

1 **Snowmelt as a determinant factor in the hydrogeological behavior of**
2 **high mountain karst aquifers: The Garcés karst system, Central**
3 **Pyrenees (Spain).**

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17

18 **Abstract**

19 Time series of environmental tracers (groundwater stable isotope composition, electrical
20 conductivity and temperature) and concentration breakthrough curves of artificial
21 tracers (uranine, eosine, amino-G and naphtionate) were analyzed to characterize fast
22 preferential and slow matrix in-transit recharge flows in the Paleocene-Eocene
23 limestone aquifer of the Ordesa and Monte Perdido National Park. This is an alpine
24 karst system drained by a water-table cave, a rare hydrological feature in high mountain

25 karst systems with similar characteristics. Snowmelt favors the areal recharge of the
26 system. This process is reflected in the large proportion of groundwater flowing through
27 the connected porosity structure of the karst aquifer, which amounts the 75% of the total
28 system water discharge. From the perspective of water resources recovery, the water
29 capacity of the fissured-porous zone (matrix) represents 99% of the total karst system
30 storage. The volume associated to the karst conduits is very small. The estimated mean
31 transit times are 9 days for conduits and 475 days for connected porosity. The short
32 transit times, even the longer ones, mean high vulnerability of the karst system to
33 contaminants and great impact of climate change.

34

35 Key words: Water isotopes; Dye tracers; Transit time; Recharge; Alpine karst
36 hydrology; Parque Nacional de Ordesa y Monte Perdido.

37

38 **1. Introduction**

39 High mountain zones are known as "water towers" because they generate the main
40 water resources feeding the lowland depending ecosystems (Viviroli et al., 2007). When
41 these mountains constitute or host an aquifer, then the collected water resources remain
42 in the basin for a longer time, because aquifers regulate the basin discharge. Thus,
43 mountain aquifers provide a strategic water resource in dry seasons, especially in semi-
44 arid regions. From the perspective of water resources availability, carbonated aquifers
45 are those arousing the most interest as they represent 15.2% of the land surface and
46 provide drinking water to about 10% of the world's population (Stevanovic, 2019).
47 Globally, 31.1% of all the surface exposures of carbonate rocks occur in plains, whereas
48 68.9% do in hills and mountain zones (Goldscheider et al., 2020).

49 A good characterization of the karst system behavior is essential to manage correctly
50 aquifer discharge. The hydrodynamical response of karst aquifers greatly depends both
51 on the type of recharge and on their underground geological structure (Audra and
52 Palmer, 2011; 2015). On the one hand, when the aquifer recharge is uneven and
53 occurring quickly with high flow rates, the hydrodynamical perturbation propagates
54 through the system and generates flash discharges. As a result, the epiphreatic zone
55 experiences often back flooding events with dry periods in between, thus developing
56 drains with looping profiles. An example is the Hollock cave (Bögli, 1980), one of the
57 most extensive limestone caves currently known, which is developed inside the alpine
58 Austro-German Hochifen-Gottesacker karst system (Goldscheider, 2005; Göppert and
59 Goldscheider 2008; Chen and Goldscheider 2014). On the other hand, when recharge
60 enters the system in a diffuse and regular way as a consequence of a poorly permeable
61 surface cover over the epikarst, then the aquifer water table remains fairly stable with
62 time, and the principal drain of the system, also known as water table cave, develops at
63 the water table level, as it happens in the Cobre cave system in the Cantabrian
64 Mountains (Rossi et al., 1997).

65 In high mountain zones, where the snow covers the ground surface during long periods
66 or even the whole year, the thaw may produce a diffuse recharge process in the
67 underlying karst systems (Meeks and Hunkeler, 2015). This process, that would emulate
68 the role played by the aforementioned permeable soil cover, may be also very important
69 in glacierized catchments, although only a few researchers have studied the relations
70 between alpine glaciers and the underlying karst aquifers (Smart, 1996; Gremaud et al.
71 2009; Zheng et al. 2015). Diffuse recharge may favor small changes in elevation of the
72 water table, which is an essential condition for the development of horizontal cave
73 passages (Gabrovsek and Dreybrodt, 2001; Kaufmann 2002). Nevertheless, the role

74 played by snow accretion and melting dynamics in high mountain karst aquifers in
75 relation to the development of cave patterns is little known. In fact, many of the studies
76 in this regard have been carried out in caves whose recharge areas are below 2000 m
77 a.s.l. (Häuselmann, 2019), up to an elevation in which rainfall infiltration dynamics
78 dominates recharge. As a result, most of the described caves present a looping cave
79 pattern. The very few geomorphological descriptions of water-table caves developed in
80 high mountain zones reflect the difficulties inherent to the exploration activities
81 imposed by the harsh working conditions in high elevation alpine zones.

82 This work is devoted to characterize the hydrogeological behavior of the alpine Garcés
83 water table cave and the associated bare karst system that are developed at elevations
84 between 2000 and 3300 m a.s.l.. The characterization is conducted through a
85 multidisciplinary approach that includes physicochemical and hydrological monitoring
86 of the karst system discharge, and the use of environmental and fluorescent dye tracers
87 testing techniques. The Garcés water table cave drains the highest aquifer karst in
88 Western Europe, the alpine Paleocene-Eocene limestones of the Ordesa and Monte
89 Perdido National Park, in the Central Southern sector of the Pyrenees. The terminal
90 point of the Garcés cave is the Garcés spring, whose discharge generates the known
91 Cola de Caballo (*Water Horsetail*) waterfall and highlights the significant natural
92 heritage value of the study zone (Ortega-Becerril et al., 2019).

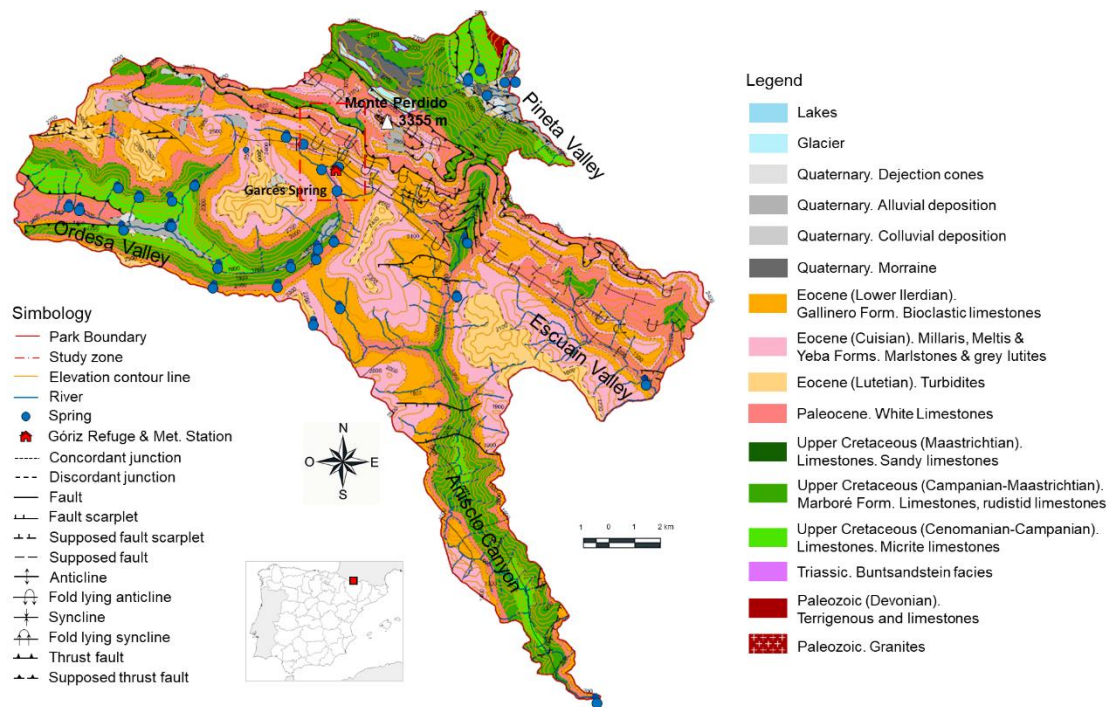
93

94 **2. The study area**

95 **2.1 Geographical and climatic settings**

96 The study area is located in the Ordesa and Monte Perdido National Park (PNOMP
97 from the initials in Spanish), in the Central-Southern Pyrenees (Fig. 1). The PNOMP

98 constitutes the highest karst system in Western Europe. It contains several peaks above
 99 3000 m a.s.l. The Monte Perdido (3348 m a.s.l.) is its highest point.



100

101 Fig. 1. Geological map of the the Ordesa and Monte Perdido National Park (PNOMP).

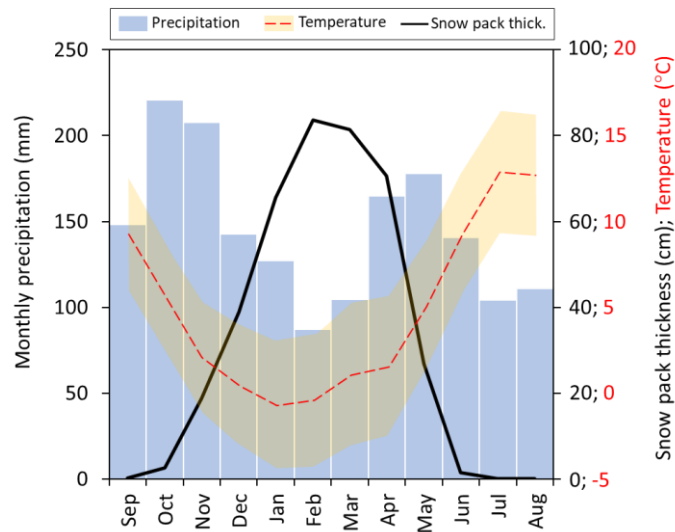
102 The study zone is indicated by the red dashed line. Modified from Lambán et al. (2015).

