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Statistical assessment of respirable and coarser size ambient aerosol sources and their timeline trend profile determination: A four year study from Delhi

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ABSTRACT

A reliable identification of sources and their relative time dependent contributions to ambient aerosol load is an important air pollution research problem. Given the inherent complexity of contributing sources in urban/mega-cities, an appropriate statistical investigation is needed to characterize sources and to understand their timeline trend profiles. Daily average ambient particulate matter (PM) loads, PM_{10} (aerodynamic diameter <10 μm) and coarser particulate matter (CPM: aerodynamic diameter >10 μm) were collected once a week over 4 years at a receptor site in Delhi. The samples were analyzed to quantify the presence of 17 marker elements. Time series data of PM loads, and that of associated marker elements was subjected to Positive Matrix Factorization (PMF) to identify sources and to quantify their contributions to each PM fraction with reference to the associated marker elements. The resolved time series data of each contributing source was further subjected to Ensemble Empirical Mode Decomposition (EEMD) analysis to extract their timeline trend profile over four years in CPM and PM_{10} load. Three sources contributed to the CPM load: anthropogenic (15%), primary crustal (59%), and fine crustal dust (26%). Four sources contributed to the PM_{10} load: coarser grain crustal material (9%), fine grain crustal material (12%), industrial and vehicular emissions (23%), and wind assisted transport and re-suspension of surface dust (56%). The timeline trend of sources contributions to CPM and PM_{10} displayed a non-linearity. The unique composite-PM_{10} source contributed maximum to the ambient PM_{10} load. Distinct underlying processes of this source involved convective re-suspension and city-wide cleaning associated upliftment of surface deposits back into the ambient environment.

1. Introduction

Ambient aerosols have a significant bearing, direct or indirect, on the state of the surrounding environment: climate forcing through light absorption and scattering (Bellouin et al., 2005; Hansen et al., 2011); atmospheric visibility (Singh and Dey, 2012; Tandon et al., 2013); atmospheric chemistry and photochemistry (Dentener et al., 1996; Andreae and Crutzen, 1997; Dickerson et al., 1997); cloud formation and precipitation (Rosenfeld et al., 2008; Kumar and Yadav, 2014) and nutrient dispersion (Reheis et al., 1995). Aerosol air pollution also impacts human health adversely by inducing respiratory ailments among inhabitants exposed to respirable aerosol arising from combustion sources (Dockery and Pope, 1996; Kandlikar and Ramachandran, 2000; Pope III, 2000). In recent years, a considerable increase in ambient aerosol load has been reported from many urban areas in developing countries due to increased economic development activities and related impact on aerosol emitting sources. The activity of the sources manifest a significant temporal variability in ambient aerosol load and their composition (diversity of organic compounds, biological materials and transition elements) from region to region (Griffin, 2007; Yadav et al., 2013a, 2013b). To have a reliable and meaningful understanding about the time dependent variations in the contributions

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from the identified aerosol sources, it is essential to have estimates of their timeline trend profile (TTP) in an investigation done over long time span (Tandon and Attri, 2011). Conventionally, identification and characterization of aerosol contributing sources has been done using multivariate statistical analysis of chemical constituents (variables) present in aerosol samples (Srivastava and Jain, 2008; Tandon et al., 2010). Ground based investigations done over a short time span are limited to reveal, at best, the identity of the contributing sources without providing any understanding about their TTP. Obtained TTP of the respective contributing sources of different aerosol size fractions enables the appraisal of their temporal variability; a first step towards addressing the issue of aerosol air pollution. However, to obtain a statistically significant TTP estimates at 95% confidence ($\pm 2\sigma$) requires an appropriate accounting of the presence of multi-scale cyclic influences (monthly, seasonal and annual) present in the collected aerosol load over time, which appear in the form of oscillations in aerosol temporal load profiles (Weatherhead et al., 1998).

In India, the ambient environment of New Delhi has a dubious distinction of being one of the most polluted cities of the world. The ambient aerosol load exceeds air pollution criteria threshold concentration on most of the days in all seasons, and aerosol contributing sources manifest time dependent complexity in their activity. To the best of our knowledge no ground based investigation has been attempted to determine the TTP following the identification of the aerosol contributing sources to different size fractions in Delhi region. Most of the investigations remained focused on the source identification by using multivariate statistical analysis of selected chemical constituents. Furthermore, such studies were mainly short termed (Karar and Gupta, 2007; Srivastava and Jain, 2008; Tandon et al., 2008; Khillare and Sarkar, 2012).

