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Research Paper

Chemical Characterization and Source Apportionment of Fine Particulate Matter ($PM_{2.5}$) at a Rural Site in North Central India

APARNA SATSANGI[1], AWNI AGARWAL[1], ANITA LAKHANI[1] and K MAHARAJ KUMARI[1,]*
[1]Department of Chemistry, Faculty of Science, Dayalbagh Educational Institute, Dayalbagh, Agra, India

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Characterization of $PM_{2.5}$ (fine particulate) matter was conducted at Sikandarpur (27.26 °N, 78.00 °E), a rural site in North Central India in summer (April 2015 to June 2015) and winter (December 2015 to February 2016) seasons. $PM_{2.5}$ mass and concentrations of major water-soluble inorganic ions (WSIIs: Cl^-, SO_4^{2-}, NO_3^-, Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}) were determined. $PM_{2.5}$ mass was categorized into 3 levels: <60, 60-100 and >100 $\mu g\ m^{-3}$ (average = $88.7 \pm 27.2\ \mu g\ m^{-3}$). Correlation analysis of secondary inorganic species (SO_4^{2-}, NO_3^- and NH_4^+) showed the existence of $(NH_4)_2SO_4$, NH_4NO_3 and NH_4Cl, which were more predominant during winter season when NH_4^+ concentrations were high. During winter, the concentrations of SIA (secondary inorganic aerosol) were influenced by local air masses. Clusters of backward air mass trajectories were simulated from HYSPLIT Model which revealed three clusters and originated from various directions and source regions. Clusters 1 and 3 showed the impact of air masses from local sources while cluster 2 was mainly influenced by long range transport. Positive matrix factorization (PMF) was employed for source apportionment. Three factors were isolated as potential sources: soil dust (42.3%), combustion (26.3%) and secondary aerosol formation (31.4%), respectively.

Keywords: $PM_{2.5}$; Water-Soluble Inorganic Ions; Secondary Inorganic Aerosols; Long Range Transport

Introduction

Aerosol particles are a mixture of chemical constituents that are directly released into the atmosphere from anthropogenic as well as natural resources or may be formed as secondary species from the primary emissions through chemical reactions. They are mostly composed of water-soluble inorganic ions, elements and carbonaceous species. In recent years, fine particles ($PM_{2.5}$) have been increasingly characterized and considered to play a vital role in climate change and visibility reduction (Habeebullah, 2016).

Epidemiologically, fine particles are considered to cause respiratory infections, lung cancer and cardiovascular diseases (Pope *et al*., 2002; Cao *et al*., 2017). Therefore, it is important to examine the chemical characteristics and determine the sources of fine particulate matter.

In India, Indo-Gangetic plain is a highly polluted and populated region, where high concentrations of pollutants are a combination of long range transport from distant places and local emissions. The enhancement in urbanization and industrialization that has taken place at a rapid rate, resulted in the deterioration of atmospheric air quality in the Indo-Gangetic Plain (Sharma *et al*., 2014). Although numerous studies report $PM_{2.5}$ chemical composition in urban areas of the region (Ram *et al*., 2011; Pachauri *et al*., 2013, Pant *et al*., 2015), there are very few measurements from rural areas (Kulshrestha *et al*., 2009; Srivastava *et al*., 2014) which form a vast part of this plain.

Complete chemical characterization of atmospheric particulate matter provides better understanding of atmospheric pollutants in rural areas where emission sources are limited. Hence, the present study was planned to characterize the chemical composition of atmospheric constituents and to carry out its source apportionment.

**Author for Correspondence: E-mail: maharajkumari.k@rediffmail.com*

Materials and Methods

The Study Area

The study was carried out at Sikandarpur Mustkil (27.26° N, 78.00° E, rural site), a village in the North Central region of India. It is situated in a densely populated and one of the most polluted state; Uttar Pradesh, which lies in the Indo-Gangetic plains of Northern India. This region lies in a semi-arid zone adjacent to the Thar Desert with subtropical type of climate. Two common seasons: 'summer' and 'winter' show extreme variation in temperature. Summers are very hot (May and June being the hottest months, with maximum temperature crossing 40°C) while during winters the weather is very cold (during December to February, temperature varies from 3° to 15°C). Winter season frequently experiences heavy fog which disturbs daily life and hinders visibility. It is the result of its unique topography which is characterized by adverse dispersion conditions, especially in winters when mixing layer depth is low and thermal inversion is common.

Sikandarpur Mustkil is spread over an area of 355 hectares. Most of its area is used for agricultural activities where mainly rice, wheat and seasonal vegetables are cultivated. Majority of the houses in this village have cattle yard where the inhabitants keep cows, buffaloes, goats. The people in this village cook food on indigenous cooking stoves (chulhas) using cowdung cakes and wood. It does not have large industries or heavy traffic around, but biomass burning emissions are frequent in winter season, as coal and wood burning are used commonly for domestic heating. The major road is about 2 kilometres from this village, which itself is flanked by large agricultural fields on both sides. It is, thus, away from the influence of urban activities. This was the main reason for its choice as the sampling site (representative of rural site). This village has few residential houses but does not have any industries within a diameter of about 10 kms. The traffic density is low as people residing mostly use bicycles and motorcycles as the means of transport. The location of sampling site has been shown in Fig. 1.

