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UNATTACHED FRACTION OF RADON DECAY PRODUCTS AS A CRUCIAL PARAMETER FOR RADON DOSIMETRY IN POSTOJNA CAVE

DELEŽ PROSTIH RADONOVIH RAZPADNIH PRODUKTOV KOT KRITIČEN PARAMETER V DOZIMETRIJI RADONA V POSTOJNSKI JAMI

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Abstract UDC: 546.29:551.44(497.4 Postojna)

Janja Vaupotič & Ivan Kobal: Unattached fraction of radon decay products as a crucial parameter for radon dosimetry in Postojna Cave

Short-term summer and winter monitoring was carried out at the lowest point in Postojna cave, on air concentrations on radon (C_{Rn}) and radon decay products (C_{RnDP}), the equilibrium factor (F) and unattached fraction of radon decay products (f_{un}), barometric pressure (P), relative air humidity in the cave (RH) and air temperature outside (T_{out}) and in the cave (T_{in}), with the emphasis on f_{un}. Dose conversion factors (DCF) for mouth and nasal breathing were calculated from the f_{un} values (ranging from 0.10 to 0.68) and effective doses for the employees in the cave were obtained. These significantly exceed the doses based on the ICRP-65 methodology now in use.

Key words: radon, radon decay products, unattached fraction, dose conversion factors, karstic caves.

Izvleček UDK: 546.29:551.44(497.4 Postojna)

Janja Vaupotič & Ivan Kobal: Delež prostih radonovih razpadnih produktov kot kritičen parameter v dozimetriji radona v Postojnski jami

V letih od 1998 do 2001 smo v zraku Postojnske jame na najnižji točki merili koncentracijo radona (C_{Rn}) in radonovih kratkoživih razpadnih produktov (C_{RnDP}), ravnotežni faktor (F), delež nevezanih radonovih razpadnih produktov (f_{un}), zračni tlak (P), relativno vlažnost zraka v jami (RH) in temperaturo zraka v jami (T_{in}) ter zunaj (T_{out}). Poseben poudarek je bil na f_{un} in na njegovi odvisnosti od vremenskih razmer. Vrednosti f_{un} so bile v širokem razponu, od 0,10 do 0,68. Z uporabo novega dozimetrijskega modela smo na osnovi izmerjenih vrednosti f_{un} izračunali dozne pretvorbene faktorje in ugotovili, da so bili znatno višji od 5 mSv/WLM, to je vrednosti, ki jo priporoča metodologija ICRP-65. Tako so dejanske doze, ki jih prejmejo jamski vodiči, v povprečju za faktor 8 poleti in za faktor 2,7 pozimi višje od vrednosti, ki jih dobimo po dosedanji metodologiji ICRP-65.

Ključne besede: radon, radonovi razpadni produkti, delež nevezanih radonovih razpadnih produktov, dozni pretvorbeni faktor, kraške jame.

INTRODUCTION

A regular radon monitoring has been performed in Postojna Cave since 1995, because high concentrations of the radioactive noble gas radon (^{222}Rn) in air were detected there (Kobal et al. 1978; Kobal et al. 1986; Kobal et al. 1987; Kobal et al. 1988). The monitoring programme was modified several times and eventually optimised by measuring air radon concentration with etched track detectors (Urban and Schmitz, 1993) at the inner railway station and at the lowest point (Vaupotič et al. 1998; Vaupotič et al. 2001; Vaupotič et al. 2002; Vaupotič et al. 2003a). The effective doses received by employees due to working in the radon-rich cave atmosphere, are estimated on the basis of radon concentrations and the time spent in the cave. For that purpose the ICRP-65 methodology (ICRP 1994a) was applied, taking 5 mSv/WLM as dose conversion convention. The doses are reported semi-annually and annually to the Health Inspectorate at the Ministry of Health which then limits the time to be spent by an employee in the next half year. WLM (working-level-month) above means the exposure gained by 170 hours breathing in the air with the activity concentration of short-lived radon decay products of 1 WL, which is defined as ^{218}Po, ^{214}Bi, ^{214}Pb/^{214}Po being in a radioactive equilibrium (F = 1) with 100 pCi/L (3,700 Bqm^{-3}) of ^{222}Rn, resulting in alpha energy concentration of 1.3×10^{5} MeV/L.

This methodology has been applied without any concern until recently when preliminary measurements of the unattached fraction of short-lived radon decay products, the crucial dosimetric parameter (Hofmann et al. 1996a; Hofmann et al. 1996b; James 1988; Porstendörfer 2002), revealed much higher values (Butterweck et al. 1992; Vaupotič et al. 2003b; Vaupotič & Kobal 2004) than 0.03, the value presently included in the ICRP-65 methodology, and hence much higher doses are expected than estimated by the present methodology (Butterweck et al. 1992; Ortega & Vargas 1996; El-Hussein 1996; El-Hussein et al. 1998; Chen et al. 1998; Solomon 2001; Vaupotič & Kobal 2003; Vaupotič & Kobal 2004). In order to check this point, over the period 1998-2001 several 5 to10 day continuous measurements were made on air concentrations of radon (C_{Rn}) and radon decay products (C_{RnDP}), the equilibrium factor (F) and unattached fraction of radon decay products (f_{un}), barometric pressure (P), relative air humidity in the cave (RH) and air temperature outside (T_{out}) and in the cave (T_{in}), with the emphasis on f_{un} at the lowest point.

This paper discusses results of the monitoring, with an emphasis on the temporal variation of f_{un} and its dependence on meteorological parameters and the operational regime of the cave. Effective doses are calculated, based on f_{un}, and compared with doses previously obtained according to the ICRP-65 methodology.

