Development and operational performance of a single calibration chamber for radon detectors

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Abstract

This work shows the design, setup and performance of a new single radon detector calibration chamber developed at the University of Huelva (Environmental Radioactivity Group). This system is based on a certified radon source and a traceable reference radon detector, which allows radon concentrations inside the chamber radon to be obtained in steady state conditions within a range of 400-22 000 Bq m^{-3} with associated uncertainties in the range of 4%.

In addition, the development of a new *ad hoc* calibration protocol (UHU-RC/01/06 "Rachel"), which is based on the modelling of radon concentration within the chamber, allows it to be used without the reference detector. To do that, a complete characterization and calibration of the different leakage constants and the flow meter reading have been performed. The accuracy and general performance of both working methods for the same chamber (i.e., with and without the reference detector) have been tested by means of their participation in an intercomparison exercise involving five active radon monitors.

Keywords: Radon chamber model, calibration protocol, leakages, active detector inter-comparison, traceability, flow-through radon source.

1. Introduction

Radon and its short-lived decay products in the atmosphere are the most important contributors to human exposure to ionizing radiation from natural sources. This contribution is around 1.1 mSv y^{-1}, which represents 50% of total dose, [1]. However, the determination of radon concentration in the air requires the use of special detectors due to its noble gas nature. These detectors require in turn specific standard and reference materials to assure the quality of the analytical results. Different kinds of active and passive detectors have been developed for radon and progeny measurement and their performance is well documented in literature ([2], [3] and [4]). In order to calibrate these detectors, many radon source reference materials have been proposed ([5], [6] and [7]), coupled with different calibration methods. Nevertheless, the most standard method for radon detector calibration is the exposure to a steady state radon concentration in a sealed chamber under controlled conditions, using a reference radon detector, which must be traceable to a first standard.

Many radon chambers have been constructed in order to perform radon detector calibration according to different criteria: radon concentration range control, volume, material, radon source, reference radon detector, humidity and temperature control, aerosol concentration, etc. The proposed use has been not only to perform research into radon and its decay product behaviour but also to carry out the routine detector calibration for field use [8]. All these chambers offer as a common factor the use of a reference detector in order to accurately know the true radon concentration inside the chamber, because the presence of unconsidered leakages introduces unpredictable deviations of experimental performances from theoretical results.

One of the main aims of this work is to obtain an equation to describe the temporal evolution of the radon activity concentration within a calibration chamber (analytically considering the effect of leakages) as a function of several system characteristic parameters, according to the operation mode. This way, it is possible to get an adequate design of the chamber in accordance with our necessities and a useful tool to control the radon concentration inside the calibration chamber. This model could be used either to automate the radon concentration control system or to operate the radon chamber without a radon reference detector.

In order to calibrate this model and characterize the radon chamber and all implicated systems a new procedure, the protocol UHU-RC/01/06 "Rachel" has been developed, allowing a theoretical reference concentration traceable to a first standard to be obtained.

2. Radon concentration model.

The analytical model describing the radon concentration inside the chamber is based on a balance equation. The different source and sink terms involved in this balance are: the amount of radon dispensed by the radon generation system, E_0 (Bq/s) the radioactive decay, λ_{Rn} (s^{-1}) the vent flow, q_1 (m^3/s) and the leakages of the different systems, λ_v. Therefore, following [9]:

$$\frac{dC_{Rn}(t)}{dt} = \frac{E_0 + q_1 C_{Rn}^{air}}{V} - \lambda_{Rn} C_{Rn}(t) - \lambda_v C_{Rn}(t) - \frac{q_1}{V} C_{Rn}(t)$$
$$i.c.: C_{Rn}(t=0) = C_{Rn}^0 \qquad (1)$$

where C_{Rn}^{air} is the background radon concentration at the laboratory. The difference of equation (1) against the referred model is the consideration of the presence of leakages inside the chamber. This phenomenon has been modelled by means of the leakages constant, λ_v, which represents the escape probability of a radon atom, per time unit, through possible fissures, pores and loose connections [10]. This parameter represents the contribution of all implicated systems, which includes the container, λ_v^c, and the radon generation system, λ_v^{FB}, leakages.

