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Natural radioactivity of bottled mineral waters of southern Poland

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Abstract Activities of ^{238}U, ^{234}U, $^{(226228)}Ra$, Ra, and ^{224}Ra as well as total α and β activities of 23 bottled mineral waters of south Poland are presented. The activities vary from a few tenths to a few mBq dm^{-3} for uranium and to several hundred mBq dm^{-3} for radium isotopes. The activities of ^{40}K were calculated from chemical analyses of potassium and checked for several mineral waters by gamma spectrometry with HPGe detector. Correlations between water mineralisation and activities of ^{40}K, ^{226}Ra, total alpha and total beta are observed. The radiological annual doses are calculated for all investigated waters assuming the consumption of 2 L per day.

1. Introduction

More than 10 years ago in Poland mineral water was only in special health resorts available available and a few shops, but nowadays it is to the whole population. The mineral water industry is growing rapidly both in terms of the volume of water produced and the variety of water types. The reason for this rapid growth is the fact that mineral waters contain significant quantities of health beneficial ions such as magnesium, iron, calcium, etc., which in many cases have properties.medicinal However, "there are two sides to every coin", with the increase in mineralisation, the content of natural radioactive elements such as radium, uranium, polonium, etc. also increases. These isotopes not only have an adverse effect biochemically, but also radiologically. For this reason, the International Health Organisation (WHO, 1998) has issued a recommendation that a given water is fit for consumption if the annual dose induced by drinking it at a rate of 2 litres/day not would exceed 0.1 mSv. In practice, the rule of thumb is that a given water is fit for consumption if its global alpha and beta activities did not exceed 0.1 and 1 Bq dmrespectively $^{-3}$ (WHO, 1998).

There are many publications on natural radioactive in isotopes bottled mineral waters, but each is devoted to only a partial problem related to the issue at hand. In the works of Sajo- Bohus et al. (1996); Sanchez et al. (1998); Surbeck (1995); Rangiel et al.(2002); Kitto et al. (2005) were concerned with the study of global alpha and beta in radiation activity bottled mineral waters sold in , respectively. Venezuela, Mexico, Spain, France, Portugal and the United StatesIn prach Bonotto (2003); Marović et al. (1996); Kralik (2003); Oliveira (2000); Laura & Godoy (2002); Somlai et al. (2002) measured radium isotope contents (^{228}Ra and ^{226}Ra) in waters sold in South America, Croatia, Spain, Austria, Brazil and Hungary, respectively. Two papers on natural in radioactivity some bottled mineral waters (Melnikov et al. 2000; Skwarzec et al. 2003). have been in publishedPoland The first paper paper discussed discussed radium isotopes, and the second uranium and polonium , from isotopeswhich the annual radiological doses received by were estimated. ingestion of these waters The present work not only extends the scope of the study of the radioactivity parameters of waters, but also undertakes a study of newly produced bottled mineral waters and, in particular, addresses the following issues:

- determination of global alpha/beta , radioactivityisotope content ^{238}U, ^{234}U, $^{228)}$Ra, ^{226}Ra, $^{224)}$Ra and ^{40}K;
- investigating the correlation between the contents of the aforementioned isotopes and water mineralisation;
- calculation of radiological doses received by ingesting the waters .tested

2. Measurements

A minimum of 9 litres of test water was used for the measurements. Depending on the mineralisation, a different amount of water was allocated for the analysis of the isotopes of the element in question, e.g. when the mineralisation was below 1,000 mg dm^{-3}, the amount of water needed for the analysis of radium and uranium isotopes was between 3 and 5 litres, respectively, while above ,000 1the amount of water could be between 1 mg dm^{-3} litre litres; the remaining water was allocated for the measurement of global alpha and beta radioactivity. The methods of analysis used in the work are briefly described below.and 3

