

Research Paper

Drinking water quality, exposure and health risk assessment for the school-going children at school time in the southwest coastal of Bangladesh

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ABSTRACT

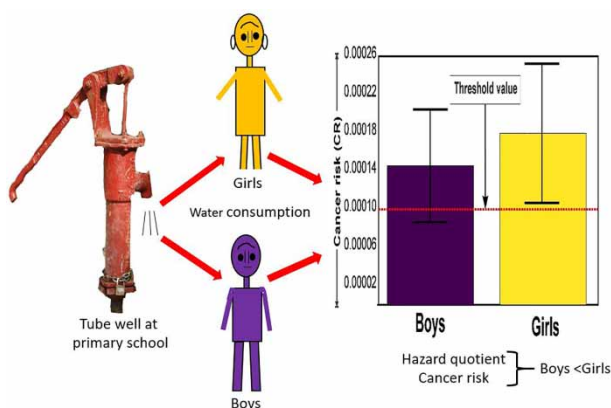
Scarcity of safe drinking water in the coastal regions throughout the world has long been recognized due to hydrological vulnerability and natural disaster, which is severe in developing countries like Bangladesh. This study focuses on trace metal(loid)s contamination and their associated health risks for primary school children from the consumption of tubewell water at school time in the vulnerable southwest coastal region of Bangladesh. The average content of electrical conductivity (EC), turbidity, chloride, total dissolved solids (TDS), hardness, iron (Fe), and manganese (Mn) were $1,983.6 \pm 1,434.6 \mu\text{S cm}^{-1}$, $10.46 \pm 10.3 \text{ NTU}$, 676.3 ± 648.1 , $1,089.1 \pm 788.6$, 560.6 ± 326.6 , 2.18 ± 1.99 , and $0.19 \pm 0.36 \text{ mg L}^{-1}$, respectively, which exceeded their respective health-based guideline values. The concentrations of arsenic (As), cadmium (Cd), lead (Pb), and zinc (Zn) were lower than the World Health Organization provisional guideline values. Spearman's correlation analysis revealed that the EC of groundwater is dependent on TDS, chlorides, and other cations contributing to hardness, while turbidity results from the Fe content in groundwater. The hazard quotients (HQs) of As, Fe, Mn, and Zn intake were lower than unity for both boys and girls, indicating no non-carcinogenic risks to the children. However, cancer risks (CRs) from As exposure through drinking water were 1.5 and 1.8 times higher than the provisional safe value of 10^{-4} for boys and girls, indicating a lifetime cancer risk to the school-going children. Therefore, prompt and effective monitoring is a crying need to ensure water's continuous usability for drinking purposes in the study area.

Key words: coastal region, health risk assessment, Khulna, primary school children, tubewell water

HIGHLIGHTS

- The EC, chloride, TDS, and hardness of drinking water were beyond the guidelines.
- Iron and manganese content were higher than the WHO guideline values.
- pH has a negative correlation with all the other variables.
- Although arsenic content was low, it poses a lifetime cancer risk to the children.

GRAPHICAL ABSTRACT



INTRODUCTION

In Bangladesh, groundwater or tubewell water (TWW) is used as the primary source for drinking, including cooking, irrigation, and many other domestic utilities (Kormoker *et al.* 2020). Approximately, one-third of the world's population uses TWWs as a primary source of drinking water (DW) and other domestic purposes where about 35–77 million people are facing adverse health-related consequences including cancers through consumption of elevated levels of As in DW. Additionally, 35 million inhabitants in coastal areas are facing a crisis in supply of fresh DW due to saltwater intrusion (Zahid *et al.* 2008; Edmunds *et al.* 2015; Talukder *et al.* 2016; Kormoker *et al.* 2020). In addition, coastal areas are much flatter and more vulnerable to sea-level rise, flooding, and saltwater intrusion into the freshwater aquifer, which influences geochemistry and physicochemical properties of groundwater (GW) such as pH, EC, TDS, hardness, turbidity, chloride content, and contamination of many other trace metal(loid)s. The GW qualities are also affected by the changes of local topography, hydrology, squeezing and catchment system, atmospheric amalgamation, different surface and subsurface geochemical processes (Milovanovic 2007; Vasanthavigar *et al.* 2010), and therefore, water quality monitoring and risk assessment is important, particularly in the vulnerable coastal regions in the world including Bangladesh where significant salt intrusion and climate change are causes for major concern.

Recently, a study was conducted on DW contamination by As, Fe, and chloride, at primary schools in a coastal area of Bangladesh, and were found to pose lifetime cancer and non-cancer risks to the school-going children by As contamination (Rahman & Hashem 2019). Rahman *et al.* (2016) reported that the DW quality (pH, EC, TDS, hardness, chloride, and trace metal(loid)s) from the primary schools in the Magura district (western Bangladesh) does not meet the standard recommended by the provisional guideline value of the WHO and the Bangladesh drinking water standard (BDWS). Moreover, water quality was also monitored in Pakistan and India and serious health risks for children were reported (Baig *et al.* 2016; Joardar *et al.* 2021).

Drinking water that does not meet regulatory guidelines can cause serious diseases in humans, especially to the children who are more vulnerable to infection. Chloride and salinity in DW cause bladder (about 80%), colon and rectal cancer (about 38%), and heart disease by complexing with other organic compounds (Cantor *et al.* 1998; Hildesheim *et al.* 1998; Simpson & Hayes 1998), and are a particular risk to children who have soft cells and tissues. Also, both excessive and inadequate hardness can harm people since deficient calcium and magnesium levels cause colon and rectal cancer (Yang *et al.* 1999), and cardiovascular disease (Crawford 1972; Bernardi *et al.* 1995).

Apart from the physicochemical properties, trace metal(loid)s such as As, Fe, Mn, Zn, Pb, and Cd were found in excess content in coastal groundwater (Rahman *et al.* 2015; Islam *et al.* 2017; Rakib *et al.* 2020). Different statistical tools (e.g., Spearman's normal and non-normal distribution, Kolmogorov–Smirnov test, etc.) are used for interpreting a significant correlation among the physicochemical variables (parameters) of water to predict their role in the environment (Kormoker *et al.* 2020). A study from Bangladesh showed that exposure to As $> 50 \mu\text{g L}^{-1}$ via DW was associated with reduced intellectual function in children compared with children exposed to As $< 5.5 \mu\text{g L}^{-1}$ (Wasserman *et al.* 2006). Overexposure to Fe and Mn can cause adverse health risks, including Parkinson's disease, Huntington's disease, cardiovascular disease,

hyperkeratosis, diabetes mellitus, pigmentation changes, Alzheimer's disease, kidney, liver, respiratory, and neurological disorders (Goldhaber 2003; Wasserman *et al.* 2006; Torres-Agustín *et al.* 2013; Ghosh *et al.* 2020; Rahman *et al.* 2021). Consequently, the health risk assessment model has long been used as an essential tool in determining human health risks for trace metal(loid)s intake through DW (Rahman *et al.* 2019). Most of the previous studies evaluated As concentration in DW from households and did not consider the additional exposure from other DW sources as children usually drink water from tubewells at school during school time. Thus, the risk is under- or overestimated as only a single DW source was considered. Often the tubewell located in the schools contained an elevated concentration of contaminants, including As ($98.3 \pm 7.93 \mu\text{g L}^{-1}$) (Joardar *et al.* 2021), which reflects the importance of monitoring water quality and associated risk (cancer and non-cancer).

