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Carbonaceous aerosols and pollutants over Delhi urban environment: Temporal evolution, source apportionment and radiative forcing

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HIGHLIGHTS

- Very high $PM_{2.5}$ (>200 μg m^{-3}) levels over Delhi during agricultural burning period.
- Enhanced levels of carbonaceous, sulphate and nitrate aerosol.
- OC/EC suggests dominance of biomass burning and SOA formation.
- BC highly contributes (50~70%) to the aerosol radiative forcing.

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ABSTRACT

Particulate matter ($PM_{2.5}$) samples were collected over Delhi, India during January to December 2012 and analysed for carbonaceous aerosols and inorganic ions (SO_4^{2-} and NO_3^-) in order to examine variations in atmospheric chemistry, combustion sources and influence of long-range transport. The $PM_{2.5}$ samples are measured (offline) via medium volume air samplers and analysed gravimetrically for carbonaceous (organic carbon, OC; elemental carbon, EC) aerosols and inorganic ions (SO_4^{2-} and NO_3^-). Furthermore, continuous (online) measurements of $PM_{2.5}$ (via Beta-attenuation analyser), black carbon (BC) mass concentration (via Magee scientific Aethalometer) and carbon monoxide (via CO-analyser) are carried out. $PM_{2.5}$ (online) range from 18.2 to 500.6 μg m^{-3} (annual mean of 124.6 ± 87.9 μg m^{-3}) exhibiting higher night-time (129.4 μg m^{-3}) than daytime (103.8 μg m^{-3}) concentrations. The online concentrations are 38% and 28% lower than the offline during night and day, respectively. In general, larger night-time concentrations are found for the BC, OC, NO_3^- and SO_4^{2-}, which are seasonally dependent with larger differences during late post-monsoon and winter. The high correlation ($R^2 = 0.74$) between OC and EC along with the OC/EC of 7.09 (day time) and 4.55 (night-time), suggest significant influence of biomass-burning emissions (burning of wood and agricultural waste) as well as secondary organic aerosol formation during daytime. Concentrated weighted trajectory (CWT) analysis reveals that the potential sources for the carbonaceous aerosols and pollutants are local emissions within the urban environment and transported smoke from agricultural burning in northwest India during post-monsoon. BC radiative forcing estimates result in very high atmospheric heating rates (~1.8–2.0 K day^{-1}) due to agricultural burning effects during the 2012 post-monsoon season.

1. Introduction

Carbonaceous aerosols (elemental carbon, EC; or equivalent black carbon, BC; organic carbon, OC) constitute a large fraction (~20 to 70%) of atmospheric aerosols and play a crucial role in the visibility degradation, formation of haze, atmospheric heating and adverse human health (Seinfeld and Pandis, 1998; Ramanathan and Carmichael, 2008; Chung et al., 2012; Ancelet et al., 2013; Pachauri et al., 2013). EC, as primary pollutant, is emitted from incomplete combustion of biomass burning, fossil fuel and carbon-contained materials and, due to its strong absorbing nature, is the most important driver to aerosol-induced global warming (Hansen et al., 2005; Bollasina et al., 2011; Bond et al., 2013) enhancing the melting of Himalayan

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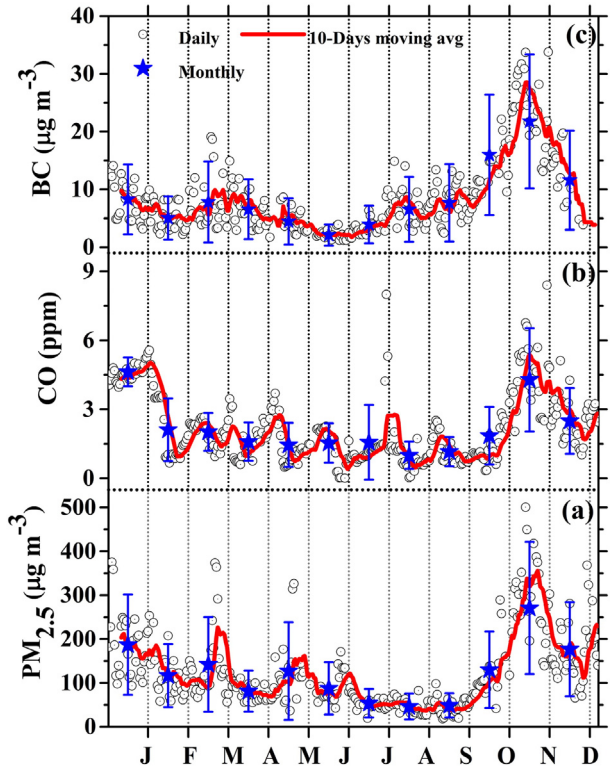

