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## Climatic and palaeoceanographic changes during the Pliensbachian (Early Jurassic) inferred from clay mineralogy and stable isotope (C-O) geochemistry (NW Europe).

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**TITLE**

Climatic and palaeoceanographic changes during the Pliensbachian (Early Jurassic) 2 inferred from clay mineralogy and stable isotope (C-O) geochemistry (NW Europe)

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1 **Climatic and palaeoceanographic changes during the Pliensbachian (Early Jurassic)**  
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16  
17 Abstract

18 The Early Jurassic was broadly a greenhouse climate period that was punctuated by short warm  
19 and cold climatic events, positive and negative excursions of carbon isotopes, and episodes of  
20 enhanced organic matter burial. Clay minerals from Pliensbachian sediments recovered from  
21 two boreholes in the Paris Basin, are used here as proxies of detrital supplies, runoff conditions,  
22 and palaeoceanographic changes. The combined use of these minerals with stable isotope data

23 (C-O) from bulk carbonates and organic matter allows palaeoclimatic reconstructions to be  
24 refined for the Pliensbachian. Kaolinite/illite ratio is discussed as a reliable proxy of the  
25 hydrological cycle and runoff from landmasses. Three periods of enhanced runoff are  
26 recognised within the Pliensbachian. The first one at the Sinemurian-Pliensbachian transition  
27 shows a significant increase of kaolinite concomitant with the negative carbon isotope  
28 excursion at the so-called Sinemurian Pliensbachian Boundary Event (SPBE). The Early/Late  
29 Pliensbachian transition was also characterised by more humid conditions. This warm interval  
30 is associated with a major change in oceanic circulation during the Davoei Zone, likely  
31 triggered by sea-level rise; the newly created palaeogeography, notably the flooding of the  
32 London-Brabant Massif, allowed boreal detrital supplies, including kaolinite and chlorite, to be  
33 exported to the Paris Basin. The last event of enhanced runoff occurred during the late  
34 Pliensbachian (Subdonosus Subzone of the Margaritatus Zone), which occurred also during a  
35 warm period, favouring organic matter production and preservation. Our study highlights the  
36 major role of the London Brabant Massif in influencing oceanic circulation of the NW European  
37 area, as a topographic barrier (emerged lands) during periods of lowstand sea-level and its  
38 flooding during period of high sea-level. This massif was the unique source of smectite in the  
39 Paris Basin. Two episodes of smectite-rich sedimentation ('smectite events'), coincide with  
40 regressive intervals, indicating emersion of the London Brabant Massif and thus suggesting that  
41 an amplitude of sea-level change high enough to be linked to glacio-eustasy. This mechanism  
42 is consistent with sedimentological and geochemical evidences of continental ice growth  
43 notably during the Latest Pliensbachian (Spinatum Zone), and possibly during the Early  
44 Pliensbachian (late Jamesoni/early Ibex Zones).

45

46 Keywords

47 Early Jurassic, Pliensbachian, runoff, clay minerals, stable isotope, glacio-eustasy

## 48 1. Introduction

49           The Early Jurassic was a period of major environmental and biological changes  
50 contemporaneous with the break-up of Pangaea and intense volcanic phases linked to the  
51 CAMP (Central Atlantic Magmatic Province) and the Karoo-Ferrar large igneous provinces  
52 (Little and Benton, 1995; Wignall, 2001; Wignall et al., 2005; Jourdan et al., 2008; Schaltegger  
53 et al., 2008; Dera et al., 2011a; Guex et al., 2016). During the Early Toarcian, prominent organic  
54 matter (OM) accumulations in marine sediments, associated with anoxic conditions, were  
55 recorded in many parts of the world, defining the well-known and extensively studied Toarcian  
56 Oceanic Anoxic Event (T-OAE). A remarkable positive shift of the  $\delta^{13}\text{C}$  values was triggered  
57 by OM storage in oceanic sediments (Jenkyns, 1988, 2010; Al-Suwaidi et al., 2010; Suan et al.,  
58 2011; Izumi et al., 2012; Krencker et al., 2014). Within this positive carbon isotope excursion  
59 a strong negative carbon isotopic excursion (CIE) of -7‰ in average amplitude has been  
60 recognised in terrestrial organic matter and in marine sediments (Hesselbo et al., 2000a;  
61 Schouten et al., 2000; Kemp et al., 2005; van de Schootbrugge et al., 2005; Hesselbo et al.,  
62 2007; Hesselbo and Pieńkowski, 2011). Intense volcanic activity from the Karoo-Ferrar  
63 magmatic province coupled with an increase of runoff conditions and temperature triggering  
64 oceanic methane injection and consequently light carbon release is a prominent suggestion to  
65 explain this negative excursion (Hesselbo et al., 2000a; Hesselbo et al., 2007; Suan et al., 2011;  
66 Hermoso and Pellenard, 2014). New data from the Pliensbachian interval show that two other  
67 excursions in the  $\delta^{13}\text{C}$  biogenic carbonate and organic matter content occurred: 1) a negative  
68 excursion (-2‰) at the Sinemurian-Pliensbachian boundary, namely the Sinemurian-  
69 Pliensbachian Boundary Event (SPBE) 2) a positive excursion (+2‰) during the Late  
70 Pliensbachian corresponding to the Late Pliensbachian Event (Korte and Hesselbo, 2011; Silva  
71 et al., 2011; Franceschi et al., 2014; Silva and Duarte, 2015; Gómez et al., 2016; Peti et al.,  
72 submitted). Most of these events are coeval with variations of seawater temperatures calculated

73 from  $\delta^{18}\text{O}$  values of bulk carbonate and calcified organisms. Warm periods occurred during the  
74 Late Sinemurian and the Early-Late Pliensbachian transition (e.g. IbeX to early Margaritatus  
75 Zones; Korte and Hesselbo, 2011; Silva and Duarte, 2015; Martinez and Dera, 2015; Gómez et  
76 al., 2016), while a cooler phase was recognised during the Late Pliensbachian mainly on the  
77 basis of  $\delta^{18}\text{O}$  data measured from belemnite rostra (Rosales et al., 2004; Dera et al., 2009b;  
78 Korte and Hesselbo, 2011; Martinez and Dera, 2015; Gómez et al., 2016), brachiopods and  
79 bivalves shells (Suan et al., 2010; Korte and Hesselbo, 2011) and from sedimentological and  
80 palynological evidences (Price, 1999; Suan et al., 2011). Together with available  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$   
81 data, clay minerals could be used as an informative palaeoclimatic proxy, especially to better  
82 constrain continental weathering and humidity/aridity variations (Robert and Kennett, 1994;  
83 Vanderaveroet et al., 1999; Ruffell et al., 2002; Pellenard et al., 2014). The kaolinite/illite ratio  
84 is considered to be a reliable proxy of humid conditions during the Jurassic (Chamley, 1989;  
85 Hallam et al., 1991; Pellenard and Deconinck, 2006; Raucsik and Varga, 2008; Dera et al.,  
86 2009a; Hesselbo et al., 2009; Hermoso and Pellenard, 2014; Pellenard et al., 2014). Kaolinite  
87 contents have been used to build long-term palaeohumidity curves for the Pliensbachian-  
88 Toarcian interval (Dera et al., 2009a). At the scale of the Tethyan domain, global enrichments  
89 of kaolinite during the Davoei Zone and during the early Toarcian (Falciferum to Bifrons  
90 Zones) were interpreted as periods of enhanced runoff on landmasses, while decreasing  
91 proportions of this mineral during the late Pliensbachian, suggested a colder and drier period,  
92 in agreement with the  $\delta^{18}\text{O}$  fluctuations (Dera et al., 2009a, 2009b, 2011a; Martinez and Dera  
93 2015; Gómez et al., 2016). In more detail, a significant and brief increase of kaolinite was also  
94 identified at the base of the Serpentinum Zone, synchronous with the negative shifts in carbon  
95 isotopes ( $\delta^{13}\text{C}_{\text{org}}$  and  $\delta^{13}\text{C}_{\text{carb}}$ ), and associated with an increase of temperature measured from  
96 the  $\delta^{18}\text{O}$  bulk carbonate (Hermoso and Pellenard, 2014). Again, the enrichment of kaolinite

97 supports enhanced runoff and continental weathering during a brief warming episode and the  
98 reliability of clay minerals as sensitive climate proxies.

99         Despite these relationships between clay mineralogy and geochemical proxies in the late  
100 Pliensbachian and Toarcian, links between  $\delta^{13}\text{C}$  disruption, sea temperature change, and  
101 kaolinite enrichment still remain poorly described in the remaining Lower Jurassic. In the  
102 present study, we cover the entire Pliensbachian stage through a high-resolution analysis of clay  
103 mineralogy and carbon and oxygen isotopes in multiple sections from the NW European  
104 domain well-constrained by ammonites and/or calcareous nannofossils. Relationships between  
105 clay mineralogy,  $\delta^{13}\text{C}$  excursions, and  $\delta^{18}\text{O}$  fluctuations are carefully examined in order to  
106 decipher the role of the carbon cycle and temperature evolution on continental weathering  
107 changes. Palaeoceanographic changes, eustatic variations, and detrital sources, are also  
108 discussed in relation to palaeoclimate changes.

109

## 110 2. Geological setting

111         During the Early Jurassic, the NW Tethyan domain corresponded to the extension of the  
112 Tethys Ocean which was characterised by an archipelago, composed of large islands inherited  
113 from the Variscan orogeny surrounded by epicontinental seas (Thierry et al., 2000). The NW  
114 Tethyan domain was also connected to the Arctic Sea and the Panthalassa Ocean through the  
115 Viking and Hispanic corridors implying different water mass influences (Dera et al., 2009b).  
116 This domain is generally subdivided into two parts, with the Mediterranean area, and a NW  
117 European domain that includes the Paris and the Wessex basins where boreholes and sections  
118 analysed in this study are located (Fig. 1).

119         The Paris Basin is an intracratonic basin surrounded by four landmasses, namely the  
120 London-Brabant Massif to the north, the Armorican Massif to the west, the Central Massif to

121 the south and the Bohemian Massif to the east. Located in the southern part of the Paris Basin,  
122 to the north of the Central Massif, the Sancerre borehole (Callovian to Carboniferous) was  
123 drilled during the “Géologie Profonde de la France” programme (Deep Geology of France,  
124 Lorenz et al., 1987). From the end of the Sinemurian to the Lower Pliensbachian, sediments are  
125 dominantly clayey limestone, while the Upper Pliensbachian is marked by claystone. The  
126 Pliensbachian-Toarcian boundary is characterised by some limestone beds. The Lower  
127 Toarcian strata comprise marl and organic-rich shale beds. Previous mineralogical and  
128 geochemical investigations were performed by Delavenna et al. (1989), Hermoso et al. (2009,  
129 2012, 2013) and Hermoso and Pellenard (2014). From this core, 283 samples were analysed by  
130 X-ray diffraction.

131 The Montcornet borehole (A901 site) is located close to the Ardennes Massif in the  
132 northern part of the Paris Basin. Reaching 1100 metres in depth, it was drilled by Andra (the  
133 French National Radioactive Waste Management Agency) from Turonian chalks to the  
134 Devonian basement. The Lower Pliensbachian is composed of marl and limestone beds with  
135 pyritised burrows and phosphate nodules in the upper part. The Upper Pliensbachian comprises  
136 marl and claystone with some diagenetic siderite-rich nodules and thin accumulations of bivalve  
137 shells interpreted as tempestites. The Pliensbachian-Toarcian boundary is poorly defined  
138 biostratigraphically, but likely corresponds to the occurrence of limestone beds, coeval with the  
139 "Grès Médioliasiques" Fm, recognised in the rest of the Paris Basin and interpreted as a  
140 regressive facies (Brigaud et al., 2014). The Toarcian is characterised by locally organic-rich  
141 and laminated shale. An abundant fauna including nektonic (ammonites and belemnites) and  
142 benthic organisms (bivalves, brachiopods, crinoids) occurs throughout the core. Previous  
143 analyses have been performed on biostratigraphy, magnetostratigraphy (Yang et al., 1996;  
144 Moreau et al., 2002) and mineralogy (Debrabant et al., 1992). 197 samples have been analysed



145 for clay mineralogy and 213 samples for isotope geochemistry. In addition, 50 new specimens  
146 of ammonites have been collected to help refine the biostratigraphy.

