

The middle-late Aalenian event: A precursor of the Mesozoic Marine Revolution

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- 1 The middle-late Aalenian event: a precursor of the Mesozoic Marine Revolution
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Abstract

16 The Aalenian was a time marked by profound environmental and carbon cycle 17 changes. Still, the scarcity of detailed studies hinders a better understanding of the triggering 18 mechanisms and the larger-scale context of Lower to Middle Jurassic environmental 19 perturbations. This study provides an unprecedented high-resolution biostratigraphically wellconstrained carbon isotope record ($\delta^{13}C_{org}$ and $\delta^{13}C_{carb}$) for the upper Toarcian-lower 20 21 Bajocian interval of two expanded limestone-marl alternation successions in France (French 22 Subalpine Basin) and Chile (Andean Basin). The comparison with available records from the 23 Tethyan and Boreal domains highlights that medium-term carbon isotope fluctuations are 24 reproducible across different palaeoceanographic settings from both hemispheres, providing 25 for the first time compelling evidence for recurrent perturbations of the global carbon cycle 26 during the Aalenian. Combined with a review of geological events, climate modes, abundance and diversity of major fossil groups, and trophic conditions inferred from the calcareous nannofossil record, our study fills the gap in our understanding of global environmental changes in a so-far poorly documented Middle Jurassic stage, intercalated between the early Toarcian Oceanic Anoxic Event and the early Bajocian carbonate crisis. Importantly, this compilation indicates that the Aalenian was a pivotal time interval of environmental perturbations, likely triggering the Mesozoic Marine Revolution.

- **Keywords:** Carbon isotope stratigraphy; Recurrent carbon cycle perturbations; Biotic events;
- 35 Jurassic

1. Introduction

The Mesozoic was punctuated by several episodes of environmental changes associated with climatic instabilities, severe biotic crises, and perturbations of the global carbon cycle expressed as prominent carbon isotope excursions (CIEs) in marine and terrestrial inorganic and organic materials (e.g., Jenkyns et al., 2002; Dera et al., 2011; Bodin et al., 2015; Korte et al., 2015; Storm et al., 2020). These perturbations were often associated with the development of marine anoxia and the deposition of organic-matter rich strata and are commonly reported as Oceanic Anoxic Events (OAEs, see Jenkyns, 2010 for a review). Over the last decades, the Toarcian-OAE (or T-OAE; e.g., Hesselbo et al., 2000; van Breugel et al., 2006; Suan et al., 2011; Bodin et al., 2016; Xu et al., 2017; Fantasia et al., 2018; Krencker et al., 2020) and the early Bajocian Event (e.g., Suchéras-Marx et al., 2013; Bodin et al., 2017, 2020) have attracted much attention because they represent such episodes of global environmental change associated with large-amplitude CIEs.

The T-OAE was one of the most extreme hyperthermal events of the Mesozoic, likely triggered by intense volcanic activity in the Karoo-Ferrar Province and associated massive release of greenhouse gases into the atmosphere-ocean system (e.g., Courtillot, 1994; Duncan et al., 1997; Pálfy and Smith, 2000; Svensen et al., 2007; Suan et al., 2008). The defining benchmark for the T-OAE is a negative CIE thought to result from the injection of isotopically light volcanogenic, thermogenic, and/or biogenic carbon into the exogenic reservoirs (e.g., Hesselbo et al., 2000; Kemp et al., 2005; McElwain et al., 2005; Cohen et al., 2007; Svensen et al., 2007; Suan et al., 2008; Them et al., 2017; Krencker et al., 2019). The early Bajocian Event is characterized by faunal and floral turnovers in pelagic ecosystems, which may represent part of the Mesozoic Marine Revolution (Vermeij, 1977; Wiggan et al., 2017; Suchéras-Marx et al., 2019; Rojas et al., 2021). These changes are associated with a global carbon cycle perturbation expressed as a positive CIE, thought to reflect elevated rates of organic carbon burial linked to enhanced primary productivity (Bartolini et al., 1999; Jenkyns et al., 2002; Hesselbo et al., 2003; O'Dogherty et al., 2006; Suchéras-Marx et al., 2013; Giraud et al., 2016; Erba et al., 2019; Bodin et al., 2020). Magmatic pulses related to major tectonic rearrangement during the early Middle Jurassic (Bartolini and Larson, 2001; Bill et al., 2001; Labails et al., 2010) have been proposed as a potential trigger of the early Bajocian Event. The injection of CO₂ and/or methane clathrates into the atmosphere may have induced a cascade of environmental feedbacks, including greenhouse conditions, enhanced hydrological cycle and weathering, marine eutrophication, and shallow water carbonate production crisis (e.g., Suchéras-Marx et al., 2015; Bodin et al., 2017).

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Bracketed in between the Toarcian and the Bajocian, the Aalenian has received much less attention, although it was likely a time of environmental instabilities marked by carbon cycle perturbations and marine biotic turnovers (e.g., O'Dogherty et al., 2006; Sandoval et al., 2008; Gómez et al., 2009). Hence, some important information can be gained from this

bridging time interval to put the T-OAE and early Bajocian Event into a broader context and better understand global carbon cycle dynamics during the Mesozoic.

Here, we present a high-resolution biostratigraphic (calcareous nannofossils), sedimentological and geochemical (stable carbon isotopes, Rock-Eval pyrolysis) study of two expanded successions, namely Le Brusquet (French Subalpine Basin, SE France) and Agua de la Falda (Andean Basin, N Chile). These successions were previously dated by ammonite faunas and spanned the upper Toarcian–lower Bajocian interval. The main objective of this paper is to assess the potential causal links between the carbon isotope perturbations and palaeoenvironmental changes significantly affecting the carbonate factory, as depicted from various inorganic, organic, and biotic proxies. Specifically, this study provides (i) detailed carbon isotope records of bulk organic matter and carbonates, (ii) a correlation of the studied sections with coeval sites using nannofossil biostratigraphy and carbon isotope stratigraphy, and (iii) an evaluation of the stratigraphic reproducibility of the carbon isotope variations and an appraisal of the influence of local processes on the global signal.

2. Geological setting

Le Brusquet and Agua de la Falda localities offer extended and continuously exposed strata of upper Toarcian–lower Bajocian age and include both the Toarcian/Aalenian and the Aalenian/Bajocian boundaries, according to previously established ammonite biozones (Le Brusquet: Mouterde et al., 1966; Caloo, 1970; de Graciansky et al., 1993; Agua de la Falda: von Hillebrandt and Schmidt-Effing, 1981; Marticorena and Tapia, 1982). The successions are composed of marly limestone-marl alternations, deposited in the French Subalpine Basin and the Andean Basin, respectively (Fig. 1).

The development of the French Subalpine Basin began during the Middle to Late Triassic (e.g., de Granciansky et al., 1993; Razin et al., 1996; Mariotti et al., 2012). During the Middle Jurassic, the basin was a transitional zone between the epicontinental sea of the Paris Basin and the deeper Ligurian Tethys, along the NW margin of the Tethys (Fig. 1; Ferry and Rubino, 1989; Thierry, 2000). The Le Brusquet succession is now exposed in SE France (Fig. 1), ~10 km NE of the town of Digne-les-Bains along the Bouinenc stream and the D22 road linking the villages of Marcoux and Draix (GPS coordinates, base of the section: 44°08′10.07′N; 06°18′14.19′E). In the studied area, Middle Jurassic sedimentary successions were covered by at least 3 km of Upper Jurassic to Eocene sedimentary deposits and are part of the Digne thrust sheets, which overthrusted autochthon strata towards the SW (Gidon and Pairis, 1992; Lickorish and Ford, 1998). The Le Brusquet succession was buried under ca. 3 km overburden (Haccard et al., 1989) and was likely impacted by burial diagenesis and complex tectonic history. These transformations are also suggested by the degree of organic matter maturation (oil window-beginning of gas window stage; Levert, 1989) of Middle Jurassic deposits in the Digne area.

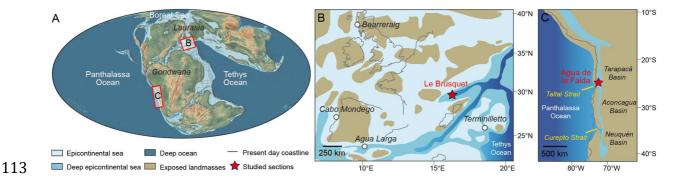


Fig. 1: (A) Global Middle Jurassic palaeogeographic reconstruction (after Scotese, 2014), with the location of the French Subalpine (B) and Andean (C) basins. (B) Palaeogeographic map of the western Tethys (after Thierry, 2000) with the location of Le Brusquet section (French Subalpine Basin, France) and other comparative key sites discussed in the text: Cabo Mondego (Lusitanian Basin, Portugal), Agua Larga (Subbetic Basin, Spain), Bearreraig (Hebrides Basin, the United Kingdom), and Terminilletto (Umbria-Marche Basin, Italy). Note that the central Tethys was not yet open during the Aalenian. (C) Palaeogeographic map of the Andean Basin, which was divided into the Tarapacá, Aconcagua and Neuquén subbasins. The location of the Agua de la Falda section and the position of the communicating straits with the Panthalassa Ocean are shown.

