Application of the Closed Compartment Model (CCM) to a radon source characterization

A. Noverques*, M. Sancho, B. Juste, G. Verdú.
Institute for Industrial, Radiophysical and Environmental Safety (ISIRYM), Universitat Politècnica de València. Camino de Vera s/n 46022 Valencia (Spain)
*ainome@iqn.upv.es

Abstract. This research provides a general overview of the radon behaviour in air. The main objective is to analyse the radon emission from three uranium sources with different activities (1.84 kBq, 4.27 kBq and 45 kBq) in two different air enclosed volumes (30 L and 120 L). As a result of uranium decay, its progeny, radon, is released into the air, where is measured continuously by a Radon Scout Plus. Deposits used in experimental tests are made of high-density polyethylene, hermetically sealed which allow radon concentration inside. Obtained results fit an exponential equation (Closed Compartment Model) with an R^2 greater than 0.99. This mathematical model allows the prediction of radon levels in air as a time function. Moreover it also provides the exposure rate of all three sources in the different air volumes tested.

KEYWORDS: Radon, air, Closed Compartment Model, Radon Scout Plus

1 INTRODUCTION

Considered a carcinogen by the World Health Organization (OMS) [1], radon, a descendant of uranium and radium, is naturally present in soil, water and building materials. This gas is the most important constituent of the natural radiation, for more than 50% that a person receives per year as well as the leading cause of lung cancer, behind smoking. Its control and subsequent mitigation is essential for the health of individuals.

According to the high concern regarding the protection from radon, European legislation has been updated in order to reduce radon concentration exposure as much as possible. In 2013, the Directive 2013/59/EURATOM [2] was published, establishing a new legislation limit for air radon concentration set at 300 $Bq \cdot m^{-3}$ in dwellings, public buildings and workplaces.

There are some studies that estimate air radon concentration evolution according to different mathematical models by considering the diffusion from soil and building materials and air exchange simultaneously [3], by a dynamic model of Radon Generation, Entry and Accumulation indoor, describing different sources [4, 5] or estimating indoor concentration based on the radon flux in soil and groundwater [6]. Additionally, other models as the Closed Compartment Model [7] explains radon concentration increase by time variations in a closed location, isolated from the environment. This mathematical model, could predict radon behavior in air according to its radon source activity.

From the different mathematical models available for the prediction of radon in air, in this research and, according to previous investigations [8], the Closed Compartment Model (CCM) has been selected in order to characterize radon sources and anticipate its concentration in air.

Therefore, the main objective of this research is the analysis of the radon behavior of radon emanating from three different sources in two tanks of volumes 30 L and 120 L. It is proved that the results obtained of air radon concentration can be adjusted to the Closed Compartment Model in order to predict the levels of radon in air as a function of time and as well as determining the maximum reached radon concentration in each deposit and the exposure rate of each source.

2 METHODS AND MATERIALS

2.1 Experimental procedure

In this research, an analysis of different sources of radon in different air volumes of air has been carried out. Subsequently, the effect of air volume on the radon exposure rate is also studied. Below, the used sources are detailed as well as the deposits in which the airborne radon measurements have been made.

2.1.1 Radon source and experimental device

In this research, three different uranium sources have been characterized. As a result of uranium decay process, radon is exhaled, which is alpha emitter. These sources will therefore be used as a source of radon in different experiments. Each of them presents a different activity value, appointed in increasing order:

- Source 1: 1.84 kBq
- Source 2: 4.27 kBq
- Source 3: 45 kBq

The experimental air chambers used in this investigation are two high density polyethylene deposits: one of 120 L volume and a smaller one, of 30 L. Both of them are impermeable to radon and are sealed from the environment.
Inside the deposits, radon sources are placed just in the bottom and above it, there is a metallic support where the detector is measuring air radon concentration in a continuous way. The diagram of the experimental devices is shown in Figures 1 and 2:

Figure 1. Experimental device: large deposit of 120 L volume.

