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1 New insights into the early Bajocian (Middle Jurassic) carbon cycle perturbation

2

3 Baptiste Suchéras-Marx<sup>1</sup>, Guillaume Suan<sup>2</sup>, Fabienne Giraud<sup>3</sup>, Emanuela Mattioli<sup>2</sup>, Hassan M.

4 Khozyem<sup>4</sup>, Jean-Charles Mazur<sup>1</sup>, Alicia Fantasia<sup>5</sup>, Jorge E. Spangenberg<sup>6</sup>, Thierry Adatte<sup>7</sup>

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6 <sup>1</sup> Aix Marseille Univ, CNRS, IRD, INRAE, CEREGE, Aix-en-Provence, France

7 <sup>2</sup> Univ Lyon, Univ Lyon 1, ENSL, CNRS, LGL-TPE, 69622 Villeurbanne, France

8 <sup>3</sup> Univ Grenoble Alpes, Univ Savoie Mont Blanc, CNRS, IRD, Univ Gustave Eiffel, ISTerre,

9 38000 Grenoble, France

10 <sup>4</sup> Department of Geology, Faculty of Science, Aswan University, Aswan 81528, Egypt

11 <sup>5</sup> Department of Geosciences, University of Fribourg, 1700 Fribourg, Switzerland

12 <sup>6</sup> Institute of Earth Surface Dynamics, University of Lausanne, Géopolis, CH-1015 Lausanne,

13 Switzerland

14 <sup>7</sup> Institute of Earth Sciences, University of Lausanne, Géopolis, CH-1015 Lausanne,

15 Switzerland

16

17 Keywords: early Bajocian, carbon isotopes, carbonate, organic matter, phosphorus, western

18 Tethys

19

20 Abstract

21 Mesozoic strata record numerous negative and positive carbon isotope excursions (CIE). The

22 Middle Jurassic records a negative ~0.5‰ CIE at the Aalenian-Bajocian boundary followed by

23 a positive ~1.5‰ CIE covering the entire early Bajocian. Although these CIEs are recorded in

24 northern and southern western Tethys and may reflect perturbations to the global carbon cycle,

25 they remain to remain poorly addressed yet. In this study, we present new geochemical and

26 sedimentological data from the Chaudon-Norante section in France and Murtinheira section in

27 Portugal in order to better constrain the origin of the lower Bajocian CIEs. Associated with the

28 previously published  $\delta^{13}\text{C}_{\text{bulk carbonate}}$  we present new  $\delta^{13}\text{C}_{\text{org}}$  records, as well as total

29 phosphorus content, phosphorus accumulation rates (AR), and CaCO<sub>3</sub>, siliciclastics and  
30 organic matter data. Contrarily to previous interpretations, our results show no evidence for a  
31 carbonate production crisis during the early Bajocian. A slight increase in siliciclastics and  
32 phosphorus AR would argue for oceanic fertilization but without a parallel increase in organic  
33 matter AR. The obtained stratigraphic  $\delta^{13}\text{C}_{\text{bulk carbonate}}$  vs.  $\delta^{13}\text{C}_{\text{org}}$  patterns are distinctive and  
34 mimic previous box modeling results simulating an increase in productivity forced by higher  
35 rates of riverine in phosphorus and weathered-carbon input burial. The subsequent organic  
36 carbon burial produced counter-greenhouse conditions, which in turn produced a cooling by  
37 CO<sub>2</sub> uptake. Our results indicate that the lower Bajocian event presents several similarities  
38 with the upper Valanginian positive CIE. A tidy comparison of both events may help for a better  
39 understanding of the origin and consequence of such carbon cycle perturbations.

40

#### 41 1. Introduction

42 The Jurassic and Cretaceous strata are punctuated by many positive and negative carbon  
43 isotope excursions (CIEs) that have been most often attributed to major climatic,  
44 environmental, tectonic and/or biologic perturbations (e.g., Jenkyns et al., 2002; Erba, 2004;  
45 Dera et al., 2011; Friedrich et al., 2012). Many of these CIEs mark oceanic anoxic events  
46 (OAE), relatively short-lived periods (hundreds of kyr) of widespread oxygen depletion in  
47 marine waters. Some OAEs are characterized by a negative CIE, like the Toarcian OAE (T-  
48 OAE; e.g. Hesselbo et al., 2000; Suan et al., 2010) or the early Aptian OAE (OAE 1a; e.g.,  
49 Erba and Tremolada, 2004; Westermann et al., 2013). These latter OAEs were likely initiated  
50 by global warming resulting from massive <sup>13</sup>C-depleted carbon input such as methane hydrates  
51 (Hesselbo et al., 2000), thermogenic methane or volcanic CO<sub>2</sub> (e.g., Larson and Erba, 1999;  
52 Suan et al., 2008; Adloff et al., 2020; Matsumoto et al., 2022). In contrast, the Cenomanian-  
53 Turonian OAE (OAE 2) is characterized by a marked positive CIE, the widespread occurrence  
54 of organic-rich deposits, and oxygen-depleted sedimentary facies (e.g., Schlanger et al., 1987;  
55 Bomou et al., 2013). This latter event also likely occurred during a global warming trend (e.g.,  
56 Friedrich et al., 2012) interrupted by a temporary cooling, possibly driven by increased organic

57 carbon burial (Jarvis et al., 2011). This positive CIE has been classically linked to increased  
58 primary productivity and organic carbon burial, removing  $^{13}\text{C}$ -depleted organic matter from the  
59 oceanic reservoir. OAE2 was hence likely initiated by an increase in export productivity that  
60 has been linked to a rapid increase in weathering and hence in nutrient input to oceanic waters  
61 (Blätter et al., 2011; Monteiro et al., 2012; Pogge von Strandmann et al., 2013) or by increased  
62 fertilization from large upwelling areas (Trabucho Alexandre et al., 2010).

63 Although the geochemical expression of these Mesozoic events is different and are evidently  
64 either associated with positive or negative, they have not been evenly studied yet. The CIE  
65 related to the T-OAE, the Weissert event (late Valanginian) and the OAE 2 have been recorded  
66 in many inorganic and organic substrates from various depositional settings, where  
67 independent biotic and geochemical proxies have also been generated. In comparison, the  
68 lower Bajocian records two CIE that are under-documented although those might correspond  
69 to global lithospheric, climatic and biogeochemical perturbation. The former event is a  $\sim 0.5\text{‰}$   
70  $\delta^{13}\text{C}$  negative CIE documented in several localities close to the Aalenian-Bajocian boundary,  
71 as initially shown by Bartolini et al. (1999) who recorded such an excursion in bulk carbonate  
72 from central Italy. More recently, such an event has been also recorded in bulk carbonate from  
73 Spain (O'Dogherty et al., 2006), Portugal (Suchéras-Marx et al., 2012), Saudi Arabia and  
74 Oman (Al-Mojel et al., 2018), but it is absent in available bulk carbonate records in France and  
75 Morocco (Suchéras-Marx et al., 2013; Bodin et al., 2017). A negative CIE has also been  
76 reported in belemnite calcite in Murtinheira section (Lusitanian Basin, Portugal) and in the  
77 Trotternish section (Isle of Skye, Scotland) (Jenkyns et al., 2002), but both records are difficult  
78 to discuss due to a poor biostratigraphical precision in the samples from Portugal (see fig. 7 in  
79 Jenkyns et al., 2002) and to the record of a single negative value in the upper part of the section  
80 – at the end of the Discites ammonite Zone, first ammonite Zone of the Bajocian – in Scotland.  
81 Importantly, a  $>2\text{‰}$ , negative CIE is documented in fossil wood (coal and charcoal) from the  
82 poorly dated mixed marine and fluvio-deltaic strata putatively attributed to the Aalenian-  
83 Bajocian boundary in Yorkshire, UK (Hesselbo et al., 2003), suggesting that the corresponding

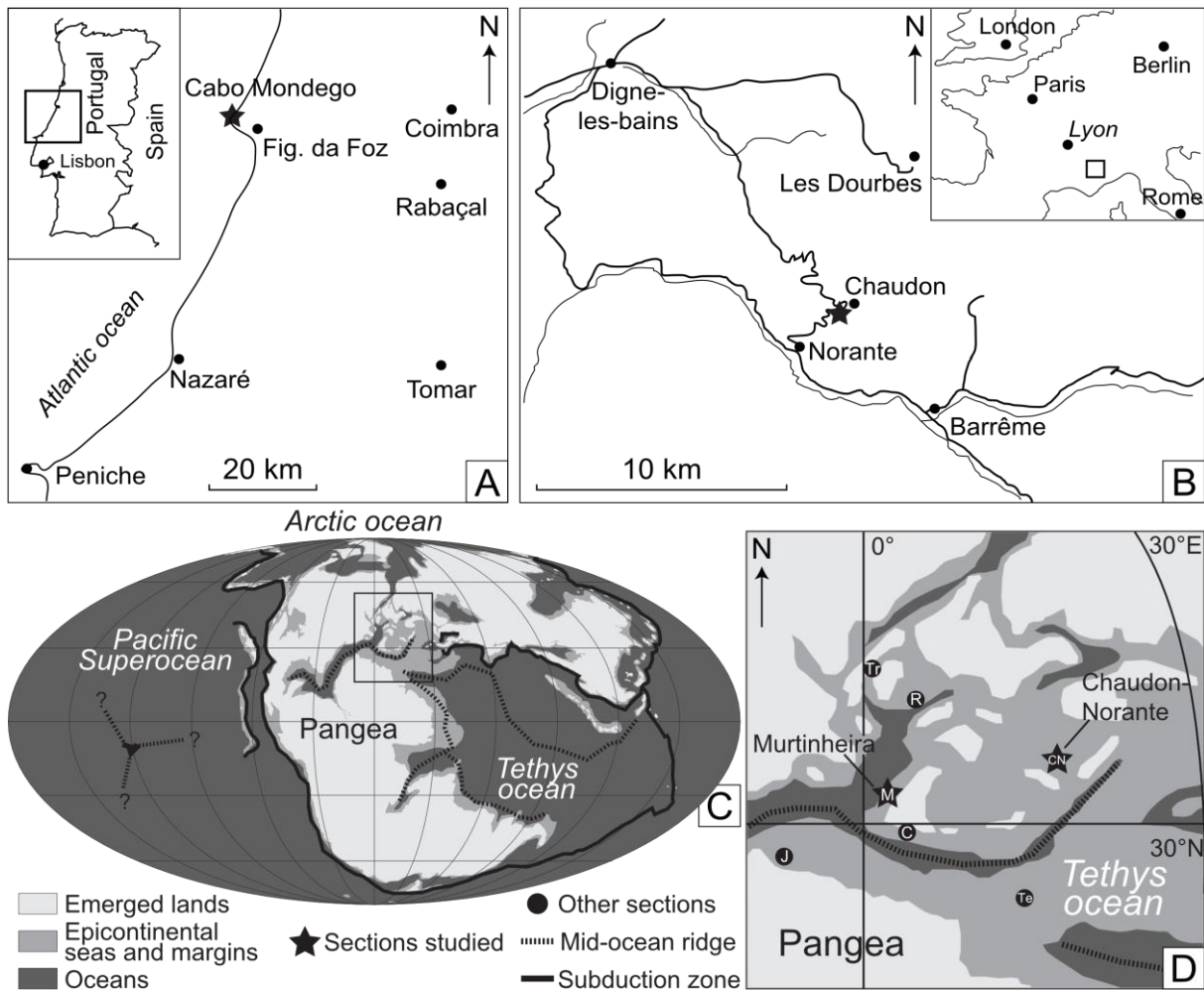
84 carbon cycle perturbation has impacted both the oceanic and the atmospheric carbon  
85 reservoirs.

