

Dew water chemical composition and source characterization in the IGP outflow location (coastal Bhola, Bangladesh)

Mohammad Shohei^{1,2} · Hasina Akhter Simol³ · Elizabeth Reid⁴ · Jeffrey S. Reid⁴ · Abdus Salam¹

Received: 28 March 2017 / Accepted: 22 May 2017 / Published online: 6 June 2017
© Springer Science+Business Media Dordrecht 2017

Abstract Dew water samples were collected during the winter season (December, 2014 to January, 2015) at an outflow location from the Indo-Gangetic Plain (IGP) to the Bay of Bengal (Coastal Bhola, Bangladesh). Physical properties of the dew water, including pH and electrical conductivity (EC), were measured. The concentrations of water soluble ions (Cl^- , SO_4^{2-} , NO_3^- , HCO_3^- , Na^+ , K^+ , Ca^{2+} , and Mg^{2+}) and trace metals (Zn, Fe, Mn, Cu, Cr, Pd, and Ni) were also measured. Source characterization of the chemical species was done by correlation analysis, enrichment factor analysis, percent source contribution calculation, and air mass trajectory analysis. The average pH and EC of the collected dew water were 6.81 and $154.80 \mu\text{S cm}^{-1}$, respectively. The average concentration of SO_4^{2-} and NO_3^- was 264.10 and $222.20 \mu\text{eq L}^{-1}$, respectively. The concentrations of water soluble ions followed the sequence: $\text{Ca}^{2+} > \text{Cl}^- > \text{SO}_4^{2-} > \text{Na}^+ > \text{NO}_3^- > \text{Mg}^{2+} > \text{K}^+ > \text{HCO}_3^-$. The concentrations of trace metals ranged in order with $\text{Zn} > \text{Fe} > \text{Mn} > \text{Cu}$, while the concentrations of Cr, Pb, and Ni were below detection limit in dew water. Regression analysis showed significant correlations among sea, soil, and anthropogenic species. High enrichment factors of SO_4^{2-} , NO_3^- , Zn, Mn, and Cu indicates anthropogenic sources. Percent source contribution

of different species showed significant anthropogenic contribution for Cl^- (1.80%), SO_4^{2-} (83.5%), and NO_3^- (99.3%). Air mass trajectory analysis supported that the regional urban pollutions have significant influence on the dew water chemistry at the coastal Bhola, Bangladesh.

Keywords Dew water · Water soluble ions · Trace metals · Air mass trajectory · Enrichment factor

Introduction

The Indo-Gangetic Plain (IGP) encompasses 21% of the Indian subcontinent and hosts over 1/8th of the world's population. The rapidly expanding economy in the IGP results in a wide range of emissions, including those from fossil fuel and biomass burning, industries, transport, mining, urbanization, and agricultural activities. Winter in this region is generally characterized by high aerosol optical depth (AOD), formation of persistent fogs, regional haze, and dew formation (Nair et al. 2007). Aerosol particles in the IGP region include those transporting in from overland and over ocean and are aged with continental as well as pristine ocean aerosol particles (Niranjan et al. 2006; Eck et al. 2001). The high aerosol loads make a significant contribution to the dew water chemical composition. Importance of the dew water chemistry has been reported in recent years (Lekouch et al. 2010; Yadav and Kumar 2014; Wentworth et al. 2016). Dew formation depends on the structure as well as chemistry of the substrate surface where condensation occurs (Beysens 1995). Contamination in condensed dew water is occurred by dry deposition, wet deposition (large fog droplet), and dissolution of gases from the atmosphere. Dew water collection is challenging, with many techniques for dew water collection having been reported, e.g., on wooden block (Duvdevani 1947), glass plate

✉ Abdus Salam
asalam@gmail.com

¹ Department of Chemistry, Faculty of Science, University of Dhaka, Dhaka 1000, Bangladesh

² Department of Chemistry, University of Iowa, Iowa, IA 52242, USA

³ Centre for Advance Research in Science (CARS), University of Dhaka, Dhaka 1000, Bangladesh

⁴ Naval Research Laboratory, 7 Grace Hopper Ave., Stop 2, Monterey, CA 93943, USA

(Janssen and Römer 1991; Kidron 1998), nylon mesh (Zangvil 1996), aluminum sheet (Baier 1966), polyethylene sheet (Wagner et al. 1992), and Teflon sheet (Takeuchi et al. 2002).

Chemical composition of the dew water is strongly affected by the atmospheric condition of the sampling locations, e.g., temperature, humidity, dry deposition (Beysens et al. 2006; Takeuchi et al. 2003). Dew water is usually weakly acidic, having a pH close to 7, but can be more acidic (5.41–6.00) in urban locations. Concentrations of calcium and ammonium were found to be higher in previous studies (Table 1). Aerosol particles in the atmosphere act as condensation nuclei in the formation of dew and fog, making wet deposition a major pathway transporting pollutants from the atmosphere to the biosphere (Polkowska et al. 2008; Rubio et al. 2012; Lynam et al. 2015). Therefore, the chemical composition of dew water provides insight into local pollution and has been undertaken in most regions, including USA, Europe, Japan, China, etc. (Beysens et al. 2009; Polkowska et al. 2008; Xu et al. 2015). Unfortunately, there is very limited information on chemical composition of dew water in IGP region (Lakhani et al. 2012; Singh et al. 2006; Yadav and Kumar, 2014), even though winter time air pollution seriously impacts dew water chemical composition in IGP region. Severe air pollution situation in Bangladesh has already been reported (Salam et al. 2003a, 2008). For coastal Bhola, an IGP outflow location, chemical characterization of the aerosol particles was done, and relatively high concentrations of some chemical species were observed (Salam et al. 2003b). We decided to undertake a study of the dew water composition at this IGP outflow location in coastal Bhola, Bangladesh, and assess the impact of the aerosol particles on the dew water.

