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Thoron standard source

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HIGHLIGHTS

- Produce a thoron source that provides a reproducible stream of thoron using standard thoron detectors and a thorium nitrate solution.
- Uses the standard RAD7 equipment to bubble a solution to produce the thoron.
- Uses gamma spectroscopy to predict the percentage of thoron that escapes.
- Initial condition investigated that shows that the system reaches a steady state after less than 10 min.

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ABSTRACT

Thoron (^{220}Rn) has been identified as a possible health concern in specific places such as monazite processing plants and (rare-earth) mines. The short half-life of thoron (55.8 s) makes thoron calibration sources and thoron chambers less common than the corresponding radon (^{222}Rn) ones. In this paper an inexpensive and straight forward but accurate thoron source is described that can easily be set up in typical nuclear environmental laboratories. The source of thoron is a solution of $Th(NO_3)_4$ in water. Thoron is extracted by bubbling air through the solution using an aerator. The gamma rays from the solution are measured at the same time. The thoron activity concentration in the exit stream follows from the reduction in the intensity of the gamma rays from the progeny of thoron over time.

1. Introduction

In this paper we will term ^{220}Rn and ^{222}Rn, thoron and radon respectively. The health risks of radon are well known and have been studied for several decades (BEIR VI (Committee on Health Risks of Exposure to Radon), 1999). The dose due to thoron has been underestimated as it was expected that its short half-life would usually lead to decay before it reaches the atmosphere. Several recent studies have indicated that there is a need to investigate thoron (Radiation Protection Dosi, 2010) especially in specific areas such as monazite mines and heavy mineral processing plants. The renewed interest in thorium reactors (Dolan, 2017) which will require an increase in the processing of minerals containing thorium, will make the need for thoron studies even more important.

One of the major difficulties in studying thoron concentrations and exhalation rates is the lack of thoron chambers and reliable standard sources compared to the many radon chambers that exist around the world. Specialised standards such as the one developed by PTB (Rottger et al., 2010, 2014) are costly and unlikely to be widely copied. Several publications (Agarwal et al., 2014; Janik and Yonehara, 2015; Sumesh et al., 2013; Kanse et al., 2013; Wang and ZhangGuo, 2017; Calamosca and Penzo S, 2010; Tan et al., 2014) have looked at this issue and have discussed the possible uncertainties in measurements by commonly used instruments which were primarily designed to measure radon such as the RAD7 ([13] www.Durridge.com.).

In this article, an inexpensive and straight forward method is described that can be used as a reliable thoron source. The method is a variant of the ideas applied in the PTB method and the radon in water standard developed by NIST (Collé, 1997).

2. Materials and method

Many commonly used radon detectors such as most alpha track devices, cannot distinguish between thoron and radon. The RAD7 monitor is one of the most widely used continuous radon monitors that can differentiate between thoron and radon. The detector contains a

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surface barrier detector that measures radon and thoron by detecting the energy of the alpha particles from ^{218}Po and ^{216}Po that results from the decay of radon and thoron, respectively. As with most thoron measurement systems, the short half-life of the thoron requires corrections which in the case of the RAD7 depend on the volume of the pipes and the desiccant as well as a correction for the decay inside the chamber of the detector system. A thoron measurement is based on the detection of alpha particles from the decay of ^{216}Po. A requirement for thoron detection is that the ^{216}Po ions reach the alpha detector. This prerequisite is not always met due to the very short half-life (0.14s) of the ^{216}Po (Tan et al., 2014).