86 The large positive CIE first documented by Corbin (1994) and Bartolini et al. (1996) in the lower  
87 Bajocian is better documented. This CIE covers the entire lower Bajocian with the positive shift  
88 interval corresponding to the Discites (i.e., earliest lower Bajocian ammonite Zone) to top of  
89 Propinquans (i.e., third lower Bajocian ammonite Zone, also called Sauzei) ammonite zones,  
90 and the plateau interval covers the whole Humphriesianum (i.e., fourth and latest lower  
91 Bajocian ammonite Zone) ammonite Zone. The return to pre-excursion  $\delta^{13}\text{C}$  values is recorded  
92 in upper Bajocian strata, within the Niortense ammonite Zone (i.e., earliest upper Bajocian  
93 ammonite Zone). This  $\delta^{13}\text{C}$  positive excursion is essentially recorded in western Tethys sites,  
94 namely in France (Corbin, 1994; Brigaud et al., 2009), Italy (Bartolini et al., 1996; Bartolini et  
95 al., 1999), Portugal (Suchéras-Marx et al., 2012), Spain (O'Dogherty et al., 2006; Aguado et  
96 al., 2017) and Morocco (Bodin et al., 2017). The  $\delta^{13}\text{C}$  positive excursion was recently  
97 documented in the Arabian platform of the southwestern Tethys (Al-Mojel et al., 2018). This  
98 positive CIE was reported in several bulk carbonate records (all above references), in  
99 belemnite calcite from the Isle of Skye in Scotland (Jenkyns et al., 2002) and, more recently,  
100 in brachiopod calcite from the Murtinheira section in Portugal (Ferreira et al., 2019). However,  
101 the lower Bajocian CIE has been also recorded in organic carbon material in several localities  
102 in Morocco (Bodin et al., 2017; Bodin et al., 2020), therefore bringing some insights into the  
103 geographical extent of the carbon cycle perturbation in organic material. The magnitude of the  
104 perturbation in bulk carbonate ranges from  $\sim 0.5\text{‰}$  in Portugal up to  $\sim 3\text{‰}$  in Saudi Arabia but  
105 is comprised between  $1\text{‰}$  and  $1.5\text{‰}$  in most localities. It ranges about  $3\text{‰}$  in organic matter in  
106 Morocco, the sole region with such data. The positive CIE is timely concomitant with possible  
107 global temperature decrease based on glendonite records in high latitudes (Price, 1999) or  
108 global temperature increase according to belemnites  $\delta^{18}\text{O}$  (Dera et al., 2011). This time interval  
109 also records important lithospheric changes with the Pacific plate formation (Bartolini and  
110 Larson, 2001), the Alpine Tethys opening (Bill et al., 2001), and an accelerated spreading rate  
111 in the Central Atlantic (Labails et al., 2010). Finally, this time interval also witnesses an

112 important turnover in ammonite biodiversity (O'Dogherty et al., 2006), a relevant diversification  
113 of coccolithophores (Cobianchi et al., 1992; Suchéras-Marx et al., 2015; Giraud et al., 2016)  
114 and of dinoflagellate (Wiggan et al., 2017) groups.

115 The current documentation of the lower Bajocian CIEs is insufficient to develop a satisfactory  
116 mechanistic model of the underlying carbon cycle perturbations. In this study, we attempt to  
117 document more thoroughly the lower Bajocian carbon isotope composition and sedimentation  
118 rates. The results presented here are based on the Chaudon-Norante section (Subalpine  
119 Basin, France) and the Murtinheira section at Cabo Mondego (Lusitanian Basin, Portugal)  
120 deposits (Fig. 1) with both bulk carbonate and organic matter carbon isotopes, phosphorus,  
121 calcium carbonate, organic matter, and siliciclastic accumulation rates following latest age  
122 models. These data are combined with published primary productivity records to discuss the  
123 impact of sedimentation rates and phosphorus input on the carbon isotopic composition of both  
124 oceanic biosphere and oceanic reservoirs.

125



126

127 Fig. 1: Location (star) of (A) the Murtinheira section at Cabo Mondego, Portugal and (B) the  
 128 Chaudon-Norante section, France. C. World paleogeographic map derived from Blakey  
 129 (2008). D. Western Tethys paleogeographic map derived from Blakey (2008) showing the  
 130 location of Murtinheira and Chaudon-Norante and of other sections discussed in the text: Tr.  
 131 Trotternish, Scotland; R. Ravenscar Group composite section, England; C. Casa Blanca,  
 132 Spain; Te. Terminilietto, Italy; J. Jebel Bou Kendill, Morocco.

133

## 134 2. Geological settings

### 135 2.1. Murtinheira section at Cabo Mondego

136 The Murtinheira section at Cabo Mondego is located on the Atlantic coast of Portugal near  
 137 Figueira da Foz in the Lusitanian Basin (Fig. 1A). The Lusitanian Basin was open toward the  
 138 north, the south and the west and bounded eastward by the Iberian Meseta and the Lusitanian

139 carbonate platform (Fig. 1C-D). This section exposes a key Middle Jurassic succession  
140 chosen as the global stratotype section and point (GSSP) of the Bajocian (Pavia and Enay,  
141 1997) and the auxiliary stratotype section and point (ASSP) of the Bathonian (Fernandez-  
142 Lopez et al., 2009). The section age model is based on ammonite (Ruget-Perrot, 1961;  
143 Fernandez-Lopez et al., 1988; Henriques et al., 1994) and nannofossil (Ferreira et al., 2019)  
144 biostratigraphy. The studied interval is spanning the uppermost part of the upper Aalenian  
145 (Concavum ammonite Zone) to the upper part of the lower Bajocian (Humphriesianum  
146 ammonite Zone) and is composed of alternations of limestones and marlstones (Fig. 2). A  
147 more exhaustive description of the section is presented in Suchéras-Marx et al. (2012)  
148 alongside with the  $\text{CaCO}_3$  (wt%) and the  $\delta^{13}\text{C}_{\text{bulk carbonate}}$  data.

149

## 150 2.2. Chaudon-Norante section

151 The Chaudon-Norante section is located in southeastern France in the Ravin de Coueste near  
152 Digne-les-Bains in the Subalpine Basin (Fig. 1B). The French Subalpine Basin was bounded  
153 northward by the Jura carbonate platform, westward by the Central Massif and the Ardèche  
154 carbonate platform, and southward by the Provence carbonate platform (Fig. 1C-D).  
155 Unfortunate candidate for the GSSP of the Bajocian, the Chaudon-Norante section age model  
156 is based on ammonite (Pavia, 1983) and nannofossil (Erba, 1990) biostratigraphy. The studied  
157 interval is spanning the uppermost part of the Aalenian (Concavum ammonite Zone) to the  
158 upper part of the lower Bajocian (Humphriesianum ammonite Zone) and is composed of  
159 alternations of limestones and marlstones / calcareous to argillaceous marlstones (Fig. 3). A  
160 more exhaustive description of the section is presented in Suchéras-Marx et al. (2013)  
161 alongside with the  $\text{CaCO}_3$  (wt%) and the  $\delta^{13}\text{C}_{\text{bulk carbonate}}$  data.

162

## 163 3. Analytical methods and calculation

### 164 3.1. Organic matter analysis: TOC and $\delta^{13}\text{C}_{\text{org}}$

165 A total of 80 samples (32 samples from Murtinheira and 48 samples from Chaudon-Norante)  
166 were analyzed for their total organic carbon (TOC) contents and carbon isotope composition



167 of organic matter ( $\delta^{13}\text{C}_{\text{org}}$ ). The analyzed samples correspond to limestones, argillaceous  
168 limestones, calcareous marlstones, marlstones or argillaceous marlstones. Prior to analysis,  
169 around 500 mg of powdered bulk sediment was mixed in 2 mL of distilled water and 5 to 8 mL  
170 of 2N HCl and react overnight at ambient temperature to remove calcium carbonate. The  
171 residue was rinsed with distilled water and centrifuged twice to remove the supernatant,  
172 reacted again with 2 mL of distilled water and 5 to 8 mL of 2N HCl and left overnight in a 90-  
173 95°C water bath to remove more refractory inorganic carbon phases. The residue was finally  
174 rinsed three times with distilled water to reach neutrality and oven-dried at 50°C.  
175 The TOC was measured by gas chromatography using an elementary analyzer Thermo-  
176 Finnigan Flash EA 1112 at the Université de Lausanne, Switzerland. The TOC is expressed in  
177 weight percent of the rock analyzed (wt.%) and the uncertainty is about 0.1 wt.%. The carbon  
178 isotopic ratio  $\delta^{13}\text{C}_{\text{org}}$  was measured using a Thermo Fischer Scientific Delta V Plus isotope  
179 ratio mass spectrometer at the Université de Lausanne, Switzerland. The results were  
180 standardized using Glycine, Urea, Graphite-24 and Pyridine and are expressed in per mil  
181 Vienna Pee Dee Belemnites (‰ VPDB).

