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4 **Carbon cycle history through the Jurassic-Cretaceous boundary:**
5 **a new global $\delta^{13}\text{C}$ stack**

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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
24 isotope records from the Pacific, Tethyan, Atlantic, and Boreal realms, to produce a new global $\delta^{13}\text{C}$
25 stack for the Late Oxfordian through Early Hauterivian interval. Our new data from Hungary is
26 consistent with the global $\delta^{13}\text{C}$ stack. The stack reveals a steady but slow decrease in carbon isotope
27 values until the Early Valanginian. In comparison, the Late Jurassic–Early Cretaceous $\delta^{13}\text{C}$ curve in GTS
28 2012 shows no slope and little variation. Aside from the well-defined Valanginian positive excursion,
29 chemostratigraphic correlation during the Jurassic–Cretaceous boundary interval is difficult, due
30 to relatively stable $\delta^{13}\text{C}$ values, compounded by a slope which is too slight. There is no clear isotopic
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 conditions and lessened primary production. However, this contradicts the reported
34 increase in $^{87}\text{Sr}/^{86}\text{Sr}$ ratios suggesting intensification of weathering (and a decreasing contribution of
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
46 of the Mesozoic greenhouse world (Föllmi, 2012; Price et al., 2013). This is, in part, due to the lack of
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
56 background of several calpionellid diversification events (Remane, 1986; Michalík et al., 2009) and an
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 jacobii Zone.
83 Hence, Wimbledon et al. (2011) , tentatively suggest that several markers have the potential to help
84 define any putative Jurassic-Cretaceous boundary.

85 Carbon isotope stratigraphy is useful both to help understand past global environmental and
86 biotic change that affected carbon cycle, and as a correlation tool. For example, the GSSP for the base
87 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
92 Late Jurassic carbonate carbon isotopic composition shifts from $\delta^{13}\text{C}$ values of around 2.5‰ in the
93 Kimmeridgian to values near 1.0‰ in the Late Tithonian–Early Berriasian. A change to lower $\delta^{13}\text{C}$
94 values was identified to occur within Magnetozones M18–M17 and within the B/C Calpionellid Zone
95 (Weissert and Channell, 1989). The low $\delta^{13}\text{C}$ values of the earliest Cretaceous contrast with the more
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
107 negligible variation associated with the boundary. Dzyuba et al. (2013) reported a positive $\delta^{13}\text{C}$ shift
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; Fózy, 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–
116 Cretaceous boundary. To this end, we also developed a new global stack of carbonate $\delta^{13}\text{C}$ curves for
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
123 environmental events is also made to help elucidate controls on the global $\delta^{13}\text{C}$ stack.

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)
155 and Galácz (1975) established the presence of repeated gaps due to non-deposition. Within the "horst
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 "Prédikálószték" ("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**

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

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

179 For this study, stable isotope analyses of 165 bulk carbonate samples were taken from sections
180 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 $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$
185 was better than $\pm 0.1\%$, based upon duplicate sample analyses.

186 **4. Results**

187 *4.1. Biostratigraphy*

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

194 diagnostic Late Jurassic ammonites from the Lókút section are shown in Figure 4. The belemnite fauna
195 allowed the recognition of four belemnites assemblages (TiBA-I to TiBA-IV) for the Tithonian part (Fózy
196 et al., 2011). Range charts showing the distribution of the complete ammonoid and belemnite fauna
197 were presented in Fózy 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 *Chitinoidea* Zone (Fig. 2). Two samples containing *Chitinoideidae* together with a few specimens of
202 *Praetintinnopsella* sp., were placed in the *Praetintinnopsella* 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*
208 zonal boundary, following the criteria of Remane et al. (1986). In comparing the Lókút ammonite
209 assemblages with calpionellid data, a general agreement is demonstrated where the data overlap. For
210 example, the first appearance of chitinoideids coincides with the base of the *Microcanthum* Zone (e.g.,
211 Benzaggagh et al., 2010).

