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Short Communication

Natural Radioactivity of Soil and Sediment Samples Collected from Postindustrial Area

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Abstract

Contents of a typical postindustrial pond that was used 12 years previously as a tank for collecting waste water pumped from a coal mine was evaluated on the basis of 19 samples collected from the pond and surrounding area. Measurements of radium, thorium, and potassium, plus cesium activity, were performed with the use of a germanium detector – HPGe 4020 by examining photons with energy in the range 0.5 MeV to 3 MeV. In consequence, the concentrations of such radioisotopes as ^{226}Ra, ^{232}Th, ^{40}K and, ^{137}Cs were determined by their decay products, but the radiation effect of natural radioisotopes was estimated by I_1 and I_2 coefficients. The investigation shows the most contaminated residue in the sediments at the bottom of the settling tank.

Keywords: Gamma spectrometry, Ra-226, Th-232, Cs-137, K-40, I_1 and I_2 factors

Introduction

One of the most important contributors to Vistula River contamination is coal mines. Salty mine water from several mines is pumped from underground and is collected in dosing tanks. Mine sewage contains a lot of coal dust, minerals, and some radioactive isotopes.

The existence of radioactive elements in water provokes spreading contamination onto the surface of the ground. After settling the suspension, the partially purified water is dosed in rivers. During many years of exploitation, sediments were accumulated in a tailing pond, mostly on its bottom. These deposits include plenty of natural radioisotopes, in particular those of uranium series. Very often their concentration is higher than average values in this area [1].

Rontok Duży pond is a reservoir with a significant sedimentation lagoon [2]. It was created in 1977 with the aim of protecting the Vistula, which is about 200 meters from the pond.

The settling pond is located on the edge of Oswiecimska Valley in the Silesia region, in Rudoltowice, Poland.

Exploitation of the pond lasted from 1977 to 1997. The fact that near the pond are many fields and woods raises the question of whether it is still a hazard for neighboring areas. The aim of this paper is to discuss this problem, having checked the radioactive pollution by selecting several examination points representing this area.

Method of Measurement

Sampling Collection

The most important measurement points were chosen at the bottom of the reservoir: near the edge and in the middle of the pond. Additional soil samples were collected from the neighborhood of the settling tank: northeast and northwest from the fields, below the water-supplying area of the reservoir, from the dike between the main reservoir and a buffer pond, from the western and northern parts of the

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embankment and from the woods to the north. All of them were taken from 10 centimeters depth. The samples evaluated in this paper were collected in 2007.

Several samples of soil and several of sediments were taken as reference probes from an open reservoir about 10 kilometers from Rontok Duży Pond.

Sampling locations are marked on the map (Fig. 1).

Gamma Spectrometric Measurement

All samples collected in this investigation were dried for 16 hours at 105ºC and closed in aluminum containers [3-5]. We assumed that after 14 days radium and its short-lived decay products reached radioactive equilibrium. Then their activity was measured by an HPGe 4020 high-purity germanium detector constructed by Canberra. All samples of soil and sediments were prepared in the same way as the measurement.

During examination, photons with energy in range from 0.5 MeV to 3 MeV were analyzed with 40% efficiency. The energy resolution of the detector was equal to 0.95 keV for the 122 keV peak and 1.8 keV for the 1,332 keV one.

The concentration of radioactive isotopes like ^{226}Ra, ^{228}Th, ^{40}K, and ^{137}Cs were determined by their decay products. Radium activity was determined by ^{214}Bi (609.3 keV, 1,120.3 keV, and 1,764.5 keV), thorium by ^{208}Tl (583.2 keV and 2,614.5 keV) and content of ^{232}Th (in secular equilibrium with ^{228}Th) was obtained from its decay isotopes balance. The concentration of potassium was determined from its 1460.8 keV energy and cesium from 661 keV [3, 6].

In all measurements, counts were collected over a period of 20 hours.

