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Natural radioactivity of groundwater from the Przerzeczyn-Zdrój Spa

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Abstract. The present authors performed investigations of natural radioactivity in groundwater from the Przerzeczyn-Zdrój Spa. Some of the waters are regarded as medical and are used for balneological purposes. Samples from seven groundwater intakes were collected 5 times over a period of 8 years (1999–2007). In order to obtain necessary data, two different nuclear spectrometry techniques were applied: α spectrometry and liquid scintillation spectrometry. The activity concentrations of ^{222}Rn varied in the range from 15±2 Bq/l to 154±22 Bq/l. The results of activity concentrations of ^{226,228}Ra varied from below 10 mBq/l to 30±1.5 mBq/l and from below 30 mBq/l to 60±4 mBq/l, respectively. Activity concentration lower than minimum detectable activity (MDA) was obtained for 3 samples for ^{226}Ra and 4 for ^{228}Ra determinations out of 7 investigated samples. The uranium content in the studied samples was determined once and the value ranged from 4.5±0.6 mBq/l to 13.6±1.2 mBq/l for ^{238}U and from 17.1±0.9 mBq/l to 52.2±2.8 mBq/l for ^{234}U. All obtained values for uranium isotopes showed activity concentrations above MDA. The activity ratios ^{234}U/^{238}U, ^{222}Rn/^{226}Ra and ^{226}Ra/^{238}U and the correlations between different isotopes concentrations were evaluated.

Key words: radon • radium • uranium • groundwater • medicinal water

Introduction

The aim of the investigations was to evaluate activity concentrations of naturally occurring radioactive isotopes in seven presently exploited groundwater intakes in the Przerzeczyn-Zdrój Spa. The characteristics of both the studied intakes and of the intakes previously used for balneological purposes together with a short specification of the water are presented in Table 1.

Radioactivity of groundwater is mainly connected with the presence of dissolved long-lived uranium ^{234,238}U and radium ^{226,228}Ra isotopes. In waters of shallow circulation, which are usually also of low mineralization, gaseous isotope radon ^{222}Rn plays a major role. Tritium ^{3}H, potassium ^{40}K and carbon ^{14}C isotopes which are also present in the waters affect the radioactivity only insignificantly. However, they are present in small concentrations in almost all groundwaters, especially in waters of current infiltration. Other natural radioactive isotopes, i.e. ^{232}Th, ^{210}Pb, ^{210}Po, ^{7}Be, ^{22}Na, ^{87}Rb, are practically meaningless in the groundwater environment [6, 15].

Localization of the Przerzeczyn-Zdrój Spa on the geological sketch of SW part of Poland is shown in Fig. 1. Przerzeczyn-Zdrój is the only spa situated on the Fore-Sudetic block. Geological structures of this unit are the continuation of the Sudety Mts. structures and constitute a part of the NE side of the Fore-Sudetic block fault.

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Table 1. Characteristics of intakes, medicinal waters and remaining groundwaters exploited in the past and currently by the Przerzeczyn-Zdrój Spa based on data from 1937–2004, from archive materials of the Spa. Bold font indicates specific components of waters giving them therapeutic properties