147 Additional clay mineral data from two sections (Corbigny, South Paris Basin, France and  
148 Charmouth, Wessex Basin, U.K) have been used for further comparisons (Fig. 1).

149

### 150 3. Materials and methods

#### 151 3.1. Clay mineralogy

152 A total of 480 samples have been analysed using X-Ray Diffraction (XRD). After a  
153 gentle crush, powdered samples were decarbonated with a 0.2M HCl solution. The fraction  
154 below two microns (clay sized particles), was extracted with a syringe after decantation of the  
155 suspension during 1h35 following the Stokes law, and then centrifuged. Ammonia water was  
156 added to samples from Sancerre to help their deflocculation as recommended by Holtzapffel  
157 (1985). Clay residue was then smeared on oriented glass slides and run in a Brucker D4  
158 Endeavour diffractometer with CuK $\alpha$  radiations, LynxEye detector and Ni filter under 40 kV  
159 voltage and 25 mA intensity (Biogeosciences laboratory, University of Burgundy). Scanning  
160 from the goniometer ranges from 2.5° to 28° for each analysis. Three runs were performed for  
161 each sample to discriminate clay phases: 1) Air-drying; 2) Ethylene-glycol solvation; 3) Heating  
162 at 490°C, as recommended by Moore and Reynolds (1997). Clay minerals were identified using  
163 their main diffraction ( $d_{001}$ ) peak and by comparing the three diffractograms obtained. Clay  
164 minerals identified are the following: R0 type illite-smectite mixed-layer (17 Å based on  
165 glycolated run condition) referred to smectite for the following sections; R1 type illite-smectite  
166 mixed-layer (around 11.5 Å on air-drying conditions and 13 Å after ethylene-glycol solvation);  
167 chlorite (14.2 Å, 7.1 Å, 4.7 Å and 3.54 Å peaks); illite (10 Å, 5 Å, 3.33 Å); kaolinite (7.18 Å  
168 and 3.58 Å peaks). Estimated proportions of each clay mineral were performed using the

169 MacDiff 4.2.5 software (Petschick, 2000) on glycolated diffractograms. As the main ( $d_{001}$ ) peak  
170 of kaolinite and the ( $d_{002}$ ) peak of chlorite are overlapping, a deconvolution procedure was  
171 applied on the ( $d_{004}$ ) peak area of chlorite (3.54 Å) and ( $d_{002}$ ) peak area of kaolinite (3.57 Å) to  
172 estimate properly both minerals proportions using the 7.1 peak ( $d_{001\text{kaolinite}} + d_{002\text{chlorite}}$ ). Chlorite  
173 percentage was calculated using the mean between ( $d_{001}$ ) and ( $d_{002}$ ) chlorite peak areas  
174 considering that chemical nature in chlorite impact the ( $d_{001}$ )/( $d_{002}$ ) ratio (Moore and Reynolds,  
175 1997). The K/I ratio based on ( $d_{001}$ ) peak area of each minerals was systematically calculated.  
176 Scanning Electronic Microscope (SEM) observations were performed with a JEOL JSM 6400F  
177 Field Emission SEM (ARCEN platform of the LICB laboratory of the University of Burgundy).

### 178 3.2. Geochemical analyses

179 Isotope analyses on bulk carbonate oxygen and carbon ( $\delta^{18}\text{O}_{\text{Carb}}$  and  $\delta^{13}\text{C}_{\text{Carb}}$ ) were  
180 performed using a carbonate preparation device Kiel IV coupled with a Delta V Plus mass  
181 spectrometer (ThermoFisher) at the Biogeosciences laboratory (University of Burgundy). 130-  
182 900µg of rock were cleaned with ethanol, finely powdered and dried at 50°C for 12 hours in a  
183 stove. Carbonates were reacted with purified orthophosphoric acid at 70°C. Samples were  
184 calibrated to V-PDB using NBS-19. Replicates were regularly made to evaluate reproducibility  
185 which are respectively of 0.04‰ and 0.08‰ for  $\delta^{13}\text{C}_{\text{Carb}}$  and  $\delta^{18}\text{O}_{\text{Carb}}$ .

### 186 3.3. Statistical analyses

187 The SiZer analysis (Significance of Zero crossings of smoothed estimates e.g. Marron  
188 and Chaudhuri, 1998) was used on the K/I ratio to define significant changes in kaolinite  
189 content. This method consists on the analysis of the derivative of a smoothed curve obtained  
190 from a kernel type regression using different levels of smoothing (noted  $h$ ). SiZer method  
191 compiles derivative curves with their 95% confidence intervals. The morphology of the curves  
192 is analysed on the second derivative to identify wide patterns (bulge, hollow) and short patterns

193 (peak) suggested from the biostratigraphic framework. Results are presented on a SiZer map  
194 indicating significant changes in the curve morphology of the investigated parameter. The  
195 abscissa characterises the level of smoothing (window size  $h$  in log scale) and the ordinate the  
196 depth. Long amplitude changes in the K/I ratio are detected on large window analysis (*i.e.*  $h =$   
197  $\log(1)$ ), whereas short amplitude changes are detected on short window analysis (*i.e.*  $h = \log$   
198  $(0)$ ). Bulges are shown in red, hollows in blue. Grey surfaces are interpreted as not statistically  
199 significant.

200

## 201 4. Results

### 202 4.1 Biostratigraphic framework

203 The biostratigraphic framework has been recently revised at Sancerre (see Peti et al., in  
204 press). At Montcornet, new investigations have been conducted to complete the study of  
205 Moreau et al., (2002). All ammonites chronozones are recognised in the Pliensbachian stage  
206 except the Spinatum Zone. Ammonites and biozonation boundaries are precisely reported on  
207 Fig. 2 and synthetically reported on Figs. 3 and 4 (grey intervals are considered as intervals of  
208 uncertainties). The detail of identified ammonites and their biostratigraphic attribution are  
209 reported in the supplementary file (Suppl. Mat. Fig. 1).

210

### 211 4.2 Clay mineralogy

#### 212 4.2.1. Sancerre borehole (South Paris Basin)

213 Clay mineral assemblages from the Sancerre borehole mainly comprise kaolinite (13-  
214 50%), illite (24-57%), R1 Type illite-smectite mixed-layer (IS R1; 13-29%) and chlorite (7-  
215 24%; Fig. 3). The main features are coeval variations between: 1) kaolinite and chlorite, and;

216 2) illite and IS R1. As a consequence, kaolinite and illite contents are inversely related, and  
217 these minerals also display the greatest variations, which exceed 20% throughout the series.  
218 Therefore, the K/I ratio can be used as a reliable proxy of mineralogical changes. Based on the  
219 K/I ratio and the associated SiZer map, two major changes are observed during: 1) the Davoei  
220 Zone (Lower Pliensbachian; interval Ⓐ on Fig. 3), and; 2) the Spinatum Zone (Late  
221 Pliensbachian; interval Ⓑ on Fig. 3). Minor changes are visible: 1) around the Sinemurian-  
222 Pliensbachian boundary (interval ① on Fig. 3), and; 2) during the Margaritatus Zone, where  
223 three cycles are observed within the Stokesi (interval ② on Fig. 3) Subnodosus (interval ③ on  
224 Fig. 3) and Gibbosus (interval ④ on Fig. 3) Subzones. Both major and minor changes of the  
225 K/I ratio are observed with the SiZer analysis that statistically validate their occurrence.

#### 226 4.2.2 Montcornet borehole (North Paris Basin)

227 Similar clay mineral assemblages are recognised at Montcornet, including kaolinite (2-  
228 54%), illite (9-58%), IS R1 (0-19 %), and chlorite (5-31%; Fig. 4). Two intervals showing  
229 smectite (up to 78%) are however identified during the Ibex Zone (interval ① on Figure 4) and  
230 before and during the "Grès Médioliasiques" Fm (interval ② on Fig. 4). The K/I ratio shows  
231 three major changes during the Pliensbachian: 1) around the Ibex-Davoei transition (interval Ⓐ  
232 on Fig. 4); 2) after the Margaritatus Zone (interval Ⓑ on Fig. 4), and; 3) during the "Grès  
233 Médioliasiques" Fm (interval ③ on Fig. 4). The first two changes in the K/I ratio are validated  
234 by the SiZer analysis. The net and brief increase of the K/I ratio during the "Grès  
235 Médioliasiques" is associated to a marked lithological change expressed by the appearance of  
236 bioclastic limestones with reworked mud clasts (772.4-776.8 m), characteristic of more  
237 proximal and high-energy facies (Fig. 2). SEM observations have been performed on one  
238 bioclastic limestone bed (772.75 m) and reveal typical authigenic vermiform kaolinite in the

239 porosity (Fig. 5 A-C). Comparison has been made with one kaolinite-rich sample from  
240 claystone at 812 m, considered as representative of detrital clayey sediments, where only typical  
241 detrital shapes for clay particles have been observed, associated with rare calcitic bioclasts and  
242 framboidal pyrite (Fig. 5 D).

#### 243 *4.2.3 Additional sections (Corbigny, South Paris Basin; Charmouth, Wessex Basin)*

244 Computed K/I ratio from clay mineralogy data of the Corbigny section (see Dera, 2009)  
245 shows low values during the Jamesoni and Ibex Zones and an increase during the Davoei Zone  
246 (Fig. 6). In the Charmouth section, a low K/I ratio is also registered during the Jamesoni Zone,  
247 while a slight increase starts in the Ibex Zone (Fig. 6). No data are currently available above  
248 this level.

249

#### 250 *4.3 Carbon and oxygen isotope geochemistry (Montcornet borehole)*

251 The bulk carbonate carbon isotope ( $\delta^{13}\text{C}_{\text{carb}}$ ) signal recorded through the Pliensbachian  
252 shows several significant changes (Fig. 3). A negative excursion of at least 1‰ (from 1.1‰ to  
253 0‰) is observed between 929.4 m and 908 m from the Raricostatum Zone (Upper Sinemurian)  
254 to an undefined zone between the Jamesoni and Ibex Zones (Lower Pliensbachian). A recovery  
255 phase between 908 m and 892 m is marked by a positive trend with values which are going up  
256 from 0‰ to 1.5‰, covering the possible upper Jamesoni Zone and of the lower Ibex Zone. The  
257 Valdani Subzone (Ibex Zone) is characterised by a plateau from 892 m to 878 m with values  
258 around 1.5‰. A second positive shift around 2‰ is observed between 878 m and 869.5 m from  
259 the top of the Valdani Subzone and the top of the Ibex - lowermost part of the Davoei Zone,  
260 before values decrease to 1.5‰ between 869.5 m and 866 m. Then a first negative shift with an  
261 amplitude of 1.8‰ is observed between 866 m and 862.5 m (Maculatum Subzone, Davoei  
262 Zone) followed by a low rebound from 862.5 m to 854m with values ranging from -0.25‰ to

263 0.25‰ (Capricornus and lower Figulinum Subzones, Davoei Zone). A second strong decrease  
264 (at least -3‰) is observed between 854 m and 846 m in the Figulinum Subzone (Davoei Zone)  
265 and keep fluctuating between -0.5‰ and -4‰ to 794 m. From 794 m,  $\delta^{13}\text{C}$  values increase to  
266 0.6‰ and fluctuate up to 1.2‰ to 782 m. Then, values slightly decrease down to 0.4‰ (773 m)  
267 and increase to 1.6‰ upward. From 766m, a net decrease of values is marked by two steps with  
268 the first one going down to 0‰ and the second one to -4‰.