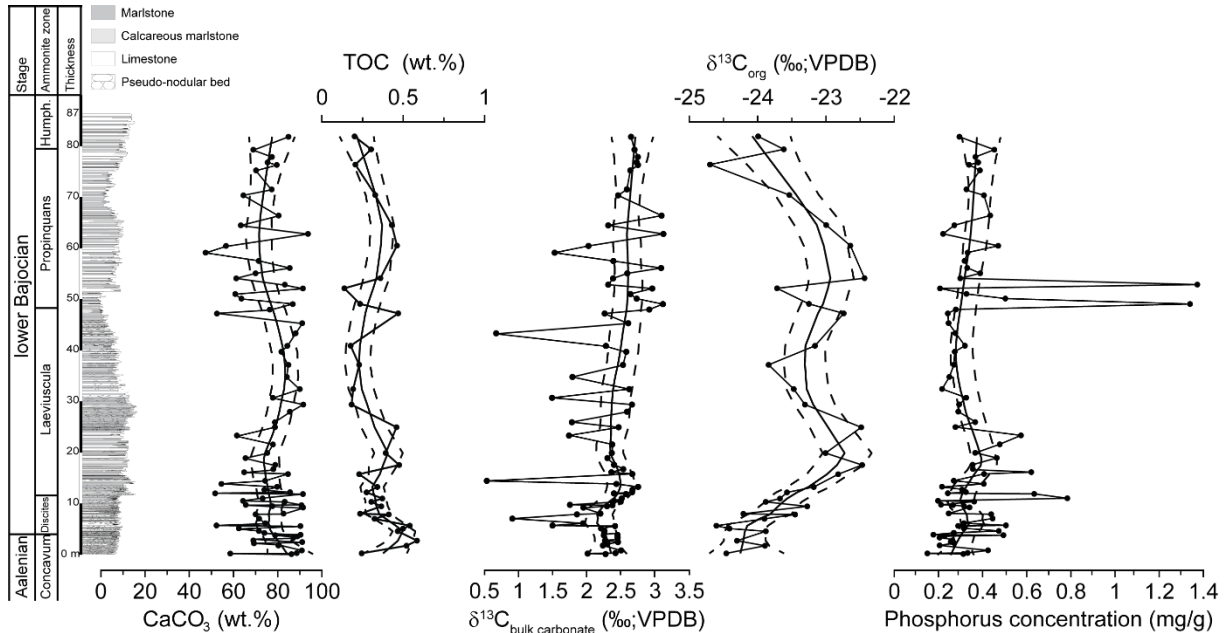
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### 183 3.2. Total phosphorus content

184 A total of 193 samples (79 samples from Murtinheira and 114 from Chaudon-Norante) were  
185 analyzed for their total phosphorus contents ( $P_{\text{tot}}$ ) using the ascorbic acid method (Mort et al.,  
186 2007). About 100 mg of powdered bulk sediment for each sample was first dried at 45°C. Once  
187 dry, 1 mL of  $\text{MgNO}_3$  was mixed to the samples and heat in a furnace at 550°C for 2 h. Once  
188 cooled, the residue was reacted with 10 mL of 1N HCl for 16 h under constant shaking. The  
189 solution was filtered with a 63  $\mu\text{m}$  filter and diluted 10 times. The filtered solution was then  
190 mixed with ammonium molybdate and potassium antimonyl tartrate forming phosphomolybdic  
191 acid. This acid was reduced with ascorbic acid forming a deep blue liquid. The total phosphorus  
192 content depends on the intensity of the blue color, which was measured using a Perkin Elmer  
193 UV/Vis Spectrophotometer Lambda 25. The concentration of  $\text{PO}_4$  was determined by

194 calibration with standard solutions (BD47 and NU81) with known concentration, is expressed  
 195 in mg/g.

196



197

198 Fig. 2: Stratigraphic variations of  $\text{CaCO}_3$  (wt.%), TOC (wt.%),  $\delta^{13}\text{C}_{\text{bulk carbonate}}$  (‰, VPDB),  
 199  $\delta^{13}\text{C}_{\text{org}}$  (‰, VPDB) and phosphorus concentration (mg/g) in the Murtinheira section.

200 The thin lines show the LOESS-smoothed curves (smoothing factor of 0.3) and the 95%CI  
 201 (dashed lines) calculated using PAST3 (Hammer et al., 2001).

202

### 203 3.3. Accumulation rates

204 Using both available  $\text{CaCO}_3$  data for both sections (from Suchéras-Marx et al., 2012; 2013)  
 205 and TOC and [P] quantified in this study, a set of derived accumulations rates (AR;  $\text{g/m}^2/\text{a}$ )  
 206 were calculated. Sedimentation rates (SR;  $\text{m/Ma}$ ) were determined using the  
 207 astrochronological time scale previously obtained at Chaudon-Norante by Suchéras-Marx et  
 208 al. (2013; alternative solution based on Gradstein et al. 2020 in supplementary data). SR was  
 209 computed at the scale of ammonite Zones in order to apply the same calculation in both  
 210 Chaudon-Norante and Murtinheira. The calculated variables are the  $\text{CaCO}_3$  AR,  
 211 Siliciclastics AR,  $\text{C}_{\text{org}}$  AR and PAR (i.e., Phosphorus AR).  $\text{C}_{\text{org}}$  AR was calculated based on  
 212 TOC, whereas Siliciclastics AR is calculated as the residues of the sum of  $\text{CaCO}_3$  AR and

213  $C_{org}$  AR. The dry bulk rock density was calculated based on the  $CaCO_3$  (wt.)/Density linear  
214 relation established in both sections by Suchéras-Marx et al. (2014):

215  $[CaCO_3] \times SR \times Density = CaCO_3 \text{ AR}$

216  $[TOC] \times SR \times Density = C_{org} \text{ AR}$

217  $[P] \times SR \times Density = PAR$

218  $(1 - [CaCO_3] - [TOC]) \times SR \times Density = \text{Siliciclastics AR}$

219

## 220 4. Results

### 221 4.1. Relative concentrations and isotopic records

#### 222 4.1.1. The Murtinheira section (Fig. 2)

223 The TOC values at Murtinheira are very low, ranging from 0.14 to 0.58 wt.%, and show a  
224 general decreasing trend stratigraphically upwards, with the highest values recorded near the  
225 Aalenian-Bajocian boundary. The lower part of the Laeviuscula ammonite Zone records few  
226 higher values around 0.5 wt.%, whereas minimum values are recorded in the middle and upper  
227 part of the Laeviuscula ammonite Zone. Overall, TOC and  $CaCO_3$  contents (Figs. 2 and 3) are  
228 negatively correlated, and TOC is very low, indicating that TOC values are mostly controlled  
229 by a variable dilution by calcium carbonate contents.

230 Organic carbon isotope values ( $\delta^{13}C_{org}$ ) values range between  $-22.4\text{‰}$  and  $-24.7\text{‰}$ , with the  
231 lowest values occurring in the Concavum and lower part of the Discites ammonite zones, and  
232 maximum value occurring in the Propinquans ammonite Zone. The  $\delta^{13}C_{org}$  values increase  
233 markedly by about  $\sim 2\text{‰}$  across the transition between the Discites–Laeviuscula ammonite  
234 zones and decrease down to  $-24.7\text{‰}$  at the end of the Propinquans ammonite Zone. The  
235 overall pattern can be described as a  $\sim 2\text{‰}$  positive CIE from the Aalenian to the Propinquans  
236 ammonite Zone, interrupted by a  $\sim 1\text{‰}$  negative CIE in the upper part of Laeviuscula ammonite  
237 Zone, and followed by a second  $\sim 1\text{‰}$  negative CIE in the upper part of the Propinquans  
238 ammonite Zone. Based on organic matter observations of 8 samples in light microscopy, the  
239 palynomorphs at Murtinheira are dominated by opaque phytoclasts, with subordinate amounts

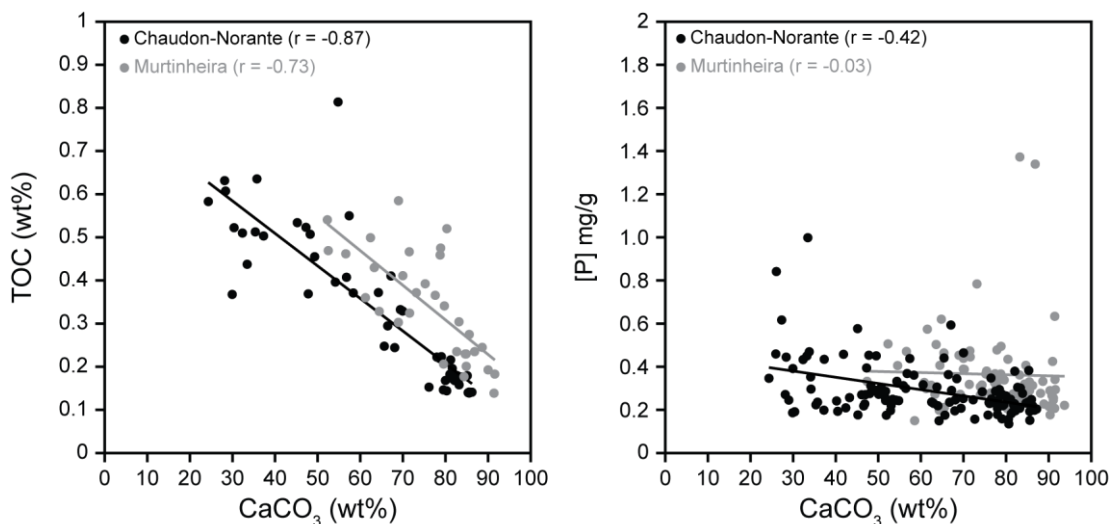
240 of translucent phytoclasts and rare terrestrial sporomorph (spore and pollen grains) and rare  
 241 marine palynomorphs (dinoflagellate cysts).

242 Phosphorus contents [P] record markedly different stratigraphic patterns. Most values are  
 243 comprised between 0.2 and 0.5 mg/g, except few samples recording higher concentration (up  
 244 to 1.37 mg/g). The [P] show a poor linear correlation with  $\text{CaCO}_3$  or TOC contents ( $r = -0.03$ ).

245 The smoothed [P] profile shows two intervals of increasing values within the Discites ammonite  
 246 Zone and in the lower part of the Laeviuscula ammonite Zone. Phosphorus contents decrease  
 247 down to around 0.3 mg/g in the middle part of the Laeviuscula ammonite Zone and increase  
 248 gradually up to almost 0.4 mg/g in the lowermost part of the Humphriesianum ammonite Zone.

249 The [P] show a poor linear correlation with  $\text{CaCO}_3$  or TOC contents ( $r = -0.03$ ; Fig. 3).

250



251

252 Fig. 3:  $\text{CaCO}_3$  (wt.%) versus TOC (wt.%) plot (left-hand side) and  $\text{CaCO}_3$  (wt.%) versus [P]  
 253 (mg/g) plot (right-hand side) with Chaudon-Norante samples in black and Murtinheira samples  
 254 in grey. There is a clear negative linear correlation between  $\text{CaCO}_3$  and TOC values, with  
 255 marlstones being more enriched in organic carbon. However, TOC is low suggesting important  
 256 effect of measurement uncertainties which is about 0.1 wt.%. There is no relationship between  
 257  $\text{CaCO}_3$  and [P] values at Murtinheira, whereas a moderately good negative linear correlation  
 258 is apparent for Chaudon-Norante, with marlstones being slightly enriched in [P].

