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A PRELIMINARY THORON SURVEY IN THE UPPER MIDWEST

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ABSTRACT

Although thoron (^{220}Rn) was recognized as a potential source of radiation exposure in homes twenty years ago, only a few homes in the United States have been measured for thoron. Early indoor surveys suggested that thoron concentrations would generally be low, highly variable, and make a poor surrogate for thoron progeny dose. However, recent reports of significant thoron interference with some radon (^{222}Rn) detectors have renewed interest in thoron. In the upper Midwest, a region where three of the North American radon-lung cancer studies took place, basement living spaces near thoron-generating soil are common raising the possibility of thoron interference in those studies. Passive, integrating, radon-thoron discriminating detectors (ATDs) and non-discriminating detectors were developed and exposed for 90 days in 76 Minnesota homes and 19 eastern Iowa homes at interior locations where ATD radon measurements are routinely made. The overall thoron and radon distributions are lognormal with geometric mean (geometric standard deviation), and arithmetic mean of 20 (1.8), 35 Bq m^{-3} respectively and 140 (2.6), 210 Bq m^{-3}. Thoron and radon were correlated, especially for basement measurements where both isotopes had higher concentrations than on first floors. In the nondiscriminatory ATD, the median percentage of tracks attributable to thoron was 17%. This contamination was enough to cause some ATD results to be false positives for the current radon action level in some of the homes measured. The thoron interference would create a contamination of the radon-related dose estimate that averages about 10%, assuming current equilibrium ratios and dose model estimates are correct.

INTRODUCTION

During the 1980's, the discovery of unexpectedly high concentration of radon gas (^{222}Rn) in some US homes led to a rapid expansion of residential radiation measurements. In addition to ^{222}Rn, a few measurements were made for the presence of ^{220}Rn, the radon isotope commonly called thoron (Schery 1985, 1990). Thoron, like radon, can pose a health risk because it can generate progeny that can deliver dose to lung and other tissues. However, the short half-life of thoron limits its transport from sources like soil and building materials to indoor living spaces. Thus, it was generally believed that thoron concentrations indoors would be low and have large spatial variation making thoron exposures difficult to assess (Nero 1988). However, one decay product of thoron, ^{212}Pb, can accumulate significant potential alpha energy concentrations (PAEC) even at low thoron concentrations because it has a long half life. These characteristics of the thoron decay chain led early researchers to focus their surveys on ^{212}Pb rather than thoron to better quantify the potential radiation dose related to thoron. They found that thoron can generate a significant fraction of the total PAEC in homes and can be an important fraction of the dose delivered to the lungs (Schery 1985, 1990, Dudney et al. 1990, Martz et al.1990, Tu et al. 1992). However, since few measurements of thoron or its progeny have been made in homes since the early 1990s, its spatial and temporal distribution indoors is largely unknown.

Thoron can interfere with the detection of radon in detectors that do not use energy discrimination and that allow rapid gas entry into their sensitive volume. Scintillation cells, ion chambers, and alpha track registration detectors (ATDs) are susceptible to thoron interference. One type of commercially-available ATDs was found to be almost equally sensitive to thoron and radon (Pearson and Spangler 1990, Tokonami et al. 2001). However, thoron has been generally ignored both as a source of dose and as an interference in US home radon measurements.

In recent years, interest in thoron has been refreshed with the discovery of elevated thoron indoors in China and the possible effect that thoron interference may have had on some radon-lung cancer epidemiology studies. (Shang et al. 1997, Wiegand et al. 2000, Tokonami et al. 2004, Yamada et al. 2005) Many of the epidemiologic studies in North America used an ATD model that is susceptible to thoron interference (Tokonami et al. 2001, Field et al. 2006).

The primary purpose of this work is to assess the interference of thoron with the typical radon measurements made in the Upper Midwest for home radon assessment or epidemiological studies.

METHODS

These measurements used selection and exposure protocols that were similar to the three radon-lung cancer studies from the region (Field et al. 2006). They also conformed to EPA guidance for ordinary long-term home radon measurements. Detectors that were sensitive only to radon and detectors that were sensitive to both radon and thoron were exposed side-by-side in frequently occupied living spaces for 90 days.

