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Tritium concentration in bottled drinking water and internal dose assessment

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Abstract

This study aims to measure tritium (^{3}H) concentrations in different bottled drinking water around the world, understand the regional properties and their relation to tritium concentrations, and evaluate radiation dose from the consumption of bottled drinking water. A total of 41 different brands from 23 locations have an activities range of < 0.04–1.31 Bq L^{-1}. The countries located in the northern latitudes have the higher observed value which is consistent with the latitude effect. There is a high chance that other control factors could also affect the activity concentration of the drinking water, such as the continental effect, seasonal effect, or human activities. The maximum annual effective dose of 1.7×10^{-5} mSv was calculated using the highest value.

Keywords Bottled mineral water · Tritium · Annual effective dose

Introduction

People are usually exposed to ionizing radiation from both naturally occurring and anthropogenic sources whether internal or external [1, 2]. The main pathways radiation could travel through the body are ingestion, inhalation, or through the skin and result in accumulation of the dose inside [1, 3]. Typically, the public receives a radiation dose of about 0.3 mSv annually due to radionuclides of natural origin in their diet, including drinking water which contributes to around 0.01 mSv of the estimated dose due to the ingestion of naturally occurring radionuclides [3].

Mineral water and groundwater have the probability of containing some naturally occurring radioactive material (NORM) [4]. In addition, artificial radionuclides could appear in drinking water from several sources due to various causes, for instance, accidental or regular discharges from nuclear facilities, the medicine industry, military activities, and dispersion of atomic nuclear weapons fallout [3–5].

Tritium is a radioisotope of hydrogen with a half-life of 12.3 years [6]. This radionuclide emits very low-energy beta radiation that cannot penetrate the layer of human skin and can be absorbed by some materials such as plastic, glass, or metal [6]. Most natural tritium is from nuclear reactions between nitrogen or oxygen atoms in the upper atmosphere

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and secondary cosmic rays [7]. Furthermore, tritium can originate from various anthropogenic nuclear activities such as nuclear reactors and nuclear fuel reprocessing plants, which both release tritium into the environment. During the period of nuclear weapon testing from 1952 to 1963, tritium in northern Hemisphere precipitation was found to be drastically increased, covering the natural tritium signal in the atmosphere for decades before returning to the natural level [5, 8]. Tritium mainly exists in the form of tritiated water (HTO) and is produced in various ways to enter the environment [9]. Its presence in water sources could indicate industrial contamination [10].

After the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident which occurred in Fukushima prefecture, several studies about the behavior of tritium in terrestrial areas have been reported [11, 12]. There are also investigations about the tritium in drinking water sources conducted in several countries. Usually, the activity concentration that was found is lower than the WHO recommend guidance level. Tritium concentration in precipitation peaks in spring increases with latitude, which was observed in the northern Hemisphere and is known as the latitude effect [13]. It also tends to decrease in the summer season [5]. In addition, tritium concentrations in continental precipitation areas had always been higher compared to maritime precipitation, and the effect is called the continental effect [7].

Several investigations were conducted to study the radionuclide contents in bottled drinking water [14, 15]. Bottled water consumption also has a trend to increase in recent years as shown in Fig. 1. Kinahan et al. have reported on the assessment of the radionuclides in bottled drinking water which the brands were available in Japan [2]. Activity concentrations of ^{234}U, ^{235}U, ^{238}U, ^{226}Ra, ^{228}Ra, ^{222}Rn, ^{210}Po, ^{210}Pb, ^{40}K, ^{3}H, ^{14}C, ^{134}Cs, and ^{137}Cs were determined in 20 different brands of samples, and ^{234}U, ^{235}U, ^{238}U, ^{226}Ra, ^{228}Ra and ^{210}Po radionuclides activity concentration above the minimum detection activity value were determined. The estimated dose was below 0.1 mSv y^{-1} which is the WHO recommendation guidance level and ^{228}Ra contributes the most to the estimated value. Rožmarić et al. also reported on the natural radionuclides in bottled drinking waters produced in Croatia and their contribution to radiation dose [14]. Dizman & Mukhtarli studied tritium concentrations and consequent doses in bottled natural and mineral waters sold in Turkey and Azerbaijan [15]. However, the information on tritium in bottled drinking water is still very scarce regardless.

