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Methods for the experimental study of ^{220}Rn homogeneity in calibration chambers

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Manuscript Details

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Title	Methods for the experimental study of ^{220}Rn homogeneity in calibration chambers
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Abstract

This work presents two experimental methods for the evaluation of ^{220}Rn homogeneity in calibration chambers. The first method is based on LSC of the ^{220}Rn decay products captured in silica aerogel. The second method is based on application of solid state nuclear track detectors facing the air of the calibration chambers. The performances of the two methods are evaluated by dedicated experiments. The repeatability of the method, estimated as relative standard deviation of the LSC measurements of ten silica aerogel samplers exposed under the same conditions is found to be 1.6%. Both methods are applied to study thoron homogeneity in a 50 L empty AlphaGuard emanation and calibration container with its fan turned on, and it was found that the ^{220}Rn distribution is homogeneous within 10%. Both methods are also applied to test the thoron homogeneity in the BACCARA chamber at IRSN during a thoron calibration exercise. The results show that, at the centre of the chamber where the inputs of the sampling systems of the instruments were put close to each other, the thoron inhomogeneity is less than 10%. However, regions of higher thoron concentrations are clearly identified near the walls and the upper part of the chamber, with ^{220}Rn concentrations being up to 60% higher compared to the concentration at the reference point. These results highlight the importance of the control and assessment of thoron homogeneity in thoron calibrations and in the cases when radon monitors are checked for thoron influence.

Keywords	Thoron (^{220}Rn), Thoron calibration, Thoron homogeneity, LSC, nuclear track detectors
Manuscript category	Radiation Measurements
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Suggested reviewers	marco capogni, Luigi Tommasino

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Dear Editor,

Please find enclosed the revised version of our manuscript (Ref: ARI_2019_310) entitled: Methods for the experimental study of ^{220}Rn homogeneity in calibration chambers, by authors: K. Mitev, P. Cassette, D. Pressyanov, S. Georgiev, Ch. Dutsov, N. Michielsen, B. Sabot. This is the first revision of the manuscript.

I attach also a separate file, in which we have replied to all the comments and suggestions made by the reviewers. We would like to thank the reviewers for their comments and suggestions, which were very constructive and helped to improve the manuscript.

For all the correspondence regarding this manuscript, please do not hesitate to contact me as a corresponding author.

Sincerely yours,

Krasimir Mitev

/On behalf of all the co-authors/

Response to Comments & suggestions of Reviewer #1

A well written paper worthy of publication. I have made a few comments and suggestions for minor changes.

Thank you for this encouraging opinion.

Abstract

Repeatability of NTD ? - Our long-term experience with NTD of Kodak-Pathe LR-115/II indicates that the repeatability within a set of detectors exposed at identical conditions is within the Poisson track counting statistics.

Chapter 2. Evaluation of thoron homogeneity by LSC of silica aerogel

- The following logical rearrangement of test sessions is proposed – accepted, thank you.
- Ref to BIPM SI brochure. – corrected to 10 min, thank you.
- What do you mean? To enable LS cocktail to be fast absorbed in the aerogel for LSC measurement ? – That is exactly what was meant, Thank you.

“The silica aerogel was crushed and sieved (from ~0.5 mm to 3.0 mm) to get grains with suitable size – not too large so they can fit in the sampler without punctuating the closing filters at the same time not too small (e.g. dust) to enable LS cocktail to be fast absorbed in the aerogel for the LSC measurement after the exposure.”

- Results ? under which point of view ? which type among the three mentioned ?

Thank you for these questions, we have added some more comments in the text to elucidate how we judged the performance of the filters. In short, what we did is to compare the net LS counting rate, which is expected from the thoron activity in the sampler (available from Eq. 3) with the measured net LS counting rate. This way we found that the glass microfiber filters always give higher measured counting rates, which we interpret as inability to stop (filter) entirely the thoron progeny to penetrate in the sampler from the outside. The membrane filters (both types ADVANTEC and Millipore), however, showed good agreement of the net LS counting rates (when placed two of them on each side of the sampler), and that was our criterion to choose membrane filters.

- Usually the repeatability is given as standard deviation and not as maximum deviation. Furthermore, if you want to separate the variability components due to the thoron sampling and counting statistics, you should subtract (quadratically) the repeatability of the count per unit mass for a single sampler, that you can assume as a Poisson variable. So, I guess the real repeatability is lower than 2.3%.

Thank you for this suggestion, which we accepted and presented the repeatability as a relative standard deviation.

We agree with the referee that it is in principle possible to perform analysis of the different contributions to the overall variance. In this particular case we prefer to stay with the “overall” variance, since it is instructive what one can expect from the application of the method as a whole, including: careful preparation of the samplers, weighing, transfer of aerogel from the samplers to the cocktail, LSC measurement, etc.

Chapter 3. Evaluation of thoron homogeneity by SSNTDs

Please, add a reference. ; Please, add a reference - The corresponding references were added in the revised manuscript.

➤ What about the second one ??? - The second one is the calibration exercise at BACCARA.

Figure captions:

- Which filter type ? Millipore filters, that was added in the caption.
- What is the difference between the two vials? There is no difference between the vials (same vials, same aerogel, same LS cocktail), we simply made a snapshot of two of them.

Response to Comments & suggestions of Reviewer #2

Dear Authors,

You present results of a study of two different experimental methods to measure homogeneity of thoron concentrations in calibration chambers. I regard this as an important contribution to the quality assurance of thoron calibration and measurement. The quality of the first method that involves liquid scintillation counting seems to be high. However, I have a few comments regarding the presentation of the results, where I am not sure what is meant.

According to my understanding, the second method that makes use of solid-state nuclear track detectors seems to rely on several assumptions where I am not sure if they are justified. Please see my specific comments.

(Page numbers refer to the numbers of pages in the Word document, with the Introduction starting on page 3.)

page 3, "from thoron series": I suggest to write "decay series" or "decay chain" instead of "series" in order to clarify what is meant. - changed to thoron decay series, thank you.

page 3, Aerogel sampling in general: The aerogel acts as a sink to the thoron concentration (in contrast to the SSNTD and also in contrast to actively sampling measurement devices such as the AlphaGuard in pump mode if the outlet air goes back into the chamber). I guess that this effect is negligible. However, this should be assessed and mentioned. We think that each volume of the chamber, which is available for thoron, acts as a sink of the thoron concentration, as long as thoron decays happen everywhere. It is difficult to evaluate the degree to which a given volume in the chamber acts as a thoron sink, since this depends on the transport of the air through the volume, the air circulation in the chamber etc. However, given the high air circulation rate needed to ensure thoron homogeneity, we also think that the effect of the presence of the sampler on the thoron distribution in the chamber is negligible. In addition, the thoron activity distribution inside the sampler depends on the ratio of the thickness of the sampler to the diffusion length (Eqs 2 and 3). If $L/2L_D \ll 1$, then the thoron distribution in the sampler tends to uniform distribution. According to our experience, ratios $L/2L_D \ll 0.4$ are a good compromise, in this particular case of the samplers the ratio is $L/2L_D \approx 0.17$, which leads to a 1.4% deviation of the activity in the middle of the sampler compared to the activity on its surface. This is the maximal deviation and this result imply almost uniform thoron distribution inside the sampler, hence the sampler acts in the same way as the other air volumes in the chamber. This is also an argument for choosing thinner samplers with thickness 1 cm (see below).

page 5, "The best results were obtained": What does "best results" mean? What are the criteria to make a result a good one? And how was this tested? – we had the same question from Reviewer #1. Please see the reply to him, as well as the revised version of the manuscript in this part.

page 5, "Ultima Gold LLT LS cocktail": As far as I know there are LSC cocktails based on polar liquids ("water-based") and nonpolar liquids, where radon and thoron are retained to a higher degree in nonpolar cocktails. (For example, such nonpolar cocktails are used for measuring radon in water: The radon is transferred from the water to the cocktail because it is retained there much stronger.) What type of cocktail is the cocktail that you used? - We have tried both types of LS cocktails. Our non-polar cocktail was a home-made toluene-based (Toluene+PPO+Bis-MSB) cocktail, which is not miscible in water and which has excellent radon solubility. We have a long term experience with this cocktail for radon measurements, but in this case we observed something which looks like excitation of the cocktail (similar to chemi-luminescence) when it gets in contact with the aerogel (not shown in the manuscript). The Ultima Gold LLT cocktail, which is a commercially available polar cocktail and which contains surfactants, does not exhibit the drawback of the non-polar cocktail. It enters easily in the aerogel (the ultra sonic bath is necessary to remove the bubbles from the aerogel) and allows reliable LS counting. That is why we have chosen to use the polar cocktail UG LLT.

page 6, "The results are shown in Table 1 and indicate that the thinnest samples have highest net counting rate per unit mass.": Is this not obvious from Equations 2 and 3? The aerogel close to the surface gets the highest activity of thoron whereas the aerogel in the middle of the sampler gets the smallest activity. Therefore, thin packs of aerogel will have the highest specific activity (activity per mass). However, I think for the LSC measurement not the specific activity (or net counting rate per mass) but the total activity (or net counting rate) matters.

If we take a look at the equation which describes the thoron activity profile inside the sampler (Eq. 2), we see that $C(x)$ behaves like $\cosh(x)$, which states that the activity will be higher close to the surface and will tend to decrease in the middle of the sampler. Thus, the thinner the sample the higher the specific activity. We agree that what matters in the LSC is the total activity, thus larger volumes of aerogel would be beneficial as there is a tradeoff sensitivity vs. volume. However, we cannot make the volumes too large as we want to place the entire content of one sampler *easily* in one standard LS vial (which volume is 22 ml) and, for practical purposes, we do not want to fill the vial entirely with aerogel. That is why we believe that the optimal samplers are with 1 cm thickness.

Figure 3: It looks as if the aerogel gets translucent in the LSC cocktails. Is this true? I think that matters for the LSC measurement. If it was not translucent, decays of thoron and thoron progeny that stay in the aerogel, which is soaked with LSC cocktail, cannot be detected by the LSC counter. – Yes! The aerogel, when it is filled with LSC cocktail becomes translucent and the thoron progenies sucked in the aerogel are detected by the LSC counter.

