Hydrogeochemical Characteristics, Quality Assessment and Health Impact Analysis of Groundwater for Drinking in the University of Chittagong, Bangladesh

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Groundwater is the major source of water supply for drinking in the Chittagong University (CU) campus. A comprehensive analysis, in terms of physico-chemical, geochemical, trace metals and biological parameters, was performed to ensure the suitability of groundwater for drinking. According to Piper, Chadha, Pie and Schoeller diagrams, Ca-Mg-HCO₃ type water was found both in winter and summer seasons. Gibb's plot specified that majority of the samples fell in the rock dominance. The multivariate analysis indicated that the hydrochemistry was governed by both the geogenic and anthropogenic sources. Comparison of physico-chemical factors, ionic compositions and trace metals results with standard values suggested that majority of the parameters were within the acceptable limit, except pH and Fe, for both seasons. Additionally, drinking water quality index (WQI) showed that groundwater could be categorized as 'excellent to good' rank in the 92% and 84% samples of summer and winter seasons, respectively. Though coliform were found in very few samples, however, no carcinogenic and non-carcinogenic health risk was observed. Collectively, CU campus groundwater is fit for drinking purpose. However, reduction of iron and removal of coliform are recommended. The present study may provide local authorities with insights into making rational decisions for sustainable groundwater management.

Key words: Chittagong University; drinking water; multivariate analysis; physico-chemical; trace metals; water quality index

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Water, an echo to life, undoubtedly, is an unavoidable and undeniable component on earth. It is considered as an important resource key element to sustain life on the planet [1]. 97% of water exists in oceans and is not drinkable, while 3% is regarded as fresh water where surface and ground water comprise about 0.3% [2]. Crucial for public health with the usage in drinking, domestic, and industrial purposes, affordability as well as safe water are basic rights and highly indispensable to our daily life [3,4]. About one third of the populations depends on groundwater for drinking purpose [5], however in Bangladesh, groundwater contributes approximately 98% to its population for potable purpose [6,7]. About 80% of diseases are caused by waterborne diseases, which lead to the deaths of 5 million people per year in Bangladesh [8]. About 6.5 to 24.4 million people are facing serious vulnerability because of contaminated water. Additionally, shortage of safe water in several regions of Bangladesh has worsened the scenario [6].

Groundwater is regarded as one of the prime sources for supplying water for industrial, domestic, and agricultural purposes not only in Bangladesh but also in the whole world. However, hasty urbanization, innate geochemical processes, and farming or agricultural activities are causing groundwater contamination. Day by day, the quality of groundwater is worsening, possessing high electrical conductivity, turbidity, total

hardness, salinity, trace metals, total dissolved solids, and even the presence of bacteria in different parts of Bangladesh [9]. Hence, regular monitoring and proper assessment are highly required in terms of physical, chemical, biological characteristics, and the presence of trace metals coupled with water evaluation indices, multivariate analysis, geostatistical modeling, and health risk assessment to save lives and the environment. Countries around the world are concerned over the quality of drinking water though their approach for providing safe drinking water to the citizens differ in a diverse array in terms of standard, pretreatment, posttreatment, residual disinfectants, distribution, and so on [10-25]. However, being a largely agriculture-based economy and lying in a delta region, Bangladesh warrants a great concern over the drinking water quality. researchers Therefore, many have evaluated quality different groundwater in Divisions (administrative headquarters which further split into several districts) of Bangladesh including Chittagong [26-32], Dhaka [33-40], Khulna [41-44], Rajshahi [28, 45-50], Sylhet [51], Rangpur [52], Barisal [35], and Mymensingh [34]. Commercial jarred and bottled water which are sold for drinking purposes in Bangladesh were also evaluated in terms of public health significance [53, 54]. Furthermore, the water ranking of Sylhet Agricultural University (SAU) [55] and Mawlana Bhasani Science and Technology University Campus of Tangail [56], as well as primary schools of Magura [57]. Rajshashi [58], Tangail [38], and Satkhira [44] were evaluated for suitability of consumption. However, most of the studies only evaluated the water quality parameters in terms of the concentration only, whereas a few of them analyzed the essential groundwater quality parameters coupled with multivariate analyses of true status [26, 27, 59, 35, 36, 40, 41, 43, 45, 47, 55]. Moreover, many of the studies analyzed only physicochemical parameters, including a few trace metals [26-30, 32, 34-36, 38, 40, 41, 43-47, 49-56, 60-64], microbiological perspective only [8, 39, 51, 65, 66], or only a few trace metals [26, 37, 47, 58], where the quality assessment of water in all perspectives were not revealed. The groundwater chemistry is an essential parameter for evaluating potential exploitable water of an aquifer [67]. Keeping in mind that geochemical processes, which control water quality of groundwater, are identified by the ion chemistry [68]. Piper diagram is useful for outlining hydro-chemical characteristics of groundwater [69]. Piper diagram is applied to investigate the geochemical analysis and quality of groundwater [70]. Chadha diagram, Gibbs plot, Schoeller diagram, and Scatter diagram are estimated for determining geochemical evolution [71, 72]. Variable applied math techniques like cluster analysis (CA) and Hydrogeochemical Characteristics, Quality Assessment and Health Impact Analysis of Groundwater for Drinking in the University of Chittagong, Bangladesh

principal component analysis (PCA) are also used for drawing information of covariance and relationship among the parameters [72]. CA links samples and parameters in groups through inter-sample similarities, while PCA reduces the dimensions of huge sets of data for better understanding [64, 73]. Pearson's correlation coefficient method is employed for determining correlation amid different criteria [65]. In addition, water quality index (WQI) is taken into appraise because it is the simplest technique of measurement of water quality [33]. It gives an overall water quality ranking or comparative overview [74]. From the large variation of WQIs, it is selected by assessing input variables and desired results [26]. Moreover, semivariogram and kriging models are useful for interpolating the dependence or autocorrelation among the spatial patterns of unceasingly varied parameters [68]. The non-carcinogenic and carcinogenic effects via ingestion of drinking water are given prime importance naturally.

Chittagong, dubbed as the commercial capital, has been facing an inadequate supply of drinking water [75]. The Chittagong University (CU) campus, a fastgrowing hilly suburb 25 km away from the port city of Chittagong, was chosen as our study area owing to its rapid infrastructural development [66], broadly unplanned solid waste disposals [75], close proximity to ship breaking coastal areas [76], rigorous fertilizerdependent agricultural activities on its various hilly premises, and unscientific laboratorial and medical waste disposal systems, among others. Needless to say, waste is forever with us, but our exposure to it should not be in a life-threatening way.

To the best of our knowledge, two literatures have been found on the evaluation of surface water at CU [65, 77], whereas a single study had been done on drinking water in the CU campus on the basis of bacteriological test only [65]. An integrated approach is highly required for the quality assessment of applying hydrogeochemical, GIS, and multivariate statistical methods. Considering the above-mentioned factors, the CU campus was chosen as our study area (Figure 1) and an integrated approach was employed to assess the water quality suitable for human consumption, and samples were collected in the winter (November, 2018 -February, 2019) and summer (April, 2019 – July, 2019) of 2018-2019. Taken together, this study aimed to evaluate the groundwater quality of the CU campus based on physico-chemical, trace metals, and microbiological parameters coupled with water quality indices, geostatistical modeling, multivariate analysis, and health impact analysis.

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Figure 1. Map of the study area, indicating sampling stations

MATERIALS AND METHODS

Study Area and Sample Collection

CU, with 1754 acres of hilly and flat areas, is regarded as the largest campus in Bangladesh. It has 27,839 students, 872 faculty members, and 2,046 non-academic staff (CU website). The study area extends from $91^{\circ}47'46.302''$ to $91^{\circ}47'6.042''$ E (longitude) and $22^{\circ}28'45.865''$ to $22^{\circ}28'7.614''$ N (latitude).

Groundwater is used for drinking purpose at the CU campus where generally pumps are used for collection. The pumped groundwater is stored in storage tanks and then supplied to the residential halls, dormitories, and family quarters. The depth of the wells at the CU campus is about 500 m (Engineering office, CU). Sterilized plastic and glass bottles are used to collect tap water according to APHA [78]. Methods used in the presented study are represented in Table 1.

