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Public health vulnerability due to the exposure of dissolved metal(oid)s in tap water from a mega city (Dhaka, Bangladesh)

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Abstract

Tap water is the sole source of water in urban cities but gradually being polluted by potentially toxic elements due to unplanned urbanization. The south Asian megacity, Dhaka (capital of Bangladesh) encounters drastic water pollution resulting mainly from anthropogenic differential factors. Consumption of such polluted water distributed to residents by pipelines can trigger health risks. Therefore, this study investigated the public health vulnerability associated with dissolved metal(oid)s in tap water collected across Dhaka city. The concentrations of Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Hg and Pb in measured tap water ranged from 8-156, 7-73, 400-20100,

12-110, 7-101, 12-136, 12-908, 0.03-9.75, 1-5, 0.22-1.30, and 8-118 $\mu\text{g/L}$, respectively. Among the observed elements, Pb, Cr, and Fe concentrations in 18%, 26%, and 75% of sampling sites, respectively, exceeded the standard guideline criteria of Bangladesh. Entropy-based water quality index demonstrated that ~12% of sampling sites possessed water unsuitable for drinking and other household works. The dominant sources of water pollution in this region are industrial effluents and domestic sewage. Both non-carcinogenic and carcinogenic health risks are invoked mostly from ingestion of Co and Pb. Regular consumption of this tap water without further pretreatment may result in detrimental health consequences to both children and adults due to the physiological accumulation of toxic elements over time. This study highlighted a comprehensive scenario of the potentially toxic elements in the tap water of Dhaka city, which will allow policymakers to take adequate measures for sustainable water quality management.

Keywords: Tap-water; Dhaka city, Bangladesh; Dissolved potentially toxic elements; Water quality appraisalment; Health risks estimation.

1. Introduction

Dhaka is ranked as the 19th megacity (300 km² area) around the world with more than 18 million people and a high population density (~23,234 people/km²) (United Nations, 2016). Ensuring a safe water supply to this vast population is a real challenge. Dhaka Water Supply and Sewerage Authority (DWASA) is currently running with a daily production capacity of ~ 2,550-million-liter water per day using 887 deep wells and three water treatment plants (Haque, 2006; DWASA, 2019). In addition, more than 2000 private water pumps are abstracting water from different depths (DWASA, 2013; 2019). Those waters are distributed to city dwellers through the pipelines called tap-water.

Tap water (commonly known as faucet water, running water, or municipal water) is substantially distributed to the stakeholders of Dhaka City through the extraction of groundwater (78%) or the treatment of the surface water (22%). Almost 95% of Dhaka City dwellers depend upon this tap water for their daily household and other seminal purposes, including drinking, bathing, cooking, washing, toilet flushing, construction, and gardening (DWASA, 2019). Also, the supplied tap water is being consumed by lower and lower-middle-income families without any purification (Sharmin et al., 2020). Rising water demand by increasing population causes the

annual incremental rate of groundwater extraction (Rahman et al., 2013), which leads to the depletion of groundwater table at least 2-3m per year (Haq, 2006; Hoque et al., 2007; Hossain et al., 2018; Bodrud-Doza et al., 2019a, b). Heavy industrialization, rapid urbanization, transportation, uncontrolled sewage discharge, and intensive agricultural activities adversely influenced the water supply, aquifer recharge, and peripheral rivers around Dhaka City (Rahman et al., 2013; Islam and Azam, 2015; Islam et al., 2016; Hossain et al., 2018; Bodrud-Doza et al., 2019b; Fakhri et al., 2017, 2018a, b; Ghasemidehkordi et al., 2018).

Sustainable Development Goal-6 (SDG-6), aims to ensure availability and sustainable management of fresh water and sanitation for all by 2030 (UN Water, 2018). Still, more than 80% of people in Bangladesh lack clean and safe water (Yeazdani, 2016), and the uncontrolled existence of metal(oid)s like Cr, Cd, Pb, As, etc., are making the SDS-6 goal harder to achieve (UNICEF, 2009). Some metal(oid)s such as iron, zinc, and copper are necessary for physiological activities, but their bioaccumulation in excess is not desirable. On the contrary, some metal(oid)s such as arsenic, lead, cadmium, chromium, and mercury have no valuable functions in the body, and their increased concentration can cause toxic effects on the body tissues (Armitage et al., 2007; Yuan et al., 2011; Singh, et al., 2011; Gbadamosi, et al., 2018). Exposure to toxic metals may trigger numerous health risks including neurotoxicity, cardiovascular disease, renal problem, and reproductive failure (Fakhri et al., 2018a, b). Lead and Cd are well known for their long residence time in the human body and causing gastrointestinal inflammation, blood cerebral diseases, and pain in the bone (Itai-Itai disease) (Flora et al., 2012; Fagerberg et al., 2017). Therefore, continuous monitoring of these toxic elements in water and other environmental matrices is a prerequisite to safeguard human health. However, the types and severity of toxic effects are not proportional to the concentrations of toxic elements. Some toxic elements with low concentration exhibit much more significant health effect than others with high concentration (Adel et al., 2016; Çelebi et al., 2014; Saha and Zaman, 2013; Atapour 2012; Malakootian et al., 2014). The adverse effects of toxic elements may also vary depending on the physiological aspects of human health.

