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Levels of naturally occurring radioisotopes in local and imported bottled drinking water available in Québec City, Canada

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Highlights

- Naturally occurring radionuclides (^{210}Po, ^{226}Ra, ^{228}Ra ^{230}Th, ^{232}Th, ^{234}U, ^{235}U and ^{238}U) were measured in 15 bottled water brands available in Québec City, as well as in municipal tap water.
- Temporal analyses were done by sampling the bottled water in two separate periods in two consecutive years.
- The analyses revealed variations in U and Ra activities, demonstrating the need to routinely monitor each radionuclide.

- In most samples, ^{226}Ra was the major contributor to dose and calculated committed doses were systematically higher for younger individuals.

Abstract

Consumption of local and imported bottled water in Canada has greatly increased during the past three decades. While the presence of natural radioactivity is often overlooked when dealing with the water quality of these bottled products, it could contribute substantially to the uptake of radionuclides especially when sourced from regions with higher radioactivity levels compared to where it is consumed. In this study, the activity of several naturally occurring radionuclides (i.e., ^{210}Po, ^{226,228}Ra, ^{230,232}Th, ^{234,235,238}U) were measured in bottled water available in Québec, Canada after sample pretreatment and analysis by either radiometric or mass spectrometry approaches. ^{230,232}Th and ^{228}Ra concentrations were below minimum detectable activity levels in all samples tested. Analytical results for ^{234}U, ^{235}U, ^{238}U, and ^{226}Ra showed concentrations that ranged from 0.38 to 115 mBq/L, (2.2–313) x 10^{-2} mBq/L, 0.48–58.4 mBq/L, and 1.1–550 mBq/L, respectively. ^{210}Po was detected in only 5 samples and its activity ranged from 2 to 26 mBq/L. To determine variability in activity within brands, the same brands of bottled water were purchased during two consecutive years and analyzed. The possible radiological impact of the consumption of these types of water was assessed based on different drinking habit scenarios. Some of the imported water brands showed higher activity concentrations than local sources or tap water, suggesting that individuals drinking predominantly imported bottled water would receive a higher radiation dose than those who drink mainly local water.

Previous Next

Keywords

Uranium; Radium-226; Thorium; Polonium-210; Water; ICP-MS/MS

1. Introduction

According to Statistics Canada (Statistics Canada, 2018), the number of Canadian households using bottled water as a primary source of drinking water has increased by 150% between 1999 and 2005. A national study of water consumption habits by Jones et al. (2006)

indicated that 27% of the population uses bottled water as a primary source of drinking water in Canada. An example of growing demand is demonstrated by the results of a local study performed in southwestern Ontario, Canada. This study indicated that 34% of the population exclusively drank bottled water, and 14.5% drank both bottled and tap water, with 10–75% of all cold water consumed in the previous day being bottled (Pintar et al., 2009). Bottled water is classified based on the origin of the water it contains: artesian, volcanic, well, glacier, spring, mineral and purified, the last three categories representing 80% of the volume of bottled water sold worldwide in 2021 (Bouhlel et al., 2023). These waters can also be still or sparkling due to the presence of natural/added carbon dioxide. In 2021, sparkling/carbonated water represented approximately 10% of the bottled water market worldwide (Bouhlel et al., 2023). While local sources are available, a portion of the bottled water in Canada is imported. In December 2022, Canadians consumed about C$95 million worth of carbonated soft drinks, other beverages, and ice in Canada - including imported bottled water (Petruzzi, 2022).

Contaminant-free water quality is a crucial consumer confidence factor. The sustained increase in bottled water consumption in some countries has been partly attributed to consumer trust in its health and safety aspects over other drinking water sources (Doria, 2006). Among the possible contaminants studied in water sources (e.g., microorganisms, inorganic and organic chemicals, disinfection byproducts), the presence of natural radionuclides is often overlooked.

In an extensive review of the drinking water quality in Europe, Flem et al. (2018) reported radionuclide concentrations in bottled water ranging from <0.001 to 0.146 μg/L for thorium (Th) and <0.001–229 μg/L for uranium (U) in bottled water (n=884). Much lower ranges (<0.001 to 0.033 and <0.001–56.2 μg/L) were reported for tap water (n=579). All of the radionuclides measured, with the exception of ^{210}Po, are incorporated in the skeleton and have been shown to cause skeletal cancers. Linsalata, 1994 reported skeletal percentages of total body burden of 55, 83, 70–95, and ≥70%, for ^{232}Th, ^{238}U, ^{226}Ra, and ^{210}Pb (which decays to ^{210}Po), respectively. It follows that monitoring them properly is important to estimate absorbed doses and risks following their ingestion.

While the number of reports on the presence of natural radionuclides in bottled water worldwide has increased in recent years (Chmielewska et al., 2020), the number of samples analyzed is still small compared to the number of groundwater samples analyzed. Based on their assessment of radium isotopes in samples of bottled and medicinal water, Chmielewska *et al.* recommended that these types of water be more frequently surveyed for natural radionuclides. Recently, Piñero-García et al. (2022) published a report on the

radiological impact of naturally occurring radionuclides in bottled water available in the Swedish market. They reported that of the 26 brands studied, only three exceeded the threshold value for drinking water (0.1 mSv/y) - mainly due to the presence of ^{234,238}U and ^{228}Ra.

Very few Canadian studies have investigated the presence of radioactivity in bottled water. In 2000, a survey of bottled water available in Manitoba showed two samples with significantly higher than background levels of radioactivity, however, activity levels were not reported (Pip, 2000). Using accelerator mass spectrometry (AMS), Zhou et al. (2019) reported levels of ^{226}Ra for 7 brands of bottled water purchased in the Ottawa, Ontario region. The levels reported varied from the detection limit (3 fg, ~0.1 mBq) to 1377 fg/L (50 Bq/L), a highly unusual value. In a recent review article dealing with radionuclides contamination in Canada, Berthiaume (2023) noted that monitoring of radionuclides is sparse in most Canadian provinces; only one drinking water study (1984–2004) was performed in the province of Québec. This reported an arithmetic mean of 4.4 mBq/L for ^{226}Ra, which was well below the Canadian national guideline (500 mBq/L).

While the exposure to radionuclides such as uranium originates mostly from food, the contribution from water can be important. For example, the daily intake of uranium is extremely variable (*i.e.*, 31–98%) and is greatly dependent on its concentration in drinking water (Zamora et al., 2002). With respect to accumulation in the body, Larivière et al. (2013) were able to demonstrate a correlation between the average uranium concentration in water and in the bones of individuals that have consumed it. Given there is not much literature on radioactivity in bottled water, it is not surprising that most Canadians do not realize that a portion of their natural radioactivity intake may result from the ingestion of this water.

In Canada, bottled water is regulated as a food, and therefore must comply with the Food and Drugs Act and Regulations (Food and Drug Regulations, 2023). Under these regulations, the geographical location of the water source, the total dissolved mineral salts, and the total fluoride must be expressed on the label. In 2002, Health Canada and the Canadian Food Inspection Agency (CFIA) began consultations about the specific requirements for some other parameters, including radionuclide content (CFIA, 2002). However, to the best of our knowledge any identified requirement was never implemented within regulation.

However, the CFIA (2002) noted that "Radium-226 is the only *(sic)* radionuclide found naturally in water, and it occurs only in underground water, not in surface water. Because this standard is in the Guidelines for Canadian Drinking Water Quality, it would also apply to prepared water" such as bottled water. The present study and others show that the

veracity of this statement fails when tested against the results of bottled water surveys available in Canada. Maximum acceptable concentrations (MAC) in drinking water for selected natural radionuclides are presented in Table 1.

Table 1. National and provincial maximum acceptable concentration (MAC) in drinking water for various naturally occurring radionuclides.

Radionuclide	Canadian MAC[b] (mBq/L)	Ontario MAC[c] (mBq/L)	Québec MAC[d] (mBq/L)
Radium-226	500	600	500
Radium-228	200	500	N.R.
Thorium-230	N.R.	400	N.R.
Thorium-232	N.R.	100	N.R.
Uranium-234	250[a]	4000	250[a]
Uranium-235	10[a]	4000	10[a]
Uranium-238	250[a]	4000	250[a]
Polonium-210	N.R.	200	N.R.

a Based on its chemical nature, the regulation stipulates 20 μg L^{-1} which translate into the activities presented in the last column of the table based on isotopic abundance of 0.000054 (^{234}U), 0.007204 (^{235}U), and 0.992742 (^{238}U) (Health Canada, 2019).

b (Health Canada, 2009).

c (Ministry of the EnvironmentOntario, 2003).

d Compilation of Québec Laws and Regulations, 2023.

