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Radon Doses in Slovenian Schools Based on Alpha Spectroscopy†

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Abstract

Indoor air concentrations of radon and radon short-lived decay products, equilibrium factor, unattached fraction of radon short-lived decay products (f_{un}), relative humidity and temperature have been measured in Slovenian schools, with an emphasis on f_{un} as the crucial parameter in dose assessment. f_{un} ranged from 0.03 to 0.21. Dose conversion factors, based on these values, exceeded 5 mSv/WLM, the value recommended by ICRP-65, in all schools.

Key words: radon unattached fraction, dose conversion factors.

Introduction

Radon is an α-radioactive noble gas originating in the Earth's crust, from where it travels toward the surface by diffusion and, mostly by being transported by thermal waters or carrier gases such as methane, helium and carbon monoxide.[1] There are three isotopes: ^{222}Rn (radon with a half-life τ = 3.8 days) originates from ^{226}Ra in the ^{238}U decay chain; ^{220}Rn (thoron, τ = 56 seconds) from ^{224}Ra in the ^{232}Th decay chain; and ^{219}Rn (actinon, τ = 3.9 seconds) from ^{223}Ra in the ^{228}U decay chain. Only the ^{222}Rn isotope (hereafter simply called radon) has a half-life long enough to appear in indoor air (both in homes and at workplaces) at activity levels potentially hazardous for our health. Radon further decays (α, E_α = 5.5 MeV) into its short-lived decay products (RnDP): ^{218}Po (α decay, τ = 3.1 min, E_α = 6.0 MeV), ^{214}Pb (β-γ decay, τ = 26.8 min), ^{214}Bi (β-γ decay, τ = 19.9 min) and ^{214}Po (α decay, τ = 164 μs, E_α = 7.7 MeV). Short-lived decay products originate as positive ions, which soon recombine with air molecules to form clusters, with an activity median aerodynamic diameter (AMAD) of 0.8 nm (called the unattached fraction of RnDP). Later, some of them attach to dust particles and water droplets to form aerosols with AMAD around 200 nm (called the attached fraction of RnDP). In calm and clean air, a radioactive equilibrium between Rn and RnDP may be reached in about three hours, i.e., from 1 Bq of Rn, 1 Bq of each RnDP is generated. In reality equilibrium is never reached because of air movement and plate-out of aerosols on walls and floors. The equilibrium factor, F, measures the state of radioactive equilibrium, and is in the range 0.40 – 0.50 in indoor air.

When breathing in air contaminated by Rn and RnDP, Rn is inhaled and exhaled, and only a minor fraction decays in the lungs.[2] On the other hand, RnDP appear in the form of clusters and aerosols which are deposited on the walls of airways in the mouth, nose and lungs, where they further decay and damage the nearby tissue. For this reason, we are mainly concerned about RnDP (mostly α-active ^{218}Po and ^{214}Po because of high α-energies) and not about Rn which contributes only about 5% to the lung dose, although we use the general notion of radon dosimetry.

It is relatively simple to measure long-term average Rn concentration in air by exposing track-etched detectors. In contrast, there is no commercially available device to obtain long-term average RnDP concentration. Therefore, it is general practice in radon dosimetry to measure Rn and then to translate its concentrations into RnDP concentration using F = 0.40, as recommended by ICRP-65 methodology.[3] The exposure to Rn and/or RnDP is expressed in an old though convenient unit, WLM (working-level-month). 1 WLM is the exposure received by 170 hours breathing in air with an activity concentration of RnDP of 1 WL, which is defined as ^{218}Po, ^{214}Bi, ^{214}Pb/^{214}Po in radioactive equilibrium (F = 1) with 100 pCi/L (3700 Bqm^{-3}) of ^{222}Rn, resulting in an alpha energy concentration of 1.3×10^5 MeV/L.[1] The effective dose according to ICRP-65, E_{65}, is then calculated using the equation:

$$E_{65} = \frac{C^t_{Rn} \times F}{3700} \times \frac{t}{170} \times DCF \qquad (1)$$

