RadoNorm
Managing risks from radon and NORM

Deliverable 2.2

General outline of the design of the reference fields and the measurement strategy for thoron decay products

Work Package 2

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Executive Summary

Due to its short half-life, the exhalation of thoron (Rn-220) from the ground and the migration of thoron into houses is usually negligible from the viewpoint of radiation protection - especially compared with the dominating contribution of radon (Rn-222) and its progenies to the dose. Several field studies reported a significant contribution of thoron exhaling from building material. Rather often and even in outdoor atmosphere, measurements of radon progenies (Po-218, Pb-214, Bi-214 and Po-214) give the evidence of thoron progeny (Po-216, Pb-212, Bi-212 and Po-212) being present at the same time. The half-life of thoron gas is short (56 s) but the half-lives of its progeny are far longer than those of radon.

According to Article 100, the Council Directive 2013/59/EURATOM requires EU member states to take appropriate actions to identify and evaluate existing exposure situations considering exposure from thoron and to determine the corresponding occupational and public exposures. Requirements from Article 103 equally apply to thoron exposure and warrant a strategic consideration of thoron and its progeny in national Radon Action Plans. The prerequisites for the identification, verification and calibration of appropriate radiation protection equipment for the determination of thoron / thoron decay products exposure are, compared to vast experiences with radon monitoring, less advanced.

In order to prepare for surveys of indoor thoron / thoron progeny concentrations at work or at home, metrological, structural and organisational capabilities for the thoron / thoron decay product calibration need to be established. The dose from thoron exposure is mainly due to the exposure to its progeny. Because of the short half-life of thoron, no uniform distribution of it can be expected, rather higher concentrations only close to the place of origin. However, the thoron progeny has significantly longer half-lives compared to the radon progeny and thus, in relation to the equilibrium equivalent activity concentration, the potential alpha energy concentration is higher by a factor of 14. The focus of a dose assessment clearly is on thoron progeny and their measurement.

Currently, no single EU member state country has the expertise or capabilities to provide the required infrastructure and competence for a thoron progeny calibration. To provide the access to calibration facilities to stakeholders in every country is at least questionable in the viewpoint of limited resources and may constitute unnecessary duplication. This pan-European project RadoNorm therefore pioneers the access to metrology capabilities and services across borders.

In the opinion of the consortium, the necessity to hold available reliable (traceable, quality-assured) measurement and thus evaluation of thoron exposures and its progeny in Europe remains. Therefore, the establishment of a thoron / thoron progeny reference calibration chamber following the DIN 61577-4 specification in a laboratory accredited to ISO / IEC 17025 is a prerequisite for the verification and calibration of instruments for the determination of thoron exposure.

The deliverable at hand summarises the current state of the design of the reference fields and the measurement strategy for thoron decay products that had been considered and discussed since the start of the project. The report gives a snapshot of the actual knowledge, discussed issues and further foreseen activities.

Table of content

List of figures

List of Tables

1. Introduction

The exposure of the public to the naturally occurring isotopes Rn-222 and Rn-220 (in this work also referred as “radon” and “thoron” respectively), is a critical aspect of legal metrology in Europe, as the European Directive 2013/59/EURATOM states to consider both isotopes in the dose assessment to natural radiations.

Although radon is more commonly encountered due to its longer half-life, the necessity for reliable and traceable measurement of thoron and its decay products remains essential. Unlike radon, where the properties of the ground beneath a building are the primary source of indoor air contamination, thoron's short half-life limits its migration to short distances. This means that building materials are the main contributors to thoron's presence inside confined spaces, potentially affecting just the materials used in nearby wall construction rather than the entire building. An increasing interest to know the contribution of certain building materials can be seen. To assess whether thoron poses a concern, it is important to determine the thorium activity concentration in building materials, or more effectively, the thoron exhalation rate. However, this approach can be challenging in existing buildings. A simpler method is to directly measure thoron gas activity concentration using a basic detector and, if needed, proceed with quality-assured and traceable methods.

Due to its short half-life of 55.6 seconds, thoron does not disperse evenly in the indoor space where it is released. Typically, high thoron concentrations are found only near its source, such as the walls. Consequently, it is difficult to establish a representative, averaged value of thoron concentration in indoor air during measurements. On the contrary, its decay products (Figure 1) have longer half-life time and have the time to diffuse more homogeneously in the indoor space.

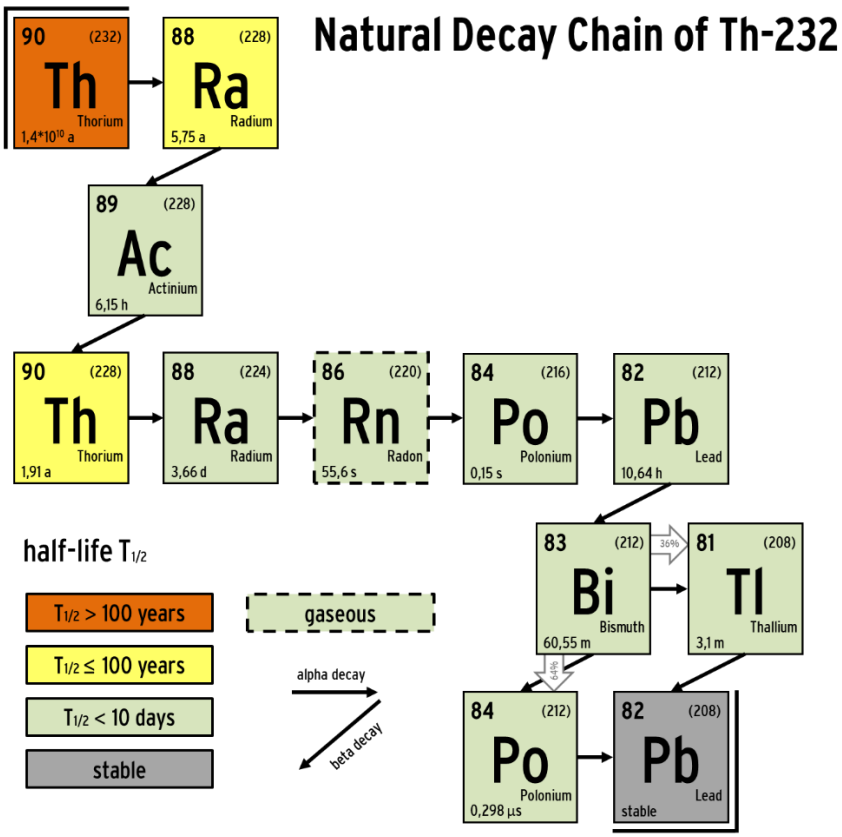

Figure 1 – Natural Decay Chain of Th-232 [BfS, Section Radon metrology]

The dose caused from an exposure to thoron and its decay products, as the radiobiological effects, are mainly due to the transfer of alpha energy from the decay of thoron gas decay products. So, when considering the hazards associated with thoron, the situation parallels that of radon. However, some differences exist. Radon's decay products follow the decay of the gas itself, whereas thoron decay products can persist beyond the complete decay of thoron gas. It is due to the fact that some of the thoron progenies have a longer half-life time: 10.64 h for Pb-212 and 60.55 min for Bi-212. Consequently, due to the decay law, secular equilibrium between thoron and its decay products cannot be achieved, making it impossible to calculate a reference equilibrium factor or to evaluate the activity concentration and potential alpha energy of thoron decay products based solely on thoron gas measurements. Only the direct detection of the decay products allows the direct dose calculation, without measuring or assuming an equilibrium factor between gas and decay products. Moreover, for thoron gas and decay products atmosphere, the equilibrium factor is inappropriate to describe a certain equilibrium as a large spatial and temporal variability in relation to the position of observance in a room needs to be accepted due to the short half-life of thoron. Moreover, the fraction of unattached thoron decay products differs significantly from that of radon decay products; the longer half-life of thoron decay product Po-212 increases its likelihood of attaching to aerosols, resulting in a relatively low unattached fraction.

To assess the significance of a given thoron / thoron decay product activity concentration for radiation protection, it is crucial to recognize that when radon and thoron have the same activity concentrations, the potential alpha energy concentration of thoron decay products, having the same activity as thoron, is 14 times greater than that of radon decay products being in secular equilibrium with radon. Notwithstanding that it is a very limited situation, due to, as discussed above, half-life ratios between thoron and its decay product, in specific cases, it is necessary to accurately measure either both thoron and its decay products or thoron decay products, at least, in addition to radon, for the determination and evaluation of radiation exposure due to both radon isotopes occurrence.

The establishment of a thoron decay products reference calibration chamber, adhering to DIN 61577-4, specifications and housed within a laboratory accredited to ISO/IEC 17025, traceable to national standards is central to ensure the verification and calibration of relevant measurement instruments. Such an infrastructure is essential for providing quality-assured data on thoron exposure necessary for compliance with the Council Directive. In the past, the German National Metrological Institute (NMI) Physikalisch-Technische Bundesanstalt (PTB) developed and operated a primary standard for the activity concentration of thoron in air [1] and also pioneered the concept of a German thoron progeny chamber [2]. Also, colleagues from French CEA and IRSN developed a new thoron atmosphere reference measurement system at the LNE-LNHB in the frame of the European MetroRadon project [3].

Despite the EURATOM mandate, the current metrological landscape in Europe lacks the comprehensive infrastructure required for thoron calibration [4]. No EU member state currently possesses all the requisite capabilities and resources independently, leading to potential inefficiencies and redundancies if individual countries attempt to develop such facilities alone. In response, the project RadoNorm is pioneering a shared approach to establish metrology capabilities and services across borders, thereby optimizing resources and expertise.

This deliverable aims to provide a general outline of the design of the reference fields and the measurement strategy for thoron decay products.

2. Reference atmosphere design

The accurate assessment and calibration of thoron decay products require a well-defined reference atmosphere, which serves as the baseline for experimental and measurement processes. In this report, by “reference atmosphere” is considered the ensemble of thoron decay products and influential

parameters that affect it, such as temperature, pressure, relative humidity, aerosols and air flows or turbulences. Monitoring and controlling these variables is essential to ensure the reliability and precision of thoron and its progeny measurements, particularly in the context of potential alpha energy concentration (PAEC) assessments. This chapter outlines the parameters that define the reference atmosphere alongside the methodologies employed to simulate and maintain consistent atmospheric conditions necessary for calibration. Additionally, it discusses the challenges and solutions associated with measuring thoron decay products, focusing on the integration of continuous sampling and real-time monitoring techniques to enhance measurement accuracy and instrument calibration.

2.1 Definition of the reference atmosphere

The “reference atmosphere” is the ensemble of thoron decay products and influential parameters that affect it, such as temperature, pressure, relative humidity, aerosols and air flows or turbulences. For each instrument, more calibration points are required, so it is necessary to create different reference atmospheres with different parameter values. For each calibration point, all the parameters must be homogeneous and stable for a defined period of time.

In this chapter the parameters that define the reference atmospheres are discussed.

2.1.1 Thoron decay products

To determine the PAEC of a thoron atmosphere, several radionuclides have to be considered for analysis, depending on the measurement technique and sampling method. Due to alpha and gamma radiation emitted by several radionuclides, it is possible to use spectrometric methods. While because of the alpha and beta or gamma radiation, a detection of decay products in samples with non-spectrometric methods is also conceivable, e.g., by using liquid scintillation counting (LSC) or thermoluminescence detectors (TLD). The following table gives a list of energies of Rn-220 and its short-lived decay products that could be considered for measuring with alpha or gamma spectrometers as well as for calculating the contribution to counts rate in case of LSC or TLD. The recommended method is based on continuous sampling and on-line measurement.

Table 1 – Basic data of thoron and its decay products [5]

Nuclide	Decay type	Absolute efficiency (%)	Energy (MeV)
Rn-220	α	~100.0	6.404
	X	0.114	0.550
Po-216	α	100.0	6.288
Pb-212	β	5.2	0.159
	β	82.5	0.3345
	β	12.3	0.574
	X	5.9	0.011
	X	27.9	0.080
	γ	43.3	0.239
Bi.212	α	25.1	6.051
	α	9.7	6.090
	β	55.5	2.254
	γ	6.6	0.727
Tl-208	β	24.5	1.293
	β	21.8	1.526
	β	48.7	1.803
	γ	6.3	0.277
	γ	22.6	0.511
	γ	84.5	0.583
	γ	12.4	0.861
	γ	99.8	2.614
Po-212	α	100.0	8.784

2.1.2 Influencing parameters

The parameters that can influence the thoron decay products atmosphere are the following:

- Aerosol parameters (especially amount and size distribution)
- Temperature
- Pressure
- Relative humidity
- Air flow (turbulence)

It is essential that these parameters are controlled and stay stable during the calibration period.

2.1.2.1 Aerosol

Shortly after their formation, thoron progeny nuclides rapidly interact with trace gases and vapours, occurring in less than one second, forming clusters that grow into particles approximately 1 nm in size, known as unattached progeny. Attachment of these particles to existing aerosols occurs within 1 to 100 seconds, resulting in attached decay products, as illustrated in Figure 2.

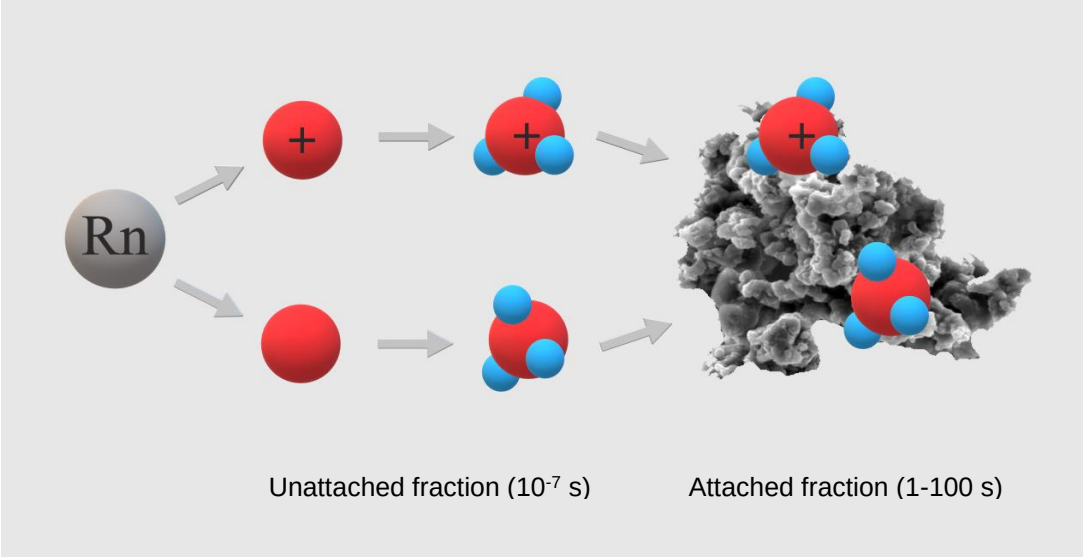

Figure 2 – Generation of radioactive aerosol

Data on the activity size distributions of the thoron decay products, Pb-212, are relatively limited. Due to the longer half-life of Pb-212 compared to the short-lived decay products of Rn-222, the aerosol size of attached Pb-212 is likely to be larger than that of Rn-222 decay products [6]. The longer half-life allows Pb-212 atoms to attach to aerosols during a longer period of time allowing aerosols to grow to larger particle sizes. However, measurements indicated that the median diameters of the accumulation mode for Pb-212 and the radon decay products Pb-214 are similar, at least in 'typical' indoor air. For this reason, in the next figures some data regarding radon are shown.

Furthermore, it must be taken into consideration that under different environmental conditions the aerosols size distribution can be slightly different. Both, outdoor and indoor aerosol sources can influence the size distribution. Figure 3 and Figure 4 show typical indoor aerosol distributions. In Figure 5 the distribution of aerosols size in an active coal mine is presented.

For the simulation of a typical indoor aerosol size distribution the Activity Median Aerodynamic Diameter (AMAD) has to be in the range of 0.2 µm with concentrations of 10^{10} particles/m^3 (between 10^8 to 10^{12} particles/m^3) [7].

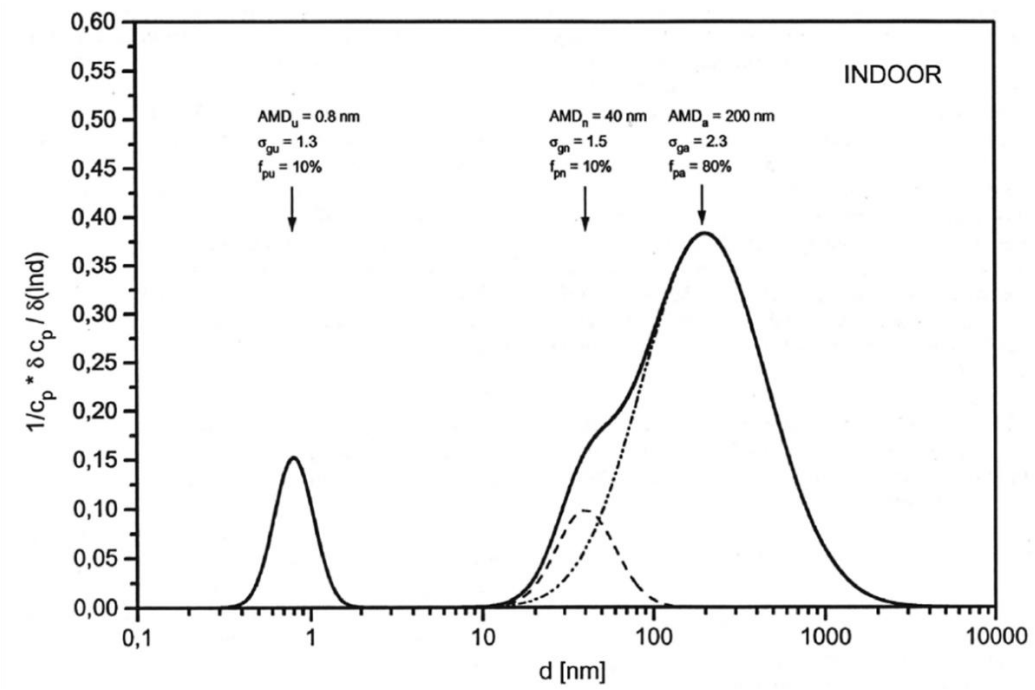

Figure 3 – Typical relative size distribution of the PAEC of radon decay products in indoor air without additional aerosols, taken from [8]

