

EXPERIMENTAL AND MODELING STUDIES OF THORON DECAY PRODUCTS IN OUTDOOR AIR NEAR THE GROUND SURFACE*

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ABSTRACT

To better understand the physical behavior of thoron decay products in outdoor air at ground level, and the resulting exposure-dose coefficient, measurements of the concentrations of thoron (^{220}Rn) gas and its short-lived decay products (^{216}Po , ^{212}Pb , ^{212}Bi) in attached-to-aerosol and unattached states were carried out at a semi-arid field site in central New Mexico under varying atmospheric conditions. A high volume air sampler (100 L min^{-1}) with a single 635 mesh stainless steel screen (50% penetration for 4 nm particles) and a glass fiber filter were employed. Analysis of the 105 outdoor measurements yielded the following average values: 9 Bq m^{-3} for the ^{220}Rn activity concentration; 22 nJ m^{-3} for the concentration of potential alpha energy, 0.03 for the equilibrium factor, 0.15 for the unattached fraction of potential alpha energy, and 14 nSv h^{-1} for the effective dose rate. The activity concentration of thoron gas and the unattached fraction of potential alpha energy are similar to those of radon and its decay products at this site but the equilibrium factor and dose rate are smaller. The thoron decay products measurements are interpreted using the computer code TPOUT, which applies one-dimensional eddy transport to calculate the vertical concentrations of ^{220}Rn and its decay products as a function of atmospheric stability, aerosol concentration, terrain roughness, and surface wind speed. This code successfully predicted the observed trends of unattached and attached decay products concentrations with height and other factors. For example, the analysis showed that the primary reason for the very small equilibrium factor is the mixing of ^{212}Pb to much greater heights than the corresponding mixing for the parent isotope ^{220}Rn . The resulting effective dose rate is found to be relatively invariant with dry deposition factors such as terrain roughness, surface wind speed, and even the ambient concentration of aerosol particles. Projection of the present results to conditions typical of temperate climates suggests an average annual outdoor effective dose from thoron decay products of about 0.025 mSv , which is a significant component of the total outdoor dose.

INTRODUCTION

Although the radon isotope ^{220}Rn (thoron) has been a traditional object of study in atmospheric science (Jacobi and Andre, 1963; Butterweck *et al.*, 1994; Doi and Kobayashi, 1994), it has received relatively less attention than the isotope ^{222}Rn (radon) in relation to environmental dose studies. It has rather been assumed, based on little experimental study, that the dose from thoron decay products outdoors can be neglected in comparison with that from radon decay products.

The United Nations Report of 1982 (UNSCEAR, 1982) estimated that the activity concentration of thoron decay products outdoors was about a factor of 10 lower than that of radon decay products, but did not attempt to assess the comparative doses. For indoor air the report estimated that the contribution from ^{220}Rn decay products to the effective dose was on the average one-fifth of that from ^{222}Rn decay products. The United Nations Report of 1993 (UNSCEAR, 1993) provided some additional information about thoron in outdoor air. It acknowledged that the activity concentration of thoron gas close to the surface of the ground in many cases can exceed that of radon. However, for dose estimation the report recommended the assumption of the same average concentration

of 10 Bq m^{-3} for both thoron and radon gas in outdoors at the human breathing level. Schery and Grumm (1992) reviewed the subject of environmental thoron, but could not find sufficient information to assess outdoor dose. Tso and Li (1987) measured thoron decay products concentrations outdoors in Hong Kong, but without particle size information their dose estimates were very approximate. Reineking *et al.* (1992) reported outdoor measurements of thoron decay products. Their measurements are some of the few to provide particle size information important for assessing dose, although their paper did not deal directly with this subject.

The research reported here is aimed at fulfilling the need for more comprehensive measurements of thoron and its decay products at an outdoor site. Such data should help in better understanding the physical behavior of thoron and its decay products under varying meteorological conditions, and assessing the dose delivered to the human lungs. We include measurements of thoron decay products in both the attached-to-aerosol and unattached states, and apply dose conversion factors derived from current recommendations of the International Commission on Radiological Protection (ICRP) to estimate the resulting effective dose. Auxiliary data for meteorological conditions are also given, to enable comparisons to be drawn with model predictions of the one-dimensional eddy correlation code TPOUT, a version of the radon code RPOUT (Schery *et al.*, 1992) modified for application to thoron and its decay products. Once calibrated with these data (from the studied site at Socorro, NM), the model can be used to project thoron decay products concentration and effective dose under conditions at other sites that may be different.

