

# **Measurement of Granite Countertop and Other Building Materials Radon Emanation**

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## **Abstract**

Granite countertops and other natural materials used in building construction may have concentrations of uranium series radionuclides that are higher than the local average background. Further, the concentration within the material may be highly variable and may contain inclusions that have concentrations much higher than the material average. Such materials have the potential to emanate radon to the extent that the radon concentration of indoor air is significantly increased. A study of radon emanation from polished granite sheets was performed and two simple tests were devised to determine the radon emanation rate. A generic calculation for the radon concentration potential increase was proposed, but is no substitute for direct measurement of the room air concentration.

## **Introduction**

Concern for radon emanation in news and Internet reports prompted several studies of radon emanation from granite countertops and caused similar concern for other building structural and decorative materials (Kitto et al, 2008) (Brodhead, 2008) (Sundar et al.,2003). Uranium (U-238) and its subsequent progeny radon (Rn-222) are naturally occurring radioactive materials that are found in nearly all natural materials at about 1 pCi/g, but with large variations by material and locality. Radon may emanate from the material at a rate dependent upon material properties and surface area. Radon emanation into a closed structure may increase the indoor radon concentration depending upon the exchange rate of the indoor and outdoor air.

The U.S. Environmental Protection Agency (EPA) and other national bodies recommend or require that the homes and occupied structures maintain the indoor radon concentration below a specified limit. EPA recommends a limit of 4 pCi/L.

## **Radon Emanation Theory**

Fick's laws can describe the diffusion of one gas into another if the diffusing gas is sufficiently dilute. Fick's first law in one dimension states that the flux of the gas across a surface is proportional to the product of the diffusion coefficient of the gas and the concentration gradient across the surface,

$$J = -D \frac{dC}{dx}, \quad 1$$

$J =$	Flux in molecules $\text{m}^{-2} \text{s}^{-1}$
$D =$	Diffusion coefficient in $\text{m}^2 \text{s}^{-1}$
$C =$	Concentration in molecules $\text{m}^{-3}$

$x =$	Distance from the surface in m
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Fick's second law describes the change in concentration with time,  $t$ , at a location,

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2}. \quad 2$$

The solution to eqn 2 is

$$C_{x,t} = C_0 \operatorname{erfc}\left(\frac{x}{2\sqrt{Dt}}\right). \quad 3$$

The function  $\operatorname{erfc}$  is the complimentary error function and  $C_0$  is the concentration at the source surface. Eqns 2 and 3 show that the flux across the surface decreases over time as the concentration in the diffusion volume increases. Some authors call the reduction in flux back diffusion (Christopher et al., 1999). Eqn 3 is important for a closed system in which  $C_{x,t}$  approaches  $C_0$  at which point net diffusion across the surface approaches zero. An open system in which the air in the diffusing volume is well mixed and receives a continuous in-flow of clean air is of particular interest since most occupied volumes, where radon concentration may be of concern, fall into this category.

The radon concentration of a well-mixed, open system having an air exchange rate,  $E$ , and receiving a constant source of radon atoms,  $S$ , each second can be described by

$$\frac{dC}{dt} = \frac{S}{V} - \frac{CE}{V} - \lambda C. \quad 4$$

$\lambda =$	Radon decay constant in $\text{s}^{-1}$
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Eqn 4 integrates to

$$C = \frac{S}{E + \lambda V} \left(1 - e^{-\left(\frac{E}{V} + \lambda\right)t}\right). \quad 5$$

The radioactive decay can be neglected in eqn 5 if the exchange rate is large relative to  $\lambda V$ . For example, at one air change each hour,  $E = V$ , and for  $\lambda = 7.55 \times 10^{-3} \text{ h}^{-1}$ , the decay constant is negligible in the denominator and exponent in eqn 5. The diffusion equations indicate that in a well-mixed, open system where the change from initial to final concentration is small, there is little change in flux across the surface, so within limit, the change in flux can be ignored.

### **Materials and Methods**

Emanation rates for various materials were established in flow-through chambers using various materials and standards and two separate continuous radon concentration monitors (CRM). An accredited radon calibration laboratory calibrated the CRMs. One

radon emanation standard was obtained from National Institute for Science and Technology (NIST) and the other standards were materials that were evaluated by two other investigators. Fresh, radon-free air was drawn through the test chamber at a constant rate. The air was rendered radon-free by passing through an activated carbon filter. The filter was of sufficient volume that the break-through time was longer than 10 Ra-222 half-lives.

