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Impacts of the high loadings of primary and secondary aerosols on light extinction at Delhi during wintertime

S. Tiwari [a], A.K. Srivastava [a,*], D.M. Chate [b], P.D. Safai [b], D.S. Bisht [a], M.K. Srivastava [c], G. Beig [b]

[a] Indian Institute of Tropical Meteorology (Branch), Prof Ramnath Vij Marg, New Delhi, India
[b] Indian Institute of Tropical Meteorology, Dr Homi Bhabha Road, Pashan, Pune, India
[c] Department of Geophysics, Banaras Hindu University, Varanasi, India

HIGHLIGHTS

- First study on contribution of different species to extinction coefficient.
- Scattering type aerosols were dominated by ~76% than absorbing type aerosols.
- The largest contribution was observed for organic carbon (~46%).
- Lowest contribution was observed for ammonium nitrate (~4%).

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ABSTRACT

High emissions of anthropogenic aerosols over Indo-Gangetic Plain (IGP) inspired continuous measurements of fine particles ($PM_{2.5}$), carbonaceous aerosols (BC, OC and EC), oxides of nitrogen (NO_x) and estimation of light extinction (b_{ext}) and absorption (b_{abs}) coefficients over Delhi during high pollution season in winter from December 2011 to March 2012. During study period, the mass concentrations of $PM_{2.5}$, BC and NO_x were 186.5 ± 149.7 μg m^{-3}, 9.6 ± 8.5 μg m^{-3} and 23.8 ± 16.1 ppb, respectively. The mass concentrations of OC and EC were studied by two different techniques (i) off-line (gravimetric method) and (ii) semi-continuous (optical method) and their mean mass concentrations were 51.1 ± 15.2, 10.4 ± 5.5 μg m^{-3} and 33.8 ± 27.7, 8.2 ± 6.2 μg m^{-3}, respectively during the study period. The ratios of mass concentration of OC to EC in both cases were in between 4 and 5. The source contribution of carbonaceous aerosols in $PM_{2.5}$ estimated over 24hrs, during day- and night-time where motor vehicles accounted for ~69%, 90% and 61% whereas coal combustion accounted for ~31%, 10% and 39%, respectively. The estimated mean values of b_{ext} and b_{abs} over the station were 700.0 ± 268.6 and 71.7 ± 54.6 Mm^{-1}, respectively. In day and night analysis, b_{ext} is ~37% higher during night-time (863.4 Mm^{-1}) than in day-time (544.5 Mm^{-1}). Regression analysis between b_{ext} and visibility showed significant negative correlation ($r = -0.85$). The largest contribution in the light extinction coefficients was found to be due to organic carbon (~46%), followed by elemental carbon (~24%), coarse mode particles (~18%), ammonium sulfate (~8%) and ammonium nitrate (~4%). The individual analysis of light extinction due to chemical species and coarse mode particles indicates that scattering type aerosols dominated by ~76% over the absorbing type. The aforementioned results suggest that the policy-induced control measures at local administration level are needed to mitigate the excess emissions of carbonaceous aerosols over IGP region which ranks highest in India and elsewhere in worldwide.

1. Introduction

The Indo-Gangetic plain (IGP) is the most populous and one of the highly polluted regions in the world, surrounded by various anthropogenic sources such as burning of fossil fuels and agricultural residues, vehicular emissions etc. Its unique topography along

* Corresponding author.
E-mail address: atul@tropmet.res.in (A.K. Srivastava).

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with atmospheric stability and calm winds during winter period are responsible for the higher level of pollution over the region during the period (Srivastava et al., 2012a,b; Tiwari et al., 2013a,b; Rehman et al., 2011). Delhi, the fourth most populous megacity with 19 million inhabitants is located in the western IGP region in northern part of India (http://www.indiaonlinepages-com/population/delhi-population.html). Tremendous growth of population, urbanization, industrialization, human migration as well as meteorological conditions over the station increased the atmospheric pollution levels, especially during winter which causes visibility degradation, dense fog, smog and haze every year (Gautam et al., 2007; Singh et al., 2008; Lal et al., 2013).

Carbonaceous aerosols are emitted from the combustion of fossil and biomass fuels and are more abundant component in fine particulate matter (i.e. $PM_{2.5}$). They can be usually classified into three different categories: organic carbon (OC), elemental carbon (EC) and carbonate carbon (CC). In OC, carbon is associated with organic compounds emitted either directly (anthropogenic emissions) from the source, or by condensation due to atmospheric oxidation of volatile organic compounds (i.e. secondary OC) in the atmosphere. On the other hand, EC is formed during the combustion of hydrocarbons and is essentially non-volatile at ambient temperature (Jeong et al., 2004; Hussain et al., 2007; Rogge et al., 1998; Sheesley et al., 2003). Often, EC has inter-changeable relationship with black carbon (BC) (Turpin and Huntzicker, 1991) and are typically light absorbing (Jacobson, 2004, 2006). Mineral dust is one of the good sources for CC, which is not given much consideration due to low atmospheric mass concentration. In the urban areas, diesel automobiles, especially heavy duty trucks are the main source of EC in the atmosphere (Gray and Cass, 1998). In the Indian context, however, very few studies have been conducted for the measurement of total carbonaceous aerosols (Satsangi et al., 2012; Tiwari et al., 2013a; Srivastava et al., 2014). Venkataraman et al. (2005) reported the relative contributions of fossil fuel, open biomass and bio-fuel combustion in BC mass are ~25%, 33% and 42%, respectively whereas ~13%, 43% and 44% in OC mass, respectively. In another study, Stone et al. (2010) have estimated only 4% contribution of OC from diesel combustion whereas large contribution from biomass burning (21%) to OC over the Himalayas.