The interest in thoron has necessitated the need for a standard thoron source or thoron chambers to calibrate detectors. Solid thoron sources have the same disadvantage as solid radon sources: the release depends on the temperature and pressure in an undetermined and irreducible way or may just be low (Wang and ZhangGuo, 2017). For this reason we have developed a thoron source that uses a thorium nitrate solution in water. The short thoron half-life makes the exhalation of thoron from the solution too small to be used directly. This problem is solved by bubbling air through the solution to release the thoron very similar to the method used in the RAD H_2O, an accessory of a RAD7 detector to measure radon concentrations in water (www.Durridge.com.; Talha et al., 2008). The problem with this method is the uncertainty in the fraction of thoron that is successfully removed by the bubbling. A simultaneous measurement of the gamma rays released from the solution from the decay chain of ^{232}Th is made to quantify this similar to what has been done in other work (Rottger et al., 2010, 2014; Wang and ZhangGuo, 2017). The relevant set-up to do this is shown in Fig. 1 with details on traceability given in Table 1. Room air is pumped through a flowmeter into a vial containing a solution of $Th(NO_3)_4$. An aerator converts the flow into a spray of small bubbles in the vial. The bubbles collect the gaseous component in the solution (thoron) and remove it from the vial. The activity of this solution is measured via the emitted gamma rays detected with a 7.5 cm diameter, 7.5 cm long cylindrical NaI(Tl) crystal connected via an MCA to a desktop computer. The air leaves the vial, and passes passively a small drying tube and a RAD7 monitor.

By removing Thoron from the solution, the intensity ratio is affected between gamma rays emitted in the decay of ^{232}Th prior (especially ^{228}Ac) and post thoron (such as by ^{208}Tl) since post-thoron gamma rays are removed from the vial whereas the prior-thoron gammas, remain in the solution. Hence the change in intensity ratio is a measure of the fraction of the formed thoron that is removed. The decay scheme (Fig. 2) of thorium, compared to one of the ^{238}U decay series, has the advantage of strong gamma rays from decays prior to thoron in the decay series (specifically ^{228}Ac) as well as decays from thoron progeny in the decay chain, such as ^{208}Tl. *The fact that the gamma rays originating from before and after thoron in this chain are both present (see Table 2), indicates that their difference in intensity is a measure of the quantity of thoron that is released and is the basis for the method described in this paper.*

Table 2 lists the gamma ray energies in the thorium decay chain that are relevant to this study. An added complication is the half-life of ^{212}Pb (10.6 h) which means that the measurement has to be taken over several hours.

To produce a well characterised source, 10.2 ± 0.14 g of Th $(NO_3)_4.6H_2O$, the common form of thorium nitrate at room temperature (Benz et al., 1987), was dissolved in 1.325(2) litres of water. A sample of 28.7 ± 0.14 ml of this solution was poured into a glass bottle with a volume of 40 ml, a standard bottle supplied as part of the RAD H_2O system of Durridge ([13] www.Durridge.com.). The bottle was not filled to the brim since we wanted to avoid bubbles in the exit pipe. Assuming that the thorium nitrate was pure and in secular equilibrium, the thorium nitrate mass in the bottle implies a concentration of 354 ± 5 Bq. As a check, the bulk of the solution was analysed in a Marinelli Beaker with an HPGe system using the method described in (Talha et al., 2008, 2010) resulting in an activity concentration of 11 460 ± 180 $Bq.L^{-1}$.

This result implies that the sample bottle contains an activity of 331 ± 5 Bq. This value is reasonably close to the calculated value of about 354 Bq, but does not agree with that value. We do not have a reliable history of the salt and its purity. Therefore, **we will use the measured value based on the HPGe result** in the rest of this paper. A new sample of $Th(NO_3)_4$ would solve this discrepancy, but our old sample has the advantage that secular equilibrium has been reached since the activity concentration extracted from the ^{228}Ac and ^{208}Tl peaks differed by less than 3%. It should be noted that for the validity of our method, secular equilibrium between ^{232}Th and ^{228}Th is not required.

Air was bubbled through the solution using the stainless steel aerator that is supplied as part of the radon in water accessory (RAD H_2O) of the RAD7 system. The aerator has a glass tube that forces air through a glass frit inside the solution as shown in Fig. 1. The RAD7 pump could be used to bubble the air through the solution, but we preferred to use an external pump with a flow meter to allow us flexibility to test various pumping speeds.

Gamma-ray spectra were measured with a standard 7.5 cm × 7.5 cm NaI(Tl) detector(REXON) connected via a ScintiSPEC™ Multi Channel Analyser to a personal computer. The winTMCA (https://detectionsupport.) software was used to operate the MCA. The system showed slight gain drift (Gilmore, 2008) which can be a major problem when analysing data taken over several hours as in this work. Therefore, gain stabilisation was used to keep the background peak at 1460 keV (from ^{40}K) in more or less the same channel by changing the fine gain every 6 min. The software allows for this to be done automatically but we preferred to do this using a batch file which allows us to interrogate the amount of drift and to choose a longer period for a better peak to form before changing the gain.