Parameters	Methods/Instruments
Temperature, pH, Total Dissolved Substances (TDS), and	Combometer (Hanna portable combometer, Model no:
Electrical Conductivity (EC)	HI9813-6)
Salinity	Hand Refractometer (Model no: REF201/211/201bp)
Turbidity	Turbidity Meter (Model no: TU-2016)
Total Hardness (TH)	EDTA Titrimetric Method
Chloride	Argentometric Method
Free CO ₂ , Total Acidity (TA) and Bicarbonate (HCO ₃ ⁻)	Titrimetric Method
Nitrate (NO ₃ ⁻), Sulfate (SO ₄ ²⁻) and Phosphate (PO ₄ ³⁻)	Ultraviolet Spectrophotometer
Na K Ca and Mg	Inductively Coupled Plasma Optical Emission
Iva, K, Ca, and Mg	Spectrometry (ICP-OES)
Al, Fe, Mn, As, Cr, Pb, Cd, Hg, Co, Cu, Ni, Mo, Se, Ag,	Inductively Coupled Plasma Mass Spectrometry (ICP-
Zn, Sb, U, and Sn	MS)
Total Aerobic Count (TAC)	Pour Plate Method
Total Coliform Count (TCC) and Total Fecal Coliform (TFC)	Most Probable Number Method

Table 1. Methods followed in the present study

Physico-chemical Characteristics

Temperature, Total Dissolved Solids (TDS), Electrical Conductivity (EC), and pH were analyzed at the spot from where samples were collected. Temperature, TDS, EC, and pH were determined by a combometer (Hanna portable combometer, Model no: HI 9813-6), turbidity was determined by a turbidity meter (Model no: TU-2016), and salinity was determined by a hand refractometer (Model no: REF201/211/201bp). Total Hardness, chloride, and free CO₂ were determined by titrimetric methods according to APHA [78]. HCO₃⁻ was determined by titrating with HCl. Total acidity value was also determined by following the modified APHA protocol [78]. To assess total acidity, 50 mL of each sample was titrated with 0.02 N NaOH (V mL), using phenolphthalein as an indicator [79]. It was calculated using the following equation (1):

$$Total Acidity = \frac{V \times 0.02 \times 50 \times 1000}{Sample Volume}$$
(1)

An UV-visible spectrophotometer was used to determine nitrate (NO_3^{-}) [78] and phosphate (PO_4^{3-}) concentrations [80]. Sulfate (SO₄²⁻) concentration was determined by turbidimetric method with a little modification [78]. Briefly, a conditioning reagent (25 mL of glycerol + 15 mL of conc. hydrochloric acid + 50mL of isopropyl alcohol (95%) + 37.5 g of sodium chloride in a 250 mL volumetric flask) and a standard sulphate solution (1.479 g of sodium sulphate in 1L DW) were prepared. Then, 0, 10, 20, 30, and 40 mL of the standard solution and 5 mL of the conditioning reagent were added into five different 100 mL standard flasks to make the calibration solutions. A pinch of barium chloride was also added in the solutions. For the sample preparation. 20 mL of a sample with all other reagents (mentioned above to prepare standard solution) were added into a 100 mL standard flask. Then, sulfate concentration was determined at the wavelength of 420 nm.

Trace Metals Determination

Na, K, Mg, and Ca were measured by Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES: Shimadzu 9820) following a method explained elsewhere [81]. Different wavelengths were used: Na at 588.983 nm, K at 7666.455 nm, Ca at 315.880 nm, and Mg at 279.071 nm. Al, Fe, Mn, As, Cr, Pb, Cd, Hg, Co, Cu, Ni, Mo, Se, Ag, Zn, Sb, U, and Sn were monitored by Inductively Coupled Plasma Mass Spectroscopy (ICP-MS: 7900 Agilent ICP-MS, Agilent Technologies Inc.). EPA (200.8) method was followed for the preservation as well as to determine metal contents [82]. Hydrogeochemical Characteristics, Quality Assessment and Health Impact Analysis of Groundwater for Drinking in the University of Chittagong, Bangladesh

A calibration standard solution was prepared by mixing a standard solution and a reagent containing 1% (v/v) nitric acid. The R² value was > 0.999 for the calibration curve of each trace metal. The water samples were digested with 1% nitric acid. The *CRM* accuracy and spiking recovery were within 80-120%.

Microbial Test

Pour plate technique was followed to assess Total Viable Count (TVC) [83]. Samples were inoculated onto nutrient agar medium in duplicates and incubated (37° C, 24 to 48 hours). Colony meter was used to count colonies. For determining total coliform count (TCC) and total fecal coliform (TFC), most probable number (MPN) method was employed using fermentation tube [83]. 0.1, 1, and 10 ml samples were inoculated onto the three rows of the test tubes, respectively. In the first and second row of test tubes, 10 ml of single-strength MacConkey broth was added, and in the third row double-strength medium was used. Incubation was done at 37° C for TCC and 45° C for TFC. After 24 h, tubes were considered as positive for coliforms if they contained acid and gas.

Geostatistical Modeling

For the evaluation of structural analysis and spatial interpolation, semivariogram models are established. Kriging is used for the integration of spatial correlation and the dependence to predict a known variable which follows a well-established methodology [40]. There are various kriging methods (ordinary, universal, simple etc.), among which ordinary kriging (OK) was found to be the most accurate during calculation and prediction [84]. ArcGIS, version 10.5 was utilized for geostatistical modeling.

Statistical Analysis

Descriptive statistical analysis, Pearson's correlation, PCA, and CA were calculated by SPSS-23.0. Piper and Schoeller diagrams were plotted using Grapher 14. Gibbs, Chadha, Pie and Scatter diagrams were plotted by MS Excel-16. Spatial analysis was modelled by ArcGIS 10.5.

Water Quality Index (WQI)

WQI was evaluated by applying well established method known as weighted arithmetic [85].

Health Risk Assessment (HRA)

To assess health risk, US EPA (1989) guidelines were followed. Health Quotient (HQ) was estimated for Fe,

As, Cr, and Mn only as concentrations of other trace metals were too low. HQ was calculated using the following equation (2):

$$HQ = \frac{CDI}{RfD}$$
(2)

RfD is the reference dose (mg/kg/day) of a trace metal as suggested by *US EPA* [86]. *CDI* stands for chronic daily intake (mg/kg/day), which was calculated using the following equation (3):

$$CDI = \frac{C \times IR \times EF \times ED}{BW \times AT}$$
(3)

Where, *C* is the concentration of a trace metal in groundwater (mg/L); *IR* is the human water ingestion rate (L/day); *ED* is the exposure duration (years); *EF* is the exposure frequency (days/year); *BW* is the average body weight in (kg); and *AT* is the averaging time (AT= $365 \times ED$).

For HQ < 1, no significant health risk of noncarcinogenic effects. However, for HQ values more than unity (HQ > 1), non-carcinogenic health risk is anticipated [87]. Hazard index (HI) was calculated for HRA which is the summation of the HQ values of Fe, As, Cr, and Mn. HI value less than unity (HI < 1) indicates a lower possibility of undesirable noncarcinogenic health effects in case of lifetime exposure. For HI > 1, unexpected non-cancer health effects are evident [88]. Cancer risk (CR) assessment was done using the following equation (4) [89].

$$CR = CDI \times CSF \tag{4}$$

CSF (Cancer slope factor) values for As and Cr are 1.5 and 0.5 mg/kg/day, respectively (*US EPA's* Integrated Risk Information System-*IRIS*). *CR* value between 10^{-4} and 10^{-6} is considered as the acceptable *CR*

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[88]. From our list (Fe, As, Cr, and Mn), carcinogenic trace elements (As and Cr) were considered for determining *CSF*.

RESULTS AND DISCUSSION

Physico-chemical Characteristics

A comparative statistical evaluation of different physico-chemical parameters of drinking water in the CU campus during winter and summer is shown in Table 2 with BSTI and WHO standards. During the winter, the mean temperature was 24.26 ± 0.25 °C, varying from 23.10 to 26.20°C, while during the summer the mean temperature was 29.76 \pm 0.28°C, varying within the range of 29.30-30.20°C, indicating that all samples remained within the range of BSTI and WHO prescriptions (20-30°C). TDS is the characterization of the presence of organic and inorganic matter [90]. During the winter, the range of TDS lay within 44.0 to 90.70 mg/L with the mean value of 56.42 ± 4.88 mg/L, while the values varied from 37.33 to 87.0 mg/L with the average value of 49.0 ± 15.05 mg/L during the summer. During both seasons, TDS was within the acceptable limits of BSTI (1000 mg/L) and WHO (500 mg/L) (Table 2). Water samples in all the sampling stations were found as freshwater type having the mean concentration of TDS less than 1000 mg/L [91]. Pure water is a good insulator and has poor conductivity. The mean EC was $63.33 \pm 6.67 \,\mu$ S/cm during the winter and $60 \pm 24.12 \,\mu\text{S/cm}$ during the summer, ranging from 40.0 to 110.0 μ S/cm and 40.0 to 120.0 μ S/cm during the winter and summer, respectively. In both seasons, EC values were within the limit (500 μ S/cm). EC values less than 1000 μ S/cm are classified as very weakly mineralized water defined by Detay et al. [92] and EC values of 0-250 μ S/cm indicate low salinity according to WHO [91].