212 Within the Hárskút HK-II section, the first beds above the radiolarite provided ammonites (Fig.
213 5) characteristic of the latest Kimmeridgian *Beckeri* Zone. Higher up the complete succession from the
214 *Hybonotum* to the *Ponti* Zone was documented by Fózy (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
231 the Occitanica to Boissieri ammonite zones. The overlying Lower Valanginian strata are condensed, but
232 yielded rich assemblages from the Pertransiens and Campylotoxus zones. Stable isotope analyses
233 revealed a well-defined positive $\delta^{13}\text{C}$ excursion in the Valanginian strata, identified as the Weissert
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

250 Measurements of the carbon isotope composition of bulk carbonate yielded positive $\delta^{13}\text{C}$
251 values throughout the sections examined. At Lókút, values around 2.5‰ characterise the lower,
252 Kimmeridgian part of the section, followed by a gradual negative shift, reaching a minimum of 0.0‰
253 within the Lower Berriasian. Higher up-section, a return towards more positive values up to 0.7‰ is
254 observed. Biostratigraphic data (Vigh, 1984; Főzy et al., 2011; Grabowski et al., 2010a) together with
255 magnetostratigraphic data (Grabowski et al., 2010a) allow us to accurately place the low point seen in
256 the carbon isotope curve within these schemes. This minimum appears in the upper part of

257 magnetosubzone M19n2n and towards the middle of calpionellid Zone B (i.e. the alpina Subzone) (Fig.
258 2). The oxygen isotope data at Lókút are more variable and range from ~ 0.0 to -3.2‰ . The highest
259 $\delta^{18}\text{O}$ values occur at the base of the section. Although showing a degree of scatter, isotope values
260 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
263 negative shift, reaching a minimum of 0.9‰ within the Lower Berriasian. Following this, a return
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
266 from ~ -1.8 to 0.3‰ . The most positive $\delta^{18}\text{O}$ values occur close to the base of the section and show
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 $\delta^{13}\text{C}$ 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,

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

284 Despite the differences in amplitude and offsets in absolute $\delta^{13}\text{C}$ values, there is in general a
285 good agreement of long-term $\delta^{13}\text{C}$ trends in all the sections compared, correlated on the basis of their
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).
289 Given the large distances between the sites (Fig. 7) it is notable that the overall shape the $\delta^{13}\text{C}$ curves
290 are similar in some intervals. The $\delta^{13}\text{C}$ decline through the Late Jurassic and across the Jurassic–
291 Cretaceous boundary, stable values in the Berriasian and a major Early Cretaceous positive $\delta^{13}\text{C}$
292 excursion, i.e. the Valanginian Weissert event, are clearly recognisable in all sections covering this
293 interval. With respect to the isotope data from Lókút Hill (Fig. 2), the $\delta^{13}\text{C}$ decline through the Late
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
300 amplitude $\delta^{13}\text{C}$ shifts (e.g., La Chambotte, eastern France) are potentially affected by a combination of
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)
303 high amplitude $\delta^{13}\text{C}$ variation is to be expected. Another noisy record is derived from the
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
306 interval studied (consistent for example with the Lókút section) the high-frequency changes in $\delta^{13}\text{C}$
307 most probably result from a mix of diagenetic and local environmental effects (Colombié et al., 2011).

308 Few $\delta^{13}\text{C}$ records across the Jurassic–Cretaceous boundary have been derived from organic
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
312 Tithonian is evident. Likewise a declining marine $\delta^{13}\text{C}_{\text{org}}$ trend is seen in DSDP site 534A data reported
313 by Falkowski et al. (2005) from the Tithonian, before a pronounced positive event is seen associated
314 with the Valanginian (Patton et al., 1984). Those Late Jurassic and Early Cretaceous $\delta^{13}\text{C}_{\text{org}}$ data derived
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.

319 In order to separate the anomalous, the regional and the global trends, an average $\delta^{13}\text{C}_{\text{carbonate}}$
320 stack was developed (Fig. 8), based on the sections compared and presented here (Fig. 6). The new
321 global $\delta^{13}\text{C}$ stack (Fig. 8) is used to visualise and identify those globally synchronous shifts in $\delta^{13}\text{C}$ that
322 can be applied for global correlation. The $\delta^{13}\text{C}$ stack does not include any estimated or calculated
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.

