RADON, THORON AND THEIR PROGENY IN LANCASTER PA HOMES

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Two radon isotopes are commonly found in US homes. Conventional wisdom holds that ^{220}Rn (thoron) concentrations are lower than ^{222}Rn (radon) concentrations and pose a smaller health risk in most living spaces. An earlier survey of radon and thoron gas in 30 Lancaster, PA homes found some with elevated concentrations of both isotopes. That survey found some living spaces with higher short-term concentrations of thoron than radon. To improve the estimate of the long-term risk from exposure to these gases, we took both short and long-term measurements in 20 homes. In addition to grab samples of radon and thoron progeny in two different seasons, we measured the long-term gas concentrations and surface deposited activity to estimate contributions to the potential airborne dose from radon and thoron progeny. The following averages were found: long-term average radon concentration 395 Bq m^{-3} (10.7 pCi/L) with an estimated radon progeny equilibrium ratio 0.24; long-term thoron concentration 120 Bq m^{-3} (3.2 pCi/L) and progeny equilibrium ratio 0.02. Estimated average effective doses calculated from long-term gas measurements and short-term progeny measurements are: radon progeny 7.3 mSv yr^{-1} and thoron progeny 0.6 mSv yr^{-1}.

INTRODUCTION

Residential radiation measurements in the US taken during the 1980's occasionally included both ^{222}Rn and ^{220}Rn (Schery 1985, 1990). To distinguish these two isotopes we will follow the common practice of using the nicknames radon for the former and thoron for the latter. Thoron, like radon, can pose a health risk because its progeny can deliver radiation 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).

Significant potential alpha energy concentrations (PAEC) can accumulate indoors even at low thoron concentrations because one of its decay products, ^{212}Pb, has a long half life. Early surveys concentrated on thoron progeny measurements rather than thoron measurements to better quantify the potential environmental radiation doses. They found that thoron can generate a significant fraction of the total PAEC in homes and can be an

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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.

Interest in thoron has been revived with the discovery of elevated thoron indoors in China. An earlier study of the radon and thoron in Lancaster suggested that there might be high thoron homes there (Stewart 2007). These cases raise the question of the effects of thoron on the potential dose to individuals and on “radon” measurement technologies used in risk assessment. (Shang et al. 1997, Wiegand et al. 2000, Tokonami et al. 2004, Yamada et al. 2005, Steck 2006).

While the dose is primarily delivered by the progeny for both radon isotopes, measurements of radon gas are most common in residential risk assessment since they are easier to make, especially over extended periods of time. While gas measurements in homes typically average over days to months, typical progeny measurements are grab samples that collect the radionuclides over brief periods of time (5 to 10 minutes). This practice introduces substantial uncertainty in exposure assessment because both gas and progeny have strong temporal variation on diurnal to seasonal scales. Integrating passive progeny measurement technologies show promise for improving radiation risk assessment in homes (Steck et al. 2007).

The primary goals of this study were to measure the long term average concentrations of radon and thoron gas, to take grab samples of their progeny in two seasons in order to estimate the doses available in select Lancaster PA homes, and to test the field performance of passive progeny dosimeters.

METHODS

To estimate the long-term airborne radon and thoron progeny in the living spaces, we placed track-etch gas and progeny detectors for 90 day exposures. Grab samples were taken of the gas and airborne progeny upon placement of these detectors, and then again upon retrieval. Estimates of the long-term equilibrium ratios (F_R, F_T) were based on the averages of measurements from these grab samples. Finally, long-term radon progeny PAEC and dose were estimated from the long-term gas concentrations and the average grab sampled F’s.

Twenty homes, located in Lancaster County, PA, were measured for radon, thoron and their progenies. The locations were selected from a prior study (Stewart 2007) to ensure measurable results. The sites in the houses were selected based on the lowest lived-in level. An equal number of basements and first floors were sampled. The initial grab samples were taken and the long-term tests were placed between 8/18/2007 and 11/26/2007. The final grab samples were taken and the long term tests were collected between 12/19/2007 and 2/25/2008.

