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Research Article

Radioactivity measurements and radiation dose evaluation in tap waters of Central Italy

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Consumption of drinking water is very important for human nutrition and its quality must be strictly controlled. A study of radioactivity content in tap water samples collected in the Central Italy was performed in order to check the compliance with recent European regulations. Gross alpha and beta activity, ^{226}Ra, ^{238}U and ^{234}U concentrations were measured. Gross alpha and beta activities were determined by standard ISO 9696 and ISO 9697; for ^{226}Ra determination liquid scintillation was used. ^{238}U and ^{234}U concentrations were determined by alpha spectrometry after separation from matrix by extraction chromatography and electroplating. Recommended WHO guideline activity concentrations for drinking water (0.1 and 1.0 Bq/L for gross alpha and gross beta activity, respectively) are exceeded in two cases for gross alpha activity and are not exceeded in any case for gross beta activity. The concentrations (mBq/L) of ^{226}Ra, ^{238}U and ^{234}U ranged from <1.70 to 15.3, 0.65 to 48.8 and 0.780 to 51.5, respectively. Effective dose due to the uranium isotopes and radium was calculated for children and adults using the dose coefficients reported by EC Directive 96/29 EURATOM and annual water intake. For all class ages, the doses are quite similar and much lower than 0.1 mSv/year.

Keywords: Dose assessment / Radioactivity / Radium / Tap water / Uranium

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1 Introduction

Due to the importance of water in human nutrition, its quality must be strictly controlled. Studies of drinking water for human consumption must be performed in order to guarantee a low-level content of radioactivity. Radiological safeguards of drinking water are based on the control of natural and anthropogenic radionuclide concentrations.

Uranium is slightly soluble in natural water; it occurs mainly as two isotopes ^{238}U and ^{234}U, with small amounts of ^{235}U. The ^{234}U concentration often exceeds that ^{238}U because ^{234}U is more easily leached from mineral surfaces [1 – 4]. Although four radium isotopes occur in natural waters, only ^{226}Ra and ^{228}Ra have sufficiently long half-lives to permit appreciable concentrations. Radium concentrations (expressed as radioactivity concentration of ^{226}Ra) are generally quite low (<1 Bq/L) [5] nevertheless its quantification is desirable due to its higher toxicity: ^{226}Ra and its progenies ^{210}Pb and ^{210}Po partially accumulate in bones leading to rather long residence times in the human body [6]. In addition, several progenies of ^{226}Ra decay by alpha particle emission which increases the committed doses from internal radiation [7].

Among the radionuclides produced by uranium and thorium, radon plays an important role: as a matter of fact it is a noble gas which can easily diffuse in the environment. Moreover, due to its poor solubility in water, it can be found in waters which are in contact with rocks having high uranium (radium) concentrations. Three radon isotopes exist, but only ^{222}Rn is generally taken into consideration as it presents a sufficiently long half time (3.82 days) and can produce an accumulation in the environment.

Natural alpha emitters are the most dangerous radionuclides when ingested [8, 9] particularly, uranium and radium compounds are very toxic for human beings.

The European Directive 98/83/EC [10] replaces the old drinking water regulation of 1980 with the aim to strengthen consumers security concerning drinking water quality. According to the European Directive, the total dose

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Abbreviation: TID, total dose indicative

