Barremian–early Aptian charophyte biostratigraphy revisited

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Abstract

New Barremian–early Aptian charophyte biozonations are proposed here based on the charophyte succession in the Maestrat Basin (Iberian Chain, Spain). Biostratigraphic analysis distinguished two biozonations, European and Eurasian, which were compared to establish correlations. This is the first time that Early Cretaceous charophyte biozones are calibrated by means of strontium-isotope stratigraphy, enabling their correlation with the coeval marine realm (ammonite biostratigraphy). The European charophyte biozonation is formed of two partial range biozones, whose index species were endemic from the Cretaceous Tethyan Archipelago (present-day Europe and Northern Africa):

(1) Globator maillardii var. trochiliscoides (early Barremian) and (2) Ascidiella cruciata-Pseudoglobator paucibracteatus. The base of the A. cruciata-P. paucibracteatus biozone was calibrated with an oyster shell sample whose \({^{87}}\text{Sr}/^{86}\text{Sr}\) is 0.707482, translating to a late early Barremian age. The total time span of this latter biozone is late early Barremian–early Aptian. The Eurasian biozonation involves a novel Lower
Cretaceous charophyte biostratigraphy. It has more resolution than the European biozonation, as it is composed of three partial range biozones whose index species were well-distributed in Eurasia. From oldest to youngest these are: (1) Atopochara trivolvis var. triquetra, (2) Hemiclavator neimongolensis var. neimongolensis, and (3) Clavator grovesii var. jiuquanensis. The A. trivolvis var. triquetra biozone characterises the early Barremian, being almost equivalent to the European G. maillardii var. trochiliscoides biozone. The base of the H. neimongolensis var. neimongolensis biozone is marked by the first appearance datum (FAD) of *Hemiclavator neimongolensis* var. *neimongolensis* and was calibrated with an oyster shell whose $^{87}\text{Sr}/^{86}\text{Sr}$ is 0.707481 corresponding to a late early Barremian age. The top of the H. neimongolensis var. neimongolensis biozone coincides with the FAD of *Clavator grovesii* var. *jiuquanensis*. The base of the C. grovesii var. jiuquanensis biozone was dated with an oyster sample sample collected 25 m below the FAD of *C. grovesii* var. *jiuquanensis*. The $^{87}\text{Sr}/^{86}\text{Sr}$ value of this sample is 0.707489, which translates into an early late Barremian age. Thus, the H. neimongolensis var. neimongolensis biozone spans the late early Barremian-early late Barremian interval. The top of the Clavator grovesii var. jiuquanensis biozone (late Barremian–early Aptian) is marked by the FAD of *Clavator grovesii* var. *lusitanicus*. The newly proposed Eurasian charophyte biozonation will facilitate the correlation between non-marine basins in one of the largest continental areas on Earth.

**Keywords:** Charophyta, Clavatoraceae, Lower Cretaceous, Iberia, China, Intercontinental correlation, Biochronology

1. **Introduction**

The fructifications (utricles) of the fossil charophyte family Clavatoraceae are widely used for the biostratigraphic characterisation of continental Late Jurassic-Early
Cretaceous successions (e.g., Grumbast 1974, Wang and Lu 1982, Mussacchio 1989, 2000, Schudack 1987, 1993, Martín-Closas 1989, Martín-Closas and Alonso-Millán 1998, Feist et al. 1995, Trabelsi et al. 2016). A number of Barremian-lower Aptian charophyte biozonations have been proposed in Europe (e.g., Grumbast 1974; Feist et al. 1995, Riveline et al. 1996, Mojon 1996, 2002) and China (e.g., Wang and Lu 1982, Peng et al. 2003, Yang et al. 2008). Despite the extensive work carried out on clavatoracean biostratigraphy in Europe and Asia, the different biozones proposed have not been numerically dated or correlated between continents. The relative ages of previous proposals were determined from the correlation with foraminifera found in marine deposits, which occur interbedded within continental successions (e.g., Combes et al. 1966, Martín-Closas and Salas, 1988, 1994, Mojon 2002, Martín-Closas et al. 2009). Li et al. (2020) dated charophyte assemblages from Chinese basins to the late Barremian-early Aptian time by using previously published chemostratigraphic data and by considering the age assigned to similar charophyte assemblages in European basins.

In Europe, two Barremian biozones were first proposed by Grumbast (1974) spanning the early Barremian and the late Barremian, respectively. These biozones were the bases for the later proposals (e.g., Feist et al. 1995, Mojon 1996). The most used present-day European Barremian biozonation was proposed by Riveline et al. (1996) and later modified by Martín-Closas et al. (2009). These authors used assemblages and ages similar to those reported by Grumbast (1974).

In Asia, different charophyte biostratigraphic charts have been proposed for the Chinese basins. Wang and Lu (1982) defined two biozones that have Hauterivian-early Barremian and late Barremian age ranges, respectively. Later on, Peng et al. (2003) combined these two biozones into a single biozone spanning the whole Barremian, and Yang et al. (2008) considered this latter biozone to also span the Hauterivian Stage. This relatively low-resolution charophyte biostratigraphy contrasts with the proposals
for other Cretaceous stages whose time span is similar to that of the Barremian. This is
the case, for example, with the late Campanian-late Maastrichtian European and Asian
charophyte biostratigraphy that involve three biozones (e.g., Vicente et al. 2015, Li et
al. 2019).

This paper presents new European and Eurasian charophyte biostratigraphic
frameworks for the Barremian-early Aptian, based on the clavatoracean succession
identified in the Maestrat Basin (Iberian Chain, Spain) and the extensive data on
clavatoracean biogeography available for this time span elsewhere (e.g., Martín-Closas
and Wang 2008, Martín-Closas 2015, Pérez-Cano et al. 2020). The analysis of this
charophyte record along with a continuous Barremian succession enabled us to
determine the first appearance datum (FAD) of several species from the Barremian,
improving the resolution of the Barremian biostratigraphy. Moreover, the identification
of the FAD of Eurasian-distributed taxa (i.e., *Atopochara trivolis* var. *triquetra*,
*Clavator grovesii* var. *jiuquanensis*, *C. harrisii*, *C. calcitrapus*, and *Hemiclavator
neimongolensis* var. *neimongolensis*; Martín-Closas and Wang 2008, Martín-Closas
2015) enabled their use as index species, improving the correlation between Europe and
Asia.

The utility of a biostratigraphic framework increases when it is faithfully
calibrated to a time scale. Charophyte biozonations from other ages have been
calibrated against the Geomagnetic Polarity Time Scale (GPTS), which translates into
numerical ages. This has been performed for the Campanian–Danian charophyte
biozonations of the Pyrenees (Galbrun et al. 1993, Vicente et al. 2015) and the Chinese
basins (Li et al. 2016, 2019), as well as for the Eocene biozonation of the Ebro Basin
(Sanjuan et al. 2014). However, the calibration of Late Jurassic-Early Cretaceous
charophyte biozones in southern European basins with the GPTS is not straightforward
due to the Alpine Orogeny-related remagnetisation of the Mesozoic record (e.g.,
Moreau et al. 1992, Gong et al. 2009). Consequently, the calibration of the Barremian-early Aptian biostratigraphy presented here was performed by numerically dating marine intercalations using strontium isotope stratigraphy (SIS), a methodology extensively used for the calibration of marine records (e.g., Steuber 1999, Williamson et al. 2012, Frijia et al. 2015, Bover-Arnal et al. 2016, Caus et al., 2016).

2. Geological setting

The Maestrat Basin was part of an intracratonic rift system that developed in the NE part of the Iberian plate (Fig. 1A–B) during the Mesozoic as a consequence of the breakup of Pangaea and the opening of the Central Atlantic Ocean (Salas and Casas 1993, Salas et al. 2001). According to Salas et al. in Martin-Chivelet et al. (2019), there were three different stages of rifting followed by three post-rift phases in the Iberian rift system: (1) Late Permian–Triassic rifting, (2) Early–Middle Jurassic post-rift, (3) Late Jurassic–middle Berriasian rifting, (4) late Berriasian–Hauterivian post-rift, (5) Barremian–early Albian rifting and (6) Late Cretaceous post-rift. The Maestrat Basin is associated with the Late Jurassic–middle Berriasian and the Barremian–early Albian rifting stages. Listric extensional faults and palaeotopographic highs subdivided the basin into several sub-basins, from north to south, namely El Perelló, Morella, Oliete, Las Parras, Galve, Salzedella, Orpesa, Penyagolosa and Cedramán, with different subsidence rates and sedimentary features (Fig. 1B; Salas and Guimerà 1996, Salas et al. in Martin-Chivelet et al. 2019). The study areas are located within the Morella Sub-basin (Fig. 1B–D). The sedimentary deposition in this sub-basin ranged from non-marine carbonate successions, mainly found along the northern margin of the half-graben, to hemipelagic successions occurring in the central zone of the sub-basin (Canérot et al. 1982, Salas 1987). Later, during the Alpine orogeny (Late Cretaceous–
early Miocene), the Iberian rift system was inverted to form the Iberian Chain (Nebot and Guimerà 2016, Guimerà 2018).

The Barremian sedimentary record of the Morella Sub-basin is subdivided into five formations: Cantaperdus, Artoles, Morella, Cervera del Maestrat and Xert (Fig. 2; Canérot et al. 1982, Salas 1987, Salas et al. 2001, Bover-Arnal et al. 2016). The present study was carried out along the Cantaperdus, Artoles and Morella formations (Fig. 2).

The non-marine lacustrine Cantaperdus Formation is mainly composed of alternating charophyte-rich marls and limestones. It is up to 400 m-thick in the northern area of the Morella Sub-basin and becomes thinner towards the central part of the basin, where it laterally changes into the Artoles Formation. The Artoles Formation, which also overlies the Cantaperdus Formation throughout the sub-basin, is characterized by coastal to shallow open marine marls and limestones rich in oysters (Salas 1987, Salas et al. 2001, Bover-Arnal et al. 2016). This unit is up to 700 m thick in the central parts of the sub-basin, with its thickness decreasing to less than 200 m in the marginal areas.

Laterally and above the Artoles Formation, there is the Morella Formation, a non-marine unit composed of grey to red and purple clays and sandstones (Canérot et al. 1982, Salas, 1987, Gàmez et al., 2003). Towards the depocentre of the sub-basin, the Morella Formation becomes progressively more brackish-influenced, laterally passing to the Cervera del Maestrat Formation that contains mixed carbonate-siliciclastic coastal and shallow marine deposits (Canérot et al. 1982, Salas, 1987, Salas et al. 2001). The Xert Formation corresponds to a fully marine succession composed of marls and limestones rich in orbitolinids. The Morella, Cervera del Maestrat and Xert formations
have been dated to the late Barremian using ammonites and SIS (Garcia et al. 2014, Bover-Arnal et al. 2016).

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3. Palaeobotanical setting

Since the middle of the 20th century, the taxonomic studies carried out by Louis Grambast (Grambast 1966a, b, 1967, 1968, 1969, 1970, 1974) and later continued by Martín-Closas and Grambast-Fessard (1986), Martín-Closas (1989, 2000), Schudack (1993), Climent-Domènech et al. (2009) and Pérez-Cano et al. (2020) have illustrated the high biodiversity of the charophytes occurring in the Lower Cretaceous non-marine record of the Maestrat Basin. These studies showed that the extinct Clavatoraceae family dominated the non-marine settings of the basin during that period. The stratigraphic continuity of the non-marine record of the Maestrat Basin provides an excellent context to understand the evolution and phylogeny of the clavatoraceous species, enabling the identification of evolutionary lineages with biostratigraphic significance (Grambast, 1966a, 1968, 1974; Martín-Closas, 1989, 1996, 2000). The species identified in the studied sections are summarised in Table 1 and shown in Plates 1 and 2.

