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## Methylmercury in tailings ponds of Amazonian gold mines (French Guiana): Field observations and an experimental flocculation method for *in situ* remediation

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### Abstract:

Sites of monomethylmercury (MMHg) production in Amazonian regions have been identified in hydraulic reservoirs, lake sediments and wetlands, but tailings ponds have not yet received sufficient attention for this purpose. This work evidenced high MMHg production within the water column and the interstitial water of two tailings ponds of French Guiana Au mines located; (i) in a small scale exploitation (*Combat*) where Hg was used for Au amalgamation, and (ii) in an industrial on-going Au mine (*Yaoni*) processing without Hg. The (MMHg)<sub>D</sub> maximum (2.5 ng L<sup>-1</sup>) occurred in the oxic water column above the sediment–water interface (SWI) of the most recent tailings pond (*Combat*), where the substrate was fresh, the redox transition was sharp and the pool of total Hg was large. In the *Yaoni* pond, the (MMHg)<sub>D</sub> maximum concentration (1.4 ng L<sup>-1</sup>) was located at the SWI where suboxic conditions prevailed. Using the (MMHg)<sub>D</sub> concentration as a proxy for Hg methylation rates, the present results show that Hg methylation may occur in various redox conditions in tailings ponds, and are favored in areas where the organic matter regeneration is more active.

A 3-month long laboratory experiment was performed in oxic and anoxic boxes filled with high turbidity waters from the *Combat* Au mine to simulate tailings ponds. Slaked lime was added in an experimental set (2 mg L<sup>-1</sup>) and appeared to be very efficient for the reduction of suspended particulate matter (SPM) to environmentally acceptable concentrations. However, at the end of the experiment, large (MMHg)<sub>D</sub> concentrations were monitored under treated anoxic conditions with the (MMHg)<sub>D</sub> maximum located at the SWI above the Fe-reducing zones. No (MMHg)<sub>D</sub> was detected in oxic experiments. The use of slaked lime for SPM decantation appears to be an efficient and non-onerous process for Au miners to avoid Hg methylation in tailings ponds when it is combined with rapid drainage of the mine waters. A subsequent human intervention is however necessary for the recovery of soil structure through the cover of dried ponds with organic rich materials and reforestation to avoid the stagnation of rain waters and the occurrence of anoxia.

**Research highlights :**

► Two tailings ponds of French Guiana Au mines were studied. ► High monomethylmercury (MMHg) production was found within water column and interstitial water. ► A laboratory experiment simulating tailings ponds was performed in oxic and anoxic conditions. ► Slaked lime addition decreased suspended particles but not MMHg concentrations. ► Remediation must consider decantation, ponds drainage and reforestation to limit Hg methylation.

**Key-words :** Mercury, Methylmercury, Gold placers, Tailing ponds, Remediation

## 53 **1. Introduction**

54 Gold mining activities, located mostly in tropical regions, are responsible for more  
55 than 10% of global mercury emissions (Smodis, 2006). The local ecological concern  
56 of this anthropogenic input depends mainly on the sites and the rate of Hg  
57 transformation into its toxic methylated species, and its transfer and bioamplification  
58 as monomethylmercury (MMHg) in the aquatic food chains (Akagi et al., 1995;  
59 Boudou et al., 2005; Carmouze et al., 2001; Dolbec et al., 2000; Frery et al., 2001;  
60 Pfeiffer et al., 1993; Veiga et al., 1999; Watras, 1992; Watras et al., 1994). This  
61 process and the associated toxicological concerns for inhabitants, whose diet relies  
62 on fish, have been evidenced in the Amazon plain and the Guyana plateau by many  
63 authors (Akagi et al., 1995; Carmouze et al., 2001; Dolbec et al., 2000; Frery et al.,  
64 2001). Sites of MMHg production in these regions have been commonly identified in  
65 hydraulic reservoirs, lake sediments and wetlands (Coquery et al., 2003; Dominique  
66 et al., 2007; Guimaraes et al., 2000a; Guimaraes et al., 2000b; Kehrig et al., 2003;  
67 Roulet et al., 2001a). Most of these sites constitute biogeochemical reactors with  
68 sharp redox transitions and where large concentrations of electron acceptors  
69 (dissolved organic matter (OM), sulfates, and iron oxides) favor bacterial activity  
70 (Fitzgerald and Lamborg, 2003; Fleming et al., 2006; Lamborg et al., 2008; Muresan  
71 et al., 2008; Ullrich et al., 2001)

72 Alluvial gold mining activities, using elemental Hg for Au-Hg amalgamation, have  
73 been highlighted to be the major source of Hg for tropical environments during Au-Hg  
74 amalgamation and the burning of amalgams (Lacerda and Salomons, 1998). In  
75 addition, the release of large suspended particles, when soils are flushed to take off  
76 light particles for the concentration of heavy gold-rich fractions, is also an important  
77 diffusive source of Hg for downstream hydrosystems (Lacerda, 1997; Roulet et al.,

