

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

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1	A global palaeoclimatic reconstruction for the Valanginian based on clay mineralogical and
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High-resolution clay mineral and $\delta^{18}O_{carb}$ analyses have been performed on three sections of 29 30 Valanginian age (Early Cretaceous), from northwestern (~20–30°N) and southern (~53°S) Tethyan realms. The data have been integrated in a large set of published mineralogical (clays), and 31 geochemical ($\delta^{18}O_{carb}$, $\delta^{18}O_{bivalve}$, $\delta^{18}O_{lenticulina}$, $\delta^{18}O_{belemnite}$, and TEX₈₆) data from 17 sections, 32 situated between ~45°N and ~53°S. Based on this data set, we provide new insights into 33 Valanginian climate conditions from a wide range of palaeolatitudes. We highlight climate trends 34 within specific areas and identify main climate belts. In the Proto-North Atlantic realm (~15–17°N) 35 large amounts of smectite together with the occurrence of palygorskite testify for a semi-arid 36 climate belt throughout the Valanginian. Significant variations in kaolinite content in the 37 northwestern Tethyan realm (~20 to 30°N) suggest that this realm has been more sensitive to 38 changes in hydrological cycling and subsequent erosion and runoff processes during the early to 39 early-late Valanginian transition interval. This time interval, that is coincident with the pronounced 40 positive and globally recognized carbon isotope excursion (CIE; the Weissert episode), documents 41 the wet conditions of a humid subtropical climate belt. Temperature ranges derived from δ^{18} O 42 records suggest climate warming by about 3-5±0.9°C in the northern hemisphere, during the 43 Weissert episode. This is followed by a global cooling of about ca. 1–5±0.9°C in both hemispheres, 44 during the late Valanginian. 45

We postulate that these patterns were due to interplays between tectonic and orbital factors, which have controlled the distribution of regional palaeoclimate belts during the Valanginian. The semi-arid belt expressed in the Proto North Atlantic realm has probably been induced by its specific palaeogeographic configuration and the existence of isolated basins, which were each characterized by strong and continuous thermohaline circulation. An eccentricity paced monsoon-like system might have played a significant role in maintaining the subtropical belt around the northwestern Tethyan realm, up to the mid latitudes (~35–45°N). Superimposed on these regional processes, a widespread magmatic pulse in Paraná-Etendeka volcanic activity might have stimulated greenhouse conditions, impacting marine biota and favouring the development of a dense vegetation cover on the continent. An increase in sedimentary burial of 13 C depleted organic carbon on the continents may have contributed to the progressive global positive carbon isotope excursion. In the medium term, it has also probably led to a significant decrease in atmospheric CO₂, as testified by the global cooling recorded during the late Valanginian in both hemispheres and the probable concomitant extension of polar ice caps.

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

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64 **1. Introduction**

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

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

While such results may provide a rather clear image of the evolution of climate during the Valanginian, new and contrasting findings have challenged this particular view, rendering the reconstructions of palaeoclimate conditions that prevailed during the Valanginian more elusive. The most striking controversial examples are the TEX₈₆ records from the Proto North Atlantic (~15– 17°N) and the southern Tethyan (~53°S) realms, which document high and rather stable sea-surface temperatures (SST of ~33 to 34°C and ~26°C, respectively) through the entire Valanginian (Littler et al., 2011). As a consequence, these authors have re-evaluated the earliest Cretaceous as a warm and stable period. In addition, recent petrological and geochemical analyses on the complex sequence of ikaite-calcite precipitation leading to glendonite formation suggest that glendonites document past events of methane release and oxidation in polar settings rather than cold-water conditions (e.g., Morales et al., 2017). Similarly, the origin of dropstones in the geological record can be related to non-glacial mechanisms such as gravity processes or wood rafting, implying that these deposits cannot consistently be interpreted as the result of ice rafting and therefore cool climate (Bennett et al., 1994, 1996; Doublet and Garcia, 2004).

