Proc Indian natn Sci Acad **78** No. 3 September 2012 pp. 523-533

Status Report

Carbonaceous Aerosols Over Northern India: Sources and Spatio-temporal Variability

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This article presents a synthesis of the chemical and optical properties of ambient aerosols studied from selected sites (high altitude and urban) in Northern India. Atmospheric carbonaceous and water-soluble inorganic species account for nearly two-third of the total aerosol mass during the winter time, whereas their contribution is only 35 % during summer. The mass concentrations of carbonaceous species (EC, OC and WSOC) at urban sites are an order of magnitude higher than those at a high altitude site. Several factors, namely source variability, emission strength, secondary aerosol formation and boundary layer dynamics, contribute to the atmospheric concentrations of carbonaceous species and their temporal variability. Based on diagnostic ratios, K^+/OC and OC/EC, biomass burning emission (wood-fuel and post-harvest agricultural-waste burning) can be identified as major sources of carbonaceous aerosols. The large variability in the WSOC/OC ratio suggests significant contribution from secondary organic aerosols. The formation of secondary aerosols and their hygroscopic growth has implications to the scattering properties of aerosols, a process that can be invoked for the fog-haze formation during the wintertime over Northern India.

Key Words : Northern India; Atmospheric Aerosols; Biomass Burning Emission; Carbonaceous Species; Temporal Variability

Introduction

Atmospheric aerosols, an important component of the atmosphere, are derived from a variety of sources (natural and anthropogenic) and comprise of wide-range of particles having different chemical composition, size (0.001 μm to 100 μm), shape and optical properties. They are mainly confined to the lower troposphere wherein intense vertical and horizontal mixing takes place. As a consequence, large-scale temporal and spatial heterogeneity in aerosol loadings of the atmosphere is observed. The abundance of aerosols in the atmosphere is either quantified by their total mass concentration (μg m^{-3}) or by an optical measurement referred to as aerosol optical depth (AOD). Atmospheric aerosols have short residence time (few days to a week) and are projected to have a regional to global impact on the radiation budget. Atmospheric aerosols produce a net cooling effect (negative forcing), however these estimates are associated with large degree of uncertainty and the present level of scientific understanding still remains in the range-medium to low (IPCC, 2007). The large uncertainty in the estimation of radiative forcing is partially attributed to the lack of knowledge on the spatio-temporal variability of chemical composition and optical properties of absorbing and scattering aerosols on a regional scale. A recent study suggests that uncertainty in the estimation of radiative forcing is due to relative increase in the mass concentrations of absorbing black carbon (BC) which is considered to be larger than the overall increase in the abundance of scattering anthropogenic aerosols (Myhre, 2009).

Carbonaceous aerosols, a ubiquitous component of Earth' atmosphere contribute ~20-70% of total aerosol mass over urban regions (De Gouw and Jimenez, 2009; Ramachandran *et al*., 2009), and are known to influence the solar radiation budget, hydrological cycle and climate on a regional to global scale via direct and indirect effects (Menon *et al*., 2002; Srivastava *et al*., 2012; Venkataraman *et al*., 2005). In addition, several hazardous gases (e.g. carbon mono-oxide) and un-combusted hydrocarbons (volatile organic compounds, VOC and polycyclic aromatic hydrocarbons, PAH) are produced during emissions of carbonaceous aerosols and thus, affect respiratory, lung systems and human health (Lighty *et al*., 2000; Nel, 2005).

Carbonaceous aerosols are derived from two major sources: those directly emitted from the biomass burning emission, vehicular and coal-based industries (referred as primary organic aerosols, POA) and those formed in the atmosphere from the volatile-organic compounds (VOCs) via gas-phase oxidation with oxidizing agents (referred as secondary organic aerosols, SOA, Jimenez *et al*., 2009).

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Carbonaceous aerosols derived from biomass burning (BB) emission are enriched in organic carbon (OC) compared to elemental carbon (EC), thus, providing relatively higher OC/EC ratios. In contrast, vehicular emissions produce relatively small fraction of OC and large EC resulting in lower OC/EC ratios. Hence, OC/EC ratios can be used as a tracer for biomass burning and vehicular emissions, provided secondary organic carbon contribution is negligible.

Considering large spatio-temporal variability in aerosol chemical composition associated with the emission, regional meteorology and transport processes, in-situ measurements of aerosol chemical composition and optical properties can be used for the better estimation of direct aerosol radiative forcing on a regional scale. Furthermore, ground and long-term measurements of atmospheric chemical constituents can also be used for the validation of aerosol optical properties retrieved from the satellite data.