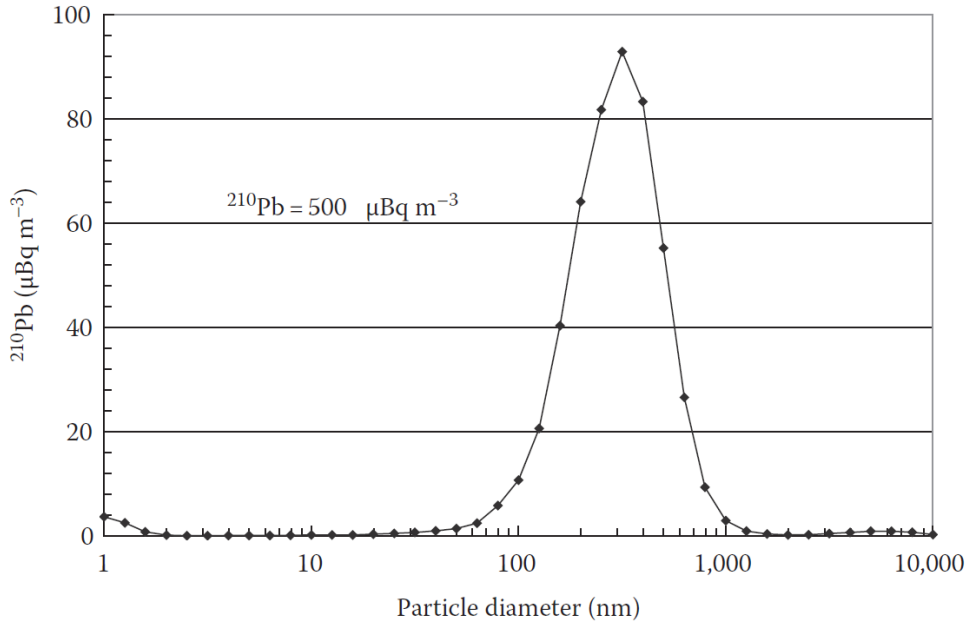

Figure 4 – Indoor aerosol particle distribution (dwelling) monitored via Pb-210 taken from [9]

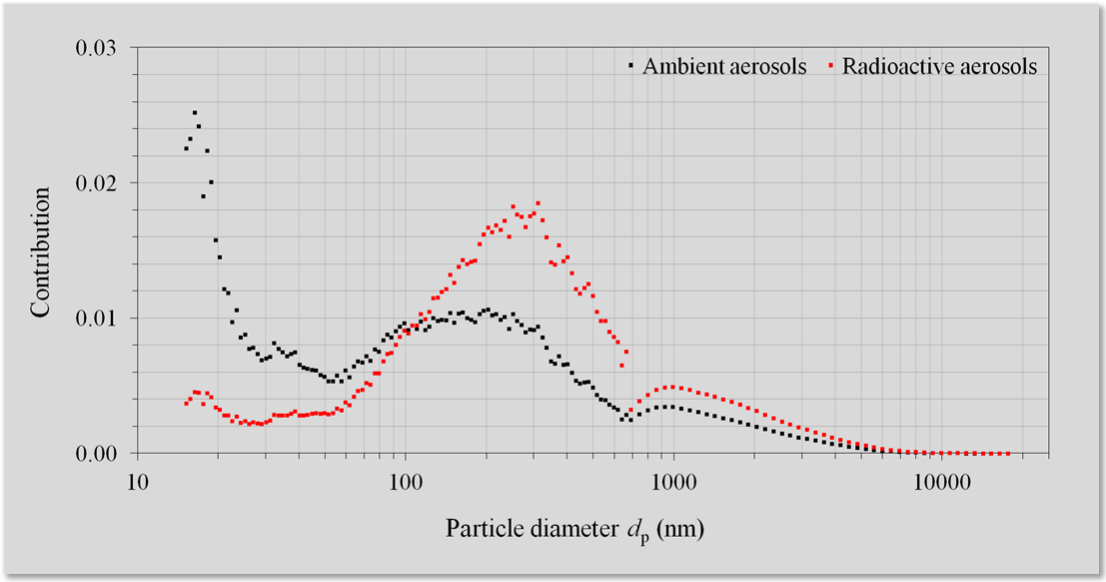

Figure 5 – The count and activity contributions of ambient and radioactive aerosols measured in an active underground coal mine close to the road head during heading machine operation for the aerodynamic shape factor of χ = 1.1, and the particle density of ρ_p = 1.4 g/cm^3 [10]

For experimental purposes, typically sodium chloride [11], carnauba wax [2] or incense [12, 13] is used for the generation of aerosol particles that recreates the typical indoor conditions (Figure 6).

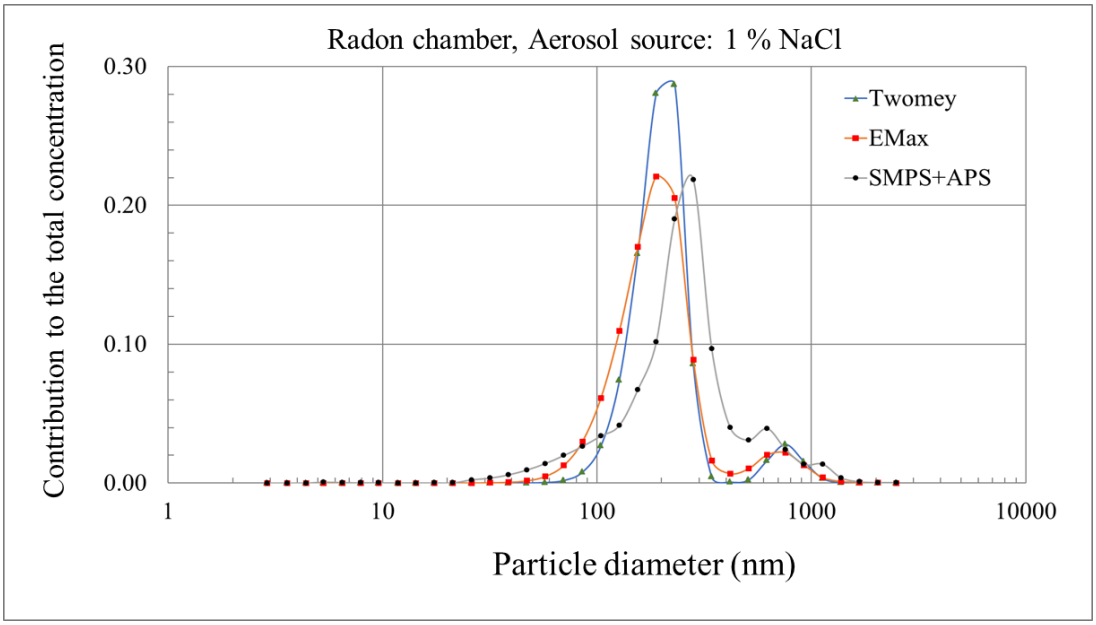

Figure 6 – The count and activity contributions of radioactive aerosols measured by different methods (Twomey, EMax and based on the data related to the size distribution of ambient aerosol SMPS+APS) in the radon chamber of the Central Mining Institute (GIG), Poland [14].

Sodium chloride as the primary constituent of sea salt accounts for a major fraction of the total atmospheric aerosol loading. Aerosol particles produced by nebulisation of aqueous NaCl solutions and subsequent drying are usually of cubic shape [15].

2.1.2.2 Temperature

Temperature has a significant influence on the behaviour of thoron and its decay products. As temperature rises, the diffusion of gases, including thoron, becomes more rapid, leading to a quicker and more uniform distribution of thoron within controlled environments such as calibration chambers. However, this same process requires vigilant monitoring to ensure consistent measurements throughout testing periods.

Furthermore, temperature variations can alter how thoron decay products adhere to aerosol particles by increasing their kinetic energy and movement. This change in behaviour is crucial when determining the potential alpha energy concentration. Additionally, changes in temperature can affect the deposition, or "plate-out," rates of thoron decay products on surfaces, as warmer conditions may modify material characteristics and airflow dynamics. Maintaining thermal stability is essential not only for the consistency of measurements but also for the calibration precision of the instruments employed.

2.1.2.3 Pressure

Pressure, by affecting air density, plays a critical role in thoron behaviour and its measurement accuracy. Increases in pressure lead to denser air, which generally slows the diffusion of thoron gas. This change can influence the uniformity of thoron concentrations in enclosed spaces such as calibration chambers, making it imperative to maintain appropriate pressure conditions.

The interaction between thoron decay products and aerosols is also impacted by pressure changes, as variations in air density can alter collision rates and attachment effectiveness, influencing overall energy concentration readings. Additionally, stable pressure is fundamental for ensuring the reliability and sensitivity of measurement tools, as fluctuations can compromise data integrity.

2.1.2.4 Relative humidity

Relative humidity (RH) directly impacts the prevalence and size of aerosol particles, which play a crucial role in the behaviour of thoron decay products. Elevated humidity facilitates aerosol growth, thereby enhancing the likelihood of decay products attaching to these particles, which can complicate precise measurement. On the other hand, low humidity levels may suppress aerosol formation, affecting attachment rates and potentially altering the decay dynamics within a system.

RH can also affect electrostatic interactions and the potential for aerosols to coagulate, both of which have implications for thoron decay products behaviour. Moreover, humidity levels can influence surface interactions, where high RH might increase decay products deposition due to condensation on surfaces, affecting the fidelity of results. Thus, maintaining consistent relative humidity is key to minimising uncertainties in measurement.

2.1.2.5 Ventilation

Ventilation is essential for maintaining a balanced environment for thoron and its decay products. Effective ventilation strategies distribute thoron evenly throughout calibration chambers, minimising the risk of localized concentration discrepancies and thus supporting the establishment of a consistent reference atmosphere for testing and calibration.

The rate of ventilation can significantly influence the accumulation or removal of thoron and decay products, impacting their equilibrium states and the overall decay process. While excessive ventilation may lead to variable conditions, insufficient ventilation could cause stagnation and misrepresentative concentration levels. Therefore, achieving an optimal balance in ventilation is crucial to ensure representational and reliable data collection.

2.2 Standards and recommendations

The EN 61577-4:2014 standard provides guidelines for the design and operation of equipment used to create reference atmospheres containing radon isotopes, specifically Rn-222 (radon) and Rn-220 (thoron), as well as their decay products. Therefore, this norm can be used as base for the formulation of the reference fields and the measurement strategy for thoron decay products.

The climate chamber must operate within the condition ranges specified in the standard EN 61577-4:2014, as detailed in the following table:

Table 2 – Calibration condition ranges specified in the standard EN 61577-4:2014

Influence quantity	Reference condition	Calibration condition
Temperature	20 ºC	18 ºC - 24 ºC
Relative humidity	65 %	40 % - 75 %
Atmospheric pressure	1013 hPa	860 hPa - 1060 hPa
Ambient gamma dose rate	-	< 0.25 µSv h^{-1}
Unattached fraction	-	< 0.25
Volume number of aerosols	10^{10} particles·m^{-3}	10^{8} particles·m^{-3} - 10^{12} particles·m^{-3}
Aerosol size (AMTD or AMAD)	0.2 µm	0.1 µm - 0.5 µm

Climate chambers capable of achieving broader ranges of environmental conditions are valuable for testing measurement instruments under non-standard, yet realistic, field conditions. This extended capability is exemplified in the EN 61577-4:2014 standard, which outlines the necessary specifications and requirements. The ranges are reported in Table 3.

Table 3 – Tests with variation of the influence quantities specified in the standard EN 61577-4:2014.

Influence quantity	Range of values
Temperature	- 25 ºC – + 50 ºC
Relative humidity	10 % – 100 % (condensing)
Atmospheric pressure	800 hPa – 1080 hPa
Ambient gamma dose rate	To be defined
Rn-222 activity concentration (for Rn-220 or Rn-220 DP measuring instruments)	10 $Bq \cdot m^{-3}$ – 1 $MBq \cdot m^{-3}$
Rn-220 activity concentration (for Rn-222 or Rn-220 DP measuring instruments)	10 $Bq \cdot m^{-3}$ – 1 $MBq \cdot m^{-3}$
EEC_{Rn-222} (Rn-220 measuring instruments)	2 $Bq \cdot m^{-3}$ – 1 $MBq \cdot m^{-3}$
EEC_{Rn-220} (Rn-222 measuring instruments)	2 $Bq \cdot m^{-3}$ – 100 $Bq \cdot m^{-3}$
Unattached fraction	up to 0.9
Aerosol concentration	10^{8} particles$\cdot m^{-3}$ – 10^{12} particles$\cdot m^{-3}$
Aerosol size (AMTD or AMAD)	10^{-3} µm – 10 µm
EEC: Equilibrium Equivalent Concentration *DP: Decay Products* *AMAD: Activity Median Aerodynamic Diameter* *AMTD: Activity Median Thermodynamic Diameter*	

2.2.1 ICRP 137

In the context of aerosol data for thoron decay products, ICRP Publication 137 can also be considered. Annex A of this publication details the aerosol characteristics (and dose coefficients) pertinent to thoron decay products.

The reference aerosol parameter values for indoor workplaces are shown in the following table:

Table 4 – Reference aerosol parameter values for indoor workplaces suggested by ICRP.

Exposure scenario	unattached fraction	Mode i	f_{pi}	AMTD or AMAD
Indoor workplace	0.02	nucleation	0.14	0.04 µm
		accumulation	0.86	0.2 µm
fpi: fraction of attached potential alpha energy concentration for mode i				

Of particular relevance to the design of a thoron decay products calibration chamber is the comparison between EIC 61577-4:2009 reference conditions and the aerosol parameters assumed by ICRP 137 [17]. The ICRP provides a comprehensive data set for these aerosol parameters, particularly concerning indoor environments. Notably, Po-212 is unlikely to be present in the nucleation mode due to its longer half-life, meaning it predominantly appears as attached decay products.

2.3 Measurand

2.3.1 PAE and PAEC

The potential alpha energy (PAE) ε_p is the total alpha energy emitted during the decay of products nuclides along the decay chain of Rn-220 (**Fehler! Verweisquelle konnte nicht gefunden werden.**) down to ^{208}Pb [16] and thus characterises thoron decay products atmospheres. The unit of potential alpha energy concentration is Jm^{-3}.
Thoron decay products includes Po-216, Pb-212, Bi-212, P-212 and Tl-208.They are contributing to the PAE except Tl-208, which subjects to beta decay. With $\varepsilon_{p,Pb-212} = \varepsilon_{p,Bi-212}$ the calculation follows [1]:

$$\varepsilon_{p,Rn-220} = \varepsilon_{p,Po-216} \cdot N_{Po-216} + \varepsilon_{p,Pb-212} \cdot (N_{Pb-212} + N_{Bi-212}) + \varepsilon_{p,Po-212} \cdot N_{Po-212}$$

where N is the number of the respective nuclides.

The potential alpha energy concentration (PAEC) C_p refers to the concentration of any mixture of thoron decay products nuclides contributing their own PAE per Becquerel released for complete decay.

$$C_{p,Rn-220} = \frac{C_{Po-216}}{\lambda_{Po-216}} \cdot \varepsilon_{P.Po-216} + \left(\frac{C_{Pb-212}}{\lambda_{Pb-212}} + \frac{C_{Bi-212}}{\lambda_{Bi-212}}\right) \cdot \varepsilon_{p,Pb-212} + \frac{C_{Po-212}}{\lambda_{Po-212}} \cdot \varepsilon_{P.Po-212}$$

where C is the activity concentration of the respective decay products (= measurand) and λ the radioactive decay constant.

Po-216 and Po-212 are each in radioactive secular equilibrium with their parent nuclide due to the short half-life:

$C_{Po-216} = C_{Rn-220}$

and

$C_{Po-212} = C_{Bi-212}(1 - p_\Sigma)$

with the transition probabilities p_k for the α-decays of Bi-212 with $p_\Sigma = \sum_k p_k$

Table 5 – PAE of the radionuclides Rn-220, Po-216, Pb-212, Bi-212 and Po-212. No uncertainties are provided in [17]

Radionuclide	PAE per atom [10^{-10} J] according to [16]	PAE per Bq [10^{-10} J] according to [17]	Share of total PAE per Bq according to [18]
Rn-220	0.0334 [ICRP 50]	2.65	-
Po-216	0.0233641 ± 0.00081 (k=1)	$5.1 \cdot 10^{-3}$	$7 \cdot 10^{-6}$
Pb-212	0.0125036 ± 0.00081 (k=1)	691	0.913
Bi-212	0.0125036 ± 0.00081 (k=1)	65.5	0.087
Po-212	0.0140754 ± 0.00071 (k=1)	$6.2 \cdot 10^{-9}$	$8 \cdot 10^{-12}$

Considering the relevance for the effective dose calculation, the contribution of Pb-212 is by far the most relevant for the potential alpha energy calculation, with a share of 0.913 of the total PAE [18, 19]. Thus, there is an option to only monitor the intake of Pb-212 [20]. The contribution of thoron gas inhalation is negligible [17]. Still, dose calculations in [17] consider the intake of Pb-212 as well as Bi-212 and Rn-220.

Under specific conditions, where there is a high Rn-220 activity concentration but a low amount of accumulated Pb-212 in the air due to a high ventilation rate with Rn-220 rich exhaust air that is often filtered to prevent Pb-212 inclusion, the high number of Po-216 atoms can theoretically be the main source of PAEC. However, this situation usually leads to a negligible contribution to PAEC because of the short half-life of Po-216 and the absence of decay product equilibrium. This is more likely to occur during the initial few hours after thoron begins to flow into the chamber (see figure 6). Consequently, for proper calibration, it is necessary to wait at least three days to ensure equilibrium and accurate measurement. Furthermore, a well-established way of sampling decay products from air is continuous or grab sampling, e.g., on a filter and / or wire-mesh with a known volume flow and duration. In case of taking a grab sample from a decay product atmosphere into a filter for measurements outside the atmosphere, no Rn-220 and no Po-216 will be observable in the sample anymore (see decay chain and half-lives, **Fehler! Verweisquelle konnte nicht gefunden werden.**). Po-216 with its short half-life would only be accessible in continuous sampling measurements within the atmosphere. As a consequence, in such special cases a method is necessary to include Po-216, which by definition is a part of the thoron PAEC. On the other hand, in typical indoor environments under less extreme conditions, the influence of Po-216 can be neglected.

2.3.2 Thoron dynamic and equilibrium factor

According to [17], the equilibrium factor for thoron and its decay products is the ratio of the PAEC for the actual mixture of the thoron decay products, to that which would apply assuming activity of decay products are equal to the activity of thoron. Hence, it is defined similar as for radon and its progenies, to some extent. However, due to the short half-life of thoron and the relatively long half-lives of its decay products, Pb-212 and Bi-212, as well as the branching in the decay chain, true radioactive equilibrium in an Rn-220 atmosphere cannot be achieved according to the decay law. Nevertheless, a "dynamic" equilibrium can be established if the reduction in thoron activity due to radioactive decay is balanced by a sufficient inflow of thoron into the chamber. This assumes an efficient source of thoron and accounts for the branching of Bi-212 decay. It's important to differentiate this dynamic state from radioactive secular equilibrium, as potential consequences of any interventions (discussed further in the section 3.2.2.) may differ. Over time, the plate-out of airborne particles onto surfaces means that later decay products from Pb-212 onwards are more likely to deposit on surfaces rather than remain airborne. The extent of this plate-out, and therefore the composition of decay products in the air, depends on factors such as chamber size, surface area, ventilation, and climate conditions.

When the inflow of thoron is stopped, e.g. after taking a sample of from the atmosphere, no stable ratios between the Rn-220 decay products can be observed within the sample either (Figure 7). However, a radioactive transient equilibrium is obtained between Pb-212 and Bi-212. The activity ratio between Bi-212 and Pb-212 is equal to about 1.11, while Bi-212 is in secular equilibrium with Po-212 and Tl-208 considering the relevant decay ratio.

