



Radon decay products and 10–1100 nm aerosol particles in Postojna Cave

M. Bezek, A. Gregorič, and J. Vaupotič

Department of Environmental Sciences, Jožef Stefan Institute, Ljubljana, Slovenia

Correspondence to: M. Bezek (mateja.bezek@ijs.si)

Received: 28 September 2012 – Published in Nat. Hazards Earth Syst. Sci. Discuss.: –

Revised: 31 January 2013 – Accepted: 15 February 2013 – Published: 28 March 2013

Abstract. At the lowest point along the tourist route in Postojna Cave, the activity concentration of radon (^{222}Rn) decay products and the number concentration and size distribution of aerosol particles in the size range of 10–1100 nm were monitored, with the focus on the unattached fraction (f^{un}) of radon decay products (RnDPs), a key parameter in radon dosimetry. The total number concentration of aerosols during visits in summer was lower (700 cm^{-3}) than in winter (2800 cm^{-3}), and was dominated by $< 50\text{ nm}$ particles (related to unattached RnDPs) in summer and by $> 50\text{ nm}$ particles (related to the attached RnDPs) in winter. This explains the higher f^{un} values in summer (0.75) and the lower winter measurement (0.04) and, consequently, DCF_D values of 43.6 and 13.1 mSv WLM^{-1} respectively for the calculated dose conversion factors. The difference is caused by an enhanced inflow of fresh outside air, driven in winter by the higher air temperature in the cave compared to outside, resulting in the introduction of outside aerosol particles into the cave.

1 Introduction

In a great number of karst caves worldwide, elevated radon (^{222}Rn) levels in the air have been observed (Cigna, 2005; Field, 2007; Vaupotič, 2010). Radon α -transformation (half-life $t_{1/2} = 3.82\text{ days}$) is followed by a radioactive chain of its short-lived radon decay products (RnDPs) (Nero, 1988): ^{218}Po (α , 3.05 min) \rightarrow ^{214}Pb (β/γ , 26.8 min) \rightarrow ^{214}Bi (β/γ , 19.7 min) \rightarrow ^{214}Po (α , 164 μs). Initially, they appear mostly as positive ions (Chu and Hopke, 1988; Pagelkopf and Porstendörfer, 2003; Porstendörfer and Reineking, 1992), which react with molecules of trace gases and vapours

(mostly water) in the air and form small clusters, which are then neutralised (Chu and Hopke, 1988; Pagelkopf and Porstendörfer, 2003) and attach to the background aerosol particles (Dankelmann et al., 2001; Hopke, 1996; Pagelkopf and Porstendörfer, 2003; Papastefanou, 2008). Eventually, RnDP aerosols mostly show a bimodal size distribution one in the 1–10 nm size range (referred to as unattached RnDPs) and the other between 100 and 500 nm (attached RnDPs) (Gründel and Porstendörfer, 2004; Porstendörfer and Reineking, 1992; Porstendörfer et al., 2000). Total concentration of RnDP in the air is reported as equilibrium-equivalent activity concentration ($C_{\text{RnDP}}^{\text{A}}$, Bq m^{-3}) expressed by the following (Nero, 1988):

$$C_{\text{RnDP}}^{\text{A}} = 0.1065 C_{^{218}\text{Po}}^{\text{A}} + 0.515 C_{^{214}\text{Pb}}^{\text{A}} + 0.379 C_{^{214}\text{Bi}}^{\text{A}}, \quad (1)$$

where C^{A} (Bq m^{-3}) stands for the individual activity concentrations of ^{218}Po , ^{214}Pb and ^{214}Bi . Because of its short half-life, ^{214}Po activity is equal to the activity of ^{214}Bi and is therefore already included in the last term of Eq. (1). Due to air movement and deposition of RnDP to surfaces, individual activity concentrations of ^{218}Po , ^{214}Pb and ^{214}Bi are always lower than that of radon (C_{Rn}^{A}), and secular equilibrium between radon and RnDP is never reached in the ambient air, its degree being expressed by the equilibrium factor (F):

$$F = C_{\text{RnDP}}^{\text{A}} / C_{\text{Rn}}^{\text{A}}. \quad (2)$$

On the world average, RnDPs contribute more than half to the effective dose (the radon contribution is minor) received by members of the general public from all natural radioactivity (UNSCEAR, 2000) and are a significant cause of lung cancer, second only to cigarette smoking (Darby et al., 2005). For this reason their levels in the living and working environment, including karst caves, are of great concern to us and are

a research challenge. While the exposure of tourists is negligible during their visit to a cave, it may easily reach high levels for cave employees who spend the majority of their working time underground (Field, 2007; Kávási et al., 2003; Somlai et al., 2009; Vaupotič et al., 2001).

For the general purposes of radiation protection, the International Commission on Radiological Protection (ICRP), in its Publication 65, recommends for radon dosimetry a dose conversion factor (DCF_E) of 4 mSv WLM^{-1} at home and 5 mSv WLM^{-1} in the workplace (ICRP, 1993), as the values deduced from epidemiological studies. Nevertheless, dose conversion factors are subject to constant investigation and upgrading, and an increase of their recommended values is foreseen in the near future (Harrison and Marsh, 2012; ICRP, 2010). A value of 1 WLM (working-level month) corresponds to the exposure from 170 h of breathing air in which the potential α -energy concentration of RnDP ($E_{\alpha\text{RnDP}}$) is $1.3 \times 10^8 \text{ MeV m}^{-3}$. $E_{\alpha\text{RnDP}}$ (MeV m^{-3}) is expressed through the activity concentrations (C^A , Bq m^{-3}) of ^{218}Po , ^{214}Pb and ^{214}Bi (Nero, 1988):

$$E_{\alpha\text{RnDP}} = 3690 C_{^{218}\text{Po}}^A + 17830 C_{^{214}\text{Pb}}^A + 13120 C_{^{214}\text{Bi}}^A. \quad (3)$$

