



Variability in aerosol optical properties over an urban site, Kanpur, in the Indo-Gangetic Plain: A case study of haze and dust events



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ABSTRACT

In this study, we report on three important optical parameters, viz. absorption and scattering coefficients (b_{abs} , b_{scat}) and single scattering albedo (SSA) based on one-year chemical-composition data collected from an urban site (Kanpur) in the Indo-Gangetic-Plain (IGP) of northern India. In addition, absorption Ångström exponent (AAE) was also estimated in order to understand the wavelength dependence of absorption and to decipher emission sources of carbonaceous aerosols, in particular of black carbon. The absorption and scattering coefficients ranged between 8.3 to 95.2 Mm^{-1} ($1 \text{ Mm}^{-1} = 10^{-6} \text{ m}^{-1}$) and 58 to 564 Mm^{-1} , respectively during the study period (for $n = 66$; from January 2007 to March 2008) and exhibit large seasonal variability with higher values occurring in winter and lower in the summer. Single scattering albedo varied from 0.65 to 0.92 whereas AAE ranged from 0.79 to 1.40 during pre-monsoon and winter seasons, respectively. The strong seasonal variability in aerosol optical properties is attributed to varying contribution from different emission sources of carbonaceous aerosols in the IGP. A case study of haze and dust events further provide information on extreme variability in aerosol optical parameters, particularly SSA, a crucial parameter in atmospheric radiative forcing estimates.

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1. Introduction

The ubiquitous presence of aerosols in the atmosphere affects radiation budget of the Earth, directly by scattering and absorbing solar radiation, or indirectly through modification of microphysical properties and thus, influences climate on a local as well as global scale (IPCC, 2013). The impacts of aerosols on radiation budget, hydrological cycle and climate are significant, but remain highly uncertain (Gadhavi and Jayaraman, 2010; IPCC, 2013; Menon et al., 2002a; Menon et al., 2002b; Ramanathan et al., 2001; Satheesh and Moorthy, 2005). The abundance of atmospheric aerosol and its constituents, their physico-chemical and optical properties are highly variable with respect to time and space (Ram and Sarin, 2010; Ram et al., 2012b; Ramanathan and Carmichael, 2008; Satheesh and Moorthy, 2005). Carbonaceous aerosols, an ubiquitous component of atmospheric aerosols, can contribute about 20–70% of particulate matter over urban environment and are mainly associated with fine-fraction of aerosols (i.e. aerodynamic diameter $\leq 2.5 \mu\text{m}$, defined as $\text{PM}_{2.5}$ aerosols) (Fuzzi et al., 2006; Jimenez et al., 2009; Ram and Sarin, 2011; Ram and Sarin, 2015). Carbonaceous aerosol play a significant role in regional atmospheric chemistry, visibility and air-quality (Ram et al., 2012a; Rengarajan et al.,

2011; Srivastava et al., 2014) and also contribute to the formation of haze and fog events (Huang et al., 2014; Kang et al., 2013; Ram and Sarin, 2011; Ram et al., 2012a; Tan et al., 2016; Tiwari et al., 2014).

Carbonaceous aerosols mainly include organic aerosols (OA) which account for the majority of aerosol mass whereas contribution of elemental carbon (EC), or black carbon (BC), is relatively low ($\leq 10\%$) (Ram and Sarin, 2010; Ram et al., 2010b; Ram et al., 2014; Tripathi et al., 2005). Although, both organic carbon (OC) and EC are emitted from the same emission sources, they have different optical, chemical and physical properties (Lan et al., 2013; Ram and Sarin, 2009; Schwarz et al., 2008). Black carbon (BC) is also called as elemental carbon or graphitic carbon and is a principal light absorbing species in atmospheric aerosols. However, light absorption due to BC varies temporally and geographically depending on emission sources and their emission strength (Andreae and Gelencser, 2006; Bond and Bergstrom, 2006; Bond et al., 2013; Bond et al., 2006; Ram and Sarin, 2009).