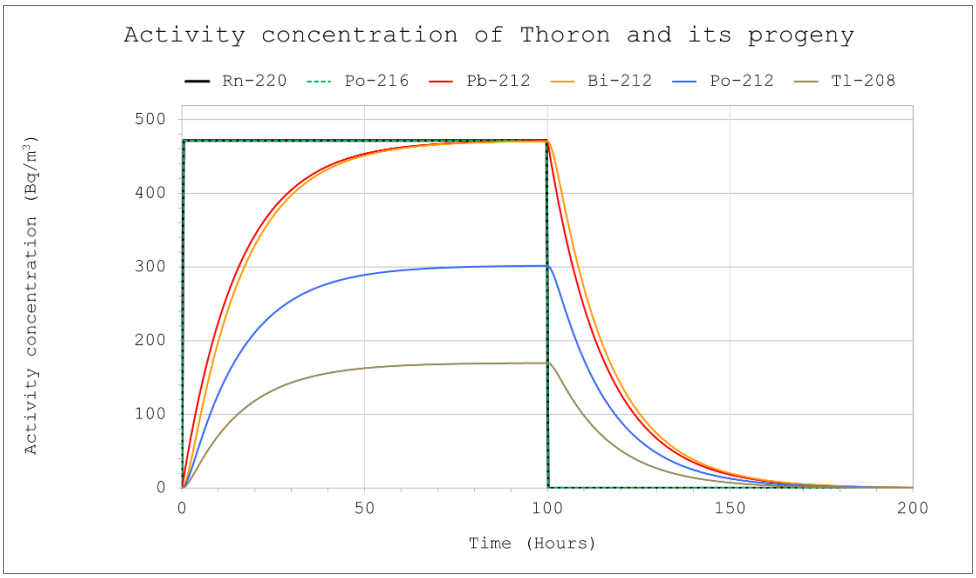

Figure 7 – Example of a thoron decay product activity concentration in a chamber when constant inflow of thoron gas is assured (up to 100 h) and when thoron supply was stopped (e.g. decay products have been collected on a filter). Thoron supply rate 12 Bq/s. Chamber volume 2 m³.

2.4 Atmosphere conditioning

To effectively calibrate an instrument, it is necessary to establish a homogeneous and stable reference atmosphere within the chamber for a certain amount of time, aligning with the chosen calibration method. For instance, a thoron decay products calibration chamber setup may consist of a climate test chamber designed to maintain constant environmental parameters over several days and that can be operated in continuous mode. In continuous operation mode, it is possible to make minor adjustments to climate parameters without opening the chamber, thereby preserving the reference atmosphere stability.

The following steps outline the calibration process using a chamber in continuous operation mode:

1. Climate Point Approach:
   The climate control system is activated upon setting the desired climatic point. Stable environmental conditions are essential for accurately evaluating thoron decay products and their interactions with aerosols within the chamber. Limited, controlled gas exchanges may occur to set target conditions within a specific timeframe, but preserving the gas composition's integrity is crucial. Once the desired climate point is achieved, aerosols and thoron gas can be introduced
2. Maintenance of Measuring Conditions:
   The climate test chamber maintains stable environmental conditions for periods ranging from 12 hours to 14 days. Temperature and humidity are kept constant, with necessary adjustments to aerosol concentrations. This stability is key to assessing thoron decay products as they interact with aerosols. Strategic aerosol redosing helps maintain the desired particle characteristics.
3. Switching Between Climate Points:
   Transitioning between climate points may require temporary gas exchanges to adjust humidity or temperature. These adjustments are carefully managed to preserve environmental integrity. Air releases, whether for drying or gas replacement, are directed to specialized exhaust systems to prevent contamination.
4. Release of the Atmosphere and Conclusion of Calibration:
   After calibration, as routine, the chamber atmosphere is methodically released through the designated exhaust system. Once the atmosphere is evacuated, the calibration process is officially completed.

Current advancements allow for the automation and software control of continuous chamber operations. Studies can be conducted on automated control of aerosol and thoron sources to further enhance efficiency.

The selection of atmospheric conditioning systems is guided by two key objectives:

- Minimising surfaces and volumes inside the chamber:
  The additional introduction of surfaces or volumes, such as in a pipe system with cooling and heating elements, would significantly complicate the efficient generation of the aerosol and thoron gas concentration in the chamber, since the aerosols and thoron decay products required in the measuring volume tend to adhere to surfaces (a phenomenon known as the plate-out effect). This it so be avoided during the climate measurement point. Circulation over closed longer airways would also lead to increased separation of the derived products and must be avoided.
- Avoiding the disruption of the reference atmosphere:
  Air injections and/or extractions must be avoided to keep a stable and uniform atmosphere during the calibration measurements, hence it is better to operate the chamber as a closed system (or as closed as possible). Strong air flows inside the chamber must be avoided too.

The system for aerosol control plays a vital role in maintaining the desired atmospheric conditions within the calibration chamber. Aerosols are essential for the study of thoron decay products, as they serve as condensation nuclei and influence particle characteristics. An effective aerosol control system ensures the desired concentration and distribution of aerosols, facilitating precise measurements and reliable experimental results.

3. Calibration chamber design

The design of a calibration chamber for thoron decay product measurement requires careful consideration of various factors to ensure the integrity and accuracy of the reference atmosphere.

Two key factors in the choice of which systems are better for this scope, are the following:

- Minimise surfaces and volumes (objects) inside the chamber:
  thoron decay products tend to adhere to walls, so that they are not available to be measured (plate out effect), so it is necessary to minimise them.
- Minimise elements that could disrupt the atmosphere:
  avoiding air flows or using an open system creates variations in the reference atmosphere and this must be avoided as much as possible.

3.1 Dimensions

When choosing the dimensions for the construction of a thoron chamber the following points must be taken into consideration:

- Control of homogeneity:
  a compact volume is advantageous for maintaining a uniform reference atmosphere. Smaller chambers facilitate precise regulation of thoron decay products concentrations and environmental parameters, such as temperature, pressure, relative humidity, aerosol distribution, and airflow.
- Independence from wall conditions:
  the chamber must be sufficiently large to minimise the influence of wall conditions, particularly the plate-out effect where aerosols adhere to surfaces. This ensures that the reference atmosphere remains consistent and unaffected by the proximity to chamber walls.
- Accommodation of equipment:
  adequate space is necessary to place and house both the reference instruments and the devices undergoing calibration, alongside any supplementary equipment like chargers and

pumps. This prevents interference among instruments and allows for unobstructed operation and accessibility.

Therefore, striking an optimal balance between a compact volume and a sufficient space is essential. Additionally, the dimensions of the chamber are influenced by the available space in the calibration laboratory, as well as the logistics of how personnel plan to access and arrange the instruments before and after the calibration period.

3.2 Materials

When selecting materials, it is crucial to consider the following key criteria:

- Permeability: to preserve the integrity of the reference atmosphere, it is essential to use materials with low permeability to thoron. This reduces the risk of gas leakage, which could undermine the accuracy of the measurements. Suitable materials include stainless steel with metal-based seals (such as copper or aluminium) or seals made from FPM/FKM, PVC, PTFE, and PUR. Silicone must be avoided as a sealing material due to its permeability to radon and thoron.
- Chemical Resistance: materials must withstand chemical interactions with thoron decay products to ensure that environmental conditions remain stable and measurement data are not affected by material degradation. For example, to prevent corrosion of the chamber walls due to humidity, a material with corrosion resistance class ‘CRC II’ could be employed.
- Surface Smoothness: since thoron decay products and aerosols tend to adhere to surfaces (plate-out effect), thereby reducing the amount of detectable thoron decay products, it is advisable to minimise not only the surface area but also the roughness of materials used.

3.3 Simulation model

3.3.1 Characterization of atmosphere/processes inside a chamber from the calibration process perspective

The calibration process is determined by such factors as the chamber capacity, measurand and the efficiency of the measurand source. Calibration may apply to devices designed to measure only the concentration of thoron or to measure the potential alpha energy concentration (PAEC) or nuclides that are its decay products. In the case of thoron, a single injection of this nuclide is insufficient. Its constant supply is the necessary solution. Moreover, for chambers with smaller volumes, the conditions inside can be strongly influenced if the calibration involves active devices equipped with filtering systems that will decrease the activity concentration of thoron decay products. Greater stabilization of conditions can be achieved in larger chambers, but this entails the need to use sources with higher efficiency (activity). However, other factors as losses made by diffusion, particle inertia and gravitational settlement can also reduce the nuclide concentration. The model can be used for overall designing of a chamber, when the process is starting from the scratch or for simulating necessary border conditions during a calibration process.

3.3.2 Model

In order to assess the calibration conditions that can be achieved in a given facility, a mathematical model was developed for simulation of either thoron and its decay product activity concentration inside a chamber or PAEC of thoron decay products considering different boundary conditions. The model is based on set of differential equations describing successive decay, the efficiency of the source, method of parent radionuclide delivery (continuous/single injection), equilibrium ratio between a parent radionuclide and decay products at the delivery moment, the volume of the calibration chamber. There are two options considered, the closed chamber and flow-through chamber. Additionally, the possibility of reducing the activity concentrations of nuclides inside a chamber, e.g. as a result of the operation of filtering devices inside characterized by specified flow rate and filtering efficiency. The model assumes

that the nuclides inside the chamber are evenly distributed. However, it does not consider losses that may arise as a result of diffusion, gravitational settling or inertia. The details of the model are presented in the publications [21] and [22].

3.3.3 Examples of model application – PAEC and radionuclides activity concentration in a closed space

The first simulation is focused on prediction of PAEC and thoron and its decay product activity concentration in a closed chamber. Figure 8 shows a situation where, as a result of a single injection into the chamber, the initial concentration of thoron was 1 kBq/m^3. Despite the relatively high concentration of thoron, the concentration of potential alpha energy did not reach high values and systematically decreased. Focusing on thoron and its decay product activity concentration in such situation there is possibility to observe two typical cases of radioactive equilibrium, i.e. no radioactive equilibrium when thoron decayed completely but the decay products still exist and, when due to the ratio of the decay constants of Pb-212 and Bi-212, the transient radioactive equilibrium can occur between these nuclides, when the activity of bismuth reaches the activity of lead, after a period exceeds it, finally reaching the stable ratio Bi-212/Pb-212 and then decays further approximately according to the half-life of lead (Figure 9).

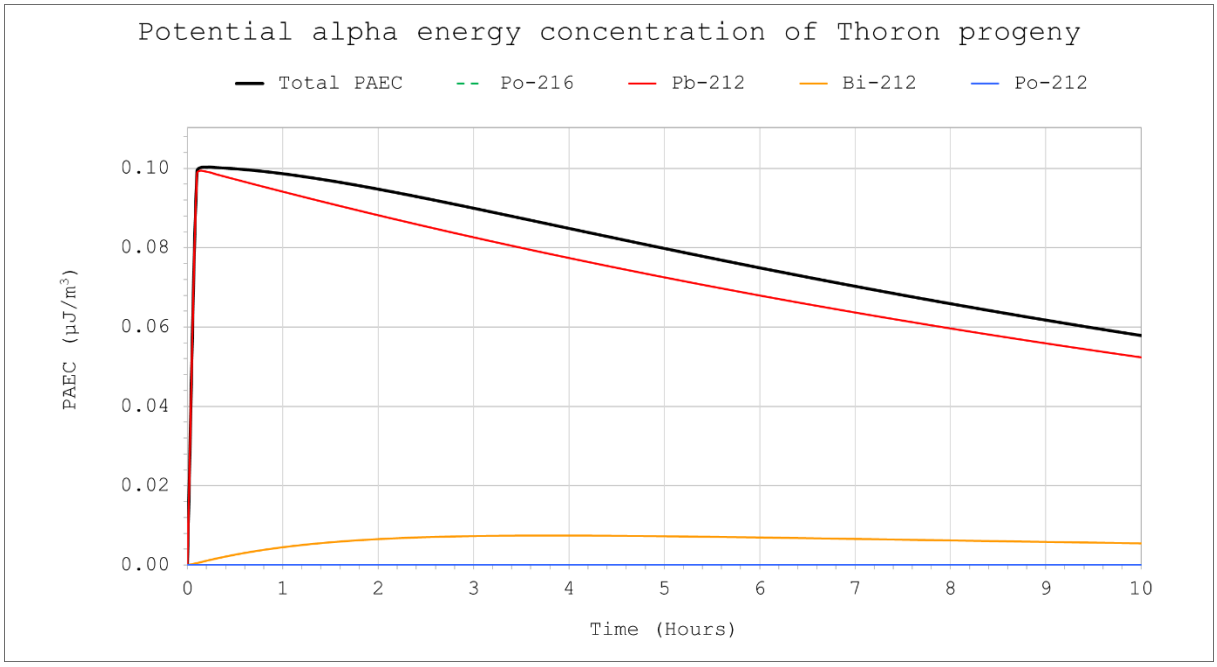

Figure 8 – Single injection of thoron. Initial thoron activity in the chamber: 1 kBq/m^3

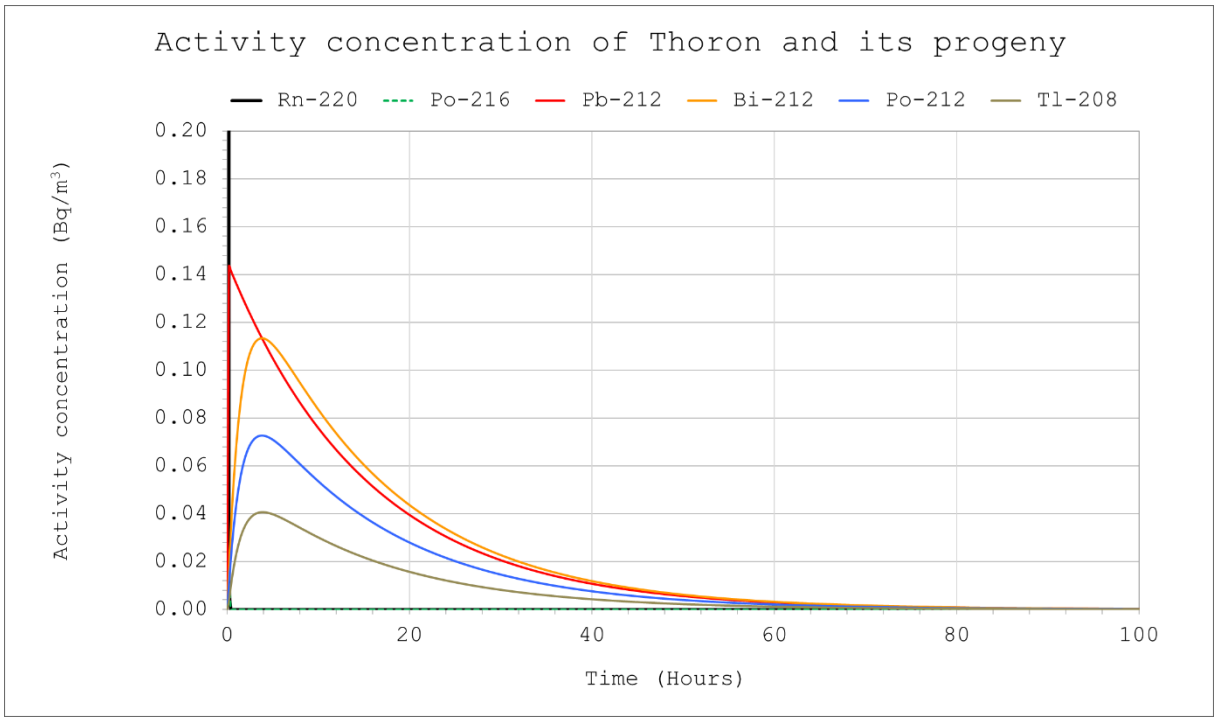

Figure 9 – Transient equilibrium. Single injection of thoron resulting in the initial thoron activity concentration of 100 Bq/m³

The situation is different in the case of systematic introduction of thoron to the chamber. Such an example is illustrated in **Fehler! Verweisquelle konnte nicht gefunden werden.**10 and **Fehler! Verweisquelle konnte nicht gefunden werden.**11 (for PAEC and radionuclides activity concentration, respectively), where thoron flows into a chamber with a volume of 1 m³ at a rate of 1 Bq/s. In such situation the decrease of thoron activity due to radioactive decay is compensated by the thoron inflow, hence in the chamber some activity of thoron still exists. This created a possibility for thoron decay products get equilibrium with thoron likely as radioactive secular equilibrium (Figure 11). However, it must be underlined that it is a dynamic situation, determined by the thoron inflow and strictly valid for confined spaces with small volumes. The difference is that while radioactive equilibrium (no matter secular, transient or no equilibrium) is ruled by the decay law and does not depend on external circumstances at all, the dynamic equilibrium depend, in general, on current local circumstances. What would happen when conditions are changed e.g. inflow of thoron is stopped is presented in the **Fehler! Verweisquelle konnte nicht gefunden werden.**2. For this example, the activity concentration thoron and its decay products under equilibrium conditions is reaching 80 Bq/m³ and the PAEC reaches a maximum value of about 6 µJ/m³ after approximately 80 hours. This time depends neither on the volume of a chamber nor on thoron rate inflow, however, the maximum concentration would be proportionally lower for larger objects (for a chamber with a volume of 20 m³ it would be 0.30 µJ/m³).

