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Article

Quality Assessment of Bottled and Unbottled Drinking Water in Bangladesh

Md. Ripaj Uddin ¹, Mayeen Uddin Khandaker ^{2,*}, Md. Jainal Abedin ³, Nahida Akter ⁴, AHM Shofiul Islam Molla Jamal ¹, Rokaia Sultana ¹, Shahnaz Sultana ¹, Mehedi Hasan ¹, Hamid Osman ⁵, Basem H. Elesawy ⁶, M. I. Sayyed ^{7,8} and A. Sulieman ⁹

- ¹ Institute of National Analytical Research and Service (INARS), Bangladesh Council of Scientific and Industrial Research (BCSIR), Dhanmondi, Dhaka 1205, Bangladesh; md.ripajuddin@gmail.com (M.R.U.); shofiuljamal@yahoo.com (A.S.I.M.J.); rokaiasultana@rocketmail.com (R.S.); shahnazsultana339@gmail.com (S.S.); hasan.mehedi013@gmail.com (M.H.)
 - ² Centre for Applied Physics and Radiation Technologies, School of Engineering and Technology, Sunway University, Bandar Sunway, Subang Jaya 47500, Selangor, Malaysia
 - ³ Faculty of Public Health, Thammasat University, Bangkok 10200, Thailand; abedinj88@yahoo.com
 - ⁴ Institute of Marine Fisheries Technology, University of Chittagong, Chittagong 4331, Bangladesh; nahida.nipaimsf@gmail.com
 - ⁵ Department of Radiology, College of Applied Medical Sciences, Taif University, Taif 21944, Saudi Arabia; ha.osman@tu.edu.sa
 - ⁶ Department of Pathology, College of Medicine, Taif University, P.O. Box 11099, Taif 21944, Saudi Arabia; b.elesawy@tu.edu.sa
 - ⁷ Department of Physics, Faculty of Science, Isra University, Amman 11622, Jordan; dr.mabualssayed@gmail.com
 - ⁸ Department of Nuclear Medicine Research, Institute for Research and Medical Consultations, Imam Abdulrahman bin Faisal University, Dammam 31441, Saudi Arabia
 - ⁹ Department of Radiology and Medical Imaging, College of Applied Medical Sciences, Prince Sattam Bin Abdulaziz University, P.O. Box 422, Alkharj 11942, Saudi Arabia; a.sulieman@psau.edu.sa
- * Correspondence: mayeenk@sunway.edu.my

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Abstract: The demand for bottled drinking water in Bangladesh is becoming popular due to the lack of safe drinking water sources. The present study was carried out to assess the water quality and public health impacts of local brands of bottled drinking water and tube-well water, compared to the imported ones. Several state-of-the-art techniques were employed to determine the contents of pH, EC, salinity, chloride, nitrates, nitrites, fluoride, sulfates, phosphates, bicarbonate, turbidity, color, total hardness, and total dissolved solids in the studied water samples. The measured data show that the level of dissolved minerals in the local branded bottled water is very low (with an exception for sample codes D1 and D8), while imported brands, Zamzam water, and tube-well water contain satisfactory level of minerals and fulfill Dietary Reference Intake requirements. Total coliform, fecal coliform, and *E. coli* were found to be absent in local brands of bottled water and tube-well water. On the other hand, concentrations of some radionuclides ⁴⁰K, ²³⁸U, ²²⁶Ra, ²¹⁰Pb, ²¹⁰Po, ²²²Rn, ¹³⁷Cs, and ²³²Th in local bottled water and tube-well water were found to be less than the permissible level given by WHO (2011). The results reveal that local bottled drinking water manufacturing companies may not be following standard guidelines and quality control steps properly. Thus, improvement in their standard operating process is urgently needed to ensure strict compliance with guidelines set by Bangladesh standards and testing institutions.

Keywords: bottled and unbottled water; physico-chemical analysis; microbial analysis; elemental analysis; water quality; dietary reference intake

1. Introduction

Water as a universal solvent is essential for the survival and basic needs of life. It should be free from pollutants, toxic substances, pathogens, and hazardous radionuclides. Without water, we can hardly live for a few days whereas without food, we can live for seven days. To get optimum fitness and alertness, every adult body requires at least 2 L of water per day [1]. All over the world, at least one billion people do not get adequate safe water, more than 800 million people live in rural areas and only 38% of them have access to safe drinking water. It has been reported that about 75% of the population in developing countries has access to fresh water supply [2]. Although about three-fourths of the Earth's surface is covered with water, globally, people face a shortage of freshwater access. Rivers, seas, lakes, tanks, rainwater, and springs are the main sources of potable water. However, freshwater crises are increasing day by day globally. Owing to the lack of access to safe drinking water, most people in developing countries face various health problems [3]. Statistics show that annually in third world countries, about two million people die because of inadequate sanitation, unsafe drinking water, and poor hygiene. Although developed nations have standard laws, programs, policies, and intentions of investments, they have failed to protect public health and sanitation from drinking water-related diseases [4]. In recent decades, even industrial nations like Sweden, Japan, Canada, the U.K, the U.S., and Australia have faced many waterborne diseases [2].

Tube well-, filtered-, and bottled water are the prime sources of drinking water for consumers [5]. Although developed nations have access to safe, good quality, and mineral-rich potable water, consumers prefer to drink bottled water. A range of water purification systems such as distillation, reverse osmosis, microfiltration, ozonation, carbon filtration, ultraviolet (UV) light, etc. is available. However, in Bangladesh, the reverse osmosis (RO) system is commonly employed by different companies to purify water to be used for drinking purposes. Note that the RO treatment completely de-mineralizes the water, which is corrosive and not suitable for drinking purposes. By drinking RO-treated water regularly over a prolonged period, the consumer may face serious health problems; such an example has been experienced in Jordan [2]. To meet the U.S. Food and Drug Administration (FDA, 2008) purified water standards, a bottled water plant is required to follow several processes. Then, under sanitary conditions, the treated water is transferred to a sealed bottle and afterward supplied to the market. Statistics show that the global consumption of bottled water was about 288 and 391 billion liters in 2012 and 2017, respectively [6]. In fact, the demand for bottled water has increased at an average rate of 7% per year over the last 30 years worldwide. This is because it has some advantageous features, such as being easy to carry or handle, available in public facilities, possessing good water quality, being free from pathogens, more palatable, refillable, transparent, and easy to dispense [6,7]. Bottled water follows model code and manufacturing guidelines, hence maintaining standard quality [8].

A large portion of the urban population in developing countries like Bangladesh prefer to drink bottled water due to the unsafe tap water supplied by the competent authorities. City dwellers in Bangladesh receive water from WASA (Water Supply and Sewerage Authority; a service-oriented autonomous commercial organization) for use in domestic or drinking purposes [9]. This organization supplies surface or groundwater from the Padma River after treatment through Water Treatment Plant (WTP) [10–12]. WASA purifies water through some processes like water intake (from the primary reservoir), aeration (increase the dissolved oxygen level), biological pretreatment (reduce the ammonia concentration), rapid sand filtration (for pH adjustment), chlorination (disinfect the water or make free from microbes), and then storage (final reservoir) ([13–15]. More than 90% of rural people use tube-well water for drinking or domestic purposes. Depending on the geographical structure, level of groundwater, or safe mineral water layer availability, tube-well depths vary from 180 to 1000 ft in various areas of Bangladesh. Tube well water

in coastal areas is usually rich in minerals and free from microbial loads due to the presence of a sandy layer or carbonate layer, or the availability of surface water in the nearby sea or river [16].

Due to its mineral-rich profile, Zamzam water has been recognized as unique and differs from natural water [17,18]. The worldwide Muslim community strongly believes that it is holy and miracle water. Zamzam water can satisfy both thirst and hunger [19]. Previous studies [17,20] have reported that this water has medicinal value for curing illness. Recently scientists reported via animal studies that Zamzam water cures cancer and tumors [17]. Due to the presence of high concentrations of various minerals [21], it is considered standard mineral water [17,18]. For instance, the presence of high levels of calcium, magnesium, sodium, etc. provides medicinal benefits, such as the reduction in risk of premature birth, prevention of heart stroke and eye diseases, and lowering of blood pressure [19]. Due to its fluoride content, it works for the prevention of dental caries and also contributes to the good health of bones and teeth [17]. To obtain variation of the mineral profile of this water with Bangladeshi water, Zamzam water is included in this study.