103

104 The PNOMP has a cold climate with mild and cool summers (AEMET/IM, 2011). The
 105 main precipitation (P) volumes registered in the PNOMP are generated by oceanic low
 106 pressure fronts arriving from the Atlantic Ocean (Lambán et al., 2015). The study area
 107 is covered by snow between October and June. The mean annual precipitation at the
 108 Góriz Meteorological station (Fig. 1, at 2200 m a.s.l.) is 1650 mm (Fig. 2). Precipitation
 109 (P) presents two peaks in autumn and spring, at 221 and 178 mm/month, respectively,
 110 and two minima in winter and summer, at 87 and 104 mm/month, respectively. At the
 111 same meteorological station the air temperature (T_{2m}) shows a seasonal evolution with
 112 maximum and minimum monthly averaged values of 13°C (July) and -0.6°C

113 (December), respectively (Fig. 2). The mean annual temperature is 4.9 °C. Both P and
 114 T_{2m} present significant altitudinal variations in the area, with mean vertical gradients of
 115 P and T_{2m} of 200 mm/km and -3.3 °C/km, respectively (Lambán et al., 2015).

116 The mean isotopic content of precipitation (δ_P) in the PNOMP shows a seasonal
 117 variation, as does the T_{2m} . The mean annual value of $\delta^{18}O$ (δ_P) in the meteorological
 118 station of Góriz is -11.25‰, and the mean amplitude (A_δ) is 4.97‰. Besides, δ_P and A_δ
 119 vary with elevation, with altitudinal slopes of -2.2‰/km and 0.9‰/km, respectively
 120 (Jódar et al., 2016b).



121

122 Fig. 2. Seasonal variation of average monthly precipitation (columns), soil accumulated
 123 snow pack thickness (thick line), and monthly average air temperature (dashed line)
 124 with the corresponding maximum and minimum values marked as a variation interval
 125 (shaded area) at the meteorological station of Góriz (2220 m a.s.l.), for the period 1981-
 126 2019

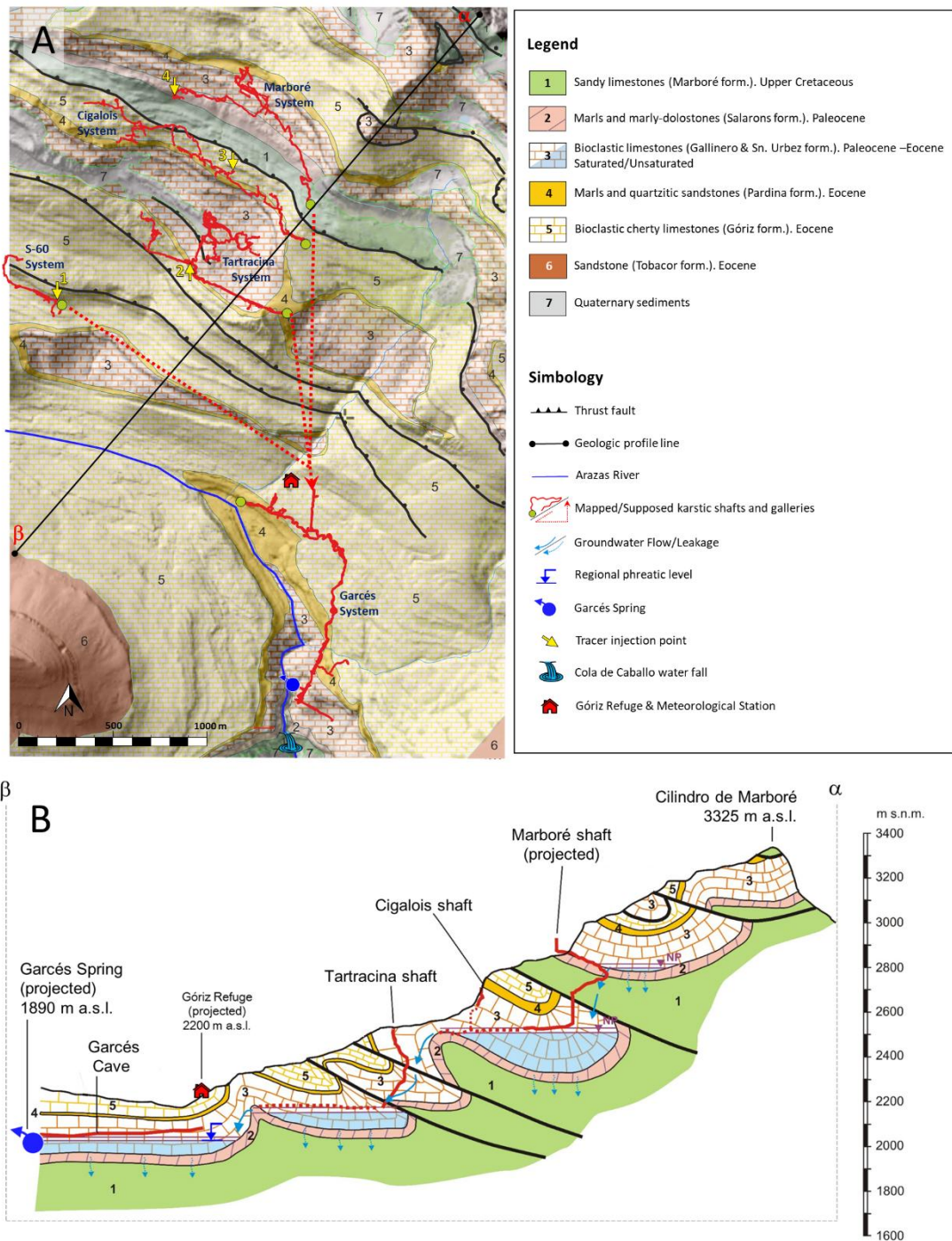
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128 **2.2 Geological and hydrogeological settings**

129 The PNOMP is located in the central-western sector of the southern Pyrenees, forming
130 part of the Sierras Interiores (*Inner Ranges*) structural unit. Its geological structure is
131 characterized by imbricated thrust sheets and associated folds of carbonate materials,
132 with a predominantly southern vergence (Séguret, 1972). The age of the different
133 materials composing the massif ranges from Upper Cretaceous (Marboré Formation) to
134 the Lower Paleocene-Eocene (Gallinero Formation) (Lambán et. al., 2015; Robador et.
135 al., 2018). The layout of the different materials by stacking several thrust sheets
136 generates a vertical development close to 1000 m from the Marboré peak to the Garcés
137 Spring (Fig 3).

138 The limestones associated to the Gallinero Formation are karstified and constitute the
139 main aquifer drained by the Garcés Spring. The calcareous Paleocene-Eocene materials
140 and their vertical disposition favor both the karstification process and the existence a
141 very thick unsaturated zone due to an imbricated group of overthrust sheets and
142 associated folds. The base of the karst aquifer is the low permeability materials of the
143 Marboré Formation (calcareous sandstone with intercalations of both quartz and lithic
144 arenites).

145 The karst system is subject of active exploration so far with the aim of proving the
146 hydrological connection between the different mapped galleries (Fig.3A). The karst
147 exploration has revealed that the network in the vadose zone is of the branchwork type
148 (Jouves et al., 2017), which is the most frequent development pattern in mountainous
149 karst massifs. In the epiphreatic zone, the karst development pattern type is of the water
150 table cave type. This pattern is associated with diffuse recharge processes, which often
151 involves a well-developed epikarst zone or a semi-confining layer that covers the deeper
152 most permeable materials. Nevertheless, a well-developed epikarst or a surficial semi-
153 confining layer do not exist in the study area



155

156 Fig. 3. Hydrogeological conditions. (A) Hydrogeological map of the study zone. The
 157 main karst cave systems of the area are marked in red. Most of them have not been
 158 completely explored. The green circle indicates the terminal sump of the corresponding
 159 karst systems. The dotted arrows indicate the unmapped hydrologic connection between

160 these karst systems with the Garcés karst system. The codes of the tracer injection
161 points correspond to those of Table 1. The map area corresponds to that of the “study
162 zone” marked in Fig. 1. (B) α - β cross-section, as defined in the hydrogeological map.

163

164 **3. Methods and materials**

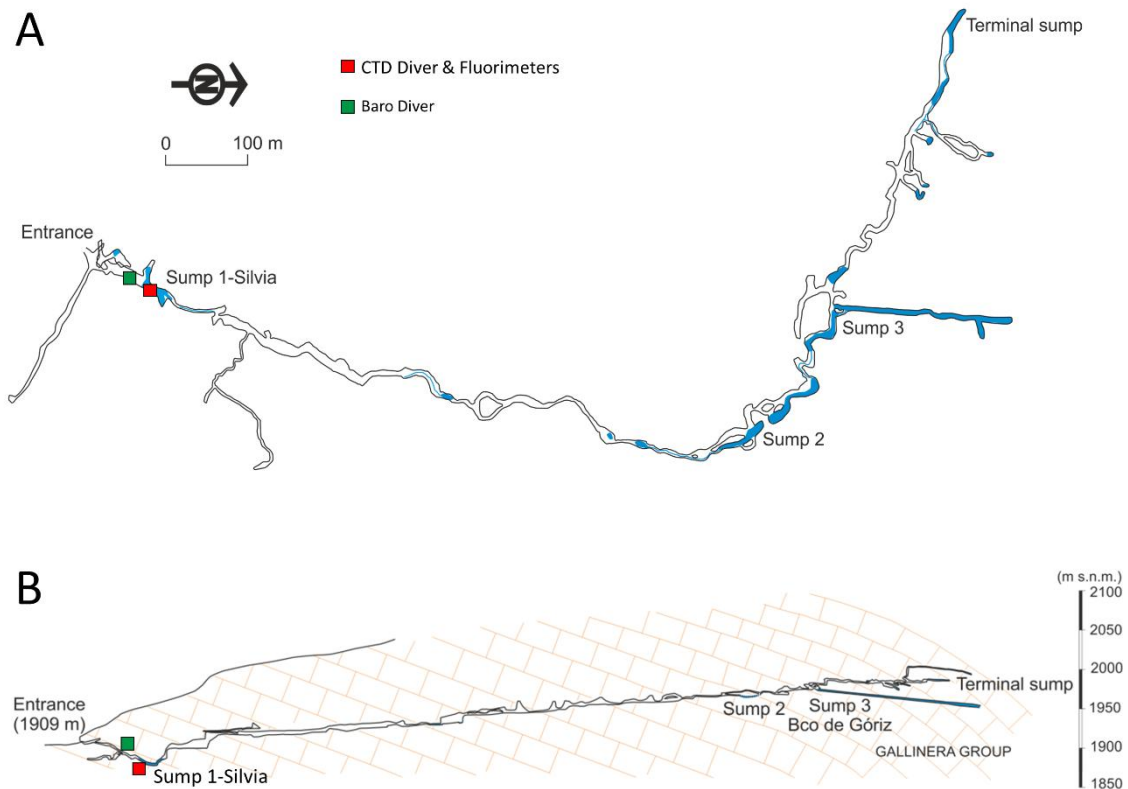
165 **3.1. Field work**

166 **3.1.1 Instrumentation of the Garcés Cave**

167 The Garcés Cave corresponds to the lowest zone of the Garcés System (Fig. 4). The
168 entrance is to an old upwelling point that has been left hang. Currently, the water that
169 overflows through Sump-1 (locally known as Silviá’s sump) discharges in the Arazas
170 river bed during the low-flows periods and through a set of trop-pleins located at
171 different levels on the wall of the left bank of the Arazas river during the high-flows
172 events triggered by both long rainy periods and high intensity rainfall events.