We collected dew water samples from December 2014 to January 2015 at this outflow location from the IGP into the Bay of Bengal. The objectives was to characterize the physicochemical properties of the dew water, to identify pollutant sources through apportionment studies, and to evaluate the influence of long range transported air pollutants on the dew water composition at this remote island location.

Experimental

Sampling location

Dew samples were collected at Bhola (22°10'01" N, 90°45'00" E), Bangladesh, a rural delta island in the Bay of Bengal (Fig. 1). This location rests in the outflow path for pollutants from the IGP region to the Bay of Bengal (Fig. 1), making it an important monitoring location. Bhola has an area of 1441 km² and a population primarily employed in agriculture and fishing. There are no major industries in Bhola Island, and no traffic was observed within 1 km of the sampling location.

Biomass burning for cooking is the primary source of local pollution.

Meteorology of coastal Bhola, Bangladesh

The climate in Bangladesh is characterized by high temperatures, excessive humidity, and distinctly marked seasonal variations in precipitation. Bangladesh can be divided into four seasons: pre-monsoon (March–May), monsoon (June–September), post-monsoon (October–November), and winter (December–February). The average meteorological conditions in coastal Bhola during different seasons have been reported by Salam et al. (2003b). The average temperature varies between 4 and 25 °C in the winter and 24 to 36 °C in the summer, with an average of 25.9 °C, and daily high temperatures generally observed in the afternoon. The highest relative humidity occurs in July (99%) and the lowest in December (36%), with light precipitation occurring in March and April, and average annual precipitation of 230 mm. Our dew water samples were collected during winter season (December, 2014–January, 2015) in Bhola, Bangladesh. The temperature ranged from 9 to 17 °C, no rain events were recorded, and air mass back trajectories (NOAA/ARL 2005) indicated transport was mostly from north to southeast during sampling period.

Sampling procedure and analysis

Dew water samples were collected nightly in a stainless steel bowl (Fig. 2) from 20 December 2014 to 30 January 2015 at the IGP outflow location in Bhola Island, Bangladesh. Our first attempts used a collector made of a poly(ethylene) sheet, but the collection efficiency was not satisfactory. Therefore, we modified our system to collect dew water using a stainless steel container and during our sampling period were able to collect ten dew water samples having adequate volume (>30.0 mL) to analyze all for parameters. On the remaining days, weather conditions did not support enough dew formation in our collector. The stainless steel container was cleaned daily with de-ionized water prior to nightly deployment, and field blanks were collected to minimize errors in analysis. The collection system was installed at the top of a 9.0-m tower, built in 2013 in collaboration with NASA AERONET and ONR Global, USA, for aerosol and precipitation studies. As there is generally no dew in daytime, our sampling period was 9 to 10 h nightly. Dew condensed on the inclined surface of the bowl and collected at the bottom, from which the samples were removed and stored daily around 6:00 a.m. to preclude evaporation due to sunlight. The collected dew water samples were filtered using Whatman 41 filter paper. Then, the samples were split, physical properties and ions were measured from one part (e.g., pH, conductivity, anions, cations, etc.), while the other part was acidified with ultra-pure nitric acid

Table 1 Comparison of dew water composition collected at different locations of the world

	Bangladesh	India ^{a,b}	China ^c	Japan ^d	Chile ^e	Jordan ^f	France ^g	USA ^h	Croatia ⁱ	Israel ^j	
	Coastal Bholra/ rural (winter)	Delhi/urban (Winter)	Agra/rural (winter and monsoon)	Changchun/ urban (summer and autumn)	Yokohama/ urban (2 years)	Santiago/ urban (4 years)	Amman/ urban (6 month)	Bordeaux/ urban (1 year)	Indiana/ urban (1 year)	Zadar/rural (3 years)	Negev/ desert (Fall)
pH	6.81 ± 0.28	6.78	6.80	6.72	5.41	6.70	5.88	6.82	6.37	6.7	7.31
EC	154.8 ± 62.4	271	NA	308	NA	256.2	29	NA	NA	195.6	510
NH ₄ ⁺	NA	1662.1	254.5	1100–1800	454	47.0	NA	34	94	50	120
Na ⁺	243.8 ± 126.7	106.8	191.5	65–160	128	157	124	3.9	7.8	573	874
K ⁺	135.6 ± 45.7	69.1	112.9	45–80	22.8	30.1	6.39	8.5	9	58.8	144
Ca ²⁺	693.3 ± 361.3	834.1	413.1	35–90	274	639	17.5	75	115	1730	2873
Mg ²⁺	205.1 ± 188.3	62.9	290.2	600–1000	66.3	230	28.8	16	11	234	390
Cl ⁻	291.2 ± 127.7	193.5	348.2	80–125	165	138	135	6.1	11	668	1076
NO ₃ ⁻	223.2 ± 131.9	131.0	120.6	140–200	92.4	45.20	8.06	25.5	38	11.5	790
SO ₄ ²⁻	264.1 ± 135.2	1381.9	359.5	1200–2000	271	464	52	16	66	80.7	1660
HCO ₃ ⁻	63.0 ± 22.3	NA	66.7	NA	40.7	NA	NA	146	88.6	45.1	842

All units are in $\mu\text{eq L}^{-1}$, except pH, and EC ($\mu\text{S cm}^{-1}$)

NA not analyzed

^a Singh et al. (2006)

^b Yadav and Kumar (2014)

^c Xu et al. (2015)

^d Okochi et al. (1996)

^e Rubio et al. (2002)

^f Jiries (2001)

^g Beysens et al. (2006)

^h Wagner et al. (1992)

ⁱ Beysens et al. (2009)

^j Kidron and Starinsky (2012)



Fig. 1 Map of Bangladesh and Coastal Bhola (marked by red dots). Red circle indicates the sampling location (Source: Google map)

(1% V/V) and stored separately for trace metal analysis. Samples were stored in the refrigerator with sealed container to avoid any chemical transformation. The field blanks were collected using a clean stainless steel collector having the same size as that used for the real sample. The collector was filled with 50 mL de-ionized water, covered with air tight lid, and then kept beside the real sampler during the whole sampling time.

