Environmental Pollution 210 (2016) 217–226

Contents lists available at ScienceDirect

Environmental Pollution

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

Changes in column aerosol optical properties during extreme haze-fog episodes in January 2013 over urban Beijing☆

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A R T I C L E I N F O

Article history:
Received 15 August 2015
Received in revised form
9 December 2015
Accepted 10 December 2015
Available online xxx

Keywords:
Beijing
Haze-fog episode
AOD
Size distribution
SSA
Radiative forcing

A B S T R A C T

Several dense haze-fog (HF) episodes were occurred in the North China Plain (NCP), especially over Beijing in January 2013 characterized by a long duration, a large influential region, and an extremely high $PM_{2.5}$ values (>500 μg m^{-3}). In this study, we present the characteristics of aerosol optical properties and radiative forcing using Cimel sun-sky radiometer measurements during HF and no haze-fog (NHF) episodes occurred over Beijing during 1–31 January, 2013. The respective maximum values of daily mean aerosol optical depth at 440 nm (AOD_{440}) were observed to be 1.21, 1.43, 1.52, and 2.21 occurred on 12, 14 19, and 28 January. It was found that the Ångström exponent (AE) values were almost higher than 1.0 during all the days with its maximum on 26 January (1.53), suggests the dominance of fine-mode particles. The maximum (minimum) aerosol volume size distributions occurred during dense HF (NHF) days with larger particle volumes of fine-mode. The single scattering albedo, asymmetry parameter, and complex refractive index values during HF events suggest the abundance of fine-mode particles from anthropogenic (absorbing) activities mixed with scattering dust particles. The average shortwave direct aerosol radiative forcing (DARF) values at the bottom-of-atmosphere (BOA) during HF and NHF days were estimated to be 112.29 ± 42.18 W m^{-2} and −58.61 ± 13.09 W m^{-2}, while at the top-of-atmosphere (TOA) the forcing values were −45.78 ± 22.17 W m^{-2} and −18.64 ± 5.84 W m^{-2}, with the corresponding heating rate of 1.61 ± 0.48 K day^{-1} and 1.12 ± 0.31 K day^{-1}, respectively. The DARF values retrieved from the AERONET were in good agreement with the SBDART computed both at the TOA (r = 0.95) and the BOA (r = 0.97) over Beijing in January 2013.

1. Introduction

China, since the 1980s, due to its rapid economic growth has resulted in great increase in anthropogenic emissions, caused serious air pollution problems (Guo et al., 2011; Zhang et al., 2014). Haze-fog (HF) episodes (with visibility < 10 km and high relative humidity (RH) > 80%) occurred frequently in China, in particular over the North China Plain (NCP) and the East China during the last decade (Liu et al., 2013; Xu et al., 2013; Wang et al., 2015). Fine aerosol particles (i.e., $PM_{2.5}$) play a dominant role in the formation and evolution of haze (Liu et al., 2013). High loading of atmospheric pollutants leads to common urban haze over major cities in China, which has greatly threatened public health (Cheng et al., 2013; Wang et al., 2015). A series of laws, regulations, standards, and measures has been implemented to reduce air pollutant emissions and to improve the air quality in China. However, the air pollution control remains a great challenge due to the complex sources and evolution processes of aerosol particles (Quan et al., 2014).

Recently, unprecedented HF events occurred in the NCP, especially over the densely populated urban areas in January 2013. The most prominent feature of this extreme HF pollution was its long lasting, extensive coverage, and high particle concentration (Che et al., 2014a,b; Quan et al., 2014; Tao et al., 2014; Sun et al., 2014, 2015; Wang et al., 2014, 2015). During this period, Beijing experienced several dense HF days and witnessed the most serious episodes of dense HF to date, which caused consternation amongst the public (http://edition.cnn.com/2013/01/19/world/asia/china-florcruz-pollution) (Che et al., 2014a,b; Zhang et al., 2014). The greatest instantaneous concentration of $PM_{2.5}$ reached 1000 μg m^{-3} in some heavily polluted areas of Beijing (Li et al., 2013; Zhang et al.,

☆ This paper has been recommended for acceptance by Eddy Y. Zeng.

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http://dx.doi.org/10.1016/j.envpol.2015.12.021
0269-7491/

2014) and the Yangtze River Delta (Wang et al., 2015). Residents of Beijing and many other cities in China were warned to stay inside in mid-January 2013 as the nation faced one of the worst periods of air quality in the recent history. The Chinese government ordered factories to scale back emissions. Air ports were shut down due to poor visibility, more than 2000 schools were closed, and hundreds of people were hospitalized with respiratory problems (Che et al., 2014b; Sun et al., 2015).

Thus, the present study aims to analyze aerosol optical and radiative properties in terms of aerosol optical depth at 440 nm (AOD_{440}), Angstrom Exponent at 440–870 nm ($AE_{440-870}$), aerosol volume size distribution (VSD), single scattering albedo (SSA), and asymmetry (ASY) parameter, together with the real (Re) and imaginary (Im) parts of refractive index (RI) over Beijing during HF and no haze-fog (NHF) days in January 2013. In addition to aerosol characteristics, shortwave (0.3–4.0 μm) direct aerosol radiative forcing (DARF) values are calculated at the earth's surface/bottom-of-atmosphere (BOA) and at the top-of-atmosphere (TOA) during HF and NHF episodes in January 2013 using the Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model (Ricchiazzi et al., 1998).

