Reasons for large fluctuation of radon and CO$_2$ levels in a dead-end passage of a karst cave (Postojna Cave, Slovenia)

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Abstract. Measurements of radon concentration were performed at three geomorphologically different locations in Postojna Cave, Slovenia. In the part of the cave open to visitors, annual average radon activity concentrations of $3255 \pm 1190$ Bq m$^{-3}$ and $2315 \pm 1019$ Bq m$^{-3}$ were found at the lowest point (LP) and in the Lepe jamne (Beautiful Caves, BC), respectively. A much higher average of $25020 \pm 12653$ Bq m$^{-3}$ was characteristic of the dead-end passage Pisani rov (Gaily Coloured Corridor, GC), in which CO$_2$ concentration also reached very high values of 4689 $\pm$ 294 ppm in summer. Seasonal variations of radon and CO$_2$ levels in the cave are governed by convective airflow, controlled mainly by the temperature difference between the cave and the outside atmosphere. The following additional sources of radon and CO$_2$ were considered: (i) flux of geogas from the Earth’s crust through fractured rocks (radon and CO$_2$ source), (ii) clay sediments inside the passage (radon source) and (iii) the soil layer above the cave (radon and CO$_2$ source).

1 Introduction

The karst cave environment is characterised by very high microenvironmental stability with essentially quasi-closed air masses (Badino, 2010), and may be considered stable in comparison to the outside atmosphere. Beyond this apparent stability, however, complex processes occur, which can today be brought to light thanks to advances in measurement techniques, data storage and data processing. All caves should be considered fragile systems, but show caves are particularly at risk because of anthropogenic impact. An understanding of cave microclimates is of great importance when studying the thermodynamics of karst processes, palaeoclimatic proxies, hydrogeological aspects of speleothems, CO$_2$ build-up and cave ecosystems (Baldini et al., 2006; Faimon et al., 2011; Kowalczyk and Froelich, 2010; Perrier and Richon, 2010; Spötl et al., 2005; Tremaine et al., 2011). The thermal and moisture characteristics of the cave air (Badino, 2010; De Freitas et al., 1982), as well as the concentration of gases ($^{222}$Rn, CO$_2$) and aerosols (Bezek et al., 2012) are mainly controlled by the degree of air exchange with the outside environment. Convective air circulation, driven by buoyancy forces created by the difference of air density between the external and internal air masses, is a major mechanism controlling air circulation in caves with more than one entrance at different elevations (Badino, 2010; Cigna, 1968; Hakl et al., 1996; Kowalczyk and Froelich, 2010; Wigley, 1967). On the other hand so-called barometric circulation, driven by the internal–external pressure difference, can be very important for caves with large volumes connected by small passages, with one entrance or with extremely small entrances (Badino, 2010; Luetscher and Jeannin, 2004; Wigley, 1967). Therefore, in addition to outside atmosphere, cave geomorphology plays an important role in cave ventilation.

Radon ($^{222}$Rn, $\alpha$-radioactive, half-life $t_{1/2} = 3.82$ days) has often been used as an excellent tracer for air circulation, since it is a noble gas and highly abundant in caves (Cigna, 2003; Cunningham and Larock, 1991; Hakl et al., 1996, 1997; Kies and Massen, 1997; Kowalczyk and Froelich, 2010; Perrier et al., 2004; Przylibski, 1999). Its half-life, suitable for the timescales on which cave ventilation takes
place, distinguishes $^{222}$Rn from the other two radon isotopes ($^{220}$Rn and $^{219}$Rn). An additional advantage is its radioactivity, which makes it relatively easy to monitor the activity concentration of radon with a very low detection limit. Variation of radon concentration in cave air arises from a balance of radon emission from cave surfaces and drip waters, its radioactive transformation, and exchange with the outside atmosphere (Wilkening and Watkins, 1976). Radon concentration in underground cave systems is also characterised by internal mixing of air masses (Perrier and Richon, 2010). In a study of 220 caves around the world, Hakl et al. (1997) reported an annual average radon concentration of 2800 Bq m$^{-3}$.

The most important parameter governing dissolution and precipitation processes in carbonate karst is CO$_2$ (Dreybrodt, 1999), so understanding CO$_2$ distribution and dynamics in caves is important for palaeoclimatic research. The dynamics of CO$_2$ in caves is governed by the distribution and intensity of its sources and (mainly) advective transport by air currents. The main sources of CO$_2$ in caves are diffusion from the epikarst, decomposition of organic matter and precipitation of calcite from supersaturated solutions. Many authors therefore include cave ventilation when modelling CO$_2$ variation over time in order to explain seasonality and trend (Baldini et al., 2008; Fernandez-Cortes et al., 2011; Milanolo and Gabrovšek, 2009; Tanahara et al., 1997). CO$_2$ was used in this study as an additional tool to characterise and explain the sources and reasons of high radon concentration and its variability in a dead-end passage in Postojna Cave. However, the exact interpretation of short-term fluctuation of the CO$_2$ level remains outside the scope of this study.

