#### Environmental changes during the onset of the Late Pliensbachian 1

- Event (Early Jurassic) in the Cardigan Bay Basin, Wales. 2
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14 Abstract. The Late Pliensbachian Event (LPE), in the Early Jurassic, is associated with a perturbation in the 15 global carbon cycle (positive carbon isotope excursion (CIE) of  $\sim 2$  %), cooling of  $\sim 5^{\circ}$ C, and the deposition of 16 widespread regressive facies. Cooling during the Late Pliensbachian has been linked to enhanced organic matter 17 burial and/or disruption of thermohaline ocean circulation due to sea level lowstand of at least regional extent . 18 Orbital forcing had a strong influence on the Pliensbachian environments and recent studies show that the 19 terrestrial realm and the marine realm in and around the Cardigan Bay Basin, UK, were strongly influenced by 20 orbital climate forcing. In the present study we build on the previously published data for long eccentricity cycle 21  $E459 \pm 1$  and extend the palaeoenvironmental record to include  $E458 \pm 1$ . We explore the environmental and 22 depositional changes on orbital time scales for the Llanbedr (Mochras Farm) core during the onset of the LPE. 23 Clay mineralogy, XRF elemental analysis, isotope ratio mass spectrometry, and palynology are combined to 24 resolve systematic changes in erosion, weathering, fire, grain size and riverine influx. Our results indicate 25 distinctively different environments before and after the onset of the LPE positive CIE, and show increased 26 physical erosion relative to chemical weathering. We also identify five swings in the climate, in tandem with the 27 405 kyr eccentricity minima and maxima. Eccentricity maxima are linked to precessionally repeated 28 occurrences of a semi-arid monsoonal climate with high fire activity and relatively coarser sediment from 29 terrestrial runoff. In contrast, 405 kyr minima in the Mochras core are linked to a more persistent, annually wet 30 climate, low fire activity, and relatively finer grained deposits across multiple precession cycles. The onset of 31 the LPE +ve CIE did not impact the expression of the 405 kyr cycle in the proxy records; however, during the 32 second pulse of heavier carbon (<sup>13</sup>C) enrichment, the clay minerals record a change from dominant chemical 33 weathering to dominant physical erosion.

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#### 35 **1** Introduction

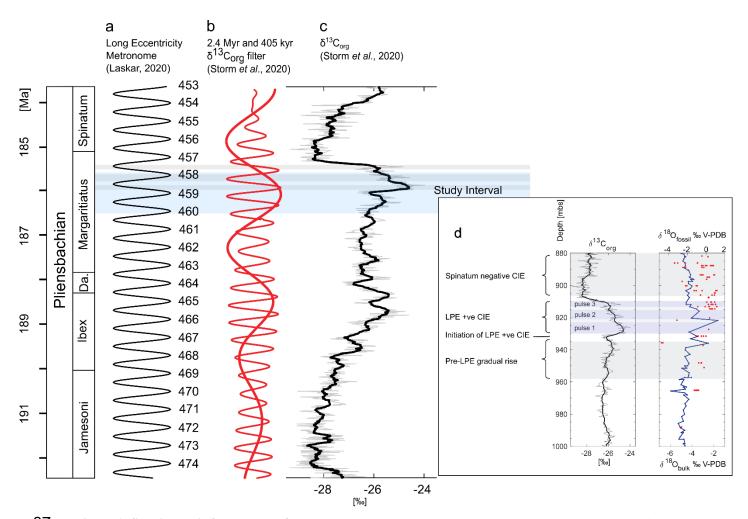
36 The Early Jurassic is a period marked by large climatic fluctuations and associated carbon-isotope excursions

- 37 (CIE's) in an overall warmer than present and high  $pCO_2$  world (McElwain *et al.*, 2005; Korte and Hesselbo,
- 38 2011; Steinthorsdottir and Vajda, 2015; Korte et al., 2015; Robinson et al., 2016). A series of small and medium
- 39 sized CIE's have recently been documented for the Sinemurian and Pliensbachian, which have mainly been

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- 40 from European, North African and North American records (Korte and Hesselbo, 2011; Franceschi *et al.*, 2014;
- 41 Korte *et al.*, 2015; Price *et al.*, 2016; De Lena *et al.*, 2019; Hesselbo *et al.*, 2020a; Mercuzot *et al.*, 2020; Storm
- 42 et al., 2020; Silva et al., 2021; Cifer et al., 2022; Bodin et al., 2023). Notable is the pronounced positive CIE in
- 43 the Late Pliensbachian, which has been called the Late Pliensbachian Event (LPE) and is linked to climatic
- 44 cooling (Hesselbo and Korte, 2011; Korte *et al.*, 2015) and a supra-regional/global sea level low stand (Hallam,
- 45 1981; de Graciansky *et al.*, 1998; Hesselbo and Jenkyns, 1998; Hesselbo, 2008). The LPE has been recognized
- 46 by a positive shift in benthic marine oxygen-isotopes (~1.5–2 per mil) (Bailey *et al.*, 2003; Rosales *et al.*,
- 47 2004,2006; Suan et al., 2010; Dera et al., 2011a; Korte and Hesselbo, 2011; Gómez et al., 2016; Alberti et al.,
- 48 2019, 2021), coeval with a positive shift in marine and terrestrial carbon isotopes (~2 per mil) (Jenkyns and
- 49 Clayton, 1986; McArthur et al., 2000; Morettini et al., 2002; Quesada et al., 2005; Rosales et al., 2006; Suan et
- 50 *al.*, 2010; Korte and Hesselbo, 2011; Silva *et al.*, 2011; Gómez *et al.*, 2016; De Lena *et al.*, 2019).
- 51 A cooler Late Pliensbachian climate has been suggested based on low  $pCO_2$  values inferred by leaf stomatal
- 52 index data from eastern Australia (Steinthorsdottir and Vajda, 2015), the presence of glendonites in northern
- 53 Siberia (Kaplan, 1978; Price, 1999; Rogov and Zakharov, 2010), vegetation shifts from a diverse flora of
- 54 different plant groups to one mainly dominated by bryophytes in Siberia (Ilyina, 1985; Zakharov *et al.*, 2006),
- and possible ice rafted debris found in Siberia (Price, 1999; Suan *et al.*, 2011). Whilst the presence of ice sheets
- 56 is strongly debated, a general cooling period (~5°C lower; Korte *et al.*, 2015; Gómez *et al.*, 2016) is evident
- 57 from several temperature reconstructions from NW Europe. A cooling is hypothesized via enhanced carbon
- 58 burial in the marine sediments, leading to lower *p*CO<sub>2</sub> values and initiating cooler climatic conditions (Jenkyns
- and Clayton, 1986; Suan et al., 2010; Silva et al., 2011; Storm et al., 2020). Direct evidence of large-scale
- 60 carbon burial in Upper Pliensbachian marine deposits has not yet been documented (Silva *et al.*, 2021).
- 61 Alternatively, cooling has been suggested to be caused by a lower sea level which would have disrupted ocean
- 62 circulation in the Laurasian Seaway, reducing poleward heat transport from the tropics (Korte *et al.*, 2015). In
- the UK region, a dome structure in the North Sea has been linked to shedding of sediments during sea level low
- 64 stands from the Late Toarcian and possibly before (Underhill and Partington, 1993; Korte *et al.*, 2015; Archer et
- al. 2019). Disruption of the ocean circulation between the western Tethys and the Boreal realm is supported by
- 66 marine migration patterns (Schweigert, 2005; Zakharov et al., 2006; Bourillot et al., 2008; Nikitenko, 2008;
- 67 Dera et al., 2011b; van de Schootbrugge et al., 2019) and numerical models (Bjerrum et al., 2001; Dera and
- **68** Donnadieu, 2012; Ruvalcaba Baroni *et al.* 2018); however, the net direction of the flows remain debated.
- An additional factor to be considered is that a strong orbital control exists on the Pliensbachian sedimentary
- 70 successions (Weedon and Jenkyns, 1990; Ruhl et al., 2016; Hinnov et al., 2018; Storm et al., 2020; Hollaar et
- 71 *al.*, 2021). Previous studies have indicated that sea level changes, possibly coupled to glacio-eustatic rise and
- fall, occurred during the LPE on a 100 kyr (short eccentricity) time scale (Korte and Hesselbo, 2011). A high-
- resolution record of charcoal, clay mineralogy, bulk-organic carbon-isotopes, TOC and CaCO<sub>3</sub> encompassing
- 74 approximately one 405 kyr cycle from the Llanbedr (Mochras Farm) borehole, Cardigan Bay Basin, NW Wales,
- 75 UK, suggested that the long-eccentricity orbital cycle had a significant effect on background climatic and
- 76 environmental change during the late Pliensbachian, particularly affecting the hydrological regime of the region
- 77 (Hollaar *et al.*, 2021). This previous research focussed on orbital forcing of environmental change for a time
- **78** lacking any large excursion in  $\delta^{13}C_{org}$ , and so unaffected by perturbations to the global carbon cycle. Here, we

- respond on the record of Hollaar *et al.* (2021) to cover two long eccentricity cycles (which we identify as
- 80 spanning from cycle E459 ±1 to the start of E457 ±1 of Laskar *et al.* 2011 and Laskar 2020), where the final
- parts of E458 and the start of E457 are interrupted by onset of the Late Pliensbachian Event (Fig. 1). This longer
- 82 record allows us to more robustly examine the influence of the long eccentricity cycle and the potential impact
- 83 of a global carbon cycle perturbation on the palaeoclimate and depositional environment. We find that the long
- 84 eccentricity forcing continued to dictate the precise timing of major environmental changes in the Cardigan Bay
- 85 Basin, including the initial step of the positive carbon isotope excursion.