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4. Materials and methods

4.1. Lithostratigraphy and charophyte sampling

Two Barremian sections from the Morella Sub-basin were logged and systematically sampled for the identification of charophyte assemblages: Herbers-Mas de Petxi (base: 40°42’39”N, 0°0’36”W; top: 40°42’34”N, 0°0’30”E) (Figs. 1C and 3A–
Samples of approximately 3 kg were taken from each marl or clay bed. In the laboratory, the samples were disaggregated using a solution of water, sodium carbonate (Na$_2$CO$_3$; useful for deflocculating the clay), and hydrogen peroxide (H$_2$O$_2$; eliminates the organic matter and helps with the disaggregation). These samples were left for a few days and then sieved through meshes with apertures of 1, 0.5, and 0.2 mm. When the samples were dried, charophyte fructifications were handpicked using a Wild M5A binocular microscope. The fructifications were measured using a Motic BA310 microscope and the Motic Images Plus 2.0 software. Selected specimens were studied and photographed with a Quanta 200 Scanning Electronic Microscope (SEM) at the Centres Científics i Tecnològics de la Universitat de Barcelona (CCiTUB). The figured specimens were deposited at the Museu de Geologia del Seminari Conciliar de Barcelona (MGSCB). The handpicked material has been kept at the Departament de Dinàmica de la Terra i de l’Oceà of the Universitat de Barcelona.

Limestone beds were also sampled and studied as c. 30 µm-thick sections cut parallel and perpendicular to the bedding surface of the sample. These sections were studied and photographed with a Motic BA 310 petrographic microscope.

4.2. Trace element concentration and strontium-isotope analysis

Eleven oyster shells were sampled for SIS. Four oysters were obtained from the Fredes section, all of them belonging to the Cantaperdus Formation (Fig. 4). In the Herbers-Mas de Petxí section, six oyster samples were collected from the Artoles
Formation and one sample was gathered from the upper part of the Morella Formation close to the top of the non-marine record (Fig. 3A–B).

Oyster shells were cleaned to remove superficial contamination and later polished and screened for diagenetic changes. Visual inspection of the shells helped to prevent the sampling of bioperforated parts, cements, and other alterations within the shell structure. The externally non-altered parts of the shells were sampled with a hand-operated microdrill equipped with a tungsten drill bit (0.3 to 0.5 mm in diameter) and homogenised in an agate mortar mill. The resulting carbonate powder from each shell was divided into two samples of ~100 mg. The first sample was employed for the Inductively Coupled Plasma-Optical Emission Spectroscopy (ICP-OES) analysis of Ca, Sr, Mg, Fe and Mn concentrations in order to perform diagenetic screening. This was carried out at the Centres Científics i Tecnològics de la Universitat de Barcelona (CCiTUB). The second sample was used for SIS, which was performed at the Unidad de Geocronología, CAI de Ciencias de la Tierra y Arqueometría at the Universidad Complutense de Madrid (Spain). Strontium (Sr) was separated from the sample by standard ion-exchange resin methods. The Sr isotope ratios ($^{87}$Sr/$^{86}$Sr) were measured on a TIMS-Phoenix® mass spectrometer. The analysis was corrected to avoid interferences from $^{87}$Rb, and the $^{87}$Sr/$^{86}$Sr ratio was normalised to the mean value of 0.1194. The analysed raw results were corrected using the standard NBS 987, which has a value of 0.710249 ± 0.000012 (2 statistical uncertainty, $n = 7$).

5. Stratigraphy and sedimentology

A sedimentological analysis was performed in the Herbers-Mas de Petxí and Fredes sections to determine the relationship between the presence or absence of charophyte species and the depositional facies. Taphonomical features such as fragmentation and abrasion, as well as the occurrence of different charophyte organs
together or even connected anatomically (i.e., fructifications and thalli), were investigated to elucidate the degree of autochthony or allochthony of the charophyte remains in the sedimentary record.

5.1. The Herbers-Mas de Petxí section

This section crops out to the south and south-east of the village of Herbers (Castelló, Spain; Fig. 1C) and contains an 800 m-thick continuous Barremian succession. The present study focuses on the lower 690 m, corresponding to the Cantaperdius, Artoles, and Morella formations (Figs. 2 and 3A–B).

The Barremian succession is deposited above an irregular karstified surface exhibiting vertical tubular structures that are interpreted as root marks marking the top of the Hauterivian Herbers Formation (Fig. 2). The lowermost part of the Barremian record (0–11 m), which corresponds to the base of the Cantaperdius Formation, consists of variegated clays that change from a greyish colour at the base to a yellowish colour in the middle and a reddish colour at the top. Similar deposits at the base of the Barremian record in the Iberian Chain have been interpreted as laterite deposits (Combes 1969).

These clays contain charophyte remains (thalli and fructifications), ostracods and molluscan fragments. Up to 0.5 m-thick packstones occur, interbedded with the clays. The charophyte assemblage in the clays is mainly composed of utricles of well-preserved *Atopochara trivolvis* var. *triquetra*, *Clavator harrisii* var. *harrisii*, *Ascidiella stellata* var. *stellata* and *A. triquetra*, as well as gyrogonites of aff. *Mesochara harrisii*. Charophyte thalli portions, *Munieria grambastii* Bystrický and *Favargerella aquavivae* Martín-Closas et Salas, have been also observed, suggesting that the charophyte assemblage is autochthonous. Utricles of *Hemiclavator adnatus* and *H. neimongolensis* var. *posticecaptus* are rare and occur abraded. They are therefore interpreted as
allochthonous components. Above these clays, the section has been divided into eight
descriptive intervals, from A to H (see Fig. 3A-B).

**Interval A** (11–161.5 m) mainly consists of charophyte-rich massive dark-grey
limestones and marls that are stacked in parasequences. The marls are frequently absent
from the parasequences, but when they do occur, they consist of thin (<20 cm) layers
that contain abundant clavatoracean utricles associated with thalli and ostracods. The
marls are interbedded with limestones whose texture varies among mudstone,
wackestone and packstone and contain abundant clavatoracean utricles and thalli,
266 ostracods, and black pebbles (<1 mm across). These limestones often show colour
mottling. The top of the packstones is characterised by the presence of root marks.
Successions with these sedimentary features have been attributed to shallow carbonate
lakes laterally associated with palustrine environments (see, e.g., Gierlowski-Kordesch
2010, Alonso-Zarza and Wright 2010). Similar sedimentary settings have been
described by Platt (1989) and Meléndez et al. (2009) in other parts of the Lower
Cretaceous Iberian Chain.

The charophyte assemblages in this interval are mainly of *A. trivolvis* var.
*triquetra*, *Clavator harrisi* var. *dongjingensis*, var. *harrisi*, and var. *reyi*, *Ascidiella*
*stellata* var. *stellata*, and var. *lata*, *A. triquetra* and, rarely, *Globator maillardii* var.
*trochiliscoides*, *Hemiclavator adnatus*, *H. neimongolensis* var. *posticecaptus*, aff.
*Mesochara harrisi* and *Porochara maestrichta*. Most samples contain well-preserved
fructifications that are associated with the charophyte thalli of *F. aquavivae*,
*Clavatoraxis* sp., *Charaxis* sp., and *M. grambasti*. Utricles of the genus *Ascidiella* are
locally found attached to *F. aquavivae* thalli portions while *C. harrisi* var. *harrisi*, *H.*
adnatus and *H. neimongolensis* var. *posticecaptus* are attached to *Clavatoraxis* sp.
phylloids. Taphonomic data show that these assemblages can be regarded as
autochthonous. Populations of *P. maestrichta* are frequently small, but, locally, this
species can dominate charophyte assemblages. Porocharacean-rich autochthonous assemblages from the Lower Cretaceous are interpreted as evidence of brackish water conditions (e.g., Martin-Closas and Grambast-Fessard 1986, Schudack 1993, Climent-Domènech et al. 2009).

**Interval B** (161.5–187 m) is formed of interbedded marls and limestones, with the marl beds being more abundant and thicker than those in Interval A. The marl layers are <50 cm thick and contain abundant charophyte thalli and fructifications (utricles and gyrogonites), ostracods, gastropods and, rarely, small agglutinated benthic foraminifera. Limestones are up to 1 m-thick and exhibit wackestone and packstone textures with a palaeontological content similar to that observed in the limestones of Interval A. Root marks at the top of the limestone beds also occur, suggesting deposition in very shallow lake and palustrine environments (cf. Alonso-Zarza and Wright 2010). As in Interval A, Interval B presents indications of lacustrine deposition with scarce palustrine intercalations and a low exposition rate. However, the thicker marl layers in Interval B compared to Interval A suggests that the freshwater alkaline lakes received higher clastic input than those described for Interval A (e.g., Meléndez et al. 2009).

The charophyte assemblage in this interval is similar to that observed in the interval A (Fig. 3A). The only differences are the first occurrences of *Echinochara lazarii* and, locally, *Globator maillardii* var. *biutricularis*. Moreover, *G. maillardii* var. *trochiliscoides* is more frequent and abundant here than in the underlying interval. The last occurrences of *A. stellata* var. *stellata* and *Hemiclavator adnatus* are also recorded in this interval. Most samples contain well-preserved fructifications that are associated with thalli, suggesting autochthony. However, the gyrogonites of *P. maestratica* and the benthic foraminifera are frequently broken and/or abraded, which indicates lateral transport.
**Interval C** (187–276 m) is mainly composed of a succession of clavatoracean-rich mudstone to packstones, resulting in a crest landscape. Occasionally, the limestones are interbedded with <20 cm-thick marl beds. The palaeontological content of the limestones and marls is similar to that of Interval A. They also show intraclasts and root marks at the top, sedimentary features that indicate lakeshore environments (e.g., Gierlowksi-Kordesch 2010; Alonso-Zarza and Wright 2010).

The charophyte assemblages in this interval are composed of *E. lazarii, A. trivolvis* var. *triquetra, C. harrisii* var. *harrisii*, and var. *reyi, Ascidiella stellata* var. *lata, A. triquetra, H. neimongolensis* var. *posticecaptus and H. neimongolensis* var. *neimongolensis* and gyrogonites of *P. maestratica* and aff. *M. harrisii*. They are mostly formed of well-preserved fructifications associated with thalli, indicating that the observed charophyte remains are autochthonous. The occurrence of porocharacean-dominated limestones interbedded with clavatoracean-rich limestones is interpreted as evidence of brackish water environments laterally associated with freshwater lakes, similar to Interval A.

**Interval D** (276–366 m) is another interval with abundant marl and comprises the uppermost part of the Cantaperdus Formation and the lowermost part of the Artoles Formation. The lower part of the interval (276–348 m) is composed of a monotonous succession of interbedded marls and packstones. While marls contain abundant charophyte fructifications and thalli, ostracods, and gastropods, limestones contain abundant charophyte thalli (mainly *M. grambastii*), few charophyte fructifications, and large intraclasts (>1 cm in diameter). Root marks and ferruginous surfaces are common at the top of the limestones. The facies slightly change in the upper part of the interval (348–366 m). The marls of this upper part contain abundant, but poorly diverse
clavatoracean utricles, together with small agglutinated benthic foraminifera, and few ostracods. These marls are interbedded with mudstone-wackestone limestones containing a few charophyte utricles and thalli as well as ostracods. They are also occasionally interbedded with oyster-bearing packstones. The lower part of the interval is interpreted as deposited in a lacustrine setting. The root-marked limestones correspond to lakeshore areas surrounded by a palustrine zone, similar to those described by Meléndez et al. (2009) and Alonso-Zarza and Wright (2010) in other Lower Cretaceous Iberian basins. Upwards, the succession shows a progressive substitution of the freshwater lake facies by the brackish and oyster-bearing coastal marine deposits of the Artoles Formation (Fig. 2), which mark the onset of the early Barremian transgression in the basin (Bover-Arnal et al. 2016).