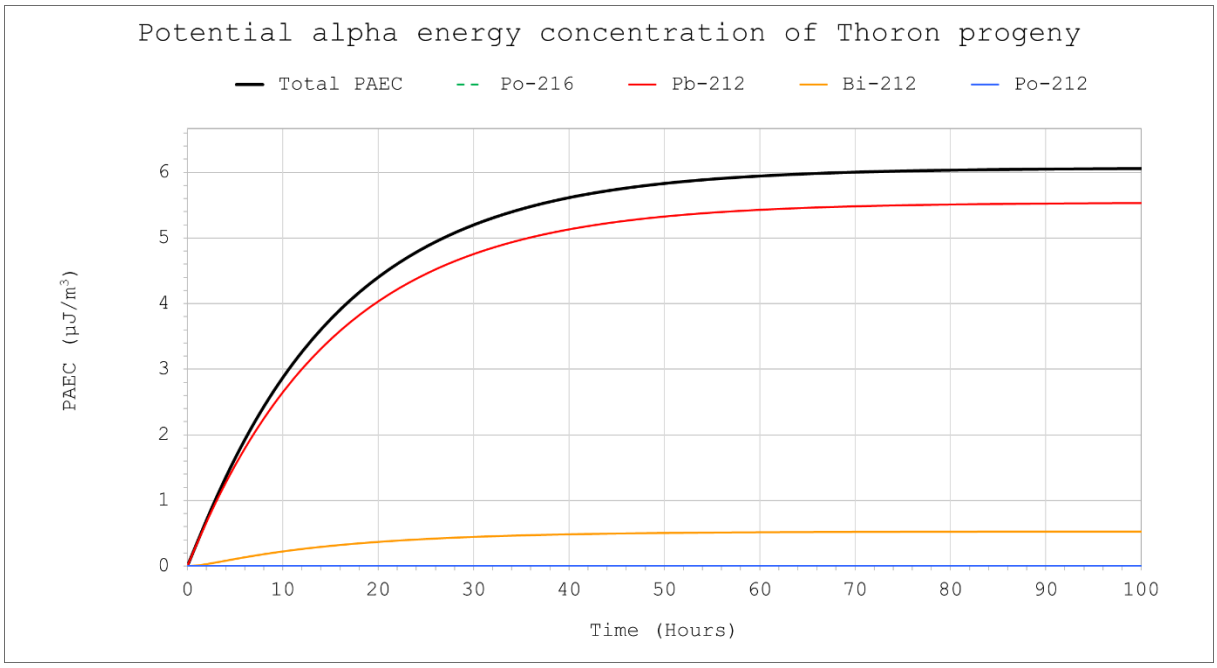

Figure 10 – Thoron decay products PAEC in a chamber, stable inflow of thoron from outside with efficiency of 1 Bq/s to the 1 m³ chamber

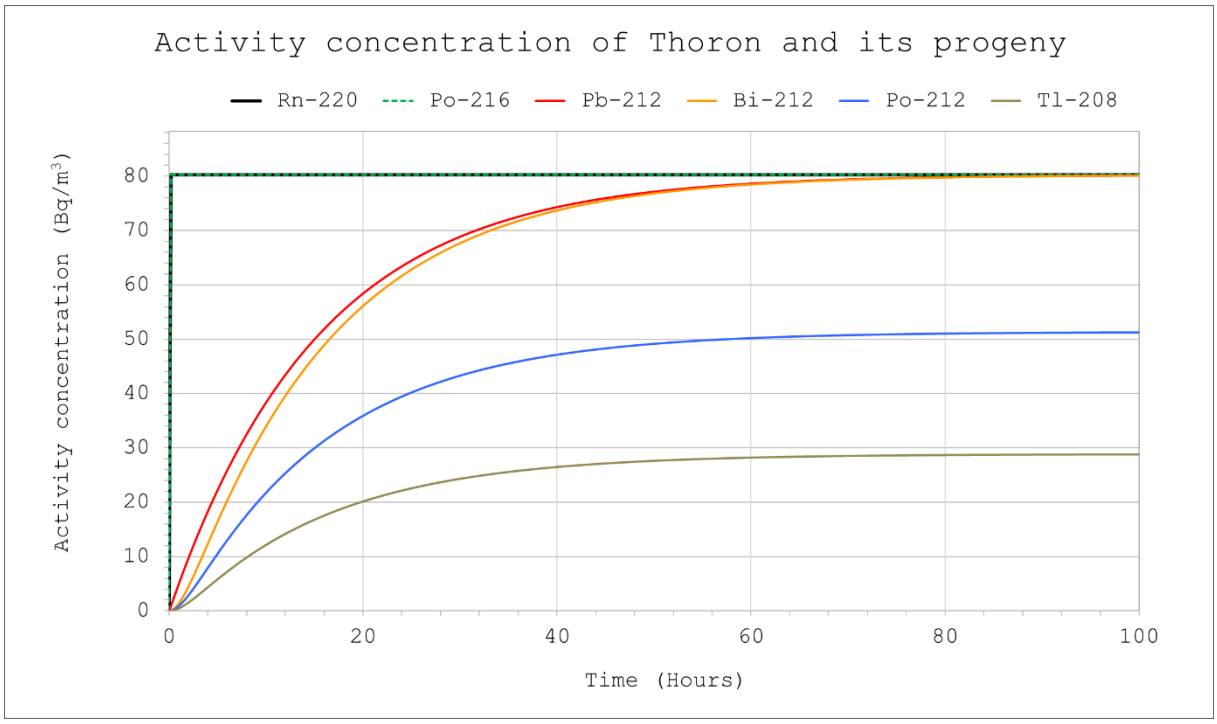

Figure 11 – Thoron and its decay products activity concentration in a chamber, stable inflow of thoron from outside with efficiency of 1 Bq/s to the 1 m³ chamber

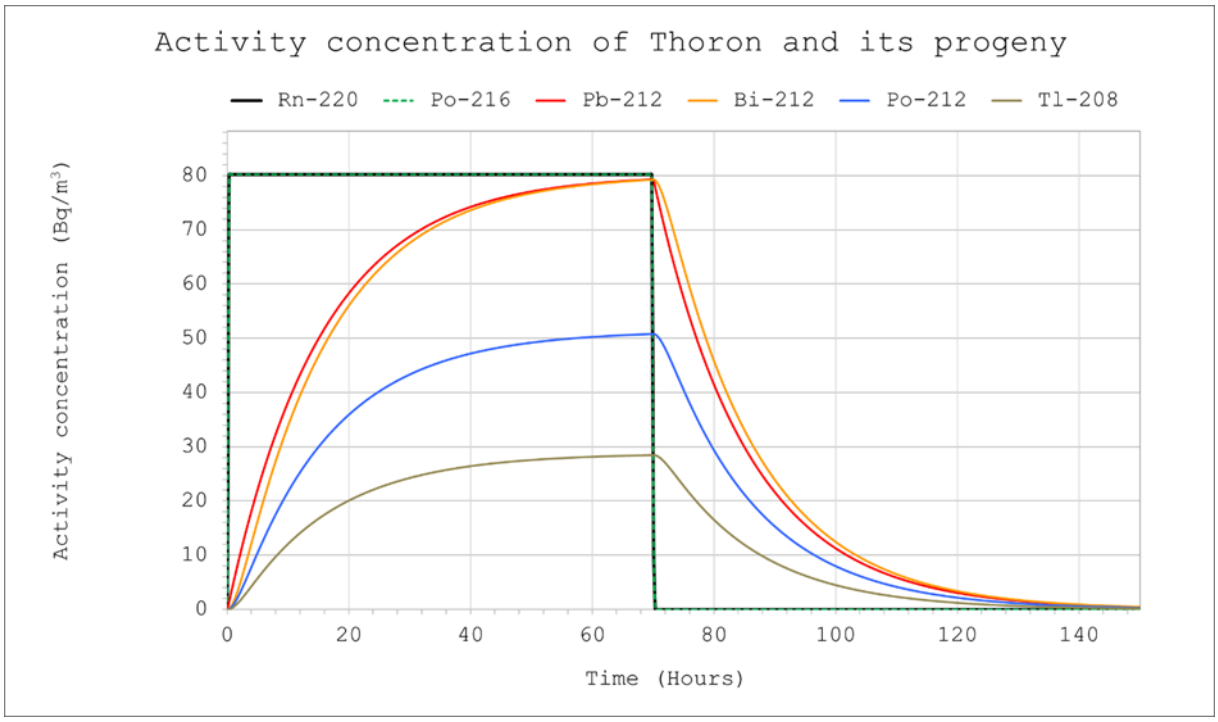

Figure 12 – Chamber volume: 1 m³, no active device, thoron inflow of 1 Bq/s was stopped at the seventieth hour

The presence of filtering devices (an active instrument calibrated) inside a chamber will reduce the concentration of potential alpha energy, to a greater extent for smaller objects. This is shown in Figure 13 and Figure 14 where it is assumed that there is a device inside the chamber equipped with filter and operating at a flow rate of 5 l/min.

All calculations and tasks related to the use of the model can be performed using an Excel file, attached to the publication: *Application_Closed_system.xlsm*

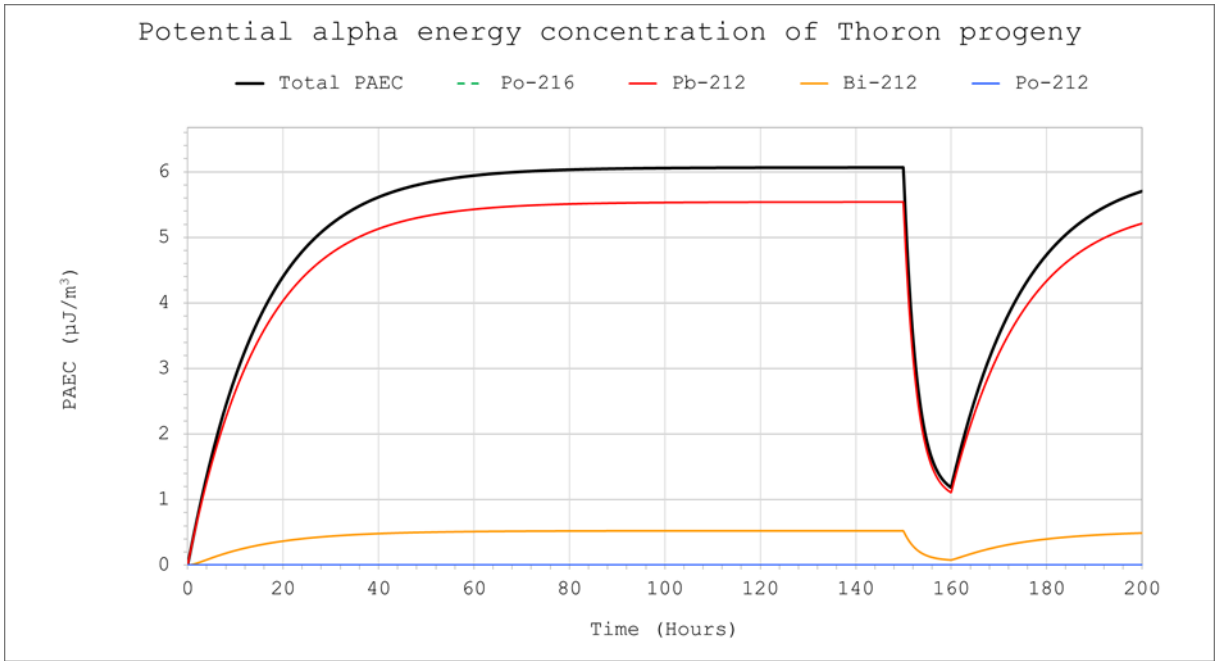

Figure 13 – Effect caused by an active device calibrated, chamber volume: 1 m³, flow rate of the calibrated active device inside the chamber equal to 5 l/min, pumping period between 150-160 hour with the filtration efficiency 100%, thoron inflow: 1 Bq/s

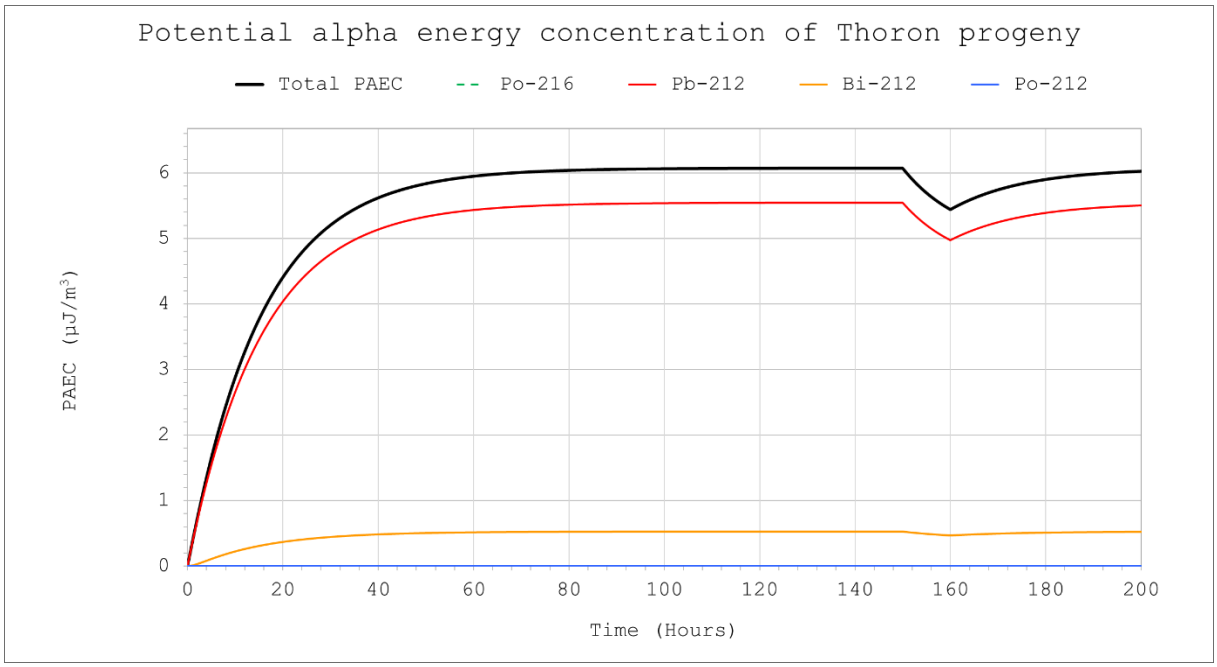

Figure 14 – Chamber volume: 20 m³, flow rate of the active device 5 l/min between 150-160 hour with the filtration efficiency 100%, thoron inflow: 20 Bq/s

3.3.4 Examples of model application – a flow-through chamber

For a flow-through chamber the model allows one to predict the effect on radionuclides activity concentration as well as PAEC when a chamber is ventilated. An example below shows the evolution of the thoron and progenies activities concentration in a chamber under conditions of: the chamber volume 10 m^3, thoron source is inside a chamber, thoron inflow 1 Bq/s, there are neither radionuclides inside the chamber at the beginning nor in inflowing air, ventilation and thoron inflow are launched in the same time. Simulation was done for different ventilation rates, relatively small when compared with the assumed chamber volume. Under such conditions, the ventilation process efficiently decreases Pb-212 and its decay products activity concentration while the impact on thoron activity concretion is negligible (**Fehler! Verweisquelle konnte nicht gefunden werden.**5, **Fehler! Verweisquelle konnte nicht gefunden werden.**6, **Fehler! Verweisquelle konnte nicht gefunden werden.**7, **Fehler! Verweisquelle konnte nicht gefunden werden.**8). This is changing calibration conditions significantly. By the way this simulation is showing how efficient ventilation can be considered as a mitigation of thoron related hazard.

