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DIURNAL AND SEASONAL VARIATIONS OF CONCENTRATION AND SIZE DISTRIBUTION OF NANO AEROSOLS (10–1100 nm) ENCLOSING RADON DECAY PRODUCTS IN THE POSTOJNA CAVE, SLOVENIA

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At the lowest point along the tourist route in the Postojna Cave, the activity concentration of radon (^{222}Rn) short-lived decay products and number concentration and size distribution of background aerosol particles in the size range of 10–1100 nm were measured. In the warm yearly season, aerosol concentration was low (52 cm^{-3}) with 21 % particles smaller than 50 nm, while in the cold season, it was higher (1238 cm^{-3}) with 8 % of <50 nm particles. Radon activity concentrations were 4489 and 1108 Bq m^{-3}, and fractions of unattached radon decay products were 0.62 and 0.13, respectively.

INTRODUCTION

In the Postojna Cave, the biggest in Slovenia and one of the biggest show caves in the world, radon (^{222}Rn) concentration is high, as in the majority of karst caves worldwide[1–5], and therefore the working time of the tourist guides is limited[6]. According to Butterweck *et al.*[7], the unattached fraction (f_{un}) of radon short-lived decay products (RnDP) in the Postojna Cave appear within 1.3–6.0 nm and the attached, within 119–289 nm. In order to reveal how f_{un}, the crucial parameter in radon dosimetry[8–10], is controlled by the presence of background aerosol particles with which RnDP are associated, number concentration and size distribution of aerosol particles in the 10–1100 nm size range were recently introduced[11]. In the present paper, measurements at the lowest point along the guided route in the cave in February, April and June are described and results presented and commented on.

MATERIALS AND METHODS

Site description

In the Postojna Cave, air temperature is in the range of 9.7–12°C all the year round and relative air humidity is at 97–99 %. In winter, when the cave temperature is higher than outside, cave air is released from the cave into the outside atmosphere due to the air draught caused by the 'chimney effect', thus allowing fresh and cold outdoor air to enter the cave through low lying openings. This effect is not operative in summer, when the outside temperature is higher than in the cave, and air draught is minimal or reversed.

Measurement of radon decay products

To monitor RnDP aerosols, an EQF3020-2 device (Equilibrium Factor Monitor-System, Sarad, Germany) was used which gives every 2 h: activity concentrations (Bq m^{-3}) of radon (C_{Rn}), individual activity concentrations of RnDP in the unattached and attached form, equilibrium equivalent activity concentration of RnDP (C_{RnDP}), equilibrium factor ($F = C_{RnDP}/C_{Rn}$) and fraction of unattached RnDP ($f_{un} = C^{un}_{RnDP}/C_{RnDP}$), as well as air temperature and humidity.

Measurement of background aerosols

The term 'background' is used here to comprise all particles: those with RnDP associated (although their contribution in the number concentration is negligible, as seen later) and those without. Their number concentration and size distribution were measured with SMPS+C instrument (Scanning Mobility Particle Sizer+Counter), Series 5.400 (Grimm, Germany). The long DMA (Differential Mobility Analyser) unit, available in the laboratory, was used, designed for the 10–1100 nm size range, although the range of 0.10–1100 nm would be preferred for the purpose of this study. The frequency of measurement is 1 in 7 min. The instrument gives the total number concentration C_b(tot) and size distribution $dC_b(d)/d\ln d$, with d, electrical mobility-equivalent particle diameter. Bearing in mind that RnDP in Postojna Cave are attached to aerosol particles bigger than 100 nm[7], we selected 50 nm as the border between the particles associated with the unattached and attached RnDP, and were interested in the fraction $x_b(<50)=C_b(<50)/C_b(\text{tot})$.

Several one-day measurements were carried out with both devices in the middle of February, middle of April and beginning of June at the lowest point along the guided walking route in the cave. The SMPS+C device was run for several hours only during morning visits, because it is not designed to be used in high air humidity and therefore its operation was kept minimised.

RESULTS AND DISCUSSION

Table 1 summarises the results of the three periods. C_{Rn} and C_{RnDP} are highest in summer (June) and lowest in winter (February), due to effective ventilation of the cave during the cold period. The inflow of fresh air in winter brings outside aerosol particles into the cave, thus resulting in elevated C_b(tot) values, as compared with about 20 times lower values in summer. The values in spring (April) are in between. This is reflected in F values which gradually decrease from winter to summer. Thus, C_{RnDP} is the highest in spring, and not in summer when C_{Rn} is the highest. The inflow of outside particles with a geometric mean of diameter of 63 ± 19 nm[12] is additionally evidenced in Figure 1, showing number size distribution in the three periods, before tourist visits started and during them. Particles of sizes grouping around 100 nm are present in all periods, although their contribution to C_b(tot) differs substantially. In winter (Figure 1a), they predominate and the concentration of particles smaller than 50 nm is very low. The passing by of tourists did not change the size distribution markedly, geometric means of diameters being 110 and 113 nm.