EXPERIMENTAL

Portable EQF3020 and EQF3020-2 devices (manufactured by SARAD, Dresden, Germany) were used to measure concentrations of radon and radon short-lived decay products, equilibrium factor, unattached fraction of decay products, air temperature and relative air humidity (Streil et al. 1996). The sampling and analysing frequency is once in two hours. The two Po isotopes are not distinguished by their alpha energies, but are analysed using a quasi spectroscopy based on measuring the total alpha activity at three appropriately chosen time intervals (Thomas 1970). The devices were in operation for 10-15 days in summer and the same in winter from 1998 to 2001. The instruments were calibrated by the manufacturer on purchase, and have since then been regularly checked at the intercomparison experiments organized annually by the Nuclear Safety Administration at Slovenia's Ministry of the Environment, Spatial Planning and Energy (Križman 2001). The hourly average values of the barometric pressure and the outdoor air temperature at the Postojna meteorological station were obtained from the Meteorology Office of the Slovenian Environmental Agency.

RESULTS AND DISCUSSION

Temporal variations of the parameters under consideration

Results obtained for summer and winter of 1999 are shown in Fig. 1 and Fig. 2, respectively. The patterns of temporal variation of parameters in summer differ substantially from those in winter. While in summer, expected (Vaupotič et al. 2002; Vaupotič et al. 2003a) diurnal fluctuations of C_{Rn}, C_{RnDP}, F and f_{un} are seen, the fluctuations in winter are sporadic, with C_{Rn}, C_{RnDP} and f_{un} showing a similar run. It is also clear that C_{Rn} and C_{RnDP} values are lower in winter (average values being 2074 Bqm^{-3} and 1128 Bqm^{-3}, respectively) than in summer (average values 4536 Bqm^{-3} and 1575 Bqm^{-3}, respectively). This is because of the so called chimney effect which in winter, when the temperature outdoors is lower than in the cave, enhances a natural draught of radon-rich air from the cave through vertical channels into the outdoor atmosphere (Kobal et al. 1988). In summer, the situation is reversed and the draught is minimal, if any. The opposite is true with F, being lower in summer (average value 0.35) than in winter (average value 0.58). The number of visitors is much higher in summer than in winter, thus causing a higher plate-out of RnDP in summer, and thus reducing F.

As seen from Fig.1, diurnal variations of f_{un} are well pronounced in summer, which is not the case in winter (Fig. 2) when these changes seem to follow the C_{Rn}, C_{RnDP} changes, though slightly advanced. Relationships C_{Rn} - f_{un}, C_{RnDP} - f_{un} and F - f_{un} are shown in Fig. 3 for summer and in Fig. 4 for winter. First of all, f_{un} values are regularly much higher in summer than in winter. An expected negative F - f_{un} correlation (Ortega & Vargas 1996; Chen et al. 1998; Paul et al. 1999) is observed, both in summer (Figs. 3c) and in winter (Figs. 4c), though more pronounced in winter. In summer, f_{un} does not seem to be related to C_{Rn} (Figs. 3a) and it slightly decreases with increasing C_{RnDP} (Figs. 3b). On the other hand, f_{un} increases with increasing both C_{Rn} (Figs. 4a) and C_{RnDP} (Figs. 4b) in winter. At this point of the study we still are not able to explain the difference in the C_{Rn} - f_{un} and C_{RnDP} - f_{un} relationships between summer and winter.

Although air temperature and humidity may play an important role in the recombination of radon decay products (Dankelmann et al. 2001; Pagelkopf et al. 2003) these two parameters are practically constant in Postojna Cave, being 8-10 ºC and above 98 %, respectively, all the year round, and therefore their effect on f_{un} has not been sought. On the other side, the dependence of f_{un} on P and T_{out} is presented in Figs. 5 and 6 for summer and winter, respectively. No influence of T_{out} on f_{un} may be determined, either in summer (Fig. 5a) or in winter (Fig. 6a). But a positive P - f_{un} correlation is observed both in summer (Fig. 5b) and in winter (Fig. 6b), though the latter one is less clear. Because low f_{un} values are found in a stagnant air (Morlier et al. 1996) it may be that higher barometric pressure makes the cave air system more dynamic. But this cannot be confirmed without measuring the velocities of air masses through the cave corridors, which are planned in the future.

In Fig. 7, f_{un} values are plotted for every second hour of a day for 7 days of a measurement in summer and in winter. Sometimes in summer (Fig. 7a), f_{un} increases in the morning, reaches a maximum value at around 10 a. m. and then either decreases or remains constant in the afternoon. One could expect f_{un} to increase after the visits have started in the morning with the running of trains and walking of tourists enhancing air movement, and to decrease in the evening when the cave is closed to visitors. Because this does not happen we may assume that the operational regime of the cave has only a minor influence on f_{un}, and the effects of meteorological and microclimatic parameters are predominant. Neither is the influence of the operational regime on f_{un} observed in winter (Fig. 7b).

Dose conversion factors

The ICRP-65 methodology (ICRP 1994a) for estimating doses due to radon and short-lived radon decay products is based on the results of epidemiological studies and recommends as dose conversion convention 4 mSv/WLM at home and 5 mSv/WLM at the workplace. A refined dosimetric approach (Porstendörfer 1996), based on a new respiratory trackt model (ICRP 1994b), has recently been proposed. The short-lived radon decay products are divided in two categories: the unattached short-lived radon decay products with the activity median aerodynamic diameter (AMAD) of 0.8 nm and the attached fraction of 200 nm. Dose conversion factors (DCF in mSv/WLM) are expressed by the unattached fraction, f_{un}, as:

$$DCF_m = 101 \times f_{un} + 6.7 \times (1 - f_{un})$$ for mouth breathing, and

$$DCF_n = 23 \times f_{un} + 6.2 \times (1 - f_{un})$$ for nasal breathing.

Application of this dosimetric model requires much more sophisticated measuring equipment (Reineking & Porstendörfer 1990; El-Hussein & Ahmed 1995; Tokonami et al. 1996; Strong & Baker 1996; Ortega & Vargas 1996; Morlier et al. 1996; Wasiolek et al. 1996; El-Hussein 1996; Chen et al. 1998; Streil et al. 1996; Paul et al. 1999; Leung et al. 2003) than the previous one, ranging from simple screen samplers to effective dosimeters (Solomon 2001). Different measuring techniques were recently compared, and their advantages and disadvantages with respect to the dose estimate commented by Wasiolek and James (Wasiolek & James 2000).