Table 1: L is given in units of cm^3 . This is probably a typo and should read cm. – Yes, it was a typo and we corrected it to "L, cm". Thank you!

Table 2: I see that the uncertainty of each single measurement is larger than the deviation from the mean. I understand that the uncertainty of the mean is reduced by repeated or multiple measurements. However, in this case you compare single measurements with each other. Therefore, the uncertainty of the single measurements (e.g. 4.0% for AG 1) should be taken into account.

By the way, I would expect that measurement results with an uncertainty of that level should deviate from the mean more strongly, just because of the stochastic nature of uncertainty, even if the measurements are perfectly reproducible. The mean value of the relative uncertainties is 2.9%. I do not know if you applied a coverage factor (e.g. coverage factor $k=2$ in order to get an expanded uncertainty which covers approximately 95% of the results), but if the uncertainties are standard uncertainties, there should only be a not too big amount of the measurement

results within a range of the mean \pm the uncertainty (about 68% in case the values are normally distributed). Maybe the uncertainties are too big?

Yes, we have been conservative in the estimation of the uncertainties of the net counting rate at the end of the exposure. This is due to the experimental method we use to deduce the net counting rates of the samples at the end of the exposure. The uncertainty of the net counting rate at the end of the exposure has several components which contribute to it, please see our reply to the next comment. We are confident that our experimental approach to determine the net LS counting rate at the end of the thoron exposure $n_0(t=0)$ is unbiased and robust and the estimate of the uncertainties is adequate. All the reported uncertainties in the tables are standard uncertainties, i.e. coverage factor was not applied ($k=1$).

page 7, "The net counting rate of each of the samples was followed for 60 h": Is there a reason why you performed the measurements for such a long time? How did you get the count rate at the beginning? Did you calculate it from the total counts within this measurement time or did you read the count rate in several short intervals and used only the first one?

Our procedure for these measurements is as follows: we place all the LS samples in the LS analyzer and we perform series of consecutive measurements of all the samples with fixed counting time for each measurement (5 minutes in each case). We repeat the measurements of all the samples in a row for 60h. Thus, we obtain a series of measurements, and we fit the data $\ln(\text{net counting rate})$ vs. t with a straight line. Hence, we determine the net counting rate at the beginning ($n_0(t=0)$) for each sample as well as the effective half-life of the nuclides in the sample. This procedure is necessary and advantageous for two reasons: 1.) Technically we cannot measure the net counting rate at the end of the exposure ($t=0$) for all the samples simultaneously; 2) We have to wait at least 4 hours after the end of the thoron exposure in order to attain secular equilibrium between Pb-212 and its short lived progeny. The uncertainty of the net counting rate at $t=0$ includes the uncertainty components due to counting statistics, background correction as well as the component due to the fitting.

page 8, "in this case ^{220}Rn and its progeny atoms": Is there a reason why you do not mention Tl-208? I see that it does not emit alpha radiation but Pb-212 does not either (at least not this isotope itself). Tl-208 was mentioned in the list, thank you.

page 8, "Parallel measurements of the activity concentrations of ^{222}Rn ": The amount of deposited progeny depends on the surface to volume ratio of the chamber and on the unattached fraction of the progeny. A 1 m^3 chamber has a smaller surface to volume ratio than the 200 l spherical chamber; therefore a smaller amount of progeny might be deposited on the walls. A small unattached fraction (because of aerosol particles in the air inside the chamber) reduces the deposition velocity; this also leads to a smaller amount of deposited progeny.

Although surface to volume ratio of the empty 1 m^3 cylindrical BACCARA chamber is somewhat smaller (within 50%) than that of the 200 L spherical chamber, we do not consider this to change significantly the conditions assumed (almost all of the ^{220}Rn progeny: ^{212}Pb , ^{212}Bi , ^{212}Po , ^{208}Tl is deposited on the internal surface of the chamber) for the following reasons: (1) In the experiment described radon/thoron monitors were put in the BACCARA chamber, which increased the surface at which progeny atoms may deposit and decreased the free internal volume, thus increasing the surface to volume ratio; (2) Experiments at much larger than 1 m^3 volumes (e. g. rooms, as in the cited reference of Harley et al. (2010)) demonstrate extreme disequilibrium between ^{220}Rn and its progeny in the air. The following sentence in the text: "Results of other authors (Harley et. al., 2010) also show extreme disequilibrium in air between ^{220}Rn and its progeny, even for much larger volumes (e.g. rooms) than the volumes of 200 L or 1 m^3 " was amended to make this more clear.

page 8, "They register alpha particles of energy within 1.5 – 4.0 MeV and incident angle $<55^\circ$ ": I wonder if this

assumption is justified. I know that the energy and angular range of LR-115 is limited but according to my understanding that only means that within this range LR-115 are best suited as a measurement device whereas outside this range the sensitivity is smaller but not zero. I do not have access to the references literature Pressyanov 2012 but this publication

Mheemeed et al., Characterization of alpha-particle tracks in cellulose nitrate LR-115 detectors at various incident energies and angles. *Applied Radiation and Isotopes* 79 (2013) 48–55.

presents results that show that LR-115 is sensitive also to higher energies (in this publication presented up to 5 MeV), especially at large angles of incidence. I think the proposition that LR-115 is not sensitive to energies above 4 MeV should be justified more strongly.

The energy and angular window of the SSNTDs depends on the etching conditions and the mode of track counting. Essential for the application is the energy of alpha particles of ^{220}Rn progeny to be outside of this window, to avoid registration of alphas from the plate-out on the detector surface. This is valid as for the conditions we use, as well as for these reported by Mheemeed et al. (2013). The corresponding paragraph in the text was revised, to make this more clear.

Highlights paper 78 Mitev et al.:

- Two new methods for checking Thoron homogeneity in chambers are suggested.
- The first method is based on trapping of Thoron progeny in aerogel and LSC.
- The second method is based on solid state nuclear track detectors.
- Both methods studied in laboratory experiments and found feasible.
- Both methods are applied in the BACCARA 2018 Thoron calibration exercise.

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4 **Methods for the experimental study of ^{220}Rn homogeneity in**
5 **calibration chambers**
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21 Roses, France.
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26
27 **Abstract**
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29 This work presents two experimental methods for the evaluation of ^{220}Rn homogeneity
30 in calibration chambers. The first method is based on LSC of the ^{220}Rn decay products
31 captured in silica aerogel. The second method is based on application of solid state
32 nuclear track detectors facing the air of the calibration chambers. The performances of
33 the two methods are evaluated by dedicated experiments. The repeatability of the
34 method, estimated as relative standard deviation of the LSC measurements of ten silica
35 aerogel samplers exposed under the same conditions is found to be 1.6%. Both methods
36 are applied to study thoron homogeneity in a 50 L empty AlphaGuard emanation and
37 calibration container with its fan turned on, and it was found that the ^{220}Rn distribution
38 is homogeneous within 10%. Both methods are also applied to test the thoron
39 homogeneity in the BACCARA chamber at IRSN during a thoron calibration exercise.
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62 The results show that, at the centre of the chamber where the inputs of the sampling
63 systems of the instruments were put close to each other, the thoron inhomogeneity is
64 less than 10%. However, regions of higher thoron concentrations are clearly identified
65 near the walls and the upper part of the chamber, with ^{220}Rn concentrations being up to
66 60% higher compared to the concentration at the reference point. These results highlight
67 the importance of the control and assessment of thoron homogeneity in thoron
68 calibrations and in the cases when radon monitors are checked for thoron influence.
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73 **Keywords:** Thoron (^{220}Rn), Thoron calibration, Thoron homogeneity, LSC, nuclear
74 track detectors.
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121 **1. Introduction**
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123 Thoron (^{220}Rn) is an isotope of the noble gas radon with 55.8 s half-life. Studies in
124 the last decade demonstrate that doses from thoron decay series cannot be considered
125 negligible and there is a need for improvement of passive methods to measure thoron
126 progeny and of the associated metrological assurance (McLaughlin, 2010; Hosoda et
127 al., 2017). The quality assurance of thoron measuring instruments and the studies of the
128 influence of ^{220}Rn on the radon measurement devices are areas of active research (Sabot
129 et. al., 2016; Röttger et. al., 2009; Röttger et. al., 2010; Röttger et. al., 2014; Tokonami,
130 2005; He et. al., 2017; Michielsen and Bondiguel, 2015). However, the short half-life
131 of ^{220}Rn makes it difficult to ensure that it is homogeneously distributed in the chamber
132 volume when thoron exposures are performed. Therefore, experimental methods able
133 to probe thoron homogeneity are highly necessary.
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147 The objective of this work is to present two newly proposed methods for evaluation
148 of thoron homogeneity and their application in the thoron calibration exercise that has
149 been carried out at IRSN in the framework of the MetroRADON Euramet EMPIR joint
150 research project. The first method is based on a capture of thoron decay products in
151 silica aerogel grains and subsequent liquid scintillation counting (LSC) of the silica
152 aerogel. The second method is based on the measurement of the density of tracks
153 formed by ^{220}Rn and ^{216}Po in Kodak Pathe LR-115/II solid state nuclear track detectors
154 (SSNTDs). The two methods are applied successfully for the evaluation of ^{220}Rn
155 homogeneity in small (50 L) calibration vessels as well as in the BACCARA chamber
156 (1 m^3) at IRSN with seven ^{220}Rn measuring instruments inside.
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170 **2. Evaluation of thoron homogeneity by LSC of silica aerogel**
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 180 This method makes use of a silica aerogel as thoron sampler and its subsequent
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 182 mixing with a LS cocktail for LSC counting. The idea of the sampler is to allow thoron
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 184 to enter freely from the environmental air into the cylindrical volume through the filters
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 186 and to stop the thoron decay products on the filters. Thus, when ^{220}Rn decays inside the
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 188 sampler, its decay products (^{216}Po , ^{212}Pb , ^{212}Bi , ^{212}Po and ^{208}Tl) will attach to the silica
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 190 aerogel and their activity in the silica aerogel will be proportional to the ^{220}Rn activity
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 192 that has entered in the cylinder. The latter is proportional to the ambient ^{220}Rn activity
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 194 concentration in the air surrounding the sampler. The usage of silica aerogel provides a
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 196 large / sufficient amount of free space in the sampler for thoron to diffuse in and at the
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 198 same time allows the effective capture of its decay products.
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201 The ^{220}Rn transport inside the sampler (assumed to have a cylindrical geometry)
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 203 can be described by the diffusion equation with a term accounting for radioactive decay.
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 205 As the thoron half-life is 55.8 s, for constant thoron concentration and exposures longer
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 207 than 10 min, a steady state diffusion can be assumed:
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$$210 \quad D \frac{\partial^2 C}{\partial x^2} - \lambda C = \frac{\partial C}{\partial t}, \text{ with } \frac{\partial C}{\partial t} = 0 \quad (1)$$