MATERIALS AND METHODS

Site

The measurements were performed at a field site (Schery *et al.* 1984; Wasiolek and Schery, 1993) about 1 km to the west of the New Mexico Tech campus in Socorro, New Mexico during February of 1994 and February of 1995. The site is located at an altitude of about 1400 m at the edge of the Rio Grande Valley near the slopes of the Socorro Peak. There are a few one story buildings within 500 m of the site. The climate is semi-arid (rainfall averages about 20 cm y^{-1}) and vegetation is sparse, limited to scattered bushes and clumps of dry grass. The soil is a gravelly sandy loam common for alluvial deposits along the Rio Grande, with a surface ^{224}Ra content of about 44 Bq kg^{-1} and average thoron flux density of about $2.5 \text{ Bq m}^{-2} \text{ s}^{-1}$. The water table is about 30 m below the surface. The monitoring of several meteorological variables during the measurements yielded the values presented in Table 1. There was no precipitation of any kind during the measurement periods.

The sampling of the thoron and thoron decay products was performed at 1 m above ground, whereas the meteorological sensors were located at 5 m, on a meteorological mast to more efficiently characterize atmospheric conditions.

Thoron Gas

The ^{220}Rn concentrations were measured semi-continuously with a custom built two-filter system (Grumm *et al.*, 1990). The system consists of a 79-L decay chamber with fixed inlet filter (Type A/E, Gelman Sciences¹) and movable exit filter (strip of Versapore acrylic copolymer with about $1.2 \mu\text{m}$ pore diameter). The inlet filter removes all thoron decay products from the incoming air stream. The exhaust filter collects the decay products from thoron decays in the chamber. Alpha spectrometry with a 900-mm² surface barrier detector is performed on the exit filter, to estimate the concentration of thoron without interference from radon decay products. The sampling protocol of 2-h sampling and 2-h counting yields a sensitivity of $1.25 \text{ counts h}^{-1}$ per Bq m^{-3} of thoron gas. The calibration of the system is performed on a routine basis with a Pylon² flow-through thoron source (Model Th-1025.) that has also been checked against equipment at other laboratories. Statistical counting error for a typical

¹GelmanSciences, 600 South Wagner Road, Ann Arbor, MI 48106.

²Pylon Electronic, 147 Colonnade Road, Ottawa Ontario, K2E 7L9

two-hour measurement at a thoron level of 10 Bq m^{-3} is $\pm 20\%$.

Thoron Decay Products

Relevant decay information for thoron and its short-lived decay products is presented in Table 2. In terms of radiation type and order of decay there are similarities between the thoron and radon decay chains. However, the short-lived radon decay products have half-lives less than 30 min each, whereas one of the thoron decay products, ^{212}Pb , has a much longer half-life of 10.6 h. Measurements of the activity-weighted size distribution for ^{212}Pb under laboratory conditions (Cheng *et al.*, 1992) yielded two distinctive size modes, one less than 2-nm diameter, and a second around 100-200 nm; commonly classified as the unattached fraction and the attached-to-aerosol fraction, respectively. In the present measurements, a single stainless steel (SS) wire mesh screen³ was used to separate unattached from attached thoron decay products. The combination of a 8.9-cm-diameter screen of 635 mesh number and a 100 L min^{-1} flow rate yielded 50% penetration, $d_p(50\%)$, for 4-nm diameter particles. Such a selection of the $d_p(50\%)$ assured almost complete (98%) collection of 1.1-nm diameter particles which are assumed to represent the median size of unattached activity, as discussed by Reineking and Porstendörfer (1990). The collection efficiency curve for the 0.2-500 nm particle-size range is presented in Fig. 1. The front side of the screen and the backup filter (Type A/E, Gelman Sciences) were counted for 8 hours, in 1-min intervals, using commercial detectors⁴ (10-cm ZnS(Ag) scintillator + photomultiplier tube, Model 43-1, and automatic counters, Model 2000, Ludlum Measurements) interfaced to a personal computer. Multiple counting systems were used to allow three samples (consisting of both a screen and filter) to be taken every day, between 7:00-8:00, 13:00-14:00 and 20:00-21:00 mountain standard time (MST), respectively. The sampling time was chosen to cover different atmospheric stability conditions, which were typically unstable during most of the day and more stable during the night.