Using the above equations, dose conversion factors were calculated for the periods under investigation, and are collected in Table 1. Generally, DCF_m and DCF_n values are much higher in summer than in winter: their average values are given in the bottom two lines of Table 1. Assuming that a tourist guide's breathing pattern consists of 50 % mouth breathing and 50 % nasal breathing, a combined value is reached of DCF, i.e., $DCF_c = 0.50 \times DCF_m + 0.50 \times DCF_n$. Table 1 displays also DCF_c and the ratio $DCF_c/5$ to facilitate comparison with the ICRP-65 convention value of 5 mSv/WLM. The table indicates an alarming point, that DCF_c values and hence the effective doses of the tourist guides in Postojna Cave, are being significantly underestimated if the present ICRP-65 methodology is followed. On average, the factor of underestimation is 8.0 in summer and about 2.7 in winter. Therefore, a thorough study of f_{un} levels in Postojna Cave and their dependence on meteorological and microclimate parameters as well as on the operational regime of the cave and breathing characteristics of the employees should be performed in order to obtain correct doses.

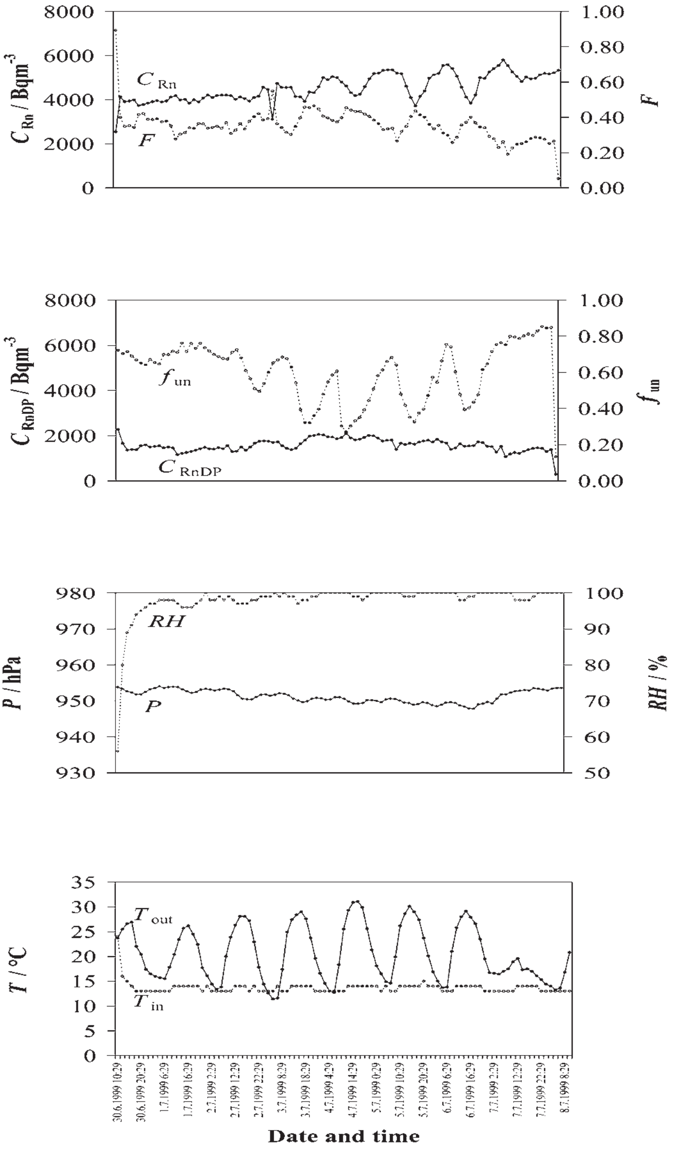

Figure 1: Results of a continuous measurement carried out at the lowest point in the Postojna Cave in the period from 30 June to 8 July 1999: concentrations of radon (C_{Rn}) and radon decay products (C_{RnDP}), equilibrium factor (F), unattached fraction of radon decay products (f_{un}), barometric pressure (P), relative air humidity in the cave (RH), and air temperature in the cave (T_{in}) and outdoor (T_{out}).

