ASSESSMENT OF THE RADIOLOGICAL SITUATION IN DIFFERENT AREAS AFFECTED BY URANIUM MINING AND URANIUM PROCESSING IN THE CZECH REPUBLIC

by

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This study presents measurements of activity concentrations of natural radionuclides in selected water and soil samples taken from areas affected by the uranium industry in the surroundings of Pribram, Straž pod Ralskem and Mydlovary, Czech Republic. In these areas, the dose equivalent rate was also determined at the sampling locations and additionally also during walkabouts in the surroundings of sludge fields. The activity concentration of water samples was 0.06 0.02 BqL^{-1} for ^{226}Ra and 0.07 0.07 mgL^{-1} for ^{238}U while the mean activity concentrations of soil samples were 74 70 Bqkg^{-1} and 80 77 Bqkg^{-1} for ^{226}Ra and ^{238}U, respectively. The average value of the dose equivalent rate was of 0.15 0.1 μSvh^{-1}. These values conform with the nature of the industrial activities which were carried out in the areas and are comparable with measurement results in similar locations worldwide.

Key words: environmental radioactivity, activity concentration, dose equivalent rate, uranium industry

INTRODUCTION

Uranium mining and uranium processing are activities requiring long-term monitoring due to the possible consequences for human health and environmental integrity, which may be affected by radiation exposure while these activities are undertaken, but also after they have come to an end. In the Czech Republic, the uranium industry has been and is still overseen by the state enterprise DIAMO, which was formerly responsible for uranium mining and processing, and now mainly for remediation and recultivation work after the last Czech uranium mine closed in April 2017 [1, 2]. The beginning of uranium mining in what is now the Czech Republic goes back to the 16th century, when the miners of silver encountered a black, shining mineral somewhat similar to silver ores which, however, contained no silver. It became later known as pitchblende and was found to contain uranium. The first uses of this mineral were associated with the colouring of glass or ceramics [3]. In the framework of a geological survey, uranium deposits were later found in different places in the former Czechoslovakia, characterized by very diverse mineral forms and concentrations, in different geological formations. Between 1945 and the mid-1990 s, uranium mining was an important industry in Czechoslovakia and as far as the production of uranium concentrate is concerned, the country held a top position in the world [4]. Mining areas existed around Jachymov, Horní Slavkov, Príbram, and Rožna – Olši [1]. The ore obtained was processed in pre-processing plants (Príbram, Jachymov) and processing plants (MAPE Mydlovary, Dolni Rožinka and Straž pod Ralskem) [5]. Whereas pre-processing consisted of mechanically breaking ore into smaller pieces and eliminating those with a lower radioactivity, processing consisted of extracting uranium with the help of acids or leaches (depending on the ore type) and producing so-called yellow cake, mostly containing triuranium octoxide (U_3O_8) [6, 7]. The assessment of the environmental consequences of these activities, and the possible health effects on people participating in the production as well as the population in the surroundings, is a very complex process, as is decision making about possible remediation in the areas affected by uranium mining and processing [8]. For the evaluation of the burden to the environment due to the contamination close to these sites, it is necessary to take into consideration the specifics of the given locations. The risk to human health arises from the inhalation or ingestion of short-lived radon decay products and long-lived radionuclides of the uranium-radium series emitting alpha-particle radiation, mostly in radioactive dust, and from external gamma-ray irradiation [9].

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Radium present in sludge deposits decays producing radon, which is then released into the atmosphere. In low-precipitation periods, sludge deposits can (partly) dry up and contaminated dust can be whirled up into the atmosphere [10]. Moreover, gamma-emitting radionuclides present in sludge may considerably affect the background dose rate in the area. Furthermore, the sludge contains toxic elements such as arsenic, molybdenum, astatine, iron, vanadium, lead and other heavy metals. These elements can enter the soil through sludge leaking and affect the ground as well as surface waters [11].

In our research, we assessed the radiological situation in three areas – Pribram, Straž pod Ralskem and Mydlovary – where chemical processing plants and sludge fields are found at different stages of operation. Some of these facilities are still (partially) operating, while others are at the stages of decontamination and remediation. In all three areas, numerous soil and water samples were taken and later analysed in the laboratory, dose equivalent rates were measured at the same locations. Our main purpose here is to compare the three main areas of uranium mining and processing in the Czech Republic, correlate the results – if possible – with the activities carried out at the different localities, and draw on literature data for further analysis.

METHODS

Study area

Three localities were chosen for this research, in which very diverse technologies of uranium mining and processing were used. These are the areas of Pribram, Straž pod Ralskem and Mydlovary, fig. 1. Maps of the individual locations with the exact sites of sampling are shown in figs. 2-4.

When uranium mining was begun in the Pribram area in the 1950 s, the building of a uranium ore treatment plant nearby was considered convenient, but later it was decided that the ore would be processed in MAPE Mydlovary. The site of the former processing plant represents one of the most significant environmental problems in the Pribram region, mainly because of high gamma exposure rates, radon emissions, and the stirring up of dust with an increased radionuclide contents. Different by-products of mining activities have been and still are deposited here. Section I still serves, among other purposes, as a receptacle for mining water pumped from one of the shafts which is kept open. The operation of Section II as a tailing pond was terminated in 1988, and technical and biological reclamation was carried out in the following years [12].

