Journal of Radiological Protection

Official journal of the Society for Radiological Protection

PAPER

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To cite this article: Chunyu He *et al* 2023 *J. Radiol. Prot.* **43** 011504

IOP Publishing J. Radiol. Prot. 43 (2023) 011504 https://doi.org/10.1088/1361-6498/acb067

Journal of Radiological Protection

RECEIVED
17 November 2022

REVISED
22 December 2022

ACCEPTED FOR PUBLICATION
5 January 2023

PUBLISHED
20 January 2023

PAPER

Impact of humidity and flowrate on the thoron measurement sensitivity of electrostatic radon monitors

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Keywords: thoron activity concentration, measurement sensitivity, electrostatic collection, sampling flowrate, absolute humidity

Abstract

The accurate measurement of thoron activity concentration is an important issue in both thoron exposure evaluation and in reducing its influence on radon measurement. For radon monitors based on electrostatic collection technique and an alpha spectrometry analyser, air humidity and sampling flowrate are key factors influencing the sensitivity of thoron activity concentration measurement. For the purpose of improving thoron measurement sensitivity and stability, theoretical derivation and experimental studies were systemically performed in this study. The results show that thoron measurement sensitivity decreases as a negative exponential function with absolute humidity increasing, and the sensitivity of thoron is much lower than that of radon under the same conditions, which is mainly caused by the small value of the concentration ratio of thoron inside to outside of the chamber. When the air exchange rate of the measurement chamber (sampling flowrate/inner volume) increases, the measurement sensitivity of thoron gas first increases rapidly and then decreases slowly after reaching its maximum at the air exchange rate of 0.24 s^{-1}. In practice, in the normal air exchange rate range (for example <0.05 s^{-1}), increasing the sampling flowrate could greatly improve the thoron measurement sensitivity, which consequently suggests an effective way to update thoron measurement under the present conditions of the monitor.

1. Introduction

Indoor radon (^{222}Rn) exposure has been epidemiologically proved to be the second cause of lung cancer after smoking (WHO 2009). However, as indicated by UNSCEAR (UNSCEAR 2019), there arestill big uncertainties, and the interference in radon measurement caused by its isotope thoron (^{220}Rn) is one of the biggest reasons. Thoron, a decay product of the ^{232}Th radioactive decay series, exists everywhere just as radon does since ^{232}Th naturally occurs in soils, rocks and building materials. Due to the short half-life (55.6 s) of thoron, its emanation is generally considered limited, and as a consequence, its contribution to public exposure is not often taken into account. However, more and more studies have suggested that thoron concentration could be high and its dose contribution might be at the same level as that of radon or even higher in some dwellings (Steinhäusler *et al* 1994, Tschiersch *et al* 2007, Tokonami 2010, Meisenberg and Tschiersch 2011). But thoron's short half-life makes it difficult to measure compared to radon. To measure radon and thoron individually in a real environment, and to assess the influence of thoron on radon measurement, the accurate measurement of thoron activity concentration is required.

Many techniques for thoron activity concentration measurement have been developed based on different kinds of detectors and systems for different purposes. For long-term or large-scale surveys of indoor levels of radon and thoron concentrations, the double alpha-track detector method is usually deployed for its low cost and no need for any power supply (Doi *et al* 1992, Zhuo *et al* 2002, Tokonami *et al* 2005, Chen *et al* 2011).

However, as a passive and integrated measurement method, it only gives the average concentration. An active method, sampling by pumps, can give hourly activity concentrations, and it can be divided into different types depending on the detection methods, such as the lucas scintillation counter, pulse ionisation chambers, and semiconductor detectors combined with electrostatic collection chambers (ECCs) (World Health Organization (WHO) 2009, ICRU 2015).

In the ECC method, thoron gas is pumped into the electrostatic chamber through a filter, where thoron decays into its progeny, positively charged parts are collected on the surface of the semiconductor, and the alpha particles emitted from thoron progeny are detected directly. Due to its excellent discrimination of alpha particles emitted from radon and thoron progeny, the ECC method is one of the most commonly used methods for thoron gas measurement nowadays. However, stable and highly sensitive measurement of thoron activity concentration is usually hard to realise because of the short half-life of thoron and the complexity of the electrostatic collection process. The thoron measurement sensitivity of those systems using the ECC method is found to be affected by humidity (Ashok Kumar *et al* 2014, Tamakuma *et al* 2021), and it is also found that a small change in the flowrate usually considerably changes the sensitivity of thoron (Sumesh *et al* 2013, Hosoda *et al* 2022). Although these issues have been somehow studied, they were investigated separately and mostly through experiment. A further systematic theoretical study combining humidity and flowrate influence is needed.