Field blanks were analyzed using the same procedure used for the dew, and results were excluded from the sample to avoid contamination error in the analysis. The pH and electrical conductivity were measured by using a pH meter (pH 211, Hanna Instruments) and conductivity meter (CM-5S, DKK-TOA Corporation). Concentration of water soluble cations (from filtered dew water) and trace metals (filtered dew water preserved with nitric acid) were determined by flame atomic absorption spectroscopy (model: AAnalyst 800, Perkin

Elmer). Concentrations of water-soluble anions were determined by ion chromatography (model: 881 compact IC pro, Metrohm). Samples were filtered with a 0.45- μm Millipore membrane filter to exclude any remaining ultrafine particles before analysis of metals and ions. Concentration of HCO_3^- was calculated from pH with Eq. (1) used by Kulshrestha et al. 2003:

$$[\text{HCO}_3^-] = 10^{-11.2+\text{pH}} \quad (1)$$

Results and discussion

Physical properties

The dew water samples were analyzed for pH and electrical conductivity (EC). While slightly turbid before filtration, indicating presence of solid particulate material, the filtered dew water samples were transparent and odorless, indicating absence of volatile odorous species. The average pH of the dew water was 6.81 ± 0.3 , nearly neutral. The acidity of dew water depends on dissolution of different gases (CO_2 , SO_x , and NO_x , etc.) and acidic aerosol (Beysens et al. 2006; Takeuchi et al. 2003), so the pH of the dew water is determined by the balance between acidic and neutralizing components, such as ammonium, bicarbonate, basic aerosol, etc. (Takeuchi et al. 2003). As a benchmark, rainwater pH is on the order of 5.6, owing to the formation of carbonic acid from CO_2 . The

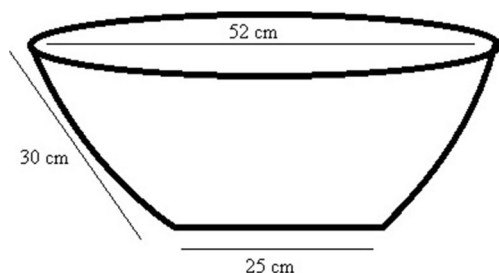


Fig. 2 Stainless steel bowl used for dew water collection (area $\approx 0.800 \text{ cm}^2$) in Coastal Bhola, Bangladesh

average EC of the collected dew water was $155 \pm 62 \mu\text{Scm}^{-1}$; corresponding to the total mineralization $119 \pm 48 \text{ mg L}^{-1}$ ($\approx 0.77 \cdot \text{EC}$). In Table 1, we compare our measured pH and EC to global dew water values. Our pH was comparable to that in Delhi and Agra (India) and rural Indiana (USA) but lower than in the other sites, except Bordeaux (France).

Chemical composition

The concentration of major ions in our dew water samples was measured and is presented in Table 1. The ionic concentrations of the measured cations and anions in our dew water samples reflect $\text{Ca}^{2+} > \text{Cl}^- > \text{SO}_4^{2-} > \text{Na}^+ > \text{NO}_3^- > \text{Mg}^{2+} > \text{K}^+ > \text{HCO}_3^-$. Concentration of measured total cations ($1.3 \times 10^3 \mu\text{eq L}^{-1}$) was 1.5 times higher than of total anions ($0.84 \times 10^3 \mu\text{eq L}^{-1}$). However, as we were unable to measure some ionic species, ion balance was not achieved in our data. From our measured ionic concentrations and the literature, in Table 1, we see that in Bhola, concentrations of NO_3^- and K^+ were higher than those in other locations, except for Negev, Israel. It is important to note here that most of the studies were conducted at urban sites. So, higher concentration of the anthropogenic species in dew water of Bhola is quite remarkable. In Bhola, K^+ was 2–3 times higher than in Agra (India), Changchun (China), and Zadar (Croatia), while NO_3^- was 1.5–2.5 times higher than in India (Delhi, Agra), Changchun (China), Yokohama (Japan), and Santiago (Chile) and much higher (5 to 27 times) than in all other locations. In Bhola, SO_4^{2-} was 3–16 times higher than in Bordeaux (France), USA (Indiana, Arkansas), and Zadar (Croatia) but lower than in all other sites. Concentration of Na^+ and Cl^- were also higher in Bhola Island samples than in most other sites, except Zadar, Croatia (closer to the sea) and Negev, Israel (desert/semi desert region).

Trace metals, including Zn, Fe, Mn, Cu, Cr, Pb, and Ni were measured in the Bhola dew water samples. The concentration order (in μgL^{-1}) of trace metals followed the sequence $\text{Zn} (345.5 \pm 154.5) > \text{Fe} (267.5 \pm 320.1) > \text{Mn} (186.2 \pm 96.5) > \text{Cu} (75.0 \pm 21.4)$, with concentrations of Cr, Pb, and Ni below minimum detection limit. The instrumental detection limits for Cr, Pb, and Ni were 100.0, 200.0, and $60.0 \mu\text{g L}^{-1}$, respectively. In the aerosol samples previously collected at the coastal Bhola site (Salam et al. 2003b), Fe and Zn concentrations were also very high. Correlation among different chemical species in dew water is given in Table 2. Significant correlation was observed between soil tracers Ca^{2+} and Mg^{2+} (0.62) indicating their common crustal origin. Correlation between K^+ and soil tracer Ca^{2+} (0.65) suggests that a significant fraction of the K^+ is of crustal origin. Strong correlation was also found between Na^+ and K^+ (0.87), Na^+ and Ca^{2+} (0.70), and Na^+ and Mg^{2+} (0.86), suggesting some fraction had marine origin. Cl^- also showed significant correlation with Na^+ (0.73) indicating probable sea salt.