2.1. Total alpha and beta radiation activities

A 1 to 3 litre sample of water was heated at about 80° C until completely evaporated. The remaining precipitate was transferred to a measuring vessel, mixed with 6ml of distilled water and 12ml of liquid scintillator in a gel, noting the time of mixing. The sample was then measured with an counter α/β with liquid scintillator (L'Annunziata et al., 2003). In these measurements, care must be taken to establish the correct value of the PSA parameter (a parameter that allows the counter to correctly separate pulses from alpha radiation from pulses from beta particles). To be able to eliminate the contribution of radon and its decay products from radium in the sample, the measurement was repeated daily for one week. The duration of each measurement was planned so that the relative uncertainty of the measurement did not exceed 5%. On the basis of results, curves were prepared of the dependence of the measured intensity including background (net) alpha and beta radiation on time. The global intensities of alpha and beta radiation when the sample was mixed with the liquid scintillator were obtained by extrapolating the aforementioned curves. An example of this procedure is shown in Figure 1.

2.2. Radium isotopes

A sample volume (2 to 3 l) was evaporated to about 1 l, then radium isotopes were precipitated together with barium in the form of sulphate. In order to purify the precipitate from other interfering isotopes such as lead, polonium, etc., the precipitate was washed repeatedly with distilled water up to a neutral reaction and dissolved in an alkaline EDTA solution of 0.25 M. The pH of the solution was then lowered to 4.5 by adding acetic acid, in this way radium and barium were again precipitated in the form of sulphate, while uranium and lead remain in solution (Tomza, 1975). After washing with distilled water, the precipitate was transferred to a measuring vessel and mixed with 12 ml of liquid scintillator in gel, the sample was measured with an counterα/β . The time instant of precipitation must be recorded to account for the build-up of radon and its products after time. Measurements were taken daily for at least 21 days. From the results measured the after time, radium isotope contents (^{228}Ra, ^{226}Ra and ^{224}Ra) were determined (Chau et al., 1997).

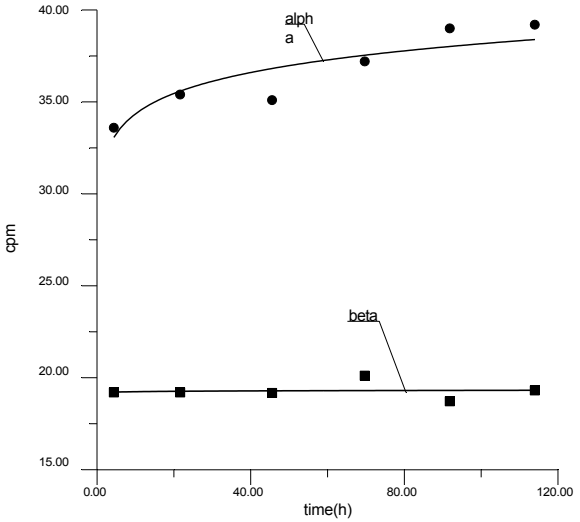

Figure 1. typical dependence measured pulse intensities from alpha and beta radiation emitted from of water sample on time *The of the remainder the evaporatedFigure 1. The typical dependence of measured count rates of total alpha and beta activities of the remainder of the evaporated water sample on time*

2.3. Uranium isotopes

To a sample of at least 3 l, a known amount of tracer uranium of about 0.1 was added Bq ^{232}U and evaporated to a volume of 0.5 l, then uranium was precipitated as $(NH_4)_2)U_{(7\ 22}$Otogether with $MnO_{()}$. The precipitate was washed with distilled water and dissolved in a 9 solutionM HCl . The solution was then passed through a 200-400 Dowex cationite columnmesh . The uranium was eluted from the column with a solution of 0.1 M HCl. The eluate was evaporated to dryness, the residue was dissolved in a solution of 1 M HCl. The uranium was re-precipitated by the addition of neodymium chloride, Mohr's salt and HF acid. The precipitated uranium was deposited on a membrane filter with a porosity of 100 nm. After drying, the uranium filter was measured with a silicon solid-state counter alpha spectrometer (Mietelski 2003; Skwarzec et al., 2001).