Two types of grab samples were collected for this study, radon and thoron progeny, and radon and thoron gas. The progeny grab samples were measured using the Ludlum Measurements Inc. Model 2000 counter and Model 43-9 alpha scintillation detector, and data were collected to computer storage using DEPOMON interfacing software. The progeny grab sampling equipment was calibrated in the laboratory with a SARAD EQF whose calibration is traceable to PTB. After taking a background count and checking the calibration using a ^{230}Th sealed source, air was pumped through a glass fiber filter for 5 minutes for radon progeny and 10 minutes for thoron progeny respectively. Then counts from the filter were taken every minute for 50 minutes or 325 minutes, for radon progeny and thoron progeny respectively. The progeny activities were calculated using the Tsivoglou method for radon progeny and the Khan-Busigin-Phillips for thoron progeny (Khan et al. 1982)

The gas grab samples were taken using the Durridge Co. Inc. RAD7. After lowering the internal relative humidity to 9%, three consecutive 20-minute tests were conducted and the results averaged.

Long-term average concentrations were measured with track-etch detectors. Radon and thoron gas detectors, conventional and encapsulated ATD's, were placed more than one meter from the external walls, and usually about 1.6 meters from the floor, but not less than one meter. The surface alpha activity of radon and thoron progeny (specifically ^{214}Po and ^{212}Po respectively) were measured using open track registration detectors fitted with several energy absorbing filters. In all but two cases, the surface deposition detectors were placed on a window pane, as close to the center as achievable. In the exceptions, they were placed on a structural steel support beam and a plastic wall fixture. These gas detectors (Steck 2006) and surface progeny detectors have been described elsewhere (Steck 2007 et al.).

RESULTS

Almost all of the distributions were more lognormal than normal, so geometric mean, geometric standard deviation and average statistics are given for the major results. The grab sample distribution statistics for the progeny measurements are given in Table 1. The spatial and temporal variations are shown in Figure 1 and 2.

Table 1: Progeny Grab Sample Measurement Statistics in Lowest Lived-in Sites

	Median(GSD) mWL	Average mWL	Equilibrium ratio Median[GSD]
Radon progeny (All 20 sites)	38 [2.1]	48	0.22 [1.4]
Thoron progeny (All 20 sites)	3.8 [1.9]	4.7	0.018 [3.4]

