



Suitability of rocks and sediments from Brzeszcze and Silesia coal mines as building materials in terms of radiological hazard

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Abstract. The isotope activity concentration of rocks and bottom sediments was evaluated based on the samples collected from sedimentation ponds and gangue repositories. Radium ^{226}Ra , thorium ^{228}Th and potassium ^{40}K activities were measured by gamma spectrometry using high-purity germanium detector – HPGe 4020. The radiation effect resulting from the presence of natural radionuclides was estimated by radiological hazard indices such as f_1 and f_2 coefficients, radium equivalent, internal and external hazard indices and absorbed dose rate. Performed measurements and calculations have shown that the bottom sediments are most contaminated. They may pose a serious radiological hazard for present and future generations.

Keywords: dosing tanks • radiological hazard • K-40 • Ra-226 • Th-228

Introduction

Exploitation of coal is inevitably bound up with the necessity of dewatering of mines and removal of gangue. Water and rocks are treated as waste products. In Polish mines, both water and rocks contain elevated concentrations of natural radionuclides, mostly radium and thorium. These isotopes in water environment, due to coprecipitation or adsorption on particles, are removed from the water column and buried in bottom sediments. Waste rocks (ca. 0.4 t of rock per ton of coal derived from the Brzeszcze and Silesia coal mines) are stored in gangue repositories and form reservoirs of elevated concentrations of radioactive isotopes [1].

Radium ^{226}Ra , thorium ^{228}Th and potassium ^{40}K activity concentrations in rocks and sediments from Brzeszcze and Silesia mines have been used for the estimation of radiological hazard associated with exposure on radiation and potential usage of these materials in its pure form in construction industry. Coefficients f_1 and f_2 , radium equivalent, internal and external hazard indices and absorbed dose rate are the most important parameters used in the appraisal of rocks and sediments.

The problem of radioactivity of classical or new building materials is widely discussed in the literature [2–5], but there is lack of information about radiation hazard related to use of waste products in building industry. Three locations of sludge storage

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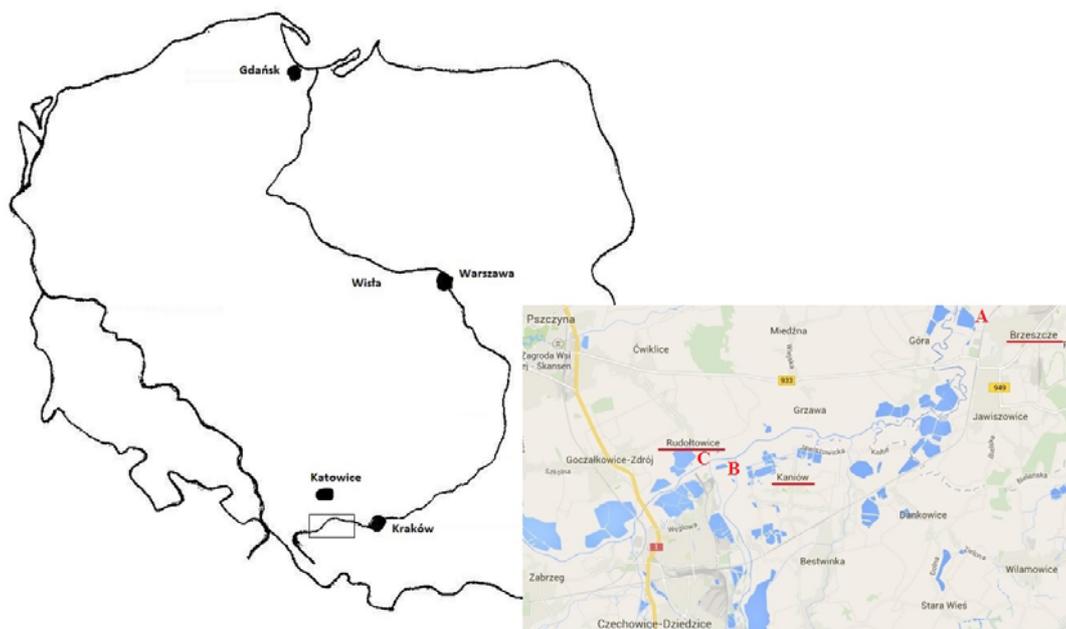


Fig. 1. Location of the studied area (Google maps).

have been chosen for the investigation of suitability of mining post-production wastes as construction materials. Two of them (Kaniów and Rontok Duży ponds) contain wastes from the Silesia coal mine, whereas in the third place wastes from the Brzeszcze mine are stored. The sedimentation ponds and stores of waste material are located in Małopolska and Silesia regions (Fig. 1). Bottom sediments and waste rocks stored in these locations have been never assessed in terms of radiological hazard arising from their application in building and construction industries.

