WP2: Influence of thoron (^{220}Rn) and its progeny on radon end-user measurements and radon calibrations

SUBG, IRSN, CEA, STUK, BEV-PTP

WP2 leader: **SUBG** (prof. Dobromir Pressyanov)

Task 2.1, "Ensuring traceability of the secondary thoron reference instruments used in the experimental research to the primary thoron measurement system at IRSN"

A2.1.1	IRSN and SUBG will establish and evaluate the reference thoron atmospheres in their reference test chambers. The range of thoron activity concentrations that can be created in both chambers will be assessed. IRSN will assess the homogeneity of the thoron atmosphere in their BACCARA 1 m^3 test chamber using numerical simulation and experimentation. SUBG will study experimentally the inhomogeneity in their 50 L test chamber.	**IRSN**, SUBG

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

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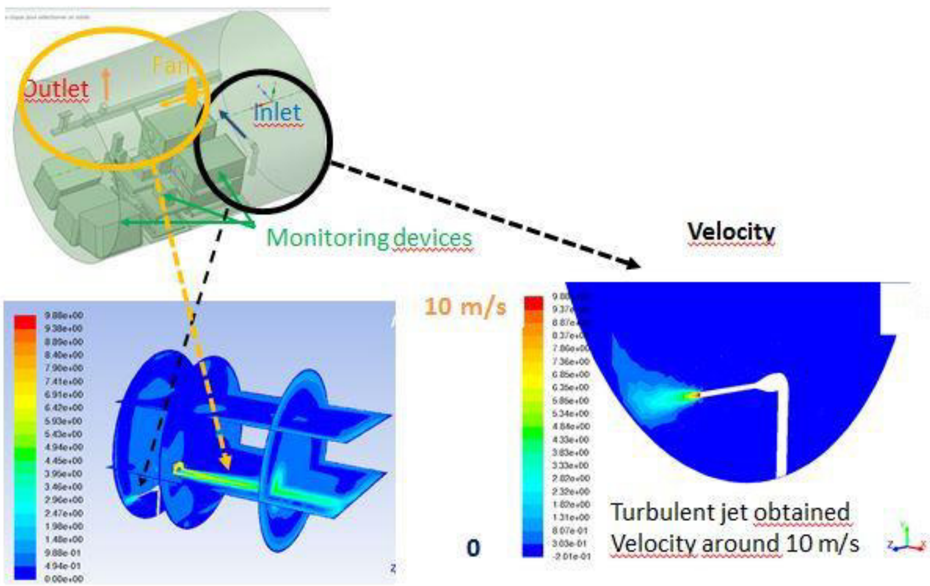
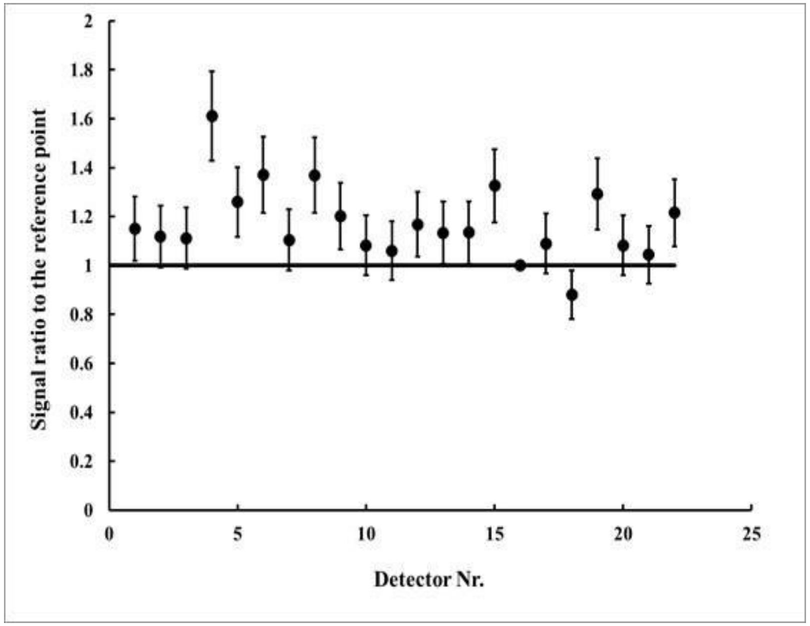
Numerical simulations and experimental results

Numerical simulations (IRSN)



Experimental results (SUBG, CEA)



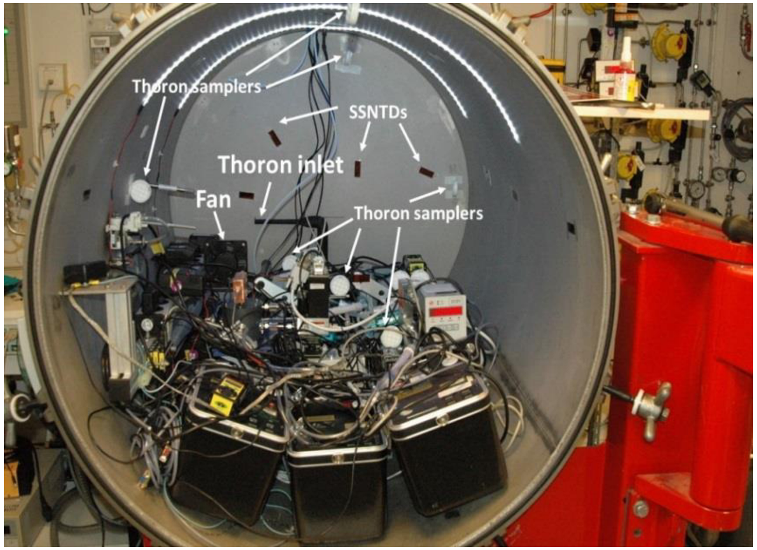
Findings (when fan operates in the exposure volume): In a 50 L exposure box thoron inhomogeneity is within 10%, while in the 1 m^3 BACCARA chamber it may be up to 60%

Task 2.1, "Ensuring traceability of the secondary thoron reference instruments used in the experimental research to the primary thoron measurement system at IRSN"

A2.1.2	IRSN, SUBG and STUK will jointly organise an exercise to calibrate their secondary thoron reference instruments for activity concentrations of 10^2-10^6 Bq/m^3 at the IRSN radon/thoron calibration laboratory at Saclay, France. Information from A2.1.1 on the homogeneity of the thoron atmosphere will be taken into account. SUBG will participate in the calibration with its reference thoron monitors – AlphaGuard 2000 RnTn Pro and RAD 7, whilst STUK will participate with its thoron monitors: AlphaGuards and Lucas cells (Pylon Inc). In addition BEV-PTP will send their AlphaGuard to IRSN for calibration.	**IRSN**, BEV-PTP, STUK, SUBG

Thoron calibration exercise was organized and carried-out in IRSN in May 2018 (IRSN, SUBG, STUK, BEV-PTP).

Thoron calibration exercise: the primary thoron measurement system (B. Sabot, et al., *Applied Radiation and Isotopes* 118 (2016) 167-174) was used as a reference



Three different constant ^{220}Rn reference atmospheres, i.e. 10 kBq.m^{-3}, 46 kBq.m^{-3} and 240 kBq.m^{-3}, have been established to cover a wide range of ^{220}Rn activity concentrations and calibrate seven instruments.

Ratios between the ^{220}Rn activity concentrations measured by the instrument and the reference activity concentration have been found close to 1 for the four AlphaGUARD and around 0.6 for the three RAD7. The calibration factor of an alphaGUARD changes with the thoron activity concentration.

The results obtained in this work reveal some discrepancies in the readings of certain types of ^{220}Rn measurement instruments and emphasize the importance of the metrological assurance of ^{220}Rn measurements.

