1	ON THE ONSET OF CENTRAL ATLANTIC MAGMATIC PROVINCE (CAMP)
2	VOLCANISM AND ENVIRONMENTAL AND CARBON-CYCLE CHANGE AT THE
3	TRIASSIC-JURASSIC TRANSITION (NEUQUÉN BASIN, ARGENTINA)
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32 Panthalassic marine basins, hypothetically reducing the magnitude of the observed 33 negative CIE in  $\delta^{13}C_{TOC}$ .

34 Increased deposition of sedimentary Hg (and Hg/TOC and Hg/Zr) in the marine 35 Neuquén Basin began stratigraphically before the negative CIE associated with the 36 end-Triassic mass extinction, and thus before the commencement, in North America 37 and Africa, of CAMP-related basaltic volcanism, but possibly coinciding with the early 38 emplacement of CAMP-associated intrusives (dykes and sills). This relative 39 chronology suggests thermal alteration of intruded country rocks and/or intrusive 40 magmatic degassing of Hg as potential major sources of elevated Hg fluxes to the 41 atmosphere at this time. 42 The Neuquén Basin experienced the development of dysoxic-anoxic marine 43 conditions across the Triassic-Jurassic transition, enabling increased preservation of

44 organic matter. Simple mass-balance calculations show that enhanced carbon burial
45 rates can explain the inferred evolution of the global exogenic carbon cycle across

46 this time-interval.

47

48 **Keywords:** Carbon-cycle change, Triassic–Jurassic mass extinction, CAMP

49 volcanism, palaeo-climate change, mercury (Hg), anoxia

50

### 51 **[1] INTRODUCTION**

52 Central Atlantic Magmatic Province (CAMP) volcanism and the associated 53 release of greenhouse gases and toxic compounds has been suggested as the 54 initiator of or driver for major climatic and environmental perturbations at the 55 Triassic–Jurassic transition (e.g. Marzoli et al., 2004; van de Schootbrugge et al., 56 2009; Deenen et al., 2010; Panfili et al., 2019). The release of carbon into the ocean-57 atmosphere system likely led to significantly elevated atmospheric  $pCO_2$  levels 58 (McElwain et al., 1999; Bonis et al., 2010; Schaller et al., 2011; Steinthorsdottir et al., 59 2011; Schaller et al., 2012; Schaller et al., 2015) and an associated increase in

60 global temperatures in the oceans and atmosphere (Korte et al., 2009; McElwain et 61 al., 1999; Ruhl et al., 2010; Bonis and Kürschner, 2012). The climatic and 62 environmental repercussions of Triassic–Jurassic carbon release are suggested to 63 have led to the end-Triassic mass extinction in the marine realm, with the loss of 64 most ammonoid and all conodont species and major extinctions in many other 65 marine invertebrate groups (e.g. Mander et al., 2008; Mander and Twitchett, 2008; 66 Hillebrandt et al., 2013, and references therein). Terrestrial ecosystems were also 67 severely affected, with major changes in vegetation (e.g. van de Schootbrugge et al., 68 2009; Mander et al., 2010; Bonis and Kürschner, 2012). The principle processes 69 leading to marine extinctions probably varied geographically, through time, and 70 between faunal groups, but enhanced atmospheric  $pCO_2$  levels likely led to the 71 Triassic–Jurassic transition being one of the strongest ocean acidification events of 72 the Phanerozoic Eon (Črne et al., 2011; Greene et al., 2012; Hönisch et al., 2012; 73 Ikeda et al., 2015).

74 Carbon release from the lithosphere to the atmosphere and ocean, associated 75 with CAMP magmatism, and the possible initiation of positive and negative feedback 76 mechanisms in the global carbon-cycle, also resulted in major changes in global 77 (bio)geochemical cycling, manifested as a series of negative and positive excursions 78 in organic and inorganic marine and terrestrial carbon-isotope records. The release 79 of <sup>13</sup>C-depleted carbon from CAMP volcanogenic degassing, possibly combined with 80 the release of bio- and thermogenic <sup>13</sup>C-depleted carbon from the dissociation of 81 seafloor methane clathrates and thermally altered subsurface organic-rich shales 82 intruded by dykes and sills, likely contributed to or caused the observed negative 83 carbon-isotope excursion (CIE) coincident with the end-Triassic mass extinction 84 (Hesselbo et al., 2002; Ruhl et al., 2009; Ruhl et al., 2011). The magnitude of this 85 negative CIE is poorly constrained and varies between 1 and 8‰ in marine and 86 continental (terrestrial and lacustrine) organic and inorganic bulk-rock and molecular 87 biomarker archives (Pálfy et al., 2001; Hesselbo et al., 2002; Galli et al., 2005; van

88 de Schootbrugge et al., 2008; Korte et al., 2009; Ruhl et al., 2009; Deenen et al., 89 2010; Ruhl et al., 2010; Whiteside et al., 2010; Črne et al., 2011; Deenen et al., 90 2011; Ruhl et al., 2011; Bartolini et al., 2012; Lindström et al., 2012; Dal Corso et al., 91 2014; Al-Suwaidi et al., 2016; Yager et al., 2017). The uncertainty in the size of the 92 observed CIE and thus the associated carbon-cycle change at this time severely 93 hampers mass-balance constraints on the magnitude and rate of carbon release 94 associated with the onset of CAMP emplacement, and related carbon release from 95 e.g. ocean-floor methane hydrate reservoirs or intruded subsurface organic-rich 96 shales, and the associated changes in the fluxes of carbon between the exogenic 97 carbon reservoirs.

98 Here, determinations of newly collected ammonites, bivalves and brachiopods 99 have allowed improved stratigraphic resolution of an expanded Triassic–Jurassic 100 boundary section in the Neuguén Basin, Argentina, and are accompanied by new 101  $\delta^{13}C_{TOC}$ , Hg (and Hg/TOC and Hg/Zr) and elemental concentration data. We show 102 that the magnitude of the observed negative CIE over the interval of the end-Triassic 103 mass extinction varies geographically, with only minor 2-3‰ negative excursions in bulk  $\delta^{13}C_{TOC}$  observed in marine basins along the western Pangaean continental 104 105 margin, bordering the eastern Panthalassic Ocean (Figure 1). This magnitude 106 change is broadly similar to the 2–4‰ negative CIEs in bulk  $\delta^{13}C_{TOC}$  observed in 107 North American and Moroccan continental sections, but it is significantly smaller than 108 the 4–6.5‰ negative CIEs observed in bulk  $\delta^{13}C_{TOC}$  records from the Laurasian 109 Seaway (Figure 1). We hence here provide a stratigraphic framework for the onset of 110 CAMP magmatic activity and discuss associated environmental consequences with 111 implications for the end-Triassic mass extinction and the magnitude of the associated 112 end-Triassic carbon-cycle perturbation.

113

# 114 [2] PALAEOGEOGRAPHY, GEOLOGICAL SETTING AND SEDIMENTARY

### 115 **DEPOSITIONAL ENVIRONMENT**

116 The Neuquén Basin is located in west central Argentina, on the eastern side of the Andes, between 34° and 41° S and covers an area of about 120,000 km<sup>2</sup> (Figure 117 118 1; Supplementary Figure 1; Howell et al., 2005). The basin has a complicated 119 tectonic history associated with the break-up of Gondwana, subduction of the proto-120 Pacific plate and the development of the Andean magmatic arc (Vergani et al., 1995, 121 and references therein). It began as a rift basin during Late Triassic to Early Jurassic 122 times and its tectonic evolution was characterized by about 220 Myr of Mesozoic-123 Cenozoic subsidence (Vergani et al., 1995; Bechis et al., 2010, and references 124 therein). Initial rifting in the Triassic was driven by extensional collapse of a Permo-125 Triassic orogenic belt and early syn-rift sequences were restricted to isolated 126 depocentres limited by normal faults (Vergani et al., 1995). The location and 127 geographical extent of the main depocentre evolved significantly through the 128 Jurassic, associated with changes in accommodation space and sediment supply in 129 response to eustatic sea-level change and tectonic processes (Vicente, 2005; 130 Vicente, 2006; Lanés et al., 2008; Riccardi et al., 2011). From the Aalenian (Middle 131 Jurassic) onward, fault-controlled subsidence was replaced by regional subsidence, 132 but several episodes of structural inversion modified the shape of the depocentre and 133 rejuvenated sedimentary source areas. The tectonic evolution of the basin is 134 reflected in a complex structural and depositional framework, and resulted in the 135 formation of several major hydrocarbon source-rock intervals, and numerous 136 reservoir zones (Vergani et al., 1995). 137 The north-south oriented Neuquén Basin contains up to 6 km of Triassic to 138 Cenozoic sediments in its most central part, which accumulated in several 139 depositional cycles representing a pre-rift phase through to foreland-basin 140 development (Howell et al., 2005). The sedimentary succession of the Atuel

- 141 depocentre studied here is situated in the northern Andean sector of the northern
- 142 Neuquén Basin (Supplementary Figure 1) and formed in the Late Triassic (Norian),

with marine sedimentary infill recording the first stages of the basin opening as a riftsystem (Bechis et al., 2010, and references therein).

145 The Triassic to Jurassic infill of the Atuel depocentre consists of siliciclastic 146 marine and continental sediments of the Arroyo Malo, El Freno, Puesto Araya and 147 Tres Esquinas Formations (Riccardi et al., 1991; Lanés, 2005; Lanés et al., 2008; 148 Bechis et al., 2010). Accommodation space vs sediment supply varied through time, 149 with syn-rift depositional conditions occurring from Rhaetian to late Early Sinemurian 150 times (marked by accommodation space >> sediment supply; Lanés, 2005). At this 151 time sediments were deposited in slope-type fan deltas in the west, and coeval 152 braided fluvial systems in the east (Lanés, 2005; Lanés et al., 2008), while active 153 normal faults controlled the palaeogeography and lateral variations of the 154 depositional systems (Bechis et al., 2010).

The Early Jurassic syn-rift deposition gave way to a sag phase in Sinemurian-155 156 Toarcian times, with accommodation space exceeding sediment supply, allowing for 157 a marked increase in the marine depositional area, a transgression of the marine 158 shelf and a deepening of the depositional environment. Marine syn-rift sediments are 159 up to ~980 m thick in the western part of the Atuel depocentre, with continental 160 deposits forming, thinning out and disappearing towards the east (Bechis et al., 161 2010). In the Neogene, the Andean orogeny deformed and uplifted the depocentre 162 infill, leading to exhumation of the syn-rift deposits and their exposure in the northern 163 sector of the Malargüe fold and thrust belt (Bechis et al., 2010, and references 164 therein).

The Upper Triassic and Lower Jurassic marine sediments of the Arroyo Malo Formation are interpreted as mainly consisting of fan-delta facies with planar laminated mudstones, fine sandstones and conglomeratic lenses deposited by lowand high-density turbidity currents and debris flows (Lanés, 2005; Lanés et al., 2008). Sediments may have entered the Neuquén Basin from two main source areas: the Chilean Coastal Cordillera that supplied immature volcaniclastic material

and cratonic areas to the east that introduced more mature material (Tunik et al.,

172 2008).

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## 174 [3] MATERIALS & METHODS

175 The Triassic–Jurassic marine sedimentary succession at Arroyo Alumbre is 176 remotely located in the Andes mountains, west of the main road from Malargüe to Mendoza (leaving the main road at El Sosneado), at 34°49'767"S – 69°53'224"W 177 178 (Google-maps coordinates: -34.827675 (South), -69.888884 (West)) (Supplementary 179 Materials and Supplementary Materials Figures 2 A–E). The section was sampled 180 during two field seasons (2014 and 2016). The Arroyo Alumbre (and associated 181 nearby Arroyo Malo) sections were previously studied and described by Riccardi et 182 al. (1988, 1997, 2004), Lanés (2005), Lanés et al. (2008), Damborenea and 183 Manceñido (2012), and Damborenea et al. (2017), who distinguished the Arroyo 184 Malo Formation at the base (0–280 m), followed by the conglomeratic lenses of the 185 El Freno Formation and the stratigraphically higher deposits of the El Cholo 186 Formation. The Triassic–Jurassic sediments of the Arroyo Alumbre/ Arroyo Malo 187 section were deposited in the northern sector of the Neuquén Basin, in the Atuel 188 depocentre (Lanés, 2005; Lanés et al., 2008; Bechis et al., 2010; Damborenea et al., 189 2017). The Triassic–Jurassic samples geochemically studied here are from ~124– 190 271 m in the Arroyo Alumbre section, and all come from the Arroyo Malo Formation 191 (Figure 2). The Triassic–Jurassic sedimentary succession at Arroyo Alumbre is 192 characterized by the monotonous occurrence of dark-coloured shales and 193 mudstones, apart from sporadic turbiditic or debris flow beds (1–10cm thick; 194 consisting of fine-grained sands to coarse-grained silts). The rare gravity flows were 195 avoided when sampling of the studied stratigraphic section, with collected samples 196 all consisting of shales and mudstones (Supplementary Figure 2 D–E). The 197 monotonous dominant lithology in the samples collected, is exemplified by relatively 198 unchanging sedimentary zirconium (Zr) concentrations (Figure 2).

