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Radiological impact of naturally occurring radionuclides in bottled water

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ABSTRACT

Consumption of bottled water is increasing year after year in Europe. Due to the local geology from where the water is extracted; bottled water could be enhanced with radionuclides. This study focuses on the activity concentrations of ^{210}Po, ^{210}Pb, ^{226}Ra, ^{228}Ra, ^{234}U and ^{238}U in bottled water available in the Swedish market, to assess the radiological impact to different age groups. The results showed that among the 26 brands studied, only three could exceed the threshold value for drinking water: 0.1 mSv/year. For two brands, the dose was mainly due to the activity concentrations of ^{238}U and ^{234}U being up to 714 and 1162 mBq/L, respectively. While for one brand, the dose was mainly due to the activity concentration of both ^{210}Po and ^{210}Pb being around 100 mBq/L. For the remainder brands, ^{228}Ra was the main contributor to the committed effective dose.

1. Introduction

According to international public health organisations, it is recommended to complete the daily liquid intake by food with at least 1.5 L or 2 L of drinking water to prevent health problems (European Commission, 2020). For that reason, drinking water must be safe and of high quality. However, the transfer of heavy metals and radionuclides from the bedrock and soils to the aquifer and drinking water reservoirs can influence the quality of drinking water, especially in areas with moderate and high natural radioactivity content in the bedrock, where the relevant radionuclides are ^{210}Po, ^{210}Pb, ^{226}Ra, ^{228}Ra, ^{234}U and ^{238}U, along with their short-lived progenies.

Consumption of bottled water is increasing in Europe year after year (Tosun et al., 2020). In Sweden, the consumption of bottled water during 2019 was around 10^8 L, where 77% was natural mineral water and 23% was spring water, with a higher preference of sparkling (74%) than still water (26%), according to the statistics of the European Federation of Bottled Water (EFBW, 2021). The highest consumption of bottled water occurred in the largest cities (Stockholm, Gothenburg, and Malmö), and it was related to the availability and differences in lifestyle, original nationality and thus population habits (Westrell et al., 2006).

Three well-defined types of bottled water are available on the European Market:

1. **Natural Mineral Water (NMW)** comes from protected underground water, characterized by the original purity and constant mineral composition. This water needs official recognition from the State's competent national authorities and registration in the Official Journal of the European Union. The water must be bottled near the source, without any modification or water processing, except for a few treatments specified by DIRECTIVE 2009/54/EC and prior authorization by the competent national authorities (European Parliament Council of the European Union, 2009). In Sweden, there are ten officially recognized brands of natural mineral water (European Commission, 2021).
2. **Spring Water (SpW)** comes from a specific and recognized natural spring that is wholesome and clean for human consumption in their natural state. This water must be bottled at the source and it must not be treated or modified with the exception of a few authorised processes. A stable mineral composition or official recognition is not required, however, it must comply with the national and EU regulations to ensure the quality of water intended for human consumption.
3. **Other Waters in Bottles (OBW)** comes from several sources like surface water, groundwater as well as municipal water supply. This water can be treated or modified to comply with national and EU drinking water regulations.

During the last decades, concern regarding the impact of naturally

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occurring radionuclides on human health has increased and as a result, the European directive 2013/51/Euratom of 22nd October included reference values as derived concentrations for naturally occurring radionuclides in water intended for human consumption (Council of the European Union, 2013). Derived concentrations are calculated for a dose of 0.1 mSv per year and considering an annual intake of water of 730 L per person (Council of the European Union, 2013).

Some studies investigating the radioactivity levels in bottled water have reported wide ranges in activity concentration of naturally occurring radionuclides (Benedik & Jeran, 2012; Calin et al., 2015; Chmielewska et al., 2020; Desideri et al., 2007; Kasic et al., 2015; Kovacs et al., 2004; Kralik et al., 2003; Perez-Moreno et al., 2020; Skwarzec et al., 2003). For instance, bottled water in Spain could contribute to the committed effective dose with up to 160 μSv/year to infants and 44 μSv/year to adults (Diaz-Frances et al., 2013). However, to our knowledge, the radionuclide concentrations in bottled water available on the Swedish market have not previously been studied.

Therefore, the aim of this study is to carry out a radiometric characterization of bottled water available on the Swedish market in order to assess the radiological impact of naturally occurring radionuclides with high radiotoxicity (^{210}Po, ^{210}Pb, ^{226}Ra, ^{228}Ra, ^{234}U and ^{238}U) on different age groups.

2. Materials and methods

2.1. Sampling

Thirty-seven bottled water samples, 23 sparkling (Sp) and 14 still water (St) of 26 different brands (11 NMW, 5 SpW and 10 OBW) available in Sweden, were purchased in local supermarkets from Gothenburg (Sweden) and catering web shops during the first quarter of 2020. The sampling covered the majority of brands available in the Swedish market whose origin was as follows: Bosnia and Herzegovina (1 brand); Croatia (1 brand); France (1 brand); Germany (1 brand); Denmark (3 brands), Norway (3 brands) and Sweden (13 brands). The bottled water was acquired in different bottle sizes: 33 cL, 50 cL, 100 cL, 150 cL and 200 cL. A representative number of bottles, for each sample, were collected to have a minimum water volume of 5 L. The samples were labelled using an Identification Code (ID) consisting of the country origin abbreviation: Bosnia and Herzegovina (BA); Germany (DE); Denmark (DK); France (FR); Croatia (HR); Norway (NO) and Sweden (SE), a number between 1 and 26 to identify each brand purchased followed by the type of the sample: St for still water and Sp for sparkling water.

2.2. Radiochemistry procedures

Analytical grade reagents were used to isolate the naturally occurring radionuclides analysed. Nitric Acid (HNO_3, 65%, Fisher Scientific™; UK) and Hydrochloric Acid (HCl, 37% Fisher Scientific™; UK) were diluted with distilled water to prepare acid solutions. Samples were homogenized and acidified to pH 2 with Nitric Acid (65%, Fisher Scientific™; UK) to prevent formation of aggregates and fixation to the bottle walls prior to the radiochemical procedure. Certified standard solution: ^{209}Po (Eckert & Ziegler® Isotope Products, USA, reference number 1895-42); ^{229}Th/^{225}Ra (National Physical Laboratory, UK, reference number R26-01-2013120068-1) and ^{232}U (National Physical Laboratory, UK, reference number R20-15-2014040121-1), were used to prepare the radioactive tracers in order to assess the chemical yield and to calculate the activity concentration of naturally occurring radionuclides (Mantero et al., 2019). After adding the tracer, samples were vigorously stirred with a magnet bar for about 2 h, to ensure the chemical equilibrium and to favour the removal of the CO_2 from the sparkling water in order to avoid any interference during the radiochemistry analysis.