Where the content of radionuclides in bottled water brands has been published, it is typically for a specific water lot and reflects the presence of radionuclides just as water was bottled at its source. However, it has also been shown that the occurrence and distribution of radionuclides in groundwater fluctuates, depending on many factors including the local geology and geochemical parameters. For example, Dematatis et al. (2020) recently

demonstrated spatial and temporal variability in radium concentrations in the Wisconsin Cambriam-Ordovician aquifer system, highlighting the importance of examining compliance data sets for temporal trends. Sadly, as no compliance data are yet available for natural radioactivity in bottled water, fluctuation in the concentrations of radioactivity for a specific brand cannot be correctly ascertained. This means consumers would not be able to know intakes resulting from consuming a certain brand for an extended period.

The objective of the present study is to determine the level of radioactivity in bottled drinking water available in the Canadian market, and to determine if activity fluctuations in the most common radionuclides should be expected. For this study branded samples were collected during two distinct periods (2021 and 2022). Finally, based on the measurements performed and bottled water consumption patterns, we estimate the radiation doses received by Canadians from bottled water in 2021 and 2022.

2. Materials and methods

2.1. Sample collection

Bottled drinking water samples were purchased from local stores in Québec City, QC, in June to August 2021 and June to August 2022 (Table 2). Approximately 3 L of each brand was bought during each year of study. The samples were left unopened at room temperature from the time of purchase to the time of analysis. Based on the information provided, bottled waters were divided into one of two categories: mineral bottled water (MBW) or spring bottled water (SBW). Municipal tap water (MTW), collected on the University campus, was also analyzed as an example of the drinking water available to the local Québec City population (see Table 2).

Table 2. Description of the brands of bottled water (type and location) and the water composition (in mg/L) provided by the manufacturer.

Brand	Water Type[a]	Bottling Location (ON)	$[Ca^{2+}]$	$[Na^{+}]$	$[Mg^{2+}]$	$[K^{+}]$	$[Cl^{-}]$	$[HCO_3^{-}]$	$[NO_3^{-}]$	$[SO_4^{2-}]$	$[F^{-}]$	TDS
A	MTW[b]	Canada	15	15	2	0.8	22	N.A.	0.3[c]	20	<0.1	N.A.
B	SBW	Canada	7	450	6	3	350	560	N.A.	0	N.A.	1000
C	MBW	Canada	42	10	12	1	60	77	0	17	9	N.A.
D	MBW	France	103	1172	10	66	235	2989	N.A.	138	<1,5	3378

Brand	Water Type[a]	Bottling Location (ON)	$[Ca^{2+}]$	$[Na^{+}]$	$[Mg^{2+}]$	$[K^{+}]$	$[Cl^{-}]$	$[HCO_3^{-}]$	$[NO_3^{-}]$	$[SO_4^{2-}]$	$[F^{-}]$	TDS
E	MBW	France	240	5,2	42	N.A.	N.A.	384	4,4	400	N.A.	1084
F	NSW	France	150	9,6	3,9	1	19,5	420	7,3	25,3	0,2	N.A.
G	MBW	France	153	180	80	N.A.	54	1250	N.A.	35	1,2	1100
H	MBW	France	549	14,2	119	N.A.	N.A.	383,7	N.A.	N.A.	N.A.	2513
I	MBW (volcanic)	France	12	12	8	6	15	74	7,3	9	N.A.	130
J	MBW	Slovenia	210	470	90	75	52	2000	<2,2	98	N.A.	3000
K	MBW	Greece	90,1	4,03	4,5	1,23	5,62	240	2,93	14,4	0,1	245
L	MBW	Romania	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	0,54	1400
M	MBW	Germany	348	118	108	11	40	N.A.	N.A.	N.A.	N.A.	2479
N	MBW	Armenia	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.
O	MBW	Italy	166	30	49	2,1	49,6	244	2,8	401	0,5	853
P	MBW	Spain	6,6	47,7	5,2	5,0	14,7	160	N.A.	N.A.	0,48	179

N.A. – Not available.

a Based on the description provided by the supplier. MTW – Municipal Tap Water; SBW – Spring Bottled Water; MBW – Mineral Bottled Water.

b Based on data provided by the Division de la qualité de l'eau, Ville de Québec (Beaulieu, 2023)

c As NO_2^{-}/NO_3^{-}

2.2. Materials

Solutions and standards were prepared using ultrapure water with a resistivity of 18.2 MΩcm obtained from a Milli-Q purification system from EDM Millipore (Etobicoke, ON, Canada). Plasma Cal standard solutions containing 1000 mg/L of U, Th, Rh, and Tl purchased

from SCP Sciences (Baie d'Urfé, QC, Canada) were used to prepare calibration solutions, internal standards, and spiking. ^{226}Ra obtained from the National Institute of Standards and Technology (NIST; Gaithersburg, MD, USA) was used as a calibration standard and to assess separation recovery. ^{209}Po obtained from NIST (Gaithersburg, MD, USA) was used as a tracer for ^{210}Po recovery. Trace metal concentrated hydrochloric and nitric acid from Fisher Scientific (Ottawa, Ontario, Canada) and ACS grade ammonia solution from VWR (Radnor, PA, USA) were used for sample preparation, elution solutions, and pH adjustments.

Elution solutions for the separation procedures were made with ethylenediaminetetraacetic acid (EDTA) diammonium salt and nitrilotriacetic acid (NTA) diammonium salt from Fisher Scientific (Ottawa, ON, Canada), ammonium chloride salt and glacial acetic acid (AcOH) from VWR (Avantor; Radnor, PA, USA), 1,2-diaminocyclohexanetetraacetic acid monohydrate (CDTA) from Sigma-Aldrich (St-Louis, MI, USA), and oxalic acid from Alfa Aesar (Tewksbury, MA, USA). These salts and the AcOH were all reagent-grade purity or higher.

A cation exchange resin (AG50Wx8, 100–200 μm mesh size, H^+ form) for radium preconcentration, a TRU resin (octylphenyl-N,N-di-isobutylcarbamoylphosphine oxide, 50–100 μm mesh size) for U and Th preconcentration, and a Sr spec resin (4,4′ (5')-di-t-butylcyclohexano-18-crown-6, 50–100 μm mesh size) for ^{210}Po preconcentration were all purchased from Eichrom Technologies (Lisle, IL, USA) as pre-packed 2 mL cartridges.

2.3. Isotopic analyses

The uranium (^{234,235,238}U), thorium (^{230,232}Th) and radium (^{226,228}Ra) isotopes studied were analyzed by inductively coupled plasma mass spectrometry (ICP-MS) on an Agilent 8900 (Agilent Technologies, Santa Clara, CA, USA). The polonium (^{210}Po) analyses were performed using an Alpha Analyst (ORTEK, Oak Ridge, TN, USA).

2.4. Sample preparation

The purchased bottles were first shaken vigorously by hand for a few seconds and then opened to release any dissolved CO_2. This process was repeated for up to 5 min, or until the water seemed flat. The water bottles were then further degassed using an ultrasonic bath (Model 15,337,426, Fisher Scientific, Ottawa, ON, Canada) for 90 min while the bottle is uncapped using the degas mode and subsequently vacuum filtered with 0.45 μm filter (Cytiva; Marlborough, MA, USA). The filtrate was finally left under vacuum for an additional 10 min to ensure complete degassing of the water. This process eliminates most of the dissolved CO_2 from the sample. These steps are necessary to avoid the presence of CO_2

during the loading of the sample into the chromatographic resins which could impact the preconcentration performances.