Asia is one of the most polluted regions in the world and is often characterized by atmospheric brown cloud formation in the Indo-Gangetic plain (IGP), the Himalayas and south-east Asia (Ramanathan et al., 2005). Aerosol optical properties exhibit a large spatial and temporal variation depending on aerosol composition, type of aerosols sources, prevailing meteorological conditions, ageing and transport of aerosols (Dey and Tripathi, 2007; Ram and Sarin, 2010; Ram and

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Sarin, 2012; Ram and Sarin, 2015). Light absorbing aerosol species enhances global warming by supplementing to the greenhouse effect, however the assessment of their climatic impact still poses a greater challenge with relatively large uncertainty (Gadhavi and Jayaraman, 2010). It is, thus, important to assess aerosol optical properties and their climatic effect on a regional scale with a combination of real-time measurements as well as satellite retrievals. The present study aims to evaluate a few crucial optical properties of aerosols, namely absorption and scattering coefficients (b_{abs} and b_{scat} , respectively) and single scattering albedo (SSA) using a one-year chemical composition data of PM_{10} samples ($n = 66$; January 2007 to March 2008) collected from an urban location, Kanpur, in the Gangetic plain of northern India. In addition, we also studied the wavelength dependence of aerosol absorption coefficient and estimated absorption Ångström exponent (α ; AAE) for different seasons to decipher emission sources of aerosols. Finally, aerosol optical parameters during haze and dust events, as a case study, have been examined with the help of real-time measurements as well network of ground-based sun photometers AERONET (Aerosol Robotic Network) at Kanpur.

2. Methodology

2.1. Ambient aerosol sampling

PM_{10} samples (particulate matter with aerodynamic diameter $\leq 10 \mu\text{m}$) were collected, during January 2007–March 2008, from Kanpur using a high-volume sampler (APM 450, Environtech Pvt. Ltd., New Delhi, India), operated at a flow rate of $1.0 \pm 0.1 \text{ m}^3 \text{ min}^{-1}$. A total of 66 PM_{10} samples were collected during the study period and details of aerosol sampling are described in Ram et al. (2010b). Of these, 22 samples were collected during wintertime (December–February), 9 samples during March, 25 samples during summer (April–June) and 10 samples during post-monsoon (October–November). July–September months represent the wet-season when south-west monsoon rain causes efficient washout of the atmosphere and hence, no aerosol samples were collected during July–September period (Ram et al., 2010b).

2.2. Site description and meteorological details

The sampling site, Kanpur (26.5°N , 80.3°E , 142 m above mean sea level), is located in the central part of the IGP. The site is mostly influenced by large-scale anthropogenic emission sources (biomass burning, fossil-fuel combustion and industrial activities) within the IGP and mineral dust transported from western India which directly influence optical properties of aerosols at Kanpur (Ram et al., 2010b). Due to shallow boundary layer in the wintertime and the Himalayan mountains parallel to the IGP, aerosols are confined to lower atmosphere resulting in a high concentration of anthropogenic aerosols (Ram and Sarin, 2011; Ram and Sarin, 2012; Ram and Sarin, 2015; Ram et al., 2010b). The average fractional contribution of carbonaceous aerosols in the IGP is about 30–35% of PM_{10} ; however, it can be as high as 70% of $\text{PM}_{2.5}$ during the wintertime (Ram and Sarin, 2011; Ram et al., 2010b). It is noteworthy to mention here that variability in aerosol composition, absorption and scattering properties over IGP is important to decipher climate-relevant optical properties and radiative forcing (Ram et al., 2012b).