Figure 2. Situation map – Pribram

Figure 3. Situation map – Straž pod Ralskem

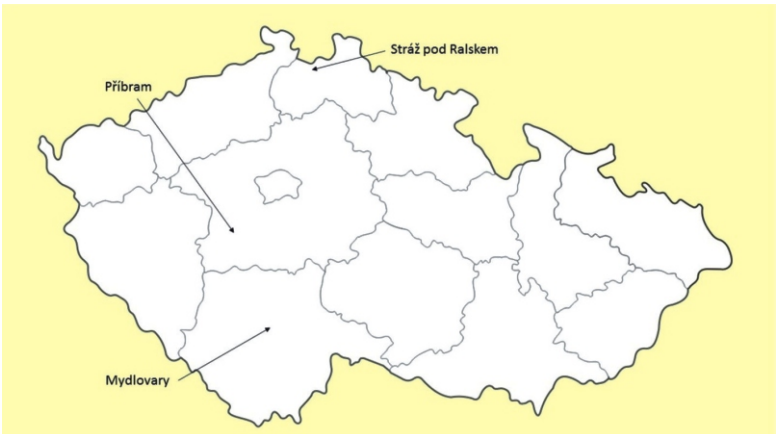

Figure 1. Three location of research: Pribram, Straž pod Ralskem, and Mydlovary

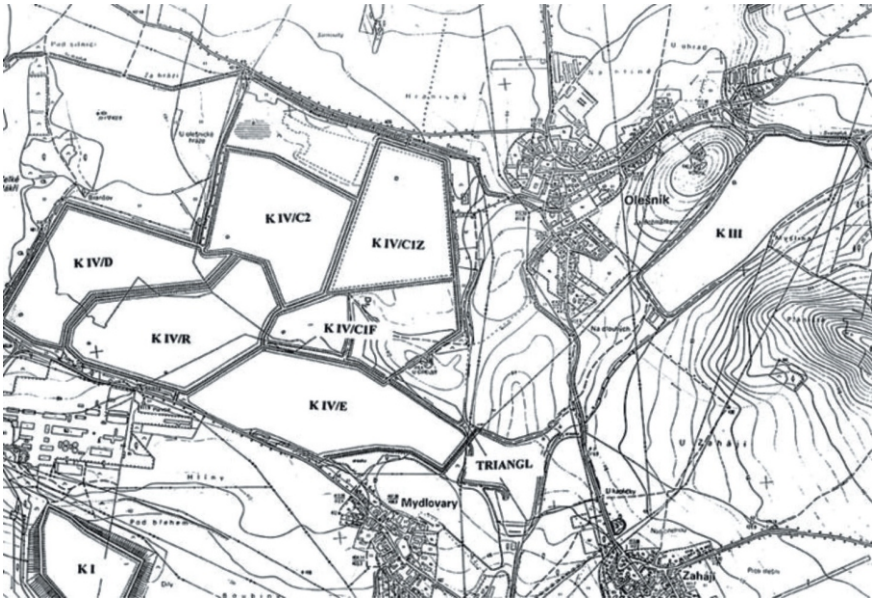

Figure 4. Situation map – Mydlovary

The deposits at Straž pod Ralskem were exploited by both conventional and in situ leach methods. The latter in particular led to the accumulation of radioactive sludge in large tailing ponds which have been only partly remediated. Deposit I receives all kinds of contaminated materials such as ferrous and non-ferrous metal, used protective equipment, contaminated wood, piping plastics, building debris, and sediments from a creek contaminated by radionuclides. Deposit II serves exclusively as a repository for neutralized sludges [13].

In the case of the MAPE Mydlovary uranium ore treatment plant, which in the late 1960 s to early 1990 s produced most of the yellow cake in Czechoslovakia, exploited lignite mining sites were used for waste disposal. The tailing ponds here cover an area of approximately 285 hectares, where 36 million tons of sludge with 2320 tons of uranium (and 10^{14} Bq ^{226}Ra) are stored. Since the processing was terminated in 1991, extensive sanitation and reclamation work has been carried out in the area. Tailing ponds I, III, IV/D and E have been completely covered, whereas the ponds IV/C2, C1Z, C1F and R remain partly open. This area has continued to be used for the storage of mining by-products. In the long term, the open areas will serve to accumulate and purify sludge waters, and to store by-products of pond maintenance and disposal operations [14].

Sampling and sample preparation

A total of 56 samples was collected (30 soil samples, 12 surface water samples and 14 water samples from hydrologic monitoring boreholes, *i. e.*, ground water samples) from the selected locations. The samples were taken from September to December 2016, GPS coordinates of the sampling points are given in tabs. 1-3. The sites for taking surface water samples were chosen considering the likelihood of contamination, but certain restrictions in admission to the potential sampling sites also had to be respected.

Soil samples were placed into polyethylene bags. A sample of 20 cm 20 cm 10 cm was first taken with the help of a shovel, from a 10 cm ground depth. From this primary sample, the top 5 cm litter layer was separated and discarded. Each soil sample thus had a total volume of 2000 cm^3, mass ca. 5.5 kg. The samples were dried at 105 °C until no further

Table 1. Samplings sites in the locality Pribram

Sample number	GPS co-ordinates	Description of the locality	Material
1	49°41'21.6"N 14°03'44.6"E	Wastepond I – Bytiz, centre of the dam	Soil
2	49°41'27.9"N 14°03'38.5"E	Wastepond I – Bytiz, north-east	Soil
3	49°41'26.1"N 14°03'25.1"E	Wastepond I – Bytiz, north	Soil
4	49°41'23.1"N 14°03'10.2"E	Wastepond I – Bytiz, north-west	Soil
5	49°41'14.4"N 14°03'05.7"E	Wastepond I – Bytiz, west	Soil
6	49°41'06.2"N 14°03'14.2"E	Wastepond I – Bytiz, south-west	Soil
7	49°41'07.1"N 14°03'26.5"E	Wastepond I – Bytiz, south	Soil
8	49°41'10.4"N 14°03'44.6"E	Wastepond I – Bytíz, south-east	Soil
9	49°41'11.7"N 14°03'47.1"E	Bytízsky brook – above Pribram I	Water
10	49°41'21.3"N 14°03'54.6"E	Bytízsky brook – bypass channel	Water
11	49°41'19.7"N 14°03'48.9"E	Pribram I – inflow	Water
12	49°41'19.7"N 14°03'48.9"E	Pribram I – outflow	Water

