Journal of Radiological Protection

Official journal of
the Society for
Radiological Protection

PAPER

Variations in radon dosimetry under different assessment approaches in the Altamira Cave

IOP Publishing | Society for Radiological Protection Journal of Radiological Protection
J. Radiol. Prot. **40** (2020) 367–380 (14pp) https://doi.org/10.1088/1361-6498/ab6fd2

Variations in radon dosimetry under different assessment approaches in the Altamira Cave

Carlos Sainz[1,2], Daniel Rábago[1,4], Enrique Fernández[1], Jorge Quindós[1], Luis Quindós[1], Alicia Fernández[1], Ismael Fuente[1], Jose Luis Arteche[3], Luis Santiago Quindós[1,2] and Santiago Celaya[1,2]

[1] Radon Group, University of Cantabria, C/Cardenal Herrera Oria s/n 39011, Santander, Spain
[2] The Cantabrian International Institute for Prehistoric Research (IIIPC), Spain
[3] Spanish Meteorological Agency, AEMET, Spain

E-mail: daniel.rabago@unican.es

Received 10 December 2019, revised 20 January 2020
Accepted for publication 24 January 2020
Published 28 February 2020

Abstract
The atmosphere of caves is a special environment where it is necessary to take into account some particular characteristics when assessing the radon dose. The equilibrium factor (F) between radon and its progeny, and especially its unattached fraction (f_p), is a key parameter in radon dose evaluation. In order to consider the specific features of the atmosphere in the Altamira Cave, the radon and particle concentrations have been measured. The mean annual radon concentration inside the cave over the period 2013–2019 is around 3500 Bq m^{-3} with a standard deviation of 1833 Bq m^{-3} and this exhibits seasonal variations. This value surpasses all international (WHO, IAEA, ICRP) upper action and reference levels (occupational and non-occupational). Dose rate levels expressed in μSv h^{-1} were estimated for four different equilibrium scenarios between radon and its progeny ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po. The most recent dose conversion factors have been used and the contribution made to the dose by the unattached fraction of radon progeny f_p has been also assessed from the particle concentration. The results suggest that the mean annual dose levels show variations of up to 500% due to the range of F and the f_p considered in this study. Given the high radon concentrations usually found in show caves, the best way to reduce this variability and its

[4] Author to whom any correspondence should be addressed.

associated uncertainty in dose assessment is to conduct specific studies aimed at determining both F and f_p.

Keywords: radon dose, progeny, dose conversion factor, cave, Altamira

(Some figures may appear in colour only in the online journal)

1. Introduction

Radon gas (^{222}Rn) comes from radioactive decay of the radium (^{226}Ra) present in varying amounts in almost all the materials of the Earth's crust (Hofmann *et al* 2012, Baskaran 2016). Its gaseous nature allows it to diffuse, whether carried by air inside the soil or by water flowing in underground aquifers, from the original source to the atmosphere, becoming a common component of the air in all the land regions of the planet (ICRP 1993, World Health Organization 2009). Concentrations in air are found to range over several orders of magnitude, with minimum values in the outside air but increasing with the rate of production and level of insulation in restricted and enclosed places (Nazaroff and Nero 1988). Radon is transformed by radioactive alpha decay into solid progeny with short half-lives, of the order of minutes and less, for example ^{214}Po with a half-life of 160 μs. The behaviour of these progeny is complex: on the one hand they can be deposited on macroscopic surfaces in the environment but on the other they may remain in the air. Of the fraction of progeny that are not so deposited, and remain in the air, part is attached to aerosols and is called the attached fraction, with a wide dimensional spectrum from a few nanometres to more than a micron (Reineking and Porstendörfer 1988, Butterweck-Dempewolf *et al* 1997); on the other hand radon progeny may also be found that are not attached to aerosols, the so-called unattached fraction (f_p).

When a mixture of radon gas and its progeny is breathed in, the fact that radon is a noble gas means that only an insignificant fraction is retained in the respiratory tract. However, the solid nature of the progeny means that they remain fixed in different regions of the respiratory tract, depending on their size (Reineking and Porstendörfer 1988). Bearing in mind the condition of alpha emitters of some of these progeny, it can be clearly understood that it is the progeny and not the radon that are primarily responsible for irradiating the lungs (ICRP 2017). For this reason, in order to make an appropriate estimate of the dose received by inhalation of radon and its progeny, it is essential to know both the equilibrium factor F, defined as the ratio between the equivalent equilibrium concentration (EEC) of radon, assessed from the concentrations of the radon progeny and of radon gas C and the unattached fraction of progeny f_p.

Continued inhalation of air with radon in dwellings and workplaces poses a scientifically proven risk to health, which depends mainly on the levels and duration of exposure. In 1988 the International Agency for Research on Cancer (IARC), included radon in the main category of carcinogens (Class I) as a human carcinogen with extensive scientific backing (IARC 1988). More recently, the World Health Organization (WHO), with the cooperation of more than a hundred scientists and experts on radon, created an international project that resulted in the publication of the *WHO Handbook on Indoor Radon: a Public Health Perspective* (World Health Organization 2009), which brings together the main problematic aspects of radon globally. The section on health effects states that exposure to radon is the second main cause of lung cancer, after smoking, in the most developed countries. More recent studies confirm the link between the exposure to radon and risk of lung cancer worldwide (Gaskin *et al* 2018).

