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THORON ACTIVITY LEVEL AND RADON MEASUREMENT BY A NUCLEAR TRACK DETECTOR

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Radon activity concentrations in the air were measured with LR-115 nuclear track detectors at three locations in Osijek. The respective equilibrium factors and the effective dose equivalents were determined. Indoor concentrations were from 9.8 to 58.2 Bq m^{-3} and relative errors of the track etching method were near 19 per cent. The indoor alpha potential energy of the radon and thoron progenies was measured with an ISD detector. Independent measurements, performed with a Radhome semiconductor detector, showed that the indoor thoron concentration was nearly 20 per cent of the radon one.

Key terms: effective dose equivalent, equilibrium factor, indoor radon concentrations

The ^{222}Rn (radon, Rn) and ^{220}Tn (thoron, Tn) radioisotopes of the noble gas radon belong to the decay chains of ^{238}U and ^{232}Th, and the parent atoms can be found in all natural materials (1). Radon is released from the earth's surface and building materials into the air, where the radon activity concentration can be slightly higher than or close to that of thoron. Because of a significant difference in their alpha decay constants ($2.09\text{x}10^{-6}$ s^{-1} and $1.26\text{x}10^{-2}$ s^{-1} for radon and thoron, respectively) and different short-lived daughters, the thoron concentration value is commonly neglected by atmospheric radon measurements, which seems unjustifiable.

Indoor and outdoor radon concentrations in Osijek, Croatia, have been measured by means of LR-115 solid state nuclear track (SSNT) detectors since 1985 (2). In 1985 and 1986, the seasonal outdoor radon concentrations varied from 2 Bq m^{-3} (September) to 16 Bq m^{-3} (January), the mean 7.5 Bq m^{-3}. However, the average indoor radon concentration was 22.5 Bq m^{-3} (3).

In order to determine the dose equivalent of radon and its daughters, two SSNT detectors were used in bare and filtered states and a relationship between the equilibrium factor (F) and alpha track densities of the detectors was derived (4).

The equilibrium factor (5) is defined as the ratio of the equilibrium equivalent concentration of radon (c_E) to the actual activity concentration of radon in the air (c_0), i.e.:

$$F = \frac{c^E}{c_0} \quad (1)$$

where c_E is the radon concentration in equilibrium with its daughters which has the same potential alpha energy concentration as the given non-equilibrium mixture of the radon daughters in the air. The equilibrium equivalent concentration of the radon daughters is:

$$c_e = f_1c_1 + f_2c_2 + f_3c_3 \quad (2)$$

where c_1, c_2 and c_3 are ^{218}Po, and ^{214}Pb and ^{214}Bi concentrations, respectively, and f_1=0.105, f_2=0.516, f_3=0.380. In addition to the processes of disintegration and ventilation, a deposition (plateout) was also included into concentration equations of radon and its progeny, and a more realistic relationship for F was obtained (5, 6).

The equilibrium factor measurements done with the Integrated System for Dosimetry (ISD) detector (CRPM, France) have been also presented here and the contribution of thoron in indoor radon concentration measurements in Osijek has been considered.

METHODS

Alpha activity concentrations of radon and its progeny were measured by two LR-115 (Kodak-Pathé) detectors in bare and filtered states. The detector cup, whose diameter and length were 9.6 and 9 cm, was covered with filter paper of 0.078 kg m^{-2} surface density. The filter had high permeability for radon but did not allow its daughters to pass, since they were mainly in the ionic form. The detectors were exposed in the Osijek residential areas Blokcentar, B (fourth floor of a building, concrete and brick), and Retfala, R (single house, brick, first floor and outdoors) and in the nearby village of Tiborjanci, T (single house, brick, first floor), for eight weeks. They were etched in 10 per cent NaOH aqueous solution at 600 °C (333 K) for 120 min. Afterwards, the detector tracks were counted visually using a microscope of magnification 10x16.

