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Evaluation of physicochemical and trace metal qualities of rainwater in the south-eastern region of Bangladesh

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ABSTRACT

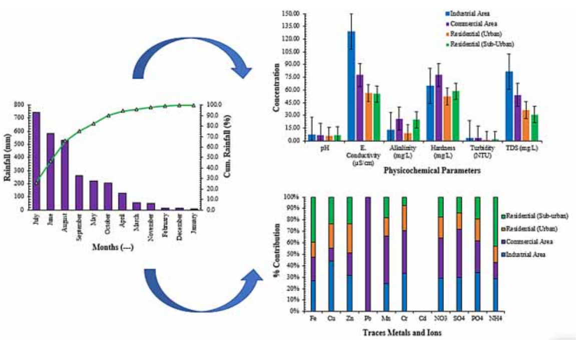
The chemical characteristics of rainwater in the south-eastern region of Bangladesh were investigated in this study in addition to identifying the potential sources of different precipitation constituents, which have often been unexplored or not well covered in the literature. Rainwater pH, major ions and trace metals were measured in samples collected from five different locations with different land-use patterns of the Chattogram Metropolitan area (CMA) during two rainy seasons. The results of this study showed variability in rainwater quality across the sites signifying site-specific influences. The mean concentration of all measured physicochemical parameters, ions and trace metals in rainwater samples was also found to be significantly lower compared with the drinking water quality standard of Bangladesh and WHO guidelines. The correlation matrix and principal component analysis (PCA) indicated that NO_3^- and SO_4^{2-} originated from anthropogenic sources, while the average concentration of trace metals found in rainwater was exhibited in the following order: Zn>Cu>Fe>Cr>Mn>Pb>Cd. The findings of this study could be used as a reference to further investigate the influences of industrial, urban and agricultural emissions that regulate the chemical characteristics of the atmosphere in particular areas of study.

Key words: chemical composition, commercial area, industrial area, rainwater, south-eastern Bangladesh, statistical analysis

HIGHLIGHTS

- Chemical composition of rainwater is investigated.
- Influence of land-use pattern on rainwater quality is reported.
- The concentrations of trace metals are found in order of Zn > Cu > Fe > Cr > Mn > Pb > Cd.
- Anthropogenic sources and environmental conditions are found that the controlling sources affecting the chemical composition of rainwater.

GRAPHICAL ABSTRACT



INTRODUCTION

Rainwater is first and foremost natural water used in many countries as an alternate source of water for human survival. Although rainwater is relatively safe, the chemical composition at the point of collection is often influenced by the local environment and atmospheric conditions. Consequently, different locations may have different elemental concentrations that are key to the quality and suitability of uses. Rainwater chemistry is formed through the dynamic interaction between atmospheric (e.g., cloud) dynamics and microphysical actions together with a sequence of chemical reactions in the atmosphere caused by rainout and washout (Bertrand *et al.* 2008; Mohanakumar 2008; Al-Khashman 2009; Mix *et al.* 2019). As noted by Wang & Han (2011), rainwater chemistry signifies the chemical characteristics of the atmosphere through which it falls, which helps to understand and investigate the soluble elements that exist in rainwater. Moreover, the chemical composition of rainwater provides useful information about the relative contributions of various contaminants in the atmosphere, and their regional dispersion (Kulshrestha *et al.* 2003; Wu *et al.* 2012). It also assists to investigate site-specific characteristics, such as the influence of emissions from industrial, urban and agricultural sectors, and the biogeochemical factors that elaborate the physical processes regulating the chemical characteristic of the atmosphere in regional areas and natural biogeochemical cycle (Salve *et al.* 2008; Tiwari *et al.* 2008). For example, Moreda-Piñeiro *et al.* (2014) reported that the chemical composition of rainwater in a sub-urban area of Spain is influenced by sea, terrestrial and anthropogenic sources. Furthermore, the influence of sea salts, car exhaust, industrial dust and secondary aerosols on chemical composition of rainwater has been identified by Huston *et al.* (2012). Given the likelihood of the adverse effects of local environment and atmospheric conditions on the characteristics of rainwater, the chemical composition of rainwater should be investigated to identify the possible sources of different precipitation constituents in a particular region.

Chattogram, a commercial capital, is one of the largest and busiest port cities in Bangladesh and is situated in the eastern coastal zone of Bangladesh (Brammer 2014). The city is home to nearly 4.5 million people, and the sources of water are a mix of surface and groundwater. However, groundwater near the coastal belt observes substantially higher salinity in the dry period due to the scarcity of rainfall along with arsenic seen in a few cases shallow tube wells of households (Akter & Ali 2011; Jabed *et al.* 2018). Accordingly, people mostly depend on water supplied by the Chattogram Water Supply and Sewerage Authority (CWASA) for both domestic and drinking purposes that are intermittent and only cover around 69% of the demand of city dwellers. The river water quality near the city area is unsuitable due to the lack of wastewater treatment facilities and poor solid waste management system. As such, CWASA has to look further upstream, to search for comparatively less polluted surface water deemed suitable for the treatment of wastewater (Ahmed *et al.* 2010; Hossen & Jishan 2018). However, at present, taking poor quality surface water and groundwater with high concentration of salinity and arsenic compared with the levels recommended by Bangladesh drinking water quality standards and WHO guidelines, rainwater is the only practical option remaining for the area particularly given the annual mean precipitation 2,488 mm during the monsoon season in Bangladesh.

In the past years, much research investigations have been devoted to identifying the physicochemical quality of rainwater worldwide (see Al-Khashman 2009; Huston *et al.* 2012; Moon *et al.* 2012; Vialle *et al.* 2012; Morales-Pinzón *et al.* 2015; Mimura *et al.* 2016; Bharti *et al.* 2017). While the potential for rainwater harvesting methods, reliability and opportunities across Bangladesh are increasingly being studied (e.g., Islam *et al.* 2010; Karim 2010; Alam *et al.* 2012; Akter & Ahmed 2015; Karim *et al.* 2015; Bashar *et al.* 2018; Rahaman *et al.* 2019; Akter *et al.* 2020), the chemical characteristics of rainwater and the variation of relative contributions of different atmospheric pollutants concerning different land uses remain generally unexplored considering its merit and suitability of uses. In Bangladesh, particularly in the southern-eastern region of the country (e.g., Chattogram), no such study is found at present. This limitation and gap have motivated the need to undertake a comprehensive investigation on the chemical characteristics of rainwater in the Chattogram region across various social and economic zones of the city (e.g., industrial, commercial, residential and urban areas). The primary aim of this study, therefore, is to gain a better understanding of the physicochemical and trace metal qualities of rainwater across varying geographical locations of Chattogram in addition to identifying the potential sources of different precipitation constituents in the region.

