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Continuous monitoring of radon gas as a tool to understand air dynamics in the cave of Altamira (Cantabria, Spain)

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ABSTRACT

The use of radon as an atmospheric tracer in the Altamira Cave over the past 30 years has provided relevant information about gaseous exchanges between the Polychromes Room, the adjoining Chambers inside the cave, and the outside atmosphere. The relatively simple physico-chemical behaviour of radon gas provides a marked advantage over other tracer gases that are usually present in high concentrations in hypogeous environments, such as CO_2. Two types of continuous radon measurement were undertaken. The first involves active detectors located in the Hall and Polychromes Room, which provide radon concentration values at 1-hour intervals. In addition, nuclear solid track etched detectors (CR-39) are used in every chamber of the cave over 14-day exposure periods, providing average radon concentrations. In this paper we show some of the specific degassing and recharge events identified by anomalous variations in the concentration of radon gas in the Polychromes Room. In addition, we update knowledge regarding the degree of connection between chambers inside the cave and with the outside atmosphere. We verify that the connection between the Polychromes Room and the rest of the cave has been drastically reduced by the installation of the second closure in 2008. Except for point exchanges with the Crossing zone generated by a negative temperature gradient in that direction, the atmosphere of the Polychromes Room remains stable, or else it exchanges matter with the outside atmosphere through the karst interface. The role of radon as a tracer is demonstrated to be valid both to reflect seasonal cycles of degassing and recharge, and to analyse shorter (daily) period fluctuations.

1. Introduction

The Cave of Altamira is known worldwide for the ancient rock art that it contains. The pictorial representations dating back about 15,000 years, mainly concentrated in the so-called Polychromes Room, are rank among the best artistic manifestations in the history of Humanity, their value being not only symbolic, but also technical and representative (Lasheras et al., 2014). Altamira Cave was named a World Heritage Site in 1985 by UNESCO (http://whc.unesco.org/en/list/310).

Since its discovery in around 1868, the cave has been visited by a large number of people attracted by the beauty and realism of the pictorial representations it contains. In 1977, the great influx of visitors led to the decision to close the cave in order to conserve its cultural heritage due to increase in CO_2, temperature and lighting which promote the development of Lampenflora (Pfendler et al., 2018). During this first closure, scientific studies began to investigate how best to conserve the paintings. These studies included a detailed analysis of both the geological and environmental setting of the paintings, and the impact that the presence of visitors exerted on their state of conservation (Villar et al., 1984b, 1986). These studies provided the first integral view of the physico-chemical conditions under which the paintings were preserved, and provided the first models to explain the dynamics of gaseous exchanges between the various chambers inside the cave and between the cave and the outside environment (Villar et al., 1983, 1984a, 1985; Villar, 1986). Thanks to the systematic acquisition of further environmental data, and to the improvement of the theoretical models over successive research projects, we currently have quite a complete and complex vision of the main thermal, hydric and atmospheric processes that affect the conservation of the paintings, and also of the possible impacts that the ingress of visitors would have on those processes (Soler et al., 1999; Sánchez-Moral et al., 1999, 2010).

All work to date has used the concentration of radon gas (^{222}Rn) inside the cave to investigate the atmospheric dynamics of the Altamira Cave, (Fernández et al., 1986; Lario et al., 2005; Cuezva et al., 2011; Garcia-Anton et al., 2014). It is well known that radon is a natural radioactive gas that comes from the disintegration of the ^{226}Ra present in variable amounts in most rocks and soils of the earth's crust; radon tends to accumulate in poorly ventilated ground or underground enclosures (Quindós et al., 1991). Radon gas has been used as a tracer of dynamic processes in aqueous and aerial media on

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numerous occasions (Fernández et al., 1984; Frisia et al., 2011; Kowalczk and Froelich, 2010; Sainz et al., 2016). The relatively simple physico-chemical behaviour of radon gas means that it has a marked advantage over other tracer gases, such as CO_2, which are usually present in high concentrations in hypogeous environments (Batiot-Guilhe et al., 2007; Nazaroff and Nero Jr, 1988). The generation of radon gas inside the cave can be considered practically constant as there are no processes that significantly alter either the concentration of ^{226}Ra in the rock or the emission of radon over its surfaces. For this reason, positive increments in radon concentration inside a rock chamber can only be due to its increased isolation with respect to the outside atmosphere, which typically has very low radon concentrations, or to a supply of air from adjacent chambers containing significantly higher concentrations of the gas. Likewise, reductions in radon concentration in such a chamber can only be produced by radioactive decay or due to an air supply containing a lower concentration of the gas, usually from more superficial or external locations. Radon is a noble gas, which makes it highly unlikely that its concentration reduces by means of chemical reactions with the environment in which it is found (Nazaroff and Nero Jr, 1988).

Over the past 30 years, the use of radon as an atmospheric tracer in Altamira Cave has provided relevant information about gaseous exchange between the Polychromes Room, the adjoining chambers inside the cave, and the exterior atmosphere. During the 1980s the first estimates of the rates of air exchange between the Polychromes Room and the large hall inside the cave entrance, called the Hall, were made (Fernández et al., 1986). By also using the CO_2 concentrations, they proposed a model of air exchange which was driven by the temperature gradient between the two chambers at different times of the year. In 2008, this connection was significantly altered with the installation of a second seal door designed to reduce the ingress of organic matter into the Polychromes Room and to stabilise its temperature. Later studies provided detailed descriptions of the exchange processes between the Polychromes Room and the exterior through the karst host rock (Sánchez-Moral et al., 2009, 2012; Somavilla et al., 1978).