The Andean Basin was an elongated back-arc basin along the SW Gondwana margin, which developed in response to extensional and/or transtensional tectonics related to the subduction of the proto-Pacific plate and the formation of the Andean magmatic arc during the Jurassic and Early Cretaceous (Uliana et al., 1989; Vicente, 2005; Charrier et al., 2007). Despite the development of the volcanic arc to the west, a connection with Panthalassa Ocean was likely possible through marine connecting straits during the Middle Jurassic (Gröschke et al., 1988; Prinz et al., 1994). The Agua de la Falda succession is exposed in N Chile (Fig. 1), near the Potrerillos locality in the Atacama Region (GPS coordinates: 26°30'34.03''S; 69°20'34.5''W). The succession was likely not too strongly affected by burial diagenesis and eventually by the tectonic history related to the formation of the Andean Cordillera since this was not the case for older deposits of Toarcian age in the same area (Fantasia et al., 2018).

3. Material and methods

3.1. Sampling

Le Brusquet and Agua de la Falda sections were measured and sampled for geochemical analyses with a vertical resolution of 45–90 cm and 5–20 cm, respectively. In parallel, the lithology, sedimentary features, fossil content, and ichnofossil associations were described for each level sampled. The samples showing clear evidence of diagenetic neoformed, recrystallized calcite, or containing macroscopic allochems were avoided. About 10–20 g of macroscopically homogeneous sample were selected, dried at 45°C, crushed, and powdered using an agate mortar at the Department of Geoscience, Aarhus University prior to analysis.

3.2. Calcareous nannofossil biostratigraphy

Smear slides for calcareous nannofossil identification were prepared at the LGL-TPE, Université Claude Bernard Lyon 1. A total of 76 samples from Le Brusquet and 20 from the Agua de la Falda were selected at regular space intervals in the marl-limestone alternations. A small amount of crushed rock was mixed with water before spreading onto a cover slide (Bown and Young, 1998). Cover slides were then mounted onto the microscope slide using Rhodopass B resin (polyvinyl acetate). Sample slides were then studied using a Leica DM750P optical microscope at 1000x magnification. Two transverses of the slide (12.8 mm²) were scanned and the encountered nannofossils were counted. The slides were further analysed to recover rare species. The preservation state of the nannofossils was evaluated based on etching and overgrowth of the specimens, using the preservation index and criteria proposed by Roth (1994).

3.3. Carbon and oxygen isotopes

Carbon and oxygen isotope compositions ($\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$, % VPDB) of aliquots of powdered whole-rock samples (Le Brusquet: n=395, Agua de la Falda: n=41) were determined using a Thermo Fisher Scientific (Bremen Germany) Gas Bench II connected to a Delta Plus XL isotope mass spectrometer at the Institute of Earth Surface Dynamics of the University of Lausanne (IDYST–UNIL). The measured $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ values were normalized to VPDB scale using reference laboratory standard (Carrara Marble, $\delta^{13}C = +2.05\%c$, $\delta^{18}O = -1.7\%c$) and international reference materials NBS18 carbonatite ($\delta^{13}C = -5.01\%c$, $\delta^{18}O = -23.01\%c$) and NBS19 limestone ($\delta^{13}C = +1.95\%c$, $\delta^{18}O = -2.2\%c$). Analytical uncertainty (1σ) monitored by replicate analyses of the laboratory standard Carrara Marble was not greater than $\pm 0.05\%c$ for $\delta^{13}C$ and $\pm 0.1\%c$ for $\delta^{18}O$.

Carbon isotope compositions of bulk organic matter ($\delta^{13}C_{org}$, ‰ VPDB) were determined on decarbonated (treatment with 10% HCl) powdered whole-rock samples (Le Brusquet: n= 689, Agua de la Falda: n= 303) by using an elemental analyser Carlo Erba 1108 connected to a Thermo Fisher Scientific Delta V Plus isotope ratio mass spectrometer at IDYST–UNIL. The repeatability and intermediate precision of the $\delta^{13}C_{org}$ values were better than 0.05‰.

3.4. Rock-Eval pyrolysis

Total organic carbon (TOC, wt.%) content, mineral carbon (MinC, wt.%) content, hydrogen index (HI, mg HC/g TOC, HC=hydrocarbons), oxygen index (OI, mg CO₂/g TOC), and T_{max} (°C) were measured on powdered whole-rock samples (Le Brusquet: n= 639, Agua de la Falda: n= 298) using a Rock-Eval 6 (Espitalié, 1985; Behar et al., 2001) at the Institute of Earth Sciences of the University of Lausanne (ISTE–UNIL). Measurements were calibrated using the IFP160000 standard. T_{max} estimates of the thermal maturation level of the organic matter (Espitalié et al., 1977; Peters, 1986). HI values were not interpreted for TOC \leq 0.2 wt.%, and T_{max} values were rejected when $S_2 \leq$ 0.3 mg HC/g avoiding misinterpretation of flat thermograms (Peters, 1986). The parameter Q_{carb} = 7.976*MinC (Jiang et al., 2017) was used to estimate the proportion of carbonate with MinC values as the sum of mineral carbon obtained during pyrolysis and oxidation phases (Lafargue et al., 1998).

4. Results

- *4.1. Lithological description and biostratigraphy*
- *4.1.1. Le Brusquet*

The upper Toarcian-lower Bajocian at Le Brusquet is about 400 m-thick and dominated by dm- to m-thick alternations of offshore marl and marly limestone (Fig. 2). The succession shows relatively monotonous lithology and is expanded compared to previously documented contemporaneous European sections (Fig. 3; e.g., Sandoval et al., 2008; Gomez et al., 2009; Price, 2010; Korte et al. 2015). The lithostratigraphy and ammonite biostratigraphy of equivalent successions in Le Brusquet area have been previously described by Mouterde et al. (1966), Caloo (1970) and de Graciansky et al. (1993), and the following ammonite zones have been recognised: Aalensis (late Toarcian), Opalinum (early Aalenian), Murchisonae (middle Aalenian), Concavum (late Aalenian), and Discites (early Bajocian). The ammonite biostratigraphy is complemented here by nannofossil data (Fig. 3). The base of the succession is dominated by dark grey marl, intercalated with few yellowish lenticular, nodular, dm-thick limestone beds (Fig. 2A), assigned to the uppermost Toarcian (Aalensis zone). The transition towards the Aalenian is marked by a progressive evolution to rhythmic marly limestone-marl alternations (Fig. 2A). The lower Aalenian (Opalinum zone) is composed of marly limestone-dominated rhythmic alternations, with few dm-thick lenticular limestone beds at the base. A 5 cm-thick brown clayey layer is intercalated at 80 m. The middle Aalenian (Murchisonae zone) is composed of massive marly limestone-marl rhythmic alternations (Fig. 2B), with the limestone becoming progressively thinner and more marly towards the top of the interval. The upper Aalenian (Concavum zone) shows marl with rhythmic intercalations of thin marly limestone beds (Fig. 2C). The lower Bajocian (Discites zone) is composed of marl, which grades into regular limestone-marl alternations (Fig. 2C).

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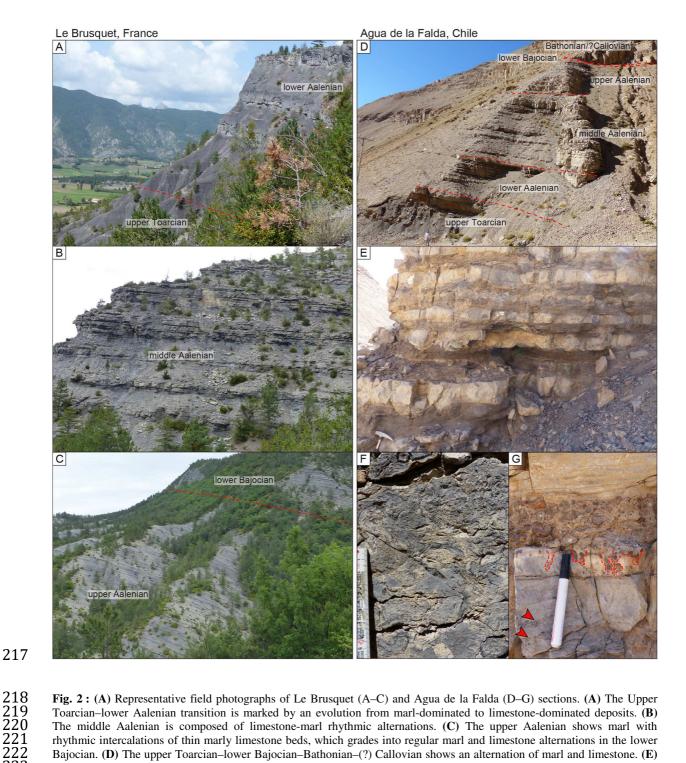


Fig. 2 : (**A**) Representative field photographs of Le Brusquet (A–C) and Agua de la Falda (D–G) sections. (**A**) The Upper Toarcian–lower Aalenian transition is marked by an evolution from marl-dominated to limestone-dominated deposits. (**B**) The middle Aalenian is composed of limestone-marl rhythmic alternations. (**C**) The upper Aalenian shows marl with rhythmic intercalations of thin marly limestone beds, which grades into regular marl and limestone alternations in the lower Bajocian. (**D**) The upper Toarcian–lower Bajocian–Bathonian–(?) Callovian shows an alternation of marl and limestone. (**E**) Close-up view of the lower Aalenian marl (sometimes finely laminated)-limestone alternations. (**F**) Close-up view of the limestone marker bed at 26 m, which is almost entirely composed of oysters. (**G**) The upper Aalenian–Bajocian transition is marked by a limestone bed with bivalve concentrations (red arrows). The top of the bed, representing the unconformity between the lowermost Bajocian and the Bathonian–(?) Callovian, is a hard ground with borings (dashed lines) infilled with phosphatic clasts from the phosphorite conglomerate bed. This bed is the basis of a Bathonian–(?) Callovian succession composed of silicified limestone.

