Chemical Characteristics of Water Soluble Components of Fine Particulate Matter, PM$_{2.5}$, at Delhi, India

Suresh Tiwari$^1$, Manoj K. Srivastava$^2$, and Deewan S. Bisht$^1$

$^1$Indian Institute of Tropical Meteorology-Pune, New Delhi Branch, Prof. Ram Nath Vij Marg, New Delhi-110060

$^2$Department of Geophysics, Banaras Hindu University, Varanasi – 221005

Abstract

Aerosol samples in the size range up to 2.5µm were collected from January to December, 2005 at New Delhi, India, as a pilot experiment and analysed for organic (Oxalate and Formate) and inorganic (Sulfate, Ammonium, Nitrate, Potassium, Chloride, Sodium, Calcium and Magnesium) chemical components. Initial results show that the annual mean PM$_{2.5}$ concentration was 98.7µg/m$^3$, which varied between 38 to 285µg/m$^3$. The water soluble inorganic fraction constituted only 9% of PM$_{2.5}$ with SO$_4^{2-}$, NH$_4^+$ and NO$_3^-$ being the dominant ions followed by potassium. Annual cycle shows higher concentration of PM$_{2.5}$ during winter season (October to January) and the lowest during monsoon. It is attributed to the enhanced production of aerosols and prevailing meteorological conditions. The PM$_{2.5}$/PM$_{10}$ ratio (0.86) coupled with the Hy SPLIT air-mass back trajectories indicated that PM$_{2.5}$ was dominated by fine particles, when the winds passed through the eastern azimuth, where many industries and major thermal power plants are located.

Introduction

Atmospheric aerosols have received much attention during the last two decades due to their potential influence on the global radiation budget and adverse impacts on human health and agriculture. Particulate matters with aerodynamic diameters less than 2.5µm (PM$_{2.5}$) have especially been found associated with increasing respiratory illness, carcinogens (Dockery and Pope, 1994), asthma (Anderson et al., 1992) and ultimately in increasing number of premature deaths. Many epidemic studies have linked airborne concentrations of PM$_{2.5}$ and PM$_{10}$ with a variety of health problems, including the morbidity as well as mortality (Wang et al., 2003). The high levels of PM$_{2.5}$ have also been associated with amenity problems such as visibility degradation associated with haze (Milne et al., 1982).

Aerosols, which are a complex mixture of elemental carbon, organic carbon, ammonium, nitrates, sulphates, mineral dust, trace elements, water vapour etc., have been examined for their composition and sources in urban and rural environments (e.g. Chow et al., 1994). Water-soluble fraction of atmospheric aerosol of hygroscopic nature, contains many important compounds, and can change size, composition, number-density and lifetime of aerosols (Novakov and Penner, 1993; IPCC, 1995; USEPA, 1996; Jacobson et al., 2000). Also, water-soluble aerosols increase the solubility of toxic organic compounds by acting as surface active reagents and therefore increasing their toxicity to human health apart from altering their optical properties. Results show that, on an average, about 185 to 483x10$^6$ ton
global aerosol load is emitted from anthropogenic activities per year, such as transportation, stationary combustion, industrial processes, solid waste disposal and other miscellaneous sources (Bridgman, 1990). Most of these sources are concentrated in the Northern Hemisphere, especially the areas downwind of the industrial sites, slash-and-burn agriculture regions, and overgrazed grasslands. Due to its characteristic economic, industrial and demographic nature, the atmospheric pollutants from the Indian region have different chemical compositions than the aerosols found in the developed parts of the globe. Some studies in Asian countries including India have even reported the organic, inorganic fractions, heavy metals and other chemical characterisation of the atmospheric aerosols (Khemani et al., 1982, 1985; Smirnov et al., 1998, Zhang and Friedlander, 2000, Kumar et al., 2003; Sharma and Shaily, 2005; Patel et al., 2006, Rengarajan and Sarin, 2006; and Rengarajan et al., 2007).

