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Environmental impact and magnitude of paleosol-
 1
     carbonate carbon-isotope excursions marking five
 2
     early Eocene hyperthermals in the Bighorn Basin,
 3
     Wyoming
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26 Abstract

27 Transient greenhouse warming events in the Paleocene and Eocene were associated 28 with the addition of isotopically-light carbon to the exogenic atmosphere-ocean 29 carbon system, leading to substantial environmental and biotic change. The 30 magnitude of an accompanying carbon isotope excursion (CIE) can be used to 31 constrain both the sources and amounts of carbon released during an event, and also 32 to correlate marine and terrestrial records with high precision. The Paleocene Eocene 33 Thermal Maximum (PETM) is well documented, but CIE records for the subsequent 34 warming events are still rare, especially from the terrestrial realm.

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36 Here, we provide new paleosol-carbonate CIE records for two of the smaller 37 hyperthermal events, I1 and I2, as well as two additional records of ETM2 and H2 in 38 the Bighorn Basin, Wyoming, USA. Stratigraphic comparison of this expanded, high-39 resolution terrestrial carbon isotope history to the deep-sea benthic foraminiferal isotope records from ODP Sites 1262 and 1263, Walvis Ridge, in the southern 40 41 Atlantic Ocean corroborates that the Bighorn Basin fluvial sediments record global 42 atmospheric change. The \sim 34-m thicknesses of the eccentricity-driven hyperthermals 43 in these archives corroborate precession-forcing of the ~7-m thick fluvial overbankavulsion sedimentary cycles. Using CALMAG bulk-oxide mean-annual-precipitation 44 45 reconstructions, we find similar or slightly wetter than background soil moisture 46 contents during the four younger hyperthermals, in contrast to soil drying observed 47 during the PETM using the same proxy, sediments, and plant fossils.

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49 The magnitude of the CIEs in soil carbonate for the four smaller, post-PETM events 50 scale nearly linearly with the equivalent event magnitudes documented in marine 51 records. In contrast, the magnitude of the PETM terrestrial CIE is at least 5‰ smaller 52 than expected based on extrapolation of the scaling relationship established from the 53 smaller events. We evaluate the potential for recently documented, non-linear effects 54 of pCO_2 on plant-photosynthetic C-isotope fractionation to explain this scaling 55 discrepancy. We find that the PETM anomaly can be explained only if background 56 pCO_2 was at least 50% lower during most of the post-PETM events than prior to the 57 PETM. Although not inconsistent with other pCO_2 proxy data for the time interval, 58 this would require declining pCO_2 across an interval of global warming. A more 59 likely explanation of the PETM CIE anomaly in pedogenic carbonate is that other 60 environmental or biogeochemical factors on the terrestrial CIE magnitudes were not 61 similar in nature or proportional to event size across all of the hyperthermals. We 62 suggest that contrasting regional hydroclimatic change between the PETM and 63 subsequent events, in line with our soil proxy records, may have modulated the 64 expression of the global CIEs in the Bighorn Basin soil carbonate records.

65

66 **1** Introduction

67 During the late Paleocene and early Eocene around 60 to 50 million years ago 68 massive amounts of carbon were released in pulses into the ocean-atmosphere 69 exogenic carbon pool causing a series of transient global warming events, known as 70 hyperthermals (Kennett and Stott, 1991; Cramer et al. 2003; Zachos et al., 2005; 71 Lourens et al., 2005). These events represent the best paleo-analogs for current 72 greenhouse gas warming, despite the very different background climatic, atmospheric, 73 and geographic conditions, and potentially the different time scales on which they 74 occurred (Bowen et al., 2006; Zachos et al., 2008; Cui et al., 2011; Bowen et al. 75 2015). The largest of the hyperthermals, the Paleocene-Eocene Thermal Maximum 76 (PETM) at 56 million years ago, is known to have caused severe climatic and marine 77 and terrestrial biotic change (Thomas, 1989; Gingerich, 1989; Kennett and Stott, 78 1991; Koch et al., 1992), comprehensively reviewed in McInerney and Wing (2011). 79 Recently, records of the secondary hyperthermals (i.e., ETM-2/H1, ETM3/K) became 80 available (Cramer et al. 2003; Lourens et al. 2005; Nicolo et al. 2007; Abels et al. 2012; Chen et al. 2014; Lauretano et al. 2015), while their environmental and biotic 81 82 impact has yet to be resolved (Sluijs et al., 2009; Stap et al., 2010a,b; Abels et al., 83 2012; D'Haenens et al. 2014).

84

85 All hyperthermals are characterized by a distinct geochemical signature, a negative 86 carbon isotope excursion, indicating that the carbon released to the exogenic carbon 87 pool during these events had a dominant biogenic origin (Dickens et al., 1995). The 88 potential biogenic sources range from plant material to methane. With the carbon 89 isotope excursions, and independent constraints on the mass of carbon release, it 90 should be possible to identify the source. The mass can be constrained by several 91 approaches, for example quantifying ocean acidification, or pCO_2 by proxy, either 92 direct (e.g., epsilon p) or indirect (e.g., SST) (Dickens et al., 1997; Dickens, 2000;

93 Bowen et al., 2004; Ridgwell, 2007; Panchuk et al., 2008; Zeebe et al., 2009), though 94 the uncertainty with these approaches is large (Sexton et al., 2011; DeConto et al., 95 2012; Dickens, 2011). Nevertheless, in theory, if there was a single source of carbon 96 for all CIE, the scaling with mass should be predictable. This requires that, firstly, the 97 exact size of the carbon isotope excursions (CIEs) in the global exogenic carbon pool 98 during hyperthermal events be well constrained and, secondly, the factors that 99 fractionating C-isotopes between the substrate reservoirs and organic and carbonate 100 proxies be well understood (Sluijs and Dickens, 2012).

101

102 Paleosol or pedogenic carbonate is precipitated from CO₂ that stems from respiration 103 of roots and plant litter in the soil and from atmospheric CO₂ diffusing into the soil. 104 Plant CO₂ from C₃ plants is typically fractionated by -16 to -24‰ compared to atmospheric CO₂ (O'Leary, 1988). Paleosol carbonate is a mix of both isotopically-105 106 distinct sources, modified by fractionation associated with diffusion, carbonate 107 equilibrium, and calcite precipitation, and therefore registers values between -7 and -11‰ in non-hyperthermal conditions in Paleogene soils covered by C₃ vegetation. 108 109 Paleosol carbonate records the atmospheric carbon isotope excursions related to the 110 Paleocene-Eocene Thermal Maximum (PETM), though amplified with respect to 111 marine carbonate (Bowen et al., 2004). This amplification has been attributed to 112 increased soil productivity and humidity during the hyperthermal events (Bowen et al., 2004; Bowen and Bowen, 2008), by changing plant communities (Smith et al., 113 114 2007), and by higher pCO_2 (Schubert and Jahren, 2013).

115

116 In a recent study, the carbon isotope anomalies associated with ETM2 and H2 were 117 documented in paleosol carbonate, allowing for comparison of the terrestrial 118 amplification of the CIEs relative to the PETM (Abels et al. 2012). An apparent linear 119 scaling of the marine and terrestrial carbon isotope excursions for the PETM, ETM2 120 and H2 events was invoked to suggest that all three events may have reflected a 121 common mechanism of global change. Interpretation of this signal is complicated, 122 however, by shifting background climate conditions between the events, which are 123 separated by close to 2 million years of gradual greenhouse warming (Zachos et al., 124 2008; Littler et al., 2014), and by the fact that the observed relationship did not 125 converge on the origin, leaving the carbon isotope scaling associated with smaller 126 events (e.g., I1 and I2) uncertain.

