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► To cite this version:

Amandine Sartégou, Pierre-henri Blard, Regis Braucher, Didier Bourlès, Patrick Sorriaux, et al. Late Cenozoic evolution of the Ariège River valley (Pyrenees) constrained by cosmogenic $^{26}\text{Al}/^{10}\text{Be}$ and $^{10}\text{Be}/^{21}\text{Ne}$ dating of cave sediments. *Geomorphology*, 2020, 371, pp.1-20. 10.1016/j.geomorph.2020.107441 . hal-02953947

HAL Id: hal-02953947

<https://hal.science/hal-02953947>

Submitted on 2 Oct 2020

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1 **Late Cenozoic evolution of the Ariège River valley (Pyrenees) constrained by**
2 **cosmogenic $^{26}\text{Al}/^{10}\text{Be}$ and $^{10}\text{Be}/^{21}\text{Ne}$ dating of cave sediments**

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18

19 **Abstract**

20

21 Despite increasing knowledge on the orogenic phases of the Pyrenees, the Neogene
22 evolution of the range remains poorly constrained. The central Pyrenees, particularly the
23 Ariège River valley and its terrace systems and glacial extensions, are key to reconstructing
24 Pyrenean evolution during the Neogene. However, few terrace relics remain on the piedmont
25 edges. To overcome this limitation and temporally extend the dataset obtained from terraces,
26 we focus on alluvium-filled horizontal epiphreatic passages developed in limestone karstic
27 networks. These landforms record the transient position of former local base levels during the
28 process of valley deepening, similar to fluvial terraces. The alluvium fills of the studied caves
29 in the Tarascon-sur-Ariège area, in the transition zone between the upper range and the
30 piedmont, therefore enable the reconstruction of the geodynamic evolution of the Ariège
31 River valley. All studied caves are developed on at least eight levels. Based on burial
32 durations determined by $^{26}\text{Al}/^{10}\text{Be}$ and $^{10}\text{Be}/^{21}\text{Ne}$ terrestrial cosmogenic nuclide dating of

33 samples from epiphreatic levels, our results indicate Langhian to Messinian (~13–5 Ma)
34 incision rates averaging 48 m Myr⁻¹. However, the obtained record does not allow us to
35 accurately retrace the Pliocene evolution of the area due to the lack of known caves at
36 corresponding levels. Moreover, raised local base levels during glacial phases both make the
37 record more complex and call into question the methodological approach in terms of potential
38 internal sediment remobilization and mixing related to implied re-flooding periods.

39

40 **Keywords**

41 Cave deposits, cosmogenic burial dating, ²⁶Al, ¹⁰Be, ²¹Ne, incision rates, Pyrenees,
42 karst geomorphology.

43

44 **Highlights**

- 45 • We identify eight regional cave levels in a Neogene valley in the eastern Pyrenees
- 46 • ²⁶Al/¹⁰Be, ¹⁰Be/²¹Ne burial durations obtained for intrakarstic alluvium
- 47 • Langhian-Messinian valley incision rates were ~48 m Myr⁻¹
- 48 • Palaeodenudation rates increased by one order of magnitude since the Miocene
- 49 • Cave re-flooding events preclude a post-Pliocene reconstruction of the Ariège valley

50 **1- Introduction**

51 Landscapes evolve via interactions between internal and external forcings.
52 Deciphering these retroactions rely on the knowledge of the deformation, exhumation and
53 sedimentation chronologies. The Pyrenees, an intraplate collisional range, were built by the
54 shortening of continental thinned margins during the convergence between Iberia and Eurasia
55 lasting from the Late Cretaceous to the Early Miocene. Because of the duration of the
56 orogenic growth and of its sedimentary evolution that both happen during the Cenozoic
57 climatic cooling, the Pyrenees appear particularly well-suited to investigate coupling and
58 retroactions between climate and tectonics and to attempt quantifying accretion and erosion
59 fluxes. In the case of the Pyrenees, most recent studies have focused on their evolution during
60 Quaternary glaciation phases (e.g., Stange et al., 2013; Delmas et al., 2015; Crest et al., 2017).
61 However, the geomorphological evolution of the Pyrenees during the Neogene is less
62 constrained, and several uplift and incision scenario remain debated (e.g., Babault et al., 2005,
63 Calvet and Gunnell, 2008; Gunnell et al., 2009; Bourdet, 2014; Monod et al., 2016a;
64 Sartégou, 2017; Sartégou et al., 2018). Recent works have focused on incision rates by dating
65 alluvial terraces (e.g., Stange et al., 2012, 2013, 2014; Delmas et al., 2015; Nivière et al.,

66 2016) and the Lannemezan megafan (Mouchené et al., 2017). However, methodological
67 limitations and the limited preservation of alluvial terraces preclude constraining incision
68 rates prior to the Mio-Pliocene transition. Therefore, the upstream history of valley incisions,
69 particularly those excavated into limestone massifs, may prove useful.

70 Incision events can be recorded via horizontal karstic systems, and some epiphreatic
71 levels are established in direct relation with their former base levels. Due to variations in these
72 former base levels, such karstic systems are staggered, and the alluvial deposits they record
73 enable the valley incision history to be traced based on their burial duration (e.g., Hauselmann
74 and Granger, 2005; Mocochain et al., 2009; Plan et al., 2009; Sartégou et al., 2018). However,
75 valley evolution is often complex; the Pyrenees have undergone intense glaciation phases that
76 marked the landscape and may have impacted the incision records.

77 Several studies have focused on the Niaux cave and its surroundings, and the
78 Tarascon-sur-Ariège area (France), has thus become fairly well known. This region is located
79 at the transition zone between the upper Pyrenees and the piedmont. At the surface, the spatial
80 and chronological footprints of six Würmian (MIS 2 to 5) and pre-Würm glaciation stages are
81 constrained by *in-situ* ^{10}Be dating of glacial margin deposits (Delmas, 2009). The stratigraphy
82 and sedimentology of the Niaux cave fills and U-Th dating of several stalagmite floors
83 provide a chronological framework for the last 350 kyr and the first evidence of multiple
84 glaciation cycles in Pyrenean valleys (Sorriaux, 1981, 1982; Bakalowicz et al., 1984).
85 Nevertheless, these data comprise indirect dates for interglacial stalagmite floors, and no
86 study has established a link between the surface sedimentary archives on the slopes of the
87 Ariège River catchment and the intrakarstic deposits.

88 The aim of this study is to date alluvium injection phases in karstic networks of the
89 Tarascon-sur-Ariège area and estimate the minimum formation age of large phreatic galleries
90 in relation to the floor altitude of neighbouring valleys (Sorriaux et al., 2016). We explored 8
91 epiphreatic levels in the Ariège River catchment, surveyed 21 horizontal caves, and collected
92 alluvial deposits in 11 caves for cosmogenic nuclide analyses (^{10}Be , ^{26}Al , and/or ^{21}Ne). The
93 limitations of our methodologies and the importance of our selection of dating objects is given
94 particular focus. Notably, the significant influence of cave re-flooding episodes that caused a
95 partial evacuation of the previous sediments within a cavity is investigated. We show that
96 karstic landforms record the history of the Ariège River valley incision, and that the data
97 acquired in this region cannot explain the late Neogene valley via a simple topographic uplift
98 mechanism. The evolution of the palaeodenudation rates, that increased by an order of
99 magnitude between the Miocene and the Quaternary, are also discussed.

100 2- Geological and geodynamical contexts

101

102 2.1- Lithological and structural contexts of the central-eastern Pyrenees

103 The Pyrenees (southwestern Europe) formed along the boundary between the Iberian
104 and European plates due to the Late Cretaceous-Cenozoic inversion of a Cretaceous
105 transcurrent hyper-extended rift created during the opening of Biscay Bay (e.g., Lagabrielle et
106 al., 2010). This mountain range forms a 400-km-long geographical barrier oriented N110°
107 (e.g., Choukroune, 1992). Pyrenean structures extend along the southern shore of the Biscay
108 Bay to the Asturias Mountains in the west. The Axial Zone of the Pyrenees comprises various
109 terrains of Precambrian and Palaeozoic ages that were folded during the Carboniferous (355–
110 295 Ma) Hercynian orogeny (Engel, 1984).

111

Figure 1

112 The source of the Ariège River is Lac Noir (2,400 m above sea level, a.s.l.), in the
113 Font-Nègre cirque. In its upper course, the Ariège River crosses the crystalline Hospitalet
114 massif towards the northeast before its confluence with the Oriège and Lauze rivers. It then
115 flows northwest through the Tabe and Montcalm massifs in the bottom of a glacial trough
116 before reaching Tarascon-sur-Ariège (Fig. 1). The Ariège River receives the waters of several
117 tributaries such as the Najar (confluence at Savignac), Aston (confluence at Les Cabannes),
118 Vicdessos (confluence at Tarascon), and Saurat (confluence at Tarascon), and brings water to
119 crystalline massifs. After its confluence with the Aston, the Ariège River crosses the North
120 Pyrenean fault and incises the strongly deformed and faulted Cretaceous limestone of the
121 North Pyrenean Zone. Beyond Tarascon, the Ariège River flows towards the north, and then
122 enters the Garonne at Portet-sur-Garonne (south of Toulouse), from where it connects to the
123 Atlantic Ocean.

124

125

Table 1

126

127 The lithostructural framework along the Ariège is complex (see supplementary
128 material for a detailed geological description). The massifs are strongly folded and faulted
129 (Fig. 1). The Ariège river drains quartz-crystalline zones in its southern distal part (in the
130 upper chain) before incising downstream limestone-rich areas in the North-Pyrenean zone,
131 where numerous karsts and caves are developed.

132

Figure 2

133

134 **2.2- Quaternary history of the Ariège catchment**

135 During glacial periods, astronomically driven climate fluctuations triggered the
136 development of the Ariège glacier, whose dynamics contributed to downstream valley
137 incision (e.g., Goron, 1937). These events are recorded in the studied caves, through
138 morphologies and sediments. The composite glacier of the Ariège basin was therefore one of
139 the largest Pleistocene glaciers on the northern slope of the Pyrenees. At the maximum of the
140 last glacial period, between 25 and 20 ka, the Ariège glacier reached an altitude of
141 approximately 1,300 m a.s.l. at the confluence with the Aston glacier, implying an ice
142 thickness of 800 m. Only the highest limestone massifs, including those of Quié de Lujat,
143 emerged from this ice surface (Fig. S1, supp. info). Moreover, the ice diffused towards
144 neighbouring valleys on all sides. In the Vicdessos valley, glaciers only reached 1,000 m a.s.l.
145 in the valley of Génat and did not exceed 500 m a.s.l. in the Niaux glacial basin. North of the
146 basin, ice thicknesses exceeded 400 m, implying that the glacier front must have been much
147 further downstream and may have reached the Foix-Montgaillard basin (Delmas, 2009).

148 **3- Karst features and cave systems**

149 We investigated several massifs in the vicinity of Tarascon-sur-Ariège (Fig. 2; Table
150 1), three of which are karstified (Cap de la Lesse, Sédour, and Calamès). The karst maximum
151 altitude is 1,189 m a.s.l., and the Ariège flows at 470 m a.s.l. at Tarascon. Consequently, in
152 these Cretaceous massifs affected by tight folds and inverse faults (Sorriaux, 1982), nine to
153 ten staggered levels have been identified along nearly 900 m of elevation. Unlike the Têt
154 River valley (Sartégou et al., 2018), karstic levels are vertically connected in some massifs of
155 the Ariège River valley, particularly Cap de la Lesse. Epikarstic morphologies are difficult to
156 study in this area due to the presence of vegetation.

157

158 **3.1- Cap de la Lesse massif**

159 In the Cap de la Lesse massif, two karstic networks belonging to the large Niaux-
160 Lombrives-Sabart network are notably developed (Fig. 3). Although they are small in extent
161 (5 km²), their accessible gallery network extends more than 14 km. This karst is located
162 between the deep Vicdessos and Ariège valleys (520 and 480 m a.s.l. at their respective valley
163 floors). The upper part comprises the Niaux and Lombrives caves and the upper galleries of
164 the Sabart cave, whilst the lower part includes the lower galleries of the Lombrives cave and

165 two large halls of the Sabart cave; upper and lower parts are connected by large vertical shafts
166 in Lombrives cave and by two large chimneys that seem to rise towards the Cap de la Lesse
167 summit in the Niaux system (Abîme Martel, 967 m a.s.l., and Salon Noir, 780 m a.s.l.; Fig. 3).
168 The entire Niaux karstic system is currently dry (Sorriaux, 1982) and belongs to the
169 infiltration zone, from which it is fed by seeps only. However, Sorriaux (1982) showed that
170 this karstic system was previously crossed three times by more or less rapid flows, which
171 delivered gneisses and granites from the Pyrenean Axial Zone and North Pyrenean primary
172 massifs. The galleries were therefore repeatedly and sometimes totally filled, then partially to
173 completely emptied (Sorriaux, 1982). Three alluvial formations have been identified by
174 Sorriaux (1982) as: the "alluvial formation of Lombrives" (F1), "conglomerates of the gallery
175 of the Counts of Foix" (F3) and "alluvial formation of Niaux" (F5); site names 'F-' refer to
176 the sites where these formations are best represented. The alluvial formation of Niaux exists
177 only throughout the upper stage of the system, whereas the other two are ubiquitous
178 throughout the system. Because these fillings must have been deposited in previously formed
179 galleries, karstogenesis was anterior to the oldest deposits.

180 The "alluvial formation of Lombrives", the oldest by relative chronology, was
181 rubified, which produced kaolinite in the east and northeast sections of the karst (Sorriaux,
182 1982). Because these alluvial sediments were deposited via turbulent currents or debris flows
183 (Sorriaux, 1982), sediment particle sizes, sedimentary structures, and erosion patterns enable
184 reconstruction of the dominant flow direction. The "alluvial formation of Lombrives" and
185 "conglomerates of the gallery of the Counts of Foix" (upper floor) correspond to flow from
186 the Vicdessos towards the Ariège (west to east), in agreement with the slight eastward slope
187 of the Niaux-Lombrives system (Sorriaux, 1982).

188 *Figure 3*

189 Sorriaux (1982) postulated that during the Last Glacial Maximum (Delmas et al.,
190 2012), when coalescent glaciers covered the massifs almost entirely, water flowed through the
191 hypogean karst system from east to west, i.e., from Ariège to Vicdessos. During the following
192 Garrabet deglaciation stage, waters abandoned the upper levels of the karst due to glacial
193 retreat and associated glacial disjunctions. Melt waters of the Ariège and Vicdessos then
194 joined the Lombrives lower level and flowed to Sabart. During the final stages of the Garrabet
195 cycle, the surface of the Ariège glacier only reached an altitude of 600 m a.s.l., and the
196 Vicdessos glacier was confined to its high valley. Therefore, at that time, waters could only
197 penetrate the system at the level of the Lombrives cave inlet.

198 Subsequent to the alluvial deposits, these galleries also contain red clay formations
199 that testify to the occurrence of lakes. This necessitates that the previously described glacial
200 levels do not strictly correspond to the stepped galleries.

201 Dating of the alluvial sediments deposited in this karst system therefore better
202 constrains its functionality and geomorphological evolution.

203

204 **3.2- Sédour, Calamès, and Quié de Lujat massifs**

205 The Sédour and Calamès massifs (Figs. 4 and 5, respectively) were originally
206 constituents of the same limestone units, but have since been intersected and progressively
207 entrenched by the Saurat River. This implies that data obtained in the few preserved cavities
208 (due to receding walls) can be correlated. Alluvial deposits in these massifs are highly altered
209 and rubified, with crystalline pebbles.

210 Inventoried cavities of the Sédour massif (Fig. 4) indicate their consistent staging,
211 though less-accessible cavities do not contain intrakarstic sediments. The most developed
212 cavity is the Bédeilhac cave, which contains rupestrian art and is the only cave in the
213 investigated massifs with galleries wide and high enough to have served as an aircraft hangar
214 during World War II. Hangar installation at the entrance of the cavity exposed a large section
215 of alluvial deposits, which range from fairly rubified medium sands to altered crystalline
216 pebbles. Clay deposits are also numerous, possibly indicating phases of transient damming.
217 Chrono-stratigraphic correlation of these deposits is difficult; although correlating this and the
218 Niaux-Lombrives levels seems reasonable, although the presence of faults and the proximity
219 of a formerly exploited Triassic gypsum formation render this correlation speculative.
220 However, numerous circulations in the epiphreatic regime (lapiés, circular pools, vault
221 channels, and scallops) provide opportunities for improving the correlation relationships.

222

223

Figure 4

224 The Bouicheta cave opens on the western flank of the Sédour massif and developed
225 mainly along a NE-SW axis over a length of approximately 85 m. Because this cavity has
226 undergone several excavations and has served as a sheep fold, the organization of the
227 sedimentary deposits has been disturbed. Nevertheless, old stalagmitic floors surmount silty
228 deposits, and an important concretion episode is preserved.