Significant correlation between anionic species SO_4^{2-} and NO_3^- and their individual correlation (at $p = 0.03$) with biomass marker K^+ indicates their anthropogenic sources. Correlation is also obtained between different anionic (SO_4^{2-} , Cl^- , NO_3^-) and cationic species (K^+ , Ca^{2+} , Mg^{2+}) indicating their common sources or association of acidic and basic species after neutralization. Among trace metals, only Fe and Zn show strong correlation with each other. Mn shows strong correlation with SO_4^{2-} and NO_3^- . Among other trace metals, only Cu shows significant correlation with anionic species (Cl^-). No significant correlation was obtained between trace metals and other metals (Na, K, Ca, Mg, etc.). Correlation analysis of trace metals suggests anthropogenic sources as they showed little correlation with soil and/or sea-derived species.

Table 2 Correlation among different chemical species present in dew water

	Na^+	K^+	Ca^{2+}	Mg^{2+}	Cl^-	NO_3^-	SO_4^{2-}	Fe	Cu	Mn	Zn
Zn	-0.03	0.40	0.14	0.25	0.53	0.22	0.61*	0.69**	0.18	0.06	1
Mn	0.02	0.53	0.46	-0.03	0.47	0.66**	0.82**	-0.38	-0.24	1	
Cu	0.33	0.45	0.43	0.31	0.67**	0.01	-0.11	0.43	1		
Fe	-0.03	-0.21	-0.19	0.14	-0.07	-0.33	-0.09	1			
SO_4^{2-}	0.23	0.56*	0.43	0.013	0.63*	0.73**	1				
NO_3^-	0.20	0.57*	0.69**	-0.06	0.73**	1					
Cl^-	0.73**	0.96**	0.72**	0.39	1						
Mg^{2+}	0.86**	0.32	0.62*	1							
Ca^{2+}	0.70**	0.65**	1								
K^+	0.87**	1									
Na^+	1										

** $p < 0.01$; * $p < 0.03$ at Coastal Bhola, Bangladesh (an IGP outflow location to the Bay of Bengal)

Fractional acidity and neutralization factor

Fractional acidity of the dew water was calculated using the following formula (Yadav and Kumar 2014):

$$FA = [H^+]/[SO_4^{2-} + NO_3^-] \quad (2)$$

FA is 0 for maximum and 1 for no neutralization situation.

The average calculated value of FA in this study was 0.003 (highest 0.01, lowest 0.002), which indicates effective neutralization of acidic components (NO_3^- , SO_4^{2-} , etc.) in the dew water by basic components (NH_4^+ , Ca^{2+} , Mg^{2+} , and K^+). The contribution of basic components to the neutralization process can be determined by calculating the neutralization factor (NF), via the equation (Yadav and Kumar 2014):

$$NF_x = [X]/[NO_3^- + SO_4^{2-}] \quad (3)$$

where X is the species whose neutralization factor is to be calculated.

The order by scale (larger to smaller) of neutralization factors found in dew water of Bhola Island is NF_{Ca} (1.42) > NF_{Mg} (0.42) > NF_K (0.28). Dew forms near the ground, and thus collects more neutralizing particles (Ca^{2+} , Mg^{2+}) from soil than does water that forms at higher elevations (rain, cloud, etc.) (Yadav and Kumar 2014). Unfortunately, while NH_4^+ is another important neutralization species in dew water, its concentration was not measured in the current study.

Enrichment factor of major ions and trace metals

Enrichment factors were calculated to assist in identifying the origin of elements/ions in dew water (Kulshrestha et al. 1996). The calculation compares elemental ratios between ions/elements collected in the sample to reference material ratios. In coastal areas like Bhola, Na is commonly taken as a reference element for seawater since it is assumed to be primarily marine contribution in coastal regions (Kulshrestha et al. 2003). Al and Ca are typical lithophilic elements normally used as reference elements for continental crust (Cao et al. 2009). $EF_{sea\ water}$ and EF_{crust} of major cations and anions were calculated by the following equations, using Na and Ca as reference materials:

$$EF_{sea\ water} = (X/Na)_{dew}/(X/Na)_{sea} \quad (4)$$

$$EF_{crust} = (X/Ca)_{dew}/(X/Ca)_{soil} \quad (5)$$

where X is the concentration of the species of interest. $(X/Na)_{sea}$ is the ratio from seawater composition (Kidron and Starinsky 2012), and $(X/Ca)_{crust}$ is the ratio from crustal composition (Taylor 1964).

An EF value much less than or much higher than 1 is considered to be diluted or enriched respectively relative to the reference source (Safai et al. 2004). $EF_{sea\ water}$ and EF_{crust} values are listed in Fig. 3.