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128 Here, we extend the existing record of three hyperthermals from the Bighorn Basin 129 with data documenting two new CIEs (I1 and I2). We further report additional records 130 of the ETM2 and H2 CIEs within the Basin and analyze bulk oxides in thick (>0.75 131 m) soils to reconstruct soil moisture values through these greenhouse-warming events. 132 We compare our records with the new benthic foraminiferal records generated for 133 Ocean Drilling Program Site 1263 at Walvis Ridge, Atlantic Ocean (Lauretano et al., 134 2015), and a bulk sediment carbon isotope record from ODP Site 1262 (Zachos et al., 135 2010; Littler et al., 2014), Walvis Ridge, to investigate coeval carbon isotope change 136 and registration of multiple CIEs in the different carbonate proxies. We analyze these 137 records in the context of recently characterized dependence of plant carbon isotope 138 fractionation on atmospheric CO₂ partial pressure (Schubert and Jahren, 2012), 139 including scenarios that allow for changing background conditions across the late 140 Paleocene-Early Eocene.

- 141
- 142 **2** Material and Methods

Pedogenic carbonate nodules were sampled at 12.5 cm spacing where present after removal of the weathered surface in the West Branch and Creek Star Hill sections located in the McCullough Peaks area of the northern Bighorn Basin, Wyoming (USA; Fig. 1). Sediment samples from soil-B horizons for reconstruction of Mean Annual Precipitation (MAP) are from the same sections and from the Upper Deer Creek section of Abels et al. (2012).

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150 Micritic parts of the nodules were cleaned and ground to powder, while spar was 151 taken out after crushing the nodule in few pieces. Carbon isotope ratios of carbonate 152 micrite were measured using a SIRA-24 isotope ratio mass spectrometer of VG 153 (vacuum generators) at Utrecht University (Netherlands). Prior to analysis, samples 154 were roasted at 400°C under vacuum before reaction with dehydrated phosphoric acid 155 in a common-bath system for series of 32 samples and 12 standards. Carbon isotope ratios are reported as δ^{13} C values, where δ^{13} C = (R_{sample}/R_{standard} - 1), reported in per 156 mil units (‰), and the standard is VPDB. These isotope ratio measurements are 157 158 normalized based on repeated measurements of in-house powdered carbonate 159 standard (NAXOS) and analytical precision was calculated from inclusion of three

160 IAEA-CO1 standards in every series of 32 samples. Analytical precision is $\pm 0.1 \%$ 161 for $\delta^{13}C(1\sigma)$, whereas variability within individual paleosols averaged 0.2‰.

162

163 To calculate CIE magnitudes, carbon isotope records are first detrended to exclude the 164 influence of the long term Paleocene to early Eocene trends. The CIE magnitudes are 165 then calculated as the difference between pre-excursion carbon isotope values and 166 excursion values within the core of the main body (Table 2; Supporting Information). 167 Standard errors are calculated using variability in background and excursion values.

168

169 **3 Results**

170 **3.1 Bighorn Basin**

High-resolution pedogenic carbonate carbon isotope records are constructed for the 171 172 lower Eocene of the Willwood Formation in the McCullough Peaks area, northern 173 Bighorn Basin, Wyoming (USA; Fig. 1). Previous work included the Upper Deer 174 Creek (UDC) section, where the carbon isotope excursions of ETM2 and H2 175 hyperthermal events were located (Abels et al. 2012). Here, we analyze two parallel 176 sections, the Creek Star Hill (CSH) and West Branch (WB) sections, separated by 1 to 177 2 kilometers from the UDC section (Fig. 1). The isotope record is extended upwards 178 in the WB section and downwards in the Deer Creek Amphitheater section (DCA; 179 Abels et al. 2013). We construct a composite stratigraphic section by connecting the 180 four sections via lateral tracing of marker beds in the field, such as the P1 to P8 purple 181 soils in the ETM2-H2 stratigraphic interval (Abels et al., 2012).

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183 The carbon isotope record of paleosol carbonate of the McCullough Peaks (MCP) 184 composite section shows four carbon isotope excursions (CIEs; Fig. 2). The lower 185 excursions of ~ 3.8 and $\sim 2.8\%$ in magnitude (see methods for CIE magnitude 186 calculation) have previously been related to the ETM2/H1 and H2 events (Abels et 187 al., 2012) and are shown to be similar in the parallel Upper Deer Creek, West Branch, 188 and Creek Star Hill sections. This confirms the presence and regional preservation of 189 these CIEs in the Willwood Formation. The two younger carbon isotope excursions 190 are ~ 2.4 and $\sim 1.6\%$ in magnitude and both located in the West Branch section (Fig. 191 2). These excursions likely relate to the CIEs of the I1 and I2 events that occur in the

subsequent 405-kyr eccentricity maximum after ETM2/H1 and H2 (Cramer et al.,2003).

194

195 Besides these CIEs, several intervals show less-well-defined negative carbon isotope 196 excursions of ~0.5-1%; two below ETM2 at MCP meter levels 95 and 145, two 197 above H2 at meter levels ~260 and ~290, and one above I2 at meter 400. This scale of 198 variability is harder to detect as the carbon isotopes show background variability of 199 $\sim 1\%$ (2 σ), possibly noise related to local environmental factors. The spacing between 200 the CIEs and the low amplitude variability in the MCP section is on average ~34 m. 201 Bandpass filtering of this scale of variability specifically shows a strong coherent 202 variation through the ETM2 to I2 interval (Fig. 3).

203

204 Precession-forcing of overbank-avulsion lithological cyclicity in the Willwood 205 Formation was recently substantiated with data from the Deer Creek Amphitheater 206 section (Abels et al., 2013). In the DCA section, the cyclicity occurs at a scale of \sim 7.1 207 meters. In the three sections now covering ETM2-H2, the cyclicity has a very similar 208 average thickness of \sim 7.1 meters. The precession cyclicity comprises heterolithic 209 sandy intervals showing little pedogenic imprint alternating with mudrock intervals 210 showing intense pedogenesis. In the precession forcing sedimentary synthesis, the 211 heterolithic intervals are related to periods of regional avulsions and rapid 212 sedimentation, while the mudrocks are related to periods of overbank sedimentation 213 when the channel belt had a relatively stable position (Abels et al., 2013). This scale 214 of sedimentary cyclicity is also observed higher in the West Branch section. Average 215 climatic precession cycles in the Eocene last ~20 kyr resulting in ~7.1 meters of 216 sediment. This gives an average sedimentation rate of ~ 0.35 m/kyr, resulting in ~ 96 217 kyr for the 34 meter cyclicity observed in the carbon isotope records in the ETM2-I2 218 interval. This is in line with ~100-kyr eccentricity forcing of individual hyperthermals 219 and a 405-kyr eccentricity forcing of the ETM2-H2 and I1-I2 couples.