To improve the thoron sensitivity of the ECC monitors and the stability of thoron measurement, a complete study on the two factors working on thoron sensitivity is carried out, and the corresponding theoretical derivation and experimental verification were performed systematically in this study.

2. Materials and methods

2.1. Theoretical derivation

Figure 1 shows a schematic of the collection and detection process of an ECC monitor for thoron measurement. During the sampling process, thoron gas is pumped into the measurement chamber. In the electrostatic chamber, ^{220}Rn decays into ^{216}Po particles, which are mostly positively charged, and the positively charged ^{216}Po particles could be collected on the Si-PIN detector at the bottom of the chamber as a result of the electrostatic effect, and the alpha particles emitted from ^{216}Po could be detected by the detector. The alpha spectrum obtained is analysed and recorded by a multi-channel analyser as well as a micro controller unit. The reason why only ^{216}Po is included without other thoron decay products is the relatively long half-lives of ^{212}Po (10.6 h) and ^{212}Bi (60.6 min), which will result in a long response time to the concentration change.

Assuming that the thoron gas is uniformly distributed and the electrostatic collection process is stable, the thoron activity concentration in the measurement chamber $C_1(t)$ (Bq m^{-3}) could be as follows:

$$\frac{\mathrm{d}C_1(t)}{\mathrm{d}t} = \frac{qC_1^0}{\mathrm{V}} - \left(\lambda_1 + \frac{q}{\mathrm{V}}\right) C_1, \tag{1}$$

where C_1^0 is the thoron activity concentration in the environment (Bq m^{-3}) and is assumed to be constant, q is the sampling flowrate (m^3 s^{-1}), V is the inner volume of the measurement chamber (m^3), and λ_1 is the radioactive decay constant of ^{220}Rn (s^{-1}). With $C_1(0) = 0$, the thoron concentration in the chamber $C_1(t)$ is as follows:

$$C_1(t) = \frac{qC_1^0\left[1 - \exp\left(-\left(\lambda_1 + \frac{q}{V}\right)t\right)\right]}{V\left(\lambda_1 + \frac{q}{V}\right)}. \tag{2}$$

If λ_2 is the radioactive decay constant of ^{216}Po (s^{-1}), the activity concentration of ^{216}Po in the measurement chamber $C_2(t)$ (Bq m^{-3}) can be given by the following equation:

$$\frac{\mathrm{d}C_2(t)}{\mathrm{d}t} = \lambda_2 C_1(t) - \left(\lambda_2 + \frac{q}{V}\right) C_2(t). \tag{3}$$

Because $C_2(0) = 0$, the concentration of ^{216}Po in the chamber $C_2(t)$ can be written as:

$$C_2(t) = \frac{q\lambda_2 C_1^0}{V}\left[\frac{1}{\left(\lambda_1 + \frac{q}{V}\right)\left(\lambda_2 + \frac{q}{V}\right)} - \frac{\exp\left(-\left(\lambda_1 + \frac{q}{V}\right)t\right)}{\left(\lambda_1 + \frac{q}{V}\right)\left(\lambda_2 - \lambda_1\right)} - \frac{\exp\left(-\left(\lambda_2 + \frac{q}{V}\right)t\right)}{\left(\lambda_2 + \frac{q}{V}\right)\left(\lambda_1 - \lambda_2\right)}\right]. \tag{4}$$

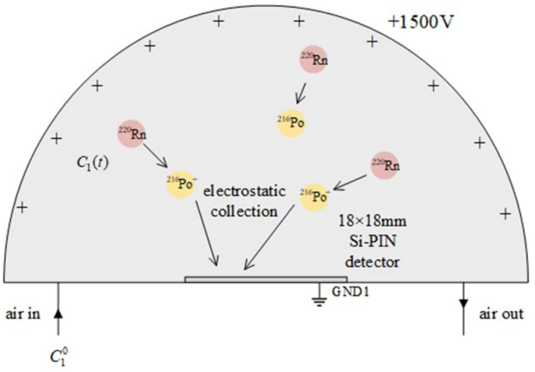

Figure 1. Schematic of the collection and detection process of an ECC monitor for thoron measurement.