Ammonite, bivalve and brachiopod samples were collected for

200 biostratigraphical constraints and published in Damborenea et al. (2017) and

- 201 Riccardi (2019). Bulk samples were collected and analysed for chemostratigraphy
- 202 ( $\delta^{13}C_{TOC}$ ), organic carbon and pyrolysis (Rock-Eval) analyses, mercury analyses
- 203 (suggested previously to reconstruct CAMP-activity; Percival et al., 2017), and
- 204 elemental/ palaeoenvironmental study (Hand-Held X-Ray Fluorescence (hh-XRF)).
- 205 Detailed description of methods is provided in the on-line Supplementary Information206 section.
- 207

# 208 [4] RESULTS & DISCUSSION

# 209 [4.1] INTEGRATED BIO- & CHEMOSTRATIGRAPHY OF THE TRIASSIC-

# 210 JURASSIC TRANSITION AT ARROYO ALUMBRE/ ARROYO MALO

211 The studied Triassic–Jurassic succession at Arroyo Alumbre/ Arroyo Malo

212 (Neuquén Basin, Argentina) contains relatively diverse marine invertebrate

213 macrofossils, such as ammonites, bivalves and brachiopods (Figure 2 and 3;

Riccardi et al., 2004; Damborenea and Manceñido, 2012 and references therein;

215 Damborenea et al., 2017; Riccardi, 2019).

216 The presence of marine Triassic strata in Argentina was initially proposed on

217 the basis of bivalves and brachiopods found in marine deposits near Cerro Chachil

and near Piedra del Águila (both in the Neuquén Province; Groeber, 1924, 1929;

219 Groeber et al., 1953). These fossils, however, were later recognized as Early

Jurassic in age (Leanza, 1948; Frenguelli, 1948; Leanza, 1992; Manceñido and

Leanza, 1993; Pérez et al., 1995). Upper Triassic (Norian and/or Rhaetian) marine

fossils (mollusc and brachiopod faunas) were subsequently reported from Arroyo

223 Malo, in the river Atuel area of the Southern Mendoza province, in deposits below the

- lower Hettangian sediments that had a fauna pertaining to the *Psiloceras*
- *rectocostatum* Zone (Riccardi et al., 1991; Riccardi et al., 1997; Riccardi et al., 2004;
- 226 Riccardi, 2008; Manceñido, 2002; Damborenea and Manceñido, 2012; Riccardi,

227 2019). A late Rhaetian age for these deposits was confirmed by the occurrence of 228 Choristoceras cf. marshi (Riccardi and Iglesia Llanos, 1999; Riccardi, 2008, 2019). 229 Upper Triassic and Lower Jurassic marine faunas described for the Arroyo Malo/ 230 Arroyo Alumbre sections were found throughout the succession (Figure 2; 231 Damborenea & Manceñido, 2012; Damborenea et al., 2017; Riccardi, 2019). 232 Bivalves are the most numerous and diverse group, with at least 15 species (and 233 almost as many families; Damborenea and Manceñido, 2012; Damborenea et al., 234 2017). Apart from the bivalves and brachiopods, this fauna also includes Triassic and 235 Jurassic ammonoids, three gastropod species, a cnidarian, and some microfossils 236 (foraminifers and ostracods) (Damborenea and Manceñido, 2012). A few 237 impressions of plant remains are also present (suggested to be of Zuberia and 238 Equisetales affinity amongst others (Riccardi et al., 1997, 2004; Damborenea and 239 Manceñido, 2012).

240 New ammonite finds were recently described in Riccardi (2019), as part of a 241 wider review of cephalopod fossils from this section. Triassic cephalopods were 242 found *in situ* in the lower part of the section (at 2–73 m), including the heteromorph 243 ammonoids Rhabdoceras suessi, ?Peripleurites and the coleoid Aulacoceras cf. 244 carlottense, suggesting a late Norian age. An ex situ specimen of the latest Rhaetian 245 Choristoceras marshi was found at ~135 m in the section (Riccardi, 2019). Given the 246 steep nature of the outcrop, and the non-overturned nature of the bedding, this 247 specimen likely originated from this level or from higher up the succession; this is 248 consistent with the *in situ* occurrence of Rhaetian nannofossils up to ~10m above this 249 level (Angelozzi personal communication).

The *ex situ* find of the earliest Jurassic ammonite *Psiloceras tilmanni* at ~176 m
suggests the occurrence of the *tilmanni* Subzone of the *tilmanni* Zone
stratigraphically at or above this level. The *in situ* record of *Psiloceras* cf. *primocostatum* and *P.* cf. *polymorphum*, and the *ex situ* record of *P.* cf. *pressum*

between 188–197 m are indicative of the *planocostatum* Subzone of the *tilmanni* 

Zone, and/or the *primocostatum* Zone of the Andean zonation (or the *polymorphum*Zone in the North American zonation; Guex, 1995). The *in situ* record of *Psiloceras rectocostatum* at ~230 m in the section indicates the *rectocostatum* Zone. The
Andean *primocostatum* and *rectocostatum* (and the North American *polymorphum*)
zones all temporally overlap with the European *planorbis* Zone (Riccardi, 2008; von
Hillebrandt et al., 2013).

The  $\delta^{13}C_{TOC}$  record presented here across the Triassic–Jurassic transition at 261 262 Arroyo Malo starts at ~124 m, extending to ~271 m (Figure 2). The observed values in  $\delta^{13}C_{TOC}$  vary between ~ -26.5‰ and ~ -29.2‰ and the record is marked by 263 264 significant and relatively abrupt ~  $1-2\infty$  fluctuations superimposed on 265 stratigraphically more extensive trends (Figure 2). The lowest Jurassic ammonites 266 recorded in situ, occur ~9 m above a distinct negative carbon-isotope excursion 267 (CIE). The position of this negative CIE, relative to the ammonite record, combined with the more stratigraphically extensive trends in  $\delta^{13}C_{TOC}$ , is remarkably similar to 268 269 what is observed for other Triassic–Jurassic boundary sections, such as at St 270 Audries Bay (Bristol Channel Basin, UK) or New York Canyon (Nevada, USA) 271 (Figure 3; Hesselbo et al., 2002; Hesselbo et al., 2004; Bartolini et al., 2012). The 272 record at Arroyo Malo is, however, 4-5 times more expanded compared to St 273 Audries Bay and the changes in magnitude of  $\delta^{13}C_{TOC}$  are generally smaller. The 274 integrated bio- and chemostratigraphical framework for the Arroyo Malo section 275 presented here allows for direct high-resolution correlation with Triassic-Jurassic 276 records elsewhere (Figure 3).

277

#### 278 [4.2] CAMP MAGMATISM RECORDED IN THE NEUQUÉN BASIN (ARGENTINA)

279 Mercury (Hg) analyses are increasingly being used to constrain the

280 stratigraphical occurrence of large igneous province (LIP) volcanism in the

281 Phanerozoic history of the planet (Percival et al., 2018 and references therein).

282 Gaseous mercury has a relatively short residence time in the present-day

283 atmosphere, of 0.5–2 years (Schroeder and Munthe, 1998; Blum et al., 2014; Pyle 284 and Mather, 2003). The element can be sequestered into marine and terrestrial 285 sediments through several pathways, but a major route into the sedimentary realm is 286 after uptake by and deposition with organic matter (for which reason sedimentary Hg 287 concentrations are commonly normalized against the sedimentary total organic 288 carbon concentration (Hg/TOC)). Despite its promise, numerous important questions 289 remain to be answered before Hg can be claimed as an unequivocal fingerprint of 290 LIP volcanism, as well as an understanding of why some sedimentary records 291 document clear Hg enrichment signals whilst others do not. Of particular importance 292 is evaluating the impact of different volcanic styles on the global mercury cycle, as 293 well as the role played by depositional processes in recording global Hg-cycle 294 perturbations (Percival et al., 2018 and references therein).

295 Nevertheless, evidence from multiple sections suggests that Central Atlantic 296 Magmatic Province volcanism likely had a major impact on the global mercury cycle, 297 and relatively elevated sedimentary Hg and Hg/TOC values are reported from 298 geographically widespread continental and marine sedimentary basins in the USA 299 (Nevada), Canada (Partridge Island), East Greenland (Jameson Land), Austria 300 (Eiberg Basin), the UK (Bristol Channel Basin), and Morocco (Argana Basin) (Figure 301 1; Thibodeau et al., 2016; Percival et al., 2017). Accumulation of enhanced 302 sedimentary Hg at the Triassic–Jurassic transition appears to have begun close to 303 the time of the end-Triassic mass extinction, contemporaneously with the initial 304 emplacement of CAMP basalts in the Argana Basin and the High Atlas Mountains 305 (Morocco). Subsequent uppermost Triassic and Lower Jurassic peaks in mercury 306 levels appear to be pulsed, and are possibly aligned, age-wise, with the major CAMP 307 basalt flows as reported from North America and Morocco (Percival et al., 2017). 308 Mercury (Hg) analyses on the expanded Upper Triassic to Lower Jurassic 309 Arroyo Alumbre sedimentary succession in the Neuquén Basin show a relative 310 increase in Hg concentrations from on average ~10 ppb below ~169m in the studied

311 section, increasing up to ~60 ppb at ~189 m (Figure 2). As stated above, Hg 312 drawdown into marine sediments can be controlled by variations in the organic-313 matter flux to the sediment-water interface; sedimentary Hg concentrations are 314 therefore commonly normalized against TOC (Percival et al., 2015). Also Hg/TOC 315 values in the studied succession (in samples with TOC > 0.25%; lower TOC values 316 are impacted by increasingly large analytical uncertainties) increase from ~169m 317 upward, suggesting an increased Hg-flux to the sediment-water interface, 318 independent of sedimentary organic-matter enrichment (Figure 2).

319 Sedimentary Hg concentrations could alternatively have resulted from 320 changes in the depositional environment and associated changes in sediments 321 supplied, and the rate of sedimentation. However, all studied samples represent a 322 monotonous shale or mudstone lithology, void of major stratigraphic changes as 323 exemplified by relatively constant sedimentary Zr concentrations (Figure 2). A major 324 increase in Hg/Zr values, onsetting at ~169 m in the studied succession, therefore 325 suggests that lithology plays no major role in the stratigraphical evolution of 326 sedimentary Hg concentrations (Figure 2). The data-set presented here suggests 327 that elevated sedimentary Hg (and Hg/TOC & Hg/Zr) values initiated before the 328 negative CIE associated with the end-Triassic mass extinction (Figure 2).

329 Mercury in the present day not only released from volcanogenic degassing on 330 land or along mid-ocean ridges (Pyle and Mather, 2003; Bowman et al., 2015), but 331 also by the thermogenic alteration of subsurface organic-rich deposits (O'Keefe et 332 al., 2010). CAMP-associated dykes and sills intruded sedimentary basins in South 333 America and western Africa up to 100 kyr predating the earliest known age of CAMP 334 basaltic extrusion (Davies et al., 2017; Heimdal et al., 2018). Hence, elevated Hg 335 loading (leading to increased sedimentary Hg, Hg/TOC and Hg/Zr levels) at a time 336 apparently before the negative CIE associated with the end-Triassic mass extinction, 337 may suggest that mercury was not only released into the atmosphere through direct 338 volcanogenic degassing, but possibly also expelled via intrusions of CAMP-

associated dykes and sills into subsurface organic-rich shales (Figures 2 and 6;
Percival et al., 2015; Jones et al., 2019). Volcanic Hg measurements from fumaroles
and diffuse soil emissions associated with CO<sub>2</sub> (rather than SO<sub>2</sub>) degassing (e.g.,
Bagnato et al., 2014) also suggest that some degree of magmatic Hg degassing
within the crust, associated with intrusive (rather than purely extrusive) magmatism,
may also play a role.

Observed peak Hg and Hg/TOC values of ~50 ppb and ~150 ppb/%, 345 346 respectively, in Arroyo Alumbre are, however, lower than those observed in 347 contemporaneous sediments from New York Canyon and Greenland (Figure 1; 348 Thibodeau et al., 2016; Percival et al., 2017), which might reflect (I) higher sediment 349 accumulation rates (albeit without a significant lithological change) in the Neuquén 350 Basin at the time (and associated dilution of the Hg signal), (II) a relatively small 351 contribution of terrestrially sourced organic matter to the basin, with an associated 352 lower Hg-shuttling capacity (Them et al., 2017), and/or (III) the geographically closer 353 and/or downwind position of the Greenland and New York Canyon localities relative 354 to CAMP igneous centres (Figure 1).

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## 356 [4.3] MAGNITUDE OF THE END-TRIASSIC CARBON-CYCLE PERTURBATION

357 The observed negative carbon-isotope excursion (CIE) at the end-Triassic 358 mass extinction directly predated the onset of CAMP basalt eruptive flows in the 359 Fundy, Newark and Hartford Basins (North America; Cirilli et al., 2009; Whiteside et 360 al., 2010), but coincided with the onset of CAMP basalt emplacement in the Argana 361 Basin and High Atlas Mountains (Morocco; Deenen et al., 2010; Dal Corso et al., 362 2014; see also a review by Korte et al., 2019 and references therein; Panfili et al., 363 2019). The onset of eruption of the major CAMP basalt flows at the time of the end-364 Triassic mass extinction was, however, likely preceded by intrusions of major dykes 365 and sills, feeding the oldest basalt extrusives, up to ~100 kyr earlier (Marzoli et al., 366 2011; Dal Corso et al., 2014; Davies et al., 2017; Panfili et al., 2019).

367 Carbon-cycle evolution during the end-Triassic mass extinction and across the 368 Triassic–Jurassic transition has in recent years been intensely studied in marginal 369 marine and continental basins across Asia, northern Africa, Europe and North and 370 South America (see Schobben et al., 2019 and references therein). The magnitude 371 of the  $\delta^{13}C_{TOC}$  negative CIE at the level of the end-Triassic mass extinction generally 372 varies between -4 and -6.5‰ in European marine basins. Carbon-isotope records of 373 marine skeletal material ( $\delta^{13}C_{Calcite}$ ) do not exist across the whole span of this time-374 interval, possibly partly due to extinction and/or reduced preservation of calcite-375 secreting organisms under low pH conditions (Črne et al., 2011; Greene et al., 2012; 376 Hönisch et al., 2012; Ikeda et al., 2015). Many bulk carbonate carbon-isotope 377  $(\delta^{13}C_{CARB})$  records across Europe and the north-western Tethys, however, show only 378 a much smaller 1–3‰ negative CIE (Clémence et al., 2010; Črne et al., 2011; Korte 379 and Kozur, 2011; Felber et al., 2015). Similarly, bulk carbonate carbon-isotope  $(\delta^{13}C_{CARB})$  records in the eastern Tethys (Tibet and the Qiangtang Basin, China) 380 show a 1.5–4‰ negative shift (Yin et al., 2006; Hu et al., 2020). Some  $\delta^{13}C_{CARB}$ 381 382 records, however, show a much larger negative shift, of 6-8‰, at the level of the 383 end-Triassic mass extinction, but these localities are notably marked by the effects of 384 diagenetic carbonate precipitation and/or cementation (central and northern Italy: 385 Bachan et al. (2012), Bachan and Payne (2015); Doniford, Somerset, UK: Clémence 386 et al. (2010)), and/or carbonate deposition in a relatively restricted basin (such as 387 recorded in the Csővár section, Hungary; Pálfy et al., 2001), and/or carbonate 388 deposition in a local calcium-rich lagoonal setting (such as the Wadi Milaha and Wadi 389 Niqab sections of the United Arab Emirates; Al-Suwaidi et al., 2016; Ge et al., 2018). 390 Interestingly, the latter two sections in the United Arab Emirates, as well as the Csővár section in Hungary only show a minor 2–3‰ negative CIE in  $\delta^{13}C_{TOC}$  (Figure 391 392 1; Pálfy et al., 2001; Ge et al., 2018).

