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Evaluation of tritium, gross alpha and gross beta radioactivity levels in tap and bottled drinking water in Singapore

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Abstract
With the aim of assessing the radiological impact on public health from water consumption, tritium and gross α-β radioactivity levels were determined in tap and bottled drinking water consumed in Singapore using ultra-low level liquid scintillation counting. Tritium and gross α activities were < MDA values, while gross β activity levels varied between 0.228 and 0.258 Bq/L in tap water samples. For bottled drinking water, the activity concentrations of tritium, gross α and gross β ranged from < MDA–1.59 Bq/L, < MDA–0.437 Bq/L and < MDA–1.33 Bq/L respectively. The annual total effective doses were also estimated for both children and adults due to intake of radionuclides from consumption of tap and bottled water. Our results showed that consumption of tap and bottled water presents insignificant radiological risk to the Singapore population.

Keywords Singapore · Tritium · Gross α-β · Tap water · Bottled water

Introduction

Water is essential for human survival, playing a critical role in maintaining many of the body's physiological functions. Due to its daily consumption and ability to transport pollutants, radiometric investigations of drinking water have been the subject of many environmental studies [1, 2]. Most of the radioactive contents in drinking water can be attributed to radionuclides originating from the natural decay series of ^{238}U and ^{232}Th, in addition to ^{40}K. Artificial radionuclides like ^{137}Cs, ^{90}Sr and ^{3}H can also be introduced into water sources from nuclear testing and accidents [3]. Many of these abovementioned radionuclides that are found in water are alpha (α) or beta (β) emitters. Because of their higher ionizing power compared to gamma radiation, α and β radionuclides produce more detrimental effects when they enter the human body. Exposure to high levels of these radionuclides for extended periods could potentially result in serious health effects such as cancer [4]. Therefore, it is of paramount importance to monitor drinking water supplies to ensure they are void of concerning levels of radionuclides.

The measurement of gross α-β activity is typically utilized as a rapid screening tool for evaluating radioactivity in drinking water. An overall quantification of total radioactivity levels due to α and β radiation in drinking water is achieved with the use of relatively fast, inexpensive, and simple radioanalytical methods. Further investigations to identify individual radionuclides are usually only required when elevated gross α or gross β activity levels are detected. The recommended WHO guideline levels for gross α and gross β in drinking water are 0.5 Bq/L and 1.0 Bq/L (excluding ^{40}K) respectively [5]. Below these reference levels of gross activity, the drinking water is considered to be acceptable for human consumption. By definition, gross α activity is the total activity of all α-emitters excluding gaseous radon while gross β activity is the total activity of all β-emitters excluding tritium [6].

Tritium is a radioisotope of hydrogen, with a half-life of 12.3 years, that emits low-energy beta radiation (E_{max}: 18.6 keV). It is produced naturally in the upper atmosphere from interactions of cosmic rays with atmospheric

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gases. Major anthropogenic sources such as past nuclear weapons testing, nuclear fission reactors, spent fuel reprocessing plants and tritium production facilities have in fact contributed larger amounts of tritium in the environment [7]. The world's largest nuclear reprocessing facility at La Hague was reported to release 11,400 TBq of tritium into the English Channel in 2020 [8]. The release of treated wastewater containing diluted tritium into the Pacific Ocean from the Fukushima Daiichi Nuclear Power Plant (FDNPP) is also expected to take place over several decades. Tritium exists primarily as radioactive tritiated water (HTO), entering water sources such as groundwater, rivers, lakes and oceans [9]. The HTO can circulate and spread through the water cycle, potentially contaminating drinking water supplies and ecosystems. The ingestion, inhalation or absorption of large quantities of tritium have being shown to pose potential radiological health risk to humans [10].

In Singapore, the two key sources of drinking water consumed by the population are tap and bottled water. Most of the bottled water commercially sold in Singapore are imported from other countries which tend to be more susceptible to elevated radioactivity. For instance, imported natural mineral waters from other countries can have higher radioactivity levels due to prolong contact with rocks and soils containing naturally occurring radioactive materials (NORM) like uranium, thorium and radium. In addition, many of these natural mineral waters are bottled directly at the source without significant treatment that would remove radionuclides. Therefore, it is crucial to monitor the radioactivity levels in these imported bottled drinking water to ensure that they are within international radiological safety standards. It is also important to understand that radioactivity data reported in tap water in other countries would not accurately reflect the actual levels that will be seen in Singapore's tap water as the radioactivity levels present in local tap water would be unique to Singapore. Thus, it remains necessary to conduct studies to gather the true radioactivity levels of tap water in Singapore especially when tap water is still the primary form of drinking water in majority of households in Singapore. To the best of our knowledge, no studies have been conducted till date to determine radioactivity levels in bottled drinking water and potable tap water in Singapore which would allow us to evaluate the radiation dose exposure to the population from water consumption. Here, in this work, we reported the first study carried out in Singapore to determine tritium, gross α and gross β activity levels in bottled and tap water using ultra-low level liquid scintillation counting (LSC). The age-dependent annual total effective doses due to consumption of drinking water were also evaluated for children and adults in the population to assess the radiological impact on public health.

