

# 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

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24 **ABSTRACT**

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

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<sub>2</sub>)  
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<sub>2</sub> 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

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

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., 2019).

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  $^{202}\text{Hg}/^{198}\text{Hg}$  ratio as  $\delta^{202}\text{Hg}$  (where  $\delta^{202}\text{Hg}$  (‰) =  $[(^{202}\text{Hg}/^{198}\text{Hg})_{\text{sample}}/(^{202}\text{Hg}/^{198}\text{Hg})_{\text{standard}} -$   
182  $1] \times 1000$ ). 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  $\delta$  value (e.g.,  $\delta^{199}\text{Hg}$ ) and the MDF predicted  
185 using the measured  $\delta^{202}\text{Hg}$  value and the kinetic MDF law; this difference is typically  
186 reported as  $\Delta^{199}\text{Hg}$  for odd isotope MIF.

187 Gaseous elemental Hg emitted from volcanoes enters the atmosphere with no  
188 measurable MIF ( $\Delta^{199}\text{Hg} = 0\text{‰}$ ; Fig. 1a) and with negative to zero MDF ( $\delta^{202}\text{Hg} = -2\text{‰}$   
189 to  $0\text{‰}$ ; 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  $\Delta^{199}\text{Hg}$ ) and much more negative  $\delta^{202}\text{Hg}$  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  $\Delta^{199}\text{Hg}$  and  $\Delta^{201}\text{Hg}$   
203 values, or “positive MIF” and less negative  $\delta^{202}\text{Hg}$ ), 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

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 ( $\delta^{13}\text{C}_{\text{org}}$ ) 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  $\delta^{13}\text{C}_{\text{org}}$  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  $\delta^{13}\text{C}$  (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

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

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 Italcementi 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 ( $\delta^{13}\text{C}$ ); 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 calcisphaerae. 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 biostratigraphic 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<sub>2</sub> flow at 350 ml/min, heated in a drying step to  
513 300°C for 40 s, then decomposed for 300 s at 800°C. Following combustion, evolved gases  
514 were carried through a heated (600°C) 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 600°C 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<sub>2</sub>SO<sub>4</sub> (v/v) and ~1% KMnO<sub>4</sub> (w/w), which oxidized the Hg<sup>0</sup> gas to Hg<sup>II</sup>.  
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<sub>2</sub>OH-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<sub>2</sub>OH-HCl and diluted to 1–2 ng/g with a matrix matched solution. Mercury was  
563 introduced to the plasma as Hg<sup>0</sup> after SnCl<sub>2</sub> reduction and Hg<sup>0</sup> 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 <sup>204</sup>Pb  
569 was monitored using <sup>206</sup>Pb, but was always negligible (correction never altered the  
570 calculated δ<sup>204</sup>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  $2\sigma$  (standard error mean) of sample replicates or the  $2\sigma$  (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  $\delta^{13}\text{C}$  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  $\delta^{13}\text{C}$  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

594 Canyon using both methods, and results were within the analytical uncertainty for both  
595 TOC and  $\delta^{13}\text{C}_{\text{org}}$  measurements, supporting either method as a viable means of TOC and  
596  $\delta^{13}\text{C}_{\text{org}}$  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  $\text{CO}_2$  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  $\pm 0.08$  on average, or approximately  $\pm 3\%$   
608 of the measured value ( $1\sigma$  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<sub>3</sub>PO<sub>4</sub>. 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<sub>2</sub>. Samples  
620 were then run on a Picarro CRDS coupled to an Automate preparation device, which  
621 sparges the solution with N<sub>2</sub> gas to drive CO<sub>2</sub> 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<sub>3</sub>. 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,  $\delta^{202}\text{Hg}$ , and  $\Delta^{199}\text{Hg}$  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  $\sim 12 \pm 4$  ppb  
655 ( $n = 4$ ; 1 s.d.). This value rises to  $\sim 50 \pm 21$  ppb ( $n = 14$ , 1 s.d.) during the ETE, and then  
656 decreases to  $11 \pm 10$  ppb ( $n = 100$ , 1 s.d.) during the Early Jurassic. Ratios of Hg/TOC have  
657 average values of  $48 \pm 15$  ppb/wt.% ( $n = 4$ ; 1 s.d.) during the pre-extinction interval, rise  
658 to an average of  $244 \pm 139$  ppb/wt.% ( $n = 14$ ; 1 s.d.) during the ETE, and decrease to an  
659 average value of  $92 \pm 48$  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  $\delta^{202}\text{Hg}$  values during the Rhaetian and early Hettangian are between  $\sim 0\text{‰}$  and  $-1.5\text{‰}$ ,

662 becoming progressively more negative upsection;  $\Delta^{199}\text{Hg}$  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  $\sim 29 \pm 15$  ppb ( $n = 38$ , 1 s.d.), during the ETE (correlated using the  
671 negative CIE in organic C isotopes) average Hg contents are  $19 \pm 15$  ppb ( $n = 13$ ; 1 s.d.),  
672 and during the Early Jurassic Hg contents are  $42 \pm 29$  ppb ( $n = 29$ ; 1 s.d.). Ratios of  
673 Hg/TOC during the pre-extinction interval are on average  $43 \pm 27$  ppb/wt.%TOC ( $n = 38$ ;  
674 1 s.d.), during the ETE are  $102 \pm 77$  ppb/wt.% ( $n = 13$ ; 1 s.d.), and during the Early Jurassic  
675 are  $18 \pm 20$  ppb/wt.%TOC ( $n = 29$ ; 1 s.d.). Ratios of Hg/TOC rise to  $\sim 150$  ppb/wt.%  
676 coincident with the onset of the negative C isotope excursion (meter  $\sim 12$ ) and then decrease  
677 to an average of  $\sim 20$  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

684 sampling or TOC methodological differences; the implications for this difference are  
685 discussed in the figure caption.

686 Values of  $\delta^{202}\text{Hg}$  at St. Audrie's Bay are similar throughout the measured section  
687 and range between  $-1.50\text{‰}$  and  $-0.50\text{‰}$ ;  $\Delta^{199}\text{Hg}$  values are negative during the Rhaetian  
688 ( $-0.20\text{‰}$  to  $-0.40\text{‰}$ ), increase to about  $-0.10\text{‰}$  during the ETE, and decrease again during  
689 the Hettangian ( $-0.20\text{‰}$  to  $-0.40\text{‰}$ ).

