A geochronological approach for cave evolution in the Cantabrian Coast
(Pindal Cave, NW Spain)

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with 5 figures and 2 tables

Summary. Some of the oldest speleothems in the North Cantabrian Coast (Spain) are reported for the first time in this work. Pindal Cave is developed at 24 m above sea level, in a karstic massif reaching its highest surface in a marine terrace (rasa) located at 50–64 m above the present sea level. Several phases of evolution were previously recognized into the cave, including block collapse of the roof, episodic flooding and detrital sedimentation, and chemical precipitation of at least four speleothem generations over both alluvial and collapse deposits. Three of these speleothem generations have been dated by U/Th. The first generation yielded ages from 124.2 ± 1, 5 ka BP to 73.1 ± 0.9 ka BP, giving a minimum age for the main detritic sediments in the cave. The second one is not dated. The third generation gives an age of 371 ± 0.4 ka BP (mathematically corrected to 2.7 ± 0.9 ka BP), while the youngest generation, with actively growing stalagmites in the cave, basal ages of 200 years BP are estimated by counting annual laminae. The data suggest a tentative maximum elevation rate close to 0.2 mm/yr for the Cantabrian Margin in this area, although further chronological studies will be needed to check this hypothesis.


Résumé. Dans ce travail sont présentées des données géochronologiques obtenus d’un des plus vieux spéléothèmes dans la Côte Cantabrique de l’Espagne. La Grotte de Pindal se trouve à 24 m d’altitude sur la mer, dans un massif karstique culminé par une ancienne surface d’abrasion marine élevée jusqu’à 50–64 m d’altitude. La recherche géomorphologique accomplit préalablement dans la cavité nous avait permis différents phases d’évolution, avec procès de collapse du plafond, des épisodes d’inondation et entrée des galets fluviaux, et de précipitation chimique de spéléothèmes qui peuvent être groupés dans quatre phases. Trois phases d’spéléothèmes ont été datées par U/Th. L’âge minime de la première phase se trouve entre 124,2 ± 1,5 ka BP et 73,1 ± 0,9 ce qui apporte aussi des âges minimes pour les dépôts fluviales les plus anciens. Des données géochronologiques pour la seconde phase n’ont pas pu être obtenues. La troisième phase est datée en 371 ± 0,4 ka BP (corrigée à 2,7 ± 0,5 ka BP). Le contage de laminae sur une stalagmite
1 Introduction

Geomorphological research along the Cantabrian Coast from the beginning of the 20th century described the presence of marine terraces called “rasa” (FLOR 1983, MARY 1983). Marine terraces are present along more than 200 km along the Asturias Shore and appear mostly on Paleozoic bedrock. They reach elevations of 260 m in the East and drop to 60 m in the West reflecting a differential uplift along the coast related to the Cenozoic reactivation of former faults (ALVAREZ-MARRÓN et al. 2003, 2005). Therefore, tectonic uplift is clearly involved in the origin of these terraces, although the timing of uplift and the interaction between climatic, eustatic and tectonic factors in this area still remains unknown.

In the eastern area of Asturias, the marine terraces are well preserved on quartzitic bedrock of Ordovician age as well as on limestone bedrock of Carboniferous age. Karstic massifs showing important exokarstic and endokarstic features are well developed beneath these marine terraces. A good example of terraces appearing at 50–64 m above sea level is the one where Pindal Cave is developed. Pindal Cave shows important and well preserved Paleolithic paintings and geomorphological and environmental research has been carried out there in recent years (JIMÉNEZ-SÁNCHEZ et al. 2002, 2004, 2005, STOLL et al. 2005a, b). However, a lot of questions remain unknown, especially related to the Quaternary evolution of the cave together with the prehistoric occupation.

Geomorphological research projects developed in the Pindal Cave allowed us to propose a qualitative model of evolution of the cave and the surrounding areas (JIMÉNEZ-SÁNCHEZ et al. 2002, 2004). Preliminary palaeoclimatological research (STOLL et al. 2005a, b), together with geochronological data obtained from speleothems of Pindal Cave (JIMÉNEZ-SÁNCHEZ et al. 2005) allowed us to establish the evolution of the karst and also to give a chronological reference for the evolution of the Cantabrian Coast in this area.

