

Highlights

- A new system for preparation of radon-in-water reference materials is developed
- The system allows to prepare LSC sources traceable to a radon primary standard
- The system allows to prepare gamma-spectrometry sources traceable to a radon primary standard
- The system allows direct comparison of the radon-in-water measurement methods.

Towards a Radon-in-water primary standard at LNHB

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Abstract

Despite the widespread radon-in-water measurements, no primary radon-in-water standard currently exists. The objective of this work is to address this gap by developing a system for producing radon-in-water reference materials. This system enables the quantitative preparation of radon-in-water concentrations with traceability to a primary radon standard created by the fixed solid angle method.

The operation of the system is based on cryogenic, loss-free transfer of radon from the defined solid angle radon standard to water. The system allows for the radon loss-free preparation of liquid scintillation and gamma spectrometry samples from the same water containing radon in a single experiment. The liquid scintillation samples can be used for absolute radon-in-water measurements using the triple-to-double coincidence ratio (TDCR) method, as well as for calibrating commercial liquid scintillation analysers for radon-in-water measurements.

This paper describes the design and functionality of the system, as well as the results of performance tests conducted during the critical steps of radon-in-water production. Furthermore, feasibility is demonstrated through pilot metrological tests of the entire system.

Keywords: Radon-in-water, ²²²Rn, Primary standard, Liquid scintillation, TDCR, Gamma spectrometry

1. Introduction

Measurement of ²²²Rn activity in water is important in various fields, including dosimetry, radiation protection, geology, and volcanology. Human exposure to ²²²Rn coming from drinking water sources is considered to contribute to the overall radiation dose for the general population. Consequently, many countries have established reference levels for radon concentration in drinking water. For instance, the European Union has set a parametric value of 100 Bq·L⁻¹ for radon-222 in water intended for human consumption (Council Directive 2013/51/EURATOM). The United States Environmental Protection Agency recommends that the concentration of radon in drinking water supplied by water providers should be below 148 Bq·L⁻¹ (Environmental Protection Agency, 1999).

Various methods have been developed for measuring radon-in-water, including emanometry, gamma-ray spectrometry (GS), liquid scintillation counting (LSC) and track-etch techniques. Among these methods, the three most commonly used for demonstrating compliance with legislation are LSC, GS, and emanometry. Despite the significance, extensive

usage, and long-standing history of radon-in-water measurements, there is currently no primary standard for radon-in-water concentration. This lack of a primary standard hinders the establishment of a traceability chain for radon-in-water measurements, as the calibrations are often performed using other nuclides (such as ^{226}Ra standards), which may introduce certain challenges.

For instance, in LSC radon-in-water measurements, calibration sources are typically based on ^{226}Ra solutions and rely on the assumption of secular equilibrium between ^{226}Ra and ^{222}Rn . However, as highlighted by Salonen (2010), if the ^{226}Ra solutions are stored for long periods, long-lived decay products of ^{222}Rn (such as ^{210}Po , ^{210}Pb and ^{210}Bi) can accumulate in the solution. Thus if the ^{226}Ra solution is used for calibration without purification, the activities of these decay products (mainly ^{210}Po) may introduce interference in the LSC calibration (Salonen, 2010). Also, as pointed out by Kitto et al. (2009) regarding the LSC calibration, “the inability to detect transfer losses and radioactive decay, in standardization cocktails that contain radon in equilibrium with ^{226}Ra , limits the reliability of radon results obtained from groundwater samples that do not contain supporting ^{226}Ra ”. In an attempt to avoid this shortcoming, Kitto et al. (2010) prepared radon-in-water standards using a ^{226}Ra -loaded filter sandwiched in polyethylene sheeting placed in a water-filled vials. However, this is still a ^{226}Ra -based standard and, furthermore, the water in the vials that held the encapsulated ^{226}Ra sources were found to contain only 86% of the known activity, after full ingrowth had been attained Kitto et al. (2010).

On the other hand, the calibration of gamma-spectrometry systems is a well-established procedure that usually involves bulk sources with homogeneously dispersed activities in suitable calibration geometries. However, it is important to note that the ^{222}Rn activity in the water is typically estimated from the activities of its short-lived decay products (such as ^{214}Pb and ^{214}Bi) under secular equilibrium. These decay products cannot be assumed to be uniformly dispersed in the water beforehand. For instance, there may be deposition of radon progenies on the walls of the measurement container, leading to differences between the calibration and measurement conditions, and consequently introducing bias in the results.

The precise comparison of liquid scintillation counting (LSC), gamma-spectrometry, and emanometry for radon-in-water measurements is hindered by difficulties associated with water sampling procedures. These procedures are susceptible to radon loss, which can introduce significant variability in the measurement results. Additionally, other interference-causing phenomena are frequently encountered in radon-in-water intercomparisons. Consequently, all these factors may contribute to relatively large variations (e.g., greater than 10%) observed in the results of radon-in-water intercomparisons, even within a single method (see, for example, Jobbágy, 2019, 2020; Kito, 2009; Tzoupko, 2020).

Therefore, from a metrological standpoint, there is a clear need for a pure ^{222}Rn -in-water standard or reference material that ideally possesses traceability to a primary ^{222}Rn standard.

In this work, we have attempted to develop a system that enables the production of radon-in-water reference material with radon concentration traceable to a primary standard. In the following sections, we will describe the concept of the system constructed at LNHB, France, its performance, as well as the results from the pilot experiments conducted with it.

