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title: **Natural radionuclides in drinking water supplies**

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INTRODUCTION

Groundwater from different aquifers is often used as a source of a drinking waters at many locations. It is usually stated, that such water supply are more safer for the public due to smaller possibility of contamination as surface ones. On the other hand, waters from underground reservoirs are containing more minerals usually, together with natural radionuclides. We were interested in investigation of the water purification processes, as radionuclides may be accumulated in the waste materials produced during water treatment.

For the preliminary investigations, eight different water supplies were taken into account, five of them were underground ones, while 3 others were surface water supplies. The latter ones were investigated to compare the balance of natural radionuclides in treatment processes, which in all of the cases were similar. The simplified description of the treatment method is following. At first gravel/sand filters have been used for the removal of mechanical suspention, afterwards the oxidation of the water has been applied in most of the cases to remove iron, manganse and sulphur (a co-precipitation or adsorption of natural radionuclides may occur at that stage). The sediments, containing iron hydroxide and manganese dioxide were filtered again at the gravel filters and finally a chlorine was added to the as the desinfecting agent. From time to time gravel filters are backwashed for the removal of the residues. Sludges are transported to the sedimentation tanks, where iron hydroxide and manganese dioxide are settled, together with adsorbed or co-precipitated natural radionuclides. Naturally occurring radionuclides can accumulate in these sludges enhanced concentrations, therefore these waste materials should be considered often as NORMs. Most abundant radionuclides, present in sludges are usually radium isotopes (Ra-226 and Ra-228). Due to some reports, elevated concentrations of radium are observed in waters with low oxygen content (Vornehm, 2009), which parameter can be correlated with hydrogeological conditions in the aquifer.

Level of acceptable radioactivity in the drinking water is regulated or recommended by different legal acts of recommendations. In the USA, the EPA regulation (EPA, 2001) was established for Ra-226 concentration as a permissible level 5 pCi/l (0,185 Bq/l). If concentration of radium is exceeding 3 pCi/l (0,111 Bq/l) measurements of Ra-228 concentration must be done. When radium concentration is above the permissible level, than the water treatment must be done for removal of radioactive nuclides. On the other hand, recommendation of the WHO (WHO, 2004) are following. At first the gross alpha and beta activity should be screened and in case, if gross alpha activity is below 0,5 Bq/l and gross beta activity doesn't exceed 1 Bq/l, such water can be used as a drinking water. Only in cases, when those values are higher as recommendations, analysis of particular radionuclides should be done. In European Union, the Drinking Water Directive (Drinking Water Directive, 1998) has been issued, followed by the Radon in Water Directive (Directive 2001/928/Euratom, 2001), regulating radon issues in drinking water supplies. Drinking Water Directive requirements are following – the only directly mentioned radionuclide is tritium H-3, which permissible level is 100 Bq/l. For all other radionuclides following provision is applied, that the total dose due to ingestion of radioactivity (called Total Indicative Dose — TID), shouldn't be higher as 0,1 mSv/year. Conversion factors from ingestion to dose were published in Euratom Directive (Euratom, 1996), in which whole population has been divided into six age groups, characterised by different conversion factors. In the Radon in Water Directive, recommended level of radon is established at level 100 Bq/l while permissible level is as high as 1000 Bq/l. Additional requirement of that Directive is the monitoring of Pb-210 and Po-210 when the recommended value is exceeded.

SAMPLING

Eight different water supplies, located in Upper Silesia region, were taken into preliminary investigations, five of them were underground ones, while 3 others were surface water supplies. In each of the cases, the raw water samples were taken, before any treatment and at least one sample of drinking water, after full treatment. The volume of particular water sample was 20 l, to have enough sample for preconcentration and measurements of all required radionuclides. Sampling has been done in August and September 2009 in the Upper Silesia Province in southern Poland. In three cases also sludges have been sampled from the water treatment installations, in which waters from underground were treated. The usual amount of the sludge was 2 liters, to enable gamma spectrometry of the sample.

RESULTS

20 samples of raw and treated waters have been take into analyses as well as 3 samples of sludges. Measurements of natural radionuclides in waters have been performed and results are shown in Table 1. Results of measurements for tritium, strontium and cesium were very low, usually below the detection limit for the particular analysis, therefore are not shown.

Analysis of results of natural radionuclides in water samples is showing the following pattern, that concentrations of all these radionuclides in waters from surface supplies are low (see samples 3, 5 and 11). It is different for samples from underground wells (samples 7, 9, 14, 15, 17 and 19). It can be seen, that concentrations of particular natural radionuclides can vary a lot and it is related to the conditions in particular aquifers.

During sampling three samples of sludges were taken for the analysis. Samples were measured with application of gamma spectrometry and results are presented in Table 2.

Table 2. Results of measurements of natural radionuclides in sludges from water treatment installations – results in Bq/kg of the dry mass.

No	Sampling site	Ra-226	Unc. 2 δ	Ra-228	Unc. 2 δ	Ra-224	Unc. 2 δ	Pb-210	Unc. 2 δ	U-238	Unc. 2 δ
1.	Konopisko (water No. 7 and 8)	2390	340	1496	76	366	28	345	44	360	90
2.	Dąbrowski (water No. 14 and 13)	1630	160	982	51	425	27	104	19	< 150	-
3.	Zwonowice (water No. 19 and 20)	905	45	510	25	108	15	55	15	< 160	-

It can be seen, that all of the sludges are showing enhanced activities of natural radionuclides. Most important radionuclides are radium isotopes, co-precipited and adsorbed on the sediments, while uranium U-238 was found only in one of the samples, probably being the effect of the uranium adsorption on the iron hydroxide. The presence of Ra-224 and Pb-210 is either a reason of its co-precipitation from the water, or more probably it is due to ingrowth of decay products of Ra-226 and Ra-228 in the sludge.