393 The expanded Arroyo Alumbre section studied here also shows only a minor 394 2‰ negative CIE in  $\delta^{13}C_{TOC}$  across the level of the end-Triassic mass extinction

395 (Figure 2). Importantly, other successions from marine basins bordering the western 396 Pangaean continental margin (along the eastern Panthalassic Ocean; Figure 1), are 397 also marked by only a minor 2–3‰ negative CIE over this interval (Figures 1 and 4). 398 This distinct palaeogeographic disparity between the magnitude of the  $\delta^{13}C_{TOC}$ 399 negative CIE between the north-western Tethys Ocean and the Eastern Panthalassic 400 marine basins may give crucial clues concerning regional Triassic–Jurassic climatic 401 and environmental conditions.

402 The Triassic–Jurassic transition was possibly marked by a tripling to 403 quadrupling of atmospheric  $pCO_2$  concentrations, based on stomatal density analyses and pedogenic carbonate  $\delta^{13}$ C (McElwain et al., 1999; Schaller et al., 2011, 404 405 2012; Steinthorsdotttir et al., 2011). Elevated and increasing atmospheric  $pCO_2$ 406 concentrations linked to CAMP-associated greenhouse gas release likely had a 407 profound impact on global climate, primarily on temperature and precipitation, with 408 significant warming and possible decreased humidity in some coastal marine regions 409 and continental interiors, respectively (Korte et al., 2009; Bonis et al., 2009; Bonis et 410 al., 2010; Ruhl et al., 2010; Bonis and Kürschner, 2012; Steinthorsdottir et al., 2012). 411 Triassic–Jurassic continental configuration, with the Pangaean supercontinent 412 spanning both hemispheres, would have affected regional climatic and environmental 413 conditions (Kent et al., 2014). This continental configuration, combined with elevated 414 atmospheric pCO<sub>2</sub> and average global temperatures at that time, likely prevented 415 effective penetration of humidity and precipitation into the vast continental interior, 416 leading to strongly reduced rainfall (and elevated evaporation over precipitation 417 rates) across the central and western Pangaean landmass (Figure 1; Chandler et al., 418 1992; Huynh and Poulsen, 2005). Extremely warm and dry conditions likely affected 419 western Pangaean continental interiors between the latitudes of 60° north and south 420 (Chandler et al., 1992; Huynh and Poulsen, 2005), possibly in response to poleward 421 expansion of the Hadley cells and associated subtropical climate belts (Figure 1), as

422 also predicted by models for anthropogenic greenhouse-gas warming (Hu and Fu,

423 2007; Lu et al., 2007; Previdi and Liepert, 2007; Kang and Lu, 2012).

424 The climatic effect of the Pangaean continental configuration, combined with 425 elevated Triassic–Jurassic greenhouse conditions, may have profoundly impacted 426 the occurrence and dominant type of vegetation across the central and western part 427 of the landmass, with the mid-latitudes possibly largely characterized by desert-like 428 conditions, with very limited to no vegetation and more temperate vegetation-types 429 only occurring at higher latitudes (>60° N and S; Rees et al., 2000; Sellwood and 430 Valdes, 2006, 2008). The modelled reduced to non-existent vegetative cover across 431 central and western Pangaea would likely have affected weathering rates, sediment 432 supply, and the supply of terrestrial organic matter to marginal marine basins.

433 The atmospheric flux of humidity from the oceans into continental interiors, and 434 associated precipitation and plant growth are, however, likely to have been much 435 more common along the eastern margin of Pangaea, with wetter conditions and lush 436 vegetation along most of the north-western Tethyan realm (Rees et al., 2000; Barrón 437 et al., 2006; Sellwood and Valdes, 2006, 2008; Gómez et al., 2007; McElwain et al., 438 2007; Bonis et al., 2009; Cirilli et al., 2009; van de Schootbrugge et al., 2009; Bonis 439 and Kürschner, 2012; Lindström et al., 2012; Pieńkowski et al., 2014; Cirilli et al., 440 2018).

Osmium-isotope ratios are significantly elevated across the Triassic–Jurassic transition (Cohen and Coe, 2007; Kuroda et al., 2010), reflecting a relative increase in the proportion of radiogenic isotopic species, suggesting overall enhanced continental chemical weathering rates through this time-interval, likely in response to elevated and enhanced atmospheric  $pCO_2$  and hypothesised accelerated hydrological cycling around eastern Pangaea.

447 Marginal marine basins along the sparsely vegetated western edges of the
448 Pangaean landmass have significantly expanded sedimentary successions across
449 the Triassic–Jurassic transition, with the negative CIE at the end-Triassic mass

450 extinction generally expressed over a stratigraphic thickness of 4–6 metres (Figure 3; 451 this study; Williford et al., 2007; Bartolini et al., 2012; Thibodeau et al., 2016; Yager 452 et al., 2017). By contrast, and despite elevated precipitation and weathering levels, 453 the stratigraphic thickness recording the negative CIE associated with the end-454 Triassic mass extinction in most of the north-western Tethyan marginal marine 455 basins, which were surrounded by well-vegetated coastal regions and hinterlands, is 456 much more limited and generally only 30-80cm thick in siliciclastic-dominated 457 successions (Hesselbo et al., 2002; Ruhl et al., 2009; Lindström et al., 2012; Felber 458 et al., 2015). This pattern suggests that, in addition to a potentially larger 459 accommodation space in the back-arc basins of western Pangaea (or possible 460 depositional gaps or hiatuses in European sections), vegetation cover in eastern 461 Pangaea may have played a role in stabilizing soil profiles, controlling sediment 462 budgets and the supply of sediments to the marine realm at that time.

The prolonged Late Triassic and Early Jurassic potential scarcity in vegetation cover across western Pangaea (Figure 1; Sellwood and Valdes, 2006; 2008) likely resulted in only limited terrestrial organic-matter supply to nearby marginal marine basins. In contrast to the Tethyan realm, sedimentary organic matter in these basins may therefore have been largely marine in origin, before, during, and after the negative CIE associated with the end-Triassic mass extinction.

469 Marine and terrestrial organic matter generally has a significant 4–5‰ 470 difference in its stable carbon-isotope composition (Figure 4 and 5). This difference is 471 exemplified by the 3–5‰ more positive Early Jurassic  $\delta^{13}$ C values of long-chain (C<sub>31</sub>

472 to C<sub>35</sub>) *n*-alkane molecular biomarkers from terrestrial higher plant leaf-waxes, *vs* 

473 marine/aquatic algal-sourced short-chain (C<sub>17</sub> to C<sub>21</sub>) *n*-alkanes (e.g. Xu et al.,

474 2017*a*), including across the end-Triassic mass extinction interval in the Eiberg

475 Basin, Austria (Figure 4; Ruhl et al., 2011). Mixing, or a changing relative supply, of

476 marine and terrestrial organic matter into sediments can therefore impact the bulk

477 organic carbon-isotope ( $\delta^{13}C_{TOC}$ ) composition of such sediments (Suan et al., 2015).

478 Although such mixing by no means entirely explains observed perturbations in  $\delta^{13}C_{TOC}$  across major global change events (Xu et al., 2017*a*), it can, however, affect 479 480 the magnitude of such excursions (Suan et al., 2015). Triassic-Jurassic palynofacies, hydrogen index (HI) and nitrogen- ( $\delta^{15}N$ ) isotope data from marine 481 482 basins in the northwestern Tethys realm do indeed show significant changes in origin 483 of the sedimentary organic matter and kerogen (Bonis et al., 2009; Clémence et al., 484 2010; Paris et al., 2010; Ruhl et al., 2010; Bonis and Kürschner, 2012; Lindström et 485 al., 2012). Hydrogen indices can unfortunately, however, not be analysed for the 486 studied sedimentary succession at Arroyo Alumbre, likely due to a high level of 487 thermal maturation of the sediments.

488 The  $\delta^{13}C_{TOC}$  values of sedimentary organic matter in the most negative part of 489 the negative CIE associated with the end-Triassic mass extinction level are similar, -490 29 to -31‰, in both the north-western Tethys and Panthalassic marginal marine 491 basins, probably reflecting predominantly marine constituents due to enhanced productivity (Figure 4). The ~3‰ more positive  $\delta^{13}C_{TOC}$  values of sedimentary 492 493 organic matter in levels pre- and postdating the negative CIE in the north-western 494 Tethys realm, relative to similar coeval stratigraphy in Panthalassic marginal marine 495 basins, likely reflects increased terrestrial-marine mixing (with a relatively larger 496 input of terrestrial organic matter; Figures 4 and 5). The possibly limited vegetation 497 cover and/or standing biomass in the western Pangaean hinterland, and the 498 therefore possibly (significantly) limited terrestrial organic-matter supply to eastern 499 Panthalassic marine basins may have resulted in limited to no source mixing in these 500 localities pre- and succeeding the negative CIE. Because of this situation, changes in 501  $\delta^{13}C_{TOC}$  of the sedimentary organic matter in eastern Panthalassic marginal marine 502 basins across the Triassic–Jurassic transition may therefore better reflect true 503 changes in the exogenic carbon cycle, in this case marked by a 2–3‰ negative 504 carbon-isotope shift, similar in magnitude to that observed in bulk marine carbonate  $(\delta^{13}C_{CARB})$  in the north-western and eastern Tethyan realm, and the terrestrial records 505

506 of North America and Morocco (Figure 1 and 4). The larger amplitude negative CIE 507 of 5–6.5‰ in the north-western Tethyan region may thus illustrate a predominantly 508 aquatic organic-matter supply to marine sediments during the negative CIE, 509 contrasted by the overall mixing of terrestrial and marine organic matter before and 510 after the CIE (Figure 5), resulting in the observed elevated magnitude of the end-511 Triassic negative CIE in this region (Figure 4).

512 The carbon-isotope composition of molecular biomarkers from higher plant leaf waxes ( $\delta^{13}C_{n-alkanes}$ ) and macrofossil wood ( $\delta^{13}C_{wood}$ ) in the continental Newark and 513 514 Hartford basins also shows only a modest ~3 and 4‰ negative CIE, respectively, 515 concomitant with the end-Triassic mass extinction (Whiteside et al., 2010). Similar 516 data from coeval sediments in the Eiberg Basin (Austria), however, show a major 7-517 8‰ negative CIE in terrestrially derived long- and short-chain *n*-alkanes (Figure 4; 518 Ruhl et al., 2011). This large-amplitude excursion may then reflect either (I) changes 519 in the dominant terrestrial vegetation (as shown from palynological studies of the 520 same samples; Bonis et al., 2009), and associated differences in biological fractionation for light (<sup>12</sup>C) carbon, (II) increased biological fractionation for light (<sup>12</sup>C) 521 522 carbon in response to enhanced hydrological cycling during the end-Triassic mass 523 extinction negative CIE (Ruhl et al., 2011), and/or (III) relatively increased biological 524 fractionation for light (<sup>12</sup>C) carbon in terrestrial plants, in response to elevated 525 atmospheric pCO<sub>2</sub> following CAMP volcanism (McElwain et al., 1999). 526 Alternatively, the  $\delta^{13}C_{TOC}$  record at Arroyo Alumbre may not accurately reflect the magnitude of change in  $\delta^{13}$ C of exogenic carbon reservoirs during the end-527 528 Triassic mass extinction, due to (1) preferential degradation of the more labile and 529 <sup>13</sup>C-depleted marine organic matter from the end-Triassic mass extinction interval 530 during thermal maturation of the sediments, or (2) reduced terrestrial biological fractionation for light (<sup>12</sup>C) carbon in response to reduced hydrological cycling across 531 532 the western Pangaean landmass during the time of the end-Triassic mass extinction. 533 Although both mechanisms are not totally rejected here, they are both considered

534 unlikely because multiple model studies suggest that extremely arid, desert-type 535 conditions were likely present between ~60° North and ~60° South in western 536 Pangaea (Figure 1); consequently, meaningful vegetation cover or standing biomass 537 may have been absent in the hinterland of the study area. The impact of terrestrial 538 vegetation (and potentially associated differences in fractionation during thermal 539 maturation and/or under conditions of changing hydrological cycling) on bulk 540 sedimentary organic carbon-isotope ratios may therefore have been negligible and 541 may not have impacted the magnitude of the observed negative CIE at the end-542 Triassic mass extinction interval in the Argentinian succession. The likelihood of the 543 observed 2‰ negative CIE associated with the end-Triassic mass extinction in the 544 Arroyo Alumbre section reflecting true global carbon-cycle change is further 545 corroborated by the similar magnitude negative CIE observed in other sections from 546 eastern Panthalassa marginal marine basins (Figure 1, 4 and 5).

547

### 548 [4.4] ENHANCED TRIASSIC-JURASSIC CARBON RELEASE AND BURIAL

549 Mesozoic global-change events, such as oceanic anoxic events (OAEs) are 550 marked by the widespread development of anoxic-euxinic conditions and the 551 associated sequestration of carbon into organic-rich black shales (Jenkyns, 2010). 552 The massive burial of isotopically light carbon at these times led to a prolonged 553 positive excursion in  $\delta^{13}$ C in global exogenic carbon reservoirs, observed most 554 obviously in the case of OAE 2 at the Cenomanian–Turonian boundary (at ~94 Ma) 555 and during the Early Toarcian OAE (T-OAE) (at ~183 Ma; Jenkyns and Clayton, 556 1986; Jenkyns, 1988; Tsikos et al., 2004; Xu et al., 2018). The T-OAE is furthermore marked by a major negative perturbation in global  $\delta^{13}$ C records superimposed on the 557 558 prolonged positive carbon-isotope excursion, possibly as a result from isotopically 559 light carbon released as methane from ocean-floor methane-clathrates (Jenkyns et 560 al., 2002; Hermoso et al., 2009; Hermoso and Pellenard, 2014; Xu et al., 2018; 561 Storm et al., 2020). The duration and magnitude of CIEs associated with Mesozoic

562 OAEs are ultimately controlled by the pattern of contemporaneous carbon release
563 and carbon sequestration into marine and lacustrine sediments (Jenkyns, 2010; Xu
564 et al., 2017*a*).

