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RADIOLOGICAL OF NATURAL AND MINERAL DRINKING WATERS IN SLOVENIA

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Various types of water were collected in Slovenia and analysed in order to assess the radiation doses from ^{238}U, ^{234}U, ^{228}Ra, ^{226}Ra, ^{210}Pb and ^{210}Po for three different age groups of the population. It was found in all cases that the calculated median committed effective dose from the investigated radionuclides for each population group was well below the recommended value of 100 μSv y^{-1}, ranging from 4 to 7 μSv y^{-1} for adults, from 8 to 10 μSv y^{-1} for children and from 6 to 7 μSv y^{-1} for infants. Of the investigated groups of the population children are the most exposed with the highest absolute doses of 19.1 and 18.7 μSv y^{-1} after drinking a certain bottled brand of mineral and natural water, respectively. The contribution of each particular radionuclide to total doses varied among different water types and within each type, as well as between different age groups.

INTRODUCTION

In most European countries, there is an increasing tendency to replace tap water of satisfactory quality for human consumption with commercial bottled natural (BN) and mineral water. Moreover, various beverages are also prepared from mineral water, not ordinary tap water. Systematic studies on the radiological characterisation of drinking water started after 1993, when the recommendations of the Guidelines for drinking water quality issued by the World Health Organization (WHO) were published[1]. These guidelines state that drinking water is safe from the radiological point of view if within the range of normal consumption (2 l per day), the annual dose rate originating from the presence of radioactive nuclides does not exceed 0.1 mSv. UNSCEAR reports[2, 3] estimated that exposure to natural sources contributes >98 % of the radiation dose to the population (medical treatment not taken into account). The main contribution to dose is largely due to the presence of naturally occurring radionuclides of both the uranium and thorium decay series. Owing to their high radiotoxicity, the contributions of ^{210}Po and ^{228}Ra to the dose are more pronounced. The dose contributions of the radionuclides due to water consumption found in literature are in the order: ^{210}Po>, ^{228}Ra>, ^{210}Pb>, ^{226}Ra>, ^{234}U>, ^{238}U>, ^{224}Ra> and ^{235}U[4]. Increased concerns regarding the radiological quality of drinking water have led to an increased demand for real data assessment. The old drinking water regulation 980/778/EEC from 1980[5] in which neither radioactivity nor uranium was mentioned was replaced by the European Directive 98/83/EC in 1998[6]. In this Directive, the reference dose level of the committed annual effective dose due to drinking water consumption is 0.1 mSv. The Directive points out that the total indicative dose must be evaluated excluding tritium, ^{40}K, ^{14}C, radon and its decay products, but including all other radionuclides of the natural decay chains. The maximum values for radon and long-lived radon decay products such as ^{210}Pb and ^{210}Po are proposed in the European Commission Recommendation 2001/928/Euratom[7]. Uranium is covered by the Directive, although its contribution to the dose is minor due its small dose conversion factor. Uranium is known as a toxic heavy metal and its intake in water should be limited by consideration of its toxicity to the kidney[8]; therefore, it has to be regulated and controlled. The WHO set a limitation of 2 μg l^{-1} in its 1998 report[9], but later[10] changed this stringent limit to 15 μg l^{-1}; the USA[11] set the limit to 20 μg l^{-1}. Recently, Germany set the uranium limit of 2 μg l^{-1} for mineral water considered suitable for infants[12]. Considering the importance of water for human consumption, its quality has to be assured and regularly controlled. The assessment of the radiological quality of natural or bottled drinking and mineral waters is also important in view of the assessment and reduction of the radiation exposure of the population. For practical purposes, the recommended screening levels for drinking water below which no further actions are required are 0.1 Bq l^{-1} for gross alpha activity and 1 Bq l^{-1} for gross beta activity. If these values are exceeded, determination of particular radionuclides dissolved in drinking water needs to be performed. In 2004, the WHO published the third edition of its guidelines for drinking water[10] in which the recommended