Name of intake	Specific medicinal waters					Other exploited groundwaters		
	Radon-sulphide rich water		Radon water		Sulphide water			
	Spring "Siarczkowe"*	Borehole no. 2	Borehole no. 8	Borehole no. 13	Borehole no. 9	Borehole no. 11	Borehole no. 15	Borehole no. 10
Depth (m.b.g.l.)***	6	77	80	79	151	40	98	182
Water bearing horizon (m.b.g.l.)***	5–6	69.5–77.0	72–80	65–79	53–151	14–40	91–98	20–
Lithology	Alluvial sand and silt	Granodiorite, serpentinite	Rubble, gneiss	Granodiorite, serpentinite, gneiss	Gneiss, serpentinite	Serpentinite	Gneiss	–
Stratigraphy	Quaternary	Palaeozoic	Neogene Proterozoic	Palaeozoic Proterozoic	Proterozoic Palaeozoic	Palaeozoic	Proterozoic	–
Discharge (m^3/h)	1.08	3.7	3.5	3.25	3.7	1.2	4.3	–
Depression (m)	0.5	9.0	7.3	42.6	9.0	14.7	32	natural outflow
T (°C)	8.0–12.3	8.4–14.4	10.0–11.8	9.2–12.7	10.8–12.1	9.5–12.2	11.7–13.7	11.0–11.3
pH	7.3–8.10	7.4–8.1	7.3–8.6	7.3–8.3	7.0–8.5	7.6–8.6	7.1–8.3	7.8–8.0
TDS (mg/dm^3)	291–456	372–563	518–575	488–567	344–454	318–573	417–556	532–565
Hydrochemical type	HCO_3-Ca-Mg-(Na)	HCO_3-Ca-Mg	HCO_3-Mg-Ca	HCO_3-Mg-Ca	HCO_3-Mg-Ca	HCO_3-Mg-Ca	HCO_3-Mg-Ca	HCO_3-Mg-Ca
HCO_3^- content (mg/dm^3)	208–305	233–341	278–374	291.6–399.0	208.7–293.1	190.3–398.0	317.0–399.6	354.0–387.4
SO_4^{2-} content (mg/dm^3)	3.0–19.5	3.5–27.5	19.3–36.6	7.0–21.0	3.0–12.35	9.0–25.7	7.0–12.8	17.1–31.3
Cl^- content (mg/dm^3)	6.4–17.2	10.3–35.4	12.0–16.0	3.4–6.9	7.1–14.2	5.2–13.8	3.5–7.0	10.6–11.5
Ca^{2+} content (mg/dm^3)	34.1–64.9	35.2–66.1	47.1–53.9	41.1–56.7	33.1–44.5	15.7–56.7	36.4–52.0	48.3–51.3
Mg^{2+} content (mg/dm^3)	9.4–23.7	19.1–31.0	41.4–43.7	38.0–41.8	18.9–28.7	22.8–42.0	37.2–42.8	41.4–44.8
Na^+ content (mg/dm^3)	18.0–39.6	20.0–28.7	14.0–20.0	17.0–19.5	19.0–23.0	16.0–22.0	17.0–18.5	14.1–17.4
H_2S content (mg/dm^3)	**0.34–3.06**	**0.43–2.21**	0.0	0.0	**0.68–2.04**	0.0	0.0	0.0
^{222}Rn content (Bq/dm^3)	**33.3–92.5**	**18.5–133.2**	**81.4–215.7**	**62.9–122.1**	**37–188****	7.4–74.0	14.2–47.4	25.9–48.1

* – (Sulphide spring) not exploited since November 1991.