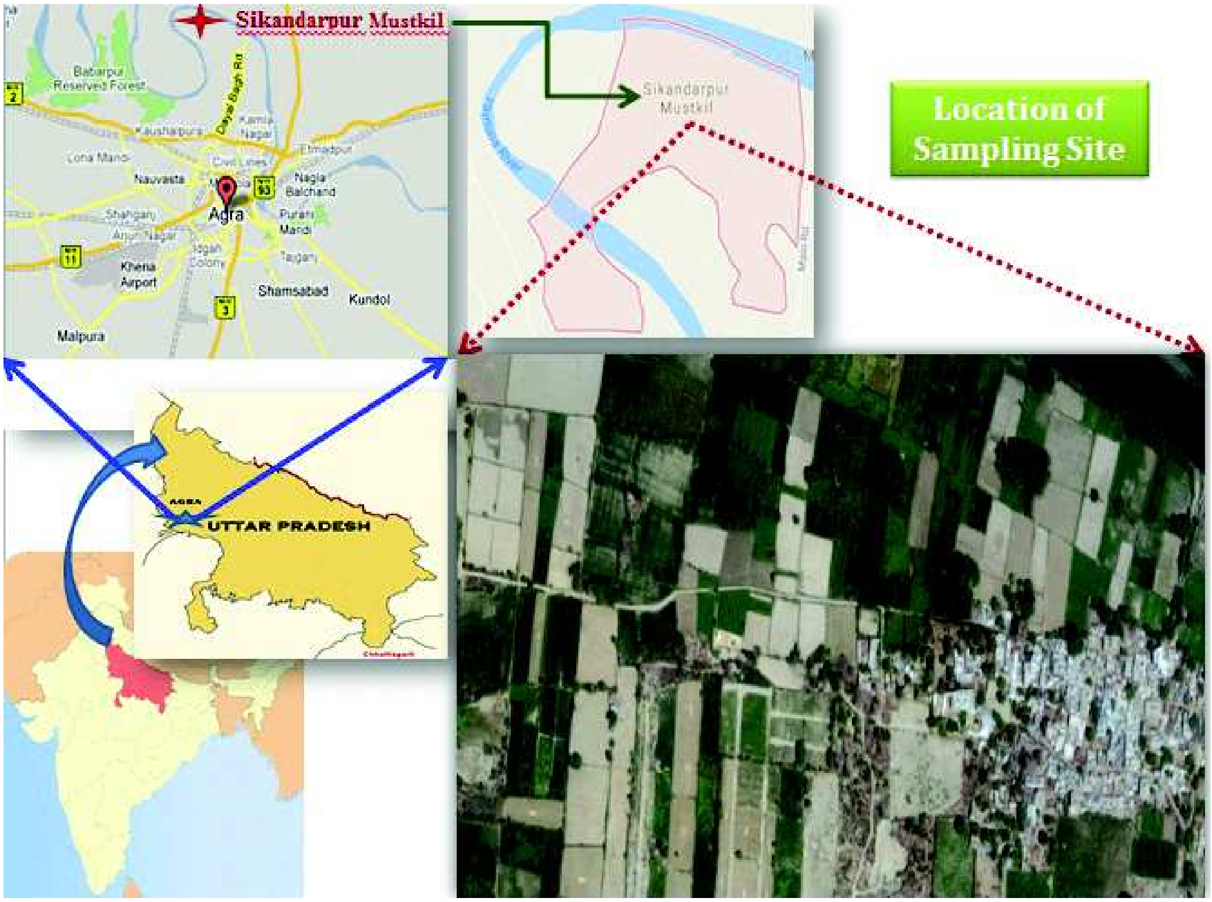


Fig. 1: Map showing position of Sampling Site

Sampling

$PM_{2.5}$ samples were collected during summer (April 2015 to June 2015) and winter (December 2015 to February 2016) seasons on the roof of a house in Sikandarpur at about 10 feet above the ground level. This single storied building was not surrounded by either tall buildings or trees. For sample collection, quartz fibre filters (QFF, 47 mm, Pallflex) were weighed and placed in Fine Particulate Sampler (Model: APM 550EL; Make: M/s Envirotech Instruments Private Limited) for 24 hours at a flow rate of 1.0 m^3 hr^{-1}. Prior to sampling, QFFs were placed for 3 hours in a muffle furnace at 900°C to remove volatile contaminants. The collected filters were stored in a refrigerator at approximately 4°C until extraction for chemical analysis.

$PM_{2.5}$ Mass Determination

$PM_{2.5}$ mass concentration was determined by the gravimetric analysis of exposed filters using the electronic microbalance. Each filter was stabilized at constant temperature (25±1°C) and relative humidity (50±1%) for 48 hours before and after sampling, and the net particle loading was obtained as $PM_{2.5}$ mass in µg m^{-3} by the subtraction of pre-sampling weights from the post-sampling weights. The typical uncertainty of gravimetric analysis is ±20 µg, which represents ±5% of the total $PM_{2.5}$ mass of the field samples.

Analysis of Water Soluble Inorganic Ions (WSIIs)

WSIIs were quantified by the extraction of one-half of each filter paper in deionised water by ultra-sonication for 45 minutes. The extract was then divided into two parts: one part was treated with chloroform for the analysis of anions while another part was preserved with 1% nitric acid (v/v) for the cation analysis (Kumar *et al.*, 2006).