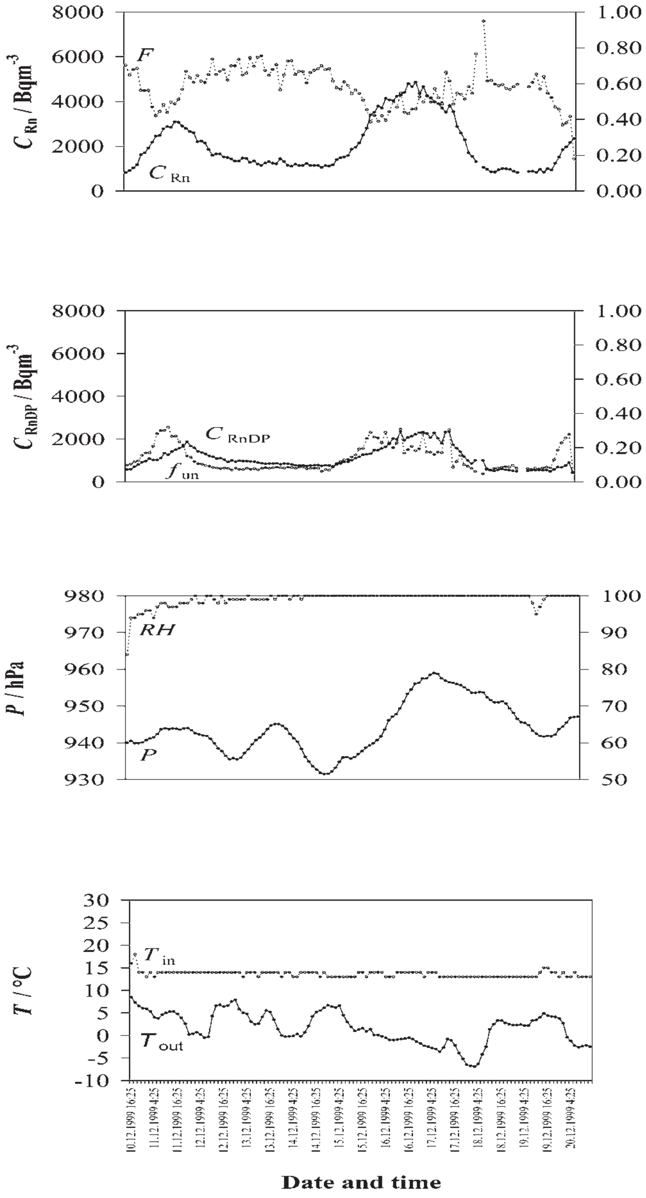

Figure 2: Results of a continuous measurement carried out at the lowest point in Postojna Cave in the period of 10-20 December 1999: concentrations of radon (C_{Rn}) and radon decay products (C_{RnDP}), equilibrium factor (F), unattached fraction of radon decay products (f_{un}), barometric pressure (P), relative air humidity in the cave (RH), and air temperature in the cave (T_{in}) and outdoor (T_{out}).

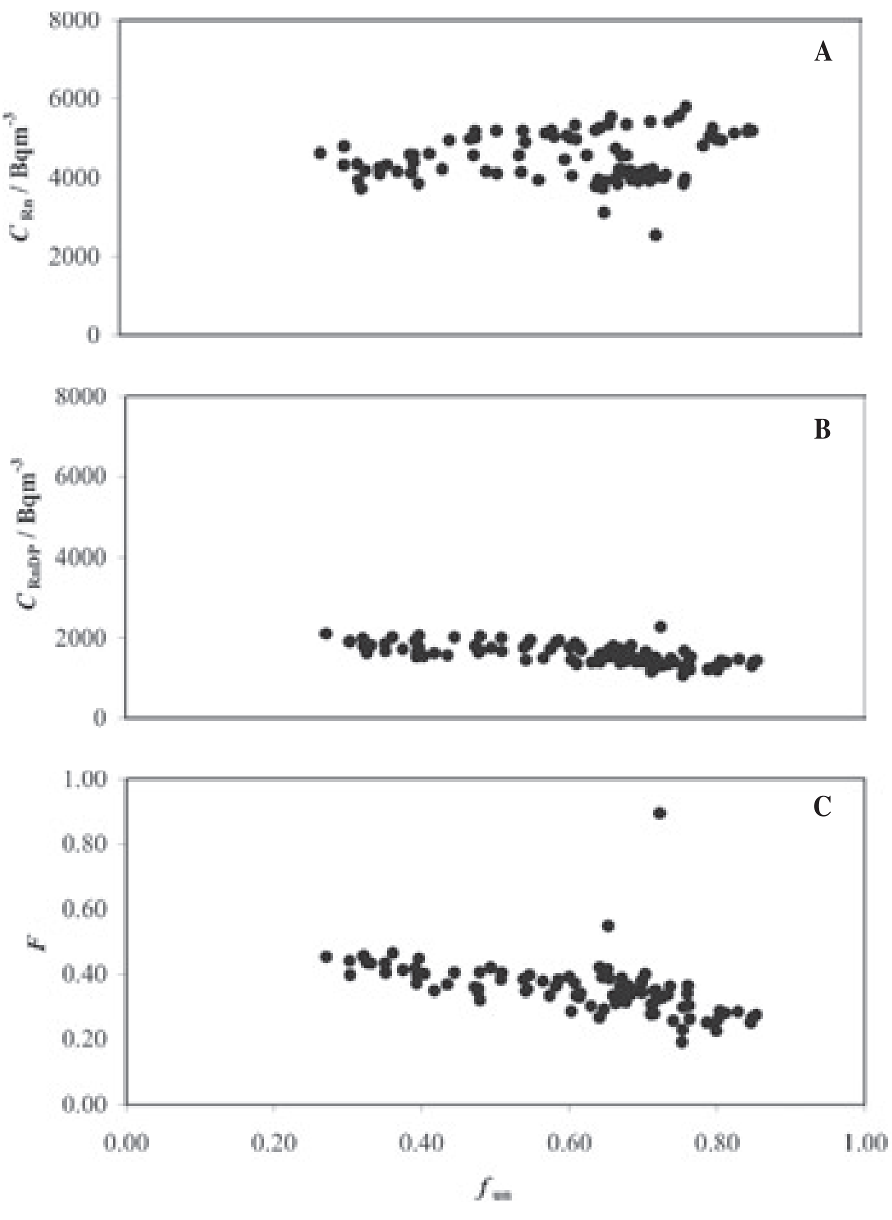

Figure 3: Relationships between f_{un} and a) C_{Rn}, b) C_{RnDP} and c) F for the measurement from 30 June to 8 July 1999.