220

We produce mean annual precipitation (MAP) estimates across the ETM2-I2 interval with the CALMAG method that uses bulk oxide ratios in soil-B horizons (Nordt and Driese, 2010). Conservatively the method reconstructs soil moisture contents in these ancient soils. Ideally, soil-B horizons thicker than 1 meter should be used for this 225 proxy (Adams et al., 2011). We measured all 59 soil-B horizons thicker than 1m, 226 where possible in multiple, parallel sections. In addition, we measure 24 soil-B 227 horizons between 0.5 and 1 m. Our estimates from the 83 individual soils show a 228 stable soil moisture regime in the early Eocene Bighorn Basin with mean annual 229 precipitation estimates of around 1278 mm/yr (2σ 132 mm/yr; Fig. 2). All except one 230 soil-B horizon thicker than 1.25 m fall in this range. Soil-B horizons below 1.25 m 231 thickness occasionally show drier outliers, of which three are below 1000 mm/yr. 232 There are no striking changes during ETM2, H2, I1, or I2 hyperthermal events. The 5 233 soils that contribute to our ETM2 reconstructions show a potentially slightly enhanced 234 soil moisture contents with reproduced annual rainfall of 1337 (2σ 88 mm/yr), while 235 the 11 soils in H2 show 1267 mm/yr (±166), not different from reconstructions for 236 background climate states. There are slightly more dry outliers both in as well as just 237 out the hyperthermals, especially H2, but it should noted that these intervals also have 238 denser sampling, because of the replication of data for these intervals in three parallel 239 sections.

240

241 3.2 Walvis Ridge

242 For comparison of time equivalent carbon isotope change, we use existing and new 243 benthic foraminiferal Nuttallides truempyi records from Site 1263 (McCarren et al., 244 2008; Stap et al., 2010a; Lauretano et al., 2015), the shallowest site of Walvis Ridge 245 with a paleodepth of ~1500 meters. For Site 1263, because N. truempyi specimens are 246 absent in the main body of the PETM, the benthic record includes data for the 247 infaunal species Oridorsalis umbonatus, which is isotopically similar (McCarren et 248 al., 2008). The O. umbonatus data cover most of the CIE though no shells were 249 recovered from the lowermost portion of the clay layer. Data for the ETM2-H2 events 250 are from Stap et al. (2010a) and for I1-I2 from Lauretano et al. (2015). Benthic 251 foraminifera are mostly absent within the Elmo clay layer at Site 1263. A compilation 252 of all Walvis Ridge sites shows very similar benthic carbon isotope excursion values 253 for ETM2 (Stap et al., 2010a). Therefore, we use the next shallowest Site 1265 254 (paleodepth ~1850 m) to cover the missing ETM2 peak excursion values in Site 1263. 255 The data from N. truempyi at Site 1263, generated at 5-cm resolution across the I1 and 256 I2 events, show benthic CIEs of 0.88‰ for I1 and 0.73‰ for I2 (Fig. 3).

258 As a framework for correlation, we plot the long, high-resolution bulk carbonate 259 carbon isotope record from ODP Site 1262 (Zachos et al., 2010) and benthic carbon 260 isotope record from ODP Site 1263 (Fig. 3). Site 1262 is the deepest Site from the 261 ODP Leg 208 Walvis Ridge transect with an approximate paleodepth of 3600 meters. 262 Site 1262 carbon isotope record is orbitally tuned (Westerhold et al., 2008) and 263 capture all Eocene CIE, PETM, ETM2, H2, and I1 and I2 events (Zachos et al. 2010; 264 see also Littler et al. 2014), though the PETM is clearly truncated due to dissolution 265 (Zachos et al. 2005).

266

267 **3.3 CIE comparison with fixed background** *p***CO**₂

268 The new records show that CIE magnitudes of both terrestrial and marine substrates 269 decrease progressively across the 5 hyperthermal events (Fig. 4). For the four smaller 270 events, the pedogenic carbonate and benthic foraminifera record are strongly, linearly 271 correlated ($r^2 = 0.97$). The data for the larger PETM event, however, deviate strongly 272 from this trend. As described above, it has previously been observed that Eocene 273 hyperthermal pedogenic carbonate CIEs are generally amplified in magnitude relative 274 to their marine counterparts (Bowen et al. 2004; Smith et al. 2007; Schubert and 275 Jahren, 2013). The new data suggest that the mechanisms leading to this amplification 276 were stronger, relative to the size of the event, for the smaller events than for the 277 PETM.

278

279 We evaluate this observation in the context of one mechanism, sensitivity of land 280 plant photosynthetic ¹³C-discrimination to change in pCO_2 , which may affect the C-281 isotope offset between marine and terrestrial substrates differently among events. We 282 conduct two sets of model experiments, adopting a common framework for both 283 based on the assumption that the carbon sources and nature of environmental change 284 during each event were comparable. Although this assumption is likely over-285 simplistic, it allows us to evaluate the effects of the photosynthetic discrimination 286 mechanism in isolation and directly evaluate its potential contribution to CIE 287 expression in the new terrestrial records. Specifically, we assume that for each event 288 the CIE magnitude in the atmosphere $(D\delta_{a,h})$ is equal to the CIE magnitude in marine 289 (benthic) records. We also assume that peak pCO_2 change for each hyperthermal 290 (Dp_h) is a linear function of marine (benthic) CIE magnitude, which is to some extent supported by the temperature change derived from $D\delta^{18}O$ scaling with $D\delta^{13}C$ (Stap et al. 2010a; Lauretano et al. 2015), such that

293 $Dp_h = Dp_{PETM} \times D\delta_{a,h} / D\delta_{a,PETM}.$ (1)

As a starting point for our analysis, we use C-isotope data from leaf wax lipids that constrain the magnitude of the PETM CIE within Bighorn Basin plants ($D\delta_{p,PETM} \approx -$ 4.2%; Smith et al., 2007). Decomposing the plant CIE into

$$D\delta_{p,PETM} = D\delta_{a,PETM} - D\Delta_{PETM},\tag{2}$$

where Δ is photosynthetic C-isotope discrimination, we solve for the change in discrimination during the PETM (+0.8‰ using the Walvis Ridge benthic data to estimate $D\delta_{a,PETM}$).

301

302 For any background pCO_2 condition prior to the PETM ($p_{bkg,PETM}$) we can calculate an 303 estimate of plant carbon isotope discrimination ($\Delta_{bkg,PETM}$) using equation 6 of 304 Schubert and Jahren (2012). This idealized value corresponds to fractionation for 305 plants under experimental conditions that are not water or light limiting and is used 306 throughout our modeling when we refer to values of Δ . Adding this value and $D\Delta_{PETM}$ 307 we obtain an equivalent value for PETM photosynthetic discrimination, Δ_{PETM} . We 308 then invert the photosynthetic discrimination equation to find the PETM pCO_2 309 concentration (p_{PETM}) that gives the estimated discrimination:

310

$$p_{PETM} = (\Delta_{PETM} \times a / b + \Delta_{PETM} \times c - a \times c) / (a - \Delta_{PETM}), \tag{3}$$

311 where a = 28.26, b = 0.21, and c = 25 are empirically optimized parameter values (Schubert and Jahren, 2012). Although environmental and physiological factors 312 313 almost certainly caused the actual, absolute magnitude of plant carbon isotope 314 discrimination in the Paleocene-Eocene Bighorn Basin to be different from the Δ 315 values calculated here, our results depend only on the change in \varDelta between 316 background and hyperthermal conditions, and thus on the assumption that the form of 317 the discrimination equation accurately describes the response of Bighorn Basin plants. 318 Below, we discuss how changes in other environmental parameters during 319 hyperthermals may compromise this assumption. We used this approach to calculate values of p_{PETM} and change in PETM pCO_2 (Dp_{PETM}) across a range of assumed 320 321 background pCO_2 conditions from 250 to 3,000 ppmv (Figure given in Appendix B). 322