Table 1. Minimum, maximum, arithmetic mean and arithmetic standard deviation values of total number concentration of background aerosol (C_b(tot)), fraction of particles smaller than 50 nm (x_b(<50)), activity concentration of radon (C_{Rn}), equilibrium equivalent activity concentration of radon short-lived decay products (C_{RnDP}), equilibrium factor between radon and its decay products (F) and fraction of unattached radon decay products (f_{un}), at the lowest point in the Postojna Cave in morning hours of 22 February, 12 April and 4 June, 2011.

	C_b(tot)/ cm^{-3}	$x_b(<50)$	C_{Rn}/ Bq m^{-3}	F	C_{RnDP}/ Bq m^{-3}	f_{un}
February 22						
AM	1238	0.08	1108	0.75	831	0.13
ASD	160	0.02	78	0.01	64	0.02
min	984	0.04	1026	0.74	771	0.11
max	1512	0.12	1182	0.76	898	0.15
April 12						
AM	853	0.51	3293	0.58	1915	0.36
ASD	283	0.13	201	0.02	188	0.07
min	329	0.2	3099	0.57	1756	0.29
max	1802	0.65	3501	0.78	2639	0.41
June 4						
AM	52	0.21	4489	0.27	1213	0.62
ASD	21	0.18	44	0.02	93	0.02
min	28	0	4458	0.26	1147	0.6
max	108	0.57	4520	0.28	1279	0.63

The situation was different in summer (Figure 1c), when passages of visiting groups (shaded region in Figure 2), enhanced both C_b(tot) and $x_b(<50)$ considerably, mainly contributed by particles smaller than 20 nm; they both decreased to their initial values between two groups. These small particles, presumably clusters of water molecules or water nano droplets, are obviously produced by visitors' breathing. Changes in the concentration of the <50 nm particles (to which unattached RnDP are associated) did not affect f_{un}, and the expected inverse relationship between C_b(tot) and f_{un}[13] was not observed. During the entire period shown in Figure 2, f_{un} remained practically unchanged (region between two vertical lines in Figure 3), although during the visit of the first group $x_b(<50)$ increased from about 0.10 to 0.50. Short-time changes in size distribution of aerosol particles may not be expected to cause immediate change in f_{un}, i.e. redistribution between unattached and attached Po, Pb and Bi atoms. It rather influences only the newly born Po, Pb and Bi atoms and clusters. Creation of these atoms by radioactive transformations, their neutralization, clustering, and attachment to[14] and detachment from background aerosol particles by recoil take time. Calculations have shown[15] that a time delay of hours[16] necessarily appears between a change in particle size distribution of aerosol particles and a change in f_{un}. Therefore, a true response of f_{un} on the fast changes in background aerosol in the case was masked or even totally obscured. It is only seen in slow changes, such as when comparing winter and summer situation. Then, both f_{un} – C_b(tot)[13] and f_{un} – F[17–19] inverse relationships are evident (Table 1).

In addition, during the period of using the SMPS+C device only three readings from the EQF device were obtained, because its analysis frequency is only once in 2 h, and therefore the relationship between f_{un} and $x_b(<50)$ is difficult to evaluate correctly.

In spring (April), size distribution (Figure 1b) before visits was similar to that in winter (Figure 1a), but with a tail toward smaller diameters. Then, the contribution of particles smaller than about 30 nm started to increase already prior to visits. Particles of <30 nm size are also emitted when boiling water[12]. Therefore, one may speculate that in spring, when a weak ventilation is still operative, warmer and humid outside air entering the cave

