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Carbon cycle history through the JurassicCretaceous boundary: A new global 13C stack

Watanabe, Sayaka

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4	Carbon cycle history through the Jurassic-Cretaceous boundary:
5	a new global $\delta^{13}\text{C}$ stack
6	Gregory D. Price ¹ , István Főzy ² , József Pálfy ^{3, 4}
7	¹ School of Geography, Earth & Environmental Sciences, Plymouth University, Drake Circus, Plymouth,
8	PL4 8AA, United Kingdom (g.price@plymouth.ac.uk)
9	² Department of Paleontology and Geology, Hungarian Natural History Museum, POB 137, Budapest, H-
10	1431 Hungary (fozy@nhmus.hu)
11	³ Department of Physical and Applied Geology, Eötvös Loránd University, Pázmány P. sétány 1/C,
12	Budapest, H-1117 Hungary (palfy@nhmus.hu)
13	⁴ MTA-MTM-ELTE Research Group for Paleontology, POB 137, Budapest, H-1431 Hungary
14	
15	ABSTRACT
16	We present new carbon and oxygen isotope curves from sections in the Bakony Mts. (Hungary),
17	constrained by biostratigraphy and magnetostratigraphy in order to evaluate whether carbon isotopes
18	can provide a tool to help establish and correlate the last system boundary remaining undefined in the
19	Phanerozoic as well provide data to better understand the carbon cycle history and environmental

20 drivers during the Jurassic-Cretaceous interval. We observe a gentle decrease in carbon isotope values 21 through the Late Jurassic. A pronounced shift to more positive carbon isotope values does not occur 22 until the Valanginian, corresponding to the Weissert event. In order to place the newly obtained stable 23 isotope data into a global context, we compiled 31 published and stratigraphically constrained carbon isotope records from the Pacific, Tethyan, Atlantic, and Boreal realms, to produce a new global δ^{13} C 24 25 stack for the Late Oxfordian through Early Hauterivian interval. Our new data from Hungary is consistent with the global δ^{13} C stack. The stack reveals a steady but slow decrease in carbon isotope 26 values until the Early Valanginian. In comparison, the Late Jurassic–Early Cretaceous δ^{13} C curve in GTS 27 28 2012 shows no slope and little variation. Aside from the well-defined Valanginian positive excursion, 29 chemostratigraphic correlation durSchning the Jurassic-Cretaceous boundary interval is difficult, due to relatively stable δ^{13} C values, compounded by a slope which is too slight. There is no clear isotopic 30 31 marker event for the system boundary. The long-term gradual change towards more negative carbon 32 isotope values through the Jurassic-Cretaceous transition has previously been explained by increasingly 33 oligotrophic condition and lessened primary production. However, this contradicts the reported increase in ⁸⁷Sr/⁸⁶Sr ratios suggesting intensification of weathering (and a decreasing contribution of 34 35 non-radiogenic hydrothermal Sr) and presumably a concomitant rise in nutrient input into the oceans. 36 The concomitant rise of modern phytoplankton groups (dinoflagellates and coccolithophores) would 37 have also led to increased primary productivity, making the negative carbon isotope trend even more 38 notable. We suggest that gradual oceanographic changes, more effective connections and mixing 39 between the Tethys, Atlantic and Pacific Oceans, would have promoted a shift towards enhanced 40 burial of isotopically heavy carbonate carbon and effective recycling of isotopically light organic matter.

- 41 These processes account for the observed long-term trend, interrupted only by the Weissert event in
- 42 the Valanginian.
- 43 **Keywords:** Late Jurassic; Early Cretaceous; chemostratigraphy; carbonate carbon cycle history

44 **1. Introduction**

45 The Jurassic-Cretaceous transition is a relatively poorly understood interval in the development of the Mesozoic greenhouse world (Föllmi, 2012; Price et al., 2013). This is, in part, due to the lack of 46 47 an agreed upon, chronostratigraphic framework for the Jurassic-Cretaceous boundary (Zakharov et al., 48 1996; Wimbledon et al., 2011; Michalík and Reháková, 2011; Guzhikov et al., 2012; Shurygin and 49 Dzyuba, 2015). It is a time of contentious biotic changes, for which opinions have ranged from proposal 50 of a putative mass extinction (Raup and Sepkoski, 1984) or a regional event (Hallam, 1986) or non-51 event (Alroy, 2008; Rogov et al., 2010). Using large taxonomic occurrence databases, several recent 52 studies (particularly of tetrapods) have re-examined the Jurassic-Cretaceous boundary, and note a 53 sharp decline in diversity around the Jurassic-Cretaceous boundary (Barrett et al., 2009; Mannion et al., 54 2011; Upchurch et al., 2011; Tennant et al., 2016). Further, the boundary interval is characterized by 55 elevated extinction and origination rates in calcareous nannoplankton (Bown, 2005) set against a background of several calpionellid diversification events (Remane, 1986; Michalík et al., 2009) and an 56 57 evolutionary rise of the modern plankton groups, notably dinoflagellates and coccolithophores 58 (Falkowski et al., 2004). The system boundary also presents persistent stratigraphic correlation 59 problems, which explains why the Jurassic-Cretaceous boundary is the only Phanerozoic system 60 boundary for which a GSSP (Global Stratotype Section and Point) remains to be defined (Wimbledon, 61 2008; Wimbledon et al., 2011). The problems in global correlation of the Jurassic–Cretaceous boundary 62 arise from the lack of an agreed upon biostratigraphical marker, in part related to general regression 63 leading to marked provincialism in different fossil groups. The Tethyan based ammonite definition for 64 the base of the Cretaceous has been the base of the Jacobi Zone (e.g., Hoedemaeker et al., 1993), 65 although the base of which falls within the middle of relatively long sub-Boreal Preplicomphalus Zone

66 and the Boreal Nodiger Zone. Other definitions of the Jurassic-Cretaceous boundary (see Grabowski, 67 2011; Wimbledon et al., 2011) include the base of Grandis ammonite Subzone, in the lower part of 68 calpionellid Zone B, almost coinciding with the base of magnetozone M18r (Colloque sur la Crétacé 69 inferieur, 1963) or the boundary between Grandis and Subalpina ammonite subzones, correlated with 70 the middle part of calpionellid Zone B and the lower part of magnetozone M17r (Hoedemaeker, 1991). 71 Due to scarcity of ammonites in many Tethyan Tithonian and Berriasian successions, calpionellids have 72 been used as the main biostratigraphic tool in some studies (e.g., Horváth and Knauer, 1986; Blau and 73 Grün, 1997; Houša et al., 2004; Boughdiri et al., 2006; Michalík et al., 2009; Grabowski et al., 2010a). 74 The base of Calpionella Zone (B Zone) and the sudden appearance of a monospecific association of 75 small, globular Calpionella alpina (referred to by authors as the alpina "acme", Remane 1985; Remane 76 et al., 1986) is sometimes used as an indicator of the Jurassic-Cretaceous boundary. The base of 77 reversed-polarity chron M18r has also been suggested as a convenient global correlation horizon near 78 the clustering of these possible biostratigraphic-based boundaries (Ogg and Lowrie, 1986). The 79 recognition of this magnetozone across provincial realms (e.g., Ogg et al., 1991; Houša et al., 2007; 80 Grabowski et al., 2010a) has enabled inter-regional correlations. In the GTS2012, Ogg and Hinnov 81 (2012a) utilize the base of chron M18r for assigning the numerical age (145.0 Ma) to the top of the 82 Jurassic. Notably, the base of chron M18r which falls within the middle of the Berriasella jacobi Zone. 83 Hence, Wimbledon et al. (2011), tentatively suggest that several markers have the potential to help 84 define any putative Jurassic-Cretaceous boundary.

Carbon isotope stratigraphy is useful both to help understand past global environmental and biotic change that affected carbon cycle, and as a correlation tool. For example, the GSSP for the base of the Eocene Series is defined by a negative excursion in the carbon isotope curve (Aubry et al., 2007).

