Radon events in the U-mine environment and related radiation exposure

Milko J. Križman, Jože Rojc, Josef E. Peter

Abstract. The term "radon event" indicates here a sudden appearance of enhanced radon concentrations, observed like well expressed peaks in time series of radon concentrations. The peaks are superimposed on normal diurnal periodical curves. The characteristics of radon events are high peak values, a rather short duration and a low radon equilibrium factor. Since radon events appear only in the environment near significant radon emission sources, they were investigated in more detail in the case of the former Žirovski Vrh uranium mine (Slovenia), using the existing network of continuous radon progeny measuring devices. Eight different types of radon events were identified in the vicinity of the U-mine disposal sites, lasting for some hours and with the range of their peak levels of equilibrium equivalent concentrations (EEC) of radon from a few Bq·m⁻³ to over 200 Bq·m⁻³. Exposures to radon events in units of Bq·h·m⁻³ were estimated for adult individuals of the reference group. They resulted in relatively high effective doses of the range 1–5 μ Sv per a single event, thus exceeding, e.g. the total effective dose for the public due to radioactive discharges from most nuclear facilities during the whole year.

Key words: radon • uranium mine • environment • radon event • effective dose

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Introduction

Radon concentrations in the natural environment in stable weather conditions have more or less diurnal periodical behaviour, with clear minima during the day and maxima during the night [7]. When boundary layer stabilizes during late afternoon, night and early morning, radon is trapped within the inversion layer and its level increases. During the day, a vertical mixing is the major process for restoring low concentrations of radon and its short-lived progeny, due to a dilution with clean air from above the mixing layer. In unstable and windy weather, the near-ground radon concentrations are quite low due to efficient mixing, and the differences between minimum and maximum daily levels are rather small.

Within the area where several radon sources are present (e.g. U-mine area with tailings piles, mine ventilation shafts, etc.), an industrial radon diurnal behaviour is, in principle, the same as for natural radon, but in some meteorological conditions it might be modified. Namely, a carefully designed network of active radon monitors enabled to detect sharp and high peaks in time series of radon concentrations that were superimposed on normal periodical curves [3, 5]. These radon peaks are an indication of a sudden appearance of radon with enhanced concentration and they have never been reported in relation to the uranium mine and to radiation exposure. They are termed here as

Uranium deposits at Žirovski Vrh were discovered in the early 1960s. Mining and milling were carried out much later for a relatively short period from 1985 to 1990. During the operational period, 452 tonnes of yellow cake were produced from the ore with a moderate content of uranium (near 0.1% U₃O₈). The industrial complex was located mainly in a deep valley and all disposal sites were either situated on or lean against the hill slopes. Frequent and strong temperature inversions (about 55% of the time of the year, temperature gradient mostly 0.02–0.03 K/m) are important characteristics of the site. After cessation of the mining operation, the underground mine was closed and not ventilated any more. Several disposal sites with bare areas remained, among them a large waste rock pile and a large tailings pile. Each of them had an area of 4 hectares and both were significant sources of radon. At Żirovski Vrh, inhalation of industrial radon from the uranium mine area was found to be the most important exposure pathway for a long period of time since it contributed up to 80% of the annual mine-related effective dose to the public [4] or about 0.30 mSv [2]. This value dropped after the decommissioning of the mine and the milling plant and after the restoration of the main disposal sites down to less than 0.1 mSv per year. The total exposure of the population living in the U-mine surroundings was estimated - based on environmental radioactivity measurements, including indoor radon and external radiation - to be on average 5.5 mSv per year [2].

The aim of this paper was to present the existence of radon events identified in the environment of the former uranium mine and their characteristics. Beside this evidence of a dynamic impact to the environment, radiation exposures to the public were estimated, caused by various single radon events. Such exposures were likely to be received by a representative individual of the nearby reference groups. So far the exposures have never been evaluated yet.

Materials and methods

Measurement devices for continuous monitoring of radon short-lived decay products were originally designed by the former GSF Institute (now Helmholtz Zentrum München, Neuherberg, Germany). The instruments measured radon equilibrium equivalent concentrations (EEC) and alpha spectra on hourly basis, while local temperatures and flow rate through the filter were recorded every 10 min. A unique comprehensive network for continuous measurements was set up in the early 1990s in the frame of the Slovenian-Bavarian bilateral research and technical collaboration [5, 6]. Measurements were performed in the period that followed mining and milling cessation and preceded the final restoration of disposal sites. The data files recorded by the radon devices were stored locally in files, covering at least a 10-day period. Fourteen automatic stations were installed at key locations, on-site uranium mining area (within disposal sites and on their perimeters), and

off-site, including settlements along the affected valleys and a reference point [2, 6]. The software was developed for display and analysis of the data.