173 Two Diver® (Van Essen Instruments, 2016) devices were installed inside the Garcés
174 Cave: a Baro-Diver just after the main cave entrance to register the air pressure and
175 temperature variations in the cave and a CTD-Diver in Sump-1 to measure the
176 variations of groundwater column and the corresponding groundwater temperature
177 (T_{GW}) and electrical conductivity (EC_{GW}). Besides, two fluorimeters GGUN-FL24 were
178 installed in Sump-1 to be able to measure 4 different fluorescent dye tracers at the same
179 time. All the installed devices were programmed for measuring hourly their
180 corresponding variables. The spring discharge is obtained as a function of the measured
181 groundwater column through an empirical relationship.

182



183

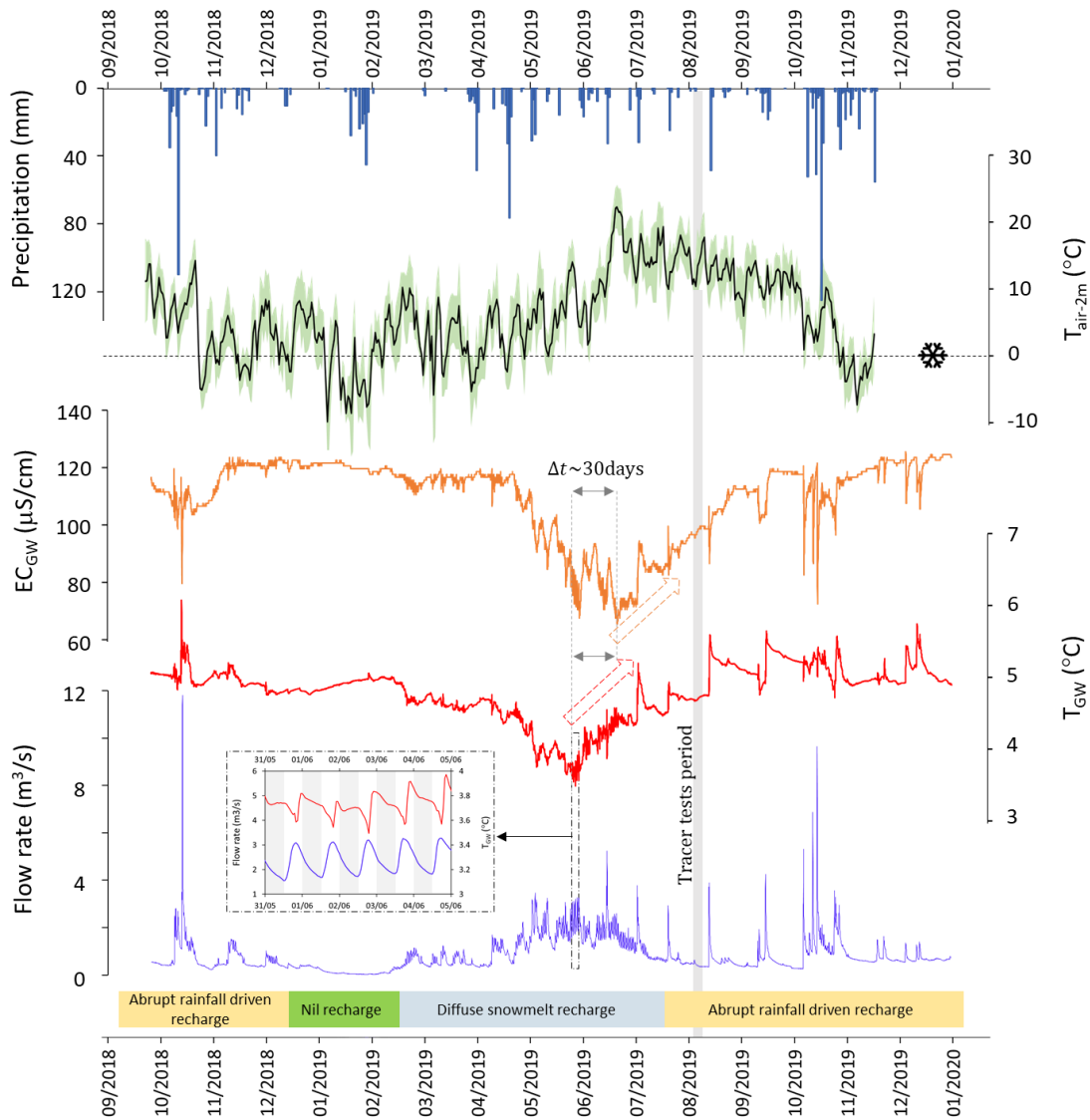
184 Fig. 4. Plan view (A) and cross section (B) of the Garcés karst system. The location of
 185 field instruments is also indicated.

186

187 3.1.2 Groundwater sampling

188 Since Jun 2018, periodical monthly GW sampling has been carried out in Sump-1 (Fig.
 189 4). In all cases, the EC_{GW} , pH and T_{GW} were measured on site (Fig. 5). The isotope
 190 content in the water samples were determined at the Stable Isotopes Laboratory of the
 191 UAM (Universidad Autónoma de Madrid). The $\delta^{18}O$ and δ^2H contents were analysed by
 192 pyrolysis in a Thermo Scientific FlashEA™ 1112 HT Elemental Analyzer coupled to a
 193 DELTA V™ IRMS System. The measurement errors for $\delta^{18}O$ and δ^2H are ± 0.1 and
 194 $\pm 1\%$, respectively. All results are given relative to the V-SMOW standard.

195



196

197 Fig. 5. From top to bottom: daily precipitation (mm); air temperature at 2 m ($T_{\text{air-2m}}$; °C)
 198 at the Góriz meteorological AEMET station, with the greenish lines indicating the
 199 maximum and minimum temperature interval (the horizontal dashed line with the
 200 snowflake symbol indicates the freezing temperature of 0 °C); electrical conductivity
 201 (EC_{GW} $\mu\text{S}/\text{cm}$) and temperature (T_{GW} °C) of groundwater in Sump-1; and Garcés Spring
 202 discharge (m^3/s) throughout the monitoring period. The inset shows the effect of the
 203 snowmelt and refreezing daily cycles on both the spring discharge and groundwater
 204 temperature for the selected time slice. The vertical grey shaded area indicates the time
 205 period in which the dye tracer tests were conducted.

206

207 3.1.3 Tracer tests

208 In August 2019, the members of the Otxola Espelogroup injected four fluorescent dye
 209 tracers simultaneously at different points of the Paleocene-Eocene karst system.
 210 Uranine, eosine, amino-G and naphthionate were injected into the Marboré, Cigalois,
 211 Tartracina and S60 karst systems (Fig. 3A), respectively. The distances between the
 212 tracer injection and recovery points vary between 2.8 and 4 km, and the elevation gap is
 213 between 235 and 790 m (Table 1). The time in which the dye tracer tests were
 214 conducted coincides with the low flow period of the aquifer system (Fig. 5).

215 Table 1: Summary of the dye tracer tests characteristics

Tracer test code	Injection point	Injected tracer	Injection point elevation ^(a) / depth ^(b) (m a.s.l.) / (m b.g.l.)	$\Delta Z^{(c)}$ (m)	Tracer mass injected (kg)	$C_{\max}^{(d)}$ (ppb)	Tracer test distance ^(e) (km)
1	S-60	Naphtionate	2222 / 20	302	1.0	12.07	3.2
2	Tartracina	Amino-G	2155 / 415	235	1.5	12.88	2.8
3	Cigalois	Eosine	2710 / 0	790	0.5	1.13	3.2
4	Marboré	Uranine	2520 / 120	600	0.5	0.97	4.0

(a) Meters above sea level (b) Meters below ground level; (c) Elevation gap between the tracer injection and recovery points; (d) Maximum concentration of the breakthrough curve; (e) Distances along the transects, shown in Fig. 3

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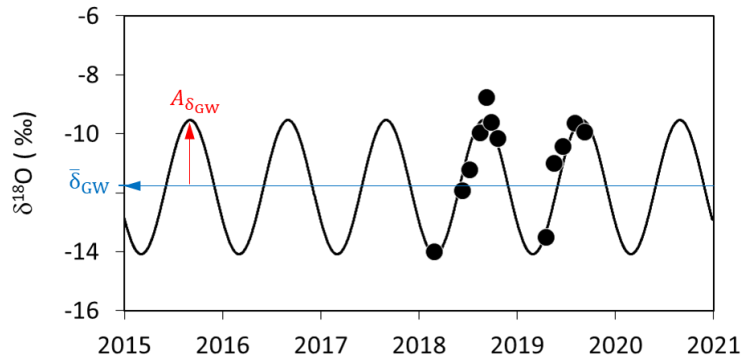
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218 3.2 Estimation of groundwater transit time and recharge elevation

219 3.2.1 Seasonal varying isotopic content input function

220 The isotopic content of GW (groundwater) in Sump-1 shows a seasonal dependence that
 221 can be described by a sinusoidal function $\delta_{GW}(t)$, with $\bar{\delta}_{GW}$ and $A_{\delta_{GW}}$ the
 222 corresponding mean value $\bar{\delta}$ and amplitude A_{δ} , respectively (Fig. 6).