A number of field studies, based on land campaigns and ship-borne measurements, have documented strong spatio-temporal variability in aerosol chemical composition, optical properties and radiative forcing over the Indian region (Aloysius *et al*., 2008; Das *et al*., 2009; Das *et al*., 2008; Ganguly *et al*., 2006; Kumar *et al*., 2010; Kumar *et al*., 2008a; Kumar *et al*., 2008b; Moorthy *et al*., 2005; Nair *et al*., 2007; Niranjan *et al*., 2006; Pant *et al*., 2006; Ram and Sarin, 2009, 2010; Ramachandran *et al*., 2006; Raman *et al*., 2011; Rengarajan *et al*., 2007; Sharma *et al*., 2000; Srivastava *et al*., 2009; Sudheer and Sarin, 2008; Tare *et al*., 2006; Tripathi *et al*., 2006). As a part of Indian Space Research Organization-Geosphere Biosphere Programme (ISRO-GBP), observations made from urban sites (Hisar, Allahabad), a high-altitude site (Manora Peak), during December, 2004, have provided some insight on the aerosols chemical composition over the Indo-Gangetic Plain (IGP) (Kulshrestha *et al*., 2009; Lal *et al*., 2008; Ram *et al*., 2012b; Raman *et al*., 2011; Rengarajan *et al*., 2007).

However, a systematic long-term simultaneous data on atmospheric concentrations of EC, OC and water-soluble organic carbon (WSOC) from Northern India are lacking in the literature. Recent studies on carbonaceous aerosols, absorption properties and chemical composition over urban (Kanpur: Jan 2007-March 2008 and Patiala: Oct 2008 to May 2009) and high-altitude sites (Mt Abu: May 2005-Feb 2006 and Manora Peak: Feb 2005-July 2008) in northern India (Rajput *et al*., 2011; Ram *et al*., 2012a; Ram and Sarin, 2009, 2010, 2011; Ram *et al*., 2008, 2010a; Ram *et al*., 2012b; Ram *et al*., 2010b, c) fill this gap to some extent. The objectives of these short-term campaign and long-term systematic studies are to understand spatio-temporal variability in optical properties, mass concentration and chemical composition of ambient aerosols. In this article, we review studies on the chemical composition of aerosols from selected locations in India with the major emphasis on the characterization of carbonaceous aerosols and their absorption properties over India.

Results

Aerosol Chemical Composition

The chemical composition and optical properties of atmospheric aerosols are highly variable in space and time due to the changes in the source characteristics (types and emission strength), meteorological conditions and transport of aerosols. The study on the chemical characterization of ambient aerosols provides an effective tool to understand the formation of secondary aerosols, atmospheric chemical processes, transport and the aging of aerosols. For example, studies over Indian regions suggest that carbonaceous aerosols contribute to the majority (~30-60%) of the total aerosol mass followed by water-soluble inorganic species (WSIS) and mineral aerosols over urban and rural atmospheres (Behera and Sharma, 2010b; Krishna, 2012; Rajput *et al*., 2011; Ram *et al*., 2012a; Ram and Sarin, 2010, 2011; Ram *et al*., 2010b; Rengarajan *et al*., 2007; Rengarajan *et al*., 2011; Satheesh, 2012; Tare *et al*., 2006). Aerosols at high-altitude are governed by the long-range transport and hence, their chemical composition is governed by the transport patterns, secondary formation and chemical processing in the atmosphere (Carrico *et al*., 2003; Cong *et al*., 2008; Cozic *et al*., 2008; Decesari *et al*., 2010; Hegde *et al*., 2007; Kumar and Sarin, 2009; Ram *et al*., 2008, 2010a). Fig. 1 shows the chemical composition of aerosols collected at urban and high-altitude sites in Northern India.

The PM_{10} mass concentration exhibits a large temporal variability and ranges from 40 to 310 μg m^{-3} during a one-year sampling period at Kanpur (Ram *et al*., 2010b). Total carbonaceous aerosols (TCA) and water-soluble inorganic species contribute nearly two-third of the PM_{10} mass during wintertime whereas their contribution is only ~35% during summer (Ram *et al*., 2010b). Total carbonaceous aerosol (TCA) abundance is estimated by summing up organic matter (OM) and elemental carbon (EC) concentrations. The OM abundance is taken as 1.6 times that of organic carbon measured in aerosols (i.e. OM=1.6 × OC). Although, a wide range of values for OM/OC ratios (1.2 to 2.1) have been reported in the literature (Turpin and Lim, 2001), a value of 1.6 for OM/OC ratio have been suggested for urban aerosols (Cao *et al*., 2003; Ram and Sarin, 2011; Rengarajan *et al*., 2007; Rengarajan *et al*., 2011). The contribution of TCA was as high as 60% of the PM_{10} mass for some of the samples collected during winter months. The boundary layer dynamics, varying sources and their emission strength, secondary aerosol

formation and transport of aerosols all contribute to the seasonal trend in aerosol mass concentrations at Kanpur (Ram *et al*., 2010b). Based on the chemical tracers (K^+ concentrations, K^+/OC and OC/EC ratios), biomass burning emission (wood-fuels and agricultural waste) has been identified as a major source of carbonaceous aerosols at urban and high-altitude sites in northern India (Ram and Sarin, 2010, 2011; Ram *et al*., 2010a; Ram *et al*., 2010b).

