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1 **Sources, transport, and accumulation of mercury in the northwestern**
2 **Mediterranean margin sediments during the Industrial Era**

3

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17

18

19 **Abstract**

20 Sources and pathways of the Hg accumulated in the sediments of the Gulf of Lion (GoL) and its
21 adjacent marine areas (Northwestern Mediterranean) have been explored using sediment grab
22 samples, sediment cores, and sediment trap samples. The main source of Hg along this margin is the
23 Rhône River, whose suspended sediments settle mainly in the prodelta area but also along the mid-
24 shelf, then reaching the continental rise *via* wave resuspension and cascading of dense shelf waters.
25 Seaward, these riverine particles are mixed with carbonated ooze conveyed to the bottom by the
26 biological pump. The Hg is enriched in surface sediments of the GoL with decreasing concentrations
27 westward and seaward from the Rhône prodelta to the continental rise. Dated cores from the Rhône
28 prodelta give access to riverine sediments deposited over the last 400 years and show that the Hg
29 concentration time trend resembles the evolution of coal consumption in France, peaking during the

30 AD 1960s decade. Similar trends were observed in sediment cores collected along the GoL slope and
31 submarine canyons along with the preservation of traces of erosion and sediment instability events.
32 Seaward, on the continental rise, the Hg concentration distribution suggests the deposition of fine
33 planktonic-derived material and particles episodically advected from the shelf during deep cascading
34 pulses. Anthropogenic Hg accumulated in GoL and continental rise sediments during the
35 Industrialized Era is ~150 Mg (tons), two-thirds of which are buried in the Rhône prodelta area.
36 Significant correlations are found between Hg and organic matter in GoL sediments, but the
37 relationships differ between areas and are disrupted by the inputs of anthropogenic Hg and by the Hg
38 availability in the Rhône River watershed during the pre-industrial period. The HgT availability in
39 waters appears to be the limiting factor to the Hg enrichment of the particulate organic matter.
40 Methylmercury (MMHg), which represents on average 0.3% of the total Hg, was positively
41 correlated to total Hg. Their distributions suggest *in situ* MMHg formation and a more effective net
42 Hg methylation for surface sediment or particles collected in traps compared with particles buried in
43 the sedimentary column.

44

45 **Keywords:** *mercury, sediment, prodelta, shelf, western Mediterranean*

46

47 **Highlights:**

- 48 1. Sources and pathways of the Hg accumulated in the sediments of the Gulf of Lion (GoL) and
49 its adjacent marine areas (Northwestern Mediterranean) have been explored
- 50 2. Anthropogenic Hg accumulated in the GoL sediments during the Industrial Era is ca. 150 Mg,
51 mainly from the Rhône River
- 52 3. In surface sediment, the Hg enrichment factor decreases from 10.7 in the Rhône prodelta to
53 1.5 in the continental rise
- 54 4. The chronological trend of Hg is similar to the coal consumption peaking AD 1962
- 55 5. The monomethyl mercury abundance is a function of the total Hg content with proportions
56 decreasing with increasing sediment depth

57

58 1. Introduction

59 Sediments of continental margins are major geochemical sinks for natural and anthropogenic
60 mercury (Hg) (e.g., Cossa et al., 1996; Fitzgerald et al., 2007; Mason et al., 2012). On the global
61 scale, the Hg accumulated in continental margin sediments is estimated to be $\sim 200 \text{ Mg a}^{-1}$ which is
62 one-third of the Hg deep sediment burial (Outridge et al., 2018). From very early on, concentration
63 profiles in sedimentary deposits have been used to track historical Hg contamination of coastal areas.
64 Many chronological records faithfully reflect Hg depositional fluxes during the Industrial Era and
65 even during the entire historical period (Bothner et al., 1980; Smith and Loring, 1981; Asmund and
66 Nielsen, 2000; Johannessen et al., 2005; Elbaz-Poulichet et al., 2011; Boutier et al., 2012; Oliveri et
67 al., 2016). This manner of reconstructing historical Hg deposition is based on the assumption that the
68 sediment does not undergo too extensive physical or biological mixing and that Hg is not translocated
69 during the early diagenesis of organic matter (OM). In the coastal areas, where accumulation rates are
70 high, these conditions are thought to be fulfilled (Gobeil and Cossa, 1993; Cossa et al., 1996;
71 Fitzgerald et al., 2018; Cooke et al., 2020). However, a good knowledge of depositional conditions,
72 including sedimentation rates (SRs) and biological mixing, is required to establish a realistic
73 chronology, to sort out the anthropogenic Hg fraction from the “natural” one, and ultimately quantify
74 the Hg accumulation over the years.

75 In the northwestern Mediterranean continental margin, the Gulf of Lion (GoL), comprising a
76 large continental shelf, slope, and rise, densely incised by submarine canyons, is under the influence
77 of natural and anthropogenic Hg inputs from atmospheric and riverine sources, especially from the
78 Rhône River (RR), the largest river discharging in the western Mediterranean (Mermex Group, 2011;
79 Cossa et al., 2022). The present article aims to provide the inventory of Hg accumulation, establish a
80 chronology of the Hg deposition for the last 400 years, and identify the main sources and transport
81 paths. Our specific objectives are (i) providing the geographical distribution of Hg concentrations in
82 surface sediment of the various regions of the GoL, (ii) establishing the chronology of the Hg
83 deposition in the last 400 years, (iii) identifying the main Hg sources and main transport modes, and
84 (iv) quantifying the anthropogenic Hg accumulation in GoL sediments during the Industrial Era (post
85 AD 1850). For that purpose, we have analyzed material collected in sediment traps, surface sediment
86 samples, including coastal lagoons, and dated sediment cores distributed from the Rhône prodelta to

87 the continental rise and in submarine canyons. The interpretation of our results has benefited from the
88 contribution of numerous studies which cover the geological context, sedimentary characteristics, and
89 anthropogenic influences on the study area (e.g., Zuo et al., 1991, 1997; Miralles et al., 2005, 2006;
90 Rabineau et al., 2005; Roussiez et al., 2006; Cathalot et al., 2010; Canals et al., 2013; Fanget et al.,
91 2013; Cossa et al., 2017, 2018a; Nizou et al., 2019; Dennielou et al., 2019; Estournel et al., 2023;
92 Durrieu de Madron et al., 2020, 2023).

93

94 **2. Study site**

95 The GoL is characterized by a wide shelf and a steep continental (gradients up to 10° at the upper
96 slope to 2° at 1800 m) dissected by a network of submarine canyons down to 2000 m (Berné and
97 Gorini, 2005; Dennielou et al., 2009) (Fig. 1). Sedimentary materials delivered to the GoL derive
98 mainly from riverine sources (the RR and several smaller coastal rivers), but also atmospheric
99 deposition (Cathalot et al., 2010; Marion et al., 2010; Zebracki et al., 2015; Wu et al. 2018). The RR
100 inputs, which contribute to more than 90 % of the total river-borne particles discharging into the GoL
101 (Bourrin et al., 2006), dominate the deposition on the shelf, upper continental slope, and canyons
102 (Aloïsi et al., 1979; Zuo et al., 1997), whereas the contribution from the atmosphere (mostly driven
103 by the biological pump) increases seaward, to become nowadays the dominant particle source on the
104 lower continental slope and rise (Durrieu de Madron et al., 2000). The SRs vary within three orders
105 of magnitude, from 0.2 mm a^{-1} at the continental rise (Zuo et al., 1991) up to 60 cm a^{-1} in the
106 nearshore part of the RR mouth, called the Roustan lobe, which is the most recent prodelta of the
107 Rhône (Charmasson et al., 1998; Touzani and Giresse, 2002; Beaudouin et al., 2005; Vella et al.,
108 2005; Fanget et al., 2013).

109 Within the Roustan lobe, the RR particle transport was estimated to be $\sim 9.6 \cdot 10^6 \text{ Mg a}^{-1}$
110 (Antonelli and Provansal, 2002) and the accumulation to be $\sim 5.8 \cdot 10^6 \text{ Mg a}^{-1}$ (a value calculated from
111 the accumulation rate expressed in $\text{m}^3 \text{ a}^{-1}$ by Sabatier et al. (2006) multiplied by a mean density of 1.7),
112 whereas the total accumulation of sediment in the GoL sediment margin is estimated to $10 \pm 4 \cdot 10^6 \text{ Mg}$
113 a^{-1} (Zuo et al., 1997). These figures mean that all the RR particulate discharge settles down in our study
114 area and that $\sim 60\%$ of it is deposited within 5 km of the river mouth. It has been suggested earlier that
115 this percentage would be close to 75%, and that two-thirds of the deposited sediments are more or less
116 resuspended depending on the water depth (Aloisi et al., 1984; Radakovitch et al., 1999; Touzani and

117 Giresse, 2002; Estournel et al., 2023). The intense reworking of sedimentary material was quantified by
118 the thickness of the surface mixed layer, which is > 30 cm in the proximal prodelta (Zuo et al, 1997).
119 The Rhône prodeltaic sediments consist of laminated organic-rich pelitic deposits, which show
120 alternations of more- sandy and more-carbonaceous layers. In these layers, seasonal and flood
121 signatures are recorded, with organic-rich layers characterized by high terrigenous inputs associated
122 with pollen grains typical of the RR watershed vegetation (Beaudouin et al., 2005; Fanget et al., 2013).
123 According to the same authors, flood events create distinct sedimentary deposits, which are
124 superimposed to long-term trends in accumulated sediments, including climatic and man-made changes
125 in RR (channelization, damming, reforestation, etc.).

126 On the GoL shelf, storms and dense shelf-water cascading events prevent long-term sediment
127 deposition (Guillen et al., 2006; Durrieu de Madron et al., 2008; Durrieu de Madron et al., 2023). The
128 general westward water mass circulation directs most of the export of suspended sediment towards
129 the southwestern exit of the GoL through re-suspension and deposition cycles along the inner shelf
130 induced by waves and an anti-clockwise circulation (Millot, 1991; Palanques et al., 2006; Durrieu de
131 Madron et al., 2008). This results in distinct depositional and erosional zones at the annual scale
132 (Estournel et al., 2023) and net westward transport of the sediment toward the shelf edge of the
133 westernmost submarine canyons. A part of the sediments entering the GoL canyons forms temporary
134 deposits in their upper/middle reaches, which trap particulate OM and associated trace elements
135 (Buscail et al., 1997). They are periodically resuspended during intense dense shelf-water cascading
136 events and flushed to deeper canyon reaches and further down to the continental rise, especially
137 through the Cap de Creus (CdC) Canyon (De Geest et al., 2008; Puig et al., 2008; Palanques et al.,
138 2012; Stabholz et al., 2013). Thus, the CdC Canyon is the largest outlet of sediment temporarily
139 trapped on the GoL shelf, and consequently of uppermost importance for assessing the contamination
140 transfer from the industrialized Rhône valley to the northwestern Mediterranean deep ecosystems.
141 Nearby, the Bourcart Canyon (BC, Fig. 1) is a less active canyon infilled by sea-level fall and Last
142 Glacial Maximum-derived sediments (Gaudin et al., 2006).

143 On the outer shelf, upper slope, interfluves, and the continental rise, sediments have
144 accumulated during the last sea-level fall and ensuing stillstand. Coarse-grained bioclastic carbonates
145 have accumulated on the outer shelf (e.g., Rabineau et al., 2005) and submarine canyon heads
146 (Gaudin et al., 2006), fine-grained prodeltaic mud on the upper slope, and the canyon interfluves
147 (e.g., Tesson et al., 1990), and detrital lithogenic sediments at the base of the slope (e.g., Droz et al.,

148 2006). Beyond about 1700 m water depth, the continental rise is mainly occupied by two large
149 sediment bodies, the Pyreneo-Catalan Sediment Ridge and the Rhone Deep Sea Fan. They consist of
150 heterogeneous material mainly deposited during low stands of the sea through gravitational processes
151 (turbidites and mass transport deposits) (e.g., Droz et al., 2001). Acoustic images of the sea-floor and
152 shallow cores show that some turbiditic activity persisted during the Holocene (last 10 ka), as well as
153 some erosion (Denniellou et al., 2006; 2009; Lombo Tombo et al., 2015). Seaward at the canyon
154 mouths and lobe areas, sediments are characterized by superficial heterogeneous sandy material
155 intercalated in hemipelagic carbonate ooze, although in some regions, over-consolidated muds are
156 present (e.g., Droz et al., 2001; Dennielou et al. 2009). These sedimentary facies are not documented
157 in terms of anthropogenic imprints. Recent modeling of sediment transport on the GoL shelf and
158 slope confirms the current knowledge of the pluri-annual characteristics of the shelf sediment
159 dynamics which includes the massive storage of sediment at the RR mouth, the accretion of the mid-
160 shelf mud belt, the sediment transfer through the CdC Canyon, and the magnitude of sediment
161 accumulations (Estournel et al., 2023).