No effort was made to reduce background radiation by shielding.

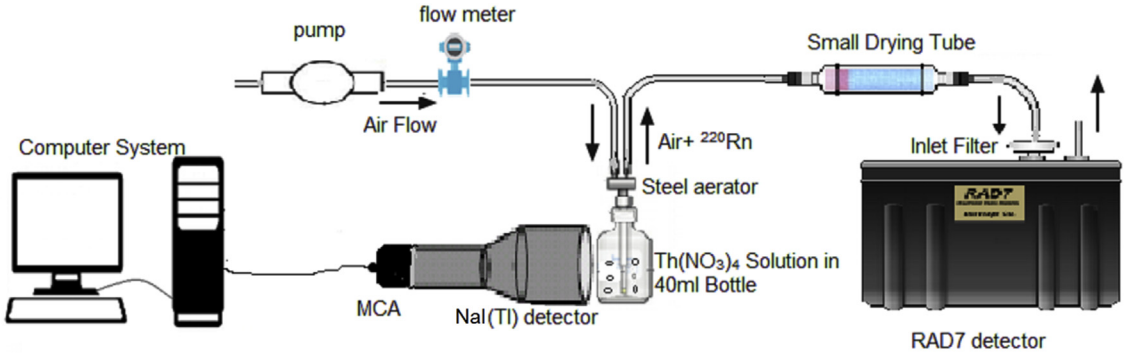

Fig. 1. The set-up used to measure the gamma rays from a solution while simultaneously bubbling air through the solution to provide a stream of air containing thoron that can be measured with the RAD7 as indicated.

Table 1
Description on instruments used and information about traceability.

Equipment used	Information	Reading accuracy
Air Pump	Brailsford Model TD-3LS	Specifications not important
Flow meter	Dwyer Model MMA-21	4% of Full Scale- See www.dwyer-inst.com that states that it meets "requirements of EU Directive 2011/65/EU (RoHS II)"
Steel Aerator, pipes and drying tube and bottle are standard RAD7 equipment sold as part of their H2O system.	www.Durridge.com	The volume of the pipes and drying are less than 30 cm^3. The exact value is not important for our purpose.
The Th $(NO_3)_4.6H_2O$ in the solution was supplied by Fluka AG Buchs, Switserland.	The concentration of the ^{224}Ra that produces the thoron was NOT based on the amount of Thorium Nitrate, but on the HPGe measurement of a solution of this material.	The accuracy of this solution is based on the method of ref 16 that was cross referenced with a KCl solution and a reference source supplied by National Metrology Laboratory of South Africa
Rad7 used for thoron detection based on the alpha detection from the decay of ^{216}Po.	www.Durridge.com Note that we are not trying to check the accuracy of the RAD7 but simply using the RAD7 as a check on our prediction.	Thoron measurements has an uncertainty of about 30% according to procedures described in manual (Revision 7.3.3. © 2015 DURRIDGE Company.). Calibrated by Durridge on an annual basis -

