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Mercury contents and isotope ratios from diverse depositional environments across the Triassic–Jurassic boundary

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1 Mercury contents and isotope ratios from diverse depositional environments

2 across the Triassic–Jurassic Boundary: Towards a more robust mercury proxy

3 for large igneous province magmatism

4 **Joyce A. Yager[1,^], A. Joshua West[1], Alyson M. Thibodeau[2], Frank A. Corsetti[1],**

5 **Manuel Rigo[3], William M. Berelson[1*], David J. Bottjer[1*], Sarah E. Greene[4*], Yadira**

6 **Ibarra[1,5*], Flavio Jadoul[6*], Kathleen A. Ritterbush[7*], Nick Rollins[1*], Silvia Rosas[8*],**

7 **Pietro Di Stefano[9*], Debbie Sulca[1,10*], Simona Todaro[9*], Peter Wynn[1,11*], Laura**

8 **Zimmermann[12*], Bridget A. Bergquist[12]**

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10 *[1]University of Southern California, Department of Earth Sciences*

11 *[2]Dickinson College, Department of Earth Sciences*

12 *[3]University of Padova, Department of Geosciences*

13 *[4]University of Birmingham, School of Geography, Earth and Environmental Sciences*

14 *[5]San Francisco State University, College of Science and Engineering*

15 *[6]Dipartimento di Scienze della Terra 'Ardito Desio', Universitá degli Studi di Milano*

16 *[7] University of Utah, Department of Geology and Geophysics*

17 *[8]Pontificia Universidad Católica del Perú, Geological Engineering Program*

18 *[9]Università degli Studi di Palermo, Department of earth and marine science (DiSTeM)*

19 *[10]University of California, Davis, Department of Environmental Toxicology*

20 *[11]University of Washington, Department of Earth and Space Sciences*

21 *[12] University of Toronto, Department of Earth Sciences*

22 *These authors are listed in alphabetical order

23 ^Corresponding author: joyceannyager@gmail.com

ABSTRACT

Mercury is gaining prominence as a proxy for large igneous province (LIP) volcanism in the sedimentary record. Despite temporal overlap between some mass extinctions and LIPs, the precise timing of magmatism relative to major ecological and environmental change is difficult to untangle, especially in marine settings. Changes in the relative contents of Hg in sedimentary rocks through time, or 'Hg anomalies', can help resolve the timing of LIP activity and marine extinctions. However, major questions remain unanswered about the fidelity of Hg as a proxy for LIP magmatism. In particular, depositional (e.g., redox) and post-depositional (e.g., oxidative weathering) processes can affect Hg preservation in marine sediments. These factors pose challenges for confidently using Hg as a fingerprint of volcanism. Here, we use the Hg anomaly at the Triassic–Jurassic boundary to explore the opportunities and challenges associated with two approaches that may help build a more robust interpretation of the Hg proxy: (1) measurements from sediments from diverse depositional environments, including lithologies with low Hg and organic carbon content, and (2) the simultaneous use of Hg stable isotope ratios. We present and compare Hg records from five geographically disparate Upper Triassic–Lower Jurassic marine sections that represent nearshore, mid-shelf, deep-water, and carbonate platform settings. These sedimentary sections span the emplacement of the Central Atlantic magmatic province (CAMP) and the associated end–Triassic extinction (ETE). Total organic carbon contents, carbonate contents, and Hg contents and stable isotope compositions demonstrate the multiple ways in which different depositional environments impact how Hg anomalies are expressed in ancient marine sedimentary rocks. Although we observe an increase in Hg/TOC during the ETE in each

2

47 section, the pattern and duration of Hg enrichment differ notably between sections, and the

48 timing is not always coincident with CAMP activity, illustrating how the depositional filter

49 complicates the use of Hg/TOC ratios alone as a fingerprint of LIP magmatism. In

50 addition, Hg isotope measurements support a volcanic origin for these Hg

51 anomalies during the ETE, suggesting CAMP was the Hg source during the extinction

52 interval. These data support the use of Hg isotopes to help distinguish Hg loading that

53 results from LIP magmatism on a global scale and emphasize the importance of making Hg

54 proxy measurements from diverse depositional environments.

55

56

57 **1. INTRODUCTION**

58   The prevalence of mass extinctions coincident with large igneous province (LIP)

59 activity motivates a thorough understanding of the connections between magmatism and

60 global environmental change (e.g., Bond and Wignall, 2014). Extinctions may be caused

61 or enhanced by the cascade of effects following the release of volcanic gases (e.g., S, CO_2)

62 in high quantities over geologically short periods of time. These gases perturb the Earth

63 system, potentially causing ocean acidification, ocean anoxia, and warming (or cooling),

64 all of which may be detrimental to metazoan life (e.g., Bond and Wignall, 2014).

65 Similarities between mass extinction–LIP emplacement intervals and modern

66 anthropogenic CO_2 emissions underscores their relevance today (e.g. Foster et al., 2018).

67 However, correlation between terrestrial sections (where LIPs often occur) and marine

68 sections (where many of the extinction records occur) is typically not straightforward,

69 motivating further work to better understand the relationship between LIP emplacements

3

70 and related extinctions. Mercury preserved in sedimentary rocks may help link LIP

71 magmatism with biotic and other environmental changes observed in the marine

72 sedimentary record, because Hg can be measured in the same sedimentary records that

73 preserve paleontological evidence (e.g., Grasby et al., 2016, 2019; Percival et al., 2015;

74 Sanei et al., 2012; Scaife et al., 2017; Shen et al., 2019a, b; Thibodeau et al., 2016, amongst

75 many other recent studies).

76   In the absence of anthropogenic perturbation, Hg is primarily introduced to the

77 atmosphere, ocean, and sedimentary record through volcanic inputs (e.g., Fitzgerald et al.

78 2014). In the geologic record, periods of high volcanic activity and associated increased

79 input to the Earth system may be reflected in sedimentary intervals with unusually high Hg

80 enrichments, termed 'Hg anomalies' (Sanei et al., 2012). Since Hg is often associated with

81 organic matter in sediments, recent literature has focused on identifying Hg anomalies

82 based on ratios to total organic carbon (TOC; reviewed by Grasby et al., 2019). The ease

83 of measuring Hg and TOC contents in sedimentary rocks has led to a rapid proliferation in

84 the application of this proxy across Phanerozoic LIP and mass extinction events.

85   As its application has grown, so too has the understanding of the complexity of

86 factors that can influence Hg contents and Hg/TOC ratios in sedimentary rocks. The

87 reliability of Hg contents as a proxy for LIP volcanism is significantly impacted by its host

88 phase, which is often organic matter but can also be clays or sulfur species (e.g., Shen et al

89 2019a,b). Redox conditions and diagenesis impact the formation and preservation of these

90 host phases and are all at least partially controlled by depositional environment.

91 Additionally, post-depositional fluid flow and weathering can redistribute Hg, altering

92 primary contents (Charbonnier et al., 2020). These multiple confounding factors may erase

4

93   the record of volcanic loading or generate Hg enrichments unrelated to volcanism (Shen et

94   al., 2019a), raising fundamental questions about whether increases in Hg content and/or

95   Hg/TOC ratios in sedimentary sections can be used as a simple and reliable fingerprint of

96   secular changes in volcanism.

97   The recognition of complexities is a vital step in the development of robust

98   geochemical proxies for past environmental change; an important next step is to explore

99   approaches and techniques that may provide for reliable interpretations despite the inherent

100   complexity (Elderfield, 2011). In this study, we explore two of the many possible directions

101   that may help develop a more robust Hg proxy. First, we ask whether measurements from

102   rock types that are low in Hg and TOC may help to broaden the applicability of the proxy,

103   as different depositional environments may differ in their deposition of sulfides, clays, and

104   TOC, and indirectly Hg contents. Mercury measurements are scarce in organic-poor

105   lithologies, in part because they are more difficult to measure and in part because prior

106   work has established thresholds of TOC content below which Hg/TOC ratios are regarded

107   as unreliable; however, this seems likely to be a product of ill-suited TOC methods and

108   difficulty interpreting low TOC samples, and we address both here. Second, and in tandem,

109   we evaluate how Hg isotopes may add vital but widely under-utilized information for

110   fingerprinting volcanism in the sedimentary record. Understanding how Hg isotope ratios

111   in sedimentary rocks are affected by local and regional cycling and depositional

112   environments remains incomplete yet critical to extracting the most from the

113   complementary information that they offer.

114   We address these two major questions by focusing on the Triassic–Jurassic

115   boundary (TJB), which is associated with one of the major Phanerozoic mass extinctions

116   and one of the iconic Hg anomalies that has enabled linking of the end-Triassic extinction

117   (ETE) with LIP activity associated with the Central Atlantic magmatic province, or

118   (CAMP) (Percival et al., 2017; Thibodeau et al., 2016). To this end, we present new records

119   of Hg content and isotopic compositions from diverse depositional settings across this time

120   interval. In the sections that follow, we begin with a review of the Hg proxy and its

121   complexities; we follow by introducing TJB sedimentary sections that are focus of our

122   study before presenting new records and discussing the implications of our data for use of

123   the Hg proxy.

124

125   **2. BACKGROUND**

126   **2.1 Hg as a proxy for LIP magmatism in the sedimentary record**

127   Volcanic emissions comprise ~75% of present-day natural Hg emissions, making

128   them the primary natural source of Hg to surficial reservoirs on Earth (Pyle and Mather,

129   2003). Mercury has an approximately 1 year residence time in the atmosphere, which

130   enables its global distribution and deposition (Schroeder and Munthe, 1998; Lindberg et

131   al., 2002; Douglas et al., 2008). From the atmosphere, Hg deposits into terrestrial and

132   marine environments where it can be cycled back to the atmosphere between terrestrial and

133   marine environments, with ultimately a major sink in marine sediments. Today, Hg has a

134   deep-ocean residence time of ca. 350 years, much shorter than the whole-ocean mixing

135   time (ca. 1000 years), and the surface ocean has an even shorter residence time (Gill and

136   Fitzgerald, 1988). As a result, periods of intense volcanism can increase the global

137   atmospheric Hg load and leave a systematic imprint on marine and terrestrial systems.

138   Once emitted, volcanogenic Hg can enter the marine system via atmospheric

139   deposition or terrestrial runoff, and in both cases Hg is thought to be transferred to marine

140   sediments mostly complexed with organic compounds and other particles (e.g., Kongchum

141   et al., 2011; Zhong et al., 2008). Thus, Hg contents in marine sediments are highly

142   dependent on the local organic load and/or dilution by non-Hg-bearing detrital sediments

143   (Grasby et al., 2013), and local effects may dominate the Hg signature in coastal marine

144   sediments. Although Hg is often associated with organic matter, in modern and ancient

145   sediments it can also be associated with sulfides (e.g., Huerta-Diaz and Morse, 1992; Shen

146   et al., 2019a; Shen et al., 2020) or clays (e.g., Them et al., 2019), depending on the redox

147   conditions during and immediately after deposition.

148   Due to the particle-reactive nature of Hg accumulation in sediments, Hg contents

149   are typically normalized to a host phase in order to identify changes in input to the

150   atmosphere-ocean system, such as that from volcanic loading. Given the predominant

151   associated of Hg with organic matter, total organic carbon (TOC) contents are widely used

152   for normalization to ensure that observations of high Hg contents are not simply a product

153   of high sedimentary TOC deposition in sediments. Observations of increases in Hg/TOC

154   in multiple stratigraphic sections are hypothesized to reflect globally increased

155   magmatism, and can help to relate the timing of magmatism with environmental and biotic

156   changes recorded in the same sections (e.g., Percival et al., 2017; Grasby et al.,2 019).

157   Previous studies applying the Hg proxy have focused on sedimentary rocks with

158   significant TOC content, which can easily be analyzed via thermal combustion methods.

159   Previous work (recently reviewed by Grasby et al (2019)) excluded Hg/TOC ratios from

160   samples with <0.2% TOC. This threshold is presumably a holdover from analytical

161 methods that are unable to resolve lower TOC (e.g., Rock-eval pyrolysis); subsequent

162 studies have adopted this cutoff regardless of the TOC analysis method and its detection

163 limit. Many methods, such as those that use elemental analyzers, are capable of quantifying

164 low TOC contents and have been reported in other studies (e.g., Fujisaki et al., 2018; Galy

165 et al., 2007, 2008; Hilton et al., 2010; Scheingross et al., 2021) Here, we use a method that

166 can quantify low TOC samples and further describe its capabilities and limitations in the

167 supplement. Quantifying Hg/TOC ratios in low-TOC samples could open possibilities of

168 working in a wider range of lithologies across a wider range of depositional environments

169 than those available when restricted to samples with 0.2 wt.% TOC. Records from diverse

170 lithologies across the same period of time could help to avoid 'false positive' Hg anomalies

171 related to depositional artifacts. However, the challenge with this approach is not just

172 analytical; the meaning of Hg/TOC values in samples with very low TOC remains to be

173 fully understood.

174 **2.2 Hg isotopes and the volcanic Hg proxy**

175 Mercury isotopes can help identify the source of Hg in sediments and/or the pathway

176 of deposition (i.e., atmospheric versus runoff). In particular, isotopic information can

177 potentially identify anomalously high Hg contents originating from LIP magmatism

178 (Thibodeau and Bergquist, 2017; Bergquist 2017). Mercury has seven stable isotopes and

179 undergoes both mass dependent fractionation (MDF) and mass independent fractionation

180 (MIF). Mass dependent fractionation in Hg isotopes is typically reported using the

181 ^{202}Hg/^{198}Hg ratio as δ^{202}Hg (where δ^{202}Hg (‰) = [(^{202}Hg/^{198}Hg)$_{sample}$/(^{202}Hg/^{198}Hg)$_{standard}$ −

182 1] × 1000). The NIST 3133 standard is widely used for Hg isotope measurement and is

183 adopted here (Blum and Bergquist, 2007). Mass independent fractionation is measured as

184   the difference between another measured δ value (e.g., δ^{199}Hg) and the MDF predicted

185   using the measured δ^{202}Hg value and the kinetic MDF law; this difference is typically

186   reported as Δ^{199}Hg for odd isotope MIF.