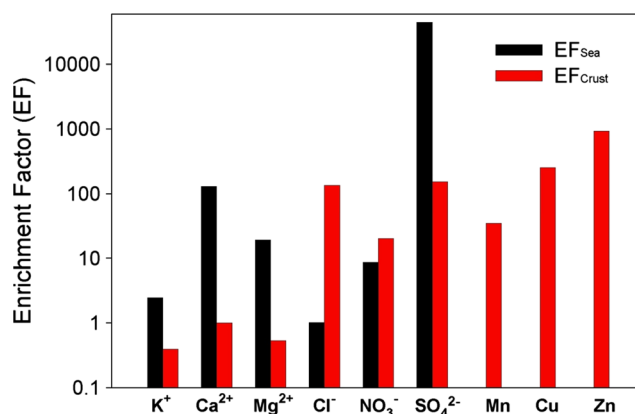


Fig. 3 Enrichment factor of major ions and trace metals in dew water at Coastal Bhola, Bangladesh

Cl^- has $EF_{sea\ water}$ value of 1.02 but has an EF_{crust} 135.5, which clearly indicates its marine origin. And, although the Cl^-/Na^+ value of dew water (1.19) is slightly higher than of sea water (1.16), this merely suggests very small contributions from other sources of Cl^- , such as K^+ . The average EF_{crust} and $EF_{seawater}$ value of K^+ and Mg^{2+} suggested primarily crustal sources, with partial contribution from marine sources. The high EF_{crust} and $EF_{seawater}$ values of SO_4^{2-} and NO_3^- indicated that both were enriched in dew water compared to soil and seawater, suggesting that their primary sources was anthropogenic.

The EF value of trace metals with respect to the crustal material (e.g., Al, Fe) is often used to evaluate the degree of anthropogenic influence (Huang et al. 2013). Enrichment factors (EF) of trace metals were calculated in this study using Fe as reference material by the following equation (Tripathee et al. 2014):

$$EF = (X/Fe)_{dew}/(X/Fe)_{crust} \quad (6)$$

where X is the concentration of the species of interest. The ratios of $(X/Fe)_{crust}$ are taken from Salam et al. (2003a).

Elements with an $EF > 10$ are considered to be of mainly anthropogenic origin (Huang et al. 2013). Based on EF, the trace elements can be classified as non-enriched ($EF = 1$ to 10), moderately enriched ($EF = 10$ to 100), and highly enriched ($EF > 100$) (Al-Momani 2003). Enrichment factors of trace metals in dew water collected from Bhola (see Fig. 3) suggest that their source is mainly anthropogenic ($EF > 10$). Moreover, EF of Cu and Zn were greater than 100, indicating highly enriched conditions, suggesting severe contamination due to human activities (Duce et al. 1975).

Source contributions

Marine contribution

In order to determine marine contribution to the chemical species present in dew water, the sea salt fraction (SSF) and

non-sea salt fraction (NSSF) have been calculated. Bhola is an island surrounded by the Bay of Bengal, so significant marine contribution is expected in dew water. The equations used for calculating SSF and NSSF are taken from (Kulshrestha et al. 1996)

$$\%SSF = 100(X/Na)_{\text{sea}}/(X/Na)_{\text{dew}} \quad (7)$$

$$\%NSSF = 100-(\%SSF) \quad (8)$$

where X is the concentration of the species of interest, and $(X/Na)_{\text{sea}}$ is the ratio from seawater composition (Kidron and Starinsky 2012).

Na is used as reference, as this is a coastal site, and nearly all the sodium is of marine origin. From the value of %SSF, significant marine contributions are found for Cl^- (97.47%), SO_4^{2-} (11.57%), K^+ (40.53%), and Mg^{2+} (5.23%) (see Fig. 4). The higher %NSSF value of SO_4^{2-} (88.43%), K^+ (59.47%), Mg^{2+} (94.77%), Ca^{2+} (99.23%), and NO_3^- (99.99%) suggest that these mostly came from non-marine sources.

Crustal and anthropogenic contribution

In order to estimate the crustal contribution and anthropogenic contribution of different species present in dew water, crustal fraction (%CF) and anthropogenic fraction (%AF) were calculated using the following equations (Lu et al. 2011):

$$\%CF = 100(X/Ca)_{\text{soil}}/(X/Ca)_{\text{dew}} \quad (9)$$

$$\%AF = ((100-SSF(\%))-CF(\%)) \quad (10)$$

where X is the concentration of the species of interest. Here, Ca is used as reference element for continental crust, since it is a natural source for lithophilic elements and its composition in soil is stable (Cao et al. 2009).

Potassium occurs in the coarse particles of soil, but fine particles of potassium could result from biomass burning, industrial sources, or sea spray. Here, the primary source of NSSF K^+ was considered crustal, as we were unable to

distinguish between crustal sources and biomass/wood combustion in the present study (Cao et al. 2009; Yadav and Kumar 2014). The source contribution for different species is shown graphically in Fig. 4. The calculated value of %CF suggested small contribution of soil originated SO_4^{2-} , Cl^- , and NO_3^- in dew water. The %NSSF of K^+ and Mg^{2+} is considered as having pure crustal origin (Lu et al. 2011; Safai et al. 2004). Natural dust makes an important contribution to the ions present in dew water as the crustal sources of Ca^{2+} , Mg^{2+} , and K^+ are dust aerosols, originating from soil, which are suspended in the lower layer of the atmosphere, and transported on our collection surface by wind/deposition (Xu et al. 2015). Ca^{2+} and Mg^{2+} could be derived from the dissolution of both primary and secondary minerals ($CaCO_3$, $CaCO_3 \cdot MgCO_3$, and $CaSO_4 \cdot 2H_2O$) commonly present as aerosol particles (Yadav and Rajamani 2006). From the values of (%AF) in Fig. 4, we see that while a small fraction of Cl^- (1.8%) potentially comes from anthropogenic sources, SO_4^{2-} and NO_3^- have the highest anthropogenic contribution with %AF values of 83.5 and 99.3%, respectively.