Materials and methods

Sample collection

A total of 35 bottled drinking water samples of different brands and countries of origin were obtained directly from importers, as well as purchased from supermarkets and local grocery stores between November 2023 and April 2024 for analysis of tritium and gross α-β activities. 6 tap water samples were also collected in the same period from household taps from six different locations (Sengkang, Tampines, Clementi, Toa Payoh, Yishun, Queenstown) in Singapore for analysis. These two sources represent the main sources of drinking water consumed by children and adults in Singapore. In this study, the age range of children is defined from 1 to 17 years old and for adults is > 17 years old. Two portions of at least 500 mL of each water sample were collected and transported back to the National Centre for Food Science for tritium and gross α-β measurements respectively. For gross α and gross β measurements, samples were acidified with 3 M HNO_3 to pH 1–2 to prevent any adsorption of radionuclides onto the inner walls of the bottles before experimentation.

Radioactivity measurements of tritium

For tritium analysis, the water samples were subjected to a distillation process to remove interferences from other radionuclides. 100 mL of each sample was mixed thoroughly with 0.1 g of $KMnO_4$ and 0.5 g of NaOH in a 250 mL round bottom flask. The alkaline permanganate solution helps to oxidise radionuclides that are present in the volatile chemical forms such as radioiodine and radiocarbon to non-volatile forms prior to sample distillation. The distillation was performed at a temperature of 100 °C. The first 30 mL fraction of distillate was discarded, and the next 30 mL fraction was collected. An 8 mL aliquot was mixed with 12 mL liquid scintillation cocktail (Ultima Gold uLLT) in a 20 mL Teflon-coated scintillation vial. The vials were shaken vigorously for several minutes, and subsequently kept in the dark for 16 h before LSC measurements.

Tritium measurements were performed with the ultra-low level Quantulus 1220 (Perkin Elmer) liquid scintillation counter for 2000 min (200 min × 10 cycles). As tritium levels in the environment tend to be low, we opted to perform tritium counting for an extended time to lower the MDA. The spectra were acquired by WinQ and analyzed by EasyView software. The detection efficiency was measured using a tritium standard solution prepared by spiking a known amount of tritium from a tritium reference

source (Eckert & Ziegler, USA) in 8 mL tritium-free water and mixed with 12 mL liquid scintillation cocktail. For this sample to cocktail ratio, the detection efficiency was determined to be 18%. The background counting was performed using 8 mL tritium-free water mixed with 12 mL liquid scintillation cocktail. The counting window for tritium measurements was optimized to achieve the largest Figure of Merit (FOM), which was calculated by taking the square of the detection efficiency divided by background (E^2/B) and determined to be in the channel range of 40–160 (FOM of 453.8). Tritium activity concentrations of the water samples were calculated based on the following equation:

$$A_T = \frac{C_{\mathrm{Spl}} - C_{\mathrm{Bkg}}}{E \times V \times 60} \tag{1}$$

where A_T is the tritium activity concentration (Bq/L), C_{Spl} and C_{Bkg} are the count rates (CPM) of water sample and background respectively, E is the detection efficiency, V is the volume of water sample (L), and 60 is the conversion factor from CPM to CPS. The tritium activity concentrations were also presented in tritium units (TU), whereby 1 TU is approximately equivalent to 0.118 Bq/L.

The minimum detectable activity (MDA) for tritium measurements was calculated using the following equation:

$$\mathrm{MDA} = \frac{2.71 + 4.65\sqrt{C_{\mathrm{Bkg}} \times t}}{E \times V \times t \times 60} \tag{2}$$

Based on a counting time of 2000 min and sample volume of 8 mL, the MDA values for tritium measurements in this study were evaluated to be 1.00–1.05 Bq/L.

A tritium quench curve (Fig. 1) was also generated by varying the ratios of sample to organic scintillator in 10 different vials and spiking a known tritium activity into each vial [11]. The quench levels of the water samples were determined by means of the Spectral Quench Parameter of External Standard or SQP(E), derived from irradiating the samples with a ^{152}Eu gamma source for 1 min after initial tritium counting of samples. If the water samples display significantly different SQP(E) values, the detection efficiencies could be accurately determined using the tritium quench curve.

