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A global palaeoclimatic reconstruction for the Valanginian based on clay mineralogical and geochemical data

Guillaume Charbonnier, Stéphanie Duchamp-Alphonse, Jean-François Deconinck, Thierry Adatte, Jorge Spangenberg, Christophe Colin, Karl Föllmi

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27 **Abstract**

28

29 High-resolution clay mineral and $\delta^{18}\text{O}_{\text{carb}}$ analyses have been performed on three sections of
30 Valanginian age (Early Cretaceous), from northwestern ($\sim 20\text{--}30^\circ\text{N}$) and southern ($\sim 53^\circ\text{S}$) Tethyan
31 realms. The data have been integrated in a large set of published mineralogical (clays), and
32 geochemical ($\delta^{18}\text{O}_{\text{carb}}$, $\delta^{18}\text{O}_{\text{bivalve}}$, $\delta^{18}\text{O}_{\text{lenticulina}}$, $\delta^{18}\text{O}_{\text{belemnite}}$, and TEX_{86}) data from 17 sections,
33 situated between $\sim 45^\circ\text{N}$ and $\sim 53^\circ\text{S}$. Based on this data set, we provide new insights into
34 Valanginian climate conditions from a wide range of palaeolatitudes. We highlight climate trends
35 within specific areas and identify main climate belts. In the Proto-North Atlantic realm ($\sim 15\text{--}17^\circ\text{N}$)
36 large amounts of smectite together with the occurrence of palygorskite testify for a semi-arid
37 climate belt throughout the Valanginian. Significant variations in kaolinite content in the
38 northwestern Tethyan realm (~ 20 to 30°N) suggest that this realm has been more sensitive to
39 changes in hydrological cycling and subsequent erosion and runoff processes during the early to
40 early-late Valanginian transition interval. This time interval, that is coincident with the pronounced
41 positive and globally recognized carbon isotope excursion (CIE; the Weissert episode), documents
42 the wet conditions of a humid subtropical climate belt. Temperature ranges derived from $\delta^{18}\text{O}$
43 records suggest climate warming by about $3\text{--}5\pm 0.9^\circ\text{C}$ in the northern hemisphere, during the
44 Weissert episode. This is followed by a global cooling of about ca. $1\text{--}5\pm 0.9^\circ\text{C}$ in both hemispheres,
45 during the late Valanginian.

46 We postulate that these patterns were due to interplays between tectonic and orbital factors,
47 which have controlled the distribution of regional palaeoclimate belts during the Valanginian. The
48 semi-arid belt expressed in the Proto North Atlantic realm has probably been induced by its specific
49 palaeogeographic configuration and the existence of isolated basins, which were each characterized
50 by strong and continuous thermohaline circulation. An eccentricity paced monsoon-like system
51 might have played a significant role in maintaining the subtropical belt around the northwestern
52 Tethyan realm, up to the mid latitudes ($\sim 35\text{--}45^\circ\text{N}$). Superimposed on these regional processes, a

53 widespread magmatic pulse in Paraná-Etendeka volcanic activity might have stimulated greenhouse
54 conditions, impacting marine biota and favouring the development of a dense vegetation cover on
55 the continent. An increase in sedimentary burial of ^{13}C depleted organic carbon on the continents
56 may have contributed to the progressive global positive carbon isotope excursion. In the medium
57 term, it has also probably led to a significant decrease in atmospheric CO_2 , as testified by the global
58 cooling recorded during the late Valanginian in both hemispheres and the probable concomitant
59 extension of polar ice caps.

60

61 *Keywords:* hydrolysing conditions; temperature range; palaeoclimate belt; volcanic activity; global
62 reconstruction; Valanginian.

63

64 **1. Introduction**

65

66 Historically, the Cretaceous (145 ± 0.8 – 66 ± 0.05 Ma; Ogg et al., 2016) has been referred to a
67 period of greenhouse Earth, characterized by warm and equal conditions (e.g., Hallam, 1985; Frakes
68 et al., 1992). However, in the last decades, numerous studies have documented evidences for the
69 existence of shorter episodes of considerable palaeoclimatic and palaeoenvironmental changes (e.g.,
70 Jenkyns, 2010; Föllmi, 2012). The Valanginian time interval represents such a dynamic phase. In
71 sedimentary archives, important changes in the global carbon cycle are expressed by a significant
72 positive carbon isotope excursion (CIE) at the early–late Valanginian transition, known as the
73 “Weissert episode” (e.g., Cotillon and Rio, 1984; Weissert, 1989; Erba et al., 2004; Bornemann and
74 Mutterlose, 2008; Charbonnier et al., 2013). The CIE is preceded by and associated with several
75 phases of carbonate platform drowning (Schlager, 1989; Föllmi et al., 1994, 2006; Graziano, 1999),
76 as well as with widespread turnovers (from oligotrophic to mesotrophic assemblages) and crises in
77 pelagic (planktonic and benthic organisms) and neritic (bioconstructions) carbonate biota (Channell
78 et al., 1993; Erba and Tremolada, 2004; Duchamp-Alphonse et al., 2007, 2014; Barbarin et al.,

79 2012). Numerous indicators of cooler conditions such as glendonites, dropstones, tillite deposits and
80 ice rafted debris, have been recognized in Valanginian sediments in Australian, Canadian,
81 Spitsbergen, and Siberian basins (e.g. Kemper and Schmitz, 1975; Price, 1999; Alley and Frakes,
82 2003; Price and Nunn, 2010; Alley et al., 2019; Vickers et al., 2019). Furthermore, changes in
83 calcareous nannofossil assemblages (Melinte and Mutterlose, 2001), oxygen isotope data (Van de
84 Schootbrugge et al., 2000; Pucéat et al., 2005; McArthur et al., 2007; Barbarin et al., 2012;
85 Meissner et al., 2015) and more recently, Mg/Ca measurements (Price et al., 2018), have evidenced
86 an overall 2 to 4°C cooling trend within the northern hemisphere during the late Valanginian, which
87 may suggest the presence of a polar ice cap (McArthur et al., 2007; Price and Passey, 2013). In
88 parallel, clay mineral records from the northwestern Tethys have highlighted short-term
89 Valanginian palaeoclimate fluctuations in low to mid latitudes, with warm and humid conditions
90 during the Weissert episode, and drier conditions during the cooler phase in the late Valanginian
91 (Duchamp-Alphonse et al., 2011; Westermann et al., 2013; Charbonnier et al., 2016). An important
92 magmatic pulse related to the volcanic activity of the Parana-Etendeka traps has been postulated to
93 have triggered the Valanginian environmental perturbations (Föllmi et al., 1994; Erba et al., 2004;
94 Martinez et al., 2015; Charbonnier et al., 2017). Massive volcanism and the associated released of
95 CO₂ in the ocean-atmosphere system may have accelerated the hydrological cycle, intensified
96 silicate weathering, and fertilized the coastal environments; thereby affecting both marine and
97 terrestrial ecosystems (Föllmi et al., 1994; Erba et al., 2004; Charbonnier et al., 2017).

98 While such results may provide a rather clear image of the evolution of climate during the
99 Valanginian, new and contrasting findings have challenged this particular view, rendering the
100 reconstructions of palaeoclimate conditions that prevailed during the Valanginian more elusive. The
101 most striking controversial examples are the TEX₈₆ records from the Proto North Atlantic (~15–
102 17°N) and the southern Tethyan (~53°S) realms, which document high and rather stable sea-surface
103 temperatures (SST of ~33 to 34°C and ~26°C, respectively) through the entire Valanginian (Littler
104 et al., 2011). As a consequence, these authors have re-evaluated the earliest Cretaceous as a warm

105 and stable period. In addition, recent petrological and geochemical analyses on the complex
106 sequence of ikaite-calcite precipitation leading to glendonite formation suggest that glendonites
107 document past events of methane release and oxidation in polar settings rather than cold-water
108 conditions (e.g., Morales et al., 2017). Similarly, the origin of dropstones in the geological record
109 can be related to non-glacial mechanisms such as gravity processes or wood rafting, implying that
110 these deposits cannot consistently be interpreted as the result of ice rafting and therefore cool
111 climate (Bennett et al., 1994, 1996; Doublet and Garcia, 2004).

112 This controversy in the interpretation of climate evolution during the Valanginian is
113 promoted by the lack of high-resolution climate reconstructions in the southern hemisphere based
114 on geochemistry and clay mineralogy (Ditchfield, 1997; Price and Nunn, 2010; Littler et al., 2011;
115 Price and Passey, 2013). Here, we provide new insights into Valanginian climate conditions from a
116 wide range of palaeolatitudes, in order to highlight main trends within specific realms, and
117 emphasize the presence and localization of main climate belts. This study is based upon new clay
118 mineral and $\delta^{18}\text{O}_{\text{carb}}$ data from the northwestern ($\sim 20\text{--}30^\circ\text{N}$) and southern ($\sim 53^\circ\text{S}$) Tethys realms.
119 These data have been integrated into a set of published clay, $\delta^{13}\text{C}$, $\delta^{18}\text{O}$, and TEX_{86} data from sites
120 situated between $\sim 45^\circ\text{N}$ and $\sim 53^\circ\text{S}$ (Table 1, Fig. 1). Our multiproxy approach helps specifying
121 weathering intensity on continents and documenting humid and arid climate belts. It is also useful to
122 re-evaluate previous geochemical results and propose coherent palaeotemperature patterns on a
123 global scale.

124

125 **2. Data compilation and method**

126

127 Published and unpublished geochemical and mineralogical data from the Valanginian have
128 been compiled within a sizeable database presented in Tables 1A and B. In detail, 1924 oxygen
129 isotope measurements represented by $\delta^{18}\text{O}$ values of whole-rock carbonate ($\delta^{18}\text{O}_{\text{carb}}$), hand-picked
130 benthic foraminifera ($\delta^{18}\text{O}_{\text{lent}}$: *Lenticulina* spp.), bivalves ($\delta^{18}\text{O}_{\text{biv}}$), and belemnite samples ($\delta^{18}\text{O}_{\text{bel}}$),

131 37 TEX₈₆ analyses, as well as 257 clay mineral data, have been gathered from 17 reference sections.
132 These published data have been complemented by 69 new geochemical and 347 new mineralogical
133 analyses on three reference sections, situated in the northwestern (Barranco del Garranchal and
134 Molinas de las Oicas sections, Betic cordillera, south-east Spain) and the southern (ODP Site 765C)
135 Tethyan realms (Fig. 1). Lithological, palaeogeographical and depositional settings of the selected
136 sections are given in Table 1. These sections are located in the southern part of the Boreal (~35–
137 45°N), the northwestern (~20–30°N) and southern (~53°S) Tethyan, and the Proto North Atlantic
138 (~15–17°N) realms (Table 1 and Fig. 1). They all benefit from robust biostratigraphic and/or
139 magnetostratigraphic constraints as well as detailed carbon-isotope stratigraphies, which provide a
140 most useful tool for regional to global correlations given their characteristic evolution (Wendler,
141 2013).

142 The composition of clay mineral assemblages selected in the database has been analyzed by
143 XRD method on the <2 µm clay fraction of sediments using the classical analytic procedure
144 described by Moore and Reynolds (1997). Kaolinite, illite, chlorite, palygorskite, smectite, and
145 illite/smectite irregular mixed-layers (R0) were identified in the sections. The R0 I-S minerals,
146 which contain more than 60 % of smectite layers, have been included in the smectite group and will
147 hereafter be called “smectite”. Oxygen and carbon stable isotope data selected here have been
148 obtained using isotope ratio mass spectrometry and reported in the delta notation (δ) as the per mil
149 (‰) deviation relative to the Vienna Pee Dee Belemnite standard (VPDB). Detailed methodologies
150 have been described in the Supplementary Online Materials (SOM). These data have been obtained
151 from various laboratories using different instruments and standards, which may introduce possible
152 bias for a global compilation (see Table S1 in Supplementary Online Materials). Therefore, in order
153 to maximize the applicability of the data, LOESS smoothing curves (i.e. locally weighted scatterplot
154 smoothing, Cleveland and Devlin, 1988), have been used for the mineralogical and geochemical
155 datasets, for each highlighted realm (Table 1 and Figs. 2–5). Within their 95 % confidence levels
156 (statistical precision of 5 %, 2σ ; Figs. 2-5), they have the potential to highlight main regional

157 palaeoenvironmental trends.

158

159 **3. Results**

160

161 3.1. Northern hemisphere data

162

163 3.1.1. Northwestern Tethyan realm (~20°–30°N)

164 Kaolinite, smectite, $\delta^{18}\text{O}_{\text{carb}}$ and $\delta^{18}\text{O}_{\text{bel}}$ LOESS smoothing curves are plotted with the
165 carbon isotope stratigraphy and the biostratigraphic scheme in Fig. 2.

