NUKLEONIKA 2010;55(4):439–444 ORIGINAL PAPER

First results of measurement of equilibrium factors *F* and unattached fractions f_p of radon progeny in Czech dwellings

Karel Jílek,
Josef Thomas,
Ladislav Tomášek

Abstract. The unattached fraction of radon decay product clusters f_p and equilibrium factor F are dose relevant parameters in all dosimetric approaches to dose calculation. In the past, three year continuous weekly measurements of unattached and attached activity of radon daughter product and air exchange rate were carried out during heating season in 30 occupied typical Czech family houses. The results indicated significantly different weekly averages of equilibrium factor F and f_p for houses located in towns compared those in villages. Due to this fact, approximately a 10% average increase of equivalent lung dose rate was estimated in the detriment in towns. Average values of equilibrium factor F and f_p were 0.40 and 8.6% in urban houses and 0.32 and 10.7%, respectively in rural houses. Based on the measurements of mean values of f_p, average effective dose conversion factors (DFC) in units of mSv per working level month (WLM) were estimated to be 15.0 mSv/WLM in urban houses and 15.9 mSv/WLM in rural houses, respectively.

Key words: unattached fraction f_p • dose • air exchange rate • indoor aerosol concentration • rural/urban area • day/night

K. Jílek✉, J. Thomas
National Radiation Protection Institute,
28 Bartoškova Str., Praha 4-140 00, Czech Republic,
Tel.: +420 226 518 120, Fax: +420 241 410 215,
E-mail: karel.jilek@suro.cz

L. Tomášek
Institute of Computer Science,
Academy of Sciences of the Czech Republic,
28 Bartoškova Str., Praha 4-140 00, Czech Republic

Received: 30 June 2009
Accepted: 30 December 2009

Introduction

The unattached fraction of radon decay product clusters f_p and equilibrium factor F are dose relevant parameters in all dosimetric approaches to dose calculation. Direct measurements of f_p and F under different ambient conditions allow a comparison of the influence of ambient conditions to relative dose contribution [4].

The results of sensitivity analysis applied to the classical Jacobi-Porstendörfer (J-P) room model [5], generally describing dynamics of both unattached and attached short-lived radon progeny in the room, indicate air exchange rate (ACH), deposition q_f and attachment X rate of unattached radon daughter product as the key quantities influencing most strongly the behaviour of observed values f_p and equilibrium factor F in a room. The most important role in processes affecting values of deposition q_f and attachment X rate plays, besides ACH, predominantly aerosol size distribution and total aerosol concentration.

In the past, three year weekly continuous measurements of unattached and attached activity concentration of each radon progeny and of ACH were performed during heating season in thirty occupied typical Czech family houses.

Primarily, the purpose of the present paper is to introduce the results of direct measurements of F and f_p under different living conditions and to estimate the influence of these conditions to dose contribution.

We also focused on the calculation of the key model parameters X, q_f and ACH in measured houses in order to independently estimate the most probable values of

F and f_p by means of the room model and to compare them with those obtained from direct measurements.

Since there are well known practical and principal problems arising from long-term measurements of aerosols in occupied dwellings with any scanning mobility particle sizer (SMPS) caused by evaporating of wide spread butyl alcohol saturator used in particle condensation nuclei counters into the room, aerosol particle size distribution and aerosol concentration were not systematically directly measured. Nevertheless, aerosol concentration Z could be estimated indirectly from measured values f_p by means of Porstendörfer's approximate formula ($f_p = 414/Z$) [6].

Material and methods

All measurements were conducted in thirty occupied typical Czech or two-storey family houses since October to March during heating season. The houses were chosen randomly and they were equipped with a central heating system. Originally, we intended to divide them according to assumed different ambient living conditions from the aerosol concentration and its size distribution standpoint in structure as follows: towns-villages, smokers-non smokers. Unfortunately, after finishing all measurements we had to distinguish only basic living conditions according to the house location, i.e. town-village and newly according to daily time period.

To detect the assumed influence of different occupant's indoor activities and outdoor changes (traffic density, weather conditions etc.) on behaviour of the observed parameters f_p and F we divided each daily 24 h taking measurement time period into "day" from 6 a.m. to 6 p.m. and into "night" taking from 6 p.m. to 6 a.m., respectively. Exposure duration took minimally one week in each house.