The concentration of major ions (F^-, Cl^-, NO_3^-, SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}) was quantified by Ion Chromatography (Dionex ICS 1100). Filtered extract (filtered using 0.45 µm PTFE syringe filter, Pall Co. Ltd., USA) was used for detection of ions. Anions were detected by the system equipped with guard column (AG11), analytical column (IonPac AS11), and anion self-regenerating suppressor (ASRS 300, 4 mm) using eluent (6 mMNaOH). Analysis of cations was carried out by the system equipped with guard column (CG12A), analytical column (IonPac CS12A), and cation self-regenerating suppressor (CSRS 300, 4 mm) using eluent (20 mM methane sulphonic acid). The recoveries of ions were in the range of 80-90% and were calculated after spiking of target ions at different concentrations (Verma *et al.*, 2017).

Quality Assurance and Uncertainty

All the reported ion concentrations were corrected for field blanks. The filters that were brought to the field and placed in the samplers but no air was pumped through these were considered to be blank filters. These blank filters were analysed together with the collected samples, following similar analytical procedures. The obtained values were routinely subtracted from the sample concentrations.

Multi-ionic standards were used to test the linearity and to calibrate response of the instrument. Calibration was done using a range of 5 multi-ion standard concentration values. Spiking was done to check the ionic concentrations by using 1 ppm standard solution and to estimate uncertainties. This led to the peak area variation of less than 5%. Detection limit of each ion was estimated as three times the standard deviation of the blank (Miller and Miller, 2005). Quantified detection limits (ng m^{-3}) of the ionic species were as follows: F^-: 5.0, Cl^-: 17.2, NO_3^-: 9.5, SO_4^{2-}: 12.8, Na^+: 2.5, NH_4^+: 8.7, K^+: 16.8, Mg^{2+}: 3.2 and Ca^{2+}: 7.2.

Meteorological Parameters

Surface meteorological parameters like temperature, relative humidity, wind speed and direction were obtained from the website: *www.wunderground.com*. The variation of meteorological parameters (temperature, relative humidity and wind speed) has been shown in Fig. 2.

During winter season, the average temperature varied from 8.0 to 17.0°C, RH varied from 62.0% to 94.2% while the wind speed was low ranging from 0.6 to 1.0 m sec^{-1}. In summer season temperature was high ranging from 29.0 to 36.0°C, RH varied from 16.0% to 48.0% and wind speed ranged from 1.2 to 3.9 m sec^{-1}. Wind direction was mostly from west or northwest and at other times it was from

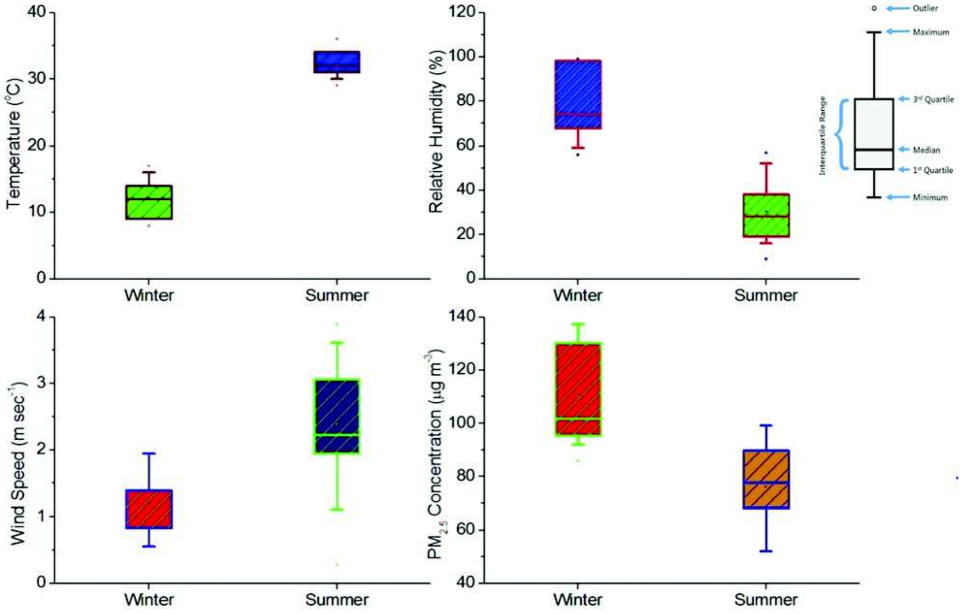


Fig. 2: Variation of $PM_{2.5}$ and Meteorological Conditions

southwest and east of India.

Positive Matrix Factorization (PMF) Technique

Multivariate receptor models are the mathematical approaches that are used to quantify the contribution of sources to samples based on the composition or fingerprints of the sources in environmental data. Positive Matrix Factorization (PMF) is also a multivariate factor analysis tool which is employed to identify the emission source contributions. EPA PMF 5.0 (Norris and Duvall, 2014) was used to analyse the data in the present study. A total of 10 chemical components were used as input parameters, including $PM_{2.5}$, major cations (Na^+, NH_4^+, K^+, Mg^{2+} and Ca^{2+}), and major anions (F^-, Cl^-, NO_3^-, SO_4^{2-}).

