



## Carbonaceous and inorganic species in atmospheric aerosols during wintertime over urban and high-altitude sites in North India

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[1] Synchronous sampling of bulk-aerosols, carried out during wintertime from the two strategically located sites in North India, reveals that total suspended particulates (TSP) over an urban site (Hisar: 29.2°N 75.7°E; 219 m asl) ranged from 67 to 396  $\mu\text{g m}^{-3}$ ; in contrast, TSP at Manora Peak (a high-altitude station: 29.4°N 79.5°E; 1950 m asl) was relatively low (range: 13.7 to 42.7  $\mu\text{g m}^{-3}$ ). At Hisar, on average, water-soluble ionic species (WSIS, range: 14.1 to 78.3  $\mu\text{g m}^{-3}$ ) contribute nearly one-fourth by weight to TSP, with dominant contribution from  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{NH}_4^+$ . The time series analysis over a span of 30 days shows somewhat uniform distribution of organic carbon/elemental carbon (OC/EC) ratio centering around  $8.5 \pm 2.2$  at this urban site; and the water-soluble organic components (WSOC range: 6.7 to 42.0  $\mu\text{g m}^{-3}$ ) account for 11.5 % to the TSP concentration. Both WSOC and OC exhibit significant positive correlation with water-soluble  $\text{K}^+$  ( $r = 0.88$  and  $0.79$  respectively), suggesting their dominant contribution from biomass burning. At Manora Peak, the chemical composition of ambient aerosols show characteristically lower abundances of WSIS (range: 2.0 to 9.9  $\mu\text{g m}^{-3}$ ) and WSOC (range: 1.4 to 6.0  $\mu\text{g m}^{-3}$ ); together they account for one-third of the TSP. The characteristic low abundances of OC (range: 2.8 to 6.9  $\mu\text{g C m}^{-3}$ ) and EC (range: 0.34 to 1.4  $\mu\text{g C m}^{-3}$ ) at this high-altitude site and their significant correlation with  $\text{K}^+$  and  $\text{SO}_4^{2-}$  suggest contribution from long-range transport of anthropogenic species. This study represents a first comprehensive data set for documenting the chemical characteristics of ambient aerosols and source apportionment of EC, OC, WSIS and mineral dust over urban and high-altitude sites in north India, an important data set required for the south Asian region. If the observed OC/EC ratios far greater than  $\sim 2$  (unlike reported values in the literature for urban sites) and the semi-empirical estimates of secondary OC are typical of the annual average abundances in the ambient aerosols over north India, then the temporal and regional analyses of primary and secondary OC using the existing emission models require reassessment for this region.

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### 1. Introduction

[2] In the present-day scenario of growing anthropogenic activities, a comprehensive study on the chemical characterization of atmospheric aerosols is essential to assess their impact on regional environment degradation, human health and climate change. [Dockery *et al.*, 1993; Harrison and Yin, 2000; IPCC, 2007]. The principal contribution to the atmospheric radiative forcing and climate change involves black carbon (also known as elemental carbon, EC), organic carbon (OC) and sulphate aerosol particles [Charlson and Heintzenberg, 1995; Chung *et al.*, 2005; Jones *et al.*, 2005]. The primary sources of EC are biomass burning and combustion of fossil fuels [Goldberg, 1985]; almost 50%

of its global emissions are estimated to arise from the latter source [Cooke and Wilson, 1996]. The primary sources of OC are either anthropogenic or biogenic in nature; whereas its secondary source results from the condensation of semi-volatile organic vapors to particle surfaces and via atmospheric photo-oxidation reactions of precursor gases such as terpene [Odum *et al.*, 1996; Griffin *et al.*, 1999; Kanakidou *et al.*, 2005]. The role of black carbon and organic aerosols is getting increasingly recognized in the atmospheric chemistry and radiative forcing models [IPCC, 2007]. The organic aerosols have potential to scatter sun's radiation and to reduce the hygroscopicity of inorganic species, thus causing variations in the light scattering property of aerosols with change in relative humidity [Sjogren *et al.*, 2007]. In particular, the water-soluble organic compounds (WSOC) can alter the surface properties of aerosols from hydrophobic to hydrophilic and thus influence their optical properties as well as atmospheric cycles of several important trace gases (such as ozone); and eventually the number

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