Measurement instruments

For measurement of unattached and attached activity concentration of each short-lived radon daughter progeny, we used a continuous monitor Fritra 4 (J. Plch – SMM, CZ). The monitor Fritra 4 has built-in a memory buffer allowing more than 4000 records taking each 120 min. This measuring interval included 10 min sampling and 110 min taking computational procedure with implemented 4 alpha counting intervals providing minimum detectable activity (MDA) for each radon progeny in the following ratio (^{218}Po:^{214}Pb:^{214}Bi) = (1: 0.6: 0.5) stepwise 40 Bq·m^{-3}, 12 Bq·m^{-3}, 15 Bq·m^{-3}. The MDA for measured radon concentration was estimated to be about 15 Bq·m^{-3}. Unattached activity concentration of each radon progeny was measured from the screen with mesh 300 and cut-off diameter d_{50} = 4 nm and the attached activity concentration of each radon progeny from a Millipore filter type AA, 0.8 μm placed behind the screen.

The description of the monitor Fritra 4 alone in more details and quality assurance from the measurement of unattached and attached activity concentration for each radon progeny and from the measurement indoor average aerosol concentration standpoint is given elsewhere [2].

Radon measurements were performed by means of the well-known continuous monitor Alphaguard (Genitron, D) certified in a German reference chamber at the Physikalisch-Technische Bundesanstalt (PTB) in Braunschweig. During all measurements, we used its 60 min measuring diffusion mode. Both the monitors were placed mostly in living rooms at each house.

Theoretical background

Equilibrium factor F and f_p as dose relevant parameters

The lung equivalent dose H from inhalation of short-lived radon progeny can be written by means of measured time integral of radon concentration as follows:

(1) $$H = \text{DFC} \cdot \frac{T_{\exp}}{170} \cdot F \cdot \frac{a_v}{3700}$$

where: DFC is the dose conversion factor [mSv/WLM]; F is the equilibrium factor; a_v is the average radon concentration [Bq·m^{-3}]; $T_{\exp}$ is the exposition duration [h].

According to calculations by Marsh and Birchall [4], the DFC in Eq. (1) can be expressed as a linear function of unattached part of equilibrium equivalent concentration of radon progeny f_p as follows:

(2) $$\text{DFC}(f_p) = 11.35 + 0.43 f_p$$

where f_p expressed as the percentage of total potential alpha energy concentration (PAEC) of the radon progeny mixture.

Finally, considering Eqs. (1) and (2) and assuming "the same" breathing rate, the influence of two different ambient conditions to dose can be expressed in terms of relative change of equivalent dose rate H_1/H_0 as follows:

(3) $$\frac{H_1}{H_0} = \frac{F_1 \cdot (k + f_{p,1})}{F_0 \cdot (k + f_{p,0})}$$

with subscripts 0 and 1 representing two different ambient conditions and k – representing the ratio 11.35/0.43 from Eq. (2).

Attachment rate X, deposition rate q_f and air exchange rate ACH

The attachment rate X and deposition rate q_f (plate-out) and ACH can be calculated, in principle, simultaneously from the measured unattached and attached activity concentration of each radon daughter progeny by means of an algebraic inversion of the model [9] expressed for a room in a steady state as follows:

(4) $$\begin{aligned} a_{j,f} &= \lambda_{i\text{-}1}(a_{j\text{-}1,f} + R_{j\text{-}1} a_{j\text{-}1,a}) / (\text{ACH} + \lambda_j + q_f + X) \\ a_{j,a} &= \frac{(\text{ACH} a_{j,o} + (1 - R_{j\text{-}1})\lambda_j a_{j\text{-}1,a} + X a_{j\text{-}1,f})}{(\text{ACH} + \lambda_j + q_a)} \\ a_{0,i} &= (Q_{\text{Rn}} + \text{ACH} a_{0,o}) / (\lambda_0 + \text{ACH}) \end{aligned}$$

where initial conditions $a_{0,i} = a_{0,i,f}$, $a_{0,i,a} = 0$, $a_{j,o,f} = 0$; a_j – activity concentration of *j*-th radon daughter product [Bq·m^{-3}]; λ_j – decay constant of *j*-th radon daughter product [h^{-1}]; R_j – fraction of recoiled atoms of *j*-th nuclide from aerosol particles; 1st subscript $j = 0, 1, 2, 3, 4$ stepwise ^{222}Rn, ^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po; 2nd subscript *i*, *o* – denotes indoor/outdoor; 3rd subscript *f*, *a* – denotes unattached/attached activity *j*-th radon daughter product; Q_{Rn} – sum of radon entry rates into a house [Bq·m^{-3}·h^{-1}].