The charophyte assemblages of Interval D are mainly composed of *E. lazarii*, *G. maillardii* var. *trochiliscoides*, and var. *biutricularis*, *A. trivolvis* var. *triquetra*, *C. harrisii* var. *dongjingensis* (last occurrence), var *harrisii*, and var. *reyi*, *Clavator calcitrapus* var. *jiangluoensis*, and var. *calcitrapus* (first occurrence), *Hemiclavator neimongolensis* var. *neimongolensis*, and aff. *M. harrisii*. Occasionally, they also include *Ascidiella stellata* var. *lata*, *A. triqueta*, *H. neimongolensis* var. *posticecaptus* (last occurrence) and *P. maestratica*. Most samples show well-preserved utricles and gyrogonites, occasionally attached to thalli. They are thus considered to be autochthonous. The gyrogonites of *P. maestratica* show abrasion, indicating reworking or lateral transport.

**Interval E** (366–450 m) represents the lower part of the Artoles Formation (Fig. 3B). It begins with 30 m of interbedded marls and wackestone limestones rich in well-preserved gyrogonites of *P. maestratica*, small agglutinated benthic foraminifera, and ostracods, indicating brackish water settings, as interpreted for Intervals A and C. The marl layers can also contain scarce utricles of *E. lazarii*. These porocharacean-rich beds
are overlaid by 54 m of mudstone to packstones that contain agglutinated benthic foraminifera, miliolids, oyster shells, gastropods, and rare porocharacean gyrogonites. In these facies, porocharacean gyrogonites are slightly abraded, suggesting lateral transport. These marine limestones are interpreted as shallow water facies that were deposited in marginal coastal areas of a carbonate shelf (e.g., Tucker 1985).

Accordingly, Interval E shows an evolution of depositional environments from brackish water settings dominated by porocharaceans at the base to shallow marine settings at the top.

**Interval F** (450–490 m) belongs to the Artoles Formation at the base and to the Morella Formation at the top (Fig. 3B). The lower 28 m (450–478 m) are composed of >2 m-thick marls interbedded with 0.5–1 m-thick limestones. The palaeontological assemblage found in the marls includes abundant charophyte thalli and clavatoracean fructifications, ostracods, and molluscs. The limestones contain ostracods, gastropods, bivalves, dasycladaleans, benthic foraminifera, and rare clavatoracean fructifications and thalli. The top of the limestones is characterised by a ferruginous hardground surface, locally encrusted by oysters. The upper 12 m (478–490 m) of the interval shows a very distinct facies succession, characterised by repetitive interbedded grey and red clays. These clays contain abundant and diverse charophytes associated with ostracods, gastropods, and lituolid foraminifera. The lithofacies have strong similarities with those described by Gàmez et al. (2003) for the Morella Formation in the depocentre of the Maestrat Basin (Salzedella Sub-basin; Fig. 1B). Consequently, these clays are ascribed to the Morella Formation. The depositional setting of Interval F is interpreted to have evolved from marginal coastal marine settings in the lower 28 m and from brackish non-marine environments in the upper 12 m.

The charophyte assemblages are rich in *E. lazarii*. Less abundant charophytes include *G. maillardii* var. *trochiliscoides*, and var. *biutricularis*, *A. trivolvis* var.
*triquetra*, *C. harrisii* var. *reyi*, *Ascidiella cruciata* (first occurrence), *H. neimongolensis* var. *neimongolensis*, *Pseudoglobator paucibracteatus* (first occurrence), aff. *M. harrisii*, and *P. maestratica*. Fertile whorls of *E. lazarii* are frequently preserved, and two utricles were found anatomically attached the corresponding thalli, i.e., *Charaxis spicatus* Martin Closas et Diéguez, suggesting autochthony. The other fructifications of the assemblage (*G. maillardii* var. *trochiliscoides* and var. *biutricularis*, *A. trivolvis* var. *triquetra*, *C. harrisii* var. *harrisii* and var. *reyi*, *A. cruciata*, *H. neimongolensis* var. *neimongolensis*, *P. paucibracteatus*, aff. *M. harrisii* and *P. maestratica*) are well-preserved or slightly eroded, which suggests paraautochthony.

--- Please insert Fig. 3B near here ---

**Interval G** (490–611 m) corresponds to the upper part of the Artoles Formation in the Herbers-Mas de Petxí section (Fig. 3B). It mainly consists of marls alternating with cross-bedded packstone-grainstone limestones. The marls frequently contain bivalve moulds. The limestones are lenticular and show a lateral thinning from 2 m in the central parts to 0.3 m at the edges of the bed. The bioclastic components of the limestones mainly correspond to oysters, gastropods, echinoids, dasycladales, and benthic foraminifera. At the base, the limestone beds contain abundant mud pebbles. The occurrence of cross-bedding, lateral thinning of the beds, and the presence of intraclasts at the base, indicates that these limestones correspond to channel-fill deposits formed in shallow marine settings (e.g., Tucker 1985, Bover-Arnal and Strasser 2013). Above the channelised beds, a 7 m-thick laminated mudstone limestone with thin calcified filaments occurs. The laminas of this mudstone are a millimetre-thick and contain benthic foraminifera and very small mollusc bioclasts. This deposit represents a microbial algal mat formed in intertidal to shallow subtidal areas, resembling the one
reported by Tucker (1985) and Bover-Arnal and Strasser (2013). Thus, the interval is attributed to a deposition in a tidal flat.

The charophyte assemblages of this interval are mainly composed of well-preserved utricles of *Echinochara lasarrii*, which are considered autochthonous. Small populations of *G. maillardii var. biutricularis* and *A. cruciata* are well-preserved to slightly eroded, suggesting parautochthony.

**Interval H** (611–680 m) is ascribed to the Morella Formation, being the second interval attributed to this formation in Herbers-Mas de Petxí (Fig. 3B). It starts with a 1 m-thick red clay interval followed by a 1.5 m-thick charophyte-rich limestone. The following 37 m are characterised by up to 5 m-thick layers of variegated clays (grey, red, ochre, and purple) that contain abundant charophyte remains (thalli and fructifications), ostracods, and agglutinated benthic foraminifera. Red and purple clays show mottled areas and contain abundant root marks. Thin (3–20 cm-thick) wackestone-packstones occur, interbedded with the clays. These limestones are rich in charophyte utricles and thalli, found sometimes anatomically connected, as well as ostracods and molluscs. The uppermost 30.5 m of the interval (649.5–680 m) are composed of a succession of grey and red clays interbedded with packstones that are rich in oysters and ostracods and frequently exhibit bioturbated tops. The microfossil content of the clays resembles that of the basal part of the interval, mainly containing charophyte fructifications and thalli, ostracods, and agglutinated benthic foraminifera. The top of this stratigraphic interval is marked by a 2 m-thick, cross-bedded and bioturbated packstone limestone rich in bivalve fragments and ostracods, similar to the channel-fill deposits observed in the underlying interval.

Similar clay-dominated deposits occur in the type locality of the Morella Formation, where they are associated with conglomerates and sands, thus being interpreted as deposited in a mudflat environment (Gàmez et al. 2003). In the studied
section, lacustrine limestones occur interbedded with clays and sandstone, unlike in the type locality. These limestones may correspond to temporary lakes or ponds that were formed in poorly-drained areas of mudflats, such as those reported by Bádenas et al. (2018) in the Tithonian–Berriasian record from the Galve Sub-basin (Fig. 1B). In the upper part of the interval (649.5–680 m), the clay deposits change upwards into channel-fill ostracod-rich limestones that mark the beginning of a long-term late Barremian–early Aptian transgression (Bover-Arnal et al. 2016). This interval represents the last non-marine deposits in the studied section.

The charophyte assemblage is very similar throughout the interval and strongly resembles that observed in Interval F. In this regard, Atopochara trivolvis var. trivolvis, Clavator grovesii var. jiuquanensis, and C. harrisii var. harrisii appear in this interval, but are absent in Interval F. Porochara maestratica is found in Interval F but is absent in interval H. E. lazarii, A. trivolvis var. triquetra and var. trivolvis, C. harrisii var. reyi and var. harrisii are abundant, well-preserved and occasionally attached to the thalli portions, suggesting autochthony. The utricles of G. maillardii var. trochiliscoides and var. biutricularis, Clavator grovesii var. jiuquanensis, A. cruciata, H. neimongolensis var. neimongolensis, P. paucibracteatus, and gyrogonites of aff. M. harrisii are less abundant and occasionally slightly abraded, indicating short lateral transport and paraautochthony. Benthic foraminifera are abundant and well-preserved, indicating a marine influence, which is commonly described for mudflat settings (c.f. Bádenas et al. 2018).

5.2. The Fredes section

The Fredes section is exposed between kms 8 and 9 of the road CV-106 (Fig. 1D) and consists of a ~100 m-thick continuous Barremian succession that belongs entirely to the Cantaperdus Formation. It lies on the Herbers Formation, Hauterivian in age (e.g.,
Salas et al. 2001). The basal laterite of the Cantaperdius Formation consists of ~2.5 m of yellowish and red clays that are topped by a mudstone limestone with root marks. The charophyte assemblage in the laterite is mainly composed of *A. trivolvis* var. *triquetra*, *A. stellata* var. *stellata*, *H. neimongolensis* var. *posticecaptus*, and gyrogonites of *P. maestratica* and aff. *M. harrisii*. Thalli portions belonging to *Clavatoraxis* sp., *Favargerella aquavivae* and *Munieria grambastii* are rare, suggesting a parautochthonous assemblage. Above the laterite, the section is divided into five descriptive intervals A–E (Fig. 4). The laterite and the lower 10 m of the section are repeated as the section is faulted.

**Interval A** (2.5–10.25 m and 13–26 m) is characterised by the occurrence of lenticular limestone beds interbedded with marls. The marls form up to 1 m-thick beds that are rich in charophyte thalli and fructifications, ostracods, molluscs and, frequently, agglutinated benthic foraminifera, dasycladaleans, and vertebrate teeth. The limestones are up to 3 m-thick (most commonly 1.5 m-thick). Their base is erosive and characterised by lag deposits with a packstone texture containing molluscs, ostracods, intraclasts, and rare charophyte thalli. Above the lag, the limestone is a mollusc-rich wackestone-mudstone, forming a thinning-upwards sequence. Root marks as well as nodular and intraclastic fabrics are observed at the top of the limestones.

The marls are interpreted as deposited in shallow lakes. The co-occurrence of freshwater and brackish water organisms suggests that these lakes were probably established in a coastal area that experienced alternating periods of freshwater and marine influences. The overlying lenticular limestone beds are very similar to other carbonate channel-fill deposits associated with lacustrine and palustrine areas (e.g., Platt 1989, Alonso-Zarza and Calvo 2000). The occurrence of intraclasts and the similarity of the fossil contents of the limestones and the underlying marl intervals suggest the reworking of the previously deposited shallow lake deposits during the sedimentation of
the channel-fill deposits. Nodular textures and root marks at the top of these lenticular beds indicate the abandonment of the channel and the establishment of palustrine conditions (see, e.g., Alonso-Zarza and Wright 2010).

The charophyte assemblage found in the marls is mainly composed of the utricles of *A. trivolvis* var. *triquetra*, *A. stellata* var. *stellata* and var. *lata* (first occurrence), *A. triquetra* and *H. neimongolensis* var. *posticecaptus*. These utricles are well-preserved and frequently occur associated with or attached to thalli (*Clavatoraxis* sp., *F. aquavivae* and *M. grambastii*), suggesting autochthony. On the other hand, *G. maillardii* var. *trochiliscoides*, *C. harrisii* var. *dongjingensis*, *C. harrisii* var. *harrisii*, aff. *M. harrisii* and *P. maestratica* are rare and frequently abraded, and found associated with well-preserved dasycladalean and benthic foraminifera. This latter charophyte assemblage is considered paraautochthonous in the sample.

