



Natural radioactivity levels in commercialized bottled drinking water and their radiological quality assessment

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ABSTRACT

This work presents radiological data on most commonly sold bottled water brands in the urban areas of Bangladesh. A significant fragment of the population use bottled water as their source of daily water intake. It is known from the literature that some varieties of bottled waters contain natural radionuclides in higher concentration than the traditional municipal tap water. In this study, concentrations of naturally occurring radioactive materials, particularly ²²⁶Ra, ²²⁸Ra, and ⁴⁰K, and its annual effective dose with its stochastic effects were determined in available brands of bottled mineral water manufactured in Bangladesh. The measurement was performed by gamma-ray spectroscopy using a hyper-pure germanium detector. The activity concentration of the radiologically important nuclides ²²⁶Ra and ²²⁸Ra were found within the permissible limits of 1 and 0.1 Bq l⁻¹, respectively, recommended by World Health Organization (WHO) in almost all samples. The annual effective doses caused by natural radioactivity of ²²⁸Ra for the infants (0–1 year) and adolescent (12–17 year) groups exceeded the recommended WHO guideline level of 0.1 mSv y⁻¹ for drinking water in most of the cases. Particularly for those age groups, the estimated doses are extremely high. In general, the carcinogenic risk due to natural radioactive doses for all bottled water samples were found below the acceptable limit of 10⁻³.

Keywords: Bottled water; Natural radioactivity; HPGe γ -ray spectrometry; Annual effective dose; Lifetime cancer risk

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1. Introduction

Water is a vital component for life support and a significant amount of radionuclides intake in humans that may get into the environmental aquatic system from natural and human activities. Therefore, its quality is of great importance to human health. Owing to the increased demand and consumption of bottled water around the world, there has been a growing concern about the quality of this corporate product. Groundwater derived from very deep aquifers by drilled wells is the main source utilized by the manufacturers of bottled water in Bangladesh. Most of the bottled water passed off as mineral water in Bangladesh.

Radioactive matters are omnipresent on Earth and environment that contain radioactive isotopes which enter human body via ingestion of food, water, etc. and also by inhalation. These isotopes are originated from primordial radionuclides, cosmogenic radionuclides, radionuclides produced in natural decay series, and anthropogenic radionuclides [1]. Natural decay series radionuclides (^{238}U , ^{232}Th , and ^{235}U) represent the most significant source of ionizing radiation on Earth, which contribute about 83% to the total effective dose received by the global inhabitants [1–3]. On the other hand, radioactive isotope ^{40}K contributes only 16% to the annual effective dose [2,4].

In water, radioactive substances can liquefy and contaminate potable water reserves. On the other hand, disposal of industrial effluents, wastes (domestic and industrial), such as sewage sludge and mining effluents, are other causes of groundwater pollution. Therefore, a remarkable portion of radiation exposure received by human being around the globe is due to drinking of contaminated and polluted water [5]. Water sourced from ground and communal tap water, is not free of natural radioactive isotopes from ^{238}U , ^{232}Th decay series, and ^{40}K [6]. As a decay product of uranium and thorium, ^{226}Ra and ^{228}Ra are the most common isotopes present in all environmental media such as rock, soil, and drinking water [7]. These isotopes are readily soluble into groundwater in contact with sands or soils, and their distribution in groundwater depends on the role of uranium and thorium contents in the aquifer, local geology of soil or rock, the geochemical properties of the aquifer solids, and the half-lives of it [8–10]. The actual mechanism involved in the infiltration of radium into groundwater is by dissolution of aquifer solids, by direct recoil across the liquid–solid boundary at the time of its formation, by radioactive decay of its parent substance in the solid and by desorption [8,11–13].

It is well-known that any types of radiation exposure carry some degree of health risk. Due to their high

radiotoxicity, the ^{226}Ra (alpha emitter) and the ^{228}Ra (beta emitter) recognized as carcinogens and are of key concern [3,14–19]. Radium in water may pose critical biological effect and can cause a life-threatening health hazard [16,20] when it is used for drinking. Since radium is chemically analogous to calcium, it has the potential to cause great harm by replacing calcium in bones [16,21,22]. Internally deposited radium can cause carcinomas and other disorders due to the emission of alpha and beta particles upon their decay. Those particles have the tendency to alter the genes in the cells (mutation) and cause variety of bone cancers [8,9,16]. According to the United State Energy Protection Agency (USEPA) model for ^{226}Ra and ^{228}Ra , additional lifetime risks linked to drinking water containing 0.185 Bq l^{-1} (5 pCi l^{-1}) is about 1 in 10,000 (if 10,000 people consume 2 l of this water per day for 50 y, one extra deadly cancer would be projected among the 10,000 exposed persons) [23].

Recommended WHO guideline levels for radionuclides ^{226}Ra and ^{228}Ra in drinking water is 1 and 0.1 Bq l^{-1} , respectively. In accordance with this guidance, drinking water is safe from the radiological viewpoint if the annual exposure dose caused from the presence of radionuclides does not exceed 0.1 mSv y^{-1} [24–26]. This value is based on average water consumption of 2 l d^{-1} . Due to the adverse health effects caused by environmental radiation created from bottled water and growing social concern, a remarkable number of investigators are engaged in the measurement of radioactivity in this water worldwide [3,5,6,8,10,18,19,25–32]. Moreover, various international organizations are working regarding this issue [2,4,14,24,33–44].