Table 1. Results of investigations for water samples

		Radium isotopes				^{210}Pb		Uranium isotopes			
			mBq/l				mBq/l		mBq/l		mBq/l
No.	**Sampling site**	**^{226}Ra**	**δ ^{226}Ra**	**^{228}Ra**	**δ ^{228}Ra**	**^{210}Pb**	**δ ^{210}Pb**	**^{238}U**	**δ ^{238}U**	**^{234}U**	**δ ^{234}U**
1	Tychy – treated water	<1,1	-	<6,6	-	<3,2	-	0,10	0,09	0,10	0,09
2	Pszczyna - treated water	<1,1	-	<6,6	-	<3,2	-	<0,05	-	<0,05	-
3	Goczałkowice – raw water	<1,1	-	<6,6	-	<3,2	-	1,01	0,32	1,18	0,35
4	Dziećkowice- treated water	<1,1	-	<6,6	-	<3,2	-	0,64	0,15	0,92	0,19
5	Dziećkowice – raw water	<1,1	-	<6,6	-	<3,2	-	0,99	0,18	2,19	0,31
6	Mikołów – treated water	<1,1	-	<6,6	-	<3,2	-	0,36	0,10	0,56	0,14
7	Konopisko – raw water	27,9	3,0	18,9	10,0	3,3	3,3	0,76	0,16	7,02	0,79
8	Konopisko - treated water	12,6	2,0	<6,6	-	4,0	3,3	0,15	0,04	0,42	0,07
9	Niegowonice raw water	60,2	5,0	12,7	10,0	5,7	3,4	11,48	0,82	18,70	1,29
10	Niegowonice treated water	60,9	5,0	<6,6	-	5,1	3,3	11,27	0,65	18,31	1,03
11	SUW Maczki raw water	<1,1	-	<6,6	-	<3,2	-	8,72	0,62	14,33	0,97
12	SUW Maczki treated water	<1,1	-	<6,6	-	<3,2	-	6,55	0,55	11,76	0,92
13	Dąbrowski treated water	25,8	3,0	65,6	20,0	3,5	3,3	4,59	0,57	11,75	1,26
14	Dąbrowski raw water	33,8	4,0	88,0	20,0	3,6	3,3	3,56	0,37	9,11	0,81
15	SUW Bielszowice raw water	42,1	4,0	48,0	20,0	3,7	3,3	3,87	0,48	8,39	0,89
16	SUW Bielszowice treated water	15,3	2,0	17,8	10,0	<3,2	-	3,70	0,34	8,26	0,66
17	Łabędy raw water	17,0	2,0	13,3	10,0	3,3	3,3	31,60	2,44	58,23	4,41
18	Łabędy treated water	11,2	2,0	8,7	7,6	3,3	3,3	34,19	2,52	60,77	4,40
19	Zwonowice raw water	5,0	1,5	<6,6	-	<3,2	-	0,09	0,05	0,25	0,05
20	Zwonowice treated water	<1,1	-	<6,6	-	<3,2	-	0,08	0,05	0,11	0,05
		LLD	^{226}Ra=	1,1	mBq/l	LLD ^{210}Pb	3,2	mBq/l	LLD ^{238}U=	0,05	mBq/l
		LLD	^{228}Ra=	6,6	mBq/l				LLD ^{234}U=	0,05	mBq/l

DISCUSSION

Results of investigations are showing, that the treatment of the raw water for the producing the drinking water may lead to the creation of sludges with enhanced natural radioactivity. On the other hand, far more important is the concentration of radionuclides in the drinking water after purification. The drinking of the water with elevated content of natural radionuclides may cause the increase of the effective dose for the population.

It can be clearly seen, that in some cases estimated annual doses are higher as 0,1 mSv/year. It also became obvious, that there are two critical groups in the population — babies and teenagers, for whom the number of cases, when the value 0,1 mSv/year has been exceeded was significant. In contrary, for adults we found no examples of the exceeding of the Total Indicative Dose limit. It means, that applying the WHO approach, all of investigated waters would be classified as a drinking waters. This point is very crucial, as application of the approach accordingly to EU Directive is leading to different conclusions. It means also, that relying on the screening method of gross alpha and gross beta activity may cause the underestimation of the annual dose.

CONCLUSIONS

1. Analyses of water samples, taken from different water supplies, are showing following pattern — usually concentrations of radionuclides in surface waters are low, while in waters from underground wells natural radiunuclides are often present.
2. Screening method for the assessment of radioactivity of the drinking water, it means measurement of gross alpha and gross beta activity is appear to be inadequate for this purpose, especially for underground waters.
3. The most important radionuclide, taking into considerations a dose for the population, is radium isotope Ra-228. The critical age groups are babies and adolescents.
4. In our opinion, a different approach should be applied for the monitoring of waters from surface supplies and underground ones. For surface waters the screening might be good enough to classify these waters.
5. For the monitoring of underground waters, the monitoring should be based on measurements of the most important natural radionuclides — Ra-228, Ra-226 and Pb-210, additionally uranium isotopes are sort of important ones, if gross alpha activity is high.
6. More attention must be focused in the future on investigation of sludges, as till now these waste materials are disposed into the environment without any measures againt the environmental pollution.
7. Additionally radon concentration in waters from underground water supplies should be monitored and mitigated, if necessary.

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