166 The smoothed records of kaolinite and smectite range from 16 to 39 % and from 21 to 43 %,
167 respectively. The early Valanginian (*Tirnovella pertransiens*–*Busnardoites campylotoxus* ammonite
168 Zones) is characterized by relatively stable values with averages of 27 and 36 %, despite a small
169 decreasing/increasing trend in the kaolinite/smectite percentages at the base of the section. Near the
170 early–late Valanginian transition, i.e. where the Weissert event starts (*B. campylotoxus*–*Saynoceras*
171 *verrucosum* transition Zones), the kaolinite record shows an increasing trend, reaching a maximum
172 of 39% at the $\delta^{13}\text{C}$ climax, whereas the smectite curve displays a drop of 13 % (from 34 to 21 %). A
173 significant decrease in kaolinite abundances to 16 % is observed throughout the early late
174 Valanginian (*S. verrucosum*–*Neocomites peregrinus* zones). Smectite reaches contents of up to 41
175 % in this interval. Finally, the upper part of the section, which covers the late Valanginian interval
176 (*N. peregrinus*–*Criosarasinella furcillata* zones), shows relatively stable values in kaolinite and
177 smectite contents, with averages of 22 and 40 %, respectively.

178 The smoothed $\delta^{18}\text{O}_{\text{carb}}$ record is quite stable in the early Valanginian (Fig. 2), ranging from
179 -2.0 to -1.8‰ . The oxygen isotope data show a decreasing trend in the early–late Valanginian
180 transition, and reaches their lowest values (-2.4‰) at the $\delta^{13}\text{C}$ peak. The late Valanginian is
181 characterized by rather scattered $\delta^{18}\text{O}_{\text{carb}}$ values. However, a long-term positive trend is discernible,
182 going from -2.4 to -1.8‰ (amplitude of 0.6‰). The smoothed $\delta^{18}\text{O}_{\text{bel}}$ data show relatively stable

183 values (between -0.3 and -0.2 ‰) for the early and early-late Valanginian interval, while a
184 subsequent increase towards more positive ones (from -0.2 to 0.3 ‰) is observed in the remainder
185 of the late Valanginian.

186

187 3.1.2. Proto North Atlantic realm (~ 15 – 17°N)

188 Kaolinite, smectite, $\delta^{18}\text{O}_{\text{carb}}$ and TEX_{86} LOESS smoothing curves are shown in Fig. 3.

189 Throughout the Valanginian, clay mineral assemblages are largely dominated by smectite
190 (between 70 and 87 %) with minor contents of kaolinite (between 1 and 8 %) and palygorskite
191 (between 1 and 5 %). The smoothed $\delta^{18}\text{O}_{\text{carb}}$ record shows rather stable values (between -3.7 and
192 -3.5 ‰) in the early Valanginian. It is characterized by a prominent negative trend, ranging from
193 -3.6 to -4.5 ‰ at the early-late Valanginian transition, followed by a long-term positive trend
194 (from -4.5 to -3.6 ‰), in the late Valanginian. The smoothed TEX_{86} data displays quite stable
195 values (between 0.91 and 0.93) along the Valanginian interval.

196

197 3.1.3. Southern part of the Boreal realm (~ 35 – 45°N)

198 Kaolinite, smectite, $\delta^{18}\text{O}_{\text{lent}}$, $\delta^{18}\text{O}_{\text{biv}}$, and $\delta^{18}\text{O}_{\text{bel}}$ LOESS smoothing curves are reported in
199 Fig. 4.

200 The clay mineral assemblages are largely dominated by kaolinite (>70 %) in the early
201 Valanginian, and by smectite (up to 80 %) in the late Valanginian. Therefore, the early-late
202 Valanginian transition is characterized by the notable transition from kaolinite to smectite-
203 dominated sedimentation of clays.

204 The $\delta^{18}\text{O}_{\text{lent}}$ and $\delta^{18}\text{O}_{\text{biv}}$ signals show short-term fluctuations (from -0.70 to -1.58 ‰) in the
205 lower Valanginian. A subsequent increase to 0.53 ‰ is documented for the early-late Valanginian
206 transition, while in the early part of the late Valanginian, the $\delta^{18}\text{O}_{\text{lent}}$ and $\delta^{18}\text{O}_{\text{biv}}$ values display
207 rather stable values (from -0.39 to -0.16 ‰). Further up section, in the latest Valanginian interval,
208 they rise from -0.28 to 0.20 ‰. The smoothed $\delta^{18}\text{O}_{\text{bel}}$ curve shows a number of short-term

209 variations in the early and early–late Valanginian as well, with values ranging from -0.9 to -0.2 ‰.
210 It depicts a long-term positive trend from -0.6 to 0.4 ‰ in the remainder of the late Valanginian.

211

212 3.2. Southern Tethyan realm ($\sim 53^\circ\text{S}$)

213

214 The evolution of kaolinite, smectite, $\delta^{18}\text{O}_{\text{carb}}$, and TEX_{86} LOESS smoothing curves are
215 shown in Fig. 5.

216 The clay mineral fraction is dominated by smectite ($>90\%$), while kaolinite is absent. The
217 smoothed $\delta^{18}\text{O}_{\text{carb}}$ data are marked by a trend towards more positive values in the late Berriasian–
218 early Valanginian interval (from -2.3 to -1.6 ‰). They show rather stable values (between -1.7 and
219 -1.6 ‰) during the early–late Valanginian transition, and depict a small decreasing trend (from -1.6
220 to -1.3 ‰) along the late Valanginian, which is concomitant to a slight decreasing trend (from 0.66
221 to 0.63) in the smoothed TEX_{86} record.

222

223 4. Discussion

224

225 4.1. Significance of clay mineral and oxygen isotope signals

226

227 4.1.1. Diagenetic overprint

228 Mineralogical signals hold palaeoenvironmental information, which have possibly been
229 modified by diagenetic processes (e.g., Friedman and O’Neil, 1977; Chamley, 1989; Frank et al.,
230 1999; Thiry, 2000). Particularly, clay mineral compositions are sensitive to burial diagenesis, which
231 may be responsible for the progressive illitization and chloritization of smectite in marly interbeds
232 and limestone beds, respectively (Friedman and O’Neil, 1977; Deconinck and Debrabant, 1985).
233 Incipient illitization is characterized by the appearance of R1-type illite smectite mixed-layers as
234 soon as the maximum burial temperature reaches $60\text{--}70^\circ\text{C}$ (Šucha et al., 1993; Lanson et al., 2009).

235 In the studied sections, illitization processes have not been identified (Chamley et al., 1983;
236 Duchamp-Alphonse et al., 2011; Morales et al., 2015; Charbonnier et al., 2016) and smectite is
237 largely abundant instead. Besides, Rock-Eval analyses performed on selected samples indicate that
238 T_{\max} values never exceed 435 °C (Burtner and Warner, 1986; Morales et al., 2015; Charbonnier et
239 al., 2020), thus indicating a low thermal influence, below the oil window. These T_{\max} values are
240 consistent with the occurrence of irregular illite-smectite (I-S) R0 minerals with a high proportion
241 of smectite layers (>50 %), which indicates a weak thermal diagenesis (Dellisanti et al., 2010).
242 Therefore, it is reasonable to state that the impact of diagenesis on clay mineral signals was rather
243 limited, and that trends in this proxy reflect palaeoenvironmental conditions.

244 Published $\delta^{18}\text{O}_{\text{bel}}$ signals used here have already been previously evaluated for their capacity
245 to depict primary palaeotemperature signals using trace element geochemistry,
246 cathodoluminescence, and scanning electron microscope observations (Price et al., 2000, 2018; van
247 de Schootbrugge et al., 2000; Price and Mutterlose, 2004). The authors highlight no significant
248 diagenetic or compaction overprints, and suggest that the $\delta^{18}\text{O}_{\text{bel}}$ signal reflects the isotopic
249 composition of intermediate waters, belemnite being now considered as nektobenthic organisms
250 living below the thermocline (50 to 250 m) (e.g., Wierzbowski, 2002; Price et al., 2009, 2012;
251 Alberti et al., 2012; Jenkyns et al., 2012; Mutterlose et al., 2012; Meissner et al., 2015).

252 Extreme care is needed in the evaluation of the history of sediment preservation when using
253 $\delta^{18}\text{O}_{\text{carb}}$ as a palaeoenvironmental proxy. Primary whole-rock oxygen isotope ratios are sensitive to
254 differential compaction processes, as well as early meteoric and burial diagenesis. They all may
255 lead to fluid-rock interaction, neof ormation/recrystallization of the carbonate phases and thus,
256 isotopic resetting with later fluids. Rhythmic alternations of green/black marls and/or cherty layers
257 and limestones are typical sediments of (hemi-) pelagic basins and are specifically present as typical
258 facies throughout the Early Cretaceous. Some authors show that this bimodal lithologic pattern is
259 strongly affected by two contrasting diagenetic pathways during the early diagenesis (more
260 weathering-resistant limestone beds versus softer interbeds) (Reinhardt et al., 2000; Westphal et al.,

2000). Model simulations of Ricken (1986, 1987) suggest that the calcium carbonate cement in the limestone beds was sourced from calcium carbonate dissolution in the interlayers during the early diagenesis prior to the compaction. They demonstrate that the possible effect of differential compaction and early diagenetic changes acting on the precursors of these rhythmic marl-limestone alternations, may have seriously disturbed the original primary signal of $\delta^{18}\text{O}_{\text{carb}}$ signal (Westphal et al., 2008). Cyclostratigraphic investigations reveal that marl-limestone alternations in the Valanginian sediments mainly originate from cyclic changes of humidity/aridity conditions linked to Earth orbital cycles rather than diagenetic overprint (Cotillon et al., 1980; Sprovieri et al., 2006; Charbonnier et al., 2013; Martinez et al., 2013, 2015). Diagenetic overprint may also causes decreases of primary $\delta^{18}\text{O}_{\text{carb}}$ and $\delta^{13}\text{C}_{\text{carb}}$ values through the precipitation of variable amounts of diagenetic cements depleted in ^{18}O and ^{13}C , respectively (e.g., Veizer, 1983; Jenkyns and Clayton, 1986; Jenkyns, 1995; Godet et al., 2016; Benamara et al., 2020). Cross plots of $\delta^{18}\text{O}_{\text{carb}}$ - $\delta^{13}\text{C}_{\text{carb}}$ of the northwestern Tethyan, Proto North Atlantic, and southern Tethyan realms, reveal lack of correlations ($R^2=0.02$, 0.03 , and 0.17 , respectively) (Fig. 6). Besides, no negative or positive covariance exists between the $\delta^{18}\text{O}$ signal and the CaCO_3 contents in the ODP 765C (southern Tethyan realm) ($R^2=0.03$, Fig. S1 in supplementary online materials, SOM) as it would be expected from strong burial overprint (Frank et al., 1999). The $\delta^{18}\text{O}_{\text{carb}}$ values studied herein, are similar to those of open marine carbonates (from -4.0 to -1.0 ‰) and fall generally within the range of $\delta^{18}\text{O}$ values for sediments from the Early Cretaceous period (Van de Schootbrugge et al., 2000; Godet et al., 2006; Stein et al., 2011). Burial diagenesis overprint has not significantly affected the primary $\delta^{18}\text{O}_{\text{carb}}$ signal as suggested by the well-preserved clay mineral assemblage in the signal. Although this reinforces the assumption that the $\delta^{18}\text{O}_{\text{carb}}$ trends reflect an original signal, we cannot totally exclude a potential diagenetic overprint on $\delta^{18}\text{O}_{\text{carb}}$ absolute values, and thus on reconstructed palaeotemperatures. Consequently, we only consider the trends of the long-term LOESS-smoothing $\delta^{18}\text{O}_{\text{carb}}$ curves. In the same way as for published $\delta^{18}\text{O}_{\text{bel}}$ signals, and because the whole-rock carbonate fraction is mainly composed of planktic calcareous fossils, we interpret such trends as

287 average surface temperature changes (Godet et al., 2006; Duchamp-Alphonse et al., 2007; Bodin et
288 al., 2009; Stein et al., 2011).

289 Stable isotope analyses of foraminifera have been used to reconstruct temperatures at
290 various depths of the water column, depending on the habitat of the studied taxa. Because the shell
291 of *Lenticulina* spp. is prone to be filled with sediments, the $\delta^{18}\text{O}_{\text{lent}}$ can be contaminated either, by
292 the sediments itself either, by calcite dissolution processes associated with shell fragmentations.
293 Selected specimens of *Lenticulina* genera have previously been examined by scanning electron
294 microscopy (SEM) to assess the degree of diagenetic overprint (Morales et al., 2015). The authors
295 concluded that foraminifera are composed of primary calcite crystals, since SEM images of shell
296 structures show tests, pores, and chambers, free of secondary calcite, mural pores depicting well-
297 defined edges (Morales et al., 2015). *Lenticulina* is a cosmopolitan genus, with low ecological
298 requirements, that occupied a wide range of microhabitats (from epifaunal to deep infaunal habitats)
299 (Reolid et al., 2013). The $\delta^{18}\text{O}_{\text{lent}}$ signal might therefore reflect a vertical temperature gradient
300 between these microhabitats and absolute values in $\delta^{18}\text{O}_{\text{lent}}$ should be considered with extreme
301 caution. Consequently, we use the isotope trends as a benthic signal, in the only case that they are in
302 well agreement with $\delta^{18}\text{O}_{\text{carb}}$, and $\delta^{18}\text{O}_{\text{bel}}$ ones.

303

304 4.1.2. Kaolinite and smectite as palaeoclimate proxies

305 Kaolinite and smectite represent the major clay components in the studied sections.
306 Palygorskite is observed only at the DSDP Sites retrieved in the Proto North Atlantic realm.

