Scientific paper

# Concentrations of $^{222}$Rn and $^{220}$Rn in Indoor Air

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## Abstract

Radon ($^{222}$Rn) and thoron ($^{220}$Rn) were surveyed in indoor air of 15 public buildings and 10 homes in Slovenia in November 2004 by exposing Rn–Tn discriminative etched track detectors. The following ranges of the activity concentration were obtained for public buildings: 98–2680 Bq m$^{-3}$ for $C_{Rn}$, 19–1330 Bq m$^{-3}$ for $C_{Tn}$ and 0.11–0.72 for the $C_{Tn}/C_{Rn}$ ratio. In homes, the $C_{Rn}$ range was 12–374 Bq m$^{-3}$ and all $C_{Tn}$ values, except one of 29 Bq m$^{-3}$, were below the detection limit of 10 Bq m$^{-3}$. Monthly effective doses in public buildings due to Rn ($E_{Rn}$) and Tn ($E_{Tn}$) were in the ranges 0.05–1.45 mSv and 0.01–0.47 mSv, respectively, and in homes, 0.018–0.548 and 0.009–0.095 mSv, respectively. The $E_{Tn}/E_{Rn}$ ratio in public buildings was in the range 0.07–0.47.

**Keywords:** Radon ($^{222}$Rn), thoron ($^{220}$Rn), indoor air, effective dose estimate, radon-prone area

## 1. Introduction

There are three primordial radioactive decay chains in the earth's crust:[1] the thorium chain starting with $^{232}$Th, the uranium chain starting with $^{238}$U, and the actinium chain starting with $^{235}$U (Figure 1). In each chain, decay of radium results in formation of radon with accompanying α particles. Only a fraction of radon atoms (called emanation coefficient or emanation power) succeed in leaving the mineral grain and entering the void space. From there, radon travels through the medium either by diffusion or, more effectively and to longer distances, carried by geogas (dissolved in water, He, $N_2$, $CH_4$, $CO_2$, etc)[2] On its way, it accumulates in underground rooms (mines, karst caves, fissures, basements) and eventually enters the atmosphere, and hence appears in air of our living and working environments. Thus, the level of radon activity in the environment is dependent on (i) the contents of elements of the three decay chains, (ii) radon emanation and (iii) radon exhalation. The emanation depends on the recoil length which is, for $^{222}$Rn atoms, 40–60 nm in minerals and 60 μm in air (and is lower for $^{220}$Rn atoms because of the lower recoil energy),[3] and on the grain size. Exhalation is affected by geophysical and hydro-meteorological parameters.[2] Along its path, radon as a noble gas is not subject to chemical reactions but to α-decay. Over a landscape with normal contents of $^{232}$Th and $^{238}$U (in the range 7–50 Bq kg$^{-1}$),[4] usually only $^{222}$Rn (also called radon, Rn) appears at a measurable level in the environmental air because of its relatively long half-life ($t_{1/2}$ = 3.82 days), as compared with half-life of $^{220}$Rn (also called thoron, Tn, $t_{1/2}$ = 55.6 s) and, especially, half-life of $^{219}$Rn (also called actinon, $t_{1/2}$ = 3.9 s). Thus, $^{222}$Rn activity concentrations are usually found in the following ranges:[4] from several kBq m$^{-3}$ to several MBq m$^{-3}$ in soil gas, from several 10 Bq m$^{-3}$ to several kBq m$^{-3}$ in indoor air (worldwide average is 45 Bq m$^{-3}$), and from several Bq m$^{-3}$ to several 10 Bq m$^{-3}$ in outdoor air (worldwide average is 7 Bq m$^{-3}$).

Although radon is used as a tracer in studying movement of air and water masses on local and global scales,[5] as a tool in mineral exploration,[6] as an indicator of activity of fault zones[7] and as an earthquake precursor,[8] we are mostly concerned about its negative aspects. It has become clear[4] that, on the world wide average, breathing air contaminated by radon and thoron 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 radon is a major cause of lung cancer, second only to cigarette smoking.