565 The duration of the global exogenic carbon-cycle perturbation and the 566 associated observed negative carbon-isotope excursion (CIE) in marine and 567 terrestrial sedimentary archives at the end-Triassic mass extinction is relatively short 568 (between ~20–85 kyr; Deenen et al., 2010; Schoene et al., 2010; Ruhl et al., 2010; 569 Whiteside et al., 2010; Yager et al., 2017), much shorter than the negative CIEs that 570 characterize the Permo-Triassic boundary interval, the T-OAE and the Paleocene-571 Eocene Thermal Maximum (PETM), each of which lasted several 100 kyr (Lourens et 572 al., 2005; Shen et al., 2011; Huang and Hesselbo, 2014).

573 Triassic–Jurassic climatic and environmental change, in response to CAMP 574 emplacement and associated carbon greenhouse gas release, would have 575 significantly impacted local and regional depositional environments. Some shallow-576 marine basins along Triassic–Jurassic continental margins witnessed a significant 577 shift to reducing water-column conditions, with even euxinic (sulphidic) conditions in 578 the photic zone (Richoz et al., 2012; Jaraula et al., 2013; Kasprak et al., 2015; Jost et 579 al., 2017). Climatic and environmental change, and an associated increase in global 580 weathering rates at this time (Cohen and Coe, 2007; see also discussion in Ruhl et 581 al., 2016), may have enhanced the supply of nutrients to marginal marine basins and 582 coastal regions, as postulated for OAEs (Jenkyns, 2010). Many Triassic-Jurassic 583 marginal marine basins are indeed characterised by increased sedimentary total 584 organic carbon (TOC) concentrations commencing around the level of the end-585 Triassic mass extinction, possibly in response to enhanced marine primary 586 productivity and redox changes favouring enhanced preservation (Hesselbo et al., 587 2004; Ruhl et al., 2009; Xu et al., 2017b).

588 The Neuquén Basin was possibly also marked by a shift towards more 589 reducing water-column conditions at that time, allowing the drawdown of redox-

sensitive trace metals, such as molybdenum (Mo). In oxic conditions, Mo exists as soluble molybdate ( $MoO_4^{2-}$ ) that adsorbs onto Mn oxides, which only slowly precipitate. In sulfidic (euxinic) waters, however, molybdate dissociates into thiomolybdate anions, which are rapidly reduced to highly reactive Mo(IV) sulfides that precipitate out of solution, leading to sedimentary Mo enrichment (Dahl et al., 2013).

596 Sedimentary Mo concentrations are largely below the hand-held-XRF detection 597 limit (of ~3 ppm) in the lower part of the studied succession (Figure 2) and Mo/Zr 598 ratios are consequently not meaningful. Higher up in the succession Mo values do, 599 however, increase up to 20 ppm in strata representing the Triassic–Jurassic 600 transition (Figure 2; Supplementary Data File). The observed sedimentary Mo values 601 are not as enriched as in euxinic shales (Dahl et al., 2013), but high Mo fluxes from 602 the water column to the sea floor may have been masked by overall high 603 sedimentation rates in the Neuquén Basin. Furthermore, sulphur (S) and S/Zr values 604 are elevated above detection limit stratigraphically above 169 m. peaking from 181-605 188 m in the studied succession (Figure 2), the stratigraphical interval with most positive  $\delta^{13}C_{TOC}$  values. The monotonous nature of the studied sedimentary 606 607 succession suggests that the evolution of elevated S and S/Zr values are unrelated 608 to lithology; increased S and S/Zr values may rather have resulted from elevated 609 levels of sedimentary pyrite, possibly accumulating in the sediments as micron-scale 610 pyrite framboids precipitated in the water column as well as forming diagenetically 611 below the sea floor. Combined, the elevated total organic carbon (TOC), Mo and S 612 concentrations stratigraphically coinciding with the end-Triassic mass extinctions and 613 associated carbon-cycle perturbations, do suggest that the Neuquén Basin 614 developed more reducing water column conditions (Figure 2) at the same time as 615 marginal marine basins in Europe and North America. 616 Importantly, although the Triassic–Jurassic transition was marked by a

617 prolonged negative shift in the  $\delta^{13}$ C of exogenic carbon reservoirs, extending over

the Hettangian-Early Sinemurian interval (Hesselbo et al., 2002; van de 618 619 Schootbrugge et al., 2005; Ruhl et al., 2010b; Whiteside et al., 2010; Hüsing et al., 2014; Peti and Thibault, 2017; Xu et al., 2017b; van de Schootbrugge et al., 2019; 620 621 Hesselbo et al., 2020; Storm et al., 2020), the 'initial' negative CIE characterizing the 622 end-Triassic mass extinction is separated from this long-term negative shift by a globally observed pronounced positive excursion in  $\delta^{13}$ C (Figures 2, 3 and 6; 623 Hesselbo et al., 2002). This positive excursion in the  $\delta^{13}$ C of exogenic carbon 624 625 reservoirs stratigraphically coincides with the emplacement of the volumetrically 626 major CAMP basalt extrusives in North America, Africa and Europe (Panfili et al., 627 2019), and the development of anoxic-euxinic conditions in several marginal marine 628 basins at that time (Richoz et al., 2012; Blackburn et al., 2013; Jaraula et al., 2013; 629 Kasprak et al., 2015; Jost et al., 2017; Kent et al., 2017). This shift to more positive 630 carbon-isotope values is observed globally in carbonate and marine and terrestrial 631 organic matter, including primary skeletal calcite and terrestrial higher-plant 632 molecular *n*-alkanes (Figures 2, 3 and 6; Hesselbo et al., 2002; Yin et al., 2006; 633 Ward et al., 2007; Williford et al., 2007; Guex et al., 2008; van de Schootbrugge et 634 al., 2008; Korte et al., 2009; Ruhl et al., 2009; Whiteside et al., 2010; Bartolini et al., 635 2012; Lindström et al., 2012; Felber et al., 2015; Yager et al., 2017; Ge et al., 2018; 636 Hesselbo et al., 2020). Enhanced marine productivity and preservation in 637 geographically more widespread anoxic-euxinic settings at this time, with the 638 associated elevated sequestration of isotopically light carbon into marine marls and 639 shales, may have acted as a powerful negative feedback in the global climate system 640 and carbon cycle. This phenomenon, which would have potentially driven the carbon 641 in the ocean–atmosphere system to more <sup>13</sup>C-enriched values, manifestly 642 complicates geochemical and mass-balance constraints on the magnitude and 643 source of isotopically light carbon released at this time. 644 The 2–3‰ exogenic carbon-cycle perturbation at the time of the end-Triassic

645 mass extinction likely resulted from carbon released primarily from (1) CAMP-basalt

volcanogenic carbon degassing (with a possible C-isotopic composition of ~-4 to 8‰; Macpherson and Mattey, 1994), (2) thermogenic methane release by the
intrusion of CAMP-related dykes and sills into subsurface organic-rich shales (with a
C-isotopic composition of ~-35 to -45‰), and/or (3) biogenic methane release from
destabilizing sea-floor methane hydrates (with a C-isotopic composition of ~-60‰)
(Coleman et al., 1981; Berner and Faber, 1987; Whiticar, 1999).

652 Previous studies have suggested that the negative CIE at the time of the end-Triassic mass extinction was relatively short in duration, with a rapid  $10^3-10^4$  yr onset 653 654 (Deenen et al., 2010; Schoene et al., 2010; Ruhl et al., 2010; Whiteside et al., 2010; 655 Yager et al., 2017). Simple mass-balance calculations presented here suggest that a ~2.5‰ negative shift in global exogenic  $\delta^{13}$ C can be achieved with the release of 656 657 ~20000 Gt C from methane hydrates or a larger mass of carbon as thermogenic 658 methane or from volcanogenic sources (Figure 6; Supplementary Figure 3). Re-659 shuffling of carbon between Triassic–Jurassic carbon reservoirs, such as through the 660 oxidation of terrestrial biomass, may have further contributed to a negative shift in 661 global ocean-atmosphere carbon-isotope values. The doubling to tripling of 662 atmospheric  $pCO_2$  (from ~1000 to ~2000 ppmv based on stomatal-density analyses, or from ~2000 to ~6000ppmv based on pedogenic carbonate  $\delta^{13}$ C (McElwain et al., 663 664 1999; Schaller et al., 2011; Steinthorsdottir et al., 2011; Schaller et al., 2012; Schaller 665 et al., 2015), combined with a significant and rapid reduction in seawater pH (Črne et 666 al., 2011; Hönisch et al., 2012; Ikeda et al., 2015) may, however, suggest that a 667 much larger quantity of greenhouse gases was released at this time. Therefore, 668 either (1) large amounts of carbon whose isotopic signature was similar to that of the 669 ocean-atmosphere system (such as volcanogenic CO and CO<sub>2</sub>) were released, or 670 (2) large amounts of isotopically depleted carbon (such as thermo- or biogenic 671 methane (CH<sub>4</sub>)) were released, but this was largely offset by contemporaneous burial 672 of isotopically light organic matter, resulting in a net reduction in the amplitude of the observed negative shift in  $\delta^{13}$ C of global exogenic carbon reservoirs. A simple mass-673

balance calculation suggests that  $\sim 13 \times 10^5$  Gt of organic carbon, with a carbon-674 675 isotope composition of ~-25‰, had to have been buried over about 200 kyr, to explain the observed positive excursion in  $\delta^{13}$ C (following the end-Triassic mass 676 677 extinction and associated negative CIE) around the Triassic–Jurassic transition. 678 Such a carbon-burial flux, approximately half of the current yearly anthropogenic 679 carbon release, may be an underestimate of the actual figure. Even higher rates of 680 organic-carbon burial may have taken place at the time, explaining the short duration 681 and relatively small (2–3‰) magnitude of the 'initial' negative CIE at the end-Triassic 682 mass extinction interval. Actual quantitative analytical constraints on global (organic) 683 carbon-burial rates across the Triassic-Jurassic transition do not currently exist. 684 Accurate and precise constraints on (changes in) atmospheric  $pCO_2$ , and ocean pH 685 levels are essential to constrain the Triassic-Jurassic carbon-cycle evolution and 686 evaluate the magnitude and rate of change in carbon fluxes between the exogenic 687 carbon reservoirs, in response to CAMP magmatism.

688

## 689 **[5] CONCLUSIONS**

690 A bio- and chemostratigraphically constrained and expanded Triassic–Jurassic 691 boundary succession from the Arroyo Alumbre section in the Rio Atuel Region of the 692 Neuquén Basin, Argentina, is here described. Carbon-isotope analyses of 693 sedimentary organic matter ( $\delta^{13}C_{TOC}$ ) in this biostratigraphically well-constrained 694 section can be reliably used to correlate Triassic–Jurassic boundary successions 695 between the northern and southern hemisphere and the Panthalassic and Tethyan 696 realms. The level of the end-Triassic mass extinction, located with the help of newly 697 discovered ammonoid faunas, is marked by a limited (2–3‰) magnitude negative CIE, similar to that found in other eastern Panthalassic basins as well as  $\delta^{13}C_{\text{CARB}}$ 698 699 records in the Tethys realm, but significantly smaller than observed in contemporaneous Tethyan  $\delta^{13}C_{TOC}$  negative CIEs of 5–6‰. 700

Possibly arid climatic conditions, extending between 60° northern and southern latitudes across the western Pangaean landmass, may have resulted in a predominantly marine organic matter flux to the sedimentary realm in eastern Panthalassic basins, and mixing and changing marine and terrestrial sources of organic matter can explain the observed larger amplitude negative CIE in  $\delta^{13}C_{TOC}$ records from the Tethyan realm.

Relative enrichment of sedimentary Hg (and Hg/TOC) in the marine Neuquén Basin initiated before the end-Triassic mass extinction and associated negative CIE, and before the emplacement of CAMP basalt flows began in North America and Africa. Sedimentary Hg enrichments do appear to correlate with the onset of CAMPassociated dyke and sill emplacement, suggesting thermal alteration of intruded sedimentary piles or intrusive magma degassing as a potential major source of elevated Hg fluxes to the atmosphere at this time.

714 The Neuquén Basin may have been marked by the development of reducing 715 water-column conditions allowing a build-up of organic matter on the sea floor around 716 the Triassic–Jurassic transition as a result of increased marine primary productivity 717 and/or increased preservation. Combined with similar observations across the 718 Panthlassic margin and the northwest Tethyan seaway, the data from Argentina 719 suggest potentially significantly elevated organic-carbon burial rates in a global 720 context. Simple mass-balance calculations show that enhanced carbon burial rates 721 at this time, beginning at or directly succeeding the time of the end-Triassic mass 722 extinction, and in line with the major emplacement phase of CAMP basalts, can 723 explain the observed evolution of the global exogenic carbon cycle across the 724 Triassic–Jurassic transition.

725

#### 726 FIGURE CAPTIONS

Figure 1 Triassic–Jurassic continental configuration and the geographical
extent of Central Atlantic Magmatic Province (CAMP) magmatism (shown by the

729 brown shading), modified after Korte et al. (2019) and references therein. The magnitude of the  $\delta^{13}C_{TOC}$  negative CIE, concomitant with the end-Triassic mass 730 731 extinction, is given in *blue* for the Pangaean realm, in *black* for the Tethyan realm, 732 and in green for the terrestrial realm, whereas the magnitude of this CIE in  $\delta^{13}C_{CARB}$ 733 is given in *purple* (this study; Palfy et al., 2001; Hesselbo et al., 2002; Galli et al., 734 2005; Yin et al., 2006; Williford et al., 2007; van de Schootbrugge et al., 2008; Korte et al., 2009; Ruhl et al., 2009; Deenen et al., 2010; Whiteside et al., 2010; Deenen et 735 736 al., 2011; Črne et al., 2011; Bartolini et al., 2012; Lindström et al., 2012; Dal Corso et 737 al., 2014; Al-Suwaidi et al., 2016; Ge et al., 2018; Yager et al., 2017; Hu et al., 2020). 738 Values for maximum sedimentary Hg (ppb) and Hg/TOC (ppb/%) values are given 739 with the coloured stars (this study; Thibodeau et al., 2016; Percival et al., 2017). The 740 geographical extension of arid climate belts and vegetation patterns across the 741 western and central Pangean landmass, at the time of the Triassic-Jurassic 742 transition, and the potentially expanded Hadley Cells under conditions of enhanced 743 greenhouse warming, are from Chandler et al., 1992; Rees et al., 2000; Huynh and 744 Poulsen, 2005; Sellwood and Valdes, 2006; Hu and Fu, 2007; Lu et al., 2007; Previdi 745 and Liepert, 2007; Kang and Lu, 2012; Sellwood and Valdes, 2008.