Exposure to the radon event is defined as a time integral of the equilibrium equivalent concentration (EEC) of radon for the duration of the event $(0 \rightarrow T)$ and expressed in terms of Bq·h·m⁻³. The dose conversion factor (DCF) for radon decay products was taken according to the dose convention at work 8.0 nSv/Bq·h·m⁻³, derived from ICRP Publication 65 [1]. The effective dose (*E*) for a single radon event, received by a representative person of the public (nearby inhabitant) is estimated by the formula:

(1)
$$E_{\text{event}} = \text{DCF} \cdot \int_0^T \text{EEC}(t) dt$$

The EEC levels during the particular events and the duration of such events were obtained from the recorded data, measured continuously at the points of interest. Simultaneously, measurements were performed at the relevant reference locations, outside the affected area, indicating the background level. Only the difference between both data sets was taken into account for the assessment of the effective dose.

Results and discussion

During unstable windy weather radon events are generally very weak and cannot even be detected. The events with high radon or EECs were identified easily in stable weather conditions. Radon events are characterized by peak values of EEC or ²²²Rn concentrations, with an increasing rate of these concentrations, and with a low equilibrium factor. They differ also by their duration, locations of appearance, time of the day and season of their appearance.

EEC peak values during the radon event ranged usually from 50 Bq·m⁻³ up to 200 Bq·m⁻³ and more (see Table 1, column 3) and exceeded maximum levels of the natural radon background for several times. They usually appeared in the late afternoon or in the early evening when temperature inversion establishes and also in the early or late morning, when temperature inversion decays.

Besides the peak value, an increasing rate of EEC is another parameter for recognition of the radon event. The EEC concentrations of industrial radon are rising far more rapidly than those of radon of natural origin, resulting in the time series diagrams as steep and high peaks. In the case of intensive events, this rate is of the order of 5 Bq·m⁻³/h to 100 Bq·m⁻³/h (see Table 1, column 4). Sharp increases of EEC are typical of the warm period of the year.

A low equilibrium factor, if compared to that of radon of natural origin, is also a characteristic feature for a radon event. It is of the order of 0.1–0.3 and can be analysed with alpha spectrometry of air filters or simultaneous measurements of the EEC and ²²²Rn concentrations. A low equilibrium factor of radon was also measured on the site of uranium mine disposals at Crossen in Saxony, in the range of 0.03–0.25 [8].

Radon from the uranium mine sources was dispersed to the environment and its impact to the environment

No.	Location of the events, air movement, and period of appearance	Max EEC (Bq·m ⁻³)	Max ∆EEC/t (Bq·m ⁻³ /h)	Event duration (h)	Net EEC exposition* (Bq·h·m ⁻³)	Effective dose* (µSv)
1	Waste rock pile: foothills, downstream airflow, late afternoon, May–September	270	120	8	660	5.3
2	Waste rock pile: plateau plateau ground inversion, early evening, May–September	205	65	8	550	4.4
3	Waste rock pile: plateau, inversion over plateau, morning, winter	135	30	10	480	3.8
4	Tailings pile: foothills, stationary downstream airflow, early evening, May–September	105	10	5	300	2.3
5	Tailings pile: foothills stationary downstream airflow, early morning, winter	55	4	8	140	1.1
6	Tailings pile: plateau ground inversion decay, late morning, early spring	65	25	4	95	0.8
7	Tailings pile: upwards upstream airflow, late morning, winter	60	20	8	160	1.3
8	Tailings pile: plateau horizontal transport from plateau, late morning, winter	50	6	5	75	0.6

Table 1. The identified radon events, their location and appearance, their characteristics, and the resulted effective doses

* Calculated for the radon event, lasting for 5 h background EEC subtracted.

was not constant in regard to time development and locations of appearance.

Types of radon events

In the case of the Żirovski Vrh mine, altogether eight radon events were identified that differed by their intensity, duration time, radon equilibrium factor and magnitude of radiation exposure. They appeared only at certain locations and certain time and season of the year. The most significant radon events were detected at foothills of the disposal sites (piles). Time series of EEC, recorded on the sites and at reference locations simultaneously, are shown for a comparison and for the event evaluation.

Five types of radon events were related to the tailings pile at the Boršt site and its vicinity, located about 150 m above the lateral valley. The cross-section of the area with indicated measurement points is presented in Fig. 1. The bare pile, a strong radon source with 4 TBq of ²²²Rn per year generated radon events with radon expositions of 50–300 Bq·h·m⁻³ per event.

