High radon levels in subterranean environments: monitoring and technical criteria to ensure human safety (case of Castañar cave, Spain)

Miriam Alvarez-Gallego^{*1}, Elena Garcia-Anton¹, Angel Fernandez-Cortes^{1,2,3}, Soledad Cuezva^{1,2}, Sergio Sanchez-Moral¹

¹Department of Geology, National Museum of Natural Sciences (MNCN-CSIC), 28006 Madrid, Spain

²Geomnina Natural Resources SLNE, 28003 Madrid, Spain

³Department of Earth Sciences, Royal Holloway, University of London, Egham, Surrey TW20 0EX, UK

*Corresponding author: (mag@mncn.csic.es)

ABSTRACT

Castañar cave contains the highest radon gas (^{222}Rn) concentration in Spain with an annual average of $31.9 \text{ kBq}\cdot\text{m}^{-3}$. Seasonal variations with summer minimums and maximum values in fall were recorded. The reduction of air-filled porosity of soil and rock by condensation or rainfalls hides the radon exchange by gas diffusion, determining this seasonal stair-step pattern of the radon activity concentration in underground air. The effective total dose and the maximum hours permitted have been evaluated for the guides and public safety with a highly detailed radon measurement along 2011 and 2012. A network of 12 passive detectors (kodalphas) has been installed, as well as, two radon continuous monitoring in the most interesting geological sites of the subterranean environment.

A follow up of the recommended time (max. 50 minutes) inside the underground environment has been analysed since the reopen to public visitors for not surpassing the legal maximum effective dose for tourists and guides. Results shown that public visitors would receive in fall a 12.1% of the total effective dose permitted per visit, whereas in summer it is reduced to 8.6%, while the cave guide received a total effective dose of 6.41 mSv in four months.

The spatial radon maps allow defining the most suitable touristic paths according to the radon concentration distribution and therefore, appropriate fall and summer touristic paths are recommended.

KEY WORDS

Radon exposures, risk assessment, prevention, subterranean workplaces, effective dose, cave monitoring.

1. INTRODUCTION

The confined environments, such as mines, caves, tunnels or catacombs, generally can accumulate radon gas (^{222}Rn) to reach harmful concentrations due to low ventilation (Gunn, 2003). Radon originates from the decay of radium (^{226}Ra) by alpha-emission in the natural chain of uranium (^{238}U) and it is exhaled from the rock or soil and accumulated in these poor ventilated sites at subsurface. Several comprehensive reviews have provided extensive regional and worldwide data set of ^{222}Rn measurements in underground air based on a statistically-representative number of caves, exhibiting a wide range of mean radon concentrations; $0.3\text{-}10.6 \text{ kBq}\cdot\text{m}^{-3}$ (Halk et al., 1992), $4.5\text{-}8.9 \text{ kBq}\cdot\text{m}^{-3}$ (Hyland and Gunn, 1994), $2.8 \text{ kBq}\cdot\text{m}^{-3}$ (Halk et al., 1995, 1997), $0.5\text{-}0.8 \text{ kBq}\cdot\text{m}^{-3}$ (Solomon et al., 1996), $2.5 \text{ kBq}\cdot\text{m}^{-3}$ (Cigna, 2005) or, recently, $0.5\text{-}8.3 \text{ kBq}\cdot\text{m}^{-3}$ (Somlai et al., 2011). The highest measured concentration in

a natural cave seems to be the 155 kBq·m⁻³ recorded by Gunn et al. (1991) at Giants Hole in Derbyshire, England. But some other examples of extreme ²²²Rn gas concentrations have been reported, including; 88 kBq·m⁻³ recorded in a show cave from northern Greece (Papastefanou et al., 2003), 84 kBq·m⁻³ in Hollow Ridge Cave, a wild cave from Florida (Kowalczyk and Froelich, 2010), or Castañar cave with 50.5 kBq·m⁻³ (Lario et al., 2006) being the purpose of this study.

Focusing in subsurface workplaces with high concentrations of ²²²Rn, such as those mentioned above, increases the lung cancer health hazard due to the alpha radiation dose delivered by the radon decay products that deposited in the respiratory tract (ICRP, 1987, 2010; UNSCEAR, 2006; Craven and Smit, 2006). Therefore, tourists, cavers and tour guides of this kind of subterranean sites are exposed to a wide range ionizing radiation from ²²²Rn gas and its progeny (Field, 2007; Kavasi et al., 2010). In the case of touristic subsurface sites, an environmental radon monitoring programme is required to make decisions on needed action for radiation protection of workers (guides) and tourists (recreational and professional cavers) from natural radon (IAEA, 2003). The monitoring approaches and activities on radon management strategy must be discussed for each environmental situation on a case-by-case basis and in accordance to the legal constraints regarding the air quality at the workplaces, in terms of the radon reference levels by effective dose criteria of Sv·y⁻¹ as well as by radon concentration in air of Bq·m⁻³.

The most recent European directive (Council of the European Union, 2014) has updated the basic safety standards for protection against the dangers arising from exposure to ionising radiation, in accordance to the recommendations of the International Commission on Radiological Protection (ICRP, 2007; Wrixon, 2008). Previously, in its Publication 65, the ICRP (1994) recommended an action level of 500–1500 Bq·m⁻³ for the annual average radon concentration at workplaces. Now, the European Union (EU) establishes that the reference levels for the annual average activity concentration in air shall not be higher than 300 Bq·m⁻³, and the current recommendations for occupational (workers) and public effective dose limits are 20 and 1 mSv in a year, respectively. This new legal framework compels the Member States, through transposing to its national regulation, to promote action to identify sites with radon concentrations (as an annual average) exceeding these reference level, as well as to perform technical (preventive or corrective) measures to reduce the existing radon concentrations or the exposure to ionizing radiation.