Very limited studies have been found on the assessment of TWW qualities and their subsequent risk to humans, particularly for children in the coastal region of Bangladesh where safe DW supply is still quite challenging as a result of lack of fresh-water aquifers, high salinity, and high cost of water treatment technologies (Kormoker *et al.* 2020). In addition, the natural disasters and anthropogenic activities result in worsening water quality in the study area day by day, which could affect the physical and mental health of the coastal population, where children are more vulnerable compared with adults.

Therefore, the aims of this study are to assess (i) the contamination of salinity-induced physicochemical water quality parameters including trace metal(loid)s contamination, (ii) statistical correlation analysis (Spearman's normal and non-normal distribution) was performed to characterize the inter-relationships among the tested parameters, and (iii) carcinogenic and non-carcinogenic health risk assessment of children via the consumption of trace metal(loid)s such as As, Fe, Mn, Cd, Pb, and Zn in TWWs from 35 primary schools at school time exposure to ensure safe and potable water supply and protect future students from carcinogenic health-related consequences.

MATERIALS AND METHODS

Description of the study area

The study area is in the southwest coastal region of Bangladesh, known as the Khulna district, primarily an industrial city. It covers an area of 4,389.11 km² with nine Upazilas (sub-districts), namely Terokhada, Batiaghata, Dacope, Dumuria, Dighalia, Koyra, Paikgachha, Phultala, and Rupsha (BBS 2011). Three major rivers (Bhairab, Rupsha, and Posur) are flowing through this district in the east, while on the southern side, the world's largest mangrove forest, the Sundarbans, is situated.

The geology of this region is characterized by Pleistocene-Modhupur clay and underlying fluvial-deltaic sediment of the Holocene. The sediment is mainly composed of quartz with some plagioclase and potassium feldspars, with some fragments of volcanic, metamorphic, and sedimentary rocks (Uddin & Lundberg 1999; Halim *et al.* 2010). Meanwhile, the hydrogeology of the Khulna region has been described as a three aquifer system consisting of a semiconfined, shallow Holocene aquifer (extend 100 m below ground level) that is vertically separated from two Pleistocene aquifers (extending 200 and 300 m below ground level) (Burgess *et al.* 2011). The contaminants, especially inorganic pollutants such as As, Mn, Pb, and Cd in aquifer, originate from the interaction between water and rocks belonging to different geological times, and the concentration of pollutants depends on several factors, including water-rocks chemistry, surface and groundwater interactions, the geologic setting of the preferred pathways for groundwater flow, and any other sources for water pollution (Yousif & El-Aassar 2018).

Khulna has an annual average temperature of 26.3 °C (79.3 °F) and monthly means varying between 12.4 °C (54.3 °F) in January and 34.3 °C (93.7 °F) in May. The annual average rainfall at Khulna is 1,809.4 mm (71.24 in.), in which approximately 87% of the annual average rainfall occurs between May and October.

Sampling and laboratory analysis

The groundwater samples were collected from 35 randomly selected tubewells located in 35 primary schools (Figure 1) in the Khulna district of Bangladesh. The study area was selected on the basis of its hydrological and geological vulnerability, natural calamity, and salt intrusion, which can contribute to the scarcity of safe drinking water. The selected tubewells were under the open sky and may sometimes contain dust or any other suspended particles, and therefore, the water samples were collected after the first 10 minutes of continuous discarding so that the samples could be free from secondary contamination and represent the actual layer of the individual tubewells. Three samples were collected from each sampling point (35 selected school's tubewells and three samples per school) for physicochemical and trace metal(loid)s analyses. Among three samples (each of 500 mL), two were taken into the high-density polyethylene bottles for As, Pb, Cd, Mn, Fe, and Zn analyses which are

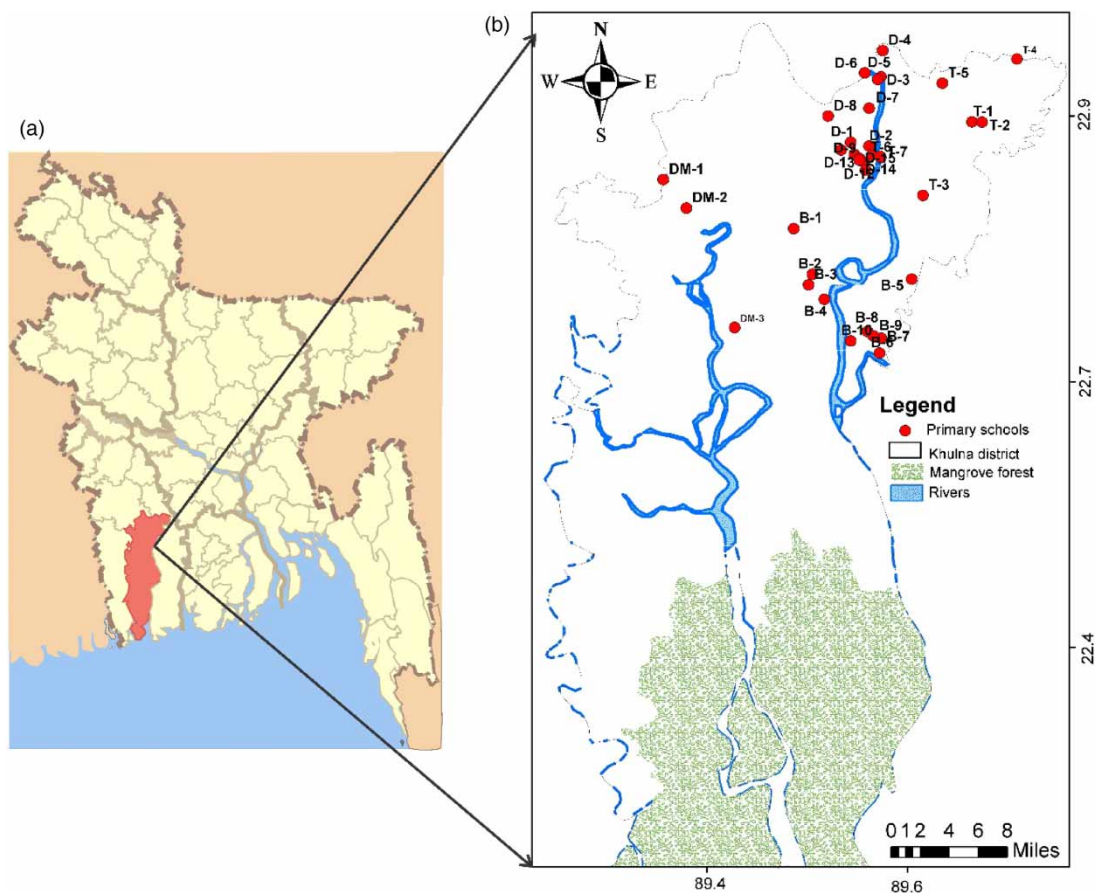


Figure 1 | Study area (a) location of the Khulna district in Bangladesh; (b) study area showing the locations of 35 primary schools in southwestern (coastal region) Bangladesh.