In actual fact, it is not radon and thoron themselves, but their short-lived decay products that contribute the great majority of the dose. α decay of Rn ($E_\alpha$ = 5.49 MeV) results into the formation of radioactive aerosols of Rn short-lived decay products (RnDP)[1]: $^{218}$Po ($E_\alpha$ = 6.00 MeV), $^{214}$Pb, $^{214}$Bi and $^{214}$Po ($E_\alpha$ = 7.69 MeV) (Figure 1). Because $t_{1/2}$ of Rn is much longer than $t_{1/2}$ of each RnDP, radioactive equilibrium between Rn and RnDP (when the activity of each RnDP is equal to the activity of Rn) may in principle be reached. Due to air movement and plate-out of aerosols, equilibrium is only partly reached and is expressed by the so called equilibrium factor $F_{Rn}$, a fraction number between 0.20 to 0.80 in the indoor air. If not measured, ICRP[9] recommends $F_{Rn}$ = 0.40 to be taken for dose estimates.

Tn short-lived decay products (TnDP) are formed by α decay of thoron ($E_\alpha$ = 6.29 MeV): $^{216}$Po ($E_\alpha$ = 6.78 MeV), $^{212}$Pb, $^{212}$Bi ($E_\alpha$ = 6.07 MeV) and $^{212}$Po ($E_\alpha$ = 8.78 MeV) (Figure 1). Because $t_{1/2}$ of Tn is much shorter than $t_{1/2}$ of $^{212}$Pb, radioactive equilibrium between Tn and TnDP cannot be reached and the corresponding equilibrium factor $F_{Tn}$ is far lower than $F_{Rn}$, measured values being mostly below 0.20.[10–12] While spatial distributions of Rn, RnDP and TnDP in a room are homogeneous, the concentration of Tn, due to its short half-life, decreases away from the source – either floor, wall or ceiling.[13–15] Therefore the position of the measuring device is very important in obtaining reliable data.

Following breathing in air, RnDP and TnDP are partly deposited on the walls of airways, where the energy released during α decay (energies of β and γ decays are much lower) is absorbed in tissue, and the resulting equivalent dose is responsible for lung cancer.

As in other countries, also in Slovenia great attention has been paid to Rn while Tn has been ignored. Rn, first measured in the air of the Žirovski vrh uranium mine in 1969,[16] has been surveyed nation-wide[17] in almost all kindergartens and schools, in 1000 randomly selected dwellings, all underground mines, 50 karst caves, 5 major spas, 10 major water supply plants, 26 major hospitals, 8 major wineries and some others. In more than 50 buildings, radon problems have been successfully mitigated.[18] In contrary, this is the paper reporting on indoor Tn in Slovenia. Measurements are described, Rn and Tn concentrations in indoor air reported and effective doses commented on.

## 2. Experimental

### 2. 1. Description of Sites

Two kinds of buildings were chosen: 15 public buildings, in which elevated Rn levels had been found by previous measurements,[19] and 10 homes, at sites where regular environmental radioactivity monitoring (including outdoor Rn) is being carried out. Public buildings were: 1 kindergarten, 7 elementary schools, 2 health care centres, 2 railway stations, 1 post office, 1 customs office, 1 re-