I_1 and I_2 Factors Calculation

Radiation effect of natural radioisotopes could be estimated by two coefficients, I_1 and I_2, proposed by the European Commission [7]. The first one (I_1) determines natural isotope abundance like ^{40}K, ^{226}Ra, and ^{232}Th, and the second one (I_2) is equal to ^{226}Ra concentration.

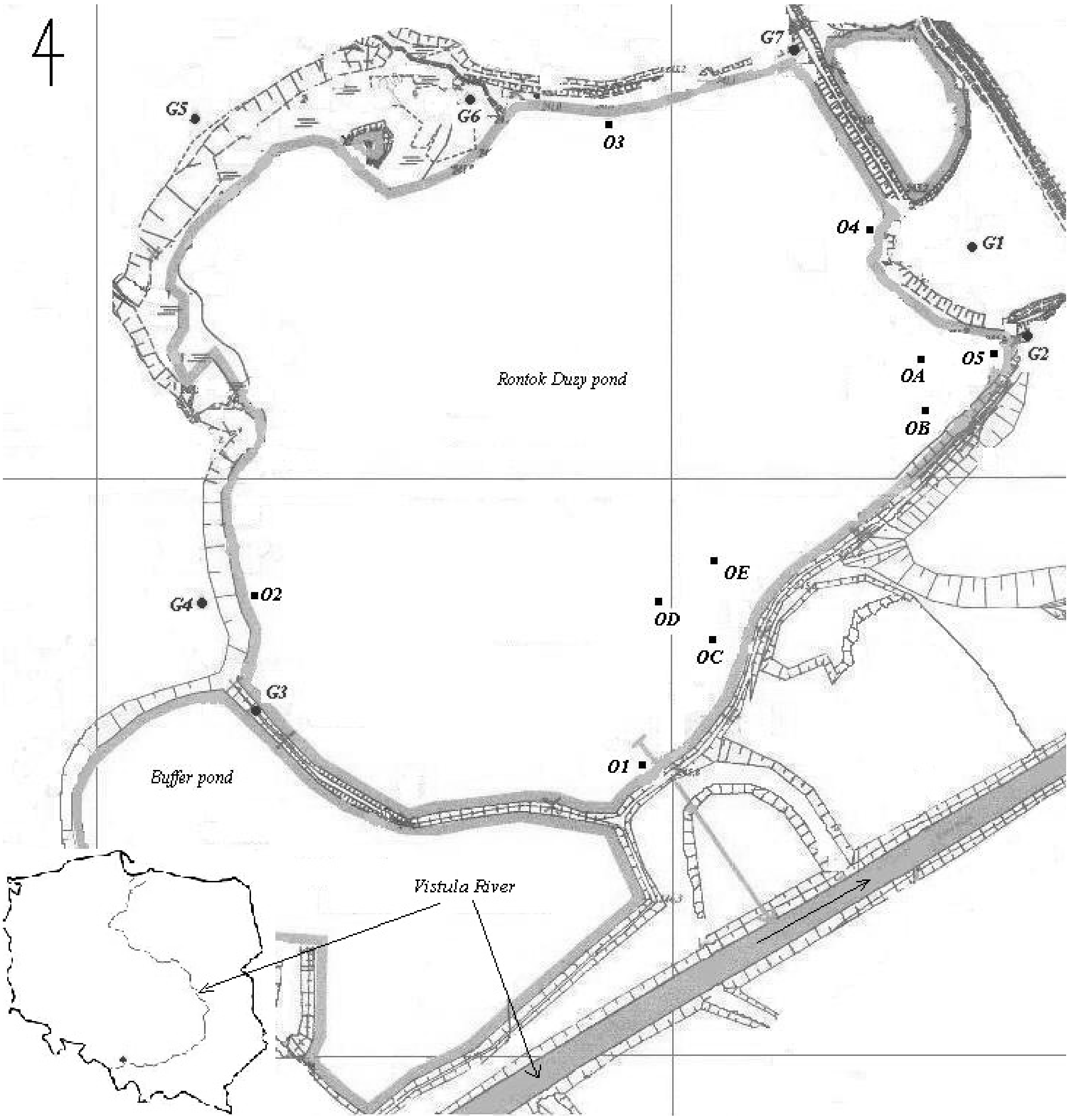

Fig. 1. The investigated area with location of sampling points (O – sediment; G – soil).