746

747 Figure 2 Bio- and chemostratigraphical, and geochemical data across the 748 Triassic-Jurassic transition in the Arroyo Alumbre succession of the Neuquén Basin, 749 Argentina. Lithological columns presented here are from Riccardi et al. (2004), and 750 Damborenea and Manceñido (2012). Ammonoids collected from the studied section 751 constrain the age of the studied succession to span the Triassic-Jurassic transition 752 (Damborenea et al., 2017; and newly collected samples in Riccardi, 2019). Bivalve 753 diversity patterns show a significant decrease across the Triassic–Jurassic transition 754 (Damborenea et al., 2017 and references therein). An abrupt and stratigraphically short-lived 2–3‰ negative excursion in  $\delta^{13}C_{TOC}$  suggests that the stratigraphic 755 756 interval (in pink) represents the global end-Triassic mass extinction (this study).

757 Sedimentary zirconium (Zr) concentrations (this study) are relatively constant 758 throughout the studied succession, suggesting limited lithological change in the 759 studied shales and mudstones of the Arroyo Alumbre succession. Elevated 760 sedimentary Hg, Hg/TOC and Hg/Zr enrichments in strata stratigraphically preceding this level suggest earlier CAMP-related magmatic activity (this study). Elevated total 761 762 organic carbon (TOC), and redox-sensitive trace and major element concentrations 763 (molybdenum (Mo and Mo/Zr) and sulphur (S and S/Zr)) suggest potential 764 environmental change and a possible shift toward more reducing conditions across 765 the end-Triassic mass extinction (this study).

766

767 Figure 3 Bio- and chemostratigraphic and geochronological framework across 768 the Triassic–Jurassic transition in the Neuquén Basin (Argentina; this study), the 769 Bristol Channel Basin (UK; Hesselbo et al., 2002 and references therein), Levanto 770 (Peru; Yager et al., 2017), New York Canyon (USA; Bartolini et al., 2012 and 771 references therein; Thibodeau et al., 2016) and the Eiberg Basin (Austria; Ruhl et al., 772 2009; Hillebrandt et al., 2013). Ash-bed ages from Levanto (Peru) and New York 773 Canyon (USA) are from [A, B, C, D] Wotzlaw et al. (2014), [E, F, G] Schaltegger et 774 al. (2008), Schoene et al. (2010), Wotzlaw et al. (2014), [H, I] Guex et al., 2012, [J] 775 Schoene et al. (2010).

776

 $\delta^{13}C_{TOC}$ ,  $\delta^{13}C_{CARB}$  and  $\delta^{13}C_{n-alkane}$  data across levels recording the end-777 Figure 4 778 Triassic mass extinction and associated negative carbon-isotope excursion (CIE), 779 from marine successions in the Tethyan and Panthalassic realms and from terrestrial 780 successions in North America and northern Africa (this study; Hesselbo et al., 2002; 781 Galli et al., 2005; Williford et al., 2007; Ruhl et al., 2009; Deenen et al., 2010; Črne et 782 al., 2011; Deenen et al., 2011; Ruhl et al., 2011; Bartolini et al., 2012; Lindström et 783 al., 2012; van de Schootbrugge et al., 2013; Dal Corso et al., 2014; Felber et al., 784 2015; Thibodeau et al., 2016; Suarez et al., 2017; Yager et al., 2017).

785

[A] The magnitude of the negative carbon-isotope excursion in  $\delta^{13}$ C of 786 Figure 5 total sedimentary organic carbon ( $\delta^{13}C_{TOC}$ ), bulk-rock carbonate ( $\delta^{13}C_{Carb}$ ) and leaf-787 788 wax *n*-alkane biomarkers ( $\delta^{13}C_{\text{leaf-wax }n-\text{alkanes}}$ ), with references referring to: (1) 789 Kennecott Point, Queen Charlotte Islands, Canada (Ward et al., 2001); (2) New York 790 Canyon, Nevada, USA (Ward et al., 2007); (3) Arroyo Alumbre, Argentina (This 791 study); (4) Pucara Basin, Peru (Yager et al., 2017); (5) Japan (Fujisaki et al., 2018); 792 (6) Newark & Hartford basins, USA (Whiteside et al., 2010); (7) Argana Basin, 793 Morocco (Deenen et al. (2010), Dal Corso et al. (2014)); (8) Fundy Basin, Canada 794 (Deenen et al., 2011); (9) Montenegro (Črne et al., 2011); (10) Hungary (Palfy et al., 795 2001); (11) Italy (Galli et al., 2005); (12) Southern Tibet, China (Yin et al. 2006); (13) 796 Danish Basin, Denmark (Lindström et al., 2012); (14) German Basin, Germany (van 797 de Schootbrugge et al., 2008); (15) Italy (Bachan et al., 2012); (16) St Audries Bay, 798 Bristol Channel Basin, Somerset, UK (Hesselbo et al., 2002); (17) Restentalgraben, 799 Eiberg Basin, Austria (Ruhl et al., 2009); (18) Kuhjoch, Eiberg Basin, Austria (Ruhl et 800 al., 2011); (19) United Arab Emirates (Ge et al., 2018). [B] Model explaining how 801 organic-carbon source mixing pre- and post- the end-Triassic mass extinction could 802 have affected the magnitude of the associated negative carbon-isotope excursion 803 (CIE).

804

805 Figure 6 Stratigraphical framework showing the long-term evolution of the exogenic carbon cycle based on  $\delta^{13}C_{TOC}$  data from the marine Arroyo Alumbre 806 807 (Neuquén Basin, Argentina) and St Audries Bay (Bristol Channel Basin, UK) sections, marine  $\delta^{13}C_{SKELETAL CALCITE}$  from Lavernock Point, Watchet and St Audries 808 809 Bay (Bristol Channel Basin, UK), terrestrial  $\delta^{13}C_{TOC}$  from the continental Newark and 810 Hartford Basins (USA) (this study; Hesselbo et al., 2002; Korte et al., 2009; Ruhl et 811 al., 2010; Whiteside et al., 2010). Stratigraphical correlation between these sections 812 is also based on this study, Deenen et al. (2010), Whiteside et al. (2010), and Hüsing

813 et al. (2014). Also shown are reconstructed pCO<sub>2</sub> data from the Newark and Hartford 814 Basins (USA) (Schaller et al., 2011, 2012, 2015), and Hg (and Hg/TOC) data from 815 the (Neuguén Basin, Argentina, this study and Percival et al. (2017)). The 816 stratigraphic occurrence of lavas of the Central Atlantic Magmatic Province (CAMP) 817 and the age of the end-Triassic mass extinction are based on Davies et al. (2017), 818 Kent et al. (2017 and references therein), and references in Xu et al. (2017b). The 819 stratigraphic occurrence of elevated organic-carbon contents suggestive of low-820 oxygen conditions in marine basins during Triassic–Jurassic boundary times is based on references given in Section [4.4]. The computed  $\delta^{13}$ C evolution of exogenic 821 822 carbon reservoirs across the Triassic–Jurassic transition is based on simple mass-823 balance calculations, following different models as explained in the Supplementary 824 Information section, incorporating the combined timed volcanogenic carbon release (with  $\delta^{13}C = -5\%$ ), biogenic methane release from ocean-floor clathrates (with  $\delta^{13}C =$ 825 -60‰), and elevated organic-carbon burial (with  $\delta^{13}C = -35\%$ ). 826

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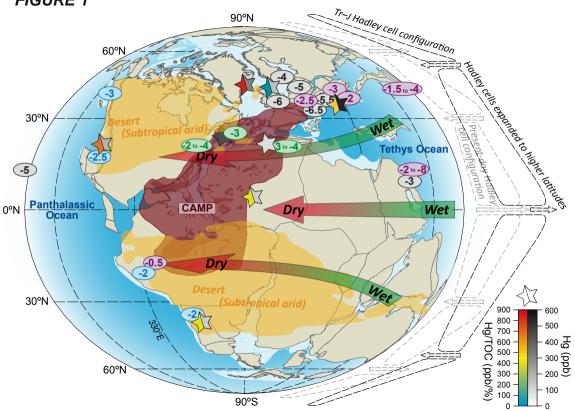
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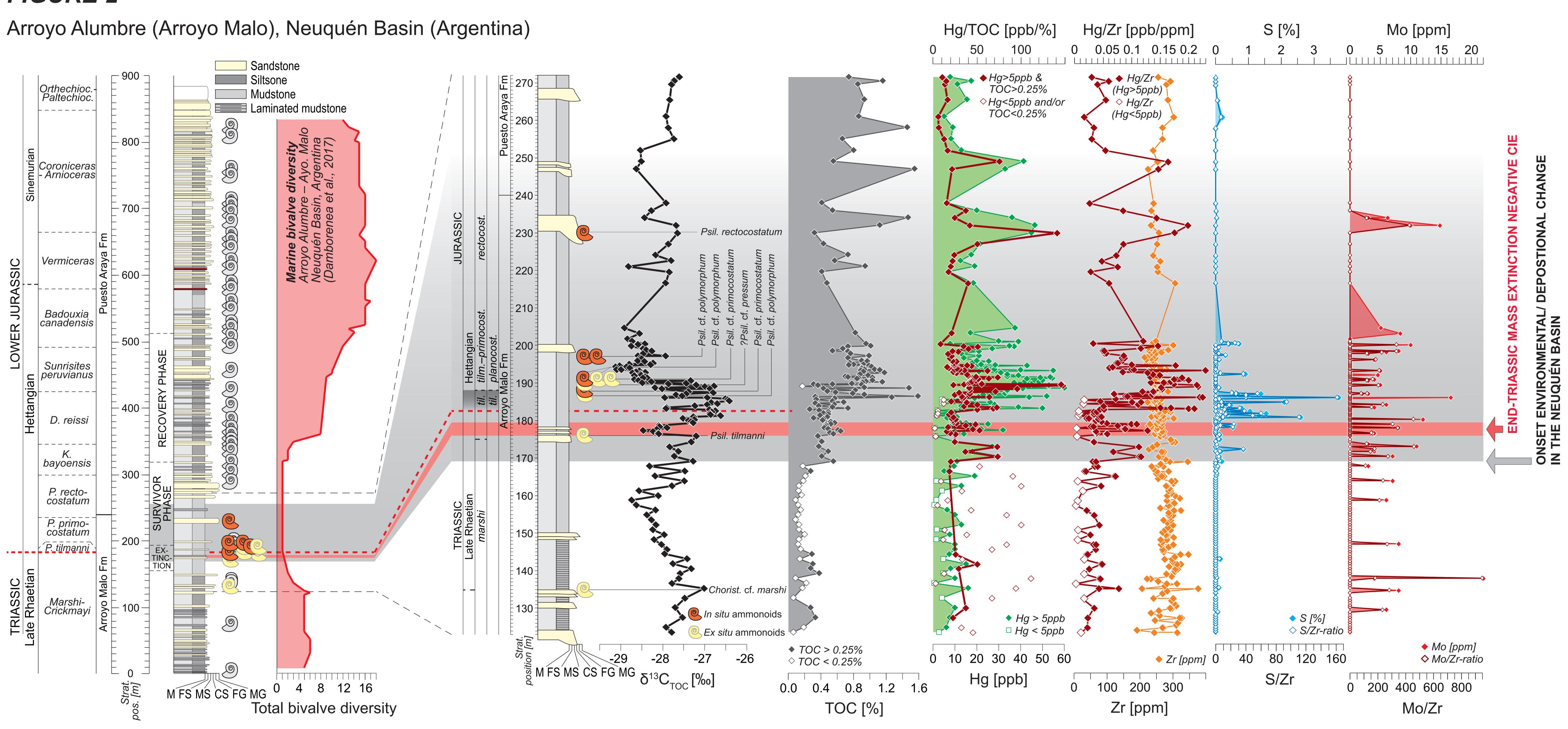
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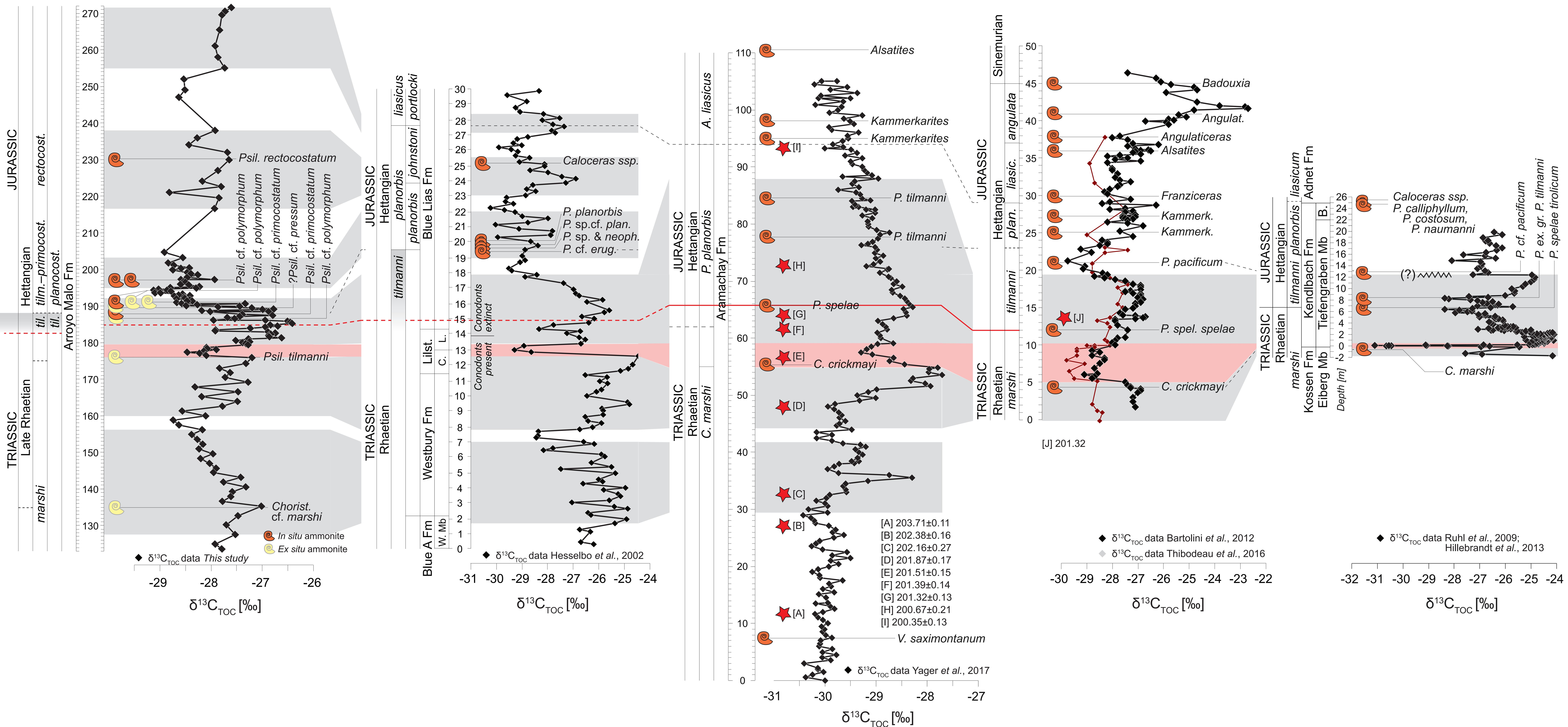
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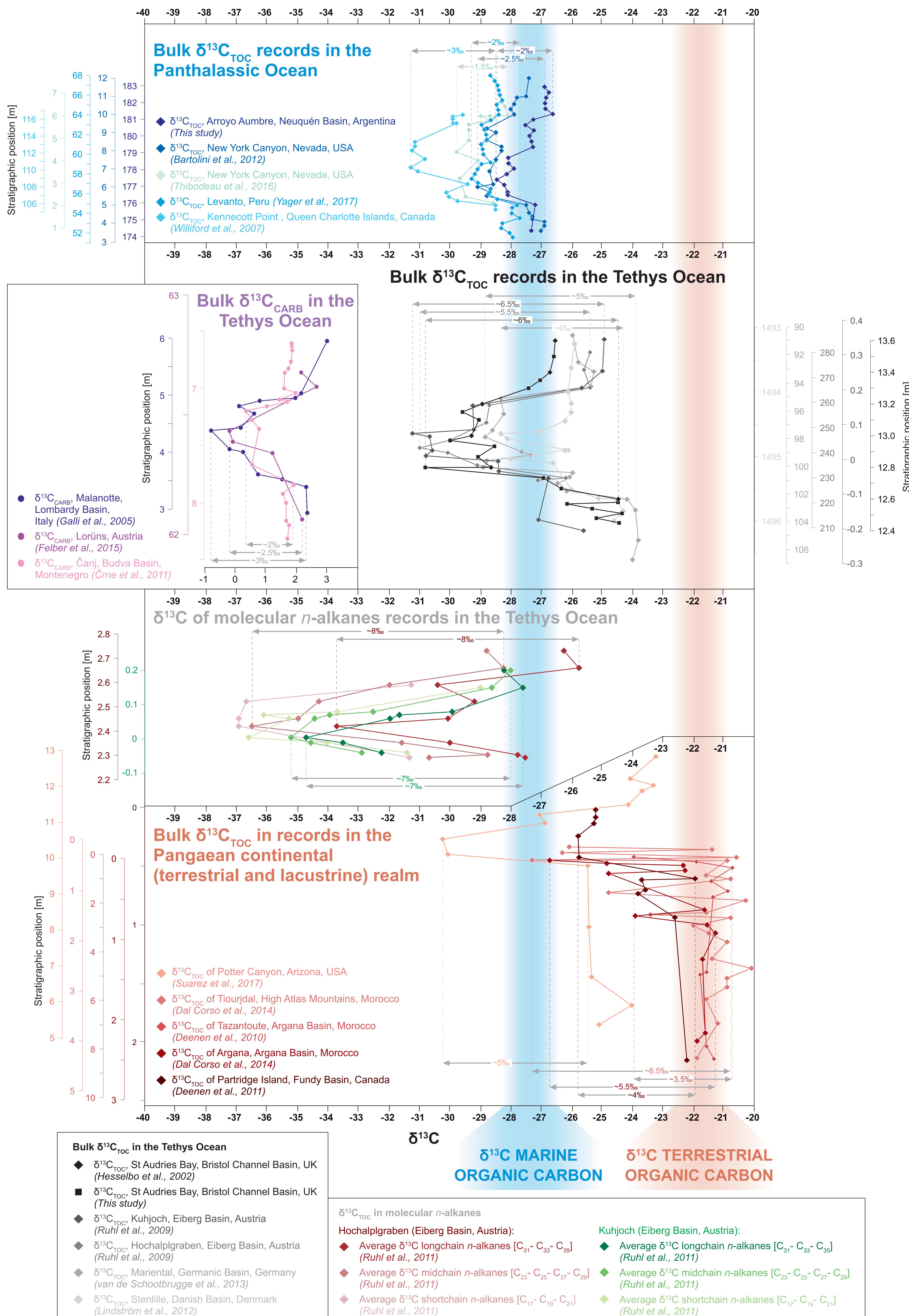