334 The stack clearly shows a decline in $\delta^{13}\text{C}$ throughout the Late Jurassic–Early Cretaceous,
335 reaching a minimum in the Early Cretaceous in magnetochron M12, near the base of the Campylotoxus
336 Zone (see also Weissert et al., 1998). The positive excursion in the Valanginian, (the Weissert event), is
337 plainly evident. Our data from Hárskút (Fig. 3) clearly reveals the positive excursion in the Valanginian
338 (see Főzy et al., 2010). The width of the grey envelope partly reflects the sampling density within the
339 data set, and additional data could certainly modify the picture. Nevertheless, for intervals with similar
340 data coverage, the outline of the envelope and its width may help evaluate the relative importance of

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

347 In comparison, the composite Late Jurassic–Early Cretaceous $\delta^{13}\text{C}$ curve in GTS 2012 shows little
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
358 towards the middle of calpionellid Zone B (the Alpina Subzone) (Figs. 2, 3) is not resolved in the $\delta^{13}\text{C}$
359 stack. Likewise, the positive Boreal $\delta^{13}\text{C}$ shift immediately above the Jurassic–Cretaceous boundary
360 correlated to Tethyan records recorded by Dzyuba et al. (2013) is also not resolvable in the $\delta^{13}\text{C}$ stack.

361 *5.2. Comparison and interpretation of $\delta^{13}\text{C}$ trends*

362 Our newly obtained stable isotope data from Lókút and Hárskút (Figs. 2, 3), taken together with
363 the $\delta^{13}\text{C}$ stack, as noted above, shows a shifts towards negative values throughout the Late Jurassic–
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,
366 increases in volcanic activity and episodic rapid methane release from gas hydrates contained in
367 marine sediments. Large negative excursions in marine carbonate $\delta^{13}\text{C}$ are often associated with
368 period boundaries and mass extinctions (Kump, 1991). Given the typically abrupt nature of isotope
369 excursions related to inferred methane fluxes (e.g., Menegatti et al., 1998), this mechanism appears
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
380 *Nannoconus* spp. and *Conusphaera* spp. correlating with low $\delta^{13}\text{C}$ values. Oligotrophic conditions in the
381 Tethyan seaway have been linked to drier climates and a sea level low during the latest Jurassic (e.g.,
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., Haq,
385 2014) may in part correlate with aridity, lower inputs of nutrients and the gradual negative $\delta^{13}\text{C}$ shift. A
386 kaolinite minimum is known from all over Europe and associated with a major Late Jurassic “dry event”
387 (e.g., Hallam et al. 1991, Abbink et al. 2001; Rameil, 2005; Schnyder et al., 2006). Rameil (2005),
388 inferred from cyclostratigraphy, the duration of the dry phase, as defined on the Jura platform, to be
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
391 transition phase (Rameil, 2005). However, the decline in $\delta^{13}\text{C}$ seen is not a short interval associated just
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 $\delta^{13}\text{C}_{\text{org}}$
396 trends derived from woody material and charcoal, noted above, to the marine carbonate $\delta^{13}\text{C}$ stack
397 clearly supports the notion that the surface ocean and atmosphere behaved as coupled reservoirs at
398 this time.

399 In contrast, the Sr isotope record for this interval (Fig. 8) (e.g., Jones et al., 1994; McArthur et al.,
400 2004; Bodin et al., 2009; Wierzbowski et al., 2012) shows a trend towards more radiogenic values from
401 a long-term low at the Callovian-Oxfordian boundary to a peak in the Barremian. This variation in Sr-
402 isotopes possibly reflects a change in the balance of flux from relatively non-radiogenic Sr derived from
403 mid-ocean ridge hydrothermal activity to relatively radiogenic Sr derived from continental weathering
404 (including changes in both total riverine flux and the isotopic composition of the flux). The $^{87}\text{Sr}/^{86}\text{Sr}$ low
405 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-
409 Oxfordian minimum $^{87}\text{Sr}/^{86}\text{Sr}$ ratios observed (Fig. 8). Indeed, the data Cogné and Humler (2006) do
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
419 be a major factor governing the evolution of marine $^{87}\text{Sr}/^{86}\text{Sr}$. Indeed, phosphorus flux rates (Föllmi,
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
423 latest Jurassic to Early Cretaceous (post the Late Jurassic “dry event”) are also observed (e.g., Thiede
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 and
428 Jenkyns, 2001).

