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ENVIRONMENTAL HAZARD IN THE AREA OF THE FORMER URANIUM ORE MINE "PODGÓRZE" IN KOWARY

The heap used in the past by an abandoned uranium mine "Podgórze" is situated closely to the Jedlica stream. The heap contains naturally occurring radioactive materials (NORM). The study was aimed at determining the environmental hazard posed by the NORM waste. Concentrations of hazardous elements and gamma radiation were determined. Trace elements contained in the waste material may be dangerous to waters, soil and plant cover in this area. It was found that due to a considerable concentration and migration properties, uranium poses the most serious hazard to the environment constitutes.

1. INTRODUCTION

Mining and processing industries are the leading producers of waste, collected in heaps and in sedimentation ponds. The waste materials are the source of hazardous heavy metals, metalloids and radioactive isotopes, which penetrate the environment. Polish uranium mining industry [1], concentrated in the Sudeten, has its own specific character. At first easily accessible beds were mined, but they soon were depleted. Therefore, new mines, i.e. in Kowary ("Podgórze") and in Radoniów, were built [1], [2].

The uranium ore bed in Kowary was discovered in 1950 and as early as in the fall of this year its exploitation began. It was located on the northern slope of the Karkonosze Mts., in the tectonic area where granites are in contact with the rocks adjacent to metamorphic rocks. The bed was formed as a consequence of tectonic fracture across gneiss and schist packages [3]–[6]. It was a vein of hydrothermal origin [4]–[6]. Uranium content amounted to 0.1–0.3% by weight [3], [7]. Concentration of uranium was classified as average [5], [6] or even low [3], [7]. Uranium minerals were accompanied by the variety of other heavy metal minerals, among others by hematite, pyrite, chalcopyrite, calcite and barite [5]. As early as in the fall of 1958, the bed was depleted, and the mine – closed. Excavations of the mine were spontaneously flooded [3].

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The operation of the “Podgórze” mine may be characterized as follows [3], [4], [7], [8]:

- Uranium mining period – from 1951 up to 1958.
- Amount of pure uranium in the ore mined – 196.5 t.
- Amount of material tipped – 228 500 m^3.
- Amount of materials in heaps – 88500 m^3.
- Length of horizontal excavations – 32200 m.
- Length of vertical excavations – 520 m.
- Mining operations were carried out to 660 m below the ground level.

In the first period of mining operations, there was tipped overburden, then also too lean ore, i.e. materials with uranium contents below 0.1% [1], [2], [9], [10]. Due to weather conditions uranium and radium were released, and together with other heavy metals penetrated the heaps, waters and the surrounding grounds [9], [11]. Erosion of heaps resulted in an increase of overall radioactivity of the Jedlica River water [3], and along its banks the concentration of uranium amounted to 4–7 ppm [12]. In the seventies of the twentieth century, an increased radioactivity was found in the Jedlica Valley, 20 km below the heap [13].

Mining operations generated the so-called NORM waste, characterized by an increased activity of naturally occurring radioactive materials and an increased dose-equivalent compared with a natural radiation background. The waste materials are hazardous to the natural environment, mainly due to contamination of underground [14]–[19] and surface [13], [16] waters with natural radioactive isotopes.