$C_{Rn}{}^t$ is average Rn concentration for the time period t (in hours) spent by a person in the room surveyed. DCF is a dose conversion factor, recommended by ICRP-65 as 5 mSv/WLM.[3]

In a recently proposed, refined dosimetric approach based on a new lung model [4,5], DCF is not taken from ICRP-65 but is rather expressed through the fraction of unattached RnDP, f_{un}, by:

$$DCF_m = 101\times f_{un} + 6.7\times(1 - f_{un}) \qquad (2)$$

for mouth breathing,
and

$$DCF_n = 23\times f_{un} + 6.2\times(1 - f_{un}) \qquad (3)$$

for nasal breathing.

The dose conversion factor for a person, DCF_c, is then composed of DCF_m and DCF_n by taking into account the fraction of mouth (x) and nasal (1 – x) breathing, i. e.,

$$DCF_c = x \times DCF_m + (1 - x) \times DCF_n. \qquad (4)$$

If DCF_c is used instead of 5 mSv/WLM, the resulting effective dose is denoted by E_{un}, to be differentiated from E_{65}:

$$E_{un} = \frac{C^t_{Rn} \times F}{3700} \times \frac{t}{170} \times DCF_c \qquad (5)$$

In the Slovenian Radon Programme, radon was surveyed in 890 elementary and high schools in the period from 1992–94.[6–16] In 78 cases, where indoor air radon concentrations exceeded the national radon limit of 400 Bqm^{-3},[17] additional and more intensive investigations were carried out and effective doses of teachers and students were calculated according to ICRP-65 methodology,[3] using equation (1). In 15 buildings radon problem has been mitigated and doses of personnel successfully reduced to an acceptable level.

In order to check the role of the unattached fraction of RnDP in radon dosimetry, i. e., to compare E_{un} to E_{65}, in the period 1998–2003, we carried out 7–15 day continuous measurements of indoor air concentrations of radon and radon short-lived decay products, equilibrium factor and unattached fraction of radon short-lived decay products in 30 Slovenian schools. Air relative humidity and temperature were also monitored, as two parameters affecting f_{un}.[18–22] In this paper, the influence of environmental parameters and of the working regime on the measured quantities, with emphasis on f_{un} and consequently on E_{un} received by the personnel, have been sought and commented on with regard to the previously obtained E_{65} values.

Experimental

In the study, portable SARAD EQF3020 and EQF3020-2 devices (manufactured by SARAD, Dresden, Germany) have been used. They measure concentrations of radon (C_{Rn}) and radon short-lived decay products (C_{RnDP}) and equilibrium factor (F), as well as air temperature (T) and relative air humidity (RH).[23] The sampling and analysing frequency is once in two hours. A filter separates aerosols with AMAD below 10 nm from those above, enabling measurements of unattached (f_{un}) and attached fractions of RnDP. Alpha spectrometry is used to detect ^{218}Po and ^{214}Po, based not on their α-energies but on the difference in their half-lives, by measuring the total α-activity on the filter at different appropriately selected times.[24] A device has been operated for 7-15 days in one or several rooms of the selected school.

The instruments were calibrated by the manufacturer at the time of purchase, and have since then been checked by the manufacturer every two years. In addition, they have been regularly included in the intercomparison experiments organized annually by the Slovenian Nuclear Safety Administration at the Ministry of Environment and Spatial Planning.[25]

Results and discussion

Temporal variation of the measured parameters and some of their relationships

Figure 1 shows time variations of: a) concentration of radon (C_{Rn}) and equilibrium factor (F), b) concentration of radon short-lived decay products (C_{RnDP}) and unattached fraction of short-lived radon decay products (f_{un}), and c) relative air humidity (RH) and air temperature (T), as recorded during a measurement in the s-MO-S3-01 school. C_{Rn}, C_{RnDP} and F show diurnal fluctuations,[1,11,12] with maxima

