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Chemical composition and source apportionment of PM_{2.5} and PM_{2.5–10} in Trombay (Mumbai, India), a coastal industrial area

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ABSTRACT

PM_{2.5} and PM_{2.5–10} concentrations, elemental constituents, and sources in a densely populated coastal industrial area (Trombay, Mumbai) were investigated in 2010 and 2011. The PM_{2.5} and PM_{2.5–10} concentrations were 13.50–71.60 and 22.40–127.78 $\mu\text{g}/\text{m}^3$, respectively. The daily PM_{2.5} concentrations exceeded the Indian Central Pollution Control Board limit (60 $\mu\text{g}/\text{m}^3$) several days in winter. Of the elements analyzed, Si then Al had the highest concentrations in PM_{2.5–10}, but black carbon then Si had the highest concentrations in PM_{2.5}. The element concentrations varied widely by season. Al, Ca, Fe, Si, and Ti concentrations were highest in summer, Cl, Mg, and Na concentrations were highest in the monsoon season, and the other trace metal concentrations in both PM_{2.5} and PM_{2.5–10} were highest in winter. The PM_{2.5} and PM_{2.5–10} sources were apportioned by positive matrix factorization. PM_{2.5} and PM_{2.5–10} had six dominant sources, crustal material (8.7% and 25.3%, respectively), sea salt spray (6.1% and 15.0%, respectively), coal/biomass combustion (25.5% and 13.8%, respectively), fuel oil combustion (19.0% and 11.2%, respectively), road traffic (17.7% and 12.6%, respectively), and the metal industry (10.6% and 7.0%, respectively). Anthropogenic sources clearly contributed most to PM_{2.5} but natural sources contributed most to PM_{2.5–10}.

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Introduction

Air pollution in India is very severe. Accelerating industrialization, transport use, urbanization, construction, economic growth, and energy demands have caused air quality to deteriorate severely, especially in terms of particulate matter (PM) in Indian cities (CPCB, 2010; Tiwari et al., 2015). The International Agency for Research on Cancer classes PM in outdoor air as a Group I carcinogen (Hamra et al., 2014). Atmospheric PM pollution, especially PM_{2.5} (PM with aerodynamic diameters $\leq 2.5 \mu\text{m}$) and PM_{2.5–10} (PM with aerodynamic diameters of 2.5–10 μm), is a major current problem because of the adverse effects PM has on human health and the environment. PM_{2.5} and PM₁₀ exceeding the Indian Central Pollution Control Board (CPCB) annual limits have been found in most Indian cities (Banerjee, Murari, Kumar, & Raju, 2015). There is great interest in PM because of increasing PM concentrations in the atmosphere and the negative effects of PM on atmospheric chemistry, visibility, coastal ecosystems (because of air–water exchange), and the global climate (Ramanathan, Crutzen, Kiehl, & Rosenfeld, 2001;

Yatkin & Bayram, 2008). Exposure to PM is associated with diseases ranging from lower respiratory tract infections to cerebrovascular disease (Hamra et al., 2014; Mimura et al., 2014). Worldwide, about 8% of lung cancer deaths, 5% of cardiopulmonary deaths, and 3% of respiratory infection deaths have been attributed to PM exposure (Cohen et al., 2005). The World Health Organization uses PM as an indicator when evaluating air pollution that damages health (An, Hou, Li, & Zhai, 2013; Tao, Mi, Zhou, Wang, & Xie, 2014).

Chemically characterizing PM is very important to epidemiological and source apportionment studies, and allows radiative forcing associated with PM to be quantified in climate studies. PM is a complex mixture of anions, cations, mineral dust, trace elements, organic carbon, elemental carbon, and water. The relative abundances of these components vary widely, spatially, and temporally, because of the natures of the sources (Li, Wiedinmyer, & Hannigan, 2013). PM_{2.5} is mainly emitted during combustion, so is emitted in engine exhaust and during fossil fuel combustion for power generation, biomass combustion, and metal smelting (Cheng, Liu, & Chen, 2010). PM_{2.5–10} has non-wind-suspended and wind-suspended components. Anthropogenic sources, such as construction activities, open-cast mining, and mechanical processes (e.g., tire and brake emissions), contribute to the non-wind-suspended component. Natural sources, such as volcanic emissions, agricultural tilling, sea spray, biological particles, surface soil, and dust on paved

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areas, contribute to the wind-suspended component (Cheng et al., 2015).

It is important to identify the contributions of the different sources to decrease or control PM pollution. Source apportionment studies are indispensable when attempting to identify PM sources and quantify their contributions. Multivariate receptor models are very popular tools for source apportionment studies. Such models have been used to identify and quantify PM sources around the world (Bove et al., 2014; Geng et al., 2013; Gummeneni, Yusup, Chavali, & Samadi, 2011; Karnae & John, 2011; Minguillón et al., 2014; Seneviratne et al., 2011; Sharma et al., 2014). Principal component analysis/absolute principal component scores, edge analysis, chemical mass balance, and positive matrix factorization (PMF) are examples of receptor models. PMF is a powerful model capable of resolving PM sources without prior knowledge of the sources (Cohen, Crawford, Stelcer, & Bac, 2010; Mooibroek, Schaap, Weijers, & Hoogerbrugge, 2011). PMF has many advantages over other models (Ancelet, Davy, Trompetter, & Markwitz, 2014).