187   Gaseous elemental Hg emitted from volcanoes enters the atmosphere with no

188   measurable MIF (Δ^{199}Hg = 0‰; Fig. 1a) and with negative to zero MDF (δ^{202}Hg = –2‰

189   to 0‰; e.g., Zambardi et al., 2009; Szponar et al., 2020; Si et al., 2020). After oxidation of

190   elemental Hg to more reactive and particle-reactive species, Hg can acquire odd MIF

191   mostly via photochemical reduction either in cloud droplets, surface waters, or on surfaces

192   in terrestrial settings (Thibodeau and Bergquist, 2017; Bergquist 2018). This results in

193   gaseous elemental Hg in the atmosphere having negative odd MIF compositions and

194   oxidized Hg species having positive odd MIF signatures (Blum et al., 2014). Because much

195   of the Hg accumulated in terrestrial soils is from uptake of gaseous elemental Hg by

196   vegetation (Zhou et al., 2021), terrestrial soils and surfaces typically preserve negative odd

197   MIF values (i.e., negative Δ^{199}Hg) and much more negative δ^{202}Hg or "negative MDF" in

198   biomass and soils due to the preferential uptake of the lighter Hg isotopes (Demers et al.,

199   2013). Nearshore sediments that acquire Hg through terrestrial runoff thus typically exhibit

200   negative MIF and more negative MDF Hg isotope values (Demers et al., 2013; Zheng et

201   al., 2016; Thibodeau and Bergquist, 2017; see Fig. 1a). In contrast, settings further from

202   the continent usually contain small odd-isotope excesses (i.e., positive Δ^{199}Hg and Δ^{201}Hg

203   values, or "positive MIF" and less negative δ^{202}Hg), which are from the deposition of

204   oxidized Hg species (Thibodeau and Bergquist, 2017; Bergquist and Blum, 2007; Sonke

205   2011).

206   Thibodeau et al. (2016) measured Hg isotopes in shallow marine sediments

207   spanning the Triassic–Jurassic boundary (TJB) and speculated that the absence of MIF

208   observed in these rocks during the end–Triassic extinction was due to increased volcanic

209   loading of Hg associated with the central Atlantic magmatic province (CAMP). Since then,

210   other studies have reported Hg isotope values associated with Hg anomalies during other

211   LIPs, including at the end–Permian (Grasby et al., 2017; Shen et al., 2019a,b; Wang et al.,

212   2018), during the Toarcian ocean anoxic event (Them et al., 2019), across the Ordovician–

213   Silurian boundary (Gong et al., 2017), and at the Cretaceous–Paleogene boundary (Sial et

214   al., 2016). Since volcanic Hg often initially exhibits negative MDF and MDF can be altered

215   by many other environmental processes, MDF is not as diagnostic as MIF in fingerprinting

216   magmatically sourced Hg. Here we focus our discussion on MIF signatures, which are

217   thought to be more useful for fingerprinting volcanic Hg.

218   **2.3 Complications in using the Hg proxy for LIP magmatism**

219   Although Hg contents and isotopes are a promising proxy in many respects, a

220   growing number of studies are revealing that multiple, complex processes may influence

221   the preservation and magnitude of Hg and Hg/TOC anomalies. Release of Hg during LIP

222   activity may be influenced by magmatic style as well as non-volcanic sources such as fires

223   on land or release of sedimentary Hg by magmatic intrusions and emplacement (Grasby et

224   al., 2017). Periods of LIP activity were also likely characterized by both intrusive and

225   extrusive magmatism, and by eruptions that ranged from the subaerial to the submarine. It

226   is unclear how these different styles of eruption would have affected the release and

227   deposition of Hg and the resulting signals recorded in sedimentary records. It is possible

228   that subaerial, explosive volcanism may have led to more Hg in globally distributed

229 depositional environments, while submarine and effusive volcanism may not have

230 perturbed the global Hg cycle to the same extent (Percival et al., 2018).

231   Additional complexity in Hg records also results from depositional and diagenetic

232 processes (Percival et al., 2018; Charbonnier et al., 2020), including local redox

233 fluctuations (Shen et al., 2019a). Some apparent anomalies may be better explained by

234 local redox variability than by volcanic emissions of Hg (Them et al., 2019; Shen et al.,

235 2019a), and the known association between Hg and sulfur in many sedimentary rocks

236 (Huerta-Diaz and Morse, 1992; Shen et al., 2020) and clays (Manceau et al., 2018; Shen et

237 al., 2019a, b) raises questions about whether TOC normalization alone accounts for the key

238 processes driving Hg enrichment or depletion in some environments (Sanei et al., 2012;

239 Shen et al., 2019a; Shen et al., 2020).

240   Given the complexity of Hg contents and Hg/TOC ratios in marine sediments, Hg

241 isotopes may offer key complementary information for fingerprinting LIP magmatism.

242 Despite a large number of studies measuring Hg contents and Hg/TOC ratios in a range of

243 depositional environments, there are few studies that utilize Hg isotopes to understand the

244 origin of Hg anomalies across depositional settings. A systematic investigation of Hg

245 isotopes in different depositional settings from the sedimentary record is important to

246 further understand the role of LIP magmatism (among other processes) in driving changes

247 in sedimentary Hg. Understanding such effects is particularly important because multiple

248 records of Hg, Hg/TOC, and Hg isotopes from the same time period are necessary to

249 demonstrate that perturbations to the Hg cycle are indeed global and not local (Percival et

250 al., 2017). Understanding site-dependent effects associated with depositional environment

251 may be critical to evaluating similarities and differences between sections. Here, we

252 explore how the depositional environment affects Hg contents and the Hg isotope signal

253 across four different sections spanning the TJB, when a well-defined Hg anomaly has been

254 previously identified and tied to LIP activity (Thibodeau et al., 2016; Percival et al., 2017).

255 **2.4 The Triassic–Jurassic Transition: A testbed for the Hg proxy**

256 The CAMP (Marzoli et al., 1999) was a subaerial magmatic province emplaced

257 over approximately one million years during the Late Triassic and Early Jurassic (Davies

258 et al., 2017; Blackburn et al., 2013; Knight et al., 2004). Intrusive activity began ca. 100

259 kyrs before the dominant extrusive activity (Davies et al., 2017). Emplacement was likely

260 characterized by 3–4 main pulses (Blackburn et al., 2013) of durations somewhere between

261 1–100 kyrs (Schaller et al., 2012; Berner and Beerling, 2007).

262 The end–Triassic extinction (ETE; 201.56 Ma) coincided with CAMP activity

263 (Marzoli et al., 2004; Schoene et al., 2010; Wotzlaw et al., 2014). The record of the ETE

264 is largely preserved in marine sediments, in which direct evidence of CAMP activity at

265 many localities is scarce. Thus, our understanding of the ETE and the TJB transition can

266 be greatly enhanced by the successful application of the Hg proxy, if Hg provides a

267 fingerprint of CAMP volcanism that can be related directly to the marine record of

268 extinction across multiple stratigraphic sections (Percival et al., 2017; Thibodeau et al.,

269 2016). However, records of Hg contents from this time are different at each section that

270 has been measured to date. Mercury contents and Hg/TOC differ by orders of magnitude

271 from section to section, relative increases (i.e., anomaly severity) in each section are

272 different, and the relative timing of anomalies based on chemo- and biostratigraphy and

273 the extinction record are not consistent, raising questions about how increases in Hg or

274 Hg/TOC can be directly interpreted as fingerprints of LIP volcanism (Kovács et al., 2020;

275 Lindström et al., 2019; Percival et al., 2017; Ruhl et al., 2020; Thibodeau et al., 2016).

276   In this study, we target five sedimentary sections that span the TJB and represent a

277 range of depositional environments, from shallow through deep marine (here, we use the

278 term "deep marine" to refer settings that are still on the continental margin but well below

279 storm wave base in basinal settings). We also consider a carbonate platform. We report Hg

280 contents, Hg/TOC ratios, and Hg isotopes from these localities and explore how differences

281 and similarities in their Hg records may be tied to depositional setting and associated

282 sedimentary processes.

283

284 **3. OVERVIEW OF LOCALITIES**

285   We collected samples and measured Hg from five open marine sections (although

286 we note some debate exists for St. Audrie's Bay) spanning the Triassic–Jurassic boundary

287 from disparate paleogeographic locations (Fig. 1a) and from a range of depositional

288 settings with variable expected influences on the Hg proxy (Fig. 1b). We deliberately chose

289 depositional settings characterized by lithologies that have been widely used in applying

290 the sedimentary Hg proxy (specifically, rocks with substantial TOC) as well as those that

291 have been less well studied, including carbonate-dominated rocks with low TOC. In

292 working with low-TOC carbonates, our goal was to explore both analytical and interpretive

293 limits.

294   Most samples have a corresponding thin section for further lithologic comparison.

295 Each locality has a well-characterized bio- and/or chemostratigraphic context; the position

296 of the ETE and Triassic–Jurassic boundary are denoted in Figs. 2–7 and are based on

13

297   previous studies. We correlated each section and demarcated the end–Triassic extinction

298   based on the negative carbon isotope excursion seen in organic matter at each section (e.g.,

299   Yager et al., 2017; their fig. 2). There is currently a lively debate regarding the source of

300   the organic carbon isotope excursion (e.g., Lindström et al. 2012), the ability to correlate

301   it between ocean basins (Fox et al., 2020), and its relationship to the ETE extinction.

302   However, given the presence of the initial carbon isotope excursion in Peru (Yager et al.,

303   2017), Canada (Williford et al, 2007), Nevada (Guex et al., 2004; Ward et al., 2007), Japan

304   (Fujisaki et al., 2018), Argentina (Ruhl et al., 2020), and many European sections (e.g.,

305   Lindström et al. 2012; Fox et al. 2020) and that in each of those sections it coincides with

306   the onset of the mass extinction, we think using a biostratigraphic and C isotope-based

307   correlation is reasonable. In any case, we note that different possible correlations, such as

308   those suggested by Lindström et al. (2019) and Zaffani et al. (2018), will not change our

309   conclusions.

310   **3.1 New York Canyon, Nevada, USA**

311   The New York Canyon area of Nevada, USA includes a well-studied Panthalassic

312   record from the Triassic–Jurassic boundary (e.g., Taylor et al., 1983; Guex et al., 2004 and

313   references therein). The section is exposed on Ferguson Hill, which forms the northern

314   portion of Muller Canyon, is the auxiliary global stratigraphic section and point for the

315   period boundary (Bown et al., 2007; Morton and Hesselbo, 2008). The Gabbs Formation

316   and overlying Sunrise Formation comprise limestone, silty shale, and silty marl (Fig. 3),

317   with detailed biostratigraphy (the ammonite *Psiloceras spelae*; Guex et al., 2004)

318   indicating the Triassic–Jurassic boundary within the upper Gabbs Formation. Several

319   studies of the organic carbon (C) isotope signature ($\delta^{13}C_{org}$) at the Ferguson Hill section

320   (Bartolini et al., 2012; Guex et al., 2004; Ward et al., 2007; Corsetti et al., 2015) document

321   a negative excursion in $\delta^{13}C_{org}$ at the extinction horizon.

322   Upper Triassic rocks of the Gabbs Formation are interpreted to represent a robust

323   carbonate ramp (the Mount Hyatt Member), overlain by a finer-grained middle-shelf

324   deposit (the uppermost Muller Canyon Member). Lower Jurassic rocks first appear within

325   the Muller Canyon Member of the Gabbs Formation and persist into the increasingly thick

326   and resistant beds rich in limestone and chert, in the overlying Ferguson Hill Member of

327   the Sunrise Formation (Schoene et al., 2010), which are interpreted to represent a middle-

328   ramp setting on a distally-steepened bioclastic habitat (Ritterbush et al., 2014; Taylor et

329   al., 1983). We interpret the rocks in this section as indicative of a local upward-shallowing

330   trend, which began in the Late Triassic and continued into the Early Jurassic (Sinemurian).

331   The paucity of inorganic carbonate and other benthic bioclastic content throughout the

332   Muller Canyon Member has been attributed to either a global drop in carbonate saturation

333   (but see Greene et al., 2012) or to a loss of carbonate producers during the extinction

334   interval (Kiessling et al., 2007; Ritterbush et al., 2014). Overall, the New York Canyon

335   area represents deposition along the mid to inner shelf (Fig. 3) (Ritterbush et al., 2014).

336   **3.2 St. Audrie's Bay, UK**

337   The St. Audrie's Bay section has been well studied for over 100 years (e.g.,

338   Richardson, 1911, 1906, 1905). The ammonite *Psiloceras spelae,* which denotes the base

339   of the Jurassic (Guex et al., 2004, Guex et al., 2012), is absent in the section, so we rely on

340   the presence of the aforementioned negative CIE and subsequent positive shift in $\delta^{13}C$ (e.g.,

341   Yager et al., 2017; Lindstrom et al., 2012) to place the Triassic–Jurassic boundary at ~15

342   m in the St. Audrie's Bay section (similar to Hesselbo et al., 2004; we note the possibility

343 that this isotope excursion may also reflect a period of subaerial exposure as suggested by

344 Fox et al., 2020, but argue that this would not fundamentally change our correlation).

345   The section at St. Audrie's Bay includes dramatic changes in depth, TOC (wt.%),

346 and calcium carbonate content (wt.%). Hesselbo et al. (2004) provided a thorough

347 summary of the depositional changes seen at the section, which we briefly summarize here

348 (we followed the stratigraphy from Hesselbo et al., 2004, for our sampling and use their

349 stratigraphic column in Fig. 4). The lowermost ~2 m comprise the Williton Member of the

350 Blue Anchor Formation, which consists primarily of mudstone, has marine trace fossils

351 and bivalves, and is interpreted as shallow marine (Hesselbo et al., 2004; Mayall, 1981).