Key outcomes of Task 2.1

- **The testing of radon monitors for thoron sensitivity should include reference (secondary) thoron monitor that is calibrated with, or traceable to a primary thoron measuring system. We recommend the reference instrument to be placed inside the exposure chamber throughout the exposure;**

- **Thoron homogeneity in the chamber should be checked during exposure or in another experiment under the same exposure conditions;**

- **The ratios between the ^{220}Rn activity concentrations measured by the instrument and the reference activity concentration measured by the IRSN reference instrument is close to 1 for the four AlphaGUARDs and around 0.6 for the three RAD7s.**

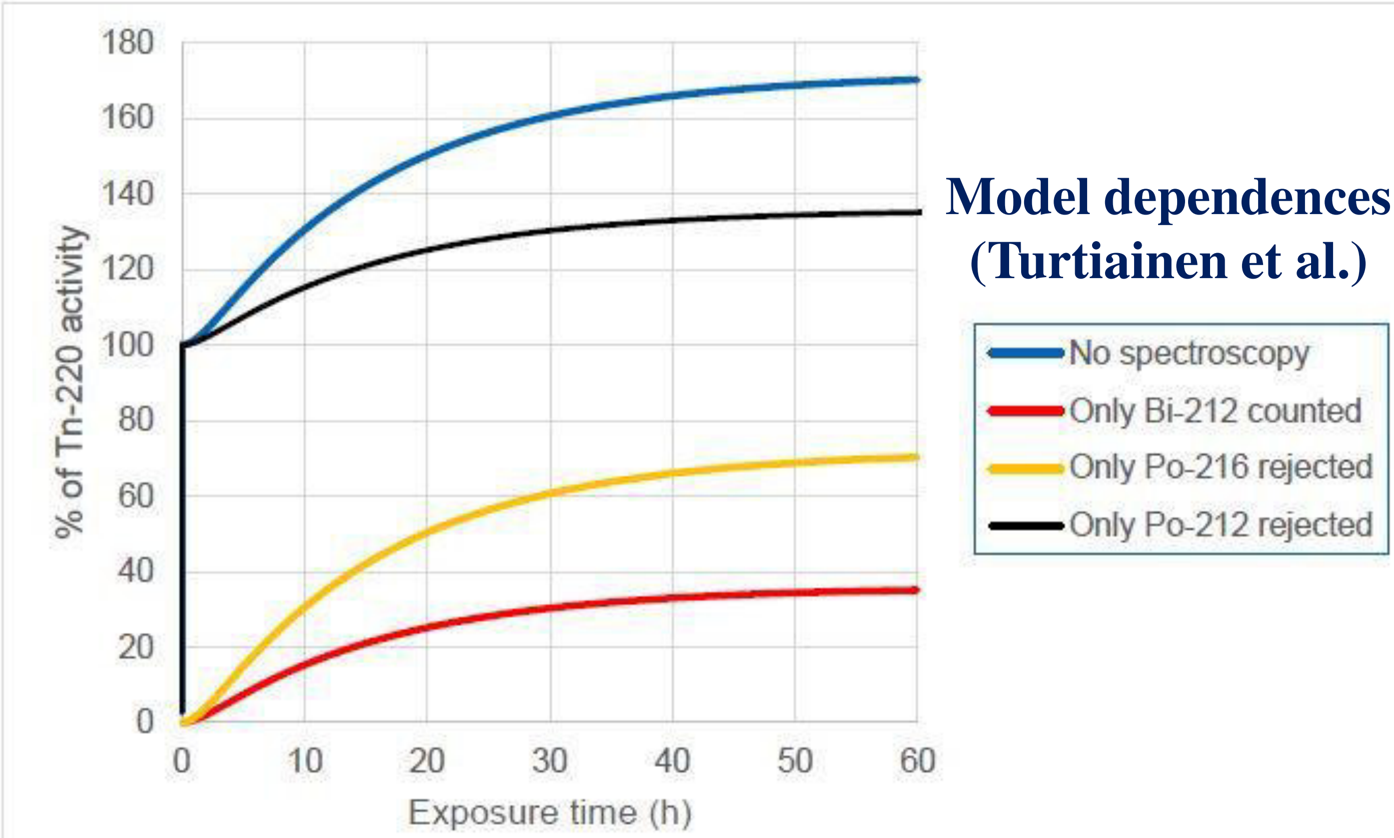
Task 2.2, “Investigation of the influence of thoron on radon measurements and calibrations”

A2.2.1	IRSN, SUBG and STUK will study the influence of thoron on active radon monitors. Measurements will be performed in both thoron and radon plus thoron atmospheres using the secondary reference instruments calibrated in A2.1.2. At least 10 instruments available at IRSN, CEA, SUBG, STUK will be studied for example AlphaGUARD (different types), DoseMan, radhomeHR3, BARASOL, monitors with Lucas cells, RAD7 etc. Theoretical models and analysis will be employed by IRSN, SUBG and STUK to determine, understand and potentially correct for the influence of thoron on the performance of the radon monitors.	**IRSN**, CEA, STUK, SUBG

16 Active monitors tested ($CI = \frac{E_{Rn}}{E_{Tn}} \times 100\%$)

- $CI \leq 5\%$: 5 (out of 16)
- $5\% < CI \leq 20\%$: 6 (out of 16)
- $CI > 20\%$: 5 (out of 16)

Thoron interference of active monitors may depend on the exposure time, due to the contribution from thoron progeny (^{216}Po, ^{212}Pb, ^{212}Bi+^{212}Po/^{208}Tl)



Model dependences (Turtiainen et al.)

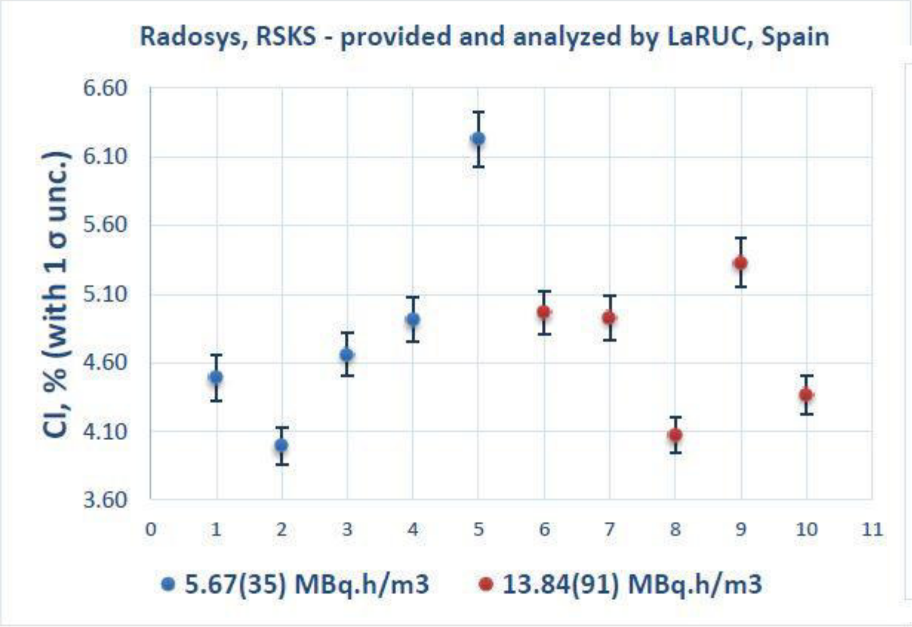
Task 2.2, “Investigation of the influence of thoron on radon measurements and calibrations”