As in other caves, the seasonal loading and discharging of gases in the Altamira Cave – particularly in the Polychromes Room – are easily visualised by continuous monitoring of radon and CO_2 concentrations (Cuezva et al., 2011). In deep caves, with a single point of connection with the outside, usually the entrance, concentrations of radon and CO_2 are at a maximum in summer, and at a minimum in winter. These dynamics can be easily explained by the mass displacements that are caused by differences in air density between the cave interior and exterior at different times of year. However, the Cave of Altamira is quite shallow – the overburden above its ceilings varies from 2 to 10 m, although it can reach 18 m in places (Elez et al., 2013). Its karstic configuration favours gaseous exchange with the exterior through parts of the cavity other than the main entrance. For this reason, the dynamics of tracer gas concentrations (such as Rn and CO_2) can be very different, or even opposite to what is usually observed in deep caves.

This overall dynamics of gas exchange within Altamira Cave has various exceptions, such as in the event of an abrupt change or inversion of the temperature gradient between the outer and inner atmosphere of the cave. This article outlines some specific degassing and recharges events identified by anomalous variations in radon gas concentration in the Polychromes Room. In addition, knowledge about the degree of connection between Chambers inside the cave and between these and the outside atmosphere is updated.

2. Material and methods

2.1. Location and characteristics of the Altamira Cave

The Altamira Cave is located in Santillana del Mar (Cantabria, Spain) at an elevation of more than 150 m above sea level. Its entrance faces North and the cave as a whole is oriented northwest. It is a rather shallow cave, with a difference in elevation of around 16 m between the level of the entrance and its deepest point of (Fig. 1).

About 25 m from the entrance, just beyond the second artificial door, is a junction, called El Cruce ('The Crossing'). Ahead lies the access to the deeper chambers of the cave, while to the left, descending about 2 m *via* some stairs, is the access to the Polychromes Room. This chamber is about 15 m long and 7 m wide, with an average height of less than 3 m; it is here that the spectacular coloured pictorial representations on the ceiling were made, which led to the cave being dubbed the 'Sistine Chapel' of Quaternary Art (Déchelette, 1908; Elez et al., 2013) The volume of the Polychromes Room is approximately 342 m^3 and the ceiling area is 159 m^2.

The microclimate inside the cave is characterised by very stable temperatures throughout the year, with a thermal oscillation in the Polychromes Room of less than 2 °C. This oscillation is described by a sinusoidal wave that is out of phase and damped with respect to the annual thermal oscillation outside. This is due to the layer of rock between the ceiling of the chamber and the ground surface immediately above.

2.2. Radon gas concentration measurements

Two types of continuous radon measurement are made inside the Cave of Altamira. The first involves active detectors located in the Hall and Polychromes Room, which provide radon concentration at 1-hour intervals. In addition, in every chamber of the cave, nuclear solid track etched detectors (CR-39) provide averaged radon concentrations over exposure periods of two weeks.

RadonScout monitors (SARAD GmbH) are used for the continuous radon measurements, their sensors are Si detectors that converts the energy yielded by alpha emissions of radon and its progeny into electrical impulses whose magnitude is proportional to the energy absorbed. This device performs alpha spectrometry to measure the amount of radon in the air. Measurements can be made at intervals of between 1 and 3 h, and are written to memory, allowing storage of data series more than two months long. Concentrations can be measured in the range 0 to 10 MBq/m^3. The uncertainty associated with the measurement performed by this device varies from 20% to 10% in the concentration range of 100–1000 Bq/m^3. Each RadonScout monitor is calibrated every six months in LaRUC radon chamber described below, using an AlphaGUARD (SAPHIMO GmbH) device as reference traceable to the PTB (*Physikalisch Technische Bundesanstalt*) ^{222}Rn chamber (Germany) of using a primary PTB gaseous ^{222}Rn activity standard. In addition, detection efficiency is verified in the data download performed every two weeks.

The main difficulty presented by the continuous measurement of the radon concentration in underground environments is the continuously high relative humidity of the cave atmosphere. In addition to possible damage to the electronic components of the monitors, high humidity can modify detection efficiency in electrostatic radon descendant collection systems (George, 1996). In order to minimise the influence of ambient humidity on the operation of continuous monitors, they are placed in a low density waterproof plastic bag, which allows the transfer of radon through its surface by diffusion. Following the procedure of Moreno et al. (2015), it has been demonstrated in a controlled atmosphere that the radon measurement inside the bag

