A Study on the Understanding of Chemical Compositions of Deposited Fog Water over Central Indo-Gangetic Plain in India

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ABSTRACT

Fog preserves information about air pollution by pausing air movement and capturing chemical compositions of pollutants into fog droplets. The current study presents a chemical analysis of fog water collected in January 2021 at a rural site, Arthauli, located over the central Indo-Gangetic Plain (IGP) in India. Fog water collected on 11 foggy days during 1st to 21st January 2021, were analyzed for anions (SO$_4^{2-}$, Cl$^-$, NO$_2^-$, NO$_3^-$), and cations (NH$_4^+$, Na$^+$, Ca$^{2+}$, Mg$^{2+}$, K$^+$) using Ion Chromatography. pH of fog water ranged from 6.4 to 6.8 (Mean = 6.6 ± 0.1), depicting its alkaline nature. Total ionic composition (TIC) comprised majorly of NH$_4^+$ (35%), followed SO$_4^{2-}$ (23%), Ca$^{2+}$ (15%), Cl$^-$ (10%), NO$_3^-$ (8%), Mg$^{2+}$ (3%), and Na$^+$, K$^+$, NO$_2^-$ (2% each). Back-trajectories analysis indicated air movement from western IGP passing over major urban areas before reaching the sampling site. NH$_4^+$ had the highest neutralizing factor in fog samples (1.13), followed by Ca$^{2+}$ (0.48) and Mg$^{2+}$ (0.07). Analyses from enrichment factors, correlation coefficients, and principal components suggest that NH$_4^+$, NO$_3^-$, NO$_2^-$, SO$_4^{2-}$, and Cl$^-$ were from anthropogenic sources like agricultural activities, factories, vehicular emissions, brick kilns, thermal plants, fossil fuels combustion, and, biomass burning, while Ca$^{2+}$, K$^+$, and Mg$^{2+}$ were from the crustal source from agricultural fields.

Keywords: Fog, Chemistry, Indo-Gangetic Plain, Major ions, Neutralization

1 INTRODUCTION

It is well documented in literature that fog is a suspension of very small, usually microscopic water droplets in the air, reducing visibility at the Earth’s surface (Fuzzi et al., 1996; Gultepe et al., 2007; Nieberding et al., 2018). Fog droplets are formed on aerosol particles as relative humidity (RH) increases and ambient temperature decreases (Lakhani et al., 2007). Fog supports the processing of aerosols by providing a site of interaction for the chemical species present in the atmosphere, involved in new particle formations, their incorporation into fog droplets, scavenging of soluble trace gases (ammonia, nitric acid), and removal of particles from the atmosphere by droplet deposition (Pandis et al., 1990; Ali et al., 2004; Lakhani et al., 2007).

Deposition of fog water has an environmental and economic impact. Fog water has multiple effects on vegetation, health of living organisms, and historical buildings and transport due to visibility reduction (Fuzzi et al., 1996; Ali et al., 2004; Lakhani et al., 2007; Nieberding et al., 2018; Nath and Yadav, 2018). Fog water contains 5–20 times higher ion concentration than rainwater (Anderson et al., 1999; Beiderwieden et al., 2007). As a result, the investigation of fog chemistry demands a better understanding of potential deposition mechanisms. Several studies on fog chemistry have been carried out in different parts of the world (Waldman et al., 1982; Munger et al., 1983; Jacob et al., 1984; Igawa et al., 2001; Bridges et al., 2002; Błaś et al., 2010; Liu et al., 2015; van Pinxteren et al., 2016; Nieberding et al., 2018; Wang et al., 2019) and in urban areas of India like Delhi (Khemani et al., 1987; Ali et al., 2004; Nath and Yadav, 2018), Assam (Kulshrestha et al., 2005), and Agra (Lakhani et al., 2007).
One of the world’s most populated and polluted regions is the Indo-Gangetic Plain (IGP), located at the foothill of the Himalayas in the Indian sub-continental (Jat and Gurjar, 2021). In winter, the occurrence of fog is a widespread event over IGP (Saikh and Das, 2023; Singh et al., 2011; Sawaisarje et al., 2014; Ghude et al., 2017). Earlier studies reported a continental haze situated over IGP in winter, noticed by satellite observations (Di Girolamo et al., 2004; Ramanathan and Ramana, 2005; Badarinath et al., 2009; Kharol et al., 2011; Das et al., 2015a). This wintertime haze contained a significantly high amount of aerosols that were venting out from IGP to the Bay of Bengal, and the occurrence of fog caused a pause in the movement of this haze over IGP and preserves information regarding the formation and compositions by trapping all the atmospheric pollutants within the fog layer (Ganguly et al., 2006). The deposited fog water brings down all the pollutants within fog layer (Das et al., 2015b; Ramachandran et al., 2020; Ojha et al., 2020).

Investigations of chemical composition of deposited fog water provide an opportunity to improve our current understanding of the winter haze over IGP. Earlier studies on fog chemistry were mainly focused on the urban regions of IGP; thereby, a gap is created in our understanding regarding the downwind regions. The present study focuses on the chemical compositions of deposited fog water collected at Arthauli, a rural site situated on the way of winter haze movement over IGP. Having a weaker strength of emissions of anthropogenic aerosols, a rural site like Arthauli exhibits a relatively lower loading of aerosols. However, the movement of winter haze can cause enhancement of aerosol loading. Present investigation on fog water provides a new direction of fog chemistry on the role of occurrences of fog over rural site on the scavenging of aerosols transported from urban regions like Delhi and Kanpur. Our analysis reports on the fog water characteristics and possible sources of major ions present within it, which affects the overall nature of fog in the central IGP.