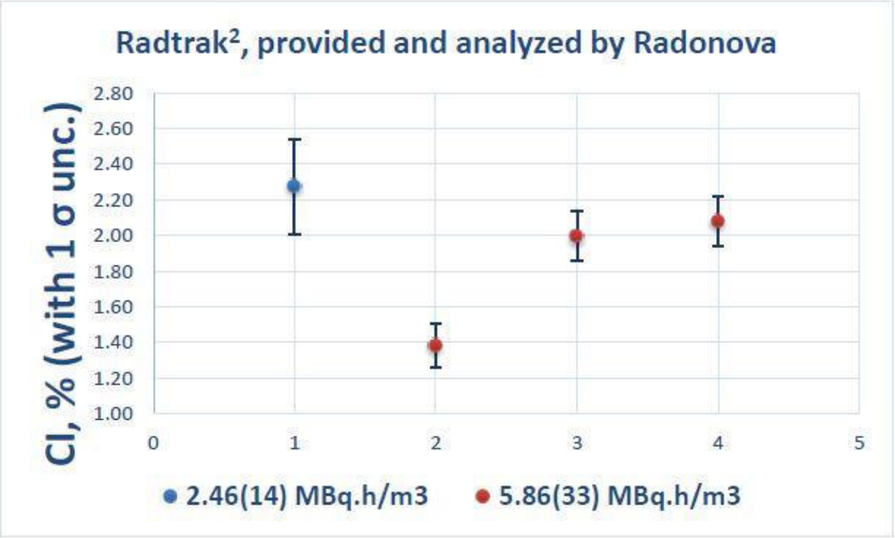
A2.2.2	CEA, IRSN, STUK, and SUBG will study the influence of thoron on passive integrating radon detectors. Measurements will be performed in both thoron and radon plus thoron atmospheres using the secondary reference instruments calibrated in A2.1.2. At least 10 detectors available at IRSN, CEA, SUBG and STUK will be studied, for example diffusion chambers with different alpha-track detectors, E-Perm electret chambers, compact disks/DVDs, etc. Theoretical models and analysis will be employed by CEA, IRSN, STUK, and SUBG to determine, understand and potentially correct for the influence of thoron on the performance of the radon monitors.	**SUBG**, CEA, IRSN, STUK

19 passive detectors tested: 17 commercially available + 2 DVD based

- CI ≤ 5%: 8 (out of 19)
- 5% < CI ≤ 20%: 9 (out of 19)
- CI > 20%: 2 (out of 19)

Individual variations among different detectors of one kind were observed



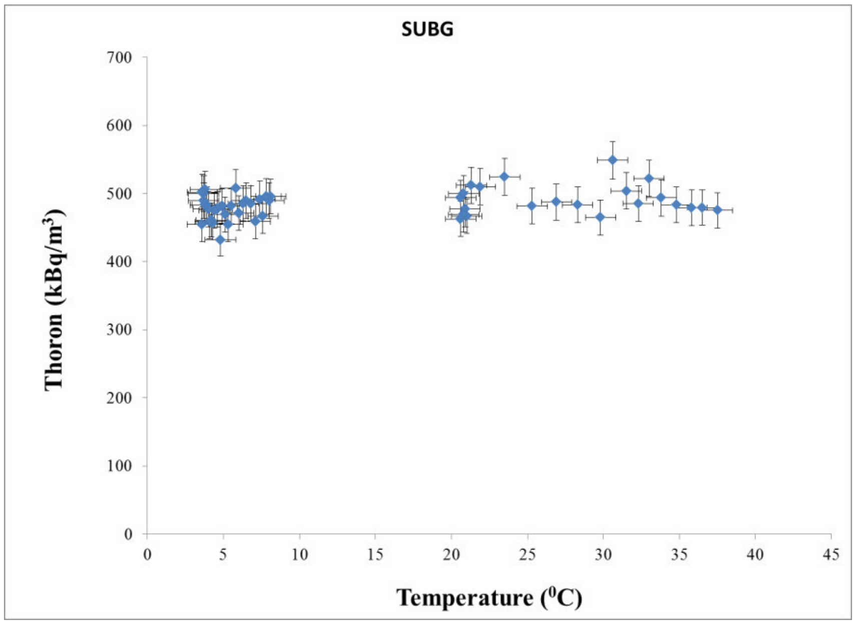


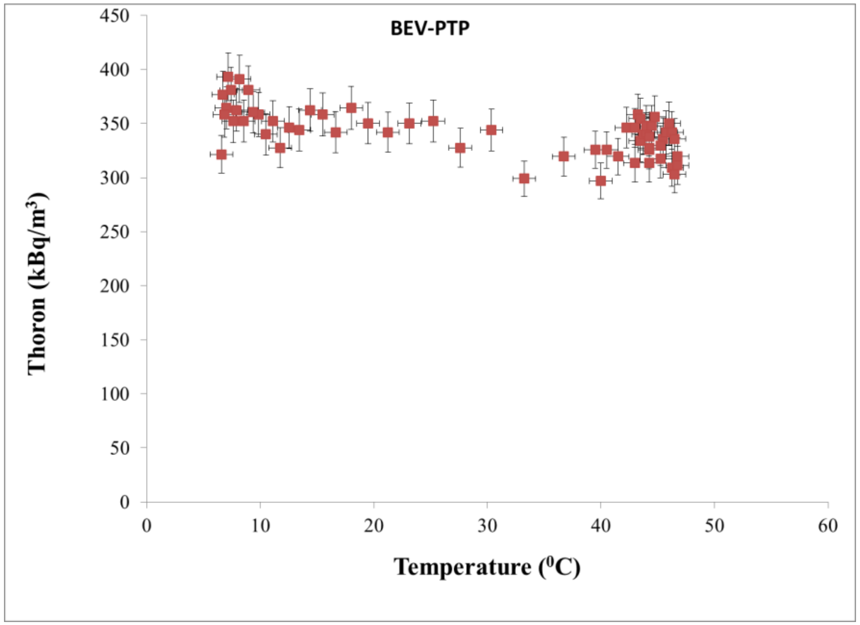
Task 2.2, “Investigation of the influence of thoron on radon measurements and calibrations”

A2.2.3	Using SUBG's laboratory facilities and the secondary reference instruments calibrated in A2.1.2, SUBG and BEV-PTP will study experimentally the response of radon/thoron measurement instruments/detectors available at SUBG and BEV-PTP at different environmental temperatures (between +5 °C and +45 °C) in both thoron and radon plus thoron atmospheres, under both static and dynamic regimes. SUBG and BEV-PTP will also study the response of radon/thoron measurement instruments under different radon/thoron concentrations ratios. Theoretical models and analysis will be employed by SUBG and BEV PTP to determine, understand and potentially correct for the influence of thoron on the performance of the radon monitors.	**SUBG**, BEV-PTP

The response of active monitors (SUBG, BEV-PTP) and passive detectors (SUBG, Radonova) was studied at different temperatures (between 5 ^{0}C and 45 ^{0}C)

A2.2.3: Two reference monitors (AlphaGUARD PQ2000 RnTn Pro) of SUBG and BEV-BTP were experimentally tested in SUBG for temperature influence on the response to thoron





Slight positive correlation. Bias in 5-45 ^{0}C within 5%

Slight negative correlation. Bias in 5-45 ^{0}C within 10%

Temperature dependence of diffusion chambers with Kodak-Pathe LR-115/II detectors