162

163 **3. Methods**

164 **3.1. Sampling**

165 Twenty-one sediment cores were collected in the RR prodelta, continental shelf, CdC Canyon and
166 BC canyons, and the continental rise (Fig. 1) using various interface- and box-corers. In addition, we
167 used a Kullenberg piston corer to have access to longer cores. Information on sampling dates,
168 locations, water depths, and lengths of the cores are given in Supplementary Information (Table
169 SI.1). The cores were sliced every 0.5 or 2 cm from the surface to 100 cm, then every 4 cm down to
170 the bottom of the cores. Subsamples were frozen (-18°C), freeze-dried, and stored under cold (+4°C)
171 and dark conditions until analysis. Additional 196 surface sediments (0-2 cm) were collected with an
172 Ekman grab sampler in the nearshore areas as a part of the French monitoring network database
173 (RNO/ROCCH website) and were subjected to similar treatments before analyses.

174 Time series of settling particles were collected along the GoL continental rise (Stas. I and L) at
175 20 m above the bottom and at the head of the CdC Canyon (Sta. HD) at 30 m above the bottom (Fig.
176 1) using Technicap® sediment traps PPS53 and PPS3 models, respectively. The distance from the
177 bottom was chosen to reduce the flux of particles coming from local resuspension, which could

178 interfere with the signal (i.e., composition) of downward particles settling through the water column.
179 Nine samples were collected in traps I and L in spring 2008, and 25 in trap HD from autumn 2004 to
180 spring 2005. Station coordinates, water depth, and details about the mooring are given in
181 Supplementary Information (Table SI.2). Sediment trap cups were filled with buffered Hg-free
182 formaldehyde; the collected material was stored at +4 °C after sieving through a 1 mm nylon mesh to
183 retain the large swimming organisms that occasionally enter the traps during sampling. It was then
184 precisely divided into sub-samples for subsequent analyses using a wet sample divider (WSD-10,
185 McLane®). A subsample from each trap was freeze-dried and stored in the dark until analysis.

186 **3.2. Dating and age models**

187 The dating of cores has been established based on ^{210}Pb and ^{137}Cs vertical profiles in combination
188 with sedimentological proxies (Fanget et al., 2013). These authors also used one ^{14}C date for the
189 bottom of core RHS-KS57. Sedimentation rates have been calculated using both constant flux-
190 constant sedimentation (CFCS) and constant initial concentration (CIC) models (Appleby and
191 Oldfield, 1978) based on ^{210}Pb activities measured according to Radakovitch and Heussner (1999).
192 Published SR values are presented in Table SI.3 and unpublished data are detailed in SI.1, with the
193 periods covered by each core.

194 **3.3. Chemical analyses**

195 Total carbon (C_t), organic carbon (C_{org}), and total nitrogen (N_t) were determined using freeze-dried
196 and homogenized subsamples of sediments with an elemental analyzer (Model CN 2000, LECO®)
197 after acidification with 2 M HCl (overnight, at 50°C) for C_{org} to remove carbonates (Cauwet et al.,
198 1990). The precision (6 replicates of a sample) for C_{org} and C_t analyses was 2 %, and 0.3 % for N_t .
199 Concentrations are expressed as the weight percent of dry sediment (% dry weight). Calcium
200 carbonate content was calculated from mineral carbon ($C_t - C_{\text{org}}$) using the molecular mass ratio
201 $\text{CaCO}_3:\text{C} = 100:12$.

202 Sub-samples for total Hg (HgT) determinations were freeze-dried; sediment aliquots (~50-100
203 mg) were weighed in Ni boats and loaded in a semi-automatic Hg analyzer (AMA-254, Altec®)
204 according to Guédron et al. (2011). Within this instrument, the samples are first heated in a furnace at
205 550 °C under an atmosphere enriched in O_2 and the produced elemental Hg vapor is trapped onto a
206 gold amalgamation device. This Hg is subsequently thermally (600 °C) released from the trap as an
207 instant pulse and quantitatively measured by atomic absorption spectrophotometry. Our analytical

208 precision, appraised from replicate measurements ($n = 16$) of the certified reference material (CRM)
209 MESS-3, a marine sediment certified for total metal content from the National Research Council of
210 Canada ($0.091 \pm 0.009 \mu\text{g g}^{-1}$), was 8 %; the accuracy, expressed in terms of the recovery rate of the
211 CRM, was 109 %, and the detection limit, defined as 3 times the standard deviation of 6 blanks, was
212 $0.007 \mu\text{g g}^{-1}$. We report the HgT concentrations in the sediment samples on a dry weight basis after
213 correction for the salt content of the sediment approximated from the measured porosity value of
214 each sample and the water column salinity.

215 Monomethyl Hg (MMHg) determinations were performed by isotopic dilution, derivatization
216 (propylation), and gas chromatography-inductively coupled plasma mass spectrometry (GC-ICP-MS)
217 (Monperrus et al., 2005). This method was slightly modified by Abi-Ghanem et al. (2011) for marine
218 sediments. A known quantity of MM^{202}Hg (the CRM ERM-AE670 from the European Commission)
219 was added to the sediment aliquot ($\sim 300\text{mg}$). The MMHg was extracted using 6M HNO_3 (Suprapur,
220 Merck®), and propylated with 100 μL of 4% solution of sodium tetrapropylborate (Galab®). The
221 propylated MMHg was then extracted into 300 μL of iso-octane and the detection was carried out by
222 GC-ICP-MS (GC-Focuswith X-series, Thermo Electron®). The detection limit (3 times the standard
223 deviation of 6 blank values) was better than 15 %. The accuracy, expressed in terms of the recovery
224 rate of the CRM IAEA-405 ($5.49 \pm 0.30 \text{ ng g}^{-1}$), an estuarine sediment certified for MMHg, was 104
225 % and the precision (6 replicates of the CRM) was 0.02 ng g^{-1} . Aluminum (Al) concentrations were
226 determined by atomic absorption spectrophotometry (SpectrAA 600, Varian) after the total
227 dissolution of the sediment with a mixture of HCl, HNO_3 , and HF in hermetically sealed Teflon
228 (PFA) reactors according to the protocol described by Chiffoleau et al. (2004).

229 **3.4. Mercury inventories**

230 The calculation of the anthropogenic Hg (Hg_{anthr}) accumulated in the GoL sediments during the
231 Industrial Era (i.e., post AD 1850) has been performed by summing for each core the amount of
232 Hg_{anthr} calculated in each sedimentary layer for which we obtained Hg concentrations, then by
233 combining the results obtained for each of individual regions (proximal and distal deltas, inner and
234 outer shelves, CdC Canyon, and continental rise). The Hg_{anthr} concentrations were calculated as the
235 difference between measured Hg (HgT) and the pre-industrial Hg concentration in the same core
236 (Table SI.4). The areas of each region are taken from the scientific literature (Estournel et al., 2023),
237 and the proximal prodelta area is defined by an envelope of 3 km on either side of the mouth of the

238 Rhône River between the isobaths 10 and 40 m, whereas the distal part by an envelope between 40
 239 and 100m. The Hg_{anthr} inventories for each core are calculated as

$$240 \quad \sum_{k=0}^n \binom{n}{k} \rho [Hg_{anthr}] V (1 - \beta)$$

241 with ρ the density, β the porosity, V the volume of the layer, and $[Hg_{anthr}]$ the concentration of the
 242 anthropogenic (post-AD 1850) Hg in the layer, n being the number of layers in the core. Hg
 243 background is the pre-industrial Hg concentration.

244

245 **4. Results**

246 **4.1. Mercury in surface sediments**

247 *4.1.1 Lagoons and inner-shelf areas*

248 The results presented in this section concern the surface sediment (0-2 cm) collected within the near-
 249 shore zone by depth < 50 m all along the GoL coasts from the Spanish-French border to the Cap
 250 Sicié, West of Toulon (Suppl. Info. Fig. SI.1) and consist of 196 sediment grab samples that include
 251 harbors and lagoons. The HgT concentrations ranged from low (< 0.01 $\mu g g^{-1}$) to very high values
 252 (up to 4.90 $\mu g g^{-1}$), consistent with the nature of the bottom which includes both erosion and
 253 accumulation zones, with pristine and human-impacted levels, respectively (Fig. SI.1). Seventy-five
 254 percent of the concentrations ranged from 0.01 to 0.20 $\mu g g^{-1}$ with a mean of $0.08 \pm 0.02 \mu g g^{-1}$ which
 255 is comparable with the Hg levels found in other near-shore areas of the Mediterranean (e.g.,
 256 Barghigiani et al., 1996; Di Leonardo et al., 2006). The highest concentrations were found in the
 257 harbor area of Marseille.

258 *4.1.2. Rhône prodelta, shelf, slope, and continental rise*

259 The means of HgT concentrations of the surface sediments of the proximal and distal parts of the Rhône
 260 Prodelt, the shelf, and the continental rise ranged from 0.06 to 0.29 $\mu g g^{-1}$ (Table 1). These
 261 concentration levels were always higher than the levels measured at the base of the cores (data shown
 262 below) and attest to Hg enrichments. The highest mean HgT concentration was found in the Rhône
 263 prodelta region and the lowest on the shelf break and the continental rise. The sediments of the
 264 continental slope, including CdC Canyon, present intermediate values. Our results from CdC Canyon

265 surface sediments (Table 1) are in the range of the HgT concentration published earlier for the same
 266 canyon and in its southern open slope, i.e., $0.071 \pm 0.010 \mu\text{g g}^{-1}$ (Durrieu de Madron et al., 2020).

267 **Table 1.** Surface sediments (0-1 cm). Organic carbon, C/N, Ca, Al, and HgT (mean \pm 1 standard deviation,
 268 number of determinations in brackets). Rhône prodelta (RD_P: proximal part includes ME, 62C, and Stas. 1-4;
 269 RD_D: distal part includes 61D, 94-21, and Sta. 5), Gulf of Lion shelf (SH_E: east part includes Sta. 6, PR12,
 270 PR14, PR15, and PR18, 136, 95-19, TYR12; SH_W: west part includes Stas. 7-16, PR2-4, PR10-11), head of the
 271 Cap de Creus Canyon (HD includes Stas. 17-23), head of the Bourcart Canyon (STKI19), Cap de Creus
 272 Canyon (cores G and H), continental slope (KIGC25 and TYR20), and continental rise (cores I, L, and
 273 KIGC1&2).

Core/ Region	C _{org} (% d.w.)	C/N (at. Ratio)	Ca (%)	Al (%)	HgT ($\mu\text{g g}^{-1}$)
Rhône prodelta					
RD _P	1.71 ± 0.14 (6)	10.9 ± 0.5 (4)	11.9 ± 2.9 (4)	5.32 ± 0.18 (5)	0.160 ± 0.032 (6)
RD _D	1.56 ± 0.37 (3)	11.6 ± 0.6 (3)	-	5.53 ± 0.32 (3)	0.289 ± 0.076 (3)
Gulf of Lion shelf					
SH _E	1.06 ± 0.15 (7)	9.9 ± 0.6 (7)	-	5.47 ± 0.52 (8)	0.171 ± 0.043 (8)
SH _W	0.89 ± 0.36 (15)	9.0 ± 1.0 (10)	9.3 ± 3.4 (10)	5.37 ± 0.59 (10)	0.075 ± 0.037 (15)
Head of canyons					
HD	0.60 ± 0.26 (7)	8.9 ± 1.1 (7)	13.7 ± 3.4 (3)	5.06 ± 1.07 (7)	0.060 ± 0.036 (7)
STKI19	-	-	-	-	0.055 ± 0.007 (2)
Cap de Creus Canyon					
G	0.71 ± 0.07 (2)	9.7 ± 0.5 (2)	10.8 ± 0.1 (2)	6.40 ± 0.20 (2)	0.104 ± 0.008 (2)
H	0.74 ± 0.03 (2)	9.6 ± 0.4 (2)	11.1 ± 0.1 (2)	6.11 ± 0.05 (2)	0.102 ± 0.005 (2)
Continental slope					
KIGC25	-	-	-	-	0.062 ± 0.002 (2)
TYR20	0.69 (1)	-	-	4.2 (1)	0.141 ± 0.011 (3)
Continental rise					
I	0.46 ± 0.02 (2)	10.0 ± 0.1 (2)	17.0 ± 1.9 (2)	4.53 ± 0.68 (2)	0.056 ± 0.007 (2)
L	0.39 ± 0.02 (2)	11.2 ± 1.3 (2)	20.6 ± 0.6 (2)	3.60 ± 0.23 (2)	0.072 ± 0.011 (2)

KIGC2&5 - - - - 0.058 ± 0.002 (2)

274

275 **4.2. Mercury in sediment traps**

276 The HgT concentrations of samples collected in the sediment trap at the head of CdC Canyon (Trap
 277 HD, Fig. 1; Table SI.2) varied from 0.05 to 0.11 $\mu\text{g g}^{-1}$. The means of HgT, Al, Ca, C_{org}
 278 concentrations, and C:N atomic ratios are given in Table 2. Temporal variations of these parameters,
 279 the sand proportion, and $\delta^{13}\text{C}$ are illustrated in figure 2. From October 2004 to January 2005, HgT
 280 concentrations remained fairly constant but dropped down during March 2005, then concentrations
 281 increased to reach a level similar to the beginning of the sampling period. These variations are
 282 parallel to those of C_{org} and inverse to those of Ca, the proxy for carbonates, and inverse with the
 283 sand proportion (Fig. 2). In March, the increase in coarse material, low C_{org}, and high Ca
 284 concentrations suggest the input of carbonate decomposed material. On the contrary, from April 2005
 285 to the end of May, low C/N and high $\delta^{13}\text{C}$ values indicate the input of more recent biogenic marine
 286 particles (Fig. 2). On the continental rise (traps I and L), the time series were limited to 9 samples
 287 collected in spring 2008 (from 28 March to 6 June and from 4 April to 4 July for Stas. I and L,
 288 respectively). The particles collected were carbonate and OM-rich (Table 2), which suggests a
 289 biogenic origin, consistent with the increasing marine influence on the continental rise compared to
 290 the shelf area (see section 2); this biogenic marine material has HgT average concentrations
 291 significantly higher (t-test, $p < 0.01$) than at Sta. HD (Table 2).