78 2001b). Indeed, these suspended particles have high Hg concentration since they  
79 accumulate Hg from long term *in situ* weathering of rock, natural and recent industrial  
80 atmospheric emissions (Baeyens et al., 1991; Guedron et al., 2006; Schroeder and  
81 Munthe, 1998). To avoid downstream river contamination, environmental policy  
82 requires that gold-miners concentrate mine tailings into tailing ponds for particle  
83 sedimentation. Such tailing ponds are comparable to small-scale hydraulic reservoirs  
84 since sediment stratification occurs and redox gradients can establish rapidly.  
85 Recently, several authors pointed out high methylmercury levels in Au and Hg mine  
86 tailings, which indicated that methylation of metallic Hg in the tailings can occur at  
87 significant levels, consistently with the identification of sulfate-reducing bacteria  
88 (SRB) in the same type of environments (Gray et al., 2004; Gray et al., 2006;  
89 Ikingura et al., 2006; Winch et al., 2008). However, no information is available on the  
90 possible occurrence of Hg methylation in tailing ponds themselves. In French Guiana  
91 environments, the high density of clay particles associated with OM, Fe or Al oxides  
92 (Do Nascimento et al., 2004; Guedron et al., 2006; Roulet and Lucotte, 1995) and  
93 combined with elevated temperatures (25-35°C) and a acidic pH (4-5) (Barret, 2004)  
94 favor geochemical conditions and nutrient stocks propitious for mercury bacterial  
95 methylation (Ullrich et al., 2001). Here we address the question of the location and  
96 significance of mercury methylation in mine tailing ponds from two French Guiana  
97 gold mines, the *Combat* and *Yaoni* mines, and we test the efficiency of a common  
98 waste water flocculent (slaked lime) for particles decantation and its associated  
99 impact on mercury methylation.

100

## 101 **2. Sites, material and methods**

### 102 ***2.1. Environmental settings***

103 Two tailing ponds were monitored on different gold mines located in French  
104 Guyana, a region where the climate is tropical with an annual average rainfall of 4000  
105 mm (Barret, 2004).

106 The first tailing pond (*Combat*) is located on a former goldmine site (52°23'W,  
107 4°34'N), which was exploited at the beginning of the 1950's by artisanal gold miners  
108 who used Hg for gold amalgamation (Fig. 1). Macroscopic Hg droplets were identified  
109 in these gold mined hydromorphic soils (Guedron et al., 2009). This mine is located  
110 on the 'Amina series' of the Guiana Proterozoic shield consisting primarily of dark  
111 schist facies and thin sandstones (Egal et al., 1994). This site has recently been  
112 exploited for a second time by the CMB (Boulanger Mining Company). During the  
113 exploitation, an experimental pond of 10 m long, 8 m wide and 1 to 2 m deep was  
114 dug and filled with mine water (pH 4 to 5) of around 0.1 g L<sup>-1</sup> suspended particulate  
115 matter (SPM) load. The mean total Hg concentration of the SPM was 0.90 µg g<sup>-1</sup>.  
116 The *Combat* pond was monitored during December 2006, 6 months after its filling.

117 The second tailing pond (*Yaoni*) is located on the Coralie track, in the *Yaoni* placer  
118 (52°24'W, 4°31'N), which is a large alluvial gold mine of the CMB still in exploitation  
119 (Fig. 1). This mine is located on the 'Paramaca formation' of the Guiana proterozoic  
120 shield consisting of metavolcanic rocks (Egal et al., 1994). Gold is exploited by  
121 gravimetric methods using a shaking table to segregate gold from ore sands. The  
122 tailing pond was monitored during December 2006, two years after its filling. The  
123 pond dimensions were 300 m long, 100 m wide, 1.4 to 5 m deep. The water pH  
124 ranged between 4 and 5, the SPM load was 3.3 g L<sup>-1</sup>, and the mean total Hg  
125 concentration of the SPM was 0.50 µg g<sup>-1</sup>.