4.1.2. Agua de la Falda

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The Agua de la Falda locality in the Potrerillos area was selected because it presents one of the most expanded and well-exposed upper Toarcian-lower Bajocian succession in Chile, offering a globally-distributed insight of the environmental conditions. It is composed of a 52 m-thick alternation of marl and marly limestone (Fig. 2). The Agua de la Falda outcrop belt can be followed laterally to the Quebrada El Asiento section (~2.5 km away from Agua de la Falda), which was previously described for its lithostratigraphy and ammonite biostratigraphy (von Hillebrandt and Schmidt-Effing 1981; Marticorena and Tapia 1982; Perez 1982). In addition, the T-OAE interval has been identified further down in the succession (Fantasia et al., 2018), bracketing the stratigraphic framework of the studied succession. The ammonite biostratigraphy is based on the scheme developed for the Andean regions and correlated with the European Standard Ammonite zones (von Hillebrandt and Schmidt-Effing, 1981; von Hillebrandt and Westermann, 1985; Riccardi, 2008). Specifically, the following ammonite assemblage zones were distinguished in the Potrerillos area: (i) Pleydellia cf. fluitans (~Aalensis), (ii) Bredyia manflasensis (~Opalinum), (iii) Zurcheria groeberi (~Murchisonae), (iv) Puchenquia malarguensis (~Concavum), (v) horizon of Podagrosiceras maubeugei (~Aalenian/Bajocian boundary). Pseudotoites cf. sphaeroceroides indicates the lowermost Bajocian, (vi) (?) Xenocephalites sp. and (?) Epistrenoceras sp. of late Bathonian to Callovian age (von Hillebrandt and Westermann, 1985).

The base of the section (Aalensis zone, upper Toarcian) is composed of wavy to nodular marly limestone beds, rarely bioclastic, alternating with thin marl interbeds. Those alternations grade up-section into a dark brown marl-dominated interval, which contains limestone nodules and large ammonites (unidentifiable, ~30 cm diameter) and alternates with nodular marly limestone beds. The marl-dominated interval is overlain by massive dm-thick bioclastic limestone beds, equivalent to the beds 8 described by von Hillebrandt and Schmidt-

Effing (1981) at the Toarcian–Aalenian transition. The rest of the succession is dominated by wavy to nodular, bioclastic (wackestone to packstone), bioturbated, marly limestone beds, which alternate with dark brown, organic-matter rich, thin-bedded to laminated, marls (Fig. 2E). The marls contain limestone nodules and mm-scale phosphatic peloids. Several firm grounds are present. The macrofossil content in the bioclastic limestones consists of bivalves (mostly oysters), brachiopods (mostly terebratulids), crinoids, ammonites, and scarce belemnites and gastropods. The shells are poorly sorted, wholly or almost entirely fragmented, and are oriented along cross bedding, or are normally graded. At 26 m, a 60 cmthick normally-graded limestone bed with an erosive base and almost composed by articulated oysters (Lopha sp.), is a marker bed in the studied succession (Fig. 2F). This coquina bed is equivalent to bed 12 described by von Hillebrandt and Schmidt-Effing (1981), which is assigned to a middle-late Aalenian age (Murchisonae-Concavum zones). The limestone and marl become silty to sandy from 40 m upwards, and the proportion of marl relative to limestone increases. The silty marl-dominated interval is capped by a 20 cm-thick erosivebased limestone bed including thin-shelled bivalves accumulated in distinct layers (Fig. 2G). The upper part of the limestone bed presents borings filled with phosphatic clasts. This is overlain by a cm-thick phosphorite conglomerate bed (Fig. 2G), laterally discontinuous with an erosive base containing heterometric, sub-angular to rounded, poorly sorted phosphate clasts (Fig. 2G). The limestone bed below the conglomerate corresponds to bed 16 described by von Hillebrandt and Schmidt-Effing (1981), which was assigned to an early Bajocian age. The conglomerate of early Bajocian age is disconformably overlain by silicified limestone (Fig. 2G), with ripple laminations, of late Bathonian–(?) Callovian age described as bed 17 by von Hillebrandt and Schmidt-Effing, 1981. Most of the Bajocian interval is hence largely condensed and/or missing, and marked by reworked deposits.

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4.2. Calcareous nannofossils

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At Le Brusquet, calcareous nannofossils were identified in 76 samples throughout the upper Toarcian-lower Bajocian interval providing a suite of biostratigraphic events (see Supplementary Information and Fig. S1), whereas the analysed samples at Agua de la Falda were all barren. At Le Brusquet, the dominant calcareous nannofossil taxa are represented by Schizosphaerella, Biscutaceae (including various species of Similiscutum, Biscutum and Discorhabdus), several species of Lotharingius, Watznaueria with a cross structure in the central area (namely: W. colacicchii, W. contracta, and Watznaueria aff. W. contracta), Watznaueria with a bridge in the central area (W. communis and several morphotypes of W. britannica defined in Giraud et al., 2006), and Watznaueria without structures in the central area (W. fossacincta and Watznaueria aff. W. manivitiae; Fig. 3). The assemblage composition shows important variations across the Aalenian (Fig. 4). The base and intermediate, carbonate-rich part of the succession (Aalensis to middle Murchisonae zones; upper Toarcian to middle Aalenian) are characterized by the lowest nannofossil content and by the dominance in nannofossil assemblage of Schizosphaerella spp. (an incertae sedis, commonly considered to be a dinoflagellate cyst; Bown, 1987). The coccolith fraction, although very rare, is dominated by Biscutaceae, with a peak at around 140-160 m (Murchisonae zone; middle Aalenian), while Lotharingius coccoliths show an inverse trend. However, being the dominant coccolith forms, such inverse trend in Biscutaceae and Lotharingius can be due to the closed-sum effect. The carbonate-rich interval also bears micrometre-sized plant debris commonly observed in the smear slides. A sudden change in the assemblage composition is observed from ~220 m up to the top of the section (middle Murchisonae to Discites zones; middle Aalenian to lower Bajocian), where nannofossil amounts progressively increased and Watznaueria spp. became dominant. In particular, this

- 306 shift to *Watznaueria* spp. dominance is driven by elevated proportions (20 to 50%) of those
- forms bearing a cross structure in the central area.

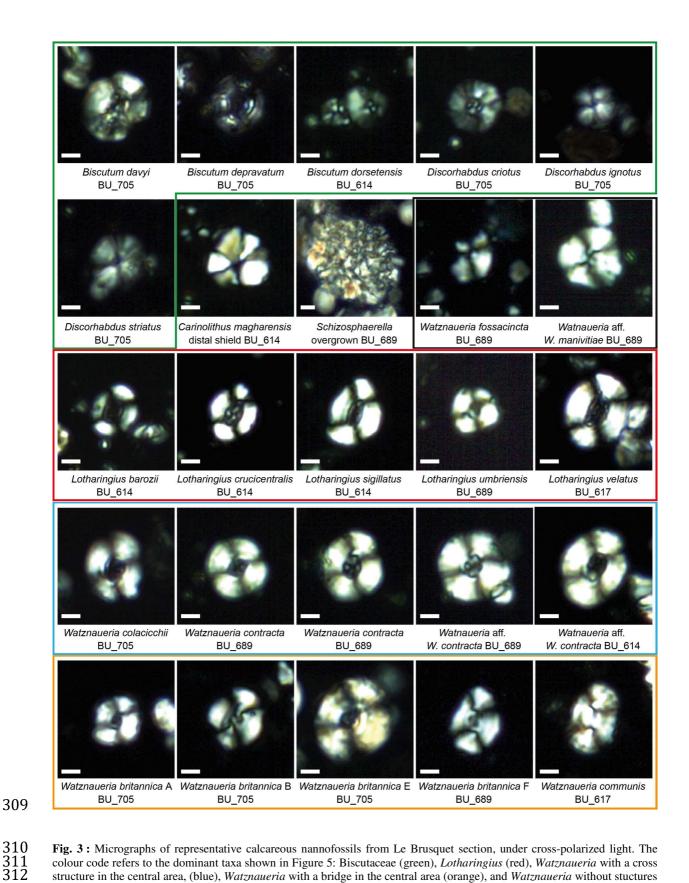


Fig. 3: Micrographs of representative calcareous nannofossils from Le Brusquet section, under cross-polarized light. The colour code refers to the dominant taxa shown in Figure 5: Biscutaceae (green), Lotharingius (red), Watznaueria with a cross structure in the central area, (blue), Watznaueria with a bridge in the central area (orange), and Watznaueria without stuctures (black).