352 Meters ~2–12 comprise the Westbury Formation, which consists of mudstone and siltstone

353 with carbonate concretions and phosphatic conglomerate (Hesselbo et al., 2004;

354 Richardson, 1911, 1906). The Westbury Formation is characterized by relatively low TOC

355 (although note scale difference for TOC between Figs 3–6), and carbonate is typically in

356 diagenetic concretionary horizons. The Westbury Formation is generally interpreted as

357 deeper and further offshore compared to the Williton Member, but the uppermost Westbury

358 Formation documents a shallowing (Hesselbo et al., 2004). The Cotham Member of the

359 Lilstock Formation overlies the Westbury Formation, comprises mudstones and

360 limestones, and is interpreted as a shoreface equivalent to the Westbury Formation

361 (Hesselbo et al., 2004). The Cotham Member contains an erosional surface, desiccation

362 cracks, and ooids, representing a substantial shallowing relative to the rest of the section,

363 subaerial exposure, and potentially non-marine conditions (Hesselbo et al., 2004 and

364 references therein). The negative CIE occurs in the Cotham Member, and thus this interval

365 likely represents the ETE. Although not found at St. Audrie's Bay, sections nearby contain

16

366 stromatolites in association with the negative excursion (Ibarra et al., 2016), and their

367 widespread occurrence suggests the absence of fauna and further supports the interpretation

368 of this interval as representing the ETE. The Langport Member of the Lilstock Formation

369 comprises limestone and mudstone and is interpreted as fully marine but relatively shallow

370 based on wave-generated ripples (Hallam, 1960; Richardson, 1911; Swift, 1995). Above,

371 the Blue Lias Formation comprises organic-rich shale and limestones that are likely a

372 product of diagenetic mobilization of carbonate (Hallam, 1964). Burrowing intensity varies

373 substantially in the Blue Lias Formation, and carbonate-rich beds typically reflect less

374 organic-rich, oxygenated settings while the more organic rich facies are interpreted to

375 reflect an anoxic sediment water interface (Hesselbo et al., 2004). Fossils and pyrite are

376 common and suggest marine but potentially euxinic conditions in the water column or

377 sediment water interface (Hesselbo et al., 2004; Wignall, 2001).

378   In summary, the shallowest portion of the St. Audrie's Bay section is during the

379 ETE, while the remainder of the exposed Rhaetian and Hettangian are relatively deeper but

380 with very different TOC and carbonate contents, likely due to differences in conditions at

381 the sediment water interface and during early diagenesis.

382 **3.3 Lombardy Basin, Southern Alps, Italy**

383   The Lombardy Basin (Southern Alps, Italy) is a rift-related basin developed during

384 the Mesozoic. The Upper Triassic to Lower Jurassic succession (upper Norian to

385 Sinemurian) comprises a mixed marly-carbonate to carbonate succession deposited on the

386 western margin of Tethys (e.g., Jadoul et al., 2012; Zaffani et al., 2018). The lower portion

387 of the Calcari di Zu Formation (i.e., Zu Limestones) is upper Norian–Rhaetian and contains

388 common bivalves and foraminifera. Cycles of dark marls transition upwards into limestone

389   (Zu-1 Member; Fig. 5; Fig S1). These beds successively develop into centimeter-scale

390   carbonate beds interlayered with very thin, shale-rich intercalations (Zu-2 Member; Fig. 5;

391   Fig S1) (Galli et al., 2007; Zaffani et al., 2018). These cyclic packages comprise

392   bioturbated and fossiliferous wackestones and packstones, with patch reefs, sponges, and

393   foraminifers overlain by cyclic packages consisting of thick marl and micritic fossil-rich

394   limestones (Zu-3 Member; Fig. 5; Fig S1) containing benthic foraminifers, bivalves,

395   calcareous sponges, and corals (Galli et al., 2007; Zaffani et al., 2018). The Malanotte

396   Formation unconformably overlies the Calcari di Zu Formation and represents the

397   outermost and deepest portion of the carbonate ramp environment. It consists of thin-

398   bedded micritic limestones and mudstones, with rare bioclasts at the base and abundant

399   reworked ooids in the upper part as it grades into the Albenza Formation (Galli et al., 2007).

400   The Albenza Formation represents a regional progradation of the Hettangian carbonate

401   platform, characterized by amalgamated beds of oolitic and bio-intraclastic grainstones,

402   which are locally dolomitized (Galli et al., 2007). This carbonate platform slowly drowned

403   during the Hettangian into an open subtidal shelf environment, represented by the Sedrina

404   Formation, which comprises mudstones–wackestones and thin marl intercalations, with

405   common black chert nodules, sponge spicules, brachiopods, pelagic bivalves, and rare

406   ammonites (Bersezio et al., 1997). The Sedrina Formation grades into the Sinemurian–

407   Pliensbachian Calcare di Moltrasio Formation (i.e., Moltrasio Limestones; Fig. 5; Fig S1),

408   which is dominated by gray marl limestones and black cherts with slumps and calcarenitic

409   beds, representing a fully pelagic environment (Bersezio et al., 1997).

410   We sampled the Lombardy Basin succession at four sections that would allow us

411   to sample the mid-Rhaetian (Zu Limestone) through the mid-Hettangian (Moltrasio

18

412   Formation) and into the Sinemurian; no single section covers the entire Rhaetian–

413   Hettangian. We sampled the Zu Limestone at the Brumano Section, which exposes ~200

414   m of stratigraphy that extends from the upper Norian (Zu-1 Member) to the mid-Rhaetian

415   (middle of Zu-3) (Zaffani et al., 2018). We sampled ~120 m of this section from Zu-2 (just

416   above the Norian–Rhaetian boundary) to the top of the Zu-3a sub-member at the top of the

417   section (Muttoni et al., 2010). We sampled the upper Zu Formation (sub-members 3b and

418   3c) up to the base of the Malanotte Formation at the Italcementi Quarry. The Triassic–

419   Jurassic boundary is placed within the Zu-3c sub-member (Zaffani et al., 2018). There are

420   multiple sections at the Italcementi Quarry, and we sampled from the inactive quarry (~100

421   m total stratigraphy), corresponding to the same sites used by Zaffani et al. (2018). Only

422   the very base of the Malanotte Formation is exposed at the Italcimenti Quarry; we sampled

423   the remainder of this formation along the Valcava-Torre di Busi road (the ~30 m Malanotte

424   section from Galli et al., 2005). Finally, we sampled the upper portion of the Malanotte

425   Formation extending through the Albenza and Sedrina formations into the Moltrasio

426   Formation at Val Adrara (corresponding to the well-studied section described and studied

427   by Bachan et al., 2012; Galli et al., 2007; Van de Schootbrugge et al., 2008; Jost et al. 2017

428   and others). We sampled the lower ~350 m of the Val Adrara section. To generate a

429   composite stratigraphic section for the Lombardy Basin samples used in this study, we

430   correlated between sections based on formation boundaries, biostratigraphy, and

431   chemostratigraphy ($\delta^{13}C$); a detailed correlation between these sections is available in the

432   supplementary information.

433   **3.4 Panormide platform, Sicily, Italy**

434   The Triassic and Lower Jurassic shallow water carbonates from Sicily are

435   dolostones and limestones that crop out in the Sicilian Maghrebian chain and in the

436   subsurface of the foreland areas of the Hyblean plateau and Sicily Channel (Antonelli et

437   al., 1988; Patacca et al., 1979). These rocks were deposited in a wide carbonate embayment

438   that was flanked by deep water basins in the southwestern branch of Tethys (Di Stefano et

439   al., 2015; Zarcone et al., 2010). During the Late Triassic, the carbonate platform edge was

440   a Dachstein-type reef complex dominated by hypercalcified coralline sponges that are now

441   well exposed in the Palermo and Madonie Mountains and in central-southern Sicily (Di

442   Stefano et al., 2015). The reef complex transitioned to an escarpment and to a deep water

443   basin, while the back reef zone was characterized by extensive lagoons. Westward, a

444   transition to evaporite facies (sabkha-type) characterized the innermost zone of the

445   carbonate platform (Lo Cicero, 1986). The shallow water carbonate sedimentation lasted

446   until the late Sinemurian–Pliensbachian, when the platform experienced a drowning.

447   In Sicily, the Triassic–Jurassic transition in peritidal settings has been documented

448   in a few areas from the Hyblean foreland and in the Sciacca area (Cacciatore et al., 2006;

449   Patacca et al., 1979). More recently, a well exposed and continuous peritidal succession

450   ~430 m thick, encompassing the Triassic–Jurassic boundary, was described in detail in

451   Northwestern Sicily (Mt Sparagio, San Vito Lo Capo Peninsula; Todaro et al., 2018, 2017).

452   In this section, the peritidal limestones are organized in shallowing upward cycles (1.5 m

453   thick on average) characterized by subtidal, intertidal and supratidal facies (D'Argenio,

454   1974; Strasser, 1991). Based on the fossil content in the subtidal facies, the Mt Sparagio

455   section is divided into three informal units: unit A records the common occurrence of very

456   large megalodontid shells (up to 30 cm in size), coral patch reefs, and benthic foraminifers

457 such as *Triasina hantkeni* Majzon, *Auloconus permodiscoides* (Oberhauser), *Aulotortus*

458 *sinuosus* (Weynschenk), *Aulotuortus* sp., *Glomospirella* sp., *Trochammina* sp.,

459 *Frondicularia* sp., *Nodosaria* sp. (Todaro et al., 2017). Some horizons contain calcareous

460 algae, classified as *Cayeuxia* sp. and *Orthonella* sp. The presence of *Triasina hantkeni*

461 indicate this unit is Rhaetian in age. Unit B shows the same benthic foraminifera

462 assemblage as unit A, with differences in the macro benthic community, as corals disappear

463 and a change in the size of megalodontids is observed (Todaro et al., 2017, He et al., 2020).

464 The upper boundary of unit B is identified based on the last occurrence of the Rhaetian

465 benthic community and by the presence of an oolitic horizon. Above, there is a ~10 m thick

466 fossil-poor interval that contains only small calcispherae. Unit C consists of oligotypic

467 facies characterized by a bloom of the alga *Thaumatoporella parvovesiculifera* in

468 association with *Aeolisaccus* sp, and, higher up, by *Siphovalvulina* sp. This fossil

469 assemblage is considered typical of the Hettangian–Sinemurian interval (Barattolo and

470 Romano, 2005, Boudagher-Fadel and Bosence, 2007).

471 **3.5 Levanto, Peru**

472   The Levanto section in northern Peru spans the Upper Triassic and Lower Jurassic;

473 the ~105 m section is continuous for ca. four million years. The entirety of the outcrop

474 consists of the Aramachay Formation, which, at this location, comprises carbonate-rich

475 mudstones with abundant intercalated ash beds (Fig. 6). U–Pb dating of zircons of the ash

476 beds and ammonite biostratigraphy (Guex et al., 2012; Schaltegger et al., 2008; Schoene

477 et al., 2010; Wotzlaw et al., 2014) have constrained the durations of the Rhaetian and

478 Hettangian, and the chronology provides a valuable biostratigrahic and temporal

479 framework for geochemical work at the section. The section is well below storm wave base

480 and is characterized by little bioturbation, which we interpret as indicative of anoxic

481 conditions at the sediment water interface during deposition, persisting from the Rhaetian

482 through Hettangian. Previous work on carbon isotopes, TOC, and carbonate content (Yager

483 et al., 2017) record a typical Triassic–Jurassic boundary carbon isotope profile. Unlike

484 many Tethyan sections, Levanto is characterized by lower TOC during the Hettangian

485 relative to the Rhaetian. Carbonate content remains relatively high (~60%) through much

486 of the section but is variable, particularly at the top of the section, where carbonate is

487 concentrated in thick beds.

488 The first ~57 meters of the continuous section is characterized by alternating thin

489 and thick-bedded intervals, though this alternation has little influence on sedimentary

490 structures or lithology as observed in thin sections (Yager et al., 2017). At approximately

491 57 m, this alternation changes to meter-scale differences, and eventually (~80 m) grades

492 into concretionary bedding and finally into meter-scale concretions (~85). Thin sections

493 contain carbonate-replaced radiolarians, rare foraminifera, sponge spicules, and low

494 contents of coarse detrital material (<5%). Pyrite is absent in the section, in stark contrast

495 to St. Audrie's Bay. In summary, the Levanto section records a relatively static depositional

496 environment spanning ca. four million years and has robust geochronological and

497 biostratigraphic constraints.

498

499 **4. MATERIALS AND METHODS**

500 **4.1 Sample processing**

501 Samples were collected from outcrop and then cut to remove weathered material,

502 veins, and modern marine-derived debris (specifically from St. Audrie's Bay samples).

503   Portions of samples were crushed in a jaw crusher and pulverized either in a stainless steel

504   disk crusher (New York Canyon samples) at the University of Southern California, Los

505   Angeles, CA USA (USC), in a tungsten carbide ball mill at USC (St. Audrie's Bay

506   samples), or at Actlabs in Ontario, Canada (Levanto and Lombardy Basin samples). Long

507   term blank testing indicated none of the powdering processes resulted in Hg contamination.

508   **4.2 Hg contents**

509   For the St. Audrie's Bay, New York Canyon, and Levanto samples, total Hg was

510   measured using a Hydra IIc at The University of Toronto (Toronto, Canada), using similar

511   methods to those from Thibodeau et al. (2016). In the Hydra IIc, weighed sub-samples of

512   powdered rocks were combusted under O_2 flow at 350 ml/min, heated in a drying step to

513   300ºC for 40 s, then decomposed for 300 s at 800ºC. Following combustion, evolved gases

514   were carried through a heated (600ºC) catalyst tube to remove possible interferences (e.g.,

515   halogen compounds, sulfur oxides, nitrous oxides) and Hg was captured on a gold trap

516   while combustion gases were removed from the detection cell. The gold trap was heated

517   for 30 s at 600ºC to release Hg, which was then carried to the detection cell where

518   absorbance from a mercury lamp is measured at 253.7 nm. For this study, instrumental

519   response was calibrated using the NIST 3133 Hg standard in a 0.25% L-cysteine solution.