The Council Directive 96/29/Euratom of 13 May 1996 on basic safety standards for the protection of the health of workers and the general public against the dangers caused by ionizing radiation contains special clauses on radiation hazards posed by natural isotopes [20]. The UE Member Countries are obliged to adapt their legal regulations to the Directive [20]. In Poland, on November 29, 2000, the Sejm amended the Atom Law act to fulfil the requirements of the European legislation [21]. In the executory provisions of the act, i.e. in the government order on the management of radioactive waste and used nuclear fuel [22], the content of radioactive substances was adopted as the criterion for their classification as radioactive wastes. Particular categories of radioactive wastes are classified based on activity and concentration of radioisotopes. The legal regulations imposed on slightly radioactive rock materials, containing natural radionuclides, specify that if the sum of the maximum concentrations of the radioisotopes in diverse components of the waste material does not exceed 10 for a representative sample of 1 kg, then – according to the Appendix No. 1 to the government order [22] – the rock material is not classified as a radioactive waste. In the case of mining waste, containing increased concentration of natural radionuclides, the clause enables a complicated legal procedure provided by the Atom Law to be avoided [23]. On the other hand, waste materials with increased concentration of natural radionuclides are not taken into account in environmental protection regulations. So, the legal status of such waste materials is not formally given, in spite of the fact that due to their ionizing radiation they are dangerous to the environment.

2. STATE OF ROCK WASTE MATERIALS ON HEAPS

Currently, the area of the former mine is a typical abandoned industrial terrain, without any installations, by with heaps of gangue rock, small amounts of lean ore and fragments of concrete and brick structures. The heaps are scarcely covered with self-sown plants. In 1997, flood damaged a dewatering channel of heaps [17] which resulted in the returning of the Jedlica River to its old channel [8].

As a consequence of geological conditions, the uranium deposit was opened with drifts. In a close vicinity of those drifts, the dumping ground was situated. A current state of the rock waste dumping ground is as follows:

- **Drift No. 16** – close to the outlet there is the rock heap, approx. 3500 m^3 in volume, partially afforested, not endangered by washing away [3], [4].
- **Drift No. 17** – heap situated below the forest road contains approx. 15 000 m^3 of rock material. It is, to a considerable degree, afforested with greenwood. Water from the drift flows to the Jedlica River [3], [4].
- **Drift No. 18** – rock heap around the drift contains approx. 3200 m^3 of gangue [3].
- **Drifts Nos. 19 and 19a** – below the drift there are heaps with rock material, being within the period of 1958–1965 gradually utilized [3], [4], [8]. When approx. 140000 m^3 of the rock was recycled as building aggregates, the heaps lost their original shape [7]. The remaining material was partially arranged, by levelling of the surface and cutting terraces. Unfortunately, irregular slopes of the heap were not protected against washing out, and heavy rainfalls washed considerable amounts of the material into the river [4]. The rock material consists mainly of gneiss and mica schist, which may contain uranium [3].

After the mining operations were finished, the Drift No. 19a was used as a radon inhalation chamber of the nearby Cieplice health-resort [24].

In 2005, in the upper course of the Jedlica River, there were carried out fragmentary river bed regulation and bank stabilization works, aimed at protecting the river banks against erosion and at preventing further dispersion of radioactive rock material. The river in this place flows at the foot of the heap. In this section, both banks and bed of the river are not much damaged.

The abandoned heaps of overburden rock are situated in the mountain landscape, close to the Karkonosze National Park and the Rudawy Landscape Park within the "protected landscape area" of the Karkonosze–Izerskie Mountains. This forces us to undertake many actions aimed at preventing the environment degradation and reclaiming terrains. The landscape values in this area should be improved. Reclamation, as a complex action reducing a negative impact of various pollutants on the environment, is indispensable for minimizing the following environmental hazards associated with:

- effect of a direct radiation on people and animals,
- possible migration of heavy metals and radioactive elements to the Jedlica River,
- contamination of soil, surrounding the heaps, with heavy metals and radioactive elements,
- stability of heaps made of debris, rock and earth,
- assimilation and continuous accumulation of heavy metals in plants involved in the food-chain (in particular, concentration of mutagenic and carcinogenic substances in soil and in bottom deposits).

3. PURPOSE OF STUDY

The study was aimed at assessing environmental hazards posed by waste disposal sites of the abandoned uranium mine "Podgórze" in Kowary.