At high-altitude sites, the total suspended particulate (TSP) mass also exhibits large temporal variability; varying from 15 to 430 μg m^{-3} (at Mt Abu) and 15 to 270 μg m^{-3} (at Manora Peak) (Ram *et al*., 2008, 2010a). Simultaneous measurements of aerosol optical depth (AOD), at Manora Peak, also exhibit a significant increase during summer months (Ram *et al*., 2010a; Sagar *et al*., 2004). On annual-scale, TCA and WSIS contribute nearly 25% and 10% of TSP mass at Manora Peak, respectively (Fig. 1). The chemical analyses of ambient aerosols from high-altitude sites suggested the dominance of mineral aerosols throughout the sampling period (Carrico *et al*., 2003; Decesari *et al*., 2010; Hegde *et al*., 2007; Kumar and Sarin, 2009; Ram *et al*., 2008, 2010a; Rastogi and Sarin, 2005). However, the dominance of mineral aerosols (to TSP and AOD values) is significantly pronounced during summer months (April-June) under the prevailing south-westerly winds when long-range transport of aerosols originating from the desert regions in the middle-East and the Thar Desert (in western India) (Ram *et al*., 2008, 2010a; Rastogi and Sarin, 2006, 2009). The long-range transport of mineral aerosols was established with the help of water-soluble Ca (Ca^{2+}) in aerosols and back trajectory analysis of the air-masses reaching at respective sampling locations (Carrico *et al*., 2003; Decesari *et al*., 2010; Ram *et al*., 2010a).

Carbonaceous Species Measurements in Aerosols

The determination of light absorbing carbon (LAC) mass based on optical (e.g. Aethalometer, particle soot absorption photometer, PSAP) and a thermal method (e.g. thermo-optical EC-OC analyzer) is referred as BC and EC, respectively. However, measurement of LAC mass by two different methods suffers from large degree of uncertainty and difference in mass concentration by a factor of two to four (or even more) is not very uncommon (Hitzenberger *et al*., 2006; Ram *et al*., 2010c). Thus, an absolute method for the accurate determination of LAC by optical method is still lacking (Reisinger *et al*., 2008). However, recent studies suggest that BC measurement by a single particle soot photometer (SP2) provides reliable quantification of light absorbing carbon mass (Gao *et al*., 2007; Moteki and Kondo, 2007; Schwarz *et al*., 2006; Schwarz *et al*., 2008). In the following sections, we will discuss the measurements of mass of light absorbing carbon based on optical (i.e. Aethalometer) and thermo-optical method.

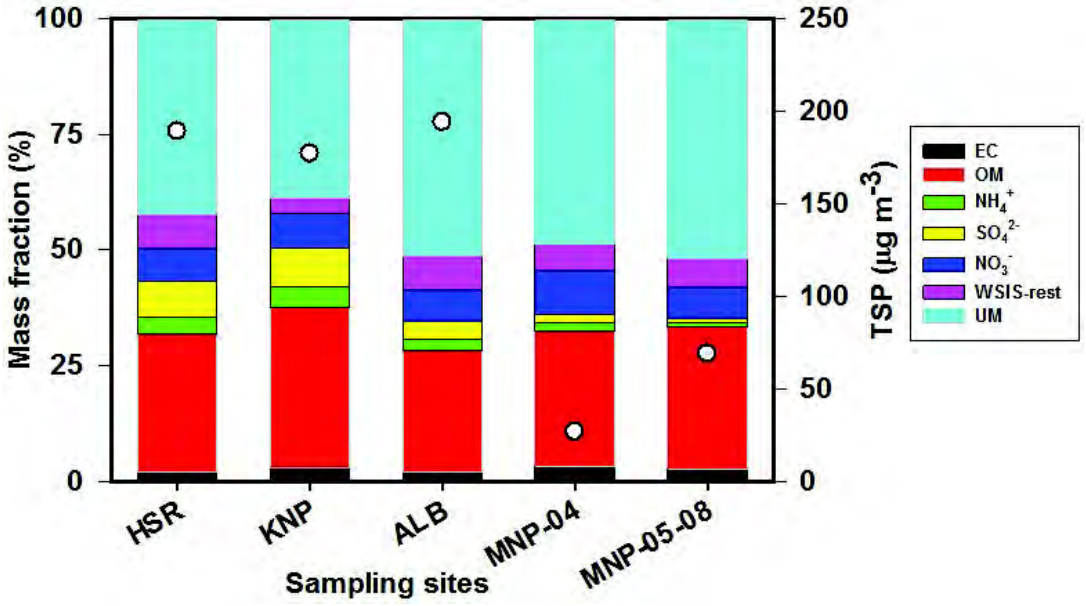

Fig. 1: Chemical composition of aerosols collected at urban and high-altitude sites in India. OM refers to the organic matter (=f × OC), WSIS-rest (sum of Cl^-, K^+, Mg^{2+} and Na^+) and unaccounted mass (UM) is defined as TSP – OM – EC – WSIS-rest. The OC to OM conversion factor (f) was taken as 1.6 for urban and 1.8 for high-altitude site. The open circles (y-axis scale) represent TSP mass for HSR (Hisar; Dec 2004), ALB (Allahabad; Dec 2004) and MNP (Manora Peak), and PM_{10} mass for KNP (Kanpur; 2007-2008). The chemical composition at MNP for Dec 2004 campaign (MNP-04) and that for years 2005-2008 (MNP-05-08) is shown separately