The selected configuration for the radon generation system is based on a feedback circuit with a flow-through radon source (Fig. 1). This source is based on a dry powder with an emanation factor and ^{226}Ra activity certified. Thus, it is possible to see that the source term introduced into the chamber is not dependent on the applied flow in the feedback circuit; it is only dependent on the ^{226}Ra activity, A_{Ra}. So, it is possible to write this source term as: $E_0 = \lambda_{Rn} A_{Ra.}$

The vent flow plays a very important role in the radon concentration inside the chamber as it controls the rate of air renewal into the chamber. It is important to take into account that both the radon diffusion in the air and the spatial situation of the different source and sink terms have not been considered in this study. It is possible to do that once an air homogenization system inside the chamber is provided.

Under these conditions, it is possible to solve the equation (1) in an analytical way and to obtain a temporal exponential dependence, (2), modulated by the effective constant of the chamber, λ_{eff}, which includes the dependence with the radon decay, the leakages and the vent flow, (3):

$$C_{Rn}(t) = C_{Rn}^{sat} - (C_{Rn}^{sat} - C_{Rn}^{0})\, e^{-\lambda_{eff} t} \qquad (2)$$

where

$$\lambda_{eff} = \lambda_{Rn} + \lambda_{v} + \frac{q_1}{V} \qquad (3)$$

$$C_{Rn}^{sat} = \frac{E_0 + q_1 C_{Rn}^{air}}{\lambda_{eff} V} \qquad (4)$$

C_{Rn}^{Sat}, the saturation concentration (Bq/m^3), is the steady state radon concentration for an applied vent flow. This one will be the working concentration for detector calibrations and the inter-comparison described below.

The dependence obtained for the saturation concentration, (4), implies that, working with the same source, a large volume chamber will present a lower concentration than a small chamber. The presence of leakages also provides a drop in the working concentration inside the chamber.

3. Systems description

In figure 2, a scheme of the implicated systems in the calibration chamber operation is shown. Following the labelling of each system, they are all detailed in this part with the principal characteristics described.

3.1 Radon generation system

This element is used to feed the radon inside the chamber. It is based on a reference certified material, Rn-1025-20 (Pylon electronics, Canada). It consists of a dry source and has a ^{226}Ra activity of 23.7 kBq (± 4%) and an emanation factor of 100% for flow through within a range of 0 - 10 L/min. In the selected configuration, feedback, the radon activity provided per time unit is E_0 = 49.7 mBq/s (± 4%). Finally, the radon-air flow across the source is pressure-driven by a small pump with a nominal flow of 7 L/min. The background radon concentration at lab was monitored during six months; the average measured concentration (19 ± 7 Bq/m^3) was used as C_{Rn}^{air} input for the model.

3.2 The chamber

The laboratory-made radon chamber container consisted of a 0.220 m^3 barrel made of high-density polyethylene (HDPE). This material has a diffusion coefficient of $1.8 \cdot 10^{-12}$ m^2 s^{-1}, which can assure a good impermeability level for thickness larger than 1.5 mm [11]. Several detectors can be simultaneously introduced within the chamber due to its large volume, thus allowing the performance of calibrations and inter-comparisons under exactly the same operational conditions.

The maximum saturation concentration is reached when the vent flow and the leakages are zero, equations (3) and (4). Under these ideal conditions, that concentration would be in the range of 100 kBq m^{-3}. The presence of leakages will diminish this value drastically, as shown in part 5. A small ventilator homogenizes the air distribution inside the chamber, also assuring an appropriate mixing of gases.

3.3 The concentration control system

The purpose of this system is to renew the air inside the chamber with low (background) radon contents until a selected value of radon concentration is reached. The control parameter is the pressure-driven vent flow, which works within a range of 0 – 7 L min^{-1}. The flow is monitored using a controller flow meter (GFC17A-BADL2, Aalborg, USA).

The minimum saturation concentration is reached when the vent flow reaches its maximum. Thus, this concentration value is about 400 Bqm^{-3} and the influence of leakages will be small.

