EXPOSURE TO RADON AT UNDERGROUND WORKPLACES IZPOSTAVLJENOST RADONU NA PODZEMNIH DELOVNIH **MESTIH**

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Abstract

Aim: The main aim of this contribution is to review the radon (²²²Rn) surveys carried out over the last two decades at underground workplaces in Slovenia in coal mines, karstic caves, water supply plants, wineries and hospitals. **Methods:** Alpha scintillation cells, etched track detectors, an AlphaGuard PQ2000 multiparameter radon monitor and EQF 3020 and EQF 3020-2 radon and radon progeny monitor systems were used to measure concentrations of radon and radon short-lived decay products, equilibrium factor and unattached fraction of radon decay products in air.

Conclusions: (1) Radon levels are low in coal mines; (2) elevated radon levels can be present in karstic caves; prior to a longer stay in a karstic cave, the radon level should be checked and, if necessary, stay in the cave limited; (3) although elevated radon levels are frequently found at water supply plants, attendance times at underground workplaces are short and the effective doses low; care is necessary for longer maintenance works; (4) under normal working regimes in a winery, exposure to radon in underground facilities is low; (5) radon levels are low in the majority of basement rooms in hospitals, but precautions are necessary in old buildings where the floor may not always be a sufficient barrier to Rn entry, and indoor radon levels may be elevated.

Key words: radon, radon short-lived decay products, underground workplaces, effective doses

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Izvleček

Cilj: Glavni namen je bil podati pregled preiskav radona (²²²Rn) na podzemnih delovnih mestih v zadnjih dveh desetletjih, in sicer v premogovnikih, kraških jamah, vodnih zajetjih, vinskih kleteh in bolnišnicah.

Metode: Uporabljali smo komplementarno merilno opremo, s katero smo merili koncentracijo radona (Rn) in radonovih kratkoživih razpadnih produktov (RnDP), faktor radioaktivnega ravnotežja med Rn and RnDP ter delež prostih RnDP, in sicer: alfa scintilacijske celice, detektorje jedrskih sledi, radonski merilnik AlphaGuard PQ2000 ter kombinirana merilnika EQF 3020 in EQF 3020-2 za Rn in RnDP.

Zaključki: (1) Koncentracije Rn v zraku rudnikov so bile nizke. (2) V kraških jamah lahko naletimo na zelo visoke koncentracije Rn v zraku, zato je potrebno pred daljšim zadrževanjem ali delom v jami predhodno izmeriti radon, delo v jami načrtovati in, če je potrebno, časovno omejiti; (3) v podzemnih prostorih vodnih zajetij lahko naletimo na povišane koncentracije Rn v zraku, ker pa je tu zadrževalni čas delavcev kratek, so dobljene efektivne doze nizke; vendar je potrebno daljša vzdrževalna dela načrtovati in, če je potrebno, omejiti delovni čas; (4) pri normalnem delovnem režimu so v vinskih kleteh koncentracije Rn v zraku nizke; previdnost je potrebna samo pri izvajanju del ob izključenem prezračevanju; (5) na večini delovnih mest v kletnih prostorih bolnišnic je izpostavljenost radonu zadovoljivo nizka – previdnost je potrebna le pri starejših zgradbah, v

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katerih morda tla niso bila kakovostno izvedena ali pa je prišlo z leti do poškodb talne plošče, s čimer se je radonu olajšal dostop v prostor.

Klju~ne besede: radon, radonovi kratko'ivi razpadni produkti, podzemna delovna mesta, efektivne doze

1 Introduction

Recent epidemiological studies in homes indicate that in Europe (1) the radioactive noble gas radon (^{222}Rn) or Rn) together with its short-lived decay products (RnDP) accounts for about 9 % of deaths due to lung cancer and about 2 % of all deaths due to cancer. It originates in rocks and soils by radioactive decay of radium (226 Ra) in the uranium (238 U) decay chain (2). Because uranium is widely distributed, though at low levels, all over the earth's crust, radon is present everywhere in the environment, the highest activity concentration being in soil gas, i. e., from several hundred kilo Bq m^{-3} to several mega Bq m^{-3} (activity of 1 Bq equals 1 disintegration per second). Radon emanates from its place of origin in a mineral grain, into soil gas or water present in the void space. Then, dissolved in either a carrier geogas or thermal-mineral water and driven by advection, it moves towards the surface and eventually reaches the atmosphere (3). There, it is rapidly diluted and its resulting concentration in the outdoor air is low, in the range of 5–50 Bq m⁻³. On the other hand, it accumulates in closed underground places (karst caves, mines, tunnels), as well as in basements and ground floors of the living and working environment. In the indoor air of a building, its concentration depends on the geology and pedology of the site, shape, size and quality (less often building material) of the structure, and living habits of residents and the working regime (4). Because of its decay, Rn (α -decay, half-life, $t_{1/2} = 3.82$ days) is always accompanied by its short-lived decay products (RnDP): ²¹⁸Po (α-decay, $t_{1/2} = 3.05$ min), ²¹⁴Pb (β / γ-decay, $t_{1/2} = 26.8$ min), ²¹⁴Bi (β/γ-decay, $t_{1/2} = 19.7$ min) and $\frac{1}{2}$ ¹⁴Po (α -decay, $t_{1/2}$ = 164 μs). RnDPs are present attached to aerosols or as unattached nanosize clusters. Although theoretically possible, radioactive equilibrium between Rn and RnDP is never reached in the actual environment and activity concentrations of RnDP are lower than that of Rn, as described by the equilibrium factor F, which has a value between 0.40 and 0.60 in indoor air.