MATERIALS AND METHODS

The rainwater chemistry was identified by conducting extensive field and laboratory tests based on various water quality parameters (e.g., pH, turbidity, alkalinity, hardness, major ions and trace metals). The resulting chemical composition of rainwater obtained in the study area was then compared with the existing standards of drinking water quality parameters applicable to the study region.

Study area

The city of Chattogram is a coastal city and commercial hub located on the south-eastern side of Bangladesh. The Chattogram Metropolitan area (CMA) is four times greater than the Chattogram City Corporation (CCC) area, as illustrated in Figure 1 (22°6′41″N–22°33′38″N, 90°41′E–92°2′E) (Mia *et al.* 2015). The city is bounded by the Bay of Bengal on the west, by the Halda River on the north-east and, by the Karnaphuli River to the south-east. The busiest seaport of Bangladesh is located at the estuary of the Karnafuli River (Mia *et al.* 2015). Given the location of the port facility, the city is business-friendly, with hundreds of the oldest and largest industries of Bangladesh established here (BBS 2013). Indeed, the CMA is one of the critical coastal regions of the country with a population of around 4.5 million people, density 6,000/km^2, with a growth rate of 2.06% per annum (BBS 2011).

From a geographic perspective, the central city area is situated on flat land, with low height hillocks mainly towards the north with a peak elevation level of 351 m in Sitakunda (Roy *et al.* 2020). A tropical monsoon climate mostly dominates the weather conditions of the city. According to data from the Chattogram station of the Bangladesh Meteorological Department (BMD), between 2,400 and 3,000 mm annual rainfall occurs in the Chattogram region, of which 80% occurs between May and September each year (see Figure 2). The mean annual relative humidity is 73.7% which varies each month between 58% in January to 86% in August (BMD 2013). During the dry weather season (November to March), typically, a mild cold wind blows from the north to the south of the country with very little precipitation. The average maximum and minimum air temperature vary between 32.3 and 13 °C, in summer and winter, respectively (BMD 2013).

Five locations were selected in this study in addressing different land uses for collecting rainwater samples in which three are in the city centre (see Figure 1). At the same time, the remaining two sites were located outside of the city centre, one is an industrial area named Kumira and the other is a sub-urban residential area named Pahartali. Generally, in a field of study such as this, there will exist residential, commercial and industrial characteristics. This is the reason why in this study, all aspects of the city based on their particular use of land were selected. In particular, rainwater parameters of General Electric Company (GEC) and Agrabad locations belonging to commercial land use might have been influenced by the road traffic environment due to the volume of high traffic. However, the highest traffic-induced noise pollution was recorded at the GEC intersection and Agrabad circle (Mia *et al.* 2015; Uddin 2018). As such, the growing concern for city dwellers is air pollution, mostly caused by automobile emissions since the volume of vehicles is increasing daily. In 2010, the number of registered vehicles was 84,391, not including several thousand non-registered vehicles (Shamsher & Abdullah 2013).

Suspended particulate matter (SPM), sulphur dioxide (SO_2) and nitrogen oxides (NO_x) are the three major components of air pollution in Chattogram city with SPM being the most concentrated pollutant (Mia *et al.* 2015; Hossen *et al.* 2018; Masum & Pal 2020). The Pahartali site is mainly residential in nature where most of the Bangladesh Railway residence is located. Kumira is an industrial area located in the north of the CMA, the Bay of Bengal on one side and hills on the other. Also, various cement, glass and steel manufacturing industries supporting shipbuilding are situated here. As such, the soil, water and air around the shipbuilding industry are highly polluted by toxic substances (Abdullah *et al.* 2013; Hasan *et al.* 2013; Hossain *et al.* 2016). The sampling site of Chittagong University of Engineering & Technology (CUET) is situated in Raozan Upazila, outside of CMA boundary, which is considered a pristine site with relatively little pollution compared with the other sampling sites. The area here is predominantly sub-urban with a relatively clean environment in which agricultural land use prevails along with academic activities.

Sample collection

Rainwater samples were collected on an event basis, in which 95 wet-only rainwater samples were collected from two rainy seasons (June 2018 to October 2018 and June 2019 to October 2019). In 2018, the average monthly rainfall was 235.83 $\pm$ 280.23 mm with a maximum intensity of 791 mm, while in 2019, the average monthly rainfall was 219.75 $\pm$ 321.49 mm with a maximum intensity of 1,108 mm (BMD). The rainwater collector, as shown in Supplementary Material, Figure S1, was placed on the building roof at each rainwater sampling site described above with varying heights ranging between 10 and 20 m. The first flush tank was incorporated to separate the rainwater after the first flush using a ballcock. In this study, the water from the rainwater tank was used for analysis. In the rainwater tank, there is also a ballcock that is used to prevent the passing of the remaining rainwater residing in the catchment after filling up the rainwater tank. This is because,