323 Building from this framework, our first set of model experiments assume an invariant 324 background pCO_2 value across all five events to evaluate whether the non-linear 325 response of changing photosynthetic discrimination to a range of Dp_h magnitudes 326 across the events can explain the non-linear CIE scaling observed in the terrestrial 327 records. Using $p_{bkg,h} = p_{bkg,PETM}$ and the Dp_h values estimated for each event, we 328 calculated $D\Delta_h$ for each event using the previously referenced photosynthetic 329 discrimination equation. We then apply equation 2 to each event to calculate an 330 estimate of $D\delta_p$ for and compare the implied plant CIE magnitude ($CIE_p = 0 - D\delta_p$) 331 with the observed soil carbonate CIEs to evaluate whether these scale proportionally 332 across all 5 events. If change in plant discrimination explains the non-linear scaling of 333 the paleosol carbonate CIE magnitudes (CIE_c) , assuming all other soil/environmental 334 influences scale proportionally with event magnitude, then we expect that for all 335 events;

$$CIE_{c,h} = CIE_{p,h} \times \beta_1 + \beta_o. \tag{4}$$

Nowhere within the range of background pCO_2 values tested here is this the case (Fig. 5), suggesting that changing photosynthetic discrimination in isolation and under the assumption of near-constant background pCO_2 cannot explain the variation in CIE expression in Bighorn Basin soil carbonates. The exercise shows that large changes in absolute background pCO_2 values do not significantly impact the results.

342

343 3.4 Impact on CIE magnitudes of variable background *p***CO**₂

For our second set of experiments, we allow background $pCO_2(p_{bkg})$ to change across the study interval and evaluate the p_{bkg} conditions required to reconcile the observed pattern of soil carbonate CIE magnitudes with the marine record. Our initial assumptions and estimates of PETM discrimination and pCO_2 change are as described in section 3.3.

349

Here we assume that equation 4 does describe the relationship between plant and soil carbonate CIEs, and that there are no fixed offset effects (i.e. $\beta_o = 0$, all factors that affect the size of the carbonate CIEs relative to the plant CIEs scale linearly with event size). It follows that the plant CIE magnitude for each event is

354
$$D\delta_{p,h} = D\delta_{p,PETM} \times D\delta_{c,h} / D\delta_{c,PETM}.$$
 (6)

355 We then calculate the change in photosynthetic discrimination for each event as:

356 L

$$D\Delta_h = D\delta_{a,h} - D\delta_{p,h} \tag{7}$$

357 We now have two differences, Dp_h and $D\Delta_h$, for each event. From the photosynthetic discrimination equation, we can write 358

 $D\Delta_h = \frac{ab(p_{bkg,h} + Dp_h + c)}{a + b(p_{bkg,h} + Dp_h + c)} - \frac{ab(p_{bkg,h} + c)}{a + b(p_{bkg,h} + c)}.$ 359 (8)

360 This can be rearranged to give:

361
$$b^{2}p_{bkg,h}^{2} + b(2a + 2bc + bDp_{h})p_{bkg,h} =$$

362 $a^{2} + 2abc + abDp_{h} + b^{2}c(Dp_{h} + c) - \frac{a^{2}bDp_{h}}{D\Delta_{h}},$ (9)

363 a quadratic which can be solved to obtain the background pCO_2 value required for 364 each hyperthermal to give linear scaling between CIE_p and CIE_c across the events (at 365 any prescribed value of $p_{bkg,PETM}$).

366

367 The analysis suggests that the non-linear scaling of the soil carbonate CIEs relative to 368 the marine record can be explained across the entire range of assumed $p_{bkg,PETM}$ conditions through changes in photosynthetic ¹³C-discrimination forced by 369 370 hyperthermal pCO_2 increase over a varying background pCO_2 conditions (Fig. 6). For any assumed PETM background pCO_2 , our results require a > 50% decrease in 371 372 background pCO_2 during the ~2 Myr interval separating the PETM and ETM2. The 373 analysis requires sustained, low background pCO_2 which rises gradually across the 374 two subsequent events, before a more abrupt increase prior to the I2 event. Across 375 most of the range of initial conditions evaluated the results require non-hyperthermal 376 background pCO_2 values substantially lower than $p_{bkg,PETM}$ throughout the Early 377 Eocene. The fractional change in pCO_2 required, relative to PETM background conditions, is lower for higher assumed $p_{bkg,PETM}$, but larger absolute changes in pCO_2 378 379 are required for these cases.

380

381 4 Discussion

382

Fluvial sedimentary archives of the Bighorn Basin 4.1

383 The presence of five carbon isotope excursions demonstrates that the river floodplain 384 sedimentary successions in the Bighorn Basin firmly record these global atmospheric 385 events. The two new parallel series in the Bighorn Basin confirm the presence of 386 ETM2 and H2 (Abels et al., 2012). The records of the I1 and I2 events represents the 387 first equivalents in fluvial strata. In the terrestrial realm, a CIE has been found in coal388 seams in the Fushun Basin, China, which has been related to I1 (Chen et al., 2014),

- 389 while I2 has not yet been recorded in any other terrestrial record.
- 390

391 The bulk oxide CALMAG proxy data have been proposed to reflect mean annual 392 precipitation (MAP) through its influence on soil mineral weathering and cation 393 leaching (Nordt and Driese, 2010; Adams et al. 2011). Here, we conservatively use 394 the method as a proxy for soil moisture rather than mean annual precipitation. The 395 data indicate no or slight increases in soil moisture during the four early Eocene 396 hyperthermals. This strongly deviates from observations of paleohydrologic change 397 for the PETM in the northern and southern Bighorn Basin, where the same proxy 398 indicates a decrease in soil moisture (Kraus and Riggins, 2007; Kraus et al., 2013), 399 consistent with a soil morphology index (Kraus et al., 2013), and analysis of fossil 400 leaves (Wing et al., 2005; Kraus et al., 2013). This would suggest that the regional 401 climatic and/or environmental response to the PETM differed from the post-PETM 402 hyperthermals.

403

404 Besides precipitation, temperature, vegetation, and sediment type and rates also have 405 a large impact on soil moisture and changes in CALMAG geochemical data should be 406 considered in light of changes in these factors (Kraus et al., 2013). For the four 407 younger hyperthermals, there are no temperature or vegetation data available for the 408 Bighorn Basin, while the impact of sediment type and rates needs to be investigated 409 for all five hyperthermals. In that sense, it thus remains uncertain whether the 410 observed opposite CALMAG changes between PETM and the four post-PETM 411 hyperthermals relate to diametrically opposed precipitation trends or environmental 412 (depositional) trends.

413

414 The precession forcing of the 7-m thick overbank-avulsion sedimentary cycles (Abels 415 et al., 2013) is in line with ~100-kyr and 405-kyr eccentricity forcing of the carbon 416 cycle changes in the ETM2 to I2 stratigraphic interval (Fig. 3). Mudrock intervals 417 with well-developed purple and purple-red paleosols occur predominantly in the 418 eccentricity maxima, while the minima seem to be richer in sand. This could point to 419 a more prolonged relatively stable position of the channel belt on the floodplain, 420 causing less coarse clastic deposition on the floodplains, during eccentricity maxima 421 (Abels et al., 2013). Such an effect could have occurred in combination with or due to

422 more intense pedogenesis under warmer and wetter climates. However, in this 423 interval, the eccentricity-related change is dominated by the hyperthermal events and 424 corroboration of the eccentricity impact is needed from an interval lacking 425 hyperthermals.