Sampling sites and measurement protocols

To date, 76 homes in Minnesota and 19 homes in Iowa have been measured for thoron. The selection and sampling criteria were slightly different in the two states. In Minnesota, towns for the survey were selected on the basis of their surface equivalent thorium concentration (eTh). The data presented by Darneley et al. 2003, shows that the equivalent thorium surface content of the upper Midwest ranges from extremely low to roughly two thirds of the maximum observed in North America. The surface radiation measurements from the NURE program were aggregated over each Iowa and Minnesota county to estimate the geographic thoron potential of sites in the upper Midwest (Duval and Riggle, 1999). While some towns with low eTh were included, the emphasis in Minnesota was on towns with high eTh since this better reflected the eTh content in Iowa. Figure 1 shows that the sampled locations included high and low eTh contents. Inside those Minnesota regions homes were selected randomly from phone directories and enrolled via telephone. In Iowa, the high radon houses were selected because their primary study required elevated radon concentrations. All Iowa houses were in the same county and are estimated to have the same eTh.

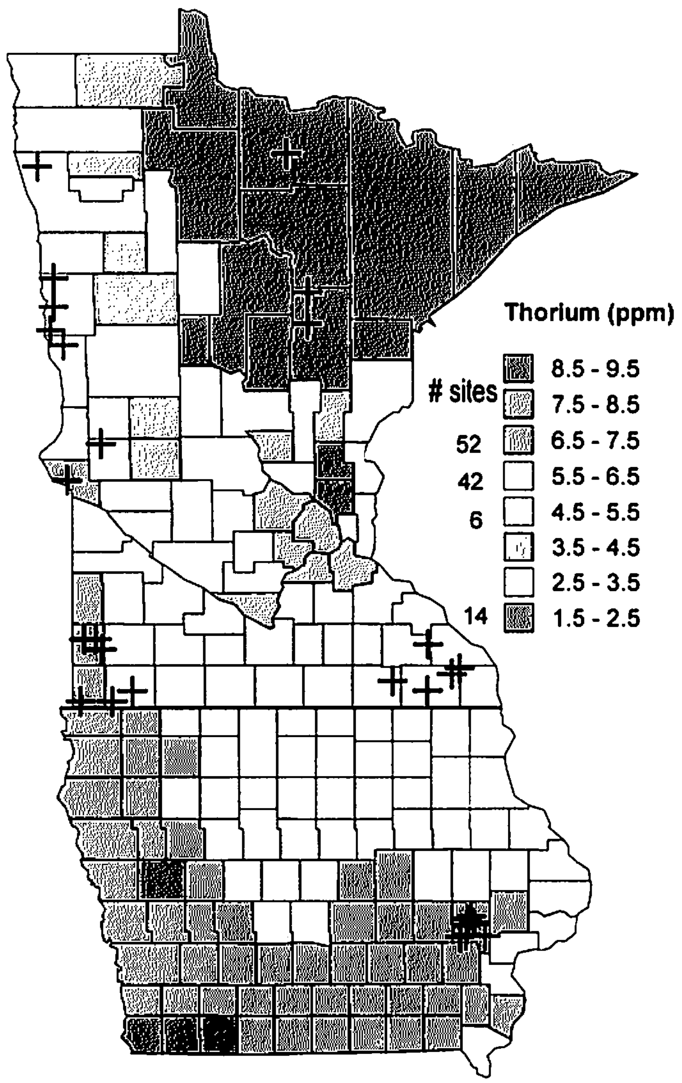

Figure 1: Map of the estimated thorium content of the surfaces of Minnesota and Iowa with the location of the selected houses shown. The number of samples in each concentration category is shown next to the legend.

In Minnesota only one measurement was made in each house while in Iowa multiple measurements were made in each house. All measurements took place in living spaces. In Minnesota, the measurement was made in the lowest floor where someone spent 10 or more hours per week. Homeowners placed the detectors following directions that were based on EPA placement protocol. In Iowa, the detectors were placed and retrieved by a trained technician following EPA protocol. All measurements were long-term, 90 days or more. The measurements usually took place in the heating season but many extended into the non-heating season.