Table 2. Descriptive statistics for drinking water quality at the CU campus during winter and summer

Parameters	Unit	Winter	· (n=12)	Summe	er (n=12)	WHO standards	BSTI standards
i urumeters		Range	Mean ± SD	Range	Mean ± SD		
Temperature	(°C)	23.10-26.20	24.26±0.25	29.30-30.20	29.76±0.28	20.0-30.0	20.0-30.0
TDS	(mg/L)	44.0-90.70	56.42±4.88	37.33-87.0	49.0±15.05	500.0	1000.0
EC	(μ S/cm)	40.0-110.0	63.33±6.67	40.0-120.0	60.0±24.12	500.0	500.0
Turbidity	(NTU)	0.26-11.46	2.04±0.91	0.28-9.64	3.08±3.48	5.0	10.0
pH	-	5.70-6.70	6.26±0.11	5.80-6.80	6.36±0.33	6.5-8.5	6.5-8.5
Salinity	(ppt)	0.0	0.0	0.0	0.0	-	-

TH	(mg/L)	14.0-42.0	21.78±2.87	12.0-44.0	20.33±9.18	300.0	200.0-500.0
ТА	(mg/L)	4.0-28.0	13.0±2.10	6.0-50.0	20.0±13.91	-	-
Free CO ₂	(mg/L)	4.40-17.60	7.52±1.10	6.60-24.20	14.30±5.56	-	-
Chloride	(mg/L)	0.95-8.84	3.70±0.60	2.0-8.33	4.44±1.97	250.0	150.0-600.0
Nitrate	(mg/L)	0.0-1.78	1.13±0.17	0.0-1.81	1.18 ± 0.68	45.0	10.0
Sulfate	(mg/L)	0.01-0.06	0.03 ± 0.02	0.01-0.04	0.03 ± 0.01	250.0	400.0
Phosphate	(mg/L)	0.01-0.05	0.03±0.01	0.01-0.04	0.03 ± 0.01	0.5	6.0
Bicarbonate	(mg/L)	14.0-50.30	25.61±11.75	15.30-40.90	27.06±8.30	200.0	-
Na	(mg/L)	4.45-15.84	9.55 ± 2.96	5.16-15.14	9.58 ± 2.72	200.0	200.0
K	(mg/L)	0.54-1.05	0.78 ± 0.17	0.49-1.0	0.77 ± 0.17	12.0	12.0
Ca	(mg/L)	2.45-8.89	5.81 ± 2.26	2.68-8.23	6.0 ± 2.21	200.0	75.0
Mg	(mg/L)	1.87-6.0	3.75 ± 1.48	1.34-6.19	3.67 ± 1.56	150.0	30.0-35.0
Al	(µg/L)	1.31-8.13	3.03 ± 2.02	1.01-11.50	3.0 ± 2.80	200.0	200.0
Fe	mg/L	0.79-1.87	1.14 ± 0.35	0.71-1.65	1.14 ± 0.30	0.30	0.3-1.0
Mn	(µg/L)	0.40-88.23	29.67 ± 23.22	1.33-75.13	22.17 ± 20.13	100.0	100.0
As	(µg/L)	0.01-0.84	0.12 ± 0.24	0.03-0.73	0.12 ± 0.20	10.0	50.0
Cr	(µg/L)	0.0-2.55	1.36 ± 0.98	0.0-2.51	1.48 ± 0.84	50.0	50.0
Pb	(µg/L)	0.53-3.28	1.27 ± 0.82	0.49-6.10	1.99 ± 2.08	50.0	10.0
Cd	(µg/L)	0.03-2.85	0.42 ± 0.80	0.02-2.06	0.46 ± 0.65	3.0	5.0
Hg	(µg/L)	0	0	0	0	1.0	1.0
Co	(µg/L)	0.01-1.19	0.28 ± 0.32	0.07-1.61	0.58 ± 0.55	50.0	50.0
Cu	(µg/L)	0.37-10.27	2.38 ± 2.75	0.39-13.10	3.47 ± 4.24	1000.0	1000.0
Ni	(µg/L)	0.0-4.90	2.83 ± 1.54	0.0-6.0	3.31 ± 2.09	70.0	100.0
Мо	(µg/L)	0.0-0.01	$\begin{array}{c} 0.0007 \pm \\ 0.0030 \end{array}$	0.0-0.43	0.07 ± 0.14	10.0	-
Se	(µg/L)	0.0-0.80	0.16 ± 0.22	0.0-0.38	0.10 ± 0.13	10.0	10.0
Ag	(µg/L)	0	0	0	0	100.0	20.0
Zn	(µg/L)	8.23-262.18	60.32 ± 73.78	4.69-137.40	26.25 ± 38.53	3000.0	5000.0
Sb	(µg/L)	0.01-0.15	0.04 ± 0.04	0.02-0.15	0.04 ± 0.04	20.0	-
U	(µg/L)	0.003-0.025	0.014 ± 0.006	0.003-0.020	0.014 ± 0.004	30.0	-
Sn	(µg/L)	0.53-0.62	0.57 ± 0.03	0.47-0.83	0.58 ± 0.10	-	2.0

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Lower EC indicated a lower dissolution of aquifer minerals, salinity intrusion, and nutrient content. Turbidity varied from 0.26 to 11.46 NTU during the winter and 0.28 to 9.64 NTU during the summer, with corresponding means of 2.04 ± 0.91 and 3.08 ± 3.48

NTU, respectively. All the values were within allowable limits (5.0 and 10.0 NTU for WHO and BSTI, respectively), except one location (SS-10) in both seasons and two locations (SS-10 and SS-12) in the winter that exceeded the acceptable limits, which might

be due to the aggregation of dissolved constituents in the water storage tank and also in the supply line. Furthermore, it could be stated that there is no seasonal effects as the values that exceeded the limits were found at SS-10 in the summer. Consuming a highly turbid water would be a potential health risk, which makes it the foremost drinking water quality parameter [93]. pH has very little direct impact on human health [94]. During the winter, the range of pH was 5.70 to 6.70 and the mean value was 6.26 ± 0.11 , while in the summer the range was 5.80 to 6.80 and the mean value was 6.36 \pm 0.33. Water in this area is slightly acidic where the mean pH values for both seasons were very close to minimum values (6.5) of the permitted range reported by WHO and BSTI. Acidic pH is possibly derived mainly from free CO_2 and HCO_3^- concentrations [95, 96]. Conversely, recharge of infiltration from acidic rainwater, oxidation of pyrite in sediments [97], presence of clay minerals, and temperature effect cannot be ruled out at this moment [98]. Nearly neutral water was previously reported for the supply water of Chittagong city, which is comparable to our findings [99]. Water which is acidic or weakly acidic in nature indirectly indicates the presence of very low amounts of inorganic sediments or domestic and agricultural wastes [100]. Nonetheless, acidic water contributes to corrosion of metals coupled with disinfection efficiency, which is responsible for indirect effects on humans [101]. The mean values of total acidity were found to be 13 ± 2.10 and 20 ± 13.91 mg/L in the winter and summer with the range of 4.0 to 28.0 mg/L and 6.0 to 50.0 mg/L, respectively. Free CO₂ concentration is responsible for acidity of water by increasing hydrogen ions in water (Equation 7) and is able to liquefy some elements [102] (Ca, Mg, Na, K, Si) and other species (HCO₃⁻). Here, the content of free CO₂ varied from 4.40 to 17.60 mg/L and 6.60 to 24.20 mg/L with the mean values of 7.52 ± 1.10 and 14.30 ± 5.56 mg/L in the winter and summer, respectively.

$$CO_2 free + H_2O \leftrightarrow H_2CO_3 \leftrightarrow H^+ + HCO_3^-$$
 (7)

Salinity in drinking water may cause hypertensive disorders [65]. Zero salinity (0.0 ppt) was found from all of the sampling stations for both seasons. Though Chittagong is a coastal region, nevertheless saline surface water intrusion has not occurred in the presented area. The mean values of total hardness (TH) were found to be 21.78 ± 2.87 mg/L and 20.33 ± 9.18 mg/L and the ranges were 14.0 to 42.0 mg/L and 12.0 to 44.0 mg/L during the winter and summer, respectively. The results indicated that the water of the CU campus is soft (<75 mg/L) in nature according to classification of TH reported elsewhere [103]. Our findings were more or less agreed with the previous studies for the groundwater of Chittagong City [75, 99], whereas largely differed from Chittagong EPZ [104] and ship breaking area

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(Sitakundo) of Chittagong due to serious pollution [105]. Noteworthy, insignificant temporal variations were observed in this study.

Ionic Composition of Water

The analysis of several cations and anions is represented in Table 2. Na⁺, K^+ , Ca^{2+} , Mg^{2+} , and Al^{3+} were obtained as the major abundant cations in the study area. Among them, Na⁺ contributed the highest concentrations (9.55 \pm 2.96 and 9.58 \pm 2.72 mg/L for the winter and summer, respectively) but lay below the acceptable limit (200 mg/L). In the human body, it is an obligate nutrient for some necessary functions like nerve and muscle functions [106]. Ca²⁺ and Mg²⁺ control the hardness of water [107]. The mean content of Ca^{2+} and Mg^{2+} was 5.81 ± 2.26 and 3.75 ± 1.48 mg/L in the winter, and 6.0 \pm 2.21 and 3.67 \pm 1.56 mg/L in the summer, which were within the prescribed limits set by WHO (200 and 150 mg/L) and BSTI (75 and 150 mg/L) as well. Therefore, the water was found as soft, caused by the low amounts of Ca^{2+} and Mg^{2+} . The values of K^+ and Al^{3+} were also found within acceptable range (Table 2). HCO₃⁻, Cl⁻, NO_3^{-} , SO_4^{2-} , and PO_4^{3-} were found as the major anions of this study area. Among them, the concentration of HCO_3^- ion was higher. The mean content of HCO_3^- was 25.61 ± 11.75 and 27.06 ± 8.30 mg/L in the winter and summer, respectively, which was also within the prescribed limit fixed by WHO (200 mg/L). Carbonate weathering and carbonic acid dissolution could be the most probable reason for it [108]. If chloride concentration in water exceeds 100 mg/L, it is regarded as saline water or brackish [109]. In the CU campus, chloride was much lower $(3.70 \pm 0.60 \text{ mg/L} \text{ for winter})$ and 4.44 ± 1.97 mg/L for summer) than this value. Nitrate is considered as a disease causing parameter like hypertension in adults and blue baby syndrome in infants [92], however the concentration of nitrate was found within the permissible range. Very insignificant mean concentrations of sulfate and phosphate were detected in both seasons. The abundance of ions in the drinking water could be sequenced as: Na⁺, Ca²⁺, Mg²⁺, K⁺, and Al³⁺ for cations, and HCO₃⁻, Cl⁻, NO₃⁻, SO₄²⁻, and PO4³⁻ for anions. It is hypothesized that the low contents of Na⁺, Ca²⁺, Mg ²⁺, HCO³⁻, and Cl⁻ in the drinking water are explained through the lack of salinity burden from the sea, comparatively lower extraction, moderate to heavy rainfall due to tropical monsoon climate in Chittagong, or due to low rock weathering [110]. Moreover, temporal variations were found insignificant in our study.