### 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  $\sim 6$  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  $7 \pm 4$  ppm ( $n = 11$ ,  
699 1 s.d.), TOC average of  $0.2 \pm 0.1\%$  ( $n = 26$ ; 1 s.d.), and Hg/TOC average of  $25 \pm 7$  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  $\sim 3$  ppb) in  
704 resistant, carbonate-rich beds and higher (between  $\sim 5$  and 14 ppb) in marly, mudstone beds.

705 Samples from the Italcementi quarry section (Fig. S1) have average Hg contents of  
706  $4 \pm 3$  ppb ( $n = 25$ , 1 s.d.; not including 32 samples below detection limit), TOC contents



707 of  $0.1 \pm 0.04$  wt.% ( $n = 22$ , 1 s.d.) and Hg/TOC ratios of  $44 \pm 26$  ( $n = 22$ ; 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  $\sim 2 \pm 1.5$   
711 ppb ( $n = 18$ ; 1 s.d.), TOC contents of  $0.08 \pm 0.16$  wt.% ( $n = 27$ ; 1 s.d.) and Hg/TOC values  
712 of  $44 \pm 25$  ppb/wt.% ( $n = 16$ ; 1 s.d.).

713 The Val Adrara section (Fig. S1) contains average Hg contents of  $2.7 \pm 1.4$  ppb ( $n$   
714  $= 19$ ; 1 s.d.), TOC concentrations of  $0.03 \pm 0.03$  wt.% ( $n = 15$ ; 1 s.d.) and Hg/TOC of  $140$   
715  $\pm 119$  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  $33 \pm 14$  ppb during the Rhaetian pre-extinction interval, ( $n = 84$ ; 1 s.d.),  $37 \pm$   
721  $24$  ppb ( $n = 14$ ; 1 s.d.) during the negative CIE and ETE Hg concentrations, and  $36 \pm 19$   
722 ppb following the ETE during the Hettangian ( $n = 96$ ; 1 s.d.). Hg/TOC during the pre-  
723 extinction interval is  $24 \pm 25$  ppb/wt.% ( $n = 84$ ; 1 s.d.), during the ETE is  $30 \pm 29$  ppb/wt.%  
724 ( $n = 14$ ; 1 s.d.), and following the ETE is  $64 \pm 36$  ppb/wt.% ( $n = 96$ ; 1 s.d.). There is an  
725 increase in Hg/TOC at  $\sim 57$  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  $\delta^{202}\text{Hg}$

730 are between about  $-1.30\text{‰}$  and  $-0.30\text{‰}$  for the measured section, with a possible excursion  
731 from  $-0.3\text{‰}$  to  $-1.5\text{‰}$  during the late Rhaetian and ETE;  $\Delta^{199}\text{Hg}$  is slightly positive during  
732 the Rhaetian ( $\sim 0.1\text{‰}$ ) and near zero without discernable MIF during the ETE and  
733 Hettangian ( $\pm 0.05\text{‰}$ ).

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

#### 745 **5.5 Consistent observations between sections**

746 Using organic C isotopes as a correlation tool, at all four sections Hg/TOC rises  
747 during the negative CIE and, by inference, during the contemporaneous ETE. At New York  
748 Canyon and St. Audrie's Bay, Hg/TOC decreases during the Early Jurassic after its peak  
749 associated with the CIE and ETE (Fig. 7). In contrast, at Levanto, Hg/TOC increases during  
750 the negative CIE and ETE and then increases further in the early Jurassic, remaining  
751 elevated for the remainder of the section (Fig. 7). In the Lombardy Basin, Hg/TOC is  
752 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  $\Delta^{199}\text{Hg}$ ) during the ETE. New York Canyon and Levanto  
762 record a slightly negative shift in  $\delta^{202}\text{Hg}$  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 ( $\Delta^{199}\text{Hg}$ )  
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 deposition  
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 noisy 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  $\delta^{202}\text{Hg}$  values coincident with upward shallowing through the  
827 section. The increasingly negative  $\delta^{202}\text{Hg}$  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  $\delta^{202}\text{Hg}$  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;  $201.63 \pm 0.029$   
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 is 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 contents. 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<sub>2</sub>-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



1071 and Lombardy sections, Hg/TOC remains elevated for much longer, well into the  
1072 Hettangian. The well-constrained absolute chronology from Levanto shows that the long-  
1073 lasting elevated Hg/TOC persists well after the youngest U–Pb date of CAMP activity.  
1074 These results suggest abundant caution is needed when inferring the timing of mass  
1075 volcanism from Hg anomalies in sedimentary rocks, particularly from a single stratigraphic  
1076 section, and in the example studied here, even with the benefit of multiple sections.

1077 Mercury isotope signatures across the studied sections are consistent with the  
1078 expected signatures based on each site’s depositional environment. Shallow, nearshore  
1079 sites have negative  $\delta^{202}\text{Hg}$  and  $\Delta^{199}\text{Hg}$  values, and the New York Canyon data are  
1080 consistent with existing interpretations of shallowing upwards at this locality. In the deeper  
1081 basinal setting of Levanto, slightly positive MIF are indicative of an atmospheric and  
1082 marine-influenced Hg source, but  $\delta^{202}\text{Hg}$  values suggest some continental influence on  
1083 MDF signatures. At each site, MIF is near or approaches zero during the ETE and CAMP  
1084 magmatism, supporting the idea that a large source of volcanic Hg may have overwhelmed  
1085 the surficial Hg cycle at the time, leading to a notable reduction in the Hg MIF signature  
1086 recorded in sedimentary rocks. Differences between Hg isotope behavior during this time  
1087 interval versus the end-Permian raise the intriguing possibility of whether Hg isotopes offer  
1088 clues about volatile recycling associated with massive volcanism in the geologic past.  
1089 Overall, our work suggests that, despite the complex processes that are increasingly  
1090 recognized as influencing sedimentary Hg records, the Hg proxy can still be informative  
1091 — but that it can be most reliably applied by studying sections that span diverse  
1092 depositional environments and by including measurements of Hg isotope ratios as well as  
1093 concentrations.