Radon and Thoron measurement devices

Pairs of high sensitivity alpha track detectors, which had been developed for measuring atmospheric radon concentrations, were adapted to discriminate between radon and thoron (Steck et al. 1999.) In this system, one of the ATDs is enclosed in a thin polyethylene film to exclude thoron (closed detector) while the other ATD has a coarse metal grid and cloth covering the large entry port allowing for rapid gas ingress. The detector's radon and thoron response was measured through multiple exposures in two chambers in our laboratory. The radon response of the open and closed ATDs were determined in a room which contains no measurable thoron but can have constant radon concentrations of about 1 k kBq m^{-3} [25 pCi/L] . The radon concentration in the room is measured hourly with calibrated CRM (Durridge RAD-7) The radon response of the open ATD has also been calibrated in national radon tests for more than 10 years. The smaller thoron chamber has a mixture of 95% thoron and 5% radon. We confirmed the thoron calibration of the open ATDs through an intercomparison with National Institute of Radiological Sciences in Chiba, Japan. The closed detector has a response of 5.7 tracks cm^{-2} (kBq hr $m^{-3})^{-1}$to radon and less than 0.05 tracks cm^{-2} (kBq hr $m^{-3})^{-1}$ to thoron. The open detector has a thoron response of 5.2 tracks cm^{-2} (kBq hr $m^{-3})^{-1}$ and a radon response of 5.7 tracks cm^{-2} (kBq hr $m^{-3})^{-1}$.

The statistical uncertainty in the individual ATD track density is approximately 10%. Since The closed detector's track density depends on radon exposure alone while the open detector's track density reflects the radon and thoron. Hence, the thoron exposure uncertainty depends on the radon exposure as well as the thoron exposure. For example for a 90 day exposure, the thoron LLD at 37 Bq m^{-3} [1 pCi/L] was 10 Bq m^{-3} [0.2 pCi/L] while at a radon concentration of 400 Bq m^{-3} [10 pCi/L] the thoron LLD was 100 Bq m^{-3} [2 pCi/L].

RESULTS

Table 1 shows a summary of the results. Some caution must be used in interpreting and comparing results because site selection was not completely random. Thus the distributions may not be representative of the entire region. Some of the distributions are neither normal nor lognormal.

Table 1. The distribution of indoor radon (^{222}Rn) and thoron (^{220}Rn) measured in selected Minnesota and Iowa homes.

Data Set	N	^{222}Rn Mean[1]	^{222}Rn GM[2] (GSD)	^{220}Rn Mean[1]	^{220}Rn GM[2,3] (GSD)
All	114	210 [5.7]	140 (2.6)	35 [1.0]	20 (1.8)
Basement	36	320 [8.5]	210 (2.6)	55 [1.5]	21
First +	74	165 [4.4]	115 (2.4)	29 [0.8]	10
MN	76	145 [3.9]	90 (2.4)	39 [1.1]	28 (2.1)
Basement	23	230 [6.3]	140 (2.6)	75 [2.0]	27
First +	51	110 [2.9]	80 (2.2)	25 [0.7]	8
IA	38	340 [9.2]	300 (1.6)	30 [0.8]	17 (2.6)
Basement	13	460 12.4]	430 (1.4)	25 [0.7]	15 (2.5)
First +	23	290 [7.8]	260 (1.6)	35 [1.0]	17 (2.0)

[1]Units are Bq m^{-3} [pCi/L]
[2] Lognormal distribution; Geometric mean and (geometric standard deviation).
[3] When neither normal nor lognormal distributions are appropriate only the median value is given

Figure 2 shows that the overall radon and thoron distributions were close to lognormal. This set of houses has elevated radon concentrations compared to the regional average (Steck 2005, Field et al 2000).