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 212 where C is the ^{220}Rn activity concentration, D is the diffusion coefficient of ^{220}Rn in the
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 214 material, λ is the ^{220}Rn decay constant, x is the space coordinate along the cylinder axis
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 216 and t is the time variable. The solution of the above equation in plate parallel geometry
 217
 218 along the axis of the sampler is:
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$$221 \quad C(x) = C_{out} \frac{\cosh\left(\frac{2x-L}{2L_D}\right)}{\cosh\left(\frac{L}{2L_D}\right)} \quad (2)$$

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 223 and the ^{220}Rn activity in the sampler (A_{Tn}) is given by
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$$226 \quad A_{Tn} = 2C_{out}V \frac{L_D}{L} \tanh\left(\frac{L}{2L_D}\right), \quad (3)$$

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239 where C_{out} is the outside ^{220}Rn activity concentration (assuming that the filters are
240 transparent to thoron), L and V are the height and the volume of the cylinder and
241 $L_D=(D/\lambda)^{1/2}$ is the diffusion length of ^{220}Rn in the material (air or silica aerogel). Taking
242 into account that the diffusion coefficient of ^{220}Rn in air is $D=10^{-5}$ m²/s (Ishimori et. al.,
243 2013) which gives $L_D=3.0$ cm, from Eq. 2 it follows that one can expect non-
244 homogeneous thoron distribution inside the sampler. Thus, it is important, as shown
245 hereafter, to choose carefully the thickness of the samplers to ensure best performance.
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249 Specially designed thoron samplers were developed as shown in Fig 1. The sampler
250 consists of a cylindrical body and two end caps, which serve to fix and support two air
251 filters (bottom and top) at the ends of the cylinder (Fig. 1a). The inner and the outer
252 diameters of the cylinder are 4.4 cm and 4.7 cm, respectively. The diameters were
253 chosen to fit the diameter of the filters ($d=4.7$ cm). The silica aerogel was crushed and
254 sieved (from ~0.5 mm to 3.0 mm) to get grains with suitable size – small enough so
255 they can fit in the sampler without punctuating the closing filters and, at the same time,
256 large enough (e.g. larger than dust particles) to allow the aerogel to absorb the LS
257 cocktail quickly in order to perform the LSC measurement as soon as possible after the
258 exposure. The silica aerogel is placed in the cylinder (Fig. 1b) and the sampler is closed
259 tight (Fig. 1c). All plastic parts of the thoron sampler are locally made with a 3D printer.
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278 In order to evaluate the thoron homogeneity during the calibration exposures the
279 thoron samplers can be positioned at any point of interest in the calibration chamber.
280 After the end of the thoron exposure, the samplers are removed and the silica aerogel
281 from each sampler is carefully transferred in a high performance LS glass vial. The
282 mass of the transferred silica aerogel is determined by weighing the LS vials. The vials
283 are then filled with 15 mL Ultima Gold LLT LS cocktail and placed for 10 min in an
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298 ultrasonic bath in order to facilitate the full penetration of the scintillation cocktail in
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300 the silica aerogel and to remove air bubbles from it (Fig. 3).
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302 Different types of filters were tested - glass microfiber filter with an equivalent
303 pore size of 1.2 μm and thickness 260 μm (LLG-Labware 9045867), mixed cellulose
304 ester membrane filter with an equivalent pore size of 0.2 μm and thickness 130 μm
305 (ADVANTEC A20A047A) and mixed cellulose ester membrane filter with 0.3 μm
306 equivalent pore size and thickness 150 μm (Millipore PHWP04700). The performance
307 criterion for the choice of the filter is the agreement between the measured net LS
308 counting rate and the expected net LS counting rate from the activity, absorbed in the
309 aerogel, which is determined from Eq. 3.
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319 The results from the tests showed that net LS counting rate obtained with the glass
320 microfiber filters is always higher than the net LS counting rate expected from the
321 absorbed activity (Eq. 3). Therefore, we conclude that the used glass microfiber filters
322 do not completely stop the thoron progeny from penetrating in the samplers. The best
323 agreement between the measured and the expected net LS counting rate was obtained
324 with two membrane filters placed on each entry of the sampler. Both types of filters
325 ADVANTEC A20A047A and Millipore PHWP04700 show excellent performance.
326 Scanning electron microscope images of the used membrane filters are shown in Fig.
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337 2.

338 In order to choose the optimal thickness, three different samplers were produced
339 with $L=1.0$ cm, 1.5 cm and 2.0 cm. Six samplers (two of each thickness) were exposed
340 to thoron in a 50 L AlphaGuard calibration container with an AlphaGuard PQ2000 PRO
341 (Rn/Tn) reference monitor placed inside. The ^{220}Rn activity concentration during the
342 exposure was $C_{out}=612(61)$ kBq/m³ and the exposure duration was 68.2 h. The samplers
343 were placed at the bottom of the vessel close to each other with a special focus on ^{220}Rn
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357 freely reaching each sampler and passing through the filters. After the exposure the
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359 silica aerogel from each sampler was transferred in a high performance glass vial and
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361 the vials were measured on a RackBeta 1219 LS counter (Wallac, Finland). The results
362
363 are shown in Table 1 and indicate that the thinnest samples have the highest net
364
365 counting rate per unit mass. Considering the results in Table 1 and noting that the net
366
367 LS counting rate is due to several thoron progenies (^{212}Pb , ^{212}Bi , ^{212}Po and ^{208}Tl) it can
368
369 be concluded that samplers with thickness $L=1$ cm which contain around 5 g of silica
370
371 aerogel provide sufficiently good sensitivity of the method to be applied for the
372
373 evaluation of ^{220}Rn homogeneity.
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377 The most important characteristic of the proposed method from the point of view of
378
379 practical applications is its repeatability. It is studied in this work with the experimental
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381 set-up shown schematically in Fig. 4 and on the photo in Fig. 5. The set-up consists of
382
383 a powerful fan (gas-flow $2.5 \text{ m}^3/\text{min}$) mounted to a tube. Inside the tube there are 10
384
385 thoron samplers divided in two groups (AG1 to AG5 mounted closer to the fan and
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387 AG6 to AG10 just behind them, see Fig. 4a and Fig 5a). The samplers are positioned
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389 with their filter-sides parallel to the air-flow (see Fig. 4b and Fig 5b). The entire system
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391 is placed in a 50 L calibration container (Fig. 6) with one thoron sampler placed at the
392
393 exit of the tube perpendicular to the gas-flow (AG11) and another sampler placed
394
395 outside beside the tube (AG12), see Fig. 4a and Fig. 6. The thoron inlet is positioned
396
397 right in front of the fan and the container is closed hermetically. The gas-flow of the
398
399 fan ($150 \text{ m}^3/\text{h}$) is chosen large enough to guarantee that all the samplers in the tube
400
401 (AG1-AG10) are exposed to the same thoron concentration and the volume refresh rate
402
403 (50 times per min) is sufficient to assume that AG11 and AG12 are also exposed to the
404
405 same concentration. The duration of the ^{220}Rn exposure was 66 h and the thoron activity
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407 concentration in the container was $474(47) \text{ kBq}/\text{m}^3$. After the exposure, the silica
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416 aerogel from the samplers was transferred and measured on the RackBeta 1219 LS
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418 counter as described above. The net counting rate of each sample was followed for 60
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420 h and the net counting rates at the moment of the end of exposure are evaluated. The
421
422 results, presented in Table 2, show that the relative standard deviation of the net
423
424 counting rates per unit mass of the samplers in the tube (AG1-AG10) is 1.6% and their
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426 variations are fully within the estimated uncertainties. Moreover, the sampler AG11,
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428 which is in front of the tube and is perpendicular to the air stream also agrees well with
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430 the mean value within its estimated uncertainty. The same is also true for the sampler
431
432 AG12, which is behind the tube (see Fig. 4a and Fig. 6). These results suggest a
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434 repeatability of the method of the order of 1.6%, which is an excellent repeatability for
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436 the evaluation of thoron homogeneity in ^{220}Rn calibrations.
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439 440 441 **3. Evaluation of thoron homogeneity by SSNTDs**

