Coll. Antropol. **32** (2008) Suppl. 2: 99–104
Original scientific paper

Levels of Nanosize Radon Decay Products in Indoor Air: A Comparison for Different Environments

Janja Vaupotič

Radon Center, Department of Environmental Sciences, »Jožef Stefan« Institute, Ljubljana, Slovenia

ABSTRACT

The fraction of unattached (f_{un}) nanosized ^{222}Rn short-lived decay products in indoor air has been measured in different environments and the following ranges obtained: 0.03–0.25 in kindergartens, 0.03–0.18 in schools, 0.08–0.20 in wineries, and 0.12–0.68 in a karst cave. The dependence of f_{un} on the environmental conditions (air temperature, relative humidity and pressure), as well as on the working regime has been studied and will be discussed. Applying the dosimetric approach, dose conversion factors were calculated, based on the measured f_{un} values, and compared to the values, obtained from epidemiological studies and currently recommended to be used in radon dosimetry.

***Key words**: radon, decay products, unattached fraction, indoor air, dose conversion factors*

Introduction

The radioactive noble gas radon (^{222}Rn isotope, α decay, $t_{1/2}$ = 3.82 days) is always accompanied by its short-lived decay products (RnDP): ^{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 after neutralization appear in air as nanosize clusters, the so-called unattached RnDP (in the size range 0.5–3 nm), which sooner or later, depending on the environmental conditions (air temperature, humidity, concentration and size distribution of aerosols), attach to aerosols and form attached RnDP in the size range between 200 and 800 nm[1]. Because of plate-out of aerosols on the walls and floor of a room, as well as air movement and entry of fresh air, radioactive equilibrium between RnDP and Rn in indoor air is only partly achieved and is expressed as a fraction between 0 and 1, called the equilibrium factor[2], *F*.

It has become well known that, on the world wide average, breathing air with radon short-lived decay products contributes more than half to the effective dose a member of the general public receives from all natural radioactive sources of ionizing radiation, and that they are a major cause of lung cancer, second only to cigarette smoking[3]. To quantify the detrimental effects of Rn and/or RnDP to the human health, the so called dose conversion factor (*DCF*) is required, which relates the exposure to RnDP (expressed either in WLM or Bq m^{-3} h) and effective dose (expressed in mSv). The old but still widely used unit, 1 WLM (working-level-month) is the exposure resulting from 170 hours breathing air with an activity concentration of short-lived radon decay products of 1 WL (working-level). 1 WL[2] was originally defined as the activity concentrations of RnDP which are in radioactive equilibrium (F = 1) with 100 pCi L^{-1} (3700 Bq m^{-3}) of ^{222}Rn, resulting in a potential alpha energy concentration of 1.3 × 10^{8} MeV m^{-3}. *DCF* values have been obtained from epidemiologic studies on uranium miners. At present, the International Commission for Radiological Protection (ICRP) in Publication 65[4] recommends 5 mSv WLM^{-1} for working and 4 mSv WLM^{-1} for living environments. The epidemiology-based *DCF* will be hereafter in the text denoted by DCF_{E}.

Birchall and James[5] elaborated a dosimetric approach to calculate the dose conversion factor based on f_{un}; it will be hereafter in the text denoted by DCF_{D}. Their 'best estimate' of DCF_{D} for the indoor air conditions in dwellings was 15 mSv WLM^{-1}. It thus appears that DCF_{D} is 3-fold

Received for publication July 30, 2008

higher than DCF_E = 5 mSv WLM^{-1}. The discrepancy between DCF_D and DCF_E values has not been fully clarified, the most probable reason originating in too high a value being chosen for the radiation weighting factor[6], w_α. In addition, Porstendörfer[7] has shown that DCF_D can be calculated separately for mouth (DCF_{Dm}) and nasal (DCF_{Dn}) breathing, using empirical equations:

$$DCF_{Dm} = 101 \times f_{un} + 6.7 \times (1 - f_{un}) \qquad (1)$$

$$DCF_{Dn} = 23 \times f_{un} + 6.2 \times (1 - f_{un}) \qquad (2)$$

In this paper, the results of our studies on f_{un} in 29 rooms of kindergartens, 26 rooms of elementary and high schools, 4 rooms in wineries and at two points in the Postojna Cave are reported. DCF_D values, calculated based on the Porstendörfer formulae (Eqs. 1 and 2), are discussed and compared with the DCF_E value recommended by ICRP-65.

