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# Radon leakage in LSC vials: Material-dependent analysis for utilization and reutilization

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ABSTRACT

Liquid scintillation counting (LSC) is a method commonly used for natural occurring radionuclides measurements in water, in which radon concentration is measured after placing water samples in LSC vials that contain scintillation cocktails. Due to the low levels of activity that are involved in natural environments, it is required to characterize any radon loss during the measuring process in order to minimize errors and uncertainties. Hence, radon leakage in different types of vials (plastic and plastic lined with Teflon) is studied. This analysis reported that, after one and a half radon half-lifes, plastic and Teflon lined vials, compared with the values expected for no radon leakage, presented deviations in radon concentration over the 7% and 3%, respectively. Additionally, the diffusion of radon through the vials' wall is checked, as well as if it remains embedded in the polymeric matrix. This study confirmed that radon remains embedded in the polymeric matrix and can diffuse back to the vial, having the plastic vials a greater contribution. The possibility to re-utilize single-use vials is also studied from characterizing the presence of radon depending on the vial's material.

## 1. Introduction

Liquid scintillation counting (LSC) is an effective and efficient radiometric method applied both in beta and alpha emitters measurements (Hidex 600 SL, n.d.). It is used in several applications such as the decommissioning of nuclear facilities, in routine monitoring works for medical research and nuclear facilities, as well as in rapid analysis for emergency management (Hou, 2018). It is also used in the analysis of environmental processes and to determine naturally occurring radionuclides with low levels of radiation, such as $^{222}$Rn, from now on radon. Due to the simplicity of LSC sample's preparation and the high sensitivity of the method, radon in water is usually measured by this technique. LSC working principle consists of the detection of low intensity light that is produced when the energy originated during a radioactive decay interacts with the scintillator (Hidex 600 SL, n.d.). The sample preparation consists of filling the LSC vial with scintillation cocktail and inserting the water that contains radon into the core of the cocktail previously disposed.

Radon migrates within a medium based simultaneously on diffusive and advective flows, which are respectively expressed by Fick's and Darcy's laws. When the mobility of the gas occurs towards areas with lower concentration, the transport is by diffusion. The advective flow depends on the permeability of the medium, meaning its capacity to transmit the gas upon pressure gradients (Mollo et al., 2018). Furthermore, the permeability depends on the combined effect of radon diffusion and adsorption processes (Fernández et al., 2004). Due to radon gas not being highly soluble in water, the radon contained in it, when it is placed inside of plastic vessels, can be lost through its surface by these radon transport processes: adsorption or diffusion. Previous literature points out that glass containers do not loss radon during storage periods, in contrast when using high density polyethylene (HDPE) for radon storage, that up to 10% of it may be lost per day (Saito, 1983). Plastic vials (HDPE) used in LSC are considered low diffusive to radon, even that previous bibliography indicates that, for long time storages, they might induce high uncertainties into the analysis due to a slow radon loss (Hou, 2018). As LSC samples mix radon in water with scintillator cocktails, due to radon being more soluble in organic solvents than in water, organic scintillation cocktails are applicable for radon analysis and may prevent this radon leakage. Anyway, some radon losses can be expected through the mentioned transport processes.

Due to the low levels of radiation involved when measuring natural occurring radionuclides any loss or leakage may influence the measuring process. This has driven to studies that analyzed the tightness of the vials used as containers to measure radon in water samples (Vitz, 1991), where differences between glass and plastic vials had been reported. This aligns with the statement about the necessity to check radon

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tightness of each vials' material, particularly when the samples cannot be measured up to one day or longer after its sampling (Jobbágy et al., 2019).