The complexity and size of Postojna Cave, with its numerous known and unknown entrances at different levels and a long and highly ramified cave system, makes this cave a fascinating study site for a variety of physical and environmental studies (Bezek et al., 2012; Gosar et al., 2009; Gregorič et al., 2011; Kobal et al., 1988; Mulec et al., 2012; Šebela et al., 2010; Šebela and Turk, 2011; Vaupotič, 2008). In general ventilation of Postojna Cave is characterised by convective airflow, controlled mainly by the temperature difference between the cave and the outside atmosphere, as discussed by Gregorič et al. (2011) for one measurement location in the Velika Gora (Great Mountain) chamber of Postojna Cave. Different ventilation regimes in the cold and warm periods of the year are responsible for the observed seasonal pattern of radon concentration with low winter and high summer levels, as already reported for this (Gregorič et al., 2011; Kobal et al., 1988; Vaupotič, 2008) and several other caves (Gillmore et al., 2002; Kowalczk and Froelich, 2010; Nagy et al., 2012; Perrier and Richon, 2010; Przylibski, 1999; Tanahara et al., 1997; Wilkening and Watkins, 1976).

In one passage of Postojna Cave, known as the Pisani rov, very high radon levels have been observed (annual mean 25 020 $\pm$ 12 653 Bq m$^{-3}$) which are comparable to the highest radon levels of 32 246 Bq m$^{-3}$ (annual mean) measured in Castañar de Ibor karst cave in Spain (Lario et al., 2006), although the rest of the cave is characterised by radon levels similar to world average values (Hakl et al., 1997). This paper presents the results of 18 months (March 2011–September 2012) of continuous measurements of radon concentration in three passages in Postojna Cave, including Pisani rov, in which CO$_2$ concentration was additionally measured in two periods: 1 March–9 May and 28 June–1 September 2012. The aim of this study is to reveal the geophysical processes and geomorphological characteristics of cave passages that are responsible for very significant differences in the amplitude of fluctuations of radon and CO$_2$ levels.

2 Site and methodology

2.1 Site description

The Postojna Cave System (Fig. 1a), with its 20 570 m of known passages, is the second longest of 10 000 registered karst caves in Slovenia and one of the most visited show caves in Europe. The passages were formed at two main levels. The river Pivka sinks at the lower entrance to the cave at 511 m above sea level (m a.s.l.) (Fig. 1b) and the active river passages are mostly smaller than the higher ones. The river bed is composed mostly of gravels derived from the Eocene flysch. The entrance to the main, currently dry, passage is situated at 526.5 m a.s.l. and is 10 m high and 6 m wide. This entrance is also used as a tourist entrance. The cave passages have developed in an approximately 800 m thick layer of Upper Cretaceous bedded limestone situated
Table 1. Geomorphological characteristics of cave passages with measurement locations.

<table>
<thead>
<tr>
<th>Cave passage</th>
<th>Abbrev.</th>
<th>Level/m a.s.l</th>
<th>Tourist route</th>
<th>Geomorphological characteristics and sediments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lepe jame (Beautiful Caves)</td>
<td>BC</td>
<td>526</td>
<td>15 m away</td>
<td>– narrow solutional fissure, created along a fault plane</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>– loam in the fissure</td>
</tr>
<tr>
<td>Lowest point</td>
<td>LP</td>
<td>508</td>
<td>yes</td>
<td>– wide passage</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>– thin layer of loam</td>
</tr>
<tr>
<td>Pisani rov (Gaily Coloured Corridor)</td>
<td>GC</td>
<td>529</td>
<td>no</td>
<td>– 920 m long dead-end passage</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>– grey loam originating from weathered flysch rocks along the whole passage</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>– 145 cm thick profile at the end of the passage: fine-grained sediments (yellowish brown silts to clays with dark stains in the upper part showing cubic to columnar disintegration with Fe stains on the fractures) covering the collapse boulders and massive flowstone</td>
</tr>
</tbody>
</table>

