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Original article

Chemical characterization and source apportionment of aerosol at an urban area of Central Delhi, India

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ABSTRACT

The concentrations of organic carbon (OC), elemental carbon (EC), water soluble inorganic ionic components (WSIC), and major & trace elements of PM₁₀ were studied in Delhi, an urban site of the Indo Gangetic Plain (IGP), India during January 2013 to June 2014. The average mass concentration of PM₁₀ recorded as $249.7 \pm 103.9 \mu\text{g m}^{-3}$ (average \pm standard deviation) with a range of 61.4–584.8 $\mu\text{g m}^{-3}$. The strong seasonal variation was noticed in the mass concentration of PM₁₀ and its chemical composition with maxima during winter (PM₁₀: $293.9 \pm 95.6 \mu\text{g m}^{-3}$; OC: $30.5 \pm 13.7 \mu\text{g m}^{-3}$; EC: $15.2 \pm 7.4 \mu\text{g m}^{-3}$) and minima during monsoon (PM₁₀: $143.9 \pm 36.3 \mu\text{g m}^{-3}$; OC: $19.9 \pm 16.2 \mu\text{g m}^{-3}$; EC: $7.4 \pm 5.4 \mu\text{g m}^{-3}$). The average concentration of major and trace elements (Na, Mg, Al, P, S, Cl, K, Ca, Si, Cr, Ti, As, Br, Pb, Fe, Zn and Mn) was accounted for ~18.5% of PM₁₀ mass. Results of Positive Matrix Factorization (PMF) model, HYSPLIT4 trajectory model, PSCF analysis and cluster analysis provide region of sources and its strength and types of sources of PM₁₀ over Delhi. Positive PMF provides that the major source of PM₁₀ are soil dust (22.7%) followed by secondary aerosols (20.5%), vehicle emissions (17.0%), fossil fuel burning (15.5%), biomass burning (12.2%), industrial emissions (7.3%) and sea salts (4.8%) at the observational site of Delhi. The cluster analysis of air mass trajectories calculated by HYSPLIT model indicates that the air mass approaches to the observational site mainly from 4 sides (north-western IGP, Pakistan (10%); north-western IGP, Northwest Asia (45%); eastern IGP (38%); Pakistan and Arabian Sea (6%)) during study. Potential Source Contribution Function (PSCF) analysis also supports the cluster analysis indicating that the concentration of PM₁₀ mass contributed, is mainly from IGP region (Uttar Pradesh, Haryana and Punjab etc.), Afghanistan, Pakistan and surrounding areas.

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1. Introduction

Atmospheric aerosols significantly affect the atmospheric chemistry, ambient air quality, visibility and the Earth's radiation budget. During the last decades, air pollution has become an increasing concern, especially in urban areas, due to adverse effect on human health (Pope and Dockery, 2006; Ho et al., 2007). Several studies have revealed that aerosols, especially fine mode (particulate matter having an aerodynamic diameter $\leq 2.5 \mu\text{m}$) particles,

can lead to serious human health effects like cardiovascular and respiratory disorders (Dockery and Pope, 1994). Respiratory and cardiovascular illnesses related to particulate matter (PM) exposure have been well documented (Ramgolam et al., 2009; Pope et al., 2009). Therefore, identification of PM sources is necessary to develop air quality improvement strategies in order to be able to control and reduce ambient PM concentrations through targeted action (Waked et al., 2014). To address this issue, many tools have been used for the identification and quantification of PM sources (Paatero and Tapper, 1994; Paatero, 1997; Ulbrich et al., 2009).

Identification and quantification of different type sources that contributes to ambient concentration of pollutants is one of the major issues of the urban air quality research. Hence, the development and application of improved tools are required for the identification and apportionment of atmospheric aerosols. Receptor modeling offers a method to complete the process of

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measured of the pollutant concentrations at a sampling site (Hopke, 1991). Recently, PMF model has been improved significantly and a new approach was developed by Paatero (Paatero and Tapper, 1994; Paatero, 1997), using a least squares approach. PMF solves the problem arising in factor analysis by integrating non-negativity constraints in the optimization process and utilizing the error estimates for each data value as a point-by-point weight (Begum et al., 2004). PMF has been applied successfully worldwide for such studies (Polissar, 1998; Kim and Hopke, 2004; Lee and Hopke, 2006; Karanasiou et al., 2009). Different models including principal component analysis/absolute principal component scores (PCA-APCS), edge analysis (UNMIX), chemical mass balance (CMB) and PMF have been applied by several researchers to identify and establish the sources contributing to ambient air. Rizzo and Scheff (2007) compared the magnitude of source contributions resolved by each model and examined correlations between PMF and CMB resolved contributions. They observed that the major factors correlated well and were similar in magnitude. Additionally, PMF resolved source profiles were generally similar to measured source profiles. Recently, Callén et al. (2009) carried out source apportionment of PM₁₀ in Zaragoza, Spain by three multivariate receptor models based on factor analysis: PCA-APCS, UNMIX and PMF. Special attention was paid to the models comparison in order to determine which were more adequate for the apportionment. They concluded that greater requirements of measure of uncertainty in PMF permitted to obtain better results than with the other two models: PCA-APCS and UNMIX. Therefore, in the present study, source apportionment of PM₁₀ has been carried out using PMF model.