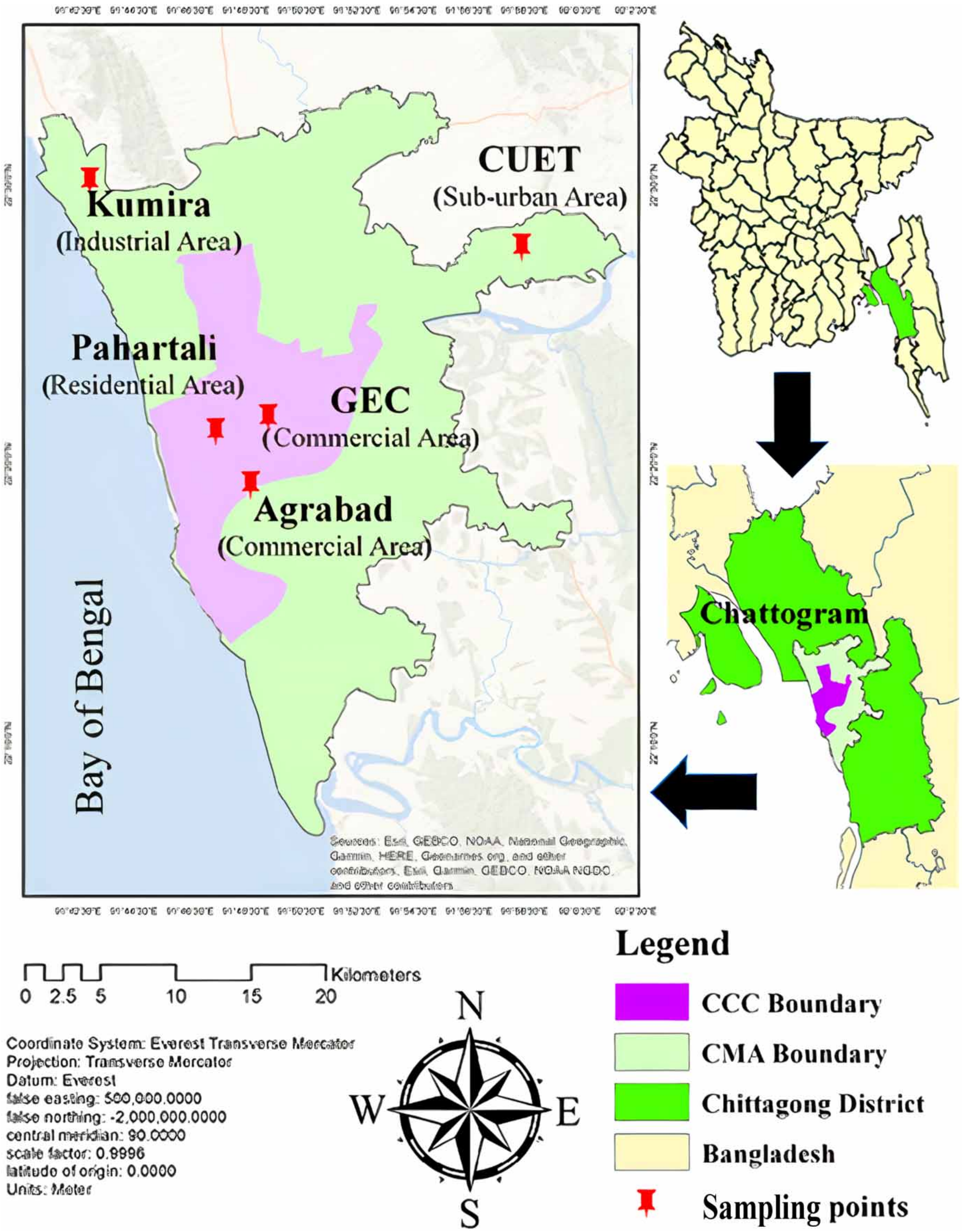


Figure 1 | Map of the studied area showing the location of the sampling site.

during the period between the rainfall and collection, there is a possibility of contamination caused by birds and insects since the catchment was exposed (open), whereas the rainwater tank was closed. In this study, the initial water that was flushed was not considered for testing since it was agreed that the first flush would be omitted concerning contamination (Zhu *et al.* 2004;

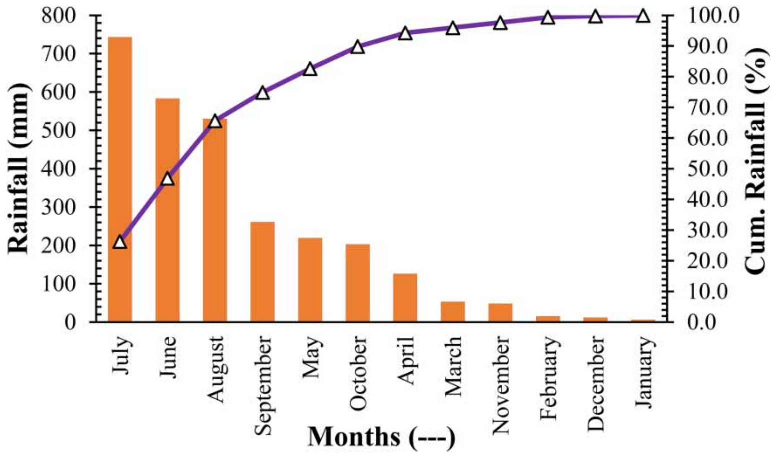


Figure 2 | The monthly average rainfall scenario in the study area during the period (1982–2019) (Source: BMD).

Kus *et al.* 2010; Gikas & Tsihrintzis 2012). Between the two rainfall events, the rainwater tank was physically cleaned while the dry deposition of pollutants was mainly influenced by atmospheric fall-out, which was unavoidable. Immediately after each rainfall event, the rainwater samples were collected using a 500 mL polypropylene bottle. Before collecting the rainwater, the bottles were thoroughly washed using distilled water to avoid signs of cross-contamination.

Sample analysis

To investigate the chemical composition of rainwater in the study area, physicochemical parameters, trace metals and major ions were considered within this work. Adopting the well-established approach of rainwater quality investigations as reported in the literature (see Li *et al.* 2011; Alam *et al.* 2012; Huston *et al.* 2012; Amponsah *et al.* 2015; Morales-Pinzón *et al.* 2015; Mimura *et al.* 2016; Bharti *et al.* 2017; Majumdar *et al.* 2020), physicochemical parameters, that is, pH, electrical conductivity (EC), total dissolved solids (TDS), alkalinity, hardness and turbidity, trace metals such as Fe, Cu, Zn, Pb, Mn, Cr and Cd and major ions, that is, NO_3^-, SO_4^{2-}, PO_4^{3-} and NH_4^+ were selected and measured in this study. The analytical parameters and method for assessing water quality of rainwater samples are provided in Supplementary Material, Table S1. *In situ* measurements of pH, EC and TDS were done using a Smart Digital pH meter and HQ40D Multi-meter. The pH meter was calibrated using 4.0, 7.0 and 10.0 pH buffer solutions. All samples were stored at 4 °C for later analysis. Before analysis, the samples were filtered using No. 1 Whatman® filter paper to remove any insoluble particles. While the experimental set up was conducted adhering to the well-established guidelines, the control test using synthetic water was not performed in this study due to the experimental limitations, albeit further study investigating control tests would be clearly desirable to identify if the dry deposition still affects the sample after 10 L disposal of rainwater. The standard analytical protocol following quality control and quality assurance was maintained throughout the experiments conducted in the Environmental Engineering Laboratory at CUET.

