Deposition of short-lived Rn-222 decay products on solid surfaces

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Literature data for the last two decades on deposition velocities of unattached radon decay products has been analyzed. A long-term experiment on investigation of the variation range of the deposition velocity of short-lived ^{222}Rn decay products on a surface (dwelling walls) was conducted for actual living conditions in Germany. The influence of the different parameters (atmospheric pressure, indoor and outdoor air temperatures, humidity, turbulence in the near-surface air, radon concentration activity, heating, ventilation regimes, and season) on the deposition velocity was statistically analyzed. Mean value and the deposition velocity range were found to be 0.05 cm · s^{-1} (0.02–0.09 cm · s^{-1}) for unattached ^{218}Po and ^{214}Pb. Significant correlation between the unattached ^{218}Po deposition velocity and indoor air temperature, humidity, and season was found.

Introduction

Knowledge of basic transfer mechanisms of the radon daughter decay product (DDP) in air is necessary when solving applied problems in many fields of science. The transfer processes influence the concentration activity of radioactive aerosols and gases inside buildings and determine regularities in their behavior.

In air, DDP can be in a free or linked state, the latter implies "attached to aerosol particles." By the free state we mean molecular state of chemical compounds of the radon decay products with vapor of NO, NO_2, H_2O, oxygen, and different gaseous impurities in the indoor air. Diameter of free radon DDP varies from 0.5 to 3 nm (Ref. 1). Particles of this size are transferred in air and deposit on surfaces mainly due to Brownian motion or turbulent diffusion, if it exists.

Surface deposition is the most important process in reduction of the indoor concentration activity of radioactive aerosol particles, which lead to radioactive unbalance between radon and its decay products.[2–4] Quantitative estimates of the surface deposition of radon DDP are important for metrology,[5–9] radiation ecology, and dosimetry.[10,11]

Deposition processes of the radon daughter products are characterized by the deposition velocities of their free (v^f) and attached (v^a) fractions, which can depend on many factors. To date, such factors as electric field,[9,12,13] surface material,[12,14] surface geometry,[12,13] position of the surface relative to particle stream,[15] and indoor air turbulence[12,16–18] are thoroughly studied.

Calculated and experimental data[12] have shown that the influence of turbulence in the naturally ventilated houses on the deposition velocity is much lower than expected. An essential difference in the deposition velocity was found only under low turbulence conditions observed in a thermally isolated chamber or in dwelling houses.

Deposition velocity is proportional to the diffusion coefficient, which in its turn depends on particle linear dimensions. The diameter of DDP when they get attached to aerosol particles becomes on average two orders of magnitude larger,[3] and, correspondingly, the diffusion coefficient reduces. Therefore, the attached fraction of the radon decay products does not significantly contribute to the deposition processes. The unattached radon DDP fraction in air is presented mainly by the short-lived ^{218}Po and ^{214}Pb isotopes (90% and more), therefore, other isotopes (^{214}Bi) are usually neglected in the deposition processes.

Mean values and ranges of deposition velocity of the unattached radon DDP obtained by different scientists theoretically or experimentally, are summarized in Table 1.

In 1998 in Germany, deposition velocity measurements were conducted under real dwelling conditions in different seasons.[18] The measurement results well agree with numerical calculations[15,24–26] and measurements conducted in a radon chamber,[5] though they are an order of magnitude lower than the values reported in Refs. 19–23.

Unfortunately, deposition velocity measurements[18] conducted in different dwelling houses are single, and do not allow tracing temporal dynamics of desired parameters and determining the influence of meteorological conditions on them.

Differences in deposition velocities of unattached ^{218}Po and ^{214}Pb, established in Refs. 18 and 27 (Table 1), also attract some attention, though earlier they were believed to be equal, that was experimentally proved.[21]