In a city like Delhi, with over 15 million inhabitants contributing towards the anthropogenic aerosols, coupled with the desert dust aerosols from the northwestern region (Singh et al., 2006), a very high level of ambient particulate loadings is expected. An earlier study on air pollution and health in India from the World Bank (1995) shows that small particulate emission (PM$_{10}$) and gases (like SO$_2$) are responsible for 95% of health damage (Khillare et al., 2004). A Delhi based non-government organization (NGO) has estimated an increase in premature deaths from 40,351 in 1991-92 to 51,779 in 1995 due to air related pollutions, out of which Delhi alone contributed to 7491 and 9859 during the same period suggesting one death per hour, on an average, due to the polluted air in the capital of India (Agarwal, 1992). A recent study conducted by Pande (1999) revealed that Delhi’s air pollution is responsible for over 40% of the emergency hospital admissions with respiratory and heart problems. In the present study, we have studied the chemistry of PM$_{2.5}$ over Delhi using some discrete data samples. Particularly, the water–soluble chemical components have been studied in details.

**Site description and aerosol collection**

Delhi (28°35’N; 77°12’E, 218 m above msl) experiences a severe weather swing between different seasons: from hot and humid weather in summer to cold and dry weather during winter. The prevailing wind throughout the year is easterly, northerly and northwesterly, and it is strongest during summer. Apart from such swings of weather during the annual cycle, the entire northern part of India, especially the Indo-Gangetic Plain, experiences a thick foggy weather during winter and show low boundary layer height. During such conditions, pollutants could not be dispersed or mix with free troposphere. The impact of such conditions is discernible as poor visibility and high levels of pollutants in this region. During the pre-summer and summer season, dust storm events also affect the climate of Delhi (Singh et al., 2006).

The sampling of aerosols for this study was carried out at about 15m above the ground level, on the rooftop of a building situated in the thoroughly urbanized central...
part of Delhi. To be mentioned here that the sampling area is primarily a residential area, and no large pollutant source exists nearby which could have influenced the sampling site directly. Sampling location is given on the road map of Delhi in Fig.1.

Sampling of PM$_{2.5}$ was done by using Fine Particulate Sampler (APM 550; Envirotech Instruments Pvt. Ltd, design approved by USEPA). The initial flow rate was kept at about 1 m$^3$/hr which subsequently reduced when the filters were loaded with fine particles. A 47mm quartz micro-filter (Whatman, UK) was used for sampling. The sampling duration for each sample was ~6 hrs [1100hrs to 1700hrs] and reflects the characteristics of the air pollution during the daytime. Total 14 samples of PM$_{2.5}$ were collected from January to December 2005. Table-1 shows the considered dates in this study, which are fairly distributed during various seasons.

**Table-1: Dates of Observation**

<table>
<thead>
<tr>
<th>Sample Number</th>
<th>Dates</th>
</tr>
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<tbody>
<tr>
<td>1</td>
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</tr>
<tr>
<td>2</td>
<td>10.01.05</td>
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<tr>
<td>3</td>
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<td>4</td>
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<td>5</td>
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<tr>
<td>6</td>
<td>18.05.05</td>
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<td>8</td>
<td>16.06.05</td>
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<td>12</td>
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<td>13</td>
<td>22.12.05</td>
</tr>
<tr>
<td>14</td>
<td>27.12.05</td>
</tr>
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</table>
For the chemical analysis, quartz micro-fibber filters were extracted with ultrapure water via ultrasonication and analyzed for water soluble ions. The ions, viz., organic (formate and oxalate), inorganic anions (chloride, nitrate and sulfate) and various cations (sodium, ammonium, potassium, calcium and magnesium) are obtained using Ion Chromatograph (IC). For quality assurance, calibrations of the IC were performed before and after the analysis. Field blanks were also obtained at regular intervals for improvement of the final results. These field blank filter papers are weighed and analyzed for the various chemical species. Values obtained by these field blanks were used for the correction of the final results.