primarily found in TWWs in the study area where As is being reported as a major concern. Industrial activities including Fe and steel-alloy, and Pb-battery production, lead smelter, fireworks, burning of organo-Mn compounds containing petrol, power plants, textile, painting and dyeing industries, galvanization, etc. as well as urbanization were significantly developed at the study area, which could impact background concentration of As, Pb, Cd, Mn, Fe, and Zn in groundwater. Another sample (1.0 L) was taken for other physicochemical analyses, including pH, EC, TDS, turbidity, chloride, alkalinity, and hardness. All samples were kept in the refrigerator at 4 °C until the experiment was complete. A global positioning system (GPS; Garmin eTrex 10) was used for the location of each sampling point, and details of the study area and sampling points are provided in Supplementary Table S1. Trace metal(loid)s were quantified using atomic absorption spectroscopy (AAS; SpectraAA 220, Varian, Australia). A detail of the sampling and analytical procedures can be found in our previous reports (Rahman *et al.* 2016; Rahman & Rahaman 2018). All analysis was conducted by following the American Public Health Association (APHA 2012) method. Briefly, As was measured by the hydride generation (HG-AAS) method using a T-shaped quartz absorption cell VGA 76/77 (Varian). The argon served as a carrier gas (gas flow rate was 0.1 L min⁻¹) at the wavelength of 193.7 nm and the slit width of 0.5 nm. The reductant agent was sodium borohydride (NaBH₄-0.6% w/V), and the acid reagent was HCl (5 M). The Fe, Mn, Pb, Cd, and Zn contents were measured by direct flame (F-AAS) using air-acetylene at the wavelength of 248.3, 279.5, 283.3, 228.8, and 213.9 nm, respectively. Mean and standard deviations were calculated from the results of the analysis of the three samples per sampling point. Three times the standard deviation of the blank samples was used for determining the limit of detections (LODs) for As, Fe, Mn, Pb, Cd, and Zn. The LODs of As, Fe, Mn, Pb, Cd, and Zn were observed to be 0.0005, 0.009, 0.009, 0.003, 0.004, and 0.006 µg L⁻¹, respectively, which seems to be good enough to measure the lower level of As, Fe, Mn, Pb, Cd, and Zn of the aqueous solutions.

Health risk estimation

In this study, the non-carcinogenic health risk was assessed based on chronic daily intake (CDI) of metals and hazard quotient (HQ) for both boys and girls using the US EPA recommended method (US EPA 2004, 2011).

The CDI is calculated according to the following equation (US EPA 2011):

$$CDI = \frac{(C \times IR_{\text{water}} \times EF \times ED)}{(BW \times AT)} \quad (1)$$

where C is the concentration of As, Fe, Mn, and Zn (mg L^{-1}) in drinking water from each primary school, IR_{water} is the ingestion rate of water ($1.0 \text{ L day (d)}^{-1}$) for children at each primary school; EF is the exposure frequency (230 d year^{-1}), ED is exposure duration (5 years), AT is the average time (1,150 days), BW is the body weights for 6–10 year-old school children (mean body weights of boys and girls are approximately 24.68 and 20 kg, respectively) (NCHS 2001). In this study, we have considered 1.0 L water consumption during school time to estimate health risk, although children usually consume 2.1 L of water daily (Hossain *et al.* 2013). Exposure duration was considered based on school opening days annually.

The Pb and Cd were excluded from health risks and statistical calculation because their concentrations were below the instrument detection limit (DL).

The HQ is estimated using the following equation (US EPA 2004):

$$HQ = \frac{CDI}{RfDo} \quad (2)$$

$$HI = \sum HQ = HQ_{As} + HQ_{Fe} + HQ_{Mn} + HQ_{Zn} \quad (3)$$

where RfDo is the oral reference dose ($\text{mg kg}^{-1} \text{ d}^{-1}$), and the RfDos for As, Fe, Mn, and Zn were 0.0003, 0.7, 0.14, and 0.3 $\text{mg kg}^{-1} \text{ d}^{-1}$, respectively (US EPA 2020). When the value of $HQ < 1$, the exposed population is safe from specific harmful effects of trace metal(loid)s, but $HQ > 1$ may have adverse health effects on the exposed population. The hazard index (HI) is a cumulative metric that considers the combined contribution of HQs of individual metals. $HI > 1$ indicates a chance of non-carcinogenic health risk, while $HI \leq 1$ indicates no possible health risk (US EPA 2001).

The cancer risk (CR) from the consumption of As-contaminated DW is estimated employing the following formula (US EPA 2004):

$$CR = CDI \times SF \quad (4)$$

where SF is the oral slope factor ($\text{mg kg}^{-1} \text{ d}^{-1}$) and the SF for As is $1.5 \text{ mg kg}^{-1} \text{ d}^{-1}$ (US EPA 2020).

Data analysis

The calculations of human health risk parameters and other general calculations were performed using Microsoft Excel 2016. The descriptive statistics were performed using SPSS version 22.0. Shapiro–Wilk and Kolmogorov–Smirnov tests were carried out to select a suitable correlation analysis method that indicated a mix of the normal and non-normal distributions of different parameters in the dataset. According to the Kolmogorov–Smirnov test, pH and hardness were normally distributed ($p > 0.05$), while other parameters were not normally distributed ($p < 0.05$; Supplementary Table S2). The Shapiro–Wilk test also indicated that pH is normally distributed, while others were non-normally distributed. Therefore, a non-parametric (Spearman's correlation) analysis was performed to identify the inter-relationships among the studied parameters (Schober *et al.* 2018).

RESULTS AND DISCUSSION

The descriptive statistics of the studied physicochemical parameters and trace metal(loid)s content are tabulated in Table 1, compared with the national and international standards.

Table 1 | Descriptive statistics ($n = 35$) and comparison of observed value with its standard value provided by different organizations