This paper is structured as follows: Section 2 describes the methodologies used. Subsequently, Section 3.1 presents the back-trajectory analysis of air-parcel over Arthauli. In Section 3.2, the characterization of fog chemistry for the major ions is done. Section 3.3 includes Subsections 3.3.1, 3.3.2, and 3.3.3, where source assessment of ions in the deposited fog water is analyzed with the help of enrichment factor, Principal Component Analysis (PCA), and correlation analysis, followed by conclusions in Section 4.

2 METHODOLOGY

2.1 Sampling Site
Deposited fog water was collected from 1st to 21st January 2021 at Arthauli (25.9535°N, 85.1050°E), a rural site situated over central IGP in India. The measurement site is selected well within a village named Arthauli under Vaishali district of Bihar state in India, as shown in Fig. 1(a). Vaishali is situated at the bank of Gandak (in the west) and the Ganga (in the south) rivers. In addition, the region is located between two high-altitude areas; the Himalayas in the north and the Satpura range in the south, making the measurement site a unique region on the movement of cold wind movement from west to east IGP in winter. The nearest industrial area is in the capital, Patna, about 60 km (southern direction) from the measurement site. A coal-based thermal power plant is at a distance of about 50 km (north-eastern direction), and a few brick chimneys are found nearby, as marked in Fig. 1(b). The sampling was carried out on the roof of a residential house at a height of about 10 m from the ground, and it was surrounded by the agricultural fields mainly covered with wheat crops in their early vegetative stage at the time of sampling when new leaves unfurl with a low rate of trans-evaporation. Therefore, there could be a negligible contribution to the chemical compositions of fog water from these crops. Being a rural site, the village has no major industries with fewer vehicular movements. A few constructive works were going on sparsely within the village.

2.2 Prevailing Meteorological Conditions
The central IGP experiences harsh winter due to the western disturbances (Ali et al., 2004). Diurnal variations of meteorological parameters (RH, temperature, wind speed, visibility, surface solar radiation, and planetary boundary layer height (PBLH)) during the study period are shown.
Fig. 1. Location of measurement site Arthauli (red star). The sampling site is situated between two hilly regions, the Himalaya range and, Vindhya and Satpura range, and at the bank of two rivers- Gandak and Ganga. Thermal plants, factories, and brick chimneys are also marked near the sampling site.

in Fig. 2. The data were retrieved from Indian Monsoon Data Assimilation and Analysis (IMDAA), a regional atmospheric reanalysis over the Indian subcontinent. IMDAA is a collaboration among Met Office (MO), U.K., National Centre for Medium Range Weather Forecasting (NCMRWF), India, and the Indian Meteorological Department (IMD), which is funded by National Monsoon Mission (NMM) project of the Ministry of Earth Sciences (MoES), Government of India. The horizontal grid resolution of this reanalysis model is 12 km (0.12° × 0.12°), and it also has 63 vertical levels, which reach to a height of approximately 40 km (Rani et al., 2021).

The shaded region in the figure indicates the period of fog water collections. The mean values of meteorological parameters like RH, temperature, wind speed, visibility, solar radiation, and PBLH during the sampling period are observed as 93 ± 4.2%, 14 ± 1.9°C, 4 ± 0.7 m s⁻¹, 16 ± 5 km, 115 ± 16 W m⁻², and 100 ± 50 m respectively. PBLH was noticed to be very low, indicating relatively lower ventilation with a weaker convective process favoring fog formation. Additionally, calm wind conditions also enhance the accumulation of water vapor in the atmosphere in a highly humid environment, which helps the fog droplets to be exposed for a more extended period in the atmosphere and deposit by absorbing significantly higher concentrations of air pollutants. It is worth to mention here that the visibility in Fig. 2 is over 10 km during foggy conditions because the data retrieved is for 12 km grid, however, collection of fog water was done in local conditions.

2.3 Sample Collection

Deposited fog water samples at the ground were collected between night 20:00 hours and morning 08:00 hours. During the campaign, 11 dense fog episodes occurred, and deposited fog water samples were collected in a significant amount of about 107 ± 70 mL. The fog water samples were collected using a sterile tray washed with double autoclaved water and placed 2 m above the rooftop, following the protocol mentioned in Kulshrestha et al. (2005). Care was taken to
Fig. 2. Diurnal variation of meteorological data at the measurement site (a) RH and temperature, (b) wind speed and visibility, and (c) surface solar radiation and planetary boundary layer height. The shaded regions present sampling periods of fog water collected at Arthauli from 20:00 hours at night to 08:00 hours in the morning.

avoid any contamination from the roof surface. During the sampling period, wind speed was calm. It is worth mentioning here that the calm, stagnant wind in a foggy atmosphere helps to settle down the fog droplets within the tray. A few millilitres of fog water were used to measure its pH immediately after collection. Collected samples were then transferred to sterilized polyethylene bottles and kept within ice gel packs to maintain low temperatures. After the campaign, samples were moved to the laboratory and analyzed within 15 days.

2.4 Sample Analysis

From each sample, 10 mL of deposited fog water was filtered with sterile filters of 0.22 µm pore size (MF-Millipore Mixed Cellulose Ester Membrane, Merck Life Science Private Limited, India). This filtrate was then divided into two parts of 5 mL; one part was used for the analysis of anions (SO$_4^{2-}$, Cl$^-$, NO$_2^-$, NO$_3^-$), and the other part was used for cation analysis (NH$_4^+$, Na$^+$, Ca$^{2+}$, Mg$^{2+}$, K$^+$). The major water-soluble ions were analyzed with ion chromatography (Eco IC, Metrohm, Herisau, Switzerland), which consists of an anion exchange column (Metrosep A Supp 5 250/4.0) and cation exchange column (Metrosep C6 150/4.0). Weak base eluent Na$_2$CO$_3$ (3.2 mM) and NaHCO$_3$ (1.0 mM) and a suppressor 0.2 N H$_2$SO$_4$ were used for anion detection. Weak acid eluent HNO$_3$ (1.7 mM) and dipicolinic acid (1.7 mM) were used for cation detection. Standard solutions were
prepared of three concentrations (5 ppm, 25 ppm, and 50 ppm) and used to quantify a calibration curve. The anion and cation analysis flow rates were kept at 0.7 mL min⁻¹ and 0.9 mL min⁻¹, respectively. The sample reproducibility was ±0.2 ppm (Fernandes et al., 2018).