The material emanation rate was calculated from the formula

$$C = \frac{S}{E}, \quad 6$$

where

C =	Concentration in Bq m <sup>-3</sup>
S =	Source emanation rate in Bq s <sup>-1</sup>
E =	Volume exchange rate in m <sup>3</sup> s <sup>-1</sup>

Eqn 6 is eqn 5 under equilibrium conditions when radioactive decay and change in flux are negligible in the flow-through system. The equilibrium concentration of the flow-through system is equal to the emanation rate divided by the exchange rate, i.e., a volume of air at the equilibrium concentration replaces an equal volume at the same concentration.

There are two advantages of the flow-through method over the closed volume method of measuring emanation rate. One is that equilibrium is reached quickly if the exchange rate is on the order of 10% of the total volume (90% equilibrium in 24 h). The other advantage is it is not necessary to know the volume of the chamber. It is necessary to measure the exchange rate (flow rate into the chamber) and to maintain the exchange rate low enough to obtain sufficient sensitivity for the radon concentration measurement. The flow rate in these measurements was made with a calibrated electronic mass flow meter.

A NIST radon emanation source was used to calibrate all systems using the method of (Kotrappa et al., 2004). The source was rated at 372.7 Bq s<sup>-1</sup>.

The characterized materials and the NIST source were used to test two methods for simple field measurement of emanation rate. The first method was an activated carbon packet below an overturned, shallow disposable dish<sup>1</sup>. The dish was sealed to the surface using KY Jelly (other potential sealants were potentially harmful to granite). The second method was a square of activated carbon cloth covered by a sheet of plastic<sup>2</sup>.

The packet comprised 30 g of activated carbon in a 10x10 cm Tyvek envelope. The covering dish was made of plastic and had a volume of 1.1 L and enclosed a surface area of 0.05 m<sup>2</sup>.

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<sup>1</sup> Copyright AirChek, inc.

<sup>2</sup> Patent applied for.

The activated carbon cloth was 0.3 m<sup>2</sup> and weighed 13 g. The carbon cloth was covered with Kraft paper of the same dimensions and taped to the surface with black electrical tape after covering with aluminum foil of the same dimensions.

The collection method for the two systems is well-established with the best known example the Large Area Activated Charcoal Canisters (EPA 2008) collectors for radon emanation from soil. The concentration on the charcoal follows

$$\frac{dC}{dt} = S - \lambda C, \quad 7$$

which integrates to

$$C = \frac{S}{\lambda} (1 - e^{-\lambda t}). \quad 8$$

The tests of the methods showed that both methods closely followed eqn 8. The methods reach 90% equilibrium in 10 days. The tests need run only to the point where sufficient sensitivity is obtained for the test. The capacity of the charcoal is sufficient to ignore back diffusion from the charcoal.

## Results and Discussion

The two field methods for measuring radon flux from a surface were calibrated against a NIST emanation source and both accumulator and flow-through methods of measuring radon flux. The agreement with a series of measurements was with a coefficient of variation near 5% for all devices.

The data for the activated carbon packet system showed more scatter than the activated carbon cloth method. The smaller collection area of the activated carbon packet method caused the increased scatter. The results for the carbon packet method changed if the collection region was repositioned on a material that exhibited variation in exposure rate across the surface, while sufficient carbon cloth was used to cover the entire test surface.

The scatter shown by the carbon packet method reveals the problem with making a single radon emanation measurement on a surface exhibiting variation in exposure rate. Several measurements are necessary to obtain a good estimate of the total emanation from the surface, if it is impractical to measure emanation from the total surface. It is left to the investigator define a 'good' estimate based on the requirements of and the intent of the measurement.

This study was initiated to develop a practical method for determining the radon emanation rate from granite countertops to determine the potential radon concentration increase in a home from the installed granite. The concentration increase would be predicted using eqn 6 and an assumed air exchange rate.

For example, in the case of a flux of  $10 \text{ Bq m}^{-2} \text{ s}^{-1}$  from  $3 \text{ m}^2$  of surface would be  $30 \text{ Bq s}^{-1}$ . Given a room volume of  $12 \text{ m}^3$  and a room air exchange rate of  $1.2 \text{ m}^3 \text{ h}^{-1}$ , the equilibrium radon concentration according to eqn 6 is  $1500 \text{ Bq m}^{-3}$ .