259

## 260 4.1.2. The Chaudon-Norante section (Fig. 4)

261 At Chaudon-Norante, TOC contents are comparable to Murtinheira with values ranging from  
262 0.14 wt.% to 0.81 wt%. The TOC contents have higher concentrations in marlstones than in  
263 limestones (Figs. 3 and 4). The phosphorus concentration is ranging between 0.1 and  
264 0.5 mg/g, with the exception of few samples reaching almost 1 mg/g. Both the raw and  
265 smoothed [P] profiles show a slight increase around the Discites-Laeviuscula ammonites  
266 zones boundary (Fig. 4). The [P] shows a moderately good negative linear correlation with  
267 CaCO<sub>3</sub> contents ( $r = -0.42$ ) but there are no clear stratigraphic trends.

268 The  $\delta^{13}\text{C}_{\text{org}}$  values range between  $-24.5\text{‰}$  and  $-27\text{‰}$ , with lowest values occurring in the  
269 Concavum ammonite Zone and highest values occurring in the upper part of the Laeviuscula  
270 ammonite Zone (Fig. 3). The  $\delta^{13}\text{C}_{\text{org}}$  profile shows a first increasing trend within the Concavum  
271 ammonite Zone interrupted by a negative CIE around the Aalenian-Bajocian boundary, values  
272 increase again by 2‰ in the Discites and Laeviuscula ammonite zones up to a maximum  
273 around the Laeviuscula-Propinquans ammonite zones boundary. The  $\delta^{13}\text{C}_{\text{org}}$  values then  
274 decrease up to the top of the section, thus forming a broad, 2‰ positive CIE, but do not reach  
275 pre-excursion values (Concavum ammonite Zone and at the Aalenian-Bajocian boundary).  
276 Based on organic matter observations of 8 samples in light microscopy, the palynomorphs at  
277 Chaudon-Norante are dominated by phytoclasts (opaque and translucent) and amorphous  
278 organic matter, with rarer terrestrial sporomorphs (spore and pollen grains) and marine  
279 palynomorphs.

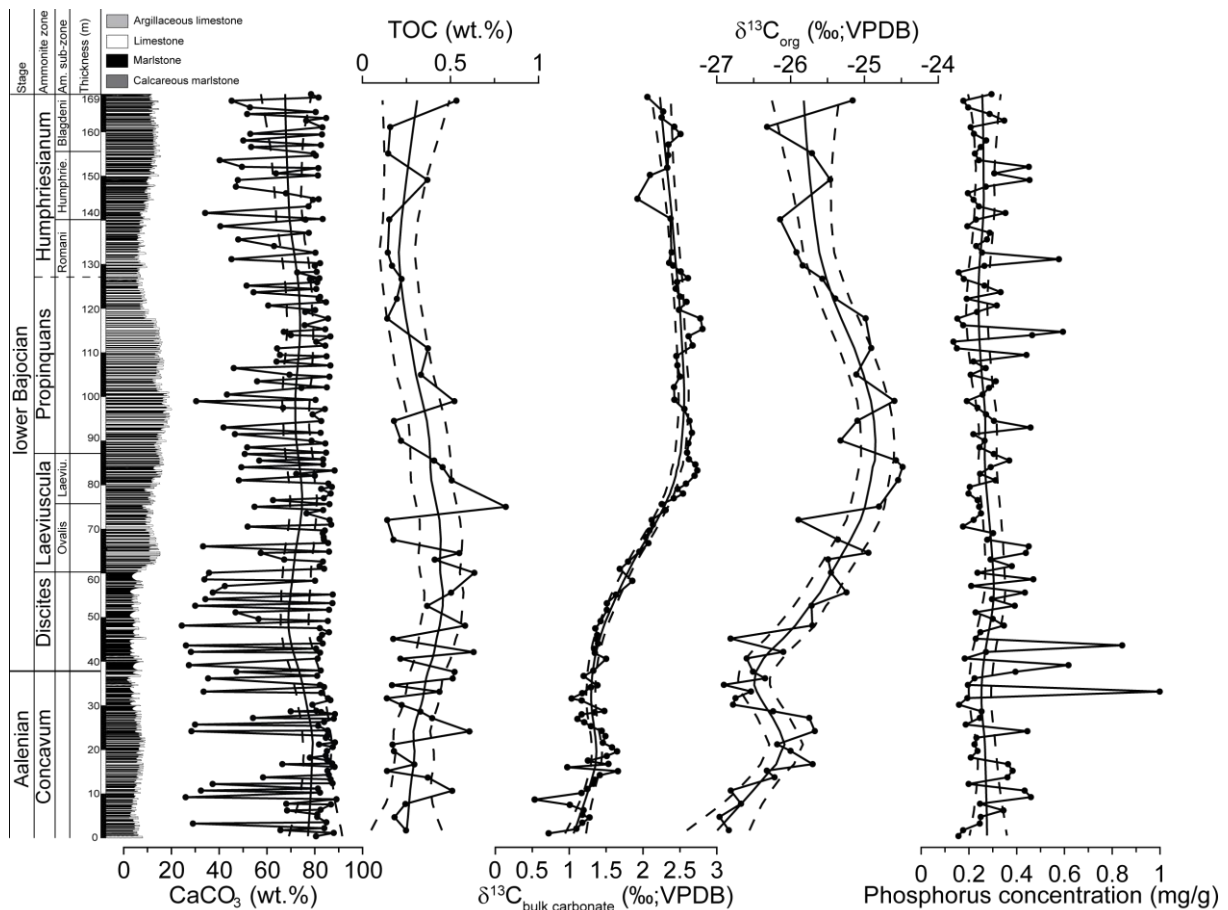
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## 281 4.2. Accumulation rates

## 282 4.2.1. The Murtinheira section (Fig. 5)

283 The SR determined at the ammonite Zone level show lowest values of  $\sim 11.5$  m/Myr in the  
284 Discites Zone. This interval is also marked by low wt.% CaCO<sub>3</sub>, resulting in an important drop  
285 in CaCO<sub>3</sub> AR. The low phosphorus AR results from the drop in sedimentation rate despite the  
286 increase in phosphorus concentration. The Laeviuscula Zone conversely has the highest  
287 sedimentation rates. Hence, all sedimentary fractions considered here are increasing in terms

288 of accumulation rates. However, only  $\text{CaCO}_3$  AR is very high all along this ammonite Zone.  
 289 The siliciclastics AR,  $\text{C}_{\text{org}}$  Ar and phosphorus AR show a peak in the lower part of the  
 290 *Laeviuscula* ammonite Zone. Finally, in the *Propinquans* ammonite Zone, two distinct patterns  
 291 are observed. The  $\text{CaCO}_3$  and phosphorus ARs tend to increase between the base and the  
 292 top of the ammonite Zone. Conversely, siliciclastics and  $\text{C}_{\text{org}}$  ARs tend to decrease between  
 293 the base and the top of this ammonite Zone. Overall, siliciclastics and  $\text{C}_{\text{org}}$  ARs have very  
 294 similar trends and  $\text{CaCO}_3$  is the most important sediment component in term of accumulation  
 295 rates. Finally,  $\text{C}_{\text{org}}/\text{P}_{\text{tot}}$  (molar ratio) is always below the Redfield ratio 1:106. Alternative results  
 296 using Gradstein et al. (2020) age model is presented in supplementary material  
 297 (Supplementary fig. 1). This solution is not described in this study.

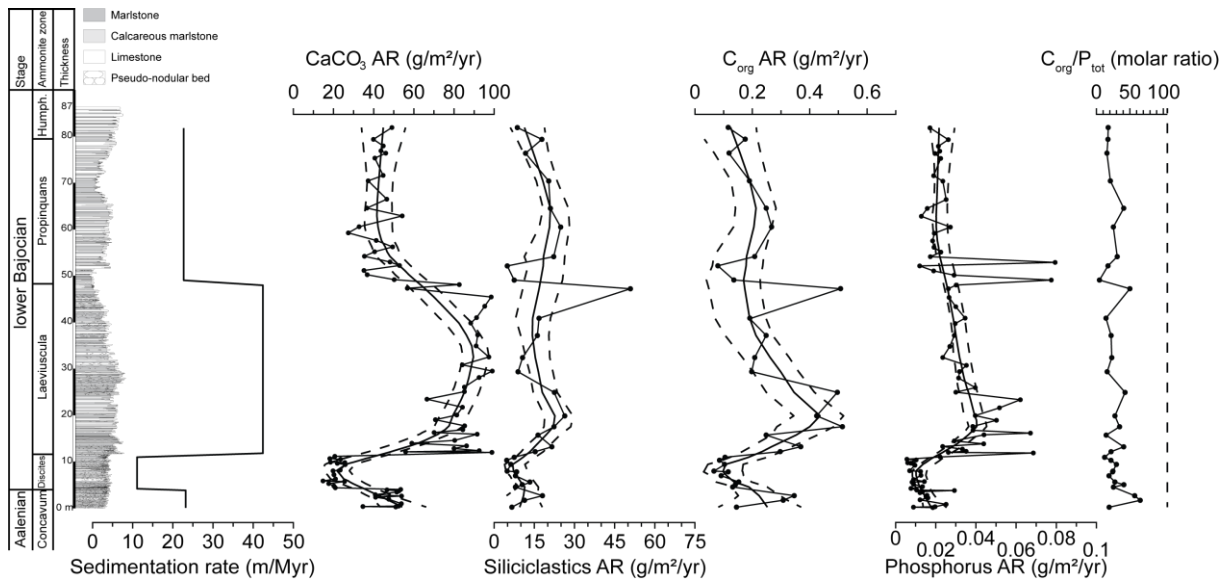


298  
 299 Fig. 4: Stratigraphical variations of  $\text{CaCO}_3$  (wt.%), TOC (wt.%),  $\delta^{13}\text{C}_{\text{bulk carbonate}}$  (‰, VPDB),  
 300  $\delta^{13}\text{C}_{\text{org}}$  (‰, VPDB) and phosphorus concentration (mg/g) at Chaudon-Norante.

301 The thin lines show the LOESS-smoothed curves (smoothing factor of 0.3) and the 95%CI  
 302 (dashed lines) calculated using PAST3 (Hammer et al., 2001).

303

304



305

306 Fig. 5: Stratigraphic changes in sedimentation rate at Murtinheira (based on Suchéras-Marx et  
 307 al., 2013 age model) and the resulting  $\text{CaCO}_3$  accumulation rates (AR;  $\text{g/m}^2/\text{a}$ ),  
 308 Siliciclastics AR ( $\text{g/m}^2/\text{a}$ ),  $\text{C}_{\text{org}}$  AR ( $\text{g/m}^2/\text{a}$ ), and phosphorus AR ( $\text{g/m}^2/\text{a}$ ), along the  $\text{C}_{\text{org}}/\text{P}_{\text{tot}}$   
 309 molar ratio. The thin lines show the LOESS-smoothed curves (smoothing factor of 0.3) and the  
 310 95%CI (dashed lines) calculated using PAST3 (Hammer et al., 2001). The dashed line in  
 311  $\text{C}_{\text{org}}/\text{P}_{\text{tot}}$  data corresponds to the Redfield ratio (1:106).