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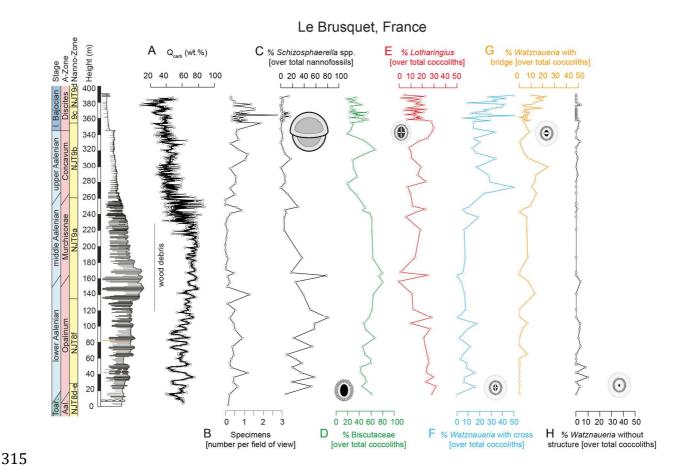


Fig. 4: (A) Carbonate content (Q_{carb}) for Le Brusquet section is shown next to (B) the calcareous nannofossil abundances, and the percentages of the dominant taxa, i.e., (C) *Schizosphaerella*, (D) Biscutaceae, (E) *Lotharingius*, (F) *Watznaueria* with a cross structure in the central area, (G) *Watznaueria* with a bridge in the central area, and (H) *Watznaueria* without structures. Note the significant increase in *Watznaueria* across the middle–late Aalenian. Wood debris are particularly present in the middle Aalenian.

4.3. Organic matter and carbonate contents

4.3.1. Le Brusquet

At Le Brusquet, TOC contents are comprised between 0.1 and 1.0 wt.%, with maximum values in the Aalensis and Opalinum zones (Fig. 5). The TOC values are high (average 0.7 wt.%) up to the Opalinum–Murchisonae zonal transition before gradually decreasing up to the lower Murchisonae zone. This is followed by a scattered increase of the TOC in the upper Murchisonae zone, with values up to 0.8 wt.% in the marl and minimum values of 0.1 wt.% in the limestone. This is followed by steady TOC contents up to the top of

the section, with values fluctuating around 0.5 wt.%. HI (2–87 mg HC g^{-1} TOC; average 21 mg HC g^{-1} TOC) and OI (18–284 mg CO₂ g^{-1} TOC; average 61 mg CO₂ g^{-1} TOC) values are relatively low throughout the section (Fig. 5), with the highest HI and lowest OI recorded from the Aalensis zone up to the lower Murchisonae zone. This is followed by a decrease of the HI and generally higher OI (\leq 284 mg CO₂ g^{-1} TOC) up to the top of the section. The T_{max} values are uniform within 494–498 °C. Q_{carb} values fluctuate between 7.8 and 87.3 wt.% and broadly mirror TOC values, with a long-term increasing trend from the base of the section up to the Murchisonae–Concavum zonal transition. This gradual increase in Q_{carb} values is expressed as a change to more massive marly limestone-marl rhythmic alternations. This is followed by a decrease in the Concavum zone and relatively steady values up to the top of the section (Fig. 5).

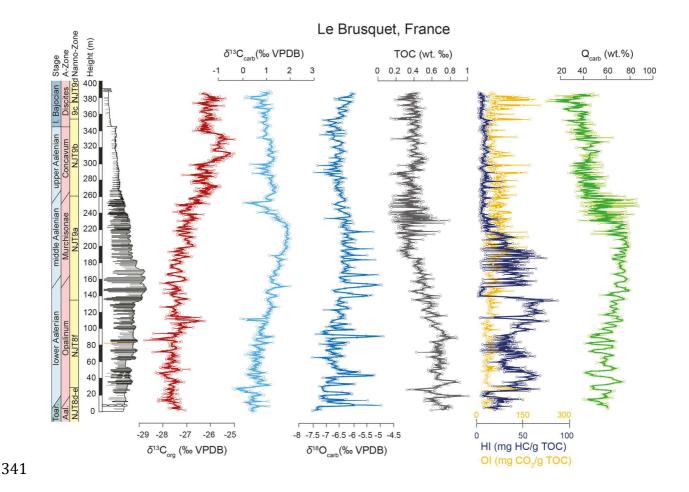


Fig. 5: Isotopic composition ($\delta^{13}C_{org}$, $\delta^{13}C_{carb}$, $\delta^{18}O_{carb}$), total organic carbon (TOC) content, hydrogen index (HI), oxygen index (OI), and carbonate content (Q_{carb}) at Le Brusquet. The trend lines correspond to a 4-points moving average.

4.3.2. Agua de la Falda

At Agua de la Falda, TOC content is generally higher than at Le Brusquet, with values up to 1.9 wt.% (Fig. 6).

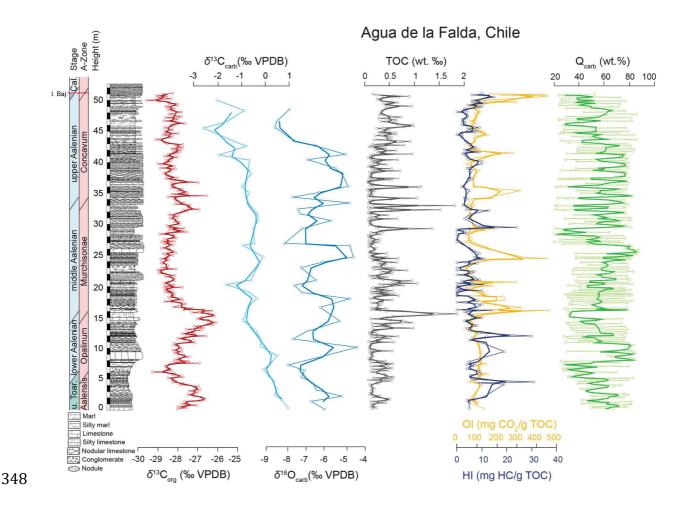


Fig. 6: Isotopic composition ($\delta^{13}C_{org}$, $\delta^{13}C_{carb}$, $\delta^{18}O_{carb}$), total organic carbon (TOC) content, hydrogen index (HI), oxygen index (OI), and carbonate content (Q_{carb}) at Agua de la Falda. The trend lines correspond to a 4-points moving average.

A gradual increase is recorded up to the Opalinum–Murchisonae zonal transition, with a maximum of 1.9 wt.% in the laminated marl. This is followed by a decrease at the base of the Murchisonae zone and a slight increase up to the middle Murchisonae zone. This trend is followed by a gradual decrease of the TOC values up to the upper Murchisonae zone, and

then a scattered increase across the Murchisonae–Concavum zonal transition. TOC values show a general increase in the Concavum zone, followed by a decreasing trend near the Bajocian–Bathonian–(?) Callovian disconformity surface. HI values are generally low (\leq 31 mg HC g⁻¹ TOC; average 6 mg HC g⁻¹ TOC), whereas OI values are moderately high (\leq 449 mg CO₂ g⁻¹ TOC; average 116 mg CO₂ g⁻¹ TOC), and both index fluctuate throughout the section (Fig. 6). Q_{carb} values vary between 0.2 and 83.8 wt.%, and show a high scatter which refects the alternation between marl and limestone. Three intervals of decreasing Q_{carb} values are recorded at the Toarcian/Aalenian boundary, in the Opalinum zone, in the upper Murchisonae zone, and in the Concavum zone, respectively (Fig. 6).

4.4. Carbon and oxygen isotopes

4.4.1. Le Brusquet

At Le Brusquet, the $\delta^{13}C_{org}$ values range between -29.9 and -24.9% (Figs. 5 and 7) and show a long-term gradual increase of about 3.0% from the upper Toarcian to the uppermost Aalenian, which culminates with maximum values in the Concavum zone. Distinct $\delta^{13}C_{org}$ medium-term fluctuations are superimposed on this long-term positive trend. The $\delta^{13}C_{org}$ profile shows a $\sim 1\%$ negative excursion near the Toarcian/Aalenian boundary, followed by a $\sim 1\%$ increase in the Opalinum zone, reaching values up to -26% with a first maximum in the upper Opalinum zone. Two distinct $\sim 1.5\%$ negative shifts are recorded at the Opalinum–Murchisonae zonal transition and in the Murchisonae zone, followed by an increase in $\delta^{13}C_{org}$ values reaching -25.5% at the Murchisonae–Concavum zonal transition. The $\delta^{13}C_{org}$ shows two $\sim 1.0-1.5\%$ negative shifts in the Concavum zone that interrupt a progressive increase to a maximum value of -24.9%. The Aalenian/Bajocian boundary is marked by a decrease in $\delta^{13}C_{org}$ (Fig. 5).

The $\delta^{13}C_{carb}$ values vary between -0.5 and 2.0% (Fig. 5) and show a general increasing trend from the Aalensis zone up to the middle Murchisonae zone, which mimics the increase of carbonate content in the section. The increasing trend is followed by a shift to more negative $\delta^{13}C_{carb}$ in the upper part of the section, where the lithology changes toward marl-dominated deposits. The $\delta^{18}O_{carb}$ values fluctuate between -7.5 and -1.7% (Fig. 5), displaying broadly a similar trend as the $\delta^{13}C_{carb}$ values.

4.4.2. Agua de la Falda

At Agua de la Falda, $\delta^{13}C_{org}$ values range between -29.6 and -25.1%, with consecutive medium-term shifts of $\sim 1.0-2.5\%$ magnitude (Figs. 6 and 7). The $\delta^{13}C_{org}$ profile shows a $\sim 2\%$ negative shift at the Toarcian/Aalenian boundary, followed by an increase of up to -26% at the Opalinum–Murchisonae zonal transition, associated with high TOC. The Murchisonae zone shows a broad stepped $\sim 2.5\%$ negative $\delta^{13}C_{org}$ shift, followed by an increase to a maximum value of -27% near the Murchisonae–Concavum zonal transition, coupled with an increase in TOC. The $\delta^{13}C_{org}$ values show two $\sim 1.5\%$ negative shifts in the lower Concavum zone and a decrease near the Aalenian/Bajocian boundary. These negative shifts are followed by a marked increase to the maximum value of -25.1% below the lower Bajocian–Bathonian–(?) Callovian disconformity surface (Fig. 6).