The scintillation cocktail used for radon in water measurements in this work corresponds with the Hidex MaxiLight + cocktail. When using this scintillator, after the preparation of the sample, 3 h must pass until radon is in equilibrium with the short-lived alpha-emitters of its progeny and then, the measurement can be initiated. This time is equivalent to six half-lives of each short-lived radon progeny, period necessary to reach the secular equilibrium between radon and progeny (L'Annunziata, 2023). These 3 h are the minimum time elapsed between preparation and measurement, however, when the measurements are part of field duties the sample needs to be transported to the location of the detector, inducing even greater delays. The vials used for radon determination by LSC can be made from different materials such as plastic, plastic lined with Teflon or glass. Glass vials are resistant to diffusion; however, they are fragile, which can pose a problem during field duties. In contrast, plastic and Teflon lined vials, due to their greater resistance to any potential impact, do not present this issue. Furthermore, plastic vials offer ultra-low backgrounds and are significantly more affordable (Vials, n.d.). These observations lead to consider in this work plastic and Teflon lined vials for the radon measurements. Hence, the LSC vials to study correspond with the ones that are not considered gastight to radon: plastic and plastic lined with Teflon, which, in case of measurement delays, might introduce uncertainties to the tests.

As radon loss can interfere in the results, a hypothesis of how it occurs is considered. The premises are that radon might be diffusing through the vial's wall and that this diffusion could be influenced by the container's material. In the same way that the radon contained in the water tends to scape by migration through the polyethylene wall (Hou, 2018), it might be happening also in presence of scintillation cocktail in spite of being radon more soluble. This leads to question how radon behaves depending on the material of the vial when it contains the water sample mixed with the scintillator and if these vials can be re-utilized after its first use. Then, the main objectives of this work are to evaluate the radon leakage in LSC vials that are used for radon in water measurements; to study the influence of vial's material in possible leakage and to check if single-use vials can be re-used in further measurements.

## 2. Methods and materials

The methodology used during the research consists of three experiments: the first one to test radon leakage from plastic (HDPE), Teflon lined plastic and glass vials, a second experiment to study the diffusion of radon through the plastic and Teflon lined vial's wall, and a third test to check the presence of radon in plastic and Teflon lined vials' wall after its first use.

Water samples enriched in radon are used for all the set of experiments. To enrich the radon content of the water samples, tap water is placed inside of an airtight container together with a hermetically sealed radon source of 92.94 $kBq{\cdot}kg^{-1}$ of activity, that acts as a radon source during a time around 15 days in which water reached a value of 6000 Bq/L of radon concentration. The activity of the radon source was determined by measuring the activity of $^{214}Pb$ (in equilibrium with radon) by gamma-spectrometry in a germanium detector (Noverques, 2022).

### 2.1. Verification of radon leakage from the vials

This study intends to analyze if the radon concentrations of LSC vials containing a mixture of radon in water and scintillation cocktail evolves as it would be expected by following the decay law. To confirm that radon loss occurs in LSC vials, an experimental procedure is planned. After filling the vials with a sample of radon in water and closing them, the container is measured repeatedly over the time. By this, the evolution of radon in water concentration of the container is determined, which can be compared to the expected evolution that would take place according to the decay law. Two types of material are tested to study radon leakage: vials made of high-density polyethylene (plastic) and the same plastic coated with Teflon. As glass vials are highly resistant to radon diffusion (Vials, n.d.), it is assumed that radon leakage does not occur. In order to verify that glass vials are gas-tight to radon, additionally, the analysis of the evolution of radon in the vials is also performed for these vials. The analysis of the three materials: plastic, plastic Teflon lined, and glass vials is made by LSC, measuring them in a scintillation detector Hidex 600 SL. The vial's preparation for these measurements consists of filling these 20 mL containers of total capacity with 12 mL of Hidex MaxiLight + scintillation cocktail, and sampling 8 mL of the water containing radon that is introduced immediately in the core of the liquid. To ensure that the radon contained in the vial is in equilibrium with its $\alpha$-emitter short lived progeny ($^{218}Po$ and $^{214}Po$), the vials are left to rest for 3 h until radon and progeny are in equilibrium. Once this time is elapsed, they are measured each 2 h and a half repeatedly over the time for 6 days to monitor the mentioned evolution.