Attachment rate

Under realistic indoor conditions, the observed attachment rate *X* in a room in "steady state" can be approximately calculated only on the base of measurement of unattached and attached activity concentration ^{218}Po as follows [5].

(5) $$X = \frac{a_{1ia}}{a_{1if}} \lambda_1$$

where: $a_{1i,a}$, $a_{1i,f}$ are the activity concentration attached, unattached respectively ^{218}Po [Bq·m^{-3}]; λ_1 is the decay constant ^{218}Po [h^{-1}] or in the case of measurement the proper aerosol size distribution is as follows:

(6) $$X = \int_0^\infty \beta(d) Z(d) \mathrm{d}d$$

where: $\beta(d)$ – attachment coefficient of attached radon progeny to aerosol as a function of particle diameter *d* [cm^3·h^{-1}]; $Z(d)$ – aerosol size distribution [cm^{-3}].

According to [1], attachment coefficient $\beta(d)$ can be calculated as follows:

(7) $$\beta(d) = \frac{2\pi D_0 d}{\frac{8D_0}{dv_0} + \frac{d}{2\delta_0}}$$

where: D_o is the diffusion coefficient 6.8×10^{-2} cm^2·s^{-1}; v_0 is the mean thermal velocity 1.72×10^4 cm^{-1}; $\delta_o = d/2 + s_o$ where $s_o = 4.9 \times 10^{-6}$ cm is the mean free path of the unattached decay product clusters.

Plate-out

According to the model, the deposition rate q_f, of unattached daughter products can be calculated approximately under realistic indoor conditions (ACH $<< X + \lambda_1$) as follows:

(8) $$q_f = \lambda_1 \frac{a_0}{a_{1f}} - (\lambda_1 + X)$$

with all symbols as mentioned in previous text Eqs. (4)–(5).

Air exchange rate

Under the assumptions of the model introduced by a set of Eq. (4), the ACH can be calculated both by means of an algebraic inversion given previously, and by means of numerical iterations applying the set of Eq. (4).

On the basis of our previous experiments carried out in a radon chamber of National Radiation Protection Institute (NRPI), Praha we developed a code calculating ACH, in principle, as follows.

Input data for the code were measured at lower and upper limits of ratio activity concentrations, stepwise Rn/^{218}Po, ^{214}Pb/^{218}Po, ^{214}Bi/^{218}Po and at lower and upper limit estimates of *X*, q_f and ACH. The code provides stepwise outputs in a form of sets of required parameters *X*, q_f, ACH and recoil *R* fulfilling all input conditions. The most probable value of ACH is chosen from the set where values *X* and q_f are the closest to the corresponding value *X* and q_f, calculated according to Eqs. (5) and (8), respectively. A large number of tests under the defined ACH in the radon chamber indicated overall agreement of the calculated and true values of ACH up to acceptable 30%.

The differences between measurements were evaluated using the Student-test and assuming log-normal distribution.

Results and discussion

The results of confirmatory data analysis from all measured houses ($N = 30$) in the form of weekly geometric means (GM), corresponding geometric standard deviation (GSD), 95% confidence interval (95% CI) for desired quantities of interest and *p*-value are summarized in Tables 1 and 2.

In Table 1 are shown the results of the analysis applied to measured data for compared time periods day/night. From Table 1, by means of *p*-values can be seen that all quantities of interest are not significantly different, except ACH.

The observed statistically significant approximate average 30% increase of ACH during day compared to night was not surprising. Due to this fact, a higher indoor aerosol concentration coming from outdoor air is assumed and implicitly higher attachment rate *X* (see Eq. (6)) is expected and also measured.

The results of attachment rate *X* calculated according to Eq. (5) can be approximated by their basic parameters (GM = 24 h^{-1}, GSD = 1.63) for time period day and (GM = 20 h^{-1}, GSD = 1.41) for time period night, respectively. These results agree fairly well with published data. According to [1], attachment rate *X* is ranging from 20 to 50 h^{-1} in poor ventilated houses (ACH < 0.5 h^{-1}) without additional aerosol sources.