screening level for gross alpha activity was increased from 0.1 to 0.5 Bq l^{-1}. Owing to the increasing tendency in the consumption behaviour of the population to replace surface tap water of sufficient quality for human consumption with commercial bottled drinking natural and mineral water, several studies to assess the radioactivity levels in bottled drinking and mineral water were performed around Europe[4, 13–29], while data on drinking water quality in Slovenia are very scarce. In Slovenia, monitoring of natural radionuclides in ground water in the vicinity of the former uranium mine Žirovski vrh as well as monitoring of ground and drinking water in the vicinity of the nuclear power plant Krško is performed regularly. Only limited information was obtained on the levels of natural radionuclides in drinking water collected in the area of Šoštanj, where the thermal power plant is located[30–32].

The experimental design of this study was to determine the activity concentrations of ^{238}U, ^{234}U, ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po in Slovenian bottled drinking and mineral water, as well as in tap water. The studied samples included bottled drinking and mineral water purchased on the market in Ljubljana. The present study was focused only on water samples originating from Slovenia.

MATERIALS AND METHODS

There are many companies in Slovenia producing natural drinking and natural mineral water from bedrock aquifers of different depths. Tap water was collected from chosen cities in Slovenia. Figure 1 shows the sampling points of the waters investigated.

For this study, three of the most frequently sold natural mineral waters and eight bottled drinking waters 'off the shelf' were analysed. Seven tap waters were analysed. Although, two mineral waters, BM2 and BM3 and two bottled drinking waters BN7, BN8, (Figure 1) coming from the same region in the north-east of Slovenia they have different commercial names and they differ from each other chemically as well as in origin and depths of bedrock or exploitation. BM2 belongs to the so-called Mg-Na-HCO_3-SO_4 type of natural mineral water with very high Mg content (1000 mg l^{-1}) and with the total content of dissolved solid minerals of 13.2 g l^{-1} and exploitation depths between 275 and 606 m, while BM3 belongs to Na-Mg-Ca-HCO_3-SO_4 type with the total content of dissolved minerals of 1.5 g l^{-1} and exploitation depths between 170 and 302 m[33]. Natural bottled mineral (BM) waters BN7 and BN8 have much lower total content of dissolved minerals and shallowness depth of exploitation. During the sampling period, the authors analysed different batches of the same producer bought in different shops but no differences in activity concentrations were obtained (results not shown).

All reagents used in the analysis were of analytical grade. The tracer solutions (^{232}U, ^{209}Po, ^{133}Ba) and ^{228}Ra standard solution used in the study were prepared from calibrated solutions purchased from