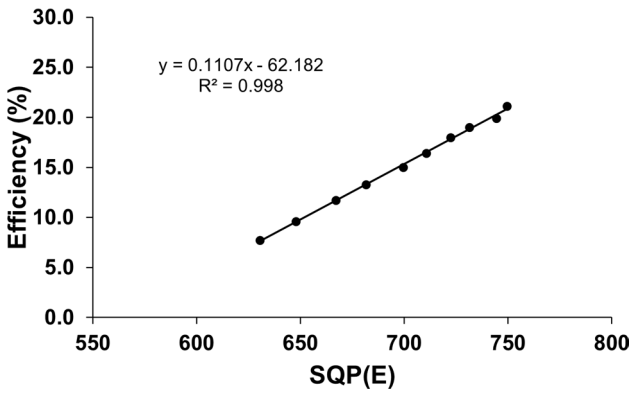

Fig. 1 Quench curve for tritium measurements

Radioactivity measurements of gross *α-β*

For gross α-β analysis, the total evaporation method was applied to the water samples. 100 mL of each acidified water sample was evaporated to dryness on a hotplate. The heating was performed at a temperature of less than 80 °C, to minimize volatilization of α and β radionuclides. The amount of total dissolved solids (TDS) after evaporation of the water samples were also measured as high solid contents can lead to attenuation of counts, leading to underestimation of the total α-β activity concentrations. The residue was then redissolved in 8 mL 0.1 M HNO_3 and mixed with 12 mL liquid scintillation cocktail (OptiPhase HiSafe 3). For samples (high TDS contents) with residues that cannot be completely dissolved in 0.1 M HNO_3, a smaller sample volume of 25 mL or 50 mL was utilized for evaporation. Background sample was prepared by mixing 8 mL 0.1 M HNO_3 with 12 mL liquid scintillation cocktail. The counting vials were shaken vigorously for a few minutes and kept in the dark for at least 16 h before measurements on the Quantulus 1220 liquid scintillation counter. All the water samples were measured for gross α and gross β simultaneously for 1000 min (200 min × 5 cycles). The spectra were gathered by WinQ and analyzed by EasyView software.

The Quantulus 1220 is equipped with a Pulse Shape Analyzer (PSA) to discriminate between α and β pulses. Optimal PSA setting was established by measuring ^{241}Am and ^{90}Sr/^{90}Y standard solutions, prepared in the same sample to scintillation cocktail composition as real samples using ^{241}Am and ^{90}Sr/^{90}Y reference sources (Eckert & Ziegler, USA), at different PSA levels. The percentages of α-spillover and β-spillover were plotted against PSA levels. The optimum PSA in this study corresponds to intersection of the two spillover curves, which is observed to be at PSA 62 with a minimum spillover of 5.5% (Fig. 2). Detection efficiencies were determined to be 100% for gross α and 97% for gross β using ^{241}Am and ^{90}Sr/^{90}Y standard solutions respectively. The gross α and gross β activity concentrations were calculated using Eq. 1, while the MDA values for gross α and gross β were determined using Eq. 2. The MDA values for gross α and gross β varied depending on the volume of sample used for the evaporation process. Typically, MDA for gross α is 0.020 Bq/L and MDA for gross β is 0.048 Bq/L for a 100 mL sample volume measured for 1000 min.

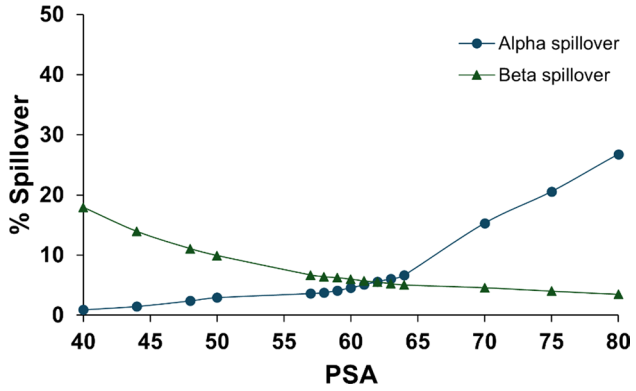

Fig. 2 Spillover curves for determination of optimum PSA

Assessment of annual committed effective dose

The annual committed effective doses due to intake of tritium were calculated for different age groups using the following equation:

$$D_T = A_T \times \mathrm{CR}_W \times \mathrm{DCF}_T \quad (3)$$

where D_T is the annual committed effective dose due to ingestion of tritium (Sv/yr); A_T is the activity concentration of tritium (Bq/kg); DCF_T is the dose coefficient factor of tritium which is 4.8×10^{-11} Sv/Bq (children) and 1.8×10^{-11} Sv/Bq (adult) [12]; CR_W is the annual consumption rate of drinking water and estimated to be 350 L/year for children and 730 L/year for adults [13].

The annual committed effective doses from intake of gross α-β were calculated based on the following equation [14]:

$$D_{\mathrm{GAB}} = A_a \times \mathrm{CR}_W \times \mathrm{DCF}_a \times 2 \quad \text{(for both gross } \alpha \text{ and gross } \beta\text{)} \quad (4)$$

where D_{GAB} is the annual committed effective dose due to ingestion of gross α-β (Sv/yr); A_a is the gross α activity concentration (Bq/L). Since more than 50% of the annual dose from water consumption can be attributed to the α-emitting radionuclide ^{226}Ra [15], DCF_a is the dose coefficient factor of ^{226}Ra. DCF_a corresponds to 9.6×10^{-7} Sv/Bq (children) and 2.8×10^{-7} Sv/Bq (adults) [12].