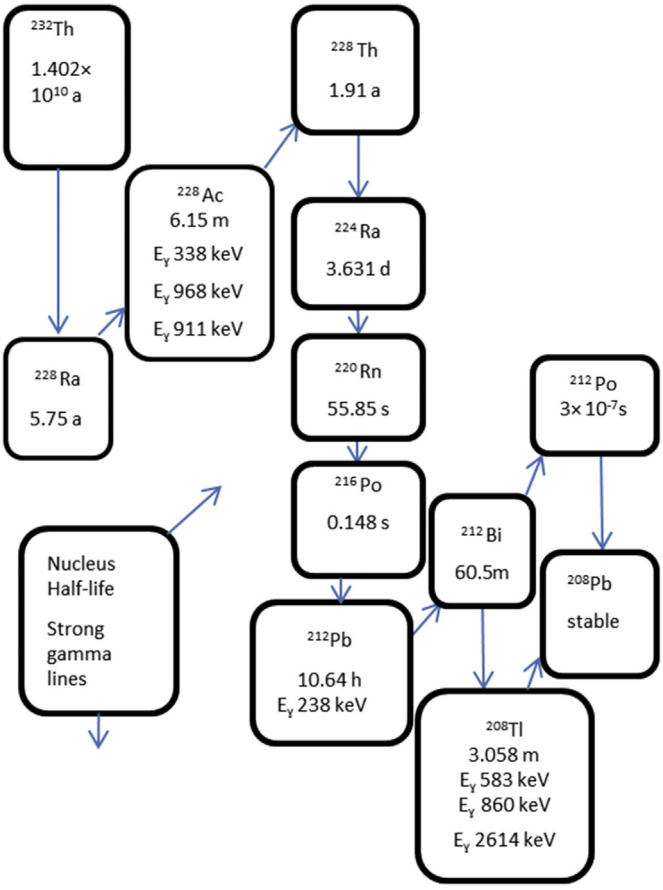

Fig. 2. The ^{232}Th decay series indicating the half-lives as well as gamma ray energies (in keV) that are relevant to this work. More accurate energy values are presented in Table 2. Note the two branches in the ^{212}Bi decay which precedes the ^{208}Tl decay (http://www.nucleide.org/D).

The background was fairly constant and can be subtracted without the complications of a set-up with lead blocks. This was in the spirit of the suggested thoron source-simple, cost effective and fairly accurate. Note that the method described in this work does not require an efficiency calibration of the detector for the geometry used since we use the intensity ratios of gamma lines. There is also no need for any correction to take into account coincidence summing that is present in, for example, the 2614 keV line (http://www.nucleide.org/D). In order to check for consistency, an MCNP-X (Pelowitz, 2005) simulation was done for our set-up. The geometry for the partially filled bottle and the detector (as far as is available from the manufacturer) was used. A fit was made to the simulated relative efficiency, ε_{rel} above 100 keV in the form

$$\varepsilon_{rel} = a\left(\frac{E}{E_0}\right)^{-b}$$

where a and b are constants, E_0 is arbitrarily set at 1 keV and E is the energy. The *absolute efficiency* was then obtained by measuring the counts in the peak at 1460 keV from ^{40}K for a known activity in a 9.96 g solution of KCl in a sample bottle.

3. Results

Fig. 3 shows the spectrum measured during the first hour of measurement and also the spectrum obtained after 65 h of pumping. The bubbling does not affect the non-gaseous progeny but does remove thoron from the solution and therefore the gamma ray peaks, such as the 583 keV and 2614 keV peaks from ^{208}Tl that originate from the decay of thoron progeny, are reduced in intensity. The rate of reduction depends mainly on the decay of ^{212}Pb with a half-life of 10.6 h. The peaks originating from the ^{232}Th decay chain *prior* to the formation of thoron, such as the 911 keV line from ^{228}Ac, are not affected. The reduction in count rate and hence the rate at which thoron is removed, can be extracted from the peaks *post* the thoron decay. The 583 keV and 2614 keV peaks are obvious candidates. The 583 keV peak has the advantage that the count rate is noticeably higher which leads to a smaller uncertainty in the counting statistics. The disadvantages of this peak are the non-linear background and the contribution of the 511 keV annihilation peak to this peak which is not affected by the pumping. After testing both peaks, it was found that the 2614 keV peak was the more reliable one for our set-up. This may well be different if an HPGe detector is used.

Spectra were collected for periods of one hour. Fig. 4 shows the count rate in the 2614 keV peak as a function of the time for a period of 14 h. The figure also shows the count rate in the (nominal) 911 keV peak. Both count rates have been normalised for clarity. The broad "911 keV" peak actually runs from about 800 keV to 1070 keV and includes contributions from the 964 and 968 keV peaks which also stem from the decay of ^{228}Ac and can be included in this peak. However, the peak also contains some counts from the 860 keV line which is from the decay of ^{208}Tl. This makes only a small contribution to the peak since the branching ratio for this decay is only 0.045. The count rates in the 911 peak and the 338 keV peak remain nearly constant while pumping and the decrease in count rate for the 583 and 2614 keV peaks gives us confidence that the reduction in the count rate is indeed as a result of the removal of the thoron.

