

# Optimization of AlphaGuard AquaKIT set-up for analysis of radon in water using stainless-steel bottles and its validation through IAEA standard samples

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## Abstract

In this study, a system composed of an ionization chamber AlphaGUARD (PQ2000), an AquaKIT set and an AlphaPUMP was calibrated to measure  $^{222}\text{Rn}$  in water. First, the system was checked for leakages using a NIST traceable  $^{226}\text{Ra}$  source. Then, we tested polyethylene terephthalate (PET) and stainless-steel bottles for sample storage estimating radon loss over time. Results showed that stainless-steel was a better suitable material for sample storage. Additionally, system setup was further validated by measuring IAEA reference materials issued within the framework of several interlaboratory intercomparisons. Finally, the procedure was applied to bottled mineral and tap waters collected in Sevilla (Spain) including annual effective dose for ingestion for adults children, and infants.

Keywords: Radon in water; Certified material; PET; stainless steel; AlphaGUARD; effective dose

## 1. Introduction

Ingestion of water or inhalation of air containing degassed radon from water increases exposure to  $^{222}\text{Rn}$  dissolved in drinking water (World Health Organization (WHO), 2018). Accurate analyzing of  $^{222}\text{Rn}$  in drinking water is therefore essential to evaluate the risk of exposure. This risk has led to changes in European legislation through the Council Directive 2013/51/Euratom of 22 October 2013 (Council Directive 51/EURATOM, 2013) which establishes a range of between 100 and 1000 Bq  $\text{l}^{-1}$  for radon in drinking water. Those limits have been introduced into Spanish legislation (Real Decreto 314/2016, 2016) setting a limit for radon concentration in drinking water of 500 Bq  $\text{l}^{-1}$ .

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World Health Organisation gives no recommended technique in its guidelines to measure radon concentration in drinking water (World Health Organization (WHO), 2018). However, a limit of detection of  $10 \text{ Bq l}^{-1}$  must be achieved according to the European Council directive 2013/51/Euratom. Several direct and indirect techniques have been used for determining  $^{222}\text{Rn}$  levels in water: gamma-ray spectrometry, emanometry and liquid scintillation counting (Jobbágy et al., 2017; Pujol and Pérez-Zabaleta, 2017; Wiedner et al., 2018). Emanometry is based on transferring radon dissolved in water to air. A water sample is degassed by air circulation or an inert gas flow transferring  $^{222}\text{Rn}$  to a measurement system. Therefore, several detection techniques may be applied to determine  $^{222}\text{Rn}$  concentration activity, including alpha scintillation, an ionisation chamber, or a silicon detector. All of the detection techniques comply with the required detection limit (Jobbágy et al., 2017).

Measurements results in a laboratory are sensitive to sampling, transport and storage (Gruber et al., 2009). Besides sampling, storage conditions, storage time and container material strongly influence the results. Different materials have been studied for containers founding that polylactic biopolymer (PLBP) and polyethylene terephthalate (PET) are better materials than polyethylene (HD/LDPE) (Jobbágy et al., 2019b; Lucchetti et al., 2016; Pujol and Pérez-Zabaleta, 2017). However, there is a need to explore the use of new, shatterproof and recyclable materials as radon containers in order to avoid accidental breaking and keep radon inside, particularly for use in in-situ analysis (Jobbágy et al., 2019b).

On the other hand, ISO 13164-1:2013 declares that method validation is essential for laboratory analysis (ISO 13164-1, 2013). There were international efforts to make primary radon standards using both the common  $^{226}\text{Ra}$  calibration approach and new radon-in-water standard (Felice, 2007; Forte et al., 2006; ISO 13164-1, 2013; Mertens et al., 2020; Mostafa et al., 2017). Furthermore, ISO 13164-1:2013 also emphasizes the importance of participating in proficiency tests or interlaboratory comparisons (Celaya González et al., 2018; Jobbágy et al., 2019a). In addition, this ISO standard also states that method validation should be carried out periodically through analysis of reference materials. However, the use of certified materials is not widespread in the literature.

In this work, a technique based on an ionization chamber for the measurement of  $^{222}\text{Rn}$  activity concentration in water has been developed, tested, verified and applied to tap and bottled drinking water samples.

