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The Importance of Nanosize Aerosols of Radon Decay Products in Radon Dosimetry*

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Air concentrations of radon and radon short-lived decay products, equilibrium factor, unattached fraction of radon decay products (f_{un}), barometric pressure, air temperature and relative air humidity in the Postojna Cave and in a kindergarten were measured, with the emphasis on f_{un}. Applying the dosimetry model, dose conversion factors were calculated on the basis of f_{un} values obtained (ranging from 0.09 to 0.65 in the cave and from 0.03 to 0.25 in the kindergarten) and shown to exceed significantly 5 mSv WLM^{-1}, the value proposed on the basis of epidemiological studies.

INTRODUCTION

Radon is a radioactive noble gas formed by radioactive decay of radium in natural radioactive decay chains in the Earth's crust, *i.e.*, ^{222}Rn isotope (α decay, half-life, $t_{1/2}$ = 3.82 days) from ^{226}Ra in the ^{238}U chain, ^{220}Rn isotope (α decay, $t_{1/2}$ = 55 s) from ^{224}Ra in the ^{232}Th chain, and ^{219}Rn isotope (α decay, $t_{1/2}$ = 3.92 s) from ^{223}Ra in the ^{235}U chain.[1] Only a small fraction of radon (described as the emanation coefficient) enters the space between mineral grains and, thus, has the possibility to travel away from the source, carried either by carrier gases (methane, carbon dioxide) or by water.[2] It accumulates in underground rooms (karst caves, mines) and eventually reaches the living and working environment. Of the three radon isotopes, only ^{222}Rn (also simply called radon) is usually of concern to humans, since it appears at a considerable level in the atmosphere because of its long half-life compared with half-lives of ^{220}Rn (thoron) and ^{219}Rn (action).[1] Therefore, following common practice, hereafter 'radon' (or Rn) refers to ^{222}Rn.

Radon α decays and is always accompanied by its short-lived decay products (RnDP) in the following sequence: ^{218}Po (α decay, $t_{1/2}$ = 3.10 min), ^{214}Pb (β/γ decay, $t_{1/2}$ = 26.8 min), ^{214}Bi (β/γ decay, $t_{1/2}$ = 19.9 min), and ^{214}Po (α decay, $t_{1/2}$ = 164 μs). Initially, these products are positively charged free ions which, sooner or later, depending on environmental conditions, are partly neutralized in the air by recombination with small ions in the air, electron scavenging by OH radicals, or charge transfer by molecules of lower ionization potential. They form nanosize clusters, the so-called *unattached* RnDP, with the activity median aerodynamic diameters (AMAD) of less than 10 nm.[3–5] They further attach to aerosol particulates, forming *attached* RnDP, the radioactive aerosols with AMAD between 10 and 1000 nm.[6] Due to plating-out of aerosols to the walls and floor of a room, as

* Dedicated to Professor Nikola Kallay on the occasion of his 65th birthday.

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well as air movement and entry of fresh air, radioactive equilibrium between RnDP and Rn (when their activity concentrations expressed in Bq m^{-3} are equal) is only partly reached and is expressed as a fraction between 0 and 1, called the *equilibrium factor*, *F*.[1] When breathing air contaminated by Rn and RnDP, part of the RnDP is deposited on the walls of respiratory airways, where it decays, releasing the energy to the tissue, thus damaging it and eventually causing cancer. Computational simulations have shown that deposition patterns of RnDP in the lung[7] depend strongly on the size distribution of RnDP and RnDP-bearing aerosols, which therefore play a crucial role in radon dosimetry.[8–12]

The detrimental effect of Rn and RnDP can be quantified on the basis of the radiation dose received by breathing air containing Rn and RnDP. For this purpose, a dose conversion factor (DCF) has been introduced, which is defined as the ratio between the weighted equivalent dose to the lung (assuming the radiation weighting factor for α particles, $w_\alpha = 20$, and the tissue weighting factor for lung, $w_{\text{lung}} = 0.12$) expressed in mSv, and 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 of 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 concentration of ^{218}Po, ^{214}Bi and ^{214}Pb (^{214}Po), which are in radioactive equilibrium (F = 1) with 100 pCi L^{-1} (3700 Bq m^{-3}) of ^{222}Rn, resulting in an alpha energy concentration of 1.3×10^5 MeV L^{-1}.[1] DCF values may be obtained either on the basis of the results of epidemiologic studies (hereafter denoted by DCF$_E$) or calculated applying dosimetric models (hereafter denoted by DCF$_D$). It is now generally accepted and recommended by the International Commission for Radiological Protection (ICRP) in Publication 65[13] that 5 mSv WLM^{-1} for working and 4 mSv WLM^{-1} for living environments should be used for DCF$_E$ in radon dosimetry.