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Table 1

Source	Deposition velocity of unattached ^{218}Po (^{214}Pb), cm/s	Notes
Hengde et al., 1982 (Ref. 19)	0.4	Measurements by proportional counter
Scott, 1983 (Ref. 20)	0.14–0.5	Measurements by tracking detectors
Toohey et al., 1984 (Ref. 21) – ^{218}Po and ^{214}Pb deposition velocity ratio	0.4(0.4) 1	Measurements by proportional counter
Rudnick et al., 1986 (Ref. 22)	0.23 1.3	Without ventilation Under ventilation
Leonard, 1995 (Ref. 23)	0.1–0.6	Extrapolation of measurement results obtained in a 0.28 m^3 chamber to dwelling houses
McLaughlin et al., 1984 (Ref. 5)	0.04	Measurements with a surface-barrier detector in a small radon chamber
Knutson et al., 1983 (Ref. 24)	0.033–0.066	Numerical calculations
Brager et al., 1991 (Ref. 25)	0.014–0.079	Numerical calculations
Nazaroff et al., 1992 (Ref. 26)	0.02–0.08	Numerical calculations
Gadgil et al., 1992 (Ref. 15)	0.02–0.04	Numerical calculations
Vanmarcke et al., 1991 (Ref. 27) – ^{218}Po and ^{214}Pb deposition velocity ratio	3	Theoretical calculations
Schmid et al., 1999 (Ref. 18) – ^{218}Po and ^{214}Pb deposition velocity ratio	0.02–0.10 (0.004–0.021) 4	Measurements with a semiconductor (Si) α-detector in dwelling houses in different seasons

Thus, the effects of meteorological conditions on velocity of the unattached radon DDP deposition onto solid surfaces as well as relations between unattached ^{218}Po and ^{214}Pb deposition velocities remain poorly studied today.

In this work, we have attempted to answer these questions through a long-term experiment.

The procedure of the experiment

In the experiment we used a room in one of the BfS buildings (Federal Office for Radiation Protection) of 118 m^3 volume and 162 m^2 total surface area. Measurements were conducted from March to July 2001.

Deposition velocity measurement

Surface deposition velocity is generally determined by the ratio of the flux of radiative aerosol particles onto this surface to the concentration activity of these particles in the indoor air. In this work, the flux was determined from the surface activity of settled unattached radon DDPs. Neglecting the contribution of the unattached ^{214}Bi into the deposition process, the surface deposition velocity of unattached short-lived radon decay products (v^f) was calculated by the equations[12]:

$$v_1^f = A_1 \frac{\lambda_1}{a_1^f} \text{ for } ^{218}\text{Po } (i = 1), \quad (1)$$

$$v_2^f = (A_4 - A_1) \frac{\lambda_2}{a_2^f} \text{ for } ^{214}\text{Po } (i = 2), \quad (2)$$

where v_i^f is the deposition velocity of unattached ith radionuclide, m/s; a_i^f is the concentration activity of unattached ith radionuclide in air, Bq/m^3; λ_i is the decay constant, s^{-1}; A_i is the surface (deposited) activity of ^{218}Po ($i = 1$) and ^{214}Po ($i = 4$), Bq/m^2.

Surface activity was measured with a semiconductor (Si) α-detector incorporated into the wall surface so that the sensitive detector surface was in level with the wall surface, because the measurement geometry could influence the particle deposition velocity. With the same purpose, the detector was placed more than thirty centimeters from the room corners. By using the pulse analyzer, we obtained the total number of pulses separately for ^{218}Po and ^{214}Po, subtracted background values and then calculated the surface activity (Bq/m^2) by the formula

$$A_i = N_i/(t\varepsilon F), \quad (3)$$

where N_i stands for the total number of pulses for the ith radionuclide; t is measurement time, s; $\varepsilon = 0.5$ is a detection efficiency of the silicon α-detector; $F = 4.5 \cdot 10^{-4}$ m^2 is the sensitive surface area of the detector.

Concentration activity of the unattached fraction of the radon DDP in indoor air was measured using the well known wire screen method and the Markov algorithm.[12]

The deposition velocity values calculated by Eqs. (1)–(2) are daily average estimates.

Influence of different factors

In this work we have studied the influence of different factors on the deposition velocity of the

unattached ^{218}Po and ^{214}Pb onto solid surfaces (dwelling walls). We considered the following factors of influence: atmospheric pressure, indoor and outdoor air temperatures, air humidity, radon indoor concentration activity, rates of indoor heat exchange and air exchange, season. Such parameters as radon concentration activity, indoor air humidity, and atmospheric pressure were continuously measured through the experiment with the multifunctional Alpha-GUARD radiometer. The values obtained were averaged over a day to conduct the correlation analysis of the results.