This study aims to measure tritium concentrations in different bottled drinking water commercially available around the world, understand the regional properties and their relation to tritium concentrations, and assess radiation dose from the consumption of bottled drinking water.

Therefore, this study was expected to contribute information on tritium in bottled drinking water from various brands across the world to the public for the safety monitoring of the foodstuffs.

Material and methods

Sampling

Bottled drinking water samples of 41 different brands from 23 locations: New Zealand, Chile, South Africa, Fiji, Indonesia, Cameroon, Brunei, Thailand, Philippines, Oman, Spain, USA, Italy, Bulgaria, Hungary, Austria, France,

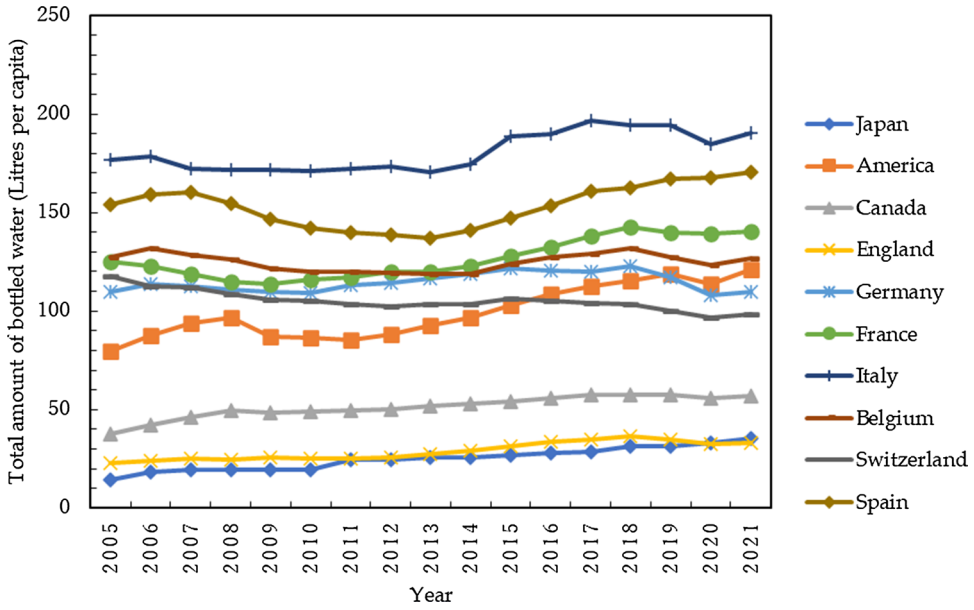

Fig. 1 The trend of bottled water consumption in different countries (The mineral water association of Japan, 2021) [16]

England, Northern Ireland, Scotland, Norway, Finland, and Dubai were purchased. The locations of the samples were distributed across the different continents which were shown in Fig. 2. This gives the wide distribution geographically. Since April 2015, the samples have been collected continuously.

Tritium measurement

Each sample was removed from the organic materials by distillation process to prevent the interference of organic compounds, and then the filtrated water was electrolyzed to be enriched with tritium using a solid polymer electrolyte membrane (XZ001, De Nora Permelec Ltd.) [7]. The enriched sample once again was distilled. An aliquot of 50 mL was mixed with 50 mL of a liquid scintillation cocktail (Ultima Gold LLT, PerkinElmer) in a 145 mL, low-diffusion polyethylene vial (LDPE vial) with an inner Teflon coating. In the study of bottled drinking water, counting efficiencies were 24.4% and the relative uncertainty was less than 9.3%. The limit of detection of tritium (LOD) was around 0.04 Bq L^{-1} which can be determined using the following equation [9];

$$\mathrm{LOD} = \frac{(2.71 + 4.65\sqrt{R_b \times t})}{V_s \times \%\mathrm{Eff} \times 60 \times t} \times 100,000 \quad (1)$$

where R_b represents the counting rate of the blank sample, t is the counting time, Vs is the volume of the water sample, and Eff is the % of the counting efficiency.