269 The  $\delta^{18}\text{O}_{\text{carb}}$  signal recorded from the bulk carbonate of Montcornet shows more  
270 scattered values, yet some trends are observed and detailed in the supplementary material  
271 (Suppl. Mat. Fig. 2). Values range from -6.8 to -1.7‰. No covariation is visible between carbon  
272 and oxygen isotope data.

273

## 274 5. Discussion

### 275 5.1. Influence of diagenesis

#### 276 *5.1.1 Clay mineral diagenesis*

277 Lower Jurassic deposits from the Paris Basin are recognised to have sustained a low to  
278 moderate burial diagenesis as shown by apatite fission tracks indicating a burial temperature  
279 slightly greater than 60°C (Blaise et al., 2014).

280 At Montcornet, several lines of evidence including the occurrence of R0 type illite-  
281 smectite mixed-layer (IS R0) in two intervals and mean  $T_{\text{max}}$  values below 430°C indicating  
282 immature organic matter (Disnar et al., 1996), confirm a weak influence of thermal diagenesis  
283 linked to burial. IS R0 are known to be particularly sensitive to burial diagenesis and transform  
284 into IS R1 by illitisation processes with temperatures over 60-70°C (Merriman and Frey, 1999;  
285 Lanson et al., 2009; Środoń et al., 2009; Dellisanti et al., 2010).

286 In the Sancerre borehole, the influence of burial diagenesis on clay assemblages is more  
287 questionable as IS R0 have not been identified. These minerals were either absent during  
288 sedimentation or have been transformed through incipient illitisation processes. However, mean  
289 T<sub>max</sub> values below 435°C indicate also an organic matter immaturity in the south of the Paris  
290 Basin (Hermoso et al., 2012). Consequently, as the occurrence of IS R0 is consistent with such  
291 T<sub>max</sub> values (Dellisanti et al., 2010), these minerals were likely absent during sedimentation.

292 The kaolinite/illite ratio decreases with depth in all investigated sites (Figs. 3,4 and 6).  
293 This observation could raise the question of a potential illitisation of kaolinite associated to  
294 burial diagenesis (Ruffell et al., 2002; Worden and Morad, 2003). In such cases, illitisation of  
295 kaolinite with depth is generally progressive, while in the two studied boreholes the observed  
296 decrease of kaolinite is abrupt, rather indicating a palaeoenvironmental control. In addition, in  
297 the two boreholes, the main striking feature is the coeval decrease of the K/I ratio during the  
298 same ammonite biozone (i.e. Davoei Zone; Figs. 3 and 4). If the decrease of kaolinite with depth  
299 had been triggered by burial diagenesis, alteration at the same a stratigraphic horizon in  
300 different locations would have been an unlikely outcome. We suggest therefore that in both  
301 boreholes the influence of burial diagenesis on the clay mineral assemblages is negligible.

302 The occurrence of authigenic kaolinite is open to discussion as it is commonly  
303 associated with porous rocks such as limestone and sandstone. This mineral grows in pore  
304 spaces during eo- and meso-diagenesis by flushing of meteoric waters (Lanson et al., 2002;  
305 Worden and Morad, 2003). In both boreholes, there is no obvious relationship between  
306 lithology and clay mineralogy (Figs. 3 and 4). However, in a bioclastic limestone bed at 772.75  
307 m in the Montcornet borehole, a high proportion of kaolinite (up to 55%) characterises the clay  
308 fraction (Figs. 2 and 4). Scanning Electronic Microscopy (SEM) observations revealed typical  
309 authigenic vermiform kaolinite in this sample (Fig. 5 A-C), suggesting that a large part of  
310 kaolinite content from this bed is related to fluid circulation. Other SEM observations from

311 clayey and limestone beds show common detrital shape morphologies, suggesting that kaolinite  
312 formed by authigenesis is restricted to only one porous interval (Fig. 5C).

### 313 *5.1.2 Influence of diagenesis on the isotopic signal*

314 The carbonate fraction of the Pliensbachian marl and claystone of the Montcornet  
315 borehole is a mixture of cemented micro-crystalline calcite (micrite) and thin bioclasts; these  
316 were avoided in the process of microsampling. Measured isotopic composition of calcite in bulk  
317 rocks ranges from -4.2 to 2.7 ‰ for the  $\delta^{13}\text{C}$  and from -6.8 to -1.7 ‰ for the  $\delta^{18}\text{O}$ , and are  
318 consistent with values previously published from other Pliensbachian and Toarcian sections  
319 (Jenkyns et al., 2002; Silva et al., 2011; Léonide et al., 2012; Hermoso et al., 2013; Duarte et  
320 al., 2014). In Jurassic claystones, characterised by poor permeability and low burial  
321 temperatures, the primary carbon isotope signal is frequently well-preserved during early and  
322 late diagenetic processes (e.g. Hermoso et al., 2009; Lavastre et al., 2011; Pellenard et al.,  
323 2014). Similar coeval  $\delta^{13}\text{C}_{\text{carb}}$  fluctuations recognised in other Pliensbachian series well-dated  
324 by ammonites (e.g. Silva et al., 2011; Duarte et al., 2014) highlight the negligible influence of  
325 diagenesis. However, scattered and anomalous negative values of  $\delta^{13}\text{C}_{\text{carb}}$  are observed  
326 throughout a very argillaceous interval between 789 m and 862 m, corresponding to the Late  
327 Pliensbachian (Davoei and Margaritatus Zones). This interval contains frequent red nodules of  
328 siderite with occasional Zn-mineralisation (sphalerite), (Fig. 2) likely due to reducing  
329 conditions occurring during early diagenesis, even if late fluid circulations cannot be ruled out.  
330 The disturbed  $\delta^{13}\text{C}_{\text{carb}}$  signal obtained here which prevents the use of bulk carbonate carbon  
331 isotope values for palaeoenvironmental reconstructions is mainly explained by low carbonate  
332 contents.

333 The  $\delta^{18}\text{O}_{\text{carb}}$  values in the Montcornet borehole are scattered, particularly in the 789 m  
334 to 862 m siderite-nodule-bearing interval indicating thorough fluid circulation.. Trends are yet



335 observed at the base and the top of the borehole with higher values between 905 m and 890 m  
336 (base Ibex Zone) and 790 m and 775 m (Spinatum Zone?), which may suggest cooler  
337 temperatures during these two intervals. Nevertheless, bulk oxygen isotope data have to be used  
338 with caution here as they are much more sensitive to diagenetic effects than carbon isotope data  
339 (Marshall, 1992; Jenkyns et al., 2002).

340

## 341 5.2. Significance of the clay mineral record

342 Clay mineral assemblages are composed of a mixture of illite, IS R1, occasional IS R0,  
343 kaolinite and chlorite, and this composition is a very common feature of Early Jurassic  
344 sediments from European basins (Delavenna et al., 1989; Deconinck and Bernoulli, 1991;  
345 Debrabant et al., 1992; Duarte, 1998; Deconinck et al., 2003; Jeans, 2006; Raucsik and Varga,  
346 2008; Dera et al., 2009a; Brański, 2012; Lézin et al., 2013; Hermoso and Pellenard, 2014).  
347 These clay minerals are considered to be chiefly inherited from detrital sources, because burial  
348 diagenesis is low and authigenesis only observed in one porous interval. No evidence of major  
349 tectonic rejuvenation of landmasses is reported for the Pliensbachian in the Paris Basin, which  
350 is thought to have been broadly a quiescent period in an extensional subsidence context (Brunet  
351 and Le Pichon, 1982; Guillocheau et al., 2000; Beccaletto et al., 2011). Consequently, clay  
352 mineral supplies were likely controlled by variations in eustasy, climate, and sediment source  
353 area.

354 Clay mineralogy is partly dependent on the nature of weathered rocks outcropping on  
355 emerged landmasses, where it may originate from either contemporaneous soils or old  
356 sediments. Although proximal continental areas were the main sources, more distal sources  
357 could also be expected, because clay minerals are transportable over long distances by marine  
358 or aeolian currents. The main landmasses surrounding the Paris and the Wessex basins were the

359 London-Brabant Massif to the north prolonged by the Rhenish Massif eastward, the Bohemian  
360 Massif to the east, the Massif Central to the south prolonged by the Armorican Massif  
361 westward, the Cornubian-Welsh High and Irish Massif to the west and the Scottish Massif to  
362 the northwest (Figs. 1 and 7). Erosion of these emerged lands, mainly composed of Palaeozoic  
363 igneous, metamorphic and sedimentary rocks, would have favoured inputs of primary clay  
364 minerals such as illite or chlorite. Weathering of the crystalline basement, may have also  
365 provided IS R1, smectite (IS R0) and kaolinite. This latter mineral may be reworked from the  
366 basement sedimentary rocks together with illite and chlorite (Deconinck et al., 2003; Dera et  
367 al., 2009a).

#### 368 *5.2.1 Origin of smectite*

369 Interestingly, smectite is only reported in the Montcornet borehole for the Ibex Zone  
370 and around the Pliensbachian-Toarcian transition. Differential sedimentation of clay minerals  
371 is responsible for the increase of kaolinite, commonly observed during regression phases in  
372 association with proximal facies, whereas smectite and mixed-layer minerals are more  
373 frequently recorded during associated with maximum flooding surfaces (Chamley, 1989;  
374 Ruffell et al., 2002; Pellenard and Deconinck, 2006; Godet et al., 2008). Inferred large-scale  
375 transgressive-regressive (T/R) facies cycles from European domains (Hardenbol et al., 1998;  
376 Jacquin et al., 1998) are reproduced on Figures 3 and 4. These so-called ‘second-order’ order  
377 T/R cycles (T4/R4, T5/R5) match well with the sedimentological pattern and depositional  
378 environments observed through the two boreholes, in particular for cycles defined in the  
379 Tethyan domains. A maximum flooding surface is reported within the lower Margaritatus Zone  
380 (Stokesi Subzone). Regressive maxima are at the top of the Ibex Zone for the Tethyan domains  
381 (or at the top of the Davoei Zone for the Boreal domains) and the top of the Spinatum Zone  
382 (Pliensbachian/Toarcian boundary). The Spinatum Zone was not recognised in the Montcornet  
383 borehole, but the bioclastic limestone beds between 772 and 777 m are attributed to an

384 equivalent of the "Grès Médioliasiques Fm" well known throughout the Paris Basin, dated to  
385 the Spinatum Zone and corresponding to a regressive maxima (Hermoso et al., 2013; Brigaud  
386 et al., 2014). Surprisingly, these regressive maxima correspond to the two smectite-rich  
387 intervals with proportions reaching more than 50% (Fig. 4). Smectite was exclusively restricted  
388 to the northern part of the Paris Basin until the Late Jurassic (Debrabant et al., 1992; Pellenard  
389 and Deconinck, 2006), which suggests it likely originated from the London-Brabant and  
390 Rhenish massifs. Compared to other massifs inherited from the Variscan orogeny, the London-  
391 Brabant and Rhenish-Eiffel massifs including the Ardennaise platform should constitute  
392 moderate and flat relief during the Jurassic. This morphology was favourable to the formation  
393 of smectite-rich soils under semi-arid to temperate climates with strong seasonality (Chamley,  
394 1989; Paquet and Clauer, 1997), whereas illite, chlorite and IS R1 could be formed on higher  
395 relief with steeper slopes and less pronounced weathering. A reported regional uplift with  
396 moderate average speed of unroofing of the London-Brabant and Rhenish-Eiffel massifs  
397 throughout the Jurassic (Vercoutere and Van den Haute, 1993) may also have played a role for  
398 denudation of relief and the peneplanation favourable for the storage of smectites. During  
399 regressive phases, the smectite-rich soils from low relief landmass and previously smectite-rich  
400 sediments stored on platforms were preferentially eroded, and are hypothesized to have  
401 supplied adjacent areas of the basin (Fig. 7A). By contrast, during sea-level highstand, flooding  
402 of the flat London-Brabant Massif prevented smectite-rich sedimentation in the Paris Basin  
403 (Fig. 7B). This uncommon relationship between smectite sedimentation and low sea-level is  
404 only observed in the north of the Paris Basin and the Boulonnais area from the Callovian to the  
405 Tithonian (Schnyder et al., 2000; Pellenard and Deconinck, 2006; Deconinck and Baudin, 2008;  
406 Hesselbo et al., 2009). The present study shows that as early as the Pliensbachian, the London-  
407 Brabant Massif exhibited a flat morphology by comparison with other emerged landmasses.  
408 Eustatic sea-level variations can thus explain here some regional clay mineralogical features,

409 but is insufficient to explain the several kaolinite changes, correlated between the four studied  
410 sites.