## 126 **2.2. Sampling procedure**

127 Field and laboratory sample collection and treatment were performed using ultra-  
128 clean techniques (Cossa and Gobeil, 2000) and analytical methods applied for water  
129 analyses. All materials in contact with samples were acid-washed (5 days in 20%  
130 HNO<sub>3</sub> v/v, then 3 days in HCl 10 % v/v) and rinsed several times with demineralized  
131 water (Milli-Q<sup>®</sup>) before use. Polyethylene gloves were used for handling operations.  
132 Clean Teflon<sup>®</sup> (FEP) bottles were stored in double polyethylene bags until use. Acid  
133 washed peepers were degassed in a proof box during 15 days with Hg-free Ar. They  
134 were then disposed during 15 days in the tailing ponds and 3 months for the  
135 laboratory experiments. Interstitial water was extracted immediately after sampling by  
136 tipping water in the peeper cells. The sulfide-accumulating zone (SAZ) was identified  
137 with sulfide sensitive sellotape, through the formation of a surface darkening Ti-S  
138 complex (Jezequel et al., 2007). Superficial pond water samples were collected by  
139 PE-gloved-hand in Teflon<sup>®</sup> (FEP) bottles which were three times rinsed with the pond  
140 water at each sampling site prior to the sample collection. Aliquots for dissolved  
141 methyl mercury ((MMHg)<sub>D</sub>) were filtered with Sterivex<sup>®</sup>-HV 0.45 μm sterile filters in  
142 the field. After the first 50 mL had been discarded, the filtrates were collected in  
143 Teflon<sup>®</sup> (FEP) bottles (rinsed beforehand several times with filtrate), acidified (HCl 0.5  
144 % v/v, Seastar<sup>®</sup>) and kept in two polyethylene (PE) bags until analysis. In addition to  
145 samples for Hg speciation analyses, aliquot samples were collected to determine the  
146 concentration of constituent sulfides, and reduced iron. After the first 10 mL had been  
147 discarded, the filtrates were collected in Pyrex<sup>®</sup> glass bottles, rinsed beforehand  
148 several times with filtrate and analyzed in the field.

### 149 **2.3. Laboratory experiments**

150 Stream waters were sampled in the *Combat* creek within 2 L Teflon<sup>®</sup> (FEP) bottles  
151 using ultra clean techniques and frozen before use. Around 4 kg of soil samples were

152 collected from the *Combat* gold mine, several meters from the monitored *Combat*  
153 decantation pond to have similar material as in the field measurements. Collected  
154 soils were identified as the mineral horizon of a contaminated hydromorphic soil of  
155 the *Combat* gold mining flat.

156 Two boxes each containing four separated compartments of 33 cm height, 8 cm  
157 width and 20 cm length were filled with stream waters sampled in the *Combat* stream  
158 mixed with  $2 \pm 0.1$  kg of soil sample. Chemical grade  $\text{Ca}(\text{OH})_2$  (slaked lime) was  
159 added ( $2 \text{ mg L}^{-1}$ ) to the first box, with 2 compartments set in open-air (oxic  
160 compartments F1O and F2O) and two sealed compartments degassed with Hg-free  
161  $\text{N}_2$  (anoxic compartments F1A and F1B). The second box was used for controls in  
162 the same conditions without slaked lime (C1O and C2O for oxic compartments and  
163 C1A and C2A for anoxic compartments). A peeper was placed in each compartment.  
164 The atmosphere above the water in the sealed compartments was renewed with a  
165 low flux of Hg-free  $\text{N}_2$ . Both boxes were wrapped with aluminium foil to limit light  
166 penetration and algae formation, and set at room temperature ( $25 \pm 2^\circ\text{C}$ ) during ~3  
167 months (103 days). pH and conductivity were followed only in the open-air  
168 compartments during the experiment time. These measurements were performed in  
169 anoxic compartments at the beginning and the end of experiment to avoid the  
170 introduction of  $\text{O}_2$ .

#### 171 **2.4. Analytical measurements**

172 Samples were analyzed for  $[(\text{MMHg})_{\text{D}}]$  by cold vapor atomic fluorescence  
173 spectrometry (CVAFS) after conversion of all mercury species into  $\text{Hg}^0$  (Bloom and  
174 Fitzgerald, 1988), using a Tekran<sup>®</sup> (Model 2500) mercury detector. For  $(\text{MMHg})_{\text{D}}$   
175 determination we used the hydruration method described by (Tseng et al., 1998),  
176 modified by Cossa et al. (2009). These quantifications were performed after checking



177 for possible interference with internal spikes (Coquery et al., 2003). The accuracy  
178 was checked using a certified reference material (CRM) ERM-AE670 (IRMM -  
179 European Commission). The detection limit, defined as 3 times the standard  
180 deviation of the blanks ( $SD_{\text{blk}}$ ), was  $0.01 \text{ ng L}^{-1}$ . The reproducibility varied from 10%  
181 to 15% according to the concentration level.