2. Materials and methods

2.1. Site description

The measurements were conducted on the terrace of the Institute of Atmospheric Physics (IAP) (Lat: 39.98N, Long: 116.38E; Elev: 92 m asl), Chinese Academy of Sciences' (CAS) in Beijing, which is a 11-storyed building (about 30 m high from the ground level). Beijing, the capital of the People's Republic of China with about 20 million residents, and its population density at the end of 2012 was 1261 people per km^2. In addition, the significant increase in the number of vehicles in operation has led to an increasing number of HF episodes with low visibility days (Che et al., 2014b; Zhang et al., 2014). Numerous studies have been conducted to investigate the sources, compositions, and evolution mechanism of haze pollution over megacities in China (Li et al., 2013; Che et al., 2014a,b; Quan et al., 2014; Tao et al., 2014; Sun et al., 2014, 2015; Wang et al., 2014, 2015; Zhang et al., 2014). The meteorological data recorded over Beijing in January 2013 has been discussed by several previous authors (e.g., Che et al., 2014b; Sun et al., 2015 and references therein) and hence no repetition (see Supplementary data, S1 and Fig. S1).

2.2. Aerosol Robotic network (AERONET)

The Cimel automatic sun and sky radiometer (CE–318), which is a ground-based remote sensing, part of AErosol RObotic NETwork (AERONET), used for measuring direct sun and diffuse sky radiances within the 340–1020 nm and 440–1020 nm spectral ranges, respectively with a 1.2 full field of view (Holben et al., 1998). More details about the instrument, working operation, and data retrieving method have been described by several earlier researchers (Holben et al., 1998; Xia et al., 2006; Alam et al., 2012; Che et al., 2014b; Adesina et al., 2014). The sun measurements were used to accurately calculate the AOD, AE, and water vapor (WV); while spectrally dependent SSA, ASY, complex RI, absorption AE (AAE), and extinction AE (EAE) were obtained from both sky radiance almucantar measurements and direct sun measurements. An inversion algorithm was used to retrieve particle VSD (dV(r)/dlnr) for the column of atmosphere within 22 bins for particle radius (r) from 0.05 to 15 μm. The detailed aerosol properties retrieved were used for calculating broad band fluxes within the spectral range from 0.3 to 4.0 μm.

For the present study, we have used AERONET level 2.0 data (http://aeronet.gsfc.nasa.gov/) from both direct sun and inversion products for the time period during 1–31 January, 2013. The data period is rather typical in terms of pollution suffered from dense HF events and therefore, the study of aerosol optical and radiative properties in this region is of great importance. We used AOD at four wavelengths (440, 675, 880, and 1020 nm) along with $AE_{440-870}$, SSA, ASY, and complex RI. The uncertainty in AOD retrieval under cloud free conditions was $<\pm 0.01$ for wavelengths >440 nm, and $<\pm 0.02$ for shorter wavelengths, which is less than the $\pm 5\%$ uncertainty for the retrieval of sky radiance measurements (Dubovik et al., 2000). SSAs were expected to have an uncertainty of 0.03–0.05 depending on aerosol type and loading; while the errors in RI were estimated to be 30–50% for the Im-RI and ± 0.04 for the Re-RI (Dubovik et al., 2000, 2002; Singh et al., 2004; Alam et al., 2012; Valenzuela et al., 2012). Note that only the SSA and complex RI of almucantar retrievals with $AOD_{440} > 0.4$ were retained in this study, to avoid large inversion errors from the limited information content (Dubovik et al., 2002). The detailed retrieval accuracy, calibration, and uncertainties have been discussed by numerous authors (Dubovik et al., 2002; Singh et al., 2004; Prasad et al., 2007; Alam et al., 2012; Adesina et al., 2014; Valenzuela et al., 2012) and hence not presented here to avoid repetition.

2.3. Estimation of DARF from SBDART model

The direct aerosol radiative forcing (DARF) at the TOA or the BOA is defined as the difference in the net (down minus up) solar flux (solar plus long wave; in W m^{-2}) with and without aerosol, i.e.,

$$\Delta F = (F_{a\downarrow} - F_{a\uparrow}) - (F_{0\downarrow} - F_{0\uparrow}) \tag{1}$$

where ΔF denotes the net irradiance (downwelling minus upwelling; in W m^{-2}) and F_a and F_0 denote the global irradiances with and without aerosols, respectively either at the TOA or the BOA. The arrows indicate the direction of the global irradiances (down↓ and up↑).

In this study, we have chosen the atmosphere without aerosols as the reference case. Thus, we have computed the net flux at the TOA and the BOA separately, within the wavelength range from 0.3 to 4.0 μm, both with and without aerosols, using the SBDART model (Ricchiazzi et al., 1998), which is a discrete ordinates radiative transfer model. This model was developed by the atmospheric science community and has been widely used for radiative transfer calculations by several authors (e.g., Alam et al., 2012, 2014; Adesina et al., 2014; Prasad et al., 2007; Valenzuela et al., 2012). Based on the prevailing weather conditions and measured parameters, we used the 'Mid-latitude winter' atmospheric model. The optical parameters used for the DARF estimations were the spectral values of AOD, SSA, and ASY obtained from the Cimel Sunphotometer of Beijing AERONET site. With respect to the vertical distribution of aerosol, we used the SBDART profile, which takes into account the aerosol-loaded atmosphere fitting an exponential decay to the AOD derived by Sunphotometer. Other input parameters in the model include solar zenith angle, which is calculated using a small code in the SBDART model by specifying a particular date, time, latitude, and longitude. The total column ozone concentration and surface albedo values were obtained from the Ozone Monitoring Instrument (OMI) onboard NASA's Aura satellite reflectivity data through the Giovanni online data system, developed and maintained by the NASA GES DISC (http://disc.sci.gsfc.nasa.gov/giovanni).

The atmospheric rate (HR) was computed for each layer, based on finite difference estimates of the irradiance divergence at each pair of levels (Liou, 2002):