292 **Table 2.** Sediment traps (moored 20 m above the bottom). Organic carbon, C/N, $\delta^{13}\text{C}$, Ca, Al, and HgT (mean
 293 ± 1 standard deviation, number of determinations in brackets). (*) The HgT mean differs from the two others
 294 (t-test, $p < 0.01$).

	C _{org} (%)	C/N _{at}	Ca (%)	HgT ($\mu\text{g g}^{-1}$)	HgT flux ($\mu\text{g m}^{-2} \text{a}^{-1}$)
HD	1.46 ± 0.09 (25)	9.3 ± 0.7 (25)	11.1 ± 1.39 (25)	0.089* ± 0.024 (25)	637
I	3.43 ± 0.71 (9)	7.4 ± 0.9 (9)	24.8 ± 3.0 (9)	0.123 ± 0.008 (9)	25
L	3.46 ± 0.68 (9)	7.8 ± 1.4 (9)	20.8 ± 2.9 (9)	0.120 ± 0.023 (9)	22

295

296 **4.3. Mercury in the sediment cores**

297 *4.3.1. Rhône prodelta (cores ME, RHS-KS22, RHS-KS57, 61C, 62C, and 94-21)*

298 In the RR prodelta, six cores were collected at depths from 20 m (ME) to 98 m (61C) (Fig. 1). The
299 locations, water depth, core length, type of corer, and date of sampling are indicated in Suppl. Info.
300 (Table SI.1). The SRs (Table SI.3) varied largely decreasing seaward from 20-60 cm a⁻¹ in the
301 proximal prodelta area to 0.2-0.6 cm a⁻¹ at 20 km away seaward (Zuo et al., 1997; Radakovitch et al.,
302 1999; Charmasson et al., 1998; Beaudouin et al., 2005; Fanget et al., 2013; Miralles et al., 2005). In
303 the 32-cm long ME, the Hg concentrations are quite homogeneous ranging from 0.19 to 0.27 µg g⁻¹
304 and averaging 0.23 ± 0.03 µg g⁻¹ (n = 13); this core located near the mouth of the RR gave access to
305 the sediments accumulated during the year before the collection in September 2011. The surfaces of
306 the piston cores RHS-KS22 and RHS-KS57 were perturbed during the sampling. The former was 835
307 cm long and the second 770 cm (Fig. 1). Core RHS-KS22, collected in the proximal prodelta at 0.65
308 km South of ME, by 42 m water depth, gave access to a time window of AD 1953-2008 (Fanget et
309 al., 2013). Core RHS-KS22 was 200 m seaward from another piston core (KTR05, 738 cm long)
310 collected in 1992 (Cambon et al., 1997), for which an SR of 17.5 cm a⁻¹ gives access to sediments
311 deposited after AD 1960 (Beaudouin et al., 2005). Based on the same assumptions and the ¹³⁷Cs
312 distribution, the SR of core RHS-KS22 is ca. 16 cm a⁻¹. The HgT concentrations in core RHS-KS22
313 vary widely from 0.08 to 0.77 µg g⁻¹ averaging 0.29 ± 0.15 µg g⁻¹ (n = 84). The general pattern of the
314 HgT concentration profile with depth in Core RHS-KS22 (Fig. 3a) indicates a steady increase with
315 increasing depth, superimposed by high-frequency variations depending on the grain size distribution
316 of the sediments. Low HgT concentrations occurred within the sandy layers especially for depths ca.
317 350 and 755 cm, whereas high HgT concentrations are related to high RR discharge periods (Suppl.
318 Info., Fig. SI.2). Core RHS-KS57, collected by 79 m depth 2.3 km south of RHS-KS22, allows
319 assessing the Hg deposition of the last four centuries (Fanget et al, 2014). The HgT concentrations in
320 core RHS-KS57 varied widely from 0.02 to 0.79 µg g⁻¹ (n = 200). The HgT profile date is illustrated
321 in Figures 3a and 4. Noteworthy is that the HgT concentrations started strongly to increase at the
322 beginning of the XXth Century and peaked during the AD 1960-70 decade. In cores 61C, 62C, and
323 94-12, HgT concentrations vary from 0.19 to 0.75 µg g⁻¹ (n = 59) and also peaked during the decade
324 AD 1960-70 (Fig. 3b).

325 *4.3.2. Gulf of Lion shelf (cores I36, 95-19, TYR-12, 95-21, TYR-23, 95-9A, KIGC25, TYR20)*

326 Six box-cores were collected along the GoL shelf, west of the RR, between Rhône prodelta and the
327 head of the CdC Canyon (Fig. 1). HgT concentrations converged to $0.03 \mu\text{g g}^{-1}$ in the deepest parts of
328 all the cores (Fig. 5a). In the upper part of the 6 cores, the most striking feature was the higher HgT
329 levels in cores close to the Rhône prodelta, namely cores I36, 95-19, and TYR12; they ranged
330 between 0.15 and $0.20 \mu\text{g g}^{-1}$, whereas more westward they were close to $0.10 \mu\text{g g}^{-1}$. In addition,
331 the maximum concentration values of HgT decreased steadily westward from $0.21 \mu\text{g g}^{-1}$ (I36,
332 TYR12), 0.12 - $0.17 \mu\text{g g}^{-1}$ (95-19, 95-21), to 0.08 - $0.10 \mu\text{g g}^{-1}$ (TYR23, 95-9A).

333 Located in interfluves of GoL submarine canyons, cores KIGC25 and TYR20 (Fig. 1) exhibited
334 profiles of contrasting HgT distributions (Fig. SI.3, Suppl. Info.). HgT distribution in core KIGC25
335 showed a homogeneous layer from surface sediment to 10 cm averaging $0.06 \pm 0.01 \mu\text{g g}^{-1}$ and below
336 a very smoothed signal decreasing down to $0.014 \mu\text{g g}^{-1}$ at 70 cm. On the other hand, the distribution
337 in the short core TYR20 exhibited a sharp decrease from 0.15 at the surface to $0.02 \mu\text{g g}^{-1}$ at 12 cm
338 depth. These patterns suggest a massive input of material in a short period for KIGC25 and a rather
339 slow, more regular, sediment accumulation for TYR20. No age models are available from these cores
340 and the patterns of the vertical distribution suggest very variable sedimentary conditions and
341 sediment sources on the interfluve.

342 4.3.3. *Canyons (cores G and H) and continental rise (cores I, L, and KIGC2 & 5, Fig. 6)*

343 In the CdC Canyon sediments HgT concentrations range from 0.02 to $0.12 \mu\text{g g}^{-1}$ and the two profiles
344 were similar (Fig. 5b), with increasing concentrations at the beginning of the XIXth Century and
345 maxima occurring between AD 1950 and 1980 (Fig. 5b). The surface sediments were homogenous
346 over the upper 3.5 cm, averaging $0.11 \pm 0.01 \mu\text{g g}^{-1}$ in the AD 1994-2002 period. On the GoL
347 continental rise (cores I and L), HgT concentrations ranged from $0.03 \mu\text{g g}^{-1}$ at the base of the cores
348 to $0.08 \mu\text{g g}^{-1}$ at the surface (Fig. 6) and their temporal evolutions were similar during the period AD
349 1800–1970, showing a steady increase with time. However, in the youngest deposits, HgT
350 concentrations diverged; they continued to increase toward the surface in Core L, whereas a sub-
351 surface minimum appeared on the Core I profile (Fig. 6). For the most distal collected cores (KIGC2
352 & 5, Fig. 1), and for which no age model was available, we observed high HgT contents in the first 8
353 cm below the surface followed by almost constant values ($0.03 \mu\text{g g}^{-1}$) down to the bottom of the
354 core (Fig. 6b).

355 4.4. *Monomethyl mercury*

356 MMHg concentrations have been determined in 7 sediments cores including those from the Rhône
357 prodelta (RHS-KS57, 61C, 62C), the GoL shelf (I36), the CdC Canyon (H and G), and the
358 continental rise (L). In addition, surface sediment from the GoL shelf and material collected in the
359 sediment traps I and L were also analyzed for their MMHg content. The overall average for the 370
360 samples analyzed (\pm one standard deviation) is $0.56 \pm 0.62 \text{ ng g}^{-1}$ (i.e., 0.3% of the HgT), ranging on
361 more than two orders of magnitude from 0.01 to 3.20 ng g^{-1} .

362

363 **5. Discussion**

364 *5.1. Tracing Hg: terrestrial vs marine sources and transport*

365 The geographical gradients of Hg concentrations highlight the sources of Hg-enriched particulate
366 material distributed all along the GoL sediments. Recent simulation using based on a
367 hydrosedimentary model (Estournel et al., 2023) shows the (i) massive sediment accumulation near
368 the Rhône prodelta, (ii) the net sediment advection westward through resuspension, (iii) the accretion
369 along the mid-shelf, and (iv) the impact of dense shelf water cascading on sediment resuspension and
370 erosion inside the Cap de Creus Canyon (see section 2). Here we show that the Hg sedimentary
371 distribution is consistent with the general pattern of the sediment dynamic in the GoL, and in
372 particular with the transfer of particles from the coast towards deeper parts of the margin during
373 dense shelf water formation events and their subsequent cascading (Durrieu de Madron et al., 2023).

374 *5.1.1. The Rhône River as the main Hg source*

375 The highest HgT concentrations have been measured in lagoons, in the inner shelf, harbors, and
376 especially in the Rhône prodelta region. On the shelf, the maximum concentration values of HgT
377 decreased steadily westward from $0.21 \mu\text{g g}^{-1}$ (I36, TYR12), $0.12\text{-}0.17 \mu\text{g g}^{-1}$ (95-19, 95-21), to 0.08-
378 $0.10 \mu\text{g g}^{-1}$ (TYR23, 95-9A) (Fig. 1, Fig. SI.1) with significantly higher Hg in the eastern part of the
379 shelf compared to the western part (t -test, $p < 0.01$, Table 2). This implies that the main Hg source for
380 the whole area is the RR, which is a sort of truism for one of the most industrialized rivers in Europe
381 documented for its Hg contamination (Poulier et al., 2019). This marked trend likely is consistent
382 with the increasing proportion of material from catchments of Languedoc rivers in the western part of
383 the GoL compared to the eastern part (Révillon et al., 2011).

384 *5.1.2. Contrasting Hg accumulation on continental shelf and slope*

385 Low HgT values were encountered in surface sediment at the head of the canyons (HD and STKI19), in
386 environments where resuspension and ephemeral sediment deposition prevail (Palanques et al., 2012),
387 making unlikely long-term sedimentary accumulation, a hypothesis consistent with the conclusion by
388 Courp and Monaco (1990). Sta. STKI19 is located in the distal domain of the head of BC (478 m),
389 which constitutes a bypass or erosive area as fine-grained sediments are winnowed away by the strong
390 currents (Gaudin et al., 2006). Similarly, the head of CdC Canyon contains coarse material (DeGeest et
391 al., 2008), which is usually associated with low Hg content (i.e., Vane et al., 2020). However, a
392 relatively high mean concentration for the continental slope sediments was found (Sta. TYR20); this
393 HgT value is associated with a high C_{org} concentration and low lithogenic content (Table 2). On
394 continental rise stations, HgT concentrations along the continental rise are as low as the sediments at
395 the head of the CdC Canyon with an overall mean of $0.062 \pm 0.014 \mu\text{g g}^{-1}$. There, low C_{org} , high C/N,
396 high Ca, and low Al values of surface sediment suggest a more mineralized OM and an increasing
397 proportion of carbonates relative to lithogenic material, which testifies to the increasing importance of
398 biogenic material from the water column in the seaward direction.