2.5 Data Analysis
Data quality control was performed as mentioned in the literature (Rodhe and Granat, 1984; Safai et al., 2004; Liu et al., 2015). Spearman’s correlation between the sum of anions and the sum of cations was 0.99 (p < 0.001), suggesting credible data quality. The ratio of \( \sum \text{anions} / \sum \text{cations} \) was calculated using Eq. (1), as shown below

\[
\frac{\sum \text{anions}}{\sum \text{cations}} = \frac{\sum_{i=1}^{n} [\text{SO}_4^{2-} + \text{Cl}^- + \text{NO}_2^- + \text{NO}_3^-]}{\sum_{i=1}^{n} [\text{NH}_4^+ + \text{Na}^+ + \text{Ca}^{2+} + \text{Mg}^{2+} + \text{K}^+]}
\]

where \( n \) is the number of samples, and the unit of ion concentration is mEq L⁻¹. The ratio of total anions and total cations was 0.76, indicating consideration of most of the major ions influencing the fog chemistry; the remaining percentage could be due to bicarbonate (HCO₃⁻) and oxalate (C₂O₄²⁻) (Li et al., 2011; Liu et al., 2015; Kaskaoutis et al., 2022).

2.6 Non-Sea Salt Concentration
Non-sea salt concentration has been estimated to quantify the non-marine part of the compositions. The amount of non-sea salt fraction of ions (nssX) was calculated using the following Eq. (2) (Keene et al., 1986; Nieberding et al., 2018):

\[
nssX = [X]_{\text{sample}} - \left[ \frac{X}{\text{Na}^+} \right]_{\text{sea}} \times [\text{Na}^+]_{\text{sample}}
\]

where \([X]\) is the concentration of ions in the samples in mEq L⁻¹, \( X/\text{Na}^+ \) are the ratios of concentration in the sea (µEq X/µEq Na⁺), mentioned in Table 5.

2.7 Neutralization Factor (NF)
NF helps to understand the neutralization of the acidic components in the fog water by the cations (NH₄⁺, Mg²⁺, Ca²⁺). The major acid-forming anions are (SO₄²⁻ and NO₃⁻), which are neutralized by the cations. NF was calculated by using the following Eq. (3) (Saxena et al., 1996; Ali et al., 2004; Nath and Yadav, 2018):

\[
NF(X) = \left( \frac{[nssX^+]}{[\text{NO}_3^-] + [nssSO_4^{2-}]} \right)
\]

where ‘X’ is the cation of the fog sample for which neutralizing factor is to be calculated, and ‘nssX’ is the non-sea salt component of the cation calculated by using Eq. (2), and the unit of ion concentrations are in mEq L⁻¹. In this study, NF was estimated from the major ions present at our measurement site and compared with previous studies, as mentioned in Table 3.

2.8 Enrichment Factor
Enrichment of ions in the fog water samples was assessed by normalization of elemental concentrations of ions present within the samples against natural abundance (Hissler and Probst, 2006). EF is commonly used to examine the source contribution of major ions (Liu et al., 2015; Wang et al., 2019). The ratios of the composition of ions in sea and soil are mentioned in Table 5.
Na\(^+\) is commonly used as the tracer element of marine sources, and EF\(_{sea}\) of the ion was estimated using the following Eq. (4):

\[
\text{EF}_{sea} = \frac{\frac{X}{Na^+}_{fog}}{\frac{X}{Na^+}_{sea}}
\]

(4)

where EF\(_{sea}\) is the EF of an ion in fog relative to an ion in sea; \(X\) is ion in fog; \([X/Na^+]_{fog}\) is the ratio of composition in fog (mEq X/mEq Na\(^+\)); and \([X/Na^+]_{sea}\) is the ratio of composition in sea (\(\mu\)Eq X/\(\mu\)Eq Na\(^+\)) (Keene et al., 1986; Liu et al., 2015).

Similarly, EF\(_{soil}\) of an ion in fog relative to the ion in the soil is estimated by using Ca\(^{2+}\) as the tracer element of the continental source using the following Eq. (5):

\[
\text{EF}_{soil} = \frac{\frac{X}{Ca^{2+}}_{fog}}{\frac{X}{Ca^{2+}}_{soil}}
\]

(5)

where \(X\) is ion in fog; \([X/Ca^{2+}]_{fog}\) is the ratio of composition in fog (mEq X/mEq Ca\(^{2+}\)); and \([X/Ca^{2+}]_{soil}\) is the ratio of composition in soil (\(\mu\)Eq X/\(\mu\)Eq Ca\(^{2+}\)) (Taylor, 1964; Liu et al., 2015; Wang et al., 2019).

Fog samples are mainly influenced by marine, crustal, and anthropogenic sources, and their contributions can be estimated by using the following equations (Cao et al., 2009; Lu et al., 2011; Liu et al., 2015):

\[
\text{Sea Salt Fraction (S.S.F\%) } = \left(\frac{X}{Na^+}_{sea} \times \frac{X}{Na^+}_{fog}\right) \times 100
\]

(6)

\[
\text{Crustal Fraction (CF\%) } = \left(\frac{X}{Ca^{2+}}_{soil} \times \frac{X}{Ca^{2+}}_{fog}\right) \times 100
\]

(7)

\[
\text{Anthropogenic Fraction (A.P\%) } = 100(\%) - [\text{SSF\%} + \text{CF\%}]
\]

(8)

It is important to mention here that if SSF(\%) is greater than 100, the difference between 100 and CF(\%) is considered as SSF(\%), and for the identical case of CF(\%), recalculated CF(\%) is carried out following the similar method of SSF(\%). In this study, CF(\%) of K\(^+\) and Mg\(^{2+}\) is calculated with this phenomenon.