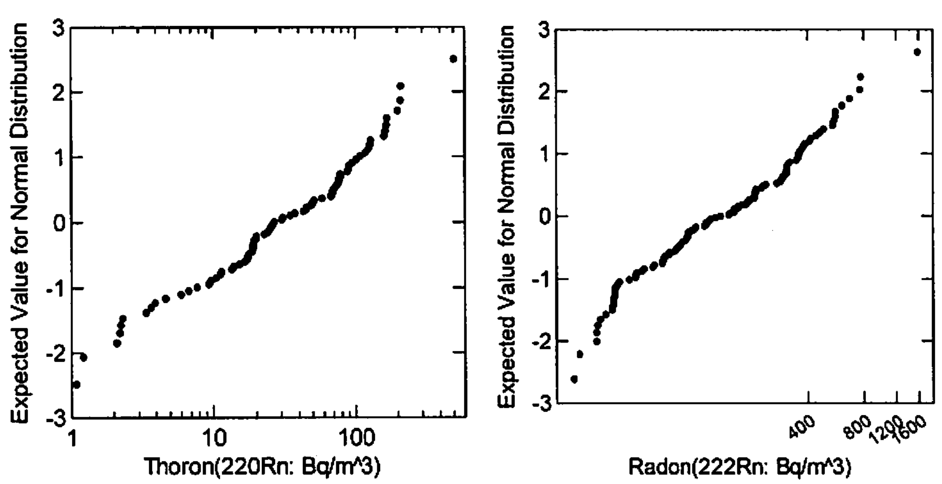

Figure 2 Probability plots of thoron and radon samples.

Since thoron concentrations were calculated from the track density difference between the open and closed ATDs, elevated radon concentrations created high LLDs for thoron concentrations. The average LLD for individual open ATDs was 79 Bq m^{-3} [2.1 pCi/L] with a range from 9 Bq m^{-3} [0.2 pCi/L] to 460 Bq m^{-3} [12 pCi/L]. Figure 3 shows the distribution of the ratio of thoron-created tracks to radon-created tracks in the open, nondiscriminatory ATDs

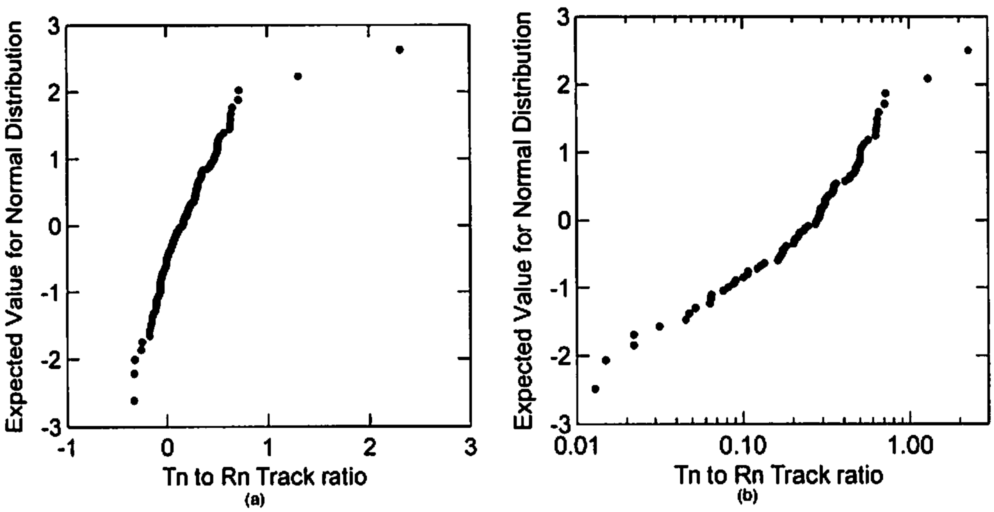

Figure 3. The distributions of the ratio of tracks caused by thoron to those caused by radon in detectors exposed side-by-side. Negative values are not plotted in (b).

Figure 4 shows that, in this sample, there is little correlation between the local eTh content of the soil and either indoor thoron or radon concentrations. Note that the samples from the low eTh region had both low thoron and radon.