4. MATERIALS AND METHODS

4.1. MATERIALS

Material for analyzes was sampled from the heaps of waste located below the Drafts No. 19 and No. 19a of the "Podgórze" mine, in the areas of plant vegetation. The samples were taken at a height of 1 m above the terrain surface, from the areas where gamma radiation equivalent dose amounted to 500–2400 nSv/h. The measuring points and sampling sites are shown in the figure.

4.2. METHODS OF ANALYSIS OF SOIL MATERIALS

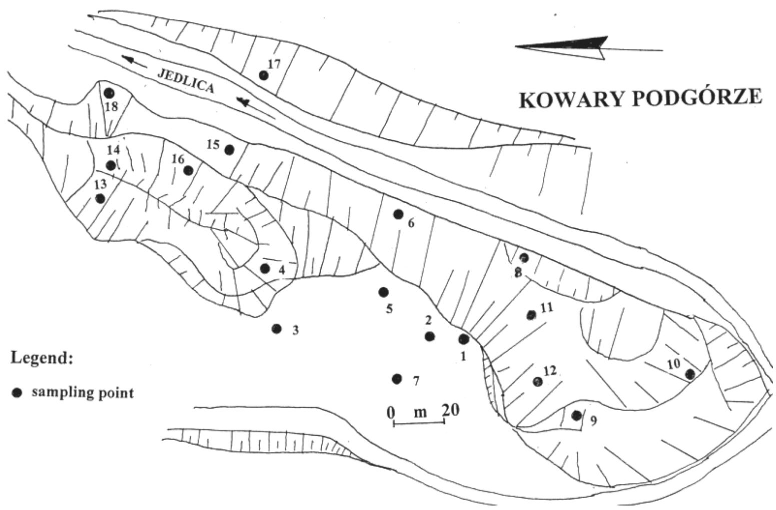
According to the standards in force, the samples of soil were analyzed for:

- basic physical and chemical properties (grain composition, moisture content, pH of water extracts),
- content of heavy metals (Cd, Cu, Ni, Pb, Zn, As, Cr, Co and Ba) and toxic elements.

Grain composition was determined by means of a sieve analysis, according to the PN-R-04033 standard requirements.

According to the method recommended by STRZYSZCZ [25], the reaction of the waste material was determined potentiometrically, by measuring pH of raw samples in their original graining.

Chemical composition of the samples was determined with the XRF method, using the sample preparation equipment and the PW 2400 type fluorescent X-ray spectrometer of Philips.



Localization of sampling points on heaps of "Podgórze" mine

The gamma radiation equivalent doses were measured by means of a portable apparatus Thermo Eberline ESM, the FH 40 GL 10 type, equipped with an ionization chamber. The apparatus was calibrated and certified by German Bundesamt für Strahlenschutz.

Water extracts were prepared according to requirements set by the government order (DzU Nr 162, item 1116).

5. DISCUSSION OF RESULTS

Petrographical composition of the samples was diverse. The samples varied in their grain composition, weathering and proportion of particular components. Generally, rock grains are irregular in shape and sharp-edged.

Sieve analysis of the samples collected in six various points of the heap proved that the material consists of:

- 0.05–2 mm sand, 25–30% by weight,
- 2–75 mm gravel, 50–65% by weight,
- > 75 mm cobbles, 5–10% by weight.

In the post-industrial areas, the succession of plants proceeds slowly and is a prerequisite for the reconstruction of natural environment. Plants, colonizing the heap are severely controlled by unfavourable conditions. The soil-forming processes consist in proceeding weathering of rock material, and succeeding plants promote formation of regosol. In all the samples examined, the proportion of biotite, the important component of soil, reached approx. 3%. Nevertheless, on post-industrial terrains, the rate of soil formation depends mainly upon anthropogenic factors [26]. The properties of the soil formed, such as water capacity, absorbing capacity and buffering properties, depend upon granulometric composition of the material [27], [28].