In contrast, Birchall and James[14] developed and elaborated the dosimetric approach of calculating DCF$_D$, based on a refined, recently proposed human respiratory tract model.[15] They also showed the parameters that most affect DCF$_D$, ranked in the following order: unattached fraction of RnDP (f_{un}), nucleation aerosol size, nucleation fraction, the unattached aerosol size, *etc*.[16,17] By taking $w_\alpha = 20$ and $w_{\text{lung}} = 0.12$, DCF$_D$ values in the range 8–32 mSv WLM^{-1} were obtained under different conditions, with 15 mSv WLM^{-1} as the 'best estimate' for the indoor air conditions in dwellings, which is higher by a factor of 3 than DCF$_E$. It was also shown[16] that the relationship of DCF$_D$ to f_{un} may be approximated empirically by:

$$\text{DCF}_\text{D} = 11.35 + 0.43 \times f_{\text{un}} \qquad (1)$$

Reasons for disagreement between DCF$_E$ and DCF$_D$ values were recently well reviewed by Birchall and Marsh.[18] They have not been fully clarified, but are argued to be probably due to too high values chosen for w_α and w_{lung}.[18–20]

In addition, Porstendörfer[21] proposed expressions to calculate DCF$_D$ for mouth (DCF$_{Dm}$) and nasal (DCF$_{Dn}$) breathing:

$$\text{DCF}_\text{Dm} = 101 \times f_{\text{un}} + 6.7 \times (1 - f_{\text{un}}) \qquad (2)$$

$$\text{DCF}_\text{Dn} = 23 \times f_{\text{un}} + 6.2 \times (1 - f_{\text{un}}) \qquad (3)$$

For a person with a fraction x of mouth breathing and a fraction $(1 - x)$ of nose breathing, a combined dose conversion factor (DCF$_{Dc}$) is then calculated as:

$$\text{DCF}_\text{Dc} = x \times \text{DCF}_\text{Dm} + (1 - x) \times \text{DCF}_\text{Dn} \qquad (4)$$

While the epidemiology-based DCF$_E$ is recommended by the ICRP-65[13] to be used in general radon dosimetry, Publication 66[15] suggests the use of dosimetry-based DCF$_D$ for research purposes only, when dependence of the radiation dose on environmental parameters is sought.

As nanosize RnDP particles play a crucial role in radon dosimetry, the interest in f_{un} studies is rapidly growing. Nonetheless, while the database on Rn in various environments has become very rich, reports on f_{un} values are sparse, because the measuring techniques are complicated and devices have only recently become available in the market.[22] This paper reports on measurements of f_{un}, together with activity concentrations of Rn and RnDP, and F in two typical environments: (i) air in the Postojna Cave (where enhanced exposure to radon may easily occur)[23] and (ii) air in a selected kindergarten (where children, the most vulnerable population, are exposed to radon).[24] We discuss the influence of meteorological parameters on the measured values (with emphasis on f_{un}), comment on DCF$_D$ values obtained on the basis of measured f_{un}, and compare them with 5 mSv WLM^{-1}, in order to show the discrepancy between the epidemiology-based and dosimetry-based dose conversion factors in real environments.

EXPERIMENTAL

Measuring Sites

One measuring site was at the lowest point of the guided tourist tour of the Postojna Cave, the largest of 12 show caves in Slovenia, visited by about half a million tourists a year. An electric train takes visitors from the entrance to the railway station in the cave, wherefrom they start a walking tour which, in about 1.5 hours, brings them back to the railway station. Visits are scheduled for every full hour from 9