between two important Dinaric faults (Šebela et al., 2010). The cave passages are mostly horizontal. The system has six known entrances; however, unknown connections to the surface along corrosively widened fissures undoubtedly exist. There is no forced ventilation in the cave, and air is only exchanged via natural air flow through the numerous cracks, passages and breathing holes (Gams, 1974) connecting the cave with the outside atmosphere. In the interior part the temperature is relatively stable at around 10°C. Measurements were performed at three locations (Fig. 1a). Geomorphological characteristics of the passages are summarised in Table 1. The first location is inside a narrow solutional fissure, created along a fault plane in the Lepe jame (Beautiful Caves, BC), 526 m a.s.l. and about 15 m off the guided tourist route. The second location is situated at the lowest point (LP) of the tourist route, at 508 m a.s.l., where the passage widens significantly. The distance between the two locations is about 250 m. The third measurement location lies at the end (529 m a.s.l.) of the 920 m long Pisani rov (Gaily Coloured Corridor, GC), which deviates from the main passage to the north. This passage is not a part of the tourist route. It terminates below the slopes of a collapse doline where the bottom is filled by sediments at 535 m a.s.l. (Šebela and Čar, 2000). Along the whole passage grey loam originating from weathered flysch rocks can be found. The roughly 145 cm thick profile of fluvial sediments situated at the end of GC consists of fine-grained sediments (yellowish brown silts to clays with dark stains in the upper part showing cubic to columnar disintegration with Fe stains on the fractures) covering the collapse boulders and massive flowstone (Zupan Hajna et al., 2008). The deepening of the collapse doline interrupted the continuation of GC towards the north (Šebela and Čar, 2000). The smallest thickness of the cave ceiling is about 30 m. Between BC and Črna jama (Black Cave) (Fig. 1a), an artificial tunnel is closed by doors that are opened only during occasional tourist visits. The ventilation from Črna jama does not have a significant impact on our monitoring locations.

2.2 Instrumentation

At all three measurement locations, radon activity concentration ($C_{\text{Rn}}$) was measured continuously once an hour from March 2011 to September 2012. Radon measurements at BC were performed using a Radim 5 monitor (SMM Company, Czech Republic), which is mainly designed for radon measurements in indoor air. It determines radon concentration by measuring gross alpha activity of the radon decay products $^{218}\text{Po}$ and $^{214}\text{Po}$, collected electrostatically on the surface of a semiconductor detector. The lower limit of detection is about 50 Bq m$^{-3}$ and the sampling frequency is twice an hour. Hourly averages are calculated for further data evaluation in order to correspond to measurements performed using other types of measuring devices.

At the LP and GC locations, Barasol probes (MC-450, ALGADE, France) were used for radon measurements. The probe is primarily designed for radon measurements in soil gas, and it therefore has a higher lower limit of detection (about 500 Bq m$^{-3}$) than the Radim 5 monitor. It gives radon concentration based on alpha spectrometry of radon decay products in the energy range of 1.5 MeV to 6 MeV using an implanted silicon detector. The detector sensitivity is 50 Bq m$^{-3}$ per 1 imp h$^{-1}$ with a sampling frequency of once an hour. It also records temperature and relative atmospheric pressure.

In both instruments data are stored in the internal memory and are transferred to a personal computer. The instruments
are checked regularly, using a portable AlphaGuard radon monitor (Saphymo, Germany) as a reference instrument, and were calibrated in the Radon Chamber at the Henryk Niewodniczański Institute of Nuclear Physics, Polish Academy of Sciences, Kraków, Poland (Kozak et al., 2009).

Continuous measurements of CO\(_2\) concentrations were performed in two periods, spring and summer 2012, at the GC location. CO\(_2\) is measured along with the other microclimatic parameters within broader monitoring of cave micrometeorology. A Vaisala Carbocap CO\(_2\) module GMM 221 with a measurement of interval 0–7000 ppm and an accuracy range of 1.5\% is used for the task. It is connected to a data logger with a sampling interval of 10 min. Probe operation is based on measurements of infrared absorption by CO\(_2\).

Atmospheric parameters (temperature, barometric pressure and relative humidity) were measured continuously by a DL-180THP data logger (Vollcraft, Germany) in front of the tourist entrance. Daily height of rainfall was additionally provided by the Postojna weather station of the Slovenian Environment Agency (Ministry of Agriculture and Environment of the Republic of Slovenia).

### 3 Results

On a long timescale, radon concentration in the Postojna Cave exhibits annual cycles with high summer and low winter values (Gregorić et al., 2011), reflecting the ventilation pattern. However, amplitudes of fluctuation of radon levels differ from point to point. Annual mean radon concentration recorded at LP is 3255 ± 1190 Bq m\(^{-3}\) and at BC, 2315 ± 1019 Bq m\(^{-3}\), which is in the same range as values recorded at the Great Mountain location (Gregorić et al., 2011). On the other hand, radon concentration at GC is up to ten times higher than in other cave passages (Fig. 2) with the annual mean of 25 020 ± 12 653 Bq m\(^{-3}\) (Table 2).