Table 2. Sampling sites in the locality Straž pod Ralskem

Sample number	GPS co-ordinates	Description of the locality	Material
1	50°42'25.6"N 14°45'49.8"E	Wastepond, south, 1st sample	Soil
2	50°42'33.1"N 14°45'34.1"E	Wastepond, south-west	Soil
3	50°42'44.5"N 14°45'39.1"E	Wastepond, west	Soil
4	50°42'50.8"N 14°45'52.8"E	Wastepond, north-west	Soil
5	50°42'55.3"N 14°46'26.0"E	Wastepond, north, 1st sample	Soil
6	50°42'54.5"N 14°46'38.9"E	Wastepond, north, 2nd sample	Soil
7	50°42'51.1"N 14°47'09.4"E	Wastepond, north-east	Soil
8	50°42'39.7"N 14°47'16.4"E	Wastepond, east	Soil
9	50°42'29.6"N 14°47'09.1"E	Wastepond, south-east	Soil
10	50°42'16.4"N 14°46'51.8"E	Wastepond, south, 2nd sample	Soil
11	50°42'18.6"N 14°47'12.6"E	Outlet of the Sedlicky fishpond	Water
12	50°42'19.9"N 14°47'16.5"E	Sedlicky fishpond	Water
13	50°42'23.4"N 14°47'05.6"E	Outer drainage, north	Water
14	50°42'18.2"N 14°47'02.0"E	Outer drainage, south	Water

weight change was observed. Larger solid pieces (for example stones) and plant and animal segments were removed from the sample. The sample was then processed through a fine-grain sieve. The mass of the processed sample was determined and the radioactivity analysis was carried out as described below [15].

The surface water samples were taken with the help of a sampling device and placed into two-litre PET bottles. In this procedure, attention was given to avoiding undesirable materials which could enter the sample from the surface (for example leaves, grass). Waters were sampled under relatively stable meteorological conditions – clear to partly cloudy with zero atmospheric precipitations and temperatures about 18 °C. Immediately after handing over to the laboratory, samples were stabilised and preserved with hydrochloric acid. [15].

Water samples were also taken from hydrologic monitoring boreholes (so called indication holes) existing in the network of the DIAMO state enterprise. The quality of ground water from these holes is routinely monitored in the vicinity of sludge fields in the Mydlovary area. In the case of ground waters from hydrologic monitoring boreholes, sampling involved pumping to the surface (after triple exchange of water in the hole, if this was possible, depending on the whole capacity). Because of the tedious nature of this

Table 3. Samplins sites in the locality Mydlovary

Sample number	GPS co-ordinates	Description of the locality	Material
1	49°05'20.9"N 14°20'19.0"E	Wastepond KI, east	Soil
2	49°05'25.7"N 14°19'50.4"E	Wastepond KI, weast	Soil
3	49°05'42.0"N 14°20'57.0"E	Wastepond KIV/E, south	Soil
4	49°05'56.6"N 14°20'01.6"E	Wastepond KIV/R, south-west	Soil
5	49°06'12.0"N 14°19'51.8"E	Wastepond KIV/D, west	Soil
6	49°06'18.4"N 14°20'15.6"E	Wastepond KIV/D, north	Soil
7	49°06'22.8"N 14°20'31.2"E	Wastepond KIV/C2, west	Soil
8	49°06'30.2"N 14°21'17.3"E	Wastepond KIV/C1Z, north	Soil
9	49°06'12.4"N 14°21'28.7"E	Wastepond KIV/C1Z, east	Soil
10	49°05'47.9"N 14°21'55.5"E	Exit from the area of wasteponds, direction of Olešnik	Soil
11	49°06'15.3"N 14°22'40.3"E	Wastepond KIII, south-east	Soil
12	49°06'29.9"N 14°22'32.9"E	Wastepond KIII, west (near Olešnik)	Soil
13	49°05'16.7"N 14°19'58.4"E	Gutter at wastepond KI	Water
14	49°06'00.2"N 14°19'48.8"E	Gutter at wastepond KIV/D	Water
15	49°06'22.4"N 14°21'37.0"E	Svatopluk brook (near Olešnik)	Water
16	49°06'33.4"N 14°22'31.1"E	Svatopluk brook (direction Rojdanka)	Water
17	49°06'00.8"N 14°21'55.3"E	Hydrologic borehole HV-12	Water
18	49°05'22.0"N 14°20'27.9"E	Hydrologic borehole M-22	Water
19	49°05'17.3"N 14°19'50.3"E	Hydrologic borehole M-19	Water
20	49°05'11.6"N 14°20'01.8"E	Hydrologic borehole M-20	Water
21	49°05'54.4"N 14°20'07.3"E	Hydrologic borehole M-7	Water
22	49°06'02.9"N 14°19'43.8"E	Hydrologic borehole M-9	Water
23	49°06'02.9"N 14°19'43.8"E	Hydrologic borehole M-46	Water
24	49°06'16.8"N 14°20'28.1"E	Hydrologic borehole M-12	Water
25	49°05'42.0N 14°20'57.8"E	Hydrologic borehole M-2B	Water
26	49°05'46.5"N 14°21'50.6"E	Hydrologic borehole M-31	Water
27	49°05'40.1"N 14°21'27.4"E	Hydrologic borehole M-24	Water
28	49°06'26.2"N 14°21'30.5"E	Hydrologic borehole HJ-510	Water
29	49°06'32.3"N 14°21'10.1"E	Hydrologic borehole HJ-508	Water
30	49°06'39.2"N 14°20'42.7"E	Hydrologic borehole HJ-505	Water

sampling procedure, the work was carried out with the help of the personnel in the Monitoring Centre of DIAMO. After handing over, samples were stabilised and preserved with hydrochloric acid. The samples were also filtered, since ground water from the holes contains considerable proportions of insoluble material [15].