From each sample, three different aliquots of 1.5 L were prepared to determine the naturally occurring radionuclides.

1. The first aliquot was used to determine ^{210}Po, no later than 7 days after the sampling, avoiding further in-growth and decay of ^{210}Po that would not reflect the real ingested amount, at sampling date. A known amount of ^{209}Po (ca. 50 mBq) was added as internal standard. Po was concentrated by co-precipitation with iron (III) hydroxide ($Fe(OH)_3$) (Milena-Perez et al., 2018; Thomas, 2020). The Fe^{3+} carrier solution was prepared by Iron (III) Chloride Hexahydrate (Fisher Scientific™; UK). 30 mg of Fe^{3+} carrier were added to the aliquot, the hydroxide precipitation was obtained by adjusting the pH between 8 and 9 using Ammonia Solution (25%, Fisher Scientific™; UK). After precipitation, the sample was stirred during 30 min to allow for the co-precipitation of ^{210}Po. Then, the precipitate was allowed to settle overnight. The supernatant was discarded and the precipitate was collected by centrifugation. Finally, the precipitate was dissolved in HCl (1.5M) together with 0.2 g of L-Ascorbic acid (Sigma-Aldrich, USA) to carry out the self-deposition of polonium on copper discs at 80 °C during 5h. Copper discs were measured by alpha-particle spectrometry to determine the activity concentration of ^{210}Po.
2. In the second aliquot, activity concentration of radium and uranium radionuclides were determined using known amounts of ^{229}Th (^{225}Ra) (ca. 100 mBq) and ^{232}U (ca. 50 mBq) as internal standards to assess the chemical yield and to calculate the activity concentration of these radionuclides (Perez-Moreno et al., 2020). The samples were concentrated by MnO_2 co-precipitation (Perez-Moreno et al., 2019) using Potassium permanganate ($KMnO_4$; Acros Organics, USA) and Manganese (II) Chloride 4-hydrate ($MnCl_2$; ITW reagents, USA). For that purpose, 2 mL of $KMnO_4$ (0.2 M) and 2.5 mL of 0.3 $MnCl_2$ (0.3 M) were added. After homogenization, MnO_2 precipitation was achieved increasing the pH up to 8–9 with Ammonia Solution (25%, Fisher Scientific™, UK). The aliquot was stirred during 1 h to favour the homogenization and the co-precipitation of Ra and U. Then, the precipitate was allowed to settle overnight. The supernatant was discarded and the precipitate was collected by centrifugation and then dissolved with HNO_3 (3M) and some drops of Hydrogen Peroxide (H_2O_2, 35%, Chem Lab nv, Belgium). Uranium was extracted by UTEVA resin (100–150 μm particle size, Triskem International, France) in HNO_3 (3M) according to the analytical procedure for the determination of uranium and thorium in water of Eichrom Technologies (Eichrom, 2014b), while Ra passed through the resin. To isolate Ra from all interfering elements, AG Cation-Exchange Resin Hydrogen Form (200–400 mesh; AG50Wx8, Eichrom, USA) was used following the final steps of the analytical procedure for radium determination by alpha-particle spectrometry described by Pérez-Moreno et al. (2019). Finally, radium and uranium sources were prepared by cerium fluoride micro-precipitation (Eichrom, 2014a) and barium sulphate ($BaSO_4$) micro-precipitation (Perez-Moreno et al., 2019), respectively.
3. In the third aliquot, a second determination of ^{210}Po was carried out in order to determine ^{210}Pb by alpha-particle spectrometry, using two independent aliquots (Garcia-Orellana & Garcia-Leon, 2002). The second determination was done after waiting at least six months to ensure approximately 60% of the ingrowth of ^{210}Po from ^{210}Pb. ^{210}Po activity concentration was measured in the third aliquot, having this way ^{210}Pb activity concentration.

2.3. Alpha-particle spectrometry

^{210}Po, ^{210}Pb, radium and uranium radionuclides activity concentration were measured by ULTRA Ion-Implanted-Silicon Charged-Particle detectors (AMETEK ORTEC, USA), with a resolution of 20 keV for 241Am at 5.486 MeV and 450 mm^2 surface in an Alpha Ensemble® system (AMETEK ORTEC, USA). Further details of the detector system could be found in Mantero et al. (2019). Samples were measured from 1

Table 1
Analytical Quality Control results.

Sample	Matrix	Radionuclide	Target Value	Reported Value	Z-score
IAEA-TEL-2020-03	Water	^{210}Po	97.4 ± 5	95.5 ± 11.1	0.03
		^{226}Ra	13.5 ± 0.8	14.5 ± 2.2	0.29
IAEA-TEL-2019-03	Water	^{226}Ra	7.5 ± 0.3	7.9 ± 0.5	0.14
		^{228}Ra	22.1 ± 1.0	21.15 ± 1.5	0.75
IAEA-TEL-2018-03	Water	^{210}Pb (^{210}Po)	95.6 ± 0.9	86.1 ± 5.8	1.08
CSN-CIEMAT-2010	Food ashes	^{238}U	27.3 ± 4.9	22.8 ± 2.2	0.92
		^{234}U	28.5 ± 5.1	28.9 ± 3.9	0.08

Table 2
Effective dose coefficients in μSv/Bq for internal exposure via ingestion, food or water, for specific age group: infants (1–2 y), children (7–12 y) and adults (>17 y) (ICRP, 2012).

	Infants	Children	Adults
^{210}Po	8.8	2.6	1.2
^{210}Pb	3.6	1.9	0.69
^{226}Ra	0.96	0.8	0.28
^{228}Ra	5.7	3.9	0.69
^{238}U	0.12	0.068	0.045
^{234}U	0.13	0.074	0.049

to 3 days and the alpha spectra were analysed by GammaVision for Windows version 8 (AMETEK ORTEC, USA).