Few studies attempted to assess the groundwater quality of Dhaka and the probable health risks of Dhaka city dwellers (Sharmin et al., 2020; Bodrud-Doza et al., 2020; Sabrina et al., 2013). Bodrud-Doza et al. (2020) demonstrated that the children of the western part of the city were more prone to Fe and Mn exposure. According to Sharmin et al. (2020), the sublime concentration of

Pb and As made 6.4% of groundwater unsuitable for drinking, and the residents who reside in the central-western portion of the city were more vulnerable to carcinogenic risks.

Although the recent studies focused on the groundwater quality and associated health risks in Dhaka city (Sharmin et al., 2020; Bodrud-Doza et al., 2020), a detailed investigation of supplied tap water quality in relation to dissolved metal(oid)s composition, and evaluation of associated health risks are yet to be done. The consumers of tap water in Dhaka city (~ 95% of total population) are often unsatisfied with the quality of supplied water due to its color and smell, and the levels of the toxic element are poorly known. It should be mentioned that the concentrations of metal(oid)s in the water in the water plants could be different than that of tap water due to corrosion-induced alteration of chemical composition during transportation through supply system (Faier et al., 2009; Kavcar et al., 2009; Khan et al., 2015; Shanbehzadeh et al., 2014). Considering the importance of this issue, the present study was aimed to (1) quantify the concentration of eleven metal(oid)s (Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Hg, and Pb) in tap water, (2) utilize the entropy-based water quality index for differentiating the tap water quality, (3) evaluate the potential sources and distributions of analyzed metal(oid)s using statistical and spatial mapping approaches, and (4) appraise probable cancer and non-cancer health risks for the city dwellers. To the best of our knowledge, this is the first study of its kind in the present study area. The outcomes of this study will help the government, responsible authorities, and policymakers undertake better management strategies and achieve the goal of SDG-6.

2. Experimental and Methodologies

2.1 Study area

Dhaka City stands in between 23.8103° N, 90.4125° E which is located almost in the middle of Bangladesh. According to DCC (Dhaka City Corporation), the city is covering an area of total 306.4km² and is bounded by a number of rivers, lakes, and differential water reservoirs. Major rivers in and around this city are Buriganga (south and west), Turag (northwest), Shitalakhya (northwest), Dhaleshwari (south) and Balu (east) (Islam et al., 2015). Among other water bodies Gulshan Lake, Dhanmondi Lake, Hatirjheel, Tongi-khal have a substantial impact on the hydrological cycle. A relatively flat topographic setting has the most advantageous outcome and its elevation varies from 0.5-12m to the mean sea level (Hoque et al., 2007). Total four seasons circulate throughout the year like other parts of the country; around March to May is pre-monsoon,

June to September is monsoon, October to November is post-monsoon and December to February is the dry season. The estimated average annual rainfall is over 2000 mm from which nearly 80–90% occurs during the monsoon period (Banglapedia, 2018). Monthly average temperatures range between 25–31 °C and average humidity and evaporation range between 80–90%, and 80–130 mm respectively (BMD, 2016). Water-logging, unplanned sewage system, untreated industrial waste disposal, drainage congestion, lack of management of domestic waste, etc. are a quite familiar scenario of the city for a long time (Rahman et al., 2012; Islam and Azam, 2015; Islam et al., 2016; Bodrud-Doza et al., 2020). Fig. 1 shows the exact location by coordinates of the sampling sites from Dhaka City where purple circles indicate the sampling points.

2.2 Sample collection, processing, and analysis

A total of 34 household tap-water samples were collected from thirty-four different locations (Fig. 1) of Dhaka metropolitan city. However, required sample information and ancillary data for the analyzed tap water samples are tabulated in Table 1. All samples were collected during September 2020 (monsoon). Water samples were collected in thoroughly prewashed 1L capacity high-quality polyethylene water bottles. At the time of sampling, faucets were opened for ~5 minutes; hereafter, water bottles were rinsed several times with sample water of a particular location. Sampling bottles were then completely filled with the tap-water sample and closed the cap tightly. Probable all types of precautions were taken to omit the unenvied contamination during sampling and rinsing. However, the overall course of sampling was simple random sampling where all the locations were selected randomly; thus, the process of sampling can also be deliberated as cluster area sampling. In the analytical laboratory, Flame Atomic Absorption Spectrophotometer (AAS-6800, Shimadzu Corporation, Japan) was used to analyze the elemental concentrations of Cr, Mn, Fe, Co, Cu, Zn, and Pb in the sample water after consecutive acid digestion with concentrated HNO₃ acid followed by required dilutions (Ahmed et al., 2021; Rahman et al., 2020). Besides, concentrations of Ni and Cd were determined by Zeeman-AAS (Model-GTA 120-AA240Z, Varian, Australia) whereas elemental abundances of As and Hg in tap-water samples were measured by Hydride vapor generation technique of AAS (Model-Spectra AA 220 equipped with ETC-60 & VGA-77, Varian, Australia). The quality control schemes applied in this research were, however, exactly the same as in previous studies (Ahmed et al., 2021; Habib et al., 2020; Islam et al., 2020; Ahsan et al., 2019).