312

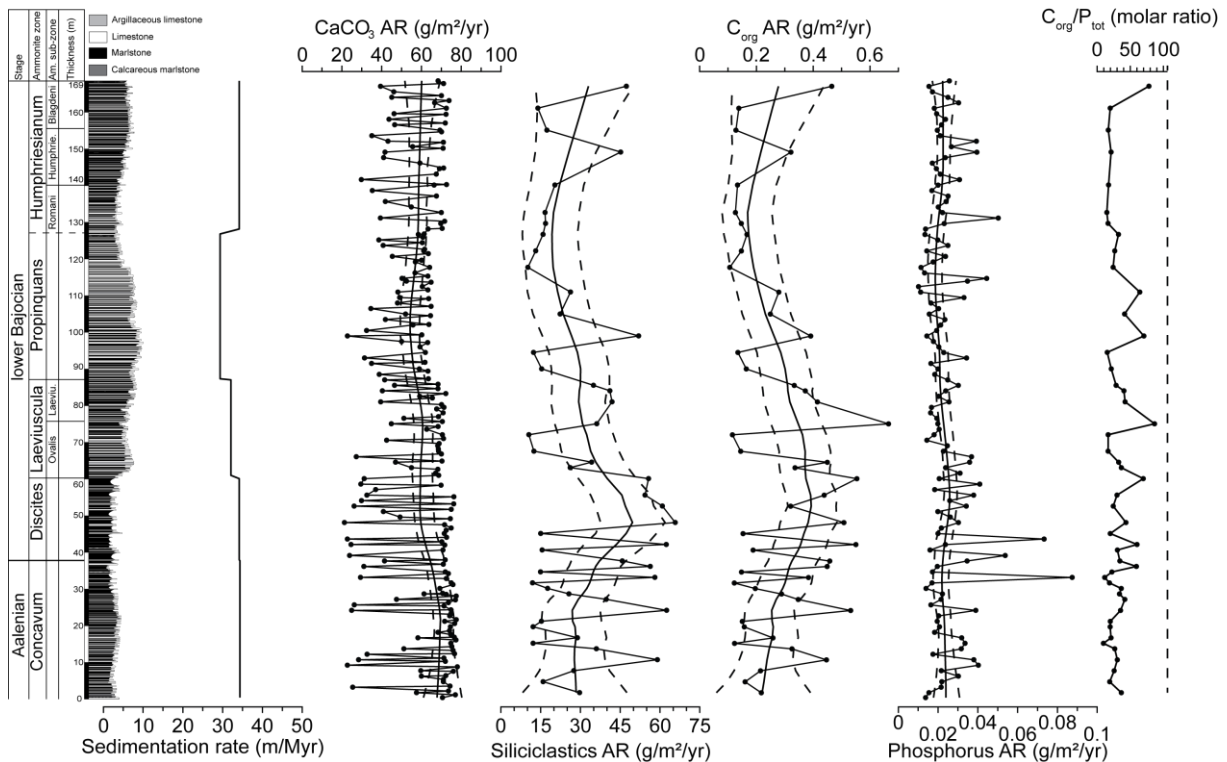
#### 313 4.2.2. The Chaudon-Norante section (Fig. 6)

314 The age model for the lower Bajocian from Suchéras-Marx et al. (2013) was built on the  
 315 astrochronology of the Chaudon-Norante section. Nevertheless, in order to compare both  
 316 sections with the same resolution, sedimentation rates are also calculated per ammonite Zone  
 317 (Fig. 6). The sedimentation rates are relatively stable all along the section with a slight  
 318 decrease in the Propinquans ammonite Zone which is the interval with rarer marlstone beds.  
 319 The  $\text{CaCO}_3$  AR is the highest sediment influx even if the clay-rich beds have in some cases a  
 320 higher siliciclastics AR. As observed in Murtinheira section, siliciclastics and  $\text{C}_{\text{org}}$  ARs have  
 321 similar trends with a maximum in the Discites ammonite Zone and a slight increase in the upper  
 322 part of the Humphriesianum ammonite Zone. Excepted few points, the end of the Concavum

323 ammonite Zone corresponds to a slight decrease in phosphorus accumulation rates whereas  
 324 the Discites ammonite Zone corresponds to a slight increase.  $C_{org}/P_{tot}$  (molar ratio) is always  
 325 below the Redfield ratio 1:106. Contrarily to the Murtinheira section, alternative results using  
 326 the Gradstein et al. (2020) age model are presented in supplementary material  
 327 (Supplementary fig. 2). This solution is not described in this study.

328

329



330

331 Fig. 6: Stratigraphic changes in sedimentation rate at Chaudon-Norante (based on Suchéras-  
 332 Marx et al., 2013 age model) and the resulting  $CaCO_3$  accumulation rates (AR;  $g/m^2/a$ ),  
 333 Siliciclastics AR ( $g/m^2/a$ ),  $C_{org}$  AR ( $g/m^2/a$ ), and phosphorus AR ( $g/m^2/a$ ), along the  $C_{org}/P_{tot}$   
 334 molar ratio. The thin lines show the LOESS-smoothed curves (smoothing factor of 0.3) and the  
 335 95%CI (dashed lines) calculated using PAST3 (Hammer et al., 2001). The dashed line in  
 336  $C_{org}/P_{tot}$  data correspond to the Redfield ratio (1:106).

337

## 338 5. Discussion

### 339 5.1. The lower Bajocian negative excursion record



340 The Aalenian-Bajocian boundary is characterized in many sections by a negative CIE (Bartolini  
341 et al., 1996; Hesselbo et al., 2003; O'Dogherty et al., 2006; Fantasia et al., 2022). This event  
342 is clearly recorded by 0.5‰ negative CIE across the Aalenian-Bajocian in the  $\delta^{13}\text{C}_{\text{org}}$  profile at  
343 Chaudon-Norante (Fig. 3) and in Morocco (Fig. 7; Bodin et al., 2017; 2020), but not at  
344 Murtinheira (Fig.2). Conversely, the CIE is hardly discernable in bulk carbonate in Chaudon-  
345 Norante and in Moroccan sites (Fig. 7), but well-expressed at Murtinheira (Figs. 3 and 7) in the  
346 lower part of the Discites ammonite Zone. Discussing the possible causes of this carbon  
347 isotope event is thus complicated due to its patchy record in the different localities and carbon  
348 substrates. The negative CIE is clearly short-lasting and has a lower amplitude than the  
349 ensuing positive CIE (Bartolini et al., 1996; O'Dogherty et al., 2006; Suchéras-Marx et al.,  
350 2013). The low magnitude and relatively short duration of this negative CIE may thus partly  
351 explain its uneven occurrence among different sites and various types of investigated material.  
352 For instance, relatively small changes in proportion of isotopically distinct OM and  $\text{CaCO}_3$   
353 sources may produce large changes in bulk organic and inorganic records (Swart and Eberli,  
354 2005; Suchéras-Marx et al., 2012; Suan et al., 2015) and may thus have blurred this small  
355 negative CIE in some Aalenian-Bajocian bulk records.

356

## 357 5.2. The lower Bajocian positive excursion record

358 Our new carbon isotope records confirm that the Bajocian positive CIE is present in organic  
359 material in Portugal (Fig. 2) and SE France (Fig. 3), and strongly suggest that this geochemical  
360 anomaly reflects a perturbation of the global exchangeable carbon reservoir. Indeed, if the  
361 positive CIE was only recorded in  $\delta^{13}\text{C}_{\text{bulk carbonate}}$ , it could have reflected a global shift in the  
362 carbonate production i.e., by change in the  $\text{CaCO}_3$  producers with more positive  $\delta^{13}\text{C}$  signature  
363 (Suchéras-Marx et al., 2012). Conversely, if the  $\delta^{13}\text{C}_{\text{org}}$  positive CIE was recorded only in  
364 Morocco (Bodin et al., 2017; 2020), local effect like the carbon residence time in the carbonate  
365 platform, or local changes in hydrology, or even change in the source and preservation of the  
366 organic matter may have been involved (Suan et al., 2015). Our concomitant record of a  
367 positive CIE in both  $\delta^{13}\text{C}_{\text{bulk carbonate}}$  and  $\delta^{13}\text{C}_{\text{org}}$  dismiss these hypotheses and confirms the large

368 geographical extent of this carbon cycle perturbation. Our new data suggest that the magnitude  
369 of the shift ranges between 1‰ and 1.5‰, in line with previous bulk organic and inorganic  
370 records, as well as brachiopod (Ferreira et al., 2019) and belemnite (Jenkyns et al., 2002)  
371 records. These latter records have an inherently low resolution and may therefore not fully  
372 capture short scale variations in  $\delta^{13}\text{C}$ . Also, changes in proportions of various inorganic and  
373 organic particles with distinct  $\delta^{13}\text{C}$  signatures may greatly distort bulk sediment records (Swart  
374 and Eberli, 2005; Suchéras-Marx et al., 2012; Suan et al., 2015). The preliminary qualitative  
375 palynological investigations of samples show no major changes in the dominant kerogen type  
376 in each section, indicating that the recorded trends are not attributable to changing contribution  
377 of distinct type of organic carbon. Nevertheless, our observations indicate that the kerogen is  
378 dominated by terrestrial opaque phytoclasts at Murtinheira, whereas phytoclasts and  
379 amorphous organic matter (AOM) dominate at Chaudon-Norante. Traditionally, structureless  
380 AOM has been considered as derived from bacteria or phytoplankton and strongly altered  
381 macrophyte remains (Pacton et al., 2011), this latter source being a volumetrically secondary  
382 component in marine sediments (Tyson, 1995). The distal position of the site and non-gelified  
383 aspect of the AOM at Chaudon-Norante could therefore indicate a marine source, which was  
384  $^{13}\text{C}$ -depleted by 2-4‰ compared to terrestrial phytoclasts during the Jurassic (Suan et al.,  
385 2015). Such a  $^{13}\text{C}$ -depletion is in good agreement with the 2‰ offset toward lower values of  
386 the Chaudon-Norante  $\delta^{13}\text{C}_{\text{org}}$  record relative to that at Murtinheira (Figs. 2 and 3). Accordingly,  
387 the  $\delta^{13}\text{C}_{\text{org}}$  record at Chaudon Norante, where  $^{13}\text{C}$ -depleted AOM kerogen occurs in higher  
388 proportions, could capture a mixed- terrestrial marine signal while the Murtinheira record,  
389 where phytoclasts dominate, may essentially captures a terrestrial signal. Such palynological  
390 observation are preliminary and qualitative, and further quantification of changing proportions  
391 of various inorganic and organic carbon sources will be required to more robustly assess the  
392 magnitude of the lower Bajocian positive CIE using bulk records.