** – Analyses performed after 1994 did not show ^{222}Rn activity concentrations higher than 74 Bq/l.

*** – m.b.g.l. – meters below ground level.

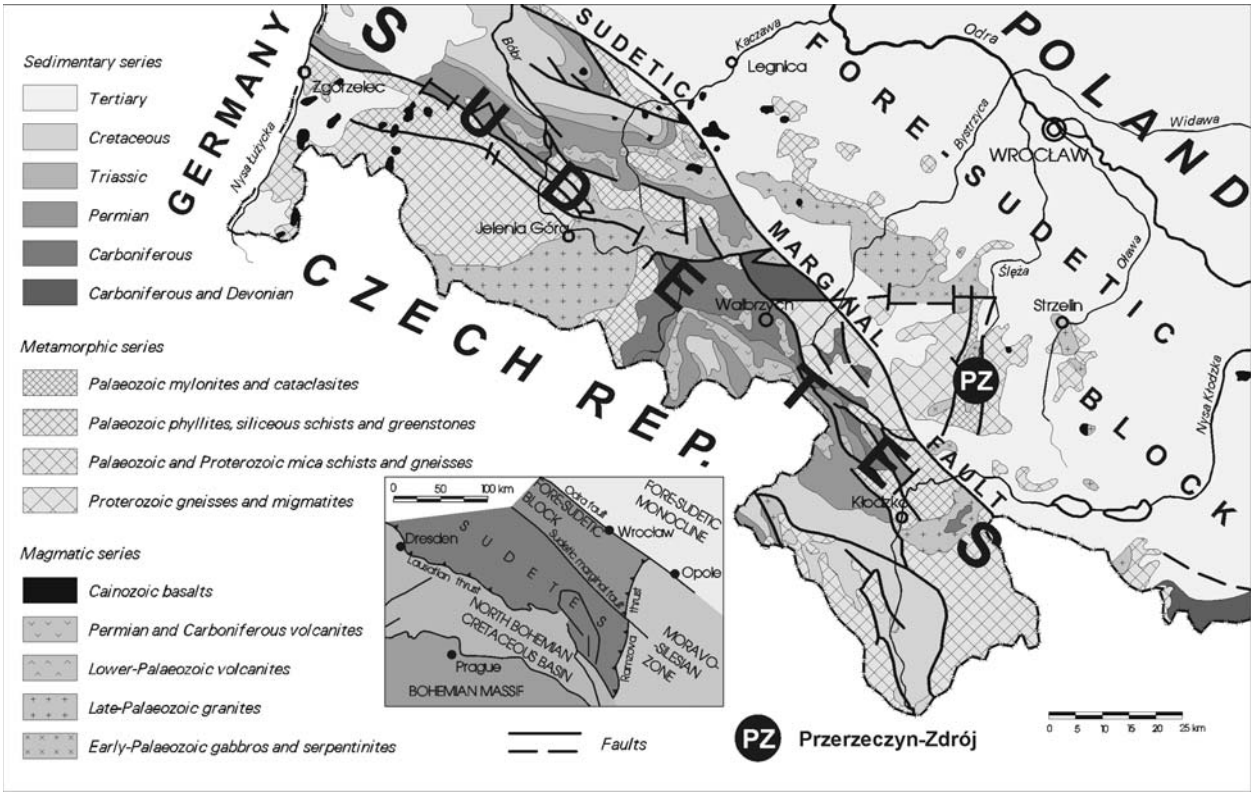

Fig. 1. Localization of the Przerzeczyn-Zdrój Spa on the geological sketch of SW part of Poland.

The Sudety Mts. have a mosaic geological structure which is a consequence of their long evolution beginning in the Precambrian. They are mainly built of crystalline rocks which due to many deformations and weathering processes are strongly cracked. This in effect favours the migration of groundwater and gases. Therefore, the Sudety Mts. together with the Fore-Sudetic block are rich in groundwater. Waters from these regions often contain radon, radium as well as uranium. This is due to the presence of the latter two isotopes in the reservoir rocks [22, 24, 25]. However, until now groundwater from the Fore-Sudetic block, unlike the Sudety Mts. waters, have not been a subject of detailed investigations, especially concerning radioactivity. Books published by Kozłowska and Przylibski [10, 23] are the only available ones in this field. This has stimulated the authors to undertake the investigations of natural radioactivity found in the groundwater of the Przerzeczyn-Zdrój Spa.

Groundwaters of the Przerzeczyn Spa recognized as medicinal ones are sulphide, radon or radon-sulphide waters of HCO_3-Mg-Ca chemical type. They flow out within Paleozoic (Carboniferous) granitoid rocks with low pyrite mineralization of the Niemcza shear zone. Granitoids are interlayered by mylonite gneisses and other rocks of upper Paleozoic age. On the depth of several dozen meters below ground level Devonian serpentinites are also present. The contact between these serpentinites and granitoids is of tectonic character. The above-mentioned rocks lie under a cover of Neogene and Pleistocene sedimentary rocks. Holocene rocks are formed locally, especially in the valleys of the Ślęża river and its tributaries [5].

The region of Southern Poland, especially the Sudety Mts., is characterized by an increased gamma radiation background [9]. This is due to not only higher radium content in the soil than in other regions, but also the presence of uranium. Groundwater flowing through reservoir rocks containing higher amounts of radioactive isotopes may, when the conditions are favourable, transport these elements to considerable distances. Moreover, water passing through rocks containing ^{226}Ra and ^{234,238}U isotopes usually become enriched in ^{222}Rn. Radon is a noble, water-soluble gas. It can be transported with water to distances usually not exceeding a hundred meters but sometimes reaching even a few hundred meters.

Measurement methods

The samples were collected in polyethylene bottles, acidified with HCl for radium and with HNO_3 for uranium determinations in order to avoid radionuclide precipitation and adsorption on the walls of the containers. Samples of radium and radon from seven water springs were collected 5 times over a period of 8 years (1999–2007). Sampling of uranium was performed once in 2007. The information about water usage is presented in Table 2.