The alpha-count data for the screen and filter combinations were analyzed for individual airborne decay products concentrations, and the airborne potential alpha energy concentrations (PAEC), using the computer code of Knutson (1989) based on the Expectation-Maximum Likelihood (E-M) algorithm by Maher and Laird (1985). Because the screens were counted from one side, correction factors for front-to-total activity and screen losses were applied (Solomon and Ren, 1992). The counting efficiency was obtained for the counting systems with a calibrated ^{230}Th 10-cm-diameter alpha source⁵ and varied from 0.29 to 0.31.

The potential alpha-energy concentration (PAEC), which is the amount of alpha-particle energy ultimately released through decay to ^{208}Pb of the thoron decay products present per unit volume of air, was calculated from the individual decay products concentrations as follows,

$$\text{PAEC} = 5.32 \times 10^{-4} \times C(^{216}\text{Po}) + 69.1 \times C(^{212}\text{Pb}) + 6.56 \times C(^{212}\text{Bi})$$

(1)

where, PAEC is in nJ m^{-3} and decay products concentrations, $C(X)$, are in Bq m^{-3} . The contribution of ^{212}Po is much smaller and is neglected here. The disequilibrium factor, F , between the decay products and the parent thoron gas concentration was calculated by,

where, PAEC is in nJ m^{-3} and concentration of thoron $C(^{220}\text{Rn})$ is in Bq m^{-3} . The unattached fraction of the PAEC, f_p , is expressed as the ratio of the potential alpha activity measured by the screen (corrected for screen collection

³Tetko, Inc., P.O. Box 346, Lancaster, NY 14086.

⁴Ludlum Measurements, Inc., 501 Oak St., Sweetwater, TX 79556.

⁵Eberline Instrument Corporation, P.O. Box 2108, Santa Fe, NM 87504

$$F = 0.0133 \times \frac{\text{PAEC}}{C(^{220}\text{Rn})}$$

(2)

efficiency, screen losses and front-to-total emission ratio) to the total potential alpha activity (including that on the backup filter).

Other Atmospheric Measurements

The meteorological observations of wind speed, air temperature, relative humidity and barometric pressure were averaged at 1 hour intervals. Commercial sensors (Model 107 temperature probe⁶, Model 03001-5 Yung wind vane⁶, barometer and hygrometer⁷, were mounted at 5 m on a mast with outputs connected to a data logger⁶ (Model 21X). Based on the standard deviation of the horizontal wind direction (EPA, 1986) the atmospheric stability classes according to Pasquill classification (extremely unstable to extremely stable, A=1 to G=7) were assigned for the individual sample periods. Particle concentration was monitored with a calibrated Rich 200 particle counter⁸ for which the manufacturer specifies a size range sensitivity of 0.0016 μm and larger.

Dosimetry

Doses from inhalations of thoron decay products are calculated by the same methods as those from radon decay products. To evaluate lung dose rate as a function of the measured values of PAEC and f_p , we used the new ICRP lung model (ICRP, 1994a). Regional lung deposition of the unattached and attached fractions of airborne thoron decay products were calculated using the software code LUDEP (Jarvis *et al.*, 1993). The resulting doses to target cells in the bronchi, bronchioles, and alveoli (per unit exposure), and the corresponding effective dose, were calculated as described elsewhere (Birchall and James, 1994; James, 1994). According to the ICRP's new model for lung dosimetry, the risk from irradiating lung tissues is determined by averaging the doses received by target cells in these three distinct tissue regions. The absorbed dose (in Gy), is calculated for each tissue. For the alpha particle radiation emitted by the thoron decay products, the absorbed dose is multiplied by the radiation weighting factor of 20, to give the "equivalent" dose (in Sv) received by each tissue. The equivalent dose received by the lungs as a whole, H_{lung} , is then given by averaging the equivalent doses received by the constituent tissues.

The effective dose, E_{lung} , for a member of the public, which is a measure of the overall risk of lung cancer (and is also expressed in Sv), is given by,

$$E_{\text{lung}}(\text{public}) = H_{\text{lung}} \times w_{\text{lung}} \times 0.3 \times 0.77,$$

(3)

where, w_{lung} is the tissue weighting factor (risk factor) of 0.12 for the lungs, the factor of 0.3 corrects for the overestimation of lung cancer risk (compared to that determined from epidemiological studies) obtained when the effective dose is calculated for a uranium miner (Birchall and James, 1994; James, 1994), and the factor of 0.77 corrects for the greater risk per unit of effective dose assumed to apply for a member of the public compared to a worker (ICRP, 1991; 1994b; see also Hopke *et al.* in press; Wasiolek and James, in press).