399 5.1.3. *Hg transport: role of organic matter and cascading*

400 In the sediments trap samples collected at Stas. HD, I, and L, the relationship between Hg and OM
401 allows us to highlight the Hg transport. The Hg vs C_{org} relationship is well described by an
402 asymptotic shape (Fig. 7a). This pattern reflects two different origins for the material collected (Fig.
403 SI.4). First, particles with low C_{org} (<1%) and HgT (< $0.13 \mu\text{g g}^{-1}$) concentrations are those of the
404 material trapped at Sta. HD during the cascading event recorded in winter/spring 2005, which
405 brought resuspended material (Canals et al., 2006; Puig et al., 2008; Tesi et al., 2010; Stabholz et al.,
406 2013; Durrieu de Madron et al., 2023) mostly composed of carbonate broken shells and OM of
407 terrigenous origin as indicated by the low $\delta^{13}\text{C}$ values (Fig. 2). In a study on the same trap material,
408 these carbonates have been shown to be depleted in several trace elements compared to the clays
409 (Cossa et al., 2014a); present results show that it is also the case for Hg. Secondly, for C_{org} content >
410 1%, i.e., outside of the cascading period, HgT concentrations remained relatively stable with small
411 variations between $0.09\text{-}0.13 \mu\text{g g}^{-1}$, in association with finer material (Fig. 2). In addition, during the
412 post-cascading spring period, more biogenic marine material, as testified by high $\delta^{13}\text{C}$ (Fig. 2), was
413 collected in the trap with similar Hg contents to the material collected before the cascading event.
414 This phenomenon may result from the Hg uptake in the water column by biogenic material formed in
415 the euphotic zone which settled in the station HD trap. Furthermore, at the sediment traps moored on

416 the continental rise during the year 2008 (Stas. I and L), high C_{org} , Ca, and low C/N values indicate
417 higher marine inputs, which come with average HgT concentrations significantly higher than at the
418 head (Sta. HD) of the CdC Canyon (Table 2).

419 In summary, the biological pump seems to scavenge the Hg present in the water column and
420 quantitatively delivers it to the traps deployed near the seafloor in deep-sea environments. Such a
421 process has already been proposed by Gehrke et al. (2009) for Hg transport to the sediments during
422 the mid-Pleistocene in the western Mediterranean. It can be inferred that in the lower 20-30 m water
423 column above the bottom (the nepheloid layer) two types of particles, and their associated Hg, settle
424 down: (i) autochthonous biogenic ones brought by vertical transfer and (ii) reworked material from
425 the shelf advected through cascading and other near-bottom transport processes. In terms of transport,
426 the advective Hg flux through the CdC Canyon ($637 \mu\text{g m}^{-2} \text{a}^{-1}$) is 25 times more intense than the
427 vertical flux on the continental rise ($24 \mu\text{g m}^{-2} \text{a}^{-1}$) (Table 2). Indeed, at the head of the canyon, 28%
428 of the total downward Hg flux occurred during the cascading period which occupied less than 15% of
429 the total sampling period.

430 5.1.4. Meanings of the various Hg-OM relationships

431 The role of OM as an important carrier of Hg in the GoL sediments is evidenced by our results.
432 Indeed, the overall correlation coefficient between Hg and C_{org} in sediments and material from traps
433 is significant ($R^2 = 0.20$, $n = 545$, $p < 0.01$, Fig. 7, Fig. SI.5) as in many other aquatic environments all
434 over the world (e.g., Clifton and Vivian, 1975; Schäfer et al., 2006; Hare et al., 2010; Sanei et al.,
435 2014). However, the Hg: C_{org} ratios stretched over more than one order of magnitude, with higher
436 ratios during the Industrial Era than before AD 1850 and decreasing seaward (Table 3) testifying to
437 diverse local and temporal situations. In surface sediments of the entire GoL, the correlation between
438 HgT and C_{org} is of linear type ($R^2 = 0.54$, $n = 29$, $p < 0.01$) with a positive regression coefficient of
439 $\sim 0.1 \cdot 10^{-4}$. Considering the entire sedimentary columns, the scatterplot exhibits different
440 relationships depending on the location and period (Fig. 7b and c). Linear HgT vs C_{org} relationships
441 are highly significant in most of the zones, but regression coefficients largely differ from negative
442 values to positive ones (from $-0.63 \cdot 10^{-4}$ to $+1.09 \cdot 10^{-4}$, Table 4). This diversity of relationships is
443 explained by the fact that, despite the proven role of OM in Hg transport, the input functions of Hg
444 and C_{org} differ over time; in other words, Hg- C_{org} relationships are disrupted by the time variations of
445 anthropogenic Hg. For example, in the sediment core of Sta. 94-21, where a decrease of Hg inputs

446 was evidenced (Fig. 3b), the Hg and C_{org} are inversely correlated, whereas in surface sediments
 447 (SHw, HD) they are positively correlated with the highest R^2 (Table 4 and Fig. 7c). In Core RHS-
 448 KS57 the HgT concentrations cover various ranges depending on the date of the depositions.
 449 Significant relationships were observed during the periods of anthropogenic Hg inputs since AD
 450 1850, namely from AD 1850 to 1897, when the increase of Hg relatively to C_{org} was low (regression
 451 coefficient $+0.3 \cdot 10^{-6}$, Fig. 7c), and between AD 1897 to 1962 (the peak of Hg contamination) but
 452 multiplied by a factor of 5 (regression coefficient $+1.6 \cdot 10^{-6}$, Fig. 7b). Keeping the C_{org} inputs
 453 constant, this would imply that anthropogenic Hg contamination in the Rhône prodelta has hastened a
 454 first time around AD 1850, but augmented especially at the beginning of the XXth Century. Before
 455 the Industrial Era (prior to AD 1850), the HgT signal in the sediment was independent of the OM
 456 content (Fig. 7c), in a similar way as observed in trapped material (Fig. 7a). This implies that, within
 457 the narrow C_{org} range (0.4-0.6%), the HgT availability in the RR waters was the limiting factor to a
 458 Hg-enrichment of the OM. Still in the same core (RHS-KS57), during the anthropogenic Hg decrease
 459 period (i.e., after AD 1962), there is a total decoupling of Hg and OM content (Fig. 7b), since
 460 organic-rich young deposits bring with them less and less Hg-contaminated particles.

461 **Table 3.** *HgT:C_{org} (mass ratios $\times 10^{-4}$) in sediments from various parts of the GoL. N: number of samples.*

Area	Core	Mean \pm 1 standard deviation (min-max), N
Rhône Prodelta	RHS-KS57-22, 94-21 (Industrial Era)	0.351 ± 0.257 (0.082 – 1.155), 170
Rhône Prodelta	RHS-KS57 (pre-industrial period)	0.079 ± 0.014 (0.0530 – 0.136), 79
Shelf	95-9A, 95-19, 95-21, TYR91	0.119 ± 0.106 (0.029 – 0.705), 64
CdC Canyon	G & H	0.124 ± 0.045 (0.036 – 0.189), 53
Continental rise	I & L	0.103 ± 0.032 (0.043 – 0.172), 39

462
 463 In summary, two sources for Hg in the GoL sediments are evidenced, continental and marine.
 464 The former derives from rivers and is dispersed on the shelf and transferred to depths *via* submarine
 465 canyons, with some contribution of eroded material from the shelf. Active Hg transfer from the shelf
 466 sediment to the deeper parts of the margin occurs during the cascading events associated with dense
 467 water formation. Seaward, the particulate Hg export is associated with marine Hg inputs from
 468 atmospheric deposition, scavenged by the biological pump and reaching the sediments with the
 469 settling material. During these particle transport processes, in part controlled by the particulate OM

470 transport, Hg overcomes phase change reactions. In this regard, it is worthwhile pointing out that the
 471 material collected in the traps was Hg-enriched, with a factor of 1.5-1.9, compared to the surface
 472 sediments below, whereas similar enrichment was much higher for the C_{org} (2.5-8.0). This suggests
 473 that during the intense OM mineralization in the benthic boundary layer before its burial into the
 474 sedimentary record, the Hg is not fully mobilized and that a large proportion is reabsorbed.

475 **Table 4.** HgT ($\mu g g^{-1}$) versus C_{org} (%) relationships in sediment cores. RD_P and RD_D : Rhône prodelta proximal
 476 and distal parts; SH_E and SH_W : GoL shelf eastern and western parts; HD : Head of the CdC Canyon. *NS*: non-
 477 significant. In surface sediments, R^2 is 0.54, the regression coefficient +0.1009, and the origin +0.0234
 478 ($p < 0.01$).

Zone	Core	Regression coefficient (Slope)	Origin	R^2	p
Rhône prodelta					
RD_P	RHS-KS22	-	-	0.03	NS
RD_P	RHS-KS57	+1.0852	-0.4138	0.43	<0.01
RD_D	94-21	-0.6289	+1.4744	0.60	<0.01
Gulf of Lion shelf					
SH_E	surface	-0.0974	+0.3146	0.71	<0.01
SH_W	surface	+0.1441	-0.0122	0.88	<0.01
SH_W	95-21&9A	+0.1111	-0.0827	0.37	<0.10
Cap de Creus Canyon					
HD	surface	+0.13334	-0.0194	0.92	<0.01
Canyon	G & H	+0.2752	-0.0897	0.65	<0.01
Continental rise					
Rise	I & L	+0.1457	-0.0136	0.66	<0.01

479

480 5.2. Chronology of Hg accumulation

481 5.2.1. Hg chronology in the prodelta sediments and coal emissions

482 Core RHS-KS57 permits a long historical record that goes back to the beginning of the XVIIth
483 Century. At the base of the core, HgT concentrations reach low values of $\sim 0.030 \mu\text{g g}^{-1}$ but, at this
484 stage, nothing indicates that this level, occurring in AD 1600, is representative of the pristine level of
485 the prodelta sediments. Rather the time trend in HgT and HgT:C_{org} ratios suggest that human
486 influence began well before the onset of the Industrial Era (insert, Fig 4a). As early as the second part
487 of the XVIIth, Hg concentrations grew from 0.03 (AD 1650) to $0.04 \mu\text{g g}^{-1}$ (AD 1850), with most of
488 the increase between AD 1650 and AD 1700. Elbaz-Poulichet et al. (2011) already observed in a
489 sediment core from the Pierre-Blanche Lagoon, a Hg broad peak during the XVIIth Century, which
490 they attributed to the expansion of gold and silver amalgamation in Europe and particularly in the
491 South of France (Cévennes). There, many auriferous rivers were exploited as soon as the Roman
492 period (Domergue et al., 2006). However, the Hg increase rate enhanced at the beginning of the
493 Industrial Era and particularly \sim AD 1900, then peaking during the AD 1960-70 decade followed by a
494 rapid decline at the end of the XXth Century and the first decade of the XXIth (Fig. 4a). The Hg
495 increase rate in core RHS-KS57 reached 25 times between AD 1850 and AD 1962 i.e., the date of the
496 maximum, with Hg concentrations varying from $0.04 \mu\text{g g}^{-1}$ to $1.02 \mu\text{g g}^{-1}$; this corresponds to an
497 increase rate of ca. $3\% \text{ a}^{-1}$ (Table 5). The decrease between AD 1962 and the first decade of the XXIth
498 Century was more rapid ($3.6\% \text{ a}^{-1}$) allowing the HgT to drop down to $0.19 \mu\text{g g}^{-1}$. This overall Hg
499 time trend resembles the evolution of coal consumption in France which started ca. AD 1830 and
500 culminated in the sixties with two mega drops during the two world wars of the XXth Century (Conus
501 and Escudier, 2006). Such time trends have already been observed for several metals in the same
502 core, especially for Pb, tracer coal residues and coal-burning emissions, and Zn (Core RHS-KS57,
503 Cossa et al., 2018a), and in sediment cores collected in the Rhône River (Ferrand et al., 2012).
504 Parallel profiles between Hg and Pb were also observed in Mediterranean sediments from a lagoon
505 nearby (85 km west) the Rhône mouth (Elbaz-Poulichet et al., 2011) to the Levantine margin of the
506 eastern basin (Azoury et al., 2013). At this latter location, Hg and Pb contaminations were associated
507 with the presence of polyaromatic hydrocarbons from combustion origin. These coincidences
508 strongly suggest a common coal-burning source for Hg and Pb. Coal consumption in France started
509 ca. AD 1820 and gradually increased to reach 60 000 Tg at the beginning of the XXth Century,
510 culminating ca. 80 000 Tg in the sixties, then dropping down until the end of the Century (Suppl.
511 Info. Fig. SI.6; Conus and Escudier, 2006). In addition, French coal consumption during the XXth
512 Century crashed during the two World wars (Fig. SI.6). The HgT concentrations in the sediments of

513 Core RHS-KS57 follow a similar time-trend, starting to increase in the second part of the XIXth
514 Century for a 100-years long period and culminating in the sixties, then decreasing progressively
515 until the end of the XXth Century (Fig. 4a). Short-term variations are superimposed to this broad
516 pattern. After the first steady increase, HgT concentrations stood between AD 1900-1920, a period
517 which comprises the First World War, and then increased again until AD 1925, followed by a drop
518 covering AD 1926-1943, a period which includes the Great Recession in Europe and the first part of
519 the Second World War. Interestingly, during the First World War, while most of the French coal
520 production crashed, the Cevennes mining (South of France), which fed the coal consumed in the
521 entire Rhône Valley, continued to increase between 1914 and 1920 (Chancerel, 2012). This situation
522 is more than likely the reason why the sedimentary Hg concentrations did not decrease during the
523 period 1914-1918 in line with the evolution of coal consumption in France (Fig. 4a). The Hg
524 evolution between AD 1930-1943 coincides with the decrease in coal consumption which has started
525 ca AD 1930 and extended until 1943. The co-evolution between HgT and coal consumption
526 continued until the end of the beginning of the XXIth Century while France was still using several Tg
527 of imported coal mainly for energy production.