112 This controversy in the interpretation of climate evolution during the Valanginian is promoted by the lack of high-resolution climate reconstructions in the southern hemisphere based 113 114 on geochemistry and clay mineralogy (Ditchfield, 1997; Price and Nunn, 2010; Littler et al., 2011; Price and Passey, 2013). Here, we provide new insights into Valanginian climate conditions from a 115 wide range of palaeolatitudes, in order to highlight main trends within specific realms, and 116 emphasize the presence and localization of main climate belts. This study is based upon new clay 117 mineral and $\delta^{18}O_{carb}$ data from the northwestern (~20–30°N) and southern (~53°S) Tethys realms. 118 These data have been integrated into a set of published clay, δ^{13} C, δ^{18} O, and TEX₈₆ data from sites 119 situated between ~45°N and ~53°S (Table 1, Fig. 1). Our multiproxy approach helps specifying 120 weathering intensity on continents and documenting humid and arid climate belts. It is also useful to 121 re-evaluate previous geochemical results and propose coherent palaeotemperature patterns on a 122 global scale. 123

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125 **2. Data compilation and method**

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

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

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

- 157 palaeoenvironmental trends.
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- 159 **3. Results**
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- 161 3.1. Northern hemisphere data
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163 3.1.1. Northwestern Tethyan realm (~20°–30°N)

164 Kaolinite, smectite, $\delta^{18}O_{carb}$ and $\delta^{18}O_{bel}$ 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 %, respectively. The early Valanginian (Tirnovella pertransiens-Busnardoites campylotoxus ammonite 167 Zones) is characterized by relatively stable values with averages of 27 and 36 %, despite a small 168 decreasing/increasing trend in the kaolinite/smectite percentages at the base of the section. Near the 169 early-late Valanginian transition, i.e. where the Weissert event starts (B. campylotoxus-Saynoceras 170 171 *verrucosum* transition Zones), the kaolinite record shows an increasing trend, reaching a maximum of 39% at the δ^{13} C climax, whereas the smectite curve displays a drop of 13 % (from 34 to 21 %). A 172 significant decrease in kaolinite abundances to 16 % is observed throughout the early late 173 Valanginian (S. verrucosum-Neocomites peregrinus zones). Smectite reaches contents of up to 41 174 % in this interval. Finally, the upper part of the section, which covers the late Valanginian interval 175 (N. peregrinus-Criosarasinella furcillata zones), shows relatively stable values in kaolinite and 176 smectite contents, with averages of 22 and 40 %, respectively. 177

The smoothed $\delta^{18}O_{carb}$ record is quite stable in the early Valanginian (Fig. 2), ranging from -2.0 to -1.8%. The oxygen isotope data show a decreasing trend in the early-late Valanginian transition, and reaches their lowest values (-2.4 %.) at the $\delta^{13}C$ peak. The late Valanginian is characterized by rather scattered $\delta^{18}O_{carb}$ values. However, a long-term positive trend is discernible, going from -2.4 to -1.8 %. (amplitude of 0.6 %.). The smoothed $\delta^{18}O_{bel}$ data show relatively stable values (between -0.3 and -0.2 ‰) for the early and early-late Valanginian interval, while a subsequent increase towards more positive ones (from -0.2 to 0.3 ‰) is observed in the remainder of the late Valanginian.

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

188 Kaolinite, smectite, $\delta^{18}O_{carb}$ and TEX₈₆ LOESS smoothing curves are shown in Fig. 3.

Throughout the Valanginian, clay mineral assemblages are largely dominated by smectite (between 70 and 87 %) with minor contents of kaolinite (between 1 and 8 %) and palygorskite (between 1 and 5 %). The smoothed $\delta^{18}O_{carb}$ record shows rather stable values (between -3.7 and -3.5 % $_{o}$) in the early Valanginian. It is characterized by a prominent negative trend, ranging from -3.6 to -4.5 % $_{o}$ at the early-late Valanginian transition, followed by a long-term positive trend (from -4.5 to -3.6 % $_{o}$), in the late Valanginian. The smoothed TEX₈₆ data displays quite stable values (between 0.91 and 0.93) along the Valanginian interval.