Parameters (unit)	This study		Water quality standard			Number of samples exceeding water quality standard			% of samples exceeding water quality standards		
	Range	Average \pm Standard deviation	WHO (WHO 2017)	US EPA (US EPA, 2009)	BDWS (ECR 1997)	WHO	US EPA	BDWS	WHO	US EPA	BDWS
pH	7.24–9.03	7.85 \pm 0.40	6.5–8.5	6.5–8.5	6.5–8.5	2	2	2	5.7	5.7	5.7
EC ($\mu\text{S cm}^{-1}$)	377–8,280	1,983.6 \pm 1,434.6	250	–	–	35	–	–	100	–	–
TDS (mg L^{-1})	261–4,554	1,089.1 \pm 788.6	600–1,000	500	1,000	19	28	19	54.3	80	54.3
Turbidity (NTU)	0.5–37	10.46 \pm 10.3	5	0.5–1	10	19	29	13	54.3	82.9	37.2
Chloride (mg L^{-1})	10–3,550	676.3 \pm 648.1	200–300	–	600	26	–	17	74.3	–	48.6
Alkalinity (mg L^{-1})	70–638	350.2 \pm 127.2	600	–	–	1	–	–	2.9	–	–
Hardness (mg L^{-1})	124–1,520	560.6 \pm 326.6	200	–	500	33	–	18	95.3	–	51.4
As (mg L^{-1})	0.0004–0.0035	0.0024 \pm 0.01	0.01	0.01	0.05	0	0	0	0	0	0
Fe (mg L^{-1})	0.17–8.58	2.18 \pm 1.99	0.3	0.3	0.3–1.0	32	32	23	91.4	91.4	65.7
Mn (mg L^{-1})	0.009–1.98	0.19 \pm 0.36	0.4	0.05	0.1	3	19	15	8.6	54.3	42.9
Pb (mg L^{-1})	<0.002	<0.002	0.01	0.015	0.05	–	–	–	–	–	–
Cd (mg L^{-1})	<0.002	<0.002	0.003	0.005	0.005	–	–	–	–	–	–
Zn (mg L^{-1})	0.005–1.32	0.23 \pm 0.41	3.0	5.00	5.0	0	0	0	0	0	0

WHO, World Health Organization; US EPA, United States Environmental Protection Agency; BDWS, Bangladesh drinking water standard.

pH and alkalinity

For DW to be considered of good quality, the pH level should be in the 6.5–8.5 range, as advised by the United States Environmental Protection Agency (US EPA), WHO (2017), and the BDWS (ECR 1997). Therefore, compatible DW should have approximately neutral characteristics. In this study, the mean pH was 7.85 ± 0.40 , which is within the recommended limits (Table 1). However, samples T-4 (pH=9.03) and T-6 (pH=8.53) contained a pH slightly higher than the recommended health-based guideline value (Supplementary Table S1), which is about 5.7% of the total TWWs in the primary schools (Table 1). Recently, Islam *et al.* (2020) reported that the average pH of groundwater in coastal regions was 7.34 and 7.44 during wet and dry seasons, respectively, which are slightly lower than this study. In addition, the pH of groundwater in the Khulna district was reported to be 7.2 (range: 6.5–7.9) (Islam *et al.* 2017), which is a bit lower than this study. However, a much lower pH value of 6.03 ± 0.61 was observed in the adjacent coastal district called Satkhira (Rakib *et al.* 2020).

At very low and high pH levels, all toxic metals' solubility may increase in groundwater, which enters into the human body through direct drinking of groundwater (Chuan *et al.* 1996; Tang *et al.* 2006). In addition, too much alkalinity influences the body's normal pH by neutralizing and inhibiting gastric juice secretion in humans (Drobnik 2000). Recently, the WHO (2017) has stipulated that the acceptable level of alkalinity in DW is 600 mg L^{-1} and all samples in our investigation were below the accepted limit, except DM-3 (638 mg L^{-1}), which is slightly higher than the recommended value of 600 mg L^{-1} (Supplementary Table S1). The mean value of alkalinity was $350.2 \pm 127.2 \text{ mg L}^{-1}$, which is more than half of the WHO-recommended value (Table 1).

Chloride leading to salinity

Different levels of chlorides cause salinity. In this study, chloride content ranged from 10 to $3,550 \text{ mg L}^{-1}$ with an average value of $676.3 \pm 648.1 \text{ mg L}^{-1}$ (Table 1) that exceeded both the WHO and BDWS limits of 200–300 and 600 mg L^{-1} , respectively. About 74.3 and 48.6% of samples surpassed the WHO and BDWS standards, respectively (Table 1). In a previous study in the Khulna district, an average chloride content of $1,776.74 \text{ mg L}^{-1}$ (range: 32.07–6,270.8 mg L^{-1}) was reported, which is

more than twice that of this study (Islam *et al.* 2017). A recent study from the nearby district (Shyamnagar, Satkhira) reported a five times higher level of chloride (average: $2,940.78 \pm 1,563.5 \text{ mg L}^{-1}$) in DW (Rakib *et al.* 2020). Also, in coastal regions (Khulna, Bagerhat, Satkhira, Patuakhali, etc.), the average chloride content was reported to be $2,005.74 \pm 2,685.5 \text{ mg L}^{-1}$ (Islam *et al.* 2020), which is also severalfold higher than this study. This extensive chloride content was considered one of the chlorides' original salinity levels since they are strongly correlated. Consequently, the salinity in DW is a concerning issue in the Khulna district since there is enough documentation on the adverse effects of salinity on human health, such as hypertension, miscarriages, skin diseases, acute respiratory infections, and diarrheal diseases (Vineis *et al.* 2011).

The elevated level of salinity might originate from NaCl, Na_2CO_3 , KCl, and CaCl_2 salts dissolved from weathering, leaching of rocks and infiltration of seawater (Rahman *et al.* 2015), tidal channel water inundation, brine shrimp aquaculture (Ayers *et al.* 2017), and industrial waste and sewage (Meride & Ayenew 2016). A study conducted in Massachusetts and Chicago, USA, which investigated two cohorts consisting of high school students with a high (272 mg L^{-1}) and low (20 mg L^{-1}) salinity level in public DW, hypothesized that systolic and diastolic blood pressure in the high region were significantly higher by 3–5 mmHg compared with the low salinity level region (Calabrese & Tuthill 1981; Vineis *et al.* 2011). The raised BP was the primary cause of cardiovascular disease which is responsible for 62% of strokes and 49% of coronary heart disease in developed countries (He & MacGregor 2007; Vineis *et al.* 2011). As a result, the increasing intrusion of salinity inland and the extent of salinity in the coastal areas are being blamed on the rising sea levels and effectively endangering the groundwater flow in coastal regions. Furthermore, salinity intrusion is accelerating gradually through tidal flooding during the wet season, leading to direct inundation by saline or brackish water, upward or lateral movement of salinity in groundwater during the dry season, and inundation via brackish water that is threatening the shrimp farming industry (Abedin *et al.* 2014; Habiba *et al.* 2014). River flows also affect the salinity such that Ganges River water is being diverted at Farakka Barrage (India-Bangladesh river system) and impacting salinity level in the southwest coastal parts of Bangladesh (Mirza 2004).