In this context, an extensive set of subterranean environments open to tourism have been documented concerning the effective dose from radon inhalation estimation, including show caves (Solomon et al., Vaupotič et al., 2001; 1996; Gillmore et al., 2002; Papachristodoulou et al., 2004; Lario et al., 2005; Sainz et al., 2007; Landridge et al., 2010), catacombs (Quarto et al., 2014), touristic mines (Przylibski, 2001; Gratan et al., 2004; Anjos et al., 2010) or touristic underground routes (Przylibski, 2000 and 2008), among many other examples. Mitigation actions were adopted in some subterranean workplaces in order to reduce the health risk of tourist guides because of the exposure to excessive concentrations of radon or its decay products, such as the introduction of forced renewal of underground air through equipping the routes with periodically activated mechanical ventilation (Gunn et al., 1991; Fijalkowska-Lichwa, 2014).

However, high ²²²Rn gas concentrations in caves cannot easily be remediated by the above procedures because of the sensitive nature of these environments, thus air ventilation in caves is regarded as unthinkable because of the likely deleterious effects on its microclimate. The control of exposure to ionising radiation through changes to working patterns should be the most suitable options, e.g., by restricting the amount of time spent in the subterranean site and limiting the accessibility to areas with dangerous radon activity of underground air, beside

subjecting cave workers to an appropriate system of radiation protection. Restrictions regarding accessibility to underground sites were suggested in some cases, including the limitation of the tourist route (Przylibski, 2001) or the reduction of daily employee's shift (Lario et al., 2006; Anjos et al., 2010). Indeed, filtering the air by the use of throwaway masks has been demonstrated that reduce the total effective dose (Wang et al., 2011).

Here, we report the results of a systematic and comprehensive spatiotemporal monitoring of radon concentrations of air in a subterranean site (Castañar Cave, Spain). This show cave was declared Natural Monument by the Spanish environmental legislation, which carries with it an expectation of sustainable tourist use. However, its subterranean atmosphere exhibits a very high level of ^{222}Rn with an annual average of $31.9 \text{ kBq}\cdot\text{m}^{-3}$ and monthly mean values always higher than $20 \text{ kBq}\cdot\text{m}^{-3}$ (Fernandez-Cortes et al., 2011a), which is two order of magnitude higher than the safety reference levels for the annual average activity concentration established by the EU authorities for workplaces. The high micro-environmental stability throughout the annual cycle ensures the outstanding state of conservation of this subterranean site and, consequently, it precludes the adoption of interventionist measures to reduce air radon activity (e.g. forced ventilation). In this context, the aim of this work was to define a focused and manageable occupational program for this subterranean worksite, setting some preventive recommendations according to the current legal constraints concerning the exposure to ionising radiation in workplaces. We propose the recommendable exposure times, duration of tourist tour, visiting schedule and timing of tourist route by defining the radon prone areas and the spatiotemporal distribution of effective dose (spot and annual accumulated) to guides and visitors. The protocol visit to the cave designed after this study, is being applied in the cavity from the cave reopen to public visitors in June 2014. Finally, an analysis and follow up of the recommendations given are being evaluated.

2. MATERIALS AND METHODS

2.1. Study site and scientific background

Castañar cave (SW Spain, $39^{\circ}37'40''\text{N}$, $5^{\circ}24'59''\text{W}$, 590 m.a.s.l.) is hosted in mixed carbonate-metapelite metamorphic facies. The cave developed by the dissolution of the dolomite beds and the extensive weathering of the slates favours collapses, which created and enlarged the cave (Alonso-Zarza et al., 2011). Castañar cave exhibits a maze pattern with a labyrinthine distribution of galleries (Figure 1) that indicate a strong structural and lithological control on its formation. The cave has only one upper natural entrance, located at a height of 9 meters above the main access gallery to the cave interior. To reduce the air exchange between the cave and the exterior atmosphere a glass door cubicle and a cabin protects the entrance by keeping its natural isolation. The depth of the galleries with respect to the present topography ranges between 15 to 55 m and the galleries are a few meters wide and approximately 2 m high. Water is sourced by diffuse recharge through overlying insoluble rocks and bedding, fractures and cleavage planes are the main paths for water circulation. Therefore, the physical-chemical features of karstic waters are mainly controlled by the petrologic and mineralogic composition of the host rock, the hydrogeologic properties of rocks that have a strong influence on the extent of water/rock interaction (Sanchez-Moral et al., 2006) and finally, the cave air exchange with the external atmosphere (Fernandez-Cortes et al., 2009a, 2011a; Alvarez-Gallego et al., 2013).