3.4 The reference radon monitor

A commercial monitor AlphaGuard PQ2000PRO (Genitron Instruments, Germany), working as an ionization chamber, has been selected for this purpose. This active system fits the instrumental requirements based on 1) good temporal resolution, 2) scarce influence of both thoron concentration and weather conditions, which can be monitored 3) factory calibration traceable to PTB standards and 4) a large dynamic range (2 - $2 \cdot 10^6$ Bq m^{-3}).

As detailed in the following section, the calibration protocol requires a strong traceability of the reference monitor. The unit used in this work was factory-calibrated against a PTB standard and the performances were tested against different reference materials such as a dry source having certified both activity and emanation factor (NIST RM 4968/CP-100) and a certified activity ^{222}Rn bulb from NPL (X2/94). Finally, this device has been validated through its participation in the ICE'94 International inter-

175 calibration experiment [12]. All the deviations from the respective reference values
176 were less than 3%.
177
178 **4. Calibration procedure: the UHU-RC/01/06 "Rachel" protocol**
179 The calibration chamber could operate independently of the reference monitor, provided
180 that the proposed model is correctly calibrated. To do that, a specific calibration
181 protocol (UHU-RC/01/06 "RA-don CH-amber mod-EL") has been developed. This
182 procedure considers five fundamental steps: a) Reference radon detector traceability
183 from a primary standard, as detailed in the previous section, b) controlled flow meter
184 calibration, c) container leakage calibration, d) determination of the radon generation
185 system leakages, and e) model validation. The last four points are detailed in the
186 following section.
187
188 *4.1. Controlled flow meter calibration*
189 The calibration of the flow meter reading deviation has been carried out via the
190 determination of the dimensionless "flow correction factor" *FCF*, defined as:

$$FCF = \frac{q_{reference}}{q_{GFC}} \qquad (5)$$

191
192 being $q_{reference}$ the true flow (measured by means of a volume totalizer with a low limit
193 of 0.7 L/min) and q_{GFC} the flow meter reading. Nine different flows were tested in order
194 to evaluate the *FCF* within a range of 1 up to 7 L min^{-1}. These experimental results were
195 used to fit the *FCF* as a function of the flow meter reading, q_{GFC} according to the
196 following experimental equation:

$$FCF = a - b \cdot \sqrt{q_{GFC}} \qquad (6)$$

197

198 The obtained fit is shown in Figure 3. It is easy to see that due to the low precision
199 associated with the low flow rate readings (i.e., the uncertainty associated with q_{GFC}),
200 flow rates less than 1 L min^{-1} should be avoided during the calibration procedure.

201

202 *4.2. Container leakages*

203 To determine the magnitude of the container leakage constant, characterized by the
204 constant λ_v^c, an initial radon concentration of 19 kBq m^{-3} was introduced into the
205 chamber. Its temporary variation was monitored for at least two weeks in order to fit the
206 measured concentration as a function of time, as shown in Figure 4. It is clear that,
207 under these conditions, the situation predicted by equation (1) should be achieved after
208 removing the terms corresponding to vent flow and source term. The reproducibility of
209 the calculus has been assured by replicating this step 5-10 times. Thus a value of λ_v^c =
210 $(2.8 \pm 0.3)10^{-7}$ s^{-1} was obtained.

211

212 *4.3. Radon generation system leakages*

213 They are characterized by the parameter λ_v^{FB}. The applied procedure was similar to that
214 previously shown in section 4.2; nevertheless, the radon generation system was working
215 all time. The chamber remained sealed for 7 days, until steady state conditions were
216 reached. In this case the equation (1) should appear with a source term. The initial
217 concentration under these conditions (for five different replicates) was about 100
218 kBq/m^3, which is also assumed to be the maximum theoretical concentration when no
219 leakages are present.

The radon generation system leakage constant was evaluated in this way as λ_v^{FB} = (7.75 ± 0.06)10^{-6} s^{-1}. It is important to see that this value is one order of magnitude higher than the container leakage constant, and it is the same order of magnitude as the radon decay constant. As shown in figure 5, this fact represents a limitation on the maximum available concentration, which should be about 22 kBq/m^3.