Electrical conductivity (EC) and total dissolved solids (TDS)

Electrical conductivity is the indication of the total dissolved substituent in water. The compounds that are dissolved into ions are called electrolytes, whereas different salts, organic and inorganic materials such as alkalis, chlorides, sulfides, and carbonates provide these ions with the ability to carry electric conductivity (Karmoker *et al.* 2018). The EC differs from pH in that it represents all active ions (negative and positive) in the water, while pH measures only the hydrogen (H^+) and hydroxyl (OH^-) ions. Generally, pH decreases as EC increases due to some active metal cations being precipitated by the media's depletion of hydroxyl ions. Pure and standard water indicates very low EC, i.e., low dissolved contaminated ions (Karmoker *et al.* 2018; Rahman & Hashem 2019). However, in this investigation, all samples exceeded the recommended EC value of $250 \mu\text{S cm}^{-1}$, where the highest and lowest values were $8,280 \mu\text{S cm}^{-1}$ (B-2) and $377 \mu\text{S cm}^{-1}$ (B-1), respectively, with an average value of $1,983.571 \mu\text{S cm}^{-1}$ (Table 1). These high values meant that the water was not of good quality and contained many dissolved impurities, which may be harmful to school-going children. The high EC indicated the presence of a higher content of different salts, organic and inorganic materials such as alkalis, chlorides, sulfides, and carbonates that provide these ions resulting in the higher EC of groundwater in the coastal areas. In the Khulna district, a previous study reported an average EC value of $3,018.65 \mu\text{S cm}^{-1}$ (range: $498\text{--}5,910 \mu\text{S cm}^{-1}$) (Islam *et al.* 2017), which is much higher than this study. In addition, about 3.5 times higher EC ($7,135.67 \pm 3,433.58 \mu\text{S cm}^{-1}$) than this study was found in Shyamnagar, Satkhira, which is an adjacent district to Khulna (Rakib *et al.* 2020).

TDS indicates the wide range of inorganic and organic minerals or various salts such as K, Ca, Mg, Na, Al, HCO_3^- , Cl^- , and SO_4^{2-} and several trace metals that are dissolved in water. High TDS could cause kidney stones, gall stones, blockage of arteries, heart disease, either a laxative or constipation effect, and potentially cancer (Meride & Ayenew 2016). The dissolved trace heavy metals (As, Pb, Be, Cd, Cr, etc.) can cause carcinogens to appear in humans.

The mean TDS content was $1,089.1 \pm 788.6 \text{ mg L}^{-1}$, but approximately 54.3 and 80% of water samples surpassed the value as prescribed by the WHO ($600\text{--}1,000 \text{ mg L}^{-1}$) and the US EPA (500 mg L^{-1}) (Table 1). Although many samples in this investigation fall in worrying levels, the highest value was $4,554 \text{ mg L}^{-1}$ (B-2) (Supplementary Table S1). However, the average TDS concentration was $1,089.1 \text{ mg L}^{-1}$, which went beyond all the maximum acceptable limits (Table 1). A significantly higher level of TDS was reported in the Khulna district (average: $1,556.05 \text{ mg L}^{-1}$) as well as Shyamnagar, Satkhira (average: $3,691 \text{ mg L}^{-1}$) (Islam *et al.* 2017; Rakib *et al.* 2020) which is higher than this study. However, the higher level of TDS concentration is found from the consequences of strong evapotranspiration, industrial toxic waste, domestic sewage, long-term

Table 2 | Quality rating of drinking water in terms of TDS (WHO 2003)

TDS (mg L ⁻¹)	Rating	Sample ID	Percent of samples
<300	Excellent	T-4, B-1, B-3, DM-1	11.4% (4 out of 35)
300–600	Good	D-7, D-8, D-10, B-8, B-10, DM-2	17.1% (6 out of 35)
600–900	Fair	D-6, D-14, B-4, B-9	11.4% (4 out of 35)
900–1,200	Poor	D-1, D-2, D-5, D-9, D-12, D-13, D-15, T-3, T-7, B-5, B-6, B-7	34.3% (12 out of 35)
>1,200	Unacceptable	D-3, D-4, D-11, T-1, T-2, T-5, T-6, B-2, DM-3	25.7% (9 out of 35)

water–rock reactions in a deteriorating condition (Chetia *et al.* 2012; Huq *et al.* 2019), which could significantly affect the drinking water quality.

The WHO (2003) categorized the preferable level of TDS in water as follows: excellent (<300 mg L⁻¹), good (300–600 mg L⁻¹), fair (600–900 mg L⁻¹), poor (900–1,200 mg L⁻¹), and unacceptable (>1,200 mg L⁻¹). In Khulna, 34.3% of TWWs are abysmal in quality, and 25.7% are unacceptable. Only 11.4% are considered excellent, while 17.1% are in a good category, which means there is a major problem with DW sources (Table 2).

Turbidity and hardness

Turbidity is another physical property of water that regulates the quality, indicates the cloudiness, and provides the media for microbial growth. It is an exhibitor of suspended materials (e.g., clay, silt, etc.), colloidal particles, inorganic (Mg, Fe) and organic chemicals, biological particles, and disease-producing organisms, including bacteria, viruses, and parasites which can cause diarrhea, gastrointestinal illness, and associated headaches (Schwartz *et al.* 2000; Hoque 2003; Muoio *et al.* 2020). Subsequently, the WHO, the US EPA, and the BDWS have established their respective safe values as 4, 0.5–1, and 10 NTU (Table 1). However, it remains a matter of significant concern that 54.3, 82.9, and 37.3% of the observed TWWs crossed these guideline values of the WHO, the US EPA, and the BDWS, respectively. The turbidity content ranged from 0.5 to 37 NTU, with an average value of 10.46 NTU, which exceeded all the guidelines (Table 1).

The hardness of water is due to Ca and Mg ions, originating from the soil, rock, sediment, and minerals. Both low and high values of hardness are harmful to the human body. A low value may cause colon carcinogens and rectal cancer (Yang *et al.* 1999) and cardiovascular disease (Crawford 1972; Bernardi *et al.* 1995) to appear since Ca and Mg can bind bile acid and fatty acid, thus affecting the creation of colon mucosa (Wargovich *et al.* 1983; Van der Meer & De Vries 1985; Pence & Bud-dingh 1988; Yang *et al.* 1999). However, several years ago, the WHO (2017) demonstrated that a high value may cause kidney stones and skin diseases such as eczema (WHO 2010). The provisional limits of hardness are 200 and 500 mg L⁻¹ provided by the WHO (2017) and the BDWS (ECR 1997), respectively. Only two samples fell within the WHO-recommended limit of 200 mg L⁻¹, while 33 (about 95.3%) exceeded the safe level. In addition, the hardness of water in the primary schools ranged between 124 and 1,520 mg L⁻¹, and the average value was 560.57 mg L⁻¹, which surpassed all the guideline values. Mahmud *et al.* (2020) reported that the total hardness of TWW was 16–178 (average: 52.03 mg L⁻¹) in the Khulna region which is 10 times lower than our study. The higher level of hardness in this study is, therefore, a threat to children's health in the investigated region.

Trace metal(loid)s content

The concentrations of As, Fe, Mn, Pb, Cd, and Zn are provided in Table 1. The increasing pattern of mean concentration of trace metal(loid)s was as follows: Fe (2.18 mg L⁻¹) > Zn (0.23 mg L⁻¹) > Mn (0.19 mg L⁻¹) > As (0.0024 mg L⁻¹) > Pb (<0.002 mg L⁻¹) = Cd (<0.002 mg L⁻¹).