307 The distribution of kaolinite versus smectite is partly controlled by their differential settling
308 behaviour, which is related to physical segregation during their transport as a function of
309 differences in size and flocculation capacity (Thiry, 2000; Adatte et al., 2002; Godet et al., 2008).
310 Indeed, kaolinite is a dense and coarse mineral that settles preferentially closer to the coast, in
311 shallow-water environments. Conversely, smectite is a fine-grained mineral that tends to settle
312 much more slowly, in deeper waters and more offshore settings. Consequently, differential settling

313 of kaolinite versus smectite may explain the distribution of clay mineral assemblages along a
314 platform/basin transect (Thiry, 2000). The sections selected here present lithologies reflecting
315 hemipelagic, pelagic, or abyssal settings (Table 1).

316 Sedimentary records in the Proto North Atlantic (~15–17°N) realm, situated in pelagic
317 environments, are characterized by high smectite contents (>80%) and the absence of kaolinite. In
318 strong contrast, kaolinite constitutes one of the main phases of the clay mineral assemblages in the
319 hemipelagic successions from the northwestern Tethyan (~20°–30°N) and southern part of the
320 Boreal (~35–45°N) realms. Based on these observations, differential settling of clay particles may
321 be postulated as a cause for relatively high smectite (kaolinite) abundances in pelagic (hemipelagic)
322 environments. However, as for several clay minerals that are known to have localized distributions,
323 the relative abundances of smectite and kaolinite follow a climate related latitudinal pattern (Ito and
324 Wagai, 2017). Smectite is predominantly formed under dry and seasonally contrasted conditions, in
325 mid- and high latitudes. Kaolinite used to precipitate under humid conditions, particularly in
326 subtropical environment. The occurrence of lateritic and bauxite deposits in the northwestern
327 Tethyan realm supports the development of intense biogeochemical weathering and the presence of
328 kaolinite. At the opposite, the presence of evaporitic and calcrete deposits in sediments from the
329 Proto-North Atlantic realm is a further argument for widespread aridity in this region during the
330 Early Cretaceous (Chumakov et al., 1995; Scotese, 2001; Hay and Floegel, 2012), and for the
331 climate-related occurrence of both high amount of smectite and palygorskite in the clay mineral
332 assemblages. Thus, the absence of kaolinite in pelagic deposits may be well explained by climatic
333 conditions and not necessarily only by differential settling processes. Therefore, the smoothed
334 smectite and kaolinite contents are proposed to depict the weathering conditions that prevailed for
335 each basin (Figs 2–4).

336 In addition, the kaolinite/smectite ratio can be also modulated by sea-level changes during
337 regressive/transgressive (proximal/distal) cycles (sources) (Adate et al., 2002). There are some
338 significant discrepancies between the recent reconstructions of Cretaceous global sea-level changes.

339 According to Haq (2014), the early Valanginian is represented by low amplitude sea level changes,
340 while the late Valanginian is characterized by a long-term sea-level rise. In their recent review, Ray
341 et al. (2019) document a progressive increase in the magnitude of sea-level changes from the late
342 Berriasian to the early Valanginian (e.g., Sahagian et al., 1996; Gréselle and Pittet, 2010;
343 Dujoncquoy et al., 2018), followed by a major sea level fall (~100-150 m) the late early
344 Valanginian (*Campylotoxus ammonite Zone*), and a significant sea-level rise at the early-late
345 Valanginian transition (Gréselle and Pittet, 2010; Dujoncquoy et al., 2018). According to these
346 authors, the late Valanginian would be characterized by a sea-level highstand marked by a series of
347 lower-amplitude fluctuations (between 25 and 50 m). Despite these discrepancies, it appears that the
348 trends in sea-level changes are poorly correlated to the distributions of kaolinite and smectite
349 contents reported herein (Fig. S2 in supplementary online materials), and when major mineralogical
350 redistributions occur, particularly within the northwestern Tethyan realm, they are usually not
351 consistent with a sea-level influence. The prominent sea-level fall reported in the late-early
352 Valanginian by Ray et al. (2019) is associated with relatively stable kaolinite and smectite contents
353 (Fig. S2 SOM), while the major sea-level rise documented at the early-late Valanginian transition
354 corresponds to a significant long-term increase in kaolinite contents. During the late Valanginian,
355 the significant sea-level rise documented by Haq (2014) is associated with increasing smectite
356 contents. In such a case, clay mineral assemblages could be influenced by sea-level changes rather
357 than climatic conditions. However, palynological investigations in the Vocontian Basin, considered
358 as a key climatic indicator, attest to a reduction of hydrolysing conditions in this realm, which is
359 rather consistent with the increased smectite pattern observed during this time interval (Kujau et al.,
360 2013).

361 Consequently, in analogy to the clay distribution in modern oceans (Ito and Wagai, 2017),
362 and because local sedimentary and diagenetic overprints have been shown to bear less weight, we
363 assume that the stratigraphic distribution of clay minerals captures general patterns in weathering
364 intensity at a basin scale. However, caution is in order due to the differential settling overprint and

365 sea-level fluctuations.

366 Furthermore, it should be noted here that halmyrolytic alteration of volcanic glass or basalt
367 may result in the neoformation of smectite (Chamley, 1989). Since widespread magmatism
368 occurred along the northwestern Australian passive margin due to rifting during the Early
369 Cretaceous, it is not excluded that smectite retrieved from Hole 765C (Fig. 5) has a volcanic origin
370 (Compton and Locker, 1992). The distribution of smectite at this site should be treated in this
371 perspective.

372

373 4.1.3. Oxygen isotopes as a potential palaeothermometer

374 Once diagenetic overprint is excluded, the $\delta^{18}\text{O}$ data obtained on carbonates from whole
375 rock samples or belemnite rostra can be related to: (i) temperatures in the environmental setting; (ii)
376 $\delta^{18}\text{O}_w$ of seawaters as a function of ice sheet volume and freshwater input; or (iii) a mixture of both
377 (e.g., Frakes and Francis, 1988; Henderson et al., 2000; Mutterlose et al., 2010; Price et al., 2012;
378 Meissner et al., 2015). In Mesozoic series, bulk-rock oxygen isotope signals have been widely used
379 to document changes in average SST, and this has been especially the case when the $\delta^{18}\text{O}$ variations
380 were in line with overall paleoenvironmental reconstructions (Godet et al., 2006; Duchamp-
381 Alphonse et al., 2007; Bodin et al., 2009; Stein et al., 2011; Charbonnier et al., 2016). We used the
382 paleo-temperature equation of Anderson and Arthur (1983) after Epstein et al. (1953) and Craig and
383 Gordon (1965):

384

$$385 \quad T(^{\circ}\text{C}) = 16.0 - 4.14 (\delta^{18}\text{O}_c - \delta^{18}\text{O}_w) + 0.13 (\delta^{18}\text{O}_c - \delta^{18}\text{O}_w)^2$$

386

387 Where $\delta^{18}\text{O}_c = \delta^{18}\text{O}$ vs. VPDB of calcite and $\delta^{18}\text{O}_w = \delta^{18}\text{O}$ vs. VSMOW of seawater. Estimating
388 $\delta^{18}\text{O}_w$ is problematic because it is variable through geological time (Hudson and Anderson, 1989).
389 We have reported $\delta^{18}\text{O}_w$ values based on the simulation of latitudinal Cretaceous surface seawater
390 $\delta^{18}\text{O}_w$ from Zhou et al. (2008) and derived $\delta^{18}\text{O}_{\text{seawater}}$ trends calculated by Price et al. (2018). We

391 used a $\delta^{18}\text{O}_w$ ranging from 0 and -0.5‰ for seawater located at a paleolatitude between ~ 38 and
392 45°N , 0.5‰ for seawater situated in the Proto-North Atlantic realm, and -1.5‰ for seawater
393 located between ~ 50 and 70°S (Zhou et al., 2008). Recently, Price et al. (2018) have demonstrated
394 that the composition of seawater from the northwestern Tethyan realm fluctuated around 1.0‰
395 with more positive $\delta^{18}\text{O}_{\text{seawater}}$ values in the *Neocomiensiformis* and *Furcillata-Loryi* Zones and
396 more negative values in the *Verrucosum* Zone. Consequently, we estimate the amplitude of SST
397 change with $\delta^{18}\text{O}_w$ values ranging from 0.4 to 1.4‰ (Price et al., 2018) (Table 2).

398

399 4.2. Latitudinal palaeoclimate reconstructions in the northern hemisphere

400

401 4.2.1. Early Valanginian

402 During the early Valanginian, both kaolinite and smectite contents as well as $\delta^{18}\text{O}_{\text{carb}}$
403 document no significant temporal changes in the studied realms (i.e. northwestern Tethyan, the
404 Proto North Atlantic, and the southern part of the Boreal) (Fig. 7). It appears that weathering
405 intensity and seawater temperatures have remained stable during this time interval. These results
406 confirm previous TEX_{86} measurements (Littler et al., 2011), palynological investigations (Kujau et
407 al., 2013), and recently reported Mg/Ca ratios (Price et al., 2018), which highlight stable
408 temperature conditions within the Proto-North Atlantic and the northwestern Tethyan realms,
409 respectively.

410 Interestingly, kaolinite and smectite distributions show significant variations according to
411 the studied basin (Figs. 2, 3, and 4). In a similar way as for modern oceans, these regional
412 modifications are tentatively explained by the presence and influence of latitudinal climate belts
413 with contrasted hydrolysing conditions on continents (Fig. 8). The high abundance of kaolinite in
414 sediments from the northwestern Tethyan (up to 27%) and southern part of the Boreal (up to 74%)
415 realms suggests the existence of an extensive humid subtropical climate belt (at least) ~ 20 to 45°N
416 (Fig. 8). By contrast, in the Proto North Atlantic realm, kaolinite is uncommon and never exceeds 8

417 % (Zemmels et al., 1972; Chamley, 1981; Chamley et al., 1983; Debrabant et al., 1984; Huff,
418 1993). Large smectite proportions (>70 %) associated with the occurrence of palygorskite might
419 instead indicate, that a semi-arid climate belt prevailed at about 15°N (Fig. 8).

420

421 4.2.2. Early–late Valanginian transition

422 The early–late Valanginian transition is characterized by a rise in kaolinite contents in the
423 northwestern Tethyan realm (up to 12 ± 5 %), which supports the development of wetter conditions,
424 as previously documented by local clay mineral, palynological and geochemical data (Kuhn et al.,
425 2005; Duchamp-Alphonse et al., 2011; Kujau et al., 2013; Charbonnier et al., 2016). Further north
426 (at ~45°N), this time interval is marked by the replacement of kaolinite by smectite (from 1 to 74
427 %), highlighting an abrupt change from high to low weathering intensities and thus, from humid to
428 dry and seasonally contrasted conditions (Fig. 7). This is well supported by change in spore and
429 pollen assemblages showing an evolution toward more arid climate (Kujau et al., 2013). These
430 spatio-temporal changes in the weathering mode may point to the southward migration of a
431 monsoon-like subtropical rain belt during the Weissert episode, as previously proposed by
432 Duchamp-Alphonse et al. (2014), Morales et al. (2015), and Charbonnier et al. (2016), based on
433 local climate reconstructions. Our reconstruction permits to postulate a southward shift of
434 minimally 15° in latitude (from 45 to 30°N) (cf., Morales et al., 2015). At the opposite, as
435 documented by the dominance of smectite (>80 %) and palygorskite (>8 %) in the Proto-North
436 Atlantic realm, climate conditions have remained stable and the semi-arid climate belt has prevailed
437 in this part (Figs. 7 and 8).

438 Interestingly, clear long-term decreases in whole rock $\delta^{18}\text{O}$ values have been observed in the
439 northwestern Tethyan (amplitude: 0.53 ± 0.18 ‰), and the Proto North Atlantic realms (amplitude:
440 0.95 ± 0.18 ‰) (Figs. 2 and 3). Such coherent trends clearly exclude a significant overprint of the
441 smoothed- $\delta^{18}\text{O}_{\text{carb}}$ curves by changes in the local water balance of $\delta^{18}\text{O}_{\text{w}}$ and salinity. Shifts from
442 more arid to more humid climate modes may explain the heavier ocean-surface $\delta^{18}\text{O}$ values because

443 of increased continental freshwater storage linked to a phase of accelerated hydrological cycling
444 (more strongly ^{18}O -depleted precipitation) (Föllmi, 2012; Wagreich et al., 2014; Wendler et al.,
445 2016). However, clay mineralogical distributions suggest the occurrence of specific climate belts
446 with contrasted weathering regime, which is not in favour of a global climate transition mode
447 between more arid to more humid conditions. Moreover, during this time interval, the calculated
448 $\delta^{18}\text{O}_{\text{seawater}}$ values from sections in France and Spain are quite stable (Price et al., 2018), which
449 highlight that the decreases in the $\delta^{18}\text{O}$ whole-rock signal have not been only linked to changes in
450 precipitation/evaporation rates. Consequently, an overall warming phase, characterized by
451 increasing seawater temperatures seems to be the most plausible explanation. In strong contrast,
452 oxygen-isotope analyses performed on the selection of hand-picked monogeneric benthic
453 foraminifera (*Lenticulina* spp.) and bivalves in the southern Boreal realm exhibit a significant
454 positive $\delta^{18}\text{O}$ excursion (Fig. 4). This trend is coeval to the abrupt change in clay mineral
455 assemblages (from kaolinite to smectite dominance), which indicate a switch from humid to dry and
456 seasonally contrasted conditions. A decrease in the flux of freshwater during time of decreased
457 humidity lead to more positive $\delta^{18}\text{O}$ values in general. Consequently, the enhanced $\delta^{18}\text{O}$ values
458 could reflect local variations in salinity due to the decreased influx of freshwater input into the
459 ocean (Morales et al., 2015) (Fig. 4). Alternatively, one might argue that benthic organisms may
460 simply reflecting colder bottom waters or a diagenetic signal.