The adverse effects of PM and increasing interest in PM studies led us to perform the study presented here in Trombay, a densely populated coastal industrial area in Mumbai, India. The Trombay site is near various large and small industrial plants, port areas, and areas affected by heavy traffic, which can all contribute to PM pollution. Little information is available on particulate-bound metal concentrations, temporal and seasonal variations in PM concentrations, or the specific sources of PM at the site. The study lasted two years, from January 2010 to December 2011, and the goals were to assess temporal and seasonal variations in $PM_{2.5}$ and $PM_{2.5-10}$ and particle-bound element concentrations and to identify and quantify the sources of PM at the study site.

Materials and methods

Site description

Trombay (19.0°N, 72.9°E) is a northeastern suburb of Mumbai, India, and covers about 25 km². The sampling site was about 20 km from Mumbai center, with the Arabian Sea on one side and an industrial area on the other. A detailed map showing the locations of the site and the surrounding industrial plants is shown in Fig. 1. The sampling site was near a coal- and oil-burning power plant, oil refineries, a fertilizer complex, and automobile, metallurgical, textile, chemical, paint, and small metal industry plants. The sampling site was also near several other industrial areas (Maharashtra Industrial Development Corporation, Talaja, Turbhe, Trans Thane Creek, and others). Mumbai port and Jawaharlal Nehru (Nhava-Sheva), the largest ports in India, handle liquid chemicals, crude oil, petroleum products, and other commodities. The sampling site was a few kilometers away from each port. Mumbai is densely populated and has worse traffic problems than other Indian cities.

Mumbai has three distinct seasons, winter (October–February), summer (March–June), and the monsoon (July–September). The mean summer and winter maximum temperatures are 32 and 30 °C, respectively, and the mean summer and winter minimum temperatures are 25 and 20.5 °C, respectively. Mumbai receives heavy rainfall (300–800 mm) during the monsoon, and mean annual rainfall is 2240 mm. Winter winds are generally light and from the north and northeast. Monsoon winds are stronger and are moist and from the southwest (from marine areas). The wind speeds at the study site were 0.5–7.5 m/s in summer and winter but as high as 17 m/s in the monsoon.

PM sampling and gravimetric analysis

Each PM sample was collected using a Gent's dichotomous sampler with a stacked filter unit containing two 47 mm diame-

ter Nuclepore polycarbonate filters (one coarse, with 8 µm pores, and one fine, with 0.4 µm pores) in series. The mean flow rate was 16 L/min. The pre-impaction stage intercepted particles with aerodynamic diameters >10 µm. The coarse filter with 8 µm pore size collected $PM_{2.5-10}$, and the fine filter with 0.4 µm pore size collected $PM_{2.5}$. Each sample was collected for 24 h, and a sample was collected twice a week from January 2010 to December 2011. Each filter was preconditioned (equilibrated for 24 h at room temperature and 50% ± 5% relative humidity) and then weighed using a Mettler AE240 electronic microbalance (Mettler Toledo, Switzerland, error ±10 µg) at least three times before and after sampling to determine the mass of particles collected and to ensure the measurement variance was low. The net PM mass was calculated by subtracting the pre-sampling filter weight from the post-sampling filter weight. The net mass was divided by the air volume to give the daily PM concentration in µg/m³.

Chemical analyses of $PM_{2.5}$ and $PM_{2.5-10}$

Elemental analyses of the particles were performed using a Xenometrix EX-6600 energy dispersive X-ray fluorescence (EDXRF, Xenometrix, Israel) spectrometer with a Rh anode (the X-ray source) with a 60 kV, 6 mA (400 W) power supply. The system had a Si (Li) detector (resolution 150 eV at 5.9 keV Mn X-ray) and various secondary targets. The primary X-ray beam spectrum was modified using the secondary targets Ge, Mo, Si, and Ti and was then used to excite the elements in a sample. The concentrations of 18 elements (Al, As, Ca, Cl, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Pb, S, Si, Ti, V, and Zn) were determined. The best sensitivity was achieved using the optimum voltages, analysis times, and currents shown in Table 1. The instrument was calibrated using thin-film Micromatter Standards, then the analyte concentrations in the samples were determined using the calibrations. The EDXRF measurement method was validated using NIST SRM 2783 (air particulate on filter media, NIST, USA) reference material. The measured concentrations were within 6% of the NIST SRM 2783 certified values.

The black carbon (BC) concentrations in the $PM_{2.5}$ samples were determined by making reflectance measurement using an EEL 43D smoke stain reflectometer (Evans Electro Selenium Limited, USA), in which light from a tungsten lamp was projected onto a well-defined spot on the sample and the reflected light measured by photocells in the black housing. Reflectances were measured at five points on each filter, and the mean value was used to estimate the BC concentration using a mass absorption coefficient of 9.7 m²/g and a formula presented by Salako et al. (2012). The reflectometer was calibrated following the appropriate instructions using a blank filter from the same batch as the filters used to collect the samples.

Source apportionment

The sources of the PM were apportioned using the US Environmental Protection Agency (EPA) PMF model version 5.0. PMF decomposed the data matrix X ($n \times m$; n and m being the numbers of samples and chemical species, respectively) into two matrices, factor contributions G ($n \times p$) and factor profiles F ($p \times m$) with non-negative constraints (p is the number of factors extracted) (Paatero, Eberly, Brown, & Norris, 2014). PMF is described in more detail elsewhere (Geng et al., 2013; Guinot, Gonzalez, De Faria, & Kedia, 2016; Minguillón et al., 2014; Pakbin, Ning, Shafer, Schauer, & Sioutas, 2011; Rahman et al., 2011). Uncertainties for the data points could affect the PMF results, and the measured uncertainty was an input parameter for the PMF analysis. The measurement uncertainty was calculated using Eq. (1) (Polissar, Hopke, Paatero, Malm, & Sisler,

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