Aethalometer Based Black Carbon (BC) Measurements

The INDOEX (Indian Ocean Experiment) study had reported the presence of high level of pollutants over the Indian Ocean, enriched in BC and transport of aerosols from south and South-east Asia during the wintertime (Jan-March 1999) (Lelieveld *et al*., 2001; Neusüß *et al*., 2002; Venkataraman *et al*., 2002). Based on these data, Satheesh *et al*. (1999) developed an aerosol model for tropical Indian Ocean and found that BC mass alone contributes 11% to composite aerosol optical depth during INDOEX study. More recently, Ramanathan *et al*. (2007) suggested that warming trends in the lower atmosphere of vertically extended atmospheric brown clouds (ABC) over the Indian Ocean could be equivalent to the recent increase in the greenhouse gases.

The measurements of black carbon (BC), using an Aethalometer, have been extensively performed over Indian regions to document the spatio-temporal variability (Babu and Moorthy, 2002; Babu *et al*., 2002; Ganguly *et al*., 2006; Latha and Badarinath, 2003; Ramachandran and Rajesh, 2007; Tripathi *et al*., 2005a; Tripathi *et al*., 2005b). Over urban locations, BC concentrations are generally less than 15 µgC m^{-3}, however, it can be as high as 60 µgC m^{-3} at some locations (Ganguly *et al*., 2006; Latha and Badarinath, 2003). During the ISRO-GBP land campaign-I (LC-I), carried out in wintertime (Dec 2004) over IGP, extensive measurements of BC mass concentration along with several other chemical and optical properties were preformed (Nair *et al*., 2007; Ramachandran *et al*., 2006; Rengarajan *et al*., 2007; Tare *et al*., 2006; Tripathi *et al*., 2005a).

Tripathi *et al*. (2005a) reported that BC mass concentration at Kanpur varied from 6 to 20 µgC m^{-3} during the field campaign (December 2004) and gave a low value of 0.76 for single scattering albedo (SSA). During the same field campaign, Ganguly *et al*. (2006) reported that BC mass concentration was as high as 60 µgC m^{-3} at Delhi (an urban location) with an average value of 29±14 µgC m^{-3} resulting in further lower value of 0.68 for SSA. Although, EC constitutes only a minor part of PM_{10} mass, it is one of the major absorbing particulate species in atmospheric aerosols. The EC/PM_{10} mass ratio, thus, provides a qualitative assessment for the absorbing nature of aerosols and helps understand the radiative impact of EC. In general, EC/PM_{10} ratio varies from 3-10% over urban locations in India (Babu and Moorthy, 2002; Babu *et al*., 2002), but it can be as high as 15% during wintertime in some parts of the IGP (Ganguly *et al*., 2006; Tripathi *et al*., 2005a).

Thermo-optical Measurements of Mass Concentrations of OC and EC

Aethalometer based measurements of BC do not provide any information on the mass concentrations of OC and WSOC. It is important to note that organic carbon is a major constituent of carbonaceous aerosol at urban locations and contributes significantly to the PM_{10} mass during wintertime. However, very few measurements of carbonaceous aerosols, especially OC and WSOC, are available over Indian regions. More recently, substantial efforts have been made in characterizing EC, OC and WSOC, OC/EC and WSOC/OC ratios in ambient aerosols over a few locations in India (mainly in northern India) (Rajput *et al*., 2011; Ram *et al*., 2012a; Ram and Sarin, 2009, 2010, 2011; Ram *et al*., 2010a; Ram *et al*., 2010b; Rengarajan *et al*., 2007). Systematic and long-term measurements on atmospheric concentrations of EC, OC and WSOC, along with selected optical properties were made available over northern India. Fig. 2 shows spatial variability of average mass concentrations of OC and EC at high-altitude, urban sites and Oceanic regions. In