Measurement methodology

Sampling collection

To estimate the radiological hazard from the waste products such as bottom sediments or rocks, the following sampling places were chosen: the gangue repositories adjoining the dosing tanks, embankments of the ponds and rivers as well as bottom sediments.

During the high water level, samples of bottom sediments were collected using a special gimlet. When the sediments were uncovered, samples were taken from the 10-cm thick surface layer. Samples were collected at the inlet area, the coastal zone and the middle part of reservoirs. Location of the bottom sediment sampling points within the investigated water reservoirs is shown in Fig. 2. The rock samples were collected from enumerated locations representing different types of rocks. The weight of each sample was equal to 1–2 kg. All samples were collected between 2009 and 2013.

Gamma spectrometry measurements

Collected samples were dried for 24 hours at 105°C, crumbled, homogenized and sieved. Then, they were

closed in aluminium cylindrical containers (70 mm in diameter, 31.5 mm height). After 21 days when the radioactive equilibrium between radium and its short-lived decay products was achieved, the activity concentration was measured using high-purity germanium detector Canberra HPGe 4020. The detector's energy resolution was equal to 0.8 and 0.2% for the 122 and 1332 keV energy peaks, respectively.

The concentration of radioactive isotopes such as radium ^{226}Ra and thorium ^{228}Th was determined by their decay products. The radium activity was determined by ^{214}Bi (609.3, 1120.3 and 1764.5 keV) and that of thorium by ^{208}Tl (583.2 and 2614.5 keV). As well, the concentration of potassium ^{40}K was determined from its peak at 1460.8 keV. The detection limit was equal to $1 \text{ Bq}\cdot\text{kg}^{-1}$ for radium ^{226}Ra , thorium ^{228}Th and potassium ^{40}K . Counts were collected for 20 hours [2, 6]. Calibration was performed using RGK-1, RGU-1 and RGTh-1 reference materials distributed by the International Atomic Energy Agency (IAEA). The relative uncertainties of the specific activity for these materials are equal to 0.33, 0.25 and 1%, respectively [7].

Indicators of radiological hazard

Measured concentrations of natural radioisotopes in rocks and bottom sediments exceed typical values recorded in soils in Poland. The assessment of their suitability as construction materials is a subject of legal regulations of the Polish Ministry Board [8]. These regulations concern ^{40}K , ^{226}Ra and ^{228}Th isotopes and are in accordance with the recommendations of European Commission [9, 10]. They define two coefficients, f_1 and f_2 , which should be calculated for each building material and compared with reference values resulting from maximum allowable concentrations of the mentioned radionuclides. Other radiological exposure indicators are

also used in the literature with the aim of better determination of risk from ionizing radiation.

It is worth to mention here about differences in thorium isotope treatment in Polish and European legislations. In the calculation of parameters determining radiological hazard, Polish regulations recommend ^{228}Th instead of ^{232}Th . Therefore, in calculations

