THE SIGNIFICANCE OF THORON (^{220}Rn) CONTRIBUTION TO NATURAL RADIATION DOSE IN MALAYSIA

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ABSTRACT

The objective of this study was to assess the significance of thoron contribution to the natural radiation dose in the environment. The indoor and outdoor radon/thoron (gas and progenies) concentrations in a few areas in Peninsular Malaysia, Sarawak and Sabah were measured based on active method using continuous radon/thoron gas and progeny monitors. Their respective concentrations and doses were compared. The mean indoor and outdoor radon gas concentrations were 23 Bq m^{-3} and 17 Bq m^{-3} respectively while the mean indoor and outdoor thoron gas concentrations were 10 Bq m^{-3} and 9 Bq m^{-3} respectively. The mean indoor and outdoor EEC (Rn) were 11.1 Bq m^{-3} and 3.2 Bq m^{-3} respectively while the mean indoor and outdoor EEC (Tn) were 3.0 Bq m^{-3} and 1.1 Bq m^{-3} respectively. Total annual effective dose (indoor and outdoor) from radon and thoron series were 0.9 mSv and 1.1 mSv respectively. Thoron contribution to the total annual effective dose was about 50%, indicating that radon and thoron are equally important in the assessment of natural radiation dose. Thus, the presence of thoron in the Malaysian environment cannot be simply ignored otherwise it will underestimate the dose receive by the people.

ABSTRAK

Objektif kajian ini adalah untuk menilai kepentingan sumbangan thoron kepada dos sinaran semula jadi di alam sekitar. Kepekatan radon / thoron (gas dan sepsis susulan) dalaman dan luaran di beberapa kawasan di Semenanjung Malaysia, Sarawak dan Sabah diukur berdasarkan kaedah aktif menggunakan radon / thoron berterusan dan monitor progeny. Kepekatan dan dos masing-masing telah dibandingkan. Mean konsentrasi gas radon dalaman dan luaran adalah 23 Bq m^{-3} dan 17 Bq m^{-3} masing-masing manakala kepekatan gas thoron dalaman dan luaran adalah 10 Bq m^{-3} dan 9 Bq m^{-3} masing-masing. EEC (Rn) dalaman dan luaran adalah 11.1 Bq m^{-3} dan 3.2 Bq m^{-3} masing-masing manakala EEC (Tn) dalaman dan luaran adalah 3.0 Bq m^{-3} dan 1.1 Bq m^{-3} masing-masing. Jumlah dos berkesan tahunan (dalaman dan luaran) daripada siri radon dan thoron adalah 0.9 mSv dan 1.1 mSv masing-masing. Sumbangan Thoron kepada jumlah dos tahunan tahunan adalah kira-kira 50%, menunjukkan bahawa radon dan thoron adalah sama penting dalam penilaian dos sinaran semula jadi. Oleh itu, kehadiran thoron dalam persekitaran Malaysia tidak boleh diabaikan sebaliknya ia akan meremehkan dos yang diterima oleh orang awam.

Keywords: radon, thoron, indoor, outdoor, dose

INTRODUCTION

Radon (Rn) and thoron (Rn) are radionuclides from U and Th decay series respectively. Radon (Rn) and its short-lived decay products or progenies have received much attentions and interest from investigators around the world as it is believed that they can cause significant radiation dose to the lungs and other tissue. On the other hand, thoron (Tn) was not given much attention due its short-lived (half-life 55.6 s) which limits its transport from sources to indoor living spaces. The presence of thoron was often neglected because it was considered that the quantity of thoron in the environment was very much less compared to radon and so it's associated dose. It was also assumed that thoron will completely decay before giving harm to the people. However, in certain situations, such as when building materials containing high ^{232}Th (^{228}Ra) were used, thoron may become a significant source of exposure. Thoron, like radon, can also pose a health risk because it can generate decay products that can deliver dose to lung and other tissues.