The example is for a single room, but in most cases, the whole house should be considered. The example is for an air exchange rate of one-tenth room volume each hours. Some commercial buildings may have exchange rates as high as 12 changes each hour, while newly constructed homes may be 0.1 air change or less each hour. While careful measurement of surface emanation rates can predict the equilibrium concentration of radon of a room, such a prediction is no substitute for measuring the actual concentration under normal circumstances. The usefulness of the methods reported here is to determine the source of radon, if concentrations above the limit are found.

### **Conclusions**

Two simple techniques for measuring radon emanation were demonstrated that allow field determination of emanation rates without instrumentation and elaborate set up and system volume determination.

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## **Uncertainties in the evaluation of the dose coming from radon in tourist caves**

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### **Abstract**

Indoor radon poses a health risk confirmed by a variety of studies. High concentrations of this gas can be found in closed enclosures depending on several factors like activity of source term, permeability of the source/indoor air inter-phase, as well as the interchange rate of external air. Low ventilation rates together with the presence of <sup>226</sup>Ra in rocks, make the tourist caves a place where medium and high radon concentrations can typically be found. With the incorporation of EURATOM basic standards for radiological protection, into the national European legislations radon have been recognised as a health risk to be controlled in workplaces. The transfer of EURATOM standards to the Spanish legislation came out on Title VII (BOE 178/2001) where radiation coming from natural sources has an analogous role as radiation emitted from artificial ones.

The most significant exposure to radon in show caves is received by tourist guides who work providing tours for the general public. The implementation of an efficient radiological protection system for this kind of workplaces must take into account that in some circumstances forced ventilation may alter the humidity inside the cave affecting some of the formations or paintings. For this reason the best option to protect workers from radon is a system based on limitation of exposure by restricting the amount of time spent in the cave. For doing so, the knowledge of main uncertainty sources in dose evaluation is of main importance.

The principal variability sources in dose assessment are the integration intervals for radon measurements and the aerosol conditions related with the unattached fraction of radon progeny. The first factor takes into account the daily and monthly variations in radon concentrations and can not be fully addressed in caves with the seasonal correction factors proposed for the determination of radon doses in houses. On the other hand, mainly due to the low particle concentration usually present inside the caves, the unattached fraction of radon progeny can be higher than in other workplaces. This factor is significant when dose assessment is approached from the dosimetric respiratory track model of ICRP.

In this work the results of radon measurements carried out monthly in different points of 7 caves located in the region of Cantabria (Spain) as well as estimations of the dose received by workers approached from different integration intervals are presented. Also the results concerning particle concentration and its relationship with unattached progeny fraction together with their implications in dosimetric calculations are discussed for some of the caves.

**KEYWORDS:** *radon, dose, tourist caves*

## 1. Introduction

When we breathe in air with a concentration of radon and radon progeny, several alpha radioactive decays take place inside the lung. This irradiation is responsible of about half of the annual average effective dose received by the human due to natural sources of radiation. [1]. Outdoor radon does not represent a significant health hazard because high concentrations are never reached. However, it becomes a problem when released into a closed or poorly ventilated enclosures like dwellings, buildings and also caves and mines. Indoor concentrations of radon and its short-lived progeny depend on several factors mainly related with the entry or production rate from various sources and the ventilation rate.

Many workplaces both above and below ground may be affected by high radon concentrations. On its 1990 publication, the International Commission of Radiological Protection, ICRP recommended that exposure to high radon levels should be considered as occupational exposure and remedial actions have to be taken in such situations [2]. Concerning the Spanish situation regarding radiation coming from natural sources, in 2001 the Spanish law incorporated EURATOM basic standards for radiological protection, which include a request at the EC Member States to determine the working places on which exposure to natural radiation is significant. On Title VII (BOE 178/2001) radiation coming from natural sources plays the same role than radiation emitted from artificial sources.

Tourist caves represent a type of workplace with particular environmental conditions that might be affected by high radon concentrations [3, 10]. In these places in which guides host visits for the general public, typical remedial actions like forced ventilation, sealing or reducing pressure in the source rock can not be used for conservation reasons. For example, forced ventilation could alter the humidity inside the cave thus affecting the paintings or geological formations that attract the tourists. So in most of the cases the only way to reduce radon exposure to guides and other workers is to apply a radiation protection system based on restrictions in the amount of time spent in the cave.