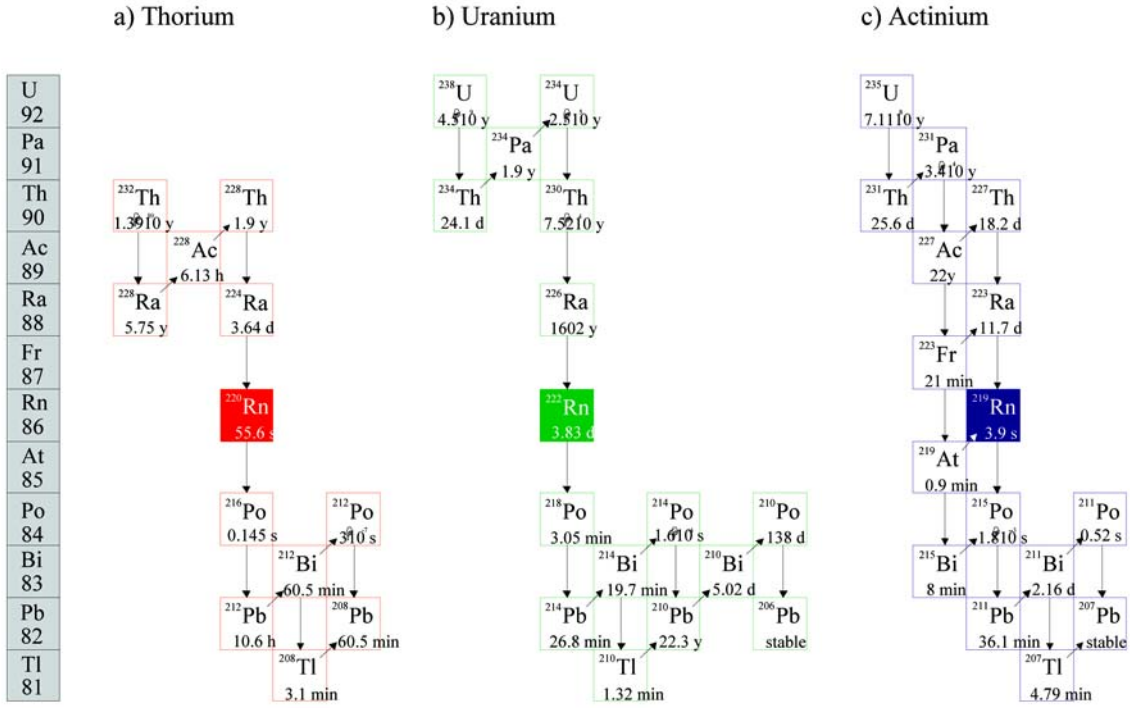


**Figure 1.** Thorium, uranium and actinium decay chains.

search institute. Railway stations were built before 1900, and all other buildings after 1950. All are one-storey buildings, made of stone, brick and concrete. Air conditioning is installed in health centres and customs office, but not in other public buildings or in homes. The latter are heated in winter by water radiators connected to a central heating system based on gas or oil. Measurements were carried out in October–November 2004.

## 2. 2. Measuring Techniques

To measure average Rn and Tn indoor air concentrations, UFO Rn–Tn discriminative dosimeters were used, provided by the Vinča Institute of Nuclear Sciences, Beograd, Serbia. After exposure, dosimeters were sent back to Beograd for development and evaluation. The dosimeter is composed of two hemispherical chambers connected by a 1-mm hole in the centre, and each fitted with a polycarbonate film. The film in one chamber detects both Rn and Tn, while the other only Rn. At the end of measurement, films are first etched chemically and then electrochemically, under conditions to show tracks caused predominantly by Rn on one film, and those caused by Tn on the other. The dosimeter is described in detail elsewhere.[13]

In several buildings Rn and Tn were also measured continuously with the RTM 2010 Rn/Tn monitoring system (Sarad, Germany), based on α spectrometry. Air is pumped through the chamber at a flow rate between 0.5 and 2.5 $dm^3\ min^{-1}$. Because of the high voltage between the chamber wall and the silicon detector, the positively charged $^{218}Po$ and $^{216}Po$, created by Rn and Tn decays, respectively, after they have entered the chamber, are deposited on the detector where they are analysed and the concentrations of their parents calculated. The frequency of analysis and data reporting can be set from once a minute to once an hour.

# 3. Results and Discussion

## 3.1. Rn and Tn Levels

Rn and Tn concentrations ($C_{Rn}$ and $C_{Tn}$, respectively) in indoor air of public buildings are listed in Table 1 (two readings failed) and those of homes in Table 2 (one reading failed). These are average values for the duration of exposure of the detector. $C_{Rn}$ values are similar to those obtained previously at the same places. $C_{Tn}$ values are seen to be significant fraction of $C_{Rn}$, the $C_{Tn}/C_{Rn}$ ratios varying from 0.11 to 0.72, with a geometric mean of 0.40 and geometric standard deviation 1.66 (Figure 2). On the other hand, Rn levels in homes were low, in only one case $C_{Rn}$ exceeding 100 Bq $m^{-3}$. Practically all $C_{Tn}$ values were below the detection limit.