Statistical analysis

Principal component analysis (PCA) is an effective multivariate statistical method which is commonly applied to determine the influence of probable sources of contaminants present in rainwater samples (Palma *et al.* 2010; Machiwal & Jha 2015). PCA was used in this study to identify the possible sources of major species in rainwater. As this is the case for other relevant studies in the literature, PCA was performed to validate the outcomes of the correlation matrix which has been also performed within this work. PCA and correlation were undertaken using the IBM Statistical Package for the Social Sciences (SPSS) version 25. Microsoft Excel 2019 was used to conduct a descriptive statistical analysis of various parameters.

RESULTS AND ANALYSIS

Physicochemical analysis

In this study, the average concentration of all physicochemical parameters investigated was found to be well below the Bangladesh standard and WHO guideline in all selected land-use locations. The descriptive statistics of physicochemical parameters are tabulated in Table 1. The average pH values ranged between 6.07 and 6.75 in all samples tested, slightly in the acidic range. The EC and TDS were recorded higher in the industrial area, indicating the presence of more ions dissolved in the rainwater at that location. Turbidity was found in a similar range in all locations, though marginally higher in the commercial area due to the presence of dust particles in the air of that region. Even though turbidity is not a serious health threat, it may create a health hazard if the suspended particles contain or have adsorbed toxic organic or inorganic compounds (Kus *et al.* 2010). The alkalinity and hardness values in the studied locations ranged from 5 to 70 and 10 to 215 mg/L, respectively. These results are consistent with that of Amponsah *et al.* (2015), which was conducted in Colombia on a similar form of rainfall pattern when compared with the current study area. The average concentration of alkalinity and hardness was found to be somewhat higher in the commercial area.

Ionic composition and pH value

Natural rainwater is normally considered to be weakly acidic with a pH value of 5.6 when the atmosphere is free from pollution (Zhao *et al.* 2013; Xiao 2016). Here, the statistical variation of pH at different land-use locations is shown in Table 1. The maximum pH (8.66) was recorded in the residential (sub-urban) area, while in contrast, the minimum value (4.68) was found in the industrial area. Given the variability and anthropogenic input at the different sites, the residential catchment in Pahartali was found to be relatively clean with no surrounding input sources and exhibited a wide range of pH compared with other sites having urban, commercial and industrial surroundings. The relatively notable variation in urban, commercial and industrial catchments in comparison with sub-urban catchments illustrates site-specific characteristics influenced by the anthropogenic input (road traffic and industrial emissions) during precipitation followed by dry deposition between rain events. Similar variations have also been reported in other studies (Li *et al.* 2004; Despins *et al.* 2009).

Descriptive statistics for major ions in rainwater of the different locations are presented in Table 2. For the four sampling locations, SO_4^{2-} was discovered to be the highest dominant major ion, with an average concentration of 192.5, 274.7, 90.3 and 89.6 μeq/L, for industrial, commercial, residential (urban) and residential (sub-urban) areas, respectively. The maximum concentration of NO_3^- and SO_4^{2-} was found in the commercial area where vehicular emissions could have influenced rainwater quality compared with other locations. In the study area, vehicles are often congested which consequently increases vehicular emissions (Shamsher & Abdullah 2013). Likewise, the results of this study, the highest concentrations of NO_3^- and SO_4^{2-}, were also found by Alves *et al.* (2018) for Campo Bom, part of southern Brazil, which is characterised by commercial and industrial activities. Sulphate was found to be the most abundant ion among the investigated ions in all sampling locations (Figure 3). The contribution of NO_3^- and SO_4^{2-} across all the sampling locations was recorded high (above 90%) as shown in Figure 3. The percentage of NH_4^+ in the residential (sub-urban) area (Figure 3(e)) was recorded somewhat higher when compared with other locations (Figure 3(a)–3(d)), where agricultural activities were evident. In contrast, the highest mean concentration (7.2 μeq/L) of PO_4^{3-} was found in the industrial area. The presence of PO_4^{3-} in rainwater samples may indicate bird or insect faeces contamination as noted by Huston *et al.* (2012).

Trace metals concentration

A statistical summary of trace metal concentrations in the rainwater samples of the different sites is presented in Table 2. Here, it can be seen that the average concentrations of all selected trace metals fall well below the Bangladesh drinking water standard and WHO guidelines. In contrast, while an elevated concentration of Zn was found in all analysed samples, the limiting value of Zn was seen to be much higher compared with other metals. Comparatively, the higher concentration of Zn and Cu was found in the industrial area. The mean concentration of copper (Cu) was 74.7, 48.2 and 35.6% greater than the commercial, residential (urban) and residential (sub-urban) areas, respectively. Metal intensive activities usually influence industrial areas, along with the widespread use of metals, such as Fe, Cu and Zn in industries, thereby reflecting higher concentrations in rainwater (Wong *et al.* 2006; Joshi & Balasubramanian 2010; Hossen *et al.* 2021). The average concentration of Fe was recorded at 47.2, 36.7, 23.3 and 69.6 μg/L in industrial, commercial, residential (urban) and residential (sub-urban) areas, respectively, and Pb was only found in the rainwater samples of the commercial area. The highest average concentration (32.3 μg/L) of Cr was found in the commercial area, while the lowest average concentration (6.5 μg/L) was recorded in the residential (sub-urban) area. Mn was found in an elevated concentration in the commercial area, while Cd was found below the detection limit in all sampling sites. Notably, atmospheric deposition is one of the significant pathways of trace metals present in urban rainwater, mostly impacted by site-specific emissions (Joshi & Balasubramanian 2010). Since vehicular emissions in the commercial area are abundant compared with those of residential areas, this pathway

Table 1 | Descriptive statistics of physicochemical parameters of rainwater samples

	Industrial area				Commercial area				Residential (urban)				Residential (sub-urban)					
Parameter	**Min**	**Max**	**Avg**	**SD**	**Min**	**Max**	**Avg**	**SD**	**Min**	**Max**	**Avg**	**SD**	**Min**	**Max**	**Avg**	**SD**	**Bangladesh Standard[a]**	**WHO[b]**
pH	4.68	7.82	6.73	0.68	6.0	7.24	6.77	0.39	5.09	6.72	6.10	0.38	5.36	8.66	6.75	0.76	6.5–8.5	6.5–8.5
TDS (mg/L)	5.59	620	81.77	121.21	7.34	227	54.24	60.42	10.20	131.00	36.51	33.86	7.62	45.40	30.99	9.95	1,000	1,000
EC (μS/cm)	7.85	873	128.79	172.61	10.34	383	77.69	89.09	14.46	184.00	56.60	49.61	18.30	107.60	55.14	16.00	…	…
Alkalinity (mg/L)	7	20	12.76	3.23	5	70	26.16	18.48	6.00	15.00	9.11	2.32	20.00	35.00	24.79	4.62	…	1,000
Hardness (mg/L)	30	170	65.00	25.41	25	215	77.81	43.81	35.00	80.00	52.50	11.15	10.00	90.00	58.54	16.41	200–500	500
Turbidity (NTU)	0.4	11.31	2.75	2.40	0.51	8.51	3.66	2.21	0.67	1.81	1.22	0.31	0.41	2.93	1.47	0.62	10	5

Note: [a]Bangladesh drinking water quality standard, ECR'1997.
[b]World Health Organization drinking water quality standard.
'…' No standards available.