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## 2. Materials and methods

### 2.1. *Sample Characteristics and Sampling Procedure*

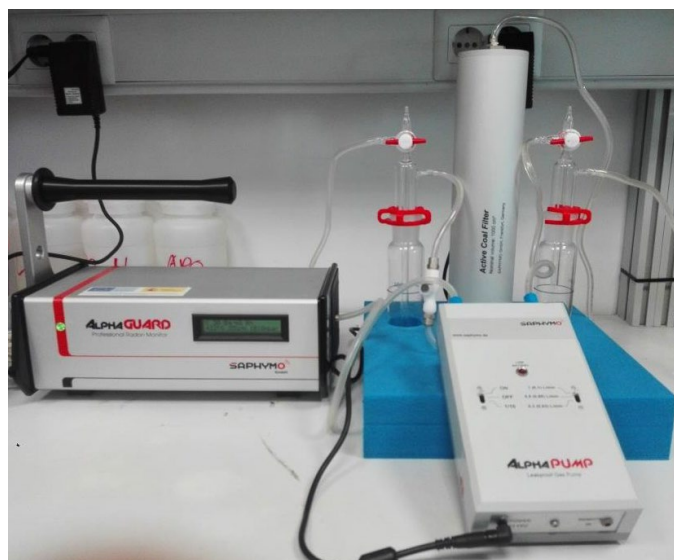
Twelve tap water samples were taken directly at taps in Sevilla city. Before taking the sample, water was let run for 15 min to obtain fresh water. The water was kept flowing for a while and filling the containers to the edge, then being immediately closed to avoid loss of radon.

Twenty-four bottled mineral water samples from two different brands (“Solan de Cabras” and “Font Natura“) were purchased from supermarkets in Sevilla. Samples of bottled mineral water were kept at room temperature and then opened in the laboratory for analysis.

### 2.2. *Equipment Setup*

A continuous active radon monitor ALPHAGUARD (Model PQ 2000 PRO) of Genitron-Saphymo, Germany was used to obtain radon concentration (Genitron Instruments, 1998). This measuring system was selected due to its calibration stability, fast reading, and accurate measurements which have been confirmed in previous studies (Alonso et al., 2015; Di Carlo et al., 2019; Kotrappa et al., 2005; Kurnaz and Atif Çetiner, 2016; Li et al., 2015; Seid et al., 2020). The detector comprises a pulse-counting ionization chamber of 0.65-l active volume with a measurement range of 2 to  $2 \times 10^6$  Bq m<sup>-3</sup>. Radon and its progeny in the air enter the active volume of the ionization chamber being detected. Additionally, the system also registers the values of temperature, humidity, and atmospheric pressure for the duration of the measurement.

Radon concentration in water was measured using an AquaKIT system (Genitron Instruments, 2008). The water sample was injected into a 500 mL container (degassing vessel). Then, a gas pump (AphaPUMP) expelled radon from the water sample by passing bubbles through it. Finally, a safety glass bottle was connected to the detector forming a closed loop, as shown in Figure 1. Additionally, an active carbon filter was plugged in the system before every measurement cycle in order to reduce background radon concentration.



**Fig. 1 - Practical setup in the laboratory.**

### *2.3. Quality control*

The calibration of AlphaGUARD was carried out using a NIST- $^{226}\text{Ra}$  standard (NIST 4973#17) with a concentration of  $1036 \pm 6$  Bq in a 1.5M HCl solution. Its  $^{222}\text{Rn}$  emanation fraction was  $0.98 \pm 0.03$  at  $21^\circ\text{C}$ . The polyethylene encapsulated  $^{226}\text{Ra}$  solution was packed into a leak tight glass bulb where  $^{222}\text{Rn}$  accumulates over time.

$^{222}\text{Rn}$  emanation standards, based on a liquid standard  $^{226}\text{Ra}$  source from CIEMAT (Spain) ( $35.68 \pm 0.24$  Bq  $\text{g}^{-1}$ ), have also been used for QC testing. The secondary standards were prepared by gravimetric dilution with deionized water of the  $^{226}\text{Ra}$  standard obtaining two 1 L solutions (STD#1:  $23.690 \pm 0.002$  Bq  $\text{m}^{-3}$ ; and STD#2:  $53.410 \pm 0.007$  Bq  $\text{m}^{-3}$ ). The secondary standards were stored in glass containers at least three weeks to ensure reestablishment of secular equilibrium between  $^{226}\text{Ra}$  with its progeny.