The measurements of ^{226,228}Ra and ^{222}Rn activity concentrations in the investigated samples were performed with the use of a 1414 WinSpectral α/β liquid scintillation counter from Wallac [34]. For ^{222}Rn determination, 10 ml of analyzed water was drawn by a disposable syringe and transferred to a scintillation vial already filled with 10 ml of a scintillation solution Insta-Fluor (Canberra-Packard). This method is based on ISO norm [29]. Radon was extracted from the water phase to the liquid scintillator phase. ^{222}Rn content in water samples was calculated from the α – part of the spec-

Table 2. Mineralization (TDS), water destination and activity concentrations of ^{222}Rn, ^{226,228}Ra and ^{234,238}U for waters from the Przerzeczyn-Zdrój Spa. For ^{222}Rn, ^{226,228}Ra and ^{234,238}U values ±SD (*n*)*

Borehole	Water destination/characteristics	TDS (mg/l)	^{222}Rn (Bq/l)	^{226}Ra (mBq/l)	^{228}Ra (mBq/l)	^{238}U (mBq/l)	^{234}U (mBq/l)
No. 2	Bathing-in sanatorium	421	69 ± 4 (5)	30.0 ± 1.5 (4)	60.0 ± 4.0 (1)	4.5 ± 0.6 (1)	20.7 ± 1.5 (1)
No. 8	Drinking water, bathing-in sanatorium	553	154 ± 22 (5)	21.0 ± 2.1 (4)	35.0 ± 2.0 (1)	11.2 ± 1.0 (1)	37.7 ± 2.6 (1)
No. 9	Bathing-in sanatorium	411	56 ± 5 (5)	< MDA	< MDA	12.7 ± 0.9 (1)	52.2 ± 2.8 (1)
No. 10	Free intake, bathing	545	36 ± 3 (5)	< MDA	< MDA	13.6 ± 1.2 (1)	49.3 ± 3.3 (1)
No. 11	Municipal water	506	15 ± 2 (5)	< MDA	< MDA	5.6 ± 0.4 (1)	17.1 ± 0.9 (1)
No. 13	Municipal water	536	87 ± 5 (5)	29.0 ± 0.7 (4)	53.0 ± 5.0 (3)	8.9 ± 0.7 (1)	41.7 ± 2.3 (1)
No. 15	Municipal water	537	18 ± 2 (5)	11.6 ± 0.9 (4)	< MDA	12.3 ± 1.4 (1)	42.4 ± 3.7 (1)

* (*n*) – number of individuals.

trum originating from this isotope and its α-radioactive daughters, taking into account the chemical recovery of radon transfer from water to the scintillation solution. MDA for ^{222}Rn determination was equal to 1 Bq/l for 1 h counting time [4].

The procedure for chemical treatment of samples for the determination of radium was based on the radiochemical preconcentration by coprecipitation with $BaSO_4$ and purification of its derivatives. This method was described in details elsewhere [21]. Measurements were performed once a day for 1 h counting time for a period of one month until secular equilibrium between ^{226}Ra and its daughters was reached. ^{228}Ra content was determined from the high-energy β component originating from its daughter ^{228}Ac which reached the radioactive equilibrium state with ^{228}Ra after 40 h. The theoretical calculations were based on Bateman equations for each radium isotope series [1]. Fitting theoretical curves to experimental points allowed to determine ^{226}Ra and ^{228}Ra contents in the studied samples. For the measurement lasting 1 h the MDA value was equal to 0.01 Bq/l and 0.03 Bq/l for ^{226}Ra and ^{228}Ra, respectively.

The determination of ^{234,238}U was performed with the use of α spectrometer 7401VR from Canberra-Packard, USA and a silicon surface barrier detector with a surface area of 300 mm^2. The radiochemical separation of uranium from other radionuclides present in the waters was based on a slightly modified procedure published by Suomela [30] and described in [11]. At the beginning of each analysis, a standard ^{232}U (NIST, USA) of known activity was added to a 0.5 l water sample. Uranium was coprecipitated with an ammoniac alkaline earth phosphate precipitate. Then, the precipitate was separated from the solution and dissolved in 8 M HCl. Uranium was separated and purified on an anion exchange resin Dowex 1 × 8 (Cl^-, 200–400 mesh). A thin α-source was prepared from uranium fraction by coprecipitation with NdF_3 [26]. Uranium was deposited on a 0.1 μm polypropylene filter (Pall Corporation, Gelman Laboratory). The chemical recoveries equaled from 65 to 85%. Samples were measured for a period of 2–3 days. The efficiency of the detector was 34% and MDA was equal to 0.5 mBq/l for ^{234,238}U isotopes for a measurement lasting for two days. The uncertainty of a single measurement was calculated as a square root of the sum of uncertainties in all quantities in quadrate.