**Figure 1: Stratigraphic framework of the Mochras borehole.** (a) 405 kyr metronome (Laskar, 2020) which shows that this study spans E459 ±1 to E457 ±1. (b) 2.4 Myr and 405 kyr filter derived from the  $\delta^{13}C_{org}$  record from Storm *et al.* (2020). A slight offset in pacing is observed in the 405 kyr metronome based on an assumed fixed 405 kyr cycle length (a), versus filtering of the 405 kyr signal from the orbital solution (b). (c)  $\delta^{13}C_{org}$ curve from the Mochras borehole (Storm *et al.*, 2020), showing the ~1.8 ‰ +CIE that marks the LPE. High resolution data are visualized in light grey and a 10-step moving average in black. The blue bar marks interval in the Mochras borehole considered in this study. The three grey shaded bars represent the three pulses in the +CIE

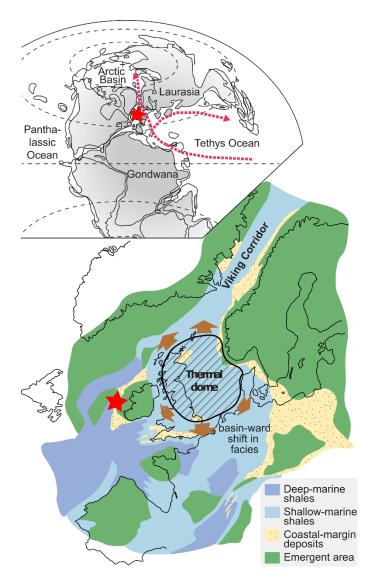
- 94 of the LPE. (d) Close-up of the  $\delta^{13}C_{org}$  (Storm *et al.*, 2020) and  $\delta^{18}O_{bulk}$  and  $\delta^{18}O_{fossil}$  (Ullmann *et al.*, 2022) from
- 95 the Late Pliensbachian of the Mochras core. A pre-LPE gradual rise is recorded in the  $\delta^{13}C_{org}$ , followed by the
- 96 initiation of the LPE +ve CIE, which consists of three pulses. After the LPE +ve CIE,  $\delta^{13}C_{org}$  values drop

- 97 recorded starting at ~910 mbs, and the Spinatum negative CIE is recorded. The  $\delta^{18}O_{bulk}$  of the Mochras core
- 98 (blue) is diagenetically altered and unlikely to preserve a palaeoclimatic imprint (Ullmann *et al.*, 2022). Also,
- $99 \qquad \text{shown are } \delta^{18}O_{\text{fossil}} \text{ values (red)}.$
- 100

# 101 2 Material

# 102 2.1 Palaeo-location and setting

- 103 Associated with the break-up of Pangea, connections between oceans via epicontinental seaways were
- 104 established during the Early Jurassic, such as the Hispanic Corridor, which connected the north-western Tethys
- and the eastern Panthalassa, and the Viking Corridor which linked the north-western Tethys Ocean to the Boreal
- 106 Sea (Sellwood and Jenkyns, 1975; Smith *et al.*, 1983; Bjerrum *et al.*, 2001; Damborenea *et al.*, 2012). The
- 107 linking passage of the NW Tethys Ocean and the Boreal Sea (south of the Viking Corridor) is the
- 108 palaeogeographical location of the Llanbedr (Mochras Farm) borehole, Cardigan Bay Basin, NW Wales, UK
- 109 (Fig. 2) referred to hereafter as Mochras. Due to the location of the Mochras succession during the Late
- 110 Pliensbachian, it was subject to both polar and equatorial influences allowing study of variations in the
- 111 circulation in the N-S Laurasian Seaway (including the Viking Corridor) prior to and across the LPE. Mochras
- 112 was located at a mid-palaeolatitude of ~35° N (cf. Torsvik and Cocks, 2017).



## 114 Figure 2: Palaeolocation of the Mochras borehole in the context of potential North Sea doming. Figure

- reprinted and adapted from Korte *et al.* (2015), which is open access
- 116 (https://creativecommons.org/licenses/by/4.0/). The Mochras borehole was located at a paleolatitude of  $\sim$ 35° N
- 117 in the Cardigan Bay Basin (Torsvik and Cocks, 2017). Circulation in the Tethys Ocean and between there and
- 118 the Boreal region influenced the depositional environment of the Mochras core (Pieńkowski *et al.*, 2021). Late
- 119 Pliensbachian sea level fall potentially resulted in occlusion of the Viking Corridor as the topography of the
- 120 North Sea dome structure disrupted circulation in the seaway (Korte *et al.*, 2015).
- 121
- 122 The depositional environment of Mochras is likely characterized by a rift setting, which is reflected by the
- 123 relatively open and deep marine facies and the evidence for below storm wave-base and contourite deposition
- 124 (Pieńkowski *et al.*, 2021), but always with a strong terrestrial influence (van de Schootbrugge *et al.*, 2005;
- 125 Riding et al., 2013) from the nearby landmasses (Dobson and Whittington, 1987). The Cardigan Bay Basin fill
- 126 was downthrown against the Early Paleozoic Welsh Massif by a major normal fault system, probably
- 127 comprising the Bala, Mochras and Tonfanau faults at the eastern and south-eastern margins of the basin in Late
- 128 Paleozoic–Early Mesozoic time (Woodland, 1971; Tappin *et al.*, 1994). The main source of detrital material is

- 129 understood to be the Caledonian Welsh Massif, followed by the Irish and Scottish landmasses (Deconinck *et al.*,
- 130 2019). Other massifs that could have influenced the provenance are the London-Brabant Massif to the south
- 131 east, and Cornubia to the south (van de Schootbrugge *et al.*, 2005), depending on the marine circulation and
- 132 sediment transport at the time.

## **133 2.2** Core location and material

- 134 The Llanbedr (Mochras Farm) Borehole was drilled onshore in the Cardigan Bay Basin (52 48' 32" N, 4 08' 44"
- 135 W) in 1967–1969, North Wales (Woodland, 1971; Hesselbo *et al.*, 2013). The borehole recovered a 1300 m
- thick Early Jurassic sequence (601.83–1906.78 metres below surface (mbs)), yielding the most complete and
- 137 extended Early Jurassic succession in the UK, being double the thickness of same age strata in other UK cores
- and outcrops (Hesselbo *et al.*, 2013; Ruhl *et al.*, 2016). The Lower Jurassic is biostratigraphically complete at
  the zonal level (Ivimey-Cook, 1971; Copestake and Johnson, 2014), with the top truncated and unconformably
- 140 overlain by Cenozoic strata (Woodland, 1971; Dobson and Whittington, 1987; Tappin *et al.*, 1994; Hesselbo *et*
- 141 *al.*, 2013). The lithology is dominated by argillaceous sediments, with alternating muddy limestone, marl and
- and the first of t
- 142 mudstone (Woodland, 1971; Sellwood and Jenkyns, 1975).
- 143 The Pliensbachian Stage in the Mochras borehole occurs between ~1250 and ~865 mbs, with the Margaritatus
- **144** Zone between ~1013 and 909 mbs (Page in Copestake and Johnson, 2014). The Pliensbachian interval
- 145 comprises alternations of mudstone (with a moderate total organic carbon [TOC]) and organic-poor limestones,
- 146 with a pronounced cyclicity at  $\sim 1 \pm 0.5$  m wavelength (Ruhl *et al.*, 2016). The Upper Pliensbachian contains
- 147 intervals that are silty and locally sandy, whilst levels of relative organic enrichment also occur through the
- 148 Pliensbachian (Ruhl et al., 2016). Overall, the Upper Pliensbachian is relatively rich in carbonate (Ruhl et al.,
- 149 2016; Ullmann *et al.*, 2022).
- 150

# 151 3 Methods

- 152 For this study, samples were taken at a ~30 cm resolution from slabbed core from 934–918 mbs for XRD and
- 153 mass spectrometry, as well as palynofacies and microcharcoal analysis. XRF analyses were made at a 1 cm
- resolution from 934–918 mbs (complete dataset deposited as Damaschke *et al.*, 2021). These new samples
- 155 complement samples and data at 10 cm resolution from 951–934 mbs published in Hollaar *et al.* (2021).