2. Methods and materials

Elements of the system

The developed system is shown schematically in Fig. 1 and in two pictures in Fig. 2. It is a close loop system which has a gamma spectrometry part (volumes V_4 to V_{16}) and a liquid scintillation part (volumes V_{17} to V_{19} and V_3). The system allows to dissolve known ^{222}Rn

activity in a known volume of water and to prepare gamma-spectrometry and LSC sources in a radon-loss free manner.

In the gamma spectrometry part there are five volumes (V_5 , V_7 , V_{11} , V_{13} and V_{15} , denoted also as Gaz1, Gaz5, Gaz9, Gaz7 and Gaz11) which are cylinders made of stainless steel which match the dimensions of volumes that are used for efficiency calibration at LNHB. The volume Gaz7 is used for the ^{222}Rn transfer from the defined solid angle method (see Fig. 3).

The liquid scintillation part consists of four volumes (V_{17} , V_{18} , V_{19} and V_3). By design, it allows to produce LSC samples by pushing the water from the system directly in LSC vials through a needle attached to the system after the valve B_8 . This is achieved as follows: After the homogenization of the radon in the water in the system the gamma spectrometry part is isolated by closing valves B_9 and B_{22} and a dispenser is attached to the system as shown in Fig. 1. The dispenser allows to push a pre-defined volume of radon-free-water in the system through the Inlet 1. Hence, the same volume of water charger with radon leaves the system through the outlet with the needle attached to valve B_6 . The key design part here is that the volume V_{19} is a tube with large length (around 3 m) and small diameter (4 mm) which prevents mixing of the pushed radon-free water with the radon-containing water available in the tube. We have checked experimentally that, pushing a radon-free-water with the dispenser does not compromise the LSC sampling, because of the lack of mixing and thus all the water in V_{19} is available for production of LSC sources.

The other elements of the system are not directly connected to the preparation of the radon-in-water sources, but are essential for the correct system operation. The vacuum pump attached to valve V_5 allows to evacuate the system to a vacuum with residual pressure $< 10^{-5}$ hPa. The reference volume $V_{\text{ref}} = 2223.6(10) \text{ cm}^3$ attached to valve B_1 is traceable to a LNE volume standard. By using V_{ref} and the reference pressure and temperature gauges attached to V_1 one can measure the volumes of the system by applying the ideal gas law. Thus, the volume of the system V_{sys} can be measured with an overall relative standard uncertainty smaller than 0.15% and the gas volumes with relative standard uncertainty smaller than 0.3%. The possibility to accurately measure the volumes of the system with SI traceability, allows to calculate the radon concentration in the water.

The Inlet 2 is used after the experiments to flush with dry air the system down to the exit after valve B_2 . This allows to quickly dry the entire system (takes about 1-2 hours) and prepare it for the vacuum pumping for further experiments.

System operation

By design, the principle steps for the operation of the system are as follows:

- All volumes are evacuated to vacuum with residual pressure lower than 10^{-5} hPa. The reference volume V_{ref} is filled with nitrogen;
- By opening consecutively the valves of the system and making pressure (P) and temperature (T) measurements at each step determines the volumes of the system by means of the ideal gas law;
- The volume V_7 (Gaz7) is then detached and attached to the defined solid angle standard (Fig. 3). There, the ^{222}Rn activity which is measured by the defined solid angle method is transferred cryogenically to Gaz7 and it is then attached again to the system. Note that the volume contains only radon, there is no air in it;
- The system is then evacuated again to vacuum with residual pressure lower than 10^{-5} hPa. This is necessary because during the detachment/attachment of V_7 the volumes V_8 and V_6 are filled with air at ambient pressure. After the evacuation the valves B_6 and B_5 are closed and the system is ready for filling with water.

