Science of the Total Environment 550 (2016) 950–960

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

A closure study of aerosol optical properties at a regional background mountainous site in Eastern China

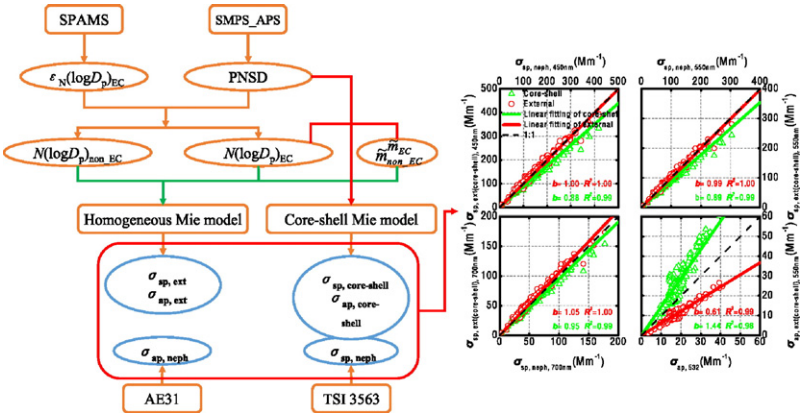
Liang Yuan [a,b], Yan Yin [a,b,*], Hui Xiao [b], Xingna Yu [a,b], Jian Hao [b], Kui Chen [b], Chao Liu [a,b]

[a] Collaborative Innovation Center on Forecast and Evaluation of Meteorological Disasters, Nanjing University of Information Science & Technology, Nanjing 210044, China
[b] Key Laboratory for Aerosol–Cloud–Precipitation of China Meteorological Administration, School of Atmospheric Physics, Nanjing University of Information Science and Technology, Nanjing 210044, China

HIGHLIGHTS

- A spherical Mie model with assumptions of external and core–shell mixtures is used.
- Two-component optical aerosol model and three scenarios of EC ratios are considered.
- Averaged size-segregated EC ratio is appropriate for absorption coefficient inversion.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 22 November 2015
Received in revised form 24 January 2016
Accepted 29 January 2016
Available online 4 February 2016

Editor: D. Barcelo

Keywords:
Aerosol optical properties
Closure study
Uncertainty
Size-segregated
Mixing state
Mt. Huang

ABSTRACT

There is a large uncertainty in evaluating the radiative forcing from aerosol–radiation and aerosol–cloud interactions due to the limited knowledge on aerosol properties. In-situ measurements of aerosol physical and chemical properties were carried out in 2012 at Mt. Huang (the Yellow Mountain), a continental background mountainous site in eastern China. An aerosol optical closure study was performed to verify the model outputs by using the measured aerosol optical properties, in which a spherical Mie model with assumptions of external and core–shell mixtures on the basis of a two-component optical aerosol model and high size-segregated element carbon (EC) ratio was applied. Although the spherical Mie model would underestimate the real scattering with increasing particle diameters, excellent agreement between the calculated and measured values was achieved with correlation coefficients above 0.98. Sensitivity experiments showed that the EC ratio had a negligible effect on the calculated scattering coefficient, but largely influenced the calculated absorption coefficient. The high size-segregated EC ratio averaged over the study period in the closure was enough to reconstruct the aerosol absorption coefficient in the Mie model, indicating EC size resolution was more important than time resolution in retrieving the absorption coefficient in the model. The uncertainties of calculated scattering and absorption coefficients due to the uncertainties of measurements and model assumptions yielded by a Monte Carlo simulation were ±6% and ±14% for external mixture and ±9% and ±31% for core–shell mixture, respectively. This study provided an insight into the inherent relationship between aerosol optical properties and physicochemical

* Corresponding author at: School of Atmospheric Physics, Nanjing University of Information Science and Technology, Ningliu Road 219, Nanjing, China.
E-mail address: yinyan@nuist.edu.cn (Y. Yin).

http://dx.doi.org/10.1016/j.scitotenv.2016.01.205

characteristics in eastern China, which could supplement the database of aerosol optical properties for background sites in eastern China and provide a method for regions with similar climate.

1. Introduction

Atmospheric aerosol particles can affect the Earth's energy balance and climate by aerosol–radiation interactions through scattering and absorbing the incident solar radiation (Charlson et al., 1992) and by aerosol–cloud interactions by acting as cloud condensation nuclei (CCN) and ice nuclei (IN) in cloud formation to modify the optical properties and life time of clouds (Andreae and Rosenfeld, 2008). Recently, it has also been found that the influences of atmospheric aerosol particles on solar radiation have an important impact on the variations of crop evapotranspiration (Valipour and Eslamian, 2014; Valipour, 2015a; Valipour, 2015b), which plays a vital role in water management with respect to global water crisis issues (Valipour, 2015c; Valipour, 2015d). Light-absorbing carbon (LAC), one of the most important components of atmospheric aerosols, which dominates the light absorption in the atmosphere (Moffet and Prather, 2009) and hence has a positive radiative forcing on the climate system (Jacobson, 2001), is one of the largest uncertainties in quantifying the global radiative forcing of aerosols.