# 156 3.1 TOC, CaCO<sub>3</sub> and bulk organic carbon isotope mass spectrometry

- 157 TOC and  $\delta^{13}C_{org}$  were measured to track the changes in the total organic fraction and the bulk organic carbon
- 158 isotope ratios in relation to the other palaeoenvironmental proxy data.
- 159 Powdered bulk-rock samples (~ 2 g) were decarbonated in 50 ml of 3.3% HCl. After this, the samples were
- transferred to a hot bath of 79 °C for 1 h to remove siderite and dolomite. Subsequently, the samples were
- 161 centrifuged and the liquid decanted. The samples were rinsed repeatedly with distilled water to reach neutral *p*H.
- 162 After this, the samples were oven-dried at 40 °C, re-powdered and weighed into tin capsules for mass
- 163 spectrometry using the Sercon Integra 2 stable isotope analyser at the University of Exeter Environment and and
- 164 Sustainability Institute (ESI), stable isotope facility on the Penryn Campus, Cornwall. Samples were run
- alongside in-house reference material (bovine liver;  $\delta^{13}$ C -28.61 and Alanine;  $\delta^{13}$ C -19.62) which was used to
- 166 correct for instrument drift and to determine the  $\delta^{13}$ C values of the samples. The  $\delta^{13}$ Corg values are reported

- relative to V-PDB following a within-run laboratory standard calibration. Total organic carbon was determined
- using the CO<sub>2</sub> beam area relative to the bovine liver standard (%C 47.24). Replicate analysis of the in-house
- 169 standards gave a precision of  $\pm <0.1 \%$  (2 SD).
- 170 The carbonate content was measured by the dry weight sample loss before and after decarbonation. The %C
- 171 content derived from the mass spectrometer was corrected for carbonate loss to derive TOC.

## 172 3.2 X-Ray Diffraction (XRD) to determine clay mineralogy

- 173 Clay mineral analysis was performed to gain insight into the relative importance of physical erosion versus
- 174 chemical weathering and related changes in the hydrological cycle.
- 175 About 2–3 g of gently powdered bulk-rock was decarbonated with a 0.2 M HCl solution. The clay sized fraction
- 176 (< 2 µm) was extracted with a syringe after decantation of the suspension after 95 minutes following Stokes'
- 177 law. The extracted fraction was centrifuged and oriented on glass slides for X-ray diffraction analysis (XRD)
- 178 using a Bruker D4 Endeavour diffractometer (Bruker, Billerica, MA, USA) with Cu Kα radiations, LynxEye
- 179 detector and Ni filter under 40 kV voltage and 25 mA intensity (Biogéosciences Laboratory, Université
- 180 Bourgogne/Franche-Comté, Dijon). Following Moore and Reynolds (1997), the clay phases were discriminated
- 181 in three runs per sample: (1) air-drying at room temperature; (2) ethylene-glycol solvation for 24 h under
- **182** vacuum; (3) heating at 490 °C for 2 h.
- 183 Identification of the clay minerals was based on their main diffraction peaks and by comparison of the three
- 184 diffractograms obtained. The proportion of each clay mineral on glycolated diffractograms was measured using
- the MACDIFF 4.2.5. software (Petschick, 2000). Identification of the clay minerals follows the methods in
- 186 Deconinck *et al.* (2019) and Moore and Reynolds (1997).

## 187 3.3 Palynofacies and microcharcoal

- 188 Palynofacies were examined to explore shifts in the terrestrial versus marine origins of the particulate organic
- 189 matter. Each ~ 20 g bulk rock sample was split into 0.5 cm<sup>3</sup> fragments, minimizing breakage of charcoal and
- 190 other particles, to optimize the surface area for extraction of organic matter using a palynological acid
- 191 maceration technique. The samples were first treated with cold hydrochloric acid (10% and 37% HCl) to remove
- 192 carbonates. Following this, hydrofluoric acid (40% HF) was added to the samples to remove silicates. Carbonate
- 193 precipitation was prevented, by adding cold concentrated HCl (37%) after 48 h. The samples were neutralized
- 194 via multiple dilution-settling-decanting cycles using DI water, after which 5 droplets of the mixed residue were
- 195 taken for the analysis of palynofacies prior to sieving. The remaining residue was sieved through a 125 μm and
- 196 10 µm mesh to extract the microcharcoal fraction.
- 197 A known quantity (111 μl) out of a known volume of liquid containing the 10–125 μm sieved residue was
- 198 mounted onto a palynological slide using glycerine jelly. This fraction, containing the microscopic charcoal, was
- analysed and the charcoal particles counted using an Olympus (BX53) transmitted light microscope (40 x10
- 200 magnification). For each palynological slide four transects (two transects in the middle and one on the left and
- right side of the coverslip) were followed and the number of charcoal particles determined. Charcoal particles
- were identified with the following criteria: opaque and black, often elongated lath-like shape with sharp edges,
- 203 original anatomy preserved, brittle appearance with a lustrous shine (Scott, 2010). These data were then scaled
- up to the known quantity of the sample according the method of Belcher *et al.* (2005).

- 205 Palynofacies were grouped broadly according to Oboh-Ikuenobe *et al.* (2005): sporomorphs, fungal remains,
- 206 freshwater algae, marine palynomorphs, structured phytoclasts, unstructured phytoclasts, black debris,
- amorphous organic matter (AOM), and charcoal (further described in Hollaar et al., 2021). The palynofacies
- were quantified on a palynological slide using the optical light microscope (40 x10 magnification) and counting
- a minimum of 300 particles per slide. Because the samples are AOM-dominated, counting was continued until a
- 210 minimum of 100 non-AOM particles were observed. We used the percentage of terrestrial phytoclasts, which
- 211 includes sporomorphs, and structured and unstructured phytoclasts, to examine changes in terrestrial organic
- 212 particle content.
- 213

## 214 3.4 X-Ray Fluorescence (XRF) to determine detrital elements

- 215 Detrital elemental ratios were examined to analyse changes in relative terrestrial influx and the type of material
- transported from the land to the marine realm. The slabbed archive halves of the Mochras borehole were
- scanned via automated X-ray fluorescence (XRF) at a 1 cm resolution for the interval 951 –918 mbs, with the
- 218 ITRAX MC at the British Geological Survey Core Scanning Facility (CSF), Keyworth, UK (Damaschke et al.,
- 2021). The measurement window was 10 s and long-term drift in the measurement values was counteracted by
- regular internal calibration with a glass reference (NIST-610). Duplicate measurements were taken every 5 m
- for a 50 cm interval to additionally verify the measured results.

## 222 3.5 Statistical analysis

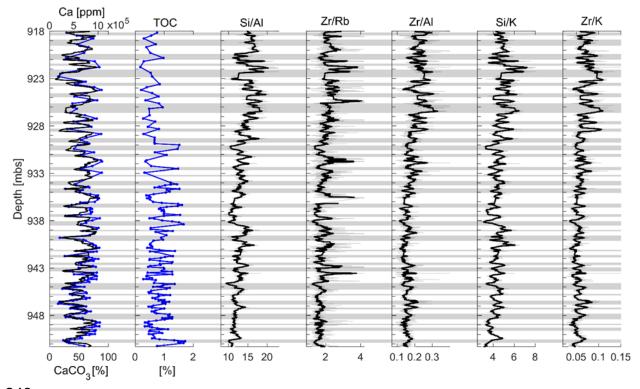
- Principal component analysis (PCA) was performed to examine a potential change in the proxy data before and after the +vie CIE. This was executed in the software PAST on the normalized dataset including microcharcoal, TOC, CaCO<sub>3</sub>,  $\delta^{13}C_{org}$ , S/I, K/I, primary clay mineralogy, Si/Al, Zr/Rb. The samples before the +ve CIE (951.0– 930.4 mbs) and the samples after the +ve CIE (930.3–918.0 mbs) are grouped to examine a potential difference in the sedimentary composition before and after the +ve CIE. A Pearson's correlation was executed in Matlab R2017b. The *p* value tests the hypothesis of no correlation against the alternative hypothesis of a positive or negative correlation (significance level at  $p \le 0.05$ ).
- 230
- 231 4 Results

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# 233 4.1 TOC, CaCO<sub>3</sub> and bulk organic carbon isotope ratio mass spectrometry