Results and Discussion

Variation in the concentration of ionic mass and gravitational mass of PM$_{2.5}$:

Fig. 2 shows the daily variation in the mass concentration and measured ionic mass along with standards of PM$_{2.5}$. The average loading of PM$_{2.5}$ was found to be 98.71 µg/m$^3$, which is approximately two and half times higher than the Indian National Ambient Air Quality Standard (NAAQS; 40 µg/m$^3$) and almost six times higher than the United State Environmental Protection agency (USEPA) standard (i.e. 15 µg/m$^3$). The average loading of PM$_{2.5}$ showed highest deposition during winter (285 µg/m$^3$) and lowest during monsoon (38 µg/m$^3$) season. The high concentration of PM$_{2.5}$ during winter is expected to be caused by the combined effect of the elevated emissions from fossil fuel and coal burning and the prevailing meteorological conditions such as low temperature, wind speed, and mixing height. The lower inversion layer caused by low mixing height limits the dilution and dispersion of fine pollutants during winter season (Chatterjee et al., 2007).

**Fig.2:** Daily variation in the mass concentration and ionic mass along with the Indian and US standard level of urban PM$_{2.5}$

Relation between gravimetrical PM$_{2.5}$ mass concentration and the summation of ionic mass of analyzed water soluble inorganic chemical constituents was studied and found that only 9% of PM$_{2.5}$ was made up by water soluble inorganic fraction. There are presumably the negative discrepancies due to the abundance of
elemental carbon, organic matter, un-dissolved mineral dust, trace metals, and water etc. Similar negative discrepancy was also found by Hueglin et al., (2005) for an urban side in Switzerland. The mass contributions for ionic mass (30%) were organic matter, black carbon (48%), mineral dust, trace metals (7%) and unknown and calculated water (15%).

The relative percentage contribution of each ionic constituent to the total ionic mass concentration was calculated and found that the contribution of secondary component, \( \text{SO}_4^{2-} \), was 51% (4.74µg/m\(^3\)) to the total ion mass, while the anthropogenic acidic components, which are in fine fraction, contributed approximately 75%. \( \text{NH}_4^+ \) and \( \text{NO}_3^- \) were the second and third highest water soluble components followed by potassium and oxalate. The contribution of alkaline inorganic components (\( \text{Ca}^{2+}, \text{Mg}^{2+}, \text{Na}^+, \text{K}^+ \), and \( \text{NH}_4^+ \)) was approximately 25% to the total ionic mass. The calcium ion, which is mostly a soil generated components in the atmosphere, accounted for only ~2% of the total ion mass for PM\(_{2.5}\). This is obvious in this size range as it is generally available in atmosphere in the coarse mode fraction. Similarly, the contribution of magnesium (1%) was also negligible in PM\(_{2.5}\).