In public building #13, Rn and Tn were continuously recorded for a month. Hourly values of $C_{Rn}$, $C_{Tn}$ and $C_{Tn}/C_{Rn}$ are shown in Figure 3. Since the measurement period was different from that in Table 1, values in Figure 3a cannot be compared with those in the Table.

Although both $C_{Rn}$ and $C_{Tn}$ show pronounced diurnal variations, their fluctuations differ, and the resulting $C_{Tn}/C_{Rn}$ ratio is far from being constant but changes from 0.05 to 0.70. This finding, though very interesting, cannot be interpreted at the moment and more experiments will be needed to understand it.

While elevated Rn levels in public buildings have been explained in terms of geology and building quality,[17,19] at the present time, we have no firm understanding of elevated Tn levels. Neither can we explain the value of $C_{Tn}/C_{Rn}$ ratio. In order to obtain the necessary knowledge, more systematic Tn measurements will be needed, carried out in various buildings at places of different geological characteristics and under different meteorological conditions. In addition to measuring average concentrations over longer periods (as done in this

**Table 1.** Rn ($C_{Rn}$) and Tn ($C_{Tn}$) concentrations in public buildings

| Number | Public building at | Duration | $C_{Rn}$ / Bq $m^{-3}$ | $C_{Tn}$ / Bq $m^{-3}$ |
|---|---|---|---|---|
| 1 | Jožef Stefan Institute Ljubljana | 11.08. – 29.11.04 | 98 ± 8 | 19 ± 30 |
| 2 | Unec | 26.10. – 25.11.04 | 655 ± 51 | 400 ± 191 |
| 3 | Babno Polje | 26.10. – 25.11.04 | 392 ± 33 | 210 ± 121 |
| 4 | Idrija | 26.10. – 26.11.04 | 236 ± 22 | 100 ± 78 |
| 5 | Ljubljana | 27.10. – 22.11.04 | 860 ± 66 | 308 ± 245 |
| 6 | Ribnica | 28.10. – 25.11.04 | 1710 ± 120 | 996 ± 472 |
| 7 | Novo mesto | 28.10. – 17.11.04 | 2680 ± 190 | 1330 ± 735 |
| 8 | Kranj | 29.10. – 18.11.04 | 1210 ± 90 | 530 ± 345 |
| 9 | Radovljica | 29.10. – 18.11.04 | 711 ± 57 | 264 ± 212 |
| 10 | Komen–1 | 02.11. – 24.11.04 | 452 ± 39 | – |
| 11 | Komen–2 | 02.11. – 24.11.04 | 334 ± 31 | – |
| 12 | Divača–1 | 02.11. – 24.11.04 | 973 ± 75 | 108 ± 275 |
| 13 | Divača–2 | 02.11. – 24.11.04 | 932 ± 72 | 342 ± 268 |
| 14 | Kozina | 02.11. – 24.11.04 | 254 ± 25 | 183 ± 91 |
| 15 | Sežana | 02.11. – 24.11.04 | 633 ± 51 | 335 ± 191 |

**Table 2.** Rn ($C_{Rn}$) and Tn ($C_{Tn}$) concentrations in homes

| Number | Public building at | Duration | $C_{Rn}$/Bq m$^{-3}$ | $C_{Tn}$/Bq m$^{-3}$ |
|---|---|---|---|---|
| 1 | Trbovlje | 18.03. – 21.06.05 | 12 ± 2 | < 10 |
| 2 | Koèevje | 21.03. – 17.06.05 | 374 ± 28 | – |
| 3 | Novo mesto | 21.03. – 04.07.05 | 70 ± 6 | < 10 |
| 4 | Ribnica na Pohorju | 23.03. – 01.07.05 | 30 ± 4 | < 14 |
| 5 | Maribor | 23.03. – 01.07.05 | 31 ± 4 | < 26 |
| 6 | Idrija | 31.03. – 17.06.05 | 61 ± 6 | < 10 |
| 7 | Murska Sobota | 23.03. – 01.07.05 | 54 ± 5 | < 10 |
| 8 | Ljubljana | 29.03. – 21.06.05 | 28 ± 4 | < 10 |
| 9 | Lokev pri Se ani | 30.03. – 17.06.05 | 88 ± 8 | < 10 |
| 10 | Todra | 01.04. – 21.06.05 | 53 ± 6 | 29 ± 20 |