2.4. Isotope ^{40}K

Potassium is an alkaline metal located in the first group of the periodic table, so its chemical precipitation from a water sample is difficult. The isotope ^{40}K is a natural beta radioactive isotope. When decaying, this isotope also emits 11 % gamma quanta with an energy of 1464 keV (Lederer et al., 1967). Potassium 40 can therefore be determined by gamma spectrometry. In the work of Jodłowski (2005), the found to detection threshold of potassium 40 by gamma spectrometry an semiconductor detector withHPGe using a large Marrinelli measuring vessel with a volume equal to 0.78 l was be 14 mg $dm^{(-3)\ of}$ total potassium, assuming that the abundance of potassium 40 is 0.012 %. For gamma-ray spectrometric measurements

water should be evaporated so that the total potassium content per litre is 2 to 3 greater than the detection threshold given above. for determination of Several mineral waters with total potassium concentrations ranging from 8 mg to 52 mg . Based on the measured content of the isotope were selected ^{40}K by spectrometrygamma ^{40}K and its abundance table value (0.012 %), the total potassium content per litre of water was converted. The resulting total potassium values and its values provided by the water manufacturer, as well as the measured specific activities of the ^{40}K isotope for several selected mineral waters, are summarised in Table 1.

Table 1. Total potassium concentrations by manufacturer, measured specific the isotope activities of ^{40}K and converted potassium concentrations in some mineral waters

Table 1. Label concentrations of potassium, determined contents of ^{40}K and calculated values of potassium in the several mineral waters

Name of the water	Potassium according to the manufacturercontent [mg dm^{-3}].	Assayed activity ^{40}K [mBq dm^{-3}]	Recalculated potassium content [mg dm^{-3}].
Staropolanka	52	1364± 150	43.3± 4.8
Muszynianka	10,1	290± 50	9.2± 1.6
Aquarel	8,75	223± 31	7.1± 1.0
R8	48,5	1368± 150	43.4± 4.8

It can be seen from this table that all converted total potassium values are slightly lower than the manufacturer's values. This fact could be due to the partial loss of potassium through evaporation, adsorption of potassium on the walls of the vessels and the transfer of the sample from the glass beakers to the measuring vessel. For these reasons, it was decided to convert the isotope content of ^{40}K in the mineral waters from the amounts of total potassium determined by chemical analysis and the abundance value of - 0.012 %. The calculated specific activities of the isotope ^{40}K for the investigated waters are summarised in Table 2.

3. Measurement results and discussion

The results of the radiometric analysis for the 23 bottled mineral waters and their ionic as reported on the bottle labels are presented in 2 and composition Tables(TDS) 3. Due to the lack of uncertainty in the ionic composition reported on the bottle labels, so the authors of this paper were not able to estimate this uncertainty on the total dissolved content of the solids waters . studiedThe results of the radium isotope analysis contained in this paper are generally consistent with the results of the analysis in the work of Melnikov et al. (2000).

Table 2. Total alpha and beta radiation activities and uranium, radium and potassium the bottled mineral waters tested.isotope contents of

Table 2. Total alpha, beta activities and contents of uranium, radium and potassium isotopes in the investigated bottled mineral waters.