182 Total particulate mercury ( $\text{HgT}_P$ ) was determined by cold vapor atomic absorption  
183 spectrometry after thermal decomposition of the sample using an automatic solid  
184 analyzer (Altec®, model AMA-254). The detailed procedure is described elsewhere  
185 (Cossa et al., 2003; Cossa et al., 2002). The detection limit ( $3 SD_{\text{blk}}$ ) was  $0.005 \text{ mg}$   
186  $\text{kg}^{-1}$  and the reproducibility better than 5%. The method is known as the standard  
187 method N° 7473 of the US-EPA. The accuracy of the determination for ( $\text{HgT}_P$ )  
188 concentrations were estimated using CRM 7002 (Czech Metrological Institute); the  
189 repeated analyses never exceeded the published concentration ranges ( $0.090 \pm$   
190  $0.012 \text{ } \mu\text{g g}^{-1}$ ).

191 Dissolved reduced iron ( $\text{Fe}^{\text{II}}$ ) and sulfide ( $\text{S}^{\text{II}}$ ) concentrations were measured in the  
192 field with a Hach® (model DR/850) colorimeter (methods 8146 and 8131 for  $\text{Fe}^{\text{II}}$  and  
193  $\text{S}^{\text{II}}$ , respectively). Detection limits ( $3 SD_{\text{blk}}$ ) were  $0.01 \text{ mg L}^{-1}$  for both  $\text{Fe}^{\text{II}}$  and  $\text{S}^{\text{II}}$   
194 concentration measurements.

195 Eh, pH and conductivity profiles were performed *in situ* using a Sentron® Eh meter  
196 (model Argus with probe 67597), a Heito® pH meter (model, PPD 26) and a WTW®  
197 conductimeter (model 340i), respectively.

198

### 199 **3. Results and discussion**

#### 200 ***3.1 Mercury methylation in tailing ponds - field measurements***

201 Dissolved MMHg concentrations were larger in the *Combat* tailing pond than in the  
202 *Yaoni* pond (Mean  $\pm$  SD =  $1.02 \pm 0.74$  and  $0.24 \pm 0.50$  ng L<sup>-1</sup>, range = 0.27 to 2.53  
203 and 0.01 to 1.44, respectively - Fig. 2). [(MMHg)<sub>D</sub>] in both ponds were in the range of  
204 concentrations reported in the hypolimnion waters of Petit Saut reservoir, Sinnamary  
205 basin in French Guiana (0.06 to 1.19 ng L<sup>-1</sup> - (Muresan, 2006). In both ponds, the  
206 sulfide accumulation zones (SAZ) were identified from the sediment-water interface  
207 (SWI) to the deep sediments in both profiles (Fig. 2). S<sup>II</sup> concentration measured in  
208 the field (data not shown) superimposed very well to the identified SAZ for both  
209 ponds, but concentrations were low, often close to detection limit of the method,  
210 ranging between 0.01 to 0.03 mg L<sup>-1</sup> with weak variation with depth.

211 The most striking features for the [(MMHg)<sub>D</sub>] vertical distributions were (i) the  
212 elevated concentrations in the water column especially in surface waters (0.92 and  
213 0.32 ng L<sup>-1</sup>, for *Combat* and *Yaoni* respectively) and (ii) the fact that they exceeded  
214 concentrations monitored in the sediment pore waters. The occurrence of maximum  
215 (MMHg)<sub>D</sub> concentrations located above and at the SWI for *Combat* (2.5 ng L<sup>-1</sup>) and  
216 *Yaoni* (1.4 ng L<sup>-1</sup>) pond respectively, indicates a source (production) of MMHg. In  
217 addition, these (MMHg)<sub>D</sub> peaks were located above the SAZ associated within high  
218 Eh and slightly negative Eh for *Combat* and *Yaoni* ponds respectively (Fig. 2). In the  
219 case of the *Combat* pond, methylmercury peaks occurred well above the ferri-  
220 reduction zone, and consequently above the sulfate-reducing one. The occurrence of  
221 a (MMHg)<sub>D</sub> maximum in hypoxic environment above the SWI has previously been  
222 described in marine lagoon sediments by Muresan et al. (2007). These observations  
223 suggest that the main methylation can occur disconnected from ferri-reducing  
224 bacteria (FRB) or SRB main activity sites, which are below the SWI (Fig. 2). This  
225 distribution pattern is consistent with the recent findings of active mercury

226 methylation in the oxygenated water columns of the open ocean (Cossa et al., 2009;  
227 Sunderland et al., 2009). Inorganic mercury methylation in hypoxic aquatic  
228 environment, even if the mechanism for this (these) reaction(s) are still unknown,  
229 may occur in “micro-niches” sheltering suboxic environments (Amouroux et al., 2009;  
230 Lin and Jay, 2007). The large particulate load is suggestively a great carrier for  
231 bacterial metabolism and thus for methylation in the water column, since they are  
232 mainly composed of clay size organic matter and Fe or Al oxides which are the main  
233 carrier for Hg in tropical soils (Guedron et al., 2009).