528 It is worthwhile pointing out that in Core RHS-KS57, Pb concentrations peak at the end of the
529 seventies (Cossa et al., 2018a), whereas Hg peaked ten years before (Fig. 4a). In addition to coal,
530 other sources of Pb (and Hg) exist. The various Hg vs Pb relationships allow sorting out these
531 sources depending on the period (Fig. 8a). Until the beginning of the XXth Century Hg and Pb
532 covaried with low regression coefficients (0.001-0.002), during the 1900-1970 period, Hg increased
533 steeply relative to Pb (regression coefficient 0.018), then Hg decreased tremendously and Pb
534 remained high (regression coefficient 0.006). After AD 1990, concentrations of both metals dropped
535 down, with a regression coefficient regaining its initial value. These variations suggest that during
536 the 1970-1990 period, a secondary source of Pb existed. Indeed, with the advent of the oil age, the
537 Pb has been more and more used as a gasoline additive, and its emission culminated in AD 1975,
538 then decreased to cease in AD 2000 (Suppl. Info. Fig. SI.7a). The influence of Pb emission from
539 gasoline consumption has been recorded in the sediment cores from CdC Canyon as shown by the
540 low values of $^{206}\text{Pb}:^{207}\text{Pb}$ and $^{206}\text{Pb}:^{208}\text{Pb}$ isotopic ratios (Fig. SI.7b). The reason for the shift
541 between the maxima of Hg and Pb, therefore, resides in the multiplicity of recent Pb sources, which
542 are illustrated in figure 8b. The low $^{206}\text{Pb}:^{207}\text{Pb}$ ratios (<1.175) indicate the gasoline source and
543 occur with constant Hg concentrations. $^{206}\text{Pb}:^{207}\text{Pb}$ ranging from 1.175 to 1.190 indicate coal sources

544 (Fig. SI.7b) and occur with high Hg concentrations (Fig. 8). In addition, Figure 8b allows us to
 545 characterize HgT concentrations in pre-industrial Pb sediments ($^{206/207}\text{Pb} \geq 1.20$), averaging $0.035 \pm$
 546 $0.003 \mu\text{g g}^{-1}$ ($n = 33$) and $0.013 \pm 0.003 \mu\text{g g}^{-1}$ ($n = 9$) for riverine and marine sources, respectively,
 547 two values that can be taken as reliable estimates of the pre-industrial Hg levels.

548 **Table 5.** Period covered by the cores and time trends observed from the HgT concentration profiles for the
 549 various cores.

Station	Period (AD)	Hg time trend (% a ⁻¹)	
		Increase	Decrease
Rhône Prodelta			
RHS-KS22	1973 – 2008	–	2.0
RHS-KS57	1600 – 1850	0.3	–
	1850 – 1963	3.0	–
	1964 – 2008	–	3.6
61C	1943 – 1963	1.2	–
	1964 – 2008	–	2.8
62C	1963 – 2008	–	2.7
94-21	1943 – 1961	1.9	–
	1962 – 1993	–	2.2
Gulf of Lion Shelf			
95-19	1925 – 1980	3.0	–
95-21	1925 -1970	2.5	–
Cap de Creus Canyon			
G	1850 – 1963	1.0	–
	1963 – 2005	–	0.2
H	1850 – 1963	1.2	–
	1963 – 2008	–	0.5
Continental Rise			
I	1850 – 2005	0.5	–
L	1850 – 2005	0.4	–

550

551 The entire (835 cm-long) Core RHS-KS22 covers a 36-year-long period (AD 1972-2008)
552 which corresponds to the upper 90 cm of Core RHS-KS57. In this time interval, the main trend of Hg
553 concentrations is similar between the two cores (Fig. 3a), showing a decrease between the seventies
554 and the time of core collections of roughly a factor of 3-5 depending on the core. This allows
555 calculating a Hg concentration decreasing rate of ca. 2% per year since the seventies (Table 5).
556 Noteworthy, the Hg profile in the RHS-KS57 core exhibited roughly constant concentration since the
557 end of the nineties (Fig. 3), which may result from the mixing of the top of the sediment collected
558 with a piston corer. A detailed examination of HgT variations in Core RHS-KS22 indicates that low
559 Hg concentrations coincide with sandy layers (see especially depths 350 and 755 cm in Fig. SI.2). In
560 addition, besides the sandy influence, a positive correlation between the HgT concentrations and the
561 RR discharge, especially with the Durance tributary discharge ($R^2 = 0.44$, $n = 83$, $p < 0.001$) has been
562 found using the monitoring database of the French rivers (Hydrofrance web site). This relationship
563 suggests a possible effect of the resuspension of Hg-rich fine sediment of the RR with increasing
564 river discharge.

565 5.2.2. *Repercussion in the prodelta sediments of the recent Hg decrease in the Rhône River*

566 The three short box-cores (94-21, 61C, and 62C) collected in the distal part of the Rhône prodelta
567 (Fig. 1) allow documenting more accurately the Hg evolution of the second part of the XXth Century;
568 the values of their SRs range from 0.45 to 0.70 cm a⁻¹ (Table SI.3). Thus, the period covered by the
569 three cores starts at the end of the Second World War. Cores 94-21 and 61C showed the highest Hg
570 concentrations in the early sixties peaking at 0.75 and 0.58 $\mu\text{g g}^{-1}$; the highest HgT concentration
571 (0.69 $\mu\text{g g}^{-1}$) was observed at ~AD 1962) (Fig 3b). The HgT concentrations rapidly decrease
572 consistently towards the sediment-water interface in the three cores, attaining at the sediment surface
573 (i.e., ~2008 AD) 0.19 and 0.28 $\mu\text{g g}^{-1}$ in core 62C and 61C, respectively. Noteworthy, the higher HgT
574 concentration of core 61C corresponds to a homogenous layer probably resulting from the mixing of
575 the top 10 cm of the sediment, equivalent to ~20 years of deposition. From the three profiles (Fig.
576 3b), rates of Hg decrease in the last 46 years is 1.6 % per year, a similar rate as calculated for cores
577 RHS-KS22 and RHS-KS57. If the Hg decrease continues at the same linear rate, the pre-industrial
578 Hg level might be reached after ~20 years; however, the decrease does not seem to be linear and an
579 exponential model would probably be more appropriate to describe the Hg time trend since soils and

580 sediments of the RR basin are still Hg-enriched from the anthropogenic Hg left from the deposition
581 of the Industrial Era. In brief, the Hg concentrations measured in the sediments of the six cores
582 collected in the Rhône prodelta exhibit a 1.6-2 % annual decrease during the last 50 years. They also
583 show that the most recently deposited sediments (AD 2008-2011) have a similar Hg concentration
584 range ($0.13\text{-}0.27 \mu\text{g g}^{-1}$) as the annual mean of Hg concentrations in the RR suspended particles
585 collected near the river mouth (Arles), which consistently ranged from 0.1 to $0.2 \mu\text{g g}^{-1}$ for the period
586 2011-2016 AD (Poulier et al., 2019; Delile et al., 2022), in agreement with the Hg content of recent
587 deposits at RR mouth (RDp: $0.16 \pm 0.03 \mu\text{g g}^{-1}$, Table 2).

588 *5.2.3. Hg chronology in the slope and rise sediments*

589 Canyon sediments receive inputs from the GoL shelf, especially during cascading events. Deep
590 cascading phenomena play a major role in the shelf export of particulate material, including metals,
591 as well as their redistribution in the basin by deep convection (Durrieu de Madron et al., 2023). In the
592 CdC Canyon sediments (cores G and H), HgT concentrations start to increase at the beginning of the
593 XIXth Century to culminate between AD 1950 and 1980 (Fig. 5b). The surface sediments are
594 homogenous over the upper 3.5 cm, averaging $0.11 \pm 0.01 \mu\text{g g}^{-1}$ in the AD 1994-2002 period. This
595 chronology matches that of core RHS-KS57 strongly suggesting that the CdC Canyon is the main
596 conduit for the exportation of particles from the RR. Hg concentrations in non-dated core STKI19,
597 collected in the BC, varied from 0.02 to $0.08 \mu\text{g g}^{-1}$ with a rapid change in HgT concentrations
598 between 29 and 30 cm (Fig. 5c). The X-ray imaging and the ^{210}Pb activity performed on the same
599 core (Gaudin et al., 2006) allowed identifying a 30-cm thick structureless upper sequence and a lower
600 sequence. The presence of Hg-contaminated sediments, associated with measurable ^{210}Pb activities,
601 only in the upper sequence of the sediments supports that the distal part of the BC head is subject to
602 the accumulation of modern sediments. In the absence of a real age model for this core, we are
603 constrained to hypothesize that this distal part of the head of BC has received a massive input of Hg-
604 enriched sediment, accumulated on the shelf break thru the Industrial Era during a period of dense
605 water cascading at the shelf edge, such as those described by Gaudin et al. (2006). Our results
606 support the conclusion of these authors that the modern sedimentary activity of BC is made of
607 alternative phases of deposition and erosion.

608 The ^{210}Pb inventories in continental rise sediments (cores I and L) suggest that they receive
609 most of their material from downward settling in the upper water column, but also from particles

610 episodically advected from the margin during deep cascading pulses (Tesi et al., 2010; Palanques et
611 al., 2012; Puig et al., 2013; Durrieu de Madron et al., 2023). The presence of planktonic material is
612 attested by the coarse carbonate material, including foraminifera and pteropods, in core I, and finer
613 planktonic-derived material in core L (Cossa et al., 2014a). The chronology of Hg deposition in core
614 I is parallel to that of core RHS-KS57 suggesting the influence of RR inputs on the continental rise
615 through the CdC Canyon. However, the time resolution obtained for core L does not permit access to
616 similar details, since the last 70 years are covered with only three measurements (Fig. 6a and b).
617 Recent anthropogenic Hg inputs and bioturbation likely control the shape of the Hg concentration
618 increase toward the sediment surface. A similar interpretation can be applied to cores KIGC2 and 5,
619 the upper 10 cm of which consisting of carbonated biogenic mud and SRs are in the order of 0.002
620 cm a^{-1} (Denniellou et al., 2009). Thus, we can reasonably hypothesize that the HgT enrichment in the
621 top of these cores on the continental rise sediments is due to anthropogenic Hg accumulated in the
622 Industrial Era (Fig. 6b) and that the Hg profiles are affected by biomixing.

623 **5.3. Anthropogenic Hg: sources and quantification**

624 The HgT concentration distributions observed in the near-shore surface sediments (Fig. SI.1) are
625 consistent with those of the most recent survey by Bouchoucha et al. (2018) which identifies the
626 Marseille harbor as a highly Hg-contaminated area of the GoL coast. The surface sediment collected
627 in the Marseille area presented elevated concentrations ranging from 0.30 to 4.90 $\mu\text{g g}^{-1}$, with the
628 highest levels found in the sediments collected in front of the main outflow of the sewage of the city
629 of Marseille. These concentration levels exceed the Environmental Assessment Criteria for Hg in the
630 sediments (0.20 $\mu\text{g g}^{-1}$) of the Convention for the Protection of the Marine Environment (OSPAR),
631 i.e., the Hg concentration in the environment below which no chronic biological effects are expected
632 to occur in marine organisms (Webster et al., 2009). Besides this extreme value, HgT concentrations
633 were higher than the pre-industrial levels and have to be considered Hg-enriched. They comprise
634 lagoons under local Hg inputs, such as the Thau lagoon near which cement and coal-gas plants have
635 been located. To provide an order of magnitude of the Hg-enrichment of surface sediments we have
636 calculated enrichment factors ($\text{EF} = (\text{HgT}/\text{Al})_{\text{surf}} / (\text{HgT}/\text{Al})_{\text{base}}$). For the regions defined in the Table
637 1 caption, apart from lagoons and harbors, the EF values increase from 1.5-3.7 on the continental
638 rise, to 2.3-4.7 in the CdC Canyon, 2.9-6.4 on the GoL shelf, and 6.3-10.7 in the Rhône prodelta,
639 pointing out the RR as the main source of these Hg-enrichments. The Rhône prodelta proximal (RD_P)

640 and the distal parts (RD_D) can be distinguished based on their Hg mean concentrations (t-test,
641 $p < 0.01$, Table 2).