393

394 5.3. Similarities with other Mesozoic CIEs

395 The triggers of the CIEs might be interpreted differently according to their inferred timescales.  
396 According to Gradstein et al. (2020), the  $\delta^{13}\text{C}$  increase interval is more or less lasting the whole  
397 Discites and *Laeviscula ammonite* zones thus corresponding to 580 kyr, and the plateau is  
398 lasting 580 kyr. The increasing phase and the plateau have thus the same duration and the  
399 whole perturbation lasts a little longer than one million years. The duration of ammonite zones  
400 is estimated based on the assumption of an equal duration of subzones and thus it is  
401 dependent on the number of subzones in the sub-Boreal zonation scheme (Gradstein et al.,  
402 2020). The cyclostratigraphic study of Ikeda et al. (2016) a total duration of ~4 Myr for the  
403 Bajocian-Callovian interval, but no other biostratigraphic information is available for the  
404 condensed radiolarites section they studied. This poor biostratigraphic age control makes the  
405 former results impossible to consider in our framework. According to the cyclostratigraphy  
406 established by Suchéras-Marx et al. (2013) at Chaudon-Norante, the positive isotope shift  
407 lasted 1.357 Myr and the plateau 2.724 Myr for a total of 4.082 Myr, thus approximately 4 times  
408 longer than the duration stated in Gradstein et al. (2020). These differences in the time  
409 estimation of the early Bajocian are critical and further data are necessary to definitively get to  
410 a consensual solution.

411 Following the timescale of Suchéras-Marx et al. (2013), the duration of the positive carbon  
412 isotope excursion is similar to that of the Weissert event (lasting some 5.84 Myr; Martinez et  
413 al., 2015) and much longer than other Mesozoic carbon isotope excursions associated with  
414 OAEs, which last around ~1 Myr (e.g., T-OAE, Ait-Ito et al., 2018; OAE1a, Malinverno et al.,  
415 2010; OAE2, Boulila et al., 2020). The negative excursion at the Aalenian/Bajocian boundary  
416 based on  $\delta^{13}\text{C}_{\text{org}}$  data is estimated to last between 520 kyr and 570 kyr (i.e., 27 cycles of 20 ka  
417 or 14 cycles of 37 ka or 41 ka or 5 cycles of 100 ka and 3 cycles of 20 ka). Such a duration seems  
418 incompatible with a short-lived event such as asteroid impact or methane clathrates release  
419 (e.g., Dickens et al., 1995).

420 Thus, the carbon isotope perturbations can be described as a ~550 ka negative excursion  
421 followed by a positive CIE composed of a phase of increasing values lasting 2.5 times longer  
422 than the negative excursion, but shorter than the plateau phase; and a plateau phase ~2 times

423 longer than the increasing phase. The Aalenian-Bajocian negative CIE observed in several  
424 (although not all) records therefore likely reflects a relatively short-lived carbon cycle  
425 perturbation, whereas the lower Bajocian positive CIE recorded in bulk carbonate, fossil calcite  
426 and organic matter is one of the most pronounced and long-lived carbon cycle perturbations  
427 of the Jurassic Period.

428 Altogether, the long, cumulated duration of the two CIEs is compatible with long-lived  
429 geological perturbations, such as tectonically-controlled climatic (e.g., global tectonic, large  
430 igneous province) or oceanographic changes (e.g., oceanic connections). The possible  
431 mechanisms involved are discussed in the following section.

432

#### 433 5.4. Possible environmental drivers of the lower Bajocian positive excursion

##### 434 5.4.1. Higher fertility and enhanced organic matter burial

435 Positive CIE in the geological record have been classically attributed to the increased burial of  
436  $^{13}\text{C}$ -depleted organic carbon in the oceans, thereby increasing  $\delta^{13}\text{C}_{\text{DIC}}$  values (e.g., Vincent  
437 and Berger, 1985; Schlanger et al., 1987). This increased organic carbon burial could reflect  
438 higher organic matter production and supply or enhanced preservation in the sediment favored  
439 by anoxic conditions and is thus may be partly compatible with some key features of Bajocian  
440 sediments despite the fact no black shales are observed in this study. Indeed, the lower  
441 Bajocian strata record some of the most important biotic events of the Mesozoic plankton  
442 revolution, notably the diversification of the major coccolithophores genus *Watznaueria*  
443 (Cobianchi et al., 1992; Giraud et al., 2016), the emergence of planktic foraminifera  
444 (BouDagher-Fadel, 2015), an increase in abundance of the dinoflagellate genus *Dissiliodinium*  
445 (Wiggan et al., 2018), increase in calcareous nannoplankton accumulation rates (Suchéras-  
446 Marx et al., 2015) and derived calcareous nannoplankton  $\text{CaCO}_3$  (Suchéras-Marx et al., 2012).  
447 There is an increase in the geographical distribution of radiolarian deposits (Bartolini et al.,  
448 1999) although a major radiolarian turnover is dated earlier in the middle-late Aalenian  
449 (Bartolini et al., 1999; Aguado et al., 2008). The early Bajocian is also associated with a major  
450 ammonite turnover (O'Dogherty et al., 2006). Moreover, abundant bioclastic deposits of

451 crinoids and bivalves in carbonate platforms in France and Spain point to the dominance of a  
452 filtering community in neritic environments, hence to nutrient-rich surface waters (Dromart et  
453 al., 1996; Thiry-Bastien, 2002; Aurell et al., 2003; Brigaud et al., 2009; Molina et al., 2018).  
454 Finally, the Chaudon-Norante succession records abundant *Zoophycos* that likely also reflects  
455 higher organic matter influx to the sediment (Olivero, 1994). Overall, these paleontological data  
456 suggest that strata recording the lower Bajocian positive CIE were deposited during an interval  
457 of increased primary productivity. Such an important increase in productivity would have been  
458 logically sustained by higher siliciclastic and phosphorus inputs from weathering or increase  
459 of widespread upwelling in the western Tethys. The former hypothesis is in line with the major  
460 rise of siliciclastic AR and PAR recorded at Murtinheira (Fig. 5), but at odds with the Chaudon-  
461 Norante data (Fig. 6) showing a small increase in both siliciclastic AR and C<sub>org</sub> AR and almost  
462 no change in PAR, which could explain the relative low increase in Nannofossil AR. Thus, even  
463 if paleontological data suggest a higher primary productivity during the early Bajocian, the  
464 fertilization model from continental weathering does not seem fully compatible with the new  
465 sedimentological and geochemical data presented here. Conversely, the fertilization could be  
466 related to upwelling development in the western Tethys. However, our data cannot  
467 unambiguously discriminate between the upwelling model for explaining the ocean fertilization  
468 from other mechanisms. According to a previous study on radiolarites, the Western Tethys and  
469 Alpine Tethys did not see the development of upwelling-related high productivity, and  
470 information about North Atlantic are not available (Baumgartner, 2013).

471 The lower Bajocian successions investigated in this paper and in previous works (Raucsik,  
472 1999; Bodin et al., 2020) have low to very low TOC contents and C<sub>org</sub> AR (Figs. 2 and 3, 5 and  
473 6). Furthermore, lower Bajocian strata are unnoticeable in terms of occurrence of petroleum  
474 source rock (Kendall et al., 2009). Nevertheless, a stratigraphically thin (~10 cm) black shale  
475 level ('Gaetani level') at Alpe Turati in Italy has been reported (Erba et al., 2019), which may  
476 stratigraphically correspond to part of the increasing phase of the positive CIE. Moreover,  
477 "black flysch" have been dated as early Bajocian in Poland, and lower Bajocian black shales  
478 have been reported in Alaska (Imlay, 1976; Barski et al., 2012). To our knowledge, however,

479 the lower Bajocian strata do not yield widespread black shales levels. Importantly, the  $C_{org}/P_{tot}$   
480 molar ratios in both studied sites are always lower than the Redfield ratio 1:106 (Figs. 5 and  
481 6), which suggest overall stable, oxic conditions throughout the deposition of the corresponding  
482 strata (Suan et al., 2012). This is also coherent with geochemical results from La Losillas  
483 section in Spain, which suggest fluctuating but overall oxic conditions during the early Bajocian  
484 (Molina et al., 2018). Although investigation of oxygenation conditions and rates of carbon  
485 burial in extra-Tethyan sites is warranted, all these data are thus at odds with the hypothesis  
486 of increased organic matter preservation under poor oxygenation as the main trigger of the  
487 Bajocian CIEs, as proposed for some comparable negative and positive CIEs associated with  
488 OAEs.

489

#### 490 5.4.2. Carbonate crisis

491 Alternatively, the positive CIE could be related to a major neritic carbonate crisis, which would  
492 have decreased the burial of relatively  $^{13}C$ -enriched material (Bartolini and Cecca, 1999; Bodin  
493 et al., 2017). This hypothesis is not supported by our new records showing no major decrease  
494 in  $CaCO_3$  AR in both sections studied here (Figs. 5 and 6; regardless of the age model used,  
495 see SupplFigs. 1-2; Gradstein et al., 2020). However, the carbonate factory was dominated by  
496 neritic production by the Middle Jurassic (Suchéras-Marx et al., 2012) thus by carbonate  
497 platforms. Hence, the state of productivity in such environments is more critical to evaluate the  
498 impact of carbonate burial on the lower Bajocian carbon cycle. In eastern France, the early  
499 Bajocian was a period a medium  $CaCO_3$  accumulation rates (Dromart et al., 1996). A published  
500 compilation of carbonate platforms sedimentation rates does show a peculiar carbonate crisis  
501 (Andrieu et al., 2016). The early Bajocian seemed to be an intermediate state between an  
502 Aalenian (or upper Aalenian) carbonate production low and a upper Bajocian high (Dromart et  
503 al., 1996; Andrieu et al., 2016; Fantasia et al., 2022). This crisis, firstly proposed in Italian  
504 platforms (Bartolini and Cecca, 1999), is evidenced by a reduction in  $CaCO_3$  accumulation  
505 rates in the High Atlas of Morocco (Bodin et al., 2017) and Provence platform of SE France  
506 (Léonide et al., 2007). In these regions and in many other carbonate platforms, abundant

507 crinoids and, to a lesser extent, other filtering organisms are common to dominant carbonate  
508 producers during the early Bajocian (e.g. Burgundy-Ardèche, France, Dromart et al., 1996;  
509 Lusitanian basin, Portugal, Azerêdo, 1998; Jura, France, Thiry-Bastien, 2002; Basque-  
510 Cantabrian and Iberian basins, Spain, Aurell, 2003; Betic basin, Spain, O'Dogherty et al., 2006;  
511 Molina et al., 2018; United Arab Emirates, Hönig and John, 2015; northwestern Paris basin,  
512 France, Andrieu et al., 2016). This carbonate fossils are associated in many localities with  
513 ooids (e.g. Burgundy-Ardèche, France, Dromart et al., 1996; Lusitanian basin, Portugal,  
514 Azerêdo, 1998; Jura, France, Thiry-Bastien, 2002; United Arab Emirates, Hönig and John,  
515 2015; northwestern Paris basin, France, Andrieu et al., 2016). Thus, carbonate production in  
516 neritic environments was mixed between heterozoan and microbial production (Andrieu et al.,  
517 2016) and dominated by grains rather than mud. At Murtinheira and Chaudon-Norante, the  
518 carbonate fraction is dominated by mud (Pavia, 1983; Suchéras-Marx et al., 2012) but those  
519 pelagic sections are the far reaching end of the carbonate export thus too far to receive grains.  
520 Some of the localities recording a marked decrease in carbonate accumulation are also  
521 characterized by a relative sea-level rise that could explain local carbonate crisis due to  
522 drowning (e.g. Provence, France, Léonide et al., 2007; Morocco, Bodin et al., 2017). The effect  
523 of the drowning could have been more effective due to environmental perturbation such as  
524 eutrophication. Those conditions may have then favor heterozoan over photozoan platforms.  
525 However, heterozoan platforms are less efficient in carbonate production and thus may have  
526 been incapable to keep-up. Nevertheless, this carbonate crisis was not global and did not shut  
527 down all type of neritic carbonate production and export to the basin. Hence, a carbonate crisis  
528 may have contributed to the lower Bajocian positive CIE but was certainly not its main driver.

