A new thoron atmosphere reference measurement system

Benoit Sabot, Sylvie Pierre, Nathalie Michielsen, S. Bondiguel, Sylvain Bondiguel, Philippe Cassette

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B. Sabot[a,*], S. Pierre[a] , N. Michielsen [b], S. Bondiguel[b], P. Cassette[a]

[a] CEA, LIST, Laboratoire National Henri Becquerel (LNE-LNHB), Bât 602 PC111, CEA Saclay 91191 Gif-Sur-Yvette, Cedex, France

[b] Institut de Radioprotection et de Sûreté Nucléaire (IRSN), PSN-RES, SCA, LPMA, Centre de Saclay, Gif-sur-Yvette, 91192, France

Abstract

A new thoron reference (^{220}Rn) in air measurement system is developed at the LNE-LNHB with the collaboration of the IRSN. This measurement system is based on a reference volume with an alpha detector which is able to directly measure thoron and its decay products at atmospheric pressure. In order to improve the spectrum quality of the thoron progenies, we have applied an electric field to catch the decay products on the detector surface. The developed system is a portative device which can be used to measure reference thoron atmosphere such as the BACCARA chamber at IRSN (Picolo, 1999). As this system also allows the measurement of radon (^{222}Rn) in air, it was validated using the radon primary standards made at the LNE-LNHB. This thoron measurement system will be used, at IRSN, as a reference instrument in order to calibrate the thoron activity concentration in the BACCARA facility.

Keywords: thoron; Rn-220, Rn-222; radon; primary standard

[*] Corresponding author. Tel.: + 33 (0)1 69 08 41 04; fax: + 33 (0)1 69 08 26 19.

E-mail address: benoit.sabot@cea.fr, benoit.sabot@hotmail.fr, (B. Sabot)

1. Introduction

Thoron (^{220}Rn) is a decay product of ^{232}Th which is present in the Earth's crust. Due to its short half-life (55.8 s) (Bé et al., 2004) the thoron emanation from the ground is generally considered negligible. In France, the thoron is assumed to be 1/10 of the radon (^{222}Rn) concentration, but measurements in houses are rarely done. Recent measurements undertaken in Korea (Kim, 2005) and in Indian dwellings (Baciu, 2005) have shown that the thoron activity concentration may be similar or even higher than the radon concentration due to the building materials. More recently, a study done by Michielsen (2014) has shown that some commercial radon detectors results are affected by thoron. The interest on thoron measurement might also increase in the future if ^{232}Th is used in nuclear reactors.

Currently there are many commercial devices to measure radon and thoron concentrations in air. It is necessary to calibrate these detectors with a well-defined and traceable reference atmosphere (IEC, 2006), (IEC, 2009) (IEC, 2014). The LNE-LNHB has already a radon ($T_{1/2}$ = 3.8 days) primary standard system based on a solid angle method with a frozen radon source (Picolo, 1996). It has been upgraded in 2012 to produce radon standard with a relative standard uncertainty of 0.3 %. Radon standards are stored into vials and sent to other laboratories to calibrate detectors (such as IRSN with BACCARA). Unfortunately, due to the short half-life of thoron, it is not possible to use the same calibration method. Currently there is only one known thoron reference standard at the PTB (Physikalisch-Technische Bundesantalt) in Germany which is based on a gamma spectrometry of a thin ^{228}Th source to determine the thoron production (Röttger, 2010). This article presents a new thoron reference measurement system which is a portable device based on an alpha spectrometry of thoron and its decay products.

2. Measurement method

Thoron is a radioactive gas decaying through alpha transition to produce solid decay products which decay through alpha and beta transitions. From the thoron decay chain presented in figure 1, one can see the possibility to detect not only thoron gas using an alpha detector but also the decay products.