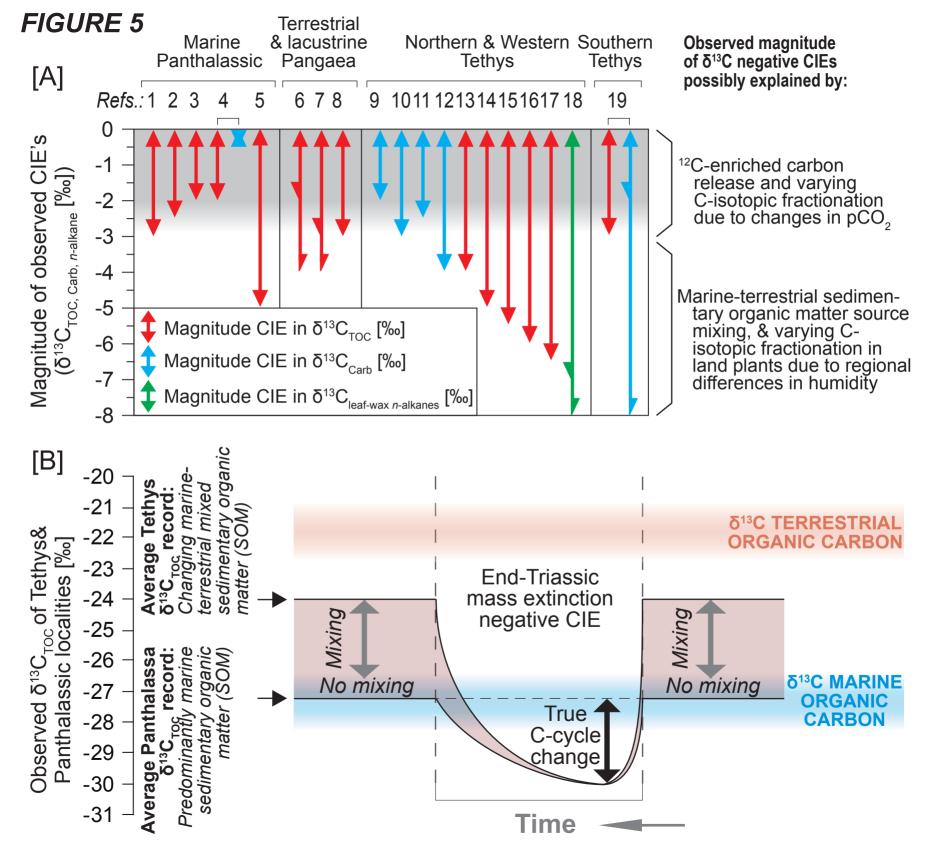
Arroyo Malo, Neuquén Basin (Argentina)

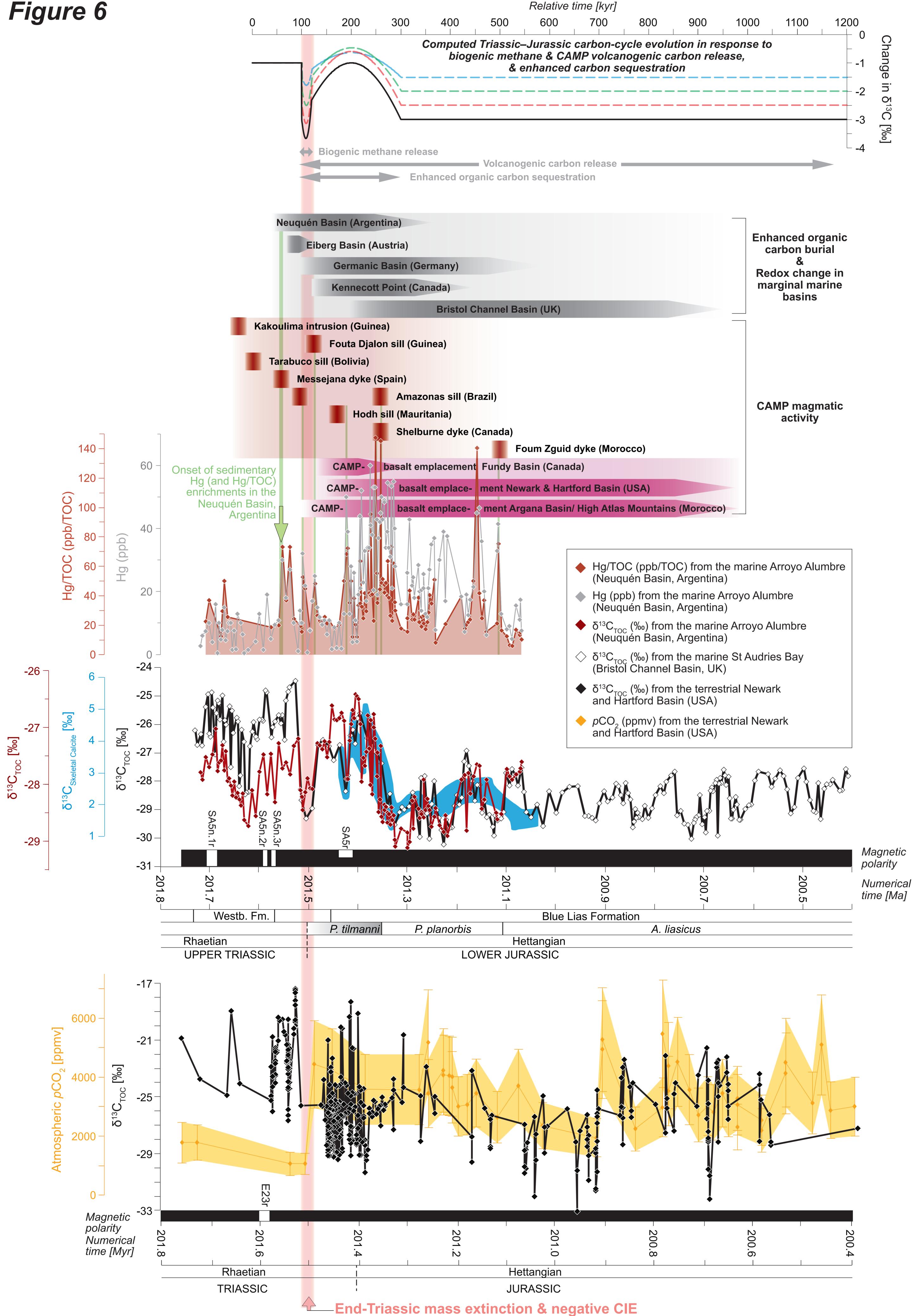


# Kuhjoch, Eiberg Basin (Austria)



**δ**<sup>13</sup>**C** 





1	SUPPLEMENTARY INFORMATION
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3	ON THE ONSET OF CENTRAL ATLANTIC MAGMATIC PROVINCE (CAMP)
4	VOLCANISM AND ENVIRONMENTAL AND CARBON-CYCLE CHANGE AT THE
5	TRIASSIC-JURASSIC TRANSITION (NEUQUÉN BASIN, ARGENTINA)
6	
7	Ruhl, M. <sup>1,2</sup> , Hesselbo, S.P. <sup>3</sup> , Al-Suwaidi, A. <sup>4</sup> , Jenkyns, H.C. <sup>2</sup> , Damborenea, S.E. <sup>5</sup> ,
8	Manceñido, M.O. <sup>5</sup> , Storm, M. <sup>2,6</sup> , Mather, T.A. <sup>2</sup> , Riccardi, A.C. <sup>5</sup>
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21	Africa
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### 29 [1] MATERIALS & METHODS: $\delta^{13}C_{TOC}$ , TOC, hh-XRF & Hg ANALYSES

30

### 31 **[1.1] Materials**

The Triassic–Jurassic sedimentary succession at Arroyo Alumbre was sampled during two field seasons (2014 and 2016). The Arroyo Alumbre (and associated nearby Arroyo Malo) sections were previously studied and described by Damborenea and Manceñido (2012, and references therein), who distinguished the Arroyo Malo Formation at the base (0–280m), stratigraphically overlain by the conglomeratic lenses of the El Freno Formation and the subsequent deposits of the El Cholo Formation.

39 The section is located high up in the Andes mountains, west of the main road 40 from Malarguë to Mendoza (leaving the main road at El Sosneado), at 34°49'767''S – 41 69°53'224"W (Google-maps coordinates: -34.827675 (South), -69.8888884 (West)) 42 (Supplementary Materials Figures 1, 2-a, 2-b). The base of the relatively steep 43 section is located at ~2395m above sea-level, on the western side of the Arrovo 44 Alumbre valley (Supplementary Figures 2-c, d, e). The Arroyo Alumbre valley is a 45 minor side-valley of the Arroyo Malo valley, which itself connects to the Rio Atuel to 46 the south. The section can be accessed from El Sosneado along a ~38 km dirt-road 47 along the Rio Atuel, followed by a ~1 hr hike (Supplementary Figure 2-a,b).