PMF decomposes a matrix of speciated data of samples into 2 matrices: 1) Factor contributions and 2) Factor profiles. This helps to understand the contribution of factors or sources which influence the selected data set (Paatero and Hopke, 2003; Paatero *et al*., 2014; Brown *et al*., 2015). The aim of PMF is to identify the number of factors, species profile of each factor, and the amount of mass contributed by each factor to each sample individually. The basis of data quality categorization is signal to noise ratio (S/N). The species which have $S/N \geq 2$ are categorized as having strong data quality, while the species which have S/N between 0.2 to 2 are categorized as having weak data quality and the species with S/N ratio below 0.2 are known to represent bad data quality. Weak and bad species are not likely to provide enough variability in concentration, and therefore contribute to the noise in the results. Such values must be excluded from further analysis, however, none of the species showed weak or bad data quality in the present study. More details related to EPA PMF version 5.0 are described in EPA PMF User Guide (2014). Several researches have used PMF for source apportionment of $PM_{2.5}$ (Gugamsetty *et al*., 2012; Cheng *et al*., 2014; Lu *et al*., 2016).

Transport and Dispersion Simulation

Air mass transport is studied from backward trajectories that are simulated to identify the sources and transport pathways of the chemical components in the atmospheric particulate matter. They may also provide information on the nature of particles present in the air. These air mass backward trajectories were simulated using HYSPLIT Model (version 4, revision February 2016; Draxler and Rolph, 2003) that are based on meteorological data from NOAA ARL (National Oceanic Administration Analysis Air Resources Lab) for the sampling location (27.26°N and 78.00°E). The trajectory was computed at 500 m for 72 hours at an interval of 6 hours (0000, 0600, 1200 and 1800 hours daily for the year 2015). 72 hours was chosen for backward trajectory as most of the secondary species have lifetime within this time period (Wojcik and Chang, 1997). The trajectories were developed at an altitude of 500 m above ground level for all to provide reasonable representation of the air mass and did not cause any unnecessary interruption of the air flow in the region.

The trajectories clusters provide the most accurate and best representation of air mass classifications according to the Euclidean distance between the trajectories. The clustered transport routes could be used to accurately identify the trajectory of air masses which originated from different source regions. This indicates the direction of the potential source of ionic species in $PM_{2.5}$. The air masses that had origin from nearby or local areas (Clusters 1 and 3) have been classified and termed as 'short ranged trajectories'. These trajectories are confined to the nearby areas of about 100 to 500 km only and originate from all directions. Such trajectories originate in all the seasons but are frequent during winter season. Contrarily, trajectories that originate from longer distances were said to have the influence of long range transport (Clusters 2) and were termed as 'long ranged trajectories'. These trajectories primarily originated from southwestern and western regions which are common in summer season. They may travel long distances of greater than 1000 km or even more before reaching the site under investigation. Percent contribution of water soluble ions in different clusters has been categorized to study its influence on ionic composition. Each ionic species showed characteristic variation depending upon the associated meteorological conditions and air mass transport.

Results and Discussion

$PM_{2.5}$ Mass Concentrations

$PM_{2.5}$ mass concentrations varied from 31.5 to 137.3 $\mu g\ m^{-3}$ with an average of 88.7±27.2 $\mu g\ m^{-3}$. In winter, $PM_{2.5}$ concentrations varied from 85.8 to 137.3 $\mu g\ m^{-3}$ with an average of 110.0±18.4 $\mu g\ m^{-3}$ while in summer, it varied from 52.0 to 99.0 $\mu g\ m^{-3}$ with an average of 76.1±15.8 $\mu g\ m^{-3}$. High $PM_{2.5}$ concentrations in winter may be associated with high RH, low mixing layer, low temperature and low wind speed. These stagnant conditions inhibit dispersion. Contrarily, high temperature and low relative humidity with greater mixing layer depth in summer favour dispersion leading to lower $PM_{2.5}$ mass.

These mass concentrations were higher than the NAAQS (National Ambient Air Quality Standards by CPCB, 2009) value of 60.0 $\mu g\ m^{-3}$ set as standard for $PM_{2.5}$ mass for rural and residential areas in India (CPCB, 2009). 67% of samples exceeded NAAQS standards of $PM_{2.5}$ concentrations in summer season while all $PM_{2.5}$ concentrations were above NAAQS standard in winter season.

$PM_{2.5}$ samples were classified into three categories on the basis of mass: (i) less than 60 $\mu g\ m^{-3}$, (ii) between 60 and 100 $\mu g\ m^{-3}$ and, (iii) greater than100 $\mu g\ m^{-3}$. Descriptive statistics (mean, median, 25% and 75% percentile, maximum, minimum and standard deviation) of $PM_{2.5}$ were recorded for the three categories. The average $PM_{2.5}$ mass concentrations in the three categories were 48.2±11.1 $\mu g\ m^{-3}$, 85.2±10.7 $\mu g\ m^{-3}$ and 122.4±11.1 $\mu g\ m^{-3}$, respectively. Mass of 20% of the aerosol samples in summer was less than 60 $\mu g\ m^{-3}$. 45% samples of winter season and 80% of summer samples contributed to medium ranged $PM_{2.5}$ mass while 55% samples of winter season had high mass (greater than 100 $\mu g\ m^{-3}$). Samples collected during dust have been excluded because they had very high PM mass. Air mass trajectories do not show the influence of SW winds but north western flow of winds as seen from clusters generated from HYSPLIT Model. High $PM_{2.5}$ concentrations in winter may be associated with high RH, low mixing layer depth, low temperature and low wind speed. These stagnant conditions inhibit dispersion. Contrarily, high temperature and low relative humidity with greater mixing layer depth in summer favours dispersion leading to lower $PM_{2.5}$ mass.