429 The relatively short-lived arid episode (or dry phase *sensu stricto*, Rameil, 2005) and possible
430 linked short-term sea-level fall and rise (e.g., Haq, 2014) appears not to be reflected in the Sr-isotope
431 curve. The short duration of arid conditions and presumed reduction in continental weathering and
432 change in $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, is unlikely to be resolvable over such a short timescale as inputs and outputs
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,
438 Adatte et al., 1996) are evidently not expressed within the $\delta^{13}\text{C}$ record (Weissert and Mohr, 1996; Price
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
444 and sequestration of isotopically light marine carbon (^{12}C) may not be fully applicable for this interval.
445 Likewise, given the evolutionary rise of the modern plankton groups through Late Jurassic–Early
446 Cretaceous time one would anticipate an overall increase in $\delta^{13}\text{C}$ values in marine carbonates (e.g.,
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
452 relatively isotopically heavy carbon) could have dampened changes in $\delta^{13}\text{C}_{\text{DIC}}$ expected from elevated
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,
485 2012) may also have played a role in buffering the $\delta^{13}\text{C}$ record. The dominance of arid conditions on
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

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

494 *5.3. Oxygen isotopes and palaeoenvironmental change*

495 The preservation of primary $\delta^{13}\text{C}$ values during carbonate diagenesis is quite typical, and is
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,
498 commonly result in a change in oxygen isotope ratios leading to relatively light $\delta^{18}\text{O}_{\text{carb}}$ values (Hudson,
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 (Fózy et al., 2011) we are confident that the
503 trends do reflect a primary signal, independent of diagenesis. Increasingly negative $\delta^{18}\text{O}$ values are
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
519 cooling event is seen (a shift to more positive $\delta^{18}\text{O}$ values), followed by cooling through the Valanginian.
520 The Hauterivian shows a return to warmer conditions. Shorter term trends through the Jurassic-
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
524 Late Jurassic and Early Cretaceous, any recognisable correlation with the $\delta^{13}\text{C}$ curve is lacking. For
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 $\delta^{13}\text{C}$ 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 $\delta^{18}\text{O}$
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,

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

537

538 **6. Conclusions**

539 The $\delta^{13}\text{C}$ data from Hungary are consistent with other isotope stratigraphies and indicate that
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
542 the $\delta^{13}\text{C}$ record is challenging due to relatively stable $\delta^{13}\text{C}$ values showing a slope which is too slight.
543 The Berriasian minimum and the return to more positive values seen in our data from Lókút and
544 Hárskút is not resolved in the global $\delta^{13}\text{C}$ stack. Oxygen isotopes point to warming through the Late
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
548 Late Jurassic and Early Cretaceous any recognisable correlation with the $\delta^{13}\text{C}$ curve is lacking.

549 The Late Jurassic $\delta^{13}\text{C}$ 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 $\delta^{13}\text{C}$ 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
557 changes in $\delta^{13}\text{C}_{\text{DIC}}$ expected from elevated weathering and increased organic carbon burial rates
558 (particularly in marginal seas). We envisage also well-mixed parts of the ocean, perhaps as a result of
559 connections established between the Tethys and Central Atlantic, and the full opening of the Hispanic
560 Corridor effectively linking the Atlantic and Pacific Oceans. This scenario reconciles the apparently
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
564 of elements such as silica and phosphorus from the continents to the oceans (e.g., Föllmi, 1995)
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
572 counterbalances the effects of the “Mesozoic plankton revolution”.