In recent years, the awareness of importance of thoron in terms of its radiological impact and contribution to radiation doses has been acknowledged. Several investigations concerning thoron have been performed by researchers in a few countries worldwide (Chen *et al,* 2011; Hosoda *et al,* 2017; Kudo *et al,* 2015; McLaughlin *et al,* 2011; Omori *et al,* 2016; Ramachandran, 2010; Saidou *et al,* 2015; Shang *et al,* 2005; Sorimachi *et al,* 2009, 2012; Sulaiman *et al,* 1994; Tokonami *et al,* 2001, 2004). Several studies found that thoron can be a significant contributor to the radiation dose in residential buildings (Ramola *et al,* 2012; Janik *et al,* 2013; Kudo *et al,* 2015). Enhanced thoron concentrations have been reported in Japanese traditional mud houses (Doi, 1992) and Chinese cave dwellings (Tokonami *et al,* 2004).

In Malaysia, not many studies on thoron and its decay products have been conducted and data are scarcely available. Earlier assessment by Sulaiman *et al* (1994) using gross alpha and delay counting method, showed that thoron present in the environment along with radon and its concentration was quite significant. The objective of this study was to further assess the contribution of thoron to natural radiation dose in Malaysia environment. In this work, radon and thoron (gas or progenies) were measured in a few areas in Peninsular Malaysia, Sarawak and Sabah using instruments equipped with alpha spectroscopy system. This paper reports results of the measurements. Data analysis and discussions were emphasized on thoron contribution and the dose received by the people.

MATERIALS AND METHODS

Study area

The measurement locations are shown in Figure 1. The indoor radon/thoron gas and progenies measurements were carried out in brick houses or hotels. Measurements in dwellings were conducted mostly in living room. The detectors were placed on a table or cupboard. The outdoor measurements were performed either on ground surface or at 1 m height (by using a tripod stand). As far as possible, the measurements were conducted for a period of about 24 hours in order to obtain a representative average radon and thoron concentration for one day.

Radon/thoron gas and progenies were measured based on active method using detectors model RTM1688-2 and DosemanPro respectively. Both equipments were manufactured and calibrated by Sarad (Germany). Both detectors consist of the same common features such as membrane pump, USB interface, rechargeable battery, semiconductor detector and alpha spectroscopy system (which can discriminate alpha energy peaks emitted by radon and thoron progenies). The difference between those two detectors is the RTM1688-2 has a larger detector chamber for sampling of radon and thoron gas while DosemanPro came with a filter holder for collection of radon and thoron progenies.

The air containing radon and thoron gas will be drawn by the pump and transferred into detector chamber (RTM1688-2). Radon and thoron gas decayed to progenies by emitting alpha particles which will be subsequently counted by the semiconductor detector and analysed using Radon Vision software. The radon and thoron gas concentrations were reported in Bq m^{-3}. While, the sampling of radon and thoron progenies was performed by pumping the air passes through the filter paper. The deposited radon and thoron progenies on the filter will automatically be counted by the semiconductor detector (DosemanPro) and also analysed using Radon Vision software. The radon and thoron progenies concentrations were reported in term of equilibrium equivalent concentration (EEC) in Bq m^{-3}.

Figure 1: Radon and thoron measurement locations

RESULTS AND DISCUSSION

Radon and thoron gas

Table 1 presents the results of radon and thoron gas measurements. The mean radon gas concentrations were 23 ± 10 Bq m^{-3} and 10 ± 5 Bq m^{-3} for indoor and outdoor respectively while for thoron gas the mean concentrations were17 ± 4 Bq m^{-3} and 9 ± 4 Bq m^{-3} for indoor and outdoor respectively. The mean radon gas concentrations indoor was lower than global average of 39 Bq m^{-3} while for outdoor the mean concentrations was similar (UNSCEAR, 2000). The mean thoron gas concentrations indoor was higher than global average while for outdoor the mean concentrations was comparable. The global average of thoron for both indoor and outdoor was 10 Bq m^{-3} (UNSCEAR, 2000). The results indicated that thoron gas was present and measurable in Malaysian environment even though the detector was placed 1 m above the ground or floor. Ratios of thoron to radon gas were in the range of 0.30-0.67 and 0.30-0.86 for indoor and outdoor air respectively. The results also showed that thoron gas concentrations were normally lower than radon gas.