88 To serve both purposes, Late Jurassic–Early Cretaceous carbon isotope stratigraphies have been 89 developed extensively from pelagic sediments of the Tethys Ocean and Atlantic (e.g., Weissert and 90 Channell, 1989; Bartolini et al., 1999; Katz et al., 2005; Tremolada et al., 2006; Michalík et al., 2009; 91 Coimbra et al., 2009; Coimbra and Olóriz, 2012). Weissert and Channell (1989) documented how the Late Jurassic carbonate carbon isotopic composition shifts from δ^{13} C values of around 2.5‰ in the 92 Kimmeridgian to values near 1.0‰ in the Late Tithonian–Early Berriasian. A change to lower δ^{13} C 93 94 values was identified to occur within Magnetozones M18–M17 and within the B/C Calpionellid Zone (Weissert and Channell, 1989). The low δ^{13} C values of the earliest Cretaceous contrast with the more 95 96 positive values obtained from the Valanginian (Lini et al., 1992; Hennig et al., 1999; Weissert et al., 97 1998; Duchamp-Alphonse et al., 2007; Főzy et al., 2010). Such variation has led to the idea that carbon 98 isotopes may be useful in adding to the characterisation of the Jurassic-Cretaceous boundary (e.g., 99 Michalík et al., 2009; Dzyuba et al., 2013; Shurygin and Dzyuba, 2015) although others (e.g., Ogg and 100 Hinnov, 2012a) note the lack of significant geochemical markers. Changes in the Late Jurassic–Early 101 Cretaceous carbon isotope record are interpreted to reflect decelerated global carbon cycling and 102 ocean productivity (Weissert and Mohr, 1996) and have been variously linked to changes in sea level, 103 aridity and temperature (e.g., Weissert and Channell, 1989; Ruffell et al., 2002a; Tremolada et al., 2006; 104 Föllmi, 2012). Other carbon isotope records through the Jurassic–Cretaceous boundary show 105 somewhat different trends. For example, Michalík et al. (2009) documented a minor (<0.5‰) negative 106 excursion in the latest Jurassic (Late Tithonian), whilst some Boreal records (e.g., Žák et al., 2011) show negligible variation associated with the boundary. Dzyuba et al. (2013) reported a positive δ^{13} C shift 107 108 immediately above the Jurassic–Cretaceous boundary. The significance of Jurassic–Cretaceous carbon 109 isotope stratigraphies is underlined by correlation needs for the yet-to-be-defined GSSP.

110 In this study we report new carbon isotope data for the Late Jurassic–Early Cretaceous from 111 two sections, Lókút Hill and Hárskút in Hungary (Figs. 2, 3). Both sections are well constrained by 112 ammonite (Figs. 4, 5), belemnite (Vigh, 1984; Horváth and Knauer, 1986; Főzy, 1990) and calpionellid 113 (Horváth and Knauer, 1986; Grabowski et al., 2010a) biostratigraphy. Magnetostratigraphy is also 114 available for Lókút Hill (Grabowski et al., 2010a). The aim of this study is to assess whether a consistent 115 pattern in carbon isotope variation can be established, particularly with respect to the Jurassic-Cretaceous boundary. To this end, we also developed a new global stack of carbonate δ^{13} C curves for 116 117 the Jurassic–Cretaceous transition (from the Late Oxfordian to Early Hauterivian), based on the two 118 newly obtained curves and a global compilation of 30 published curves from this interval. We use this 119 global stack to evaluate the possible controls on carbon isotope variation (similar to the approach 120 taken by Wendler (2013) for the Late Cretaceous) and the correlation potential of carbon isotope 121 stratigraphy. Comparisons to a range of other climate proxies (including the oxygen isotopic 122 composition of fossil belemnites derived from a range of low and mid Tethyan palaeolatitude sites) and environmental events is also made to help elucidate controls on the global δ^{13} C stack. 123

124 2. Geological setting

125 The studied Hungarian sections are situated ca. 6 km apart from each other in the 126 southwestern part of the central Bakony Mountains (Fig. 1) that belongs to the Transdanubian Range, 127 which in turn forms part of the Bakony Unit in the Austroalpine part of the AlCaPa terrane (Csontos 128 and Vörös, 2004). This complex structural unit stretches from the Eastern Alps to the Western 129 Carpathians. Its Mesozoic sedimentary succession is thought to have deposited on the southern 130 passive margin of the Penninic ocean branch of the western Neotethys (Csontos and Vörös, 2004) (Fig.

131 1). In the lowermost part of the studied sections, the cherty Lókút Radiolarite Formation crops out 132 (Figs. 2, 3). The overlying unit consists of red and yellowish, well-bedded nodular limestone (Pálihálás 133 Limestone Formation), which passes gradually into light grey, less nodular, ammonite-rich facies 134 (Szentivánhegy Limestone Formation). The uppermost part of both sections (Figs. 2, 3) are made up of 135 white, thin-bedded, Biancone-type limestone (Mogyorósdomb Limestone Formation). The boundaries 136 between these formations are gradational. A brief description of these lithostratigraphical units is 137 given in Császár (1997). The studied section at Lókút (referred to as the hilltop section) ranges in age 138 from the late Oxfordian to Berriasian, whereas at Hárskút (section HK-II) upper Kimmeridgian to 139 Berriasian strata are exposed.

140 The entire Jurassic succession of Lókút Hill (exposed in three disjunct sections, of which the 141 hilltop section is the youngest) is the most complete and thickest Hettangian to Tithonian succession of 142 Transdanubian Range, deposited in a deep, pelagic environment (Galácz and Vörös, 1972). In the 143 "horst and graben" palaeogeographic model proposed by Vörös and Galácz (1998), this locality 144 represents a site of typical basinal deposition. The Upper Jurassic–lowermost Cretaceous strata (Fig. 2) 145 are exposed on the southwestern edge of the top of Lókút Hill in an artificial trench (47° 12' 17" N, 17° 146 52' 56" E). The beds gently dip (20°) to the north. Biostratigraphic data from the Tithonian part of the 147 section were first provided by Vigh (1984), later amended and complemented by late Oxfordian and 148 Kimmeridgian cephalopod data by Főzy et al. (2011). In addition, Grabowski et al. (2010a) developed a 149 calpionellid biostratigraphy and magnetostratigraphy for the Tithonian–Berriasian part of the section. 150 Bed numbers of Grabowski et al. (2010a) are still visible, allowing correlation with these data and our 151 isotope results.

152 At Hárskút, two measured Late Jurassic-Early Cretaceous sections (referred to as HK-II and HK-153 12) are exposed on the opposite sides of a small valley, the Közöskút Ravine (Fig. 3). In the ravine itself, 154 a Lower to Middle Jurassic Ammonitico Rosso-type succession crops out. Studies by Fülöp et al. (1969) and Galácz (1975) established the presence of repeated gaps due to non-deposition. Within the "horst 155 156 and graben" palaeogeographic model (Vörös and Galácz, 1998), these strata represent intermittent 157 deposition on an elevated submarine high. Overlying the extremely lacunose Middle Jurassic and the 158 cherty Lókút Radiolarite Formation, the Upper Jurassic limestone succession is more complete. The 159 studied profile (HK-II) is a c. 10 m high natural cliff, also known as "Prédikálószék" ("Pulpit", 47° 09' 160 53,4" N, 17° 47' 7,36" E). It offers excellent outcrop of the fossiliferous Upper Jurassic to lowermost 161 Cretaceous limestone units. For the uppermost Kimmeridgian–Tithonian part of the section, Főzy (1990) 162 established an ammonite-based biostratigraphy, whereas for the Berriasian part of the same profile, 163 calpionellid and ammonite stratigraphy was provided by Horváth and Knauer (1986). The section HK-II 164 described in the present paper is situated a few hundred meters west of a complementary section (HK-165 12), which recently was subject of a detailed integrated stratigraphic study by Főzy et al. (2010), who 166 demonstrated the Late Valanginian positive carbon isotope excursion, known as the Weissert event 167 (Erba et al., 2004).

168 3. Material and methods

A substantial cephalopod fauna was collected from Lókút in 1962–1964 by a team of the Hungarian Geological Institute under the supervision of J. Fülöp. Our re-measuring and re-sampling of the section yielded additional specimens. Cephalopods of the studied section are housed in the Department of Palaeontology of the Hungarian Natural History Museum and partly in the Museum of

the Hungarian Geological and Geophysical Institute. Perusal of the original documentation allowed us
to accurately reconstruct the source beds of the specimens collected nearly 50 years ago and partly
published by Vigh (1984). Ammonites (Figs. 4, 5) are preserved throughout both sections as internal
moulds As the fauna consists of solely Mediterranean (i.e. Tethyan) elements, the ammonite
biostratigraphic zonation of Enay and Geyssant (1975) and Olóriz (1978) were used. Belemnites of
stratigraphical value were collected only from the Tithonian of the Lókút section.

