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Atmospheric carbonaceous aerosols from Indo-Gangetic Plain and Central Himalaya: Impact of anthropogenic sources

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ABSTRACT

In the present-day scenario of growing anthropogenic activities, carbonaceous aerosols contribute significantly (~20–70%) to the total atmospheric particulate matter mass and, thus, have immense potential to influence the Earth's radiation budget and climate on a regional to global scale. In addition, formation of secondary organic aerosols is being increasingly recognized as an important process in contributing to the air-pollution and poor visibility over urban regions. It is, thus, essential to study atmospheric concentrations of carbonaceous species (EC, OC and WSOC), their mixing state and absorption properties on a regional scale. This paper presents the comprehensive data on emission sources, chemical characteristics and optical properties of carbonaceous aerosols from selected urban sites in the Indo-Gangetic Plain (IGP) and from a high-altitude location in the central Himalaya. The mass concentrations of OC, EC and WSOC exhibit large spatio-temporal variability in the IGP. This is attributed to seasonally varying emissions from post-harvest agricultural-waste burning, their source strength, boundary layer dynamics and secondary aerosol formation. The high concentrations of OC and SO_4^{2-}, and their characteristic high mass scattering efficiency, contribute significantly to the aerosol optical depth and scattering coefficient. This has implications to the assessment of single scattering albedo and aerosol radiative forcing on a regional scale.

1. Introduction

Atmospheric carbonaceous aerosols over south and south-east Asia have been a subject of major debate over the past two decades because of their potential impact on the regional air quality, climate and hydrological cycle (Bollasina et al., 2011; Menon et al., 2002; Ramanathan and Carmichael, 2008; Ramanathan et al., 2007). Among the major anthropogenic sources, biomass burning (agricultural-waste and wood-fuel), industrial and vehicular emissions have contributed significantly to the total aerosol burden of the atmosphere and in particular to the carbonaceous species over northern India (Babu and Moorthy, 2002; Ram and Sarin, 2011; Ram et al., 2010a,b; Rengarajan et al., 2007; Tripathi et al., 2005; Venkataraman et al., 2005).

Emission products of biomass burning and fossil-fuel combustion are characterized by different type of chemical species, namely organic carbon (OC) and elemental carbon (EC, also known as black carbon (BC) or soot) (Andreae and Gelencser, 2006). EC and BC are interchangeably used in literature; however, we will use EC in this paper when measured by an EC–OC analyzer and BC when it is measured by an Aethalometer. Although OC and EC are emitted from same sources, they have different physical, chemical and optical properties (Andreae and Merlet, 2001; Ram and Sarin, 2009). Black carbon is a major absorbing species in the ambient atmosphere, ranking next to carbon dioxide (CO_2) and methane (CH_4) in contributing to the atmospheric radiative forcing (IPCC, 2007). The warming potential of EC can influence the atmospheric circulation pattern and the cloud-precipitation efficiency over south and south-east Asia (Bollasina et al., 2011; Menon et al., 2002; Padma Kumari and Goswami, 2010; Wang et al., 2009). In contrast, OC represents carbon fraction associated with non-refractory organics (low-molecular weight hydrocarbons and its derivatives) and mainly scatters the solar radiation. However, recent studies suggest that a small component of OC (such as humic-like substances, HULIS and Brown Carbon), emitted during biomass burning emissions can absorb solar radiation at lower wavelength of the spectrum (Andreae and Gelencser, 2006; Kirchstetter et al., 2004).

A large spatio-temporal variability in aerosol chemical composition, optical properties and radiative forcing has been reported

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over Indian region. The varying nature and strength of emission sources (anthropogenic and natural), transport pathways of chemical constituents and boundary layer dynamics are the dominant factors that contribute to the pronounced seasonal variability (Aloysius et al., 2008; Das et al., 2009, 2008; Ganguly et al., 2006; Nair et al., 2007; Niranjan et al., 2006; Pant et al., 2006; Ram and Sarin, 2009, 2010; Ramachandran et al., 2006; Rengarajan et al., 2007; Srivastava et al., 2009, 2012; Tare et al., 2006; Tripathi et al., 2006). However, a long-term systematic data on atmospheric concentrations of EC, OC and water-soluble organic carbon (WSOC) from Northern India is still lacking in the literature. In addition, relatively high concentrations of OC and SO_4^{2-} contribute significantly to the total aerosol mass over the urban atmosphere of northern India during the wintertime (Kulshrestha et al., 2009; Ram and Sarin, 2011; Ram et al., 2010b; Rengarajan et al., 2007, 2011; Tare et al., 2006). Furthermore, relatively high mass scattering efficiency of OC and SO_4^{2-} (3.8 ± 0.5 and 6.1 ± 2.1 m^2 g^{-1}, respectively) (Magi, 2009, 2011) would lead to a significant increase in total scattering coefficient. Thus, direct aerosol radiative forcing depends largely on the relative fractions of scattering-OC and -SO_4^{2-} compared to absorbing-EC (i.e. OC/EC and SO_4^{2-}/EC ratios) in the ambient aerosols (Novakov et al., 2005; Ramana et al., 2010).

This manuscript presents a review and synthesis of the abundance pattern of carbonaceous species (OC, EC and WSOC) from the central Himalaya and selected urban sites in the Indo-Gangetic Plain (Ram and Sarin, 2010, 2011; Ram et al., 2008, 2010a,b; Rengarajan et al., 2007; Satsangi et al., 2010). The primary objective is to assess the spatio-temporal variability in the abundance pattern and emission sources of carbonaceous species over Northern India. In addition, we discuss their climatic relevance based on OC/EC and SO_4^{2-}/EC ratios from urban sites in the Indo-Gangetic Plain and high-altitude site in Central Himalaya.