The activity concentration of ^{210}Po and ^{210}Pb at sampling date was calculated based on the measurements of ^{210}Po at two different times and solving Bateman's equations, as proposed by Garcia-Orellana and Garcia-Leon (2002). In addition, radium samples were also measured at one month and six months after preparation by alpha particle spectrometry in order to determine the activity concentration of ^{226}Ra and ^{228}Ra according to calculations given by Pérez-Moreno et al. (2019). Activity concentrations of ^{238}U and ^{234}U were calculated by alpha-particle spectrometry, according to the equations described by Thomas (2020). In addition, the mass concentration of uranium (μg/L) was calculated considering the radioactive decay to transform the activity concentration of uranium (mBq/L) to mass concentration (μg/L), using a conversion factor of 0.0803 (Milena-Perez et al., 2021). The radiochemistry procedures applied in this study were validated by participating in proficiency test exercises and obtaining satisfactory results (Mantero et al., 2019). Table 1 summarizes some results obtained during the validation and analytical quality control of the radiometric determination. Minimum Detectable Activity (MDA) was calculated according to the ISO 11929-3:2019. MDA was approximately 0.2 mBq/L for ^{210}Po, ^{210}Pb and ^{238,234}U and 0.7 mBq/L for ^{226,228}Ra, based on the average values of measuring times as well as chemical yields.

2.4. Committed effective dose

Assessment of the committed effective dose from one year's consumption of bottled water was calculated using ingestion dose coefficients from publication 119 of International Commission on Radiological Protection (ICRP, 2012), presented in Table 2, according to the following equation:

$$E\ (\mu Sv\,/\,y) = \sum a_i \cdot V \cdot e_i$$

$$\text{where } \begin{cases} a_i \text{ is the activity concentration for } i:th \text{ radionuclide } [Bq/L] \\ V \text{ the annual water intake per age group } [L] \\ e_i \text{ is the effective dose coefficient for } i:th \text{ radionuclide } [\mu Sv/Bq] \end{cases}$$

For comparison between brands, the dose assessment was performed assuming that all the consumed drinking water during one year is from the same brand. The annual water consumption used was 150 L for infants; 350 L for children and 730 L for adults. These values were chosen from the food habits surveys carried out by Swedish National Food Agency, according to the 95 percentile of drinking water consumption per age group in Sweden (Livsmedelsverket, 2018).

2.5. Statistical analyses

The assessment of uncertainties for the radiometric measurements were calculated according to error propagation theory and considering a confidence level of approximately 68% (k = 1). The average values are shown together with the standard deviation. Box and whisker diagrams were plotted in Excel using the inclusive median quartile calculation method, outlier are not shown in the diagram in order to make easier the comparison between the different types of bottled water. According to Box and whisker diagram options in Excel, it is considered outlier points that lie either below the lower whisker line or above the upper whisker line. This diagram is valuable to show the dataset distribution based on the median, quartiles and range. SPSS Statistics version 25 was used to apply a Mann–Whitney *U* test to compare the median values of each radionuclide from the different types of water to analyze if there were significant differences between them.

3. Results and discussions

3.1. Naturally occurring radionuclides in bottled water

Table 3 shows the activity concentrations and ratios of the measured radionuclides in bottled water together with the mass concentration of uranium and mineral composition labelled in the bottles. Table 4 shows the range, average and the median values in bottled water in Sweden as well as the results from other studies carried out in Europe.

Low levels of ^{210}Po were detected in bottled water, with an average value and median value of 11 ± 25 mBq/L and 2 mBq/L, respectively. These results were comparable to the ^{210}Po levels reported in literature (see Table 4). ^{210}Po concentration usually ranges from 1 to 30 mBq/L in groundwater and drinking water, though, in some areas with high background levels of naturally occurring radionuclides, like Finland, ^{210}Po levels up to 7600 mBq/L have been reported (IAEA, 2017). In three brands (SE-15-Sp, SE-17-Sp and SE-22) the activity concentration of ^{210}Po was higher than 30 mBq/L, the maximum activity concentration was measured in sample SE-22-Sp (120 ± 9 mBq/L), which is higher than the derived concentration for ^{210}Po stated by the European directive 2013/51/Euratom (100 mBq/L). ^{210}Pb activity concentration was higher than 0.2 mBq/L in 89% of the bottled water analysed, with an average activity concentration of 19 ± 30 mBq/L and a median value of 7 mBq/L. The results were similar to the values reported in Spain and slightly higher than the concentrations detected in Austria and Slovenia (see Table 4).

The activity concentration ratio of ^{210}Po/^{210}Pb ranged from 0.1 to 3.3 with an average value of 0.5 ± 0.7. A general tendency was that the activity concentration of ^{210}Pb was higher than ^{210}Po levels, except for samples SE-19-St and SE-21-St, indicating that the ^{210}Po in the bottled water was due to the ingrowth from ^{210}Pb. In addition, the results pointed out a disequilibrium between ^{210}Po and ^{210}Pb, except for sample SE-22-Sp, which could be considered in secular equilibrium. The disequilibrium between ^{210}Po and ^{210}Pb is controlled by many factors like the low solubility of Po and its short half-life (138.376 days) as well as local processes in the water source such as strong association with suspended particles, biological uptake, and the influence of geochemical properties (IAEA, 2017; Zhong et al., 2020). On the other hand, the achievement of the secular equilibrium will be influenced by the time elapsed between bottling time and time of measurement. However, a correct decay assessment to evaluate the source was not possible since the bottling date is unknown.

To better understand the high levels of ^{210}Pb in SE-22, it is necessary to analyze the lithology of the bedrock, the mineral composition of the aquifer together with the geochemistry of ^{210}Pb. SE-22 is a NMW whose

Table 3
Sample description: Identification code (ID) consisting of country code*, brand identification number (1–26) and sample type (St: still and Sp: sparkling) as well as water group (Natural mineral water (NMW); spring water (SpW) and Other Waters in Bottles (OBW). Activity concentration of measured naturally occurring radionuclides detected in bottled water, activity concentrations ratios and uranium mass concentration together with associated uncertainties (k = 1). Mineral composition labelled in the bottles.