4.4. Model validation

The validation of the calibration model was performed by using six different exposures to constant radon concentrations (24 h each) covering a range of 0.4 to 10 kBq/m^3. A calibration factor was obtained, which is defined as:

$$CF = \frac{C_{reference}}{C_{\mathrm{mod}\,el}} \qquad (7)$$

where $C_{reference}$ is the reference concentration determined by using the certified reference monitor. Figure 6 shows the experimental results for *CF* as a function of the used flow rates. In all cases, the *CF* value is close to one, and all the deviations from the reference value were less than 5%.

The associated uncertainty relative to the predicted radon concentration is about 8%; nevertheless, when flow rates are less than 0.5 L/min, it can rise to 12%. By comparing this uncertainty with the true deviation value, 5%, it is possible to see that the uncertainty concerning the model has been overestimated.

Some concern on the usefulness of the calibration procedure could arise as indoor radon natural concentrations are usually below the concentration range here used. However, this way of working (which is quite common in the calibrations of both radon and external dose rate meters) is possible based on the usually high range of linearity of radon monitors and detectors.

5. Active detector inter-comparison

This experiment was carried out to test the feasibility of this chamber to perform detector calibrations, using the AlphaGUARD as reference radon monitor, and including the model predictions as results to compare.

In this test, the performances of five active radon monitors from three Spanish laboratories were compared. All of them (excepting the ALPHAGUARD) are based on ^{218}Po electrodeposition. These detectors use two different sampling methods: radon diffusion and flow-through pressure-driven. Details of the used detectors are provided in Table 1.

Two different six-hour constant concentration exposures under normal operation conditions were used: 438 ± 63 and 669 ± 98 Bq/m^3. These concentrations were fixed by selecting the adequate vent flow, 6.7 ± 0.15 and 4.0 ± 0.15 L/min respectively.

The results are summarized in Figure 7, which shows the good stability of all the detectors used in this test. For three detectors, the deviation from the reference value was more than 5 % for both exposures, possibly as a consequence of a longer elapsed period since their last calibration (as compared to that of the reference detector).

The detector number 4 has shown the best behaviour from the entire detector set with a deviation from the reference value less than 3% under both exposure conditions. However, the radon chamber model, calibrated by means of UHU-RC/01/06 “Rachel” Protocol, has shown close agreement with the reference, showing a deviation of less than 3%.

6. Conclusions

The design and development of a robust radon detector calibration chamber has been shown. The calibration of a theoretical model for the prediction of radon concentration within the chamber allows the use of the calibration setup both with and without the reference detector. It has been shown that the greater effects on the accuracy of results are related to the lack of precision of the reading in the air flow of the concentration control system. On the contrary, explicitly introducing the air leakages from the chamber into the model allows us to get a close agreement between predicted and measured results.

The use of the calibration chamber in an inter-comparison exercise for active detectors allowed the location of minor disagreements (i.e., greater deviations from reference value) between monitored detectors, possibly as a consequence of the longer length of time since their last calibration for some of them. On the contrary, very close agreement was found between the predicted (model) and the monitored (reference monitor) concentrations, as expected.

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8. References

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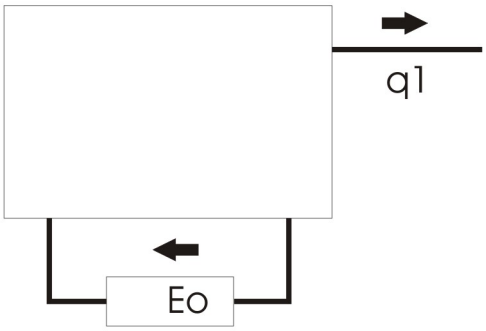
Table 1. Code, commercial denomination and manufacturer of the detectors employed in the inter-comparison exercise.