On the other hand, Birchall and James (1994) and Marsh et al. (2002) elaborated on a dosimetric approach to calculating the dose conversion factor (DCF_D), which is used mainly for research purposes. They also showed that the parameter most affecting DCF_D is the fraction (f^{un}) of unattached RnDPs, defined as follows (Nero, 1988):

$$f^{\text{un}} = \frac{C_{\text{RnDP}}^{\text{Aun}}}{C_{\text{RnDP}}^A}, \quad (4)$$

where $C_{\text{RnDP}}^{\text{Aun}}$ is the equilibrium-equivalent concentration of unattached RnDPs, obtained if activity concentrations of only the unattached ^{218}Po , ^{214}Pb and ^{214}Bi are taken into Eq. (1). Furthermore, they expressed DCF_D based on f^{un} with an empirical formula:

$$DCF_D = 11.35 + 43 f^{\text{un}}. \quad (5)$$

In Postojna Cave, radon and RnDPs have been monitored systematically since 1995 in order to estimate the radiation doses of personnel and keep them below an acceptably low level (Vaupotič et al., 2001). In addition to elevated radon levels, f^{un} values are also higher in the cave – ranging from 0.40 to 0.80 in summer and from 0.10 to 0.20 in winter (Vaupotič, 2008a) – than in kindergartens (Vaupotič, 2007), schools (Vaupotič and Kobal, 2006) and wineries (Vaupotič, 2008b). As in the Carlsbad Caverns (Cheng et al., 1997) and in an experimental room (Meisenberg and Tschiersch, 2009), high f^{un} values were assumed to result from low aerosol concentration in the cave (Butterweck et al., 1992). The activity size distribution of RnDPs in Postojna Cave was found to be bimodal, with the unattached fraction in the range 1.3–6.0 nm and the attached fraction in the range 119–289 nm (Butterweck et al., 1992).

In order to reveal how levels of unattached and attached RnDPs (a crucial parameter in radon dosimetry) (Hofmann and Koblinger, 1990; Nikezić et al., 2006) are controlled by the presence of background aerosol particles with which RnDPs are associated, measurement of number concentration and size distribution of aerosol particles in the 10–1100 nm size range has recently been introduced (Iskra et al., 2010).

Background aerosols are formed in the atmosphere by various natural processes (e.g. rock grinding and crushing, volcanic eruptions, wildfires) and anthropogenic influences (e.g. biomass and fossil fuel burning, traffic emissions). The concentration of background aerosols in outdoor air depends strongly on emission factors and meteorological conditions (Colbeck, 1998). In general, the highest number concentrations are observed in urban areas during winter, caused by biomass and fossil fuel burning, while in summer the levels are usually lower. In the indoor (living) environment the main sources are human beings and their activities.

Background aerosols were measured in urban and rural outdoor air during our previous work (Smerajec and Vaupotič, 2012). The average total number particle concentration ($C_b^N(\text{tot})$) and geometric mean particle diameter (d_{GM}) obtained in the urban environment was $14\,750 \pm 7220 \text{ cm}^{-3}$ and $46 \pm 10 \text{ nm}$, respectively. In the rural environment $C_b^N(\text{tot})$ of $7800 \pm 5250 \text{ cm}^{-3}$ and d_{GM} of $63 \pm 19 \text{ nm}$ were measured (Smerajec and Vaupotič, 2012).

In the present paper, simultaneous measurements of the activity concentration of radon decay products and the number concentration and size distribution of aerosol particles at the lowest point along the guided route in summer and winter are described, and the results presented with a commentary.

2 Materials and methods

2.1 Site description

Postojna Cave is a part of a larger cave system in Slovenia, which also includes the Pivka, Magdalene, Black and Island caves (Fig. 1) and has a total length of about 20 km. It is one of the biggest show caves in the world. Postojna Cave is a horizontal cave with passages developed on two levels. The Pivka river sinks at the lower entrance to the cave at 511 m a.s.l. (above sea level). The entrance to the main, currently dry passage is situated at 529.5 m a.s.l. (Šebela, 1998). This entrance is used as a tourist entrance. As in the majority of karst caves, the only ventilation is natural air circulation. Air temperature is practically constant at around 10–11 °C all year round, and relative air humidity is in the range 94–100 %. Air flow differs considerably in summer and winter. In winter, when the cave temperature is higher than outside, cave air is released from the cave into the outdoor atmosphere due to the air draught caused by the “chimney effect”, thus allowing fresh and cold outside air to enter the cave through

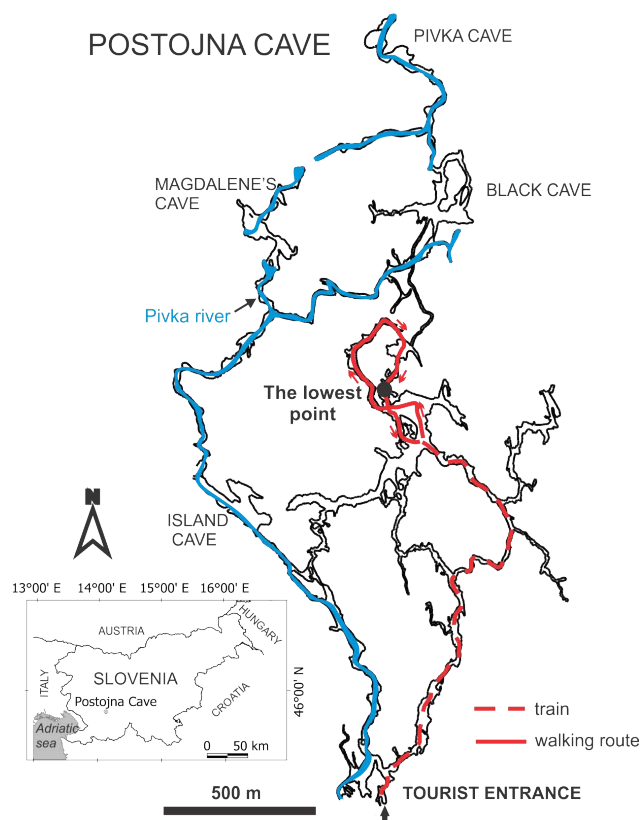


Fig. 1. Map of the Postojna Cave system showing the cave railway, walking route and the lowest point (our measurement site).

low-lying openings (Gregorič et al., 2011). This effect is not produced in summer, when the outside temperature is higher than in the cave and the air draught is minimal or reversed. In any case, the air speed is low, never exceeding 1.5 m s^{-1} along the main entrance corridor, and is much less or zero in other parts of the cave. As a result of the different natural ventilation regime, the cave interior, at least along the main air paths, is less wet in winter than in summer.