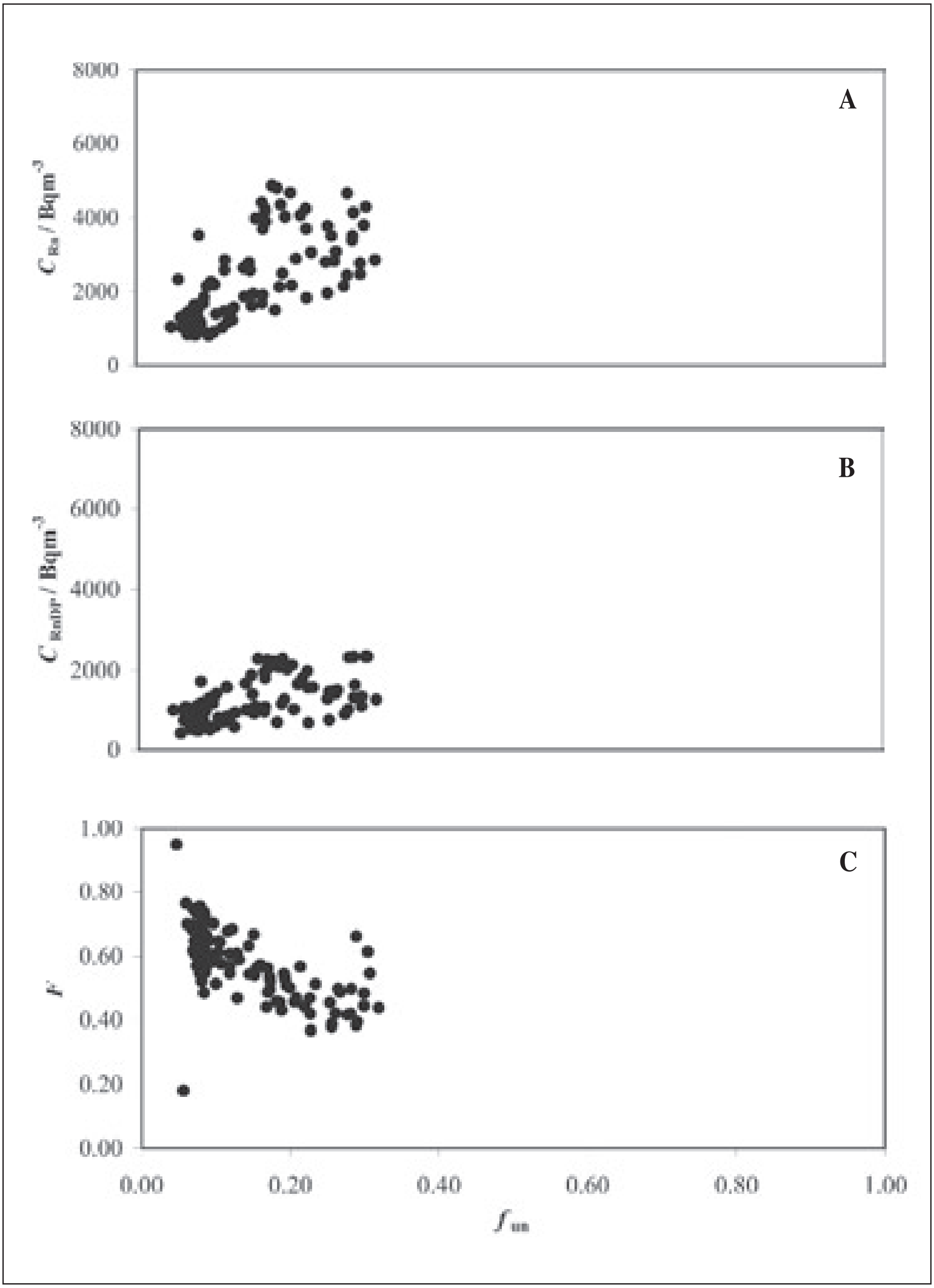

Figure 4: Relationships between f_{un} and a) C_{Rn}, b) C_{RnDP} and c) F for the measurement 10-20 December 1999.

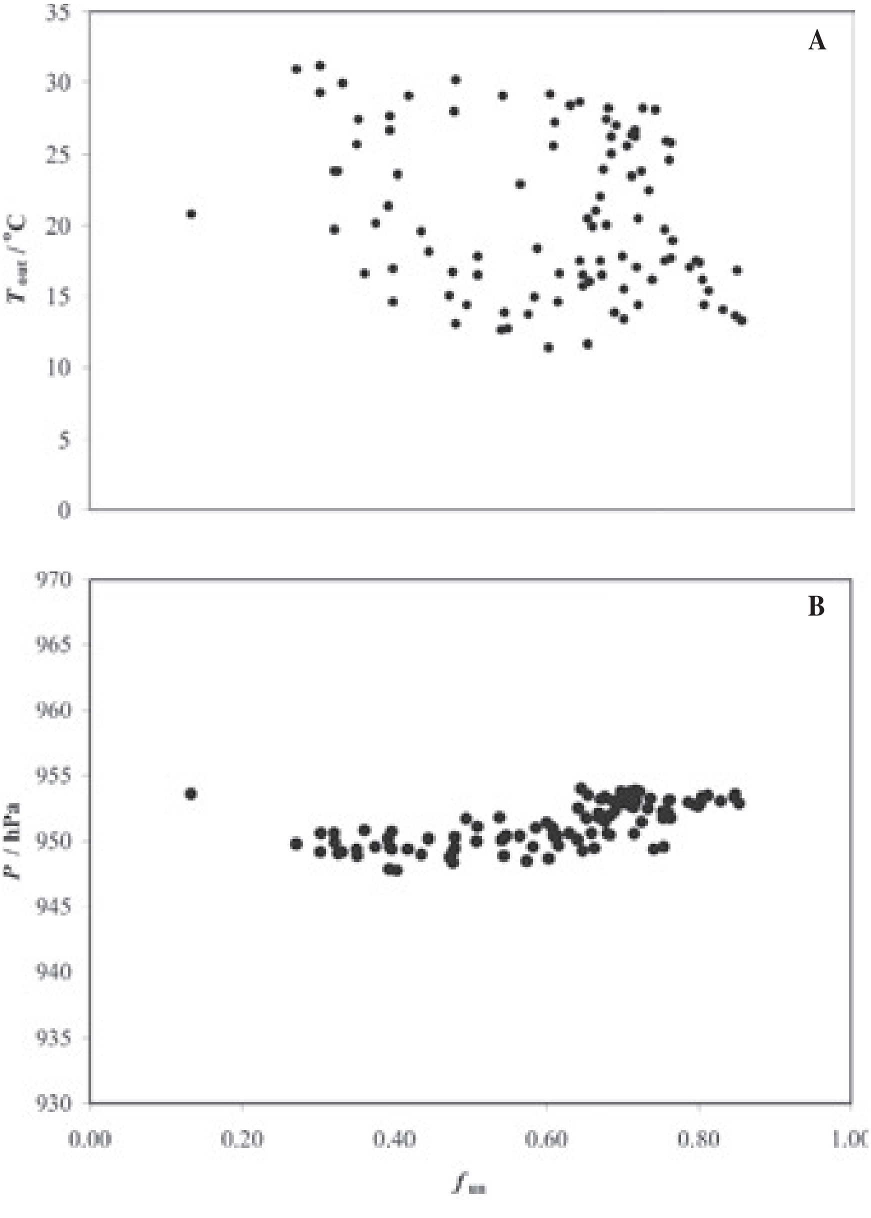

Figure 5: Relationships between f_{un} and a) T_{out}, and b) P for the measurement 30 June to 8 July 1999.