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

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

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

The $\delta^{18}O_{\text{lent}}$ and $\delta^{18}O_{\text{biv}}$ signals show short-term fluctuations (from -0.70 to -1.58 ‰) in the lower Valanginian. A subsequent increase to 0.53 ‰ is documented for the early–late Valanginian transition, while in the early part of the late Valanginian, the $\delta^{18}O_{\text{lent}}$ and $\delta^{18}O_{\text{biv}}$ values display rather stable values (from -0.39 to -0.16 ‰). Further up section, in the latest Valanginian interval, they rise from -0.28 to 0.20 ‰. The smoothed $\delta^{18}O_{\text{bel}}$ 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.
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212	3.2. Southern Tethyan realm (~53°S)
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214	The evolution of kaolinite, smectite, $\delta^{18}O_{carb}$, 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 $\delta^{18}O_{carb}$ 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

to -1.3 %) along the late Valanginian, which is concomitant to a slight decreasing trend (from 0.66

- to 0.63) in the smoothed TEX₈₆ record.
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- 223 **4. Discussion**
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- 4.1. Significance of clay mineral and oxygen isotope signals
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- 4.1.1. Diagenetic overprint

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

In the studied sections, illitization processes have not been identified (Chamley et al., 1983; 235 Duchamp-Alphonse et al., 20111; Morales et al., 2015; Charbonnier et al., 2016) and smectite is 236 largely abundant instead. Besides, Rock-Eval analyses performed on selected samples indicate that 237 238 T_{max} values never exceed 435 °C (Burtner and Warner, 1986; Morales et al., 2015; Charbonnier et al., 2020), thus indicating a low thermal influence, below the oil window. These T_{max} values are 239 consistent with the occurrence of irregular illite-smectite (I-S) R0 minerals with a high proportion 240 of smectite layers (>50 %), which indicates a weak thermal diagenesis (Dellisanti et al., 2010). 241 Therefore, it is reasonable to state that the impact of diagenesis on clay mineral signals was rather 242 limited, and that trends in this proxy reflect palaeoenvironmental conditions. 243

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

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

2000). Model simulations of Ricken (1986, 1987) suggest that the calcium carbonate cement in the 261 limestone beds was sourced from calcium carbonate dissolution in the interlayers during the early 262 diagenesis prior to the compaction. They demonstrate that the possible effect of differential 263 264 compaction and early diagenetic changes acting on the precursors of these rhythmic marl-limestone alternations, may have seriously disturbed the original primary signal of $\delta^{18}O_{carb}$ signal (Westphal et 265 al., 2008). Cyclostratigraphic investigations reveal that marl-limestone alternations in the 266 267 Valanginian sediments mainly originate from cyclic changes of humidity/aridity conditions linked 268 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 269 decreases of primary $\delta^{18}O_{carb}$ and $\delta^{13}C_{carb}$ values through the precipitation of variable amounts of 270 diagenetic cements depleted in ¹⁸O and ¹³C, respectively (e.g., Veizer, 1983; Jenkyns and Clayton, 271 1986: Jenkvns, 1995: Godet et al., 2016; Benamara et al., 2020). Cross plots of $\delta^{18}O_{carb}$ - $\delta^{13}C_{carb}$ of 272 the northwestern Tethyan, Proto North Atlantic, and southern Tethyan realms, reveal lack of 273 correlations (R^2 =0.02, 0.03, and 0.17, respectively) (Fig. 6). Besides, no negative or positive 274 covariance exists between the δ^{18} O signal and the CaCO₃ contents in the ODP 765C (southern 275 Tethyan realm) (R^2 =0.03, Fig. S1 in supplementary online materials, SOM) as it would be expected 276 from strong burial overprint (Frank et al., 1999). The $\delta^{18}O_{carb}$ values studied herein, are similar to 277 those of open marine carbonates (from -4.0 to -1.0 %) and fall generally within the range of δ^{18} O 278 values for sediments from the Early Cretaceous period (Van de Schootbrugge et al., 2000; Godet et 279 al., 2006; Stein et al., 2011). Burial diagenesis overprint has not significantly affected the primary 280 $\delta^{18}O_{carb}$ signal as suggested by the well-preserved clay mineral assemblage in the signal. Although 281 this reinforces the assumption that the $\delta^{18}O_{carb}$ trends reflect an original signal, we cannot totally 282 exclude a potential diagenetic overprint on $\delta^{18}O_{carb}$ absolute values, and thus on reconstructed 283 palaeotemperatures. Consequently, we only consider the trends of the long-term LOESS-smoothing 284 $\delta^{18}O_{carb}$ curves. In the same way as for published $\delta^{18}O_{bel}$ signals, and because the whole-rock 285 carbonate fraction is mainly composed of planktic calcareous fossils, we interpret such trends as 286