The first publication of the International Commission on Radiological Protection (ICRP) given over exclusively to protection from radon exposure was ICRP 65, published in 1993 (ICRP 1993). A distinction was established at that time between dwellings and workplaces as sites of exposure, with clearly differentiable characteristics. This classification was maintained in successive publications about radon, until the current version, ICRP 137 (ICRP 2017), which describes a wider range of exposure scenarios. Since 1993 the concepts of dose conversion factor (DCF) or dose coefficient have been considered necessary to convert the levels of exposure to radon and its progeny [expressed in working level months (WLMs) or Bq m^{-3} h] into equivalent dose values (expressed in mSv), and therefore into the risk of lung cancer resulting from this exposure. These factors can be estimated in two independent ways. One is the so-called dosimetric approach, based on biokinetic and dosimetric models of the respiratory tract after inhalation of radon progeny (Marsh *et al* 2017), which is recommended by the ICRP for all radioactive elements that give rise to internal irradiation, except for radon. For the case of radon, and until the end of 2017, the ICRP recommended the epidemiological approximation based on the 'dose conversion conventions' which, in simplified form, means a comparative analysis between the risk of lung cancer in populations exposed to radon and the overall cancer risk in survivors of the atomic bombs, exposed largely to gamma radiation. Discrepancies between the factors given by the two approximations have been greatly reduced by the new revised risk coefficients set out in ICRP 115 in 2010 (ICRP 2010, Vaillant and Bataille 2012). The ICRP publication of 2010 presented an update of the analysis of the epidemiological studies into risks of lung cancer from exposure to radon and its progeny in the population as a whole. The main result was to evaluate the risk factor per unit of exposure in 5×10^{-4} WLM^{-1}, which is nearly double the risk estimated in 1993. As a consequence of the above, the recommended DCF was changed to 12 mSv WLM^{-1} for workers (Cigna 2004, ICRP 2010). For this reason, publication 137 of the ICRP recommends, for the first time in its history, the use of dose coefficients derived from the dosimetric approximation, including radon and its progeny, in the set of radioactive elements deposited inside the organism.

It is well known that radon exposure in caves visited by tourists is a special scenario because of the characteristics of the air inside a cave (Gillmore *et al* 2000). Normally, the mean particle concentration (Z) of the atmosphere inside a cave is much less than in the air outside, which leads to unattached fractions of progeny f_p that are higher than those found in other workplaces or in dwellings (Sainz *et al* 2007). Furthermore, the surface/volume ratio is usually rather low, which decreases deposition on surfaces, the main mechanism by which progeny in the air are reduced, and thus increasing the equilibrium factor F. Although the relation between F and f_p is usually an inverse (Vargas *et al* 2000), the DCF derived from this latter may be very high because of the high dosimetric charge of the unattached radon progeny, ranging from 1 to 10 nm in size (Porstendörfer 2002).

The Altamira Cave is world renowned for the quantity, diversity and quality of its cave paintings, which led to its designation as a UNESCO World Heritage Site in 1985 (http://whc.unesco.org/en/list/310). The concentration of radon in the interior exhibits the seasonal behaviour typical of surface caves (Sainz *et al* 2018, Wang *et al* 2019), related to exchange of masses of air with the outside, with maximum values in winter that may reach 8000 Bq m^{-3} and minima in the summer of around 400 Bq m^{-3}. The conservation of its cave art makes the use of forced ventilation systems to reduce these concentrations quite unfeasible, as this would seriously affect the conditions of humidity and temperature in which the paintings are preserved.

All these specific characteristics of Altamira, and the general features of most caves visited by tourists, make it necessary to develop methods for assessing personal dosimetry, to regulate exposure time as efficiently as possible. This paper sets out different methodologies for dose assessment to estimate dose received inside the Altamira Cave using yearly and monthly time scales.

2. Materials and methods

2.1. Description of the cave and sampling points

The Altamira Cave is in the higher section of limestone hills near Santillana del Mar, on the western coast of Cantabria (northern Spain), at 4° 7′ 11″ W, 43° 22′ 37″ N ($X = 409\,289$, $Y = 4803\,279$; UTM 30N, ETRS89). It is a shallow cave with a north-facing entrance at an elevation of 152 m above sea level, with S-shaped passages and a total length of 270 m, and the lowest point with respect to the entrance at 16 m (Elez *et al* 2013).

2.2. Radon gas concentration measurements

The radon concentration in the Altamira Cave was measured in two ways. First, radon was measured at hourly intervals by continuous radon monitors in the Hall, Polychromes Room and the Well Room. Second, CR-39 solid state track etched detectors were also used, giving integrated values for radon concentration over time, in the previously mentioned rooms and also in the Polychromes Corridor, the Walls Room and the Grave Room, as shown in figure 1. The locations of the CR-39 detectors in the rooms were at a height above the ground of 1 m. CR-39 detectors and radon monitors placed in the same room were close together in order to compare results and identify any operational problems. The radon monitors and etched track detectors were replaced every 2 weeks.

The radon monitor used for continuous measurement is the Radon Scout (SARAD GmbH). This monitor operates on the basis of a high-voltage chamber and a silicon detector, into which the radon diffuses. The concentration of radon is found from the energy left by the ^{218}Po alpha particles, which adhere to the surface of the detector after radon decay due to the electric field generated in the chamber. The sensitivity of the detector is 1.8 cpm at 1 kBq m^{-3}. The range of measurement is up to 10 MBq m^{-3}. Working conditions range from −10 °C to 40 °C and from 0% to 100% non-condensing relative humidity. The statistical uncertainty associated with this device varies from 20% to 5% in the concentration range 100–3000 Bq m^{-3}.