2.5. Separation procedures

2.5.1. Uranium and thorium

The procedure used for the pre-concentration and isolation of uranium and thorium isotopes is based on a modified version of the method developed by Dalencourt et al. (2020) , using a 2 mL cartridge TRU resin (Table 3). A nominal flow rate of 4 mL/min was used throughout the whole separation process. 300 mL of degassed mineral water was acidified with 20 mL of conc. HNO_3 to obtain a final 320 mL solution at a concentration of 1 M HNO_3. The resin was first conditioned (Step 1) using 20 mL of 1 M HNO_3 before loading the entirety of the sample onto it (Step 2). The glassware and funnel used for the preparation and transfer of the sample were rinsed three times with 10 mL of 1 M HNO_3. The rinses were pooled and used as a column rinse (Step 3). Uranium and thorium were then eluted together using 30 mL of 0.1 M $(NH_4)HC_2O_4$/0.025 M HCl (pH adjusted to 2.78±0.05) (Step 4). The samples were subsequently analyzed without any modification by ICP-MS/MS after the addition of rhodium as an internal standard and using calibration curves made using the same matrix (0.1 M $(NH_4)HC_2O_4$/0.025 M HCl). The instrumental conditions used are presented in the supporting information (ESI, Table S1).

Table 3. Uranium and thorium preconcentration/separation method.

Steps	Description	Solution	pH	Volume (mL)
1	Conditioning	1 M HNO_3	N/A	20
2	Loading	1 M HNO_3	N/A	320
3	Rinse	1 M HNO_3	N/A	30
4	Stripping	0.1 M $(NH_4)HC_2O_4$/0.025 M HCl	2.78	30
5	Cleaning	0.1 M $(NH_4)HC_2O_4$/0.025 M HCl	2.78	5

2.5.2. Radium

Ra separation/preconcentration was performed using a procedure developed and validated by Dalencourt et al. (2018). Throughout the separation procedure, the flow rate was maintained at 6.5 mL/min as recommended by the authors. The method was used without

any modifications and is summarised in Table 4. A 2 mL cartridge of AG50W-X8 (Eichrom Technologies) resin was conditioned with 10 mL of 0.062 M $(NH_4)_2$EDTA at pH=4.8 (Step 1). The sample was then loaded on the resin (300 mL, 0.062 M $(NH_4)_2$EDTA, (Step 2). After the loading step, the cartridge was rinsed with 20 mL of 0.060 M CDTA/0.040 M AcOH at pH=5 (Step 3) followed by a rinse of 20 mL of 0.374 M NH_4Cl at pH=5 (Step 4) and a final rinse of 20 mL of 0.062 M $(NH_4)_2$EDTA at pH=6 (Step 5). As mineral water samples are usually rich in minerals, exhaustive rinsing of the resin was needed to remove possible interference during analysis. Ra was finally eluted with 10 mL of $(NH_4)_2$NTA at pH=10 (Step 6). The collected fraction was analyzed by ICP-MS/MS after the addition of Tl as an internal standard. Calibration curves, covering the range from 30 to 5000 mBq/L, were prepared in the same matrix as the Ra stripping fraction. The instrumental conditions used are presented in the supporting information (ESI, Table S2).

Table 4. Radium preconcentration/separation method.

Steps	Description	Solvent	pH	Volume (mL)
1	Conditioning	0.062 M $(NH_4)_2$EDTA	4.8	10
2	Sample loading	0.062 M $(NH_4)_2$EDTA	4.8	300
3	Rinse 1	0.040 M CDTA/0,060 M AcOH	5	20
4	Rinse 2	0.374 M NH_4Cl	5	20
5	Rinse 3	0.062 M $(NH_4)_2$EDTA	6	20
6	Ra stripping	0.124 M $(NH_4)_2$NTA	10	10

2.5.3. Polonium

^{210}Po preconcentration was performed using a Sr spec resin (Table 5). The procedure is based on one published by Eichrom Technologies for the determination of ^{210}Pb and ^{210}Po in water (Eichrom Techlogies, 2014). The flow rate was maintained at 2 mL/min throughout the procedure. 166 mL of degassed mineral water was acidified to 2 M HCl and spiked with ^{209}Po as a tracer to obtain an activity of 200 mBq/L. The resin was conditioned with 10 mL of 2 M HCl (Step 1) before loading the sample (Step 2). The glassware used for sample preparation and the transfer were rinsed with 10 mL of 2 M HCl thrice and pooled to rinse the columns (Step 3). Po was stripped from the resin using 30 mL of 0.05 M HCl (Step 4). Po was spontaneously deposited on to 1 mm silver discs (Alfa Aesar; Tewksbury, MA, USA) for

4h using a procedure described elsewhere (Blanchet-Chouinard and Larivière, 2021) with a home-made Teflon disc holder. The samples were counted for 24h by alpha spectrometry.

Table 5. Polonium preconcentration/separation method.

Steps	Description	Solvent	Volume (mL)
1	Conditioning	2M HCl	10
2	Sample loading	2M HCl	200
3	Rinse 1	2M HCl	20
4	Stripping	0,05M HCl	30

2.6. Committed effective dose (E)

We calculated the committed effective dose from the annual consumption of bottled water using ingestion dose coefficients recommended by the International Commission on Radiological Protection (Table 6) (Eckerman et al., 2013).

Table 6. Effective dose coefficients in nSv/mBq for internal exposure via ingestion for three targeted ages.

Radionuclide	5 years old	10 years old	Adult
^{210}Po	4.4	2.6	1.2
^{226}Ra	0.62	0.80	0.28
^{228}Ra	3.4	3.9	0.69
^{230}Th	0.31	0.24	0.21
^{232}Th	0.35	0.29	0.23
^{234}U	0.088	0.074	0.049
^{235}U	0.085	0.071	0.047
^{238}U	0.080	0.068	0.045

The committed effective dose (E, μSv/y) was calculated using the following equation:

$$E = \sum A \cdot V \cdot e_r \qquad \text{Equation 1}$$

where A represents the activity (mBq/L) for a specific radionuclide, V is the annual water intake (L) and e_r is the effective dose coefficient this radionuclide (nSv/mBq). To compare different brands, the dose evaluation was conducted under the diverse scenarios that are presented below. For all scenarios, a 5-year-old-child, a 10-year-old child, and an adult were considered.

Scenario A. In this scenario, only bottled water is consumed at a nominal volume of 2 L per day for a whole year, representing an annual intake of 730 L. This nominal volume is used in the calculation of maximum acceptable concentrations (MAC) (Health Canada, 2009).

Scenario B. In this scenario, bottled water consumption based on Canadian data published by Jones et al. (2006) were used to estimate the annual volume of water consumed per age group. We used the following average consumptions:

- a 5-year-old child consumes about 283 L.
- a 10-year-old child consumes about 338 L.
- An adult (15 years and older), consumes about 420 L.

Scenario C. In this scenario, bottled water is consumed at an annual nominal volume of 72.9 L which is the reported value per capita for the consumption of packaged water in Canada in 2022 (Ridder, 2022). The remaining average per-person water consumption (*i.e.*, 657.1 L) is tap water.

For all scenarios tested, radiological measurement data from both 2021 and 2022 were used, when available. Thus, the effective dose estimates reported are calculated as the average of both years except for Brands I and P since we were unable to collect bottles in 2021 for both brands due to supply issues.

3. Results and discussion

3.1. Naturally occurring radionuclides present in bottled water

Table 7 shows the measured activities for Po, Ra, Th, and U isotopes in available bottled water brands for the two collection periods in 2021 and 2022. The compiled results are also compared, with respect to range, average and median, with other international studies concerning the presence of naturally occurring radionuclides in bottled water (Table 8). For the 2021 sampling campaign, instrumental issues with the alpha-spectrometry instrument prevented timely analyses for ^{210}Po, so 2021 ^{210}Po results are not reported in Table 7. As

imports were affected by the COVID-19 crisis, some brands were not available or were only available in limited quantities during some collection periods (Brands H, I, and P).

Table 7. Radionuclide activity concentration (mBq/L) and standard uncertainty (k=1) of tap and local and imported spring and mineral bottled water available in Québec, Canada (n=3).