ID	*Water Group*	^{210}Po (mBq/L)	^{210}Pb (mBq/L)	^{226}Ra (mBq/L)	^{228}Ra (mBq/L)	^{238}U (mBq/L)	^{234}U (mBq/L)	^{210}Po/^{210}Pb	^{234}U/^{238}U	U (μg/L)	*Ca^{2+} (ppm)*	*Na^{+} (ppm)*	*K^{+} (ppm)*	*Mg^{+} (ppm)*	*HCO_3 (ppm)*	*SO_4^{2-} (ppm)*	*Cl^{-} (ppm)*	*F^{-} (ppm)*
BA-01-Sp	NMW	<0.2	0.7 ± 0.1	7.9 ± 4.6	14 ± 8	9.1 ± 1.7	47 ± 2		5 ± 1	0.7 ± 0.1	*248.5*	*598*	*16.8*	*41.3*	*1806*	*490*	*93.7*	
DE-02-Sp	NMW	0.32 ± 0.04	3.5 ± 0.5	<0.7	6 ± 2	<0.2	0.3 ± 0.1	0.09 ± 0.02			*25*	*12.8*		*3*		*45.9*	*30.8*	
DE-02-St		<0.2	<0.2	<0.7	3.2 ± 0.9	<0.2	0.2 ± 0.1				*29.2*	*14.9*	*1.5*	*3.1*	*32.1*	*46.5*	*34.7*	
DK-03-St	NMW	0.62 ± 0.06	11 ± 2	0.7 ± 0.4	1.9 ± 1.2	<0.2	0.6 ± 0.1	0.06 ± 0.01			*36*	*10*		*3*	*120*	*3*		
DK-04-Sp	OBW	0.28 ± 0.04	<0.2	10 ± 5	6.5 ± 3.8	1.4 ± 0.1	2.7 ± 0.2		1.9 ± 0.2	0.1 ± 0.01	*130*	*22*	*2*	*18*	*420*	*50*	*35*	*0.2*
DK-05-Sp	OBW	0.2 ± 0.1	0.5 ± 0.1	8 ± 1	16.2 ± 2.9	3.8 ± 0.4	4.8 ± 0.4	0.4 ± 0.2	1.3 ± 0.2	0.3 ± 0.04								
DK-05-St		<0.2	0.4 ± 0.1	8 ± 1	18 ± 3	3.8 ± 0.4	4.8 ± 0.4		1.3 ± 0.2	0.3 ± 0.04								
FR-06-St	NMW	0.5 ± 0.1	2.3 ± 0.3	6 ± 2	8.4 ± 2.9	24 ± 1	26 ± 1.3	0.24 ± 0.04	1.1 ± 0.1	1.9 ± 0.1	*80*	*6.5*	*1*	*26*	*360*	*14*	*10*	
HR-07-Sp	NMW	<0.2	25 ± 2	21 ± 5	N.d.	2.9 ± 0.3	7 ± 1		2.5 ± 0.4	0.23 ± 0.03	*114*	*805*	27.1	*43*	2246	*116.1*	*262*	0.9
HR-07-St		0.4 ± 0.1	0.6 ± 0.1	7 ± 4	3.5 ± 2.3	12 ± 1	42 ± 2.1	0.6 ± 0.2	3.6 ± 0.3	0.9 ± 0.1	*63.8*	*1.8*	*0.6*	*32*	*381*	*7.2*	*2.9*	*0.02*
IT-08-Sp	NMW	1.7 ± 0.1	6.6 ± 0.7	77 ± 5	23 ± 9	92 ± 4	85 ± 3.5	0.26 ± 0.03	0.9 ± 0.1	7.4 ± 0.43	*164*	*31.12*		*49.5*	*243*	*402*	*49.4*	
NO-9-St	SpW	1.6 ± 0.1	14 ± 3	2.5 ± 1.1	7.9 ± 3.8	1.7 ± 0.2	2.3 ± 0.2	0.11 ± 0.02	1.3 ± 0.1	0.14 ± 0.02	*10.7*	*1.1*	*0.6*	*0.4*	*36.7*	*3*	*0.4*	
NO-10-Sp	OBW	<0.2	2.0 ± 0.4	2.3 ± 1.5	N.d.	0.8 ± 0.1	1.4 ± 0.1		1.7 ± 0.3	0.07 ± 0.01								
NO-11-Sp	OBW	0.2 ± 0.1	0.95 ± 0.15	<0.7	1.5 ± 1.0	0.6 ± 0.1	2.9 ± 0.2	0.20 ± 0.15	5 ± 1	0.05 ± 0.01								
SE - 12-Sp	NMW	0.8 ± 0.2	<0.2	4 ± 2.2	9.4 ± 3.1	0.2 ± 0.1	0.8 ± 0.1		3.4 ± 1.2	0.02 ± 0.01	*3.5*	*210*	*2*	*0.8*	*522*	*4*	*16*	*3.5*
SE - 13-Sp	SpW	6.0 ± 0.5	89 ± 13	6 ± 1.4	12 ± 3	1.2 ± 0.7	3.8 ± 0.9	0.07 ± 0.01	3.1 ± 1.9	0.10 ± 0.06	*15*	*20*	*< 2*	*3.1*		*8*	*34*	*0.3*
SE - 13-St		1.2 ± 0.1	7.0 ± 1.6	5 ± 1.7	14 ± 5	1.6 ± 0.1	1.7 ± 0.1	0.18 ± 0.04	1.1 ± 0.1	0.13 ± 0.02								
SE - 14-Sp	OBW	1.1 ± 0.1	7.4 ± 0.6	1.2 ± 0.8	4.2 ± 2.8	1.5 ± 0.1	1.5 ± 0.1	0.15 ± 0.02	1.0 ± 0.1	0.12 ± 0.01	*30*	*150*	*90*	*10*				
SE - 15-Sp	NMW	34 ± 3	49 ± 5	20 ± 5	N.d.	579 ± 25	938 ± 40	0.7 ± 0.1	1.6 ± 0.1	46 ± 3	*65*	*21*	*5*	*15*	*240*	*20*	*49*	*1.3*
SE - 15-St		20 ± 1	46 ± 4	19 ± 3	24 ± 5	600 ± 96	1001 ± 34	0.44 ± 0.04	1.7 ± 0.3	48 ± 8								
SE - 16-Sp	NMW	4.6 ± 0.3	15 ± 1	3.1 ± 1.3	6.8 ± 3.2	2.5 ± 0.2	2.2 ± 0.2	0.31 ± 0.03	0.9 ± 0.1	0.20 ± 0.02	*15*	*20*		*3*		*9*	*30*	*0.3*
SE - 16-St		2.1 ± 0.1	<0.2	2.8 ± 1.1	3.8 ± 1.8	1.7 ± 0.1	2.2 ± 0.2		1.3 ± 0.1	0.14 ± 0.01								
SE - 17-Sp	OBW	31 ± 2	49 ± 4	23 ± 3	7.9 ± 1.6	694 ± 49	1150 ± 80	0.6 ± 0.1	1.7 ± 0.2	56 ± 5								
SE - 17-St		2.3 ± 0.2	34 ± 3	23 ± 7	17 ± 6	714 ± 52	1162 ± 83	0.07 ± 0.01	1.6 ± 0.2	57 ± 6								
SE - 18-Sp	OBW	0.4 ± 0.2	1.9 ± 0.2	1.3 ± 0.5	3.4 ± 1.4	0.9 ± 0.2	1.8 ± 0.3	0.2 ± 0.1	1.9 ± 0.5	0.07 ± 0.02	*60*	*240*	*320*	*10*				
SE - 19-Sp	SpW	1.9 ± 0.4	4.2 ± 0.4	7.9 ± 1.7	13 ± 3	48 ± 1.9	49 ± 2	0.5 ± 0.1	1.0 ± 0.1	3.9 ± 0.2	*49*	*32*	*2.2*	*5.5*	*80*	*48*	*75*	*0.36*
SE - 19-St		2.6 ± 0.5	1.4 ± 0.1	7.5 ± 1.6	11 ± 3	44 ± 2	44 ± 2	1.9 ± 0.4	1.0 ± 0.1	3.5 ± 0.3								
SE - 20-Sp	NMW	3.4 ± 0.7	5.1 ± 0.5	0.9 ± 0.6	4.6 ± 3.4	2.5 ± 0.2	3.0 ± 0.2	0.7 ± 0.2	1.2 ± 0.1	0.21 ± 0.02	*32*	*1.3*	*0.51*	*1.2*		*6.7*	*2.1*	*< 0.1*
SE - 21-Sp	OBW	5.7 ± 1.2	12 ± 1	3 ± 1	8.3 ± 2.9	2.8 ± 0.6	2.5 ± 0.3	0.5 ± 0.1	0.9 ± 0.2	0.23 ± 0.06								
SE - 21-St		1.3 ± 0.1	0.4 ± 0.1	5.9 ± 1.6	21 ± 6	2.4 ± 0.3	2.6 ± 0.3	3.3 ± 0.6	1.1 ± 0.2	0.19 ± 0.03								
SE - 22-Sp	NMW	120 ± 9	101 ± 9	6 ± 2	17 ± 7	27 ± 4	51 ± 4	1.2 ± 0.1	1.9 ± 0.3	2.2 ± 0.4	*2*	*1.5*	*0.5*	*0.6*		*1.4*	*0.8*	*0.1*
SE - 22-St		75 ± 5	115 ± 11	9.2 ± 1.8	22 ± 5	24 ± 2	46 ± 2	0.6 ± 0.1	1.9 ± 0.2	1.9 ± 0.2								
SE - 23-Sp	SpW	1.1 ± 0.1	9.5 ± 1.1	38 ± 5	52 ± 9	<0.2	0.6 ± 0.3	0.11 ± 0.02			36	82	6.8	9.4				0.59
SE - 23-St		0.31 ± 0.04	2.5 ± 0.4	40 ± 6	50 ± 9	<0.2	0.4 ± 0.2	0.13 ± 0.03										
SE - 24-Sp	SpW	6.8 ± 0.6	11 ± 1	5.8 ± 1.5	13 ± 4	20 ± 3	33 ± 2.6	0.6 ± 0.1	1.6 ± 0.3	1.6 ± 0.3	31	15		3.9			20	0.8
SE - 25-Sp	OBW	7.9 ± 0.8	11 ± 1	2.7 ± 1.3	9.2 ± 5.2	22 ± 4	38 ± 2	0.7 ± 0.1	1.7 ± 0.3	1.8 ± 0.3								
SE - 26-Sp	OBW	<0.2	0.5 ± 0.1	1.5 ± 0.6	3.1 ± 1.5	1.3 ± 0.2	2.2 ± 0.3		1.8 ± 0.3	0.10 ± 0.02								