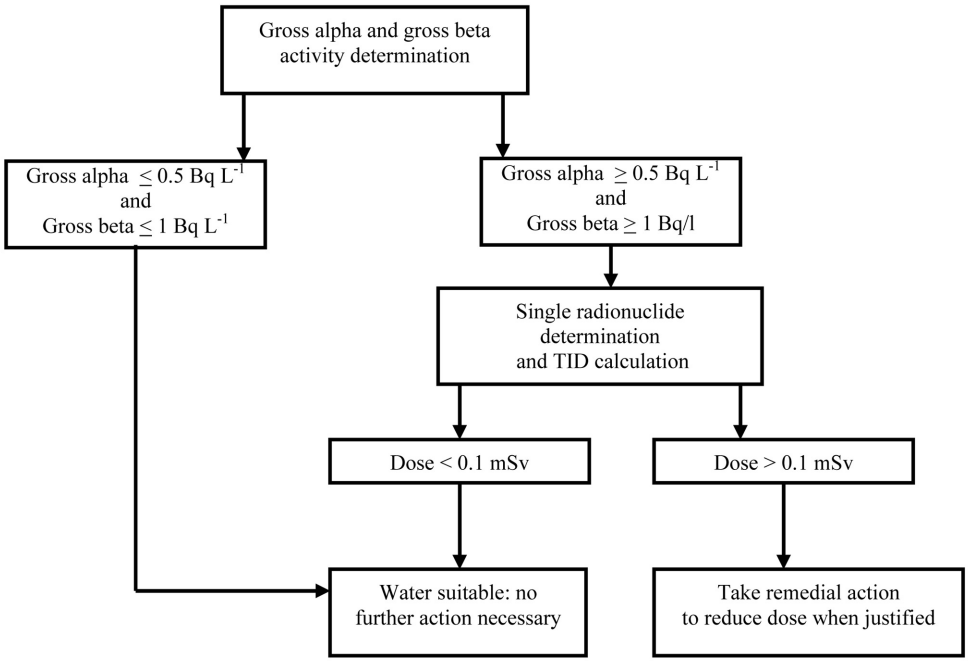

Figure 1. Application of guidelines values for radionuclides in drinking water based on the committed effective dose of 0.1 mSv/year [9].

indicative (TID) has to be determined and the parameter value of TID is 0.1 mSv/year. It is specified that TID excludes tritium, ^{40}K, radon and radon decay products; maximum concentration value for radon (100 Bq/L) is separately proposed in the European Commission Recommendation 2001/928/EURATOM [11]. European Directive 98/83/EC does not require further analyses if the estimated TID is below 0.1 mSv/year(Fig. 1).

Gross alpha and gross beta activities are determined and used for conservative TID estimation. Alpha activity is mostly due to the uranium isotopes (^{234}U, ^{235}U and ^{238}U) and to ^{226}Ra. Beta activity, to a large extent, is usually due to ^{40}K and short-lived daughters of ^{238}U (^{234}Th and ^{234m}Pa). In ‚*Guidelines for Drinking Water Quality*', 2nd edition 1993 [12], guideline values were fixed at 0.1 and 1.0 Bq/L for alpha and beta activity, respectively. In the 3rd edition published in 2004 [9], the total alpha activity value was raised to 0.5 Bq/L. In any case, a correct TID estimation presupposes the direct measure of the radionuclides that may be present in the water on the basis of general and specific knowledge regarding an aquifer.

Because today it is not clear the method of TID calculation from the gross alpha and beta activities, in this paper the radiation dose evaluation is based on the determination of ^{238}U, ^{234}U and ^{226}Ra activity concentrations.

The goal of this study was to obtain a representative estimation of radioactivity content in tap water samples collected during the summer 2006 in the Marche region (Central Italy). Moreover, the radiation dose contribution of the three radionuclides for different classes of age of the public was evaluated due to the drinking water consumption.

2 Materials and methods

2.1 Sampling

Sixty-eight tap water samples (3 L) collected drained by local Health Agency (ASUR). The samples were acidified with conc. HNO_3 to avoid precipitations, polymerizations and colloid formations.

The investigation involved the Northern part of Marche region, a district adjacent to the Republic of S. Marino, an area quite homogeneous, characterized mainly by the mountains and hills of the ‚Umbro-Marchigiana' stratigraphic series of a sedimentary marine origin. The water sample code and location of their springs can be seen in Fig. 2.

Taking into account the dry residue contents ranging from 172.08 to 662.40 mg/L, 80% of the samples were of the oligomineral class (residue from 50 to 500 mg/L), all the others were of the medium-mineral class (residue >500 mg/L).

2.2 Methods

2.2.1 Gross alpha and beta determination

The gross alpha and beta activity was determined following the thick source method [13, 14]. In this method, it is first