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444 The other approach we investigated to evaluate the homogeneity of ^{220}Rn in
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446 chambers is based on the use of bare SSNTDs, placed at different points inside the
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448 chamber. These detectors register alpha particles that reach the detector surface with
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450 energy and incident angle within certain registration window specific for each type of
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452 SSNTDs. Normally, the air contains a mixture of isotopes (in this case ^{220}Rn and its
453
454 progeny atoms ^{216}Po , ^{212}Pb , $^{212}\text{Bi}+^{212}\text{Po}/^{208}\text{Tl}$, ^{212}Po is always in equilibrium with ^{212}Bi).
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456 However, in an exposure chamber volume a substantial part of the progeny atoms is
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458 deposited on the internal walls (George et al., 1983). The deposited fraction is higher
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460 when a fan creates air turbulence inside the chamber (Cheng et al., 1990) (the described
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462 experiments were made in this mode). Parallel measurements (Pressyanov, 2002) of the
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464 activity concentrations of ^{222}Rn and its progeny in 200 L spherical volume with a fan
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466 operating inside showed that the deposited fractions are 94.0%, 99.7% and 99.9% for
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475 ^{218}Po (half-life 3.05 min), ^{214}Pb (half-life 26.8 min) and ^{214}Bi (half-life 19.9 min),
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477 respectively. When ^{220}Rn + progeny is created in the chamber, due to the longer half-
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479 life of ^{212}Pb (half-life 10.64 h) and ^{212}Bi (half-life 60.55 min) one can expect that
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481 practically all of the ^{212}Pb and $^{212}\text{Bi}+^{212}\text{Po}$ atoms are deposited on the walls and their
482
483 air fraction is negligible. Results of other authors (Harley et. al., 2010) also show
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485 extreme disequilibrium in air between ^{220}Rn and its decay products ^{212}Pb and ^{212}Bi ,
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487 even for much larger volumes (e.g. rooms) than the volumes of 200 L or 1 m³.
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489 Therefore, within the present approach, we assume that the isotopes in the air are ^{220}Rn
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491 and ^{216}Po and that ^{216}Po , due to its short half-life of 0.15 s, it has of the same volume
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493 distribution as ^{220}Rn .
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497 In our experiments SSNTDs of Kodak-Pathe LR-115 type II were used. They
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499 register alpha particles within an energy and angular window of registration that
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501 depends on the etching conditions and the mode of counting. These conditions in our
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503 case were etching with 10% NaOH at 60 °C for 100 min, washing with water for 30
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505 min and washing for 2 min in still 50% ethanol and visual counting by microscope -
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507 only of tracks that created holes through the 12 μm sensitive layer of this type of
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509 detectors were counted. As described elsewhere (Pressyanov, 2012), under these
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511 conditions the detectors register alpha particles of energy within 1.5 – 4.0 MeV and
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513 incident angle <55° to the normal. The air volumes from which the alpha particles of
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515 different isotopes can be detected are schematically shown in Fig.7. What is essential
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517 for this application is that the alpha particles from the ^{220}Rn progeny atoms deposited
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519 on the detector surface cannot be detected, as their energy is well above the upper
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521 energy threshold of the window for registration. From Fig.7 it follows that, to avoid
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523 contribution from the atoms deposited on the chamber internal surface, the SSNTD face
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525 should look to air being at a distance of at least 8 cm from any surface – i.e. outside the
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534 range of ^{212}Po alphas. Under these conditions the tracks will be due only to the alpha
535 particles from the sources in the air (^{220}Rn and ^{216}Po) and the registered tracks will
536 originate from the isotopes that are in a small volume within a distance of 2.4 – 5.0 cm
537 from the detector surface (“detection volumes”, see Fig. 7).
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542
543 Respecting the above rule, Kodak-Pathe LR-115/II detectors were used in two
544 experiments. The first was in 50 L cylindrical exposure chamber in which 33 pieces of
545 detectors were placed. The map of the “detection volumes” is shown in Fig. 8. The
546 exposure was made at the laboratory facility, described elsewhere (Pressyanov et. al.,
547 2017). The exposure was made at an average ^{220}Rn concentration of 800(50) kBq m⁻³
548 for 220 min. During exposure the ^{220}Rn concentrations were followed by a reference
549 instrument AlphaGUARD PQ2000 PRO (Rn/Tn). The analysis of the results showed
550 that the distribution of ^{220}Rn in the air is homogeneous within 10%. The variations
551 between the results can be explained by the detector reading uncertainty and sub-
552 volumes of significantly higher or lower concentrations cannot be identified. Similar
553 homogeneity (i.e. within 10%) of the empty 50 L AlphaGuard calibration volume with
554 its own fan turned on is obtained by the LS counting of the silica aerogel (not shown).
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570 **4. Application of the methods during the thoron calibration exercise, performed** 571 **at BACCARA chamber at IRSN** 572

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574 A thoron calibration exercise was carried out in May 2018 in the framework of the
575 MetroRADON Euramet project using the IRSN reference radon chamber called
576 BACCARA. The BACCARA chamber is a 1 m³ stainless steel chamber with a thoron
577 reference instrument attached to it (Sabot et. al., 2016), which is used to create reference
578 radon and thoron atmospheres for calibration of radon and thoron measuring devices.
579 During the thoron calibration exercise, four AlphaGuards and three RAD7 instruments
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593 were placed in the chamber and their thoron measurement performance was checked
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595 against the reference instrument (Fig. 9). The thoron calibration exercise and its results
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597 will be described in detail elsewhere. In order to test the thoron homogeneity in the
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599 BACCARA chamber during the calibration exercise 12 silica aerogel thoron samplers
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601 and 22 pieces of SSNTDs were placed at different positions in the chamber (Fig. 9).
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603 The SSNTDs were placed in all parts of BACCARA internal volume, but respecting
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605 the stated above rule for detector position. The thoron exposure duration was 48 h and
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607 the ^{220}Rn activity concentration in the chamber was 46 kBq/m^3 . In order to avoid the
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609 effect of possible ^{220}Rn inhomogeneity during the calibration, the inputs of the
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611 instruments sampling systems were put close to each other as much as possible to the
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613 extent of forming a sampling point in the chamber (Fig. 10). The silica aerogel samples
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615 and the SSNTDs were placed around the sampling and all other parts of the chamber,
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617 trying to cover the upper part of the chamber (Figures 11 and 12) as well as the space
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619 around the sampling points and around and between the detectors (Fig. 13).
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623 After the end of the exposure, the silica aerogel samples were treated as described
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625 above. The activity in the samples is measured on a Wallac Guardian LS counter at
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627 LNHB and the results are shown in Table 3. For better visualization, the values obtained
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629 with the different samplers are shown relative to the reference point in Figures 11-13.
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631 The results in Table 3 and Fig. 11 show that the differences between the ^{220}Rn
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633 concentrations in positions “C”, near the center of the chamber, close to the
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635 instruments’ sampling points, do not exceed 10%. But, one can observe higher
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637 differences for positions “W”, far from the center of the chamber, for example the ^{220}Rn
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639 concentrations near the upper wall of the chamber (AT7) are up to 61% higher than
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641 those in the center (Fig. 11). Similarly, the thoron concentration near the walls of the
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643 chamber are higher than those at the reference point (see, for example AT4, AT5, AT11
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652 and AT7 in Fig. 11 and 12). The ^{220}Rn concentration below the instruments (AT8) are
653 also higher than those in the reference point (Fig. 13). Overall, it seems that the ^{220}Rn
654 concentration near the sampling point of the instruments is homogeneous, but there are
655 volumes in the chamber near the walls, the entry point of the thoron and in front of the
656 fan where much higher ^{220}Rn concentrations are observed.
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663 The SSNTD results reveal signs of inhomogeneity with up to about 60% deviation
664 from the reference point (Fig. 14). It should be noted also that the silica aerogel
665 sampling method and the SSNTD method give consistent results for the points where
666 the two methods can be applied (see Fig. 12). For example, the detection volume of the
667 highest deviation ratio 1.61(0.18) corresponds to SSNTD No 4 and it is close to the
668 space where LSC of silica aerogel (AT5) also showed high deviation (ratio of 1.55).
669 Similarly, SSNTD No 7 and AT11, which are close at the back of the chamber, give
670 very close results (Fig. 12). The two methods clearly demonstrate that there can be high
671 thoron inhomogeneity near the walls of the chamber.
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683 684 **5. Conclusions**