234 Alternating TOC-enhanced and Ca-rich lithological couplets occur on a metre scale through the studied interval 235 with TOC and CaCO<sub>3</sub> having a strong negative correlation (r = -0.64, p = 0.001). TOC content fluctuates in the 236 range 0.17–1.72 wt% (mean 0.8 wt%) and the highest fluctuations of TOC content are found from 939–930 237 mbs. The CaCO<sub>3</sub> content fluctuates in opposition to TOC and varies between 14 and 89 %. The studied interval 238 is generally high in CaCO<sub>3</sub> (mean 58 %) (Fig. 3). The  $\delta^{13}C_{org}$  displays a minor (~0.5 ‰) shift towards more 239 positive values at ~944 mbs (as reported in Storm et al., 2020; Hollaar et al., 2021). At ~ 930 mbs an abrupt 240 shift of ~1.8 ‰ (Figs. 1, 4; Storm et al., 2020) indicates the onset of the Late Pliensbachian Event (LPE) in the 241 Mochras core. In agreement with this, the results of the present study show a shift from ~ minus 27 per mil to ~ 242 minus 25.15 per mil between 930.8 and 930.4 mbs (Fig. 4). The  $\delta^{13}C_{org}$  data presented here have been divided 243 into three phases: the pre-LPE gradual rise, followed by the +ve CIE, which is subdivided into pulses 1, 2 and 3

- 244 (Fig. 1). After the onset of the positive  $\delta^{13}C_{org}$  excursion, the TOC content drops to the lowest values (from 0.85
- 245 % before and 0.6 % after the +ve CIE on average), but the 1 metre fluctuations continue (Figs. 3, 4). No overall
- change in the CaCO<sub>3</sub> content is observed through the positive carbon-isotope excursion (Fig. 3).





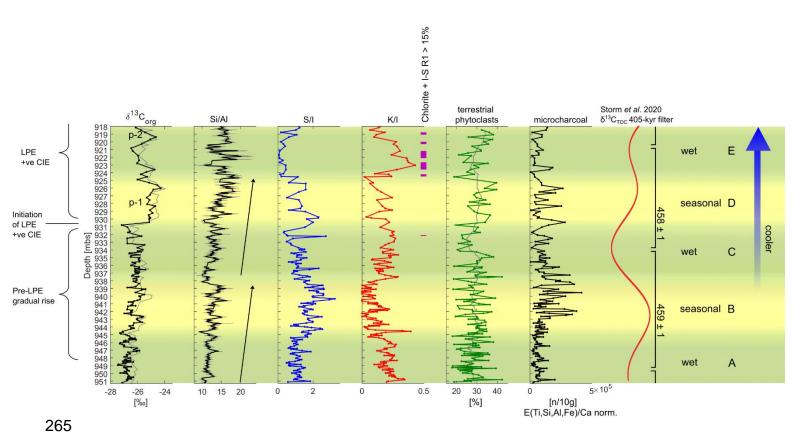
### Figure 3: Detrital ratios over the Ca-rich and TOC-enhanced lithological couplets for the studied

interval. Overview of Ca (black, derived from Ruhl *et al.* 2016), CaCO<sub>3</sub> (blue), and TOC content of the studied
interval 951–918 mbs. The grey shading represents the TOC-enhanced beds and the unshaded bands mark the
Ca-rich (limestone) beds. The detrital ratios reflect the silt to fine sand fraction (Si, Zr) versus the clay fraction
(Rb, Al, K). Two increasing upward cycles are observed in the Si/Al and Zr/Rb ratios. The pattern observed in
all detrital ratios (except the Ti/Al) is similar and likley reflects overall upwards coarsening.

255

## 256 4.2 Clay minerals

- 257 XRD analysis shows that the main clay types found in this interval are illite, random illite-smectite mixed-layers
- 258 (I-S R0) [hereafter referred to as smectite], and kaolinite. Illite and kaolinite co-fluctuate in the interval studied
- 259 here, and are directly out of phase with smectite abundance. Chlorite and R1 I-S are present in minor
- proportions, but reach sporadically higher relative abundance (> 10 %) from ~ 932 mbs upwards, with sustained
- 261 >10% abundance at ~925–918 mbs (Fig. 4 and SI Fig. 1). The relative abundances of smectite and illite and of
- kaolinite and illite are expressed by the ratio S/I and K/I respectively. These ratios were calculated according to
- the intensity of the main diffraction peak of each mineral.



## Figure 4: Synthesis diagram showing the climatic swings observed in tandem with the long eccentricity

267 cycle. The studied interval (Upper Pliensbachian Margaritatus Zone) comprises part of the pre-LPE gradual rise, 268 the initiation of the LPE +ve CIE and pulse 1 and 2 ( $\delta^{13}C_{org}$  data in black from this study and in light grey from 269 Storm et al. (2020)). Five climatic phases (A-E) are interpreted from the Si/Al, smectite/illite, kaolinite/illite, 270 chlorite and I-S R1 abundance and the microcharcoal abundance. In tandem with the 405 kyr cycle (Storm et al., 271 2020) climatic state of a year-round wet climate, low fire activity and fine-grained sediments across multiple 272 precession cycles (phase A and C) alternates with a climatic state that includes repeated precessionally driven 273 states that are semi-arid, with high fire activity and coarser sediments (phase B and D). The top of the record 274 (phase E) indicates increased physical erosion (chlorite + I-S R1, kaolinite) relative to chemical weathering. In 275 terrestrial phytoclast column grey line = 10-step moving average.

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## 278 4.3 Organic matter

279 The type of particulate organic matter, and more specifically the abundance in the marine versus terrestrial

- origin of the particles, fluctuates on a metre scale from 18–42 % (Fig. 4, SI Fig. 2). Palynofacies indicate that
- the type of organic matter does not change in relation to the metre-scale lithological facies cycles (no correlation
- between percentage terrestrial phytoclasts and TOC or CaCO<sub>3</sub>). No large and abrupt changes are recorded in the
- terrestrial/marine proportions, but the proportion of terrestrial phytoclasts has 4 high phases: between 944.6 and

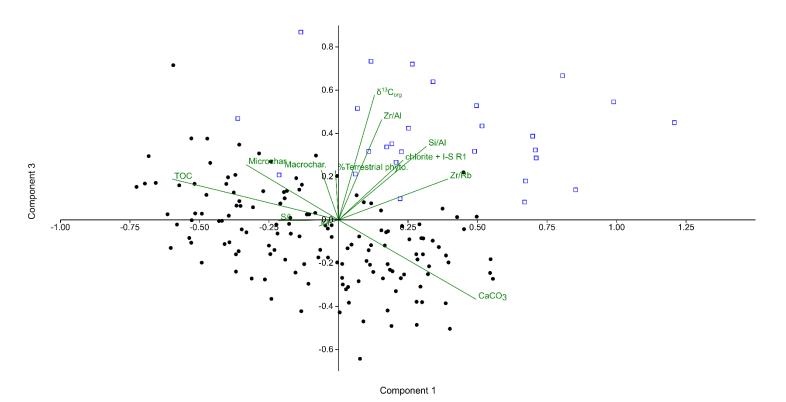
- 284 942.0 mbs, 937.5 and 934.9 mbs, 930.4 and 925.4 mbs, and 920.3 and 918.0 mbs (SI Fig. 2). The first and
- 285 second high phase falls within the + 0.5 % positive swing in the  $\delta^{13}C_{org}$  whilst the latter two high phases
- correspond to pulse 1 and pulse 2 in the +ve CIE. Amorphous organic matter (AOM) is very abundant, followed
- by unstructured phytoclasts, with lower proportions of structured phytoclasts and charcoal (SI Fig. 3). Charcoal
- particles make up a relatively large proportion of the terrestrial particulate organic matter (~10 % on average)
- and ~3.5 % on average of the total particulate organic matter fraction (SI Fig. 3). Only sparse marine and
- terrestrial palynomorphs were observed (SI Fig. 3).
- 291 To assess the character of the observed fluctuations in microcharcoal abundance, whether changes in
- microcharcoal can be related to enhanced runoff from the land and/or organic preservation, or if the
- 293 microcharcoal signifies changes in fire activity on land, the charcoal record was corrected for detrital influx. We
  294 adjust the charcoal particle abundances using the XRF elemental record, normalizing to the total terrigenous
- influx following Daniau *et al.* (2013) and Hollaar *et al.* (2021). The stratigraphic trends in the normalized
- microcharcoal for  $E_{ter}/Ca$ , Si/Al, Ti/Al and Fe/Al remain the same (SI Fig. 4). The absolute number of
- 297 microcharcoal particles decreases, with raw mean charcoal particles  $1.06 \times 10^5$  per 10 g and E<sub>ter</sub>/Ca normalized
- 298 mean 9.7 x  $10^4$  n/10g, Ti/Al normalized 6.4 x  $10^4$  n/10g, Si/Al normalized 7.7 x  $10^4$  n/10g, Fe/Al normalized
- $9.8 \times 10^4$  mean number of microcharcoal particles per 10 g (SI Fig. 4). The number of microcharcoal particles per
- 300 10 g processed rock decreases when correcting for terrestrial run-off changes, hinting that perhaps part of the
- 301 'background' microcharcoal is related to terrestrial influx; the normalisation also shows that the observed
- 302 patterns in microcharcoal abundances are not influenced by changes in terrestrial runoff and taphonomy. Hence,
- the highs and lows in the microcharcoal record can be interpreted to represent changes in the fire regime on
- 304 land. The microcharcoal abundance fluctuates strongly in the record presented here; however, no clear
- difference in microcharcoal content has been observed before and after the onset of the +ve CIE.
- 306