Table 1: Radon (Rn) and thoron (Tn) gas concentrations indoor and outdoor.

Area	Rn Gas, Bq m^{-3}	Tn Gas, Bq m^{-3}	Ratio Tn/Rn	Note (description of measurement)
Indoor				
Bangi	23	7	0.30	1 m, carpeting
Kajang	33	16	0.48	1 m, ceramic tile
Kuantan	12	8	0.67	1 m, ceramic tile
Mean ± a	23 ± 11	10± 5	0.30-0.67	
Outdoor				
Bangi	20	6	0.30	1 m, open ground
Kajang	14	12	0.86	1 m, car porch- ceramic tile
Mean ± a	17 ±4	9 ± 4	0.30-0.86	

There were situations where thoron concentrations were comparable or higher than radon. This situation was normally observed at night and early morning when the atmosphere was stable. High thoron gas was also found when measurement performed close to the source (e.g., on the ground surface, on the floor or near the wall). High thoron gas concentrations have been reported in Japan, Korea, China, Canada and Hungary with ratio of thoron to radon gas concentrations ranged from 0.97-35.3 (Tokonami, 2010) indicating that in certain areas in those countries thoron gas concentrations were much higher than radon gas concentrations. Saidou *et al.,* (2015) reported that high thoron concentrations up to 300 Bq m^{-3} have been measured in houses in southwestern Cameroon. A mean thoron gas concentration of about 400 Bq m^{-3} in Nagoya (Japan) was reported by Guo *et al* (1992).

High thoron gas concentrations were related to high Th (Ra) concentrations in soil or building material. As Malaysian soil and building materials contain slightly higher ^{228}Ra (Omar, 2000), significantly high thoron in soil gas concentrations was expected. However, less thoron concentrations were measured in air due to its short half-life (55.6 s) where its concentrations reduced sharply with distance from the source (ground/floor/wall) compared to radon (half-life 3.8 d). If there was no continuous supply of thoron gas, its concentrations will become negligible after 5 min exhaled out of the ground/wall/floor surfaces. This will result in the spatial distribution of thoron gas concentration is highly inhomogeneous in a room and make it difficult to assess thoron progenies concentrations based on thoron gas measurements or vice versa. Large spatial variations of thoron gas indoors have been noted by Tokonami (2010) and Hosoda *et al* (2017).

As thoron present in indoor air is produced primarily as a result of emanation from building materials, the highest concentrations are likely to be found close to the walls and floors. Thoron emanation from building materials might also vary within different rooms of a house because of differences in the building material properties. Thoron (and radon) levels in closed environment are also affected by the degree of exchange with outdoor air. While in the open air, thoron (and radon) levels are influenced by the balance between the exhalation rate from its source and dilution processes in the atmosphere. Wind will carry thoron gas (and radon gas) away from its exhalation point while rainwater will reduce or prevent thoron (and radon) exhalation from the ground. Low or minimum concentrations were measured in the afternoon while high or maximum concentrations were measured during nighttime or early morning.

Radon and thoron progenies

Table 2 showed the results of measurements of radon and thoron progenies in term of equivalent equilibrium concentration (EEC). Most of the measurement results showed that EEC thoron were generally lower than radon with exception a few locations showing the opposite. Ratio EEC thoron to radon ranging from 0.09-1.60 and 0.02-0.80 for indoor and outdoor respectively. Even in Sabah and Sarawak, which have been reported to have low soil radioactivity (Sulaiman *et al,* 2015), EEC thoron and radon also low but their ratios were quite significant (range 0.2-0.9). The ratios were high especially on the ground surface measurement. About 10% of the indoor measurements showed higher average thoron progenies compared to radon progenies. UNSCEAR (2000) reported that ratio thoron to radon progenies indoor ranging from 0.01-0.50 while for outdoor the ratio ranging from 0.01-0.08. Ramachandran (2010) reported ratios of thoron to radon progenies in a few countries ranging from 0.1-1.3. High ratio of thoron to radon progenies were also reported by Guo *et al.,* (1992) and Doi *et al.,* (1994).