Nowadays, there has been a propensity among the people for tap water to be replaced by commercial bottled water in their daily diet due to the inadequate tap water quality and awareness of waterborne diseases. The local market of Bangladesh especially in urban areas is flooded with various brands of bottled drinking water. Bottled water is also widely used for the preparation of liquid milk from powder milk for children in lactation age. These corporate labeled waters are purified by physical and chemical methods to eliminate solid particles, to kill the *Pseudomonas*, *E. coli* and other biological objects such as bacteria, fungi, etc. The type of treatment applied to purify different brands of bottled mineral water in Bangladesh does not finger the problem of radioactivity. Furthermore, the majority of the bottled water producers in Bangladesh are not aware about the manufacturing process of bottled water in accordance with the guidelines of WHO and the International Bottled Water Association [38]. Alam et al. [45] reported the radioactivity levels of

²²²Rn, ²²⁶Ra, ²³²Th, and ⁴⁰K in well, tube well, and tap water in Chittagong region, Bangladesh. However, the data regarding the radioactivity concentration of ²²⁶Ra, ²²⁸Ra, and ⁴⁰K and its annual effective dose with its stochastic effects in the bottled drinking water are lacking. Therefore, the assessment of radioactivity levels in the bottled drinking water available in Bangladesh market and the associated doses are needed to be determined to ensure safety use by the population. Alongside, these measurements can be useful as base-line values for the estimation of the internal radiation doses with the intention of protecting the population from radiation hazard.

2. Materials and methods

2.1. Geology and hydrology of the study areas

2.1.1. Geology

The study areas are situated at the Madhupur Tract, the southern tip of a Pleistocene terrace. Two distinct geological segments named Madhupur clay of the Pleistocene age and alluvial deposits of recent age wrap the area and vicinity. The study area which has the featured topography and drainage is exposed by the oldest sediment Madhupur clay. Other major geographic characteristics are the low-lying bogs and marshes situated in and around the areas. The subsurface sedimentary chain (explored depth of up to 300 m), point up three separate units in which Madhupur clay of the Pleistocene age is one of them,

featured by reddish plastic clay with silt and very fine sand particles. This Madhupur clay composed of medium to coarse yellowish brown sand and occasional gravel unconformably superimpose the Dupi Tila formation of the Plio-Pleistocene age [46].

2.1.2. Hydrology

The main source of water is the Dupi Tila sands aquifer from where bottled water is drawn. The aquifer overlies by the Madhupur clay with a width of 8–45 m and varies in thickness from 100 to 200 m with averages of 140 m. The extraction of groundwater has increased more than 900% over the last 30 year; as a result, the level of water has gone down by 6 m in the last seven year. In 2011, the water table was 52 m beneath the mean sea level. In these circumstances, the bordering rivers act as sources of recharge as the Dupi Tila sands are exposed along the riverbeds [46]. Vertical percolation of rain and flood water, seepage from water mains and the drainage system, and leakage from standing water bodies within areas are the other sources of recharge from where most of the bottled water abstracted from drilled wells.

2.2. Sample collection and preparation

Ten brands of domestically produced bottled drinking waters with similar volume were used in this study which is available in the local market of Bangladesh. To confirm a broad and prevalent

Table 1

Chemical–physical parameters of different brands of bottled drinking water samples (mg l⁻¹) according to the manufacturers

Parameters	Bottled water brand code									
	BW-1	BW-2	BW-3	BW-4	BW-5	BW-6	BW-7	BW-8	BW-9	BW-10
TDS	<250	<500	<250	<250	<500	<250	<250	<250	<250	<500
As	0	0	0	0	<0.01	0	0	0	0	<0.01
Cd	<0.003	0	<0.003	0	<0.003	0	<0.003	<0.003	0	<0.002
Na	–	–	<4	–	–	–	–	–	–	–
Cl ⁻	<250	<250	<250	<20	<250	<20	<250	<250	<60	<250
F ⁻	–	–	<1	–	–	–	<1	–	0	<1
Ca	–	<75	<35	–	–	–	–	–	<50	–
Pb	<0.01	0	<0.01	0	<0.01	0	0	<0.01	–	<0.01
NO ₃	<4.5	<0.5	<2.5	<2.5	<4.5	<2.5	<4.5	<4	<0.002	<4
NO ₂	0	0	0	0	0	0	0	0	0	0
Hg	–	–	0	–	–	–	–	–	–	–
CN	0	0	0	0	<0.01	0	0	0	–	<0.01
Mg	–	<35	–	–	–	–	–	–	–	–
Cl	–	0	–	–	–	–	–	–	–	–
p ^H	6.4–7.4	6.4–7.4	6.4–7.4	–	–	6.4–7.4	6.4–7.4	6.4–7.4	6.4–7.4	6.4–7.4
Total hardness	–	–	–	–	–	–	–	–	<100	–

representation, the best-sold brands were selected to measure the natural radioactivity levels. Three samples of each brand (total of 30 samples) were collected randomly from the local markets in and around Dhaka city. This city was chosen for its huge population density and approachability (14.6 million inhabitants) [47]. Most of the bottled water factory is located in and around Dhaka city and supplies them all over the country. In order to keep the brand names anonymous, the bottled water samples were given a code from BW-1 to BW-10 and this convention was used throughout the study. According to the information given on the labels of the bottled water, nonradioactive contaminants/chemical parameters were noted (Table 1).

