in fine and coarse mode (Schuster et al., 2015). However, most researches focus on the estimated aerosol components and fail to develop the new species retrieval (e.g. Li et al., 2015; Zhang et al., 2015; Xie et al., 2014).

Recently, many studies find that organic matter (OM), composed of

* Corresponding author.
E-mail address: lizq@radi.ac.cn (Z. Li).

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water-insoluble (WIOM) and water-soluble organic matter (WSOM), played a important role and accounts for about 20–50% in aerosols (Sun et al., 2016; Chan and Yao, 2008; Cao et al., 2007, 2004, 2003; Putaud et al., 2004; Saxena and Hildemann, 1996). But only a few studies separated the OM based on remote sensing measurements (Xie et al., 2017; Choi and Ghim, 2016). Xie et al. (2017) added the sea salt and particulate organic matter based on the method of Li et al. (2013) to extend the species to seven kinds, but the weak hygroscopic characteristics of organic matter are not included in the algorithm. Similarly, the water-soluble and insoluble organic carbons in fine mode are considered in the research of Choi and Ghim (2016), but the water-soluble organic carbon is hard to distinguish from inorganic salts without regard to aerosol hygroscopic growth.

In this study, we separate the OM (including WIOM and WSOM) from aerosol using the strong scattering and weak hygroscopic characteristics of OM (Li et al., 2014a,b), and improve the classification of aerosol species in condition of volume balance. This study fill up the gap of measurement mode between chemical analysis and remote sensing and decrease the estimated component biases. The methodology of inversion algorithm described in section 2. Section 3 shows the results and comparisons with chemical analysis, and conclusions and discussions are listed in section 4.

2. Methodology

2.1. Hygroscopicity characteristics of OM components

In order to retrieve the OM component, we separate the aerosol size distribution into fine and coarse mode in advance. Due to the anthropogenic OM mainly from fine mode, the algorithm in this study is not involved the OM component in coarse mode (e.g. fungus, spore). Therefore, in coarse mode there are only two components including dust-like (DU) and aerosol water content (AW_c), while five components in fine mode (i.e. BC, WSOM, WIOM, ammonium nitrate-like (AN) and AW_f) is a challenge in this study.

For estimating the aerosol components in the fine mode, hygroscopic difference between OM and AN component as an extra constrained condition is applied to the algorithm coupled with the condition of light-absorbing difference. Fig. 1 shows the geometric growth factors (GF) of typical inorganic salts and OMs (including WIOM and WSOM) in laboratory, and the GFs of these components present a significant difference (Sjogren et al., 2007; Prenni et al., 2003). GFs of inorganic salts are all larger than 1.5 (Hu et al., 2010), while that of OMs are much smaller than inorganic salt with mean value of 1.1 (Varutbangkul et al., 2006) except the low content of malonic acid and levoglucosan components in the aerosols (Jing et al., 2016; Liu et al., 2016). Therefore, the OM regards as the low hygroscopic components can be distinguished with AN. However, the OM mixture includes WSOM and WIOM, which can be only distinguished by the phase state and slight difference in imaginary part of atmospheric aerosol complex refractive index (CRI). Thus, the empirical relationship of the ratio of WSOM in the total OM is used to constrain the iteration process for the fine mode, which is descripted in section 2.3.1.

2.2. The aerosol composition model and component separation

The algorithm consists of two parts, including the estimation of atmospheric aerosol CRI (Zhang et al., 2017), denoted by $m = n - ik$, and components (this study). For the first part, the aerosol CRIs are separated into fine and coarse mode and iterated through fitting measurements with the inputs of aerosol optical depth (τ), absorbing aerosol optical depth (τ_a) and volume particle size distribution (VPSD). τ at five wavelengths (440, 500, 675, 870, 1020 nm) and τ_a at four (440, 675, 870, 1020 nm) are used in the retrieval process, and then fine- and coarse-mode CRIs at four wavelengths can be obtained and is applied to the aerosol component retrievals. In this study, the aerosol CRIs and