To represent outdoor exposure, a mean breathing rate of 1.2 $\text{m}^3 \text{h}^{-1}$ was assumed. This is ICRP's

⁶Campbell Scientific, Inc. P.O. Box 551, Logan, UT 84321.

⁷Davis Instruments, 3465 Diablo Ave., Hayward, CA 94545.

⁸Environment One Corporation, 2773 Balltown Road, Schenectady, NY 12309.

recommended value for an adult male (ICRP, 1994a). Fig. 2 shows the dose conversion coefficients in sievert (Sv) per unit potential alpha energy exposure, PAEE (in J h m⁻³), as functions of the activity median thermodynamic diameter (AMTD) of the associated aerosols, assuming typical decay products disequilibria of 1 : 0.0002 : 0 for the unattached fraction, and 0 : 0.02 : 0.005 for the attached fraction.

To calculate the effective dose, E_{lung}, from the experimental data the following expression was used,

$$E_{\text{lung}}(\text{public}) = [f_p \times \text{DCF}_u + (1 - f_p) \times \text{DCF}_a] \times \text{PAEC},$$

(4)

In this study we assumed that "unattached" thoron decay products are characterized by particles of diameter 1.1 nm (NRC, 1991; Reineking and Porstendörfer, 1990). The resulting dose conversion factor, DCF_u, applied to the measured airborne concentration of unattached potential alpha energy was 2.7 Sv per J h m⁻³ (corresponding to 9.76 mSv WLM⁻¹). To characterize the "attached" fraction of potential alpha energy, we assumed an aerosol with activity median thermodynamic diameter (AMTD) of 0.22 μm. This is based on a "typical" AMTD of 0.15 μm found for radon decay products in indoor air (NRC, 1991), with an assumed hygroscopic growth of approximately 50% in the respiratory tract. The resulting dose conversion factor, DCF_a, applied to the measured airborne concentration of attached potential alpha energy was 0.38 Sv per J h m⁻³ (corresponding to 1.37 mSv WLM⁻¹).

Model for Outdoor Decay Products Concentrations

The model calculations predicting concentrations of unattached and attached thoron decay products were performed with the computer code TPOUT. This is a BASIC program designed to calculate the vertical distribution of thoron and its decay products outdoors based on observable environmental conditions. This one-dimensional eddy-transport code is a modified version of the earlier code RPOUT (Schery *et al.*, 1992; Schery and Wasiolek, 1993) dealing with radon and its decay products. The steady-state vertical distribution of thoron and its unattached and attached decay products are calculated based on thoron flux from soil, the turbulent diffusivity K(z), the aerosol concentration, terrain roughness and meteorological conditions. As an example, if n₀(z) is the atomic thoron concentration as a function of height, the unattached n_{1u}(z) and attached n_{1a}(z) concentrations of ²¹⁶Po are calculated from,

$$\lambda_0 n_0(z) + \frac{\partial}{\partial z} \left(K(z) \frac{\partial n(z)_{1u}}{\partial z} \right) - \lambda_1 n(z)_{1u} - \chi n(z)_{1u} = 0$$

(5a)

$$\chi n(z)_{1u} + \frac{\partial}{\partial z} \left(K(z) \frac{\partial n(z)_{1a}}{\partial z} \right) - \lambda_1 n(z)_{1a} = 0,$$

(5b)

where, λ₀ is the decay constant for thoron, λ₁ is the decay constant for the ²¹⁶Po, χ is the aerosol attachment rate, and z is the height above ground. The full equations for the analogous problem of radon decay products (only the value of the decay constants must change) are given in Schery and Wasiolek (1993). The atmospheric model assumes the existence of a thin laminar sub-layer just above the ground, where K(z) assumes the single-particle

diffusion coefficient as appropriate for the unattached or attached modes. The terrain roughness, atmospheric stability and wind speed, are used to calculate the sub-layer depth and deposition velocity. After the program finds the sub-layer depth and the deposition velocity, the single-particle diffusion coefficients for the unattached and attached decay products must be entered. The program also requires the input of the flux density of thoron gas from soil, aerosol concentration (which is assumed constant with height), and upper boundary height. The major difference between TPOUT and RPOUT is that TPOUT uses a finer grid spacing to deal with the larger decay constants of ^{220}Rn and ^{216}Po , and the upper boundary conditions for ^{212}Pb must be modified to take into account that its half-life is much larger than that for ^{220}Rn and ^{216}Po , causing ^{212}Pb to penetrate to much greater heights. For the majority of calculations, the upper limit of calculation for ^{216}Po was set at 10 m and for all other radionuclides at 1000 m. The decay products concentration values are used to calculate the effective dose at reference height of 1 m above ground using dose conversion factors described earlier. Any other input parameter values not specifically listed here (turbulent diffusivity, recoil coefficient, attachment coefficient, etc.) were set at the default options listed in Schery and Wasiolek (1993).