- A source of degassed and deionized water is fed to the system through Inlet 1 with all the valves being closed. Valve B₂₄ is opened and the water is allowed to enter through the gamma spectrometry volumes step by step, all the way down to valve B₁₃. Then the same is done through the LSC part and volume V₄ all the way down to valve B₁₀. The glass parts in volumes V₁₆ and V₁₈ are used for visual inspection of the process of filling and homogenization of the system. Note that, as long as deionized water is fed to the system which volume has been evacuated to vacuum, the entire volume of the system is filled without residual air in it.
- The bottom of the Gaz 7 volume is frozen in liquid nitrogen (see Fig. 2 right) in order to form a solid source on the bottom of the volume. The liquid nitrogen is then removed and the water is let in the volume by opening simultaneously valves B₁₂ and B₁₃. There is a transient “radon on the rocks” period, until the water melts and absorbs the radon from the bottom of Gaz11. The valve B₂₄ remains open in this period. Note that the radon cannot escape from the system, because of the large distance it needs to travel through the gamma spectrometry and the LSC volumes. When an equilibrium is attained the valve B₁₄ is closed and the radon is contained (and dissolved) in the water in the system in the volume V_{sys} confined between the valves B₆, B₈, B₂₄ and B₂₆.
- Then the air-tight pump (V₉) is switched on for homogenization of the radon activity in the system. The pump allows to change the direction of the circulation, which is done typically 4 times during the homogenization, in order to avoid the potential existence of dead volumes during the homogenization. Thus, a radonated water is created with homogeneous radon distribution in it.
- After the homogenization the valves in the gamma-spectrometry part (B₉ to B₂₂) are closed and the volumes Gaz1, Gaz5, Gaz 7, Gaz9 and Gaz11 are ready to be detached from the system for further gamma-spectrometry measurements and/or radon-in-water source dissemination.
- The LSC part is further used to prepare LSC samples. A needle is attached to valve B₈ and a dispenser is connected to the system (see Fig. 4, left) and the valve B₂₄ is opened again. The dispenser is typically set to push 5 mL of water per push and three preliminary injections are made in order to fill completely the volume of the needle with the radon-containing water from the system.
- There are two techniques to perform LSC measurement of ²²²Rn-in-water. The first technique uses water miscible cocktail, in which the cocktail absorbs the water and forms a homogeneous mixture. The other technique uses water immiscible cocktail which is covering the water in the vial, forming a two phases system (in fact the system in the vials is a three-phases system as there are water, LS cocktail and air in the vial). The radon is transferred from the water to the LS cocktail by vigorous shaking. The two phases cocktail is known to show instabilities in certain cases as shown by Salonen (2010b). Owing to the fact that we are performing an absolute measurement by the TDCR method, we aim at the most accurate measurement with the smallest possible uncertainty. Thus any sources of instability in the system should be avoided. For this reason, water miscible cocktail and one-phase samples have been used in the current study. The LSC samples that are prepared in the experiments are a mixture of 5 mL H₂O charged with radon with 19,5 mL UGAB LS cocktail. In all the experiments PE-PTFE vials with aluminum foil-lined caps were used.
- The LSC sources are prepared by pushing 5 mL of water charged with radon in each LSC vial below 10 mL of LSC cocktail (see Fig. 4). The precise determination of the quantity of water delivered to the vial is done by weighing the vials before and after the water transfer. Eventually, the vials are filled completely with LSC cocktail and carefully closed with aluminum foil-lined caps in order to avoid any air bubbles remaining in them. There are two key elements for this procedure to ensure that it is radon loss-free: First, the element of pulling water charged with radon is completely avoided. The pulling of water charged with radon is

suspected to be a source of radon loss, as there is always under-pressure below the pulling surface and this may cause radon degassing. Second, after the transfer to the LSC vial, the water charged with radon stays always below a centimeter of organic LSC cocktail, which prevents radon loss during the weighing stage.

- The LSC samples that are prepared in these experiments are a mixture of 5 mL radonated H₂O with 19.5 mL UGAB LS cocktail. In all the experiments PTFE vials with aluminum foil-lined caps were used. All LSC sources were carefully filled in order to avoid residual air bubbles in the vials.

- After the preparation of the gamma-spectrometry and the LSC samples, the water from the system is evacuated and the system is flushed with dry air to dry well the internal parts.

The entire process of the preparation of radon-in-water samples can be completed in one day (including the radon standardization through the defined solid angle method). The preparation of the system for a new experiment takes another 24 h (which includes assembly, drying, reaching $< 10^{-5}$ hPa vacuum and system volume measurements).

3. Results

In what follows we will present the results of the pilot tests of the new system.

Testing of reproducibility of the preparation of LSC samples.

In order to test the reproducibility of the process for preparing LSC samples, we conducted four different experiments. For each experiments, ²²²Rn was transferred to water with the system. LSC samples were then prepared and the variance of the transferred H₂O mass was estimated by weighing and measuring of the net counting rate, i.e. blank and decay corrected LS counting rate of the samples using a commercial LS counter (Wallac 1214). The results of these experiments are shown in Table 1 (weighing) and Table 2 (LS counting).

The results in Tables 1 & 2 demonstrate that, with the developed system, we can achieve a good repeatability in preparing LS samples. The repeatability is better than 1% when estimated based on the relative standard deviation of the transferred mass or the mass specific counting rate of the prepared samples.

Absolute measurement of the ²²²Rn mass specific activity in the LSC samples by TDCR counting

The LSC samples are measured on two TDCR systems equipped with different electronics which are described by Sabot et. al (2022). The first system consists of a MicroTDCR counter and a nanoTDCR analyzer (Yantel, USA) and allows four simultaneous TDCR acquisitions with two different coincidence counting windows and two different dead times. The other system consists of a MiniTDCR and CAEN DT5721 digitizer. The digitized data is stored in list mode format and is later analyzed with a dedicated code (Dutsov, 2021).

Coincidence window of 40 ns and extendable dead time is used in all measurements. The counting rate data is corrected for background, decay during measurement and for accidental coincidences (Dutsov et al., 2021).

Due to the relatively short half-life of ²¹⁴Po, a correction for its decay during the dead time has to be applied. Two approaches were used for this purpose – analytical efficiency calculation and extrapolation to zero dead time. The analytical calculation is based on an analytical evaluation of the probability that a ²¹⁴Po nucleus will decay within the dead time of the counter and will be described in a separate publication. The extrapolation to zero-dead time is implemented by making measurements with different dead-time base duration in the case of the

nanoTDCR analyzer and by analyzing the list mode data with different dead-time base durations in the case of CAEN digitizer. The variation of the counting rate as a function of the dead-time base duration and the extrapolation to zero-dead time, are shown in Figs. 5 and 6.

Measurement of the ^{222}Rn mass specific activity in the LSC samples by a commercial LSC counter

The LSC samples are measured also on a commercial LSC counter (RackBeta1214). The counter was previously calibrated for ^{222}Rn measurements with samples that were standardized by TDCR counting as described in the previous part. The objective of these measurements was to study the transfer of the unit to commercial LSC analyzers and to compare the results with the primary measurement methods.