There is an added difficulty associated with continuous measurement of radon concentration in environments with a high level of relative humidity, as in the case of this cave. Humidity may change the detection and gathering of progeny within the chamber (George 1996). In order to minimise this influence, the radon monitors are placed in highly radon-permeable plastic bags, which allow radon to pass through but block the entry of humidity to the device. Also, the monitors are replaced with others of identical characteristics every 2 weeks. Additionally some punctual measurements of thoron (^{220}Rn) concentration inside of the cave were performed with the Thoron Scout device (SARAD GmbH) under the same conditions as for the continuous radon measurements. Results showed values below the detection limit (50 Bq m^{-3} for 1 h of integration time interval), so the contribution of thoron to the radon measurements is considered to be negligible.

The etched track detectors used comprise a diffusion chamber and a sensor area made of CR-39 plastic (model RSKS, Radosys Ltd). This type of passive detector measures exposure

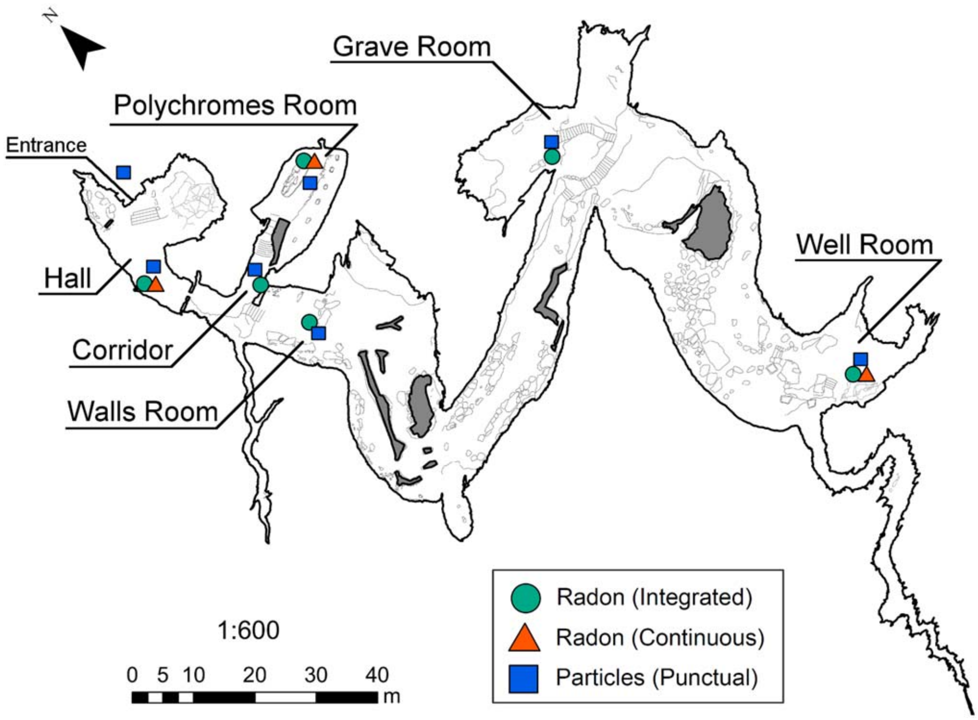

Figure 1. Floor plan of the Altamira Cave, with the sampling points for radon and particle concentration.

to radon, from which mean radon concentration can be found if the exposure time is known. Alpha particles from the radon gas diffuse into the chamber containing the detector and, by the progeny generated inside, leave marks on the detector which are etched through a chemical development process with a solution of distilled water and sodium hydroxide (NaOH) at a concentration of 6.25 mol l^{-1} and a temperature of 90 °C, for a period set by the manufacturer (but normally 4.5 h). Counting of track density (tracks/mm^2) is done automatically with a microscope. The diffusion chamber used in this model is a conducting plastic cylinder with an effective volume of 29 cm^3, which acts as a filter for radon progeny and allows only radon gas to enter. The typical sensitivity for this kind of detector is 2.0 tracks/cm^2 for an exposure of 1 kBq m^{-3} h. The uncertainty is approximately 10% for exposures in the range 500–1500 kBq m^{-3} h. The calibration factor which allows conversion from track density to exposure is given by the manufacturer; however, this factor was obtained independently in the radon chamber of the Laboratory of Environmental Radioactivity of the University of Cantabria (LaRUC).

All radon concentration measurements carried out were subject to strict quality control. The LaRUC is accredited according to UNE-EN ISO/IEC 17025:2005 for measurements of radon in air, which implies rigorous quality control of the entire measurement process, including periodic international intercomparison exercises. The radon monitors are periodically checked against a traceable international standard device in the radon chamber of LaRUC in a stable radon atmosphere. The specifications of the chamber, the calibration procedure for the radon monitors and the assessment of their response time is analysed in detail in Fuente *et al* (2018).

2.3. Measurement of particle concentration

Particle concentration was measured at various points both inside and outside the Altamira Cave over a period of 2 weeks. This parameter was measured by means of a condensation particle counter (CPC; TSI Model 3007, TSI Incorporated). Air in the device is pumped at a rate of 700 $cm^3 min^{-1}$ and passes throughout a porous wick containing liquid isopropyl alcohol. The flow enters the device through a bypass in two flows, one of 100 $cm^3 min^{-1}$, which is used for the analysis, while the rest (600 $cm^3 min^{-1}$) is ignored. This flow is independent of the form of measurement chosen for the device. After 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. This system can detect particle concentrations in the range of 0 to 100 000 particles/cm^3. The measurements carried out with this CPC give concentration in particles/cm^3 for particles with a size below 1 μm, with an uncertainty of 20%.