RESULTS AND DISCUSSION

The 105 sets of measurements performed at the New Mexico Tech (NMT) site in Socorro during February of 1994 and 1995 yielded the results summarized in Table 3. The frequency distribution for thoron gas concentration is presented in Fig. 3. This is approximately log-normal with a median value of 7.5 Bq m^{-3} and a geometric standard deviation of 1.6 Bq m^{-3} . Thoron typically moves horizontally only several hundreds of meters before decay (Schery and Grumm, 1992), so its concentration can be very dependent on local soil and meteorological conditions. However, the mean (9 Bq m^{-3}) and median (7.5 Bq m^{-3}) thoron concentrations measured at the NMT site are very close to the average outdoor thoron concentration adopted in the United Nations Report (UNSCEAR, 1993) of 10 Bq m^{-3} . Furthermore, the average wind speed, atmospheric stability and aerosol particles concentration (Table 1) are not anomalous, so our results for thoron and its decay products should be fairly typical for rural areas in temperate climates.

The low equilibrium between thoron and its decay products is explained by the presence of ^{212}Pb in the thoron decay chain. Since ^{212}Pb has a much longer half-life ($T_{1/2} = 10.6 \text{ h}$) than the parent isotope thoron ($T_{1/2} = 55 \text{ s}$), ^{212}Pb can be carried to much greater heights than thoron, diluting its concentration at ground level. Due mainly to variation in this vertical mixing a diurnal variation was noted in both the total PAEC and the calculated 1-m effective dose rate. The mean values for the total PAEC and the effective dose rate for samples taken between 7:00 and 8:00 (MST) were 32 nJ m^{-3} and 20 nSv h^{-1} , respectively, and for samples taken between 13:00 and 14:00 (MST) were 9 nJ m^{-3} and 7 nSv h^{-1} , respectively. Another clear trend observed in the experimental data was a decrease in the unattached fraction of PAEC with increasing aerosol concentration (Fig. 4). This was expected and has been observed before for radon decay products, since a higher aerosol concentration increases the rate at which the unattached decay product, usually a free ion, can attach to a particle.

Since there have been few earlier measurements of thoron decay products outdoors, it is difficult to compare our results with other studies. The 23 samples taken on the roof of a building in Göttingen (Germany) by Reineking *et al.* (1992) yielded on average ^{220}Rn concentration of 3.3 Bq m^{-3} , PAEC of 6.4 nJ m^{-3} , equilibrium factor of 0.03 and a ratio of the unattached ^{212}Pb activity concentrations to total ^{212}Pb activity concentrations of 0.057. ^{212}Pb is the most important isotope contributing to the total PAEC and their lower unattached value might be explained by the lower aerosol concentration at the NMT site in comparison to the urban environment of Göttingen.

Measurements in Hong Kong (Tso and Li, 1987), where the predominant geological material is thorium (^{232}Th) rich decomposed granite, yielded averaged values of PAEC of 25 nJ m^{-3} (1.19 mWL), which is very close to the PAEC found at the NMT site.

Calculations with TPOUT were carried out to provide further interpretation of the experimental data. The

initial calculations (base case) were done with no adjustment of input parameters, and the wind speed, atmospheric stability, and aerosol concentration were fixed at the mean experimental values listed in Table 1. The experimental flux density of $2.5 \text{ Bq m}^{-2} \text{ s}^{-1}$ was used and a terrain roughness parameter of 20 cm was employed based on earlier radon measurements at this site (Schery and Wasiolek, 1993). Fig. 5 shows the resulting predictions for the vertical concentrations of thoron and its decay products. The ^{220}Rn and ^{216}Po fall off more rapidly with height in comparison with the other isotopes due to their shorter half-lives. There is more attached ^{212}Pb and ^{212}Bi than unattached since these relatively long-lived isotopes have sufficient time at these moderate particle concentrations to become predominantly attached. Such predictions are in agreement with some previous observations (Doi and Kobayashi, 1994; Butterweck *et al.*, 1994). Column four of Table 4 (base case) lists the predictions for a 1-m height for the thoron concentration, total PAEC, the unattached fraction of PAEC, the equilibrium factor, and the effective dose rate. Agreement with experimental data is better for some variables than others, with the most significant disagreement occurring with the underprediction of the equilibrium factor by about factor of four. Such disagreement is not unexpected, given the approximations involved in the model, and the lack of adequate experimental information to specify turbulent diffusivity and aerosol concentration as a function of height. For example, only the crude classification of atmospheric stability based on the standard deviation of the horizontal wind direction (EPA, 1986) at one height is used to specify the turbulent diffusivity when ideally sounding data are needed. The important trend in the experimental data of decreasing unattached fraction with increasing aerosol concentration is predicted fairly well by the model as shown in Fig. 4. For the model calculation, in Fig. 4, the input parameter of aerosol concentration (only) was varied from its base value as given in Table 1.