223



224

225 Fig. 6. Observed (circles) and EPM fitted (line) $\delta^{18}\text{O}$ content in groundwater in Sump-1.

226 $\bar{\delta}_{\text{GW}}$ and $A_{\delta_{\text{GW}}}$ are the sinusoidal mean value and amplitude, respectively.

227

228 The seasonal behaviour observed in $\delta_{\text{GW}}(t)$ is the consequence of both a small aquifer
 229 transit time and the seasonal dependence of the isotopic content of precipitation $\delta_{\text{P}}(t)$.

230 The seasonal variation in $\delta_{\text{P}}(t)$ shows lighter and heavier isotopic compositions in
 231 summer and winter, respectively, which agrees with the well-established relationship

232 between isotopic fractionation and temperature (Clark and Fritz, 1997). As it happens to
 233 GW, $\delta_{\text{P}}(t)$ can be also described by a sinusoidal function with the corresponding $\bar{\delta}_{\text{P}}$

234 and $A_{\delta_{\text{P}}}$. Aside, $\bar{\delta}_{\text{P}}$ and $A_{\delta_{\text{P}}}$ depend on elevation. This can be approached by a linear
 235 relationship between these two variables and elevation. $\bar{\delta}_{\text{P}}(Z)$ and $A_{\delta_{\text{P}}}(Z)$ are known as

236 the “Isotopic Altitudinal Line” (IAL) and the “Amplitude Altitudinal Line” (AAL),
 237 respectively, and have to be determined empirically on site (Poage and Chamberlain,

238 2001; Jódar et al., 2016a,b; Herms et al., 2019).

239 The seasonal variation of the isotopic contents, $\delta_{\text{P}}(t)$ and $\delta_{\text{GW}}(t)$, allows estimating a
 240 mean response time of the aquifer (τ) by solving the convolution integral (Eq. 1) in the

241 framework of lumped flow models (Małoszewski and Zuber, 1982). Eq. 1 describes the

242 aquifer discharge $\delta_{\text{GW}}(t)$ in terms of both $\delta_{\text{P}}(t)$ and a system response function $g(t)$.

$$\delta_{GW}(t) = \int_0^{\infty} \delta_P(t-t')g(t') dt' \quad (1)$$

243 This solution is for invariable geometry. In the case of hydraulic changes the system is
 244 represented by the drainage/refilling porosity (often called effective porosity), which
 245 may be different, depending on capillary hysteresis in the unsaturated zone, and often
 246 much less than total porosity. The transfer of a conservative tracer depends on total
 247 porosity or on total matrix and fracture porosity in the case of dual porosity media. In
 248 this transfer case, the response time is the average transit /renovation time, which results
 249 from a transit time distribution in the flow and transport system.

250 In some cases, $g(t)$ can be expressed as an algebraic expression of a few parameters
 251 that include τ , the first moment of the transit time distribution (Małoszewski and Zuber,
 252 1982; Amin and Campana, 1996). Jódar et al. (2016b) demonstrated that the
 253 Exponential-Piston model (EPM) was a good enough transit time distribution to
 254 describe the behaviour of the PNOMP aquifer. This model is actually the sequential
 255 combination of two different lumped models: (1) The Piston Flow Model (PFM) and (2)
 256 the Exponential Model (EM). The first model represents an aquifer system through
 257 which the tracer migrates with a constant velocity in the profile (i.e. piston flow),
 258 whereas the latter model considers an aquifer system with an exponential distribution of
 259 transit times. As a result, EPM describes an aquifer system consisting of two parts in
 260 line, regardless of their particular sequence, one with volume V_{PFM} and piston flow
 261 transit time distribution, and another part with volume V_{EM} with exponential
 262 distribution of transit times. The analytical expression describing the EPM system
 263 response function is provided by Eq. 2

$$g(t) = \begin{cases} 0 & t < \tau \left(1 - \frac{1}{\eta}\right) \equiv t_\tau \\ \frac{1}{\tau} \eta e^{-\frac{\eta}{\tau} t + \eta - 1} & t \geq t_\tau \end{cases} \quad (2)$$

264

265 where η is the fraction of the total volume with the exponential distribution of transit
 266 times. In other words, $\eta = (V_{EM} + V_{PFM}) / V_{EM}$. For $\eta = 1$, EPM equals to EM. For $\eta \rightarrow \infty$,
 267 EPM reduces to PFM.

268 In the case of EPM, there is an analytical solution to the convolution integral, given by
 269 (Jódar et al., 2014)

$$\delta_{GW}(t) = \frac{A_{\delta_P}(Z_R) \cdot \kappa^2}{\kappa^2 + \omega^2} \cdot \left(\sin(\omega(t - (t_0 + t_\tau)) + \varphi) - \frac{\omega}{\kappa} \cos(\omega(t - (t_0 + t_\tau)) + \varphi) \right) + \bar{\delta}_{GW} \quad (3)$$

270

271 where $\kappa = \eta / \tau$ and $A_{\delta_P}(Z_R)$ is the amplitude of the seasonal variation of the isotopic
 272 content of precipitation evaluated at the recharge zone elevation Z_R .

273 To estimate $A_{\delta_P}(Z_R)$, the method proposed by Jódar et al. (2016b) is applied. It can be
 274 summarized in a three steps approach:

- 275 1. Estimation of $\bar{\delta}_{GW}$. This is done by fitting a sinusoidal function to the observed
 276 values of the isotopic content in groundwater at the sampling point (Fig. 6).
- 277 2. Estimation of Z_R . This is done by evaluating the IAL (isotopic altitudinal line)
 278 function and comparing with the previously obtained $\bar{\delta}_{GW}$ value.
- 279 3. Estimation of $A_{\delta_P}(Z_R)$ by evaluating the AAL (amplitude altitudinal line)
 280 function for Z_R (Fig. 6). Both the IAL (Eq.4) and AAL (Eq.5) functions have
 281 been characterized by Jódar et al (2016b) for the PNOMP.

$$Z_R = -458.28 \cdot \overline{\delta^{18}\text{O}_{\text{GW}}} - 2981.60 \quad (4)$$

$$A_{\delta_P} = 0.0009 \cdot Z_R + 3.08 \quad (5)$$

282

283 3.2.2 Pulse tracer input function

284 All the tracer tests conducted in the Paleocene-Eocene karst system of the PNOMP
 285 consisted of a direct pulse tracer injection in an active phreatic tube. This type of
 286 instantaneous tracer injection, with initial concentration C_0 , is mathematically
 287 represented by a Dirac delta δ function (Eq. 6). The convolution integral (Eq. 1, where
 288 $\delta_{\text{GW}}(t)$ and $\delta_P(t)$ are replaced, without loss of validity, by $C_{\text{out}}(t)$ and $C_{\text{in}}(t)$, respectively)
 289 is solved to simulate the tracer breakthrough at the outlet of the system. In this case, the
 290 dispersion flow model (DM) (Małoszewski and Zuber, 1982) is used as the system
 291 response function, whose analytical expression is given by Eq. 7,

$$C_{\text{in}}(t) = C_0 \delta(t) \quad \forall t \quad (6)$$

$$g(t) = \Gamma \cdot t^{-3/2} e^{-\left(\frac{1}{4P_D} \left(\frac{\tau+t}{\tau}\right)\right)} \quad t \geq 0 \quad (7)$$

292

293 where Γ is defined by mathematical convenience as:

$$\Gamma = \sqrt{\frac{\tau}{4\pi P_D}} e^{\left(\frac{1}{2P_D}\right)} \quad (8)$$

294

295 P_D is the inverse of the Péclet number (Pe). Pe is a dimensionless number defined as the
 296 ratio between the advective and the diffusion/dispersion characteristic times. If $\text{Pe} > 1$,
 297 then the advective flux is the most important transport mechanism, and conversely.

298 As in the case of EPM, the analytical solution to the convolution integral for DM does
 299 exist (Jódar et al., 2014) and is given by

$$C_{out}(t) = C_0 \cdot \Gamma \cdot t^{-3/2} e^{-\left(\frac{1}{4P_D} \left(\frac{\tau+t}{\tau}\right)\right)} \quad t \geq 0 \quad (9)$$

300

301 **3.3 Time series analysis**

302 The statistical methods used in this work are the autocorrelation and the cross-
 303 correlation functions. Both methods are presented and commented briefly, followed by
 304 the hydrogeological interpretation.

305 The autocorrelation function $r_X(k)$ provides a normalized measure of the temporal
 306 dependence of the successive terms. In this work, there is a daily basis for the time
 307 series $X(t)$ (Lambrakis et al. 2000). It is defined as

$$r_X(k) = \frac{C_X(k)}{C_X(0)} \quad (10)$$

$$C_X(k) = \frac{1}{N} \sum_{j=1}^{N-k} (X_j - \bar{X})(X_{j+k} - \bar{X}) \quad (11)$$

308

309 where N , X_j and \bar{X} are respectively the length, the j^{th} term and the average value of the
 310 time series $X(t)$, and k is the time lag.

311 The correlogram $C_X(k)$ allows estimating the memory capacity of the system (Mangin,
 312 1984; Pulido-Bosch et al., 1995). The memory capacity is defined as the time lag
 313 required by the system to forget the initial time series values. From a pragmatic
 314 perspective, this happens when $r_X(k)$ is below a certain correlation threshold value
 315 comprised between 0.1 and 0.2 (Benavente et al., 1985). The slope of the correlogram
 316 function provides information of both, the karstification degree of the hydrogeological

317 system and the groundwater reserves in the aquifer (Mangin, 1984; Padilla and Pulido-
318 Bosch, 1995). This can be analysed from two extreme situations:

- 319 • Steep correlogram slope with short decorrelation time lag, which is associated to
320 well-developed karst network aquifers with a fast response and therefore a low
321 memory effect.
- 322 • Slightly decreasing correlogram slope with autocorrelation values above the
323 correlation threshold value over a long time lag. This situation corresponds to
324 poorly developed-activated karst network aquifers, with a large water storage
325 and hence a large behavioural inertia or strong memory response.