461 Applying the equation of Anderson and Arthur (1983) and using various $\delta^{18}\text{O}_w$ values
462 according to the latitude (Zhou et al., 2008; Price et al., 2018 and see chapter 4.1.3), results in
463 temperature increases of about $3\pm 0.9^\circ\text{C}$ in the northwestern Tethyan and about $5\pm 0.9^\circ\text{C}$ in the Proto
464 North Atlantic (Table 2). As such, warming appears much more moderate in the northwestern
465 Tethyan compared to the Proto-North Atlantic. This confirms the lower temperature variability
466 recently obtained by belemnite Mg/Ca ratios in the Subbetic Domain and the Vocontian basin
467 during the Weissert Episode (Price et al., 2018). However, these temperatures and changes therein
468 are in contradiction with TEX_{86} records from the Proto North Atlantic realm, which report stable

469 temperature conditions during the early-late Valanginian transition period (Littler et al., 2011). The
470 causes for this difference can be related to a diagenetic alteration of the $\delta^{18}\text{O}_{\text{carb}}$ values recorded by
471 the whole rock signal (see chapter 4.1.1). Alternatively this discrepancy may be related to a
472 seasonal bias (Sluijs et al., 2006). The TEX_{86} proxy is based on the chrenarchaeotal membrane lipid
473 distribution in sedimentary rocks, which is dependent on SST (Schouten et al., 2003). However, the
474 highest fluxes of chrenarchaeotal lipids coincide with periods of high phytoplanktonic productivity
475 and are recorded during summer months (Sluijs et al., 2006). Therefore, the cell number in the
476 water column depends on the season, and the TEX_{86} signal may record an average of the high
477 summer SSTs (Sluijs et al., 2006), prone to changes with relatively small amplitudes. In all cases,
478 changes in SST conditions observed in the oxygen-isotope records attest to a warming phase in the
479 northern hemisphere. Together with the weathering patterns documented by clay mineral data, they
480 highlight temperate to semi-arid conditions in the Proto North Atlantic, and humid and subtropical
481 conditions related to a monsoon-like system in the northwestern Tethyan (Fig. 8).

482

483 4.2.3. Late Valanginian

484 During the late Valanginian, kaolinite contents remain low within the Proto North Atlantic
485 and the southern part of the Boreal realms (<5 %), while they significantly decrease (from 39 to 16:
486 23 ± 5 %) in the northwestern Tethyan realm (Fig. 7). This latter realm appears more sensitive to
487 changes in hydrological cycling and subsequent erosion and runoff processes, which is consistent
488 with the idea of the presence of a monsoon-like system. In a wider perspective, smectite dominates
489 clay mineral assemblages, which points to a diminution in the weathering intensity in the northern
490 hemisphere, related to the reduction of hydrolysis conditions.

491 The composite whole rock $\delta^{18}\text{O}$ values in the northwestern Tethyan and in the Proto North
492 Atlantic realms show long-term trends towards heavier values (amplitudes of 0.58 ± 0.18 ‰ and
493 0.88 ± 0.18 ‰, respectively) (Figs. 2 and 3). These trends probably indicate declines in
494 palaeotemperatures of $\sim 3\pm 0.9$ and $5\pm 0.9^\circ\text{C}$, respectively (see chapter 4.1.3 and Fig. 7). The

495 difference between whole-rock and TEX₈₆ records can be explained by a substantial seasonal bias
496 as suggested above (see chapter 4.2.2). Oxygen-isotope records obtained on the benthic
497 foraminifera (*Lenticulina* spp.) and bivalves in the southern part of the Boreal realm exhibit similar
498 trends than those documented by the $\delta^{18}\text{O}_{\text{carb}}$ (amplitude: 0.48 ± 0.18 ‰). They imply a cooling of
499 $2^{\circ}\text{C}\pm 0.9^{\circ}\text{C}$ of bottom waters during the latest Valanginian (Fig. 6), which is in the same range of
500 magnitude as those previously obtained from the oxygen-isotopic composition and Mg/Ca ratio of
501 belemnites in the northwestern Tethyan realm ($\sim 2\pm 0.6^{\circ}\text{C}$, McArthur et al., 2007; Price et al., 2018)
502 and oxygen-isotopic composition of benthic fish teeth ($\sim 3^{\circ}\text{C}$, Pucéat et al., 2003; Barbarin et al.,
503 2012). These are also consistent with combined $\delta^{18}\text{O}_{\text{bel}}$ data sets from the Boreal realm, which
504 document a significant cooling (~ 2 to $\sim 4\pm 0.6^{\circ}\text{C}$) from 35 to 65° N, during the late Valanginian
505 (Meissner et al., 2015) (Fig. 7).

506 From a global perspective, it appears that during the late Valanginian, a warm temperate belt
507 probably developed in the Boreal, at the expense of the subtropical humid belt of the northwestern
508 Tethys (Fig. 8). Finally, a stable semi-arid climate belt in the Proto-North Atlantic (~ 15 - 17°N)
509 occurred throughout the Valanginian (Fig. 8). Interestingly, the palaeoclimatic belts that are
510 proposed herein, and based upon high-resolution mineralogical and geochemical data are consistent
511 with those proposed by Chumakov et al. (1995) and Scotese (2001) for the Early Cretaceous, which
512 were based on sedimentological, mineralogical and paleontological evidences. The occurrence of
513 lateritic and bauxite deposits in the northwestern Tethys highlights a humid subtropical belt at ~ 20 –
514 35°N of latitudes. A semi-arid belt is defined in the Proto North Atlantic, with the presence of
515 evaporites and calcretes. Finally, a warm temperate belt is documented in the high latitudes from
516 ~ 35 to 65°N (Fig. 8).

517 518 4.3. Palaeoclimatic reconstruction in the southern hemisphere

519
520 At ODP Site 765C ($\sim 53^{\circ}\text{S}$), the stable amounts of smectite throughout the entire

521 Valanginian may have resulted from the alteration of basalt and volcanic ash derived from the
522 northwestern Australian passive margin (see chapter 4.1.2). However, the clay mineral assemblages
523 in the Weddell Sea basin (~60°S) document the presence of alternating wet and arid seasons
524 (Robert and Maillot, 1990), thus suggesting that installation of a temperate belt in this realm (Fig.
525 8). Interestingly, the smoothed $\delta^{18}\text{O}_{\text{carb}}$ data displays no significant variations throughout the early
526 and early-late Valanginian transition (Fig. 5), documenting rather constant temperature conditions
527 (Fig. 7). These results are consistent with seawater palaeotemperature estimations obtained from the
528 Austral Basin (Argentina, ~60°S), based on $\delta^{18}\text{O}_{\text{bel}}$ signals, and indicating constant temperatures of
529 ~25°C during the early Valanginian (Gómez-Dacal et al., 2019). During the late Valanginian, the
530 composite $\delta^{18}\text{O}_{\text{carb}}$ signal of ODP Site 765C shows trend towards heavier values (amplitudes of
531 $0.3 \pm 0.18 \text{ ‰}$) (Fig. 5), which may indicate a decline in palaeotemperatures of $\sim 2 \pm 0.9^\circ\text{C}$. Such
532 cooling event appears to be in agreement with the slight cooling trend documented by TEX_{86} data in
533 the ODP Site 766 ($\sim 1^\circ\text{C}$; Littler et al., 2011) (Fig. 5) and the occurrence of *cheirolepidiaceae* leaves
534 and pollen in the Gippsland and Otway Basins (southeastern Australia) (Tosolini et al., 2014),
535 which testify for cool and moist conditions in the region during that time interval. It is also
536 confirmed by recent chronostratigraphic investigations of sedimentary sequences from the southern
537 and western margins of the Eomanga Basin (~70°S), which indicate a late Valanginian age for
538 glaciogenic facies, and more particularly for the Living Tillite Member (Alley et al., 2019). In all
539 cases, these results draw the scenario of a global cooling in both hemispheres during the late
540 Valanginian.

541

542 4.4. Valanginian climate conditions: a complex interplay between regional versus global
543 mechanisms

544

545 Our compilation of published and unpublished clay mineral and $\delta^{18}\text{O}_{\text{carb}}$ data allows for a
546 better appraisal of global climate conditions during the Valanginian. The low- to mid-latitudes of

547 northern hemisphere are characterized by the presence of two main rainfall belts: a humid
548 subtropical one associated with a monsoon-like system bringing significant precipitations into the
549 Tethyan realm, as testified by the important proportions of kaolinite in the area from sites between
550 (at least) ~20 to 45°N; and a sub-arid one, characterized by dry conditions in the Proto North
551 Atlantic realm, as documented by prominent proportions of smectite and palygorskite at sites ~15–
552 17°N. Temperate conditions may have prevailed around 45°N and up to ~60°N.

553 Both hemispheres present distinct features, with a lower temperature gradient through the
554 early Valanginian in the southern hemisphere than in the northern one, as recently proposed by
555 Gómez-Dacal et al. (2019). The distribution of climate sensitive sediments also demonstrates that a
556 complex system develops in the northern hemisphere whereas low- to mid-latitudes of the southern
557 hemisphere may probably be under the broad influence of an arid climate belt (Chumakov et al.,
558 1995) (Fig. 8). The climatic asymmetry of hemispheres existed throughout the geological history
559 (Chumakov, 2004). These general climate conditions are probably related to the overall continental
560 configuration and particularly the distribution of the oceanic gateways, which modulate circulation
561 pattern and heat transfer at regional and global scales (Poulsen et al., 2001, 2003; Hotinski and
562 Toggweiler, 2003; Donnadieu et al., 2006; Goddérís et al., 2014). One of the causes of the
563 discrepancies in climate evolution between both hemispheres is certainly the fact that the northern
564 hemisphere was mainly continental, with marine environments mainly restricted to several smaller
565 basins, whereas the southern hemisphere is divided into one larger continent and a very large ocean.
566 Particularly, during the earliest Cretaceous, new seaways formed between the Tethyan, the Boreal,
567 and the Proto North Atlantic realms, due to the fragmentation of the Pangaea (Le Pichon and
568 Huchon, 1984; Dercourt et al. 1986). A new connection allowed for the establishment of the
569 westward Tethys Circum global Current (TCC) (Pucéat et al., 2005), which derived from tropical
570 epicontinental seas around ~20–30°S and brought warm and saline waters to the proto-North
571 Atlantic Ocean. During the Valanginian, the combination of warm saline surface waters and
572 restricted palaeogeography in the Proto North Atlantic Ocean probably acted as preconditioning

573 factors for the establishment of a stable and semi-arid belt (Barron et al., 1985; Ziegler et al., 1987;
574 Hay, 2000, 2008). Under such conditions, the climate is also more sensitive to monsoonal dynamics
575 due to atmospheric pressure gradients (e.g., Hay, 2008). This may have been the case within the
576 northwestern Tethyan realm, where the arrangement of continents north and south of the wide
577 Tethys seaway provided the ideal geographic configuration for significant land–sea pressure
578 contrasts via the different thermal inertia of land and ocean in this area. These conditions may have
579 helped maintaining seasonally reversing movements of air mass, heat and precipitation (Oglesby
580 and Park, 1989), as supported by the important kaolinite deposition in Tethyan and Boreal
581 sediments in contrast to the Proto North Atlantic (Charbonnier et al., 2016) (Figs. 7 and 8).