**Ionic variability:**

The ionic abundance in PM\(_{2.5}\) (µg/m\(^3\)), on an average, showed the general trend for inorganic ions as \( \text{SO}_4^{2-} > \text{NH}_4^+ > \text{NO}_3^- > \text{K}^+ > \text{Cl}^- > \text{Na}^+ > \text{Ca}^{2+} > \text{Mg}^{2+} \) whereas for the organic species as oxalate > formate. The variation of measured ionic species of PM\(_{2.5}\) on different days during the study period is shown in Fig. 3. Variation in the mass concentration of chloride and sodium were observed to be significant. Being far from the ocean, non-sea salt ions are dominating the aerosol chemistry comprising about 91% of the total water soluble ions. Sulfate, ammonium, oxalate and potassium showed similarity in their variation. However, calcium showed the different variation and formate and nitrate showed the similar behavior. As obvious from Fig. 3, \( \text{SO}_4^{2-} \) and \( \text{NH}_4^+ \) formed the most abundant part of water soluble fraction of inorganic species in PM\(_{2.5}\). Together, \( \text{SO}_4^{2-} \) and \( \text{NH}_4^+ \) accounted for ~6% of overall PM\(_{2.5}\) mass, but they were ~62% of the PM\(_{2.5}\) water soluble inorganic ion mass. About 97% of the total sulfate was from non-sea salt fraction. The \( \text{SO}_4^{2-} \) was significantly correlated with \( \text{NH}_4^+ \) \((r=0.91, p<0.0001, \text{Fig. 4})\). The inclination and the low intercept of the regression line strongly suggested that \( \text{NH}_4^+ \) and the most of \( \text{SO}_4^{2-} \) were probably present as \((\text{NH}_4)_2\text{SO}_4\) particles. It indicates often sufficient ammonia in the ambient air of Delhi, to neutralize the sulphuric acid formed from oxidation of sulphur dioxide. The mean \( \text{NH}_4^+ \) concentration during the study period was 5.76µg/m\(^3\) and it further suggests that both these ions were originated from the local anthropogenic sources in Delhi. Whereas, some part of these ions might have also been incorporated in the ambiance of Delhi from the nearby industrial locations.
Fig. 3: Daily variation of measured ionic species of PM$_{2.5}$
In order to identify the transport patterns of air parcels for higher PM$_{2.5}$ loading that may be arriving from different source regions to the sampling site, five-day HYSPLIT back trajectories were analyzed for all the considered days. Norman et al. (2001) have studied air mass trajectories during the monsoon period and found that the transport of chemical species was influenced by the prevailing winds. The wind associated with the low pressure system originated in the Bay of Bengal and moving towards inland or the easterly wind near the foothills of Himalayas, associated with the monsoon trough could carry the pollutants from the Industrial belt located in the central part of India, eastern Uttar Pradesh and its neighborhood to the areas in downwind. Two days (on 16$^{th}$ and 22$^{nd}$ December 2005) were chosen to show the trajectory analysis since these days show higher concentration of PM$_{2.5}$. The obvious dates during winter seasons showed higher PM$_{2.5}$ mass, amongst the measured dates, due to prevailing meteorological concentrations, such as lower mixing layer height. Trajectories for the considered two days (Fig. 5a and 5b, respectively) at 500m, 1000m and 1500m, when PM$_{2.5}$ mass concentration found higher, were computed using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT, Version 4) model developed by National Oceanic and Air administration/Air Resources Laboratory (NOAA/ARL,USA) (Draxler and Hess, 1997). Air mass trajectory analysis showed that air parcels were impacted with the emissions from the surrounding industrial locations. As seen from the figure 5a, five days long air mass trajectories ending on December 16 were originated from west and north-west azimuth for all the height levels, which turned to be from the east azimuth in last phase. It was found that the concentration of PM$_{2.5}$ was the highest (285µg/m$^3$) on December 16. This indicates towards the impact of sources from farther locations situated at cross-country boundary as well as adjoining industrialized places, such as

![Fig. 4: Relation between Sulfate and Ammonium in PM$_{2.5}$](image-url)
Panipat (Haryana) and Ghaziabad (Uttar Pradesh) and other locations in the east of the sampling site. Similarly, air mass trajectories ending on December 22 at Delhi showed that winds were predominating easterlies with respect to Delhi, where the three thermal power plants (Rajghat, Indraprastha and Badarpur) are located. For this day, the PM$_{2.5}$ concentration was 137 µg/m$^3$. Chemical analysis showed that the concentrations of sulphate and ammonium were higher when wind blew from east azimuth as compared to those from the other directions and indicates the influence of emissions of precursor gases from sources located in the east.

Fig.5a: Air mass back trajectories at different heights for Delhi on December 16$^{th}$, 2005
Fig.5b: Air mass back trajectories at different heights for Delhi on December 22nd, 2005

Mass ratio between PM$_{2.5}$ and PM$_{10}$:

The ratio of PM$_{2.5}$ to PM$_{10}$ is used to determine the contribution of fine particulate to the respirable particulate matter with aerodynamic particle size less than 10µm. Fig.6 shows the ratio of PM$_{2.5}$ / PM$_{10}$ for different stations in India as well as important places around the globe. The data for PM$_{10}$ at Delhi was taken from the Central Pollution Control Board data bank. The mean PM$_{2.5}$ / PM$_{10}$ ratio was found to be 0.86, which is remarkably higher value. Such higher values are expected to be due to the high concentration of PM$_{2.5}$ in the atmosphere at Delhi region attributed mainly to the combustion of fossil fuel during winter. As mentioned in Section 5.1, the low level inversion is one of the important causes responsible for higher concentration of PM$_{2.5}$ during winter.
Chemical Characteristics of Water Soluble Components of Fine Particulate Matter, PM$_{2.5}$, at Delhi, India: Suresh Tiwari, Manoj K. Srivastava, and Deewan S. Bisht