234 In the *Combat* tailing pond, a second peak ( $0.8 \text{ ng L}^{-1}$ ) was monitored in the  
235 sediment pore water around -15 cm (Fig. 2). The occurrence of this secondary peak  
236 in the reducing SAZ and below the  $\text{Fe}^{\text{II}}$  peak (-4 cm, Fig. 2) suggests that Hg  
237 methylation is likely due to FRB or SRB. This feature is similar to numerous  
238 observations in aquatic sediment (*i.e.*, (Benoit et al., 1999; Lamborg et al., 2008) and  
239 suggests an optimum sulfide concentration favoring *in situ* methylation. This  
240 hypothesis is consistent with the theory according to which the bioavailability of  
241 inorganic mercury for SRB methylation is controlled by the presence of neutral Hg-S  
242 complexes (Benoit et al., 2003; Benoit et al., 2001). According to Muresan et al.  
243 (2007), this also suggests that the sub-surface peak monitored in the *Yaoni* pond at  
244 the SWI results from the  $(\text{MMHg})_{\text{D}}$  diffusion from the sediment interior, where SRB  
245 are active, which is trapped by oxides at the very top of the sediment surface layer.

246 The difference in  $(\text{MMHg})_{\text{D}}$  distribution between the two vertical profiles as well as  
247 the larger  $(\text{MMHg})_{\text{D}}$  production in the *Combat* relative to the *Yaoni* tailing pond can  
248 be attributed to the different age and composition of the two substrates leading to a  
249 more active OM oxidation in the *Combat* pond. First, it can be argued that the  
250 *Combat* pond is the most recent and is richer in fresh substrate (including OM)

251 favoring bacteria activity. Second, the sediment consolidation and stratification is also  
252 less established than in the *Yaoni* pond facilitating water diffusion and advection.  
253 These remarks are supported by the sharp Eh transition above the SWI in the  
254 *Combat* pond in comparison to the progressive Eh transition monitored in *Yaoni* (Fig.  
255 2). Finally, according to the gold mining past of the *Combat* site (Guedron et al.,  
256 2009), the Hg enrichment of sediments and the presence of macroscopic droplets  
257 implies a larger Hg pool, which is potentially bioavailable for bacterial methylation  
258 (Dominique et al., 2007). In opposition the use of Hg in the *Yaoni* mine is restricted  
259 by the mining processes which do not use Hg for Au-Hg amalgamation and the Hg is  
260 mainly related to geogenic and atmospheric origin.

### 261 **3.2 Effect of slaked lime addition on Hg methylation in experimental tailing** 262 **ponds**

263 The results from the test of the effect on particle decantation with slaked lime  
264 addition ( $2 \text{ mg L}^{-1}$ ) were very conclusive; the SPM concentration largely decreased  
265 from  $320 \pm 40 \text{ mg L}^{-1}$  to  $27 \pm 5 \text{ mg L}^{-1}$  only a few minutes after slaked lime  
266 addition. SPM values obtained after treatment were in the range of concentrations  
267 measured in the *Combat* Creek before the beginning of the goldmining activities ( $25$   
268  $\pm 16 \text{ mg L}^{-1}$  – unpublished data) and of reported concentrations for three French  
269 Guiana creeks un-impacted by goldmining activities ( $5 - 38 \text{ mg L}^{-1}$  – Muresan et al.,  
270 (2008).

271 As expected, the slaked lime treatment induced a significant change in the  
272 chemical conditions in the experimental boxes. In the treated compartments (F10,  
273 F20, FIA and F2A), pH reached nearby 11 at the beginning of the experiment, while  
274 the untreated sediment slurry was between 4 and 5 (C10, C20, C1A and C2A).  
275 Figure 3 illustrates the progressive chemical stabilization of soil particles with water

276 during the three months of experimentation in the “open air” compartments. In both  
277 control compartments (*i.e.*, C1O and C2O), the pH progressively increased by about  
278 1 pH unit over the first 60 days then stabilized, while conductivity slightly decreased.  
279 In contrast, in both experimental compartments where slaked lime was added (*i.e.*,  
280 F1O and F2O), a sharp decrease of pH (from 10.8 to 8) and conductivity (from 380 to  
281 140) was monitored in the first days and even hours after flocculent addition (Fig. 3).  
282 Then, until the end of the experiment, pH progressively decreased to a stabilized  
283 value (7.7), while conductivity progressively increased during the same period from  
284 130 to ~ 180  $\mu\text{S cm}^{-1}$ . At the end of the experiment, pH and conductivity monitored in  
285 the water column of anoxic treated compartments were in the same range of oxic  
286 treated compartments. Eh measurements revealed an air leak in the two control  
287 compartments (C1A and C2A), thus, the following results and discussion will not  
288 consider these two so-called anoxic control compartments.