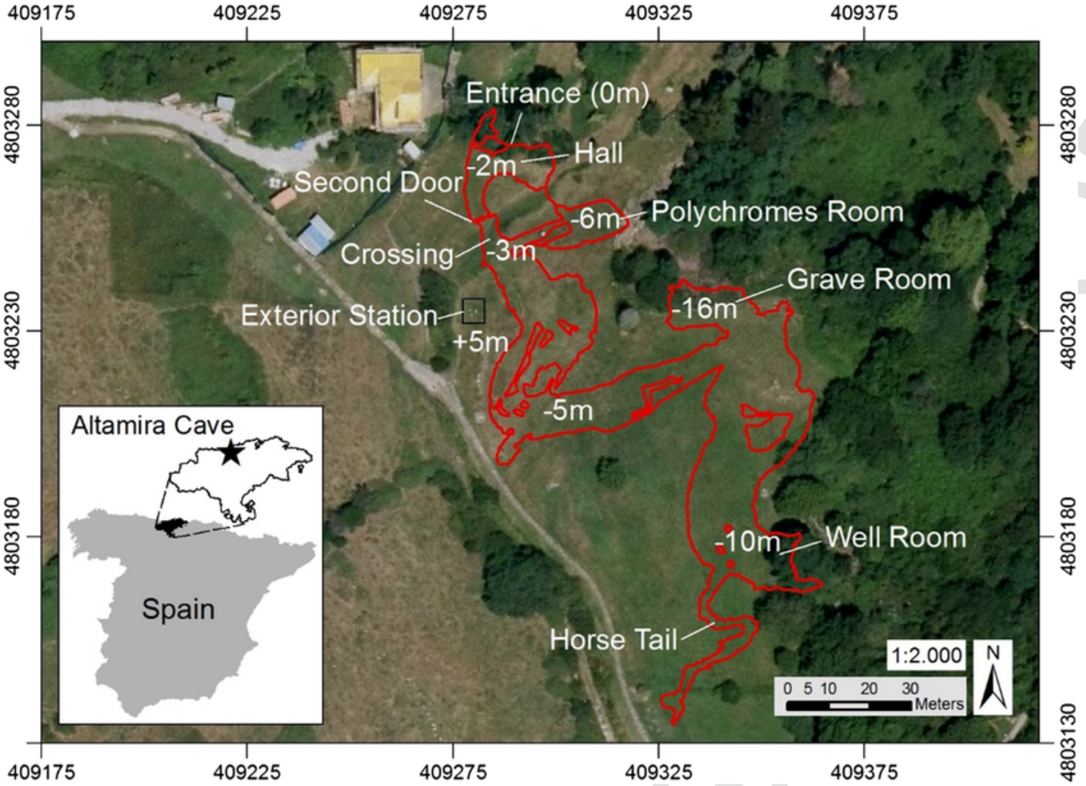

Fig. 1. Plan of the Altamira Cave superposed on an orthophoto. The differences in height shown are relative to the elevation at the cave entrance.

implies an initial delay in the response to changes in concentration between 3 and 6 h. For this reason, the first ten measurements are discarded.

In contrast, track etch detectors CR-39 (Radosys Ltd.) were used to obtain integrated values of the radon concentration every 14 days. Measurements using these devices are based on the effect of α particles on different plastic materials. Radon and its progeny emit α particles that produce a track in the detector. The plastic material is placed in a container and radon diffuses through the container preventing the entrance of its progeny. At the end of the measurement period, the chamber is sealed and transferred to the laboratory, where it is analysed.

In the laboratory, the track etch detectors (consisting of a plastic polymer) are chemically treated and the number of tracks per unit area of the detector is counted under a microscope using an automatic optical reader. Microscope is checked, with a tolerance range, every time it is used with a traceability slide where tracks density is known. Once the calibration factor of the equipment is known, the number of tracks per unit area allows the radon concentration to be calculated. The exposure time of these detectors varies depending on the range of concentrations to be measured. The calibration factor is provided by the manufacturer, however we do a double check using our radon chamber and the reference radon monitor mentioned above.

All radon measurements are subject to a double quality control on a continuous and periodic basis. The Laboratory of Environmental Radioactivity of the University of Cantabria (LaRUC) is accredited according to UNE-EN ISO/IEC 17025:2005 for integrated radon in air measurements with CR-39, which implies rigorous quality control of the entire measurement process, including periodic international intercomparison exercises. In this way continuous radon measurements with RadonScout monitor are controlled. In addition, all available measurement systems are periodically checked in the radon chamber at LaRUC, by exposing them to known concentrations of radon. This chamber is a cubic container of 1 m^3 volume, located in the Faculty of Medicine of the University of Cantabria. The walls of the chamber consist of 3.25 mm thick stainless steel plates welded together to ensure a tight seal. The top plate acts as a removable cover for the insertion and removal of equipment and sources, and has three holes that allow exposure to sets of track etch detectors without having to open the whole assembly.

The radon chamber is equipped for simultaneous exposure to various radon detection systems, and has electrical connections to the required power sources. It also has valves to allow gas exchange with the outside air, so allowing exposure to radon concentrations to be varied as required. Homogeneity of the radon concentration inside the chamber is achieved using a small fan, ensuring that concentration differences within the chamber are less than 3%. The tightness of the assembly is guaranteed by seals made of materials with a low diffusion coefficient for radon gas; by this means, an exchange rate with the exterior of lower than 0.01 h^{-1} is achieved.

2.3. Continuous monitoring of air pressure, CO$_2$ concentration and air temperature

For carbon dioxide and air temperature measurements, there are six stations (Fig. 2) located at different points in the Cave (Exterior, Hall, Crossing, Polychromes Room, Grave Room and Well Room). Each station contains several thermometers to measure air temperature at different heights, CO$_2$ concentration, relative humidity and atmospheric pressure. Data loggers record the values of each variable every 15 min, with daily online downloads of data. A station outside the cave records the same parameters in the outdoors air.