While extensive surveys of radon in indoor air in dwellings and workplaces have been carried out in most developed countries, much less attention has been paid to underground rooms, despite the fact that elevated Rn levels are expected here. However, underground places have not been ignored in Slovenia. In a nationwide indoor radon programme, radon in air has been monitored in 730 kindergartens (5), 890 schools (6) and 1000 randomly selected homes (7) together with a large number of underground workplaces, where elevated radon levels can be expected.

In this paper, we reported radon levels in workplaces in Slovenian non-uranium underground mines, karst caves, water supply plants, wineries and hospitals. The effective doses estimated for the employees are discussed.

2 Experimental

2.1 Survey methods

Different instruments have been used to measure radon concentration in air, the choice depending on the purpose of measurement.

- To obtain instantaneous Rn concentration, different size alpha scintillation cells (8) were used. The cells were calibrated with a standard $^{226}\mathsf{RaCl}_{2}$ solution (National Institute of Standards and Technology (NIST Standard Reference Material 4953D), according to the Rushing procedure (9, 10). Using the same procedure, cells were checked monthly. Cell efficiencies lie around 1.4 \times 10⁻³ s⁻¹ Bq⁻¹ m³, which gives a lower limit of detection of $10-30$ Bq m⁻³ at a $1-2$ min⁻¹ background and 30 minutes counting time. Air was sampled directly into a cell, which was transported to the laboratory, where gross alpha radiation was counted in PRM 145 counters (AMES, Slovenia) three hours after sampling, when equilibrium between radon and its short-lived decay products was reached.
- Average radon concentration was obtained by exposing etched-track detectors provided by various manufacturers: Forschungszentrum Karlsruhe (Germany), Radonlab (Oslo, Norway), and National Institute of Radiological Sciences (Chiba, Japan). After 1 to 3 months exposure, detectors were

mailed back to the manufacturer for development and evaluation of the results. At these exposure times the lower limit of detection was 3 to 5 Bq $m⁻³$. The detectors were calibrated by the manufacturers.

- An AlphaGuard PQ2000 multiparameter radon monitor (Genitron, Germany) was used to measure radon concentration, air temperature and barometric pressure continuously, with a frequency of one per hour over a period of 5–20 days in order to determine diurnal fluctuations of Rn concentration. The lower limit of detection was 50 to 100 Bq m–3. The instrument was calibrated in the manufacturer's radon chamber before delivery.
- EQF 3020 and EQF 3020-2 radon and radon progeny monitor systems (Sarad, Germany) were used to measure continuously (with a frequency of once every two hours) concentrations of Rn and RnDP, equilibrium factor between Rn and RnDP and unattached fraction (f_{un}) of RnDP together with air temperature and humidity, over a period of 5 to 20 days in order to determine diurnal fluctuations of these parameters. The lower limit of detection was 30 to 80 Bq m^{-3} . The instruments were calibrated before delivery and recalibrated every two years in the manufacturer's radon chamber.

In order to comply with the quality assurance - quality control recommendations, the devices were checked regularly at the inter-comparison experiments organized annually by the Slovenian Nuclear Safety Administration (11), and at each site, radon concentration was also measured with alpha scintillation cells calibrated with a NIST standard as described above. Results obtained with the cell and the other devices agree within experimental errors.