Table 2: EEC radon (Rn) and thoron (Tn) indoor and outdoor.

Area	EEC Rn, Bq m^{-3}	EEC Tn, Bq m^{-3}	Ratio EEC Tn/Rn	Note (description of measurement)
Indoor				
Bangi	15.9	5.3	0.33	1 m, carpeting
Kajang	8.6	2.9	0.34	1 m, ceramic tile
Kuantan	17.9	5.8	0.32	1 m, carpeting
Kemaman	1.6	2.5	1.60	1 m, carpeting
Cameron Highlands	19.0	2.1	0.11	1 m, ceramic tile
Ipoh	10.7	2.4	0.22	1 m, carpeting
Sungkai	18.4	1.6	0.09	1 m, ceramic tile
Langkawi	3.2	2.5	0.78	1 m, ceramic tile
Bintulu	4.7	2.0	0.43	1 m, carpeting

Mean	11.1 ± 6.9	3.0 ±1.5	0.09-1.60	
Outdoor				
Bangi	6.5	2.2	0.34	1 m, open ground
Kuantan	8.2	3.4	0.41	1 m, open ground
Kemaman	1.3	0.4	0.31	1 m, open ground
Cameron Highlands	1.8	0.4	0.22	1 m, open ground
Ipoh	10.3	0.3	0.03	1 m, open ground
Sungkai	1.8	1.3	0.72	ground surface
Langkawi	0.5	0.4	0.80	ground surface
Pasir Gudang	4.8	2.8	0.58	1 m, open ground
Kota Kinabalu	0.9	0.8	0.89	ground surface
Ranau	0.5	0.4	0.80	ground surface
Kuching	0.7	0.2	0.22	1 m, open ground
Serian	1.4	0.3	0.23	1 m, open ground
Mean ± a	3.2 ± 3.4	1.1 ± 1.1	0.03-0.89	

It's important to note that, high EEC thoron was contributed by one of the thoron progenies i.e. ^{212}Pb (half-life 10.6h) which is still present or available in the air even though its parent (^{220}Rn) has completely decayed. Lead-212, due to its longer half-life may be transported and distributed homogeneously throughout a room. Similar to their parents, high or maximum concentrations of thoron (and radon) progenies were also measured during nighttime or early morning and low or minimum in the afternoon. From the results, it is obvious that thoron (and progenies) concentrations in air were significant in Malaysian environment for both indoor and outdoor especially when measurement conducted near floor/wall/ground. Measurement of thoron concentrations close to walls or floors is relevance for human exposure assessment as during sleep people generally breathe air close to walls or floor.

It is recommended to measure both radon and thoron concentrations, because measurement of radon only can lead to inaccurate assessment and underestimate its associated dose. This is due to thoron may interfere in the detection of radon when using certain type o detectors that do not use energy discrimination and that allow rapid gas entry into their sensitive volume (Tokonami, 2001). These include solid state nuclear track detector (SSNTD), electrets and flow-through detectors such as scintillation cell and ion chamber.

Effective Dose

As most of the harms to human are contributed by radon and thoron progenies, their respective concentrations were used to assess the doses to the lung. UNSCEAR (2000) dose conversion factor (DCF) of 9 nSv/(Bq.h.m^3) and 40 nSv/(Bq.h.m^3) for exposure to radon and thoron progenies respectively and occupancy factor of 0.8 (indoor) and 0.2 (outdoor) have been used. The calculated annual effective dose is shown in Table 3. Total annual effective dose (indoor and outdoor) from radon and thoron progenies were 0.9 mSv and 1.1 mSv respectively. An attempt to estimate the annual effective dose from radon gas concentrations (by using established equilibrium factor) also produced comparable results i.e., 0.6 mSv and 0.2 mSv for indoor and outdoor respectively. While for thoron, since thoron gas is not homogeneously distributed and the equilibrium factor was not well established, the dose from thoron gas cannot accurately be assessed.