The pumping was continued for more than two days and the count rates in the 2614 peak for later times are plotted in Fig. 5. The nett counts in the first hour in the 2614 keV peak was 6800 while the average count rate after 60 h was around only 1300 counts per hour.

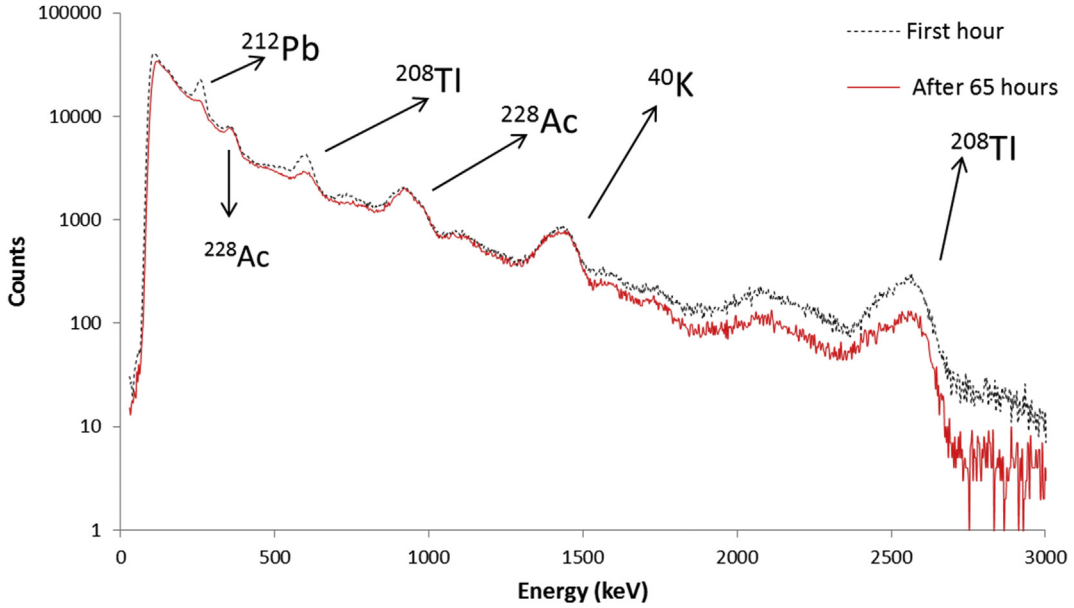

Fig. 3. Spectrum from the Solution during the first hour of measurement and after 65 h.

Table 2
Gamma ray lines in the spectrum of the ^{232}Th decay series that are relevant to this study and visible on Fig. 3. The Branching Ratios are extracted from ref (http://www.nucleide.org/D). The ^{208}Tl values take the branching from ^{212}Bi to ^{208}Tl into account.

Nucleus	Gamma ray energy (keV)	Branching Ratio
^{212}Pb	238.632(2)	0.436(5)
^{228}Ac	338.320(5)	0.114(4)
^{208}Tl	583.187(2)	0.305(11)
^{208}Tl	860.53(2)	0.0446(4)
^{228}Ac	911.196(6)	0.262(4)
^{228}Ac	968.960(9)	0.159(5)
^{40}K (Background in room)	1460.822(6)	0.1055(11)
^{208}Tl	2614.511(10)	0.3584(7)

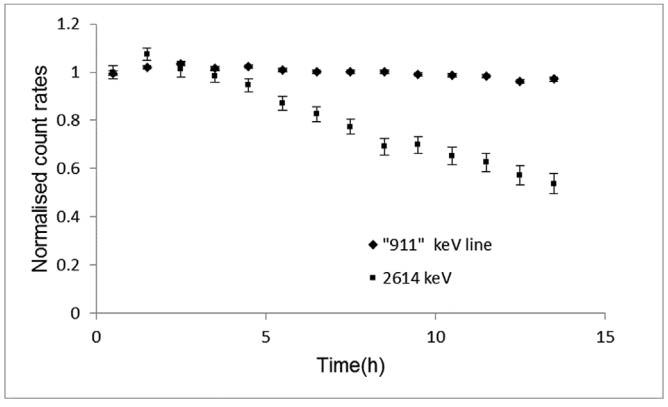

Fig. 4. Count rates in the "911" keV peak (diamonds) compared to the 2614 keV peak (squares).