Measurement of the ^{222}Rn mass specific activity in the gamma-spectrometry samples

The gamma spectrometry samples (Gaz volumes) are intended to be measured on the high-purity germanium detectors (HPGe) available at LNHB. These measurements aim to study the transfer of the unit to radon-in-water measurements with HPGe systems as well as to study potential sources of bias due to a possible non-homogeneous distribution of the radon progeny in the samples. A N-type HPGe detector is used (ORTEC, Planar detector, 50% det efficiency, carbon windows, 10 cm lead shielding), which has precise efficiency calibrations for measurements with such sources at distances 0.45 cm and 10 cm (the reference measurement geometry for this detector). For instance, the efficiency calibration for the measurement at 0.45 cm is shown in Fig. 7. The efficiencies for the measurement at 20 cm are obtained by efficiency transfer calculations using the ETNA software (Piton, 2000). The ^{214}Pb 295 keV and 352 keV full energy absorption peaks were used for the analysis. An example of the results obtained in these measurements is shown in the three last lines of Table 3.

The gamma-spectrometry results in Table 3 show a difference between the measurements at 10 cm and 20 cm and that on 0.45, which is significant in view of the estimated uncertainties. Similar results were obtained in the other tests (not shown). Our hypothesis is that this difference is due to the deposition of the radon progeny on the walls of the container, which creates differences between the calibration and measurement conditions, and hence a bias in the activity estimate. Such an effect should be more pronounced in close measurements geometries, which is what is observed in Table 3. Research is ongoing in order to provide a better understanding of this phenomenon.

Comparison between the results of the measurement methods and the production method.

The fundamental capability of the new system that was aimed with its design is to allow direct comparison of the different radon-in-water measurement techniques. This is illustrated hereafter in a pilot experiment, in which a radon source is produced by the fixed solid angle methods and transferred to water with the new system. Gamma-spectrometry and LSC sources were prepared and measured as described in the previous sections. The results of the comparison between the three methods is shown in Fig. 7 and Table 3.

Overall, it can be considered that there is a good agreement between the measurements within the estimated uncertainties. The only outlier is the γ -spectrometry measurement in close geometry (0.45 cm), which is suspected to be due to inhomogeneity of the radon progeny distribution, as suggested above. It should be noted here that the primary measurement methods

(the defined solid angle method and the TDCR counting) allow to measure the ^{222}Rn activity with rather low relative standard uncertainty, typically of the order of 0.5%. Thus, this type of comparison can be considered as a strong experimental test of the consistency between the different measurement techniques.

4. Conclusions

In this work, we present the design and initial tests of a new system for producing radon-in-water reference material that is traceable to a primary ^{222}Rn standard. The preliminary tests of the system confirm the feasibility of the design and demonstrate the potential to establish traceability to the ^{222}Rn primary standard defined by the defined solid angle method. Additionally, the system allows for the measurement of prepared radon-in-water concentration using another absolute method, specifically TDCR counting.

Furthermore, the system enables the preparation of gamma-spectrometry and LSC sources from the same water loaded with radon in a manner that avoids ^{222}Rn losses. This capability facilitates a sensitive comparison of results obtained using these measurement methods. The presented comparison results indicate that the developed system has the potential to serve as a cornerstone in establishing a radon-in-water traceability chain from in-situ laboratory measurements to the primary ^{222}Rn standard. Moreover, the system helps identify subtle effects that may introduce bias in measurements, such as variations in gamma-spectrometry results in close measuring geometries.

Ongoing research aims to explore the full potential of the system. This includes assessing the repeatability of radon-in-water production, conducting a more detailed comparison of radon-in-water measurement methods across a range of concentrations, and experimentally evaluating approaches for disseminating the ^{222}Rn -in-water concentration unit in laboratory measurements.

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Declaration of competing interest

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

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Figure captions

Figure 1: A scheme of the experimental system.

Figure 2. Pictures of the system during its development. Left: measurement of the volumes of the system and preparation for filling with water; Right: end of the cryogenic radon transfer just before filling the system with water.

Figure 3. Left: Transfer of ^{222}Rn to the Gaz7 volume after its standardisation by the fixed solid angle method (Picolo, 1996; Sabot et al., 2016); Right: The Gaz7 volume which is used to transfer the ^{222}Rn from the fixed solid angle method to the new system. The Gaz7 volume is further attached to the system for production of a radon-in-water source (see Fig. 2, right).

Figure 4. Preparation of a LSC source with the new system. Left: The dispenser for water pushing is attached to the system. Middle and right: snapshots of different steps during the preparation of the LSC source. Please note that the glass vial in the picture is used for illustration, while PTFE vials were used throughout the measurements.

Figure 5. TDCR measurement with the MiniTDCR and the CAEN digitizer. Top: Variation of the counting rate of the logical sum of double coincidences (D) as a function of the dead-time base duration and extrapolation to zero dead-time. Bottom Activity of the sample vs. the dead-time base duration as obtained by the analytical method.

Figure 6. TDCR measurement with the MicorTDCR and the nanoTDCR digitizer. Top: Variation of the counting rate of the logical sum of double coincidences (D) as a function of the dead-time base duration and extrapolation to zero dead-time. Bottom Activity of the sample vs. the dead-time base duration as obtained by the analytical method.

Figure 7. The efficiency calibration curve for the γ -spectrometry measurement of the 50 cm^3 volume (Gaz 11) in close measuring geometry (0.45 cm from the detector).

Figure 8. Comparison between the results of the different methods obtained in one of the pilot experiments with the new system.