Method validation was assessed using IAEA proficiency test QC samples with known  $^{226}\text{Ra}$  activity (IAEA-TEL-2014-03 Sample 02:  $14.21 \pm 0.06$  Bq  $\text{kg}^{-1}$ ; and IAEA-TEL-2014-03 Sample 03:  $17.9 \pm 0.1$  Bq  $\text{kg}^{-1}$ ). Quality control samples were allowed to stand for 25 days to allow ingrowth of  $^{222}\text{Rn}$  in the containers.

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Multiple blank measurements were obtained and a detection limit of 5.9 Bq l<sup>-1</sup> was calculated (Currie, 1968), which is lower than 10 Bq l<sup>-1</sup>.

#### 2.4. Measurement method

First, 100 ml of the sample was injected into the degassing vessel using a syringe. After sample injection, the gas cycle was closed, and the air pump was switched on being radon extracted from water and transferred to the monitor all through the closed gas cycle. All drops generated in the degassing process would go into the gas cycle being deposited in the safety vessel. The background of the empty setup was measured for 40 minutes before each measurement was carried out. The flow rate of the pump was 0.3 l min<sup>-1</sup> for 10 minutes. Then, the pump was turned off and the activity concentration of <sup>222</sup>Rn was recorded every 10 minutes for the next 40 minutes. The activity concentration of <sup>222</sup>Rn in the water samples was calculated using the following equation:

$$C_{water} = \frac{C_{air} \cdot \left( \frac{V_{system} - V_{sample}}{V_{sample}} + k \right) - C_0}{1000} \quad (1)$$

where  $C_{water}$  is the <sup>222</sup>Rn concentration in the water sample (Bq m<sup>-3</sup>),  $C_{air}$  is the <sup>222</sup>Rn concentration in the setup after expelling <sup>222</sup>Rn from water (Bq m<sup>-3</sup>),  $C_0$  is empty setup background (Bq m<sup>-3</sup>),  $V_{system}$  is the interior volume of the measurement setup (ml),  $V_{sample}$  is the volume of the water sample (ml), and  $k$  is the <sup>222</sup>Rn distribution coefficient.

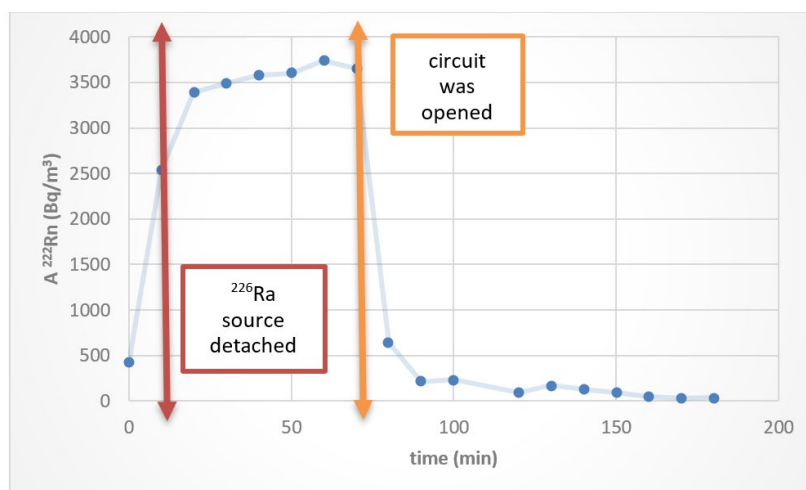
The radon distribution coefficient  $k$  varies inversely with the temperature and it was calculated by the following equation (Li et al., 2015):

$$k = 0.1057 + 0.405 \cdot e^{-0.0502 \cdot T} \quad (2)$$

### 3. Results and discussion

### 3.1. System optimization and validation

AquaKIT measurement equipment included several glass vessels granting a hermetically sealed enclosure of  $^{222}\text{Rn}$  emanated from water samples. However, leakages in the system can cause incorrect measurements. For this reason, the system was first checked for leaks using a NIST- $^{226}\text{Ra}$  standard source.