520   Blank absorbance was <2% of typical sample signals and always <4%. NIST 3133 was

521   periodically combusted and analyzed alongside samples to determine precision, with

522   results within 5% of the nominal values. Samples measured more than once are reported

523   as averages, which typically have reproducibility better than 10%. External standards NIST

524   SRM 1944 and 1646a were combusted alongside samples to check measurement accuracy.

525 Errors on Hg content measurements are estimated to be 10% (2sd) based on reproducibility

526 of samples and external standards.

527 Lombardy Basin and preliminary Panormide Domain samples were analyzed for

528 total Hg concentrations using an MA-3000 Direct Mercury Analyzer (Nippon Instruments

529 Corporation, Tokyo, Japan) thermal decomposition analyzer at USC. Similar to the Hydra

530 IIc, the MA-3000 combusts powdered samples, uses a catalyst to remove interferences,

531 traps Hg on a gold trap, and measures the amount of Hg after release from the trap by

532 absorbance at 253.7 nm. The MA-3000 allows for combusting multiple aliquots of a single

533 sample in order to accumulate enough Hg for analysis after release from the gold trap. This

534 approach is well suited to samples with low Hg contents, such as the rocks from the

535 Lombardy Basin. Total sample masses of up to 5 g were analyzed by combusting up to 20

536 ~250 mg sub-samples. Even with this approach, several samples contained too little Hg to

537 obtain robust results, because the detection limit of 1.5 ng could not be reached with a

538 reasonable number of combustions for some samples. Samples from the Lombardy basin

539 that were below the detection limit are noted in the supplementary dataset. No samples

540 from the Panormide domain in Sicily were above the detection limit, and results from this

541 section are not reported here. We consider these to represent the analytical limit of Hg

542 content measurement via the widely-used thermal decomposition method, though other

543 analytical methods might allow Hg quantification in such rocks.

544 **4.3 Hg isotope measurements**

545 Mercury isotope measurements followed the analytical methods from Thibodeau et

546 al. (2016). Briefly, Hg was extracted and purified from samples using the Hydra IIc without

547 the gold trap (with a longer decomposition time than that used for Hg concentration

548 measurements). Mercury was trapped by directly sparging the gas outflow with elemental

549 Hg into ~10% H_2SO_4 (v/v) and ~1% $KMnO_4$ (w/w), which oxidized the Hg0 gas to HgII.

550 To ensure removal of residual Hg in the furnace, after each sample combustion, 50 µL of

551 Milli-Q was combusted and lines were heated with a heat gun to transfer any residual Hg

552 to the trap. NIST 3133 and 1646a were combusted and trapped as procedural standards and

553 blanks.

554 Procedural blanks were <0.02 ng/g, which is <1–2% of the sample Hg. Mercury

555 recovery was checked by neutralizing an aliquot of each solution with NH_2OH-HCl

556 immediately after vapor trapping and measuring its concentration using a Tekran 2600 cold

557 vapor atomic fluorescence spectrometer (Tekran instruments, Toronto, Canada). The ~10%

558 variation in sample recoveries reflects the uncertainty in concentration analyses and sample

559 heterogeneity. Mercury isotope analyses were conducted with a cold vapor multi–collector

560 inductively coupled mass spectrometer (Neptune Plus MC–ICP–MS, Thermo-Finnigan,

561 San Jose, CA, USA) at the University of Toronto. Sample solutions were neutralized with

562 NH_2OH-HCl and diluted to 1–2 ng/g with a matrix matched solution. Mercury was

563 introduced to the plasma as Hg0 after $SnCl_2$ reduction and Hg0 vapor separation on a gas–

564 liquid separator. A thallium internal standard and standard-sample bracketing with NIST

565 3133 were used to correct for instrumental mass bias, and an in-house Hg standard (J.T.

566 Baker Chemicals) was measured multiple times in each analytical session to determine

567 external reproducibility of the method. Signal concentrations and intensities of all

568 bracketing standards and samples matched within 10%. Isobaric interference from ^{204}Pb

569 was monitored using ^{206}Pb, but was always negligible (correction never altered the

570 calculated $\delta^{204}Hg$). On-peak blank corrections were made on all Hg and Pb masses.

571   All samples and procedural standards were measured at least twice, and sample

572   isotope values are reported as the mean of duplicate or triplicate measurements (Tables S2

573   and S3). Average values for procedural standards are reported and are consistent with

574   previous values for these standards (Thibodeau et al., 2016). Sample errors are reported as

575   either the 2σ (standard error mean) of sample replicates or the 2σ (standard deviation)

576   external reproducibility of the in-house J.T. Baker Hg standard, whichever is larger.

577   **4.4 TOC and TIC**

578   **TOC:** For samples from Levanto (Yager et al., 2017), St. Audrie's Bay, and New York

579   Canyon, sub-samples of powders (~1 g) were treated with 1M HCl at 70°C for four hours

580   to remove carbonate from the samples. Samples from the Italian sections (Brumano,

581   Valcava Torre di Busi, and Italcementi) were treated the same way, but with 1 hr

582   decarbonation times; our tests of decarbonation times indicate 1 hr heat treatments yield

583   the same results for $\delta^{13}C$ and TOC as up to 8 hr decarbonation times (unpublished data).

584   The samples then settled for 24 hours, the supernatant was decanted, and the decarbonated

585   powder was rinsed three times with repetitive settling and decanting before being dried

586   overnight at 50°C (after Galy et al., 2007). For dolomite-containing Val Adrara samples,

587   this decarbonation procedure failed to fully remove carbonate, a problem identified by

588   many unrealistically high $\delta^{13}C$ values for organic carbon (e.g., greater than –5‰).

589   Additional sub-samples were treated with 3M HCl, agitated before heating (to expose all

590   sample to acid), and then heated at 70°C for 1 hour to ensure complete carbonate removal.

591   Acidified samples from Val Adrara were then centrifuged (2,000 RPM in an 8 inch

592   diameter centrifuge) for 2 minutes in lieu of letting samples settle. Reported results for

593   Italian samples are from this decarbonation procedure. We tested samples from New York

26

594 Canyon using both methods, and results were within the analytical uncertainty for both

595 TOC and $\delta^{13}C_{org}$ measurements, supporting either method as a viable means of TOC and

596 $\delta^{13}C_{org}$ measurement.

597   For all samples, TOC (wt.%) was determined on decarbonated powder using

598 Picarro cavity ring down spectroscopy (CRDS; G2131-I; Santa Clara, CA, USA) coupled

599 to a Picarro Liaison (A0301) and a Costech Elemental Combustion System (EA 4010,

600 Valencia, CA, USA). Picarro measurements of CO_2 concentrations were calibrated to

601 determine the percent of organic carbon in samples using the USGS 40 standard (L-

602 glutamic acid) weighed at five different sample masses and run at the beginning and end

603 of each set of ~15 samples. Determination of the reported sample TOC took into account

604 the amount of carbonate lost during decarbonation (see further detail in supplemental

605 information and Fig. S2). Uncertainty was determined by replicate analysis of samples and

606 standards (typically ~2 replicates per sample; see Supplementary Table 1). Standard

607 deviation of replicates for measured %TOC was ± 0.08 on average, or approximately $\pm 3\%$

608 of the measured value (1σ standard deviation). Because of potential errors introduced by

609 small amounts of sample loss during liquid decarbonation, including via solubilization of

610 organics (Galy et al., 2007), our conservative estimate of uncertainty is $\pm 10\%$ of the

611 measured value for reported TOC. We elaborate further on our methods for TOC

612 measurement in the supplement. Importantly, our methods do not have the same 0.2% TOC

613 detection limit reported in many previous studies (typically measured via Rock-eval) and

614 reviewed by Grasby et al. (2019), since we are able to calibrate the amount of organic

615 carbon in our samples well within a standard curve (see supplemental information).

616 **TIC:** Depending on the sample's carbonate content, 3–200 mg of sample was weighed into

617 10 mL glass Exetainer vials with rubber septa caps. Vials were evacuated and acidified

618 with 1 mL 30% H_3PO_4. Samples and standards were heated for 80 minutes in a water bath

619 at 70°C to ensure that C associated with all carbonate phases was released as CO_2. Samples

620 were then run on a Picarro CRDS coupled to an Automate preparation device, which

621 sparges the solution with N_2 gas to drive CO_2 into the analyzer. In-house carbonate

622 standards Optical calcite (OPT) and AR15 were run at different masses to calibrate total

623 inorganic carbon (TIC). Errors were calculated by replicate analyses of samples and

624 standards. Average error (1σ) for TIC measurements was ±0.04, or <1% of the measured

625 value. Our reported carbonate contents (wt.%) were calculated from measured TIC,

626 assuming all inorganic carbon in our samples is $CaCO_3$. No dolomite or other carbonate

627 phases were observed in thin sections of the NYC, SAB, and Levanto samples. Dolomite

628 is present in the Lombardy Basin samples, potentially introducing some uncertainty into

629 carbonate content values for this section. These independent TIC analyses also allow for

630 some constraint on the mass loss during decarbonation, which assist in understanding

631 potential error introduced to TOC measurements (see supplemental information).

632

633 **5. RESULTS**

634   We report Hg contents, TOC contents, carbonate contents, and Hg isotope

635 compositions in the supplemental data set. We analyzed 193 samples for Hg contents and

636 18 samples for Hg isotopes from the Levanto section, 80 samples for Hg contents and 13

637 for Hg isotopes from St. Audrie's Bay, and 81 samples for Hg contents from New York

638 Canyon, complementing the previously published 35 content and 35 isotope measurements

639 from Thibodeau et al. (2016). The New York Canyon samples in this study are all

640 stratigraphically above those previously published in Thibodeau et al. (2016). From the

641 Lombardy Basin, we analyzed Hg contents in 24 samples from the Brumano section (11

642 yielding Hg above the detection limit), 57 from the Italcementi section (25 above the

643 detection limit), 21 from Valcava Torre de Busi (18 above the detection limit), and 29 from

644 Val Adrara (19 above the detection limit).

645 In Figs 3–6 we plot total Hg, TOC contents, Hg/TOC, δ^{202}Hg, and Δ^{199}Hg for each

646 section with stratigraphy. In each figure, 'pre' denotes the pre-extinction interval (i.e., the

647 available Rhaetian stratigraphy until the onset of the negative C isotope excursion), ETE

648 is denoted (i.e., from the onset of the negative CIE to the TJB), and 'post' denotes post-

649 extinction (i.e., Hettangian and above, where available). Figure S3 contains selected cross

650 plots of carbonate content, Hg, Hg/TOC, and TOC, with data separated by relative time

651 intervals (pre-ETE, ETE, and post-ETE).

652 **5.1 New York Canyon**

653 In Fig. 3 and Fig. S3a–c we report data from Thibodeau et al. (2017) and additional

654 data from this study. Prior to the ETE, samples have average Hg contents of ~12 ± 4 ppb

655 (n = 4; 1 s.d.). This value rises to ~50 ± 21 ppb (n = 14, 1 s.d.) during the ETE, and then

656 decreases to 11 ± 10 ppb (n = 100, 1 s.d.) during the Early Jurassic. Ratios of Hg/TOC have

657 average values of 48 ± 15 ppb/wt.% (n = 4; 1 s.d.) during the pre-extinction interval, rise

658 to an average of 244 ± 139 ppb/wt.% (n = 14; 1 s.d.) during the ETE, and decrease to an

659 average value of 92 ± 48 ppb/wt.% (n = 100; 1 s.d.) during the post-extinction interval.

660 Mercury isotope were originally reported and discussed in Thibodeau et al. (2017). The

661 δ^{202}Hg values during the Rhaetian and early Hettangian are between ~0‰ and –1.5‰,

662 becoming progressively more negative upsection; Δ^{199}Hg values are near zero during the

663 ETE and in the early Hettangian, but deviate from zero (in both negative and positive

664 directions) during the Hettangian. The New York Canyon section is truncated in the

665 Rhaetian, so it was not possible to measure additional samples from before the extinction

666 from this studied section. Mercury and TOC contents are not correlated during the ETE but

667 are correlated ($r^2 = 0.50$) in the Hettangian samples (Fig. S3b).

668 **5.2 St. Audrie's Bay**

669 Figures 4 and S3d–f report data from St. Audrie's Bay. During the Rhaetian,

670 average Hg contents are ~29 ± 15 ppb (n = 38, 1 s.d.), during the ETE (correlated using the

671 negative CIE in organic C isotopes) average Hg contents are 19 ± 15 ppb (n = 13; 1 s.d.),

672 and during the Early Jurassic Hg contents are 42 ± 29 ppb (n = 29; 1 s.d.). Ratios of

673 Hg/TOC during the pre-extinction interval are on average 43 ± 27 ppb/wt.%TOC (n = 38;

674 1 s.d.), during the ETE are 102 ± 77 ppb/wt.% (n = 13; 1 s.d.), and during the Early Jurassic

675 are 18 ± 20 ppb/wt.%TOC (n = 29; 1 s.d.). Ratios of Hg/TOC rise to ~150 ppb/wt.%

676 coincident with the onset of the negative C isotope excursion (meter ~12) and then decrease

677 to an average of ~20 ppb/wt.% during the Hettangian. Mercury and TOC are correlated,

678 particularly during the ETE ($r^2 = 0.51$) and the Hettangian ($r^2 = 0.90$; Fig S3e). Percival et

679 al. (2017) also reported Hg data from St. Audrie's Bay, which are plotted in Fig. 4; their

680 TOC data are from Hesselbo et al. (2002). Mercury contents from Percival et al (2017)

681 match well with our data, and TOC data are similar in the two studies, but the interval

682 during the negative carbon isotope excursion differs slightly, resulting in a difference in

683 Hg/TOC ratios. This discrepancy could be due to heterogeneity in the section during

30

684 sampling or TOC methodological differences; the implications for this difference are

685 discussed in the figure caption.