Trace Metals

Fe, Mn, As, Cr, Pb, Cd, Hg, Co, Cu, Ni, Mo, Se, Ag, Zn, Sb, Sn, and U were measured in this study (Table 2). The mean concentrations of Fe were 1.14 ± 0.35 mg/L in the

winter and 1.14 ± 0.30 mg/L in the summer, while the ranges were 0.79-1.87 and 0.71-1.65 mg/L, respectively. In both seasons, Fe values exceeded the acceptable limits set by WHO and BSTI. It is hypothesized that Fecontaminated water which is used for drinking, domestic, and cooking purposes has the potential to cause human health hazards. Fe in groundwater is unfavorable due to taste of the water, also to cause staining. High concentrations of Fe may lead to stained clothes, discolored plumbing fixtures, hence corroded, and sometimes add an "oxidized" taste [111]. Excessive iron content can damage cell structure by producing free radicals and leads to high risks of carcinogenesis [112]. Furthermore, high concentrations of iron can lead to nausea, diarrhea, and cardiovascular diseases; even liver and kidney might be affected (hemochromatosis, cirrhosis, and arrhoithmias) [44]. Recently, Fe content out of the acceptable limit had also reported in Chittagong [44]. The high concentration of iron can be anticipated by the dissolution of rocks and ferruginous minerals and domestic and industrial sewage effluents [113]. Groundwater of Bangladesh is contaminated with arsenic, which poses a threat to health of many areas [114, 115]. However, As concentrations in this study area were very low for both seasons. On the other hand, Hg, Mo, and Ag were not detected in our investigation. All other trace metals were found in very small quantities and within permissible limits according to Hydrogeochemical Characteristics, Quality Assessment and Health Impact Analysis of Groundwater for Drinking in the University of Chittagong, Bangladesh

WHO and BSTI standards (Table 2), which indicates the water is safe for drinking. The mean content of trace downward metals followed orders: Fe>Zn> Mn>Ni>Cu>Cr>Pb>Sn>Cd>Co>Se>As>Sb>U during the winter and during the summer it was Fe>Zn>Mn>Cu>Ni>Cr>Pb>Co>Cd>As>Se>As>Sb >U. Insignificant temporal variations were observed in terms of mean concentration. Taken together, the water is safe for drinking purposes in terms of physicochemical parameters, giving slight concerns for acidic pH and excess iron concentrations.

Microbiological Assessment

For microbiological assessment, five sampling stations were randomly selected. TVC (cfu/ml) detected in the water samples are presented in Figure 2. Significantly higher TVC count was recorded during the summer at sites SS-06, SS-08 and SS-11 compared to the winter.

The possible reason for higher TVC during the summer may be due to the enzymatic activity of microorganisms for heavy proliferation which increases at elevated temperatures [116]. Followed by TVC, TCC count was also higher in the summer at SS-08 (43 MPN/100 mL). TFC was found at SS-06 and SS-08 in the summer only.

Table 3. Permissible bacterial count limits of according to WHO and BSTI standards

Parameters	WHO standards	BSTI standards
Total viable count (TVC)	1×10 ³	0
Total coliform count (TCC)	0	0
Total fecal coliform (TFC)	0	0



Figure 2. Results of microbiological analyses of water samples at the CU campus during the winter and summer. (Units of TVC, TCC and TFC are cfu/ml, MPN/100 ml, and MPN/100 ml, respectively)

The TFC values of these sampling stations were 04 and 23 MPN/100 mL, respectively (Figure 2), which exceeded the permissible limits (Table 3) according to WHO and BSTI standards, and water from these sample stations was not suitable for drinking purposes. Increased temperature might be the prime reason for this high TCC and TFC values [114]. Total fecal coliform could also have been originated from environmental sources such as organic matters, soils or biofilms.

Geochemical Classification and Ionic Composition

A piper diagram is a tool used for hydro-geological studies with visualized graphical representation [118–120]. In the diagrams presented in Figure 3, no dominant

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cation was found in the cation ternary diagram (left) for both seasons, while HCO₃⁻ was the only dominant anion (right). Importantly, from the central diamond shape diagram Ca-Mg-HCO₃ type of water was found in both seasons (Figure 3). This kind of water in the study area could be present as a temporary hard water. This could be attributed to rock-water inter-relationship [59].

Chadha proposed a classification diagram for the hydrochemical analysis of natural water [121]. From the Chadha diagram in Figure 4, Ca-Mg-HCO₃ type of water was likely to be dominant during both seasons, except only one sample in the winter and three samples in the summer.



Figure 3. Piper trilinear diagram illustrating chemical compositions of water samples



Figure 4. Chada diagram demonstrating hydrochemical classifications of water samples

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Figure 5. Pie diagrams of median values of major ions in the summer (a, b) and winter (c, d)

The Chadha plots are likely to support the obtained data from the piper diagrams significantly. On the other hand, Cl⁻ and NO₃⁻ surpassed 19% and 3%, respectively, in both seasons. Overall, pie charts (Figure 5) showed a similar type of water (Ca-Mg-HCO₃ type), which is well in agreement with the piper and Chadha diagrams.

A Schoeller diagram represents a semilogarithmic diagram of the concentrations of groundwater samples which represents the visual comparisons among the concentrations of ions in different sample areas. Main ionic constituents of the groundwater samples were plotted in Schoeller diagrams (Figure 6). In this study, the Schoeller diagrams revealed the dominance of Na⁺, Ca²⁺, and Mg²⁺ as well as HCO₃⁻ in the groundwater during both seasons (Figure 6). Interestingly, the dominance of Na was not observed in the piper and Chadha diagrams, nevertheless a moderate dominance of Na⁺ was observed in pie diagrams.



Figure 6. Schoeller diagrams of the samples of CU for both seasons

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Figure 7. Gibbs plots showing the mechanisms governing groundwater chemistry during the summer (a, b) and winter (c, d)

Gibbs Plot

Gibbs plots were plotted to have deeper insights concerning hydrochemical processes and groundwater chemistry [122]. Plots drawn with (Na+K)/(Na+K+Ca) and Cl/(Cl+HCO₃) versus TDS indicated that the groundwater samples (70.83% and 58.33 %) fell mostly in the rock-dominant zone, which might be controlled by carbonate dissolution and hydrolysis of silicate within the aquifers in both the winter and summer (Figure 7).

On the other hand, a few samples (29.17% and 41.67%) were found in the precipitation-dominant zone in both seasons. The chemical compositions of very low-saline waters (Na and Cl) (see pie charts and Scholler diagrams in Figures 5 and 6, respectively)

were controlled by the amount of dissolved salts furnished by precipitation [113].

Groundwater Contamination and Source Identification

Principle Component Analysis (PCA)

PCA was analyzed for the reduction of the data set presented by scree plot approach (Supplementary Figure 1). For analyzing the compositional pattern of the variables, PCA was conducted. PCA also reduced the data set to some factors that were highly influencing through the elimination of insignificant data [123]. Here, we report only the R-mode PCA analysis in the present study due to small area and population. From PCA, five components with eigenvalues > 1 (Supplementary

Figure 1) were taken to explain 78.983% and 80.886% of total variance of the winter and summer, respectively. The use of eigenvalue > 1 is recommended for verifying five loading factors. From the data set of the winter, 36.947%,

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15.507%, 10.170%, 9.222%, and 7.138% of variances were explained by PC1, PC2, PC3, PC4, and PC5, respectively, while for the summer variances were 35.373%, 15.076%, 13.161%, 9.467%, and 7.809%, respectively (Table 4).