Name of the mineral water	Global activity [mBq dm^{-3}].		Radium isotope activities [mBq dm^{-3}]			Uranium [mBq dmisotope activities $^{-3}$].		Isotope activity [mBqdm^{-3}].
	Alfa	beta	^{224}Ra	^{226}Ra	^{228}Ra	^{328}U	^{324}U	^{40}K
Żywiec	2.2± 0.6	53 8±	≤0,5	1.3± 0.4	29 3±	0.61± 0.09	1.66± 0.15	–
Dobrawa	139± 16	67 7±	≤0,5	66 7±	10 3±	5.1± 0.5	22.8± 1.2	26,7
Staropolanka	592± 60	1784± 200	≤0,5	525± 30	276± 20	7,68±	16,94	1364± 150*
Krynicznka	96 6±	233± 11	≤0,5	73 6±	96 9±	2.86± 0.27	3.51± 0.29	211
Nałęczowianka	16 3±	83 9±	≤0,5	2.4± 0.4	16 2±	0.81± 0.09	1.19± 0.11	170
Muszynianka	55 7±	367± 32	≤0,5	44 5±	45 7±	4.66± 0.30	10.22± 0.44	290± 50*
Multivital	86 8±	140± 10	≤0,5	8.0± 1.0	14 2±	19.2± 3.5	74.7± 10.2	37,8
Laguna	23 4±	132± 12	≤0,5	22 3±	8.0± 1.0	10.04± 0.42	12.13± 0.47	–
Muszyna	206± 14	184± 12	≤0,5	47 5±	45 7±	1.72± 0.29	4.67± 0.48	170
Piwniczanka	155± 15	592± 39	≤0,5	57 5±	39 4±	2.20± 0.20	2.09± 0.20	563
Kropla Beskidu	11 2±	88 8±	≤0,5	4.0± 0.6	5.0± 1.0	2.8± 0.4	6.4± 0.7	48,5
Ustronianka	100± 10	161± 14	≤0,5	16 2±	≤1,0	10.54± 0.76	16.92± 1.22	–
Kinga	9,06	160	≤0,5	3.16± 0.38	5.33± 0.71	4.43± 0.4	7.79± 0.56	72,8
Jurassic	At	on	≤0,5	12 3±	≤10	1.34± 0.13	7.42± 0.34	78,8
Aquarel	100± 10	212± 18	≤0,5	17.8± 1.6	19.3± 2.0	0.57± 0.08	0.75± 0.09	223± 31*
Oasis	18,33	86,82	≤0,5	0.8± 0.1	8.2± 1.1	6.79± 0.47	10.82± 0.63	15,7
Polaris	40,37	406	≤0,5	20.0± 1.4	11.8± 1.1	4.23± 0.42	4.47± 0.42	92,6
Jan	18,63	474	≤0,5	9.3± 1.0	9.2 1±	3.94± 0.61	5.36± 0.51	113,7
Zuber	938± 104	10537 470±	18.5± 2.5	437± 26	393± 25	2.30± 0.60	3.07± 0.70	10064
Kazimierka	on	on	≤0,5	0.8± 0.1	12.5± 1.7	5.0± 0.48	9.51± 0.70	126
Hermes	19.7± 2.3	87.2± 7.6	≤0,5	3.78± 0.44	17.74± 2.30	5.1± 0.4	7.1± 0.4	42,5
Wielka Pieniawa	625± 51	1062± 70	≤0,5	249± 16	169± 12	4.4± 1.2	19.0± 2.7	1069± 150
Mazovia	on	on	≤0,5	1.38± 0.16	2.1± 0.28	on	on	236,4

* Measured isotope content of ^{40}K by gamma-ray . spectrometryon - not analysed

Table 3. Ionic composition of bottled mineral waters collected from labels
Table 3. Ion composition of the investigated bottled mineral waters after bottle labels