The cave is open to visitors every day from 10:00 a.m. to 04:00 p.m. LT (local time) in winter and from 09:00 a.m. to 06:00 p.m. LT in other seasons. The daily number of visitors is around 3500 in summer and from 50 to several hundred in winter, totalling around half a million a year. Visitors ride an electric train for the first 2 km from the entrance to the train stop in the cave, walk 1.8 km on a figure-of-eight route and return back to the train in about an hour and a half. Our study site is located at the lowest point of the path (508 m a.s.l., Fig. 1), approximately 500 m on from the train stop along the guided route. This is also the point of regular radon monitoring for the purpose of radon dosimetry of the cave employees. The path is paved with a special concrete containing silica sand to prevent it from becoming slippery when wet.

The major source of particulate matter in the cave air is the inflow of fresh outside air in winter. Deposition of dust

is observed on surfaces in the main corridor all the way from the entrance to the train stop, but no further. Another source is the railway. Particles originate from the rusting of iron parts, grinding of sand, and rotting and damage to wooden sleepers. They are lifted and resuspended by the air draught caused by the train running at a speed of 1.6 m s^{-1} through the narrow corridors and galleries. Human activity, both visits and maintenance work on the cave infrastructure, is also a potential source of particulate matter.

2.2 Radon decay products

Individual activity concentrations (C^A , Bq m^{-3}) of ^{222}Rn , ^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po have been measured using the EQF3020-2 device (Equilibrium Factor Monitor System, Sarad, Germany). Air is pumped for 6 min at a flow rate of $2.4 \text{ dm}^3 \text{ min}^{-1}$ over a metal mesh grid on which aerosol particles smaller than 5 nm (considered as unattached RnDPs) are separated from those above this size (considered as attached RnDPs), and the two fractions are deposited electrostatically on two separate 150 mm^2 semiconductor detectors. Gross α activity is measured over three consecutive intervals within 110 min of the end of pumping, and, applying the Markov method (Markov et al., 1962; Streil et al., 1996), individual activity concentrations of ^{218}Po , ^{214}Pb and ^{214}Bi in the unattached and attached fractions are obtained. The device also gives the equilibrium-equivalent activity concentration of RnDPs (C_{RnDP}^A), the equilibrium factor between Rn and RnDPs (F), the fraction of unattached RnDPs (f^{un}) and potential α -energy concentration of RnDPs ($E_{\alpha\text{RnDP}}$), as well as air temperature and relative humidity.

In order to facilitate comparison of the number concentration of background aerosols and concentrations of ^{218}Po , ^{214}Pb and ^{214}Bi aerosols, the activity concentrations (C^A) of radionuclides are converted into their number concentrations (C^N), by applying the radioactivity law equation:

$$C^A = \lambda \times C^N, \quad (6)$$

with

$$\lambda = \ln 2 / t_{1/2}. \quad (7)$$

The obtained number concentrations of ^{218}Po , ^{214}Pb and ^{214}Bi atoms (cm^{-3}) are denoted by, respectively, $C_{^{218}\text{Po}}^{\text{Nun}}$, $C_{^{214}\text{Pb}}^{\text{Nun}}$ and $C_{^{214}\text{Bi}}^{\text{Nun}}$ for unattached RnDPs and $C_{^{218}\text{Po}}^{\text{Natt}}$, $C_{^{214}\text{Pb}}^{\text{Natt}}$ and $C_{^{214}\text{Bi}}^{\text{Natt}}$ for attached RnDPs.

2.3 Background aerosols

The term “background” is used here to comprise all particles in the cave air, both with and without associated RnDPs (although the contribution of the latter in the number concentration is negligible, as will be seen later).

The number concentration and size distribution of background aerosol particles were measured with a SMPS+C

instrument (scanning mobility particle sizer + counter), Series 5.400 (Grimm, Germany). For this purpose, the long DMA (differential mobility analyzer) unit, designed for the 10–1100 nm size range, was used. The DMA unit separates charged particles based on their electrical mobility, which depends on the particle size and electrical charge: the smaller the particle and the higher its electrical charge, the higher its mobility. Particles enter the CPC (condensation particle counter) unit containing a heater saturator in which alcohol vapour molecules condense onto the entering particles, thus causing them to grow into droplets. The droplets are then detected with a laser beam (dynamic light scattering detection) and counted. The frequency of measurement is once in 7 min. The instrument gives the total number concentration C_b^N (tot) and size distribution $dC_b^N(d)/d \ln d$ (with d the electrical mobility equivalent particle diameter). Because the DMA unit is not designed for sizes below 10 nm, and bearing in mind that RnDPs in Postojna Cave are attached to aerosol particles bigger than 100 nm (Butterweck et al., 1992), we selected 50 nm as the boundary between unattached and attached RnDPs. Thus, we are interested in concentrations of particles smaller than 50 nm ($C_b^N(< 50)$), those bigger than 50 nm ($C_b^N(> 50)$) and the fraction of the smaller particles, defined as

$$x_b = C_b^N(< 50)/C_b^N(\text{tot}). \quad (8)$$

Several 5–10 day measurements were carried out with EQF devices in summer 2009 and winter 2010 at the lowest point along the guided walking route in the cave. Within these periods, the SMPS+C instrument was also used, but only for a few hours during morning visits, because the instrument is not designed for such high air humidity and its operation was therefore minimised.

3 Results and discussion

3.1 Previous results

Previous measurements (Vaupotič, 2008a; Vaupotič et al., 2001) indicated that the difference in air temperature outside and inside the cave played a dominant role in governing both diurnal and seasonal variations of the environmental conditions in the cave. Radon levels were lowest in winter, when the cave temperature is higher than outdoors, because radon-rich air is released from the cave into the outdoor atmosphere and fresh outdoor air with low radon concentration is driven into the cave. This effect is not produced in summer, when the outdoor air temperature is higher than in the cave, and the resulting radon levels in the cave air are higher.