During the measurement period, positive ^{216}Po particles are collected on the detector and the integrated alpha spectrum is recorded. The integrated count of 6.8 MeV alpha particles emitted from ^{216}Po during the measurement period T satisfies:

$$D_T = \int_0^T C_2(t)\,\mathrm{d}t \cdot V \cdot \eta_{\text{detection}} \cdot \varepsilon_{\text{collection}}(\text{AH}) = T \cdot S \cdot C_1^0, \tag{5}$$

where $\eta_{\text{detection}}$ is the detection efficiency of the detector, $\varepsilon_{\text{collection}}(\text{AH})$ is the effective collection efficiency of ^{216}Po, which might be a function of absolute humidity (AH, g m^{-3}), and S (cps/(Bq m^{-3})) is the measurement sensitivity of thoron, defined as the count rate per unit activity concentration. It can be given by the formula below:

$$S = \frac{D_T}{T \cdot C_1^0} = V \cdot \frac{q\lambda_2 \cdot \eta_{\text{detection}} \cdot \varepsilon_{\text{collection}}(\text{AH})}{V}\left[\frac{1}{\left(\lambda_1 + \frac{q}{V}\right)\left(\lambda_2 + \frac{q}{V}\right)} - \frac{1 - \exp\left(-\left(\lambda_1 + \frac{q}{V}\right)T\right)}{T\left(\lambda_1 + \frac{q}{V}\right)^2\left(\lambda_2 - \lambda_1\right)} - \frac{1 - \exp\left(-\left(\lambda_2 + \frac{q}{V}\right)T\right)}{T\left(\lambda_2 + \frac{q}{V}\right)^2\left(\lambda_1 - \lambda_2\right)}\right]. \tag{6}$$

If $T \geqslant 3600$ s, the second and third terms in the square bracket above will be zero, and equation (6) can be rewritten as:

$$S \approx V \cdot \frac{q\lambda_2 \cdot \eta_{\text{detection}} \cdot \varepsilon_{\text{collection}}(\text{AH})}{V\left(\lambda_1 + \frac{q}{V}\right)\left(\lambda_2 + \frac{q}{V}\right)}. \tag{7}$$

Since $\eta_{\text{detection}}$ (nearly 50%) and V are constant for a certain system, formula (7) shows that the thoron measurement sensitivity is mainly affected by q and $\varepsilon_{\text{collection}}$, while $\varepsilon_{\text{collection}}$ is mostly influenced by AH from others' results. However, due to the complex behaviour of thoron progeny in the electrostatic field and the difficulty of directly describing the complete influence of humidity on $\varepsilon_{\text{collection}}(\text{AH})$ theoretically, the influence of air humidity on thoron sensitivity is usually measured experimentally nowadays.

Furthermore, formula (7) also shows that the effect of sampling flowrate on thoron sensitivity essentially comes from the air exchange rate of the measurement chamber, which is defined as the ratio of sampling flowrate and measurement chamber volume ($x = q/V$), and the complex relationship between thoron sensitivity and air exchange rate x (s^{-1}) can be given by the following formula:

$$S \approx V \cdot \frac{x\lambda_2 \cdot \eta_{\text{detection}} \cdot \varepsilon_{\text{collection}}(\text{AH})}{\left(\lambda_1 + x\right)\left(\lambda_2 + x\right)}. \tag{8}$$

2.2. Experiment measurement

The schematic diagram of the experimental measurement system setup is shown in figure 2. Experiments were carried out in a thoron chamber (Zhang *et al* 2010), which is a stainless-steel temperature-and-humidity-controllable box with an inner effective volume of 250 l (HASUC HS-250B China). The temperature inside can be adjusted from 0 °C to 100 °C with an uncertainty of 0.5 °C, and the relative humidity can be adjusted from 30% to 98% with an uncertainty of 3%. Thoron sources were provided by