The water samples were placed within radon impermeable cylindrical polypropylene containers (Marinelli beaker) which eliminate leaks and minimize gamma-ray attenuation. The containers were cleaned by deionized water and soaked with weaken HCl and air dried to avoid sample contamination. After shaking the bottle of water, a 500-ml from each sample was transferred into a clean container and sealed with electrical tape to prevent any leaks so that the gaseous radon isotopes cannot escape. The samples were labeled with its code, preparation date, weight (etc.) and were stored for 4 weeks (more than seven half-lives of ^{222}Rn) in order to reach secular equilibrium of ^{226}Ra with its daughter products prior to gamma spectroscopy [48,49].

2.3. Instrumentation and measurement of radioactivity

In this experiment, a high-purity germanium (HPGe) semiconductor γ -ray spectrometer from Canberra was employed. The detector relative counting efficiency was 28.2% and an energy resolution of 1.8 keV FWHM at the 1,332.5 keV peak of ^{60}Co . The detector is shielded by a cylindrical lead guard (10 cm) with a permanent base and a changeable cover to minimize the peripheral γ -ray background in the measured spectrum. The HPGe detector was connected to the amplifier and MCA electronics contained within the NIM BIN for data acquisition. The Genie 2000 software was used to fit region of interests (ROIs) and identify the peaks. The ROIs were confirmed using eyes and making sure that no peaks had been excluded.

A cylindrical multi-nuclide gamma source (initial radioactivity: 5.046 μCi ; reference date: 1 September 2013; nature of source: solid; source package: 500 ml plastic Marinelli beaker) from Isotope Products Laboratories, Valencia, California, USA, containing ^{241}Am (59.541 keV), ^{109}Cd (88.040 keV), ^{57}Co (122.061 keV; 136.474 keV), ^{203}Hg (279.1,952 keV), ^{113}Sn (391.698 keV), ^{85}Sr (514.007 keV), ^{137}Cs (661.657 keV), ^{88}Y

(898.042 keV), and ^{60}Co (1,173.228 keV; 1,332.492 keV) was used for detector energy calibration and the absolute photo-peak efficiency evaluation [50]. A polynomial fitting function was used to fit the measured efficiencies, and the activity of the samples was determined by using these fitted efficiencies. In order to reduce the counting error, adequately long period (86,000 s) was allowed to count the sample. The actual activity of the samples was acquired by subtracting the background counts for the same counting time as the sample. Three samples from each brand of bottled water were analyzed and their weighted mean activity has been considered as the representative value of each brand.

The activity concentrations of ^{226}Ra , ^{228}Ra , and ^{40}K for each sample were derived from their respective characteristic γ -lines. Sufficiently discriminated and intense γ -lines of the relevant radionuclides were used for the reduction of uncertainties in activity determination. The activity concentrations of ^{226}Ra was determined by the gamma-ray lines at 351.93 keV (35.6%) from ^{214}Pb and 609.32 keV (45.49%) from ^{214}Bi , whereas the concentrations of ^{228}Ra was dogged via the gamma-ray lines at 583.19 keV (85.0%) from ^{208}Tl and 911.21 keV (25.8%) from ^{228}Ac . The single transition 1,460.822 keV (10.66%) was used to determine the activity concentrations of ^{40}K . Weighted mean approaches of the different values acquired for the individual gamma-ray energy values were used to derive the final activity concentration [50,51].

2.4. Statistical analysis

Most of the data were presented as the mean \pm standard deviation and subjected to statistical analysis using IBM SPSS version-20 software. Arithmetic mean (AM), geometric mean (GM), and standard deviation (SD) was determined by descriptive statistical analysis. One-way analysis of variance (ANOVA) was used to compare the data of activity concentrations among the different brands of bottled water. Statistically significant variations were verified between the individual means at $p < 0.05$ by employing post hoc Tukey HSD test.

3. Results and discussion

3.1. Radionuclide activity concentrations

The activity concentrations for the radionuclides (^{226}Ra , ^{228}Ra , and ^{40}K) in the samples were calculated by Eq. (1) [52]:

$$A_c = \frac{N \times 1,000}{\epsilon_\gamma \times I_\gamma \times T_s \times M_s} \quad (1)$$

where A_c is the activity in the sample (Bq l^{-1}), N is the net counts of the consequent photo-peak, ϵ_γ is the detection efficiency of the HPGe detector at the respective γ -ray energy, I_γ is the gamma-ray emission probability (intensity) of the corresponding γ -ray energy, T_s is the sample counting time in seconds, and M_s is the volume of the sample (ml), and 1,000 is the conversion factor from milliliter to liter.