The $\delta^{13}C_{carb}$ values fluctuate between -3.3 and 1.0% (Fig. 6) and show a long-term decreasing trend throughout the section, interrupted by more positive values across the Murchisonae–Concavum zonal transition. The $\delta^{18}O_{carb}$ values fluctuate between -8.6 and -4.3% (Fig. 6).

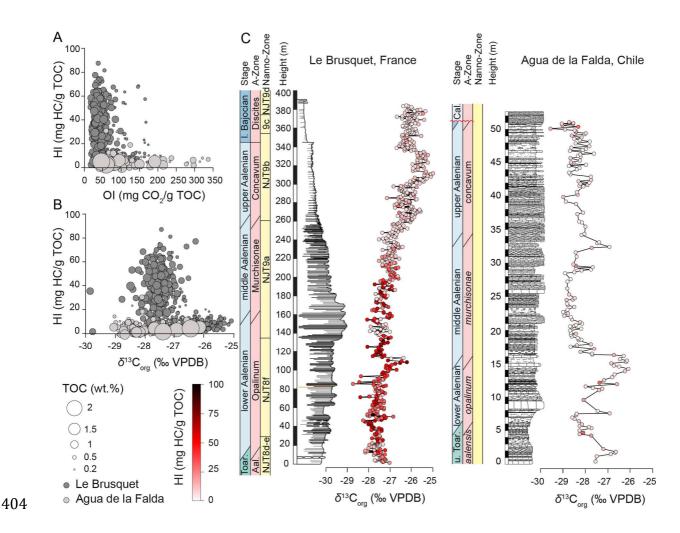


Fig. 7: (A) Scatterplot of the hydrogen index (HI) versus the oxygen index (OI) in relation with the total organic carbon (TOC) content for Le Brusquet and Agua de la Falda sections. (B) Relationship between $\delta^{13}C_{org}$ and hydrogen index for Le Brusquet and Agua de la Falda sections. (C) Bulk organic matter isotopic composition ($\delta^{13}C_{org}$) for Le Brusquet and Agua de la Falda sections, compared to the stratigraphic evolution of the hydrogen index (HI) values. Note the absence of clear relationship between the type/preservation of the organic matter and the $\delta^{13}C_{org}$ trend.

5. Discussion

5.1. Calcareous nannofossils

5.1.1. Biostratigraphic constraints

The chronostratigraphic frame and age assignment provided by ammonites for Le Brusquet section (Caloo, 1970; de Graciansky et al. 1993) is confirmed by our new calcareous nannofossil biostratigraphy (Fig. S1). In particular, the occurrence of *W. contracta* and *C.*

magharensis approximate the Toarcian/Aalenian boundary, which correlates to the base of the NJT8 zone of the Ferreira et al. (2019) biochronological scheme (also used in the GTS 2020). The middle Aalenian was identified by the consecutive FOs of *W. britannica* morphotype A and *W. communis* (NJT9a zone). The Aalenian/Bajocian boundary and basal Bajocian were recognised by the FO of *R. escaigii* and the LCO of small *Similiscutum* (NJT9c zone).

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Calcareous nannofossil data are also available for the Agua Larga (Spain, Sandoval et al., 2008) and Terminilletto (Italy, Baumgartner et al., 1995) sections, where whole-rock δ¹³C_{carb} have been measured (Aguado et al., 2008; Sandoval et al., 2008; Morettini et al., 2002). The nannofossil and carbon isotope data of these sections were compared with the new dataset from Le Brusquet and Agua de la Falda (Fig. 8). To ensure accurate comparisons with the studied sections, nannofossil data from Terminilletto (studied by one of us, EM) and from Agua Larga (Aguado et al., 2008) were reconsidered in the light of recently published taxonomic revision (see the synthesis in Ferreira et al., 2019). In particular, the specimen of W. contracta shown by Aguado et al. (2008; figure 5-39) is larger than 7 µm; thus it should be considered as a W. aff. W. contracta according to Tiraboschi and Erba (2010). Aguado et al. (2008) reported a sharp increase in proportions of W. contracta in the Agua Larga section from 59.40 m. After reconsideration, this increase may instead correspond to the inception of W. aff. W. contracta. If so, the record of the Agua Larga section is consistent with that of Le Brusquet (Fig. 8). Likewise, the specimens of W. britannica shown by Aguado et al. (2008; figure 5-44) might correspond to a transitional form close to W. manivitiae. We hence added the FO of W. aff. W. manivitiae as an alternative to the FO of W. britannica reported by Aguado al., 2008 (Fig. 8). et

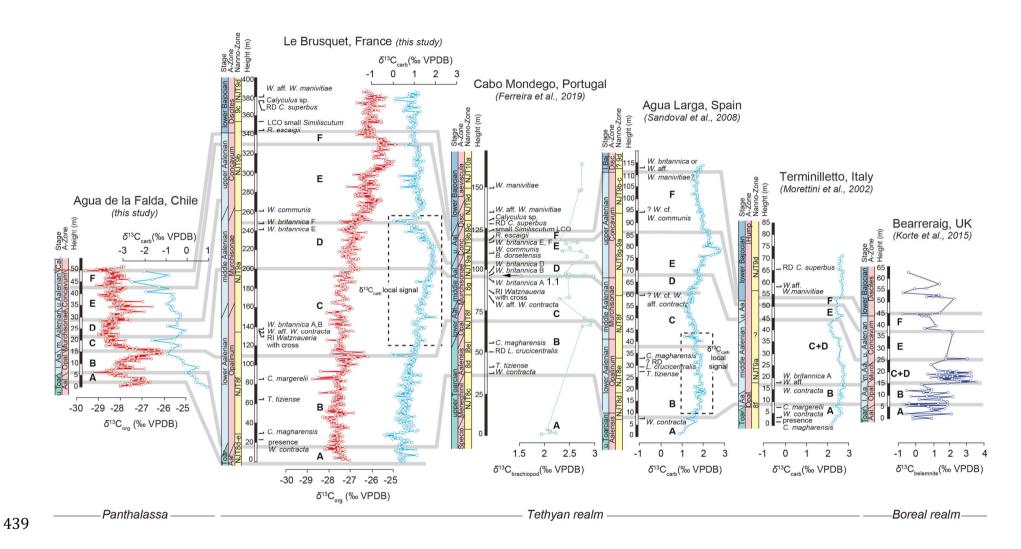


Fig. 8: High-resolution carbon isotope records (δ^{13} C_{org}, δ^{13} C_{carb}) of the upper Toarcian–lower Bajocian interval of Le Brusquet and Agua de la Falda sections, and comparison with available key records from the European area: Cabo Mondego, Lusitanian Basin, Portugal (Ferreira et al., 2019); Agua Larga, Subbetic Basin, Spain (Sandoval et al., 2008); Terminilletto, Umbria-Marche Basin, Italy (Morettini et al., 2002); Bearreraig, Hebrides Basin, the United Kingdom (Korte et al., 2015). Note that the grey lines correspond to correlations proposed in this study, based on biostratigraphy and chemostratigraphy and highlights carbon isotope shifts (intervals A–F) that can be correlated between the different sites from the Panthalassa, and Tethyan and Boreal realms. At Le Brusquet and Agua Larga, the box with dashed line indicates the interval where the δ^{13} C_{carb} signal is likely influenced by the carbonate ooze exported from adjacent platforms.

The comparison between nannofossil events at Le Brusquet, Agua Larga, and Terminilletto shows that some events can be traced across the three sections (Fig. 8). The nannofossil record of Agua Larga and Terminilletto allows a tentative application of the nannofossil zones of Ferreira et al. (2019) that provides correlation with Le Brusquet record (Fig. 8). In particular, the occurrence of *W. contracta* is relatively consistent between the three sections, and similarly to Le Brusquet, *W. aff. W. contracta* and *W. britannica* A occur close to each other at Terminilletto. According to our stable isotope and biostratigraphic data, the FO of *W. aff. W. manivitiae* is consistent between the three sections and further allow potential recognition of subzone NJT9d despite the lack of observation of the LO of *Calyculus* spp. at Agua Larga and Terminilletto (Fig. 8).

5.1.2. Palaeoenvironmental implications

Although calcareous nannofossils are very rare at Le Brusquet (Fig. 5), the percentage of the dominant taxa can be interpreted in terms of palaeoenvironmental changes, especially in the carbonate-rich part of the section between 130 and 220 m. Calcareous nannofossil abundances (Fig. 4B) provide raw information on the general trends. The Aalensis to Opalinum zones (upper Toarcian to lower Aalenian, 0–130 m) display moderate values of nannofossil abundances along with relatively high percentages of shallow-dweller taxa, proliferating either in times of relative oligotrophy (such as *Schizosphaerella* spp.; Fig. 4C) or meso-eutrophic conditions of surface waters (such as Biscutaceae and *Lotharingius* spp.; Fig. 4D–E). *Schizosphaerella* was likely better adapted to overall oligotrophic conditions in shallow-waters, where nutrients were probably recycled in times of intensified storms (Mattioli and Pittet, 2004; Aguado et al., 2008; Reggiani et al., 2010). However, some authors (Claps et al., 1995; Erba, 2004; Tremolada et al., 2005) interpreted *Schizosphaerella* as a

deep-dweller taxon, proliferating in time of surface-water oligotrophy. Biscutaceae are commonly associated with eutrophic environments both in the Jurassic and in the Cretaceous, thrivring in high-fertility surface-waters (Roth and Bowdler, 1981; Premoli Silva et al., 1989; Watkins, 1989; Erba, 1992; Mattioli and Pittet, 2004; Tremolada et al., 2006; Mattioli et al., 2008). *Lotharingius* probably dwelled in shallow-waters and was adapted to moderately high nutrient concentrations (Pittet and Mattioli, 2002; Mattioli and Pittet, 2004; Olivier et al., 2004; Tremolada et al., 2005; Mattioli et al., 2008). Overall, calcareous nannofossil assemblage at the base of the Le Brusquet section can be considered as indicative of alternating high- and low-fertility conditions in surface waters.