Bulk density of the ground skeleton varied within the range of 1350–1580 kg/m^3. Mineralogical analysis proved that the material was composed of dust, sand, crushed rock grains (gneiss, schist, amphiboles and pyroxene) and organic matter.

pH of the waste materials sampled from the heaps varied from 6.0 up to 7.8. The reaction is associated with the presence of carbonates, silicates and basic ions. Ferric sulphides, mainly pyrite and marcasite, occurring in small amounts [10], are responsible for soil acidification. The released cations may bond H^+ ions, or may be adsorbed by the clay minerals formed. They may also be washed out of the material. Final products of the decomposition of clay minerals are ionic forms of aluminum and iron. Chemical composition of anthropogenic soils of an abandoned "Podgórze" mine is given in the table.

Table

Chemical composition of anthropogenic grounds of abandoned "Podgórze" mine

Sample No.	Rock material sampled from the heaps										
	(mg/kg d.m.)										Dose equivalent
	Pb	As	Ba	Cr	Cu	Ni	Zn	Cd	Co	U	(nSv/h)
1	52	197	318	36	41	39	14	9	16	402	1200
2	119	48	161	21	39	22	76	8	15	38	2400
3	236	31	120	33	52	31	20	3	18	41	670
4	119	50	142	37	23	23	76	8	15	59	630
5	118	37	183	61	32	20	83	7	11	31	650
6	65	38	165	24	31	40	30	5	19	50	650
7	80	87	173	108	36	39	80	6	14	113	1000
8	41	47	165	34	29	37	59	6	12	62	780
9	683	85	247	25	22	24	93	7	10	33	500
10	213	52	163	46	21	24	103	7	9	28	550
11	108	45	200	63	17	23	72	7	9	24	530
12	153	46	250	37	22	20	72	7	9	31	550
13	41	50	144	30	33	32	44	5	10	42	500
14	736	199	1263	43	38	50	310	8	36	115	1200
15	66	47	154	28	30	34	39	2	15	55	600
16	74	55	222	50	38	36	41	8	14	152	1080
17	51	89	129	38	26	41	32	3	17	45	650
18	70	48	138	47	29	30	49	6	11	32	530

Concentrations of some of the elements, e.g. As, Cd, Co, Cu, Ni, Pb, found in the waste material of heaps, considerably differ from the Polish averages. In particular, concentrations of As, Pb and Cd, i.e. the metals extremely dangerous for living organisms, are higher. So, the results of chemical analyses of gangue accompanying the uranium beds confirmed the reports found in [10], [29]–[32].

In almost all the samples of anthropogenic soils, the concentrations of As, Cd, Co, Cu, Ni and Pb exceeded the permissible values for Polish arable grounds [33]. Moreover, in some of the samples, the concentrations of such trace elements as Ba, Cr, Zn were too high.

A separate issue is radiological hazard for the environment caused by the NORM waste collected in heaps of the former "Podgórze" mine. In some areas of the Sudeten, the concentration of uranium reached 13.3 mg/kg of dry material [33]. However, in the samples examined, this level was exceeded ten or even twenty times. In one case, it was even 30 times higher.

Taking into account that most of uranium minerals are vulnerable to weathering and that some microorganisms are able to decompose uranium compounds, e.g. in granite rock [34], [35], it can be inferred that uranium can travel through soil and hence is considered to be the element most dangerous for the environment. In the same way, the environment is contaminated with other members of the uranium series. Due to its high-power radiation, radium should be considered as extremely dangerous element of the series.

Assuming the permissible maximum concentrations of trace metals (Pb, As, Ba, Ni, Cd, Co) in soil as the criterion of hazard, as specified in the order of the Ministry of the Environment [36], it should be concluded that any of the samples collected from the heaps does not comply with the requirements set for arable land of the B class. Moreover, approx. 30% of the material sampled exceed the permissible maximum concentration of at least one of the following metals: As, Pb and Ba, assumed to be the standards for the quality of industrial area, mineral excavation and roads.