As an example, at the lowest point the following average values were obtained in 1999 (Vaupotič, 2008a; Vaupotič et al., 2001) in summer: C_{Rn}^A of $4540 \pm 600 \text{ Bq m}^{-3}$, C_{RnDP}^A of $1580 \pm 280 \text{ Bq m}^{-3}$, F of 0.35 ± 0.09 and f^{un} of 0.60 ± 0.16 , and in winter: C_{Rn}^A of $2070 \pm 1160 \text{ Bq m}^{-3}$,

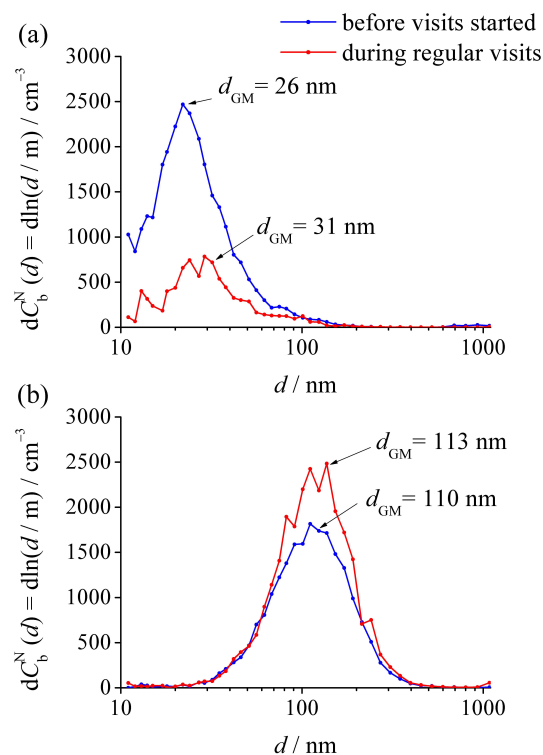


Fig. 2. Size distribution ($dC_b^N(d) = d \ln(d/m)$) of background aerosols at the lowest point in Postojna Cave (a) in summer 2009 and (b) in winter 2010: before visits started and during regular visits.

C_{RnDP}^A of $1130 \pm 530 \text{ Bq m}^{-3}$, F of 0.58 ± 0.11 and f^{un} of 0.14 ± 0.08 .

3.2 Present summer results

For one of the measurements carried out in summer, Fig. 2a shows size distribution of background aerosols before visits started and during regular visits. A low-size peak of bimodal distribution (Butterweck et al., 1992) was not observed, because our instrument is designed for the 10–1100 nm range. Their geometric mean values of 26 and 31 nm do not differ significantly. The decrease in concentration from 2700 to 700 cm^{-3} is mainly due to the smaller particles as seen in Fig. 3a, showing a constant concentration of $> 50 \text{ nm}$ particles and a steady decrease of $< 50 \text{ nm}$ particles during visits. This is presumably because the smaller particles are preferentially deposited on the cave surfaces (Papastefanou, 2008; Porstendörfer, 1984), caught by clothing (Balcazar et al., 1999; Rovenská et al., 2008) and taken up by the lungs (Hofmann and Koblinger, 1990) of tourists walking through the corridor at the lowest point, with a cross section of about 25 m^2 .

Figure 4a shows radon activity concentration (C_{Rn}^A), the equilibrium factor (F), the equilibrium-equivalent activity concentration of RnDPs (C_{RnDP}^A) and the fraction of

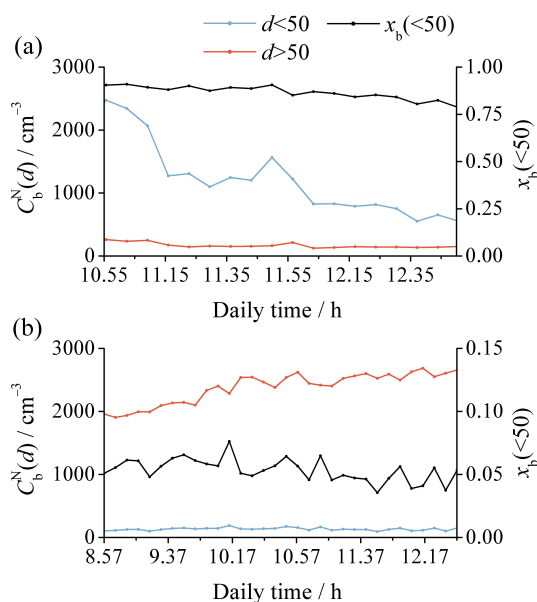


Fig. 3. Time run of number concentrations ($C_b^N(d)$) of background aerosol particles smaller than 50 nm ($C_b^N(<50)$) and bigger than 50 nm ($C_b^N(>50)$), and the fraction of smaller ones ($x_b(<50)$) at the lowest point in Postojna Cave (a) in summer 2009 and (b) in winter 2010.

unattached RnDPs (f^{un}). As expected from previous monitoring in summer (Vaupotič, 2008a), C_{Rn}^A and f^{un} are high and F is low. During the period when background aerosols were also monitored, f^{un} values were around 0.75 (Fig. 4a), resulting in a DCF_D value (Eq. 5) of 43.6 mSv WLM⁻¹. For this time interval, Fig. 5a shows the individual activity concentrations of the unattached and attached RnDPs, and the related number concentrations of the unattached and attached RnDPs, ²¹⁸Po, ²¹⁴Pb and ²¹⁴Bi, respectively. The measurement frequency of the EQF device is much lower (once in 2 h) than that of the Grimm device (once in 7 min), and therefore the time variations of the parameters measured with the two cannot be adequately compared. The number concentration of the unattached ²¹⁸Po atoms is lowest, because it has the shortest half-life (cf. Eq. 6) in comparison to ²¹⁴Pb and ²¹⁴Bi. On the other hand, the ratio of the numbers of the unattached and attached ²¹⁸Po atoms was initially 26, far higher than those of ²¹⁴Pb and ²¹⁴Bi, which were 5.3 and 0.65 respectively. There are several reasons for this high ratio for ²¹⁸Po. Because of its short half-life in comparison to ²¹⁴Pb and ²¹⁴Bi, it has less time available to become attached to atmospheric particles. The second reason is based on the values of diffusion coefficients. These are in the range of 0.024–0.039 cm² s⁻¹ for the charged RnDP species and 0.068–0.085 cm² s⁻¹ for the neutral species (Porstendörfer and Mercer, 1979). At a radon activity concentration of about 5500 Bq m⁻³, as in our case (Fig. 4a), 13 % of ²¹⁸Po species are estimated to be charged (Chu and Hopke, 1988). As a