Determination of the activity concentration by gamma-ray spectrometry

Soil samples were submitted to the Laboratory of Dosimetry and Monitoring Radioactivity SUJCHBO, v. v. i. Kamenna for analysis. The laboratory is accredited by the Czech Institute for Accreditation in accordance with the standard ČSN EN ISO/IEC 17025:2005. Samples were transferred to Marinelli vessels and introduced in an appropriate geometry into the CANBERRA 35 PLUS analyser, whose main component was a CANBERRA GC3018 Hyper-Pure Germanium detector. Calibration was carried out and data were collected. After the measurement time, which was selected so that the measured value would be statistically significant, data were transferred to a desktop computer with GAMWIN software. If the sample activity did not reach statistical significance, the result is reported as *less than minimally detectable activity* (MDA). The GAMWIN software first performs a qualitative analysis, which results in the identification of certain isotopes by their gamma energies. Then follows the quantitative analysis in which the mass activity is determined for each radionuclide whose line has been identified in the spectrum. In this paper, we focus on ^{226}Ra and ^{238}U. These two cannot be measured directly, but their activities were assessed from the decay of their decay products ^{214}Pb, decay energy 352 keV, and ^{234}Th, decay energy 92.6 keV, resp. The background was measured periodically every three months and the net area of the background peak of the most recent measurement was subtracted from the net area of the radionuclide peak. The MDA is dependent on many factors (measuring time, radionuclide energy, background, *etc*.), but in most cases it was 0.001 mgL^{-1} and 0.03 BqL^{-1} for ^{238}U and ^{226}Ra, [15].

Determination of uranium concentration in water samples

A volume of the 0.5 ml sample was evaporated on a platinum dish at 105 °C, after which a homogeneous mixture of 98 % sodium fluoride and 2 % lithium fluoride was added, and the mixture was melted in a muffle furnace at 105 °C. After cooling down, the sample was measured as previously described. Samples with high uranium content were diluted and those with low uranium content were concentrated by evaporation of a suitable sample volume. The concentration of uranium was then calculated according to the following formula [16]

$$c \quad \frac{y_{\text{sample}} \; m_{\text{U}}}{(y_{\text{with addition}} \quad y_{\text{sample}})\, V}$$

where c [mgL^{-1}] is the concentration of uranium in water, V [L] – the volume of the water sample used for analysis, y_{sample} – the fluorometer reading measuring the sample as it is, $y_{\text{with addition}}$ – the fluorometer reading measuring the sample with an additional known quantity of uranium, m_{U} [mg] – the mass of the uranium added to the sample

Determination of the radium activity concentration in water samples

Determination of the ^{226}Ra activity per unit volume was carried out in accordance with standard PNU 830501 [17]. Radium was separated and concentrated by precipitation with barium sulphate and lead sulphate. The precipitate produced was transferred to test tubes and centrifugated. An ammoniacal solution of Chelaton 3 was then added and the precipitate was dissolved by heating on a sand bath. Barium-radium sulphate precipitate was produced by adding glacial acetic acid. The precipitate separated was mixed with the scintillator liquid detecting alpha-particles. The sediment was dried at 105 °C. The determination was carried out as previously described. The activity per unit volume was calculated according to the aforementioned formula adapted for radium instead of uranium [18].

Determination of the photon dose equivalent rate

For the measurement of the photon dose equivalent rate at the soil sampling sites, a FH 40G radiometer was used. This device is a proportional counter for gamma energies of 45 keV – 1.3 MeV. It is designed for the measurement of photon dose equivalent rates of 0.1 Svh^{-1} to 0.99 Svh^{-1}. The measurement of the photon dose equivalent rate was carried out at two different levels above the ground surface: 0.05 m and 1 m. Adherence to the same measurement level at all the points of measurement was controlled by a yardstick. Measurements of the photon dose equivalent were provided once at each height level for a time period of five minutes. The resulting values are presented as the average of values obtained during the time of measurement.

For the measurement of the dose equivalent level during walkabouts in the surroundings of sludge fields, a RT-30 spectrometer was used. This device is a NaI(Tl) semiconductor counter for gamma energies of 20 keV – 3

MeV. It is designed for the measurement of photon dose equivalent rates of 0.01 Svh^{-1} to 10 Svh^{-1}. The measurement route was adjusted to the terrain and accessibility. The measurement of the dose equivalent was carried out at a level of 1 m above the ground surface. The data from the instrument were transferred to the PC with the help of the GeoView software and summarized in tables. In the course of the measurements, the GPS values were automatically assigned to the values measured. After interconnection of these data with a map base, this outcome can be used for research purposes [19].

RESULTS AND DISCUSSION

Activity concentration of radium and concentration of uranium in water samples

The results for activity concentrations of radium and concentration of uranium in water samples are summarized in tab. 4. The values of the uranium concentration ranged from 0.001 to 0.275 mgL^{-1}, with an average of 0.07 0.07 mgL^{-1}. The highest values of the uranium concentration, which were found in samples from the inflow to the water treatment plant Pribram I (5.81 mgL^{-1}) and the Bytízský brook before that inflow (1.37 mgL^{-1}) were intentionally not included in this range. These samples were taken for the illustration of the efficacy with which the mine water processing is carried out.