The different trace metal(loid)s present in DW may have severe effects on both children and adults. A moderate Fe is essential for humans, but various problems may arise in terms of Fe being present in either very low or high amounts. For instance, low Fe content causes anemia in children and adults in developing countries, alters the brain's metabolism, neurotransmission, myelination, and gene and protein profiles (Beinner *et al.* 2005; Walker *et al.* 2007). In contrast to low iron levels, too much may affect water quality (less dissolved oxygen) and human health (kidney stones). Besides, the excessive level of Fe in DW and blood could damage the cells in the gastrointestinal tract, preventing them from adequately regulating Fe absorption. A high Fe level plays a significant role in producing atherosclerosis, Alzheimer's dementia, and

neurodegenerative disorders (Sieliechi *et al.* 2010). Interestingly, in this study, some TWWs contained a meagre amount of Fe, while others contained a relatively high amount. About 92% (32 out of 35) and 66% (23 out of 35) of groundwater exceeded the safe levels stipulated by WHO and US EPA (0.3 mg L^{-1}) and BDWS ($0.3\text{--}1.0 \text{ mg L}^{-1}$), among which sample D-5 indicated the highest content of Fe 8.58 mg L^{-1} (Table 1). In Bangladesh's coastal region, the average Fe content was reported to be 6.24 ± 8.41 and $5.13 \pm 7.02 \text{ mg L}^{-1}$ during wet and dry seasons, respectively (Islam *et al.* 2020), which are about 2–3 times greater than this study. In addition, a study from Shyamnagar and Assasuni in the Satkhira district reported about two times higher level of Fe content (4.9 ± 4.76 and $3.59 \pm 2.50 \text{ mg L}^{-1}$, respectively) in groundwater (Rahman *et al.* 2019; Rakib *et al.* 2020).

The average concentration of Zn was $0.23 \pm 0.41 \text{ mg L}^{-1}$ with a range between 0.005 and 1.32 mg L^{-1} (Table 1). Surprisingly, none of the samples surpassed the provisional limits of the WHO (3.0 mg L^{-1}), the US EPA (5.0 mg L^{-1}), and the BDWS (5.0 mg L^{-1}), indicating that the water is safe for drinking purposes. In a nearby district of this study area, Zn concentration was reported to be $0.42 \pm 0.26 \text{ mg L}^{-1}$ (Rakib *et al.* 2020), which is about twice that of this study. However, in Assasuni Upazila of the Satkhira district, groundwater was contaminated with 0.034 mg L^{-1} Zn (Rahman *et al.* 2019), which is much lower than this study.

Various investigations have been conducted on the effects of Mn on children's health and behavior through exposure to DW. The wide range of threats included cognitive, behavioral, and neuropsychological problems, memory loss or intellectual impairment of school-aged pupils (Bouchard *et al.* 2011; Khan *et al.* 2011, 2012; Oulhote *et al.* 2014). In addition, Khan *et al.* (2012) experimented on Mn exposure through DW, documenting the behavior and academic achievement of school children, and hypothesized that a 6.4% score loss (lower performance) was very evident in language learning (Bangla and English), science and mathematics when compared with non-exposed children. At the school level, the high rate of failure in English and Mathematics in the rural areas of Bangladesh has been evident for a long time, particularly in those regions where rural people rely heavily on groundwater through which natural manganese is transported. It is not only Bangladesh that is affected by this problem; other countries have observed the same issue. For example, in China, children exposed to Mn were found to exhibit significantly more unsatisfactory school performance in mathematics and language compared with non-exposed village children (Zhang *et al.* 1995; Khan *et al.* 2012). Moreover, exposure to Mn can alter children's classroom behavior and lead to symptoms of irritability, aggression, impulsivity, and so on (Khan *et al.* 2011). Furthermore, infant mortality within a 1-year age was remarkable after mothers had been exposed to Mn during their pregnancy in Bangladesh (Hafeman *et al.* 2007). Being so hazardous, the WHO (2017), the US EPA (2009), and the BDWS (ECR 1997) have set recommendations for the tolerance level of 0.4, 0.05, and 0.1 mg L^{-1} , respectively. About 8.6% (3 out of 35), 54.3% (19 out of 35), and 42.9% (15 out of 35) of samples surpassed the guideline values of the WHO (2017), the US EPA (2009), and the BDWS (ECR 1997), respectively (Table 1). The average Mn was $0.19 \pm 0.36 \text{ mg L}^{-1}$, which exceeded the guideline values of the US EPA and the BDWS. Therefore, this scenario is liable to cause the above-mentioned diseases, notably, intellectual impairment and infant mortality, which has significantly threatened Bangladesh's economic development, given that it will rely on investment in its human potential. In the groundwater of Satkhira (Asasuni Upazila), the Mn content was found to be 0.14 mg L^{-1} (Rahman *et al.* 2019), which is very close to the Mn content found in this study. However, in coastal areas, groundwater contained an average Mn content of $0.47 \pm 1.41 \text{ mg L}^{-1}$ (Islam *et al.* 2020), which is about three times larger than this study.

Other trace metal(loid)s, namely As, Pb, and Cd, were present in very minute quantities. The average As concentration was $0.0024 \pm 0.01 \text{ mg L}^{-1}$ with a range from 0.0004 to 0.0035 mg L^{-1} (Table 1). Although groundwater in Bangladesh is highly contaminated with As (reported previously), none of the samples exceeded the provisional limits of the WHO, the US EPA, and the BDWS in the present study (Table 1). However, about 17 times higher (than WHO provisional guideline) As concentration (0.0168 mg L^{-1}) was reported in TWW in primary schools in the Satkhira district (Rahman & Hashem 2019). Another study from the Shyamnagar sub-district in the Satkhira district reported 0.0166 mg L^{-1} As in groundwater which is about 1.6-fold higher than the WHO guideline value (0.01 mg L^{-1}) (Rakib *et al.* 2020). The low As concentration in the current study area was probably favored by nearly neutral pH (7.85 ± 0.40) of groundwater. It is well known that under aerobic conditions and neutral pH, the release of arsenic into aqueous media is low since As is strongly absorbed on iron oxides (Pal *et al.* 2009). Another reason may be the carefully chosen sample sites (As-contaminated sites avoided by following the previously published report) where installation of new tubewells had been carried out by government institutions or NGO's in the study area.

The Pb and Cd concentrations were below the AAS detection limit, indicating safe DW in terms of Pb and Cd in the study area. Previous study also reported that Cd in groundwater was below detection level and very limited Pb

concentration ($<0.034 \text{ mg L}^{-1}$) in the coastal region (e.g., Satkhira, Noakhali district, and Chandpur districts) (Zahid *et al.* 2008; Rahman *et al.* 2015, 2019) which is similar to our study area. The low level of Pb and Cd in the study area might be related to the geology of the study area. The sediment is mainly composed of quartz with some plagioclase and potassium feldspars, with some fragments of volcanic, metamorphic, and sedimentary rocks (Uddin & Lundberg 1999; Halim *et al.* 2010). However, detailed data are not available for the release and retention of Pb and Cd in the study area. Another possibility is that a very low amount of Cd and Pb minerals are present in the aquifer, and probably the slightly basic nature of water inhibits their release into groundwater.

Correlations among studied parameters

Correlation analysis provides information about the sources, chemistry, and strength of correlation among the contaminants in groundwater. Therefore, Spearman's correlation coefficients for physicochemical properties and trace metal(loid)s of groundwater quality in the study region are presented in Table 3 to establish the strength of their relationships.