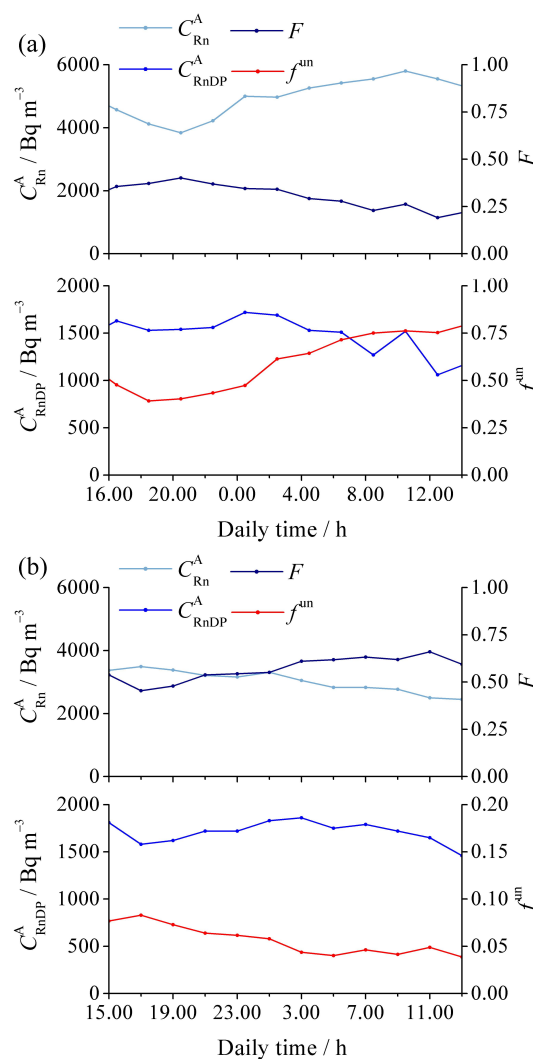


Fig. 4. Results of continuous radon measurement at the lowest point in the Postojna Cave (a) in summer 2009 and (b) in winter 2010: radon activity concentration (C_{Rn}^A), equilibrium factor between radon and its decay products (F), equilibrium-equivalent activity concentration of radon decay products (C_{RnDP}^A) and fraction of unattached decay products (f^{un}).

result of lower mobility, the attachment of these charged species is hindered. This effect cannot be estimated for ²¹⁴Pb and ²¹⁴Bi, because the data are not available. At the end of our measurement, the ratios of numbers of unattached versus attached ²¹⁸Po, ²¹⁴Pb and ²¹⁴Bi atoms were 6.6, 5.0 and 0.86, respectively. The value for ²¹⁸Po decreased drastically, concomitantly with the falling concentration of the < 50 nm particles (Fig. 3a), while the other two did not change significantly.

Concentrations of ²¹⁸Po, ²¹⁴Pb and ²¹⁴Bi atoms are lower by several orders of magnitude than the total concentration of background aerosols. We therefore assume that the reason for high f^{un} values in the cave air is not simply the

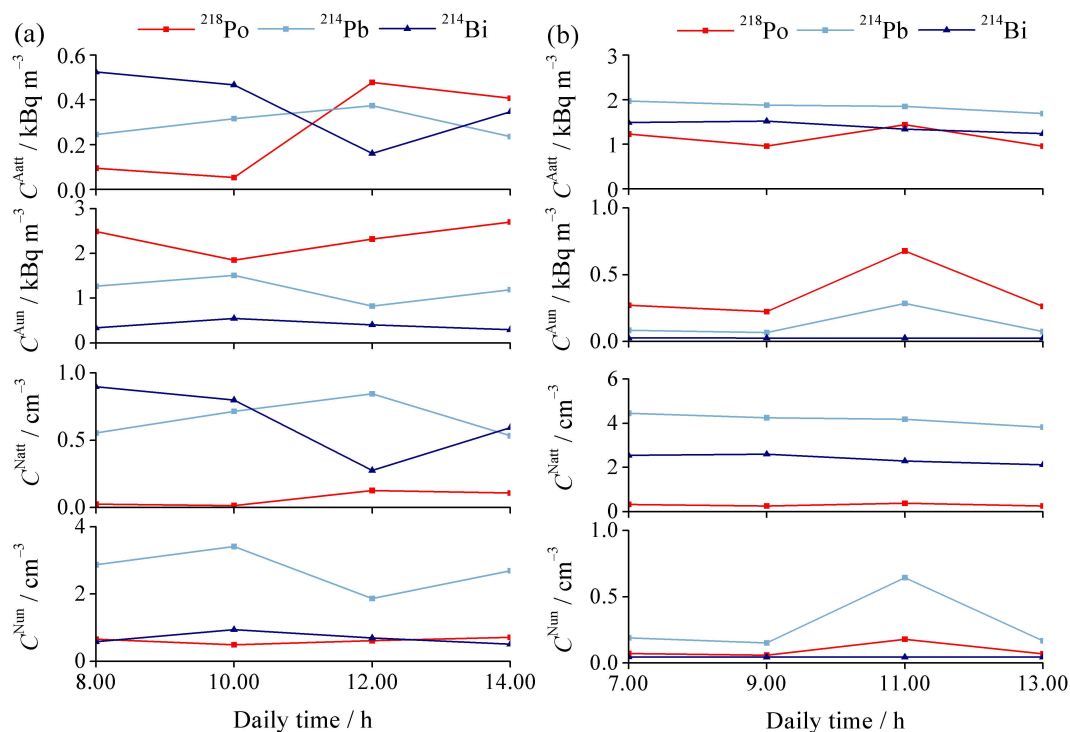


Fig. 5. Activity concentrations and number concentrations of ^{218}Po , ^{214}Pb and ^{214}Bi in the time interval of Fig. 4, when background aerosols were also monitored, from top to bottom: activity concentrations in the unattached form ($C_{^{218}\text{Po}}^{\text{Aun}}$, $C_{^{214}\text{Pb}}^{\text{Aun}}$ and $C_{^{214}\text{Bi}}^{\text{Aun}}$), activity concentrations in the attached form ($C_{^{218}\text{Po}}^{\text{Aatt}}$, $C_{^{214}\text{Pb}}^{\text{Aatt}}$ and $C_{^{214}\text{Bi}}^{\text{Aatt}}$), number concentrations in the unattached form ($C_{^{218}\text{Po}}^{\text{Nun}}$, $C_{^{214}\text{Pb}}^{\text{Nun}}$ and $C_{^{214}\text{Bi}}^{\text{Nun}}$) and number concentrations in the attached form ($C_{^{218}\text{Po}}^{\text{Natt}}$, $C_{^{214}\text{Pb}}^{\text{Natt}}$ and $C_{^{214}\text{Bi}}^{\text{Natt}}$) (a) in summer 2009 and (b) in winter 2010.