After the purification of these waters in the water treatment plant Pribram I, at the point of release into the environment, the uranium concentration is 0.081 mgL^{-1}. In 46 % of samples taken, the values of the uranium concentration were lower than the detection limit.

The radium activity concentration measured in these water samples ranged from 0.030 to 0.093 BqL^{-1}, on average 0.06 0.02 BqL^{-1} (again excluding values of the ^{226}Ra activity in samples from the inflow to the water treatment plant Pribram I – 0.244 BqL^{-1} and the Bytizsky brook before that inflow – 0.187 BqL^{-1} which are not released into the environment without processing). In 58 % of samples taken, the activity values per unit volume were lower than the detection limit. A conservative approach was employed when handling values under the detection limit. At those points of the monitoring grid where no activity was detected, the detection limit was adopted as the value when calculating the average. The results obtained are very similar to those of our previous study [20].

The values found here are also comparable to those reported by Rapantova *et al.* [4], who focused on the assessment of mine water quality in shut-down uranium mines in the Czech Republic, where the ^{238}U concentration in most locations studied reached up to 0.45 mgL^{-1} and in the case of ^{226}Ra lay between 0.03 and 1.85 BqL^{-1} with few exceptions.

When our values are compared with those found around Mydlovary by Tomašek *et al*. [11] eight years earlier, namely average uranium concentrations between 0.007 and 4.5 mgL^{-1} (maximum values being 0.01 to 7.5 mgL^{-1}), and average radium concentrations mostly between 0.159 and 3.2 BqL^{-1} (with some exceptionally high values up to 6.4 BqL^{-1}), there is an obvious reduction in those radioactivity concentrations, which is associated with remediation and recultivation work in the processing plant area.

Table 4. Results of radium and uranium concentration in water

Sample No.	Area	Radium concentration [BqL^{-1}]	Uranium concentration [mgL^{-1}]
1	Pribram	0.197	1.370
2	Pribram	0.040	0.079
3	Pribram	0.244	5.810
4	Pribram	0.059	0.081
5	Straž pod Ralskem	<0.030	0.002
6	Straž pod Ralskem	<0.030	0.002
7	Straž pod Ralskem	<0.030	<0.001
8	Straž pod Ralskem	<0.030	<0.001
9	Mydlovary	<0.030	0.176
10	Mydlovary	<0.030	0.275
11	Mydlovary	<0.030	<0.005
12	Mydlovary	<0.030	0.003
13	Mydlovary (bore)	<0.030	<0.001
14	Mydlovary (bore)	<0.030	<0.001
15	Mydlovary (bore)	0.083	<0.001
16	Mydlovary (bore)	0.030	<0.001
17	Mydlovary (bore)	<0.030	0.001
18	Mydlovary (bore)	0.093	0.002
19	Mydlovary (bore)	0.041	0.103
20	Mydlovary (bore)	0.049	<0.001
21	Mydlovary (bore)	0.084	<0.001
22	Mydlovary (bore)	<0.030	<0.091
23	Mydlovary (bore)	<0.030	<0.105
24	Mydlovary (bore)	<0.030	<0.001
25	Mydlovary (bore)	0.077	0.004
26	Mydlovary (bore)	0.040	<0.001

A comparison of our results with those of other studies is shown in tab. 5.

The measured radium activity concentrations may be compared with derived radioactivity concentrations in water designed for human consumption in accordance with the European Council Directive 2013 /51/ EUROATOM [21]. These are of 3 BqL^{-1} and 0.5 BqL^{-1} for ^{238}U and ^{226}Ra, respectively. These values are calculated for a dose of 0.1 mSv at an intake of 730 litres annually. For the calculation of the total effective dose from the water ingestion, one can calculate that in an adult individual, the uranium concentration in water of 0.12 mgL^{-1} (equivalent 3 BqL^{-1}) corresponds to the total indicative annual dose of 27 Sv. An activity concentration of radium of 1 BqL^{-1} corresponds to the total effective annual dose of 49 Sv. The measured values can also be compared with recommended values of radionuclide contents in drinking water according to the World Health

Table 5. Comparison of radium and uranium concentration in waters with those of other studies

Location	^{226}Ra concentration [mBqL^{-1}]	^{238}U concentration [μgL^{-1}]
Czech Republic	30-93	1-275
Czech Republic [4]	300-1850	Up to 450
Czech Republic [11]	Up to 6400	Up to 7500
India [22]	5.2-38.1	0.1-3.75
Kazakhstan [23]	–	7.8-1250
Kyrgyzstan [24]	<10 to 890	1-213
Austria [25]	Up to 190	0.05-160

Organization of 10 BqL^{-1} for ^{238}U and 1 BqL^{-1} for ^{226}Ra [26]. In accordance with the valid Czech legislation [27], within the framework of the systematic measurement and evaluation of the content of natural radionuclides in water, it is necessary to determine the effective dose if the levels of the total alpha-particle activity per unit volume can exceed 0.2 BqL^{-1}. However, the values mentioned do not take into account the chemical toxicity of uranium.

The values measured within the framework of our research here are all under the established Czech [27], European [21] as well as international limits [26] and about half the samples were beyond the detection limit.