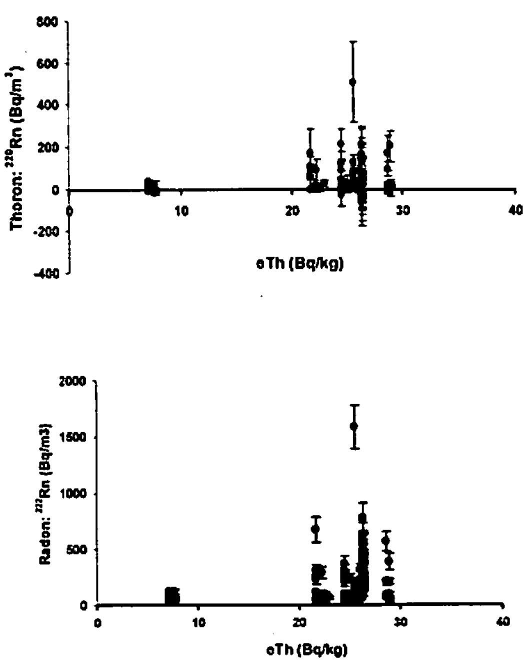

Figure 4. Thoron and radon concentration dependence on local surface soil thorium content.

Figure 5 shows the correlation between indoor radon and thoron in this region. The correlation is strongest in the Minnesota, particularly in basements.

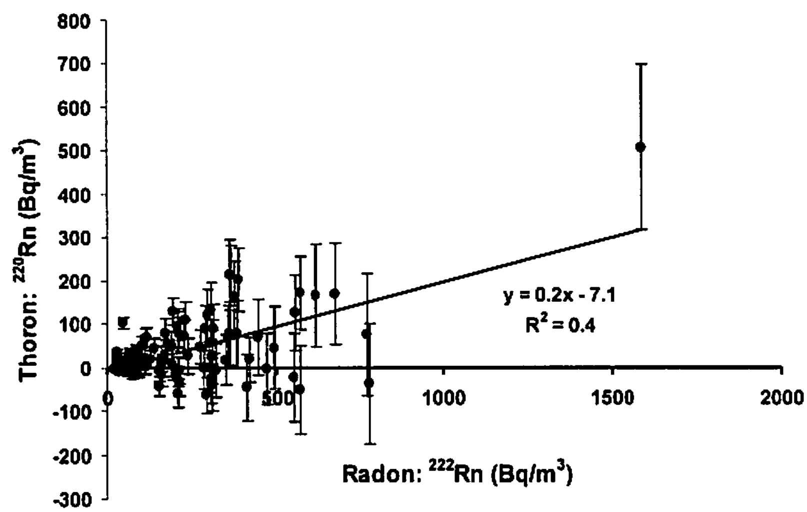

Figure 5 Thoron and radon concentrations at the indoor sites

DISCUSSION

Thoron-radon measurement interference

The primary purpose of this study was to gauge the interference that thoron may cause in typical radon measurements done for home risk assessment or as part of an epidemiological study. The magnitude of the interference will depend on the relative thoron to radon sensitivity of the technology. In the US, most long-term measurements are made with ATDs; at least one widely-used model is known to be quite sensitive to thoron.

For ATDs, the ratio of the track difference between the open ATD and the closed ATD tracks serves as a measure of interference. Using this measure, the median contribution of thoron to the open ATD track density was 17% with a range from -30% to 230%. (The thoron to radon concentration ratios would only be 5% higher since the relative sensitivity of the open detector was only 5% higher for radon.) Other ATD models or devices with different sensitivity would show a different interference statistic. For example, using the RADTRAK sensitivity reported by Tokonami (Tokonami et al. 2001), the interference ratio statistics for the sites in the present study would have a median of 13% thoron contamination with a range from -20% to 150%.

The additional signal created by thoron may have a significant impact on home measurements in regions where the median home's radon may be near the EPA action level. Five homes had radon concentrations below the action level but ATD results above the action level due to thoron contributions.

Indoor thoron concentration distributions

Thoron concentration distribution parameters (median, variation) give a general measure of indoor thoron in this region. Thoron concentrations were generally higher in the basement, particularly at the Minnesota sites. The thoron concentrations at individual sites suffered from substantial uncertainties when the radon concentration was high and the thoron concentration was low. Hence accurate individual site thoron concentrations are only available when thoron concentrations were about the same or higher than the radon concentration. Thoron concentrations exceeding the LLD were observed at 20% of the sites, 11% in Iowa and 23% in Minnesota. Sixty percent of those sites were first floors. The maximum thoron concentration observed was 510 Bq m^{-3} [13.7 pCi/L] in a basement recreation room.