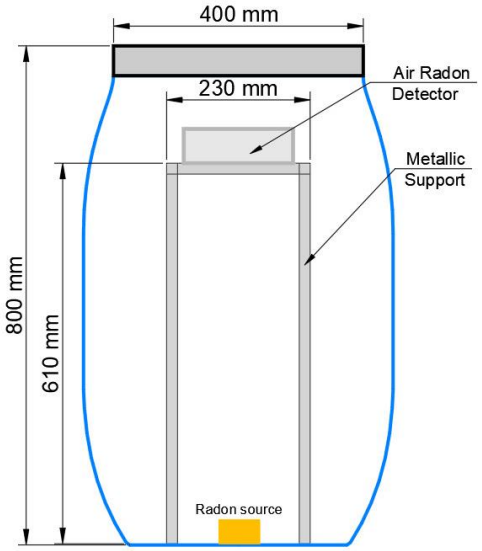
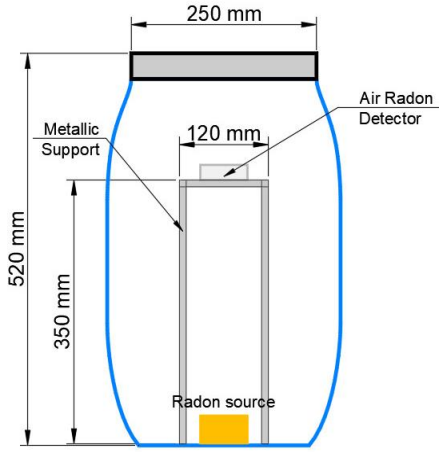


Figure 2. Experimental device: small deposit of 30 L volume.



2.1.2 Air radon detector

For this research, air radon concentration inside both deposits has been measured with the Radon Scout Plus detector (SARAD Company). It is a semi-conductor based continuous radon monitor, with a high voltage detector inside, operating in a passive diffusion mode. The used technique is based in gross alpha detection. Air radon concentration is obtained through the short-lived descendants of radon that are produced in its decay inside the chamber. Its range measurement varies from 0 to 2 MBq/m^3 with an error ± 5% or smaller, within the whole range, according to manufacturer. Additionally, the instrument works in the diffusion mode, so that a possible influence of thoron can be excluded. Relative humidity, air temperature and barometric pressure are also measured simultaneously. The detector response does not show any sensitivity against environmental changes in the atmosphere. (Sarad Manual, 2017).

2.2 Theoretical model for radon concentration in air: Closed Compartment Model

In this research, the Closed Compartment Model is applied in order to characterize radon sources as well as to predict its concentration in air during a certain exposure time.
This model is widely used to analyze radon behavior in air in a closed location, isolated from the environment. The CCM equation is:

$$\frac{dC_{Rn}}{dt} = \phi - \lambda \cdot C_{Rn} \quad (1)$$

Where C_{Rn} is radon concentration in water (Bq·m^{-3}), ϕ is the radon exposition rate in Bq·m^{-3}·h^{-1}, and λ is a decay constant (h^{-1}) which includes the two contributions shown in equation (2): λ_{Rn}, which is the radon decay constant (h^{-1}) and λ_v, which is the system leakage constant (h^{-1}).

$$\lambda = \lambda_{Rn} + \lambda_v \quad (2)$$

Applying the constant variation method, the equation for the evolution of radon concentration results in the following expression:

$$C_{Rn}(t) = C_0 \cdot \exp(-\lambda \cdot t) + \frac{\phi}{\lambda} \cdot (1 - \exp(-\lambda \cdot t)) \quad (3)$$

Where C_0 (in Bq·m^{-3}) is radon concentration at the initial time and t is the exposure time (h). Rearranging terms and assuming that the initial value of radon concentration in air is null, the equation of the Closed Compartment Model to be applied in this research is:

$$C_{Rn}(t) = \frac{\phi}{\lambda} \cdot (1 - \exp(-\lambda \cdot t)) \quad (4)$$