For this study, stable isotope analyses of 165 bulk carbonate samples were taken from sections
at Lókút (hilltop) and Hárskút (HK-II) (Figs. 2, 3). The average spacing of samples was ~0.15 m.

181 Subsamples, avoiding macrofossils and sparry calcite veins, were then analysed for stable isotopes.

182 Carbonate powders were analysed on a GV Instruments Isoprime Mass Spectrometer with a Gilson

183 Multiflow carbonate auto-sampler at Plymouth University, using 250 to 400 micrograms of carbonate.

184 Isotopic results were calibrated against the NBS-19 standard. Reproducibility for both δ^{18} O and δ^{13} C

185 was better than ±0.1‰, based upon duplicate sample analyses.

186 **4. Results**

187 *4.1. Biostratigraphy*

Based on the rich and relatively well-preserved ammonite fauna which was collected bed-bybed, high-resolution biostratigraphical subdivision of the lower, cephalopod-bearing part of the Upper Jurassic–lowermost Cretaceous section was possible (Főzy et al., 2011). Above the lowermost beds of probable Oxfordian age, a relatively complete succession of the Kimmeridgian Platynota, Strombecki, Divisum, Compsum, Cavouri and Beckeri zones was recognised, which is followed by the Tithonian Hybonotum, Darwini, Semiforme, Fallauxi, Ponti and Microcanthum zones. Representative and age-

diagnostic Late Jurassic ammonites from the Lókút section are shown in Figure 4. The belemnite fauna
allowed the recognition of four belemnites assemblages (TiBA-I to TiBA-IV) for the Tithonian part (Főzy
et al., 2011). Range charts showing the distribution of the complete ammonoid and belemnite fauna
were presented in Főzy et al. (2011).

198 From the Lókút section Grabowski et al. (2010a) published detailed calpionellid biostratigraphic 199 data. Their lowermost samples analysed were assigned to the Early Tithonian Parastomiosphaera 200 malmica Zone, whereas the overlying 3 m of the Szentivánhegy Limestone Formation belongs to the 201 Chitinoidella Zone (Fig. 2). Two samples containing Chitinoidellidae together with a few specimens of 202 Practintinnopsella sp., were placed in the Practintinnopsella Zone. Higher upsection, the remanei 203 Subzone (or A1), and the intermedia (or A2) Subzone of the Crassicollaria Zone is identified (Grabowski 204 et al., 2010a). The calpionellid assemblage of the next bed is mainly composed of Calpionella alpina 205 and Crassicollaria parvula and was therefore assigned to the Early Berriasian alpina Subzone of the 206 Calpionella Zone (Grabowski et al., 2010a). Therefore, Grabowski et al. (2010a) place the Tithonian-207 Berriasian boundary (and thus the Jurassic–Cretaceous boundary) at the Crassicollaria/Calpionella zonal boundary, following the criteria of Remane et al. (1986). In comparing the Lókút ammonite 208 209 assemblages with calpionellid data, a general agreement is demonstrated where the data overlap. For 210 example, the first appearance of chitinoidellids coincides with the base of the Microcanthum Zone (e.g., 211 Benzaggagh et al., 2010).

Within the Hárskút HK-II section, the first beds above the radiolarite provided ammonites (Fig.
5) characteristic of the latest Kimmeridgian Beckeri Zone. Higher up the complete succession from the
Hybonotum to the Ponti Zone was documented by Főzy (1990). Similarly to Lókút, the Upper Tithonian

215 seems less complete, or at least not as well documented by means of ammonites. The Durangites and 216 Microcanthum Zones could not be separated (Főzy, 1990). Although the upper part of the section 217 yielded only very poorly preserved ammonites, Horváth and Knauer (1986) recognised all of the 218 Mediterranean standard ammonite subzones, including the Jacobi, Grandis, Occitanica and Boissieri 219 Zones (Fig. 3). Horváth and Knauer (1986) also recognised the presence of minor gaps on the basis of 220 successive faunas, particularly in the Grandis Zone as well as within the Occitanica and Boissieri Zones. 221 The calpionellid assemblages identified by Horváth and Knauer (1986) at Hárskút (Fig. 3) 222 allowed the recognition of the intermedia Subzone of the Crassicollaria Zone as well as the Berriasian 223 alpina, elliptica, simplex and oblonga Subzones. Therefore, Horváth and Knauer (1986) place the 224 Tithonian/Berriasian boundary at the Crassicollaria/Calpionella zonal boundary, following the criteria 225 of Remane et al. (1986). In comparison with calpionellid data from Lókút, a general agreement is seen, 226 as the same succession of calpionellid assemblages have been identified, significantly also across the 227 Jurassic–Cretaceous boundary.

228 An integrated stratigraphic analysis of the overlying, higher part of the Lower Cretaceous 229 (Berriasian–Hauterivian), exposed in the HK-12 section, was carried out by Főzy et al. (2010). They 230 identified the Calpionella Zone at the base of the section, and a nearly complete sequence spanning the Occitanica to Boissieri ammonite zones. The overlying Lower Valanginian strata are condensed, but 231 232 yielded rich assemblages from the Pertransiens and Campylotoxus zones. Stable isotope analyses revealed a well-defined positive δ^{13} C excursion in the Valanginian strata, identified as the Weissert 233 234 event. These data are integrated with those reported in this study from the Tithonian and Berriasian of 235 Hárskút HK-II section.

236 *4.2. Calibration with magnetostratigraphy*

237 Grabowski et al. (2010a) recently published integrated magneto- and biostratigraphies of the 238 upper part of the Lókút section. The observed 6 reverse and 5 normal polarity intervals were 239 correlated with magnetochrons M21r through to M18r spanning the Jurassic-Cretaceous boundary. On 240 the basis of calpionellid biostratigraphy, Grabowski et al. (2010a), place the Jurassic–Cretaceous 241 boundary between beds no. 44 and 45, and based on reference sections (e.g., Ogg et al., 1991; Houša 242 et al., 2004) the boundary therefore appears in the middle part of normal polarity magnetosubzone 243 M19n2n (Fig. 2). Consequently, Grabowski et al. (2010a), correlate the magnetic polarity intervals from 244 the Jurassic–Cretaceous boundary down and up the section. This approach indicates that the 245 magnetozone M19r occurs entirely within the intermedia subzone (A2) in the Upper Tithonian, which is 246 consistent with other studies (e.g., Ogg et al., 1991). Likewise the M21n2n/M21r magnetosubzones fall 247 within the Fallauxi Zone, in agreement with Ogg and Hinnov (2012a). For the Hárskút section no 248 magnetostratigraphic data are available.

249 4.3 Stable carbon and oxygen isotope stratigraphy

Measurements of the carbon isotope composition of bulk carbonate yielded positive δ¹³C
values throughout the sections examined. At Lókút, values around 2.5‰ characterise the lower,
Kimmeridgian part of the section, followed by a gradual negative shift, reaching a minimum of 0.0‰
within the Lower Berriasian. Higher up-section, a return towards more positive values up to 0.7‰ is
observed. Biostratigraphic data (Vigh, 1984; Főzy et al., 2011; Grabowski et al., 2010a) together with
magnetostratigraphic data (Grabowski et al., 2010a) allow us to accurately place the low point seen in
the carbon isotope curve within these schemes. This minimum appears in the upper part of

magnetosubzone M19n2n and towards the middle of calpionellid Zone B (i.e. the alpina Subzone) (Fig. 2). The oxygen isotope data at Lókút are more variable and range from ~ 0.0 to -3.2‰. The highest δ^{18} O values occur at the base of the section. Although showing a degree of scatter, isotope values become increasingly more negative, reaching -3.2‰ towards the top of the section.