### 2.9 Computation of Back-trajectories

Three days of air parcel back-trajectory analysis was performed using the HySplit-4 (Hybrid-Single Particle Integrated Trajectory Model 4) to identify the possible source regions. Back-trajectories were computed from 1\(^{st}\) to 21\(^{st}\) January 2021 at an initial height 100 m above the ground over the sampling site and are shown in Fig. 3. The fire data were retrieved from NASA’s FIRMS (Fire Information for Resource Management System) (https://earthdata.nasa.gov/firms), which provides near real-time (NRT) fire locations obtained from images taken by Visible Infrared
Fig. 3. Three days air parcel backtrajectory at the measurement site (pink star). Fire data represents orange color as nominal confidence and red color as high confidence are also shown over the Indian subcontinent during the sampling period.

Imaging Radiometer Suite (VIIRS-375m) from the Suomi NPP and NOAA-20 platforms. Red dots depict fire with high confidence, and orange dots show the same with nominal confidence (Giglio et al., 2016).

3 RESULTS AND DISCUSSION

3.1 Back-trajectory Analysis

Three days of air parcel back-trajectories were observed to be originating mainly from the north-western part of IGP and blown over major urban areas like Delhi, Agra, and Kanpur before coming to the sampling site over central IGP (Fig. 3). In earlier studies, it has been reported that the winter haze containing a large number of human aerosols emitted from human activities is transported from northwest IGP (Venkataraman et al., 2005; Nair et al., 2007; Kaskaoutis et al., 2014; Das et al., 2015b; Ramachandran et al., 2020; Ojha et al., 2020). The present sampling site is located on the path of this long-range transportation of the winter haze. In addition, fire spots indicate high biomass burning along the path of this haze, as shown in Fig. 3.

3.2 Fog Chemistry Characterization

$\text{TIC} = 2.77 \pm 1.84 \text{ mEq L}^{-1}$ over the measurement site. $\text{NH}_4^+$ (35%) was present in the highest percentage of equivalent concentration, followed by $\text{SO}_4^{2-}$ (23%), $\text{Ca}^{2+}$ (15%), and $\text{Cl}^{-}$ (10%), and all the other ions were present below 10%, as shown in Fig. 4 and Table 1. The pH value reflects the acid-base balance in fog water. Conventionally, fog is considered acidic when pH is below 5 and alkaline when pH is 6 or above (Collett et al., 2002). In the present study, the pH of the deposited fog water ranged from 6.4 to 6.8, with a mean value of 6.6 ± 0.1, depicting its alkaline nature. A high amount of ammonium and calcium in deposited fog water raised the pH at Arthuali. A similar nature of fog water was also reported at Delhi and Agra in India, as mentioned
A comparison of ionic concentration in fog water at different sites across the world is presented in Table 2. It is important to note that the pH of fog water in Delhi has decreased over the years, inclining from an alkaline to acidic nature. The pH of fog water collected at Delhi was 7.2 in 1985 (Khemani et al., 1987), and then it decreased to 6.61 in 2000–2003 (Ali et al., 2004); furthermore, it declined to 5.38 in 2014–2015 (Nath and Yadav, 2018). In thirty years, a 35% decrease in the pH of fog water was noticed, shifting towards acidic nature over Delhi. Such a decrease in pH is due to increased sulfate and nitrate ions. Acidic fog events were reported over Mt. Lu, Southern China (Sun et al., 2015), NE Taiwan (Beiderwieden et al., 2007), and Daekwanreung, South Korea (Kim et al., 2006), which is due to the presence of a high concentration of $SO_4^{2-}$ and $NO_3^-$. These ions are present in the fog water from various anthropogenic sources like brick chimneys, factories and thermal plants with coal combustions, and vehicular emissions (Lakhani et al., 2007; Li et al., 2011). In the present study, these acid-forming anions ($SO_4^{2-}$ and $NO_3^-$) are present in fog water at Arthauli.
Table 2. Comparison of ionic concentration (mEq L⁻¹) in fog water at different sites.