Air mass trajectories analysis

Backward air mass trajectories were calculated for selected sampling days (for samples having the highest and lowest concentrations of SO_4^{2-} , NO_3^- , and trace metals) with the NOAA HYSPLIT model (hybrid single-particle lagrangian integrated trajectory model) (<http://www.arl.noaa.gov/>). The 48-h air mass trajectories were calculated at 500, 1500, 2500 meters AGL (above ground level). Figure 5a represents trajectories for the sample having the lowest concentrations of SO_4^{2-} , NO_3^- , and trace metals. That sample had 6.2 times lower concentration of ($SO_4^{2-} + NO_3^-$) than average and 5.8 times lower concentration of trace metals (sum of concentration of all trace metals) than average. From Fig. 5a, the calculated air masses at all levels were coming from the Bay of Bengal. On this sampling day, dew water was significantly influenced by clean air from over ocean, hence the observed very low concentrations of measured chemical species. The second case, having a lower than average concentration of ($SO_4^{2-} + NO_3^-$), is shown in Fig. 5b. Here, the air mass was coming from the IGP region to the sampling location carrying polluted air masses. The third case, Fig. 5c, represents sampling days when the concentration of ($SO_4^{2-} + NO_3^-$) was 1.25 times higher than average. Back trajectories for this case show that air masses came from the north (500 MAGL) and west directions (1500 and 2500 MAGL) on that sampling day. Figure 5d represents the fourth case: sampling days when concentration of ($SO_4^{2-} + NO_3^-$) was 1.63 times higher than average. Figure 5e represents our fifth case: the sampling day when the concentration of trace metals was

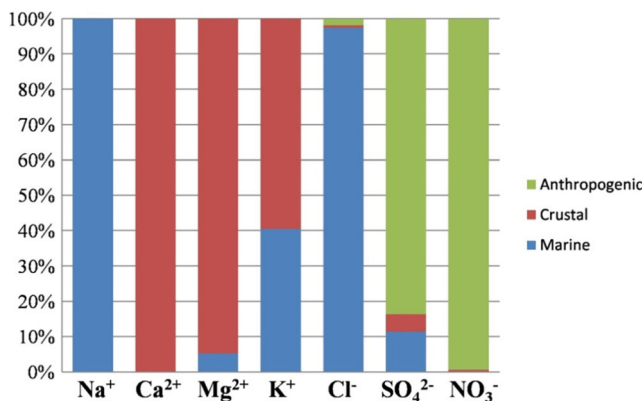
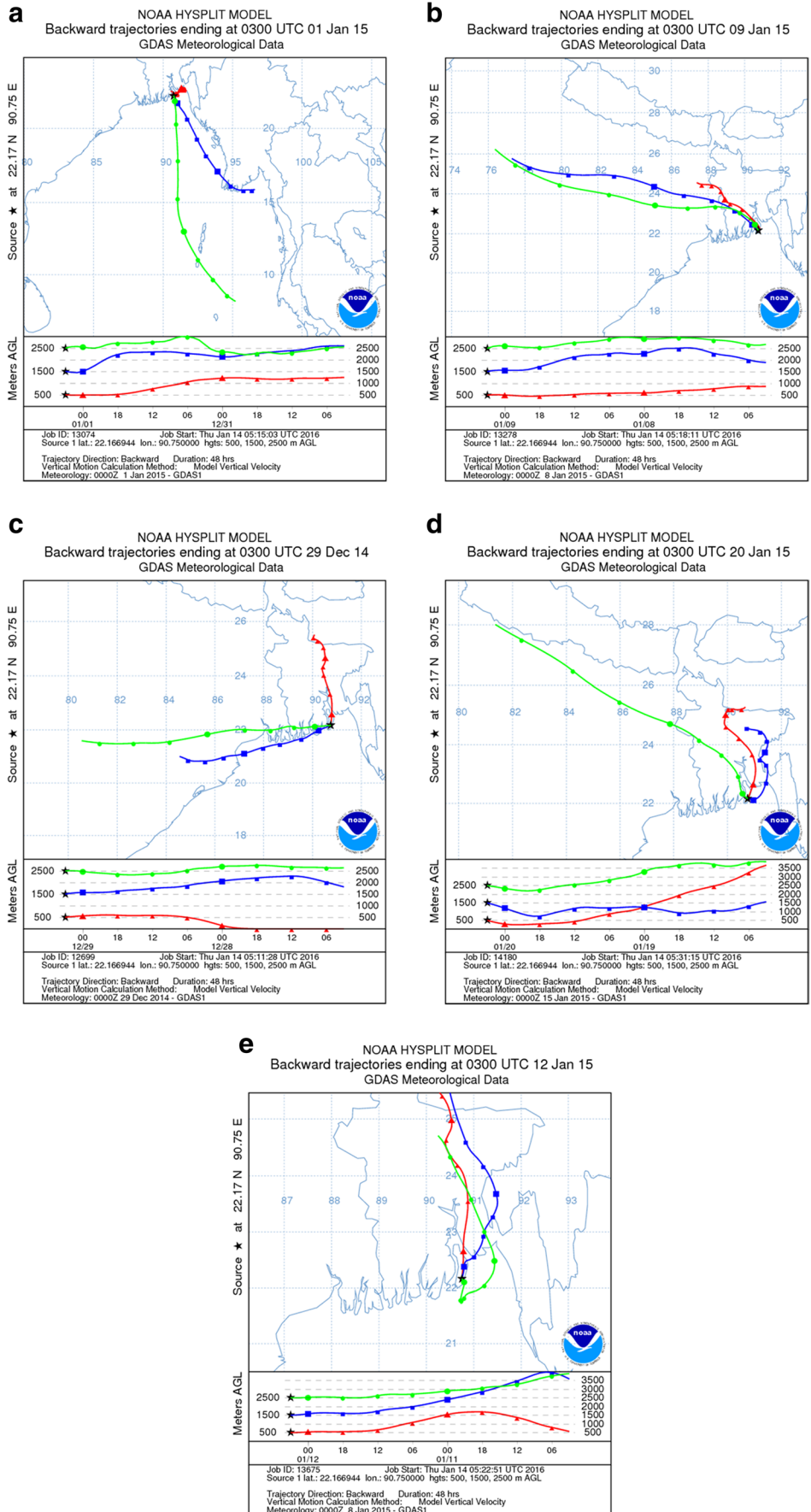