Delhi is distributed over 1484 km² with population density of 11,450 km² and surrounded by highly industrialized National Capital region (NCR) with population density of 1050 km². The ambient air quality of the mega city Delhi is deteriorating with time. The sources of atmospheric pollutants are localized and heterogeneous and depended on season (Nagpure et al., 2013; Mohan et al., 2012). The combination of factors including industries, power plants, domestic combustion of coal and biomass, and transport (direct vehicle exhaust and indirect road dust) is contributing to air pollution (Garg et al., 2006; Gurjar et al., 2004). Even the total vehicle population in Delhi has amounted to more than that of three metros, Mumbai, Kolkata and Chennai, put together. To meet the challenge of urban growth in Delhi, several steps (introduction of compressed natural gas (CNG) in public transport, relocation of industries and introduction of Metro transport etc) have been initiated. The present source apportionment study of aerosol, can help the stakeholders and policymakers to understand the influence of regional and local sources of PM₁₀ in urban areas and thus to identify effective emission control strategies to improve the ambient air quality.

In this work, we report chemical characteristics and source apportionment of PM₁₀ mass concentration in an urban site of Central Delhi, India during January 2013 to June 2014. The PMF receptor model was applied for the source apportionment using chemical composition (organic carbon, elemental carbon, major ions, metals and trace elements) of PM₁₀ mass.

2. Methodology

2.1. Site description

PM₁₀ samples ($n = 86$) were collected (5–6 samples in a month) periodically at sampling site of the CSIR-National Physical Laboratory (CSIR-NPL), New Delhi [(28°38'N, 77°10'E; 218 m above mean sea level (amsl)], India (Fig. 1) during January 2013 to June 2014. The sampling site is amenable to free wind flow from all the directions.

The sampling location represents a typical urban atmosphere, surrounded by huge roadside traffic (~100 m) and agricultural fields in the southwest direction (~500 m). There are different small, medium and large scale industries in and around Delhi. Since we consider traffic could be one of the major sources of pollutants in mega city like Delhi, it is to be reminded that the total number of registered vehicles in the city was in the order of 7.77 million in 2012–13 (Delhi Statistical Handbook, 2013). Roadside vehicle, industrial emission and biomass burning etc. could be the major sources of carbonaceous aerosols and several other pollutants. The occasional occurrence of dust storms may contribute the presence of mineral dust significantly to the aerosol loading in summertime (Ram et al., 2010; Sharma et al., 2014b; Kumar and Sarin, 2009; Ram et al., 2012). This area is under the influence of air mass flow from North-east to North-west in winter and from South-east to South-west in the summer (Goyal and Sidhartha, 2002; Sharma et al., 2014a). In addition, Delhi experienced severe fog and haze weather conditions and poor visibility during wintertime. The temperature of Delhi varied from minimum (monthly average: 12.7 °C) in winter (November to February) to maximum (monthly average: 35.2 °C) in summer (March to June). The average rainfall in Delhi during monsoon (July to October) was in the order of ~735 mm.

2.2. Sampling method

PM₁₀ samples were collected on quartz fiber filters (pre-combusted at 550 °C at least 5 h before desiccated and sample collection) by using Fine Particle Sampler (APM 550, Make: M/s. Envirotech, India) at 10 m height (above ground level). Ambient air was passed through a quartz filter (QM-A; 47 mm) at a flow rate of 1 m³ h⁻¹ (accuracy ± 2%) for 24 h during the sampling period. The flow meter of the sampler was calibrated (with the accuracy of ±2% of Full Scale) with Air Flow Calibrator traceable to National Standard. The QM-A filters were weighed before and after the sampling during the experiment in order to determine the mass of the PM₁₀ collected. The amount of PM₁₀ (μg m⁻³) was calculated on the basis of the difference between initial and final weights of the QM-A filters measured by a micro balance (M/s. Sartorius, resolution: ± 1 μg) and by dividing the amount of total volume passed during the sampling. After collecting samples, filters were stored under dry condition at -20 °C in the deep-freezer prior to analysis.

2.3. Chemical analysis

Analysis of OC and EC of ambient PM₁₀ samples ($n = 86$) have been carried out by OC/EC carbon analyzer (Model: DRI 2001A; Make: Atmoslytic Inc., Calabasas, CA, USA) following the USEPA Method 'IMPROVE Protocol' with negative pyrolysis areas zeroed. The principle of the OC/EC carbon analyzer (DRI 2001A) is based on the preferential oxidation of OC and EC at different temperatures in which the sample is heated to four temperature plateaus (140, 280, 480 and 580 °C) in pure helium and three temperature plateaus (580, 740 and 840 °C) in 98% helium and 2% oxygen. Its function relies on the fact that OC can be volatilized from the sample deposit in a non oxidizing helium atmosphere, while EC must be combusted by an oxidizer. The principal function of the optical component (laser reflectance and transmittance) of the analyzer is to correct for pyrolysis, charring of OC compounds into EC. The thermal optical reflectance (TOR) charring corrections are not necessarily the same, owing to charring of organic vapors within the QM-A filter (Chow et al., 2004). Approximately 0.536 cm² area of QM-A filter was cut using the proper punch and the values are reported as μg cm⁻² as given by the instrumental

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