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Radon levels after restoration of the U-mine disposal site

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Abstract. After cessation of the underground mining of uranium ore and production of uranium concentrate at Žirovski Vrh (Slovenia) in the period 1985–1990, two permanent surface disposal sites remained, namely, tailings pile and mine waste rock pile. Both disposal sites were of equal size of 4 hectares and were significant sources of radon. Their final restoration was designed in compliance with the condition of dose constraint for the public and authorized limits for radon exhalation from the remediated piles. In the late summer of 2008, a restoration of the mine waste pile was finished. Radon releases were reduced significantly by constructing an effective radon barrier of well compacted clayey material and a thick complex protective cover layer constructed over it. Radon exhalation rate from the mine waste area was lowered from primary level of 0.7 $Bq/m^2 \cdot s$ to natural levels (0.01 $Bq/m^2 \cdot s$), and consequently, ambient radon levels also decreased on the site and nearby environment. The average radon contribution from the remaining U-mine sources was estimated on the basis of the environmental measurements of radon concentrations; they dropped from initial 7–9 Bq/m^3 to approximately 3 Bq/m^3. Further reduction of outdoor radon concentrations is expected after 2010, since the restoration of another disposal site will have been completed by the end of this year. Public exposure due to industrial radon after the first phase of restoration satisfactorily meets the dose constraint level of 0.3 mSv/y, since it decreased to less than 0.1 mSv/y.

Key words: uranium mine • restoration • radon exhalation rate • radon concentrations • public exposure

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Introduction

Uranium mining and milling at Žirovski Vrh (Slovenia) ceased in 1990. During the 5 year operation, 452 tonnes of yellow cake were produced from uranium ore with a moderate content of uranium (< 0.1% U_3O_8). Two permanent surface disposal sites, a tailings pile at the Boršt site and a mine waste rock pile at the Jazbec site, both of an area of 4 hectares remained after cessation [2], not including several smaller and temporal disposal sites of mine waste, all situated on the hill slopes. The average content of ^{226}Ra in the mine waste rock is 0.7 kBq/kg and in the tailings 8.6 kBq/kg. Surface disposal sites were significant emission sources of radon, with the order of magnitude amounting to several TBq/y.

The Žirovski Vrh uranium mine is located in the sub-alpine region of Slovenia, in the valleys of the Brebovščica and Todraščica streams, and surrounded by steep slopes of the hills with the heights up to 900 m. The Alpine climate is the reason for a large amount of precipitation, with an annual average of 1700 mm and temperature inversion appearing 200 or more days a year. Strong inversions could last continuously for several days and significantly affect the enhancement of the ambient radon concentrations within the uranium mine area and its nearby environment.

The lower part of the Jazbec mine waste rock pile (foothills) begins in the Brebovščica valley at 427 m, while

Table 1. Composition of the final complex cover layer of the mine waste pile at the Jazbec site and characteristics of single layers

Thickness	Layer type	Material
–	Vegetation	Grass and bushes
0.25 m	Vegetation layer	Locally available soil, humus
0.5 m	Interim layer	Crushed Carnian solid sandstone 0/63 mm, coarse fraction
0.8 m	Protecting layer	Crushed Carnian solid sandstone 0/63 mm, fine fraction
0.4 m	Radon barrier	Weathered Carnian clayey sandstone/clay sand (compacted)
Up to 80 m	Waste rock pile	Mine waste rock

its plateau is located at 500 m; this height coincides with the height of an average temperature inversion layer and exhaled radon from the pile was mostly captured there. As regards radon, mine waste disposal site used to have the highest impact on the enhancement of outdoor radon concentrations in the valley and consequently also on the public exposure. Restoration of this pile started in 2006 with a recasting of its shape (lowering the inclination of a pile slope). Mine waste rock from two smaller temporary disposal sites was transported completely to the main disposal site at the Jazbec site, resulting in a final pile of 1.9 million tonnes of material with a grade of about 70 g/tonne U_3O_8. Adequately, shaped pile was then covered with an effective sealing layer for radon, of compacted Carnian clayey sandstones from a local deposit, what substantially decreased radon exhalation rate. The sealing layer was then covered additionally with a combined layer(s) of inert materials and locally available soil, with an overall thickness of 2 m (Table 1).