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

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

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304 4.1.2. Kaolinite and smectite as palaeoclimate proxies

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

The distribution of kaolinite versus smectite is partly controlled by their differential settling behaviour, which is related to physical segregation during their transport as a function of differences in size and flocculation capacity (Thiry, 2000; Adatte et al., 2002; Godet et al., 2008). Indeed, kaolinite is a dense and coarse mineral that settles preferentially closer to the coast, in shallow-water environments. Conversely, smectite is a fine-grained mineral that tends to settle much more slowly, in deeper waters and more offshore settings. Consequently, differential settling of kaolinite versus smectite may explain the distribution of clay mineral assemblages along a platform/basin transect (Thiry, 2000). The sections selected here present lithologies reflecting hemipelagic, pelagic, or abyssal settings (Table 1).

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

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

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

Consequently, in analogy to the clay distribution in modern oceans (Ito and Wagai, 2017), and because local sedimentary and diagenetic overprints have been shown to bear less weight, we assume that the stratigraphic distribution of clay minerals captures general patterns in weathering intensity at a basin scale. However, caution is in order due to the differential settling overprint and 365 sea-level fluctuations.

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

372

4.1.3. Oxygen isotopes as a potential palaeothermometer

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

384

385
$$T(^{\circ}C) = 16.0 - 4.14 \left(\delta^{18}O_{c} - \delta^{18}O_{w} \right) + 0.13 \left(\delta^{18}O_{c} - \delta^{18}O_{w} \right)^{2}$$

386

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

391	used a $\delta^{18}O_w$ 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 $\delta^{18}O_{seawater}$ values in the <i>Neocomiensiformis</i> and <i>Furcillata-Loryi</i> Zones and
396	more negative values in the Verrucosum Zone. Consequently, we estimate the amplitude of SST
397	change with $\delta^{18}O_w$ 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

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401 4.2.1. Early Valanginian

- 10

During the early Valanginian, both kaolinite and smectite contents as well as $\delta^{18}O_{carb}$ 402 document no significant temporal changes in the studied realms (i.e. northwestern Tethyan, the 403 Proto North Atlantic, and the southern part of the Boreal) (Fig. 7). It appears that weathering 404 intensity and seawater temperatures have remained stable during this time interval. These results 405 confirm previous TEX₈₆ measurements (Littler et al., 2011), palynological investigations (Kujau et 406 al., 2013), and recently reported Mg/Ca ratios (Price et al., 2018), which highlight stable 407 408 temperature conditions within the Proto-North Atlantic and the northwestern Tethyan realms, respectively. 409