Here, pH showed a negative correlation with all the other variables, except As and Mn. Although the correlations were negative, pH vs. turbidity ($r = -0.72$), pH vs. alkalinity ($r = -0.42$), Fe ($r = -0.48$), and Zn ($r = -0.54$) were significant. However, pH vs. As ($r = 0.05$) and pH vs. Mn ($r = 0.18$) correlations were very weak and insignificant. Therefore, it is apparent that pH conversely affects the physicochemical properties, Fe, and Zn chemistry in groundwater in the study area. A very strong correlation of EC with TDS ($r = >99$), chloride ($r = 0.98$), and hardness ($r = 0.83$) was observed. In addition, a significant and strong correlation of TDS vs. chloride ($r = 0.98$), TDS vs. hardness ($r = 0.83$), turbidity vs. hardness ($r = 0.35$), and chloride vs. hardness ($r = 0.87$) was observed (Table 3). Consequently, it can be inferred that the EC of groundwater is dependent on TDS, chlorides, and other cations contributing to hardness.

Among the trace metal(loid)s, only Fe vs. Zn ($r = 0.35$) correlation was significant but weak. However, trace metal(loid)s, for example Fe, showed a significant and strong correlation with turbidity ($r = 0.77$), a moderate correlation with hardness ($r = 0.50$), and a weak correlation with chloride ($r = 0.35$) (Table 3). Therefore, it might be inferred that turbidity is contributed by Fe content in groundwater.

To check the regional impact on the correlation of the studied parameters, Spearman's correlation was performed for samples in Digholia (D region), Terokhada (T region), Batiaghata and Dumuria (B region) Upazilas in the Khulna district. In all regions, pH indicated negative correlations with most of the studied parameters, while in the D region, pH vs. alkalinity ($r = 0.64$) (Supplementary Table S3), in the T region, pH showed a positive correlation with alkalinity ($r = 0.25$), As ($r = 0.66$), and Zn ($r = 0.54$) (Supplementary Table S4), and in the B region, it is with alkalinity ($r = 0.17$), As ($r = 0.31$), and Mn ($r = 0.16$) (Supplementary Table S4). Interestingly, in all regions, pH vs. alkalinity correlation was positive, but the strength of correlations was significantly different. Thus, it is apparent that pH controls the alkalinity of groundwater differently in different regions. Similarly, other parameters indicated a difference (either small or large) in correlations with other parameters in each region (Supplementary Tables S3–S5). Therefore, it can be said that the groundwater chemistry differs from one region to other regions, even in a small study area.

Human health risks

Human health risks were estimated through the non-carcinogenic (HQ) and carcinogenic risk (CR) assessment method provided by the US EPA. The non-carcinogenic health risks of school-going children due to exposure to As, Fe, Mn, and Zn from DW were estimated in terms of CDI, HQ, and HI for school-going children, as presented in Figures 2–4, respectively. The highest CDI was observed for Fe (Boys: 0.09 ± 0.08 ; Girls: $0.11 \pm 0.10 \text{ mg kg}^{-1} \text{ d}^{-1}$), while the lowest was observed for As (Boys: 0.00010 ± 0.0002 ; Girls: $0.00012 \pm 0.0003 \text{ mg kg}^{-1} \text{ d}^{-1}$) in DW. Considering gender differences, girls intake a larger amount of each element than boys, indicating that girls are more vulnerable to trace metal pollution in DW in the study area (Figure 2).

The HQs from As, Fe, Mn, and Zn intake and HIs were lower than unity for both boys and girls (Figure 3). These indices imply that neither the individual trace elements nor a combination poses a threat to the school-going children. Among the studied elements, the highest HQ was from As intake and the decreasing patterns of HQs of individual trace metals were as follows: As (average: 0.32 ± 0.78 and 0.40 ± 0.96 for boys and girls, respectively) $>$ Fe (average: 0.13 ± 0.12 and 0.16 ± 0.14 for boys and girls, respectively) $>$ Mn (average: 0.05 ± 0.11 and 0.07 ± 0.13 for boys and girls, respectively) $>$ Zn (average: 0.03 ± 0.06 and 0.04 ± 0.07 for boys and girls, respectively) (Figure 3).

Table 3 | Spearman's correlation among the physicochemical parameters and trace metal(loid)s content in tubewell water samples

Parameters	pH	EC ($\mu\text{S cm}^{-1}$)	TDS (mg L^{-1})	Turbidity (NTU)	Chloride (mg L^{-1})	Alkalinity (mg L^{-1})	Hardness (mg L^{-1})	As (mg L^{-1})	Fe (mg L^{-1})	Mn (mg L^{-1})	Zn (mg L^{-1})
pH	1										
EC ($\mu\text{S cm}^{-1}$)	-0.15	1									
TDS (mg L^{-1})	-0.15	1.0**	1								
Turbidity (NTU)	-0.72**	0.19	0.19	1							
Chloride (mg L^{-1})	-0.08	0.98**	0.98**	0.19	1						
Alkalinity (mg L^{-1})	-0.42*	-0.30	-0.30	0.18	-0.37*	1					
Hardness (mg L^{-1})	-0.23	0.83**	0.83**	0.35*	0.87**	-0.49**	1				
As (mg L^{-1})	0.05	-0.09	-0.09	-0.06	-0.12	-0.02	-0.11	1			
Fe (mg L^{-1})	-0.48**	0.32	0.32	0.77**	0.34*	0.06	0.50**	0.02	1		
Mn (mg L^{-1})	0.18	0.19	0.19	0.05	0.21	-0.34*	0.35*	0.33	0.30	1	
Zn (mg L^{-1})	-0.54**	0.01	0.01	0.48**	-0.05	0.30	0.05	0.30	0.36*	0.27	1

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

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

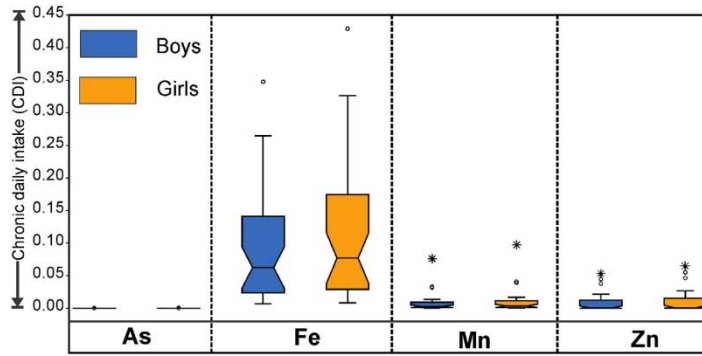


Figure 2 | Chronic daily intake (CDI) of As, Fe, Mn, and Zn ($\text{mg kg}^{-1} \text{d}^{-1}$) through water ingestion for the school-going children of primary schools in the Khulna district, Bangladesh.