2.3 Water quality index

Shannon (1948) expressed the concept of entropy as a criterion of measuring the information or uncertainty that can predict the outcome of a probabilistic occurrence (Guey-Shin et al., 2011). Herein, the following steps are involved to appraise the water quality by using the entropy principle (Islam et al., 2020). For estimating the entropy weight for m ($i = 1, 2, \dots, m$) number of water samples with n ($j = 1, 2, \dots, n$) number of analyzed parameter, Eigen-value matrix X can be computed by eq. (1).

$$X = \begin{bmatrix} X_{11} & X_{12} & \cdots & X_{1n} \\ X_{21} & X_{22} & \cdots & X_{2n} \\ \vdots & \vdots & \ddots & \vdots \\ X_{m1} & X_{m2} & \cdots & X_{mn} \end{bmatrix} \dots\dots\dots (1)$$

To diverge the influences of various units and contents of analyzed parameters, efficiency type (eq. 2) normalization approach (Pei-Yue et al., 2010) was obtained to convert the eigen value matrix (X) into a standard grade matrix (Y) (eq. 3).

$$Y_{ij} = \frac{X_{ij} - (X_{ij})_{\min}}{(X_{ij})_{\max} - (X_{ij})_{\min}} \dots\dots\dots (2)$$

$$Y = \begin{bmatrix} Y_{11} & Y_{12} & \cdots & Y_{1n} \\ Y_{21} & Y_{22} & \cdots & Y_{2n} \\ \vdots & \vdots & \ddots & \vdots \\ Y_{m1} & Y_{m2} & \cdots & Y_{mn} \end{bmatrix} \dots\dots\dots (3)$$

Then the ratio of the analyzed parameter index (P_{ij}), information entropy (e_j), and entropy weight (ω_j) can be calculated by eqs. (4-6).

$$P_{ij} = \frac{Y_{ij}}{\sum_{i=1}^m Y_{ij}} \dots\dots\dots (4)$$

$$e_j = -\frac{1}{\ln(m)} \sum_{i=1}^m (P_{ij} \times \ln P_{ij}) \dots\dots\dots (5)$$

$$\omega_j = \frac{1 - e_j}{\sum_{j=1}^n (1 - e_j)} \dots\dots\dots (6)$$

Quality rating scale for the analyzed parameter (j) can be calculated from the obtained data (C_j) and the standard data (S_j) by eq. (7).

$$q_j = \frac{C_j}{S_j} \times 100 \dots\dots\dots (7)$$

From eqs. (6) and (7), EWQI can be determined by eq. (8).

$$EWQI = \sum_{j=1}^n \omega_j q_j \dots\dots\dots (8)$$

Here, the estimation of two significant parameters including information entropy (e_j) and entropy weight (ω_j) are tabulated in Table 2 those are required to elucidate the water quality in terms of EWQI. According to the EWQI value, water samples can be classified into five grades based on the suitability of human consumption. Herein, grade one ($EWQI < 50$) is excellent to use for drinking purpose; followed by grade two ($50 \leq EWQI < 100$) which is good and suitable for drinking; grade three ($100 \leq EWQI < 150$) is moderate and suitable for domestic, irrigation, and industrial uses, albeit require further treatment for using in drinking purpose, grade four ($150 \leq EWQI < 200$) is earmarked as poor and not suitable for drinking; and lastly, grade five ($EWQI \geq 200$) which is considered as extremely poor quality water and thoroughly inadmissible for human consumption (Islam et al., 2020; Siddique et al., 2021; Ahmed et al., 2021).

2.4 Statistical approaches

The measured analytical parameters were evaluated statistically by SPSS-software (Version-20, IBM-Corporation, Armonk, NY, USA). To determine the important factor, inter-relationship, specific pollution source of the tap-water, multivariate statistical methods (Pearson correlation analysis, cluster analysis, and principal component analysis) were applied in the present study (Khan et al., 2019; Ahsan et al., 2019; Islam et al., 2018). Principal component analysis (PCA) with a Varimax rotation method was utilized to extract principal components (PCs) from the sampling points, to evaluate spatial variations and possible source of pollution in groundwater, and to determine the degree of pollution (Bodrud-Doza et al., 2016; Islam et al., 2018). On the other hand, cluster analysis was performed with Ward algorithmic method and rescaled linkage distance of similarity calculates the similarity among elemental components from different sources with the help of various sample groups (Lattin et al., 2003; Bhuiyan et al., 2016). Moreover, the dendrogram contributed by giving a visual summary of a different cluster as well as their proximity (Bodrud-Doza et al., 2016). Pearson correlation analysis was executed to show the relevant associations among the analyzed parameters in a certain cluster (Islam et al., 2019).

2.5 Health risk estimation

Health risk estimation is the likelihood of any given enormity of adverse health effects amid a specific time (Bortey-Sam et al., 2015; Bodrud-Doza et al., 2020). This estimation is usually based on the risk level ascertainment and disclosed by carcinogenic and non-carcinogenic health

risks (USEPA, 2009). Direct oral ingestion and dermal absorption by the skin are generally considered for the evaluation of health risks induced by trace elements in water (Zeng et al., 2015). According to USEPA (2004), the exposure doses for direct ingestion ($ADD_{\text{ingestion}}$) and dermal absorption (ADD_{dermal}) are expressed as bellow:

$$ADD_{\text{ingestion}} = \frac{C_w \times IR \times Abs_g \times EF \times ED}{BW \times AT} \dots\dots\dots(9)$$

$$ADD_{\text{dermal}} = \frac{C_w \times SA \times K_p \times EF \times ET \times ED \times 10^{-3}}{BW \times AT} \dots\dots\dots(10)$$