The Triassic–Jurassic sedimentary succession at Arroyo Alumbre represents a fully marine and stratigraphically expanded record (Figure 1). 42 samples (RA-1 to RA-42) were collected in 2014, from ~187m to ~272m in the section, and 160 additional samples (AA-1 to AA-160) were collected in 2016, from ~123m to ~205m in the section (Supplementary Figures 2-d,e).

53

### 54 [1.2] Fossil collections

All Triassic–Jurassic fossil material collected and described from the fossiliferous
levels of the Arroyo Malo Formation at the Arroyo Alumbre/ Arroyo Malo localities, as

57 described in Damborenea et al. (2017, and references therein) and (Riccardi (2019,

and references therein), are deposited in the collections of the Invertebrate

59 Palaeontology Department of the La Plata Natural Sciences Museum (MLP).

60

### 61 [1.3] Bulk-rock organic $\delta^{13}$ C ( $\delta^{13}$ C<sub>TOC</sub>) analysis

62 One gram of homogenized sample was treated with 40mL cold HCI (3 molar) to 63 dissolve the carbonate. Samples (dissolved in 3 molar HCI) were then put on a hot 64 plate for 2 hours at 80°C. Samples were subsequently rinsed 4 times with distilled 65 water to reach neutral pH. Samples were then oven-dried (at  $30^{\circ}$ C) and again 66 powdered. Around 1-15mg (depending on organic-carbon concentration) of 67 homogenized de-carbonated sample residue was weighed into a 8×5-mm tin capsule  $\delta^{13}C_{TOC}$  analyses. The  $\delta^{13}C_{TOC}$  value of the samples was measured at the Stable 68 69 Isotope Laboratory of the Open University (Milton Keynes, UK), with a Thermo 70 Scientific Flash 2000 HT Elemental Analyzer (EA) coupled to a Thermo Scientific 71 MAT253 isotope ratio mass spectrometer via a Conflo IV open split interface. The 72 Thermo Scientific Flash 2000 HT EA has a MAS2000 carousel and between the 73 carousel and the EA sits a Thermo No Blank Device (NBD), allowing for the single 74 sample purging with helium. The EA is also equipped with a Thermal Conductivity 75 Detector (TCD). Automated dilution of sample gas with the Conflo IV open split 76 interface allows for high dynamic range of C+N content and controls the introduction 77 of the "reference" gases. Isotope ratios are reported in standard delta notation 78 relative to Vienna PDB. 79 Analytical precision was checked with the routine analysis of four internal and

80 referenced laboratory standards (urea 020914MAG, IAEA-CH-6, NIST 8573,

81 IRR041), showing measured average  $\delta^{13}C_{TOC}$  values of -45.24‰, -10.39‰, -26.52‰,

82 -23.58‰, respectively, and standard deviations of 0.85‰, 0.12‰, 0.28‰, 0.49‰.

83

### 84 [1.4] Rock-Eval: Total organic carbon (TOC)

85 Total organic carbon (TOC) analyses were performed using a Rock-Eval 6 86 Standard Analyser unit (Vinci Technologies), with pyrolysis and oxidation ovens, a 87 flame ionization detector and infrared cell, at the Department of Earth Sciences, 88 University of Oxford. The S1, S2, TMax, S3co/ S3co2 (New Oxygen Index), S4co/ 89 S4co2 (Residual Organic Carbon), S5 (Mineral Carbon) are measured to calculate 90 the Total Organic Carbon (TOC, in %), Hydrogen Index (HI, in mg HC/g TOC), and to 91 characterize the kerogen types and other parameters. Laboratory procedures 92 followed Behar et al. (2001). Around 50 mg of homogenized sample were weighed 93 into crucibles and measured following a temperature profile of 300-650°C and 300-94 850°C for the pyrolysis and oxidation ovens, respectively. The IFP 160000 Reference 95 Standard and the in-house SAB134 (Blue Lias organic-rich marl) standard were 96 regularly measured (every 10 samples for the in-house standard). The standard 97 deviation on TOC and HI of the in-house SAB134 and the reference IFP160000 98 standards is, respectively, 0.065% and 0.066% (TOC) and 22.65 mg HC/g TOC and 99 10.64 mg HC/g TOC (HI). The studied sediments are likely over-mature, with 100 measured Tmax values of up to 577°C and with Production Index (PI) values being 101 close to 1 in many samples and over 0.4 in all samples (Supplementary Materials). 102

103 [1.5] Mercury (Hg) analyses

104 Sedimentary mercury concentrations were analyzed on a RA-915 Portable 105 Mercury Analyzer with PYRO-915 Pyrolyzer (Lumex), at the Department of Earth 106 Sciences, University of Oxford, adapted from the methods described in Bin et al. 107 (2001) and Percival et al. (2015). For each sample, about 50±2 mg of rock powder 108 was weighed out and the precise mass recorded. For Hg-poor samples, about 100±2 109 mg of rock powder were analyzed. The material was heated in the Pyrolyzer to 110 700°C, volatilizing the mercury in the sample. The mercury gas was subsequently 111 entered into the Analyzer, where its abundance was measured, allowing calculation 112 of Hg concentration in the original mass of sample. The machine was calibrated

before use with six standards of peat (NIMT/UOE/FM/001 – Inorganic Elements in
Peat) with Hg concentration of 169±7 ppb, of varying masses between 20 and 80
mg. During analysis, a further peat sample was analyzed after every ten rock
samples to ensure continuity.

Previous Hg analyses from the Lower Jurassic sediments in the Arroyo 117 118 Alumbre succession of the Neuguén Basin also showed relatively elevated Hg and 119 Hg/TOC values (Percival et al., 2017). The expanded upper Triassic to lower 120 Jurassic data-set presented here shows that some samples from the upper Triassic 121 part of the succession are characterized by low Hg and/or TOC concentrations 122 (Figure 2). The relative uncertainty in Hg and TOC analyses increases in samples 123 with low respective concentration values. Hg/TOC values from samples with TOC < 124 0.25% and Hg < 5 ppb are therefore considered not trustworthy. Relatively elevated 125 sedimentary Hg and Hg/TOC values, from samples with TOC > 0.25% and Hg > 5 126 ppb, already onset before the negative CIE associated with the end-Triassic mass 127 extinction (Figure 2).

128

### 129 [1.6] Hand-Held X-Ray fluorescence (hh-XRF)

130 Hand-held X-Ray Fluorescence (hh-XRF) analysis was performed with the 131 Olympus Innov-X Delta Premium XRF Analyzer, under climate-controlled laboratory 132 conditions. The analyses were executed on both the flat surface of several cm<sup>3</sup>-133 sized, hand specimens, as well as on the powdered material of each sample, 134 throughout the studied section. The HH-XRF was run in 2 Beam Mining Mode (DS-135 6000) (each beam had a 40-sec measurement window; at 15 kV and 40 kV, 136 respectively), and standard calibration to a manufacturer-delivered standard metal 137 clip was performed at the onset of each run. Additionally, the NIST-2702 and NIST-138 2781 certified reference standards were regularly measured for quality control on the 139 measurements, for calibration for long-term drift and measurement offset and to 140 constrain measurement errors for the individual elements of interest. Frequent

141 measurements of the NIST-2702 standard show for calcium an average of 0.35%

- 142 (NIST-2702 = 0.34%), with a standard deviation of 0.01%; for sulphur an average of
- 143 0.83% (NIST-2702 = 1.5%), with a standard deviation of 0.01%; and for molybdenum
- an average of 11ppm (NIST-2702 = 11.4ppm), with a standard deviation of 1.1ppm.
- 145 Frequent measurements of the NIST-2781 standard show for calcium an average of
- 4.72% (NIST-2781 = 3.9%), with a standard deviation of 0.05%; for sulphur an
- average of 1.10%, with a standard deviation of 0.02%, and for molybdenum an
- average of 26.0ppm (NIST-2781 = 46.7ppm), with a standard deviation of 1.7ppm.
- 149

### 150 [2] SIMPLE MASS-BALANCE CACULATIONS ON TRIASSIC–JURASSIC

### 151 CARBON-CYCLE EVOLUTION

152 The required release of isotopically light carbon, and the required changes in 153 the fluxes between exogenic carbon reservoirs to explain the observed carbon cycle 154 evolution across the Triassic–Jurassic transition, including the ~2.5‰ negative 155 carbon isotope excursion (CIE) at the end-Triassic mass extinction and the 156 subsequent positive CIE straddling the Triassic–Jurassic boundary, is tested using a 157 simple mass balance calculation, under scenarios (1-4. Supplementary Figure 3): 158 159 •  $d(\delta(Ex))/dt = ((F_{additional}/M_{ex}) \times (\delta_{additional} - \delta_{ex})) + ((F_{in}/M_{ex}) \times (\delta_{in} - \delta_{ex})) -$ 160  $((F_{out}/M_{ex}) \times (\delta_{out} - \delta_{ex}))$ 161 162 M<sub>ex</sub> = Mass of exogenic carbon reservoirs (in Gt)  $\delta_{ex} = \delta^{13}C$  composition of exogenic carbon reservoirs (in ‰) 163 \_ 164 - F<sub>in</sub> = Flux in (amount of carbon added to exogenic carbon reservoirs (in Gt/ 165 1000 yr) -  $\delta_{in} = \delta^{13}C$  composition of F<sub>in</sub> (in ‰) 166 - F<sub>out</sub> = Flux out (amount of carbon extracted from exogenic carbon reservoirs 167 (in Gt/ 1000 yr) 168

169	$-\delta_{out} = \delta^{13}C$ composition of $F_{out}$ (in ‰)
170	- F <sub>additional</sub> = Additional flux in (additional amount of carbon added to exogenic
171	carbon reservoirs (in Gt/ 1000 yr)
172	$- \delta_{\text{additional}} = \delta^{13} \text{C composition of } F_{\text{additional}} \text{ (in \%)}$
173	
174	(with $M_{ex}$ and $\delta_{ex}$ at t=0 given as 60,000 Gt and -1 ‰, respectively, and other
175	parameters forced as given in Supplementary Figure 3).
176	
177	[3] SUPPLEMENTARY FIGURES
178	Supplementary Figure 1 Early Jurassic (Pliensbachian–Toarcian) basin
179	geometry and facies distribution of the Neuguén Basin, Argentina (modified after
180	Riccardi et al., 2011).
181	
182	Supplementary Figure 2-A Location of the Arroyo Alumbre section in the Neuquén
183	Basin, Argentina (images from Google Earth).
184	
185	Supplementary Figure 2-B Location of the Arroyo Alumbre section in the Neuquén
186	Basin, Argentina (images from Google Earth).
187	
188	Supplementary Figure 2-C Outcrop photos of the Arroyo Alumbre section in the
189	Neuquén Basin, Argentina.
190	
191	Supplementary Figure 2-D Outcrop photos of the Arroyo Alumbre section in the
192	Neuquén Basin, Argentina, with the stratigraphic position of the studied samples.
193	
194	Supplementary Figure 2-E Outcrop photos of the Arroyo Alumbre section in the
195	Neuquén Basin, Argentina, with the stratigraphic position of the studied samples.
196	

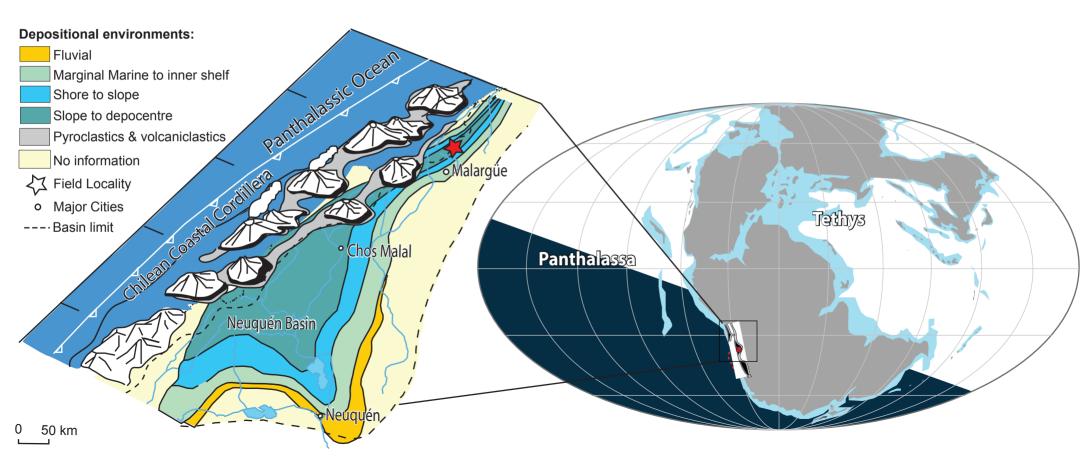
198 reservoirs across the Triassic-Jurassic transition, based on simple mass-balance 199 calculations. The impact of timed volcanogenic-carbon release (with  $\delta^{13}C = -5\%$ ), 200 biogenic methane release from dissociation of ocean-floor clathrates (with  $\delta^{13}C = -$ 60‰), and elevated organic-carbon burial (with  $\delta^{13}C = -35\%$ ), are evaluated 201 202 separately. Colours represent the different scenarios and model outputs given in 203 Figure 6 of the main text. 204 205 [4] REFERENCES 206 Behar, F., Beaumont, V., De B. Penteado, H.L., Rock-Eval 6 Technology: 207 Performances and Developments. Oil & Gas Science and Technology – Rev. 208 IFP 56 (2), 111–134 (2001). 209 Bin, C., Xiaoru, W., Lee, F.S.C., Pyrolysis coupled with atomic absorption 210 spectrometry for the determination of mercury in Chinese medical materials. 211 Anal. Chim. Acta 447, 161-169 (2001). 212 Damborenea, S.E., Echevarría, J., Ros-Franch, S., Biotic recovery after the end-213 Triassic extinction event: Evidence from marine bivalves of the Neuguén Basin, 214 Argentina. Palaeogeography, Palaeoclimatology, Palaeoecology 487, 93-104 215 (2017). 216 Damborenea, S.E., Manceñido, M.O., Late Triassic bivalves and brachiopods from 217 southern Mendoza, Argentina. Revue de Paléobiologie, Genève 11, 317-344 218 (2012). 219 Percival, L.M.E., Ruhl, M., Hesselbo, S.P., Jenkyns, H.C., Mather, T.A., Whiteside, 220 J.H., Mercury evidence for pulsed volcanism during the end-Triassic mass 221 extinction. PNAS 114, 7929-7934 (2017). 222 Percival, L.M.E., Witt, M.L.I., Mather, T.A., Hermoso, M., Jenkyns, H.C., Hesselbo, S.P., Al-Suwaidi, A.H., Storm, M.S., Xu, W., Ruhl, M., Globally enhanced 223 224 mercury deposition during the end-Pliensbachian and Toarcian OAE: A link to