In Table 2, activity concentrations of all samples were summarized, where we found that the radioactive isotopes ^{226}Ra , ^{228}Ra , and ^{40}K were present in all samples. The results of this study showed the radioactivity levels of ^{226}Ra in the aforesaid bottled mineral waters ranged from 31.1 ± 7.2 to $86.4 \pm 4.8 \text{ mBq l}^{-1}$, which are well below the WHO (2011) [24] guideline level of 1 Bq l^{-1} . Our findings are comparatively low and comparable with some literature data reported by

Rozmaric et al. from Croatia [3]; Ismail et al. from Jordan [6]; Jankovic et al. from Serbia [8]; Wallner and Jabbar from Austria [10]; Vasile et al. from Hungary [19]; Fredj et al. from Tunisia [25]; Ushko et al. from Belarus [26]; Chau and Michalec from Poland [27]; Beyermann and Bunger, from Germany [28]; and Joksi et al. from Bolkan cities in Serbia [29] and quite higher than some literature values reported by Benedik and Jeran from Slovenia [18]; Ndontchueng et al. from Cameroon [30]; and Godoy and Godoy from Brazil [31] (Table 3). Conversely, the activity concentrations of our studied bottled waters were within the range of well, tap, and tube well water of Chittagong region of the country [45].

^{228}Ra activity concentrations are found in the range of 22.6 ± 3.8 to $71.0 \pm 14.2 \text{ mBq l}^{-1}$, which are rather low and comparable with the findings of some

Table 2
Activity concentrations of ^{226}Ra , ^{228}Ra , and ^{40}K in different brands of bottled water samples

Sample code	Parameters	Radioactivity concentrations		
		^{226}Ra (mBq l^{-1})	^{228}Ra (mBq l^{-1})	^{40}K (Bq l^{-1})
BW-1 (3)	Range	81.2 ± 7.4 – 90.7 ± 6.6	49.5 ± 9.4 – 66.8 ± 7.6	2.57 ± 0.1 – 3.78 ± 0.07
	AM \pm SD	86.4 ± 4.8	59.3 ± 8.9	3.1 ± 0.6
	GM	86.3	58.8	3.0
BW-2 (3)	Range	69.4 ± 8.6 – 78.6 ± 7.5	57.6 ± 2.6 – 77.3 ± 8.3	4.52 ± 0.08 – 8.75 ± 0.06
	AM \pm SD	73.8 ± 4.6	65.9 ± 10.2	6.4 ± 2.1
	GM	73.7	65.4	6.2
BW-3 (3)	Range	42.3 ± 9.4 – 67.4 ± 8.7	43.7 ± 8.7 – 79.5 ± 4.5	5.36 ± 0.6 – 7.46 ± 0.2
	AM \pm SD	58.1 ± 13.8	62.8 ± 18.0	6.2 ± 1.1
	GM	56.9	61.0	6.1
BW-4 (3)	Range	23.3 ± 8.5 – 32.4 ± 6.2	19.4 ± 5.5 – 26.8 ± 2.8	1.66 ± 0.3 – 4.13 ± 0.08
	AM \pm SD	31.1 ± 7.2	22.6 ± 3.8	2.8 ± 1.3
	GM	30.5	22.4	2.6
BW-5 (3)	Range	42.1 ± 8.6 – 63.8 ± 7.4	56.1 ± 10.3 – 62.6 ± 6.8	5.05 ± 1.2 – 7.37 ± 0.7
	AM \pm SD	52.5 ± 10.9	58.6 ± 3.5	5.9 ± 1.3
	GM	51.8	58.6	5.8
BW-6 (3)	Range	32.7 ± 5.7 – 46.3 ± 8.3	27.3 ± 3.4 – 36.4 ± 2.6	3.92 ± 0.1 – 5.08 ± 0.04
	AM \pm SD	38.3 ± 7.1	32.2 ± 4.6	4.5 ± 0.6
	GM	37.8	32.0	4.5
BW-7 (3)	Range	59.3 ± 5.4 – 63.8 ± 9.2	40.6 ± 6.2 – 68.8 ± 2.8	2.33 ± 0.3 – 5.44 ± 0.1
	AM \pm SD	63.2 ± 3.7	57.9 ± 15.2	3.8 ± 1.6
	GM	63.2	56.4	3.6
BW-8 (3)	Range	59.7 ± 7.5 – 87.4 ± 3.8	55.2 ± 5.6 – 82.7 ± 3.4	2.57 ± 0.8 – 4.72 ± 0.2
	AM \pm SD	74.5 ± 13.9	71.0 ± 14.2	3.9 ± 1.1
	GM	73.6	70.0	3.7
BW-9 (3)	Range	52.6 ± 7.8 – 71.4 ± 5.4	57.4 ± 8.9 – 64.6 ± 5.1	5.34 ± 0.05 – 6.96 ± 0.04
	AM \pm SD	59.2 ± 10.6	60.5 ± 3.7	6.4 ± 0.9
	GM	58.6	60.4	6.3
BW-10 (3)	Range	25.6 ± 4.6 – 43.5 ± 3.5	27.7 ± 7.2 – 41.7 ± 8.5	3.48 ± 0.3 – 6.79 ± 0.1
	AM \pm SD	32.9 ± 9.4	33.3 ± 7.4	5.0 ± 1.7
	GM	32.1	32.8	4.8

Notes: The values in parenthesis indicate the number of samples analyzed. AM \pm SD denotes arithmetic mean \pm standard deviation and GM stands for geometric mean.

