Gamma spectrometric measurements of radon daughter ^{214}Bi in surface waters

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RESUMEN

Se han realizado medidas por espectrometría gamma de ^{214}Bi y ^{208}Tl en agua y sedimentos del río Odra, Polonia, y en algunos de sus afluentes. Los resultados obtenidos indican que el flujo del radón a partir del sedimento es significativamente mayor que el radón producido por el ^{226}Ra presente en el agua. La presencia de sedimentos saturados de agua y el flujo de agua subterránea fomentan la migración del radón y del torón.

Los efectos de anomalías de origen antropogénico del ^{226}Ra de la minería del carbón y del ^{137}Cs de Chernobyl son insignificantes.

PALABRAS CLAVE: Radón, río, agua, gamma, espectrometría.

ABSTRACT

Gamma spectrometric measurements of ^{214}Bi and ^{208}Tl have been performed in water and bottom sediments of the Odra river, Poland, and some of its tributaries. Obtained results indicate that radon flux from the bottom sediment is significantly greater than radon produced by ^{226}Ra present in water. Presence of water saturated sediments and influx of ground waters promote both radon and thoron migration.

Effects of antropogenic coal mining ^{226}Ra and Chernobyl ^{137}Cs anomalies are insignificant.

KEY WORDS: radon, river, water, gamma, spectrometry.

INTRODUCTION

Odra river and some of its tributaries have been chosen for gamma spectrometric investigations due to the fact that two significant radioactive anomalies are located with the Odra river catchment area. ^{226}Ra anomaly is connected with coal mining and ^{137}Cs with Chernobyl event. The other suspected sources of radionuclides were Variscan rocks and copper mining of the Sudetes and its foreland.

ANALYTICAL METHOD

Measurements have been performed using Exploranium GR-320 gamma spectrometer connected by 30 m long cable with the NaI (Tl) detector borehole-probe supplied with 0.5 °C ^{137}Cs source installed in the base of the detector unit (requires no license in most countries). Results have been controlled using standard GPX-21A detector.

Impulses supplied by the detector units have been classified using channels 70-204 of the 256 channels of the spectrometer covering the energy window 850-2810 keV.

The problem of stabilization of energy windows of channels was solved by means of continuous measurement of cesium 662 keV photons in the band RO/1 covering channels 51-60 (600-730 keV). Gain parameter responsible for fitting channels to energy windows was continuously updated using least-squares fit of a Gaussian Cs peak shape every time the 5000 level of Cs counts was exceeded. This ensured that System Gain was always correct and selected channels corresponded to the desired energy windows.

Three selected bands (Regions of Interests ROI) corresponding to energy windows of radionuclide peaks ^{40}K, ^{214}Bi and ^{208}Tl has been set up.

Instrument was set to ASSAY evaluation which uses Calibration Coefficients computed (by the manufacturer) during calibration on the Test Pads to display data at the end of the sample period as:

TOTAL COUNT in ppm eU.
POTASSIUM in %K ,
URANIUM in ppm,
THORIUM in ppm.

Table 1

Band	Radionuclide peak		Channels	Energy window (keV)	Sensitivity
RO/2	^{40}K40	1460 keV	109-122	1370-1570	0.661 cps/%
RO/3	^{214}Bi	1760 keV	129-142	1660-2810	0.067 cps/ppm
RO/4	^{208}Tl	2620 keV	179-204	2410-2810	0.025 cps/ppm

Displayed data were recalculated into activity units (Bq/kg).

Measurement of ^{214}Bi and ^{208}Tl peaks is commonly applied for evaluation of equivalent uranium(eU) and thorium(eTh) concentrations in geophysical prospecting. Taking into account the fact (see Tables 2 and 3) that radon and thoron are closer to these radionuclides in the decay series it is obvious that obtained results are more trustworthy for these gases than for uranium and thorium. Due to this fact many radon anomalies have been interpreted in the past as uranium ones. Measurements of cesium have been performed using detector without cesium source. In spite of the ground surface anomalies reaching tens of kBq/m^2 in the catchment area, the concentration of ^{137}Cs in rivers was below detection limit.

Radionuclides content of bottom sediments and water have been measured. If the river was deep enough measurements have been done in water 0.8 m under water surface and at least 0.7 meter above bottom. Next, the measurement at the bottom surface was done.