229 The Moutons cave developed along the same axis as the Bouicheta cave, but over a
230 length of approximately 20 m. Inside a low room nearly as long as it is wide, large portions of

231 stalagmitic floors are present. The walls of this room present deep, circular pool shapes
232 indicating an ancient epiphreatic regime, and a conduit leads to alluvial deposits.

233 Cavities in the Calamès massif (Fig. 5) are relatively short. The Quarry cave, at the
234 base of the limestone cliff along Saurat River, developed over a length of ~5 m, although the
235 wall has retreated by ~30 m due to anthropic exploitation. The remaining vault section is
236 fairly wide. It seems to be a drain that formed in an epiphreatic regime. The observed lapiés
237 may have formed on the banks of an aggraded river, and fills are alluvial deposits (highly
238 altered pebbles surmounted by graded sandy levels) that contain intercalated relics of
239 stalagmitic floors. A lacustrine and deltaic level that originated in the Bédeilhac palaeolake
240 faces the cavity. These deposits comprise varves with silty strata, some sandy and gravelly
241 occurrences, and pebbled levels. Two morainic blocks are located at the top of the sequence.

242 *Figure 5*

243 Cavities on the south side of the massif are slightly more developed. The length of the
244 Enchantées cave is approximately 45 m, and its shape displays numerous indices of
245 epiphreatic regime with corrosion at the vaults. Its abundant fillings comprise rubified
246 sandstone claddings on the walls and vaults, and smaller clasts are altered. Two erratic blocks
247 lie at the porch entrance.

248 Further upstream, the limestone cliff opens onto several small porches. Here, three
249 small, short caves (~50 m in length) are presently well separated. However, due to their
250 shared NW-SE orientations and the similarity of the observed morphologies, this gallery was
251 probably intersected by the recession of the wall of the Esplourgues cave. Similar to the
252 Enchantées cave, flowstone formation has been intense and stalagmitic floors are inserted in
253 varied fillings ranging from laminated strands to powdery pebbles clad to the walls. The
254 morphologies of the walls and vaults are characteristic of epiphreatic flows. However, at the
255 most perched entrance, fluid escape structures are observed as coarse to highly altered and
256 rubified pebbly deposits.

257 The Quié de Lujat massif is poorly karstified compared to the other massifs in the
258 study area, and few relics of horizontal conduits remain. Typical morphologies of epiphreatic
259 circulations, including scallops and deep circular pools, are visible at the entrance to the
260 cavity, indicating a possible resurgence. Various cemented coarse alluvial deposits remain,
261 including highly altered and rubified crystalline pebbles. This cavity, perched at 1,100 m
262 a.s.l., was below the maximum level of the Ariège glacier during the Foix glacial cycle
263 (Sorriaux et al., 2016).

264 Splouga de Souloubrié, in the eastern slope of Quié de Lujat massif, is a cavity that
265 also presents many typical flow morphologies (scallops and circular pools). The main gallery
266 is obstructed by a rockfall, which may truncate the small network.

267 **4- Materials, methods, and sampling strategy**

268

269 **4.1- Description of the studied caves**

270 Many caves in the studied area (Fig. 6; Table 1) are hidden and difficult to access due
271 to vegetation cover. These caves are also subject to heightened protection measures due to
272 their archaeological interest or their importance to the preservation of specific troglobite
273 species. Thus, only three cavities in Calamès massif, three in Sédour massif, two in Quié de
274 Lujat massif, and two in Cap de la Lesse massif could be sampled.

275

276 **4.1.1- Epiphreatic model and chronology of valley deepening**

277 The epiphreatic model of Audra and Palmer (2011, 2013), on which our work is based,
278 considers three different zones: the vadose zone, the epiphreatic zone (in which the water
279 table fluctuates), and the phreatic zone. This model assumes that recharge occurs via sinking
280 streams, dolines, and the epikarst, a branching cave system established in the vadose zone
281 above the water table. Vadose passages include shafts and canyons. Groundwater flows along
282 a relatively gentle gradient to springs at the water table level in nearby valleys. Most phreatic
283 passages are tubular and form at or slightly below the water table, although many exhibit
284 vertical loops. During floods, phreatic passages may be unable to transmit all the incoming
285 water, causing the development of complex looping overflow routes in the epiphreatic zone.
286 Large phreatic passages develop when the erosional base level remains at a given elevation
287 for an extended period of time (Audra, 1994) and when erosional benches developed. When
288 the base level drops and the surface river erodes downward, phreatic passages tend to drain
289 through diversion routes, and the former phreatic passages indicate the former base level.

290

Figure 6

291 The use of karstic systems as markers of palaeo-base levels relies on the localization
292 of certain key morphological factors (Fig. 6B, D), such as characteristic morphologies formed
293 under phreatic or epiphreatic conditions. Cavities of the Tarascon valley mostly contain
294 intrakarstic sediments emplaced by river circulation (see Fig. 6 and Section 3). Allogenic
295 sediments trapped in the karst systems reflect the geomorphological evolution of both the

296 surface and underground. However, deposits are better preserved underground, especially
297 when concretions, stalagmitic floors, or landslides seal previous deposits.

298 *Figure 7*

299 In most cases, the alluvial sediments we encountered in the cavities were collected and
300 analysed. Reconstituting the stratigraphy is often complex, as only a few residues remain in
301 the visited galleries, except for the larger caves such as Bédeilhac (Fig. 7), Lombrives, and
302 Niaux (Figs. 8, 9). Therefore, we only sampled alluvium cemented against walls that were
303 clearly related to characteristic flow morphologies. In the larger networks of Bédeilhac,
304 Niaux, and Lombrives, more substantial and continuous deposits were sampled.

305 *Figure 8*

306 *Figure 9*

307 **4.1.2- Attempts to date multiple fillings**

308 In the Niaux-Lombrives-Sabart cave system, fillings are predominantly preserved as
309 discontinuous deposits on the walls of the galleries and in their anfractuosités or connected to
310 the ceiling (Fig. 8). Discontinuities between deposits, limited accessibility, and darkness due
311 to large volumes complicate research on the stratigraphy of these fillings. Because the
312 deposits are occasionally juxtaposed, several generations of alluvial fillings in the Niaux and
313 Lombrives caves are recognizable. Alluvial deposits also show characteristic flow
314 morphologies. Based on the relative chronology and sizes of the observed deposits, large
315 water flows may have filled the galleries without completely removing in-place sediments.
316 These flows were powerful enough to transport blocks (Sorriaux, 1982). Various generations
317 of sediments were collected in areas where stratigraphic and geometric relationships were
318 obvious, the objective being to improve the absolute and relative chronologies of the multiple
319 filling episodes. Samples were also collected in areas previously sampled for petrological
320 studies (Sorriaux, 1982) to obtain additional information via particle size distributions and
321 thin-section observations.

322

323 **4.2- Burial dating with cosmogenic nuclides**

324 **4.2.1. Systematics**

325 Rocks or soils exposed to cosmic radiation at the Earth's surface accumulate terrestrial
326 cosmogenic nuclides (TCNs, such as ^{26}Al , ^{10}Be , or ^{21}Ne) in quartz during their exposure. TCN
327 concentrations in such materials can be calculated as (Braucher et al., 2011):

$$C(t, \varepsilon) = \frac{P_n \cdot \left(1 - \exp\left(-t \left(\frac{\rho \varepsilon}{L_n} + \lambda\right)\right)\right)}{\frac{\rho \varepsilon}{L_n} + \lambda} + \frac{P_{\mu s} \cdot \left(1 - \exp\left(-t \left(\frac{\rho \varepsilon}{L_{\mu s}} + \lambda\right)\right)\right)}{\frac{\rho \varepsilon}{L_{\mu s}} + \lambda} + \frac{P_{\mu f} \cdot \left(1 - \exp\left(-t \left(\frac{\rho \varepsilon}{L_{\mu f}} + \lambda\right)\right)\right)}{\frac{\rho \varepsilon}{L_{\mu f}} + \lambda}, \quad (1)$$

where $C(t, \varepsilon)$ is nuclide concentration (at g^{-1}), t exposure time (yr), ε denudation rate (cm yr^{-1}), and ρ the rock/soil density (g cm^{-3}). P_n , $P_{\mu s}$, and $P_{\mu f}$ are the production rates and L_n (160 g cm^{-2}), $L_{\mu s}$ ($1,500 \text{ g cm}^{-2}$), and $L_{\mu f}$ ($4,320 \text{ g cm}^{-2}$) (Braucher et al., 2011) the attenuation lengths of neutrons and slow and fast muons, respectively, and λ is the radioactive decay constant (yr^{-1}) of the considered TCN.

The burial dating method is based on the analysis of materials that were initially exposed to cosmic rays before being buried to $>10 \text{ m}$ depth, where *in-situ* muonic production is reduced to 1% of the total surface production (Braucher et al., 2011). This burial halts the *in-situ* production of TCNs, and their concentrations then evolve by radioactive decay (^{10}Be , ^{26}Al) or remain constant (^{21}Ne). Thus, measuring the concentrations of two cosmogenic nuclides with contrasting half-lives (e.g., ^{26}Al - ^{10}Be or ^{10}Be - ^{21}Ne) permits computation of the burial duration. The $^{26}\text{Al}/^{10}\text{Be}$ ratio can be used to date burial events that occurred during the last $\sim 5 \text{ Myr}$ (e.g., Granger & Muzikar, 2001), and $^{10}\text{Be}/^{21}\text{Ne}$ to investigate those within the last 15 Myr (e.g., Sartégou et al., 2018).

Burial dating requires that the pre-burial cosmogenic nuclide ratios are known (Blard et al., 2019a; b) and that the surface exposure of quartz, in which TCNs are measured, was long enough to accumulate measurable cosmogenic radionuclide concentrations. In a denudation regime, sediment transport time must have been negligible compared to the exposure duration on a surface prior to burial, and the sediment storage duration within a catchment must have been limited. Finally, burial must have been rapid and deep enough to avoid post-burial production of cosmogenic nuclides. Consequently, this method is well-suited to dating the filling of caves by detrital sediments (e.g., Häuselmann and Granger, 2005; Häuselmann et al., 2015; Sartégou et al., 2018; Häuselmann et al., 2020). Caves provide a near-ideal configuration for such burial duration determinations because post-burial exposure to cosmic rays is negligible in most cases. Furthermore, the galleries of the caves studied herein are always overlain by more than 50 m of rocky material, rendering post-burial production negligible.

In denudation regimes, pre-burial TCN concentrations of river sands are considered to have reached steady state (Lal, 1991), and represent the average of all quartz-bearing rocks outcropping upstream in the drainage basin (Brown et al., 1995). Using the approach of Blard et al. (2006), pre-burial average denudation rates can be determined for an entire drainage

360 basin by relating the average erosion rate $\bar{\varepsilon}$ to the palaeo-concentration \bar{C} at the moment of
 361 burial as:

$$362 \quad \bar{\varepsilon} = \sum_{i,x} \frac{\bar{P}_i L_i}{\rho \bar{C}}, \quad (2)$$

363 where x is the location in the catchment, i refers to the different TCN production pathways
 364 (see Eq. (1)), L_i are attenuation lengths for each particle, and \bar{P}_i are the mean basin-wide
 365 cosmogenic nuclide production rates, computed from the arithmetic mean of spallogenic and
 366 muogenic production using specific scalings for each production pathway as a function of
 367 elevation and latitude. Here, pre-burial denudation rates were computed using ^{10}Be
 368 concentrations (after correcting for radioactive decay) by using the following production
 369 rates: $P_n = 4.11 \pm 0.19$ atoms $\text{g}^{-1} \text{yr}^{-1}$, $P_{\mu s} = 0.011 \pm 0.001$ atoms $\text{g}^{-1} \text{yr}^{-1}$, and $P_{\mu f} = 0.039 \pm$
 370 0.004 atoms $\text{g}^{-1} \text{yr}^{-1}$ (Braucher et al., 2011; Martin et al., 2017). We corrected for mass
 371 shielding by using the equation of Dunai (2010). Muon contributions based on Braucher et al.
 372 (2011) were scaled for altitude only. We calculated production rates for each cell of a 30-m
 373 ASTER digital elevation model using a Matlab script and the CREp tool (Martin et al., 2017)
 374 and a rock density of 2.5 g cm^{-3} . Areas without surficial quartz-bearing rocks or not linked to
 375 the stream network were excluded from the calculations. No correction was performed for ice
 376 cover because it only represents a very small proportion of our study area.

377 Some studies have incorrectly used pre-burial cosmogenic nuclide ratios equal to the
 378 production rate ratio when computing burial ages from paired cosmogenic nuclides. In the
 379 case of slowly eroded material ($\ll 1 \text{ mm yr}^{-1}$), the pre-burial ratio may be lower than the
 380 production ratio, and such a simplification can result in overestimated burial ages. To avoid
 381 any bias, accurate burial age calculations should consider the elevation of the quartz-rich
 382 watershed and solve the following equation (Blard et al., 2019a, b):

$$383 \quad \frac{P_1}{N_1} e^{-\lambda_1 \cdot t_{\text{burial}}} - \frac{P_2}{N_2} e^{-\lambda_2 \cdot t_{\text{burial}}} = \frac{\lambda_1 - \lambda_2}{f}, \quad (3)$$

384
 385 where f is a spatial scaling factor representative of the latitude and altitude of the watershed,
 386 t_{burial} is the burial duration (yr), and P_1 and P_2 are the production rates (at $\text{g}^{-1} \text{yr}^{-1}$), N_1 and N_2
 387 the concentrations (at g^{-1}), and λ_1 and λ_2 the radioactive decay constants (yr^{-1}) of two
 388 cosmogenic nuclides ‘1’ and ‘2’, respectively.

390 In this study, we employed the following production rates (subscripts indicate the
 391 mass number of the nuclide): for P_{10} , we used the most up-to-date global averaged value of
 392 4.11 ± 0.19 at g a^{-1} available in the CREp calculator (Martin et al., 2017); for P_{21} , we used

393 17.0 ± 0.9 at g a^{-1} deduced from the $^{21}\text{Ne}/^{10}\text{Be}$ production ratio of 4.12 ± 0.17 (Kober et al.,
394 2007); and for P_{26} , we used 27.3 ± 0.2 at g a^{-1} deduced from the $^{26}\text{Al}/^{10}\text{Be}$ production ratio of
395 6.61 ± 0.52 (Nishiizumi et al., 2007; Braucher et al., 2011). The half-lives of ^{10}Be and ^{26}Al are
396 1.387 ± 0.120 and 0.717 ± 0.017 Myr, respectively (Chmeleff et al., 2010; Korchinek et al.,
397 2010; Granger, 2006; Samworth et al., 1972). The spatial scaling factor f was computed
398 following Stone (2000). Since Equation (3) cannot be analytically solved, we numerically
399 computed t_{burial} using a Monte Carlo approach (10^4 draws), considering that the cosmogenic
400 nuclide concentrations have normal distributions (N_{26} , σ_{26}) and (N_{10} , σ_{10}), where σ_{26} and σ_{10}
401 are their analytical uncertainties. The mean and standard deviation of the 10^4 solutions
402 obtained for t_{burial} were computed to obtain the best solutions and their respective uncertainties
403 (see Table 3 for ^{26}Al - ^{10}Be durations). In the case of the ^{10}Be - ^{21}Ne pair, Equation (3) can be
404 solved analytically (Blard et al., 2019a, b), and the burial age was then computed as:

$$405 \quad t_{\text{burial}} = \frac{-1}{\lambda_{10}} \cdot \ln \left[\frac{N_{10}}{P_{10}} \cdot \left(\frac{P_{21}}{N_{21}} + \frac{\lambda_{10}}{f} \right) \right]. \quad (4)$$

406
407
408 For both the ^{26}Al - ^{10}Be and ^{10}Be - ^{21}Ne pairs, burial ages were computed using the
409 present-day watershed elevation of 1,990 m a.s.l. Hypothetical palaeo-elevations of 0 and
410 4,000 m a.s.l. were also tested to evaluate the impact of elevation changes on the burial ages,
411 which induced a maximum variation of 10% in the computed burial ages. As an acceptable
412 uncertainty, reported burial ages were computed using the modern catchment elevation.
413 Temporal variations in the geomagnetic field integrated over the investigated time periods are
414 assumed to be negligible compared with the total uncertainty in the data.

415 416 **4.2.2. Measurement of in-situ cosmogenic ^{26}Al and ^{10}Be**

417
418 All ^{26}Al and ^{10}Be measurements were performed at CEREGE (Aix-en-Provence,
419 France). ^{10}Be and ^{26}Al were extracted from the rock samples following the chemical
420 procedure of Brown et al. (1991) and Merchel and Herpers (1999). Pebbles were crushed and
421 sieved, and the 250-1,000 μm fraction was processed. Quartz was concentrated by magnetic
422 separation and selective dissolution in a 1:2 HCl - H_2SiF_6 mixture. Quartz was decontaminated
423 for atmospheric ^{10}Be by dissolving 30% by mass via three successive HF leaching processes.
424 Pure quartz was spiked with 100 μL of an in-house ^9Be carrier ($3,025 \pm 9$ ppm; Merchel et al.,
425 2008) and an adapted amount of a commercial 985- $\mu\text{g/g}$ ^{27}Al carrier. The natural ^{27}Al

426 concentrations in the dissolved samples were measured by inductively coupled plasma optical
427 emission spectrometry (ICP-OES) on a Thermo Scientific ICP6500. These analyses were
428 performed using replicate aliquots, two emission lines (394 and 396 nm, axial sight), and two
429 standards (Merck, 999 ± 2 mg/L, and Fluka TraceCert, 985 ± 2 $\mu\text{g/g}$). Dissolution was
430 completed in concentrated HF.