<table>
<thead>
<tr>
<th>Sampling site</th>
<th>Sampling year</th>
<th>pH</th>
<th>Cl⁻</th>
<th>NO₂⁻</th>
<th>NO₃⁻</th>
<th>SO₄²⁻</th>
<th>Na⁺</th>
<th>K⁺</th>
<th>Ca²⁺</th>
<th>Mg²⁺</th>
<th>NH₄⁺</th>
<th>Reference</th>
</tr>
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<tr>
<td>Arthauli, India</td>
<td>2021</td>
<td>6.6</td>
<td>0.29</td>
<td>0.04</td>
<td>0.23</td>
<td>0.63</td>
<td>0.06</td>
<td>0.06</td>
<td>0.42</td>
<td>0.07</td>
<td>0.97</td>
<td>This study</td>
</tr>
<tr>
<td>New Delhi, India</td>
<td>2014–2015</td>
<td>5.38</td>
<td>7.81</td>
<td>0.45</td>
<td>4.55</td>
<td>11.73</td>
<td>0.78</td>
<td>0.80</td>
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</tr>
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<td>6.61</td>
<td>0.05</td>
<td>–</td>
<td>0.03</td>
<td>0.24</td>
<td>0.05</td>
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<td>0.12</td>
<td>0.02</td>
<td>0.31</td>
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<td>0.43</td>
<td>–</td>
<td>0.09</td>
<td>0.13</td>
<td>0.19</td>
<td>0.01</td>
<td>0.34</td>
<td>0.16</td>
<td>1.89</td>
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<td>–</td>
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<td>0.18</td>
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<td>0.34</td>
<td>0.15</td>
<td>1.70</td>
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<td>–</td>
<td>0.01</td>
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<td>0.04</td>
<td>0.03</td>
<td>0.07</td>
<td>0.02</td>
<td>0.17</td>
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<td>2010</td>
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<td>–</td>
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<td>–</td>
<td>0.18</td>
<td>0.34</td>
<td>0.02</td>
<td>0.01</td>
<td>0.06</td>
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<td>0.04</td>
<td>–</td>
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<td>0.01</td>
<td>0.03</td>
<td>0.02</td>
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<td>–</td>
<td>0.26</td>
<td>0.15</td>
<td>0.09</td>
<td>0.02</td>
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<td>–</td>
<td>0.07</td>
<td>0.19</td>
<td>0.01</td>
<td>0.03</td>
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<td>0.73</td>
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<td>0.01</td>
<td>–</td>
<td>0.08</td>
<td>0.20</td>
<td>0.00</td>
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<td>0.02</td>
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<td>2009</td>
<td>4.8–6</td>
<td>0.01</td>
<td>–</td>
<td>0.11</td>
<td>0.06</td>
<td>0.03</td>
<td>0.01</td>
<td>0.09</td>
<td>0.01</td>
<td>0.06</td>
<td>Huo et al. (2011)</td>
</tr>
</tbody>
</table>

Closely related with a correlation coefficient of 0.99 (p < 0.001), indicating they were simultaneously released from similar anthropogenic sources, pushing the pH of fog water to exhibit a more acidic nature.

In the present study, SO₄²⁻ (23%) was the second most abundant ion in fog water at the measurement site (Table 1). It is considered a major acidifier in fog (Kim et al., 2006; Wang et al., 2019; Aikawa et al., 2001). Therefore, the strength of neutralization of acid components in fog water by cations was found to be highest in NH₄⁺ (1.13), followed by Ca²⁺ (0.48) and Mg²⁺ (0.07). NH₄⁺ was the most abundant ion in all fog events in the current study. NH₄⁺ mainly comes from animal waste decomposition, use of mineral fertilizers, and biomass burning (Klimont, 2001; Nieberding et al., 2018). The sampling site had a high concentration of NH₄⁺, indicating the above-mentioned possible sources. It is worth mentioning that animal rearing is a common practice of residents in the village, and biomass burning frequently occurred over central IGP; as it was winter season and temperature was very low during the sampling period, villagers also had the practice of setting fire to keep themselves warm (Nath and Yadav 2018) and Ali et al. (2004) have also reported NH₄⁺ as a cation with highest neutralization strength over Delhi (Table 3).

The neutralization was supported by a strong positive correlation between NH₄⁺ vs. NO₃⁻ (R² = 0.85, p < 0.001) and NH₄⁺ vs. SO₄²⁻ (R² = 0.84, p < 0.01), as shown in Fig. 5. In addition, molar ratio of NH₄⁺/SO₄²⁻ and NH₄⁺/NO₃⁻ are 1.5 and 4.2 respectively, indicating the increased formation of (NH₄)₂SO₄ and NH₄NO₃ (Rastogi et al., 2014; Kaskaoutis et al., 2022; Sheoran et al., 2022). CaSO₄ and Ca(NO₃)₂ could also be possible neutralization product in fog as Ca²⁺ and SO₄²⁻ (R² = 0.60, p < 0.05) and Ca²⁺ and NO₃⁻ (R² = 0.60, p < 0.05) are positively correlated. Both ammonium and calcium...
Table 3. Comparison of Neutralization Factor of ions in fog water with previous studies.

<table>
<thead>
<tr>
<th>Sampling site</th>
<th>NF</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Arthauli, India</td>
<td>1.13</td>
<td>This study</td>
</tr>
<tr>
<td>New Delhi, India</td>
<td>0.73</td>
<td>Nath and Yadav (2018)</td>
</tr>
<tr>
<td>Delhi, India</td>
<td>1.45</td>
<td>Ali et al. (2004)</td>
</tr>
<tr>
<td>Delhi, India*</td>
<td>9.36</td>
<td>Khemani et al. (1987)</td>
</tr>
<tr>
<td>Agra, India*</td>
<td>1.75</td>
<td>Lakhani et al. (2007)</td>
</tr>
<tr>
<td>Assam, India*</td>
<td>2.82</td>
<td>Kulshrestha et al. (2005)</td>
</tr>
<tr>
<td>Poland*</td>
<td>0.63</td>
<td>Blas et al. (2010)</td>
</tr>
<tr>
<td>Germany*</td>
<td>1.07</td>
<td>van Pinxteren et al. (2016)</td>
</tr>
<tr>
<td>China*</td>
<td>0.91</td>
<td>Nieberding et al. (2018)</td>
</tr>
</tbody>
</table>

Fig. 5. Spearman’s correlation of major ions in fog water at Arthauli, where, * - p < 0.05, ** - p < 0.01, *** - p < 0.001, and × - no significance. Correlation coefficient values are indicated by size and color.

Table 3 shows the comparison of Neutralization Factor (NF) of ions in fog water with previous studies. NH₄⁺ play an important role in neutralizing acidity caused by sulfate and nitrate. NF of NH₄⁺ is highest at different locations in India (Table 3). This neutralization by NH₄⁺ was due to increased production in winter. Ammonium has volatile characteristics, and it is entrapped within a low boundary layer and absorbed by fog water. Ca²⁺ was the third most abundant ion in fog water and contributed 15% to the TIC in our study. It is mainly emitted from construction sites. As few construction activities were going on sparsely near the village, it can lead to an increase in the concentration of Ca²⁺ in the deposited fog samples at the measurement site.
### Table 4. Sea salt and non-sea salt composition of fog water (mEq L⁻¹).