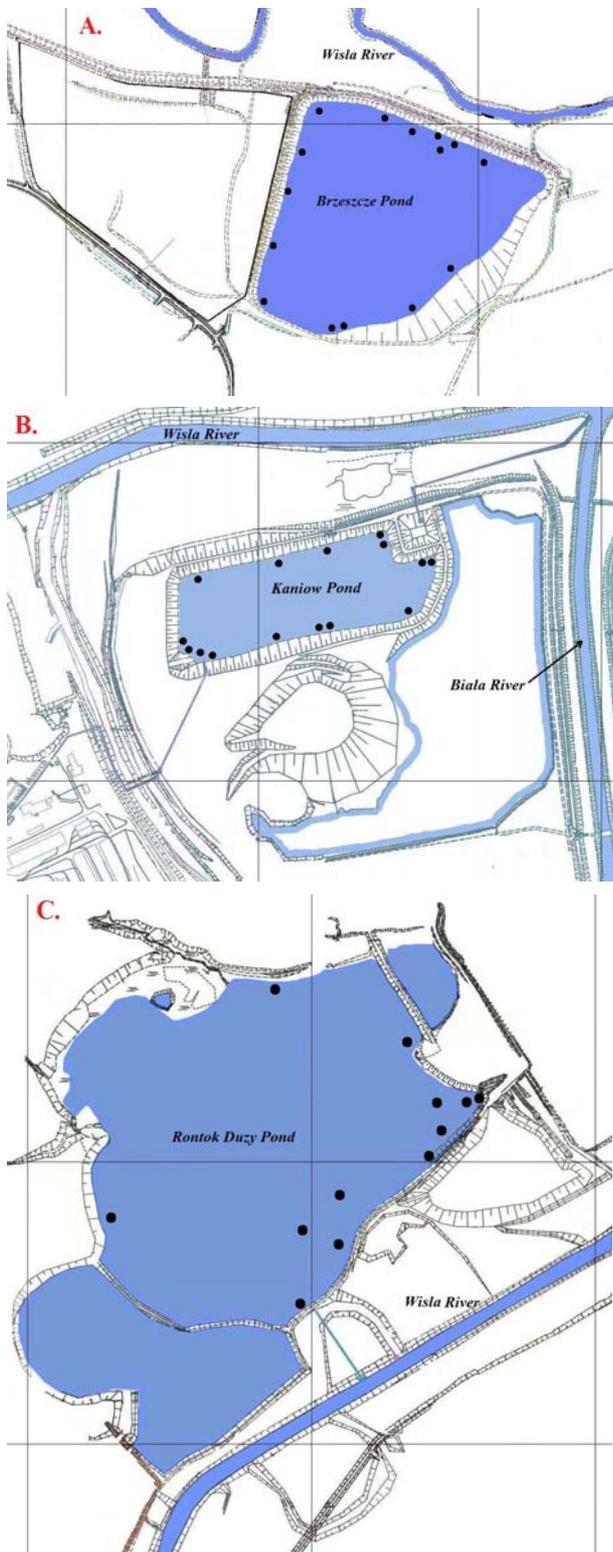


Fig. 2. The investigated area: (A) Brzeszcze pond, (B) Kaniów pond and (C) Rontok Duży pond. Black dots represent the individual sampling points of bottom sediments.

of all radiological exposure indices, the ^{228}Th activity was consequently used in place of ^{232}Th .

f_1 and f_2 factors

The f_1 activity factor defines the content of natural radioactive isotopes such as radium ^{226}Ra , thorium ^{228}Th and potassium ^{40}K in materials and wastes used as construction materials. It describes the full body exposure to gamma radiation from these radionuclides. The f_2 factor informs indirectly about the radiation exposure of radon and its short-lived decay products [8, 9].

The f_1 and f_2 factors are defined by the following equations:

$$(1) \quad f_1 = \frac{C_K}{3000 \text{ Bq} \cdot \text{kg}^{-1}} + \frac{C_{\text{Ra}}}{300 \text{ Bq} \cdot \text{kg}^{-1}} + \frac{C_{\text{Th}}}{200 \text{ Bq} \cdot \text{kg}^{-1}}$$

$$f_2 = C_{\text{Ra}}$$

where C_K , C_{Ra} and C_{Th} are the activity concentrations of ^{40}K , ^{226}Ra and ^{228}Th (in $\text{Bq} \cdot \text{kg}^{-1}$), respectively.