Year	Brand	[^{234}U]	[^{235}U]	[^{238}U]	[^{226}Ra]	[^{228}Ra]	[^{230}Th]	[^{232}Th]	[^{210}Po]
		mBq/L	mBq/L	mBq/L	mBq/L	mBq/L	mBq/L	mBq/L	mBq/L
2021	**A (CA), Tap**	<MDA	< MDA	0.122±0.006	19±3	N/A	< MDA	< MDA	N/A
	B (CA)	0.66±0.02	(2.2±0.3) x 10^{-2}	0.441±0.009	19.1±0.1	N/A	< MDA	< MDA	N/A
	C (CA)	6.4±0.2	(11.2±0.2) x 10^{-2}	3.06±0.06	14.3±0.7	N/A	< MDA	< MDA	N/A
	D (FR)	1.18±0.07	(3.0±0.1) x 10^{-2}	0.66±0.01	172±6	N/A	< MDA	< MDA	N/A
	E (FR)	19±2	(70±6) x 10^{-2}	14.4±0.1	39±2	N/A	< MDA	< MDA	N/A
	F (FR)	26±2	(95±9) x 10^{-2}	19±2	21.6±0.3	N/A	< MDA	< MDA	N/A
	G (FR)	115±5	(213±8) x 10^{-2}	40.9±0.1	22.3±0.2	N/A	< MDA	< MDA	N/A
	H (FR)	N/A	N/A	N/A	17±1	N/A	< MDA	< MDA	N/A
	I (FR)	N/A	N/A	N/A	N/A	N/A	< MDA	< MDA	N/A
	J (SI)	2.0±0.2	(5.0±0.3) x 10^{-2}	1.03±0.06	17.3±0.6	N/A	< MDA	< MDA	N/A
	K (GR)	7.5±0.2	(17.4±0.2) x 10^{-2}	3.84±0.09	20±5	N/A	< MDA	< MDA	N/A
	L (RO)	13.8±0.2	(40.6±0.8) x 10^{-2}	8.2±0.2	319±4	N/A	< MDA	< MDA	N/A
	M (DE)	21.3±0.5	(50±1) x 10^{-2}	10.3±0.3	24±1	N/A	< MDA	< MDA	N/A
	N (AM)	38±1	(101±2) x 10^{-2}	19.3±0.5	22.1±0.1	N/A	< MDA	< MDA	N/A

Year	Brand	[^{234}U]	[^{235}U]	[^{238}U]	[^{226}Ra]	[^{228}Ra]	[^{230}Th]	[^{232}Th]	[^{210}Po]
		mBq/L	mBq/L	mBq/L	mBq/L	mBq/L	mBq/L	mBq/L	mBq/L
	O (IT)	113±5	(379±14) x 10^{-2}	77±3	93±3	N/A	< MDA	< MDA	N/A
	P (ES)	N/A	N/A	N/A	N/A	N/A	< MDA	< MDA	N/A
2022	**A (CA), Tap**	0.15±0.01	(0.65±0.03) x 10^{-2}	0.126±0.009	0.8±0.1	< MDA	< MDA	< MDA	< MDA
	B (CA)	0.38±0.07	(2.25±0.08) x 10^{-2}	0.48±0.02	5.1±0.6	< MDA	< MDA	< MDA	< MDA
	C (CA)	5.7±0.5	(14±2) x 10^{-2}	2.8±0.2	4.2±0.3	< MDA	< MDA	< MDA	< MDA
	D (FR)	1.1±0.2	(3.8±0.6) x 10^{-2}	0.6±0.1	550±10	< MDA	< MDA	< MDA	7.4±0.6
	E (FR)	13.3±0.9	(44.8±0.6) x 10^{-2}	8.7±0.1	28±5	< MDA	< MDA	< MDA	< MDA
	F (FR)	24.6±0.2	(82.9±0.8) x 10^{-2}	15.7±0.2	7.4±0.1	< MDA	< MDA	< MDA	< MDA
	G (FR)	88±7	(201±2) x 10^{-2}	40±2	12±2	< MDA	< MDA	< MDA	< MDA
	H (FR)	45.9±0.5	(94±2) x 10^{-2}	17.9±0.1	10±2	< MDA	< MDA	< MDA	6±3
	I (FR)	28.3±0.9	(102±5) x 10^{-2}	21.5±0.2	1.1±0.7	N/A	< MDA	< MDA	6±3
	J (SI)	1.9±0.1	(8.5±0.2) x 10^{-2}	1.01±0.02	4.8±0.3	< MDA	< MDA	< MDA	< MDA
	K (GR)	11±2	(35±5) x 10^{-2}	4.0±0.5	14±1	< MDA	< MDA	< MDA	< MDA
	L (RO)	14.5±0.2	(35.7±0.6) x 10^{-2}	6.9±0.2	310±6	< MDA	< MDA	< MDA	6±3
	M (DE)	14.3±0.7	(45.9±0.8) x 10^{-2}	9.9±0.1	5.3±0.5	< MDA	< MDA	< MDA	< MDA
	N (AM)	40.9±0.7	(84±2) x 10^{-2}	15.7±0.1	7.5±0.2	< MDA	< MDA	< MDA	< MDA

Year	Brand	[^{234}U]	[^{235}U]	[^{238}U]	[^{226}Ra]	[^{228}Ra]	[^{230}Th]	[^{232}Th]	[^{210}Po]
		mBq/L	mBq/L	mBq/L	mBq/L	mBq/L	mBq/L	mBq/L	mBq/L
	O (IT)	78.3±0.5	(313±3) x 10^{-2}	58.4±0.7	80±4	< MDA	< MDA	< MDA	< MDA
	P (ES)	4.8±0.1	(29.8±0.8) x 10^{-2}	3.8±0.1	188±4	< MDA	< MDA	< MDA	26±6
2021	**MDA**	0.02	(0.02) x 10^{-2}	0.005	1	N/A	(0.5) x 10^{-3}	0,04	N/A
2022	**MDA**	0.02	(0.02) x 10^{-2}	0.005	0.3	100	(0.5) x 10^{-3}	0,04	2

MDA – Minimum detectable activity; N/A – Not analyzed; CA – Canada, FR – France, SI - Slovenia, GR-Greece, RO-Romania, DE - Germany, AM - Armenia, IT – Italy, ES - Spain.

Table 8. Activity of detected naturally occurring radionuclides in bottled water available in various countries.

Country		^{234}U (mBq/L)	^{238}U (mBq/L)	^{226}Ra (mBq/L)	^{210}Po (mBq/L)	Reference
Canada (*n*=28)	Range	0.38–115	0.48–58.4	1.1–550	6–26	This study
	Average	27±33	15±19	72±128	10±9	
	Median	14	9	20	6	
Italy (*n*=68)	Range	0.2–79	0.2–89	10–52.5	0.04–21	Desideri et al. (2007)
	Average	12±15	9±15	15±11	2±4	
	Median	N.R.	N.R.	N.R.	N.R.	
Romania (*n*=10)	Range	N.R.	40–170	29–450	N.R.	Calin et al. (2015)
	Average	N.R.	84±33	136±116	N.R.	

Country		^{234}U (mBq/L)	^{238}U (mBq/L)	^{226}Ra (mBq/L)	^{210}Po (mBq/L)	Reference
	Median	N.R.	84	110	N.R.	
Spain (*n*=59)	Range	1–173	1–80	1–194	1–49	(Díaz-Francés et al., 2013; Pérez-Moreno et al., 2020)
	Average	19±30	11±15	26±43	7±11	
	Median	9	7	7	2	
Slovenia (*n*=11)	Range	3–173	1–53	1.1–32	0.2–2.1	Benedik and Jeran (2012)
	Average	35±51	13±16	11±9	0.9±0.6	
	Median	13	5	11	0.6	
France (*n*=142)	Range	1.6–1000	1.3–230	11–450	2.1 25	(Franques et al., 2013)
	Average	49±55	20±19	105±86	9±4	
	Median	16	8.5	53	3.5	
Greece (*n*=16)[a]	Range	4.1–44.8	4.1–44.8	0.6–22.1	N.R.	Karamanis et al. (2007)
	Average	16±13	16±13	5±5	N.R.	
	Median	10	10	4	N.R.	
Germany (*n*=400)	Range	N.R.	1–1300	1–290	N.R.	Obrikat et al. (2004)
	Average	N.R.	16.5	19.6	N.R.	
	Median	N.R.	1.4	7	N.R.	
Sweden (*n*=37)	Range	0.2–1162	0.2–714	0.7–77	0.2–120	Piñero-García et al. (2022)
	Average	129±332	92±215	12±15	11±25	
	Median	3	3	6	2	

Country		^{234}U (mBq/L)	^{238}U (mBq/L)	^{226}Ra (mBq/L)	^{210}Po (mBq/L)	Reference
Hungary (*n*=18)	Range	11–92	9–98	4–2940	3–19	Kovács et al. (2004)
	Average	38±27	35±30	273±684	7±6	
	Median	27	23	47	4	
Poland (*n*=65)	Range	0.4–1.5	0.4–1.5	3–641	0.3–3	(Chmielewska et al., 2020; Skwarzec et al., 2003)
	Average	0.8±0.4	0.8±0.4	95±155	1±1	
	Median	0.8	0.8	39	0.7	
Japan (*n*=20)	Range	0.13–260	0.12–240	8.5–130	1–4.9	Kinahan et al. (2020)
	Average	15±61	14±56	94±58	2±1	
	Median	0.3	0.5	120	2	

N.R. – Not reported.

a ^{234}U and ^{238}U were reported as ^{234}U +^{238}U.