Table captions

Table 1. Mean mass (\bar{m}) and relative standard deviation of the mass in an individual sampling (δ_m) of the water transferred to the LSC cocktail in each experiment.

Table 2. Mean (decay and background corrected) counting rate (\bar{n}_0) and relative standard deviation of the individual counting rate (δ_{n_0}) obtained in the LSC measurements of the prepared samples.

Table 3. Mass specific activities of the ^{222}Rn -in-water as determined by HPGe measurements on three different distances. The values in the brackets represent the overall estimated standard uncertainties.

Figure 1

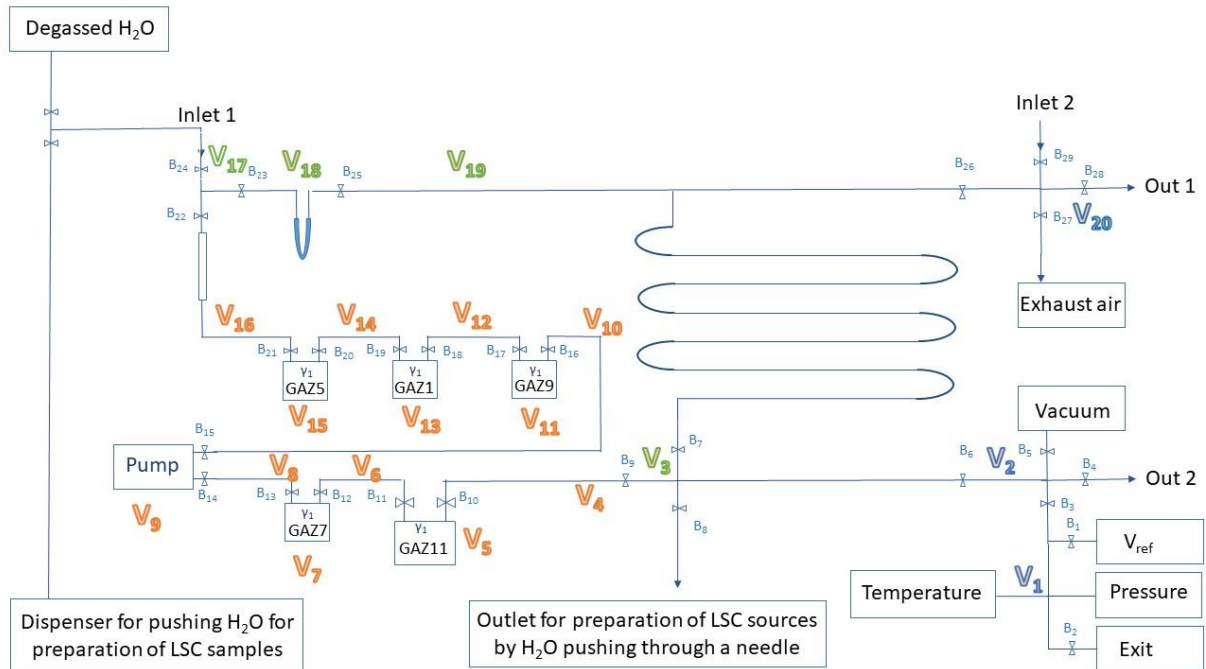


Figure 2



Figure 3

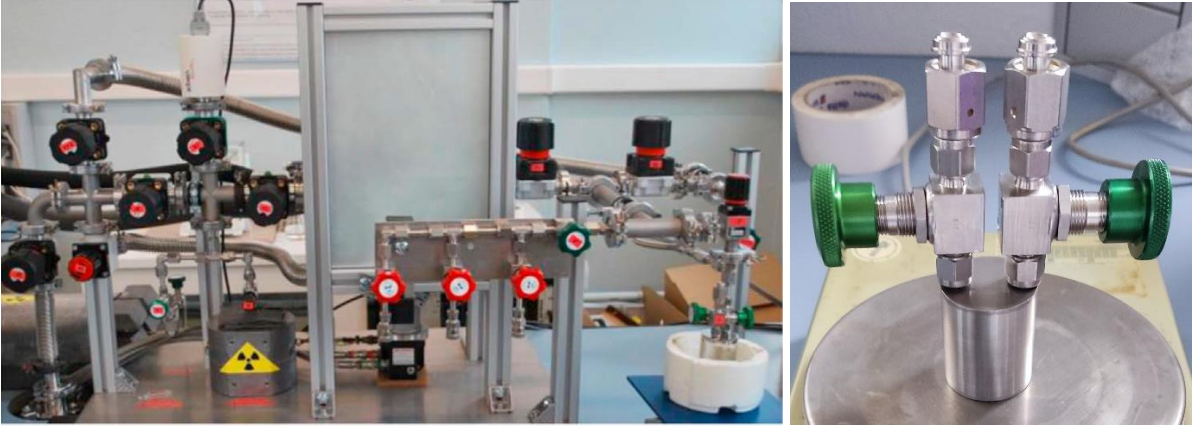


Figure 4



Figure 5

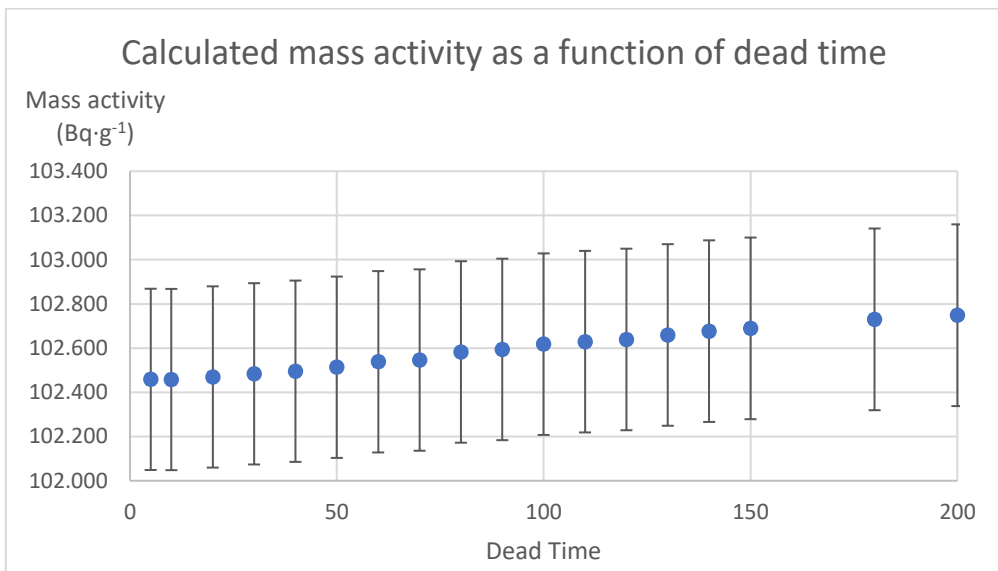
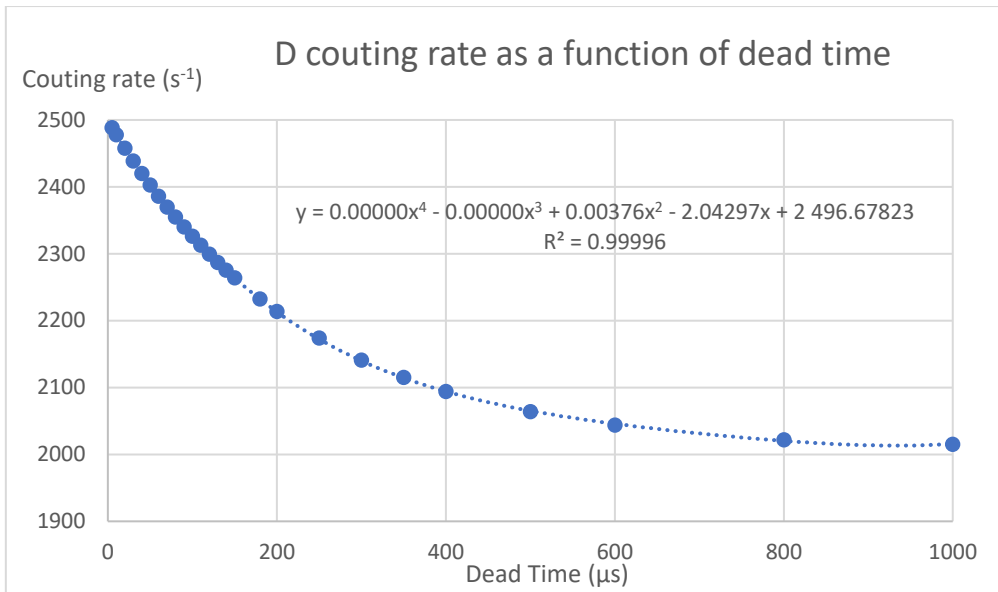