Few indoor thoron measurements have been reported in the literature; most research has focused on thoron progeny measurements. Schery, who reported thoron measurements at 25 indoor sites in 5 states, observed lower and more diverse concentrations (Schery 1985). The sites were mainly in southern and western United States where grab sample measurements were made, presumably during the day. The geometric mean and geometric standard deviation was 6 Bq m^{-3} and 4.2 respectively for that survey compared to 20 Bq m^{-3} and 1.8 for this upper Midwest survey.

Following the discovery of some very high indoor thoron regions in China (Shang et al. 1997, Wiegand et al. 2000, Yamada et al. 2006), new reports are beginning to appear in the literature that give a picture of indoor thoron in a variety of climates and housing construction (Kim, Shang , and Martinez). The ranges of concentrations in these reports overlap with those found in the upper Midwest.

Thoron correlations

The estimated equivalent thorium soil content (eTh) does not appear to be a useful guide to elevated indoor thoron concentrations since the regression coefficient was low (R^2~0.02). The highest correlation (R^2 ~0.1) was found in Minnesota basement sites.

There have been few reports about correlations between indoor radon and thoron. Schery reports a good correlation between the two gas concentrations; R^2~0.5 (Schery 1985, 1990 Li and Schery 1992) which is comparable to the correlation found in this study;

R^2~0.4. The radon-thoron correlation is best in Minnesota basements; R^2~0.8. However, poor correlation was observed between radon and thoron in houses with soil walls in Gansu Province China (Tokonami et al. 2004, Shang et al. 2005, Yamada et al. 2006). In those houses, the radon concentration was generally lower and the thoron concentration was higher than in the upper Midwest houses. The difference may reflect differences in building materials and radon/thoron entry mechanisms in the two regions.

Simultaneous, time integrated measurements of both thoron and its progeny are difficult. No surveys of correlations between the two have been reported in the US. A report of recent work suggests that a central estimate of the equilibrium ratio between thoron and its progeny in US homes may be 0.02 (Harley and Chittaporn 2006). This value is similar to one reported for homes in China (Tokonami et al. 2004, Yamada et al. 2006) but much less than the value given in UNSCEAR 2000. Since the correlation between thoron and its progeny at individual sites is not good, the ratio is only useful for making estimates for a "typical" house.

Implications for epidemiological studies

Epidemiological studies with small sample size depend on good correlation between the surrogate that they measure and the disease risk factor. In the case of radon and lung cancer, one would like to have a good surrogate for the energy delivered to the lungs of each individual over an extended period in the past. Other factors that interfere with the surrogate measure - dose correlation tend to obscure the trend between surrogate and disease (Steck and Field 2006).

Thoron could interfere in several ways. If a thoron insensitive device is used to measure the radon, then the dose from thoron, which presumably carries some risk of causing the disease, is not included in the surrogate measurement. As an example, the retrospective radon reconstruction technique which uses implanted ^{210}Po as a surrogate is insensitive to thoron progeny.

Most epidemiological studies have used ATD tracks, interpreted as radon exposure, as the dose surrogate. How much of a problem might thoron be in this case? For illustration, let's assume that the current estimate for the thoron equilibrium factor (~0.03) is correct, the microdosimetry models for both radon and thoron progeny are reasonably good, and that the ATDs used for measurements are roughly equally sensitive to both thoron and radon. Then, for the radon and thoron concentrations in the upper Midwest survey, the thoron contamination would represent an average 10% error in the radon-related dose estimate based on a nondiscriminatory ATD. But in regions or houses where either thoron concentrations are higher or radon concentrations are lower, thoron may be more of a problem. For example, some North American epidemiological studies have been done in regions where the radon concentrations are low (Field et al. 2006). If the thoron levels observed in the present study region, where the typical radon concentration is ~100 Bq m^3, were present in those other epidemiological study regions, where radon concentrations are a factor of 2 to 5 lower, then the possibility for serious thoron interference with the studies' analysis is possible.

CONCLUSIONS

Substantial concentrations of thoron were found in upper Midwest living spaces. Thoron generated tracks added 17% on average to the tracks from radon in the ATD used in this study. Thoron interference has the potential to cause significant problems for some sensitive radon assessment applications. However, the magnitude and distribution of time-averaged thoron and thoron progeny indoors needs to be studied in a wider sample of houses.

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