**Interval B** (26–40.75 m) is formed of two different parts. The lower part (26–36 m) is characterised by decimetre-thick limestone beds, frequently showing vertical root marks and nodular fabrics at the top, which are interbedded with thinner layers of marl, all of them rich in charophyte thalli and fructifications as well as ostracods. The upper part of the interval (35–40.75 m) starts with a thin marl bed containing abundant charophyte remains (utricles, gyrogonites and thalli) together with abundant agglutinated benthic foraminifera, molluscs and echinoid fragments. Above the marl layer, the interval consists of repeating limestone parasequences composed of a basal massive up to 2 m-thick wackestone that contains porocharacean gyrogonites, benthic foraminifera, echinoid fragments, and rare clavatoracean remains, followed by a 0.2 m-thick nodular limestone.

The lower part of the interval (26–35 m) is interpreted as a deposition in freshwater shallow lakes that were later subaerially-exposed and, thus, the original rock texture was modified by the establishment of palustrine conditions (see, e.g., Freytet
and Plaziat 1982, Alonso-Zarza and Wright 2010). The upper interval (35–40.75 m) was probably deposited in coastal environments, with changing salinity and frequent desiccation and emersion, as demonstrated by the presence of nodular limestones (see similar examples in Alonso-Zarza and Wright 2010).

The charophyte assemblage identified in the marls of this interval resembles that described in the previous interval, but with a slightly different preservation. The utricles of *G. maillardii* var. *trochiliscoides* and, locally, *G. maillardii* var. *biutricularis* (first occurrence) and *A. trivolvis* var. *triquetra* are abundant and well-preserved (indicating autochthony), while those of *C. harrisii* var. *harrisii*, *A. stellata* var. *stellata* and var. *lata*, *A. triqueta*, *H. neimongolensis* var. *posticecaptus*, aff. *M. harrisii* and *P. maestratica* are scarce and can be abraded. The benthic foraminifera associated with the charophytes are also abundant and well-preserved, suggesting a process of time averaging between the deposition of the two different assemblages (freshwater and the brackish to marine water, respectively).

---Please insert Fig. 4 near here---

**Interval C** (40.75–71.5 m) is characterised by marl beds interbedded with limestones. The marls are up to 2.5 m-thick and contain charophyte thalli and fructifications (utricles and gyrogonites), ostracods, mollusc and echinoid fragments, agglutinated benthic foraminifera, dasycladales and vertebrate teeth. The limestones are 0.25 to 1.5 m-thick and frequently display rootlet marks or nodular fabrics at the top. There are three types of deposits: (1) massive clavatoracean-rich limestones containing abundant clavatoracean utricles and thalli, ostracods, and molluscs, (2) massive limestones rich in benthic foraminifera and fragments of molluscs, dasycladales, and echinoids, and (3) lenticular cross-bedded bioclastic limestones with molluscs,
dasycladaleans, and benthic foraminifera that exhibit a basal lag deposit and become more fine upwards into a wackestone containing the same fossil content. This interval is interpreted as coastal-marginal settings with alternating freshwater and brackish to marine water influences. Root marks and nodular fabrics observed at the top of some of the limestone beds indicate palustrine settings with subaerial exposition (e.g., Freytet and Plaziat 1982, Alonso-Zarza and Wright 2010). The lenticular cross-bedded bioclastic limestones observed at the top of the interval correspond to channel-fill deposits, probably formed in a coastal marine-influenced area.

The charophyte assemblages in this interval are mainly composed of well-preserved utricles of *G. maillardii* var. *trochiliscoides* (rarely *G. maillardii* var. *biutricularis*) and *A. trivolvis* var. *triquetra*. Occasionally, *E. lazarii* (first occurrence), *C. harrisi* var. *dongjingensis*, var. *harrisi*, and var *reyi* (first occurrence), *A. stellata* var. *stellata* (last occurrence), and var. *lata*, *A. triqueta* and *H. neimongolensis* var. *posticecaptus* (last occurrence) and var. *neimongolensis* (first occurrence), aff. *M. harrisi*, and *P. maestratica* do also occur. As in the underlying intervals A and B, samples with a higher abundance and better preservation of charophyte remains contain scarce, abraded and/or fragmented remains of marine organisms, while those with a higher abundance of marine organisms contain scarce and frequently abraded charophyte remains.

**Interval D** (71.5–88 m) is composed of thick limestone beds that are characterised by the frequent presence of oncoids and intraclasts. The marl layers interbedded with these limestones are very thin and contain charophyte thalli and fructifications, ostracods, molluscs and, occasionally, vertebrate teeth, dasycladaleas, and miliolids. The base of the interval (71.5–82 m) consists of two 5 m-thick parasequences, the bottom of which displays two up to 1 m-thick marl beds that are separated by a 0.5 m-thick charophyte-rich packstone limestone. Above this, each parasequence continues
with a 3 m-thick massive or laminated wackestone limestone containing large (~1 cm in
diameter) black pebbles and oncoids. The upper part of Interval D (82–88 m) is
distinguished by the occurrence of limestone beds with a massive fabric at the base that
becomes nodular to brecciated at the top. These limestones are interbedded with thin
layers of marl.

The whole interval is interpreted as a shallow lacustrine to palustrine succession.

Black pebbles suggest more hydrodynamic lakeshore areas (e.g., Gierlowski-Kordesch
2010). The interbedding of nodular and brecciated limestones indicates palustrine facies
(e.g., Alonso-Zarza and Wright 2010), while the massive limestones correspond to well-
oxygenated lacustrine deposits (e.g., Gierlowski-Kordesch 2010). The absence or
scarcity of remains from marine organisms indicates a deposition in freshwater settings.

The charophyte assemblage in this interval is mainly formed of well-preserved *A.
trivolvis* var. *triquetra* and, occasionally, superficially eroded specimens of *G.
maillardii* var. *trochiliscoides* and var. *biatricularis, A. stellata* var. *lata, A. triquetra, H.
neimongolensis* var. *neimongolensis*, aff. *M. harrisii* and *P. maestratica*. *E. lazarii* is
rare, but showing complete fertile whorls. In the uppermost part of the interval (82–88
m), charophyte biodiversity increases and the assemblages are composed of well-
preserved fructifications of the aforementioned taxa together with those of *Clavator
grovesii* var. *gautieri* and *C. harrisii* var. *reyi* that are associated with thalli (*F.
aquavivae, Clavatoraxis* sp., and *Charaxis* aff. *spicatus*), sometimes in anatomical
connection, indicating an autochthonous assemblage.

-----------------------------Please insert Fig. 5 near here-----------------------------

**Interval E** (88–98 m) is dominated by massive (locally nodular) clavatoracean-
rich marl beds that are up to 1.7 m-thick and contain ostracods, and molluscs. These are
interbedded with up to 1 m-thick limestones. Based on textures and fossils, two
microfacies can be distinguished: (1) oyster-rich grainstones that also contain
gastropods and benthic foraminifera, and (2) wackestone limestones rich in
porocharacean gyrogonites that also contain less-abundant molluscs and benthic
foraminifera. The whole interval is interpreted as coastal shallow lakes, laterally-
associated with brackish water and marine environments.

This interval contains richer charophyte assemblages than those observed in the
underlying interval, including *E. lazarii*, *G. maillardii* var. *trochiliscoides*, and var.
*biutricularis*, *A. trivolvis* var. *triquetra*, *C. grovesii* var. *gauteri*, *C. harrisii* var.
dongjingensis (last occurrence), var. *harrisii*, and var. *reyi*, *Clavator calcitrapus* var.
*jiangluoensis*, and var. *calcitrapus* (first occurrence), *A. stellata* var. *lata*, *A. triquetra,*
*H. neimongolensis* var. *neimongolensis*, and rarely also *P. maestratica* and aff. *M. harrsii*. The utricles and gyrogonites are generally well-preserved and associated with
charophyte thalli (*Charaxis* sp., *Clavatoraxis* sp., *F. aquavivae*, and *M. grambastii*),
suggesting autochthony of the charophyte assemblage.

**6. Strontium-Isotope Stratigraphy (SIS)**

**6.1. Preservation of the original $^{87}\text{Sr}/^{86}\text{Sr}$ signature**

Evaluation of the preservation state of low-Mg calcite shells is a primary
requirement in SIS. Diagenetic processes during burial may modify the original
$^{87}\text{Sr}/^{86}\text{Sr}$ ratio, resulting in an erroneous derivation of the age. The analysis of major and
trace elements is an effective tool to evaluate any changes in biogenic samples (e.g.,
biogenic samples commonly result in a decrease in Sr concentration and an increase in
Fe and Mn concentrations as well as in the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio (Brand and Veizer 1980,
Wenzel 2000). However, these trends were not observed in our results, which are summarised in Table 2. Except for sample OyFr1, which is clearly altered due to diagenesis, the Sr concentrations are >700 ppm (Table 2). A Sr concentration > 700 ppm is usually considered a minimum value for discriminating between non-diagenetically-altered samples and diagenetically-altered samples (e.g., Boix et al., 2011; Frijia et al., 2015). Moreover, in most samples, the Mn and Fe concentrations are above the threshold values (50 ppm and 250 ppm, respectively) that discern between diagenetically-altered and non-diagenetically-altered samples (e.g., Steuber 1999, 2001). However, high Fe and Mn concentrations have also been linked to sedimentary processes. Schneider et al. (2009) indicated that in marginal marine conditions, Fe and Mn concentrations can be affected by freshwater influx and suggested cut-off values of Mn <250 ppm and Fe <700 ppm for oysters found in such settings. On the other hand, low Mn and Fe concentrations can also occur in diagenetic calcite (Steuber et al. 2005, Boix et al. 2011, Frijia et al. 2015, Bover-Arnal et al. 2016, Caus et al. 2016). Therefore, low Mn and Fe concentrations and high Sr concentrations have to be used cautiously as indicators of non-diagenetic overprint of biogenic low-Mg calcite. In our dataset, 6 out of the 10 samples (OyFr2, OyFr21c, OyH3, OyH4, OyH6, and OyH7; Table 2) have Mn and Fe concentrations that are below the threshold values (Mn < 250 ppm and Fe < 700 ppm) distinguishing between diagenetically-altered and non-diagenetically-altered oyster shells from marginal marine environments (e.g., Schneider et al., 2009; Horikx et al., 2014). Following Brasier et al. (1994), Denison et al. (1994) and Rosales et al. (2001), we used the Mn/Sr and Fe/Sr ratios to discern between diagenetically-altered and non-diagenetically-modified samples (Table 2). Ratios of Mn/Sr <0.5 and Fe/Sr <0.3 were proposed by Brasier et al. (1994) to distinguish between altered and non-altered carbonates. Based on this criterion, only
three samples, OyFr21c, OyH4 and OyH6 (Table 2), preserved non-altered $^{87}\text{Sr}/^{86}\text{Sr}$ values and were finally used to derive the numerical ages.

The discarded samples OyH3 and OyH7 show Sr-isotope ratios of 0.707515 and 0.707504, respectively, which are not in agreement with the marine $^{87}\text{Sr}/^{86}\text{Sr}$ ratios published for the Barremian Stage. According to McArthur et al. (2012), the Barremian Sr-isotope values range between 0.707471 and 0.707432, whereas McArthur et al. (2020) give a range between 0.707470 and 0.707426. The oyster shells OyFr2, OyH3 and OyH7 discarded were collected from beds that contain abundant and well-preserved charophytes (Figs. 3B and 4). Therefore, the isotopic signal of these samples was probably modified by freshwater influence (see e.g., Benito et al. 2020; Table 2).