From this equation, the maximum radon concentration value in air to be achieved in the system can be calculated by the following expression:

$$C_{Rn\,max} = \frac{\phi}{\lambda} \quad (5)$$

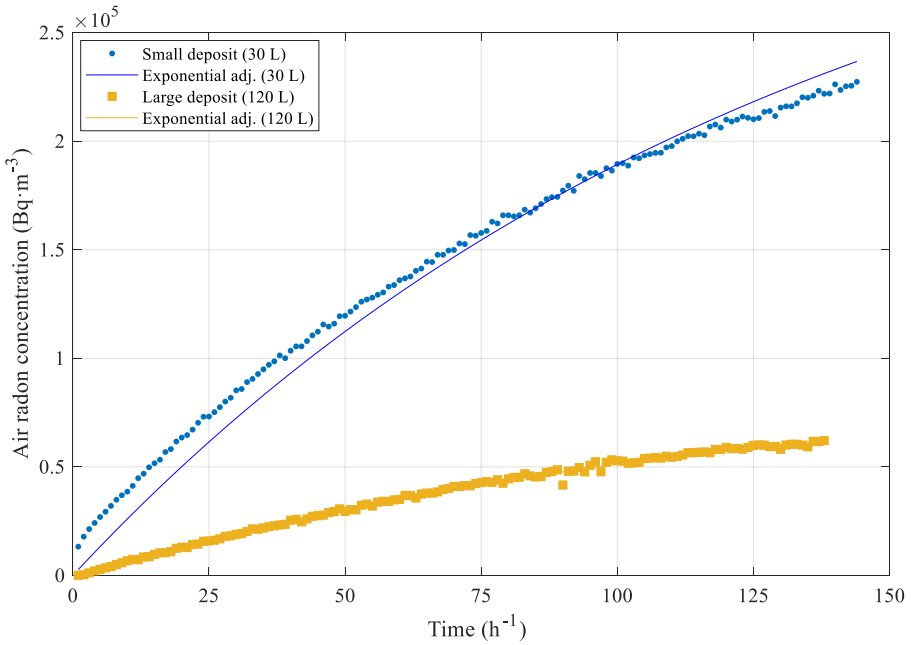
The leakages in the system, for both deposits are considered negligible, so the value of the decay constant is set considering $\lambda = \lambda_{Rn} = 0.00756\ h^{-1}$. This hypothesis has been assumed for the three sources and in both volumes tested.

3 RESULTS

3.1 Analysis of air radon concentration evolution from source 1

Initially, radon concentration in air due to the source 1(1.84 kBq) emission has been measured in both deposits (30 L and 120 L). Results are shown in Figure 3.

Figure 3. Air radon concentration from radon source 1 (1.84 kBq) in two different deposits (30 L and 120 L)



As it is shown, air radon concentration inside the large deposit is lower than in the smaller one due to the air volume difference. The smaller the amount of air available, the higher radon concentration is

accumulating, reaching $2.22 \cdot 10^5$ Bq·m^{-3} in the small volume, whereas the radon concentration reached for the large one has been $6.21 \cdot 10^4$ Bq·m^{-3}. Three points from the two tests have been selected in order to calculate the relative difference between both as:

$$Relative\ difference(\%) = \frac{[Rn]_{Small\ deposit} - [Rn]_{Large\ deposit}}{[Rn]_{Small\ deposit}} * 100 \quad (6)$$

Results are shown in Table 1.