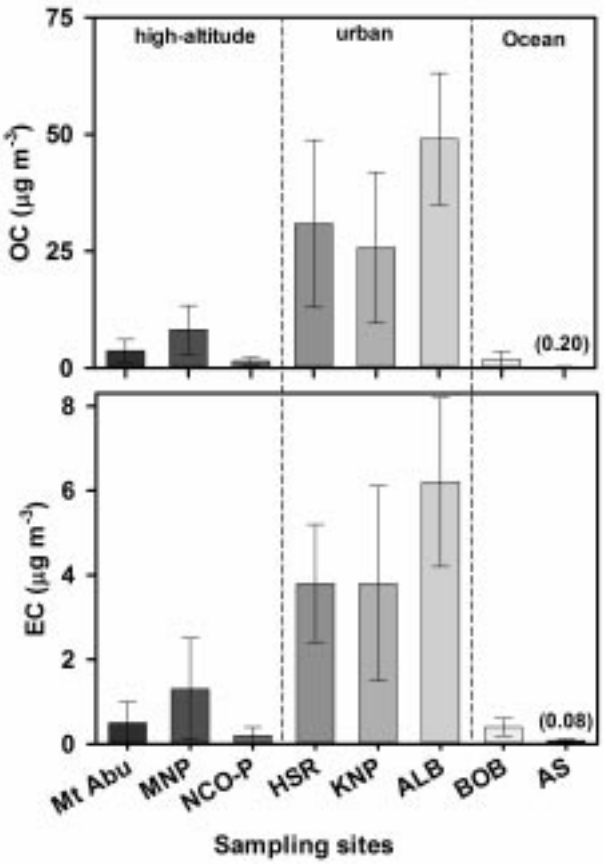

Fig. 2: Spatial variability of mass concentrations of OC and EC at high-altitude, urban, rural sites and Oceanic regions. The sites are abbreviated as: MNP (Manora Peak), NCO-P (Nepal Climate Observatory-Pyramid), HSR (Hisar), KNP (Kanpur), ALB (Allahabad), BOB (Bay of Bengal) and AS (Arabian Sea). The concentrations of EC and OC over Arabian Sea are very low and the values are given in the parenthesis

the following sections, we discuss the spatiotemporal variability in the mass concentrations of OC, EC, OC/EC and WSOC/OC ratios over northern and western India, high-altitude sites and oceanic regions separately.

Northern India

The mass concentrations of OC, EC and WSOC at urban sites in the IGP are an order of magnitude higher is than those at the high-altitude sites (Ram *et al*., 2012a; Ram and Sarin, 2010; Ram *et al*., 2008, 2010a; Ram *et al*., 2010b). In contrast, OC concentration is nearly uniform at urban sites with the highest value of OC at Allahabad. A recent data over the two winter seasons (Jan-Feb 2007 and Dec-Feb 2008) from Kanpur, an urban site at the centre of the IGP, also exhibits similar abundance patterns for OC and EC (as observed during Dec 2007, Ram *et al*., 2010b).

Enhancement in their mass concentrations of carbonaceous species is largest during wintertime when biomass burning emission strength is highest and a shallow boundary layer height helps in trapping the aerosols (Ram *et al*., 2010b). Increase in the biomass burning emission strength is also reflected by the seasonal trend in the mass concentrations of OC and EC at Kanpur when concentrations are a factor of two-to-three higher in the wintertime (compared to summer months) (Ram *et al*., 2012a). However, OC and EC mass concentrations at Kanpur are relatively lower than those reported at Agra (Satsangi *et al*., 2010). The average concentrations of OC and EC are 60.9±40.5 μg m^{-3} (range: 20.4 to 147.4 μg m^{-3}) and 7.5±4.6 μg m^{-3} (range: 1.4 to 20.7 μg m^{-3}), respectively at the suburban site (Agra) during Jan-Oct 2009 (Satsangi *et al*., 2010). A characteristic feature of the carbonaceous species in aerosol samples from the urban and high altitude sites is also reflected in the OC/EC ratios (Av=7.8 ± 3.4) (Fig. 3). This is in sharp contrast to their mass ratio from a rural site, influenced by coal-based emissions, in the north-eastern part of India (range: 2.1-4.0, Av=3.1 ± 0.6, n=7) (Ram and Sarin, 2010; Ram *et al*., 2010b).

Western India

The mass concentrations of OC and EC varied from 4.6 to 28.0 μg m^{-3} and 0.3 to 4.4 μg m^{-3}, respectively over an urban site (Ahmedabad) for the samples collected during Jan-Dec 2003 (Rastogi and Sarin, 2009). In a recent study, Rengarajan *et al*. (2011) reported that OC and EC concentrations varied from 11 to 39 μg m^{-3} and 1.5 to 5.7 μg m^{-3} during wintertime (Dec 2006-Jan 2007) at Ahmedabad. In contrast, the mass concentrations of OC and EC were relatively lower during summer months (Mar-June 2007) and ranged from 4.5 to 18.9 μg m^{-3} (av.: 7.9 μg m^{-3}) and 0.51-3.1 μg m^{-3} (av.: 0.99 μg m^{-3}), respectively at Ahmedabad. The mass concentrations as well as