261 At Hárskút (HK II), there is overall more isotopic variability (Fig. 3). Carbon isotope values of 262 around 1.5‰ characterise the lower (Upper Kimmeridgian) part of the section, followed by a gradual negative shift, reaching a minimum of 0.9‰ within the Lower Berriasian. Following this, a return 263 264 towards more positive values is once again observed. At the top of the section, carbon isotope values 265 of 1.7‰ are recorded. The oxygen isotope data are much more variable in this section, too, and range from ~ -1.8 to 0.3‰. The most positive δ^{18} O values occur close to the base of the section and show 266 267 significant scatter; oxygen isotope values become increasingly more negative towards the top of the 268 section.

269 **5. Discussion**

270 5.1. Towards a new global δ^{13} C stack

271 In order to place the newly obtained stable isotope data from Lókút and Hárskút into a broader 272 context, we compiled 31 published Late Jurassic-Early Cretaceous carbon isotope curves, covering the 273 Oxfordian to Hauterivian interval (Fig. 6, Table 1). From the literature we gleaned those carbonate 274 carbon isotope data which have adequate stratigraphic constraints, so that magneto and/or bio-275 chronostratigraphic calibration and correlation is possible. Reference was made to biostratigraphic 276 schemes (e.g., Hoedemaeker, 1991; Remane, 1986; Wimbledon et al., 2011) that allow Tethyan–Boreal 277 correlations as well as correlations to magnetostratigraphic data. All stratigraphic data were evaluated,

so that the compilation of Gradstein et al. (2012) (e.g., Ogg and Hinnov, 2012a; 2012b) could be used.
Hence, the top of the Jurassic is the base of chron M18r with a the numerical age of 145.0 Ma. These
carbon isotope data are dominated by pelagic basinal locations, within Tethys and the Atlantic Ocean
(Table 1, Fig. 7). These successions have often been focused upon because of one or more of the
following: their completeness, the fine grained pelagic carbonate sediments suitable for isotope work,
lack of or limited diagenesis and available biostratigraphy and/or magnetostratigraphy.

Despite the differences in amplitude and offsets in absolute δ^{13} C values, there is in general a 284 good agreement of long-term δ^{13} C trends in all the sections compared, correlated on the basis of their 285 286 biostratigraphic and/or magnetostratigraphic framework. There are similar trends in our data from 287 Hungary compared with datasets from other Tethyan, Atlantic and Pacific locations (Weissert and 288 Channell, 1989; Weissert and Mohr, 1996; Katz et al., 2005; Coimbra and Olóriz, 2012; Žák et al., 2011). Given the large distances between the sites (Fig. 7) it is notable that the overall shape the δ^{13} C curves 289 are similar in some intervals. The δ^{13} C decline through the Late Jurassic and across the Jurassic– 290 Cretaceous boundary, stable values in the Berriasian and a major Early Cretaceous positive $\delta^{13}\text{C}$ 291 292 excursion, i.e. the Valanginian Weissert event, are clearly recognisable in all sections covering this interval. With respect to the isotope data from Lókút Hill (Fig. 2), the δ^{13} C decline through the Late 293 294 Jurassic is distinct.

295 Differences in absolute values and amplitude most likely reflect a number of factors including 296 local influences on the water chemistry such as nutrient levels and primary productivity, fluvial 297 influences supplying isotopically lighter and more variable DIC, sediment reworking, and the varying 298 contribution of diagenetic cements. Other differences arise potentially from low sampling resolution or

299 analysis of poorly constrained or correlated sections. Those sections that show generally high amplitude δ^{13} C shifts (e.g., La Chambotte, eastern France) are potentially affected by a combination of 300 301 sedimentology, diagenesis and the influence of varying supply of isotopically light DIC (Morales et al., 302 2013). As La Chambotte represents platform lagoonal and open-marine facies (Morales et al., 2013) high amplitude δ^{13} C variation is to be expected. Another noisy record is derived from the 303 304 stratigraphically well constrained Kimmeridgian of the Swiss Jura (Colombié et al., 2011). Although, 305 Colombié et al. (2011) showed that a long-term negative trend characterizes the entire Kimmeridgian interval studied (consistent for example with the Lókút section) the high-frequency changes in δ^{13} C 306 307 most probably result from a mix of diagenetic and local environmental effects (Colombié et al., 2011).

Few δ^{13} C records across the Jurassic–Cretaceous boundary have been derived from organic 308 309 carbon (e.g., Wortmann and Weissert, 2000; Morgans-Bell et al., 2001; Falkowski et al., 2005; Nunn et 310 al., 2009; Hammer et al., 2012). The highly detailed curve for the Kimmeridge Clay in Dorset (Morgans-311 Bell et al., 2001) ends within the Lower Tithonian, but a declining trend from the Kimmeridgian to Tithonian is evident. Likewise a declining marine $\delta^{13}C_{org}$ trend is seen in DSDP site 534A data reported 312 313 by Falkowski et al. (2005) from the Tithonian, before a pronounced positive event is seen associated with the Valanginian (Patton et al., 1984). Those Late Jurassic and Early Cretaceous $\delta^{13}C_{org}$ data derived 314 315 from woody material and charcoal (Nunn et al., 2009, 2010; Pearce et al., 2005; Gröcke et al., 2005) 316 also reveal a long-term decline in carbon-isotopes through the Late Jurassic and a positive Valanginian 317 excursion closely matching the marine carbon-isotope curves. Notably a lack of data is apparent for the 318 latest Tithonian and earliest Berriasian.

In order to separate the anomalous, the regional and the global trends, an average $\delta^{13}C_{carbonate}$ 319 320 stack was developed (Fig. 8), based on the sections compared and presented here (Fig. 6). The new global δ^{13} C stack (Fig. 8) is used to visualise and identify those globally synchronous shifts in δ^{13} C that 321 can be applied for global correlation. The δ^{13} C stack does not include any estimated or calculated 322 323 average, but instead shows all data of the curves with a grey envelope indicating the range of absolute 324 values. Using the available magnetostratigraphy and biostratigraphy as tie-points for alignment of the 325 records, the curves were plotted onto the same scale, adjusted to the data from DSDP 534A of 326 Tremolada et al. (2006), Bornemann and Mutterlose (2008) and Katz et al. (2005) in order to visualize 327 similarities and differences. Some error may be incorporated here, particularly for shorter isotope 328 records, even when combined biostratigraphy is available, as for example magnetostratigraphic 329 resolution may be not fine enough to allow for multiple tie-points or variable sediment accumulation 330 rates need to be estimated. Notably the data from Lókút and Hárskút do not fall outside of the stack. 331 These data (from Lókút and Hárskút) are from a pelagic settings, consistently seen elsewhere. Although 332 carbon isotope data from shallower marine settings (e.g., Colombié et al., 2011) also see similar trends 333 attesting to the robustness of the carbon isotopic signal.

The stack clearly shows a decline in δ¹³C throughout the Late Jurassic–Early Cretaceous,
reaching a minimum in the Early Cretaceous in magnetochron M12, near the base of the Campylotoxus
Zone (see also Weissert et al., 1998). The positive excursion in the Valanginian, (the Weissert event), is
plainly evident. Our data from Hárskút (Fig. 3) clearly reveals the positive excursion in the Valanginian
(see Főzy et al., 2010). The width of the grey envelope partly reflects the sampling density within the
data set, and additional data could certainly modify the picture. Nevertheless, for intervals with similar
data coverage, the outline of the envelope and its width may help evaluate the relative importance of

and reproducibility of possible global isotopic trends. The well constrained Valanginian event contrasts with much of the earlier record, in particular the Early Tithonian, where the width of the grey envelope is larger, potentially reflecting local influences on the water chemistry, sediment reworking, diagenesis combined with stratigraphic uncertainty. Hence, aside from the well-defined Valanginian event, chemostratigraphic correlation using the δ^{13} C record from the Late Jurassic–earliest Cretaceous is challenging due to relatively stable δ^{13} C values, a broad envelope, compounded by a slope too slight.