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686 Two experimental methods for evaluation of ^{220}Rn homogeneity in calibration
687 chambers are presented. The first method is based on LSC of thoron short-lived decay
688 products captured in silica aerogel and the second method is based on application of
689 SSNTDs. The performance of the two methods is evaluated in several dedicated
690 experiments where it is shown that the repeatability of the method based on LSC of
691 silica aerogel is within 1.6%. Both methods are applied to study thoron homogeneity in
692 a 50 L empty AlphaGuard emanation and calibration container with its fan turned on,
693 and it was found that thoron distribution is homogeneous within 10%.
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711 Both methods are applied successfully to test the thoron homogeneity in the
712 BACCARA chamber during the thoron calibration exercise that was carried out at
713 IRSN, France in the framework of the MetroRADON Euramet EMPIR project. The
714 results show that, at the centre of the chamber, where the inputs of the instruments
715 sampling systems were put close to each other, the thoron inhomogeneity is less than
716 10%. However, regions of higher thoron concentrations are clearly identified near the
717 walls and the upper part of BACCARA, with ^{220}Rn concentrations being up to 60%
718 higher compared to the concentration at the reference point. These results highlight the
719 importance of the control and assessment of thoron homogeneity in thoron calibrations
720 and in the case when radon monitors are checked for thoron influence. The assessment
721 of ^{220}Rn homogeneity will be particularly important in the case of checking passive
722 radon monitors for thoron influence. That is because, contrary to the case of active
723 monitors, there are no inputs of the passive detectors, so they cannot be put together
724 near a common sampling point in the calibration chamber. Thus, an experimental
725 method for checking thoron homogeneity as those described in this work will be highly
726 necessary.
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888 **Figure captions:**
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892 **Fig. 1.** A photograph of a thoron sampler. a) – Empty sampler, with bottom air filter,
893 ready to be filled with silica aerogel and the top end cap with top air filter
894 mounted; b) the sampler filled with silica aerogel; c) closed sampler ready to
895 be placed in thoron chamber.
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900 **Fig. 2.** Scanning electron microscope images of the Millipore membrane filters used
901 for the samplers. a) and b) –surface view, c) and d) – cross-sectional view.
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904 **Fig. 3.** High performance LS glass vials (20 ml) filled with silica aerogel and Ultima
905 Gold LLT LS cocktail. The photograph is taken after the vials had been placed
906 for 10 min in a ultrasonic bath. When filled with LSC cocktail, the aerogel
907 becomes translucent.
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912 **Fig. 4.** Schematic view of the geometry used to test the repeatability of the silica
913 aerogel method. a) Schematic view of the exposure vessel, the ^{220}Rn inlet, the
914 fan and the tube with the thoron samplers; b) Illustration of the arrangement
915 of the thoron samplers in the tube.
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922 **Fig. 5.** Photographs of the experimental arrangement used to test the repeatability of
923 the silica aerogel method: the fan, the tube and the thoron samplers can be
924 seen in the picture.
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928 **Fig. 6.** Photograph of the exposure set-up used to test the repeatability of the silica
929 aerogel method. The fan and the tube with the samplers are placed in a 50 L
930 vessel. Additional samplers are placed in front and near the tube.
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934 **Fig. 7.** Air-volumes from which alpha particles can be detected by LR-115/II
935 SSNTD. The volumes are shaped taking into account the fact that energy
936 window for track registration get narrow when the incident angle is increasing.
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947 **Fig. 8.** The picture shows the spots in the chamber volume from where activity can
948 be detected by the placed in a grid Kodak Pathe LR-115/II SSNTD
949
950 be detected by the placed in a grid Kodak Pathe LR-115/II SSNTD
951 (“detection” volumes).
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954 **Fig. 9.** A photograph of the BACCARA chamber showing the experimental set-up
955 used in the thoron calibration exercise. The thoron inlet nozzle and the fan
956 used to homogenize the air in the chamber are indicated as well as the
957 positions of some of the thoron samplers and the SSNTDs.
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962 **Fig. 10.** Photograph showing the sampling point, where the inlets of the thoron
963 measuring instruments are located. There are thoron samplers (AT1, AT2 and
964 AT6) and a SSNTD (No 16) around the sampling point. The percent in the
965 boxes with the sampler number indicate the sampler’s readings relative
966 deviation with respect to the reference (AT1).
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973 **Fig. 11.** Position and relative deviation with respect to the reference position of the
974 readings of the thoron samplers in the upper half of the BACCARA.
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977 **Fig. 12.** Position and relative deviation with respect to the reference position of the
978 readings of the thoron samplers in the upper half of the BACCARA and near
979 the sampling point.
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983 **Fig. 13.** Position and relative deviation with respect to the reference position of the
984 readings of the thoron samplers located below and between the ^{220}Rn
985 measuring instruments.
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990 **Fig. 14.** The ratio of the SSNTD signal (net track density) to the signal of the detector
991 that is at closest distance to the reference point (detector No 16).
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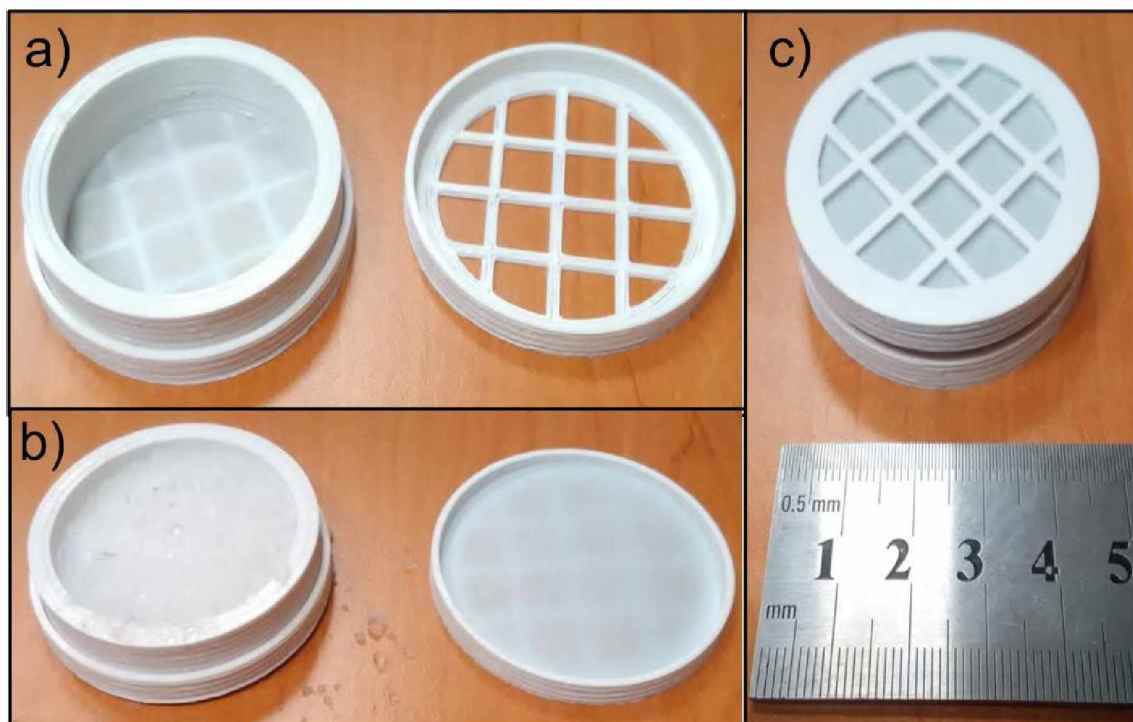