Table 3

Comparison of ^{226}Ra and ^{228}Ra concentration in drinking water with the reported values of other countries

Country	^{226}Ra concentration range (mBq l ⁻¹)	^{228}Ra concentration range (mBq l ⁻¹)	References
Croatia (bottled)	0.67–52.1	<0.05–35.8	[3]
Jordan (bottled)	3,520–5,500	400–2,400 (^{228}Ac)	[6]
Serbia (bottled)	<70		[8]
Austria (bottled)	2.0–211	<5–236	[10]
Slovenia (bottled)	2.4–17	1.9–5.3	[18]
Hungary (bottled)	4.1–89	6.3–76	[19]
Bangladesh (Chittagong) (well, tap and tube well)	12–82		[23]
Tunisia (bottled)	1,260–1,770	<380–1,490 (^{228}Ac)	[25]
Belarus (bottled)	<5–622	<10–2080	[26]
Poland (bottled)	0.8–525	≤10–393	[27]
Germany (bottled)	0.7–1,690	4.6–930	[28]
Serbia (bottled)	<26–60	<3–100	[29]
Cameroon (bottled)	6.98 ± 1.72–38.2 ± 5.71	5.12 ± 1.53–28.10 ± 7.28	[30]
Brazil (bottled)	27	97	[31]
Bangladesh	31.1 ± 7.2–86.4 ± 4.8	22.6 ± 3.8–71.0 ± 14.2	Present study

literature values published by Rozmaric et al. from Croatia [3]; Ismail et al. from Jordan [6]; Wallner and Jabbar from Austria [10]; Benedik and Jeran from Slovenia [18]; Vasile et al. from Hungary [19]; Fredj et al. from Tunisia [25]; Ushko et al. from Belarus [26]; Chau and Michalec from Poland [27]; Beyermann and Bunger from Germany [28]; Joksi et al. from Bolkan cities in Serbia [29]; and Godoy and Godoy from Brazil [31]; and below the WHO (2011) recommended limit of 0.1 Bq l⁻¹. However, some elevated ^{228}Ra activity levels were found in three brands of bottled waters (BW-2, BW-3, and BW-8).

Subsequently, there is a significant ($p < 0.05$) difference in activity concentrations of ^{226}Ra and ^{228}Ra were observed between some brands of bottled water samples. This may be attributed due to the fact that the geological origins of these investigated bottled waters are different and presumably drawn from various depths and would have passed through dissimilar geological layers [30]. Similarly, asymmetrical sharing of radium activity concentrations in these bottled water may depend on the geochemistry of parent nuclides, the interaction between water and the solid phases with which it comes into contact during its movement through the terrestrial crust and the physical and chemical properties of each brands of water sample [25,30]. The chemical–physical parameters of bottled water samples given in the bottle labels are listed in Table 1.

On the other hand, the radioactivity concentration for ^{226}Ra was found slightly higher than ^{228}Ra in the

same brands of samples (Table 2). However, there are no significant differences ($p > 0.05$) were observed between the radioactivity concentrations of ^{226}Ra and ^{228}Ra among the same brands of bottled water samples. This indicates that both radium isotopes are more or less equally soluble in water [53].

Furthermore, the obtained ^{40}K concentration values were found higher than those of radium nuclides. However, no significant variation ($p > 0.05$) in ^{40}K concentration was found among the brands of bottled water. This high ^{40}K activity in the study area may be due to local environmental conditions and geomorphology of the adjacent area from which there may be an elevated contribution of ^{40}K . Potassium is an abundance element in all environmental media. However, the isotope ^{40}K is radiologically less important compared to radium isotopes because it is homeostatically controlled in the human body and also an essential element [54].

3.2. Annual effective doses estimation

Radionuclide could directly reach to the gastrointestinal tract by ingestion of water and subsequently transported to the body fluids. High radiotoxicity makes an adverse effect to human health. Therefore, monitoring of individual annual effective dose needs particular attention due to its stochastic effect. In this study, the annual effective dose to a person owing to ingestion of natural radionuclides from the consumption of bottled water was estimated using Eq. (2) [32]:

$$D_{ef} = A_c \times W_{ac} \times D_{cf} \tag{2}$$

where D_{ef} is the annual effective dose ($Sv\ y^{-1}$) to an individual, A_c is the activity concentration of each radionuclides in the ingested drinking water ($Bq\ l^{-1}$), W_{ac} is the annual consumption of drinking water ($l\ y^{-1}$), D_{cf} is the ingestion dose conversion factor (committed effective dose per unit intake via ingestion) for members of the public ($Sv\ Bq^{-1}$), and values were taken from IAEA safety standard series, 2011 (Table 4) [55]. Considering the different consumption rates of water for people of different ages (since both W_{ac} and D_{cf} are age-dependent), the calculation for the annual effective dose for ^{226}Ra , ^{228}Ra , and ^{40}K was carried out separately for six age groups from 0 to 1 year, 1 to 2 year, 2 to 7 year, 7 to 12 year, 12 to 17 year, and >17 year with water consumption of 250, 300, 350, 440, 550, and 730 l per year, respectively (the consumption rate for drinking water is assumed similar for the consumption of bottled water).

