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ENVIRONMENTAL ASPECTS OF POST MINING URANIUM WASTES DEPOSITED IN RADONIOW, POLAND

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Abstract: Post mining uranium wastes from Radoniów were analysed and chemical, mineralogical andpetrographic characteristics were performed. Additionally, the size parameters of dumps and direction of water runoff were estimated using LIDAR data. Bioaccumulation of uranium in different plants covering dump's surface was shown.

Keywords: *uranium, post mining wastes, bioaccumulation, LIDAR*

Introduction

The Radoniów deposit is located in the northern part of the Izera Metamorphic Complex (Poland). This unit is composed of granitic gneisses, quartz-biotiteschists, mylonites and breccias with trace amounts of sulphides. Primary and secondary uranium minerals have been found. The primary minerals are pitchblende, uraninite with blue fluorite, and hematite. Between secondary minerals, metatorbernite, metauranocircite, metaautunite and uranophane were recognized (Piestrzynski et al., 2006). Uranium mining has started in 1954 and was continued until 1960.

Post-mining wastes were deposited in the dump located near the mine. Originally the dump covered an area of about 25 000 m^2 and γ-measurements made in 1971 showed elevated radiation (about 500 μR/h (~5 μSv/h)), reaching a maximum dose rate of 2000 μR/h (about 20 μSv/h) (Piestrzynski et al., 1996). During the next twenty years the dump was reexploited for local purposes (mainly for construction of local

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roads and sometimes for buildings). In 1994 the average noted dose rate was about 80μR/h with maximum 105 μR/h (about 0.8–1.05 μSv/h).

All data collected by Piestrzynski et al. (1996) show that the rocks deposited on the dump reveal a significant amount of uranium. The available data are not sufficient to present quantitative figures of uranium content in the dump and its dispersion around the dump.

In presented research we try to quantify material collected in two dumps (actually, after reexploitation two dumps exist – one small and the second much larger) and describe mineral and chemical character of the deposited wastes.

Materials and methods

Sampling

Wastes were collected from two dumps located in Radoniow (Large Dump –N 51°00’13.39;E 15°29’00.16, Small Dump - N 51°00’15.55; E 15°28’53.68). Plants were collected from the surface of dumps.

XRD Analyses

A Philips X’Pert PW 3020 X-ray diffraction analyzer was utilized to characterize the mineral composition of wastes. Co radiation (K-Alpha1 [Å]-1.78901 K-Alpha2 [Å]-1.79290 K-Beta [Å]-1.62083;K-A2 / K-A1 Ratio-0.50000) was used at 30 mA, and 40 kV.

Petrography

Light petrographic microscopy of thin section of polished waste sample was used for petrographic characteristic.

Chemical analysis

Uranium in wastes was analyzed using the ICP-MS technique according to the procedure described by Chajduk et al. (2013). Extraction analysis with EDTA, phosphate buffer and 0.11 M acetic acid (pH 3.0) was done for determination of uranium bioavailability. Uranium and other metals in plants were analyzed in Polish Geological Institute-National Research Institute using the ICP-MS technique according to the certified procedure used in Central Chemical Laboratory (Polish Centre of Accreditation certificate No. AB 283).

Electron Microscopy

Scanning Electron Microscopy analysis was done in the Paleozoology Department of Museum and Institute of Zoology, Polish Academy of Sciences. HITACHI S-3400N Scanning Electron Microscope equipped with an EDS X-ray microanalyser, (Superdry II detector) was used for examination of polished section of wastes and crushed wastes (fraction 250–500 μm). Acceleration voltage was 20.0 kV. Thin sections of polished waste sample were examined.

Analysis of the dump size and direction of water runoff

The pictures taken in 2011 in the LAS format and containing data from laser scanning ground – LIDAR with spatial resolution of 12 points per square meter were used for analysis. Basing on the point of altitude (altitude above sea level), triangulation model TIN (Triangulated Irregular Network) was created. It was then converted into a GRID model based on a grid. Resolution of this model was 0.5 m. Analysis was carried out in the Space Research Centre of Polish Academy of Sciences.

Results and discussion

Waste dumps in Radoniow are localized in the vicinity of Radoniow village and they are surrounded by agricultural lands (Fig. 1). The surface of dumps is is tightly folded (especially the surface of the large dump) and clear signs of erosion are visible (Fig. 2).