Table 2 | Statistical representation of each tested ion and trace metal in rainwater samples

Location\parameter		NO_3^-	SO_4^{2-}	PO_4^{3-}	NH_4^+	Fe	Cu	Zn	Pb	Mn	Cr	Cd
Industrial area	Min	BDL	BDL	0.6	BDL	BDL	BDL	110.0	BDL	1.0	2.0	BDL
	Max	87.9	812.5	20.5	24.1	440.0	620.0	630.0	BDL	25.0	94.0	BDL
	Avg.	25.5	192.5	7.2	3.9	47.2	82.8	240.0	BDL	8.4	29.2	BDL
	CV (%)	75.2	112.7	77.9	125.1	179.2	148.8	52.5		73.6	64.5	
Commercial area	Min	7.1	BDL	1.6	0.8	BDL	BDL	8.0	BDL	1.0	2.0	BDL
	Max	164.4	1,062.5	10.1	3.2	100.0	60.0	320.0	42.0	38.0	72.0	BDL
	Avg.	30.5	274.7	6.0	1.9	36.7	20.9	175.6	8.3	14.3	32.4	BDL
	CV (%)	107.4	106.3	71.2	64.4	57.0	63.5	42.1	194.1	72.3	54.1	
Residential (urban)	Min	BDL	BDL	0.6	0.8	10.0	BDL	80.0	BDL	1.0	4.0	BDL
	Max	51.9	270.8	12.6	4.0	40.0	480.0	530.0	BDL	15.0	52.0	BDL
	Avg.	16.4	90.3	3.9	2.0	23.3	42.9	219.3	BDL	5.5	18.8	BDL
	CV (%)	73.4	94.7	99.6	52.8	29.4	263.7	54.6		76.5	70.6	
Residential (sub-urban)	Min	BDL	BDL	0.3	0.8	20.0	10.0	60.0	BDL	1.0	2.0	BDL
	Max	50.0	333.3	25.9	26.6	320.0	310.0	570.0	BDL	12.0	13.0	BDL
	Avg.	15.2	89.6	4.0	5.7	69.6	53.3	192.1	BDL	6.1	6.5	BDL
	CV (%)	72.5	113.0	141.1	139.1	91.5	111.1	57.7		46.5	46.3	
Bangladesh Standard, ECR'97		800	8,300	190	…	300–1,000	1,000	5,000	50	100	50	5
WHO		800	5,000	…	…	…	200	…	10	…	50	3

Note: Unit of all ions are in μeq/L and trace metals in μg/L.
BDL stands for below detection limit; '…' No standards available.

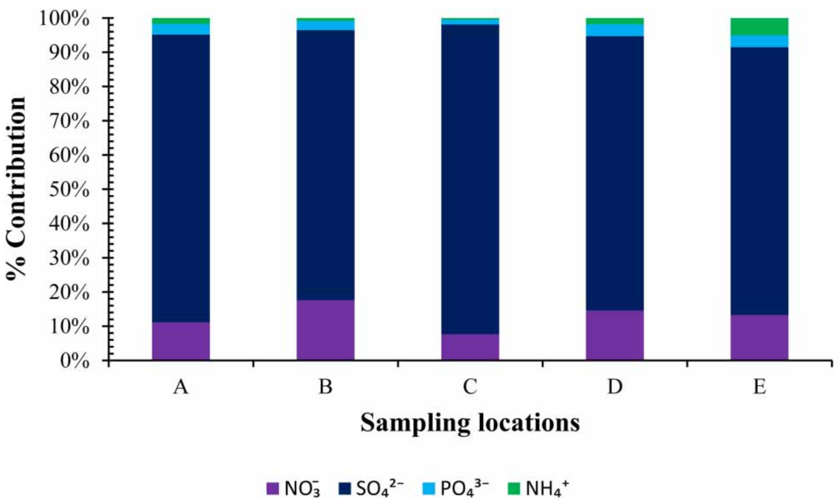


Figure 3 | Frequency distribution of major ions of rainwater samples: (a) industrial area, (b) commercial area (GEC), (c) commercial area (Agrabad), (d) residential (urban) and (e) residential (sub-urban).

(atmospheric deposition) could contribute to the elevated concentration of trace metals at the commercial site (Gunawardena *et al.* 2013).

Nevertheless, it was observed that the average concentration of trace metals in all analysed samples followed a decreasing order of Zn>Cu>Fe>Cr>Mn>Pb>Cd. The percentages of total concentration for selected trace metals in five sampling locations are shown in Figure 4. Zn was found as the most abundant trace metal compared with the investigated metals those measured in this work (Figure 4). Here, Cu and Zn together accounted for 80% of the total concentration in the industrial area. In comparison with the total concentrations of Cu and Zn in the industrial area, 70% accounted for the commercial area (GEC), 68% for the commercial area (Agrabad), 84% for the residential (urban) area and 74% for the residential (sub-urban) area.