6.2. Strontium ratios and the derived numerical ages

Non-diagenetically-altered samples used for SIS were collected from the Cantaperdius Formation (sample OyFr21c) and the Artoles Formation (samples OyH4 and OyH6) (Figs. 3A-B and 4; Table 2). The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios obtained from the low-Mg calcite oyster shells from these units vary from 0.707481 ± 0.000012 to 0.707489 ± 0.000012 (Table 2). Taking into account the stratigraphic context, these values were converted into numerical ages using the ‘LOWESS 5 fit 26 03 13’ look-up table of McArthur, which is linked to the Geologic Time Scale 2012 (GTS2012) of Gradstein et al. (2012), and the ‘LOESS 6 16 03 2020’ look-up table of McArthur (J. McArthur, personal communication, 2021), which is linked to the Geologic Time Scale 2020 (GTS2020) of Gradstein et al. (2020). In both Geologic Time Scales, the whole-time range of the samples is latest Hauterivian–late Barremian (Table 2). The reason why these $^{87}\text{Sr}/^{86}\text{Sr}$ values translate into such a wide time span is due to the slow increase in the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios between the early Barremian and the early late Barremian (see, e.g., Jones and Jenkins 2001, McArthur et al. 2001, 2012, 2020, Mutterlose et al. 2014, Wan
et al. 2019). However, the data available on charophyte biostratigraphy at the base of the studied sections constrains the age of these samples to the Barremian (e.g., Martín-Closas and Salas 1994, Martín-Closas 2000).

According to the numerical ages derived from the ‘LOWESS 5 fit 26 03 13’ look-up table of McArthur, which is linked to the GTS2012, the $^{87}\text{Sr} / ^{86}\text{Sr}$ ratios indicate preferred ages of between 129.80 and 129.00 Ma, dating the analysed specimens to the early Barremian–early late Barremian. Considering the statistical uncertainty (2 s.e.), the total age range for the samples is 130.95–128.25 Ma (latest Hauterivian–late Barremian).

The oldest low-Mg calcite sample was obtained from the Cantaperdius Formation in the Fredes section (sample OyFr21c; Fig. 4). The $^{87}\text{Sr} / ^{86}\text{Sr}$ ratio of this sample is $0.707481 \pm 0.000012$, translating into a preferred numerical age of 129.80 Ma (+1.15/-0.60). This numerical age corresponds to the late early Barremian (Ogg and Hinnov, 2012). The two other analysed samples were obtained from the Herbers-Mas de Petxí section (Fig. 3A–B). Sample OyH4 was obtained from the lower part of the Artoles Formation and its $^{87}\text{Sr} / ^{86}\text{Sr}$ ratio is $0.707482 \pm 0.000012$, which translates into a preferred age of 129.70 Ma (+1.20/-0.60), i.e., early Barremian (Ogg and Hinnov, 2012). The youngest low-Mg calcite shell sample was OyH6, which was obtained from the topmost part of the Artoles Formation, specifically, 1.5 m below the second interval of the Morella Formation in the Herbers-Mas de Petxí section (Fig. 3B). The $^{87}\text{Sr} / ^{86}\text{Sr}$ value for this sample is $0.707489 \pm 0.000012$, which translates into a preferred age of 129.00 Ma (+0.80/-0.75), corresponding to the early late Barremian (Ogg and Hinnov, 2012).

On the other hand, utilizing the ‘LOESS 6 16 03 2020’ look-up table of McArthur, which is associated to the GTS2020, the preferred numerical ages fall within the early Barremian and range between 126.10 and 125.05 Ma (Table 2). Taking into
account the statistical uncertainty, however, the total time span for the sample is 126.65–124.20 Ma (latest Hauterivian–early late Barremian according to Gale et al. 2020). However, this recent numerical calibration shows inconsistencies. For example, according to the ‘LOESS 6 16 03 2020’, the preferred numerical age derived for sample OyH6 is 125.05 Ma (Table 2) and falls within the early early Barremian. Sample OyH6 is 24 m below the first appearance of Clavator grovesii var. jiuquanensis (Fig. 3B), which has been correlated with the Toxanclyoceras vandenheckii ammonoid biozone (late Barremian) in the Subalpine Chains (Martín-Closas et al. 2009). According to Gale et al. (2020), the base and top of the Toxanclyoceras vandenbeckii Zone is dated to 124.4 and 123 Ma, respectively. This would imply that the 24 m of stratigraphic succession found between sample OyH6 and the the first occurrence of Clavator grovesii var. jiuquanensis (Fig. 3B) would record 0.65 My. Such a significant sedimentary gap or slow rate of sedimentation seems unlikely given the overall expansion and large thickness of the synrift succession studied (Fig. 3A-B). In consequence, the biozonations presented herein follow the GTS2012, which provides a more consistent chrono-biostratigraphic framework (Fig. 6), and is in agreement with previous correlations between charophyte assemblages and ammonoid biostratigraphy (Martín-Closas et al. 2009).

---Please insert Table 2 near here---

### 7. Barremian-early Aptian charophyte biozonation

The new data obtained from the two Barremian sections studied enabled us to refine the Barremian-early Aptian European charophyte biozonation and extend its use to most of the Cretaceous Tethyan Archipelago (mainly present-day Europe and North Africa). Moreover, the new data led us to propose a new Barremian-early Aptian...
Eurasian charophyte biozonation, whose index species were widely distributed from Europe to China. The two biozonations are directly correlated to one another in Figures 3A–B, 4 and 6.

7.1. The Barremian–early Aptian European charophyte biozonation

The first proposal of a Barremian–early Aptian European charophyte biozonation was from Grambast (1974) and was later modified by Martín-Closas and Salas (1994), Riveline et al. (1996), and finally by Martín-Closas et al. (2009). The current biozonation (after the revision of Martín-Closas et al., 2009) is composed of two partial range biozones that are characterised at their bases by the FAD of *Atopochara trivolvis* var. *triquetra* (early Barremian) and *Ascidiella cruciata* and *Pseudoglobator paucibracteatus* (late Barremian–early Aptian; Fig. 6), respectively. Herein, we propose to modify this biozonation by only using species that are known to have been endemic in the Cretaceous Tethyan Archipelago. Following this criterion, *Globator maillardii* var. *trochiliscoides* is proposed here as the index species of the early Barremian charophyte biozone instead of *A. trivolvis* var. *triquetra*. This new index species was extensively distributed and appears to have been endemic in Europe and most of the Cretaceous Tethyan Archipelago (Martín-Closas, 2000; Martín-Closas and Wang 2010; Pérez-Cano et al., 2020; Sanjuan et al. 2021), while *A. trivolvis* var. *triquetra*, which had a cosmopolitan distribution during the Barremian (Martín-Closas and Wang, 2008), is reserved for the Eurasian biozonation (see below). By contrast, the late Barremian–early Aptian *Ascidiella cruciata*-Pseudoglobator *paucibracteatus* biozone is kept unchanged in the new proposal (Fig. 6), since these index species are, so far, unique to the Cretaceous Tethyan Archipelago.

*Globator maillardii* var. *trochiliscoides* biozone
Definition. Partial range biozone defining the interval between the FAD of
Globator maillardii var. trochiliscoides and the FAD of Ascidiella cruciata and
Pseudoglobator paucibracteatus (Figs. 3A–B and 6).

Species assemblage: Echinochara lazarii, Globator maillardii var. trochiliscoides
(dominant over G. maillardii var. biutricularis), G. maillardii var. biutricularis,
Atopochara trivolvis var. triquetra, Clavator grovesii var. gautieri, C. harrisii var.
dongjingensis, C. harrisii var. harrisii (dominant over var. dongjingensis and var. reyi),
C. harrisii var. reyi, C. calcitrapus var. jiangluoensis, C. calcitrapus var. calcitrapus,
Ascidiella stellata var. stellata, A. stellata var. lata (dominant over var. stellata), A.
triquetra, A. iberica var. iberica, Hemiclavator adnatus, H. neimongolensis var.
posticecaptus, H. neimongolensis var. neimongolensis, and Pseudoglobator fourcadei.

Remarks. A biozone based on G. maillardii var. trochiliscoides was previously
proposed by Mojon (1996) to characterise the late Barremian–early Aptian. The same
author proposed the G. maillardii var. mutabilis biozone for the lower Barremian.
However, the populations of G. maillardii observed in the oldest assemblages studied
herein (in the lower part of the Cantaperdius Formation) do not show the basal utricle
features characteristic of G. maillardii var. mutabilis (cf. Martin-Closas 2000), fully
belonging to var. trochiliscoides instead. This criterion of homogeneous populations of
G. maillardii var. trochiliscoides was used to establish the base of the G. maillardii var.
trochiliscoides biozone. However, some utricles (<10%) of G. maillardii var.
birutricularis may occur in particular G. maillardii var. trochiliscoides populations of
this biozone, while G. maillardii var. biutricularis can be more frequent than G.
maillardii var. trochiliscoides or even dominant in the overlying Ascidiella cruciata-
Pseudoglobator paucibracteatus biozone, where it can form homogeneous populations
(e.g., Vicente and Martin-Closas 2013).
The *C. harrisii* populations in the *G. maillardii* var. *trochiliscoides* biozone are dominated by *C. harrisii* var. *harrisii*, which is the typical variety in this biozone and frequently forms homogeneous populations (Martín-Closas 2000 and references therein). However, some utricles of *C. harrisii* var. *dongjingensis* and *C. harrisii* var. *reyi* are repeatedly found in the studied samples to be in association with *C. harrisii* var. *harrisi*. *C. harrisii* var. *dongjingensis* was typical and dominant between the late Berriasian and late Hauterivian (Martín-Closas 2000). *C. harrisii* var. *reyi* occurs for the first time in this biozone inside populations dominated by *C. harrisii* var. *harrisi*.

However, *C. harrisii* var. *reyi* is typical and dominant in the overlying *C. ruciata*- *Paucibracteatus* biozone, where it forms homogeneous populations (e.g., Vicente and Martín-Closas 2013).

*C. calcitrapus* var. *jiangluoensis* occurs in the lower part of the *G. maillardii* var. *trochiliscoides* biozone, while *C. calcitrapus* s.s. occurs in the upper part of the biozone. Therefore, the gradualistic evolution of *C. calcitrapus* described by Pérez-Cano et al. (2020) allows more precise dating throughout this biozone.

Two more species are also characteristic of this biozone, although they have not been identified in the stratigraphic sections studied here. These are *Ascidiella iberica* var. *iberica* (Grambast) Martín-Closas ex Schudack, which occurs in association with *A. trivolvis* var. *triquetra* in several localities of the Iberian Chain in Spain and in the Wealden Basin in England (Martín-Closas 2000 and Feist et al. 1995, respectively), and *Pseudoglobator fourcadei* Grambast, which has been exclusively described in the Prebaetic Chain (south-eastern Spain), where it occurs in association with *A. trivolvis* var. *triquetra* and *H. adnatus* (Martín-Closas 2000).

**Biostratigraphic correlations.** In the Iberian Chain, *G. maillardii* var. *trochiliscoides* has been correlated with the orbitolinid foraminifer *Palaeodictyoconus cuvillieri* (Combes et al., 1966). In the Subalpine Chains, *G. maillardii* var.
*trochiliscoides* has been correlated with the orbitolinid assemblage composed of *Valserina broennimanni*, *Eopalorbitolina charollaisi*, *Orbitolinopsis debelmasi*, *Cribellopsis elongata* and *Paracoskinolina hispanica*, corresponding to the *Nicklesia nicklesi* ammonite Zone (Martín-Closas et al. 2009).

**Calibration based on SIS.** No calibration is available for the base of the biozone, while its top coincides with the base of the next biozone, which is calibrated as explained below.

**Age.** Early Barremian.

**Ascidiella cruciata-Pseudoglobator paucibracteatus biozone**

**Definition.** Time interval defined between the FAD of *Ascidiella cruciata* and *Pseudoglobator paucibracteatus*, and the FAD of *Clavator grovesii* var. *lusitanicus*.