Interestingly, kaolinite and smectite distributions show significant variations according to the studied basin (Figs. 2, 3, and 4). In a similar way as for modern oceans, these regional modifications are tentatively explained by the presence and influence of latitudinal climate belts with contrasted hydrolysing conditions on continents (Fig. 8). The high abundance of kaolinite in sediments from the northwestern Tethyan (up to 27 %) and southern part of the Boreal (up to 74 %) realms suggests the existence of an extensive humid subtropical climate belt (at least) ~20 to 45°N (Fig. 8). By contrast, in the Proto North Atlantic realm, kaolinite is uncommon and never exceeds 8 % (Zemmels et al., 1972; Chamley, 1981; Chamley et al., 1983; Debrabant et al., 1984; Huff,
1993). Large smectite proportions (>70 %) associated with the occurrence of palygorskite might
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., 2005; Duchamp-Alphonse et al., 2011; Kujau et al., 2013; Charbonnier et al., 2016). Further north 425 426 (at ~45°N), this time interval is marked by the replacement of kaolinite by smectite (from 1 to 74 %), highlighting an abrupt change from high to low weathering intensities and thus, from humid to 427 dry and seasonally contrasted conditions (Fig. 7). This is well supported by change in spore and 428 pollen assemblages showing an evolution toward more arid climate (Kujau et al., 2013). These 429 spatio-temporal changes in the weathering mode may point to the southward migration of a 430 monsoon-like subtropical rain belt during the Weissert episode, as previously proposed by 431 Duchamp-Alphonse et al. (2014), Morales et al. (2015), and Charbonnier et al. (2016), based on 432 local climate reconstructions. Our reconstruction permits to postulate a southward shift of 433 minimally 15° in latitude (from 45 to 30°N) (cf., Morales et al., 2015). At the opposite, as 434 documented by the dominance of smectite (>80 %) and palygorskite (>8 %) in the Proto-North 435 Atlantic realm, climate conditions have remained stable and the semi-arid climate belt has prevailed 436 in this part (Figs. 7 and 8). 437

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

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

Applying the equation of Anderson and Arthur (1983) and using various $\delta^{18}O_w$ values 461 according to the latitude (Zhou et al., 2008; Price et al., 2018 and see chapter 4.1.3), results in 462 temperature increases of about 3±0.9°C in the northwestern Tethyan and about 5±0.9°C in the Proto 463 North Atlantic (Table 2). As such, warming appears much more moderate in the northwestern 464 Tethyan compared to the Proto-North Atlantic. This confirms the lower temperature variability 465 recently obtained by belemnite Mg/Ca ratios in the Subbetic Domain and the Vocontian basin 466 during the Weissert Episode (Price et al., 2018). However, these temperatures and changes therein 467 are in contradiction with TEX₈₆ records from the Proto North Atlantic realm, which report stable 468

temperature conditions during the early-late Valanginian transition period (Littler et al., 2011). The 469 causes for this difference can be related to a diagenetic alteration of the $\delta^{18}O_{carb}$ values recorded by 470 the whole rock signal (see chapter 4.1.1). Alternatively this discrepancy may be related to a 471 472 seasonal bias (Sluijs et al., 2006). The TEX₈₆ proxy is based on the chrenarchaeotal membrane lipid distribution in sedimentary rocks, which is dependent on SST (Schouten et al., 2003). However, the 473 highest fluxes of chrenarchaeotal lipids coincide with periods of high phytoplanktonic productivity 474 and are recorded during summer months (Sluijs et al., 2006). Therefore, the cell number in the 475 476 water column depends on the season, and the TEX₈₆ signal may record an average of the high summer SSTs (Sluijs et al., 2006), prone to changes with relatively small amplitudes. In all cases, 477 478 changes in SST conditions observed in the oxygen-isotope records attest to a warming phase in the northern hemisphere. Together with the weathering patterns documented by clay mineral data, they 479 highlight temperate to semi-arid conditions in the Proto North Atlantic, and humid and subtropical 480 conditions related to a monsoon-like system in the northwestern Tethyan (Fig. 8). 481

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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.