<table>
<thead>
<tr>
<th></th>
<th>Maximum</th>
<th>Mean</th>
<th>Minimum</th>
<th>St. Deviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cl⁻</td>
<td>0.20</td>
<td>0.07</td>
<td>0.02</td>
<td>0.05</td>
</tr>
<tr>
<td>Cl⁻(nss)</td>
<td>0.35</td>
<td>0.21</td>
<td>0.10</td>
<td>0.09</td>
</tr>
<tr>
<td>K⁺</td>
<td>0.00</td>
<td>0.00</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>K⁺(nss)</td>
<td>0.14</td>
<td>0.06</td>
<td>0.01</td>
<td>0.04</td>
</tr>
<tr>
<td>Ca²⁺</td>
<td>0.01</td>
<td>0.00</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>Ca²⁺(nss)</td>
<td>0.89</td>
<td>0.41</td>
<td>0.12</td>
<td>0.25</td>
</tr>
<tr>
<td>Mg²⁺</td>
<td>0.04</td>
<td>0.01</td>
<td>0.00</td>
<td>0.01</td>
</tr>
<tr>
<td>Mg²⁺(nss)</td>
<td>0.14</td>
<td>0.05</td>
<td>0.02</td>
<td>0.04</td>
</tr>
<tr>
<td>SO₄²⁻</td>
<td>2.39</td>
<td>0.63</td>
<td>0.11</td>
<td>0.70</td>
</tr>
</tbody>
</table>

Na⁺ and Cl⁻ are considered of marine origin (Nieberding et al., 2018). In addition, fossil fuel combustion, biomass burning, and waste incineration are also considered as other sources of Cl⁻ (Li et al., 2011; Wang et al., 2011; Kaskaoutis et al., 2022). At our sampling site, the Cl⁻/Na⁺ ratio was found to be 4.8 ± 2.8, much higher than its ratio in sea (1.16) (Keene et al., 1986). Therefore, one can conclude that Cl⁻ has an additional source other than marine like biomass burning, previously reported in a high biomass burning laden site in Greece during winter (Kaskaoutis et al., 2022). A similar result with a higher ratio [Cl⁻/Na⁺ (9.97)] was also reported in fog water collected in Delhi in 2014–2015 (Nath and Yadav, 2018). K⁺ (2%) also makes a minor contribution to TIC; it is mainly emitted from biomass burning (Li et al., 2011); however, a few literatures reported its emission from crustal sources (Lakhani et al., 2007; Ali et al., 2004; Nath and Yadav, 2018). Mg²⁺ originated from marine and crustal sources like road dust and long-range dust transport (Nieberding et al., 2018). It also contributed 3% to the TIC in our samples.

The concentration of non-sea salt components of ions in fog water samples is mentioned in Table 4. SO₄²⁻ (0.63 mEq L⁻¹) had the highest mean non-sea salt composition, followed by Ca²⁺ (0.41 mEq L⁻¹), Cl⁻ (0.21 mEq L⁻¹), Mg²⁺ (0.05 mEq L⁻¹), and K⁺ (0.06 mEq L⁻¹). The ratios of nssSO₄²⁻/SO₄²⁻, nss-Ca²⁺/Ca²⁺, nss-Cl⁻/Cl⁻, nss-Mg²⁺/Mg²⁺, and nss-K⁺/K⁺ are 97.5%, 99.2%, 74.3%, 77.1%, and 96.7% respectively, which indicates least contribution from marine sources in overall fog deposition at our measurement site. Moreover, back-trajectory analysis (Fig. 3) suggested that the direction of air movement was from the western sector. The measurement site is situated at the central IGP, which is highly influenced by continental sources like factories, vehicular emissions, biomass burning, etc. As Arthauli has a significantly less number of such sources, it can be easily concluded that these aerosol compositions are transported along with winter haze from urban regions like Delhi and Kanpur, which is scavenged by fog droplets over measurement site.

### 3.3 Source Assessment of Ions in Fog Water

#### 3.3.1 Enrichment factor

A higher (lower) than 1 EF value indicates enrichment (dilution) of the ions in the environment relative to the reference source composition, as in Table 5. In the present study, Cl⁻, NO₃⁻, SO₄²⁻, K⁺, Ca²⁺, Mg²⁺, and NH₄⁺ have higher EFsea values suggesting they were enriched compared to the sea. On the other hand, EFsoil of Na⁺, K⁺ and Mg²⁺ had a relatively lower EF value than soil reference source, indicating dilutions from crustal source composition. Cl⁻, NO₃⁻, SO₄²⁻, and NH₄⁺ were also enriched relative to a soil reference source. The possible sources for the enrichment of these ions during winter are increased agricultural crop residue burning, solid waste, and biofuel burning, including lower boundary layer height, which traps the pollutants near the ground (Bisht et al., 2015; Dumka et al., 2017). High concentrations of inorganic ions impact visibility degradation and form extreme atmospheric conditions like fog or haze, usually observed over polluted sites like Delhi (Dumka et al., 2017) and China (Cao et al., 2012).

The enrichment factor in percentage of equivalent concentration for anthropogenic (AF), crustal (CF), and sea salt (SSF) is shown in Fig. 6. Na⁺ and Ca²⁺ are considered tracers of marine and crustal sources, respectively. NH₄⁺, NO₃⁻, SO₄²⁻, and Cl⁻ were mainly from anthropogenic sources.
Table 5. Enrichment factors relative to seawater and soil for fog water collected over Arthauli.