low concentration of background aerosols (Butterweck et al., 1992; Cheng et al., 1997; Meisenberg and Tschiersch, 2009), but rather the dominant contribution of their <50 nm particle fraction, with which unattached RnDPs are associated. This assumption is further supported by Fig. 6a: in a volume unit, the total surface area of all < 50 nm particles, with which unattached RnDPs are associated, is from one to eight times lower than that of the bigger particles, but on the other hand the number of smaller ones is four to ten times higher than the number of bigger ones. Thus, the probability of RnDP atoms being associated with smaller particle-clusters is favoured, without even mentioning that the ratio of the number of surface atoms, as adsorption sites, versus the total number of atoms on the smaller particles is substantially higher than on the bigger ones (Oberdörster et al., 2005).

The substantial decrease in x_b (< 50) during morning hours (Fig. 3a) could result in an increase in f^{un} , but it did not, and in fact the opposite was observed (Fig. 4a). Actually, such rapid changes (lasting a matter of minutes) in size distribution are not expected to cause immediate change in f^{un} , i.e. redistribution between unattached and attached Po, Pb and Bi atoms. Rather, they will only influence newly formed Po, Pb and Bi atoms and clusters. Creation of these atoms by radioactive transformations takes

time; also, their neutralisation, clustering, and attachment to (Tokonami, 2000) and detachment from background aerosol particles by recoil are processes with definite values of rate constants. Taking this into account, calculations would show (Nikolopoulos and Vogiannis, 2007) that a time delay of some hours (Wang et al., 2011) necessarily occurs between a change in particle size distribution of background aerosols and a change in f^{un} . Therefore, a true dependence of f^{un} on rapid changes in background aerosols in our case was masked or even totally obscured. A similar situation has been observed in an indoor environment (Smerajec and Vaupotič, 2012).

3.3 Present winter results

For a measurement carried out in winter, Fig. 2b shows the size distribution of background aerosols before visits started and during regular visits. The related geometric mean diameters are similar – 113 and 110 nm respectively – and are substantially higher than in summer (Fig. 2a). Because our measurements of the number size distribution in outdoor air show a geometric mean of 63 ± 19 nm (Smerajec and Vaupotič, 2012), we may assume that in winter the inflow of fresh air brings outside aerosol particles into the cave. In contrast to the situation in summer, the total concentration

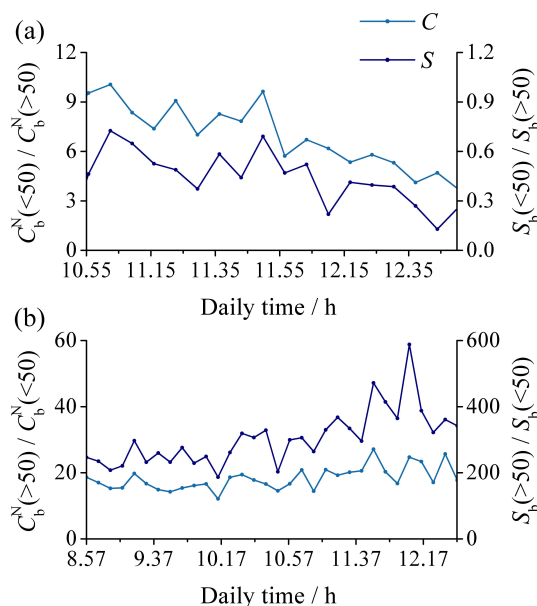


Fig. 6. (a) Ratio of the number concentrations of aerosols smaller than 50 nm ($C_b^N(<50)$) to those of bigger ones ($C_b^N(>50)$), and ratio of the total surface area of all particles smaller than 50 nm ($S_b(<50)$) to the total surface area of all particles bigger than 50 nm ($S_b(>50)$) in a volume unit for the measurement in summer 2009 and (b) ratio of the number concentrations of aerosols larger than 50 nm ($C_b^N(>50)$) to those of smaller ones ($C_b^N(<50)$), and ratio of the total surface area of all particles larger than 50 nm ($S_b(>50)$) to those smaller than 50 nm ($S_b(<50)$) in a volume unit for the measurement in winter 2010.

gradually increases during visits from 2000 to 2800 cm⁻³, with an increase in $x_b(>50)$ and $x_b(<50)$ remaining constant (Fig. 3b). Higher concentration in winter cannot be understood as a result of higher air ionisation due to the radioactivity present (Pashenko and Dublyansky, 1997), because C_{Rn}^A is lower in winter than in summer. The air inflow is assumed to slightly decrease during morning hours due to a decrease in the difference between temperature in the cave and outside. The increase in the total aerosol concentration in morning hours cannot therefore be caused by the inflow of outside air but can only originate from the particles re-suspended and transported by tourists walking on the paved path, which is relatively drier in winter than in summer. As a result of larger particles dominating in winter, $x_b(<50)$ values are substantially lower in winter (Fig. 3b) than in summer (Fig. 3a).