The two main restrictions of measurements existed:

- in the case of shallower water the result for water, can be treated only as approximate one since the influence of the bottom sediments cannot be neglected,

Table 2

Half lifes and 99% equilibrium times for ^{238}U decay series

Radionuclide	Half life	Unit	99% Equilibrium	Unit
^{238}U	4,468	10^9 years		
^{234}Th	24,1	days	160,1	days
^{234}Pa	1,18	minutes	7,8	minutes
^{234}U	2,48	10^5 years	1,6	10^6 years
^{230}Th	7,52	10^4 years	5,0	10^5 years
^{226}Ra	1602	years	1,0	10^4 years
^{222}Rn	3,825	days	25,4	days
^{218}Po	3,05	minutes	20,3	minutes
^{214}Pb	26,8	minutes	178,1	minutes
^{218}At	2	seconds	13,3	seconds
^{214}Bi	19,7	minutes	130,9	minutes
^{214}Po	1,64	10^{-4}seconds	10,9	10^{-4} seconds
^{210}Tl	1,32	minutes	8,8	minutes
^{210}Pb	22,3	years	148,2	years
^{210}Bi	5,02	days	33,4	days
^{210}Po	138,3	days	918,8	days
^{206}Tl	4,19	minutes	27,8	minutes

Table 3

Half lifes and 99% equilibrium times for ^{232}Th decay series

Radionuclide	Half life	Unit	99% Equilibrium Time	Unit
^{232}Th	1,39	10^{10} years		
^{228}Ra	5,75	years	38,2	years
^{228}Ac	6,13	hours	40,7	hours
^{228}Th	1,913	years	12,7	years
^{224}Ra	3,64	days	24,2	days
^{220}Rn	55,6	seconds	369,4	seconds
^{216}Po	0,145	seconds	1,0	seconds
^{212}Pb	10,64	hours	70,7	hours
^{212}Bi	60,5	minutes	402,0	minutes
^{212}Po	3,04	10^{-7} seconds	20,2	10^{-7} seconds
^{208}Tl	3,1	minutes	20,6	minutes

- in few cases the river current was too rapid and it was impossible to place detector on the bottom.

Influence of the thickness of water layer on the measurements of the gamma photons emitted by granitic bottom is shown in the Figure 1.

Data for this figure have been collected at the solid granitic layer where radon emanation (and ^{214}Bi, ^{208}Tl migration) cannot be very high. It is visible that the granitic bottom of 1% K, 0,7 ppm eU and 2 ppm eTh content can influence measurements in water at the distance of 0.7 meters. Decrease of ^{40}K radiation and reduction of ^{214}Bi/^{40}K and ^{208}Tl/^{40}K ratio to zero is especially significant since in contrary to ^{214}Bi and ^{208}Tl no emanation and migration to adjacent water occurs in the case of ^{40}K .

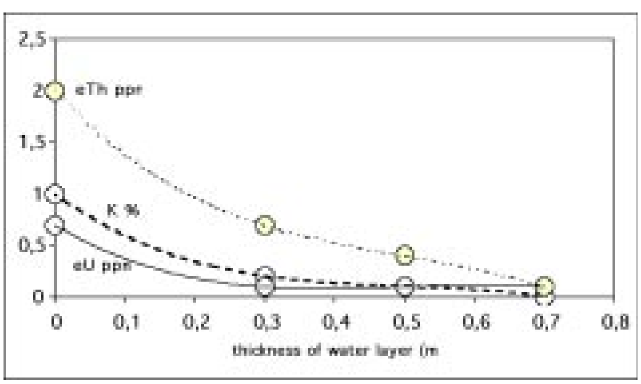

Fig. 1. Decrease of ^{40}K, ^{214}Bi and ^{208}Tl the granitic bottom radiation (expressed in concentration units) with distance.

AREA UNDER INVESTIGATION

Odra river is the second (after Vistula) river of Poland. It starts in the Czech Republic and arrives to Poland as a river of 55 m^3/s flow reaching the value of 540 m^3/s in its lower course.

Odra river in its upper course receives waters form the part of the Upper Silesian Coal Basin (USCB), where mining exploitation reaching in the past up to 200 mln tons of coal per year takes place. Mining activity results in discharging 20 MBq of ^{226}Ra and 10 MBq of ^{228}Ra per day to Odra river (Chalupnik *et al*., 2001). Taking into account the flow of 55m^3/s it should make no more than 4Bq/m^3 ^{226}Ra and 2 Bq/m^3 of ^{228}Ra in Odra water coming from USCB mines. It is significantly lower than the natural groundwater radium content reaching tens and even hundreds of Bq/m^3 (see Hess *et al*. 1985, Solecki 1997).