For building and waste materials, regulation of the Polish Ministry Board gives admissible values of f_1 and f_2 coefficients, depending on the type of construction and its purpose:

- 1) $f_1 = 1$; $f_2 = 200 \text{ Bq} \cdot \text{kg}^{-1}$ for the building materials used in buildings intended for human or livestock accommodation,
- 2) $f_1 = 2$; $f_2 = 400 \text{ Bq} \cdot \text{kg}^{-1}$ for waste products used in buildings placed on built-up area or for levelling of land designated for construction area,
- 3) $f_1 = 3.5$; $f_2 = 1000 \text{ Bq} \cdot \text{kg}^{-1}$ for waste products used in buildings and for levelling of land designated for construction area not listed in 2), and
- 4) $f_1 = 7$; $f_2 = 2000 \text{ Bq} \cdot \text{kg}^{-1}$ for waste products used in underground construction (railways and road tunnels).

Additionally, on the areas where industrial wastes were used for levelling, construction of roads or sport and recreation objects the absorbed dose rate at 1 m above ground level should be reduced to value not exceeding $0.3 \mu\text{Gy} \cdot \text{h}^{-1}$, in particular by covering an additional layer of other materials.

Radium equivalent

Radium equivalent Ra_{eq} is defined by equation [11]:

$$(2) \quad \text{Ra}_{\text{eq}} = C_{\text{Ra}} + 1.47C_{\text{Th}} + 0.077C_K$$

where C_K , C_{Ra} and C_{Th} are the activity concentrations of ^{40}K , ^{226}Ra and ^{232}Th (in $\text{Bq} \cdot \text{kg}^{-1}$), respectively.

It is assumed that $\text{Ra}_{\text{eq}} = 370 \text{ Bq} \cdot \text{kg}^{-1}$ is a limit value below which the radiological exposure is treated as negligible.

External and internal hazard indices

The value of the external exposure index H_{ex} is defined by equations [10, 11]:

$$(3) \quad H_{ex} = \frac{C_{Ra}}{370 \text{ Bq} \cdot \text{kg}^{-1}} + \frac{C_{Th}}{259 \text{ Bq} \cdot \text{kg}^{-1}} + \frac{C_K}{4810 \text{ Bq} \cdot \text{kg}^{-1}}$$

where C_K , C_{Ra} and C_{Th} are the activity concentrations of ^{40}K , ^{226}Ra and ^{232}Th (in $\text{Bq} \cdot \text{kg}^{-1}$), respectively.

The external hazard index is closely related to the radium equivalent Ra_{eq} . The value of $H_{ex} = 1$ corresponds to the radium equivalent Ra_{eq} equal to $370 \text{ Bq} \cdot \text{kg}^{-1}$. At this index value, radiological exposure is treated as negligible.

Apart from external index, an internal hazard index H_{in} is defined [10, 11]. It expresses a threat from radon and its short-lived decay products and is defined by the equation:

$$(4) \quad H_{in} = \frac{C_{Ra}}{185 \text{ Bq} \cdot \text{kg}^{-1}} + \frac{C_{Th}}{259 \text{ Bq} \cdot \text{kg}^{-1}} + \frac{C_K}{4810 \text{ Bq} \cdot \text{kg}^{-1}}$$

where C_K , C_{Ra} and C_{Th} are the same as mentioned earlier.

The limit value of this coefficient is 1, which corresponds to half of the maximum permissible concentration of radium ^{226}Ra .

Absorbed dose rate 1 m above the ground level

Total absorbed dose rate D is defined by equation [12]:

$$(5) \quad D = 0.462C_{Ra} + 0.604C_{Th} + 0.041C_K$$

where D is the absorbed dose rate at 1 m above the ground level (in $\text{nGy} \cdot \text{h}^{-1}$), C_K , C_{Ra} and C_{Th} are the activity concentrations of ^{40}K , ^{226}Ra and ^{232}Th (in $\text{Bq} \cdot \text{kg}^{-1}$), respectively.

In calculations, the presence of other radionuclides such as cesium ^{137}Cs , strontium ^{90}Sr and other isotopes from uranium series is neglected because of their low concentration in materials and thereby a small contribution to the absorbed dose.

Results and discussion

Results of radium ^{226}Ra , thorium ^{232}Th and potassium ^{40}K activity measurements and calculated indices of radiological hazard for waste rocks and bottom sediments are presented in Table 1.