<table>
<thead>
<tr>
<th>Ion</th>
<th>EF_{sea}</th>
<th>EF_{soil}</th>
<th>([X/Na^+]_{fog})</th>
<th>([X/Ca^{2+}]_{fog})</th>
<th>([X/Na^+]_{sea})</th>
<th>([X/Ca^{2+}]_{soil})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cl(^-)</td>
<td>4.6</td>
<td>275.0</td>
<td>5.3</td>
<td>0.9</td>
<td>1.16</td>
<td>0.00</td>
</tr>
<tr>
<td>NO_3(^-)</td>
<td>467510</td>
<td>298.7</td>
<td>4.7</td>
<td>0.6</td>
<td>0.00</td>
<td>0.02</td>
</tr>
<tr>
<td>SO_4^{2-}</td>
<td>106.3</td>
<td>86.4</td>
<td>12.9</td>
<td>1.6</td>
<td>0.12</td>
<td>0.00</td>
</tr>
<tr>
<td>Na(^+)</td>
<td>1.0</td>
<td>0.3</td>
<td>1.0</td>
<td>0.2</td>
<td>1.00</td>
<td>0.57</td>
</tr>
<tr>
<td>K(^+)</td>
<td>44.2</td>
<td>0.3</td>
<td>1.0</td>
<td>0.1</td>
<td>0.02</td>
<td>0.50</td>
</tr>
<tr>
<td>Ca(^{2+})</td>
<td>156.0</td>
<td>1.0</td>
<td>6.9</td>
<td>1.0</td>
<td>0.04</td>
<td>1.00</td>
</tr>
<tr>
<td>Mg(^{2+})</td>
<td>5.0</td>
<td>0.3</td>
<td>1.1</td>
<td>0.2</td>
<td>0.22</td>
<td>0.56</td>
</tr>
<tr>
<td>NH_4(^+)</td>
<td>199444</td>
<td>4694</td>
<td>19.9</td>
<td>2.8</td>
<td>0.00</td>
<td>0.00</td>
</tr>
</tbody>
</table>

[X] - major ions; [X/Na\(^+\)]_{sea} - ratio of ions in sea (Keene et al., 1986); [X/Ca\(^{2+}\)]_{soil} - ratio of ions in soil (Taylor, 1964).

Fig. 6. Source contribution in fog water calculated through enrichment factor using major ions concentration (mEq L\(^{-1}\)).

with AF values of 99.9%, 99.6%, 97.9%, and 77.9% respectively. K\(^+\) and Mg\(^{2+}\) were found to be from crustal sources with CF values of 97.7% and 80.2%. SSF values of Cl\(^-\) and Mg\(^{2+}\) were 21.8% and 19.9% indicating some marine source contribution. Similarly, the SSF value of K\(^+\) (2.3%) and SO_4^{2-} (0.9%) shows that they have very low contributions from marine sources.

3.3.2 Principal component analysis (PCA)

PCA is widely used in atmospheric studies as a multivariate statistical process to estimate the strength and the relative contribution of natural and anthropogenic sources based on the chemical compositions of samples (Liu et al., 2015; Wang et al., 2019; Cao et al., 2009; Balasubramanian et al., 2001). In the present study, PCA has been applied using SPSS (IBM SPSS) to determine the important source factors. Varimax rotated PCA with Kaiser Normalization was used to characterize fog water samples into five factors, as mentioned in Table 6, which explains 99% of the variance in the data set. The first factor had a high variance value of 53% for Cl\(^-\), NO_3\(^-\), SO_4^{2-}, and NH_4\(^+\), suggesting common anthropogenic sources like factories, thermal plants, agricultural activities, vehicular emissions, and biomass burning (Jaiprakash et al., 2017; Sheoran et al., 2022).

The second factor is dominated by Mg\(^{2+}\), Ca\(^{2+}\), and K\(^+\), with a variance value of 29%, indicating major contributions from the crustal source (Rajput et al., 2018; Sheoran et al., 2021). The third factor had a variance of 12% for Cl\(^-\) and Na\(^+\), indicating its contributions from a marine source. In addition, a positive correlation was observed between Na\(^+\) and Cl\(^-\) \((R^2 = 0.58, p = 0.059)\); however, it was not significant, thus indicating additional sources for Cl\(^-\) like biomass burning, supported by a strong correlation of Cl\(^-\) with K\(^+\) \((R^2 = 0.75, p < 0.01)\) (Kaskaoutis et al., 2022). PCA depicts concentration of Cl\(^-\) was derived from dual sources, i.e., anthropogenic (PC1) and marine (PC3).
Table 6. Varimax rotated principal component analysis with Kaiser Normalization of major ions in deposited fog water. Values above 0.5 are marked with bold text.

<table>
<thead>
<tr>
<th></th>
<th>PC1</th>
<th>PC2</th>
<th>PC3</th>
<th>PC4</th>
<th>PC5</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cl^-</td>
<td>0.59</td>
<td>0.34</td>
<td>0.61</td>
<td>0.34</td>
<td>0.15</td>
</tr>
<tr>
<td>NO_2^-</td>
<td>0.06</td>
<td>0.28</td>
<td>0.46</td>
<td>0.83</td>
<td>0.13</td>
</tr>
<tr>
<td>NO_3^-</td>
<td>0.97</td>
<td>0.18</td>
<td>0.16</td>
<td>0.05</td>
<td>0.02</td>
</tr>
<tr>
<td>SO_4^{2-}</td>
<td>0.98</td>
<td>0.16</td>
<td>0.91</td>
<td>0.27</td>
<td></td>
</tr>
<tr>
<td>Na^+</td>
<td>0.29</td>
<td>0.87</td>
<td>0.10</td>
<td>0.19</td>
<td>0.30</td>
</tr>
<tr>
<td>K^+</td>
<td>0.29</td>
<td>0.87</td>
<td>0.10</td>
<td>0.19</td>
<td>0.30</td>
</tr>
<tr>
<td>Ca^{2+}</td>
<td>0.13</td>
<td>0.94</td>
<td>0.19</td>
<td>1.7</td>
<td></td>
</tr>
<tr>
<td>Mg^{2+}</td>
<td>0.51</td>
<td>0.96</td>
<td>0.21</td>
<td>0.7</td>
<td></td>
</tr>
<tr>
<td>NH_4^+</td>
<td>0.98</td>
<td>0.01</td>
<td>0.07</td>
<td>0.99</td>
<td></td>
</tr>
<tr>
<td>Variance (%)</td>
<td>53</td>
<td>29</td>
<td>13</td>
<td>3</td>
<td>1</td>
</tr>
<tr>
<td>Cumulative (%)</td>
<td>53</td>
<td>82</td>
<td>95</td>
<td>98</td>
<td>99</td>
</tr>
</tbody>
</table>