THE PROBLEM WITH TEMPERATURE DEPENDENCE OF RADON DIFFUSION CHAMBERS WITH ANTI-THORON BARRIER

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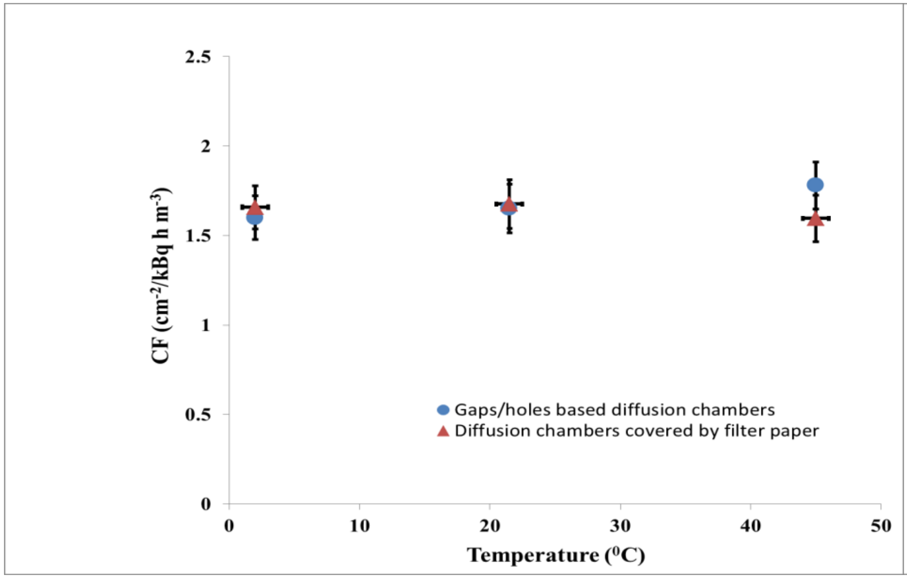
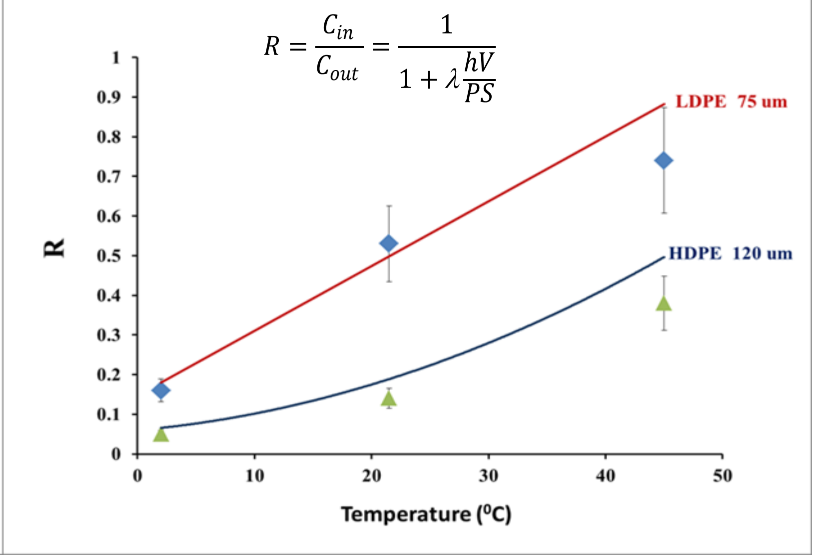


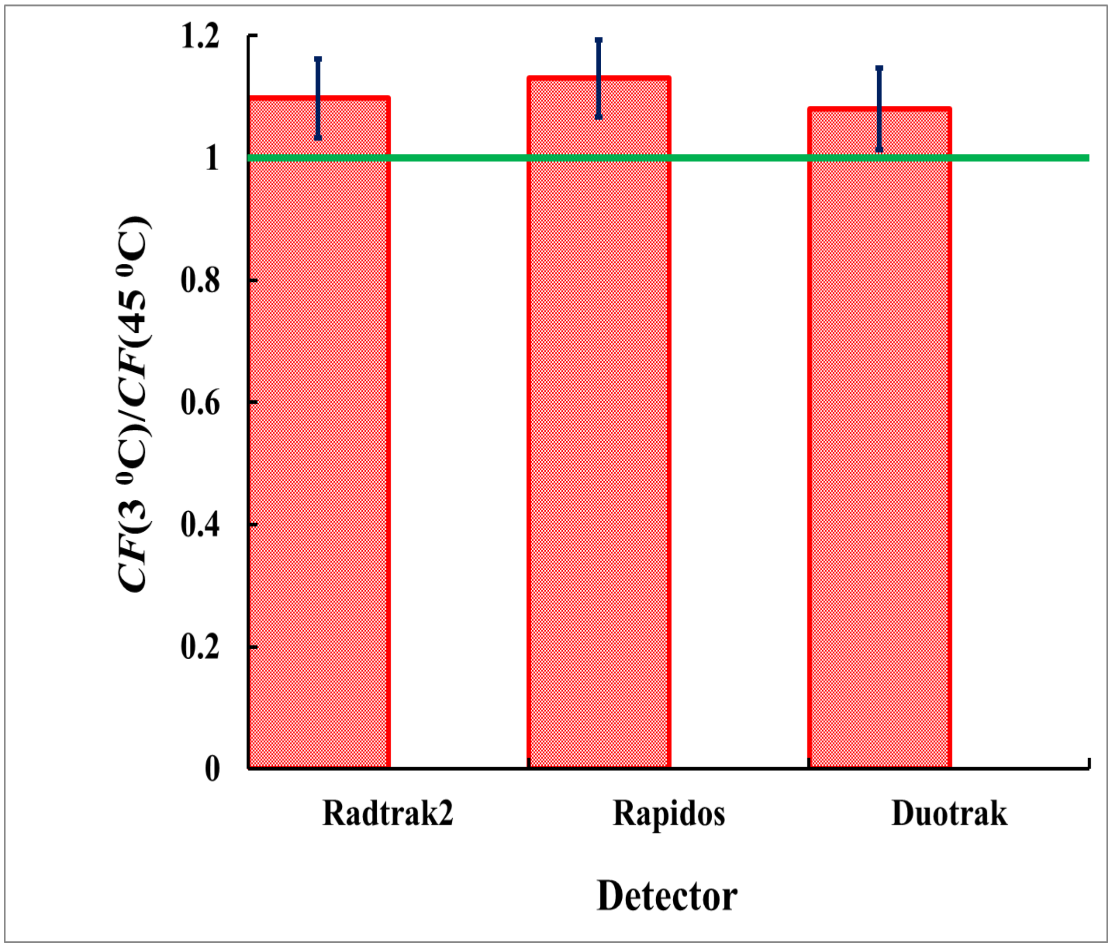
Anti-thoron barrier: gaps/pin holes



$$R = \frac{C_{in}}{C_{out}} = \frac{1}{1 + \lambda \frac{hV}{PS}}$$

Anti-thoron barrier: polymer foil

The ratio of calibration factors at 3 ^{0}C to 45 ^{0}C of some RADONOVA chambers with CR-39 detectors. From 3 to 45 ^{0}C the calibration factor drops by 8 – 13%



Key outcomes of Task 2.2

- Different types of radon monitors/detectors show different thoron interference. The thoron interference of some passive detectors may vary between different detectors of one type.
- The manufacturers of radon monitors/detectors should perform cross-interference testing for their radon instruments and should include this information in the specifications of the instrument.
- The cross-interference tests should be planned taking into account the possible contribution from thoron progeny (^{216}Po, ^{212}Pb, $^{212}Bi+^{212}Po/^{208}Tl$). For instance after the end of thoron exposure the integrated passive detectors should be left for at least three days at low thoron/radon levels before analysis. This is needed to leave thoron progeny deposited in their volume to decay.
- **A minimum of three days test with a high thoron activity concentration (around 10 kBq.m^{-3} or more) is recommended to determine an accurate final *CI* of the active monitors, instead of the 4 hours at 1000 Bq.m^{-3} required in the IEC 61577-2 standard.**
- Some radon detectors show temperature dependence in their response.