642 The inventories of the anthropogenic Hg accumulated during the Industrial Era (Hg_{anthr}) are
643 summarized in Table 6 and totaled an average of 152 Mg. Pre-industrial background Hg
644 concentrations, of 0.013 to 0.035 $\mu\text{g g}^{-1}$ depending on the zone, have been subtracted in order to only
645 take into account the Hg_{anthr} fractions (Table SI.4). Put in perspective, this total amount, equally
646 distributed over the 150 years of the Industrial Era, gives a virtual rate of 1 Mg accumulated each
647 year. This figure is close to the current Hg accumulated annually in the sediments of the GoL (0.9
648 Mg a⁻¹ according to Cossa et al., 2018b). On the other hand, it is 0.5% of the Hg buried annually in
649 all the ocean margins (200 Mg a⁻¹ according to Outridge et al., 2018), which is huge for an area that
650 represents less than 1‰ of the total surface of the ocean margins.

651 **Table 6.** Quantities of Hg_{anthr} (Mg) accumulated in various parts of the sediment of the GoL and its continental
652 rise. The calculation method is given in Material and Methods (section 3.4.) and Table SI.4.

Proximal prodelta	Distal prodelta	Inner shelf	Outer shelf	CdC Canyon	Continental rise	Total
63	33	10-25	6-30	1	20	132-172

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654 5.4. Methylmercury sources

655 MMHg present in the surficial sediments may originate from MMHg borne with settling material.
656 However, there are enough pieces of evidence from which it appears that the MMHg concentrations
657 in aquatic sediments reflect the net microbiological methylation-demethylation rates within the
658 sediment itself (e.g., Parks et al., 2013). Here, MMHg concentrations were systematically positively
659 correlated to the HgT regardless of the location, with R² always statistically significant ($p < 0.01$)
660 varying from 0.30 to 0.94 (Fig. SI.8). Noteworthy is that the slopes of the MMHg vs HgT
661 relationships were maxima with the particles from the sediment traps. Since MMHg is always less
662 than 1 % of the HgT, HgT can be used as a proxy of the inorganic Hg^{II} which constitutes the
663 substratum for bacterial methylation. Thus, we used a model resembling Michaelis–Menten kinetics
664 synthesizing the enzymatic methylation and demethylation reactions to explore the MMHg vs HgT
665 relationship as earlier proposed (Cossa et al., 2014b). Even empirical, such fits offer the advantage of
666 a mechanistic foundation (i.e., enzymatic processes governing Hg methylation).

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$$MMHg = \frac{a \times HgT}{Km + HgT}$$

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where “*a*” is the saturation MMHg concentration and “*K_m*” estimates the HgT concentration, which corresponds to MMHg half-saturation. This parameter can be used as a proxy of Hg methylation efficiency, as *K_m* increases, methylation efficiency decreases. It appears from data in Table 7, that net methylation efficiency is maximum for samples collected in the sediment traps or at the surface of the sediment, and one order of magnitude smaller than *K_m* values calculated for sediment cores. At the same time, the magnitude of the variance of MMHg explained by the HgT (i.e., *R*²) is smaller in these “younger” particles suggesting that for them the methylation efficiency is not primarily controlled by HgT, but more probably governed by other processes, such as microbial activity and inorganic Hg availability for methylator microorganisms (e.g., Cossa et al., 2014b). In sediment cores, *K_m* varied in a smaller range from 16.7 to 43.1 μg g⁻¹. The highest methylation efficiencies are found in the proximal part of the Rhône prodelta, an area known for its high rates of OM mineralization driven by sulfate-reducing bacteria (e.g., Pastor et al., 2011) also known as Hg methylators. For comparison purposes, we have calculated a similar *K_m* (~ 25 μg g⁻¹) for a 15 cm sediment core collected in the adjacent Balearic Abyssal Plain in the western Mediterranean basin. Altogether, these results suggest that Hg net methylation is less efficient for particles buried in the sedimentary record, supporting the idea that the water column is a main source for MMHg in marine environments and that, during sediment burial, demethylation seems prevailing albeit the remanence of an MMHg background at depth. Our results support the general proposition by Jonsson et al (2012): “...a combination of thermodynamic and kinetic effects of Hg^{II} solid-phase dissolution and surface desorption control the Hg^{II} methylation rate in sediments and cause the large observed differences in *K_m*-value.” This suggests that the surficial sediment, where OM is not matured, provides a medium where Hg bounds to OM are weaker than deeper in the sediment where OM sulfidization offers stronger ligands to Hg. In addition, the recently deposited sediments are MMHg-enriched in the foraging zone of meiofauna organisms, favoring by this way its biomagnification into benthic food webs.

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Table 7. Parameters of the Michaelis-Menten relationships between MMHg and HgT. *K_m* (μg g⁻¹) values were calculated for an “*a*” value of 0.056 μg g⁻¹ estimated based on more than 1400 couple values (Cossa et al., 2014b). *Italics* for sediment trap samples; *RD_P* and *RD_D*: Rhône prodelta proximal and distal parts; *SH*: GoL shelf; *CdC*: Cap de Creus; *Core WB* was collected in the Balearic Abyssal Plain in the Western Mediterranean (Cossa et al., 2021).

Zone	Core/trap	Km	R ²	p
RD _p	RHS-KS57	16.7 ± 5.5	0.89	<0.001
RD _D	61C-62C	31.5 ± 7.0	0.61	<0.001
SH	I36	19.3 ± 0.3	0.92	<0.001
SH	Surface	4.0 ± 0.8	0.70	<0.001
CdC Canyon	G & H	43.1 ± 14.9	0.77	<0.001
Continental rise	L	28.4 ± 12.0	0.94	<0.001
Continental rise	I-L Traps	2.4 ± 1.2	0.19	<0.05
Abyssal Plain	WB	25.4 ± 15.0	0.34	<0.01

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699 **6. Summary and conclusions**

700 1. Anthropogenic Hg has been identified in the Gulf of Lion sediments. In surface sediments, the Hg-
701 enrichment factors decrease from 10.7 in the Rhône prodelta to 1.5 in the continental rise. The Hg
702 distribution in surface sediments clearly shows an E-W gradient. The Rhône River is pointed as the
703 major anthropogenic Hg source for the Gulf of Lion sediments.

704 2. Anthropogenic Hg accumulated since the beginning of the Industrial Era is ca. 150 Mg, mainly
705 from the Rhône River. The majority of this accumulation has occurred in the prodelta sediments (ca.
706 100 Mg).

707 3. The chronological trend of anthropogenic Hg accumulated in the Rhône prodelta sediments
708 indicates an abrupt increase at the beginning of the Industrial Era parallel to the coal consumption in
709 France starting in AD 1830, exhibiting a maximum ~ AD 1962, then decreasing at the end of the
710 XXth Century.

711 4. The Cap de Creus Canyon is a focusing path and a site of (transient/ephemeral) accumulation for
712 Hg transported from the shelf to the continental rise, especially during deep cascading events; 20 Mg
713 of anthropogenic Hg are accumulated in the continental rise sediments. The marine water column of
714 the Gulf of Lion is also a Hg source for the sediments; its contribution increases seaward and quite
715 significantly in the continental rise sediments.

716 5. The OM is the main carrier of Hg to the sediments. Before the Industrial Era, in the riverine
717 sediments settled in the proximal prodelta, the Hg:C_{org} mass ratios averaged $0.08 \pm 0.01 \cdot 10^{-4}$, whereas
718 in the currently settling particles collected along the continental rise, which are mostly composed of
719 plankton, Hg:C_{org} mass ratios average $0.04 \pm 0.01 \cdot 10^{-4}$. In both cases, crossed a threshold of OM, the
720 HgT concentrations remained unchanged (~ 0.04 and $0.12 \mu\text{g g}^{-1}$, in riverine deposited sediment of the
721 prodelta and marine plankton collected in the sediment traps, respectively) regardless of the C_{org}
722 contents. These observations suggest that Hg-enrichment of the scavenging OM is limited by the Hg
723 available in the water. On the contrary, in the proximal delta sediments and during the Industrial Era,
724 the HgT and OM covary on a wide range of concentrations, with Hg:C_{org} mass ratios reaching up to
725 $1.15 \cdot 10^{-4}$ in AD 1962; since the beginning of the XXIth Century, the values are stabilized around
726 $0.25 \cdot 10^{-4}$.

727 6. The monomethyl mercury abundance in the sediment is a function of its total Hg content but with
728 proportions decreasing with increasing depth. Meanwhile, the settling particles trapped near the
729 bottom and the surficial sediments present the highest net methylation efficiencies.

730

731 **Data availability**

732 Data has been made available in Electronic Supplementary (EXCEL file “*Data_Hg_GoL_(Cossa et al.,*
733 *2023)*”).

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739 determinations in cores KIGC and STKI. Cores TYR12, TYR20, and TYR23 have been collected
740 during the EROS-2000 (European River Ocean System) project supported by the European Union
741 through the Environment and Climatological and MAST Programs and grab samples and cores 94-
742 21, 95-9A, 9519, 95-21 and I36 have been collected in the framework of the "Réseau National

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744 Ministry for the Environment (<http://www.ifremer.fr/envlit/surveillance/rno.htm>).

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Table captions

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1083 **Table 1.** Surface sediments (0-1 cm). Organic carbon, C/N, Ca, Al, and HgT (mean \pm 1 standard
 1084 deviation, number of determinations in brackets). Rhône prodelta (RD_P: proximal part includes ME,
 1085 62C, and Stas. 1-4; RD_D: distal part includes 61D, 94-21, and Sta. 5), Gulf of Lion shelf (SH_E: east part
 1086 includes Sta. 6, PR12, PR14, PR15, and PR18, I36, 95-19, TYR12; SH_W: west part includes Stas. 7-
 1087 16, PR2-4, PR10-11), head of the Cap de Creus Canyon (HD includes Stas. 17-23), head of the
 1088 Bourcart Canyon (STKI19), Cap de Creus Canyon (cores G and H), continental slope (KIGC25 and
 1089 TYR20), and continental rise (cores I, L, and KIGC1&2).

1090 **Table 2.** Sediment traps (moored 20 m above the bottom). Organic carbon, C/N, $\delta^{13}\text{C}$, Ca, Al, and
 1091 HgT (mean \pm 1 standard deviation, number of determinations in brackets). (*) The HgT mean differs
 1092 from the two others (t-test, $p < 0.01$).

1093 **Table 3.** HgT:C_{org} (mass ratios $\times 10^{-4}$) in sediments from various parts of the GoL. N: number of
 1094 samples.

1095 **Table 4.** HgT ($\mu\text{g g}^{-1}$) *versus* C_{org} (%) relationships in sediment cores. RD_P and RD_D: Rhône prodelta
 1096 proximal and distal parts; SH_E and SH_W: GoL shelf eastern and western parts; HD: Head of the CdC
 1097 Canyon. NS: non-significant. In surface sediments, R^2 is 0.54, the regression coefficient +0.101, and
 1098 the origin +0.023 ($p < 0.01$).

1099 **Table 5.** Period covered by the cores and time trends observed from the HgT concentration profiles
 1100 (\uparrow increase, \downarrow decrease) for the various cores.

1101 **Table 6.** Quantities of Hg_{anthr} (Mg) accumulated in various parts of the sediment of the GoL and its
 1102 continental rise. The calculation method is given in Material and Methods (section 3.4.) and Table
 1103 SI.4.

1104 **Table 7.** Parameters of the Michaelis-Menten relationships between MMHg and HgT. K_m ($\mu\text{g g}^{-1}$)
 1105 values were calculated for an “ a ” value of $0.056 \mu\text{g g}^{-1}$ estimated based on more than 1400 couple
 1106 values (Cossa et al., 2014). Italics for sediment trap samples; RD_P and RD_D : Rhône prodelta proximal
 1107 and distal parts; SH: GoL shelf; CdC: Cap de Creus; Core WB was collected in the Balearic Abyssal
 1108 Plain in the Western Mediterranean (Cossa et al., 2021).