Where, C_w , IR, EF, SA, ET, ED, BW, AT, Abs_g , and K_p represent the concentrations of trace elements (μgL^{-1}), ingestion rate (L.day^{-1}), exposure frequency (days.year^{-1}), exposed skin area (cm^2), exposure time (h.day^{-1}), exposure duration (in years), body weight (in kg), average time for non-carcinogens (days), gastrointestinal absorption factor, and dermal permeability coefficient (cm.h^{-1}), respectively. The values of each parameter are tabulated in Table S1. Besides, non-carcinogenic health hazard can be calculated from the estimation of Hazard Quotient for both oral ingestion and dermal exposure from the following eqs. (11-13).

$$HQ_{\text{ingestion}} = \frac{ADD_{\text{ingestion}}}{R_f D_{\text{ingestion}}} \dots\dots\dots(11)$$

$$HQ_{\text{dermal}} = \frac{ADD_{\text{dermal}}}{R_f D_{\text{dermal}}} \dots\dots\dots(12)$$

$$R_f D_{\text{dermal}} = R_f D_{\text{ingestion}} \times Abs_g \dots\dots\dots(13)$$

Here, $R_f D_{\text{ingestion}}$ and $R_f D_{\text{dermal}}$ are the reference doses for individual elements ($\mu\text{g.kg}^{-1}.\text{day}^{-1}$) which are also presented in Table S1 (USEPA, 2011, 2004). However, the Hazard Quotient (HQ) evaluation considers the possible degree of harmfulness for each type of hazard whereas the total potential non-carcinogenic health hazard can be calculated by Hazard Index (HI) from the following eq. (14).

$$HI = \sum_{i=1}^n (HQ_{\text{ingestion}} + HQ_{\text{dermal}}) \dots\dots\dots(14)$$

When the HI value is less than 1, no potential hazard for water pollution, contrariwise, when the value is more than 1, there is a probable risk of hazardous impact to the water. Consequently, non-carcinogenic health hazards can be considered, only when the values of HQ and HI are >1 . Moreover, exposure to more than one metal contaminant may cause additive and/or interactive effects, and hence, accretive health effect from multiple metals' exposure can be calculated by summing Total Hazard Quotient (THQ) value of the individual metal that expressed as Total Target Hazard Quotient (TTHQ) from the following eq. (15) (Fakhri et al., 18a):

$$TTHQ = TTHQ_{Cr} + TTHQ_{Mn} + TTHQ_{Fe} + TTHQ_{Co} + TTHQ_{Ni} + TTHQ_{Cu} + TTHQ_{Zn} + TTHQ_{As} + TTHQ_{Cd} + TTHQ_{Hg} + TTHQ_{Pb} \dots\dots\dots(15)$$

TTHQ value >1 introduces the likelihood of untoward health consequences and indicates the concernment of further investigation approaches and probable preclusive measures whereas TTHQ <1 represents no possible risk of health hazard from the exposure of analyzed metals at present consumption rates (Fakhri et al., 2018a, b).

On the other hand, Carcinogenic Risk (CR) for both oral ingestion and dermal exposure can be estimated from the eq. (16) and (17) whereas total carcinogenic risk can be calculated from the eq (18). Hence, the equations are as follows:

$$CR_{\text{ingestion}} = ADD_{\text{ingestion}} \times CSF_{\text{ingestion}} \dots\dots\dots(16)$$

$$CR_{\text{dermal}} = ADD_{\text{dermal}} \times CSF_{\text{dermal}} \dots\dots\dots(17)$$

$$CR_{\text{total}} = CR_{\text{ingestion}} + CR_{\text{dermal}} \dots\dots\dots(18)$$

Here, CSF is the Cancer Slope Factor (mg/kg/day)⁻¹. In this work, the CR_{ingestion} was estimated for Cr, Ni, As, Cd, and Pb; and CR_{dermal} was estimated for only As (as standard dermal CSF for other metals were not available). However, the CSFs are 0.00038, 0.00091, 0.0015, 0.041, and 0.0085 (mg/kg/day)⁻¹ for ingestion intake respectively, whereas, dermal CSF for As is 0.00366 (mg/kg/day)⁻¹ (Fakhri et al., 2018a, b; Saha et al., 2017; De Miguel et al., 2007; Gao et al., 2019; Siddique et al., 2021; Ahmed et al., 2021). The acceptable or tolerable range of carcinogenic risks is 1.0×10⁻⁶ to 1.0×10⁻⁴ (USEPA, 2004, 2011).

3. Results and Discussion

3.1 Metal(oid)s in tap water and their distributions

Concentrations of metal(oid)s (Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Hg, and Pb) in the analyzed tap water samples are tabulated in Table 2. The concentration of Cr was found with an average of 43.1±44.5 µg/L, which is slightly lower than the national and international recommended limits (Table 2, Fig. 2). However, Cr concentration in some earmarked samples, including W-12 (Bosila), W-20 (Demra), W-1 (Abdullahpur), W-8 (ECB), W-33 (Goran), and W-9 (Monipur school), W-22 (Matuail), W-27 (Badda), W-34 (Mohammadpur) were >2 times higher than the prescribed limits (WHO 2004, 2011; ECR, 1997; EPA 2001; EU 1998; BIS 1991) (Table 2).