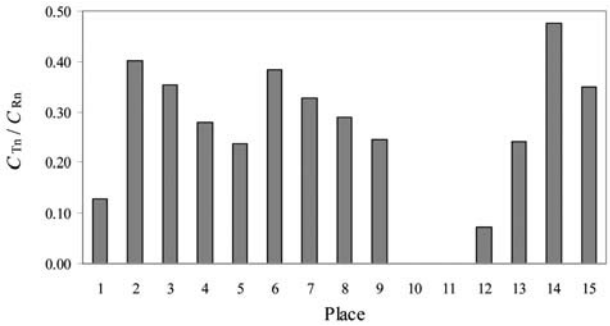


**Figure 2.** $C_{Tn}/C_{Rn}$ ratios in public buildings, measured in October–November 2004.

study), it will be also necessary to follow $C_{Rn}$ and $C_{Tn}$ at hourly intervals. From their diurnal variations, dependence will be obtained not only on the meteorological parameters but also on the heating and ventilation regime, and on working regime in public buildings and living habits in homes.

## 3. 2. Effective Doses Due to Rn and Tn Decay Products

Effective doses are calculated by using dose conversion factors which convert exposure to Rn and Tn, expressed in WLM or J h m$^{-3}$, into the effective dose in mSv gained during the period of exposure. WLM (working-level-month) is an old but still widely used unit, for practical reasons. An exposure of 1 WLM is gained by 170-hour breathing in air with either RnDP or TnDP concentration of 1 WL, where 1 WL means concentrations of RnDP or TnDP that result into concentrations of potential α energy of $1.3\times10^8$ MeV m$^{-3}$. $C_{Rn}$ and $C_{Tn}$ are converted into WL for Rn ($WL_{Rn}$) and Tn ($WL_{Rn}$) by using equations:[10,11]

$$WL_{Rn} = C_{Rn}\times F_{Rn}/3700$$

$$WL_{Tn} = C_{Tn}\times F_{Tn}/275,$$

where $F_{Rn}$ and $F_{Tn}$ are equilibrium factors of RnDP and TnDP, respectively. While ICRP methodology[12] recommends $F_{Rn} = 0.40$ for dose calculations, there is no such recommendation for $F_{Tn}$. In addition, $F_{Tn}$ cannot be defined simply since TnDP are homogeneously distributed in a room while Tn is not. Depending on ventilation rate, it may attain very different values,[11] but mostly they are much lower than $F_{Rn}$ values.[10,125] Therefore for correct dose estimates the concentration of TnDP ($C_{TnDP}$), rather than that of Tn, should be used.[20] Not having $C_{TnDP}$ data in our study, we took as a compromise $F_{Tn} = 0.07$,[10] which is close to the mean value measured in India.[11] Effective doses due to RnDP ($E_{Rn}$) and TnDP ($E_{Tn}$) were then calculated using the expressions:

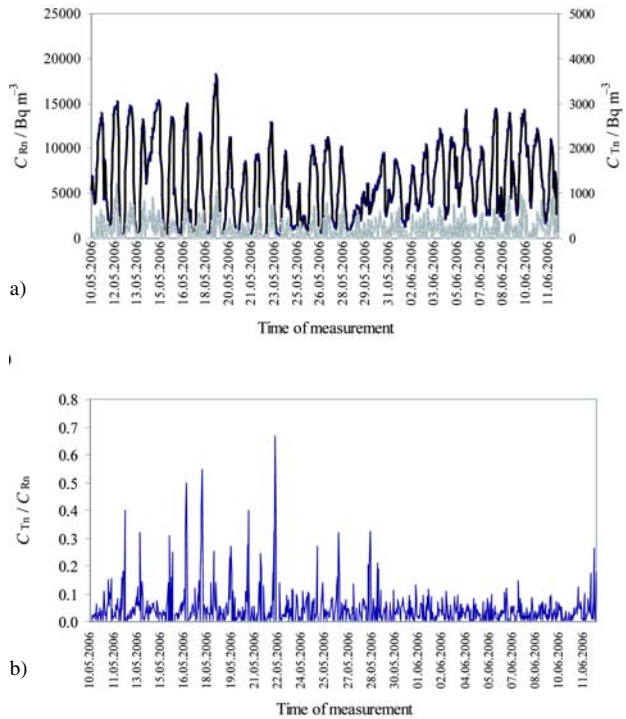


**Figure 3.** Results of continuous monitoring of Rn and Tn in public building #13 from May 10 to June 11, 2004, using RTM-2010 device: a) $C_{Rn}$ and $C_{Tn}$ time series; b) $C_{Tn}/C_{Rn}$ ratio time series.