Comprehensive adjustment of the model to optimize parameters for best predictions of the data is beyond the scope of the presented paper. However, some variation of input parameters was carried out to study the sensitivity of the model predictions. Table 4, columns five through nine, shows some representative results. One input parameter at a time was varied (indicated by bold characters), usually by a factor of two, from the base value. The results are informative. The predicted results are relatively insensitive to variation in wind speed and terrain roughness. On the other hand, the predicted thoron concentration, total PAEC, and dose rate are quite sensitive to variation in both the thoron flux and atmospheric stability. The implication for understanding variation of thoron dose rate from one location to another is that the dose rate is controlled primarily by the atmospheric stability and the local thoron flux, and only secondarily by wind, terrain roughness, and even aerosol concentration. Activity concentrations would be expected to scale with thoron flux density from soil. High stability would be expected to result in low vertical mixing and higher ground concentrations. Wind speed, terrain roughness, and aerosol concentration primarily control deposition to the ground. Therefore, we conclude that the decay products removal caused by dry deposition is apparently less significant for controlling decay products concentrations than the dilution caused by longer-lived ^{212}Pb mixing vertically. Table 4 (improved calculations, last column) shows an example of the improved prediction of the mean experimental data that can result from variation of the input parameters from the base case. For this calculation, the parameters were varied by amounts that are consistent with their overall uncertainty. There is a significant improvement in most predictions, but, for example, the prediction of the equilibrium factor is still too low by about a factor of two. The residual differences between experimental data and predictions probably indicate the degree of approximation inherent in this one-dimensional model.

The 1-m thoron concentration at the site is very close to the world average (UNSCEAR, 1993), and the average atmospheric stability is near neutral. Given the relative insensitivity of the dose rate to the other parameters, our result of 14 nSv h^{-1} should therefore approximate a world average value for temperate climates. Assuming an outdoor occupancy factor of 0.2 (UNSCEAR, 1993), this corresponds to an average annual dose from exposure to thoron decay products outdoors of about 0.025 mSv. For comparison, UNSCEAR (1993) estimates an average annual dose from outdoor radon decay products to be 0.13 mSv. Though smaller than the dose from radon decay products, the outdoor dose from thoron decay products is still significant. For example, it is comparable to some other commonly referenced doses such as the world wide annual effective dose per caput from diagnostic nuclear medicine of 0.03 mSv (UNSCEAR, 1993).

CONCLUSIONS

1. Experimental measurements at 1-m height above the ground at an outdoor site at Socorro, NM yielded average values for thoron concentration, total PAEC, and the resulting dose rate of 9 Bq m^{-3} , 22 nJm^{-3} , and 14 nSv h^{-1} . The average value of the unattached fraction of thoron decay products potential alpha energy was 15%.
2. A diurnal pattern was observed in the calculated effective dose rate, with an average value of 19 nSv h^{-1} early in the morning, and 6 nSv h^{-1} in the afternoon.
3. The eddy-diffusion model (TPOUT) successfully represented gross features of the data, and certain experimental trends. However, the approximate nature of the model, and the lack of complete input information (such as vertical sounding data to specify atmospheric stability) led to predictions of specific variables only to within about factor of two.
4. Based on the collected experimental data, the estimated average annual dose from outdoor exposure to thoron decay products was about 0.025 mSv assuming an outdoor occupancy factor 0.2 ($14 \text{ nSv h}^{-1} \times 8760 \text{ h} \times 0.2$). The observed environmental conditions at the site in Socorro suggest that this dose estimate should also apply to other locations in temperate climates, given a similar thoron flux density from the ground of about $2.5 \text{ Bq m}^{-2} \text{ s}^{-1}$. Although smaller than current estimates of the dose from radon decay products, thoron decay products thus contribute a significant part of the total dose from natural radiation in the outdoor environment.