However, Fe in all the analyzed tap water samples was found to be significantly higher than the international permissible limits and ~75% of the samples possess much higher Fe than the national recommended value for drinking water (WHO 2004, 2011; ECR, 1997; EPA 2001; BIS 1991) (Table 2, Fig. 2). Cobalt concentrations in several samples including W-7, W-8, W-17, W-26, W-32, W-33, and W-34 were found slightly higher than the permissible limit of WHO, (2011) and USEPA (2004) (Table 2). The concentration of Ni was found within the range of 7-101 µg/L with an average of 30 ± 24 µg/L. Here, only a single sample W-12 (Bosila) was ascertained slightly upper than the national water quality guideline (ECR, 1997) whereas W-22, W-33, and W-34 possess higher Ni-content than the recommended value of WHO (2011) (Table 2). Concomitantly, Cu, Zn, As, and Hg concentrations were determined much lower than the national and international permissible limits (Table 2, Fig. 2). Cd concentrations for all the samples were lower than the national recommended value (ECR, 1997), albeit ~33% of the analyzed samples possess slightly higher Cd-content compared to the permissible limit of WHO (2004, 2011) (Table 2, Fig 2). The average Pb concentration was found 32.9 ± 28.4 µg/L which was upper than the guideline value of WHO (2011), EPA (2001), and EU (1998) (Fig. 2). Besides, six samples (~18%) including W-3 (Ashkona), W-4 (Dakkhinkhan), W-9 (Monipur School), W-14 (Khilgoan), W-26 (Notunbazar), and W-32 (Shankar) were obtained significantly higher than the accepted limit proposed by the ECR (1997) (Table 2).

Moreover, Table 2 also represents the comparison between some existing literature data (Zakir et al., 2020; Saha et al., 2019; Rahman et al., 2013; Bodrud-doza et al., 2020; Kumar et al., 2019; Maw et al., 2020; Abeer et al., 2020; Muhammad et al., 2011; Mohammadi et al., 2019; Qiao et al., 2020; Dong et al., 2020; Nguyen et al., 2020; Le bot et al., 2016; Momot et al., 2005; Szuster-Janiaczek et al., 2018; Abdeldayem, 2019; Opoku et al., 2020; Egbueri et al., 2020) and the determined heavy metal(oid)s concentrations in the tap water samples of the present study. In comparison to literature data, the mean concentration of Fe, Cr, Pb, Co, Ni, Cd were obtained significantly higher than the tabulated literature data, contrariwise, As concentration in the present study were much lower than the literature data. Hence, the above discussions unravel that in the present study the sublime concentrations of Fe, Cr, and Pb in the analyzed tap-water samples are indicating a pernicious threat to the consumers whereas the concentrations of Co, Ni, and Cd were significantly higher than the listed previous studies (Table 2).

Consequently, distributions of both analyzed metal(oid)s and the total population number of the specific sampling area are represented by spatial distribution maps in Fig. 3. The population distribution map shows that the Central-Northern portion of Dhaka city is a significantly most populated area and thus if a particular carcinogenic or non-carcinogenic heavy metal is counted as predominant there, it will affect a huge population.

However, the sublime concentration of Cr, Fe, Ni, Cu, and Cd were found in the Southern part of the city. Principally, several industrial effluent disposals in the surface water including chemical industry, textile industry, production of dyes, wood preservation, leather tanning, chrome plating, manufacturing of various alloys might be liable for the intemperance concentration of Cr, Ni, Cu, and Cd in the tap water where the concentration of Fe naturally depends on the groundwater aquifer (Tumolo et al., 2020; Hossain et al., 2015; Zhitkovich, 2011). Moreover, South-Eastern areas are predominant with several export-oriented industries including textiles, apparel, leather, jute, cement, ceramics, steel mills, etc. (Britannica, 2019). A high concentration of Mn was determined in the three different portions of Dhaka city such as W-3 (Eastern), W-7 (Central), and W-18 (Western). Albeit, naturally occurring Mn in the groundwater is considered as the principal source of Mn in the tap-water (Frisbie et al., 2012). The maximum affluence of Co, Cu, and Pb were found in the Western part of the city and thus, the populations of these areas are at the risk of excessive exposure to these elements. Industrial effluents, agricultural and urban run-off, and the geology and geochemistry of a particular area might be responsible for the surpassed concentration of these elements in the tap-water (Atashi et al., 2009). Furthermore, Ni and Pb concentrations were also determined significantly orient in the Northern part whereas the ascendant concentration of Zn was determined in the Eastern part of Dhaka city. Several sampling points from different parts of the city including W-14 (Khilgaon), W-15 (Maradia), W-21 (Jatrabari), and W-23 (Kalshi) were obtained high concentration of Hg which indicating the sources of tap-water of these locations probably contaminated with hazardous waste (Verma et al., 2018). Albeit, in some cases, pipeline complications may be induced due to the contact of soft acidic water with the household plumbing, faucets, and water fixtures that results in corrosion and might be incrementing the concentration of several elements including Fe, Ni, Cu, and Pb in the tap-water (McNeill and Edwards, 2001; WHO, 2005; Björn et al., 2003; USEPA, 2011).

Hence, the spatial distribution of the trace elements revealed that sampling point W-7 (Kafrul), W-21 (Jatrabari), and W-33 (Goran) are immensely loaded with four different types of

trace metal(oid)s and followed by sampling point W-14 (Khilgaon); W-27 (Badda), and W-32 (Shankar) are also stately loaded with three diverse types of trace elements, respectively. As a result, the populations of the above-mentioned areas are at the highest possible risks of metal-inducing hazards.