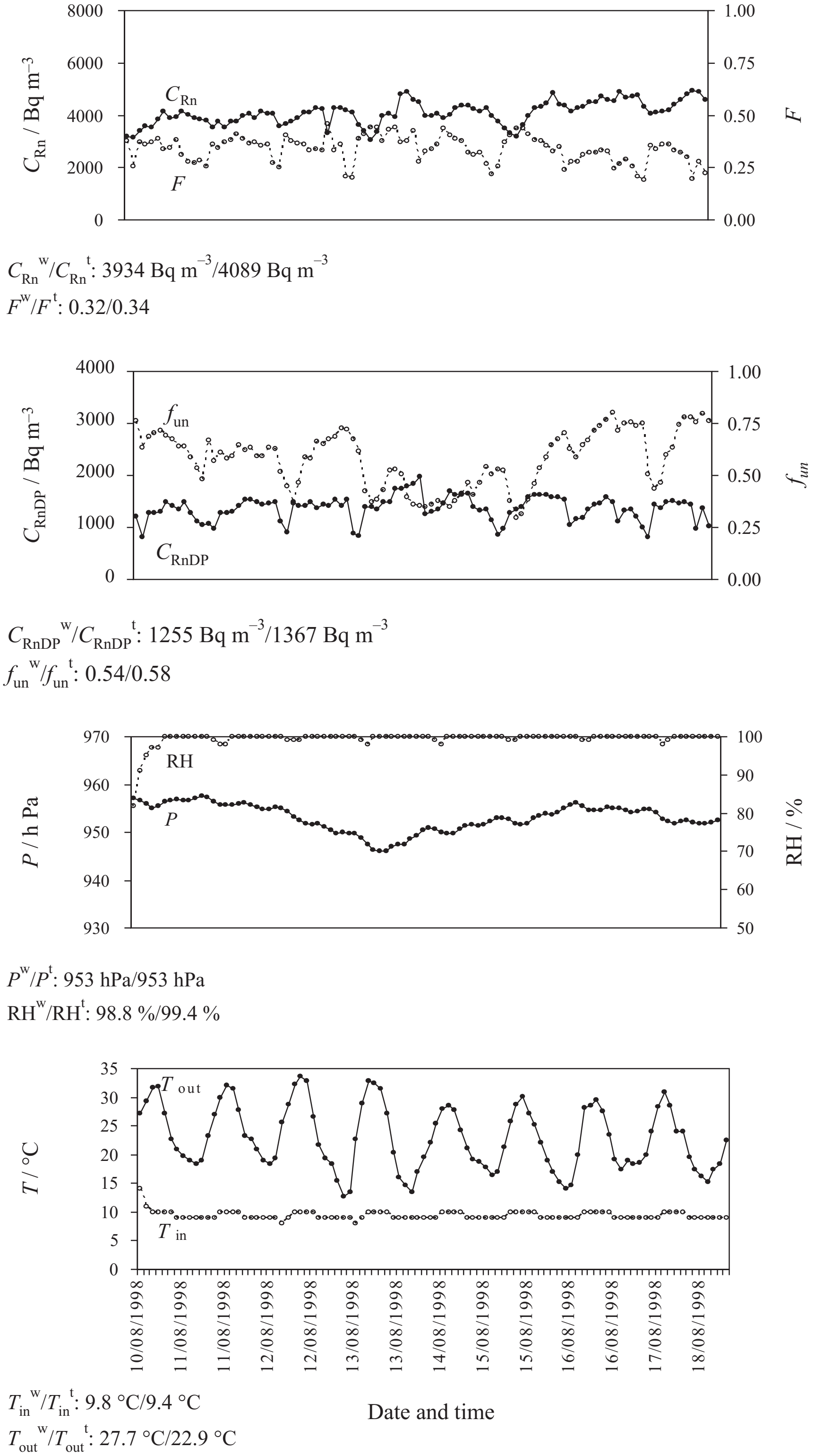
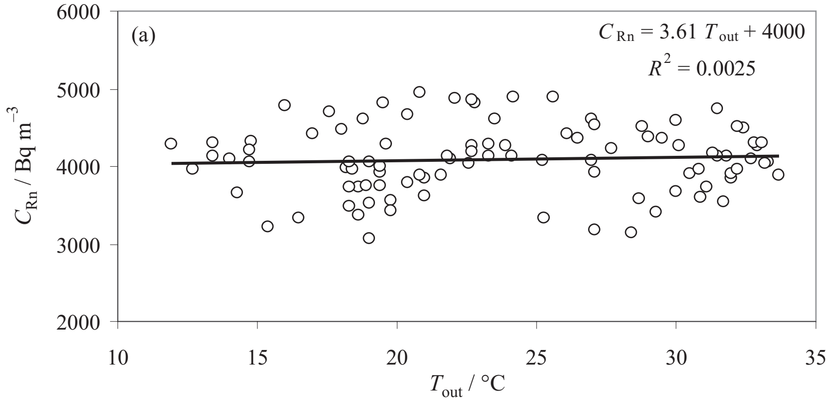


Figure 1. The lowest point in Postojna Cave, August 10–18, 1998: concentrations of radon (C_{Rn}) and radon decay products (C_{RnDP}), equilibrium factor (F), unattached fraction of radon decay products (f_{un}), barometric pressure (P), relative air humidity in the cave (RH), and air temperature in the cave (T_{in}) and outdoors (T_{out}). Average values during the whole period of measurement (denoted by t – *total*) and during the working hours only (denoted by w – *working*) are quoted for each parameter.