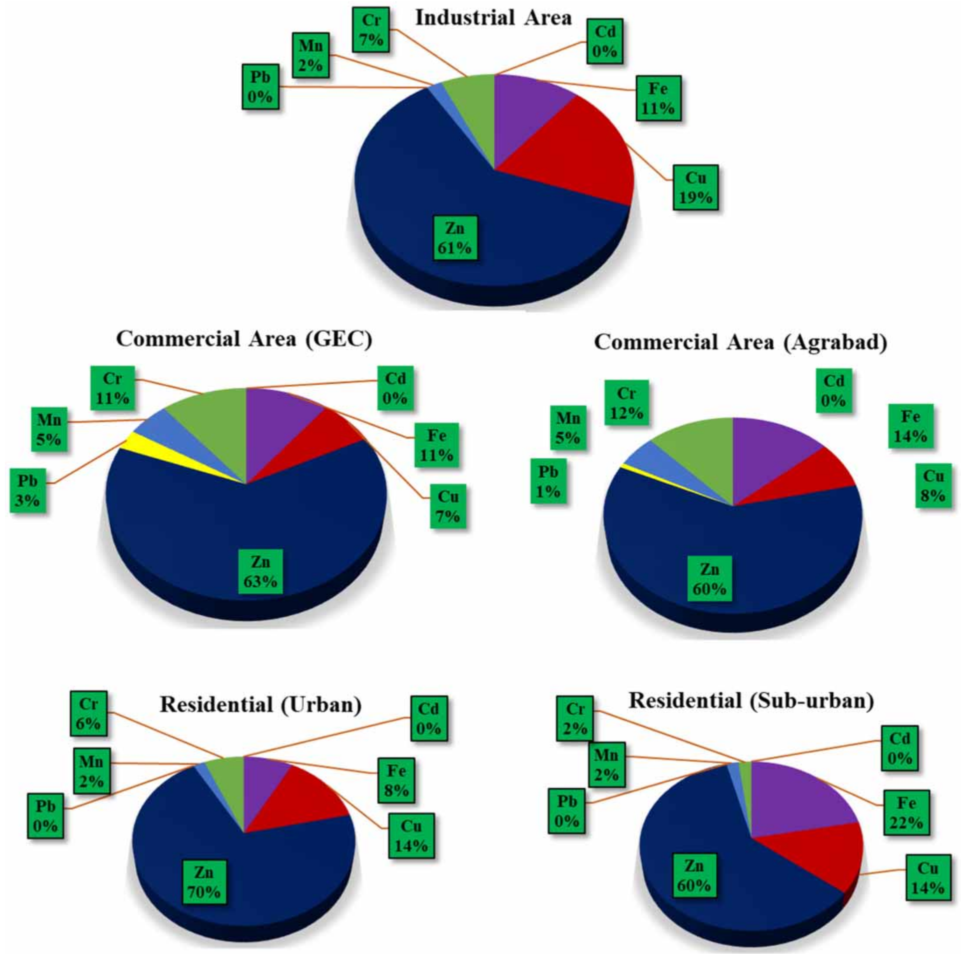


Figure 4 | Percentage contribution of each tested trace metal in rainwater samples, subjected to the different sampling locations.

Worldwide comparison

The precipitation analysis of this study was compared with the data collected from other areas of Bangladesh and worldwide. In Table 3, the physicochemical characteristics of rainwater measured for this study area were compared with those observed worldwide in other studies. The pH of rainwater samples was in the range between 4.68 and 7.82, with a mean value of 6.63 ± 0.62 (i.e., in the acidic range), which reveals similar characteristics as reported in other world regions. The concentrations of TDS and EC were found to be lower within this work as compared with those observed in other areas such as Sylhet (Bangladesh), Haryana (India), Loess Plateau (China) and Pereira (Colombia) but somewhat larger than Ayanfuri (Ghana), Jeju (Korea) and South-Western France, as reported in Table 3. The measured values of alkalinity and hardness for the tested area were higher compared with those reported in other parts of the world, except for Pereira (Colombia) (see Table 3). Similarly, the concentration of turbidity was seen to be much lower than those observed worldwide, but slightly higher compared with those in another city of Bangladesh (i.e., Sylhet), Colombia and France.

The differences and variability among different sites with respect to the city, country, region and worldwide are reflected in the results, which demonstrates the in-depth investigation prior to the collection of rainwater at the potential sites of this study. The mean concentrations of ions and trace metals in the study area for all samples are presented in Table 4 in addition to those reported for other regions globally. Overall, the concentration of nitrate (NO_3^-) was found to be much lower in the investigated area of this study in comparison with those observed in other sampling sites worldwide, but slightly higher than those observed for Ayanfuri (Ghana), as reflected in Table 4. The sulphate concentration (161.8 μeq/L) was observed to be slightly higher when compared with those reported in the literature for similar sampling locations globally, except for Tai'an

Table 3 | Comparison of physicochemical characteristics of rainwater between this study and worldwide

Parameter\ location	This study	Sylhet, Bangladesh (a)	Haryana, India (b)	Loess Plateau, China (c)	Ayanfuri, Ghana (d)	Jeju, Korea (e)	Pereira, Colombia (f)	South-western France (g)
pH	6.63 ± 0.62	7.6	6.85	7.48	6.62	5.2	7.96	6.5
TDS	52.3 ± 73.2	80	105	61.3	9.30	23.4	…	…
EC	81.3 ± 105.6	…	195	94.28	15.75	36	65.71	56.2
Alkalinity	19.3 ± 13.01	13.2	…	…	10.24	…	23.5	…
Hardness	65.3 ± 30.7	23	32	…	9.94	…	29.2	16
Turbidity	2.5 ± 2.02	0.56	11	4.5	5.98	4.8	1.45	2.4

Note: All units are in mg/L, except pH, EC (μS/cm) and turbidity (NTU); '…' No data available.
(a) Alam *et al.* (2012); (b) Bharti *et al.* (2017); (c) Wu *et al.* (2017); (d) Amponsah *et al.* (2015); (e) Moon *et al.* (2012); (f) Morales-Pinzón *et al.* (2015); (g) Vialle *et al.* (2012).

Table 4 | Comparison of ions (μeq/L) and trace metals (μg/L) in rainwater between Chattogram, Bangladesh and other locations

Parameter	This study	Ghore El-Safi, Jordan (a)	Oleiros, Spain (b)	Kolkata, India (c)	Ayanfuri, Ghana (d)	Tai'an, China (e)	Brisbane, Australia (f)	Juiz de Fora, Brazil (g)
NO_3^-	21.9 ± 22.9	67.3	31.5	202	16.77	64.3	25.8	28.7
SO_4^{2-}	161.8 ± 220.5	112.4	72.5	65	314.5	238.2	33.3	8.0
PO_4^{3-}	5.3 ± 5.2	…	…	…	8.85	…	3.16	…
NH_4^+	3.4 ± 5.7	75.4	32.5	…	…	167.1	…	…
Fe	45.1 ± 56.2	430	11.4	14	510	41.2	68	…
Cu	48.5 ± 85.6	73	2.1	3.7	…	7.9	21	89.9
Zn	203.7 ± 107.9	210	55.7	26	…	85.2	770	…
Pb	5.1 ± 12.9	66	0.51	0.68	270	5.9	5.4	11.8
Mn	9.2 ± 7.7	…	6.4	7.2	280	14.2	8.7	49.7
Cr	22.7 ± 18.0	3.1	0.28	0.71	…	0.06	0.53	…
Cd	BDL	52	…	0.12	120	0.61	…	0.17

Note: BDL stands for below detection limit; '…' No data available.
(a) Al-Khashman (2009); (b) Moreda-Piñeiro *et al.* (2014); (c) Majumdar *et al.* (2020); (d) Amponsah *et al.* (2015); (e) Li *et al.* (2011); (f) Huston *et al.* (2012); (g) Mimura *et al.* (2016).

in the Republic of China (238.2 μeq/L). The observed values of NH_4^+ are seen to be much lower in precipitation as reported here, compared with those observed in the sampling sites worldwide. The $PO_4{}^{3-}$ concentration was slightly lower than that of Ayanfuri (Ghana), but higher than those reported for Brisbane (Australia).