At LP radon concentration follows significant annual cycles, with average values of 1742 ± 666 Bq m\(^{-3}\) in winter months (period D, Table 2) and constantly high summer concentrations of 4130 ± 370 Bq m\(^{-3}\) (average of summer 2011 (B) and 2012 (F), Table 2). The biggest variations between minimum and maximum values are, as expected, characteristic of transitional periods in spring and autumn.

Situated in a side branch of the main passage, characterised by cracked and faulted rocks and possibly connected to an unknown passage, the BC location exhibits a far more variable radon concentration pattern, not only on an annual scale but also from year to year. Changes of behaviour of radon concentration were, for example, observed after floods in 2010 (Gregorić and Vajpotič, 2011). Concentration during the period discussed in this paper shows a different seasonal pattern from the LP location, with the highest values in spring and autumn, reaching up to about 5500 Bq m\(^{-3}\) (Fig. 3), whereas the concentration in summer usually remains below 4000 Bq m\(^{-3}\). Minimum radon levels are still found in the cold period of year (period D, Table 2). The radon concentration at BC exhibits high variation throughout the whole measurement period. Short periodic cycles (24 h) can be observed at BC in the transitional periods, spring and autumn, as a result of the changing ventilation regime during cold nights, when outside temperature drops below cave temperature (10\(^\circ\)C), and warm sunny days with outside temperature above cave temperature.

The GC location is, in contrast to LP and BC, characterised by much higher radon levels in summer, reaching up to 44 500 Bq m\(^{-3}\), with a mean summer value of 35 857 ± 3259 Bq m\(^{-3}\) in 2011 and 33 038 ± 3015 Bq m\(^{-3}\) in 2012 (Table 1). During the cold part of the year, however, radon concentration in the periods of active ventilation drops below 500 Bq m\(^{-3}\) – the lowest of all three measurement locations (Fig. 3). The mean value in winter is 8 684 ± 7 648 Bq m\(^{-3}\).

CO\(_2\) levels at the GC location show similar seasonal characteristics in spring and summer as radon levels, with mean values of 1522 ± 614 ppm and 4689 ± 294 ppm, respectively. Higher fluctuation of CO\(_2\) levels is observed in spring, which represents a transitional period in terms of the ventilation regime. During this period high correlation (\(R^2 = 0.91\)) is observed between CO\(_2\) and \(^{222}\)Rn concentrations, pointing to a common driving force (i.e. cave ventilation) (Fig. 4). In summer, CO\(_2\) levels remain high, consistent with decreased ventilation. However, different behaviour of these two gases is observed, reflecting in a weaker correlation (\(R^2 = 0.69\)) than in spring.

#### 3.1 Spatial differences in radon levels

Linear correlation of radon levels between the GC and LP locations, with coefficient of determination (\(R^2\)) 0.85, can be observed throughout the annual cycle, as seen in Fig. 5a and b. From Fig. 5a, where the point colour represents daily mean outside temperature, it can be noted that the lowest radon levels are usually observed at outside temperatures between 0 and 10\(^\circ\)C, while the highest radon levels are typical for days with daily mean outside temperatures around 15\(^\circ\)C. On extremely cold winter days, when \(T_{\text{out}}\) remains below 0\(^\circ\)C for several days, slightly higher radon levels are observed at both locations (GC and LP). Conversely, during extremely warm summer days (\(T_{\text{out}} > 23\,\text{\(^\circ\)}\)), radon levels slightly below maximum are observed. Atmospheric pressure (Fig. 5b), on the other hand, does not show a significant role in controlling radon levels in the cave. Therefore, although the amplitude of fluctuation of radon concentration at GC is some orders of magnitude higher than at LP, it is obvious that both locations are subject to a similar ventilation pattern.

In contrast to GC and LP, the measurement location at BC shows slightly different behaviour, with radon levels being roughly in the same range as at the LP location. A moderate correlation (\(R^2 = 0.46\)) is observed between radon concentrations at BC and GC in winter months, when fresh outside
Fig. 2. Time series of radon concentration at three measurement locations (Pisani rov – $C_{\text{GC}}^{\text{Rn}}$, Lepe jame – $C_{\text{BC}}^{\text{Rn}}$ and the lowest point – $C_{\text{LP}}^{\text{Rn}}$) and CO$_2$ concentration recorded at the GC location. Time series of main atmospheric parameters controlling radon concentration in the cave: outside air temperature ($T_{\text{out}}$), air pressure ($P_{\text{out}}$) and daily height of rainfall ($h_r$). Radon concentration and atmospheric parameters are expressed in hourly values, 24-h weighted average smoothing is applied to radon concentration and temperature; rainfall is expressed as absolute daily values.