a.m. to 6 p.m. in spring, summer and autumn, and every other full hour from 10 a.m. to 4 p.m. in winter. The temperature of the cave air is practically constant all the year round (8–10 °C) and relative humidity is close to 100 %. Elevated Rn levels in the cave were discovered decades ago[25] and regular radon monitoring was recently introduced. The work-



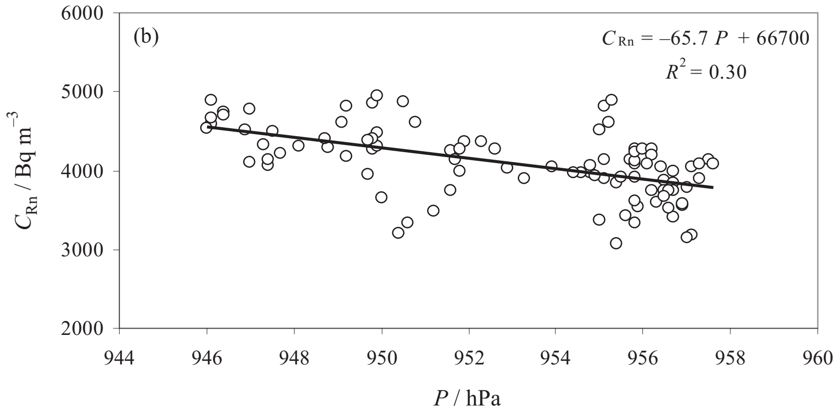


Figure 2. The dependence of radon concentration (C_{Rn}) on: (a) outdoor air temperature (T_{out}) and (b) barometric pressure (P) at the lowest point in the Postojna Cave, August 10–18, 1998.

ing time of tourist guides has been limited to ensure sufficiently low exposure to radon.[25] The cave environment is therefore of special interest. Measurements within this study were carried out in summer when Rn levels are known to be highest.[25,26]

The second measuring site was a room in a kindergarten in which our survey within the national radon project revealed elevated radon levels.[27] It was built of prefabricated elements in 1980 and is laid directly on the ground, without a basement underneath. There is no air-conditioning, and the rooms are ventilated by opening the doors and windows. During winter, rooms are heated with hot-water radiators connected to a gas central heating system. Working hours are from 5.30 a.m. to 5.00 p.m. Children spend some time outdoors, the length depending on the weather and season. Measurements for this study were carried out in winter when Rn levels are known to be highest.[28]

Measuring Techniques

A portable Sarad EQF3020 device (manufactured by Sarad, Dresden, Germany) was used. It measures activity concentrations of Rn and RnDP, F and f_{un}.[29] It also records air temperature and relative air humidity, the two parameters affecting f_{un}.[30–33] The frequency of sampling and analyses is once every two hours. The device was used several times for 10-15 days at the lowest point in the Postojna Cave in summer and in the selected kindergarten room in winter.

The instrument was calibrated by the manufacturer on purchase, checked regularly during the inter-comparison experiments organized annually by the Slovenian Nuclear Safety Administration at the Ministry of the Environment and Spatial Planning,[34] and re-calibrated every two years in the manufacturer's radon chamber.

RESULTS AND DISCUSSION

Postojna Cave

The results of measurements of concentrations of radon (C_{Rn}) and radon decay products (C_{RnDP}), equilibrium factor (F), unattached fraction of radon decay products (f_{un}), barometric pressure (P), relative air humidity in the cave (RH) and air temperature in the cave (T_{in}) and outdoors (T_{out}) as a function of time, carried out at the lowest point in the Postojna Cave in August 1998, are shown in Figure 1, together with their average values for the whole period of measurement (denoted by t for *total* average, *e.g.*, C_{Rn}^{t}) and the working hours only (denoted by w for *working* average, *e.g.*, C_{Rn}^{w}).

Rn concentration always exceeded the Slovenian national tolerance limit[35] of 400 Bq m^{-3} (Figure 1). Its