582 Superimposed on these regional mechanisms, many studies suggested a link with the
583 intensification of the Paraná-Etendeka (South America-South Africa) volcanic activity (Lini et al.,
584 1992; Föllmi et al., 1994; Erba et al., 2004). An update of the Valanginian/Hauterivian
585 astrochronological time scale confirms a potential link between the main phase of Paraná-Etendeka
586 volcanic activity (-134.6 ± 0.6 ; -134.3 ± 0.8 Ma; Thiede and Vasconcelos, 2010; Janasi et al., 2011)
587 and the beginning of the Weissert episode (-135.22 ± 1 Ma) (Martinez et al., 2015). Moreover, a set
588 of mercury anomalies in Valanginian sediments recovered from localities outside and in European
589 sections demonstrated that a main phase of magmatism occurred at the onset of the Weissert
590 episode (Charbonnier et al., 2017, 2020). Our study highlights a coherent fingerprint of the Paraná-
591 Etendeka emplacement on climate change across the Weissert Episode. Indeed, the global warming
592 observed during the Weissert episode is the typical consequence of increased emissions of
593 greenhouse gases from a LIP volcanism. In contradiction to Price et al. (2018), it appears that
594 elevated $p\text{CO}_2$ levels during the Weissert episode and associated warming may have actually
595 accelerated the hydrological cycle and enhanced nutrient input from continents to ocean. Ocean
596 fertilization associated with excess of CO_2 in surface waters (i.e., surface-ocean acidification) may
597 have induced marine carbonate production crisis in both in neritic and pelagic domains (Föllmi et
598 al., 1994; Weissert et al., 1998; Erba et al., 2004; Weissert and Erba, 2004; Barbarin et al., 2012;

599 Duchamp-Alphonse et al., 2014). On the continent the development of greenhouse conditions
600 probably led to the implantation of an important vegetation cover from mid to high latitudes, as
601 highlighted by widespread coal deposits in the sedimentary records (Ziegler et al., 1987; McCabe
602 and Totman Parish, 1992; Scotese, 2001; Westermann et al., 2010) (Fig. 8). Particularly, dense
603 conifer forests of Valanginian age were documented in the Australian (~53–85°S) and Canadian
604 domains (~74–79°N) (Dettmann et al., 1992; McLoughlin and Hill, 1996; McLoughlin et al., 2002;
605 Galloway et al., 2013, 2015; Tosolini et al., 2014) (Fig. 8). The presence of extensive polar forests
606 probably induced positive feedbacks between climate and vegetation, reducing surface albedo and
607 increasing adsorption of solar radiation at high latitudes (Upchurch et al., 1998). During the
608 Valanginian these factors might have also played an important role in maintaining high-latitude
609 warmth, which might have induced the presence of a stable warm temperate belt in high latitudes
610 (Fig. 8). This is supported by clumped-isotope analyses from belemnites, which testify for warm
611 polar conditions (10–20°C) at 60–65°N (Price and Passey, 2013). The long-lasting and global burial
612 of terrestrial organic carbon consuming a significant amount of atmospheric CO₂, may have
613 contributed to the global cooling observed in both hemispheres, during the last stages of the
614 Weissert episode (Figs. 7 and 8). In fossil plant material, from the late Valanginian, Gröcke et al.
615 (2005) attested cool terrestrial conditions caused by a decrease in atmospheric *p*CO₂ levels.
616 Stratigraphic and sedimentological evidences suggest that this global cold event may have help
617 developing the Livingston glaciation observed in high southern latitudes during the late
618 Valanginian–Hauterivian interval (Alley et al., 2019). One cannot exclude that this time interval
619 might also be punctuated by limited extension of polar ice cap in the high northern latitudes, as
620 previously documented by the occurrence of glendonites, tillites, and dropstones described in
621 Svalbard (~65–70°N; Kemper and Schmitz, 1975; Price and Nunn, 2010; Vickers et al., 2019), in
622 the Sverdrup Basin (~74–79°N; Kemper, 1987; Price, 1999), and in Siberia (Price, 1999). Such cold
623 conditions could have caused the formation of a new deep-water ocean current system through the
624 opening of the Greenland-Norwegian seaway (Pauly et al., 2012).

625

626 **5. Conclusions**

627

628 The set of published and unpublished mineralogical and geochemical data presented in this
629 study allows to show the reliability of clay and $\delta^{18}\text{O}_{\text{carb}}$ data as good palaeoclimate proxies for the
630 Valanginian.

631 In the northern hemisphere, the kaolinite and smectite distributions obtained through the
632 Valanginian show significant variations, which can be tentatively explained by the presence of
633 latitudinal climatic belts characterized by contrasted hydrolysing conditions. It appears that stable
634 semi-arid conditions prevailed through the Valanginian in the Proto North Atlantic realm (~15–
635 17°N), and a humid subtropical climatic belt was present between (at least) ~20 and 45°N. Spatio-
636 temporal changes in the weathering mode (from ~30 to 45°N), from humid to dry and seasonally
637 contrasted conditions, clearly points to the southward migration of a monsoon-like rain tropical belt
638 in the northwestern Tethyan realm during the Weissert episode. The general trends in the $\delta^{18}\text{O}_{\text{carb}}$
639 records reveal that the northern hemisphere was characterized by an overall warming phase at the
640 early–late Valanginian transition, with temperatures increasing between ~3 and $5\pm 0.9^\circ\text{C}$. Our data
641 suggest also a significant and generally long-term cooling trend throughout the late Valanginian
642 between ~1 and $5\pm 0.9^\circ\text{C}$ in both hemispheres. In strong contrast, it appears that the amplitude of
643 climatic changes recorded during the Valanginian was probably less pronounced in the southern
644 hemisphere than in the northern one.

645 Climate evolution during the Valanginian resulted from the complex interplay between
646 regional, global, palaeogeographic and palaeoceanographic impacts and changes therein, which
647 influenced the exchange of currents, and especially heat flow, and which triggered the development
648 of a dense vegetation cover on the continents, capable of preserving excess carbon in the form of
649 terrestrial organic matter (leading to the formation of widespread coal deposits). However, further
650 investigations are needed in the southern hemisphere and at a global scale, to better detail the
651 climate regime of the Valanginian.

652

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654

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660

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662

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1104 **Figure captions**

1105
1106 **Fig. 1:** World palaeogeographic map of the Early Cretaceous (modified after Blakey, 2010).
1107 Locations of the studied sections in four depositional environments at several palaeolatitudes (see
1108 Table 1): (1) Wawal core (Polish Basin), (2) Angles section (Vocontian Basin), (3) Orpierre section
1109 (Vocontian Basin), (4) Barranco del Garranchal section (Subbetic Basin), (5) Molina de las Oicas
1110 section (Subbetic Basin), (6) DSDP Hole 603B (Continental rise Cape Hatteras), (7) DSDP Hole
1111 534A (Blake Bahama Basin), (8) DSDP Hole 535 (Gulf of Mexico), (9) ODP Hole 765C (Argo
1112 abyssal plain), (10) Speeton section (Yorkshire), (11) Northwest Germany composite section
1113 (Lower Saxony Basin), (12) Northern Germany composite section (Proto-North Sea Basin), (13)
1114 Vergol/La Charce sections (Vocontian Basin), (14) Vocontian composite section (Vocontian Basin),
1115 (15) Subbetic composite section (Subbetic Basin), (16) Umbria Marche composite section (Umbria
1116 Marche Basin), (17) Bersek Quarry (Gerecse Mountains), (18) Kozoskut ravine section (Bakony
1117 Mountains), (19) Cehegín section (southern Iberian continental margin) (20) ODP Hole 766A
1118 (Gascoyne and Cuvier abyssals plains).

1119
1120 **Fig. 2:** Synthesis diagram showing kaolinite, smectite, $\delta^{18}\text{O}_{\text{carb}}$ and $\delta^{18}\text{O}_{\text{bel}}$ LOESS smoothing

1121 curves from the northwestern Tethyan realm with the carbon isotope stratigraphy against the
1122 biostratigraphic scheme. LOESS, i.e. locally weighted scatterplot smoothing (Cleveland and Devlin,
1123 1988). For the calculation of LOESS smoothing curve (red line) an α value of 0.15 has been
1124 applied. Blue curves indicate the 95 % confidence interval. SST: Sea Surface Temperature, SWT:
1125 Sea Water Temperature.

1126

1127 **Fig. 3:** Synthesis diagram showing kaolinite, smectite, $\delta^{18}\text{O}_{\text{carb}}$ and TEX_{86} LOESS smoothing
1128 curves from the Proto North Atlantic realm with the carbon isotope stratigraphy against the
1129 biostratigraphic scheme. LOESS, i.e. locally weighted scatterplot smoothing (Cleveland and Devlin,
1130 1988). For the calculation of LOESS smoothing curve (red line) an α value of 0.15 has been
1131 applied. Blue curves indicate the 95 % confidence interval. SST: Sea Surface Temperature.

1132

1133 **Fig. 4:** Synthesis diagram showing kaolinite, smectite, $\delta^{18}\text{O}_{\text{lent}}$ and $\delta^{18}\text{O}_{\text{bel}}$ LOESS smoothing curves
1134 from the southern Boreal realm with the carbon isotope stratigraphy against the biostratigraphic
1135 scheme. LOESS, i.e. locally weighted scatterplot smoothing (Cleveland and Devlin, 1988). For the
1136 calculation of LOESS smoothing curve (red line) an α value of 0.15 has been applied. Blue curves
1137 indicate the 95 % confidence interval. SST: Sea Surface Temperature, SWT: Sea Water
1138 Temperature.

1139

1140 **Fig. 5:** Synthesis diagram showing kaolinite, smectite, $\delta^{18}\text{O}_{\text{carb}}$ and TEX_{86} LOESS smoothing
1141 curves from the southern Tethyan realm with the carbon isotope stratigraphy against the
1142 biostratigraphic scheme. LOESS, i.e. locally weighted scatterplot smoothing (Cleveland and Devlin,
1143 1988). For the calculation of LOESS smoothing curve (red line) an α value of 0.15 has been
1144 applied. Blue curves indicate the 95 % confidence interval. SST: Sea Surface Temperature.

1145

1146 **Fig. 6:** Cross-plot of the $\delta^{13}\text{C}_{\text{carb}}$ and the $\delta^{18}\text{O}_{\text{carb}}$ values from the northwestern Tethyan, Proto North
1147 Atlantic and southern Tethyan realms.

1148

1149 **Fig. 7:** Evolution of kaolinite contents and temperature ranges during (i) the early Valanginian, (ii)
1150 the early-late Valanginian transition, and (iii) the late Valanginian. Palaeogeographic map are from
1151 Blakey (2010).

1152

1153 **Fig. 8:** Synthetic map (modified after Blakey, 2010) illustrating the Valanginian palaeoclimatic
1154 belts supported by geochemical and mineralogical data during the early Valanginian, the early-late
1155 Valanginian transition, and the late Valanginian. Location of Berriasian, Valanginian and
1156 Hauterivian coal deposits are reported after McCabe and Totman-Parrish (1992). Distribution of
1157 conifer forest are from Dettmann et al. (1992); McLoughlin and Hill (1996); McLoughlin et al.
1158 (2002); Galloway et al. (2013, 2015); and Tosolini et al. (2014). Distribution of glendonites,
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1162

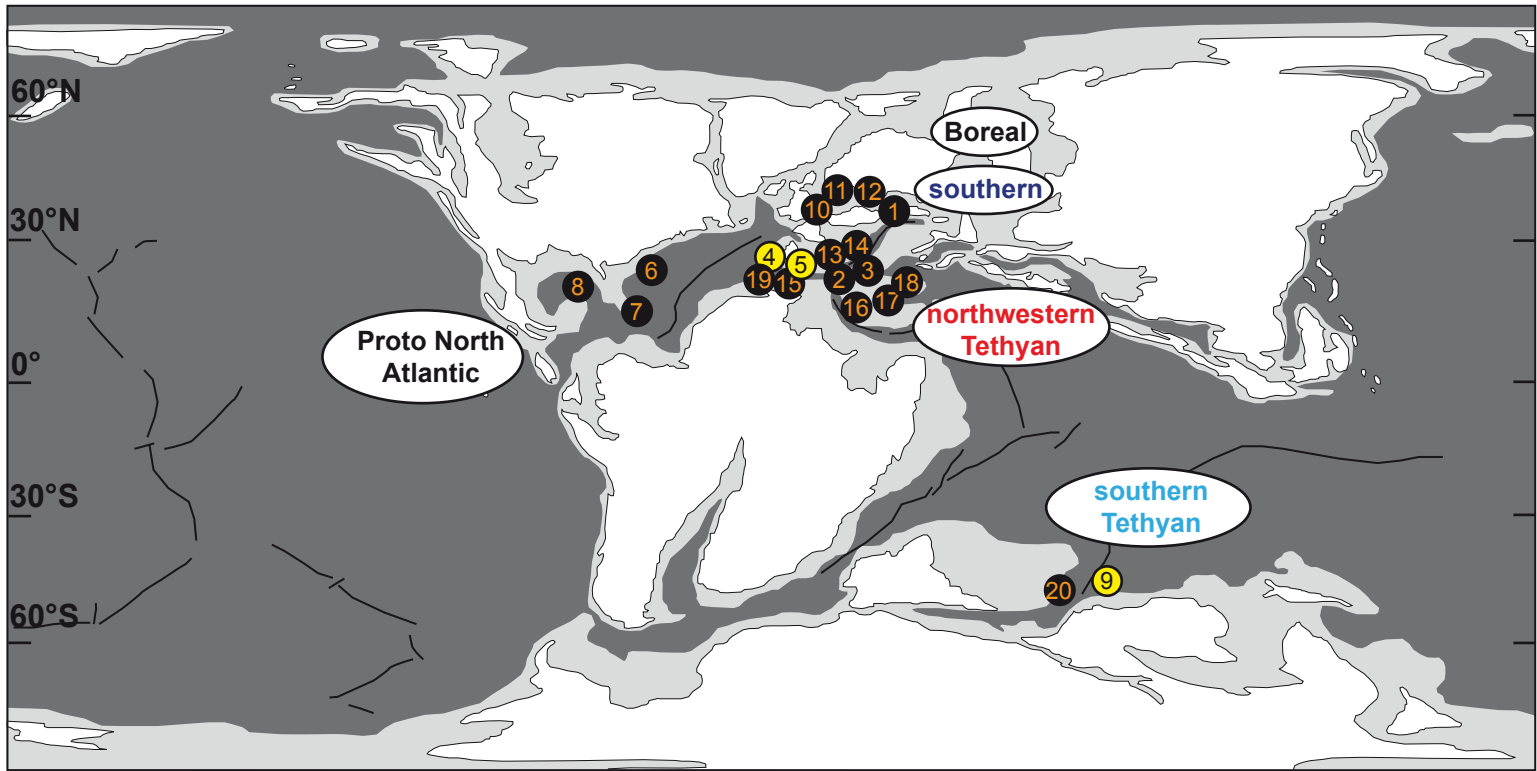
1163 **Table captions**

1164

1165 **Table 1:** Mineralogical, and geochemical data set compilation.