### 2.2. Radon diffusion test

The second test aims to determine if there is a radon leakage through the vial's wall when it remains filled with the radon in water sample and scintillation liquid. A setup is designed to confirm that radon escapes through the vial's wall. The configuration is based on the closed compartment model (CCM) (Sainz et al., 2016), and consists of an airtight container connected to a Real-time Continuous Radon Monitor RAD7, forming a closed loop as shown in Fig. 1. The test is performed in plastic and Teflon lined vials containing radon in water enriched at 2000 Bq/L mixed with the scintillator and vials containing a standard solution of $^{226}Ra$ in equilibrium with radon at 900 Bq/L.

After putting the vial in the center of the container, it is closed and the RAD7 Monitor is turned on to register the evolution of the radon concentration in the air of the closed loop for 250 h. Additionally, to differentiate the tests from the background of the setup, another test is carried out when no LSC vial is inside of the container that acts as a chamber. The evolution of the concentration of radon in air is recorded, registering any contribution of radon from the vials that would verify that there is radon diffusion though the vials' walls.

### 2.3. Radon presence after a first vials' use

The LSC vials are examined after its first use to review if they are suitable for their utilization in further LSC measurements. The aim of this test is to check if, when reusing vials, there could be any radon contribution to the mixture coming from LSC vials. First, vials made of both plastic and Teflon lined, that were filled for 6 days with scintillator and water with a radon concentration of 6000 Bq/L, are emptied. After that, they are rinsed with distilled water, filled back following the 12/8 mL proportion with distilled water and scintillator liquid, and they are measured for the first time after the 3 h required to reach the

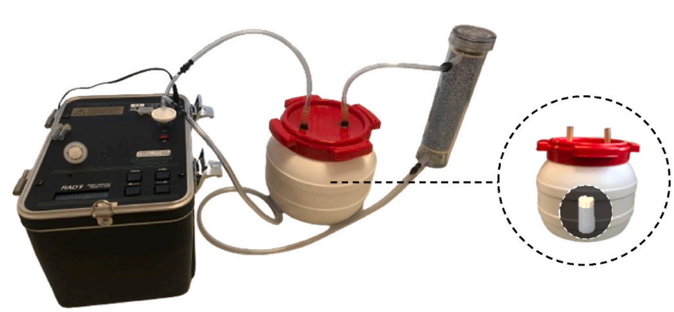

**Fig. 1.** Radon diffusion test setup.

equilibrium between radon and its progeny. Radon presence in each type of vial is recorded from monitoring the evolution of radon concentration by measuring them repeatedly each 2 h in the Hidex 600 SL detector. The concentration of radon in the refilled vials is analyzed in order to register any radon contribution from the vials wall or if it decays as it would be expected by following the decay law.

## 3. Results

This section presents the results obtained from the study of LSC vials including the discussion of the different tests performed.

### *3.1. Verification of radon leakage from the vials*

Radon loss in vials composed of plastic, plastic coated with Teflon and glass is studied by measuring sealed vials repeatedly and monitoring the evolution of their radon in water concentrations. Fig. 2 presents the data evolution for each type of material and displays together the evolution of radon concentration that, following the decay law, would be expected if there was no radon loss. As the difference between values measured and the expected trend tends to increase over the time, additionally, Fig. 2 presents zoomed of the last 50 h of each test.

It is noticeable that plastic and plastic coated with Teflon vials present less concentration than the theoretical decay. This behavior suggests that part of the radon enclosed in the vial is escaping from the control volume. Meanwhile, glass vials follow the tendency of the expected decay, indicating that in this type of vial there is no noticeable radon loss throughout the 140 h of duration of the test. The interval chosen corresponds to 1.5 radon half-times, period that is not expected to be exceeded between the sampling and measurement, even when performing field duties. This observation agrees with the findings of previous works, which stands that, even though glass vials introduce greater intrinsic background than plastic vials, they prevent radon diffusion through the material (Jobbágy et al., 2019). Analyzing the difference between last radon concentration values (the final 10 h of the trials) and the corresponding expected ones, glass vials present a mean deviation under the 1%, meanwhile plastic and plastic coated with Teflon vials reach values over the 7% and 3%, respectively. The observable differences between the experimental and theoretical evolution results in a variation when fitting the data and comparing it to the