The fourth factor had a variance of 3% for NO_2^- indicating another anthropogenic source like fossil fuels combustion and waste incineration. It is worth mentioning here that the villagers burned significant biomass near the sampling site to keep themselves warm. The fifth factor (PC5) did not contribute much to the factor analysis. This result is broadly consistent with source attribution concluded from back-trajectories analysis.

3.3.3 Correlation analysis

In this study, Spearman’s correlation analysis was performed to observe the relationship among various ions in the fog water samples, as shown in Fig. 5. A strong correlation coefficient value between NH_4^+ and Cl^- ($R^2 = 0.80$, $p < 0.01$) was observed, suggesting their emission from biomass burning, as NH_4Cl is used as a fertilizer in agricultural fields. NH_4Cl also dominated the chemical compositions during fog episodes in Delhi (Acharja et al., 2022). Ca^{2+}, Mg^{2+}, and K^+ show a high correlation with each other, and the observed correlation coefficient between Ca^{2+} and Mg^{2+} was 0.95 ($p < 0.001$), between Ca^{2+} and K^+, was 0.88 ($p < 0.001$), and, between Mg^{2+} and K^+ was 0.93 ($p < 0.001$). These high correlation coefficient values indicated that they have a common crustal source. Similar results showing Ca^{2+}, Mg^{2+}, and K^+ were coming from crustal sources have been observed in Agra (Lakhani et al., 2007) and Delhi (Ali et al., 2004; Nath and Yadav, 2018). K^+ also had significant correlations with SO_4^{2-} ($R^2 = 0.79$, $p < 0.01$), NO_3^- ($R^2 = 0.77$, $p < 0.01$), and Cl^- ($R^2 = 0.75$, $p < 0.01$), suggesting its emissions from brick kilns, fertilizers, thermal plants, and biomass burning (Choudhary et al., 2018; Kaskaoutis et al., 2022).

4 CONCLUSIONS

A campaign was conducted from 1–21 January 2021 at a rural site, Arthauli (25.9535°N, 85.1050°E), of central IGP to collect fog water samples by a deposition method. Eleven fog water samples were collected during the sampling period. West-to-east movement of winter haze was noticed over IGP, and the occurrences of fog over central IGP paused this haze movement. Therefore, fog droplets get exposed for a longer duration to absorb the major ions from the air. Due to the location of the sampling site as well as wind-blown from the western direction, the contribution of marine sources was significantly less.

The major findings of the present study are as follows:

i. The average pH value of the collected fog water was 6.6 ± 0.1, exhibiting alkaline in nature, which might be due to the dominance of cations (NH_4^+, Na^+, Ca^{2+}, Mg^{2+}, K^+) over anions (SO_4^{2-}, Cl^-, NO_3^-, NO_2^-).

ii. The order of abundance of major ions in fog water order was NH_4^+ > SO_4^{2-} > Ca^{2+} > Cl^- > NO_3^- > Mg^{2+} > Na^+ > K^+ > NO_2^- at Arthauli.

iii. The contribution of dominant ions NH_4^+ (35%), SO_4^{2-} (23%), Ca^{2+} (15%), and Cl^- (10%) to the total ionic species of the fog water samples was nearly 83%.
iv. Acidity neutralization was found to be maximum in NH$_4^+$ (1.13), followed by 0.48 of Ca$^{2+}$ and 0.07 of Mg$^{2+}$.

v. Back-trajectory analysis suggested long-range transportation of aerosols coming to central IGP from western IGP blowing over major cities like New Delhi, Agra, Kanpur, etc., indicating the possible source regions of these major ions in the fog water over Arthauli.

vi. PCA indicated four distinguishable types of sources and grouped Cl$^-$, NH$_4^+$, NO$_3^-$, and SO$_4^{2-}$ in the first group (anthropogenic), Ca$^{2+}$, K$^+$, and Mg$^{2+}$ in the second group (crustal), Cl$^-$ and Na$^+$ are in third group (marine), NO$_2^-$ is in the fourth group (other anthropogenic sources).

vii. Enrichment factor and correlation analysis indicated that NH$_4^+$, NO$_3^-$, SO$_4^{2-}$, and Cl$^-$ were from anthropogenic sources, including activities like agriculture, use of fertilizers, fossil fuels combustion, waste disposal, thermal plants, brick chimneys, factories, and biomass burning, while Ca$^{2+}$, K$^+$, and Mg$^{2+}$ were from crustal sources from agricultural fields.

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AUTHOR CONTRIBUTION

Shahina Raushan Saikh contributed to the investigation, methodology, writing original draft, data curation, and software. Md Abu Mushtaque helped by contributing to the analysis and writing the original draft, and Sanat Kumar Das helped with conceptualization, validation, resources, investigation, supervision, writing review and editing, and project administration.

REFERENCES


eastern United States, as sampled from three high-elevation sites along the Appalachian Mountains. Atmos. Environ. 33, 5105–5114. https://doi.org/10.1016/S1352-2310(99)00193-4