The objectives of this study are to assess the quality of local branded bottled water in Bangladesh, and compare it with imported brands, tube well water, and Zamzam water. This study also aims to obtain radiological and microbial status in some selected bottled water brands.

2. Materials and Methods

2.1. Sample Collection and Preservation

For this study, local tap water supplied by Dhaka Water Supply & Sewerage Authority (DWASA), Dhaka, Bangladesh, tube-well water (from a coastal area of BD); bottled water, including two imported brands and 23 commercially available indigenous branded water samples were collected during three seasons in 2018. According to the indication on their body labels certified by Bangladesh Standard and Testing Institution (BSTI), all of the collected bottled water had a shelf-life of one year. To keep the brand names anonymous, the water samples were given a numerical code from 1 to 9. All bottles were made of Polyethylene Terephthalate (PET) with plastic screw caps. After taking the sample, the bottles were labeled accurately, mentioning the name, location, collection time, date, etc., and then stored in a refrigerator at 4 °C as well as under dark conditions until the analysis was completed.

2.2. Sample Preparation and Analysis

The collected samples were analyzed in the laboratory by properly maintaining the Laboratory Quality Management System (LQMS). The samples were shaken well before testing the parameters and analyzed as per the standard methods [22] listed in Table 1. To find out the representative natural pH, EC, salinity, chloride, turbidity, color, and TDS, respective data were recorded immediately after collection of each sample within 5–10 min at the sampling spot by portable multi-meter. Chlorides, nitrates, and fluoride were tested by Ion Chromatograph (IC). The sulfates and phosphates were determined by using a UV spectrophotometer. Bi-carbonate of the samples was determined by 0.2 N HCl where phenolphthalein was used as the indicator. Chlorine was determined by acetic acid, potassium iodide, and 0.1 N NaOH where phenolphthalein was used as an indicator. A complexometric method was followed to determine the Total Hardness (TH) of water. Use was made of Eriochrom Black T and Na₂-EDTA indicator as a complexing agent. Equation (1) was used to calculate the TDS of each sample:

$$\text{TDS (mg/L)} = [(a - b) \times 1000] \div \text{Sample volume} \quad (1)$$

where, b = weight of evaporating dish and a = weight of evaporating dish and dried residue.

Table 1. Analytical methods for measurement of water quality parameters.

Parameter Name	Instruments or Devices	Method Used	Method
Color & Odor	Color-Meter	Gravimetric	APHA 2017 [22]
Turbidity, TDS, Salinity & EC	Portable Multi-meter (SENSION™156)	-	APHA 2017
pH	pH meter (HannaHI-255)	-	APHA 2017
E. Coli, Total Coliform & Fecal Coliform	Leminary Flow, Derham tube	Count	APHA 2017
Na & K	Flame Photometer	Photo Flame	APHA 2017
Nitrate, Nitrate, Chloride, Fluoride & Sulfate	IC, HIC-10A(super), Shimadzu, Japan	Ion-exchange	APHA 2017
Bi-carbonate, Chlorine	Burette	Titration	APHA 2017
Al, Ca, Mg, Fe, As, Hg, Cu, Cr, Ni, Co, Zn, Ag, Ba, Mo, Pb, Cd, and Mn	Atomic Absorption Spectrophotometer	Atomic Absorption	APHA 2017
⁴⁰ K, ²³⁸ U, ²²⁶ Ra, ²¹⁰ Pb, ¹³⁷ Cs, ²³² Th, ²¹⁰ Po and ²²² Rn	NexION 2000 ICP-MS, PerkinElmer	ICP mass spectrometer	APHA 2017

2.3. Quality Control and Instrumental Analysis

Water quality analysis was performed carefully following the ISO/IEC 17025: 2017 guidelines. Calibrated glassware such as burette, pipette, beaker, and volumetric flask was used by properly rinsing with deionized water several times after being cleaned by diluted 2% (*v/v*) HNO₃ overnight. For sample weighing, a calibrated digital electrical balance (HR-200, Max 210 g, d = 0.1, A&D Company Limited, Tokyo, Japan; Model N92, D0001) was used. During the sample preparation, analytical grade traceable certified reference materials (CRM) reagents (Scharlau, Barcelona, Spain and Sigma Aldrich, Hamburg, Germany) such as HClO₄, HNO₃, deionized water (Barnstead International, Manchester Center, VT, USA, Model-D7071, Resistance 18.2 MΩ cm, Conductivity 0.2 μS/cm at room temperature) were used. Certified reference materials' (Sigma-Aldrich, Hamburg, Germany) stock standards (concentration 100 ± 4 mg/L) were used for instrumental calibration. By spike recovery, duplicate check, independent standard check, and blank check validity were ensured during the work. These analytical results' accuracy and precision were checked with triplicate analysis through the NIST (National Institute of Standards and Technology, Gaithersburg, MD, USA) traceable CRM, and also checked with a quality control chart where upper and lower level confidence limit was 99%.

During turbidity measurement, stock metals standard (Scharlau, Barcelona, Spain) solutions were used to prepare calibration standards. The stock standards were diluted to a certain concentration of calibration standards, which was then used to detect different metals and anions by AAS, ICP-MS, and IC techniques. The detection limits of these metals, such as Na, K, Ca, Mg, Fe, As, Hg, Cu, Cr, Pb, Cd, Mn, Al, Zn, Ni, Co, Ag, Ba, and Mo were 1.0, 1.0, 0.2, 0.1, 0.2, 0.05, 0.1, 0.005, 0.01, 0.001, 0.05, 0.1, 0.3, 0.07, 0.00005, 0.05, 0.02, 1.0, and 0.070 mg/L, respectively. Before metal analysis, a water sample (100 mL) was digested after adding 2 mL of concentrated HNO₃ (67–70%) on a hot plate and evaporated to reduce the volume to 40 mL. Then distilled water was added up to the mark and filtered through a Whatman filter paper. Sodium and potassium concentrations were measured by using a flame photometer. During the radionuclides analysis by ICP-MS, a mixed standard (K, U, Ra, Pb, Cs, Th, Po and Rn in 1% HNO₃), Ultrapure HNO₃ 69.5% (*w/w*) (Fluka) solutions, Argon gas 99.999% purity, and dilutions were prepared with ultra-pure water. The detection limits of these nuclear species such as the K, U, Ra, Pb, Cs, Th, Po, and Rn were 0.01 μg/L. The unit is converted by using the relation 15 μg/L = 180 mBq/L, and the relevant equation with detailed information is available in Yasmin et al. (2018) [23].

2.4. Microbiological Analysis

For determining total bacterial load, the collected water samples were diluted up to 10^{-4} dilutions and cultured following the pour plate technique. Therefore, 1 mL of samples was transferred to a sterile screw-capped vial containing 9 mL distilled water to make a dilution of 1:10 or 10^{-1} . Then using sterile pipettes, 1 mL of the sample from 10^{-1} was transferred to a vial containing 9 mL distilled water to get a dilution of 1:100 or 10^{-2} . In this way, the collected water samples were serially diluted up to 10^{-4} dilutions. Afterward, 1 mL of each dilution was plated by a sterile pipette and covered with a petri dish. Respective sterile melted media was poured into the plates and subsequently, the plates were rotated to spread the media evenly. After solidifying the media, the plates were inverted and incubated overnight at 37 °C. The number of colonies was counted by a colony counter. Total plate count was calculated by multiplying the colonies on the plate with the dilution factor. The mean value of the total plate count of each dilution was calculated as total counts. Besides this, collected water samples were examined to determine the total coliform count according to the MPN method [22] and further screened to check the presence of *Escherichia coli*. Therefore, water samples were inoculated in LB broth (yeast extract 0.5%, tryptone 1%, NaCl 1%) at 37 °C for the enrichment and were observed after overnight incubation. Eosine Methylene Blue (EMB) agar has been used as a selective media for detecting *E. coli*.