2. Meteorology and emission sources in the Indo-Gangetic Plain

The regional topography, meteorology and emission sources (natural and anthropogenic) essentially dictate the spatio-temporal variability in mass concentrations of ambient aerosols and their chemical composition. The local rainfall is also important for the efficient wash-out of the atmosphere (in-cloud and/or below-cloud scavenging). Wet-precipitation in the Indo-Gangetic Plain (IGP) occurs mainly during the south-west (SW)-monsoon (late June–September) and accounts for ~70% of the annual rainfall over India. Based on the regional meteorology, we have assessed the temporal variability in concentrations of carbonaceous species spread over four seasons, described here as winter (December–March), summer/pre-monsoon (April–June), monsoon (late June–September) and post-monsoon (October–November). During the period of SW-monsoon (July–September), the ambient atmosphere is relatively clean; whereas, in the wintertime, the shallow boundary layer height, 500–800 m (Nair et al., 2007) confines aerosols within the lower atmosphere.

The atmospheric abundance of aerosols is intimately related to the wind speed, whereas an increase in relative humidity (RH) favors coagulation/condensation of water-soluble aerosols. At a critical value of RH, particles tend to grow into bigger droplets and are subsequently removed from the ambient atmosphere. The relative humidity in the IGP attains close to 100% (Av ~ 70%) when winds are generally weak (<2 m s^{-1}) and mostly localized during the wintertime and post-monsoon (Ram et al., 2010b). The stagnant atmosphere, relatively high concentrations of anthropogenic aerosols and their hygroscopic growth may lead to formation of fog-haze conditions during wintertime over the IGP (Gautam et al., 2007; Ram et al., 2012a; Ramachandran et al., 2006; Ramanathan and Ramana, 2005; Tare et al., 2006). In general, fog-haze events succeed after intense biomass burning emissions in Northern India (as seen by the fire counts and MODIS image) (Badarinath et al., 2009; Ram and Sarin, 2011) and are characterized by high abundance of carbonaceous and inorganic aerosols (Ram and Sarin, 2011).

During summer months, RH is generally low (~25–60%) with ambient temperature close to ~40–45 °C. Winds are relatively strong and are northwesterly with occasional component from the northeast and southwest. The surface level low pressure, changes in the wind regimes and increase in the ambient temperature favor convective mixing and long-range transport of aerosols; thus, leading to a rapid decrease in the concentrations of carbonaceous species in the lower atmosphere during summer months (Mishra and Tripathi, 2008; Singh et al., 2005).

Biomass burning (wood-fuel and agricultural waste burning), vehicular and industrial emissions are among the major sources of carbonaceous aerosols over Northern India. Emission from bio-fuel consumption is estimated to be 247–584 Tg yr^{-1} (1 Tg = 10^{12} g) and accounts for ~75% of the total emission in India (Habib et al., 2004). The agricultural crop-residue burning contributes remaining 25% of the emissions. Black carbon emissions from these sources range from 65 to 760 Gg yr^{-1} (Habib et al., 2004). However, these estimates are based on emission inventories and type of fuels; thus resulting in large degree of uncertainty. The coal-based emissions are highly localized in eastern part of the IGP (Prasad et al., 2006; Reddy and Venkataraman, 2002). Thus, emissions from coal-based thermal power plants and brick-kilns represent an important source of carbonaceous aerosols over Northern India.

Rice and wheat, the two major cereal crops in Northern India, are seasonally grown in the land area covering nearly 12 million hectares (Mha) (Singh et al., 2011). These crops are cultivated in the monsoon season (June–September) and during January to early February, respectively. The cultivation area used for rice and wheat production in Punjab state alone accounts for ~20% (2.6 Mha) of the total agricultural area of the IGP. The two crops produce 100 Million tones of the rice straw, with almost 75% of the straw is disposed by the open burning in the field. The wheat residue burning (during April–May) constitutes only 8% of total biomass burning emission and rest (~92%) is from the rice-straw burning in Punjab. The emission factors (EFs) for various chemical and particulate species have been estimated as 34.66 g/kg (for CO); 2.63 g/kg (for NOx); 0.41 g/kg (CH_4); 3.99 g/kg (for PM_{10}; aerosols with aerodynamic diameter ≤ 10 μm) and 3.76 g/kg (for $PM_{2.5}$; aerosols with aerodynamic diameter ≤ 2.5 μm) from wheat residue burning (Badarinath et al., 2006). Based on these emission factors, Badarinath et al. (2006) had estimated that total emissions from Punjab State contribute ~13 Gg of PM_{10} (or equivalent to 12 Gg of $PM_{2.5}$). Likewise, emissions from paddy residue burning are estimated to be 30 Gg of PM_{10} (or equivalent to 28.3 Gg of $PM_{2.5}$) (Badarinath et al., 2006). A recent study by Rajput et al. (2011) has suggested that emissions of elemental and organic carbon (EC and OC) are factor of 4–5 lower during wheat-straw burning compared to rice-straw burning. Relatively high abundance of particulate species (EC, OC and $PM_{2.5}$) emitted from rice-straw burning during October–November is attributed to the high moisture content and poor combustion efficiency.

3. Methodology

3.1. Study sites in the Indo-Gangetic Plain and central Himalaya

The Indo-Gangetic Plain (hereafter referred as IGP; Fig. 1), extending from (21.75 °N, 74.25 °E) to (31.0 °N, 91.5 °E), is