686   Values of δ^{202}Hg at St. Audrie's Bay are similar throughout the measured section

687 and range between –1.50‰ and –0.50‰; Δ^{199}Hg values are negative during the Rhaetian

688 (–0.20‰ to –0.40‰), increase to about –0.10‰ during the ETE, and decrease again during

689 the Hettangian (–0.20‰ to –0.40‰).

690 **5.3 Lombardy Basin**

691   The Lombardy Basin results are separated into the four respective sections (Fig.

692 S1) and are reported in a composite section in Fig. 5. Considered together, the Lombardy

693 sections exhibit low Hg contents (all <15 ppb, and contents greater than ~6 ppb found only

694 in the Brumano section) and low TOC. Many samples contain so little Hg that the 1 ng

695 detection limit would have required an estimated 10–20 separate combustions, implying a

696 content well below 1 ppb (as noted above, we did not analyze samples with such little Hg).

697 Marly samples contain slightly more Hg and sometimes elevated TOC.

698   At the Brumano section, samples have an average Hg content of 7 ± 4 ppm (n = 11,

699 1s.d.), TOC average of $0.2 \pm 0.1\%$ (n = 26; 1 s.d.), and Hg/TOC average of 25 ± 7 wt.% (n

700 = 10; 1 s.d.). The Brumano section, which is the deepest of the composite Italian sections,

701 deepens upwards, with less carbonate in samples from further up-section. Many carbonate-

702 rich samples lower in the section were below the detection limit for Hg measurements.

703 Overall, the Brumano section Hg contents are low (below the detection limit to ~3 ppb) in

704 resistant, carbonate-rich beds and higher (between ~5 and 14 ppb) in marly, mudstone beds.

705   Samples from the Italcementi quarry section (Fig. S1) have average Hg contents of

706 4 ± 3 ppb (n = 25, 1 s.d.; not including 32 samples below detection limit), TOC contents

31

707 of 0.1 ± 0.04 wt.% (n = 22, 1 s.d.) and Hg/TOC ratios of 44 ± 26 (n = 22; 1 s.d.). The two

708 high (>50) Hg samples ITZ62 and ITZ64 are from marly beds. Carbonate-rich beds exhibit

709 somewhat lower Hg contents.

710 The Valcava Torre dei Busi section (Fig. S1) has average Hg contents of ~2 ± 1.5

711 ppb (n = 18; 1 s.d.), TOC contents of 0.08 ± 0.16 wt.% (n = 27; 1 s.d.) and Hg/TOC values

712 of 44 ± 25 ppb/wt.% (n = 16; 1 s.d.).

713 The Val Adrara section (Fig. S1) contains average Hg contents of 2.7 ± 1.4 ppb (n

714 = 19; 1 s.d.), TOC concentrations of 0.03 ± 0.03 wt.% (n = 15; 1 s.d.) and Hg/TOC of 140

715 ± 119 ppb/wt.% (n = 15; 1 s.d.). Although the highest Hg/TOC values are found above the

716 Triassic–Jurassic boundary from the Hettangian portion of the Val Adrara section, a less

717 pronounced rise in Hg/TOC is synchronous with the TJ boundary.

718 **5.4 Levanto**

719 In Figs 6 and S3g–i we report data from the Levanto section. Mercury contents are

720 on average 33 ± 14 ppb during the Rhaetian pre-extinction interval, (n = 84; 1 s.d.), 37 ±

721 24 ppb (n = 14; 1 s.d.) during the negative CIE and ETE Hg concentrations, and 36 ± 19

722 ppb following the ETE during the Hettangian (n = 96; 1 s.d.). Hg/TOC during the pre-

723 extinction interval is 24 ± 25 ppb/wt.% (n = 84; 1 s.d.), during the ETE is 30 ± 29 ppb/wt.%

724 (n = 14; 1 s.d.), and following the ETE is 64 ± 36 ppb/wt.% (n = 96; 1 s.d.). There is an

725 increase in Hg/TOC at ~57 meters, coincident with the negative CIE in organic carbon

726 isotopes (Yager et al., 2017). Mercury and Hg/TOC are negatively correlated with

727 carbonate (Fig S3h–i) during the ETE and Hettangian. Correlations between Hg and TOC

728 are weaker than for the St. Audrie's Bay and New York Canyon sections (Fig. S3g–i);

729 elevated Hg/TOC is expressed during the ETE and into the Hettangian. Values of δ^{202}Hg

are between about $-1.30‰$ and $-0.30‰$ for the measured section, with a possible excursion from $-0.3‰$ to $-1.5‰$ during the late Rhaetian and ETE; Δ^{199}Hg is slightly positive during the Rhaetian ($\sim0.1‰$) and near zero without discernable MIF during the ETE and Hettangian ($\pm0.05‰$).

In the Rhaetian portion of the Levanto section, there is no discernible lithologic control on Hg contents based on outcrop or thin section observations (i.e., neither high nor low Hg contents appear to be related to specific lithologic composition). In contrast, during the Hettangian, alternations in thick and thin bedded strata appear to impact Hg contents and Hg/TOC. Samples high in Hg are compacted and contain less carbonate and slightly higher TOC, while samples that are uncompacted and high in CARB contain less Hg and lower Hg/TOC (Fig. S4). At ~57 m, carbonate content variability increases and samples with lower carbonate contents are associated with higher Hg concentrations (Fig. S5). We note proximity of samples to ash beds did not have any discernible effect on Hg contents, and in fact the Rhaetian portion of the section includes more conspicuous ash beds than the Hettangian, indicating ash bed presence does not control Hg contents in these samples.

5.5 Consistent observations between sections

Using organic C isotopes as a correlation tool, at all four sections Hg/TOC rises during the negative CIE and, by inference, during the contemporaneous ETE. At New York Canyon and St. Audrie's Bay, Hg/TOC decreases during the Early Jurassic after its peak associated with the CIE and ETE (Fig. 7). In contrast, at Levanto, Hg/TOC increases during the negative CIE and ETE and then increases further in the early Jurassic, remaining elevated for the remainder of the section (Fig. 7). In the Lombardy Basin, Hg/TOC is briefly elevated during the pre-ETE interval, decreases and then increases again during the

753 ETE and negative isotope excursion, and then rises to the highest observed values during

754 the early Hettangian (although these very high values from the Val Adrara section are

755 associated with very low TOC). Thus, St. Audrie's Bay and New York Canyon both exhibit

756 relatively well-defined Hg anomalies, while those at Levanto and in the Lombardy Basin

757 appear more protracted and less clearly defined. Levanto and Lombardy also share

758 somewhat enigmatic brief spikes in Hg/TOC in the late Rhaetian, prior to the ETE and C

759 isotope excursion.

760   At St. Audrie's Bay, New York Canyon, and Levanto, each section also exhibits

761 negligible or low MIF (near zero Δ^{199}Hg) during the ETE. New York Canyon and Levanto

762 record a slightly negative shift in δ^{202}Hg during the Late Rhaetian and ETE, while low

763 sampling resolution in Hg isotopes at St. Audrie's Bay may obscure any similar trend at

764 this site. We were not able to measure Hg isotopes from the Lombardy sections.

765

766 **6. DISCUSSION**

767   All four sections contain elevated Hg/TOC during the ETE and negative CIE, when

768 compared to pre-extinction values. During the same interval, MIF of Hg isotopes (Δ^{199}Hg)

769 is near zero or shifts towards zero, suggesting an increase in Hg deposition derived from

770 subaerial volcanism associated with CAMP activity during the ETE. Below, we detail the

771 implications of elevated Hg/TOC and low MIF and discuss the caveats from our study and

772 next steps for using and understanding the Hg proxy.

773

774 **6.1 Mercury isotopes support a magmatically derived Hg signal at the ETE**

775   At New York Canyon, Levanto, and St. Audrie's Bay, elevated Hg/TOC values

776   during the ETE coincide with Hg isotope signals that exhibit negligible MIF (Fig. 7),

777   consistent with a large increase in volcanic Hg that overwhelmed the surface reservoirs in

778   a way that resulted in minimal MIF producing photoreduction prior to deposition

779   (Thibodeau and Bergquist, 2017). Outside of the extinction interval, Hg isotopes are

780   consistent with normal Hg cycling through surface reservoirs, acquiring positive and

781   negative MIF (e.g., Bergquist and Blum, 2007; Blum et al., 2014; Thibodeau and

782   Bergquist, 2017 and references therein).

783   In more detail, deposition of Hg in many terrestrial systems is dominated by

784   gaseous elemental Hg via uptake by vegetation and subsequent litterfall that results in

785   terrestrial biomass and soils acquiring negative MIF and MDF. Thus, marine sediments

786   close to continents and dominated by terrestrial input often reflect this source of Hg. In

787   contrast, marine sediments farther from land that are dominated by atmospheric depositon

788   of HgII species have positive MIF and less negative MDF. Our results for MIF and MDF

789   outside of the ETE are consistent with varying mixtures of Hg sourced from the atmosphere

790   (positive MIF, less negative MDF) and Hg sourced from terrestrial runoff (negative MIF,

791   more negative MDF). The Hg MIF signatures are also consistent with the expectation for

792   the depositional environment of each section — with slightly positive MIF at the deeper

793   (Levanto) section, likely more influenced by atmospherically derived Hg, and more

794   negative MIF at the shallower sections (St. Audrie's Bay, New York Canyon), more

795   influenced by terrestrial sources (Figs. 3–4 and 6). The St. Audrie's Bay section, which is

796   shallow enough to have been exposed subaerially during the ETE, shows the most negative

797   MIF of any section studied here. In summary, when Hg/TOC ratios are relatively low

798   (typically before and after the ETE), each section bears a MIF composition consistent with

799   its major source of Hg input, with near-shore sections displaying terrestrial negative MIF

800   (which may also be related to fires and biomass burning as suggested by Grasby et al.,

801   2019; e.g., at St. Audrie's Bay) and more offshore sections having positive MIF values

802   reflected of atmospheric deposition of HgII species (positive MIF for offshore sections;

803   e.g., Levanto) (Figs 2 and 8).

804   In contrast, when Hg/TOC is elevated during ETE Hg/TOC anomalies, MIF values

805   approach zero, consistent with Hg input from unaltered volcanic sources (Thibodeau and

806   Bergquist, 2017). Importantly, the absence or decrease in the extent of MIF coincides with

807   the intervals of elevated Hg/TOC in each section. When the elevated Hg/TOC periods

808   cease, the two shallower sections also see a return to odd MIF values, although with

809   different timings. At St. Audrie's Bay, the Hg/TOC peak ends relatively abruptly, and

810   negative MIF returns in the first sample measured from the Hettangian. At New York

811   Canyon, there is a somewhat more gradual return to background Hg/TOC extending into

812   the earliest Hettangian, and the absence of MIF follows suit. The Levanto record is

813   different, and exhibits elevated and noisey Hg/TOC persisting throughout the Hettangian

814   and positive MIF does not reoccur after the ETE (discussed in more detail below).

815   Altogether, the disappearance of MIF is remarkably consistent across the three different

816   sections, suggesting that absence or decrease of MIF may be a general feature of TJB Hg

817   anomalies. This observation lends support to a global magmatic source for the Hg

818   anomalies at this time (consistent with the conclusions of Thibodeau et al., 2017).

819   Like the MIF signal, the Hg isotope MDF compositions largely reflect the

820   depositional setting of each section. We observe negative MDF in all sections (Figs 3–4

821   and 6), but this signature is less pronounced at Levanto, which likely had less relative

822   terrestrial influence relative to St. Audrie's Bay and New York Canyon given its deeper

823   depositional setting and positive MIF values prior to the ETE. Closer proximity to the

824   continents results in more continental influence and more influence from biomass and soil,

825   resulting in more negative MDF compositions. At New York Canyon, there is a general

826   trend towards more negative δ^{202}Hg values coincident with upward shallowing through the

827   section. The increasingly negative δ^{202}Hg values may reflect an increase in the relative

828   amount of continentally derived material versus atmospheric deposition. Unlike the MIF

829   signal, we do not see systematic shifts in δ^{202}Hg values associated with the Hg/TOC

830   anomalies in any of the three sections studied here; this is not surprising given volcanic Hg

831   also has negative MDF that overlaps with terrestrial Hg and atmospheric HgII.

832

833   **6.2 Comparison to Hg isotope records from other mass extinction and LIP intervals**

834   Mercury isotope records are also available from other sedimentary sections

835   deposited during LIP activity (summarized in Fig. 8). Across these studies, as in the case

836   of the ETE, MIF signatures broadly reflect the depositional setting (e.g., as in Fig. 2).