Results and discussion

Tritium activity concentrations

The radioanalytical method employed in this study to determine tritium activity concentrations in water samples using liquid scintillation counting was first validated with tritium certified reference material IARMA-009 (International Atomic Reference Material Agency) to ensure the accuracy and precision of our test method. As shown in Table 1, the tritium activity concentration obtained for IARMA-009 in our laboratory was within a relative bias of ± 5% of the certified value. In addition, our method was further validated through regular participation in proficiency test exercises organized by the IAEA (International Atomic Energy Agency). For the IAEA-TERC-2023-01 proficiency test, our reported value for tritium in Sample 1 was also within the range of the target value and an acceptable Zeta-score of < 1 was attained.

Table 1 Target and measured activity concentrations (Bq/L) of tritium in IARMA certified reference material and IAEA-TERC-2023-01 proficiency test sample

Reference material	Tritium		
	Target value (Bq/L)	Measured value (Bq/L)	Zeta-score
IARMA-009	25.4 ± 0.8	26.4 ± 1.9	–
IAEA-TERC-2023–01 Sample 1	29.0 ± 1.5	29.6 ± 2.6	0.19[a]

[a]Zeta-score was calculated using the target value and its uncertainty as Z-score was not available

The validated LSC method was applied to determine the radioactivity levels of tritium in bottled drinking water as well as local tap water. Although a tritium quench curve was generated in this study, correction for the detection efficiency was not required as no significant quenching was observed in the water samples. This was evident from comparable SQP(E) values of the samples to the tritium standard solution (Table 2). The lack of quenching in the measured samples is expected as the distillation process would have removed any impurities and interfering materials. As shown in Table 2, of the 35 bottled drinking water samples investigated, 11 samples exhibited trace levels of tritium. The tritium activity concentrations in these 11 samples ranged from 1.09 to 1.59 Bq/L (9.24–13.47 TU), while the remaining samples displayed activity levels less than the MDA values. Sample B22 from Norway yielded the highest tritium activity of 1.59 Bq/L amongst the water samples. Tritium levels examined for 6 household tap water samples in this study were all found to be below the MDA for tritium. The low activity concentrations of tritium detected were generally not of a major concern since these levels are far below the WHO recommended guidance level of 10,000 Bq/L for tritium in drinking water [13], and about two orders of magnitude lower than the tighter EU reference value of 100 Bq/L [16].

One explanation for the presence of trace levels of tritium in some of the bottled drinking water samples tested could be attributed to naturally occurring tritium in the environment. Studies have shown that formation of cosmogenic

Table 2 Tritium activity concentrations (Bq/L) in bottled and tap water in Singapore measured by liquid scintillation counting

Sample	Country of origin	A_T (Bq/L)	A_T (TU)	MDA_T (Bq/L)	SQP(E)
B1	France	< MDA	< MDA	1.01	719.63
B2	Singapore	< MDA	< MDA	1.01	721.01
B3	Italy	1.14 ± 0.10	9.66 ± 0.85	1.01	721.11
B4	Sweden	< MDA	< MDA	1.05	723.12
B5	Poland	< MDA	< MDA	1.05	722.25
B6	China	< MDA	< MDA	1.05	722.33
B7	Fiji	< MDA	< MDA	1.05	721.29
B8	Italy	1.21 ± 0.09	10.25 ± 0.76	1.05	722.03
B9	Slovenia	< MDA	< MDA	1.05	720.08
B10	Norway	< MDA	< MDA	1.05	721.47
B11	France	< MDA	< MDA	1.01	719.98
B12	Italy	< MDA	< MDA	1.01	722.69
B13	Sweden	< MDA	< MDA	1.01	722.00
B14	Turkey	1.49 ± 0.08	12.63 ± 0.68	1.01	722.28
B15	Switzerland	1.55 ± 0.10	13.14 ± 0.85	1.01	723.04
B16	Iceland	1.36 ± 0.08	11.53 ± 0.09	1.01	720.51
B17	Norway	1.26 ± 0.11	10.68 ± 0.93	1.01	721.57
B18	Austria	< MDA	< MDA	1.01	720.03
B19	Germany	1.26 ± 0.12	10.68 ± 1.02	1.01	718.19
B20	Indonesia	< MDA	< MDA	1.01	721.35
B21	Hungary	< MDA	< MDA	1.01	721.14
B22	Norway	1.59 ± 0.10	13.47 ± 0.85	1.01	720.92
B23	Greece	< MDA	< MDA	1.01	721.98
B24	South Korea	< MDA	< MDA	1.01	719.20
B25	Canada	1.32 ± 0.11	11.19 ± 0.93	1.01	720.86
B26	Taiwan	< MDA	< MDA	1.01	720.99
B27	Malaysia	< MDA	< MDA	1.01	721.47
B28	Poland	< MDA	< MDA	1.01	721.28
B29	Malaysia	< MDA	< MDA	1.01	720.68
B30	New Zealand	< MDA	< MDA	1.01	721.49
B31	Indonesia	< MDA	< MDA	1.01	723.41
B32	Finland	1.09 ± 0.10	9.24 ± 0.85	1.02	720.06
B33	Japan	< MDA	< MDA	1.04	719.23
B34	Japan	< MDA	< MDA	1.04	721.39
B35	Japan	1.46 ± 0.14	12.37 ± 1.19	1.04	719.05
T1	Sengkang	< MDA	< MDA	1.01	722.98
T2	Tampines	< MDA	< MDA	1.01	723.62
T3	Clementi	< MDA	< MDA	1.00	727.93
T4	Toa Payoh	< MDA	< MDA	1.00	734.55
T5	Yishun	< MDA	< MDA	1.00	723.09
T6	Queenstown	< MDA	< MDA	1.00	721.32
Mean ± SD (all samples, $n = 41$)[a]		1.10 ± 0.17	9.32 ± 1.44		