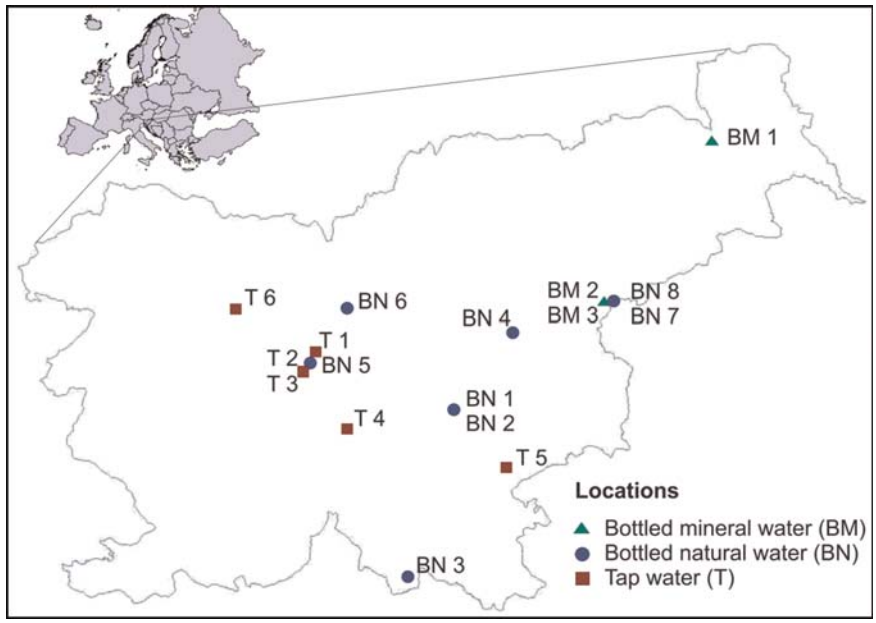

Figure 1. Locations of tap water samples and bottling facilities for natural and mineral waters from Slovenia.

Analytics, Inc. (Atlanta, GA, USA). The producer maintains traceability to the National Institute of Standards and Technology.

Instruments

An alpha spectrometer (CANBERRA's Alpha Analyst^{TM}) with passivated implanted planar silicon semiconductor detectors with an active area of 450 mm^2 and 28 % efficiency for 25-mm diameter discs was used for alpha-particle spectrometry measurements. The calibration of the detectors was made with a standard radionuclide source, containing ^{238}U, ^{234}U, ^{239}Pu and ^{241}Am (code: 67978-121), obtained from Analytics, Inc. A coaxial HP Ge detector was used for measurement of the gamma-emitting nuclides ^{133}Ba (E_γ=356.01 keV, P_γ=62.05 %) and ^{228}Ac (E_γ=911.20 keV, P_γ=25.8 %). For evaluation of gamma spectra, the Hyperlab[34] program was used. A Canberra LB 4110 low-background gas-flow proportional counter was used for the determination of beta emitter ^{210}Pb via its decay product ^{210}Bi.

Radiochemical separation of investigated radionuclides

For determination of uranium radioisotopes a known amount of ^{232}U tracer (~0.3 Bq) was added to the water sample which was acidified with concentrated HNO_3 (3 ml of acid per 1 l of sample). The volume of water was up to 2 l. Uranium was preconcentrated from the water samples by coprecipitation with iron(III) hydroxide at pH 9–10 using an ammonia solution[35]. The precipitate was separated by decantation, centrifugation, washed with distilled water and dissolved in concentrated nitric acid. The solution was adjusted with distilled water to 3 M HNO_3 and loaded onto an UTEVA column (Eichrom Industries Inc.)[36] pre-conditioned with 5 ml 3 M HNO_3. The column was then washed with 3 M HNO_3. Thorium radioisotopes were stripped from the column with 9 and 5 M HCl. Uranium radioisotopes were eluted with 15 ml of 1 M HCl. The microcoprecipitation method with neodymium fluoride was used for thin source preparation for alpha spectrometric determination[37, 38]. The neodymium fluoride suspension was filtered through a 25-mm diameter 0.1-μm polypropylene filter. The dry filter was mounted on a stainless steel disc.

The analytical scheme for determination of ^{226}Ra was adapted from Lozano *et al.*[39]. The procedure is based on coprecipitation of Pb(Ra)(Ba)SO_4. The water sample of up to 2 l was transferred to a glass beaker and acidified with concentrated H_2SO_4 (10 ml of sulphuric acid per 1 l of sample). After addition of ^{133}Ba tracer together with Ba-carrier, the sample was stirred for ~30 min. With stirring, 30 mg Pb^{2+} was added in portions to allow good coprecipitation of radium and barium. After settling, the suspension was centrifuged and washed with distilled water. The $PbSO_4$ precipitate containing radium and barium was dissolved in 4 ml 0.1 M EDTA, prepared in 0.5 M NaOH. For ^{226}Ra determination 250 μg of 0.3 mg ml^{-1} Ba^{2+} solution was added together with 4 ml of saturated Na_2SO_4 solution. With stirring, 1:1 acetic acid solution was added until pH 4-5 was reached, thus precipitating $BaSO_4$, while Pb^{2+} ions remained in solution. Immediately after, 0.2 ml of a 0.125 mg ml^{-1} $BaSO_4$ suspension was added, acting as a seeding precipitate to obtain small particles. The suspension was allowed to settle for 30 min and filtered through a 25 mm 0.1-μm polypropylene filter. The filter with $BaSO_4$ deposit was dried and mounted on a stainless steel disc and measured by gamma–ray spectrometry for ^{133}Ba yield determination and by alpha–particle spectrometry for determination of ^{226}Ra.