Value of 40Bq/m^3 of radium reported by Muras and Olszewski (1993) for Odra water contrary to their opinon seems to be evidently the natural background one.

However in the case of smaller tributaries which transport mining waters to the Odra river radium concetration in water and bottom sediments may be significantly higher. Values of hundreds of Bq/m^3 in small rivers of the USCB have been measured (Chalupnik *et al*., 2001).

In its middle course the Odra river flows across Chernobyl ^{137}Cs anomaly of Opole and next receives its southern tributaries from the Sudety Mts. area where the

Variscan orogen with uranium rich granites is being eroded. Locally copper and uranium mining took place there. However, the known radium content of river waters in this area (54 Bq/m^3 for the Kaczawa river after Muras and Olszewski 1993 is significantly lower than in the case of the USCB).

RESULTS

Results of gamma spectrometric measurements of ^{214}Bi and ^{208}Tl of the bottom sediments and waters are shown in the Figure 2 and 3 respectively.

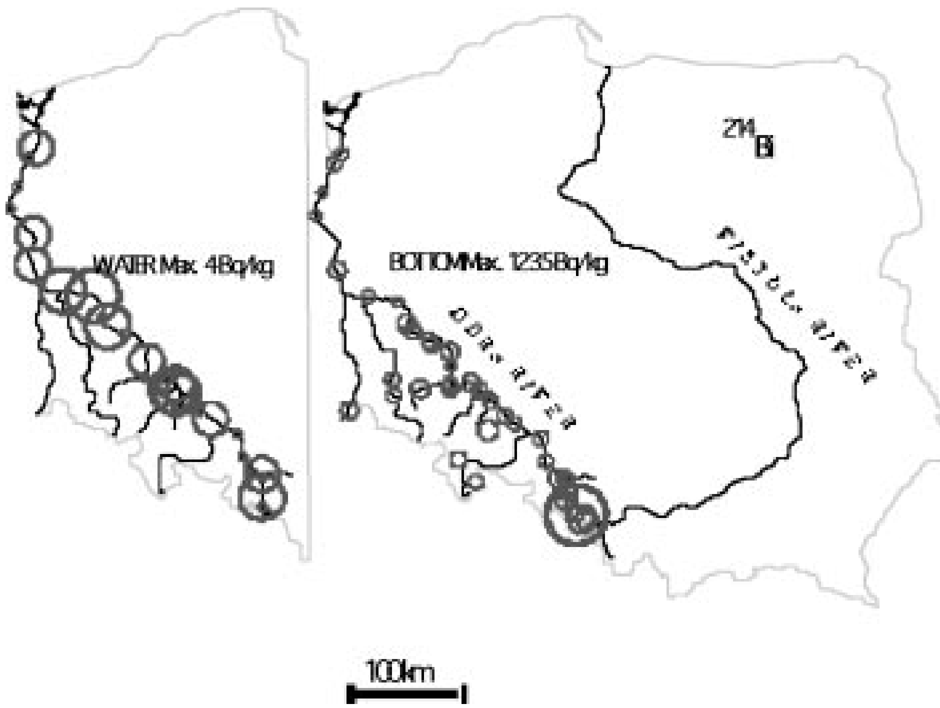

Fig. 2 ^{214}Bi in waters and bottom sediments of the Odra river and some of its tributaries (circles area proportional to values).