Fig. 1

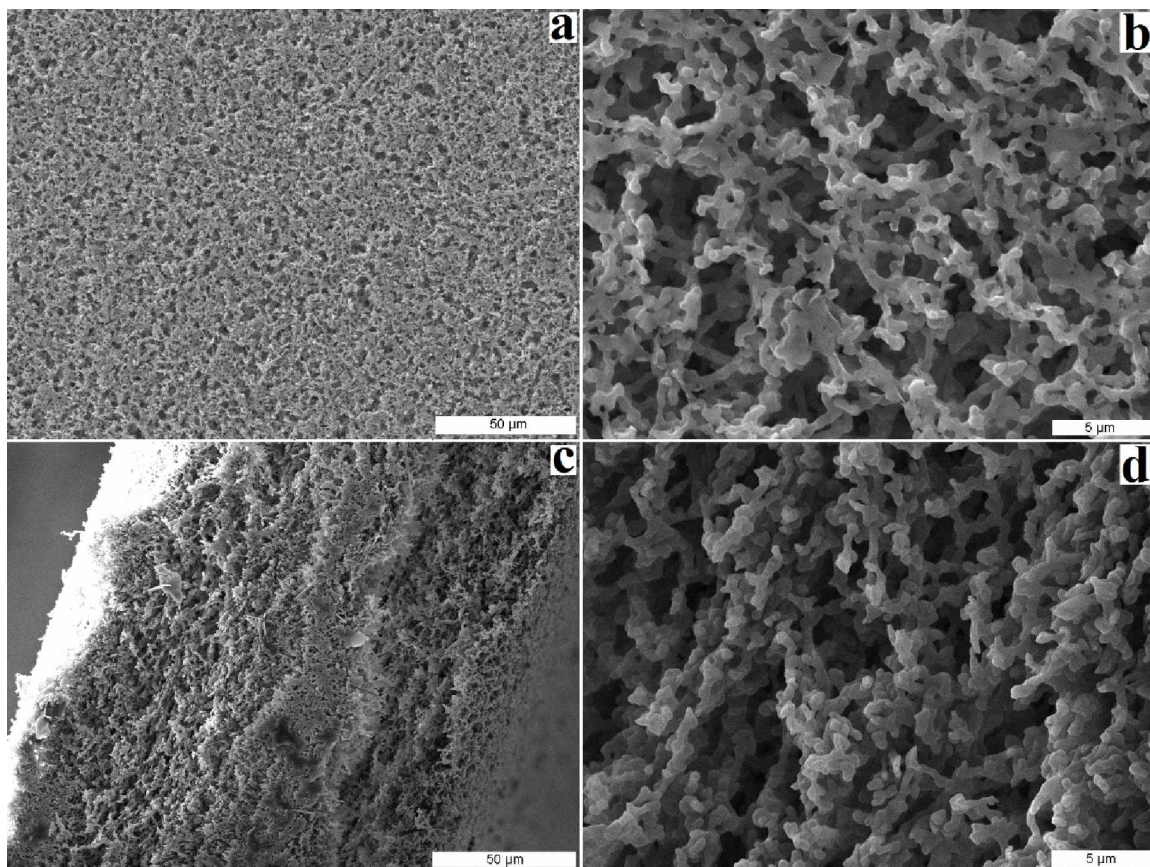


Fig. 2



Fig. 3

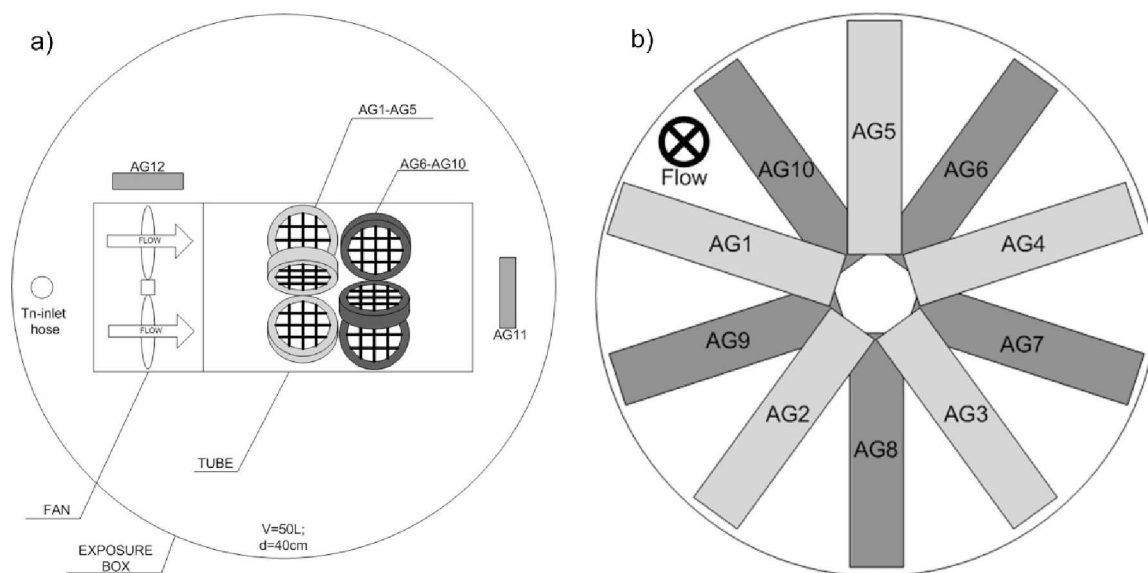


Fig. 4



Fig. 5



Fig. 6

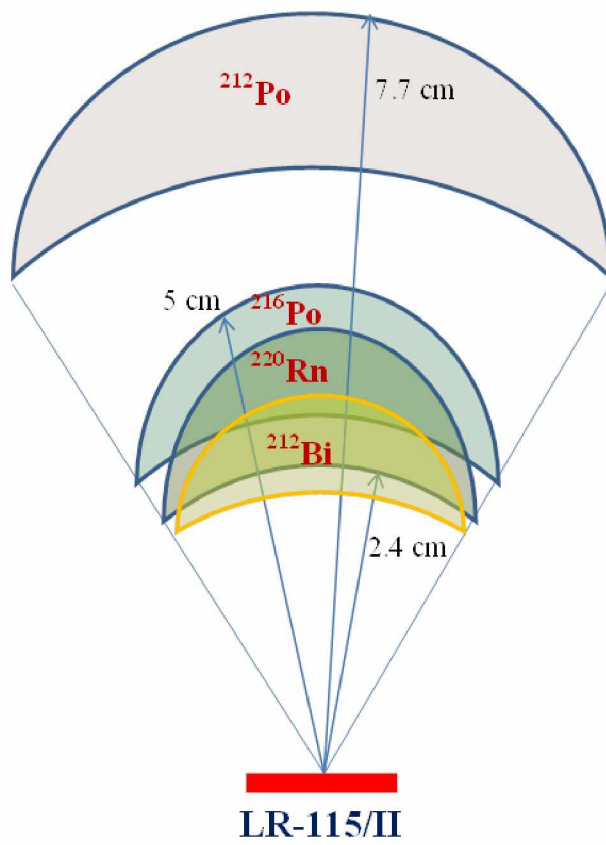


Fig. 7

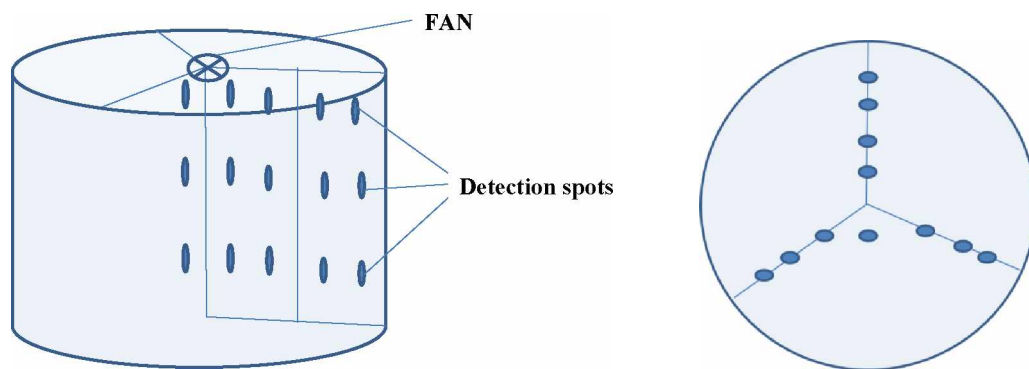


Fig. 8

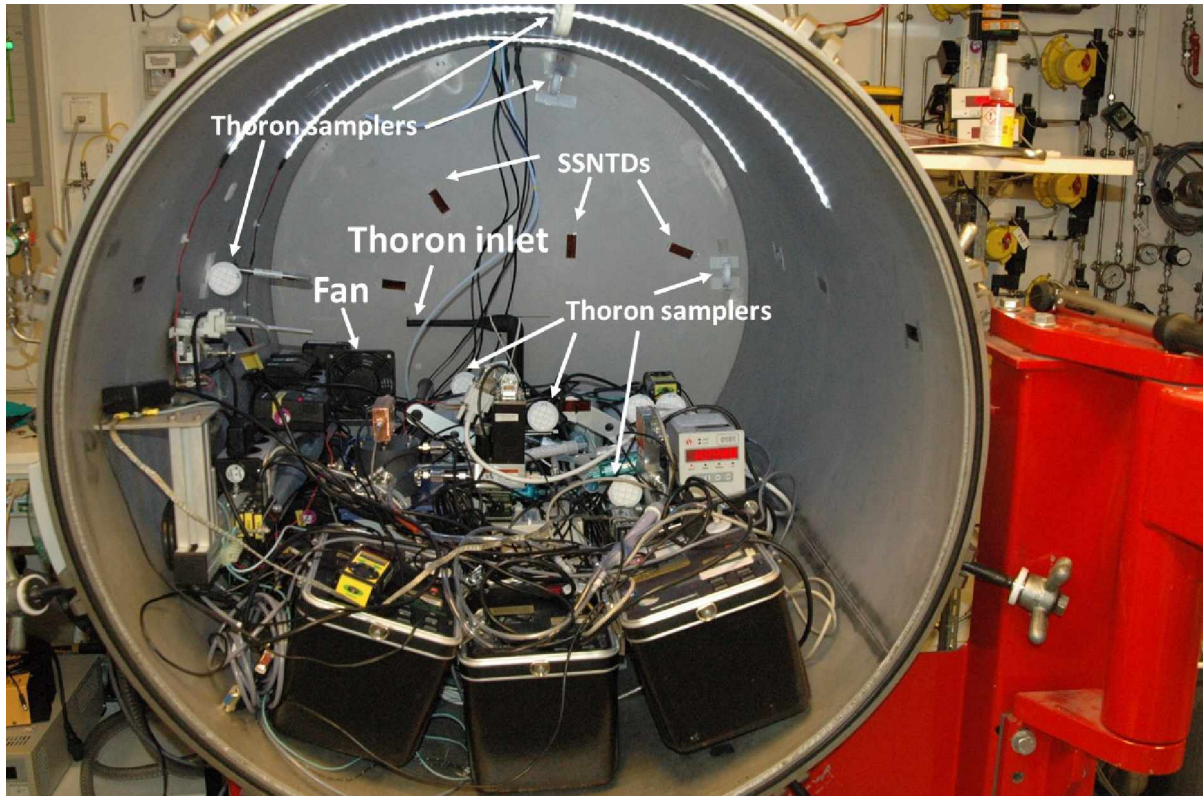