2.4. Dose estimate

To estimate the dose from inhalation of radon progeny it is necessary to assess exposure to radon progeny, which requires knowing the EEC, which is that activity concentration of radon in radioactive equilibrium with its short-lived decay products that has the same potential alpha energy concentration as the non-equilibrium mixture (Hofmann *et al* 2012). This magnitude, expressed in working level (WL), corresponds to the same potential alpha energy concentration as for a mixture in equilibrium with a radon concentration of 3700 Bq m^{-3}, and is related to the concentration of radon gas C (Bq m^{-3}) via the equilibrium factor F:

$$\mathrm{EEC} = \frac{CF}{3700}. \tag{1}$$

From the EEC a total value for a given time period, that is the equivalent equilibrium exposure E_{eq}, can be calculated and expressed in the classic unit WLM as:

$$E_{\mathrm{eq}} = \frac{\mathrm{EEC}\ \Delta t}{170} \tag{2}$$

where Δt (h) is the exposure period considered and the value 170 corresponds to the number of working hours per month considered in the original definition of WLM.

The effective dose H (mSv) received over the exposure period can be calculated via the following expression:

$$H = E_{\mathrm{eq}}\mathrm{DCF} \tag{3}$$

where DCF (mSv WLM^{-1}) is the dose conversion coefficient or DCF. Combining equations (1) and (2), this expression can be rewritten as:

$$H\ (\mu\mathrm{Sv}) = 1.6 \times 10^{-3}\, C\ \mathrm{DCF}\ F\ \Delta t. \tag{4}$$

Evaluating H from the mean concentration values C of radon, and assuming $\Delta t = 1$ h, gives mean dose rate values (μSv h^{-1}) for the different periods studied (monthly and yearly). The reason for using these units in the measurements of the dose rate is to facilitate, in terms based on the order of magnitude of the typical environmental dose rates, values that can be easily used to estimate the total dose received for cave entries which practically never exceed a duration of 1 h. Bearing in mind that radon concentration can be measured fairly accurately, the biggest sources of uncertainty in this evaluation are DCF and F. To properly determine the equilibrium factor, as well to measure the activity of radon gas, it is necessary to know the activity of each of the progeny, which involves expensive equipment that is not always available. It is therefore common practice to use representative values found in the literature. In the case of caves, some review studies give values of *F* between 0.2 and 0.9 (Cigna 2004), and it is common to use a value of 0.4 as recommended by ICRP 137 for caves visited by tourists (ICRP 2017). In order to more easily compare our results with other studies made previously in Altamira Cave, this study will consider, for each risk assessment approximation, the values of 0.2, 0.4, 0.7 and 0.9, to cover the range of equilibrium factors described for caves in the literature. Despite there being no experimental evidence in the case of the Altamira Cave, a value between 0.7 and 0.9 could be plausible given the low levels of ventilation in the cave, especially in the autumn–winter period (Sainz *et al* 2018).

In order to illustrate the wide variation that may be found in the assessment of the dose, depending on the conversion factor chosen, this study compares two different approximations for the risk derived from exposure to radon in the Altamira Cave, using in each case the four possible values of *F* described above.

2.4.1. Model ICRP 137. The most recent ICRP publication includes recommendations on radon and dosimetry evaluations in the dosimetric model described in the publication ICRP 66 (ICRP 1994) for different exposure scenarios. In particular, it carries out a review of different studies of radon exposure in caves, which gives information about the most relevant parameters of the dosimetric models, such as the unattached fraction of the progeny, the equilibrium factor *F*, the dimensional spectrum of the aerosols and the humidity conditions. As a consequence, the new recommended conversion coefficient for activities in caves visited by tourists is 20 mSv WLM^{-1} (ICRP 2017).

2.4.2. Dosimetric approximation based on unattached fraction (DAUF). There are many statistical applications of the dosimetric model of ICRP 66 (ICRP 1994) to a variety of situations which take into account the characteristics of atmospheric aerosols and other factors such as the rate and type of respiration or the level of physical activity (Harrison and Marsh 2012); these eventually give rise to a variety of DCFs applicable to each situation studied. In order to obtain a more detailed approximation of radon exposure in the Altamira Cave, concentrations of particles in the air were used to evaluate the unattached fraction of progeny, f_p. Taking the semi-empirical approximation obtained by Porstendörfer (2001) and used in prior work (Sainz *et al* 2007)

$$f_p = \frac{414}{Z} \tag{5}$$

where Z (cm^{-3}) is the concentration of particles in the air. Given that the atmosphere of the Altamira Cave has very a low concentration of aerosols, the use in this study of equation (5) is limited to periods in which the particle concentration is above 500 cm^{-3}. For values of Z below this, the unattached fraction is significantly higher than 1, which is not plausible in an atmosphere with as little ventilation as the inside of the Altamira Cave (Sainz *et al* 2018). The value of f_p can be used to evaluate the dose coefficient via the following expression (Porstendörfer 2002):

$$\text{DCF (mSv WLM}^{-1}) = 6.1 + 42 f_p. \tag{6}$$

This equation was obtained for indoor sites with a ventilation level below 0.5 h^{-1} and without additional aerosol sources. In this way the size of the particles is displaced towards values of 200 nm, and there is a large number of the larger particles. The mean respiration rate

Table 1. Mean and standard deviation (SD) of the mean monthly concentrations of radon for July 2013 to August 2019 in the Polychromes Room and in the Altamira Cave in general.