The air temperature probes inside and outside are Pt100 models, with 3 × 6 mm-diameter wires, 40 mm long INOX AISI 316. Temperature is measured to a resolution of 0.01 °C, certified to an accuracy of 0.06 °C. The probes used to determine CO$_2$ concentration are

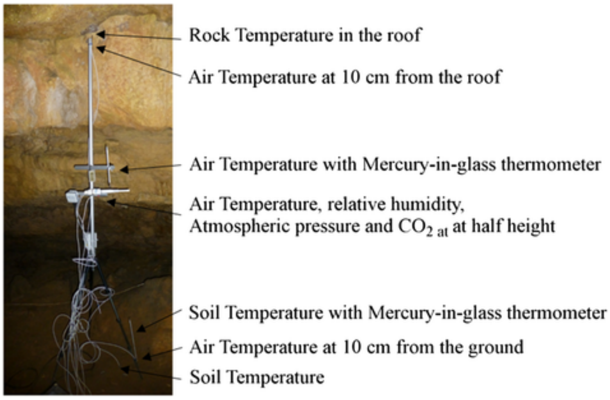

Fig. 2. Sampling station in the Hall, indicating the various sensors used for environmental monitoring of the cave.

EE82 Series instruments, are accurate to within 100 ppm and a resolution of 1 ppm. The pressure sensor is a Delta OHM HD 9408, with an accuracy of 0.4 hPa at 20 °C, and a resolution of 0.01 hPa.

2.4. Air density calculation

As mentioned above, temperature gradients lead to variations in air density which cause convective movements due to gravity. In order to study episodes of gas exchange between the Polychromes Room and the exterior, the corresponding densities were evaluated over the entire time series available. The calculations differ slightly for the external and internal air, taking into account the high moisture content and CO_2 in the latter case. The expressions used are based on the following references (Garcia-Anton et al., 2014; Buck, 1981).

2.5. Estimates of exchange rates

In order to assess exchange rates between the Polychromes Room and other cave chambers or the exterior, the simplified model of Wilkening, used by Fernández et al. (1986), was used. This mathematical model considers that the radon concentration in a chamber varies according to the quantity emitted through the surface of the rocks, the quantity that disappears by disintegration, and the variations produced by exchange with air masses containing different concentrations from other chambers and/or the outside atmosphere.

The exchange rate can be calculated using the following expression, assuming that it becomes practically nil when the radon concentration reaches its maximum value (Fernández et al., 1986):

$$Q = \lambda \cdot V \cdot \frac{\left(C_{\max} - C\right)}{\left(C - C'\right)} \qquad (1)$$

where

Q is the exchange rate (m^3/h)

λ is the radon disintegration constant ($0.0076\ h^{-1}$)

V is the volume of the Polychromes Room ($342\ m^3$)

C is the radon concentration in the Polychromes Room (Bq/m^3)

C' is the radon concentration in the chamber with which it exchanges (Bq/m^3)

$C_{\max}$ is the maximum annual radon concentration in Polychromes Room (Bq/m^3)

To estimate the exchange rate with the outside air, it can be assumed that radon concentration is practically zero in the free atmosphere, therefore $C' \approx 0$.

3. Results and discussion

3.1. Annual distribution of air temperature

As indicated above, the air temperature field inside the Altamira Cave is fundamentally determined by the annual thermal oscillations of the outside air. These oscillations are transmitted into the cave through varying thicknesses of rock (Elez et al., 2013), which causes the damping and lags of the thermal waves to be different in each of the interior chambers (Monteith and Unsworth, 2013).

Fig. 3 shows air temperatures in the Exterior, Hall, Crossing and Polychromes Room between 2013 and 2016. The thermal wave is propagated through the rock, which attenuates its amplitude and causes a lag between the outer thermal wave and the inner one. Each of the temperature time series inside the cave was shifted in time to the point of maximum correlation with respect to the outside temperature, and by this means information about the lag between waves is obtained. The lag for the Hall is 1 month, for the Crossing, 3 months and for the Polychromes Room, 4 months, according to Villar et al. (1984b) and Villar (1986). Table 1 shows the monthly average air temperature at half height in the Exterior, Hall and Polychromes Room.

In spite of the remarkable regularity in the annual oscillation in temperature in every chamber, there are a number of degassing and recharge episodes that are related to abrupt variations in the thermal gradients between the interior and exterior parts of the cave.

3.2. Annual distribution of Rn and CO_2 concentrations

As for evolution over time, there is a clear seasonal pattern in both the Hall and Polychromes Room. Concentrations in both are lowest between June and October; and highest between November and April when the exchange between the cave and the outside air is at a minimum (see Fig. 4). The monthly Rn and CO_2 concentrations in the Hall and in the Polychromes Room are shown in Table 1. In general, measurements in the Hall are lower than in the Polychromes Room, though this trend can be reversed during transition periods. During these transition periods, like March, oscillations in Rn concentration increase, especially near the mouth of the cave.

Correlations between the various chambers were calculated from the continuous data series of air temperature ΔT, pressure ΔP and density $\Delta\rho$. The gradients between the Exterior and the two chambers studied (Hall and Polychromes Room) are represented in Fig. 4, alongside the concentrations of the tracer gases, CO_2 and Rn. Table 2 shows the linear correlation coefficients obtained for a 95% confidence interval and highlights how radon and carbon dioxide behaviours are similar. There are small (r about − 0.3) and medium (r − 0.5) negative correlations between air pressure and gas concentration in the Hall and Polychromes Room, respectively. On the other hand, there are high negative correlations with temperature (r around − 0.8), and high positive correlations with air density (r around 0.8) – both higher in Polychromes Room. This indicates that air temperature and density gradients dominate the behaviour of gases within the cavity.