The composite whole rock δ^{18} O values in the northwestern Tethyan and in the Proto North Atlantic realms show long-term trends towards heavier values (amplitudes of 0.58±0.18 ‰ and 0.88±0.18 ‰, respectively) (Figs. 2 and 3). These trends probably indicate declines in palaeotemperatures of ~3±0.9 and 5±0.9°C, respectively (see chapter 4.1.3 and Fig. 7). The

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

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

517

518 4.3. Palaeoclimatic reconstruction in the southern hemisphere

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520

At ODP Site 765C (~53°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 $\delta^{18}O_{carb}$ 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 $\delta^{18}O_{bel}$ 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 $\delta^{18}O_{carb}$ signal of ODP Site 765C shows trend towards heavier values (amplitudes of
531	0.3±0.18 %o) (Fig. 5), which may indicate a decline in palaeotemperatures of ~2±0.9°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 (~1°C; Littler et al., 2011) (Fig. 5) and the occurrence of <i>cheirolepidiaceae</i> 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.

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542 4.4. Valanginian climate conditions: a complex interplay between regional versus global543 mechanisms

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545 Our compilation of published and unpublished clay mineral and $\delta^{18}O_{carb}$ data allows for a 546 better appraisal of global climate conditions during the Valanginian. The low- to mid-latitudes of northern hemisphere are characterized by the presence of two main rainfall belts: a humid subtropical one associated with a monsoon-like system bringing significant precipitations into the Tethyan realm, as testified by the important proportions of kaolinite in the area from sites between (at least) ~20 to 45°N; and a sub-arid one, characterized by dry conditions in the Proto North Atlantic realm, as documented by prominent proportions of smectite and palygorskite at sites ~15– 17°N. Temperate conditions may have prevailed around 45°N and up to ~60°N.

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

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

582 Superimposed on these regional mechanisms, many studies suggested a link with the intensification of the Paraná-Etendeka (South America-South Africa) volcanic activity (Lini et al., 583 1992; Föllmi et al., 1994; Erba et al., 2004). An update of the Valanginian/Hauterivian 584 astrochronological time scale confirms a potential link between the main phase of Paraná-Etendeka 585 volcanic activity (-134.6±0.6; -134.3±0.8 Ma; Thiede and Vasconcelos, 2010; Janasi et al., 2011) 586 587 and the beginning of the Weissert episode (-135.22 ± 1 Ma) (Martinez et al., 2015). Moreover, a set of mercury anomalies in Valanginian sediments recovered from localities outside and in European 588 sections demonstrated that a main phase of magmatism occurred at the onset of the Weissert 589 episode (Charbonnier et al., 2017, 2020). Our study highlights a coherent fingerprint of the Paraná-590 Etendeka emplacement on climate change across the Weissert Episode. Indeed, the global warming 591 observed during the Weissert episode is the typical consequence of increased emissions of 592 greenhouse gases from a LIP volcanism. In contradiction to Price et al. (2018), it appears that 593 elevated pCO_2 levels during the Weissert episode and associated warming may have actually 594 accelerated the hydrological cycle and enhanced nutrient input from continents to ocean. Ocean 595 fertilization associated with excess of CO₂ in surface waters (i.e., surface-ocean acidification) may 596 have induced marine carbonate production crisis in both in neritic and pelagic domains (Föllmi et 597 al., 1994; Weissert et al., 1998; Erba et al., 2004; Weissert and Erba, 2004; Barbarin et al., 2012; 598