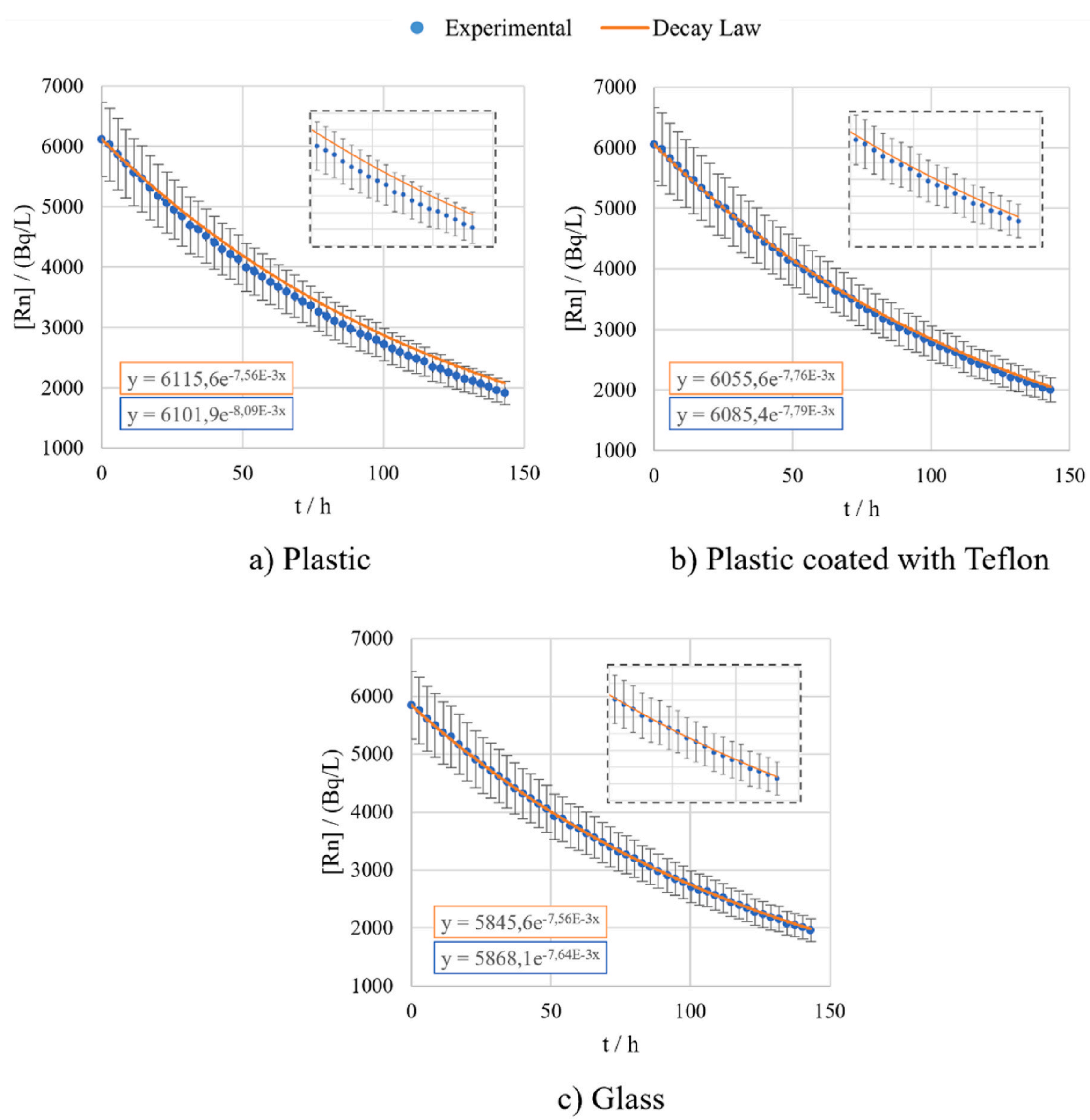


**Fig. 2.** Radon concentration evolution of the vials: a) Plastic, b) Plastic coated with Teflon and c) Glass.

expected evolution from the decay law. The fitting equations have been included in Fig. 1, corresponding the one highlighted in blue with the experimental fit and the orange highlighted with the theoretical equation. Table 1 summarizes the fittings, where the radon decay constant ($\lambda_{Rn}$) is compared with the exponent observed from the fitting ($\lambda_{obs}$) and presented together with the associated relative error ($\varepsilon_r$).