Figure 6

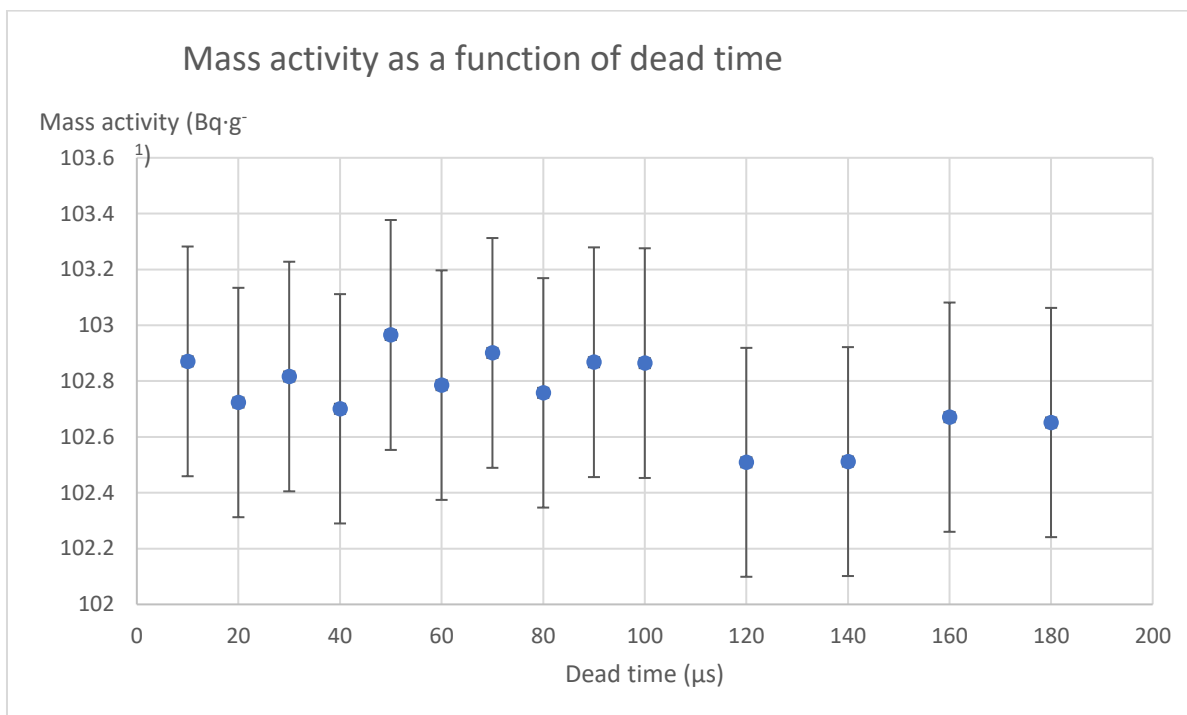
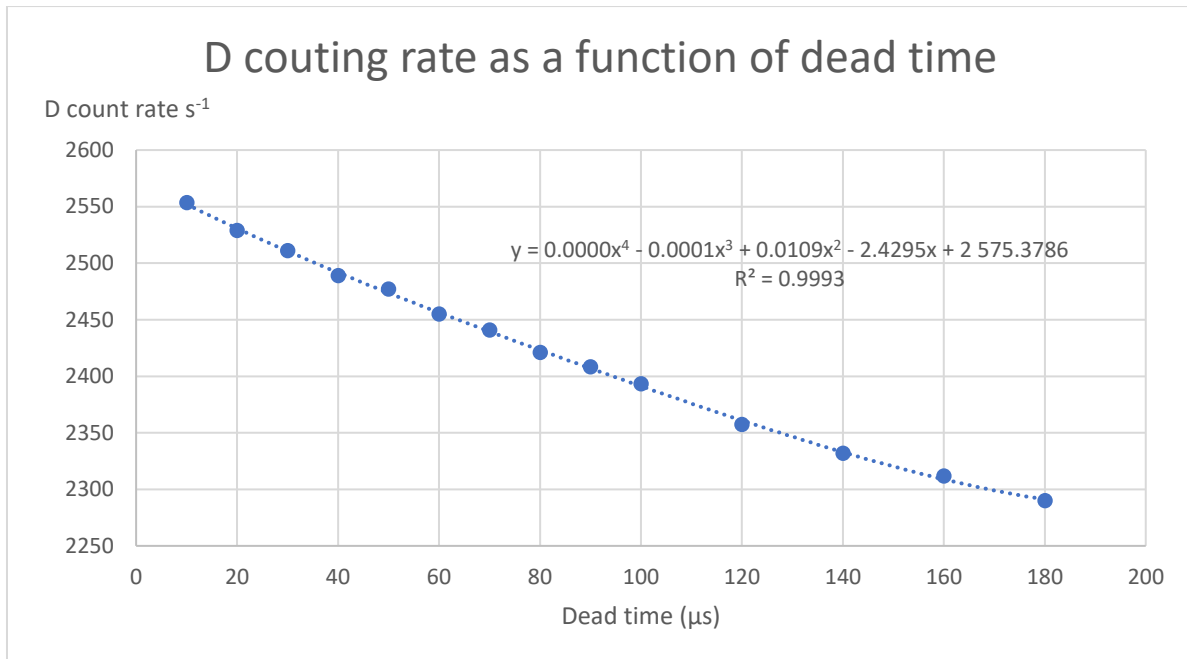


Figure 6

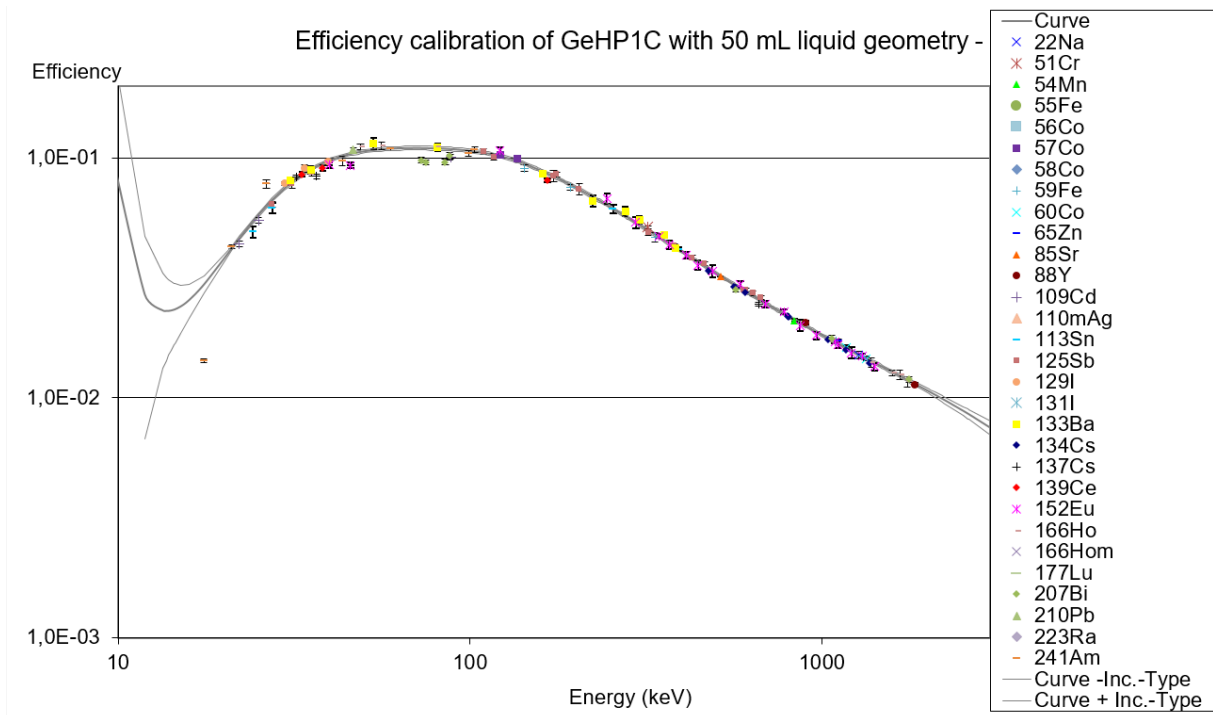


Figure 7.