837   However, the end–Triassic is distinct in that MIF is negligible or shifts towards zero during

838   the Hg/TOC anomaly, at least for the localities studied thus far, while similar systematic

839   shifts are not seen during other time periods. For example, there are several records of Hg

840   isotopes for the end–Permian mass extinction, with several deep-water sections (Buchanan

841   Lake, Sverdrup Basin, Canada; Daxiakou, Shangsi, Gujo-Hachiman) showing positive

842   MIF, and a platform carbonate section (Meishan, China) exhibiting negative MIF,

843   particularly during and after the increase in Hg/TOC (Grasby et al., 2017; Shen et al.,

844   2019b; Wang et al., 2018). However, changes in the MIF signature during the end–Permian

845   Hg/TOC anomaly are inconsistent. At Gujo-Hachiman, MIF decreases much as it does in

846   the end–Triassic sections, but there is little change in MIF of the other end–Permian

847   sections (Buchanan Lake, Xiakou) and a shift from positive to negative MIF is observed at

848   Meishan.

849   The limited Hg isotope data from other time intervals also present a mixed picture.

850   During the Toarcian Ocean Anoxic Event, data from a near-shore section suggest a shift

851   from slightly positive (0.06‰ to 0.07‰) to negative MIF (less than –0.10‰) during the

852   Hg anomaly and carbon isotope excursion (Them et al., 2019). At the Ordovician–Silurian

853   boundary, no significant change in the MIF signal was found during two apparent Hg/TOC

854   anomalies in deep water (Gong et al., 2017) or from S-rich shale sections (Shen et al.,

855   2019a) in China (however, the potential of LIP emplacement at this interval is regarded as

856   circumstantial by some studies; e.g., Shen et al. 2019). Across the Cretaceous–Paleogene

857   transition, the MIF signature during Hg excursions range widely (Sial et al., 2016). In some

858   cases, the lack of consistent isotopic signatures associated with Hg/TOC anomalies might

859   indicate a non-volcanic origin of the anomaly. In other cases, such as during the end–

860   Permian, volcanically derived Hg may have been subject to environmental transformation

861   after its release, imparting characteristic MIF depending on depositional setting (as

862   suggested by Thibodeau and Bergquist, 2017). At the end–Triassic, the rapid volcanic

863   release may have overloaded any MIF-inducing environmental processes prior to

864   deposition. If this interpretation is correct, the new data presented here show that this

865   overloading generated negligible MIF signature in sections across a wide range of

866 depositional settings at the end–Triassic. Determining whether this feature is observed in
867 all end–Triassic localities will require additional analyses.

868   The apparent contrast between the end–Triassic and end–Permian suggests that Hg
869 isotopes may not provide a "smoking gun" to distinguish whether apparent anomalies are
870 associated with LIP-derived Hg (Bergquist, 2017). Yet the differences in Hg behavior
871 between time intervals may also hold general clues about the fate of volatiles released by
872 different LIPs — an important question with implications for the environmental effects and
873 ultimately for the causes of mass extinction. Why the fate of volcanically released Hg might
874 have differed during the end–Permian versus the end–Triassic remains to be understood
875 but raises intriguing questions. For example, was the difference because of more rapid
876 volatile release during CAMP? Or did the Siberian trap emplacement release Hg from coal
877 beds and organic-rich sediments with MIF during the end–Permian? To date, the number
878 of studies reporting Hg isotope data from sedimentary rock records lags far behind those
879 presenting Hg contents and Hg/TOC ratios. Though Hg isotopes are much more
880 challenging to measure, they may offer unique insights that are simply missing from
881 concentration data alone. The consistency of the MIF patterns observed in this study at the
882 end–Triassic suggests that a concerted effort to add isotopic information may be important
883 to realizing the full scope of the Hg proxy.

884

885 **6.3 Elevated volcanogenic Hg persists after CAMP magmatism in some Triassic-**
886 **Jurassic sections**

887   The apparent ubiquity of elevated Hg/TOC during the ETE (Fig. 9; Percival et al.,
888 2017) suggests that elevated Hg may be a global signal during this interval of time,

889   presumably relating to CAMP volatile emissions. However, when viewing the Hg and

890   Hg/TOC 'anomalies' section by section, a number of complexities become apparent. The

891   Levanto section, which has a high-resolution chronology from ash bed U–Pb ages, seemed

892   a promising opportunity for testing the correspondence between CAMP timing (as

893   constrained by the U–Pb age dates) and increases in Hg and Hg/TOC contents. However,

894   Hg contents do not align with CAMP U–Pb ages in this section; instead elevated Hg begins

895   ca. 150 kyrs (ca. 201.5 Ma) after the oldest dated CAMP basalts (Fig. 10; 201.63 ± 0.029

896   Ma; Davies et al., 2017). Meanwhile, the youngest U–Pb dated CAMP basalts are $201.1 \pm$

897   0.071 Ma, but at the Levanto section elevated Hg persists until the top of the section at ca.

898   200.1 Ma. Similar elevated Hg/TOC also persists into the Early Jurassic in the Lombardy

899   sections (Fig. 9). These mismatches in timing between the Hg signal and CAMP activity

900   are a clear indication that increases in Hg contents and Hg/TOC are imperfect proxies for

901   magmatism in the sedimentary record, especially when looking to this proxy for temporal

902   correlation.

903   The effect of intrusive vs. extrusive magmatism on Hg release (e.g., Percival et al.

904   2017) does not on its own explain the timing discrepancy in the onset of the Hg anomaly

905   from Levanto. The earliest dated phases of CAMP activity were intrusive (Davies et al.,

906   2017), possibly releasing less Hg than expected from extrusive volcanism. However, only

907   the first dated CAMP basalt is intrusive (red CAMP age in Fig. 10), and several ages for

908   CAMP extrusives also pre-date the increase in Hg/TOC seen at Levanto (Fig. 10), requiring

909   another explanation for the mismatch in timing at the onset of the Hg anomaly in this

910   section.

911   The long persistence of the anomaly at Levanto is also unexpected. Prolonged

912   supply of CAMP-related Hg, e.g., from the continents, could be one explanation. However,

913   if Levanto were receiving Hg from terrestrial runoff, or from atmospheric deposition, we

914   would expect to see a characteristic Hg isotope signal; instead, MIF remains near zero

915   throughout the higher and noisier high Hg/TOC interval. Another explanation would be that

916   CAMP activity lasted longer than suggested by currently available U/Pb dates; however,

917   similar non-MIF, high Hg/TOC samples do not persist into the Hettangian at the St.

918   Audrie's Bay or New York Canyon sections, so it seems unlikely that the long-lasting

919   signal at Levanto simply reflects more CAMP-derived atmospheric Hg deposition

920   extending into the Hettangian.

921   A more plausible explanation for the extended period of high Hg/TOC at Levanto

922   and the Lombardy basin is that depositional and/or diagenetic processes in the local

923   environments controlled Hg and TOC concentrations. These processes may also explain

924   the delayed onset of the Hg anomaly at Levanto. In the section that follows, we consider

925   these effects more deeply.

926

927   **6.4 Depositional and diagenetic controls on Hg contents and Hg/TOC ratios in**

928   **sedimentary rocks**

929   Recent studies have recognized and investigated the importance of the Hg host

930   phase when applying Hg contents and Hg/TOC ratios as a proxy for LIP magmatism (Shen

931   et al., 2019a, b, 2020; Keller et al., 2020; Kovács et al. 2020), as well as the role of

932   weathering in altering depositional Hg contents (Charbonnier et al., 2020). We removed

933   surficial, altered material from our samples and did not observe the kind of systematic

934 effects anticipated from weathering (Charbonnier et al., 2020), so we do not see weathering

935 as a likely control on the changes in Hg and Hg/TOC observed through the sections we

936 have studied. However, differences in depositional environment likely played an important

937 role. Since the depositional environment and diagenetic history control the presence of the

938 host phases of Hg, they ultimately control the Hg contents. We can therefore expect that

939 Hg concentrations of most if not all sedimentary sections will be primarily controlled by

940 depositional environment and in turn host phases of Hg, and secondarily controlled by

941 secular changes in the global surficial Hg pool.

942 *6.4.1 Plausible host phases for Hg in relation to depositional environment of each section*

943 Each section in this study exhibits a different depositional environment, and in

944 some cases these changed through time. The depositional and diagenetic histories of each

945 section and subsection impacted the TOC, S, clay, and carbonate contents, and thus

946 ultimately the Hg contents. While we lack detailed characterization of Hg associations in

947 each section, we can make some general observations based on Hg-TOC correlations and

948 independent information about each section, such as presence or absence of sulfide

949 minerals. In the New York Canyon section, the host phase for Hg in the post-extinction

950 interval is likely dominantly TOC (based on their close correlation). At St. Audrie's Bay,

951 Hg in likely hosted by TOC. In the Lombardy basin sections, TOC may be the dominant

952 host, although it seems likely that carbonate dilution plays a larger role in the measured Hg

953 contents than at other sections. The Levanto section Hg contents are negatively correlated

954 with carbonates, suggesting dilution may play a major role in determining the Hg contents

955 in these rocks. The host phase for Hg at the Levanto section may be clays, as TOC appears

956   to play a minor role (based on the low correlations) and there is no evidence abundant S

957   (based on the absence of sulfide phases in thin sections).

958   *6.4.2 Possible paleo-environmental controls on depositional effects*

959   Depth and proximity to continental sources may influence Hg contents in the

960   sedimentary record (e.g., Them et al., 2019; see their fig. 6). The rapid changes in

961   depositional environment seen at shallow sections mean environmental parameters that can

962   affect both Hg and TOC, as well with other rock materials including carbonate, may affect

963   Hg concentrations and Hg/TOC ratios. We illustrate these changes in Fig. 11 and briefly

964   discuss some below, in terms of their potential influence on the ETE Hg records studied

965   here.

966   Carbonate deposition and diagenesis may have important effects on geochemical

967   proxies, especially in carbonate-rich rocks (typical of many marine sediments), (e.g.,

968   Arvidson and Morse, 2014). Dilution occurs whereby increased carbonate content leads to

969   lower TOC and Hg contents. Mercury contents can be calculated on a carbonate-free-basis

970   to correct for dilution (see Fig S4–6), but this correction does not remove the high Hg and

971   Hg/TOC in the Hettangian at Levanto and Lombardy. Another important process in

972   carbonate rich rocks is the migration of carbonate during early diagenesis, possibly leaving

973   organic C and associated Hg behind. In the Hettangian and ETE portions of the Levanto

974   section, Hg is enriched in samples from thin beds with lower carbonate content and

975   depleted in carbonate-rich beds (Fig. SI 5), and carbonate and Hg contents are negatively

976   correlated (Fig. 7h). These patterns could be due to migration of carbonate and/or dilution,

977   but on its own, the bed-to-bed alternating enrichment and depletion does not explain the

978   overall relatively elevated Hg/TOC throughout this interval at Levanto. We note that at any

979 rate these aren't particularly elevated contents (e.g., see fig. 11 from Charbonnier et al.,

980 2020).

981   Processes controlling organic C preservation are also expected to influence

982 Hg/TOC ratios. Organic C in marine sediments is subject to degradation that greatly

983 depends on water depth, local productivity, oxygen exposure time at the sediment water

984 interface, and sedimentary reworking (see also Charbonnier et al., 2020). Changes in depth

985 at St. Audrie's Bay may have contributed to the very low TOC (and potentially high

986 Hg/TOC) during the ETE at this section. Although there is no evidence for similar changes

987 in depth at Levanto that would have led to preferential TOC loss and the observed Hg/TOC

988 enrichment in the Hettangian, it is difficult to rule out all possible changes that could have

989 affected organic C preservation (e.g., Charbonnier et al., 2020).

990   Widespread anoxia and/or euxinia associated with the Triassic–Jurassic boundary

991 (Schootbrugge et al. 2008; Jost et al., 2017), like many other extinction events, may have

992 favored the deposition of sulfides and associated Hg enrichment (e.g., Bower et al., 2008;

993 Han et al., 2014; He et al., 2020). Large sedimentary pyrites are found in the Hettangian at

994 St. Audrie's Bay (Hesselbo et al., 2004; Wignall, 2001). Such an increase in anoxia would

995 likely affect sedimentary S deposition, and therefore may have also affected sedimentary

996 Hg deposition.

997 *6.4.3 Interpretation of Hg/TOC ratios from low-TOC samples*

998   The complexity of host phases and depositional effects on Hg content is magnified

999 when interpreting Hg/TOC ratios in samples with particularly low TOC contents. While

1000 previous studies have regarded high Hg/TOC ratios in samples with TOC <0.2 wt.% as

1001 artifacts (see Section 2.1) or difficult to interpret, we instead suggest that they reflect an

1002   additional, and important, layer of complexity arising from the processes that sequester Hg

1003   and its associated elements in sediments. High Hg/TOC ratios associated with low

1004   TOC content are not artifacts per se, in that they do represent Hg enrichment relative to

1005   TOC. However, at low TOC values, small changes in TOC can lead to large changes in

1006   Hg/TOC despite little change in Hg contetns. The more relevant question is whether these

1007   represent the enrichment of Hg in association with TOC, or the increasing importance of

1008   Hg hosted in other phases — emphasizing the need to better understand Hg distribution in

1009   sedimentary rocks, especially in those with low TOC content (see also Shen et al., 2020).

1010   Recent studies have greatly expanded our understanding of the role of S sequestration in

1011   Hg 'anomalies' (Shen et al., 2019a; Shen et al., 2020), and demonstrated that sediments

1012   deposited in highly-reducing, highly sulfidic environments sequester Hg to an extent that

1013   obscures any global Hg signal (Shen et al., 2019a). Similar micro-scale study of carbonate-

1014   rich, sulfide-poor lithologies such as those from Levanto and the Lombardy Basin could

1015   help illuminate the controls on Hg distribution in these low TOC rocks.

1016   *6.4.4 Summary of depositional effects*

1017   Mercury records in marine sedimentary rocks across the ETE, and at other

1018   extinction events, are likely affected by a combination of all the processes discussed above.

1019   Ocean anoxia and euxinia (from CO_2-induced global warming and increased nutrient

1020   runoff), increased Hg, and increased S are all possible direct and indirect consequences of

1021   CAMP, and elevated Hg and Hg/TOC may be affected by a combination of these effects.

1022   Further, changes in the carbonate system (e.g., Greene et al., 2012), global redox (e.g.,

1023   Schootbrugge et al., 2013; Kasprak et al., 2015; Jost et al., 2017) or changes in delivery of

1024   clay (e.g., from increased weathering) are all downstream consequences of LIP magmatism

1025 that also could have impacted Hg delivery and/or preservation. Thus we expect a general

1026 connection between LIP activity and Hg, as observed consistently in our ETE sections, but

1027 the causes of that enrichment, and the timing of Hg anomalies, may be complex. Notably,

1028 the persistent Hg enrichment in the Hettangian that we see at both Levanto and Lombardy

1029 seems difficult to explain by one depositional effect alone, since the elevated Hg/TOC

1030 ratios are observed in carbonate-rich, low-TOC samples in Lombardy but the carbonate-

1031 poor, higher TOC samples from the Hettangian at Levanto.