**Species assemblage.** *Echinochara lazarii*, *Globator mallardi* var. *trochiliscoides*, *G. maillardii* var. *biutricularis* (dominant over var. *trochiliscoides*), *Atopochara trivolvis* var. *triquetra* (dominant variety upon var. *trivolvis*), *Atopochara trivolvis* var. *trivolvis*, *Clavator grovesii* var. *gautieri*, *C. grovesii* var. *jiuquanensis*, *C. harrisii* var. *harrisii*, *C. harrisii* var. *reyi* (dominant over var. *harrisii*), *Hemiclavator neimongolensis* var. *neimongolensis*, *Ascidiella cruciata*, *A. iberica* var. *inflata* and *Pseudoglobator paucibracteatus*.

**Remarks.** Riveline et al. (1996) defined this biozone as a partial range biozone described between the FAD of *A. cruciata* and the FAD of *C. grovesii* var. *lusitanicus*. Later, Martín-Closas et al. (2009) redefined this biozone as a total range biozone limited between the FAD of *A. cruciata* and *P. paucibracteatus* and the last appearance datum (LAD) of *P. paucibracteatus*. The redefinition as a partial range biozone in this paper follows the original characterisation of the late Barremian Ascidiella cruciata partial range biozone of Riveline et al. (1996) but using the FAD of *A. cruciata* and *P.*
paucibracteatus. This avoids possible problems of (1) overlapping between the
Ascidiella cruciata-Pseudoglobator paucibracteatus biozone and the overlying Clavator
grovesii var. lusitanicus biozone if the LAD of *P. paucibracteatus* is found to be
younger than the FAD of *C. grovesii var. lusitanicus* and (2) occurrence of an undefined
interval between the two biozones. Such poorly defined biostratigraphic intervals are
usually designed as interval biozones.

*A. trivolvis var. trivolvis* occurs in this biozone. However, it never forms
homogeneous populations but it is found associated with *A. trivolvis var. triquetra*,
which is the dominant variety in the Cruciatia-Paucibracteatus biozone. Homogeneous
populations of *A. trivolvis var. trivolvis* are typical of the overlying Clavator grovesii
var. corrugatus biozone (Riveline et al. 1996). *Ascidiella iberica var. inflata* (Grambast-
Fessard) Martín-Closas has not been identified in the studied sections. However,
Martin-Closas (2000) reported it from beds interbedded with *Palorbitolina lenticularis*
in other sections from the Maestrat basin.

**Biostratigraphic correlations.** In the Subalpine Chains, *P. paucibracteatus* has
been observed in a marl level above a grainstone bed containing *Paleodictyoconus
cuvillieri*, *Paleodictyoconus actinostoma* and *Cribrellopsis neoelongata* (Mojon, 1988).
Martin-Closas et al. (2009) found *P. paucibracteatus* in marl beds interbedded with
marine layers alongside an orbitolinid assemblage composed of *Orbitolinopsis kiliani*,
*O. cuvillieri*, *O. buccifer* and *Palorbitolina lenticularis*. These authors correlated this
assemblage with the Barremian ammonite biozones of *Toxancyloceras vandenheckii*,
*Gerhardtia sartousiana*, *Hemihoplites feraudianus* (now the upper subzone of
*Gerhardtia sartousiana* according to Reboulet et al. 2018), *Imerites giraudi* and
*Martelites sarasini*. In the Iberian Chain, *A. cruciata* has been repeatedly correlated
with *P. lenticularis* (Martin-Closas, 2000 and references therein).
Calibration based on SIS. In the Herbers-Mas de Petxí section, the FAD of *P. paucibracteatus* occurs in a marl bed at the basal part of the Artoles Formation (sample H124; Fig. 3B), which is located 5 m below the stratigraphic layer where sample OyH4 was collected (Fig. 3B). An $^{87}$Sr/$^{86}$Sr ratio of 0.707482 ± 0.000012 was obtained for this oyster shell, translating into a preferred age of c. 129.70 Ma (+1.20/-0.50) (Table 2) for the base of this biozone. No calibration is available yet for the top of the biozone.

**Age.** Late early Barremian–early Aptian.

7.2. The Barremian–early Aptian Eurasian charophyte biozonation

**Atopochara trivolvis var. triquetra biozone**

**Definition.** Time interval defined between the FAD of *Atopochara trivolvis* var. *triquetra* and the FAD of *Hemiclavator neimongolensis* var. *neimongolensis*.

**Species assemblage (Family Clavatoraceae) in the basins of the Cretaceous Tethyan Archipelago.** *Echinochara lazarii*, *Globator maillardii* var. *trochiliscoides* (dominant over *G. maillardii* var. *biutricularis*), *G. maillardii* var. *biutricularis*, *Clavator grovesii* var. *gautieri*, *Asciidiella stellata* var. *stellata*, *A. stellata* var. *lata* (dominant over *A. stellata* s.s.), *A. triquetra*, *A. iberica* var. *iberica*, *Hemiclavator adnatus*, *H. neimongolensis* var. *posticecaptus*, and *Pseudoglobator fourcadei*.

**Species assemblage (Family Clavatoraceae) in all the Eurasian basins.**

*Atopochara trivolvis* var. *triquetra*, *Clavator harrisi* var. *dongjingensis*, *C. harrisi* var. *harrisi* (dominant over var. *dongjingensis* and var. *reyi*) and *C. calcitrapus* var. *jianglouensis*.

**Remarks.** Riveline et al. (1996) characterised it as a partial range biozone that was defined between the FAD of *A. trivolvis* var. *triquetra* and the FAD of *Asciidiella cruciata* in the European biozonation. Due to the cosmopolitan distribution of *A. trivolvis* var. *triquetra* (Martin-Closas and Wang 2008), this biozone is redefined herein...
as a Eurasian biozone, with its upper boundary modified to the FAD of *H. neimongolensis* var. *neimongolensis*.

The FAD of *A. trivolvis* var. *triquetra* seems to occur at least in the late Hauterivian, where it appeared to be associated with *A. trivolvis* var. *ancora* (Fig. 3A). However, it is not until the base of the Barremian when *A. trivolvis* var. *triquetra* formed homogenous populations. This criterion, i.e., the occurrence of homogeneous populations of this anagenetic variety, was used here to define the base of the *A. trivolvis* var. *triquetra* biozone.

**Biostratigraphic correlations.** *A. trivolvis* var. *triquetra* frequently appears with *G. maillardii* var. *trochiliscoides* in the Iberian Chain (e.g., Combes et al. 1966, Martin-Closas and Salas 1994, Martin-Closas 2000) and in the Subalpine Chains (Martin-Closas et al. 2009). Thus, it has been correlated with the same orbitolinid species and ammonite biozones as those for the *G. maillardii* var. *trochiliscoides* biozone.

**Calibration based on SIS.** The upper boundary of this biozone was calibrated in the Fredes section with an $^{87}\text{Sr}/^{86}\text{Sr}$ ratio obtained from a low-Mg oyster shell belonging to sample OyFr21c (Fig. 4 and Table 2). The $^{87}\text{Sr}/^{86}\text{Sr}$ value is 0.707481 ± 0.000012, translating into a preferred numerical age of 129.80 Ma (+1.15/-0.60).

**Age.** Early Barremian.

**Hemiclavator neimongolensis var. neimongolensis biozone**

**Definition.** Partial range biozone defining the interval between the FAD of *Hemiclavator neimongolensis* var. *neimongolensis* and the FAD of *Clavator grovesii* var. *jiuquanensis*.

**Species assemblage (Family Clavatoraceae) in the basins of the Cretaceous Tethyan Archipelago.** *Echinochara lazarii, Globator mallardi* var. *trochiliscoides, G. maillardii* var. *biutricularis, Clavator grovesii* var. *combei, C. calcitrapus* var.
calcitrapus, Ascidiella stellata var. lata, A. triquetra, A. cruciata, A. iberica var. inflata, and Pseudoglobator paucibracteatus.

**Species assemblage (Family Clavatoraceae) in all the Eurasian basins.**

Atopocha trivolvis var. triquetra, Clavator harrisii var. dongjingensis, C. harrisii var. harrisii (dominant variety of this species), C. harrisii var. reyi, C. calcitrapus var. jiangluoensis, and Hemiclavator neimongolensis var. neimongolensis.

**Remarks.** The base of the biozone is marked by the first homogeneous population of *H. neimongolensis* var. *neimongolensis*. In the Herbers-Mas de Petxí section, mixed populations of *H. neimongolensis* var. *posticecaptus* and *H. neimongolensis* var. *neimongolensis* occur in the uppermost part of the *A. trivolvis* var. *triquetra* biozone (e.g., samples H36–H38; Fig 3A), while the first homogeneous population of *H. neimongolensis* var. *neimongolensis* occurs a few metres above (sample H39; Fig 3A).

The wide biogeographic range of *H. neimongolensis* var. *neimongolensis* and its quick expansion from Iberia to China (Martín-Closas 2015) make this species a useful tool to perform intercontinental correlations of non-marine successions.

Homogeneous populations of *C. calcitrapus* var. *jiangluoensis* can be found at the base of the Hemiclavator neimongolensis var. neimongolensis biozone, while homogenous populations of *C. calcitrapus* s.s. are found only in the upper part of the biozone. In fact, the *C. calcitrapus* populations observed in the H. neimongolensis var. neimongolensis biozone frequently contain both *C. calcitrapus* var. *jiangluoensis* and *C. calcitrapus* s.s., as well as intermediate morphotypes.

**Calibration based on SIS.** The FAD of the index species in the Fredes section occurs in sample Fr21c (Fig. 4) in association with oyster shells (sample OyFr21c; Table 2). The $^{87}\text{Sr}/^{86}\text{Sr}$ value obtained for this sample is $0.707481 \pm 0.000012$, which gives a preferred age of 129.80 Ma (+1.15/-0.60).

**Age.** Late early Barremian–early late Barremian.
**Clavator grovesii var. jiuquanensis biozone**

**Definition.** Partial range biozone comprising the interval between the FAD of *Clavator grovesii var. jiuquanensis* and the FAD of *Clavator grovesii var. corrugatus*.

**Species assemblage (Family Clavatoraceae) in the basins of the Cretaceous Tethyan Archipelago.** *Echinochara lazarii*, *Globator maillardii var. trochiliscoides*, *G. maillardii var. biurricularis* (dominant over var. *trochiliscoides*), *Ascidiella cruciata*, *A. iberica var. inflata*, and *Pseudoglobator paucibracteatus*.

**Species assemblage (Family Clavatoraceae) in all the Eurasian basins.**

*Atopochara trivolvis* var. *triquetra* (dominant over var. *trivolvis*), *A. trivolvis* var. *trivolvis*, *Clavator grovesii var. jiuquanensis*, *Clavator harrisii var. harrisii*, *C. harrisii var. reyi* (dominant over var. *harrisii*), and *Hemiclavator neimongolensis* var. *neimongolensis*.

**Remarks.** *C. grovesii var. jiuquanensis* was used by Wang and Lu (1982) to define the Perimneste ancora–Clypeator jiuquanensis assemblage biozone (former taxonomy for *Atopochara trivolvis* var. *ancora* and *C. grovesii var. jiuquanensis*, respectively) with a Hauterivian-lower Barremian age range in the Chinese basins. Similar ages were suggested for the *Mesochora stipitata-Clypeator jiuquanensis-Flabellochara hebeiensis* assemblage biozone (the latter a synonym of *C. harrisii* var. *harrisii*) defined by Peng et al. (2003). Yang et al. (2008) proposed a Clypeator jiuquanensis biozone that characterized the Hauterivian and the Barremian. However, recent data about the age of this taxon in China also suggest a late Barremian-early Aptian age (Li et al. 2020), similar to the ages observed in Europe.