air enters the cave through the large tourist entrance, and warmer cave air rises through narrow vertical cracks and channels and exits into the outside atmosphere. Radon concentration at BC ($C_{\text{BC}}^{\text{Rn}}$) is higher than at LP ($C_{\text{LP}}^{\text{Rn}}$) in autumn and lower in spring (Fig. 3). The negative correlation ($R^2 = 0.26$) observed when daily mean $T_{\text{out}}$ exceeds 20°C (Fig. 5c) indicates that other parameters take control of ventilation at BC and the air connection between LP and BC is cut off.

3.2 Influence of outside atmospheric parameters in governing radon levels in the cave

Correlation between radon levels at each location with outside atmospheric parameters – outside temperature ($T_{\text{out}}$), pressure ($P_{\text{out}}$) and accumulated rainfall in the last 7 days ($h_{r-7}$) (Table 2) – reveals that $T_{\text{out}}$ has the highest influence on radon levels. The response of fluctuation of radon concentration on $T_{\text{out}}$ is comparable at the GC and LP locations in all periods except the summer months (periods B and F), when radon concentration at BC and LP decreases with increasing $T_{\text{out}}$.

Correlation between $P_{\text{out}}$ and $P_{\text{cave}}$ (pressure in the cave) was discussed by Šebela and Turk (2011) in the study of climate characteristics of Postojna Cave, where simultaneous pressure variations at the surface and at three locations in the underground was shown. Therefore, air pressure may in this case have an influence on radon concentration only by the effect of “barometric pumping” of radon from the pore space (Perrier and Richon, 2010). However, no significant influence of $P_{\text{out}}$ can be observed, possibly due to the obscured effect of pressure changes by airflows driven by temperature gradients.

The most sensitive measurement location for rainfall with respect to radon concentration seems to be BC, where significant changes of radon levels after heavy flooding in 2010 (Gregorič and Vaupotič, 2011) provide further evidence for high sensitivity to precipitation. Rainfall acts in two ways at this location: firstly, by reducing the air connection between the outside atmosphere and the cave atmosphere due to saturation of soil pores on the surface (thus causing an increase of radon concentration) and, secondly, by reducing radon exhalation from rock surface and cracks (decrease of radon concentration shown about a week after heavy rainfall) (Gregorič and Vaupotič, 2011). On the other hand, no significant correlation with rainfall was observed for the GC location.

3.3 Estimation of radon source

The ventilation characteristics of a cave can be reflected in the fluctuation of radon concentration provided the turnover time is shorter than about five mean lives of $^{222}\text{Rn}$.
(approximately four weeks) (Kowalczyk and Froelich, 2010). Cave ventilation represents the proportion of cave air exchanged per time unit. If the cave atmosphere is just pulled back and forth due to an alternating ventilation regime during transitional periods in spring and autumn, there is no air exchange in the deeper parts of the cave. This period can be called a stagnant ventilation period and could be the reason for higher radon levels at BC in spring and autumn. By contrast, ventilation is considered active when the cave remains in one ventilation regime for long enough in comparison to the time that air is retained in the cave (i.e. residence time) (Faimon et al., 2011).

According to Wilkening and Watkins (1976) and Perrier et al. (2004), the temporal evolution of radon concentration at a given location in the cave can be described as

$$\frac{dC_{Rn}^{cave}}{dt} = \frac{S}{V} \Phi - \lambda C_{Rn}^{cave} - v(C_{Rn}^{cave} - C_{out}),$$  \hspace{1cm} (1)

where \(S\) (m²) and \(V\) (m³) are, respectively, the total surface area and volume of the cave; \(\Phi\) (Bq m⁻² h⁻¹) represents the radon exhalation rate from the rock surface; \(\lambda\) (h⁻¹) is the radioactive decay constant of \(^{222}\)Rn; \(v\) (h⁻¹) is the cave ventilation rate; and \(C_{out}\) (Bq m⁻³) is the radon concentration in the outside air. Note that the radon concentration in the outside air is on the order of tens of Bq m⁻³ and thus negligible in comparison to the radon concentration in the cave air.

As the total area and volume of the cave are very hard to determine without exact measurements, we considered the radon source for different locations separately:

$$\Phi_{ch} = \frac{S_{ch}}{V_{ch}} \Phi,$$ \hspace{1cm} (2)

where \(\Phi_{ch}\) (Bq m⁻³ h⁻¹) is the radon source in a specific chamber, either GC or LP; \(S_{ch}\) (m²) and \(V_{ch}\) (m³) are, respectively, the surface area and volume of the chamber; and \(\Phi\) (Bq m⁻² h⁻¹) represents the radon exhalation rate from the rock surface.