Table 1. Relative difference calculation in three different exposition times for radon source 1

	Radon source 1 (1.84 kBq)		
	Air radon concentration in the small deposit (Bq·m^{-3})	Air radon concentration in the large deposit (Bq·m^{-3})	Relative difference (%)
Exposition time t = 1 h	$1.32 \cdot 10^4$	67	99.49
Exposition time t = 30 h	$8.52 \cdot 10^4$	1.8910^4	77.81
Exposition time t = 96 h	$1.85 \cdot 10^5$	$5.24 \cdot 10^4$	71.75
Exposition time t = 138h	$2.22 \cdot 10^5$	$6.21 \cdot 10^4$	72.01

As presented, the radon transfer from the source in a small air volume such as 30 L causes a much higher accumulation of Rn, as it was expected, because the same amount of radon is diluted in a lower volume of air. As the test progresses and the gas accumulates, the difference in concentration in the two tanks decreases from 99.49% to 72%. This is probably due to the stabilization of radon diffusion rate with time.

Likewise, in both cases the Closed Compartment Model (CCM) has been applied to determine if the results can be fitted to this mathematical model regardless of the volume of air in which radon accumulates. As it can be seen in Figure 3, for both the large and small deposits, the results follow an exponential trend which allows predicting the air radon concentration as a time function (in hours). In addition, the values obtained are adjusted to the CCM with an R^2 greater than 0.99. The CCM equations for each deposit and for source 1 are shown in Table 2.

Table 2. Application of the closed compartment model for radon measurements in air emitted by source 1

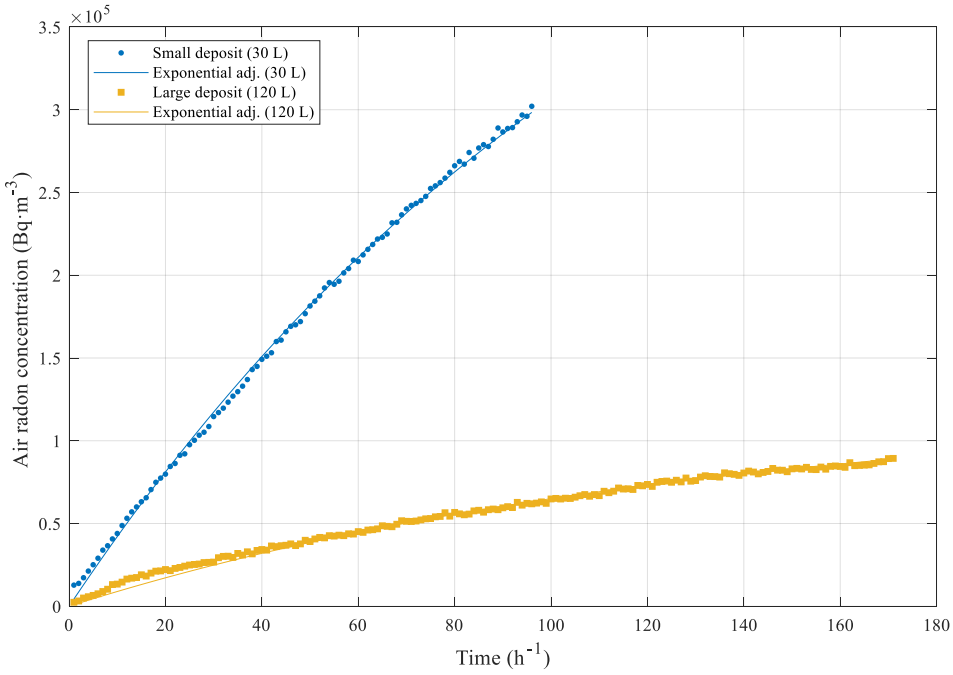
	Radon source 1: 1.84 kBq
Small deposit	$C_{Rn}(Bq \cdot m^{-3}) = 3.57 \cdot 10^5 \cdot (1 - \exp(-0.00756 \cdot Time))$
Large deposit	$C_{Rn}(Bq \cdot m^{-3}) = 9.67 \cdot 10^4 \cdot (1 - \exp(-0.00756 \cdot Time))$

From these equations it is possible to predict the levels of radon in air as a function of time (in hours) and the maximum value of radon concentration in air that will be reached. For the small deposit, the maximum value is $3.57 \cdot 10^5$ Bq·m^{-3} while for the large tank, it is $9.67 \cdot 10^4$ Bq·m^{-3}

3.2 Analysis of air radon concentration evolution from source 2

This section analyses the results of radon in air due to exposure of source 2 in the sampled air volumes. Results are shown in Figure 4.