1094

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1111

1112 **FIGURE CAPTIONS**

1113 **Fig. 1.** [A] [A] Late Triassic paleogeography with paleogeographic locations of studied  
1114 sections, adapted from Kuroda et al. (2010). 1: St. Audrie's Bay, UK; 2: New York Canyon,  
1115 Nevada; 3: Lombardy Basin, Italy; 4: Mt. Sparagio, Sicily, Italy; 5: Levanto, Peru. [B]  
1116 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  $\delta^{13}\text{C}_{\text{org}}$ , Hg, TOC, Hg/TOC,  $\delta^{202}\text{Hg}$  (MDF), and  $\Delta^{199}\text{Hg}$  (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  $\delta^{13}\text{C}_{\text{org}}$ , Hg, TOC, Hg/TOC,  $\delta^{202}\text{Hg}$  (MDF), and  $\Delta^{199}\text{Hg}$  (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  $\delta^{13}\text{C}_{\text{org}}$ , 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  $\delta^{13}\text{C}_{\text{org}}$ , Hg,  
1159 TOC, CC, Hg/TOC,  $\delta^{202}\text{Hg}$  (MDF), and  $\Delta^{199}\text{Hg}$  (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

1163 **Fig. 7.** Hg/TOC data and Hg isotope data (MIF) plotted with stratigraphy for St. Audrie's  
1164 Bay, Levanto, and New York Canyon related based on organic C isotope correlation;  
1165 horizontal gray bars indicate ETE. Stratigraphic column for New York Canyon is from  
1166 Thibodeau et al. (2016); stratigraphic column from St. Audrie's Bay is from Hesselbo et  
1167 al. (2004); stratigraphic column from Levanto is from Yager et al. (2017). Data from the  
1168 lower portion of the New York Canyon section is from Thibodeau et al. (2016).

1169

1170 **Fig. 8.** Summary of Hg isotope data in this and previous studies.

1171

1172 **Fig. 9.** Hg and Hg/TOC from this study with summary of anoxia/euxinia proxies. Gray bar  
1173 denotes ETE based on the initial carbon isotope excursion in Fig. 2. Black boxes denote  
1174 possible euxinia at those sections, which do not align with high Hg/TOC or Hg or at a  
1175 single point in time. Mt. Sparagio is from  $\delta^{34}\text{S}_{\text{cas}}$  data from He et al. (2020); Lombardy  
1176 basin data is from  $\delta^{238}\text{U}$  from Jost et al. (2017); St. Audrie's Bay data is from pyrite  
1177 framboids reported in Hesselbo et al. (2004); New York Canyon data is from Larina et al.  
1178 (unpublished data); Levanto data is from Yager et al. (unpublished data).

1179

1180 **Fig. 10.** Levanto section Hg/TOC data (this study) plotted using the age model from Yager  
1181 et al. (2017) and displayed with CAMP U–Pb age dates from Davies et al. (2017). High  
1182 Hg/TOC was expected to overlap with CAMP U–Pb ages, but elevated Hg persists for  
1183 much longer and does not begin coincident with the earliest known CAMP ages.

1184

1185 **Fig. 11.** Schematic displaying example ways LIP magmatism may impact the Hg cycle  
1186 with respect to measured Hg contents and Hg/TOC.  
1187