Radon exhalation from bedrock might be a continuous source of this gas to cave atmosphere, since the slates hosting the cave volume contains $39.2 \pm 5.2 \text{ mg}\cdot\text{kg}^{-1}$ of total uranium, emitting $487.2 \pm 22.3 \text{ Bq}\cdot\text{kg}^{-1}$ (Garcia-Guinea et al., 2013). However, the weathering leakage processes of the bedrock also favours the remobilization of radionuclides via leaching and their later

settlement into the cave environment associated to mineral phases of cave deposits, in particular abundant hydrous silica-uranyl coatings covering carbonated speleothems with radionuclides of ^{238}U natural decay series (Garcia-Guinea et al., 2013). Enrichment in rare earth elements has also been observed in some other growth layers of stalactites, which confirms the intense leaching of ions in the past coming from the overlying metapelitic rocks (Fernandez-Cortes et al., 2011b). These long-lived radionuclides of the radium radioactive decay chain are responsible of the continuous regeneration of radon gas inside the cave and, consequently, the maintaining of an outstanding natural environmental radioactivity of this subterranean site and the detection of spatially anomalies of radon activity.

The high radon activity of air in Castañar cave is maintained with monthly mean values always above $20 \text{ kBq}\cdot\text{m}^{-3}$ throughout year (Fernandez-Cortes et al., 2011a) and, therefore, well above the current EU reference levels for the annual average activity concentration in air at workplaces ($300 \text{ Bq}\cdot\text{m}^{-3}$). That implies the need for specific studies with continuous monitoring of cave air as a means to detect problems of overexposure to environmental radiation and correct problems as they occur. The first study to assess the calculation of the annual effective dose received by guides showed values (27.4 mSv) higher than the maximum effective dose recommended by authorities (Lario et al., 2006). These authors proposed two management measurements to reduce these doses, including the reduction of the time of visit to a maximum of 60 min, and opening the cave door 1 hour before the entrance of the guides and visitors. These management measurements were effective, as they led to a decrease of 10–12% in ^{222}Rn in the cave atmosphere during visits and prevented the guides from being overexposed. However, the second measure was not finally implemented because the prolonged opening of cave access during visits provokes forced ventilation that also generates a pCO_2 -imbalance between cave air and seepage water. The equilibrium is re-established by CO_2 -degassing of seepage water, but a continuous and intense depletion of CO_2 level in the cave atmosphere would alter the thermodynamically equilibrium ranges for pCO_2 that determines the mineral deposition rates and the type of calcium-carbonated phases that contributes in the current speleothems growth (Sanchez-Moral et al., 2006; Fernandez-Cortes et al., 2009b; Sanchez-Moral et al., 2012). Other management guidelines for visits to Castañar cave have been recently proposed in order to prevent additional contamination of radon daughters that are attached by electrostatic attraction to clothes, skin and hair of people (Martin-Sanchez et al., 2013).

Figure 1. Castañar cave map including the spatial distribution of the 12 passive radon dosimeters (kodalphas) and two Radims giving a ^{222}Rn continuous monitoring located in Nevada chamber and Gallery entrance. In addition, includes the level curves and the cave speleothems accumulation (brown line) and the lakes/gours (light blue).

2.2. Instrumentation for radon activity monitoring of cave air

Radon gas measurements are usually preferred to radon progeny measurements because of their relative simplicity and cost effectiveness. For this study, the radon gas concentration was monitored every half hour during the biennium 2011-2012 using two Radim 5WP monitors (SMM Company, Prague, Czech Republic and distributed by GT-Analytic KG Innsbruck, Austria), which are mainly designed for radon measurements in indoor air. These monitors determine radon concentration by measuring gross α -activity of the radon decay products ^{218}Po and ^{214}Po , collected electrostatically on the surface of a Si-semiconductor. The lowest activity detectable is $80 \text{ Bq}\cdot\text{m}^{-3}$, for 1-hour measurements with a statistical error equal to $\pm 20 \%$, and the maximum is $150 \text{ kBq}\cdot\text{m}^{-3}$. The instrument response is $0.4 (\text{imp}\cdot\text{h}^{-1})/(\text{Bq}\cdot\text{m}^{-3})$. The monitors were checked regularly on field (inside cave) using a Pylon AB5 scintillator-photomultiplier with a continuous radon detector (Pylon Electronics Inc., Ottawa, Canada), as a reference instrument periodically calibrated that provide highly-accurate measurements (lowest detectable activity of $24.8 \text{ Bq}\cdot\text{m}^{-3}$, a sensitivity of $0.041 \text{ cpm} (\text{Bq}\cdot\text{m}^{-3})^{-1}$, accuracy of $\pm 4\%$ and nominal background of 0.4 cpm).

Radon continuous monitors were installed at two cave locations: Gallery entrance and Nevada chamber, 15 m and 120 m far from the cave entrance and -16 m and -37 m vertical depth from the surface, respectively (Figure 1). Monitors were placed at a height of 10 cm above cave floor. Radon concentration was every half hour registered in order to make our dose estimation more precise, inasmuch as the daily-averaged concentration used for radiation dose calculation usually differs from radon levels during working hours.

Spatial distribution of radon activity of cave air was determined by Kodalpha nuclear track-etch passive detectors (GT-Analytic Innsbruck, Austria) manufactured by Dosirad (Pierrelatte, France). The ^{222}Rn -sensitive part is a solid state LR-115 type film (Kodak) built of a $100 \mu\text{m}$ thick

polyester base that is coated with a 12 μm thin film of red coloured cellulose nitrate. The sensitivity of the LR115 films is about 1.5-2.4 nuclear tracks $\cdot\text{cm}^{-2}$, for a total ^{222}Rn exposure of 1 kBq $\cdot\text{h}\cdot\text{m}^{-3}$. The saturation level of films is approximately 600 tracks $\cdot\text{mm}^{-2}$ that corresponding to a maximum exposure of 70 MBq $\cdot\text{h}\cdot\text{m}^{-3}$. The lower detection threshold is 2 kBq $\cdot\text{h}\cdot\text{m}^{-3}$.