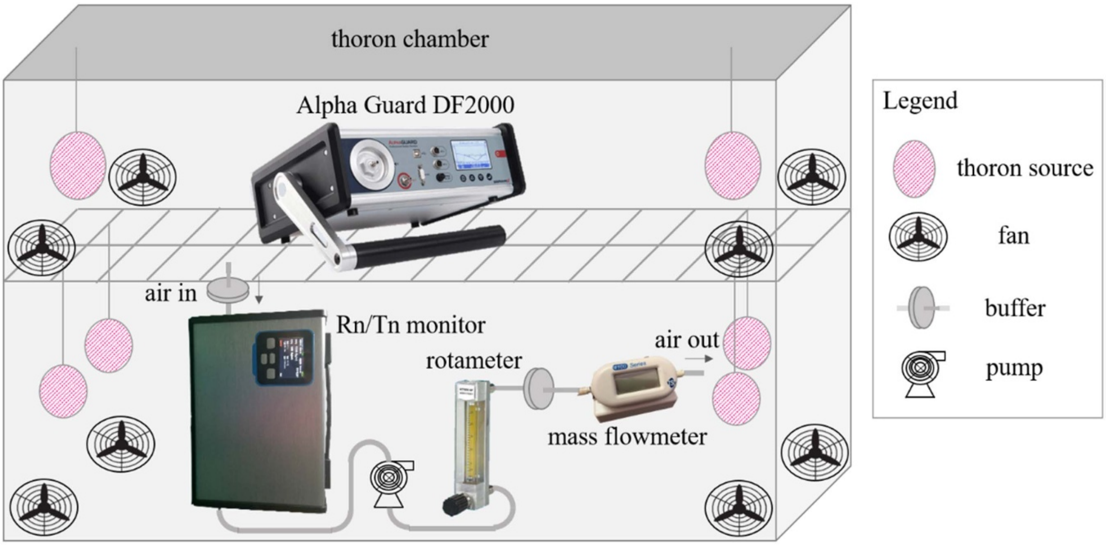

Figure 2. Schematic diagram of the experimental measurement system.

lantern mantles with high ^{232}Th content hanging in the thoron chamber evenly (Sorimachi *et al* 2009, Wang *et al* 2017). Eight fans are used to guarantee a homogeneous thoron atmosphere in the experiment chamber.

An NRM-P01 radon monitor (Sairatec, China) is used in the experiments, which is an active-type monitor consisting of a semiconductor detector and an ECC with 1.25 l inner volume. An 18 × 18mm Si-PIN detector (S3204-09 Hamamatsu Co, Japan) is located at the centre of the bottom, and +1500 V high voltage is applied on the chamber wall to form an electrostatic field. The measurement range is nearly from 2 Bq m^{-3}–4.0 × 10^8 Bq m^{-3} for radon activity concentration and can discriminate different alpha particles from radon and thoron progeny. To make it suitable for our experiment, the monitor was modified by removing its built-in pump and membrane drying tube, and pumps with different flowrates were used outside.

Different flowrates from 0.08 min l^{-1} to 25 min l^{-1} was realised by the combined use of a rotameter (Zhenxing, China) and different diaphragm pumps (Lianhezhongwei technology, China). The flowrate was recorded by a mass flowmeter (TSI4046, USA), and the temperature and the relative humidity were given automatically by the temperature and humidity sensor (Sensirion AG, Switzerland) in the ECC. An AlphaGUARD DF2000 (Saphymo, France) monitor was used to measure the thoron activity concentration in the thoron chamber as a reference, which is calibrated and compared at a stable flowrate of 2 l min^{-1} as it settled especially in the thoron measurement mode of the instrument, and the measurement result can be traced back to the national thoron gas standard of China. To study the influence of flowrate and humidity on thoron measurement, two series of experimental tests were carried out with one factor kept stable and the measurement cycle set to 60 min during the experiment.

3. Results and discussion

3.1. The relationship between thoron measurement sensitivity and absolute humidity

While studying the impact of humidity on the thoron measurement sensitivity of the monitor, the sampling flowrate was fixed at 0.5 lmin^{-1}, the concentration of thoron ranged from 10 000 Bq m^{-3} to 120 000 Bq m^{-3}, and experiments were launched at eight different absolute humidities from 0.13 g m^{-3} to 24.67 g m^{-3}. The absolute humidity from 3.79 g m^{-3} to 24.67 g m^{-3} was achieved by adjusting the temperature and the relative humidity of the thoron chamber, while desiccants (Drierite, USA) were used additionally for achieving 0.13 g m^{-3} with the corresponding factor as 1.2 (Ma *et al* 2012). At each absolute humidity condition, the temperature and the humidity were kept stable for more than 8 h.