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Figure 1

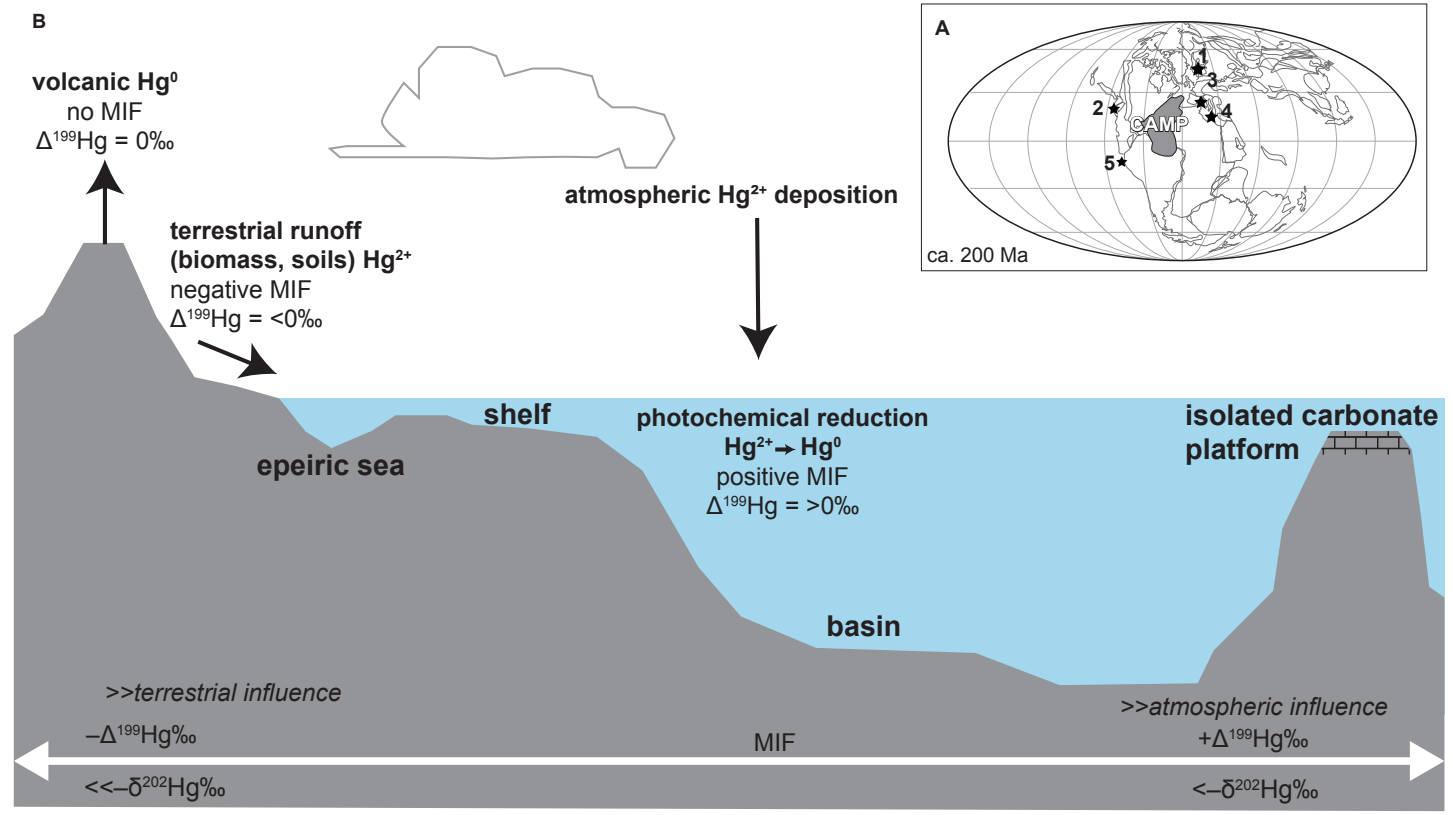