Figure 4. Air radon concentration from radon source 2 (4.27 kBq) in two different deposits (30 L and 120 L)



Once again, the results obtained show that the smaller the volume of air, the greater the accumulation of radon inside the deposit and therefore, the higher the exposure rate. In the small deposit, the air radon concentration reached $3.02 \cdot 10^5$ Bq·m^{-3} while in the 120 L volume deposit, the reached concentration is lower, $6.19 \cdot 10^4$ Bq·m^{-3}. The slope of the line is much steeper than for the large tank, as a higher source activity in a smaller volume of air causes a much faster accumulation of radon. Additionally, three points have been selected from the Figure 4 from which the relative difference has been calculated according to Equation 6.

Table 3. Relative difference calculation in three different exposition times for radon source 2

	Radon source 2 (4.27 kBq)		
	Air radon concentration in the small deposit (Bq·m^{-3})	Air radon concentration in the large deposit (Bq·m^{-3})	Relative difference (%)
Exposition time t = 1 h	$1.28 \cdot 10^4$	$2.41 \cdot 10^3$	81.15
Exposition time t = 30 h	$1.15 \cdot 10^5$	$2.67 \cdot 10^4$	76.71
Exposition time t = 96 h	$3.02 \cdot 10^5$	$6.19 \cdot 10^4$	79.51

The air radon concentrations for the experimental tests with source 2 are higher than those of source 1, as it would be expected since this source is 4 times higher than source 1. Higher source activity, higher concentration of radon in air at different times.
With regard to the relative differences, in this case, values range between 81.15% and 79.51%, being more constant than for source 1. It can be observed that orders of magnitude of the differences in both source 1 and 2 are very similar. This suggests that the speed of radon diffusion is not significantly influenced by the concentration of radon in the air. On the other hand, it is verified that the smaller the volume of air, the greater the accumulation of radon

Results, in both cases, as shown in the Figure 4, are adjusted to an exponential equation that corresponds to the Closed Compartment Model with an R^2=0.99. The CCM equations for each deposit are shown in Table 4.

Table 4. Application of the Closed Compartment Model for radon measurements in air emitted by source 2

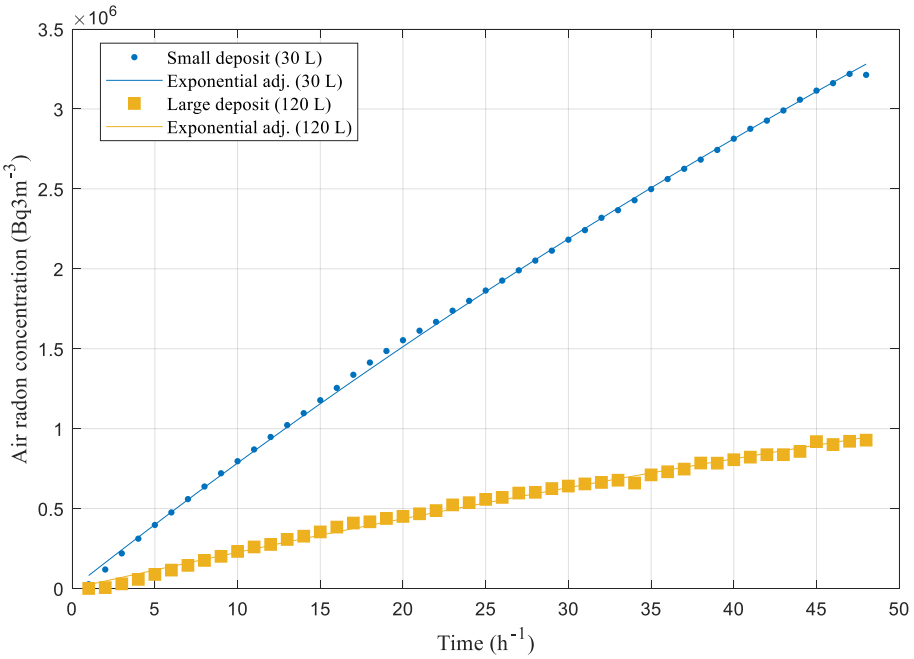
	Radon source 2: 4.27 kBq
Small deposit	$C_{Rn}(Bq \cdot m^{-3}) = 5.78 \cdot 10^5 \cdot (1 - \exp(-0.00756 \cdot Time))$
Large deposit	$C_{Rn}(Bq \cdot m^{-3}) = 1.24 \cdot 10^5 \cdot (1 - \exp(-0.00756 \cdot Time))$