In the present study, the mass of $PM_{2.5}$ particles and its chemistry, BC, NO_x, extinction coefficient (b_{ext}) along with the meteorological parameters were analyzed over Delhi during winter period (1st December 2011 to 30th March 2012). This is the first attempt to understand the impact of aerosol mass and its chemical composition on the light extinction coefficient over the study region. The main objectives of the present study are: (i) to examine the contribution of mass of atmospheric aerosols, carbonaceous fraction and water-soluble inorganic species in $PM_{2.5}$; (ii) to estimate the contribution of b_{ext} along with absorption and scattering coefficients for their day-time and night-time variations, and (iii) to investigate the relationship between b_{ext} and visibility.

2. Sampling site and instrumentations

The experimental site Delhi is located in the north western part of the IGP with Thar Desert in the west, which is the single largest contributor to the mineral dust aerosols in the region (Srivastava et al., 2011; Todd et al., 2007). During winter, the occurrence of fog, calm atmospheric conditions and low level boundary inversion are experienced over the station. Samples of aerosols and other pollutants were collected in the premises of Indian Institute of Tropical Meteorology (Branch), located in central part of New Delhi (28.38 °N, 77.10 °E and ~235 m amsl), India. The population density of Delhi is ~11,297 km^{-2} in 2011, having approximately 19 million inhabitants spread over 1484 km^2 area. It has a sub-tropical climate with extremely hot temperatures during summer (April–May–June) and moderately cold temperature in winter (December–January–February–March).

Samples of $PM_{2.5}$ were collected by medium volume air sampler (gravimetric: off-line; APM 550, Envirotech Pvt. Ltd., India) once in a week during day-time (1000–1800 h) and night-time (1900–0700 h). Details of the instrumentation and the chemical analysis technique are given elsewhere (Tiwari et al., 2009; Srivastava et al., 2012a). Further, aerosol mass concentrations ($PM_{2.5}$ and PM_{10}) were also measured by online samplers (beta attenuation: optical method) from Thermo Andersen, USA, Inc., Series FH 62 C14 (C14 BETA) during the study period. Thermal-optical transmittance based semi-continuous OC-EC instrument (Sunset Lab, USA: Model-4L) was used for the measurement of organic and elemental carbon (Tiwari et al., 2013a). Black carbon, was measured continuously using an Aethalometer (Model AE-31, Magee Scientific Company, Berkeley, CA, USA) with high temporal resolution (5-min interval) using quartz fiber filter tape transmission at an 880 nm wavelength (Tiwari et al., 2013b). Chemiluminescence NO–NO_2–NO_x analyzer (Model 42*i* Thermo electron Corporation, US) was used for monitoring of NO_x. Meteorological parameters (including visibility) during study period were obtained from India Meteorological Department, New Delhi which were recorded near the sampling site (~500 m) in the campus of Indian Agricultural Research Institute; however, rainfall data was collected inside the premises of the institute. The boundary layer mixing depths for Delhi was obtained from the stability time series data available on the NOAA Air Resources Laboratory web server (http://www.arl.noaa.gov/ready.html). The general equation is adopted for conversion of ppb to μg m^{-3} for NO_x is as below:

$$\mu g\ m^{-3} = (ppb)*(12.187)*(M)/(273.15 +^{\circ} C) \quad (1)$$

Where, M is molecular weight of NO_x. An atmospheric pressure of 1 atm is assumed.

3. Results and discussion

3.1. Status of primary aerosols over Delhi during winter

Day to day variability of mass concentrations of $PM_{2.5}$, BC and NO during study period over Delhi are shown in Fig. 1. The monthly mean concentrations of $PM_{2.5}$, BC and NO measured from different techniques are depicted in Table 1. The seasonal mean mass concentration of $PM_{2.5}$ was found to be 186.5 ± 149.7 μg m^{-3} with on-line sampling and 151.7 ± 30.9 μg m^{-3} with off-line sampling during the study period. The mean mass concentration of on-line $PM_{2.5}$ mass was ~23% higher than that of off-line $PM_{2.5}$, which might be due to (i) difference in sampling protocol for on-line and off-line and (ii) chocking of the pores of the filter media in off-line sampling due to high humidity, especially during foggy nights. Monthly variability analysis shows on-line $PM_{2.5}$ mass fraction is relatively higher in December (108%) and January (14%) as compared to off-line $PM_{2.5}$ mass. On the other hand, during February and March, on-line $PM_{2.5}$ mass fraction is found to be relatively lower by about 36% and 16%, respectively as compared to off-line $PM_{2.5}$ mass. Results are attributed due to the influence of meteorological parameters, especially boundary layer condition and wind speed. During stable atmospheric condition, the pollutants released by different sources are trapped in lower atmosphere resulting in enhancement of the pollution level, whereas during the unstable conditions, strong horizontal winds disperse the trapped atmospheric pollution levels due to air mass transportation (Dutkiewicz et al., 2009; Srivastava et al., 2012a). In recent studies, Tiwari et al. (2013b) have also reported the impact of boundary