The typical activities of natural radionuclides in rocks and sediments at the area of Poland are given in Radiological Atlas of Poland 2005 [13]:

- radium ^{226}Ra – from 4.2 to $116 \text{ Bq} \cdot \text{kg}^{-1}$; mean $25 \text{ Bq} \cdot \text{kg}^{-1}$,
- thorium ^{232}Th – from 3.6 to $82.8 \text{ Bq} \cdot \text{kg}^{-1}$; mean $23.4 \text{ Bq} \cdot \text{kg}^{-1}$,
- potassium ^{40}K – from 60 to $1028 \text{ Bq} \cdot \text{kg}^{-1}$; mean $408 \text{ Bq} \cdot \text{kg}^{-1}$.

As shown in Table 1, the measured activities of these three radionuclides in bottom sediment samples collected from all investigated tanks exceed

Table 1. Results of radium ^{226}Ra , thorium ^{232}Th and potassium ^{40}K activity measurements and calculated radiological hazard indices for waste rocks and bottom sediments on the area of investigations (*number of samples; **average relative uncertainties)

Parameters	Kaniów (14*)		Brzeszcze (10*)		Kaniów (15*)		Rontok Duży (12*)		Brzeszcze (15*)	
	Waste rocks		Bottom sediments		Bottom sediments		Bottom sediments		Bottom sediments	
^{226}Ra [Bq/kg]	14.0–85.9 (5.2%**)	30.9–75 (6%)	29.7–58.5 (4.6%)	1043–35 490 (3%)	96.7–95 510 (3%)	107.9–3588 (3%)	96.7–95 510 (3%)	48.9–26 150 (4.4%)	60.1–1817 (3.6%)	107.9–3588 (3%)
^{232}Th [Bq/kg]	9.8–85.3 (5.2%)	27.5–50 (5.6%)	24.4–71.1 (4.8%)	584–18 740 (3.6%)	48.9–26 150 (4.4%)	60.1–1817 (3.6%)	48.9–26 150 (4.4%)	249–1528 (10.6%)	299–561 (7.4%)	60.1–1817 (3.6%)
^{40}K [Bq/kg]	22.1–739 (10.6%)	337–607 (4.8%)	94.8–712 (4.4%)	423–1620 (10%)	249–1528 (10.6%)	299–561 (7.4%)	249–1528 (10.6%)	0.76–449.6 (4.8%)	0.85–21.2 (2.5%)	299–561 (7.4%)
f_1 [-]	0.1–0.9 (3.4%)	0.35–0.70 (3.2%)	0.28–0.73 (3%)	6.4–212.6 (2.5%)	0.76–449.6 (4.8%)	0.85–21.2 (2.5%)	0.76–449.6 (4.8%)	96.7–95 510 (3%)	107.9–3588 (3%)	0.85–21.2 (2.5%)
f_2 [Bq/kg]	14.0–85.9 (5.2%)	30.9–75 (4.6%)	29.7–58.5 (4.8%)	1043–35 490 (3%)	96.7–95 510 (3%)	107.9–3588 (3%)	96.7–95 510 (3%)	212.6–134 000 (2.5%)	239.5–6295 (2.5%)	107.9–3588 (3%)
Ra_{eq} [Bq/kg]	30.1–251.1 (3.4%)	97.3–195.2 (3.4%)	80.7–203.7 (3%)	1896–63 170 (2.5%)	212.6–134 000 (2.5%)	239.5–6295 (2.5%)	212.6–134 000 (2.5%)	0.57–359.4 (2.5%)	0.64–16.8 (2.5%)	239.5–6295 (2.5%)
H_{ex} [-]	0.08–0.67 (3.4%)	0.26–0.52 (3.2%)	0.22–0.54 (3%)	5.1–168.6 (2.5%)	0.57–359.4 (2.5%)	0.64–16.8 (2.5%)	0.57–359.4 (2.5%)	0.83–617.6 (2.5%)	0.93–26.5 (2.5%)	0.64–16.8 (2.5%)
H_{in} [-]	0.12–0.90 (3.4%)	0.34–0.73 (3.4%)	0.33–0.70 (3%)	7.9–264.5 (2.5%)	0.83–617.6 (2.5%)	0.93–26.5 (2.5%)	0.83–617.6 (2.5%)	0.098–60 (2.5%)	0.11–2.77 (2.5%)	0.93–26.5 (2.5%)
D [$\mu\text{Gy/h}$]	0.013–0.115 (3.6%)	0.045–0.090 (3.6%)	0.036–0.092 (3%)	0.84–27.8 (2.5%)	0.036–0.092 (3%)	0.11–2.77 (2.5%)	0.036–0.092 (3%)			0.11–2.77 (2.5%)