289 Large  $[(\text{MMHg})_{\text{D}}]$  were monitored in the two anoxic compartments (*i.e.*, F1A and  
290 F2A) where slaked lime was added (Mean  $\pm$  SD =  $0.65 \pm 0.84$  and  $0.56 \pm 0.36$   $\text{ng L}^{-1}$ ,  
291 range = 0.002 to 2.27 and 0.37 to 1.52, respectively for F1A and F2A - Fig 4).  
292 Sulfides were not detectable for both anoxic compartments and  $\text{Fe}^{\text{II}}$  concentration  
293 profiles exhibited very low concentrations ( $<0.05$   $\text{mg L}^{-1}$ ) in the water column and a  
294 sharp increase at the SWI to reach largest values in the sediment at - 4 cm.  
295  $[(\text{MMHg})_{\text{D}}]$  maxima in both F1A and F2A compartments were monitored on the SWI  
296 and were in the same range of  $[(\text{MMHg})_{\text{D}}]$  maxima measured in *Yaoni* and *Combat*  
297 ponds. Large  $[(\text{MMHg})_{\text{D}}]$  (Mean  $\pm$  SD =  $0.26 \pm 0.16$  and  $0.46 \pm 0.07$   $\text{ng L}^{-1}$ ,  
298 respectively for F1A and F2A) were also monitored in the water column at weak Eh  
299 values (-90mV). These large  $[(\text{MMHg})_{\text{D}}]$  in the water column may results from the  
300 desorption of MMHg from particles after slaked lime addition or from *in situ*

301 methylation. In the beginning of the experiment, the increase in pH and conductivity  
302 resulting from the  $\text{Ca(OH)}_2$  dissolution may lead to aluminium oxide particle  
303 dissolution (Hind et al., 1999) and to the release of sorbed elements, including  
304 MMHg, from the particles surface through the water column. Such Al (hydr)oxides  
305 were identified in the field for the same soils as carrier phases for Hg (Guedron et al.,  
306 2009). Nevertheless, observations made in the oxic boxes are in contradiction with  
307 the hypothesis of MMHg release through the solution since no  $(\text{MMHg})_D$  was  
308 monitored in the water column of treated oxic boxes where similar desorption may  
309 have occur (Fig 5). Thus, the hypothesis of the *in situ* methylation is more probable  
310 and we can reasonably speculate that a larger accessibility of dissolved elements  
311 (e.g., DOC, sulfates, nitrates, etc.) after slaked lime treatment would be stimulating  
312 for bacteria metabolism and furthermore for methylation under suboxic conditions as  
313 observed in lake tropical water columns below the oxycline (Coquery et al., 2003). In  
314 addition, the occurrence of  $(\text{MMHg})_D$  peaks above the  $\text{Fe}^{\text{II}}$  maxima and in absence of  
315 sulfides suggests a Hg methylation by FRB rather than SRB (Fig. 4). Nevertheless,  
316 the absence of controls under anoxic conditions limits this interpretation. The  
317 diffusion of MMHg in the water column from the sediment can also be suggested.  
318 Since laboratory measurements were done in the dark, the rates of demethylation  
319 are thus reduced in the water column since photodemethylation is impossible. In the  
320 field, demethylation would certainly be larger in the water column after slaked lime  
321 addition since the absence of particle could not limit UVB penetration.

322 In opposition to anoxic compartments,  $[(\text{MMHg})_D]$  were very low and close to the  
323 detection limit in all “open-air” experiments with and without slaked lime addition (Fig  
324 5). In parallel, Eh and  $\text{Fe}^{\text{II}}$  concentration profiles didn’t show any significant vertical  
325 pattern. Such oxic conditions are not propitious for bacterial methylation (Benoit et

326 al., 2003). In addition, as discussed previously for the anoxic boxes, at pH 11,  
327 aluminium oxide dissolution occurs, while iron(III) hydroxide do not (Hind et al., 1999;  
328 Schwertmann and Cornell, 2000). Then, when the solution reach the equilibrium (pH  
329 7.7), Al adsorbs on the various solid phases (Charlet et al., 1993) or Al- (hydr)oxides  
330 may re precipitate because of the low solubility of aluminium hydroxide below pH 9.5.  
331 The formation of such Fe and Al hydroxides, which are known to be great adsorbants  
332 for dissolved Hg and MMHg, may limit the presence of both Hg and MMHg in solution  
333 (Feyte, 2007).