### 411 *5.2.2 Significance of the K/I ratio*

412 The K/I ratio shows two main significant changes at the investigated localities: 1) a  
413 sharp increase at the transition between the Ibex and Davoei Zones, and; 2) a decrease in the  
414 Spinatum Zone (Fig. 6). The abrupt increase of the K/I ratio at the base of the Davoei Zone  
415 coincides with the onset of the T5 2<sup>nd</sup> order transgressive cycle (Tethyan domains) and remains  
416 high up to the top of the R5 regressive cycle (Figs. 3 and 4). Looking at large scale trends of  
417 K/I ratio in the Sancerre and Montcornet boreholes, regressive facies trends are not  
418 characterised by an increase of kaolinite but rather by a decrease (Ibex and Spinatum Zones),  
419 and no evidence linking kaolinite maxima and regressive maxima can be identified. Kaolinite  
420 variations and inferred K/I ratio are unlikely to be forced by a unique eustatic control.

421 Clay minerals are known to reflect climate through continental weathering and runoff  
422 in modern environments and in deep time (Chamley, 1989; Hallam et al., 1991; Paquet and  
423 Clauer, 1997; Thiry, 2000; Dera et al., 2009a). Kaolinite is preferentially formed at low latitudes  
424 under hot and wet tropical climate, allowing strong hydrolysing processes to take effect (Paquet  
425 and Clauer, 1997; Thiry, 2000). By contrast, chlorite and illite are formed in drier climates  
426 where physical erosion prevails (Chamley, 1989). Therefore, many authors classically use  
427 kaolinite as a reliable indicator of humid tropical climatic belt, whereas chlorite and illite are  
428 used as proxies of cold and dry conditions or associated to arid environments. Evolution of clay  
429 mineral assemblages from the Pliensbachian highlight covariation of kaolinite and chlorite  
430 (Figs. 3 and 4), which is a very unusual feature and difficult to reconcile in terms of coeval  
431 formation in a soil governed by a unique climate. Moreover, as discussed by Thiry (2000),  
432 formation of kaolinite-rich laterite and erosion of clay into the oceanic domain require long  
433 durations, estimated to more than 1 Myr. The studied area was located between 30° and 40° of

434 palaeolatitude (Thierry et al., 2000). On the basis of GCM models and lithological and  
435 palaeontological data, the region was subjected to humid to semi-arid climates and  
436 megamonsoonal conditions (Rees et al., 2000; Dera et al., 2009a). Evidence of  
437 contemporaneous kaolinite-rich soils surrounding basins in the north Tethyan basins has not  
438 been demonstrated, and some authors point to the reworking of kaolinite from Palaeozoic rocks  
439 instead as the main source (Hurst, 1985; Jeans, 2006). Kaolinite is, for example, reported in  
440 Devonian and Carboniferous rocks as detrital and/or authigenic mineral derived from closely  
441 emerged landmasses (Crasquin, 1983; Hillier et al., 2006; Shaw, 2006). The palaeogeography  
442 of the north Tethyan domains, as an archipelago with landmasses partly or completely drowned  
443 by an epicontinental sea, has also probably reinforced the high proportions of kaolinite in the  
444 studied basins by comparison with other basins. Periods of strong chemical and physical erosion  
445 by intensive runoff on these continents have therefore likely contributed to increase inherited  
446 kaolinite content of contemporaneous marine sediments, while illite and IS R1 corresponded to  
447 the background of the clay sedimentation. Therefore, we suggest that the K/I ratio is a reliable  
448 proxy to estimate the runoff on continents and inferred humidity/aridity changes that strongly  
449 influenced continental weathering during the Pliensbachian (cf. Ruffell et al., 2002).

### 450 *5.2.3 Illite, chlorite and kaolinite sources*

451 In the Montcornet and Sancerre boreholes, the clay fraction of the Pliensbachian  
452 sediments display unusual features: 1) chlorite is abundant reaching 20% on average, while  
453 Jurassic rocks from European sedimentary basins show generally at most 10% of chlorite in  
454 their clay fraction (e.g. Ruffell et al., 2002; Deconinck et al., 2003; Jeans, 2006; Pellenard and  
455 Deconinck, 2006; Schnyder et al., 2006); 2) parallel changes in illite and chlorite proportions  
456 are not observed, whereas these primary minerals usually show coeval changes in most  
457 sedimentary series, and: 3) instead a co-variation of kaolinite and chlorite is surprisingly  
458 highlighted here, even though these two minerals cannot be formed under the same climatic

459 conditions. This coeval kaolinite- and chlorite-rich composition suggests either two distinct  
460 sources or a common detrital source with both kaolinite and chlorite inherited from ancient  
461 rocks.

462         The similar temporal evolution of kaolinite and chlorite suggest a common source. The  
463 absence of co-variations of chlorite and illite and the co-variation of kaolinite and chlorite could  
464 thus be explained by the reworking of authigenic chlorite and kaolinite formed in pore-spaces  
465 of old sandstones notably described in the boreal domain along the Fennoscandian Shield (Mørk  
466 et al., 2003; Dera et al., 2009a). We suggest therefore that at least a part of kaolinite and chlorite  
467 can be considered as tracers of boreal sources, specifically during periods of high sea-level,  
468 when the London-Brabant Massif was flooded (Fig. 7B). By contrast, during periods of sea-  
469 level fall, kaolinite and chlorite from boreal domains were likely trapped between the  
470 Fennoscandian Shield and the London-Brabant Massif, this latter feeding the northern part of  
471 the Paris Basin with smectite (Montcornet, Fig. 7A). High proportions of kaolinite and chlorite  
472 occur throughout the Pliensbachian succession of the Cleveland Basin (Jeans, 2006), suggesting  
473 continuous input of reworked kaolinite and chlorite from boreal domains, an observation that  
474 strengthens this interpretation (Fig. 7).

475

### 476 5.3. Climatic events and runoff during the Pliensbachian

#### 477 *5.3.1 Relationships between clay mineralogy and C-O-isotopes*

478         In order to test relationships between clay mineral changes and carbon cycle events in  
479 the Tethyan domain, correlations have been attempted between the four studied sections using  
480 ammonite biostratigraphy and carbon-isotope chemostratigraphy and by comparing the K/I  
481 ratio, considered here as a proxy for runoff on land, to significant negative and positive  
482 excursions in carbon isotopes (Fig. 6). Sancerre and Montcornet boreholes cover the top

483 Sinemurian to lowermost Toarcian in the Paris Basin, while Charmouth (Wessex Basin) and  
484 Corbigny (Paris Basin) sections provide additional data for the lower Pliensbachian.

485         During the Late Sinemurian and most of the Early Pliensbachian (up to the Ibex Zone),  
486 a low K/I ratio is observed. At the Sinemurian-Pliensbachian boundary, a weak increase in the  
487 K/I ratio is recorded at Sancerre (Fig. 6). A coeval increase of kaolinite content is consistently  
488 reported in the Lusitanian Basin (Caniço et al., 2015). This trend is not observed at Montcornet  
489 and Charmouth (Fig. 6). It could be explained by either a condensation or hiatus in the  
490 Sinemurian-Pliensbachian boundary interval of the two latter sections. Indeed, the Taylori  
491 Subzone (first Subzone of the Jamesoni Zone) is not recognised at Montcornet and a hiatus is  
492 highlighted at Charmouth (Hesselbo and Jenkyns, 1995). This mineralogical change occurred  
493 within a negative carbon isotope excursion measured on organic matter in the Sancerre borehole  
494 (Peti et al., submitted) and on bulk carbonate at Montcornet (Fig. 6). This negative excursion,  
495 at the so-called Sinemurian-Pliensbachian Boundary Event (SPBE), was identified from  
496 belemnite carbon-isotope data and organic matter/wood  $\delta^{13}\text{C}$  in U.K. basins (Korte and  
497 Hesselbo, 2011; Price et al., 2016; Ruhl et al., 2016) and can also be seen in earlier compilations  
498 (Jenkyns et al., 2002). The same event has been described in the Lusitanian Basin (Duarte et  
499 al., 2014), the Asturian Basin (Armendáriz et al., 2012; Gómez et al., 2016) and the Trento  
500 platform (Franceschi et al., 2014). This negative shift is reported Fig. 8 from the belemnite  
501 carbon isotope compilation of Martinez and Dera (2015).

502         From the top of the Ibex Zone to the lower Davoei Zone, the K/I ratio dramatically  
503 increase in Montcornet and Sancerre (Fig. 6). This major mineralogical change coincides with  
504 a decrease in  $\delta^{13}\text{C}_{\text{org}}$  and  $\delta^{13}\text{C}_{\text{carb}}$  values (Fig. 6) and follows a sea-level lowstand expressed by  
505 the occurrence of smectites on the southern borders of the London-Brabant Massif (LBM). This  
506 major mineralogical change is consistent with the general increase of the kaolinite input into  
507 the NW European domain within the Davoei Zone (Dera et al., 2009a). The K/I increase is also

508 concomitant with more negative  $\delta^{18}\text{O}$  values measured from belemnite rostra, suggesting  
509 warming of sea water (Rosales et al., 2004; Dera et al., 2011a; Armendáriz et al., 2012; Martínez  
510 and Dera, 2015; Gómez et al., 2016, Fig. 8).

511 From the top of the Davoei Zone to the top of the Margaritatus Zone, the K/I ratio  
512 remains high at Sancerre and Montcornet with slight variations following three cycles in both  
513 boreholes (Fig. 6). These clay mineral cycles likely reflect variations of the runoff triggered by  
514 climate in the Milankovitch frequency-band as observed in some Cretaceous sedimentary series  
515 (Moiroud et al., 2012; Ghirardi et al., 2014). These runoff cycles are roughly correlated with  
516 carbon isotope cycles from organic matter in the Sancerre borehole.

517 The upper Pliensbachian (Spinatum Zone) is characterised by a sharp decrease of the  
518 K/I ratio (Fig. 6), also reported by Dera et al. (2009a). This change precedes an increase in  
519  $\delta^{13}\text{C}_{\text{carb}}$  values and is coeval with maximal  $\delta^{18}\text{O}$  values. Similar high  $\delta^{18}\text{O}$  values, recorded in  
520 the Lusitanian Basin (Oliveira et al., 2006; Suan et al., 2010), in Spain (Rosales et al., 2004;  
521 Gómez et al., 2016), in the Cleveland Basin (Korte and Hesselbo, 2011), and in the Western  
522 Carpathians (Arabas and Schlögl, 2015), suggest that low runoff occurred during a cooler  
523 period (Fig. 8).