Fig.6: The ratio of PM$_{2.5}$/PM$_{10}$ at Delhi and different stations in India and elsewhere

In the Indian region, however, the ratio values are low, in general, due to the dominance of PM$_{10}$, which are contributed by the wind-driven desert-dust. However, the PM$_{2.5}$ concentrations are mainly due to the combustion processes. The ratio values at Delhi are comparably higher than those reported of Raipur (Patel et al., 2006), and Kanpur (Sharma and Shaily, 2005) where enormous burning of fossil fuels by various industries as well as thermal power plants and various industries like tanneries are located, respectively. This higher value also indicates towards very high contribution of PM$_{2.5}$ particle formation at Delhi. Ratio values at Zurich (Hueglin et al., 2005), Beijing (Wang et al., 2007), and Taipei (Mao et al., 2007) were almost comparable with those observed at Delhi. This indicates more concentrations of fine size fraction in the ambiance of Delhi with respect to the total aerosol loadings and suggests more anthropogenic activities for the cause. At Beijing and Taipei, although the fine particle concentration was more as compared to Delhi, however, it is due to equally high concentration of coarse particle; that the ratio of PM$_{2.5}$ to PM$_{10}$ at these two cities was comparable to that observed at Delhi.

Factor Analysis:

In order to study the origin of these aerosols and the components dominating in it, factor analysis was undertaken. Factor loadings for each variable have been calculated by principal component analysis (Khare et al., 2004). In general, factor loadings greater than 0.5 are considered significant in source apportionment studies. The factor loadings with the variance accounted by the various factors for Eigen value >1 are presented in Table-2. The data is synthesized into three major factors, explaining about 77.3% of the total variance. The division of information into
different groups (common factors) can be interpreted as the variables listed within the same factor that might have the same origin.

**Table- 2:** Factor analysis of ionic mass concentration of water soluble components in PM$_{2.5}$

<table>
<thead>
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<th>Factor 1</th>
<th>Factor 2</th>
<th>Factor 3</th>
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<tbody>
<tr>
<td>Calcium</td>
<td>-</td>
<td>-</td>
<td>0.86</td>
</tr>
<tr>
<td>Chloride</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Formate</td>
<td>-</td>
<td>0.85</td>
<td>-</td>
</tr>
<tr>
<td>Potassium</td>
<td>0.75</td>
<td>-</td>
<td>0.59</td>
</tr>
<tr>
<td>Magnesium</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Sodium</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Ammonium</td>
<td>0.92</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Nitrate</td>
<td>0.51</td>
<td>0.81</td>
<td>-</td>
</tr>
<tr>
<td>Oxalate</td>
<td>0.91</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Sulphate</td>
<td>0.94</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Eigenvalue</td>
<td>4.3</td>
<td>2.2</td>
<td>1.3</td>
</tr>
<tr>
<td>% Variance</td>
<td>43.0</td>
<td>21.9</td>
<td>12.4</td>
</tr>
<tr>
<td>Cumulative</td>
<td>43.0</td>
<td>65.0</td>
<td>77.4</td>
</tr>
</tbody>
</table>

The first factor comprises approximately half of the total variance (43%) and consists of potassium, ammonium, nitrate, oxalate and sulfate. The high loading of these ions in this factor is associated with the influence of secondary pollutants in the atmosphere, from the industries such as from thermal power plant as well as from automobile emissions. Ammonia (NH$_3$) is the most abundant gaseous alkaline component in the atmosphere. Particulate ammonium (NH$_4^+$) found in the atmosphere originates from NH$_3$ and its mean concentration in PM$_{2.5}$ was found to be 1.01 µg/m$^3$. The neutralization of acidic species like SO$_4^{2-}$ and NO$_3^-$ by NH$_4^+$ is very critical from acid deposition point of view. The major sources for ammonia gas include animal farming, use of fertilizers and organic decomposition. Ammonia is not transported to very long distances as it is rapidly converted into NH$_4^+$ aerosols at a rate of 30% h$^{-1}$ (Asman and Van Jaarsveld, 1991). As per early study pointed out by Khemani *et al.* (1982) for this region, the cation accumulates in the fine particle size and correlates well with SO$_4^{2-}$, suggesting that NH$_4^+$ ion were present in the atmosphere in the form of salt such as (NH$_4$)$_2$SO$_4$.