Composition of Water Soluble Inorganic Ions (WSIIs)

Water soluble inorganic ions (WSIIs) constitute a major part of atmospheric particulate matter; these ionic species (acidic and basic) react together and result in neutralization of atmospheric particulate matter. Ion balance (ratio of the sum of cation to anion; $\Sigma C/\Sigma A$; $\mu eq\ m^{-3}$) is an indicator of acidity or alkalinity of the atmosphere. The ratio ranged from 0.7 to 1.6 (1.2±0.2) and was greater than 1, indicating alkaline nature of aerosols as reported in earlier studies in this region (Kulshrestha *et al*., 1998; Parmar *et al*., 2001; Kumar *et al*., 2007; Satsangi *et al*., 2013). Ratio of greater than 1 indicates deficiency of anions, probably due to exclusion of CO_3^{2-}, HCO_3^- and organic acids from chemical analysis. The sum of WSIIs ranged from 15.8 to 50.5 μg m^{-3} (average: 33.6±8.0 μg m^{-3}). Among cations, NH_4^+ (6.8%) showed highest concentration followed by Ca^{2+} (4.6%), Na^+ ~ K^+ (4.4%) and Mg^{2+} (0.7%) while among anions, NO_3^-, SO_4^{2-} and Cl^- concentrations were similar (5.9, 5.4 and 5.3%). F^- (0.4%) was present in very low concentration.

High, Medium and Low Levels of $PM_{2.5}$ Mass Concentrations

Figure 3 depicts the average chemical composition of fine particulate aerosol for the entire study period categorized into 3 levels of $PM_{2.5}$ mass. A distinct variation is noticed in three levels. Comparison of PM levels based on *t* test suggests that all the three levels are distinct datasets and show statistically significant difference ($p < 0.001$, 95% significance level) when compared amongst each other. Air mass backward trajectory analysis shows that short ranged trajectories are local in origin and had high mass concentrations. while long ranged trajectories (associated with clean air) showed low concentrations. Meng *et al*. (2007) and Liang *et al*. (2016) have also reported similar results.The fraction with high $PM_{2.5}$ mass (> 100 ìg m^{-3}) showed dominance of SIA (52%) as compared to medium and low $PM_{2.5}$ mass levels (40% and 39%), respectively.

In order to show the impact of air masses, trajectory clusters were simulated using HYSPLIT model. The resultant trajectories resolved in three clusters (group of trajectories). Each cluster is representative of a specific pattern and direction of air mass transport. The contribution of each cluster in different seasons has been calculated (Fig. 4). The difference in composition may be attributed to the origin of trajectories from different source areas.

Cluster 1 represented maximum percent of the air masses predominantly from north and north-western direction accounting for 62% (403 trajectories) of the total trajectories formed. These trajectories travelled moderate distances (about 400