Fig. 9

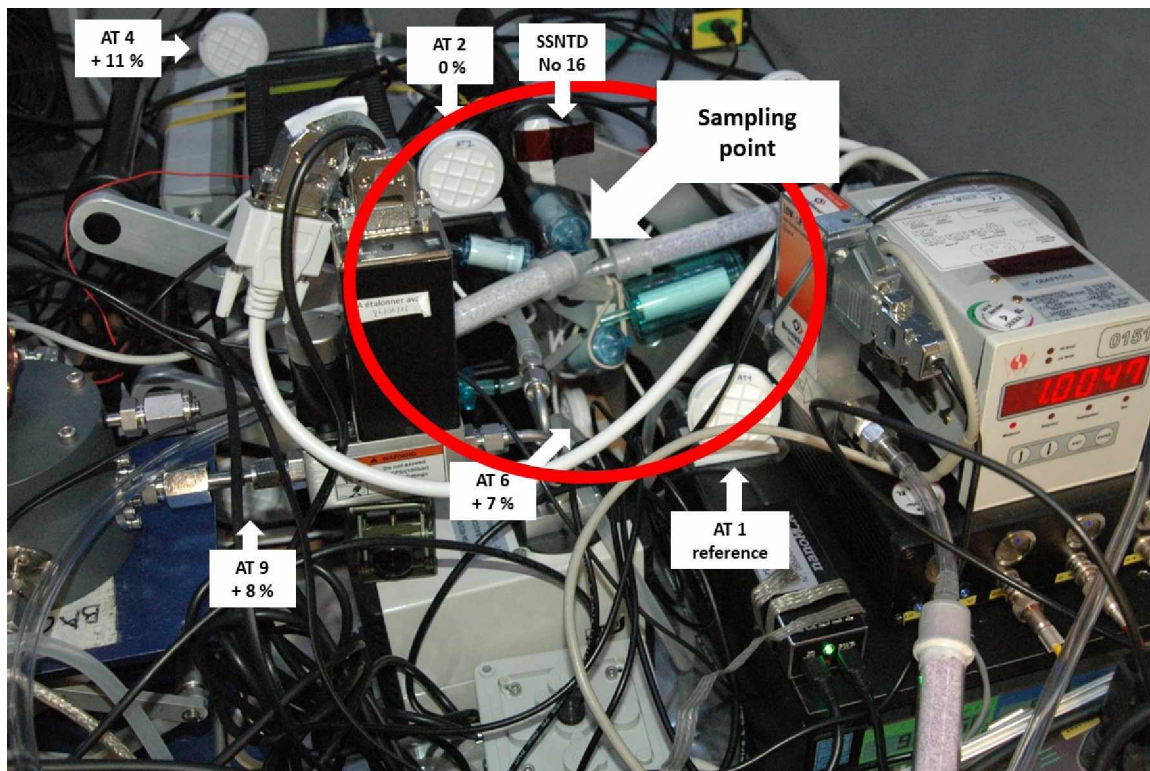


Fig. 10

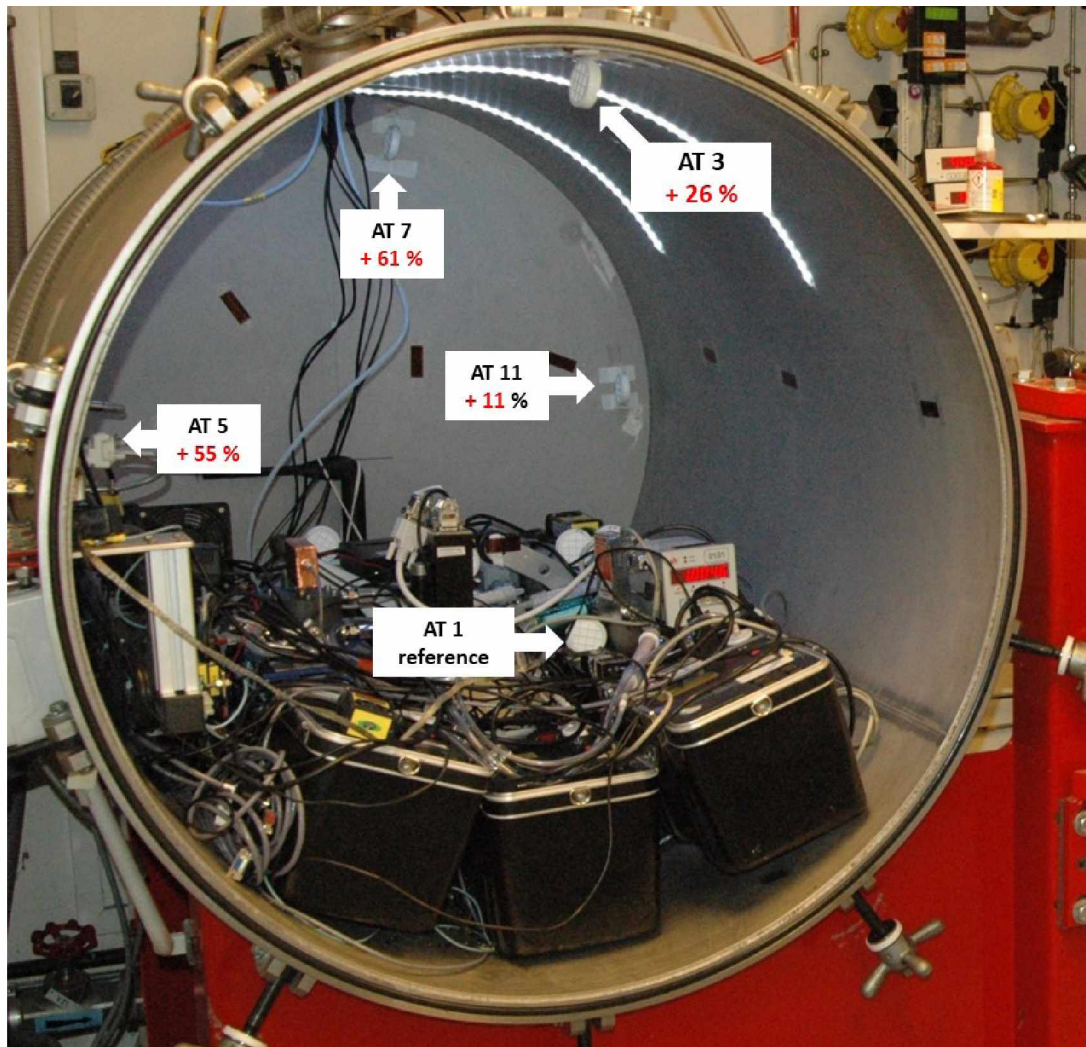


Fig. 11

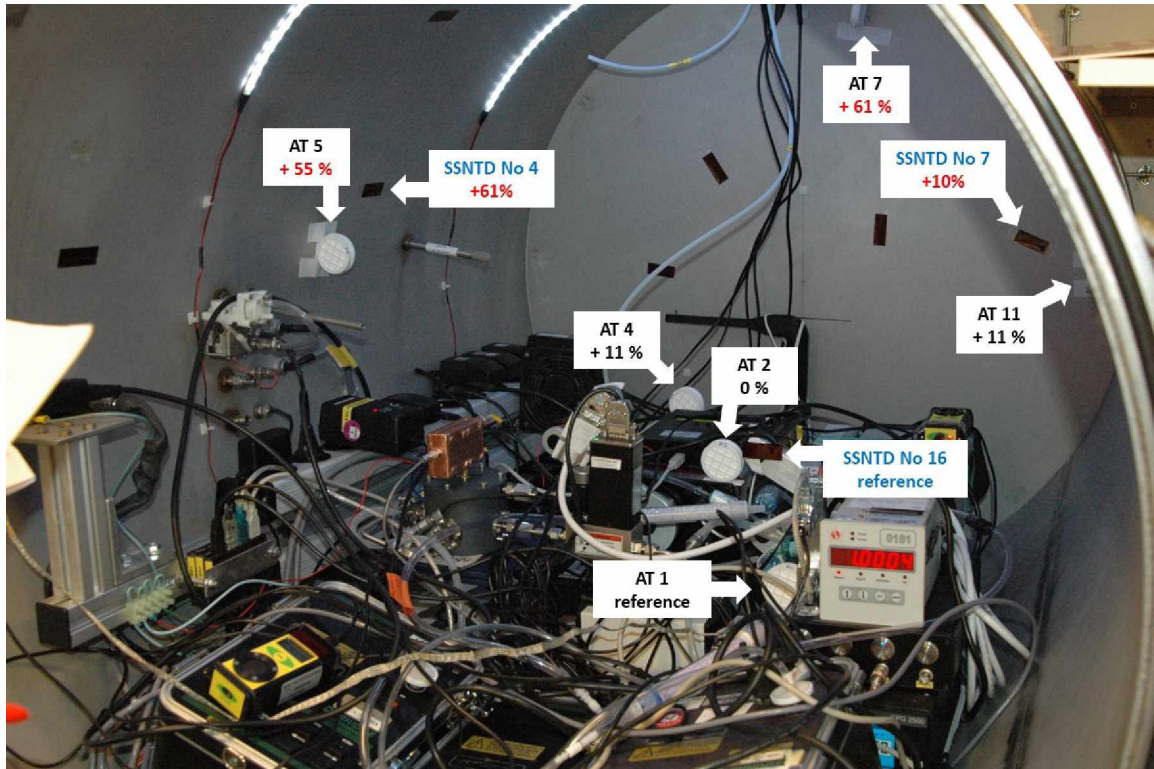


Fig. 12

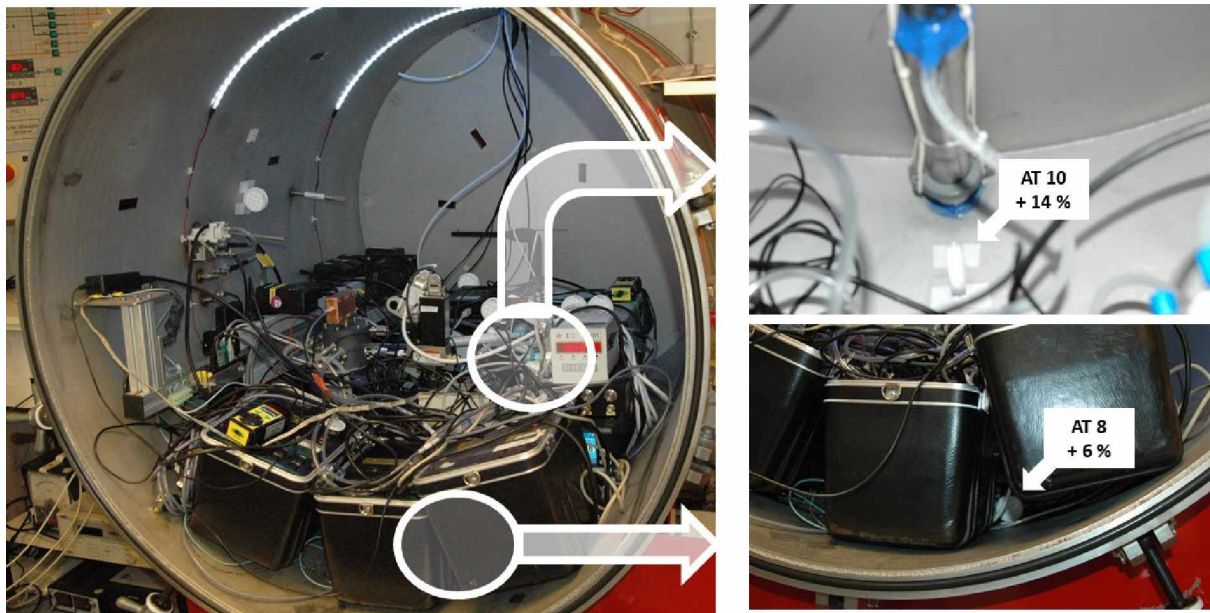
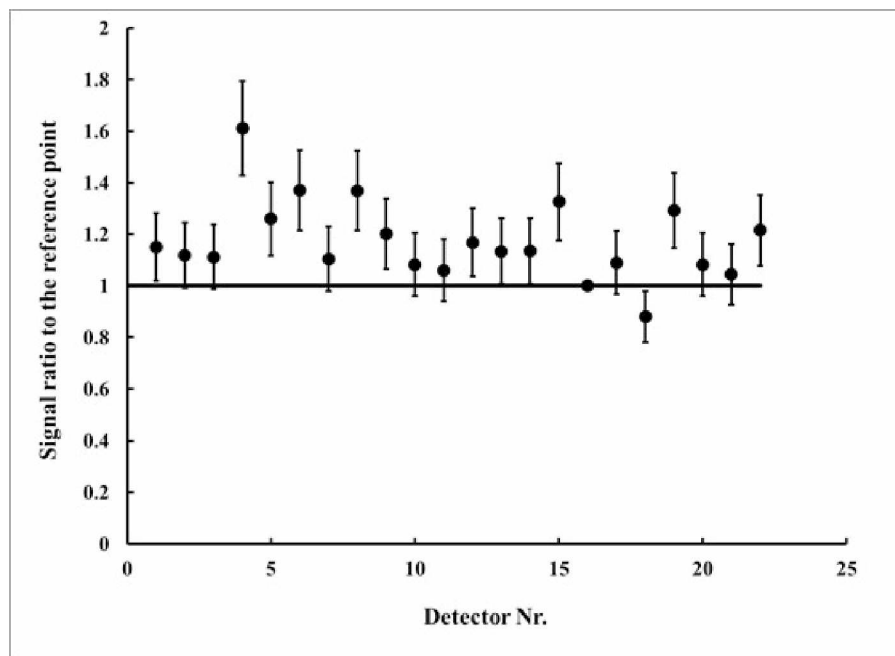


Fig. 13.