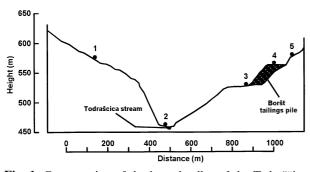


Fig. 1. Cross-section of the lateral valley of the Todraščica stream with the elevated position of the Boršt tailings pile and with indicated measurement locations.

Figure 2 shows the case of a radon event appearing within the plateau height when inversion layer was formed just above the pile plateau. The duration of the event on the plateau was shorter than at locations below the pile since the temperature inversion lasted for a shorter period at higher altitudes.

Medium levels of radon exposition per event $(160 \text{ Bq}\cdot\text{h}\cdot\text{m}^{-3})$ were typical of the upwards zone, outside the pile. Such events were rarely observed, only during moderate thermal buoyancy (Fig. 3). They appeared in the late morning and are the only radon events recorded during the day.

The intensive radon event of 300 Bq·h·m⁻³ was observed in a zone below the disposal site. A stationary airflow was moving downwards the plateau and the pile slope, rich with fresh radon of low equilibrium factor. Figure 4 shows the radon event, observed at foothills of the tailing pile at Boršt in the warm period of the year (May–September).

The lowest radon exposition $(50 \text{ Bq} \cdot \text{h} \cdot \text{m}^{-3})$ belonged to the event, appearing at a location at the opposite site

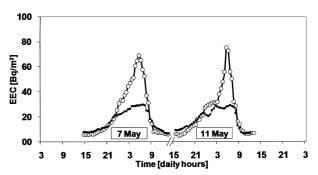


Fig. 2. Two radon events $(-\circ-)$ identified at the location of the Boršt pile plateau (location no. 4 in Fig. 1) during short term morning inversions; local background levels $(-\blacksquare-)$ were also recorded.

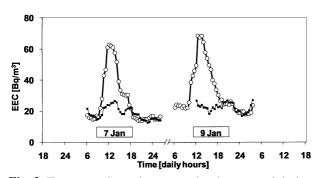


Fig. 3. Two successive radon events $(-\circ-)$ measured during the day at the location uphill the Boršt tailings pile (location no. 5 in Fig. 1) in the case of thermal buoyancy; local background levels $(-\bullet-)$ are shown for a comparison.

of the valley (location no. 1 in Fig. 1): radon was being transported horizontally from the pile to the 0.8 km distant environment.

Another three types of events are related to the mine waste rock pile at Jazbec. The pile extends from the foothill – located in the main valley of the Brebovščica stream – to the upper end of the plateau, situated at a relative height of about 70 m above the valley (Fig. 5).

The most significant type of radon event is shown in Fig. 6 and appeared in the late afternoons from May to September. The highest estimated EEC expositions ranged from 300–600 Bq·h·m⁻³ per single event and lasted up to 8 h. The shape of the event curve led to an assumption that an additional source of radon was present. It was confirmed that the outlet of the drainage system below the pile, discharged timely radon, collected in the pipelines within the pile body. Radon exited from the drainage system when outdoor temperatures exceeded 14°C.

The EEC increasing rates were high, within the interval of 65–120 Bq·m⁻³/h. Such radon events have been frequently observed in the late afternoons, in the phase of development of a temperature inversion layer, exclusively during the warm period of the year.

A further strong radon event, identified in the nearby vicinity of the Jazbec pile plateau (location no. 1 in Fig. 5), usually appeared later in the evening, when the height of the temperature inversion layer reached the elevated plateau, quite frequently in the warm period of the year. The corresponding EEC peak value exceeded $200 \text{ Bq}\cdot\text{m}^{-2}$ and net radon exposition was estimated up to 550 Bq·h·m⁻³, as indicated in the sixth column of Table 1.

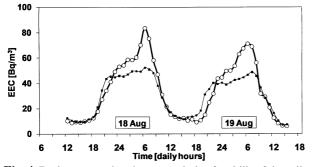


Fig. 4. Radon events $(-\circ-)$ as recorded at foothills of the tailings Boršt pile (location no. 3 in Fig. 1), in the late morning, during long lasting inversion; local background levels $(-\bullet-)$ are also presented.

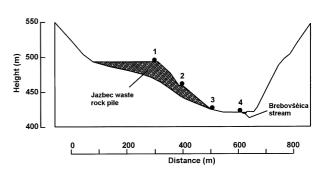


Fig. 5. Cross-section of the main valley of the Brebovščica stream with the position of the Jazbec waste rock pile and with indicated measurement locations.