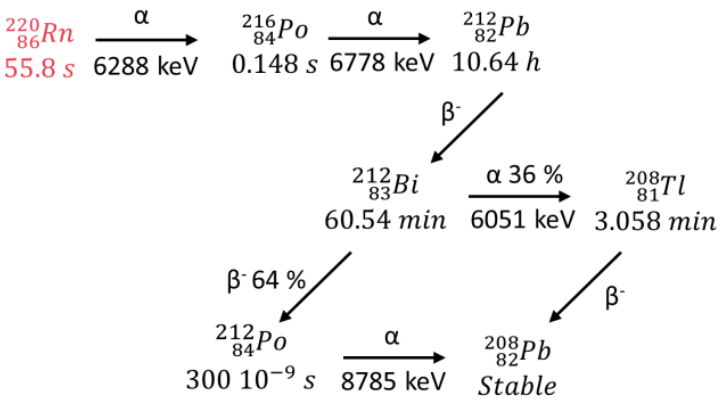


Fig. 1. Thoron decay chain with half-life and emitted particles, data are from Bé *et al.* (2004)

In order to improve the quality of the alpha spectra, it has been decided to use the electric properties of thoron decay products which are positively charged (Dua, 1981) to drive them onto the detector surface by applying an electric field, as shown on figure 2. However, many factors may reduce the amount of charged decay products such as air impurity (Porstendörfer, 2005), humidity (Porstendörfer, 1979) and the influence of these factors need to be addressed.

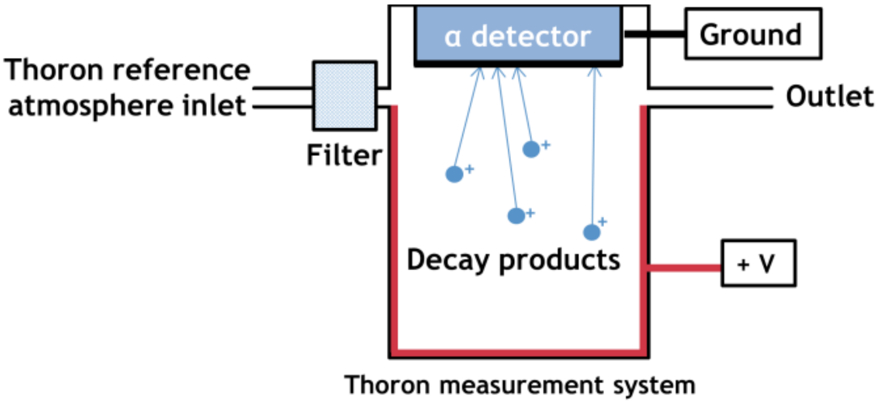


Fig. 2. Diagram of the thoron reference measurement system. The decay products are caught on the detector surface, whereas the gas fills the entire volume.

Different simulations were performed with Comsol Multiphysics (Comsol, 2015) and MCNPX (Pelowitz, 2011) to optimize the measurement system. The optimized properties are listed below.

1. The detection volume is a small cylinder of 1 cm high with 4 cm diameter.
2. A large surface silicon barrier detector is used as the top surface of the cylindrical volume to allow good gas detection efficiency.
3. The small size of the volume has been chosen to reduce the path of alpha particle emitted by the gas. The Monte Carlo simulations show that it prevents any detection efficiency variation due to pressure, humidity and temperature changes.
4. The detector surface is connected to the ground while the rest of the volume surface is a metallic electrode at the 2 kV potential. This induces a maximum electric field of 200 $kV.m^{-1}$ in the volume.
5. The electric field is strong enough to catch all the decay products produced in the volume chamber even if the air flow is 1 $L.min^{-1}$. From Comsol simulations, it was possible to deduce that the maximum time required to catch a particle with a single charge is 80 ms. In reality the decay products can have more than one charge, so they will be caught faster. This delay is much lower than the mean residence time of the thoron progenies inside the volume.
6. A filter is placed at the entrance of the detection volume in order to be sure that all the solid decay products produced outside of the volume are trapped.

As a result, this geometry can be used to detect both the alpha particles emitted by the gas and by the decay products. Since all the alpha particles that reach the detector are counted, and the detection volume is small, the counting efficiency can be calculated using its geometry and a Monte Carlo method. As radon (^{222}Rn) has the same properties than thoron, a radon standard can be used to validate the counting efficiency calculation results.