Code	Model	Manufacturer
1	RTM 2100	Sarad
2	RTM 2100	Sarad
3	Doseman	Sarad
4	1027	SNC
5	Scout	Sarad
6	Radon Chamber Model	Huelva University
Reference	AG PQ2000 PRO	Genitron

363 **Figure 1**



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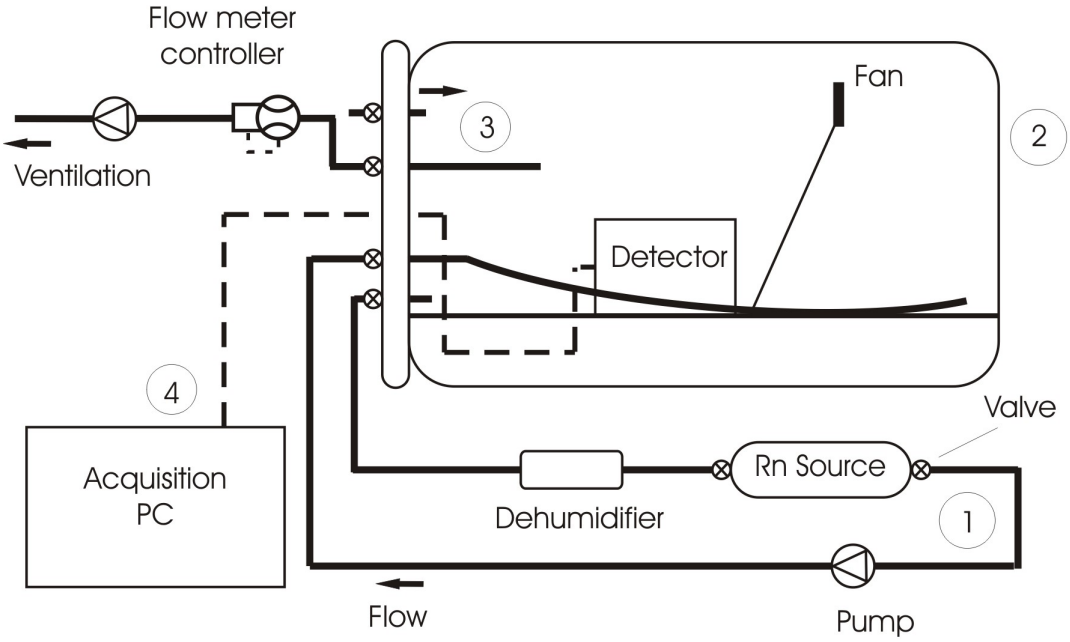
366 **Figure 2**



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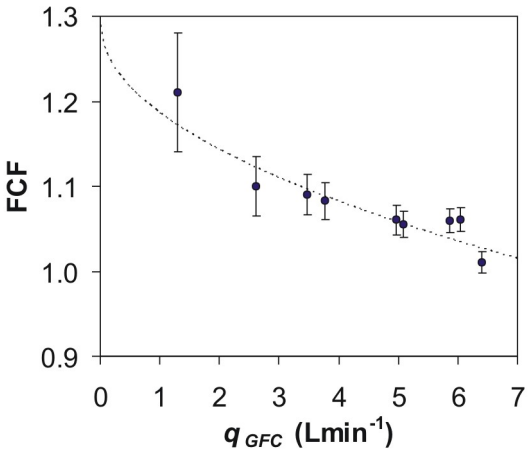
369 **Figure 3**



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373 **Figure 4**

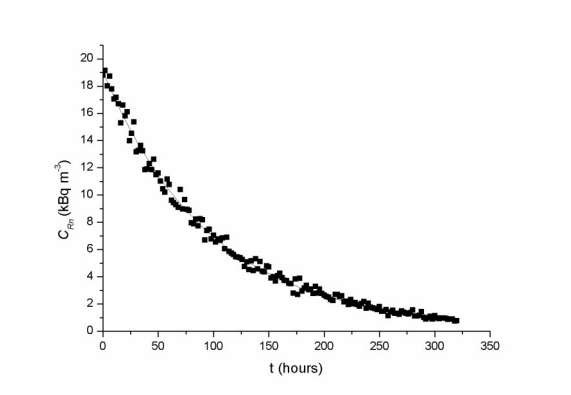

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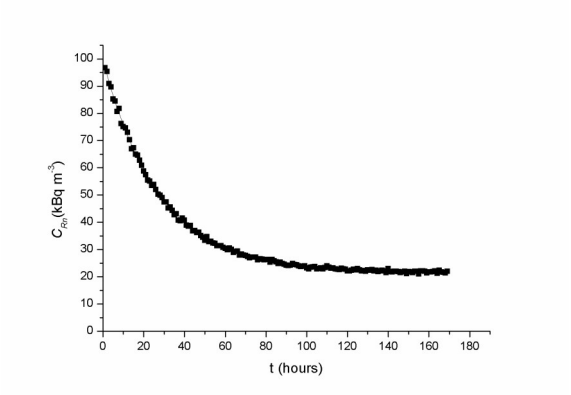
Figure 5