The Opalinum to middle Murchisonae zones (lower to middle Aalenian, ~130–220 m) is marked by the lowest relative abundances in nannofossils, decreasing proportions of *Schizosphaerella* spp. and low percentages of *Lotharingius* spp., whereas Biscutaceae show the highest values (Fig. 4C–E). Moreover, this interval also corresponds to the most carbonate-rich part of the succession and with the highest concentration in micrometer-sized wood debris observed in smear slides (Fig. 4). Such observations may suggest either increased continental runoff and bio-limiting elements input to epicontinental basins or relative proximity from source areas at times of platform progradation. Whatever the cause, the observed dominance of Biscutaceae may have been triggered by enhanced continent-derived nutrient input.

The dominant features observed in the middle Murchisonae to Discites zones (middle Aalenian to lower Bajocian, ~220–400 m) comprise the overall decreasing trend in Biscutaceae proportions and the progressive and prominent increase in *Watznaueria* bearing a cross in its central area (Fig. 4D and F). This trend encompasses an increase in *Watznaueria* bearing a bridge in the central area (Fig. 4G), which were likely opportunistic forms, proliferating in times of global eutrophication related to major oceanic reorganisation (Giraud

et al., 2016). Alternatively, they may have developed the ability to bloom in nutrient-rich environments previously unexploited by other species (Suchéras-Marx et al., 2015). Likewise, major changes have been documented in the siliceous microplankton, with a major radiolarian turnover across the middle–late Aalenian (Yao, 1997; Bartolini et al., 1999; Aguado et al., 2009). This faunal change has been interpreted as the biological response to palaeotectonic rearrangements in the Western Tethys linked to the central Atlantic spreading, which likely induced palaeoceanographic modifications, with global ocean chemistry changes and eutrophication (e.g., Aguado et al., 2008; Sandoval et al., 2008).

5.2. Effects of diagenesis and different carbonate phases on the isotope records

The potential diagenetic overprint on the marly limestone-marl alternations was evaluated at Le Brusquet and Agua de la Falda to assess if the $\delta^3 C_{carb}$ and $\delta^8 O_{carb}$ data could be used as primary environmental indicators. Diagenetic processes on pelagic carbonates generally have less influence on the carbon isotopes than oxygen isotopes (e.g., Schrag et al., 1995). The organic matter mineralization releases 13 C-depleted dissolved inorganic carbon (DIC). Therefore, recrystallization-neomorphism of carbonate phases during burial diagenesis will lower the $\delta^{13}C_{carb}$ values (e.g., Weissert et al., 1979; Marshall, 1992; Schrag et al., 1995). Actually, Le Brusquet section is composed of micrite-rich deposits, which are less prone to diagenetic fluid migration and carbon buffered. At Le Brusquet, the moderate correlation between $\delta^{13}C_{carb}$ and Q_{carb} values (r=0.45, p<0.001) suggests a relationship between the whole-rock carbonate content and the carbon isotope composition. The long-term marine carbon isotope record is mainly governed by changes in the organic carbon fluxes relative to the total carbon in the different carbon pools (e.g., Berner, 1987). Long-term $\delta^{13}C_{carb}$ records can however potentially be influenced by regional carbon cycling processes. These include

the incorporation of carbonate particles with varying carbon isotope compositions exported from platforms and pelagic mud (e.g., Swart and Eberli, 2005; Godet et al., 2006; Bodin et al., 2016). It has however been demonstrated that the contribution of nannofossil carbonate to the global oceanic carbon cycle is negligible during the Middle Jurassic (Suchéraz-Marx et al., 2012) and hence did likely not significantly contribute to the δ^3 C_{carb} record of Le Brusquet. Long-term fluctuations in the contribution of riverine DIC fluxes to the ocean can also induce shifts in the δ^{13} C_{carb} and δ^{18} O_{carb} profiles (e.g., Kump and Arthur, 1999; Kemp et al., 2020), which are decoupled from global changes in the carbon cycle. Given the difference between the carbon isotope record from Le Brusquet and other European sites, it is hence not excluded that the δ^{13} C_{carb} profile from Le Brusquet likely do not solely reflect changes in the global but also the superimposed influence of local carbon cycling processes. Furthermore, in the studied area, Middle Jurassic sedimentary successions were covered by at least 3 km of Jurassic to Eocene sediments and underwent a complex regional alpine tectonic history (Digne Nappe; Levert, 1989; Lickorish and Ford, 1998), pointing to the influence of burial diagenesis on the deposits. At Le Brusquet, the relationship between $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ values (r=0.57, p<0.001) together with generally low $\delta^{18}O_{carb}$ values suggest a significant overprint of burial diagenesis, precluding the use of the $\delta^{18}O_{carb}$ as a reliable environmental proxy. The influence of burial diagenesis is further supported by high T_{max} values (494–498°C), which indicate mature organic matter (gas window; Espitalié et al., 1985).

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At Agua de la Falda, the correlation between $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ values is very weak (r=0.19, p=0.12), suggesting that burial diagenesis had no significant influence on the carbon isotope values. This low diagenetic overprint is also supported by the lack of correlation between $\delta^{13}C_{carb}$ and Q_{carb} values (r=0.18, p=0.11). Nonetheless, the long-term negative $\delta^{13}C_{carb}$ trend is paralleled by a lithological transition towards the shallowest facies observed

throughout the succession and the increasing proportion of detrital material. This suggests that diagenetic processes, including organic matter oxidation and incorporation of 13 C-depleted carbon and/or changes in the relative content of carbonate ooze, may have driven the negative δ^{13} C_{carb} trend without any link with the global signal. Interpretation of δ^{18} O_{carb} values is excluded from the discussion since they are particularly low, suggesting diagenetic overprint related to dissolution and recrystallization-neomorphism (Schrag et al., 1995). The absence of S₂ peak hampers any reliable interpretation of T_{max} values and the influence of thermal diagenesis.

5.3. Evaluation of the degree of preservation of the organic matter

At Le Brusquet, burial diagenesis impacted the type of preserved organic matter. The more refractory terrestrial organic matter was likely preferentially preserved during burial diagenesis than the labile marine organic matter (e.g., de Lange et al., 1994; Arndt et al., 2013). Low HI and OI values (averages 21 mg HC g^{-1} TOC and 61 mg CO₂ g^{-1} TOC, respectively; Fig. 5) and high T_{max} values (494–498°C) well within the gas window point to overmature organic matter in these rocks. This high thermal maturity prevented the assessment of the organic matter type in the HI vs. OI plot (Fig. 7A). Although a minor contribution of overmature phytoplankton-derived marine organic matter can not be excluded, the occurrence of microscopic and macroscopic fossil wood particles (e.g., between 130-220 m) suggests a predominance of terrestrial organic matter, likely conveyed to the French Subalpine Basin from the surrounding emerged landmasses such as the Massif Central. At Agua de la Falda, the source of the preserved organic matter is difficult to assess because it has been overprinted by diagenetic processes (S₂< 0.3 mg HC/g and therefore not reliable T_{max} values). The absence of smectite in the Pliensbachian–Toarcian interval from the same area

(Fantasia et al., 2018) confirms that burial diagenesis affected the deposits. However, this overprint was likely not too strong in the studied area as indicated by the presence of kaolinite (Fantasia et al., 2018), which supports higher thermal burial than smectite before transformation into illite or chlorite (e.g., Burtner and Warner, 1986; Chamley, 1989).

Nonetheless, although the studied successions underwent diagenetic overprint related to the complex diagenetic and tectonic history of the studied areas, similar medium-term shifts and range of $\delta^{13}C_{org}$ values were recorded at both sites, suggesting that the carbon isotope composition of the preserved organic matter may not have been strongly modified. Furthermore, the isotope composition of the kerogen is generally preserved from low thermal maturation degrees until relatively high degree of metamorphism (e.g., McKirdy and Powell, 1974; Schoell, 1984; Spangenberg and Macko, 1998) No high thermal gradients were reached at Le Brusquet and Agua de la Falda, supporting the use of the $\delta^{13}C_{org}$ as an environmental signal. Additionally, the absence of a relationship between the preservation of the organic matter type (HI values) and the $\delta^{13}C_{org}$ values at both localities (Fig. 7) sustained the organic carbon isotope records a an environmental proxy.