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REFERENCES

- Birchall, A., and A.C. James. Uncertainty analysis of the effective dose per unit exposure from radon progeny and implications for ICRP risk-weighting factors. *Radiat. Prot. Dosim.* 53(1/4): 133-140; 1994.
- Butterweck, G., A. Reineking, J. Kesten and J. Porstendorfer. The use of the natural radioactive noble gases radon and thoron as tracers for the study of turbulent exchange in the atmospheric boundary layer-case study in and above a wheat field. *Atmospheric Env.* 28(12): 1963-1969; 1994.
- Cheng, Y.S., Y.F. Su, J. Newton and H.C. Yeh. Use of a graded diffusion battery in measuring the activity size distributions of thoron progeny. *J. Aerosol Sci.* 23(4): 361-372; 1992.

Doi, M. and S. Kobayashi. Vertical distribution of outdoor radon and thoron in Japan using a new discriminative dosimeter. *Health Phys.* 67: 385-392; 1994.

Environmental Protection Agency (EPA). Guideline on air quality models. EPA-450/2-78-027R, Research Triangle Park, NC; 1986.

Grumm, D., S.D. Schery and S. Whittlestone. Two-filter continuous monitor for low levels of ^{220}Rn and ^{222}Rn . In: *Proceedings of the 1990 International Symposium on Radon and Radon Reduction Technology*, Atlanta, GA. Research Triangle Park, NC; EPA/600/9/91/026C, Vol. 3: 3-37; 1991.

P.K. Hopke, B.Jensen, C.S. Li, N. Montassier, P. Wasiolek, A. Cavallo, K. Gatsby, R. Socolow, and A.C. James. Assessment of the exposure to and dose from radon decay products in normally occupied homes. *Environ. Sci. & Technol.* (In press).

International Commission on Radiological Protection (ICRP). 1990 Recommendations of the ICRP. Oxford: Pergamon Press: ICRP Publication 60. *Ann. ICRP* 21(1/3); 1991.

International Commission on Radiological Protection (ICRP). Human respiratory tract model for radiological protection. Oxford: Pergamon Press: ICRP Publication 66. *Ann. ICRP* 24(1-3); 1994a.

International Commission on Radiological Protection (ICRP). Protection against radon-222 at home and at work. Oxford: Pergamon Press: ICRP Publication 65. *Ann. ICRP* 23(2); 1994b.

Jacobi, W. and K. Andre. The vertical distribution of radon 222, radon 220 and their decay products in the atmosphere. *J. Geophys. Res.* 68: 3799-3813; 1963.

James, A.C.. Dosimetry of inhaled radon and thoron progeny. In: *Internal Radiation Dosimetry*. O.G. Raabe, ed. Madison, WI: Medical Physics Publishers: 161-180; 1994.

Jarvis, N.S., A. Birchall, A.C. James, M.R. Bailey and M.D. Dorrian. LUDEP 1.0: Personal computer program for calculating internal doses using the new ICRP respiratory tract model. UK National Radiological Protection Board, NRPB-SR246, Chilton, UK; 1993.

Knutson, O.E. Personal computer program for the use in radon/thoron progeny measurements. Environmental Measurements Laboratory, EML-517, New York, NY; 1989.

Maher, E.F. and N.M. Laird. EM algorithm reconstruction of particle size distribution from diffusion battery data. *J. Aerosol Sci.* 16: 557-570; 1985.

National Research Council (NRC). Comparative dosimetry of radon in mines and homes. National Academy Press, Washington, DC; 1991.

Reineking, A. and J. Porstendörfer. "Unattached" fraction of short-lived Rn decay products in indoor and outdoor environments: an improved single-screen method and results. *Health Phys.* 58(6): 715-727; 1990.

Reineking, A., G. Butterweck, J. Kesten and J. Porstendörfer. Thoron gas concentration and aerosol characteristics of thoron decay products. *Radiat. Prot. Dosim.* 45 (1/4): 353-356; 1992.

Schery, S.D., D.H. Gaeddert, and M.H. Wilkening. Factors affecting exhalation of radon from a gravelly sandy loam. *J. Geophys. Res.* 89(D5): 7299-7309; 1984.

Schery, S.D. and D. Grumm. Thoron and its progeny in the atmospheric environment. In: Gaseous Pollutants: Characterization and Cycling. John Wiley&Sons, New York, NY; 1992.

Schery, S.D., R. Wang, K. Eack and S. Whittlestone. New models for radon progeny near the earth's surface. Radiat. Prot. Dos. 45 (1/4): 343-347; 1992.