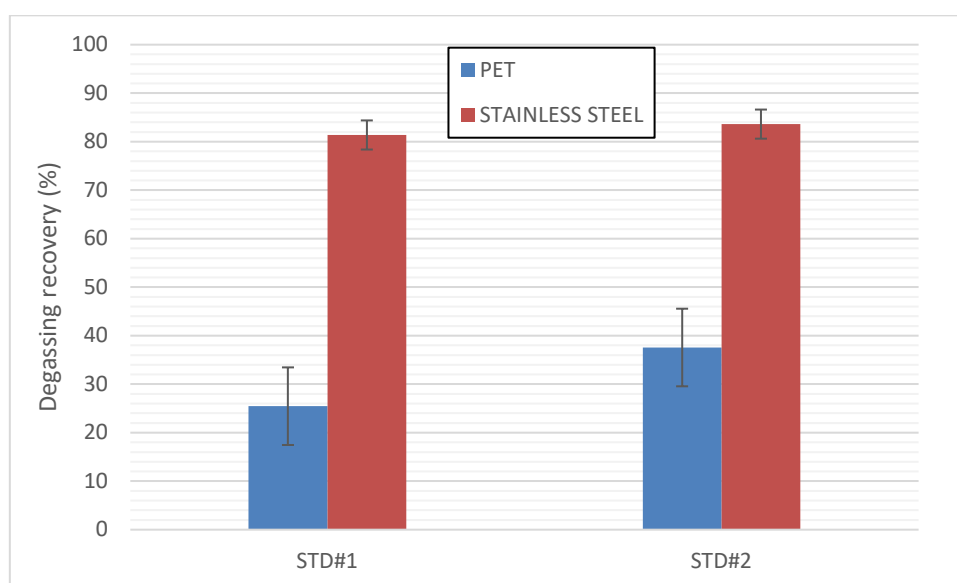
**Fig. 2 - Activity concentration of  $^{222}\text{Rn}$  for NIST standard source as a function of time.**

The system was first cleaned of any residual radon from previous measurements, so the activated carbon filter was switched on, and the pump was also switched on with a maximum flow rate of  $1 \text{ l min}^{-1}$  until a radon concentration of less than  $5 \text{ Bq m}^{-3}$  was reached. After that, NIST- $^{226}\text{Ra}$  standard source was attached to the system, and AlphaGUARD and AlphaPUMP were switched on. After 10 min the NIST- $^{226}\text{Ra}$  standard source was detached, pump was switched off, and AlphaGUARD remained measuring for another 60 min (see Fig.2). Finally, system was opened to clean it. It can be concluded that system was kept airtight during 60 min of measurement.

On the other hand, the degassing process was also checked. 100 ml of  $^{222}\text{Rn}$  secondary standards (STD#1 and STD#2) were directly injected into the degassing vessel. The samples were measured following the method described above. The degassing efficiency was calculated by comparing the

measured and actual activity concentration of secondary standard samples (STD#1 and STD#2), obtaining a high value (>97.0 %).

Then, the material of containers for water samples measurement was tested. Previous studies suggested that PET is much suitable for storing water for the assay of radon. To check this fact, two  $^{222}\text{Rn}$  secondary standards (STD#1 and STD#2) were collected in 0.5 l PET coke bottles. The secondary standards were stored in those PET containers at least three weeks to ensure reestablishment of secular equilibrium between  $^{226}\text{Ra}$  with its progeny. Next, samples were measured following the method previously described. As shown in Fig. 3,  $^{222}\text{Rn}$  degassing recovery was too low for both samples. Because degassing efficiency was found to be optimal, low recovery must be due to container. The main factor affecting radon loss rate of a given material is a combined effects of both the radon adsorption and diffusion processes during sample storage (Fernández et al., 2004; Lucchetti et al., 2016). Not only container material but its cap also has to be radon tight (Jobbágy et al., 2019b).



**Fig. 3 – Degassing recovery (%) of  $^{222}\text{Rn}$  for secondary standard sources (STD#1 and STD2) in PET and stainless-steel containers.**

Radon chambers were usually made of stainless steel (De Simone et al., 2015; Mostafa et al., 2017). For that reason, 0.4 l stainless steel bottles were purchased for this test, as the alloy contains aluminium

with a very small diffusion length. The results showed that a significant improvement in radon recovery was achieved for standard samples (see Fig. 3).