## **307 4.4 Detrital elemental ratios (XRF)**

308 Strong similarities are observed between the fluctuating ratios of Si/Al, Si/K, Zr/Rb, Zr/Al and Zr/K (Fig. 3). 309 The elements Al, Rb and K sit principally in the clay fraction (e.g. Calvert and Pederson, 2007), whereas Si and 310 Zr are often found in greater abundance in the coarser fraction related to silt and sand grade quartz and heavy 311 minerals (Calvert and Pederson, 2007). The ratios all show clear metre-scale fluctuations, and these are 312 superimposed on two increasing-upward trends observed in both the Si/Al and the Zr/Rb, followed by a drop 313 and rise to peak values in the latest part of phase D and phase E above the onset of the +ve CIE (Figs. 3, 4). A 314 parallel trend is observed between the clay ratios (XRD) and elemental ratios Si/Al and Zr/Rb (Fig. 3). Phases of 315 high S/I correspond to the peaks in the two coarsening upward sequences, whereas phases of high K/I 316 correspond to the low phases in the two coarsening upward sequences. After the +ve CIE onset (in phase E) this 317 relationship turns around, and an enrichment in the kaolinite/illite ratio corresponds to the elemental ratios, 318 where highest kaolinite relative abundance is observed in parallel with elemental ratios suggesting maximum 319 coarse fraction.

# 321 4.5 PCA analysis

- 322 The proxy datasets ( $\delta^{13}C_{org}$ , TOC, percentage terrestrial phytoclasts, microcharcoal, smectite/illite,
- 323 kaolinite/illite, abundance of chlorite and R1 I-S, Si/Al, Zr/Rb, Zr/Al) were normalized between 0–1 and run for
- 324 PCA analysis in PAST. Sixty-four percent of the variance is explained by the first three axes (PCA-1 27.7 %,
- **325** PCA-2 19.7 %, PCA-3 15.3 %) inside the 95 % confidence interval.
- 326 PC-1 mainly explains the anti-correlation of TOC and CaCO<sub>3</sub>. PC-2 shows the anti-correlation of K/I and S/I.
- 327 Positive loadings were observed for S/I, microcharcoal, macrocharcoal and CaCO<sub>3</sub>. For PC-2, negative loadings
- 328 were observed for K/I, abundance of chlorite + I-S R1. PC-3 shows strong positive loadings (> 0.3) for  $\delta^{13}C_{org}$ ,
- 329 Si/Al and Zr/Al.
- Plotting PC-1 (y-axis) over PC-3 (x-axis) shows that the samples after the onset of the +ve CIE are grouped to
- the top of the y-axis (more associated with S/I compared to K/I) and to the right of the x-axis (more associated
- 332 with primary minerals, phytoclasts, and higher Si/Al, Zr/Rb and Zr/Al) (Fig. 5).





- **334 the LPE +ve CIE in the Mochras core.** PCA plot of PC-1 and PC-3: all samples before the onset of the LPE
- +ve CIE are marked in black closed circles and the samples after the onset of the LPE +ve CIE are marked in
- blue open squares.
- 337
- 338 5 Discussion

- 339 Figure 1 provides the context for the LPE 'cooling event' at Mochras set within the background record. Shifts in
- bulk  $\delta^{18}O_{carb}$  are coeval to the  $\delta^{13}C_{org}$  change to heavier isotopic values (~930 mbs) and reach a maximum in the
- 341 Margaritatus Zone (>1 ‰) (Ullmann *et al.*, 2022). The bulk oxygen-isotope excursions of Mochras are affected
- 342 by diagenesis and are deemed unlikely to reflect environmental conditions (Ullmann *et al.*, 2022). However,
- 343 oxygen isotope data from marine benthic and nektobenthic molluscs and brachiopods show heavier values
- during the late Margaritatus Zone concurrent with a positive shift in  $\delta^{13}C_{org}$ , indicating cooling during the LPE
- in the nearby Cleveland Basin (Robin Hood's Bay and Staithes) (Korte and Hesselbo, 2011) and this trend is
- also observed in several European sections (e.g. Korte *et al.*, 2015). The duration of the +ve CIE has been
- estimated as ~0.4–0.6 Myr in the Cardigan Bay Basin (Ruhl et al., 2016; Storm et al., 2020; Pieńkowski et al.,
- 348

2021).

349

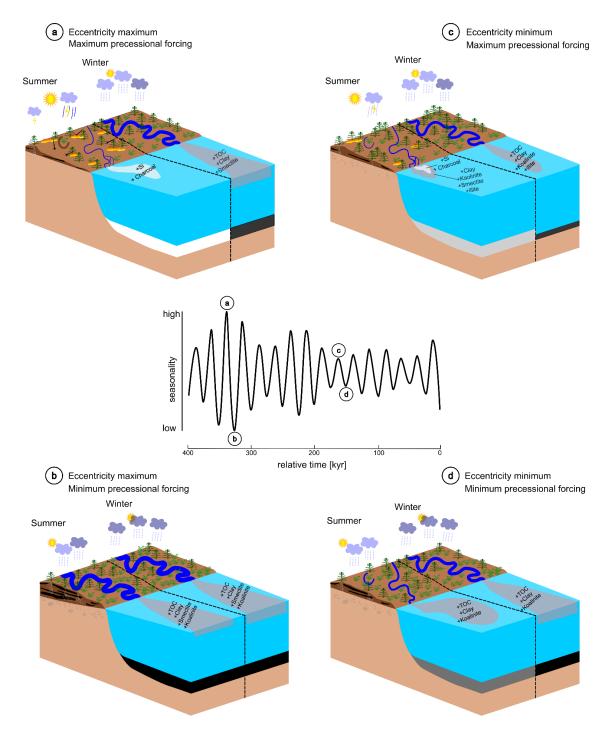
# 350 5.1 Background sedimentological and environmental variations

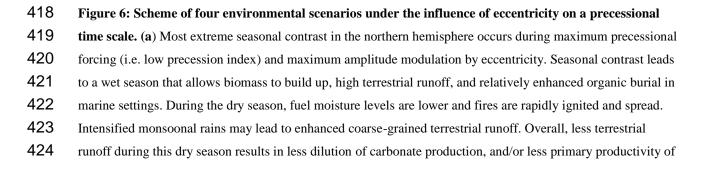
351 The Mochras succession shows metre-scale alternating TOC-enhanced and Ca-rich lithological couplets 352 (mudstone/limestone; Fig. 3). Previous assessments of the palaeoenvironmental signature of these TOC-353 enhanced and Ca-rich couplets indicate strongly that the different depositional modes are driven by orbital 354 precession (Ruhl et al., 2016; Hinnov et al., 2018; Storm et al., 2020; Hollaar et al., 2021; Pieńkowski et al., 355 2021). Precession driven changes in monsoonal strength have been suggested to influence the deposition and 356 preservation of TOC and carbonate in the Cardigan Bay Basin (Ruhl et al., 2016), although the impact may have 357 been expressed, at least partially, by changes in strength of bottom currents in the seaway as a whole 358 (Pieńkowski et al., 2021).

359 The preservation of primary carbonate is poor in the Mochras borehole, making it complex to determine in 360 detail the relative importance of carbonate producers for the bulk carbonate content (Ullmann et al., 2022). 361 However, Early Jurassic, pelagic settings in the Tethys region often received abiotic fine-grained carbonate from 362 shallow marine carbonate platforms (Weedon, 1986; Cobianchi and Picotti, 2001; Krencker et al., 2020) and 363 partly via carbonate producing organisms (such as coccolithophores in zooplankton pellets) (Weedon, 1986; van 364 de Schootbrugge et al., 2005, e.g. Weedon et al., 2019; Slater et al., 2022). Coccolithophores are often poorly 365 preserved and recrystallized (Weedon, 1986; Weedon et al., 2019; Slater et al., 2022). The organic matter found 366 in the studied section of the Mochras borehole varies between 18 and 42% terrestrial phytoclasts (Fig. 4). 367 Phytoclasts are common, but palynomorphs are relatively sparse and poorly preserved. Marine amorphous 368 organic matter is the main constituent in the present study of particulate organic matter in unsieved macerated 369 samples, in the interval studied here (951-918 mbs). Examination of variations in the terrestrial/marine 370 proportions of organic matter, shows no correspondence between the type of organic matter and the TOC-371 enhanced or Ca-rich lithological alternations. However, previous research has indicated that the percentage of 372 terrestrial phytoclasts show precession forcing independent of the lithological couplets (so out of phase with 373 precession scale changes in Ca-TOC content) between 951-934 mbs in the Mochras core (Hollaar et al., 2021). 374 Such orbital forcing of the terrestrial vs marine proportions of organic matter were also found in Early Jurassic 375 sediments of Dorset, and were similarly independent of the lithological facies (Waterhouse, 1999). Terrestrial 376 phytoclast content show a weak expression of long-eccentricity driven variations in the section studied (Fig. 4).