Task 2.3, “Development of techniques to reduce the influence of thoron on radon measurements and calibrations”

A2.3.1	STUK and SUBG will undertake a literature review of potential techniques and materials to reduce the influence of thoron on radon measurements and calibrations. Based on these findings, STUK and SUBG will perform an analytical analysis of the different techniques/materials and will identify the most promising ones, based on the effectiveness of the relative differentiation between thoron and radon.	**STUK**, SUBG

Review of potential techniques and materials to reduce the influence of thoron on radon measurements and calibrations

Olli Holmgren[1], Tuukka Turtiainen[1], Krasimir Mitev[2] and Dobromir Pressyanov[2]

[1]STUK, [2]SUBG

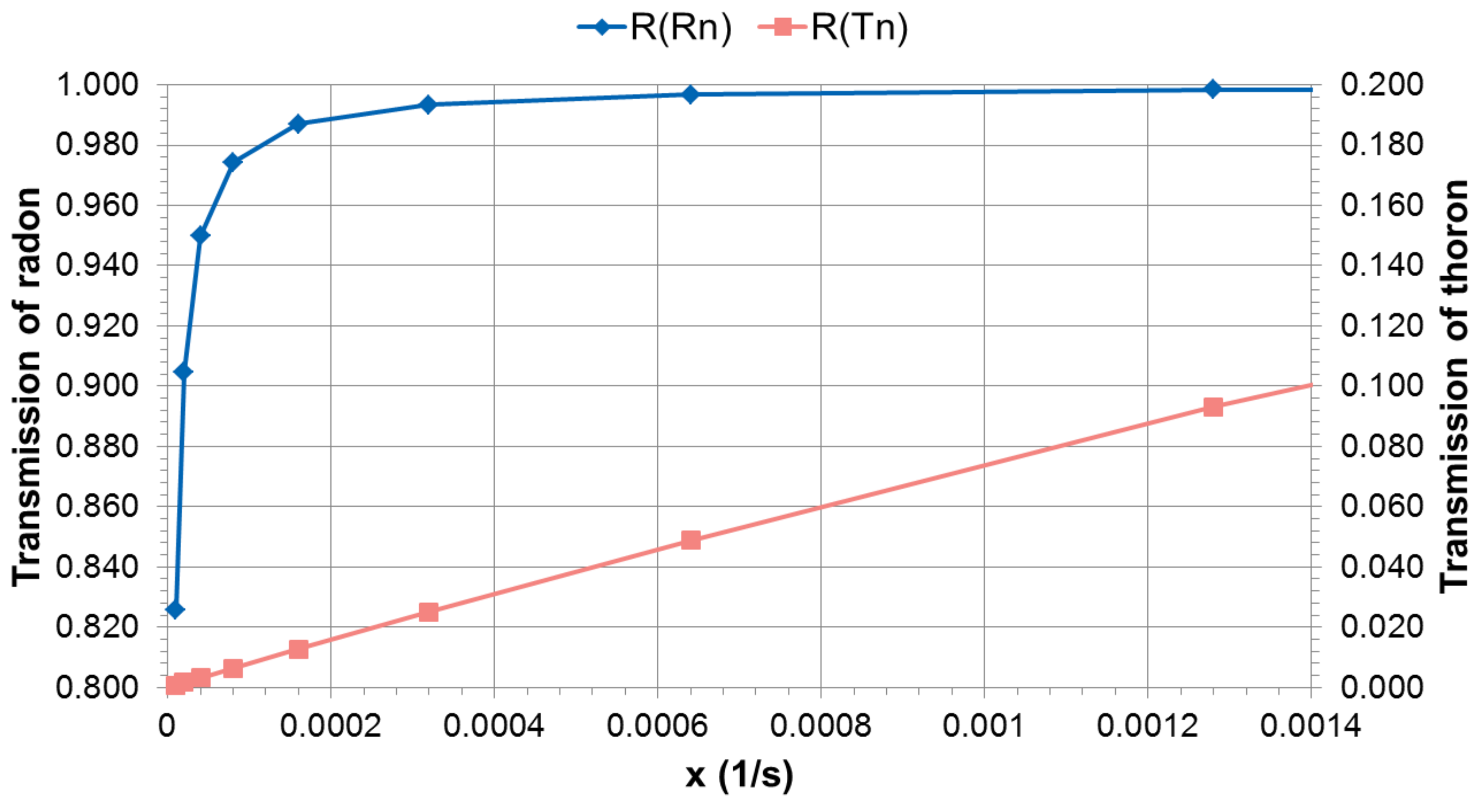
MetroRadon WP 2, Report on the activity A2.3.1

Basic methods to reduce the influence of thoron:

- Passive (diffusion barriers: diffusion through gaps/pin holes or through polymer foils);
- Active (delay lines, spectroscopy discrimination, counting in two or more time intervals)

Transmission (R) of radon and thoron through diffusion barriers:

$$R = \frac{C_{in}}{C_{out}} = \frac{1}{1+\lambda\tau} = \frac{1}{1+\frac{\lambda}{x}}$$



Gaps/holes: $x = \frac{D_{air}A}{Vd}$ **Polymer membrane:** $x = \frac{PS}{hV}$

Task 2.3, "Development of techniques to reduce the influence of thoron on radon measurements and calibrations"

A2.3.2	SUBG will evaluate the properties of different filters/foils/ membranes identified in A2.3.1 that could potentially serve as selective thoron barriers and will evaluate their radon permeability. SUBG will initially undertake a literature survey of radon (^{222}Rn) permeability data. SUBG will then perform an experimental study of the permeability of radon (^{222}Rn), using it as a thoron (^{220}Rn) surrogate, because both isotopes have the same solubility and diffusion coefficient. Using the radon permeability data, the thoron permeability of the various materials will be evaluated and the most promising materials identified.	**SUBG**

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Article

Partition Coefficients and Diffusion Lengths of ^{222}Rn in Some Polymers at Different Temperatures

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Task 2.3, “Development of techniques to reduce the influence of thoron on radon measurements and calibrations”

A2.3.3	SUBG and CEA will develop and then characterise one or more selective barriers for ^{220}Rn identified in A2.3.2. It is well known that radon is soluble in some polymers and this solubility can be used to design/select a polymer membrane that allows diffusion of radon and not thoron. Using data from A2.3.2, the nature and thickness of the membranes will be carefully chosen so that the diffusion time is much greater than the thoron half-life, but much less than the half-life of radon. The performance of such barriers will be experimentally tested on the CEA and SUBG facilities and expressed, for example, as attenuation factors for both gases.	**SUBG**, CEA

A workshop in the framework of activity A.2.3.3 of WP 2 of MetroRADON on **“Transport of Radon and Thoron in Polymers”** was organized (head organizer: K. Mitev) on 21-22 March 2019 at the Faculty of Physics, Sofia University “St. Kliment Ohridski” (**SUBG**)

Task 2.3, “Development of techniques to reduce the influence of thoron on radon measurements and calibrations”

A2.3.4	Based on the results from A2.3.1-A2.3.3, SUBG, IRSN, STUK, CEA and BEV-PTP will develop recommendations on the construction of radon monitors that are not sensitive to thoron including the technical concepts / solutions aimed at reducing thoron-related bias in the radon signal in existing monitors. SUBG, IRSN, STUK, CEA and BEV-PTP will also develop recommendations for tests to check the sensitivity of radon monitors and detectors to thoron.	**SUBG,** BEV-PTP, CEA, IRSN, STUK

Passive methods (by diffusion barriers)

- Diffusion through polymer foils (can eliminate thoron influence, eliminates also humidity);
- Diffusion through small gaps/holes (depending on the construction can reduce thoron influence to < 10%, however, detectors are affected by humidity);