### 334 ***3.3 Good practices for remediation***

335 In opposition to field observation, Hg methylation under oxic conditions was not  
336 observed in laboratory experiment with and without addition of slaked lime even after  
337 3 months of experiment. Based on these results, we can suggest that the methylation  
338 process under oxic conditions may be dependent on microbiological and  
339 geochemical parameters, which may have been modified in the lab experiments.  
340 Even if the production of MMHg under anoxic or suboxic conditions is evidenced in  
341 presence of slaked lime, the use of slaked lime as flocculent to enhance particles  
342 decantation rates have to be considered.

343 First, the rapid decrease of SPM after slaked lime addition to concentrations found  
344 in un-impacted creeks of French Guiana fits with the environmental policy  
345 requirements. Second, since the decantation yields are rapid, the drainage of  
346 decanted mine waters can also be done faster, allowing to avoid the formation of  
347 reducing conditions where MMHg production is the largest. In addition, since field  
348 observation showed that methylation occurred in the oxic water column, the particle  
349 decantation coupled with a rapid drainage of the pond may both increase

350 photodemethylation by facilitating UVB penetration and limit Hg methylation in the  
351 water column by reducing water residence time.

352 Following the drainage of the ponds two aspects have to be considered. First,  
353 even if the chemical equilibrium of treated waters is rapid allowing a rapid drainage of  
354 the pond, the final pH of treated waters is 2 to 3 pH units higher (7-8) than in stream  
355 waters (5). Thus, miners would have to dilute these treated waters with those of the  
356 mine diverted canals before it re-integers the main stream. Second, when the ponds  
357 are drained and dried, the surface compaction of thin particles limits vegetation  
358 growth and natural forest succession is effectively stalled (Bradshaw, 1997). In this  
359 case, tailing ponds are subject to be filled during intense rain event and/or to  
360 superficial erosion leading to the re-suspension of particles. Thus, the success of  
361 rehabilitation depends on human intervention to restore soil fertility, which comprises  
362 reconstitution of the physico-chemical characteristics of the soils and the recovery of  
363 biological functions (especially microbial ones - Schimann et al., 2007). Schimann et  
364 al., (2007) demonstrated that 8 years after the introduction of legumes in a gold  
365 mined site of French Guiana, microbial activities were similar to those of natural  
366 forested soils. Thus, the revegetation of ponds will increase soil drainage and limit  
367 the occurrence of anoxia in soil pore waters which maybe restrictive for Hg  
368 methylation.

369 To resume, the rehabilitation of a gold mined site must be done rapidly after the  
370 exploitation, by fast decantation with slaked lime, drainage of decanted waters and  
371 covering the dried pond with OM rich materials (e.g., organic horizons remove at the  
372 beginning of mining operations) and reforestation, to prevent MMHg production and  
373 particulate emission through superficial erosion.

374



#### 375 **4. Summary and conclusion**

376 Tailing ponds of industrial and small scale artisanal Amazonian gold mines  
377 can be assimilated to small scale biogeochemical reactors such as hydraulic  
378 reservoirs since similar geochemical conditions providing the occurrence of large  
379 dissolved methylmercury concentrations in both water column and sediment pore  
380 waters were evidenced in this study. The main feature in the comparison of both  
381 studied ponds is the large (MMHg)<sub>D</sub> concentrations measured under various redox  
382 conditions (i.e., anoxic, suboxic and (hyp)oxic conditions) in both sediment and water  
383 column of the recent *Combat* tailing pond. The age and the substratum quality of the  
384 ponds were shown to be major parameters for mercury methylation since fresh OM,  
385 Hg content, sharp redox transitions enhance bacteria metabolism as much as  
386 methylating activities.

387 Laboratory experiments to test the efficiency of slaked lime addition on  
388 particles flocculation showed that the increase of decantation yield was rapid and  
389 efficient. No methylation occurred under oxic conditions, while large (MMHg)<sub>D</sub>  
390 concentrations were measured under suboxic and anoxic conditions after slaked lime  
391 addition. It was concluded that the easiest way for gold miners to limit Hg methylation  
392 is to prevent the occurrence of reducing conditions by coupling decantation and  
393 drainage. The use of slaked lime appears to be a simple and not onerous solution in  
394 remediation process. The coupling of decantation, rapid drainage of the pond and  
395 human intervention to restore soil fertility, and reforest the site are the most effective  
396 processes to avoid Hg methylation during the mining activities.

397

398

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403

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## Figure captions

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585 Figure 1. French Guiana map and location of the studied gold mine sites.

586

587 Figure 2. Field observations. Vertical profiles of dissolved monomethylmercury  
588 ((MMHg)<sub>D</sub>), redox potential (Eh) and reduced iron (Fe<sup>II</sup>) in water column  
589 and in sediment pore water of Combat (dark triangles) and Yaoni (dark  
590 squared) decantation basins, sulfide-accumulating zone (SAZ, grey color)  
591 and sediment-water interface (0cm).