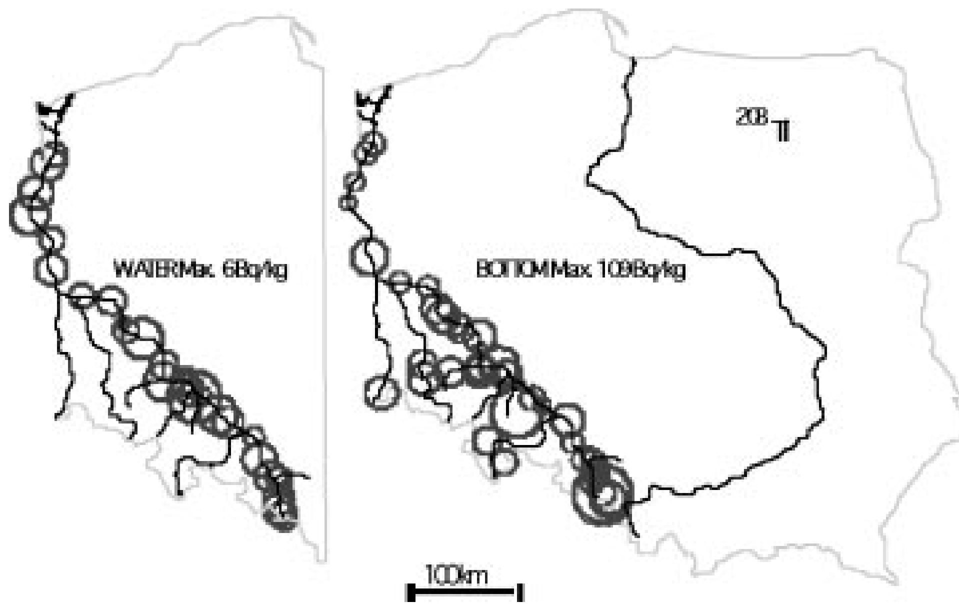

Fig. 3. ^{208}Tl in waters and bottom sediments of the Odra river and some of its tributaries (circles area proportional to values).

Two greatest values as high as 1235 and 162 Bq/kg of ^{214}Bi for bottom sediments have been measured in one of the small creeks of the USCB area. The other results vary in the range of 0-51 Bq/kg where the higher results were obtained in the points where granitc bedrock of natural (in smaller creeks) or artificial (Odra river engineering constructions) origin occurs. ^{208}Tl activity reaches its maximum of 109 Bq/kg in the same small creek of the USCB area. Granitic bedrock results are in the range of 75-32 Bq/kg for both radionuclides.

The majority of the Odra river sediments are of low radionuclides content reaching maximum values of 31 and 38 Bq/kg for ^{214}Bi and ^{208}Tl respectively , beeing sometimes below detection limit. Results for bottom sediments of tributaries coming from the Sudety Mts. are slightly higher. Single increased results obtained for the Odra river sediments are connected with occurrence of redeposited granitic material derived from the river engineering constructions.

Potassium content of bottom sediments does not exceed 0.4%.

In the case of the Odra river water the maximum values of 4 and 6 Bq/kg, for ^{214}Bi and ^{208}Tl respectively have been measured in the middle part of its course. The lowest values being below detection limit have been found both in the upper and lower part of the Odra river course. In most cases it was ^{40}K and ^{214}Bi which fall below detection limit. Control HP Ge laboratory measurements of ^{226}Ra and ^{228}Ra in the Odra river water samples have shown that activities of these radionuclides are much lower than that of ^{214}Bi and ^{208}Tl being below detection limit of 0.01 and 0.05 Bq/kg respectively.

Significant disequilibrium exists between radon progeny and radium both in ^{238}U and ^{232}Th series. It can be explained assuming that ^{214}Bi and ^{208}Tl in the Odra river water are remnants of the radon flux released from the bottom sediments (see Fig.4). Bottom sediments of the mature river in the middle part of its course are of smaller grain size and reach significant thickness.

Moreover the upper part of the sediment is water saturated and mobilized by the current. As a result significant volume of the Odra river water can be treated as ground water from the Rn emanation point of view. On the other hand direct influx of ground waters into Odra river channel cannot be excluded in this area, where moraine hills dominate of the river valley. In the case of ground waters values of tens or even hundreds of Bq/kg can be expected (see C.Cosma *et al.,* 2001, L.Toscani *et al*., 2001, A.T. Solecki, 2001).

Decrease of radon progeny activities in the lower course of the Odra river is connected with occurrence of vast marshes

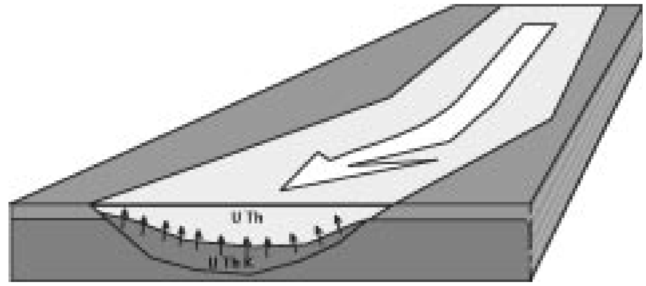

Fig.4. Radon migration from alluvial sediments.

on both sides of the river where water purification and adsorption of pollutants takes place.

CONCLUSIONS

In situ gamma spectrometric measurements of bottom sediments and water of the Odra river have shown that radon flux from the bottom sediments and accumulation of radon progeny in water takes place. As a result significant disequilibrium with radium in water occurs.

This process is controlled not only by direct radon emanation from thick water saturated sediments but probably by ground water influx into the river channel as well.

Coal mining of the USCB is the most significant antropogenic source of radionuclides but its environmental influence is limited to small creeks in the catchment area.

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