Table 4. Princi	ple com	ponent anal	ysis of	drinking	water of	quality	parameters	at the	CU	camp	ous
							1				

Parameters	Winter						Summer					
	PC1	PC2	PC3	PC4	PC5	PC1	PC2	PC3	PC4	PC5		
Temperature	0.389	0.569	-0.327	0.548	0.158	0.437	0.653	0.220	0.275	-0.194		
TDS	0.929	-0.080	0.194	0.187	0.072	0.794	0.039	0.189	-0.323	0.070		
EC	0.957	-0.094	0.168	0.107	0.126	0.776	0.041	0.185	-0.290	0.023		
Turbidity	0.181	-0.342	0.090	-0.088	-0.036	0.760	0.170	0.174	0.154	0.114		
pН	0.800	-0.264	-0.387	-0.096	0.164	0.793	-0.353	0.296	0.030	0.355		
TH	0.900	-0.170	0.175	0.191	0.110	0.908	0.034	-0.326	0.171	-0.008		
ТА	-0.685	0.397	0.242	0.269	0.280	-0.068	0.482	0.116	-0.499	-0.639		
Free CO ₂	-0.396	0.518	0.210	0.433	0.234	0.159	0.626	0.624	0.057	-0.132		
Chloride	-0.002	-0.596	0.504	0.268	0.476	0.142	0.713	-0.324	0.515	-0.080		
Nitrate	-0.621	-0.082	0.403	-0.259	0.232	-0.261	0.701	0.437	-0.398	0.125		
Sulfate	0.385	0.546	-0.067	0.156	0.160	0.275	0.323	-0.713	-0.244	-0.257		
Phosphate	0.270	0.320	0.196	-0.524	-0.190	0.661	0.256	0.234	0.164	0.484		
Bicarbonate	0.852	0.041	0.095	0.403	0.080	0.821	0.141	0.009	-0.068	-0.135		
Na	0.508	0.299	-0.116	-0.435	0.594	0.710	-0.276	-0.358	-0.407	0.101		
K	-0.753	0.017	0.097	-0.018	-0.343	-0.796	0.109	-0.059	-0.377	0.054		
Ca	0.704	-0.107	-0.051	-0.497	0.103	0.721	-0.163	0.361	0.071	0.363		
Mg	0.811	0.076	-0.339	-0.432	0.166	0.859	-0.344	0.155	0.054	0.199		
Al	0.417	0.453	0.535	-0.136	-0.540	0.022	-0.115	0.522	-0.357	0.119		
Fe	0.471	-0.722	-0.094	0.244	-0.298	0.072	-0.588	0.389	0.363	-0.331		
Mn	0.783	0.318	0.332	0.027	-0.013	0.374	0.649	-0.435	0.438	0.010		
As	0.843	0.397	0.166	-0.030	-0.209	0.718	0.112	-0.450	0.382	-0.012		
Cr	-0.843	0.303	-0.083	-0.200	0.143	-0.877	0.185	0.275	-0.017	0.093		
Pb	0.513	0.230	0.618	-0.221	-0.028	-0.611	-0.033	-0.078	0.352	0.271		
Cd	0.293	-0.700	0.454	0.149	0.276	-0.588	-0.262	-0.392	-0.091	0.630		
Co	-0.235	-0.180	0.733	0.174	0.146	-0.680	0.106	-0.405	-0.008	0.416		
Cu	-0.196	-0.448	-0.404	0.165	-0.050	-0.358	-0.263	0.460	0.637	-0.189		
Ni	-0.905	0.012	0.228	-0.045	0.262	-0.722	0.285	0.270	0.145	0.085		
Se	0.148	0.380	-0.409	0.517	0.334	0.276	-0.695	-0.058	-0.232	-0.488		
Zn	-0.135	-0.211	-0.085	-0.596	0.567	-0.349	-0.532	0.159	0.478	-0.352		
Sn	0.049	0.870	0.151	-0.101	0.127	0.029	0.262	0.729	0.006	0.170		
Eigen Value	11.084	4.652	3.051	2.767	2.141	10.612	4.523	3.948	2.840	2.343		
% Total variance	36.947	15.507	10.170	9.222	7.138	35.373	15.076	13.161	9.467	7.809		
Cumulative % variance	36.947	52.453	62.623	71.845	78.983	35.373	50.449	63.610	73.076	80.886		

N.B: Values in bold denote significant positive loadings

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Figure 8. Component plot in a rotated space of PCA



Figure 9. Cluster analysis for ionic relationships in both seasons

Three classes of factor loadings are specified here, which are strong, moderate, and weak with the values in that order of > 0.75, 0.75-0.50, and 0.50-0.30[114]. The first 3 PCs contributed 62.623% and 63.610% of the entire variance for the winter and summer, respectively (Figure 8). PC1 (eigenvalues=11.084 and 10.612, most significant variable) was positively dominant with strong to moderate ratio which contains TDS, EC, pH, TH, HCO_3^- , Na, Ca, Mg, Mn, As, and Pb in both seasons, and turbidity and PO_4^{3-} in the summer. Negatively strong to moderate loadings were dominated by TA, NO_3^- , K, Cr, and Ni during the winter, and K, Cr, Pb, Cd, Co, and Ni during the summer. These factors are explained by natural hydrogeochemical process,

dissolution of carbonate, and leaching into aquifers [68]. PC2 (eigenvalues=4.652 and 4.523) was the second significant variable showing strong to moderate loadings dominated by temperature, free CO₂, SO₄²⁻, and Sn in the winter, and temperature, free CO₂, Cl⁻, and NO₃⁻ in the summer. Negatively Cl⁻, Fe, and Cd dominated in the winter, while Fe, Se, and Zn dominated in the summer. These factors can be explained by the sources through domestic, manure, fertilizer, and sewage water [124]. In PC3 (eigenvalues=3.051 and 3.948), moderate positive loadings of Cl⁻, Al, Pb, and Co were in the winter, and free CO₂, Al, and Sn in the summer, and negative dominance by SO42- in the summer indicated anthropogenic pollution from domestic and agricultural facts [68]. For PC4 (eigenvalues=2.767 and 2.840), temperature and Se were the dominant factors in the winter, while Cl⁻ and Cu that were the moderately dominant factors in the summer might represent the sources were domestic and weathering of rocks [125]. Finally, PC5 (eigenvalues=2.141 and 2.343) showed moderate positive loadings for Na and Zn, and negative loading for Al in the winter, and Cd was the only plus dominant element in the summer, which were probably from mixed natural and anthropogenic facts [126].

Cluster Analysis (CA)

Cluster analysis (CA) is an empirical method for identifying similarities and dissimilarities among different classes by forming clusters. R-mode CA was carried out in this study area (Figure 9). In both seasons, as an output of cluster analysis, three clusters were found in a dendrogram. Cluster 1 contained temperature, TDS, EC, pH, TH, PO₄³⁻, HCO₃⁻, Na, K, Ca, Mg, Fe, Pb, and Sn for both seasons, coupled with Al and SO₄²⁻ during the winter, as well as free CO₂, Cl⁻, Cr, Ni, NO₃⁻, and TA during the summer. These results revealed the possible mixing of ions, interactions of soil and groundwater, leaching of fertilizers into subterranean layers of waterbearing leaky rocks and their fractures, industrial pollution from the surroundings, carbonate rocks weathering, physico-chemical reactions, minerals dissolution, domestic and agricultural pollution, and atmospheric and vehicular pollution [40, 127]. Cluster 2 consisted of Cd, Co, Cl⁻, Cu, Zn, turbidity, As, and Se in the winter, and Mn, As, turbidity, Cu, Zn, Al, and Se in the summer, which represented the dissolution of minerals and domestic, agricultural, and industrial pollution. Finally, Cluster 3 included TA, free CO₂, Cr, Ni, and NO₃⁻ during the winter, while just Cd and Co during the summer, which could be influenced of physico-chemical reactions and industrial, domestic, and agricultural pollution [118]. A good agreement between PCA and CA was observed, having very little variations.

Correlation Matrix (CM) of Water Quality Parameters

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The correlation matrix (CM) identified the relation and variation among the GW samples through physicochemical parameters and groundwater pollution sources [68]. Significant and non-significant trends were observed in the correlation matrix. Significant and positive trends indicate the relation between two variables in such a way that they move in the same direction, and when two variables move in inverse directions it indicates a non-significant trend. Significant and positive trends for the winter and summer are presented in Supplementary Table 1. For most of the parameters, strong (P<0.01) and significant (P<0.05) correlations were found. The correlation matrix showed that inter-parameter relationships agreed with the PCAs significantly. TDS and EC significantly correlated with pH, TH, HCO₃⁻, Ca, Mg, Mn, As, Pb, and Cd during the winter, while significantly correlated with turbidity, pH, TH, PO₄³⁻, HCO₃⁻, Na, Ca, and Mg during the summer (Supplementary Table 1), which denoted similarity to PC1 in the cluster analysis. Significant correlations of turbidity, pH, TH, PO43-, and HCO3-with other parameters (Supplementary Table 1) also indicated similar sources of the PC1 values. Temperature correlated with SO₄²⁻, HCO₃⁻, and Se in the winter, and turbidity, free CO₂, Cl⁻, Mn, and Sn in the summer. These might have been almost the same sources described in PC2. On the other hand, significant correlations that existed between Al and Pb and Co were matched with PC3 in groundwater sources described earlier. On the whole, the correlation matrix mostly agreed with the PCAs.

Ion Dominance

Scatter diagram is an important tool for expounding hydrogeochemical processes on the basis of ions and ionic combinations [128]. Na⁺, Ca²⁺, and Mg²⁺ were found as the leading cations (pie and Schoeller diagrams, Figures 5 and 6, respectively) in our study area. The Ca²⁺/Mg²⁺ ratio of 1 indicates dolomite liquefaction and >2 indicates silicate mineral weathering [129]. In the study area, 8.33% of the samples had Ca²⁺/Mg²⁺ ratios less than 1 and 75% had ratios between 1 and 2, indicating dissolution of dolomite and calcite due to higher concentrations of Ca²⁺ and Mg²⁺. However, 16.67% of the samples had ratios over 2, which indicated silicate dissolution [129]. In Figure 10(a), the scatter diagram of (Ca²⁺+Mg²⁺) vs (SO₄²⁻+HCO₃⁻) makes public that majority of the samples fell above 1:1 equiline, inferring dolomite dissolution being the primary process, and only 4 samples fell below the line, which indicates silicate weathering [130–132].