Fig. 1. Temporal evolution of daily-mean online measurements of $PM_{2.5}$ mass concentration (a), carbon monoxide (b) and BC mass concentration (c) during January to December 2012. The ten-day moving average along with monthly means are also shown. The vertical bars correspond to one standard deviation from the monthly mean.

glaciers via deposition over the snow and ice packs (Menon et al., 2010; Kaspari et al., 2011). On the other hand, OC, which is scattering rather than absorbing in nature, is directly released into the atmosphere from combustion and/or biogenic sources (primary OC; POC) or produced from gas-to-particle conversion of volatile organic compounds (secondary OC; SOC) (Pandis et al., 1992; Turpin and Huntzicker, 1995; Satsangi et al., 2012). OC can be also a mixture of several organic compounds, such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) some of which being carcinogenic in nature (Li et al., 2008; Feng et al., 2009). Recent studies (Andreae and Gelencsér, 2006; Bahadur et al., 2012; Chung et al., 2012) have shown that OC may also have a strong absorbing tendency in the UV-to-visible spectrum and significant influence on the aerosol radiative forcing (ARF).

Carbonaceous aerosols, either from human or natural emissions, are very abundant over Indian sub-continent and adjoining oceanic regions contributing to formation of the Atmospheric Brown Clouds (ABC) and Asian pollution outflow (Ramanathan et al., 2005, 2007; Lawrence and Lelieveld, 2010 and references therein). Due to their important role in degradation of visibility, formation of haze and fog over Ganges Basin, heating of the lower and mid-tropospheres and melting of Himalayan glaciers, carbonaceous aerosols and gases in the form of OC, BC, CO are systematically examined over India (Babu and Moorthy, 2002; Tripathi et al., 2005; Venkataraman et al., 2005; Rengarajan et al., 2007; Satsangi et al., 2010; Ram and Sarin, 2010, 2011, 2015; Sharma et al., 2010; Ram et al., 2012; Bisht et al., 2013; Dumka et al., 2013; Verma et al., 2013; Tiwari et al., 2013a, 2014a; Pipal et al., 2014a,b; Gadhavi et al., 2014; Safai et al., 2014; Srivastava et al., 2014) via multiple ground-based instrumentation and techniques as well as satellite remote sensing. The climatic response of carbonaceous aerosols on the weakening of the summer Indian monsoon has attracted the global scientific interest (Meehl et al., 2008; Cowan and Cai, 2011) and current modelling approaches deal with this issue over south Asia and Tibetan Plateau (Lau et al., 2006; Randles and Ramaswamy, 2008; Ganguly et al., 2012).

The current work examines a year-long (Jan–Dec 2012) set of Particulate Matter < 2.5 μm ($PM_{2.5}$) concentrations in the Delhi urban environment, which are analysed for the carbonaceous species (OC, EC) along with inorganic ions (SO_4^{2-} and NO_3^-). Furthermore, continuous real-time measurements of $PM_{2.5}$, BC mass concentration and CO support the analysis by providing the diurnal patterns, daily, monthly and seasonal averages. The main objectives of the current work are i) to improve the current knowledge of carbonaceous aerosols and pollutants over Delhi during a year with severe post-monsoon biomass burning, ii) to quantify the relative contribution of OC/EC in $PM_{2.5}$, iii) to understand the seasonality of carbonaceous aerosols and the relative importance of primary versus secondary OC sources, iv) to reveal the hotspot areas that mostly contribute to the high carbonaceous aerosol concentrations and, v) to examine the impact of BC on radiative forcing. In this respect, the current work uses various instrumentation and techniques and provides an extensive literature overview of the OC, EC and OC/EC over urban, rural, remote and elevated sites over the globe.