3. Results and Discussion

3.1. Discussion on Macro and Micro Minerals

At the International Balneological Congress in Bad Nauheim, Germany, the mineral water idea was first suggested in 1911, and it was recommended that water having more than 1000 mg/L of minerals was to be pursued as mineral water. Mineral water has several amounts of dissolved minerals ions such as cations (Na^+ , K^+ , Mg^{2+} , Ca^{2+}), anions (Cl^- , HCO_3^-), and specific medicinal value compounds. These dissolved mineral ion contents provide several health benefits. Lower values of TDS express the deficiency of essential minerals in water compliance with the standard limits of drinking water guidelines set by various international agencies. Calcium, magnesium, and sodium are potential nutrient minerals for human health. Calcium is a potential mineral for bone development, magnesium protects from cardiovascular disease, and potassium is beneficial to muscles and the nervous system. We talk of hard water with high concentrations of calcium (Ca), magnesium (Mg), bicarbonate (HCO_3^-), and sulfate (SO_4^-) appearing at mg/L concentrations, which contains high levels of minerals [1]. Hard water is also found to be protective against osteoporosis, decreased cognitive function in the elderly, decreased birth weight, cancer, and diabetes mellitus. The soft water (mineral content almost zero) can substantially contribute to the low concentration of lithium (Li), molybdenum (Mo), selenium (Se), and boron (B) at $\mu\text{g/L}$ via daily intake of water (which is more acidic). Such kinds of dietary nutrient minerals—Ca, Mg, Na, K, Fe, Zn, Cu, Cr, I, Co, Mo, and Se—are vital for the physiological and physicochemical functioning of the human body [23,24]. These elements contribute to metabolic catalysis (Zn, Cu, Se, Mg, Mn, Mo), water and electrolyte balance (Na, K, Cl), affect bone and membrane structure (Ca, P, Mg, F), hormone functions (I, Cr), and oxygen binding (Fe) [24]. Low intake of Ca and Mg contribute to rickets in children and osteoporosis in women worldwide [25,26].

In the world malnutrition index, it has been mentioned that the highest malnutrition problems are faced by children and women in Bangladesh. Zinc, iodine, and iron deficiency problems are highly experienced by Bangladeshi children. Pregnant and lactating women suffer from anemia [16,26]. Calcium and magnesium can be fulfilled with a balanced food diet. However, while consumer food has failed to supply these nutrients, drinking water can serve as a suitable alternative for these mineral sources. Minerals are easily dissolved in water so these minerals-rich drinking water sources can achieve the target of ideal dietary intakes [16,27]. For this purpose, bottled water can be beneficial if

they contain these minerals in significant levels. The WHO has recommended that a minimum level of some mineral nutrients should be present in drinking water; the nutrients' names and values [1] are listed in Table S1. On the other hand, Table S2 lists some common and rare chemical ingredients usually found in different types of mineral water; the information is taken from ref. [28].

3.2. Discussion on IBWA, FDA, EU, and EPA Regulations for Bottled Water

The International Bottled Water Association (IBWA) is an industrial advocacy organization that was established in 1958 in Alexandria, Virginia. They set an industry standard document called IBWA Model Bottled Water Regulation that was first issued in 1982, but the latest revision was published in March 2005 (Model Code, 2005) to promote their product [6]. The Model Code has six rules, which extensively highlight water quality protection features [29]. The Model Code emphasizes the definitions of the different kinds of bottled water to create water quality protection policies, followed by good manufacturing practices and operations, sources (free from coliform bacteria), hazard control policies, water transportation systems, and ways to protect water sources from contamination [6]. The Code also recommends that for microbiological contaminants, bottles should be tested weekly, and for other contaminants annually; these criteria are also found in EPA Regulation [30–32].

The US has a tradition of good health security, sanitation, and sewage disposal system [32]. The EPA has gradually monitored the supplied (to the public) water quality and sanitation to control both synthetic compounds and microbial problems. Routine checks are a mandate of the EPA, which is why after a certain period, they examine the microbial (pathogens and viruses) status and disinfection process. The FDA accepts the [33] code as the final part of the Model Code for Federal Regulations. The federal government considers bottled water as a food product for general consumption; thus, the bottled water industry has to abide by US consumer protection law and standards of water quality according to FDA Regulations (21, 40 CFR, 2006). The bottlers must label their bottles with details such as water source, manufacturer name, and water volume (21 CFR, 2006). When the bottled water contains less than 1.0 gm of any nutrient, it is not mandatory to display their provided products' universal nutrition table on the label (21 CFR 101.13, 2014). However, the bottlers must display their product's nutrient table as consumer information (21 CFR 101.9 (j), 2014). If the bottled water contains more than 1.0 gm of any nutrient that has a nutritional benefit or health safety such as "chlorinated" or "little-fat" or "fluoridated", it is mandatory to display the product nutrients table (21 CFR, 2006). IBWA (2017) and FDA (2008) regulations are almost the same. The EU directive (2009/54/EC) suggests that "natural mineral water" can be marked by its nature and original purity that is protected from all risk of pollution. It has also assessed some certain effects by their nutrient contents and trace elements [29,34]. Processing and treatment of any bottled water is discouraged by health experts, and also rejected by European Economic Community Mineral Water Regulations. According to the presence of mineral contents, European brands have been classified as low, moderate, and highly mineralized bottled water [35]. There is some variability between US and European bottled water, but both of them possess higher mineral contents than that of Bangladeshi brands. As per their natural character, some mineral water can be marked as "dietary foods for special health benefits and medicinal value". The mineral ingredients' concentration should not exceed the standard prescribed level. Table 2 shows the mineral classification and their specification, as prescribed by FDA (21CFR165.110, 2014) and European regulations (Directive 80/777/EEC; 2009/54/EC).

Table 2. Mineral water classification and their specification [28,36].

Classification	Specification
Rich in mineral water	Mineral content less than 1500 mg/L
Mineral water	Mineral content less than 1000 mg/L
Low mineral water	Mineral content less than 500 mg/L
Very low mineral water	Mineral content less than 50 mg/L
Chloride water	Chloride content not less than 200 mg/L
Bicarbonate water	Bicarbonate content not less than 600 mg/L
Sulfate water	Sulfate content not less than 200 mg/L
Fluoride water	Fluoride content not less than 2 mg/L
Sodium water	Sodium content not less than 200 mg/L
Magnesium water	Magnesium content not less than 50 mg/L
Calcium water	Calcium content not less than 150 mg/L
Iron water	Bivalent iron content not less than 1 mg/L
Oxygen water	Oxygen content not less than 100 mg L ⁻¹
Acidic water	Free carbon dioxide content not less than 250 mg/L
CO ₂ -rich, carbonized water or sparkling water	Free carbon dioxide content not less than 1000 mg/L
Radon or radioactive water	≥74 Bq/L
Silica water	H ₂ SiO ₃ content less than 70 mg/L
Thermal water	Temperature (°C) not less than 20 (°C)
Coldwater	Temperature (°C) not less than 20 (°C)
Vitamins water	B12, B9, and B3 (Voda, 2017, www.vodanaturalna.pl (accessed on 11 July 2020))
Flavors water	Strawberry and calcium, lemon vitamin and C, apple and zinc, grapefruit and magnesium peach fiber, and coconut and l-carnitine (ABC Consulting, 2013).
Low sodium diet water	Sodium content less than 20 mg/L
Drinking water	Mineral content should be 1% weight of the water and also free from pathogen, nucleosides, and contaminants.
Purified water	Distilled water or deionized water.
Infant food water	Sodium or chloride content less than 20 mg/L
	Fluoride content less than 0.7 mg/L
	Nitrate content less than 10 mg/L
	Nitrite content less than 0.02 mg/L

A comparison of Zamzam water, local bottled water, tap water, imported branded water, and distilled water with permissible limits of drinking water quality parameters set by different agencies have been assembled in Tables 3 and 4.

Table 3. Comparison of the water quality parameters between Zamzam, local bottled water, tap water, imported brand mineral water, and distilled water [17,21,37,38].

