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Radium-226 concentration in spring water sampled in high radon regions

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ABSTRACT

Water ^{226}Ra concentration in springs was measured in regions with high indoor radon: Ural, North Caucasus (Russia), Niska Banja (Serbia), Piestany (Slovakia), and Issyk-Kul (Kyrgyzstan). This paper presents the results for ^{226}Ra concentration above 0.03 Bq l^{-1}. Radium in water could indicate indoor radon problem in the region and water investigation is useful at the initial stage of radon survey. Even low ^{226}Ra concentration in water (0.1–0.6 Bq l^{-1}) caused high ^{226}Ra activity in travertine (up to 1500 Bq kg^{-1}), which resulted in indoor radon concentration above 2000 Bq m^{-3} (Niska Banja).

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1. Introduction

For natural springs it is possible to find radium in water in a wide range of concentrations—from traces to significant values. Historically, most attention has been focused on ^{226}Ra and it is still the most important isotope. Radium is present in low-salinity solution as the uncomplexed cation Ra^{2+}. The weak complex of radium with chloride, sulphate and carbonate anions may be present in concentrated water, but will not affect the radium solubility or mobility to any extent in low-salinity water. It suggested that alpha recoil is a process for transferring radium from solid to aqueous phase. Once in solution, radium concentrations are controlled by sorbtion–desorbtion with the aquifer surfaces and, in water with high sulphate concentrations, by co-precipitation with insoluble sulphates (Dickson, 1990).

Radium in natural water springs presents two main problems in radiological protection. The first problem is the direct ^{226}Ra ingestion during water consumption. It is especially significant for spa resorts where the patients want to improve their health and pay more attention to the possible harmful factors. The second problem is concerning the fact that the elevated radium concentration in water springs can be an indicator of elevated ^{226}Ra concentration in deep geological formations or the source of possible soil contamination by ^{226}Ra and high radon concentration in the atmosphere of the dwellings situated over the contaminated place. Due to high radiotoxicity, ^{226}Ra is characterized by low action level (e.g. 0.5 Bq l^{-1} for drinking water in Russia) and a high-sensitivity technique is required.

2. Materials and methods

The modified method of ^{226}Ra concentration measurements is based on a well-known emanation technique (Lucas et al., 1990; Salih et al., 2000; Schubert et al., 2006): the specific activity of ^{226}Ra is determined by radon volume activity measurement in a sealed system during water sample bubbling. The advantage of this method is lack of any preliminary chemical or physical radium concentration and a small volume of sample (about 0.4 l). The measuring steps are as follows:

- degassing, sealing, and storage of water sample during at least 2 weeks in the flask, in which a sample was collected;
- absorption of background ^{222}Rn using the active coal filter cartridge in the inner volume of measuring system consisting of radon monitor AlphaGUARD (Genitron Instruments, Germany) based on pulse ionization chamber, set of glass vials AlphaKIT and air pump AlphaPUMP;
- transfer a water sample to a degassing vessel and air bubbling through the sample;
- measurement of ^{222}Rn concentration in a sealed system (at least 10 values of radon concentration for random error minimization);
- calculation of the specific activity of ^{226}Ra (A_m) by the following equation:

$$A_m = C\left(\frac{V_{system} - V_{sample}}{V_E} + k\right) - C_0\frac{V_{system} - V_{sample}}{V_{sample}} \quad (2.1)$$

where C– is the average radon concentration in the measuring system after expelling the radon, C_0– the background radon concentration in the measuring system, V_{system}– the inner volume of the measurement system, V_{sample}–volume of the water sample, and k– the radon distribution coefficient dependent on temperature.

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Table 1
Specific activities of ^{226}Ra and ^{222}Rn in water sampled in high radon regions and estimated effective doses relating to ^{226}Ra.