The calculated annual effective doses were shown in Table 5. The age-dependent annual effective dose for ^{226}Ra , ^{228}Ra , and ^{40}K varied from 11.7 ± 3.8 (>17 y) to 68.5 ± 22.4 (0–1 y) $\mu Sv\ y^{-1}$, 26.4 ± 8.4 (>17 y) to 401.8 ± 127.3 (0–1 y) $\mu Sv\ y^{-1}$, and 19.9 ± 5.7 (12–17 y) to 75.9 ± 21.9 (0–1 y) $\mu Sv\ y^{-1}$, respectively. The dose values of ^{226}Ra and ^{40}K for all age groups were found below the WHO-recommended limit of $0.1\ mSv\ y^{-1}$ [24], except one brand (BW-1) for radio isotope ^{226}Ra showed little bit higher annual effective dose subjected to infant (0–1 y) group. On the other hand, the dose values of ^{228}Ra for the infant (0–1 y) and adolescent (12–17 y) groups exceeded the WHO guideline level of $0.1\ mSv\ y^{-1}$ for drinking bottled water in almost all cases (Fig. 1). Pertaining to the stages of human growth, the infants (≤ 1 y) and adolescent (12–17 y) periods are crucial in bone deposition and formation because more active calcium deposition and mineralization of bones occur during this period. This result indicates that those population groups are more likely vulnerable and subjected at risk (Fig. 1) due to their intensive bone growth in these stages. Therefore,

special care could be taken to avoid these bottled waters exceeding the recommended WHO values for radioisotopes. Among the radionuclides, ^{228}Ra is the main contributor to the intake dose, with a contribution of about 74% for infants (0–1 y) and 70% for adolescent (12–17 y) of the total estimated dose (Fig. 2) which is due to their higher dose conversion factor. Considering the high damaging effect of ^{226}Ra and ^{228}Ra , their existence in water leading to related health risks demand special awareness among people. Therefore, it is advised to pay more attention to the quality control of the bottled waters, as the radium could be replacing calcium in developing bones of infants and adolescent. This work by no means can be considered as a complete one; hence, more studies in this context are enviable especially in ^{222}Rn .

3.3. Dose-related stochastic effects

Long term exposure of ionizing radiation in biological cell/tissues is one of the etiologies for radiation-induced cancer. [52]. USEPA [56] suggested method was applied to evaluate the carcinogenic potencies due to the consumption of bottled water. The following Eq. (3) [52,57] was used to calculate the mortality and morbidity cancer risk and presented in Table 5.

$$ELCR = A_{ir} \times A_{ls} \times R_c \tag{3}$$

where ELCR, A_{ir} , A_{ls} , and R_c are the lifetime cancer risk, annual intake of radionuclide (Bq), average span of life (74 y), and mortality/morbidity risk coefficient (Bq^{-1}), respectively. The mortality and morbidity cancer risk coefficient for the consumption of tap water taken from USEPA [56] were used for lifetime cancer risk calculation. The mortality and morbidity cancer risk was varied from 1.1×10^{-5} to 3.2×10^{-5} and 1.7×10^{-5} to 4.6×10^{-5} , respectively, for radionuclide ^{226}Ra . Similarly, ^{228}Ra showed 3.4×10^{-5} to 7.3×10^{-5} and 3.2×10^{-5} to 10.2×10^{-5} for the above-mentioned cancer risk. These results suggested that the values are low

Table 4
Dose conversion factors ($Sv\ Bq^{-1}$) due to ingestion of radionuclides for different age groups⁽⁵⁵⁾

Radionuclides	Age groups					
	0–1 y	1–2 y	2–7 y	7–12 y	12–17 y	>17 y
^{226}Ra	4.7×10^{-6}	9.6×10^{-7}	6.2×10^{-7}	8.0×10^{-7}	1.5×10^{-6}	2.8×10^{-7}
^{228}Ra	3.0×10^{-5}	5.7×10^{-6}	3.4×10^{-6}	3.9×10^{-6}	5.3×10^{-6}	6.9×10^{-7}
^{40}K	6.2×10^{-8}	4.2×10^{-8}	2.1×10^{-8}	1.3×10^{-8}	7.6×10^{-9}	6.2×10^{-9}

Table 5
Age-dependent annual effective dose ($\mu\text{Sv y}^{-1}$) and lifetime cancer risk due to the ingestion of natural radionuclide from the bottled water