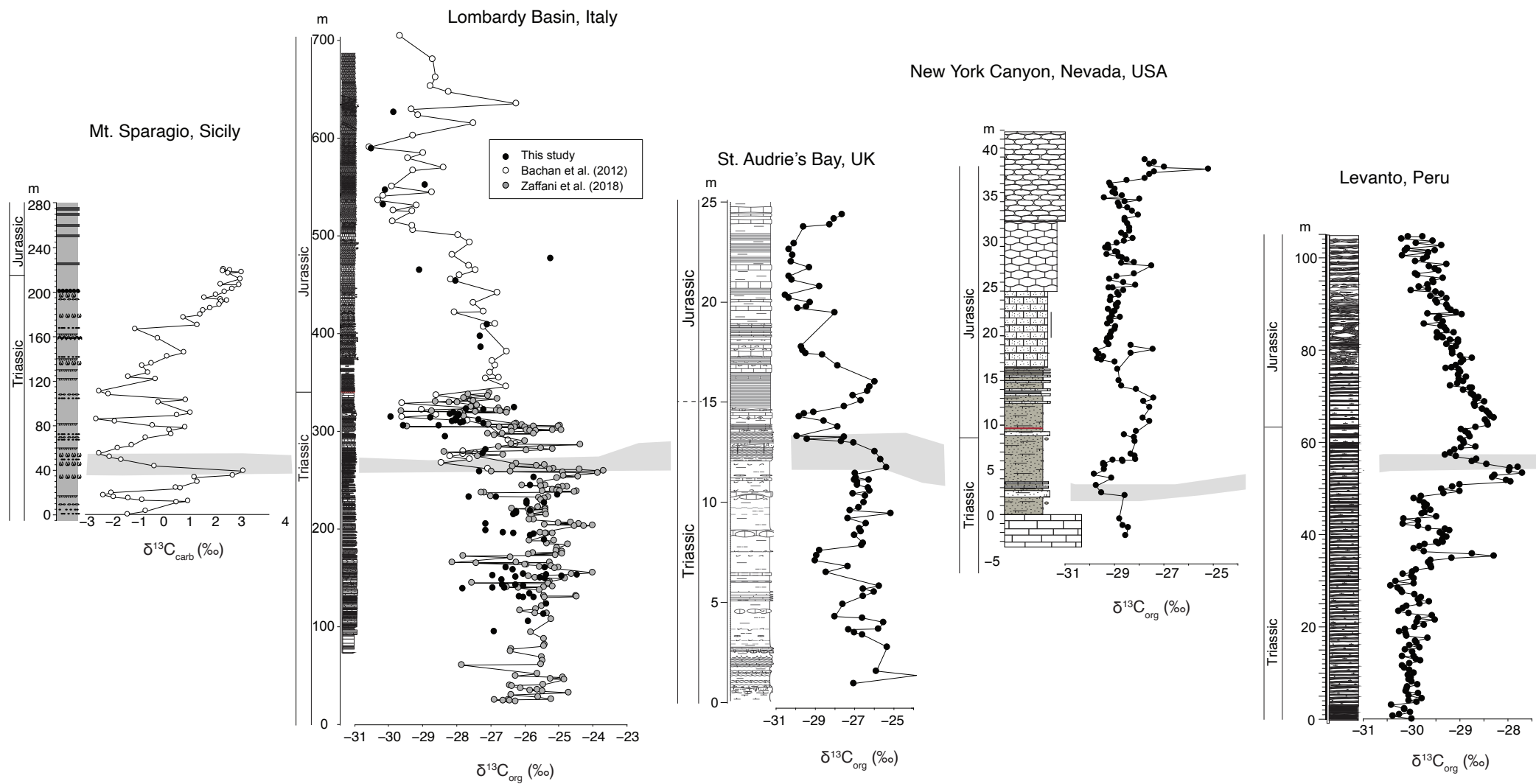
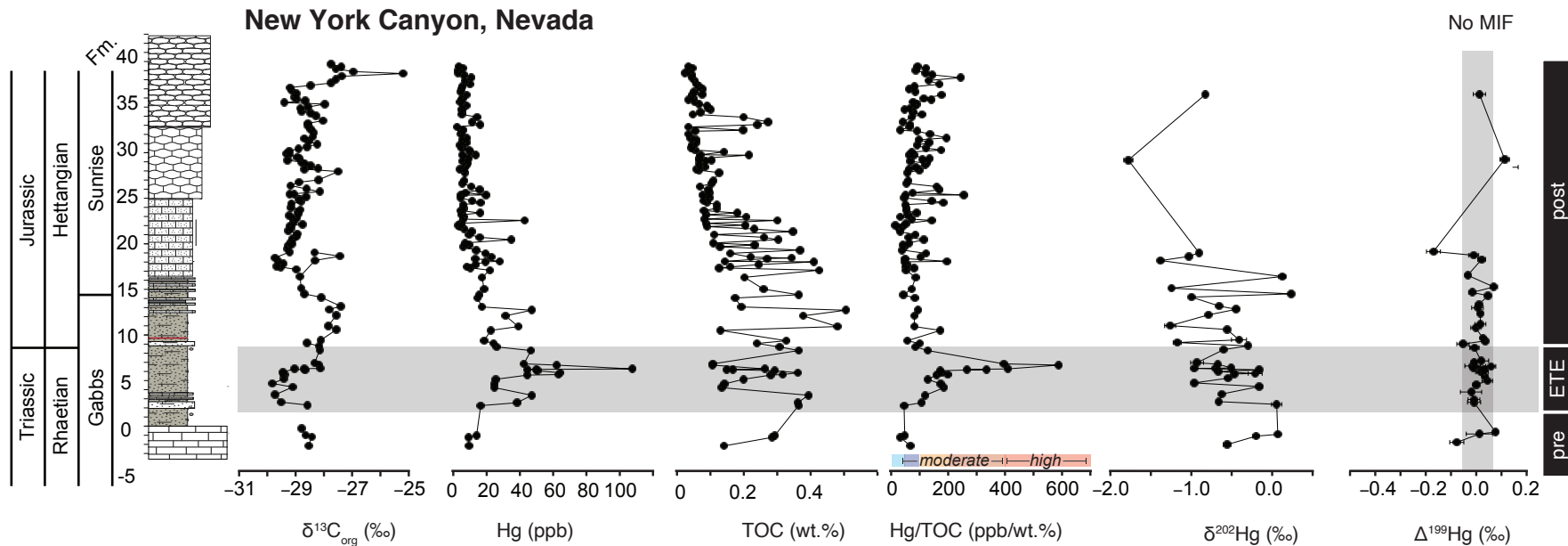