The aim of this paper is to show the first radiometric results obtained by U-Th isotopes in the Pindal Cave. These data provide the first absolute age constraints on the genesis and evolution of Pindal Cave, which may be representative of karst development in similar geomorphologic settings in the central Cantabrian Margin in northern Spain. The results also shed light on the relative uplift rate of the marine terrace (50–64 m) which hosts the cave and imply a strong role of neotectonics in terrace uplift. Finally, the results also suggest the coupling of speleothem growth rates and climatic cycles.

2 Geological and geomorphological setting

Pindal Cave is located to the East of Asturias (NW Spain), 4°30’W, 43°23’N (Fig. 1). The Cave is 590 m long (314 m open to tourist use, where this study has been developed), and presents an E-O and 110° N (ONO-ESE) trending. The maximum altitude is located to the W (30 m above the sea, +6 m above the entrance) and the minimum altitude is located to the E (6 m above the
sea level, -18 m under the entrance). The width of the cave ranges from 2 to 50 m, while the vertical distance from ceiling to bottom ranges from 1.5 m to 11 m (Obeso et al. 1996). From a geological point of view, it is developed in Carboniferous bedrock, Barcaliente Limestone, located in the Ponga-Cuera Unit of the Cantabrian Zone (Martínez García et al. 1980, Marquín 1989).

Previous geomorphological research in the area (Jiménez-Sánchez et al. 2002, 2004, 2005) showed that the landscape results from fluvial, mass wasting, karstic and marine processes (Fig. 2a) with the outstanding presence of two erosion marine surfaces ("rasas"): Pimiango (125–170 m) and Pindal (50–64 m) (Fig. 2b). The origin of the Cave, developed in a karstic massif reaching its highest surface in Pindal Rasa, is mainly controlled by two sets of subvertical fractures trending E-W (F1), the bedding planes, dipping 70° to the North and 3 joint systems: D1 (subhorizontal or 3° dipping to the E); D2 (N35E and dipping 35° to the SE) and D3 (N15E and dipping 70° to the NW).

Geomorphological research previously developed in the cave showed the presence of fluvio-karstic, gravity, chemical precipitation, anthropogenic and other features of mixed origin, whose spatial distribution is presented in the sketch of Fig. 3.

**Fluvio-karstic features** are represented by roof pendants, a fluvial channel, phreatic tubes and paleochannels, and 6 levels of alluvial deposits. Paleochannels are associated with deposits of the highest alluvial levels and are located at 0.5 m, 1.6 m, 3.5 m and 5.5 m above the present channel. These deposits are interpreted as the result of denudation by torrential systems developed on the quartzitic range of Pimiango Rasa, located at 125–170 m above sea level (Fig. 2b).
Fig. 2. a) Geomorphological setting of Pindal Cave (modified from Jiménez-Sánchez et al. 2002); b) Ideal cross section from S to N in the area, showing the relationships between the geomorphological features of both marine terraces (Pimiangos Rasa and Pindal Rasa) and the bedrock lithology.
Speleothems have been classified in *dripstone, flowstone and complex forms* (Fig. 3). According to morphometric and superposition criteria, four generations of speleothems have been distinguished in the cave (Fig. 4): 1 – Stalagmite and complex speleothems (e.g. flowstones more than 2 m wide and 3 m high covering fluvial detritic sediments and limestone blocks fallen down from roof and walls, 2 – Speleothems of decimetric to metric size covering speleothems from the first generation, 3 – Speleothems of decimetric to centimetric size covering the former ones and 4 – Actively growing speleothems, with porous white calcite and typically 2–5 cm in height, growing over speleothems of previous generations. It is interesting to remark that the first phase includes all the flowstones plus some stalagmites, but geomorphological evidence suggests that flowstones were only created during the first generation.

Finally, evidence of processes of instability by *gravity action* has been described affecting ceiling as well as the cavity floor. Isolated blocks and accumulations of blocks coming from ceiling fracturing along the previously described discontinuities are present inside the cave. Evidences of cave floor subsidence or collapse are also documented as metric scarps, incisions, closed depressions as well as rotated speleothems. Gravity processes are inferred to be previous, synchronous and posterior to chemical precipitation episodes, since deposits include broken and fractured speleothems. The instability of the cave floor is probably related to the development of a lower active level whose full length is probably greater than the one identified by the speleological studies.