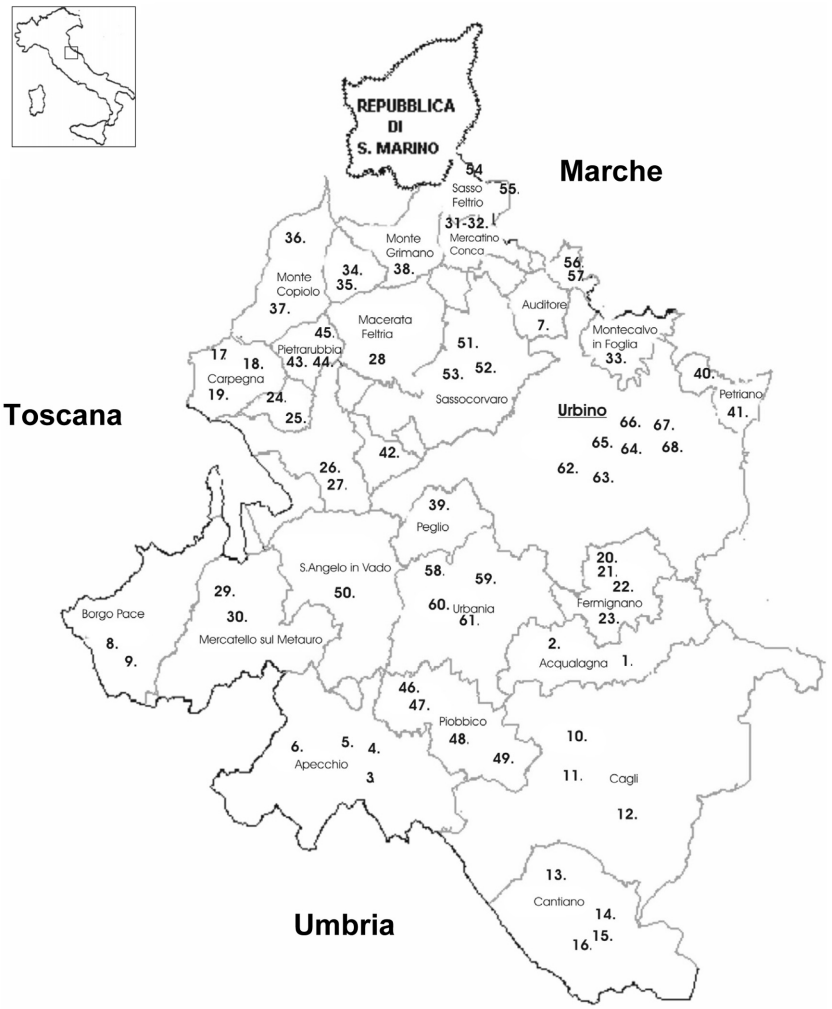

Figure 2. Sampling area in the Marche region (Central Italy).

necessary to calculate the volume of sample required to produce a mass of solid residue of approximately 200 mg. Then the sample is acidified for stabilization purpose, evaporated almost to dryness, converted to the sulphate form and ignited at 350°C. The residue is transferred to a tarred 50 mm diameter steel planchet. Sources were then counted for 80 000 s for both alpha and beta activity. Measurements were performed immediately after the preparation of the sample to minimize the influence of ^{226}Ra daughter ingrowth. Alpha activity was measured by a ZnS(Ag) detector previously calibrated against an alpha emitting source, and the beta activity was done by a low background ($6.2 \times 10^{-3}\,s^{-1}$) beta counter (Canberra-2404, USA) previously calibrated against a ^{90}Sr/^{90}Y standard source. The gross beta activities measured by this method include the ^{40}K contribution.

2.2.2 ^{238}U and ^{234}U isotopes determination

Uranium activity concentration was determined by alpha spectrometry after separation from the matrix [15–17].

Concentrated HNO_3 was added to 150 mL of water sample until the concentration reached 2 M. After the addition of ^{236}U known activity as the standard internal yield and the heating, the solution was passed through a preconditioned polyethylene microporous-trioctylphosphine oxide column which allows a chemical separation. After washing with 2 M HNO_3, 1 M HCl to eliminate thorium and few millilitres of water, uranium was eluted with 1 M $(NH_4)_2CO_3$ solution.

First the elution solution was dried and mineralized and the residue was dissolved in conc. H_2SO_4 and transferred into an electrolytic cell. Then uranium isotopes were electroplated from ammonium sulphate solution at pH 4 and measured by an alpha spectrometry system equipped with silicon detectors (Canberra) for 86 000 s. The mean counting efficiency of the instrument was 31.7 ± 3.1% with a background of $2 \times 10^{-6}\,s^{-1}$ in the energy region of interest. Taking into account the mean chemical yield of 61.0%, the MDA of the method was 0.06 mBq/L for both ^{238}U and ^{234}U.