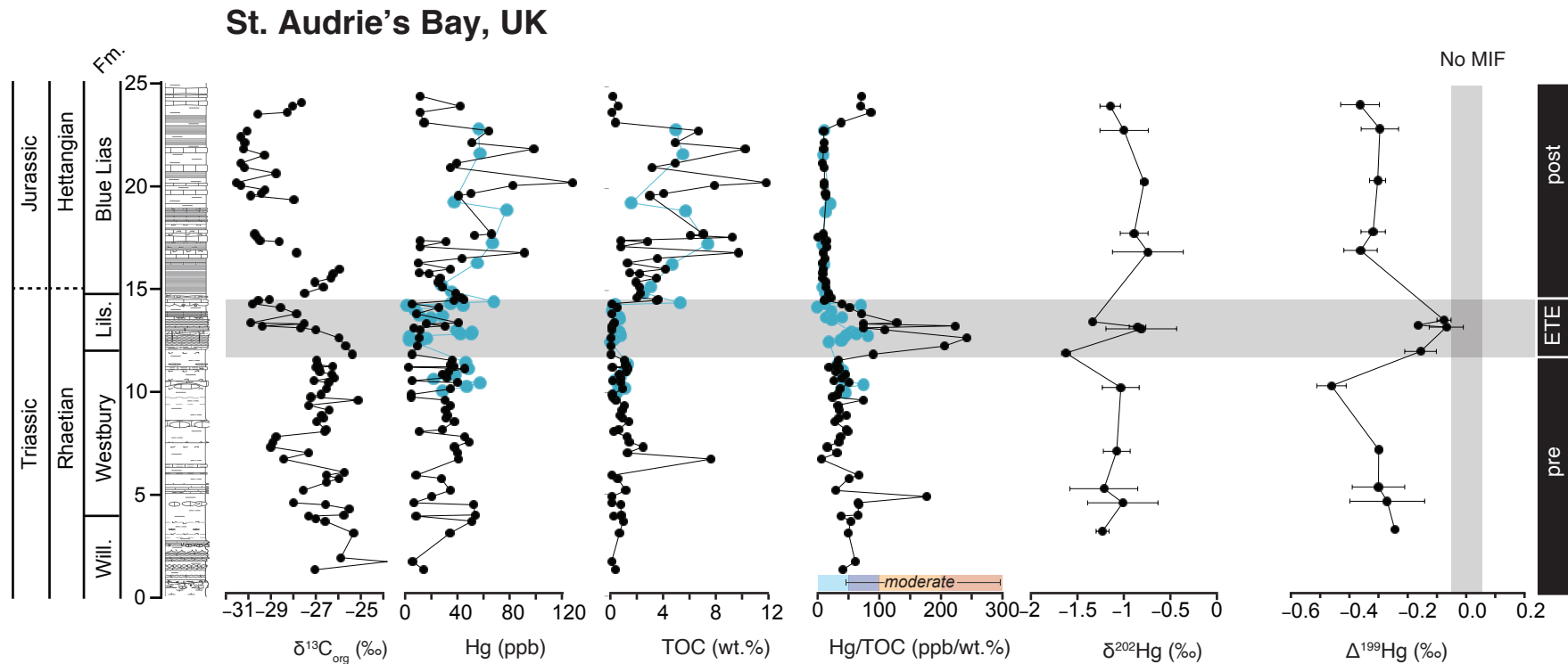
FIGURE 2



**FIGURE 3**

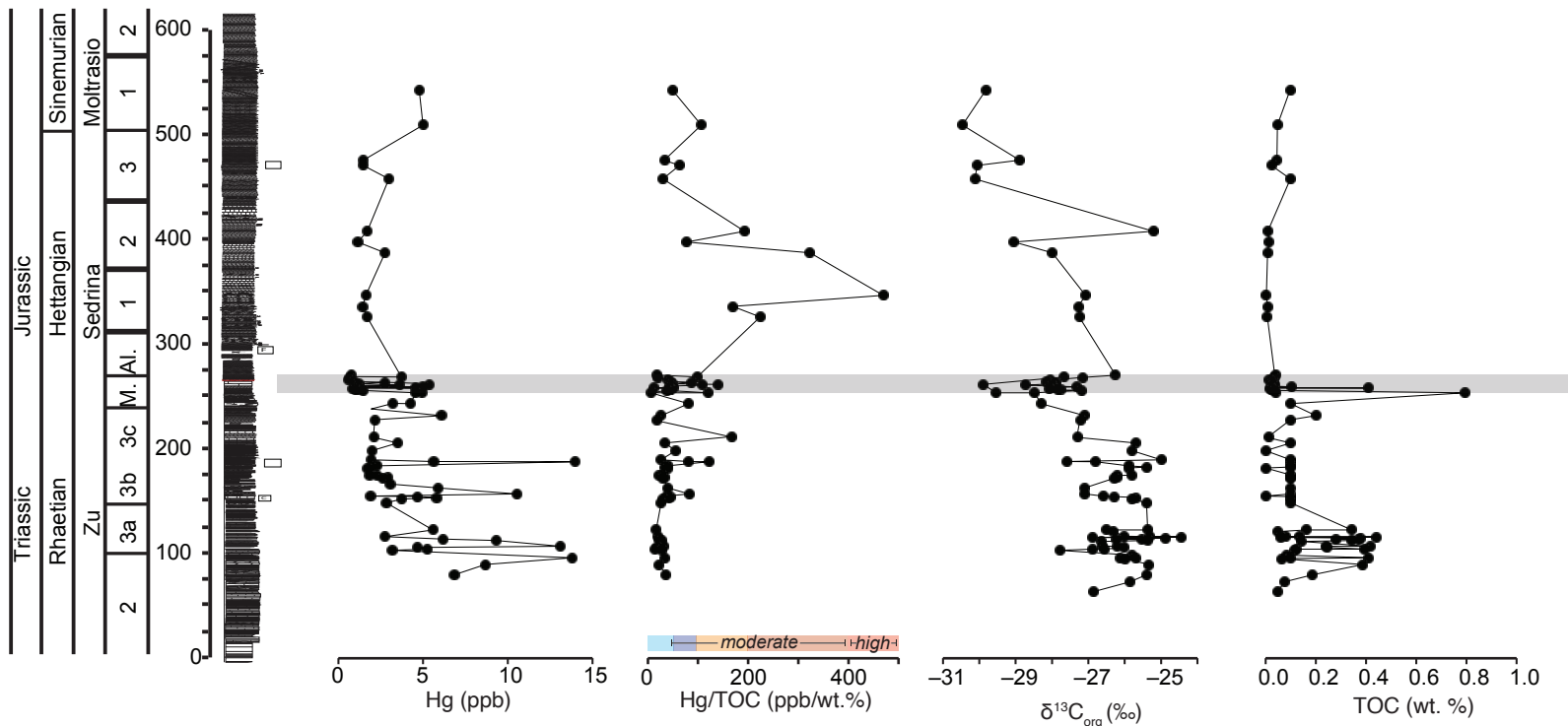


**FIGURE 4**



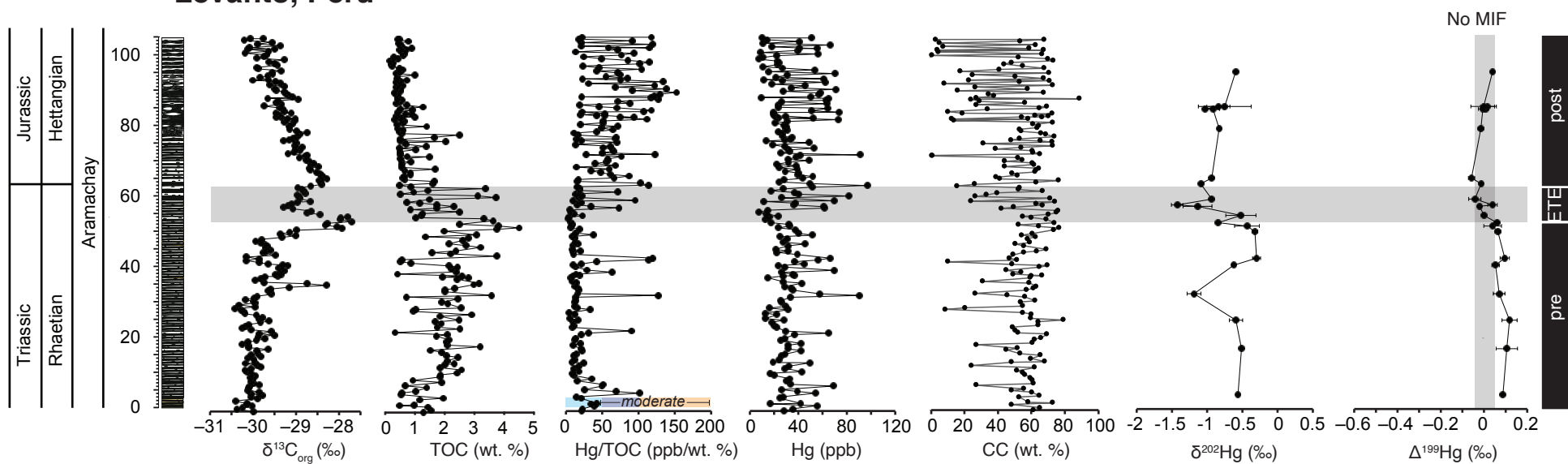