Materials and Methods

Measurement sites

Radon was surveyed in the indoor air of practically all the kindergartens (730) and schools (890) in Slovenia, within the national radon program, between 1990 and 1994[8,9]. For the study of f_{un}, those kindergartens and schools were selected in which elevated radon levels had been observed or were expected, based on geology[10]. Measurements, lasting for 1–2 weeks at each place, were carried out in 29 rooms of 13 kindergartens (in the period from March 1998 to February 2001) and in 26 rooms of 16 schools (in the period from March 1998 to April 2001).

Because of the elevated radon levels, permanent radon monitoring at the lowest point and the railway station in the Postojna Cave was introduced in 1995[11]. Based on the radon concentrations measured, working time for the employees in the cave is limited in order to keep their effective doses acceptably low. Measurements of f_{un}, lasting for 1–3 weeks were carried out at two places, at the lowest point (LP) of the cave in summer (when radon levels are highest: August 10–18, 1998; June 30 – July 8, 1999; July 19 – August 3, 2001) and in winter (when radon levels are lowest: December 14–22, 1998; December 10–20, 1999), and at the railway station (RS) in summer (June 3–18, 2001).

In the context of the radon survey in underground premises of eight major Slovene wineries, f_{un} was monitored for 1–2 weeks in the period from April to July, 2002.

Measuring technique

Portable SARAD EQF3020 and EQF3020-2 devices (SARAD, Dresden, Germany) were used. Air is pumped for 6 min at a flow rate of 2.4 dm^3 min^{-1} over a metal mesh grid where unattached and attached RnDP are separated and then deposited electrostatically on two separate 150 mm^2 semiconductor detectors. Applying the Markov method[12], the device gives Rn concentration in Bq m^{-3} (C_{Rn}), and, separately for the unattached and attached form of RnDP, the following: individual concentrations in Bq m^{-3} (C_{218Po}, C_{214Pb}, $C_{214Bi} = C_{214Po}$), equilibrium equivalent activity concentration in Bq m^{-3} (defined as: $EEC_{RnDP} = C_{RnDP} = 0.1065C_{218Po} + 0.515C_{214Pb} + 0.379C_{214Bi}$), potential alpha energy concentration in MeV m^{-3} (defined as: $PAEC = 3690C_{218Po} + 17830C_{214Pb} + 13120C_{214Bi}$), F (defined as $F = PAEC/(34640\ C_{Rn})$, and f_{un} (defined as $f_{un} = PAEC_{un}/(PAEC_{un} + PAEC_{att})$, with subscripts 'un'– unattached and 'att' – attached)[13].

The data was transferred to a personal computer in the laboratory for evaluation. Origin 6.1 Data Analysis and Graphing Software was used for statistical data evaluation and presentation.

Results

Diurnal variations of C_{Rn}, C_{RnDP}, F and f_{un} for a selected school are presented in Figure 1. Values of C_{Rn}, C_{RnDP} are highest during the night, decrease during a working day and gain higher values again during the night. Values are kept constantly higher during weekends, e. g., from 14.4. to 16.4. High values of F are accompanied by low values of f_{un}. Similar situations were observed in all kindergartens and schools, although the absolute values of the measured parameters were different and so were the amplitudes of daily fluctuations. Table 1 shows arithmetic mean (AM) and arithmetic standard deviation values (ASD) of the measured parameters, calculated for the whole period of measurements.