**Supplementary Figure 3** Model output of the  $\delta^{13}$ C evolution of exogenic carbon

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### **SUPPLEMENTARY FIGURE 1**



### SUPPLEMENTARY FIGURE 2 A

Triassic–Jurassic boundary section at Arroyo Alumbre: 34°49'767"S – 69°53'224"W (Google-maps coordinates: -34.827675, -69.888884)

Drive from El Sosneado, along the Rio Atuel, to the entrance of the Arroyo Malo Valley (dirth road)



Walking route to the Triassic–Jurassic transition in the Arroyo Alumbre valley (Side valley of the Arroyo Malo, which enters into the Rio Atuel)

Arroyo

Rio Atuel

Triassic–Jurassic boundary section in the Arroyo Alumbre valley

— Car parking along the main dirt-road or in a small lake/salt bed close to the entrance of the Arroyo Malo valley

### SUPPLEMENTARY FIGURE 2-B

Triassic–Jurassic boundary section at Arroyo Alumbre: 34°49'767''S – 69°53'224''W (Google-maps coordinates: -34.827675, -69.888884)



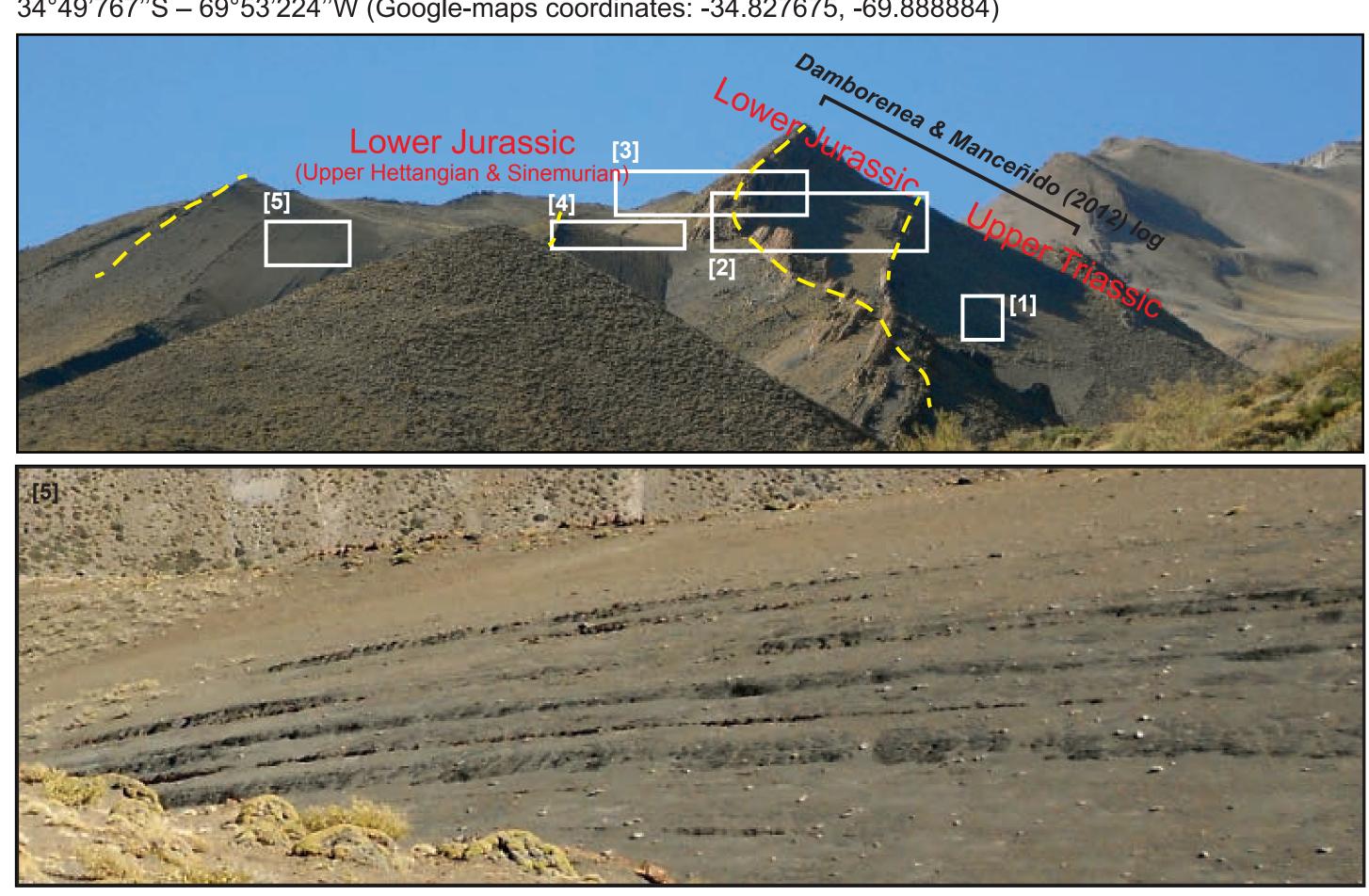
Triassic–Jurassic boundary section in the Arroyo Alumbre valley

Google Earth

Arroyo Alumbre

### SUPPLEMENTARY FIGURE 2-C

Triassic–Jurassic boundary section at Arroyo Alumbre: 34°49'767''S – 69°53'224''W (Google-maps coordinates: -34.827675, -69.888884)





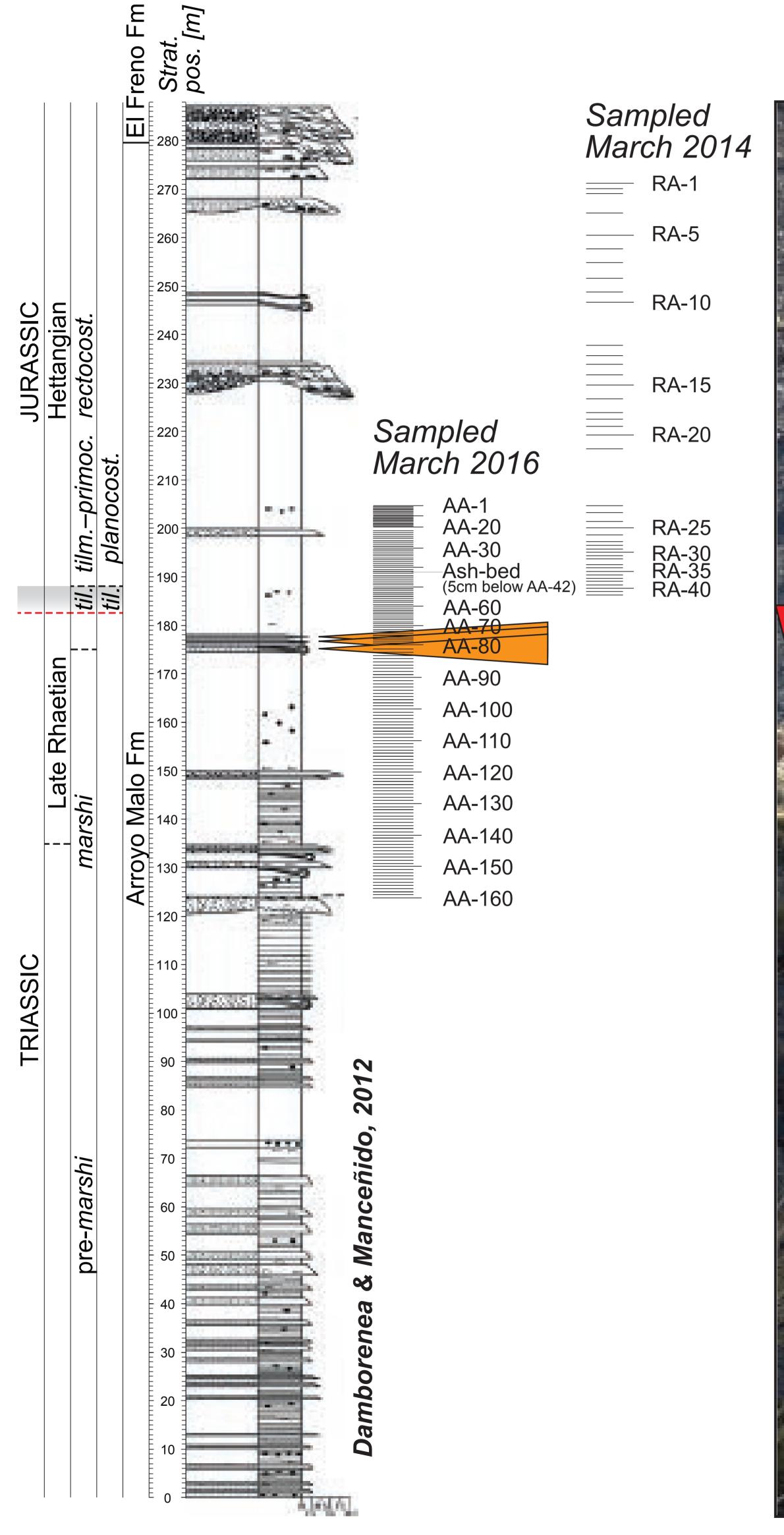






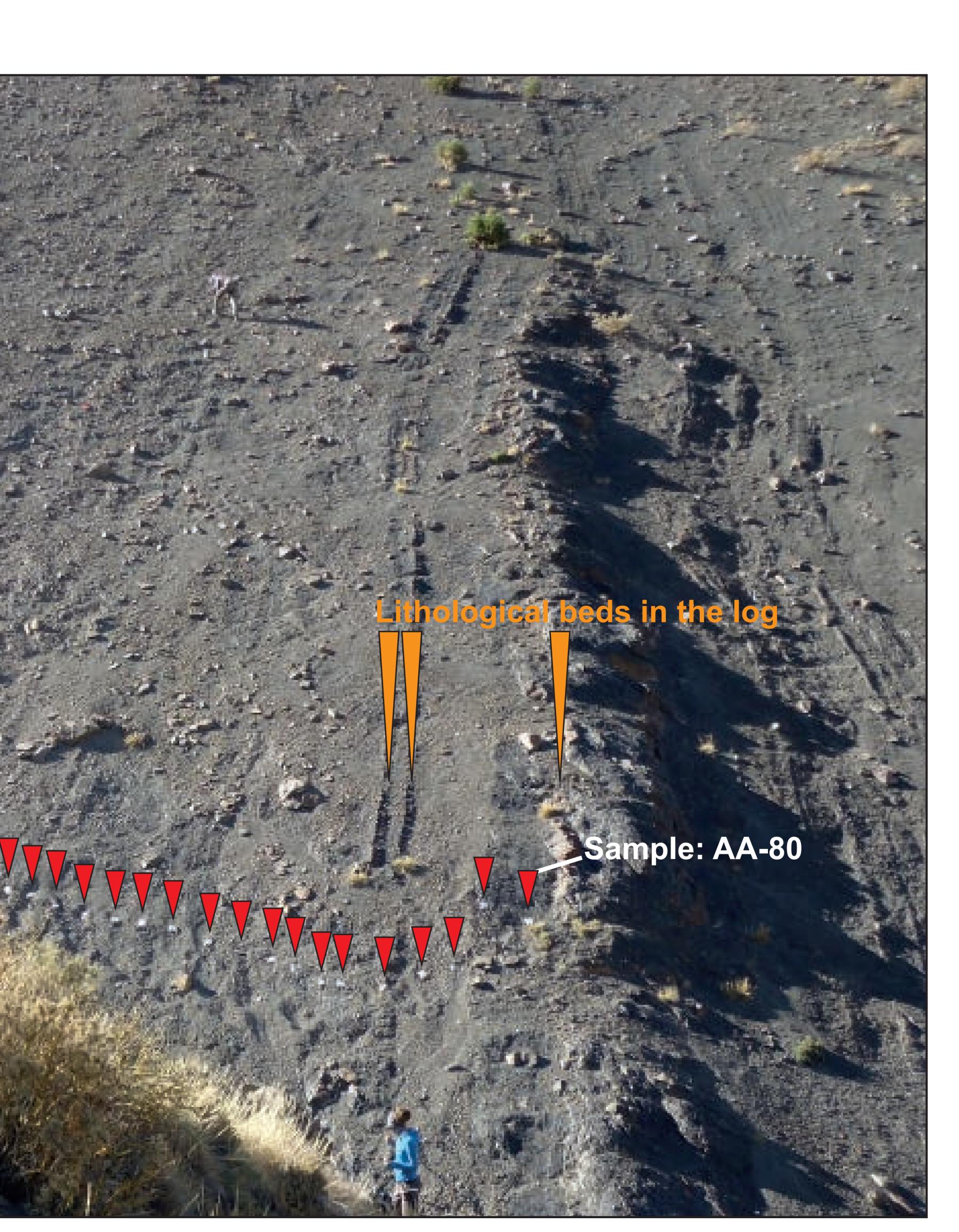
# SUPPLEMENTARY FIGURE 2-D

### Triassic–Jurassic boundary section at Arroyo Alumbre: 34°49'767''S – 69°53'224''W (Google-maps coordinates: -34.827675, -69.8888884)



Sample: AA-42

Position of ash-bed Sample: AA-43



### SUPPLEMENTARY FIGURE 2-E

### Triassic–Jurassic boundary section at Arroyo Alumbre: 34°49'767''S – 69°53'224''W (Google-maps coordinates: -34.827675, -69.888884)