The information needed for carrying out the abovementioned protecting actions is related with the specific characteristics of the cave concerning the behaviour of radon and its decay products. In order to perform a precise effective dose calculation, factors like unattached progeny fraction ( $f_p$ ), equilibrium factor (F) and particle concentration (Z) are of main importance. One of the specific characteristic of the caves is the high unattached fraction due to a particle concentration far below from the usual value in dwellings. The  $f_p$  values can be higher than 0.1, for places with low ventilation and without additional aerosol sources, with  $Z < 4 \cdot 10^3$  particle  $\text{cm}^{-3}$ .

In the present work, the results of radon measurements carried out monthly in different points of 7 caves located in the region of Cantabria (Spain), showed in Fig. 1, as well as estimations of the dose received by workers and general public are presented. Radon exposure values are calculated from different measurement integration intervals in order to stress its importance as a source of variability in dose assessment. Additionally the results concerning particle concentration and its relationship with

unattached progeny fraction together with their implications in dosimetric calculations are discussed.

## 2. Material and methods

Radon measurements were carried out within 7 caves located in the region of Cantabria in the North of Spain (Fig. 1). Radon detectors were exposed monthly along 2008 and placed inside of each cave following criteria related with most probable risk situations for workers. The analysed points were those in which guides usually spent longer periods giving explanation to the public.

CR39 track-etched detectors were used for integrating measurements. Every CR-39 detector was fastened under the cap of a cylindrical polypropylene container 55 mm high and 35 mm diameter which prevents radon decay products and also  $^{220}\text{Rn}$  from entering. Then, only alpha particles from radon that has diffused into the container and from the polonium produced inside can strike the detector. After the exposure time an etching process is performed, and the radon concentration determined by counting the tracks in a given area. Accuracy and precision of this method has been tested in a National intercomparison exercise [11], as well as in a recent European intercomparison with good results (not yet published).

Also particle concentration was also measured by means of a condensation particle counter (CPC ISI 3007). In this device, air is pumped at a rate of  $100\text{ cm}^3\text{ min}^{-1}$  and passed throughout a porous wick containing liquid isopropyl alcohol. After the exposure of the sample to the alcohol vapour, particles grow by condensation and can be detected optically with a laser light and a detection unit. With this system particle concentrations in the range of 0 to 500,000 particle  $\text{cm}^{-3}$  can be detected.

Mean annual effective doses coming from radon inhalation have been estimated by using ICRP65 dose assessment methodology [4]. The dose conversion factor (DCF) used for radon exposure was 5 mSv per WLM at work and 4 mSv per WLM for the general public, and the dose assessment is usually performed by assuming an equilibrium factor of 0.4 and indoor occupancy 2000 hours per year. In the special case of workers in tourist caves it must be taken into account that the occupational time is significantly lower than 2000 hours per year. For this reason, doses have been estimated from the real distribution of time spent by tourist guides inside each cave. On the other hand, the singular atmospheric conditions characterized by high indoor humidity and very low particle concentrations makes more appropriate the use of an equilibrium factor of 0.6.

From an alternative approach, the effective dose can be determined using the respiratory track model of ICRP 66 [5]. For doing so, the measurement of unattached fraction  $f_p$  is essential. The dependence of the  $f_p$  as a function of particle concentration  $Z$  can be approximated by the semi-empirical equation [6]:

$$f_p = 400/Z \text{ (cm}^{-3}\text{)} \quad (\text{Eq. 1})$$

on the model of ICRP 66,  $\text{DCF}_u$  for inhalation of the unattached short-lived radon progeny in mSv per WLM can be calculated from equation:

$$DCF_u = 8.4 + 64 * f_p \quad (Eq. 2)$$

### 3. Results

#### 3.1 Radon concentration

It is known that radon concentration shows monthly variations in caves [8,12]. As an illustration, Fig. 2 represents the variation in radon levels at different sectors inside the cave of Las Monedas. As it can be seen, seasonal variations are clearer at specific sectors more influenced by external changes in meteorological parameters. In each cave, from the set of values obtained monthly at different points, the annual average radon concentration has been calculated. In order to assess the influence of the integration interval in the estimation of the annual effective dose, average radon concentrations were calculated from three and six month measurements.