Finally, IUPAC (Thompson et al., 2006) and ISO (Tholen, 2008) recommendations for assessment of performance of laboratories were used including z-score test, u-test, and trueness and precision tests in two IAEA standard materials. Samples were analyzed by using the proposed method and results are shown in Table 1. The z-score and u values obtained are inside an acceptable range for trueness of the proposed method. All the results obtained an “Acceptable” status referring to precision and trueness. The analysis of IAEA-TEL-2014-03 Sample 02 provided a  $^{226}\text{Ra}$  activity of  $13.53 \text{ Bq kg}^{-1}$ , very close to the reference material reported value and within its 95% confidence interval. Referring to IAEA-TEL-2014-03 Sample 03, the value obtained with the proposed method was  $18.4 \text{ Bq kg}^{-1}$ , which was within its 95% confidence interval.

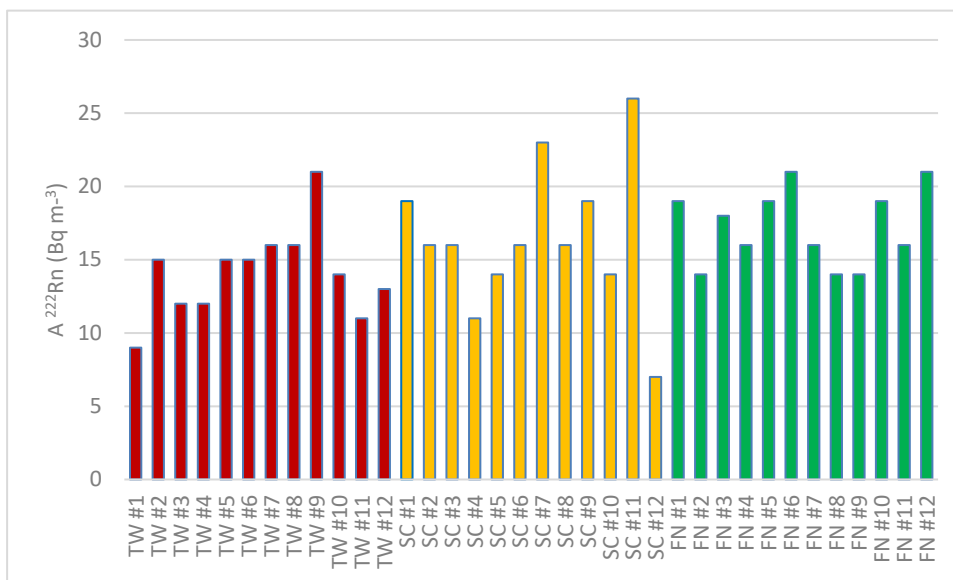
**Table 1 - Results obtained for two IAEA reference materials.**

Sample	$^{226}\text{Ra}_{\text{IAEA}}$ ( $\text{Bq kg}^{-1}$ )	$u(\text{A})_{\text{IAEA}}$ ( $\text{Bq kg}^{-1}$ )	$^{226}\text{Ra}_{\text{measured}}$ ( $\text{Bq kg}^{-1}$ )	$u(\text{A})_{\text{measured}}$ ( $\text{Bq kg}^{-1}$ )	z-score	u	Trueness	Precision P (%)
IAEA-TEL-2014-03 Sample 02	14.21	0.06	13.53	0.74	0.91	0.92	A	5.5
IAEA-TEL-2014-03 Sample 03	17.9	0.1	18.34	0.89	0.49	0.50	A	4.9

### 3.2. Radon measurement in tap and mineral water samples



The activity concentrations of  $^{222}\text{Rn}$  measured in tap (TW) and bottled natural mineral water (SC and FN) samples studied are given in Fig. 4.



**Fig. 4 - Radon concentration (Bq m<sup>-3</sup>) measured in tap (TW) and bottled mineral waters (SC and FN).**

The activity concentrations of  $^{222}\text{Rn}$  varied for tap water samples from 9 to 21 Bq m<sup>-3</sup>. The average of  $^{222}\text{Rn}$  was found as 14.1 Bq m<sup>-3</sup>. The concentrations of  $^{222}\text{Rn}$  in bottled mineral samples varied from 7 to 27 Bq m<sup>-3</sup> with an average of 16.8 Bq m<sup>-3</sup>. It was clear that all water sample collected show values below 100 Bq l<sup>-1</sup>, the recommended reference value of EU legislation, and below 500 Bq l<sup>-1</sup> from Spanish legislation. Also, the average  $^{222}\text{Rn}$  activity concentration was lower than those reported from different countries (Algeria - 7.00 Bq l<sup>-1</sup>; Egypt - 2.00 Bq l<sup>-1</sup>; Spain – 1.20 Bq l<sup>-1</sup>; China – 0.09 Bq l<sup>-1</sup>; Turkey – 0.04 Bq l<sup>-1</sup>) (Amrani, 2002; Dueñas et al., 1999; Seid et al., 2020; Yong et al., 2020; Yousef, 2018).

