Science of the Total Environment 622–623 (2018) 481–492

Contents lists available at ScienceDirect

Science of the Total Environment

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

Column-integrated aerosol optical properties of coarse- and fine-mode particles over the Pearl River Delta region in China

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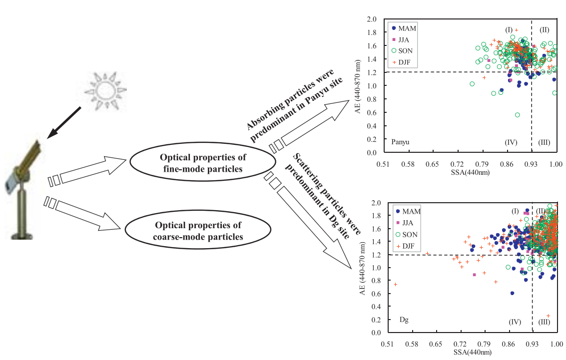
HIGHLIGHTS

- Coarse-mode particles presented low AOD but a strong absorption property for the solar light spectrum.
- Fine-mode absorbing particles were predominant in Guangzhou, fine-mode scattering particles were dominant in Dongguan.
- The aerosol properties over the PRD region were strongly modified by condensation growth.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 9 August 2017
Received in revised form 2 November 2017
Accepted 29 November 2017
Available online xxxx

Editor: Xuexi Tie

Keywords:
Fine- and coarse-mode particles
Aerosol optical properties
Size distribution
Pearl River Delta region

ABSTRACT

The sun-photometer data from 2011 to 2013 at Panyu site (Panyu) and from 2007 to 2013 at Dongguan site (Dg) in the Pearl River Delta region, were used for the retrieving of the aerosol optical depth (AOD), single scattering albedo (SSA), Ångström exponent (AE) and volume size distribution of coarse- and fine-mode particles. The coarse-mode particles presented low AOD (ranging from 0.05 ± 0.03 to 0.08 ± 0.05) but a strong absorption property (SSA ranged from 0.70 ± 0.03 to 0.90 ± 0.02) for the wavelengths between 440 and 1020 nm. However, these coarse particles accounted for < 10% of the total particles. The AOD of fine particles (AODf) was over 3 times as large as that of coarse particles (AODc). The fine particles SSA (SSAf) generally decreased as a function of wavelength, and the relatively lower SSAf value in summer was likely to be due to the stronger solar radiation and higher temperature. More than 70% of the aerosols at Panyu site were dominated by fine-mode absorbing particles, whereas about 70% of the particles at Dg site were attributed to fine-mode scattering particles. The differences of the aerosol optical properties between the two sites are likely associated with local emissions of the light-absorbing carbonaceous aerosols and the scattering aerosols (e.g., sulfate and nitrate particles) caused by the gas-phase oxidation of gaseous precursors (e.g., SO_2 and NO_2). The size distribution exhibited bimodal structures in which the accumulation mode was predominant. The fine-mode volume showed positive dependence on AOD (500 nm), and the growth of peak value of the fine-mode volume was higher than that of the coarse

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https://doi.org/10.1016/j.scitotenv.2017.11.348
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volume. Both the AOD and SSA increased with increasing relative humidity (RH), while the AE decreased with increasing RH. These correlations imply that the aerosol properties are greatly modified by condensation growth.
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1. Introduction

Anthropogenic aerosols affect the wide range of cloud and precipitation systems on Earth by scattering or absorbing solar radiation, or by serving as cloud condensation nuclei or ice nuclei, both of which modify the formation, microphysics, lifetime and the reflectivity of clouds (Seinfeld and Pandis, 2006). This has been shown to significantly influence the radiative and energy budget and even the hydrological cycle of the earth–atmosphere system (IPCC, 2007; Guo et al., 2014; Lee et al., 2016). However, the consideration of aerosol effects in the climate model is still limited (Anderson et al., 2003), mainly due to the large spatial and temporal variability of the physical and chemical properties of aerosols and the non-uniform distribution of the radiative forcing (IPCC, 2013). Therefore, the understanding of the impacts of aerosols on the environment and climate is largely dependent on the investigation of the spatial and temporal distributions of the particles and on the accurate calculation of their optical and radiative properties.