After shaking to make a homogenous cocktail, the vials were stored inside the low background liquid scintillation counter (LSC-LB5, Nihon RayTech). Tritium radioactivity measurements were done with LSC for 1,000 min (50 min × 10 times × 2 cycles), and counting efficiencies were determined using secondary standard tritium solution (118.17 ± 0.39 Bq L^{-1}, reference date: December 20, 2020) which was certified by tritium solution (SRM 4361C, NIST) [7].

Solid polymer electrolysis (SPE) system

In this study, an electrolytic apparatus, Tripure (XZ001, De Nora Permelec Ltd. Japan) was used. The diagram of the system shown in Fig. 3 included two water reservoirs, the semiconductor with the cooling function that has the Peltier

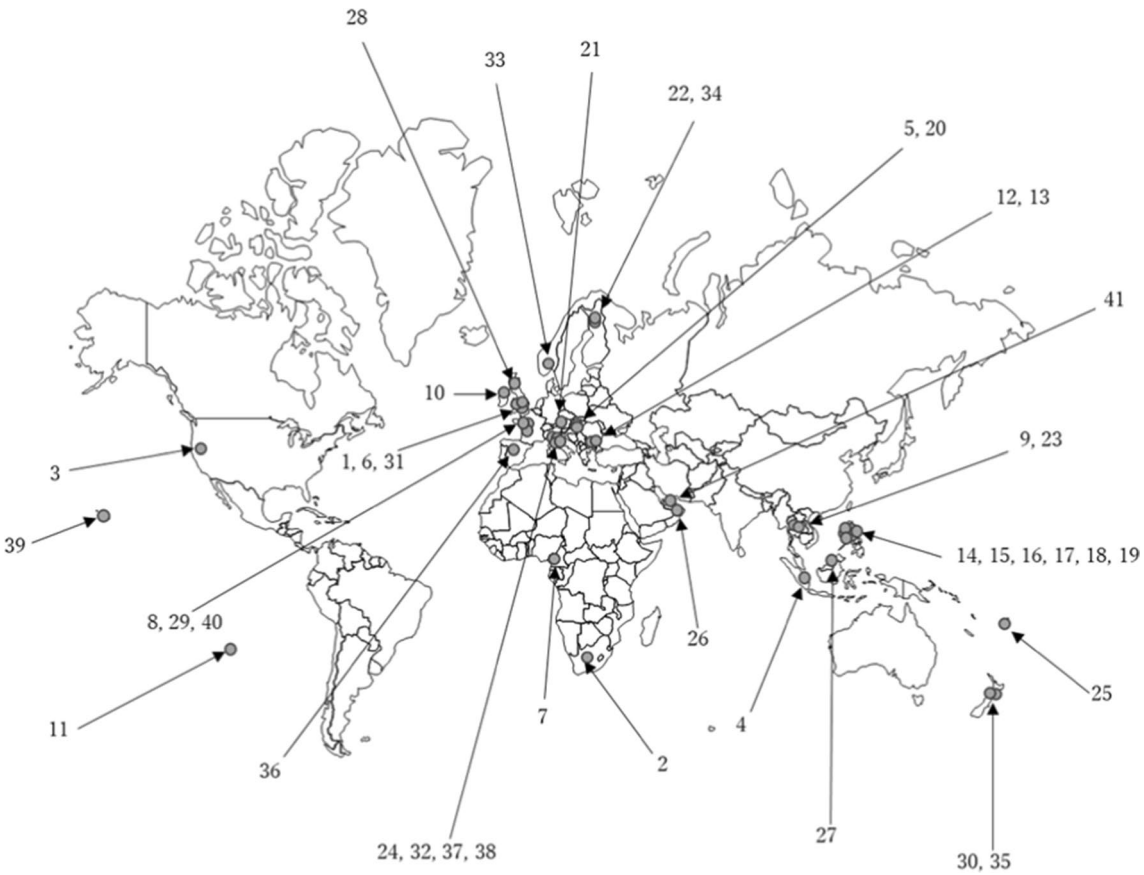

Fig. 2 Location of 41 bottled water samples

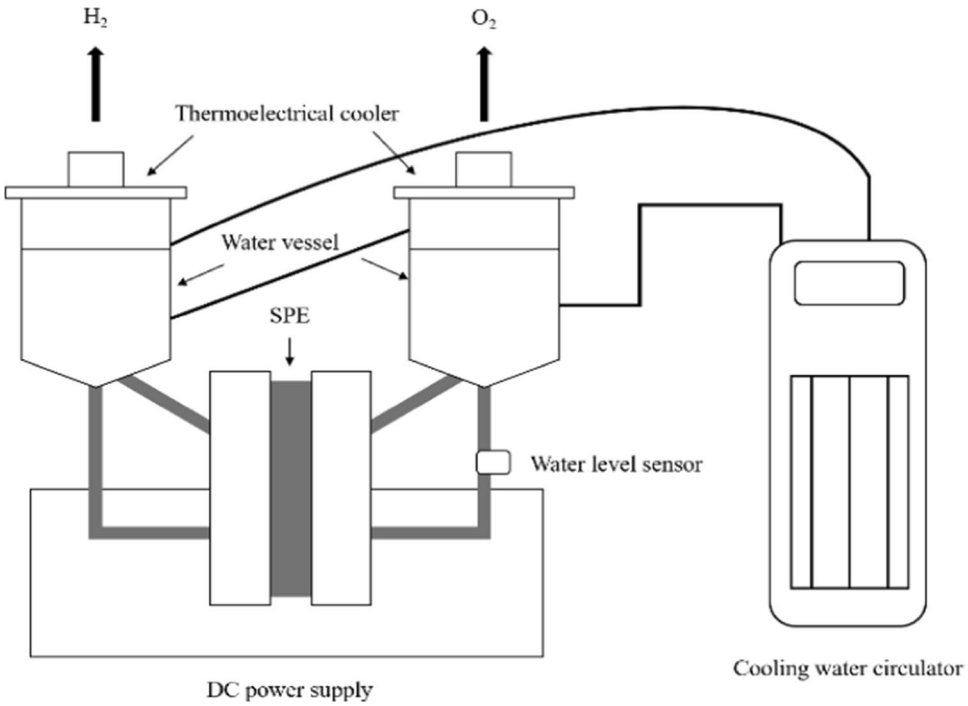

Fig. 3 The schematic diagram of the solid polymer electrolytic apparatus

effect, the electrolysis cell, and a DC power supply [17]. At the anode made of titanium, the sample water is oxidized to oxygen gas and hydrogen ions. Hydrogen ions are converted to hydrogen gas at the cathode by a solid polymer electrolyte membrane that consists of perfluoro-sulfonic acid. This membrane has the function to prevent the mixing of H_2 and O_2 produced by the electrolysis of water [18]. The two types of gases are released from each side of the water vessel through the thermoelectric cooler to avoid the loss of water. The water level sensor is set to shut down automatically to not exceed the lower limit of the water volume.