Degassing in the Polychromes Room reaches its maximum expression during the summer, when the average inside air temperature is much lower than outside. If exchange occurred only at the cave entrance, the colder, denser air in Polychromes Room and other deeper Chambers would barely reach the entrance, since these chambers lie at much lower levels than the cave mouth. For these reasons, radon and carbon dioxide concentrations are at a minimum in summer, while, they peak in winter when the karst matrix becomes impermeable through the combined effect of accumulated moisture in the

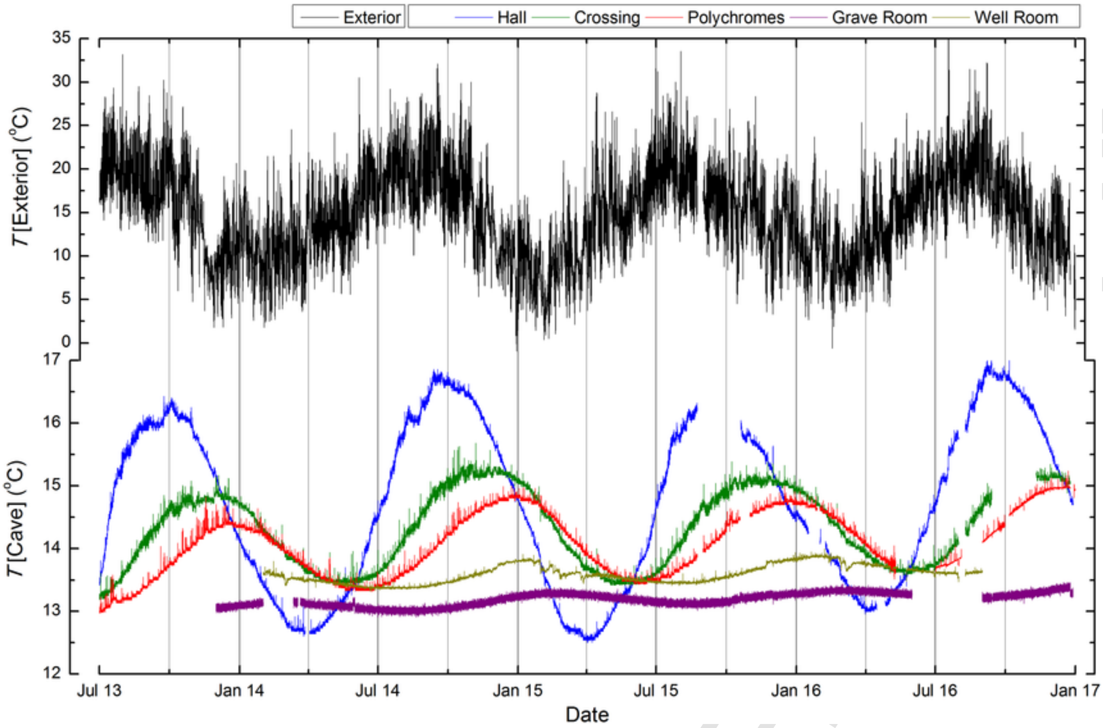

Fig. 3. Temperature at half height inside and outside the cave for the period 2013–2016.

cracks and fissures, and the higher density of the outside air. These observations accord with the general air dynamics described by other authors (Garcia-Anton et al., 2014).

Having demonstrated the capacity of continuous radon monitoring to describe the movement of air masses inside the cave and air exchanges with the outside air over annual periods, we analysed the degassing and recharge events evident over shorter periods. There are two periods in the year when fluctuations in Rn and CO_2 concentrations are significant, namely the season changes from spring to summer and from summer to autumn. During these periods, abrupt variations in trace gas concentrations can be observed over a single day or even just a few hours, which may be related to the highly variable diurnal alternations in external temperatures. We present an analysis of the period May 20 to June 9, 2015, where varieties of the above-mentioned short-period fluctuations were observed.

Fig. 5 shows the difference in air pressure, temperature and density between the outside atmosphere, the Polychromes Room and the Hall for the period 20 May to 10 June 2015. Radon concentrations in the Hall and Polychromes decreased from 5000 Bq/m^3 to below 1000 Bq/m^3. At this time of year the temperature of these two chambers is practically the same – as can be seen on a large scale in Fig. 3, or in detail in Fig. 5. There is a difference in their average temperatures over this period of less than 0.1 °C.

Chamber degassing occurs when the air density gradient is negative, *i.e.*, the density of the outside air is lower than the density of the indoor air ($\rho_{ext} < \rho_{cave}$). The denser air inside the cave tends to descend to lower levels through the pores and cracks of the karst, so favouring the observed decrease in the concentration of Rn. The greater degree of connection between the Hall and the Exterior means that degassing and recharge of this chamber is more pronounced than for the Polychromes Room. A strong correlation is found between the difference between Exterior-Hall densities and the concentration of Rn. The results of the correlation analysis are shown in Table 3.