529

#### 530 5.4.3. Data-model comparison

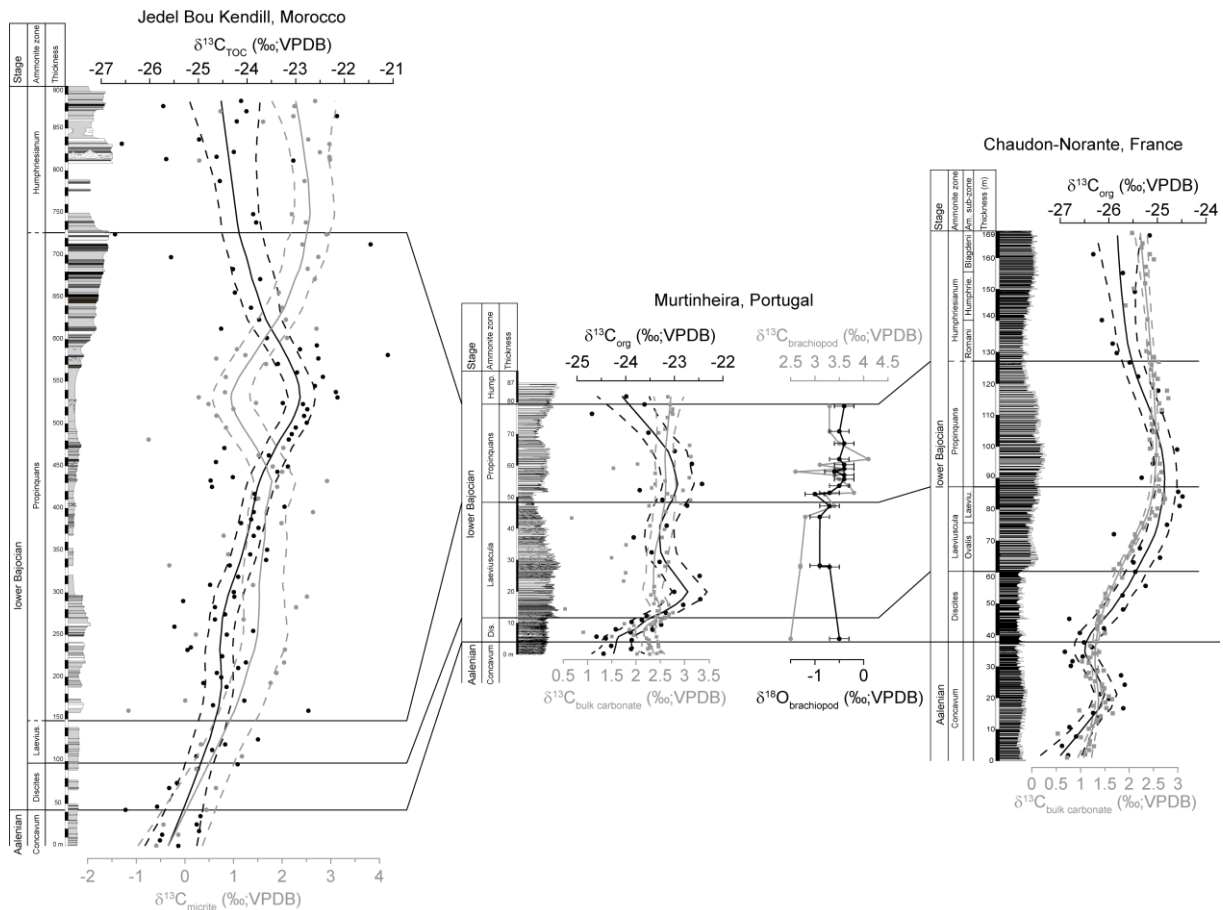
531 Comparing the  $\delta^{13}\text{C}_{\text{bulk carbonate}}$  and  $\delta^{13}\text{C}_{\text{org}}$  in different locality may help to further constrain the  
532 possible causes of the positive CIE using the box models proposed by Kump and Arthur  
533 (1999). The stratigraphic changes in  $\delta^{13}\text{C}_{\text{bulk carbonate}}$  vs  $\delta^{13}\text{C}_{\text{org}}$  seem to differ substantially  
534 between localities related to local conditions and differences in carbon sources, as mentioned

535 in section 5.1. However, there are two patterns observed in the three sections shown in Fig. 7,  
536 where the  $\delta^{13}\text{C}_{\text{org}}$  profile systematically exhibits a slightly larger positive CIE and an earlier  
537 return to pre-excursion values (basal Propinquans ammonite Zone) than that recorded by  
538  $\delta^{13}\text{C}_{\text{bulk carbonate}}$ . Such a decoupling is incompatible with a simple model of productivity increase  
539 driven by a sole increase in phosphate delivery rate to the ocean, which predict a synchronous  
540 positive CIE in  $\delta_{\text{org}}$  and  $\delta_{\text{carb}}$  (model fig. 3 in Kump and Arthur, 1999). Our results and published  
541 data seem more consistent with a model of coupled increase in rate of riverine phosphate and  
542 weathered carbon delivery to the oceans (model fig. 9 in Kump and Arthur, 1999). In the model,  
543 the negative CIE results from the enhanced delivery of  $^{13}\text{C}$ -depleted carbon from weathering,  
544 whereas the increase in organic carbon burial is rather related to the combined effects of higher  
545 riverine phosphate delivery initiating the positive CIE and a  $\text{pCO}_2$  decrease. Consequently, this  
546 version of the model predicts first a  $\sim -0.5\text{‰}$  negative CIE similar to that observed across the  
547 Aalenian-Bajocian boundary followed by a  $\delta_{\text{org}} \sim 1.5\text{‰}$  positive CIE synchronous with a  $\delta_{\text{carb}}$   
548  $\sim 1\text{‰}$  positive CIE. Thus, like for the lower Bajocian positive CIE, the CIE amplitude is higher  
549 in  $\delta_{\text{org}}$  than in  $\delta_{\text{carb}}$ . Finally, the  $\delta_{\text{org}}$  tends in the model to start decreasing back to pre-CIE  
550 values earlier than  $\delta_{\text{carb}}$ , as observed in our sections.

551 The modelled is driven by a pulse of increased organic matter burial, for which, as mentioned  
552 earlier, there is evidence in the early Bajocian. Alternatively, an increase in organic carbon  
553 accumulation might have occurred in basins that have not been investigated for their TOC  
554 contents or that have since then disappeared. Such an organic matter burial forces a decrease  
555 in  $\text{pCO}_2$  that would induce a cooling because of counter-greenhouse conditions. This  
556 alternative prediction is in line with the  $\sim 120$  ppm  $\text{pCO}_2$  decrease previously calculated for the  
557 lower Bajocian between the Aalenian-Bajocian boundary and the Propinquans ammonite Zone  
558 (Bodin et al., 2020). This is also in line with the paleotemperature estimates based on  $\delta^{18}\text{O}$  of  
559 brachiopods at Murtinheira, which suggest a  $2.5^\circ\text{C}$  cooling (i.e.,  $\sim 0.5\text{‰}$  increase in  $\delta^{18}\text{O}$ ;  
560 Ferreira et al., 2019) of bottom waters in the Propinquans ammonite Zone (Fig. 7). The isotopic  
561 data are not perfectly in line with the model in terms of magnitude, which depends on input  
562 rates and  $\text{pCO}_2$  decrease; besides, according to this model a temperature decrease should



563 rather have occurred in the Discites and Laeviuscula ammonite zones. This discrepancy could  
564 be explained by a potential CO<sub>2</sub> increase at the Aalenian-Bajocian boundary, as suggested by  
565 stomatal data from the Yorkshire coast (Hesselbo et al., 2003), which would have delayed the  
566 temperature decrease, or simply by the relatively low resolution of brachiopod data in this  
567 interval. According to bivalve  $\delta^{18}\text{O}$  data from France, bottom water temperatures increased  
568 between Propinquans and Humphriesianum ammonite zones (Brigaud et al., 2009), which  
569 could support an increase in pCO<sub>2</sub> following the event of enhanced productivity and burial of  
570 organic matter (Kump and Arthur, 1999). This 'cool snap' echoes previous observations based  
571 on fossil wood and glendonite occurrences (Philippe and Thevenard, 1996; Price, 1999; Rogov  
572 and Zakharov, 2010) suggesting a cooling in the Bajocian *sensu lato*, which would require  
573 further stratigraphic refinement. The CO<sub>2</sub> release at the Aalenian-Bajocian boundary related to  
574 global plate tectonic and Pacific Ocean plate production (Bartolini and Larson, 2001; Labails  
575 et al., 2010) may have induced a rapid warming (Gomez et al., 2009; Korte et al., 2015; Ferreira  
576 et al., 2019) at the origin of the increase in the rate of phosphate and weathering carbon release  
577 to the oceans from localized riverine input. However, the rate of delivery of limiting elements  
578 fertilizing parts of the oceans was slow enough to not have any damaging impact on the oceans  
579 like during OAE 2 (i.e., Cenomanian-Turonian boundary). On the contrary, the fertilization  
580 promoted diversification and productivity of plankton in the lower Bajocian event (Bartolini et  
581 al., 2019; Giraud et al., 2016). The Aalenian to lower Bajocian CIEs are very similar to the  
582 Weissert event in term of duration (Martinez et al., 2015), productivity increase (Gréselle et al.,  
583 2011; Duchamp-Alphonse et al., 2014; Mattioli et al., 2014; Shmeit et al., 2022) and possible  
584 cooling event (Barbarin et al., 2012; Cavaheiro et al., 2021). Thus, a similar cause may be the  
585 most probable origin of both these events although it needs to be identified (e.g. volcanic  
586 activity proposed by Erba et al. (2004) but excluded by Shmeit et al. (2023)).