In Table 1, a comparison of three-month integrated values with the annual mean is showed. Taking as a reference value  $1000 \text{ Bq m}^{-3}$ , which is the action level for radon concentration in workplaces established by IAEA in 1996 [7] it can be observed that all the studied caves present some points above this value, although all the annual means are below. It can be found significant relative differences as high as 90 % with the annual mean. In some cases, when the measurements are carried out in summer period, the annual exposition to radon can be overestimated by factors up to 2. On the other hand, mean values coming from winter or spring seasons can lead to an underestimation of similar magnitude. Fig. 3 illustrates the abovementioned differences.

As it would be expected, integrated values obtained from six-month measurements fit better to the monthly averaged annual radon concentration, as it can be seen in Fig. 4. However, also in this approach, overestimations up to 75 % can occur when the measurements are carried out during the second half of the year, and underestimations up to 400 % can be observed when the measurements are performed during the first half of the year.

#### 3.1 Dose assessment

According to the above mentioned ICRP65 methodology and to the information available about the real time spent by each tourist guide at different sectors of the studied caves, the annual effective dose was estimated. As it can be seen in Table 2, only three workers received doses above the general public's limit of 1 mSv per year. In the same way, Table 3 shows the dose coming from radon inhalation received by public in one typical visit for each cave.

From ICRP's human respiratory model point of view, the differences on aerosol conditions can modify the dose conversion factors. For the most usual aerosol conditions in homes of  $f_p = 0.08$  and equilibrium factor of 0.4, a DCF of 14 mSv per WLM has been obtained by Marsh et al. [9]. This DCF can significantly increase in caves, where particle concentration is very low and subsequently values of  $f_p$  as high as 0.8 can be found. The uncertainties in the calculation of DCF can be high using the

dosimetric model because it involves the use of parameters like weighting factors for alpha particles and lung tissues which are difficult to determine accurately. In spite of this consideration, the great differences observed between the DCF's obtained from both models show the main relevance on unattached fraction of radon progeny in the dose calculations.

In the present work, particle concentration was measured monthly at different sectors inside the caves. Table 4 shows a summary of values together with the corresponding dose conversion factors for the unattached fraction ( $DCF_u$ ) according to dosimetric model. Particle concentrations as low as  $100 \text{ cm}^{-3}$  were found in some points, which mean  $DCF_u$ 's from 3 to 8 times higher than that obtained from epidemiological approach. This would mean that in some work cases, radiological protection actions should be taken because doses up to 6 mSv per year could be possible.

#### **4. Conclusions**

With this study the annual mean radon concentration have been determined from monthly measurements at different points of several tourist caves. The results indicate that for an accurate dose assessment for workers under realistic situations the most detailed knowledge of the indoor radon levels evolution is needed. Approximations made from usual longer integration intervals as 3 or 6 months can lead to over or underestimations of the dose received by people working in the caves.

On the other hand, the extremely low particle concentrations found inside the caves can lead to higher doses coming from radon progeny inhalation than those received by people in workplaces with similar radon levels. Finally, in some cases particle concentrations lower than  $400 \text{ cm}^{-3}$  have been found which would mean an unattached fraction higher than 1. Maybe the relationship used in Eq. 1 can be questioned under special environmental conditions like those present in some tourist caves. This issue is now under analysis with new measurements of the unattached fraction in the studied caves.

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## Captions of figures and tables

Figure 1: Location of the studied caves in the Cantabria region in Spain

Figure 2: Continuous radon measurements in the Castillo cave during a 10 days period.

Figure 3: Continuous radon measurements in the Monedas cave during a 10 days period

Table 1: Integrated radon concentration and mean annual effective dose in several places inside each cave.