In this experiment, the initial volume of sample water was set at 800 mL. The electrolysis was started at a constant current of 50 A until the water was reduced to approximately 100 mL and then operated at 20 A until the volume reached the water level sensor at about 55 mL. The relationship between tritium concentration in sample water and water volume during electrolysis was defined by the following equation:

$$\frac{V_f}{V_i} = \left(\frac{T_f \times V_f}{T_i \times V_i}\right)^{\beta} \tag{2}$$

where T_i and T_f are initial and final tritium concentrations, V_i and V_f are initial and final water volumes, respectively, and β is the separation factor of tritium.

Normally, the separation factor β has been influenced by isotope fractionation due to evaporation, inside pressure of the electrolytic cell, vapor pressure of sample water during electrolysis, etc. Therefore, it is common to determine the apparent separation factor, β_a, by an experiment. Additionally, the tritium enrichment factor (Z_T) was defined using the following equation:

$$Z_T = \frac{T_f}{T_i} = \left(\frac{V_i}{V_f}\right)^{\left(1-\frac{1}{\beta_a}\right)} \tag{3}$$

In our previous studies, the initial water volume was set at 800 mL and the operation time was approximately less than 50 h [5].

Estimation of internal dose of bottled mineral water

The annual effective dose for tritium was determined according to the following equation:

$$D = D_{WI} \times D_{CF} \times Y \times C_A \tag{4}$$

where D is the annual effective dose (Sv) from the ingestion of tritium in the water, C_A is the tritium concentration in the water (Bq L^{-1}), D_{WI} is the daily consumption of water (the value was estimated to be 2 L d^{-1}) [10], D_{CF} is the dose conversion factor of ^3H for adults, (1.8×10^{-11} Sv Bq^{-1}) and Y is the ingestion period (365 d) [1, 3].