The work presented involves collection of PM_{10} and coarser particulate matter (CPM: aerodynamic diameter > 10 μm) in a time series over four years (2006–2009) in Delhi region. The samples were further analyzed to quantify 17 marker elements. Positive matrix factorization (PMF) analysis of PM_{10}, CPM loads and associated marker elements was done to determine the number of sources contributing to the respective aerosol size fraction. The determination of TTPs required an accounting of multi-scale cycles present in the aerosol loads and their contributing sources. Time series data obtained from PMF analysis for the PM_{10}, CPM loads, and that for the respective contributing sources was subjected to Ensemble Empirical Mode Decomposition (EEMD) to extract their respective TTP. EEMD circumvents the presence of cyclic influences by stepwise extraction of embedded multi-scale cycles as Internal Mode Functions (IMFs) from the respective time series and obtain TTP. The obtained TTPs adequately provide the time dependent variability in the respective loads (CPM, PM_{10}) and in their contributing sources. The TTPs of the respective contributing sources is a valued information, which can be used to evolve a mitigation strategy to lower the ambient aerosol air pollution.

2. Experimental

2.1. Description of the sampling site

The capital region of Delhi is geographically located between 28°25′–28°53′ N and 76°50′–77°22′ E at 216 m above mean sea level in the northern India. It has a semi-arid climate influenced by the Himalayan mountainous ranges located to the north and the Thar-Desert to the west. The hot peninsular region lies to the south and sub-humid Gangetic plains to the east. The population of Delhi region is 16,753,235 (Census of India (2011). Many factors contributes to the ambient aerosol load in Delhi region such as wind-blown dust transported from the great Indian Thar desert by S-SW winds during summer; lowering of planetary boundary layer (PBL) during winter and combustion linked emissions from vehicles, thermal power plants, biomass burnings (Yadav and Rajamani, 2004, 2006). Additional factors may also include construction activity and re-suspension of surface deposited dust through city-wide street cleaning practice (Tandon et al., 2008). The sampling location (Fig. 1) selected for this investigation, Jawaharlal Nehru University (JNU) is a known receptor site (Singh et al., 1997).

2.2. Sampling and chemical analysis

The respirable dust sampler (Envirotech, model – APM 460 BL), operated at constant flow rate 1.1–1.2 m^3 min^{-1}, was used to collect aerosol size fractions in a time sequence, over 24 h. PM_{10} samples were collected on a pre-baked quartz filters and CPM (aerodynamic diameter >10 μm) in a cup attached to an improved cyclone configuration having a sharp cut off (D50 at 10 μm). The samples were collected once every week, over a span of four years, starting from January 2006 to December 2009. Mass concentration of the samples was determined gravimetrically, using an electronic microbalance with an accuracy to measure 0.01 mg. The collected samples were stored under controlled temperature at −18 °C prior to their analysis. The average monthly load for a given month was derived from the weekly sample's load. The samples in each size fraction were analyzed to estimate the presence of seventeen marker elemental variables (Al, Ba, Ca, Cd, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Pb, Sr, Ti, V, Zn) using a Inductively Coupled Plasma Atomic Emission Spectrometer (Jobin Yvon make, model Ultima 2). Presence of Si in CPM load was analyzed by solution-A method. The details of the methods for Silica analysis, sample digestion protocol for estimating the elements in CPM and PM_{10} is explained elsewhere (Yadav and Rajamani, 2004; Pruseth et al., 2005). In case of PM_{10} Si could not be determined as the filter matrix (Quartz) contained Si. However, the use of Quartz filter was necessary for the analysis of the PM_{10} samples for estimating the presence of elemental carbon and total carbon using thermal/optical method.

2.3. Positive matrix factorization based analysis to identify CPM and PM_{10} sources

The identity of the sources and their contributions (μg m^{-3}) to the CPM and PM_{10} load was estimated by Positive Matrix Factorization (PMF) by taking into account the estimated marker elements concentrations as variables (Paatero and Tapper, 1994). Details of method, and other relevant information obtained from PMF analysis (uncertainty of each species, species weights, Q values for data and PMF resolved Q values) of CPM, PM_{10} and species data are provided in section 1. of the supplementary material of this manuscript. The sources contributing to the CPM and PM_{10} loads were characterized on the basis of the attributes of the grouped marker elements in the segregated factor loadings obtained from PMF analysis. PMF analysis was done on 71 samples of each size fraction (CPM and PM_{10}). The samples corresponding with the known specific events such as festivals of Diwali involving large-scale fireworks were not included in the analysis.

2.4. Ensemble empirical mode de-composition (EEMD) analysis of CPM, PM_{10} loads and their contributing factors (sources) to determine TTPs

The EEMD method, an improved version of Empirical Mode Decomposition (EMD) algorithm developed by Huang et al. (1998a), was used to analyze the time series data of CPM, PM_{10}, and their contributing sources (Huang et al., 1998a, 1998b; Wu et al., 2011). The method has been applied to extract the presence of multi-scale