1032 Better understanding of depositional effects, and distinguishing between them, may

1033 provide a more robust foundation for using Hg contents as a proxy for magmatism. What

1034 is clear is that the relatively poor correspondence between CAMP dates and the timing of

1035 the Hg/TOC anomaly at Levanto casts doubt the use of Hg contents from a single site to

1036 fingerprint the timing of discrete pulses of magmatism directly (cf. Percival et al., 2017),

1037 at least in the absence of independent constraints on sedimentary reworking of Hg.

1038

1039 **6.5 Next steps**

1040 Our examination of Hg in different depositional environments across the Triassic–

1041 Jurassic boundary corroborates other recent studies noting the complexity in depositional

1042 and diagenetic controls on Hg concentrations and Hg/TOC ratios in sedimentary rocks

1043 (Faggetter et al., 2019; Percival et al., 2018; Shen et al., 2019a,b, 2020; Them et al., 2019;

1044 Charbonnier et al., 2020). At the same time, our results show that, despite the complexity,

1045 there may be a reliable signal of CAMP magmatism recorded by the combination of

1046 Hg/TOC and Hg isotopes across a range of depositional environments at the end–Triassic.

1047 When considered together, Hg isotopes and contents may provide valuable information

1048   about LIP magmatism in sedimentary sections, but the numerous processes influencing Hg

1049   contents and Hg/TOC ratios suggest caution is warranted when applying these

1050   measurements alone, especially when using single-site records and when seeking to

1051   identify detailed timing.

1052   Future studies of the Hg proxy in deep time should aim to investigate where in the

1053   rock Hg is found, specifically what minerals host Hg and what other elements it is

1054   associated with in each sample (e.g., organic C versus S; Shen et al., 2020). As suggested

1055   by Grasby et al. (2019), total S measurements may help elucidate the role of S sequestration

1056   in the Hg anomalies. Similarly, carefully relating Hg contents and Hg/TOC ratios to other

1057   geochemical records (e.g., of local anoxia), as well as investigating sequence stratigraphic

1058   changes within a record, provide opportunities for better informing interpretation of

1059   apparent Hg anomalies. Further understanding may come from exploring more Hg records

1060   when LIP magmatism is not expected. Finally, more connection to research on Hg in

1061   modern marine sediments could help to elucidate how Hg is delivered to and preserved in

1062   sedimentary environments.

1063

1064   **7. CONCLUSIONS**

1065   We studied five sections spanning the Triassic–Jurassic boundary, and in the four

1066   sections with measurable Hg (excluding the carbonate platform of Mt. Sparagio (Sicily)

1067   which had Hg below the detection limit), Hg/TOC increases in association with the ETE

1068   and CAMP emplacement. At New York Canyon and St. Audrie's Bay, peaks in Hg/TOC

1069   are well defined and correspond well with the timing of the end–Triassic extinction as

1070   inferred from stratigraphic relationships and organic C isotopes. In contrast, at the Levanto

47

and Lombardy sections, Hg/TOC remains elevated for much longer, well into the Hettangian. The well-constrained absolute chronology from Levanto shows that the long-lasting elevated Hg/TOC persists well after the youngest U–Pb date of CAMP activity. These results suggest abundant caution is needed when inferring the timing of mass volcanism from Hg anomalies in sedimentary rocks, particularly from a single stratigraphic section, and in the example studied here, even with the benefit of multiple sections.

Mercury isotope signatures across the studied sections are consistent with the expected signatures based on each site's depositional environment. Shallow, nearshore sites have negative δ^{202}Hg and Δ^{199}Hg values, and the New York Canyon data are consistent with existing interpretations of shallowing upwards at this locality. In the deeper basinal setting of Levanto, slightly positive MIF are indicative of an atmospheric and marine-influenced Hg source, but δ^{202}Hg values suggest some continental influence on MDF signatures. At each site, MIF is near or approaches zero during the ETE and CAMP magmatism, supporting the idea that a large source of volcanic Hg may have overwhelmed the surficial Hg cycle at the time, leading to a notable reduction in the Hg MIF signature recorded in sedimentary rocks. Differences between Hg isotope behavior during this time interval versus the end-Permian raise the intriguing possibility of whether Hg isotopes offer clues about volatile recycling associated with massive volcanism in the geologic past. Overall, our work suggests that, despite the complex processes that are increasingly recognized as influencing sedimentary Hg records, the Hg proxy can still be informative — but that it can be most reliably applied by studying sections that span diverse depositional environments and by including measurements of Hg isotope ratios as well as concentrations.

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Acknowledgements

1111

FIGURE CAPTIONS

Fig. 1. [A] [A] Late Triassic paleogeography with paleogeographic locations of studied sections, adapted from Kuroda et al. (2010). 1: St. Audrie's Bay, UK; 2: New York Canyon, Nevada; 3: Lombardy Basin, Italy; 4: Mt. Sparagio, Sicily, Italy; 5: Levanto, Peru. [B] Relative depositional settings of study sites and expected Hg, TOC, and Hg isotope data at

1117 each site based on relative depth and energy of environment. This is a simplified

1118 representation of how Hg enters the marine record, with particular attention to Hg isotopes

1119 and relative contributions based on proximity to the continent and depth, and these sites

1120 were thousands of kms apart during actual deposition. Based on Thibodeau and Bergquist

1121 (2016) and references therein.

1122

1123 **Figure 2.** Carbon isotope correlation of sections studied here. Mt. Sparagio data and

1124 stratigraphic column is from Todaro et al. (2018); Lombardy basin data is from Zaffani et

1125 al. (2018), Bachan et al. (2012), and this study; St. Audrie's Bay data is from this study

1126 and stratigraphic column is from Hesselbo et al. (2004); New York Canyon data is from

1127 Thibodeau et al. (2017) and this study; Levanto data is from Yager et al. (2017). Note that

1128 this correlation forms the basis for Fig. 9. Gray shape denotes the initial negative isotope

1129 excursion. Triassic–Jurassic boundary is placed at the first occurrence of *P. spelae* for the

1130 Levanto and New York Canyon sections and is based on Todaro et al. (2018), Zaffani et

1131 al. (2018), and Hesselbo et al. (2004) for the Mt. Sparagio, Lombardy basin, and St.

1132 Audrie's bay sections, respectively.

1133

1134 **Figure 3.** Stratigraphic column from New York Canyon, Nevada (Corsetti et al. 2015) with

1135 δ^{13}Corg, Hg, TOC, Hg/TOC, δ^{202}Hg (MDF), and Δ^{199}Hg (MIF, vertical gray bar denotes

1136 no MIF) from this study and Thibodeau et al. (2016). Horizontal gray bar denotes ETE.

1137 Colored bar on Hg/TOC plot adapted from fig. 12 of Charbonnier et al. (2020), and

1138 provides relative guide for Hg/TOC anomalies.

1139

1140   **Figure 4.** Stratigraphic column from St. Audrie's Bay, UK (Hesselbo et al. 2004) with

1141 δ^{13}Corg, Hg, TOC, Hg/TOC, δ^{202}Hg (MDF), and Δ^{199}Hg (MIF, vertical gray bar denotes

1142   no MIF) from this study. Horizontal gray bar denotes ETE. "Will" refers to Williton

1143   member; "Lils." Refers to Lilstock Fm. Horizontal gray bar denotes ETE. Mercury data

1144   from Percival et al. (2017) and Hg/TOC data (calculated from Hesselbo et al., 2002 and

1145   Percival et al., 2017) are also shown. Note that TOC data was measured using different

1146   methods. The discrepancies between both sections in TOC measurements, particularly in

1147   the Lilstock formation, result in a large discrepancy between Hg/TOC measurements.

1148   These may be due to TOC methodologies, heterogeneity in the section from bed to bed, or

1149   both. Colored bar in Hg/TOC plot as in Fig. 3.

1150

1151   **Figure 5.** Composite stratigraphic heights from Italian sections (for lithologic logs for

1152   individual sections, see SI Fig. 2). Formation name and section name are displayed, with

1153 δ^{13}Corg, Hg, TOC and Hg/TOC. Zu = Calcari di Zu Formation; M = Malanotte Formation;

1154   Moltrasio = Calcare di Moltrasio Fm; stratigraphic logs are based on Galli et al. (2007),

1155   Jadoul et al. (2012), Zaffani et al. (2018), and this study. Horizontal gray bar denotes ETE.

1156   Colored bar in Hg/TOC plot as in Fig. 3.

1157

1158   **Fig. 6.** Stratigraphic column from Levanto, Peru (Yager et al., 2017) with δ^{13}Corg, Hg,

1159   TOC, CC, Hg/TOC, δ^{202}Hg (MDF), and Δ^{199}Hg (MIF; vertical gray bar denotes no MIF)

1160   from this study. Horizontal gray bar denotes ETE. Colored bar in Hg/TOC plot as in Fig.

1161   3.

1162

Fig. 7. Hg/TOC data and Hg isotope data (MIF) plotted with stratigraphy for St. Audrie's Bay, Levanto, and New York Canyon related based on organic C isotope correlation; horizontal gray bars indicate ETE. Stratigraphic column for New York Canyon is from Thibodeau et al. (2016); stratigraphic column from St. Audrie's Bay is from Hesselbo et al. (2004); stratigraphic column from Levanto is from Yager et al. (2017). Data from the lower portion of the New York Canyon section is from Thibodeau et al. (2016).

Fig. 8. Summary of Hg isotope data in this and previous studies.

Fig. 9. Hg and Hg/TOC from this study with summary of anoxia/euxinia proxies. Gray bar denotes ETE based on the initial carbon isotope excursion in Fig. 2. Black boxes denote possible euxinia at those sections, which do not align with high Hg/TOC or Hg or at a single point in time. Mt. Sparagio is from $\delta^{34}S_{cas}$ data from He et al. (2020); Lombardy basin data is from $\delta^{238}U$ from Jost et al. (2017); St. Audrie's Bay data is from pyrite framboids reported in Hesselbo et al. (2004); New York Canyon data is from Larina et al. (unpublished data); Levanto data is from Yager et al. (unpublished data).

Fig. 10. Levanto section Hg/TOC data (this study) plotted using the age model from Yager et al. (2017) and displayed with CAMP U–Pb age dates from Davies et al. (2017). High Hg/TOC was expected to overlap with CAMP U–Pb ages, but elevated Hg persists for much longer and does not begin coincident with the earliest known CAMP ages.

1185   **Fig. 11.** Schematic displaying example ways LIP magmatism may impact the Hg cycle

1186   with respect to measured Hg contents and Hg/TOC.