Table 3: Annual effective dose (mSv) due to radon and thoron

		Indoor	outdoor
Radon progenies	Minimum	0.10	0.03
	maximum	1.16	0.65
	Mean ± a	0.70 ± 0.44	0.20 ± 0.21
Thoron progenies	Minimum	0.45	0.06
	maximum	1.63	0.95
	Mean ± a	0.80 ± 0.42	0.30 ± 0.31

The annual effective dose from radon was comparable with value reported by UNSCEAR (2000) while annual effective dose from thoron was much higher. UNSCEAR (2000) estimated the annual effective doses from radon and thoron were 1.1 mSv and 0.1 mSv respectively. The total annual effective dose from radon and thoron progenies (indoor and outdoor) was 2.0 mSv with thoron contribution was about equal to radon. This shows that the two parameters are equally important to the natural radiation dose. The results were higher than previously estimated (Sulaiman *et al,* 1994) probably due to variation of radon and thoron concentrations in the environment and also the sensitivity equipment used.

Ratio of thoron to radon dose was 1.2 compared to UNSCEAR's value of only 0.09. Ratio of thoron to radon dose based on measurement locations are shown in Figure 2 and 3. Even though only about 10% of the indoor results showed that EEC thoron greater than EEC radon and none of outdoor results, the assessment showed that about 78% and 75% of thoron dose were higher than radon dose indoor and outdoor respectively. This is due to higher thoron DCF compare to radon. It is obvious that for the same concentration; thoron progenies will deliver four times higher dose than radon progenies. Kudo *et al,* (2015) reported that thoron dose was comparable with radon dose while Omori *et al,* (2016) found that exposure to thoron sometimes exceed exposure to radon.