C_{Rn} (kBq m^{-3})
t (hours)
0
10
20
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50
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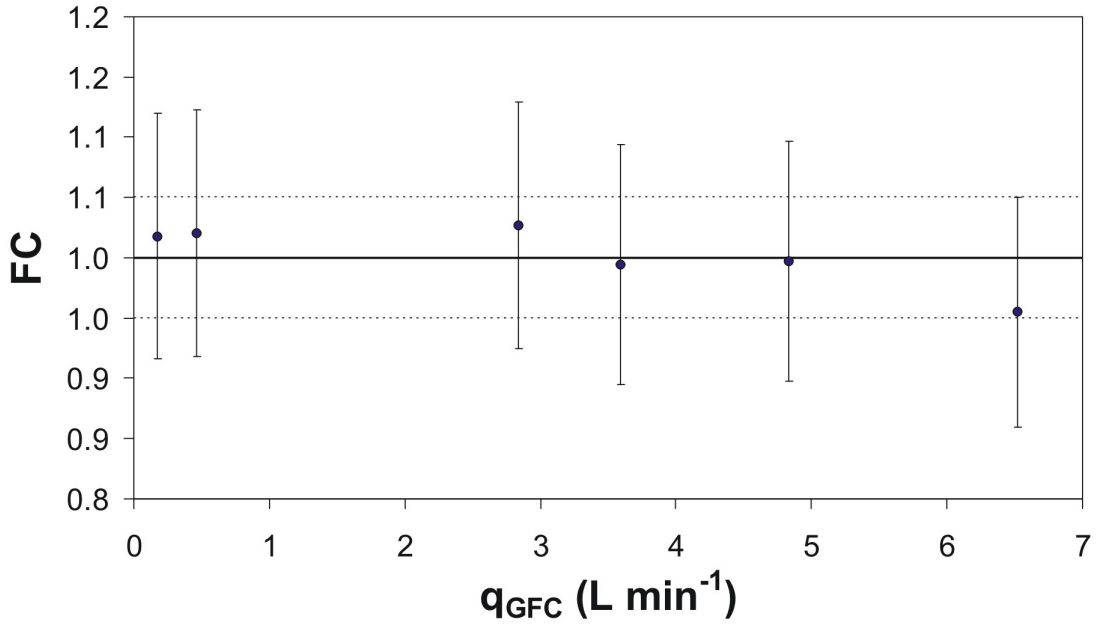
381 **Figure 6**