Name of the water	Content of individual ions [mgdm^{-3}].										
	Ca^{+2}	Mg^{+2}	Na^{+1}	K^{+1}	Fe^{+3}	Li^{+1}	HCO_3^{-1}	SO_4^{-2}	Cl^{-1}	F^{-1}	TDS*
Żywiec	27,75	8,18	8,00	nd	nd	nd	109,00	nd	4,60	0,07	185,84
Dobrawa	55,2	29,6	1,2	0,85	nd	nd	286,8	20,0	5,32	0,1	402
Staropolanka	355,1	77,3	135	52	nd	nd	1864,7	24,0	8,9	0,5	2517,5
Kryniczanka	547,9	113,48	67,3	6,7	nd	nd	2172,24	9,64	10,64	0,14	2927,63
Nałęczowianka	114,5	23,1	12,7	5,4	nd	nd	495,8	nd	8,5	0,3	696,3
Muszyna	191,8	128,7	93,2	10,1	nd	nd	1458,5	31,1	13,9	nd	1927,3
Multivital	164	25,3	5,4	1,2	nd	nd	597,0	27,1	2	nd	822
Laguna	71,56	25,56	5,8	nd	nd	nd	290,75	20,6	12,85	nd	427,12
Muszyna	482,59	51,94	47,2	6,48	nd	nd	1854,95	2,96	5,32	0,41	2451,85
Piwniczna	235	121	nd	17,9	0,2	0,34	1898	29,4	12,4	0,25	2314,49
Kropla Beskidu	46,49	19,46	37,72	1,54	nd	nd	298,94	30,28	1,77	nd	434,2
Ustronianka	78,56	15,56	5,8	nd	nd	nd	290,75	20,6	12,05	nd	423,32
Kinga	97,8	13,13	4,59	2,31	nd	nd	335,6	28,5	7	0,05	488,98
Jurassic	67,1	36,5	9,0	2,5	nd	nd	345,6	37,7	8,5	0,3	507,2
Aquarel	44,1	15,8	60	8,75	nd	nd	336,5	nd	28	0,5	519,45
Oasis	52,10	7,3	2,50	0,50	nd	nd	170,80	23,04	4,80	0,10	280,58
Polaris	102,8	16,00	11,25	2,94	nd	nd	432,7	nd	2,8	0,23	592,32
Jan	152,3	24,81	15,42	3,61	0,07	0,01	524,75	51,34	17,73	0,18	790,22
Zuber**	132,02	331,32	6495,	320	0,99	12,49	18549,47	49,51	975,08	0,4	25899,07
Kazimierka	87,68	28,86	10,0	4,0	nd	nd	387,5	35,18	7,40	0,5	561,12
Hermes	109,02	17,02	10,0	1,35	nd	nd	213,9	117,28	35,5	0,2	504,27
Wielka Pieniawa	232,1	26,1	68,5	38,7	nd	nd	1031,2	29,4	7,1	0,3	1394,7
Mazovia	38,08	15,55	95,00	7,50	nd	nd	322,50	nd	64,2	0,47	543,3

* Due to the uncertainty in lack of the ionic composition reported on the bottle , labelsso the authors of this paper not able to estimate the uncertainty on TDS

** "Zuber" water is a healing water and still contains bromine - ions 6.39 mg, iodine - 1.48 mg

nd - no data for the ion in in in the question or the question element is not present water .tested

Tables 2 and 3 show that natural radioactive isotope contents and ionic composition vary over wide ranges. For example, radium 226 and radium 228 isotope contents range from almost 1 mBq dm^{-3} to more than 500 mBq dm^{-3}, for uranium isotopes from about a fraction of mBq dm^{-3} - $^{3(}$to several mBq dm, and total mineralisation varies from about 200 mg $dm^{-3)-3-3)}$to almost 3000 mg mg dm, and exceptionally in one water mineralisation reached almost 26000 $dm^{(}$.

Bottled mineral waters contain almost no radium 224; this fact can be explained by the fact that the time between the extraction of the water and the day of sale takes more than a month, after which time the radium 224 isotope has already completely decayed ($T_{1/2}$ = 3.64 days). from It can also be seen that, in some waters, the potassium 40 isotope content exceeds more than half of the global beta activity.Table 2

Table 4 shows the values of linear correlation parameters between the analysed radioactivity and mineralisation of bottled mineral waters. This table shows that there are some linear correlations between global alpha, beta radioactivity and radium, potassium isotope contents and water mineralisation.

Table 4. parameters of linear correlation and their (a,b and u(a),u(b)) between *The values of the the uncertainties radioactivities The values of the parameters and their uncertainties (a,b and u(a), u(b)) of linear correlation between natural radioactivities and TDS of bottled mineral waters,*natural and mineralisation of bottled mineral waters *Table 4.*

Radioactivity	a± u(a)	b± u(b)	R
Integ. Activ. rad. α	0.034± 0.006	83± 33	0,61
Integ. Active. rad. β	0.41± 0.02	-90± 72	0,97
Content ^{226}Ra	0.017± 0.002	5.5± 5.1	0,97
Content ^{40}K	0.39± 0.03	156± 232	0,98

Assuming that each adult drinks an average of 2 litres of water per day, the annual radiological dose from the absorption of uranium and radium isotopes contained in a given water is calculated according to the formula (WHO, 1998):

$$D(mSv) = 730 \cdot \sum \eta_{i} \cdot W(i),$$

where: η_i - conversion factor (mSv/Bq) for the i-th isotope, values for this factor for different isotopes can be found in the paper (IAEA 1996); W_i - content of *the i-th* isotope in one litre of water [Bq dm^{-3}].