typical values for the territory of Poland. Specific activities of the individual isotopes in the bottom sediments from the Brzeszcze pond are in the range 107.9–3588 Bq·kg⁻¹ for ²²⁶Ra, 299–561 Bq·kg⁻¹ for ⁴⁰K and 60.1–1817 Bq·kg⁻¹ for ²²⁸Th. The activities of sediments from the Kaniów pond are between 1043 and 35 490 Bq·kg⁻¹ for ²²⁶Ra, 423 and 1620 Bq·kg⁻¹ for ⁴⁰K, and 584 and 18 740 Bq·kg⁻¹ for ²²⁸Th. For the Rontok Duży pond, isotope activities in bottom sediments are in the range of 96.7–95 510 Bq·kg⁻¹ for ²²⁶Ra, 249–1528 Bq·kg⁻¹ for ⁴⁰K and 48.9–26 150 Bq·kg⁻¹ for ²²⁸Th. The maximum values of activity concentration in bottom sediments of the Rontok Duży and Kaniów pond are observed in the water inflow zones. In general, in the Brzeszcze reservoir the activity concentrations in bottom sediments are lower than in the Kaniów and Rontok Duży ponds. It may reflect the different mineralogical compositions of sediments. In the Brzeszcze pond, the sediments are dominated by quartz and calcite while in Kaniów and Rontok Duży reservoirs kaolinite, muscovite and barite are the predominant mineralogical phases.

The observed variability of isotope activities of bottom sediments from individual ponds reflects also differences in chemical composition of inflowing mine waters. Water from the Silesia coal mine supplying Kaniów and, in the past – Rontok Duży pond, is enriched with radium, barium and sulphate ions. This leads to the precipitation of barium sulphate. As a consequence, the radium coprecipitation takes place. In contrast, water from the Brzeszcze mine is almost barium free and so the coprecipitation of radium is very limited. Additionally, radium and other isotopes can be buried in bottom sediments as a result of adsorption on the surface of settling particles. However, the studies of sedimentation and transport processes were beyond the scope of this work.

Specific activities of radionuclides observed in waste rock samples taken from all investigated sites fall within the range of typical values for the territory of Poland. In part of samples from the Kaniów pond, the observed level of ⁴⁰K activity was even lower. Waste rocks in all sites located near the Brzeszcze, Kaniów and Rontok Duży ponds are similar in terms of their petrography. The highest concentration of radionuclides was measured in rocks dominated by silty and mudstone shales. The lowest isotope activities were recorded in coal, independent of its origin.

According to national and international regulations expressed in terms of indices of radiological hazard (Tables 1 and 2), gangue can be used as construction material even in densely populated areas. The f_1 and f_2 factors calculated for them are lower than 1 and 200 Bq/kg, respectively. The maximum value of radium equivalent is equal to 251.1 Bq/kg, whereas the internal and external hazard indices are lower than 1. Calculated values of the individual indices allow the use of gangue stored in repositories in the construction of roads, ponds and river embankments.

Bottom sediments from the Brzeszcze retention pond are suitable for underground construction (railways and road tunnels). Calculated f_1 and f_2

Table 2. Results of radium ²²⁶Ra, thorium ²²⁸Th and potassium ⁴⁰K activity measurements and calculated radiological hazard indices for different types of waste rock samples on the area of investigations (number of samples per type of rock are given in brackets)