431 Due to its high ^{27}Al content, the abundance of muscovite in rocks of the watershed
432 forced us to regularly check the sample purification with a binocular microscope (see Section
433 4.2.3). If not removed from the quartz before total dissolution, muscovite causes the formation
434 of thick solid crusts from which it is difficult to recover the dissolved aluminium.

435 BeO and Al_2O_3 were then purified and extracted using a succession of alkaline
436 precipitations and separations on ionic-exchange resins (DOWEX 1X8 100–200 mesh,
437 DOWEX 50WX8 100–200 mesh). Full oxidization was performed at 700°C for one hour.
438 The obtained BeO and Al_2O_3 oxides were mixed with Nb and Ag conductive powders,
439 respectively, then introduced in a copper cathode to be pressed. The $^{10}\text{Be}/^9\text{Be}$ and $^{26}\text{Al}/^{27}\text{Al}$
440 ratios were measured at the French national accelerator mass spectrometry (AMS) facility
441 ASTER. Measured $^{10}\text{Be}/^9\text{Be}$ ratios were calibrated against the NIST standard SRM 4325
442 ($^{10}\text{Be}/^9\text{Be} = (2.79 \pm 0.03) \times 10^{-11}$; Nishiizumi et al., 2007), and measured $^{26}\text{Al}/^{27}\text{Al}$ ratios were
443 calibrated against the ASTER in-house standard SM-Al-11 ($^{26}\text{Al}/^{27}\text{Al} = (7.401 \pm 0.064) \times 10^{-12}$;
444 Merchel and Bremser, 2004). The half-lives used and the standardisation method employed
445 at ASTER (SM-Al-11/07KNSTD) imply that the $^{26}\text{Al}/^{10}\text{Be}$ spallation production ratio is
446 $\sim 6.61 \pm 0.52$. Reported analytical uncertainties include counting statistics, machine stability
447 ($\sim 0.5\%$ for ^{10}Be ; Arnold et al., 2010), and blank correction.

448

449 ***4.2.3. Influence of muscovite on the accuracy of cosmogenic ^{26}Al concentrations***

450 Although our procedure to separate and purify quartz from the sampled sediments was
451 efficient in most cases, it was sometimes insufficient for muscovite-rich samples (reported in
452 italics in Table 3). To quantify the potential impact of the presence of muscovite in
453 supposedly pure quartz mineral samples, we performed tests using samples LO4, LO4b, and
454 LO4c from Lombrives cave (Table 2). LO4 is a quartzite pebble from a sedimentary sequence
455 that was recovered during the carbonate dissolution step of the quartz mineral separation
456 procedure. LO4b and LO4c are derived from the sandy matrix of the same sedimentary
457 sequence, which was particularly rich in quartz. During the various separation steps,
458 muscovites were recovered from this sandy matrix. After elimination of all other accessory
459 minerals, half of the obtained pure quartz mineral sample was processed for reliable ^{26}Al

460 measurements (LO4b), whereas 141 mg of the recovered muscovites was added to the other
461 half before ^{26}Al analysis (LO4c).

462 The ^{27}Al concentration of the muscovite-loaded sample (LO4c) is 17 times higher than
463 that of the muscovite-free sample (LO4b; Table 2). Consequently, after accounting for the
464 ^{27}Al spike ($9,372.07 \pm 46.86$ ppm) added to LO4b to obtain a sufficient amount of material for
465 a reliable AMS analysis, the measured $^{26}\text{Al}/^{27}\text{Al}$ ratios differ by an order of magnitude.
466 Although the amount of added muscovite is negligible compared to ~ 20 g of dissolved quartz,
467 it significantly distorted the natural ^{27}Al concentration and the measured $^{26}\text{Al}/^{27}\text{Al}$ ratio. This
468 distortion results in an erroneous ^{26}Al concentration, and thus $^{26}\text{Al}/^{10}\text{Be}$ ratio and derived
469 burial duration. Eliminating these alumina-rich minerals is therefore paramount. A very
470 thorough separation of mineral phases is necessary for muscovite-rich samples, including high
471 intensity magnetic separation before and after HF-leaching (there is too much static electricity
472 after the $\text{HCl-H}_2\text{SiF}_6$ step), flotation, and hand-picking to remove the last leaflets. Another
473 sieving step after the last HF-leaching may be useful because most remaining muscovites are
474 smaller than $250 \mu\text{m}$.

475 *Table 2*

476 Samples reported in italics in Table 3 probably have overestimated aluminium
477 concentrations, and thus underestimated burial durations. These results are therefore
478 unreliable and are excluded from further discussion.

479

480 **4.2.4. Measurement of *in-situ* cosmogenic ^{21}Ne**

481

482 Neon isotopic concentrations in quartz were measured at the CRPG (Nancy, France)
483 noble gas laboratory. The same quartz fractions ($200\text{--}1,000 \mu\text{m}$) used for ^{26}Al and ^{10}Be
484 analyses were used for neon analysis. All samples were wrapped in tin foil (melting point, 230
485 $^\circ\text{C}$) and loaded in the carousel of the new full-metal induction furnace designed at the CRPG
486 (Zimmermann et al., 2018). The furnace and the entire purification line were then baked at
487 approximately $150 \text{ }^\circ\text{C}$ overnight under high vacuum. Subsequently, the furnace crucible was
488 degassed at $>1500 \text{ }^\circ\text{C}$ to reduce the neon blanks. Following conclusive validation tests, neon
489 was extracted in a single heating step at $\sim 1400 \text{ }^\circ\text{C}$ for 20 minutes. The purification line
490 consists of three charcoal fingers cooled with liquid nitrogen (77 K) to trap Ar, Kr, Xe, and
491 chemically active gases including N_2 , O_2 , and CO_2 (Zimmermann et al., 2015). Three Ti-
492 sponge getters also permit trapping of the remaining reactive gases (particularly N_2 , O_2 , CO_2 ,

493 and H₂). Ne was then cryofocused at 30 K before being released at 100 K and input into the
494 mass spectrometer for analysis.

495 The purified gas was input into the analyser, and ²²Ne, ²¹Ne and ²⁰Ne concentrations
496 were measured using high-sensitivity mass spectrometers: the Helix-MC+™ (two aliquots of
497 sample SO1) and the Helix-SFT™ (all other samples). Neon isotopic concentrations were
498 measured by simultaneous multi-collection on the Helix-MC+™ and by peak jumping mass
499 spectrometry with the electron multiplier of the Helix-SFT™.

500 During mass spectrometry analyses, a getter trap minimized the contribution of CO₂⁺⁺
501 on the ²²Ne signal and maintained a low H₂ abundance, which can produce an isobaric
502 interference of ²⁰NeH⁺ on ²¹Ne (Honda et al., 2015). A charcoal finger is also present in the
503 mass spectrometer to trap CO₂, H₂O, and Ar (77 K). The sensitivity of the mass spectrometer
504 was determined using an in-house gas standard of atmospheric composition. After each
505 analysis, any remaining gas was pumped. Neon isotopic concentrations were corrected for hot
506 furnace blanks and instrumental mass fractionations. Isobaric interference of H₂¹⁸O on ²⁰Ne
507 was determined for each sample by measuring H₂¹⁶O and assuming a H₂¹⁶O/H₂¹⁸O ratio of
508 495 (Honda et al., 2015); this ratio corresponds to a correction smaller than 0.1%.

509 The neon extracted from the quartz samples is generally produced from a mixture of
510 cosmogenic neon and atmospheric neon. Cosmogenic neon is mainly located in the lattice,
511 while atmospheric neon is mainly trapped in fluid inclusions and/or adsorbed on grain
512 surfaces. The position of a given sample in the three-isotope plot indicates whether it belongs
513 to the spallation-air mixing line (Fig. 10). In practice, we determined the cosmogenic
514 concentrations ²¹Ne_{cos} as (Vermeesch et al., 2015):

515

$$516 \quad {}^{21}\text{Ne}_{\text{cos}} = R_c \times {}^{20}\text{Ne}_m \times \frac{R_m - R_a}{R_c - R_a}, \quad (5)$$

517 where ²⁰Ne_m (at g⁻¹) is the measured amount of ²⁰Ne, $R_c = 0.8$ is the cosmogenic ²¹Ne/²⁰Ne
518 production ratio, R_m is the measured ²¹Ne/²⁰Ne ratio, and $R_a = 0.002905$ is the atmospheric
519 ²¹Ne/²⁰Ne ratio (Honda et al., 2015).

520 Aliquots of the quartz standard CRONUS-A measured during the same session yielded
521 a mean ²¹Ne excess of $(3.38 \pm 0.05) \times 10^8$ at g⁻¹, in agreement with the reference
522 concentration of $(3.48 \pm 0.10) \times 10^8$ at g⁻¹ (Vermeesch et al., 2015).

523

524 **5- Results**

525

526 **5.1- Cosmogenic ^{26}Al , ^{10}Be results, $^{26}\text{Al}/^{10}\text{Be}$ burial durations and palaeodenudation rates**

527 All ^{26}Al and ^{10}Be results are presented in Table 3. Most samples yielded $^{10}\text{Be}/^9\text{Be}$
528 ratios significantly above the blank value (blanks ranged from 1.5×10^{-15} to 3.0×10^{-15}).
529 These ratios indicate ^{10}Be concentrations ranging from $(8 \pm 1) \times 10^3$ to $(114 \pm 18) \times 10^3$ at g^{-1} .
530 Uncertainties on ^{10}Be concentrations range from 6 to 41%. Some samples had $^{27}\text{Al}/^{26}\text{Al}$ ratios
531 below the analytical detection limit ($\sim 2 \times 10^{-15}$; blank value, 2.2×10^{-15}), suggesting burial
532 durations longer than 6.5 Myr. These samples, for which only ^{10}Be concentrations were
533 measurable, are from the highest caves (Bouicheta, Moutons, Mirouge, and Souloubrié).
534 Otherwise, ^{26}Al concentrations varied between $(3 \pm 3) \times 10^3$ and $(34.8 \pm 4.1) \times 10^4$ at g^{-1} .
535 Uncertainties on ^{26}Al concentrations ranged from 12 to 100%.

536 $^{26}\text{Al}/^{10}\text{Be}$ burial durations ranged from 0.7 ± 0.4 to 8.8 ± 1.7 Myr. The durations
537 determined for Lombrives and Niaux caves are scattered, which is not surprising given the
538 evidence of several phases of filling. However, the burial durations calculated for Ni13-08,
539 Ni13-10, Ni13-15, Ni13-20, and Ni13-23 in Niaux cave are not internally consistent and do
540 not agree with the U-Th ages obtained by Sorriaux (1981, 1982, in press) and Bakalowicz et
541 al. (1984) for most of the stalagmitic floors that embed these alluvial formations. Indeed,
542 although they belong to the same depositional sequence that can be followed continuously
543 over a few meters, the burial durations calculated for Ni13-23 and Ni13-20 are significantly
544 different. Therefore, alluvial deposition phases in Niaux cave cannot generally be
545 discriminated. In rare cases, calculated burial durations are not consistent with the relative
546 chronology, particularly the latest deposited sequences LO1 and Ni12-02, for which the burial
547 durations are different. The most problematic finding, however, is that equivalent deposits in
548 the Niaux-Lombrives-Sabart cave system return different burial durations depending on the
549 cave considered. These inconsistencies are not observed in the Bédeilhac cave, which may be
550 part of a single level and therefore evolved under similar conditions. However, no U-Th
551 dating is available for this cavity, and fewer cosmogenic burial dates were obtained than in
552 Niaux cave. Furthermore, the stratigraphy in the Bédeilhac cave is quite ambiguous.
553 Therefore, firmly attributing the observed variations to a sampling bias is difficult.

554 *Table 3*

555 Initial concentrations obtained for samples from caves of the Calamès massif produced
556 coherent ages at approximately 3–4 Ma. However, the analysed samples (CALA 14-02, 14-
557 04, 14-05) contained abundant muscovite. Subsequent measurements of muscovite-free
558 samples taken at the same sites (CALA 14-01 and 14-03) indicate significantly longer burial

559 durations consistent with those of samples from Bédeilhac cave. Therefore, eliminating
560 muscovite is crucial before performing any cosmogenic ^{26}Al measurements.

561 Palaeodenudation rates derived from the measured ^{10}Be and ^{26}Al concentrations range
562 from 3 ± 1 to 797 ± 101 mm/kyr (Table 3).

563

564 **5.2- Cosmogenic ^{21}Ne results, $^{10}\text{Be}/^{21}\text{Ne}$ burial durations and palaeodenudation rates**

565

566 Neon isotopic concentrations and ratios, and cosmogenic ^{21}Ne concentrations are
567 reported in Table 4, and $^{10}\text{Be}/^{21}\text{Ne}$ burial durations and palaeodenudation rates in Table 5.

568

569

Table 4

570 Cosmogenic ^{21}Ne concentrations computed with Equation (5) range from (1.20 ± 0.19)
571 $\times 10^7$ at g^{-1} to $(2.20 \pm 0.26) \times 10^7$ at g^{-1} . Relative uncertainties range from 12 to 48% and
572 mainly result from the atmospheric ^{21}Ne correction. These $^{21}\text{Ne}_{\text{cos}}$ concentrations imply
573 present $^{21}\text{Ne}/^{10}\text{Be}$ ratios significantly larger than the surficial $^{21}\text{Ne}/^{10}\text{Be}$ production ratio of
574 4.12 ± 0.17 (Kober et al., 2007). These results indicate significant burial durations that range
575 from 6.8 ± 0.3 to 12.6 ± 0.3 Myr (Table 5). Paleodenudation rates computed from the
576 radioactive decay-corrected ^{10}Be concentrations range from 2.8 ± 0.3 to 4.8 ± 0.6 mm kyr $^{-1}$.

577

578

Table 5

579

580 The measured neon isotopic ratios are, within uncertainties, compatible with the
581 spallation-air mixing line in the neon three-isotope diagram, suggesting that atmospheric and
582 spallogenic neon are probably the main components (Fig. 10). However, the uncertainty of the
583 measured $^{22}\text{Ne}/^{20}\text{Ne}$ ratio is too high to ensure the absence of nucleogenic ^{21}Ne . Hence, the
584 excess ^{21}Ne concentrations computed with Equation (5) assuming binary mixing must be
585 considered as maximum cosmogenic ^{21}Ne concentrations. The potential accumulation of
586 nucleogenic ^{21}Ne from the $^{18}\text{O}(\alpha, n)^{21}\text{Ne}$ reaction in quartz can also be considered and
587 computed. The production rate of this reaction is proportional to the U and Th concentrations
588 in the quartz samples (Niedermann, 2002). Observations suggest that U and Th concentrations
589 are higher in secondary quartz coatings or diagenetic silica cements than in crystalline quartz
590 (Balco et al., 2019). Hence, to compute an upper boundary for this nucleogenic contribution,
591 we consider the higher values of reported U and Th concentrations in unetched quartz, i.e.,
592 0.40 ± 0.14 ppm U and 1.06 ± 0.49 ppm Th (Balco et al., 2019, and references therein), which

593 yield a nucleogenic ^{21}Ne production rate of $(0.10 \pm 0.03) \times 10^6$ at $\text{g}^{-1} \text{yr}^{-1}$ (Balco et al., 2019).
594 The closure temperature of the analysed quartz (~ 0.5 mm diameter) is ~ 100 °C (Shuster and
595 Farley, 2005), assuming a cooling rate of 10 °C/Myr. U-Th/ ^4He thermochronometry (Milesi et
596 al., 2020; Maurel et al., 2008) established the exhumation history of the granitic and
597 metasediment massif drained by the Ariège river. They established that this material cooled
598 below the 100 °C isotherm at 40 ± 20 Ma. Combined with the production rate of (0.10 ± 0.03)
599 $\times 10^6$ at $\text{g}^{-1} \text{yr}^{-1}$, this yields a maximum nucleogenic ^{21}Ne build-up of $(4.2 \pm 2.5) \times 10^6$ at g^{-1} .
600 This maximum nucleogenic ^{21}Ne contribution represents between 19 and 34% of the ^{21}Ne
601 excess computed with Equation (5). Given the non-linearity between the burial age and the
602 $^{10}\text{Be}/^{21}\text{Ne}$ ratio (Eq. (4)), this correction should only lower the $^{10}\text{Be}/^{21}\text{Ne}$ burial durations by
603 13 to 19% (Table 5). Because these corrected ages accounting for the maximum theoretical
604 nucleogenic ^{21}Ne contribution are compatible with the non-corrected ages at the 2-sigma
605 level, we chose to consider $^{10}\text{Be}/^{21}\text{Ne}$ burial durations not corrected for this nucleogenic ^{21}Ne
606 build-up in the following discussion.