3.2 Water quality assessment

Tap-water quality has been assessed based on two criteria: (1) Comparing the concentration of the analyzed dissolved metal(oid)s with the national and international recommended guideline values for water; and (2) Quantifying the suitability of tap water for the human consumption using entropy water quality index (EWQI). This study found that the average Fe concentration in the analyzed water samples was obtained more than 3 times higher than the national permissible limit of ECR (1997) and approximately more than 10 times higher than the international standard guideline values proposed by WHO (2011), EPA (2001) and BIS (1991) (Table 2, Fig. 2). Besides, the obtained mean concentration of Ni was found higher than the admissible limit of EPA (2001) and EU (1998) while the average abundances of Pb were determined 3 times higher than the acceptable limit of WHO (2011), EPA (2001) and EU (1998) (Table 2, Fig 2). Hence, only the mean concentration of Fe was found to transgress both the national and international permissible limit and the rest of the metal's average abundances were determined lower than the national admissible standard. However, as per the Environmental Conservation Rule (ECR, 1997), all of the analyzed tap-water samples were contaminated with the presence of excess Fe concentration whereas some specific samples are also contaminated with several trace elements (Cr, Pb, and Ni) (Table 2) in the perspective of Bangladesh.

In addition, to evaluate the pertinence of the collected tap-water samples, the entropy water quality index (EWQI) has been used as it is a more reliable and globally recognized approach compare to the assumption-based weighing method. More importantly, EWQI is the more justified and acceptable technique as it can minimize the relative error by logically identifying the weight of every analyzed parameter (Ahmed et al., 2021; Pei-Yue et al., 2010). Furthermore, the prime impactful analytical parameter can be determined by calculating these two entropy parameters (ω_j and e_j), for instance, any parameter with higher entropy weight (ω_j) and lower information entropy (e_j) value indicating that the parameter has a heavier impact on the general water quality (Siddique et al., 2021; Islam et al., 2020; Gorgij et al., 2017). The results of these two entropy parameters in

Table 2 show that Cr, Fe, and Pb have the higher ω_j value and lower e_j value, implying that these elements have maximum impact on the water quality relative to the other analyzed metal(oid)s. Hence, based on the obtained values of entropy weight (ω_j) and information entropy (e_j) (Table 2), the effects of variables on overall water quality are followed in the decreasing order: Cr>Fe>Pb>Zn>Ni>As>Mn>Hg>Cu>Cd>Co.

The value of EQWIs for every tap water sample has been calculated by using eqs. (1-8) and later, ranked these samples based on obtained EWQI values. And however, this index classification provides a firm stance of water decency by ascertaining only a single number in a relatively simple way. Here, Fig. 4a depicts the status of all the tap-water samples whereas Fig. 4b represents the distribution of EWQI-values over the different sampling points. According to the rank of EWQI standard value, sampling point W-32 (Shankar) and W-24 (Kotwali) are categorized as 'extremely poor' while W-18 (Lalbagh) and W-33 (Goran) are earmarked as 'poor' quality water and totally unsafe for human consumption without any treatment (Fig. 4a). Besides, EWQI values of sampling point W-13 (Tejgaon), W-9 (Monipur School), W-1 (Abdullahpur), W-14 (Khilgaon), and W-7 (Kafrul) are found within 100-150, thus, they are classified as 'moderate' quality water that can be used for domestic, irrigation, and industrial purposes but require further treatment for drinking; subsequently, rest of the samples are categorized as 'good' and 'excellent' quality of water which are completely suitable for drinking (Fig. 4a). Thus, ~73.6% (W: 2-6, 8, 10-12, 15-17, 19-23, 25-31, and 34) of tap-water samples are safe and suitable for direct human consumption (marked as 'excellent and good') whereas ~14.7% (W: 1, 7, 9, 13, and 14) of samples are suitable for usage in household and other purposes (marked as 'moderate'), and ~11.8% (W: 18, 24, 32, and 33) of samples are thoroughly impermissible for drinking (marked as 'poor and extremely poor') based on the EWQI rankings (Table 2 and Fig. 4). In addition, the distribution map (Fig. 4b) represents that extremely poor, poor, and moderate-quality tap water are predominant in the South-Western and Southern parts. Contrariwise, good and excellent quality tap water are flourishing in the Eastern and Northern part of the city.

3.3 Source apportionment

Principal component analysis (PCA) works as a substantial tool in finding the pattern of resemblance among a set of observations (Abdi & Williams, 2010). PCA was used to identify the possible source and groupings of the analyzed heavy metals in the collected tap water samples.