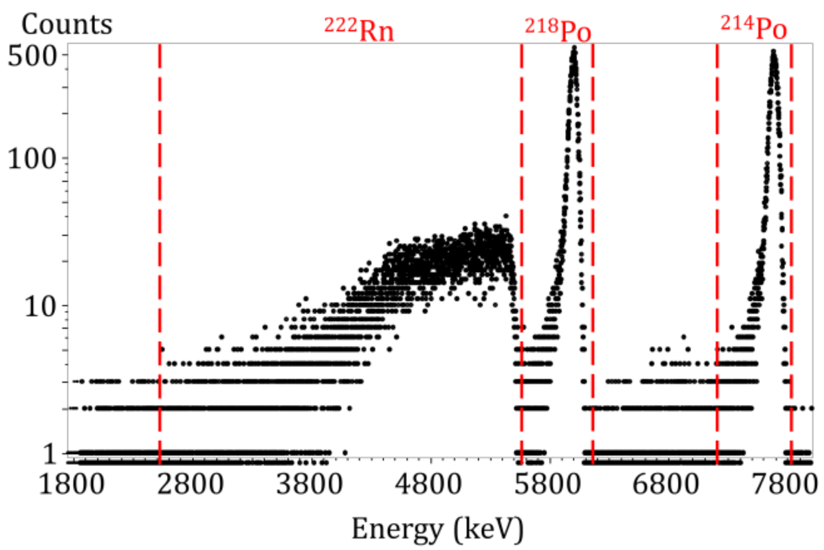
3. Measurement system validation using radon reference atmosphere

a. Experimental setup and spectra analysis

A gas dilution system has been built at the LNE-LNHB to produce reference atmosphere in terms of volume activity and to validate the thoron measurement system results. The volume of this system of 43.46 (5) L, allows the production of reference atmosphere using a primary radon standard mixed with dry and clean air. The activity concentration (Bq.m^{-3} STP, standard temperature and pressure) is known with a relative standard uncertainty of 0.3 %. The uncertainty on the radon atmosphere is the combined uncertainty of the radon primary standard activity (0.3 %) (upgraded version of the system from Picolo, 1996) and the relative uncertainty on the dilution volume (0.1 %). The radon background is negligible (less than 10^{-5} of the total activity).

With a gas humidifier it is possible to vary the relative humidity from 0 % RH to 90 % RH and the temperature and pressure are also controlled in the volume. To ensure a good mixing, two fans are placed inside the dilution volume. A pump is connected to this volume with a flow regulator to produce a closed circulation loop for the thoron measurement system. Using this closed loop it is possible to directly transfer the radon atmosphere in the thoron reference volume to validate the system efficiency calculation and also perform some variability tests.

The spectrum presented in figure 3 is obtained with the thoron measurement system for a reference radon atmosphere of 1 MBq.m^{-3} STP. Different Region Of Interest (ROI) are selected for counting the alpha decays of the ^{222}Rn, the ^{218}Po and the ^{214}Po. The same ROIs are used for each experimental result analysis.



4. **Fig. 3.** Spectra of radon atmosphere, measurement of 12000 s for an activity concentration of 1 $MBq.m^{-3}$. The selected regions of interest are those used for each measurement comparison.

a. Results obtained for a long time measurement of radon atmosphere

A radon atmosphere of 3.31 (1) $MBq.m^{-3}$ STP is created in the dilution volume. The thoron measurement system is connected to the reference atmosphere. A closed loop ensures the atmosphere circulation in the measurement system. The activity concentration is high enough to obtain high counting statistic within a short time measurement. The radon measurements in the new thoron reference system were performed during 3 days. There was a continuous monitoring (every 200 s) of every experimental parameters during the experiment. Their averages and standard deviations are presented in Table 1.

Table 1: Monitoring of experimental parameters during 3 days.