326 Despite the easy rule of thumb, the shape of r_X may depend on factors external to
327 the karst systems, such as the characteristics of the precipitation event (i.e. duration,
328 distribution along the year, frequency and intensity) (Grasso and Jeannin 1994;
329 Eisenlohr et al. 1997). As a good practise, the correlogram interpretation should be
330 supported by additional information.

331 The cross-correlation function r_{XY} , provided that it exists, shows the not always
332 evident relationship between a system input $X(t)$ and output $Y(t)$ signal time series,
333 of not necessarily the same variable, such as precipitation and the electrical
334 conductivity of the spring flow. This function is formally defined as

$$r_{XY}(k) = \frac{C_{XY}(k)}{\sigma_X \sigma_Y} \quad (12)$$

$$C_{XY}(k) = \frac{1}{N} \sum_{j=1}^{N-k} (X_j - \bar{X})(Y_{j+k} - \bar{Y}) \quad (13)$$

335

336 Where $C_{XY}(k)$ is the cross-correlogram function and σ_X and σ_Y are the standard
337 deviations of the time series $X(t)$ and $Y(t)$, respectively. If the input signal $X(t)$ into

338 the aquifer system corresponds to a random variable, then the cross-correlation
339 function provides the system response function. As can be shown from equation 13,
340 $r_{XY}(k)$ is not symmetrical if the time series are swapped (i.e. $r_{XY}(k) \neq r_{YX}(k)$).

341 The shape of $r_{XY}(k)$ helps to identify causal relationships between the input and the
342 output system functions (Larocque et al., 1998). Following this line, if $r_{XY}(k) > 0$
343 and $k > 0$, then the system input $X(t)$ affects the system output function $Y(t)$. The
344 opposite happens when $r_{XY}(k) > 0$ but $k < 0$. Additionally, when $r_{XY}(k)$ is
345 symmetrical respect the lag origin ($k = 0$), then the input signal does not influence
346 the output one. In this case, the evolution of both signals is synchronously driven by
347 a third independent signal to be identified by means of a more detailed analysis.

348 The r_{XY} function provides the time delay between the input and output functions at
349 places of maximum similarity. This delay gives an idea of the functional
350 karstification degree of the aquifer. Moreover, the length of such delay provides an
351 estimation of both the pressure pulse transfer times and the advective transit time of
352 a particle migrating through the aquifer (Panagopoulos and Lambrakis, 2006).
353 Therefore, as in the previous case, the shape of $r_{XY}(k)$ allows classifying the karst
354 aquifer as something in between two extreme cases (Mangin, 1984; Padilla and
355 Pulido-Bosch, 1995):

- 356 • Steep cross-correlation slope without or with a small delay of several days
357 are associated to well-developed karst systems, whose well organized
358 channel network favors both a short response time and a fast aquifer
359 drainage.

- 360 • A gentle slope with a long time delay of several weeks, which is associated
361 to poorly drained karst aquifers with long response times, even greater than
362 several months, and large water storage.

363

364

365 **4. Results and Discussion**

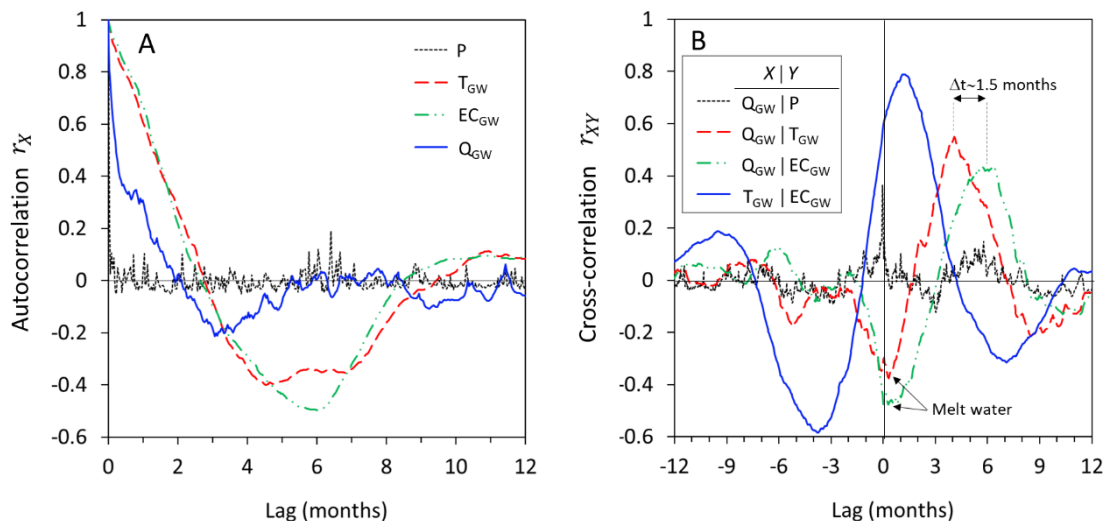
366 The measured hydrograph (Fig. 5) reveals that base flow and quick flow coexist in the
367 karst system discharge. Nevertheless, during winter and early spring, base flow clearly
368 dominates while quick flow is almost absent. At a first glance, this behaviour would
369 suggest the existence of a deficiently organized system and a poorly developed karst
370 conduits network.

371 The results of the autocorrelation analysis show a very small memory effect of 2 days
372 for P, whose autocorrelation function resembles a white noise signal (Fig. 7A). The
373 autocorrelation of Q_{GW} decreases fast and presents an initial first stage with a steep
374 slope during 10 days, which is associated to discharge through the karst conduits. After
375 that time, the slope decreases to the point in which the autocorrelation signal becomes
376 statistically not significant 50 days later. The two slopes in the autocorrelation function
377 of Q_{GW} reveal that the aquifer is a binary karst system, with a total memory effect of 1.5
378 months for Q_{GW} , which is enough to give to the system some storage that allows water
379 resources regulation capacity.

380 The fast reaction of the karst conduit driven discharge can be observed in the cross-
381 correlation function between P and Q_{GW} (Fig. 7B), giving $r_{XY}(0) = 0.35$, where the
382 reaction of the Garcés Spring discharge to precipitation becomes statistically negligible
383 after 2 days ($r_{XY} = 0.15$). Aside, the maximum cross-correlation between Q_{GW} -P is

384 lower than that for the other explored cases which consider Q_{GW} (i.e. $Q_{GW}-T_{GW}$, $Q_{GW}-$
 385 EC_{GW}), indicating that rainfall does not influence greatly the flowrate discharge of the
 386 Garcés Spring in the mid- to long-term. Moreover, the shape of this cross-correlation
 387 function shows some symmetry with respect to the lag origin, pointing out the existence
 388 of other mechanisms behind the scene, such as snowmelt, which drive Q_{GW} . In fact,
 389 there is a negative correlation r_{XY} between Q_{GW} and both T_{GW} and EC_{GW} . This points to
 390 the effect of cold and low mineralized meltwater as the main source for aquifer
 391 recharge.

392 The dissymmetry, the high values of $r_{XY}(k)$ and the similar slopes for positive k values
 393 in the other cross-correlation functions underlines the interdependence between EC_{GW} ,
 394 T_{GW} and Q_{GW} , which is related with the percolation of in-transit recharge through the
 395 unsaturated zone, where the infiltrated water is mineralized and gets warmer along the
 396 percolating flow lines. This is supported by the positive r_{XY} of the pair T_{GW} and EC_{GW}
 397 for $k > 0$. Moreover, the long delay between Q_{GW} as input function and both T_{GW} (~ 4
 398 months) and EC_{GW} ($\sim 5-6$ months) can be ascribed to the travel time of the infiltrated
 399 water pulse mostly in the unsaturated zone but also in the saturated zones (Brown,
 400 1973).



401

402 Fig. 7. Correlation dependence on lag. (A) Correlograms for precipitation (P, dotted
403 line), and groundwater discharge (Q_{GW} , line), temperature (T_{GW} , dashed line) and
404 electrical conductivity (EC_{GW} , dashed-dotted line). (B) Cross-correlograms for Q_{GW} and
405 P (dotted line), Q_{GW} and T_{GW} (dashed line), Q_{GW} and EC_{GW} (line), and T_{GW} and EC_{GW}
406 (dashed-dotted line).

407 The short delay in the cross-correlation Q_{GW} -P reflects the hydraulic effect between
408 precipitation, fast recharge through the conduit system and spring discharge, whereas
409 the other cross-correlation functions (i.e. Q_{GW} - T_{GW} , Q_{GW} - EC_{GW}) show a longer delay,
410 reflecting the effect of an environmental tracer transport effect through the aquifer
411 connected porosity structure. Moreover, cross-correlation Q_{GW} - EC_{GW} is delayed 1.5
412 months respect to Q_{GW} - T_{GW} , indicating that groundwater temperature in the connected
413 porosity zone equilibrates sooner with the respective aquifer conditions, while EC_{GW}
414 tend to be conserved but for some exchange with slow renovation water in the saturated
415 and in the saturated zone. The currently documented existence of mostly temporal
416 perched aquifers is what explains the response time difference between discharge and
417 transport.

418 The Garcés cave system is the terminal drainage of the karst structure developed in the
419 Paleocene-Eocene limestones of the PNOMP in the Ordesa valley sector. For the
420 monitoring period, the discharge of the Garcés Spring shows a seasonal behaviour
421 which is driven by the origin of recharge (Fig. 5). On the one hand, the precipitation (P)
422 events of late spring, summer and autumn generate a sudden response in terms of the
423 spring discharge (Q_{GW}), T_{GW} and EC_{GW} . During these periods, the temperature of the
424 meteoric water that recharges the aquifer is higher than T_{GW} . Besides, the EC associated
425 to precipitation (i.e. rainfall and snowmelt) infiltrating into the system, typically
426 presents a low value around 20 $\mu\text{S}/\text{cm}$ (Lambán et al., 2015), which is always lower

427 than EC_{GW} , due to physicochemical, chemical and isotopic processes during recharge.
428 Therefore, T_{GW} and EC_{GW} show pulses in response to rainfall events, of increase in
429 summer and of decrease in autumn. The synchronicity between the rainfall event and
430 the fast response of Q_{GW} , T_{GW} and EC_{GW} is due to the quick rainfall infiltration through
431 the most conductive karst features, including swallow holes and wide fractures
432 (supplementary materials).