Duchamp-Alphonse et al., 2014). On the continent the development of greenhouse conditions 599 probably led to the implantation of an important vegetation cover from mid to high latitudes, as 600 601 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 conifer forests of Valanginian age were documented in the Australian (~53-85°S) and Canadian 603 domains (~74–79°N) (Dettmann et al., 1992; McLoughlin and Hill, 1996; McLoughlin et al., 2002; 604 Galloway et al., 2013, 2015; Tosolini et al., 2014) (Fig. 8). The presence of extensive polar forests 605 606 probably induced positive feedbacks between climate and vegetation, reducing surface albedo and increasing adsorption of solar radiation at high latitudes (Upchurch et al., 1998). During the 607 608 Valanginian these factors might have also played an important role in maintaining high-latitude warmth, which might have induced the presence of a stable warm temperate belt in high latitudes 609 (Fig. 8). This is supported by clumped-isotope analyses from belemnites, which testify for warm 610 polar conditions (10–20°C) at 60–65°N (Price and Passey, 2013). The long-lasting and global burial 611 of terrestrial organic carbon consuming a significant amount of atmospheric CO₂, may have 612 contributed to the global cooling observed in both hemispheres, during the last stages of the 613 Weissert episode (Figs. 7 and 8). In fossil plant material, from the late Valanginian, Gröcke et al. 614 (2005) attested cool terrestrial conditions caused by a decrease in atmospheric pCO_2 levels. 615 Stratigraphic and sedimentological evidences suggest that this global cold event may have help 616 developing the Livingston glaciation observed in high southern latitudes during the late 617 Valanginian-Hauterivian interval (Alley et al., 2019). One cannot exclude that this time interval 618 619 might also be punctuated by limited extension of polar ice cap in the high northern latitudes, as previously documented by the occurrence of glendonites, tillites, and dropstones described in 620 Svalbard (~65–70°N; Kemper and Schmitz, 1975; Price and Nunn, 2010; Vickers et al., 2019), in 621 the Sverdrup Basin (~74–79°N; Kemper, 1987; Price, 1999), and in Siberia (Price, 1999). Such cold 622 conditions could have caused the formation of a new deep-water ocean current system through the 623 opening of the Greenland-Norwegian seaway (Pauly et al., 2012). 624

625

626 **5. Conclusions**

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

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

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

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 PA3223.
- 1103
- 1104 **Figure captions**
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Fig. 1: World palaeogeographic map of the Early Cretaceous (modified after Blakey, 2010). 1106 Locations of the studied sections in four depositional environments at several palaeolatitudes (see 1107 1108 Table 1): (1) Wawal core (Polish Basin), (2) Angles section (Vocontian Basin), (3) Orpierre section (Vocontian Basin), (4) Barranco del Garranchal section (Subbetic Basin), (5) Molina de las Oicas 1109 section (Subbetic Basin), (6) DSDP Hole 603B (Continental rise Cape Hatteras), (7) DSDP Hole 1110 534A (Blake Bahama Basin), (8) DSDP Hole 535 (Gulf of Mexico), (9) ODP Hole 765C (Argo 1111 abyssal plain), (10) Speeton section (Yorkshire), (11) Northwest Germany composite section 1112 (Lower Saxony Basin), (12) Northern Germany composite section (Proto-North Sea Basin), (13) 1113 1114 Vergol/La Charce sections (Vocontian Basin), (14) Vocontian composite section (Vocontian Basin), (15) Subbetic composite section (Subbetic Basin), (16) Umbria Marche composite section (Umbria 1115 1116 Marche Basin), (17) Bersek Quarry (Gerecse Mountains), (18) Kozoskut ravine section (Bakony Mountains), (19) Cehegín section (southern Iberian continental margin) (20) ODP Hole 766A 1117 (Gascoyne and Cuvier abyssals plains). 1118

1119

1120 Fig. 2: Synthesis diagram showing kaolinite, smectite, $\delta^{18}O_{carb}$ and $\delta^{18}O_{bel}$ 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

Fig. 3: Synthesis diagram showing kaolinite, smectite, $\delta^{18}O_{carb}$ and TEX₈₆ LOESS smoothing curves from the Proto North Atlantic realm with the carbon isotope stratigraphy against the biostratigraphic scheme. LOESS, i.e. locally weighted scatterplot smoothing (Cleveland and Devlin, 1988). For the calculation of LOESS smoothing curve (red line) an α value of 0.15 has been applied. Blue curves indicate the 95 % confidence interval. SST: Sea Surface Temperature.

1132

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

1139

Fig. 5: Synthesis diagram showing kaolinite, smectite, $\delta^{18}O_{carb}$ and TEX₈₆ LOESS smoothing curves from the southern Tethyan realm with the carbon isotope stratigraphy against the biostratigraphic scheme. LOESS, i.e. locally weighted scatterplot smoothing (Cleveland and Devlin, 1988). For the calculation of LOESS smoothing curve (red line) an α value of 0.15 has been applied. Blue curves indicate the 95 % confidence interval. SST: Sea Surface Temperature.