Experimental parameter studied	Average of the results	Standard deviation of the results	Absolute standard uncertainty on the calibration
Flow Rate ($L.min^{-1}$)	1.007	0.020	0.005
Pressure (hPa)	1013.64	0.86	0.04
Relative Humidity (% RH)	0.9	0.3	1.0
Temperature (°C)	20.99	0.30	0.04

Figure 4 shows each ROI counting results for the 60 first measurements. Each measurement is 2 000 s long and the counting are decay-corrected.

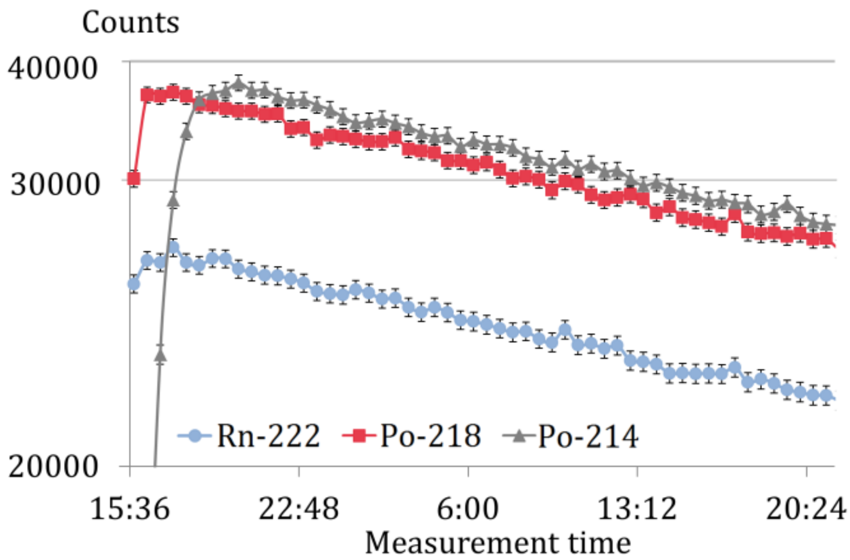


Fig. 4. Results of counting for the ^{222}Rn, ^{218}Po and ^{214}Po ROIs, the plotted results are the 60 first measurement of 2000 s for a reference atmosphere of 3.31 (2) $MBq.m^{-3}$ STP.

The uncertainty evaluation takes into account both the counting statistic and counting correction due to the radioactive decay during the measurement. The relative standard uncertainty is 0.6% for each measurement results.

The first measurements were done during radon mixing in the volume. After some time, radon becomes homogeneous in all the volume. The equilibrium between the ^{222}Rn and ^{218}Po is reached after 30 min and after 4 h with the ^{214}Po, which corresponds to the secular equilibrium of radon and its decay products. Since the activity is in a closed volume, radon and the decay products decay following the half-life of radon.

The observed variability of the results is compatible with the evaluated uncertainties.

b. Variation of pressure, temperature, humidity, flow rate and activity concentration

As previously described in § 2, this geometry has been chosen to reduce the influence of the experimental parameters. While the previous results have not shown any influence of these parameters, it is important to study their variation separately.

In the case of pressure, temperature and flow rate variation, different measurements are done for a same radon standard. In this specific case, once the reference atmosphere is created, one of the experimental parameter is changed and the measurements are repeated after 30 min of atmosphere circulation in the thoron measurement system. Using this experimental configuration, ^{218}Po and ^{222}Rn are at secular equilibrium and we are able to compare the results for each measurement.

For the activity and humidity variation, each measurement points are relative to different reference activity concentrations and the counting results are normalized to the reference standard activity for the comparison.

The results are presented in table 2. The only parameter of influence was the temperature which for both ^{218}Po and ^{222}Rn has an effect on counting. The reason for this variation is not clear yet and could be due to the warmup of the electronic. Note that Monte Carlo simulations do not reveal such influence.

Table 2: Influence of the experimental parameters on the ^{218}Po and ^{222}Rn counting.