433 On the other hand, in late autumn, winter and spring precipitation fall as snow. As a
434 result, the inflows to the system are mostly controlled by snowmelt. From January to
435 March 2019, the discharge of the Garcés Spring shows a long recession curve that
436 indicates the nil contribution of precipitation to recharge. During this period, EC_{GW}
437 remains stable and T_{GW} shows a slight increasing trend, reflecting the physicochemical
438 fingerprint of the in-transit recharge (Q_{iTR}). This corresponds to the contribution of
439 water percolating slowly through the hydrologically connected porosity of the vadose
440 zone, including small fractures and joints, to the total recharge. From March to June
441 2019 the day time gets longer, the sun elevation becomes higher and the air temperature
442 rises accordingly. As a result, the snowmelt process is activated until the snow pack on
443 the soil surface vanishes. During this period, the cold snowmelt recharges the aquifer
444 system through all the hydrologic features existing in the ground, regardless of their
445 entity (sinkholes, grikes, joints, etc.). This recharge is reflected in the increasing trend of
446 Q_{GW} , with some soft peaks, and the decreasing trend of both T_{GW} and EC_{GW} . Besides
447 the Q_{GW} and T_{GW} trends of this period, these variables also show daily fluctuations that
448 reflect the cycle of (1) morning snowmelt with the corresponding cold water fast
449 infiltration through the most conductive features, and almost downward gravity driven
450 flow through shafts and canyons, which generates both an increase in Q_{GW} and a
451 decrease in T_{GW} , and (2) the evening refreezing that cuts the cold recharge inflows from

452 snowmelt into the system. This allows T_{GW} and Q_{GW} to recover the corresponding
453 values of the system inflows from the in-transit recharge. There is a time lag of 10 h for
454 the melt-water pulse to get Sump-1. This short lag underlines the high velocity of
455 groundwater flowing through the conduits and therefore their corresponding short
456 transit times. This is closely related with the hydraulic effect highlighted by the Q_{GW} - P
457 cross-correlation function.

458 The mean isotopic content ($\delta^{18}O$) in the Garces Spring groundwater discharge is -11.81
459 ‰. The associated recharge zone elevation (Z_R , Eq. 4) is 2429 m. a.s.l.. This value is
460 consistent with those obtained by Jódar et al (2016b) for other springs in the PNOMP.
461 At elevations above 2400 m a.s.l. the snow covers the ground surface during long
462 periods of the year. Nevertheless, the summer high temperatures reduce the presence of
463 snow to the highest elevation zones of the PNOMP. Consequently, from June 2019
464 onwards Q_{GW} decreases and T_{GW} increases. Nevertheless, the evolution of EC_{GW} , which
465 has been similar to T_{GW} up to this moment, shows 1.5 month delay respect to the
466 observed increasing trend in T_{GW} . This is the consequence of the gradual extinction of
467 the snowmelt inflows into the system, while Sump-1 still receives the contribution of
468 the in-transit recharge through the unsaturated zone. This one and a half month delay
469 between EC_{GW} and T_{GW} is clearly reflected in the corresponding cross-correlogram (Fig.
470 7B).

471 It is possible to estimate the contribution of in-transit recharge to the total inflows to
472 Sump-1 using EC as an environmental tracer and conducting a water mass balance
473 given by,

$$Q_{GW} = Q_{FR} + Q_{ITR} \quad (14)$$

$$EC_{GW}Q_{GW} = EC_{FR}Q_{FR} + EC_{iTR}Q_{iTR} \quad (15)$$

474 where Q_{GW} is the groundwater discharge in the Garcés Spring and EC_{GW} is the
 475 corresponding EC which is continuously measured by the CTD-Diver in Sump-1 (Fig.
 476 5). Q_{FR} is the fast recharge entering the system though the conduits and EC_{FR} is the
 477 corresponding EC, whose value is approximated by the 20.55 $\mu\text{S}/\text{cm}$ mean EC value of
 478 precipitation in the PNOMP (Lambán et al., 2015). Q_{iTR} is the in-transit recharge
 479 contribution to the total karst system inflows and EC_{FR} is the corresponding EC value,
 480 which is estimated as the maximum EC value observed in GW throughout the
 481 monitoring period (125 $\mu\text{S}/\text{cm}$). The solution of the equation system in terms of Q_{iTR}
 482 and Q_{FR} is provided by

$$Q_{iTR} = \left(\frac{EC_{GW} - EC_{FR}}{EC_{iTR} - EC_{FR}} \right) \cdot Q_{GW} \quad (16)$$

$$Q_{FR} = Q_{GW} - Q_{iTR} \quad (17)$$

483 For the hydrological year 2018-2019, the estimated mean values of Q_{iTR} and Q_{FR} are
 484 0.61 m^3/s and 0.20 m^3/s , respectively. As the mean system discharges Q_{GW} is 0.81 m^3/s ,
 485 the contributions of Q_{iTR} and Q_{FR} to Q_{GW} are 74.9 % and 25.1%, respectively. This
 486 result highlights the main role of connected porosity and small joints and fractures as
 487 water resource storage in the Paleocene-Eocene karst system, and the importance of the
 488 in-transit recharge to release 3/4 of the total discharge to the downstream depending
 489 ecosystems. Perrin et al. (2003) found similar results for the Milandre karst system in
 490 the Swiss folded and overthrust Jura Mountains. Nevertheless, in this case, not only
 491 the epikarst played a major role as water resources storage, as the existence of a thick
 492 soil horizon provided additional water storage, while protecting the aquifer from
 493 external contaminants that may enter the system. In this line, it is important to note the

494 lack of a well-developed soil layer on top the epikarst zone in the PNOMP. This layer
495 would enhance both the water storage capacity and the protection of the groundwater
496 system in front of a contaminant spill event.

497 The water storage capacity of the hydrologic system is not the only key factor. The
498 transit time is another relevant subject of the aquifer that informs about how long takes
499 the in-transit recharge to get into the system outflow site (τ_{iTR}). This information is
500 obtained through the calibration of the EPM model parameters (τ and η ; Eq. 3) to fit
501 the observed variation of the isotopic content of GW (Fig. 6). The obtained values of η
502 and τ_{iTR} are 3.87 and 1.3 yr (475 days), respectively. The η value points out that V_{PFM} is
503 $2.87 \cdot V_{EM}$, thus accentuating the contribution of the vadose zone to the transit time
504 distribution for the whole hydrological system (see section 3.2.1). The small τ_{iTR} value
505 stresses the vulnerability of the whole hydrogeological system to the impact of climate
506 change and the associated warming trend, which will generate a decrease in the snow
507 precipitation events, thus modifying the recharge system function in terms of a decrease
508 in the snowmelt infiltration, as that obtained by Chen et al., (2018) for a high elevation
509 karst aquifer system in the northern Alps at the end of the 21st century. Moreover, in
510 keeping with those given expectations, Pardo- Igúzquiza et al., (2019) obtained for the
511 same future horizon a recharge decrease of 53% in the aquifer system of Sierra de las
512 Nieves, a privileged observatory of the early impact of climate change in continental
513 Europe, since it is the southernmost high mountain karst system of the Iberian
514 Peninsula.

515 The τ_{iTR} value can be used to estimate the water volume in the connected porosity,
516 joints and fractures (V_{iTR}) of the karst system, which is given by

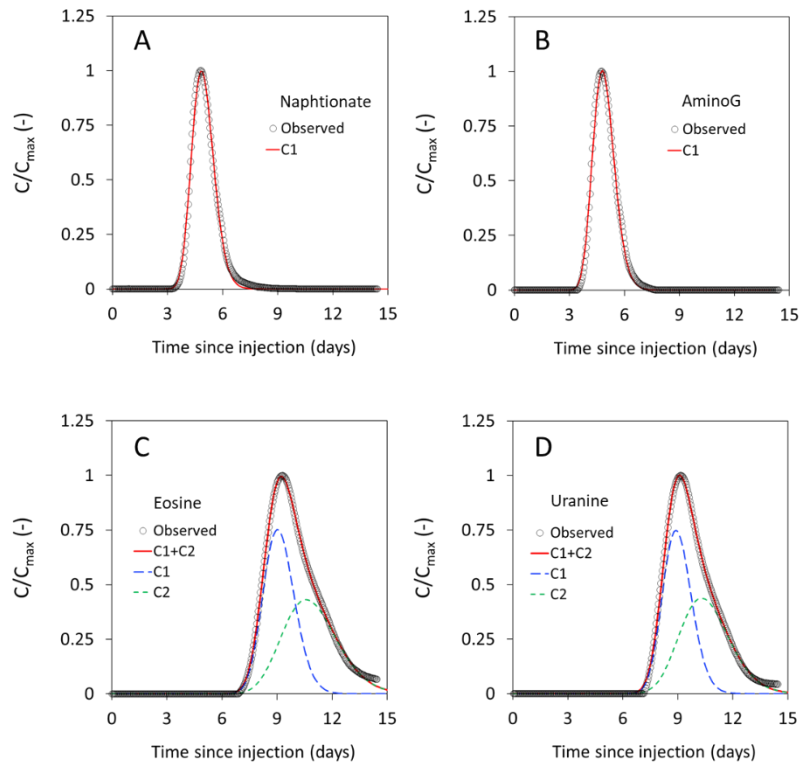
$$V_{\text{ITR}} = Q_{\text{GW}} \cdot \tau_{\text{ITR}} \quad (18)$$

517 For the hydrological year 2018-2019, the value obtained for V_{ITR} is 24.9 hm³. This
518 volume represents 39% and 24% of the mean annual water consumption in the cities of
519 Zaragoza and Barcelona, respectively (Penagos 2007; Barcelona City Council, 2018),
520 and highlights the moderate water resources regulation capacity that has the connected
521 porosity part of the aquifer, despite the associated short memory effect in Q_{GW} (Fig.
522 7A). In fact, the memory effect does not reflect correctly the regulation capacity of the
523 system, but it is more affected by fast recharge. The correlogram does not depend on the
524 values of the of the discharge series but rather on the form of the peaks (Grasso and
525 Jeannin, 1984): the sharper the hydrograph the more rapid the decrease of the
526 correlogram.