N.d. not determined. *__Country code:__ Bosnia and Herzegovina (BA); Germany (DE); Denmark (DK); France (FR); Croatia (HR); Norway (NO) and Sweden (SE).

Table 4
Activity concentration of measured naturally occurring radionuclides in bottled water in studies carried out in Europe: range, average and median values.

		^{210}Po (mBq/L)	^{210}Pb (mBq/L)	^{226}Ra (mBq/L)	^{228}Ra (mBq/L)	^{238}U (mBq/L)	^{234}U (mBq/L)	Reference
Sweden	**Range**	0.2–120	0.4–115	0.7–77	2–52	0.2–714	0.2–1162	Current study
	Average	11 ± 25	19 ± 30	12 ± 15	13 ± 12	92 ± 215	129 ± 332	
	Median	2	7	6	9	3	3	
Hungary	**Range**	3–19		4–2940		9–98	11–92	Kovacs et al. (2004)
	Average	7 ± 6		273 ± 684		35 ± 30	38 ± 27	
	Median	4		47		23	27	
Poland	**Range**	0.3–3		3–641	20–250	0.4–1.5	0.4–1.5	(Chmielewska et al., 2020;
	Average	1 ± 1		95 ± 155	78 ± 86	0.8 ± 0.4	0.8 ± 0.4	Skwarzec et al., 2003)
	Median	0.7		39	50	0.8	0.8	
Italy	**Range**	0.04–21		10–52.5		0.2–89	0.2–79	Desideri et al. (2007)
	Average	2 ± 4		15 ± 11		9 ± 15	12 ± 15	
	Median							
Romania	**Range**			29–450		40–170		Calin et al. (2015)
	Average			136 ± 116		84 ± 33		
	Median			110		84		
Bosnia and Herzegovina	**Range**			1.1–791	0.2–221	13–367		Kasic et al. (2015)
	Average			94 ± 167	40 ± 55	95 ± 97		
	Median			22	15	68		
Spain	**Range**	1–49	1.4–78	1–194	1–52	1–80	1–173	(Diaz-Frances et al., 2013;
	Average	7 ± 11	15 ± 20	26 ± 43	11 ± 13	11 ± 15	19 ± 30	Perez-Moreno et al., 2020)
	Median	2	6	7	5	7	9	
Austrian	**Range**	0.4–110	2–31	2–211	4–236	0.4–100	1–94	Kralik et al. (2003)
	Average	12 ± 23	8 ± 8	39 ± 53	48 ± 62	18 ± 26	21 ± 27	
	Median	4	5	17	21	6	6	
Slovenia	**Range**	0.2–2.1	1–9	1.1–32	1–5	1–53	3–173	Benedik and Jeran (2012)
	Average	0.9 ± 0.6	4 ± 3	11 ± 9	2 ± 2	13 ± 16	35 ± 51	
	Median	0.6	3	11	2	5	13	