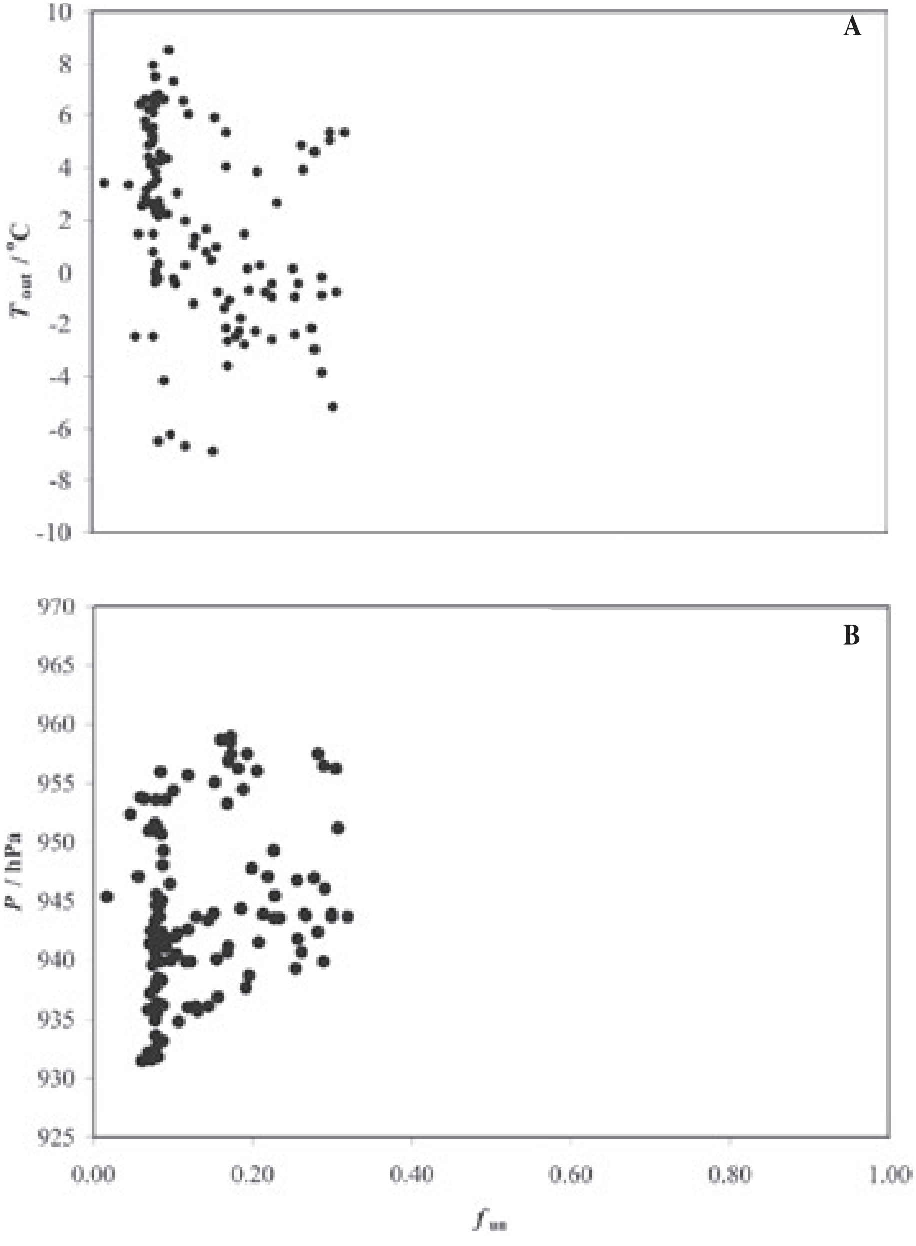

Figure 6: Relationships between f_{un} and a) T_{out}, and b) P for the measurement 10-20 December 1999.

CONCLUSIONS

The fraction of unattached radon short-lived decay products in air at the lowest point in Postojna Cave ranged from 0.10 to 0.12 in winter and from 0.56 to 0.68 in summer. For a tourist guide a combined dose conversion factor was assumed as being expressed by $DCF_c = 0.50 \times DCF_m + 0.50 \times DCF_n$. On average, DCF_c values are 8.0 times in summer and 2.7 times in winter higher than the ICRP-65 convention value of 5 mSv/WLM. Therefore, the present effective doses are by far underestimated. Systematic and thorough measurements will be required, covering longer periods under different meteorological and operational conditions, in order to provide a firm basis for re-evaluating the annual effective doses of cave employees, and most probably a modification of the present radon monitoring programme and present methodology for dose estimation will be needed.

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measuring period	f_{un}	DCF_m	DCF_n	DCF_c	$DCF_c/5$
12.01-19.01, 1998	0.12	19.0	8.4	13.7	2.7
10.08-18.08, 1998	0.58	61.4	15.9	34.7	6.9
14.12-22.12, 1998	0.10	16.1	7.9	12.0	2.4
30.06-08.07, 1999	0.60	63.3	16.3	39.8	8.0
10.12-20.12, 1999	0.14	19.9	8.6	14.3	2.9
03.07-18.07, 2000	0.56	59.5	15.5	37.5	7.5
19.07-03.08, 2001	0.68	70.1	17.6	43.9	8.8
summer - average	0.61	63.4	16.3	40.0	8.0
winter - average	0.12	18.3	8.3	13.3	2.7

Table 1: Dose conversion factors (DCF in mSv/WLM) for mouth (m) and nasal (n) breathing calculated from the unattached fraction of radon short-lived decay products, f_{un}, measured at the lowest point in Postojna Cave; also given are combined values $DCF_c = 0.50 \times DCF_m + 0.50 \times DCF_n$, as assumed for a tourist guide.