	Polychromes Room		Altamira Cave	
Month	Mean (Bq m^{-3})	SD (Bq m^{-3})	Mean (Bq m^{-3})	SD (Bq m^{-3})
January	5342	725	5316	618
February	5340	754	5230	527
March	5184	796	5077	659
April	4974	616	4872	626
May	4037	1065	3666	911
June	1431	483	1560	353
July	587	171	847	155
August	650	130	979	209
September	1257	418	1797	439
October	2469	749	3105	927
November	4801	1443	5015	886
December	5777	1223	5457	639

is held in this case to be $0.75\,m^3\,h^{-1}$. The ventilation rate found for the Altamira Cave satisfies this requirement, as can be seen in Sainz *et al* (2018).

3. Results

3.1. Radon and particle concentration measurements

The dose estimations were carried out using a database of radon concentration measurements obtained by our research group from July 2013 to August 2019. This database comprises integrated monthly values in each of the rooms within the Altamira Cave, and continuous mean hourly values in the Hall, the Polychromes Room and the Well Room. The dose was evaluated using the cave as a whole. This was done by considering the mean value of radon and particle concentration in each of the rooms, such as the Polychromes Room which contains most of the wall paintings in the cave and is the centre of special attention both for tourists visiting the cave and for those entering to perform conservation or research work.

Table 1 shows a summary of the mean monthly values for each room over the study period. It also includes the mean monthly value obtained from the average of all the rooms, considered to be representative of the interior of the Altamira Cave. These average values of the radon concentration have been evaluated from the measurements offered by the continuous monitors in the Hall, Polychromes and Well rooms, and by means of the CR-39 detectors in the rest of the rooms.

The radon concentration in the various areas within the Altamira Cave in figure 2 exhibits seasonal variations related to the exchange of air in the cave with that outside, mainly because of the air density gradients between the inside and the outside (Sainz *et al* 2018). In the case of Altamira, the radon concentrations show minimum values during the summer, when the inside air temperature is significantly lower than outside, which produces degasifying through the porous system of cracks and fissures in the karst. On the other hand, in winter, the karstic system loses most of its permeability to gas exchange with the outside, which produces an accumulation of gases within, such as CO_2 and radon. For this reason, the maximum radon concentration values in all the rooms are found during the winter months.

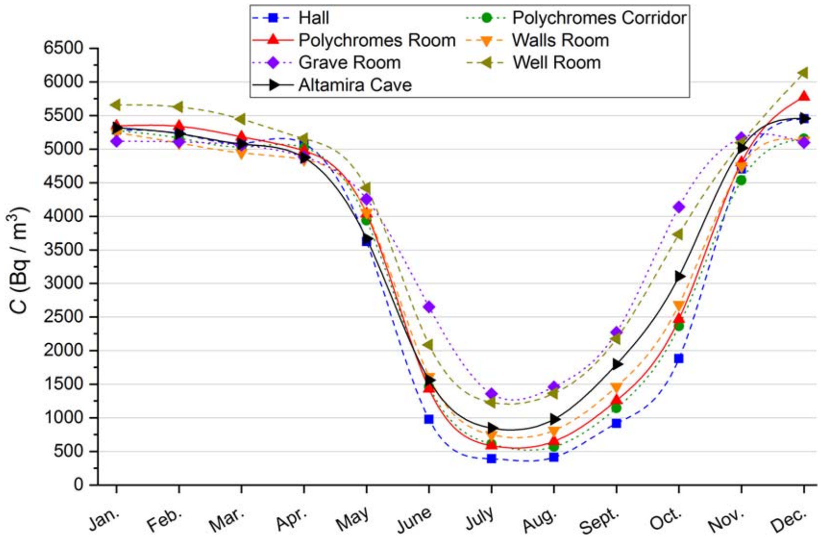

Figure 2. Mean monthly radon concentration in each of the rooms in the Altamira Cave using data from the period 2013–2019. It includes the average of all the rooms (black line).

Table 2. Mean and standard deviation (SD) of the average monthly particle concentration for the period August 2013–August 2019 in the exterior, in the Polychromes Room and in the cave, considering all the rooms monitored. The unattached fraction of radon progeny is calculated from equation (5).

	Exterior			Polychromes Room			Altamira Cave		
Month	Mean (cm^{-3})	SD (cm^{-3})	f_p	Mean (cm^{-3})	SD (cm^{-3})	f_p	Mean (cm^{-3})	SD (cm^{-3})	f_p
January	4435	2142	0.09	67	29		136	60	
February	2648	1756	0.16	94	62		146	73	
March	1968	1357	0.21	59	32		80	40	
April	3929	1985	0.11	143	151		169	205	
May	7013	4826	0.06	209	175		293	325	
June	5893	6223	0.07	724	230	0.57	807	245	0.51
July	4218	3571	0.10	806	310	0.51	833	371	0.50
August	3904	1621	0.11	1081	435	0.38	956	391	0.43
September	7313	6310	0.06	915	237	0.45	802	262	0.52
October	5003	2936	0.08	503	221		430	155	
November	4169	1684	0.10	150	136		190	155	
December	5260	1845	0.08	48	15		99	93	

On the other hand, to evaluate the dose using the dosimetry model based on the unattached fraction (DAUF) the particle concentration measurements shown in table 2 were used.