1166

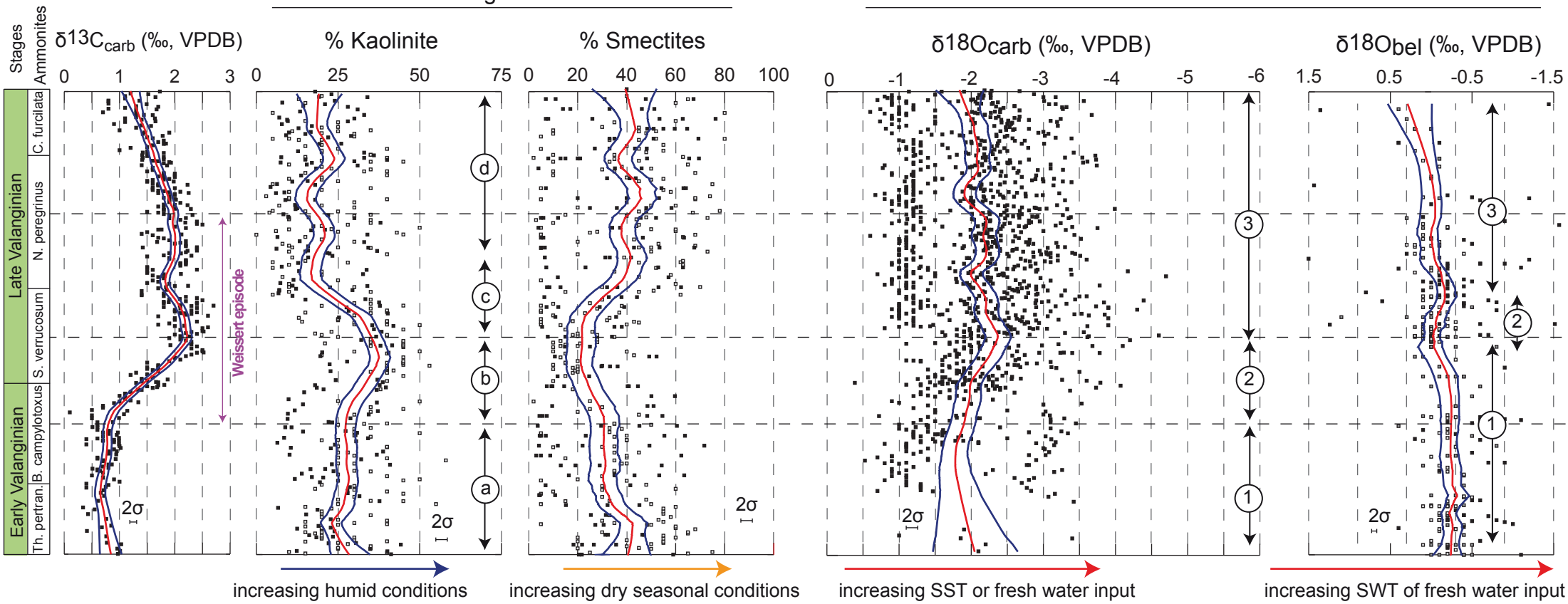
1167 **Table 2:** Seawater palaeotemperatures calculation applying the equation of Anderson and Arthur
1168 (1983) and using various $\delta^{18}\text{O}_w$ values according to the latitude (Zhou et al., 2008; Price et al.,
1169 2018).



Northwestern Tethyan realm (~20-30°N)

Mineralogical data

Geochemical data



Mineralogical and geochemical data references:

Clay mineralogy data:

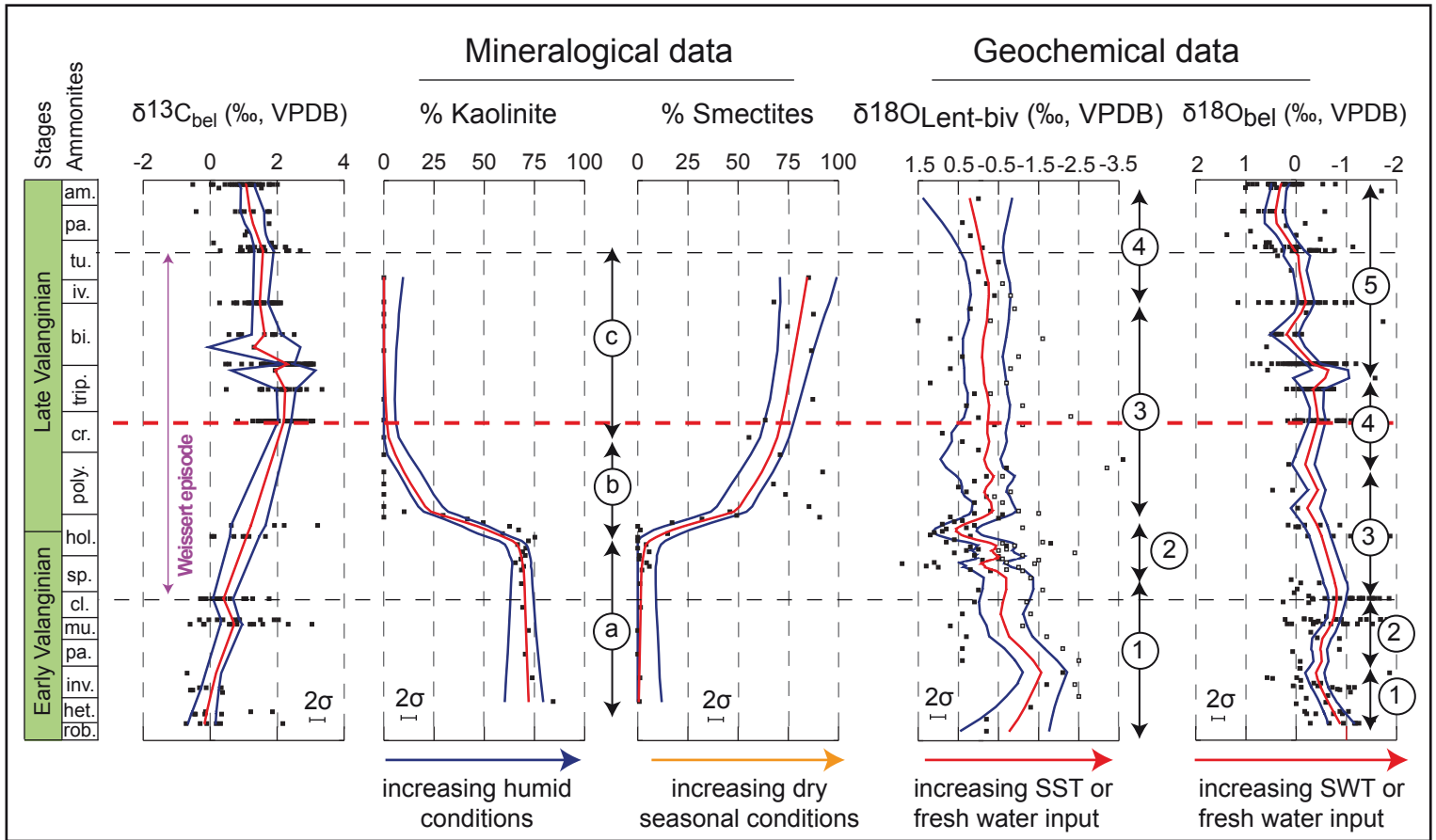
- Duchamp-Alphonse et al., 2011; Charbonnier et al., 2016
- This Study

$\delta^{13}\text{C}_{\text{carb}}$ data: Duchamp-Alphonse et al., 2007; Kujau et al., 2012

$\delta^{18}\text{O}_{\text{carb}}$ data: Sprovieri et al., 2006; Duchamp-Alphonse et al., 2007; Fozy et al., 2010; Gréselle et al., 2011; Kujau et al., 2012; Charbonnier et al., 2016; Bajnai et al., 2017; Aguado et al., 2018

$\delta^{18}\text{O}_{\text{bel}}$ data: van de Schootbrugge et al., 2000; McArthur et al., 2007; Price et al., 2018

southern part of the Boreal realm (~35-45°N)



Mineralogical and geochemical data references:

$\delta^{13}C_{carb}$ data: Meissner et al., 2015

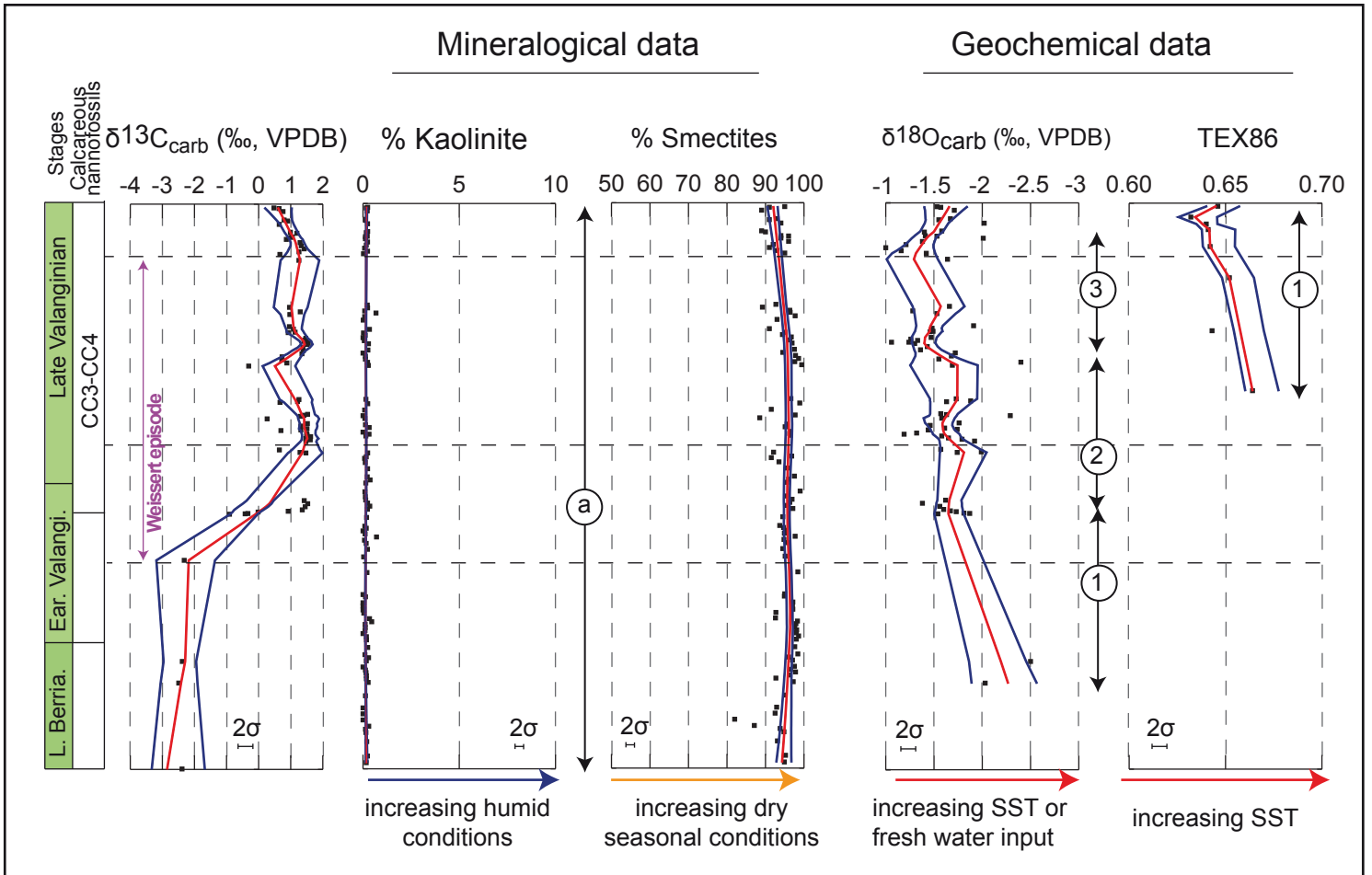
Clay mineralogy data: Morales et al., 2015

$\delta^{18}O_{biv}$ data ■ Morales et al., 2015

$\delta^{18}O_{Lent}$ data □

$\delta^{18}O_{bel}$ data: Podlaha et al., 1998; Price et al., 2000; McArthur et al., 2004; Meissner et al., 2015

southern Tethyan realm (~53°S)



Mineralogical and geochemical data references:

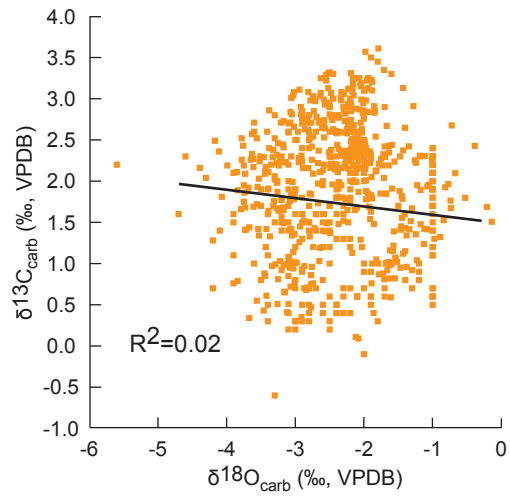
$\delta^{13}\text{C}_{\text{carb}}$ data: Charbonnier et al., 2020