Result and discussion

The radioactive activity concentrations of tritium in bottled drinking water of 41 different brands are in the range of less than 0.04 and 0.05–1.31 Bq L^{-1}. There are 12 out of 41

samples of bottled drinking water brands that yield a concentration below 0.04 Bq L^{-1}, the LOD value. The countries located in the northern latitudes have the higher observed value which is consistent with the latitude effect. The sample from Finland (67°N, 26°E) yielded the highest value of all the samples as shown in Tables 1 and 2.

As some of the previous studies have already investigated tritium concentration in water samples, Table 3 demonstrated the general range of concentrations of tritium that were found. The range of concentration found in bottled mineral from Turkey, Azerbaijan, and Serbia were slightly higher compared to the result of this study.

According to the data in Table 3, tritium concentration did not exceed the WHO recommend guidance level [10] which is comparable to our study.

Other samples which the source located in the northern Hemisphere also have a trend that has a higher concentration, especially the location in the range of 50–60 degrees in the northern latitude as demonstrated in Fig. 4.

As it was already mentioned that the majority of natural tritium occurs from the interaction of cosmic rays with atoms in the upper atmosphere. Its concentration is influenced by latitude and seasonal change and cosmic ray fluxes at different altitudes and latitudes in the atmosphere [7, 13, 19].

Additionally, the atmospheric circulation is also different as latitude changes which affected the transfer behavior of tritium in the environment. Since ^{3}H is used as one of the tracers of the water cycling in the atmosphere, including surface and underground waters and ocean circulation, therefore, the movement of water is also a very important factor for the ^{3}H behavior study [20]. The climate conditions of the troposphere and stratosphere and the seasonal changes of the boundaries between these layers of the atmosphere create the pattern of the isotopes on the earth's surface which result in the variation of ^{3}H concentration [20].

Additionally, the concentration of tritium in precipitation observed by the IAEA Global Network of Isotopes in Precipitation (GNIP) in recent years shown in Table 4 have a comparable range of tritium to this study.

The slightly higher tritium concentration in Finland sample (1.31 Bq L^{-1}), the maximum value in this study might also influence from the reason which Finland is one of the countries located around the Baltic Sea. The waste from the countries that own the nuclear power reactors falls into the Baltic Sea which is shallow and isolated from the Atlantic Ocean, and the water has low purification efficiency. The investigation was conducted in the environment of the Gulf of Finland, and the result showed tritium concentration in vegetation and water sample [28]. The highest value in the water sample is 5.77 Bq L^{-1} was found in the wastewater from the Kuzmolovsky village and 27.15 Bq L^{-1} in the vegetation of Pushkin City. This might be due to the radioactive

Table 1 Summary of tritium activity concentration in bottled water (Bq L^{-1}) which has the counting error (a) applies to all the demonstrated results of the tritium concentrations

Sample	Location	^{3}H concentration (Bq L^{-1})
1	England	0.10 ± 0.01^{a}
2	South Africa	0.09 ± 0.01
3	America	0.15 ± 0.01
4	Indonesia	0.09 ± 0.01
5	Hungary	< 0.04
6	England	1.18 ± 0.07
7	Cameroon	< 0.04
8	France	0.50 ± 0.01
9	Thailand	0.19 ± 0.01
10	Northern Ireland	0.29 ± 0.01
11	Easter island	0.16 ± 0.01
12	Bulgaria	< 0.04
13	Bulgaria	< 0.04
14	Philippines	< 0.04
15	Philippines	< 0.04
16	Philippines	< 0.04
17	Philippines	< 0.04
18	Philippines	< 0.04
19	Philippines	0.08 ± 0.02
20	Hungary	< 0.04
21	Austria	0.12 ± 0.01
22	Finland	0.65 ± 0.02
23	Thailand	0.06 ± 0.02
24	Italy	0.45 ± 0.02
25	Fiji	0.05 ± 0.02
26	Oman	0.09 ± 0.02
27	Brunei	< 0.04
28	Scotland	0.51 ± 0.01
29	France	< 0.04
30	New Zealand	0.09 ± 0.02
31	England	0.13 ± 0.02
32	Italy	0.47 ± 0.02
33	Norway	0.39 ± 0.02
34	Finland	1.31 ± 0.02
35	New Zealand	0.05 ± 0.01
36	Spain	0.11 ± 0.02
37	Italy	0.44 ± 0.02
38	Italy	0.24 ± 0.02
39	Hawaii	0.31 ± 0.02
40	France	0.55 ± 0.02
41	Dubai	0.12 ± 0.02