Considering Eq. (6) and the average attachment coefficient $\beta(d) = 0.005$ cm^3·h^{-1} published by Porstendörfer in case of poorly ventilated houses (ACH < 0.5 h^{-1}) without additional aerosol sources, we can expect an average indoor aerosol concentration $Z \approx 4800$ cm^{-3} during day and $Z \approx 4000$ cm^{-3} during night, respectively.

The results of measured deposition rate q_f calculated according Eq. (8), can be approximated by their basic parameters (GM = 23 h^{-1}, GSD = 1.48) for time period day and (GM = 22 h^{-1}, GSD = 1.35) for time period night, respectively. These measured results agree relatively well with data published by other authors 40 h^{-1} [7], 10 h^{-1} [3], 30 h^{-1} [10].

Table 1. Results of confirmatory analysis of weekly average values f_p, F, X, q_f and ACH over time periods day and night

Quantity (units)	Period	GM	GSD	95% CI		p-value
f_p	day	0.099	1.58	0.04	0.247	$p = 0.494$
	night	0.107	1.47	0.05	0.231	
F	day	0.35	1.31	0.20	0.60	$p = 0.533$
	night	0.36	1.32	0.21	0.63	
X (h^{-1})	day	24	1.63	9	64	$p = 0.130$
	night	20	1.41	10	40	
q_f (h^{-1})	day	23	1.48	11	50	$p = 0.653$
	night	22	1.35	12	40	
ACH (h^{-1})	day	0.41	1.59	0.16	1.04	$p = 0.024$
	night	0.30	1.82	0.09	0.99	

F, f_p – denote non-dimensional equilibrium factor and unattached fraction of equivalent equilibrium concentration, respectively.
X, q_f – denote attachment rate and deposition rate of unattached radon progeny.
ACH – denotes air exchange rate.

According to already cited [1] typical published average values F and f_p in indoor air range between 0.2–0.4 and 0.05–0.20, respectively for "normal" aerosol conditions $Z \approx$ (1000–20 000) cm^{-3} with mean values of $F = 0.3$ and $f_p = 0.096$. These results are in good agreement with our measured results f_p (GM = 0.099, GSD = 1.58), F (GM = 0.35, GSD = 1.31) found for time period day and f_p (GM = 0.107, GSD = 1.47), F (GM = 0.36, GSD = 1.32) found for time period night.

In Table 2 are shown the results of the analysis applied to measured data for compared locations town-village. From p-values in Table 2, it can be seen that all quantity of interest are significantly different except deposition q_f and the fact that observed mean values F, f_p, X and q_f agree very well with the published data mentioned previously.

In our opinion, the higher average value F and logically lower average value f_p in indoor air in towns compared to villages are caused mainly by higher average aerosol concentration probably in consequence of different indoor activities. This fact is indicated by dramatically higher values of attachment rate X in towns compared to villages and statistically significant lower average value ACH approximately about 30%.

If we consider the above-mentioned average attachment coefficient $\beta(d) = 0.005$ $cm^3 \cdot h^{-1}$ published by Porstendörfer, Eq. (6) and measured means of attachment rates X from Table 2, we are able to estimate the average indoor aerosol concentration $Z \approx 5200$ cm^{-3} in the case of towns and $Z \approx 3600$ cm^{-3} in the case of villages.

From Table 2, it can be also seen that the measured average value ACH is ranging around (0.3–0.4) h^{-1} in the case of both towns and villages.

If we consider the Porstendörfer's approximate formula $f_p = 414/Z$ mentioned previously (see Introduction) and the measured average values of f_p calculated for different observed conditions from Tables 1 and 2 we can estimate average value of indoor aerosol concentration as follows:

- $Z \approx 4800$ cm^{-3} in the case of towns and $Z \approx 4000$ cm^{-3} in the case of villages, respectively,
- $Z \approx 4200$ cm^{-3} for time period day and $Z \approx 3900$ cm^{-3} for time period night.

These results indicate very good agreement of both approaches used to estimate average aerosol concentration Z, i.e. by means of the measured attachment rate X and f_p, respectively and also implicitly a good accuracy of measurements of unattached and attached activity concentrations of radon daughter product by means of the monitor Fritra 4.