The particle concentration values in the various rooms within the cave show inverse evolution over time with respect to those observed for the concentration of radon gas, as shown in figure 3. In general, the maximum values are reached in summer, when there is a greater exchange of gas with the outside. On the other hand, in winter the concentration of

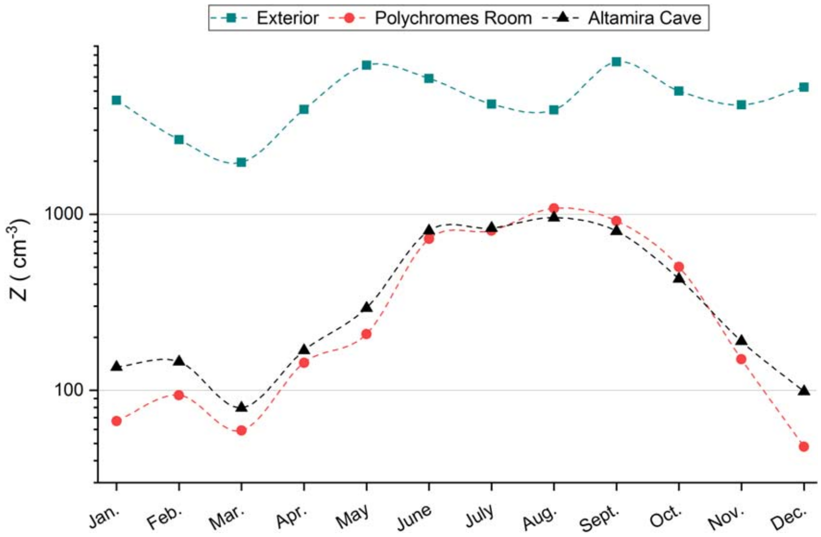

Figure 3. Mean monthly particle concentration on the outside (Exterior, outdoor air), in the Polychromes Room and in the Altamira Cave given as an average of all the rooms.

particles in suspension reaches a minimum. This behaviour is easy to understand bearing in mind that the production of particles inside the Altamira Cave is very low, and the concentrations in the air inside are always lower than in the outside air. Thus, the variations in Z in each room are a complementary indicator, together with the concentration of radon or CO_2, of the level of connection with the outside (Sainz *et al* 2018).

3.2. Dose assessment

Entrance to the Altamira Cave is carefully restricted, both to the public in general and to researchers involved in preventive conservation work on the paintings. Since 2013 guided visits by the public have been allowed on Fridays, with one or two guides passing with five visitors. During these visits, no one stays for more than 15 min in any room, so the doses received by the public are minimal. On the other hand, visits by researchers may be longer, and primarily involve the Polychromes Room, where most of the conservation effort is focused. These visits, related to sampling, recording of environmental data, state of conservation or repair or replacement of measuring instruments, are always limited in time, by the variations of parameters like air temperature and CO_2 concentration in the Polychromes Room.

Dose estimates shown in figure 4 have therefore been carried out for exposure periods of $\Delta t = 1$ h. The levels thus evaluated may serve as practical guidance for personal doses received by tourist guides, visitors or researchers.

Although the pattern of change over time in radon concentration is similar in all the rooms, the monthly values differ depending on the level of exchange of each room with those adjoining and, most importantly, the degree of connection of each room with the outside. Table 3 shows the mean monthly values of the dose rate level calculated according to each of the two approximations described. It also shows the average monthly value for the cave as a whole and the range of variation.

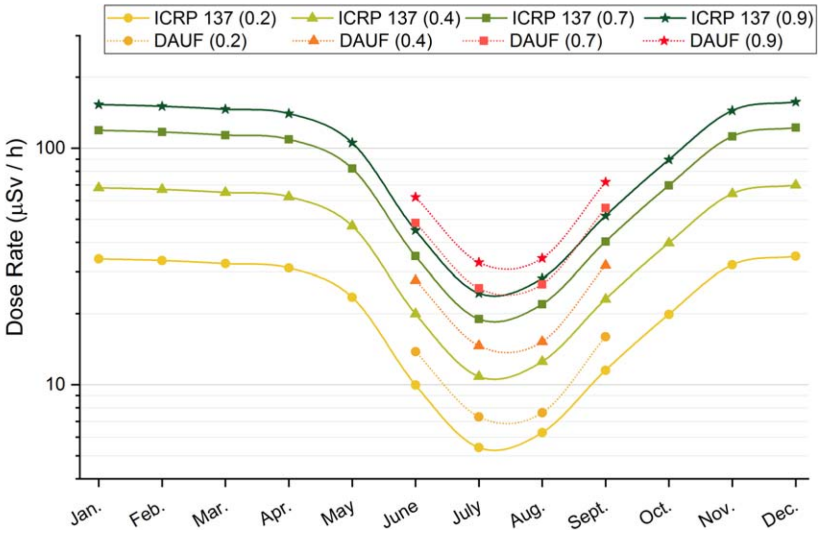

Figure 4. Dose rate expressed in μSv h^{-1}, taking into account the DCF of each approximations, 20 mSv WLM^{-1} for the ICRP 137 and 6.1 + 42 f_p mSv WLM^{-1} for the DAUF, with equilibrium factors of 0.2, 0.4, 0.7 and 0.9.