All three isotopes of uranium monitored were detected in every sample we analyzed. As concentration of uranium can be low in drinking water and that the natural isotopic abundance of ^{234}U and ^{235}U is much less than ^{238}U, these isotopes are rarely reported when dealing with U content in water measured by mass spectrometry. The fact that all three isotopes were measured above their respective quantification limits suggests that the analytical approaches selected were suitable for the monitoring of bottled drinking water. The activity concentration of ^{234}U ($t_{½}$=2.455×10^{5}y) was 0.38–115 mBq/L with a median of 14. For ^{235}U ($t_{½}$=7.04×10^{8}y) it was (2.2–379) x 10^{-2} mBq/L with a median of 0.4 mBq/L, and for ^{238}U ($t_{½}$=4.468×10^{9}y) it was 0.48–58.4 mBq/L with a median of 9 mBq/L. This range and median are comparable to those reported in other countries (Table 8). It is interesting to note that uranium activity concentrations measured in the imported brands are within the reported range presented for other national surveys.

Few values have been reported for ^{235}U in bottled water as this isotope has a very low natural abundance and a long half-life, rendering its detection challenging. The analytical procedure used in this investigation preconcentrates the sample and can achieve a low MDA which allows the proper quantification of this radioisotope. Previous studies in France have reported similar concentrations (median: 0.37 mBq/L; range: 0.056–11 mBq/L) (Franques et al., 2013). It is important to note that ^{235}U activity concentrations reported in most studies were not measured directly but calculated using the natural ratio of ^{235}U–^{238}U. In this study, none of the samples analyzed exceeded the provincial and national MAC guidelines for uranium.

As a result of the high preconcentration factor and the low MDA of the method used, ^{226}Ra ($t_{½}$=1600 y) was detected in every sample analyzed. In contrast, ^{228}Ra was not detected in any samples. This is probably because of the high MDA (100 mBq/L) achieved using our measurement method, consequence of its relatively short half-life ($t_{½}$=5.75 y), which presents a challenge when using a mass-spectrometric method. Based on the activity concentrations measured, a median ^{226}Ra activity concentration of 20 mBq/L and a range of 1.1–550 mBq/L were calculated. The activities obtained in this study are comparable to those obtained in other European studies (Table 8), although higher ^{226}Ra activity concentrations have been reported in some countries (*e.g.* Poland, Hungary). The activities reported here are similar to those reported by Zikovsky (2006) for the presence of ^{226}Ra in 216 tap samples drinking water collected between 1980 and 2004 in the province of Québec. This author reported a mean of 44±72 mBq/L (cf., 72±128 mBq/L, this study) with a median of 14 mBq/L (cf., 20 mBq/L for this study) and a maximum value of 458 mBq/L (550 mBq/L, this study). The values are also similar to historical values obtained in 21 Canadian communities between 1975 and 2016 (Chen et al., 2018). These authors reported annual averages ranging from 3.2±2.6 to 6.6±11 mBq/L for drinking water samples collected in Québec City from 1975 to 1979. Only Brand D, collected in 2022, exceeded the Québec and Canadian MAC for ^{226}Ra (500 mBq/L).

Though some samples contained relatively higher activities of ^{226}Ra (Brands D and L) than most, it was still impossible to detect ^{228}Ra above the MDA, which means this radionuclide is likely present with an activity lower than ^{226}Ra. By increasing the volume of bottled water used for the method, we could increase the preconcentration factor, which would decrease the MDA for ^{228}Ra. However, this change would have negatively impacted the extraction yield of radium onto the AG50W-X8, hence increasing the MDA. As the measurements were based on a method providing an MDA (100 mBq/L) that is lower than the MAC (200–500 mBq/L), the original protocol was deemed sufficient to determine whether ^{228}Ra exceeded those guidelines. Nevertheless, if a complete portrait of the concentration of these longer-

lived isotopes of radium in the water is required, modification to the methodology used or alternatively, the use of more conventional radiometric approaches (*e.g.* alpha-spectrometry), would be required.

While samples collected were analyzed for thorium, ^{230}Th ($t_{½}$=75,400y) and ^{232}Th ($t_{½}$=1.4×10^{10}y) were not detected in any of the samples despite the very low minimal detectable activity (MDA) achieved with the analytical procedure used (0.5 μBq/L for ^{230}Th and 40 μBq/L for ^{232}Th). This absence of Th in bottled water was expected, as Th tends to exhibit a very low solubility in aqueous media and, in its soluble form, is adsorbed onto minerals, which further reduces its presence as a dissolved ion in natural water (Porcelli and Swarzenski, 2003). Although the reported MDA could be further reduced using either higher volumes of water or detection with other instrumentation such as gamma spectrometry, the presence of Th isotopes below our MDA combined with their relatively low effective dose coefficients indicates that the effective dose resulting from Th ingestion in the bottled water samples collected would be insignificant compared to other radionuclides monitored. Moreover, the MDA obtained with the method used are orders of magnitude lower than the provincial MAC (100–400 mBq/L).

Very few samples presented detectable concentrations of Po-210 ($t_{½}$=138d) above the MDA (2 mBq/L). ^{210}Po was detected in 5 brands with activity concentrations ranging from 6 to 26 mBq/L. Subsequent measurements over time were not conducted on these samples, making it impossible to ascertain the presence of unsupported Po. Nonetheless, it is likely that the Po identified (or at least a portion) was produced *in situ* through the decay of its precursor radionuclide, ^{210}Pb. The average and median values calculated for those brands were 10±9 and 6 mBq/L, respectively. These values are consistent with those reported by other bottled drinking water studies performed in Europe (Table 8). Since the MDA for this isotope is 100 times lower than the Ontario MAC (200 mBq/L), the method used was deemed acceptable for drinking water monitoring of ^{210}Po, although activity concentrations were lower than the MDA in 66% of the samples analyzed. However, from a committed dose perspective, it could be valuable to measure ^{210}Po activities below the reported MDA since this radionuclide has the highest effective dose coefficients among the radionuclides evaluated in this study.

3.2. Comparison in radionuclides concentrations in tap water, local and imported bottled water

As stated in the introduction, while local bottled water brands are available to Canadians, most of the bottled water in Canada is imported. This is why only 2 distinct local brands

were collected as part of this study; one was labelled as spring water and one as mineral water. Tap water was also analyzed to determine its radionuclide content during the same period. For comparison purposes, only the results obtained for ^{226}Ra and ^{238}U were compared since complete data sets were available for these (*i.e.* 100% of the samples analyzed were above MDA). Since uranium measured in the collected samples exhibits isotopic ratio near the expected natural abundance (see section on isotopic variability for more details), only results for ^{238}U are presented. The average ^{226}Ra activity concentration measured in tap water (10±13 mBq/L) is of the same order of magnitude as the concentrations reported by Zikovsky in 1980–2004 (44±72 mBq/L) and Chen et al. (1.8±0 to 6.6±11 mBq/L) in 1975–1979, in tap water collected in the province of Québec (Chen et al., 2018; Zikovsky, 2006). Due to the limited number of brands (local and imported) analyzed, it is impossible to draw any robust statistical conclusions regarding the difference in activity concentrations for ^{226}Ra and ^{238}U between these sources.

Results presented in Fig. 1 show that tap and local water brands tend to contain lower activities radionuclide concentrations than imported brands. This was especially true for uranium. The calculated mean activity concentration of ^{238}U in tap water (0.124±0.002 mBq/L) and local brands of bottled water (1.7±1.4 mBq/L) was approximately 10-fold lower than in the imported brands (17±20 mBq/L). Mean ^{226}Ra concentrations in tap water (10±13 mBq/L) and local brands of bottled water (11±7 mBq/L) were also lower than in imported brands (83±135 mBq/L), by an order of magnitude. However, to statistically confirm that tap water and local bottled brands contain lower radionuclide concentrations than imported brands, a much larger data set is required.