2.2 Measurement protocol

The survey of radon at underground workplaces was designed and performed with assistance from the Radiation Protection Administration at the Ministry of Health, except for the first measurements in underground mines and karst caves. Places to be monitored were selected jointly, taking into account the elevated radon levels previously found, the radon potential based on geology and our previous experience in indoor radon levels. Prior to our survey, the management of each place was provided with general information about the radon problem at underground workplaces, and the programme of our study was ex-

plained. Underground rooms were selected jointly with the management, giving priority to those attended by larger numbers of persons for longer times. Air was sampled by alpha scintillation cells to obtain a quick and rough estimate of radon levels. Then, at the 2 or 3 points with the highest radon levels, etched track detectors were exposed for 1–3 months. In addition, at representative places, concentrations Rn (C_{p}) and RnDP (C_{RnDP}), as well as the equilibrium factor (F) and unattached fraction of RnDP (f_{un}) , were recorded continuously for 5–20 days.

3 Results

3.1 Underground non-uranium mines

High radon levels have been found in air of uranium mines and also of other underground workings, especially metal and coal mines (12, 13). Rn concentrations in the air were measured between 1978 and 1986 in the following Slovene mines: Mežica lead-zinc mine, Idrija mercury mine and Velenje-Preloge, Trbovlje, Zagorje, Hrastnik, Laško and Senovo coal mines (14). In total, about a hundred samples were taken with alpha scintillation cells. At several sites in the Mežica and Idrija mines Rn concentrations exceeded the Slovene national limit of 1000 Bq m^{-3} (15). At the time of our survey, both mines started to be shut down and the investigation was therefore not continued. Rn concentrations in all coal mines however were low, never exceeding 500 Bq m–3.

3.2 Karst caves

In 1984 and 1985, radon in air was measured with alpha scintillation cells at about five hundred sites in more than fifty Slovene caves (16), where elevated Rn levels were expected (17). Measurable Rn concentrations ranged from 2 to 6 kBq m^{-3} , although in many caves it was below the detection limit. At one site in the Postojna Cave off the tourist guided route, it was as high as 22 kBq m^{-3} (18). A further study in this cave, lasting for several years and in which various complementary measuring devices were used to obtain C_{Rn} , F, f_{un} , C_{RnDP} , as well as meteorological parameters, showed (19) that radon concentrations may reach $4-6$ kBq m⁻³ in summer. The diurnal variation of the measured parameters in summer is shown in Fig. 1. Rn and RnDP levels were lower in wintertime by a factor of about 2.

Figure 1. Radioactivity measurements at the lowest point in Postojna Cave, June 30 - July 8, 1999. Concentrations of Rn ($C_{_{\sf Rn}}$) and equilibrium factor (F); concentrations of RnDP ($C_{_{\sf RnDP}}$) and unattached fraction of RnDP (f_{un}); relative air humidity in the cave (RH) and barometric pressure (P); air temperature outdoor (T_{out}) and in the cave (T_{in}).

3.3 Water supply plants

Elevated Rn levels may also be found in air at workplaces in water supply plants (20). In 2001, Rn, RnDP and F were monitored at workplaces in 53 underground premises of water plants in Ljubljana, Grosuplje, Kočevje, Maribor, Koper, Ilirska Bistrica, Nova Gorica, Postojna, Sežana and Metlika (21). Both instantaneous and monthly average radon concentrations were found to be relatively low, exceeding 1000 Bq m^{-3} only at 4 places.

3.4 Wineries

Although wine production is an important national industry in many countries reports on radon in air at workplaces in their underground facilities are sparse (21). We focused our attention on this environment in 2002. Rn and RnDP concentrations, F and f_{un} were measured in 22 underground rooms of the larger wineries in the following cities/towns: Sežana, Koper, Vipava, Ptuj, Ormo', Ljutomer, Gornja Radgona and Maribor (23). While 1–2 month average Rn concentrations obtained with etched track detectors were below 150 Bq m^{-3} in all the rooms surveyed, alpha scintillation cells showed an instantaneous value above 1000 Bq m–3 in one room of an old winery. The reason for this disagreement is evident from Fig. 2, which shows the diurnal variation of Rn concentration. In the periods June 6–7 and 27– 28, the ventilation system was shut down, resulting in high Rn levels. The air sample was taken on June 6, when Rn concentration was highest, while etched track detector showed the average, lower, value. Time runs of the parameters monitored under the normal working regime of a new winery are shown in Fig. 3.