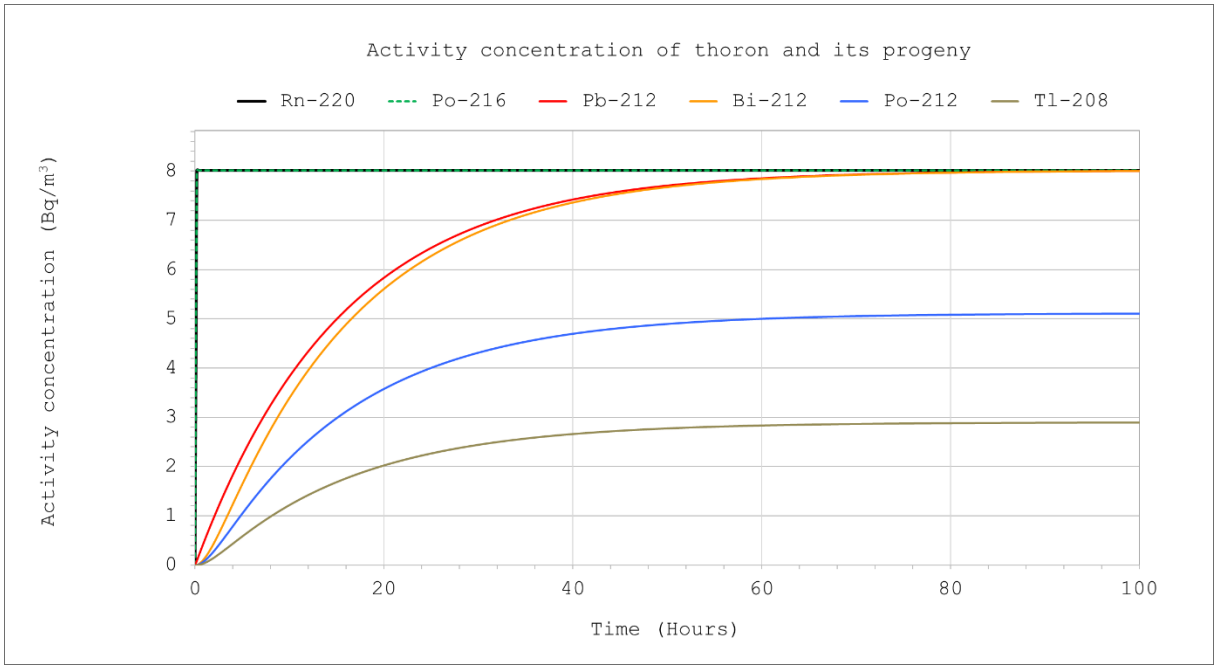

Figure 15 – Flow-through chamber volume: 10 m^3, no ventilation

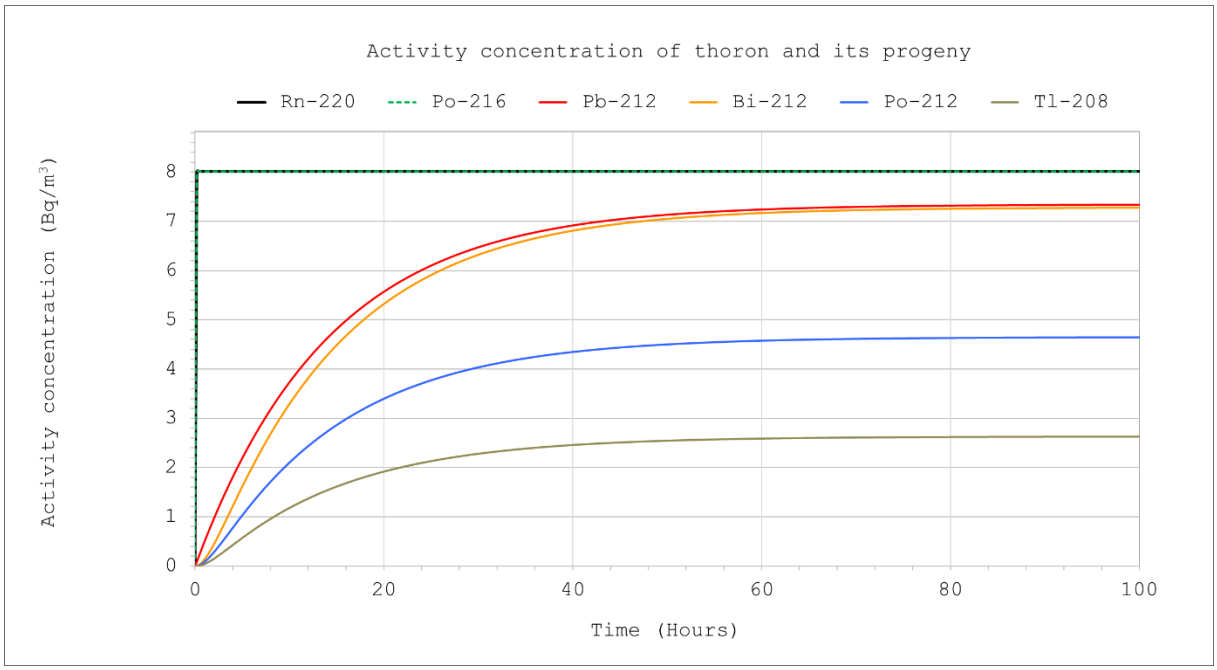

Figure 16 – Flow-through chamber volume: 10 m³, ventilation rate 1 l/min

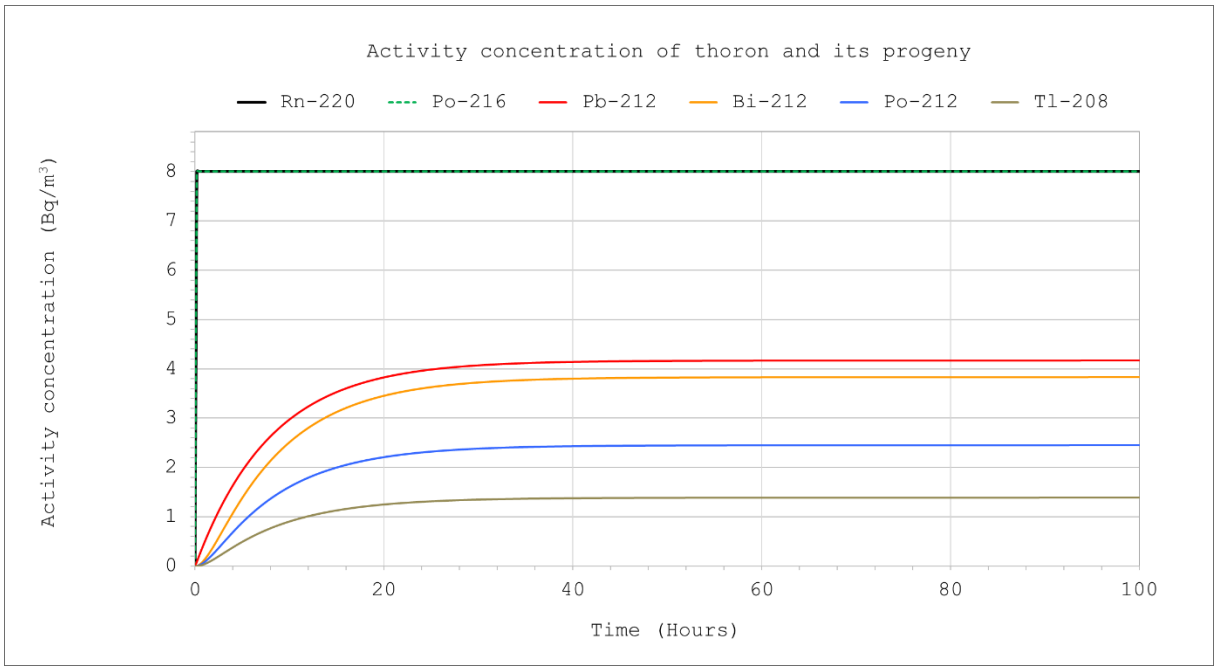

Figure 17 – Flow-through chamber volume: 10 m³, ventilation rate 10 l/min

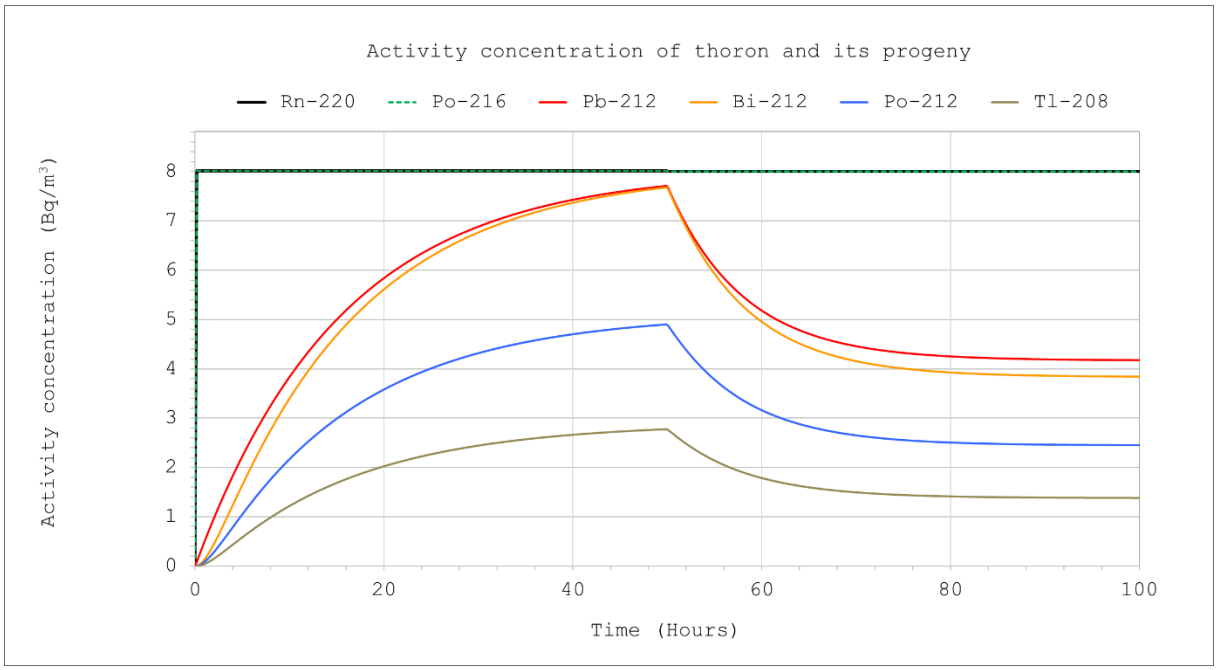

Figure 18 – Flow-through chamber volume: 10 m³, ventilation rate 10l/min, ventilation started 50 hours after the thoron inflow has been launched

3.3.5 Examples of model application – thoron source efficiency evaluation

Making some iterations using the model there is a possibility to evaluate necessary thoron inflow to get the assumed thoron and thoron decay product activity concentration in a chamber with a given volume. Evaluated, in this manner, thoron inflow into a chamber 5.5 m³ volume to get finally measurand activity concentration at the level 10 kBq/m³ should be about 720 Bq/s. Under such conditions, after about 0.1hour thoron activity (being in secular radioactive equilibrium with Po-216) reaches 10 kBq/m³ (**Fehler! Verweisquelle konnte nicht gefunden werden.**9). To get the dynamic equilibrium with Pb-212 and its decay products one has to wait at least three days (**Fehler! Verweisquelle konnte nicht gefunden werden.**20).

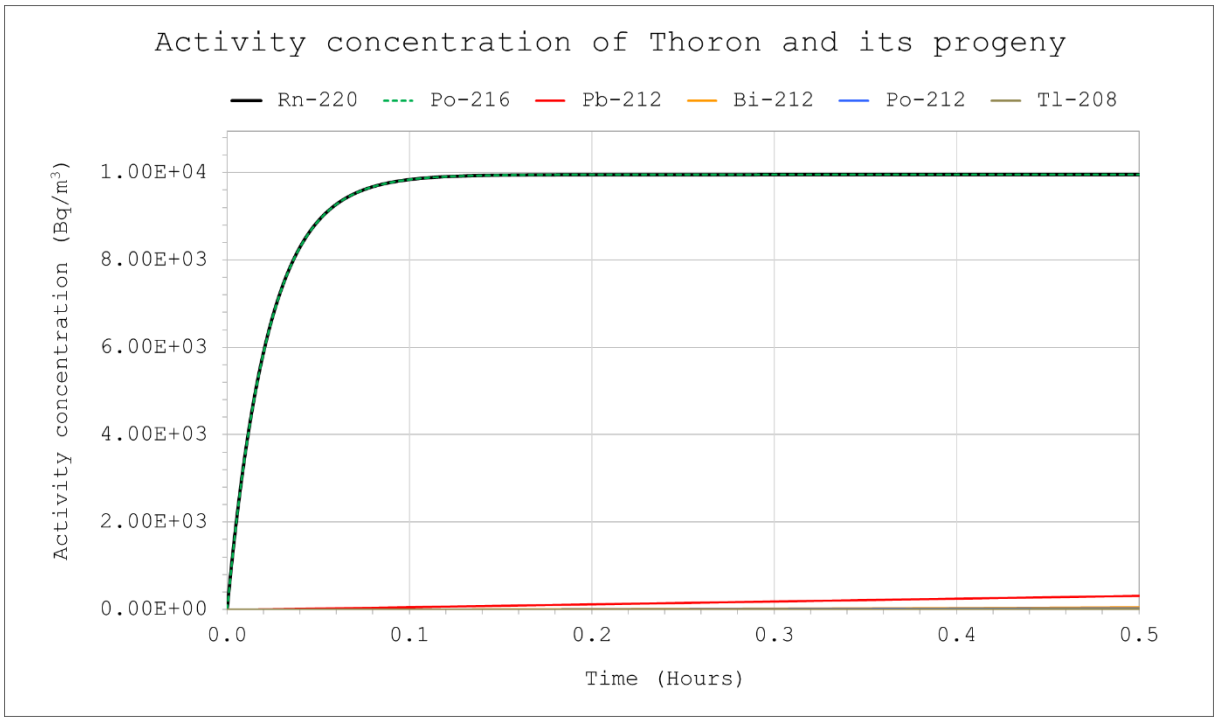

Figure 19 – Thoron activity concentration ingrowth, thoron inflow rate 720 Bq/s chamber volume 5.5 m³

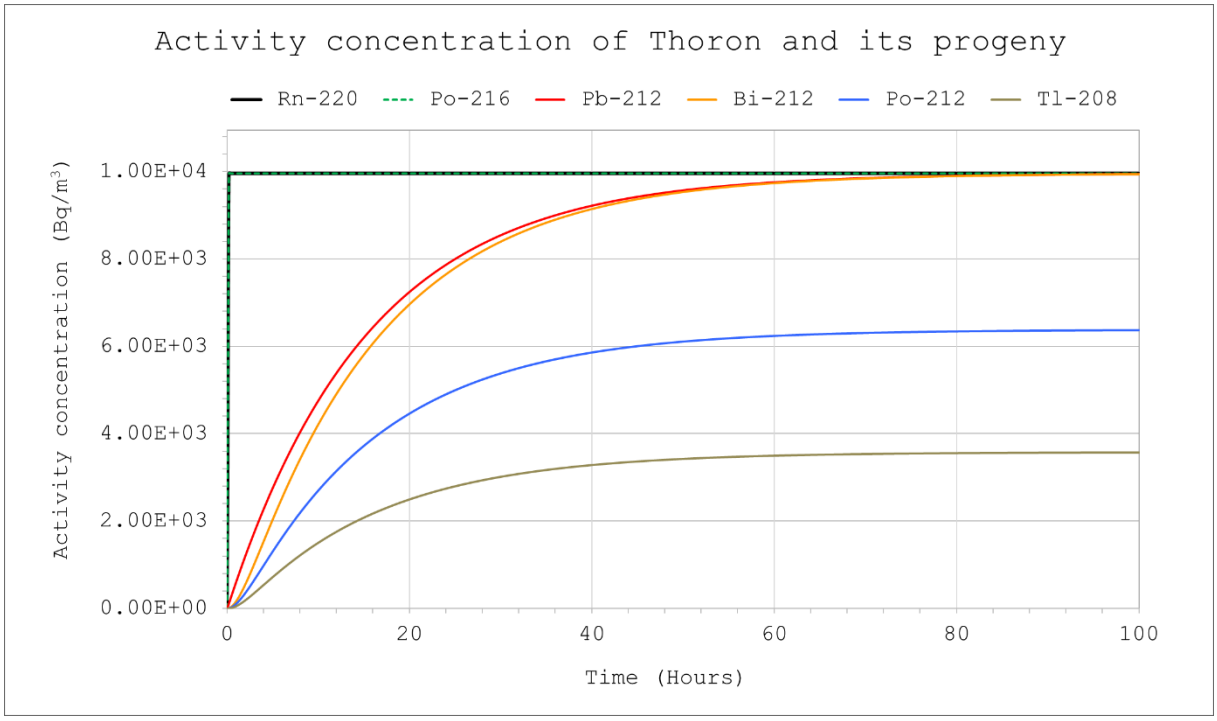

Figure 20 – Thoron and its decay product ingrowth - thoron inflow rate 720 Bq/s, chamber volume 5.5 m³

Assuming the use of thoron source as liquid solution of thorium chloride, where the air is pumped through, evaluation of Ra-224 activity concentration to get such a thoron inflow rate needs many parameters to be properly calculated. The efficiency of a thoron source in form of liquid solution expressed in Bq/s, based on measurements carried out in GIG (see the section 4.2.1), can be empirically evaluated as a source of total activity in Bq (as Ra-224) divided by 1000, assuming a source volume about 1 L and air flow through the source 2 L/min. Hence, for the above example the source activity should be at the level of 70 kBq of Ra-224 as direct parent thoron radionuclide. In practice thoron source must consist of Th-228, at least.

3.4 System for temperature control

For temperature control, as with any environmental control system, it is essential to select a system that maintains environmental stability without disrupting the reference atmosphere through unnecessary air injections or fluxes. The system must prioritize minimal interference with the reference conditions to ensure the accuracy and reliability of thoron decay products measurements. For this reason, the temperature control can be achieved using a jacket temperature control system placed on the inner walls. This design minimises additional surfaces and air fluxes, to prevent unnecessary influences on the chamber's climate.

3.5 System for pressure control

Pressure compensation mechanisms must be integrated via a valve system connected to room air and special exhaust air. The system can be controlled automatically, allowing valve adjustments only when necessary during conditioning operations to maintain chamber integrity. The system is engineered to avoid excess air movement or injections, thereby preserving the homogeneity of the reference atmosphere.

3.6 System for relative humidity control

An external conditioning unit can be used during the preconditioning phase for humidification and dehumidification. Once the chamber reaches the desired conditions, the unit is detached to maintain a stable reference atmosphere without altering the thoron concentration. Preferred solutions include a steam humidifier for humidification and a condensation dryer for dehumidification.

3.7 System for aerosol control

Aerosols can be generated using nebulization techniques to produce particles with specific size distributions and concentrations. The system can employ atomizers to disperse a dilute NaCl solution into the chamber atmosphere. Continuous monitoring of aerosol concentration and size distribution can be achieved through high-precision sensors positioned within the chamber. These sensors provide real-time data, enabling dynamic adjustments to aerosol inputs and maintaining target levels.

The aerosol control system must incorporate filters and scrubbers to prevent contamination from external sources or previous tests. These components ensure that the chamber atmosphere remains pure and free from unwanted particulates, thus preserving the integrity of experimental results.

Maintaining desired aerosol characteristics over extended periods may require a strategic resupplying approach. Such system periodically introduces additional aerosols to compensate for losses due to deposition or coagulation. This ensures that the concentration and characteristics of the aerosols remain within specified limits, supporting stable and reproducible interactions with thoron decay products.

3.8 Ventilation

To ensure homogenic dispersion of the short-lived Rn-220, uniform environmental conditions and even aerosol distribution, the chamber must include strategically positioned mixing fans with adjustable settings. Excessive airflow can cause particles to settle on surfaces (plate-out effect), potentially compromising results. Conversely, insufficient airflow may lead to atmospheric stratification, thus inhomogeneity. Hence, precise fan speed control is essential for maintaining measurement accuracy and reliability. Optimal positioning and fan power settings shall be determined through experimental evaluation to achieve the best performance.

4. Measurements strategies

In the following sections, two different measurements strategies will be shown: one developed by the BfS (that still requires some validations through experiments since the calibration chamber will be ready only in October 2025) and one method developed and tested by GIG.

4.1 BfS method

4.1.1 Source to generate a Rn-220 atmosphere

In designing an effective measurement strategy for thoron decay products, the choice of Rn-220 generating source and especially the source setup is critical for ensuring reliable assessments. Flow-through sources are particularly advantageous for this purpose. These sources typically use Th-228, which has a half-life of 1.9 years, allowing for consistent and stable generation of thoron gas. On the other hand, this Th-228 half-life leads to a considerable decrease over the years in use. The BfS Thoron Decay Product chamber has a volume of about 5.8 m³. If at least 10 kBq/m³ should be always available during the next 10 years in this chamber, a source of 380 kBq Th-228 minimum is necessary.

Flow-through sources can be integrated into experimental setups with relative ease, providing a steady flow of thoron into calibration chambers. They offer the flexibility of splitting the flow to introduce thoron simultaneously at multiple locations within the chamber, promoting homogeneity in the distribution of thoron decay products. This capability is essential for ensuring that measurements reflect representative exposure conditions.

Intense fan speeds might induce turbulent conditions that could disturb the sampling and lead to surface deposition of decay products. Instead, using a moderate fan intensity in combination with several Rn-220 outlets inside the chamber (flow divider) helps maintain a sufficiently uniform distribution of thoron decay products, particularly starting from Pb-212. For calibrations requiring focus on Po-216, measurements may need to concentrate on specific areas within the chamber where thoron and its decay products are more homogeneously distributed.

Minimising thoron transport time from the source to the calibration chamber is crucial, given thoron's short half-life. Consequently, sources shall be positioned in a way that allows to reduce the distance travelled by thoron gas, ensuring it reaches the chamber with minimal decay. Using the source or sources that are already inside the chamber or can be positioned close to it can help achieve this goal.

By employing flow-through sources and these strategic considerations, we can develop reference fields that ensure effective, accurate measurement of thoron decay products, aligning with both practical and theoretical requirements.

4.1.2 Instruments and method

At first, a Rn-220 decay products atmosphere is generated using a Th-228 flow-through source (chapter 4.1.1). In terms of efficiency and stability an equilibrium of the Rn-220 decay products would be ideal.

However, due to the long half-life of Pb-212 one would have to wait approximately 4 days to reach the equilibrium (see the section 3.3). As a compromise, waiting 1 day leads to moderate changes of the individual decay product activity concentration of 1.4% Pb-212 during 60 min which has to be considered for uncertainty calculations.

The PAEC measurement itself is done by grab sampling see the chapter 3.3.3. The sampling duration depends on the activity concentration vs. efficiency and the previously mentioned possible lack of an equilibrium has to be considered. A sampling rod with a wire-mesh on the first place to collect unattached radionuclides and a downstream filter to collect aerosol attached and remaining unattached Rn-220 decay products can be used. Collected decay products on both, the wire-mesh and filter, are measured offline using an alpha spectrometer and/or gamma spectrometer (see below chapter 4.1.3 Traceability).

From the determined nuclide activities on the filter or wire-mesh the initial activities from the grab sampling can be back-calculated and the PAE can be calculated (chapter 2.3.1). The sampling duration and volume flow during the grab sampling are measured to determine the filtered air volume and using this to calculate the PAEC by dividing the measured PAE by the filtered volume.

Additionally, the Rn-220 activity concentration can be measured separately to calculate an equilibrium factor for Rn-220 and its decay products.

To include Po-216 into the PAEC determination if necessary (see 2.3.1) the Rn-220 gas measurement can also be used for evaluation, since the activity concentrations of Rn-220 and Po-216 are equal each other as both radionuclides are in radioactive secular equilibrium. Due to the Rn-220 and Pb-216 half-life ratio under real conditions the calibration chamber is intended to be used, this assumption is always valid. Installing an online measuring system is under consideration as an alternative method to determine the alpha emitters Po-216, Bi-212 and Po-212 (as well as Po-218, Po-214 and Po-210 in a mixed Rn-222 / Rn-220 atmosphere). The online measurement could be realised using PIPS (Passivated Implanted Planar Silicon) detectors for alpha spectrometry, one in front of a wire-mesh and one in front of a filter, that are continuously sampling air. There are commercially available devices that can be adapted for this purpose.

To measure the environmental parameters described in chapter 0, the BfS thoron decay product chamber is equipped with

- Scanning Mobility Particle Sizer (SMPS) by the company TSI
- Condensation Particle Counter (CPC) by the company TSI
- Pumps
- Flowmeters
- internal sensors to measure temperature, relative humidity and atmospheric pressure (all traceable to a national standard)

4.1.3 Traceability of the Rn-220 decay products PAEC

According to the International Vocabulary of Metrology [23] the metrological traceability is defined as “property of a measurement result whereby the result can be related to a reference through a documented unbroken chain of calibrations, each contributing to the measurement uncertainty". Only National Metrological Institutes (NMI) or Designated Institutes (DI) can develop and provide primary measurement standards that are, by definition, reference measurement procedures used to obtain measurement results without relation to a measurement standard of the same kind [23]. From the metrological point of view, calibration laboratories that are not an NMI or DI do not have this opportunity of realising the traceability of its measurands without a relation to a measurement standard provided by an NMI or DI.

4.1.3.1 Alpha- and gamma spectrometry

For calibration laboratories no single Po-216 or Po-212 sources with certified activities are producible or available. As a consequence, alpha detectors used for decay product measurement cannot be calibrated directly with these alpha emitters under the same measurement conditions and geometries as the reference measurement during a calibration.