The I_1 and I_2 factors are assigned by the following equations:

$$I_1 = \frac{S_K}{3{,}000 Bqkg^{-1}} + \frac{S_{Ra}}{300 Bqkg^{-1}} + \frac{S_{Th}}{200 Bqkg^{-1}}$$

$$I_2 = S_{Ra}$$

...where: S_K, S_{Ra}, S_{Th} are activity concentrations of ^{40}K, ^{226}Ra, and ^{232}Th in $Bqkg^{-1}$, respectively.

Absorbed Dose Rate

The measured activities of ^{40}K, ^{226}Ra, and ^{232}Th in soil samples were used in calculation of the gamma-absorbed dose rate in the air at 1 m over the ground. If the influence from nuclides ^{137}Cs or ^{235}U series is ignored, the following equation is obtained [8-10]:

$$D = 0.462 S_{Ra} + 0.604 S_{Th} + 0.042 S_K$$

...where:

D is dose rate in μGyh^{-1}

S_K, S_{Ra}, S_{Th} are activity concentrations of ^{40}K, ^{226}Ra, and ^{232}Th in $Bqkg^{-1}$, respectively.

Results and Discussion

The principle result of this investigation was determination of the following radioisotope evaluations ^{40}K, ^{226}Ra, ^{232}Th, and ^{137}Cs. The concentration of enumerated isotopes is most important to estimate the radiation hazard of the discussed area [10]. The results of the activity measurements are shown in Table 1.

The most important criterion for measuring point selection was their availability for people. All of them could be divided into two groups. Containing the radionuclides in the first one is similar to natural values of the specific activity of potassium, radium, thorium, and cesium in Poland. The soil and sediment samples, taken near the edge of the settling pond, are ranked to this group. The activity of soil samples are between 33 and 155 $Bqkg^{-1}$ for ^{226}Ra, 428 and 580 $Bqkg^{-1}$ for ^{40}K, and 53 and 83 $Bqkg^{-1}$ for ^{232}Th. Similarly, sediment samples are between 31 and 75 $Bqkg^{-1}$ for ^{226}Ra, 337 and 607 $Bqkg^{-1}$ for ^{40}K, and 39 and 69 $Bqkg^{-1}$ for ^{232}Th.

The results in the second group considerably exceed the natural values of the average activity in the soil of Poland: 4.2 – 116 $Bqkg^{-1}$ for ^{226}Ra, 60 – 1,028 $Bqkg^{-1}$ for ^{40}K, and 3.6 – 82.3 $Bqkg^{-1}$ for ^{232}Th [11, 12]. The sediment samples taken from the middle of the pond and ones taken near the edge of the pond are also classified to this group because the activity of enumerated isotopes ranged between 2,680 and 13,330 $Bqkg^{-1}$ for ^{226}Ra, 249 and 504 $Bqkg^{-1}$ for ^{40}K, and 347 and 1,980 $Bqkg^{-1}$ for ^{232}Th.

The activity of any point located below the water-supplying area of the reservoir is equal to 2,260 $Bqkg^{-1}$ for ^{226}Ra, 534 $Bqkg^{-1}$ for ^{40}K, and 1,810 $Bqkg^{-1}$ for ^{232}Th.

Similarly, the anthropogenic ^{137}Cs isotope was measured. Its concentration ranged between 0.11 and 6.4 $kBqm^{-2}$ for all checked samples and it is similar to the average concentration in Poland: 0.11 – 23.68 $kBqm^{-2}$ [11]. In this case the highest activity of ^{137}Cs was observed on the northern and northwestern parts of the embankment as a result of topography.

This man-made radioactive isotope appeared in the ground after nuclear weapons tests, nuclear explosions in the stratosphere, and after the explosion of the atomic power station in Chernobyl in 1986 [8]. Those facts caused isotope ^{137}Cs to be included only in a thin layer of ground. The fall of the terrain promotes the rinsing out of cesium from the ground.