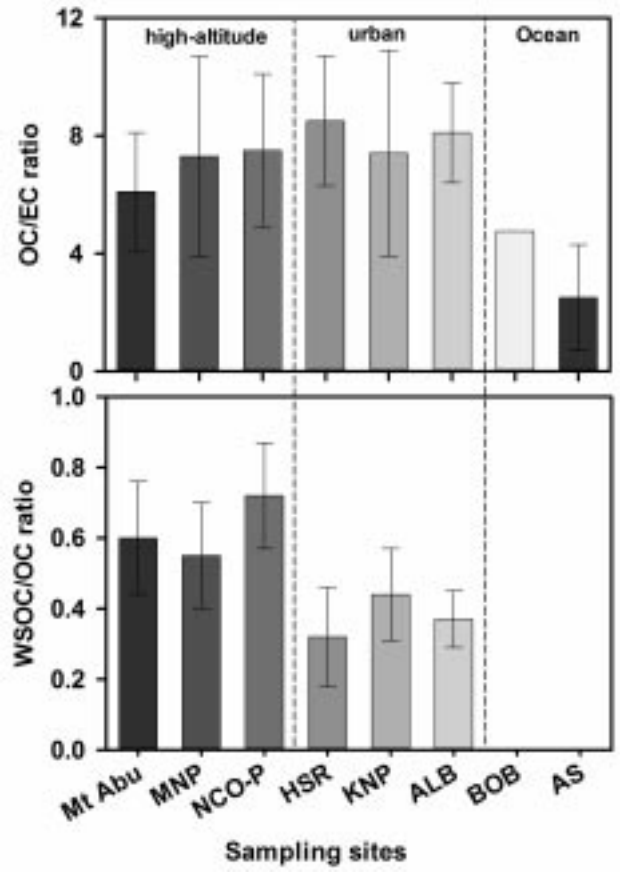

Fig. 3: Spatial variability of characteristics OC/EC and WSOC/OC ratios at high-altitude, urban, rural sites and Oceanic regions. Sampling sites abbreviation is same as given shown in Fig. 2.

fractional contributions of OC and EC at Ahmedabad are relatively lower than the urban sites in the IGP. However, OC/EC ratios at Ahmedabad are comparable to the urban sites in the IGP. For example, Rengarajan *et al*. (2011) reported an average OC/EC ratio of 6.2 (range: 4.0-8.1) at Ahmedabad for the samples collected during Mar-June 2007. Furthermore, chemical composition suggest the dominance of anthropogenic carbonaceous (~58% of $PM_{2.5}$ mass) and inorganic aerosols (~29% of $PM_{2.5}$ mass) during winter months (Rengarajan *et al*., 2011). In contrast, chemical composition at Ahmedabad is mainly dictated by mineral aerosols during summer months under the influence of long-range transport and its close proximity of the Thar Desert. It is estimated that mineral aerosol accounts for ~60-70% of the total aerosol mass throughout the year (Rastogi and Sarin, 2006).

High-altitude Sites

The mass concentrations of OC and EC varied from 0.9 to 12.3 μg m^{-3} and 0.06 to 2.3 μg m^{-3}, respectively at Mt Abu for the samples collected during May 2005-Feb 2006.

Annual average OC and EC concentrations (3.7±0.5 and 0.5±0.5 μg m^{-3}, respectively) account for ~10 and 2% of total aerosols mass at Mt Abu (Ram *et al*., 2008). The measurement at another high-altitude site, Manora Peak (in the central Himalaya) also exhibit significant seasonal variability in OC and EC concentrations. Recently, Ram *et al*. (2010a) reported that OC and EC ranged from 0.4 to 22.3 μg m^{-3} (Av.: 8.2±5.2 μg m^{-3}) and 0.14 to 7.6 μg m^{-3} (Av.: 1.3±1.2 μg m^{-3}), respectively at Manora Peak for the years 2005-2008. The average OC and EC concentrations were 4.8±1.1 and 0.9±0.3 μg m^{-3}, respectively for the samples collected during Dec 2004 during ISRO-GBP LC-II campaign (Rengarajan *et al*., 2007). The mass concentrations of carbonaceous species are relatively higher at Manora Peak than those at Mt Abu. The higher concentrations at Manora Peak are attributed to the advective transport of aerosols from the source regions in northern India (Ram *et al*., 2010a).

The Oceanic Regions (Bay of Bengal and Arabian Sea)