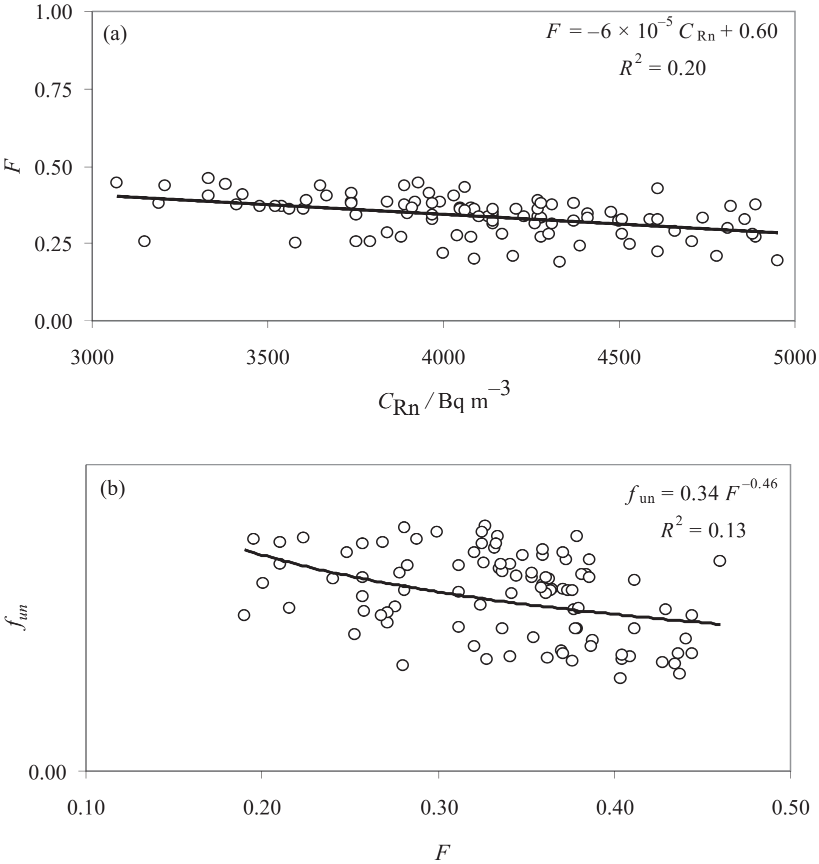


Figure 3. The dependence of: (a) equilibrium factor (F) on radon concentration (C_{Rn}) and (b) unattached fraction of RnDP (f_{un}) on the equilibrium factor (F) at the lowest point in the Postojna Cave, August 10–18, 1998.

diurnal fluctuation results from the combined effect of several parameters, such as barometric pressure and its time gradient, difference between the air temperature in the cave and outdoors, working regime and number of visitors. The cave system works as a chimney: since the cave air temperature is constant all the year round, the winter outdoor temperature is lower than in the cave, causing a natural draught of air from the cave through vertical channels into the outdoor atmosphere. The situation is reversed in summer and this draught is minimal, or zero. This is evident from Figure 2a, where no dependence of C_{Rn} on T_{out} is seen. In summer, the cave air is practically stagnant because $T_{out} > T_{in}$. Thus, at a constant Rn source – similar emanation from the cave walls and similar transport from remote caverns as in winter – radon accumulation in the cave air is enhanced in summer, resulting in concentrations 2–3 times higher in summer than in winter.[25,26] The Postojna Cave may be classified as a 'horizontal cave' for which practically no dependence of C_{Rn} on P is expected.[36] However, a weak negative correlation between C_{Rn} and P is shown in Figure 2b. As P increases, exhalation of Rn from the cave walls is reduced and so is C_{Rn}, in accordance with the general effect of pressure fluctuations on radon exhalation.[37–39] A weak negative correlation was also observed

TABLE I. Dose conversion factors for mouth (DCF_{Dm}) and nasal (DCF_{Dn}) breathing calculated from the arithmetic mean value (AM) of measured f_{un}^{w} in the Postojna Cave and in the kindergarten. For reference: DCF_E = 5 mSv WLM^{-1}

Environment	f_{un}^{w} range	f_{un}^{w} AM	DCF_{Dm} mSv WLM^{-1}	DCF_{Dn} mSv WLM^{-1}
Cave	0.09–0.65	0.54	57.6	15.3
Kindergarten	0.03–0.25	0.11	17.2	8.0

between F and C_{Rn} (Figure 3a), a phenomenon reported also by other authors.[30] This is also reflected in lower F values in summer and higher in winter (measured previously, not shown here). One reason for this is the much larger number of visitors in summer than in winter, causing a higher plate-out of RnDP and hence reducing F in summer.

f_{un} values in the cave air (Figure 1) are higher than previously observed in Slovene schools.[40] This agrees well with the data reported by Cheng *et al.*[41] for other karst caves. As expected, higher F values are accompanied by lower f_{un} values.[1,42–44] Figure 3b shows the f_{un} – F relationship observed at the lowest point, described by the power expression:[45] $f_{un} = 0.34\ F^{-0.46}$. The low f_{un} value in stagnant air[46] during the night was rapidly increased