According to these previous equations, maximum air radon concentration that could be reached by source 2 in the smaller deposit is $5.78 \cdot 10^5$ Bq·m^{-3} while for the large deposit, it is $1.24 \cdot 10^5$ Bq·m^{-3}. The higher the activity of the source, the higher the air radon concentration that could be reached.

3.3 Analysis of air radon concentration evolution from source 3

The third of the exposed radon sources has an activity of 45 kBq. Results of air radon concentration are shown in Figure 5.

Figure 5. Air radon concentration fromradon source 3 (45 kBq) in two different deposits (30 L and 120 L)



As in the previous tests, there is a greater accumulation of radon inside the small deposit due to the increase of the activity source, comparing with source 1 and 2. In this measurement, air radon concentration in the small deposit reached $3.02 \cdot 10^5$ Bq·m^{-3} while for the large deposit, the air concentration reaches a value of $8.93 \cdot 10^4$ Bq·m^{-3}. Three exposure points have also been selected in order to calculate the relative difference according to Equation 6. Results are shown in Table 5.

Table 5. Relative difference calculation in three different exposition times for radon source 3

	Radon source 3 (45 kBq)		
	Air radon concentration in the small deposit (Bq·m^{-3})	Air radon concentration in the large deposit (Bq·m^{-3})	Relative difference (%)
Exposition time t = 1 h	$2.64 \cdot 10^4$	$1.10 \cdot 10^2$	99.58
Exposition time t = 30 h	$2.18 \cdot 10^6$	$6.41 \cdot 10^5$	70.63
Exposition time t = 48 h	$3.21 \cdot 10^6$	$2.98 \cdot 10^5$	71.12

As observed, the higher the activity of the source, the higher the concentration of radon in air. In this measurement test, air radon concentration in the small deposit reached $3.21 \cdot 10^6$ Bq·m^{-3} and $2.98 \cdot 10^5$ in the large deposit. Comparing with source 1 and source 2, at the same measured time, 30 hours, in the small deposit, air radon concentration due to source 1 is $8.52 \cdot 10^4$ and $1.15 \cdot 10^5$ due to source 2 and $2.18 \cdot 10^6$ due to source 3.

Relative difference varies in this experimental test from 99.50% to 71.12%. The difference between the concentration of radon in air in the large and small tank decreases over time, as in the case of source 1. However, between the second and third point of exposure (t=30 hours and t=48 hours) the relative difference slightly varies, and the values are quite similar to those of sources 1 and 2. This would imply that both concentrations are growing in the same proportion from that point.
Results have been adjusted to the exponential equation satisfactorily with an R^2 greater than 0.99 as shown in the Figure 5. The adjusted equations for each deposit are shown in the Table 6:

Table 6. Air radon concentration due to radon source 3 (45 kBq) in two different deposits (30 L and 120 L)

	Radon source 3: 45 kBq
Small deposit	$C_{Rn}(Bq \cdot m^{-3}) = 1.08 \cdot 10^7 \cdot (1 - \exp(-0.00756 \cdot Time))$
Large deposit	$C_{Rn}(Bq \cdot m^{-3}) = 3.11 \cdot 10^6 \cdot (1 - \exp(-0.00756 \cdot Time))$

The maximum concentration value that would be reached in these two tanks is much higher than in previous tests due to the higher activity of the radon source: $1.08 \cdot 10^7$ Bq·m^{-3} in the small deposit and $3.11 \cdot 10^6$ Bq·m^{-3} in the bigger ones.