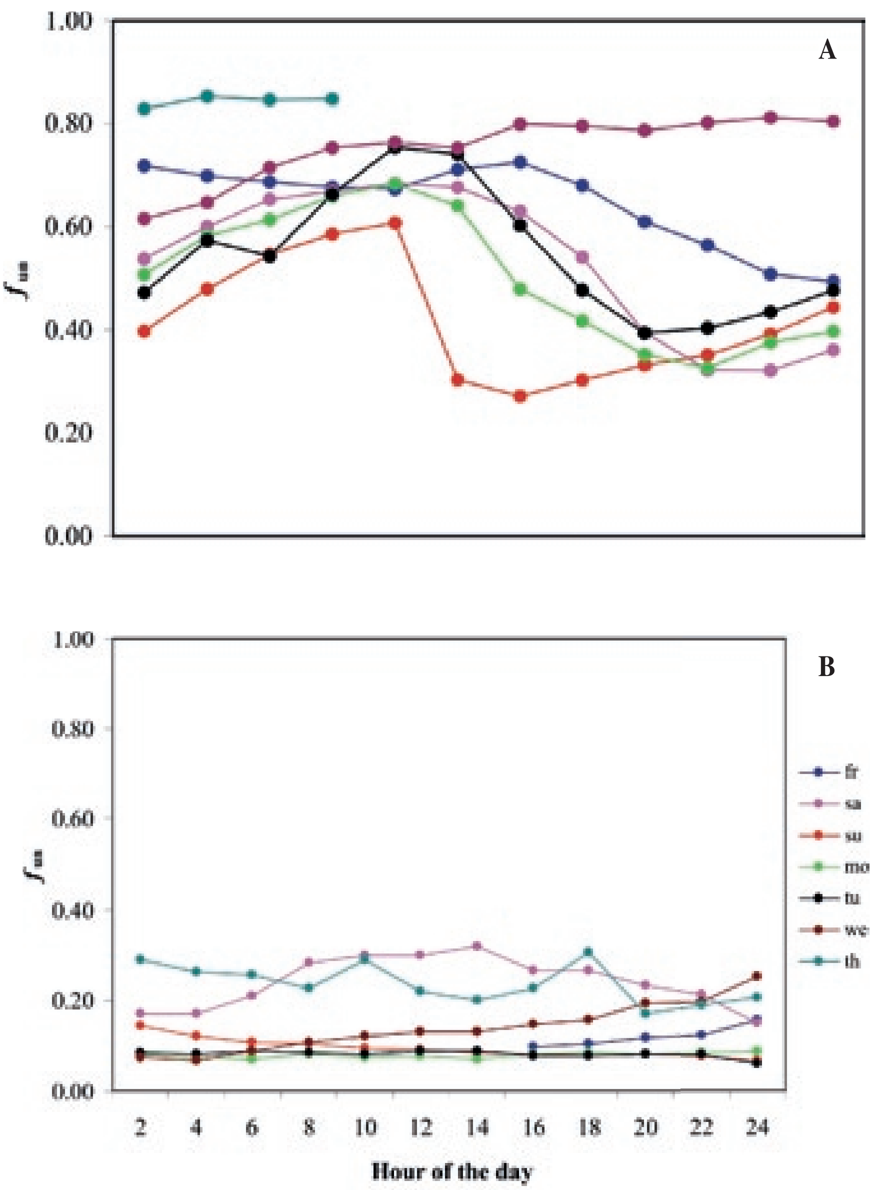

Figure 7: f_{un} values for every second hour of a day for 7-day period of the measurement a) 30 June to 8 July 1999 and b) 10-20 December 1999.

REFERENCES

Chen, C.-J. & C.-C. Liu & Y.-M. Lin, 1998. Measurement of equilibrium factor and unattached fraction of radon progeny in Kaohsiung, Taiwan.- Appl. Radiat. Isot. 49, 1613-1618.

Butterweck, G. & J. Porstendörfer & A. Reineking & J. Kesten, 1992. Unattached fraction and the aerosol size distribution of the radon progeny in a natural cave and mine atmosphere.- Radiat. Prot. Dosim. 45, 167-170.

Dankelmann, V. & A. Reineking, & J. Porstendörfer, 2001. Determination of neutralisation rates of ^{218}Po ions in air.- Radiat. Prot. Dosim. 94, 353-357.

El-Hussein, A. & A.A. Ahmed, 1995. Unattached fraction and size distribution of aerosol-attached radon progeny in the open air.- Appl. Radiat. Isot. 46, 1393-1399.

El-Hussein, A., 1996. Unattached fraction, attachment and deposition of radon progeny in indoor air.- Appl. Radiat. Isot. 47, 515-523.

El-Hussein, A. & A.A. Ahmed & A. Mohammed, 1998. Radiation dose to human respiratory trackt from inhalation of radon-222 and its progeny.- Appl. Radiat. Isot. 49, 783-790.

Hofmann, W. & T. Heistracher & I. Balásházy, 1996a. Deposition patterns of inhaled radon decay products in human bronchial airway bifurcation.- Environ. Int. 22 (Suppl. 1), S935-S940.

Hofmann, W. & M. Mainelis & A. Mohamed & I. Balásházy & J. Vaupotič & I. Kobal, 1996b. Comparison of different modeling approaches in current lung dosimetry models.- Environ. Int. 22 (Suppl. 1), S965-S976.

International Commission on Radiological Protection (ICRP). 1994a. Protection Against Radon-222 at Home and at Work: ICRP Publication 65, Pergamon Press: Oxford, 1994, pp 1-262.

International Commission on Radiological Protection (ICRP). 1994b. Human Respiratory Trackt Model for Radiological Protection. ICRP Publication 66, Pergamon Press: Oxford, 1994, pp 1-482.