The effectiveness of the site restoration was tested primarily with measurements of exhalation rates on the site area: on a bare surface before restoration, after the construction of clayey radon barrier layer and finally after a completion of the final covering [1]. Secondly, ambient radon concentrations were measured within the disposal area and in its nearby surroundings, and compared with the measurements performed before the restoration of the site [2].

Water permeability for the radon barrier layer was 10^{-8} m/s, and this characteristic implied also a lowering of radon exhalation rate below the prescribed value of 0.1 Bq/m^2 s. The thick protective layer with a surface humus cover (Table 1) prevents the drying as well as freezing of the sealing clayey material. The restoration progressed gradually and was completed in autumn 2008 (Fig. 1).

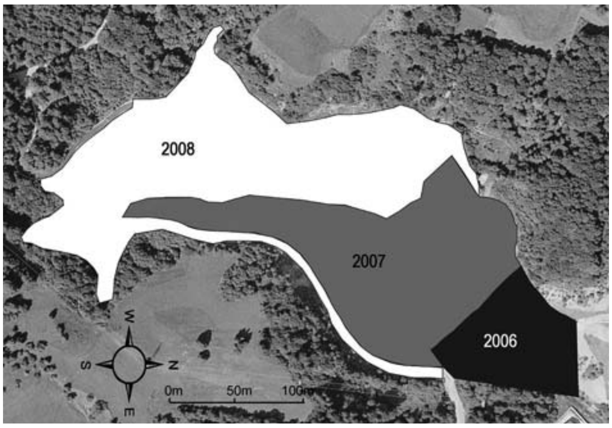

Fig. 1. Map of the mine waste rock pile with indicated restoration phases in the period 2006–2008; geometric centre of the pile lies at $X = 46°04'51''$, $Y = 14°09'32''$.

The aim of the study was to compare radon levels before and after restoration works and to establish the effectiveness of covering on the restored disposal site. The presented results refer only to the Jazbec disposal site with mine waste rock. The termination of the in-process restoration works on the other disposal site with mill tailings at Boršt is expected by the end of 2009.

Methods

Radon was sampled with passive devices such as charcoal absorbers and solid state nuclear track detectors and somewhere also with active devices. Several measurement campaigns were conducted before the restoration started, during the works and after its completion. Two kinds of measurements have been performed: radon exhalation rates from the pile and ambient radon concentrations.

For measurements of radon exhalation rate mostly the inverted cylindrical containers (Ø10 cm, $h = 15$ cm) with charcoal canister were used. Accumulation time was typically 48 h. Radon was sampled on the bare surface of the pile, on the test fields with different thicknesses of cover layer and cover materials, and on the final complex covering. Measurements were performed in all weather situations and throughout all the year to obtain a reliable annual average value. Simultaneous measurements of radon exhalation with an AlphaGUARD ionization chamber (Genitron, Germany) and with charcoal absorbers gave completely comparable results. Both methods required careful preparation of the sampling, particularly the perfect sealing of the device cylinder with the ground surface. The results obtained during heavy rain periods, or at temperatures below 0°C, or in hot and dry summer days, or during strong winds are usually questionable and were mostly disregarded. Environmental parameters in soil (soil temperature, moisture saturation) were not measured.

Passive measurements of outdoor radon concentrations were performed continuously with solid state nuclear track detectors from the Forschungszentrum Karlsruhe (Germany). Sampling period extended to 3 months due to relatively low concentrations. To obtain more reliable statistical results, three detectors were placed at each sampling location. The main measurement points are indicated on the map of the Žirovski Vrh uranium mine site (Fig. 2). Short time measurements of radon were performed several times per year with Environmental Protection Agency (EPA), USA charcoal absorbers and with the AlphaGUARD ionisation chamber.