2. Site description and measurement set up

90 (45 for daytime and 45 for night-time) $PM_{2.5}$ samples were collected during January to December 2012 (3–5 samples per month) on

Table 1
Seasonal-mean concentrations of $PM_{2.5}$, OC, EC, BC, SO_4^{2-} and NO_3^- over Delhi during day and night. The seasons are classified as follows: winter (December–January); pre-monsoon (March–June); monsoon (July–September) and post-monsoon (October–November).

Months	$PM_{2.5}$ offline	OC μg m^{-3}	EC μg m^{-3}	OC/EC μg m^{-3}	SO_4^{2-} μg m^{-3}	NO_3^- μg m^{-3}	$PM_{2.5}$ online	BC μg m^{-3}	CO ppm
Day time (10:00 to 18:00 h local time)									
Winter	204.15 ± 23.93	46.28 ± 8.22	7.27 ± 2.20	6.72 ± 1.48	31.09 ± 10.65	13.05 ± 8.14	146.28 ± 83.02	6.29 ± 4.01	3.09 ± 1.07
Summer	157.39 ± 47.19	24.60 ± 6.67	3.45 ± 1.43	7.76 ± 2.03	20.05 ± 15.05	9.67 ± 11.23	90.22 ± 21.73	2.22 ± 1.35	1.40 ± 0.70
Monsoon	115.13 ± 27.13	26.42 ± 7.14	3.73 ± 1.01	7.33 ± 1.87	17.04 ± 7.81	7.48 ± 5.66	37.40 ± 13.65	3.83 ± 1.46	0.74 ± 0.42
Post-monsoon	199.44 ± 41.55	46.58 ± 12.06	8.34 ± 2.89	6.10 ± 2.05	33.73 ± 11.90	22.88 ± 6.48	168.04 ± 72.14	11.66 ± 3.94	2.65 ± 1.40
Night time (19:00 to 07:00 h local time)									
Winter	250.81 ± 25.98	62.42 ± 11.60	14.06 ± 3.60	4.72 ± 1.21	32.33 ± 11.68	19.79 ± 7.94	216.85 ± 85.61	12.77 ± 5.86	3.68 ± 1.46
Summer	155.34 ± 49.52	30.34 ± 16.97	8.54 ± 6.41	4.01 ± 1.69	13.31 ± 7.42	5.68 ± 5.97	82.32 ± 50.49	7.70 ± 7.25	1.80 ± 0.89
Monsoon	100.86 ± 17.76	26.89 ± 8.15	4.65 ± 1.65	6.10 ± 1.86	10.84 ± 4.60	6.52 ± 2.10	35.25 ± 12.77	6.30 ± 2.91	0.82 ± 0.37
Post-monsoon	234.83 ± 35.35	53.50 ± 12.11	15.75 ± 1.93	3.39 ± 0.56	37.15 ± 13.02	28.60 ± 8.25	228.86 ± 102.55	19.44 ± 4.65	3.11 ± 1.38
All data									
Day	164.16 ± 39.93	34.05 ± 11.66	5.29 ± 2.37	7.15 ± 1.23	23.66 ± 9.12	12.15 ± 6.79	103.80 ± 57.89	5.2 ± 3.83	1.81 ± 1.06
Night	179.02 ± 64.80	41.40 ± 17.62	10.30 ± 5.69	4.58 ± 1.46	21.29 ± 12.18	13.34 ± 10.12	129.39 ± 90.59	10.59 ± 6.75	2.18 ± 1.27
Total	171.59 ± 51.61	37.73 ± 14.32	7.79 ± 3.73	5.86 ± 0.99	22.47 ± 10.23	12.74 ± 8.18	116.57 ± 73.19	7.89 ± 5.14	1.99 ± 1.16