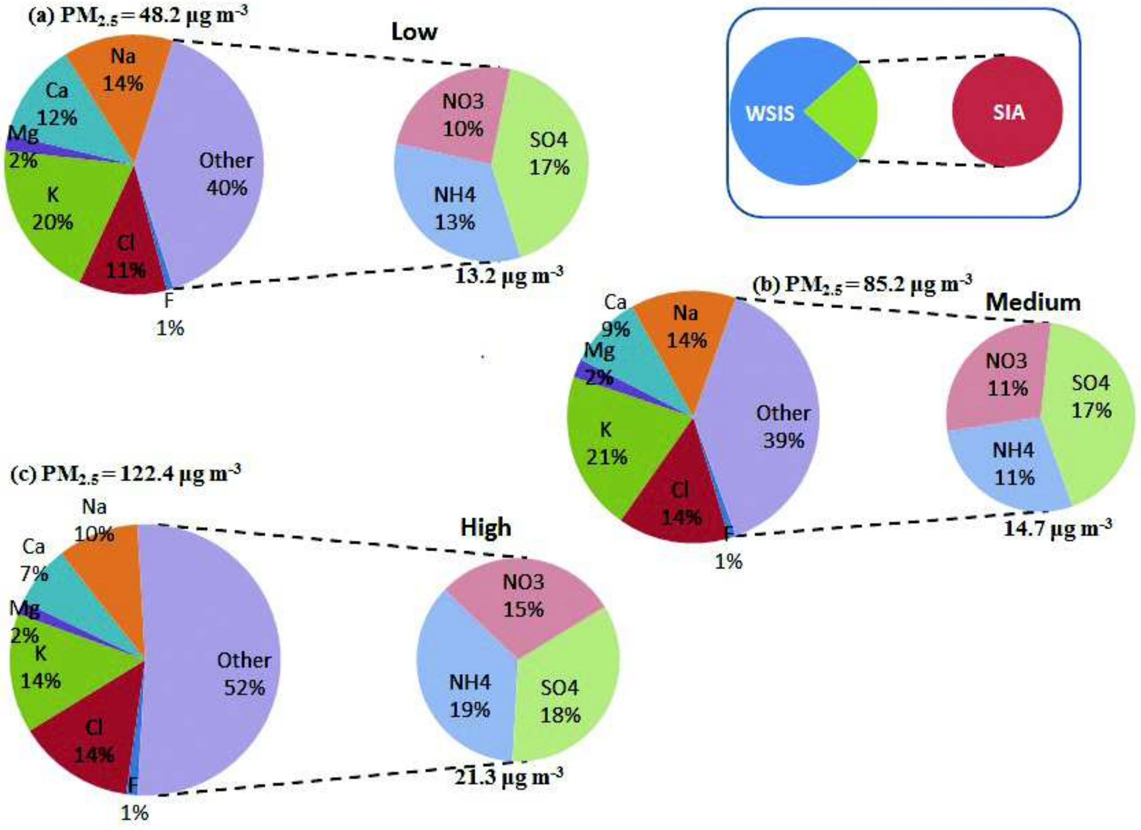


Fig. 3: WSIS in low, medium and high levels of $PM_{2.5}$

km) before reaching Sikandarpur. This region is the belt of agricultural activities. In this cluster, 45% trajectories were from summer season and were associated with moderate meteorological conditions. Wind speed is high in summer while atmospheric pressure is low favouring dispersion. 55% trajectories from winter also contributed to this cluster. Cluster 1 had medium (moderate) as well as high $PM_{2.5}$ mass concentrations due to their origin from short distances and association with localized emission sources. Cluster 2 originated from the western region of Indian subcontinent, contributing 15% (98 trajectories) of air mass back trajectories only during summer months. They originate from greater distances of more than 400 to 1100 kilometers. Few of these long ranged trajectories were associated with low RH (less than 25%) accompanied by low air pressure, high temperature and high wind speed. Trajectories of this cluster were associated with low $PM_{2.5}$ mass concentrations. Cluster 3 originated from longer distances of about 600 to 1000 kilometres contributing 23% (153 trajectories) of the total trajectories. This region has numerous coal-operated thermal power plants which leads to severe air pollution and is the belt of industrial and agricultural activities. The trajectories were mainly persistent in summer (39%) and winter (61%) seasons. This suggests dominance of local source emissions under the influence of stable meteorological parameters (low temperature, low wind speed (0.5 to 1.0 m sec^{-1}) and reduced mixing layer depth). The air pressure in the region was high during winter months but was moderate during summer months. High contribution from winter season is representative of high $PM_{2.5}$ mass concentration of this cluster and also because it is traverses the region of coal fired thermal power stations contributing to medium and high $PM_{2.5}$ mass concentrations.