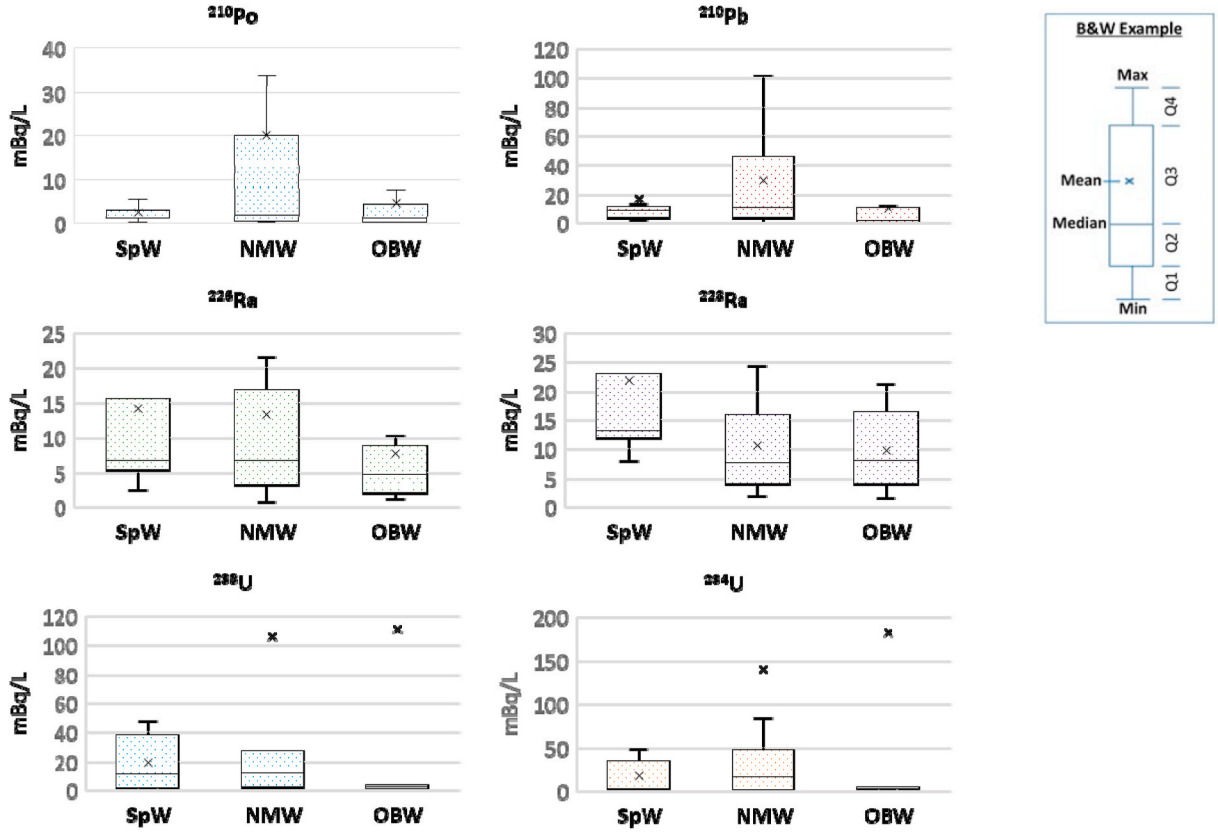

Fig. 1. *Box and whisker diagram for each of the measured radionuclides grouped by the type of bottled water: Spring water (SpW), natural mineral water (NMW) and other waters in bottles (OBW) together with a box and whisker diagram explanation. Outlier values are not shown in the diagram.*

spring dates from the ice age and it is located in Gällivare municipality in northern Sweden. The water springs up by itself after a natural purification process of more than 26 year that occurs in a granitic bedrock area (Sarlus et al., 2020). Granites rocks are known to have elevated levels of naturally occurring radionuclides like ^{238}U and its progeny (^{226}Ra, ^{220}Rn, ^{210}Pb) (IAEA, 2017). This is further supported by the fact that the municipality has observed relatively high concentrations of radon surrounding the aquifer (Gällivare kommun, 2014), which could

Table 5
Outlier values for each radionuclides in each bottled water group.

	SpW	NMW	OBW
^{210}Po	SE-24-Sp	SE-22	SE-17-Sp
^{210}Pb	SE-13-Sp	SE-22-St	SE-17
^{226}Ra	SE-23	IT-08-Sp	SE-17
^{228}Ra	SE-23		
^{238}U		IT-08-Sp	SE-17
		SE-15	SE-25-Sp
^{234}U		SE-15	SE-17
			SE-25-Sp

be an important source of ^{210}Pb due to the high solubility of ^{222}Rn in water (Szabo et al., 2020). On the other hand, the activity concentration of ^{210}Pb was higher than uranium radionuclides, highlighting geochemical and hydrological process that favors the solubility of lead. In that sense, the aquifer is locating in an important ferrous metal mining area (Sarlus et al., 2020). Therefore, the mineral composition of the area rich in metals like iron and manganese could increase the reducing condition favoring the release of ^{210}Pb from the bedrock to the aquifer (Perez-Moreno et al., 2020). However, further studies will be necessary to investigate the radionuclides speciation in the aquifer. The activity concentration of ^{226}Ra and ^{228}Ra ranged from <0.7 mBq/L to 77 mBq/L (IT-08-Sp) and from 1.5 mBq/L (NO-11-Sp) to 52 mBq/L (SE-23-Sp), respectively. The radium levels detected were lower than the values reported in other studies carried out in Europe, especially when comparing with bottled water studies from Hungary, Poland and Romania (see Table 4). High levels of radium radionuclides were detected in samples from two brands, IT-08 and SE-23. IT-08-Sp is a thermal natural mineral water with high mineral composition (see Table 4). Different studies had demonstrated that radium mobility is higher in thermal water with high mineral composition, resulting in a higher activity concentration of ^{226}Ra (Guerrero et al., 2016; IAEA, 2014; STUK, 2005). In this bottled water, the ratio ^{226}Ra/^{234}U and ^{238}U/^{234}U, taking into account the experimental uncertainties, was around one, highlighting the secular equilibrium as a result of the longevity of the aquifer. On the other hand, the bottled water from SE-23 had low levels of the others measured naturally occurring radionuclides and high levels of both ^{226}Ra and ^{228}Ra. The mineral composition is not high and the high levels of radium radionuclides could be the results of the lithology of the aquifer (clay, shale, coal (Rhaetian to Tithonian), sandstone) favouring local enrichments in radium radionuclides.

In the majority of the studied bottled water, low levels of uranium radionuclides were detected, with a median value of 3 mBq/L for both radionuclides (^{238}U and ^{234}U), being considerably less than the activities measured in bottled water in other European countries (see Table 4). However, samples from brand SE-15 and SE-17 showed an activity concentration more than 200 times higher than the median values. The origin of SE-17 water was not specified in the label, while the natural spring of SE-15 is located in Kolsva (Köping, Sweden), which is an area rich in uranium (Mantero et al., 2020). Furthermore, this bottled water also exceeded the chemical threshold value for U in drinking water (30 ppm) according to WHO and related to its chemical toxicity (Frisbie et al., 2013).