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582

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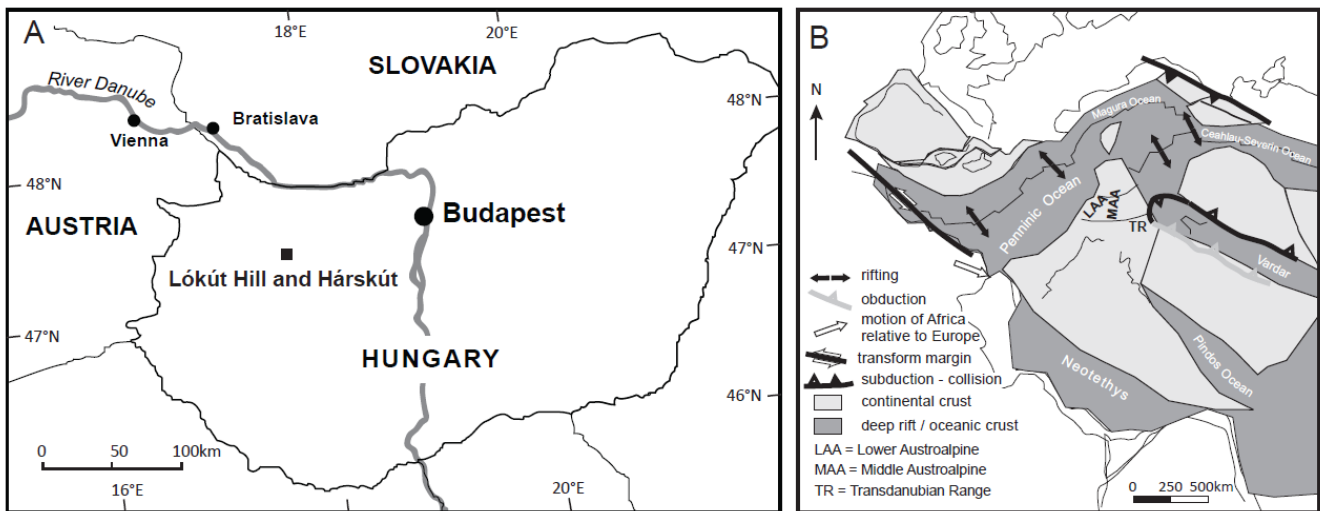
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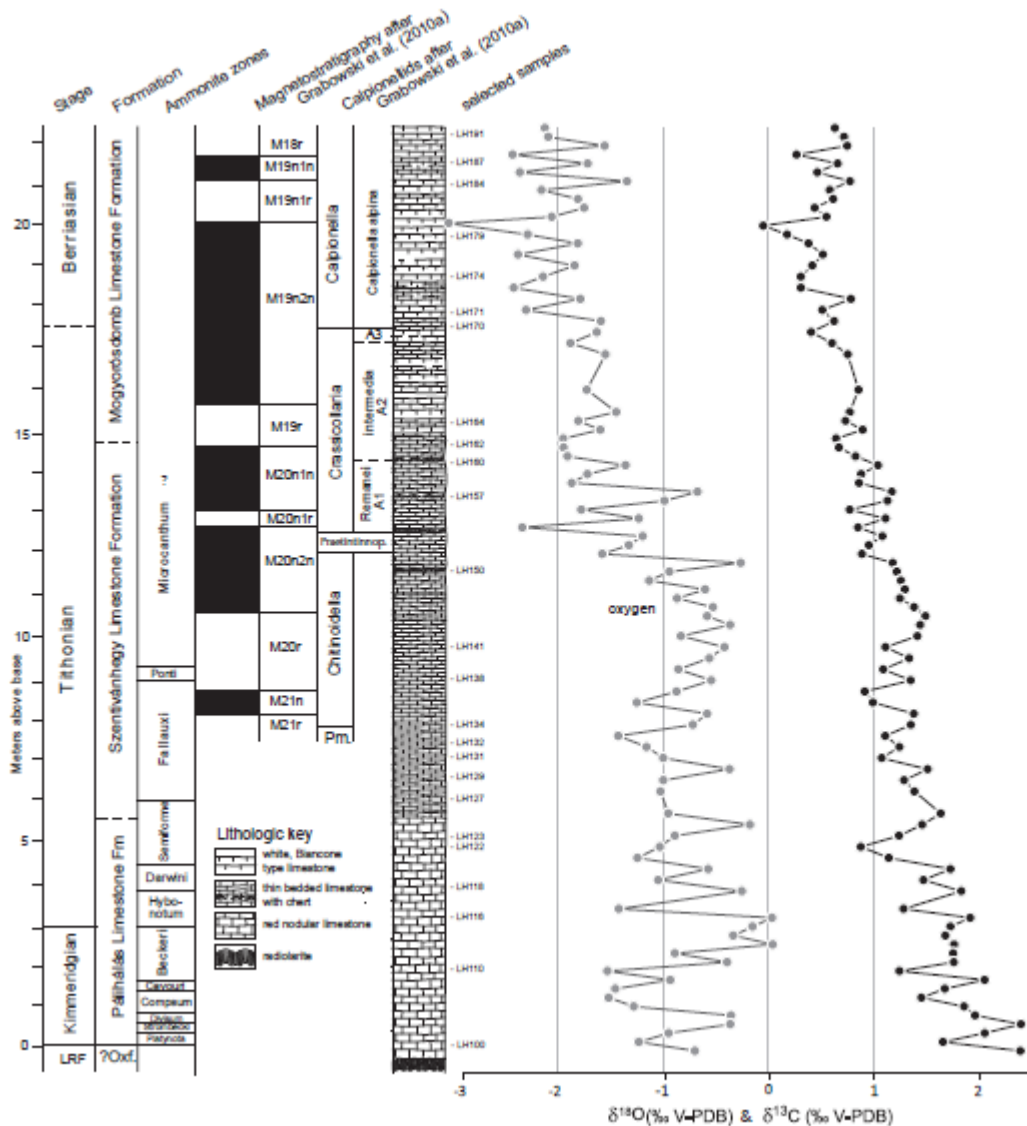
1007 **Fig. 1.** Location and palaeogeographic setting of the studied sections. A: Location of Lókút Hill and
1008 Hárskút in the Bakony Hills of the Transdanubian Range in western Hungary. B: Palaeogeographic
1009 setting of the Transdanubian Range (TR) and neighbouring units within a reconstructed Tithonian (Late
1010 Jurassic) western Tethyan palaeogeography (after Csontos and Vörös, 2004).