The experimental results of thoron measurement sensitivity at various absolute humidities are shown in table 1. For comparison, the radon sensitivity measurement of the same monitor was also carried out at different absolute humidities in the radon chamber of Peking University and the soil gas from 1 m depth in the soil was used as a radon source (Wang *et al* 2022), which realised the radon concentration of 1100–1300 Bq m^{-3}. Results of radon sensitivities at different humidities are shown in table 2, which are obtained from the count rate of 6.0 MeV alpha particles emitted from ^{218}Po.

Table 1. Experimental results of thoron measurement sensitivity at eight different absolute humidities.

Absolute humidity (g m^{-3})	Thoron sensitivity (cph/(Bq m^{-3}))
0.13 (9.6 °C, 1.4%RH)	0.10 ± 0.01
3.79 (15.3 °C, 29.1%RH)	0.075 ± 0.002
5.37 (16.7 °C, 37.9%RH)	0.070 ± 0.001
7.79 (20.2 °C, 44.7%RH)	0.058 ± 0.001
11.22 (24.1 °C, 51.5%RH)	0.045 ± 0.001
15.15 (26.9 °C, 59.5%RH)	0.037 ± 0.001
21.62 (29.0 °C, 75.6%RH)	0.032 ± 0.0002
24.67 (30.8 °C, 78.2%RH)	0.031 ± 0.0001

Table 2. Experimental results of radon measurement sensitivity at seven different absolute humidities.

Absolute humidity (g m^{-3})	Radon sensitivity (cph/(Bq m^{-3}))
0.97 (11.06 °C, 9.69%RH)	1.18 ± 0.01
1.56 (14.54 °C, 13.50%RH)	1.07 ± 0.01
3.25 (17.42 °C, 21.97%RH)	0.88 ± 0.01
6.26 (22.98 °C, 30.59%RH)	0.67 ± 0.01
12.75 (27.92 °C, 47.25%RH)	0.41 ± 0.01
16.16 (29.44 °C, 55.16%RH)	0.38 ± 0.01
21.31 (31.58 °C, 64.81%RH)	0.29 ± 0.004

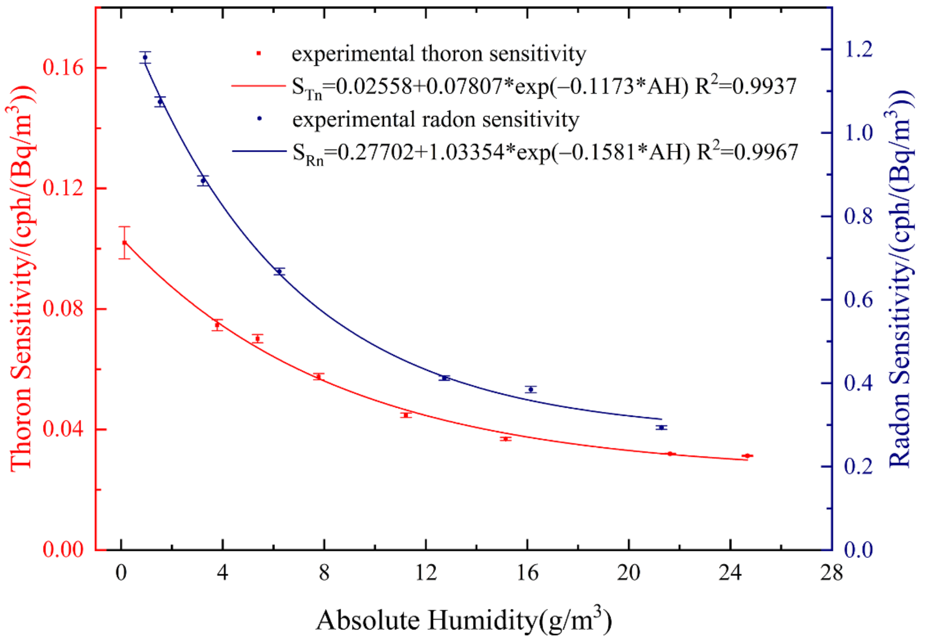

Figure 3. Experimental results and their fitted curves of the relationship between the measurement sensitivity of thoron/radon and absolute humidity.