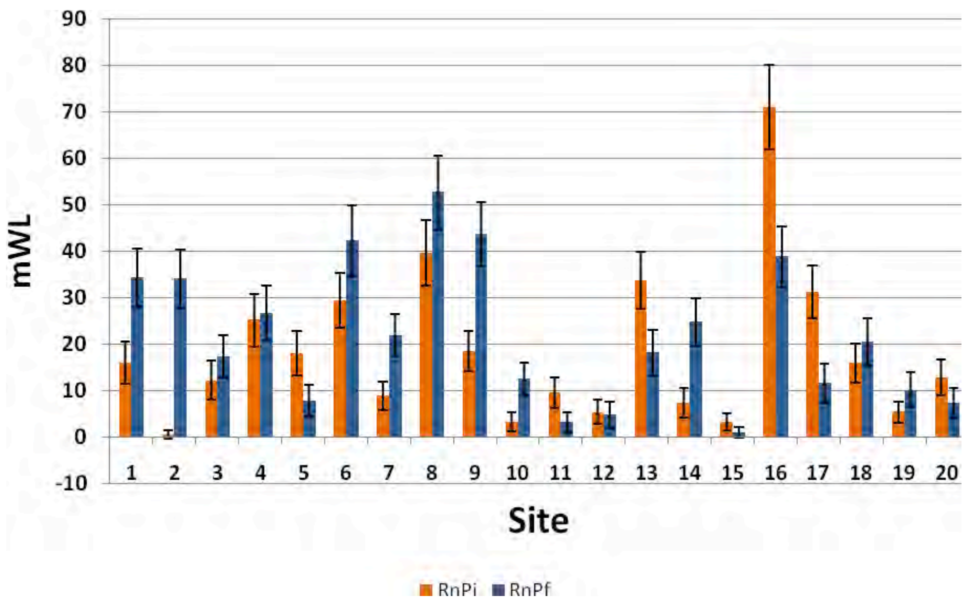


Figure 1: Radon progeny PAEC from the initial and final grab samples

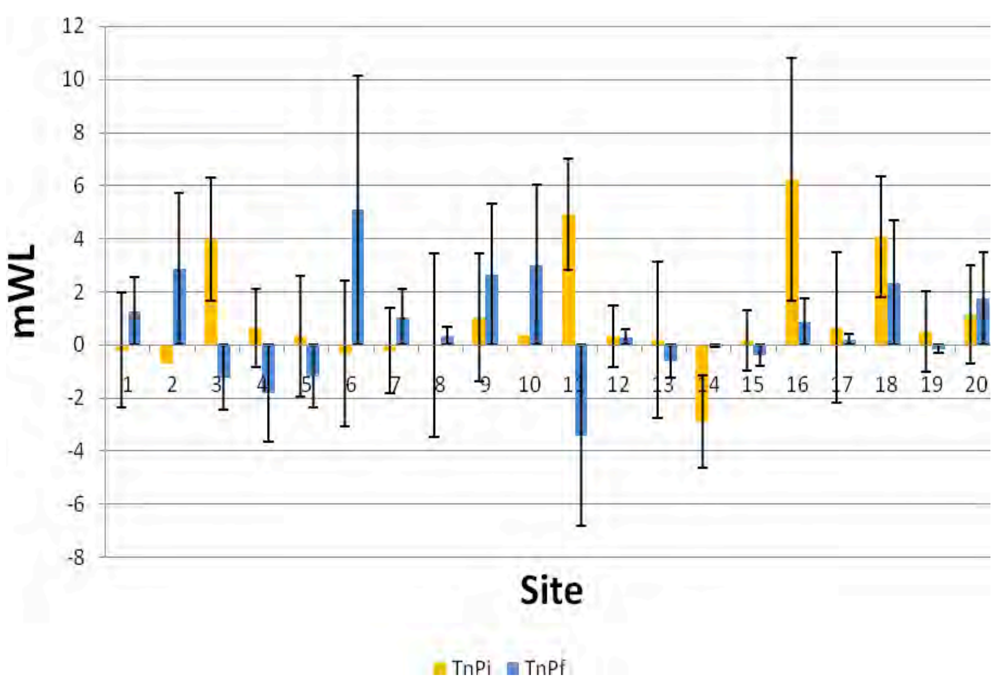


Figure 2: Thoron progeny PAEC from the initial and final grab samples

The results from the long-term track registration detectors are shown in Tables 2 and 3.

Table 2: Long-term gas measurement statistics

	Number	Median [GSD] Bq m^{-3}	Average Bq m^{-3} (pCi/L)
Radon (All sites)	20	325 [1.9]	395(10.7)
1st Floors	10		289 (7.8)
Basements	10		501(13.5)
Thoron (All sites)	20	108 [3.5]	121(3.3)
1st Floors	10		56 (1.5)
Basements	10		185 (5.0)

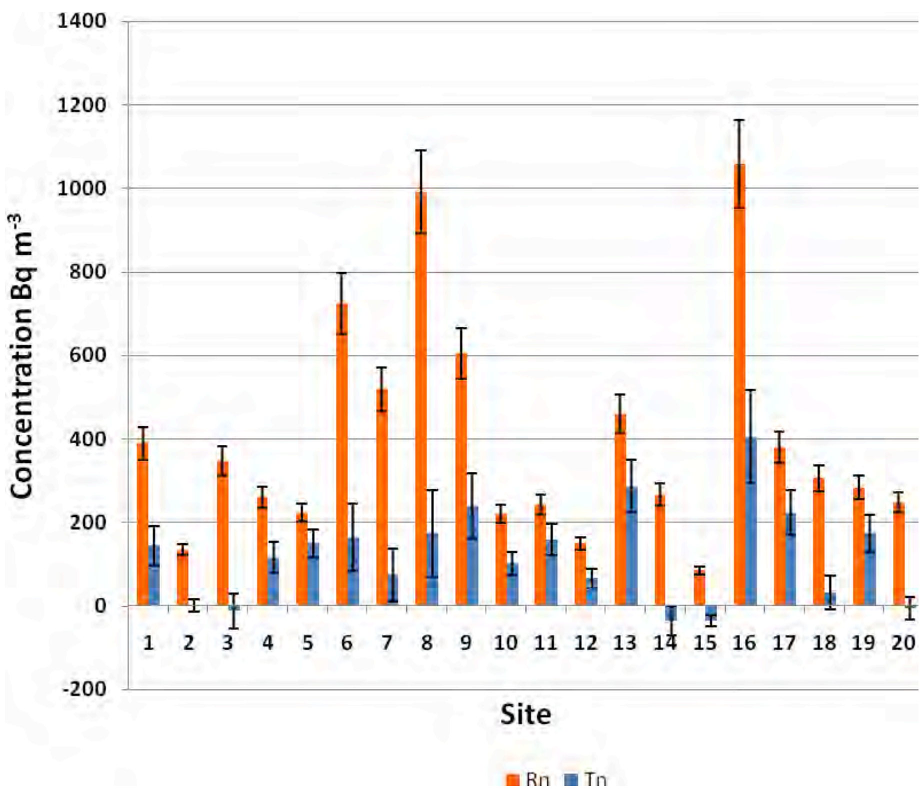


Figure 3: Site-to-site-variation of the radon and thoron gas concentrations

A statistical summary of the surface activities of ^{214}Po and ^{212}Po are given in Table 3.