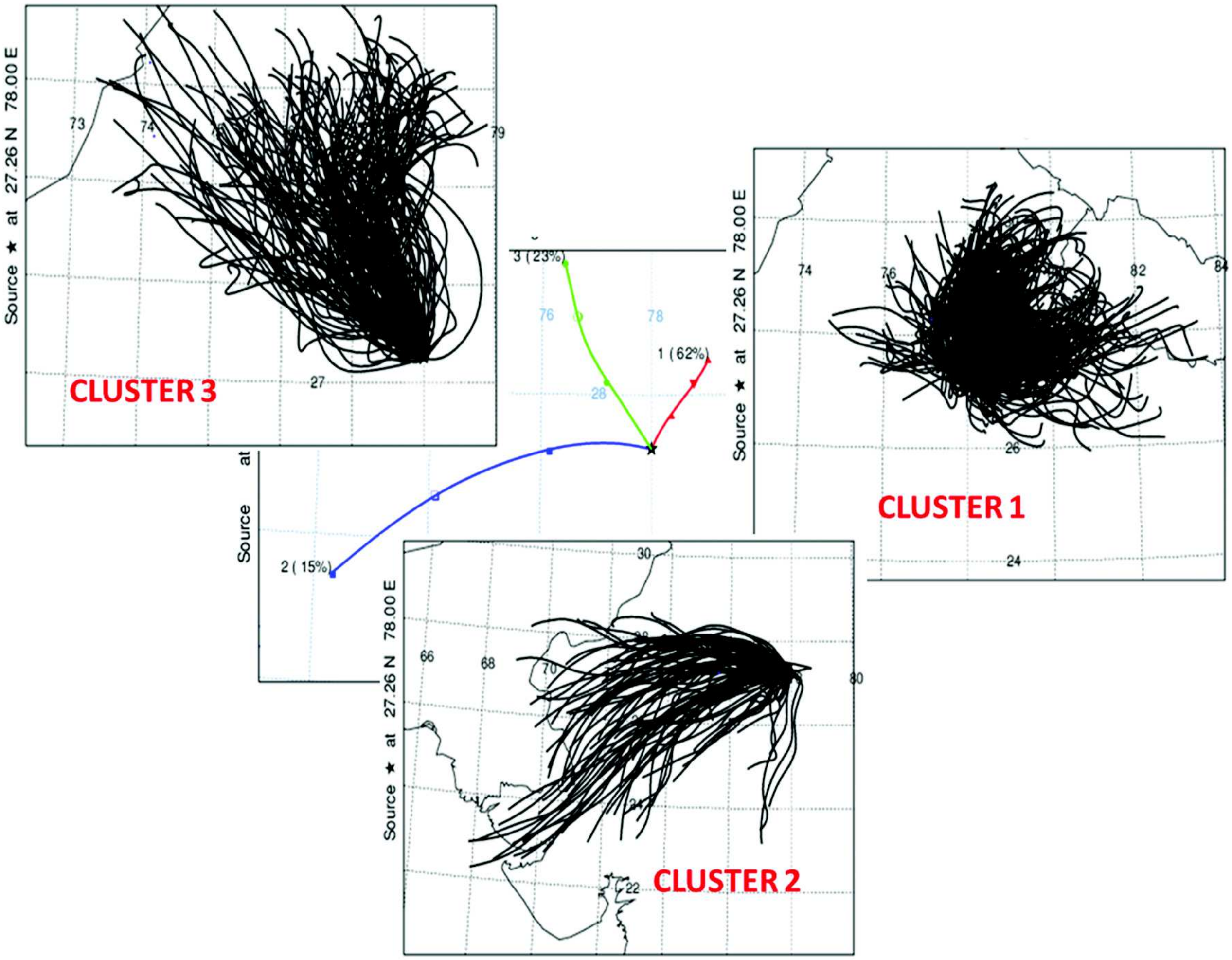


Fig. 4: Trajectory clusters and the associated trajectories

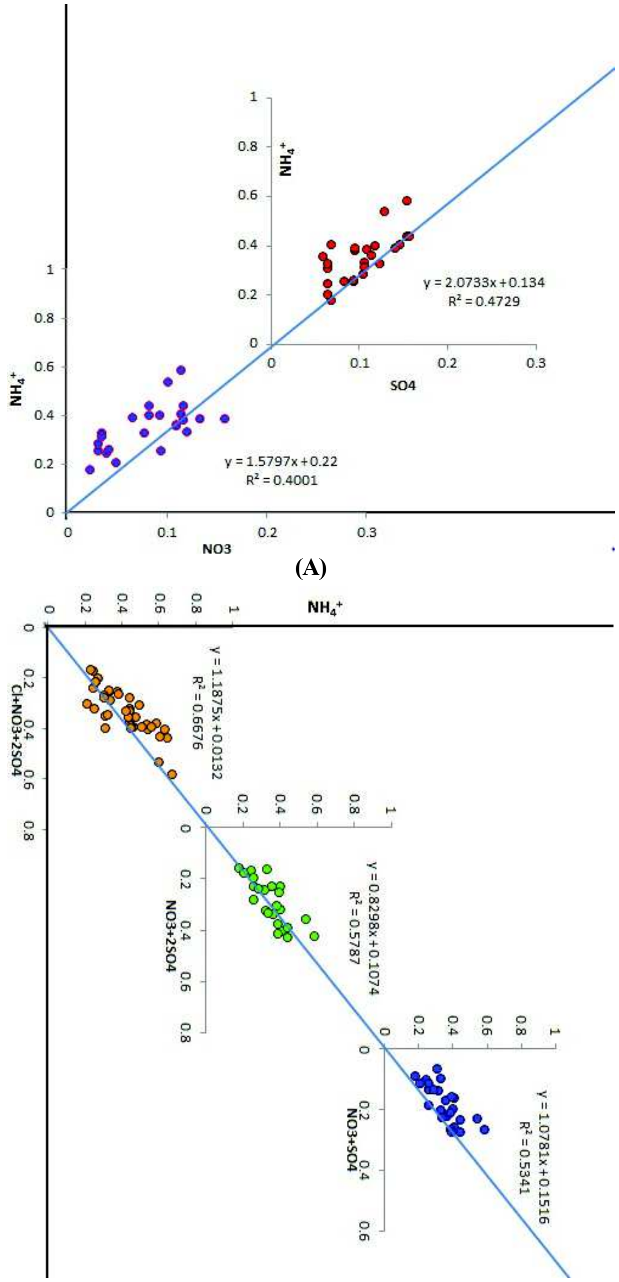
Correlation Analysis of Secondary Inorganic Aerosols

SIA (NO_3^-, SO_4^{2-} and NH_4^+) are the major constituents which form the vital hygroscopic species of fine mode particulate matter. Their chemistry depends on the formation of ammonium; mainly produced in the atmosphere by the reaction between gaseous NH_3 and acidic sulphate and nitrate particles (Yang *et al*., 2005; Pathak *et al*., 2009; Behera and Sharma, 2010).

In order to determine the relationship between NH_4^+ and the major acidic ions (Cl^-, NO_3^- and SO_4^{2-}), scatter plots of NH_4^+ with SO_4^{2-}, NO_3^-, $NO_3^- + SO_4^{2-}$, $NO_3^- + 2SO_4^{2-}$ and $Cl^- + NO_3^- + SO_4^{2-}$, were studied. The molar ratio of sulphate to ammonium for $(NH_4)_2SO_4$ and NH_4HSO_4 are 2.0 and 1.0, respectively. Figure 5A shows the regression between NH_4^+ and SO_4^{2-} indicating formation of $(NH_4)_2SO_4$. This is further confirmed by sulphate to ammonium slope of 2.2 which suggests high ammonium concentrations and complete neutralization of sulphate by ammonium (r=0.75, p=0.01) to form $(NH_4)_2SO_4$ in atmospheric particulate matter (Wang *et al*., 2006, Meng *et al*., 2016).

$(NH_4)_2SO_4$ is more stable (DRH, deliquescence relative humidity, is 79.9%), and is therefore, formed preferentially over NH_4NO_3 (DRH = 61.8%) (Seinfeld and Pandis, 2006). The formation of NH_4NO_3 takes place at a slower rate in the atmosphere. After NH_4^+ has completely neutralized SO_4^{2-}, excess NH_4^+ present in the atmosphere is available to combine with NO_3^-. Nitrate also shows a good correlation with ammonium (r=0.63, p=0.01) with a slope of greater than 1 (1.6), suggesting the formation of NH_4NO_3 in the atmospheric particulate matter (Yang *et al*., 2012; Cao *et al*., 2017). The data plot of NH_4 with (NO_3^- and SO_4^{2-}) also falls above 1:1 line indicating the neutralization of NO_3^- and SO_4^{2-} with NH_4^+, enough NH_4^+ was always present in aerosol samples to completely neutralize both NO_3^- and SO_4^{2-} and form $(NH_4)_2SO_4$ and further NH_4NO_3.