In comparison, the composite Late Jurassic–Early Cretaceous δ^{13} C curve in GTS 2012 shows little 347 348 more than the Valanginian Weissert event and slightly elevated values in the Late Tithonian (Ogg and 349 Hinnov, 2012a; 2012b). A largely unvarying carbon isotope profile through this interval within the GTS 350 2012 appears at odds with the records summarized herein. The generalized curves in GTS 2012 were 351 derived from Jenkyns et al. (2002) for the Late Jurassic and Föllmi et al. (2006) for the Early Cretaceous, 352 the latter in turn relies solely on data reported by Emmanuel and Renard (1993) for the Berriasian and 353 earliest Valanginian, and Hennig et al. (1999) for most of the Valanginian and earliest Hauterivian. In 354 comparison, our compilation includes numerous other sources for a more reliable composite curve. 355 The lack of variation through the Jurassic–Cretaceous boundary is therefore not particularly useful in 356 adding to the characterisation of the boundary. The low point and return to more positive values seen 357 in our data from Lókút and Hárskút appearing in the upper part of magnetosubzone M19n2n and towards the middle of calpionellid Zone B (the Alpina Subzone) (Figs. 2, 3) is not resolved in the $\delta^{13}C$ 358 stack. Likewise, the positive Boreal δ^{13} C shift immediately above the Jurassic–Cretaceous boundary 359 correlated to Tethyan records recorded by Dzyuba et al. (2013) is also not resolvable in the δ^{13} C stack. 360

361 5.2. Comparison and interpretation of δ^{13} C trends

362 Our newly obtained stable isotope data from Lókút and Hárskút (Figs. 2, 3), taken together with the δ^{13} C stack, as noted above, shows a shifts towards negative values throughout the Late Jurassic– 363 364 Early Cretaceous, reaching a minimum in the Early Cretaceous. Mechanisms proposed to cause global 365 shifts towards negative carbon isotope values include changes in productivity and organic carbon burial, increases in volcanic activity and episodic rapid methane release from gas hydrates contained in 366 marine sediments. Large negative excursions in marine carbonate δ^{13} C are often associated with 367 368 period boundaries and mass extinctions (Kump, 1991). Given the typically abrupt nature of isotope excursions related to inferred methane fluxes (e.g., Menegatti et al., 1998), this mechanism appears 369 370 unlikely in the studied interval. Changes in carbon isotopes may, however, be related to ecological 371 crises culminating in the disappearance of macro- and microfaunas. The Jurassic–Cretaceous boundary 372 was earlier considered to be one of the major mass extinction events during the Phanerozoic (Sepkoski 373 and Raup, 1986) with groups such as corals, brachiopods, bivalves, ammonites and fish all affected. As 374 noted above, subsequent work has downgraded the boundary to a minor extinction event at most 375 (Alroy, 2008). However, some recent studies have found evidence for a real diversity trough within 376 terrestrial dinosaurs and marine reptiles (e.g., Mannion et al., 2011). The Jurassic-Cretaceous 377 boundary interval is also characterized by significantly elevated extinction and origination rates in 378 calcareous nannoplankton (Roth, 1987; Bown, et al., 2004; Bown, 2005; Tremolada, et al., 2006). 379 Tremolada et al. (2006) document high abundances of late middle Tithonian oligotrophic taxa such as Nannoconus spp. and Conusphaera spp. correlating with low δ^{13} C values. Oligotrophic conditions in the 380 Tethyan seaway have been linked to drier climates and a sea level low during the latest Jurassic (e.g., 381 382 Hallam et al., 1991; Abbink et al., 2001; Ruffell et al., 2002b; Schnyder et al., 2006)(Fig. 8), reduced 383 runoff and reduced nutrient fluxes to the oceans, lowering the fertility of surface waters (e.g., Weissert

384 and Channell, 1989). Hence, the sea-level fall during the latest Jurassic to early Berriasian (e.g., Hag, 2014) may in part correlate with aridity, lower inputs of nutrients and the gradual negative δ^{13} C shift. A 385 kaolinite minimum is known from all over Europe and associated with a major Late Jurassic "dry event" 386 387 (e.g., Hallam et al. 1991, Abbink et al. 2001; Rameil, 2005; Schnyder et al., 2006). Rameil (2005), inferred from cyclostratigraphy, the duration of the dry phase, as defined on the Jura platform, to be 388 389 8.4 Ma (Fig. 8). However, both field observations and sedimentary log interpretation, suggest that the 390 drier phase can be subdivided into a dry phase sensu stricto lasting about 2.8 Ma, followed by a longer transition phase (Rameil, 2005). However, the decline in δ^{13} C seen is not a short interval associated just 391 392 with the Jurassic-Cretaceous boundary but one that begins in Oxfordian times and continues into the 393 Early Valanginian. The change to once again more positive carbon isotopes in the Early Cretaceous 394 Tethyan seaway in the Valanginian is therefore interpreted as a change to increasingly nutrient-rich 395 conditions and enhanced carbon cycling (Weissert and Channell, 1989). The similarity of the $\delta^{13}C_{org}$ trends derived from woody material and charcoal, noted above, to the marine carbonate δ^{13} C stack 396 397 clearly supports the notion that the surface ocean and atmosphere behaved as coupled reservoirs at 398 this time.

In contrast, the Sr isotope record for this interval (Fig. 8) (e.g., Jones et al., 1994; McArthur et al., 2004; Bodin et al., 2009; Wierzbowski et al., 2012) shows a trend towards more radiogenic values from a long-term low at the Callovian-Oxfordian boundary to a peak in the Barremian. This variation in Srisotopes possibly reflects a change in the balance of flux from relatively non-radiogenic Sr derived from mid-ocean ridge hydrothermal activity to relatively radiogenic Sr derived from continental weathering (including changes in both total riverine flux and the isotopic composition of the flux). The ⁸⁷Sr/⁸⁶Sr low in the middle Oxfordian is, however, not seen as correlatable with an obvious pulse of ocean crust

406 production (e.g., Rowley, 2002) or with the formation of a large igneous province. Wierzbowski et al., 407 (2012) do call upon fast oceanic crust spreading and opening of new ocean basins during the 408 Bathonian-Callovian-Oxfordian related to the breakup of Gondwana to account for the Callovian-Oxfordian minimum ⁸⁷Sr/⁸⁶Sr ratios observed (Fig. 8). Indeed, the data Cogné and Humler (2006) do 409 410 possibly point to higher overall seafloor spreading rates for the Late Jurassic. Notably, the Paraná-411 Etendeka large igneous province is Valanginian-Hauterivian in age with volcanic activity starting at 412 134.6 ± 0.6 Ma or at 134.3 ± 0.8 Ma (Thiede and Vasconcelos, 2010; Janasi et al., 2011) coincident with 413 the onset of the Weissert Event (Martinez et al., 2015). The Sr-isotope data at this time (Fig. 8) does 414 not show any inflections in the curve (McArthur et al., 2001). Indeed, investigations regarding the 415 spreading and production rates of oceanic ridges (e.g., Rowley, 2002; Cogné and Humler, 2006) show 416 fairly constant production rates of oceanic crust during the Cretaceous. If rates of ocean floor 417 production do not change substantially, then hydrothermal Sr fluxes should also be relatively invariant 418 over long time scales. The implication is that the source of Sr from continental weathering is likely to be a major factor governing the evolution of marine ⁸⁷Sr/⁸⁶Sr. Indeed, phosphorus flux rates (Föllmi, 419 420 1995) which are dependent on continental weathering rates, show a decrease from a high values in the 421 Late Jurassic, to a low through the Jurassic–Cretaceous boundary, and a subsequent increase through 422 into Hauterivian times. Likewise high sediment fluxes to the central North Atlantic Ocean during the latest Jurassic to Early Cretaceous (post the Late Jurassic "dry event") are also observed (e.g., Thiede 423 424 and Ehrmann, 1986). Episodes of increased hydrothermal activity are, however, not necessarily directly 425 related to rates of ocean-crust production and phenomena as ridge jumps or changes in ridge 426 orientation may substantially increase hydrothermal venting by additional fracturing of oceanic crust

427 and consequent greater access of seawater to hotter, fresher material at the ridge axis (Jones and428 Jenkyns, 2001).