Table 3: Long-term surface activity measurement statistics

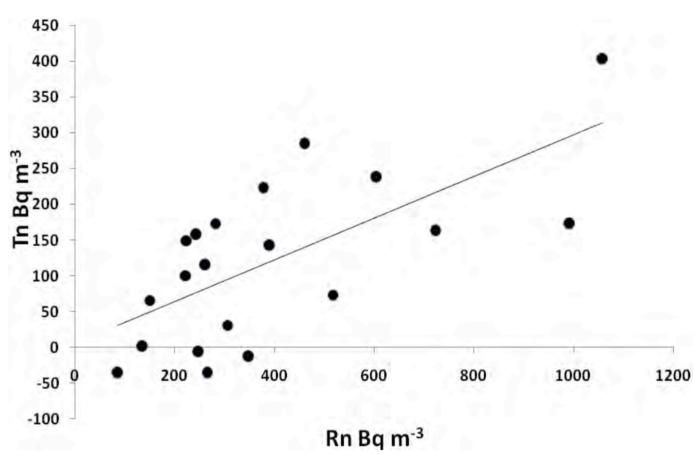
	Number	Median [GSD] Bq m^{-2}	Average Bq m^{-2}
Radon progeny(^{214}Po)	20	28 [2.0]	37
Thoron progeny (^{212}Po)	20	2.6 [2.3]	3.5

DISCUSSION

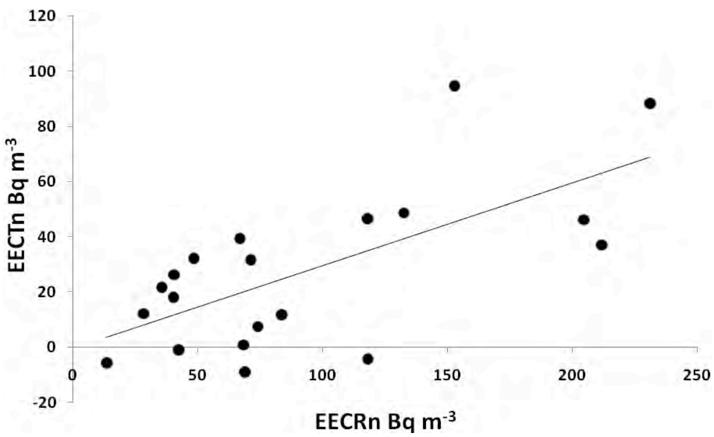
Although measurement of radon gas in homes is common today, simultaneous measurements of radon and its progeny are rare. Those kinds of measurements were made occasionally during the early research period of the 80's and 90's (Nero 1988). One reason for the rarity is the cost and difficulty of a progeny sampling protocol that would average over the temporal variation to give a representative measure of the available dose. The present work, taking place in select homes in a single locality, should contribute to the data on the relationship between radon and radon progeny, but no claims of representativeness are made for other houses. The radon is considerably elevated in these homes compared to national averages. The average equilibrium ratio (F_R), that is the fraction of the airborne Potential Alpha Energy Concentration (PAEC) compared to the maximum possible, is lower than the "rule-of-thumb" value; 0.24 vs. 0.4. Nevertheless, when one calculates the dose rate available from radon progeny using the UNSCEAR approach of Equilibrium Equivalent Concentration (EEC), the effective dose rates average to 7.3 mSv/yr and vary from 1 to 18 mSv/yr (UNSCEAR 2000).

Thoron measurements are rare in the US. Few studies report simultaneous radon, radon progeny, thoron and thoron progeny measurements. Schery's work in the mid 80's shows a reasonable correlation between radon and thoron gas concentrations; R^2~0.5 (Schery 1985, 1990 Li and Schery 1992). Our results (shown in Figure 4(a)) show general agreement with Schery's results as well as a recent study in the Upper Midwest (Steck 2006). Poorer correlations were found in Gansu Province between the two isotopes in homes where thoron dominated due to building materials (Tokonami et al. 2004, Shang et al. 2005, Yamada et al. 2006).

Correlations between thoron, radon and their progenies are not well documented. Figure 4(a) shows the relationship between the gas concentrations alone while Figure 4(b) shows correlations between the EEC values which include gas and progeny. The R^2 in both cases is approximately 0.5.