Instead, a certified Th-228 source in equilibrium with its decay products is used to calibrate the efficiency of a gamma spectrometry system for the Rn-220 decay product gamma energies (Pb-212, Bi-212, Tl-208) first. This source has to have the same geometry like the filters used for grab sampling during the measurement. In a second step, an efficiency calibration for an alpha spectrometer follows, using real grab sample at high activity concentrations and decay measurements alternating with the gamma and alpha spectrometer. The same measurement setup and sampling method as the ones used for reference measurements during calibrations will be used. Due to the secular equilibrium of the alpha emitters Bi-212 and Po-212 with gamma emitter Tl-208, the efficiency of the alpha spectrometer can be calculated from the efficiency calibrated gamma spectrometer. The alpha spectrometer in combination with the sampling equipment is then serving as a working standard.

Using an NaI well-type detector with an effectivity of around 50% it is possible to register the gamma energies of few kBq of Bi-212 (727 keV, 6.7% EP and 1621 keV, 1.5% EP). However, the energy resolution of the system does not allow to completely separate the main gamma lines of Ra-224 (241 keV, 4% EP) which is included in the Th-228 source, Pb-212 (239 keV, 43% EP and 300 keV, 3% EP) and Tl-208 (277 keV, 6% EP). Due to its high energy, the Tl-208 gamma line (2615 keV, 99,8% EP) has a high FWHM (Full Width Half Maximum) and less detector efficiency in this energy region but activity of both alpha emitters Bi-212 and Po-212 could be determined. However, after completing the BfS Thoron Decay Product Chamber different approaches will be applied and evaluated:

1. NaI well-type with gamma efficiency calibrations and measurement using Tl-208 line (and an efficiency calibration for the alpha spectrometer as described above)
2. Using a high-resolution gamma detector (HpGe detector) and full gamma spectrum analysis for the Rn-220 decay products to perform the efficiency calibrations (gamma and then alpha via Bi-212 and Po-212 spectrometry)

4.1.3.2 Sources of Rn-220 traceable to national standards

The approach to use the Rn-220 decay products included in certified radium and thorium sources for efficiency calibrations of a gamma spectrometer requires a radioactive equilibrium of the decay products and the source with the certified activity. This equilibrium can only be reached in sealed sources to prevent the Rn-220 from leaving the source. Since the spectrometric measurement setup requires a reference source with the same geometry like the samples taken from the Rn-220 decay product atmosphere, surface emanation design is chosen.

The source has to be a predecessor of Rn-220 with a half-life long enough to be usable in practice (> 1 a) and the gamma lines of the source and of the decay product chain down to Rn-220 shall not disturb the gamma spectrum evaluation of the evaluated Rn-220 decay products. Rn-220 emitting sources are listed in Table 6.

Table 6 – Radiation emission details on Rn-220 emitting sources; Data acc. to [24] energies < 50 keV and emission probabilities < 0.1% are not considered

Radionuclide	Half-life	Alpha radiation: Energy in keV (Emission probability)	Gamma or x-ray radiation: Energy in keV (Emission probability)
Th-232	1.4×10^{10} a	4013 (77%) 3954 (23 %)	59.0 (0.15%) – not easily detectable
Ra-228	5.75 a	-	-
Ac-228	6.2 h	-	911.32 (29%) 969.16 (17%) 338.37 (12%) 209.26 (5.0%) 795.07 (4.8%) 463.07 (4.6%) 270.27 (3.8%) 1587.90 (3.7%) ... and many more within the range of 60 and 1625 keV
Th-228	1.9 a	5423 (73%) 5340 (26%) 5211 (0.4%) 5177 (0.2%)	84.37 (1.2%) 215.99 (0.25%) 131.61 (0.13%) 166.41 (0.10%)
Ra-224	3.6 d	5685 (95 %) 5449 (5 %)	240.99 (4.1%) 83.78 (0.22%) – not easily detectable 81.07 (0.13%) – not easily detectable

In case of gamma spectrometry with a Th-232 source as reference the included Ac-228 could disturb evaluation of several Rn-220 decay product gamma lines. Furthermore, a Th-232 source would have to be old enough (> ca. 50 see Figure 21) to reach the radioactive equilibrium of Ra-224 which is then, after sealing of the source, in secular radioactive equilibrium with all the Rn-220 decay products after four days decay time (dominating half-life of Pb-212).

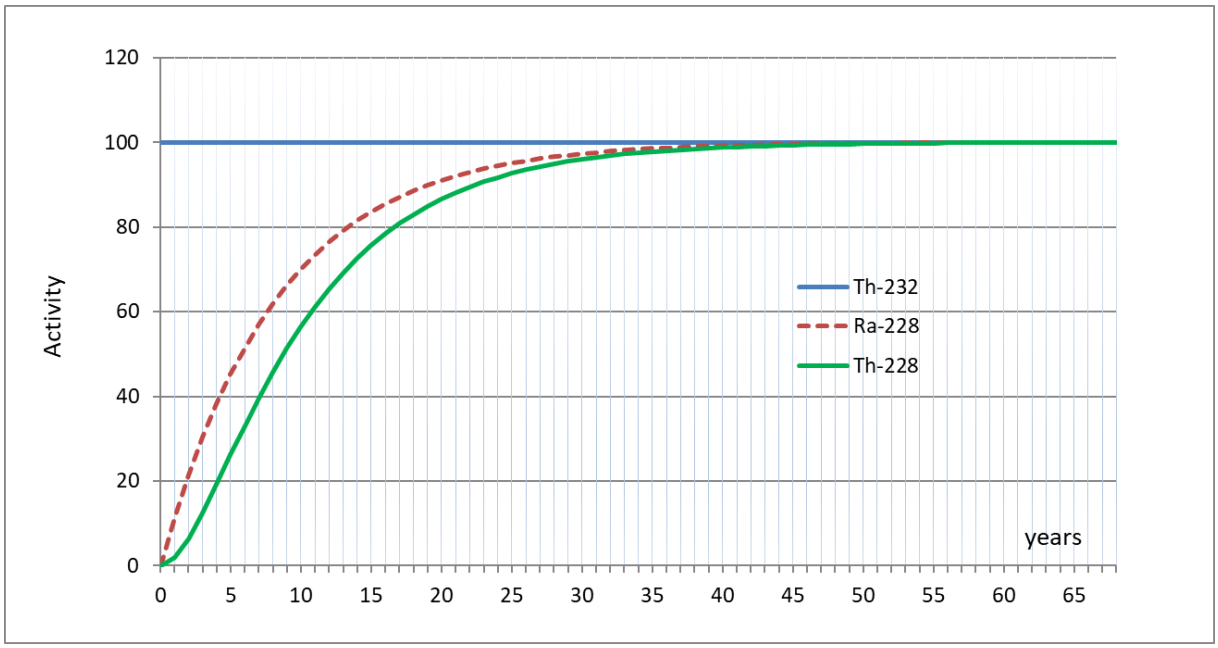

Figure 21 – Pure Th-232 source

In case of pure Ra-228 considered as a sealed source there is no option to get secular equilibrium with the Th-228 and then all Rn-220 decay products. It takes about 10 years to achieve transient equilibrium and activity of Th-228 (**Fehler! Verweisquelle konnte nicht gefunden werden.**). As for the Th-232, in case of gamma spectrometry the included Ac-228 could disturb evaluation of several Rn-220 decay product gamma lines.

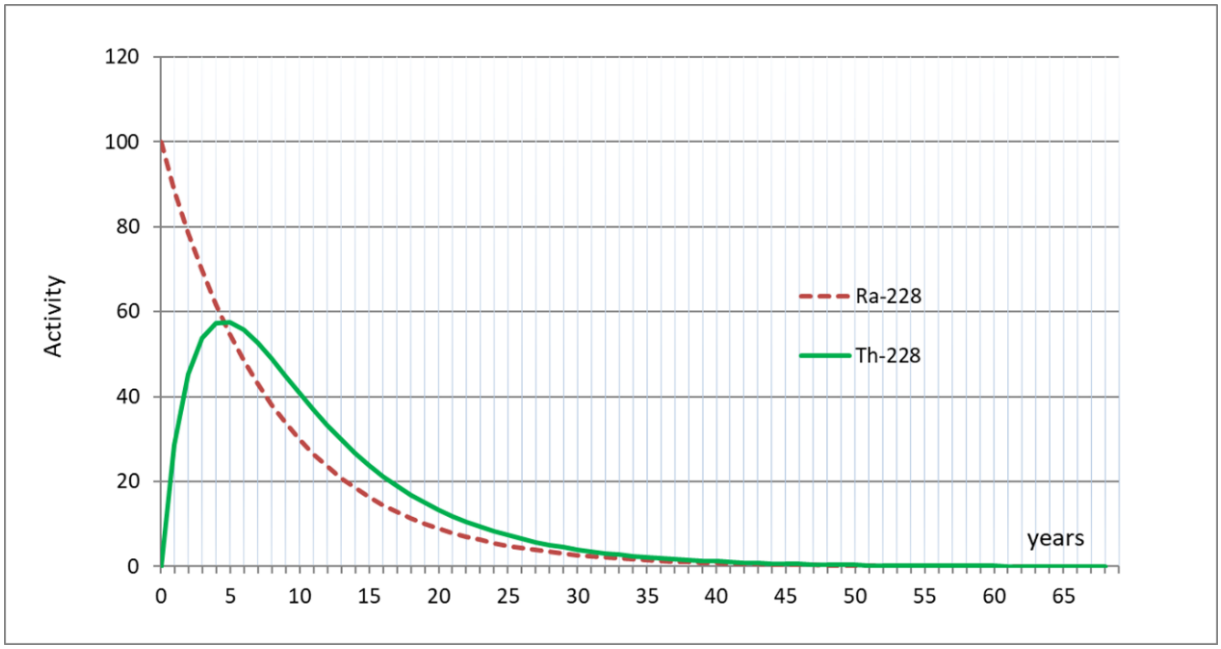

Figure 22 – Pure Ra-226 source - the activity of Th-228, initially with a value of 0, increase with time to a maximum of 0.58 on initial Ra-228- activity after about 4.55 years, and then is going to be bigger, finally reaching stable Th-228/Ra-228 ratio equal to about 1.5

In case of Th-228 new sources could be produced, sealed and certified by an NMI since aging for many years is not required. For gamma spectrometry main gamma lines of Th-228 (216 keV) and Ra-224 (241 keV) are overlapping several the main gamma line of Pb-212 (238 keV).

Pure Th-228 as a sealed source would always be in radioactive equilibrium with all Rn-220 decay products about one month after sealing. However, Th-228 sources with Th-232 impurities exist. In case of considerable Th-232 impurity, like for the Th-232 it takes about >50 years to get secular equilibrium (**Fehler! Verweisquelle konnte nicht gefunden werden.**).

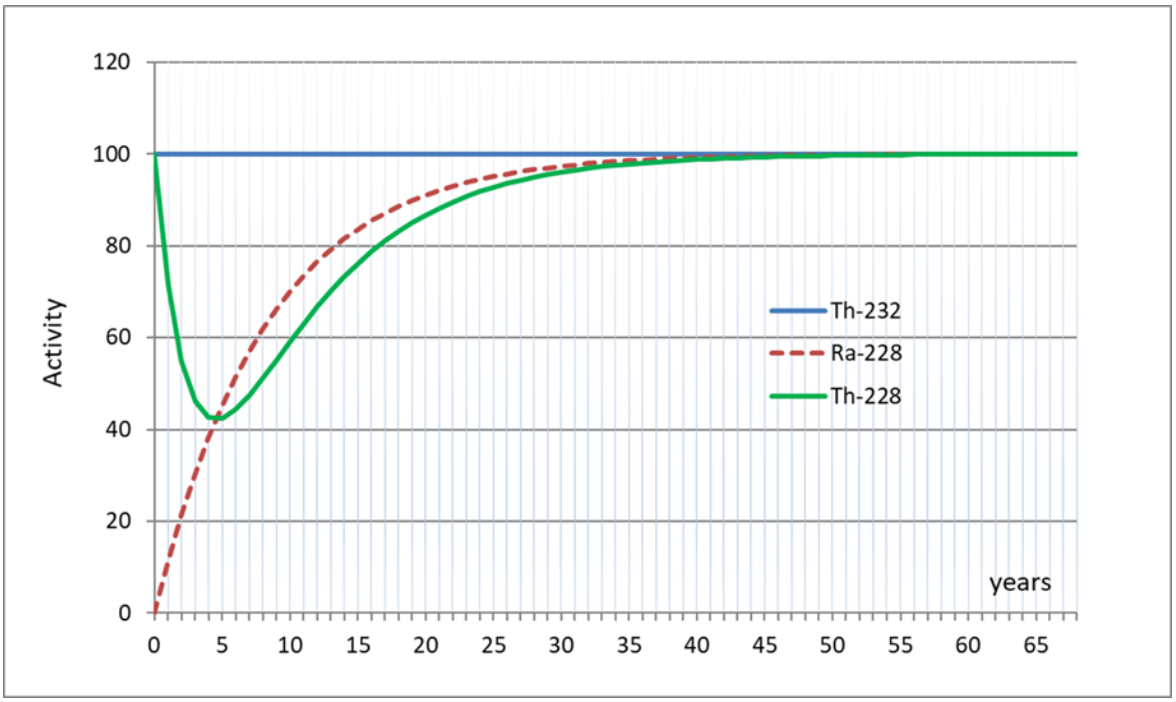

Figure 23 – A source obtained as a result of extraction of thorium isotopes – the Th-228 / Th-232 ratio, initially with a value of unity, should diminish with time to a minimum of 0.422 after about 4.55 years and then increase due to decay of Ra-228, to achieve final secular equilibrium with Th-232. In general, it is important to use a pure source and/or implement corrections of the decay and impurities

4.1.3.3 Volume flow

Since the PAEC is calculated using the volume flow during sampling, the sampling volume flow has to be traceable to national standards via calibration at an ISO EN ISO/IEC 17025 accredited calibration laboratory.

4.2 GIG method

At the Silesian Centre for Environmental Radioactivity at GIG-PIB, a procedure for calibrating meters used to measure the PAEC has been developed for its purposes. Calibration of meters involves establishing a relationship between the indications of a device being calibrated and a reference value. In general, a calibration procedure is carried out based on reference sources especially for devices measuring radon concentration. However, such a method seems to be insufficient for devices

designated to measure the PAEC of thoron decay products. In this case, the activity concentrations of Pb-212 and Bi-212 are measured and considered as reference, because the PAEC depends mainly on these nuclides (~90% for Pb-212 and ~10% for Bi-212) and contribution of other nuclides can be neglected. A liquid scintillation spectrometer is used to determine the concentration of these nuclides and corresponding PAEC based on measurement of filter activity in two different time intervals (two variables, so two equations are necessary). Since the detection efficiency of the liquid scintillation spectrometer for alpha radiation is approximately 100% and for beta radiation 98%, it was assumed that this method could be successfully used to determine the reference value of PAEC [25]. The basic data corresponding to thoron and its decay products can be found in Table 1.

4.2.1 Chamber and source

The climate chamber of the Silesian Centre for Environmental Radioactivity of GIG-PIB, where the calibration procedure is carried out, has a capacity of 17 m^3. The technical solutions used allow for manipulating the objects in the chamber from the outside, using airlocks and openings with gloves (Figure 24).

Figure 24 – Radon-thoron chamber of the Silesian Centre for Environmental Radioactive of GIG-PIB

The laboratory uses a thoron source made at GIG Silesian Centre for Environmental Radioactivity based on thorium chloride ($ThCl_4$). The transfer of thoron into the chamber was carried out by forcing air through the solution in the gas scrubber. The thoron is then transferred into the chamber in the bubbles that are formed when the air stream flows through the solution. The air flowrate through the solution is of about 2 L/min. A solution of 50 mL volume is used, where the activity of radium Ra-228 reached about 300 Bq/g. It was measured with the help of gamma-ray spectrometry based on actinium Ac-228 (Figure 25). Two laboratory scrubbers and a drier prevent particles of the solution but thoron from entering the chamber. Thoron is fed into the chamber approximately 3 days before the calibration procedure begins. During the calibration procedure, wax heaters generate aerosols in the chamber. As the aerosols begin to be generated, a fan is turned on to ensure a homogeneous atmosphere inside the chamber, and after about an hour, the calibration process begins.

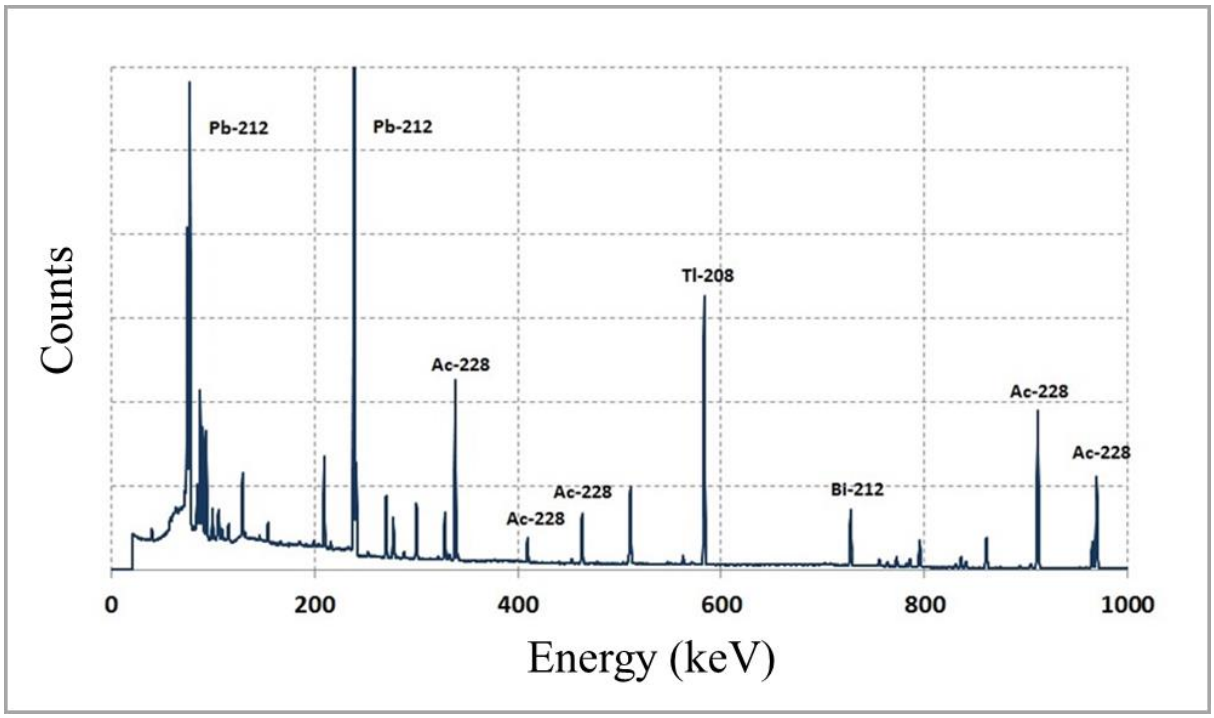

Figure 25 – Gamma-ray spectrum of thorium chloride used as a source of thoron

Generating a thoron atmosphere with a sufficiently high concentration of thoron decay products in a chamber with a capacity of 17 m^3 requires a reasonably powerful source. Due to the short half-life of thoron, the generation of a stable atmosphere in the chamber will not be ensured by a single injection of the nuclide or its brief transmission from the solution. This is because the thoron will quickly decay and its concentration will be very changeable. The solution is a constant supply of thoron at a sufficiently high activity rate. Using the source as described above, the achievable equilibrium PAEC of thoron is about 55 µJ/m^3. Using the solution of how the nuclide concentration of thoron builds up in closed facilities [21], it can be concluded that reaching such a PAEC level requires a constant inflow of thoron in the chamber at a rate of 150 Bq/s (see section the 3.3.3). Then, after about 70-80 hours from the start of thoron transfer, the PAEC concentration will reach the targeted value of 55 µJ/m^3. A large chamber provides relative stability in nuclide concentration values, provided that the airflow through the calibrated device or reference device is not too high. The airflows through the reference filter and the calibrated device were about 2 L/min, and their operating time was 60 minutes, meaning that 0.24 m^3 of air would be filtered during this time. Compared to the volume chamber of 17 m^3 this is negligible

4.2.2 Instruments and method

4.2.2.1 Reference method

Measuring the concentration of thoron decay products is much more complicated compared to measuring gaseous thoron. It usually requires a filter system to collect aerosols and a detection system to record the radiation emitted by the collected decay products on the filter. Assessing the activity concentrations of decay products in the air requires measurement of filter activity in different time intervals. Their number can be reduced if alpha or gamma-ray spectrometric systems are used, allowing identification of individual nuclides. However, scintillation counters do not provide the possibility of identifying nuclides, so it is necessary to select appropriate time intervals at which the filter activity is indicated [26-30] . The time intervals used have been modified by many researchers working on this issue over the years. Initially, attention was focused only on measuring radon decay products, but over time, the concentrations of thoron decay products began to be determined as well [6; 26; 31].