429 The relatively short-lived arid episode (or dry phase sensu stricto, Rameil, 2005) and possible linked short-term sea-level fall and rise (e.g., Haq, 2014) appears not to be reflected in the Sr-isotope 430 431 curve. The short duration of arid conditions and presumed reduction in continental weathering and change in ⁸⁷Sr/⁸⁶Sr ratios, is unlikely to be resolvable over such a short timescale as inputs and outputs 432 433 of Sr are possibly buffered too well by the large oceanic reservoir of Sr (Richter and Turekian, 1993). 434 Likewise, short-term ocean fertilisation, productivity and carbon burial events, appear also not to be 435 reflected in either the Sr-isotope or the carbon isotope curves. For example, deposition of significant 436 petroleum source rocks of Late Jurassic and Early Cretaceous age, known from Arabian-Iranian region, 437 West Siberia, the North Sea, Greenland Sea (Klemme and Ulmishek, 1991) and Mexico (the Casita Fm, Adatte et al., 1996) are evidently not expressed within the δ^{13} C record (Weissert and Mohr, 1996; Price 438 439 and Rogov, 2009; Föllmi, 2012). Paradoxically, evidence for widespread organic matter deposition in 440 the marine environment during the Valanginian is rather scarce, yet the Valanginian does show a 441 pronounced positive carbon isotope excursion (e.g., Lini et al., 1992; Channell et al., 1993; Bersezio et 442 al., 2002; Erba et al., 2004; Duchamp–Alphonse et al., 2007; Sprovieri et al., 2006; Littler et al., 2011, 443 Figs. 6, 7). Hence simple models of transient positive carbon isotope excursions associated with burial and sequestration of isotopically light marine carbon (¹²C) may not be fully applicable for this interval. 444 445 Likewise, given the evolutionary rise of the modern plankton groups through Late Jurassic–Early Cretaceous time one would anticipate an overall increase in δ^{13} C values in marine carbonates (e.g., 446 447 Falkowski et al., 2004).

448 The type of carbon burial (organic vs. carbonate carbon), accumulation rates, and areal 449 distribution of facies may instead be important factors with respect to changes in the carbon isotopic 450 signature of the Jurassic and Cretaceous oceans (Weissert, 2011; Föllmi, 2012). Mass balance models 451 for the Cretaceous (Locklair et al., 2011) suggest that elevated rates of carbonate burial (burying relatively isotopically heavy carbon) could have dampened changes in $\delta^{13}C_{DIC}$ expected from elevated 452 453 organic carbon burial rates (Weissert and Mohr, 1996; Föllmi, 2012). Indeed through the Late Jurassic-454 Early Cretaceous transition elevated rates of carbonate burial and preservation are observed (e.g., 455 Mackenzie and Morse, 1992; Berner and Mackenzie, 2011). For example, during the Late Jurassic 456 carbonate sedimentation became dominant over wide parts of the northern Tethys (Rais et al., 2007), 457 with the expansion and development of new reef sites (Leinfelder et al., 2002; Cecca et al., 2005). 458 Likewise, the surge of diversification of calcareous nannoplankton at the Jurassic-Cretaceous boundary 459 interval involved the evolution of three large and heavily calcified genera that would have greatly 460 increased the transfer and burial efficiency of carbonate (Tremolada et al., 2006). In terms of the areal 461 distribution, widespread biogenic deep-water carbonate sedimentation (Zeebe and Westbroek, 2003) 462 within a well-mixed ocean at this time would provide means to maintain a steady state between 463 carbonate-mineral burial (Locklair et al., 2011) and weathering, buffering changes in carbon cycling. In 464 contrast, earlier ocean systems (before pelagic calcifiers became increasingly abundant) were 465 dominated by biogenic shallow-water carbonate precipitation perhaps explaining why in the 466 Palaeozoic, Triassic and Early Jurassic carbon isotope anomalies (e.g., Payne et al., 2004; Hesselbo et al., 467 2007) have amplitudes of up to 6 ‰ or more.

468 Certainly organic carbon burial occurred during the Late Jurassic and Early Cretaceous, but
469 within marginal seas (e.g., Wignall and Hallam, 1991; Hantzpergue et al., 1998; Price and Rogov, 2009).

470 Deposition in marginal seas would have been initiated as eustatic sea-level peaked in the 471 Kimmeridgian–early Tithonian, followed by a lowstand across the Jurassic-Cretaceous boundary, 472 followed by a slight rise, and fall again in the Valanginian–Hauterivian (Hallam, 2001; Haq, 2014) (Fig. 8). 473 However, carbon burial within marginal seas evidently did not impact significantly on the global ocean 474 chemistry, due to the possibly relatively small size of marginal seas compared to the global ocean and 475 through efficient ocean mixing. Indeed, the Late Jurassic was a time of progressive fragmentation of 476 Pangaea (Dercourt et al., 1994) and new oceanic gateways were formed and in particular, the opening 477 of the Hispanic Corridor, connecting the Pacific to the Atlantic Ocean (Ziegler, 1988). Although the first 478 shallow-water connection between the Tethys/Atlantic Ocean and the Pacific Ocean is dated as 479 Pliensbachian–Toarcian (Aberhan, 2001) the continuous deepening of the Hispanic Corridor associated 480 with a first order sea-level rise, allowed significant water mass exchange between the two basins 481 during the Late Jurassic (Riccardi, 1991; Stille et al., 1996; Hallam, 2001, Fig. 7). Studies on reef 482 development (Leinfelder et al., 2002) for example confirm the establishment of a first true seaway 483 around the Callovian–Oxfordian boundary.

484 It has also been suggested that a decrease in organic carbon burial on the continent (Föllmi, 2012) may also have played a role in buffering the δ^{13} C record. The dominance of arid conditions on 485 486 the continent (e.g., Hallam et al., 1991; Schnyder et al., 2006) may have precluded major organic 487 carbon production and preservation. Indeed relatively large amounts of coal deposition in the earlier 488 part of the Jurassic is followed by a decline through the Jurassic–Cretaceous boundary (e.g., Bluth and 489 Kump 1991). Conversely, Westerman et al., (2010) and Kujau et al., (2012) for example, call for 490 continental organic carbon burial (i.e. coal deposition) to explain the Valanginian carbon cycle 491 perturbation. If, as noted above, the surface ocean and atmosphere behaved as coupled reservoirs at

this time, this would not preclude continental organic carbon burial as a viable means to affect carboncycling.

494 5.3. Oxygen isotopes and palaeoenvironmental change

The preservation of primary δ^{13} C values during carbonate diagenesis is quite typical, and is 495 496 likely due to the buffering effect of carbonate carbon on the diagenetic system, as this is the largest 497 carbon reservoir (e.g., Scholle and Arthur, 1980). Fluid-rock interactions during diagenesis, however, commonly result in a change in oxygen isotope ratios leading to relatively light $\delta^{18}O_{carb}$ values (Hudson, 498 499 1977). Hence, with respect to the oxygen isotope data, a diagenetic overprint affecting the samples 500 analysed and results cannot be excluded. Nevertheless, the oxygen isotope data from both sites in 501 Hungary do show a similar pattern. Furthermore, given that the isotopic trends are the same as that 502 seen from diagenetically screened belemnites from Lókút (Főzy et al., 2011) we are confident that the trends do reflect a primary signal, independent of diagenesis. Increasingly negative δ^{18} O values are 503 504 often correlated with elevated temperatures in environmental settings where continental ice volume is 505 at a minimum and evaporation or freshwater inputs are minor factors. Similar trends have been 506 observed elsewhere (e.g., Tremolada et al., 2006; Price and Rogov, 2009; Grabowski et al., 2010b), but 507 not universally as other studies found opposite trends (e.g., Emmanuel and Renard, 1993; Padden et al., 508 2002). Larger datasets through the Late Jurassic and into the Cretaceous, based on the isotopic 509 composition of fossil belemnites and brachiopods (e.g., Veizer, et al., 1999; Gröcke et al., 2003; 510 Wierzbowski, 2004; McArthur et al., 2007; Riboulleau et al., 1998; Bodin et al., 2009, 2015; Price and 511 Rogov 2009; Dera et al., 2011; Alberti et al., 2012; Price et al., 2000; 2011; 2013; Meissner et al., 2015), 512 also show a similar trends (Fig. 8). The data compiled in Figure 7 are derived from data from a range of

513 low and mid Tethyan palaeolatitudes and should, therefore, be less affected by regional (e.g., salinity-514 driven) isotopic variation. Nevertheless trends can also be linked to other factors, for example variation 515 in terrestrial water bodies and sea level variations (e.g., Föllmi 2012). If, interpreted in terms of 516 temperature, the data point to Oxfordian warming and a further peak in the middle Tithonian 517 separated by a temperature plateau. Oxfordian warming and a temperature peak in the middle 518 Tithonian is consistent with TEX₈₆ temperature data of Jenkyns et al. (2012). A possible Late Berriasian cooling event is seen (a shift to more positive δ^{18} O values), followed by cooling through the Valanginian. 519 The Hauterivian shows a return to warmer conditions. Shorter term trends through the Jurassic-520 521 Cretaceous boundary interval are less clear as belemnite oxygen isotope data in this compilation are 522 fewer and the 95% confidence interval is greater. The scatter in values here means trends must be 523 interpreted with caution. Notably, despite some considerable change in oxygen isotopes through the Late Jurassic and Early Cretaceous, any recognisable correlation with the δ^{13} C curve is lacking. For 524 525 example, during the pronounced Valanginian shift to more positive carbon isotope values (the 526 Weissert event), temperatures continue to fall, but as part of a longer term trend. The TEX₈₆ data of 527 Littler et al. (2011), also showed little recognisable correlation of temperature with the δ^{13} C curve for 528 the Valanginian.