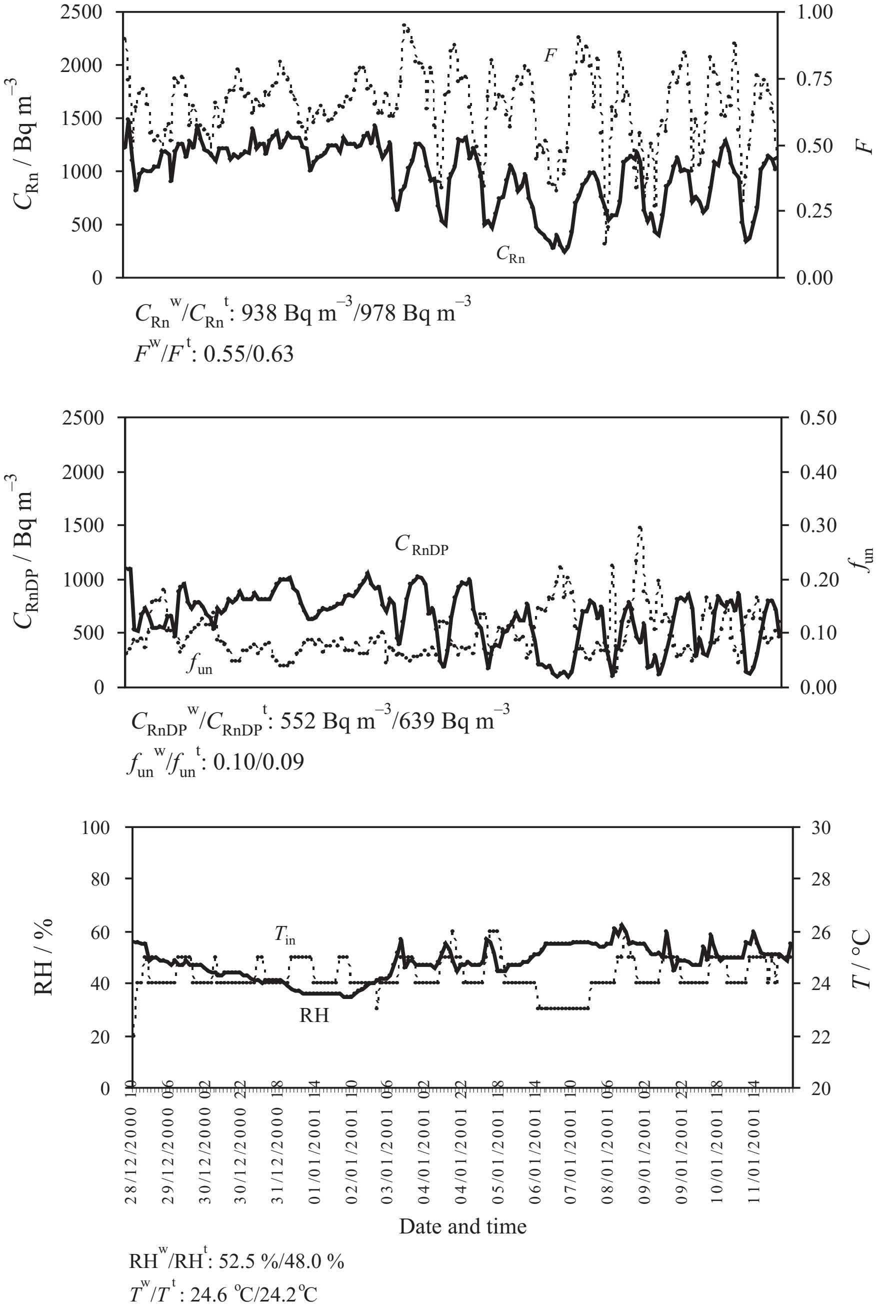


Figure 4. The kindergarten, from December 28, 2000 to January 11, 2001: concentrations of radon (C_{Rn}) and radon decay products (C_{RnDP}), equilibrium factor (F), unattached fraction of radon decay products (f_{un}), relative air humidity in the cave (RH), and air temperature (T_{in}). Average values during the whole period of measurement (denoted by t – *total*) and during the working hours only (denoted by w – *working*) are quoted for each parameter.

by visits in the morning, and started to decrease in the afternoon.