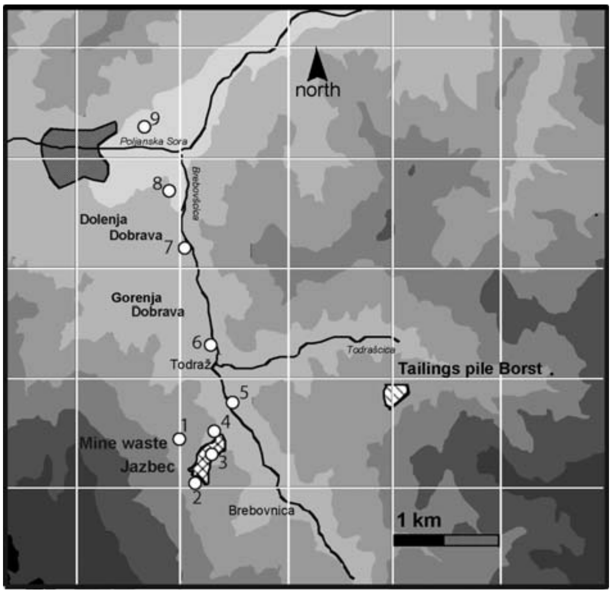

Fig. 2. Map of the Žirovski Vrh site with location of the mine waste pile and measurement points for ambient radon.

Radon short-lived decay products were monitored exclusively with active devices, mostly with ARM-200, product of Tracerlab (Germany). Measurements were being performed continuously [3], also during the entire restoration period and were intended to trace changes of concentration levels. Results were expressed as radon equilibrium equivalent concentrations (EEC). Standard deviation of the results at the EEC level of 20 Bq/m^3 is ± 10%.

Results and discussion

Radon exhalation rate

The first tests for radon barrier layer on the mine waste disposal site at Jazbec were performed with proper clay of the verified quality but from a distant location. Repeated field tests were later performed with localy available Carnian clayey sandstones: experiments showed that this material, properly treated and compacted and built in a thicker layer, could provide a similar barrier for radon as genuine clay. After the final restoration of the Jazbec site, radon exhalation rate dropped significantly from an average value of 0.73 Bq/m^2·s ± 0.49 Bq/m^2·s to only 0.011 Bq/m^2·s ± 0.007 Bq/m^2·s (Fig. 3), i.e. to less than worldwide levels 0.016 Bq/m^2·s [7]. The protective cover layer on the surface is of a material with low mass activity concentration of ^{226}Ra of only about 25 Bq/kg. Local soil from the Žirovski Vrh site contains about 60 Bq/kg of ^{226}Ra. Radon exhalation from the finally covered area originates, therefore, only from the covering material itself.

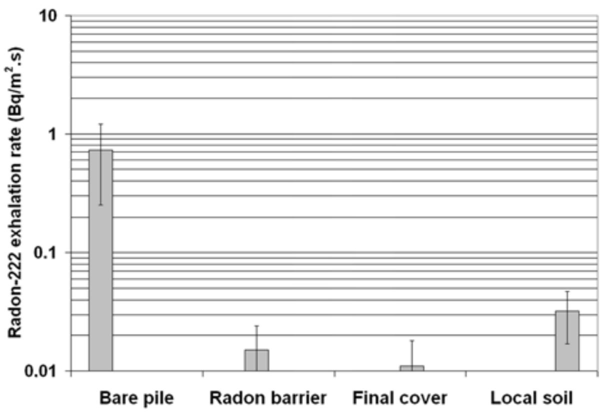

Fig. 3. Radon exhalation rate from the bare pile (first bar) was effectively reduced due to the compacted layer of clay (second bar) and further lowered with a final soil cover of low radioactivity content (third bar); exhalation rate from the local soil (fourth bar) is substantially higher.