$$E_{Rn} = WL_{Rn} \times (t/170) \times DCF_{Rn}$$

$$E_{Tn} = WL_{Tn} \times (t/170) \times DCF_{Tn}.$$

Above, $t$ is the time in hours spent by a person in the room surveyed, and $DCF_{Rn}$ and $DCF_{Tn}$ are the dose conversion factors for Rn and Tn, respectively. Doses were calculated for a month, so 170 hours were taken for workplaces in public buildings and 576 hours for homes, taking into acount 0.80 for the occupancy factor at home.[21] ICRP[9] recommends a value for $DCF_{Rn}$ of 4 mSv WLM$^{-1}$ for homes and 5 mSv WLM$^{-1}$ for workplaces. Based on an estimated ratio $DCF_{Rn}/DCF_{Tn}$ of 3.5–3.6,[20,22] in our dose calculations 1.1[22] mSv WLM$^{-1}$ and 1.4 mSv WLM$^{-1}$ were taken for Tn in homes and workplaces, respectively.

**Table 3.** Monthly effective doses due to Rn ($E_{Rn}$) and Tn ($E_{Tn}$) in public buildings

| Number | $E_{Rn}$/mSv mo$^{-1}$ | $E_{Tn}$/mSv mo$^{-1}$ | $E_{Tn}/E_{Rn}$ |
|---|---|---|---|
| 1 | 0.05 | 0.01 | 0.13 |
| 2 | 0.35 | 0.14 | 0.40 |
| 3 | 0.21 | 0.07 | 0.35 |
| 4 | 0.13 | 0.04 | 0.28 |
| 5 | 0.46 | 0.11 | 0.24 |
| 6 | 0.92 | 0.35 | 0.38 |
| 7 | 1.45 | 0.47 | 0.33 |
| 8 | 0.65 | 0.19 | 0.29 |
| 9 | 0.38 | 0.09 | 0.24 |
| 10 | 0.24 | – | – |
| 11 | 0.18 | – | – |
| 12 | 0.53 | 0.04 | 0.07 |
| 13 | 0.50 | 0.12 | 0.24 |
| 14 | 0.14 | 0.07 | 0.47 |
| 15 | 0.34 | 0.12 | 0.35 |

**Table 4.** Monthly effective doses due to Rn ($E_{Rn}$) and Tn ($E_{Tn}$) in homes

| Number | $E_{Rn}$/mSv mo$^{-1}$ | $E_{Tn}$/mSv mo$^{-1}$ | $E_{Tn}/E_{Rn}$ |
|---|---|---|---|
| 1 | 0.018 | <0.009 | |
| 2 | 0.548 | – | |
| 3 | 0.103 | <0.009 | |
| 4 | 0.044 | <0.013 | |
| 5 | 0.045 | <0.025 | |
| 6 | 0.089 | <0.009 | |
| 7 | 0.079 | <0.009 | |
| 8 | 0.041 | <0.009 | |
| 9 | 0.129 | <0.009 | |
| 10 | 0.078 | 0.028 | 0.35 |

The monthly effective doses thus calculated for public buildings and homes are listed in Tables 3 and 4, respectively. The $E_{Rn}$ values in public buildings range from 0.05 mSv to 1.45 mSv with a geometric mean of 0.32 mSv and geometric standard deviation 2.32, and $E_{Tn}$ values