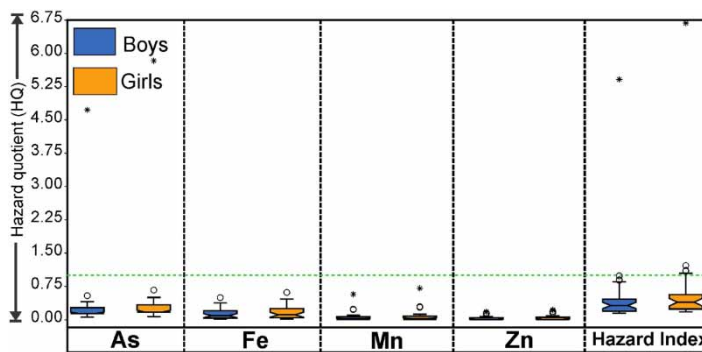


Figure 3 | Hazard quotient (HQ) of As, Fe, Mn, and Zn through water ingestion for the children of primary schools in the Khulna district, Bangladesh (dotted green line indicates the safe level for non-carcinogenic risks). Please refer to the online version of this paper to see this figure in color: <https://doi.org/10.2166/washdev.2021.016>.

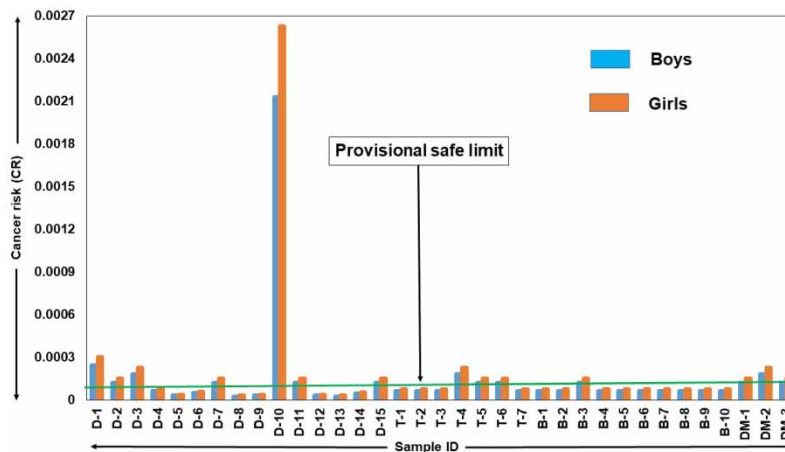


Figure 4 | Cancer risk (CR) of As through water ingestion for the school-going children of primary schools of Khulna districts in southwestern Bangladesh.

There are no absolute criteria for the acceptable number of additional cancers over a lifetime period. However, the US EPA generally adopts one additional cancer case in 1 million (i.e., 1.0×10^{-6}) as a management goal for the government to suggest the point at which management decisions should be taken. The cancer risk surpassing 1.0×10^{-4} (one case of cancer in 10,000) is considered unacceptable (US EPA, 2005). In this study, the average CR from As intake through DW was 1.5

and 1.8 times higher than the provisional safe value of 1.0×10^{-4} for boys (average: 1.5×10^{-4}) and girls (average: 1.8×10^{-4}), respectively, indicating that the students are prone to lifetime cancer risks from drinking As-contaminated water. For boys, the lowest CR from As intake from DW was found to be 2.4×10^{-5} for sample D-13, while the highest was 2.13×10^{-3} for sample D-10 (Figure 4). In addition, the lowest CR was found to be 3.0×10^{-5} through As intake from DW by sample D-8 for girls in the study area, while the highest value was 2.6×10^{-3} for sample D-10 (Figure 4). It is evident that girls are more prone to cancer risk from the drinking of As-contaminated DW than boys. About 40% of the samples (14 out of 35) showed a higher level of CR values than the provisional safe limit of 1.0×10^{-4} (Figure 4). Consequently, it is unlikely to expect that As exposure causes neurological disorders including a range of cancers of the skin, liver, lung, kidney, and bladder to the exposed population who are drinking As-contaminated water (Smith *et al.* 1992; Jain & Ali 2000; Mandal & Suzuki 2002; Ferreccio *et al.* 2013).

Limitations of this study

This preliminary estimation of the health risk to children was considered via only oral exposure through consumption of drinking water, even though some risk could be imposed through dermal exposure during hand washing. The health risk data were estimated for children (5–10 years) through intake of As-contaminated drinking water up to 5 years' exposure duration where the exposure was considered during school time only (6 h). Dermal and inhalation As-exposures are not considered in the current study. Other sources of As-exposures, for example, airborne particular matter, dust in the classroom and school premises, dietary and other supplementary foods, were not considered for the assessment of health risk in this current study. Toxic trace metals such as lead, cadmium, and chromium in drinking TWW were also not considered in this study, and these would warrant further research to provide a complete scenario of trace metals contamination in coastal areas. Additionally, larger sample sites with different frequencies (such as seasonal variations up to 5 years) would be characterized as the real cancer risk assessment in future monitoring research of children in coastal schools.

CONCLUSIONS

This study examined 35 TWWs operated in different primary schools at a vulnerable coastal region in the Khulna district, Bangladesh, for their effects on children's health; what they contained regularly breached the guidelines recommended by the WHO, the US EPA, and the BDWS both collectively and individually. The average data concerning EC, TDS, turbidity, chloride, and hardness were beyond the guidelines that could affect the physical and mental health of children. Based on TDS content, about 34.3% of TWWs are very poor in quality, and 25.7% are unacceptable. Some TWWs contain very high levels of chloride (74.3 and 48.6% of TWWs surpassed the WHO and BDWS standards), Mn (54.3 and 42.9% of TWWs exceeded the guideline values of the US EPA and the BDWS) and Fe (92 and 66% of the samples crossed the safe limit of the WHO and the BDWS), which indicates they would affect children's health.

The physicochemical variables exude both negative and positive correlations, but in most cases, chloride, EC, TDS, turbidity, and hardness had a robust correlation. Iron was attributed toward turbidity as they showed strong positive correlations. Correlations among pH and other parameters differ from region to region, indicating that the geochemistry of trace metal(-loid)s is influenced by weathering and hydrothermal and geological conditions even in a small study area.

According to human health risk assessment, there are no non-cancer health risks from As, Fe, Mn, and Zn intake to the children during school time. Based on CR assessment, As poses a lifetime cancer risk to the children through DW. Girls are more susceptible to trace metal(-loid)s pollution than boys for both non-carcinogenic and carcinogenic risk. This study indicates the physicochemical quality of TWWs is the first-tier indication of health risk. The long-term monitoring of the selected TWWs in the investigated area should be continued, and the tubewells, which deliver contaminated water, should be sealed to protect the country's future generations. The coastal population is still facing a lack of access to safe DW due to the excessive cost of the water treatment process, and therefore, this result suggests selecting a better option before installing any water-supplying device (e.g., TWWs). The better options may include installing deep tubewells to a layer where trace elements (e.g., As, Fe, Mn, etc.) are typically <WHO limit. Another option may include setting up cost-effective arsenic and iron removal plants (AIRPs) and SONO filters (one type of As treatment plant) that have evidence of reducing As and Fe contents remarkably from TWWs to be safe for drinking. Besides, before drinking water from the existing tubewells, the water quality should be checked and the associated risk that could be imposed from long-term exposure to humans, particularly to the school-going children of the coastal region in Bangladesh, should be justified.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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