For determination of ^{210}Pb and ^{210}Po in water, 25 mg Pb^{2+} of lead carrier and ^{209}Po (~0.3 Bq) tracer were added to 9 l of water and then the radionuclides coprecipitated with MnO_2[40]. Precipitation of MnO_2 was achieved by adding $KMnO_4$ and $MnCl_2$ and adjusting the pH to 9 with ammonia solution. The precipitate was then dissolved with a mixture of HCl and H_2O_2, adjusted with distilled water to 2 M HCl and loaded on a Sr resin column (Eichrom Industries Inc.). The analytical method is based on selective separation of lead and polonium by extraction chromatography with bis-4,4′(5′)-t-butyl-cyclohexano-18,6-crown ether[41]. The non-retained ions were washed from the column with 100 ml 2 M HCl. Polonium was stripped with 6 M HNO_3 while lead was removed with 6 M HCl solution. The spontaneous deposition of polonium on a 19-mm diameter silver disc was carried out at 90°C for 4 h. The Ag disc, covered on one side, was fixed in a holder and immersed in the solution[42]. Polonium radioisotopes were then measured by alpha-particle spectrometry. Lead was precipitated as lead sulphate and the beta activity of its daughter ^{210}Bi measured at equilibrium on a low background gas-flow beta proportional counter.

The analytical scheme for determination of ^{228}Ra is based on coprecipitation of (Ra)(Ba)SO_4. The water sample of 10 l was transferred to a glass beaker and acidified with concentrated H_2SO_4 (10 ml of sulphuric acid per 1 l of sample). After addition of ^{133}Ba tracer, coprecipitation of radium and barium was done with addition of 2 ml of Ba-carrier (30 mg ml^{-1}). After settling, the suspension was centrifuged and washed with distilled water. The precipitate containing radium and barium was kept for ~30 h and then ^{228}Ra was determined by gamma-ray spectrometry via its ingrown' daughter ^{228}Ac ($T_{1/2}$=6.15 h, E_γ=911.20 keV, P_γ=25.8 %). Recovery of the radiochemical procedure was

Table 1. Radionuclide activity concentrations (in mBq l^{-1}) in BN, BM waters and tap waters (T) in Slovenia.