The graph of $(Ca^{2+}+Mg^{2+})$ vs HCO_3^- (Figure 10(b)) exposed that 83.33% of the samples point fell above the 1:1 ratio line, suggesting the increased contribution of alkalis by the weathering of silicate

minerals over bicarbonate [129]. Only 4 samples (16.67%) lay below the 1:1 equiline, indicating bicarbonate zone caused by the reaction of carbonic acid with feldspar. In the presence of water, HCO₃can be produced through this reaction [133]. Na/Cl ratio of >1 was found, which indicated silicate weathering [129]. As shown in Figure 10(c), all the samples fell above the 1:1 equiline of Na⁺ vs Cl⁻ graph, which indicated the contribution of silicate weathering and ion exchange process was extensive (Hindersah et al., 2018). The diagrams of TC vs $(Na^{+}+K^{+})$ (Figure 10(d)) and TC vs $(Ca^{2+}+Mg^{2+})$ (Figure 10(e)) showed that all the samples fell above the 1:1 equiline, which also provided the indication of silicate weathering [134]. Secondary factors of anthropogenic sources were also found from the TC vs HCO_3^- plot (Figure 10(f)), where all the samples fell above the 1:1 equiline and far away from the line

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[135]. In Figure 10(g), the Na⁺ vs Ca²⁺graph shows the ion exchange reactions which led to increased Na⁺ concentration [136]. Chloro-alkaline indices (CAI-1 and CAI-2) indicated ion exchange. Positive values indicate direct exchange, while negative values indicate reverse order [132]. Negative CAI was found in this area (Figure 10(h)), which indicated the reverse ion exchange between Mg²⁺ and Ca²⁺ from GW and Na⁺ and K⁺ from host rocks [137].

Water Quality Index (WQI)

For the purpose of comprehending water grade, WQI was calculated for both seasons (Table 5). The weight values are presented in Supplementary Table 2. WQI is classified into five classes (Table 5), i.e., excellent, good, poor, very poor, and unsuitable for drinking [85].



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Figure 10. Scatter diagrams describing the ion chemistry

Based on our findings, we report here that 16.67% of the samples were excellent for drinking (Grade A) in both seasons. Good water quality (Grade B) was found in majority of the samples, 66.67% and 75.0% during the winter and summer, respectively. Poor water quality (Grade C) was observed in 16.67% and 8.33% of the samples during the winter and summer, respectively. WQI values were found to be different in both seasons as expected due to the varied values for different parameters. We found that with the increase of Fe concentration, WQI value also increases. When Fe concentration was below the permissible limit (1 mg/L of BSTI), excellent water quality (WQI=0-25) was observed. When Fe concentration was obtained between 1-1.5 mg/L, the water quality was observed as good (WQI=26-50). Moreover, at a few sampling stations, Fe content was found as >1.5 mg/L, hence poor water quality (16.67% and 8.33% during the winter and summer respectively) was observed (WQI=51-75). These observations indicate excellent to good quality of water is available at the CU campus for drinking purposes. However, poor water quality was also observed for both seasons in an insignificant fraction (SS-7 and SS-8 in the winter and SS-6 in the summer) Nevertheless, groundwater quality of CU campus in both seasons was suitable for drinking according to WQI data.

WQI category	Water quality rating	Num san	nber of nples	% of samples		Sam	Grading	
		Winter	Summer	Winter	Summer	Winter	Summer	
0–25	Excellent	2	2	16.67	16.67	SS-1 and SS-4	SS-2 and SS-4	А
26–50	Good	8	9	66.67	75.0	SS-2, SS-3, SS-5, SS-6, SS-9 to SS- 12	SS-1, SS-3, SS-5, SS-7 to SS-12	В
51–75	Poor	2	1	16.67	8.33	SS-7 and SS-8	SS-6	С
76–100	Very poor	-	-	-	-	-	-	D
>100	Unsuitable for drinking	-	-	-	-	-	-	Ε

Table 5. WQI of different sampling stations

Spatial Distribution (SD)

SD of WQI, pH, and Fe is represented in Figures 11(af). Drinking water of good quality for both seasons was dominant. Excellent water quality was observed in the northeastern region, while poor water quality was observed in the northwestern region during the winter. However, during the summer, excellent to insignificant poor water quality was observed in the eastern region. Spatial distribution of pH showed identical behaviors during both seasons, increasing in values to the southern region. Interestingly, on the basis of iron, seasonal variations were observed. The Hydrogeochemical Characteristics, Quality Assessment and Health Impact Analysis of Groundwater for Drinking in the University of Chittagong, Bangladesh

concentration of iron was relatively very much higher in the outer region during the winter, whereas during the summer the concentration did not increase highly towards the outer region as during the winter. In doing spatial analysis, physico-chemical parameters were selected based on their essential effects on water class, while cations, anions, and trace metals were selected on their chronological order of concentration. Thereby, spatial distribution maps were prepared for the parameters, namely TDS, EC, turbidity, free CO₂, TA, Na⁺, Ca²⁺, Mg²⁺, HCO₃⁻, Cl⁻, NO3⁻, Zn, Mn, Ni, and Cu for both seasons (Supplementary Figures 2(a-j), 3(a-l) and 4(a-h)).



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Figure 11. Spatial distribution maps for WQI, pH and Fe

For example, the highest TDS values were measured in the southeastern part during the winter and the southern-central part during the summer, whereas the lowest TDS values were measured in the northern part during both seasons (Supplementary Figures 2(a,b)).

Geostatistical Modeling

For the spatial analysis of geochemical data sets, Linear, Circular, Spherical, Exponential and Gaussian models are widely presented [138, 139]. Parameters like TDS, EC, Turbidity, TH, Na, Mg, K, Ca, HCO₃, Nitrate, Chloride, and Mn were considered for semivariogram modeling as they mostly influenced the geospatial factors during both seasons. Mean error (ME), mean square error (MSE), root mean error (RMSE), average standard error (ASE), and root mean square standardized error (RMSSE) were taken under consideration to evaluate the best fit models. A model is designated as the best-fit model when ME and MSE values are near to zero, RMSSE close to 1, and RMSE and ASE are as low as possible [63]. Cross validation was done to assess more accurate prediction performances, where TDS was given as an example. For TDS, ME and MSE of 0.05 and -0.04 and RMSSE of 1.02 show a good agreement of the model. The nugget, sill, lag size, nugget/sill ratio, and range of the best-fit semivariogram model are given in Tables 6 and 7. The exponential semivariogram model was found as the bestfit model in support of Turbidity, Chloride, Nitrate, Na, and Ca during the summer, and Nitrate and Na during the winter. The Gaussian semivariogram was found to be the best-fit model for TH, HCO₃⁻, K, and Mn in the summer, and TDS, EC, Turbidity, TH, HCO₃, Mg, and Mn in the winter. EC in the summer and Turbidity and K in the winter were found to be the best fit with the spherical

semivariogram model, while TDS and Mg in the summer and Chloride and Ca in the winter were the best-fit for the circular semivariogram model (Supplementary Figure 5). The Nugget/Sill ratio determines the degree of dependence, where a value less than 0.25 is regarded to be a strong spatial dependence whereas greater than 0.75 is considered as weak spatial dependence.

A moderate spatial dependence will be considered when a value lies between 0.25 and 0.75 [140]. EC, Nitrate, Na, K, Ca, and Mg exhibited strong spatial dependence for both seasons. TDS, Turbidity, and TH in summer and Mn in winter exhibited strong spatial dependences. Moderate spatial dependence was exhibited only in the winter in favor of TDS, TH, Chloride, and HCO₃⁻. Weak spatial dependence was exhibited in Chloride and Mn in the summer and Turbidity in the winter. The range varied from 0.0096 to 0.0426 km for the summer and 0.0096 to 0.05135 km for the winter. This variation might be owing to topographical factors (geometric factors, rainfall, surface runoff etc.) [63, 141]. Comparing with the CM, TDS had a strong correlation with TH (Supplementary Table 1) and they both had (Nugget/Sill ratios were 0.36 and 0.51, respectively) moderate spatial dependence. Same (weak) spatial dependence (Nugget/Sill ratios were 2.001 and 1.032, respectively) of Chloride and Mn was justified previously and a strong correlation (0.889) was found between them during the summer (Supplementary Table 1). Na, K, and Ca had strong correlations among them that established the same (strong) spatial dependence during both seasons. Comparing with spatial distribution (Supplementary Figures 3, 4, & 5), most of the parameters were found having identical spatial dependence from the northern to the southern region of this study area.