Using $f_{un}^{w} = 0.54$ from Figure 1 in equations (2) and (3), the following result was obtained: $DCF_{Dm} = 57.6$ mSv WLM^{-1} and $DCF_{Dn} = 15.3$ mSv WLM^{-1} (Table I). These values are higher by factors of 11.5 and 3.1, respectively, than the epidemiology-based value, $DCF_E =$ 5 mSv WLM^{-1}. For a tourist guide who spends the majority of her/his time in the cave talking to visitors, the large proportion of mouth breathing should be taken into account ($x \rightarrow 1$ in Eq. (4), $DCF_{Dc} \rightarrow DCF_{Dm}$) for dose estimates and hence the DCF_{Dc} value at the lowest point is 11.5 fold higher than 5 mSv WLM^{-1}. In contrast, the locomotive drivers and maintenance workers in the cave work at the other extreme, with mostly nasal breathing ($x \rightarrow 0$ in Eq. (4), $DCF_{Dc} \rightarrow DCF_{Dn}$). For them, the DCF_{Dc} values are only 3.1 fold higher than 5 mSv WLM^{-1}. Realistic DCF_{Dc} values would be obtained by using actual data of the mouth/nasal breathing ratio for different profiles of employees working in the cave.

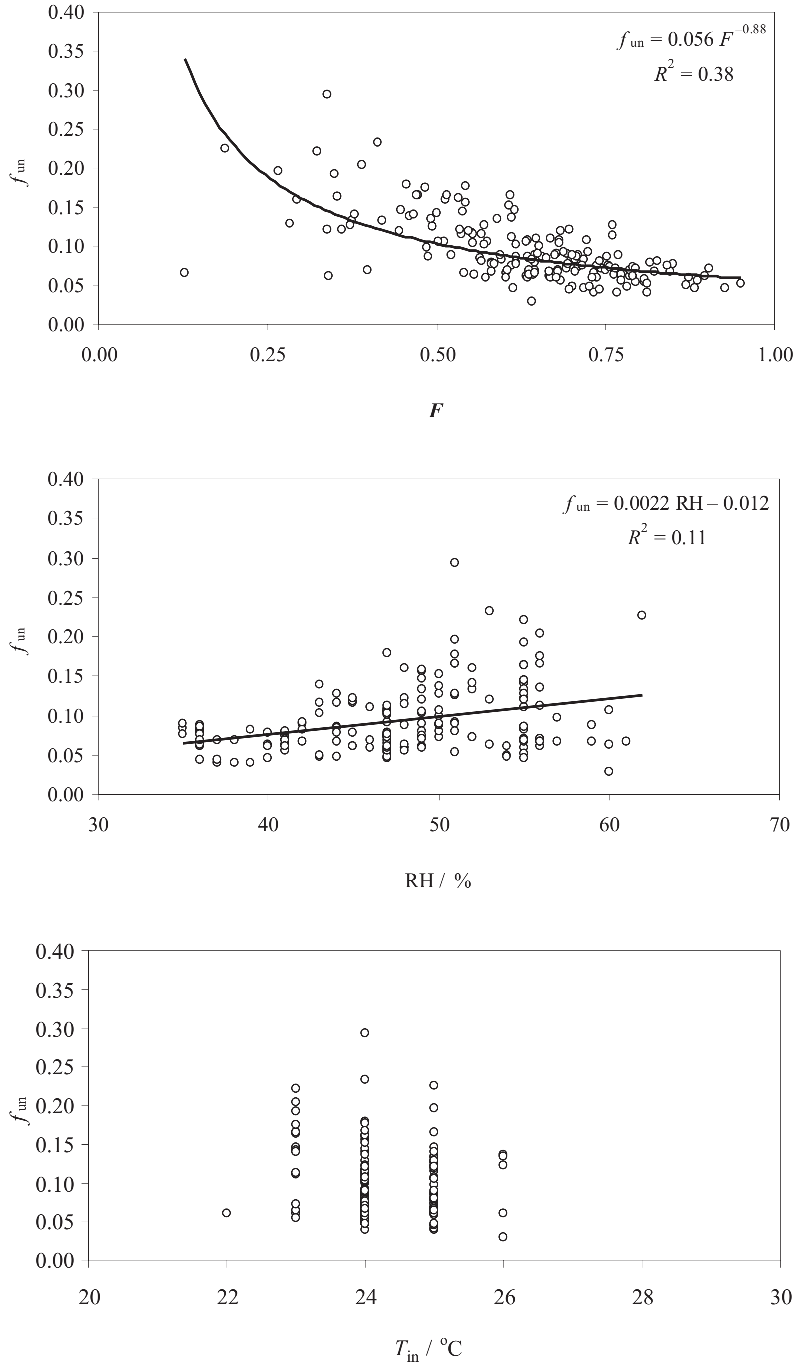


Figure 5. Dependence of the unattached fraction of RnDP (f_{un}) on: (a) equilibrium factor (F), (b) relative humidity of indoor air (RH) and (c) indoor air temperature (T_{in}) in the kindergarten, from December 28, 2000 to January 11, 2001.