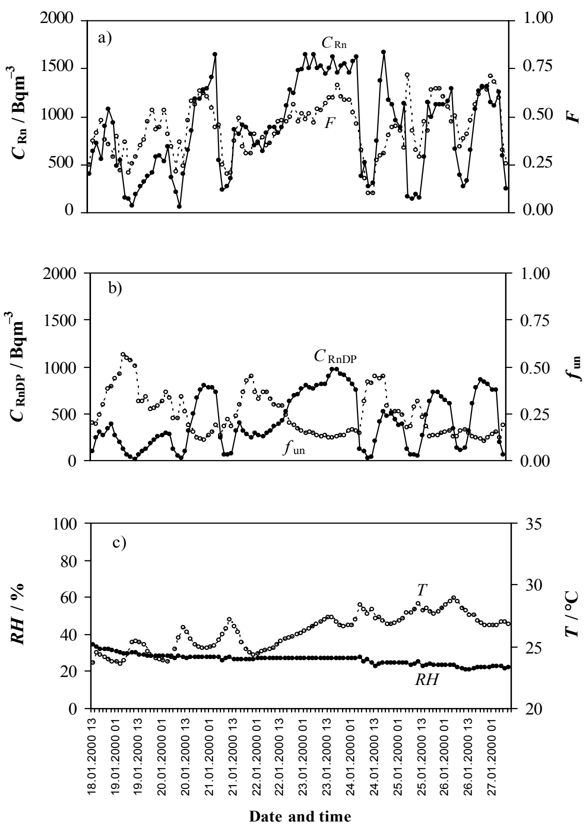

Figure 1. Temporal variations of the parameters measured in the s-MO-S3-01 school during the period January 18–27, 2000: a) concentration of radon (C_{Rn}) and equilibrium factor (F), b) concentration of radon short-lived decay products (C_{RnDP}) and fraction unattached of short-lived radon decay products (f_{un}), c) relative indoor air humidity (RH) and indoor air temperature (T).

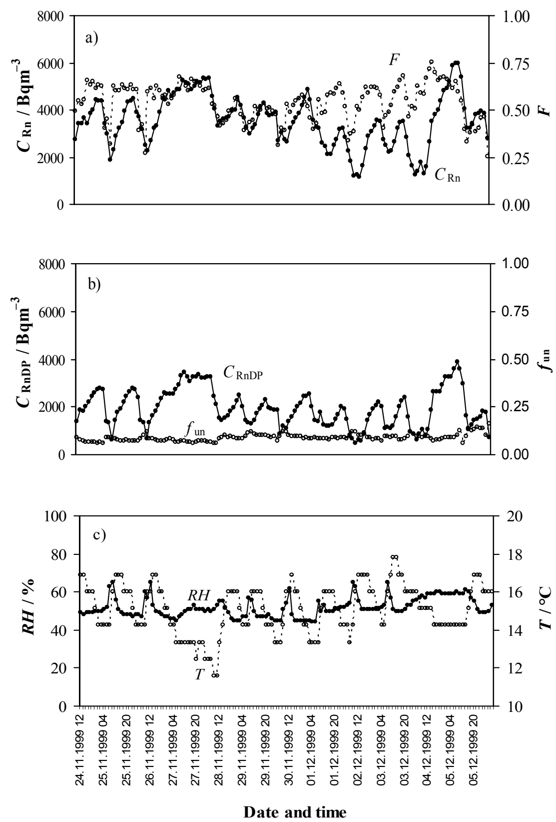

Figure 2. Temporal variation of the parameters measured in the s-SM-S2-12 school during the period January 18–27, 2000: a) concentration of radon (C_{Rn}) and equilibrium factor (F), b) concentration of radon short-lived decay products (C_{RnDP}) and unattached fraction of short-lived radon decay products (f_{un}), c) relative indoor air humidity (RH) and indoor air temperature (T).

during nights and minima in the afternoons of working days, and remain high during weekends. We have been interested mainly in the time variation of f_{un} and its dependence on other parameters, such as F, RH, T, and working regime.