PCA extracted five factors from the measured metal(oid)s in water with Eigenvalues > 1 which is shown in the scree plot (Fig. S1) (Liu et al., 2013). The initial data dimensions of the analyzed elements are reduced into five loading factors without considerable loss of data and about 69.15% of the total variance is explained by those factors (Table S2). PC1 which is accounted for 15.76% of the total variance, showed strong positive loadings for As and Hg. Arsenic and mercury contamination in the studied area might have come from paint, pharmaceutical, paper, and pulp preservatives, chlorine, and caustic soda production industries, fertilizer and pesticide industries (Aradhi et al., 2009; Krishna and Mohan, 2014; Morais et al., 2012). Pollutant disposal from this kind of pollutants may be possible in the surface water bodies of Dhaka city from where water is supplied to the households after treatment. Hence, probable inefficient removal of pollutants in the treatment plants may be another issue. PC2 explained 14.96% of the total variance showing strong positive loadings for Cr and Ni. Metal industries, leather and tanneries, painting, and cement industries are the primary sources of Cr in the water (Martin and Griswold, 2009). Ni could be present due to both anthropogenic and natural sources like vehicle exhaust, domestic effluents, or from weathering process due to disintegration of parent mineral (Tatsi et al., 2015). Both Ni and Cr can be released in the tap water system from corrosion of galvanized metallic pipes (Peng et al., 2012). The positioning of these two elements in the same PC may have been resulted from the dominance of corrosion from distribution system. On the contrary, PC3 accounted for 13.35% of total variance with the moderate loadings for Mn, Fe, and Pb. Generally, Fe and Mn occur naturally in groundwater because of dissolution from minerals (Wendland et al., 2005). They can also be generated from mining and industrial wastes. Fe and Pb might be present in water due to contamination from metal and alloy industries or these may come from poor plumbing works while transporting (Bhuiyan et al., 2011, 2016). PC4, explaining 13.28% of the total variance showed a strong positive loading for Cu and Zn. The simultaneous high loadings of these two elements may occur from galvanic corrosion (Cartier et al., 2012). Presence of Zn and Cu also suggest the influence from domestic sewage (Wang et al., 2017) or other forms of organic materials (Gonzalez et al., 2013). PC5 explained 11.79% of the total variance and showed strong negative loading for Co while strong positive loading for Cd. Presence of cadmium might be attributed to various industrial effluent of chemical, electrical, steel industries, and agricultural runoff (Wu et al., 2009; Huang et al., 2014; Wagh et al., 2018). Cobalt can be present in the aquatic system from both geogenic and anthropogenic causes. The anthropogenic causes for Co in water includes the

production of alloys and chemicals, contamination from sewage, agricultural pollution etc (Kim et al., 2006). However, the opposite loadings between Co and Cd probably reflects the alternative dissolution source or characteristics of these two metals. Similar loadings of element in a component indicates their common source of origin in the water. Depending on the level of analyzed elements, the PCA for 34 sampling sites extracted two components explaining 99.60% of the total variance (Table S3) in which most of the sampling sites loaded strongly and positively.

Based on the information derived from PCA, hierarchical cluster analysis (CA) was also employed to detect spatial similarities of the analyzed elements in the water samples (Ahsan et al., 2019) from the measured parameters (R-mode) and sampling sites (Q-mode) with Ward's method and the Euclidean distance as a measure of similarity (Fig. S2). The elements grouped in CA in each cluster mostly supports the result of PCA which gives validation of the determination.

Pearson's correlation matrices were utilized to identify the associations among the analyzed elements (Tamim et al., 2016; Hasan et al., 2020), and the results are tabulated in Table S4. The strong and significant correlations among the parameters indicate their common sources of origin(s). Most of the correlations show poor and insignificant values indicating less probability of coherence of their source of origin. However, the positioning of the elements in PCA is strongly supported by the correlation analysis since significant correlation has been observed for the parameters from the same PC or cluster (R-type). For instance, Cr and Ni which were loaded positively on PC2 also showed a significant positive association (0.526) in the correlation analysis. The moderate and significant correlation between Cr and Ni indicates the leaching from stainless steel pipes (Schwenk, 1991) or the influence by the contamination from different industries in the source water (Dessie et al., 2021). Cu and Zn which are loaded positively on PC4, showed a significant positive correlation (0.390) among them. Hence, similar source(s) for these two metals can be anticipated. As and Hg showed significant and moderately positive correlation (0.474) between them, and they were positioned in PC1. The other correlations show weak values and insignificance suggesting the absence of coherent sources for different materials.

3.4 Health risk assessment

Possible non-carcinogenic and carcinogenic health risks through ingestion and dermal pathways were estimated for both adults and children. By using risk factors, hazard quotient (HQ)

and hazard index (HI) deduced the overall potential health risk exposure originated from elemental abundances of toxic metal(oid)s in the existing samples all over Dhaka City (Fig. 5).

Non-carcinogenic health appraisals for eleven trace metal(oid)s are depicted in the Fig. 5a, for Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Hg, and Pb. In the case of, HQ_{ing} and HQ_{der} the health risk for metals seemed benign ($HQ < 1$) except for Co presence in children, which exceeded the threshold limit ($HQ = 1$) for both ingestion and dermal ($HQ_{ing} = 1.85$, $HQ_{der} = 1.96$). HQ for ingestion of Co through the intake of sample water also exceeded the safest limit for adults ($HQ_{ing} = 1.28$). Overall, calculated hazard index for non-carcinogenic risk followed a decreasing trend of $Co > Cr > As > Ni > Pb > Cd > Cu > Fe > Hg > Mn > Zn$ (Fig 5a). This indicated that tap water from all the sampling sites could induce Co-related health risk as it surpassed the safe limit of hazard index ($HI_{adult}: 1.35$, $HI_{children}: 3.94$). Even slightly higher concentration than the required level of Co for the human body can provoke potential non-carcinogenic risk, which might associate with rhinitis and dermatitis (Wang et al., 2017; Fang et al., 2014). Individually, the W-7(Kafrul) sample significantly contributed to the Co exposure (Adult: 3.06, Children: 6.09), much higher than the recommended value. For the values of Cr and As, HQ_{ing} , HQ_{der} , and HI are highly close to the threshold limit, which might impose a health risk if not maintained carefully. In case of Cr and As, Cr exhibited higher HI values which could possess greater risk for children ($HI_{children}: 0.75$) compared to adults ($HI_{adult}: 0.16$). This may be related to various skin problems, allergic inflammation (SCHER, 2015), and chromosomal aberrations (O'Brien et al., 2001; Matsumoto et al., 2006). Furthermore, total target hazard quotient (TTHQ) values (Fig. 5b), calculated by combining the HQs from all the measured metal(oid)s of which greater than 1 indicates possible non-carcinogenic human health effects (Fakhri et al., 2018a, b). The TTHQ of children for both ingestion and dermal values is 2.2 and 2.87, respectively, indicating a potential health risk for that age group. On the other hand, $TTHQ_{ingestion}$ and $TTHQ_{dermal}$ values for adults are 1.51 and 0.97, respectively, indicating the risk is higher by ingestion route for adults rather than dermal contact.