#### 524 *5.3.2 Periods of enhanced runoff*

525 Using the K/I ratio as a tracer of humid conditions, at least three periods of enhanced  
526 runoff are recognised throughout the Pliensbachian (Fig. 8): 1) the Sinemurian-Pliensbachian  
527 boundary, 2) the Lower/Upper Pliensbachian transition (Davoei/Margaritatus Zones), 3) the  
528 Subnodosus Subzone (Margaritatus Zone, Upper Pliensbachian).

529 A long-lasting  $\delta^{13}\text{C}$  negative excursion of  $\sim 2\text{‰}$ , corresponding to the SPBE,  
530 encompasses the Sinemurian-Pliensbachian boundary. This carbon excursion recorded in  
531 marine carbonates and organic matter has been reported in many localities from the Tethyan



532 domain suggesting a supra-regional or global event (Korte and Hesselbo, 2011). A source of  
533 isotopically light CO<sub>2</sub> injected into the ocean-atmosphere system is required to explain this 2‰  
534 amplitude negative carbon-isotope excursion, likely suggesting either a volcanic contribution,  
535 as proposed by Ruhl et al. (2016), or methane release, as discussed for the CIE (Carbon Isotope  
536 Excursion) of the T-OAE (Hesselbo et al., 2000a; Hesselbo et al., 2007; Hesselbo and  
537 Pieńkowski, 2011; Suan et al., 2011). A hyperthermal event was highlighted during the T-OAE  
538 associated to CO<sub>2</sub> release triggered by Karoo-Ferrar volcanic activity (Pálffy and Smith, 2000;  
539 Jourdan et al., 2008) and/or methane hydrate reservoir disruption (Hesselbo et al., 2000a;  
540 Jenkyns, 2010). The subsequent high *p*CO<sub>2</sub> favoured: 1) concomitant large organic matter  
541 accumulations (black-shale deposits) (e.g. Jenkyns, 1988, 2010; Baudin et al., 1990), and; 2)  
542 runoff and continental weathering increases, expressed by a set of isotope data (Sr, Os, Ca ...)  
543 (Jones and Jenkyns, 2001, Bailey et al., 2003; Cohen et al., 2004; Brazier et al., 2015; Ruhl et  
544 al., 2016) and sedimentological evidence, including change in clay mineral proportions  
545 (Hermoso and Pellenard, 2014). A similar increase in *p*CO<sub>2</sub> is interpreted by the  $\Delta^{13}\text{C}$  ( $\delta^{13}\text{C}_{\text{carb-}}$   
546  $\delta^{13}\text{C}_{\text{org}}$ ) during the SPBE (van de Schootbrugge et al., 2005). Organic matter-rich sediment  
547 including black shale was also deposited in dysoxic to anoxic environments during the latest  
548 Sinemurian (Rosales et al., 2004; Boussaha et al., 2014, Duarte et al., 2014). Besides the  
549 kaolinite enrichment observed in this study, proxies of continental weathering and runoff are  
550 also reported, including, for example, eutrophic conditions linked to higher nutrient input from  
551 continents preventing the widespread occurrence of the *Lithiotis* fauna (Franceschi et al., 2014).  
552 Temperature reconstructions for the SPBE are still debated but no evidence of hyperthermal  
553 temperatures has been described, in contrast to the T-OAE. A warm period is suggested during  
554 the latest Sinemurian based on belemnites from the Asturian Basin (Gómez et al., 2016),  
555 whereas a slight cooling trend is recorded by belemnites and bivalves in the Cleveland Basin  
556 (Korte and Hesselbo, 2011). However, cool bottom water temperatures described in the

557 Cleveland Basin may reflect local deep water conditions and bias temperature reconstruction  
558 of this period (Korte and Hesselbo, 2011). This event was rather interpreted as a greenhouse  
559 interval (Korte and Hesselbo, 2011), consistent with a humid climate and consequently  
560 enhanced runoff on landmasses, as supported by our data.

561 The Lower/Upper Pliensbachian transition (e.g. Davoei Zone-Stokesi Subzone) may  
562 display the most obvious change in runoff conditions during the Pliensbachian with a  
563 generalised and dramatic increase in the kaolinite input to the NW European Basins (Fig. 8).  
564 Interestingly, this period coincides with: 1) a slight change in the  $^{87}\text{Sr}/^{86}\text{Sr}$  curve characterised  
565 by a plateau, suggesting enhanced continental weathering, while a broad decrease of  $^{87}\text{Sr}/^{86}\text{Sr}$   
566 values is recognised throughout the Pliensbachian, hypothetically linked to sea-floor spreading  
567 (Jones et al., 1994; Jones and Jenkyns, 2001); 2) rising sea temperatures inferred from oxygen  
568 isotope belemnite data (Dera et al., 2009b; 2011a; Martinez and Dera, 2015; Gómez et al.,  
569 2016); 3) occasional black shale deposits in the Lusitanian Basin (Silva and Duarte, 2015); 4)  
570 a decrease in ammonite species richness observed in the NW European and Mediterranean  
571 domains likely triggered by warming (Dommergues et al., 2009).

572 These lines of evidence highlight warmer and more humid conditions at the base of the  
573 Davoei Zone. However, this interval is also characterised by a change in the detrital supply  
574 shifting to boreal sources (see section 5.2.3), and a suggested change in water mass circulation  
575 in the NW European domain. In the Davoei Zone, more radiogenic  $\epsilon\text{Nd}$  values recorded in fish  
576 teeth imply an input of water mass affected by young and more radiogenic rocks what may  
577 correspond to sources from the Panthalassa ocean or more likely from low latitudes of the  
578 Tethys Ocean (Dera et al., 2009b; Dera et al., 2015). This apparent paradox between detrital  
579 flux from the Fennoscandian Shield and radiogenic water input originating from the south may  
580 be explained by a rising sea-level favouring connections between sedimentary basins and a new  
581 current pattern (Fig. 7). The subsequent flooding of the London-Brabant Massif would allow

582 kaolinite and chlorite inputs into the Paris and Wessex basins from the north. These  
583 palaeoceanographic and palaeogeographic changes are consistent with mixing of ammonite  
584 faunas from euro-boreal domains recognised during the Davoei Zone (Dera et al., 2011b).  
585 Finally, the Lower/Upper Pliensbachian transition is characterised by two major environmental  
586 disruptions: 1) a warming episode associated to enhanced runoff, and; 2) rising sea-level and  
587 palaeoceanographic changes. These conditions contributed to explain the dramatic clay  
588 mineralogical change observed in NW European domains.

589         In the Margaritatus Zone, the K/I ratio displays weak fluctuations likely triggered by  
590 variability in runoff intensity. The maximum values (i.e. Subnodosus Subzone), coincide with  
591 the occurrence of organic matter-rich sediment and black shale deposits reported as the Late  
592 Pliensbachian Organic-Rich Preservation Interval (OMPI; Rosales et al., 2004; Silva et al.,  
593 2011; Armendáriz et al., 2012; Caruthers et al., 2014; Silva and Duarte, 2015; Neumeister et  
594 al., 2015). Maximal runoff within Subnodosus Subzone likely triggered an increase in organic  
595 productivity through the associated increase in the nutrient supply.

596

### 597 *5.3.3 Glacio-eustasy during the Pliensbachian?*

598         The possibility for glacio-eustasy should be addressed because smectite events in the  
599 north Paris Basin clearly indicate emersion of the London Brabant Massif (LBM) during sea-  
600 level fall, while the absence of this mineral highlights its flooding during sea-level rise (Figs. 7  
601 and 8). Such a great palaeogeographic change likely resulted from the flatter and lower  
602 topography of the LBM compared to other Palaeozoic massifs because of its external position  
603 in the Variscan belt. Alternating emersion and flooding of the LBM with relatively short  
604 durations (i.e. <1 Myr) and high amplitude variations of the sea-level, at least several tens of  
605 metres, appears more consistent with a glacio-eustasy model than a regional uplift. Indeed

606 regional uplift associated to the Cimmerian tectonic phases, described for the LBM, seems have  
607 been progressive with moderate speed of unroofing throughout the Jurassic from apatite fission  
608 track data (Vercoutere and Van den Haute 1993). The smectite events are recorded within the  
609 Ibex and Spinatum Zones, which correspond to cooler intervals as expressed in oxygen isotope  
610 data (Fig. 8 and Suppl. Mat. Fig. 1). This is particularly clear in the Spinatum Zone, for which  
611 time ice-cap growth has been previously suggested from sedimentological and isotope data  
612 (Price, 1999; Suan et al., 2011, Dera et al., 2011a, 2011b; Korte and Hesselbo, 2011; Arabas  
613 and Schlögl, 2015). This inference is also consistent with a relatively low  $p\text{CO}_2$  (around 900  
614 ppm) inferred from stomatal density from Araucariaceae leaves (Steinthorsdottir and Vajda,  
615 2015). In this cooler interval, the K/I ratio dramatically drops suggesting reduced humidity,  
616 slowing down of the water cycle, and subsequent low runoff (Fig. 8). Combined with the sea-  
617 level fall inferred from the uppermost part of the Pliensbachian, this situation strengthens a  
618 glacio-eustatic control. A similar situation occurred from the probably topmost part of the  
619 Jamesoni Zone to the lower part of the Ibex Zone, where low K/I ratio and smectite occurrence  
620 are observed together with higher values in bulk  $\delta^{18}\text{O}_{\text{carb}}$  within a regressive trend and a low  
621 stand of sea-level (Fig. 8 and Suppl. Mat. Fig. 2). These coincident changes are consistent with  
622 the formation of ice at high latitudes during the interval that spans the Ibex Zone, although  
623 belemnite oxygen isotope data are more scattered and do not show any clear cooling (Fig. 8).  
624 Changes in the SMOW values during the cold snap and the subsequent deglaciation, providing  
625 fresh water input and salinity variations, could also be responsible for the scattered oxygen  
626 isotope data. Recent isotope data from belemnite in the Dorset (U.K.) are consistent with a  
627 relatively cold Jamesoni-Ibex interval compared to the warmer Davoei interval (Price et al.,  
628 2016). The following warming and deglaciation, combined with the sea level rise, likely  
629 enhanced runoff and reworking of ancient kaolinite and chlorite-bearing sediments and rocks  
630 from the Fennoscandian Shield. This climatic story is consistent with oxygen isotope data

631 published in the Basque-Cantabrian and Asturian Basins (Rosales et al., 2006; Armendáriz et  
632 al., 2012).

633

## 634 6. Conclusions

635 Clay mineral and paired carbon-oxygen-isotope data have been generated from two boreholes  
636 in the Paris Basin representing an interval from the latest Sinemurian to the earliest Toarcian,  
637 with additional mineralogical data from sections in the Paris and Wessex Basins. The diversity  
638 of clay assemblages results from the complex interplay of several detrital sources, sea-level  
639 change, seaway circulation, and runoff. The following features interpreted in terms of climate  
640 and palaeoceanographic changes are highlighted:

641 - Increasing kaolinite/illite ratio characterises periods of enhanced runoff, during: i) the  
642 Sinemurian-Pliensbachian transition associated with a negative carbon excursion (SPBE); ii)  
643 the Early/Late Pliensbachian transition (Davoei Zone-Stokesi Subzone), and; iii) the  
644 Subnodosus Subzone (Late Pliensbachian, Margaritatus Zone). These periods correspond to  
645 warm and humid climates, favouring continental weathering and subsequent black shale  
646 deposition. By contrast, cooler periods with low runoff occurred during the Jamesoni - Ibex and  
647 the Spinatum Zones.