Parameters	Zamzam Water	Local Bottled Water	Tube Well Water	Imported Brand Mineral Water	Distilled Water
Appearance	Clear	Clear	Clear	Clear	Clear
Odor	Acceptable	Acceptable	Acceptable	Acceptable	Acceptable
Turbidity (NTU)	-	Nil	Nil	Nil	Nil
TDS (mg/L)	895	55.36	135	412.5	—
pH	7.73	7.19	6.83	6.5	6.85
Conductivity (µS/cm)	1390	112	275	-	0.5
Total hardness(mg/L)	300	18.75	85.47	647.54	0.2
Calcium (mg/L)	96	2.83	9.95	120	0.1
Sodium (mg/L)	133	11.08304	12.43	8.0	-
Potassium (mg/L)	43.3	0.39422	1.48	0.5	-
Magnesium (mg/L)	38	2.75	10.40	15.1	0.1
HCO ₃ (mg/L)	195.4	45.02	52.14	395	0.2
Chlorides (mg/L)	260	3.86	1.27	16	0.1

Fluoride (mg/L)	0.59	0.012	0.0	0.0	0.0	0.1
Sulfates (mg/L)	180	12.18	0.14	23.5	Nil	Nil
Nitrite (mg/L)	105	Nil	0.0	5.8	Nil	Nil
Nitrates (mg/L)	<0.01	0.45	0.0	0.0	10	10
Iron (mg/L)	0.12	<0.2	<0.2	<0.2	Nil	Nil
Copper (mg/L)	2.94	<0.1	<0.1	<0.1	Nil	Nil
Manganese (mg/L)	0.69	<0.05	<0.05	<0.05	Nil	Nil
Lead (mg/L)	2.45	<0.01	<0.01	<0.01	Nil	Nil
Chromium (mg/L)	1.68	<0.005	<0.005	<0.005	Nil	Nil
Mercury (mg/L)	-	<0.001	<0.001	<0.001	Nil	Nil
Arsenic (mg/L)	0.025	<0.005	<0.005	<0.005	Nil	Nil
Cadmium (mg/L)	0.11	<0.001	<0.001	<0.001	Nil	Nil

Table 4. Permissible limits of drinking water quality parameters set by different recognized agencies.

Parameters	Unit	WHO (2011) [39]	FDA (2008) [40]	SASO (1994) [41]	IBWA(2008) [42]	US EPA (2018) [43]	Zamzam Water (Khalid et al., 2014) [17]	BIS (2012) [44]
Color	Hazen	15	15	50	5	15	Colorless	15
Odor	-	Agreeable	3 T.O.N.	Acceptable	3 T.O.N	3 T.O.N.	Orderless	Agreeable
Turbidity	NTU	5	5	25	0.5	5	0.11	5
pH	—	6.5–9.5	—	6.5–8.5	6.5–8.5	6.5–8.5	7.5	6.5–8.5
TDS	mg/L	1000	500	1500	500	500	810	2000
Hardness	mg/L	—	—	500	—	—	303	600
Bicarbonate	mg/L	—	—	—	—	—	172	—
Ca	mg/L	—	—	200	—	—	92	200
Mg	mg/L	—	—	30–150	—	30–60	38.5	100
Na	mg/L	200	—	—	—	—	129.5	—
K	mg/L	—	—	—	—	—	44.5	—
Al	µg/L	0.2	—	—	—	200	22.04	—
Co	µg/L	70	—	—	—	—	0.38	—
Ni	µg/L	70	—	—	—	—	65.0	—
Mo	µg/L	70	—	—	—	—	27.08	—
Cl	mg/L	250	250	600	250	250	164.5	1000
Cl ₂	mg/L	—	4	0.2–1.0	0.1	4	-	4
EC	(µS/cm)	—	—	800–2300	—	-	1280	—
SO ₄	mg/L	250	250	400	250	250	123.6	400
NO ₃	mg/L	50	10	45	10	45	132.3	45
NO ₂	mg/L	3	1	—	1	1	-	—
F	mg/L	1.5	0.8–2.4	0.6–1.0	0.8–2.4	4	0.59	1.5
As	µg/L	10	10	50	10	10	9.68	50
Cd	µg/L	3	5	5	5	5	—	3
Cr	µg/L	50	100	50	50	100	1.33	50
Cu	µg/L	2000	1000	1000	1000	1300	3.63	150
Fe	µg/L	300	300	1000	300	300	85.53	300
Mn	µg/L	500	50	50	50	50	15.61	300
Pb	µg/L	10	5	50	5	0	—	100
Zn	µg/L	3000	5000	5000	5000	5000	16.6	15000
Ba	µg/L	—	—	1000	—	2000	—	—
Hg	µg/L	1.0	1.0	—	2.0	2	—	10
<i>E.Coli</i> (n/100)	Cfu	0	<2.2	0	0.0	<5%	—	0

The concentrations of water quality parameters in the studied local brands of water and Zamzam water, imported water, and tube-well water are listed in Tables 5 and 6. The statistical values of the water quality parameters of different local brands of water are listed in Table 5.

Table 5. The concentration of different quality parameters of local brands of bottled water.

Sample ID	pH	EC (μ S/Cm)	TDS (mg/L)	Na (mg/L)	K (mg/L)	Ca (mg/L)	Mg (mg/L)	SO ₄ (mg/L)	Cl (mg/L)	F (mg/L)	NO ₃ (mg/L)	HCO ₃ (mg/L)
Mum (D1)	6.45	245.48	117.33	7.81	1.67	29.07	3.29	49.88	23.95	0.0	2.93	89.32
Fresh (D2)	6.44	223	106.27	15.23	0.91	2.43	2.81	1.35	42.67	0.0	00.0	17.29
Pran (D3)	6.43	150	71.4	11.56	0.19	0.365	6.78	26.19	2.18	0.0	0.11	83.55
Aquafina (D4)	6.91	8.08	3.3	1.85	0.0	0.525	0.085	0.0	0.0	0.0	0.0	23.05
Kinley (D5)	6.81	32.95	15.4	1.63	0.0	0.185	3.965	10.86	1.31	0.0	0.0	2.88
Lilia (D6)	6.79	113.1	53.5	13.45	0.104	0.36	0.13	0.185	0.0	0.0	0.0	13.33
Jibon (D7)	6.95	145.32	69.4	9.35	0.909	5.03	4.34	0.183	6.4433	0.065	0.0	83.55
Spa (D8)	7.36	433.8	209	17.93	3.24	31	16.55	119.39	43.745	0.0	0.054	11.52
Ifad (D9)	7.58	30.0	5.9	13.8	0.0	0.15	0.04	0.16	0.0	0.205	0.325	21.61
Crystal (D10)	7.52	122.55	20.85	11.96	0.103	0.345	2.225	9.11	1.055	0.0	0.1625	18.37
Eco (D11)	7.32	160.5	76.7	12.43	0.008	0.61	2.835	37.99	2.22	0.0	0.084	28.35
Oceania (D12)	7.14	247	118	15.61	0.082	0.75	7.63	83.37	2.707	0.0	0.102	11.80
Mamia (D13)	7.79	25.8	11.8	5.21	0.0	0.18	0.04	0.13	0.965	0.0	0.68	26.89
Shanti (D14)	7.23	107	51.2	18.23	0.288	5.05	0.28	0.0	8.48	0.0	0.468	31.69
Pani (D15)	7.45	51.8	24.3	13.59	0.0	0.47	0.09	0.0	0.0	0.0	0.0	66.27
Tape water (D16)	6.83	275	135	12.43	1.68	9.95	10.4	0.14	1.27	0.0	0.0	52.14
ACEME (D17)	7.83	16.5	7.15	3.64	0.0	0.265	0.049	0.06	0.3165	0.0	0.355	11.52
Premium (D18)	7.09	173	83.05	19.1	1.009	5.9	4.94	0.29	7.3375	0.0	0.0	86.83
Perfect (D19)	7.11	7.12	2.9	1.67	0.0	0.14	0.045	0.0	0.0	0.0	0.0	14.18
Fina (D20)	7.46	250	119	36.15	0.456	0.88	0.24	1.12	3.144	0.0	0.0	42.14
Mukta (D21)	7.5	15.02	6.6	2.71	0.41	0.96	0.22	0.0	0.201	0.0	0.0	16.64
Shyamoli (D22)	7.45	269.5	128.5	21.95	0.82	1.495	0.12	0.36	6.86	0.0	7.955	72.03
Revera (D23)	7.05	103	50.14	11.24	0.87	0.58	0.14	0.33	0.56	0.0	0.45	33.13
Delta (D24)	6.98	12.05	5.17	2.36	0.11	1.54	0.44	0.54	2.11	0.0	0.14	37.45
Rangdanu (D25)	6.96	49.02	18.17	5.31	0.44	0.82	2.12	0.41	1.35	0.0	0.0	276.6
Deshbhandu (26)	7.10	85.01	42.18	4.18	0.28	0.45	2.33	1.57	0.28	0.0	0.0	17.29
Mean \pm SD	7.21 \pm 0.34	113 \pm 107	51.9 \pm 52.5	11.1 \pm 8.4	0.394 \pm 0.704	2.52 \pm 6.34	2.42 \pm 3.85	12.7 \pm 30.0	3.97 \pm 9.05	0.012 \pm 0.044	0.473 \pm 1.64	44.8 \pm 56.7

Table 6. Concentration of different quality parameters of imported water brands, Zamzam water, and tube-well water.