The experimental results and their fitted curves of the relationship between radon/thoron measurement sensitivity and absolute humidity are shown in figure 3. The results show that radon and thoron measurement sensitivities decrease clearly as absolute humidity increases, and the relationship between thoron measurement sensitivity and absolute humidity can be fitted by the following function:

$$S_{\mathrm{Tn}} = 0.02558 + 0.07807\ \exp\ (-0.1173 \times \mathrm{AH}),\ 0.1\ \mathrm{g\,m}^{-3} \leq \mathrm{AH} \leq 25\ \mathrm{g\,m}^{-3}. \tag{9}$$

While for radon, it is as follows:

$$S_{\mathrm{Rn}} = 0.2770 + 1.0335\exp\left(-0.1581 \times \mathrm{AH}\right),\ 1\,\mathrm{g\,m}^{-3} \leq \mathrm{AH} \leq 21\,\mathrm{g\,m}^{-3}. \tag{10}$$

The corresponding shapes of fitted curves are quite similar to the previous results of radon measurement studies (Ui *et al* 1998, Roca *et al* 2004, De Simone *et al* 2016, Zhang *et al* 2021). The quite nice fitting of negative exponential functions with $R^2 = 0.9937$ and 0.9967 for thoron and radon separately might result

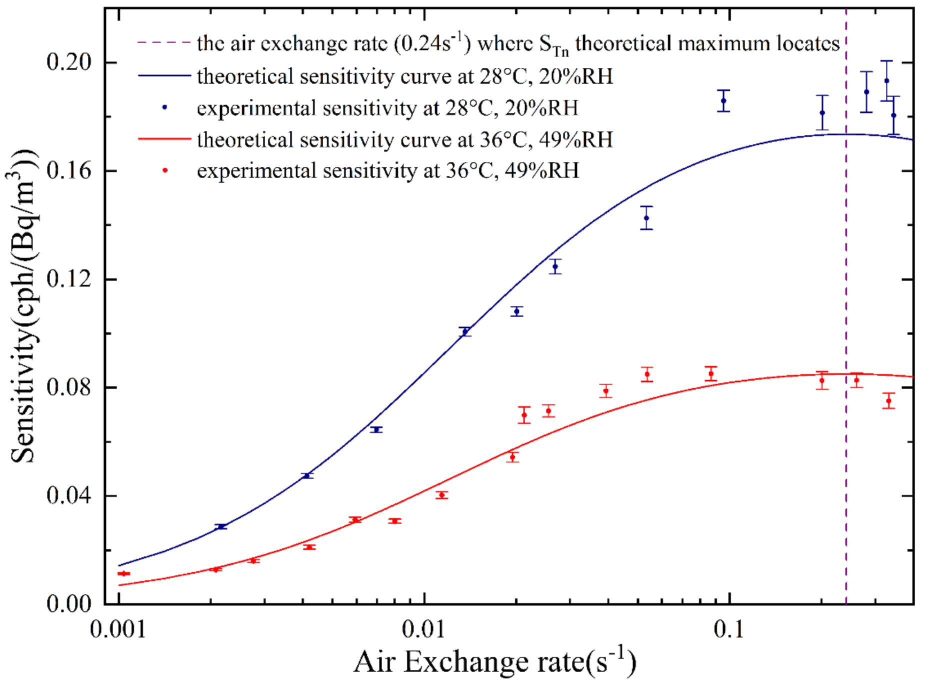

Figure 4. Experimental results and theoretical curves of the relationship between thoron sensitivity and air exchange rate.

from the neutralisation mechanism of water molecules with radon and thoron positive progeny, and it causes the percentage of charged ^{218}Po/^{216}Po to decrease exponentially with increasing AH.

Also, the results indicate that thoron sensitivity at the lowest absolute humidity (0.13 g m^{-3}) in the experiment is about 3.23 times that at the highest (24.7 g m^{-3}), and at 20 °C, 40%RH, (AH at 6.89 g m^{-3}), it is about 59% of that under dry conditions (0.13 g m^{-3}). Moreover, radon sensitivity at the lowest absolute humidity (0.97 g m^{-3}) in the experiment is about 4.07 times that at the highest (21.3 g m^{-3}).

Even though the similar tendencies of radon/thoron measurement sensitivity decreasing with AH are observed, the big difference between their values is clear in figure 3. The measurement sensitivity of thoron is much smaller than that of radon at the same absolute humidity: for example, S_{Tn} is only about 9.66% of S_{Rn} at 20 °C, 40%RH (AH at 6.89 g m^{-3}). The big difference between the half-lives of radon/thoron gas and their progeny ^{218}Po/^{216}Po might be the main reason.