Radionuclide	Sample code	Age-dependent annual effective dose ($\mu\text{Sv y}^{-1}$)							Lifetime cancer risk ($\text{LCR} \times 10^{-5}$)	
		Age group	0–1 year	1–2 year	2–7 year	7–12 year	12–17 year	>17 year	Mortality	Morbidity
^{226}Ra	BW-1		103.8 ± 5.8	24.2 ± 1.3	19.6 ± 1.1	30.3 ± 1.7	71.0 ± 4.0	17.7 ± 1.0	3.2	4.6
	BW-2		88.6 ± 5.5	20.7 ± 1.3	16.7 ± 1.0	25.8 ± 1.6	60.6 ± 3.8	15.1 ± 0.9	2.7	3.9
	BW-3		69.8 ± 16.5	16.3 ± 3.9	13.1 ± 3.1	20.4 ± 4.8	47.7 ± 11.3	11.9 ± 2.8	2.1	3.1
	BW-4		37.3 ± 8.7	8.7 ± 2.0	7.0 ± 1.6	10.9 ± 2.5	25.5 ± 5.9	6.4 ± 1.5	1.1	1.7
	BW-5		63.1 ± 13.1	14.7 ± 3.0	11.9 ± 2.5	18.4 ± 3.8	43.1 ± 8.9	10.7 ± 2.2	1.9	2.8
	BW-6		46.0 ± 8.6	10.7 ± 2.0	8.7 ± 1.6	13.4 ± 2.5	31.4 ± 5.9	7.8 ± 1.5	1.4	2.0
	BW-7		75.9 ± 4.4	17.7 ± 1.0	14.3 ± 0.8	22.2 ± 1.3	51.9 ± 3.0	12.9 ± 0.8	2.3	3.4
	BW-8		89.5 ± 16.7	20.9 ± 3.9	16.9 ± 3.2	26.1 ± 4.9	61.2 ± 11.5	15.2 ± 2.9	2.7	4.0
	BW-9		71.1 ± 12.7	16.6 ± 3.0	13.4 ± 2.4	20.8 ± 3.7	48.6 ± 8.7	12.1 ± 2.2	2.2	3.1
	BW-10		39.5 ± 11.3	9.2 ± 2.6	7.5 ± 2.1	11.5 ± 3.3	27.0 ± 7.7	6.7 ± 1.9	1.2	1.8
	AM ± SD	68.5 ± 22.4	16.0 ± 5.2	12.9 ± 4.2	20.0 ± 6.5	46.8 ± 15.3	11.7 ± 3.8	2.1	3.0	
^{228}Ra	BW-1		454.3 ± 67.9	98.6 ± 14.8	73.5 ± 11.0	101.2 ± 15.1	172.0 ± 25.7	29.9 ± 4.5	6.1	8.5
	BW-2		505.1 ± 78.3	109.7 ± 17.0	81.8 ± 12.7	112.6 ± 17.4	191.2 ± 29.6	33.2 ± 5.1	6.7	9.5
	BW-3		481.4 ± 138.1	104.5 ± 30.0	77.9 ± 22.4	107.3 ± 30.8	182.2 ± 52.3	31.6 ± 9.1	6.4	9.0
	BW-4		173.0 ± 29.2	37.6 ± 6.3	28.0 ± 4.7	38.5 ± 6.5	65.5 ± 11.1	11.4 ± 1.9	2.3	3.2
	BW-5		449.4 ± 26.7	97.6 ± 5.8	72.8 ± 4.3	100.2 ± 5.9	170.1 ± 10.1	29.5 ± 1.8	6.0	8.4
	BW-6		246.8 ± 35.2	53.6 ± 7.6	40.0 ± 5.7	55.0 ± 7.8	93.4 ± 13.3	16.2 ± 2.3	3.3	4.6
	BW-7		443.8 ± 116.1	96.4 ± 25.2	71.9 ± 18.8	98.9 ± 25.9	168.0 ± 44.0	29.2 ± 7.6	5.9	8.3
	BW-8		544.5 ± 109.0	118.2 ± 23.7	88.2 ± 17.6	121.3 ± 24.3	206.1 ± 41.3	35.8 ± 7.2	7.3	10.2
	BW-9		463.7 ± 28.4	100.7 ± 6.2	75.1 ± 4.6	103.3 ± 6.3	175.6 ± 10.7	30.5 ± 1.9	6.2	8.7
	BW-10		255.5 ± 56.6	55.5 ± 12.3	41.4 ± 9.2	56.9 ± 12.6	96.7 ± 21.4	16.8 ± 3.7	3.4	4.8
	AM ± SD	401.8 ± 127.3	87.2 ± 27.6	65.1 ± 20.6	89.5 ± 28.4	152.1 ± 48.2	26.4 ± 8.4	5.4	7.5	
^{40}K	BW-1		48.7 ± 10.0	37.7 ± 7.7	23.6 ± 4.8	17.5 ± 3.6	12.8 ± 2.6	13.9 ± 2.9	2.9	3.9
	BW-2		102.0 ± 33.9	78.9 ± 26.3	49.3 ± 16.4	36.7 ± 12.2	26.8 ± 8.9	29.1 ± 9.7	2.9	3.9
	BW-3		97.6 ± 18.0	75.6 ± 13.9	47.2 ± 8.7	35.1 ± 6.5	25.6 ± 4.7	27.9 ± 5.1	2.9	3.9
	BW-4		44.0 ± 19.8	34.1 ± 15.4	21.3 ± 9.6	15.8 ± 7.1	11.6 ± 5.2	12.6 ± 5.7	2.9	3.9
	BW-5		93.7 ± 20.1	72.5 ± 15.6	45.3 ± 9.7	33.7 ± 7.2	24.6 ± 5.3	26.8 ± 5.7	2.9	3.9
	BW-6		71.5 ± 9.2	55.4 ± 7.1	34.6 ± 4.5	25.7 ± 3.3	18.8 ± 2.4	20.4 ± 2.6	2.9	3.9
	BW-7		60.4 ± 24.7	46.7 ± 19.1	29.2 ± 12.0	21.7 ± 8.9	15.9 ± 6.5	17.2 ± 7.1	2.9	3.9
	BW-8		61.3 ± 18.1	47.5 ± 14.0	29.7 ± 8.8	22.0 ± 6.5	16.1 ± 4.8	17.5 ± 5.2	2.9	3.9
	BW-9		101.0 ± 14.2	78.2 ± 11.0	48.9 ± 6.9	36.3 ± 5.1	26.5 ± 3.7	28.8 ± 4.1	2.9	3.9
	BW-10		78.5 ± 26.6	60.8 ± 20.6	38.0 ± 12.9	28.2 ± 9.6	20.6 ± 7.0	22.4 ± 7.6	2.9	3.9
	AM ± SD	75.9 ± 21.9	58.7 ± 17.0	36.7 ± 10.6	27.3 ± 7.9	19.9 ± 5.7	21.7 ± 6.3	5.4	7.5	