Parameter	pH	Na (mg/L)	K (mg/L)	Ca (mg/L)	Mg (mg/L)	Fe (mg/L)	TDS (mg/L)	Cl (mg/L)	HCO ₃ (mg/L)	F (mg/L)	SiO ₂ (mg/L)	SO ₄ (mg/L)	NO ₃ (mg/L)
Ferrion	5.5	9.5	<1	160	4.2	-	480	22	430	-	-	33	7.8
Evian	7.2	6.5	1	80	26	0.29	345	10	360	-	15	14	3.8
Imported BMV	6.35	8	1	120	15.1	0.15	412.5	16	395	-	7.5	23.5	5.8
ZZ-1 [17]	7.73	133	43.3	96	38	0.12	835	163.3	195.4	0.72	-	124	124.8
ZZ-2 [18]	7.75	121.9	43.3	114	38.88	0.06	1011	147.5	285	0.72	-	187	124.8
ZZ-3 [21]	7.5	129.5	44.5	92	38.5	0.085	895	260	172	0.59	-	123.6	132.3
ZZ Mean Value	7.66	128.1	43.7	100.7	38.46	0.088	913.7	190.3	217.5	0.677	-	144.9	127.3
TW-1	6.9	26.19	25.78	53.04	32.98	0.46	260	230	296.73	0.5	10.12	132	2.52
TW-2	6.85	22.85	24.89	52.12	28.6	0.5	261	227	290	0.45	10.15	129	2.6
TW-3	7.02	28.3	25.01	55.2	34.56	0.55	258	235	295	0.49	10.8	135	2.78
TW Mean Value	6.92	25.78	25.23	53.45	32.05	0.50	259.7	230.7	293.9	0.48	132	2.63	10.36

TW→Tube well Water, ZZ→Zamzam Water, BMV→Brand Mean Value.

3.3. pH and Bi-Carbonate (HCO₃⁻)

According to WHO (2006) [45] guidelines, the maximum desirable limit of pH should be 6.5 to 9.5. Some organizations like EC (1993) [4], SASO (1986) [41], BIS (2012) [44], US EPA (2018) [43], and IBWA (2008) [42] have established pH limits from 6.5 to 8.5, which are listed in Table 4. The pH value of local bottled water brands was found to be 7.2 with the highest value being 7.83 and lowest at 6.43 (See in Table 5). The pH value of imported

brands, Zamzam water, and tube-well water was found as 6.35, 7.66, and 6.92, respectively (See in Table 6). Thus, the observed pH value of all the local brands, Zamzam water, and tube-well water did not exceed the prescribed limits, where the pH of imported branded water exceeded the prescribed limits.

Bicarbonate ion is the most important buffer and standby as a safeguard against acidity. The bicarbonate of local branded water was found to be 44.81 mg/L. The highest and lowest bicarbonate values were found at 276.59 mg/L and 2.88 mg/L (Table 5), and this indicates the lowest bicarbonate concentration in bottled water and did not exceed the recommended value. The concentrations of bicarbonate in imported brands, Zamzam water, and tube-well water were found as 395 mg/L, 217.47 mg/L, and 293.91 mg/L, respectively. According to the international standards listed in Table 4, the concentrations of bicarbonate in local brands, Zamzam water, imported brands, and tube-well water are within the prescribed limit.

3.4. Total Dissolved Solids (TDS)

All inorganic and organic matters or salts present in water are termed Total Dissolved Solids (TDS). The hardness of water is measured by the summation of the total amount of the content of TDS in water. However, while some of the ions are essential, others are toxic to human health. Low TDS and total hardness (TH) water contain 1–100 mg/L dissolved solids in the water [16,46]. This type of water quality was obtained from water treatment technology through distillation, reverse osmosis, and deionization. The concentrations of local brands' TDS range was 2.9–209 mg/L with a mean value of 51.9, as listed in Table 5. It is clear that only one company (D8) possessed a pleasant TDS level (209 mg/L), while the other companies did not achieve the standard level. It is to be noted that the level of TDS (2.9 mg/L) of one company (D19) was unusually low. The TDS value of imported brands, Zamzam water, and tube-well water was found to be 412.5 mg/L, 913.6 mg/L, and 259.67 mg/L, respectively. The TDS values as per IBWA (2008), WHO (2008), SASO (1994), BIS (2012), and US EPA (2018) guidelines are listed in Table 4. According to Table 4, the permissible level of TDS for mineral bottled water and drinking water are from 500 to 2000 mg/L, based on various agencies. Thus, the observed TDS value of all the local brands, Zamzam water, tube-well water, and imported branded water did not exceed the prescribed standard limits. The local brands contained a significantly lower amount of TDS compared to the other water samples in this study (see the Table 3). When the TDS level in drinking water is 0.00 mg/L, it indicates that the water has no minerals [47]. However, drinking water without minerals may neither fulfill requirements of the body nor does it have any taste. Therefore, necessary minerals in drinking water are required for a suitable taste and for fulfilling the lack of mineral demands by human health. People usually drink bottled mineral water or spring water that has a TDS value of less than 3000 mg/L.

3.5. Nitrate (NO_3^-) and Nitrite (NO_2^-)

Nitrate and nitrite are found in the nitrogen cycle and these ions are naturally originated [48]. Hemoglobin in human blood and other warm-blooded animals is highly reactive to nitrites and is prone to produce methemoglobin. It is responsible for demolishing the red blood cells' oxygen-transporting ability, which may be serious in babies under three months of age. It may result in a condition known as methemoglobinemia or "blue baby disease". Babies should not be fed water when their nitrite level exceeds 1.0 mg/L. Observations have shown that nitrite values have crossed the recommended value. Drinking water having more than 50 mg/L is the main source of the total nitrate intake in the human body. Sewage disposal of industrial discharges, domestic effluents, leaching from refuse dumps, excessive application of chemical fertilizers in agriculture, water from decayed vegetables, and atmospheric precipitation have become great sources of pollutants for drinking water [49,50]. In public drinking water, nitrate and nitrite level should not

exceed 50 mg/L and 3 mg/L, respectively. Standard guidelines listed in Table 4 have prescribed that the concentrations of NO_3^- should be 10 mg/L and 45 mg/L. NO_3^- concentration in investigated water was found to be 0.47 mg/L, which implies that the water quality did not comply with good standards. The highest and lowest NO_3^- value in local bottled water was found to be 7.9 mg/L and 0.0 mg/L, as listed in Table 6, which indicates that in bottled water, it did not exceed the recommended value. Nitrite in Zamzam water and tube-well water was found to be 123.7 mg/L and 2.63 mg/L, respectively. Standard guidelines have also prescribed the concentration of NO_2^- to be 1.0 mg/L. Obtained NO_2^- concentration in investigated water was 0.0 mg/L. The data are not shown in Tables 5 and 6 due to the low NO_2^- concentration, which did not exceed the recommended value prescribed by standard agencies.

3.6. Chloride (Cl^-) and Free Chlorine (Cl_2)

Sodium salt is liable for heart and kidney diseases. If the concentration of NaCl, CaCl_2 , and MgCl_2 exceeds the level of 250 mg/L, 500 mg/L, and 1000 mg/L respectively, it has a salty taste. FDA (2008), WHO (2011), IBWA (2008), US EPA (2018), and BIS (2012) prescribed that the chloride (Cl^-) level in drinking water should be less than 250 mg/L (see in Table 4). Chloride concentration in commercially bottled water was found to be 3.96 mg/L, which indicates a very low concentration. The highest and lowest chloride value was found to be 43.75 mg/L and 0.0 mg/L, respectively (see in Table 5). The investigated value indicates a very lower chloride concentration present in bottled water, also far below the recommended value. The sources of chlorides are agricultural run-off, rocks, wastewater from industries, oil well wastages, and effluents from wastewater treatment plants [50]. When the chlorides concentration crosses the standard level, it can contaminate freshwater, streams, and lakes, where the aquatic community can hardly survive. The obtained results of chloride concentrations in imported brands, Zamzam water, and tube-well water were 16.0 mg/L, 190.27 mg/L, and 230.67 mg/L, respectively (see in Table 6). Thus, the observed chloride values of all the local brands, Zamzam water, tube-well water, and imported branded water have not exceeded the prescribed limits. On the other hand, chlorine concentration in the investigated sample was found to be 0.0 mg/L. The detailed data of chlorine concentrations are not shown in Tables 5 and 6 due to their very low values, which certainly do not exceed the recommended values given by the standard agencies (see Table 4).

