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# Carbonaceous aerosol over semi-arid region of western India: Heterogeneity in sources and characteristics

A.K. Sudheer [a,*], M.Y. Aslam [a,b], M. Upadhyay [c], R. Rengarajan [a], R. Bhushan [a], J.S. Rathore [c], S.K. Singh [a], S. Kumar [a]

[a] Physical Research Laboratory, Ahmedabad, 380009, India
[b] Indian Institute of Tropical Meteorology, Pune, India
[c] J.N.V. University, Jodhpur, India

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ABSTRACT

Carbonaceous species (elemental carbon (EC), organic carbon (OC), water-soluble organic carbon (WSOC)) and water-soluble inorganic species ($Na^+$, $NH_4^+$, $K^+$, $Ca^{2+}$, $Mg^{2+}$, $Cl^-$, $NO_3^-$, $SO_4^{2-}$) in $PM_{10}$ and $PM_{2.5}$ from Ahmedabad and Jodhpur (urban and semi-urban locations, respectively) in western India were measured during May–September, 2011. Stable isotope composition of carbonaceous aerosol ($\delta^{13}C$ of TC) in $PM_{10}$ samples was also determined. Average EC concentration in $PM_{10}$ at Ahmedabad was 1 μg $m^{-3}$ (range: 0.34 to 3.4 μg $m^{-3}$), almost 80% of which remained in $PM_{2.5}$. Similarly, 70% of EC in $PM_{10}$ (average: 0.9 μg $m^{-3}$) resided in $PM_{2.5}$ at Jodhpur. Average OC concentration at Ahmedabad was 6.4 μg $m^{-3}$ and ~52% of this was found in $PM_{2.5}$. On the contrary, OC concentration at Jodhpur was 40 μg $m^{-3}$, 80% of which was found in coarse particles contributing substantially to aerosol mass. $\delta^{13}C$ of TC (average: −27.5‰, range: −29.6 to −25.8‰) along with WSOC/EC ratio shows an increasing trend at Jodhpur suggesting the possibility of aging of aerosol, since aging results in enrichment of heavier isotope. OC and WSOC show significant correlations with $K^+$ and not with EC, indicating biogenic origin of OC. Different size distributions are also exhibited by WSOC at the two stations. On the other hand, $\delta^{13}C$ exhibits an inverse trend with sea-salt constituents at Ahmedabad, indicating the influence of air masses transported from the western/south-western region on carbonaceous aerosol. These results suggest that a strong heterogeneity exists in the sources of carbonaceous aerosol over this region and potential sources of non-combustion emissions such as bio-aerosol that need further investigation.



## 1. Introduction

Carbonaceous aerosol, composed of elemental carbon (EC) and organic aerosol, is a major component of atmospheric particulate matter; typically constitutes 20–50% of total fine particulate matter, and can be up to 90% in tropical forested areas (Kanakidou et al., 2005). This contributes substantially to aerosol direct and indirect effects on global climate (Ramana et al., 2010) and causes adverse human health issues (Knaapen et al., 2004; Nel, 2005). EC is emitted to the atmosphere from incomplete combustion of biomass, fossil fuel and other carbon-containing materials exclusively as a primary pollutant, and on the other hand, organic carbon (OC) is either directly released into the atmosphere from combustion and/or biogenic sources (primary OC) or produced from gas-to-particle conversion of volatile organic compounds (secondary organic carbon; SOC) (Pandis et al., 1992; Turpin and Huntzicker, 1995). Large fraction of organic aerosol is water-soluble and alters hygroscopic properties of aerosol and increases their CCN activity (Asa-Awuku et al., 2011). Though this important class of aerosol component has been subject of intense research over the past decade, its sources, physico-chemical characteristics, distribution over spatial and temporal scales and formation processes remain poorly understood. This is due to the vast variety of constituents at molecular levels, numerous pathways of formation of secondary organic aerosol (SOA), heterogeneity as well as large number of sources and poor constraints on emission factors. Concentrations of EC and OC in ambient air and their relative abundances (OC/EC ratios) are critical parameters to assess direct and indirect effects of aerosol on Earth–atmosphere system.

Studies on carbonaceous aerosol from South and Southeast Asian regions have gained more attention recently in the context of regional air quality and its impact on global climate. This is due to high emissions of pollutants to atmosphere from growing economies resulting from fuel combustion, bio-mass burning from forest fires as well as wide spread use of bio-fuels like cow dung cakes, wood and agricultural wastes causing the region as hot spot (Kirillova et al., 2013; Kumar et al., 2015; Moorthy et al., 2016 and references therein). Previously, it was reported that fossil fuel combustion and bio-mass burning would contribute

* Corresponding author.
*E-mail address:* sudheer@prl.res.in (A.K. Sudheer).

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about 80% and 20%, respectively, of the carbonaceous aerosol over the Indian sub-continent that transported to the adjacent oceanic region (Novakov et al., 2000). But, subsequent studies have demonstrated the dominance of bio-mass burning sources for carbonaceous aerosol over this region based on $^{14}C$ tracer technique (Gustafsson et al., 2009). In India, several studies have been carried out on black carbon measurements in order to assess their mass concentrations, optical depth, altitude profile and radiative forcing (Babu et al., 2002; Ganguly et al., 2006; Moorthy et al., 2004; Ramachandran and Kedia, 2010; Safai et al., 2008), however, there are only a few studies undertaken for the characterization of carbonaceous aerosol providing information on their concentration, sources as well as formation processes and their influencing factors (Behera and Sharma, 2010; Ram et al., 2012; Rengarajan et al., 2007; Rengarajan et al., 2011a, 2011b; Satsangi et al., 2012). These studies were done in either urban background locations or remote high altitude regions highlighting anthropogenic sources for various aerosol constituents and atmospheric processing of ambient aerosol caused by anthropogenic emissions.