The radon source can be estimated for the summer period at the GC and LP locations when radon concentration remains constantly high – around 40 kBq m⁻³ at GC and 4 kBq m⁻³ at LP location. If we consider the summer period as a stagnant ventilation period and a constant radon concentration, Eq. (1) can be transformed to

$$\Phi_{ch} = \lambda C_{Rn,\max}^{ch},$$ \hspace{1cm} (3)

where \(C_{Rn,\max}^{ch}\) (Bq m⁻³) represents the highest radon concentration in a specific chamber. This means that in order to maintain 40 kBq m⁻³ (4 kBq m⁻³) during stable conditions in summer at the GC (LP) location, the radon source should not be less than about 300 Bq m⁻³ h⁻¹ (30 Bq m⁻³ h⁻¹).

Winter ventilation should be similar at the BC and LP locations, while during other periods of year the BC location

<table>
<thead>
<tr>
<th>period</th>
<th>min</th>
<th>(C_{Rn}) / Bq m⁻³</th>
<th>max</th>
<th>AM</th>
<th>SD</th>
<th>(R(C_{Rn}, T_{out}))</th>
<th>(R(C_{Rn}, P_{out}))</th>
<th>(R(C_{Rn}, h_{t-7}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>LP</td>
<td>A Mar–May 2011</td>
<td>1138</td>
<td>4636</td>
<td>3370</td>
<td>1154</td>
<td>0.30</td>
<td>0.06</td>
<td>0.09</td>
</tr>
<tr>
<td>B Jun–Aug 2011</td>
<td>3662</td>
<td>5030</td>
<td>4244</td>
<td>302</td>
<td>-0.30</td>
<td>-0.01</td>
<td>0.36</td>
<td></td>
</tr>
<tr>
<td>C Sep–Nov 2011</td>
<td>1608</td>
<td>4826</td>
<td>3653</td>
<td>850</td>
<td>0.60</td>
<td>-0.15</td>
<td>0.30</td>
<td></td>
</tr>
<tr>
<td>D Dec 2011–Feb 2012</td>
<td>1019</td>
<td>4019</td>
<td>1742</td>
<td>666</td>
<td>-0.55</td>
<td>0.18</td>
<td>-0.16</td>
<td></td>
</tr>
<tr>
<td>E Mar–May 2012</td>
<td>1275</td>
<td>4729</td>
<td>3164</td>
<td>980</td>
<td>0.57</td>
<td>0.31</td>
<td>0.26</td>
<td></td>
</tr>
<tr>
<td>F Jun–Aug 2012</td>
<td>2874</td>
<td>5389</td>
<td>4022</td>
<td>440</td>
<td>-0.40</td>
<td>-0.17</td>
<td>0.52</td>
<td></td>
</tr>
<tr>
<td>BC</td>
<td>A Mar–May 2011</td>
<td>185</td>
<td>4909</td>
<td>2315</td>
<td>1019</td>
<td>0.30</td>
<td>-0.06</td>
<td>0.09</td>
</tr>
<tr>
<td>B Jun–Aug 2011</td>
<td>937</td>
<td>3636</td>
<td>2039</td>
<td>654</td>
<td>-0.52</td>
<td>-0.18</td>
<td>0.23</td>
<td></td>
</tr>
<tr>
<td>C Sep–Nov 2011</td>
<td>937</td>
<td>4865</td>
<td>2925</td>
<td>804</td>
<td>-0.24</td>
<td>0.19</td>
<td>0.43</td>
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<tr>
<td>D Dec 2011–Feb 2012</td>
<td>209</td>
<td>3825</td>
<td>1543</td>
<td>828</td>
<td>0.30</td>
<td>-0.06</td>
<td>0.47</td>
<td></td>
</tr>
<tr>
<td>E Mar–May 2012</td>
<td>260</td>
<td>4059</td>
<td>1853</td>
<td>852</td>
<td>-0.14</td>
<td>-0.01</td>
<td>-0.27</td>
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</tr>
<tr>
<td>F Jun–Aug 2012</td>
<td>1093</td>
<td>4428</td>
<td>2282</td>
<td>710</td>
<td>-0.41</td>
<td>-0.04</td>
<td>0.40</td>
<td></td>
</tr>
<tr>
<td>GC</td>
<td>A Mar–May 2011</td>
<td>344</td>
<td>44578</td>
<td>25020</td>
<td>12653</td>
<td>0.47</td>
<td>0.15</td>
<td>0.08</td>
</tr>
<tr>
<td>B Jun–Aug 2011</td>
<td>25861</td>
<td>43872</td>
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is subject to mixing of different air currents, carrying cave air from different passages with various radon levels. On the other hand, occasional strong winter ventilation is expected at the GC location, in order to decrease radon concentration to lower levels than at other two locations (Fig. 3a).