607

608

609

Figure 10

610

611

612 **6- Discussion**

613

614 **6.1- Dispersion among $^{26}\text{Al}/^{10}\text{Be}$ and $^{10}\text{Be}/^{21}\text{Ne}$ burial durations in caves: implications for** 615 **cave filling dynamics and the accuracy of this dating method**

616

617 $^{26}\text{Al}/^{10}\text{Be}$ and $^{10}\text{Be}/^{21}\text{Ne}$ burial durations observed in the same caves are characterized
618 by intersample dispersions that are larger than the analytical uncertainties. In particular,
619 $^{26}\text{Al}/^{10}\text{Be}$ data from the Niaux-Lombrives-Sabart karstic system (114–142 m above Ariège
620 River) are characterized by an important age dispersion that ranges from <0.5 to >8 Ma.
621 Three causes may explain the large dispersion of this dataset.

622 First, incomplete muscovite removal may have biased ^{26}Al measurements and thus the
623 accuracy of the computed $^{26}\text{Al}/^{10}\text{Be}$ burial durations. Although we took special care during
624 muscovite removal, we cannot rule out this analytical bias that results in under- and
625 overestimated ^{26}Al concentrations and $^{26}\text{Al}/^{10}\text{Be}$ burial durations, respectively.

626 Second, the diachronous filling of these caves spanned several million years (e.g.,
627 Häuselmann, 2007; Sartégou, 2017; Sartégou et al., 2018), consistent with thin-section
628 observations of several coexisting angular and more rounded quartz populations in various
629 states of deterioration according to the deposit (Sorriaux, 1982). The more weathered quartz
630 population, *a priori* of glacial origin, were reworked during alluvial transport phases. These
631 quartz present fractured faces and grinding traces. The presence of materials of different
632 origins in the same stratigraphic layers suggests that high-energy flooding phases within the
633 cavity mixed fresh and older sediments (e.g., Häuselmann, 2007). The high energy of these
634 flows is attested by the presence of blocks ($> 1 \text{ m}^3$). The dispersion of $^{26}\text{Al}/^{10}\text{Be}$ burial
635 durations in a single cave is consistent with Sorriaux's (1982) results and suggests that the
636 studied karstic networks recorded several alluvial phases during the last glacial stages. These
637 processes are probably attributed to climatic variability, particularly the influence of
638 Quaternary glacial cycles on denudation and sediment transport dynamics. As proposed by
639 Sorriaux (1982), the glacial origin of these formations and processes indicates a base level rise
640 during the glaciation phases, as observed in the Austrian Northern Calcareous Alps (Plan et
641 al., 2009). In addition, exchanges between network levels likely occurred. Indeed, because a
642 shaft links the two levels of Lombrives cave, material from the upper level that contributed to
643 the formation of this shaft "contaminated" the lower level. The review by Audra et al. (2006)
644 presents a useful discussion on the dynamics of karst filling.

645 Third, since the analysed detrital material is probably a mixture of heterogeneous
646 sediments derived from various sources, it may have been affected by contrasted and
647 successive burial-exposure cycles within the watershed before entering the caves. Sediments
648 subjected to multiple exposure-burial cycles before their definitive burial in a cave have
649 $^{26}\text{Al}/^{10}\text{Be}$ ratios lower than the theoretical ratio used to compute the ages of the studied
650 deposit (e.g., Laureano et al., 2016; Sartégou et al., 2018). This implies that a significant
651 proportion of our computed burial durations may be overestimated. Indeed, some of the
652 $^{26}\text{Al}/^{10}\text{Be}$ burial durations derived from detrital samples are older than the U-Th ages of the
653 carbonate stalagmitic floors that bracket these deposits (Fig. 11).

654 The bias toward older ages is, in theory, larger for the $^{10}\text{Be}/^{21}\text{Ne}$ dating system than
655 for the $^{26}\text{Al}/^{10}\text{Be}$ pair because the stability of ^{21}Ne implies that the observed concentration of
656 this cosmogenic nuclide integrates all previous exposure-burial cycles. Consistently, our
657 $^{26}\text{Al}/^{10}\text{Be}$ burial durations are on average younger than $^{10}\text{Be}/^{21}\text{Ne}$ ages measured in the same
658 caves (Fig. 12, Tables 2 and 4). Hence, the youngest $^{26}\text{Al}/^{10}\text{Be}$ ages are probably more
659 representative of the last cave filling event for the Niaux-Lombrives-Sabart and Enchantées

660 caves. Indeed, the $^{26}\text{Al}/^{10}\text{Be}$ age distributions of these caves have modes respectively centred
661 at approximately 3 and 5 Ma (Fig. 12). However, this cave level is characterized by multiple
662 phases of deposits. Because the burial ages of deposits in the upper Moutons caves (385–576
663 m above Ariège River) are too old for $^{26}\text{Al}/^{10}\text{Be}$ dating, we must rely on $^{10}\text{Be}/^{21}\text{Ne}$ burial
664 durations to date the last filling of these caves, considering that these $^{10}\text{Be}/^{21}\text{Ne}$ durations are
665 probably maximum age constraints.

666

667

Figure 11

668

Figure 12

669

670

671 Our data and their potential interpretation have strong implications for the significance
672 and accuracy of the determined burial durations. To ensure that the age of karstic alluvium is
673 representative of the incision rate of a valley, different approaches and geochronological
674 methods should be combined whenever possible, including stratigraphy, karstic
675 geomorphology, geochemical and/or luminescence dating (to determine burial duration and
676 provenance), and particle size and microscopic analyses. Notably, complementary absolute
677 ages of carbonate concretions are quite useful. Electron spin resonance dating of samples
678 from the Calamès and Sédour massifs was attempted, but found to be unreliable in this area
679 (see Chapter 6 of Sartégou, 2017, in which the methodological limits are discussed);
680 therefore, we exclude those data from further discussion.

681

682 **6.2- Relationships between incision process, stepped karstic systems, the water table, and** 683 **epiphreatic galleries**

684 Our chronostratigraphic interpretation of the studied caves relies on the assumption
685 that large karstic networks formed in a single step at the water table. According to this simple
686 model, once a horizontal network has been formed, the water table drops and the old networks
687 are abandoned and fossilized. New karstic networks are then initiated at the level of the new
688 water table. Each cave level at a given elevation is thus associated with specific
689 geomorphological and climatic conditions that are favourable to its development. However,
690 network formation is likely much more complex in stepped sub-horizontal caves that bear
691 markers of successive circulation episodes in (epi-)phreatic regimes. In such cases, individual

692 cave levels may have different implications for the system development (e.g., Mocochain et
693 al., 2009; Audra and Palmer, 2011, 2013).

694 In the Ariège valley, the Niaux-Sabart-Lombrives network is typical of old karst
695 networks that underwent multiple filling phases over more than 100 kyr under the control of
696 successive glaciation stages. The speleogenesis of these networks probably simply occurred at
697 the level of the water table (records discussed in Section 3.3). However, subsequent major re-
698 flooding events seem to have profoundly reshaped some of the main karstic networks. Thus,
699 the entire geochronological dataset of the Ariège caves was not solely controlled by valley
700 incision since the Miocene. This must be considered when interpreting the significance of
701 some burial durations, such as for samples Ni12-02, Ni13-08, Ni13-10, Ni13-12, Ni13-13,
702 LS1, and LO1. As our interpretations are based on solid petrographic and morphological
703 arguments, we consider them to be sound interpretations based on the present state of
704 knowledge.

705 If these considerations are correct for large kilometric networks, confirming their
706 validity for smaller hectometric cavities (Esplourgues, Enchantées, and Bédeilhac caves) is
707 difficult because sedimentary sections are rarely visible and deposits do not appear
708 polyphased. The relatively low dispersion of the $^{26}\text{Al}/^{10}\text{Be}$ burial durations in these caves (Fig.
709 12) suggests that the filling of these caves occurred in a single depositional phase.

710 Therefore, we employ a cautious approach to interpreting our data, and consider the
711 size and shape of the gallery before extrapolating the measured burial durations to an entire
712 gallery level. In the following section, we rule out suspect $^{26}\text{Al}/^{10}\text{Be}$ and $^{10}\text{Be}/^{21}\text{Ne}$ burial
713 durations by considering gallery morphologies and stratigraphic and petrological constraints.

714

715 **6.3- Ariège River valley incision since 13 Ma (Langhian)**

716 Considering the above-mentioned potential complications, our calculated burial
717 durations indicate the likely evolution of Tarascon valley incision since the Langhian (c. 13
718 Ma), which corresponds to the burial duration derived from a sample taken from Mirouge
719 cave, the most perched sampled cavity (Fig. 12). From the Langhian to the Tortonian, the
720 range of possible incision rates deduced from $^{10}\text{Be}/^{21}\text{Ne}$ burial durations vary between 80 and
721 310 m Myr^{-1} . These rates were measured in three samples from the Mirouge, Souloubrié,
722 and Moutons caves (samples LUJ1, SO1, and MOU1). These poorly constrained results may
723 be explained by the low preservation of cavities at these elevations and the small amount of
724 alluvium to be sampled. From the Tortonian to the Mio-Pliocene transition, the range of
725 possible incision rates varies from 20 to 30 m Myr^{-1} . A "continuous" incision rate of

726 approximately 50 m Myr⁻¹ from the Langhian to the Mio-Pliocene transition is quite
727 plausible, and could have established the karst networks if it were moderated by more or less
728 pronounced stationary phases (Audra, 1994). Inducing incision rates after the Mio-Pliocene
729 transition is difficult due to perturbations introduced by glacial re-flooding events. From the
730 Tortonian (~9 Ma) to the present, several scenarios are possible. Considering the upper and
731 lower bounds of the burial durations obtained in the Massif du Calamès, we estimate
732 maximum and minimum Plio-Quaternary incision rates of ~200 and ~25 m Myr⁻¹,
733 respectively.

734 This range of incision rates is consistent with those estimated in the easternmost Têt
735 River valley (Sartégou et al., 2018), further west in the Salat watershed of the Arbas massif
736 since the Pliocene (110 m Myr⁻¹; Genti, 2015), and on the Garonne terraces (maximum rates
737 of ~46 m Myr⁻¹; Stange et al., 2014). Moreover, our dataset is consistent with reconstructions
738 of palaeo-weathering surfaces and their associated age ranges (e.g., Lagasquié, 1963; Monod
739 et al., 2016a, b). The weathering profiles associated with highly elevated and low-relief
740 surfaces in the study area may have developed between the Eocene-Oligocene and the late
741 Miocene (e.g., Calvet, 1996) and seem to be related to the Pyrenean orogeny. Monod et al.
742 (2016a, b) considered that the Aston-Beille surface was denuded by up to 50 m since the
743 Miocene and partially dissected by incision, and our dataset seems to corroborate these
744 assumptions. By extrapolation of these incision rates, the Beille plateau (1,800–2,000 m high)
745 would have formed between the Rupelian (~32 Ma) and the middle Burdigalian (~18.5 Ma).

746 Because incision during the Langhian-Serravallian period may be attributed to either
747 an undocumented uplift or a global eustatic base-level fall (as during the Langhian-
748 Serravallian transition; Haq et al., 1987), the mechanisms driving incision at that time remain
749 unknown. Variations in incision rates seem less marked in the Ariège than in the Têt River
750 valley (Sartégou et al., 2018), which may indicate a more "linear" process, despite the
751 numerous possible scenarios. This finding suggests a possible post-orogenic rejuvenation of
752 the relief during the Langhian-Messinian. On the other hand, the variable incision rates
753 observed in the Têt River valley, which was slightly glaciated and is connected to the
754 Mediterranean Sea, imply that the contribution of external forcings cannot be excluded there.
755 Further west along the Pyrenees, notably in the Eaux-Chaudes and Balaitous-Panticosa
756 massifs, Bosch et al. (2016) used thermochronological data to document significant
757 exhumation between 20 and 30 Ma. They also used apatite (U-Th)/He data to document
758 accelerated exhumation of the Balaitous-Panticosa granitic massif at ~8–9 Ma. Indeed,
759 previous studies of the southern foreland basin documented post-orogenic cooling (Jolivet et

760 al., 2007; Labaume et al., 2016; Bernard et al., 2019). Because these areas are all in the
761 central Pyrenees, this last exhumation event seems coincidental with the recorded incision.

762 Attributing incision during more recent periods (especially during the Tortonian to
763 Pliocene) to a single mechanism remains difficult. Stange et al. (2014), Delmas et al. (2015),
764 and Mouchéné et al. (2017) suggested the dominance of external forcings during the
765 Quaternary. Isostatic rebound, which is related either to surface processes such as denudation
766 (Genti et al., 2015) or to the suppression of the thick ice layer present in these internal zones
767 during the last glacial maximum (Crest, 2017), may explain part of the observed uplift.

768

769 **6.4- Palaeodenudation rates**

770

771

Figure 13

772

773 Our derived palaeodenudation rates for the Ariège River valley characterize a
774 significant increase in denudation between the Langhian (14 Ma) and the Pleistocene (2.6–0
775 Ma) (Table 3, Fig. 13). Palaeodenudation rates ranged from 3 to 10 mm kyr⁻¹ between 13 and
776 6 Ma, from 10 to 50 mm kyr⁻¹ during the Pliocene and Pleistocene, and exceed 100 mm kyr⁻¹
777 (0.1 mm yr⁻¹) during the Holocene. Modern ¹⁰Be-derived denudation rates for river sands of
778 rivers draining the Pyrénées and Massif Central are 50–300 and 50–100 mm kyr⁻¹,
779 respectively (Molliex et al., 2016; Olivetti et al., 2016; Sartégou et al., 2018).

780 Plio-Pleistocene palaeodenudation rates of 3–10 mm kyr⁻¹ are similar to those
781 observed in the nearby Têt River valley. However, the Ariège records a greater increase in
782 denudation than the Têt record (Sartégou et al., 2018). These findings suggest that increased
783 denudation was triggered by the global cooling that occurred during this time period due to
784 the appearance of high-amplitude and high-frequency Pleistocene glacial cycles (Molnar,
785 2004; Herman et al., 2013). However, the extreme increase of denudation rates in the Ariège
786 record (i.e., by nearly three orders of magnitude) is rather peculiar, and suggests that climate
787 was not the only factor influencing denudation in this basin; uplift and enhanced geodynamic
788 flux in a collisional regime are more probable forcings. This interpretation appears consistent
789 with apatite fission track data acquired in the nearby Arize, Trois-Seigneurs, Aston, and
790 Bassiès massifs (Gunnell et al., 2009; Vacherat et al., 2016). Furthermore, (U–Th)/He dating
791 of zircons and apatites show that minimal denudation occurred since the Eocene-Oligocene
792 (Morris et al., 1998; Fitzgerald et al., 1999; Sinclair et al., 2005; Denèle, 2007; Jolivet et al.,
793 2007; Gunnell et al., 2009; Vacherat et al., 2016). The agreement between long-term

794 exhumation data and our data therefore suggests a predominantly tectonic control on
795 Pyrenean incision. This interpretation is consistent with long-term ^{10}Be -derived
796 palaeodenudation rates in Central Asia, which was uplifted during the Alpine collision (e.g.,
797 Puchol et al., 2017).

798 On a shorter timescale, previous cosmogenic denudation rate estimates in the northern
799 Pyrenees were mainly derived from piedmont alluvial terraces (Stange et al., 2014; Delmas et
800 al., 2015; Nivière et al., 2016; Mouchéné et al., 2017) or from preliminary dating of alluvial
801 cave sediments (Genti, 2015; Sartégou et al., 2018). Pleistocene and Holocene alluvial
802 terraces have primarily yielded denudation rates similar to those presented herein (Delmas et
803 al., 2015; Mouchéné et al., 2017). Denudation rates determined from Ariège tributary deposits
804 (Aston-Beille catchment) are also consistent with our data (Crest, 2017).

805 Grain size-dependent ^{10}Be concentrations (e.g., Puchol et al., 2014) were not observed
806 in this work. The most probable cause of significantly higher palaeodenudation rates is
807 therefore the origin of the clasts. Certain similar lithologies are observed at separate and very
808 remote locations in the watershed. In the case of these lithologies, discriminating whether
809 pebbles originate from proximal or distal locations in the catchment and whether they were
810 produced from steeper or gentler slopes, during particular climatic events, or during stochastic
811 events (e.g., landslides, flash floods) is therefore not possible.

