



Chemical composition and source-apportionment of sub-micron particles during wintertime over Northern India: New insights on influence of fog-processing[☆]



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ABSTRACT

A comprehensive study was carried out from central part of Indo-Gangetic Plain (IGP; at Kanpur) to understand abundance, temporal variability, processes (secondary formation and fog-processing) and source-apportionment of PM₁-bound species (PM₁: particulate matter of aerodynamic diameter $\leq 1.0 \mu\text{m}$) during wintertime. A total of 50 PM₁ samples were collected of which 33 samples represent submicron aerosol characteristics under non-foggy condition whereas 17 samples represent characteristics under thick foggy condition. PM₁ mass concentration during non-foggy episodes varied from 24–393 (Avg.: 247) $\mu\text{g m}^{-3}$, whereas during foggy condition it ranged from 42–243 (Avg.: 107) $\mu\text{g m}^{-3}$. With respect to non-foggy condition, the foggy conditions were associated with higher contribution of PM₁-bound organic matter (OM, by 23%). However, lower fractional contribution of SO_4^{2-} , NO_3^- and NH_4^+ during foggy conditions is attributable to wet-scavenging owing to their high affinity to water. Significant influence of fog-processing on organic aerosols composition is also reflected by co-enhancement in OC/EC and WSOC/OC ratio during foggy condition. A reduction by 5% in mineral dust fraction under foggy condition is associated with a parallel decrease in PM₁ mass concentration. However, mass fraction of elemental carbon (EC) looks quite similar ($\approx 3\%$ of PM₁) but the mass absorption efficiency (MAE) of EC is higher by 30% during foggy episodes. Thus, it is evident from this study that fog-processing leads to quite significant enhancement in OM (23%) contribution (and MAE of EC) with nearly equal and parallel decrease in SO_4^{2-} , NO_3^- and NH_4^+ and mineral dust fractions (totaling to 24%). Characteristic features of mineral dust remain similar under foggy and non-foggy conditions; inferred from similar ratios of Fe/Al (≈ 0.3), Ca/Al (0.35) and Mg/Al (0.22). Positive matrix factorization (PMF) resolves seven sources: biomass burning (19.4%), coal combustion (1.1%), vehicular emission (3%), industrial activities (6.1%), leather tanneries (4%), secondary transformations (46.2%) and mineral dust (20.2%).

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1. Introduction

Carbonaceous aerosols constitute a substantial fraction of atmospheric fine particulate matter (PM) (Fu et al., 2010; Kanakidou et al., 2005; Kaul et al., 2011; Rajput et al., 2014c, 2017; Surratt et al., 2007; Weber et al., 2007). Based on the physico-chemical and optical properties, carbonaceous species have been classified into

organic carbon (OC) and elemental carbon (EC) (Kaul et al., 2012). They (OC, EC) play a key role in influencing the radiation budget of the earth; by scattering and absorbing the solar radiation (direct effect) or by acting as cloud-condensation nuclei (CCN), when scavenged into cloud droplets (indirect effect). It is important to mention here that secondary aerosol species viz. sulfate and nitrate also act as efficient CCN. Furthermore, ambient records (from different geographical locations) on bioaerosols with high diversity would be very important to understand and model their role as efficient CCN and IN (ice-nuclei) (Fröhlich-Nowoisky et al., 2009, 2016; Rajput et al., 2017). During wintertime, Indo-Gangetic Plain (IGP) experiences prolonged haze and fog-episodes; a

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manifestation of western disturbance, shallower planetary boundary layer height and emissions from biomass burning and fossil-fuel combustion. Continental outflow of these air-pollutants towards the foot-hills of north-eastern Himalaya and over the Bay of Bengal (BoB) is a conspicuous feature during wintertime (Rajput et al., 2013; Ramanathan et al., 2001). Dissolution of water-soluble organic and inorganic species and scavenging of insoluble species (like EC and metals) into the fog can alter overall chemical, morphological and optical properties of aerosols in particular of carbonaceous species and inorganic constituents (Gilardoni et al., 2014). Thus, assessing the influence of fog-processing is of utmost importance to understand its impact on the regional-scale atmospheric chemistry and radiative forcing (Collett et al., 1999; Hallberg et al., 1992).

IGP, located in the northern part of India, bounded by the Himalaya in its north and the Aravallis/Deccan Traps in its south, is one of the most polluted regions of India, both during the winter and the summer seasons (Chakraborty and Gupta, 2010; Jethva et al., 2005; Rajput et al., 2017, 2016c, 2014c; Singh and Gupta, 2016a). Due to its unique topography the aerosols get confined in the plain making the aerosol loading higher relative to the other parts of India (Chakraborty et al., 2017; Kumar et al., 2017). Previous studies from IGP have assessed chemical characterization and radiative forcing of ambient aerosols with not much emphasis on fog-processing of refractory and non-refractory aerosol species (Dey and Tripathi, 2007; Jethva et al., 2005; Rajput et al., 2011; Ram et al., 2010; Ramachandran et al., 2006; Satsangi et al., 2012; Srivastava and Ramachandran, 2013; Tripathi et al., 2005). In this study, we document a comprehensive scenario of fog-processing on chemical composition of PM₁ and absorption properties of EC from central part of IGP at Kanpur location. In addendum to this, a more robust source-apportionment (utilizing positive matrix factorization: PMF 5.0) of these submicron aerosols has been performed for the first-time by incorporating a lot of parameters (mineral aerosols, carbonaceous species and water-soluble inorganic species: WSIS). To better appreciate the influence of fog-processing, herein we have discussed chemical data set and several characteristic ratios in two categories: non-foggy (n = 33) and foggy conditions (n = 17). However, PMF based source-apportionment has been done with data set (matrix: 41 samples x 27 parameters) showing influence of north-westerly (data set associated with far-off air-masses have not been considered for PMF analysis; Fig. S1). In a previous study (Rajput et al., 2016a), we have addressed the issue that carbonaceous aerosols data in conjunction with other constituents would be essential for better insight into source contribution.