4 Discussion

Based on the morphology of the cave and several openings at different altitudes, we can characterise the Postojna Cave as a dynamic cave which is ventilated throughout the year. Different parts of the cave, however, exhibit different ventilation patterns based on local geomorphology. As also reported in the study of radon concentration in the Great Mountain chamber in this cave (Gregorič et al., 2011) and for several other caves (De Freitas, 2010; Spötł et al., 2005; Wilkening and Watkins, 1976), two main ventilation regimes can be distinguished in the main passage of the Postojna Cave. The measurement locations considered in this study, i.e. LP, BC and GC, have unique characteristics. Comparing the LP and BC locations, situated in the central part of the cave...
cave in galleries extending from the main passage, LP lies at 508 m a.s.l., while BC, located at 526 m a.s.l., has a specific geomorphology and microlocation – a narrow corrosively widened fissure, located 15 m from the central part of the passage, caracterised by numerous, more or less pronounced fault planes. Significant differences can therefore be found in their ventilation pattern and radon fluctuation, while the annual mean radon concentration is roughly the same for both locations. GC, on the other hand, is situated in a dead-end passage off the main passage and is therefore characterised by a higher stability of the cave atmosphere. The entrance is located below the ceiling of the main passage, and the measurement location is situated at 529 m a.s.l.

During the winter period, the difference in density between the cold outside air and warm cave air triggers a rise of warmer cave air, which exhales to the outside atmosphere through vertical cracks and fissures, thus giving space to cold, denser outside air, which enters the cave through lower entrances. This so called “chimney effect” predominates throughout the winter and can be observed through the entire cave system. On the basis of relatively good correlation between radon concentration at all three locations (Fig. 5) in winter, as well as the good correspondence between CO$_2$ and radon at the GC location (Fig. 4), it may be assumed that a current of fresh outside air comes in from the tourist entrance and river entrance, is lifted upwards, and leaves the cave through vertical cracks and openings. The artificial tunnel that connects Lepe jame with Črna jama is closed off by a door, thus preventing the airflow in direction from Črna jama. Therefore, both $^{222}$Rn and CO$_2$ concentrations at GC tend to decrease toward their atmospheric levels ($\approx 10$–$30$ Bq m$^{-3}$) for $^{222}$Rn and $\approx 380$ ppm for CO$_2$) during periods with an active chimney effect, whereas radon levels at LP, located lower than the tourist entrance, still remain higher than at GC. Owing to the remote location of BC, a slower increase of $C_{BC}^{222}$Rn in comparison to $C_{LP}^{222}$Rn is observed in spring, and a slower decrease in autumn (Fig. 3). The chimney effect is most pronounced at GC when $T_{out}$ is lower than the cave temperature ($< 10^\circ$C), but remains above 0°C. Below that temperature, the snow layer and water freezing in the upper few centimetres of the soil layer above the cave prevent the exhalation of cave air and provoke cave air isolation. Consequently, an increase of radon concentration (and CO$_2$) is observed first at GC (Fig. 5a and c) and then, at lower temperatures, also at BC and LP.

Ventilation decreases in summer and is more pronounced at the beginning of the main passage, where fresh, warmer outside air enters the cave below the ceiling and cold cave air is swept out of the cave near the bottom. When the temperature difference of outside and cave air reaches the critical point, around 10°C (at mean daily $T_{out} > 20^\circ$C), warm air enters the cave system through known and unknown entrances, fissures and cracks at higher elevations and leaves the cave through lower entrances. There is frequent strong ventilation from the BC location toward the main passage in summer (Šebela and Turk, 2011). Behind the BC location, fresh air could enter through subvertical corrosively widened fissures, possibly through some undiscovered cave chambers. On the other hand, summer ventilation has only a small effect on the LP location, and is almost undetectable at the GC location.

The results show that the intense winter ventilation regime takes control over the behaviour of gases in the cave, resulting in a high correlation between different locations as well as a good correspondence in the fluctuation of different gases (e.g. CO$_2$ and $^{222}$Rn). On the other hand, together with diminishing effect of ventilation in summer, the response of gases to other processes becomes apparent (e.g. changes of radon exhalation from the rock surface and from deeper parts of the Earth’s crust, and changes of the flux of CO$_2$ from the epikark).