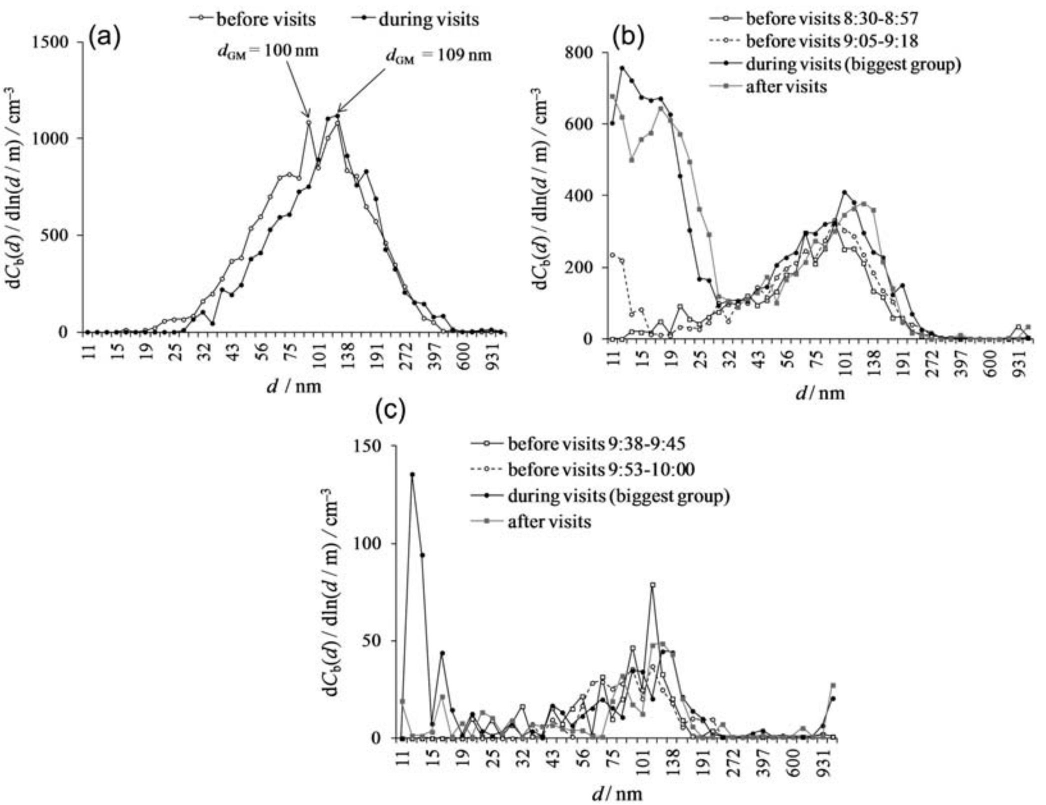

Figure 1. Size distribution ($dC_b(d)/d\ln(d/m)$) of background aerosol at the lowest point in the Postojna Cave in morning hours, before visits and during regular visits (**a**) on 22 February (**b**) on 12 April and (**c**) on 4 June 2011.

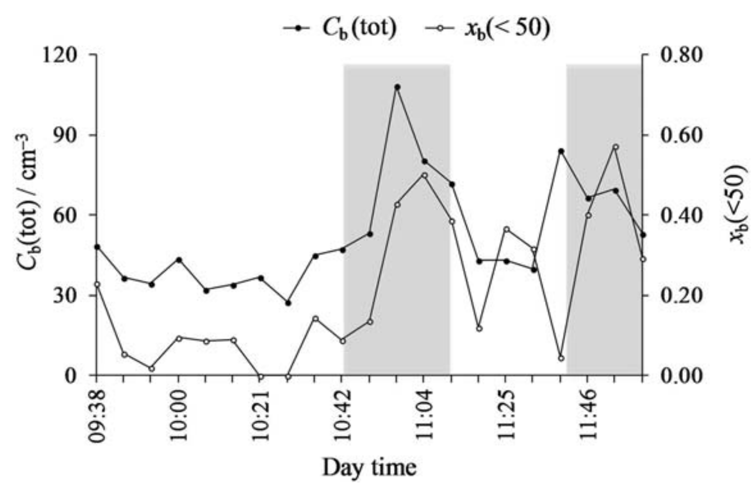

Figure 2. Time run of the total number concentrations of background aerosol (C_b(tot)) and fraction of particles smaller than 50 nm ($x_b(<50)$) at the lowest point in the Postojna Cave in morning hours on 4 June 2011.