The oceans provide a pristine environment wherein sea-salt aerosols contribute significantly to the total aerosol mass However, recent studies suggest that Arabian Sea and Bay of Bengal as well as other oceanic regions are no longer represent a pristine environment, (Kumar *et al*., 2010; Kumar *et al*., 2008a; Kumar *et al*., 2008b; Nair *et al*., 2010). For examples, the INDOEX (Indian Ocean Experiment) study has reported the presence of high level of pollutants over the Indian Ocean, enriched in BC (mixed with sulphate and organics) and transport of aerosols from south and South-east Asia during the wintertime (Jan-March 1999) (Lelieveld *et al*., 2001). Several studies also documented the influence of transport of aerosols (natural as well as anthropogenic) from continents over the AS and BOB regions (Badarinath *et al*., 2009; Kumar *et al*., 2008a; Nair *et al*., 2010). More recently, Ramanathan et al. (2007) have observed a vertically extended atmospheric brown clouds (ABC), between 0.5 to 3 km, over the Indian Ocean. Using a general circulation model, it is suggested that warming trends in the lower atmosphere, due to the ABC, could be equivalent to the recent increase in the greenhouse gases (Ramanathan *et al*., 2007). Mayol-Bracero et al. (2002) reported that average mass concentrations of OC and EC were 3.4±2.0 and 2.5±1.4 ìg m^{-3}, respectively over the northern Indian Ocean during the INDOEX study. In a recent observations made over the BOB during 19 March to 11 April 2006 as a part of the Integrated campaign for Aerosols, gases and Radiation Budget (ICARB-2006); Kumar *et al*. (2008b) reported that average mass concentrations of OC and EC were 1.9±1.6 and 0.4±0.2 ìg m^{-3}, respectively. The mass concentrations of OC and EC over BOB (range: 2.0 to 8.0 and 0.4 to 3.4 ìg m^{-3}, respectively) were 2-3 times higher during Feb 2003 campaign compared to ICRAB campaign (Sudheer and Sarin, 2008). However, concentrations of OC and EC were much lower (0.20±0.15 and 0.08±0.03 μg m^{-3}, respectively) over the Arabian Sea (AS) for measurement made during April-May 2006 (Sudheer and Sarin, 2008).

WSOC/OC Ratios as Indicator of Secondary Organic Aerosols

The WSOC/OC ratios are a unique tracer to understand secondary organic aerosols (SOA) formation mechanism (Miyazaki *et al*., 2009; Miyazaki *et al*., 2007; Ram and Sarin, 2011). It has been postulated that relatively high WSOC/OC ratios during summer months indicate contribution from SOA due to increased photochemical activity and/or aging of aerosols during the long-range atmospheric transport. The high WSOC/OC ratio is generally attributed to the atmospheric oxidation of volatile organic compounds (VOCs) via reaction of strong oxidants such as ozone and peroxide radicals through the gas-phase conversion (i.e. through SOA formation). However, studies on the measurement of WSOC concentration in ambient aerosol over the Indian region is rather sparse (Khare *et al*., 2011; Ram *et al*., 2012a; Ram and Sarin, 2010, 2011; Ram *et al*., 2010a; Ram *et al*., 2010b, c; Rengarajan *et al*., 2007; Rengarajan *et al*., 2011).

The WSOC/OC ratios are fairly uniform (~0.35-0.40) in aerosols over urban and rural sites in the IGP (Ram and Sarin, 2010). However, WSOC/OC ratios at high-altitude site (~0.55-0.65) and those in the IGP during summer months (~0.45) are higher compared to that during wintertime suggesting a significant contribution from the secondary organic aerosol (Fig. 3) (Ram *et al*., 2010a; Ram *et al*., 2010b). The SOA formation is further corroborated by the elevated WSOC/OC ratios in the daytime samples (0.66 ± 0.11) compared to that in the nighttime samples (0.47 ± 0.07) at Kanpur (Ram and Sarin, 2011). However, secondary inorganic aerosols (NO_3^-, SO_4^{2-} and NH_4^+) were prominent during wintertime (Dec-Feb), under the prevailing meteorological conditions (low ambient temperature and high relative humidity) (Behera and Sharma, 2010a, b; Ram *et al*., 2012a; Ram and Sarin, 2011; Ram *et al*., 2010b; Rengarajan *et al*., 2007). The enhanced contribution from carbonaceous and inorganic species and their hygroscopic growth could be a possible cause for the fog and haze weather conditions during wintertime over northern India (Ram *et al*., 2012b).

Aerosol Optical Properties

The absorption and scattering coefficients (b_{abs} and b_{scat}, respectively) of aerosols are the key parameters to assess direct aerosol radiative forcing and their climatic impact on a regional to global scale (Menon *et al*., 2002; Pant *et al*., 2006; Satheesh and Moorthy, 2005; Satheesh and Ramanathan, 2000; Tripathi *et al*., 2005a; Venkataraman

et al., 2005). However, the assessment of radiative forcing is associated with large uncertainty arising due to the absence of reliable and accurate measurements of BC mass concentration and the optical parameters. Generally, the Sunset Lab's thermo-optical EC-OC analyzer has been used for the measurements of OC and EC mass concentration in atmospheric aerosols (Birch and Cary, 1996a, b; Ram *et al*., 2008; Rengarajan *et al*., 2007). Recently, a novel approach for the simultaneous determination of aerosol absorption coefficient (b_{abs}, Mm^{-1}) and mass absorption efficiency (MAE, σ_{abs}, m^2g^{-1}) of EC has been proposed (Ram *et al*., 2012a; Ram and Sarin, 2009), wherein simultaneously measured optical-attenuation (ATN, equivalent to initial transmittance) of 678 nm laser source, using thermo-optical EC-OC analyzer, has been used for the determination of b_{abs} and σ_{abs} in aerosol samples. This methodology provides reliable, off-line and/or simultaneous measurements of aerosol absorption parameters along with OC and EC mass concentrations. A large spatiotemporal variability in absorption coefficient is observed over the urban sites in IGP and Manora Peak in central Himalaya.