The area surrounding Rontok Duży Pond was leveled by coal mine waste rocks. The order of Polish Ministry Board from 2007 dealing with contents of natural radioisotopes ^{40}K, ^{226}Ra, and ^{232}Th in construction materials and in industrial wastes products used in construction gives the values of I_1 and I_2 factors. Their maximum values could not be higher than 20% of 3.5 for I_1 and 1,000 $Bqkg^{-1}$ for I_2 in relation to the stripping used for leveling in unsettled areas.

For those values of I_1 and I_2 the absorbed dose rate measured 1 m over the ground and must be below 0.3 μGyh^{-1}. This condition is performed by all of the checked samples (absorbed dose rate was between 0.0662 and 0.1449 μGyh^{-1}).

Seven samples of soil were taken from the Rontok Duży reservoir neighborhood. The results of I_1 factor are between 0.522 and 1.115, and of I_2 one between 35 and 155 $Bqkg^{-1}$. All of them are similar to those factors for the reference sample (I_1=0.537; I_2=33 $Bqkg^{-1}$).

Conclusions

All of the soil and sediment samples were analyzed for the presence of the natural radioactive isotopes ^{40}K, ^{226}Ra, and ^{232}Th, as well as the anthropogenic radionuclide ^{137}Cs.

The investigation shows that higher contamination is accumulated in the sediments at the bottom of the settling tank, especially on the intersection of inflow and outflow. The distribution of natural radioactive isotopes in adherent grounds and in sediments taken near the edge of the pond is similar to the typical values in Poland. The maximum activity concentration of ^{226}Ra was equal to 13,300 $Bqkg^{-1}$ and 1,980 $Bqkg^{-1}$ for ^{232}Th measured near the slurry-supplying pipe of the reservoir.

For all samples the calculated values of I_1 were obtained in the range 0.522 to 1.115, so below the 3.5 limit. There is no propagation of radioisotopes in the area surrounding the pond, which confirms the purposefulness of the applied ground leveling operation.

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Table 1. Activity concentration of radioisotopes in soil and sediment samples collected in the neighborhood of the Rontok Duży pond.

No.	Locations	Activity concentration of radionuclides [Bqkg^{-1}]			
		^{226}Ra	^{232}Th	^{40}K	^{137}Cs
Sediments taken near edge of the pond					
O_1	drain area to the Vistula river	75±2	69±1	607±12	23.1±0.7
O_2	west part, 100 m from the end of the dyke between main reservoir and buffer pond	31±1	39±1	337±9	37.2±1.1
O_3	north part, 100 m from the dyke between main reservoir and buffer pond	40±1	57±1	510±10	83±2
O_4	northeastern part below the dyke	31±1	53±1	461±11	27.9±0.9
O_5	water supplying area of the reservoir	2260±30	1810±30	530±20	NA
Kaniów pond	reference sample	26±1	54±1	448±9	2.1±0.1
Sediments from the bottom					
O_A		2690±30	1020±20	249±20	NA
O_B		2380±40	1170±50	251±13	NA
O_C		13330±160	1980±30	340±40	72±4
O_D		4710±60	580±10	454±11	42±1
O_E		2680±30	347±7	504±19	10.2±0.6
Soil					
G_1	northeast, the corn field	40±1	58±1	529±10	42±1
G_2	water supplying area of the reservoir	42±1	65±1	580±11	13.1±0.4
G_3	the dyke between main reservoir and buffer pond	155±2	83±1	551±9	1.8±0.2
G_4	western part of embankment	37±1	54±1	491±10	6.4±0.3
G_5	northwest, the field	35±1	53±1	428±10	146±4
G_6	forest on the north	35±1	54±1	467±9	88±2
G_7	northern part of embankment	40±1	59±1	454±10	46±1
Kaniów pond	reference sample	33±1	53±1	492±10	1.7±0.2

Table 2. I_1 and I_2 factors in different locations in the neighborhood of the Rontok Duży pond.