Measurement of the potential alpha energy concentration of thoron decay products requires determining the activity of two nuclides (Pb-212 and Bi-212), so the measurement of filter activity must be carried out in two-time intervals. The measurement of the reference concentration of thoron decay products in GIG-PIB is carried out according to the scheme:

- sample collection 0-60 minutes,
- first measurement 65-125 minutes,
- second measurement 130-250 minutes.

The filtration efficiency for the filter used (Pragopor 4, nitrocellulose, porosity 0.85 µm) is 99 ± 1%. The alpha radiation detection efficiency is 100 ± 1%, and the beta radiation detection efficiency is 98 ± 1%. The potential alpha energy concentration is defined as the total energy of alpha radiation particles emitted by short-lived radon or thoron decay products following their complete radioactive decay in a given volume of gas. The filter activity is measured using a liquid scintillator (Triathler, Hidex, Finland). In this method, alpha and beta radiation are subject to detection. When air is pumped through the filter, the relationships between the activity of nuclides A_i accumulated on the filter and their radioactive activity concentration in the air C_i have the following form:

$$\frac{\partial A_1(t)}{\partial t} = FQC_1 - \lambda_1 A_1(t) \quad \text{and} \quad \frac{\partial A_i(t)}{\partial t} = FQC_i + \varepsilon_i \lambda_i A_{i-1}(t) - \lambda_i A_i\,(t) \text{ for } i \geq 2$$

where:

Q – airflow rate through the filter,

F – filtration efficiency,

λ_i – decay constant of the i-th nuclide.

The individual products of thoron decay are formed by the decay of the nuclides that precede them in the series. Hence, in the differential equations, the branch ratios ε_i indicating what part of the parent nuclide leads to its decay products must be included. The solutions to these equations can be written in the form:

$$A_i(t) = FQ \sum_{j=1}^{i} \left(\left(\prod_{k=j+1}^{i} \varepsilon_k \right) \frac{F_{ij}(t) C_j}{\lambda_j} \right)$$

At the end of pumping, the decay products in the air will no longer reach the filter, and the nuclides accumulated on the filter will continue to transform according to the law of radioactive decay, except that the initial activity will be $A_i(t_p)$, where t_p is the air sampling time. Then the activities of the nuclides A_i' on the filter will change according to the equations:

$$A_i'(t) = \sum_{j=1}^{i} \left(\left(\prod_{k=j+1}^{i} \varepsilon_k \right) G_{ij}(t) A_j(t_p) \right)$$

The F_{ij} and G_{ij} functions are included in Appendix A.

The number of recorded counts $\Re_i$ by the measurement system depends on the activity of the nuclides on the filter, the detection efficiency and the detection time. The $\Re_i$ quantities are proportional to the integrals as shown in the following formula. In the most general terms, the number of registered counts in a certain time interval $\Delta t = (t_1, t_2)$ as a result of radiation emission by the i-th nuclide, is equal to:

$$\Re_i = \eta_{Di}\left(\int_{t_1}^{t_p} A_i(t)dt + \int_{t_p}^{t_2} A_i'(t)dt\right)$$

Where:

η_{Di} – the detection efficiency of the radiation emitted by the i-th nuclide.

Depending on the situation, it is necessary to sum up the magnitudes $\Re_i$ for those nuclides whose radiation is recorded by the measuring system to obtain a result corresponding to the response obtained by the device used. After expansion, this relationship can also be presented in the following form:

$$\Re_i(t_p, t_1, t_2) = \eta_{Di} FQ\left(\sum_{j=1}^{i}\left(\left(\prod_{k=j+1}^{i}\varepsilon_k\right)\Gamma_{ij}(t_p, t_1, t_2)\frac{C_j}{\lambda_j}\right)\right)$$

The forms of the function Γ_{ij} are included in Appendix A. These relations make up the system of equations that must be solved to estimate the magnitudes of the activity concentrations C_j of the nuclides in the air, which in turn will enable the determination of the reference potential alpha energy concentration. The number of time intervals at which to measure the filter activity depends on the number of nuclides whose activity concentrations need to be determined. Finally, the solution is obtained by inverting the formed matrix based on the previous equation for different detection times.

4.2.2.2 Calibration procedure

A calibrated device and a filter device are placed in the chamber to measure the reference potential alpha energy concentration. Stabilized air flow rate Q through the filters is provided by the AP-2000 EX aspirator. Both devices are started simultaneously, and the sampling time t_p was the same. In a thoron atmosphere, the measurement of potential alpha energy concentration takes 1 hour. To evaluate the reference concentration, Pragopor-4 cellulose membrane filters (Pragochema, Prague, Czech Republic) with a pore size of 0.85 μm are used. After pumping, the reference filter is immersed in liquid scintillator and its activity is measured at successive time intervals using a Triathler spectrometer (Hidex Oy, Turku, Finland). The results obtained allow assessment of the potential alpha energy concentration inside the chamber (Cα). The vial in which the filter is immersed contains 10 ml of toluene scintillator without triton. Subsequently, the calibration factor is determined based on the reading of the calibrated device and the determined reference value of the potential alpha energy concentration.

4.2.2.3 Uncertainty budget

The uncertainty budget of the method described above consists of: the uncertainty of the air flow rate assessment through the filter, the uncertainty of the filtration efficiency assessment and the detection uncertainty associated with the liquid scintillation counter.

4.2.3 Traceability

The Silesian Centre for Environmental Radioactivity GIG-PIB is not a calibration laboratory. The laboratory is accredited by the Polish Center for Accreditation (ISO 17025) only for performing tests, not calibrations. All procedures called calibration in chapter 4.2 are performed only for the laboratory's own needs.

4.2.4 RadoThor software

The RadoThor software was developed to support calculations related to radon, thoron and their decay products. It has two functionalities. The first one is intended to determine such quantities as the potential

alpha energy concentration, equilibrium coefficients, equilibrium concentrations and conversion of units (Figure 26). The second one is used to determine the activity concentrations of radon and thoron decay products and the corresponding uncertainties based on the results of the filter activity measurements (Figure 27).

The parameters necessary to perform these tasks can also be defined by the user and stored in databases. However, to use this option, the application Access (Microsoft) must be installed.

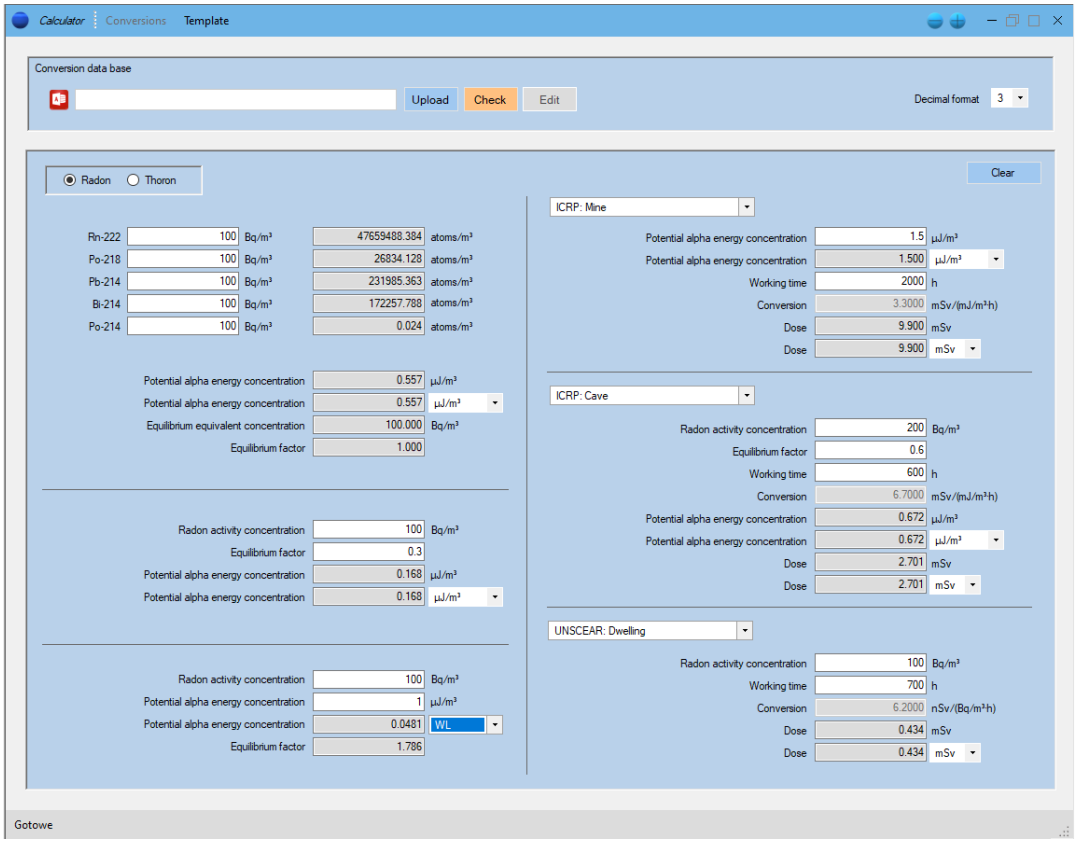

Figure 26 – Functionality 1. Calculations of activity concentrations, equilibrium coefficients and doses for radon and thoron, and their decay products

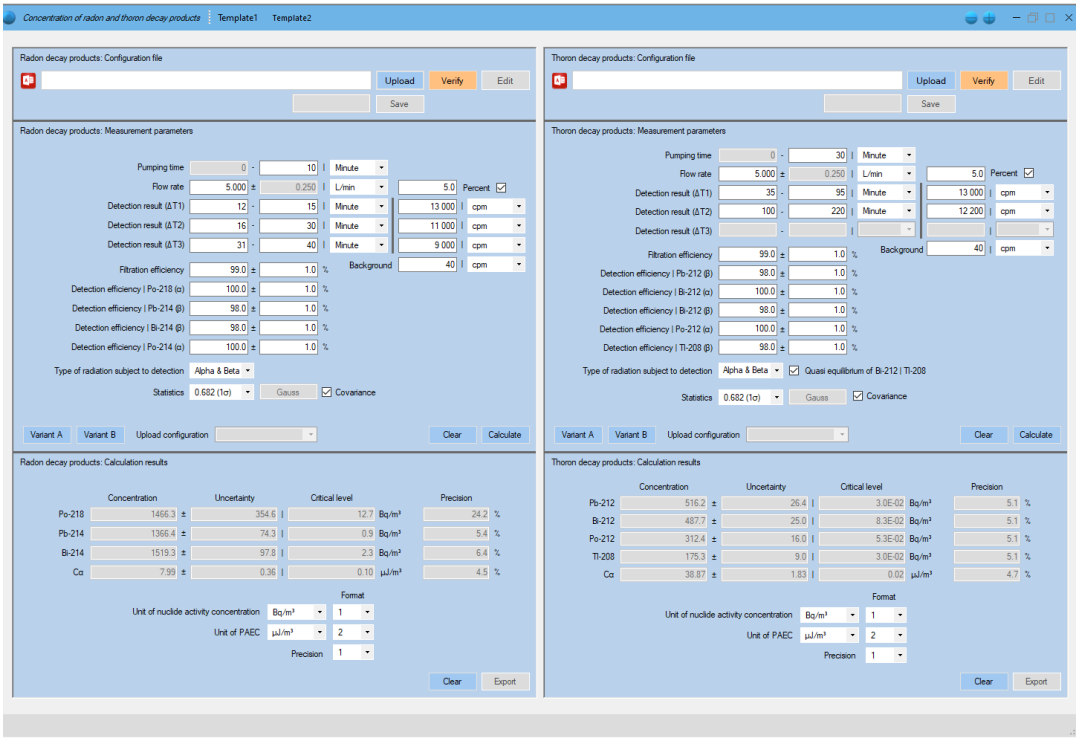

Figure 27 – Functionality 2. Measurement of activity concentrations of radon and thoron decay products

5. Conclusions

The RadoNorm project's Deliverable 2.2 outlines the essential components for developing a metrology infrastructure to address thoron decay product measurements, crucial for radiation protection considering thoron's potential public health impact.

The report emphasizes that while extensive research exists for radon, comparable methodologies for thoron are underdeveloped, underscoring a gap in existing metrological capabilities in Europe. The design and implementation of a thoron calibration chamber are pivotal developments recommended in the report, ensuring standardized, traceable, and quality-assured measurements across EU member states.

To understand if thoron and its decay products are of concern or not when considering the exposure of the public to natural radiations, it is necessary to perform measurements campaigns with tested methods and calibrated instruments. The instruments must be referred to a traceable standard and calibrated with quality assured methods.

The two different methods described in this report serve as a first base for the future set up a metrological structure for thoron decay products calibrations and measurements.

6. Outlook

Theoretical studies and literature research are essential to design a calibration chamber for the instruments that measure the thoron decay products, but since every chamber has different technical details, practical tests are essential to characterize the reference atmosphere and properly develop the method.

The models developed for simulation the conditions inside a chamber can facilitate the development of a new chamber and plan the calibration process in existing one, if any. The models developed as Excel files are attached to already published articles [21, 22]. The model for an open system is also directly available on RadoNorm webpage:

https://www.radonorm.eu/publications/other/#359-359-wpfd-top

The RadoThor software needs installation on an end-used computer. The installation files, for either 32- or 64-bits system architecture will be available on the same RadoNorm webpage. The RadoThor.zip file contains all necessary instruction and installation files. The software is for use for free based only on user registration.

Regarding the BfS method, it is important to mention that at this stage of the work it was not possible to perform experiments, because the calibration chamber is being constructed while this report is written and it will not be available for tests before the end of the RadoNorm project.

Commissioning of a thoron decay product calibration facility has not been covered by the RadoNorm project. In parallel to the project, BfS made significant efforts to design and procure such an installation and to ensure its operation in future. This internal project arrived at final status with a first operation expected in autumn 2025. The factory acceptance test is expected in June 2025. Some impressions of the status of the work are given in Figure 28.

	
Rear with auxiliary equipment for atmosphere conditioning	Front door

Figure 28 – BfS Thoron decay product chamber, status 06/06/2025 at manufacturers premise

Characterization tests that will be performed as soon as the thoron decay products calibration chamber is available and functioning at BfS:

- Homogeneity of environmental parameters
- Study on intensity and positioning of fans
- Study on the best aerosol conditions for the method
- Thoron decay products measurements
- Changes on the reference atmosphere when the chamber contains instruments (more surfaces)

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Appendix A. Forms of the functions

Function $\Gamma ij(t_p, t_1, t_2)$

The magnitude t_p is the pumping time, and t_1 and t_2 are the beginning and end of the detection time relative to when pumping began.

$\Gamma_{11}(t_p, t_1, t_2)$

- $t_2 \leq t_p$

  $\Gamma_{11}(t_p, t_1, t_2) = IntF_{11}(t_2) - IntF_{11}(t_1)$

- $t_1 > t_p$

  $\Gamma_{11}(t_p, t_1, t_2) = F_{11}(t_p, t_p)\,(IntG_{11}(t_p, t_2) - IntG_{11}(t_p, t_1))$

- $t_1 \leq t_p$ oraz $t_p < t_2$

  $\Gamma_{11}(t_p, t_1, t_2) = IntF_{11}(t_p) - IntF_{11}(t_1) + F_{11}(t_p, t_p)\,(IntG_{11}(t_p, t_2) - IntG_{11}(t_p, t_p))$

$\Gamma_{21}(t_p, t_1, t_2)$

- $t_2 \leq t_p$

  $\Gamma_{21}(t_p, t_1, t_2) = IntF_{21}(t_2) - IntF_{21}(t_1)$

- $t_1 > t_p$

  $\Gamma_{21}(t_p, t_1, t_2) = F_{11}(t_p, t_p)\,(IntG_{21}(t_p, t_2) - IntG_{21}(t_p, t_1)) + F_{21}(t_p, t_p)\,(IntG_{22}(t_p, t_2) - IntG_{22}(t_p, t_1))$

- $t_1 \leq t_p$ oraz $t_p < t_2$

  $\Gamma_{21}(t_p, t_1, t_2) = IntF_{21}(t_p) - IntF_{21}(t1) + F_{11}(t_p, t_p)\,(IntG_{21}(t_p, t_2) - IntG_{21}(t_p, t_p)) +$
  $F_{21}(t_p, t_p)\,(IntG_{22}(t_p, t_2) - IntG_{22}(t_p, t_p))$

$\Gamma_{22}(t_p, t_1, t_2)$

- $t_2 \leq t_p$

  $\Gamma_{21}(t_p, t_1, t_2) = IntF_{22}(t_2) - IntF_{22}(t_1)$

- $t_1 > t_p$

  $\Gamma_{21}(t_p, t_1, t_2) = F_{22}(t_p, t_p)\,(IntG_{22}(t_p, t_2) - IntG_{22}(t_p, t_1))$

- $t_1 \leq t_p$ oraz $t_p < t_2$

  $\Gamma_{21}(t_p, t_1, t_2) = IntF_{22}(t_p) - IntF_{22}(t_1) + F_{22}(t_p, t_p)\,(IntG_{22}(t_p, t_2) - IntG_{22}(t_p, t_p))$

$\Gamma_{31}(t_p, t_1, t_2)$

- $t_2 \leq t_p$

  $\Gamma_{31}(t_p, t_1, t_2) = IntF_{31}(t_2) - IntF_{31}(t_1)$

- $t_1 > t_p$

  $\Gamma_{31}(t_p, t_1, t_2) = F_{11}(t_p, t_p)\,(IntG_{31}(t_p, t_2) - IntG_{31}(t_p, t_1)) + F_{21}(t_p, t_p)\,(IntG_{32}(t_p, t_2) - IntG_{32}(t_p, t_1)) +$
  $F_{31}(t_p, t_p)\,(IntG_{33}(t_p, t_2) - IntG_{33}(t_p, t_1))$