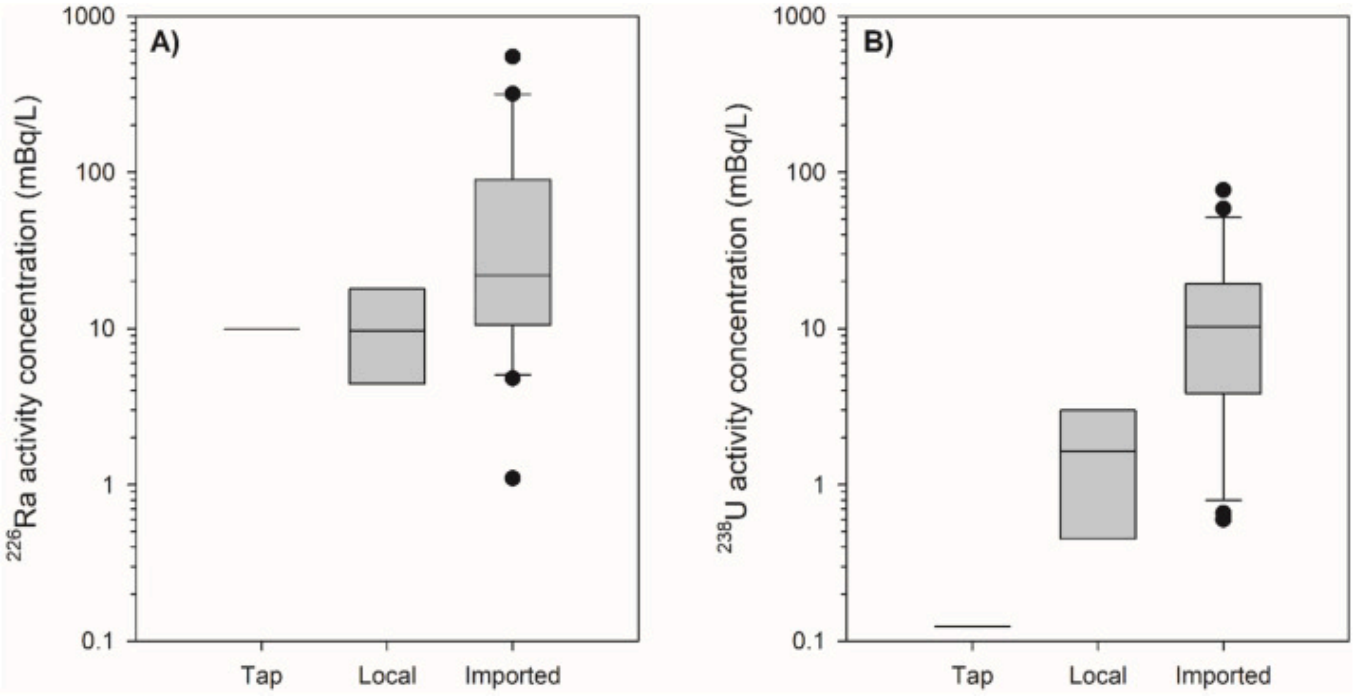

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Fig. 1. Box and whisker plot of the activity concentrations (in mBq/L) in tap (n=2), local bottled (n=4) and imported bottled (n=24) water brands for a) ^{226}Ra and b) ^{238}U.

3.3. Temporal variation in uranium and radium activity concentration

The chemical and radiological composition of water is greatly influenced by meteorological, (hydro)geological and (bio)geochemical parameters (Faraj et al., 2020). Thus, the study of radiological parameters at a specific moment in time might not be representative of the activity concentration of the radionuclide of interest throughout the entire use of a water source. To determine the degree of variability in radioactivity, the same bottled water brands were collected in 2021 and 2022 and analyzed to determine their radionuclide content. As the degree of analytical sensitivity was only sufficient to properly quantify (above the MDA) ^{226}Ra and ^{238}U, in all samples, these two isotopes were used to assess the variation in activity concentration between the 2021 and 2022 sampling period. Note that the values presented for a specific year represent the average of three replicates from the same brand lot. Fig. 2 presents the variation (2021/2022) in activity concentrations for each brand analyzed. Note that due to challenges in obtaining specific brands in 2021, Brands I and P were not measured during this sampling period. For Brand H, due to the high volume of water required for ^{226}Ra analysis, other radiological assessments were not performed. While not included in the figure, tap water exhibited the highest variation in ^{226}Ra

measured during this study (24±7) but very little variation in ^{238}U activity concentrations (0.96±0.10).

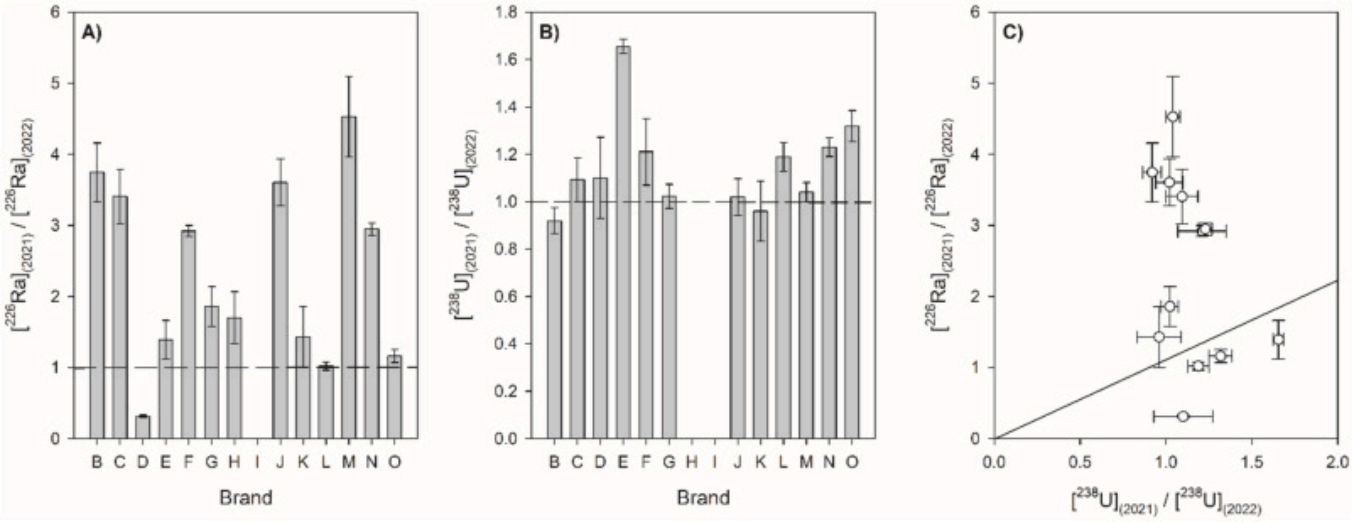

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Fig. 2. Temporal variability in the activity concentration ($[X]_{2021}/[X]_{2022}$) of A) ^{226}Ra and B) ^{238}U in the various bottled water brands investigated. C) Plot of the temporal variability for ^{226}Ra and ^{238}U for each specific brand. The dashed line in A) and B) represents the absence of variability between the two years sampled whereas the solid line in C) represents a similar change in variability between the temporal variability for ^{226}Ra and ^{238}U.

Comparing the two years of sampling, a large variability range can be observed (0.3–4.5, Fig. 2A) for ^{226}Ra activities, but not for ^{238}U (0.9–1.6, Fig. 2B). As bottled water quality can be impacted by many parameters, it is impossible to attribute the variations to a single factor. As the water collection process, including the bottling date and specific locations are not readily available to the public, it is challenging to link variations to either natural or anthropogenic events which may affect the aquifer or to changes in soil geochemistry that could impact the presence of radioactivity in the water. Even though mineral contents of bottled water remain constant, based on the information provided on the label and as required by legislation, fluctuation in underground temperature, pH, water composition and many other factors can result in variations in Ra activity (Porcelli and Swarzenski, 2003). Based on Fig. 2C, the factors influencing the presence of ^{226}Ra in the bottled water are not those favoring the presence of ^{238}U. This suggests that monitoring of individual radionuclides (and their progenies) relevant to drinking water, not just U, should be undertaken.