3.3. Degree of connection between the chambers and with the exterior

The first estimates of the rates of gaseous exchange between the Polychromes Room and the rest of the Cave were made in the 1980s (Fernández et al., 1986). The estimates were made based on the hypothesis that the most intense convective air exchange was with the Hall, occurring mainly by circulation through The Crossing. It was assumed that the restricted connection of the various chambers of the cave with the exterior meant that the predominant mechanism for mass air movement was due to air temperature gradients established inside in the cave. A second door was installed in 2008 at the point where the Hall joins The Crossing. This radically modified the route for air exchange between the Hall and the Polychromes Room; but it also highlighted the importance of the fissures and cracks in the karst as an alternative route for air exchange with the outside atmosphere (Sánchez-Moral et al., 2009). The thermal gradients between the air in the Polychromes Room and the other chambers deeper in the cave are negative throughout the year (Fig. 3). Given the other chambers are at a lower level than the Polychromes Room, air exchange between the various chambers seemed unlikely. For these reasons, estimates of the rates of gaseous exchange in the Polychromes Room are made assuming that these exchanges occurred preferentially with the exterior through the voids of the karst on the basis of historic and contemporary data on radon concentrations.

The exchange rate between the Polychromes Room and the outside was estimated from Eq. (1) and the monthly mean values of radon concentration are summarised in Table 1. The maximum annual radon concentration C_{max} is shown in Table 4, the volume of Polychromes Room is 342 m^3 and the radon concentration from the outside C' was assumed to be zero. The results of the monthly exchange rate for 1983, 2008 and for the period 2013–2016 are given in Table 4, while they are displayed graphically in Fig. 6.

Table 1
Monthly average temperature *T*, carbon dioxide [CO_2] and radon concentration C_{Rn} in the Exterior, Hall and Polychromes Chambers. The uncertainty of temperature is 0.06 °C, in case of CO_2 concentration is 100 ppm, and for radon concentration is found between 15% for values around 500 Bq/m^3 and 6% about 5000 Bq/m^3.

Month	Exterior	Hall			Polychromes room		
	T (°C)	*T* (°C)	[CO_2] (ppm)	C_{Rn} (Bq/m^3)	*T* (°C)	[CO_2] (ppm)	C_{Rn} (Bq/m^3)
Jul-13	20.30	14.52	767	216	13.12	1134	–
Aug-13	19.22	15.73	709	335	13.26	873	636
Sep-13	18.09	15.56	915	986	13.56	1180	1443
Oct-13	16.67	15.66	1479	1322	13.90	1966	2137
Nov-13	11.03	15.51	2809	4573	14.23	3289	3949
Dec-13	9.65	14.61	3288	5034	14.38	3220	4450
Jan-14	10.34	13.79	3197	5260	14.30	3317	5146
Feb-14	9.55	13.16	2932	4553	14.10	3080	4561
Mar-14	10.63	12.73	2538	3734	13.83	2844	3849
Apr-14	13.16	12.76	2954	4781	13.57	3092	4933
May-14	13.45	13.18	3237	4692	13.40	3552	4554
Jun-14	17.43	13.97	982	656	13.36	1568	1795
Jul-14	18.83	15.07	801	556	13.47	974	901
Aug-14	19.10	16.05	731	550	13.70	912	855
Sep-14	19.83	16.61	672	387	14.03	828	701
Oct-14	17.83	16.53	910	917	14.38	1289	1452
Nov-14	13.26	16.06	2650	3759	14.64	2755	3217
Dec-14	9.48	15.22	3177	4500	14.79	3335	4878
Jan-15	8.39	14.31	2939	5112	14.76	3043	5051
Feb-15	7.13	13.24	2455	4647	14.50	2566	4844
Mar-15	9.83	12.68	2649	5507	14.11	2669	5357
Apr-15	13.88	12.67	2495	4017	13.77	2661	4060
May-15	14.71	13.20	2286	3025	13.54	2624	3184
Jun-15	17.49	13.95	858	629	13.48	1354	1185
Jul-15	19.91	15.31	713	263	13.57	812	497
Aug-15	19.60	16.03	778	499	13.76	850	695
Sep-15	16.41	–	–	1310	14.11	1269	1668
Oct-15	15.37	15.80	2096	2293	14.43	2189	2584
Nov-15	13.51	15.54	2466	3115	14.62	2896	3970
Dec-15	13.05	14.80	3185	4685	14.73	3263	5087
Jan-16	10.73	14.42	3330	5020	14.69	3377	5135
Feb-16	9.37	13.82	3182	5373	14.53	3179	5554
Mar-16	9.50	13.26	2937	4944	14.24	2893	5407
Apr-16	11.55	13.05	3436	5282	13.91	3289	5252
May-16	14.78	13.45	2527	3376	13.73	2937	3905
Jun-16	17.09	14.23	1115	795	13.69	1592	1566
Jul-16	19.32	15.29	804	357	13.75	887	618
Aug-16	19.96	16.30	764	239	13.84	785	519
Sep-16	18.58	16.78	883	827	14.25	942	1081
Oct-16	14.99	16.55	1870	1928	14.67	1961	2349
Nov-16	11.02	15.96	3702	4828	14.89	3354	4913
Dec-16	11.42	15.11	3452	5049	14.97	3802	5793