The ^{234}U/^{238}U ratio ranged from 0.9 to 5.2 with an average and median value of 1.8 ± 1.1, and 1.6, respectively. Considering the experimental uncertainties, the uranium ratios in most of the bottled water analysed were higher than one, demonstrating the disequilibrium between ^{234}U and ^{238}U, which has been well documented in natural water (Borylo & Skwarzec, 2014). For example, the average uranium ratio in river water could oscillate from 1.0 to 2.1, while in groundwater uranium activity ratios from 0.5 to 9.0 were reported (Borylo & Skwarzec, 2014). Uranium disequilibrium occurs as a result of complex geochemical processes influenced by the nature of the aquifer, the uranium chemistry, the recoil process, and it could be further enhanced due to anthropogenic contributions such as migration of fertilizers (Milena-Perez et al., 2021).

3.2. Comparison between types of bottled water

Fig. 1 shows the box and whisker diagram for each of the measured radionuclide clustered by the type of bottled water. Considering the median values, the activity concentration of the measured radionuclides was comparable for the three types of bottled water. That hypothesis was also supported by the Mann–Whitney *U* test (results not showed) confirming no statistically significant differences between median values.

Average values were higher than median values, even in some cases higher than the third quartile, highlighting the presence of outliers such

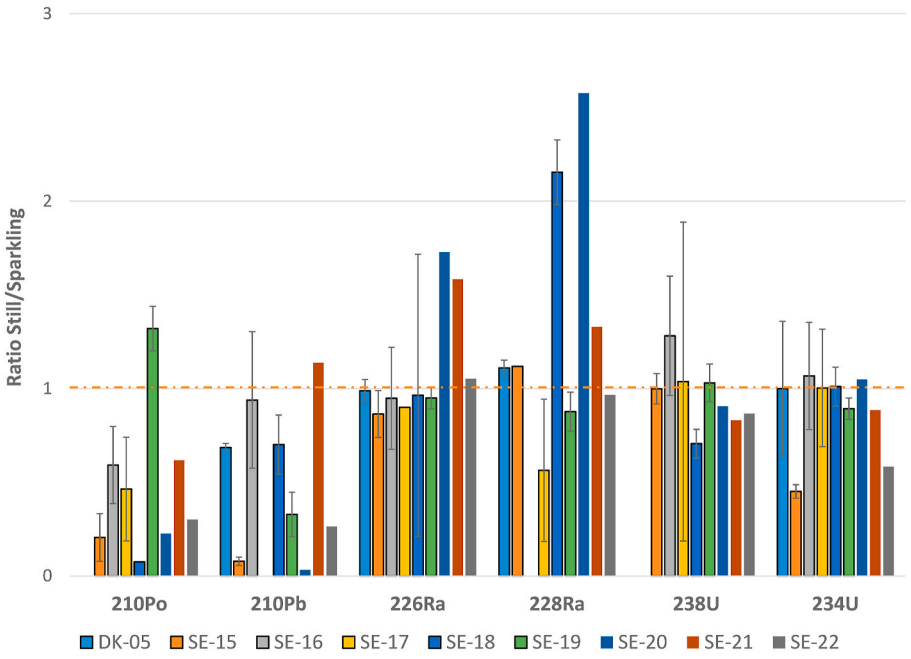

Fig. 2. Ratios of activity concentration, still to sparkling water, for nine brands and for each measured radionuclide.