812 **7- Conclusions**

813 We proposed a geochronological framework for detrital materials filling caves
814 developed in the Ariège River valley based on the $^{26}\text{Al}/^{10}\text{Be}$ and $^{10}\text{Be}/^{21}\text{Ne}$ burial
815 chronometers. Although our results display significant scatter, they are coherent with the
816 stratigraphic order and allow us to quantify the incision of the Ariège River valley since the
817 Langhian age. Incision probably began during the early Miocene, at c. 13 Ma, due to
818 geodynamic mechanisms that have not yet been identified. The average incision rate of the
819 Ariège River deduced from our cave dating is on the order of 13–50 m Myr^{-1} , similar to
820 incision rates in the Salat valley to the west (Genti, 2015) and the Têt valley to the east
821 (Sartégou et al., 2018).

822 Our data highlight a broad dispersion of burial durations within a single karstic
823 network, as illustrated by the Niaux-Lombrives network in the Cap de la Lesse massif. The
824 Niaux-Lombrives-Sabart stepped karstic system is an ancient karst with polyphased fillings
825 spanning several glaciations. During the last glacial cycle, the Ariège River valley was almost
826 completely covered by ice, which, upon melting, generated new floods in previously

827 developed karstic networks (as observed in the Alps; Audra et al., 2006; Plan et al., 2009).
828 These re-flooding events mixed newly buried sediments with sediments that had been
829 shielded from cosmic radiation for several million years. Our data therefore include notable
830 uncertainties that arise from the complexity of the investigated systems instead of analytical
831 biases. After accounting for these potential complications, our data demonstrate the
832 preponderant influence of external forcings on incision. Our results also illustrate that not all
833 stepped karstic systems are directly representative of the incision processes in nearby valleys.

834 This work demonstrates that ^{10}Be - ^{21}Ne paired nuclides are a useful complement to the
835 ^{26}Al - ^{10}Be cosmogenic pair because the burial duration method can be extended beyond 5 Ma,
836 and coherent ages can be obtained to 15 Ma.

837

838 **Acknowledgements**

839

840 This work was funded by the BRGM within the framework of the RGF Pyrenees
841 program. We especially thank the Speleological Committee of Ariège (CDS09), and in
842 particular the Spéléo Club of Haut-Sabartez (SCHS) and the Spéléo Club of the Arize (SCA,
843 Saurat cave), for sharing their invaluable knowledge of the area and their topographic dataset,
844 and Luc Wahl who accompanied us during several sampling campaigns. We thank Max Nicol
845 for his knowledge of Mirouge cave. This work was only possible with the cooperation of
846 DRAC Midi-Pyrénées, which authorized P. Sorriaux to observe and sample Niaux cave
847 within the framework of the “sampling of pre-Würmian deposits for cosmogenic nuclides and
848 U/Th dating” project. Mr. Bodin allowed us to visit and sample Lombrives cave. René Gailli,
849 the curator of the Bédeilhac cave, and Marina Laborde allowed us to walk and sample at our
850 leisure. We sincerely thank Martin Stokes, Jo De Waele, Darryl Granger, Fin Stuart, and
851 Philipp Häuselmann for constructive comments that improved the quality of the manuscript.
852 The ASTER AMS national facility (CEREGE, Aix-en-Provence, France) is supported by the
853 INSU/CNRS, the ANR through the “Projets thématiques d’excellence” program for the
854 “Equipements d’excellence” ASTER-CEREGE action, and IRD.

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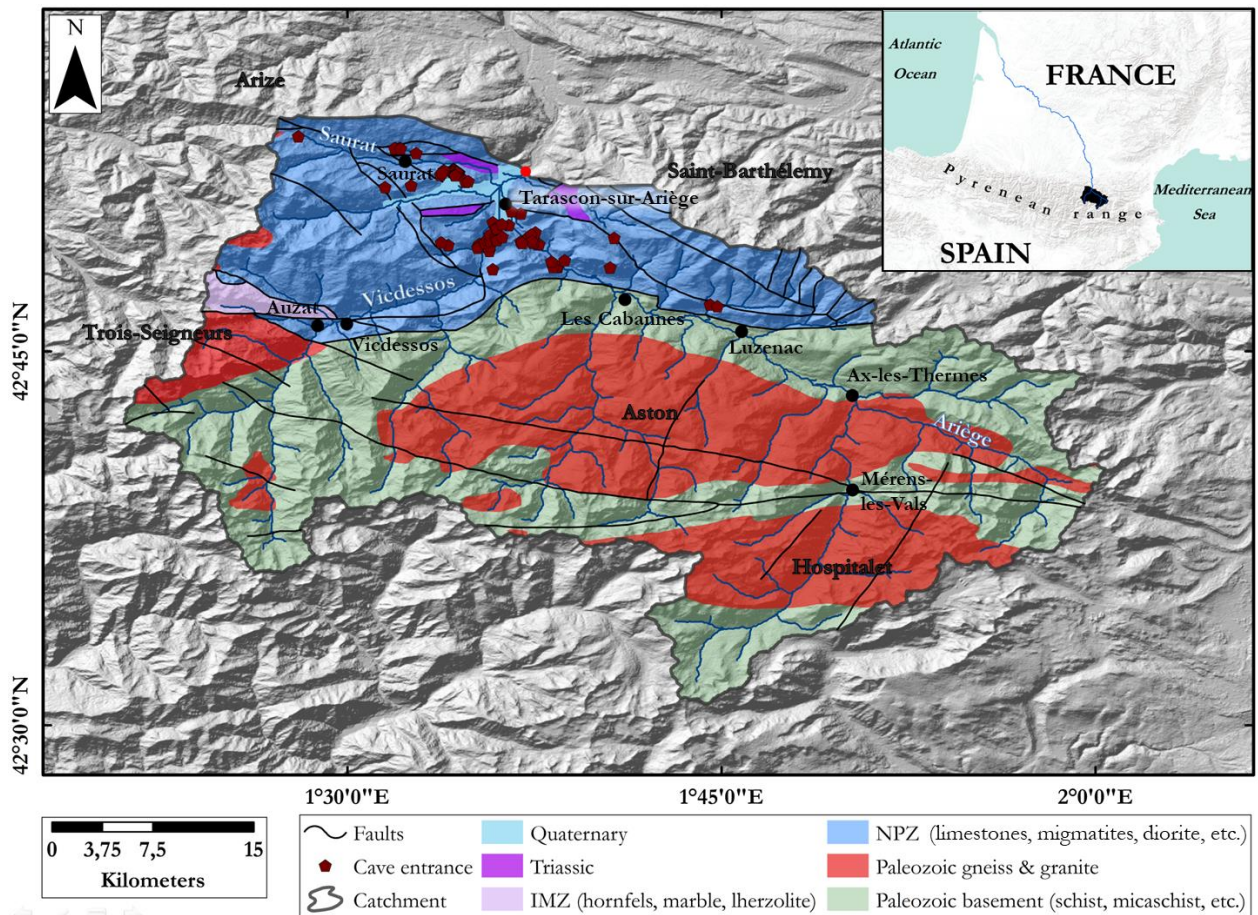
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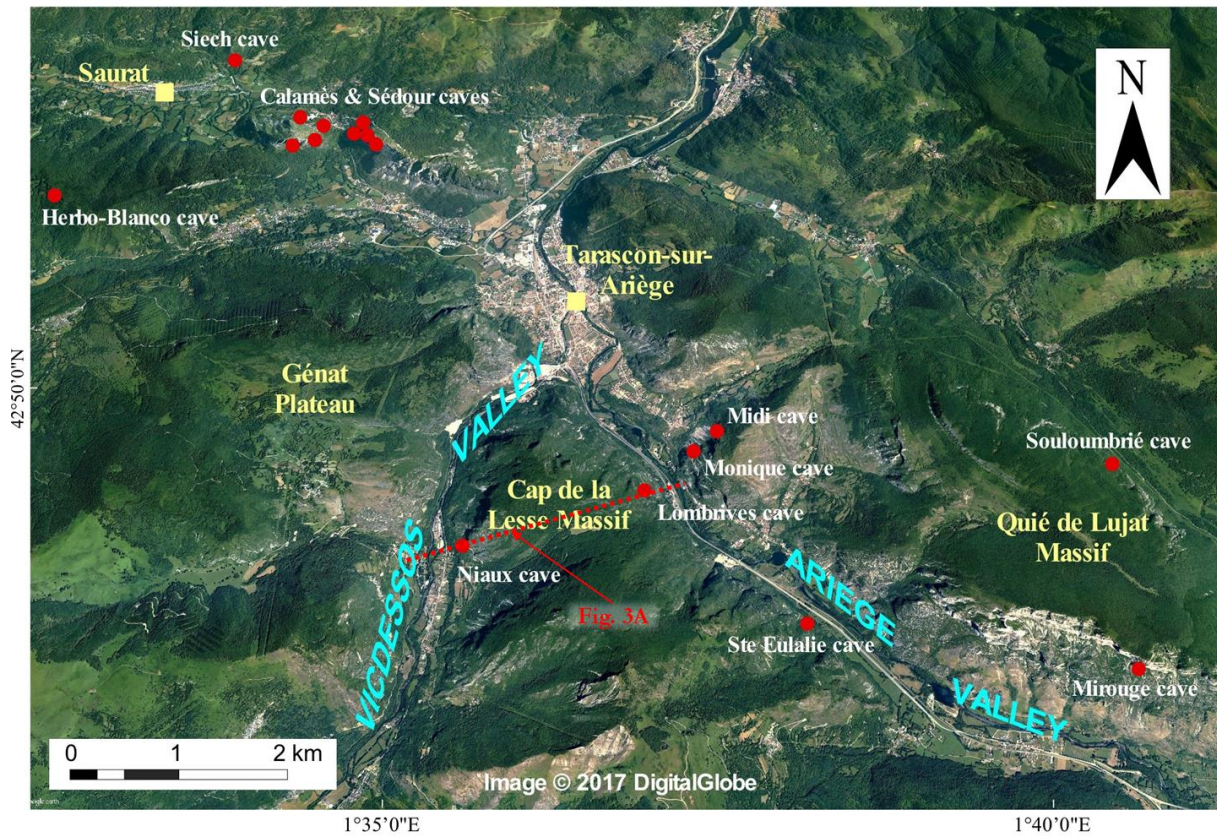
1159 **Figures**

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1162 Fig. 1. Lithostructural features of the Ariège catchment (based on IGME-BRGM data). Alluvial
 1163 sediments trapped in karstic networks originated from Palaeozoic crystalline massifs (gneisses,
 1164 granites, and metasediments) before joining the limestones of the North-Pyrenean zone (NPZ) or from
 1165 the varied lithologies of the internal metamorphic zone (IMZ). The area is tectonically complex, and
 1166 the Cretaceous limestone cliffs of the NPZ are dotted with caves that served as shelters during
 1167 prehistoric times and during religious wars. The watershed also includes glacial deposits at high
 1168 altitudes.



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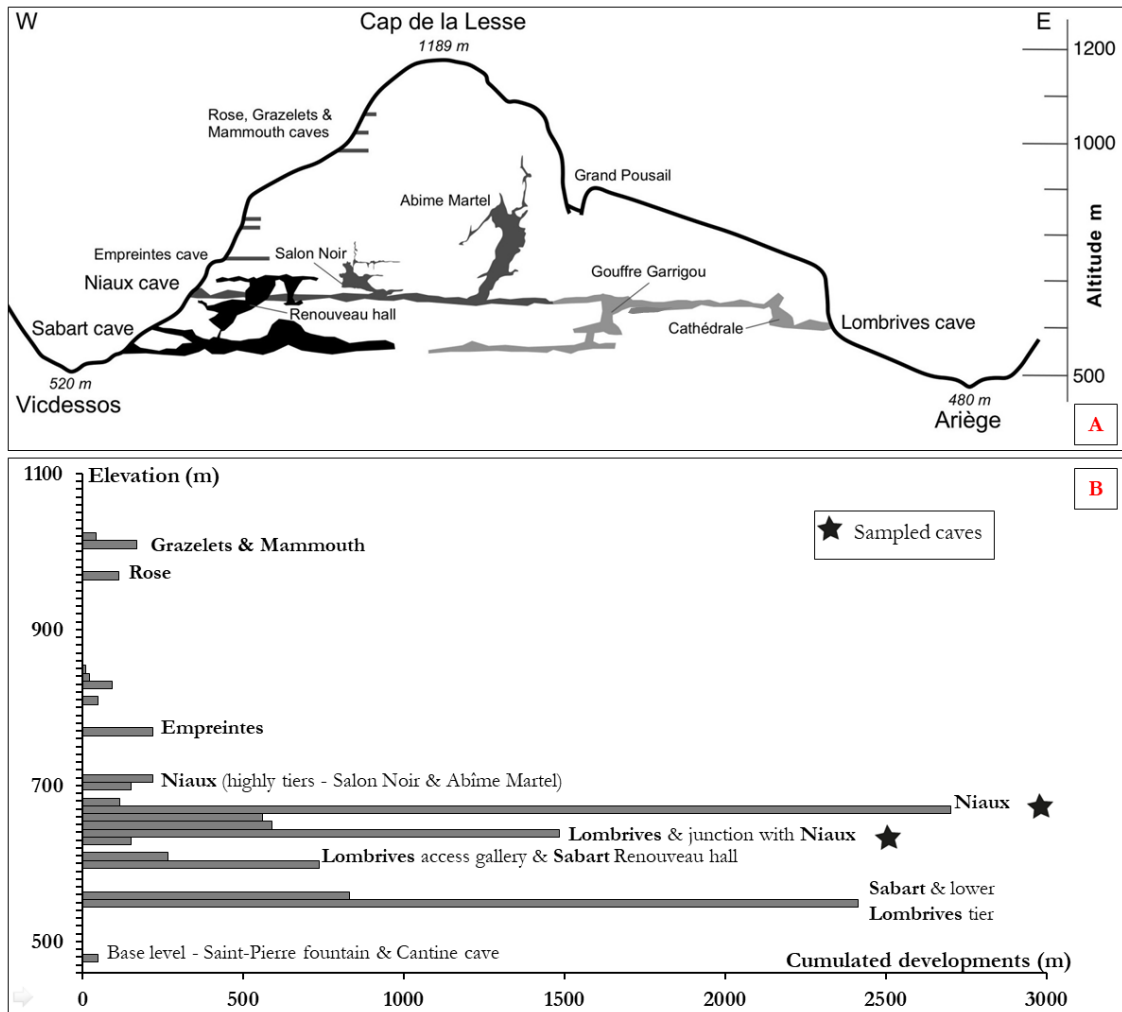
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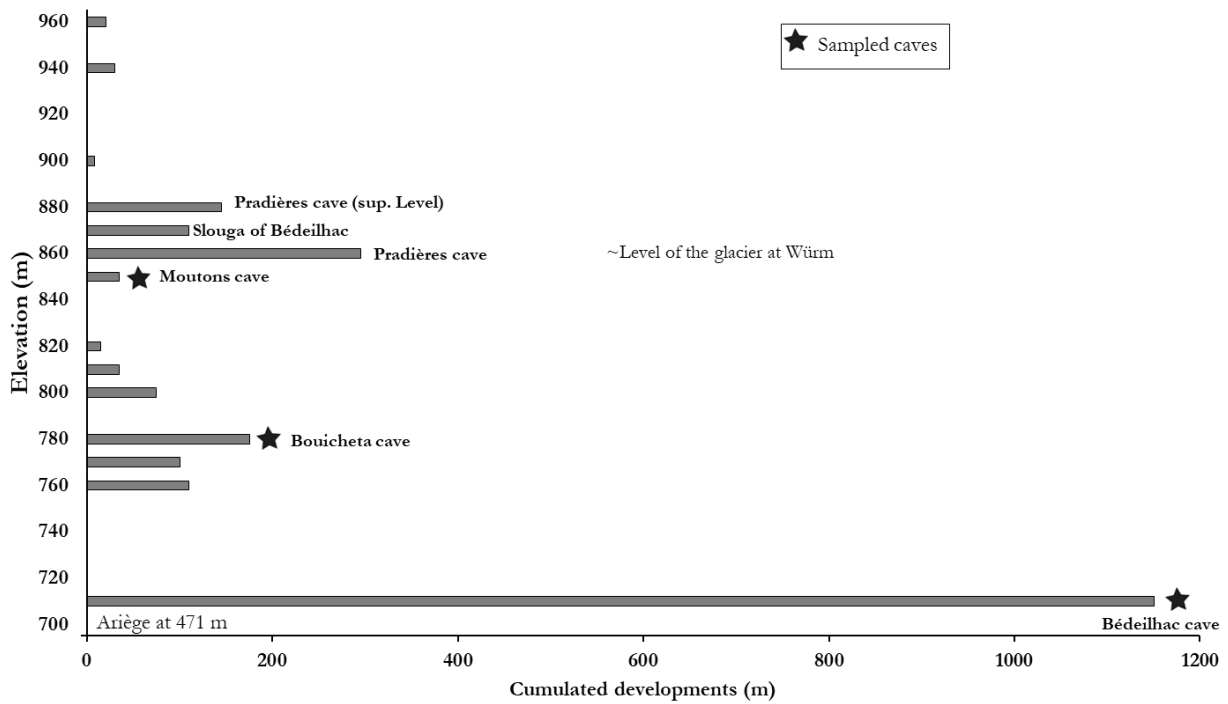
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Fig. 2. The Ariège River valley near Tarascon-sur-Ariège, highlighting the spatial distribution of the karstic network within the massifs. Epiphreatic levels are identifiable at the Quié de Lujat (east bank), Cap de la Lesse (Fig. 3), Calamès (Fig. 5), and Sédour (Fig. 4) massifs (west bank). These karstic networks developed due to variations in the levels of the Ariège River and its tributaries, the Videssos and Saurat Rivers. The perched levels have probably been trimmed over time by the removal of the flanks of the massifs.



