Abstract

The rapid growth of industrialization and increased energy demand have led to the deterioration of air quality in urban and peri-urban cities. Fine particulate matter, particularly atmospheric particulate matter (PM) with a diameter smaller than 2.5 µm ($PM_{2.5}$), significantly affects climate forcing and human well-being. At significant levels, prolonged contact with polluted air can lead to various human health-related problems. To expedite particulate matter removal strategies, sources contributing to the pollution need to be identified, apportioned, and removed. This study explored the highly time-resolved variation of the $PM_{2.5}$ bound species sources contributing to both the inorganic and organic constituents, using state-of-the-art real-time online instruments in Delhi, India. We performed the detailed long-term source apportionment, including different components of $PM_{2.5}$ (organics, elements, and black carbon), to provide valuable insights into Delhi's composition and particulate air pollution sources. We found that significant proportion of Delhi's ambient air particulate matter is attributed to the power plant (25-30%), biomass burning (BB) (25-35%), traffic emissions (20-30%), dust-related (5-15%) and industrial emissions (4-7%). Organic aerosol (OA) dominated mass fraction in all seasons, followed by other non-refractory species (SO_4^{2-}, NO_3^-, NH_4^+, and Cl^-). We observed marked seasonal and diel variability in the concentration and composition of $PM_{2.5}$ owing to the interactions of sources and atmospheric processes. Winter and post-monsoon were the most polluted periods of the year. The apportionment of organic aerosol (OA) yielded primary factors, hydrocarbon-like organic aerosol (HOA), biomass-burning organic aerosol (BBOA), and secondary factors, oxygenated organic aerosols, semi-volatile oxidized OA (SVOOA), low volatile oxidized OA (LVOOA1 and 2). On the other hand, eight sources are identified from the source apportionment of elements such as power plant dust, biomass burning, vehicular emission, secondary chloride, waste incineration, coal combustion, and industrial emission in all seasons. During the Diwali period, fireworks were identified as an additional source of pollution. The secondary chloride factor was dependent on the local wind direction (NW direction, 302–333°). Dust factor was found to dominate the elemental source apportionment in the summer season. Multiple high-loading dust storms were observed in the summer season. During the winter, power plant factors dominate the elemental source apportionment as the formation of sulfate particles was facilitated in the presence of higher RH and aerosol liquid water content (ALWC), indicating the role of rapid aqueous phase formation pathway in secondary aerosol haze formation. However, researchers have integrated data from

multiple instruments to better understand PM sources and their chemical composition to examine PM characteristics. Accordingly, in this study, source apportionment was also performed on a combined dataset consisting of non-refractory $PM_{2.5}$ organic factors, elements, and BC; we evaluated two disparate factor analytic methodologies – namely, double-PMF (D-PMF) and Tracer-conjugate PMF (TC-PMF). The D-PMF, as well as TC-PMF, improved the interpretation of organic factor sources, such as apportioning considerable contributions of LVOOA2 (85%) to the power plant factor, which is often linked with regionally carried aged organic aerosols in the organics only PMF(O-PMF). Lastly, the D-PMF results significantly agreed with TC-PMF, indicating that either of the two techniques could be used to unmix the complex variety of $PM_{2.5}$ sources in the Delhi-NCR region and, arguably, the larger Indo-Gangetic Plains also, since more than 50 % of that organic aerosol mass concentration is comprised of secondary organic aerosol (SOA). However, the underlying formation mechanism of SOA remains poorly understood in Delhi due to chemical complexities, numerous sources of organic precursors, and evolutionary processes. This study also examined the impacts of photochemical and aqueous-phase processing on factors related to SOA.