Aerosol characterization and assessment of aging processes involving stable isotope composition of carbon in ambient aerosol, along with carbonaceous species are rather limited from the Indian region (Aggarwal et al., 2013; Agnihotri et al., 2011; Pavuluri et al., 2015). Carbonaceous aerosol carry the isotopic signatures (generally expressed as $\delta^{13}C$ and defined as $1000 \times (R - R_s) / R_s$ where R and $R_s$ are $^{12}C/^{13}C$ ratios in sample and standard, respectively) of their precursors and $\delta^{13}C$ value of carbon is a useful tracer to distinguish their origin, though, complexities arise due to a broad range of values as well as potential alteration of isotopic composition during atmospheric processes (Ke et al., 2007). Hence, it is difficult to draw general conclusion on source apportionment based on $\delta^{13}C$ values only, and regional studies are highly demanding (Masalaite et al., 2015 and references therein). Northwestern India is characterized by prominent sources of natural and anthropogenic aerosols; mineral aerosol from desert region (Thar Desert) and transport of marine aerosol during southwest monsoon may have profound influence on ambient aerosol on a regional scale. In this paper, carbonaceous species of aerosol, such as, EC, OC, and water-soluble organic carbon (WSOC) are presented along with $\delta^{13}C$ of total carbon (TC, defined as EC + OC) from two locations, separated aerially by ~350 km, situated in the semi-arid/arid region of western India, documenting their characteristics and source heterogeneity.

## 2. Experimental

### 2.1. Sampling Site and Meteorological Conditions

Fig. 1 depicts the sampling locations along with representative wind trajectories during the sampling period. Jodhpur (26° 14′N, 73° 01′E) is situated in the arid region in the Rajasthan state in north-western India with a population of more than one million. The mean annual precipitation recorded at this location is ~400 mm, of which more than 90% occurring during southwest monsoon (July to September). During this time, winds are mostly westerly/south-westerly with influence of marine air parcels from the northern Arabian Sea. The sampling site is situated in the eastern region of the Thar Desert with its emission of mineral dust to the ambient air. The sampling site is within a sparsely populated residential area, ~10 km southward from the main Jodhpur city in the upwind direction during the sampling period. So, local wind direction indicates that the site is less influenced by the emissions

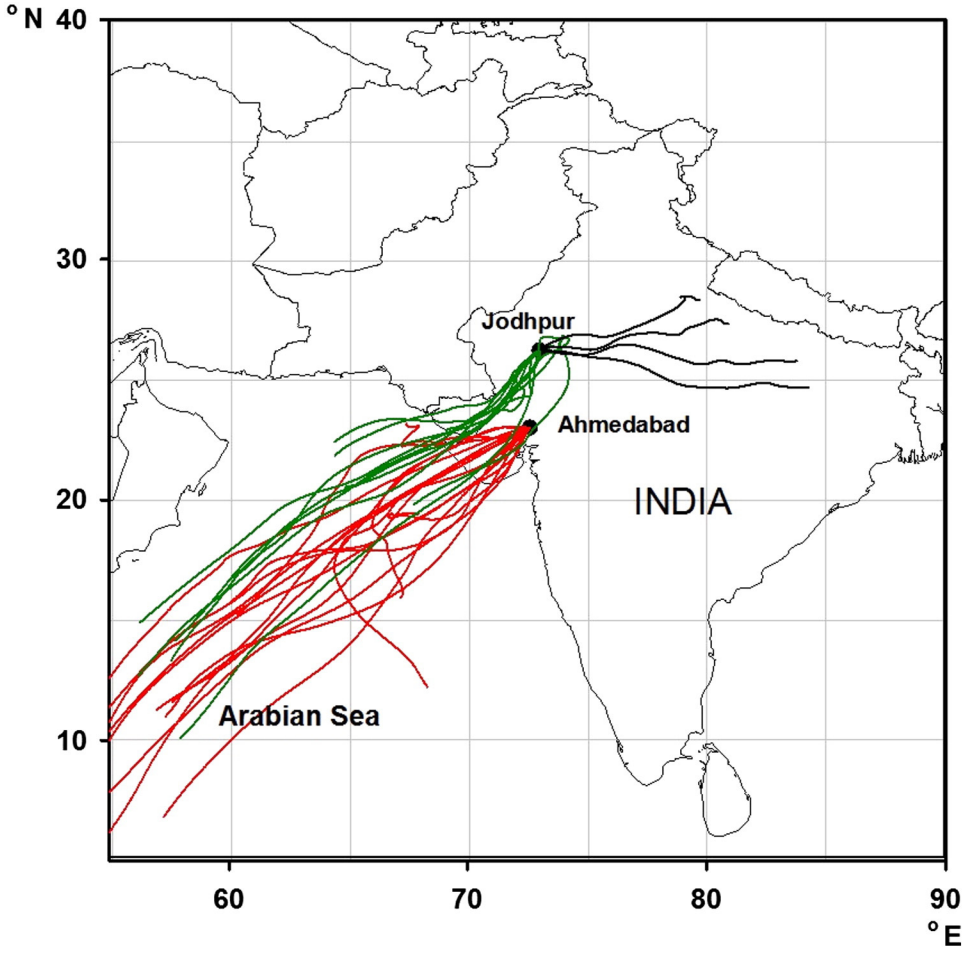


**Fig. 1.** Map showing sampling locations, Ahmedabad and Jodhpur along with 3-day back trajectories of air masses arriving at these locations during the sampling period.