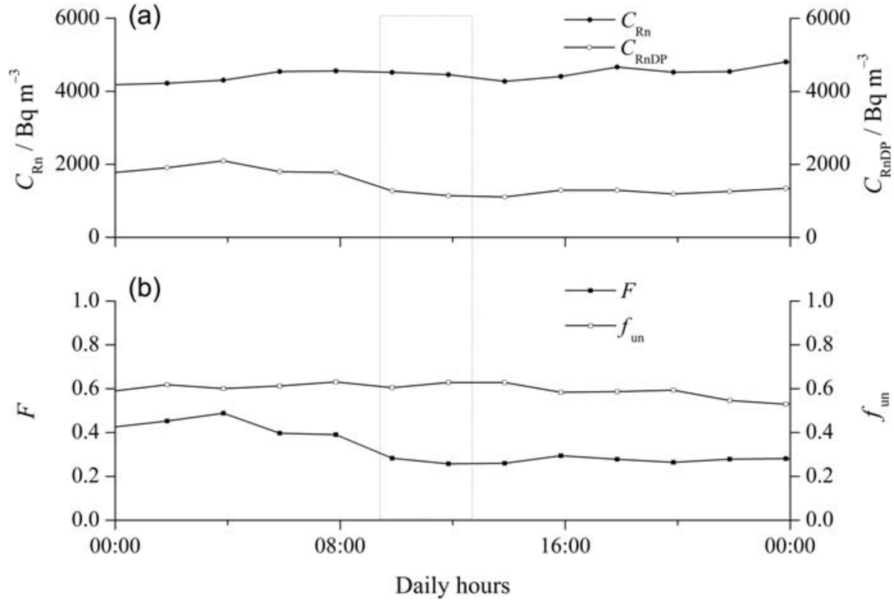

Figure 3. Time variation of (**a**) radon activity concentration (C_{Rn}) and equivalent equilibrium activity concentration of radon decay products (C_{RnDP}), and (**b**) equilibrium factor between radon and its decay products (F) and fraction of unattached decay products (f_{un}) at the lowest point in the Postojna Cave on 4 June 2011.

and flowing through corridors grows colder and water-saturated, which eventually results in water condensation. The surfaces in the cave in warm periods are actually more humid than in cold periods of the year.

CONCLUSIONS

The results have shown that fast changes in aerosol concentration of background aerosol and size distribution are not reflected in changes in the fraction of unattached RnDP, because of the time needed for radon decay products to be created, neutralised and attached. Therefore, the expected inverse relationship between total number concentration and the fraction of unattached RnDP may be observed only if two distinct seasons are compared (e.g. winter and summer), but not two periods within a day. Nonetheless, it would be beneficial to monitor RnDP aerosols with higher frequency, comparable with that of the SMPS+C device. The dependence of the fraction of unattached RnDP on the concentration of smallest particles will be clarified after measurements down to 1 nm have been extended.

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REFERENCES

1. Field, M. S. *Risks to cavers and cave workers from exposures to low-level ionizing alpha radiation from Rn-222 decay in caves.* J. Cave Karst Stud. **69**, 207–228 (2007).
2. Vaupotič, J. *Radon levels in Karst caves in Slovenia.* Acta Carsol. **39**, 503–512 (2010).
3. Hakl, J., Hunyadi, I. and Várhegyi, A. *Radon monitoring in caves.* In: Radon Measurements by Etched Track Detectors. Durrani, S. A. and Ilić, R., Eds. World Scientific Publishing Co. Pte. Ltd. pp. 261–283 (1997).
4. Cigna, A. A. *Radon in caves.* Int. J. Speleol. **34**, 1–18 (2005).
5. Somlai, J., Szeiler, G., Szabo, 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).
6. 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).
7. 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).

8. 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).
9. 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).
10. Hofmann, W. and Koblinger, L. *Monte-Carlo modeling of aerosol deposition in human lungs. Part ll: Deposition fractions ant their sensitivity to parameter variations*. J. Aerosol. Sci. **21**, 675–688 (1990).
11. Iskra, I., Kávási, N. and Vaupotič, J. *Nano aerosols in the Postojna Cave*. Acta Carsol. **39**, 523–528 (2010).
12. Smerajec, M. and Vaupotič, J. *Nano-aerosols including radon decay products in outdoor and indoor air at a suburban site*. J. Toxicol. ID 510876, **2012**, 1–31 (2012).
13. Porstendörfer, J. *Radon: measurements related to dose*. Environ. Int. **22**, S563–S583 (1996).
14. Tokonami, S. *Experimental verification of the attachment theory of radon progeny onto ambient aerosols*. Health Phys. **78**, 74–79 (2000).
15. Nikolopoulos, D. and Vogiannis, E. *Modelling radon progeny concentration variations in thermal spas*. Sci. Total Environ. **373**, 82–93 (2007).
16. 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).
17. Huet, C., Tymen, G. and Boulaud, D. *Long-term measurements of equilibrium factor and unattached fraction of short-lived radon decay products in a dwelling—comparison with PRADDO model*. Aerosol. Sci. Technol. **35**, 553–563 (2001).
18. Tokonami, S., Limoto, T. and Kurosawa, R. *Continuous measurement of the equilibrium factor and the unattached fraction fp of radon progeny*. Environ. Int. **22**, S611–S616 (1996).
19. Vaupotič, J. *Nanosize radon short-lived decay products in the air of the Postojna Cave*. Sci. Total Environ. **393**, 27–38 (2008).