2.2.3 ^{226}Ra determination

^{226}Ra activity concentration was determined by measuring its daughter ^{222}Rn after reaching the equilibrium with his mother (about 30 days). In the procedure, radon, being a noble gas, is selectively extracted by scintillation cocktail immiscible with water, whereas all the other radioisotopes, present in ionic form, are not extracted [18 – 22]. The extracted alpha and beta emitters of radon together with decay products in radioactive equilibrium were measured by a liquid scintillation counter (LSC). The sample preparation and counting procedure are described as following.

After addition of conc. HNO_3 until pH 2.7 ± 0.3, 200 mL of water samples were evaporated to 15 mL. Ten milliliters of this solution was transferred into a polyethylene vial containing 10 mL of scintillator cocktail (Optiscint, Wallac – Perkin Elmer); the internal surface of the vial was coated with Teflon which is impermeable to radon. After closing, the vial was shaken for 1 min, and the date was registered. After 30 days, one counting was performed for about 18 h using a low background LSC (Guardian-1414, Wallac). The MDA was 1.70 mBq/L.

3 Results and discussion

3.1 Alpha and beta activity

In Fig. 3, the gross alpha and gross beta activities are reported. Gross alpha values range from <18.2 to 128.2 mBq/L, gross beta values range from <41.6 to 258.6 mBq/L. The guideline values, fixed at 0.1 and 1.0 Bq/L for alpha and beta activity, respectively, in *Guidelines for Drinking Water Quality*, 2nd edition, 1993 [12], are exceeded in two cases for gross alpha activity and are not exceeded in any case for gross beta activity, whereas the guideline value fixed in the 3rd edition published in 2004 [9] at 0.5 Bq/L for alpha activity is not exceeded.

3.2 Radionuclide concentrations

Table 1 shows the ^{238}U, ^{234}U and ^{226}Ra concentrations.

As far as uranium isotopes activity concentrations is concerned, the arithmetical mean was 9.52 ± 10.9 and 12.8 ± 12.7 mBq/L for ^{238}U and ^{234}U, respectively.

The minimum ^{238}U concentration was 0.65 mBq/L and the maximum was 48.8 mBq/L. The minimum ^{234}U concentration value was 0.78 mBq/Land the maximum was 51.5 mBq/L. Samples (77.9%) show a ^{234}U/^{238}U activity ratio >1 with an arithmetical mean value of 1.45 ± 0.58. The lack of uranium isotopic equilibrium in waters is a well-known phenomenon: ^{234}U recoil and crystal damage and leaching are the main mechanism for the ^{234}U/^{238}U disequilibrium in ground water [3, 4]. These results show that the calculation of total uranium from ^{238}U concentration under equilibrium hypothesis can lead to a significant underestimation.

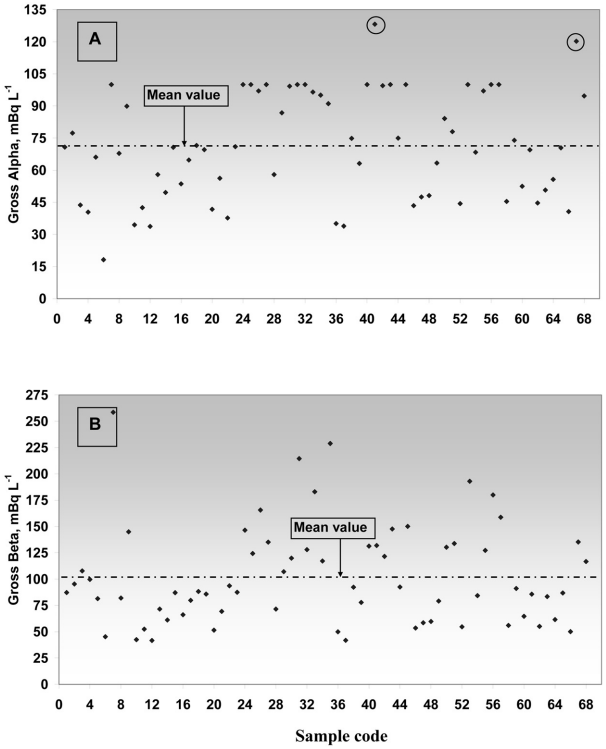

Figure 3. (A) Gross alpha and (B) gross beta activity.

The arithmetical mean of radium activity concentration resulted 2.13 ± 1.73 mBq/L. Its minimum concentration was <1.70 mBq/L and the maximum was 15.3 mBq/L.