< MDA: Less than Minimum Detectable Activity (MDA)

[a]For activity concentrations < MDA, the MDA values for tritium were used in the calculations of the mean activity concentration

tritium tends to increase with higher latitudes [17, 18]. This was consistent with findings in our study where the bottled water samples detected with tritium were observed to originate predominately from regions with higher latitudes such as Switzerland, Iceland, Norway, Germany, Canada, and Finland. Other plausible reasons for the low levels of tritium detected could be due to past atmospheric nuclear weapons testing and nuclear accidents, as well as routine

liquid discharges from nuclear power plant operations [19]. The tritium concentrations observed in Singapore's tap water were found to be relatively lower as compared to bottled water samples imported from other countries. Potable tap water in Singapore is produced from treatment of rainwater captured through waterways and reservoirs, and desalination of seawater drawn from surrounding seas. Given that Singapore is situated close to the equator (low latitude) and geographically far away from countries with nuclear activities, the level of tritium in local rainfall and seawater in nearby seas is expected to be low.

The tritium activity levels determined in bottled and tap water in our study were compared to earlier radiological studies conducted on drinking water in other countries. Akata et al. studied an assortment of bottled drinking water commercially sold in Japan and found activity concentrations of tritium ranging from < 0.04 to 1.31 Bq/L, which were in agreement with values obtained for bottled water in our study [20]. For bottled drinking water marketed in Turkey and Azerbaijan, slightly higher tritium activity levels of < MDA–3.48 and < MDA–3.19 respectively were reported [21]. Compared to a study conducted in Serbia on locally produced bottled mineral water, our values were higher than the published tritium levels of 0.023–0.046 Bq/L [22]. This is likely due to the wide range of bottled water samples investigated, leading to greater variations in tritium activities observed in our study. An unexpectedly high activity concentrations of tritium ranging from 52 to 96 Bq/L were reported by Semerjian et al. [23] in several different brands of bottled water retailed in United Arab Emirates. As the activities were within the WHO permissible limit for tritium, the authors did not probe further to identify reasons for the elevated tritium concentrations. While tritium activity levels in tap water in Singapore were found to be less than MDA of 1.00–1.01 Bq/L, a similar survey on tap water in Turkey by Turhan et al. [24] reported significantly higher tritium activities ranging from < 2.0 to 9.1 Bq/L. Through an extended period of monitoring on drinking water in Portugal, Madruga et al. [25] have established tritium contents in tap water samples to be in the range of < 0.33–7.5 Bq/L. Additionally, two separate studies conducted recently in Thailand reported tritium activities in tap water collected from various parts of Thailand to be 0.41–0.75 Bq/L and < 9.4 Bq/L [26, 27]. Marginally higher tritium levels ranging from < MDA–4.84 Bq/L were also observed in local well and spring waters that serve as sources of drinking water to the population in a different study carried out in Italy [28]. Overall, it can be seen that the tritium activity values obtained for bottled and tap water in our study were largely comparable or lower than other similar studies in the literature (Table 3).

Gross α-β activity concentrations

The 41 water samples measured for tritium were concomitantly analyzed for gross α-β radioactivity levels. The WHO reference values for gross α and gross β were the screening levels adopted for bottled and tap water investigated in this study. Similarly, satisfactory results obtained from frequent participation in various proficiency test exercises demonstrated that our LSC method for gross α-β measurements was equally reliable and accurate (Table 4). Although the Z-score was not available for the FAPAS proficiency test, our measured values for gross α-β were close to the target values. A variety of key parameters such as TDS, SQP(E), and gross α-β activity concentrations measured in the water samples were presented in Table 5. Initial screening of the samples showed no notable quenching (similar SQP(E) values to α-β standard solutions), and therefore gross α and gross β measurements were performed at the optimum PSA 62 for all the water samples. The TDS contents determined in the samples varied from 11 to 3842 mg/L, which were within the limit of 5 g/L for such a method utilizing thermal concentration and LSC [29]. It is also clear in our study that samples with higher TDS values did not lead to quenching as shown by similar SQP(E) values between samples with high TDS and those with much lower TDS contents.