3.4 Analysis of the exposition rate obtained from the CCM

As detailed above, the Closed Compartment Model predicts the concentration level in air and allows the calculation of the maximum concentration value to be reached in the system. From the CCM model, the source exposure rate value is also obtained, which is its concentration in air per unit time. These values are shown in the Table 7.

Table 7. Exposition rate results for according the CCM

	Small deposit (30 L)	Large deposit (120 L)
Φ_1, exposition rate source 1 (Bq·m^{-3}·h^{-1})	$2.70 \cdot 10^3$	$7.30 \cdot 10^2$
Φ_2, exposition rate source 2 (Bq·m^{-3}·h^{-1})	$4.37 \cdot 10^3$	$9.26 \cdot 10^2$
Φ_3, exposition rate source 3 (Bq·m^{-3}·h^{-1})	$8.15 \cdot 10^4$	$2.34 \cdot 10^4$

As it can be seen, the exposure rate in both deposits is increasing due to the source activity: source 3 (45 kBq) has a higher exposure rate ($8.15 \cdot 10^4$ and $2.34 \cdot 10^4$ Bq·m^{-3}·h^{-1}). If the results are compared between both deposits, it is noted that the higher the volume of air, the lower the exposure rate, since the same amount of radon is higher diluted in a higher volume of air.

4 CONCLUSIONS

In this research, three different radon sources (1.84 kBq, 4.27 kBq and 45 kBq) have been used for radon accumulation two deposits: 30 L and 120 L.

According to experimental results, it has been verified that the lower the activity of the source, the lower the concentration of radon in air for both the 30 L and 120 L deposits. With regard to the effect of air volume in radon concentration, it is shown for all sources that the air radon concentration, for the same source, in an enclosed location accumulates exponentially.

Analyzing the order of magnitude in the tests, it is observed that is similar for all three sources in several instants of time. This could mean a low influence of airborne radon concentration on their diffusion rate. Likewise, in all measurements, for all the sources tested as well as for both deposits, there is an exponential growth in airborne radon levels which fits the Closed Compartment Model. This adjustment has an R^2 greater than 0.99 in each test.

This mathematical model is not only a method for estimating radon levels in air but also a way of understanding its behavior and the physical parameters that characterize different sources in each situation such as emission rate, and maximum concentration that the gas can reach in that particular air volume in which the tests are carried out.

Moreover, from the CCM, the exposure rate values (concentration per unit time) can be extracted for each source. As expected, the higher the activity of the source, the higher its exposure rate. This rate varies from $2.70 \cdot 10^3$ (source 1) to $8.15 \cdot 10^4$ $Bq \cdot m^{-3} \cdot h^{-1}$ (source 3) in the small deposit and from $7.30 \cdot 10^2$ (source 1) to $2.34 \cdot 10^4$ $Bq \cdot m^{-3} \cdot h^{-1}$ (source 3) in the large one. It is also verified that this rate varies depending on the volume of air. If the volume of the deposit is greater, radon will have a greater amount of air to diffuse in and therefore there will be a lower concentration and a lower air-radon ratio.
The objectives of this investigation have therefore been effectively fulfilled. It has been shown that radon, under conditions of isolation from the outside, behaves in a way which can be adjusted to an exponential equation such as that of the CCM, and the influence of radon source activity and enclosed air volume have been analysed.

5 ACKNOWLEDGEMENTS

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