Country, region	Location	Description	Specific activity of ^{226}Ra ($Bq\,l^{-1}$)	Effective dose relating to ^{226}Ra ($mSv\,yr^{-1}$)	Specific activity of ^{222}Rn ($Bq\,l^{-1}$)
Russia, Ural	Lipovka (spa)	Radon spring	<0.03	–	540 ± 50
	Recreation Department Petushki	Drinking water	0.20 ± 0.03	4.0E−2	57 ± 6
	Recreation Department Razliv	Drinking water	0.35 ± 0.04	7.2E−2	92 ± 9
	Tyumen, Verkhniy Bor (spa)	Hot spring	0.21 ± 0.05	–	0.21 ± 0.05
Russia, Caucasus	Zheleznovodsk (spa)	Slavyanovskaya mineral water	4.1 ± 0.5	0.017	130 ± 15
	Zheleznovodsk (spa)	Smirnovskaya mineral water	2.1 ± 0.3	8.6E−3	118 ± 12
	Yessentuki (spa)	Yessentuki No. 4 mineral water	0.08 ± 0.03	3.3E−4	
	Yessentuki (spa)	Yessentuki No. 17 mineral water	0.05 ± 0.03	2.1E−4	
	Kislovodsk (spa)	Narzan dolomite mineral water	0.33 ± 0.06	1.4E−3	
	Pyatigorsk (spa)	Krasnoarmeyskiy mineral spring	0.09 ± 0.03	3.7E−4	
	Pyatigorsk (spa)	Mineral spring No. 2	0.36 ± 0.06	1.5E−3	
Kyrgyzstan, Issyk-Kul	Dzhety–Oguz spa	Drinking spring	0.97 ± 0.12	0.20	
	Dzhety–Oguz spa	Radon spring	1.8 ± 0.2	–	
Slovak Republic, Piestany	Spa, Napoleon Building	Mineral spring	1.8 ± 0.2	7.4E−3	56 ± 6
Serbia, Niska Banja	Skolska cesma	Drinking spring	0.11 ± 0.03	0.022	430 ± 46
	Kraljevo kupatilo	Hot spring	0.62 ± 0.10	–	44 ± 5
	Near Ozren Hotel	Hot spring	0.57 ± 0.09	–	148 ± 14
Serbia, other regions	Topilo spa	Drinking spring	0.08 ± 0.02	0.016	45 ± 5
	Kosovo, Gornja Stubla	Drinking spring	0.07 ± 0.02	0.014	
	Vranjska Banja	Hot spring	0.08 ± 0.02	–	
	Kosovo, Bozin bunar	Drinking spring	0.07 ± 0.02	0.014	
	Kosovo, Slatina mineralna	Drinking spring	0.11 ± 0.03	0.022	

Radon loss by decay during the measurement is neglected due to the short time of experiment. Special attention should be paid to background radon concentration and air-tightness of the system. To reach low detection limit in $0.03\,Bq\,l^{-1}$ the average value of background should be reduced to $5–10\,Bq\,m^{-3}$ and air-tightness of the system should be provided at the time of continuous bubbling of air through water. After 2–3 weeks' interval the water sample can be repeatedly measured in case of storage in the same completely filled flask.

The measurement error consists of systematic bias and random error. The systematic bias consists of AlphaGUARD calibration error (8%), error of the system interior volume assessment (1%), and error of sample volume (1%). The random error depends on a number of radon concentration measurements and a level of radon concentration in the system. The total error rarely exceeds 30%.

3. Results and discussion

The specific activity of ^{226}Ra in water was measured in five regions with high levels of radon in dwellings: Ural, Russia (Zhukovsky and Yarmoshenko, 1998); Niska Banja, Serbia (Zunic et al., 2007b); Piestany, Slovak Republic (Dubois, 2005); North Caucasus, Russia (Lezhnin et al., 2008); and Issyk-Kul, Kyrgyzstan (Zhukovsky and Termechikova, 2005). We have measured 55 water samples collected from different regions in Serbia, 24 samples collected from artesian springs in Issyk-Kul region, 9 samples taken from different spas of Caucasus, 14 samples collected from Ural springs, and one sample taken from a spa in Piestany. It was found that high radon concentration in water ($400–600\,Bq\,l^{-1}$) does not always lead to the presence of radium in the sample and the relationship between ^{226}Ra and ^{222}Rn activity in spring cannot be estimated. In one case we obtain that all radon in sample is caused by ^{226}Ra decay, but usually ^{222}Rn activity in spring is higher than ^{226}Ra ones. Table 1 shows specific activities of ^{226}Ra and ^{222}Rn in samples having activities above low detection limit. In the majority of samples presented, the radium specific activity was either below action level or a little above it. The considerable excess of action level was obtained only in 4 water springs used for medical and bathing purposes.