(a)

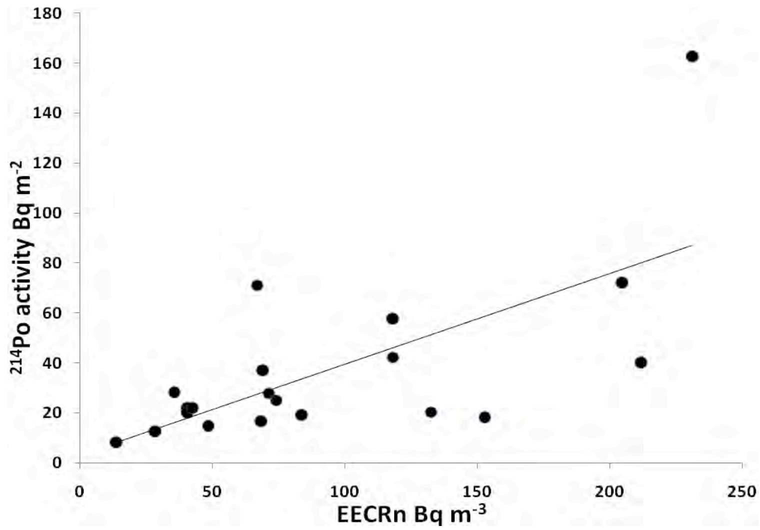


(b)

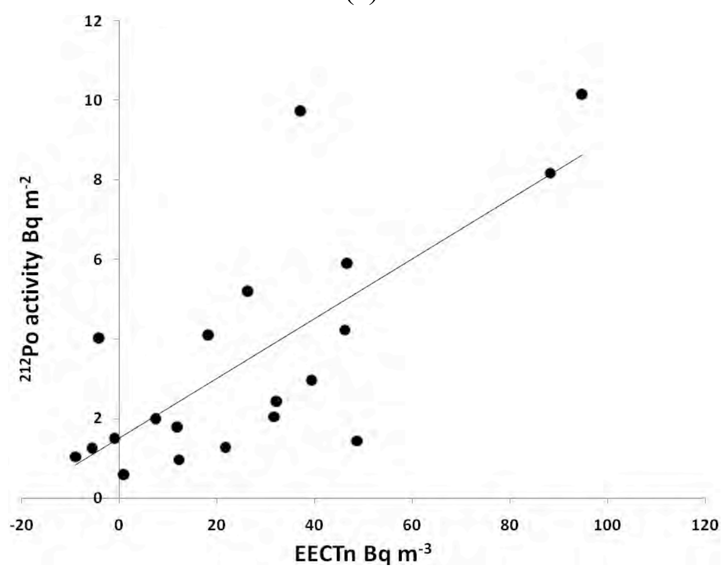
Figure 4 Correlation between (a) radon and thoron gas concentrations and (b) equilibrium equivalent radon and thoron gas concentrations.

Simultaneous measurements of both thoron and its progeny are difficult. Even in locations where thoron gas is higher than normal, thoron progeny concentrations are low. Grab samples taken with modest air flow rates and reasonable times yield progeny concentrations with substantial uncertainties as Figure 2 illustrates. No surveys of correlations between thoron and its progeny in the US have been published. An abstract 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. We found a median equilibrium ratio for thoron progeny of 0.018 but a variation of a factor of 3 around that median. Some of the variation undoubtedly arises from measurement variation and counting statistics. Nonetheless using the UNSCEAR EEC approach, we estimate that the available effective dose rate from thoron progeny to average 0.6 mSv/yr in this sample of houses.

Passive progeny dosimeters based on surface deposition hold the promise to improve risk assessment through integrating the measurement of the progeny. Passive detectors can be left in place for months tracking the average activity of surface-deposited progeny in an efficient, effective manner. New technologies have shown reasonably good results in laboratory and limited field tests (Steck et al. 2007). We deployed surface alpha track-etch detectors in this study. Figure 5 shows that the correlation between the estimated airborne progeny (EEC) and the average surface deposited activity for both the radon and thoron is fairly good (R^2 ~0.5). Part of the variability is likely due to the use of grab samples for the F values and partly due to the variation in surface deposition conditions which were not taken into account in this analysis.



(a)



(b)

Figure 5 Correlation between estimated airborne progeny and gas concentration for (a) radon and (b) thoron.

CONCLUSIONS

Elevated radon, thoron and their progenies were found in most of these homes. There was reasonable correlation between radon and thoron gas concentrations as well as the corresponding fractions of airborne progeny. Long-term measurements of activities of surface-deposited progeny show promise for estimating average airborne radon and thoron progeny dose rates in residential settings.

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