Sample	Type of sample	^{238}U	^{234}U	^{228}Ra	^{226}Ra	^{210}Pb	^{210}Po
BN1	Natural water	28 ± 3	71 ± 8	1.1 ± 0.1	32 ± 4	7.2 ± 1.2	1.1 ± 0.4
BN2		18 ± 1	57 ± 6	1.5 ± 0.3	11 ± 1	8.7 ± 1.0	2.1 ± 0.3
BN3		13 ± 2	15 ± 2	1.8 ± 0.3	1.7 ± 0.2	5.0 ± 0.7	0.9 ± 0.2
BN4		2.2 ± 0.5	3.5 ± 0.7	<0.05	2.3 ± 0.3	1.1 ± 1.0	0.43 ± 0.12
BN5		8.3 ± 1.1	12 ± 1	1.7 ± 0.2	0.14 ± 0.05	7.8 ± 4.1	2.0 ± 0.4
BN6		2.9 ± 0.5	13 ± 2	3.5 ± 0.6	6.8 ± 0.8	2.5 ± 0.6	0.41 ± 0.13
BN7		5.1 ± 0.6	14 ± 2	1.2 ± 0.1	16 ± 2	2.3 ± 0.6	0.24 ± 0.07
BN8		4.2 ± 2.8	8.7 ± 2.9	1.2 ± 0.1	15 ± 2	2.5 ± 0.6	0.60 ± 0.21
BM1	Mineral water	1.1 ± 1.6	2.8 ± 2.4	4.4 ± 0.1	2.4 ± 0.3	1.9 ± 0.9	0.39 ± 0.11
BM2		53 ± 9	173 ± 28	5.3 ± 0.5	12 ± 2	2.6 ± 0.8	0.82 ± 0.22
BM3		5.2 ± 1.8	12 ± 2	1.9 ± 0.2	17 ± 2	2.7 ± 0.7	0.57 ± 0.15
T1	Tap water	7.2 ± 0.7	8.5 ± 0.8	2.8 ± 0.5	1.0 ± 0.2	2.0 ± 0.6	0.25 ± 0.06
T2		4.8 ± 0.9	6.9 ± 1.1	2.7 ± 1.1	0.47 ± 0.10	9.7 ± 2.4	1.0 ± 0.2
T3		6.7 ± 1.1	11 ± 2	1.0 ± 0.1	1.3 ± 0.1	3.0 ± 1.3	0.77 ± 0.16
T4		8.2 ± 2.8	8.8 ± 2.9	1.2 ± 0.1	15 ± 2	5.6 ± 1.3	1.1 ± 0.2
T5		7.0 ± 0.9	11 ± 1	3.7 ± 0.5	1.4 ± 0.1	13.2 ± 0.7	1.8 ± 0.4
T6		1.1 ± 0.3	3.0 ± 0.5	0.9 ± 0.1	0.30 ± 0.03	3.3 ± 0.7	0.67 ± 0.19
T7		4.0 ± 0.4	7.5 ± 1.3	4.6 ± 0.5	1.9 ± 0.2	0.6 ± 0.5	0.66 ± 0.15

determined with ^{133}Ba. Owing to very low activity concentration of ^{228}Ra in the samples, the measurement time was >250 000s. The minimum detection activity using the Curie method[43] was determined with a confidence level of 95 %.

RESULTS AND DISCUSSION

In Table 1 results for the activity levels of ^{238}U, ^{234}U, ^{228}Ra, ^{226}Ra, ^{210}Pb and ^{210}Po in three different groups of drinking water, natural and mineral bottled water and tap water are given in mBq l^{-1}. The values obtained are relatively low and comparable with some literature data from Italy, Austria and Hungary[4, 15–19, 28, 29] and well below the limit values[6, 10]. As evident, the activity concentrations of uranium in all types of water samples analysed, except mineral water BM2, ranged from 1 to 28 mBq l^{-1} and from 3 to 71 mBq l^{-1} for ^{238}U and ^{234}U, respectively, with median levels of 6 and 11 mBq l^{-1} for these two radionuclides, respectively. In the mineral water BM2, coming from the eastern part of the country, also known as a spa region, activity concentrations of 53 mBq l^{-1} for ^{238}U and 173 mBq l^{-1} for ^{234}U were determined. Somewhat elevated levels of uranium of 13–28 mBq l^{-1} for ^{238}U and 15–71 mBq l^{-1} for ^{234}U were found also in three BN waters originating from the eastern (BN1, BN2) and southern (BN3) part of the country.

Activity concentrations of ^{226}Ra varied in the range between 0.14–32 mBq l^{-1} and like uranium can be regarded as low and comparable with data on radionuclide concentration reported for Europe. The highest absolute level of 32 mBq l^{-1} was found in BN water (BN1), which also had an elevated uranium level. Elevated activity concentrations higher than 10 mBq l^{-1} were found in two mineral waters (BM2 and BM3) coming from the eastern part of the country (Figure 1), in three BN waters [BN2, BN7, BN8 and one tap water (T4)]. All other BN waters originating from the Central part of the country, as well as tap waters, had low levels of ^{226}Ra in the range between 0.14–6.8 mBq l^{-1} and 0.3–1.9 mBq l^{-1}, respectively.