Temporal variations of the measured parameters were in principle similar in all schools, although the amplitude of their fluctuations differed substantially from school to school as well as from measurement to measurement in the same school, depending on meteorological conditions, shape, size and quality of building structure, working regime and other factors.[11,12] As another example, results obtained in the s-SM-S2-12 school are shown in Figure 2. While in the s-MO-S3-01 school, high C_{Rn} values attained overnight were reduced to below 100 Bqm^{-3} during working hours (Figure 1), in school s-SM-S2-12 this reduction was far less, and values remained above 1000 Bqm^{-3} during the whole day (Figure 2). Also daily fluctuations of F and f_{un} are much more pronounced in school s-MO-S3-01 than in school s-SM-S2-12.

Figure 3a shows an exponential decrease of f_{un} with increasing F, as observed also in other experiments.[26–33] Figure 3b shows a poor positive correlation of f_{un} with *HR,* and Figure 3c, a weak negative one with T. These relationships are very complex[34] and their interpretation is beyond the scope of this paper because no data on the number of aerosol particulates in air and its dependence on the working regime is available. In addition, RH and T ranges (which are related) were too narrow to cause apparent changes in f_{un}, because the enhancement of RnDP neutralisation rate due to an increase of RH from 20 to 30% is insignificant.[22]

Comparison of E_{65} and E_{un} effective doses

For each school, average values of C_{Rn} and f_{un} were calculated for working hours (7–14 h) for the whole period of measurement in the school. Values are collected in Table 1. f_{un} values range from 0.03 to 0.21, to be compared with the value of 0.03 implemented in the ICRP-65 methodology for calculating E_{65}. E_{65} doses were calculated by using equation (1), and E_{un} by equations (2) – (5), assuming 80% mouth and 20% of nasal breathing for a teacher. In addition to DCF_m, DCF_n and

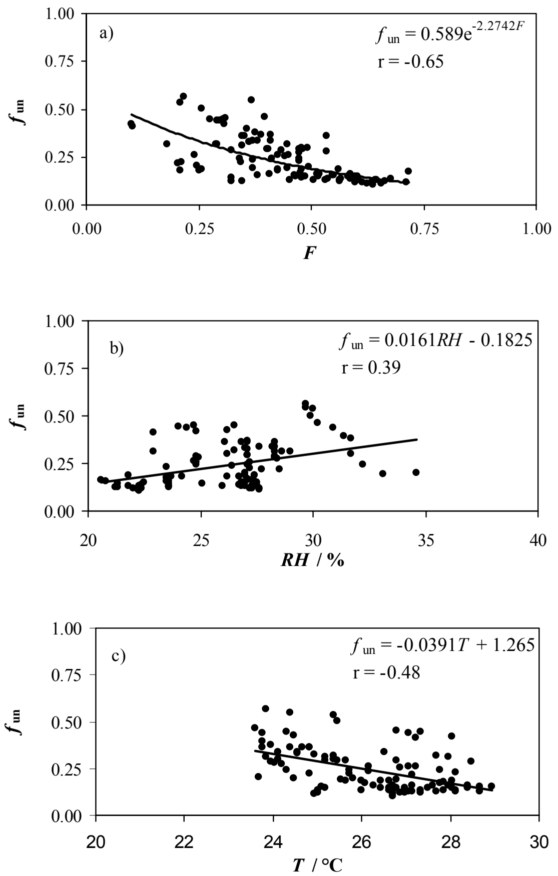

Figure 3. Dependence of f_{un} – the fraction of unattached short-lived radon decay products – on: a) *F* – equilibrium factor, b) *RH* – relative indoor air humidity and c) *T* – indoor air temperature, for the measurements in the s-MO-S3-01 school during the period January 18–27, 2000.