In 88.2% of the samples $^{234+238}$U concentration is <50.0 mBq/L, 75.0% present a ^{226}Ra concentration <1.70 mBq/L (MDA).

Uranium and radium concentrations seem to be poorly correlated.

Figure 4 shows the ^{238}U, ^{234}U and ^{226}Ra concentrations in the two samples (nos. 41 and 67) with a gross alpha activity higher than 100 mBq/L seeing the ^{226}Ra concentration representing 1.68 and 1.82% of the sum of the three alpha emitters, respectively. It confirms that uranium isotopes represent the principal source of radioactivity in water, nevertheless ^{226}Ra determination is desirable due to its higher radiotoxicity.

3.3 Radiation dose assessment

For every exposure group, the radiation dose (E, Sv) contributed from each radionuclide *via* ingestion of water was calculated based on the following equation:

$$E = K \cdot G \cdot C \cdot T$$

where K is the dose conversion factor (SvB/q) of the specific radionuclide as shown in Table 2 [23, 24]; G is the

Table 1. ^{238}U, ^{234}U and ^{226}Ra concentration (mBq/L)

Sample code	^{238}U	^{234}U	^{226}Ra	Sample code	^{238}U	^{234}U	^{226}Ra
1	3.85 ± 0.58	5.32 ± 0.80	15.31 ± 1.04	35	11.1 ± 1.66	7.98 ± 1.20	1.74 ± 0.67
2	3.66 ± 0.55	3.17 ± 0.48	<1.70	36	1.41 ± 0.21	1.64 ± 0.25	<1.70
3	8.09 ± 1.21	7.71 ± 1.16	4.71 ± 1.06	37	4.09 ± 0.61	4.32 ± 0.65	<1.70
4	6.76 ± 1.01	7.70 ± 1.16	<1.70	38	8.60 ± 1.29	14.0 ± 2.10	<1.70
5	3.26 ± 0.49	6.07 ± 0.91	<1.70	39	2.32 ± 0.35	4.90 ± 0.74	<1.70
6	0.73 ± 0.11	3.15 ± 0.47	<1.70	40	32.7 ± 4.90	35.2 ± 5.28	1.72 ± 0.67
7	17.6 ± 2.64	28.9 ± 4.33	<1.70	41	48.1 ± 7.21	51.5 ± 7.73	<1.70
8	5.03 ± 0.75	7.55 ± 1.13	3.94 ± 0.76	42	11.0 ± 1.64	21.1 ± 3.29	<1.70
9	6.70 ± 1.01	14.4 ± 2.16	2.58 ± 0.58	43	14.3 ± 2.15	19.9 ± 2.99	1.98 ± 0.56
10	0.72 ± 0.11	1.07 ± 0.16	<1.70	44	8.70 ± 1.31	16.5 ± 2.48	<1.70
11	1.33 ± 0.20	3.04 ± 0.46	<1.70	45	12.1 ± 1.81	28.1 ± 4.22	2.01 ± 0.78
12	0.87 ± 0.13	0.78 ± 0.12	<1.70	46	3.10 ± 0.47	2.79 ± 0.42	3.13 ± 0.60
13	1.67 ± 0.25	2.92 ± 0.44	<1.70	47	1.47 ± 0.22	1.10 ± 0.17	3.49 ± 0.59
14	2.86 ± 0.43	3.05 ± 0.46	<1.70	48	2.37 ± 0.36	3.72 ± 0.56	<1.70
15	2.60 ± 0.39	3.47 ± 0.52	<1.70	49	3.38 ± 0.51	2.66 ± 0.40	<1.70
16	0.65 ± 0.10	1.13 ± 0.17	<1.70	50	8.56 ± 1.28	15.8 ± 2.37	<1.70
17	4.83 ± 0.72	9.67 ± 1.45	<1.70	51	21.2 ± 3.18	27.8 ± 4.17	<1.70
18	3.85 ± 0.58	8.74 ± 1.31	2.11 ± 0.59	52	2.25 ± 0.34	2.07 ± 0.31	<1.70
19	3.99 ± 0.60	8.72 ± 1.31	<1.70	53	21.0 ± 3.15	31.4 ± 4.72	<1.70
20	1.74 ± 0.26	1.74 ± 0.26	<1.70	54	5.77 ± 0.87	9.49 ± 1.42	2.68 ± 0.61
21	3.57 ± 0.54	4.89 ± 0.73	<1.70	55	11.7 ± 1.75	22.1 ± 3.32	<1.70
22	1.03 ± 0.15	1.20 ± 0.18	<1.70	56	25.6 ± 3.84	31.3 ± 4.70	<1.70
23	1.68 ± 0.25	3.71 ± 0.56	<1.70	57	27.7 ± 4.15	38.7 ± 5.81	3.07 ± 0.59
24	8.08 ± 1.21	13.6 ± 2.04	<1.70	58	3.05 ± 0.46	1.20 ± 0.18	1.89 ± 0.43
25	6.6 ± 0.99	8.39 ± 1.26	<1.70	59	10.1 ± 1.52	10.1 ± 1.52	<1.70
26	12.1 ± 1.82	15.5 ± 2.33	<1.70	60	3.00 ± 0.45	2.44 ± 0.37	<1.70
27	16.6 ± 2.50	23.1 ± 3.47	<1.70	61	9.31 ± 1.40	9.93 ± 1.49	<1.70
28	6.34 ± 0.95	10.9 ± 1.64	<1.70	62	1.48 ± 0.22	1.99 ± 0.30	<1.70
29	13.2 ± 1.98	18.2 ± 2.73	<1.70	63	4.05 ± 0.61	3.93 ± 0.59	<1.70
30	13.6 ± 2.05	16.0 ± 2.39	3.44 ± 0.53	64	1.42 ± 0.21	3.04 ± 0.46	2.45 ± 0.47
31	20.1 ± 3.02	35.7 ± 5.36	<1.70	65	10.6 ± 1.59	13.68 ± 2.05	<1.70
32	17.98 ± 2.70	29.21 ± 4.38	<1.70	66	1.32 ± 0.20	0.88 ± 0.13	<1.70
33	9.52 ± 1.43	15.41 ± 2.31	<1.70	67	47.7 ± 7.15	43.80 ± 6.57	<1.70
34	7.23 ± 1.08	9.83 ± 1.47	<1.70	68	48.8 ± 7.32	50.91 ± 7.64	2.09 ± 0.59