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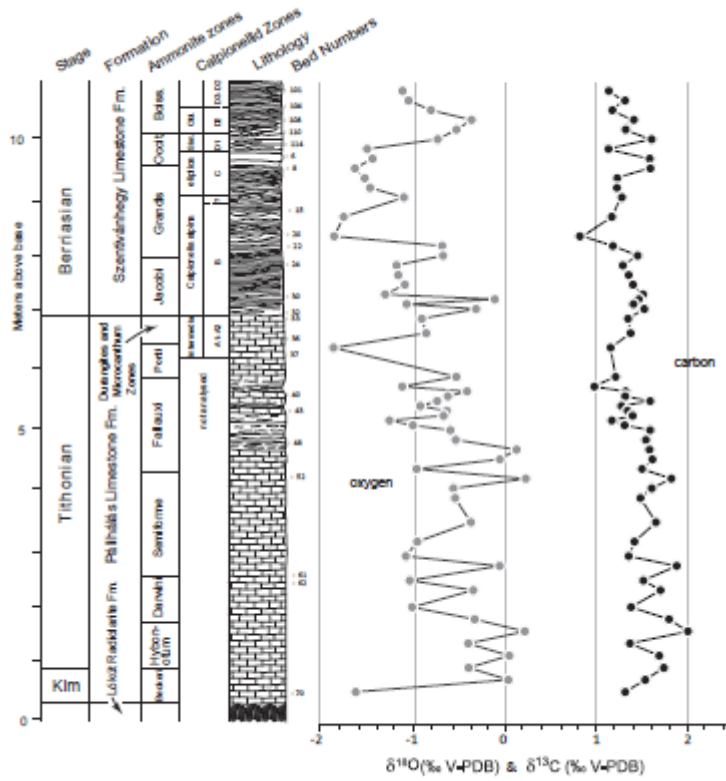
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1013 **Fig. 2.** Integrated biostratigraphy, magnetostratigraphy and carbon and oxygen isotope stratigraphy
 1014 from the Lókút section. The measured log and samples are referenced using the bed numbers of Vigh
 1015 (1984). Ammonite zones for the Kimmeridgian and Tithonian follow the zonation scheme by Enay and
 1016 Geysant (1975) and Geysant (1997). LRF = Lókút Radiolarite Formation. Pm. = *Parastomiosphaera*
 1017 *malmica* Zone. Belemnite assemblages are from Főzy et al. (2011).



1018

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.



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1024 **Fig. 4.** Representative and age-diagnostic Late Jurassic ammonites from the Lókút section. Inventory
1025 numbers of the Department of Paleontology and Geology of the Hungarian Natural History Museum
1026 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.