648 - The influence of climate forcing on clay assemblages is magnified by palaeoceanographic  
649 changes and sea level fluctuations. A dramatic change in clay mineralogy is notably observed  
650 for the Davoei Zone, characterised by kaolinite and chlorite-rich sedimentation. We suggest  
651 that these minerals were likely reworked from ancient sediments and rocks from the  
652 Fennoscandian Shield during a high sea-level period favouring connections between  
653 sedimentary basins and flooding of the London Brabant Massif. By contrast, this massif was  
654 emerged during periods of sea-level lowstand and acted as a topographic barrier.

655 - Two smectite-rich intervals occurred in the north Paris Basin during the two periods of sea-  
656 level lowstand (Ibex and Spinatum Zones). These features are limited to the margin of the  
657 London Brabant Massif, which was a unique source of smectite. These smectite events highlight  
658 recurrent emersions of the London Brabant Massif, characterised by a flat and low topography,  
659 and suggest significant amplitude of the sea-level fluctuation that could be explained by  
660 glacioeustasy. Smectite-rich intervals in the north Paris Basin may thus emphasize continental  
661 ice growth at times during the Early Jurassic (i.e. late Jamesoni/early Ibex Zones and Spinatum  
662 Zone).

663

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670

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1038 Figure captions:

1039 Figure 1. Position of studied boreholes and outcrops on the Early Jurassic palaeomap (modified  
1040 from Blakey's palaeomap: <https://deeptimemaps.com/> and Thierry et al., 2000). Abbreviations:  
1041 SM=Scottish Massif; IrM=Irish Massif; CM= Cornubian Massif; WH=Welsh High; LBM=  
1042 London-Brabant Massif; RM=Rhenish Massif; BM=Bohemian Massif; MC=Massif Central;  
1043 AM=Armorican Massif; IbM=Iberian Massif.

1044 Figure 2. Detailed lithology, sampling and ammonite biostratigraphy of the Montcornet  
1045 borehole.

1046 Figure 3. Biostratigraphy, lithology, sequence stratigraphy, clay mineralogy, K/I  
1047 (kaolinite/illite) ratio and associated SiZer map,  $\delta^{13}\text{C}$  from organic matter, in the Sancerre  
1048 borehole (Southern Paris Basin). Smoothed curves with their 95% confidence interval have  
1049 been calculated using Kernel regressions for K/I ratio and  $\delta^{13}\text{C}_{\text{org}}$ . Ammonite zone and subzone  
1050 abbreviations: Gib.=Gibbosus; Raricos.=Raricostatum; Sub.=Subnodosus;  
1051 Tenuicos.=Tenuicostatum. Grey cases correspond to uncertainties in the biostratigraphic  
1052 framework.

1053 Figure 4. Biostratigraphy, lithology, sequence stratigraphy, clay mineralogy, K/I  
1054 (kaolinite/illite) ratio and associated SiZer map,  $\delta^{13}\text{C}$  from bulk carbonate in the Montcornet  
1055 borehole (Northern Paris Basin). Smoothed curves with their 95% confidence interval have  
1056 been calculated using Kernel regressions for K/I ratio and  $\delta^{13}\text{C}_{\text{carb}}$ . Ammonite zone and subzone  
1057 abbreviations: C.=Capricornus; Gib.=Gibbosus; J.=Jamesoni; Mac.=Macdonelli;  
1058 M.=Maculatum; P.=Polymorphus; Rar.=Raricostatum; Sub.=Subnodosus. Grey cases  
1059 correspond to uncertainties in the biostratigraphic framework. The legend is the same as fig. 3.

1060 Figure 5. Scanning Electron Microscope from samples of Montcornet. A-B) views of a  
1061 limestone bed sample (772.75 m), displaying calcite mixed with detrital clays; C) zoom of the



1062 same sample (772.75 m) showing typical authigenic vermicular kaolinite formed inside  
1063 porosity; D) view of clayey sample (812.9 m) showing common detrital clay fabric with some  
1064 framboidal pyrites.

1065 Figure 6. Correlation between Sancerre and Montcornet boreholes (Paris Basin) and two  
1066 additional outcrops from Charmouth (Wessex Basin) and Corbigny (Paris Basin). Red intervals  
1067 indicate an increase in the K/I ratio while the blue intervals indicate a decrease in the K/I ratio.  
1068 Yellow intervals correspond to the main  $\delta^{13}\text{C}$  negative excursions identified in the two  
1069 boreholes (SPBE: Sinemurian Pliensbachian Boundary Event; T-OAE: Toarcian Oceanic  
1070 Anoxic Event). Ammonite zone and subzone abbreviations: B.=Brevispina; C.= Capricornus;  
1071 F.=Figulinum; Gib.=Gibbosus; Ib.=Ibex; J.=Jamesoni; L.=Luridum; Mac.=Macdonelli;  
1072 M.=Masseanum; P.=Polymorphus; M.=Maculatum; Rar./Raricos=Raricostatum; Sub.=  
1073 Subnodosus; T.=Taylori; V.=Valdani. Grey cases correspond to uncertainties in the  
1074 biostratigraphic framework.

1075 Figure 7. Palaeomaps and schematic cross-sections for clay minerals sourcing in: A) sea-level  
1076 lowstand configuration; B) sea-level highstand configuration. Abbreviations of Massifs and  
1077 discussed sections: AM=Armorican Massif; BM=Bohemian Massif; Co=Corbigny;  
1078 CB=Cleveland Basin; Ch=Charmouth; CM=Cornubian Massif; LBM=London-Brabant  
1079 Massif; IbM=Iberian Massif; IrM=Irish Massif; Mo=Montcornet; MC=Massif Central;  
1080 PB=Paris Basin; Sa=Sancerre; RM=Rhenish Massif; SM=Scottish Massif; WH=Welsh High.  
1081 Clay mineral abbreviations: C=chlorite; I=illite; K=kaolinite; S=smectite. Palaeocurrents are  
1082 based on Bjerrum et al., 2001 and Dera et al., 2015.

1083 Figure 8. Summary of clay mineral data, geochemical data, and organic matter accumulations  
1084 from the end Sinemurian to the early Toarcian, reported against numerical ages, biostratigraphic  
1085 framework based on ammonites and sequence stratigraphy of the Tethyan domain (Jacquin et

1086 al., 1998; Gradstein et al., 2012): Smectite events (this study);  $\delta^{18}\text{O}$  from belemnites (dataset  
1087 from Martinez and Dera, 2015) with red intervals corresponding to warm periods and blue  
1088 intervals to cold periods; runoff based on the K/I ratio (this study), with position of small K/I  
1089 increase (1, 2, 3) and major K/I increase (A) and decrease (B);  $\delta^{13}\text{C}$  from organic matter  
1090 (Hesselbo et al., 2000, 2007; van de Schootbrugge et al., 2005; Suan et al., 2010; Korte and  
1091 Hesselbo, 2011),  $\delta^{13}\text{C}$  from bulk carbonates (Oliveira et al., 2006; Silva et al., 2011; Duarte et  
1092 al., 2014; this study) and  $\delta^{13}\text{C}$  from belemnite data (Martinez and Dera, 2015), with yellow  
1093 intervals corresponding to main negative carbon excursions; black shale occurrence (black  
1094 point), the point size indicates their record extent (Rosales et al., 2004; Oliveira et al., 2006;  
1095 Silva et al., 2011; Armendáriz et al., 2012; Boussaha et al., 2014; Duarte et al., 2014);  $^{87}\text{Sr}/^{86}\text{Sr}$   
1096 curve (Jones et al., 1994) and  $\epsilon\text{Nd}$  curve (Dera et al., 2009b).

1097

1098 Supplementary material:

1099 Table 1: Sampled and identified ammonites in the Montcornet borehole

1100 Table 2: Clay mineral proportion in the Montcornet borehole. (IS R0: R0 type illite smectite  
1101 mixed-layer; C: chlorite; IS R1: R1 type illite smectite mixed-layer; I: illite; K: kaolinite; K/I:  
1102 kaolinite/illite ratio.

1103 Table 3: Clay mineral proportion in the Montcornet borehole. (IS R0: R0 type illite smectite  
1104 mixed-layer; C: chlorite; IS R1: R1 type illite smectite mixed-layer; I: illite; K: kaolinite; K/I:  
1105 kaolinite/illite ratio.

1106 Table 4:  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  from bulk carbonate values (expressed in ‰) in the Montcornet  
1107 borehole.

1108 Fig. 1: Biochronology from North-Western Europe and distribution of ammonites fauna in the  
1109 Montcornet borehole.

1110

1111 Fig. 2:  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  values from bulk carbonate in the Montcornet borehole (Northern Paris  
1112 Basin).