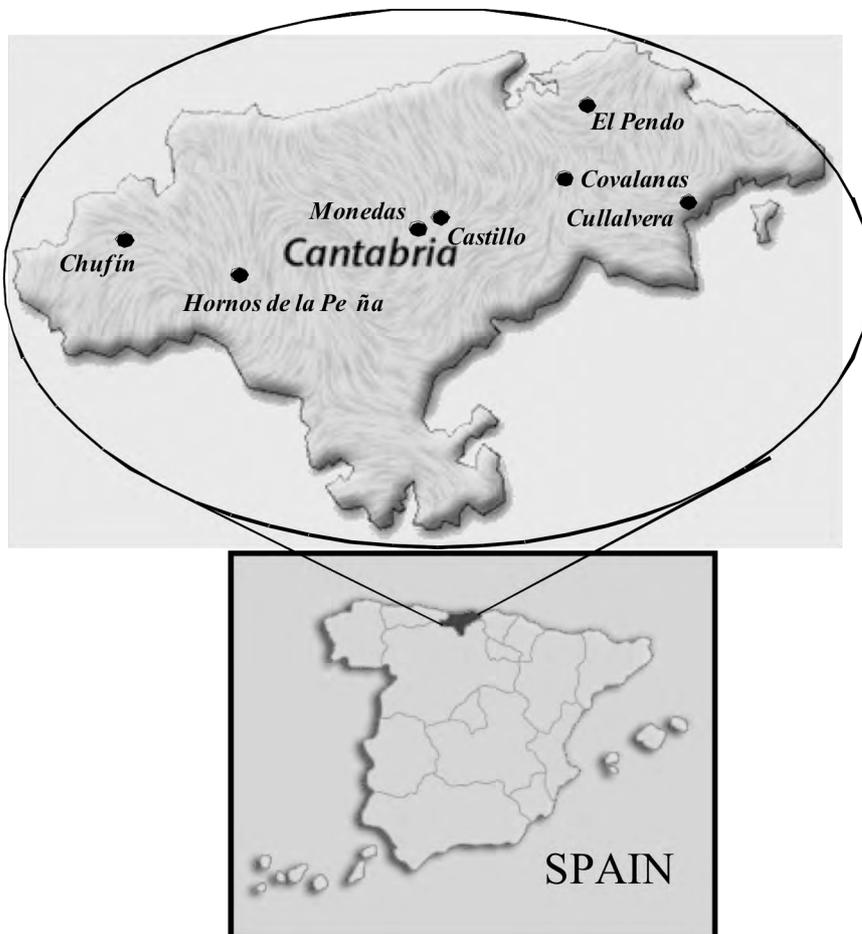


Figure 1

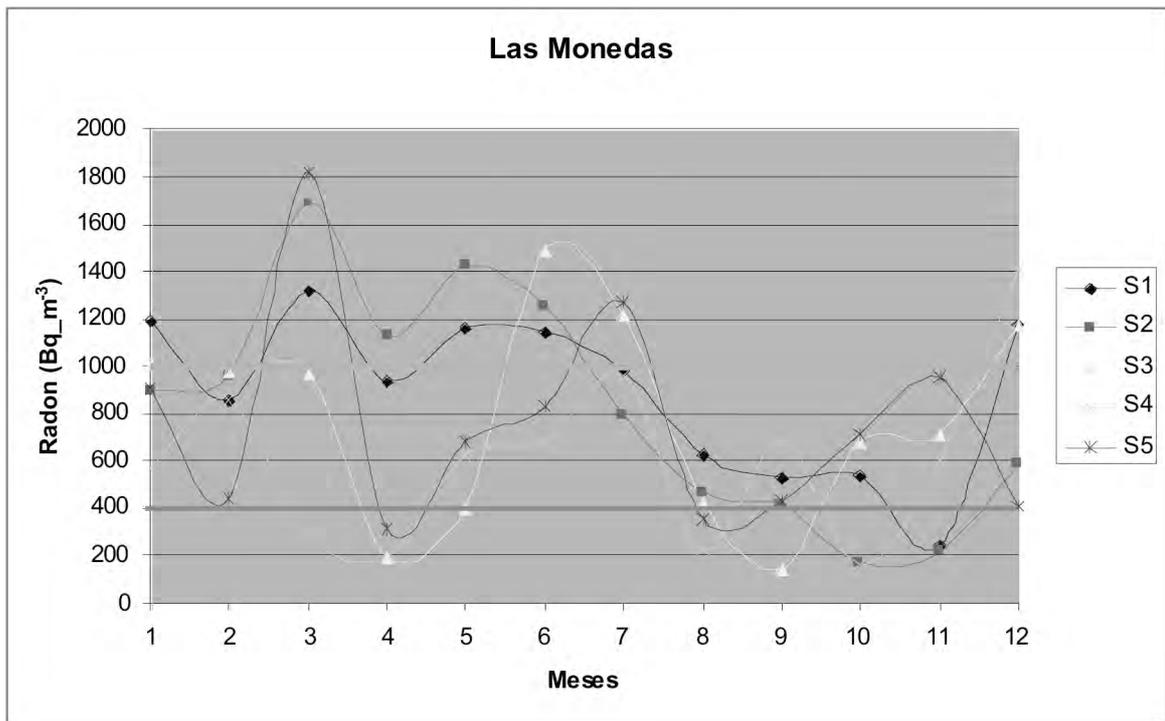


Figure 2

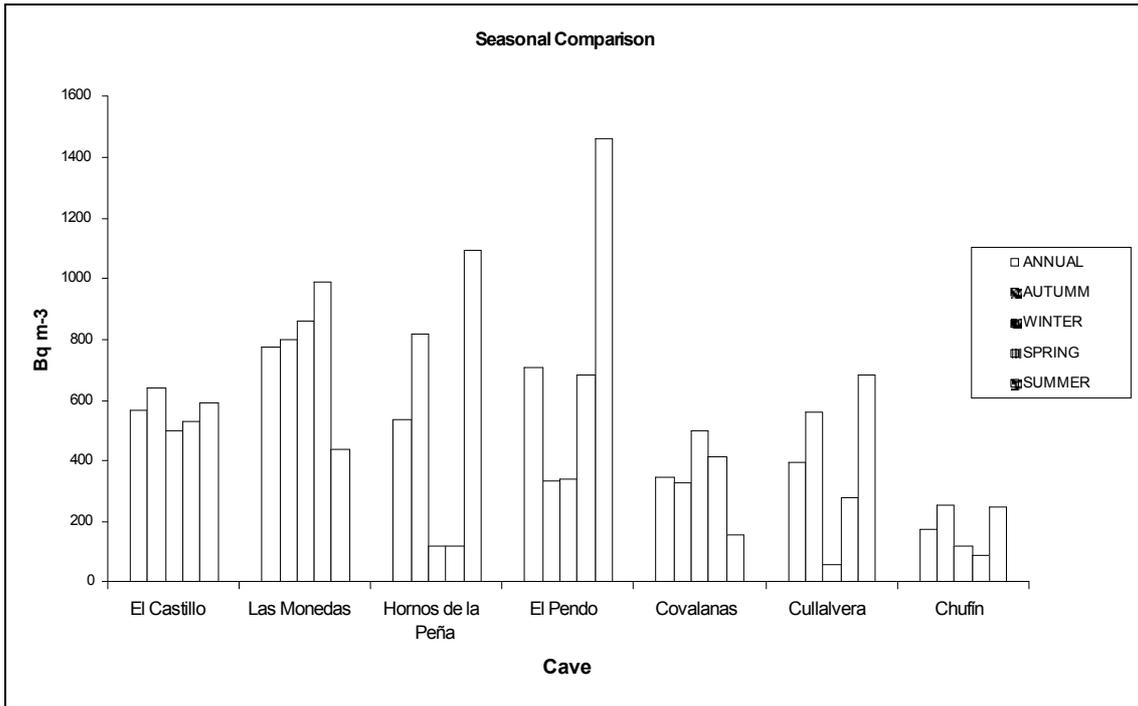


Figure 3

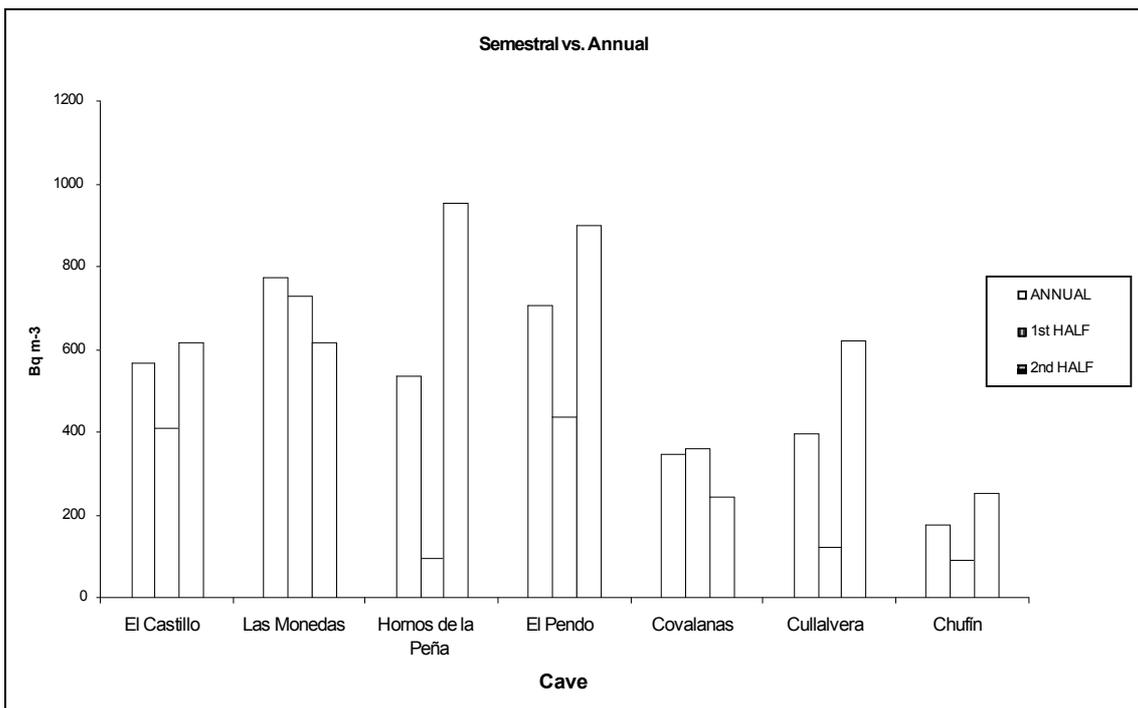


Figure 4

CAVE	MEAN RADON CONCENTRATION (Bq m <sup>-3</sup> )				
	ANNUAL	AUTUMN	WINTER	SPRING	SUMMER
<b>El Castillo</b>	571 (40- 1600)	643	500	534	595
<b>Las Monedas</b>	775 (170-1700)	804	863	995	438
<b>Hornos de la Peña</b>	539 (80-5300)	817	121	123	1096
<b>El Pendo</b>	707 (80-1700)	337	342	683	1465
<b>Covalanas</b>	350 (25-1110)	332	500	413	157
<b>Cullalvera</b>	397 (20-1600)	564	60	281	684
<b>Chufin</b>	179 (25-530)	256	121	88	250