Since the average value f_p in the measured urban houses were 8.6% and 10.6% in rural houses, then, with respect to Eq. (2), the lung DFC = 15.0 mSv/WLM should be applied in the case of urban houses and DFC = 15.9 mSv/WLM in the case of rural houses, respectively. Further, according to [4], the lung dose sensitivity per unit exposure to radon progeny contributes to more

Table 2. Results of confirmatory analysis weekly average values of f_p, F, X, q_f and ACH for location town and village

Quantity (units)	Period	GM	GSD	95% CI		p-value
f_p	town	0.086	1.58	0.04	0.247	$p = 0.003$
	village	0.107	1.40	0.05	0.231	
F	town	0.40	1.25	0.26	0.63	$p = 0.002$
	village	0.32	1.31	0.19	0.55	
X (h^{-1})	town	26	1.61	10	67	$p = 0.001$
	village	18	1.37	10	34	
q_f (h^{-1})	town	23	1.41	12	46	$p = 0.661$
	village	22	1.42	11	44	
ACH (h^{-1})	town	0.30	1.91	0.08	1.09	$p = 0.046$
	village	0.40	1.56	0.16	0.97	

The quantity f_p, F, X, q_f and ACH have the same meaning as in the previous Table 1.

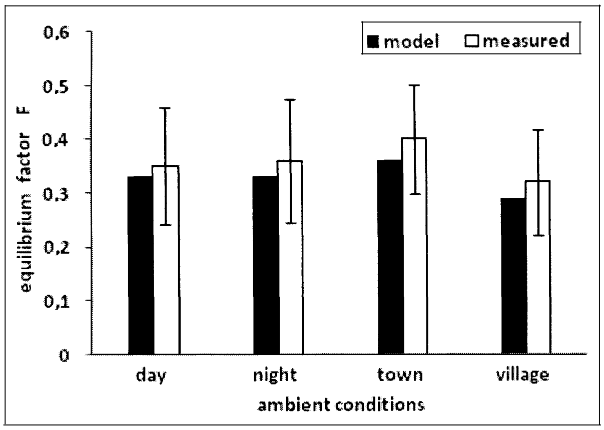

Fig. 1. Results of comparison of equilibrium factor F calculated via the room model and directly measured, respectively (with GSD).

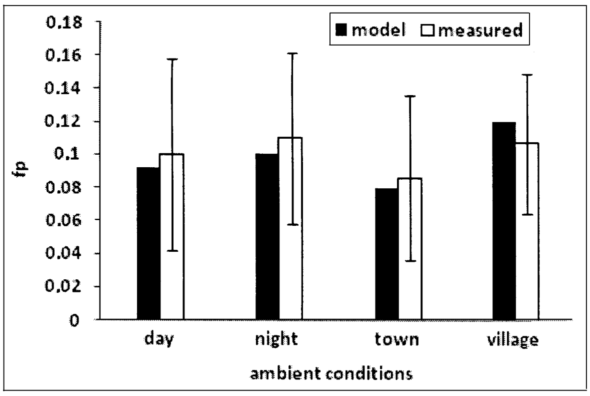

Fig. 2. Results of comparison of f_p calculated via the room model and directly measured, respectively (with GSD).

than 99% of the effective dose, and therefore the above--mentioned values of the lung DFC can represent very well the effective DFC in relevant units (mSv/WLM).

To verify the J-P model (see Eq. (4)) from the prediction of F and f_p point of view, in Figs. 1 and 2 are illustrated results of comparison of the measured values F and f_p with those calculated by means of the room model on the basis of input data, i.e. average values X, q_f and ACH (taken from Tables 1 and 2) for investigated ambient conditions. For all calculations, average value of recoil $R = 0.5$ was used [8]. The results indicate very good agreement between the directly measured values of F and f_p and those calculated by means of the room model.

Assuming "the same breathing rate", the influence of our different investigated ambient conditions (denoted 0 and 1, respectively) to relative changes of the equivalent lung dose rate H_1/H_0 is illustrated in Fig. 3.

In Fig. 3 it is seen the average value of H_1/H_0 calculated for proper observed ambient conditions applying Eqs. (1)–(3) and combined standard uncertainty acquired by application of the Law of Propagation of Uncertainty to Eq. (3).

About a 10% significant relative increase of the equivalent lung dose rate in the case of towns compared to villages was found. On the other hand, the difference in contribution to relative lung dose rate in the case of observed conditions day/night was not statistically significant.