DCF_c values, Table 1 also shows the E_{un}/E_{65} ratio for each school. It is immediately evident that the ratio is higher than 1 for all schools and that the annual effective doses estimated for the school personnel according to the ICRP-65 methodology without taking into account the unattached fraction of RnDP may therefore be highly underestimated. It would thus be advisable to use E_{un} instead of E_{65} in order to ensure being on the safe side from the radiation protection point of view. Referring to Figure 3a, we see compensating effects of f_{un} and *F*: an enhanced *F* would result in higher exposure, but the concomitant lower f_{un} will reduce it.

At present the devices to monitor f_{un} are sophisticated and expensive, and we are far from being able to use them simultaneously at a number of places for long periods of time, as required for a massive radon dosimetry. Therefore, ICRP-65 methodology based on exposing etched track detectors all the year round will still remain in general practice in the near future, and f_{un}-based radon dosimetry will be used for scientific purposes only.

Table 1. Average indoor air Rn concentrations (C_{Rn}^{t}) and fractions of unattached RnDP (f_{un}) in Slovenian schools (names are coded) for working hours (7 – 14h) during the measuring periods indicated. Dose conversion factors for mouth (DCF_m) and nasal (DCF_n) breathing are calculated using equation (2) and (3), respectively. $DCF_c = 0.80 \times DCF_m + 0.20 \times DCF_n$. E_{65} and E_{un} are calculated using equation (1) and (4), respectively.

school code	measuring period	C_{Rn}^{t} Bqm^{-3}	f_{un}	DCF_m	DCF_n	DCF_c	E_{un} / E_{65}
s-VD-S2-02a	03/02-06/02/98	126	0.03	9.5	6.7	9.0	1.79
s-PO-S2-02	06/02-13/02/98	189	0.11	17.1	8.0	15.3	3.05
s-CR-S2-03	12/03-19/03/98	251	0.12	18.0	8.2	16.1	3.21
s-LO-S2-05	19/05-29/05/98	3162	0.16	21.8	8.9	19.2	3.84
s-CE-S2-09	14/09-18/09/98	385	0.13	19.0	8.4	16.8	3.37
s-DV-S2-10	29/09-02/10/98	422	0.15	20.8	8.7	18.4	3.68
s-LO-S2-10	03/10-12/10/98	763	0.15	20.8	8.7	18.4	3.68
s-LO-S2-11	02/11-10/11/98	423	0.17	22.7	9.1	20.0	4.00
s-KR-S2-01	15/01-25/01/99	292	0.13	19.0	8.4	16.8	3.37
s-MO-S2-02	03/02-12/02/99	351	0.21	26.5	9.7	23.1	4.63
s-JU-S2-03	08/03-16/03/99	228	0.14	19.9	8.6	17.6	3.53
s-SE-S2-10	04/10-12/10/99	806	0.19	24.6	9.4	21.6	4.31
s-JA-S2-11	16/11-23/11/99	108	0.18	23.7	9.2	20.8	4.16
s-SM-S2-12	24/11-06/12/99	3304	0.09	15.2	7.7	13.7	2.74
s-DV-S2-12	06/12-09/12/99	1527	0.10	16.1	7.9	14.5	2.90
s-PO-S2-01	10/01-17/01/00	165	0.15	20.8	8.7	18.4	3.68
s-ME-S3-01	10/01-17/01/00	723	0.11	17.1	8.0	15.3	3.05
s-HD-S2-02	28/01-09/02/00	735	0.14	19.9	8.6	17.6	3.53
s-CR-S3-02	10/02-18/02/00	505	0.20	25.6	9.6	22.4	4.47
s-SM-S2-03	14/03-29/03/00	2600	0.15	20.8	8.7	18.4	3.68
s-PR-S2-12a	30/11-11/12/00	1875	0.09	15.2	7.7	13.7	2.74
s-PR-S3-12a	30/11-11/12/00	3007	0.14	19.9	8.6	17.6	3.53
s-PR-S2-12b	11/12-18/12/00	3482	0.09	15.2	7.7	13.7	2.74
s-PR-S3-12b	11/12-18/12/00	6615	0.15	20.8	8.7	18.4	3.68
s-VD-S2-02b	24/01-01/02/01	618	0.10	16.1	7.9	14.5	2.90
s-VD-S3-02	24/01-01/02/01	1557	0.21	26.5	9.7	23.1	4.63
s-ZG-S2-04a	23/03-02/04/01	3176	0.15	20.8	8.7	18.4	3.68
s-ZG-S2-04b	02/04-10/04/01	141	0.15	20.8	8.7	18.4	3.68
s-AM-S2-05	23/04-07/05/01	394	0.14	19.9	8.6	17.6	3.53
s-SM-S1-05	28/05-15/06/01	89	0.21	26.5	9.7	23.1	4.63