**377** Fossil charcoal makes up a substantial proportion of the organic fractions (11% of the terrestrial fraction) and

- **378**has previously been shown to vary considerably over long-eccentricity cycle  $459 \pm 1$  peaking in abundance
- during the phase of maximum eccentricity (Hollaar *et al.*, 2021). Microcharcoal also appears to be most
- abundant during the maximum phase of the subsequent long eccentricity cycle  $458 \pm 1$  (Fig. 4). Additionally,
- **381** K/I and S/I clay mineral ratios appear to alternate in response to long-eccentricity drivers (Fig. 4) up to 931 mbs
- where the clay mineral signature changes. Detrital clays form in soil weathering profiles and/or physical
  weathering of bedrock. Chemical weathering is enhanced in a high humidity environment with relatively high
- 384 temperatures and rainfall, when clays are formed in the first stages of soil development. In the modern day,
- temperatures and rainfall, when clays are formed in the first stages of soil development. In the modern day,kaolinite is primarily formed in tropical soils, under year-round rainfall and high temperatures (Thiry, 2000).
- 386 Smectite also occurs in the tropics, but is more common in the subtropical to Mediterranean regions, where
- humidity is still high, but periods of drought also occur (Thiry, 2000). Hence, smectite forms predominantly in
- soil profiles under a warm and seasonally dry climate (Chamley, 1989; Raucsik and Varga, 2008), and kaolinite
- in a year-round humid climate (Chamley, 1989; Ruffell *et al.*, 2002). Similarly, alternating intervals of kaolinite
- and smectite dominance were observed for the Late Sinemurian (Munier *et al.*, 2021) and the Pliensbachian of
- 391 Mochras (Deconinck *et al.*, 2019).
- 392 The predominantly detrital character of these clay minerals has been confirmed by TEM scanning of
- **393** Pliensbachian smectite minerals, which revealed the fleecy morphology and lack of overgrowth (Deconinck *et*
- *al.*, 2019). Therefore, the alternations of smectite and kaolinite are interpreted to reflect palaeoclimatic
- 395 signatures of a changing hydrological cycle, with a year-round wet climate evidenced by high K/I ratios, and a
- more monsoon-like climate with seasonal rainfall with high S/I (Deconinck *et al.*, 2019; Hollaar *et al.*, 2021;
- 397 Munier *et al.*, 2021) (See Figs. 3, and 6). The intervals with a signal for weaker seasons appears to correspond to
- 398 phases of low eccentricity in the 405 kyr cycle, and signals of greater seasonality with periods of high more
- pronounced eccentricity (Fig. 4) in the 405 kyr cycle. Between 951 and 930 mbs high K/I occurs during phases
- 400 of low long eccentricity suggesting an enhanced hydrological cycle (Hollaar *et al.*, 2021) with more intense
- 401 weathering, and enhanced fine grained terrestrial runoff to the marine record (Deconinck *et al.*, 2019). In
- 402 contrast, phases of maximum long-eccentricity appear to be smectite-rich, indicating seasonal rainfall, enhanced
- 403 fire (Hollaar *et al.*, 2021) and thus periods of droughts, and lower terrestrial runoff and subsequent lower
- dilution (Deconinck *et al.*, 2019).
- 405 Detrital elemental ratios increase accordingly during the smectite-rich phases, and are lower during kaolinite-
- 406 rich phases between 951 and 930 mbs. Detrital elemental ratios can be used to explore changes in sediment
- 407 composition (e.g. Thibault *et al.*, 2018; Hesselbo *et al.*, 2020b) and the similarity of the long-term trend in Zr/Rb
- 408 and Si/Al (Fig. 3) indicates that these elemental ratios reflect grainsize. The clay fraction (hosting Al, and Rb
- 409 (Chen *et al.*, 1999)), diminishes upwards, whereas the coarser silt to sand fraction (associated with Si (Hesselbo
- 410 *et al.*, 2020b) and Zr (Chen *et al.*, 2006)), increases upward (Figs. 3, 4). The grainsize changes inferred here
- 411 reflect two overall coarsening upwards sequences (Figs. 3, 4). These sequences may reflect changes in clastic
- 412 transport due to changes in the proximity to the shore/siliciclastic source, changes in runoff due to a changing
- 413 hydrological cycle, changes in the intensity of weathering of the bedrock, or accelerated bottom currents with
- 414 greater carrying capacity of coarser sediments.





- 425 organic plankton. (b) Minimum precessional forcing and maximum amplitude modulation of eccentricity leads
- 426 to the least seasonal contrast. Chemical weathering on land is more intense during this year-round humid
- 427 climate. And although biomass is abundant, fire is suppressed due to the high moisture status. Both seasons are
- 428 humid and have considerable terrestrial runoff, resulting in marine organic burial. (c) Moderate seasonality
- 429 occurs during maximum precessional forcing and minimum amplitude modulation of the eccentricity cycle.
- 430 During the wet season biomass grows, and during the dry season fires can occur due to drier fuel conditions.
- 431 However, due to a lesser seasonal contrast the dry conditions are less pronounced and fire is not widespread.
- 432 Runoff includes coarse- and fine-grained sediments, and charcoal during the dry-season. (d) Seasonal contrast is
- 433 low during minimum precessional forcing and minimum amplitude modulation of the eccentricity cycle. Both
- 434 seasons were humid and experienced runoff of fine-grained sediments and organic burial in marine settings.
- 435 Moderately thick soil profiles could develop under this humid climate (figure developed from Martinez and
- **436** Dera, 2015).
- 437

## 438 5.2 Depositional and environmental changes before and after the LPE +ve CIE

## 439 5.2.1 Climate forcing of the hydrological cycle

- 440 The LPE +ve CIE begins around 930 mbs in the Mochras core and encompasses the remaining part of the
- studied section (Fig. 4). We contrasted all the pre-CIE sediment signatures with those of the +ve CIE signatures
- 442 using principal components analysis which indicates distinctly different sedimentary composition and
- 443 environmental signature before and after the onset of the +ve CIE in Mochras (Fig. 5).
- 444 Before the +ve CIE onset, the clay mineral assemblage shows alternating phases of smectite and kaolinite, 445 indicating pedogenic weathering. The relative abundance of the detrital clay types observed in the studied 446 interval have the potential to hold important palaeoclimatic information regarding the hydrological cycle and the 447 relative proportion of chemical weathering and physical erosion. The hydrological cycle was forced by the 405 448 kyr eccentricity before the +CIE, with alternating eccentricity maxima linked to enhanced seasonality (smectite) 449 and eccentricity minima to an equitable wet climate (kaolinite) (Figs. 3, 6). and Higher frequency cycles are not 450 observed in the clay mineral ratios, with no precession or obliquity forcing detected in the high-resolution part 451 of the study 951–934 mbs (Hollaar et al., 2021) and no expression of the 100 kyr cycle in the record presented 452 here. The formation of developed kaolinite-rich, and to a lesser extent smectite-rich soil profiles, requires a 453 steady landscape for many tens of thousands of years, although the  $\sim 1$  Myr timescale of Thiry (2000) seems 454 excessive in our case given the clear expression of clay mineral changes through long-eccentricity cycles. Also, 455 the transportation and deposition of continental clays will occur after soil formation and add further time 456 between formation and final deposition (Chamley, 1989; Thiry, 2000). Thus, there is likely to be a lag of the 457 climatic signal observed in the marine sediments (Chamley, 1989; Thiry, 2000). However, we note that high 458 frequency climatic swings have been recorded in the clay mineral record in some instances, such as in the Lower 459 Cretaceous in SE Spain (Moiroud et al., 2012). The limestone-marl alternations there are enhanced in smectite 460 versus kaolinite and illite, respectively, reflecting precession scale swings from a semi-arid to a tropical humid 461 climate (Moiroud et al., 2012). Precession and higher frequency shifts in the clay record are likely caused by
- 462 fluctuations in runoff conditions rather than the formation of soils with a different clay fraction.