**FIGURE 5**

**Lombardy Basin, Italy (composite)**



**FIGURE 6**

**Levanto, Peru**



**FIGURE 7**

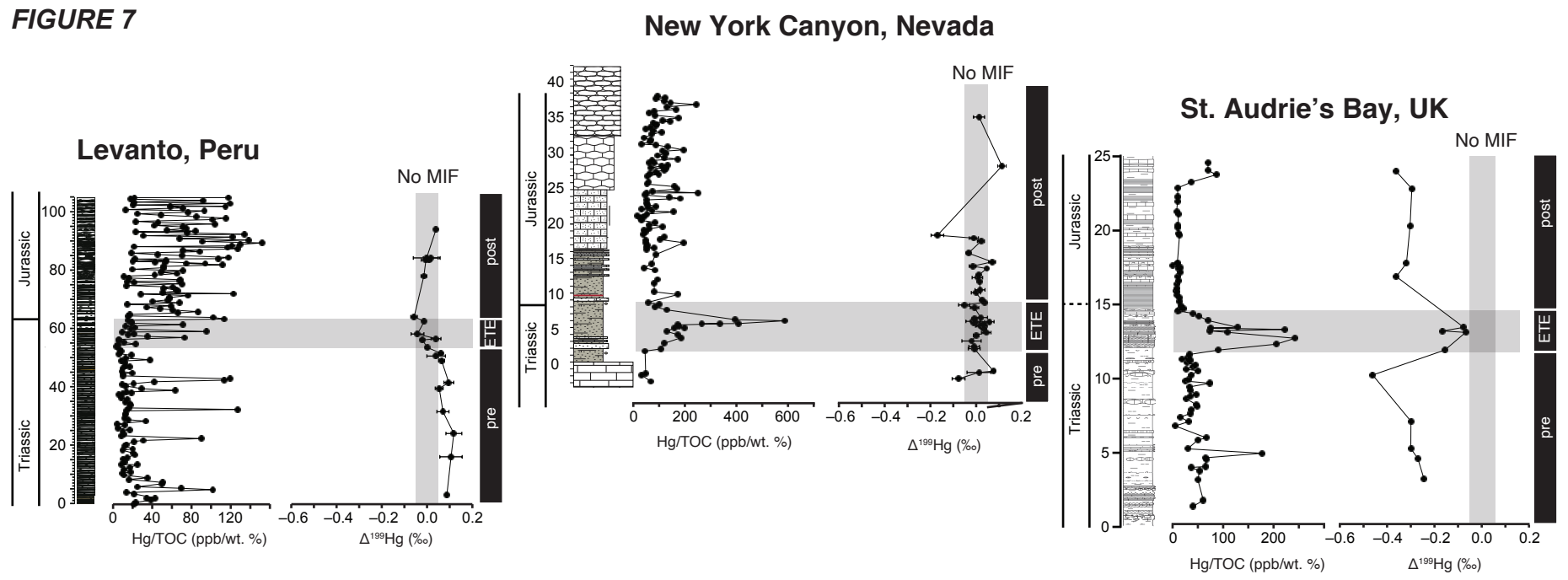
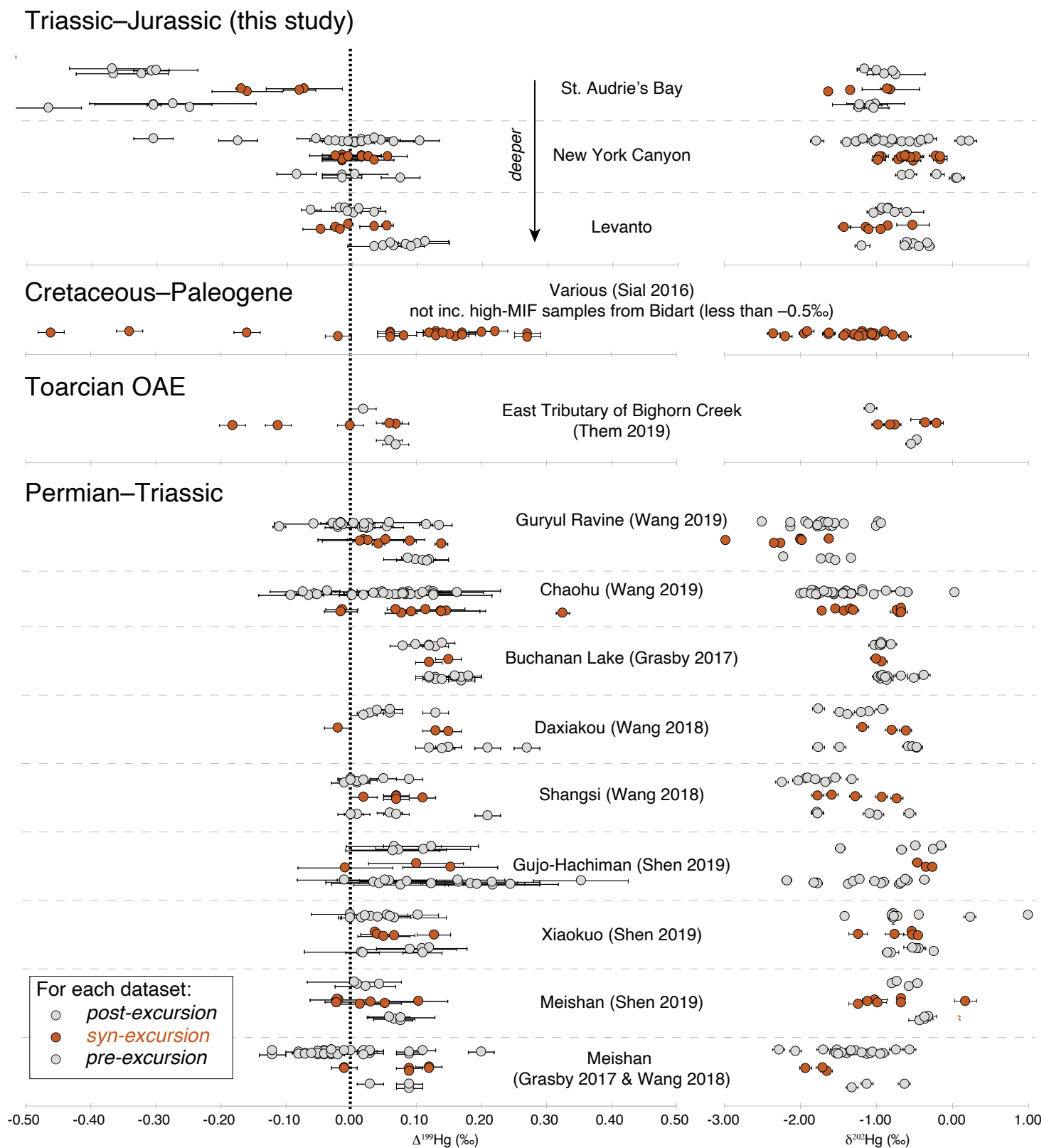