The relative errors of the exponential fittings agree with the observed deviations. Plastic vials present the highest relative error at 7.01%, meanwhile for Teflon lined and glass vials is 3.04% and 1.06%, respectively. Consequently, this test suggests that the greatest radon loss would occur in plastic vials, being reduced when they are Teflon lined and avoided in the case of using glass vials instead.

Once radon leakage is confirmed in plastic and plastic coated with Teflon vials, the next tests are made in order to determine which part of the leakage corresponds to radon diffusion through the vials' wall and which part could remain embedded in the polymeric matrix of the wall, not being in contact with the scintillator and, consequently, not being measured.

### 3.2. Radon diffusion test

This test is performed to verify that the radon present in the water sample is lost through the vials' wall. The diffusion of radon through the vials' walls is checked in vials of plastic and plastic coated with Teflon, as both materials presented radon loss during the previous test.

After placing the vials that contain radon in water samples with scintillator cocktail or the $^{226}$Ra standard solution that acts as a source of radon, inside of the container and forming the closed loop (Fig. 1), the RAD7 registers an increase of radon concentration in air. Fig. 3 presents as an example of the measurements performed the evolution of radon concentrations in air for two Teflon lined vials (Vial 1 and Vial 2), also including the measurement of the system's background. From the reported evolution, it is noticeable that, radon has accumulated in the air of the container, verifying that radon escaped by diffusion through the vials' wall. The results for the plastic vials were analogous to the shown for the Teflon lined. The significance of this radon loss will depend on the elapsed time since the sample is collected until the measurement is performed. Therefore, in the cases that there is a delay between sampling and measuring, such as in field radon measurements, where samples are transported for measurement after collection, it might be relevant to consider that radon loss is occurring and how this might affect the results.

### 3.3. Radon presence after a first vials' use

This test is carried out to determine if part of the reported radon leakage is originated from radon being retained in the polymeric matrix of the vials' wall. During the diffusion, part of the radon that penetrated through the wall could remained trapped inside. In this case, during an immediate second use of the vials, it would be observed a radon contribution from the vials wall. To verify this premise, emptied and cleaned vials of the types that presented radon leakage (plastic and plastic coated with Teflon) are monitored. By periodic LSC measurements of the radon concentrations in water of the vials that have been emptied, cleaned and refilled with scintillator and distilled water, it is possible to register if there is any radon contribution to the mixture. Fig. 4 displays the periodic LSC measurements performed, meanwhile Table 2 presents the key values obtained, where $t_1$ corresponds to the first measurement performed, $t_{1/2\ (Rn)}$ to the measurement at time equivalent to one radon half-life and Max to the highest value recorded.

**Table 1**
Key values from radon leakage fitting.

| $\lambda_{Rn}$ ($h^{-1}$) | Type of vial | $\lambda_{obs}$ ($h^{-1}$) | $\varepsilon_r$ (%) |
|---|---|---|---|
| 7.56 $10^{-3}$ | Plastic | $8.09 \cdot 10^{-3}$ | 7.01 |
| | Teflon lined | $7.79 \cdot 10^{-3}$ | 3.04 |
| | Glass | $7.64 \cdot 10^{-3}$ | 1.06 |