The arithmetic mean values of the measured parameters resulting from continuous measurements in the

TABLE 1
INDOOR AIR RADON CONCENTRATION (C_{Rn}), EQUILIBRIUM FACTOR BETWEEN RADON AND RADON DECAY PRODUCTS (F) AND UNATTACHED FRACTION OF RADON DECAY PRODUCTS (f_{un}) IN SLOVENIAN SCHOOLS AND KINDERGARTENS, WITH THE NUMBER OF VALUES (n), ARITHMETIC MEAN (AM), ARITHMETIC STANDARD DEVIATION (ASD), t-VALUE (t) AND p-VALUE (p) OF THE t-TEST

		n	AM	ASD	t	p	
C_{Rn}	schools	26	1362	1718	2.372	0.021	significantly
	kindergartens	29	584	394			different
F	schools	26	0.61	0.15	2.024	0.048	significantly
	kindergartens	29	0.53	0.15			different
f_{un}	schools	26	0.13	0.03	2.114	0.039	significantly
	kindergartens	29	0.15	0.06			different

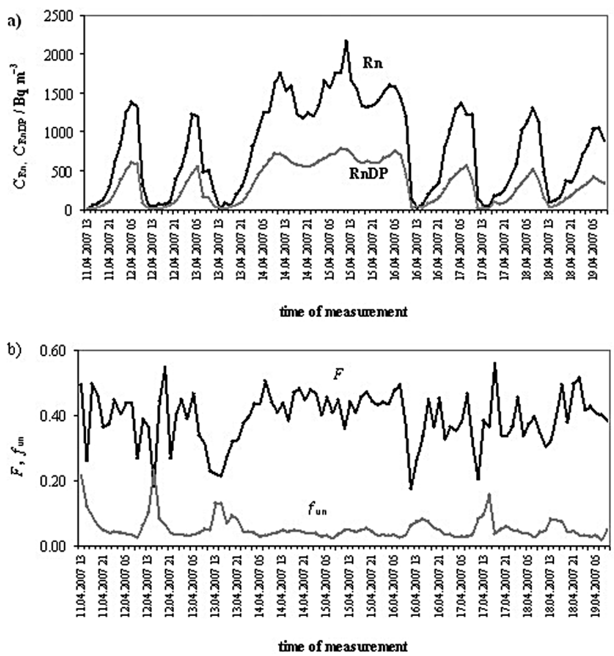

Fig. 1. Results of a continuous monitoring in a selected school, showing diurnal variations of concentrations of radon (Rn, C_{Rn}) and radon short-lived decay products (RnDP, C_{RnDP}), equilibrium factor between Rn and RnDP (F) and unattached fraction of radon decay products (f_{un}).

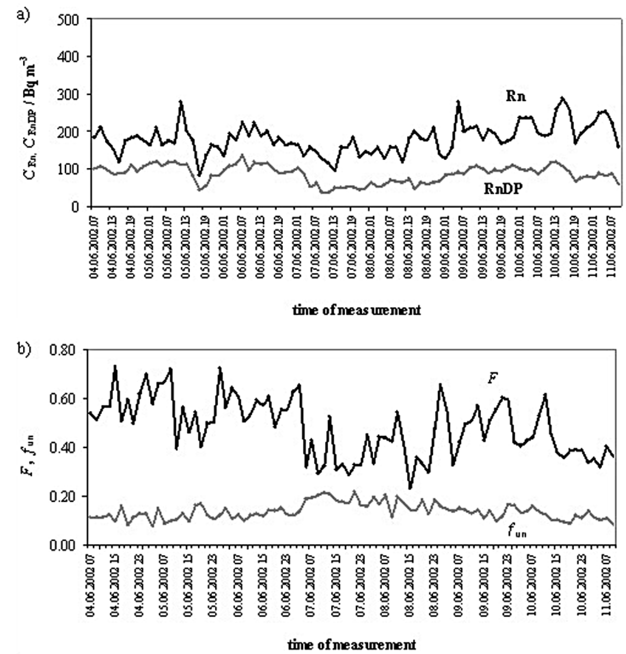

Fig. 2. Results of a continuous monitoring in a selected winery, showing diurnal variations of concentrations of radon (Rn, C_{Rn}) and radon short-lived decay products (RnDP, C_{RnDP}), equilibrium factor between Rn and RnDP (F) and unattached fraction of radon decay products (f_{un}).