Note: AM ± SD denotes arithmetic mean ± standard deviation.

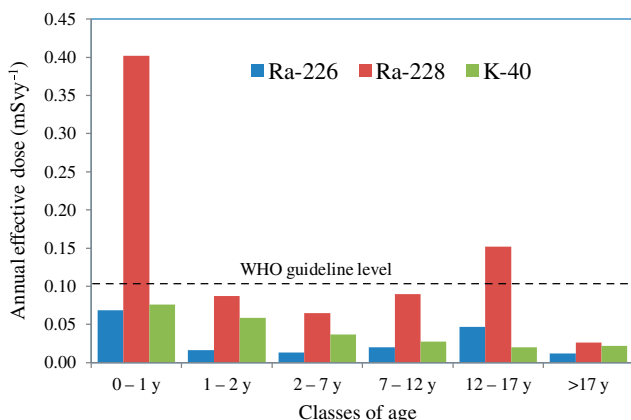


Fig. 1. Distribution of the average annual effective dose (mSv y⁻¹) due to ²²⁶Ra, ²²⁸Ra, and ⁴⁰K of different age groups.

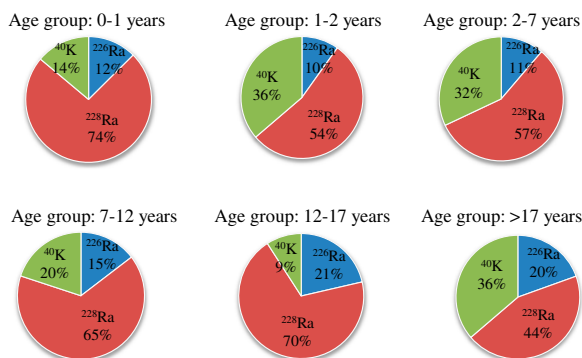


Fig. 2. Contribution of individual radionuclides in the annual effective doses for different age groups.

compared with the acceptable ELCR limit of 10⁻³ for radiological risk [52,57]. However, the radiogenic risk due to intake of ²²⁸Ra was noticed higher compared to ²²⁶Ra in these bottled water.

4. Conclusion

The most consumed brands of bottled drinking water marketed in Bangladesh were examined in order to assess their radiological quality. The measured activity concentrations of ²²⁶Ra, ²²⁸Ra, and ⁴⁰K were found well below the WHO-recommended guideline level. However, average concentrations of the radionuclides ²²⁶Ra and ²²⁸Ra were noticed very similar. The annual effective dose values of ²²⁶Ra and ⁴⁰K for all age groups were found below the WHO-recommended limit of 0.1 mSv y⁻¹. Whereas, the dose values of ²²⁸Ra for the infants (0–1 y) and adolescents

(12–17 y) groups were significantly high and exceeded in almost all cases the recommended WHO guideline level for drinking water. The stochastic effects from radionuclides (²²⁶Ra and ²²⁸Ra) are not significant. Since any kind of radiation has some degree of health hazard, it is suggested to the bottled water manufacturer to incorporate any suitable radionuclide removal method (i.e. mixed bed ion exchange, reverse osmosis, activated carbon, etc.) in their water purification process. If the drilled well indicates elevated levels of radioactivity, it is advisable to consider drilling new well into an aquifer with lower radioactivity levels. Information on the radioactivity concentrations of radionuclides should be provided in the bottled label as for the chemical–physical parameters. Additionally, regular monitoring and quality control of the radioactivity levels in bottled waters should be performed by the public sector to prevent subjecting the population to unnecessary radiation exposure. The results of this study may provide reference radiometric values of the bottled drinking water and may facilitate to establish a standard national protocol for natural radioactivity in the drinking water.

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