Table 2 Maximum annual effective dose calculated using maximum tritium concentration from the bottled mineral water sample (mSv)

Isotope	Min (Bq L^{-1})	Max (Bq L^{-1})	Median (Bq L^{-1})	Maximum annual effective dose (mSv)
^{3}H	0.04	1.31	0.16	1.7×10^{-5}

storage being located near the Kuzmolovsky village and Pushkin city being nearer to the nuclear power plant resulting in the higher content of tritium however, the value has not exceeded the maximum concentration limit.

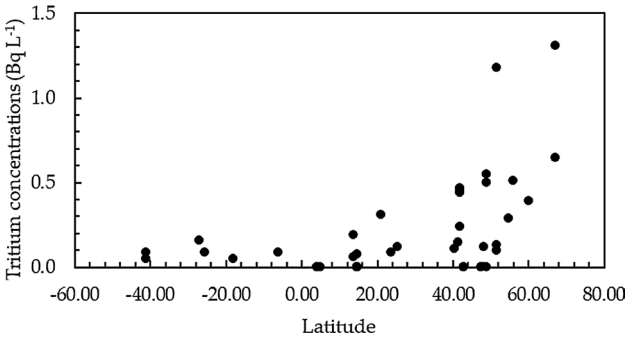

Fig. 4 Correlation between tritium concentrations and latitude of each country

Table 3 The comparison data of tritium activities in the water samples from the literature

Country	Sample	^{3}H activity concentration (Bq L^{-1})	Reference
Turkey	Bottled natural water	1.71–3.48	Dizman and Mukhtarli [15]
	Bottled mineral water	1.73–3.17	
Azerbaijan	Bottled natural water	2.21–3.04	
	Bottled mineral water	1.76–3.19	
Serbia	Bottled mineral water	0.02–0.05	Jankovic´ et al. [21]
Spain	Bottled mineral water	0.70–0.90	Palomo et al. [22]
Thailand	Tap water	0.41–0.75	Rittirong et al. [23]
Greece	Bottled water	0.60–2.00	Karamanis et al. [24]
California	Precipitation	0.15–0.66	Harms et al. [25]
Czech republic	Surface water	0.90–1.40	Mareš˘ova´ et al. [26]
	Precipitation	0.90–1.40	

Conclusions

Bottled drinking water samples of 41 different brands from New Zealand, Chile, South Africa, Fiji, Indonesia, Cameroon, Brunei, Thailand, Philippines, Oman, Spain, USA, Italy, Bulgaria, Hungary, Austria, France, England, Northern Ireland, Scotland, Norway, Finland, and Dubai were investigated. The locations of the samples were distributed across the different continents, giving the wide distribution geographically therefore, we can study the correlation between sample location and its concentration. The tritium concentration could be said to be comparable to the previous studies. The maximum annual effective dose of 1.7×10^{-5} mSv was calculated using the highest value (1.31 Bq L^{-1}). The estimated doses were less than the WHO-suggested recommendation threshold of 0.1 mSv for drinking water consumption.

Table 4 Tritium concentration data in precipitation in some monitoring stations from GNIP [27]

Sample	Location of the monitoring station	Year	^{3}H concentration (Bq L^{-1})
Precipitation	Austria (Vienna)	2021	0.57–1.63
	Cameroon (Douala)	2019	0.12–1.34
	Spain (Madrid)	2021	0.22–1.04
	Finland (Espoo)	2021	0.66–1.42
	France (Thonon-les-bains)	2021	0.51–1.44
	Philippines (Quezon city)	2018	0.06–0.18
	Thailand (Bangkok)	2020	0.12–0.21

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