3.7. Fluoride (F^-) and Sulphates (SO_4^{2-})

A very high concentration of fluorides (F^-) can be destructive and responsible for discoloring teeth and causing bone deformations while the standard level of fluoride (F^-) in drinking water is beneficial to teeth. According to the WHO (2011) and BIS (2012), the maximum permissible limit of fluoride in drinking water is 1.5 mg/L while a further highest limit of 4 mg/L is set by US EPA (2018) [43]. FDA (2008), SASO (1994), and IBWA (2008) established fluoride limits as 0.8–2.4, 0.6–1.0 and 0.8–2.4 mg/L, respectively (see in Table 4). The mean value of fluoride was found to be 0.012 mg/L, while the highest and lowest fluoride values were found to be 0.205 mg/L and 0.0 mg/L, respectively. All values show the presence of the lowest fluoride concentration in the analyzed local branded water samples. The concentrations of fluoride in imported brands, Zamzam water, and tube-well water were found to be 23.5 mg/L, 0.67 mg/L, and 0.48 mg/L respectively, as listed in Table 6. By comparing with international standard listed in Table 4, it can be mentioned that the concentrations of fluoride in local brands, Zamzam water, and tube-well water are within the prescribed limits; however, in imported water brands, these values crossed the prescribed limits. The ranges of DRI fulfillment of fluoride for adult males from imported brands, local brands, tube-well water, and Zamzam water were 70% to 82.6%, 0.0% to 5.13%, 12.5% to 22.5%, and 25.0% to 29.5%, respectively (see the Table 7). Fluoride concentration above 1.5 mg/L in drinking water causes dental fluorosis and a much higher concentration causes skeletal fluorosis, while low a concentration (approximately 0.5 mg/L)

has been proven to be protective against dental problems. Globally, in about 23 countries, health problems occur due to the consumption of fluoride-contaminated water (fluoride contaminates the water when it varies from 1.0 to 400 mg/L). The statistics show that in India, about 20 million people have faced serious fluorosis diseases and an additional 40 million people are in risk of endemic fluorosis [51].

Sulfates are many important ingredients that show the water bodies' trophic state index (TSI) and deterioration of water quality. Sulfates of calcium and magnesium form hard scales [52]. In combined form with other ions, it exerts a bitter taste and large concentrations of sulfates have a purgative effect on human health. According to the standard guidelines (Table 4), the desirable range of sulfates in drinking water should be 250 mg/L to 400 mg/L. The mean value of sulfates was found to be 12.71 mg/L, while the highest and lowest sulfates were found to be 292.25 mg/L and 0.0 mg/L (as listed in Table 6), which indicates the presence of sulfates in the lowest concentration in bottled water. The concentrations of sulfates in imported brands, Zamzam water, and tube-well water were 5.8 mg/L, 144.87 mg/L, and 132 mg/L, respectively. According to the international standards, the concentrations of sulfates in all the local brands, Zamzam water, imported brands, and tube-well water are within the prescribed limits.

3.8. Calcium (Ca) and Magnesium (Mg)

Table 5 shows the comparison of average Ca, Mg, and Na levels (in mg/L) in Bangladeshi, US, and European bottled water (% of DRI fulfillment for adult males is shown in parentheses). Table S3 shows the Dietary Reference Intakes (DRIs) [14,26,53,54] of select mineral elements.

Table 7. Comparison of average Ca, Mg, and Na levels (in mg/L) in Bangladesh, US, and European bottled water (% DRI fulfillment for adult males are shown in parentheses).

Bottled Water	Ca: Mean \pm SD (% DRI Range)	Mg: Mean \pm SD (% DRI Range)	Na: Mean \pm SD (% DRI Range)
Local brands (23 samples)	2.52 \pm 6.43 (0.01–3.1)	2.4 \pm 3.84 (0.001–6.89)	11.08 \pm 8.35(0.11– 3.01)
Imported brands (2 samples)	120 \pm 56.57 (6.15–16)	15.1 \pm 15.41(1.0–10.83)	8.0 \pm 2.12(0.43–0.79)
Tube well Water (3 samples)	53.45 \pm 1.58 (4.01–5.52)	32.05 \pm 3.09(6.81–14.4)	25.78 \pm 2.75(1.52–2.36)
Zamzam water (3 samples)	100.67 \pm 11.72 (7.08–11.4)	38.46 \pm 0.44(9.05–16.2)	128.13 \pm 5.7(8.13–11.08)
North American bottled waters [15] (1) Springwater	18 \pm 22 (0–7.6)	8 \pm 18 (0–39.58)	4 \pm 4(0–1.25)
(2) Mineral water	100 \pm 125 (0.23–31)	24 \pm 42 (0.24–54.17)	371 \pm 335(2.4– 91.25)
European bottled waters [15]	60 \pm 40(0.31–14.5)	16 \pm 19 (0.24–45.83)	13 \pm 13 (0.07–4.67)
(1)Low mineralization waters	262 \pm 139(6–57.5)	64 \pm 37 (2.14–53.33)	157 \pm 197 (0.13–55)
(2) Moderate mineralization waters	60 \pm 59 (0.38–17.6)	16 \pm 20(0.95–25)	1151 \pm 153 (60–100)
(3) High mineralization waters			

Calcium and magnesium are prime elements in rocks and soil, and their main sources are CaCO_3 , MgCO_3 , $\text{CaMg}(\text{CO}_2)_2$, $\text{CaAl}_2\text{Si}_2\text{O}_8$, CaSO_4 , $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$, $\text{Ca}_5(\text{PO}_4)_3\text{F}$, CaF_2 , and $\text{Ca}_5(\text{PO}_4)_3\text{OH}$. Calcium and magnesium are leached from soil and rocks into the water, and may exist in water in complex forms such as $[\text{CaCO}_3]$, $[\text{MgCO}_3]$, $[\text{CaSO}_4]$, $[\text{MgSO}_4]$, $[\text{MgOH}]^+$, $[\text{CaOH}]^-$, $[\text{MgHCO}_3]^+$ and $[\text{CaHCO}_3]^+$ [55]. At a temperature of 18 °C and pressure of 1000 hPa, the maximum amount of Ca content present in water is to be >1 g/L [56]. The presence of Ca and Mg in drinking water is of significance for health—bones and teeth. Ca and Mg are linked to a reduction in sudden death, and cardiovascular (ischemic) disease mortality, and most of the drinking water varies from 2:1 to 4:1. A high concentration of calcium prevents the up-taking of magnesium and thus the optimum balance of these two minerals prevails in water. Epidemiological research carried out in Europe, the US, and Russia recommend that a level of 20–30 mg/L Ca and 10 mg/L Mg in drinking water may be beneficial [56,57]. Different countries have prescribed certain levels of standard values such as 75 mg/L Ca and 30–35 mg/L Mg in drinking water to meet nutrient

deficiency [58]. According to WHO (2017) [59] guidelines, Ca content and Mg content in mineral and drinking water should range from 50–200 mg/L and single-digit to 50 mg/L, respectively. Every adult should take 1 g of Ca per day.

The calcium content in imported brands of water ranged from 80 to 160 mg/L with a mean value of 120 mg/L, while in local brands, it is ranged from 0.14 to 31 mg/L with a mean value of 2.5 mg/L. In the case of magnesium content, the range and mean value for imported brands were 4.2 to 26.0 mg/L and 15.1 mg/L, respectively; for local brands, it ranged from 0.04 to 16.55 mg/L and a mean of 2.42 mg/L. The local brands contain a significantly lower amount of calcium and magnesium concentration compared with the imported brands, Zamzam water, and tube-well water (see the Table 3). The concentration of Ca and Mg in Zamzam water were found as 96 mg/L and 36 mg/L, and in tube-well water 53.44 mg/L and 32.06 mg/L, respectively. When we considered the % of DRI fulfillment of calcium and magnesium for adults, the contribution of imported brands, Zamzam water, and tube-well water was found to be higher than the local brands (see the Table 7). The average calcium content in the imported brands can fulfill the calcium demand of DRI from 1.4% to 19.8% for adult males. For adult males, the % of DRI fulfillment by local brands ranged from 0% to 3.3% only. The % of DRI fulfillment of magnesium for adult males by imported and local brands was 1% to 16.6% and 0.002% to 7.3%, respectively (see the Table 7).