Among the alpha emitters analysed ^{210}Po had the lowest activity concentrations in the range between 0.24 and 2.1 mBq l^{-1} in all three groups of water investigated.

In all three groups of drinking water beta emitters ^{228}Ra and ^{210}Pb were also analysed. Activity concentrations of ^{228}Ra in the range between <0.05 and 5.3 mBq l^{-1} with median levels of 4.4, 2.7 and 1.4 mBq l^{-1} for BM water, tap water and BN water, respectively, were found. Activity levels of ^{210}Pb were also low in the range between 0.6 and 13.2 mBq l^{-1} with median levels of 3.8, 3.3 and 2.6 mBq l^{-1} for BN water, tap water and BM water, respectively. The highest absolute level of 13.2 mBq l^{-1} was found in tap water (T5) from the south eastern part of the country (Figure 1).

Dose estimation

Based on the results found for activity concentrations of six radionuclides in different samples of drinking waters presented in Table 1, the internal doses (committed effective dose) for three different

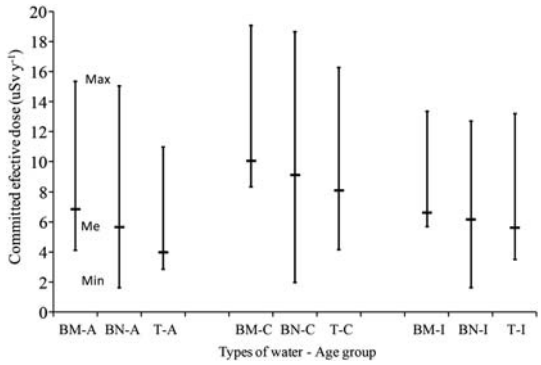

Figure 2. Committed effective doses to adults (A), children of age group 7–12 y (C) and infants (1–2 y) (I) due to radionuclide intake from drinking BN and mineral water (BM) and tap water (T) presented as median (Me), minimum (Min) and maximum (Max) levels.

Table 2. Effective dose coefficient (in μSv Bq^{-1}) and water consumption rate (in l y^{-1}) for three different age groups of the population.

Radionuclide	Effective dose coefficient (μSv Bq^{-1})[44]		
	Adults	Children (7–12 y)	Infants (1–2 y)
^{238}U	0.045	0.068	0.120
^{234}U	0.049	0.074	0.130
^{228}Ra	0.690	3.900	5.700
^{226}Ra	0.280	0.800	0.960
^{210}Pb	0.690	1.900	3.600
^{210}Po	1.200	2.600	8.800
Water consumption rate[10] (l y^{-1})	750	350	150

age groups of population [adults, children (7–12 y), infants (1–2 y)] were estimated (Figure 2) using the age-dependent annual consumption rate from the UNSCEAR report[3] and Guidelines for Drinking Water Quality[10] (Table 2), and the dose coefficients of the relevant radionuclides from the 'International Basic Safety Standards for Protection against Ionizing Radiation and for Safety of Radiation Sources'[44] (Table 2). For each water sample a conservative assumption was used, namely that yearly consumption as given in Table 2 consisted of drinking only one particular water sample, which in most cases is not true. Further, children (7–12 y) and infants (1–2 y) rarely consume mineral waters in higher amounts.