The data presented in the table prove that the material collected in the heaps constitutes a potential environmental hazard due to the content of trace elements and natural nuclides belonging to the uranium–radium series. Under the influence of atmospheric conditions, mainly due to the water and oxygen access, uranium and the products of its degradation will migrate towards the Jedlica River.

The basic issues arising from the study are as follows:

- Radiation hazard caused by the NORM-type waste was determined by direct measurement of equivalent dose of gamma radiation. The results of the measurements proved that radiation many times exceeds the background level, which calls for remedial actions.

- Very diversified contents of cadmium and lead, the elements characterized by extremely high accumulation rate, which, according to BOWEN [37], belong to the first group of the elements dangerous for the environment, unnecessary for plant growth [37]. Concentration of cadmium varies between 3 and 9 mg Cd/kg of dry material, and that of lead – from 41 to 736 mg Pb/kg, whereas the ranges detrimental for the plants [38] are 5–10 mg Cd/kg and 50–100 mg Pb/kg.

• High concentration of barium – from 120 to 318 mg/kg, and in one sample even 1263 mg/kg – may be explained by the presence of barite, feldspar and biotite in the ore.

• Moreover, it is noteworthy that uranium occurs in waste materials in a relatively high concentration.

6. CONCLUSIONS

1. Due to the presence of trace minerals in gangue, the heap poses an environmental hazard. The rock material may be the source of radioactive isotopes of the uranium–radium series, including the alpha radioactive one, posing the main risk. The isotopes migrate in the environment.

2. The results of the analyses of anthropogenic soils confirmed the data known from the references [10], [29], [30] that the gangue accompanying the uranium bed contains considerable amounts of trace elements. Concentrations of the trace elements are as follows (mg/kg of dry material):

(a) micronutrients

$$14 < Zn < 310; \quad 9 < Co < 36; \quad 17 < Cu < 52;$$

(b) elements detrimental for plats, people and animals

$$20 < Ni < 50; \; 31 < As < 199; \; 21 < Cr < 108; \; 41 < Pb < 736; \; 3 < Cd < 9; \; 24 < U < 402.$$

3. Heavy metals and radionuclides present in the environment not only create hazard of their bioaccumulation in organisms being the members of food-chains, but they also are responsible for an increase of gamma radiation over the background level.

4. Grain composition of the waste material is diversified; gravel and sand prevail.

5. The waste material and plants growing on the heaps are the main biological soil-forming factors.

6. Reclamation of the waste disposal sites is recommended to reduce their negative environmental impact that manifest itself as:

• the possibility for long-distance migration of both trace elements and radionuclides of the uranium–radium series,

• hazard caused by gamma radiation.

Reclamation should allow us to:

• provide efficient safety means, particularly in the close vicinity of Drafts No. 19 and No. 19a, subsidences and the shaft of the mine,

• reduce slumping and erosion of slopes,

- improve landscape and surface features of the terrain,
- reconstruct access roads.

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ZAGROŻENIA ŚRODOWISKA NA OBSZARZE BYŁEJ KOPALNI RUD URANOWYCH „PODGÓRZE" W KOWARACH

Zwałowisko nieczynnej kopalni rud uranu „Podgórze" gromadzące tzw. odpady NORM zostało zlokalizowane nad potokiem Jedlica. Celem badań było sprawdzenie, jak niebezpieczne dla środowiska mogą być składowane odpady. Zagrożenie oceniono, określając w próbkach materiału odpadowego zawartości uciążliwych dla środowiska pierwiastków oraz dokonując pomiaru mocy dawki promieniowania gamma. Zawarte w składowanych odpadach pierwiastki śladowe mogą stanowić zagrożenie dla wód, gleby i szaty roślinnej w rejonie składowania. Wykazano, że najpoważniejszym zagrożeniem dla środowiska naturalnego ze względu na stężenie i możliwość migracji jest uran zawarty w odpadach hałdy kamiennej.