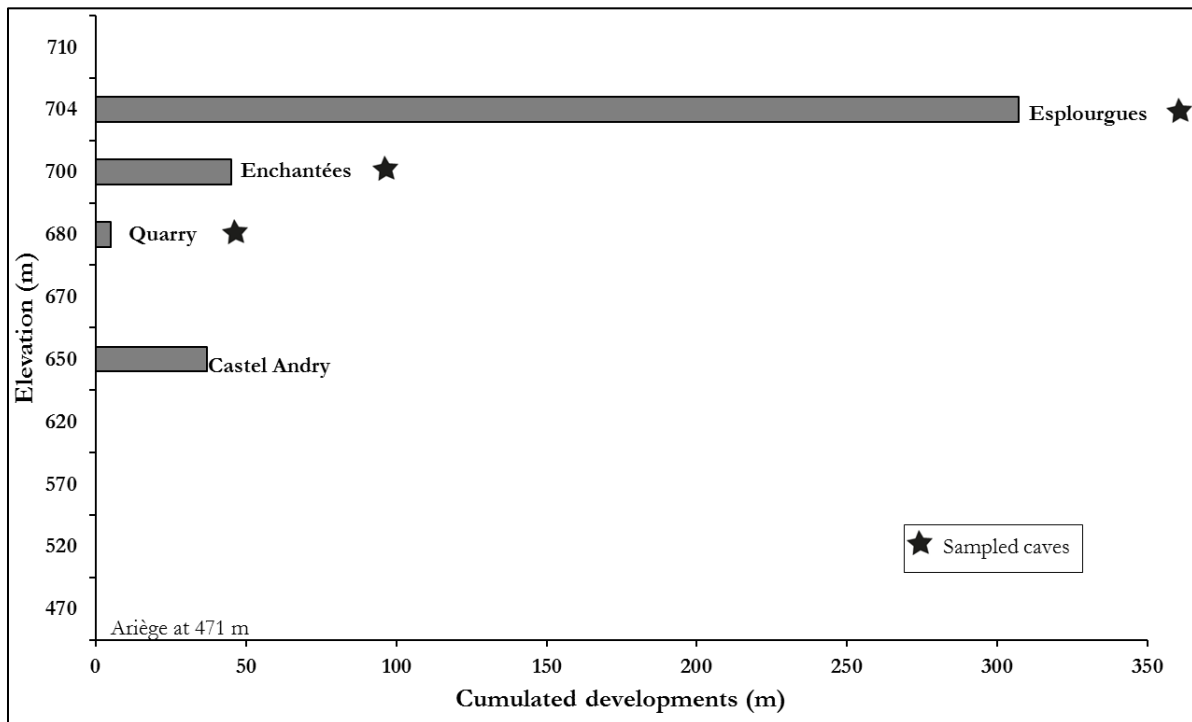
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1177 Fig. 3. Upper part (A) - Schematic cross-section of Cap de la Lesse massif (see Fig. 2 for location),
 1178 showing the spatial distribution of the karstic network. Lower part (B) - Eight epiphreatic levels,
 1179 and some galleries are vertically connected. This network developed from the Ariège and
 1180 Videssos Rivers. The floors linked to the Niaux-Lombrives-Sabart network are the most developed
 1181 (altitudinal distribution shown at bottom) and are linked by a vertically branching network that does
 1182 not originate from the surface. Perched levels were probably trimmed over time due to the removal of
 1183 the flanks of the massif.



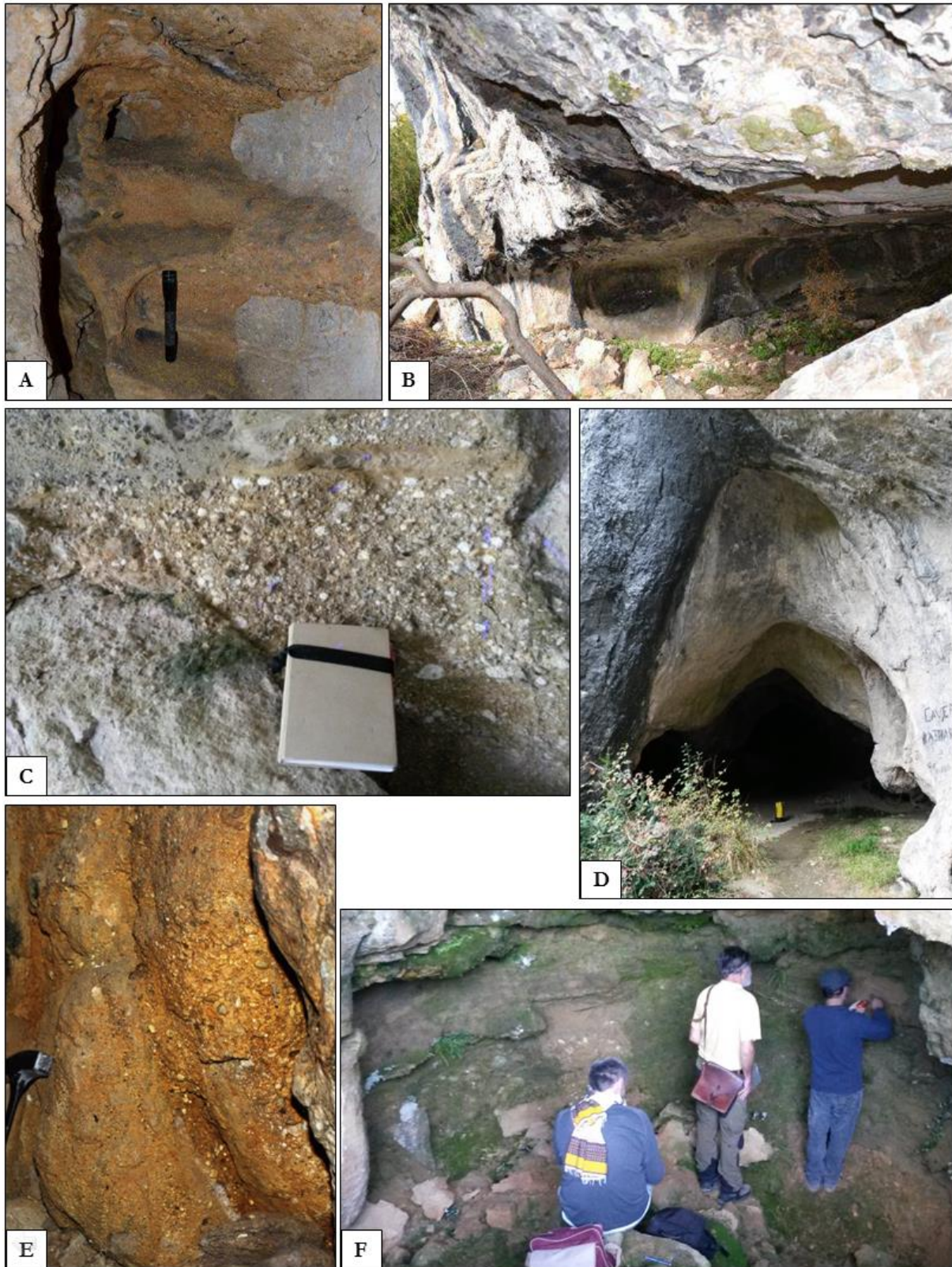
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1185 Fig. 4. Altitudinal distribution of caves of the Sédour massif. At least six epiphreatic levels are
 1186 identifiable, which formed from the Saurat River, an Ariège River tributary. The Bédailhac floor is the
 1187 most developed (due to its significant length, it is not fully represented). Perched levels were probably
 1188 rimmed over time due to the removal of the flanks of the massif.



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1190 Fig. 5. Altitudinal distribution of caves of the Calamès massif. At least four epiphreatic levels are
 1191 identifiable, which formed from the Saurat River, an Ariège tributary. This massif has been prospected
 1192 by archaeologists. Perched levels were probably trimmed over time due to the removal of the flanks of
 1193 the massif.



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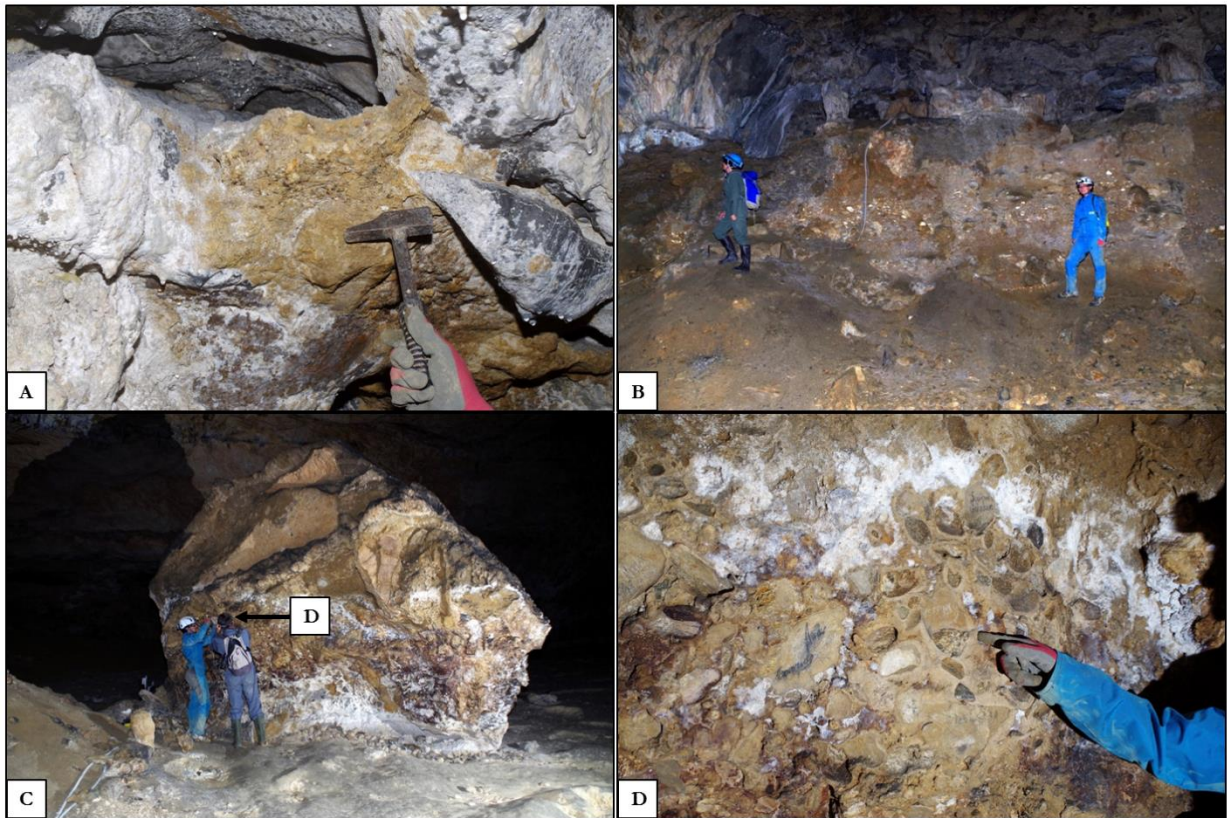
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Fig. 6. A) Well consolidated sandstone plating along the wall of the Bouicheta cave. (B) The entrance to Moutons cave: convectional niches and remnants of stalagmitic floors are visible. (C) The alluvial deposits near an entrance to the Esplourgues cave. These highly altered coarse to pebbly deposits are part of a stratigraphic sequence that includes well consolidated clayey sands. These deposits indicate an insertion and contain fluid escape structures. (D) An entrances to the Esplourgues cave, showing large niches. (E) Very coarse, rubified, and highly altered sandy deposits cemented along the wall of

1201 the Mirouge cave, in Quié de Lujat massif. (ph. By M. Nicol). (F) Sampling of medium-sand levels of
1202 the Quarry cave stratigraphic sequence. Below these sandy deposits are altered heterometric pebbles.

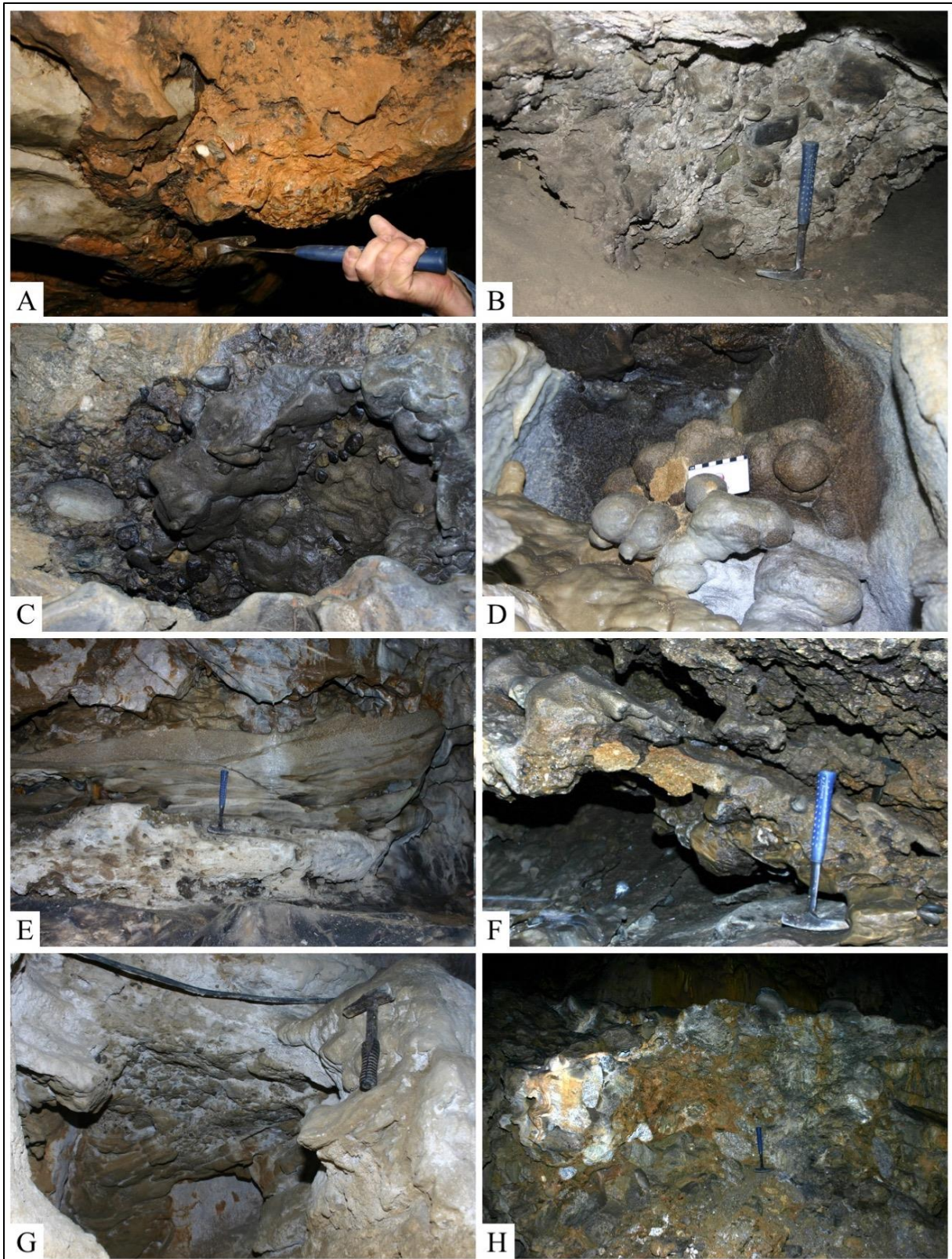


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1204 Fig. 7. Sampling in the Bédeilhac cave (ph. Sorriaux). (A) Deposits of medium to coarse sands on the
1205 wall of the Vidal gallery. (B) A section of the entry room in which a World War II aircraft hangar was
1206 installed. The thick alluvial deposits reflect high energy currents. Pebbles are altered and the deposits
1207 are rubified. (C) Collapsed stalagmitic pillar (“Tomb of Roland”). This large column has been
1208 displaced by currents and is probably very strong, as shown by its surface morphologies. (D) Granitic
1209 pebbles and medium to coarse sands are cemented at the base of the Tomb of Roland pillar.