Fig. 1. Post mining waste in the dumps in Radoniow (www.geoportal.gov.pl)

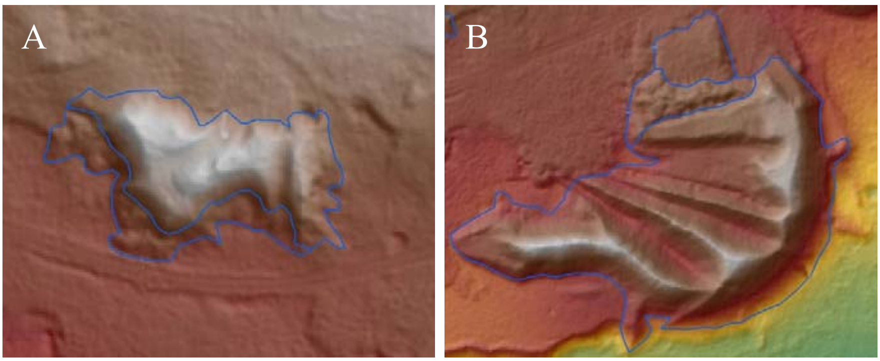

Fig. 2. Shape of small dump (A) and large dump (B)

Geometrical parameters of dumps are presented in Table 1. Total surface occupied by dumps in 2011 (7368 m^2) decreased significantly in comparison to 1971 when it was about 25 000 m^2. The total volume of dumps in 1971 is unknown and now is 44 803 m^3. Uranium concentration in waste is still high. An average uranium concentration is about 153 mg/kg with minimum about 60 mg/kg and maximum 806 mg/kg. The richer uranium wastes were found in the small dump. Total amount of uranium deposited in the dumps basing on our studies is about 17,97 Mg (volume × density (~2.65 kg/m^3)). A large amount of aggregate showing uranium concentration between 358 mg/kg and about 2000 mg/kg (single stones) was observed around the dumps in 2012. The dose rate noted on the surface of the dump in 2012 was 0.86 – 4.4 μSv/h. The minimum was similar to the radiation level noted by Piestrzynski et al. (1996) in 1994 but the maximum dose rate is only slightly lower than this one noted in 1971. Single stones around the dumps show very high radiation with the dose rate reaching 20 μSv/h.

Table 1. Dumps size parameter calculated using LIDAR data

Model LAS 0.5 m	Total	Small dump	Large dump
surface projection [m^2]	7368	1176	6192
body surface [m^2]	8621	1350	7271
altitude minimum [m]	402.3	407.1	402.3
altitude maximum [m]	414.6	414.6	414.1
height difference [m]	12.3	7.5	11.8
volume [m^3]	44803	4573	40230

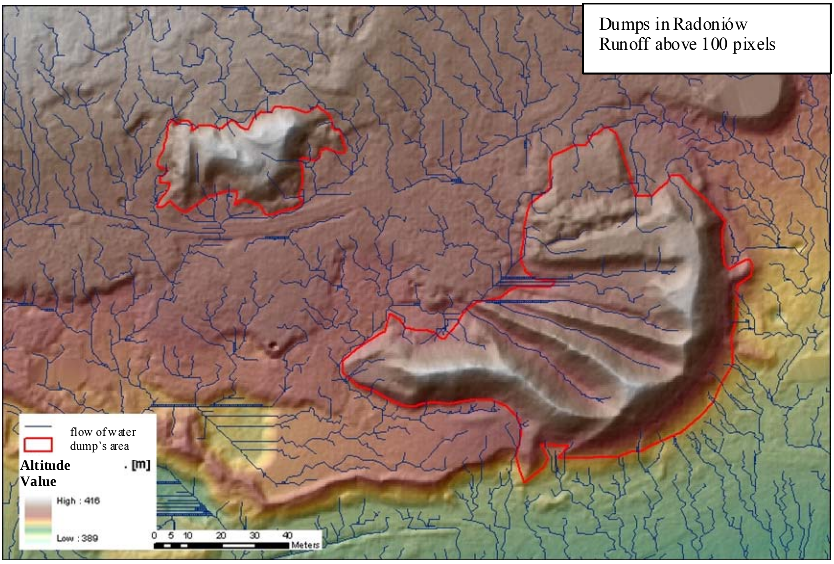

Fig. 3. The intensity of surface water runoff calculated basing on LIDAR data

The main direction of water runoff in the large dump is West, except northern part of dump where outflow to the East and North is observed. In the small dump water runoff to the South prevails. Petrographic studies of the waste materials revealed gneiss with laminar and eye texture as the main building material. Gneiss contains following minerals: quartz, K-feldspar, plagioclase and mica (mainly muscovite). In some parts of material gneiss is transformed to mylonite with thin slots filled by fluorite and iron oxyhydroxides. Ore minerals are also present between which pyrite is most abundant (Fig. 3). Pyrite occurs in gneiss and in mylonite. Uranium mineral were not found in petrographic studies.