Fig. 4 Source contribution for different species present in the dew water at coastal Bhola, Bangladesh

Fig. 5 **a** Air mass trajectories when SO_4^{2-} , NO_3^- , and trace metals were lowest. **b** Air mass trajectories when $\text{SO}_4^{2-} + \text{NO}_3^-$ was lower than the average value. **c** Air mass trajectories when $\text{SO}_4^{2-} + \text{NO}_3^-$ was higher than the average value. **d** Air mass trajectories when $\text{SO}_4^{2-} + \text{NO}_3^-$ was highest. **e** Air mass trajectories when trace metals were highest



highest in the dew water. The concentration of total trace metals in Fig. 5e was 2.02 times higher than the average total trace metal concentration. The air masses on that day came directly from north.

Bhola is a remote rural Island of the Bay of Bengal and does not host much industrial activity.

The most probable major sources of the anthropogenic species in dew water in Bhola are air masses coming from different parts of IGP, as seen through the trajectory analysis. Although, biomass burning for cooking and agriculture activities could be responsible for some anthropogenic contributions; if it is the main source, then dew water samples should have very low concentrations of SO_4^{2-} , NO_3^- , and trace metals. Along the back trajectory of the air masses (Fig. 5), there are some big cities in Bangladesh and India. Moreover, winter time post-harvest agriculture burning could also have significantly impacted aerosol as well as dew water composition over the IGP (Rajput et al. 2011). High anthropogenic contributions of SO_4^{2-} and NO_3^- in dew water (the “Crustal and anthropogenic contribution” section) is an indicator for emissions of SO_x and NO_x into the atmosphere from different sources upwind in IGP, e.g., coal combustion, biomass burning, industrial emission, and vehicular exhaust (Yadav and Kumar 2014; Rubio et al. 2002). Dissolution of precursor gases or their oxidized products is likely responsible for the very higher concentrations of NO_3^- as well as SO_4^{2-} in dew water in coastal Bhola. The sources of trace metals in dew water are likely similar contamination from aerosol particles from upwind sources. It appears that dew water in coastal Bhola during winter time is contaminated when exposed to long-range transported air masses from polluted IGP regions.

Conclusion

Dew water collected in coastal Bhola, Bangladesh, during winter time was almost neutral with an average pH of 6.81. Concentrations of NO_3^- , K^+ , Na^+ , and Cl^- in dew water were relatively higher in Bhola. Further analyses for enrichment factor, percent source contribution, and correlation analyses revealed that the elemental composition of dew water at coastal Bhola had substantial contributions from anthropogenic sources. Air mass trajectories analyses showed that regional urban pollution has a significant influence on the dew water chemical composition in this remote, rural, IGP outflow location.

Acknowledgements The authors acknowledge the financial support from the Office of the Naval Research Global (ONRG), USA, for the construction of the Bhola Observatory. Thanks to the sampling crews. Thanks also to the Centre for Advanced Research in Sciences (CARS), Dhaka University, for helping with the chemical analysis.

References

- Al-Momani IF (2003) Trace elements in atmospheric precipitation at northern Jordan measured by ICP-MS: acidity and possible sources. *Atmos Environ* 37:4507–4515
- Baier W (1966) Studies on dew formation under semi-arid conditions. *Agric Meteorol* 3:103–112
- Beysens D, Ohayon C, Muselli M, Clus O (2006) Chemical and biological characteristics of dew and rain water in an urban coastal area (Bordeaux, France). *Atmos Environ* 40:3710–3723
- Beysens D, Lekouch I, Mileta M, Milimouk I, Muselli M (2009) Dew and rain water collection in South Croatia. *Int J Chem Mol Nucl Mater Metall Eng* 3:141–147
- Beysens D (1995) The formation of dew. *Atmos Res* 39:215–237
- Cao YZ, Wang S, Zhang G, Luo J, Lu S (2009) Chemical characteristics of wet precipitation at an urban site of Guangzhou, South China. *Atmos Res* 94:462–469
- Duce RA, Hoffman GL, Zoller WH (1975) Atmospheric trace metals at remote northern and southern hemisphere sites: pollution or natural? *Science* 187:339–342
- Dudevani S (1947) An optical method of dew estimation. *Q J R Meteor Soc* 73:282–296
- Eck TF, Holben BN, Dubovik O, Smirnov A, Slutsker I, Lobert JM, Ramanathan V (2001) Column-integrated aerosol optical properties over the Maldives during the northeast monsoon for 1998–2000. *J Geophys Res* 106:28555–28566
- Huang J, Kang S, Zhang Q, Guo J, Chen P, Zhang G, Tripathee L (2013) Atmospheric deposition of trace elements recorded in snow from the Mt. Nyainqentanglha region, southern Tibetan plateau. *Chemosphere* 92:871–881
- Janssen LHJM, Römer FG (1991) The frequency and duration of dew occurrence over a year: model results compared with measurements. *Tellus Ser B* 43:408–419
- Jiries A (2001) Chemical composition of dew in Amman, Jordan. *Atmos Res* 57:261–268
- Kidron GJ (1998) A simple weighing method for dew and fog measurements. *Weather* 53:428–433
- Kidron GJ, Starinsky A (2012) Chemical composition of dew and rain in an extreme desert (Negev): cobbles serve as sink for nutrients. *J Hydrol* 420–421:284–291
- Kulshrestha UC, Sarkar AK, Srivastava SS, Parashar DC (1996) Investigation into atmospheric deposition through precipitation studies at New Delhi (India). *Atmos Environ* 30:4149–4154
- Kulshrestha UC, Kulshrestha MJ, Sekar R, Sastry GSR, Vairamani M (2003) Chemical characteristics of rainwater at an urban site of south-central India. *Atmos Environ* 37:3019–3026
- Lakhani A, Parmar RS, Prakash S (2012) Chemical composition of dew resulting from radiative cooling at a semi-arid site in Agra, India. *Pure Appl Geophys* 169:859–871
- Lekouch I, Mileta M, Muselli M, Milimouk-Melnitshouk I, Šojat V, Kabbachi B, Beysens D (2010) Comparative chemical analysis of dew and rain water. *Atmos Res* 95:224–234
- Lu X, Li LY, Li N, Yang G, Luo D, Chen J (2011) Chemical characteristics of spring rainwater of Xi’an city, NW China. *Atmos Environ* 45:5058–5063
- Lynam MM, Dvonch JT, Hall NL, Morishita M, Barres JA (2015) Trace elements and major ions in atmospheric wet and dry deposition across central Illinois, USA. *Air Qual Atmos Health* 8:135–147
- Nair VS, Moorthy KK, Alappattu DP, Kunhikrishnan PK, George S, Nair PR, Babu SS, Abish B, Satheesh SK, Tripathi SN, Niranjan K, Madhavan BL, Srikant V, Dutt CBS, Badarinath KVS, Reddy RR (2007) Wintertime aerosol characteristics over the Indo-Gangetic Plain (IGP): impacts of local boundary layer processes and long-range transport. *J Geophys Res* 112:205