For determination of D and D_0 detector track densities of the bare and filtered detectors, respectively, the concentrations of ^{222}Rn (c_0), and its daughters ^{218}Po (c_1) and ^{214}Po (c_4), the following equations were used (6, 7,):

$$D = k(c_0+c_1+c_4) \quad (3)$$

$$D_0 = k\ c_0 \quad (4)$$

where k was the detector sensitivity coefficient.

The LR–115 detector sensitivity coefficient (k) was calibrated in Alpha Instrumentation Laboratory, CRPM, Fanay (France) by means of the radon chamber with a steady-state flow of the filtered radon atmosphere. The radon concentration was measured continuously for 47.5 h by an ionization chamber and the average concentration was obtained as 2938 Bq m^{-3}. The sensitivity coefficient of the filtered detector was k=0.390 cm. A relative statistical error (ratio of the mean to standard deviation) of k was 1.4 per cent.

Theoretical considerations of the disintegration, ventilation and deposition as removal processes of the radon daughters in the room air, combined with equations 3 and 4 gave the following relationship between the equilibrium factor and track densities (6):

$$F = a \exp (bD_0/D) \tag{5}$$

where the parameters were a=14.96 and b=–7.44.

The effective dose equivalent was calculated by means of the following equation:

$$H = C_0(d_0+d_EF) \tag{6}$$

where d_0 and d_E, the dose conversion factors for radon and its daughters, were 0.33 (8) and 80 μSv y^{-1} Bq^{-1} m^3 (9), respectively.

An ISD detector also consists of an LR-115 film, but it records alpha particles emitted by the individual radon and thoron daughters separately (10). The radon progeny is collected on a filter through which air is forced by a ventilator. Alpha particles with different energies (5.3–8.7 MeV) coming from the filter pass by a system of collimators and energy-selecting absorbers of three different thicknesses (8, 23 and 36 μm). Three circular zones (with a diameter of 6 mm) of the film record N_1, N_2 and N_3 tracks belonging to alpha particles of the radionuclides (^{218}Po + ^{212}Bi), ^{214}Po and (^{214}Po + ^{212}Po), respectively (^{212}Bi and ^{212}Po are the thoron daughters; the ^{212}Bi decay gave 36 per cent alpha and 64 per cent beta particles).

Taking into account the volume of the air sampled given as the product of the volume rate (or air flow) Q and time t, the alpha potential energy of radon daughters (E_{Rn}) was calculated in accordance with the following equation (10):

$$E_{Rn} = \frac{1.6x10^{-13}}{R\,Q\,t} \{7.7N_2 + 5.99\ [N_1 - 0.36(N_3-N)_2]\} \tag{7}$$

where R is the efficiency coefficient with an appropriated value of $4.76x10^{-3}$, $Q=4.8x10^{-3}$ m^3 h^{-1}; E_{Rn} is given in J m^{-3} for t in (h).

Also, the alpha potential energy of the thoron daughters E_{Tn} was calculated in the following way (9):

$$E_{tn} = \frac{1.6x10^{-13}}{R\,Q\,t} [8.7(N_3-N_2)+6.08x0.36(N_3-N_2)] \tag{8}$$

The factors 5.99, 6.08, 7.7 and 8.7, in Equations 7 and 8 are the daughters' alpha particle energies in MeV.

Also, indoor radon and thoron concentrations were measured by a silicon semiconductor detector Radhome (CRPM, France), but without data about the daughters; the measurements were performed in time of 36 h.

RESULTS AND DISCUSSION

During 1990 radon measurements at the Osijek B, R and T locations gave the respective average indoor radon concentrations (Equation 4) of 9.8, 58.2 and 11.7 Bq m^{-3}, but the equilibrium factors (Equation 5) were 0.52, 0.46 and 0.48 and the effective dose equivalents (Equation 6) 0.41, 2.16 and 0.53 m Sv y^{-1} (taking into account an 1.0 occupancy factor).