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Figure captions

1112 **Figure 1.** Sample sites in the Gulf of Lion (GoL). CdC: Cap de Creus; BC: Bourcart Canyon; Open
 1113 circles: grab samples; filled circles: sediment cores; Red cross: sediment traps. The brown line indicates
 1114 the limit of the continental shelf (isobath 200 m); the blue lines indicate the axes of the canyons.

1115 **Figure 2.** Mercury (HgT) concentrations and chemical characteristics of the material collected in trap
 1116 HD located at the head of Cap de Creus Canyon during the period October 2004 - June 2005.

1117 **Figure 3.** Mercury (HgT) concentration profiles *versus* time in the sediment cores collected Rhône
 1118 prodelta area. (a) cores RHS-KS22 and RHS-KS57; (b) cores 61C, 62C, and 94-21.

1119 **Figure 4.** (a) Mercury (HgT), (b) organic carbon (C_{org}), and (c) aluminum (Al) concentration profiles
 1120 *versus* time in the sediment cores RHS-KS57 collected Rhône prodelta area. The insert is an enlarged
 1121 scale for HgT and HgT: C_{org} ratio. WWI and WWII refer to World War I and II, respectively.

1122 **Figure 5.** Mercury (HgT) concentration profiles *versus* time or depth in the sediment cores from the
 1123 shelf (a), the CdC Canyon (b), and BC Canyon (c).

1124 **Figure 6.** Mercury (HgT) concentration profiles in the sediment cores from the continental rise. (a)
 1125 cores I and L *versus* time and (b) cores I, L, KIGC2, and KIGC5 *versus* depth.

1126 **Figure 7.** Mercury (HgT) concentrations *versus* organic carbon (C_{org}). (a) the entire data set, and (b
 1127 and c) for particular periods and core locations. (SHw) west part of the shelf, (HD) head of the CdC
 1128 Canyon. Since HgT is expressed as $\mu\text{g g}^{-1}$ and C_{org} as %, the regression coefficient in mass should be
 1129 multiplied by 10^{-4} .

1130 **Figure 8.** Mercury (HgT) concentrations *versus* lead (Pb) concentrations according to various periods
 1131 in core RHS-KS57. (a) HgT *versus* Pb; (b) HgT *versus* stable isotope Pb ratios.

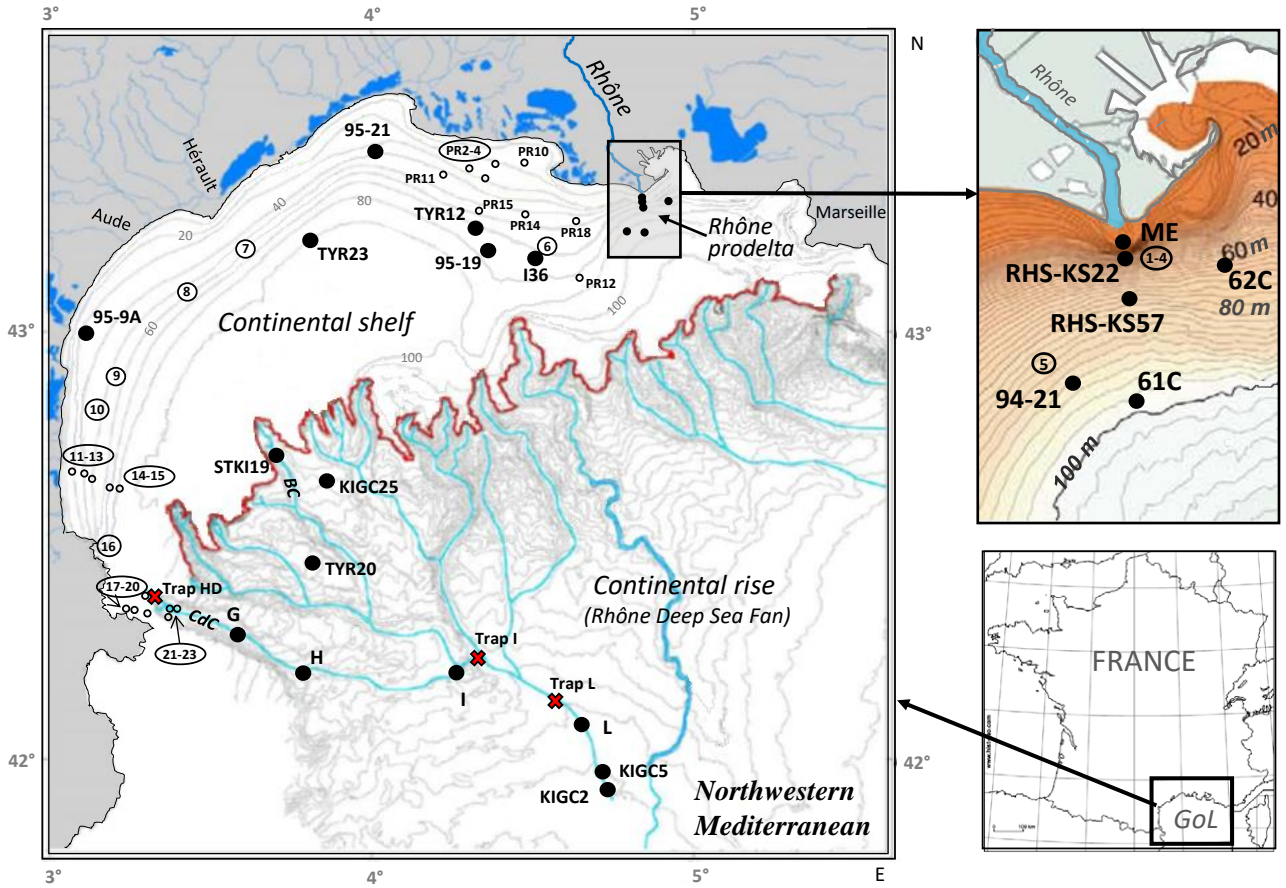
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Figures

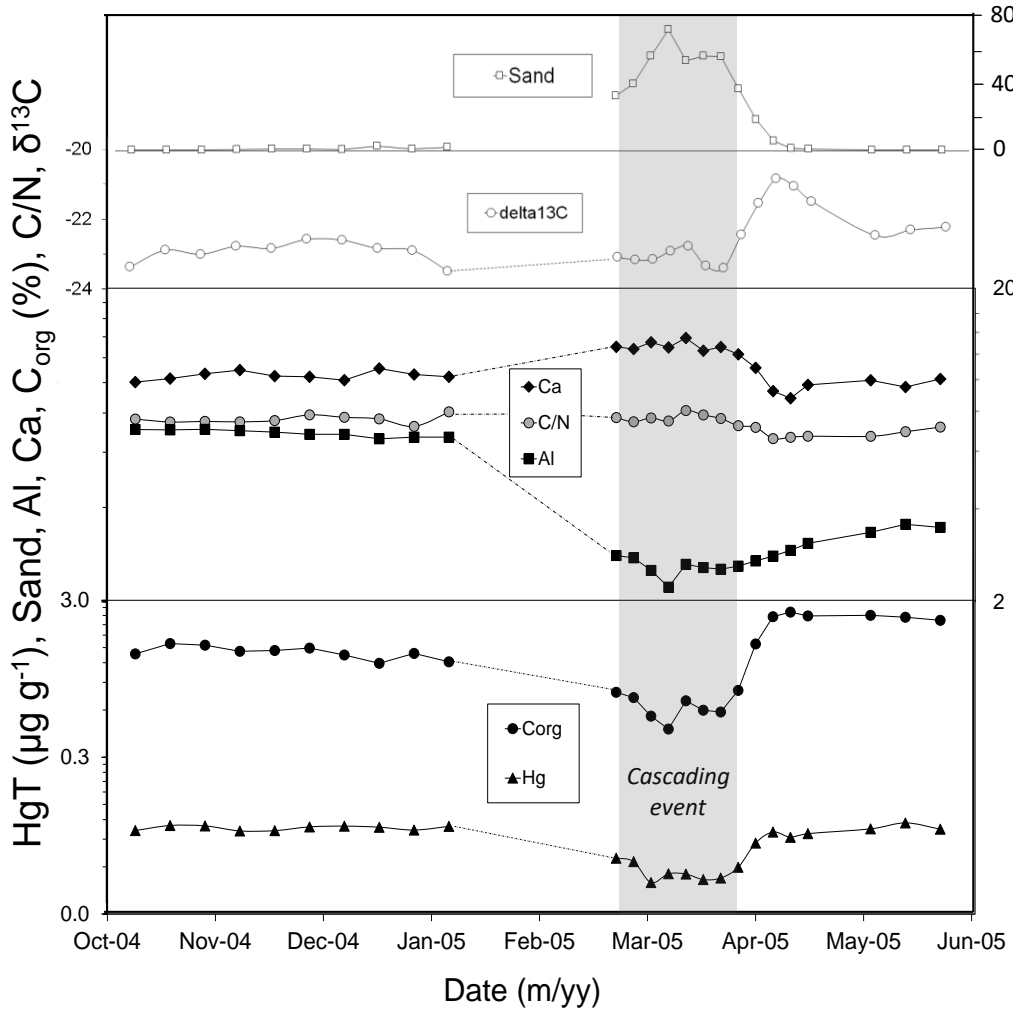


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1137 **Figure 1.**

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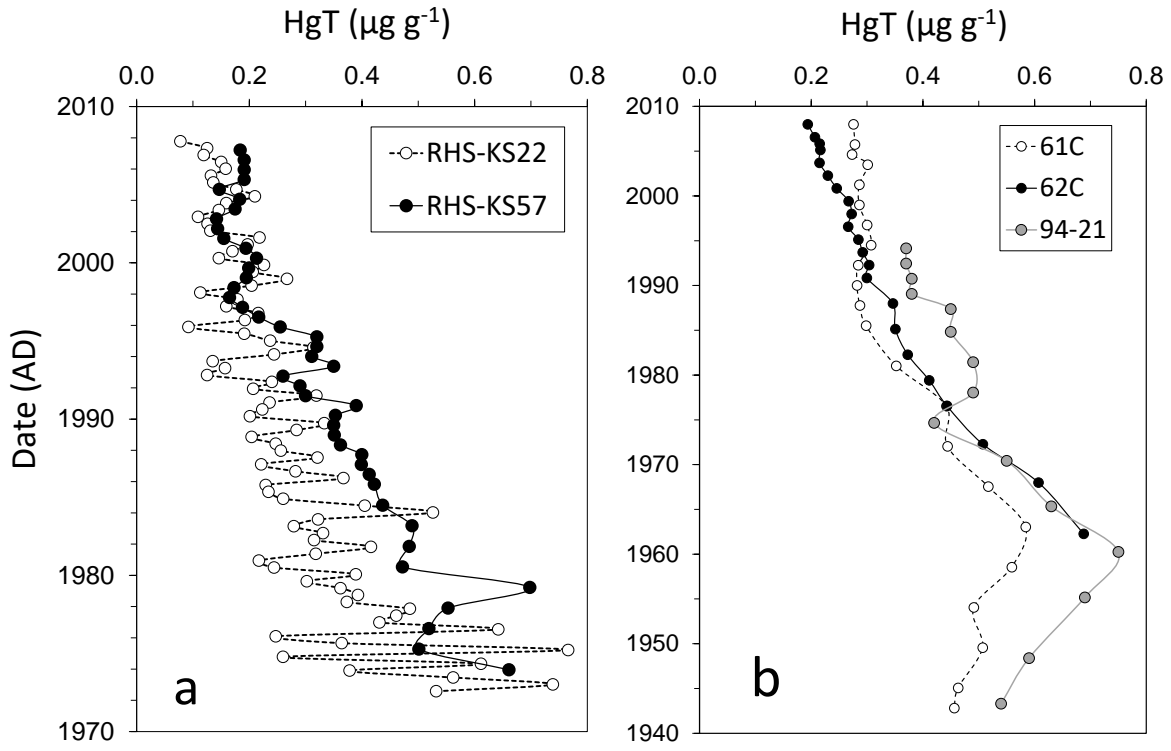
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Figure 2.