527 The fluorescent dye tracers were used to characterize the high conductive features of the
528 karst system through which the fast recharge (Q_{FR}) enters the system. The tracer tests
529 were interpreted by fitting the parameters τ and P_D of Eq. 9, which is the solution of the
530 convolution integral (Eq. 1) for the case of considering the dispersion flow model (DM)
531 as the system response function (Eq. 7). Table 2 shows the calibrated transit time (τ_{FR})
532 and the Péclet number (Pe) for each tracer test, and figure 8 shows the observed and
533 computed concentrations for every tracer test. In each case, the concentrations are
534 normalized by the observed maximum concentration of the test.

535 The tracer recovery decreases as both the distance and the elevation gap between the
536 tracer injection point and the outlet of the system increases. This result may be the
537 consequence of local channel diversions that feed non-controlled diffuse discharge
538 zones in the hydrogeological basin, according to the form of the breakthrough curves
539 (BC) for tracer tests 3 and 4 (Fig. 8C and 8D). Nevertheless, the mapped karst systems

540 show how karst drainage is locally related to fractures and thrust faults. Therefore, some
 541 mass tracer might migrate along steep tectonic structures through the low permeability
 542 Marboré Formation materials towards the underlying Upper Cretaceous aquifer
 543 (Lambán et al., 2015). This process implies the possible existence of a deep regional
 544 drainage mechanism in the PNOMP.



545
 546 Fig. 8. Observed (circles) and calibrated (continuous and dashed lines) breakthrough
 547 curves (BC) for the tracer tests conducted in this work. (A) Tracer test 1, (B) Tracer test
 548 2, (C) Tracer test 3, and (D) Tracer test 4. In the case of tracer test 3 and 4, a two-flow
 549 path model is used. Therefore, C1 and C2 correspond to the BC associated to each one
 550 of the flow paths, and C1+C2 corresponds to the sum of BC1 and BC2, which is the BC
 551 measured at the outflow site of the system.

552 The transit times are close to 5 days for tracer tests 1 and 2, and close to 9 days for
 553 tracer tests 3 and 4, in coherence with longer distance and elevation gap to travel in the
 554 case of these two latter tracer tests. In all cases, the estimated Péclet number (Pe ; Table

555 2) is much greater than 1, revealing advection as the main transport mechanism through
556 the conduits of the karst system, as one would expect from the symmetrical shape of the
557 measured BCs (Fig. 8). The minor role played by dispersion is directly related with the
558 geometrical structure of the conduits in the karst system. According to Hauns et al.,
559 (2001) and Berglund et al., (2020), the conduits geometry, especially in the overly
560 tortuous cases, is the dominant factor generating tracer dispersion at a spatial scale of
561 10^2 to 10^3 m. An advective tracer transport should be likely related with simple conduits
562 geometries. To illustrate this, the supplementary materials include some representative
563 examples of the conduits inside the PNOMP karst system. As can be shown, they
564 present a gentle slope and tubular cross section, without many irregularities, which are
565 typical features of the water table cave pattern.

566 During the tracer tests period, the groundwater flow regime in the conduits was laminar,
567 as indicated by the low Reynolds number (R_e ; Table 2). This reflects the slow discharge
568 of the connected porosity domain into the saturated zone, which is finally guided
569 through the conduits towards the aquifer discharge point in the Garcés Spring.

570 The water storage in the high conductive features can be estimated, as a first approach,
571 as follows (Małoszewski et al., 2002)

$$V_{FR} = Q_{FR} \cdot \tau_{FR} \quad (19)$$

572

573 Taking into account the τ_{FR} values obtained for the tracer test conducted in this work,
574 V_{FR} ranges between 0.076 and 0.156 hm^3 (Table 2).

575 The ratio V_{ITR}/V_{FR} is larger than 10^3 , which is consistent with the low memory effect
576 associated to the conduits by the autocorrelation of Q_{GW} . Moreover, the ratio value

577 underpins the important role played by the connected porosity of the vadose zone as
 578 water resources storage.

579 Similar results of V_{FR} and τ_{FR} were obtained by Lauber and Goldscheider (2014) for a
 580 number of tracer tests conducted in the alpine karst system of the Wetterstein Mountains
 581 during the aquifer low flow period. The short transit times indicate the existence of
 582 well-drained fractures and fissures. Despite of the similarities between the obtained
 583 results, the observed BCs in the Wetterstein Mountains karst system presented a long
 584 tail. This detail reveals that tracer explored a large karst volume before reaching the
 585 sampling (discharge) point (i.e. spring). If the injection and sampling points were
 586 connected by conduits, the corresponding BC would be much less skewed than the
 587 observed ones. In fact, unlike the PNOMP, no large cave systems are known in the
 588 Wetterstein Mountains. This result stresses the importance of speleological explorations
 589 in the karst systems, as they provide direct information of the internal geometry and
 590 actual functioning of the karts systems. They are of paramount importance for correctly
 591 defining the conceptual model of the corresponding aquifers.

592

593 Table 2: Calibrated parameters of the lumped diffusion model obtained for the different
 594 BCs measured in Sump-1.

Tracer test code	Mass recovery (%)	Mean velocity ^(a) (m/d)	R_e ^(b) (-)	V_{FR} (hm ³)	Pe (-)			τ_{FR} (d)		
					Flow Path 1	Flow Path 2	Average	Flow Path 1	Flow Path 2	Average
1	95	645	1952	0.077	133.9	--	133.9	4.96	--	4.96
2	99	571	1727	0.076	136.5	--	136.5	4.90	--	4.90
3	30	319	966	0.156	229.0	104.2	166.6	9.15	10.89	10.02
4	12	409	1236	0.152	257.9	119.1	188.5	9.00	10.56	9.78
Average	59	486	1470	0.115	189.3	111.7	156.4	7.00	10.73	7.41

(a) ratio between the tracer test distance (Table 1) and the conduit mean transit time τ_{FR} ; (b) Reynolds number estimated as $R_e=V \cdot L/\nu$, where V is the groundwater velocity, L is the channel characteristic length estimated as 0.4 m from the geometry of the flooded conduits revealed in the pictures taken during the dye tracer injection day (supplementary materials), and ν is the kinematic water viscosity, equal to 0.13 m²/d for a water temperature 4.8 °C (Engineering ToolBox, 2004), which corresponds with T_{GW} in the Garcés Spring the day of the tracers injection.

595

596 The Paleocene-Eocene limestones of the Gallinero Formation constitute the main
597 aquifer of the PNOMP. The hydrodynamic behaviour of this aquifer corresponds to a
598 binary karst system with both conduit and diffuse components. The results show that the
599 connected porosity of the formation provides 3/4 of the total groundwater discharge in
600 the Garcés Spring, while providing almost all the water storage of the aquifer (Fig. 9).
601 The porosity of the limestones is not known. Nevertheless, such parameter can be
602 estimated as a first approach as

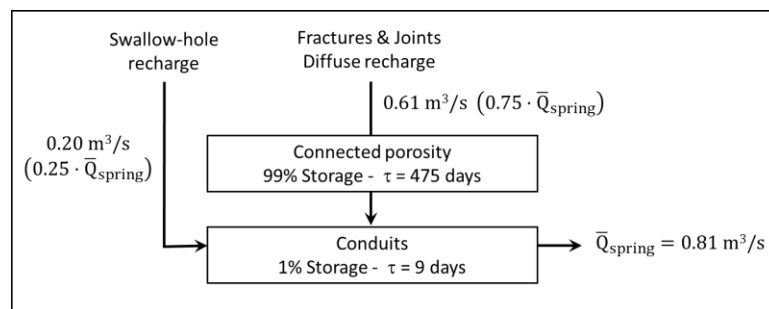
$$\phi = \frac{V_{iTR} + V_{FR}}{V_{aq}} \quad (20)$$

603 where V_{aq} is the aquifer formation volume. V_{aq} can be evaluated as the product of the
604 18.1 km^2 aquifer surface area and the aquifer thickness, estimated to be equal to the 250
605 m mean thickness of the Gallinero formation (Robador et al., 2018). Taking into
606 account these values, a connected porosity of 0.6% is obtained, which is low but still
607 consistent with those obtained for other mountain karst systems, such as 1.5% in the
608 Schneecalpe (Maloszewski et al., 2002) or 2% in the Swabian Alps (Sauter,1993). In the
609 case of the karst aquifer drained by the Garcés Spring, the associated dual-porosity
610 model, in which conduit and fracture porosity co-occur by contributing 1/4 and 3/4,
611 respectively, to the total groundwater discharge, it can be assumed that most of the flow
612 in the saturated zone is probably Darcian. Despite of that, the combination of the small
613 porosity with high permeability of the most conductive features of the karst aquifer
614 results in groundwater velocities along conduits that may have important implications
615 for contaminant transport (Worthington, 2015). The short transit times of both conduits
616 and connected porosity minimize the degradation and dilution of possible contaminants

617 entering the system, e.g. faecal bacteria that would be transported to the spring within a
618 short period of time.

619 Moreover, the low memory of the aquifer system revealed by the autocorrelation
620 analysis informed about the relatively small storage capacity of conduits along with the
621 karstification degree of the system. All these characteristics enlighten the high
622 vulnerability of the karst aquifer system drained through the Garcés Spring.

623



624

625 Fig. 9. Two-box model for the Paleocene-Eocene carbonate aquifer of the PNOMP,
626 with fraction of storage (expressed in percentage of total) and residence time in each
627 box, and groundwater flow between boxes. See text for details.

628

629 Characterizing the hydrodynamical behavior of karst systems is not easy, given the
630 heterogeneity of the karstic massifs and the complexity of the karst networks developed
631 inside, whose structure and complexity depends on the geological setting, the recharge
632 rates and time (Audra and Palmer, 2011; 2015). In high mountain areas, the
633 characterization of karst aquifers is even more difficult given the rough topography that
634 hampers the access, transportation of materials and normal operation. Besides, the
635 severe climatic conditions prevent accessing the area during long time periods. The
636 karst aquifer described here has many points in common with other karst systems in

637 high mountain zones, where diffuse infiltration of snowmelt conditions karst structure
638 and consequently its hydrodynamic behaviour (Lauber and Goldscheider, 2014).