5.4. Recurrent global carbon cycle disturbances across the Aalenian

The Aalenian carbon isotope stratigraphy was hitherto derived from δ^{13} C records of whole-rock carbonates and biogenic calcite from Tethyan and Boreal sites (Spain: Sandoval et al., 2008; Gómez et al., 2009, Italy: Morettini et al., 2002, United Kingdom: Jenkyns, 2002; Price et al., 2010; Korte et al., 2015, Portugal: Ferreira et al., 2015, 2019, Morocco: Krencker et al., 2014). Besides the Agua Larga succession in Spain, which is presently the most expanded high-resolution δ^{13} C_{carb} record for the upper Toarcian-lower Bajocian interval, the other records generally present low resolution data, restricted to short stratigraphic intervals,

and/or poor biostratigraphic constraints. Based on the correspondence between the different Tethyan and Boreal records, it has been suggested that some of the $\delta^{13}C_{carb}$ shifts may relate to supraregional controls (e.g., Price, 2010; Sandoval et al., 2008). Nonetheless, records from outside the Tethyan and Boreal domains and derived from organic matter were inexistent before our study leaving large uncertainties about the global extent of the carbon cycle perturbations and about triggering mechanisms.

Comparing our δ^3 C records and published datasets highlights that similar mediumterm δ^3 C patterns are shared between sites from both hemispheres. However, the interplay between global and local processes on the long-term carbon isotope signal varies between palaeoceanographic settings. Consecutive medium-term shifts can indeed be compared across Tethyan, Boreal, and Panthalassic sites (Fig. 8), providing compelling evidence that recurrent global carbon cycle perturbations occurred during the Aalenian. The Le Brusquet carbon isotope record not only depicts several medium-term shifts that were previously identified in Tethyan and Boral sites (e.g., Price, 2010; Sandoval et 2008) but also other undescribed fluctuations. Hence, the Aalenian δ^3 C record appears much more complex than previously thought as our results allow identifying seven intervals (A to F; Fig. 8) of global carbon cycle perturbations.

At Le Brusquet, the uppermost Toarcian is marked by a negative shift (interval A) in the $\delta^{13}C_{org}$ and $\delta^{13}C_{carb}$ values, reaching the lowest values at the Toarcian/Aalenian boundary. This trend is also observed in Chile at Agua de la Falda. This negative carbon isotope shift was previously identified in the $\delta^{13}C_{carb}$ records of hemipelagic carbonates and marine biogenic carbonates at different sites from the Tethyan and Boreal realms (Spain: Sandoval et al., 2008; Portugal: Ferreira et al., 2019; Italy: Morettini et al., 2002; United Kingdom: Price, 2010; Korte et al., 2015; France: Harazim et al., 2013; Morocco: Krencker et al., 2014). The

lower Aalenian (Opalinum zone) is marked by positive shifts in $\delta^{13}C_{org}$ and $\delta^{13}C_{carb}$ (interval B) at Le Brusquet and at Agua de la Falda and were previously identified in carbonates from Tethyan and Boreal sites (Price et al., 2010; Sandoval et al., 2008). The lower-middle Aalenian transition (Opalinum–Murchisonae zonal transition) from Le Brusquet is marked by a negative $\delta^{13}C_{org}$ excursion (interval C) which was not observed in the $\delta^{13}C_{carb}$ record. Nonetheless, a negative $\delta^3 C_{carb}$ excursion marked the Opalinum–Murchisonae transition at Agua Larga in Spain (Sandoval et al., 2008), suggesting that local conditions may have modulated the $\delta^3 C_{carb}$ record at Le Brusquet. Indeed, interval C at Le Brusquet shows the highest carbonate content associated with more massive marly limestone-marl rhythmic alternations (Fig. 5), suggesting that material exported from neritic environments may have shifted the δ^{13} C_{carb} record towards more positive values. Hence, the influence of local conditions related to different amounts of carbonate ooze exported from adjacent platforms may explain differences between the $\delta^{13}C_{carb}$ profiles from Le Brusquet and Agua Larga (e.g., Swart and Eberli, 2005; Godet et al., 2006; Bodin et al., 2016) (Fig. 8). Alternatively, a change in the organic matter type and/or preservation may explain differences between the organic and inorganic records. However, the absence of a relationship between HI and δ^{13} C_{org} values suggests that the δ^{13} C_{org} trend reflects a genuine supraregional/global signal. This interpretation is likely supported by a similar decrease in δ^{3} C values in the bulk carbonate from Spain, belemnites from the United Kingdom and brachiopod shells from Portugal (Fig. 8, interval C). At Le Brusquet, the Murchisonae zone (interval D) is marked by a distinct negative shift in the δ^{13} Corg. A similar trend can be identified in Chile at Agua de la Falda, but it is more difficult to trace in the less expanded successions (Fig. 8).

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Notably, at Le Brusquet the Murchisonae–Concavum zonal transition (middle–late Aalenian) is associated with a marked positive δ^{13} C trend (interval E) which can be identified

in both carbonates and organic matter. This is followed by a negative carbon isotope excursion across the Aalenian/Bajocian boundary (interval F) and then a return to less negative δ^{13} C values in the early Bajocian. These negative and positive excursions are traceable in both organic matter and carbonates across the different sites from the Tethys, Boreal domain, and Panthalassa (Fig. 9) and were previously identified as reflecting global events (e.g., Spain: O'Dogherty et al., 2006; Sandoval et al., 2008; Gomez et al., 2009, France: Corbin, 1994; Suchéras-Marx et al., 2013, Italy: Morettini et al., 2002; United Kingdom: Jenkyns et al., 2002; Hesselbo et al., 2003; Price, 2010, Morocco: Bodin et al., 2020).

The dataset from Le Brusquet and Agua de la Falda highlights for the first time the occurrence of recurrent perturbations of the global carbon cycle during the Aalenian. Although the long-term δ^{13} C trend seems to be influenced by local processes across the Aalenian, medium-term shifts (intervals A–F) are reproducible between different palaeogeographic settings in both hemispheres. Therefore, these shifts reflect synchronous changes in the organic and carbonates carbon and calls for the implication of global driving mechanisms.

5.5. Aalenian environmental perturbations

The evolution of environmental conditions across the Aalenian has been so far undervaluated compared to early Toarcian and early Bajocian events. However, evidences of climatic and environmental changes during the Aalenian (Fig. 9) indicate that it might represent a key time interval to better understand the wider context and environmental feedback mechanisms surrounding such major events.

Our results suggest that environmental changes occurred across the middle-late Aalenian. At Le Brusquet, a general increase in carbonate content associated with a decrease in calcareous nannofossil abundances is recorded across the middle Aalenian (Murchisonae zone; Fig. 7). A similar pattern was recorded in contemporaneous hemipelagic settings from Portugal (Ferreira et al., 2019) and Spain (Sandoval et al., 2008). Hence, the evolution of carbonate content at Le Brusquet likely represents a supraregional trend and reflects the intensity of carbonate production and export from surroundings platforms. The middle Aalenian was likely a time of high carbonate export into basinal settings from the Tethyan realm during a relative sea level fall (Hardenbol et al., 1998; Jacquin et al., 1998). A relative sea-level fall during the middle Aalenian has been inferred from the presence of unconformities and development of prograding shallow platform sediments in most Western Tethyan sites during this time interval and has been related to the Mid-Cimmerian tectonic uplift (de Graciansky et al., 1993; Durlet and Thierry, 2000; Léonide et al., 2007). Middle Aalenian platform environments surrounding the French Subalpin Basin (e.g., Beaujolais, Provence and Grands Causses subbasins) and other Tethyan sites (e.g., Bulgaria, Saudi Arabia, France, Germany) were marked by abrupt lithological changes such as the deposition of bioclastic (crinoids, brachiopods, sponges) calcarenititic limestone (Elmi and Rulleau, 1993; Ciszak et al., 1996; Rousselle, 2001; Floquet 2000), iron-rich oolithic limestones (e.g., Ohmert et al., 1996; Rulleau et al., 2001), or hiatuses (Metodiev, 1997; Le Nindre et al., 1990; Delfaud et al., 2000). Such changes in the sedimentation style during the middle Aalenian in platform settings point out to the initiation of a deterioration in environmental conditions. Importantly, these changes in the platform carbonate factory and pelagic carbonate producers correspond to the onset of a positive carbon isotope trend in bulk organic matter and bulk and brachiopod carbonate in contemporaneous Tethyan sites (Fig.9; Portugal, Spain, France).

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Severe palaeoenvironmental changes are recorded during the middle-late Aalenian (Murchisonae-Concavum zonal transition) as attested by the sharp decrease in carbonate content at Le Brusquet and in other contemporaneous successions from France, Portugal, and where the highest calcareous nannofossil abundances are observed. contemporaneous platform settings from the Tethyan area, peculiar sedimentation unknown in earlier sequences have been reported as thin-shelled bivalve buildups (Spain: Molina et al., 2018), siliceous sponge mud-mounds (Portugal: Duarte et al., 2001; France: Floquet et al., 2000; 2012), iron-rich oolithic limestones (Switzerland: Lauper et al., 2021), and hiatuses and/or condensed/reduced deposits (Arabia Saoudia: Le Nindre et al., 1990; Bulgaria: Metodiev, 1997; France: Delfaud et al., 2000; Rousselle, 2001; Germany: Ohmert et al., 1996). Such lithological changes in different palaeogeographic settings indicate a supraregional event of carbonate factory demise. The relative sea-level rise recorded during the late Aalenian in almost all series of Western Europe (de Granciansky et al., 1998; Durlet and Thierry, 2000) likely participated in the deterioration of neritic carbonate factory (e.g., Gabilly et al., 1985; Léonide et al., 2007). Nonetheless, biotic changes (Fig. 9) indicate that besides local factors, global changes in water mass circulation and chemistry across the middle-late Aalenian may have played a key role in driving the environmental changes. Specifically, this time interval saw the emergence of *Watznaueria*, in particular the forms with a cross spanning the central area, which will dominate up to the early Bajocian (Fig. 9; this study; Suchéras-Marx et al., 2015; Giraud et al., 2016). These forms have been interpreted as opportunistic, proliferating in times of environmental instability related to enhanced nutrient concentrations or temperature changes (Giraud et al., 2016). The genus Watznaueria as a whole was in fact very sensitive to paleoceanographic and paleoclimatic changes across the Aalenian/Bajocian boundary and the rise of these forms is likely related to the increase in carrying capacity in the photic zone (Suchéras-Marx et al., 2015). The