The annual committed effective doses assessed for the three different age groups drinking three different water types are summarised in Figure 2 as median values, together with the span of doses from minimum to maximum values. It can be observed that for each type of water, as well as for each population group, the range of doses is rather large, with all values; however, well below the recommended reference dose level of WHO of <100 μSv y^{-1}. Drinking BM water, the highest median internal doses were assessed for all three age groups (Me: 6.9, 10.1 and 6.6 for adults, children and infants, respectively), followed by BN water (Me: 5.7, 9.1, 6.2 μSv y^{-1} for adults, children and infants, respectively) and the lowest for a tap water (Me: 4.0; 8.1, 5.6 μSv y^{-1}). It was further observed that children (7–12 y old) are the most exposed among the investigated members of the population with maximum effective doses of 19.1 and 18.7 μSv y^{-1} after drinking BM2 or BN1, respectively. Contrary to some other similar investigations in European countries[13, 18], none of the assessed doses due to drinking any bottled water produced and sold in Slovenia, or tap water measured in this survey, exceeded the dose limit of 100 μSv y^{-1}[10].

Investigation of the contribution of particular radionuclides to the assessed internal doses to different age groups drinking three different types of water showed high variability and mostly reflect the variability of radionuclide concentrations in different water sample types. As seen from Figure 3, the contribution of both uranium radionuclides, presented as the median contribution (%) with minimum and maximum values for ^{234}U and ^{238}U, respectively, to the total ingestion dose due to drinking any type of water for all three groups of populations is well <10 %. The next lowest radionuclide contribution (Me: <20 %) is ^{210}Po, where even lower median contribution levels (<10 %) for all age groups can be observed in the case of drinking BM water. For ^{226}Ra, it can be seen that the contributions to doses vary between different water samples and within each type of water for all age groups, being low (Me: <20 and 10 %) in the case of BM waters and tap water, respectively, and up to 2 8 % (median) for BN waters, with an individual contribution (max) up to 50–60 % after drinking BN7 for all three age groups. Contrary to ^{226}Ra, the proportions of one of the most radiotoxic nuclides ^{228}Ra to total ingestion doses were on average higher due to drinking BM and tap waters and were especially high (maximum up to 77 %) for children and infants (Figure 3) for all types of waters. Among the radionuclides included in dose estimates, the proportion of ^{210}Pb is on average the highest, with median levels >30 and 40 % for all age groups drinking BN and tap waters, respectively, but relatively low proportions were found for BM waters (Me <20 %).