3.5 Hospitals

In many hospitals there are laboratories, shops, kitchens and other facilities in basements where elevated Rn levels may be expected (24, 25). In 2002, a Rn survey was carried out in hospitals in the following cities/towns: Ankaran, Begunje, Brežice, Celje, Golnik, Idrija, Izola, Jesenice, Kranj, Ljubljana, Maribor, Murska Sobota, Nova Gorica, Novo mesto, Ormož, Postojna, Ptuj, Sežana, Slovenj Gradec, Šentvid pri Stični, Topolšica, Trbovlje and Vojnik (26). In each hospital, two to four etched track detectors were exposed. In total, 207 air samples were taken from 186 rooms, and 215 etched track detectors were exposed in 198 rooms. In addition, in 12 rooms of 9 hospitals, concentrations of Rn and RnDP as well as F and f_{un} were recorded continuously for periods of 5 to 11 days.

Monthly average Rn concentrations obtained with etched track detectors were below 100 Bq m⁻³ in more than 70 % of places (Table 1) and only in 7 rooms were they above 400 Bq m^{-3} (Table 2).

Diurnal variations of Rn and RnDP concentrations in two shops with elevated levels are presented in Fig. 4. Variations were very small in the first (Fig. 4a), fluctuating by about ± 50 % around the mean value, while they were much more pronounced in the second (Fig. 4b), radon concentrations ranging from several hundred Bq m^{-3} in the morning up to several thousand Bq m⁻³ during the night. Surprisingly, both shops are on the ground floor and not in a basement. While a constantly high concentration is seen for the weekend of October 18–21 (Fig. 4b) this is not so clear for the weekend of September 27–30 (Fig. 4a). Permanent ventilation in the first shop keeps radon concentrations almost con-

Figure 2. Continuous measurement of concentrations of Rn (C_{Rn}) and RnDP (C_{RnDP}) in an old winery, using the Sarad EQF3020-2 instrument between June 6 and July 3, 2002.

stant, while the second shop is only ventilated naturally by opening doors and windows, and hence a typical diurnal variation of radon concentration occurs (27).

4 Discussion

To relate concentrations of Rn or RnDP to health, the dose conversion factor (DCF) is needed, defined as the ratio of the weighted equivalent dose to the lung (expressed in mSv) to the exposure to RnDP (expressed either in WLM, if RnDP activity concentration in air is known, or Bq m^{-3} h, if Rn activity concentration in air is known). The old but still widely used unit, 1 WLM (working-level-month), is the exposure resulting from 170 hours breathing air with an activity concentration of short-lived radon decay products of 1 WL (working-level). 1 WL was originally defined as the activity concentrations of RnDP which are in radioactive equilibrium $(F = 1)$ with 100 pCi L^{-1} (3700 Bq m⁻³) of ²²²Rn, resulting in a potential alpha energy concentration of 1.3×10⁵ MeV L⁻¹ (2). DCF values have been obtained from epidemiological studies on uranium miners. At present, the International Commission for Radiological Protection (ICRP) in Publication 65 (28) recommends 5 mSv WLM–1 for working and 4 mSv WLM–1 for living environments.

Figure 3. Continuous measurement of concentrations of Rn (C_{Rn}) and RnDP (C_{RnDP}), equilibrium factor (F) and unattached fraction of RnDP (f_{un}) in a new winery, using the Sarad EQF3020-2 instrument in the period from May 30 to June 14, 2002.

Figure 4. Continuous measurement of Rn and RnDP concentrations using the Sarad EQF3020-2 instrument: a) in the pharmacy shop of hospital 01-ID-PB between September 24 and October 2, 2002; b) in the pharmacy shop of hospital 07-NM-SB between October 14 and 25, 2002.

Table 2. Hospitals (coded names) with indoor air radon concentrations higher than 400 Bq m⁻³. Shown are: number of persons working in the rooms surveyed and their annual exposure times, monthly average radon concentrations (C_{Rn}) obtained with etched track detectors in the periods indicated (in 2002), and monthly effective doses (E) (mo stands for month), received by the personnel in the period from 23 September to 22 October.

Based on radon concentrations measured at underground workplaces, effective doses (E) received by a worker at these places were calculated applying the general formula (2):

$$
E = (C_{\text{Rn}} \times F) / 3700 \times (t/170) \times DCF.
$$
 (4)

Here, C_{Rn} is the radon concentration (in Bq m⁻³) and t is the time (in hours) spent by workers at this workplace. According to ICRP-65 (28), $F = 0.40$ and DCF = 5 mSv WLM⁻¹.

4.1 Underground non-uranium mines

Rn levels in coal mines are low, as the result of the effective ventilation needed to prevent methane explosions. Therefore no precautionary measures are needed from the radiation protection point of view. Effective doses have not been published.