The count rate was normalised to the initially expected activity of the ^{208}Tl. The initial count rate is consistent with the MCNP-X simulation that predicts an efficiency of 0.018(4) for this peak which would lead to an activity of 100 ± 20 Bq (=6800/3600)/0.018). Fig. 5 indicates that, as expected, the count rate reduces roughly in line with the half-life of ^{212}Pb (10.6 h), but tends to a constant value and not to zero, even after five half-lives of the ^{212}Pb. This indicates that a certain fraction of the thoron decays in the solution before it is pumped out. The count

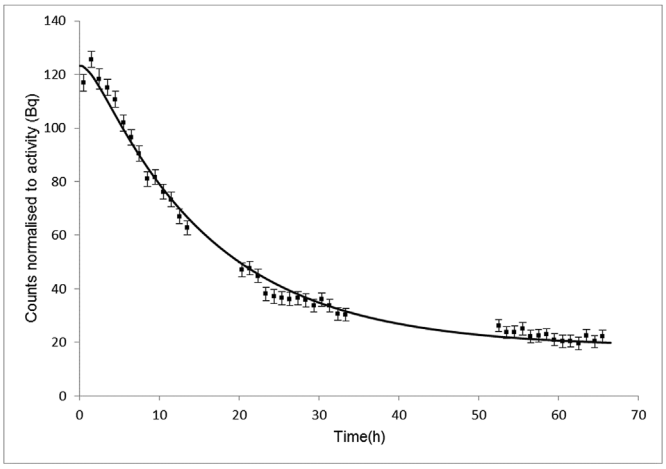

Fig. 5. The ^{208}Tl activity (line) as calculated with the Bateman equations and the (normalised) measured activity using the 2614 keV peak in Thallium, as a function of the time.

rates at times larger than 60 h show that more than 80% of the thoron is removed. To find a more quantitative result, the data has been fitted with a calculation based on the Bateman equations Bateman, 1910 for the ^{208}Tl decay.

The calculation starts with the decay series (starting with thoron) in secular equilibrium with a given concentration at t = 0 after which the feeding from the thoron decay to ^{216}Po is greatly reduced. A small feeding term from ^{220}Rn (of size *S* nuclei per second) is included that represents the thoron nuclei that decay in the water and are not pumped out. The half-life of ^{216}Po is so short that the equations can start with the decay of ^{212}Pb which has a half-life of 10.6 h that dominates the rate of reduction in the ^{208}Tl activity.

$$N_{Tl}(t) = \lambda_{Pb}\lambda_{Bi,Tl}\left[\left(N^0_{Pb}\,\frac{e^{-\lambda_{Pb}t}}{(\lambda_{Bi}-\lambda_{Pb})(\lambda_{Tl}-\lambda_{Pb})}\right) + \frac{S(1-e^{-\lambda_{Pb}t})}{(\lambda_{Pb}(\lambda_{Bi}-\lambda_{Pb})(\lambda_{Tl}-\lambda_{Pb}))}\right] + \left[\left(N^0_{Pb}\,\frac{e^{-\lambda_{Bi}t}}{((\lambda_{Pb}-\lambda_{Bi})(\lambda_{Tl}-\lambda_{Bi}))}\right) + \frac{S(1-e^{-\lambda_{Bi}t})}{(\lambda_{Bi}(\lambda_{Pb}-\lambda_{Bi})(\lambda_{Tl}-\lambda_{Bi}))}\right] + \left[\left(N^0_{Pb}\,\frac{e^{-\lambda_{Tl}t}}{((\lambda_{Pb}-\lambda_{Tl})(\lambda_{Bi}-\lambda_{Tl}))}\right) + \frac{S(1-e^{-\lambda_{Tl}t})}{(\lambda_{Tl}(\lambda_{Pb}-\lambda_{Tl})(\lambda_{Bi}-\lambda_{Tl}))}\right] + \lambda_{Bi,Tl}\times\left[\frac{N^0_{Bi}e^{-\lambda_{Bi}t}}{(\lambda_{Tl}-\lambda_{Bi})}\right] + \lambda_{Bi,Tl}\times\left[\frac{N^0_{Bi}e^{-\lambda_{Tl}t}}{(\lambda_{Bi}-\lambda_{Tl})}\right] + N^0_{Tl}e^{-\lambda_{Tl}t} \quad (1)$$