463 Directly after the initial +ve CIE shift from 930-924 mbs (Phase 1 of Fig. 4) little seems to change, and the 464 system evidently continued to respond as before to the long eccentricity forcing, despite the predicted cooling 465 (Korte and Hesselbo, 2011; Korte et al., 2015; Gómez et al., 2016). However, from around 924 mbs up to the 466 top of the studied section (Phase 2 of Fig. 4) the clay mineral assemblage displays a distinctly different 467 composition, with kaolinite dominating especially the early part of phase 2 of the LPE (Fig. 4). At the same time 468 there is an enhancement of the primary minerals illite and chlorite, and I-S R1 (Fig. 4 and SI Fig. 1). Although 469 an enhancement in detrital kaolinite indicates an acceleration of the hydrological cycle, detrital kaolinite is dual 470 in origin and can also be derived from reworking of the primary source material (Deconinck et al., 2019). If the 471 climate is cooler, chemical weathering becomes less dominant and physical erosion of the bedrock becomes the 472 main detrital source of clay minerals. In the Cardigan Bay Basin, the bedrock of the surrounding Variscan 473 massifs (such as the Scottish, Welsh and Irish massifs) were a likely source of these clays. In the Early Jurassic 474 of the NW Tethys region, Lower Paleozoic mudrocks bearing mica-illite and chlorite were emergent (Merriman, 475 2006; Deconinck et al., 2019), hence the enhancement of illite and chlorite likely indicates physical erosion in 476 the region surrounding the study site. Finally, authigenic clay particles could have been formed during burial 477 diagenesis. At temperatures between 60-70 °C smectite illitization occurs and I-S R1 is formed; however, the 478 high abundance of smectite in Mochras indicates limited burial diagenesis at that location (Deconinck et al., 479 2019). Weak-moderate thermal diagenesis is confirmed for the Pliensbachian of Mochras, with  $T_{max}$  from 480 pyrolysis analysis between 421 °C and 434 °C (van de Schootbrugge et al., 2005; Storm et al., 2020). Therefore, 481 I-S R1 in Mochras is interpreted to be derived from chemical weathering of illite (Deconinck et al., 2019). The 482 coeval increase of these primary clay minerals, I-S R1 and kaolinite, indicate that during this period physical 483 erosion dominated over soil chemical weathering (Deconinck et al., 2019; Munier et al., 2021). This is similar 484 to what has been observed for the latest Pliensbachian in Mochras previously (Deconinck et al., 2019).

485 Erosion of weathering profiles transports clay minerals (including kaolinite and smectite) to the marine realm. In 486 the ocean, the differential settling of kaolinite (near shore) and smectite (more distal) could occur based on the 487 morphology and size of clay particles (Thiry, 2000). However, comparison of long-term inferred regional sea 488 level changes from surrounding UK basins (Hesselbo, 2008) suggests that the relative proportions of smectite 489 and kaolinite are not influenced by changes in relative sea level in the Pliensbachian of Mochras (Deconinck et 490 al., 2019). On the assumption that the coarsening upward sequences at Mochras are indicative of relative sea 491 level change, it can also be argued that the proximity to shore did not impact the proportions of smectite and 492 kaolinite. Instead, we observe enhanced smectite during 'proximal' deposition and enhanced kaolinite at times

- 493 of more 'distal' deposition, the opposite of what might be expected (Fig. 4).
- 494 We suggest that the first phase of the LPE (Fig. 4, phase 1) was characterised by repeated periods of rainfall in a
- seasonal climate forced by precession in which chemical weathering (smectite formation) dominated the
- sedimentary signatures. This corresponds to maximum long-eccentricity and shows the same climatic signature
- 497 as during maximum eccentricity phases before the +ve CIE. This is then followed by a second phase (Fig. 4,
- 498 phase 2) where the climate is generally cooler, overall potentially more arid, but with rainfall throughout the
- 499 year over multiple precession cycles. This appears to have favoured deep physical erosion, owing to the
- abundance of primary clay minerals, kaolinite and I-S R1. This interval corresponds to a minimum phase in the

501 405 kyr eccentricity based on Storm et al. (2020). This interpretation is further supported by decreasing and then 502 low microcharcoal abundance, pointing to suppression of fire activity at this time.

503

#### 504 5.2.2 Climate forcing of sedimentary changes

505 Two coarsening upward cycles that predate the onset of the +ve CIE and continue for a few metres after its 506 initiation, are present in the detrital elemental ratios (best expressed in Si/Al and Zr/Rb records) (Figs. 3, 4), and 507 indicate a changing sediment influx over the studied interval. Previous study of the lithofacies of the Mochras 508 borehole has also shown the coarsening-upward sequences of 0.5-3 m thickness, which are observed to be 509 followed upwards by a thinner fining-upward succession (Pieńkowski et al., 2021). This reported fining-upward 510 part is not reflected in the elemental ratios of the two sequences shown in this study. Furthermore, the coarsest 511 phases of these sequences are approximately coeval with decreasing trends in the K/I ratio and increasing trends 512 in the S/I. This could indicate that periods of a strong monsoonal/seasonal climate (indicated by S/I) brought 513 coarser grained material to the basin, whereas periods of year-round humidity (K/I) are associated with higher 514 chemical weathering (low Si/Al). Therefore, these two coarsening upwards cycles appear to link to increasing 515 long-eccentricity. A similar mechanism has been inferred for the northern South China Sea region in the 516 Miocene, where coarser grained material is found during periods of a strong summer monsoon and relatively 517 lower chemical weathering (Clift et al., 2014). Present day studies show that bedrock erosion and associated 518 sediment transport is greater in areas with high seasonal contrast (Molnar, 2001; Molnar, 2004). Hence, the 519 Si/Al record also appears to reflect weathering and erosion conditions on land (Clift et al., 2014, 2020), driven 520 by long-eccentricity modulated climate (SI Fig.5). However, other scenarios that would influence the grain size 521 on this time scale cannot be dismissed and include changes in proximity to siliciclastic source, or changes in 522 sediment transport via bottom water currents.

- - 523 Changes in bottom water current strength and direction likely affected the depositional site of the Mochras core 524 (Pieńkowski et al., 2021) although there is as yet no consensus on the processes that likely controlled these
  - 525 palaeoceanographic parameters. In the UK region, the North Sea tectonic dome structure may have disrupted the
- 526 circulation in the N-S Laurasian Seaway (including the Viking Corridor) in the Late Pliensbachian when global
- 527 sea-levels are suggested to have been low (Haq 2018) and therefore diminished the connectivity between
- 528 western Tethys and the Boreal realm, hypothetically reducing poleward heat transport from the tropics (Korte et
- 529 al., 2015). This mechanism has also been argued to explain the later cooling observed in NW Europe during the
- 530 transition of the warmer Toarcian to the cooler Aalenian and Bajocian (Korte et al., 2015). Late Pliensbachian
- 531 occlusion of the Viking Corridor is supported by the provincialism of marine faunas at this time, showing a
- 532 distinct Euro-Boreal province and a Mediterranean province (Dera et al., 2011b). During the Toarcian, a
- 533 northward expansion of invertebrate faunal species has been found (Schweigert, 2005; Zakharov et al., 2006;
- 534 Bourillot et al., 2008; Nikitenko, 2008), indicating a northward (warmer) flow through the Viking corridor
- 535 (Korte et al., 2015). More recently, a southward expansion of Arctic dinoflagellates into the Viking Corridor
- 536 was suggested for the termination of the T-OAE (van de Schootbrugge et al., 2019), which is in agreement with
- 537 a N to S flow through the Viking Corridor suggested by numerical models (Bjerrum et al., 2001; Dera and
- 538 Donnadieu, 2012; Ruvalcaba Baroni et al. 2018) and sparse Nd-isotopes (Dera et al., 2009).