The *Clavator grovesii var. jiuquanensis* biozone comprises most of the biostratigraphic interval of the European *Cruciata-Paucibracteatus* biozone of Martín-Closas et al. (2009). However, the Eurasian biogeographic range of *C. grovesii var.*
jiuquanensis and its quick spread throughout the entire Eurasian domain (Martín-Closas 2015) favours its use as a valuable index species to perform non-marine intercontinental correlations with.

**Biostratigraphic correlations.** In the Subalpine Chains, *C. grovesii* var. jiuquanensis occurs together with *P. paucibracteatus* (Martín-Closas et al. 2009), indicating that the *C. grovesii* var. jiuquanensis biozone can be correlated with benthic foraminifera and ammonite biozones similar to those used for the Cruciatapaucibracteatus biozone.

**Calibration based on SIS.** The FAD of the index species is 24.5 m above the oyster bed from where sample OyH6 was collected in the Herbers-Mas de Petxí section (Fig. 3B). The $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.707489 $\pm$ 0.000012 obtained with this sample translates into a preferred age of 129 Ma (+0.80/-0.75).

**Age.** Late Barremian–early Aptian.

---Please insert Fig. 6 near here---

8. Discussion

8.1. The Barremian–early Aptian charophyte biozonation revisited

Two different Barremian–early Aptian charophyte biozonations, one for the Cretaceous Tethyan Archipelago (European) and the other for the whole Eurasian domain, are proposed herein (Fig. 6). The European biozonation is formed of index species that are exclusive to Europe and North Africa, whereas the Eurasian biozonation is composed of sub-cosmopolitan to cosmopolitan index species based on the biogeographic studies of Martín-Closas and Wang (2008) and Martín-Closas (2015).

The new Barremian Eurasian biozonation proposed in this paper is composed of three biozones that characterise (1) the early Barremian (the Atopochara trivolvis var.
triquetra biozone), (2) the late early Barremian–early late Barremian (the Hemiclavator
neimongolensis var. neimongolensis biozone) and (3) the late Barremian–early Aptian
(the Clavator grovesii var. jiuquanensis biozone), respectively. This provides a better
biostratigraphic resolution than the European and Chinese charophyte biozonations used
to date (e.g., Riveline et al. 1996, Martin-Closas et al. 2009 and Wang and Lu 1982,
Yang et al. 2008, respectively). This is surprising since high-resolution charophyte
biozonations are often regional, basin-wide to continent-limited (e.g., Sanjuan et al.
2014, Vicente et al. 2015, 2016 for Europe with regional subzones; Li et al. 2019 for
North China), while sub-cosmopolitan to cosmopolitan species usually usually have
lower evolutionary rates, resulting in less precise biostratigraphic subdivisions. For
instance, Li et al. (2016) proposed a late Campanian–late Maastrichtian Eurasian
Microchara cristata biozone, which was later considered a superzone and subdivided
into three regional Chinese biozones (Li et al. 2019).

The Eurasian A. trivolvis var. triquetra biozone is equivalent to the biozone with
the same index species described by Riveline et al. (1996) and Martin-Closas et al.
(2009), except that its upper boundary has been modified to the FAD of H.
neimongolensis var. neimongolensis. The H. neimongolensis var. neimongolensis
biozone was originally proposed as a regional Iberian subzone for the upper part of the
Atopochara trivolvis var. triquetra biozone by Martin-Closas and Salas (1994) and
Martin-Closas and Schudack in Riveline et al. (1996). This subzone was first used to
resolve biostratigraphic correlations within Iberia (Martín-Closas and Alonso-Millán
1998), since the index species was unknown in other European basins. Later, it was
reported to be present in the Subalpine Chains and Jura Mountains in France and
Switzerland (Martín-Closas et al. 2009). It was already known to occur in China (Wang
and Lu 1982).
The Hemiclavator neimongolensis var. neimongolensis biozone directly correlates with the upper part of the Globator maillardii var. trochiliscoides European biozone and the base of the Ascidiella cruciata-Pseudoglobator paucibracteatus European biozone (Fig. 6), indicating that the charophyte assemblage of this biozone is composed of species from both European biozones (sections 7.1 and 7.2).

*Clavator grovesii* var. *jiuquanensis* is the index species of the third Eurasian biozone (late Barremian–early Aptian). This species was previously used as an index species to characterise a Hauterivian–early Barremian biozone in Chinese basins (Wang and Lu 1982, Peng et al. 2003, Yang et al. 2008). However, more recent data from the Jiuquan Basin suggest a late Barremian–early Aptian (more probably early Aptian) age for this species (Li et al. 2020), based on a previous chemostratigraphic ($\delta^{13}$C and $\delta^{18}$O) study performed by Suárez et al. (2013) and through comparisons with the age of *C. grovesii* var. *jiuquanensis* given by Martín-Closas et al. (2009) in Europe. The derived numerical ages obtained in the present study agree with the ages previously suggested for *C. grovesii* var. *jiuquanensis* in Eurasia.

### 8.2. Comparison with other Barremian charophyte biozonations

The European Barremian biozonations proposed to date followed and modified the pioneering proposal by Grambast (1974), who defined two assemblage biozones, ‘El Mangraner’ (early Barremian) and ‘San Carlos’ (late Barremian), which were obtained after studying the clavatoracean succession in the Eastern Iberian Chain. These biozones were the base for the European or more local charophyte biozonations, such as those by Feist et al. (1995) and Riveline et al. (1996), which distinguished an early Barremian biozone and a late Barremian biozone. The biozonation by Riveline et al. (1996) changed the type of biozone from assemblage zones to partial range zones. The charophyte assemblages observed in successive European biozonations and also in the
European biostratigraphic framework described herein are more diverse but include the assemblages described by Grambast (1974).

Mojon (1996, 2002) questioned the age of these assemblages, suggesting that they were younger (late Barremian and early Aptian, respectively, M8a and M8b) and, therefore, created a third biozone for the early Barremian (M7b) based on a newly defined morphotype, *Globator trochiliscoides* subsp. *mutabilis* (Fig. 6). However, this proposal was based on a misinterpretation of the Barremian units of the Iberian Chain, as discussed by Martín-Closas and Salas (1998). Therefore, the age of these biozones given by Riveline et al. (1996) is now supported by the calibration of the charophyte assemblages performed with SIS in the present study. As a consequence, the two European biozones proposed herein, i.e., the *Globator maillardii* var. *trochiliscoides* and the *Ascidiella cruciata*-Pseudoglobator paucibracteatus biozones, can be largely correlated with the equivalent biozones of Riveline et al. (1996) (Fig. 6).

Barremian charophyte biozonations have been proposed for two more regions, China and South America (Argentina). In China, two Barremian assemblage biozones were proposed by Wang and Lu (1982): the *Perimneste ancora*-Clypeator *jiuquanensis* and the *Atopochara trivolvis* triquetra-Flabellochara *hebeiensis* biozones. The age of these biozones was used as a reference for subsequent biostratigraphic proposals (e.g., Peng et al. 2003, Yang et al. 2008). With the exception of *C. grovesii* var. *jiuquanensis* (= *Clypeator jiuquanensis*), which is now considered to date to the late Barremian-early Aptian (also in China according to Li et al. 2020), the association described within the *Perimneste ancora*-Clypeator *jiuquanensis* biozone has been documented in Europe through the *Globator maillardii* var. *steinhauseri* European biozone (see Riveline et al. 1996), which dates to the latest Berriasian–late Hauterivian. By contrast, the clavatoracean assemblage described in the late Barremian Chinese *Atopochara trivolvis* triquetra-Flabellochara *hebeiensis* biozone (= *A. trivolvis* var. *triquetra* and *Clavator*...
harrisii var. harrisii, respectively) by Wang and Lu (1982) is comparable to the
European Globator maillardii var. trochiliscoides biozone. The Chinese Atopochara
trivolvis triquetra-Flabellochara heibeiensis biozone is also characterised by the
presence of H. neimongolensis var. neimongolensis, suggesting that this Chinese
biozone includes the Eurasian Atopochara trivolvis var. triquetra and Hemiclavator
neimonglensis var. neimongolensis biozones proposed herein.

In Argentina, a late Hauterivian–early Barremian Atopochara trivolvis triquetra
assemblage zone was proposed by Musacchio (1989, 2000). This biozone is
characterised by the association of A. trivolvis triquetra (= A. trivolvis var. triquetra)
with Triclypella patagonica Musacchio (synonym of Clavator calcitrapus var.
jiangluoensis, see Pérez-Cano et al. 2020). The association of A. trivolvis var. triquetra
with C. calcitrapus var. jiangluoensis is observed in the European G. maillardii var.
trochiliscoides biozone (section 7.1) and in the Eurasian A. trivolvis var. triquetra and
H. neimongolensis var. neimongolensis biozones (section 7.2), making the correlation
between South American and Eurasian basins possible. A late Barremian biozone has not
yet been characterised in South America. The Aptian South American Flabellochara
harrisii (= C. harrisii var. harrisii) biozone distinguished by Musacchio (1989, 2000) can
be partially correlated with the European Ascidiella cruciata-Pseudoglobator
paucibracteatus biozone and with the Eurasian Clavator grovesii var. jiuquanensis
biozone.

8.3. Correlations with the marine realm

The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and derived numerical ages can be used to correlate
charophyte assemblages with ammonite biozonations. This correlation was carried out
by taking into account the $^{87}\text{Sr}/^{86}\text{Sr}$ values reported by different studies for the Tethyan
and Boreal domains, as well as the standard $^{87}\text{Sr}/^{86}\text{Sr}$ curve found in the ‘LOWESS 5 fit
26 03 13’ look-up table of McArthur, which is tied to the numerical ages and ammonoid biozonation of the GTS2012 by Gradstein et al. (2012). However, the offset between the Boreal and Tethyan Barremian $^{87}\text{Sr}/^{86}\text{Sr}$ curves (see Mutterlose et al., 2014) makes the precise correlation between the Tethyan and Boreal domains difficult for this stage.

The stratigraphically lowest sample analysed for SIS (OyFr21c) coincides with the FAD of *H. neimongolensis var. neimongolensis* in the Fredes section (Fig. 4) and, thus, with the base of the homonymous Eurasian biozone. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of this sample, 0.707481 (see Table 2), is similar to the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (0.707478 to 0.707483) reported from southern France by Bodin et al. (2009) between the upper part of the *Nicklesia pulchella* and the lower part of the *Kotetishvilia compressissima* ammonoid biozones. According to the Sr isotope curve tied to the GTS2012 used, this $^{87}\text{Sr}/^{86}\text{Sr}$ ratio correlates precisely with the boundary between the *N. pulchella* and *K. compressissima* ammonoid biozones (Fig. 6). McArthur et al. (2004) reported $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of ~0.707476 to ~0.707485 for the early Barremian *Haplocrioceras fissicostatum* Zone of the Boreal region.

The stratigraphically upper samples for SIS were obtained from the Artoles Formation in the Herbers-Mas de Petxí section. Sample OyH4 was obtained 5 metres above the FAD of *P. paucibracteatus* with a Sr isotope value of 0.707482, and was used to date the base of the Ascidiella cruciata-Pseudoglobator paucibracteatus biozone. A similar value of 0.707483 has been described for the lower part of the *K. compressissima* Tethyan ammonite biozone in south-eastern France (Bodin et al. 2009).

In addition, plotting the Sr isotope value obtained from sample OyH4 against the standard Sr isotope curve from the GTS2012 revealed a correlation with the *K. compressissima* Zone. Hence, the Sr isotope data confirm the hypothesis of Martin-Closas et al. (2009), who indirectly correlated the base of the Cruciata-Paucibracteatus biozone with the middle part of the *Holcodiscus caullaudianus* ammonite biozone (late
early Barremian). This ammonite biozone was relegated to a subzone inside the *K. compressissima* Zone in the most recent proposal of ammonite zonation for the West Mediterranean Province of the Tethyan Realm (Reboulet et al. 2018). The Sr isotope value obtained with sample OyH4 coincides with those of the *Haplocriceras fiscostatum* ammonite biozone in the Boreal Realm (values from 0.707476 to 0.707485; see McArthur et al. 2004).