639 The snowmelt is thus the process behind the geomorphological structure and
640 hydrodynamic behaviour of the PNOMP karst system. The mean persistence of the
641 snow cover during 2/3 of the hydrologic year conditions the lack of both a highly
642 developed epikarst zone and a surficial semi-confining layer. Nevertheless, the snow
643 pack plays the same role as the surficial semi-confining layer. The snowmelt flows
644 diffusely through the snow pack minimizing surface runoff. The slow snowmelt rate
645 favors water infiltration through the network of joints and fractures, thus maximizing
646 aquifer recharge while maintaining moderate drainage rates. The aquifer base flow is
647 controlled by the in-transit recharge in the saturated zone. This flow component is
648 Darcian and dominates the aquifer system discharge during 2/3 of the hydrologic year.
649 The role played by the quick flow component flowing through the conduits is only
650 relevant during late spring, summer and early autumn, which is the time when the
651 convective storms are produced in the Pyrenees (Callado and Pascual, 2005). Taking
652 into account that the conduits of a matured karst carry the majority of flow and control
653 the hydrogeological behaviour of the system (e.g., White, 1988; Ford and Williams,
654 2007), which is not the case in the PNOMP, and the existence of an epiphreatic zone
655 with a water table cave pattern, it seems reasonable to think that the PNOMP karst
656 system is still young, even containing highly conductive conduits inside.

657 In this work, the combined use of both environmental and fluorescent dye tracers along
658 with time series analysis of physicochemical and hydrological variables allowed to
659 characterize the behaviour of the karst systems drained by the Garcés Spring. The
660 applied methodology may be useful to characterize other alpine karst aquifer systems.

661

662 **5. Conclusions**

663 The snowmelt recharge controls both the geomorphological evolution and maturity of
664 the karst system and the corresponding hydrological behaviour.

665 The karst system behaves as a dual-porosity aquifer, with a highly permeable domain
666 that includes the karst conduits, and a connected porosity domain lumping joints,
667 fractures and primary inter-particle porosity. The first one controls the system response
668 during the rainfall events whereas the latter does it during the snow accumulation
669 seasons until the beginning of summer with the end of thaw.

670 The in-transit recharge controls the T and EC dynamics in the phreatic zone. Moreover,
671 the water storage in the system is almost linked to the connected porosity structure,
672 which is drained at low flow rates into the saturated zone, where the groundwater flow
673 is Darcian. Despite of that, the combination of the low porosity with the high
674 permeability of most conductive features may have important implications for
675 contaminant transport.

676 The low transit times in both the conduits and the connected porosity domains reveal
677 the high vulnerability of the karst system to contaminant issues and the impact of
678 climate change.

679

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693 study zone.

694

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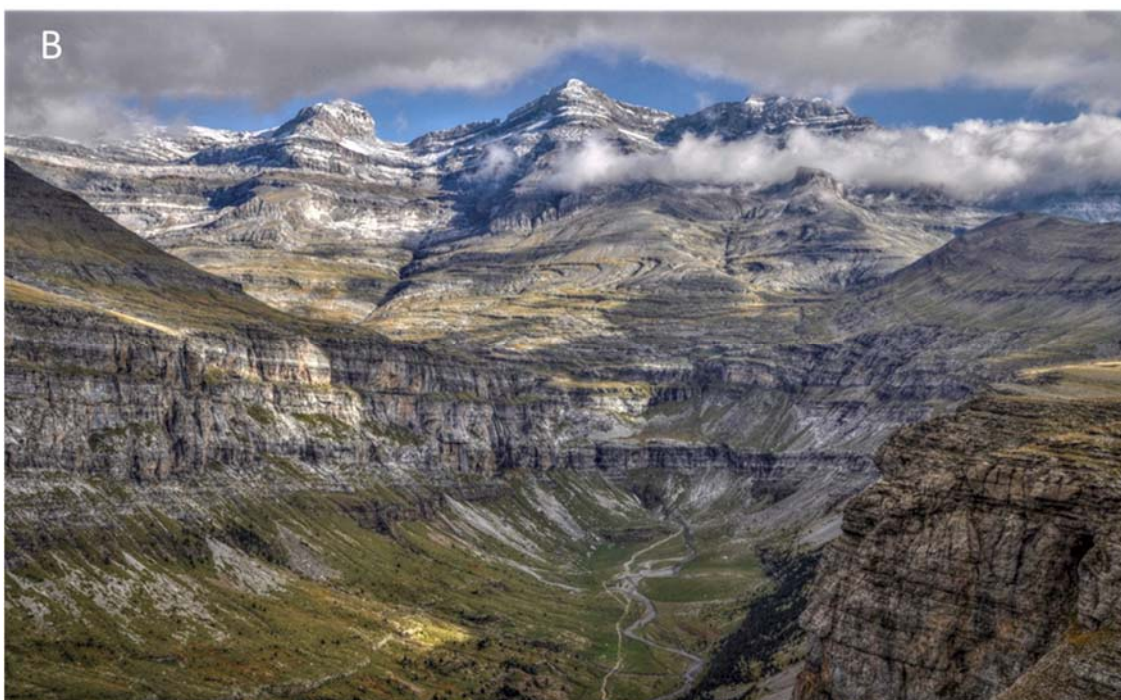
Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

The authors: The authors: Jorge Jódar, Antonio González-Ramón, Sergio Martos-Rosillo, Javier Heredia, Christian Herrera, Javier Urrutia, Yván Caballero, Ane Zabaleta, Iñaki Antigüedad, Emilio Custodio, and Luis Javier Lambán

1 **Supplementary material**



2
3 Fig. SM1. Views of the Soaso glacial cirque located in the upper Arazas River, and the
4 Monte Perdido massif. (A) April 2019. (B) September 2019

5

Garcés Cave and Spring



Fig. SM2. (A) Silvia-Coll's Sump in the Garcés Cave. (B) Gracés Spring

Cola de Caballo water fall



11

12 Fig. SM3. Cola de Caballo waterfall. (A) Nov.-2018, Chavier Lozano. (B) Mar.-2019.

13 Aug.-2019, (D) Oct.-2019

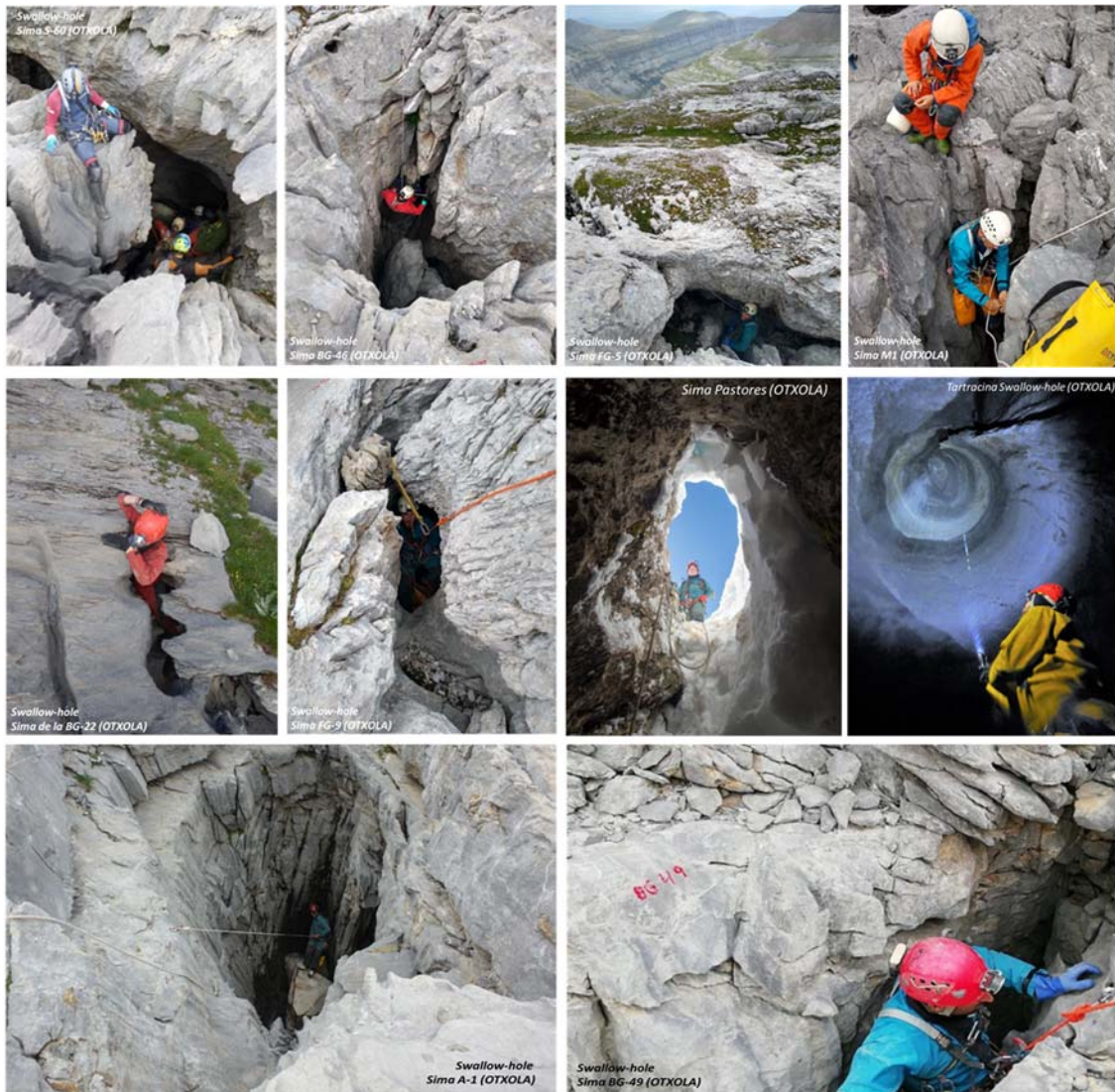
Joints and fractures



Fig. SM4. Joints and fractures in the study area

17

Swallow-holes



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Fig. SM5. Swallow-holes in the study area

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Karst galleries



Fig. SM6. Galleries in the Garcés and Tartracina karst systems.