where the λ_X is the decay constant for nucleus X and $\lambda_{X,Y}$ is the *partial* decay constant for nucleus X to decay to nucleus Y. This is important in this decay series since ^{212}Bi decays to ^{208}Tl with a probability of only 36%. N^0_X represents the initial number of nuclei of nucleus X in the solution.

The value of S, the source due to the thoron that does not get bubbled out, was varied and an overall normalisation was applied to the measured count rate in the 2614 peak as a function of the time. The best fit value for S was 51.0 Bq which implies that around 15% (51/331) of the thoron was **not** pumped out. The obtained χ^2 value is 1.3 per degree of freedom. The obtained value of S would lead to a final activity concentration of $0.36 \times S = 18$ Bq in agreement with Fig. 5 for times larger than 60 h.

The thoron nuclei in the air stream leaving the bottle can be calculated as $x \times N$ nuclei per second where x is the fraction of the thoron nuclei that is pumped out and N is the number of thoron nuclei that are created every second ($N = 331$ in our case) from the decay of ^{224}Ra.

At the flow rate of 0.6 L per minute the activity concentration(C) in the air stream leaving the bottle becomes:

$$C = \frac{x \times \lambda_{thoron} \times N}{Flow\ rate} = \frac{x \times 0.0125 \times N}{(0.6 \times 10^{-3})/60} = 350\ \text{kBq. m}^{-3} \quad (2)$$

for our values of $N = 331$ and $x \approx 0.85$. The uncertainty in C is dependent on the uncertainties in the masses of the water and $Th(NO_3)_4$, the flow rate and the fraction pumped out. This experimental set-up provides a stream of air where the thoron concentration is known fairly accurately. This can be used as a thoron standard that can be easily set-up and re-used after the ^{212}Pb has regenerated. Table 3 lists the uncertainty budget for the calculation in equation (2).

As shown in Fig. 1, the air from the aerator with this expected thoron concentration was measured with the RAD7 using the standard method described in the manual. Fig. 6 shows the thoron activity concentrations that were obtained, in good agreement with equation (2).

We also studied the initial behaviour of the system. Given an initial activity value of 331 Bq in the bottle, this implies that there are about $331/\lambda \approx 26\,500$ thoron atoms in the bottle. The equilibrium that is reached, where the source term is only around 50 Bq, implies only about 4000 thoron nuclei in the bottle. Therefore, the initial pumping should remove *more* thoron atoms than during the steady state situation. In order to check this and to see when the equilibrium state is reached, the RAD7 was set to measure in two minute intervals and was started before the pump was switched on. The resulting RAD7 readings are shown in Fig. 7 when the pump was switched on two minutes *after* the RAD7 started measuring. The figure shows that there was indeed an initial jump to a higher value than the steady state value. The equilibrium value was reached about 6 min after the pump was started.

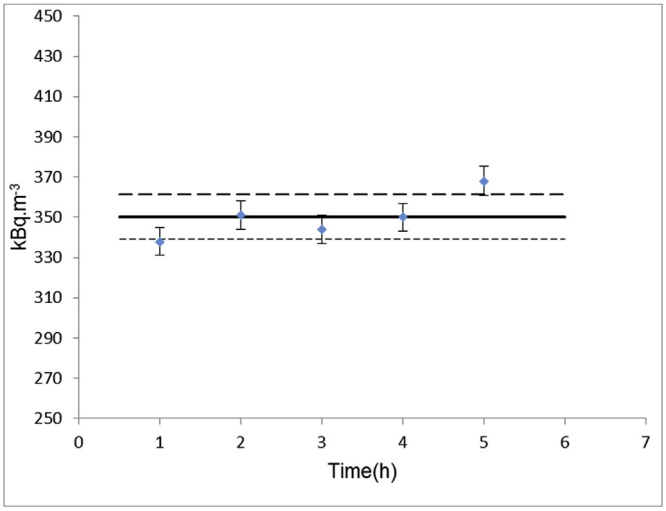

Fig. 6. The thoron concentration in the exit stream of the bubbler as obtained with the RAD7. The uncertainties are as given by the RAD7 based on statistical uncertainty. The mean value (350 ± 11 kBq.m^{-3}) is indicated by the solid line whereas the dashed lines indicate the uncertainty. The measured values correspond well with the expected value as calculated from Eq. (2).