Conclusions

Measurements of f_{un} in 30 Slovene schools have shown values between 0.03 and 0.21. Dose conversion factors, *DCF*, for mouth (m) and nasal (n) breathing were calculated and the combined (c) values for all schools obtained, assuming $DCF_c = 0.80 \times DCF_m + 0.20 \times DCF_n$. By including DCF = 5 mSv/WLM and $DCF = DCF_c$, effective doses E_{65} and E_{un}, respectively, were calculated. The ratio of E_{un}/E_{65} ranged from 1.79 to 4.63, being higher than 1 for all schools. Thus, the doses calculated according to ICRP-65 methodology without taking into account the unattached fraction of RnDP were always underestimated. Therefore, whenever the equipment is available, it is advisable to consider the measured data on the unattached fraction of short-lived radon decay products when arriving at dose estimates.

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References

1. A. V. Nero, Jr, in: W. W. Nazaroff, A. V. Nero, Jr (Eds.): Radon and its Decay Products in Indoor Air, John Wiley & Sons, New York, **1988**, pp. 1–53.
2. A. C. James, in: W. W. Nazaroff, A. V. Nero, Jr (Eds.): Radon and its Decay Products in Indoor Air, John Wiley & Sons, New York, **1988**, pp. 259–309.
3. International Commission on Radiological Protection (ICRP), *Protection against Radon-222 at Home and at Work*, ICRP Publication 65, Pergamon Press, Oxford, **1994**, pp. 1-262.
4. International Commission on Radiological Protection (ICRP), *Human Respiratory Tract Model for Radiological Protection*, ICRP Publication 66, Pergamon Press, Oxford, **1994**, pp. 1-482.
5. J. Porstendörfer, *Environ. Int.* **1996**, *22 (Suppl. 1)*, S563–S583.
6. J. Vaupotič, D. Barišić, I. Kobal, S. Lulić, *Radon in a karstic region school: concentration in soil gas and indoor,* in: B. Obelić, Z. Franjić (Eds.), Proceedings of the Fourth Symposium of the Croatian Radiation Protection Association, Croatian Radiation Protection Association, Zagreb, **1998**, pp. 349–353.
7. J. Vaupotič, M. Šikovec, I Kobal, *Health Phys.* **2000**, *78*, 559–562.
8. J. Vaupotič, *J. Radioanal Nucl. Chem.* **2001**, *247*, 291–295.
9. M. Bilban, J. Vaupotič, *Health Phys*. **2001**, *80*, 157–163.
10. J. Vaupotič, I. Hunyadi., E. Bardadács, *Radiat. Meas*. **2001**, *34*, 477–482.
11. J. Vaupotič, *Radiat. Protect*. *Dosim*. **2002**, *102*, 75–80.
12. J. Vaupotič, *Health Phys*. **2002**, *83*, 237–242.
13. A. Popit, J. Vaupotič, *Environ. Geol.* **2002**, *42*, 330–337.
14. A. Popit, J. Vaupotič, *Geologija* **2002**, *45*, 499–504.
15. J. Vaupotič, M. Andjelov, I. Kobal, *Environ. Geol.* **2002**, *42*, 583–587.
16. J. Vaupotič, I. Kobal, *Isot. Environ. Health Stud.* **2002**, *38*, 39–46.