Active method:

- Delay due to air flow in a pipe;

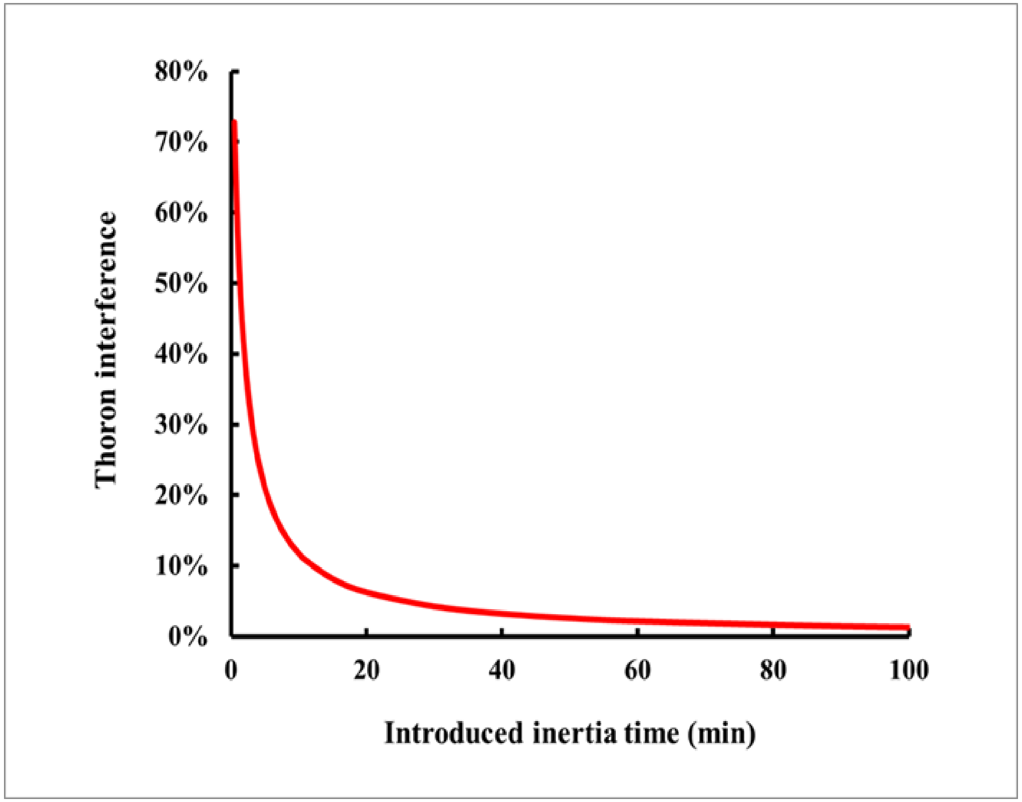
Other:

- Spectroscopy discrimination between radon and thoron or gross alpha counting in two or more time intervals (active monitors).

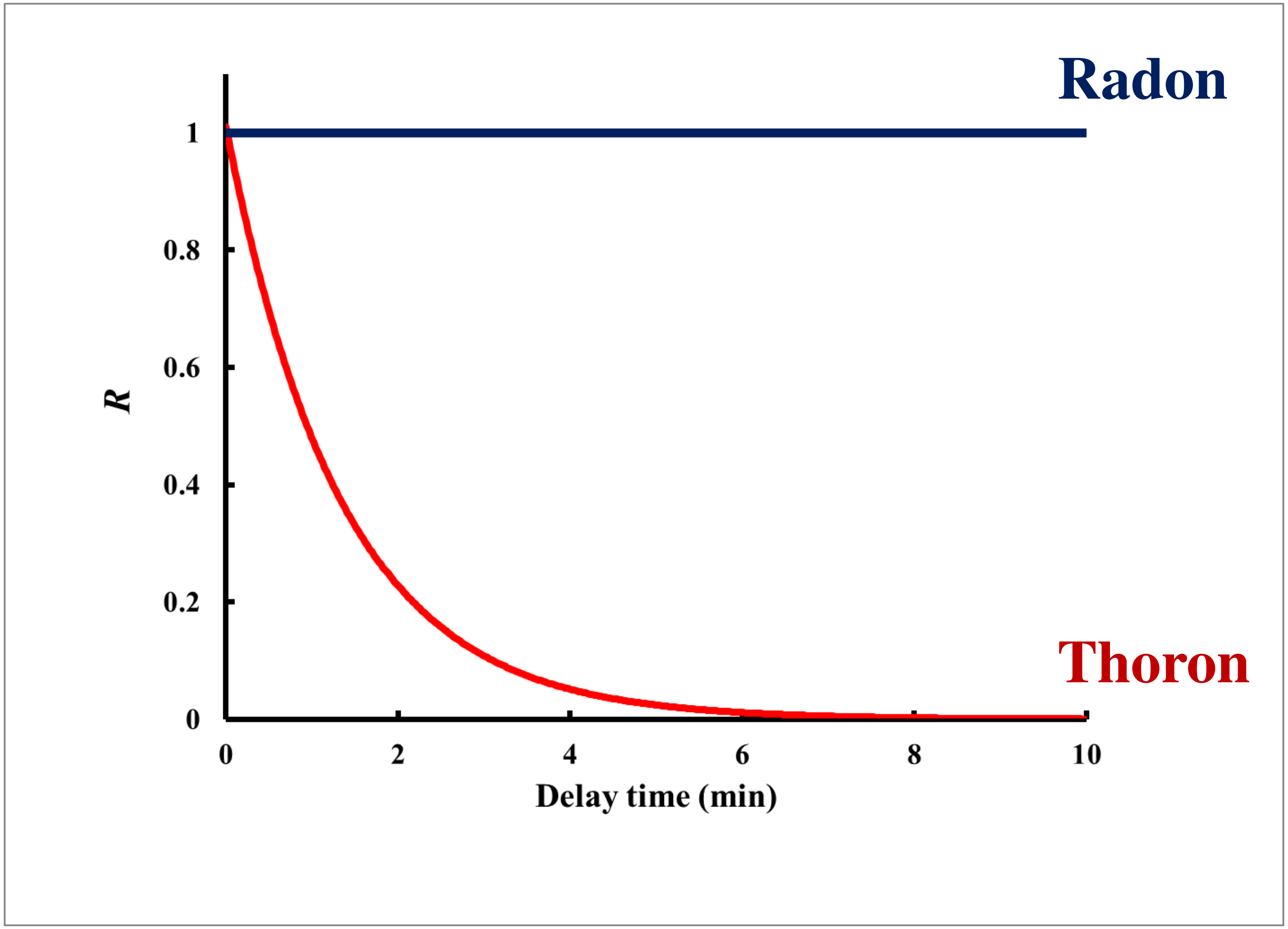
Introducing additional diffusion barrier to active radon monitors