No.	Locations	I_1 and I_2 factors Absorbed Dose Rate		
		I_1 [1]	I_2 [Bqkg^{-1}]	D [μGyh^{-1}]
Soil				
G_1	northeast, the corn field	0.600±0.008	40±1	0.0757±0.0009
G_2	water supplying area of the reservoir	0.658±0.009	42±1	0.0830±0.0009
G_3	the dyke between main reservoir and buffer pond	1.115±0.010	155±2	0.1449±0.0012
G_4	western part of embankment	0.557±0.008	37±1	0.0703±0.0009
G_5	northwest, the field	0.522±0.008	35±1	0.0662±0.0009
G_6	forest on the north	0.542±0.008	35±1	0.0684±0.0008
G_7	northern part of embankment	0.582±0.008	40±1	0.0732±0.0009
Kaniów pond	reference sample	0.537±0.008	33±1	0.0679±0.0009

References

1. AROGUNJO A.M., HOLLRIEGL V., GIUSSANI A., LEOPOLD K., GERSTMANN U., VERONESE I., OEH U. Uranium and thorium in soils, mineral sands, water and food samples in a tin mining area in Nigeria with elevated activity. Journal of Environmental Radioactivity. pp. 232-240, **2009**.
2. WOŹNIAK G., COHN E.V.J. Monitoring of spontaneous vegetation dynamics on post coal mining waste sites in Upper Silesia, Poland. Geotechnical and Environmental Aspects of Waste Disposal Sits. Sarsby & Felton eds., London, pp. 289-294, **2007**.
3. KAPALA J., TOMCZAK M., ZALEWSKI M. Radioactivity of Building Materials Available in Northeastern Poland. Polish Journal of Environmental Studies, **10**, (3), 183, **2001**.
4. KUBICA B., MIETELSKI J.W., GOŁAŚ J., SKIBA S., TOMANKIEWICZ E., GACA P., JASIŃSKA M., TULEJA-KRYSA M. Concentration of ^{137}Cs, ^{40}K, ^{238}Pu and $^{239+240}Pu$ radionuclides and some heavy metals in soil samples from two main valleys from Tatra National Park. Polish Journal of Environmental Studies. **11**, (5), 537, **2002**.
5. MIETELSKI J.W., MACHARSKI P., JASIŃSKA A., BRODA R. Radioactive Contamination of forests in Poland. Biological Trace Element Research, pp. 715-723, **1994**.
6. XINWEI L. Natural radioactivity in some building materials and by – products of Shaanxi, China. Journal of Radioanalytical and Nuclear Chemistry. **262**, (3), 775, **2004**.
7. EUROPEAN COMMISSION. Radiation Protection 112: Radiological Protection Principles concerning the Natural Radioactivity of Building Materials. Directorate – General Environment, Nuclear Safety and Civil Protection, **1999**.
8. JANKOVIĆ M., TODOROVIĆ D., SAVANOVIĆ M., Radioactivity measurements in soil samples collected in the Republic of Srpska. Radiation Measurements, **43**, 1448, **2008**.
9. LEUNG K.C., LAU S.Y., POON C.B. Gamma radiation dose rate from radionuclides in Hong Kong soil. Journal of Environmental Radioactivity. **11**, 1487, **1990**.
10. UNSCEAR, Sources and effects of ionizing radiation. Report of the United Nations Scientific Committee on the Effects of Atomic Radiation to the General Assembly, New York, USA, **2000**.
11. BIERNACKA M., ISAJENKO K., LIPIŃSKI P. Monitoring of ^{137}Cs activity concentration in soil: years 2004-2006. Central Laboratory for Radiological Protection. Constitutional activity in year 2005 report. Warsaw, pp. 18-19, **2005**.
12. PAWUŁA A. On the need for use of Radiologic Criteria in Construction Site Evaluation. Symposium: Recent Engineering Geological Problems in Poland, Instytut Geologii UAM Poznań, WIND, Wrocław, pp. 83-88, **1998** [In Polish].