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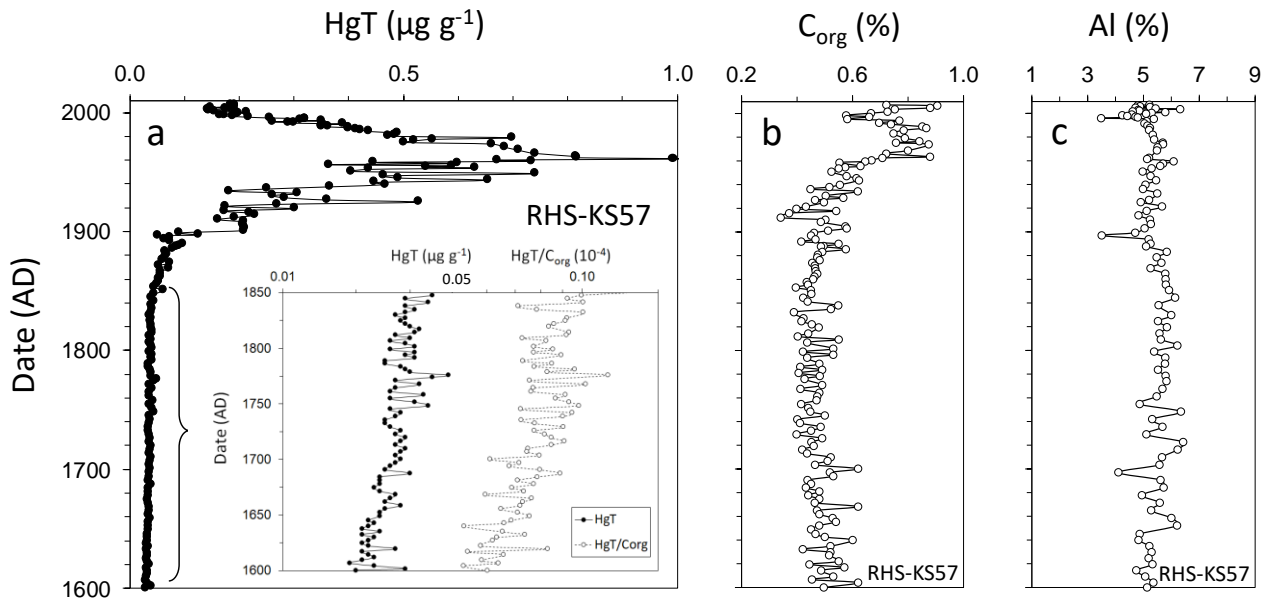
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Figure 3.

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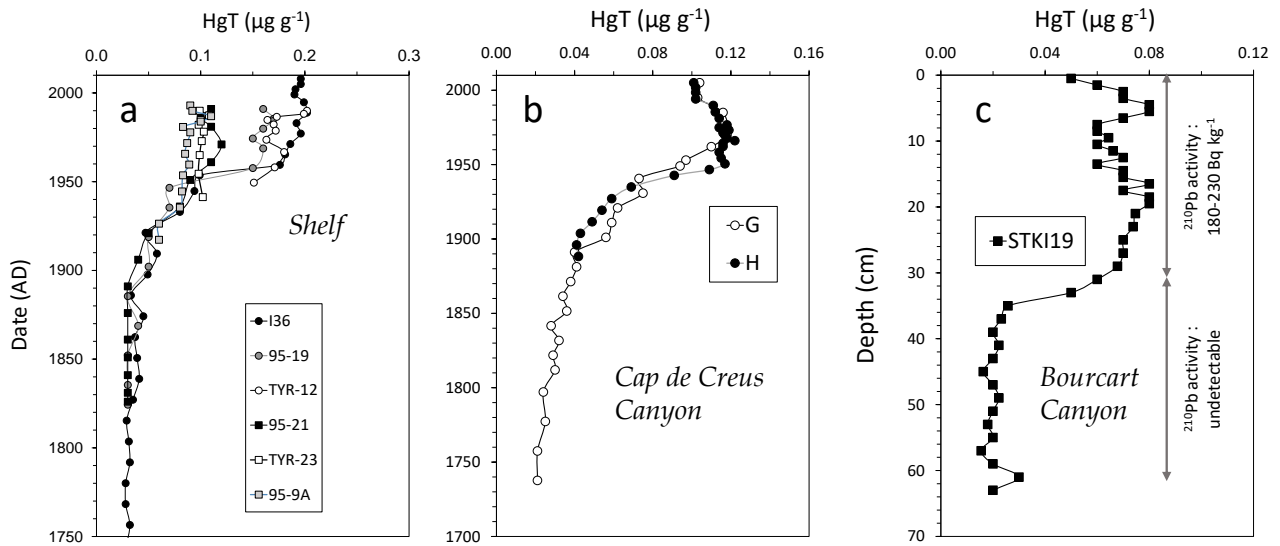
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1150 **Figure 4.**

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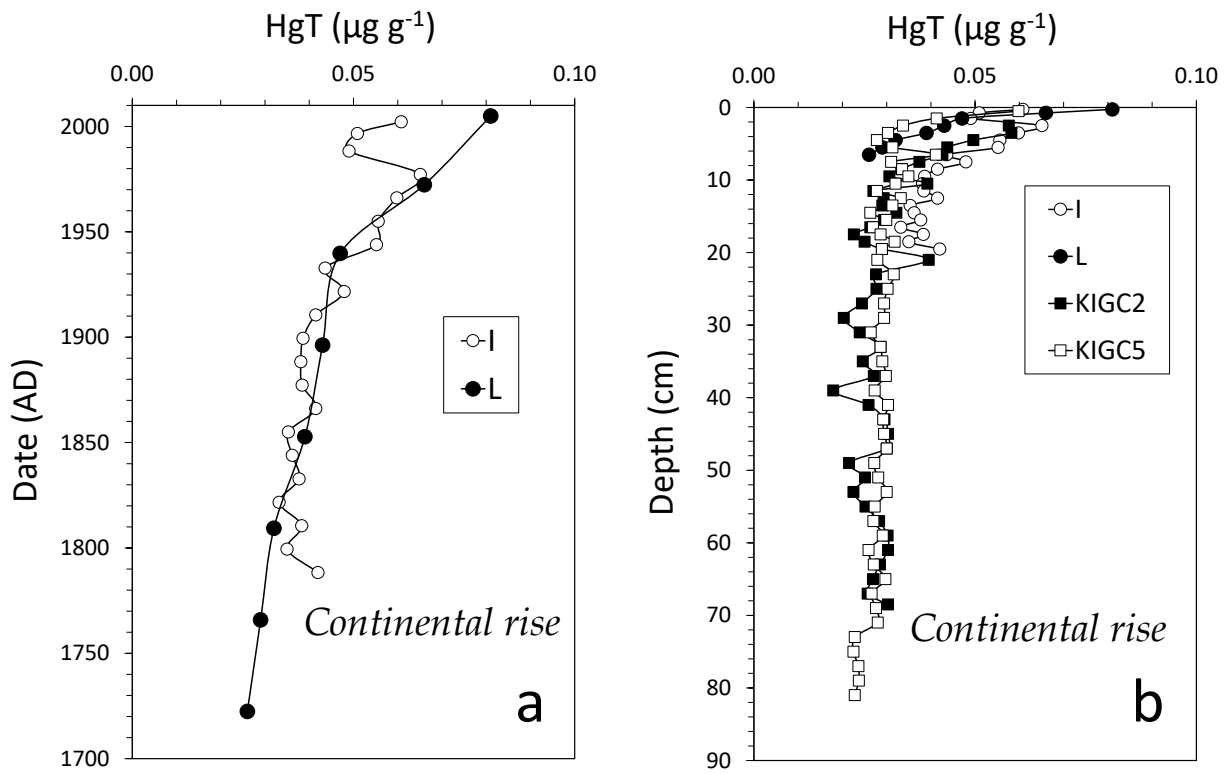
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1155 **Figure 5.**

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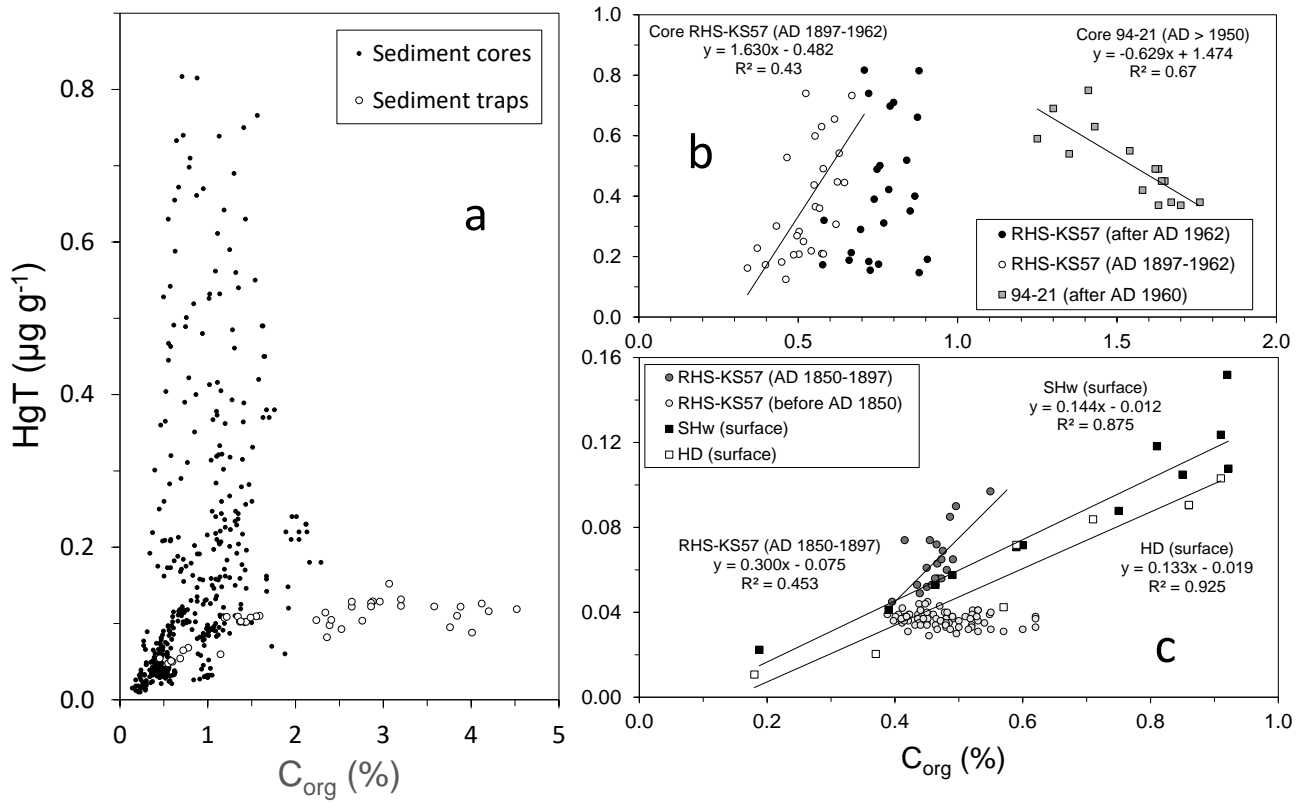
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1159 **Figure 6.**

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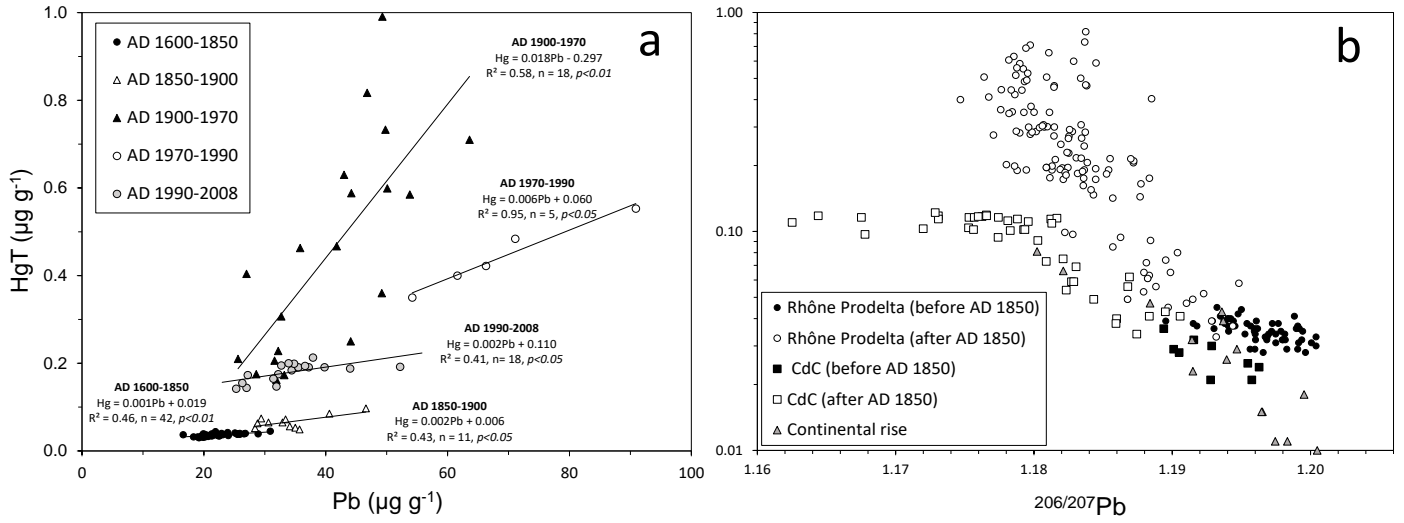
1162 **Figure 7.**

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Figure 8.