The average molar ratio of NH_4^+ to ($2SO_4^{2-} + NO_3^-$) was 1.3 confirming complete neutralization of SO_4^{2-} and NO_3^-. The regression between ($2[SO_4^{2-}] + [NO_3^-]$) and $[NH_4^+]$ also showed a strong linear correlation with a slope close to 1 (0.83) (Fig. 5b). This further indicates a completely neutralized system; and $(NH_4)_2SO_4$ and NH_4NO_3 may be the major compounds of SIA which exist in the atmosphere.



(A)

(B)

Fig. 5: Scatter plots of NH^+_4 and major acidic ions (concentrations in µequivalents)

As NH_4^+ was present in excess, relation between NH_4^+ and ($Cl^- + NO_3^- + 2SO_4^{2-}$) signifies probable existence of NH_4Cl also. The scatter plot of NH_4^+ with ($Cl^- + NO_3^- + SO_4^{2-}$) indicates that these acidic species exist in the form of $(NH_4)_2SO_4$, NH_4NO_3 and NH_4Cl. Strong correlation coefficient

(0.82) found between NH_4^+ and $(2SO_4^{2-}+NO_3^-+Cl^-)$ further confirmed the existence of these salts in the rural atmosphere.

Positive Matrix Factorization (PMF) Source Apportionment

Positive Matrix Factorization (PMF) was employed to determine the sources of $PM_{2.5}$ and to estimate their potential contributions. After the comparison of plots of a number of sources and plots of Q values (Reff *et al*., 2007), three factors were chosen which best fit the measurement data. This factor identification also explains >95% of the variability in the data set, satisfying the criterion suggested by a PMF evaluation (Chen *et al*., 2010).

The ionic concentrations of all species were input into EPA PMF 5.0 model for the identification of their potential sources. Three factors were isolated, representing potential sources which were: soil dust, combustion and secondary aerosol formation. They contributed 42.3%, 26.3% and 31.4%, respectively. The factor fingerprints of the ionic concentrations are shown in Fig. 6.

Soil Dust (Factor 1): PMF analysis showed that Factor 1 included major crustal elements (Na, K, Mg and Ca) with high loadings of Ca^{2+} and Mg^{2+}, hence, this factor suggested contribution from soil dust.These elements are the main contributors of airborne soil and road dust (Gugamsetty *et al*., 2012). Sharma *et al*. (2016) has also reported the association of Ca concentrations in $PM_{2.5}$ with its resuspension from agricultural fields or bare soils by local winds. Hence, soil dust was assigned as a source for Factor 1.

Combustion (Factor 2): Combustion activities are characterized by high loadings of K^+ and Cl^-. He *et al*., 2001 also reported Cl^- to be usually derived from coal combustion, while Duan *et al*., 2004 suggested the origin of K^+ from biomass burning. These ionic species showed high contributions in Factor 2; hence, K^+ and Cl^- may have mainly originated from coal and biomass combustion.

Secondary Aerosol Formation (Factor 3): Secondary inorganic aerosols are formed as a result of the reaction and transformation between pollutants emitted from air. An abundance of NH_4^+, NO_3^- and SO_4^{2-} was observed in this factor. NO_3^- and SO_4^{2-} may have formed by the conversion of NO_2 and SO_2 in the atmosphere while NH_4^+ may have been contributed by human activities, nitrogen fertilizers used in agricultural fields and biomass burning (Yang *et al*., 2011; Zhang *et al*., 2012).

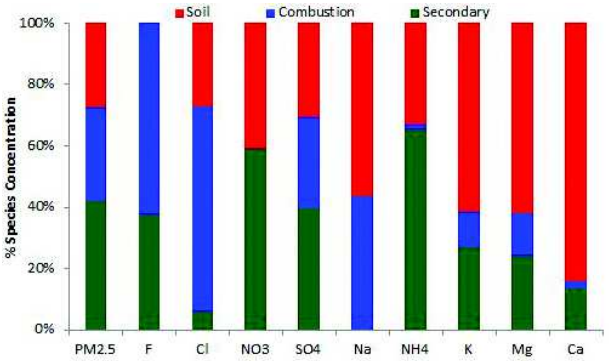


Fig. 6: Percent Contribution of each ionic species

Conclusions

$PM_{2.5}$ aerosol samples were collected at Sikandarpur, a rural site in North Central India and characterized to study the concentrations of major water soluble ionic species. $PM_{2.5}$ mass concentrations were high in winter (110.0±18.4 $\mu g\ m^{-3}$) as compared to summer (76.1±15.8 $\mu g\ m^{-3}$). Amongst ions, highest concentration was recorded for secondary inorganic ions leading to the formation of ammonium salts ($(NH_4)_2SO_4$, NH_4NO_3 and NH_4Cl) as a result of combined effect of localized activities, meteorological parameters and long range transport. Air mass cluster trajectories show that short range trajectories (Clusters 1 and 3) are associated with local emissions,while long range trajectories had transport of aerosol loadings from long range (Cluster 2). PMF analysis confirms major $PM_{2.5}$ sources to be soil dust (42.3%), combustion (26.3%) and secondary aerosol formation (31.4%), respectively.

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