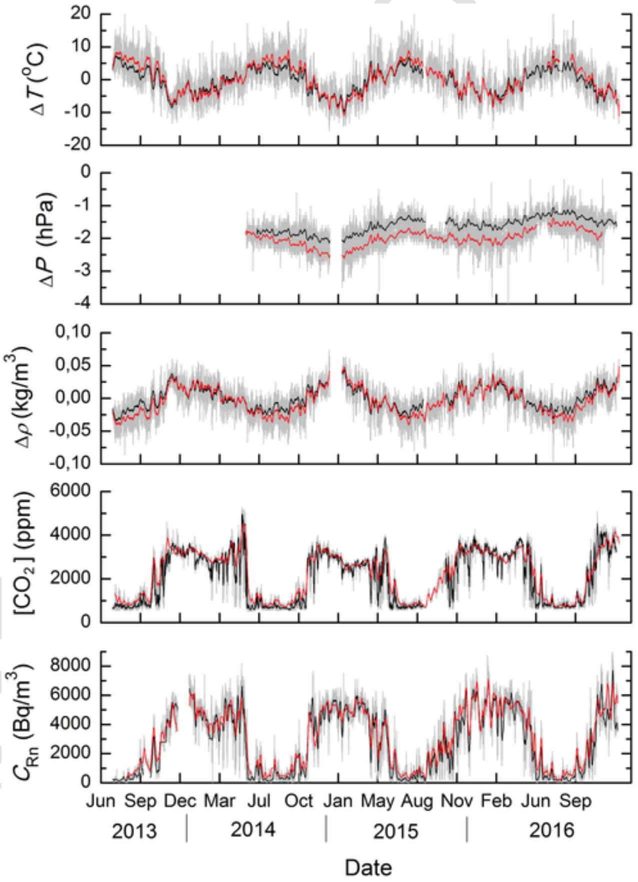

Fig. 4. Differences in temperature, pressure and density of the air between the Exterior and the Hall (black line) and Exterior and Polychromes Room (red line). Concentration of CO_2 and Rn in the Hall (black) and Polychromes (red). Graphs were constructed using smoothed weekly data. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The extraordinary increase in the rate of ventilation during the summer months might be explained by the significantly higher average temperature over this period. The greater difference in air densities in recent years, might have provoked greater degassing and enhanced connection of the chamber with the outside air. This explanation will need to be validated by collecting further data on annual cycles.

Table 2
Linear correlation coefficient, r, between the difference in air pressure, density and temperature in the Exterior and inside Altamira Cave (Hall and Polychromes Room) with tracer gases, CO_2 and Rn, obtained for a 95% confidence interval.

	$[CO_2]$ (ppm)	C_{Rn} (Bq/m^3)
ΔP (Ext.–Hall)	− 0.28 (− 0.34 to − 0.21)	− 0.34 (− 0.40 to − 0.27)
$\Delta \rho$ (Ext.–Hall)	0.77 (0.75 to 0.80)	0.75 (0.72 to 0.77)
ΔT (Ext.–Hall)	− 0.79 (− 0.80 to − 0.76)	− 0.76 (− 0.78 to − 0.73)
ΔP (Ext.–Poly.)	− 0.53 (− 0.57 to − 0.48)	− 0.51 (− 0.56 to − 0.46)
$\Delta \rho$ (Ext.–Poly.)	0.80 (0.78 to 0.82)	0.82 (0.78 to 0.84)
ΔT (Ext.–Poly.)	− 0.81 (− 0.83 to − 0.79)	− 0.82 (− 0.84 to − 0.80)

4. Conclusions

The concentrations of radon gas inside Altamira Cave can be used as an indicator of air movements both within the cave and in exchanges with the outside atmosphere. Its role as a tracer has been demonstrated to be valid both to reflect the seasonal cycles of degassing and recharge, and to analyse shorter (daily) period fluctuations.

Although air pressure and density are related magnitudes, air density gradients prove to be more sensitive indicators of convective motion than air pressure differences. Although the latter explain long-term aerodynamic changes with a relatively good precision, they are limited when it comes to predicting short-term exchanges.

We have verified that the connection of the Polychromes Room with the rest of the cave was drastically reduced by the installation of the second closure in 2008. Except for point exchanges with The Crossing generated by a negative temperature gradient in that direction, the atmosphere of the Polychromes Room remains stable, or else it exchanges matter with the exterior through the karst interface.

The annual distribution of temperature in the Hall and Polychromes Room has been updated; we observed a warming trend that could be related to a rising trend in exterior temperature. This warming could explain the increase observed in the ventilation rate between the Polychromes Room and the outer atmosphere. This needs to be corroborated with further specific analyses. Likewise, knowledge of the dynamics of the air inside the cave has been updated using radon gas as a tracer.

The distribution of the concentrations of radon and CO_2 present significant parallels. However, there are many situations of divergence of this trend, attributable to the different chemical behaviours and origins of both gases. The greater diversity and complexity of mechanisms of generation and disappearance of the carbon dioxide in the air, makes it more complicated to implement mathematical models that focus on the to evaluate quantitative exchanges of air masses between different rooms. This observation leads us to recommend radon gas concentration as a tracer, mainly due to the simpler mechanisms involved in the rises and falls in its concentration.

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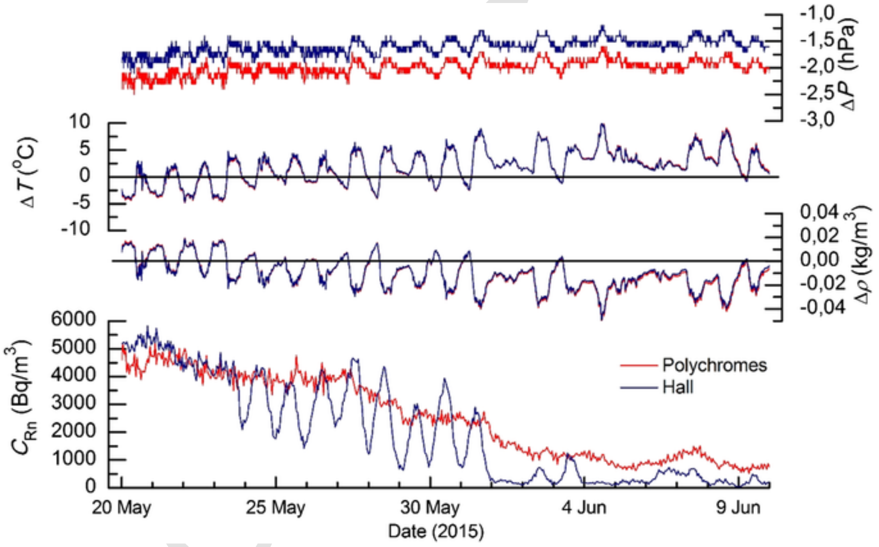