Aerosol optical depth (AOD), Ångström exponent (AE) and single scattering albedo (SSA) are important parameters for describing aerosol optical and radiative properties and they are also critical factors in evaluating the climate effects by anthropogenic and natural aerosol particles. The AOD is usually used to depict the aerosol extinction effect of a vertical column in the cloudless atmosphere, which is determined by not only aerosol extinction coefficient and mass concentration, but also by hygroscopic growth (Yoon and Kim, 2006; Howell et al., 2006). The SSA reflects the scattering and absorption of solar light caused by aerosol loading. It is crucial to precisely understand the SSA of aerosol since small error in its estimation can even change the sign of aerosol radiative forcing (Takemura et al., 2002). The AE is well known as a parameter for describing the volume fraction of fine-mode aerosols. Several field campaigns have been made during recent years largely based on ground-based remote sensing to analyze the spatial and temporal characteristics of AOD, AE and SSA (e.g., Xia et al., 2006; Liu et al., 2012; Che et al., 2014). However, these studies mainly focused on the overall optical properties of the aerosol, and analysis of properties of the coarse- and fine-modes of aerosols is still scarce in most regions of China, including the Pearl River Delta (PRD) region. Various previous studies have shown that the fine particles are more toxic and greater environmental impact than the coarse ones (Schwartz and Neas, 2000). The fine particles from anthropogenic activities made up most of the haze in the North China Plain (Che et al., 2014). In a serious haze pollution episode in this region, the fine-mode AOD was a factor of 20 larger than the coarse-mode AOD, the fine (coarse) mode SSA was relatively consistent (increased) with the wavelength, depending on the particle compositions and their mixture states (Che et al., 2015a). Therefore, the understanding of the optical and radiative properties of the coarse- and fine-modes of particles is more in-depth than that obtained from a generic analysis.

The PRD region is one of the most dynamic urban agglomerations in China. In recent years, great economic increases and social development have occurred in this region. However, a substantial part of this economic growth has relied on the high energy consumption, causing high aerosol emissions and severe air pollution. The air pollution caused by the emissions not only leads to low visibility, but also changes the optical properties of the particles (Chen et al., 2014). The haze pollution in this region is primarily due to fine particle pollution (Wu et al., 2006), of which secondary aerosols are the dominant source (Deng et al., 2013). However, the behavior of optical and radiative properties of the fine- and coarse-modes of particles in this region remains unknown. To the best of our knowledge, information on this topic is limited. Since 2007 there have been two sun-photometers set up in the observational sites of Panyu and Dg, both of which are located in the core area of this region. The main aims of this study were to: 1) analyze the spectral dependence of the coarse and fine-modes of the aerosol in terms of AOD and SSA; 2) determine the dominant particles inferred by the aerosol size and physical and chemical properties; 3) study the volume size distribution; and 4) investigate the water vapor impacts on the aerosol optical properties. Section 2 describes the datasets and methods, and Section 3 presents results and discussion. Conclusions are given in Section 4.

2. Datasets and methods

2.1. Observational sites

The Panyu site (Panyu) is a suburban site about 15 km south of downtown Guangzhou (Fig. 1). It is also a station of the Chinese Aerosol Remote Sensing Network (CARSNET) in the PRD region (Che et al., 2015a, 2015b). This station is located on top of the Dazhengang Mountain in Nancun, Panyu district of Guangzhou (23.00°N, 113.21°E, 140 m above sea level). The observational data represent the average characteristics of uniform mixing of the atmospheric compositions in this region (Wu et al., 2009). The climate is south subtropical monsoon, and the prevailing winds in winter and spring are from directions of NNW–NNE, with the highest frequency of the concurrencies occurring in N direction (Fig. 2). In contrast, the prevailing winds in summer and autumn are from directions of SSE–SE, with the majority of wind originating from the SE region. The mean wind speeds in spring, summer, autumn and winter are 1.69 ± 0.03, 1.84 ± 0.09, 1.79 ± 0.06 and 1.81 ± 0.10 m/s, respectively, with the corresponding mean relative humidity (RH) of 80.81 ± 4.22, 78.49 ± 1.63, 66.78 ± 5.35 and 69.18 ± 4.10% (Table 1).

The Dongguan site (Dg), located in the observational field of the meteorological observatory in Dongguan, is another station of the CARSNET in the PRD region. This station is about 45 km east of the Panyu site, and also belongs to a south subtropical monsoon climate. The annual mean air temperature, sunshine duration and precipitation are 22.7 °C, 1873.7 h and 1819.9 mm, respectively. The prevailing winds are from directions of NNW–NNE and N in winter and spring and from ENE–E and SSW–S in summer and autumn (Fig. 2). The mean wind speeds in spring, summer, autumn and winter are 2.54 ± 0.24, 2.68 ± 0.22, 2.37 ± 0.04 and 2.53 ± 0.16 m/s, respectively, with the mean highest RH occurring in spring (73.45 ± 3.01%) and summer (75.83 ± 2.83%), respectively (Table 1). The results above demonstrate that in the PRD region, both the minimum wind speed and the maximum moisture occurred in spring, which will strongly influence the formation of particles.

2.2. Instruments and methods

The CIMEL sun-photometer at the Panyu site (CE318, France CIMEL Company) has 5 channels (440, 670, 870, 936 and 1020 nm) in the visible to near-infrared bands and 3 polarized channels (870P1, 870P2 and 870P3). The same instrument is provided at the Dg site for direct and sky scanning of the solar radiation, which has no polarization channels. The wavelength of 936 nm was used for calculation of total precipitable water, while the remaining bands were used for the retrievals of the aerosol optical and radiative parameters. The frameworks and retrieval standards were set in accordance with Holben et al. (1998). The cloud scavenging was performed by the scheme from Smirnov et al. (2000). The AOD retrieval was acquired by the sky radiation scheme of

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