529 Of note is that the transition from arid to humid climates through the Late Jurassic and Early 530 Cretaceous may have been associated with the net transfer of water to the continent owing to the infill 531 of dried-out groundwater reservoirs in internally drained inland basins (Föllmi, 2012) and thereby 532 affecting the oxygen isotope of seawater. The prominent Late Berriasian shift to more positive δ^{18} O 533 values, could conceivably be related to the observed arid to humid climate transition, short-term sea-534 level fall (Fig. 8) and a net transfer of water towards the continent (e.g., Föllmi, 2012). Recently,

Wendler et al. (2016) also demonstrated that aquifer eustasy represents a viable alternative to explain
sea level fluctuations and consequently variation in the oxygen isotope of seawater.

537

538 6. Conclusions

The δ^{13} C data from Hungary are consistent with other isotope stratigraphies and indicate that 539 540 the Lókút and Hárskút sections record global events, as reflected in a stack of 30 individual carbon 541 isotope curves. Aside from the well-defined Valanginian event, chemostratigraphic correlation using the δ^{13} C record is challenging due to relatively stable δ^{13} C values showing a slope which is too slight. 542 543 The Berriasian minimum and the return to more positive values seen in our data from Lókút and Hárskút is not resolved in the global δ^{13} C stack. Oxygen isotopes point to warming through the Late 544 545 Jurassic interval, broadly in agreement with larger datasets through the Jurassic and Cretaceous, based 546 on the isotopic composition of fossil belemnites and brachiopods. This latter dataset point to a 547 stepwise cooling through the Valanginian. Notably, despite large changes in temperature through the Late Jurassic and Early Cretaceous any recognisable correlation with the δ^{13} C curve is lacking. 548

549 The Late Jurassic δ^{13} C decline has been explained by increasingly oligotrophic conditions in the 550 Tethyan seaway (e.g., Weissert and Channell, 1989), whilst more positive carbon isotope values in the 551 Valanginian are ascribed to increasingly nutrient-rich conditions and enhanced carbon cycling and 552 burial. However, the Jurassic–Cretaceous boundary interval is also characterized by elevated rates of 553 calcareous nannoplankton turnover and enhanced organic carbon deposition that it is not expressed 554 within the δ^{13} C record. The type of carbon burial (organic vs carbonate carbon), accumulation rates, 555 and areal distribution of facies may be the key, whereby elevated rates of carbonate burial (including

556 large and heavily calcified calcareous nannoplankton, Tremolada et al., 2006) could have buffered changes in $\delta^{13}C_{DIC}$ expected from elevated weathering and increased organic carbon burial rates 557 558 (particularly in marginal seas). We envisage also well-mixed parts of the ocean, perhaps as a result of connections established between the Tethys and Central Atlantic, and the full opening of the Hispanic 559 Corridor effectively linking the Atlantic and Pacific Oceans. This scenario reconciles the apparently 560 561 contradictory trends in carbon and strontium isotopes. The strontium isotope data through the 562 Jurassic-Cretaceous interval points to a longer term intensification of weathering (and a decreasing 563 contribution of non-radiogenic hydrothermal Sr), which would have presumably increased the transfer of elements such as silica and phosphorus from the continents to the oceans (e.g., Föllmi, 1995) 564 565 resulting in increased productivity. An increased transfer of elements is consistent with the observation 566 of high sediment fluxes to the central North Atlantic Ocean during the latest Jurassic to Early 567 Cretaceous (post the Late Jurassic "dry event"). However, there is a background evolutionary rise of 568 the modern plankton groups, notably organic-walled phytoplankton (i.e. dinoflagellates) and 569 calcareous nannoplankton (coccolithophores) in Late Jurassic-Early Cretaceous time (Falkowski et al., 570 2004). Therefore the effectiveness of the biological carbon pump and export of carbonate carbon is 571 expected to gradually increase. The carbon isotope trend is thus all the more remarkable, as its forcing counterbalances the effects of the "Mesozoic plankton revolution". 572

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Fig. 1. Location and palaeogeographic setting of the studied sections. A: Location of Lókút Hill and
Hárskút in the Bakony Hills of the Transdanubian Range in western Hungary. B: Palaeogeographic
setting of the Transdanubian Range (TR) and neighbouring units within a reconstructed Tithonian (Late
Jurassic) western Tethyan palaeogeography (after Csontos and Vörös, 2004).



Fig. 2. Integrated biostratigraphy, magnetostratigraphy and carbon and oxygen isotope stratigraphy
from the Lókút section. The measured log and samples are referenced using the bed numbers of Vigh
(1984). Ammonite zones for the Kimmeridgian and Tithonian follow the zonation scheme by Enay and
Geyssant (1975) and Geyssant (1997). LRF = Lókút Radiolarite Formation. Pm. = *Parastomiosphaera malmica* Zone. Belemnite assemblages are from Főzy et al. (2011).



- 1019 Fig. 3. Integrated stratigraphy of the Hárskút HK-II section showing the ammonite and calpionellid
- 1020 biostratigraphy (from Horváth and Knauer, 1986, Főzy, 1990) and carbon and oxygen isotope curves.
- 1021 Abbreviations: Kim. = Kimmeridgian, Occit. = Occitanica Zone; Boiss. = Boissieri Zone.



- Fig. 4. Representative and age-diagnostic Late Jurassic ammonites from the Lókút section. Inventory
 numbers of the Department of Paleontology and Geology of the Hungarian Natural History Museum
 are prefixed by INV. All figures are natural size.
- 1027 1. *Haploceras verruciferum* (Zittel, 1869), INV.2014.76, Bed LH 122, Semiforme Zone.
- 1028 2, 3. *Simoceras biruncinatum* (Zittel, 1869), INV.2014.75, Bed LH 133, Fallauxi Zone.
- 1029 4, 5. *Trapanesites adelus* (Gemmellaro, 1872), INV.2014.77, Bed LH 110-111, Compsum Zone (?).





1032 Fig. 5. Representative and age-diagnostic Late Jurassic ammonites from the Hárskút (HK-II) section.

1033 Inventory numbers of the Hungarian Geological and Geophysical Institute are prefixed by J. All figures1034 are natural size.

- 1035 1. Haploceras verruciferum (Zittel, 1869), J 10923, Bed 60, Semiforme Zone.
- 1036 2. Semiformiceras fallauxi (Oppel, 1865), J 10875, Bed 54, Fallauxi Zone.
- 1037 3, 4. Haploceras carachtheis (Zeuschner, 1846), J 10908, Bed 49, Fallauxi Zone.
- 1038 5. Semiformiceras semiforme (Oppel, 1865), J 10870, Bed 59, Semiforme Zone.
- 1039 6. *Simoceras admirandum* (Zittel, 1869), J 10965, Bed 48, Fallauxi Zone.
- 1040 7. Semiformiceras birkenmajeri Kutek & Wierzbowski, 1986, J 10367, Bed 62, Darwini Zone.
- 1041 8, 9. *Ptychophylloceras ptychoicum* (Quenstedt, 1847), J 10683, Bed 44, Fallauxi Zone.
- 1042 10, 11. Anaspidoceras neoburgense (Oppel, 1863), J 10371, Bed 64, Darwini Zone.
- 1043 12. Haploceras elimatum (Oppel, 1865), J 10600, Bed 51, Fallauxi Zone.
- 1044 13, 14. Lytogyroceras subbeticum Olóriz, 1978, J 10976, Bed 42, Ponti Zone.
- 1045 15, 16. *Discosphictoides* cf. *rhodaniforme* Olóriz, 1978, J 10363, Bed 59, Semiforme Zone.