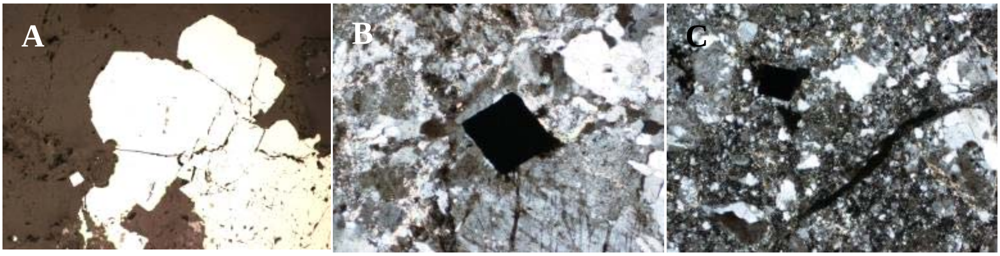

Fig. 4. Pyrite occurrence: single crystals and pyrite aggregates (A); in gneiss (B) and mylonite(C).

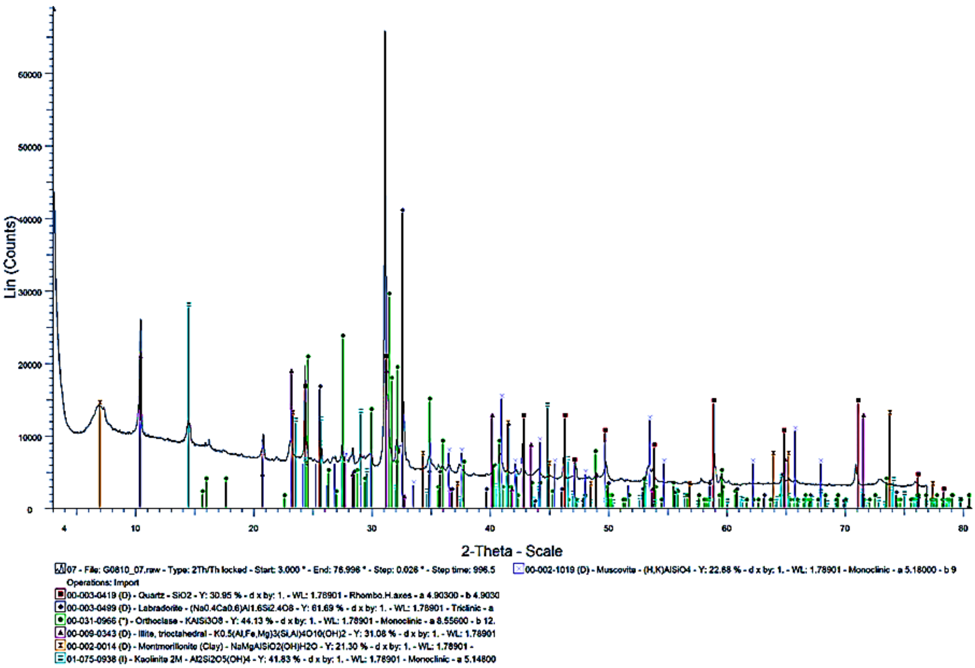

Fig. 5.XRD analysis of waste material taken from the small dump

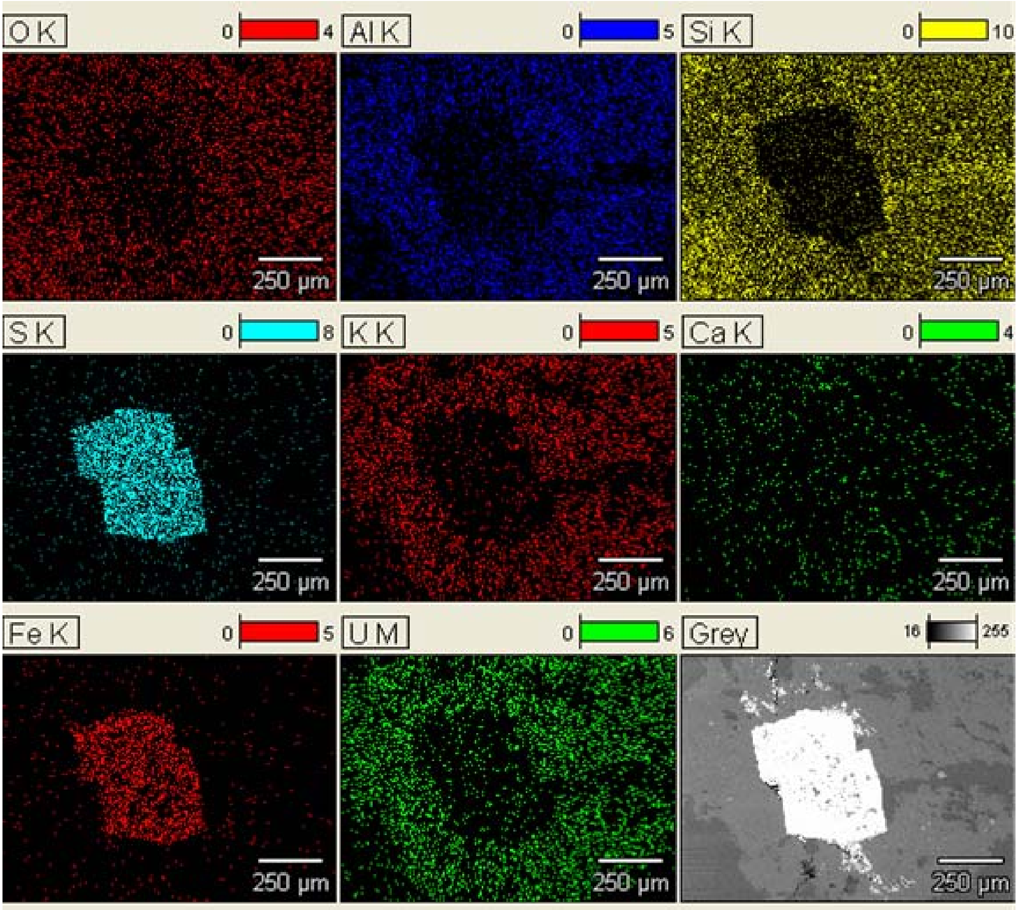

Fig. 6. Results of EDS-X ray analysis made in SEM

In XRD analysis (Fig. 5) quartz, orthoclase, montmorillonite, kaolinite, illite and labradorite were found. XRD profiles of samples from the dumps were very similar and uranium minerals were not found. Uranium presence in material from the dumps has been only shown in scanning electron microscope with the use of EDS-X ray microanalyzer. Its occurrence is strongly correlated with potassium and aluminum and uranium minerals often surrounding pyrite aggregates (Fig. 6).

Lack of detection of uranium in the XRD and petrographic analysis is a proof of its wide dispersion in waste material deposited in the Radoniow dumps. However, uranium concentration as well the level of radiation is relatively high, which brings serious environmental issues. The presence of pyrite and relatively low pH (below 6.0) facilitates leaching and bioleaching processes. We described earlier (Rewerski et al., 2013) that indigenous microorganisms living in wastes are able to oxidize iron what can cause uranium leach up to 30% of its total content in nearly neutral pH and 90%

under acidic conditions. The extraction analysis shows that 21–25% of uranium is associated with the carbonate fraction, 5.8–10.6% occurs in soluble fraction (respectively in the large and small dump) and 0.1–2.7% is bioavailable (respectively in the small and large dump).

Table 2. Uranium concentration found in plants covering the surface of dumps.

Dump	Plant		mg U/kg dry weight (ppm)