The applied methods for the determination of radium and uranium isotopes were tested during proficiency tests organized by the international and Polish authorities. The reference materials delivered by the International Atomic Energy Agency (IAEA) were analyzed during the intercalibration experiments in 2002 (for ^{222}Rn and ^{226,228}Ra) [7, 17] and in 2008 (for ^{226}Ra and ^{234,238}U) [8]. The accuracy and precision of applied methods were judged as satisfactory.

The repeatability is constantly checked during analyses of natural and spiked water samples (with addition of tracers) prepared in the laboratory.

Results and discussion

The activity concentrations of ^{222}Rn, ^{226,228}Ra and ^{238,234}U in the investigated samples are presented in Table 2.

For ^{222}Rn and ^{226,228}Ra, the data constitute the mean values with the standard deviations resulting from all obtained results. The mean activity concentrations for ^{222}Rn in the investigated samples varied from 15±2 to 154±22 Bq/l and hence, in general, seem to be in good agreement with the archival results presented in Table 1. Moreover, these values are in agreement with the range of background for the Sudety Mts. groundwaters and can be ranked as low-radon and radon waters according to classification by Przylibski [23]. Some of these waters with radon activity concentration higher than 74 Bq/l were classified as medicinal. They are exploited and used for balneological purposes in the Przerzeczyn-Zdrój Spa. On the other hand, due to low radon content these waters do not pose a threat to the health of the spa population concerning radiological protection. The effective dose from ^{222}Rn calculated on the basis of the dose conversion factor 3.5×10^{-9} Sv/Bq [18] and 0.5 liter of water consumption per day was equal to 44.1 μSv/year for ^{222}Rn mean concentration equal to 69 Bq/l. If the radon content is lower than 100 Bq/l – a level recommended by WHO [35] as the threshold value of the activity concentrations of this gaseous radioactive isotope present in a water intake – then the waters are considered suitable for consumption or utilization in households.

The mean activity concentrations of ^{226}Ra isotope varied from below 10 to 30±1.5 mBq/l and of ^{228}Ra from below 30 to 60±4 mBq/l. In reference to other values obtained by the authors for the Sudety Mts. waters these concentrations are within the average results, close to the values noted up to now for waters with similar mineralization. The samples of the studied waters have the activity concentrations of ^{238}U from 4.5±0.6 to 13.6±1.2 mBq/l, and for ^{234}U from 17.1±0.9 to 52.2±2.8 mBq/l. All obtained uranium results were higher than MDA. Concerning radiological protection, all these isotopes are not hazardous to the population of the spa. Using the dose conversion factors for radium and uranium isotopes and 2 l/day of water consumption, none of the studied waters give the total annual effective dose exceeding the WHO limit of 100 μSv/year [35]. Activity concentrations of uranium, radium and potassium depend on the total dissolved solids (TDS) of groundwater. The higher the TDS value, the higher the activity concentration of the investigated radionuclides. Especially radium, ^{226}Ra isotope, is well correlated with TDS of waters, up to $r = 0.97$ [3, 13, 32]. The uranium concentrations published for groundwaters from the intakes in Świeradów-Czerniawa health resort situated in the Sudety Mts. present higher values of up to 133±10 mBq/l for ^{238}U and up to 390±29 mBq/l for ^{234}U [12]. All the activity concentrations results obtained for the Przerzeczyn spa do not differ much from the results obtained for drinking waters both in Poland and in Europe [2, 16, 20, 27, 31, 33].

The activity ratio ^{234}U/^{238}U in the investigated samples varied in the range from 3.1±0.3 to 4.7±0.4. ^{234}U as the decay product of ^{238}U is expected to be in secular equilibrium with the activity of ^{238}U isotope. However, due to some environmental circumstances the activity ratio ^{234}U/^{238}U equal to 1 may not be achieved in water. The isotopic ratio ^{234}U/^{238}U in water higher than 1 indicates preferential leaching of ^{234}U, the daughter of ^{238}U after two other decays of ^{234}Th and ^{234}Pa. The ^{234}U isotope may be more easily leached from a crystalline matrix by increased vulnerability to water related to the oxidation of uranium from the valence state IV+ to VI+. A full description of the state of disequilibrium in water and the processes responsible for it are discussed in details in [19, 28].