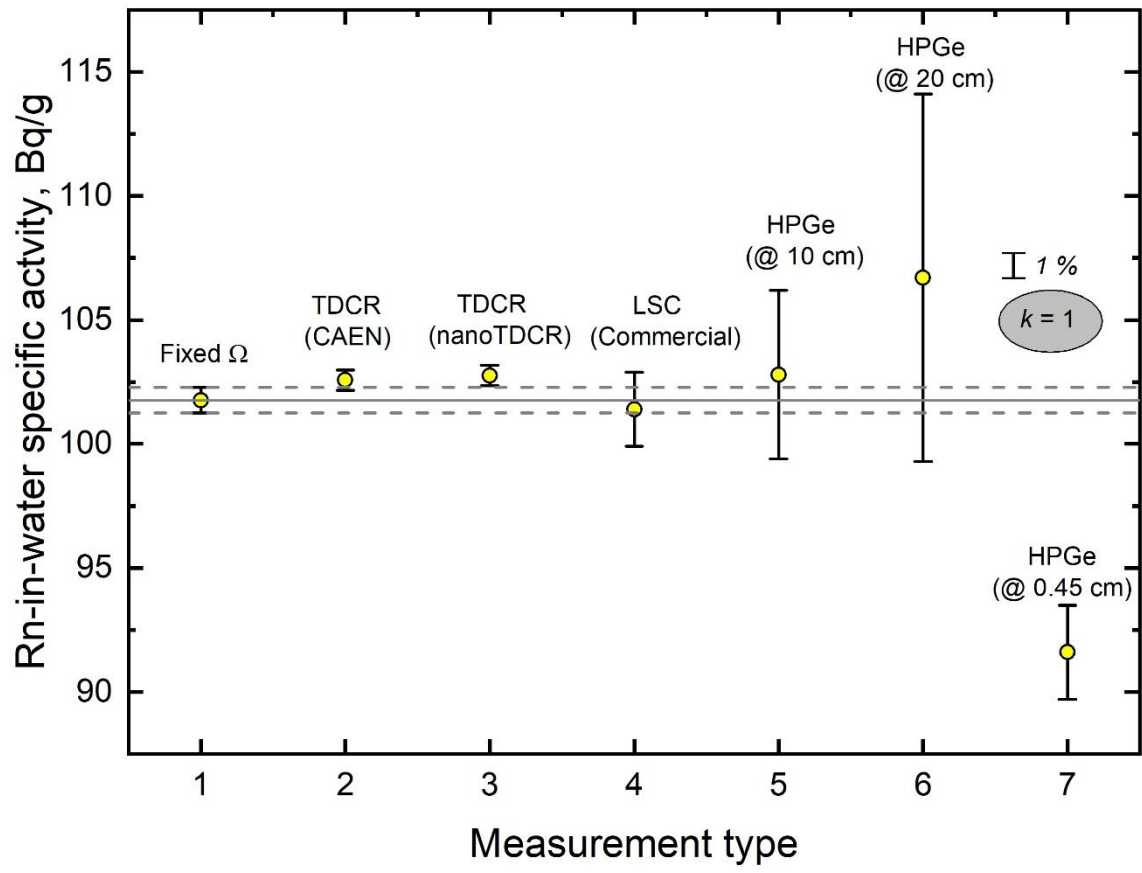


Table 1

Experiment	# samplings	\bar{m} , g	δ_m %
1	5	4.978 (11)	0.23 %
2	5	4.974 (19)	0.37 %
3	5	4.861(23)	0.47 %
4	8	5.009(12)	0.25 %

Table 2

Experiment	# samplings	\bar{n}_0 , cps/g	δ_{n_0} , %
1	5	507.2 (13)	0.25 %
2	5	1101.5 (30)	0.27 %
3	5	860.5(60)	0.70 %
4	8	334.34(65)	0.19 %

Table 3

Measurement method		Description	^{222}Rn -in-water concentration, $\text{Bq}\cdot\text{g}^{-1}$
1	Defined solid angle method	Rn-222 concentration determined by the defined Ω method and the volume of the new system	101.77(51)
2	TDCR counting	MiniTDCR counter and CAEN digitizer	102.58(41)
3	TDCR counting	MiniTDCR counter and CAEN digitizer	102.76(41)
4	LSC counting	RackBeta1214 LSC counter	101.4(15)
5	γ -spectrometry	Volume Gaz 11 (50 cm^3) at 10 cm	102.8(34)
6	γ -spectrometry	Volume Gaz 11 (50 cm^3) at 20 cm	106.0(76)
7	γ -spectrometry	Volume Gaz 11 (50 cm^3) at 0.45 cm	91.6(19)