4.2 Karst caves

Radon concentrations in karst caves are higher in summer than in winter due to the so-called "chimney effect". For example, air temperature in the Postojna Cave is between 8.0 and 9.0 °C and is practically constant all the year round. In winter, the air temperature in the cave is higher than outdoors and the cave works as a huge fire place in which the draught drives air from the cave rooms into the atmosphere.

Because of the high radon levels in the Postojna Cave, in 1995 regular and permanent Rn monitoring was required by the Radiation Protection Administration, and has been carried out ever since. The Cave management reports the effective doses for their workers in the cave for the first half of every year and for the whole year. If the effective dose for a worker for the first half of the year exceeds 2.0 mSv, that worker would spend a reduced time in the cave in the second half of the year. Based on the annual effective doses, the time to be spent the following year by workers in the cave is planned. The effective dose received by a tourist during one visit is negligible.

4.3 Water supply plants

The attendance time of workers in underground premises of the water plants is short and hence despite the elevated Rn levels, the resulting annual effective doses were acceptably low: they never exceeded 3 mSv (Fig. 5). Therefore under the present working regime no mitigation measures are needed. Nonetheless, the managements were recommended to prepare a time plan prior to every maintenance work underground, which should be accompanied by Rn monitoring and, if necessary, time limited for a given worker.

4.4 Wineries

Based on Rn concentrations obtained with etched track detectors, annual effective doses for the workers in underground rooms of wineries were estimated to range from 0.13 to 1.75 mSv. The latter value slightly exceeds 1.25 mSv, the effective dose a member of the general public receives from Rn and RnDP in a year on a worldwide average (29). Under the present operational regime in Slovene wineries no precautions from the radiation protection point of view are necessary for

Figure 5. Annual effective doses of workers in underground facilities in Slovene water supply plants in 2001 (PI – plant identity number).

workers and no mitigation measures are foreseen. We have only drawn the attention of the managements to the crucial role of ventilation in keeping low Rn levels, and not to allow works, or at least to limit working times, at underground workplaces during periods when fans are shutdown.

4.5 Hospitals

The reason for elevated indoor Rn levels in hospitals was always that the floor not properly made: either the concrete slab was of bad quality, with cracks appearing with age, or wooden boards were fixed on wooden beams laid directly on a gravel ground.

Annual effective doses were calculated for 1025 persons working in the rooms surveyed. 996 persons (94.2 %) received less than 1 mSv in a year and the remaining 59, between 1.1 and 7.3 mSv. For 16 of the latter, working in rooms with radon concentration more than 400 Bq m^{-3} , the monthly effective doses are shown in Table 2. All the rooms with annual effective doses more than 2 mSv were further investigated in order to obtain reliable data on which to decide whether mitigation measures should be undertaken. Some rooms have been already successfully mitigated and the others are in the process of mitigation.

5 Conclusions

This review of the Rn survey carried out at underground workplaces in Slovenia over the last two decades, high-

lights the following main points: (i) because of effective ventilation, no concern is necessary for miners' exposure to radon in coal mines; (ii) prior to a longer stay in a karst cave, Rn level should be checked and, if necessary, the stay in the cave should be limited – as an example: based on the semi-annual and annual effective doses of the workers in the Postojna Cave, their stay in the cave is planned ahead and working time in the cave is limited; (iii) elevated Rn levels are frequently found in underground premises of water supply plants but the effective doses received by the workers are low because of short attendance times; however, maintenance work underground should be accompanied by Rn monitoring, and the management should plan the time needed for each worker, in order to avoid too high exposure; (iv) under normal working regime in a winery, exposure of workers to Rn in underground facilities is low; care is necessary only when work is to be carried out during ventilation shut down; (v) in the majority of basement rooms in hospitals the exposure of personnel to Rn is acceptably low, nevertheless, the occupation safety officer should pay attention to rooms in old buildings where the floor is not always a sufficient barrier to Rn entry, leading to the possibility of elevated indoor Rn levels.

While radon levels cannot be reduced in karst caves because the natural environment should be preserved, at all other places Rn problem can be successfully mitigated by undertaking appropriate technical measures.

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Contributors:

Ivan Kobal participated in field measurements and data evaluation for underground mines and karst caves, while Janja Vaupotič, as the head of the Radon Center at the Jožef Stefan Institute, designed the programme and, together with her co-workers, ran the measurements in the Postojna Cave, water supply plants, wineries and hospitals, evaluated the data obtained and prepared the paper jointly with Ivan Kobal.

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List of abbreviations:

- ICRP International Commission on Radiological Protection
- Rn 222Rn isotope of radon
- RnDP short-lived decay products of ²²²Rn: ²¹⁸Po, ²¹⁴Pb, 214Bi and 214Po

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