The present dynamics of the cave are controlled by vadose conditions with infiltration and dripwater flow, chemical precipitation, collapse processes or the roof and sporadic flooding of the channel inside the cave. Water circulation at a lower active level 18 m under the entrance (6 m over the sea) is also supported by speleological evidence. A rapid downcutting of this active level is inferred from scarps, fracture and collapse development in deposits on the cave floor, suggesting that uplift is an active process at present.

3 Methodology

Speleothems, calcite wall coatings and other forms of authigenic carbonate are the material of choice for U-series dating in the cave environment because they trap uranium at the time of deposition and generally behave as a closed system to U and Th isotopes thereafter.

Four samples of speleothem from three of the four generations were taken for analysis (Fig. 3). The samples 1 and 2 were taken from a flowstone of the first generation. This flowstone is 3 m high and 3.5 m wide, although is included in a flowstone accumulation extending more than 250 m$^2$ over the cave floor. For environmental reasons, the samples were collected from a large flowstone block which had separated from the main flowstone, providing a cross sectional exposure of the outer 70 cm (Fig. 3b, 3c). The sample 1 was taken at the older part of this fragment and the sample 2 was taking close to the surface of this fragment, corresponding to the top of the speleothem (Fig. 3c). Therefore, these samples can provide a minimum age for the oldest generation of speleothems in the cave. Complete stalagmites were sampled from generations 3 (sample 3, 15 cm x 4 cm) and 4 (sample 4, 8 cm x 3 cm) in order to give a geochronological approach for these two generations of speleothems.

The actively growing stalagmite of phase 4 (sample 4) was sliced and polished and the thickness of laminations was estimated from high-resolution scanned digital images. U-series analysis
Fig. 3. Geomorphological map of the Pindal Cave (modified from Jiménez-Sánchez et al. 2002). a) General view of speleothems from generations 1, 2 and 4; b) Flowstone of generation 1 (note the fracture surface on the top of the picture); c) Location of samples 1 and 2 in the block separated from the outer part of the flowstone depicted in 2b.

was applied to samples 1, 2, 3. These samples were lightly ground, dissolved in strong mineral acids, and U and Th isotopes were isolated by ion-exchange chromatography using the procedure described by Bischoff et al. (1988). Samples were dissolved and spiked with $^{230}\text{U}$ and $^{229}\text{Th}$ and passed through an additional anion exchange column further to remove Fe and other cations. Th was stripped from the column using 6N HCl, and U was then stripped using Teflon-distilled H$_2$O. 10 μl of 0.15N H$_3$PO$_4$ was added to the Th fraction. Both Th and U fractions were evaporated to dryness, converted to nitrate form using 8N HNO$_3$, and loaded onto the sample side of a double-Re filament for analysis on a Finnigan MAI 261 solid-source mass spectrometer (thermal ionization mass spectrometry, or TIMS) configured with adjustable faraday collectors and an ion counter. U isotopes were measured using a peak-hopping mode, using the sequence $^{234}\text{U}, ^{238}\text{U}, ^{235}\text{U}, ^{236}\text{U}, ^{238}\text{U}$. Ion currents on the $^{238}\text{U}$ beam were generally greater than 10-12 a. Th isotopes were measured using a peak-hopping mode on the ion counter. Count rates for the $^{229}\text{Th}$ and $^{230}\text{Th}$ peaks were generally greater than 100 cps and occasionally reached 500 cps which represents excellent ionization efficiency for the 1 to 5 pg Th loads measured here. Age equations and half-lives for U-series age calculations are given in Bischoff et al. (1995).
4 Results and discussion

Table 1 shows the radiometric results for samples 1, 2 and 3. Samples from speleothems of the generation 1 (1 and 2) gave respective results of 124.2 ± 1.5 ka BP and 73.1 ± 0.9 ka BP, $^{230}\text{Th}/^{232}\text{Th}$ equal to 14, indicative of little detrital contamination with $^{230}\text{Th}$. The speleothem from the third generation yielded an age of 3.71 ± 0.4 ka BP. The ratio $^{230}\text{Th}/^{232}\text{Th}$ is 4, indicating a detrital contamination that biases the date to older ages. Therefore, the date was mathematically corrected to 2.7 ± 0.5 kyr (see explanation in Table 1).