### 3.3. Annual effective dose

Internal exposure due to radon in tap and bottled mineral water samples may be caused by ingestion (drinking water containing radon) or by inhalation (breathing radon gas in indoor air exhaled from water). The associated annual effective dose was estimated to evaluate radiological risk for infants (1–

2 y), children (7–12 y), and adults (> 17 y) through the ingestion annual effective dose  $E_{ING}$  (mSv  $y^{-1}$ ) (UNSCEAR, 2000):

$$E_{ING} \left( \frac{mSv}{y} \right) = C \times DC_{ING} \times IW \quad (3)$$

where  $C$  is the concentration of  $^{222}\text{Rn}$  measured in the mineral water samples ( $\text{Bq l}^{-1}$ ),  $DC_{ING}$  is the dose conversion factor for ingestion and  $IW$  is an annual intake of water ( $\text{l y}^{-1}$ ), suggested by UNSCEAR for infants, children, and adults are  $150 \text{ l y}^{-1}$ ,  $350 \text{ l y}^{-1}$ ,  $500 \text{ l y}^{-1}$  and respectively. UNSCEAR suggested the value of  $DC_{ING}$  for infants, children, and adults as  $23 \times 10^{-6} \text{ mSv Bq}^{-1}$ ,  $5.9 \times 10^{-6} \text{ mSv Bq}^{-1}$  and  $3.5 \times 10^{-6} \text{ mSv Bq}^{-1}$ , respectively.

**Table 2 - Annual effective radiation doses for ingestion due to the consumption of tap (TW) and mineral water samples (SC and FN).**

Sample	$E_{adults}$ (mSv $y^{-1}$ )	$E_{children}$ (mSv $y^{-1}$ )	$E_{infants}$ (mSv $y^{-1}$ )
TW #1	$1.6 \cdot 10^{-5}$	$1.9 \cdot 10^{-5}$	$3.1 \cdot 10^{-5}$
TW #2	$2.6 \cdot 10^{-5}$	$3.1 \cdot 10^{-5}$	$5.2 \cdot 10^{-5}$
TW #3	$2.1 \cdot 10^{-5}$	$2.5 \cdot 10^{-5}$	$4.1 \cdot 10^{-5}$
TW #4	$2.1 \cdot 10^{-5}$	$2.5 \cdot 10^{-5}$	$4.1 \cdot 10^{-5}$
TW #5	$2.6 \cdot 10^{-5}$	$3.1 \cdot 10^{-5}$	$5.2 \cdot 10^{-5}$
TW #6	$2.6 \cdot 10^{-5}$	$3.1 \cdot 10^{-5}$	$5.2 \cdot 10^{-5}$
TW #7	$2.8 \cdot 10^{-5}$	$3.3 \cdot 10^{-5}$	$5.5 \cdot 10^{-5}$
TW #8	$2.8 \cdot 10^{-5}$	$3.3 \cdot 10^{-5}$	$5.5 \cdot 10^{-5}$
TW #9	$3.7 \cdot 10^{-5}$	$4.3 \cdot 10^{-5}$	$7.2 \cdot 10^{-5}$
TW #10	$2.5 \cdot 10^{-5}$	$2.9 \cdot 10^{-5}$	$4.8 \cdot 10^{-5}$
TW #11	$1.9 \cdot 10^{-5}$	$2.3 \cdot 10^{-5}$	$3.8 \cdot 10^{-5}$
TW #12	$2.3 \cdot 10^{-5}$	$2.7 \cdot 10^{-5}$	$4.5 \cdot 10^{-5}$
SC #1	$3.3 \cdot 10^{-5}$	$3.9 \cdot 10^{-5}$	$6.6 \cdot 10^{-5}$
SC #2	$2.8 \cdot 10^{-5}$	$3.3 \cdot 10^{-5}$	$5.5 \cdot 10^{-5}$
SC #3	$2.8 \cdot 10^{-5}$	$3.3 \cdot 10^{-5}$	$5.5 \cdot 10^{-5}$
SC #4	$1.9 \cdot 10^{-5}$	$2.3 \cdot 10^{-5}$	$3.8 \cdot 10^{-5}$