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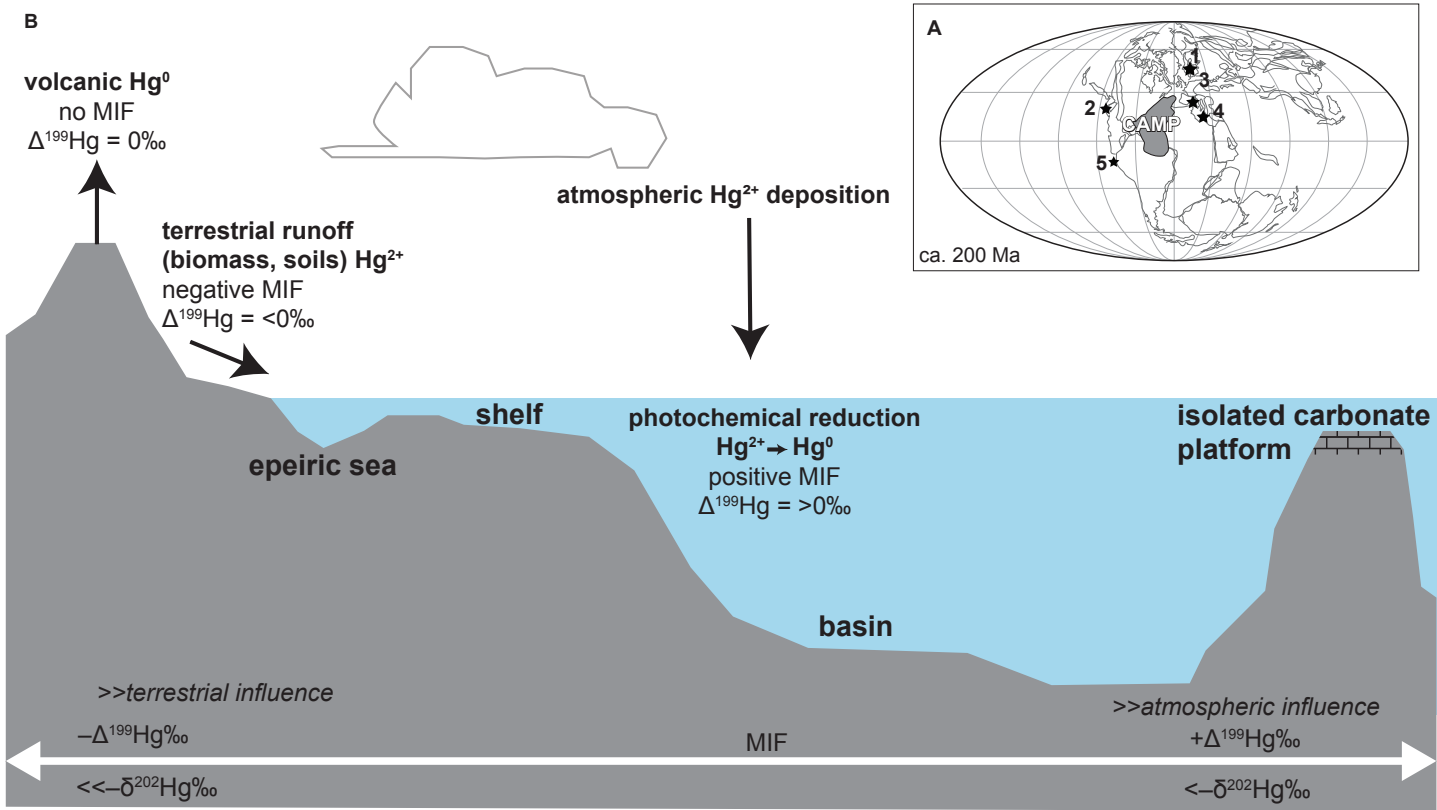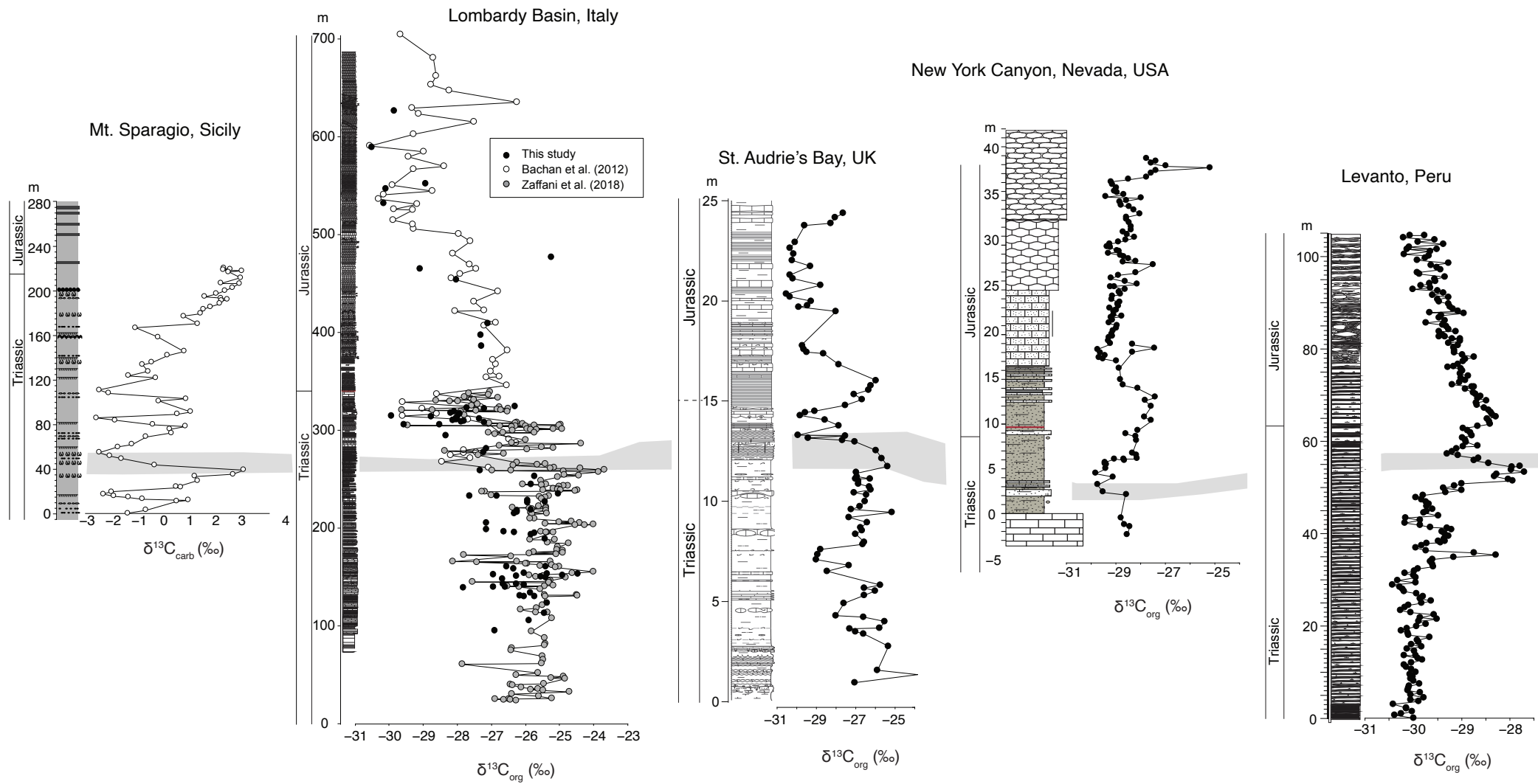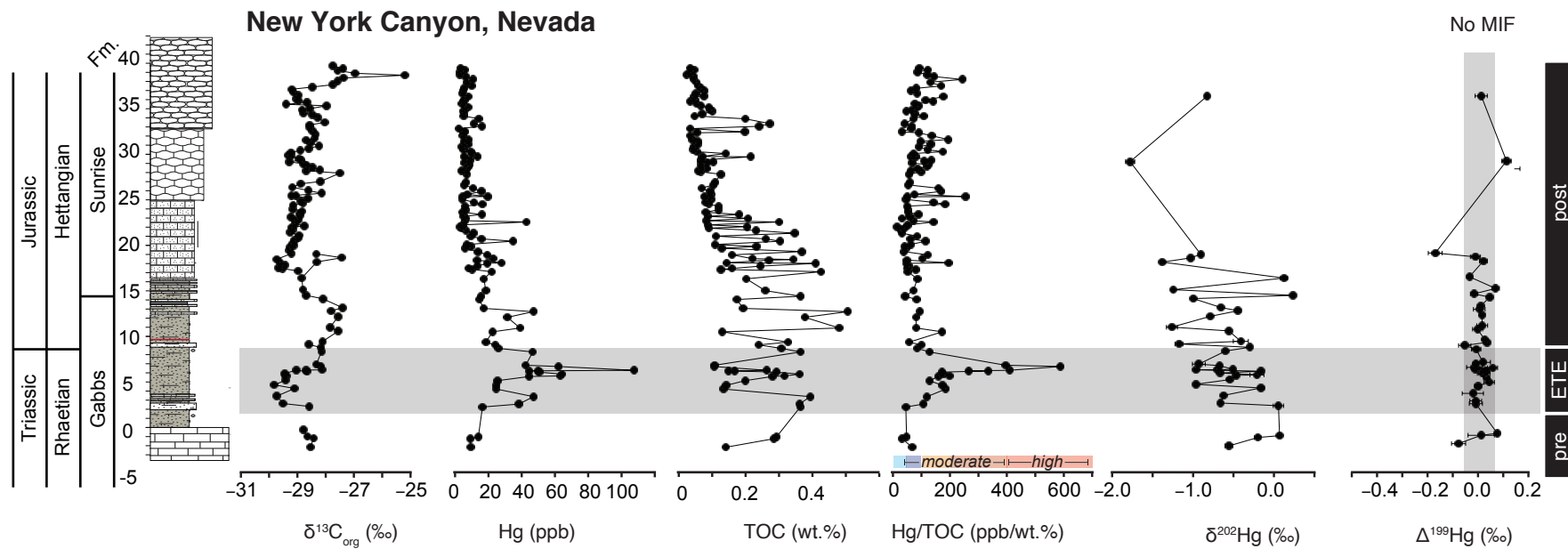1580   10.1038/s43017-021-00146-y
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Figure 1

B

volcanic Hg⁰
no MIF
$\Delta^{199}Hg = 0‰$

**terrestrial runoff
(biomass, soils) Hg²⁺**
negative MIF
$\Delta^{199}Hg = <0‰$

atmospheric Hg²⁺ deposition

A

CAMP

ca. 200 Ma

shelf

epeiric sea

**photochemical reduction
Hg²⁺ → Hg⁰**
positive MIF
$\Delta^{199}Hg = >0‰$

**isolated carbonate
platform**

basin

>>*terrestrial influence*
$-\Delta^{199}Hg‰$

MIF

>>*atmospheric influence*
$+\Delta^{199}Hg‰$

$<<-\delta^{202}Hg‰$

$<-\delta^{202}Hg‰$

FIGURE 2

Mt. Sparagio, Sicily

δ¹³C_carb (‰)

Lombardy Basin, Italy

- This study
- ○ Bachan et al. (2012)
- ● Zaffani et al. (2018)

δ¹³C_org (‰)

St. Audrie's Bay, UK

δ¹³C_org (‰)

New York Canyon, Nevada, USA

δ¹³C_org (‰)

Levanto, Peru

δ¹³C_org (‰)

FIGURE 3

New York Canyon, Nevada

δ13C_org (‰)   Hg (ppb)   TOC (wt.%)   Hg/TOC (ppb/wt.%)   δ202Hg (‰)   Δ199Hg (‰)

FIGURE 4

St. Audrie's Bay, UK

δ13C_org (‰)   Hg (ppb)   TOC (wt.%)   Hg/TOC (ppb/wt.%)   δ202Hg (‰)   Δ199Hg (‰)

FIGURE 5

Lombardy Basin, Italy (composite)
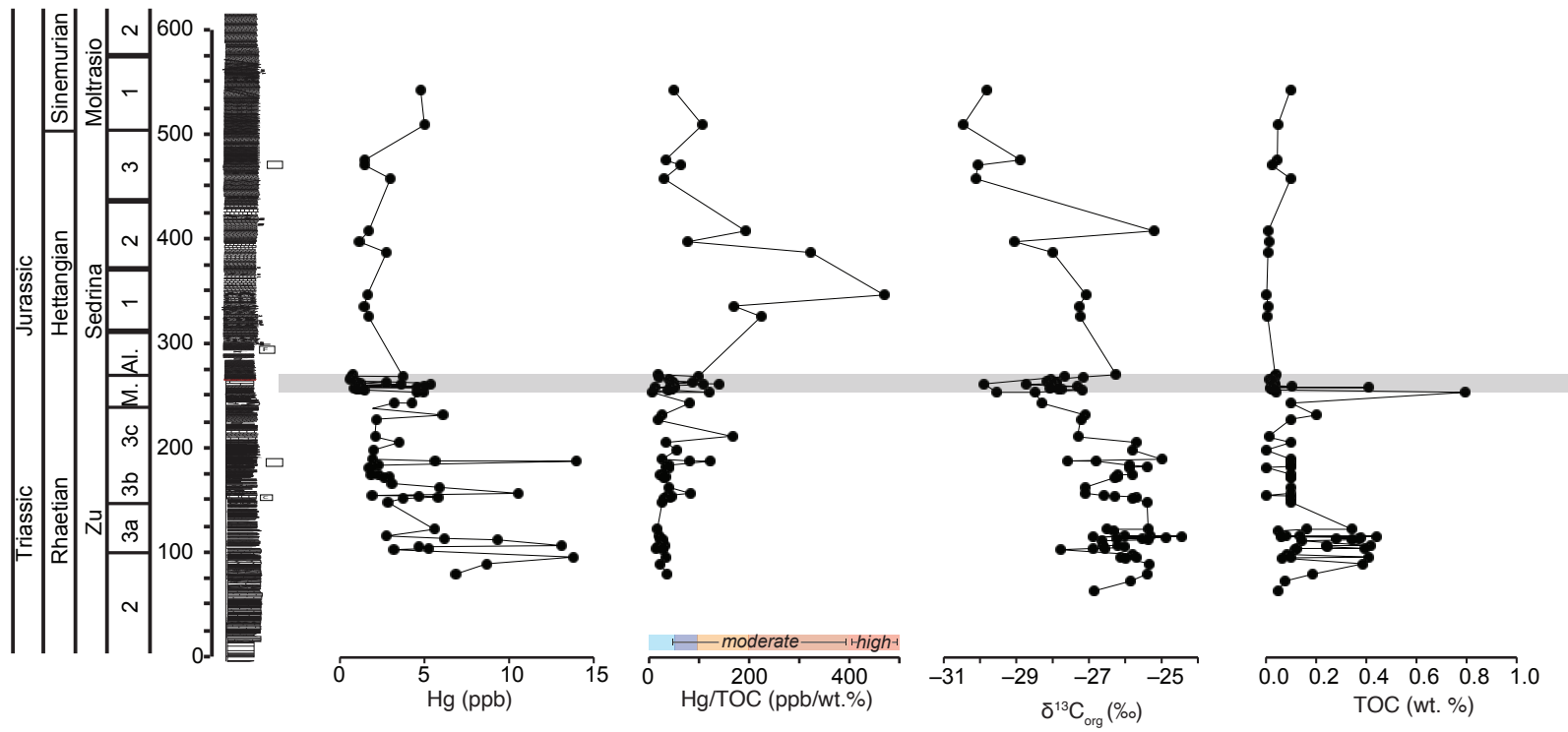
FIGURE 6

Levanto, Peru

FIGURE 7

FIGURE 8

Triassic–Jurassic (this study)

St. Audrie's Bay

deeper

New York Canyon

Levanto

Cretaceous–Paleogene

Various (Sial 2016)
not inc. high-MIF samples from Bidart (less than –0.5‰)

Toarcian OAE

East Tributary of Bighorn Creek
(Them 2019)

Permian–Triassic

Guryul Ravine (Wang 2019)

Chaohu (Wang 2019)

Buchanan Lake (Grasby 2017)

Daxiakou (Wang 2018)

Shangsi (Wang 2018)

Gujo-Hachiman (Shen 2019)

Xiaokuo (Shen 2019)

Meishan (Shen 2019)

Meishan
(Grasby 2017 & Wang 2018)

For each dataset:
○ *post-excursion*
● *syn-excursion*
○ *pre-excursion*

-0.50   -0.40   -0.30   -0.20   -0.10   0.00   0.10   0.20   0.30   0.40   0.50
$\Delta^{199}Hg$ (‰)

-3.00   -2.00   -1.00   0.00   1.00
$\delta^{202}Hg$ (‰)

FIGURE 9

Mt. Sparagio, Sicily

Lombardy Basin, Italy

St. Audrie's Bay, UK

New York Canyon, Nevada, USA

Levanto, Peru

● Hg ppb
○ Hg/TOC

FIGURE 10

FIGURE 11