Table 1. U-series analyses by Thermal ionization mass spectroscopy (TIMS), and derived dates of speleothems in the Pindal Cave, NW Spain. Half-lives of $^{234}\text{U}$ and $^{230}\text{Th}$ taken from Cheng et al. (2000). Correction for detrital contamination, assuming average crustal composition for the detritus, that all the $^{232}\text{Th}$ is in the detrital fraction and that $^{230}\text{Th}/^{232}\text{Th} = 1$. Only sample 3 showed sufficient detrital contamination to correct the date (see criteria in Bischoff & Fitzpatrick 1990).

<table>
<thead>
<tr>
<th>Sample</th>
<th>USGS Lab</th>
<th>U (ppm)</th>
<th>$^{234}\text{U}/^{238}\text{U}$</th>
<th>$^{230}\text{Th}/^{232}\text{Th}$</th>
<th>$^{230}\text{Th}/^{234}\text{U}$</th>
<th>Date (kyr)</th>
<th>Corrected date</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>03-92</td>
<td>0.0325 ± 0.0007</td>
<td>1.16 ± 0.004</td>
<td>21</td>
<td>0.696 ± 0.0046</td>
<td>124 ± 1.5</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>04-8</td>
<td>0.228 ± 0.0047</td>
<td>1.06 ± 0.0041</td>
<td>14</td>
<td>0.493 ± 0.0042</td>
<td>73.1 ± 0.89</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>04-9</td>
<td>0.48 ± 0.0002</td>
<td>1.33 ± 0.01</td>
<td>4</td>
<td>0.034 ± 0.0043.7</td>
<td>3.7 ± 0.4</td>
<td>2.7 ± 0.5</td>
</tr>
</tbody>
</table>

4.1 Karst evolution and Geochronological data

The origin of the Pindal Cave is mainly controlled by two fractures trending E-W (F1) and their intersection with the subhorizontal joint system (D1). The opening of the Cave took place under phreatic conditions, with phreatic tubes trending along the cave axis, followed by vadose conditions. These vadose conditions are represented by 1 – the block collapse from the roof related to the intersection of the three joint systems; 2 – the episodic flooding and accumulation of detritic sediments coming from the denudation of Pimango Rasa Range (125–170 m), located to the South, and 3 – the chemical precipitation of speleothems over both alluvial and collapse deposits. No evidence of freshwater-seawater interaction has been found in the cave morphology. Phreatic cave passages must have initiated in the fluvial system once the Pindal terrace (50–64 m) was above sea level. Vadose incision requires base level lowering, which may have occurred during terrace uplift or may have resulted from sea level fall after the terrace reached its current position. Geomorphological evidence suggest the idea of a relatively fast elevation of the karstic massif relative to sea level, with an active fluvial stream under phreatic conditions simultaneously downcutting the massif. Episodes of downcutting would be separated by episodes of rapid block collapse leading to cave widening.

Based on the geochronological results obtained from speleothems (Table 1 and Fig. 4), the karst development in the area has a minimum age of 124.2 ± 1.5 ka BP (sample 1). The oldest sample comes from the 70 cm of the outer part of a stalagnite deposited on top of the flowstone, and can be considered as a minimum age for cessation of major flowstone deposition. As a consequence,
we can constrain the minimum age of the cave and hence, estimate the age of the marine terrace in which it is developed. These topics, together with the evolution of the cave, will be discussed in the following sections.

[Diagram of cave features and sedimentary phases]

Legend:
- Bedrock (Montaña Limestone)
- Discontinuities (F1, D1 and So)
- Collapse Blocks
- Alluvial deposits
- Other detrital deposits
- Speleothems:
  - Phase 1 (124.2 ± 1.5 Kyr BP)
  - Phase 2 (undated)
  - Phase 3 (371 ± 0.4 Kyr BP)
  - Phase 4 (Younger than 200 yr)
  - Stalagmite

Fig. 4. Diagrammatical sketch showing the relationships between cave features and the four generations (phases) of speleothems together with geochronological results (the age of the third generation is mathematically corrected to 2.7 ± 0.5 kyr).