3.9. Sodium (Na) and Potassium (K)

Sodium and potassium are major elements in soil and rocks. Sodium and potassium leach into groundwater from soil and rock, and are present in water in complex forms, such as $[\text{NaSO}_4]^-$, $[\text{NaCO}_3]^-$, $[\text{KCO}_3]^-$, $[\text{KHCO}_3]$, and $[\text{NaHCO}_3]$ [60]. KCl, KAlSi_2O_6 , and $\text{KCl}\cdot\text{MgCl}_2\cdot 6\text{H}_2\text{O}$ are major potassium minerals. The range of sodium concentrations in imported brands was found to be 6.5–9.5 mg/L with a mean value of 8.0 mg/L (Table 6), while the respective data in local brands were 1.63–36.15 mg/L and 11.08 mg/L. The range of potassium concentrations and mean value are 0.0 to 3.25 mg/L and 0.39 mg/L in local brands, and <1.0 to 1.0 mg/L and 1.0 mg/L in imported brands. The concentrations of Na and K in Zamzam water were found to be 128.13 mg/L and 25.78 mg/L, and 28.3 mg/L and 43.7 mg/L in tube-well water (Table 6). The American Public Health Association (APHA) has recommended that drinking water should contain a maximum level of 20 mg/L Na for individuals on a severely restricted Na diet (500 mg of Na per day). In this regard, both the local and imported brands of water often have been found to exceed the 20 mg/L limits suggested by APHA [22]. This indicates that an individual with a sodium-restricted diet should avoid some of the local and imported brands of water. The ranges of DRI fulfillment of sodium for adult males by imported brands, local brands, tube-well water, Zamzam water were found to be 0.43% to 0.79%, 0.11% to 3.01%, 1.52% to 2.36%, and 8.13% to 11.08% respectively (see the Table 7). The DRI contributions (from local: 0% to 0.072% and imported: 0.01% to 0.02%) of potassium for an average adult male indicates that none of these bottled water sources contains a significant level of potassium. The ranges of DRI fulfillment of potassium for adult males from tube-well water and Zamzam water were 1.66% to 2.14%, and 0.92% to 0.98%, respectively (see the Table 7). According to WHO (2017) [59] guidelines, a high Na concentration is very dangerous for children, and deficiency of potassium can lead to vertigo. The WHO (2017; 2008) also prescribed that sodium concentration in drinking water is 200 mg/L and there is no limit for potassium content in drinking water.

3.10. The Status of Minerals in Imported Bottled Waters

The US has classified bottled water as spring water and mineral water. Spring water contains a very low amount of minerals (Ca, Mg, and Na) with ranges of 0–76 mg/L, 0–95 mg/L, and 0–15 mg/L, respectively [15]. Local brands of bottled water also contain a low amount of calcium and magnesium than the Bangladesh standards of 200 mg/L and 100 mg/L. Calcium and magnesium are essential macronutrients for human health [15]. By

continuous consumption of local brands of bottled water, the consumer may be deprived of essential mineral nutrients. The intake of minerals through the consumption of bottled water is clinically important (WHO, 2017; 2008). The ranges of DRI fulfillment of (Ca, Mg and Na) for adult males from spring water were 0.0% to 7.6%, 0.0–39.58%, 0.0–1.25%, respectively (see the Table 7), which were slightly higher than that of the local brands. The US mineral water contributes a greater % of DRI fulfillment. For an adult male, they can fulfill % of DRI up to 31%, 54.17%, and 91.25% of Ca, Mg, and Na intake (see the Table 7). This shows that the US and European mineral waters fulfill a significant portion of the dietary mineral requirements through the consumption of different mineralized levels. Low mineralized local bottled water can fulfill DRI values of Ca, Mg, and Na with the ranges: 0.31% to 14.5%, 0.24% to 45.83%, and 0.07% to 4.67%, while the high mineralized water can contribute to DRI fulfillment up to 17.6%, 25% and 100% of Ca, Mg, and Na, respectively (see the Table 7).

3.11. Iron (Fe) and Copper (Cu)

Iron (Fe) and copper (Cu) are important nutrient elements and excess Fe and Cu from drinking water may cause intestinal disorders and liver damage. The Fe and Cu contents in drinking and mineral water vary from 0.01 to 20 mg/L and 1 to 100 µg/L, respectively. Iron can be found in colloid form in tap and surface water. Minerals are present in ionic form in water, and the human body can easily absorb these ions from water than food-stuff. At least about 10 to 50 mg/L iron is required daily and is present normally in the ferrous or bivalent form in groundwater [61,62]. Plants and animals both need iron, especially for oxygen transport in the blood of animals. Infants and young face anemia or iron deficiency because it is essential for metabolism. Iron appears to be a nuisance than a potential health hazard. The presence of iron in water with a concentration of 0.1 mg/L as ferrous iron and 0.2 mg/L as ferric iron as a bitter or astringent taste. Water used in industrial processes usually contain less than 0.2 mg/L iron. According to standard guidelines (Table 4), the highest desirable limit of iron in drinking water should be 0.3 mg/L. The mean value of Fe and Cu in the investigated sample was found to be less than 300 µg/L. Copper is insoluble in water at high pH and soluble at low pH. The permissible limit of copper concentration in drinking and mineral water is 1 mg/L. The detailed data of Fe and Cu in the analyzed samples (also for other minerals and chemicals Pb, Cd, Cr, Mn, Hg, and Zn, as described in the following sub-sections), are not shown in Tables 5 and 6 due to their low concentrations, and the values did not exceed the recommended value of standard agencies.

3.12. Lead (Pb) and Cadmium (Cd)

According to standard guidelines of BIS (2012), WHO (2008;2017), SASO (1994), FDA (2008), IBWA (2008), and US EPA (2018), the highest desirable limit of lead in drinking water should be 0.1 mg/L, 0.01 mg/L, 0.03 mg/L, 0.05 mg/L, 0.005 mg/L, and 0.0 mg/L, respectively. The mean value of Pb in the investigated sample was found to be less than 10 µg/L, and this value indicates that the water is harmless.

Cadmium is a carcinogenic element that is very toxic and has no known biological role. Scientists believe that excessive concentration of cadmium may cause liver and kidney damage, anemia, and death [36]. Brittle bone disorder has occurred due to Cd. According to standard guidelines (Table 4), cadmium with a level of 3–5 µg/L is not harmful to health. The mean value of Cd in the investigated sample was found to be less than 3 ppb, and this value indicates that the water is harmless.

3.13. Manganese (Mn) and Chromium (Cr)

Manganese exists in soil and sediment in the form of MnO₂ and Mn₂O₃. Referring to the [63] guidelines, the standard limit of Mn in drinking water is 0.1 mg/L. Less than 50 µg/L of Mn in drinking water can be drinkable, but a high level of Mn in water can be

poisonous [64]. A study shows that chlorosis disease occurs due to a lack of Mn content in the human body. Chromium is leached into water from soil, rock, and burning of fuels (coal, oil). Chromite (FeCr_2O_4) is the main source of Cr minerals. In water, chromium can be found as ions from Cr^{+6} and Cr^{+3} but the Cr^{+3} ionic form has no toxic effect. The Cr^{+6} ionic form has a toxic effect, and prolonged consumption can damage the liver, kidneys, nerves, and the blood circulatory system [21]. The permissible limit of chromium concentration in drinking water is $50 \mu\text{g/L}$. The concentration of Cr and Mn in the investigated sample was found to be at a pleasant level and below the limiting values set by different standard agencies.

3.14. Mercury (Hg) and Zinc (Zn)

According to standard guidelines (Table 4), mercury with a concentration of 1–10 ppb is not harmful to health. The mean value of Hg in the investigated sample was found to be less than 1–10 ppb, and this value indicates that the water is nontoxic to health. The standard limit of Hg in drinking water is less than $0.5 \mu\text{g/L}$ [2,64].

Zinc is beneficial for metabolism, and essential for healthy growth and a strong immune system [65]. Zinc in drinking water is generally derived from the galvanized coatings of piping [21]. According to standard guidelines as listed in Table 4, Zn with concentration of 3–5 mg/L is not harmful but it can impart a metallic taste or milky appearance to water. The mean value of Zn in the investigated sample was found to be less than 3 mg/L, which indicates that the water is harmless for health.