CI = (8.8 ± 1.3) % → CI=(4.23 ± 0.84) %

Inertia in the response, introduced by diffusion barriers against thoron



Delay lines: $Delay\ time = \frac{Volume\ of\ the\ line}{Air\ flow{-}rate}$



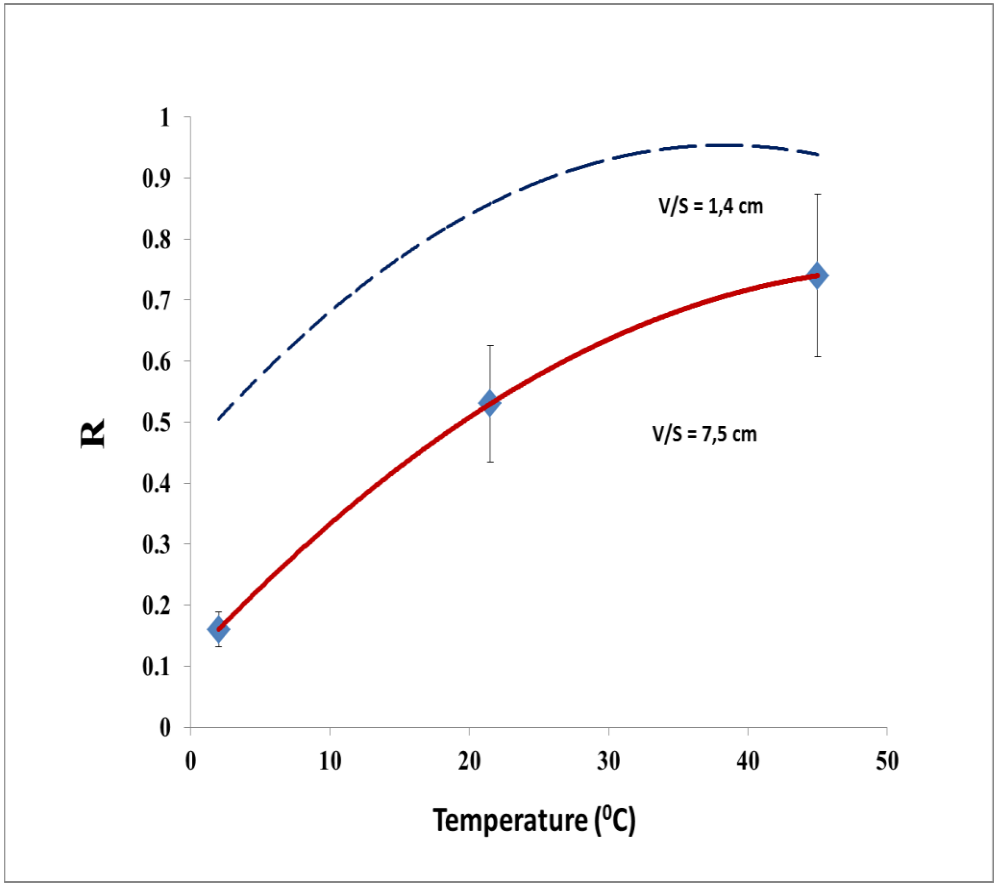
The temperature dependence of radon transmission through polymer membranes is a problem that gives an (surprising) opportunity, because:

… many radon detectors have temperature dependence of the response which is reciprocal to that of the radon transmission (R), i.e. decreasing with the increase of the temperature. Such detectors are e.g.:

- The most widely used track detectors: CR-39. These detectors show fading, and the (fading) decrease of the signal is larger at higher temperature (see e. g. H. Enomoto and N. Ishigure, 2011; M. Caresana et al., Radiat. Meas. 45 (2010) 183–189).
- Some detectors that employ radon absorption/adsorption (e.g. detectors based on activated charcoal; radon film badges (Tommasino et al., 2009) etc.);

Surprising outcome (not planned): The temperature dependences introduced by polymer anti-thoron barriers and that of many radon detectors are reciprocal. This can be used to reduce/eliminate the temperature dependence, the thoron influence and also the humidity influence (Patent Appl. Bulg. Nr. 112897, WIPO Appl. Reg. Nr. PCT/BG2020/000003).

Experimental results with a membrane of 75 μm thick low density polyethylene foil and *V/S* = 7.5 cm

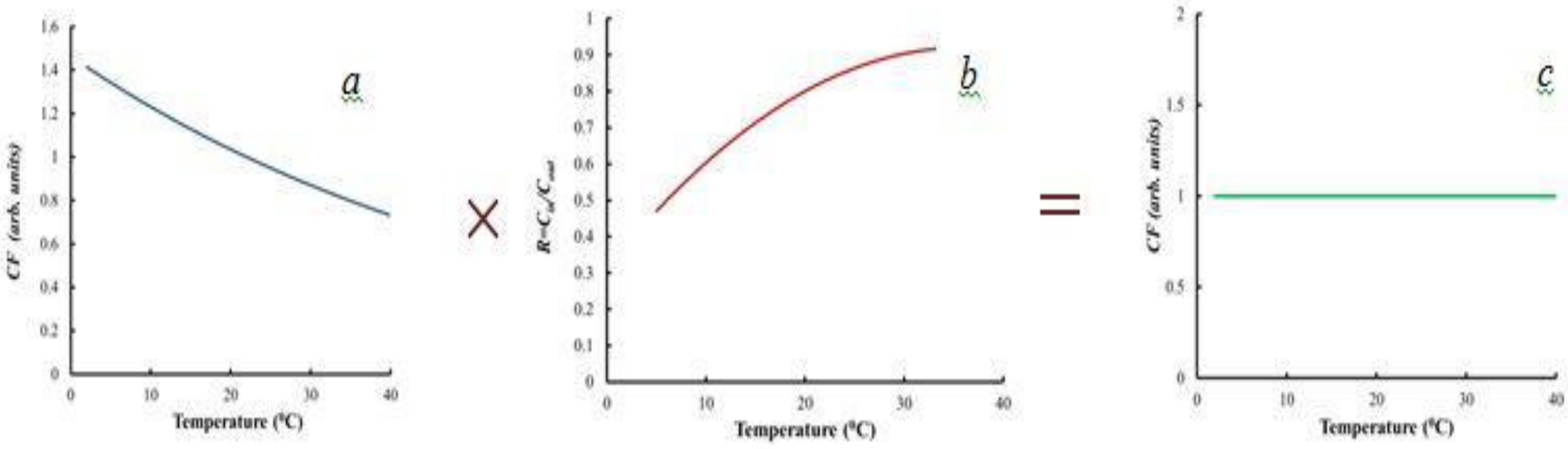


The temperature dependence might be altered by varying $\frac{hV}{S}$ and the polymer material. When $h \ll L_D$:

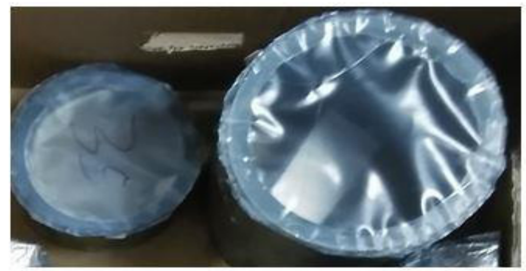
$$R = \frac{c_{in}}{c_{out}} = \frac{1}{1+\frac{\lambda h V}{SP}},$$

Theoretical modeling revealed, that by using different materials, ***h, V, S*** the temperature dependence of ***R*** can be somewhat reduced but not eliminated.