4.2 An approach to the initiation of flowstone deposition

As shown by former geomorphological research in the area, roof pendants and phreatic tubes as well as paleochannels with fluvial sediments suggest that the cave passages probably opened up under phreatic conditions (Jiménez-Sánchez et al. 2002). However, geomorphological evidence shows that the base level must have dropped allowing the onset of vadose conditions, which favoured the widening of the cave by roof blocks collapse and perhaps by some additional downcutting yielding a total vertical development of 7–11 m from ceiling to base level of flowstones.
Ceiling morphology (following a subhorizontal dipping controlled by the joint system) is indicative of block falling in most parts of the cave. However, collapse block accumulations are only found in the central part of the cave and few isolated blocks can be found on top of the flowstones. Therefore, it can be deduced that the cave was under vadose conditions before the onset of major flowstones.

Considering that the dated sample 1 corresponds to a flowstone surface, obviously the onset of flowstone deposition took place in earlier times. The flowstone is estimated to have a total thickness of 3 m. If the rate of deposition between 124-73 ka were extrapolated over the entire thickness, the basal age would be of about 290 ka; if the deposition rate were 2x faster or slower than the one represented from 124-73 ka, the basal ages would range between 210 ka and 460 ka (Table 2). From a palaeoclimatic point of view, the interval between 124 and 73 ka coincides with

Table 2. Estimated basal age of flowstone for a 3 m thickness and inferred minimum uplift rate (mm/yr).

<table>
<thead>
<tr>
<th>Case</th>
<th>Basal Age (ky)</th>
<th>Minimum uplift rate (mm/yr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 Continuous average accumulation</td>
<td></td>
<td></td>
</tr>
<tr>
<td>a At same rate as inferred for 124-73 ka</td>
<td>292</td>
<td>0,08</td>
</tr>
<tr>
<td>b At double the rate inferred for 124-73 ka</td>
<td>208</td>
<td>0,12</td>
</tr>
<tr>
<td>c At half the rate inferred for 124-73 ka</td>
<td>459</td>
<td>0,05</td>
</tr>
<tr>
<td>2 Deposition only during interstadials (at rate as 124-73 ka)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>a Deposition when sea level &gt; -50m</td>
<td>716</td>
<td>0,03</td>
</tr>
<tr>
<td>b Deposition when sea level &gt; -75m</td>
<td>422</td>
<td>0,06</td>
</tr>
</tbody>
</table>

the penultimate interglacial, a time of overall warmer climates, more like modern conditions. In contrast, during glacial times ocean temperatures were lower by 8 °C off the northern coast of Spain which would have decreased precipitation and vegetation cover shifted from deciduous forest to tundra and alpine vegetation types (FLINT 1971, CLIMAP 1981). This combination of reduced infiltration and reduced soil CO₂ would have decreased the delivery of CaCO₃ for flowstone deposition. Consequently, it may not be coincidence that the flowstone became fractured during the development of this particular 124 ka horizon because this date may represent the onset of deposition following a hiatus during the penultimate glacial period. Studies in other European flowstones have indicated a strong climatic control over depositional and non-depositional periods on speleothems (e.g. BAKER et al. 1995). Consistent with this reasoning, the third generation of stalagmites includes those with Holocene ages, formed during the present interglacial (Fig. 5).

If the inner layers of the flowstone were deposited mainly during previous interstadials, then the linear extrapolation of the depositional rate beyond 124 ka would significantly underestimate the true basal age of the flowstone. An alternative approach consists in the calculation of the depositional window as that during interglacials, defined as sea level within 50 m below the present one, consistent with the sea level curve for the 124-73 ka time of most recent deposition (Fig. 5)
Following this model, 3 m of growth at a rate similar to the one estimated for the last interglacial, would result in flowstone initiation during the interglacial commencing at about 710 ka (Table 2).