2. Materials and method

2.1. Site description and aerosol sampling

IGP (Indo-Gangetic Plain) stretches from north-western to north-eastern region in India (Rajput et al., 2013, 2014a). Shallower boundary layer height and massive prevailing emissions during wintertime leads to haze and thick fog episodes over entire IGP. The sampling site (Kanpur; 26.30 °N; 80.14 °E; 142 m amsl.) is situated in central IGP. IGP is a densely populated and highly polluted region in the country. Major anthropogenic sources include vehicular emissions, biomass burning and industrial activities. As far as the quantification of these emission sources is concerned, there is a great challenge owing to plausible reasons like year-to-year varying contribution of biomass burning emission, secondary aerosol formation etc. (Chakraborty et al., 2015; Rai et al., 2016; Rajput et al., 2016a). It is important to mention here that for the first time, in this study, we have made a rigorous attempt to characterize all

components (major) i.e. mineral aerosols, carbonaceous aerosols (OC, EC) and water-soluble inorganic species (WSIS) of PM₁ (particulate matter of aerodynamic diameter ≤ 1.0 μm) during wintertime from IGP (at Kanpur).

Low-volume (calibrated flow rate: 10 LPM) aerosol samplers (indigenously designed and developed by our group at Indian Institute of Technology Kanpur: IITK) with PM₁ size cut-off (Gupta et al., 2010; Kumar and Gupta, 2015) were deployed on the terrace of CESE (Centre for Environmental Science and Engineering) building in the campus of IITK. These PM₁ samplers have been extensively utilized in many previous studies e.g. (Rai et al., 2016; Rajput and Gupta, 2016; Rajput et al., 2015, 2016a; Singh and Gupta, 2015, 2016a, 2016b). The aerosol samples were collected onto pre-combusted (450 °C) quartz-fiber filters (47 mm diameter) for 8 h (Indian Standard Time: 2300 h–0700 h). A total of 50 samples were collected over a period of four months (drier season) from November 2014 to February 2015. Out of these, 33 samples are representative of non-foggy nights, whereas remaining 17 samples represent aerosol collection during thick foggy episodes. Soon after their retrieval from the sampler, filters were stored at -19 °C until analysis.

2.2. Gravimetric determination of PM₁ and EC-OC analysis

Following standard analytical protocol (Rajeev et al., 2016; Rajput et al., 2016a, 2017; Rajput and Gupta, 2016), the mass concentrations of PM₁, elemental carbon (EC), organic carbon (OC), water-soluble organic carbon (WSOC), water-soluble inorganic species (WSIS: NH₄⁺, Na⁺, K⁺, Mg²⁺, Ca²⁺, Cl⁻, NO₃⁻ and SO₄²⁻) and metals (Fe, Al, Ca and Mg) have been measured in this study. In brief, the PM₁ mass was measured gravimetrically on a high-precision micro-balance post to equilibration of filters at 37 ± 2% RH (relative humidity) and temperature of 24 ± 2 °C (for nearly 12 h). Mass concentrations of OC and EC have been determined on a carbon analyzer (Sunset Laboratory) using NIOSH protocol (National Institute for Occupational Safety and Health) (Birch and Cary, 1996; Rajput et al., 2017). The analytical uncertainty of 2% on the determination of total carbon (TC = OC + EC) was assessed by analyzing potassium hydrogen phthalate (KHP; n = 10). To allocate carbonate carbon (CC) peak and deduce its value, several aerosol samples (n = 7) were de-carbonated in hydrochloric acid (HCl) (Cachier et al., 1989; Rajput et al., 2017). Data suggests that CC always contributes less than 4% of the total carbon (OC + EC + CC) in PM₁. The area under CC peak was used to correct for OC concentration in aerosols as reported in earlier publications (Kaul et al., 2011; Rajput et al., 2017). Filter-based blank concentration (n = 10) of OC has been deduced from the measured concentration in aerosol samples. The detection limit of OC and EC is 0.24 and 0.06 ng m⁻³ (Table 1).

2.3. Determination of water-soluble organic carbon (WSOC) and inorganic species (WSIS)

For determination of water-soluble organic carbon (WSOC) and WSIS, 1–2 punches (3.14 cm²) of each sampled filter (pre-soaked overnight) was extracted (sonicated) with 30–40 mL of Milli-Q water (resistivity: 18.2 MΩ cm). The resulting water extracts were filtered (through 0.22 μm, Millipore) into amber colored glass vials, and analyzed for WSOC on a total organic carbon analyzer (TOC Shimadzu model 5000A). The linear calibrations for TC and IC were achieved (R² > 0.99) on TOC analyzer with potassium hydrogen phthalate (KHP) and a sodium carbonate-bicarbonate mixture (Na₂CO₃+NaHCO₃), respectively. Replicate injections (3–5 times) of water extracts revealed reproducibility within ±3%. The water-soluble inorganic species (WSIS: NH₄⁺, Na⁺, K⁺, Mg²⁺, Ca²⁺, Cl⁻,

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