426

427 4.2 Marine-terrestrial correlations

428 The benthic carbon isotope record of the I1 and I2 events at Site 1263 reveal very 429 similar patterns as in the bulk and benthic carbon isotope record of Site 1262 (Zachos 430 et al. 2010; Littler et al. 2014) at both eccentricity and precession time scales, as was 431 indicated previously for ETM2 and H2 (Stap et al., 2009). These records even capture 432 very detailed features such as the short-term pre-ETM2 and pre-H2 excursions, and a 433 similar pattern in the I2 excursion. These patterns were clearly driven by changes in 434 the carbon isotope ratio of the atmosphere-ocean exogenic carbon pool as related to 435 precession forcing (Stap et al, 2009).

436

437 Some of these precession-scale details are also captured by the pedogenic carbonate 438 carbon isotope record from the Bighorn Basin suggesting their global nature (Fig. 3). 439 A pre-ETM2 excursion occurs in the McCullough Peaks composite at meter 183, 440 while the shape of the I2 excursion is remarkably similar to the marine records. Main 441 differences on these depth scale plots are the relative expanded CIE intervals and 442 short recovery phases between H1 and H2 and between I1 and I2 in the Bighorn Basin 443 with respect to the Atlantic Ocean records. Sediment accumulation rates were 444 influenced by carbonate dissolution in the events and carbonate overshoot after the 445 events in the marine realm. At the same time, in the Bighorn Basin sedimentation 446 rates might have been higher during the events due to increased sediment budgets and 447 subsequently lower during their recovery phases. These processes might cause the expanded CIEs and contracted recovery phases in the Bighorn Basin with respect to 448 449 the marine records when comparing on depth scale.

450

451 **4.3 Pedogenic carbon isotope excursions**

452 Deciphering the true scale and timing of ocean-atmosphere $\Delta \delta^{13}$ C during 453 hyperthermal events is hampered by environmental impacts on carbon isotope 454 fractionation between marine and terrestrial substrates and their proxies (Sluijs and 455 Dickens, 2012). Our comparison of pedogenic carbonate and marine carbon isotope 456 excursions across the five hyperthermal events shows that although each of the CIEs 457 is amplified in magnitude in the soil carbonate records, the PETM soil carbonate CIE 458 magnitude is anomalously small relative to the pattern of amplification seen for the 459 other events. Use of other marine records in this comparison provides similar results. 460 Changes in photosynthetic ¹³C-discrimination alone cannot explain the anomalously small PETM soil carbonate CIE if we assume that background pCO_2 conditions were 461 462 similar across each of the events (Fig. 5). This mechanism can explain the soil carbonate CIE scaling across the events if there were substantial changes in 463 464 background pCO_2 , but the required changes involve a > 50% decline in pCO_2 from the end of the Paleocene to the Early Eocene. This pattern is not inconsistent with 465 466 independent pCO_2 proxy data from this time interval, but the existing records are too 467 variable and imprecise to provide clear support for or conclusively refute our result 468 (Jagniecki et al. 2015).

469

470 Reconciling the pattern of pCO_2 change inferred in our analysis with known changes 471 in global climate of the early Eocene is more challenging. The dramatic reduction in 472 pCO_2 we estimate following the PETM would be expected to align with a decrease in 473 global temperatures. Although transient cooling has been documented during the ~ 2 474 Myr following the PETM (Wing et al., 1999), temperatures had recovered to at least 475 pre-PETM levels by the time of the ETM2, and thereafter continued to warm toward 476 the peak Cenozoic values of the Early Eocene Climate Optimum (Zachos et al. 2008). 477 Benthic oxygen isotope data of Walvis Ridge, Atlantic Ocean, show a ~1°C increase 478 of deep-sea temperature between PETM and ETM2 baseline values (Littler et al., 479 2014). The substantially lower background pCO_2 values required by our analysis for 480 ETM2 and the subsequent hyperthermals would thus imply that non-CO₂ greenhouse 481 gases or other mechanisms drove long-term global climatic change during the Early 482 Eocene. This is one possible reading of the record of terrestrial CIE amplification 483 across Early Eocene hyperthermals, and suggests that this record may embed valuable 484 information on long-term changes in atmospheric pCO_2 , but it is necessary to 485 acknowledge that the interpretations derived here assume that other local,

environmental influences on the terrestrial CIE magnitudes were similar in nature andproportional to event size across all of the hyperthermals.

488

489 Many other factors may potentially modulate the expression of the global 490 hyperthermal CIEs in the Bighorn Basin pedogenic carbonate records, including 491 changes in temperature effects on carbon isotope fractionation, changes in mixing 492 ratios of atmospheric and organic-derived CO₂ in soils, and changes in vegetation 493 composition (Bowen et al., 2004; Smith et al., 2007). If each of these factors 494 responded primarily to CO₂-driven hyperthermal global change then it is reasonable 495 to assume a proportional, though perhaps non-linear, magnitude of effect across the 496 suite of events. Our data, however, suggest at least one potential forcing factor for 497 these effects, soil moisture, changed in a fundamentally different way during the 498 PETM than during the four younger and smaller hyperthermals (Fig. 2). There is clear 499 indication of soil drying during the PETM based soil development and chemical 500 proxies in line with plant results (Kraus and Riggins, 2007; Kraus et al. 2013). The 501 data presented here for the subsequent ETM2-I2 events show unchanged or slightly 502 increased soil moisture levels.

503

504 Soil moisture, likely reflecting more general changes in local hydroclimate, would be 505 expected to influence the soil carbonate CIE records through changes in gas-phase 506 permeability of the soil matrix (with wetter soils trapping more organic-derived CO₂, leading to lower carbonate δ^{13} C values), influences on ecosystem productivity (with 507 wetter soils supporting higher productivity, soil respiration, and lower $\delta^{13}C_c$), and 508 509 changes in plant photosynthetic discrimination (with greater soil water availability increasing discrimination and reducing $\delta^{13}C_c$; Kohn et al. 2010; Diefendorf et al. 510 511 2010). Soil moisture differences between the PETM and younger hyperthermals could 512 also have led to distinct plant community changes affecting the respective CIEs in 513 pedogenic carbonate (Smith et al. 2007).

514

Evaluating just one of these potential changes, the reconstructed shift in precipitation inferred from PETM proxy data (a reduction in mean annual precipitation from ~1400 mm/year to ~900 mm/year; Kraus et al. 2013; this study) would, based on data documenting modern relationships between precipitation and photosynthetic discrimination (Kohn et al. 2010; Diefendorf et al. 2010), equate to a reduction in 520 plant discrimination (and thus $CIE_{c,PETM}$) of ~0.9 to ~1.2%. Our data suggest that 521 changes in precipitation were negligible during the younger hyperthermals, thus this 522 effect could explain ~1‰ of the observed 5‰ PETM CIE_c anomaly. Clearly this 523 points to the need for a more comprehensive analysis including the effects of 524 discordant local environmental changes on the expression of the global hyperthermal 525 CIEs in soil carbonate records, but it also suggests that in many cases these effect 526 sizes may be modest relative to those arising from pCO_2 -driven changes in 527 photosynthetic discrimination.

