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RADIONUCLIDES IN SOME SPRING MINERAL WATERS IN SERBIA

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Abstract

Radiochemical analyses of some natural mineral bottled waters from different location in Serbia were investigated. Concentrations of all present naturally occurring radionuclides, ^{238}U, ^{234}U, ^{232}Th, ^{230}Th, ^{228}Th, ^{228}Ra and ^{226}Ra and $^{234}U/^{238}U$, $^{226}Ra/^{230}Th$, $^{228}Th/^{232}Th$, and $^{228}Ra/^{228}Th$ activity ratios were calculated and discussed. Uranium series disequilibria in the hydrosphere occur due to geochemical differentiation processes resulting with different mobility of the radionuclides from the same series. We have investigated radioactive disequilibrium in the spring waters Crni Guber, Čibutkovica and Studenica, originated from metamorphic rock area. High content of radium isotopes (^{226}Ra, ^{228}Ra) in analysed natural spring waters indicates contribution from other non-water sources, probably environmental sediment.

Introduction

Consumption of spring mineral water is continuously increasing during last few decades. Waters coming from deeply located sources in the earth crust may transport natural radioactive isotopes (including radium isotopes) under certain geological conditions. Two of them (^{226}Ra, ^{228}Ra) are of real menace from radiological point of view, because when disintegrating they create series of daughter alpha and beta radioactive elements. Permanent consumption of mineral water with higher concentration of radium isotopes might be dangerous for human health. Increased contents of ^{226}Ra were observed in some mineral waters, used as drinking waters in France where the highest concentration was 2.7 Bq/l, Portugal 2.2 Bq/l, Germany 1.8 Bq/l, Brazil 3.5 Bq/l [1]. Activity concentrations for drinking water, recommended by WHO, are 0.1 Bq/l for gross *alpha* and 1 Bq/l for gross *beta* activity. These recommendations must be appled to routine operational conditions of existing or new water supplies. For natural spring water we have to measure naturally occurring radionuclide ^{40}K, which makes up about 0.01 % of natural potassium (WHO, 1996).

We have investigated several natural spring waters. In these spring waters gross *alpha* and gross *beta* activity exceed recommended values of 0.1 Bq/l and 1Bq/l, respectively, for drinking and mineral water. These are carbon acid waters with similar chemical composite and same pH value (6.5).

Experimental

Alpha spectrometry procedure for uranium and thorium isotopes included: sampling, preliminary samples treatment, ion-exchange chemical separation, ion-exchange purification of separated elements, thin-layer source preparation and alpha spectrometry measurements.[3]

After collection water samples have been acidified, evaporated and ashed at 550°C. ^{232}U and ^{229}Th have been aded in a quantity of about 0.1 Bq as a tracer to each sample, for radiochemical yield recoveries.

Radiochemical separation and purification was done on ion-exchenge resin DOWEX 1x8, 100-200 mesh. Electroplating of purified fractions, was used to made thin-layer radioactive sources. After electrodeposition, a thorium source was covered by vyns-foil. Samples have been concretrated by evaporating 10 l water to 450 ml for gamma analysis. After preparation, samples have been sealed in the acril containers for 4 weeks to reash radioequilibrium between ^{228}Ra and ^{228}Ac and ^{226}Ra and ^{222}Rn.

Low-level activity measurements have been done by the use of Canberra 2004 alpha-spectrometry counting system, including vacuum chamber (20 mbar), PIPS-detector (300 mm^2 surface), with: counting efficiency 15%, at 25mm distance; multichannel energy scale 9.1 keV/ch, resolution 24 keV for ^{241}Am.

The counting time required had to be a few days, that is long enough to ensure an accurate result.

The gamma activity of the samples has been counted using high purity Ge detector,with counting relative efficiecy 23 %.

Results and Discussion

Spring water Crni Guber exhibits a disequilibria mostly in the ^{232}Th series (Table1.). The ^{226}Ra content, that should be in equilibrium with ^{238}U, exceeds the value expected for the equilibrium. The equilibrium means that neither gain nor loss of any parent or daughter in a decay chain are not possible. The ^{230}Th isotope was not found in this sample. However, the ^{226}Ra and ^{228}Ra activities found in this sample are much higher than those of their parents, because of the same chemical behaviour of different isotopes of an element. In the ^{232}Th series the $^{228}Th/^{228}Ra$ disequilibrium is much higher than expected from present ^{232}Th. High ^{228}Th content may derive from ^{228}Ra. Since chemical properties of uranium and radium are quite different, mobility of the parent and product isotopes are different in most chemical environments. ^{232}Th is insoluble in water environments. However, its daughter nuclide ^{228}Ra is much more soluble. The $^{228}Th/^{232}Th$ and $^{228}Ra/^{228}Th$ ratios are 126.7 and 7.7.

Table 1: Radioactivity ratios of $^{234}U/^{238}U$, $^{228}Th/^{232}Th$, $^{226}Ra/^{230}Th$ and $^{228}Ra/^{228Th}$ in mineral waters

	$^{234}U/^{238}U$	$^{228}Th/^{232}Th$	$^{226}Ra/^{230}Th$	$^{228}Ra/^{228}Th$
Crni Guber	1.24	126.7	/	7.7
Čibutkovica	0.95	54.3	38.5	12.4
Studenica	1.24	13.6	50000	/

In the Čibutkovica spring water a disequilibria has been found, both in the ^{238}U and ^{232}Th series. The ^{226}Ra content, that should be related to the ^{238}U one, exceeds very much the equilibrium value. The ^{228}Th, ^{228}Ac and ^{228}Ra concentrations are much higher than expected from present ^{232}Th amount. In the decay process from ^{232}Th to

^{228}Th, the ^{228}Ra radionuclide ($t_{1/2}$ = 5.75 y) is in the middle. Since chemical properties of radium and thorium are different, high ^{228}Th content may derive from ^{228}Ra content. The ^{228}Th/^{232}Th, ^{226}Ra/^{230}Th and ^{228}Ra/^{228}Th ratios are 54.3, 38.5 and 12.4, respectively.

The results obtained here show that the rise in the activity level corresponds to the ^{226}Ra isotope, for the Studenica water, since the ^{226}Ra/^{230}Th ratio was found to be 50000. Corresponding ratio for the Čibutkovica water is 38.5. The ^{228}Ra isotope was not found in this water, so because of that the ^{228}Ra/^{228}Th ratio was missed. For comparison, the least ^{228}Th/^{232}Th ratio (of only 13.6) was found in the Studenica water.

Conclusion

The uranium series disequilibria can occur due to geochemical differentiation processes resulting with different mobility of the radionuclides from the same series. High content of radium isotopes (^{226}Ra, ^{228}Ra) in analysed natural spring waters indicates contribution from other non-water sources, probably environmental sediment.

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