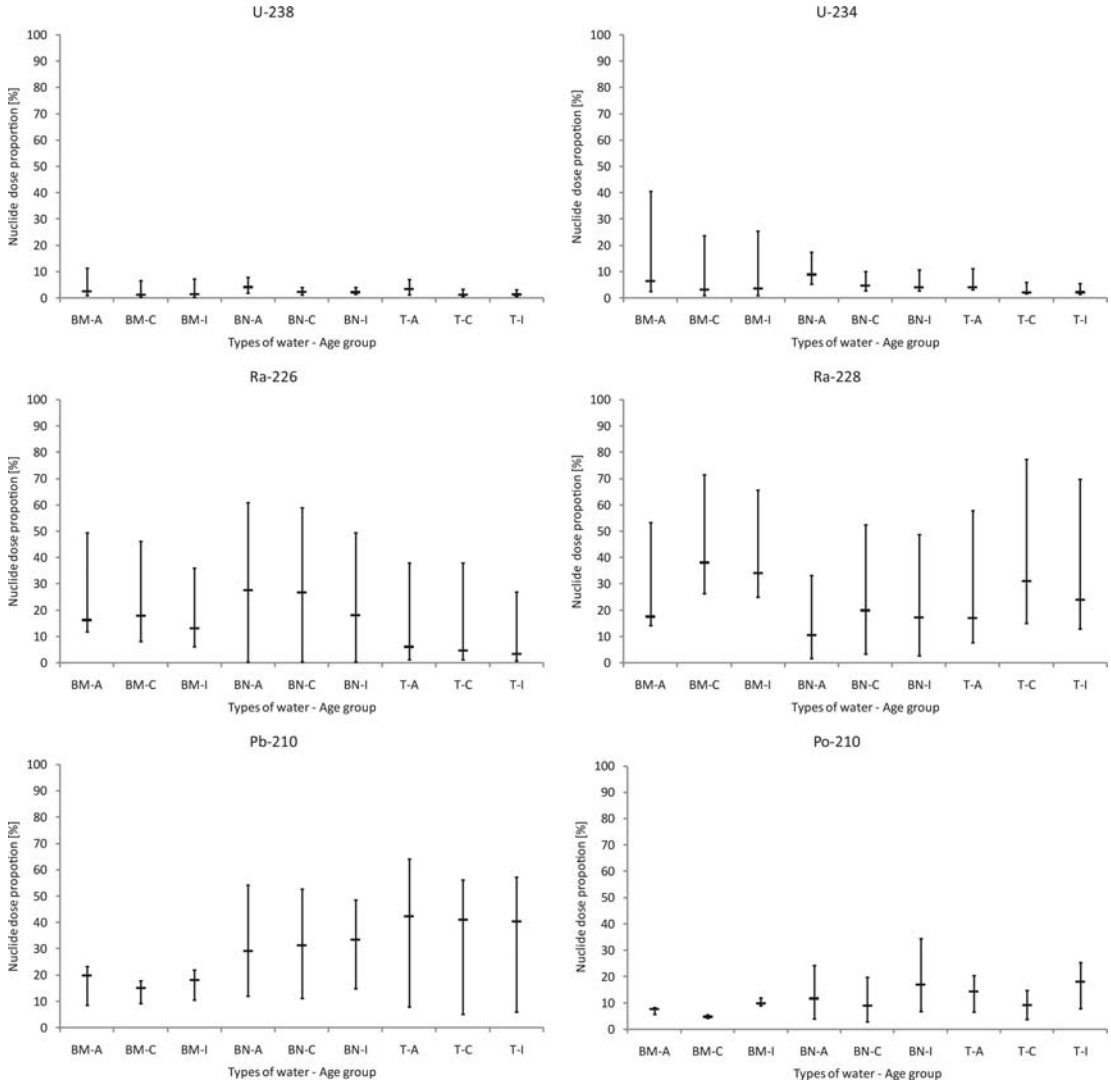

Figure 3. Proportion of radionuclide specific dose (%) due to drinking BM, natural water (BN) and tap water (T) presented as median (Me), minimum (Min) and maximum (Max) proportion level for adults (A), children (C) and infants (I).

CONCLUSIONS

Presented study was the first one in which all the BN and mineral waters produced and sold in Slovenia, together with some tap waters coming from different regions of the country were analysed for natural alpha (^{238}U, ^{234}U, ^{226}Ra, ^{210}Po) and beta (^{228}Ra, ^{210}Pb) emitting radionuclides, and the committed effective doses for different age groups were calculated. On average, the lowest proportion of the nuclide dose was found for ^{238}U and ^{234}U, followed by ^{210}Po. ^{210}Pb was the radionuclide with highest median contribution for tap water and BN water for all age groups, followed by ^{226}Ra and ^{228}Ra. However, due to its high radiotoxicity, ^{228}Ra was found to contribute much higher median as well as absolute dose proportions of the dose (>70 %) for children and infants in comparison to adults after drinking all three types of water. It can be concluded that drinking any type of BN and mineral water as well as the tap waters included in this survey, does not significantly contribute to the internal dose received by different age groups. This study further

showed that tap waters from different regions vary considerably in radionuclide activity concentrations and thus internal doses, and an extension of this survey is needed to provide additional information. There are many individual, small springs used for water supply which were not included in this survey.

The control of the quality of drinking water should be established based on the revised Directive, which suggested that the same standards as for drinking water should be applied to BM water. That is not only important for users on Slovenian market, but also of vital importance to producers, to be able to export the products to European market.

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