4. Discussion

The large fraction of the thoron that is pumped out compared to the exhalation from solid sources (e.g. Wang et al. in ref 10) makes our system as depicted in Fig. 1 a practical system with an uncertainty that can be made very small if required. In our set-up the uncertainty is mainly dependent on the accuracy of the flow rate and the extraction of the gamma intensities. As mentioned, after 60 h the nett counts in the 2614 peak is around 1300 counts per hour which implies an uncertainty of about 3% when the background subtraction is taken into account. This is more than adequate for the purpose of providing a reliable thoron source with an accuracy of a few percent. If lead shielding is added to reduce the background, this uncertainty could be reduced, but this would also complicate the system.

We experimented with several pumping speeds and various aerators. In all cases the system worked well when a correction is made for the pump speed. A faster pumping speed did not significantly increase the percentage of thoron that was removed from the solution and led to a lower concentration in the exit stream since a similar number of thoron nuclei was removed but these nuclei would now occupy a larger volume. Measurements showed that pumping for about 12 h is enough to fix the source strength for the thoron that is left in the bottle.

5. Conclusion

A simple, cost efficient thoron source based on air pumped through a solution of thorium nitrate in water has been developed. The simple and robust experimental set-up provides a thoron source with a high concentration and with good repeatability when compared to solid sources. The thoron that is removed from the solution by pumping is deduced from the activity of the thoron in the solution and the fraction

Table 3
Factors influencing the uncertainty based on equation (2).

Quantity	Value	Assigned standard uncertainty	Relative amount (%)
Fraction of Th removed	0.85	0.2 (based on a χ^2-change of 1 per degree of freedom in the fit shown in Fig. 5)	1
λ_{thoron}	0.01242	0.00006	
N -Number of radioactive nuclei in water.	331	5 based on the counting statistics in the HPGe. A systematic uncertainty for the HPGe and mass measurements of 10% is estimated.	32
Flow rate	0.6	0.1	67

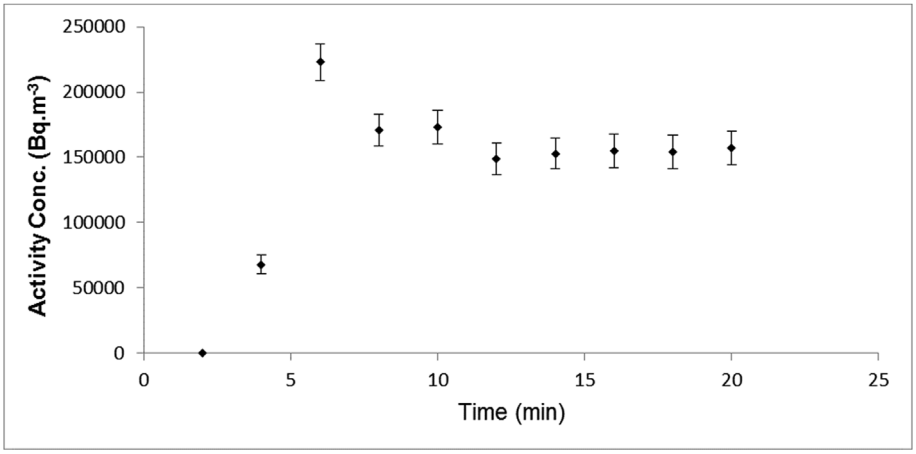

Fig. 7. Activity concentration as measured by the RAD7 during the initial few minutes of pumping for periods of 2 min. The pump was started at around t = 2 min.

of this thoron that decays in the solution by a straight forward gamma measurement of the thoron progeny.

Acknowledgements

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