Rock	²²⁶ Ra [Bq/kg]	²²⁸ Th [Bq/kg]	⁴⁰ K [Bq/kg]	f_1 [-]	f_2 [Bq/kg]	Ra _{eq} [Bq/kg]	D [μGy/h]	H _{ex} [-]	H _{in} [-]
Kaniów									
Coal (2)	14.0 ± 1.4	9.8 ± 1.0	22.1 ± 6.4	0.103 ± 0.016	14.0 ± 1.4	30.1 ± 2.0	0.013 ± 0.010	0.080 ± 0.006	0.118 ± 0.008
Claystone (3)	27.6 ± 1.6	28.4 ± 1.4	48.9 ± 6.8	0.250 ± 0.010	27.6 ± 1.6	73.1 ± 2.6	0.032 ± 0.004	0.194 ± 0.008	0.269 ± 0.010
Silty shale (3)	85.9 ± 2.6	73.7 ± 2.6	739 ± 24	0.901 ± 0.018	85.9 ± 2.6	251.1 ± 5.0	0.115 ± 0.004	0.670 ± 0.014	0.902 ± 0.018
Mudstone shale with silica (3)	68.7 ± 2.2	85.3 ± 2.8	590 ± 20	0.852 ± 0.018	68.7 ± 2.2	239.5 ± 5.0	0.1075 ± 0.0022	0.638 ± 0.014	0.823 ± 0.016
Sandstone laminating mudstone (3)	32.8 ± 1.4	34.6 ± 1.4	443 ± 15	0.430 ± 0.010	32.8 ± 1.4	117.8 ± 2.8	0.0542 ± 0.0012	0.314 ± 0.008	0.403 ± 0.010
Brzeszcze									
Mudstone (3)	58.5 ± 2.2	71.1 ± 2.8	528 ± 22	0.727 ± 0.018	58.5 ± 2.2	203.7 ± 5.0	0.0916 ± 0.0022	0.542 ± 0.014	0.701 ± 0.016
Mudstone shale (3)	56.7 ± 2.0	55.6 ± 2.0	621 ± 20	0.674 ± 0.014	56.7 ± 2.0	186.3 ± 3.8	0.0853 ± 0.0018	0.497 ± 0.010	0.650 ± 0.016
Coal (2)	37.5 ± 1.8	24.4 ± 1.4	94.8 ± 8.2	0.279 ± 0.010	37.5 ± 1.8	80.7 ± 2.8	0.0359 ± 0.0012	0.215 ± 0.008	0.327 ± 0.012
Sandstone (2)	29.7 ± 1.6	35.9 ± 1.6	712 ± 24	0.516 ± 0.012	29.7 ± 1.6	137.3 ± 3.4	0.0646 ± 0.0016	0.367 ± 0.010	0.447 ± 0.012

coefficients for most of them are lower than 7 and 2000 Bq/kg, respectively. However, bottom sediments from Kaniów and Rontok Duży tanks may create serious problems in future because they do not meet radiological standards. As shown in Table 1, for these sediments all exposure factors are higher than recommended. For the Kaniów pond, the f_1 and f_2 factors for bottom sediments are between 14.4 and 212.6 and between 2394 and 35 490 Bq/kg, respectively. The maximum value of radium equivalent is equal to 63 170 Bq/kg, which is exceeding 170 times the value representing negligible radiological exposure. The absorbed dose rate is also very high, between 0.8 and 27.8 $\mu\text{Gy/h}$. For the Rontok Duży reservoir, the maximum value of f_1 factor is close to 450 and f_2 is greater than 95 500 Bq/kg. Both internal and external hazard indices calculated for bottom sediments are in most instances greater than 1 (maximum value are equal to 618 and 359, respectively). The maximum value of the absorbed dose rate is equal to 60.0 $\mu\text{Gy/h}$.

Conclusions

The results of the performed isotope investigations indicate the possibility of disposal of waste rocks from the Brzeszcze and Silesia coal mines. They can be used as materials for surface or underground constructions. Gangue taken from both coal mines can be used as construction material even in the densely populated areas. It could also be used for the construction of roads or ponds and river embankments and for levelling of land designated for construction area. Bottom sediments from the Brzeszcze retention pond are suitable for underground construction (railway and road tunnels).

Radiological hazard indices calculated for bottom deposits of Kaniów and Rontok Duży reservoirs exceed many times the values recommended in legal acts. These sediments in their pure form are not suitable for any use in construction industry. Reclamation of these deposits will create a serious problem in the future.

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