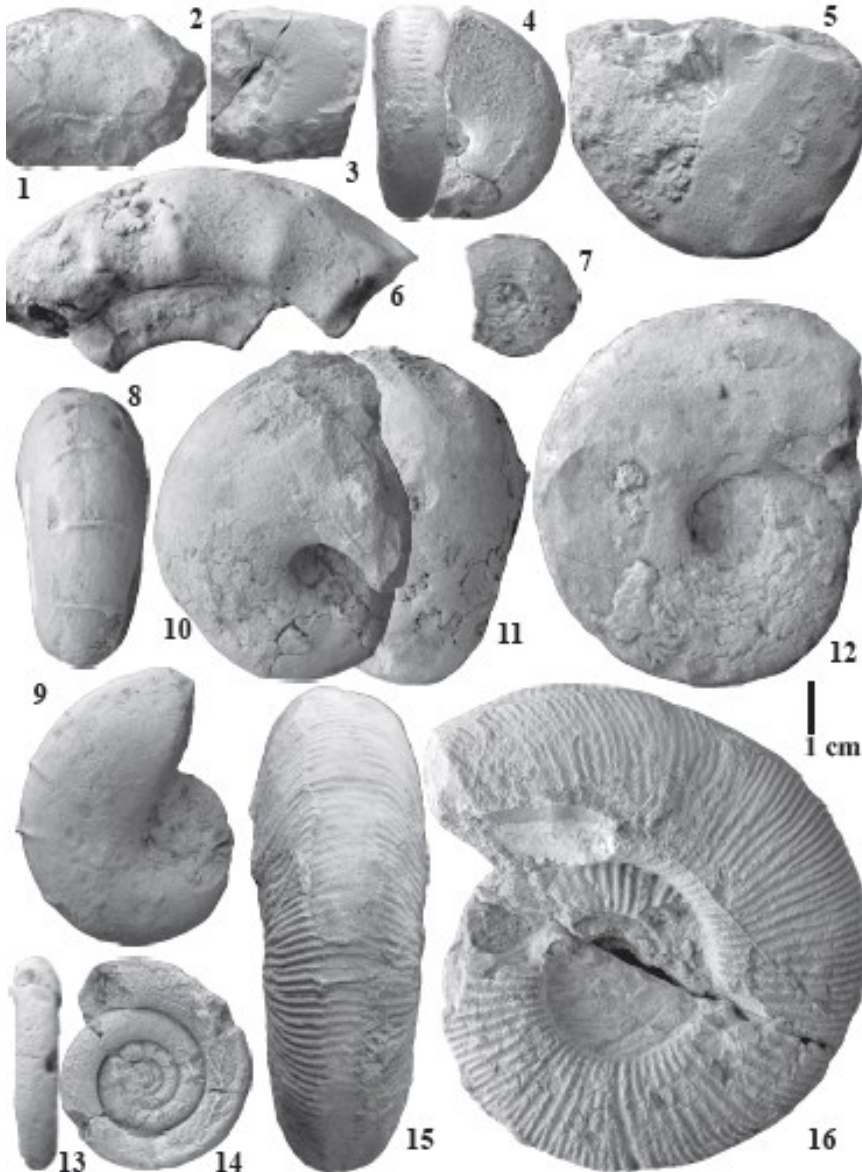
1029 4, 5. *Trapanesites adelus* (Gemmellaro, 1872), INV.2014.77, Bed LH 110-111, Compsum Zone (?).



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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 figures
1034 are natural size.

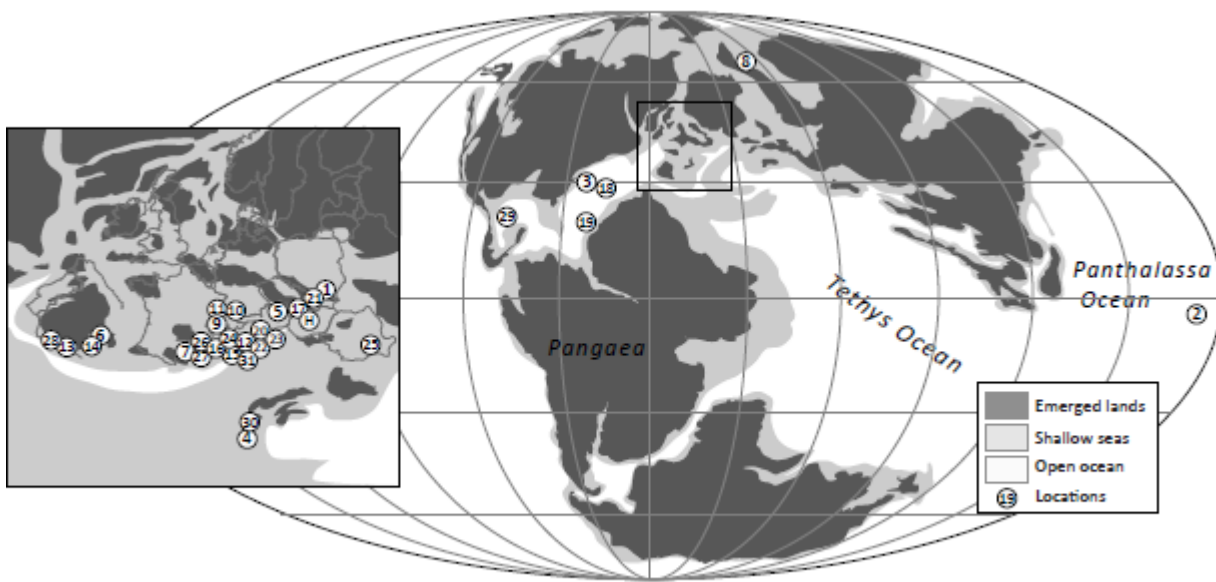
- 1035 1. *Haploceras verruciferum* (Zittel, 1869), J 10923, Bed 60, Semiforme Zone.
1036 2. *Semiformiceras fallauxi* (Oppel, 1865), J 10875, Bed 54, Fallauxi Zone.
1037 3, 4. *Haploceras carachtheis* (Zeuschner, 1846), J 10908, Bed 49, Fallauxi Zone.
1038 5. *Semiformiceras semiforme* (Oppel, 1865), 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.