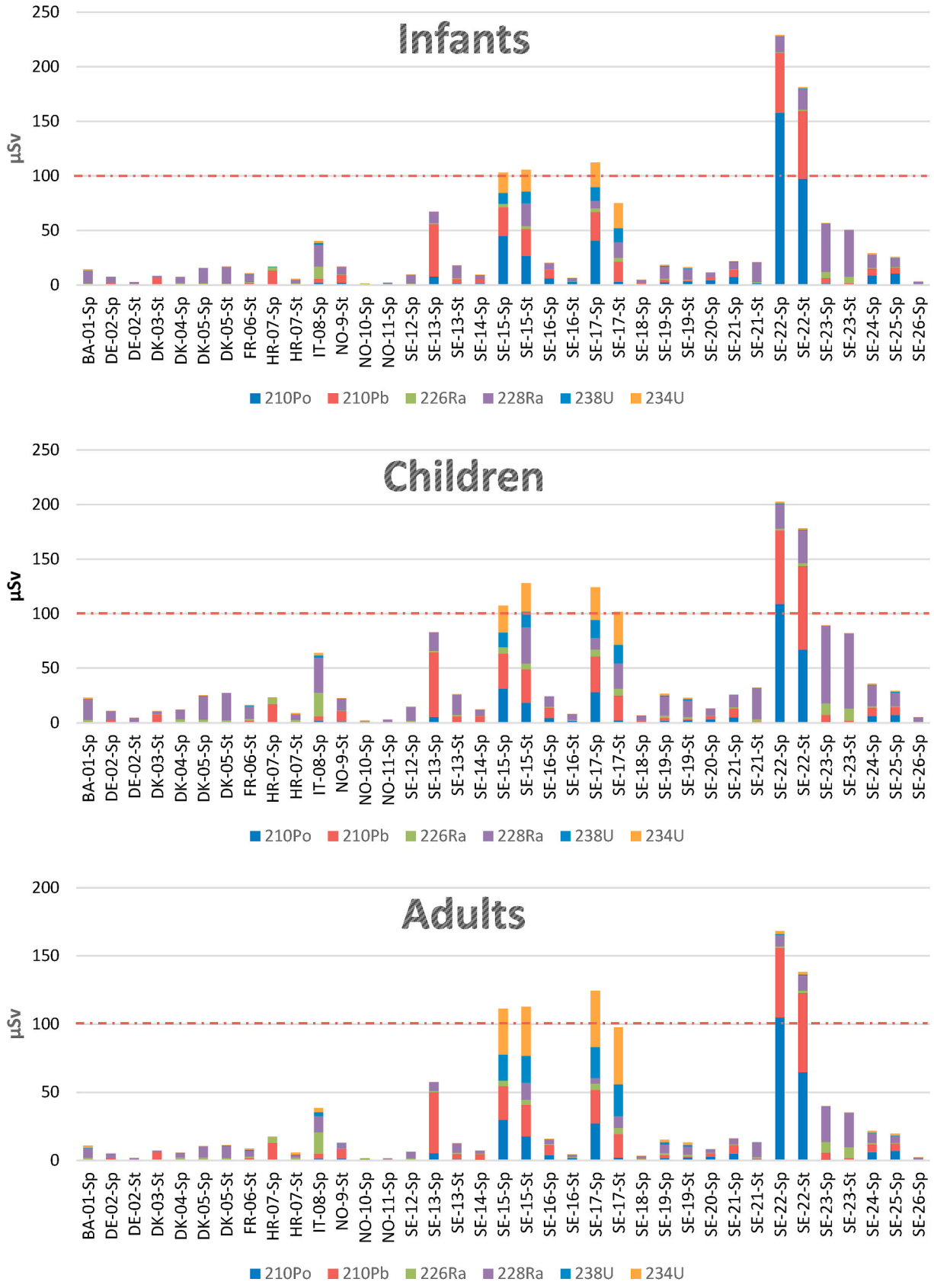

Fig. 3. *Results of the dose assessment for different age groups (top: Infants; middle: Children and bottom: Adults) from the intake of the naturally occurring radionuclides (^{210}Po; ^{210}Pb; ^{226}Ra; ^{228}Ra; ^{238}U; ^{234}U) measured in bottled water available on the Swedish market.*

as the levels of ^{238}U and ^{234}U detected in the NMW (SE-15) or OBW (SE-17). All outliers detected in each bottled water group are shown in Table 5.

Regarding ^{210}Po and ^{210}Pb, the largest spread was found in NMW while the trends for SpW and OBW were very similar. The diagram also shows that higher values of ^{228}Ra were found in SpW. In relation to uranium isotopes, the lowest dispersion was found in OBW while ^{234}U activity concentration was a little larger in NMW. Finally, the consumption of each type of bottled water results in the intake of the measured radionuclides in the following order based on the median values of activity concentration:

- NMW: ^{234}U > ^{238}U > ^{210}Pb > ^{228}Ra > ^{226}Ra > ^{210}Po.
- SpW: ^{238}U > ^{228}Ra > ^{210}Pb > ^{226}Ra > ^{234}U > ^{210}Po.
- OBW: ^{228}Ra > ^{226}Ra > ^{234}U > ^{238}U > ^{210}Pb > ^{210}Po.

3.3. Comparison between still and sparkling water

Fig. 2 shows, for nine brands, the still and sparkling activity concentration ratio for each measured radionuclide. In general, the ratios, considering the uncertainties, were around unity or lower than unity (Fig. 2). The activity concentrations of ^{238}U, ^{234}U and ^{226}Ra were very similar in both types of water. However, a clear trend pointed out an enhancement on ^{210}Pb/^{210}Po in sparkling water compared to still water. However, due to the lack of information about water processing (water batch, treatment, bottling date, etc.) it is unclear if the dissolved carbon dioxide in sparkling water may have any influence on the radioactivity levels observed, and further studies are necessary to support this hypothesis especially for ^{210}Po and ^{210}Pb.

3.4. Committed effective dose by bottled water consumption

Fig. 3 displays the committed effective dose assessment results for different age groups, resulting from the intake of the measured naturally occurring radionuclides (^{210}Po; ^{210}Pb; ^{226}Ra; ^{228}Ra; ^{238}U; ^{234}U) by drinking bottled water available on the Swedish market. The analysed bottled water showed good quality from the radiological point of view, with an average value of committed effective dose for infants, children and adults of 37 ± 51 μSv/y; 44 ± 50 μSv/y and 32 ± 44 μSv/y; respectively and a median value of 17 μSv/y, 24 μSv/y and 13 μSv/y, respectively. Despite the lower water consumption for children and infants, the committed effective dose accounted for large doses compared to adults because of the higher effective dose coefficients. Consumption of bottled water available in Sweden, favoured the intake of the detected naturally occurring radionuclides in the following order: ^{228}Ra > ^{226}Ra > ^{210}Pb > ^{238}U > ^{234}U > ^{210}Po. In fact, the dose contribution from each naturally occurring radionuclide decreases as follows, for infants and adults: ^{228}Ra > ^{210}Pb > ^{210}Po > ^{226}Ra > ^{238,234}U and for children: ^{228}Ra > ^{210}Pb > ^{226}Ra > ^{210}Po > ^{238,234}U.

The results showed that among the 26 brands studied; only the continuous ingestion of bottled water from three of them (SE-15, SE-17 and SE-22) could exceed, in all age group studied, the threshold value for drinking water 100 μSv/year. On the one hand, the bottled water from SE-15 and SE-17 were characterized by high mineral content and high levels of ^{234}U together with a non-negligible contribution from ^{210}Po, ^{210}Pb and ^{238}U. Regarding the results from SE-17, the main difference between still and sparkling water was the difference in activity concentration of ^{210}Po (Fig. 3). The reason for this difference could be due to bottling time or processing method, however, this information was not available. On the other hand, the dose contribution from SE-22 was mainly controlled by the high levels of ^{210}Po and ^{210}Pb.

4. Conclusions

The results provided by this study is valuable for the consumers in order to have a good overview on the impact of drinking bottled water from the radiological point of view, especially when large volumes of bottled water are consumed. In the majority of the samples measured, the radionuclide with the highest contribution to the ingestion dose is ^{228}Ra. However, there are three brands, SE-15, SE-17 and SE-22, that deserve special attention, since large consumption of these brands could increase the total dose received by population per year to more than 100 μSv reaching for sample SE-22 up to 229 μSv for infants, 202 μSv for children and 168 μSv for adults. The high dose levels of this water is the result of high levels of uranium radionuclides for SE-15 and SE-17 while for SE-22 it is linked to the high levels of ^{210}Po and ^{210}Pb.

There is a clear trend that points out an enhancement on ^{210}Pb and ^{210}Po in sparkling water compared to still water, but due to lack of information about water manufacturing it is a challenge to find out the reasons for this behaviour. It is important to note that bottled water with high levels of ^{210}Pb, it could be important to consider the time elapsed from bottling to consumption since the ingrowth of ^{210}Po by ^{210} Pb decay could considerably increase the dose received for consumers. However, if the water has a source of ^{210}Po (more ^{210}Po than ^{210}Pb) then the time passed will decrease the dose received by consumers.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

F. Piñero-García: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Funding acquisition. **R. Thomas:** Investigation, Resources, Writing – review & editing. **J. Mantero:** Investigation, Writing – review & editing. **E. Forssell-Aronsson:** Resources, Writing – review & editing, Supervision, Funding acquisition, Supervision. **M. Isaksson:** Resources, Writing – review & editing, Supervision, Funding acquisition, Supervision.

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