Figure 4b shows radon activity concentration, the equilibrium factor, the equilibrium-equivalent activity concentration of RnDPs and the fraction of unattached RnDPs. As expected from previous measurements in winter (Vaupotič, 2008a), C_{Rn}^A and f^{un} are lower and F is higher than in summer. The cave air in winter is less stagnant than in summer because of the inflow of fresh air, so F values would be expected to be lower than in summer. The opposite is true, however, as

a result of the higher concentration of background aerosols compared to the summer, and higher F is consequently observed in winter. C_{RnDP}^A was shown to be statistically different by t test ($t = 3.43$, $p = 0.0031$) at a significance level of 0.05 (α value). C_{RnDP}^A was higher in winter (1709 Bq m⁻³) than in summer (1480 Bq m⁻³), which is the opposite of the observed pattern of C_{RnDP}^A in previous measurements (Vaupotič, 2008a). This discrepancy could be explained by the different length of measurements (one day in this study compared to ten days in the previous study) and the amplitude of daily fluctuations of Rn and RnDP levels, caused by cave ventilation. During the period when background aerosols were also monitored, f^{un} values were around 0.04 (Fig. 4b) and the DCF_D value obtained from Eq. (3) was 13.1 mSv WLM⁻¹ (compared to 43.6 mSv WLM⁻¹ in summer). For this period, Fig. 5b shows the individual activity concentrations of unattached and attached RnDPs and the related atom number concentrations. The ratios of the numbers of unattached and attached ²¹⁸Po, ²¹⁴Pb and ²¹⁴Bi atoms are, respectively, 0.22, 0.04 and 0.018 at the start of measurement, and 0.27, 0.04 and 0.021 at the end of measurement, remaining practically constant during visits, in contrast to the summer situation, when the value for ²¹⁸Po decreased drastically. In winter too, this ratio was by far the highest for ²¹⁸Po.

Under winter conditions (Fig. 6b), the number concentration of the < 50 nm particles is about 18 times lower than that of bigger particles, and their total surface area is about 300 times smaller. The probability of ²¹⁸Po, ²¹⁴Pb and ²¹⁴Bi atoms being associated with smaller particles is thus substantially reduced compared to the summer conditions, and consequently the majority of RnDP atoms appear in attached form.

4 Conclusions

These results show why f^{un} values are higher in summer than in winter. The reason for this lies in the size distribution of background aerosol particles in the cave, not merely in their total number concentration. In summer, the contribution of particles smaller than 50 nm, with which unattached RnDPs are associated, is predominant, while in winter its contribution in the total aerosol concentration is minor. Applying the dosimetric approach, the calculated dose conversion factor – hence the dose a tourist guide or tourist receives during a visit – is more than three times higher in summer than in winter.

Acknowledgements. The study was financed by the Slovenian Research Agency within the scope of Project No. J1-0745. The authors thank Ivan Kobal for his careful reading of their manuscript and constructive remarks, and Ivan Iskra for his help in operating the Grimm instrument. The cooperation of the cave management and the technical assistance of Stanislav Glazar are appreciated.

Edited by: R. Crockett

References

- Balcazar, M., Chavez, A., Pina-Villalpando, G., Alfaro, M., and Mendoza, D.: Radon decay products attached to clothes in a nuclear laboratory, *Radiat. Meas.*, 31, 337–342, 1999.
- Birchall, A. and James, A. C.: Uncertainty analysis of effective dose per unit exposure from radon progeny and implications for ICRP risk-weighting factor, *Radiat. Prot. Dosim.*, 53, 133–140, 1994.
- Butterweck, G., Porstendörfer, J., Reineking, A., and Kesten, J.: Unattached fraction and the aerosol size distribution of the radon progeny in a natural cave and mine atmospheres, *Radiat. Prot. Dosim.*, 45, 167–170, 1992.
- Cheng, Y. S., Chen, T. R., Wasiolek, P. T., and Van Engen, A.: Radon and radon progeny in the Carlsbad Caverns, *Aerosol Sci. Tech.*, 26, 74–92, 1997.
- Chu, K. D. and Hopke, P. K.: Neutralization kinetics for polonium-218, *Environ. Sci. Technol.*, 22, 711–717, 1988.
- Cigna, A. A.: Radon in caves, *Int. J. Speleol.*, 34, 1–18, 2005.
- Colbeck, I.: Physical and chemical properties of aerosols, Thomson Science, London, 480 pp., 1998.
- Dankelmann, V., Reineking, A., and Postendörfer, J.: Determination of neutralisation rates of ^{218}Po ions in air, *Radiat. Prot. Dosim.*, 94, 353–357, 2001.
- Darby, S., Hill, D., Auvinen, A., Barrios-Dios, J. M., Baysson, H., Bochicchio, F., Deo, H., Falk, R., Forastiere, F., Hakama, M., Heid, I., Kreienbrock, L., Kreuzer, M., Lagarde, F., Makelainen, I., Muirhead, C., Oberaigner, W., Pershagen, G., Ruano-Ravina, A., Ruosteenoja, E., Rosario, A. S., Tirmarche, M., Tomasek, L., Whitley, E., Wichmann, H. E., and Doll, R.: Radon in homes and risk of lung cancer: collaborative analysis of individual data from 13 European case-control studies, *Br. Med. J.*, 330, 223–226, 2005.
- Field, M. S.: Risks to cavers and cave workers from exposures to low-level ionizing alpha radiation from ^{222}Rn decay in caves, *J. Cave Karst Stud.*, 69, 207–228, 2007.
- Gregorič, A., Zidanšek, A., and Vaupotič, J.: Dependence of radon levels in Postojna Cave on outside air temperature, *Nat. Hazards Earth Syst. Sci.*, 11, 1523–1528, doi:10.5194/nhess-11-1523-2011, 2011.
- Gründel, M. and Porstendörfer, J.: Differences between the activity size distributions of the different natural radionuclide aerosols in outdoor air, *Atmos. Environ.*, 38, 3723–3728, 2004.
- Harrison, J. D. and Marsh, J. W.: Effective dose from inhaled radon and its progeny, *Ann. ICRP*, 41, 378–388, 2012.
- Hofmann, W. and Koblinger, L.: Monte-Carlo modeling of aerosol deposition in human lungs. Part II: deposition fractions and their sensitivity to parameter variations, *J. Aerosol Sci.*, 21, 675–688, 1990.
- Hopke, P. K.: The initial atmospheric behavior of radon decay products, *J. Radioanal. Nucl. Chem.*, 203, 353–375, 1996.
- ICRP: Protection against radon-222 at home and at work, International Commission on Radiological Protection (ICRP), Publication 65, Oxford, 1993.
- ICRP: Lung cancer risk from radon and progeny and statement on radon, International Commission on Radiological Protection (ICRP), Publication 115, Oxford, 2010.
- Iskra, I., Kávási, N., and Vaupotič, J.: Nano aerosols in the Postojna Cave, *Acta Carsol.*, 39, 523–528, 2010.
- Kávási, N., Somlai, J., Kovács, T., Szabó, T., Várhegyi, A., and Hakl, J.: Occupational and patient doses in the therapeutic cave, Tapolca (Hungary), *Radiat. Prot. Dosim.*, 106, 263–266, 2003.
- Markov, K. P., Ryabov, N. V., and Stas, K. N.: A rapid method for estimating the radiation hazard associated with the presence of radon daughter products in air, *Atom. Energy*, 12, 333–337, 1962.
- Marsh, J. W., Birchall, A., Butterweck, G., Dorrian, M. D., Huet, C., Ortega, X., Reineking, A., Tymen, G., Schuler, C., Vargas, A., Vezzu, G., and Wendt, J.: Uncertainty analysis of the weighted equivalent lung dose per unit exposure to radon progeny in the home, *Radiat. Prot. Dosim.*, 102, 229–248, 2002.
- Meisenberg, O. and Tschiersch, J.: Online measurement of unattached and total radon and thoron decay products, *Appl. Radiat. Isot.*, 67, 843–848, 2009.
- Nero, A. V.: Radon and its decay products in indoor air: an overview, in: *Radon and its decay products in indoor air*, edited by: Nazaroff, W. W. and Nero, J. A. V., John Wiley & Sons, New York, 1–53, 1988.
- Nikezić, D., Lau, B. M. F., Stevanović, N., and Yu, K. N.: Absorbed dose in target cell nuclei and dose conversion coefficient of radon progeny in the human lung, *J. Environ. Radioact.*, 89, 18–29, 2006.
- Nikolopoulos, D. and Vogianis, E.: Modelling radon progeny concentration variations in thermal spas, *Sci. Total Environ.*, 373, 82–93, 2007.
- Oberdörster, G., Oberdörster, E., and Oberdörster, J.: Nanotoxicology: an emerging discipline evolving from studies of ultrafine particles, *Environ. Health Perspect.*, 113, 823–839, 2005.
- Pagelkopf, P. and Porstendörfer, J.: Neutralisation rate and the fraction of the positive ^{218}Po -clusters in air, *Atmos. Environ.*, 37, 1057–1064, 2003.
- Papastefanou, C.: Radioactive aerosols, in: *Radioactivity in the environment*, edited by: Baxter, M. S., Elsevier, Amsterdam, 171 pp., 2008.
- Pashenko, S. E. and Dublyansky, Y. V.: Generation of cave aerosols by alpha particles: critical evaluation of the hypothesis, *J. Cave Karst Stud.*, 59, 103–105, 1997.
- Porstendörfer, J. and Mercer, T. T.: Influence of electric charge and humidity upon the diffusion-coefficient of radon decay products, *Health Phys.*, 37, 191–199, 1979.
- Porstendörfer, J.: Behaviour of radon daughter products in indoor air, *Radiat. Prot. Dosim.*, 7, 107–113, 1984.
- Porstendörfer, J. and Reineking, A.: Indoor behaviour and characteristics of radon progeny, *Radiat. Prot. Dosim.*, 45, 303–311, 1992.
- Porstendörfer, J., Zock, C., and Reineking, A.: Aerosol size distribution of the radon progeny in outdoor air, *J. Environ. Radioact.*, 51, 37–48, 2000.
- Rovenská, K., Thinová, L., and Ždímal, V.: Assessment of the dose from radon and its decay products in the Bozkov Dolomite Cave, *Radiat. Prot. Dosim.*, 130, 34–37, 2008.
- Šebela, S.: Tectonic structure of Postojnska jama cave system, ZRC Publishing (Scientific Research Centre), Ljubljana, 112 pp., 1998.