**Fig. 14**

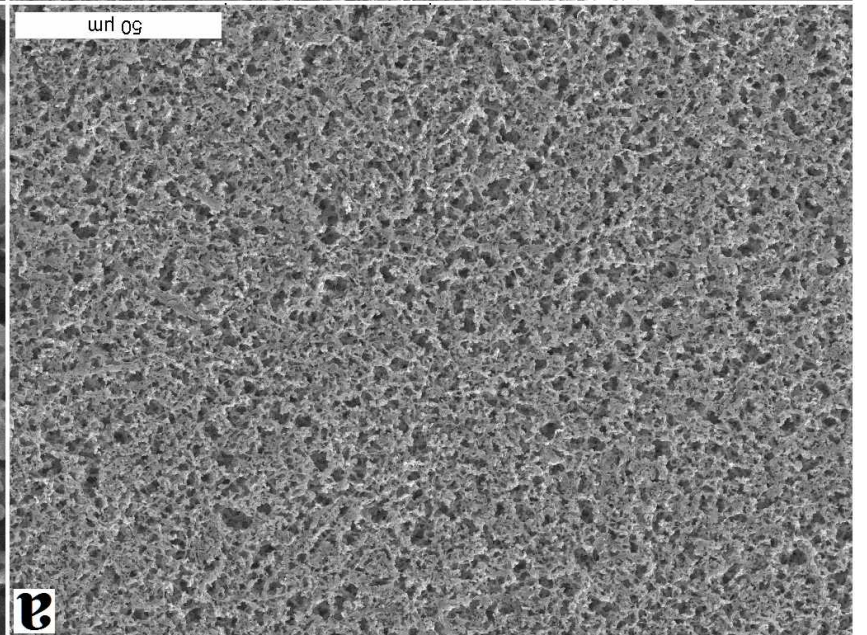
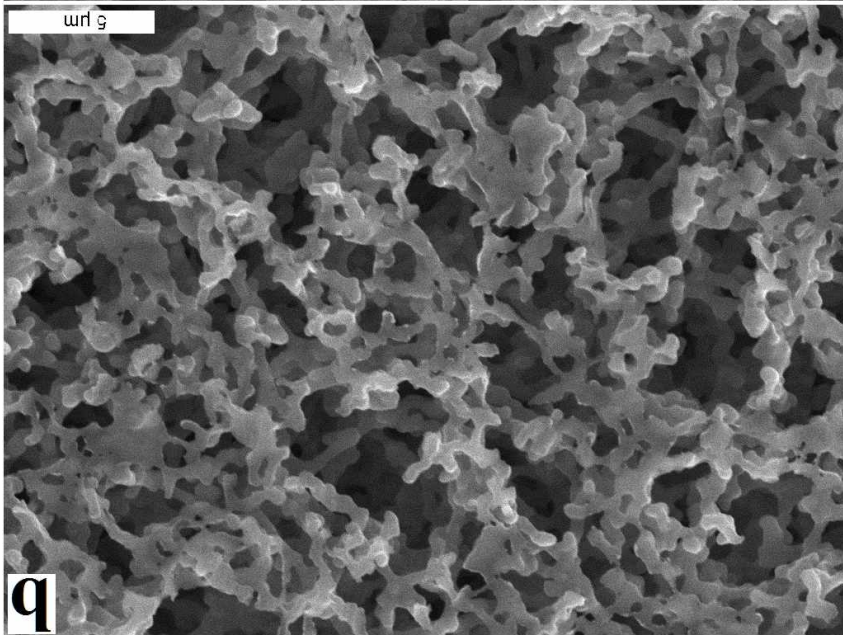
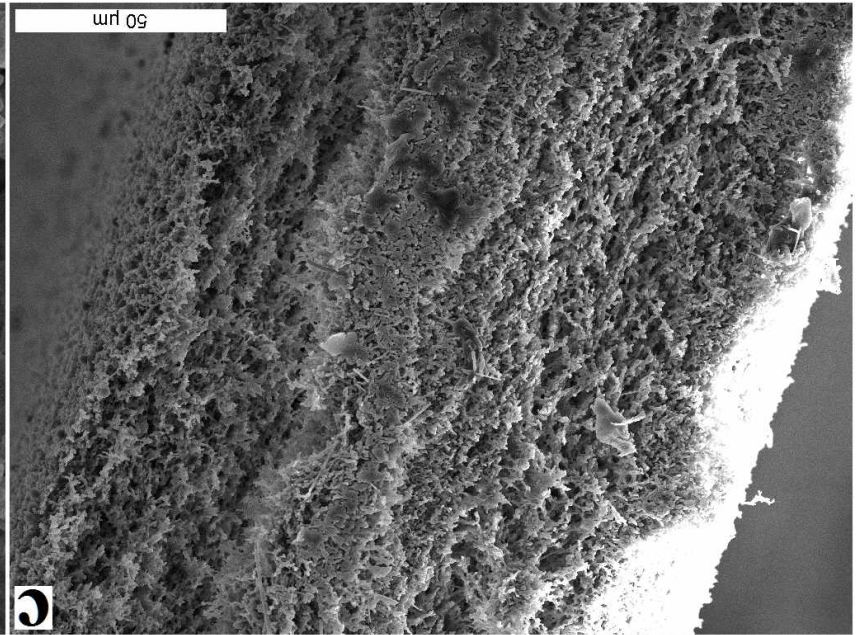
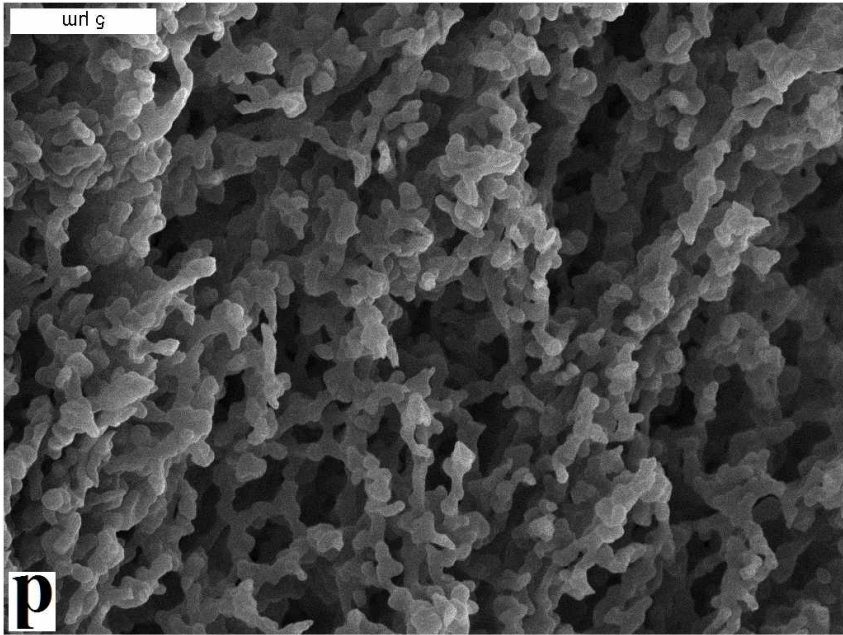


Table 1: Net counting rates per unit mass of thoron samplers with different heights exposed together to ^{220}Rn in air.

Sampler	L, cm	V, cm ³	Mass of the silica aerogel in the samplers, g	Net LS counting rate per unit mass of the silica aerogel, s ⁻¹ g ⁻¹
S1	1.0	15.21	5.421(5)	10.290(58)
S2	1.0	15.21	5.202(5)	10.415(60)
M1	1.5	22.81	8.649(5)	9.119(56)
M2	1.5	22.81	8.472(5)	9.054(58)
L1	2.0	30.41	11.932(5)	7.761(56)
L2	2.0	30.41	11.920(5)	7.714(55)

Table 2: Study of the repeatability of ^{220}Rn readings with thoron samplers. The numbers in the brackets indicate the overall estimated standard uncertainties.

Thoron sampler	Mass of the silica aerogel in the LS vial (g)	Net LS counting rate per unit mass of the silica aerogel at the end of the exposure($\text{s}^{-1}\text{g}^{-1}$)	Difference from the mean (%)
AG 1	5.014(5)	6.44(26)	1.3%
AG 2	5.064(5)	6.24(22)	-1.9%
AG 3	5.054(5)	6.40(14)	0.6%
AG 4	4.914(5)	6.31(16)	-0.8%
AG 5	5.055(5)	6.43(16)	1.0%
AG 6	5.050(5)	6.51(18)	2.3%
AG 7	5.040(5)	6.21(21)	-2.3%
AG 8	5.042(5)	6.26(12)	-1.6%
AG 9	5.043(5)	6.37(19)	0.1%
AG 10	5.021(5)	6.44(20)	1.3%
AG 11	5.062(5)	6.48(25)	1.8%
AG 12	4.884(5)	6.22(14)	-2.2%
mean AG1-AG10 \bar{x}	5.030	6.36	
std. dev. s	0.059	0.10	
$u_{bb} = \text{rel. std. dev. } s/\bar{x}$ (%)	1.2 %	1.6 %	

Table 3: Homogeneity study with thoron samplers during the thoron calibration exercise performed in the BACCARRA chamber. The numbers in the brackets indicate the overall estimated standard uncertainties.

Sample	Decay corrected net counting rate, cpm	Mass of the silica aerogel in the LS vial (g)	Specific net LS counting rate at the end of the exposure($s^{-1}g^{-1}$)	Center (C) / Walls (W)	Normalized to AT1
AT1	108.2(17)	5.346(5)	20.24(32)	C	1.00
AT2	111.9(18)	5.546(5)	20.17(33)	C	1.00
AT3	139.1(20)	5.444(5)	25.56(37)	W	1.26
AT4	122.6(20)	5.471(5)	22.41(37)	C	1.11
AT5	169.6(24)	5.402(5)	31.39(44)	W	1.55
AT6	116.2(22)	5.361(5)	21.68(41)	C	1.07
AT7	179.2(26)	5.503(5)	32.57(48)	W	1.61
AT8	119.6(24)	5.550(5)	21.54(44)	W	1.06
AT9	119.5(26)	5.454(5)	21.91(47)	C	1.08
AT10	127.7(27)	5.550(5)	23.00(50)	C	1.14
AT11	125.8(29)	5.598(5)	22.47(51)	W	1.11

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Conceptualization: K. Mitev, P. Cassette, D. Pressyanov;

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