Studies have revealed that the movement of radium is more pronounced in thermal water, which is characterized by a substantial mineral content, leading to an elevated concentration of ^{226}Ra (Guerrero et al., 2016). Comparing the activity of ^{226}Ra with reported mineral content, expressed as the total dissolved solid (TDS, in mg/L), showed that TDS was a poor indicator of the ^{226}Ra content. An investigation of the correlation between the water composition reported on the label (Table 1) and the ^{226}Ra and ^{238}U activity concentrations (Table 7) did not reveal any statistically significant correlations between the cation, anion, and TDS values reported (all $r^2 \leq 0.51$ for ^{226}Ra and ≤ 0.23 for ^{238}U).

3.4. Isotopic variability with respect to secular equilibrium

Most of the natural radionuclides present in natural water originate from either from ^{235,238}U or ^{232}Thday series apart from ^{40}K, ^{3}H and ^{14}C. Thus, in a closed and undisturbed system, a secular equilibrium could be expected from U and Th progenies. With the analytical sensitivity of the methodologies selected for monitoring radionuclides in bottled water, it is possible to determine if this equilibrium is preserved or altered. Table 9 presents the ratios of activities in bottled water collected in 2022 for ^{234}U/^{238}U, ^{226}Ra/^{234}U, and ^{210}Po/^{226}Ra.

Table 9. Ratios of activities concentrations in bottled water sampled in 2022.

Brand	^{234}U/^{238}U			^{226}Ra/^{234}U			^{210}Po/^{226}Ra		
B (CA)	0.79	±	0.19	13.42	±	4.97		N.A.	
C (CA)	2.04	±	0.35	0.74	±	0.13		N.A.	
D (FR)	1.83	±	0.77	500	±	122	0.013	±	0.001
E (FR)	1.53	±	0.12	2.1	±	0.6		N.A.	
F (FR)	1.57	±	0.03	0.30	±	0.01		N.A.	
G (FR)	2.20	±	0.30	0.14	±	0.04		N.A.	
H (FR)	2.56	±	0.04	0.22	±	0.05	0.6	±	0.5
I (FR)	1.32	±	0.05	0.04	±	0.03	5	±	17
J (SI)	1.88	±	0.14	2.5	±	0.3		N.A.	
K (GR)	2.75	±	0.96	1.27	±	0.39		N.A.	
L (RO)	2.10	±	0.09	21.4	±	0.97	0.011	±	0.006

Brand	$^{234}U/^{238}U$			$^{226}Ra/^{234}U$			$^{210}Po/^{226}Ra$		
M (DE)	1.44	±	0.09	0.37	±	0.06		N.A.	
N (AM)	2.61	±	0.06	0.18	±	0.01		N.A.	
O (IT)	1.34	±	0.02	1.02	±	0.06		N.A.	
P (ES)	1.26	±	0.06	39.2	±	1.68	0.14	±	0.04
Mean	1.8	±	0.6	39	±	128	1	±	2
Median		1.9			1.02			0.14	

N.A. – Not applicable.

The $^{234}U/^{238}U$ ratios calculated ranged from 0.79 to 2.75 with a mean of 1.8±0.6 and a median of 1.9. Those values are very close to those published by Piñero-García et al. (2022) who found a mean of 1.8±1.1 and a median of 1.6 in bottled water available to Swedish population. The breaks in the secular equilibrium could be attributed to several parameters, with decay recoil and preferential leaching of ^{234}U being the most frequently documented (Rasilainen et al., 2006; Dinh Chau et al., 2011). With the exception of Brand B, all bottled water samples tested had a$^{234}U/^{238}U$ ratio higher than 1 (even when the uncertainties were accounted for), demonstrating a disequilibrium between ^{234}U and ^{238}U.
Goldstein et al. (1997) showed that soil composition greatly impacts the $^{234}U/^{238}U$ ratio and that water containing trace levels of uranium tend to show an activity ratio closer to 2, and water with higher uranium content (≈1000 mBq/L) shows a ratio closer to 1. Since the bottled waters monitored had total uranium activity concentrations ranging from 1.7 to 136.7 mBq/L, mean and median $^{234}U/^{238}U$ values approaching 2 were expected.

The $^{226}Ra/^{234}U$ ratio is highly variable, with an average of 39±128 and a median value of 1.02. When Brands D, L, and P, which have the highest ^{226}Ra activity, were removed from the average and median calculation, the new ratio was 1.9±3.72 with a median of 0.55. This new mean ratio is similar to the ratio observed for $^{234}U/^{238}U$.

Most of the samples tested exhibited a $^{210}Po/^{226}Ra$ ratio of less than 1. Vasile et al. (2016) also found in their monitoring of European drinking water that the $^{210}Po/^{226}Ra$ ratio was typically less than 1. In a study of groundwater in India, Molla et al. (2021) reported $^{210}Po/^{226}Ra$ ratio ranging from 0.2 to 11, a range that is close to the range reported here (0.01–5).

For comparison purposes, activity concentration ratios for municipal tap water were calculated as 1.19±0.18 for $^{234}U/^{238}U$ and 5.33±1.10 for $^{226}Ra/^{234}U$. ^{210}Po was below the MDA in tap water (Brand A) in 2022.

3.5. Dose estimate from ingestion of bottled water

As stated in the Method and Materials section, several scenarios were used to calculate the effective dose received by Québec City residents who drink tap and bottled water. Fig. 3 shows the calculated committed doses as a function of age group, resulting from the intake of the quantified radionuclides by bottled drinking water available in Québec, Canada, assuming a consumption of 2 L of water per day per individual (Scenario A). Under this scenario, average committed doses were calculated as 47±66 μSv/y for 5-year-olds, 51±76 μSv/y for 10-year-olds, and 20±27 μSv/y for adults. For the same scenario, median committed doses were 17, 15, and 7 μSv/y for 5-year-olds, 10-year-olds, and adults, respectively.

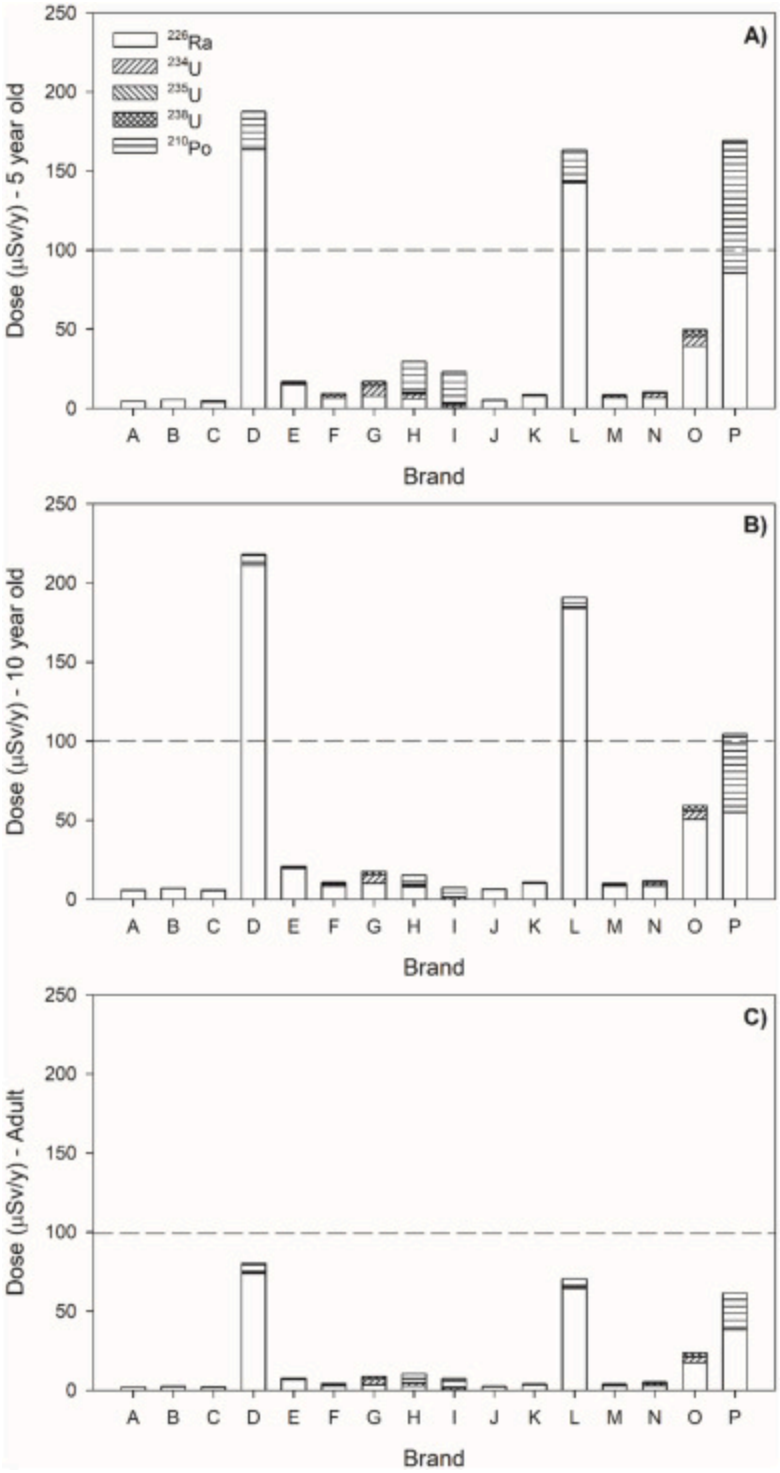

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Fig. 3. Dose assessment (µSv/y) from the intake of monitored naturally occurring-radionuclides resulting from drinking daily 2L of bottled water for a A) 5-year-old, B) 10-year-old, and C) adult.