528

529 **5** Conclusions

530 We recovered carbon isotope excursions of 2.4 and 1.6% respectively related to the 531 I1 and I2 events in floodplain sedimentary records from the Bighorn Basin, 532 Wyoming. This adds to the three earlier found CIEs, the PETM, ETM2, and H2, 533 underlining the sensitivity of these floodplain records for recording global 534 atmospheric changes. Correlations with marine records and eccentricity-forcing of 535 hyperthermals corroborate the continuity of sedimentation that occurred in the basin 536 starting above precession time scales of ~20 kyr. The 35-m short eccentricity-driven 537 hyperthermal events are in line with precession forcing of the 7-m overbank-avulsion 538 sedimentary cycles. Our CALMAG proxy-based soil moisture estimates reproduce 539 similar or slightly enhanced soil moisture contents for the younger four 540 hyperthermals, in contrast to reconstructions for the PETM. More environmental 541 reconstructions, such as from vegetation, are needed for these four younger 542 hyperthermals in the Bighorn Basin to confirm such a remarkable difference.

543

544 We find that the magnitudes of Bighorn Basin soil carbonate CIEs are linearly 545 proportional to those recorded in benthic marine records for the post-PETM 546 hyperthermals, but that the soil carbonate CIE for the PETM is ~5‰ smaller than expected based on extrapolation of the relationship observed for the other events. We 547 show that the recently characterized dependence of photosynthetic ¹³C-discrimination 548 549 on atmospheric pCO_2 could explain this PETM excursion magnitude 'anomaly', but 550 would require substantially lower background (non-hyperthermal) pCO_2 conditions in 551 the Early Eocene than at the Paleocene-Eocene boundary. That would require 552 reconciliation with globally increasing temperatures during this time interval. Local

553 environmental effects, such as the proxy-inferred reduction of mean annual 554 precipitation during the PETM, likely also modulated the expression of the global 555 hyperthermal CIEs in the Bighorn Basin soil carbonate records. The record of 556 terrestrial carbonate CIE amplification across the sequence of hyperthermals may 557 embed information on million-year changes in early Eocene pCO_2 . However, more 558 like, it records the influence of non-uniform local/regional environmental responses to 559 these events, perhaps reflecting the crossing of discrete climate system or ecological 560 thresholds during the PETM that were not reached during the smaller, subsequent 561 hyperthermals.

562

563 Appendix A.

564 Carbon isotope and soil bulk oxide results for the McCullough Peaks composite 565 section.

566

567 Appendix B.

Figure showing p_{PETM} and change in PETM pCO_2 (Dp_{PETM}) across a range of assumed background pCO₂ conditions from 250 to 3,000 ppmv.

570

571 Acknowledgements

572 H.A. acknowledges NWO-ALW for VENI grant 863.11.006. Will Clyde, Jerry 573 Dickens, Frits Hilgen, Jelmer Laks, and Appy Sluijs are thanked for discussions, 574 Arnold van Dijk, David Ecclestone, Jori Jansen, Sophie van Olst, Christine Satter, and 575 Petra Zaal for laboratory assistance, and the Churchill family of Powell, Wyoming, 576 Peter van den Berg, Francien van den Berg, Matthew Gingerich, Marijn Koopman, 577 Jort Koopmans, Sander Smeets, and Karel Steensma for field assistance. We acknowledge editor Gerald Dickens, and Brian Schubert and an anonymous reviewer 578 579 for their constructive input to the manuscript.

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582 References

- Abels, H.A., W.C. Clyde, P.D. Gingerich, F.J. Hilgen, H.C. Fricke, G.J. Bowen, and
 L.J. Lourens: Terrestrial carbon isotope excursions and biotic change during
 Palaeogene hyperthermals. Nature Geosci. 5, 326-329, 2012.
- Abels, H.A., M.J. Kraus, and P.D. Gingerich: Precession-scale cyclicity in the fluvial
 lower Eocene Willwood Formation of the Bighorn Basin, Wyoming (USA).
 Sedimentology, doi 10.1111/sed.12039, 2013.
- Adams, J.S., M.J. Kraus, and S.L. Wing: Evaluating the use of weathering indices for
 determining mean annual precipitation in the ancient stratigraphic record.
 Palaeogeogr., Palaeocl., Palaeoecol. 309, 358-366, 2011.
- Bowen, G. J., Beerling, D. J., Koch, P. L., Zachos, J. C., and Quattlebaum, T.: A
 humid climate state during the Paleocene-Eocene Thermal Maximum. Nature 432,
 495-499, 2004.
- Bowen, G. J., Koch, P. L., Gingerich, P. D., Norris, R. D., Bains, S. and Corfield, R.
 M.: Refined isotope stratigraphy across the continental Paleocene-Eocene
 boundary on Polecat Bench in the Northern Bighorn Basin *in* Paleocene-Eocene
 stratigraphy and biotic change in the Bighorn and Clark Fork Basins, Wyoming (ed
 Gingerich, P.D.) 73-88 (University of Michigan Papers on Paleontology 33,
 http://hdl.handle.net/2027.42/48328), 2001.
- Bowen, G.J.: Up in smoke: A role for organic carbon feedbacks in Paleogene
 hyperthermals. Global and Planetary Change, 109: 18-29, 2013.
- Bowen, G.J., B.J. Maibauer, M.J. Kraus, U. Röhl, T. Westerhold, A. Steimke, P.D.
 Gingerich, S.L. Wing, and W.C. Clyde: Two massive, rapid releases of carbon
 during the onset of the Palaeocene-Eocene thermal maximum. Nature Geosci. 8,
 44-47, 2015.
- 607 Chen, Z., Z. Ding, Z. Tang, X. Wang, and S. Yang: Early Eocene carbon isotope
 608 excursions: evidence from the terrestrial coal seam in the Fushun Basin, Northeast
 609 China. Geophys. Res. Lett. 41, 3559–3564, doi:10.1002/2014GL059808, 2014.
- 610 Cramer, B.S., J.D. Wright, D.V. Kent, and M.-P. Aubry: Orbital climate forcing of
- 611 d¹³C excursions in the late Paleocene-early Eocene (chrons C24n-C25n).
 612 Paleoceanography 18, 4, 1097, 2003.
- 613 Cui, Y., L.R. Kump, A.J. Ridgwell, A.J. Charles, C.K. Junium, A.F. Diefendorf, K.H.
- 614 Freeman, N.M. Urban, and I.C. Harding: Slow release of fossil carbon during the
- 615 Palaeocene-Eocene Thermal Maximum. Nature Geosci. 4, 481-485, 2011.