Experimental parameter studied	Range of variation	Influence on the ^{222}Rn and ^{218}Po counting
Flow Rate	0 to 2 $L.min^{-1}$	None*
Pressure	900 to 1050 hPa	None*
Humidity	14 to 87 % RH	None*

Activity concentration	1 to 4 $MBq.m^{-3}$ STP	None*
Temperature	21 to 38 °C	0.18 % per °C

* None means that the standard deviation of the results is much lower than the relative standard uncertainty of the measurement 0.6 %.

c. Detection efficiency evaluation using experimental setup and comparison with MCNPX results

To evaluate ^{222}Rn detection efficiency it is necessary to remove the ^{218}Po and ^{214}Po alpha peak tail from ^{222}Rn peak. The counting from tails of ^{218}Po and ^{214}Po are estimated using Colegram software as shown in figure 5 (blue line). Once the counting rate for radon peak is evaluated, the detection efficiency is calculated using the radon reference activity concentration and the detection volume. This experimental determination of the detection efficiency is repeated eighteen times; the arithmetic mean is found to be 0.320 with a standard deviation of 0.002.

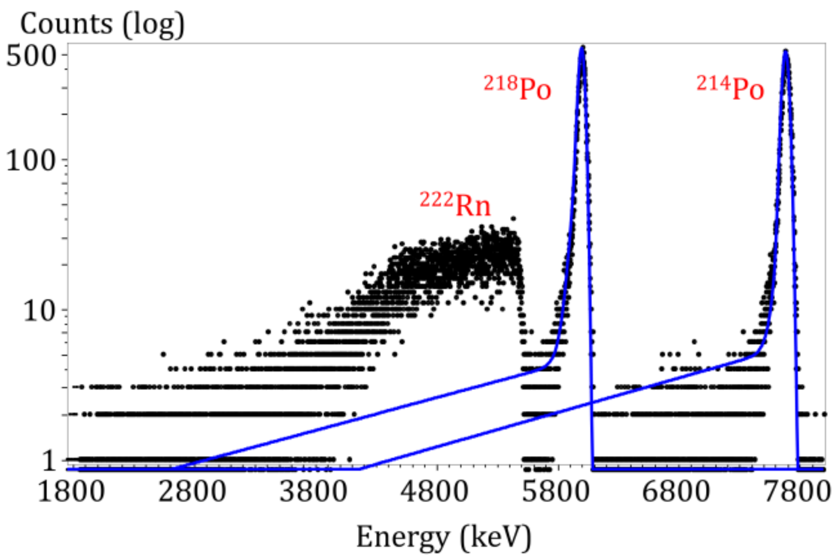


Fig. 5. Spectra of radon atmosphere with decay products fitting using the Colegram software.

On the other hand, the detection efficiency evaluated using MCNPX software is 0.3212 (3). This last uncertainty is mainly due to the volume measurement uncertainty since the statistical uncertainty and air density variation are negligible. As a result, the detection efficiency calculated with MCNPX is compatible with the experimental results within the evaluated uncertainties.

4. Measurement of thoron atmosphere

a. Experimental setup

Compared to ^{222}Rn, ^{220}Rn has a short half-life; for this reason, we use a ^{228}Th source with a constant air flow inside to produce a constant reference thoron atmosphere. For faster measurement we used a ^{220}Rn concentration of 1 $MBq.m^{-3}$STP. The reference thoron measurement system was connected to this atmosphere with a closed circulation loop. Figure 6 presents ^{220}Rn spectrum obtained during this experiment;

this spectrum is similar to the radon one. The wide peak is due to the thoron gas distributed in the whole volume and thinner peaks are due to the decay products caught on the detector surface.