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supraregional increase in Watznaueria also coincides locally with the dominance of Biscutaceae (Sandoval et al., 2008; Aguado et al., 2009), generally also interpreted as eutrophic forms developing in nutrient-rich surface waters (e.g., Roth and Bowdler, 1981; Mattioli and Pittet, 2004; Tremolada et al., 2006). The concomitant emergence of Watznaueria in different Tethyan sites (Fig. 9; Portugal, Spain, France) testifies to a global change towards eutrophic conditions in the oceanic domain, whereas the presence of Biscutaceae most likely relates to local conditions specific to each basin (Fig. 9). There is compelling evidence that the Aalenian was a time of palaeoenvironmental changes, biotic reorganisation and carbon cycle perturbation. The development of eutrophic conditions during the middle Aalenian combined to relative sea-level variations likely triggered the intiation of environmental changes while the most severe changes occurred across the late Aalenian. Indeed, our results suggest that this time interval was marked by the demise of platform carbonate factories associated to an increase in eutrophic calcareous nannofossils (genus Watznaueria) which are concomitant with the most positive carbon isotope values. The middle-upper Aalenian positive δ^{13} C excursion may hence reflect increasing productivity in sea-surface waters as previously suggested (Sandoval et al., 2008) and a carbonate productivity crisis. This middle-late Aalenian event certainly represents a precursor episode of environmental changes prior to the early Bajocian event. The increase in TOC coupled to the positive $\delta^{13}C_{org}$ values recorded at Le Brusquet and Agua de la Falda across the middle– upper Aalenian can hence be interpreted as the record of increased marine primary productivity and/or enhanced organic matter burial.

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The early Middle Jurassic was a critical time interval of evolutionary and ecological changes (Fig. 9; Table 1), major tectonic rearrangement, and major changes in Mesozoic oceanic current patterns. A shift from a Toarcian warm mode to an Aalenian cold mode has been suggested based on a compilation of oxygen isotope data (Korte et al., 2015). Though

the climatic evolution is still poorly documented across the Aalenian, rising seawater temperatures have been deduced from oxygen isotopes of belemnites from Spain (Gomez et al., 2009). However, the lack of supporting data hampers to evaluate the global or local extent of this middle Aalenian warming event and to assess its causal mechanisms. The coccolithophorid event presented in this study coincides with other important biotic changes in other planktonic groups in worldwide distributed sites (e.g., Wiggan et al., 2017). Indeed, a dinoflagellate turnover has been reported across the Aalenian, and radiolarians have a quasicompletely replaced the Early Jurassic fauna (Yao, 1997; Sandoval et al., 2008).

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Interestingly, the revolution in planktonic biota coincides with the diversification of planktivorous fish (Friedman et al., 2010; Guinot and Cavin, 2016) and cephalopods, together with the extinction of Early Jurassic ammonite taxa and their replacement by families that dominated throughout the Middle Jurassic (Fig. 9; Sandoval et al., 2001; O'Dogherty et al., 2006; Wiggan et al., 2018). Conversely, within other nekton, a crisis has been documented in belemnites (Dera et al., 2016). Moreover, bivalves, brachiopods, bryozoans, and benthic foraminifera also underwent evolutionary changes across the Aalenian (Hallam, 1976; Alméras and Fauré, 2000; Taylor and Ernst, 2008; Canales and Henriques, 2008; Gómez et al., 2009). Hence, the change observed in planktonic groups across the middle–late Aalenian likely triggered a reorganisation of the entire food chain. Our study provides compelling evidence for biotic and environmental changes in both pelagic and neritic environments at a supraregional scale, pointing out to a middle-late Aalenian event. So far, the lower Bajocian event has been seen as the most extreme episode of environmental changes in the Middle Jurassic, with a major carbonate crisis and a shift to biogenic silica production and sedimentation (Bartolini et al., 1996; 1999; Cobianchi et al., 2002; Gorican et al., 2012; 2016). However, our study indicates that palaeoceanographic, Giraud et al., palaeoenvironmental, and palaeoecological changes occurred already earlier, and that the middle–late Aalenian event may hence represent a precursor to the early Bajocian event, initiating the Mesozoic Marine Revolution. Although establishing a precise correspondence between the onset of volcanic activity and environmental changes remains to date elusive, one cannot exclude that magmatic pulses may have started earlier than previously thought, i.e., before the early Bajocian. Indeed, evidence of global warming, major biotic and carbon cycle changes during the middle–late Aalenian suggests a similar cascade of environmental feedbacks driven by volcanism known from other Mesozoic events (e.g., Caruthers et al., 2013; Bond and Wignall, 2014).

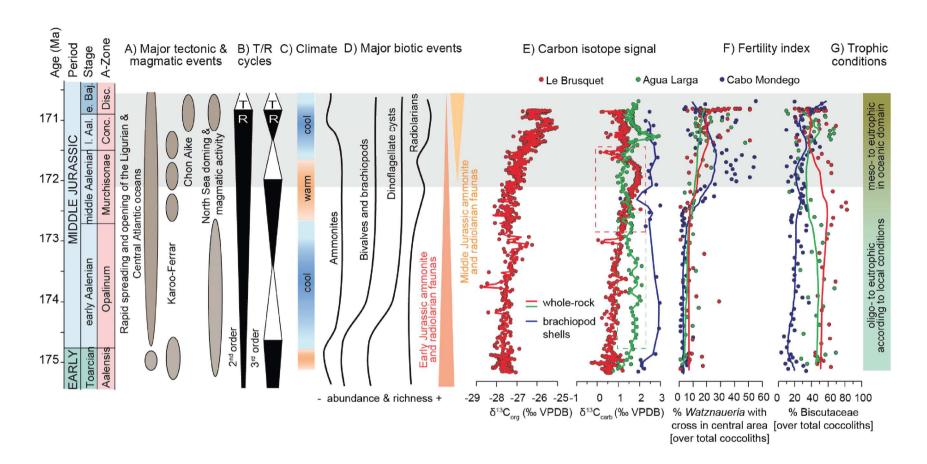


Fig. 9: Summary of key features recorded during the Aalenian time interval. (A) Timing of the main tectonic and magmatic events (after Dera et al., 2011 and reference therein). (B) 2nd and 3rd order transgressive (T) and regressive (R) sequences (Hardenbol et al., 1998). (C) Climate modes are inferred from the oxygen-isotope data from belemnites, brachiopods and bivalves (Jenkyns et al., 2002; Gomez et al., 2009; Price, 2009; Dera et al., 2011; Korte et al., 2015; Ferreira et al., 2019). (D) Major biotic events observed in different taxa, refer to the text for more details (Hallam, 1976; Yao, 1997; Alméras and Fauré, 2000; O'Dogherty et al., 2006; Sandoval et al., 2008; Taylor and Ernst, 2008; Wiggan, 2017). (E) Compilation of carbon isotopic records from Le Brusquet in France (this study), Cabo Mondego in Portugal (Ferreira et al., 2019), and Agua Larga in Spain (Sandoval et al., 2008). At Le Brusquet and Agua Larga, the box with dashed line indicates the interval where the d¹³C_{carb} signal is likely influenced by the carbonate ooze exported from adjacent platforms. The trend lines correspond to a 4-points moving average. (F) Fertility index refers to the percentage of *Watnaueria* and Biscutaceae from Le Brusquet (this study), Cabo Mondego (Ferreira et al., 2019) and Agua Larga (Sandoval et al., 2008). The trend lines are calculated using a LOWESS smoothing (α = 0.2) with the PAST software package (Hammer et al., 2001). (G) Trophic conditions are based on the proportions of *Watnaueria* and Biscutaceae. Note that the grey interval highlights the major increase in in calcareous nannofossils species indicative of fertility of oceanc water across the middle–late Aalenian.

6. Conclusions

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This study provides high-resolution carbon isotope records for the Aalenian time interval and shows for the first time a record from the southern hemisphere. Consecutive medium-term δ^{13} C fluctuations expressed in different carbon sources and identified in Tethyan, Boreal, and Panthalassic sites, suggest that the Aalenian was marked by recurrent changes in the global carbon cycle. An increase in calcareous nannofossils species related to seawater fertility coincides with an episode of platform carbonate factory demise across the middle-late Aalenian, indicating that supraregional biotic and environmental changes in both pelagic and neritic environments occurred earlier than the well-known Bajocian event. The middle-late Aalenian coincided with evolutionary and ecological changes in different taxa in worldwide-distributed sites, indicating that the middle-late Aalenian was a time of palaeoceanographic and palaeoenvironmental changes. To date, there is no consensus about the triggering mechanisms leading to these palaeoenvironmental changes. Still, tectonic rearrangement and volcanic activity during the early Middle Jurassic might have potentially played an important role. There is now compelling evidence that the Aalenian was marked by recurrent global carbon cycle perturbations associated with palaeoenvironmental and palaeoceanographic changes, which likely acted as precursors of the early Bajocian event, initiating the Mesozoic Marine Revolution.

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