The average outdoor radon concentration at the R location was 23.3 Bq m^{-3} (detectors exposed one meter above the ground) while the highest radon concentrations in January and February were about six times higher than the ones in September and October.

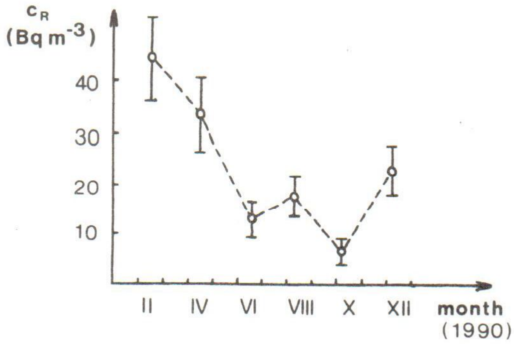

Figure 1 *Outdoor radon concentrations at location R (c_R) versus months in the year 1990. Statistical error is presented as a standard deviation.*

The significantly higher indoor radon concentration at location R than the the ones at locations B and T can be explained by the high outdoor radon concentration at the Retfala (R) location. Considering that the houses at all three locations were built of similar building materials (bricks), it is concluded that a main source of the indoor radon is the soil.

The relative statistical errors of the radon concentrations measured by an LR–115 detector were near 19 per cent.

At location T alpha potential energy of the radon and thoron progenies was measured indoors by means of an ISD detector during four weeks and the following values were obtained (Equations 7 and 8): E_{Rn}=1.52x10^{-8} J m^{-3} and E_{Tn}=9.50x10^{-9} J m^{-3}.

At the same time, in September, the LR-115 detector exposures at position T (living room) gave the radon concentration of 7.0 Bq m^{-3}, then D/D_0=2.16 and F_s=0.48 (using Equation 5). However, alpha potential energy, expressed in concentration equivalent units (1) [c_E (Bq m^{-3})=1.81x10^{8}xE_{Rn}(J m^{-3})], for c_0=7.0 Bq m^{-3} (Equation 1) gave F_{ISD}=0.39.

The discrepancy between the equilibrium factor values as well as the reason why F_s>F_{ISD} are due to the unexpectedly large measured value of the E_{Tn} relatively to the E_{Rn} (E_{Tn}/E_{Rn}=62.5%). The proposed method by two SSNT detectors (bare and filtered) for the equilibrium factor measurement is suitable only if thoron concentrations could be neglected or the E_{Tn}/E_{Rn} ratio is known. Because of a relatively large decay constant and a low permeability constant of the thoron, the filtered detector records radon concentrations (track density D_0) only, but the bare one registers tracks (D) of the radon and its daughters as well as the thoron and its progeny. Therefore, Equation 5 gives an upper limit of the F values.

Nevertheless, concentration assessments of the ^{212}Bi and precursor thoron gave lower values considering the radon ones. The equation below, as part of Equation 8, gave the ^{212}Bi atom concentration, S m^{-3}, in the air:

$$S\,(^{212}\mathrm{Bi}) = \frac{N_3 - N_2}{R\,Q\,t} \qquad (9)$$

whereas its total activity concentration was $c(^{212}Bi)=\lambda S=1.03$ Bq m^{-3} for $\lambda(^{212}Bi)=1.91\times 10^{-4}s^{-1}$.

Considering the obtained concentration of ^{212}Bi, that is the third thoron progeny ($^{220}Rn \rightarrow {}^{216}Po \rightarrow {}^{212}Pb \rightarrow {}^{212}Bi \rightarrow ({}^{212}Po \rightarrow){}^{208}Pb$), concentrations of the precursors are, of course, higher than the ^{212}Bi one or equal, in their mutual equilibrium. Therefore, at position T ($F_{Rn}=0.48$ and $F_{Tn}<1$) the thoron concentration was higher by 15 per cent than the radon concentration.