Gross α activity concentrations evaluated for the bottled drinking water samples ranged from < MDA–0.437 Bq/L, and gross β radioactivity levels were established to be between < MDA to 1.33 Bq/L. Additionally, gross α activity levels assessed in local tap water samples were all < MDA whilst gross β activities varied from 0.228 to 0.258 Bq/L. Likewise, activity concentrations of gross α-β in local tap water were lower when compared to imported bottled water samples. It is observed that only 5 bottled water samples (B1, B8, B14, B18, B27) had detectable gross α activities, with sample B14 from Turkey exhibiting the highest gross α activity of 0.437 Bq/L. These 5 samples were all less than WHO's gross α reference level of 0.5 Bq/L. In contrast, all the bottled and tap water samples apart for B13 were detected with gross β activities. The gross β activity concentrations were also found to be consistently higher than the gross α activity levels in the samples. This could be attributed to ^{40}K, a β-emitter, that occurs naturally in the environment in a fixed ratio to stable potassium. Due to potassium's high abundance in the environment, ^{40}K tends to be the dominant contributor to gross β radioactivity in drinking water. However, intake of ^{40}K is generally not seen as a health risk because potassium is an essential element for biological processes and is under strict homeostatic control in the body [40]. The WHO reference level of 1.0 Bq/L for gross β also excludes any activity contribution from ^{40}K. Interestingly, sample B14 which had elevated gross α activity was found to be the only sample in this study possessing

Table 3 Comparison of the range of activity concentrations (Bq/L) of tritium and gross α-β in drinking water from various studies reported in the literature

Country	Source	Activity concentrations (Bq/L)			References
		Tritium	Gross α	Gross β	
Japan	Bottled water	< 0.04–1.31	–	–	[20]
Turkey	Bottled water	< MDA–3.48	–	–	[21]
Azerbaijan	Bottled water	< MDA–3.19	–	–	[21]
Serbia	Bottled water	0.023–0.046	0.001–0.013	0.053–0.173	[22]
United Arab Emirates	Bottled water	52–96	–	–	[23]
Turkey	Tap water	< 2.0–9.1	–	–	[24]
Portugal	Tap water	< 0.33–7.5	–	–	[25]
Thailand	Tap water	0.41–0.75	–	–	[26]
Thailand	Tap water	< 9.4	< 0.018	0.117–0.730	[27]
Italy	Well and spring water	< MDA–4.84	–	–	[28]
Morocco	Bottled water	–	< MDA–0.211	< MDA–0.151	[29]
Saudi Arabia	Bottled water	–	0.03–0.18	0.01–0.34	[30]
Turkey	Bottled water	–	0.007–3.04	0.021–4.85	[31]
Albania	Tap water	–	0.010–0.126	0.029–0.884	[32]
Turkey	Tap water	–	0.005–0.164	0.012–0.787	[33]
Jordan	Tap water	–	< 0.082–0.484	< 0.216–0.984	[34]
Jordan	Tap water	–	< 0.110–0.724	< 0.220–0.362	[35]
Iran	Tap, well and spring water	–	0.012–0.115	0.023–0.332	[36]
Lebanon	Tap, well and spring water	–	< MDA–0.375	< MDA–0.418	[37]
Italy	Well and spring water	–	< 0.04–0.16	< 0.20–0.34	[38]
China	Well water	–	0.017–0.362	0.018–0.779	[39]
Singapore	Bottled water	< MDA–1.59	< MDA–0.437	< MDA–1.33	This study
Singapore	Tap water	< MDA	< MDA	0.228–0.258	This study

Table 4 Target and measured activity concentrations (Bq/L) of gross α-β in IAEA-TERC-2023–1 and FAPAS proficiency test samples

Reference material	Gross α			Gross β		
	Target value (Bq/L)	Measured value (Bq/L)	Z-score	Target value (Bq/L)	Measured value (Bq/L)	Z-score
IAEA-TERC-2023-01 Sample 1	27.8 ± 6.4	32.1 ± 3.9	0.67	17.3 ± 5.8	19.5 ± 0.6	0.38
IAEA-TERC-2023-01 Sample 2	13.2 ± 6.1	15.4 ± 5.2	0.36	135 ± 42	156.9 ± 1.4	0.52
FAPAS EMY 47[a]	6.29	6.23 ± 0.32	–	30.7	32.5 ± 0.6	–

[a]Standard uncertainty for target value and Z-score were not provided by FAPAS

gross β activity (1.33 Bq/L) exceeding the WHO gross β limit. This triggered us to first evaluate the radioactivity arising from the likely presence of ^{40}K in the sample. Examination of the nutrition label on B14 indicated a relatively high total potassium content of 25 mg/L. By using the ^{40}K percentage (0.012%) in total potassium and its specific activity, the activity concentration due to ^{40}K in sample B14 was calculated to be 0.80 Bq/L. Since the gross β activity in sample B14 after subtracting contribution from ^{40}K was less than 1.0 Bq/L, no further radiochemical analysis of specific radionuclides was warranted. The variations in gross α and gross β activities across different bottled water samples, including local tap water, could be attributed to differences in geological features of water sources such as mineralogical and geochemical composition of soils and rocks, differences in residence time of water in aquifers, varying environmental conditions at the water sources, and proximity of water sources to anthropogenic activities [41, 42].