4.3 The marine terrace uplift and the cave development

As commented earlier, geomorphological evidence suggests uplift in the karstic massif, with an active fluvial stream downcutting the massif under phreatic conditions. Evidence of fracture and collapse in the cave floor also suggest that uplift is an active process at present. The entrance of the cave is located 24 m above the sea level, with its active level at 6 m above the sea. A simple estimation considering 24 m as an altitude reference for the basis of the dated speleothem, and a minimum age for the Pindal Rasa close to 300 ka, gives a tentative uplift rate of 0.19 mm/yr for the Cantabrian Shore in this sector. This would give an approach to the maximum rate of relative uplift of the marine terrace in this area. Preliminary dating results in a marine terrace at 100 m altitude in the Western Asturias Coast indicate $^{21}$Ne model ages between 200 and 500 ka (Alvarez Marrón et al. 2005). These ages can be used to estimate tentative rates of uplift of 0.5 mm/yr and 0.2 mm/yr, respectively. The second one is similar to the one obtained in Pindal Rasa, under-scoring that inferred ages are very young.

Relative uplift could reflect either purely eustatic sea level fall or neotectonic uplift. Sea levels 60 m higher than present occurred most recently in the late Oligocene, at approximately 30 Ma (Kominz et al. 1998). We believe that neotectonic uplift is more plausible, since Cenozoic tectonic activity in the Cantabrian region has been widely reported by other studies (Boillot et al. 1979, Marquínez 1990, Alonso et al. 1996, Alvarez-Marrón et al. 2003). If karst and cave develop-
ment occurred shortly after subaereal exposure of the marine terrace, consistent with a phreatic origin of the cave system, then the rapid rate of accumulation of flowstones implied by our data suggest that the cavity would have been largely infilled within several millions of years given the favourable climate for karst development experienced by the region.

Our next challenge is to date the oldest speleothems in the cave, in order to know the oldest age of the karst, and therefore, the minimum age of the marine terrace. Taking into account the basal ages estimated for the speleothem from different rates of growth and the influence of sea level changes, the uplift rates for the marine terrace would take values between 0.03 and 0.12 mm/yr, therefore leading to ages between 1.9 Ma and 475 ka for a mean altitude of 57 m of Pindal Rasa (Table 2). These data, although speculative, could constitute a reference to develop further studies in the Cantabrian Shore.

4.4 The youngest stalagmites and the recent cave evolution

The period of most recent stalagmite growth may be estimated from lamination in the actively growing stalagmite. Average thickness of laminae couplets was 0.4 mm. Several factors suggest that the deposition rate of these laminae may be annual. About 70% of annual cave dripflow occurs in the five wettest winter months, and average summer dripflow rates are about a factor of 5 lower than those of winter (Stoll et al. 2005b). Given stable cave atmosphere CO₂ pressures and temperatures, and comparable average supersaturation of cave dripwaters in different seasons, this situation is expected to produce a strong seasonal alternation in the rate of calcite deposition, with the precipitation of dense rapidly growing winter bands alternating with more porous slowly growing summer bands. If the 0.4 mm reflects an annual growth rate, this stalagmite falls in the average growth rate range of stalagmites modeled and measured by Genty et al. (2001). In this case, the basal age for the actively growing stalagmite is approximately 200 years before present. This age appears to represent a change in the conditions of both the interior and exterior of the cave, resulting in deposition of more microcrystalline porous calcite, and may reflect a change in cave ventilation or land use in the karst massif above the cave. For example, increased delivery of P or organic matter related to agricultural practices (Stoll et al. 2005b) may inhibit calcite precipitation and alter crystal habit (Frisia et al. 2003).

5 Conclusions

U/Th dates on speleothems provide the first absolute age constraints on the genesis and evolution of Pindal Cave. The timescale of cave evolution may be similar for the other numerous cave systems in similar geomorphological settings in the central Cantabrian margin. A minimum age for the origin of the endokarst would be in the range of 290 ka, with a first generation of speleothems ranging from 124,2 ± 1,5 ka BP to 73,1 ± 0,9 ka BP. These geochronological data also shed light on the relative uplift rate of the marine terrace which hosts the cave and implicate neotectonics as the main process driving terrace uplift. The oldest speleothem ages imply uplift rate of the Cantabrian Margin in this area of 0,19 mm/yr, consistent with uplift rates for terraces to the west. The ages of speleothem growth also suggest a tight coupling between regional climate and speleothem accumulation rates.
Acknowledgements

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