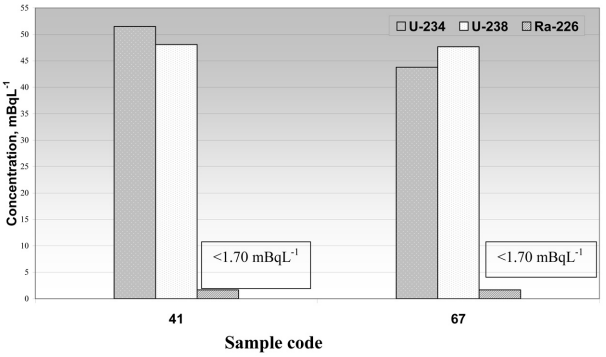

Figure 4. ^{238}U, ^{234}U and ^{226}Ra concentrations for the two samples with a gross alpha activity higher than 100 mBq/L.

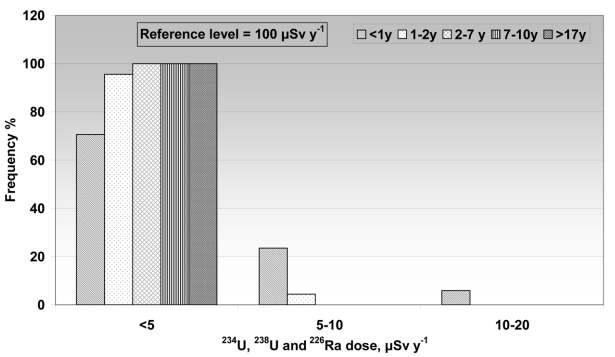

Figure 5. Three total dose classes for the different age groups.

water consumption rate (L/day) for each age group and the corresponding values for the age of <1, 1–2, 2–7, 7–10 and >17 years are 0.7, 1, 1, 1 and 2; *C* is the concentration of the specific radionuclide in water (Bq/L) and, for radionuclide concentrations below the detection limit, the MDA was considered; and *T* is the duration of consumption, here 365 days.

Figure 5 reports the radiation dose relating to ^{234}U, ^{238}U and ^{226}Ra for different age groups. None of the samples exceeds the reference level of committed effective dose (0.1 mSv/year) for drinking water. For children (>1 year) and adults, the calculated doses are quite similar and range from 0.40 to 5.79 µSv/year. For babies (<1 year), the doses are four times higher and range from 2.19 to 19.2 µSv/year.