According to formula (2), when $t \geqslant 10$ min, $\exp\left(-\left(\lambda_1+\frac{q}{V}\right)t\right)\approx 0$, therefore $\frac{C_1(t)}{C_1^0}\approx\frac{q}{V\left(\lambda_1+\frac{q}{V}\right)}\approx 0.35$ for $V = 1.25$ l at $q = 0.5$ lpm, which means only 35% of thoron can enter the chamber, and this cloud is the most important reason. The inhomogeneous distribution of thoron gas and ^{216}Po in the measurement chamber, which leads to less electrostatic collection, might be the second reason. Besides, the decay of thoron during sampling at the entrance and filter is also a possible reason.

It should be noticed that the fitted formulae acquired above can only quantitatively describe the corresponding relationships of the monitor adopted in this study. The chamber volume, the chamber and detector shape, the electric field and the sampling flowrate might affect the specific parameters in formulas (9) and (10), but the negative exponential function will be more or less similar for different electrostatic monitors.

3.2. The relationship between thoron measurement sensitivity and flowrate

When it comes to the impact of sampling flowrate on thoron measurement sensitivity, we fixed the temperature and the relative humidity at 28 °C, 20%RH (AH at 5.42 g m^{-3}) and 36 °C, 49%RH (AH at 20.34 g m^{-3}), respectively. At 5.42 g m^{-3} AH, the concentration of thoron ranged from 5000 Bq m^{-3} to 12 000 Bq m^{-3}, and the flowrate changed from 0.2 l min^{-1} to 26 l min^{-1} at 12 different levels with the corresponding air exchange rate (q/V) varying from 0.002 q V^{-1} to 0.34 s^{-1}. While at 20.34 g m^{-3} AH, the concentration of thoron ranged from 18 000 Bq m^{-3} to 33 000 Bq m^{-3}, and experiments were carried out at 16 flowrates, from 0.08 l min^{-1} to 25 l min^{-1}, with the air exchange rate from 0.001 s^{-1} to 0.33 s^{-1}. At each flowrate, the measurement was continuously carried out for more than 10 h.

The experimental results and theoretical curves, which were calculated from formula (8) with $\eta_{\mathrm{detection}} = 0.5$ and $\varepsilon_{\mathrm{collection}}$ (AH) at two different absolute humidities, are shown in figure 4. The theoretical results indicate that thoron measurement sensitivity first increases rapidly, then reaches its maximum around the air exchange rate of 0.24 s^{-1}(marked in figure 4), and decreases slowly afterwards as the air exchange rate increases. In addition, the experimental results are in good agreement with theoretical results at both absolute humidities in figure 4. The trend observed here is consistent with those results previously reported (Sumesh *et al* 2013, Hosoda *et al* 2022), which might be due to the quick decay of thoron gas and ^{216}Po before detection at lower flowrate, and after reaching its maximum, thoron gas is removed from the measurement chamber before detection. In practice, in the normal working range (such as air exchange rate $<0.05\ s^{-1}$, for a 1.25 l chamber, flowrate $<3.8\ l\ min^{-1}$), the measurement sensitivity of thoron increases sharply with increasing flowrate. For instance, when we increase the sampling flowrate from 0.5 $l\,min^{-1}$ to 1 $l\ min^{-1}$, the thoron measurement sensitivity of the ECC monitor we adopted would increase by 48%, from 0.069 cph/(Bq m^{-3}) to 0.103 cph/(Bq m^{-3}) at AH = 5.42 g m^{-3}.

4. Conclusion

Higher sensitivity and stable measurement are desired for precise measurements of thoron activity concentration. To increase thoron measurement sensitivity and to promote measurement stability, a systematic study is performed in this paper, where the relationship between thoron measurement sensitivity and humidity, as well as flowrate, is acquired through experimental measurement and theoretical analysis.

Results show that both sampling flowrate and environmental absolute humidity work on the thoron sensitivity of the ECC monitors. Increasing the sampling flowrate could be quite an effective way to achieve a more sensitive thoron measurement. However, although reducing absolute humidity can increase the measurement sensitivity of radon, it is not as effective to thoron due to its decay in the drying module during sampling. Considering the humidity correction using experiment fitting lines, the stability of thoron measurement will be upgraded greatly without using desiccants. The overall picture of thoron sensitivity influenced by humidity and sampling flowrate not only helps us with a better physical understanding but is also valuable for radon/thoron monitor design and development.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Acknowledgment

This work was supported by the National Natural Science Foundation of China (No. 11975310).

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