3.15. Molybdenum (Mo) and Arsenic (As)

WHO (2011), FDA (2008), IBWA (2008), and US EPA (2018) have prescribed a provisional guideline for arsenic in drinking water— $10 \mu\text{g/L}$ (Table 5). According to BIS (2012) and SASO (1994), the maximum permissible level is $50 \mu\text{g/L}$, and the obtained As data show a level below this limit.

The daily molybdenum dietary requirement for an adult body is 0.2 to 0.3 g. However, a high concentration of this metal causes bone deformation or slower physical development and more growth abnormalities. The WHO (2017) suggested that the permissible limit of Mo in drinking water is $70 \mu\text{g/L}$. The concentration of Mo in the investigated sample was found to be at a pleasant level and below the standard limit prescribed by different agencies.

3.16. The Status of Other Parameters

In the studied local brands, imported brands, Zamzam, and tube-well waters, other quality parameters such as color, order, hardness, alkalinity, dissolved oxygen, turbidity (NTU), phosphate, Co, Al, Ag, Ni, and Ba have not crossed permitted (Table 4) and ECR (1997) limits [66]. In addition, the data are not shown in Tables 5 and 6 due to their insignificant concentrations.

3.17. Microbial Status in the Investigated Samples

The status of total Coliform, Fecal Coliform, and *E. Coli* indicates the water quality pollution level; the permissible limit given by different agencies is listed in Table 4. IBWA (2008), WHO (2011), SASO (1994), and BIS (2012) recommended that the presence of total coliforms might be zero in 100 mL for safe water. FDA (2008) and US EPA (2018) prescribe that the presence of total coliform in drinking water should be $<2.2\%$ and $<5\%$, respectively. The WHO (2017) recommends that the permissible limit of Total Coliform and Fecal Coliform is less than 1.8 MPN/100 mL. The presence of *E. coli*, Total Coliform, Fecal Coliform in local brands, imported brands, and tube-well water were found to be 0.0 MPN/100 mL, $<1.8 \text{ MPN/100 mL}$, and $<1.8 \text{ MPN/100 mL}$, respectively, which indicates that the studied bottled water is free of microbial problem and suitable for drinking. The microbial data are not shown in any table due to their very low concentrations, and all

data are below the recommended limits given by various agencies [39–44]. Zamzam water was also found to be free of *E. coli*, Total Coliform, and Fecal Coliform, as reported in a recent study [67].

3.18. Status of Radionuclides in the Investigated Samples

Apart from physicochemical elements and toxic materials, the radioactive isotopes also contaminate drinking water. In groundwater, the concentration of natural radionuclides is connected to lithological leaching (granite and sandstone aquifers) [14]. The leaching amount is dependent on pH, temperature, water mineralization, etc. Human activities such as melting, mining, fuel combustion, agricultural fertilizer, etc. are responsible for radionuclides contamination of drinking water by [14]. Generally, the low level of radionuclides in water is safe, but a high level of radionuclides in water can cause a serious health problem. In this investigation, some radionuclides such as ^{40}K , ^{238}U , ^{226}Ra , ^{210}Pb , ^{137}Cs , ^{232}Th , ^{210}Po , and ^{222}Rn were found to be at pleasant levels, as per WHO (2017) recommendation [59]. The permissible limit for ^{40}K , ^{238}U , ^{226}Ra , ^{210}Pb , ^{210}Po , ^{222}Rn , ^{137}Cs and ^{232}Th activity in drinking water is 10 Bq/L, 180 mBq/L, 1.0 Bq/L, 200 mBq/L, 100 mBq/L, 100 Bq/L, 100 Bq/L, and 0.1 Bq/L [60,64,68–71], respectively. The radionuclides data are not shown in any table due to their very low concentrations, and all data lie below the recommended limits given by various agencies [39–44]. Concentrations of ^{238}U , ^{40}K , ^{137}Cs , and ^{232}Th in Zamzam water were reported to be 1.6 Bq/L, 3 Bq/L, 0.7 Bq/L, and 1.2 Bq/L respectively, which is higher than some agencies values [59,72,73].

3.19. Drawbacks of Bottled Water

The Food and Agriculture Organization (FAO), and the World Wildlife Foundations (WWF) agree that bottled water is not cheaper than tap water because its environmental cost is so high. Bottled water has been prescribed as a packaged food product sealed in plastic or glass containers or bottles. The plastic bottle is made of Polyvinyl Chloride (PVC) or Polyethylene Terephthalate (PET), which may contain organic (dioxins, biphenyl A, phthalates, and other organics) and inorganic (copper, antimony, lead, and nickel) contaminants that are carcinogenic agents and very harmful to human health, ecosystems, and the environment [74,75]. Depending on container type, treatment system, and storage duration, bottled water quality can be affected. The IBWA prescribes that bottles should not be reuse or refilled because they contain lethal pathogens and leaches degradable plastic into the water [76]. Reused plastic bottle releases a slow rate of degradable plastic in water per day — an average range of 1 to 3 ng per liter [8,77]. Evian and Perrier are natural spring bottled water brands imported from France. The sale price for bottles containing 330 and 750 mL of Perrier, 600 mL of Evian, and 500 mL of local brand water are BDT 126 and BDT 270, BDT 133, and BDT 15, respectively [15]. Consequently, majority of the people do not drink imported brands of water due to the high price. Seasonal variation, groundwater quality, water layer or source availability, natural disaster, and man-made activities are responsible for poor water quality [77,78]. However, this purified water can be compromised [42].

4. Conclusions and Recommendations

This study shows that imported brands, Zamzam water, and tube-well water contain more minerals than local brands of bottled water, and these mineral profiles fulfill the Dietary Reference Intakes requirements better than local brands. The obtained data show that the level of dissolved minerals in local brands of bottled water is very low, which is quite alarming, and may cause malnourishment. The local brand samples (Code D1 and D8) were good in quality to some extent but most were very poor in quality, from a scientific point of view. Local brands of bottled water in Bangladesh contain low nutrient levels, which is a matter of great concern. Another study showed that local companies neither maintain BDS-1414:2000 standards nor protect consumer rights [79]. Owing to a lack of

laws and policies for the production of quality drinking water, most companies produce distilled water, which is harmful to human health. Most water production companies do not mention the exact water source, adding derivatives and tested value of water quality parameters on bottle labels. The bottle's label also has no internal scaling, which is frowned upon by national and international standard guidelines. There is a significant difference in price between imported and local brands of bottled water. More than 50 national, regional, governmental and non-governmental industries produce drinking water with low quality.

On the other hand, drinking water-related rules and regulations in Bangladesh focus only on bacteria and toxins, while other parameters are not highlighted. However, concerning mineral balance, a good guideline is urgently required for private and public drinking water producers to preserve or improve the mineral content and mineral balance in their drinking water. Purified water should always be re-mineralized with the least level of macro-constituents such as Ca, Mg, HCO_3 , and SO_4 . A mixture of calcitic-dolomitic limestone is preferable for re-mineralization after RO [1,2]. Regulation is needed to prescribe to standard levels of minerals content. Specific tap or bottled water must have significant specific health value. Clean, safe, and re-mineralized water provides a win-win situation for public health. Thus, it should be included as an academic course at the university level in different educational phases. To provide clean and safe drinking water to all citizens, the bottled water industry should follow strong guidelines; however, currently, many of them use public water sources for their products, which are a threat to food safety. To protect the environment and ensure good health, it is vital that plastic bottled water is avoided and the use of water in glass bottles is encouraged, as is done in the EU. Globally, we need policy awareness regarding this issue, with the development and enforcement of regulations that will provide clean, safe, re-mineralized water [1]. The responsible government department should undertake strict inspection to improve water quality and improve water bottle labels to showcase the correct water characteristics. If we want safe mineral water, we must protect our water sources by keeping raw water as clean as possible, have a well-maintained distribution system, adequate treatment by including disinfection, and additional processes to remove or inactivate contaminants. Finally, strong water quality standards, regular observations, testing, operator training, monitoring, certification, reporting, placing public notices, contingency planning, adequate funding, and research are obligatory to ensure safe drinking water in Bangladesh and worldwide.

Supplementary Materials: The following are available online at www.mdpi.com/article/10.3390/w13152026/s1, Table S1: WHO recommended minimum level of some mineral nutrients should be available in drinking water, Table S2: Some common and rare chemical ingredients, Table S3: The Dietary Reference Intakes (DRIs) of selected mineral elements.

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