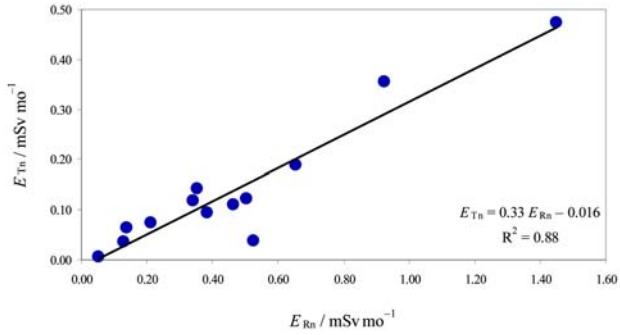


**Figure 4.** Relationship between monthly effective doses due to Tn and to Rn in public buildings.

range from 0.01 mSv to 0.47 mSv with a geometric mean of 0.09 mSv and geometric standard deviation 1.38. Thus, $E_{Tn}$ is a significant fraction of $E_{Rn}$, being on average about 1/3 $E_{Rn}$, as seen from Figure 4.

As expected, based on low Rn and Tn concentrations in homes, both $E_{Rn}$ and $E_{Tn}$ values are low there, in the range 0.018–0.129 mSv mo$^{-1}$ for Rn, while for Tn, except the value of 0.028 mSv mo$^{-1}$ all the other were below 0.025 mSv mo$^{-1}$. This situation cannot be taken as the average situation in the Slovenian homes, because the sample of 10 homes was certainly too small to be representative. Effective doses in public buildings were higher: at place 7, the monthly Rn dose even exceeding the world annual average of 1.2 mSv.[4] The $E_{Tn}/E_{Rn}$ ratio lies in the range 0.11–0.72, with a geometric mean of 0.40 and geometric standard deviation 1.66. These figures do not indicate an alarming Tn situation but rather rank Slovenia among those countries in which Tn levels are not negligible[20] and should be considered seriously in dose estimates. This will certainly be done in our future work and also the effective doses obtained so far at the radon-prone areas will be re-evaluated. A study is currently underway in which both $C_{Rn}$ and $C_{Tn}$ are monitored indoors in 150 rooms of some 50 buildings, and outdoors at 60 locations at which regular environmental radioactivity monitoring is being carried out.

## 4. Conclusion

Thoron and radon concentrations in indoor air of public building were in the range 19–1330 Bq m$^{-3}$ and 98–2680 Bq m$^{-3}$, respectively, and their ratio in the range 0.11–0.72. Radon concentrations in homes were in the range 12–374 Bq m$^{-3}$, while thoron concentrations were all, except for the value of 29 Bq m$^{-3}$, below the detection limit of 10 Bq m$^{-3}$. In public buildings, monthly effective doses due to thoron were in the range 0.01–0.47 mSv and those due to radon, in the range 0.05–1.45 mSv, with their ratios in the range 0.07–0.47. For homes, only one value of the monthly effective doses due to thoron was

0.028 mSv, while all the others below 0.025 mSv, and due to radon they were in the range of 0.018–0.129 mSv. Based on these figures, Slovenia may be considered as a country with moderate thoron levels. Nonetheless, in our future work thoron will not be ignored and the contribution from thoron will be taken into account in dose estimates.

## 5. Acknowledgments

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## Povzetek

Novembra 2004 smo preiskali radon ($^{222}$Rn) in toron ($^{220}$Rn) v zraku 15 javnih zgradb in 10 domov po Sloveniji z Rn-Tn detektorji jedrskih sledi. V javnih zgradbah smo dobili $C_{Rn}$ v območju 98–2680 Bq m$^{-3}$ in $C_{Tn}$ v območju 19–1330 Bq m$^{-3}$ z razmerjem $C_{Tn}/C_{Rn}$ v območju 0,11–0,72. V domovih smo našli $C_{Rn}$ v območju 12–374 Bq m$^{-3}$, $C_{Tn}$ pa razen v enem primeru s $C_{Tn}$ 29 Bq m$^{-3}$, pod mejo detekcije 10 Bq m$^{-3}$. Mesečne efektivne doze zaradi prisotnosti radona ($E_{Rn}$) in torona ($E_{Tn}$) so bile v javnih zgradbah v območjih 0,05–1,45 mSv in 0,01–0,47 mSv, v domovih pa v območjih 0.018–0.548 in 0.009–0.095 mSv. Razmerje $E_{Tn}/E_{Rn}$ v javnih zgradbah je bilo 0,07–0,47.