Potential carcinogenic health hazards for Cr, Ni, As, Cd, and Pb are evaluated through average daily dose with cancer slope factor (Fig. 5c). The measured parameters for carcinogenic health risk followed a decreasing trend, i.e., $Pb > As > Ni > Cd > Cr$. From Fig. 5c, Pb delineated the highest value for both age groups (adults: 8.96×10^{-4} , children: 1.20×10^{-3}), which surpassed the safe limit of 1×10^{-4} , which might be accountable for arthritis, renal dysfunction, autism, dyslexia, and other birth defects (Martin & Griswold, 2009). From Fig. 5d, calculated total cancer risk (TCR)

is also higher for Arsenic (As) which is much closer to the threshold value for both adult (TCR: 4.71×10^{-5}) and children (TCR: 9.87×10^{-5}) age group indicated that regular use of the tap-water in Dhaka City over some time would increase the probability of cancer. Except for lung, skin, and bladder cancer, chronic arsenic toxicity might cause pigmentation and keratosis (Martin & Griswold, 2009). Altogether, this study elucidates that Co can initiate potential non-carcinogenic health risks, while Pb may instigate potential carcinogenic health risks for both age groups as they exceeded the proposed safest limit of USEPA (2004) and WHO (2011). In addition, potentiality of As associated cancer risk should be a concern as the TCR for this metal(oid) is close to the safety limit. From both categories, children are found in a more vulnerable state according to the health risk estimation of every metal(oid)s.

4. Conclusions

This study assessed the tap water quality of Dhaka city based on the concentration of dissolved metal(oid)s (Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Hg, and Pb) and estimated the potential health risks of the city dwellers. The measured elemental concentrations were within the permissible limit, except Fe, Cr, and Pb. Besides, based on elemental abundances, distributions, and entropy water quality index ranking, sample water of eight different locations including W-7 (Kafrul), W-14 (Khilgaon), W-18 (Lalbagh), W-21 (Jatrabari), W-24 (Kotwali), W-27 (Badda), W-32 (Shankar), and W-33 (Goran) were identified as immensely contaminated and unsuitable for drinking. Hence, the population of these areas was highly amenable to metal-inducing health hazards. On top of that, except above-mentioned sampling points, the remaining water samples were obtained compatible for use in the household, especially for drinking purposes. The statistical analysis revealed that pollution sources could be mainly attributed to anthropogenic activities and pipeline complications; promoted by domestic sewage and industrial effluents dispensed from conventional wastewater treatment plants and sludge disposal, improper landfilling; and corrosion of pipes respectively. The total target hazard quotient suggested that children are more vulnerable compare to adults. The estimated non-carcinogenic health risks associated with oral and dermal exposures of Co, and carcinogenic health risks related to ingestion of Pb surpassed the acceptable limits. Overall, tap water of Dhaka City is contaminated with several trace elements and may impose non-carcinogenic and carcinogenic health risks for the city dwellers. Therefore, regular health check-ups are recommended to ensure healthy lives as regular metal(oid)s consumption

could have a chronic impact due to their bioaccumulation over time. Most importantly, children should be under adequate supervision for minimization of probable health risks. In addition, Dhaka Water Supply and Sewerage Authority (WASA) should constantly monitor the tap water quality in relation to metal(oid)s concentration before and after the distribution due to the pipeline corrosivity. Installing advanced water treatment plants to purify the surface water can reduce the dependency on groundwater for sustainable water supply and management. Circumstantially, this study recommended that the regulatory acts and other associated laws should be implemented appropriately to lessen anthropogenic activities and to secure a sustainable water supply and management scheme for the salubrious lives in Bangladesh.

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The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Code availability Not applicable

Authors' contributions

Sabiha Hossain and Amit Hasan Anik: Methodology, Investigation, sample collections and preparation. Rahat Khan: Conceptualization, Methodology, Validation, Writing - original draft preparation, Writing - reviewing and editing, Supervision. Farah Tasneem Ahmed and Md. Abu

Bakar Siddique: Sample analysis, data curation and interpretation. Abdul Hadi Al Nafi Khan, Narottam Saha and Abubakr M. Idris: Original draft preparation, reviewing and editing. Mahbub Alam: Reviewing, editing and supervision. All authors read and approved the final manuscript.

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Figures

Figure 1

Study area including sampling locations (Dhaka city, Bangladesh).

Compraison of measured metal(oid)s concentration with national/international recommended value

Figure 2

Comparison of measured metal(oid)s concentrations with national/international recommended values.

Figure 3

Spatial distributions of elemental abundances in tap-water of different sampling location along with the population distribution

Figure 4

Water quality assessment of the studied area through entropy water quality index (EWQI).

Figure 5

Estimated health risks indices for the tapwater from Dhaka city (Bangladesh). Non-carcinogenic: (a) health hazard indices and (b) total target hazard quotient (TTHQ); Carcinogenic: (c) health hazard indices and (d) total carcinogenic risk estimation through ingestion and dermal exposure from the elements dissolved in tap-water for children and adults.

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