Clay mineralogy data: This Study

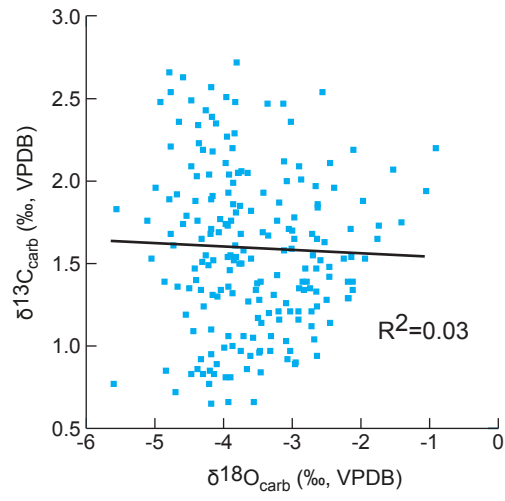
$\delta^{18}\text{O}_{\text{carb}}$ data: This Study

TEX₈₆ data: Littler et al., 2011

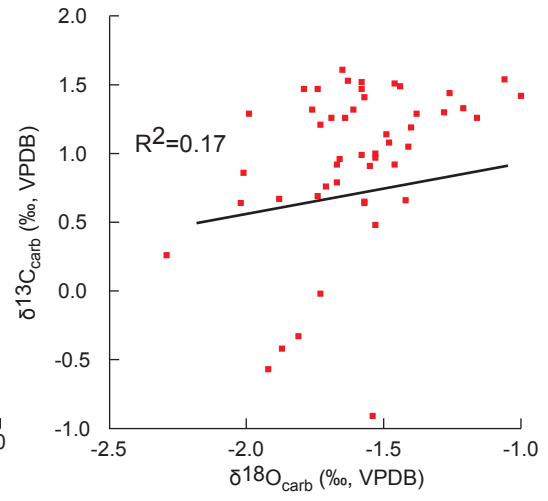
Northwestern Tethyan



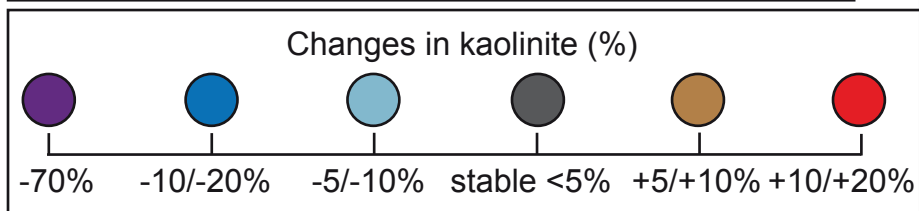
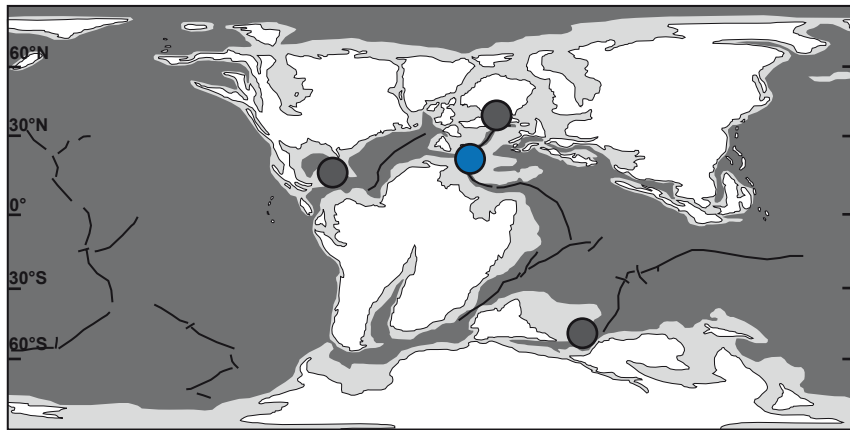
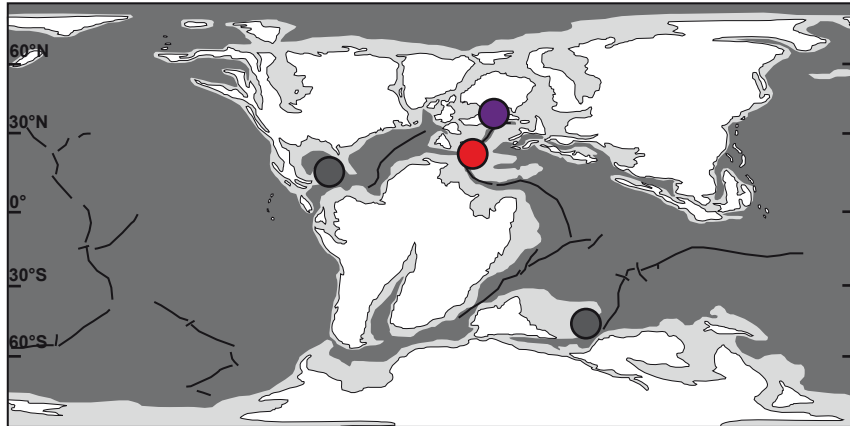
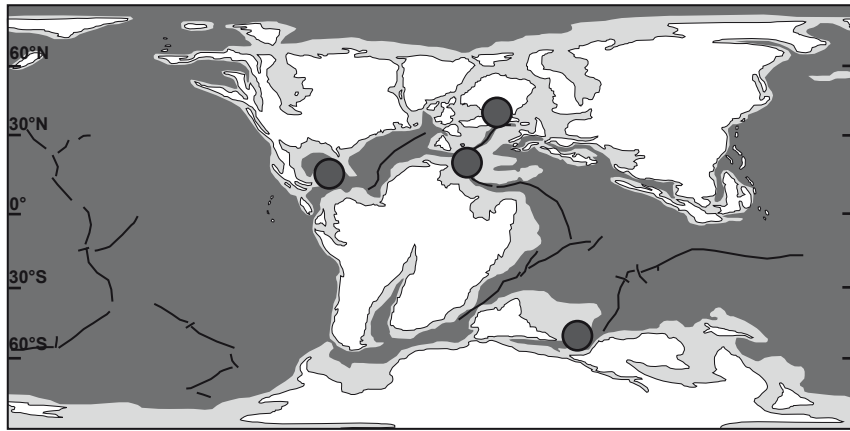
Proto North Atlantic



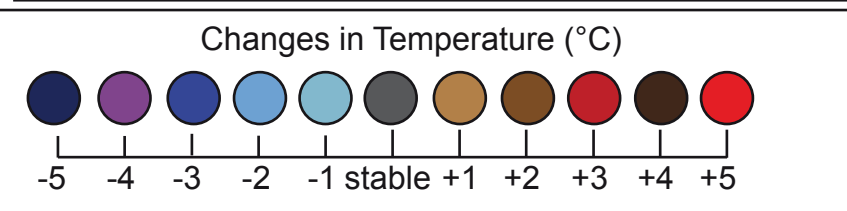
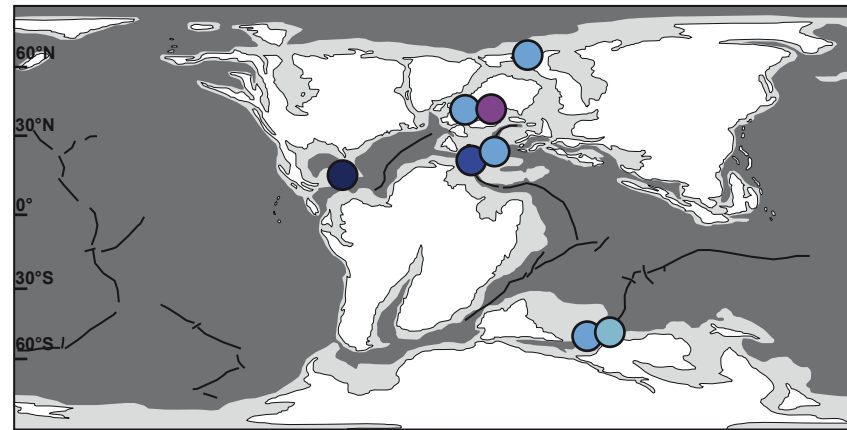
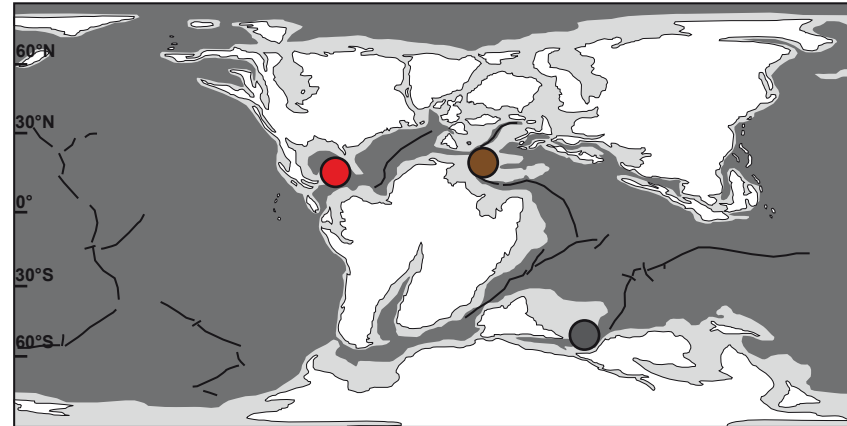
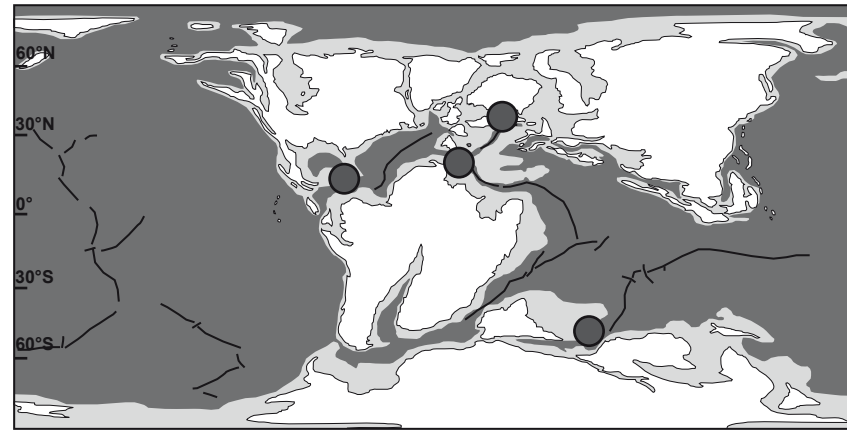
Southern Tethyan



Clay mineralogy assemblages



Temperatures



early Valanginian:

T. pertransiens
B. campylotoxus

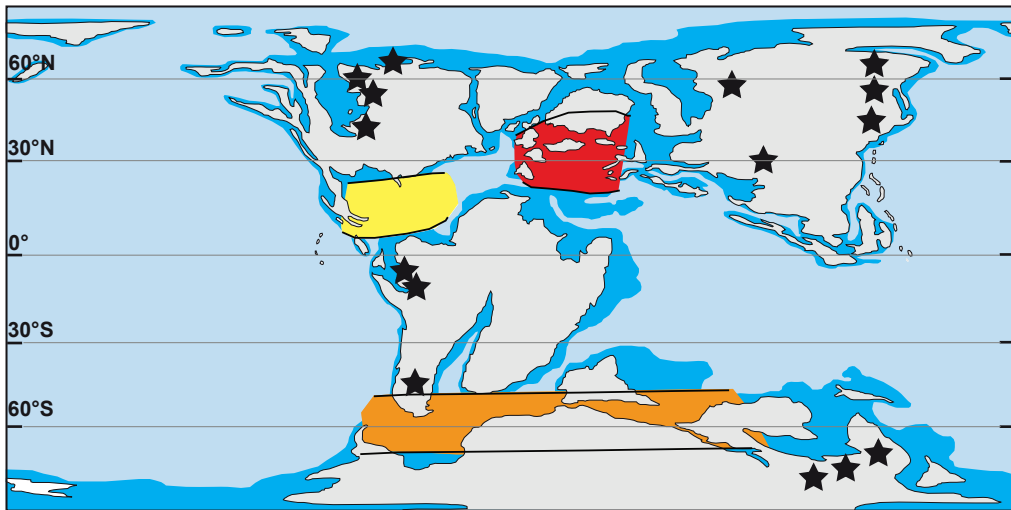
early-late Valanginian transition:

B. campylotoxus-*S. verrucosum* transition

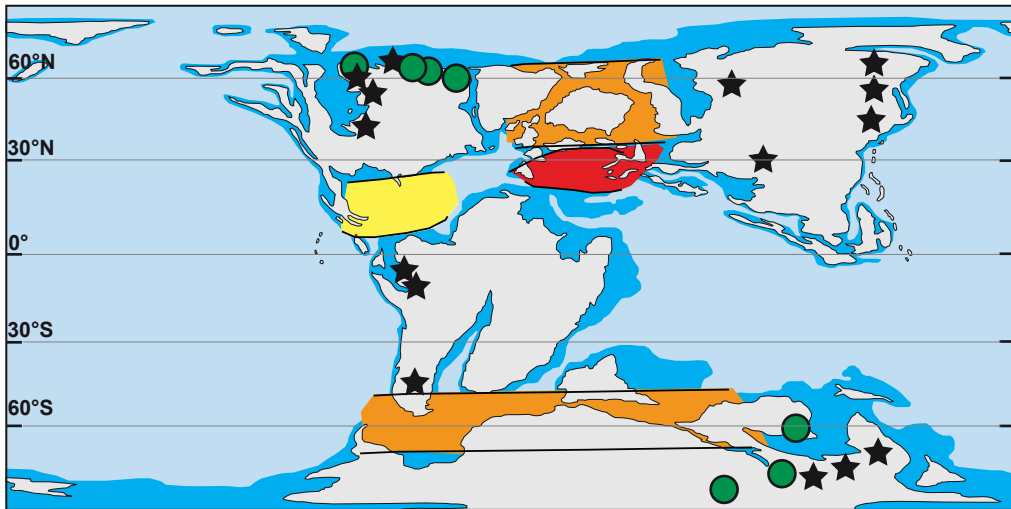
late Valanginian:

upper part *S. verrucosum*
N. peregrinus
C. furcilata

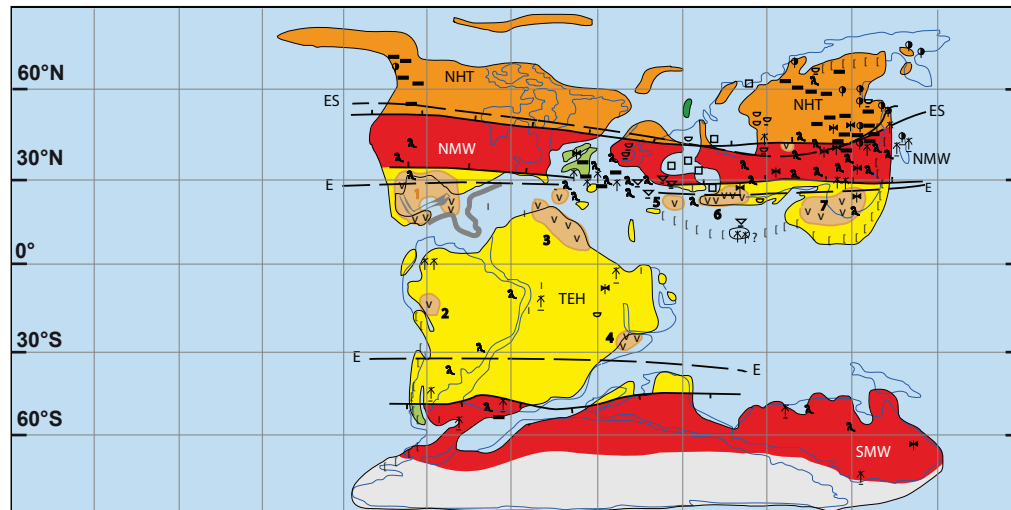
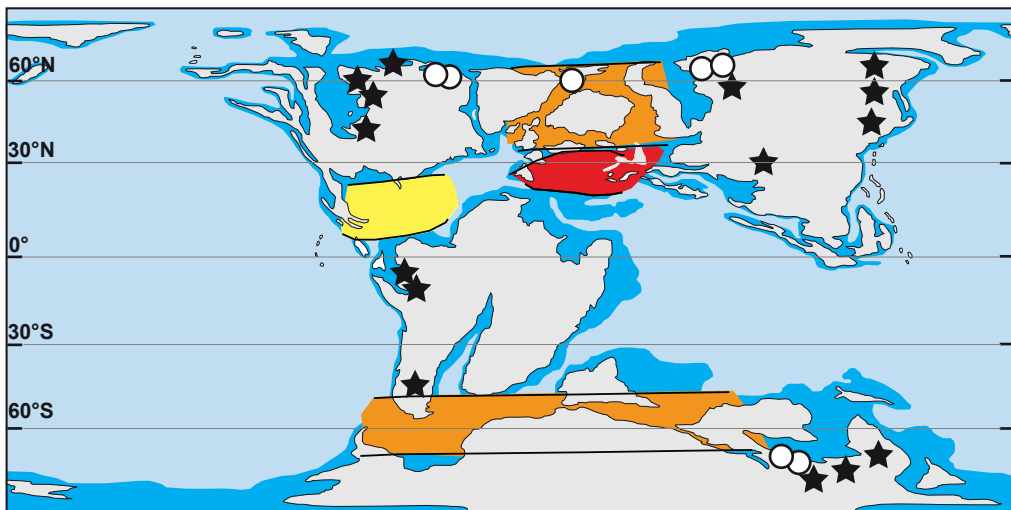
early Valanginian

















early-late Valanginian transition






late Valanginian





	high-latitude temperate belts		moderately thermophilic vegetation
	mid-latitude warm humid belts		moderately thermophilic insects
	evaporite belts		thermophilic insects
	arid belts		thermophilic vegetation
	laterite and clastic iron ores of terrestrial origin		localities of dinosaur remains
	gypsum and anhydrite deposits		coal and lignite deposits
			sedimentary and laterite bauxites
			kaolin weathering crusts or deposits

NHT: Northern High-latitude Temperate humid belt
 SHT: Southern High-latitude Temperate humid belt
 NMW: Northern Mid-latitude Warm humid belt
 SMW: Southern Mid-latitude Warm humid belt
 TEH: Tropical -Equatorial Hot arid belt
 NHA: Northern Hot Arid belt
 EH: Equatorial Humid belt
 SHA: Southern Hot Arid belt

Valanginian latitudinal belts:

	sub-arid belt		humid subtropical belt
	Warm temperate belt		

Vegetation cover:

	Distribution of conifer forests		Location of Berriasian-Valanginian-Hauterivian coal deposits
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Ice rafted indicator:


	Distribution of dropstones/glendonites/tillites
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Table 1: Mineralogical, and geochemical data set compilation

A) Mineralogical data

Section	Basin	N° section Fig. 1	Latitude	Depositional environment	Realm	lithology	Analyses	References
Wawal core	Polish Basin	1	35-40°N	proximal setting	Southern part of the Boreal	sandy and silty claystone	clay <2µm fraction	Morales et al., 2015
Angles	Vocontian Basin	2	25-30°N	hemipelagic setting	northwestern Tethyan	marl-limestone alternations	clay <2µm fraction	Duchamp-Alphonse et al., 2011
Orpierre	Vocontian Basin	3	25-30°N	hemipelagic setting	northwestern Tethyan	marl-limestone alternations	clay <2µm fraction	Charbonnier et al., 2016
Barranco del Garranchal	Subbetic Basin	4	20-30°N	hemipelagic setting	northwestern Tethyan	marl-limestone alternations	clay <2µm fraction	This study
Molina de las Oicas	Subbetic Basin	5	20-30°N	hemipelagic setting	northwestern Tethyan	marl-limestone alternations	clay <2µm fraction	This study
Site 603B	Continental rise Cape Hatteras	6	~17°N	pelagic setting	proto North Atlantic	laminated and bioturbated pelagic limestones	clay <2µm fraction	Huff, 1993
Site 534A	Blake Bahama Basin	7	~15°N	pelagic setting	proto North Atlantic	laminated and bioturbated pelagic carbonates	clay <2µm fraction	Chamley et al., 1983
Site 535	Gulf of Mexico	8	~15°N	pelagic setting	proto North Atlantic	limestone and marly limestone	clay <2µm fraction	Debrabant et al., 1984
Site 765C	Argo abyssal plain	9	~53°S	abyssal plain	southern Tethyan	pelagic claystone and nannofossil chalk	clay <2µm fraction	This study

B) Geochemical data

Section	Basin	N° section Fig. 1	Latitude	Depositional environment	Location	lithology	Analyses	References
Speeton	Yorkshire	10	~40-45°N	epicontinental sea	Southern part of the Boreal	clay formation, calcareous mudrocks	belemnites	Price et al., 2000; McArthur et al., 2004
Northwest Germany	Lower Saxony Basin	11	~38-42°N	epicontinental sea	Southern part of the Boreal	mudstones with slightly silty	belemnites	Meissner et al., 2015
Northern Germany	Proto-North Sea Basin	12	38°N	shallow marin	Southern part of the Boreal	terrigeneous brackish	belemnites	Podlaha et al., 1998
Wawal core	Polish Basin	1	35-40°N	proximal setting	Southern part of the Boreal	sandy and silty claystone	whole rock	Morales et al., 2015
Angles	Vocontian Basin	2	25-30°N	hemipelagic setting	northwestern Tethyan	marl-limestone alternations	whole rock	Duchamp-Alphonse et al., 2007
Orpierre	Vocontian Basin	3	25-30°N	hemipelagic setting	northwestern Tethyan	marl-limestone alternations	whole rock	Charbonnier et al., 2016
Vergol/La Charce	Vocontian Basin	13	25-30°N	hemipelagic setting	northwestern Tethyan	marl-limestone alternations	whole rock	Gréselle et al., 2011; Kujau et al., 2012
Composite section	Vocontian Basin	14	25-30°N	hemipelagic setting	northwestern Tethyan	marl-limestone alternations	belemnites	van de Schootbrugge et al., 2000; McArthur et al., 2007. Price et al., 2018
Composite section	Subbetic Basin	15	20-30°N	hemipelagic setting	northwestern Tethyan	marl-limestone alternations	belemnites	Price et al., 2018
Composite section	Umbria Marche Basin	16	~20°N	pelagic setting	northwestern Tethyan	white to gray pelagic limestone	whole rock	Sprovieri et al., 2006
Bersek quarry	Gerecse Mountains	17	~20°N	foreland basin	northwestern Tethyan	Grey-purple marlstone	whole rock	Bajnai et al., 2017
Kozoskut ravine	Bakony Mountains	18	~20°N	pelagic setting	northwestern Tethyan	Cherty marls and limestone	whole rock	Fozy et al., 2010
Cehegin	South Iberian Continental margin	19	~25°N	hemipelagic setting	northwestern Tethyan	marl-limestone alternations	whole rock	Aguado et al., 2018
Site 603B	Continental rise Cape Hatteras	6	~17°N	pelagic setting	proto North Atlantic	laminated and bioturbated pelagic limestones	TEX86, SSTs	Littler et al., 2011
Site 603B	Continental rise Cape Hatteras	6	~17°N	pelagic setting	proto North Atlantic	laminated and bioturbated pelagic limestones	whole rock	Bornemann and Mutterlose, 2008
Site 534A	Blake Bahama Basin	7	~15°N	pelagic setting	proto North Atlantic	laminated and bioturbated pelagic carbonates	TEX86, SSTs	Littler et al., 2011
Site 534A	Blake Bahama Basin	7	~15°N	pelagic setting	proto North Atlantic	laminated and bioturbated pelagic carbonates	whole rock	Bornemann and Mutterlose, 2008
Site 766A	Gascoyne and Cuvier abyssals plains	20	~53°S	abyssal plain	southern Tethyan	varicolored pelagic claystone and nannofossil chalk	TEX86, SSTs	Littler et al., 2011
Site 765C	Argo abyssal plain	9	~53°S	abyssal plain	southern Tethyan	pelagic claystone and nannofossil chalk	whole rock	This study

Table 2
northwestern Tethyan realm

$\delta^{18}\text{O}_w$	$\delta^{18}\text{O}_{\text{calcite}}$			$\delta^{18}\text{O}_w$	$\delta^{18}\text{O}_{\text{calcite}}$		
	early-late Valanginian transition				late Valanginian		
	-1.87‰	-2.4‰	$\Delta T^\circ\text{C}$		-2.4‰	-1.8‰	$\Delta T^\circ\text{C}$
1.0‰	28.7°C	31.3°C	2.6°C	1.0‰	31.3°C	28.4°C	2.9°C
1.4‰	30.7°C	33.4°C	2.7°C	1.2‰	32.3°C	29.4°C	2.9°C
	average		3°C		average		3°C

Proto North Atlantic realm

$\delta^{18}\text{O}_w$	$\delta^{18}\text{O}_{\text{calcite}}$			$\delta^{18}\text{O}_w$	$\delta^{18}\text{O}_{\text{calcite}}$		
	early-late Valanginian transition				late Valanginian		
	-3.65‰	-4.5‰			-4.5‰	-3.62‰	$\Delta T^\circ\text{C}$
0.5‰	35.2°C	39.7°C	4.5°C	0.5‰	39.7°C	35.0°C	4.7°C
	average		5°C		average		5°C

southern part of the Boreal realm

$\delta^{18}\text{O}_w$	$\delta^{18}\text{O}_{\text{Lent+biv}}$		
	late Valanginian		
	-0.28‰	0.2‰	$\Delta T^\circ\text{C}$
0‰	17.0°C	15.0°C	2.0°C
-0.5‰	14.9°C	13.0°C	1.9°C
	average		2°C

Southern Tethyan realm

$\delta^{18}\text{O}_w$	$\delta^{18}\text{O}_{\text{calcite}}$		
	late Valanginian		
	-1.6‰	-1.3‰	$\Delta T^\circ\text{C}$
-1.5‰	16.6°C	15.0°C	1.6°C
	average		2°C