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SC #5	$2.5 \cdot 10^{-5}$	$2.9 \cdot 10^{-5}$	$4.8 \cdot 10^{-5}$
SC #6	$2.8 \cdot 10^{-5}$	$3.3 \cdot 10^{-5}$	$5.5 \cdot 10^{-5}$
SC #7	$4.0 \cdot 10^{-5}$	$4.7 \cdot 10^{-5}$	$7.9 \cdot 10^{-5}$
SC #8	$2.8 \cdot 10^{-5}$	$3.3 \cdot 10^{-5}$	$5.5 \cdot 10^{-5}$
SC #9	$3.3 \cdot 10^{-5}$	$3.9 \cdot 10^{-5}$	$6.6 \cdot 10^{-5}$
SC #10	$2.5 \cdot 10^{-5}$	$2.9 \cdot 10^{-5}$	$4.8 \cdot 10^{-5}$
SC #11	$4.6 \cdot 10^{-5}$	$5.4 \cdot 10^{-5}$	$9.0 \cdot 10^{-5}$
SC #12	$1.2 \cdot 10^{-5}$	$1.4 \cdot 10^{-5}$	$2.4 \cdot 10^{-5}$
FN #1	$3.3 \cdot 10^{-5}$	$3.9 \cdot 10^{-5}$	$6.6 \cdot 10^{-5}$
FN #2	$2.5 \cdot 10^{-5}$	$2.9 \cdot 10^{-5}$	$4.8 \cdot 10^{-5}$
FN #3	$3.2 \cdot 10^{-5}$	$3.7 \cdot 10^{-5}$	$6.2 \cdot 10^{-5}$
FN #4	$2.8 \cdot 10^{-5}$	$3.3 \cdot 10^{-5}$	$5.5 \cdot 10^{-5}$
FN #5	$3.3 \cdot 10^{-5}$	$3.9 \cdot 10^{-5}$	$6.6 \cdot 10^{-5}$
FN #6	$3.7 \cdot 10^{-5}$	$4.3 \cdot 10^{-5}$	$7.2 \cdot 10^{-5}$
FN #7	$2.8 \cdot 10^{-5}$	$3.3 \cdot 10^{-5}$	$5.5 \cdot 10^{-5}$
FN #8	$2.5 \cdot 10^{-5}$	$2.9 \cdot 10^{-5}$	$4.8 \cdot 10^{-5}$
FN #9	$2.5 \cdot 10^{-5}$	$2.9 \cdot 10^{-5}$	$4.8 \cdot 10^{-5}$
FN #10	$3.3 \cdot 10^{-5}$	$3.9 \cdot 10^{-5}$	$6.6 \cdot 10^{-5}$
FN #11	$2.8 \cdot 10^{-5}$	$3.3 \cdot 10^{-5}$	$5.5 \cdot 10^{-5}$

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The annual effective doses for all samples are presented in Table 2. Results show that annual effective dose due to ingestion were clearly below 0.002 mSv, the recommended contribution of ingestion of radon in water for the total mean annual dose (UNSCEAR, 2000). The average of the results on this study was  $0.028 \mu\text{Sv y}^{-1}$ , for adults, and  $0.033 \mu\text{Sv y}^{-1}$ , for children, clearly above the reference values (Council Directive 59/EURATOM, 2013).

#### 4. Conclusions

The calibration and optimization of an ALPHAGUARD+AQUAKIT system performed in this study produced very good results. This was verified by the results of the measurement of IAEA

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intercomparison samples for  $^{222}\text{Rn}$  in water. The use of stainless steel containers as virtually shatterproof bottles for testing radon in water offers an alternative to the use of glass or plastic bottles. From the point of view of radiological protection, the radon levels measured in tap and bottled mineral water in the present work are lower than the minimum concentration level established by the European Commission (100 Bq l<sup>-1</sup>).

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