Table 3. Isotopic ratios: ^{234}U/^{238}U, ^{222}Rn/^{226}Ra, ^{226}Ra/^{238}U calculated for studied samples

Borehole	^{234}U/^{238}U	^{222}Rn/^{226}Ra	^{226}Ra/^{238}U
No. 2	4.6 ± 0.7	2301 ± 168	6.7 ± 0.9
No. 8	3.4 ± 0.4	7335 ± 1270	1.9 ± 0.3
No. 9	4.1 ± 0.4	–	–
No. 10	3.6 ± 0.4	–	–
No. 11	3.1 ± 0.3	–	–
No. 13	4.7 ± 0.4	2984 ± 182	3.2 ± 0.3
No. 15	3.4 ± 0.5	1580 ± 189	0.9 ± 0.1

Radon ^{222}Rn content in the studied water samples is from 1580 to 7335 times higher than the content of ^{226}Ra (Table 3). This fact confirms the high mobility of radon and its good solubility in groundwater. Moreover, it connects the origin of radon with the reservoir rocks which are the place of its buildup due to radioactive α-decay of ^{226}Ra. The dependences observed in groundwaters of the Przerzeczyn-Zdrój Spa are in good agreement with earlier results obtained by Przylibski [23].

The authors obtained interesting results for the activity ratios ^{226}Ra/^{238}U in groundwaters of the studied intakes. They ranged from 0.9 to 6.7 (Table 3). However, it should be emphasized that these results were obtained on the basis of few investigated samples. The values received may suggest that either the ^{226}Ra isotope is transported better in the studied waters than ^{238}U or (what is more likely) ^{226}Ra is situated on the surfaces of faults and cracks. Groundwater flows through them and consequently makes it easier for ^{226}Ra than for ^{238}U to be released since the latter is incorporated in the structures of minerals of reservoir rocks [14, 36].

The correlation coefficients calculated for individual isotopes are presented in Table 4. The high correlation coefficient between concentrations of ^{226}Ra and ^{228}Ra isotopes for waters from the three studied intakes, in which concentration results were greater than MDA, is equal to $r = 0.99$. From the above, it can be concluded that the chemical properties of radium, not the connection with the radioactive decay series (uranium-radium or thorium), are responsible for its content in groundwaters. Moreover, radium is washed out from cracks of reservoir rocks which it penetrates as a result of earlier nuclear decays, especially α-decay, in radioactive series. Then, it can be dissolved and adsorbed in the cracks several times depending on pH value and Eh potential conditions found on the border of groundwater/reservoir rock. Similar values of correlation coefficients

Table 4. Correlation coefficients and the number of correlated pairs

Correlated elements	Correlation coefficient	Number of correlated pairs
^{226}Ra-^{228}Ra	0.99	3
^{226}Ra-^{238}U	–0.83	4

were obtained earlier for different groundwaters from the Sudety Mts. [23].

The obtained dependence of ^{226}Ra activity concentration and the concentration of its parent isotope ^{238}U is also interesting. The obtained negative correlation ($r = -0.83$) may confirm the fact that radium is leached out from the surfaces of cracks and fissures of reservoir rocks and that it is not a product of radioactive decay of ^{238}U dissolved in groundwater. However, it is worth stressing that this relation has been obtained on the basis of only four pairs of results. Negative correlation observed between concentrations of ^{226}Ra and ^{238}U requires further investigations before a final interpretation can be postulated.

The obtained results suggest that ^{226}Ra isotope is located on the surfaces of faults and cracks of reservoir rocks where groundwater flows and from where it is released. ^{226}Ra is more easily leached than ^{238}U since the latter is incorporated in structures of minerals of reservoir rocks. A similar mechanism also causes gathering of ^{228}Ra on the surfaces of cracks and fissures. Thus, the activity concentrations of both isotopes of radium are well correlated in the studied waters, e.g. their concentrations change in a similar way.

Conclusions

The aim of the investigations was to evaluate activity concentrations of radioisotopes in seven groundwater intakes of the Przerzeczyn Spa. Waters from this region are poorly described as far as natural radioactivity is concerned. Moreover, the Sudety Mts. waters contain often higher concentrations of radon, radium and uranium isotopes, a fact that is connected with the presence of the latter two isotopes in the reservoir rocks. Analyses of natural radioactivity of the Przerzeczyn-Zdrój Spa groundwaters show considerable similarities to groundwaters of the Sudety Mts. In the studied waters radon ^{222}Rn seems to be of greatest importance. Moreover, considering other radioactive isotopes, ^{226,228}Ra and ^{234,238}U, groundwaters of the Przerzeczyn-Zdrój Spa do not differ much from waters with low mineralization from other regions of the Sudety Mts.

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