Preliminary measurements of the indoor radon concentration and of the thoron one only (without data of the progeny), performed with a Radhome semiconductor detector (CRPM, France), showed that the thoron concentration was nearly 20 per cent of the radon one.

CONCLUSION

During 1990, measurements of the indoor radon concentrations at the Osijek locations B, R and T with LR-115 nuclear track detectors, gave the average concentrations of 9.8, 58.2 and 11.7 Bq m^{-3}, the equilibrium factors of 0.52, 0.46 and 0.48 and the effective dose equivalents of 0.41, 2.16 and 0.53 mSv y^{-1}, respectively.

The indoor alpha potential energy of the radon and thoron progenies was measured by an ISD detector at position T and the respective values were $E_{Rn}=1.52\times 10^{-8}$ J m^{-3} and $E_{Tn}=9.50\times 10{-9}$ J m^{-3}. The equilibrium factor calculated by means of the E_{Tn} value was 0.39 or 19 per cent lower than the one determined by two LR-115 (bare and filtered) detectors. The reason for the discrepancy could lie in a high level of the thoron concentration because the bare detector records all the alpha particle tracks within the detector energy window.

Independent measurements, performed by a Radhome semiconductor detector, showed that the thoron indoor concentration was about 20 per cent of the radon one.

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REFERENCES

1. *International Commission for Radiation Protection.* Lung cancer risk from indoor exposure radon daughters. ICRP Publication 50, Oxford: Pergamon Press, 1989.
2. *Planinić J, Faj Z.* Equilibrium factor and dosimetry of Rn by a nuclear track detector. Health Phys 1990;59:349–51.
3. *Planinić J.* Seasonal variations of radon in Osijek. Working papers, Pecs (Hungary): Pecs-Osijek, Faculty of Education, 1987;1:15–24.
4. *Planinić J, Faj Z.* The equilibrium factor between radon and its daughters. Nucl Instrum Meth 1989;A278:550–52.
5. *Swedjemark GA.* The equilibrium factor F. Health Phys 1983;45:453–62.
6. *Faj Z, Planinić J.* Dosimetry of radon and its daughters by two SSNT detectors. Radiat Prot Dosim 1991;35:265–68.
7. *Makelainen I.* Calibration of bare LR-115 film radon measurements in dwellings. Radiat Prot Dosim 1984;7:195–97.
8. *Mauricio CLP, Tauhata L, Bertelli L.* Internal dosimetry for radon daughters. Radiat Prot Dosim 1985;11:249–55.

9. *World Health Organization.* Indoor air quality. Radon – report on a WHO working group. J Environ Radioactivity 1988;8:73–91.
10. *Duport P, Medalaine G, Zettwoog P, Pineau JF.* Energistrement des rayonnements alpha dans le dosimetre individuel et le dosimetre de site du Comissariat a l'Energie Atomique. Proceedings of the 10th International Conference on Solid State Nuclear Track Detectors. Nucl Tracks Suppl 1979;2:609.

Sažetak

MJERENJE KONCENTRACIJE AKTIVNOSTI TORONA I RADONA POMOĆU DETEKTORA NUKLEARNIH TRAGOVA

Pomoću detektora nuklearnih tragova LR-115 izmjerene su koncentracije aktivnosti radona u zraku na tri lokacije u Osijeku i određene su vrijednosti ravnotežnoga faktora te efektivni dozni ekvivalenti. Radonske koncentracije u kućama bile su od 9,8 do 58,2 Bq m^{-3}, relativna pogreška metode jetkanja bila je blizu 19%. Potencijalna alfa energija za radonove i toronove potomke izmjerena je pomoću detektora ISD. Nezavisno izvedena mjerenja s poluvodičkim detektorom Radhome pokazala su da koncentracije torona u zraku iznose blizu 20% koncentracije radona.

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Ključne riječi: efektivni dozni ekvivalenti, koncentracije radona u zatvorenim prostorima, ravnotežni faktor