- 616 D'Haenens, S., A. Bornemann, P. Claeys, U. Röhl, E. Steurbaut, R.P. Speijer: A
- 617 transient deep-sea circulation switch during Eocene Thermal Maximum 2.618 Paleoceanography 29, 370-388, 2014.
- Dickens, G.R, J.R. O'Neil, D.K. Rea, and R.M. Owen: Dissociation of oceanic
 methane hydrate as a cause of the carbon isotope excursion at the end of the
 Paleocene. Paleoceanography 10, 965-971, 1995.
- 622 Dickens, G.R., M.M. Castillo, J.C. Walker: A blast of gas in the latest Paleocene:
- 623 Simulating first-order effects of massive dissocation of oceanic methane hydrate.624 Geology 25, 259-262, 1997.
- Dickens, G.R.: Methane oxidation during the late Palaeocene thermal maximum.
 Bull.Soc.Geol.France 171, 37-49, 2000.
- Diefendorf, A.F., K.E. Mueller, S.L. Wing, P.L. Koch, and K.H. Freeman: Global
 patterns in leaf 13C discrimination and implications for studies of past and future
 climate. Proc. Nat. Acad. Sciences 107, 5738-5743, 2010.
- Gingerich, P. D: New earliest Wasatchian mammalian fauna from the Eocene of
 northwestern Wyoming: composition and diversity in a rarely sampled highfloodplain assemblage. University of Michigan Papers on Paleontology, 28: 1-97,
 1989.
- Hancock, H. J. L., Dickens, G. R., Strong, C. P., Hollis, C. J. & Field, B. D.:
 Foraminiferal and carbon isotope stratigraphy through the Paleocene-Eocene
 transition at Dee Stream, Marlborough, New Zealand. New Zealand Journ.
 Geology Geophys. 46, 1–19, 2003.
- Hollis, C. J., Dickens, G. R., Field, B. D., Jones, C. M. & Strong, C. P.: The
 Paleocene-Eocene transition at Mead Stream, New Zealand: a southern Pacific
 record of early Cenozoic global change. Palaeogeogr., Palaeocl., Palaeoecol. 215,
 313-343, 2005.
- Jagniecki, E.A., T.K. Lowenstein, D.M. Jenkins & R.V. Demicco: Eocene
 atmospheric CO2 from the nahcolite proxy. Geology 43, 1075-1078, 2015.
- Kiehl J.T., C.A. Shields: Sensitivity of the Palaeocene–Eocene Thermal Maximum
 climate to cloud properties. Phil. Trans. R. Soc. A 371, 20130093, 2013.
- 646 Kirtland Turner, S. & A. Ridgwell: Recovering the true size of an Eocene
 647 hyperthermal from the marine sedimentary record. Paleoceanography 28, 1-13.
 648 Doi. 10.1002/2013PA002541, 2013.

Kraus, M.J., F.A. McInerney, S.L. Wing, R. Secord, A.A. Baczynski, and J.I. Bloch:
Paleohydrologic response to continental warming during the Paleocene-Eocene
Thermal Maximum, Bighorn Basin, Wyoming. Palaeogeogr., Palaeocl.,

652 Palaeoecol. 370, 196-208, 2013

- Kraus, M.J. and S. Riggins: Transient drying during the Paleocene-Eocene Thermal
 Maximum (PETM): Analysis of paleosols in the bighorn basin, Wyoming.
 Palaeogeogr., Palaeocl., Palaeoecol. 245, 444-461, 2007.
- Lauretano, V., K. Littler, M. Polling, J.C. Zachos, and L.J. Lourens: Frequency,
 magnitude and character of hyperthermal events at the onset of the Early Eocene
 Climatic Optimum. Climate of the Past Discussions, in preparation.
- Littler, K., U. Röhl, T. Westerhold, and J.C. Zachos: A high-resolution benthic stableisotope record for the South Atlantic: Implications for orbital-scale changes in Late
 Paleocene–Early Eocene climate and carbon cycling. Earth Planet. Sci. Letters
 401, 18-30, 2014.
- Lourens, L. J., Sluijs, A., Kroon, D., Zachos, J. C., Thomas, E., Röhl, U., Bowles, J.,
 and Raffi, I.: Astronomical pacing of late Palaeocene to early Eocene global
 warming events. Nature 435, 1083-1087, 2005.
- McCarren, H., Thomas, E., Hasegawa, T., Röhl, U., and Zachos, J. C.: Depth
 dependency of the Paleocene–Eocene carbon isotope excursion: Paired benthic and
 terrestrial biomarker records (Ocean Drilling Program Leg 208, Walvis Ridge). *Geochemistry, Geophysics, Geosystems* 9, Q10008, doi: 10.1029/2008GC002116,
- 670 2008.
- McInerney, F.A. and S.L. Wing: The Paleocene-Eocene Thermal Maximum: A
 pertuarbation of carbon cycle, climate, and biosphere with implications for the
 future. Annu. Rev. Earth Planet. Sci. 39, 489-516, 2011.
- Meissner, K.J., T.J. Bralower, K. Alexander, T. Dunkley Jones, W. Sijp, and Ward,
 M.: The Paleocene-Eocene Thermal Maximum: How much carbon is enough?
 Paleoceanography 29, 946-963, 2014.
- Nicolo, M. J., Dickens, G. R., Hollis, C. J. and Zachos, J. C.: Multiple early Eocene
 hyperthermals: their sedimentary expression on the New Zealand continental
 margin and in the deep sea. Geology 35, 699–702, 2007.
- 680 O'Leary, M.H.: Carbon isotopes in photosynthesis. Bioscience, 38: 328-336, 1988.
- 681 Schouten, S., M. Woltering, W. I. C. Rijpstra, A. Sluijs, H. Brinkhuis, and J. S.
- 682 Sinninghe Damsté: The Paleocene–Eocene carbon isotope excursion in higher

- plant organic matter: Differential fractionation of angiosperms and conifers in the
 Arctic. Earth Planet. Sci. Lett. 258(3-4), 581–592, 2007.
- Schubert, B.A. and A.H. Jahren: The effect of atmospheric CO2 concentration on
 carbon isotope fractionation in C3 land plants. Geochimica et Cosmochimica Acta
 96, 29-43, 2012.
- Schubert, B.A. and A.H. Jahren: Reconciliation of marine and terrestrial carbon
 isotope excursions based on changing atmospheric CO2 levels. Nature
 Communications 4: 1653, doi: 10.1038/ncomms2659, 2013.
- 691 Sexton, P.F., R.D. Norris, P.A. Wilson, H. Pälike, T. Westerhold, U. Röhl, C.T.
 692 Bolton, and S. Gibbs. Eocene global warming events driven by ventilation of
 693 oceanic dissolved organic carbon. Nature 471, 349-353, 2011.
- Sluijs, A. and G.R. Dickens: Assessing offsets between the d13C of sedimentary
 components and the global exogenic carbon pool across early Paleogene carbon
 cycle perturbations. Glob. Biogeochem. Cycles 26, GB4005, doi.
 10.1029/2011GB004224, 2012.
- Smith, F.A., S.L. Wing, K.H. Freeman: Magnitude of the carbon isotope excursion at
 the Paleocene-Eocene thermal maximum: the role of plant community change.
 Earth Planet. Sci. Lett. 262, 50-65, 2007.
- Stap, L., Sluijs, A., Thomas, E., and Lourens, L. J.: Patterns and magnitude of deep
 sea carbonate dissolution during Eocene Thermal Maximum 2 and H2, Walvis
 Ridge, southeastern Atlantic Ocean: Paleoceanography 24, PA1211, doi:
 10.1029/2008PA001655, 2009.
- Stap, L., Lourens, L. J., Thomas, E., Sluijs, A., Bohaty, S., and Zachos, J. C.: Highresolution deep-sea carbon and oxygen isotope records of Eocene Thermal
 Maximum 2 and H2. Geology 38, 607-610, 2010a.
- Stap, L., L. Lourens, A. van Dijk, S. Schouten, & E. Thomas: Coherent pattern and
 timing of the carbon isotope excursion and warming during Eocene Thermal
 Maximum 2 as recorded in planktic and benthic foraminifera. Geochemistry,
- 711 Geophysics, Geosystems 11, Q11011, doi. 10.1029/2010GC003097, 2010b.
- Thomas, E.: Development of Cenozoic deep-sea benthic foraminiferal faunas in
 Antarctic waters. In J. A. Crame (ed.), Origins and Evolution of the Antarctic
 Biota, Geological Society Special Publication, 18: 283-296, 1989.