The second factor showing 21.9% of the total variance also represents a ‘strong man-made activities factor’ and is dominated by NO$_3^-$ and formate. The high loadings of NO$_3^-$ in this factor suggest an influence of nitrogen dioxide emitted by incompletely combusted fossil fuels from automobiles/diesel generators/tractors, and biomass burning which are abundant in this area, where the oxidation the gas is reported.

The third factor is comparatively minor and accounts for about 12.4% of the total variance with high loadings for natural sources such as soil containing calcium and magnesium. These ions are frequently found in soil dust. The Thar Desert, located in the west with respect to the observation location, is the major source of these alkaline
soil derived components from where they are transported by wind. Recently, Rengarajan et al., (2007) have found the soil contribution of nearly one forth by weight in the total suspended particle with the dominant ions for another north Indian urban station, Hissar, being $\text{SO}_4^{2-}$, $\text{NO}_3^-$ and $\text{NH}_4^+$. 

**Fig.7:** Contribution of ionic sources in measured mass of PM$_{2.5}$

Fig. 7 shows the contribution of ionic sources in measured mass to the daily average divided on the basis of factor analysis. The contribution of anthropogenic species was dominant (81%) in the PM$_{2.5}$ ionic mass concentration. It is produced by human activities such as burning of coal, wood, kerosene oil, and plants leaves during winter period to protect from cold (Ali et al., 2004). Apart from this, coal is also major fuel which is used in industrial activities around National Capital Region of Delhi. The results from factor analysis indicate towards the role of man–made activities in the composition of fine size aerosols over Delhi whereas contribution of mineral dust and soil aerosols, which are generally in coarse size, is very meager in the formation of fine size aerosols.

**Conclusions**

In summary, the study related to the investigation of the characteristics of the major chemical species (PM$_{2.5}$) at Delhi indicated that $\text{SO}_4^{2-}$, which produced form man-made sources, contributed approximately 75%. The annual average mass concentrations of PM$_{2.5}$ was 98.71µg/m$^3$, which is approximately two and half times and six times higher than the Indian NAAQS and USEPA standards, respectively, at
urbanized region. It suggests that particle pollution in Delhi is rather severe, especially due to fine particles. Variations of these particles were regulated mainly by
the strengths of sources and the prevailing meteorological conditions. The ratio between PM$_{2.5}$ to PM$_{10}$ was found to be high (0.86) in comparison to various other locations in India and was comparable to those reported from other locations in the world, indicating the dominance of PM$_{2.5}$, especially during the winter period. The concentrations of SO$_4^{2-}$, NO$_3^-$ and NH$_4^+$ were significantly high in the total water soluble inorganic fraction during winter. Being far from the ocean, non-sea salt ions dominated the aerosol chemistry (about 91% of the total water soluble inorganic ions). Among the detected ions, SO$_4^{2-}$ was the major chemical component of PM$_{2.5}$ (4.74 µg/m$^3$, 51% to the total ions), while NH$_4^+$ and NO$_3^-$ were the second and third highest water soluble components followed by potassium. The mean (NH$_4$)$_2$SO$_4$ concentration during the study period was 5.76 µg/m$^3$ and suggests that secondary aerosols, such as SO$_4^{2-}$, NO$_3^-$ and NH$_4^+$ were locally generated in Delhi and only a minor contribution is made by transported pollutants from the nearby industrial locations. Using the air mass trajectory analysis, the study confirmed that air parcels during winter passed through the major industrial areas located in the east of sampling site in Delhi; and sometimes, from west/north west regions. It is speculated that more attention should be paid to the regional sources of secondary pollutants while formulating pollution mitigation programmes at Delhi because these species may play a critical role in the occurrence of health hazards like increase of mortality and different types of respiratory and pulmonary diseases.

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