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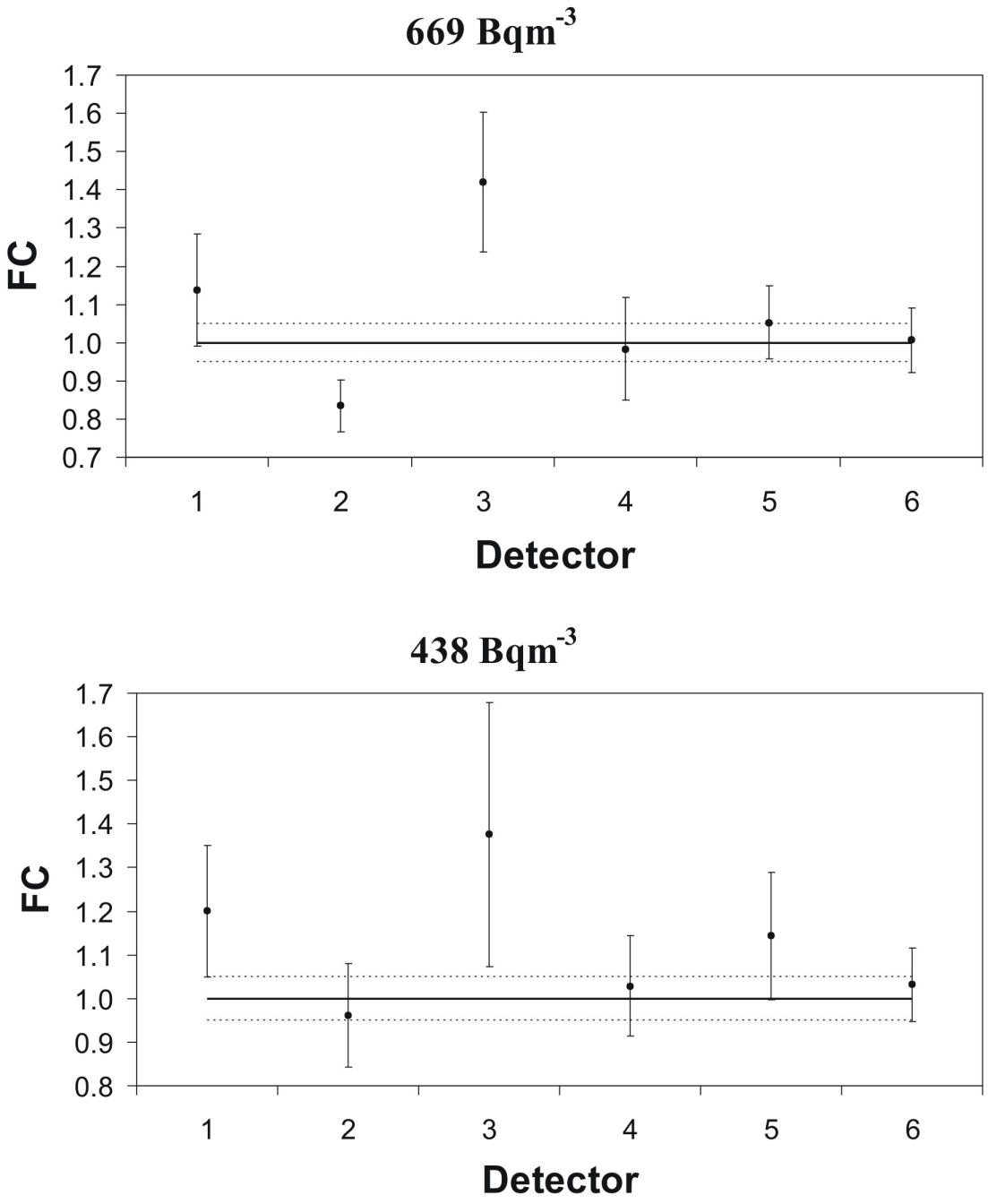
385 **Figure 7** (Opción A: Tamaño 160 mm)



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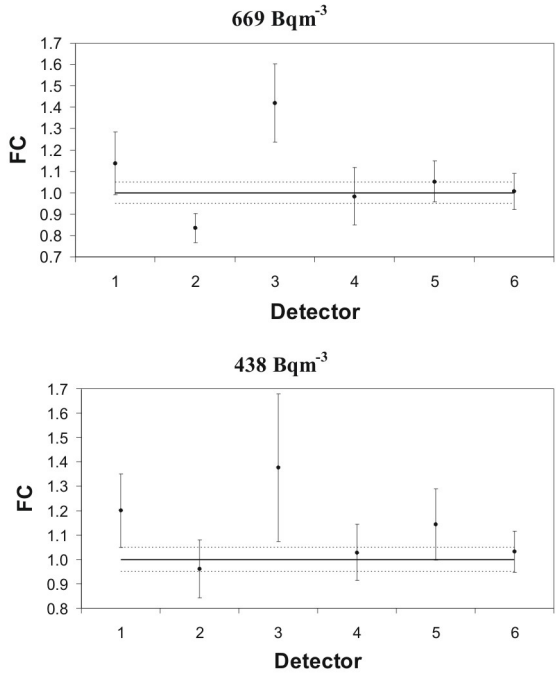
390 **Figure 7** (Opción B: Tamaño 75 mm)



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