FIGURE 8



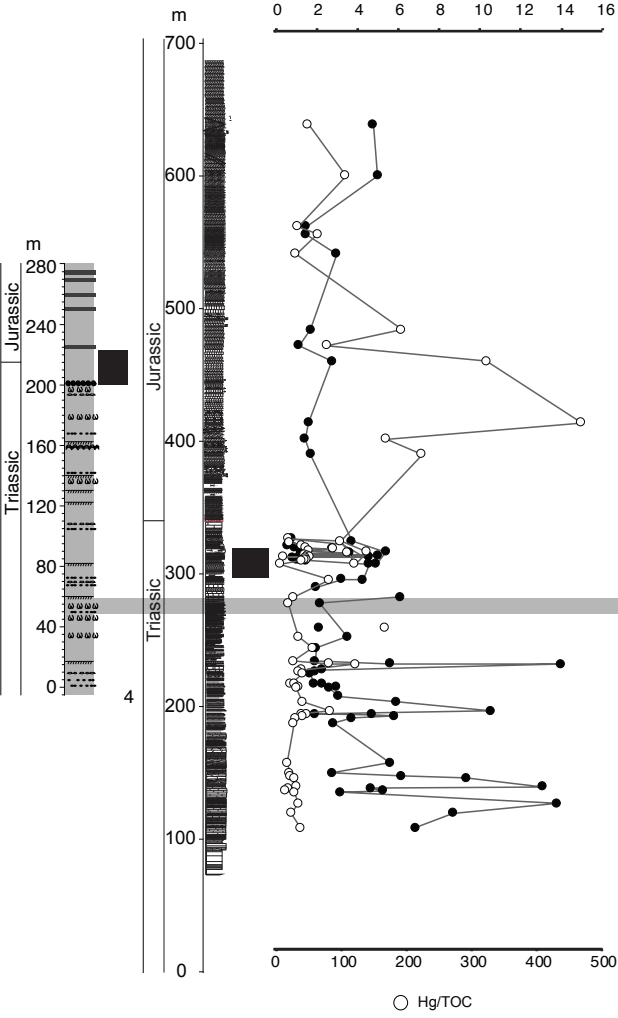
**FIGURE 9**

**Lombardy Basin, Italy**

**Mt. Sparagio, Sicily**

● Hg ppb

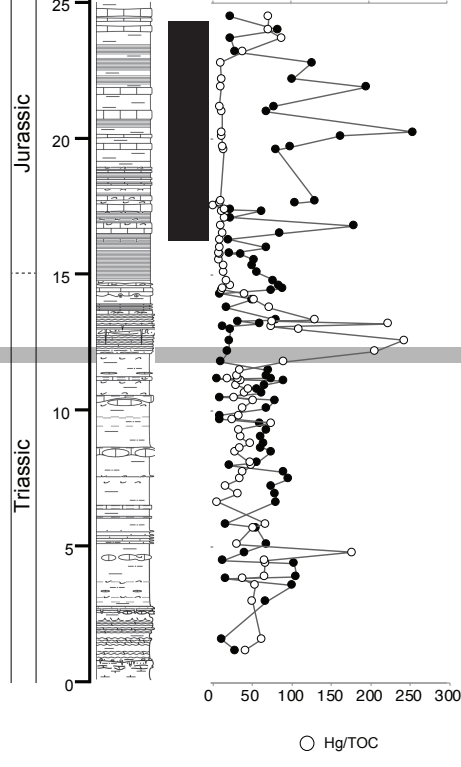
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**St. Audrie's Bay, UK**

● Hg ppb

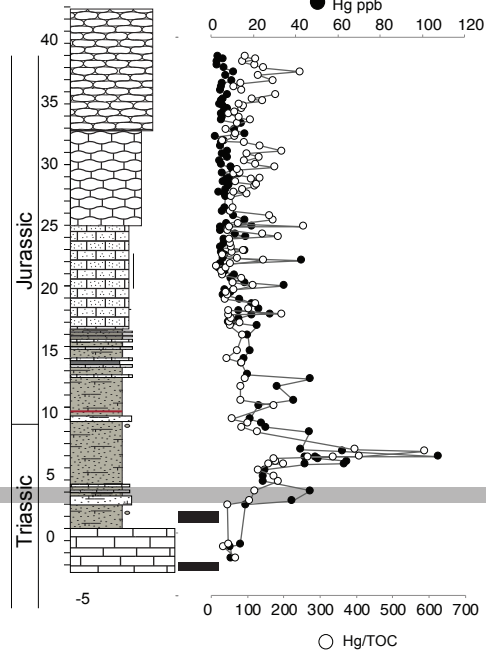
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**New York Canyon, Nevada, USA**

● Hg ppb

0 20 40 60 80 100 120



**Levanto, Peru**

● Hg ppb

0 20 40 60 80 100 120

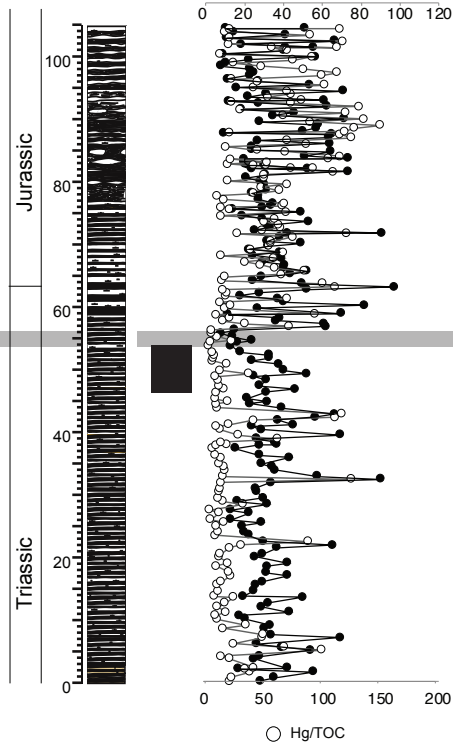
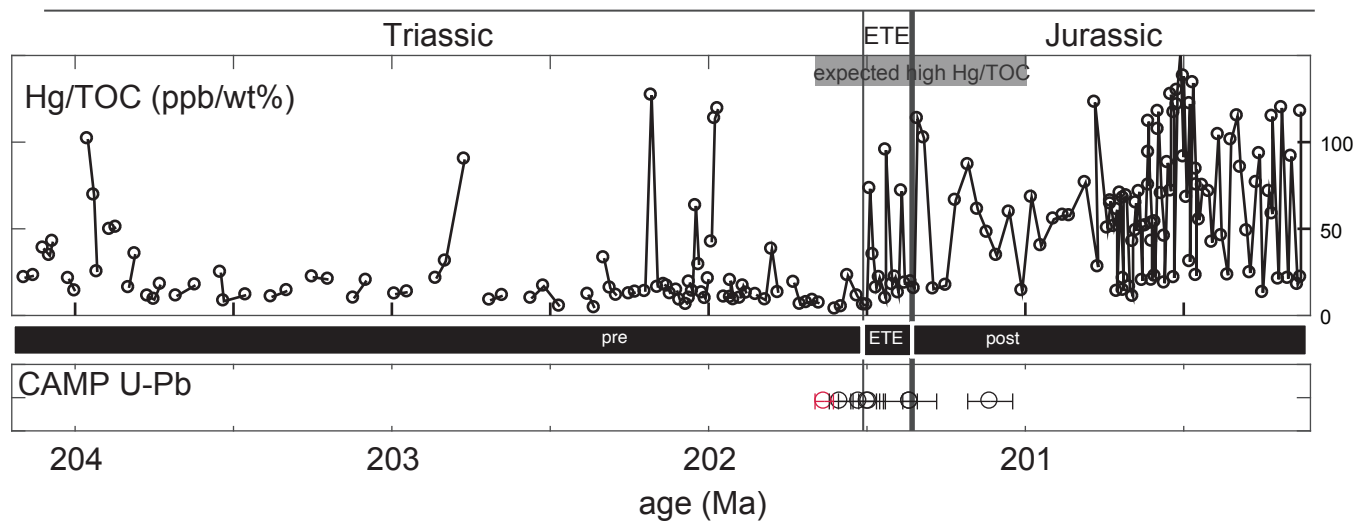


FIGURE 10



**FIGURE 11**