OyH6 was the youngest low-Mg sample used to derive the numerical ages in this study (Fig. 3B). The sample has an $^{87}$Sr/$^{86}$Sr value of 0.707489 (Table 2), which corresponds to a period from late early Barremian to early late Barremian (*sensu* Wan et al. 2019). This period is associated with stable $^{87}$Sr/$^{86}$Sr ratios (e.g., Jones and Jenkins, 2001, McArthur et al. 2012, Mutterlose et al. 2014), making it difficult to obtain a precise numerical age within this interval. The Sr isotope value of sample OyH6 falls within the range (0.707485-0.707493) obtained by McArthur et al. (2004) for the *Paracriconeras elegans* ammonite biozone in the Boreal Realm. Bodin et al. (2009) and Huck et al. (2011) reported similar mean $^{87}$Sr/$^{86}$Sr values for the Tethyan Realm as those found here for the *Moutoniceras moutonianum* ammonite biozone (latest early Barremian) and the base of the *Toxancyloceras vandenheckii* (earliest late Barremian) ammonite biozone. According to the Sr isotope curve tied to the GTS2012, the $^{87}$Sr/$^{86}$Sr value of sample OyH6 correlates with that of the early late Barremian Tethyan *Toxancyloceras vandenheckii* ammonite biozone (Fig. 6). However, sample OyH6 was obtained ~24.5 m below the FAD of *C. grovesii var. jiuquanensis* (Fig. 3B). Consequently, the base of the homonymous biozone may be younger than the numerical age obtained for this sample.

9. Conclusions
Compiled biogeographic and biostratigraphic data on clavatoracean species, with a particular focus on its succession recorded in the Maestrat Basin, established two distinct charophyte biozonations (European and Eurasian) spanning the Barremian-early Aptian period. The European charophyte biozonation is composed of two biozones whose index species were endemic in the Cretaceous Tethyan Archipelago. Its application is currently restricted to present-day Europe and North Africa. The biozones correspond to the Globator maillardii var. trochilicoideus (early Barremian) and Ascidiella cruciata-Pseudoglobator paucibracteatus (late early Barremian–early Aptian) biozones. The novel Eurasian biozonation proposed here includes the Atopochara trivolvis var. triqueta (early Barremian), Hemiclavator neimongolensis var. neimongolensis (late early Barremian–early late Barremian) and Clavator grovesii var. jiuquanensis (late Barremian–early Aptian) biozones. This new biostratigraphic proposal, whose index species are widely reported throughout Eurasia, aims to facilitate the correlation between distant continental basins (e.g., from China and Europe).

Correlation between the continental and marine realms carried out using $^{87}\text{Sr}/^{86}\text{Sr}$ values indicated that the age of the boundary between the Globator maillardii var. trochilicoideus and Ascidiella cruciata-Pseudoglobator paucibracteatus biozones of the European biozonation is roughly equivalent to that of the lower part of the Tethyan Kotetishvilia compressissima ammonoid biozone (c. 129.70 Ma). Regarding the Eurasian biozonation, the age of the base of the Hemiclavator neimongolensis var. neimongolensis biozone, which bounds the Atopochara trivolvis var. triqueta biozone below, correlates with that of the boundary between the Nicklesia pulchella and Kotetishvilia compressissima ammonite zones (c. 129.80 Ma). The base of the Clavator grovesii var. jiuquanensis biozone is constrained to the upper part of the Toxancyloceras vandenheckii ammonoid Zone (c. 129.00–128.63 Ma).
The five Barremian biozones described in this paper (including the European and Eurasian biostratigraphic frameworks) have been recognised in the stratigraphic succession studied in Herbers-Mas de Petxi. Therefore, we propose this sedimentary record cropping out in the northern Maestrat Basin to be the main reference section for Barremian charophyte biostratigraphy.

Acknowledgements

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Figure captions

Figure 1. A) Map of the main structural units of the Iberian Peninsula (modified from Salas et al. 2001). B) Palaeogeographical map of the Late Jurassic-Early Cretaceous syn-rift Maestrat and Garraf basins (modified from Salas et al. in Martín-Chivelet et al. 2019). SB, Sitges Sub-basin; EMB, El Montmell Sub-basin; EPB, El Perelló Sub-basin; SaB, Salzedella Sub-basin; OrB, Orpesa Sub-basin; PGB, Penyagolosa Sub-basin; CB, Cedramán Sub-basin; GB, Galve Sub-basin; LPB, Las Parras Sub-basin; OLB, Oliete Sub-basin; MoB, Morella Sub-basin. C) Geological map of the Herbers-Mas de Petxí area showing the location of the section logged (modified from Canérot and Leyva 1978). D) Geological map of the area of Fredes showing the location of the section logged (modified from García de Domingo and López-Olmedo 1985).

Figure 2. Lithostratigraphy of the uppermost Jurassic–Lower Cretaceous record of the Morella Sub-basin (after Salas et al. 2001, Bover-Arnal et al. 2016).

Figure 3A. Stratigraphic log of the lower part of the Herbers-Mas de Petxí section showing the location from where the samples were obtained and the distribution of the charophyte species. Charophyte biostratigraphy follows the new biozonations presented here. Geographical and geological location shown in Fig. 1C. See Figure 5 for legend.

Figure 3B. Stratigraphic log of the upper part of the Herbers-Mas de Petxí section showing the location from where samples were obtained and the distribution of the charophyte species. Charophyte biostratigraphy follows the new biozonations presented here. Geographical and geological location shown in Figure 1C. See Figure 5 for the legend.

Figure 4. Stratigraphic log of the Fredes section. Charophyte biostratigraphy follows the new biozonations presented here. Geographical and geological location are shown in Figure 1D. See Figure 5 for the legend.
Figure 6. European and Eurasian Barremian charophyte biozones and their calibrated numerical ages. Numerical ages and geomagnetic polarity were obtained from Gradstein et al. (2012). Ammonite biostratigraphy was taken from Reboulet et al. (2018) and the age ranges for the ammonite biozones were obtained from Ogg and Hinnov (2012). The previous Barremian–early Aptian European biozones (Grambast 1974, Feist et al. 1995, Mojon 1996, 2002, Riveline et al, 1996, Martín-Closas et al. 2009) and Chinese biozones (Wang and Lu 1982, Yang et al. 2008) are compared with those proposed in the present study, considering only the charophyte biozones and the age given by the different studies and not the absolute ages. The correlation with ammonite biozones was only well-established by Riveline et al. (1996) and Martín-Closas et al. (2009). The dashed line in the Eurasian C. grovesii var. jiuquanensis biozone (present study) represents the maximum numerical age of this biozone.

Table 1: Charophyte species identified in the Herbers-Mas de Petxí and Fredes sections (the Morella Sub-basin). Letters A-H and A-E are referring to the intervals described in sections 5.1 and 5.2. L: Laterite.

Table 2. Analytical results and the derived numerical ages obtained from the low-Mg calcite oyster shells analysed in the present study. See Figures 3A–B and 4 for the location from where the analysed samples were obtained. The numerical ages follow those of Gradstein et al. (2012) and are derived from the ‘LOWESS 5 fit 26 03 13’ look-up table by McArthur, and those of Gradstein et al. (2020) derived from the ‘LOESS 6 16 03 2020’ by McArthur (personal communication, 2021). ± 2 s.e., standard error; N.A., not analysed; Deg.Alt., degree of alteration; D.Alt., diagenetically-altered; Fr.Alt., freshwater-altered; N.Alt., non-altered. Samples and the analytical results used to derive the numerical ages are shown in bold.
Plate 1. A) Porochara maestratica (lateral view; sample H8; specimen no. 85579 MGSCB); B) P. maestratica (apical view; sample H8; specimen no. 85580 MGSCB);
C) aff. Mesochara harrisii (lateral view; sample H39; specimen no. 85585 MGSCB);
D) aff. M. harrisii (apical view; sample H39; specimen no. 85586); E) Echinochara lazarii (external cast of the gyrogonite; sample H95; specimen no. 85589 MGSCB); F) E. lazarii (inner series of the utricle; sample H94; specimen no 85592 MGSCB); G) E. lazarii (outer series of the utricle; sample H88; specimen no. 85601 MGSCB); H)
Globator maillardii var. trochiliscoides (lateral view; sample H27; specimen no. 85602 MGSCB); I) G. maillardii var. biutricularis (lateral view; sample H44; specimen no. 85605 MGSCB); J) Atopochara trivolvis var. triquetra (lateral view; sample H51; specimen no. 85608); K) A. trivolvis var. trivolvis (lateral view; sample H94; specimen no. 85611 MGSCB); L) Clavator grovesii var. gautieri (lateral view; sample FR27; specimen no. 85651 MGSCB); M) C. grovesii var. gautieri (adaxial view; sample FR27; specimen no. 85678 MGSCB); N) C. grovesii var. jiuquanensis (lateral view; sample H95; specimen no. 85614 MGSCB); O) C. grovesii var. jiuquanensis (adaxial view; sample H88; specimen no. 85616 MGSCB); P) C. harrisii var. dongjingensis (lateral view; sample H38; specimen no. 85617 MGSCB); Q) C. harrisii var. harrisii (lateral view; sample H38; specimen no. 85620 MGSCB); R) C. harrisii var. reyi (lateral view; sample H76; specimen no. 85622 MGSCB); S) C. calcitrapus var. jiangluoensis (apical view; sample H38; specimen no. 85623 MGSCB); T) C. calcitrapus var. calcitrapus (apical view; sample H51; specimen no. 85626 MGSCB).

Plate 2. A) Ascidiella stellata var. stellata (lateral view; sample H6; specimen no. 85677 MGSCB); B) A. stellata var. stellata (apical view; sample H6; specimen no. 85630 MGSCB); C) A. stellata var. lata (lateral view; sample H44; specimen no. 85632 MGSCB); D) A. stellata var. lata (apical view; sample H44; specimen no. 86633 MGSCB).
MGSCB); E) *Ascidiella triquetra* (lateral view; sample H27; specimen no. 85636

MGSCB); F) *A. triquetra* (apical view; sample H27; specimen no. 85637 MGSCB); G) *A. cruciata* (abaxial view; sample H94; specimen no. 85638 MGSCB); H) *A. cruciata* (apical view; sample H94; specimen no. 85639); I) *Hemiclavator adnatus* (adaxial view; sample H12; specimen no. 85641 MGSCB); J) *H. adnatus* (lateral view; sample H12; specimen no. 85642 MGSCB); K) *H. neimongolensis var. posticecaptus* (adaxial view; sample H12; specimen no. 85644 MGSCB); L) *H. neimongolensis var. posticecaptus* (lateral view; sample H12; specimen no. 85645); M) *H. neimongolensis var. neimongolensis* (adaxial view; sample H39; specimen no. 85648 MGSCB); N) *H. neimongolensis var. neimongolensis* (lateral view; sample H39; specimen no. 85649 MGSCB); O) *Pseudoglobator paucibracteatus* (lateral view; sample H96; specimen no. 85650 MGSCB).
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Charophyte species

- Total distribution
- Occurrence
- First Occurrence
- Last Occurrence

Samples

- H3  Marl (levigates)
- H6b Limestone (microfacies)
- OyH1 Oyster ($^{87}$Sr/$^{86}$Sr ratios)
### Table 1

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Table 2

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<th>Mn (ppm)</th>
<th>Fe (ppm)</th>
<th>$^{87}$Sr/$^{86}$Sr measured</th>
<th>$^{87}$Sr/$^{86}$Sr corrected</th>
<th>Deg. Alt.</th>
<th>Gradstein et al. (2012) min</th>
<th>Age (MA)</th>
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