The gross α and gross β radioactivity levels determined in bottled water in our study were higher than the reported values of 0.001–0.013 Bq/L and 0.053–0.173 Bq/L in a study by Jankovic et al. [22] on domestically produced bottled water in Serbia. Another study by Ait Bouh et al. on bottled mineral water in Morocco documented gross α and gross β activities in the range of < MDA–0.211 and < MDA–0.151 respectively [29]. Hassan et al. [30]

Table 5 Gross α-β activity concentrations (Bq/L) in bottled and tap water in Singapore measured using liquid scintillation counting

Sample	Country of Origin	TDS (mg/L)	A_α (Bq/L)	MDA_α (Bq/L)	A_β (Bq/L)	MDA_β (Bq/L)	SQP(E)
B1	France	712	0.057 ± 0.004	0.020	0.147 ± 0.006	0.049	703.6
B2	Singapore	186	< MDA	0.020	0.156 ± 0.007	0.049	701.5
B3	Italy	59	< MDA	0.020	0.072 ± 0.003	0.049	706.2
B4	Sweden	318	< MDA	0.022	0.110 ± 0.005	0.049	699.4
B5	Poland	722	< MDA	0.022	0.206 ± 0.008	0.049	699.6
B6	China	131	< MDA	0.022	0.109 ± 0.006	0.049	701.2
B7	Fiji	586	< MDA	0.022	0.260 ± 0.025	0.049	692.7
B8	Italy	1288	0.232 ± 0.019	0.043	0.416 ± 0.017	0.099	702.5
B9	Slovenia	2595	< MDA	0.043	0.133 ± 0.006	0.099	701.4
B10	Norway	18	< MDA	0.022	0.124 ± 0.006	0.049	701.8
B11	France	2524	< MDA	0.086	0.350 ± 0.017	0.198	705.2
B12	Italy	253	< MDA	0.023	0.087 ± 0.005	0.048	701.3
B13	Sweden	506	< MDA	0.024	< MDA	0.048	691.5
B14	Turkey	2420	0.437 ± 0.047	0.049	1.33 ± 0.132	0.097	702.1
B15	Switzerland	1370	< MDA	0.095	0.397 ± 0.021	0.193	705.5
B16	Iceland	174	< MDA	0.024	0.075 ± 0.003	0.048	695.5
B17	Norway	11	< MDA	0.024	0.061 ± 0.003	0.048	701.3
B18	Austria	2764	0.240 ± 0.016	0.048	0.535 ± 0.022	0.097	701.2
B19	Germany	3842	< MDA	0.048	0.525 ± 0.023	0.097	701.7
B20	Indonesia	295	< MDA	0.024	0.111 ± 0.005	0.048	698.4
B21	Hungary	372	< MDA	0.024	0.131 ± 0.004	0.048	701.4
B22	Norway	72	< MDA	0.024	0.118 ± 0.005	0.048	699.5
B23	Greece	451	< MDA	0.024	0.116 ± 0.005	0.048	704.3
B24	South Korea	132	< MDA	0.024	0.170 ± 0.008	0.048	699.2
B25	Canada	98	< MDA	0.024	0.086 ± 0.004	0.048	705.4
B26	Taiwan	29	< MDA	0.023	0.129 ± 0.006	0.048	704.2
B27	Malaysia	307	0.030 ± 0.003	0.023	0.232 ± 0.011	0.048	704.4
B28	Poland	753	< MDA	0.023	0.177 ± 0.006	0.048	698.7
B29	Malaysia	29	< MDA	0.023	0.091 ± 0.004	0.048	704.2
B30	New Zealand	187	< MDA	0.023	0.227 ± 0.011	0.048	704.4
B31	Indonesia	572	< MDA	0.023	0.383 ± 0.051	0.048	695.1
B32	Finland	188	< MDA	0.023	0.224 ± 0.009	0.048	698.8
B33	Japan	274	< MDA	0.023	0.165 ± 0.008	0.048	704.5
B34	Japan	188	< MDA	0.024	0.208 ± 0.022	0.049	698.9
B35	Japan	451	< MDA	0.024	0.310 ± 0.014	0.049	703.3
T1	Sengkang	82	< MDA	0.024	0.235 ± 0.010	0.049	701.2
T2	Tampines	112	< MDA	0.024	0.257 ± 0.014	0.049	703.6
T3	Clementi	125	< MDA	0.024	0.228 ± 0.013	0.049	702.9
T4	Toa Payoh	142	< MDA	0.024	0.258 ± 0.021	0.049	701.7
T5	Yishun	128	< MDA	0.024	0.249 ± 0.014	0.049	703.9
T6	Queenstown	129	< MDA	0.024	0.249 ± 0.012	0.049	703.9
Mean ± SD (all samples, $n = 41$)[a]			0.049 ± 0.078		0.232 ± 0.212		