Fig. 6. Summary of global carbonate δ^{13} C correlations for the Late Jurassic-Early Cretaceous. Global 1048 correlation of δ^{13} C data is based on 31 published Late Jurassic-Early Cretaceous records from the 1049 Boreal Realm, Atlantic Ocean and Tethys. The $\delta^{13}C_{carb}$ data are from bulk sediments except for the 1050 1051 Subpolar Urals and North Siberia composite data derived from belemnites from Dzyuba et al. (2013) 1052 and Price and Mutterlose (2004). The data from Cardador, Southern Spain (Coimbra et al., 2009) and 1053 Montclus, Vocontian Basin (Morales et al., 2013) is shown as a 3-point moving average. For each 1054 location a number is provided which corresponds to the section number in Table 1. Numeric ages (a 1055 linear scale), magnetostratigraphy and Tethyan Ammonite Zones are from GTS 2012 (Ogg and Hinnov, 1056 2012a; 2012b).



Fig. 7. Global and regional (inset) Late Jurassic palaeogeographic reconstruction (modified from Blakey, 1059 2015) showing the distribution of localities used to generate of the δ^{13} C stack. For each location a 1060 number is provided which corresponds to the section number in Table 1. Location H = location of 1061 Hungarian sites.



Fig. 8. A global δ^{13} C stack calibrated with magnetostratigraphy. The δ^{13} C_{carb} data are from bulk 1065 1066 sediments as shown in detail in Fig. 6, excluding data from the Subpolar Urals and North Siberia 1067 (Dzyuba et al., 2013; Price and Mutterlose, 2004) and excluding the data from La Chambotte (Morales 1068 et al., 2013) and the Kimmeridgian data from the Swiss Jura (Colombié et al., 2011). Belemnite oxygen 1069 isotope data from sources cited within the text; the Sr isotope record from Jones et al. (1994); 1070 McArthur et al., (2004) and Bodin et al., (2009); humid and arid phases from Hallam et al. (1991) and 1071 Ruffell et al. (2002b) with Jurassic "dry event" transition phase (from Rameil, 2005) and eustatic sea-1072 level curve from Haq (2014). Numeric ages, magnetostratigraphy and Tethyan Ammonite Zones are 1073 from GTS 2012 (Ogg and Hinnov, 2012a; 2012b).



Table 1 Numbered location, stratigraphical range, magneto and/or bio-chronostratigraphic control,

- 1076 lithology and source reference for published Late Jurassic-Early Cretaceous carbon isotope curves.

 Location		Stratigraphical span	Stratigraphic control	Lithology	Reference
 1.	Długa Valley, Poland	Late Oxfordian– Early Tithonian	radiolaria and calcareous dinoflagellates	nodular limestones and radiolarites	Jach et al., (2014)
2.	ODP 1149B, Pacific Ocean	Valanginian–Early Hauterivian	radiolaria and calcareous dinoflagellates	radiolarian chert and nannofossil chalk and marls	Erba et al., (2004)
3.	ODP Site 603, Atlantic Ocean	Late Berrisian- Early Hauterivian	nannofossils and magnetostratigraphy	nannofossil limestone and mudstones	Littler et al., (2011)
4.	Terminilletto, central Italy	Late Oxfordian- Late Tithonian	radiolaria	limestone and cherts	Bartolini et al., (1999)
5.	Gresten Klippenbelt, Austria	Tithonian–Early Berriasian	ammonites, calpionellids, nannofossils, magnetostratigraphy	pelagic marl- limestone cycles	Lukeneder et al., (2010)
6.	Cuber, Mallorca, Spain	Late Oxfordian– Early Berriasian	ammonites	bedded and nodular marly limestones	Coimbra and Olóriz, (2012)
7.	Berrias, France	Berriasian	ammonites calpionellids	pelagic limestones	Emmanuel and Renard, (1993)
8.	Subpolar Urals/North Sibera	LateTithonian– Late Valanginian	ammonites, magnetostratigraphy	belemnites	Dzyuba et al., (2013); Price & Mutterlose, (2004)
9.	Montsalvens, Switzerland	Late Oxfordian– Tithonian	ammonites	nodular limestones with chert	Padden et al., (2002)
10.	Gemmi, Switzerland	Late Oxfordian– Tithonian	ammonites	nodular limestones with chert	Padden et al., (2002)
11.	Gorges du Pichoux, Swiss Jura	Kimmeridgian– Early Tithonian	ammonites	lime mudstones	Colombié et al., (2011)

12.	Capriolo, Italy	Berriasian– Hauterivian	nannofossils, magnetostratigraphy	marly limestones with chert	Lini et al., (1992)
13.	Cardador, Betic Cordillera, Spain	Oxfordian– Tithonian	ammonites	bedded and nodular limestones	Coimbra et al., (2009)
14.	Cala Fornells, Mallorca, Spain	Oxfordian–Early Tithonian	ammonites	bedded and nodular marly limestones	Coimbra & Olóriz, (2012)
15.	Breggia, Switzerland	Berriasian– Hauterivian	nannofossils, magnetostratigraphy	pelagic limestone with chert	Bersezio,et al., (2002)
16.	Angles, France	Late Berriasian– Early Hauterivian	ammonites, nannofossils, calpionellids	marl–limestone alternations	Duchamp– Alphonse et al., (2007)
17.	Hlboča Slovakia	Tithonian–Early Valanginian	calpionellids, magnetostratigraphy	nodular limestone, cherty limestones	Grabowski et al., (2010b)
18.	DSDP 105, Atlantic Ocean	Tithonian– Valanginian	nannofossils	limestone and claystones	Tremolada et al. (2006); Brenneke, (1978)
19.	DSDP 534A, Atlantic Ocean	Early Tithonian– Hauterivian	nannofossils, magnetostratigraphy	limestone and claystones	Tremolada et al. (2006); Katz et al. (2005)
20.	Frisoni, Italy	Late Kimmeridgian– Early Berriasian	calpionellids, magnetostratigraphy	nodular limestone and thin bedded limestones	Weissert and Channell (1989)
21.	Brodno, Western Carpathians, Czech Republic	Tithonian–Early Berriasian	Calpionellids, nannofossils, magnetostratigraphy	pelagic limestones	Michalik et al., (2009)
22.	Xausa, Italy	Late Kimmeridgian– Berriasian	calpionellids, magnetostratigraphy	nodular limestone and thin bedded limestones	Weissert and Channell (1989)
23.	Valle del Mis, Italy	Tithonian–Early Berriasian	calpionellids, magnetostratigraphy	nodular marly limestone and thin bedded limestones	Weissert and Channell (1989)
24.	Guppen – Heuberge, Switzerland	Late Oxfordian– Early Berriasian	ammonites, calpionellids	nodular and micritic limestones	Weissert and Mohr, (1996)
25.	Bucegi	Early Valanginian–	ammonites,	pelagic limestones	Barbu,

Mountains, Romania	Early Hauterivian	nannofossils		(2014)
26. La Chambotte,	Early Berriasian–	foraminifera,	shallow-water	Morales et
France	Early Valanginian	calpionellids,	limestones	al., (2013)
27. Montclus,	Early Berriasian–	ammonites,	hemipelagic marl-	Morales et
France	Early Valanginian	nannofossils	limestones	al., (2013)
28. Puerto Escano, Spain	Tithonian–Early Berriasian	calpionellids, ammonites, magnetostratigraphy	limestones and nodular limestones	Zak et al., (2011)
29. San Lucas,	Berriasian–	calpionellids	Marls and	Adatte et al.,
Mexico	Valanginian		limestones	(2001)
30. Umbria, Italy	Berriasian– Hauterivian	calpionellids, magnetostratigraphy	limestones	Sprovieri et al., (2006)
31. Pusiano,	Berriasian–	Nannofossils,	Pelagic limestones	Channell et
Northern Italy	Hauterivian	magnetostratigraphy		al., (1993)