17. Official Journal of the Republic of Slovenia (Uradni list Republike Slovenije). *Regulations of ventilation and air-conditioning of buildings*, Uradni list RS, ULRS **2002**, *42*, pp. 4139–4161.
18. T.-C. Chu, H.-L. Liu, *Appl. Radiat. Isot*. **1996**, *47*, 543–550.
19. F. Marley, A.R. Denman, P.S. Phillips, *Radiat. Meas*. **2000**, *32*, 15–25.
20. F. Marley, *Atmos. Environ.* **2001**, *35*, 5347–5360.
21. V. Dankelmann, A. Reineking, J. Porstendörfer, *Radiat. Prot. Dosim.* **2001**, *94*, 353–357.
22. P. Pagelkopf, J. Porstendörfer, *Atmos. Environ.* **2003**, *37*, 1057-1064.
23. T. Streil, G. Holfeld, V. Oeser, C. Feddersen, K. Schönefeld, *A new microsystem based monitoring system for the continous measurement of radon and the attached and unattached fraction of the radon progeny*, in: D. Glavič-Cindro (Ed.), Symposium on Radiation Protection in Neighbouring Countries in Central Europe 1995, Radiation Protection Association of Slovenia, Ljubljana, **1996**, pp. 334–337.
24. J. W. Thomas, *Health Phys*. **1970**, *19*, 691.
25. M. Križman, *Report on the Intercomparison Experiment for Radon and Progeny in Air*; URSJV RP 47/2001, Nuclear Safety Administration, Ljubljana, **2001**, pp 1-7.
26. S. Tokonami, T. Iimoto, R. Kurosawa, *Environ. Int*. **1996**, *22 (Suppl. 1)*, S611–S616.
27. X. Ortega, A. Vargas, *Environ. Int.* **1996**, *22 (Suppl. 1)*, S149-S159.
28. C.-J. Chen, C.-C. Liu, Y.-M. Lin, *Appl. Radiat. Isot.* **1998**, *49*, 1613-1618.
29. A. Paul, S. Röttger, A. Honig, T. Sulima, A. Buchholz, U. Keyser, *Nucl. Instr. Methods Phys. Res.* **1999**, *A 434*, 303–312.
30. C. Huet, G. Tymen, D. Boulaud, *Sci. Total Environ*. **2001**, *272*, 97–103.
31. J. Vaupotič, I. Kobal, *Radon doses based on alpha spectrometry*, in: M. Franko, P. Trebše, M. Debeljak (Eds.), 13th International Symposium Spectroscopy in Theory and Practice, Book of Abstracts, Politehnika Nova Gorica, **2003**, pp. 48.
32. J. Vaupotič, I. Kobal, *Acta Chim. Slov.* **2004**, *51*, 159–168.
33. J. Vaupotič, I. Kobal, *Acta Carsologica* **2004**, *33*, 86–100.
34. K. Singh, M. Singh, S. Singh, H.S. Sahota, Z. Papp, *Radiat. Meas*. **2005**, *39*, 213–217.

Povzetek

V izbranih šolah s povišano koncentracijo radona v Sloveniji smo 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 (RH) in temperaturo zraka (T). Poseben poudarek je bil na f_{un} in na njegovi odvisnosti od delovnega režima. Vrednosti f_{un} so bile v širokem razponu, od 0,03 do 0,21. Z uporabo novega dozimetrijskega modela smo na osnovi izmerjenih vrednosti f_{un} izračunali dozne pretvorbene faktorje in ugotovili, da so bili višji od vrednosti 5 mSv/WLM, ki jo priporoča metodologija ICRP-65.