Another type of radon events existed that was not easily observed, since there were neither evidently high peak values nor a considerable increasing rate of concentrations in a time diagram. These events were recognized mostly by a low equilibrium factor, if compared to that of radon of natural origin, typically F = 0.2. Characteristics of such type of events are moderately high and uniform concentrations within the pile (location 4 in Fig. 5), lasting for 8 h or more, and a quite slow increasing rate of EEC, less than 5 Bq·m⁻³/h. Radon events of this type appeared in cold periods of the year and in stable weather conditions when the disposal site was captured within the long lasting temperature inversion layer. During the event, a stationary airflow was traversing the disposal site and consequently bearing exhaled radon within.

Summary on the radon events at the Žirovski Vrh site

During unstable windy weather the impact of the U-mine to the environment is weak and radon events cannot even be detected. Only well expressed events with high radon concentrations were identified easily, and all of them were measured in stable weather conditions. The continuous measurement devices showed the same behaviour in periodicity of diurnal concentrations as for background radon all over the year. In the vicinity of strong radon sources, irregular curve shapes in time diagrams of ²²²Rn concentrations were observed as presented in Fig. 2 to Fig. 6.

The highest EEC values of an event were observed when the entire disposal site was captured within the

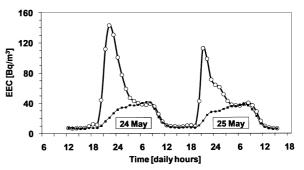


Fig. 6. Two successive radon events $(-\circ-)$ observed at the foothills of the Jazbec waste rock pile (location no. 4 in Fig. 5), in the late afternoon of the warm period of the year, with indicated local background levels $(-\bullet-)$.

boundary layer. Topographic configuration of the terrain and the position of the radon source may also play an important role. For radon sources below the average temperature inversion height (500 m is typically for the Žirovski Vrh environment), the frequency of appearance of radon events was significantly higher than for sources located at elevated positions.

The radon event is recognized by the simultaneous increase of radon or radon daughter concentrations and appearance of low equilibrium factor, easily identified by alpha spectrometry analysis. The events were regularly detected within the impact area of the radon emission sources, but not necessarily only within the controlled zone of the facility. Airflow bearing radon from the source can travel the distances of some tens up to a 100 m, i.e. to the areas freely accessed by the local people.

Usually, the intensive radon events appeared frequently in the warm part of the year. The highest exposures of the order of microsieverts were estimated for the events with the highest EEC peak values (over 100 Bq·m⁻³) and highest EEC increasing rates (over 30 Bq·m⁻³/h). Events with stationary downstream airflow bearing radon had much lower EEC peak values and smaller increasing rates, and, therefore, resulted in low potential exposures (about 1 μ Sv per event).

Radon events were observed at both disposal sites before restoration works took place. Later, after restoration of the mine waste pile in 2008, levels of industrial radon declined and the radon events at the previously monitored locations have not been recorded any more.

Effective dose due to radon events

Radiation exposures to radon events may be received by individuals who are really staying near the fence of the controlled area at times for some hours during their work (haymaking, cutting and gathering wood, picking blackberries and mushroom, fishing) at the time of the radon event. A representative person from the reference group is prone to receive an effective dose that is not entirely negligible from the radiological point of view. The exposure was of the order of magnitude of some microsieverts, ranging from 0.6 to 5.3 μ Sv per single event (see the effective dose values in Table 1). It likely exceeded 10 μ Sv, if a person having been exposed to several radon events.

External dose due to submersion in the radon cloud is four orders of magnitude lower than the above values due to inhalation dose and is estimated to be less than 0.1 nSv per event.

Conclusion

The network of automatic radon stations definitely showed that the environmental impact of mine radon is neither uniform nor constant with time and in space [3]. The exposure to radon is clearly related to radon events, short time episodes with high peaks of radon concentrations. At short distances from the source and rather far from the inhabited area, radon events are, in principle, the prevailing mode for radon exposure of members of the reference group.

Single radon events can result in radiation exposures 1 to 5 μ Sv per event, likely received by a reference person staying during his work at the accessible locations near the mine and mill waste disposal sites (radon sources). An exposure due to one single radon event at the Žirovski Vrh uranium mine is of the same order or higher as an effective dose received by a representative of the reference group living in the vicinity of the nuclear installation (e.g. at the Krško nuclear power plant) due to its radioactive discharges during the whole year.

The results on radon events at the Żirovski Vrh uranium mine and the resulting potential exposures can be applied elsewhere, particularly in cases with radon emission sources in a similar topographic situation.

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