< MDA: Less than Minium Detectable Activity (MDA)

[a]For activity concentrations < MDA, the MDA values for gross α-β were used in the calculations of the mean activity concentrations

reported gross α activities of 0.03–0.18 Bq/L and gross β levels of 0.01–0.34 Bq/L in imported bottled drinking water in Saudi Arabia. Compared to our study, higher gross α activities of 0.007–3.04 Bq/L and gross β activities of 0.021–4.85 Bq/L were reported by Kam et al. in an assessment of bottled mineral water in Turkey [31]. In addition, as shown in Table 3, our results for gross α-β activities measured in tap water in Singapore were found to be lower than

Table 6 Annual effective doses due to intake of tritium and gross α-β from consumption of bottled and tap water in Singapore for different age groups

Age category	Mean annual committed effective dose (μSv/yr)		
	Tritium	Gross α-β	Total
Children (1–17 years old)	0.019	32.93	32.95
Adult (> 17 years old)	0.015	20.03	20.05

or comparable to similar studies on tap water from Thailand [27], Albania [32], Turkey [33], and Jordan [34, 35]. Our findings were also compared against studies in the literature on other types of drinking water sources such as well and spring waters commonly consumed by people in the studied regions. Abbasi et al. [36] reported gross α and gross β activities of 0.012–0.115 Bq/L and 0.023–0.332 Bq/L in tap, well and spring waters in Iran. In another study by Ayoub et al. in Lebanon, higher gross α activities of < MDA–0.375 Bq/L and similar activity levels of < MDA–0.418 Bq/L for gross β were observed in tap, well and spring water samples investigated [37]. While gross β activity levels were close to values seen in tap water in our study, higher gross α activities were also reported in two separate studies on well and spring waters in Italy and on water samples collected from drilled wells in China [38, 39].

Evaluation of annual committed effective dose

To assess the radiation exposure to the Singapore population due to intake of radionuclides from consumption of bottled water and local tap water, the annual committed effective doses were determined using the mean activity concentrations of the radionuclides. Considering that the MDA values from our tritium and gross α-β measurements were reasonably low, for conservative evaluation, we took the MDA values for water samples which had activity concentrations < MDA for calculations of the mean activity concentrations and subsequently the effective doses. The annual committed effective doses tabulated for the different age categories were furnished in Table 6. The estimated annual effective doses due to ingestion of tritium were 0.019 μSv/yr and 0.015 μSv/yr for children and adults respectively. On the other hand, annual effective doses from intake of gross α-β were 32.93 μSv/yr for children and 20.03 μSv/yr for adults. The combined effective doses from consumption of drinking water were computed to be 32.95 μSv/yr (children) and 20.05 μSv/yr (adults). From the results, it is apparent that the radiation doses due to consumption of bottled and tap water were dominated by gross α-β while the dose contributions from tritium were negligible for both age groups. This can be mainly associated to the much lower dose coefficient factor of tritium. Although the consumption rate of drinking water for children were lower as compared to adults, the relatively higher dose coefficient factors in children used in the dose calculations translated to the higher effective doses observed in children over adults for tritium and gross α-β. The average annual effective dose values derived in our study for children and adults from water consumption were also found to be considerably lower when compared to the reference level of 0.1 mSv/yr in drinking water recommended by WHO for members of the public [5], indicating that the consumption of bottled and tap water poses insignificant radiological risk to the Singapore population.

Conclusion

In summary, we conducted the first occurrence study in Singapore to evaluate bottled drinking water as well as potable tap water consumed by the population for tritium and gross α-β radioactivity levels using ultra-low level liquid scintillation counting. The activity concentrations of tritium determined in all the water samples were far below the WHO limit of 10,000 Bq/L for tritium in drinking water. At the same time, after excluding activity contributions from the β-emitter ^{40}K, all the bottled and tap water samples analyzed for gross α-β were also found to be below the WHO reference levels of 0.5 Bq/L and 1.0 Bq/L for gross α and gross β respectively. The tritium and gross α-β activities observed in bottled and tap water in our study were largely comparable or lower than other studies on drinking water carried out in other regions in the literature. The annual committed effective doses determined for children and adults due to intake of tritium and gross α-β radionuclides from the consumption of bottled and tap water were 32.95 μSv/yr and 20.05 μSv/yr, notably lower than the WHO recommended limit of 0.1 mSv/yr for the public. Taken together, the results from our study have shown that radioactivity levels in bottled and tap water were generally low and the consumption of both sources of drinking water would constitute minimal radiological health risk to the Singapore population. These two important sources of drinking water in Singapore will continue to be rigorously monitored for compliance to radiological safety standards set by WHO, so as to ensure water safety and safeguard public health.

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Declarations

Conflict of interest The authors declare that there is no conflict of interest.

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