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Table 6: Indices of best fit semivariogram models for groundwater parameters (Summer)											
Parameter	Best fit model	Nugget	Major range	Sill	Nugget / Sill	Lag size	ME	RMSE	MSE	RMSSE	ASE
TDS	Circular	0	0.010086	316.6983	0	0.001394	0.051529	13.10129	-0.04402	1.021929	13.39042
EC	Spherical	0	0.01041	783.1614	0	0.001394	0.012186	22.60529	-0.03186	1.065495	21.99671
Turbidity	Exponential	1.954879	0.05135	20.6968	0.094453	0.025933	-0.26409	3.408597	-0.04205	1.222958	3.141594
TH	Gaussian	17.57533	0.02024	125.7208	0.139797	0.02593	1.261957	9.34424	0.044976	1.254502	7.306561
Cl	Exponential	162.0403	0.009617	80.9704	2.001229	0.00134	1.235838	13.20097	0.069323	0.837063	16.33333
NO3	Exponential	0	0.025648	0.800586	0	0.002137	-0.00308	0.652253	0.032419	1.045815	0.626859
HCO3	Gaussian	6.515808	0.009617	0.00011	59288.52	0.001564	0.003162	0.012949	0.152858	0.98206	0.011757
Na	Exponential	0	0.019776	10.90206	0	0.001648	0.575022	3.095053	0.111828	1.132395	2.484393
Κ	Gaussian	0.006835	0.023455	0.0433	0.157851	0.02593	-0.00018	0.092116	0.00448	0.850066	0.136851
Ca	Exponential	0	0.016155	6.86158	0	0.001346	-0.03472	1.353586	-0.00228	0.725098	2.082674
Mg	Circular	0	0.013844	3.906457	0	0.002571	0.001293	1.009795	-0.02167	0.895225	1.3411
Mn	Gaussian	270.1405	0.0197	261.6781	1.032339	0.02593	1.035907	24.84592	0.027977	1.216317	20.27674

Table 7. Indices of best-fit semivariogram models for groundwater parameters (Winter)

Parameter	Best fit model	Nugget	Major range	Sill	Nugget / Sill	Lag size	ME	RMSE	MSE	RMSSE	ASE
TDS	Gaussian	113.4956	0.01747	307.8422	0.368681	0.02593	2.494357	16.66004	0.074753	1.073584	15.34661
EC	Gaussian	157.047	0.01645	643.9739	0.243872	0.02593	2.777125	20.14694	0.055735	1.061965	19.78423
Turbidity	Spherical	1.089288	0.01218	0.22986	4.738919	0.001468	-0.34546	3.156218	-0.18327	2.300339	1.204348
TH	Gaussian	46.46837	0.016228	90.39035	0.514086	0.02593	1.064641	10.20637	0.048758	1.077222	9.381135
Cl	Circular	1.37228	0.009617	4.7738	0.287461	0.001346	0.153394	1.956617	0.064721	0.958507	2.179396
NO3	Exponential	0	0.042645	1.026524	0	0.00355	-0.04939	0.599078	-0.00464	0.984571	0.620404
HCO3	Gaussian	60.18368	0.019185	145.4072	0.413898	0.02593	1.69096	11.91817	0.070156	1.055436	10.82332
Na	Exponential	0	0.01692	13.21747	0	0.00141	0.456234	3.095809	0.080256	1.038131	2.854944
Κ	Spherical	0	0.019839	0.04242	0	0.025933	0.002186	0.113757	0.031141	1.011962	0.131739
Ca	Circular	0.279576	0.01298	7.295023	0.038324	0.001346	0.014808	1.390863	0.000894	0.82614	1.98053
Mg	Gaussian	0.317027	0.01548	3.91106	0.081059	0.003316	0.065336	0.877689	-0.00633	0.861919	1.2137
Mn	Gaussian	97.2826	0.020791	856.4107	0.113593	0.02593	1.232037	17.34978	0.015475	1.15389	18.10965



Figure 12. HQ values of As, Cr, Fe, and Mn, and HI values for different aged people

Health Risk Assessment

Non-carcinogenic Effect

CDI, *HQ*, *HI*, and *CR* were measured for the assessment of health risks. *RfD* values were obtained from *USEPA*. According to *USEPA*-2012, *RfD* values of Fe, As, Cr, and Mn are 0.7, 0.0003, 0.003, and 0.14 mg/kg/day, respectively [85]. Chronic daily intake (*CDI*) was estimated using Equation 5. The concentrations of respective trace metals were prerequisites for *CDI* calculation. Human water ingestion rate, *IR* was considered as 3.53 L/day for adults and 1 L/day for children. Exposure duration, *ED* was considered as 24 years for adults and 6 years for children.

Exposure frequency, *EF* was 365 days/year. Average body weight (*BW*) was 70 kg for adults and 15 kg for children. Averaging time, *AT* was obtained by $AT = 365 \times ED$. All the standard values were obtained from *USEPA* guideline [85]. *HQ* values of Fe, As, Cr, and Mn were detected at much lower values than unity, which indicated that no non-carcinogenic health risk was associated with the drinking water. *HQ* values were

ranged within 0.2, 0.06, 0.2, and 0.05 for As, Cr, Fe, and Mn, respectively. In this study area, HQ values for the entire samples were less than unity (Figure 12). From the trend lines, it was evident that HQ values were the highest for children in the winter, denoting children as more susceptible than adults. Notably, an increasing trend was viewed from samples 1 to 12 (see map for sampling stations, Figure 1) with a positive slope observed, whereas the trend was reverse for Cr. HI was estimated by summing up HQ values of Fe, As, Cr, and Mn. *HI*>1 indicates significant health risks, where 10^{-4} – 10⁻⁶ is the tolerable limit [142, 143]. From Figure 12, HI was observed having lower values than unity for adults and children in both seasons. This reveals that there was no significant non-carcinogenic risks in both seasons for all samples. However, in both seasons, children were more prone to CDI and HI, with higher values than adults.

Carcinogenic Effect

CDI and *CR* for As and Cr are presented in Supplementary Tables 3(a) and (b). In the summer, mean *CR* values of As for adults and children were 8.75

 \times 10⁻⁶ and 1.16 \times 10⁻⁵, respectively, where the values were correspondingly 9.35 \times 10⁻⁶ and 1.24 \times 10⁻⁵ for the winter. Maximum *CR* of As (8.36 \times 10⁻⁵) was found in the winter for children. It indicated that the probability of carcinogenic risks for adults was 8.75 and 9.35 in 1,000,000 for the summer and winter, respectively, and 11.6 and 12.5 for children, respectively.

CR values of Cr ranged between 0.0-0.0002, where mean *CR* for adults and children were 3.74×10^{-5} and 4.95×10^{-5} , respectively, in the summer, and 3.43×10^{-5} and 4.53×10^{-5} , respectively, in the winter. From all these *CR* values, the probability of carcinogenic risks for adults were 3.74 and 3.43 in 100,000 for the summer and winter, respectively, while for children were 4.95 and 4.53, respectively. To sum up, all *CR* values were within the acceptable levels, therefore, cancer risks can be neglected. However, it was visible that children were of higher probability for carcinogenic risks.

CONCLUSION

We have applied descriptive statistics and multivariate and geochemical analyses coupled with health impact for evaluating the groundwater quality inclusively for the first time at the CU campus for drinking purposes. Here, we conclude-

- 1. Descriptive statistics showed that only pH and Fe exceeded the standard permissible limits during the summer and winter, indicating the slight quality deterioration of GW.
- Coliform bacteria were found at SS-8 and fecal coliform bacteria were found at SS-6. During the summer, SS-6 and SS-8 can cause a threat. Due to the bacteria, both SS-6 and SS-8 are required to be given intense attention.
- Major cations were in the order of Na⁺, Ca²⁺, Mg²⁺, K⁺ and Al³⁺, while the dominant major anions were ordered as HCO₃⁻, Cl⁻, NO₃⁻, SO₄²⁻, and PO₄³⁻.
- 4. Mean concentration followed by trace metals: Fe>Zn>Mn>Ni>Cu>Cr>Pb>Sn>Cd>Co>Se>As> Sb>U in the winter, and in the summer Fe>Zn>Mn>Cu>Ni>Cr>Pb>Co>Cd>As>Se>As> Sb>U.
- 5. Ca-Mg-HCO₃ water type was confirmed by the piper trilinear diagram and the findings were also likely to be supported by Chadah plots, Pie charts and Schoeller diagrams.
- 6. Scatter diagrams indicated the dolomite dissolution as the prime source for the major cations. Silicate weathering was the reason for the abundance of

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bicarbonate. Reverse ion exchange was confirmed from CAI indices.

- 7. Five PCA were extracted where 78.983% and 80.886% of total variance were represented during the winter and summer, respectively. PCA indicated that both geogenic (rock dominance) and anthropogenic sources (domestic, manure, agricultural, and sewage water) were responsible for governing water chemistry. CM and CA were largely supported by PCA data.
- 8. Gibbs diagrams indicated rock-dominant zone in the study area. From semivariogram modelling, spatial distribution and correlation were justified and strong to moderate spatial dependence was found for majority of the estimated parameters.
- 9. Water Quality Index (WQI) indicated that the supplied water is suitable for drinking purposes in the CU campus.
- 10. Health indices were identified and suggested that no carcinogenic and non-carcinogenic effects can be observed for long term exposure.

The outcomes of the present first time allencompassing study suggest that the GW is suited for human consumption. Noticeably, water during the winter is better than that of the summer. However, excess iron in both seasons could generate metallic taste, skin problems, plumbing issues, and allow bacteria to grow. Furthermore, coliform contamination in the summer could create health problems, like diarrhea, cholera etc. Therefore, reduction of iron and complete removal of coliforms are highly recommended from this study.