Schery, S.D and P.T. Wasiolek. A two-particle size model and measurements of radon progeny near the earth's surface. J. Geophys. Res. 98 (D12): 22915-22923; 1993.

Solomon, S.B. and T. Ren. Counting efficiencies for alpha particles emitted from wire screens. Aerosol Sci. Tech. 17: 69-83; 1992.

Tso, M.W., and C. Li. Indoor and outdoor ^{222}Rn and ^{220}Rn daughters in Hong Kong. Health Phys. 53(2): 175-180; 1987.

United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). 1982 Report to the General Assembly. Ionizing Radiation: Sources and Biological Effects. United Nations, New York, 1982.

United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). 1993 Report to the General Assembly. Sources and Effects of Ionizing Radiation. United Nations, New York, 1993.

Wasiolek, P.T. and S.D. Schery. Outdoor radon exposure and doses in Socorro, New Mexico. Radiat. Prot. Dosim. 46(1): 49-54; 1993.

Wasiolek, P.T., and A.C. James. Outdoor radon dose-conversion coefficient in south-western and south-eastern United States. Radiat. Prot. Dosim. 59(4): 269-278; 1995.

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Table 1. Results of the meteorological observations during sampling at the NMT site.

Parameter	Unit	Mean	Range
Air Temperature	°C	9.1	-5.5 - 20.4
Wind Speed	m s ⁻¹	1.8	0.2 - 4.9
Pasquill Atmospheric Stability Class	-	C	A - F
Relative Humidity	%	34	13 - 80
Barometric Pressure	mm Hg	646	639 - 653
Particle Concentration	cm ³	9970	250 - 70000

Table 2. The alpha decay properties of ^{220}Rn and its major decay products.

Radionuclide	Branch (%)	Half-life	Alpha energy (MeV)	Intensity (%)
^{220}Rn		55.6 s	6.29	100
^{216}Po		0.15 s	6.78	100
^{212}Pb		10.64 h	β, γ	
^{212}Bi		60.6 m	6.05	25.2
			6.09	9.6
^{212}Po	64	2.98×10^{-7} s	8.78	100
^{208}Tl	36	3.05 m	none	
^{208}Pb		stable	none	

Table 3. Results of 105 measurements of thoron and its decay products at the NM1' site.

Parameter	Unit	Mean	Median	Range
²²⁰ Rn Concentration	Bq m ⁻³	9	7.5	2.5 - 29
PAEC	nJ m ⁻³	22	17	2 - 79
Unattached Fraction	-	0.15	0.13	0.01 - 0.46
Equilibrium Factor	-	0.03	0.03	0.01 - 0.15
Effective Dose Rate	nSv h ⁻¹	14	12	3 - 45

Table 4. Atmospheric model sensitivity study for mean experimental results.

Independent Variable	Unit	MEV	Model input values							
^{220}Rn Flux Density	$\text{Bq m}^{-2} \text{ s}^{-1}$	2.5	2.5 ^a	5	2.5	2.5	2.5	2.5	2.5	1.8 ^b
Atmospheric Stability	-	C	C ^a	C	E	C	C	C	C	E ^b
Wind Speed	m s^{-1}	1.8	1.8 ^a	1.8	1.8	3.6	1.8	1.8	1.8	1 ^b
Aerosol Particle Concentration	cm^{-3}	9970	10 ^a	10 ⁴	10 ⁴	10 ⁴	2x10 ⁴	10 ⁴	10 ⁴	3x10 ^{3b}
Terrain Roughness	cm	-	20 ^a	20	20	20	20	20	40	10 ^b
Dependent Variable	Model predicted values									
^{220}Rn Concentration	Bq m^{-3}	9	12.9 ^a	25.8	23.5	12.9	12.9	12.9	12.9	16.9 ^b
PAEC	nJ m^{-3}	22	7.9 ^a	15.8	28.3	7.6	8.0	7.7	7.7	21.2 ^b
Unattached fraction	-	0.15	0.08 ^a	0.08	0.063	0.076	0.058	0.078	0.078	0.126 ^b
Equilibrium Factor	-	0.032	0.008 ^a	0.008	0.016	0.008	0.008	0.008	0.008	0.016 ^b
Effective Dose Rate	nSv h^{-1}	14	4.5 ^a	9.0	14.9	4.2	4.1	4.3	4.3	14.3 ^b

MEV - mean experimental value

^a - base case calculation^b - improved calculation, some adjustment of input values for improved predicted values

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