Outdoor radon concentrations

Results of continuous measurements of radon decay products on the disposal site showed a constantly decreasing trend of EEC levels during all stages of the restoration period 2006–2008. Typical mean EEC levels before restoration were approximately 20 Bq/m^3 and dropped finally close to natural levels of 15 Bq/m^3 (Fig. 4). In general, EEC values should be higher in a cold period of the year due to longer lasting stable atmosphere during temperature inversions. Some observed deviations on the graph in winter time are the consequence of unexpected weather conditions – such as, for instance, less stable atmosphere, heavy precipitation, longer lasting frozen ground, etc.

Outdoor concentrations of ^{222}Rn were measured on the disposal site and in its surroundings, as shown in Fig. 2. Measurement points were extended along the trajectory of air movement [2]. The first group of five measurement spots is related to the site and its vicinity: sampling points 1 and 2 are located 0.3 km above the centre of the radon source, point 3 is on the disposal area and point 4 is located at its foothills. The nearby vicinity accessible to the public is indicated as the location 5, while other points are located downstream the main valley, at the near and distant settlements: Todraž (point 6, distance 1.3 km), Gorenja Dobrava (point 7, distance 2.3 km), Dolenja Dobrava (point 8, distance 2.8 km) and Gorenja vas (point 9, 3.6 km away from the central point of the disposal).

Before the restoration, concentrations of ^{222}Rn, appearing within the bare waste rock pile at the Jazbec site, exceeded 90 Bq/m^3 in an annual average and were

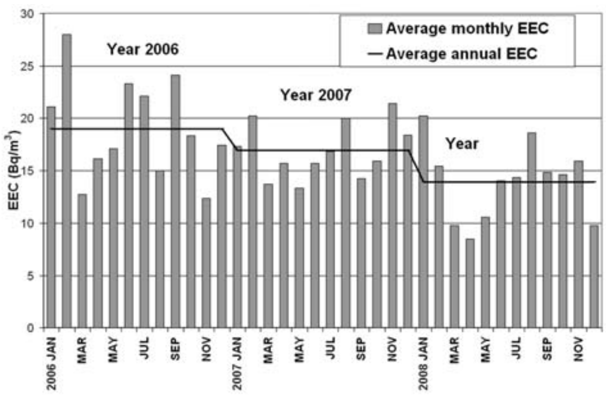

Fig. 4. EEC of radon dropped in the last year of restoration works (2008) close to natural background levels (15 Bq/m^3).

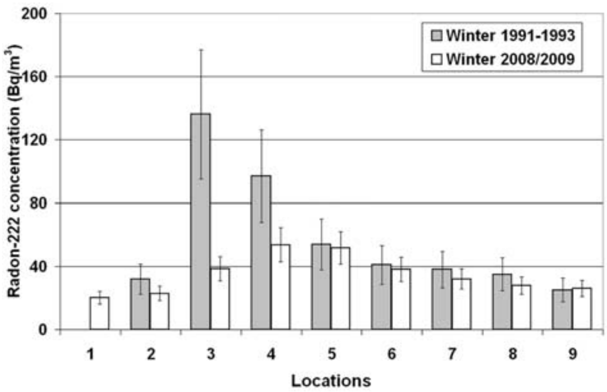

Fig. 5. Radon concentrations measured in winter time along the topographical profile of the Jazbec disposal site: before restoration (grey) and after restoration (white).

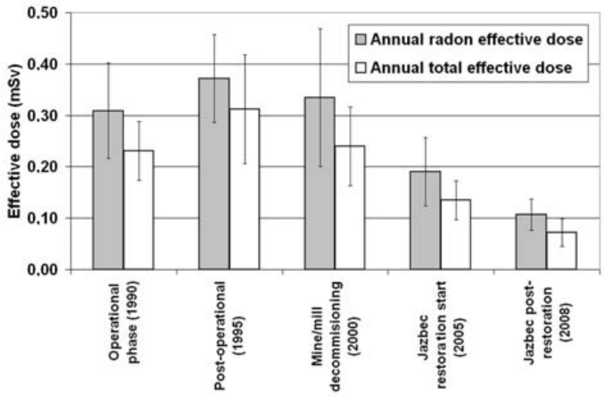

Fig. 6. Total effective dose (grey) and radon effective dose (white) to the population in different phases of the lifetime of the Žirovski Vrh U-mine.

about 140 Bq/m^3 in winter time [2]. After restoration they decreased quite close to background levels (30 Bq/m^3) as indicted in Fig. 5. Maximal hourly ^{222}Rn concentrations on the pile, measured before restoration works, had reached up to 400 Bq/m^3, but when restoration was completed, the peak values dropped below 80 Bq/m^3.