1145

Fig. 6: Cross-plot of the $\delta^{13}C_{carb}$ and the $\delta^{18}O_{carb}$ values from the northwestern Tethyan, Proto North Atlantic and southern Tethyan realms. 1148

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

1152

Fig. 8: Synthetic map (modified after Blakey, 2010) illustrating the Valanginian palaeoclimatic 1153 belts supported by geochemical and mineralogical data during the early Valanginian, the early-late 1154 Valanginian transition, and the late Valanginian. Location of Berriasian, Valanginian and 1155 Hauterivian coal deposits are reported after McCabe and Totman-Parrish (1992). Distribution of 1156 conifer forest are from Dettmann et al. (1992); McLoughlin and Hill (1996); McLoughlin et al. 1157 (2002); Galloway et al. (2013, 2015); and Tosolini et al. (2014). Distribution of glendonites, 1158 dropstones and tillites are from Kemper and Schmitz (1975), Kemper (1987), Price (1999), Alley 1159 and Frakes (2003), Price and Nunn (2010), and Alley et al. (2019). Reconstruction of the 1160 1161 palaeoclimatic belts that prevailed during the Berriasian (modified after Chumakov et al., 1995).

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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 $\delta^{18}O_w$ values according to the latitude (Zhou et al., 2008; Price et al., 1169 2018).





Northwestern Tethyan realm (~20-30°N)



Proto North Atlantic realm (~15-17°N)

Clay mineralogy data: Chamley et al., 1983; Debrabant et al., 1984; Huff, 1993

 $\delta^{13}\text{C}_{carb}$ data: Bornemann and Mutterlose, 2008

 $\delta 18O_{carb}$ data: Bornemann and Mutterlose, 2008

TEX₈₆ data: Littler et al., 2011



southern Tethyan realm (~53°S)



δ18O_{carb} data: This Study TEX₈₆ data: Littler et al., 2011





-5/-10% stable <5% +5/+10% +10/+20%

-70%

-10/-20%



-1 stable +1 +2 +3 +4 +5

-2

-3

-4

-5

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



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
Cehegín	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

Feuille1

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Table 2 northwestern Tethyan realm

1.0‰ 1 4‰	28.7°C 30.7°C	31.3°C 33.4°C	2.6°C 2 7°C	1.0‰ 1.2‰	31.3°C 32.3°C	28.4°C 29.4°C	2.9°C 2 9°C
	-1.87‰	-2.4‰	ΔT°C		-2.4‰	-1.8‰	ΔT°C
	early-late	Valanginian t	ransition		la	ate Valanginia	n
$\delta^{18}O_w$		$\delta^{18}O_{\text{calcite}}$		$\delta^{18}O_w$		$\delta^{18}O_{\text{calcite}}$	

Proto North Atlantic realm

$\delta^{18}O_w$		$\delta^{18}O_{\text{calcite}}$		$\delta^{18}O_w$		$\delta^{18}O_{\text{calcite}}$	
	early-late Valanginian transition				late Valanginian		
	-3.65‰	-4.5‰			-4.5‰	-3.62‰	ΔT°C
0.5‰	35.2°C	39.7°C	4.5°C	0.5‰	39.7°C	35.0°C	4.7°C
	aver	age	5°C		average		5°C

southern part of the Boreal realm

s ¹⁸ 0 s ¹⁸ 0							
$\delta O_w \delta O_{Lent+l}$							
	late Valanginian						
	-0.28‰	0.2‰	ΔT°C				
0‰	17.0°C	15.0°C	2.0°C				
-0.5‰	14.9°C	13.0°C	1.9°C				
	aver	2°C					

Southern Tethyan realm

	ave	2°C			
-1.5‰	16.6°C	15.0°C	1.6°C		
	-1.6‰	-1.3‰	ΔT°C		
	late Valanginian				
$\delta^{18}O_w$	$\delta^{18}O_{calcite}$				