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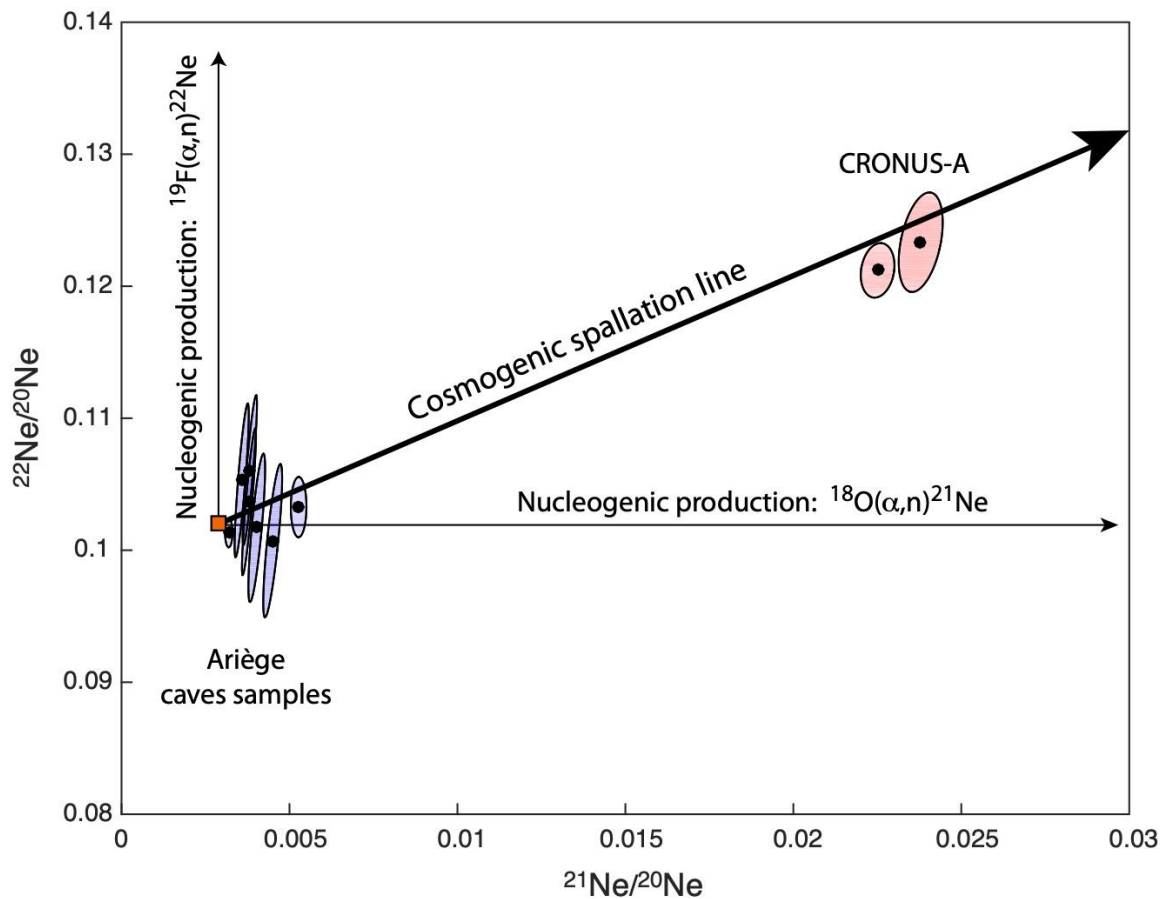
1211 Fig. 8. Three successive sedimentary deposits were investigated in the Niaux and Lombrives caves
 1212 (ph. P. Sorriaux and V. Guinot). Each deposit comprises two units: the lower units are detritic and of
 1213 alluvial origin, and thus indicate the hydrology of the network, whereas the upper units mainly
 1214 comprise calcitic deposits corresponding to drying periods when the karst evolved above the base
 1215 level. These conditions are similar to the present-day cave environment. Some stalagmitic floors have
 1216 been U-Th dated (Sorriaux, 1981; 1982; Bakalowicz et al., 1984). This reference outcrop is in Niaux
 1217 cave, a few hundred meters from the entrance at approximately 670 m a.s.l. F1 is the oldest alluvial
 1218 conglomerate; it includes a calcitic deposit (F2c) with two stalagmitic pillars, the oldest parts of which
 1219 are older than 350 ka. The basic stalagmite floor (F2c) can have a thickness of several meters slightly
 1220 further into the gallery. The tops of the pillars are covered by an alluvial conglomerate (F3) that is
 1221 sealed behind the left pillar by a stalagmitic floor (F4) and is not visible on the photograph, which
 1222 formed between 200 and 90 ka. Conglomerate F3 corresponds to a pre-Würm glacial phase, which is
 1223 poorly known at the surface (Sorriaux et al., in press). The sands (F5) at the bottom of the photo are of
 1224 fluvio-glacial origin corresponding to the Würm major glacial phase (90–20 ka), which reactivated the
 1225 karst. Here, the sands are covered by clayey-calcitic sediments (F6) corresponding to the current
 1226 drainage period.



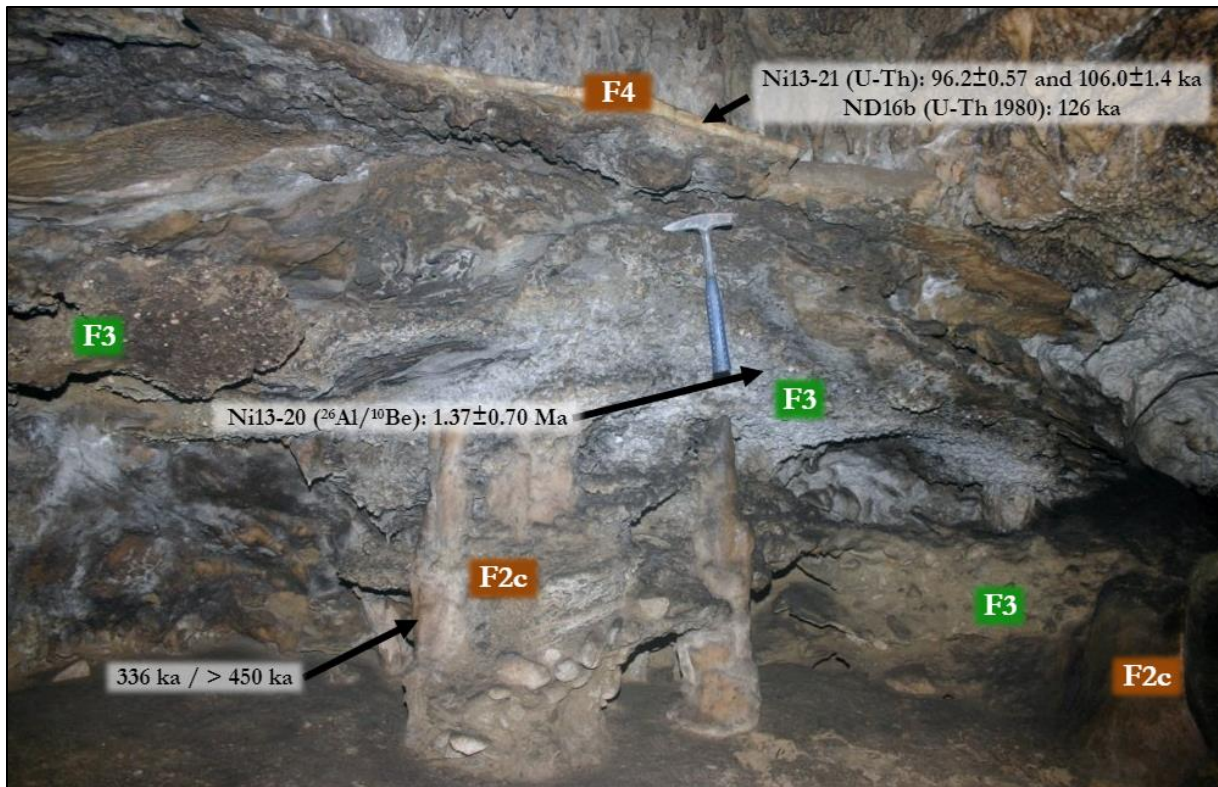
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Fig. 9. Photographs of some of the Niaux and the Lombrives cave sampling sites. The F1 and F3 formations are described in the text after Sorriaux (1982). All photos are from P. Sorriaux. (A) Site Ni13-05. (B) Site Ni13-17. (C) Site LO5 at the *Salines* location. This conglomerate is part of the F1 alluvial formation. (D) Site LO3 at the entrance of the Vierge gallery. A soutirage of the shaft is observed at this site. Medium to coarse grained sandstone balls are highly concreted. These intergranular concretions typically formed in drowned passages, not very far below the water table (shallow sumps), in which flooding was only occasional (allowing sufficient time for these sands to

1235 partly solidify). (E) Site LO7 near the Akka underground lake. The F3 alluvial formation (comprising
 1236 conglomerates surmounted by coarse- to fine-grained sandstones) seals a red clay deposit. This
 1237 sampling point clearly illustrates a channel morphology. (F) Site LO6 near Akka Lake. This F1
 1238 conglomerate is a well-consolidated conglomerate comprising gravel and crystalline pebbles in a
 1239 sandy matrix. (G) Site LO8 in the Mamelle gallery. The sample was taken from a small niche to the
 1240 left of the photograph. The material is very fine grained and hardened. Balls are noticeably altered, and
 1241 a well-indurated coarser level seals this deposit. (H) Site LO2 in the Grand Chaos room. This
 1242 formation includes highly weathered granitic pebbles in a fine to medium sand matrix. A floor seals
 1243 this formation, into which blocks of unaltered granites are inserted.



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 1245 Fig. 10. Neon three-isotope diagram of quartz samples from caves of the Ariège River catchment.
 1246 Most data from the Ariège samples (blue ellipses) are compatible with the atmosphere-spallation
 1247 mixing line, similar to the CRONUS-A quartz standards (red ellipses).



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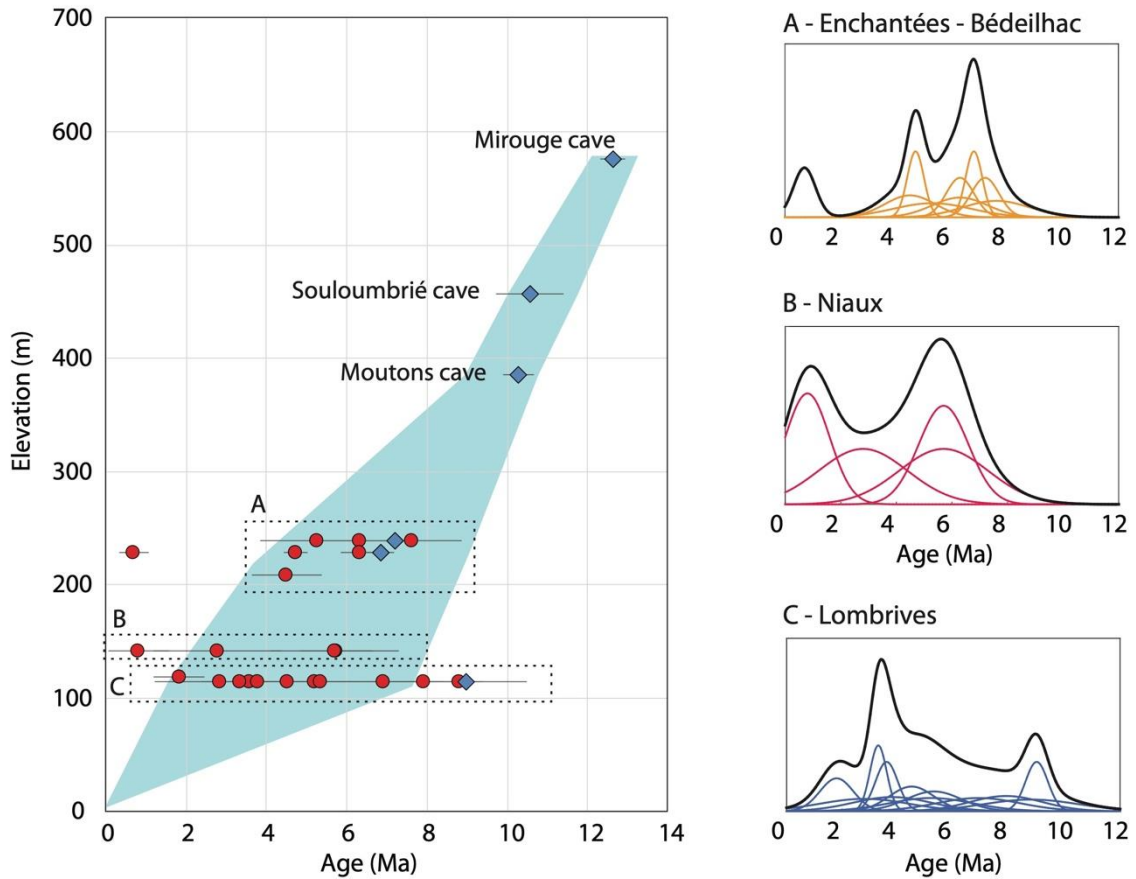
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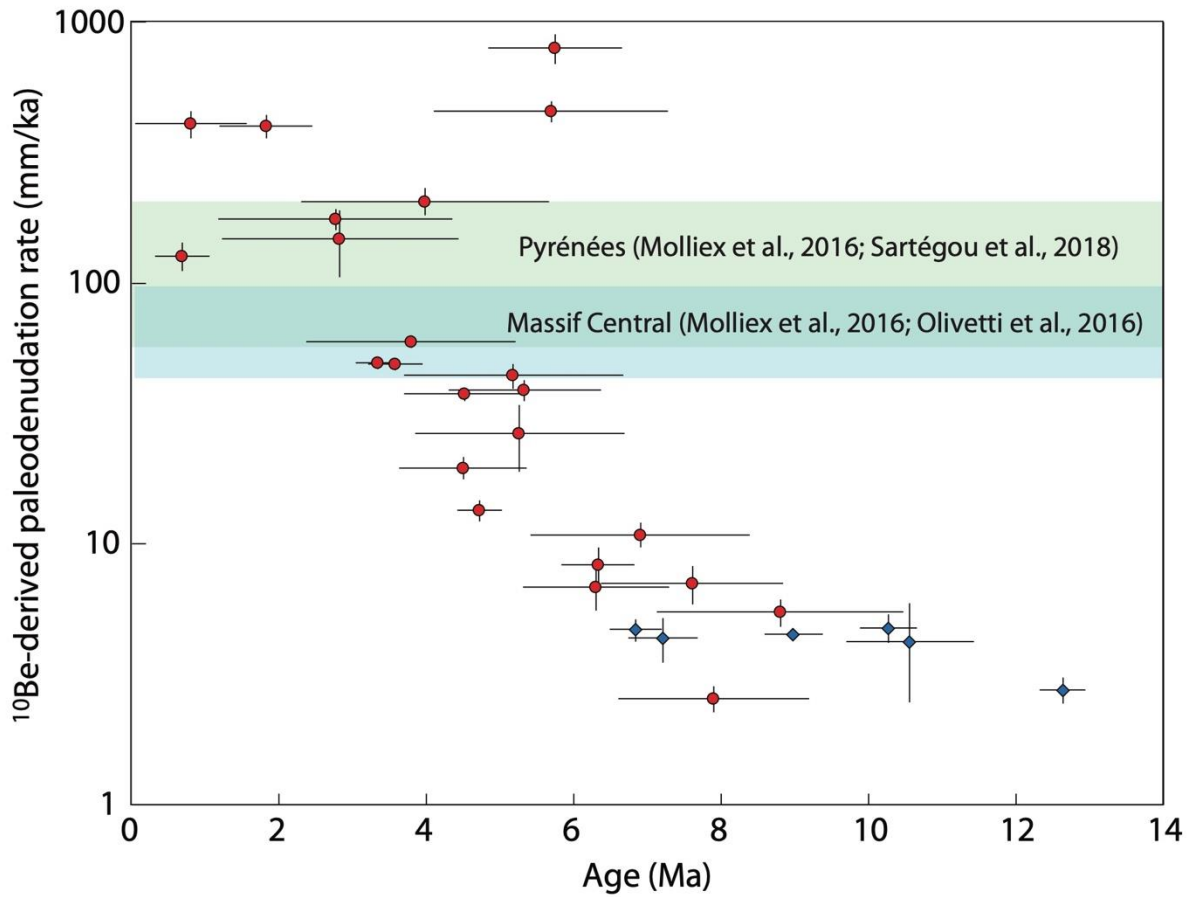
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Fig. 11. A representative photograph of polyphased fillings in Niaux cave. The F3 sandstones are sealed by stalagmitic floors dated between ~150 and ~90 ka (Ni13-21; Sorriaux et al., 2016). The roof of the calcite complex (F2c) is older than c. 336 ka outside the concretion and ~450 ka (minimum ages) at the centre of the concretion (Ni13-18; Sorriaux et al., 2016). The F3 Formation yields heterogeneous burial durations (see Ni13-15, Ni13-20, and Ni13-23, Table 3).