In comparison, the consumption of 2L of Québec City tap water (labelled as Brand A) resulted in average committed doses of radiation that were about one tenth the values

calculated for bottled water: 4±5, 6±8, and 2±3 μSv/y for 5-year-olds, 10-year-olds, and adults, respectively. For younger individuals, the exclusive consumption of bottled water at a nominal volume of 2 L/day would results in a committed dose exceeding the threshold value for drinking water of 100 μSv/y for Brands D, L and P. Fig. 3 shows that for the radionuclides detected, ^{226}Ra and ^{210}Po were the main contributors to the annual committed dose from the intake of bottled water.

Jones et al. (2006) have surveyed the Canadian population to determine the actual water intake and they reported values of 0.775, 0.925, and 1.15 L/d for a 5-year-old, a 10-year-old, and an adult, respectively. Using these values, the committed doses were recalculated and are presented in Fig. 4. In Scenario B, accounting for the lower water consumption at younger ages, only the consumption of Brand D by a 10-year-old individual would lead to a committed dose above the 100 μSv/y threshold. Although younger individuals consume less drinking water, their committed effective dose results systematically in higher doses compared to adults due to the higher effective dose coefficients used in the calculation.

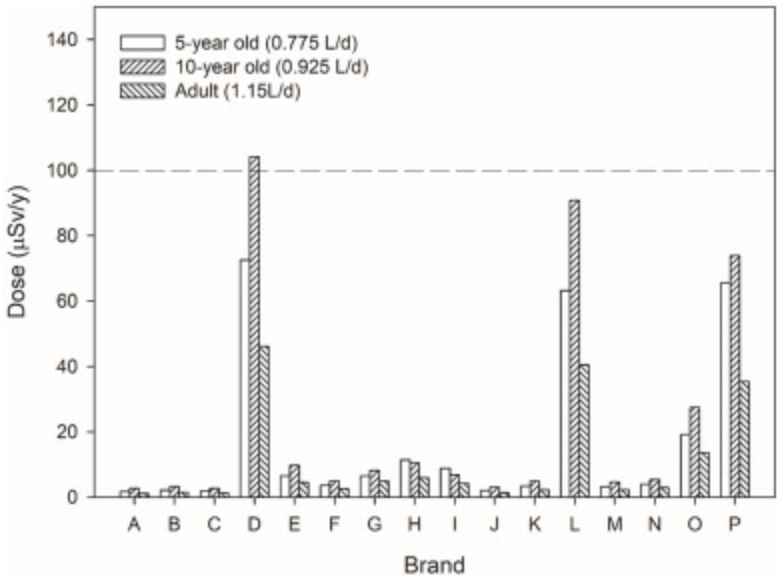

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Fig. 4. Dose assessment (μSv/y) from the intake of monitored naturally occurring-radionuclides based on reported water consumption by Jones et al. (2006) of water for a A) 5-year-old, B) 10-year-old, and C) adult.

While the water consumption data published by Jones et al. (2006) provides a more accurate portrait of Canadian drinking habits, it is very uncommon for individuals to consume exclusively bottled drinking water. Therefore, based on the reported value per

capita for the annual consumption of packaged water (72.9L) in Canada in 2022 (Ridder, 2022) with reference to the Canadian drinking water consumption, we estimated that bottled water consumption would represent approximately 26%, 22%, and 18% for 5y, 10y, and adult, respectively of the whole drinking water consumption. Based on Scenario C, committed dose were calculated and are presented in Fig. 5. For this scenario where tap water and bottled water are ingested, the drinking water threshold was never exceeded, due to the low concentrations of radionuclides measured in the municipal tap water (labelled as Brand A in this study). The highest committed dose calculated (24.6 μSv/y) would be for a 10-year-old Canadian consuming 72.9L of Brand D bottled water and 265L of municipal tap water from Québec City.

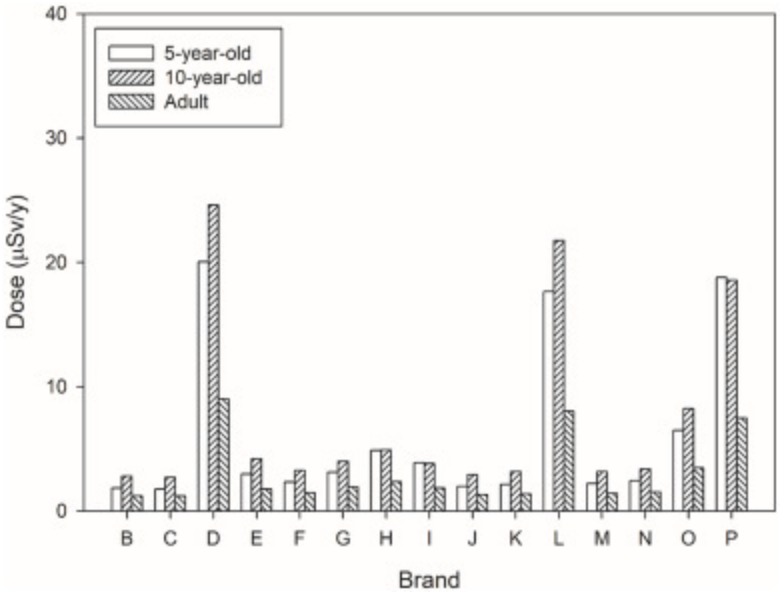

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Fig. 5. Dose assessment (μSv/y) from the intake of monitored naturally occurring-radionuclides based on the consumption of 657.1L of tap water and 72.9L of a specific bottled water brand for a A) 5-year-old, B) 10-year-old, and C) adult.

4. Conclusion

In this study, naturally occurring radionuclides (^{210}Po, ^{226}Ra, ^{228}Ra ^{230}Th, ^{232}Th, ^{234}U, ^{235}U and ^{238}U) were measured in 15 bottled water brands available in Québec City, as well as Québec City tap water. Analyses were done by sampling the bottled water for two consecutive years. The analyses revealed variations in U and Ra activities, demonstrating the need to routinely monitor each radionuclide. Irregularities in isotopic ratios were observed

in most samples especially with $^{226}Ra/^{234}U$. The activities measured were used for dose assessment of naturally occurring radionuclides in bottled water for different age groups based on recommended and reported water consumptions. In most samples, ^{226}Ra was the major contributor to dose and calculated committed doses were systematically higher for younger individuals. This assessment demonstrates that younger individuals could receive doses higher than the threshold value for drinking water of 100 μSv/y if brands containing the highest concentrations of ^{226}Ra are consumed as a primary source of water intake.

CRediT authorship contribution statement

Jean-Christophe Tremblay-Cantin: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Conceptualization. **Laurie Martin:** Writing – original draft, Validation, Methodology, Investigation. **Myriame Proulx:** Methodology, Investigation. **Nicholas D. Priest:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization. **Dominic Larivière:** Writing – review & editing, Writing – original draft, Visualization, Validation, Project administration, Funding acquisition, Formal analysis, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Dominic Lariviere reports financial support was provided by CANDU Owners Group Inc. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

The following is the Supplementary data to this article:

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Multimedia component 1.

Recommended articles

Data availability

Data will be made available on request.

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