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1058 **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 $\delta^{13}\text{C}$ 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.

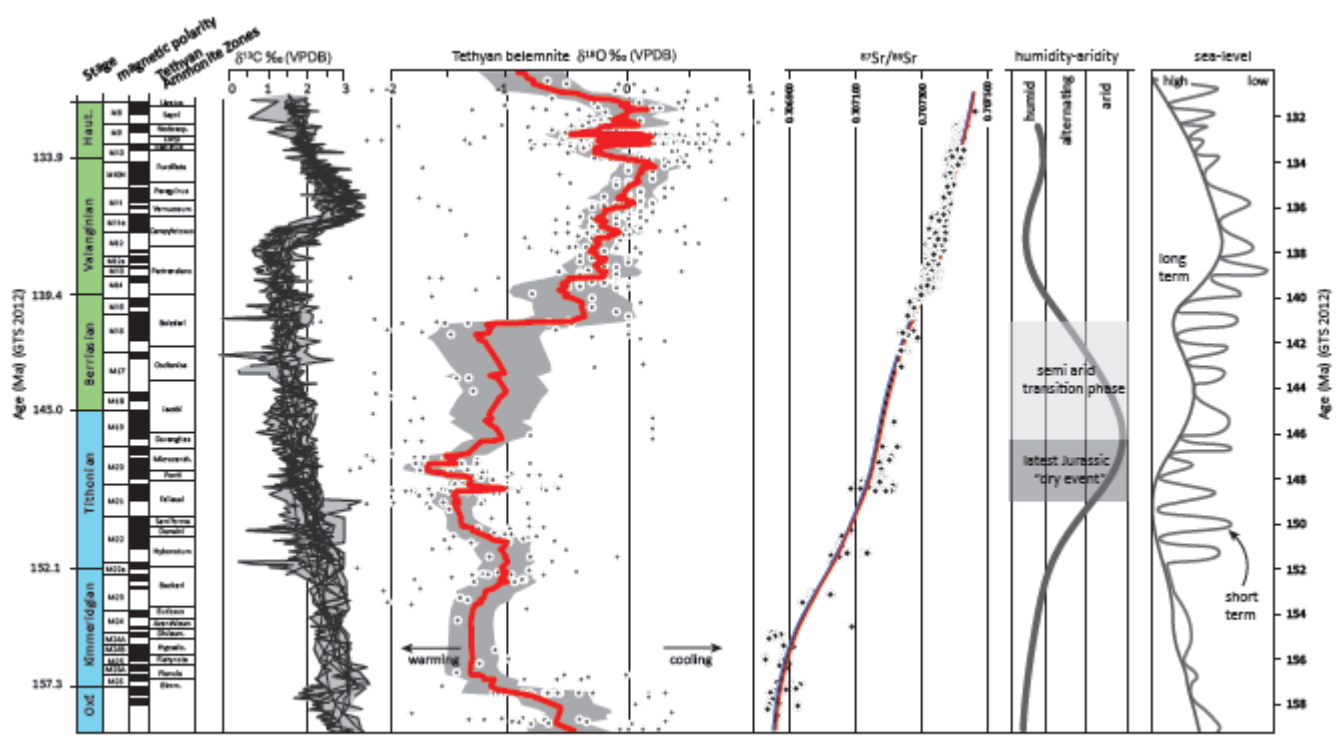
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1065 **Fig. 8.** A global $\delta^{13}\text{C}$ stack calibrated with magnetostratigraphy. The $\delta^{13}\text{C}_{\text{carb}}$ data are from bulk
 1066 sediments as shown in detail in Fig. 6, excluding data from the Subpolar Urals and North Siberia
 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).



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1075 **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.

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

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