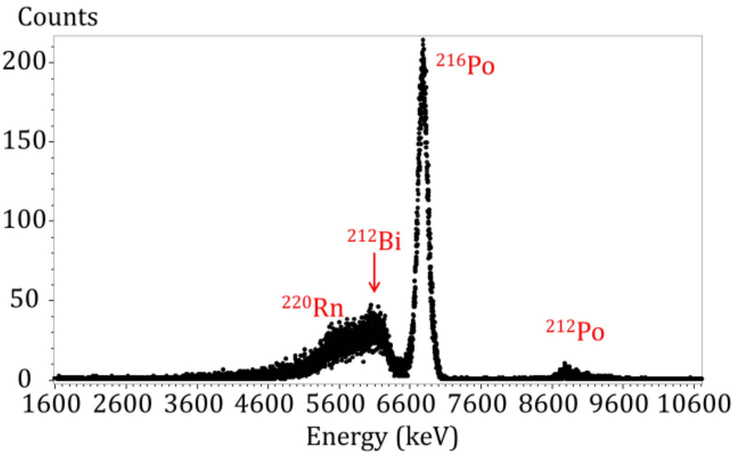
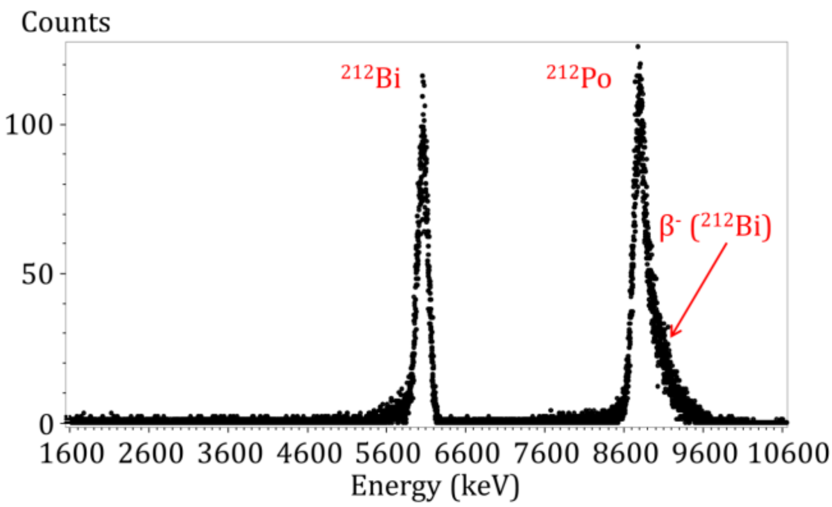


Fig. 6. Measurement of the volume activity of a constant thoron atmosphere using the new thoron measurement system.

Compared to radon, there is one difference however: ^{212}Bi peak interferes with the ^{220}Rn peak. To estimate the influence of the ^{212}Bi alpha activity, ^{212}Po peak can be used. As shown on figure 1, there is a direct secular equilibrium between ^{212}Bi and ^{212}Po. Using the decay scheme data (Bé et al., 2004) we can deduce the ratio of ^{212}Po activity and ^{212}Bi activity at the secular equilibrium $\frac{A\,(212\text{Po})}{A\ (212\text{Bi})} = 1.785(16)$.

Moreover, it is also possible to evaluate the ratio $\frac{A\,(212\text{Po})}{A\ (212\text{Bi})}$ directly with the system. After a long measurement time of thoron in the system, the detector is contaminated with ^{212}Pb which has a half-life of 10.64 h and produces ^{212}Bi. We can clean the volume with air or just close the measurement volume and wait 10 min to remove the thoron. Then, it is possible to measure only both ^{212}Bi and ^{212}Pb at secular equilibrium as shown in Figure 7. The result of this measurement is $\frac{A\,(212\text{Po})}{A\ (212\text{Bi})} = 1.753\ (17)$ which is compatible at k = 2 with the nuclides decay data.



5. **Fig. 7.** Measurement of ^{212}Bi and ^{212}Po deposited on the detector surface. The right tail of the ^{212}Po (T1/2 = 300 ns) peak is due to summing effects with the β particles emitted by ^{212}Bi since the electronic use a 2 µs shaping time.

a. Variation of the parameters of influence

The ^{222}Rn measurement does not present any variation due to the experimental parameter excepted for temperature as shown in §4.b. The same results are to be expected in the case of ^{220}Rn. However due to ^{220}Rn short half-life, it is possible that the flow rate has an influence on the measurement result.