Table 3. Dose rate expressed in μSv h^{-1} obtained from equation (4) taking into account the DCFs for workers in the tourist caves from the two approaches, considering the values of radon concentration and particle concentration in the Altamira Cave.

	ICRP 137				DAUF			
	DCF = 20 mSv WLM^{-1}				DCF = 6.1 + 42 f_p mSv WLM^{-1}			
	$F = 0.2$	$F = 0.4$	$F = 0.7$	$F = 0.9$	$F = 0.2$	$F = 0.4$	$F = 0.7$	$F = 0.9$
January	34	68	119	153				
February	33	67	117	151				
March	32	65	114	146				
April	31	62	109	140				
May	23	47	82	106				
June	10	20	35	45	14	28	48	62
July	5	11	19	24	7	15	26	33
August	6	13	22	28	8	15	27	34
September	12	23	40	52	16	32	56	72
October	20	40	70	89				
November	32	64	112	144				
December	35	70	122	157				
Mean	23	46	80	103				

Obviously, the different approximations based on ICRP 137 have a constant factor, corresponding to the ratios of the equilibrium factors used. There is also another variable factor due to the different particle concentrations recorded each month. These differences reach a maximum during autumn and winter, between October and March, and a minimum during the summer. This is explained by the fact that the minimum values of Z recorded during the period of least exchange with the outside (autumn/winter) lead to an atmosphere with a higher unattached fraction, in agreement with equation (5), and thus the DCF given by equation (6) increases significantly with respect to that found in the summer, when Z is maximum and f_p is minimum.

These results can be compared, bearing in mind the natural variations of the average monthly radon concentration due to different factors, with those obtained in previous studies conducted in the Altamira Cave. In Lario *et al* (2005), for a time of stay of the tourist guides of 349 h during a 1-year period between 1997 and 1998, the total effective dose assuming an F value of 0.7, a DCF of 10 mSv WLM^{-1} and taking the annual average radon concentration as 13.9 mSv, would correspond to 27.8 mSv if the DCF of the ICRP 137 used in this work had been used. In our study, using the same conditions as those mentioned, we would obtain a dose rate of 80 μSv h^{-1} using the DCF of ICRP 137, and 39 μSv h^{-1} if the DAUF approach is used. The total dose for a 349 h stay according to both approaches would be 27.6 mSv and 13.5 mSv, respectively. It is necessary to take into account the low significance of the calculation based on the annual average of the DAUF approach, which has been done on the basis of only 4 months, during which the average dose rates provided by this model have always exceeded those obtained with the approximation of ICRP 137.

4. Conclusions

This study provides new estimates for the radon and radon progeny dose received in the Altamira Cave. The effective doses associated with inhaling radon and its progeny have increased significantly since the first DCFs were proposed in 1993. The data used come from time series of continuous measurements made over a 6-year recording period. Given the regularity of seasonal and annual behaviour of radon concentration within the cave, these estimates may be useful for future dosimetry assessments, bearing in mind the interannual variation typical of the outside weather.

As in many other tourist caves, radon concentration experiences seasonal variations, which in the case of Altamira can differ by almost a factor of 10 between the maximum and minimum values within an annual cycle. For this reason, continuous short-term measurements are essential for an accurate assessment of the dose rates received at each period of the year, allowing a more efficient implementation of the exposure management system than that based only on annual average values.

Despite the good agreement with previous works, obtained using annual average dose rates, this work provides a greater level of detail about the monthly variations in the dose rate due to the inhalation of radon and its progeny in the Altamira Cave, which will be very useful for the individual evaluation of the doses received by visitors, researchers and tour guides at different times of the year.

The dose level for radon exposure inside the Altamira Cave was assessed, taking into account the main factors whose variation affects this assessment. Although the minimum values, which could be easily derived from the baseline approximation of ICRP 65, are those currently used in Spanish law (BOE 2001), the newest factors recommended in ICRP 137 provide guidance in more accurately interpreting, with respect to current knowledge, the scale of the doses that may be received in environments like the one studied. However, another part of Spanish law (BOE 2012) refers to an annual radon concentration average of 600 Bq m^{-3} which is exceeded six-fold in this cave.

On the other hand, the estimates given here, derived from the approximation based on the relationship between particle concentration and unattached fraction (DAUF), are intended as a first step towards more detailed future research into dosimetry addressing the specific conditions of radon exposure in the Altamira Cave. This approximation shows that the high concentrations of radon inside the Altamira Cave, together with the lower concentration of aerosols in suspension in the interior atmosphere, may lead to significantly higher doses than those received in other environments with similar radon levels.

In this study, we have shown that the effective dose rate due to the inhalation of radon progeny depends strongly on the conditions of the atmosphere in which the exposure occurs. The high concentrations of radon usually present inside tourist caves make it necessary to carry out specific studies aimed at determining both the equilibrium factor and the concentration of particles in the air, in order to reduce the uncertainty in the evaluation of the received dose.

Acknowledgments

The authors are grateful to the Altamira Cave Research Centre and Museum staff for their helpful support during the sampling surveys in the cave. We also acknowledge the essential collaboration of the Spanish Meteorological Agency (AEMET).

This work was supported by the Ministry of Education, Culture and Sport of Spain (MECD) (grant number J180052) under the Project 'Servicio de control y seguimiento de las condiciones ambientales y del biodeterioro de la Cueva de Altamira', and The Cantabrian International Institute for Prehistoric Research (IIIPC).