LAC is an individual aggregate and unmixed once it is emitted into the atmosphere, where it tends to mix with other chemical components during air mass aging (Riemer et al., 2004). Previous model studies have estimated that the direct radiative forcing of LAC at the top of the atmosphere ranged widely from $+0.17$ to $+0.80$ W/m^2 with the evolution of its mixing state (Jacobson, 2000; Chung and Seinfeld, 2002). Jacobson (2000) showed that the positive radiative forcing of LAC for the internal mixture was approximately 3 times higher than that for the external one. However, recent research showed that the light absorption of LAC was less than that predicted from observationally constrained theoretical calculations, which indicated that many climate models might overestimate warming by LAC (Cappa et al., 2012). This is due not only to the highly uneven horizontal, vertical, and temporal distributions of aerosols (IPCC, 2013) but also to the lack of knowledge about the in-situ measurements of the mixing state of LAC with other aerosol species, which has a non-negligible impact on its radiative forcing (Jacobson, 2001). Also, there are uncertainties associated with the models, such as the Mie model and the radiative transfer models, which are used to derive aerosol radiative properties from available measurements (Wex et al., 2002) and the related aerosol physicochemical properties that are closely related to the radiative forcing (Moffet and Prather, 2009). Thus, it is of great importance to verify the reliability of the outputs from an optical model with the measured aerosol properties and to evaluate the uncertainties of calculations attributed to the uncertainties of measurements and models.

As a way to investigate the inherent relationship between aerosol optical properties and physicochemical characteristics, the Mie model was applied to reconstruct aerosol optical properties with measured size spectra and chemical composition as the input parameters (Bohren and Huffman, 2008). Closure studies based on in-situ measurements and the Mie model, with two extreme assumptions of the LAC mixing state, external mixture (LAC is distinct from other aerosol particles) and internal mixture (EC is incorporated homogeneously within the non-light-absorbing component in aerosol particles), were performed to verify the reliability of the model outputs by comparing the reconstructed and measured optical properties extensively and to identify the uncertainties associated with the input parameters and numerical models. Moreover, sensitivity experiments can be conducted based on the closure (Wex et al., 2002; Cheng et al., 2006; Cheng et al., 2008; Ma et al., 2011). In China, related studies were conducted at non-urban sites in the Pearl River Delta, North China Plain (Cheng et al., 2006; Ma et al., 2011), and a mountainous site in northern China (Shen, 2012). The studies presented reliable information about the aerosol optical properties associated with the aerosol physical and chemical characteristics in China. However, such researches were based on the assumption of external or homogeneously internal mixtures for LAC and non-light-absorbing component. Meanwhile, relatively low temporal and size resolution of LAC distribution determined by traditional filter membrane samplers (Höller et al., 2003) were used, which was a critical parameter in the Mie model. Recent studies suggested that the assumption of core–shell mixture, which was an LAC core surrounded by well-mixed non-absorbing components, was more appropriate in the evaluation of aerosol direct radiative forcing (Jacobson, 2000; Bohren and Huffman, 2008; Liu et al., 2012; Liu et al., 2014). To the best of our knowledge, very few related studies have been conducted that take these three factors into consideration, especially for aerosols at a background site in eastern China.

To fill in this gap, in-situ measurements were conducted at Mt. Huang (the Yellow Mountain), a continental background mountainous site in eastern China. In this study, the term "EC" (elemental carbon) is used instead of "LAC" to refer to the light-absorbing carbon because EC is the major ingredient of LAC at Mt. Huang and no other light-absorbing carbon was considered. This paper presents a spherical Mie model with assumptions of external and core–shell mixtures in which a two-component optical aerosol model and high size-segregated EC ratio were considered. The model outputs were verified by an aerosol optical closure study using the measured aerosol optical properties. The potential uncertainties in the model associated with the assumptions were discussed. The influence of high resolution of EC size and time distribution on the modeled aerosol optical properties were investigated via two sensitivity experiments. Finally, the uncertainties of calculation attributed to the uncertainties of measurements and model assumptions were evaluated with a Monte Carlo simulation.

2. Measurements

2.1. Site description

The campaign was conducted at Mt. Huang (30.12° N, 118.19° E) from September 30 to October 8 (274–282 day of year (DOY)), 2012, at a height of approximately 900 m above mean sea level. Mt. Huang is approximately 300 km southwest of the megacity cluster of the Yangtze River Delta and is a tourist region with few industrial activities. Fig. 1 shows the average distribution of aerosol optical depth over eastern China during the observation period and the location of Mt. Huang. The relatively low aerosol optical depth values around Mt. Huang indicates that this site is less affected by regional emissions compared to surrounding regions, and therefore, can be considered as a continental background mountainous site. In-situ measurements and a model study to analyze the aerosol optical parameters at this site could have the regional representation (Yuan et al., 2013; Chen et al., 2014; Li et al., 2014).

2.2. Aerosol sampling

The data used in this study were collected in a temperature-controlled room during the campaign. A community air-sampling system, which was comprised a stainless steel pipe with a diameter of 3/4 in., put out the window to a height of approximately 8 m aboveground and a vacuum pump that was used to keep the aerosol flow rate at