The concept behind the "compensation module"

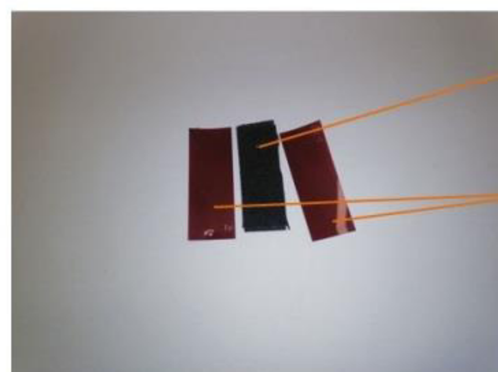


Pilot experiments



Kodak-Pathe LR-115/II

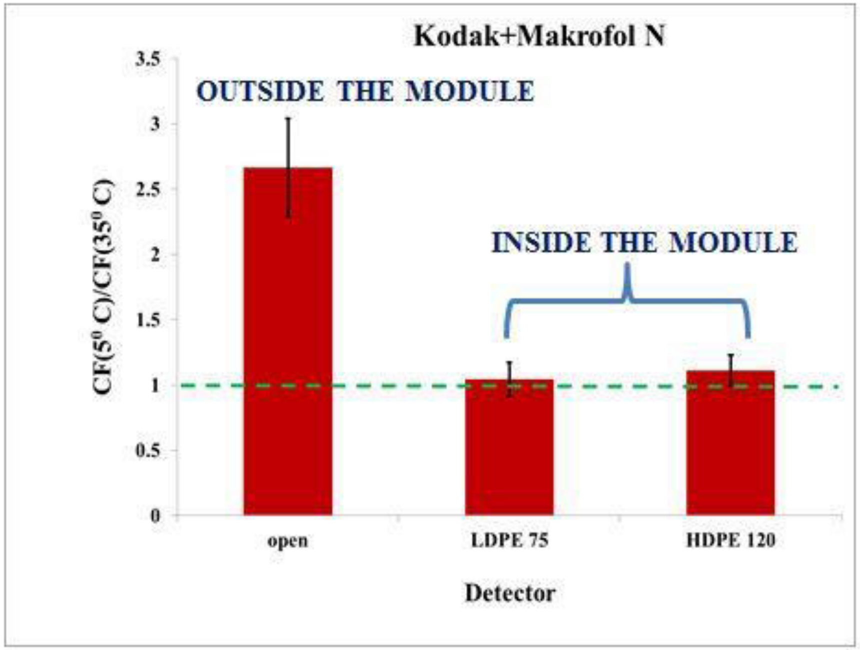
absorbers of 2×43 μm Makrofol N (Makrofol N is of unique high radon absorption ability and serves as absorber/radiator)

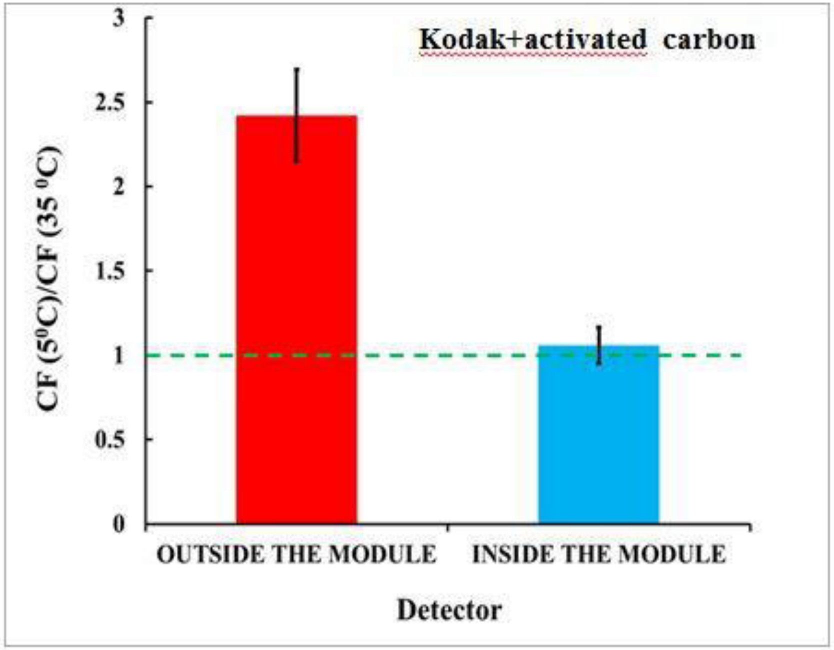


Sheet of activated carbon fibers

Kodak-Pathe LR-115/II

Experimental results





This can reduce the temperature bias + thoron interference + humidity influence

Thoron interference < 5%
Temperature bias: 10%

Thoron interference << 1%
Temperature bias: ~2-3%
and… no influence of humidity

Key outcomes of Task 2.3 (a)

- For active monitors for which fast reaction to rapidly changing concentrations is required a spectrometric discrimination between radon and thoron is probably the best option. For active monitors with active sampling, counting in two different time intervals can also be used;
- If spectral discrimination is not used but the active monitors are expected to have a fast reaction, a proper approach is to incorporate a delay line either within the instrument design or as supplementary module;
- For active monitors for which fast reaction to rapidly changing concentrations is not required, and which work in a diffusion mode, additional diffusion barrier can be used;

Key outcomes of Task 2.3 (b)

- For passive detectors diffusion barriers might be considered in the design and tested in the prototypes. If instruments are scheduled to work at high humidity we recommend diffusion barriers based on polyethylene foils of low density polyethylene;
- For monitors/detectors in which the usage of polyethylene packing is planned to reduce the thoron interference, a possible temperature bias may be introduced. Methods for handling and taking account of this bias are proposed in Deliverable 2.
- For detectors that have response decreasing with the increase of the temperature, by the new compensation module proposed (patent pending) **reduction/elimination of the influence of thoron + temperature + humidity can be achieved simultaneously.**

A2.3.5.

A2.3.5	Based on the results from A2.1.1-A2.1.2, A2.2.1-A2.2.3 and A2.3.1-A2.3.4, SUBG, IRSN, STUK, CEA, and BEV-PTP will write a report on the influence of thoron on radon monitors used in Europe including proposals for checking their sensitivity to thoron, and recommendations on the construction of radon monitors that are not sensitive to thoron together with technical approaches aimed at reducing thoron-related bias in the radon signal in existing monitors. Once the report have been agreed by the consortium, SUBG will send the report to the coordinator, who on behalf of IRSN, STUK, CEA, and BEV-PTP will then submit it to EURAMET as **D2** *'Report on the influence of thoron on radon monitors used in Europe including (i) procedures for checking their sensitivity to thoron, (ii) recommendations on the construction of radon monitors that are not sensitive to thoron and (iii) technical approaches aimed at reducing thoron-related bias in the radon signal in existing monitors'*.	**SUBG**, BEV-PTP, CEA, IRSN, STUK

Deliverable 2 accepted: June 2020

Deliverable 2 authors:

- **SUBG:** D. Pressyanov, K. Mitev, I. Dimitrova, S. Georgiev, Ch. Dutsov
- **IRSN:** N. Michielsen, S. Bondiguel, J. Malet
- **CEA:** P. Cassette, B. Sabot
- **STUK:** T. Turtiainen, O. Holmgren, R. Dehganzada
- **BEV-PTP:** M. Stietka, H. Wiedner

WP2: Research outcomes to date:

- **Papers in peer-reviewed journals:**
  - Georgiev S., Mitev, K., Dutsov, Ch., Boshkova, T., Dimitrova I., 2019. Partition coefficients and diffusion lengths of ^{222}Rn in some polymers at different temperatures. ***Int. J. Env. Res. Publ. Health***, 16, 4523.
  - Mitev K., Cassette P., Pressyanov D., Georgiev S., Dutsov Ch., Michielsen N., Sabot B., 2020. Methods for the experimental study of ^{220}Rn homogeneity in calibration chambers. ***Appl. Radiat. Isot.,*** 165, 109259
  - Pressyanov, D, Dimitrov, D., 2020. The problem with temperature dependence of radon diffusion chambers with anti-thoron barrier. ***Rom. J. Phys.,*** 65, art. 801.
- **Patent application submitted:**
  - Pressyanov, D., 2019. Bulg. Pat. Appl. Reg. Nr. 112897, priority: 19.03.2019 (assignee: Sofia University "St. Kliment Ohridski"), WIPO Appl. Reg. Nr. PCT/BG2020/000003.
- **One paper in peer-reviewed (open access) conference proceedings (33rd International AARST Radon Symposium, 2019);**
- **Two oral presentations and one poster presented on international conferences.**

Thank you!