Postojna Cave are listed in Table 2. C_{Rn} values were higher in summer than in winter at the lowest point, and higher at the lowest point than at the railway station. On the other hand, F was lower in summer than in winter at the lowest point and lower at the lowest point than at the railway station in summer.

Results of continuous measurements in a selected winery are shown in Figure 2. In contrast to kindergartens and schools, no typical diurnal fluctuations have been observed in wineries. Arithmetic mean values of C_{Rn}, F and f_{un} are collected in Table 3.

Discussion

Figure 3 shows that C_{Rn} values both in kindergartens and schools fit well lognormal distribution. Table 1 displays results of the t-test comparison of the arithmetic mean values of C_{Rn}, F, and f_{un} for the whole period of measurement. Radon levels are significantly higher in schools than in kindergartens. The probable reason lies in the age of buildings. While the majority of kindergartens were built after 1980, the school buildings are generally older. Here, due to aging, the basic concrete slab has numerous cracks and fissures and is no longer a sufficient barrier to radon. Further, there is very often no concrete slab at all in old buildings.

The difference in F levels in schools and in kindergartens most probably reflects the different working regimes at the two places. While in a classroom in a school the movement of students is minimal during class hours, children move more freely in the play room of a kindergarten, causing air movement and hence increasing pla-

TABLE 2
THE POSTOJNA CAVE: AVERAGE VALUES (ARITHMETIC MEAN AND ARITHMETIC STANDARD DEVIATION) OF RADON CONCENTRATION (C_{Rn}), EQUILIBRIUM FACTOR BETWEEN RADON AND RADON DECAY PRODUCTS (F) AND UNATTACHED FRACTION OF RADON DECAY PRODUCTS (f_{un}), MEASURED DURING DIFFERENT PERIODS AT THE LOWEST POINT (LP) AND RAILWAY STATION (RS)

Season, year (site)	C_{Rn} / Bq m^{-3}	F	f_{un}
Summer, 1998 (LP)	4090 ± 440	0.34 ± 0.06	0.58 ± 0.14
Winter, 1998 (LP)	1470 ± 1060	0.56 ± 0.10	0.10 ± 0.07
Summer, 1999 (LP)	4540 ± 600	0.35 ± 0.09	0.60 ± 0.16
Winter, 1999 (LP)	2070 ± 1160	0.58 ± 0.11	0.14 ± 0.08
Summer, 2001 (LP)	5900 ± 620	0.35 ± 0.07	0.68 ± 0.11
Summer, 2001 (RS)	3330 ± 370	0.63 ± 0.13	0.15 ± 0.05

TABLE 3
AVERAGE VALUES OF RADON CONCENTRATION (C_{Rn}), EQUILIBRIUM FACTOR BETWEEN RADON AND RADON DECAY PRODUCTS (F) AND UNATTACHED FRACTION OF RADON DECAY PRODUCTS (f_{un}) OBTAINED IN SLOVENIAN WINERIES, WITH THE RESULTING DOSE CONVERSION FACTORS FOR NASAL BREATHING (DCF_{Dn})

Winery	Date in 2002	C_{Rn} / Bq m^{-3}	F	f_{un}	DCF_{Dn} / mSv WLM^{-1}	DCF_{Dn}/5
01-S-05	23.04.–13.05.	91 ± 20	0.25 ± 0.08	0.20 ± 0.06	9.56	1.91
04-P-02	30.05.–14.06.	182 ± 37	0.49 ± 0.14	0.12 ± 0.04	8.22	1.64
06-L-02	06.06.–03.07.	998 ± 104	0.63 ± 0.16	0.08 ± 0.02	7.54	1.51
07-G-02	20.06.–04.07.	360 ± 46	0.48 ± 0.06	0.09 ± 0.02	7.71	1.54