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1255 Fig. 12. Elevation vs. filling age of the Ariège karstic networks since 14 Ma (Langhian). Burial
 1256 durations were determined from the $^{26}\text{Al}/^{10}\text{Be}$ (red dots) and $^{10}\text{Be}/^{21}\text{Ne}$ ratios (blue diamonds) of
 1257 detrital sediments filling the caves. Burial durations were discarded for poorly purified samples
 1258 containing residual muscovite. Small variations in the relative altitudes, related to gaps in the
 1259 networks, are not represented in this plot. (A–C) Burial age distributions of samples from the
 1260 Enchantées-Bédeilhac, Niaux, and Lombrives caves, respectively.



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1263 Fig. 13. ^{10}Be -derived palaeodenudation rates vs. filling age in the Ariège watershed since 14 Ma
1264 (Langhian). ^{10}Be concentrations are corrected for radioactive decay. Blue dots were obtained from
1265 samples analyzed for ^{21}Ne . The red dots were obtained from samples analyzed for ^{26}Al . Modern ^{10}Be
1266 palaeodenudation rates were obtained from modern river sands in the Pyrenees and Massif Central
1267 (Molliex et al., 2016; Olivetti et al., 2016; Sartégou et al., 2018).
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1270 **Tables**

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Massif	Cave	Cave ID	Latitude (°)	Longitude (°)	Elevation (m a.s.l.)	Relative elevation (m)	Length (m)
Cap de la Lesse	Sainte-Eulalie	ASAR115	42.809997	1.636363	570	20	62
	Lombrives network	ASAR110	42.823389	1.616056	605	114	7000
	Tunnel Lombrives	ASAR111	42.823547	1.615785	610	119	19
	Niaux network	ABOU101	42.819667	1.593750	672	142	4000
Sédour - Calamès	Bédeilhac Quarry	ASAR119	42.872949	1.560063	680	209	5
	Enchantées	ASAR118	42.869721	1.563998	700	229	307
	Esplourgues	ASAR122	42.868193	1.562145	705	234	45
	Bédeilhac	ASAR117	42.871762	1.570543	710	239	988
	Bouicheta	ABOU103	42.870497	1.569947	780	315	178
	Moutons	ABOU104	42.86925	1.57210	850	385	30
Clot la Carbonnière	Monique	ASAR113	42.828093	1.622763	650	130	110
	Midi	ASAR114	42.829686	1.625618	705	185	170
Quié de Lujat	Souloubrié	ASAR116	42.825914	1.678243	910	390	270
	Mirouge	ABOU102	42.80611	1.67556	1096	576	~10
Siech	Siech	ASAR112	42.882620	1.545914	676	70	1220
Roc de Trabinet	Herbo-Blanco	ASAR120	42.859829	1.525118	1115	425	65

1280 Table 1. Locations of visited cavities, their elevations relative to the Ariège River and its tributaries (Videssos and Saurat), and their current lengths.

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Sample	Mass (g)	[²⁷ Al] (ppm)	[²⁷ Al] × 10 ¹⁷ (at g ⁻¹)	Added spike (μL)	²⁶ Al/ ²⁷ Al (×10 ⁻¹⁴)
LO4	21.93	158.30±1.53	35.33±0.34	0	1.58±0.25
LO4b	23.96	13.98±0.09	3.12±0.02	251	2.61±0.32
LO4c	19.99	241.40±1.24	53.88±0.28	0	0.57±0.17

1282 Table 2. Aluminum measurements testing the effect of residual muscovite on observed Al contents. The Al spike has an Al concentration of 9,372 ± 47 ppm.

Sample ID	Sample (lab ID)	Cave	Facies and lithologies	Relative elevation (m)	^{10}Be (10^3 at g^{-1})	^{26}Al (10^3 at g^{-1})	$^{26}\text{Al}/^{10}\text{Be}$	Burial duration (Ma)	Paleodenudation rate (mm kyr^{-1})
ASAR117_001	BE1	Bédeilhac - Vidal gallery	Sand, coarse to very coarse, may be pebbly	239	81±16	29±11	0.4±0.2	6.31±0.99	7±1
ASAR117_002	BE2	Bédeilhac - Main Gallery	Sand, coarse to very coarse	239	35±10	21±9	0.6±0.3	5.26±1.42	27±8
ASAR117_003	BE3	Bédeilhac - Tombeau de Rolland	Amalgamated granite pebbles	239	41±7	8±4	0.2±0.1	7.61±1.24	7±1
ABOU118_001	CALA-14-01	<i>Enchantées Cave</i>	<i>Medium to coarse sandstones</i>	229	91±9	65±8	0.7±0.1	4.72±0.3	13±1
ASAR118_001b	CALA-14-01b	Enchantées Cave	Medium to coarse sandstones	229	66±11	22±4	0.3±0.1	6.33±0.49	8±1
ABOU118_002	CALA-14-02	<i>Enchantées Cave</i>	<i>Medium to coarse yellow sandstones</i>	229	61±4	82±20	1.3±0.3	3.47±0.55	211±13
ABOU118_003	CALA-14-03	<i>Enchantées Cave</i>	<i>Medium to coarse sandstones</i>	229	72±9	348±41	4.8±0.8	0.68±0.37	127±16
ABOU120_001	CALA-14-04	Esplourgues Cave	Medium to coarse sandstones	234	29±5	37±17	1.3±0.6	3.70±1.32	443±71
ABOU120_002	CALA-14-05	<i>Esplourgues Cave</i>	<i>Medium to coarse sandstones</i>	234	65±5	84±24	1.3±0.4	3.58±0.69	199±16
ABOU119_001	CALA-14-06	Bédeilhac Quarry	Sand, coarse to locally loamy	209	70±7	59±20	0.8±0.3	4.5±0.87	20±2
ASAR110_001	LO1	Lombrives-Lions Gallery	Sand, medium to very coarse	114	21±6	41±21	1.9±1.1	2.83±1.6	147±42
ASAR110_002	LO2	Lombrives (F1)	Amalgamated granite pebbles	114	29±3	3±3	0.1±0.1	8.81±1.67	5±1
ASAR110_003	LO3	Lombrives - Vierge Gallery (F1)	Sandstones, medium to coarse	114	36±2	30±10	0.8±0.3	4.52±0.83	38±2

Sample ID	Sample (lab ID)	Cave	Facies and lithologies	Relative elevation (m)	¹⁰ Be (10 ³ at g ⁻¹)	²⁶ Al (10 ³ at g ⁻¹)	²⁶ Al/ ¹⁰ Be	Burial duration (Ma)	Paleodenudation rate (mm kyr ⁻¹)
ASAR110_004	LO4	Lombrives - Vierge Gallery (F1)	Sandstones, medium to coarse	114	45±2	56±9	1.3±0.2	3.57±0.37	49±2
ASAR110_004b	LO4b	Lombrives - Vierge Gallery (F1)	Amalgamated quartzite pebbles	114	50±2	69±9	1.4±0.2	3.34±0.3	49±2
ASAR110_005	LO5	Lombrives - Salines (F1)	Amalgamated granite pebbles	114	22±2	14±8	0.6±0.4	5.18±1.48	44±5
ASAR110_006	LO6	Lombrives (F1)	Amalgamated granite pebbles	114	38±4	11±7	0.3±0.2	6.91±1.49	11±1
ASAR110_007	LO7	Lombrives (F3)	Sandstones, fine to medium	114	23±2	14±5	0.6±0.2	5.33±1.03	39±3
ASAR110_008	LO8	Lombrives - Mamelle Gallery	Sands, fine	114	98±11	16±9	0.2±0.1	7.9±1.3	3±1
ASAR110_009	Lo12-01	Lombrives (F1)	Amalgamated granite pebbles	114	8±1	B.D.L.	B.D.L.	-	-
ASAR110_010	Lo12-02	Lombrives - Vierge Gallery	Sandstones, medium to coarse	114	33±2	42±21	1.3±0.6	3.79±1.42	60±3
ASAR111_001	LS1	Lombrives - Tunnel	Sandstones, medium	119	13±1	38±9	2.9±0.8	1.82±0.63	400±43
ABOU102_001	LUI1	Mirouge cave	Medium to very coarse rubified sandstones	576	9±1	10±6	1.2±0.7	3.98±1.68	207±24
ABOU102_002	LUI2	Mirouge cave	Medium to very coarse rubified sandstones	576	114±18	B.D.L.	B.D.L.	-	-
ABOU102_003	LUI3	Mirouge cave	Medium to very coarse rubified sandstones	576	19±2	B.D.L.	B.D.L.	-	-
ABOU102_003b	LUI3b	Mirouge cave	Medium to very coarse rubified sandstones	576	8±1	B.D.L.	B.D.L.	-	-

Sample ID	Sample (lab ID)	Cave	Facies and lithologies	Relative elevation (m)	¹⁰ Be (10 ³ at g ⁻¹)	²⁶ Al (10 ³ at g ⁻¹)	²⁶ Al/ ¹⁰ Be	Burial duration (Ma)	Paleodenudation rate (mm kyr ⁻¹)
ABOU104_001	MOU1	Moutons Cave	Cemented medium to very coarse sands (yellow-ochre)	385	16±2	6±6	0.4±0.4	6.19±1.85	-
ASAR104_001b	MOU1b	Moutons Cave	Cemented medium to very coarse sands (yellow-ochre)	385	51±7	B.D.L.	B.D.L.	-	-
ASAR104_001c	MOU1c	Moutons Cave	Cemented medium to very coarse sands (yellow-ochre)	385	30±6	B.D.L.	B.D.L.	-	-
ABOU101_005	Ni-13-05	Niaux – Marbre Gallery (F2)	Amalgamated pebbles	142	55±3	26±9	0.5±0.2	5.74±0.91	797±101
ABOU101_008	Ni-13-08	Niaux – Bison Rouge Gallery	Amalgamated medium to coarse sands & pebbles	142	27±3	16±12	0.6±0.5	5.32±1.79	13±1
ABOU101_010	Ni-13-10	Niaux – Bison Rouge Gallery	Sandstones, medium to coarse	142	73±7	27±16	0.4±0.2	6.3±1.34	480±62
ABOU101_012	Ni-13-12	Niaux – Éboulis Gallery	Amalgamated quartzite & granite pebbles	142	8±1	17±8	2.1±1.1	2.77±1.59	176±17
ABOU101_013	Ni-13-13	Niaux – Éboulis Gallery	Sands, medium to very coarse, may be pebbly	142	21±2	97±27	4.7±1.4	0.8±0.76	409±50
ABOU101_015	Ni-13-15	Niaux– Gallery of the counts of Foix (F3)	Amalgamated medium to coarse sands & gneiss pebbles	142	25±4	32±16	1.3±0.7	3.69±1.54	417±46
ABOU101_017	Ni-13-17	Niaux– Gallery of the counts of Foix (F1)	Amalgamated medium sands & pebbles (quartzites, granitoids)	142	B.D.L	24±24	-	-	-
ABOU101_020	Ni-13-20	Niaux– Gallery of the counts of Foix (F3)	Amalgamated quartzite & granitoid pebbles	142	29±4	104±28	3.6±1.1	1.37±0.7	522±87

Sample ID	Sample (lab ID)	Cave	Facies and lithologies	Relative elevation (m)	^{10}Be (10^3 at g^{-1})	^{26}Al (10^3 at g^{-1})	$^{26}\text{Al}/^{10}\text{Be}$	Burial duration (Ma)	Paleodenudation rate (mm kyr^{-1})
ABOU101_023	Ni-13-23	Niaux– Gallery of the counts of Foix (F3)	Medium to very coarse sandstones	142	28±3	53±19	1.9±0.7	2.84±0.98	447±55
ASAR101_001	Ni12-02	Niaux– Carrefour Cartailhac	Sand, medium to very coarse	142	19±3	10±6	0.5±0.3	5.69±1.59	456±40
<i>ASAR103_002</i>	<i>SED-13-02</i>	<i>Bouicheta Cave</i>	<i>Cemented medium to very coarse sands (yellow-ochre)</i>	315	30±5	33±17	1.1±0.6	4.03±1.38	39±7
ASAR116_001	SO1	Souloubrié	Amalgamated smoky quartzite pebbles	460	16±7	B.D.L.	B.D.L.	-	427±78

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1284 Table 3. ^{10}Be and ^{26}Al concentrations, burial ages, and paleodenudation rates obtained for sampled caves. B.D.L., below detection limit. Burial durations were
1285 computed by solving Equation (3) with a Monte Carlo approach (Blard et al., 2019), assuming a mean watershed palaeoelevation of 1,990 m a.s.l. for the
1286 buried material (see Section 4.2.1). Derived maximum palaeodenudation rates were calculated from ^{10}Be concentrations (corrected for radioactive decay)
1287 using burial durations computed from the $^{26}\text{Al}/^{10}\text{Be}$ ratios and the production rates described in Section 4.2.1. Samples reported in italics contained residual
1288 muscovite, and the ^{26}Al concentrations reported for those samples may be inaccurate. F1, “Lombrives alluvial formation”; F3, “conglomerates of the gallery of
1289 the counts of Foix” (see Section 2.1). Additional data are available on supp. info. or on request.

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Sample	Cave	Mass (mg)	T (°C) 20 min	^{20}Ne (10^{10} at g^{-1})	$^{21}\text{Ne}/^{20}\text{Ne}$ (10^{-3})	$^{22}\text{Ne}/^{20}\text{Ne}$ (10^{-1})	Cosmogenic $^{21}\text{Ne}^a$ (10^7 at g^{-1})	Cosmogenic $^{21}\text{Ne}^b$ (10^7 at g^{-1})
LO12-02	Lombrives	287.2	1446	1.89±0.07	3.58±0.06	1.056±0.011	1.28±0.27	0.86±0.37
LUJ1	Mirouge	304.9	1446	1.38±0.05	4.49±0.09	1.004±0.013	2.20±0.26	1.78±0.36
SO1	Souloubrié	228.4	500	0.99±0.037	3.19±0.07	1.013±0.007	0.29±0.14	-
			1300	0.46±0.017	5.27±0.15	1.034±0.015	1.09±0.11	-

Sample	Cave	Mass (mg)	T (°C) 20 min	²⁰ Ne (10 ¹⁰ at g ⁻¹)	²¹ Ne/ ²⁰ Ne (10 ⁻³)	²² Ne/ ²⁰ Ne (10 ⁻¹)	Cosmogenic ²¹ Ne ^a (10 ⁷ at g ⁻¹)	Cosmogenic ²¹ Ne ^b (10 ⁷ at g ⁻¹)
						Total	1.38±0.18	0.96±0.31
MOU1	Moutons	272.7	1400	1.36±0.05	3.78 0.05	1.033±0.01	1.20±0.19	0.78±0.31
CALA-14-01b	Enchantées	250.8	1400	1.32±0.045	3.83 0.06	1.06±0.011	1.22±0.19	0.80±0.31
BE1	Bédeilhac	201.7	1400	1.18±0.04	4.02 0.09	1.013±0.012	1.32±0.19	0.90±0.32

1291 Table 4. Neon isotopic data from the sampled caves. All samples were analyzed with the Helix SFT, except SO1, which was measured with the Helix MC.

1292 ^a Cosmogenic ²¹Ne excess calculated using Equation (5) following Honda et al. (2015).

1293 ^b Cosmogenic ²¹Ne corrected for a maximum potential nucleogenic ²¹Ne build-up of $(4.2 \pm 2.5) \times 10^6$ at g⁻¹ (see Section 5.2).

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Sample	Cave	Altitude (m)	¹⁰ Be (10 ³ at g ⁻¹)	²¹ Ne (10 ⁷ at g ⁻¹)	²¹ Ne/ ¹⁰ Be	Burial duration (Ma)	Corrected burial duration [#] (Ma)	Paleodenudation rate (mm kyr ⁻¹)
LO12-02	Lombrives	114	33±2	1.28±0.27	393±84	9.0±0.4	7.8±0.8	4.5±0.2
LUJ1	Mirouge	576	9±1	2.20±0.26	2563±423	12.6±0.3	10.4±0.4	2.8±0.3
SO1	Souloubrié	460	16±7	1.38±0.18	870±376	10.6±0.9	9.1±1.0	4.2±1.7
MOU1	Moutons	385	16±2	1.20±0.19	743±148	10.3±0.4	9.0±0.8	4.8±0.6
CALA-14-01b	Enchantées	229	91±9	1.22±0.19	134±25	6.8±0.3	5.5±0.8	4.7±0.5
BE1	Bédeilhac	239	81±16	1.32±0.19	163±40	7.2±0.5	5.9±0.8	4.4±0.8

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Table 5. ¹⁰Be/²¹Ne burial durations and palaeodenudation rates calculated using Equation (4) and a watershed palaeoelevation of 1,990 m a.s.l.

Palaeodenudation rates were computed from the ¹⁰Be concentrations corrected for radioactive decay since burial.

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[#] Burial durations computed after correcting the cosmogenic ²¹Ne concentration for a maximum nucleogenic contribution of $(4.2 \pm 2.5) \times 10^6$ at g⁻¹.