A network of 12 points were homogeneously distributed along cave where nuclear track-etch passive detectors were installed and replaced every month from September 2011 to September 2012, therefore the obtained measurements indicate the monthly-averaged ^{222}Rn concentration. Detectors were placed at a height of 10 cm from the cave floor. The exposed detectors were processed and analysed by the own manufacture lab. Prior to analysis, the detectors were subjected to a chemical treatment (etching process) under controlled conditions (temperature and concentration and timing of the etching bath) to make the nuclear tracks visible using microscopy. For the analysis, nuclear track detectors were viewed under the microscope and transmitted light through the film was used to count tracks. The quality of radon measurement systems distributed by GT-Analytic for this study was certified by two laboratory intercomparisons, namely the Bundesamt für Strahlenschutz (Federal Agency of Radiation Protection) and the Paul Scherrer Institute in Switzerland. Castañar cave has a wide range of sensors installed designed for the narrow range of measurements expected for underground air parameters including temperature, relative humidity and carbon dioxide content. Detailed information about the technical specifications and locations of the complete monitoring system has been described by Fernandez-Cortes *et al.* (2009a and 2011b). For this study only time series of cave air temperature were used to compare to exterior meteorological conditions (temperature and relative humidity) in order to illustrate the seasonal pattern of air renewal during the monitoring period September 2011-September 2012. Cave air temperature was measured every 10 minutes using a high accuracy ($<0.02\text{ }^{\circ}\text{C}$) CalServ temperature PT100 1/10 DIN (compliance IEC 751: 1983: 1/10), with a measuring range of -50 to $165\text{ }^{\circ}\text{C}$ and repeatability $<0.01\text{ }^{\circ}\text{C}$. The meteorological station (HOBO Weather Station HWS) is located 20 m far from the Gallery entrance and includes, among other sensors, a 12-bit Smart Sensor (S-THB model from Onset Computer Corporation, Bourne, MA, USA) measured hourly the relative humidity and air temperature over a range from -40°C to 75°C and 0–100% RH, with an accuracy of $\pm 0.2^{\circ}\text{C}$ over 0 to 50°C , and $\pm 2.5\%$ from 10 to 90% RH, to a maximum of $\pm 3.5\%$ (above 95%). The resolution is 0.02°C and 0.1% RH at 25°C .

2.3. Calculation of radiation exposure

Although radon decay products, primarily ^{218}Po and ^{214}Po , are responsible for most of the radiation dose, radon gas concentration is generally considered a good surrogate for the radon progeny concentration and to calculate the radiation exposure. The radiation dose accrued by an individual is calculated as a function of exposure time in a radiation environment (potential α -energy exposure) and is expressed in milliSieverts (mSv). The effective dose (E, in mSv) is calculated using the equation:

$$E = C_{\text{Rn}} \cdot F \cdot t \cdot \text{DCF} \quad [1]$$

where C_{Rn} : average radon (Rn) concentration ($\text{Bq}\cdot\text{m}^{-3}$), F: average equilibrium factor, i.e. the ratio of the Rn progeny concentration (UNSCEAR, 2000) and to the radon gas concentration, t: time (hours) spent inside cave during the investigated period and DCF: dose conversion factor from ICRP ($\text{mSv}\cdot\text{m}^3\cdot\text{Bq}^{-1}\cdot\text{h}^{-1}$).

F factor has been widely studied in different conditions to evaluate the dose to persons exposed to a given concentration of ^{222}Rn when equilibrium conditions with its decay products are not attained (Chen and Marro, 2011). A default indoor value of 0.4 is recommended by

ICRP (1994), but a wide variation from 0.1 to 0.9 has been reported (UNSCEAR, 2009). For this study an F value of 0.57 was used based on the average of the equilibrium factor weighted over more than 880 measurements in caves (Cigna, 2005).

The potential α -energy exposure by radioactive decay of radon progeny is usually expressed in Working Level Months (WLM). One WL is defined as a concentration of the potential α -energy of $1.3 \cdot 10^5 \text{ MeV} \cdot \text{m}^{-3}$, corresponding to a potential concentration of the radon progeny in equilibrium ($F=1$) with $3700 \text{ Bq} \cdot \text{m}^{-3}$ and considering an occupational exposure of 170 h per month (ICRP, 1994). The commonly used approximation is that 5 mSv equal to 68 WLh. There are many complexities in equating these dose conversion factors, in particular the differences in aerosol conditions could modify the dose conversion factor, e.g. varying between 4.2 and $11.5 \text{ mSv} \cdot \text{WLM}^{-1}$ (Porstendörfer, 2001, 2002). The International Commission on Radiological Protection (ICRP, 1994) establishes the Dose Conversion Convention by comparing the detriment per unit exposure to ^{222}Rn and its progeny (expressed in WLM or Jh with the total detriment associated with unit effective dose (expressed in mSv). In the ICRP-65 report (ICRP, 1994) it is recommended the use of an epidemiological estimate for dose conversion factor (DCF) equal to $5.06 \text{ mSv} \cdot \text{WLM}^{-1}$ [$3.18 \text{ nSv} \cdot (\text{Bq} \cdot \text{h} \cdot \text{m}^3)^{-1}$] for workers and $3.88 \text{ mSv} \cdot \text{WLM}^{-1}$ [$2.44 \text{ nSv} \cdot (\text{Bq} \cdot \text{h} \cdot \text{m}^3)^{-1}$] for the public. These current dose conversion values are currently used, but the ICRP is planning to provide, in the near future, new dose coefficients per unit exposure to ^{222}Rn and its progeny for different reference conditions of domestic and occupational exposure, with specified equilibrium factors and aerosol characteristics (ICRP, 2010). In accordance to the explanation above, the effective dose from equation [1] in terms of Working Level Month is calculated as follows:

$$E_{\text{WLM}} = \sum (C_{\text{Rn}} \cdot F \cdot t)_m / 629000 \quad [2]$$

where, E_{WLM} : effective dose in terms of annual WLM, $(C_{\text{Rn}} \cdot F \cdot t)_m$ is the proportional WLM per month (m : 1...12 months) and the denominator comes from $3700 \text{ (Bq} \cdot \text{m}^{-3} \text{ per WL)} \cdot 170 \text{ (h per WM)}$ according to WLM definition.

3. RESULTS AND DISCUSSION

3.1. Seasonal and spatial signatures of ^{222}Rn concentration in the cave atmosphere

Castañar cave is characterized by a very stable microclimate, with a high radon content of air that remains constantly above $20 \text{ kBq} \cdot \text{m}^{-3}$ in the inner areas of the cave, e.g. Nevada chamber and other more distant areas of the cave entrance. During the studied period 2011-2012 the highest average ^{222}Rn level monitored up to $37.4 \text{ kBq} \cdot \text{m}^{-3}$ in November, reducing to its minimum value in August with $16.2 \text{ kBq} \cdot \text{m}^{-3}$ and with an annual average of $31.9 \text{ kBq} \cdot \text{m}^{-3}$ (Table 1). These ^{222}Rn contents are the highest recorded in any Spanish cave, either natural or show, and one of the highest concentration documented for any natural subterranean site in Europe. Thus, the minimum values registered in this show cave are approximately 5.7 times greater than the annual-averaged values found in a wide range of cave according to some review studies, e.g. $2.8 \text{ kBq} \cdot \text{m}^{-3}$ (Halk et al., 1997) or $2.5 \text{ kBq} \cdot \text{m}^{-3}$ (Cigna, 2005).

A stair-step pattern with strong seasonal shifts of radon activity concentration in underground air is distinguishable in both cave sites (Gallery entrance and Nevada chamber) throughout an annual cycle (Figure 2 and Table 1). Thus, the cave atmosphere undergoes several seasonal stages regarding the radon content:

- 1) Underground air renewal and depletion of radon content up to reach minimum levels during summer months. During late May and early summer the dry weather conditions

(air temperature higher than 17 °C and relative humidity below 50-60 %) are coeval to a gas exchange process between the cave and the outside atmosphere and, consequently, lowering radon levels. The seasonal decreases and setting of minimum radon levels during summer are more evident in the cave sites near surface (i.e. with less thickness of host-rock) such as the Gallery entrance (below 20 kBq·m⁻³) and, however, it is smoothed in inner and deeper areas, e.g. Nevada chamber (below 30 kBq·m⁻³).

- 2) Isolation of the cave atmosphere and a steady accumulation of radon during the colder and wetter seasons (generally, from November to May), reaching values higher than 31 kBq·m⁻³. This stage matches when the temperature outside is below the cave air temperature (almost steady around 17°C) and the external relative humidity above 60%, approximately (Figure 2).
- 3) A transition phase of gas recharge occurs between the above stages (renewal and isolation of cave atmosphere), once the first events of intense rain occur during each autumn and there is a water recharge of the vadose zone above the cave after the drier conditions of summer season.

Similar stages for radon content in underground air have been described in other caves (Sanchez-Moral et al., 1999; Kowalski et al., 2008; Cuezva et al., 2011). However, this pattern is contrary to the strong seasonal shifts in function of the thermal exterior-cave relationship, with high summer and low winter levels (Przylibski and Ciekowski, 1999b; Bourges et al., 2006; Perrier et al., 2007, among others).

Figure 2. Evolution of the radon (Bq·m⁻³) content of cave air during the biennium 2011-2012 at the Gallery entrance and Nevada chamber, compared to time series of main factors controlling the cave air renewal and gas exchange: temperature (°C) difference between cave atmosphere and exterior ($T_{\text{exterior}} - T_{\text{cave}}$) and exterior relative humidity (%).

According to Przylibski (1999a), the cave ventilation caused by atmospheric temperature changes plays a major role on the variations radon in underground air. Thus, the high radon concentrations are observed in the warm season of the year, when the average air temperature exceeds the average temperature of the cave interior, while low concentrations occur in the colder season when the temperature relationship is reverse.

In the case of Castañar cave, it is the partial water filling of the porous system and fissures of host rock and upper overlying soil which determines the ^{222}Rn gas exchange between the cave atmosphere and exterior (Fernandez-Cortes et al., 2011a). The connected porosity of soil and the microfracture network of bedrock carry diphasic infiltration (water plus air). Reduction of air-filled porosity of soil and rock by condensation or rainfalls hides the radon exchange by gas diffusion. Therefore, when the confined conditions are reached in the network of fissures and pores (especially during the rainy periods) a limited gas exchange prevails between the cave atmosphere and exterior, delaying the air gaseous transfer between both environments. On the contrary, the drier conditions provoke the partial opening of the porous system of upper soil and the network of host rock fissures and, consequently, an intense degasification process of the cave atmosphere occurs.