Table 2. Dose conversion factors, *K* (SvB/q) [23, 24]

Radionuclide	Age classes				
	<1 year *K*	1–2 years *K*	2–7 years *K*	7–10 years *K*	>17 years *K*
^{238}U	3.4×10^{-7}	1.2×10^{-7}	8.0×10^{-8}	6.8×10^{-8}	4.5×10^{-8}
^{234}U	3.7×10^{-7}	1.3×10^{-7}	8.8×10^{-8}	7.4×10^{-8}	4.9×10^{-8}
^{226}Ra	4.7×10^{-6}	9.6×10^{-7}	6.2×10^{-7}	8.0×10^{-7}	2.8×10^{-7}

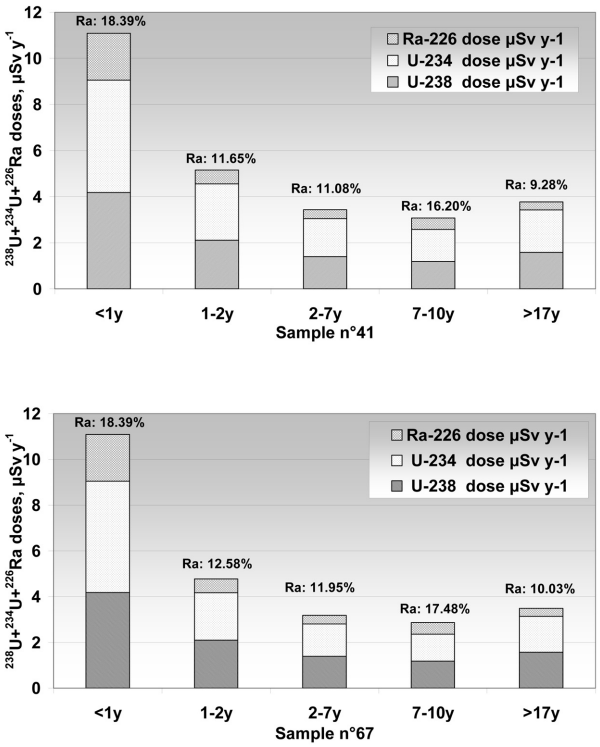

Figure 6. Contribution percent of ^{238}U, ^{234}U and ^{226}Ra to TID for the different classes of age in the sample nos. 41 and 67.

Figure 6 shows the contribution to TID of the radionuclides measured (^{234}U, ^{238}U and ^{226}Ra) in sample nos. 41 and 67 for the different classes of age. The ^{226}Ra contribution percent to the TID presents a maximum value of 20% for babies and a minimum of 10% for adults.

4 Concluding remarks

As the drinking water consumption is very important for people, we have carried out a radioactivity measurement in 68 tap water samples collected in a region of Central Italy to check the compliance with recent European and Italian regulations. Gross alpha and gross beta, ^{234}U, ^{238}U and ^{226}Ra activity concentrations for drinking water were determined and the found activity concentrations in all examined samples are under control if compared with guideline recommended by WHO.

^{238}U and ^{234}U activity concentration ranges from 0.65 to 48.8 and from <0.78 to 51.5 mBq/L, respectively. Moreover, 78% of samples exhibit an isotopic ratio higher than 1.

^{226}Ra is usually present in lower quantity than uranium and its activity concentration ranges from <1.70 to 15.4 mBq/L. In 88.2% of samples, its concentration is lower than 10.00 mBq/L(MDA).

Taking into account the total radiation dose contributed from $^{238}U + ^{234}U$ and ^{226}Ra, all samples furnish a dose lower than the reference level of committed effective dose for drinking water (0.1 mSv/year). For children (>1) and adults, the calculated doses are quite similar and ranges from 0.40 to 5.79 µSv/year. For babies (<1 year), the doses are four times higher and range from 2.19 to 19.2 µSv/year.

It is worth mentioning that the real value of the total dose should be calculated by determining all the radionuclides in water, with special regard to radium isotopes, which are very toxic for human beings especially for babies. In fact, the derived limits for gross alpha and beta activities, intended to assure compliance to the reference dose limit of 0.1 mSv/year, were derived from activity dose conversion factors for adults which are lower than those for infants.

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