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Parameters	Pearson's correlation among the parameters								
1 drameters	Winter	Summer							
Temperature	Sulfate (0.546), bicarbonate (0.534) and Se (0.755**)	Turbidity (0.516), free CO ₂ (0.573), Cl ⁻ (0.528), Mn (0.544) and Sn (.533).							
TDS	EC (0.983**), pH (0.648*), TH (0.974**), bicarbonate (0.911**), Ca (0.564), Mg (0.608*), Fe (0.539), Mn(0.697*), As (0.764**), Pb (0.520) and Cd (0.537).	EC (0.994**), turbidity (0.702*), pH (0.642*), TH (0.692*), PO ₄ ³⁻ (0.586*), HCO ₃ ⁻ (0.666*), Na (0.715**), Ca (0.567) and Mg (0.747**)							
EC	pH (0.736**), TH (0.966**), bicarbonate (0.897**), Ca (0.616*), Mg (0.684*), Mn (0.755**), As (0.772**), Pb (0.543) and Cd (0.512).	Turbidity (0.717**), pH (0.608*), TH (0.690*), PO_4^{3-} (0.536), HCO_3^{-} (0.655*), Na (0.691*), Ca (0.522) and Mg (0.737**)							
Turbidity	-	pH (0.583*), TH (0.713**), Ca (0.640*), Mg (0.768**) and As (0.560)							
рН	TH (0.705*), bicarbonate (0.652*), Ca (0.587*), Mg (0.814**), Mn (0.525) and As (0.510).	TH (0.589*), PO ₄ ³⁻ (0.675*), HCO ₃ ⁻ (0.584*), Na (0.532), Ca (0.901**) and Mg (0.898**)							
ТН	Bicarbonate (0.914**), Mg (0.573), Fe (0.573), Mn (0.677*), As (0.737**) and Cd (0.578*).	PO ₄ ³⁻ (0.573), HCO ₃ ⁻ (0.714**), Na (0.723**), Ca (0.524), Mg (0.742**), Mn (0.601*) and As (0.858**)							
ТА	Free CO ₂ (0.890**), Cr (0.688*) and Ni (0.774**).	NO ₃ -(0.858**)							
Free CO ₂	Ni (0.503)	NO ₃ ⁻ (0.657*)							
Chloride	Cd (0.800**)	Mn (0.889**) and As (0.543)							
Nitrate	Ni (0.729**)								
Sulphate	Se (0.692*)								
Phosphate	-	HCO ₃ ⁻ (0.613*), Ca (0.672*) and Mg (0.571).							
Bicarbonate	Mn (0.740**) and As (0.770**)	Mg (0.557) and As (0.614*)							
Na	Ca (0.586*) and Mg (0.754**)	Mg (0.614*) and Se (0.614*)							
К	Cr (0.663*) and Ni (0.575)	Cr (0.775**), Cd (0.574), Co (0.570) and Ni (0.570)							
Са	Mg (0.847**)	Mg (0.774**)							
Mg	Mn (0.526) and As (0.620*)	As (0.500)							

Supplementary Tables and Figures

Supplementary Table 1. Correlation matrix

Al	Mn (0.676*), As (0.740**) and Pb (0.745**)	
Fe	Cd (0.608*)	Zn (0.550)
Mn	As (0.869**) and Pb (0.613*)	As (0.744**)
As	Pb (0.633*)	-
Cr	Ni (0.803**)	Pb (0.597*) and Ni (0.807**)
Pb	Co (0.504)	Cd (0.564)
Cd	-	Co (0.785**)
Cu	-	Zn (0.840**)

** Correlation is significant at the 0.01 level (2-tailed).

*Correlation is significant at the 0.05 level (2-tailed).



(a) Winter



Supplementary Figure 1. Scree plot of the characteristic roots (Eigenvalue) of PCA























Supplementary Figure 2. Spatial distribution maps of TDS, EC, turbidity, free CO₂ and TA during both summer and winter seasons







Supplementary Figure 3. Spatial distribution maps of Na⁺, Ca²⁺, Mg²⁺, HCO₃⁻, Cl⁻ and NO₃⁻ during both winter and summer seasons





(e)

91"870"E

0.05 0.5

91'47'30'E

91148'9"E



Supplementary Figure 4. Spatial distribution maps of Zn, Mn, Ni and Cu during both winter and summer seasons



Supplementary Figure 5: Best-fitted semivariogram model for different parameter

Parameters	WHO standards (S _n)	Unit weight (W _n)
рН	6.5-8.5	0.0372
TDS	1000	0.0003
EC	500	0.0006
Turbidity	10	0.0316
ТН	500	0.0006
Chloride	600	0.0005
Nitrate	10	0.0316
Sulfate	400	0.0008
Phosphate	6	0.0527
Na	200	0.0016
К	12	0.0264
Са	75	0.0042
Mg	35	0.0090
Al	200	0.0016
Fe	1	0.3165
Mn	100	0.0032
As	50	0.0063
Cr	50	0.0063
Pb	10	0.0316
Cd	5	0.0633
Hg	1	0.3165
Со	50	0.0063
Cu	1000	0.0003
Ni	100	0.0032
Se	10	0.0316
Ag	20	0.0158
Zn	5000	6.33E-05
		$\Sigma W_{-} = 0.9997$

Supplementary Table 2. Relative weight of different parameters and their standards used for WQI determination

Sampling Station	Adult (Summer)		Child (Summer)		Adult (Winter)		Child (Winter)	
	CDI	CR	CDI	CR	CDI	CR	CDI	CR
SS-01	1.37E-06	2.06E-06	1.82E-06	2.73E-06	6.94E-07	1.04E-06	9.18E-07	1.38E-06
SS-02	1.92E-06	2.89E-06	2.54E-06	3.82E-06	1.25E-06	1.87E-06	1.65E-06	2.47E-06
SS-03	3.61E-06	5.42E-06	4.78E-06	7.16E-06	7.07E-07	1.06E-06	9.35E-07	1.4E-06
SS-04	1.42E-06	2.13E-06	1.87E-06	2.81E-06	2.27E-06	3.4E-06	3E-06	4.49E-06
SS-05	1.68E-06	2.52E-06	2.22E-06	3.33E-06	1.28E-06	1.91E-06	1.69E-06	2.53E-06
SS-06	1.89E-06	2.84E-06	2.5E-06	3.75E-06	1.32E-06	1.98E-06	1.75E-06	2.62E-06
SS-07	1.95E-06	2.93E-06	2.58E-06	3.87E-06	7.37E-07	1.11E-06	9.74E-07	1.46E-06
SS-08	6.07E-06	9.1E-06	8.02E-06	1.2E-05	4.2E-06	6.3E-06	5.55E-06	8.33E-06
SS-09	1.43E-06	2.15E-06	1.9E-06	2.84E-06	1.61E-06	2.42E-06	2.13E-06	3.2E-06
SS-10	1.56E-06	2.34E-06	2.06E-06	3.1E-06	5.56E-06	8.34E-06	7.35E-06	1.1E-05
SS-11	3.67E-05	5.5E-05	4.85E-05	7.27E-05	4.22E-05	6.33E-05	5.58E-05	8.36E-05
SS-12	1.04E-05	1.56E-05	1.38E-05	2.07E-05	1.3E-05	1.96E-05	1.72E-05	2.58E-05
Min	1.37E-06	2.06E-06	1.82E-06	2.73E-06	6.94E-07	1.04E-06	9.18E-07	1.38E-06
Max	3.67E-05	5.5E-05	4.85E-05	7.27E-05	4.22E-05	6.33E-05	5.58E-05	8.36E-05
Mean	5.83E-06	8.75E-06	7.71E-06	1.16E-05	6.24E-06	9.35E-06	8.24E-06	1.24E-05

Supplementary Table 3 (a). CDI and CR values for As in different sampling stations

Supplementary Table 3 (b). CDI and CR values for Cr in different sampling stations

Sampling Station	Adult (Summer)		Child (Summer)		Adult (Winter)		Child (Winter)	
	CDI	CR	CDI	CR	CDI	CR	CDI	CR
SS-01	6.7E-05	3.34E-05	8.82E-05	4.41E-05	8.37E-05	4.19E-05	1E-04	5.53E-05
SS-02	0.00011	5.26E-05	0.000139	6.95E-05	0.000122	6.09E-05	2E-04	8.06E-05
SS-03	0.00011	5.66E-05	0.00015	7.48E-05	7.87E-05	3.93E-05	1E-04	5.2E-05
SS-04	8.6E-05	4.28E-05	0.000113	5.65E-05	0.000129	6.43E-05	2E-04	8.5E-05
SS-05	0.00013	6.33E-05	0.000167	8.37E-05	0.000119	5.93E-05	2E-04	7.83E-05
SS-06	8.8E-05	4.4E-05	0.000116	5.82E-05	0.000101	5.03E-05	1E-04	6.64E-05
SS-07	0.0001	5.1E-05	0.000135	6.74E-05	2.99E-05	1.49E-05	4E-05	1.97E-05
SS-08	5.1E-05	2.53E-05	6.7E-05	3.35E-05	6.56E-06	3.28E-06	9E-06	4.34E-06
SS-09	5.3E-05	2.64E-05	6.97E-05	3.48E-05	0.000107	5.35E-05	1E-04	7.07E-05
SS-10	0.00011	5.39E-05	0.000142	7.12E-05	4.68E-05	2.34E-05	6E-05	3.09E-05
SS-11	0	0	0	0	0	0	0	0
SS-12	0	0	0	0	0	0	0	0
Min	0	0	0	0	0	0	0	0
Max	0.00013	6.33E-05	0.000167	8.37E-05	0.000129	6.43E-05	2E-04	8.5E-05
Mean	7.5E-05	3.74E-05	9.9E-05	4.95E-05	6.85E-05	3.43E-05	9E-05	4.53E-05