The occurrence of convective air movements can cause a reduction in ^{222}Rn activity concentration in the underground air at daily scale, for instance as described for Bear Cave , Poland (Fijalkowska-Lichwa and Przylibski, 2011). In the case of Castañar cave, other transient fluctuations of radon content of cave air, mainly noticeable during summer under drought conditions and at cave locations near surface (e.g. Gallery entrance), reveal that the sharp rises of relative humidity at exterior during several days reinforce the isolation effect of soil and host rock described above. Under these environmental conditions, the water deposition on soil by condensation increases due to the high environmental humidity and that seems to be correlated with short-term pulses of radon levels (Figure 2).

The cave monitoring studies during previous annual cycles have demonstrated that ^{222}Rn concentration of cave air also exhibits sharp short-term fluctuations depending on imbalances of cave air pressure with exterior (Fernandez-Cortes et al., 2009a). The prevailing conditions of barometric depression in weather outside or forced ventilation due to uncontrolled opening of cave entrance are responsible of these air pressure imbalances. Specially during summer months the opening of the cave entrance provokes a barometric imbalance, entailing large-scale dilution and evacuation processes by air volume changes and, consequently, the radon content in cave undergoes a fast decrease since the incoming air is comparatively depleted in this gas. Other examples of human-induced changes of in ^{222}Rn activity concentration in the underground air at daily scale have been recorded in an underground tourist routes located in part of disused mines (e.g. Fluorite Adit, Poland) related to the periods of activity and inactivity of mechanical ventilation system (Fijalkowska-Lichwa and Przylibski, 2011).

MONTHS 2011-2012	NEVADA CHAMBER [²²² Rn] Bq·m ⁻³	GALLERY ENTRANCE [²²² Rn] Bq·m ⁻³	AVERAGE of cave air [²²² Rn] Bq·m ⁻³
January	37300	35600	36400
February	36400	35300	35900
March	38000	36200	37100
April	36400	35400	35900
May	33900	31700	32900
June	29500	26700	28100
July	26400	18900	22600
August	24400	16200	20300
September	29000	24500	26700
October	34000	31000	32500
November	37700	37200	37500
December	37000	37800	37400
TOTAL	33300	30500	31900

Table 1. Monthly summary of radon content of cave air (Bq·m⁻³) over a mean biannual cycle by considering the continuous monitoring throughout the period 2011 and 2012.

Monthly average radon concentrations are shown in Table 1 from the Gallery entrance and Nevada Chamber along 2011 and 2012. The driest and warmest months (June, July and August) descend around 8.25 kBq·m⁻³ from the total annual mean, being August the most ventilated month. In contrast, in cold or/and wet months (winter and spring seasons) 5.5 kBq·m⁻³ are increased, being in this case, November with the highest radon activity of cave air. It has been spatially represented (Figure 3) the radon concentration by the use of 12 passive radon detectors (kodalphas) comparing fall and summer seasons. The spatial maps for each parameter were constructed using geostatistical gridding based on an ordinary point kriging method, with a linear variogram model and without anisotropy constraints during interpolation (distance between estimated points ranges from 3 to 5 metres). In Table 2 are shown the radiation values observed in the Figure 3.

Figure 3. Spatial distribution of the radon average concentration expressed in Bq·m⁻³ compares November 2011 and August 2012. Points 1 to 12 indicate cave locations where monthly average concentration of radon concentration was measured with nuclear-track detectors.

The spatial radon network in November 2011 shows higher accumulation than in August 2012. Thus, November 2011 recorded a monthly radon concentration of 30.3 kBq·m⁻³ compared to

August 2012 which recorded $22.7 \text{ kBq}\cdot\text{m}^{-3}$, corresponding to $7.65 \text{ kBq}\cdot\text{m}^{-3}$ of difference between both months. Especially the gas stacked at the Gallery entrance (point 12) as well as in Nevada Chamber (point 11) and points 4 and 7 recording values between $33.9 \text{ kBq}\cdot\text{m}^{-3}$ to $39.0 \text{ kBq}\cdot\text{m}^{-3}$, respectively. In contrast, in August 2012 those same points registered the lowest radon accumulations, ranging from $16.0 \text{ kBq}\cdot\text{m}^{-3}$ to $24.7 \text{ kBq}\cdot\text{m}^{-3}$. As a consequence, there is amplitude between fall and summer of $23.0 \text{ kBq}\cdot\text{m}^{-3}$ in the cave air. This may occur as the host rock at these points is more fractured and, therefore, there is a higher communication with the exterior atmosphere.

	NOVEMBER 2011	AUGUST 2012
Points	$[^{222}\text{Rn}] \text{ Bq}\cdot\text{m}^{-3}$	$[^{222}\text{Rn}] \text{ Bq}\cdot\text{m}^{-3}$
1	28505 ± 285	24205 ± 1694
2	24075 ± 241	23283 ± 1630
3	23479 ± 235	24360 ± 1705
4	38217 ± 382	22075 ± 1545
5	26494 ± 265	22257 ± 1558
6	32159 ± 643	22476 ± 1573
7	33968 ± 340	16026 ± 1122
8	23513 ± 235	27173 ± 1902
9	26587 ± 266	23545 ± 1648
10	32528 ± 325	23988 ± 1679
11	35947 ± 359	24787 ± 1735
12	39039 ± 390	18540 ± 1298
TOTAL	30376 ± 331	22726 ± 1591

Table 2. Radon concentration using passive detectors for November 2011 and August 2012 distributed along 12 different points inside Castañar cave.