- $t_1 \leq t_p$ oraz $t_p < t_2$

$$\Gamma_{31}(t_p, t_1, t_2) = IntF_{31}(t_p) - IntF_{31}(t_1) + F_{11}(t_p, t_p)\,(IntG31(t_p, t_2) - IntG_{31}(t_p, t_p)) +$$
$$F_{21}(t_p, t_p)\,(IntG_{32}(t_p, t_2) - IntG_{32}(t_p, t_p)) + F_{31}(t_p, t_p)\,(IntG_{33}(t_p, t_2) - IntG_{33}(t_p, t_p))$$

$\Gamma_{32}(t_p, t_1, t_2)$

- $t_2 \le t_p$

  $\Gamma_{32}(t_p, t_1, t_2) = IntF_{32}(t_2) - IntF_{32}(t_1)$

- $t_1 > t_p$

  $\Gamma_{32}(t_p, t_1, t_2) = F_{22}(t_p, t_p)\,(IntG_{32}(t_p, t_2) - IntG_{32}(t_p, t_1)) + F_{32}(t_p, t_p)\,(IntG_{33}(t_p, t2) - IntG_{33}(t_p, t_1))$

- $t_1 \le t_p$ oraz $t_p < t_2$

  $\Gamma_{32}(t_p, t_1, t_2) = IntF_{32}(t_p) - IntF_{32}(t_1) + F_{22}(t_p, t_p)\,(IntG_{32}(t_p, t_2) - IntG_{32}(t_p, t_p)) +$
  $F_{32}(t_p, t_p)\,(IntG_{33}(t_p, t_2) - IntG_{33}(t_p, t_p))$

$\Gamma_{33}(t_p, t_1, t_2)$

- $t_2 \le t_p$

  $\Gamma_{33}(t_p, t_1, t_2) = IntF_{33}(t_2) - IntF_{33}(t_1)$

- $t_1 > t_p$

  $\Gamma_{33}(t_p, t_1, t_2) = F_{33}(t_p, t_p)\,(IntG_{33}(t_p, t_2) - IntG_{33}(t_p, t_1))$

- $t_1 \le t_p$ oraz $t_p < t_2$

  $\Gamma_{33}(t_p, t_1, t_2) = IntF_{33}(t_p) - IntF_{33}(t_1) + F_{33}(t_p, t_p)\,(IntG_{33}(t_p, t_2) - IntG_{33}(t_p, t_p))$

$\Gamma_{41}(t_p, t_1, t_2)$

- $t_2 \le t_p$

  $\Gamma_{41}(t_p, t_1, t_2) = IntF_{41}(t_2) - IntF_{41}(t_1)$

- $t_1 > t_p$

  $\Gamma_{41}(t_p, t_1, t_2) = F_{11}(t_p, t_p)\,(IntG_{41}(t_p, t_2) - IntG_{41}(t_p, t_1)) + F_{21}(t_p, t_p)\,(IntG_{42}(t_p, t_2) - IntG_{42}(t_p, t_1)) +$
  $F_{31}(t_p, t_p)\,(IntG_{43}(t_p, t_2) - IntG_{43}(t_p, t_1)) + F_{41}(t_p, t_p)\,(IntG_{44}(t_p, t_2) - IntG_{44}(t_p, t_1))$

- $t_1 \le t_p$ oraz $t_p < t_2$

  $\Gamma_{41}(t_p, t_1, t_2) = IntF_{41}(t_p) - IntF_{41}(t_1) + F_{11}(t_p, t_p)\,(IntG_{41}(t_p, t_2) - IntG_{41}(t_p, t_2)) + F_{21}(t_p, t_p)\,(IntG_{42}(t_p, t_2)$
  $- IntG_{42}(t_p, t_p)) + F_{31}(t_p, t_p)\,(IntG_{43}(t_p, t_2) - IntG_{43}(t_p, t_p)) + F_{41}(t_p, t_p)\,(IntG_{44}(t_p, t_2) - IntG_{44}(t_p, t_p))$

$\Gamma_{42}(t_p, t_1, t_2)$

- $t_2 \le t_p$

  $\Gamma_{42}(t_p, t_1, t_2) = IntF_{42}(t_2) - IntF_{42}(t_1)$

- $t_1 > t_p$

  $\Gamma_{42}(t_p, t_1, t_2) = F_{22}(t_p, t_p)\,(IntG_{42}(t_p, t_2) - IntG_{42}(t_p, t_1)) + F_{32}(t_p, t_p)\,(IntG_{43}(t_p, t_2) - IntG_{43}(t_p, t_1)) +$
  $F_{42}(t_p, t_p)\,(IntG_{44}(t_p, t_2) - IntG_{44}(t_p, t_1))$

- $t_1 \leq t_p$ oraz $t_p < t_2$

  $\Gamma_{42}(t_p, t_1, t_2) = IntF_{42}(t_p) - IntF_{42}(t_1) + F_{22}(t_p, t_p)\,(IntG_{42}(t_p, t_2) - IntG_{42}(t_p, t_p)) +$
  $F_{32}(t_p, t_p)\,(IntG_{43}(t_p, t_2) - IntG_{43}(t_p, t_p)) + F_{42}(t_p, t_p) * (IntG_{44}(t_p, t_2) - IntG_{44}(t_p, t_p))$

$\Gamma_{43}(t_p, t_1, t_2)$

- $t_2 \leq t_p$

  $\Gamma_{43}(t_p, t_1, t_2) = IntF_{43}(t_2) - IntF_{43}(t_1)$

- $t_1 > t_p$

  $\Gamma_{43}(t_p, t_1, t_2) = F_{33}(t_p, t_p)\,(IntG_{43}(t_p, t_2) - IntG_{43}(t_p, t_1)) + F_{43}(t_p, t_p)\,(IntG_{44}(t_p, t_2) - IntG_{44}(t_p, t_1))$

- $t_1 \leq t_p$ oraz $t_p < t_2$

  $\Gamma_{43}(t_p, t_1, t_2) = IntF_{43}(t_p) - IntF_{43}(t_1) + F_{33}(t_p, t_p)\,(IntG_{43}(t_p, t_2) - IntG_{43}(t_p, t_p)) +$
  $F_{43}(t_p, t_p)\,(IntG_{44}(t_p, t_2) - IntG44(t_p, t_p))$

$\Gamma_{44}(t_p, t_1, t_2)$

- $t_2 \leq t_p$

  $\Gamma_{44}(t_p, t_1, t_2) = IntF_{44}(t_2) - IntF_{44}(t_1)$

- $t_1 > t_p$

  $\Gamma_{44}(t_p, t_1, t_2) = F_{44}(t_p, t_p)\,(IntG_{44}(t_p, t_2) - IntG_{44}(t_p, t_1))$

- $t_1 \leq t_p$ oraz $t_p < t_2$

  $\Gamma_{44}(t_p, t_1, t_2) = IntF_{44}(t_p) - IntF_{44}(t_1) + F_{44}(t_p, t_p)\,(IntG_{44}(t_p, t_2) - IntG_{44}(t_p, t_p))$

Function Fij(t, t_p)

The magnitude t_p is the pumping time, and t is the time relative to when pumping began. If $t \geq t_p$ to $t=t_p$.

$$F_{11}(t, t_p) = 1 - e^{-\lambda_1 t}$$

$$F_{21}(t, t_p) = 1 - \frac{\lambda_2}{(\lambda_2 - \lambda_1)}\, e^{-\lambda_1 t} - \frac{\lambda_1}{(\lambda_1 - \lambda_2)}\, e^{-\lambda_2 t}$$

$$F_{22}(t, t_p) = 1 - e^{-\lambda_2 t}$$

$$F_{31}(t, t_p) = 1 - \frac{\lambda_2 \lambda_3}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)}\, e^{-\lambda_1 t} - \frac{\lambda_1 \lambda_3}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)}\, e^{-\lambda_2 t} - \frac{\lambda_1 \lambda_2}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)}\, e^{-\lambda_3 t}$$

$$F_{32}(t, t_p) = 1 - \frac{\lambda_3}{\lambda_3 - \lambda_2} e^{-\lambda_2 t} - \frac{\lambda_2}{\lambda_2 - \lambda_3}\, e^{-\lambda_3 t}$$

$$F_{33}(t, t_p) = 1 - e^{-\lambda_3 t}$$

$$F_{41}(t,t_p) = 1 - \frac{\lambda_2\,\lambda_3\lambda_4}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)(\lambda_4-\lambda_1)}\,e^{-\lambda_1 t} - \frac{\lambda_1\,\lambda_3\lambda_4}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)(\lambda_4-\lambda_2)}\,e^{-\lambda_2 t} - \frac{\lambda_1\,\lambda_2\lambda_4}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)(\lambda_4-\lambda_3)}\,e^{-\lambda_3 t} - \frac{\lambda_1\,\lambda_2\lambda_3}{(\lambda_1-\lambda_4)(\lambda_2-\lambda_4)(\lambda_3-\lambda_4)}\,e^{-\lambda_4 t}$$

$$F_{42}(t,t_p) = 1 - \frac{\lambda_3\lambda_4}{(\lambda_3-\lambda_2)(\lambda_4-\lambda_2)}\,e^{-\lambda_2 t} - \frac{\lambda_2\lambda_4}{(\lambda_2-\lambda_3)(\lambda_4-\lambda_3)}\,e^{-\lambda_3 t} - \frac{\lambda_2\lambda_3}{(\lambda_2-\lambda_4)(\lambda_3-\lambda_4)}\,e^{-\lambda_4 t}$$

$$F_{43}(t,t_p) = 1 - \frac{\lambda_4}{\lambda_4-\lambda_3}\,e^{-\lambda_3 t} - \frac{\lambda_3}{\lambda_3-\lambda_4}\,e^{-\lambda_4 t}$$

$$F_{44}(t,t_p) = 1 - e^{-\lambda_4 t}$$

Function Gij(t, t_p)

The magnitude t_p is the pumping time, and t is the time relative to when pumping began.

$$G_{11}(t,t_p) = e^{-\lambda_1(t-t_p)}$$

$$G_{21}(t,t_p) = \frac{\lambda_2}{(\lambda_2-\lambda_1)}\,e^{-\lambda_1(t-t_p)} + \frac{\lambda_2}{(\lambda_1-\lambda_2)}\,e^{-\lambda_2(t-t_p)}$$

$$G_{22}(t,t_p) = e^{-\lambda_2(t-t_p)}$$

$$G_{31}(t,t_p) = \frac{\lambda_2\,\lambda_3}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)}\,e^{-\lambda_1(t-t_p)} + \frac{\lambda_2\lambda_3}{(\lambda_1-\lambda_2)\,(\lambda_3-\lambda_2)}\,e^{-\lambda_2(t-t_p)} + \frac{\lambda_2\lambda_3}{(\lambda_1-\lambda_3)\,(\lambda_2-\lambda_3)}\,e^{-\lambda_3(t-t_p)}$$

$$G_{32}(t,t_p) = \frac{\lambda_3}{\lambda_3-\lambda_2}e^{-\lambda_2(t-t_p)} + \frac{\lambda_3}{\lambda_2-\lambda_3}\,e^{-\lambda_3(t-t_p)}$$

$$G_{33}(t,t_p) = e^{-\lambda_3(t-t_p)}$$

$$G_{41}(t,t_p) = \frac{\lambda_2\,\lambda_3\lambda_4}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)(\lambda_4-\lambda_1)}\,e^{-\lambda_1(t-t_p)} + \frac{\lambda_2\,\lambda_3\lambda_4}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)(\lambda_4-\lambda_2)}\,e^{-\lambda_2(t-t_p)} + \frac{\lambda_2\,\lambda_3\lambda_4}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)(\lambda_4-\lambda_3)}\,e^{-\lambda_3(t-t_p)} + \frac{\lambda_2\,\lambda_3\lambda_4}{(\lambda_1-\lambda_4)(\lambda_2-\lambda_4)(\lambda_3-\lambda_4)}\,e^{-\lambda_4(t-t_p)}$$

$$G_{42}(t,t_p) = \frac{\lambda_3\lambda_4}{(\lambda_3-\lambda_2)(\lambda_4-\lambda_2)}\,e^{-\lambda_2(t-t_p)} + \frac{\lambda_3\lambda_4}{(\lambda_2-\lambda_3)(\lambda_4-\lambda_3)}\,e^{-\lambda_3(t-t_p)} + \frac{\lambda_3\lambda_4}{(\lambda_2-\lambda_4)(\lambda_3-\lambda_4)}\,e^{-\lambda_4(t-t_p)}$$

$$G_{43}(t,t_p) = \frac{\lambda_4}{\lambda_4-\lambda_3}\,e^{-\lambda_3(t-t_p)} + \frac{\lambda_4}{\lambda_3-\lambda_4}\,e^{-\lambda_4(t-t_p)}$$

$$G_{44}(t,t_p) = e^{-\lambda_4(t-t_p)}$$

Function IntFij(t)

Used if t >t_p, where t_p is the time to pump air through the filter.

$$IntF_{11}(t) = t + \frac{1}{\lambda_1}\, e^{-\lambda_1 t}$$

$$IntF_{21}(t) = t + \frac{\lambda_2}{\lambda_1(\lambda_2 - \lambda_1)}\, e^{-\lambda_1 t} + \frac{\lambda_1}{\lambda_2(\lambda_1 - \lambda_2)}\, e^{-\lambda_2 t}$$

$$IntF_{22}(t) = t + \frac{1}{\lambda_2}\, e^{-\lambda_2 t}$$

$$IntF_{31}(t) = t + \frac{\lambda_2\lambda_3}{\lambda_1(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)}\, e^{-\lambda_1 t} + \frac{\lambda_1\lambda_3}{\lambda_2(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} e^{-\lambda_2 t} + \frac{\lambda_1\lambda_2}{\lambda_3(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)}\, e^{-\lambda_3 t}$$

$$IntF_{32}(t) = t + \frac{\lambda_3}{\lambda_2(\lambda_3 - \lambda_2)}\, e^{-\lambda_2 t} + \frac{\lambda_2}{\lambda_3(\lambda_2 - \lambda_3)}\, e^{-\lambda_3 t}$$

$$IntF_{33}(t) = t + \frac{1}{\lambda_3}\, e^{-\lambda_3 t}$$

$$IntF_{41}(t) = t + \frac{\lambda_2\lambda_3\lambda_4}{\lambda_1(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)(\lambda_4 - \lambda_1)}\, e^{-\lambda_1 t} + \frac{\lambda_1\lambda_3\lambda_4}{\lambda_2(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)(\lambda_4 - \lambda_2)} e^{-\lambda_2 t} + \frac{\lambda_1\lambda_2\lambda_4}{\lambda_3(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)(\lambda_4 - \lambda_3)} e^{-\lambda_3 t} + \frac{\lambda_1\lambda_2\lambda_3}{\lambda_4(\lambda_1 - \lambda_4)(\lambda_2 - \lambda_4)(\lambda_3 - \lambda_4)} e^{-\lambda_4 t}$$

$$IntF_{42}(t) = t + \frac{\lambda_3\lambda_4}{\lambda_2(\lambda_3 - \lambda_2)(\lambda_4 - \lambda_2)}\, e^{-\lambda_2 t} + \frac{\lambda_2\lambda_4}{\lambda_3(\lambda_2 - \lambda_3)(\lambda_4 - \lambda_3)} e^{-\lambda_3 t} + \frac{\lambda_2\lambda_3}{\lambda_4(\lambda_2 - \lambda_4)(\lambda_3 - \lambda_4)}\, e^{-\lambda_4 t}$$

$$IntF_{43}(t) = t + \frac{\lambda_4}{\lambda_3(\lambda_4 - \lambda_3)}\, e^{-\lambda_3 t} + \frac{\lambda_3}{\lambda_4(\lambda_3 - \lambda_4)}\, e^{-\lambda_4 t}$$

$$IntF_{44}(t) = t + \frac{1}{\lambda_4}\, e^{-\lambda_4 t}$$

Function IntGij(t, t_p)

Used if t >t_p, where t_p is the time to pump air through the filter.

$$IntG_{11}(t) = -\frac{1}{\lambda_1}\, e^{-\lambda_1 (t - t_p)}$$

$$IntG_{21}(t) = -\frac{\lambda_2}{\lambda_1(\lambda_2 - \lambda_1)}\, e^{-\lambda_1 (t - t_p)} - \frac{1}{\lambda_1 - \lambda_2}\, e^{-\lambda_2 (t - t_p)}$$

$$IntG_{22}(t) = -\frac{1}{\lambda_2}\, e^{-\lambda_2 (t - t_p)}$$

$$IntG_{31}(t) = -\frac{\lambda_2\lambda_3}{\lambda_1(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)}\, e^{-\lambda_1 (t - t_p)} - \frac{\lambda_3}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} e^{-\lambda_2 (t - t_p)} -$$

$$\frac{\lambda_2}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)}\ e^{-\lambda_3(t-t_p)}$$

$$IntG_{32}(t) = -\frac{\lambda_3}{\lambda_2(\lambda_3-\lambda_2)}\ e^{-\lambda_2(t-t_p)} - \frac{1}{\lambda_2-\lambda_3}\ e^{-\lambda_3(t-t_p)}$$

$$IntG_{33}(t) = -\frac{1}{\lambda_3}\ e^{-\lambda_3 t}$$

$$IntG_{41}(t) = -\frac{\lambda_2\lambda_3\lambda_4}{\lambda_1(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)(\lambda_4-\lambda_1)}e^{-\lambda_1(t-t_p)} - \frac{\lambda_3\lambda_4}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)(\lambda_4-\lambda_2)}e^{-\lambda_2(t-t_p)} - \frac{\lambda_2\lambda_4}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)(\lambda_4-\lambda_3)}\mathrm{e}^{-\lambda_3(\mathrm{t}-\mathrm{t_p})} - \frac{\lambda_2\lambda_3}{(\lambda_1-\lambda_4)(\lambda_2-\lambda_4)(\lambda_3-\lambda_4)}\mathrm{e}^{-\lambda_4(\mathrm{t}-\mathrm{t_p})}$$

$$\mathrm{IntG}_{42}(\mathrm{t}) = -\frac{\lambda_3\lambda_4}{\lambda_2(\lambda_3-\lambda_2)(\lambda_4-\lambda_2)}\ \mathrm{e}^{-\lambda_2(\mathrm{t}-\mathrm{t_p})} - \frac{\lambda_4}{(\lambda_2-\lambda_3)(\lambda_4-\lambda_3)}\mathrm{e}^{-\lambda_3(\mathrm{t}-\mathrm{t_p})} - \frac{\lambda_3}{(\lambda_2-\lambda_4)(\lambda_3-\lambda_4)}\ \mathrm{e}^{-\lambda_4(\mathrm{t}-\mathrm{t_p})}$$

$$\mathrm{IntG}_{43}(\mathrm{t}) = -\frac{\lambda_4}{\lambda_3(\lambda_4-\lambda_3)}\ \mathrm{e}^{-\lambda_3 \mathrm{t}} - \frac{1}{\lambda_3-\lambda_4}\ \mathrm{e}^{-\lambda_4 \mathrm{t}}$$

$$\mathrm{IntG}_{44}(\mathrm{t}) = -\frac{1}{\lambda_4}\ \mathrm{e}^{-\lambda_4(\mathrm{t}-\mathrm{t_p})}$$