The trace metal concentrations reported in this study (Fe, Cu, Zn, Pb, Mn, Cr and Cd) overall were somewhat consistent, when compared with those reported worldwide (Table 4). It is evident from Table 4 that the concentration of Zn (203.7 μg/L) was one of the highest concentrations of trace metals in the study area, which was also considerably higher compared with those found in Spain, India and China, but lower than those observed in Jordan (210 μg/L) and Australia (770 μg/L). The Fe concentration (45.1 μg/L) was much lower in the investigated area when compared with those reported in Jordan, Ghana and Australia, but higher than those for Spain, India and China. The concentration of Cu (48.5 μg/L) within this study was moderately higher than that reported in Spain, India, China and Australia, but lower than that of Jordan and Brazil. The mean Pb value was found to be close to those observed in other sampling sites in Asia (i.e., India and China), but relatively much smaller than those in Jordan and Ghana. The data corresponding to Mn of this study were found to be consistent compared with worldwide observed values of Mn, however considerably smaller when compared with those observed in Ghana and Brazil. Overall, the concentration of Cd was reported lower in this study compared with the data worldwide. The Cr value was found to be somewhat higher compared with those reported globally.

Correlation matrix

The correlation matrix helps characterise the relationship among the species present in rainwater samples. To investigate the relationship with the rainwater quality parameters, Spearman's rank correlation analysis was used as shown in Figure 5. As evidenced in the graph, EC is a comprehensive indicator of the TDS in precipitation, which offers a robust correlation with TDS ($r = 1.0$). Recent work by Valappil *et al.* (2020) also confirmed that the similar correlation exists in between EC and TDS in the tropical rainforest region of northwestern Borneo. Measurements (Figure 5) also demonstrate that sulphate and phosphate are inversely related to pH (–0.57), implying that as sulphate and phosphate concentrations increase, pH reduces. Consequently, in some cases, the acidity of rainwater is supposed to increase. Since there is no apparent correlation between the pH value and NO_3^- while a relationship is present in NH_4^+ and NO_3^- indicating that NO_3^- in the rainwater sample is present as salt (NH_4NO_3) (Salve *et al.* 2008). A relationship between NH_4^+ and NO_3^- (–0.42) is indicated as expected since fertilisers often use both these components (Xiao 2016). Nitrates may potentially transfer from the air to various water sources (e.g., ground, lakes and surface water) using rainwater as the pathway (Fenn *et al.* 2013; Hoseinzadeh *et al.* 2016). In a sub-urban site in Spain, Moreda-Piñeiro *et al.* (2014) revealed that anthropogenic behaviours such as the use of NH_4NO_3 fertilisers, coal combustion and vehicle exhausts are the major sources of NO_3^- in the atmosphere.

In Figure 5, data showed that SO_4^{2-} mostly controls pH and PO_4^{3-} rather than SO_4^{2-} and NO_3^-. This result highlights that there is a presence of a somewhat noteworthy correlation among pH, SO_4^{2-} and PO_4^{3-} (–0.57). A relationship between NO_3^- and SO_4^{2-} (0.48) is also present in the samples, whereas PO_4^{3-} and NO_3^- (0.74) and PO_4^{3-} and SO_4^{2-} (0.68) exhibit strong associations. Concerning the source apportions, it has been shown that combustion processes along with the use of fuel oil with a sulphur content that occurs in industry and thermoelectric power plants are the primary sources of SO_4^{2-} and NO_3^- (Báez *et al.* 2007; Salve *et al.* 2008). Nitrate and phosphate originate from the decomposition of rocks and minerals, atmospheric deposition, agricultural and industrial activities (da Conceição *et al.* 2013). A strong correlation between SO_4^{2-} and NH_4^+ (–0.56) indicates that the available NH_3 in the atmosphere may principally react with H_2SO_4 to form $(NH_4)_2SO_4$ and NH_4HSO_4, as referred by Seinfeld & Pandis (2016).