ORCID iDs

Daniel Rábago https://orcid.org/0000-0002-2285-8767
Ismael Fuente https://orcid.org/0000-0001-7757-8511

References

Baskaran M 2016 *Radon: a Tracer for Geological, Geophysical and Geochemical Studies* (Basel: Springer) (https://doi.org/10.1007/978-3-319-21329-3)

BOE 2001 Real Decreto 783/2001, de 6 de Julio, por el que se aprueba el Reglamento sobre protección sanitaria contra radiaciones ionizantes *Boletín Oficial del Estado (BOE)* **178** 27284–393

BOE 2012 Instrucción IS-33, de 21 de Diciembre de 2011, del Consejo de Seguridad Nuclear, sobre criterios radiológicos para la protección frente a la exposición a la radiación natural *Boletín Oficial del Estado (BOE)* **22** 6833–8

Butterweck-Dempewolf G, Shuler C and Vezzu G 1997 Size distribution of the unattached fraction of radon progeny *European Conf. on Protection against Radon at Home and at Work (Prague, Czech Republic)*

Cigna A A 2004 The distribution of radon concentration in caves *Int. J. Speleol.* **32** 8

Elez J, Cuezva S, Fernandez-Cortes A, Garcia-Antón E, Benavente D, Cañaveras J C and Sánchez-Moral S 2013 A GIS-based methodology to quantitatively define an adjacent protected area in a shallow karst cavity: the case of Altamira cave *J. Environ. Manage.* **118** 122–34

Fuente M, Rabago D, Herrera S, Quindos L, Fuente I, Foley M and Sainz C 2018 Performance of radon monitors in a purpose-built radon chamber *J. Radiol. Prot.* **38** 1111

Gaskin J, Coyle D, Whyte J and Krewksi D 2018 Global estimate of lung cancer mortality attributable to residential radon *Environ. Health Perspect.* **126** 057009

Gillmore G K, Sperrin M, Phillips P and Denman A 2000 Radon hazards, geology, and exposure of cave users: a case study and some theoretical perspectives *Ecotoxicol. Environ. Safety* **46** 279–88

George A C 1996 State-of-the-art instruments for measuring radon/thoron and their progeny in dwellings-a review *Health Phys.* **70** 451–63

Harrison J D and Marsh J W 2012 Effective dose from inhaled radon and its progeny *Ann. ICRP* **41** 378–88

Hofmann W, Arvela H S, Harley N H, Marsh J W, McLaughlin J, Röttger A and Tokonami S 2012 Characteristics and behavior of radon and radon progeny *J. ICRU* **12** 55–70

IARC 1988 *Man-made Mineral Fibres and Radon* (*IARC monographs on the evaluation of carcinogenic risks to humans* 43) (Lyon: IARC)

ICRP 1993 Protection against radon-222 at home and at work. ICRP publication 65 *Ann. ICRP* **23** 15–22

ICRP 1994 Human respiratory tract model for radiological protection. ICRP publication 66 *Ann. ICRP* **24** 106–15

ICRP 2010 Lung cancer risk from radon and progeny and statement on radon. ICRP publication 115 *Ann. ICRP* **40** 27–50

ICRP 2017 Occupational intakes of radionuclides: Part 3. ICRP Publication 137 *Ann. ICRP* **46** 297–317; 447–80

Lario J, Sánchez-Moral S, Cañaveras J C, Cuezva S and Soler V 2005 Radon continuous monitoring in Altamira Cave (northern Spain) to assess user's annual effective dose *J. Environ. Radioact.* **80** 161–74

Marsh J W, Laurier D and Tirmarche M 2017 Radon dosimetry for workers: ICRP's approach *Radiat. Prot. Dosim.* **177** 466–74

Nazaroff W W and Nero A V Jr 1988 *Radon and its Decay Products in Indoor Air* (New York, NY: Wiley)

Porstendörfer J 2001 Physical parameters and dose factors of the radon and thoron decay products *Radiat. Prot. Dosim.* **94** 365–73

Porstendörfer J 2002 Influence of physical parameters on doses from radon exposures *Int. Congress Series* 1225 (Amsterdam: Elsevier) pp 149–60

Reineking A and Porstendörfer J 1988 Activity size distributions of the shortlived radon decay products and their influence on the deposition probability in the human lung *J. Aerosol Sci.* **19** 1331–7

Sainz C, Quindós L S, Fuente I, Nicolás J and Quindós L 2007 Analysis of the main factors affecting the evaluation of the radon dose in workplaces: the case of tourist caves *J. Hazard. Mater.* **145** 368–71

Sainz C, Rábago D, Celaya S, Fernández E, Quindós J, Quindós L, Fernández A, Fuente I, Arteche J L and Quindós L S 2018 Continuous monitoring of radon gas as a tool to understand air dynamics in the cave of Altamira (Cantabria, Spain) *Sci. Total Environ.* **624** 416–23

Vaillant L and Bataille C 2012 Management of radon: a review of ICRP recommendations *J. Radiol. Prot.* **32** R1

Vargas A, Ortega X and Porta M 2000 Dose conversion factor for radon concentration in indoor environments using a new equation for the *F*–*f*p correlation *Health Phys.* **78** 80–5

Wang Y *et al* 2019 High 222Rn concentrations and dynamics in Shawan Cave, southwest China *J. Environ. Radioact.* **199** 16–24

World Health Organization 2009 *WHO Handbook on Indoor Radon: a Public Health Perspective* (Geneva: World Health Organization) https://apps.who.int/iris/handle/10665/44149