Measurement of activity concentrations in soil samples

For results of the analyses see fig. 5. The values of the ^{226}Ra activity concentration for the Pribram area ranged from 32 to 316 Bqkg^{-1} (average 111 95 Bqkg^{-1}), for the Straž pod Ralskem area from 13 to 88 Bqkg^{-1} (average of 34 21 Bqkg^{-1}) and for the Mydlovary area from 34 to 254 Bqkg^{-1} (average of 82 66 Bqkg^{-1}). The highest value measured was found in a soil sample coming from the Pribram area – sludge deposition field I Bytíz southeast. The values of the ^{238}U activity concentration range from 29 to 389 Bqkg^{-1} (average of 134 112 Bqkg^{-1}) for the Pribram area, 14 to 54 Bqkg^{-1} (average of 30 12 Bqkg^{-1}) for the Stráž pod Ralskem area and 16 to 238 Bqkg^{-1} (average of 86 56 Bqkg^{-1}) for the Mydlovary area. These values are very similar to the natural background reported for the Czech Republic by UNSCEAR [28], where for the ^{238}U concentration in the soil the originally mentioned range is of 68 to 220 Bqkg^{-1} and for ^{226}Ra it is of 76 to 275 Bqkg^{-1}.

The values measured within the framework of our current study correspond to those of earlier studies. For example, in our own preceding measuring campaign in the area around MAPE Mydlovary [20], about ten years ago, the average value of the activity concentration was of 14.8 to 219.6 Bqkg^{-1} for ^{226}Ra and 22.2 to 292.6 Bqkg^{-1} for ^{238}U. At the beginning of the 1990s, however, maximum values of the uranium contents were between 60 and 680 mgkg^{-1} and those of radium contents were of 2235 to 13697 Bqkg^{-1} [29], which shows the effectivity of remediation and recultivation carried out in the meantime.

Somewhat higher than normal values of radioactivity in soil have been found elsewhere in Europe where uranium mining or processing was carried out. For example, Carvalho *et al*. [30] reported values of 230 10 Bqkg^{-1} for ^{238}U and 619 96 Bqkg^{-1} for ^{226}Ra in soil samples from central Portugal; the values in the tailing pond were in the range of tens of Bqkg^{-1}. Similar values of uranium and radium content in soil (typically hundreds of Bqkg^{-1}) are also mentioned by the same authors in their later work [31]. The assessment of the uranium mining impact on the environment and health of persons in Portugal was further considered by Carvalho *et al*. [32, 33] as well as by Pereira *et al*. [34]. The ^{226}Ra activity concentrations were also reported for the area of Stara Stara Planina in Serbia [35] where there are former uranium mines, and in other areas which the authors considered potentially affected due to their location close to the mining facilities, or because of terrain hydrographies and geochemical properties. Values of tens to hundreds of Bqkg^{-1} (45 2 to 458 20) were found here, whereas elsewhere in areas of Stara Planina in which mining activities were never carried out, the recorded values were between 95 4 and 256 8 Bqkg^{-1}. In the case of ^{238}U, values ranged from 22 to 237 Bqkg^{-1} in areas affected by uranium processes, and from 13 to 49 Bqkg^{-1} in unaf-

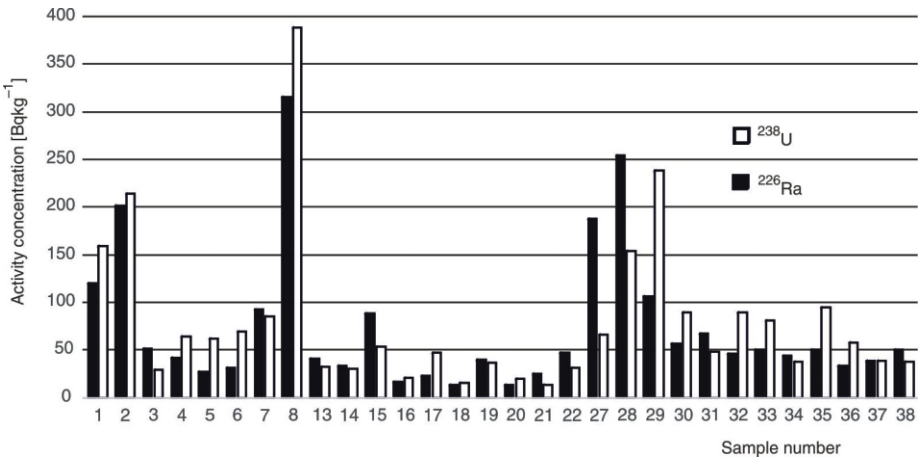

Figure 5. Results of ^{226}Ra and ^{238}U activities in samples taken

fected areas. Dimović *et al*. [36] also focused on the area of Stara Planina and measured similar activity concentrations of ^{226}Ra around the uranium mines as previously mentioned (from tens to hundreds of Bqkg^{-1}). At one site he found a value of 2600 100 Bqkg^{-1}, which was in a sample from the Mezdreja tailings pond. Activity concentrations of tens of Bqkg^{-1} for both ^{226}Ra and ^{238}U were reported by Ilić *et al*. [37], who analysed the radionuclide content of soil and clay samples in the area of the *Zbegovi* open-pit mine in Donje Crniljevo, Serbia. The average value for ^{226}Ra was 56 2 Bqkg^{-1} in soil and 61 2 Bqkg^{-1} in clay material from *Zbegovi*, for ^{238}U it was 23 8 Bqkg^{-1} in soil and <10 Bqkg^{-1} in clay material. These values for ^{238}U are somewhat higher than those presented by Marković *et al*. [38] for areas of Serbia not affected by uranium mining. Elevated ^{238}U and ^{226}Ra values were also reported by Križman *et al*. [39] in sediments in the vicinity of a uranium mine in the area of Žirovski vrch in Slovenia. Winkelmann *et al*. [40] conducted aerial measurements (with a gamma-ray spectrometric system carried by a helicopter) at several sites that contain residues from uranium mines, landfills and tailings ponds in Saxony and Thuringia in Germany, and compared these values with the results of ground measurements. The mean ^{226}Ra activity concentrations in the different waste rock dumps were found to be in the range 370 to 1600 Bqkg^{-1}. The highest mean ^{226}Ra activity concentrations for the tailings ponds were around 1300 Bqkg^{-1}. Environmental problems in connection with remediation provisions in the surroundings of the former uranium plant in Pridnieprov in Ukraine were studied by Lavrov and Voitsekhovych [41], who mentioned ^{226}Ra and ^{238}U activity concentrations of hundreds to thousands of Bqkg^{-1}. Some values measured in these areas are given in tab. 6.