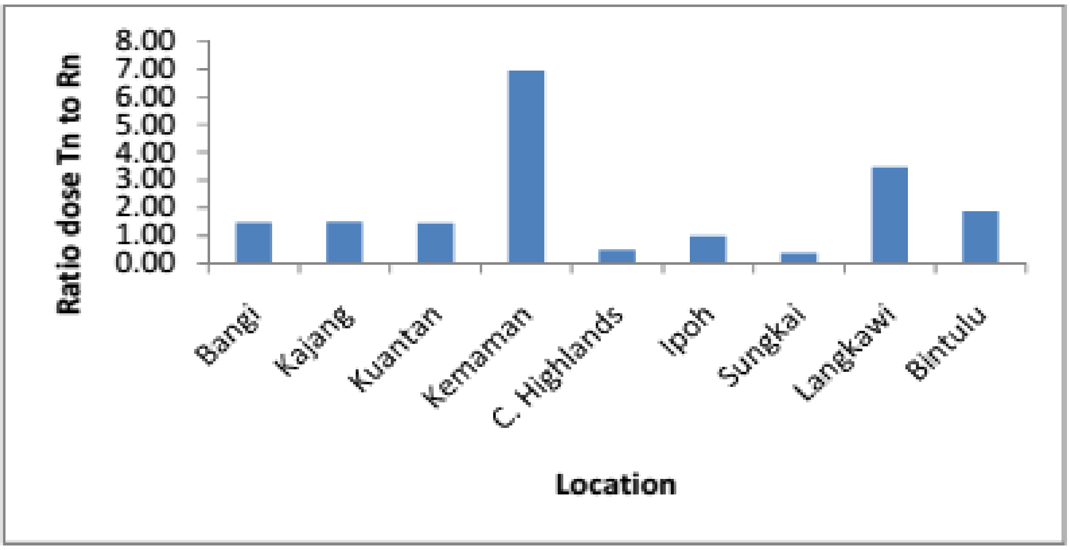

Figure 2: Ratio of thoron (Tn) to radon (Rn) dose indoor

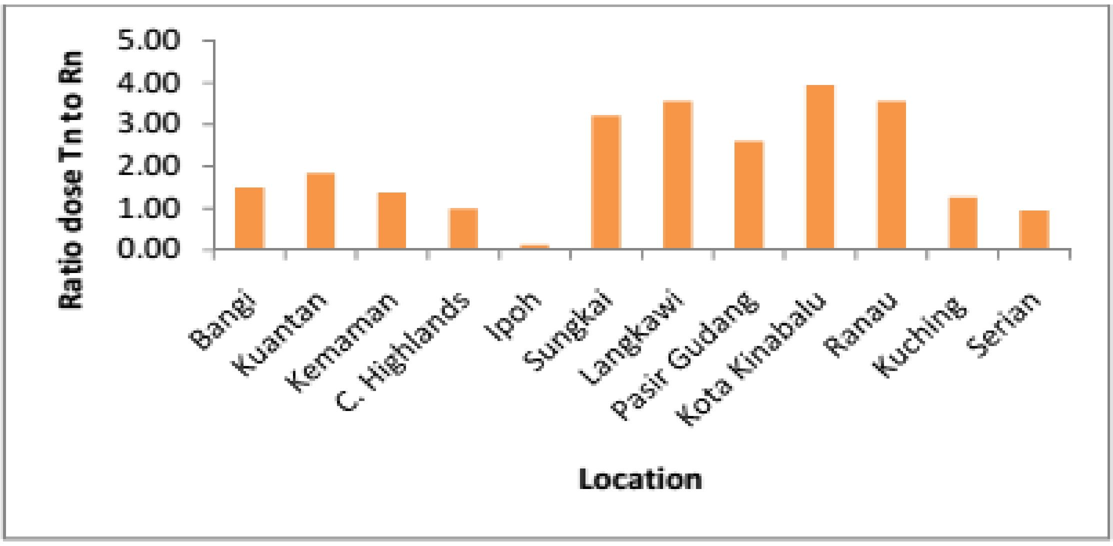

Figure 3: Ratio of thoron (Tn) to radon (Rn) dose outdoor

CONCLUSIONS

Thoron and its progenies concentrations were mostly smaller than those from radon, but in some cases their doses were comparable or higher than that from radon and contribute a significant part of the total dose from natural radiation in the environment. The estimated mean annual effective dose from radon and thoron progenies was 2.0 mSv with thoron contribution was about 50% indicating that the two parameters are equally important. The assessment also indicated that thoron contribution in Malaysian environment cannot be simply ignored because this will probably underestimate the dose receive by the people. The presence of thoron in the environment is not always, as they are often assumed to be, of negligible radiological significance.

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REFERENCES

Chen, J., Moir,D., Sorimachi, A (2011). Characteristics of thoron and thoron progeny in Canadian homes. *Radiation and Environmental Biophysics.* 50 (1): pp 85-89.

Doi, M., Kobayashi, S., and Fujimoto, K (1992). A passive measurement technique for characterization of high-risk houses in Japan due to enhanced levels of indoor and thoron concentrations. *Radiat. Prot. Dosim.* 45(1): 425-430.

Guo, Q., Shimo, M., and Ikebe, Y (1992), The study of thoron and radon progeny concentrations in dwellings in Japan. *Radiat. Prot. Dosim.* 45(1/4): 357-359.

Hosoda, M., Kudo, H., Iwaoka, K., Yamada, R., Suzuki, T., Tamakuma, Y., Tokonami, S (2017). Characteristic of thoron (Rn) in environment. *Applied Radiation and Isotopes.* 120: pp 7-10.

Janik, M., Tokonami, S., Kranrod, C., Sorimachi, A., Ishikawa, T., Hosoda, M., Mclaughlin, J., Chang, B. U., and Kim, Y. J (2013). Comparative analysis of radon, thoron and thoron progeny concentration measurements. *J. Radiat. Res.* 54: 597-610

Kudo, H., Tokonami, S., Omori, Y., Ishikawa, T., Iwaoka, K., Sahoo, S. K., Akata, N., Hosoda, M., Wanabongse, P., Pornnumpa, C., Sun, Q., Li, X., Akiba, S (2015). Comparative dosimetry for radon and thoron in high background radiation areas in China. *Radiat.Prot. Dosim,* 167 (1-3): pp 155-159, https://doi.org/10.1093/rpd/ncv235