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1114

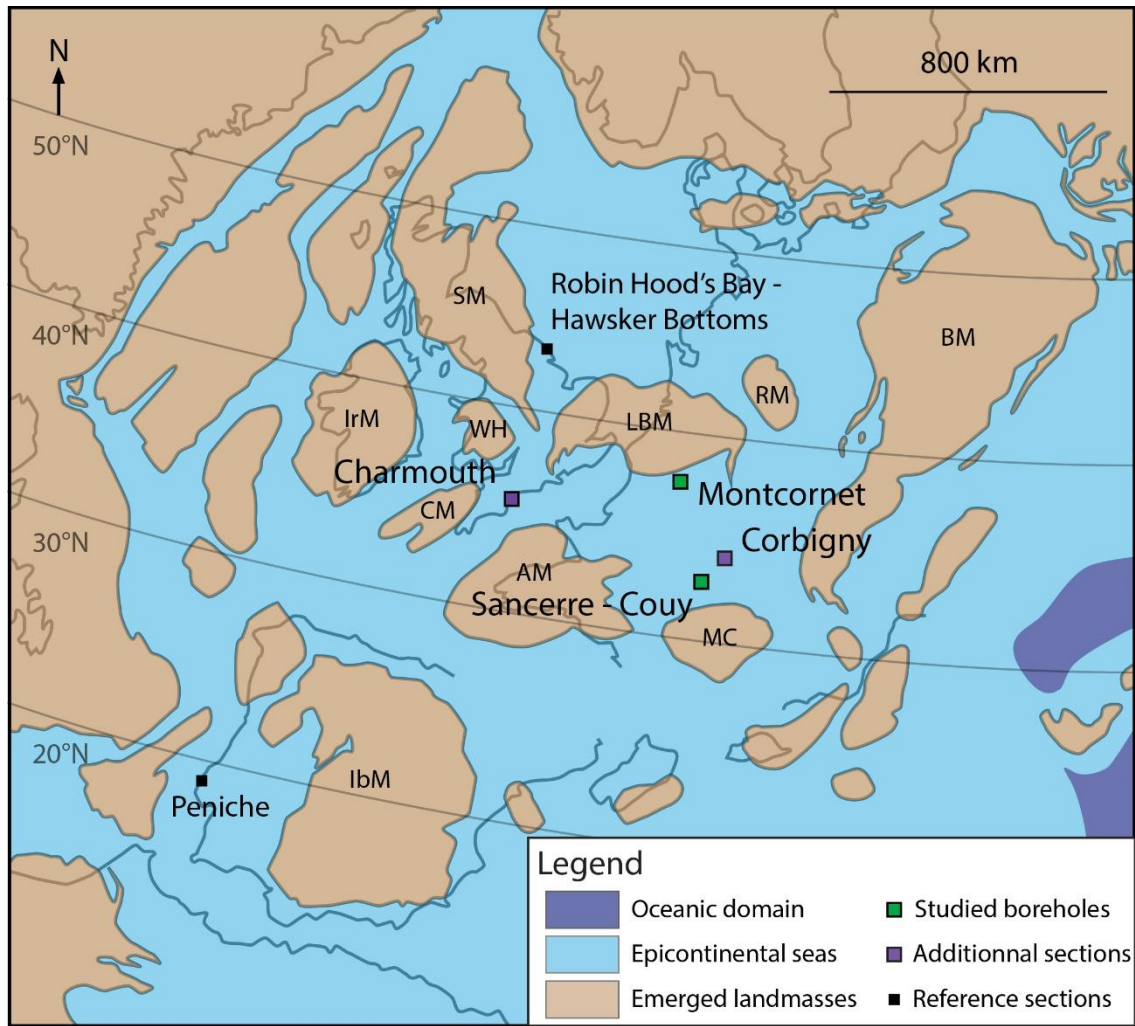


Fig 1

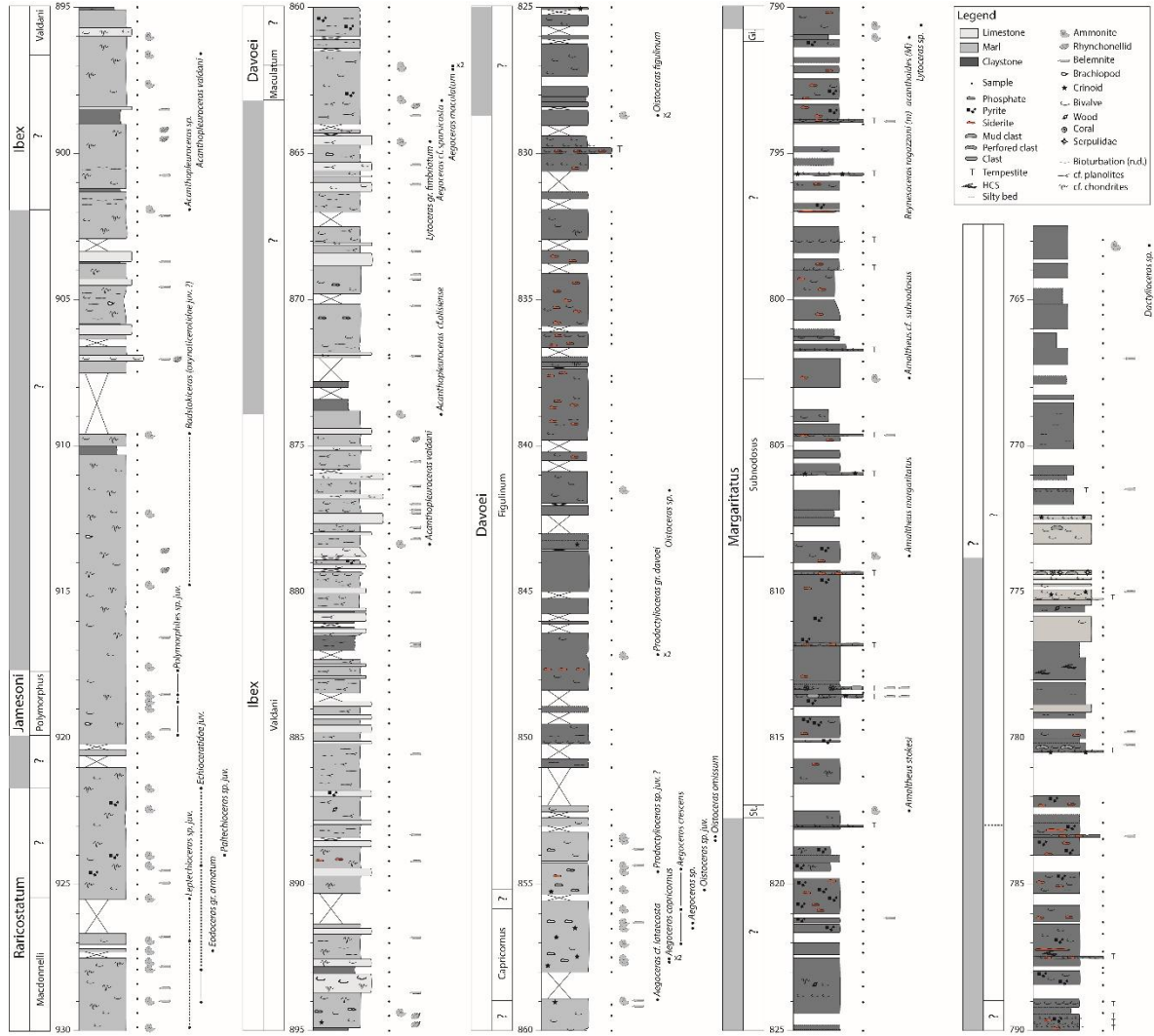


Fig 2

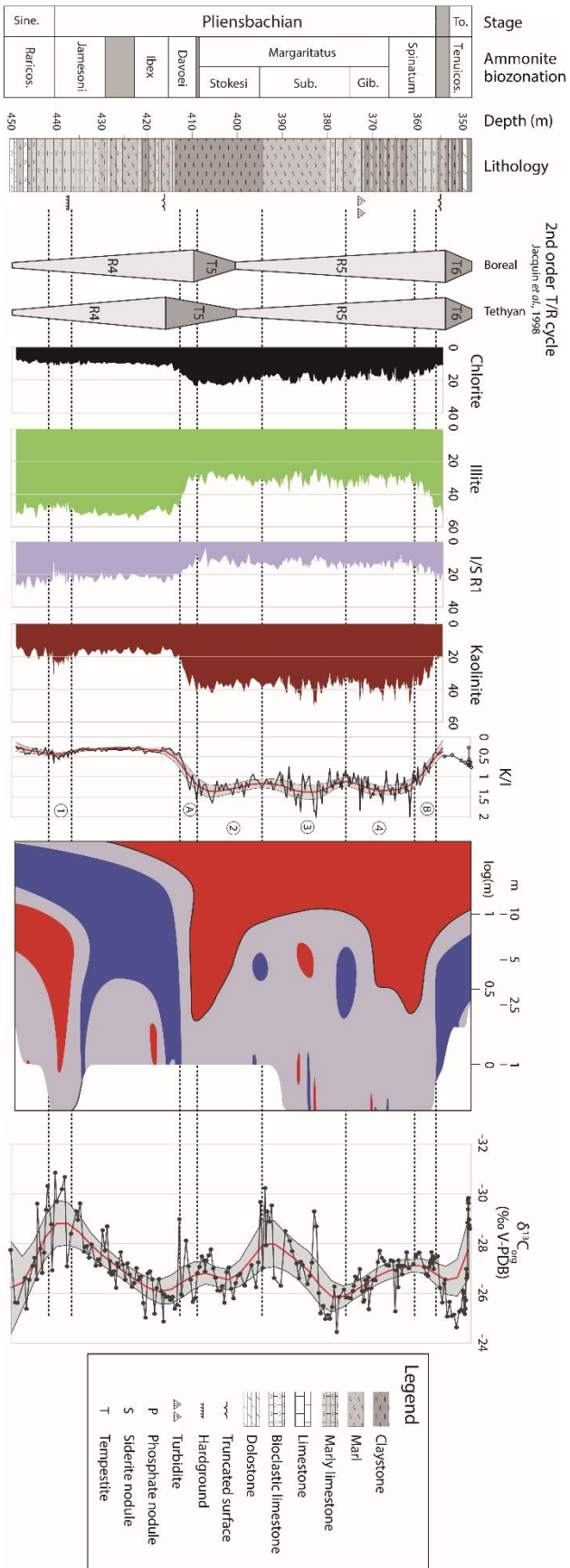


Fig 3