Table 6. Comparison of radium and uranium concentration in soils with those of other studies

Location	Activity concentration ^{226}Ra [Bqkg^{-1}]	Activity concentration ^{238}U [Bqkg^{-1}]
Czech Republic	13-316	14-389
Czech Republic [20]	15-220	22-292
Czech Republic [28]	76-275	68-220
Czech Republic [29]	2235-13697	–
Portugal [30]	230 10	619 96
Portugal [31] Mining waste heap Soil I Soil II	 2096 180 832 62 659 20	 3257 94 1526 48 588 44
Serbia [35]	45-458	36-206
Serbia [36] Soil Clay material	 <2-98 61 2	 <7-70 <10
Serbia [37] Mezdreja clay tailings	28-400 2600 100	–
Serbia [38]	22-45	22-51
Slovenia [39]	8630 340	995 80
Germany [40]	370-1600	–
Ukraine [41]	30-36500	–

Determination of the dose equivalent rate

Gamma dose equivalent rates were measured at the same sites as the soil samples were taken. The measurements were carried out at two different levels above the ground surface (0.05 m and 1 m). The results are shown in fig. 6.

At a level of 0.05 m above the ground surface, the values range from 0.18 μSvh^{-1} to 0.48 μSvh^{-1} for the Pribram area, average 0.3 0.1, from 0.08 to 0.11 μSvh^{-1} for the Straž pod Ralskem area, average 0.10 0.01, and from 0.08 to 0.14 μSvh^{-1} for the Mydlovary area, average 0.10 0.02. At a level of 1 m, the values were almost the same: 0.05 to 0.18 μSvh^{-1} for the Pribram area (average 0.1 0.1), 0.07 to 0.12 μSvh^{-1} for the Straž pod Ralskem area (average 0.10 0.02) and 0.07 to 0.14 μSvh^{-1} for the Mydlovary area (average 0.10 0.02). The average values for both levels, 0.05 and 1 m, for all three areas of interest taken together were of 0.15 0.10 μSvh^{-1}.

The average values at both levels are very close to values reported by the Monitoring Centre of DIAMO, state enterprise, *i. e*. 0.18 μSvh^{-1}. The highest radium and uranium activity values measured cor-

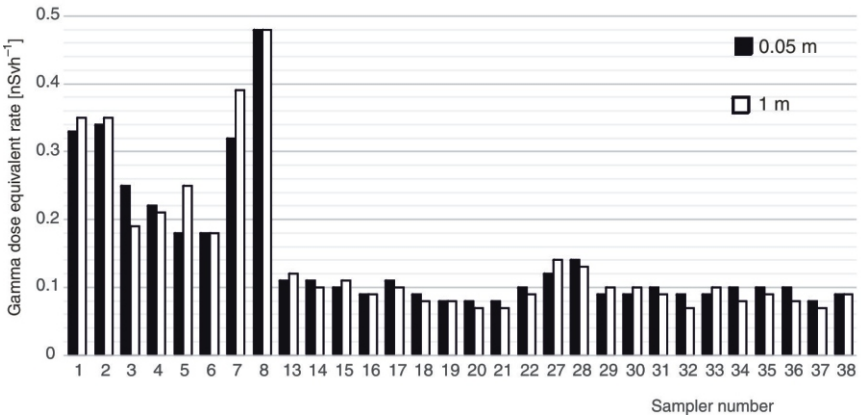

Figure 6. Dose equivelent rate in areas of interest at levels of 0.05 m and 1 m above surface

respond to the highest value of the gamma dose equivalent rate at the site of sampling; they were observed in the Pribram area, at the sludge deposit I Bytíz, southeast.

In our earlier study of the former MAPE Mydlovary area [20], measurements of the gamma dose equivalent rate at the same heights were carried out, the results from both examinations being very similar.

In the surroundings of the sludge deposits, gamma dose equivalent rates were measured along a path circumscribing the deposit. The path was adjusted to the terrain and accessibility. Measurements of the dose equivalent rate were carried out at a level of 1 m above the ground surface with the help of a yardstick. These converted data served as a basis for plotting graphs depending on particular areas of interest.

The highest values were found in the Pribram area with an average of 0.165 μSvh^{-1} and a maximum value near the southern part of the sludge deposit, part I, where the dose rate was 0.574 μSvh^{-1} (49°41'10.5"N, 14°03'42.5"E). In the Straž pod Ralskem area, the average value of the data measured was 0.040 μSvh^{-1}. The maximum was measured at the very beginning of the path near the southern part of the sludge deposit stage I, at borehole TBPK-16 (50°42'21.1"N, 14°46'04.9"E), and was of 0.080 μSvh^{-1}. In the third case, in the Mydlovary area, the average value was 0.089 μSvh^{-1}, and a maximum value of 0.396 $nSvh^{-1}$ was measured near the northern part of the KI sludge deposit, several tens of m behind the exit from the asphalt road behind the gate (49°05'34.7"N, 14°20'01.7"E).