- Smerajec, M. and Vaupotič, J.: Nanoaerosols Including Radon Decay Products in Outdoor and Indoor Air at a Suburban Site, *J. Toxicol.*, 2012, 510876, doi:10.1155/2012/510876, 2012.
- Somlai, J., Szeiler, G., Szabó, P., Várhegyi, A., Tokonami, S., Ishikawa, T., Sorimachi, A., Yoshinaga, S., and Kovács, T.: Radiation dose of workers originating from radon in the show Cave of Tapolca, Hungary, *J. Radioanal. Nucl. Chem.*, 279, 219–225, 2009.
- Streil, T., Holfeld, G., Oeser, V., Federsen, C., and Schönfeld, K.: SARAD EQF 3020: a new microsystem based on monitoring system for the continuous measurement of radon and the attached and unattached fraction of radon progeny, International Radiological Protection Association (IRPA), Regional congress on radiation protection in neighbouring countries in Central Europe, Portorož, 1996.
- Tokonami, S.: Experimental verification of the attachment theory of radon progeny onto ambient aerosols, *Health Phys.*, 78, 74–79, 2000.
- UNSCEAR: Sources and effects of ionizing radiation, United Nations Scientific Committee on the Effects of Atomic Radiation, Vol. 1, UNSCEAR 2000 Report to the General Assembly, New York, 83–156, 2000.
- Vaupotič, J.: Nano-size radon short-lived progeny aerosols in Slovenian kindergartens in wintertime, *Chemosphere*, 69, 856–863, 2007.
- Vaupotič, J.: Nanosize radon short-lived decay products in the air of the Postojna Cave, *Sci. Total Environ.*, 393, 27–38, 2008a.
- Vaupotič, J.: Comparison of various methods of estimating radon dose at underground workplaces in wineries, *Radiat. Environ. Biophys.*, 47, 527–534, 2008b.
- Vaupotič, J.: Radon levels in karst caves in Slovenia, *Acta Carsol.*, 39, 503–512, 2010.
- Vaupotič, J. and Kobal, I.: Effective doses in schools based on nano-size radon progeny aerosols, *Atmos. Environ.*, 40, 7494–7507, 2006.
- Vaupotič, J., Csige, I., Radolić, V., Hunyadi, I., Planinić, J., and Kobal, I.: Methodology of radon monitoring and dose estimates in Postojna Cave, Slovenia, *Health Phys.*, 80, 142–147, 2001.
- Wang, J., Meisenberg, O., Chen, Y., Karg, E., and Tschiersch, J.: Mitigation of radon and thoron decay products by filtration, *Sci. Total Environ.*, 409, 3613–3619, 2011.