Kindergarten

The time dependence of C_{Rn}, F, C_{RnDP}, f_{un}, RH and T_{in} recorded in the kindergarten is shown in Figure 4, together with the *total* and *working* average values. The f_{un} values are significantly lower than in the Postojna Cave and similar to those in schools.[40] C_{Rn}, C_{RnDP} and F showed diurnal fluctuations, as observed previously,[26] with maxima during nights, minima in the afternoons of working days, and remained high over the first weekend (from 29.12. to 3.1.), but low over the second (5.1.–8.1.) when, due to maintenance works, the doors and windows were kept open. While RH changed substantially, T varied by not more than ±2 °C.

Also here, a negative correlation between f_{un} and F was observed and was approximated by the power expression $f_{un} = 0.056\ F^{-0.88}$ (Figure 5a). During working hours, the plate-out of aerosols is enhanced, resulting in reduced aerosol concentration in the air[47] and thus lowering F and increasing f_{un}.

The f_{un} – RH correlation is expected to be negative:[30] higher RH causes wetting and thus increases the size of aerosol particulates, thereby increasing the attachment rate of RnDP and decreasing f_{un}. This was not observed in our measurements, which show a weak positive f_{un} – HR correlation (Figure 5b). This apparent discrepancy probably originates from the following two effects:[30] (i) water molecules enhance clustering, thus decreasing diffusion, and (ii) water molecules enhance neutralization of ^{218}Po, initially mostly present in charged form,[5,33] and thus increase the proportion of neutral particles and increase diffusion. However, an increase in RH from 45 % to 65 % is probably not enough to increase neutralization rate significantly.[5] Neutralization is also influenced indirectly by temperature because it governs RH. In our experiment no correlation between T and f_{un} was observed, as seen in Figure 5c. The f_{un} – T correlation is a result of complex environmental effects not studied in detail here, and therefore no right interpretation of this figure is possible at the moment.

Using f_{un}^{w} = 0.11 in Eqs. (2) and (3), the following values were obtained: DCF_{Dm} = 17.2 mSv WLM^{-1} and DCF_{Dn} = 8.0 mSv WLM^{-1} (Table I). The ICRP-recommended value of 5 mSv WLM^{-1} is exceeded by a factor of 3.4 for mouth and 1.6 for nasal breathing. Because the f_{un}^{w} values are lower in the kindergarten than in the cave, the disagreement between the epidemiology-based and dosimetry-based dose conversion factors is also lower.

CONCLUSIONS

Monitoring the unattached fraction of radon short-lived decay products (f_{un}) has led to values between 0.09 and 0.65, with an arithmetic mean of 0.54, in the air of the Postojna Cave, and between 0.03 and 0.25, with the arithmetic mean of 0.11, in a kindergarten. Applying the dosimetric approach, dose conversion factors for mouth (DCF_{Dm}) and nasal (DCF_{Dn}) breathing were calculated and amount to 57.6 mSv WLM^{-1} and 15.3 mSv WLM^{-1}, respectively, for the Postojna Cave, and 17.2 mSv WLM^{-1} and 8.0 mSv WLM^{-1}, respectively, for the kindergarten. These values greatly exceed the value of the epidemiology-based dose conversion factor, DCF_D = 5 mSv WLM^{-1} regularly used in radon dosimetry, and thus exemplify disagreement between the results obtained on the basis of epidemiology and those calculated using dosimetric models for these two real environments. Such results show the importance of nanosize particulates in radon dosimetry and thus contribute to nanotoxicological aspects of radon. They may further stimulate discussions[18] about parameters affecting the effective dose received by breathing in Rn and RnDP, and about the way of accounting for the disagreement between the two approaches.

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SAŽETAK

Značaj aerosola nano veličine produkata raspada radona u radon dozimetriji

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Mjerene su koncentracije radona i kratkoživućih produkata raspada radona u zraku, ravnotežni faktor, nevezani udjel produkata raspada radona (f_{un}), barometarski tlak, temperatura i relativna vlažnost zraka u Postojnskoj Jami i dječjem vrtiću, s naglaskom na f_{un}. Primjenom dozimetrijskog modela, izračunati su konverzijski faktori za doze na temelju dobivenih f_{un} vrijednosti (u rasponu od 0,09 do 0,65 u špilji i od 0,03 do 0,25 u dječjem vrtiću) i pokazano je da značajno premašuju 5 mSv WLM^{-1}, vrijednost predloženu na temelju epidemioloških studija.