The values measured here can be compared with the natural background estimated by for the Czech Republic, 0.14 μSvh^{-1}, which is employed in cases where it is or was impossible to use direct background measurements at the site. The State Institute of Radiation Protection (SURO) [42] specified that the radioactivity in the Czech Republic gives rise to gamma radiation at the ground surface level at dose equivalent rates between 0.006 and 0.245 μSvh^{-1}. In the case of the Straž pod Ralskem area, we can state that the values measured are mostly at the lower end of this range. It can therefore be assumed that radiation protection measures are effective. In the Mydlovary area, the average values of the dose equivalent rate are also mostly below the natural background, but the maximum value is considerably higher, and possibly indicates a remaining point contamination. In the area of the Pribram sludge deposits, the measured average as well as maximum values of the dose equivalent rate clearly exceed those for the natural background. However, both for Mydlovary and Pribram, the deviation from the range of natural dose equivalent rates is not extreme, *i*. *e*. about a factor of 2 at most.

The values measured can also be compared with the worldwide value of the dose equivalent rate from gamma rays, which is 0.058 μSvh^{-1} [28]. The average values in the areas of Mydlovary and more so in Pribram can obviously be considered as increased, but again by no more than a factor of 2.

Finally, we will try to assess the importance of particular exposure paths for various groups of exposed individuals. The external gamma-ray radiation is particularly of importance for the evaluation of the exposure of workers moving in the area of sludge deposits, especially in the course of carrying out recovery work in the area. These doses can be and are monitored by the responsible state company DIAMO, and their published reports indicate that they are well within the limits for professional exposure. The exposure of workers at the site in the years immediately following the shut-down of the facility were almost exclusively from gamma radiation, and their annual effective doses were 2-4 mSv, but these can be expected to have been further reduced with the progress in remediation [43].

An exposure of the population in the surrounding villages is clearly out of the question because of the distance. An exposure route that might have to be considered is the inhalation of radon and its decay products, but again it would affect only workers on-site, whereas the distance to settlements is such that radon from MAPE is not of concern for the general population. As to ingestion, given the low immission into the atmosphere and water, the possibility of affecting either workers or the population of the surrounding villages via this route would seem negligible.

If, just for the sake of a conservative estimate, we use the results of the gamma dose rate (in Gyh^{-1}) for the calculation of an annual effective dose, apply a factor of 0.7 SvGy to convert the absorbed dose in air to an effective dose for adults and assume an outdoor occupancy factor (the fraction of time spent outdoors) of 0.2 [28], then the average value of the annual effective dose could be 0.2 mSv in the Pribram area, 0.05 mSv in the Straž pod Ralskem area and 0.01 mSv for the Mydlovary area. The maximum values would be 0.7 mSv in the Pribram location, 0.01 mSv in the Straž pod Ralskem location and 0.49 mSv in the Mydlovary location.

These exposures would be well below the value of 1 mSv per year, which is established by the ICRP [44] for the general population, and certainly small in comparison with the average annual effective dose from natural sources of 2.4 mSv [45].

CONCLUSION

An evaluation of environmental contamination around uranium industry facilities in the Czech Republic was carried out through assessment of radioactivity concentrations in samples of surface water, ground water and soil, and of gamma dose equivalent rates in three areas of the Czech Republic which are known to be affected. The measured values of the ura-

nium and radium radioactivity are slightly higher but still comparable with the natural background levels. They correspond to the nature of the activities carried out in these areas and to the remediation and recultivation efforts of recent years. Somewhat higher values were found near sludge deposits, which remain long-term contamination sources. The results of our measurements and analysis are comparable with those from similar research projects implemented in the Czech Republic as well as abroad.

The measured values of the gamma dose equivalent rate suggest that workers carrying out remediation and recultivation work in the areas of interest are not at risk of exceeding dose limits for radiation workers and people living nearby are not at risk of exceeding the dose limits established for the general population. We have provided conservative estimates of doses for people that would stay in the area for relatively long times and have come to the conclusion that these are still below the acceptable doses for the general population according to the ICRP.

AUTHORS' CONTRIBUTIONS

The literature search was performed by R. Havrankova and Z. Freitinger Skalicka. Sample collection and measurement of dose rates under field conditions and related activities were carried out by E. Šimačkova. Graphs, figures and tables were created by E. Šimačkova, R. Havrankova, and J. Havranek. All authors participated in data analysis and discussion. The manuscript was compiled by R. Havrankova, E. Šimačkova, and F. Zolzer.

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ПРОЦЕНА РАДИОЛОШКОГ СТАЊА У РАЗЛИЧИТИМ ПОДРУЧЈИМА ЧЕШКЕ ПОД УТИЦАЈЕМ ВАЂЕЊА И ПРЕРАДЕ УРАНИЈУМА

Представљена су мерења концентрације активности природних радионуклида у одабраним узорцима воде и земљишта узетим са подручја под утицајем индустрије уранијума у околини Прибрама, Стража под Ралскем и Мидловари, у Чешкој. У овим областима такође је одређена јачина еквивалента дозе на локацијама узорковања и додатно у околини муљевитих поља. Концентрација активности узорака воде износила је 0.06 0.02 BqL^{-1} за ^{226}Ra и 0.07 0.07 mgL^{-1} за ^{238}U, док је средња концентрација активности узорака земљишта била 74 70 Bqkg^{-1} и 80 77 Bqkg^{-1} за ^{226}Ra и ^{238}U, респективно. Просечна вредност јачине еквивалента дозе била је 0.15 0.1 μSvh^{-1}. Ове вредности су у складу са природом индустријских активности које су се одвијале у овим областима и упоредиве су са резултатима мерења на сличним локацијама широм света.

Кључне речи: радиоактивност животне средине, концентрација активности, јачина еквивалента дозе, индустрија уранијума