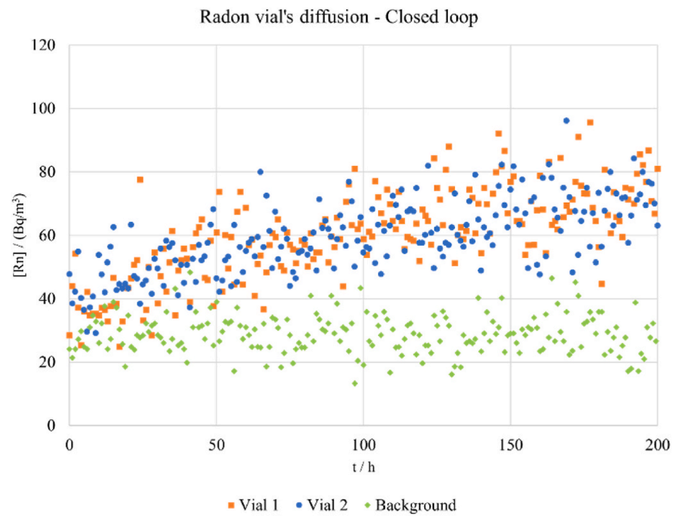


**Fig. 3.** Radon diffusion through vial's walls.

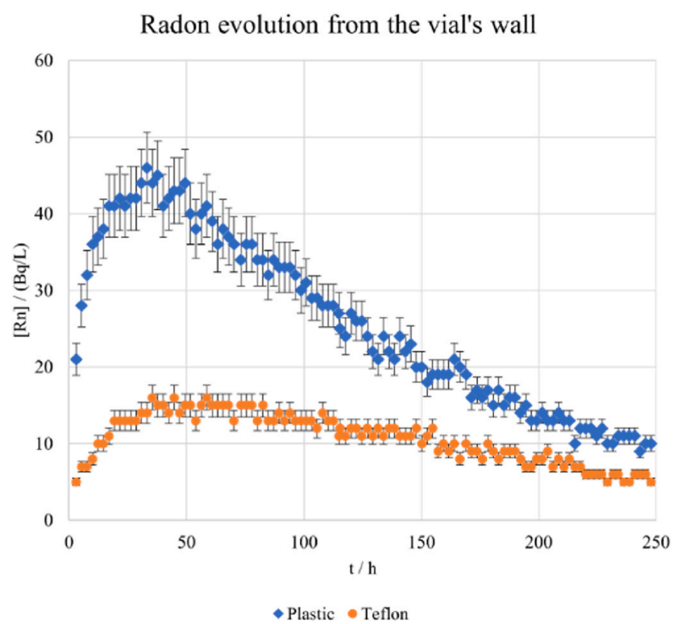


**Fig. 4.** Radon evolution from the vial's walls after refilling the vials.

**Table 2**
Key values from radon evolution after refilling the vials.

| Type of vial | $t_1$ (Bq/L) | $t_{1/2\ (Rn)}$ (Bq/L) | Max (Bq/L) |
|---|---|---|---|
| Plastic | 21 ± 2.1 | 33 ± 3.3 | 46 ± 4.6 |
| Teflon lined | 5 ± 0.5 | 14 ± 1.4 | 16 ± 1.6 |

In both cases, there is an initial increase of radon in water concentration that later begins to decay. As the radon concentration increases during the first 48 h of the test, the possibility that the measured radon originates from residues adhered to the vial's wall is ruled out. The raise observed between the first measurement compared to the value obtained after on radon half-time, suggests that the radon that stayed embed in the vial's wall may be diffusing back to the mixture. Additionally, the values reported from plastic vials are higher than in the case when the vial is coated with Teflon. This observation agrees with what would be expected after the preliminary radon loss test. As plastic vials presented the greatest radon loss, this might imply that more quantity of radon diffuses through its walls and then, a greater portion of it may be trapped in the polymeric matrix.

Considering the reusing of the vials, it could be possible as soon as the radon embedded in the polymeric matrix decays. The elapsed time selected for these range of activities to spend before re-utilizing the vials corresponds with 6 half-lives of the radionuclide (approximately 23 days), as at this time the remaining activity in the vial will be 64 times smaller, which will be considered negligible.

## 4. Conclusions

This work presents a study on the characteristics of vials of different materials used for liquid scintillation measurements. The tests performed show that, even though the LSC vials contain the scintillator cocktail where radon is highly soluble compared to the water, there is a radon leakage in vials made from plastic and plastic coated with Teflon, which does not appear in the case of glass vials. The test showed a greater radon leakage in plastic vials when compared with Teflon lined, as it presented, respectively deviations over 7% and 3% compared to the expected values for no leakage. Additionally, by the subsequent experiment, the diffusion through the vials' wall has been corroborated. Future works will focus on determine a correction factor that will allow to correct the radon concentration measurements depending on the time elapsed from the vial's preparation until it is counted.