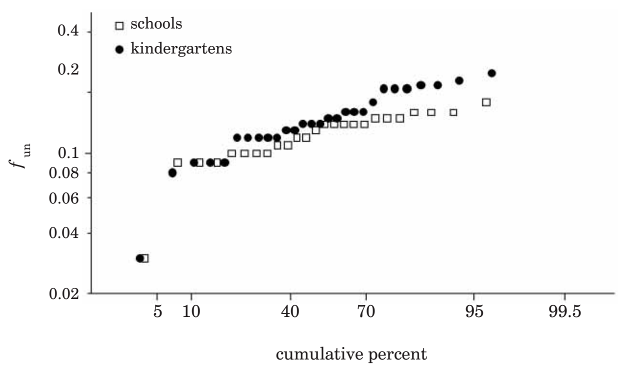

Fig. 3. Lognormal plots of the unattached fraction of radon decay products (f_{un}) in Slovenian schools and kindergartens.

te-out of aerosol and, consequently, lowering F. As expected[14,15], high F values are accompanied by low f_{un} values. The fact that f_{un} values in schools were lower than in kindergartens was therefore expected (Table 1).

Following the ICRP-65 criteria[16], the personnel in kindergartens and schools can be considered as nasal breathers and therefore for them $DCF_D = DCF_{Dn}$. If the arithmetic means of f_{un} for kindergartens and schools (Table 1) are used in Eq. 2, values of 8.72 mSv WLM^{-1} and 8.38 mSv WLM^{-1}, respectively, are obtained, being higher by factors of 1.74 and 1.68, respectively, than the ICRP recommended value of 5 mSv WLM^{-1} for workplaces.

In the Postojna Cave, the patterns (not shown here) of temporal variations in summer differ substantially from those in winter, with C_{Rn} and C_{RnDP} values being lower in winter than in summer. This is because of the so called chimney effect: in winter, the temperature outdoor is lower than in the cave, thus causing a natural draught of air from the cave through vertical channels into the outdoor atmosphere. In summer, the situation is reverse and this draught is minimal, if any. The opposite is true with F, which is lower in summer and higher in winter. The number of visitors is much higher in summer than in winter, thus causing a higher plate-out of RnDP and reducing F. Walking visitors cause more air movement in the narrow corridor at the lowest point than in the big hall at the railway station. Therefore, plate-out of RnDP is enhanced and F reduced, the effect being more pronounced in summer than in winter, and more at the lowest point than at the railway station. f_{un} values are higher in summer than in winter at the lowest point, and higher at the lowest point than at the railway station in summer. Because of the much larger number of visitors in summer than in winter, the cave air is much more disturbed in summer, resulting in enhanced f_{un} values in summer[17]. Similarly, the air is more disturbed by visitors in narrow corridors at the lowest point than in the big hall at the railway station, resulting in enhanced f_{un} at the lowest point[17]. f_{un} values in the cave are much higher than in kindergartens and schools. One of the reasons for the high values is the very low concentration of aerosols in Postojna Cave, being only (1.9–4.3) × 10^9 m^{-3}, as compared to a barite mine with (200–1600) × 10^9 m^{-3} and f_{un} in the range from 0.003 to 0.008[1]. Another reason for elevated f_{un} values could be the intensive washout of aerosols at practically 100 % relative humidity of the cave air, as observed in some spas[18].