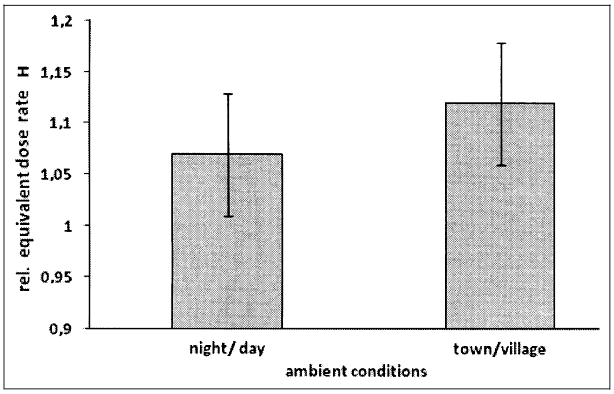

Fig. 3. Average relative contribution to lung equivalent dose rate H_1/H_0 under different investigated ambient conditions and the corresponding combined standard uncertainty.

Conclusion

The results of weekly continuous measurements of radon concentration and unattached and attached activity concentration of each short-lived radon progeny in thirty randomly chosen Czech family houses during heating season indicated a statistically significant difference of weekly averages equilibrium factor F and f_p for urban houses compared to rural. The log-normal distribution of weekly means f_p and F is represented in the case of towns as follows: f_p (GM = 0.086, GSD = 1.58), F (GM = 0.40, GSD = 1.25) and f_p (GM = 0.107, GSD = 1.40), F (GM = 0.32, GSD = 1.31), respectively in case of villages. Due to this fact, approximately a 10% average increase of equivalent lung dose rate was calculated in the detriment of towns.

The difference in mean values f_p and F during observed time period day/night was not statistically significant.

The effective dose conversion coefficients per WLM were estimated to 15.0 mSv/WLM in the case of towns and 15.9 mSv/WLM in the case of villages, respectively.

Both key parameters of the J-P room model, i.e. attachment rate X and plate-out q_f of unattached radon progeny ranged from 10 to 60 h^{-1} with the mean around (20–25) h^{-1}.

The air exchange rate ranged from 0.1 to 1 h^{-1} with the mean approximately around (0.3–0.4) h^{-1}, and its statistically significant average 30% decrease during the night compared to day was observed.

The average indoor aerosol concentration was statistically significant higher in towns ($Z \approx 5000$ cm^{-3}) compared to villages ($Z \approx 4000$ cm^{-3}). On the other hand, difference between mean indoor aerosol concentration during observed time period day ($Z \approx 4200$ cm^{-3}) was not statistically significant compared to night time period ($Z \approx 3900$ cm^{-3}).

Acknowledgment. The present work was supported by the State Office for Nuclear Safety under the Project no. 50013184/11/2006.

References

1. Commission of the European Communities Report EUR 14411EN (1993) Fifth Int Symp on the Natural Radiation Environment, ECSC-EEC-EAEC, Brussels, Luxembourg
2. Jílek K, Thomas J, Brabec M (2008) QA – programme for radon and its short-lived radon progeny measuring instruments. Radiat Protect Dosim 130;1:43–47
3. Knutson EO, George AC, Frey JJ, Koh BR (1983) Radon daughter plate-out. Part II. Health Phys 45:445–452
4. Marsh JW, Birchall A (2000) Sensitivity analysis of the weighted equivalent lung dose per unit exposure from radon progeny. Radiat Protect Dosim 87;3:167–178
5. Porstendörfer J (1984) Behaviour of radon daughter products in indoor air. Radiat Protect Dosim 7;1/4:107–113
6. Porstendörfer J (2001) Physical parameters and dose factors of the radon and thoron decay products. Radiat Protect Dosim 94;4:365–373
7. Porstendörfer J, Reineking A, Becker KH (1987) Free fractions, attachment rates, and plate-out rates of radon daughters in houses. In: Hopke P (ed) Radon and its decay products: occurence, properties, and health effects. Symposium Series no. 331. American Chemical Society, Washington DC, pp 85–300
8. Reineking A, Porstendörfer J (1990) The unattached fraction of short-lived radon decay products in indoor and outdoor environment. Health Phys 58:715–727
9. Thomas J, Jílek K, Brabec M (2010) Inversion of the Jacobi-Porstendörfer room model for the radon progeny. Nukleonika 55;4:433–437
10. Wicke A, Porstendörfer J (1982) Radon daughter equilibrium in dwellings. In: Vohra KG, Mishra UC, Pillau KG, Sadasivan S (eds) Natural radiation environment. Eastern Wiley Limited, New Delhi, pp 481–488