McLaughlin, J., Murray, M., Currivan, L., Pollard, D., Smith, V., Tokonami, S., Sorimachi, A., and Janik, M (2011). Long-term measurements of thoron, its airborne progeny and radon in 205 dwellings in ireland. *Radiat.Prot. Dosim.* 145 (2-3): pp. 189-193 doi: 10.1093/rpd/ncr067

Omar, M (2000). Natural radioactivity of building materials used in Malaysia, in Proceedings: 5th International Conference on High Levels of Natural Radiation and Radon Areas, Munich, Germany, 4-7 Sept. 2000.

Omori, Y., Prasad, G., Sorimachi, A., Sahoo, S, K., Ishikawa, T., Sagar, D. V., Ramola, R. C., Tokonami, S (2016). Long-term measurements of residential radon, thoron, and thoron progeny concentrations around the Chhatrapur placer deposit, a high background radiation area in Odisha, India. *Journal of Environmental Radioactivity.* Vol 162-163: pp 371-378. https://doi.org/10.1016/jjenvrad.2016.06.009.

Ramachandran, T.V (2010). Environmental thoron (220Rn): A review. *Iran. J. Radiat. Res.,* 8 (3): 129-147

Ramola, R. C., Gusain, G. S., Rautela, B. S., Sagar, D. V., Prasad, G., Shahoo, S. K., Ishikawa, T., Omori ,Y., Janik, M., Sorimachi, A., Tokonami, S (2012). Levels of thoron and progeny in high background radiation area of southeastern coast of Odisha, India. *Radiat.Prot. Dosim.* 152: 62-65.

Sai'dou, Tokonami, S., Janik, M., Samuel, B. G., Abdourahimi, Nkoulou, N., Emmanuel,

I (2015). Radon-thoron discriminative measurements in the high natural radiation areas of southwestern Cameroon. *Journal of Environmental Radioactivity.* 150: pp 242-246. https://doi.org/10.1016/j.jenvrad.2015.09.006

Shang, B., Chen, B., Gao, Y., Wang, Y., Cui, H (2005). Thoron levels in traditional Chinese residential dwellings. *Radiation and Environmental Biophysics.* 44 (3): pp 193199.

Sorimachi, A., Takahashi, H., Tokonami, S (2009). Influence of the presence of humidity, ambient aerosols and thoron on the detection responses of electret radon monitors. *Radiation Measurements.* 44 (1): pp 111-115. https://doi.org/10.1016/j.radmeas.2008.10.009

Sorimachi, A., Tokonami, S., Omori, Y., Ishikawa, T (2012). Performance test of passive radon-thoron discriminative detectors on environmental parameters.

Radiation Measurements. 47 (6): pp 438-442. https://doi.org/10.1016/j**T**admeas.2012.04.003

Sulaiman, I., Omar, M., Hassan, A (1994). Radon concentrations in air in Malaysia, R&D Seminar, MINT, Bangi, 7-9 November 1994

Sulaiman, I., Omar, M., Elias, M. S (2015). Natural radioactivity in soil in Sabah and Sarawak. *Journal of Nuclear and Related Technology.* 12(1): pp 38-45

Tokonami, S (2010). Why is Rn (thoron) measurement important? *Radiation Protection Dosimetry,* 141(1): pp 335-339. https://doi.org/10.1093/rpd/ncq246

Tokonami, S., Sun, Q., Akiba, S., Zhuo, W., Furukawa, M., Ishikawa, T., Hou, C., Zhang, S., Narazaki, Y., Ohji, B., Yonehara, H., and Yamada, Y (2004). Radon and thoron exposures for cave residents in Shanxi and Shaanxi provinces. Radia. Res., **162: 390396.**

Tokonami, S., Yang, M., Sanada, T (2001). **Contribution From Thoron On The Response Of Passive Radon Detectors.** *Health Physics.* 80(6): 612-615

UNSCEAR (2000). Sources, effects and risks of ionizing radiation, United Nations Scientific Committee on the Effects of Atomic Radiation, United Nation, New York.