Based on the arithmetic mean values of f_{un}, DCF_{Dm} (Eq. 1) and DCF_{Dn} (Eq. 2) values were calculated for the

TABLE 4
DOSE CONVERSION FACTORS FOR MOUTH (DCF_{DM}) AND NASAL (DCF_{DN}) BREATHING CALCULATED, USING EQS. 2 AND 3, FROM THE F_{UN} VALUES IN TABLE 2 FOR THE LOWEST POINT (LP) AND RAILWAY STATION (RS) IN THE POSTOJNA CAVE IN DIFFERENT SEASONS

Season, year (site)	f_{un}	DCF_{Dm} / mSv WLM^{-1}	DCF_{Dm}/5	DCF_{Dn} / mSv WLM^{-1}	DCF_{Dn}/5
Summer, 1998 (LP)	0.58	61.4	12.3	15.9	3.2
Winter, 1998 (LP)	0.10	16.1	3.2	7.9	1.6
Summer, 1999 (LP)	0.60	63.3	12.7	16.3	3.3
Winter, 1999 (LP)	0.14	19.9	4.0	8.6	1.7
Summer, 2001 (LP)	0.68	71.5	14.3	18.2	3.6
Summer, 2001 (RS)	0.15	20.9	4.2	8.7	1.7

lowest point in summer and winter, and in summer at the lowest point and railway station (Table 4). According to the ICRP criteria[16] only maintenance workers engaged in hard physical work in the cave may be considered as mouth breathers, and for them $DCF_D = DCF_{Dm}$, while for all others (i. e., tourist guides, souvenir vendors, locomotive drivers) $DCF_D = DCF_{Dn}$. Table 4 also displays factors by which DCF_{Dm} and DCF_{Dn} values exceed the ICRP recommended value of 5 mSv WLM^{-1} for workplaces.

The values of f_{un} in wineries are similar to those in kindergartens and schools, or even lower, although a reliable comparison is not possible because of the small number of measurements in wineries. Values are much lower than in the Postojna Cave – obviously because of higher aerosol concentrations and lower air humidity in a winery than in the cave[1,18]. In contrast to other workplaces, rooms in wineries are not attended by workers every day and therefore typical diurnal variations of the measured parameters, reflecting working regime, have not been observed. According to the ICRP criteria[16], workers in wineries may be considered as nasal breathers and for them $DCF_D = DCF_{Dn}$, calculated from Eq. 2. DCF_{Dn} values in Table 3 are 1.5-to 1.9-fold higher than 5 mSv WLM^{-1}.

Conclusion

Measurements of f_{un} in Slovenian kindergartens have shown that DCF_D is on average 8.72 mSv WLM^{-1}, which is 1.74-fold higher than the epidemiology-based value of 5 mSv WLM^{-1} currently recommended by ICRP for workplaces. For schools this factor was 1.68.

In the Postojna Cave, DCF_D values were higher than DCF_E by a factor of 1.7 in summer at the railway station, and by a factor from 3.2 to 3.6 in summer and from 1.6 to 1.7 in winter at the lowest point. For maintenance workers engaged in hard physical work this factor is 4.2 in summer at the railway station, and from 12.3 to 14.3 in summer and from 3.2 to 4.0 in winter at the lowest point.

In the four wineries, DCF_D values were in the range from 7.71 mSv WLM^{-1} to 9.56 mSv WLM^{-1} and thus between 1.5- and 1.9-fold higher than DCF_E.

The DCF_D values in the different environments contribute additional information to the general database on DCF_D, which is a prerequisite for a better understanding of and successful coping with the gap between DCF_D and DCF_E, which is not yet completely understood.

Acknowledgements

This study was financed by the Slovene Research Agency. The author thanks Prof. Ivan Kobaian for his constructive suggestions, Ms. Petra Dujmović for her assistance in field measurements and laboratory analyses, and Ms. Asta Gregorič for her technical support. The cooperation of managements and personnel of kindergartens, schools, wineries and the Postojna Cave is appreciated.