Generally, lower absorption coefficient values are typical of the high-altitude sites and higher values were obtained for the urban atmosphere. The average absorption coefficient at Manora Peak is 13.7 ± 7.3 Mm^{-1} (1 Mm^{-1}=10^{-6} m^{-1}) for the samples collected during Feb 2005-July 2008 (Ram *et al*., 2010a). Among urban sites, b_{abs} at Hisar (39.9 ± 9.1 Mm^{-1}) is lower compared to Allahabad (66.1 ± 17.2 Mm^{-1}) during wintertime (Dec 2004) and the highest was observed for rural sampling site, Jaduguda (69.7 ± 19.6 Mm^{-1}) (Ram and Sarin, 2009). In contrast, MAE of EC at Jaduguda is lowest (6.1 ± 2.0 m^2g^{-1}). The MAE values at the high-altitude site is 12.3 ± 2.9 m^2g^{-1} (Manora Peak) and is similar to those obtained at urban sites in the IGP (11.1 ± 2.6 m^2g^{-1} at Allahabad; 11.3 ± 2.2 m^2g^{-1} at Hisar and 11.3 ± 2.2 m^2g^{-1} at Kanpur). This is unlike the constant conversion factor used for MAE in most of the optical instruments for the determination of BC mass concentration. Thus, these results have significant relevance for aerosol radiative forcing estimations and climate studies over south Asian region where measurements of aerosol optical properties are still lacking in the literature.

The monthly mean AOD values ranged from 0.6-1.2 (at 550 nm) over the IGP and were persistent throughout the year (Jethva *et al*., 2005; Ram *et al*., 2012a) whereas those varied from 0.03 to 0.47 at Manora Peak (Ram *et al*., 2010a). Thus, AOD values in IGP are an order of magnitude higher compared to those in Himalayas. Generally, AOD values exhibit a similar trend in the IGP and central Himalaya with peaking in summer months (April-June), and relatively high AOD values are attributed to the transport of mineral dust originating from desert regions in middle-East and Thar Desert (in western India) (Ram *et al*., 2012a; Ram *et al*., 2010a). In addition, AOD values at Kanpur shows another maxima during post-monsoon to winter months (Oct-Jan) (Dey and Tripathi, 2008). Based on satellite retrievals, it was suggested that summertime high AOD values were associated with relatively low fine-mode aerosol fraction (FMAF) values (<0.4) and thus, suggesting the dominance of coarse-mode particles (Dey and Tripathi, 2008; Jethva *et al*., 2005). However, fine-mode aerosols (derived from anthropogenic emissions) dominate PM_{10} mass and AOD values during wintertime at Kanpur (Jethva *et al*., 2005; Ram *et al*., 2012a; Ram and Sarin, 2011; Tare *et al*., 2006).

Conclusions

Aerosols are an important component of the atmosphere and their sources (natural and anthropogenic) play a vital role in modifying the composition and, hence, the Earth's radiative balance. The divergence of absorbing and scattering characteristics of carbonaceous aerosols assumes particular importance in the polluted regions of the Indo-Gangetic Plain. During the winter season prevalence of agricultural-waste burning, fossil fuel combustion and wood-fuel for domestic heating result in enormous amount of organic carbon (OC) and elemental carbon (EC) that modify the total particulate carbon content of the atmosphere. Therefore, optical properties of aerosols assessed by the scattering coefficient and widely used for the determination of single scattering albedo (SSA – defined as the ratio of scattering coefficient to the sum of scattering and absorption coefficients) need re-evaluation for the radiative impact assessment due to aerosols. The highly acidic environment due to the presence of sulphate and nitrate aerosols over Northern India may significantly alter the morphological features of soot particles. Although the aerosol absorption properties have been mainly attributed to the occurrence of EC, the absorption from humic-like substances (HULS), formed during biomass burning emissions, needs to be investigated in the Indo-Gangetic Plain. This is important in order to quantify the total aerosol absorption and the site-specific mass absorption efficiency. The chemical reactions that occur on aerosol surfaces are also important in determining whether an aerosol particle can act as cloud condensation nuclei (CCN). The information is essential in order to infer mixing state of aerosols and their CCN activity.

Acknowledgements

The financial support received from ISRO-GBP office (Bengaluru, India) during the course of this study is thankfully acknowledged. The authors are thankful to Drs S. N. Tripathi and P. Hegde for extending logistic support for long-term sampling from Kanpur and Manora Peak, respectively.

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