- Uchikawa, J., & R.E. Zeebe: Examining possible effects of seawater pH decline on
 foraminiferal stable isotopes during the Paleocene-Eocene Thermal Maximum.
 Paleoceanography 25, PA2216, doi: 10.1029/2009PA001864, 2010.
- Wing, S.L., Bao, H., Koch, P.L.: An early Eocene cool period? Evidence for
 continental cooling during the warmest part of the Cenozoic. In: Huber, B.T.,
 Macleod, K.G., Wing, S.L. (Eds.), Warm Climates in Earth History. Cambridge
 University Press, Cambridge, UK, pp. 197-237, 1999.
- Zachos, J.C., U. Röhl, S.A. Schellenberg, A. Sluijs, D.A. Hodell, D.C. Kelly, E.
 Thomas, M. Nicolo, I. Raffi, L.J. Lourens, H. McCarren, and D. Kroon: Rapid
 acidification of the ocean during the Paleocene-Eocene Thermal Maximum.
 Science 308, 1611-1615, 2005.
- 726 Zachos, J.C., S.M. Bohaty, C.M. John, H. McCarren, D.C. Kelly, and T. Nielsen: The 727 Palaeocene-Eocene carbon isotope excursion: constraints from individual shell 728 planktonic foraminifer records. Phil. Trans. R. Soc. А 365. doi. 729 10.1098/rsta.2007.2045, 2007.
- Zachos, J. C., Dickens, G. R. & Zeebe, R. E.: An early Cenozoic perspective on
 greenhouse warming and carbon-cycle dynamics. Nature 451, 279-283, 2008.
- Zeebe, R.E., Zachos, J.C., Dickens, G.R.: Carbon dioxide forcing alone insufficient to
 explain Palaeocene-Eocene Thermal Maximum warming. Nature Geosci., 2: 576580, 2009
- 735

736 Tables

737 Table 1. Nomenclature.

a	atmosphere
bkg	background
CIE	carbon isotope excursion
CIE _c	CIE in paleosol carbonate
D	difference
$D\delta$	carbon isotope excursion magnitude
	photosynthetic C-isotope
Δ	discrimination
h	non-PETM hyperthermal
р	pCO ₂
pCO_2	atmospheric CO ₂ pressure
PETM	Paleocene Eocene Thermal Maximum

Table 2. Magnitudes of carbon isotope excursions for five Paleocene-Eocene
hyperthermal events in paleosol carbonate of the Bighorn Basin, Wyoming (USA)
and benthic foraminiferal and bulk sediment carbonate of Walvis Ridge Sites 1263
and 1262, Atlantic Ocean. Standard errors of the differences between detrended
background variability and excursion variability are given (see Methods).

Event	Bighorn Basin CIE ped.carb.	st.error	Bighorn Basin CIE n-alkanes	st.error	Walvis Ridge Sites 1263/65 CIE ben.forams	st.error	Walvis Ridge Site 1262 CIE bulk carb.	st.error
PETM	5.90	0.86	4.23	0.67	3.38	0.12	1.93	0.08
EMT2 / H1	3.78	0.56			1.30	0.18	0.89	0.05
H2	2.75	0.38			0.97	0.16	0.58	0.06
I1	2.42	0.45			0.88	0.16	0.63	0.07
I2	1.55	0.72			0.73	0.16	0.50	0.10

746 Figures



Figure 1. Location map of the sampling sites in the McCullough Peaks area of the
northern Bighorn Basin in northeastern Wyoming (USA). Background colors
denote topography, grey lines are roads. Indicated are the fossil localities and their
interpreted Wasatchian mammal zone and the four study sections. Polecat Bench in
the northwest of the study sites is the location of the PETM.



753

754 Figure 2. Carbon isotope stratigraphies of paleosol carbonate in the McCullough 755 Peaks area, Bighorn Basin, Wyoming (USA). Shown are data from the Upper Deer 756 Creek section of Abels et al. (2012), and the West Branch, Deer Creek 757 Amphitheater, and Creek Star Hill sections. Grey horizontal lines represent field-758 based tracing of marker beds P1, P4, and P8 by which the McCullough Peaks 759 composite carbon isotope stratigraphy has been constructed. To the right, Mean 760 Annual Precipitation reconstructions from the CALMAG methods are given on the 761 McCullough Peaks composite stratigraphy. Different symbols denote different 762 thickness of the soil-B horizons. Note that there is no obvious change in soil 763 moisture during the four hyperthermal events.



764

765 Figure 3. The McCullough Peaks paleosol carbonate carbon isotope stratigraphy 766 compared in depth domain to the bulk sediment and benthic foraminiferal 767 (*Nutallides truempvi*) carbon isotope stratigraphies at, respectively, ODP Site 1262 768 (left y-axis on right side; Zachos et al., 2010) and 1263 (right y-axis on right side; 769 Stap et al. 2010a; this study) at Walvis Ridge in the southern Atlantic Ocean. 770 Filters denote the ~100-kyr eccentricity band in the three records. Note that linear stretching of depth scales is sufficient to construct the figure indicating the 771 772 constant average sedimentation rates at longer time scales in both realms. At 773 smaller time scales, large sedimentation rate differences occur that in the marine 774 realm relate to carbonate dissolution during and carbonate overshoot after the 775 hyperthermal events.



Figure 4. Carbon isotope excursions (CIEs) for the PETM, ETM2, H2, I1, and I2 777 778 events in the early Eocene compared between different proxies in marine and 779 terrestrial settings. Blue squares denote benthic foraminiferal (x-axis) versus bulk 780 sediment (y-axis) CIEs at Walvis Ridge in the Atlantic Ocean. Red squares denote 781 benthic foraminiferal (x-axis) CIEs at Walvis Ridge versus paleosol carbonate (y-782 axis) CIEs in the Bighorn Basin, Wyoming (USA). Trendlines are forced through 783 the origin. Note the apparent reduced CIE for the PETM in paleosol carbonate if 784 extrapolation is used of the trendline through ETM2, H2, I1, and I2.



786

Figure 5. Carbon isotope excursions (CIEs) for the PETM, ETM2, H2, I1, and I2 events in the early Eocene compared between paleosol carbonate (y-axis) CIEs in the Bighorn Basin, Wyoming (USA) and measured and modeled plant CIE for two extreme initial pCO_2 scenarios. The plant CIE for the PETM is measured (Smith et al. 2007), those of the younger four hyperthermals modeled (see text for explanation). Note that the trendlines for both extreme pCO2 scenarios do not fit the measured CIEs in plant and pedogenic carbonate for the PETM.



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Figure 6. Model results of pCO_2 scenarios for the four younger hyperthermals finding a solution for the non-linear scaling of the soil carbonate CIEs relative to the marine record changes in photosynthetic ¹³C-discrimination forced by hyperthermal pCO_2 increase over a varying background pCO_2 conditions. A solution is found across the entire range of assumed PETM background pCO_2 conditions. Note that this requires a >50% decrease in background pCO_2 for most of the post-PETM hyperthermals.