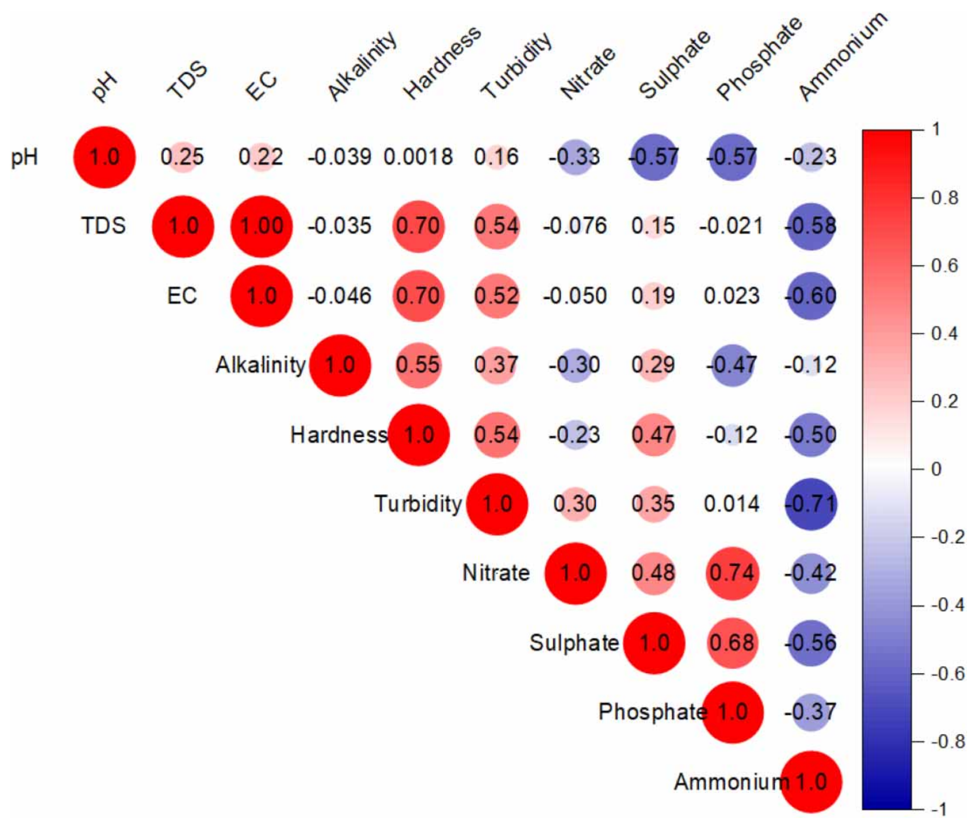


Figure 5 | Correlation matrix of important rainwater quality parameters.

Principal component analysis

The PCA of the rainwater quality parameters (see Table 5) showed three PCs with eigenvalues greater than 1 explaining about 69% of the data variability. The parameters which were correlated significantly in the correlation analysis also show strong loading in the PCA analysis. The three components were rotated using a Varimax rotation procedure.

As shown in Table 5, the first component (PC1) describes 36% of the total variance, revealing strong loading among TDS (0.943), conductivity (0.930) and hardness (0.819); also, moderate loading between pH (0.503) and turbidity (0.730). This clustering of variables points to a common origin, which in the case of rainwater, is likely related to environmental conditions. For example, recent studies (Rusydi 2018; Valappil *et al.* 2020) on rainwater concluded that TDS, EC and turbidity are physical parameters that contributed by the suspended particles in the atmosphere influenced from both natural and anthropogenic activities. The second component (PC2) consisting of nitrate (NO_3^-), sulphate (SO_4^{2-}) and phosphate (PO_4^{3-}) accounts for approximately 21% of the total variance. Here, PC2 reveals a strong loading of 0.839, 0.890 and 0.837 of the photochemical species NO_3^-, SO_4^{2-} and PO_4^{3-}, respectively. Anthropogenic activities are major sources of NO_3^- and PO_4^{3-} pollution in rainwater as discussed earlier and also reported by Fung & Lau (1998). NO_3^- and SO_4^{2-} ions also define anthropogenic influences, such as incomplete fuel combustions, automobile exhaust, coal combustion and industrial emissions, as discussed by Zhang *et al.* (2007) and Xiao (2016).

The third and final component, as shown in the figure, represents 12% of the total variance, with loadings of 0.422, 0.773 and 0.658 are pH, alkalinity and NH_4^+, respectively. The existence of NH_4^+ in the atmosphere may attribute to the utilisation of fertilisers, volatilisation of animal waste, human excreta, cattle farming, and from the burning of fossil fuels (Bouwman *et al.* 2002; Migliavacca *et al.* 2012). As noted by Colt *et al.* (2009), insufficient alkalinity could lead to incomplete nitrification and lower pH values in certain locations.

CONCLUSIONS

Detailed measurements were undertaken in this study to investigate the quality of rainwater across different locations in the south-eastern region of Bangladesh. Based on the work and findings of this study, the following conclusions are drawn:

1. The mean concentration of all tested physicochemical parameters, ions and trace metals in rainwater was found to be significantly lower compared with the drinking water quality standard of Bangladesh and WHO guidelines.
2. The concentration of nitrate (NO_3^-) was found to be much lower in the investigated area in comparison with those observed in other sampling sites worldwide. For the sampled locations, the mean concentration of nitrate (NO_3^-) and sulphate (SO_4^{2-}) was recorded as the highest in the commercial area. The observed values of NH_4^+ within this study were much lower in precipitation compared with those observed in the sampling sites from around the world.

Table 5 | Varimax rotation for PCA of selected parameters

Variables	PC1	PC2	PC3
pH	0.503	−0.196	0.422
TDS	0.943	0.129	−0.087
Conductivity	0.930	0.193	−0.066
Alkalinity	0.105	−0.100	0.773
Hardness	0.819	0.104	0.142
Turbidity	0.730	0.126	−0.003
NO_3^-	0.141	0.839	0.056
SO_4^{2-}	0.068	0.890	−0.025
PO_4^{3-}	0.142	0.837	−0.081
NH_4^+	−0.108	0.098	0.658
Eigenvalue	3.62	2.06	1.19
% of variance	36	21	12
Cumulative variance (%)	36	57	69

3. The trace metals concentrations reported in this study (Fe, Cu, Zn, Pb, Mn, Cr and Cd) were overall consistent, when compared with those reported worldwide. The mean concentration of trace metals in rainwater was exhibited in the following order: Zn>Cu>Fe>Cr>Mn>Pb>Cd in almost every sampling location.
4. Trace metals concentration, especially copper (Cu) and zinc (Zn), was predominant in the industrial area. The mean concentration of copper (Cu) in the industrial area was 74.7, 48.2 and 35.6% greater than the commercial area, residential (urban) and residential (sub-urban) area, respectively.
5. The PCA was used to determine the possible sources of major species in rainwater. The parameters that were grouped significantly in the correlation analysis also showed reliable loading in the PCA analysis. Hence, the results indicated that incomplete fuel combustion, industrial emissions, agricultural activities, and also environmental conditions were the dominant sources of the chemical composition in rainwater of the Chattogram region.

Prior to these experimental investigations, limited knowledge was available in the chemical composition of rainwater in the southern-eastern region of Bangladesh, which motivated the need for this study to be undertaken. Rainwater harvesting would be a sustainable approach to consider in the water system management for Bangladesh, given the gradual depletion of groundwater and deterioration of surface water quality. This study will provide a database of rainwater quality with varying land-use patterns, which will directly benefit stakeholders, local authorities and Government in planning and making decisions about rainwater use across the country but not limited to the study area. Additionally, the data and findings of this extensive study would be useful to engineers and researchers involved in the sustainable use of water resources across the world.

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CONFLICT OF INTEREST

The authors declare no potential conflict of interest regarding the publication of this work. In addition, the ethical issues including plagiarism, informed consent, misconduct, data fabrication and, or falsification, double publication and, or submission, and redundancy have been completely witnessed by the authors.

DATA AVAILABILITY STATEMENT

Data cannot be made publicly available; readers should contact the corresponding author for details.

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