The question of high differences in the spatial distribution of gases cannot, however, be explained simply by the ventilation characteristics of the cave, although it is obvious that the atmosphere in the Pisani rov is more stable than in the main cave passages. Stable conditions are reflected additionally in cave air temperature, with the highest daily amplitude of 0.2°C at the LP location and only 0.03°C at the GC location. A rapid increase in both radon and CO$_2$ concentrations during decreased ventilation points to strong radon and CO$_2$ sources, which could be the sum of different contributions: (i) flux of gases from the ground in faulted rocks, (ii) cave loam sediments as a radon source and (iii) deep soil layer in the collapse doline – on the surface where the Pisani rov terminates – as a source of radon and CO$_2$. The process (i), known as advection of geogas (Etiope and Martinelli, 2002), is a common source of both radon and CO$_2$.

![Fig. 5. Correlation between radon concentration at GC ($C_{RC}^{GC}$) and LP ($C_{LP}$) depending on: (a) outside air temperature (colour scale) and (b) atmospheric pressure (colour scale) and correlation between radon concentration at GC and BC ($C_{BC}^{222}$Rn) depending on: (c) outside air temperature (colour scale) ($R^2$ for summer and winter period) and (d) atmospheric pressure (colour scale). Daily mean values are used.](nat-hazards-earth-syst-sci.net/13/287/2013/)
and can act as a major radon source in fractured rocks. CO$_2$ (together with other gases) works as a carrier gas for radon (Kristiansson and Malmqvist, 1982). While numerous fault zones are found along the Pisani rov (Sebela, 1992), this process may have a substantial role in controlling the concentration of radon and CO$_2$ in this passage. The second source of radon (ii) are rock surfaces and cave sediments, from where radon diffuses to the cave air. While limestone has basically the same characteristics in all cave passages, a large amount of cave sediments at the GC location makes this part different compared to other cave passages. Clays, especially when radium is absorbed to clay minerals and ferrous oxides, may have a high emanation capacity (Miklyaev and Petrova, 2011), which determines the amount of free radon in the geologic medium. Considering the stable atmospheric conditions in the cave throughout the year, the radon emanation from clays should be constant. Process (iii) could be expressed when outside air temperature is higher than cave temperature and the chimney effect stops. Airflow in summer is decreased as the fresh outside air moves downwards through narrow fissures, soil and rock matrices. Vertical connection of the cave and outside air in summer, responsible for decreasing radon concentration at BC (and partially LP), is influenced at the GC location by the deeper soil layer in the bottom of the collapse doline. The soil in such areas on carbonate rocks (e.g. dolines, sinkholes) could become enriched with natural radionuclides (also $^{226}$Ra as a radon source) due to their migration by water, as presented in the study of natural radionuclides in Slovenian soils (Gregorič et al., 2012). The soil layer on the surface can therefore have high radon potential. Furthermore, the soil layer can be also an efficient CO$_2$ source due to biological activity, with CO$_2$ entering the cave environment by degassing from percolating water and gravity seepage through rock fractures in the thin cave ceiling. The above mechanisms, with the emphasis on process (iii), are the main reason for significant difference of gas concentrations between the Pisani rov and the main cave passages during the summer ventilation regime. On the other hand, the difference in the behaviour of radon and CO$_2$ in summer, observed by decreased correlation between them, can be explained by processes (ii) and (iii).

5 Conclusions

Investigations of radon and CO$_2$ levels carried out at three geomorphologically different locations in the Postojna Cave uncovered significant differences in the spatial distribution of radon concentration, and a high amplitude of fluctuation of radon and CO$_2$ concentrations in the Pisani rov, a dead-end passage in the cave. Concentrations of radon measured in this passage are, according to the published data, one of the highest concentrations measured in limestone caves. This research enables better understanding not only of the ventilation characteristics of Postojna Cave, but also of radon and CO$_2$ sources and mechanisms leading to very high concentrations of both gases. Significant differences in radon concentration between the main passages of the cave (up to 5400 Bq m$^{-3}$) and the Pisani rov (up to 44 600 Bq m$^{-3}$), as well as high CO$_2$ concentration at the GC location, lead to the conclusion that ventilation itself could not be the only reason for the extremely high variability in the spatial distribution of radon and CO$_2$ in Postojna Cave. Taking into account the geomorphological characteristics of cave passages, a substantial contribution to radon and CO$_2$ concentration may be represented by the deeper soil layer above this passage, formed at the bottom of the collapse doline – this effect being additionally emphasised by the thin cave ceiling. Additional radon sources with very low variability may be the clay sediments which are present along the whole of the Pisani rov.

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