587

588 Fig. 7:  $\delta^{13}\text{C}_{\text{bulk carbonate}}$  (grey squares; labelled  $\delta^{13}\text{C}_{\text{micrite}}$  in Morocco following Bodin et al., 2017)  
 589 versus  $\delta^{13}\text{C}_{\text{org}}$  (black circles; labelled  $\delta^{13}\text{C}_{\text{TOC}}$  in Morocco following Bodin et al., 2017) at Jedel  
 590 Bou Kendill (Morocco), Murtinheira (Portugal) and Chaudon-Norante (France) following a SW-  
 591 NE transect. At Murtinheira,  $\delta^{13}\text{C}_{\text{brachiopod}}$  (‰, VPDB; grey squares) and  $\delta^{18}\text{O}_{\text{brachiopod}}$  (‰, VPDB;  
 592 black circles) from Ferreira et al (2019) are plotted. The error bars for  $\delta^{18}\text{O}_{\text{brachiopod}}$  are  $\pm 2\sigma$   
 593 analytical uncertainty.

594

## 595 6. Conclusions

596 The early Bajocian was a time interval of important biologic, tectonic and climatic perturbations  
 597 that have been clearly overseen because marine strata deposited during this event lack  
 598 conspicuous black shale levels and evidence for anoxia. This time interval is also marked by  
 599 a carbon cycle perturbation in the form of a long-lasting  $\delta^{13}\text{C}$  positive excursion over the whole  
 600 lower Bajocian. In France at Chaudon-Norante and in Portugal at Murtinheira, the lower  
 601 Bajocian  $\delta^{13}\text{C}$  positive excursion was previously recorded on bulk carbonate and biogenic

602 calcite and is now also recorded on organic matter. This positive excursion reflects a long-lived  
603 and marked carbon cycle perturbation likely driven by an increase in organic carbon burial.  
604 The studied succession, however, indicate stable organic matter burial fluxes and deposition  
605 under stable oxygenation conditions, whereas an increase in erosion and productivity is  
606 supported by an increase in siliciclastics AR and PAR only in Murtinheira. The patterns seen  
607 in  $\delta^{13}\text{C}_{\text{bulk carbonate}}$  and  $\delta^{13}\text{C}_{\text{org}}$  are consistent with those previously described in box model  
608 simulating an increase in both riverine flux of phosphorus and inorganic carbon. The Bajocian  
609 carbon cycle perturbations fit particularly this solution combining both negative and positive  
610 excursions and a cooling period linked to  $\text{pCO}_2$  decrease related to an increase in primary  
611 productivity. However, this model is clearly too simplistic. There are regional variations in  
612 carbonate production with different producers on carbonate platforms (e.g., crinoids,  
613 microbialite, scleractinian) between platforms and in biological production with different marine  
614 organic producers and amount produced. Those geographical variations complexify the view  
615 of the global carbon and environmental perturbation based on too few sites of study.  
616 The lower Bajocian event was clearly different from the well-known Cenomanian-Turonian  
617 boundary  $\delta^{13}\text{C}$  positive excursion and bears more similarities with the lower Valanginian  $\delta^{13}\text{C}$   
618 positive excursion characterizing the Weissert event. Further geochemical data and especially  
619 more paleotemperature estimates and new sediment data ( $\text{CaCO}_3$ , TOC, P contents) outside  
620 the Tethys Ocean are needed to refine the Bajocian carbon cycle perturbation model.

621

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627

#### 628 Authors' contributions

629 BSM designed the study based on recommendations from FG, EM, GS and TA. BSM  
630 performed the phosphorus analyses in TA's lab at UNIL under the supervision of HMK. AF and  
631 JES performed the  $\delta^{13}\text{C}_{\text{org}}$  analyses at UNIL. JCM prepared the organic matter samples at  
632 CEREGE and GS performed the observations. BSM and GS interpreted the results. BSM and  
633 GS wrote the manuscript with contributions from FG, EM, HMK et AF.

634

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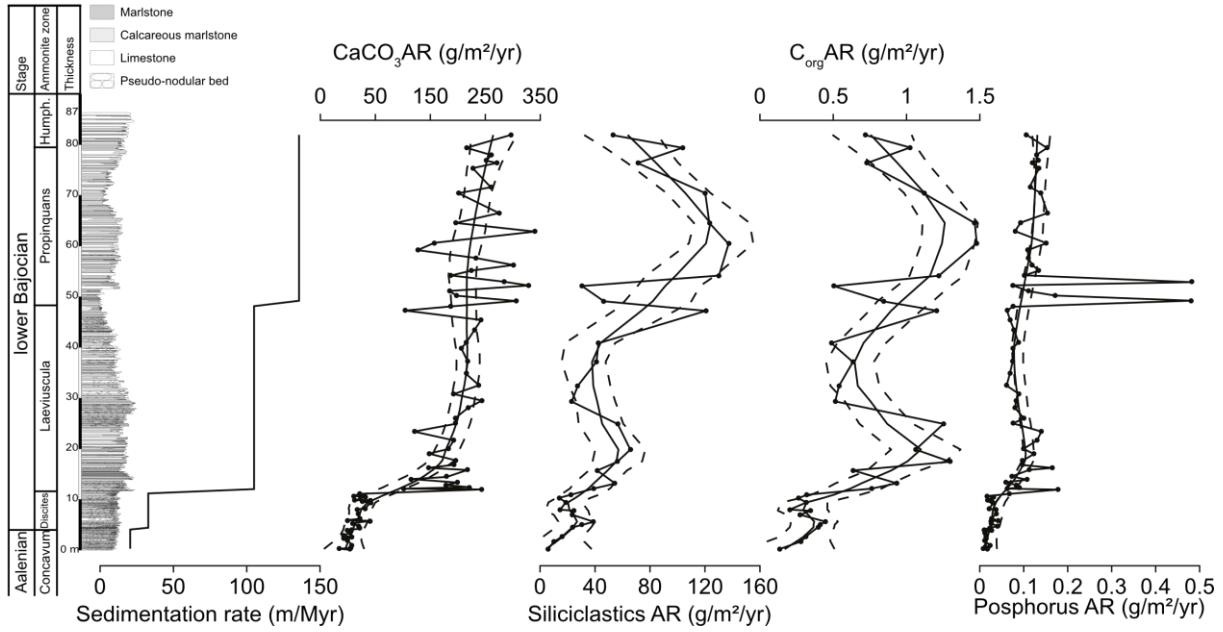
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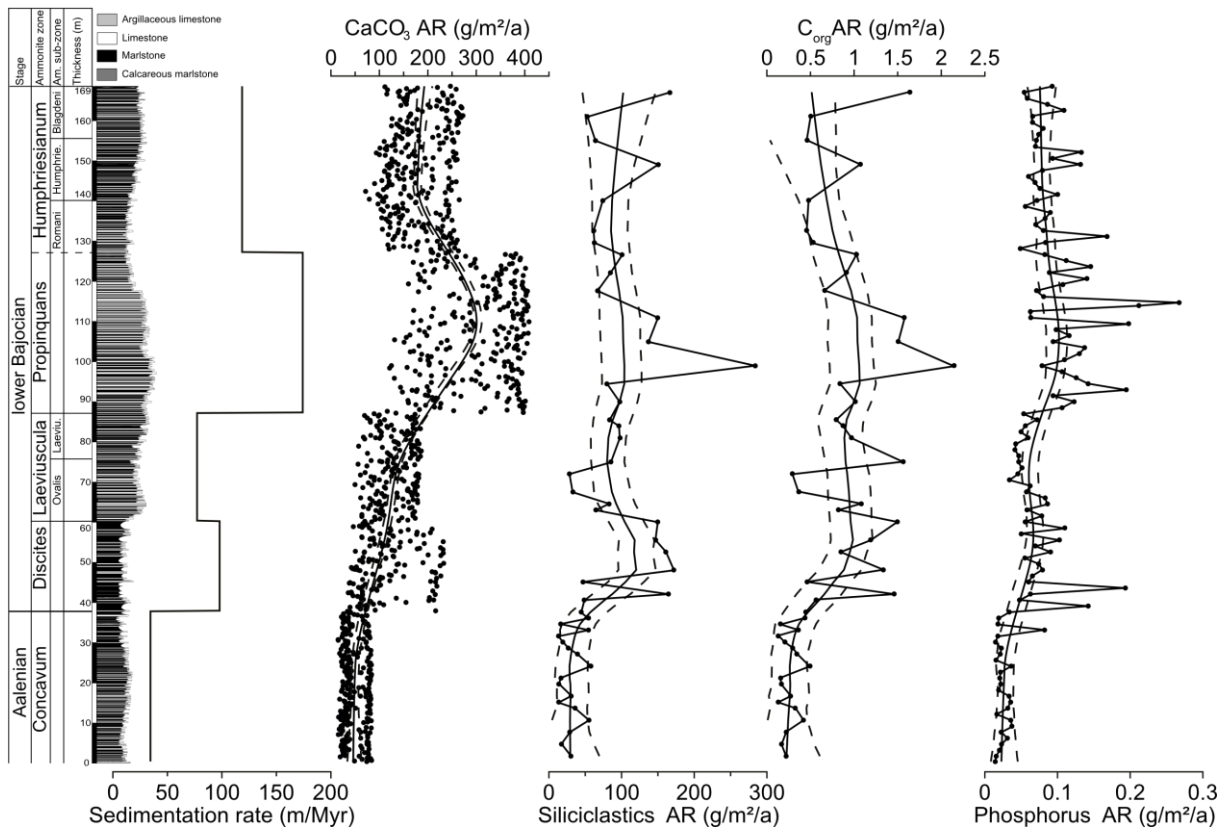


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983 Supplementary Fig. 1: Stratigraphic changes in sedimentation rate at Murtinheira (based on  
 984 Gradstein et al. (2020) age model) and the resulting CaCO<sub>3</sub> accumulation rates (AR; g/m<sup>2</sup>/yr),  
 985 Siliciclastics AR (g/m<sup>2</sup>/yr), C<sub>org</sub> AR (g/m<sup>2</sup>/yr), and phosphorus AR (g/m<sup>2</sup>/yr).

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989 Supplementary Fig. 2: Stratigraphic changes in sedimentation rate at Chaudon-Norante  
990 (based on Gradstein et al. (2020) age model) and the resulting CaCO<sub>3</sub> accumulation rates (AR;  
991 g/m<sup>2</sup>/yr), Siliciclastics AR (g/m<sup>2</sup>/yr), C<sub>org</sub> AR (g/m<sup>2</sup>/yr), and phosphorus AR (g/m<sup>2</sup>/yr).