Fig. 5. Air pressure, temperature and density differences between the external atmosphere and Polychromes Room (red line) and Hall (blue line) for the period 20 May to 10 June 2015. The radon variation in each chamber is also shown. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 3
Linear correlation coefficient r between the difference in air pressure, density and temperature in the Exterior and in the Cave (Hall or Polychromes Room) with Rn concentration obtained for a 95% confidence interval for the period 20 May to 10 June 2015 (event shown in Fig. 5).

	C_{Rn} [hall] (Bq/m^3)	C_{Rn} [polychromes] (Bq/m^3)
ΔP (Ext.–Chamber)	− 0.08 (− 0.17 to − 0.01)	− 0.11 (− 0.19 to − 0.01)
$\Delta \rho$ (Ext.–Chamber)	0.48 (0.40 to 0.54)	0.59 (0.53 to 0.64)
ΔT (Ext.–Chamber)	− 0.48 (− 0.54 to − 0.41)	− 0.59 (− 0.53 to − 0.64)

Table 4
Monthly ventilation rate Q (m^3/h) between the Polychromes Room and the outside in 1983 (Fernández et al., 1986), 2008 (Sánchez-Moral et al., 2009) and the period 2013–2016. Maximum annual radon concentration C_{max} (Bq/m^3) is also given.

Month	1983	2008	2013	2014	2015	2016
January	–	–	–	1.16 ± 0.23	1.29 ± 0.23	1.82 ± 0.27
February	4.10 ± 0.40	0.91 ± 0.21	–	1.64 ± 0.25	1.46 ± 0.24	1.49 ± 0.25
March	0.60 ± 0.19	2.03 ± 0.28	–	2.42 ± 0.30	1.07 ± 0.22	1.60 ± 0.25
April	0.38 ± 0.18	2.06 ± 0.28	–	1.32 ± 0.24	2.24 ± 0.29	1.72 ± 0.26
May	0.15 ± 0.17	3.22 ± 0.35	–	1.65 ± 0.25	3.57 ± 0.37	3.21 ± 0.35
June	5.00 ± 0.46	8.34 ± 0.66	–	8.1 ± 1.1	14.0 ± 1.0	11.90 ± 0.87
July	16.3 ± 2.8	21.0 ± 3.6	–	18.9 ± 2.2	37.0 ± 5.9	34.1 ± 5.5
August	11.17 ± 0.83	24.7 ± 4.1	22.9 ± 3.8	20.0 ± 2.3	25.7 ± 4.2	41.1 ± 6.6
September	10.81 ± 0.81	10.25 ± 0.77	8.64 ± 0.68	25.0 ± 2.8	9.18 ± 0.71	18.4 ± 1.3
October	6.18 ± 0.53	3.43 ± 0.36	4.99 ± 0.46	10.71 ± 0.80	5.01 ± 0.46	7.07 ± 0.58
November	0.96 ± 0.21	1.05 ± 0.22	1.51 ± 0.25	3.41 ± 0.36	2.35 ± 0.30	2.02 ± 0.28
December	0.23 ± 0.17	1.05 ± 0.22	1.04 ± 0.22	1.36 ± 0.24	1.26 ± 0.23	1.32 ± 0.24
C_{max} (Bq/m^3)	7250 ± 440	7250 ± 430	6240 ± 370	7440 ± 450	7560 ± 450	8730 ± 520

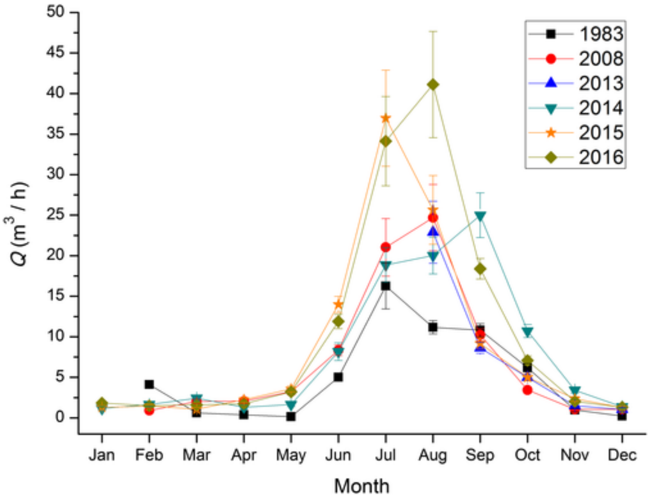

Fig. 6. Monthly variations in the ventilation rate Q (m^3/h) between the Polychromes Room and the outside in 1983 (Fernández et al., 1986), 2008 (Sánchez-Moral et al., 2009) and the period 2013–2016.

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