Air exchange (ventilation) rate in the experimental room was determined through a single injection of a large concentration trace gas (CO_2 or SF_6) with subsequent measurements of temporal exponential decrease in its concentration:

$$N(t) = N_0 e^{-\tau t}, \quad (4)$$

where N_0 stands for the initial gas concentration; τ is the indoor air exchange rate, h^{-1}. Gas concentration was measured with a gas analyser (Multi-gas monitor BRÜEL&KJAER; Type 1302).

Results and discussion

The results of investigation have shown that the deposition velocity range is the same for the unattached ^{218}Po and ^{214}Pb and equals 0.02–0.09 cm/s with a mean value of 0.05 cm/s. The values obtained for ^{218}Po well agree with the results described in Refs. 5, 15, 18, and 24–26.

Correlation analysis of the measured average daily deposition velocity values and the chosen factors of influence revealed significant dependences of the unattached ^{218}Po deposition velocity on humidity ($K = -0.92$) and temperature ($K = -0.88$) of the indoor air. Any important relations between the unattached ^{214}Pb daily deposition velocities and meteorological parameters were not found.

Table 2 gives monthly average values of the unattached ^{218}Po and ^{214}Pb deposition velocities, indoor air temperature, and humidity. As is seen, ^{218}Po deposits slower with growing temperature and humidity. For ^{214}Pb, a clear temperature and humidity dependence is not observed, though monthly averages vary up to one and a half times. Thus, the results show that deposition velocities of the unattached ^{218}Po and ^{214}Pb depend on the season (or month) with its characteristic temperature and humidity, but these dependences differ.

Deposition velocity of the unattached radon DDP is an important parameter of the model of radon DDP transfer in air. Literature experimental data (Table 1) on the difference between the deposition velocities of the unattached ^{218}Po and ^{214}Pb show that the ratio of these velocities (v_1^f / v_2^f) can be 3:1 (Ref. 27) or 4:1 (Ref. 18). Disregard to this difference can significantly affect modeling results and eventually bring researchers to false conclusions. In view of the contradictions found in literature we decided to analyze daily average and monthly average velocity ratios (v_1^f / v_2^f).

It was found that daily average (v_1^f / v_2^f) values range from 0.5 to 4 during the whole experimental period (March–July). Table 2 gives monthly average (v_1^f / v_2^f) values, which vary considerably from month to month, though the average velocity ratio for the whole experimental period equals ~1, which well agrees with the results of Ref. 21. Such behavior of (v_1^f / v_2^f) allows us to explain the contradiction in literature data. Moreover, this ratio can be below unity in a warm season.

The influence of heat exchange on the deposition velocity was studied under conditions with and without heating. It was found that deposition velocities of the unattached ^{218}Po and ^{214}Pb are dependent on indoor heat exchange rate. Any significant dependence of the ^{218}Po and ^{214}Pb deposition velocities on the atmospheric pressure and radon concentration activity in indoor air was not found.

Conclusions

By the results of the experiment performed we determined that the unattached ^{218}Po and ^{214}Pb have the same velocity variation ranges of deposition on solid surfaces, namely, 0.02–0.09 cm/s with the average level of 0.05 cm/s. The v_1^f / v_2^f value averaged over a long time period (several months) tends to unity, though daily averages can vary up to eight times in different seasons. In monthly averaged values of v_1^f / v_2^f there is a downtrend with minima in warm time of the year. Significant dependences of deposition velocity of the unattached ^{218}Po on the temperature, indoor air humidity, and heat exchange rate were found. For ^{214}Pb, the temperature and humidity dependences are weaker.

Table 2

Month	Indoor air humidity, g/kg	Indoor air temperature, °C	Deposition velocity, cm/s		Velocity ratio of unattached ^{218}Po and ^{214}Pb
			^{218}Po	^{214}Pb	
March	4.5	23	0.075	0.055	1.99
April	7.0	24	0.062	0.052	1.21
May	9.3	26	0.031	0.041	0.84
June	9.2	27	0.035	0.043	0.99
July	10.7	28	0.039	0.070	0.64

The obtained deposition velocity values can be used in modeling the processes of the radon daughter product transfer inside dwelling houses as well as in retrospective analyses of the radon doses, dwellers were exposed to in the past.

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