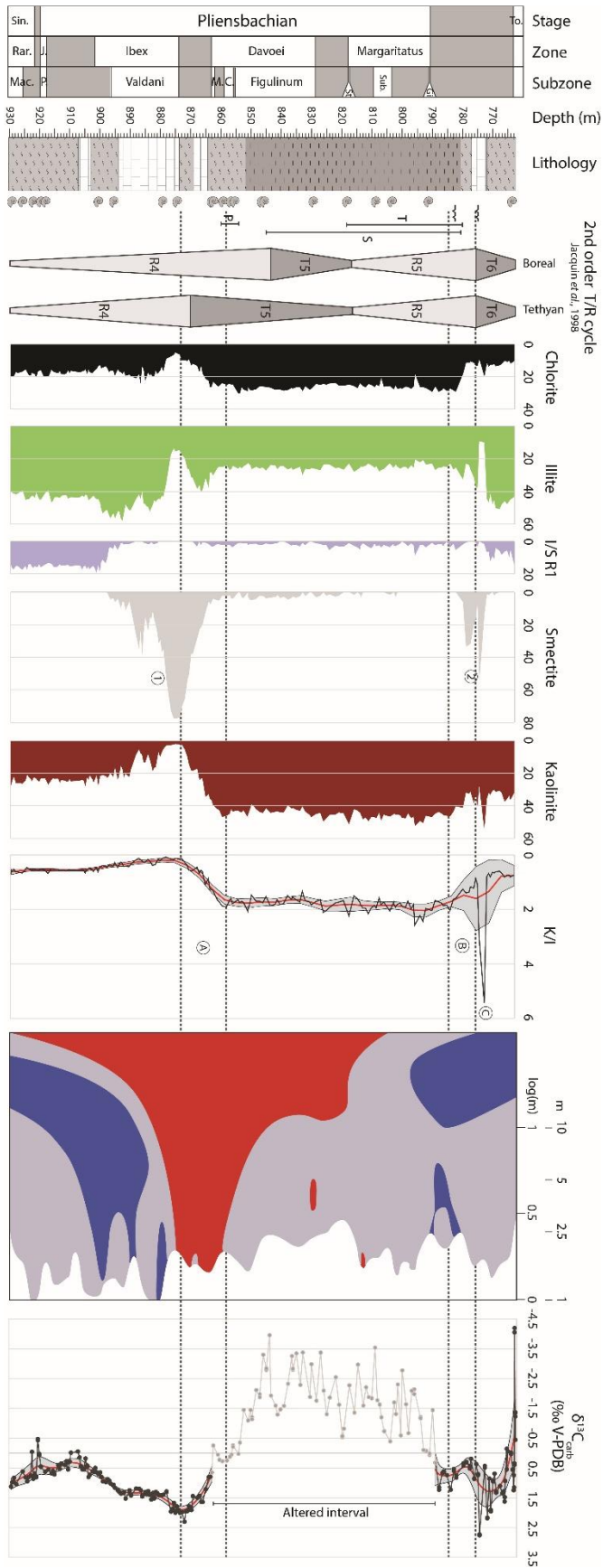


Fig 4



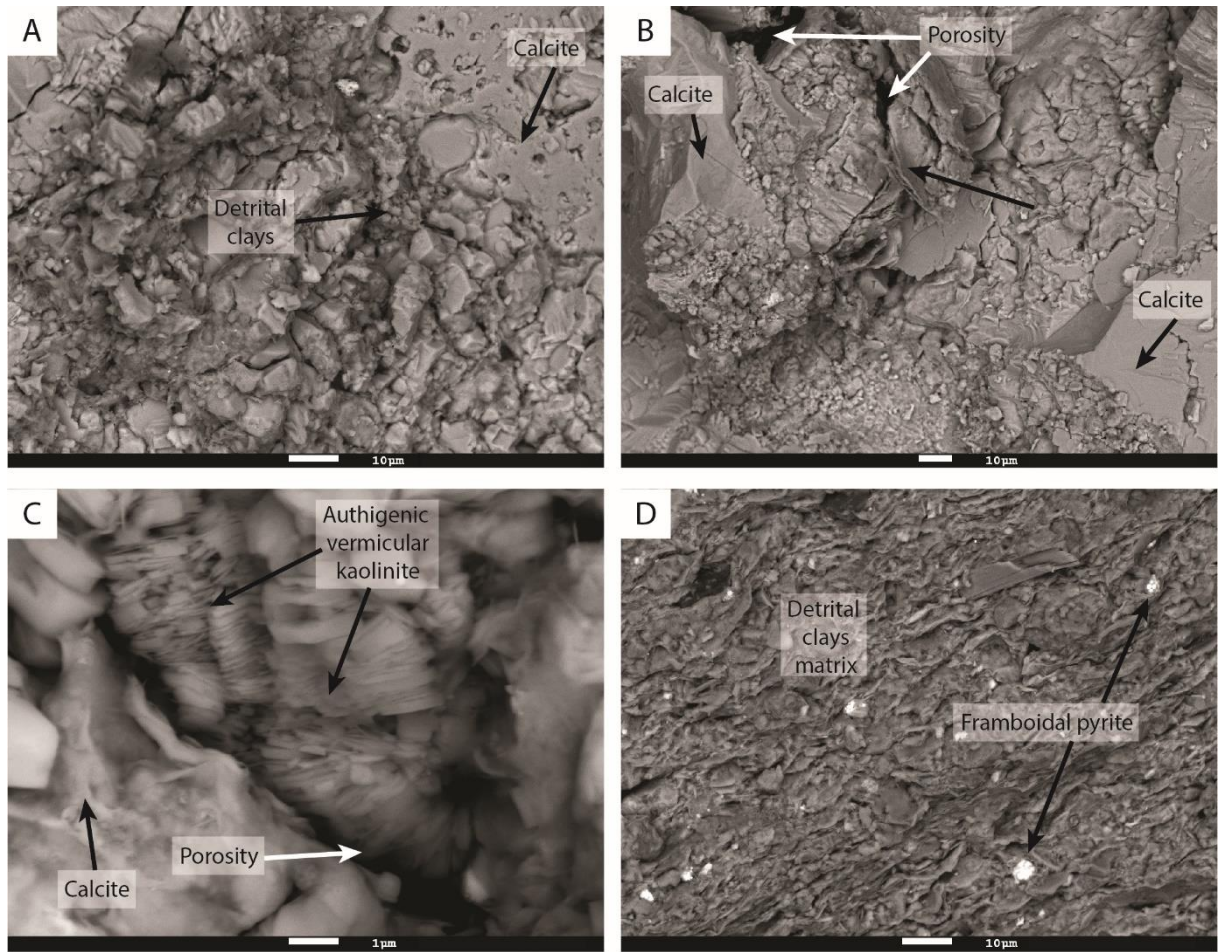


Fig 5



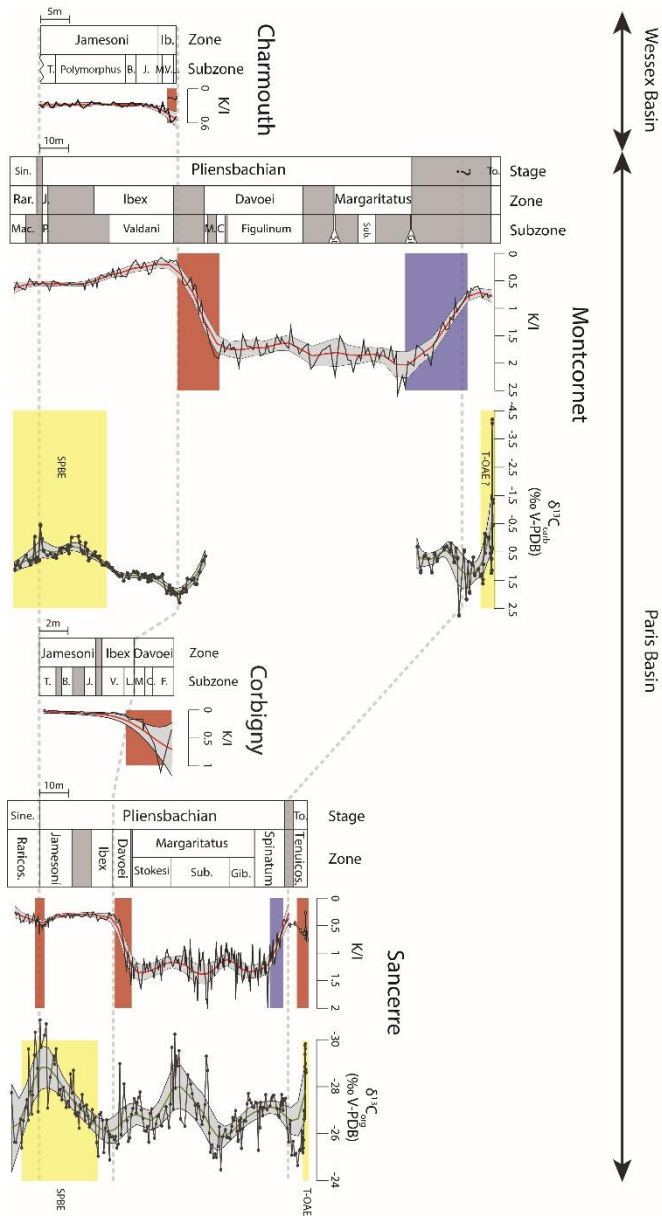


Fig 6

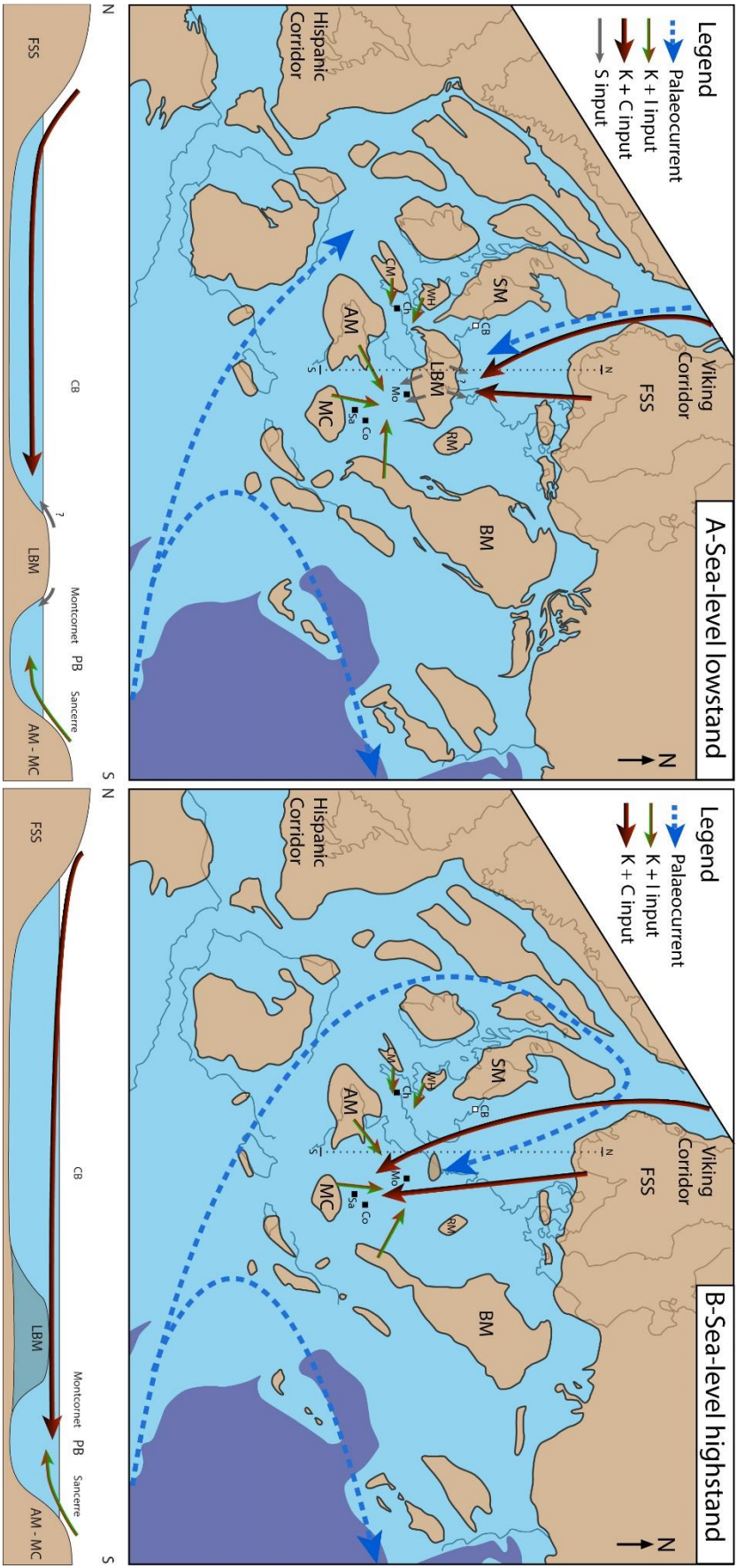


Fig 7

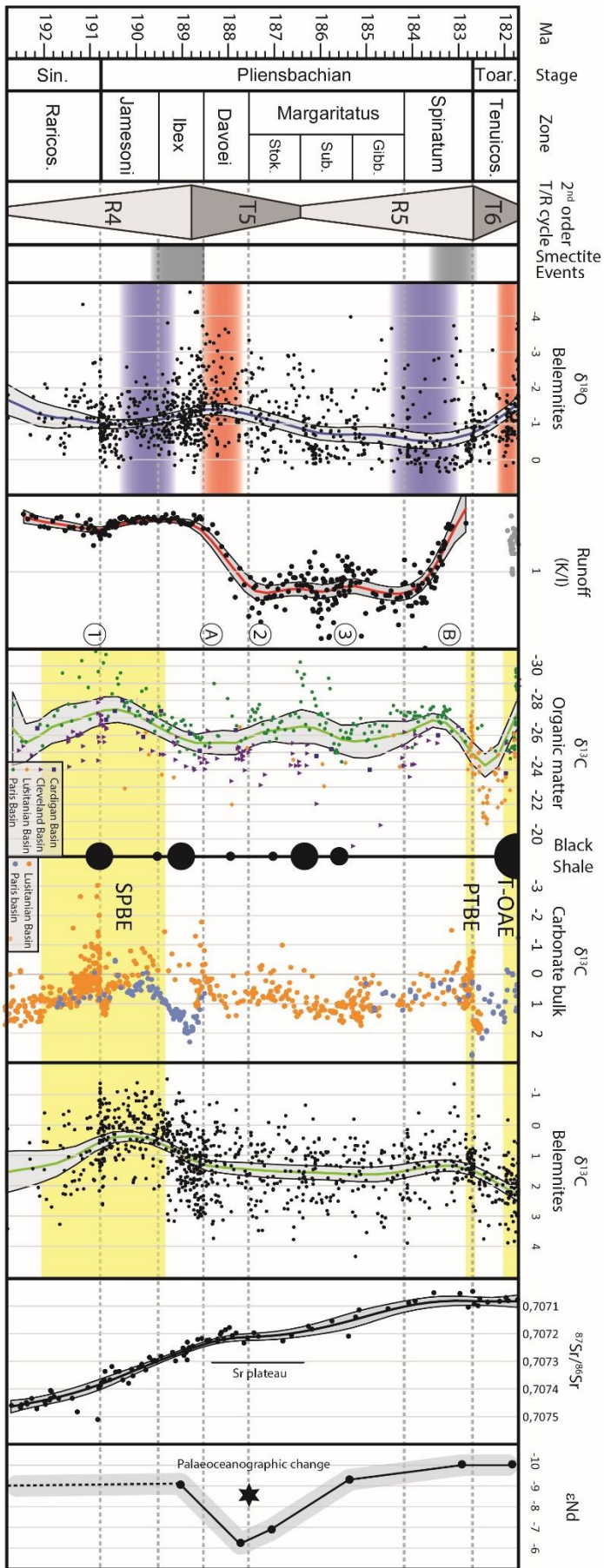


Fig 8