The radon presence in the vial's wall test revealed that part of the radon that is passing through these walls remains retained in polymeric matrix of the vial and can diffuse back when they are refilled with distilled water and scintillation liquid. Plastic vials reported greater values of radon concentration than when using plastic coated with Teflon vials. In order to reuse both type of vials, after cleaning them, it will be necessary to postpose their re-utilization during several radon half-lives until the remaining activity can be considered negligible for the next measurements.

As plastic vials are the cheapest, followed by Teflon lined vials, the use of vials made of each material will depend either on the operating conditions, the accuracy needed on the measurement and the expected delay since the sampling time until the vial's measurement. Therefore, whenever possible, it will be used glass vials. However, given its potential to break during handling and transporting them, the Teflon lined, and plastic vials will be chosen depending on the accuracy and expected delay of the measurements to perform.

## CRediT authorship contribution statement

**C. Trull-Hernandis:** Writing – review & editing, Writing – original draft, Validation, Methodology, Investigation, Conceptualization. **A. Noverques:** Writing – review & editing, Validation, Methodology, Investigation, Conceptualization. **B. Juste:** Writing – review & editing, Validation, Supervision, Methodology, Conceptualization. **M. Sancho:** Writing – review & editing, Validation, Supervision, Methodology, Conceptualization. **G. Verdú:** Writing – review & editing, Validation, Supervision, Funding acquisition.

## 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.

## Data availability

Data will be made available on request.

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## References

Fernández, P.L., Quindós, L.S., Sainz, C., Gómez, J., 2004. A theoretical approach to the measurement of radon diffusion and adsorption coefficients in radonproof membranes. Nucl. Instrum. Methods Phys. Res. Sect. B Beam Interact. Mater. Atoms 217 (1), 167–176. https://doi.org/10.1016/j.nimb.2003.09.027.

Hidex 600 SL (n.d.) Automatic Liquid Scintillation Counter Handbook..

Hou, X., 2018. Liquid scintillation counting for determination of radionuclides in environmental and nuclear application. J. Radioanal. Nucl. Chem. 318, 1597–1628. https://doi.org/10.1007/s10967-018-6258-6.

Jobbágy, V., Stroh, H., Marissens, G., Hult, M., 2019. Comprehensive study on the technical aspects of sampling, transporting and measuring radon-in-water. J. Environ. Radioact. 197, 30–38. https://doi.org/10.1016/j.jenvrad.2018.11.012.

L'Annunziata, M.F., 2023. Radionuclide decay, radioactivity units, and radionuclide mass. En Radioact. 865–883. https://doi.org/10.1016/B978-0-323-90440-7.00025-9. Elsevier.

Mollo, S., Tuccimei, P., Soligo, M., Galli, G., y Scarlato, P., 2018. Advancements in understanding the radon signal in volcanic areas: a laboratory approach based on rock physicochemical changes. In: Integrating Disaster Science and Management: Global Case Studies in Mitigation and Recovery, pp. 309–328. https://doi.org/10.1016/B978-0-12-812056-9.00018-X.

Noverques, A., 2022. Estudio del comportamiento del gas radón (222Rn) en los procesos de transferencia en agua y en aire [Doctoral dissertation]. Universitat Politècnica de València. https://doi.org/10.4995/Thesis/10251/185792.

Saito, M., 1983. Correction for loss of radon-222 in water sample caused by the use of a polyethylene bottle. Radioisotopes 32 (3), 109–112. https://doi.org/10.3769/radioisotopes.32.3_109.

Sainz, C., Rábago, D., Fuente, I., Celaya, S., Quindós, L.S., 2016. Description of the behavior of an aquifer by using continuous radon monitoring in a thermal spa. Sci. Total Environ. 543, 460–466. https://doi.org/10.1016/j.scitotenv.2015.11.052.

Vitz, E., 1991. Toward a standard method for determining waterborne radon. Health Phys. 60 (6), 817–829. https://journals.lww.com/health-physics/toc/1991/06000.