- 539 Over the European Epicontinental Shelf (EES), and the Tethys as a whole, a clockwise circular gyre likely
- 540 brought oxygenated warm Tethyan waters to the southwest shelf, with a progressively weaker north and
- 541 eastward flow due to rough bathymetry and substantial islands palaeogeography (Ruvalcaba Baroni et al.,
- 542 2018). This predominantly surface flow is modelled to have extended to shelfal sea floor depths. Only
- 543 episodically might nutrient-rich Boreal waters have penetrated south onto the EES in these coupled ocean-
- 544 atmosphere GCM model scenarios (Dera and Donnadieu, 2012). The modelling also suggests - counter-
- 545 intuitively - that the clockwise surface gyre of the Tethys extended further northwards and impacted the EES
- 546 more effectively when the Hispanic corridor was more open. The timing of the opening of the Hispanic corridor
- 547 is debated and varies from the Hettangian to Pliensbachian (Aberhan, 2001; Porter et al., 2013; Sha, 2019).
- 548 An alternative bottom current configuration was discussed for Mochras specifically wherein changes in north-549 to-south current strength (cf. Bjerrum et al., 2001) are proposed for the changes in grain size and silt or sand
- 550
- versus clay content via contour currents (Pieńkowski et al., 2021). A strong flow from the cooler and shallow
- 551 boreal waters is hypothesized to have brought a coarser grainsize fraction in suspension and as bedload, which
- 552 was then deposited in the Cardigan Bay Basin while flowing to the deeper and warmer waters of the peri-Tethys
- 553 (Pieńkowski et al., 2021). Times of a strong north to south current are proposed to be associated with more
- 554 oxygenated bottom waters (Pieńkowski et al., 2021). In contrast, when the north to south current became
- 555 weaker, less coarse material will have been carried in suspension and as bedload, and a relatively higher clay 556 proportion will have been deposited in the Cardigan Bay Basin (Pieńkowski et al., 2021). In this scenario, times
- 557 of sluggish currents are associated with low bottom water oxygenation (Pieńkowski et al., 2021) and thus
- 558 climate forcing of current strength could explain the deposition of alternating coarser and finer fractions in the
- 559 Mochras borehole (Pieńkowski et al., 2021).
- 560 Our research suggests that orbital cycles both before and during the onset of the +ve CIE have a significant 561 influence on seasonality and hydrology, affecting both fire regimes and sediment depositional character. Further
- 562 research is required to consider how long-eccentricity and obliquity cycles might interact with north-south flow
- 563 in the Cardigan Bay Basin and circulation processes. What is clear is that orbital cycles have impact on
- 564 terrestrial processes in the terrestrial sediment source areas (Hollaar et al., 2021) and led to differences in
- 565 deposition within the marine sediments in the Mochras core (Ruhl et al., 2016; Pieńkowski et al., 2021). Our
- 566 data indicate that periods of coarser sediment deposition correspond to periods that include more seasonal
- 567 climates before the onset of the +ve CIE (low kaolinite), which is in line with the hypothesized grainsize
- 568 changes caused by contour currents (Pieńkowski et al., 2021). However, after the onset of the +ve CIE, although
- 569 we suggest that the chemical weathering rate decreased, enhanced runoff and physical erosion are indicated by a
- 570 peak in primary clay minerals and K/I. Enhanced runoff could be expected to impact the thermohaline contour
- 571 currents (Dera and Donnadieu, 2012). Simultaneously, an increasingly cold climate (as indicated by enhanced
- 572 physical erosion over chemical weathering) indicates a boreal influence. It remains to be determined to what
- 573 extent orbital cycles might have the power to influence ocean circulation in the basin.
- 574 Relatively coarse sediments in the Late Pliensbachian have also been related to shallower sediment deposition in
- 575 UK basins (Hesselbo and Jenkyns, 1998; Hesselbo, 2008; Korte and Hesselbo, 2011). Around the UK area,
- 576 these regressive facies are plausibly related to enhanced sediment shedding from the North Sea dome structure
- 577 during sea level low stand across the region (Korte and Hesselbo, 2011). Sequence stratigraphy of the Lower

- 578 Jurassic of the Wessex, Cleveland and Hebrides basins (Hesselbo and Jenkyns, 1998; Hesselbo, 2008; Archer et
- 579 al., 2019) shows relative sea level changes and sand influxes in the late Margaritatus Zone in the studied basins.
- 580 Noteworthy in the Mochras borehole are phases of low  $\delta^{18}$ O of macrofossils which seem to correspond to high
- 581 phases of macrofossil wood concomitant with low sea level, suggesting a possible control of relative sea level
- 582 on the oxygen-isotope record and the source of detrital material (Ullmann et al., 2022). The broad spatial
- 583 distribution of these basins suggests that associated regression and/or sediment influx is of at least regional scale
- 584 (Hesselbo, 2008). The results presented here fall within this phase of regression (Hesselbo and and Jenkyns,
- 585 1998; Hesselbo, 2008).
- 586 In the context of the North Sea topographic dome structure (occlusion of the Viking Corridor in regional ocean 587 flow) as a possible cause of the Late Pliensbachian cooling, these facies can be interpreted to represent
- 588 shallowing upward in relatively shallow water, or the supply of coarser sediment into a deep-water system. The
- 589 doming is hypothesized to have minimized or prohibited southward flow of cooler waters from the Boreal and
- 590 northward flow from warmer waters from the Mediterranean area (Korte et al., 2015). The Mochras borehole is
- 591 situated on the southwestern flank of the dome and would have been cut-off from the northern parts of the 592
- Laurasian Seaway, including the Hebrides Basin and Cleveland Basin (Korte et al., 2015). This change in
- 593 seaway circulation could have impacted the source area of the detrital sediments in the Mochras borehole.
- 594 Superimposed on these larger-scale factors affecting grain size, orbital forcing clearly also had a strong impact.
- 595 The Cardigan Bay Basin (Mochras) is positioned about 290 km to the SW of the Cleveland Basin and at a
- 596 similar latitude, but to the W of the Wessex Basin (Ziegler, 1990; Torsvik and and Cocks, 2017), and is therefore
- 597 expected to be impacted by the same regional changes in sea level and/or sediment flux. In the Late
- 598 Pliensbachian of the Cleveland Basin, the detrital ratios of Si/Al, Zr/Al and Zr/Rb show similar coarsening
- 599 upward sequences, which have been interpreted to reflect changes in riverine transport of siliciclastic grains and
- 600 grainsize (Thibault et al., 2018). The inferred changes in sea-level in the Cleveland Basin occur at a 100 kyr
- 601 pacing (Huang and and Hesselbo, 2014; Hesselbo et al., 2020b), potentially linking the regression cycles to short
- eccentricity (Huang et al., 2010 and refs therein) and long-eccentricity (Thibault et al., 2018). This would mean 603 that eccentricity driven changes in inferred sea level change could be linked to glacioeustatic cycles during these
- 604 times (Brandt, 1986; Suan et al., 2010; Korte and and Hesselbo, 2011; Krencker et al., 2019; Ruebsam et al.,
- 605 2019, 2020b; Ruebsam and Schwark, 2021; Ruebsam and Al-Husseini, 2021). Glacioeustatic sea level changes
- 606 are discussed for the Early Jurassic and Middle Jurassic (Krencker et al., 2019; Bodin et al., 2020; Ruebsam and
- 607 Schwark, 2021; Nordt et al., 2022). A recent study on the rapid transgression observed at the Pliensbachian-
- 608 Toarcian boundary, ruled out other mechanisms that could force sea level at this time scale, such as aquifer-
- 609 eustacy, and emphasise that glacioeustatic changes in sea level are a likely possibility at times in the Early
- 610 Jurassic (Krencker et al., 2019). Therefore, our findings overall are compatible with the episodic occurrence of
- 611 continental ice at the poles (Brandt, 1986; Price, 1999; Suan et al., 2010; Korte and Hesselbo, 2011; Korte et al.,
- 612 2015; Bougeault et al., 2017; Krencker et al., 2019; Ruebsam et al., 2019, 2020a, 2020b; Ruebsam and
- 613 Schwark, 2021; Ruebsam and Al-Husseini, 2021).
- 614

#### 615 **6** Conclusions

- 616 The terrestrial environment adjacent to the Cardigan Bay Basin was strongly influenced by orbitally driven
- 617 climate forcings (particularly precession and eccentricity) and colder climate linked to the Late Pliensbachian
- Event (LPE).Long-eccentricity forcing remained strong both prior to and during the LPE. Our results identify
- five swings in the climate in the study interval in tandem with the 405 kyr eccentricity minima and maxima.
- 620 Eccentricity maxima are linked to precessionally repeated occurrences of a semi-arid monsoonal climate with
- high fire activity and relatively coarser sediment from terrestrial runoff. In contrast, 405 kyr minima in the
- 622 Mochras core are linked to a more persistent, annually wet climate, low fire activity, and relatively finer grained
- deposits across multiple precession cycles. Although the 405 kyr cycle in the proxy records persists through the
- 624 onset of the LPE +ve CIE, the expression in the clay mineralogical record changes to indicate year-round
- relatively cool and wet climate extended over multiple precession cycles driving significant erosion of bedrock.
- 626 Therefore, both the Milankovitch forcings and larger climatic shifts operate in tandem to govern changes in the627 terrestrial environment.
- 628
- **629 Data availability**: Supplementary data is available at the National Geoscience Data Centre at Keyworth
- 630 (NGDC) at (doi to be added) for the interval 934–918 mbs. All data presented for the interval 951–934 mbs is
- available at the National Geoscience Data Centre at Keyworth (NGDC) at <a href="https://doi.org/10.5285/d6b7c567-49f0-44c7-a94c-e82fa17ff98e">https://doi.org/10.5285/d6b7c567-49f0-44c7-a94c-e82fa17ff98e</a> (Hollaar *et al.*, 2021). The full Mochras XRF dataset is in Damaschke *et al.*
- **633** (2021).
- Author contribution: CMB, SPH and TPH designed the research. TPH conducted the laboratory
- 635 measurements, with JFD contributing to the XRD-measurements and MD, CU and MJ to the XRF-
- 636 measurements. TPH, CMB and SPH wrote the manuscript, with contributions from all authors.
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