- Niranjan K, Sreekanth V, Madhavan BL, Krishna MK (2006) Wintertime aerosol characteristics at a north Indian site Kharagpur in the Indo-Gangetic plains located at the outflow region into Bay of Bengal. *J Geophys Res* 111:209
- Okochi H, Kajimoto T, Arai Y, Igawa M (1996) Effect of acid deposition on urban dew chemistry in Yokohama, Japan. *Bull Chem Soc Jpn* 69:3355–3365
- Polkowska Ż, Błaś M, Klimaszewska K, Sobik M, Małek S, Namieśnik J (2008) Chemical characterization of dew water collected in different geographic regions of Poland. *Sensors* 8:4006–4032
- Rajput P, Sarin MM, Rengarajan R (2011) High precision GC-MS analysis of atmospheric polycyclic aromatic hydrocarbons (PAHs) and isomer ratios from biomass burning emissions. *J Environ Prot* 2: 445–453
- Rubio MA, Lissi E, Villena G (2002) Nitrite in rain and dew in Santiago city, Chile. Its possible impact on the early morning start of the photochemical smog. *Atmos Environ* 36:293–297
- Rubio MA, Lissi E, Herrera N, Pérez V, Fuentes N (2012) Phenol and nitrophenols in the air and dew waters of Santiago de Chile. *Chemosphere* 86:1035–1039
- Safai PD, Rao PSP, Momin GA, Ali K, Chate DM, Praveen PS (2004) Chemical composition of precipitation during 1984–2002 at Pune, India. *Atmos Environ* 38:1705–1714
- Salam A, Bauer H, Kassin K, Ullah SM, Puxbaum H (2003a) Aerosol chemical characteristics of a mega-city in Southeast Asia (Dhaka–Bangladesh). *Atmos Environ* 37:2517–2528
- Salam A, Bauer H, Kassin K, Ullah SM, Puxbaum H (2003b) Aerosol chemical characteristics of an island site in the Bay of Bengal (Bhola–Bangladesh). *J Environ Monit* 5:483–490
- Salam A, Hossain T, Siddique MNA, Alam AMS (2008) Characteristics of atmospheric trace gases, particulate matter, and heavy metal pollution in Dhaka, Bangladesh. *Air Qual Atmos Health* 1:101–109
- Singh SP, Khare P, Kumari KM, Srivastava SS (2006) Chemical characterization of dew at a regional representative site of north-central India. *Atmos Res* 80:239–249
- Takeuchi M, Okochi H, Igawa M (2002) Controlling factors of weak acid base concentrations in urban dew water: comparison of dew chemistry with rain and fog chemistry. *Bull Chem Soc Jpn* 75:757–764
- Takeuchi M, Okochi H, Igawa M (2003) Deposition of coarse soil particles and ambient gaseous components dominating dew water chemistry. *J Geophys Res* 108:4319
- Taylor SR (1964) Abundance of chemical elements in the continental crust: a new table. *Geochimica Cosmochimica Acta* 28:1273–1285
- Tripathee L, Kang S, Huang J, Sillanpää M, Sharma CM, Lüthi ZL, Guo J, Paudyal R (2014) Ionic composition of wet precipitation over the southern slope of central Himalayas, Nepal. *Environ Sci Pollut Res* 12:2677–2687
- Wagner GH, Steele KF, Peden ME (1992) Dew and frost chemistry at a midcontinent site, United States. *J Geophys Res* 97:20591–20597
- Wentworth GR, Murphy JG, Benedict K, Bangs EJ, Collett JL Jr (2016) The role of dew as a nighttime reservoir and morning source for atmospheric ammonia. *Atmos Chem Phys Discuss* 16:7435–7449
- Xu Y, Zhu H, Tang J, Lin Y (2015) Chemical compositions of dew and scavenging of particles in Changchun, China. *Adv Meteorol* 2015 Article ID 104048
- Yadav S, Rajamani V (2006) Air quality and trace metal chemistry of different size fractions of aerosols in N–NW India—implications for source diversity. *Atmos Environ* 40:698–712
- Yadav S, Kumar P (2014) Pollutant scavenging in dew water collected from an urban environment and related implications. *Air Qual Atmos Health* 7:559–566
- Zangvil A (1996) Six years of dew observations in the Negev Desert, Israel. *J Arid Environ* 32:361–371