Using the same thoron atmosphere, some measurements were done by changing the airflow inside the measurement volume. The results are presented in figure 8: we can clearly see that the counting rate of each ROI for ^{220}Rn and ^{216}Po increases as a function of the flow rate. It reaches a maximum at 1 L.min^{-1} which is the value selected for thoron measurement in the system. Since our flow rate can be regulated precisely with a relative standard uncertainty of 0.5 %, we can conclude that the flow variation will not have an impact on the measurement.

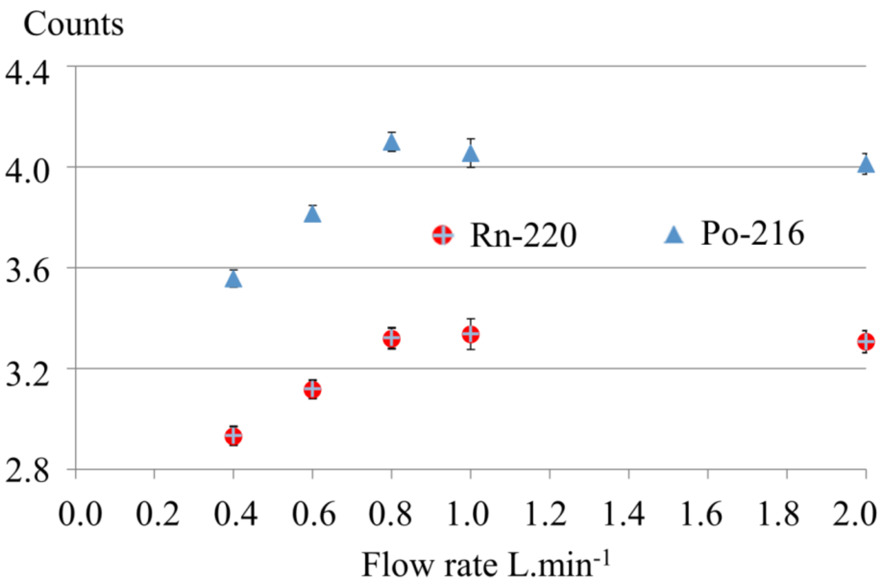


Fig. 8. Evolution of counting as a function of the flow rate in the measurement volume. The triangles correspond to ^{216}Po ROI, the circles correspond to ^{220}Rn window with ^{212}Bi removed by calculation, the crosses correspond to ^{220}Rn window with ^{212}Bi removed using experimental results.

The latest critical parameter in the measurement remains the thoron homogeneity in the volume. The simulations performed with Comsol Multiphysics (Comsol, 2015) show that thoron will be homogeneous in the volume for a flow rate of 1 L.min^{-1}. The mean residence time of thoron in the measurement volume is lower than 100 ms. However, the dynamic transfer function will be measured using helium as a tracer gas.

6. Conclusions

A portable thoron in air measurement system has been developed by the LNE-LNHB and the IRSN. This reference system has been validated using a primary radon standard. The geometrical efficiency of the system has been evaluated by calculation with a Monte Carlo method and by experiments with a radon standard; the obtained results are compatible. This geometrical efficiency can be directly used to evaluate thoron activity concentration from a reference atmosphere using direct thoron measurement for a flow rate in the volume higher than 0.8 $L.min^{-1}$. Due to the very low residence time in the measurement volume, the simulations results do not show any problem of homogeneity in the measurement volume, helium test are planned to confirm these results with experiments. As shown in this paper, the only parameter of influence is the variation of temperature while the other parameters do not have any significant effect.

The next step of the study will be the evaluation of the complete uncertainty budget of a thoron measurement; the main uncertainty factor of the method will be the counting statistics which can be reduced using long measurement time. A first test has been performed at the IRSN in a constant thoron atmosphere of 4 $kBq.m^{-3}$. With a one week measurement, counting statistic uncertainty can be negligible compared to the relative standard uncertainty of the detection efficiency (0.5 %). In this paper the results are only presented for the thoron peak but a similar study has been conducted also on ^{216}Po peak which will be used for thoron measurement in air with other uncertainty budget. The last planned step will be to perform a comparison with the ENEA (National Metrology Institute of Italy or radioactivity units) in the framework of the MetroNORM project.

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