James, A.C., 1988. The basis for health concern. In: Radon and its Decay Products in Indoor Air (Eds. W.W. Nazaroff & A.V. Nero, Jr), John Wiley & Sons, New York, pp 259-309.

Kobal, I. & M. Škofljanec, & D. Zavrtanik, 1978. Radon in Slovene karst caves.- Naše Jame 20, 41-47.

Kobal, I. & B. Smodiš & M. Škofljanec, 1986. Radon-222 air concentrations in the Slovenian karst caves of Yugoslavia.- Health Phys. 50, 830-834.

Kobal, I. & B. Smodiš & J. Burger & M. Škofljanec, 1987. Atmospheric ^{222}Rn in tourist caves of Slovenia, Yugoslavia.- Health Phys. 52, 473-479.

Kobal, I. & M. Ančik & M. Škofljanec, 1988. Variations of ^{222}Rn air concentration in Postojna Cave.- Radiat. Prot. Dosim. 25, 207-211.

Križman, M., 2001. Report on the Intercomparison Experiment for Radon and Progeny in Air.- URSJV RP 47/2001. Nuclear Safety Administration, Ljubljana, 2001;1-7.

Leung, J.K.C. & M.Y.W. Tso & J.H.C. Lam & Q.F. Zhau, 2003. Direct measurement of attachment of ^{220}Rn progeny on aerosols by atomic force microscopy.- Nucl. Instr. Methods Phys. Res. 508, 512-518.

Morlier, J.P. & M. Bisson & P. Fritsch & G. Monchaux & M. Janot & J.F. Pineau, 1996. Deposition of ^{214}Pb and nuclear aberration in respiratory trackt of rats after exposure to radon progeny under different aerosol conditions.- Environ. Int. 22 (Suppl. 1), S927-S930.

Ortega, X. & A. Vargas, 1996. Characteristics and temporal variation of airborne radon decay progeny in the indoor environment in Catalonia (Spain).- Environ. Int. 22 (Suppl. 1), S149-S159.

Pagelkopf, P. & J. Porstendörfer, 2003. Neutralisation rate and the fraction of the positive ^{218}Po-clusters in air.- Atmos. Environ. 37, 1057-1064.

Paul, A. & S. Röttger & A. Honig & T. Sulima & A. Buchholz & U. Keyser, 1999. Measurement of short-lived radon progenies by simultaneous $\alpha\gamma$ spectrometry at Geraman radon reference chamber.- Nucl. Instr. Methods Phys. Res. A 434, 303-312.

Reineking, A. & J. Porstendörfer, 1990. Unattached fraction of short-lived Rn decay products in indoor and outdoor environments: an improved single-screen method and results.- Health Phys. 58, 715-727.

Porstendörfer, J., 1996. Radon: Measurements related to dose.- Environ. Int. 22 (Suppl. 1), S563-S583.

Porstendörfer, J., 2002. Influence of physical parameters on doses from radon exposures.- Int. Congr. Ser. 1225, 149-160.

Solomon, S.B., 2001. Field tests of a radon progeny sampler for the determination of effective dose.- Sci. Total Environ. 272, 303-313.

Streil, T. & G. Holfeld & V. Oeser & C. Feddersen & K. Schönefeld, 1996. SARAD EQF 3020: A new microsystem based monitoring system for the continous measurement of radon and the attached and unattached fraction of the radon progeny. In: IRPA (International Radiological Protection Association) Regional Congress on Radiation Protection in Neighbouring Countries in Central Europe, Portorož, 1996, pp 334-337.

Strong, J.C. & S.T. Baker, 1996. Lung deposition in rodents during exposure to attached radon progeny.- Environ. Int. 22 (Suppl. 1), S905-S908.

Thomas, J.W., 1970. Measurements of radon daughters in air.- Health Phys. 19, 691-789.

Tokonami, S. & T. Iimoto & R. Kurosawa, 1996. Continuous measurement of the equilibrium factor and the unattached fraction f_p of radon progeny in the environment.- Environ. Int. 22 (Suppl. 1), S611-S616.

Urban, M. & J. Schmitz, 1993. Radon and radon daughters metrology: Basic aspects.- In: Fifth International Symposium on the Natural Radiation Environment, Tutorial Sessions.- Report EUR 14411 EN 1993;151-183.

Vaupotič, J. & P. Dujmović & I. Kobal, 1998. Radiation doses due to radon and progeny in Postojna Cave.- Acta Carsol. 27, 395-406.

Vaupotič, J. & I. Csige & V. Radolić & I. Hunyadi & J. Planinić & I. Kobal, 2001. Methodology of radon monitoring and dose estimates in Postojna Cave, Slovenia.- Health Phys. 80, 142-147.

Vaupotič, J. & P. Dujmović & I. Kobal, 2002. Doses due to radon and progeny received by workers in the Postojna Cave (from January 1, 2001 to December 31, 2001).- Jožef Stefan Institute Report IJS-DP-8537, February/2002.

Vaupotič, J. & P. Dujmović & I. Kobal, 2003a. Doses due to radon and progeny received by workers in the Postojna Cave (from January 1, 2002 to December 31, 2002).- Jožef Stefan Institute Report IJS-DP-8739, March/2003.

Vaupotič, J. & I. Kobal, 2003b. Radon doses based on alpha spectrometry. In: Book of Abstracts of the 13th International Symposium "Spectroscopy in Theory and Practice". Nova Gorica, August, 2003. O-2, pp 48

Vaupotič, J., & I. Kobal, 2004. Radon doses based on alpha spectrometry.- Acta Chim. Slovenica, 51.. 50, 159-168

Wasiolek, P.T. & A.C. James & F. Yarger, 1996. Measurements of unattached fraction of radon progeny with two different methods.- J. Aerosol Sci. 27 (Suppl. 1), S465-S466.

Wasiolek, P.T. & A.C James, 2000. Unattached fraction measuring techique and radon lung dose.- J. Environ. Radioact. 51, 137-151.