Further monitoring of radon levels in the former U-mine environment is provided for the next 5 years after restoration; after that, only institutional control of areas will be established, according to the current practices [1]. It was anticipated that the average radon concentrations outdoors, caused by the U-mine radon sources, would decrease from 7–9 Bq/m^3 as determined in the period of mine operation [4] to the values less than 3 Bq/m^3 after restoration of the site. It is difficult to precisely estimate the contribution of industrial radon from the U-mine facilities to the environment. Natural variations of radon levels and the accuracy of radon measurements require good statistics and, therefore, numerous results have to be provided.

Dosimetric implications

During the operational period and also in the pre-restoration period, radon released from the mine sources contributed up to 75–80% of the additional exposure to the public from the reference group or about 0.3 mSv/y [4]. The reference group consists of 350 inhabitants, 20% of whom are children below 15 years. The reduction of concentrations of industrial radon in the environment after of restoration has important dosimetric implications. Estimated public exposure for adult individuals of the reference group due to inhalation of radon decay products was significantly diminished in the last year and dropped to less than 0.1 mSv/y (Fig. 6). Public exposure already meets the future dose constraint level of 0.3 mSv/y for post-restoration era even before the completion of the first stage of restoration. This effective dose limitation is considerably more severe than the general limit for the public of 1 mSv/y, applied in several other countries in European Union (EU) [5].

Conclusions

Measurements of radon during and after the first phase of restoration of the disposal sites at Žirovski Vrh show definitively a lower impact to the environment. Restoration of the disposal site with mine waste rock at Jazbec resulted in the following features. Radon barrier of local clayey material with a low content of ^{226}Ra was well compacted and properly built, so that its original moisture was conserved within the cover. Radon exhalation rates significantly dropped to 0.01 Bq/m^2·s, the value that is an order of magnitude lower than the authorized level of 0.1 Bq/m^2·s and even lower than levels obtained on the natural soil in the region. Outdoor radon concentrations on the site and in the nearby environment evidently decreased. The reduction of outdoor ^{222}Rn concentrations consequently diminished the estimated annual effective dose for the reference group to less than 0.1 mSv [6], well below the prescribed authorized level for the post-restoration era and far below the general dose limit for the public.

The second disposal site, i.e. tailings pile, located at Boršt, at the air distance of 2 km from the Jazbec pile, was still in the restoration phase during the research study. Composition of its cover layer is quite similar as in the case of the Jazbec disposal site, with the exception that the radon barrier layer is made of clay with a very low water permeability of 5×10^{-10} m/s. However, this low permeability is sufficient to reduce a radon exhalation rate to less than the prescribed level of 0.7 Bq/m^2·s. Real measurements on the partly covered tailings pile with the final complex cover have already showed that the levels of radon exhalation rate were within the order of magnitude of several 10^{-2} Bq/m^2·s. Further reduction of ambient radon concentrations in the affected area is expected after 2010 when the restoration of the tailing pile will be completed till the end of this year.

Based on the results of environmental monitoring of radioactivity in the past two decades, it has been established that other factors besides radon do not affect the environment considerably. The concentrations of air dust particles with long-lived radionuclides of ^{238}U decay series became negligible after cessation of mining and milling activity. Contamination of the surface and the ground waters has been on the level of some percent of the derived concentrations for drinking water and, therefore, no discharge waters treatment has been required from the radiological point of view. External radiation on the restored disposal site equals background levels of the region and is below the prescribed limit of double background level (0.2 μGy/h).

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