2.3. Chemical composition of aerosol (EC, OC, Ionic species)

An aliquot of filter paper (area: 1.5 cm^2) was used for the measurement of EC, OC concentration on a thermo-optical EC-OC analyzer (Sunset laboratory Inc., USA) using NIOSH-5040 (National Institute of Occupational Safety and Health) based on thermal-optical transmittance (TOT) protocol (Birch and Cary, 1996; Ram et al., 2008). The analytical procedure of EC, OC consists of two stages of heating in an inert and an oxidizing medium. The first stage includes volatilization of OC

in a non-oxidizing medium through stepwise heating. In the second stage, the oven is subjected to a stepwise temperature increase (up to 900°C) in an oxidizing condition where all the fractions of pyrolyzed carbon and EC are converted to CO_2 . The evolved CO_2 in each volatilization step was converted to methane and were measured using a flame ionization detector. The thermograph obtained in the first and second stages, when corrected for pyrolyzed carbon, gives OC and EC fractions, respectively. The initial absorbance at 678 nm was used to define the split-point between OC and EC. All the reported OC concentrations are corrected for pyrolyzed and carbonate carbon (CC). The detection limits for EC and OC was reported to be 0.01 and $0.3 \mu\text{g m}^{-3}$, respectively whereas the average blank concentration for OC is $1.8 \pm 0.2 \mu\text{g m}^{-3}$ ($n = 8$). Replicate analysis of samples indicated good analytical precision, with relative percent deviations $< 3.0\%$ for OC and better than 5.0% for EC analysis ($n = 16$) (Ram et al., 2010b).

For water-soluble ionic species, one-fourth of filter area ($\sim 105 \text{ cm}^2$) is soaked in 50 ml milli-Q water (resistivity $18.2 \text{ M}\Omega \text{ cm}$) for approximately 6–8 h. An intermittent ultrasonic treatment was performed to disintegrate aerosol particles from the filter. An aliquot of water-extract was used to determine concentration of water-soluble ionic species using an ion-chromatograph. For separating anions, 1.8 mM $\text{Na}_2\text{CO}_3/1.7 \text{ mM NaHCO}_3$ eluent was used and for cations 20 mM methanesulfonic acid was used as an eluent. The replicate analysis provides a precision of better than 5% for ionic species and the concentration of blank filter was subtracted to assess the concentration of cations and anions. The analytical details of water-soluble ionic species have been described elsewhere (Ram et al., 2010b; Rengarajan et al., 2007).

2.4. Assessment of aerosol absorption and scattering coefficient

Aerosol absorption coefficient (b_{abs}) was estimated at 678 nm with the help of measured optical-attenuation (ATN) as per the methodology provided in an earlier publication (Ram and Sarin, 2009). The total uncertainty in the estimated b_{abs} values is of the order of $\sim 23\%$ arising from the ATN, area, volume of air sampled, multiple scattering and shadowing corrections (Ram and Sarin, 2009).

Scattering coefficient (b_{scat}) is calculated using the following Eq. (1):

$$b_{\text{scat}} = \sum M_i * \text{MSC}_i \quad (1)$$

where M_i represents mass concentrations ($\mu\text{g m}^{-3}$) of $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3 , organic matter (OM) and BC, and MSC_i is the mass scattering cross-section (in $\text{m}^2 \text{g}^{-1}$) of the chemical species (Bond and Bergstrom, 2006; Magi, 2009). Eq. (1) can be rewritten as

$$b_{\text{scat}} = M_{\text{ionic}} * \text{MSC}_{\text{ionic}} + M_{\text{carbon}} * \text{MSC}_{\text{carbon}} \quad (2)$$

In Eq. (2), M_{ionic} represents sum of the mass concentrations of $(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3 whereas M_{carbon} represents sum of mass concentrations of OM and BC (Magi, 2009, 2011). The mass concentration of SO_4^{2-} and NO_3^- are converted to $(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3 for calculating the scattering coefficient (Magi, 2009). A factor of 1.6 is used to convert OC to OM (Ram et al., 2010b; Rengarajan et al., 2007). Scattering coefficient was initially estimated at 550 nm in the present study as mass scattering cross-sections are reported at 550 nm and latter extrapolated to 678 nm in order to match the wavelength of absorption coefficient (Levin et al., 2010). The values of MSCs for different chemical species, used in the present study, are listed in Table 1.

2.5. Assessment of single scattering albedo (SSA; Ω)

Single scattering albedo is an important parameter which explains absorbing/scattering nature of aerosols and is used in the estimation of direct aerosol radiative forcing. The scattering and absorption characteristics of aerosols in combination with surface reflectance determine

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