The correlation between the presence of ^{226}Ra in water and high radon concentration in dwellings was found in Niska Banja and North Caucasus where the springs with radium content outflow for many years. For these places, the formation of travertine during the interaction of radium in water and limestone is typical. Fig. 1 shows that even low ^{226}Ra specific activity in water ($0.1–0.6\,Bq\,l^{-1}$) in Niska Banja can cause high radium specific activity in travertine (up to $1500\,Bq\,kg^{-1}$) (Zunic et al., 2007a) and extremely high indoor radon concentration (more than $2000\,Bq\,m^{-3}$). For North Caucasus we did not find a direct spatial correlation between radium in surveyed springs and elevated indoor radon concentration (up to $1500\,Bq\,m^{-3}$), but the travertine formation with activity up to $1700\,Bq\,kg^{-1}$ (Tokarev and Shcherbakov, 1956) and a number of outflow springs in this region allow making an assumption concerning such correlation. For Piestany we cannot make any conclusions because of insufficient data. In Ural and Issyk-Kul region most springs are artesian with radium activity below $0.03\,Bq\,m^{-3}$ and formation of travertine does not occur.

To estimate committed effective doses via ingestion for ^{226}Ra, the considered adult dose coefficient was $2.8\times10^{-7}\,Sv\,Bq^{-1}$ (ICRP, 1996). Doses presented in Table 1 were calculated by taking

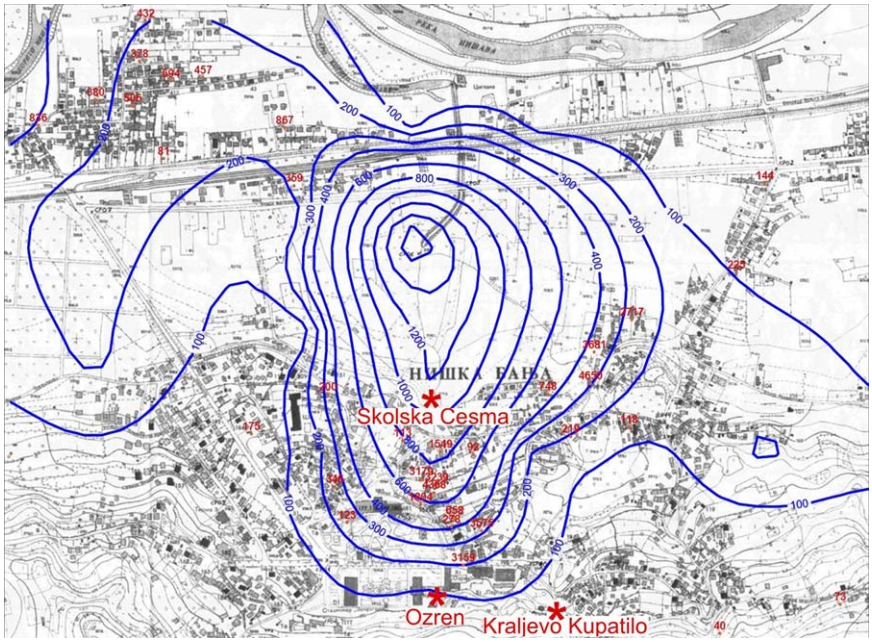

Fig. 1. Connection between the springs coordinates, isolines of ^{226}Ra concentrations in soil (Bq kg^{-1}) and average annual indoor ^{222}Rn concentrations (Bq m^{-3}) in Niska Banja, Serbia.

into account a consumption of 2 l of water per day for 1 year for drinking water and 0.7 l per day for 3 weeks for mineral water during cure process in spas. Doses from ^{226}Ra contained in hot springs and radon springs were not estimated due to another application—bathing. The action level of ^{226}Ra exceeded in a number of samples, but in general it was the mineral water for cure purposes with limited consumption; therefore, doses estimated for ingestion of radium in this waters were negligible. Only one case where the spring is used for regular drinking was found to exceed action level 0.1 mSv yr^{-1}.

4. Conclusions

In general, for the investigated regions there are no radiological protection problems due to radium consumption with drinking water. The elevated radium concentrations in water are typical for mineral water used in medical and recreation purposes. Nevertheless the presence of radium in springs can be an indicator of elevated indoor radon concentration in the dwellings of surrounding area and radium concentration measurements in springs could serve as one of the initial stages of radon survey.

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