Table 1

<b>Workers Tourist Guides</b>	<b>Dose (mSv y<sup>-1</sup>)</b>
<b>1</b>	<b>0.85</b>
<b>2</b>	<b>0.05</b>
<b>3</b>	<b>0.38</b>
<b>4</b>	<b>0.02</b>
<b>5</b>	<b>0.37</b>
<b>6</b>	<b>0.66</b>
<b>7</b>	<b>0.28</b>
<b>8</b>	<b>0.47</b>
<b>9</b>	<b>0.31</b>
<b>10</b>	<b>0.61</b>
<b>11</b>	<b>0.79</b>
<b>12</b>	<b>0.01</b>
<b>13</b>	<b>1.12</b>
<b>14</b>	<b>0.52</b>
<b>15</b>	<b>1.66</b>
<b>16</b>	<b>1.18</b>
<b>17</b>	<b>0.06</b>
<b>18</b>	<b>0.70</b>
<b>19</b>	<b>1.06</b>
<b>20</b>	<b>0.76</b>
<b>21</b>	<b>0.64</b>
<b>22</b>	<b>0.56</b>
<b>23</b>	<b>0.04</b>
<b>24</b>	<b>0.63</b>
<b>25</b>	<b>0.47</b>
<b>26</b>	<b>0.34</b>
<b>27</b>	<b>0.06</b>
<b>28</b>	<b>0.62</b>

Table 2

<b>Cave</b>	<b>Dose to the Public in one visit (mSv)</b>
El Castillo	0.0027
Las Monedas	0.0031
Hornos de la Peña	0.0088
El Pendo	0.0037
Covalanas	0.0021
Cullalvera	0.0023
Chufin	0.0015

Table 3

<b>Cave</b>	<b>Mean particle concentration (cm<sup>-3</sup>)</b>	<b>DCF<sub>u</sub> (mSv WLM<sup>-1</sup>)</b>
El Castillo	868 (100-350)	37,89
Las Monedas	1154 (600-3000)	30,58
Hornos de la Peña	3508 (440-19000)	15,70
El Pendo	1903 (400-5400)	21,85
Covalanas	1912 (370-16000)	21,79
Cullalvera	3106 (950-12000)	16,64
Chufin	3428 (1100-14000)	15,87

Table 4