The calculated annual doses by absorption of the radium and uranium isotopes contained in the study waters are shown in Figure 2; the annual dose is also plotted in this figurelimit of 0.1 mSv recommended by the WHO . From this figure, it can be seen that the annual doses of some waters exceed the dose limit for adults, so it is not recommended to consume 2 litres of these waters . per dayAlthough the isotope ^{40}K in some waters has a significant contribution to global beta radiation activity, the dose through its absorption is insignificant compared to the dose received from other isotopes (e.g. for the

of water with a concentration of ^{40}K of 563 mBq dm^{-3}, the annual dose through the absorption of this isotope is 0.0026 mSv), so that a limit on global radiation activity beta should be included along with the contribution of, for example, the .^{40}K isotope

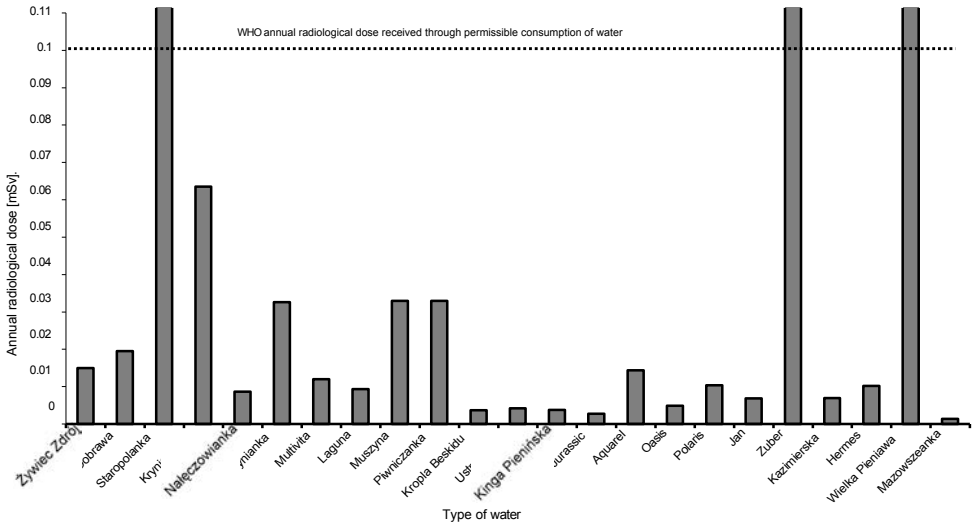

Figure 2. Annual doses from the absorption of the isotopes Ra-226, Ra-228, U-234 and U-238 through the daily consumption of 2 litres of the water in question

Figure 2. The radiological annual doses caused by the isotopes Ra-226, Ra-228, U-234 and U-238 of the water if one daily drinks 2liters

4. Conclusions

The content of natural radioactive isotopes in the tested bottled mineral waters varies over a wide range, e.g. the radium isotope content from almost 1 mBq dm^{-3} up to more than 1 000 mBq dm^{-3}. In general, it can be argued that with increasing water mineralisation, the content of radioactive isotopes increases. The correlation coefficients between water mineralisation and total radioactivityalpha, beta , ^{226}Ra and ^{40}K contents are respectively: 0.6; 0.0.97 and 0.98. In radioactive equilibrium the activities (expressed in .97; Bq dm^{-3}) of the uranium isotopes and radium should be equal , to each otherit was observed that, the activity of the uranium in the isotopes mineral waters studied is generally lower than that of the radium isotopes by at least an order of magnitude The isotope ^{40}K is not included in the radiological dose calculations, even though in some waters its content accounts for proportion of the a significant total beta activity. It is therefore proposed to give the total beta activity including potassium 40. The content of this isotope in water can be calculated from the known abundance of ^{40}K and the content of total potassium obtained from chemical analysis.

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