REFERENCES

1. BUTTERWECK G, PORSTENDÖRFER J, REINEKING A, KESTEN J, Radiat Prot Dosim, 45 (1992) 167. — 2. NERO JR AV, Radon and its decay products in indoor air: an overview. In: NAZAROFF WW, NERO JR AV (Eds) Radon and its Decay Products in Indoor Air (John Wiley & Sons, New York, 1988). — 3. UNITED NATIONS SCIENTIFIC COMMITTEE ON THE EFFECTS OF ATOMIC RADIATION (UNSCEAR), Effects and Risks of Ionizing Radiation, Report to the General Assembly (United Nations, New York, 2000). — 4. INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION (ICRP), Protection against Radon-222 at Home and at Work. ICRP Publication 65 (Pergamon Press, Oxford, 1994). — 5. BIRCHALL A, JAMES AC, Radiat Prot Dosim, 53 (1994) 133. — 6. HOFMANN W, FAKIR H, AUBIENEAU-LANIECE I, PIHET P, Radiat Prot Dosim, 112 (2004) 493. — 7. PORSTENDÖRFER J, Environ Int, 22 (1996) 563. — 8. VAUPOTIč J, KRIŽMAN M, PLANINIĆ J, PEZDIč J, ADAMIČ K, STEGNAR P, KOBAL I, Health Phys, 66 (1994) 550. — 9. VAUPOTIČ J, ŠIKOVEC M, KOBAL I, Health Phys, 78 (2000) 559. — 10. POPIT A, VAUPOTIč J, Environ Geol, 42 (2002) 330. — 11. VAUPOTIČ J, CSIGE I, RADOLIĆ V, HUNYADI I, PLANINIĆ J, KOBAL I, Health Phys, 80 (2001) 142. — 12. MARKOV KR, RYABOV NV, STAS KN, Atom Energiya, 12 (1962) 315. — 13. KNUTSON EO, Modelling indoor concentrations of radon decay products. In: NAZAROFF WW, NERO JR AV (Eds) Radon and its Decay Products in Indoor Air (John Wiley & Sons, New York, 1988). — 14. HUET C, TYMEN G, BOULAUD D, Sci Total Environ, 272 (2001) 97. — 15. VAUPOTIč J, Chemosphere, 69 (2007) 856. — 16. INTERNATIONAL COMMISSION ON RADIOLOGICAL PROTECTION (ICRP), Human Respiratory Tract Model for Radiological Protection, ICRP Publication 66 (Pergamon Press, Oxford, 1994). — 17. MORLIER JP, BISSON M, FRITSCH P, MONCHAUX G, JANOT M, PINEAU JF, Environ Int, 22 (1996) 927. — 18. VOGIANNIS E, NIKOLOPOULOS D, LOUIZI A, HALVADAKIS CP, Radiat Prot Dosim, 111 (2004) 121.

J. Vaupotič

»Jožef Stefan« Institute, Jamova 39, 1000 Ljubljana, Slovenia
e-mail: janja.vaupotic@ijs.si

RAZINE PRODUKATA RASPADANJA RADONA NANOVELIČINE U ZRAKU INTERIJERA: USPOREDBA KOD RAZLIČITIH OKOLIŠA

S A Ž E T A K

Udio nevezanih (f_{un} – unattached fraction) kratkoživućih produkata raspadanja ^{222}Rn nanoveličine, mjeren je u zraku različitih interijera te su dobiveni slijedeći rasponi: 0,03–0,25 u dječjim vrtićima, 0,03–0,18 u školama, 0,08–0,20 u vinarijama, i 0,12–0,68 u krškim špiljama. Istraživana je i raspravlja se ovisnost f_{un} o okolišnim uvjetima (temperatura zraka, relativna vlažnost i tlak), kao i o radnom režimu. Primjenjujući pristup dozimetrije, izračunati su faktori pretvorbe doze na temelju izmjerenih vrijednosti f_{un} i uspoređeni s vrijednostima dobivenima u epidemiološkim istraživanjima, koji se trenutačno preporučuju za uporabu u dozimetriji radona.