3.2. Experimental assessment of occupational exposure to alpha radiation prior to cave opening to tourism

Throughout 2011 and 2012, the scientific visits to the cave focused on the equipment control, maintenance and download data had been used to evaluate the accumulated dose that the researchers received due to the radon exposure.

Figure 4 displays the hours spend inside by a worker (researcher) along 2011 and 2012 (grey vertical lines) and reflect the difference between calculating with radon monthly average (red line) or by a continuous radon monitoring (blue line). Along, this period, 28 visits were done implying a total of 73 hours inside the underground environment. By average, each visit lasted 2h 40' with a radon average concentration of $30.7 \text{ Bq}\cdot\text{m}^{-3}$ receiving a dose of 0.37 mSv per visit.

Figure 4. Comparison between accumulated doses obtained by a continuous monitoring system or by a monthly average Rn concentration expressed in mSv.

This figure shows the importance of a continuous radon monitoring (two records per hour) rather than the use of monthly radon averages as it reduces the accumulation dose error along a complete annual cycle. The dose received by a worker considering a unique monthly average value was overestimated in compare to the dose calculated with the exact radon concentration during the time the worker is inside the underground environment. Therefore, during 2011 the monthly average measured 0.21 mSv more than the continuous detectors, whether in 2012 it increased to 0.61 mSv.

The total accumulated dose received by a worker was lower than 6 mSv, independently to the radon concentration, along both years.

3.3. Guidelines for a safety occupational program and assessment of the cave recommendations throughout 2014.

The Spanish law catalogues the workplaces in relation to the effective dose received by workers into two categories:

- Monitored zone; it sets the possibility of being exposed to an effective dose lower than 6 mSv per year. It is recommended to calculate an area dose to assess the radiation exposure.
- Controlled zone; it implies the possibility of being exposed to an effective dose between 6 and 20 mSv per year. It is permitted to be expose in a year to a total dose of 50 mSv. It is mandatory, periodical health tests.

Focusing our results in the monitored zone catalogue Figure 5 shows that in November 2011 points number 4, 7, 11 and 12 allow between 2.82 to 3.24 hours. Instead, in August 2012 those same areas allow a range between 4.45 to 6.88 hours.

Figure 5. Maximum hours permitted per month for a worker in a monitored zone (< 6 mSv per year) considering average radon concentration values. November 2011 and August 2012 are compared representing the fall and summer seasons. Data from the network of 12 passive radon detectors (kodalphas) were considered.

In Figure 6, a recommended path has been drawn in both situations to delimitate the safest areas during a visit set in 50 minutes long according to the exposure to radiation. The effective dose average in August 2012 was 0.103 mSv/h, in contrast to November 2011 which was of 0.138 mSv/h.

Figure 6. Effective dose map (mSv/h) according to a monitored zone restriction. In addition, a recommended path (red bold line) is proposed aiming to minimize the health risk of workers because of the exposure to radon or its decay products associated to a 50 minutes visit.

In winter, is not recommended to visit Nevada Chamber (point 11) instead, visiting points number 8 and 9 would be a proper alternative. In summer, two different options can be chosen. Firstly, across Nevada chamber visiting point 7 or secondly, visiting points 8 or 9. In fall, along the path the effective dose received would be 0.121 mSv, whereas in summer it will be reduced to 0.086 mSv.

Cave visits were cancelled on September 11th 2008 because of a fungal outbreak in the Gallery entrance (Jurado et al., 2010), and it remained closed to tourism during the period covered by this study. The 13th of June of 2014 the cave was adequate to reopen to touristic visitors.

The recommendations given to the cave managers implied visits with groups of up to 5 people plus a guide during a visit set in 50 minutes. Furthermore, touristic visits would be avoided

from October to March (both included) as it represents the period with the highest radon concentrations. Therefore, from the reopen to the end of September 2014 is a suitable period to assess whether the recommendations given to the cave managers have been effective.

FIRST GUIDE	TOTAL TIME SPENT IN CAVE	AVERAGE PER VISIT	TOTAL EXCESS TIME (>50 min)	EXCESS TIME PER VISIT (>50 min)	²²² Rn (Bq·m ⁻³)	DOSE (mSv)	DOSE (mSv) UP TO 50 min	NUMBER ENTRIES
jun-14	15h 15´	1h 10´	4h 25´	20´	30400	2.11	1.49	13
jul-14	14h 28´	1h 26´	6h 08´	36´	28700	1.84	1.08	10
aug-14	10h 10´	1h 07´	2h 40´	17´	24900	1.15	0.84	9
sep-14	9h 25´	1h 20´	3h 35´	30´	30100	1.32	0.80	7
TOTAL	49h 18´	1h 16´	16h 48´	26´	28500	6.41	4.22	39

Table 3. Monthly data for the presence of the first guide in Castañar cave after it reopened to tourism. Columns correspond to: total the time spent inside the cave (hours), the average per visit (hours), the total excess of time inside the cave surpassing the 50 minutes per visit (hours), the excess of time per visit (hours), the average ²²²Rn concentration (Bq·m⁻³) per visit based on Nevada chamber and Gallery Entrance data, the total dose per month (mSv), the total dose per month if the visit would have lasted 50 minutes (mSv) and the total number of entries.