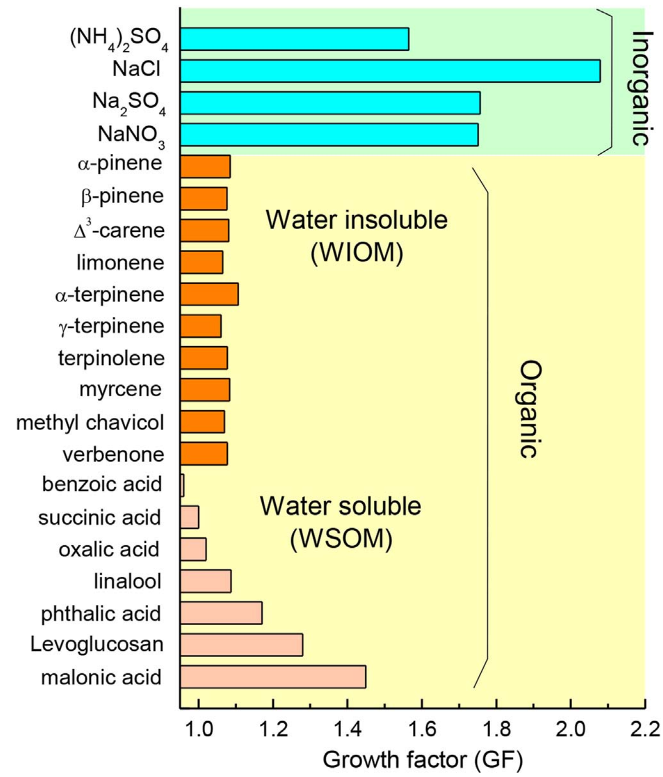

Fig. 1. Different geometric hygroscopic growth factor between inorganic and organic aerosols with RH around 85% of relative humidity. (Jing et al., 2016; Liu et al., 2016; Shi et al., 2012; Hu et al., 2010; Varutbangkul et al., 2006).

VPSD in fine and coarse mode (Zhang et al., 2016, 2017) are used as the input data for the aerosol components retrieved by remote sensing measurements. The CRI inputs include the real and imaginary parts in fine mode at four wavelengths as well as τ_a and only the imaginary part in coarse mode at 675 nm due to the low retrieved accuracy in coarse mode (Zhang et al., 2017).

For the second part, the species classification of aerosol components shows in Fig. 2 and the steps of inversion algorithm as follows: (1) estimation of CRIs for the fine and coarse modes; (2) the separation between water-soluble and water-insoluble matters; (3) estimates proportion of AN and AW_f depended on hygroscopic growth factor; (4) the separation between WSOM and AN depended on hygroscopic difference; (5) the separation between WIOM and BC depended on spectral difference; (6) estimates aerosol water content in coarse mode.

We classified the aerosol into five main species (BC, OM, AN, AW, DU) as illustrated in Fig. 2. In the five main species, OM and DU are the mixtures and thus the CRIs are determined like below. For OM, real part (n) at all wavelengths are according to Sun et al. (2007) and imaginary part (k) are according to Chen and Bond (2010). For DU, 5% (amount to 2.24% in volume percent) of hematite as light-absorbing dust and 95% of illite as light-scattering dust are mixed to calculate the CRIs of DU (Schuster et al., 2015; Lafon et al., 2004; Wagner et al., 2012). Other CRIs of BC, AN and AW and the effective densities are listed in Table 1.

2.2.1. Estimation of CRIs for the fine and coarse modes

In order to separate CRIs for different modes, we need to separate the VPSD into complete log-normal functions firstly following the VPSD breakdown method described in Cuesta et al. (2008). The multi-modal log-normal distributions fits the AERONET-retrieved VPSD by the following formula:

$$\frac{dV(r)}{dlnr} = \sum_{i=1,m} \frac{C_i}{\sqrt{2\pi}\,|ln\sigma_i|} exp\left[-\frac{1}{2}\left(\frac{lnr - lnr_i}{ln\sigma_i}\right)^2\right],\ \text{m= 1,2,\&...,} \quad (1)$$