Radoniow	berd vetch *(Vicia cracca)*	stalk	0.57
		leaves	0.73
		root	9.96
	ribwort (*Plantago lanceolata*)	flower	2.69
		stalk	2.76
		leaves	14.64
		root	56.40
	clover (*Trifolium hybridum*)	flower	0.80
		stalk	2.24
		leaves	2.83
		root	71.54
	alfalfa (*Medicago lupulina*)	flower	4.10
		stalk	2.25
		leaves	5.37
		root	0.55
	white sweet clover (*Melilotus albus*)	stalk	2.02
		leaves	3.76
		root	10.24
	orchard grass (*Dactylis glomerata*)	flower	3.65
		stalk	0.25
		leaves	3.22
		root	77.54

Therefore we decided to check accumulation of uranium in plant tissue. Plant's samples were collected in the runoff because only there was enough vegetation developed. Uranium is accumulated mainly in the roots reaching the maximum concentration at 77.54 mg/kg (dry weight) in the roots of orchard grass. Leaves accumulate much lower uranium amount and maximum of bioaccumulation was noted in ribwort's leaves (14.6 mg/kg). Surprisingly a relatively high uranium level was noted in flowers of alfalfa and orchard grass.

Distribution of uranium in the individual extraction fractions, its accumulation in plants and earlier information concerning its bioleaching by indigenous microorganisms (Rewerski et al. 2013) clearly show that uranium deposited in dumps may be disseminated in the environment causing contamination of soil, surface water and groundwater.

Conclusions

Post mining wastes deposited in Radoniow contain a significant amount of uranium. The concentration of uranium and level of radiation in some parts of dumps is still hazardous for the environment and availability of dumps should be limited. The indigenous plants accumulating uranium in leaves, e.g. ribwort, may be useful for bioremediation.

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