The guide (Table 3) spent more than 49 hours inside the cave being exposed to an effective total dose of 6.41 mSv, surpassing the effective dose limit of a monitored zone. If the visits would have lasted 50 minutes, the total effective dose would have been reduced to 4.22 mSv.

Figure 7. Comparative of the total effective dose received by the first cave-guide without time limit (light blue line) and the linear modelized effective dose that would be received with a time limit of 50 minutes (dark blue line).

Figure 7 represents a comparison between the total hours permitted before reaching the total effective dose of 6 mSv per year. If the visits would have lasted around 50 minutes, the guide could have been nearly 9 hours more inside the underground environment. Therefore, it could have been done 10.8 more entries.

4. CONCLUSIONS

Castañar de Ibor show cave contains very high ^{222}Rn levels that must be taken into consideration for the public and workers safety, therefore the restrictions given by the International Commission on Radiological Protection and the Spanish legislation must be followed carefully.

The high continuous frequency monitoring from two different points (Gallery entrance and Nevada chamber) inside the cave along 2011 and 2012 has permitted to understand the behaviour of the radon gas at a seasonal scale. Seasonal variations with summer minimums and maximum values in fall **were recorded**. Cold and wet seasons favours the accumulation inside the underground environment resulting November to be the month with the highest average concentration ($37.4 \text{ kBq}\cdot\text{m}^{-3}$), whereas hot and dry seasons allows the cave air ventilation reducing its concentration to the minimum, representing August as the month with the lowest average concentration ($20.3 \text{ kBq}\cdot\text{m}^{-3}$). **The vapour condensation or rainfalls hide the radon exchange by gas diffusion through the air-filled porosity of soil and rock discontinuities, determining the strong seasonal shifts of radon activity concentration in underground air.**

According to the spatiotemporal radon concentration map, the effective doses in a monitored zone have been calculated showing that the points with high radon concentration implies higher ionizing dose. Consequently, two touristic paths, corresponding to fall and summer conditions, have been recommended in order to avoid the higher dose areas for the safety of the guide and general public. Recommending a 50 minutes touristic visit, in fall the effective dose received would be 0.121 mSv , whereas in summer it will be reduced to 0.086 mSv .

The 13th of June 2014 the cave reopens to touristic visits, being a suitable moment to analyse if the recommendations given have been effective. The guide received an effective dose of 6.41 mSv surpassing the limit of a monitored zone in just 4 months. Visits lasted for 1:16:00 minutes, on average, instead of the 50 minutes recommended. If the guide had not surpassed the time limit recommended the effective dose would have been reduced to 4.22 mSv . Meaning that the guide could have done 10.8 more entries without surpassing the 6 mSv/year along those 4 months.

Finally, calculating the effective doses with a monthly radon average rather than with a high continuous radon monitoring implies an overestimation in the effective total dose. In 2011 the monthly averages measured 0.21 mSv more than the continuous detectors, whether in 2012 it increased to 0.61 mSv . Therefore, a high continuous monitoring frequency would represent better the reality of the subterranean environment to establish a proper visits regime.

Acknowledgments

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Figure1

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Table 1
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MONTHS 2011-2012	NEVADA CHAMBER [²²² Rn] Bq·m ⁻³	GALLERY ENTRANCE [²²² Rn] Bq·m ⁻³	AVERAGE of cave air [²²² Rn] Bq·m ⁻³
January	37300	35600	36400
February	36400	35300	35900
March	38000	36200	37100
April	36400	35400	35900
May	33900	31700	32900
June	29500	26700	28100
July	26400	18900	22600
August	24400	16200	20300
September	29000	24500	26700
October	34000	31000	32500
November	37700	37200	37500
December	37000	37800	37400
TOTAL	33300	30500	31900

Table 2
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	NOVEMBER 2011	AUGUST 2012
Points	[²²² Rn] Bq·m ⁻³	[²²² Rn] Bq·m ⁻³
1	28505 ± 285	24205 ± 1694
2	24075 ± 241	23283 ± 1630
3	23479 ± 235	24360 ± 1705
4	38217 ± 382	22075 ± 1545
5	26494 ± 265	22257 ± 1558
6	32159 ± 643	22476 ± 1573
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10	32528 ± 325	23988 ± 1679
11	35947 ± 359	24787 ± 1735
12	39039 ± 390	18540 ± 1298
TOTAL	30376 ± 331	22726 ± 1591

Table 3
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FIRST GUIDE	TOTAL TIME SPENT IN CAVE	AVERAGE PER VISIT	TOTAL EXCESS TIME (>50 min)	EXCESS TIME PER VISIT (>50 min)	²²² Rn (Bq·m ⁻³)	DOSE (mSv)	DOSE (mSv) UP TO 50 min	NUMBER ENTRIES
jun-14	15h 15′	1h 10′	4h 25′	20′	30400	2.11	1.49	13
jul-14	14h 28′	1h 26′	6h 08′	36′	28700	1.84	1.08	10
aug-14	10h 10′	1h 07′	2h 40′	17′	24900	1.15	0.84	9
sep-14	9h 25′	1h 20′	3h 35′	30′	30100	1.32	0.80	7
TOTAL	49h 18′	1h 16′	16h 48′	26′	28500	6.41	4.22	39