592

593 Figure 3. Laboratory experiments. pH and conductivity changes with time in oxic  
594 compartments with slake lime addition (F1O and F2O) and oxic control  
595 compartments (C1O and C2O) without flocculant.

596

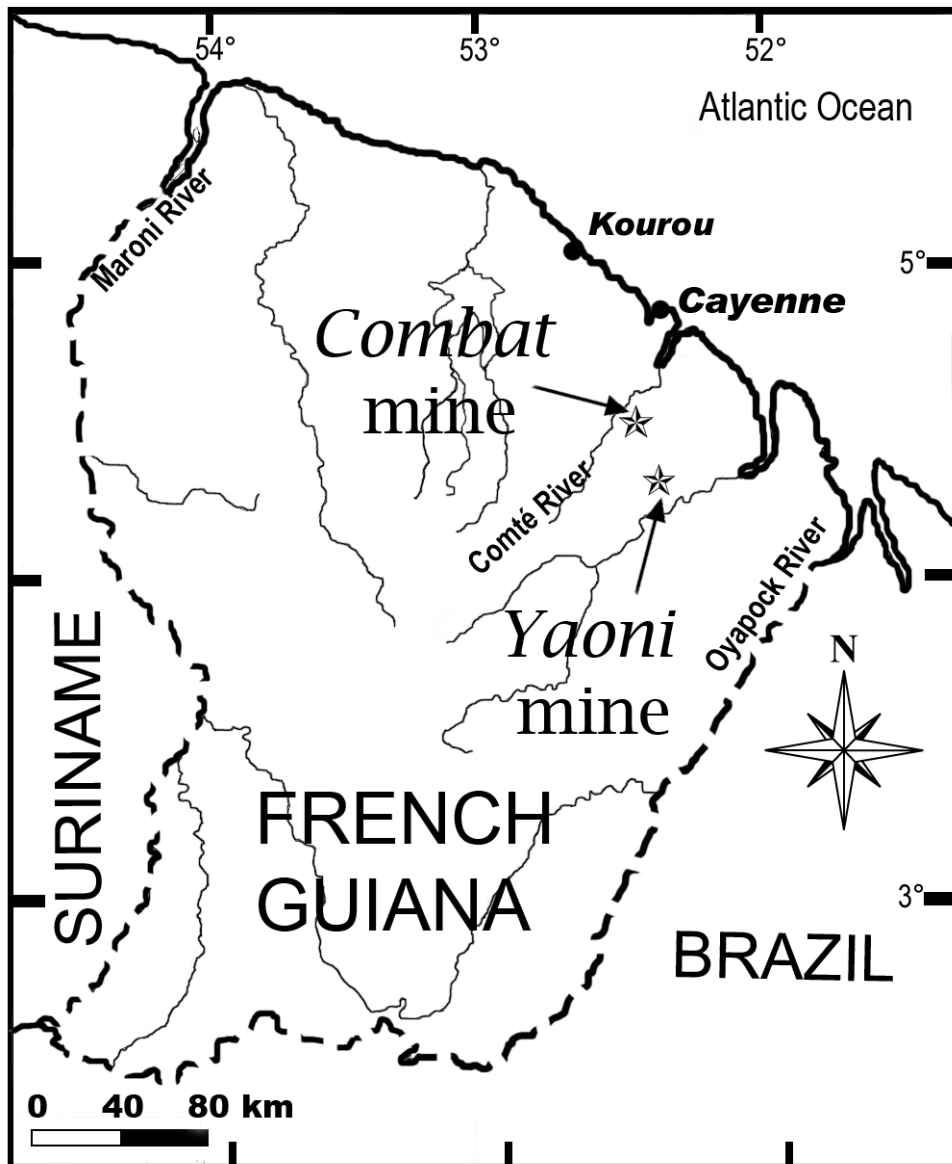
597 Figure 4. Laboratory experiments. Vertical profiles of dissolved  
598 monomethylmercury ((MMHg)<sub>D</sub>), redox potential (Eh) and reduced iron  
599 (Fe<sup>II</sup>) in water column and in sediments pore water of anoxic  
600 compartments with slake lime addition (F1A: dark diamonds and F2A:  
601 dark x-hair diamonds).

602

603 Figure 5. Laboratory experiments. Vertical profiles of dissolved  
604 monomethylmercury ((MMHg)<sub>D</sub>), redox potential (Eh) and reduced iron  
605 (Fe<sup>II</sup>) in water column and in sediments pore water of open air  
606 compartments with slake lime addition (F1O: white diamonds and F2O: x-  
607 hair diamonds) and open air control compartments (C1O: white circles  
608 and C2O: x-hair white circles) without slake lime.

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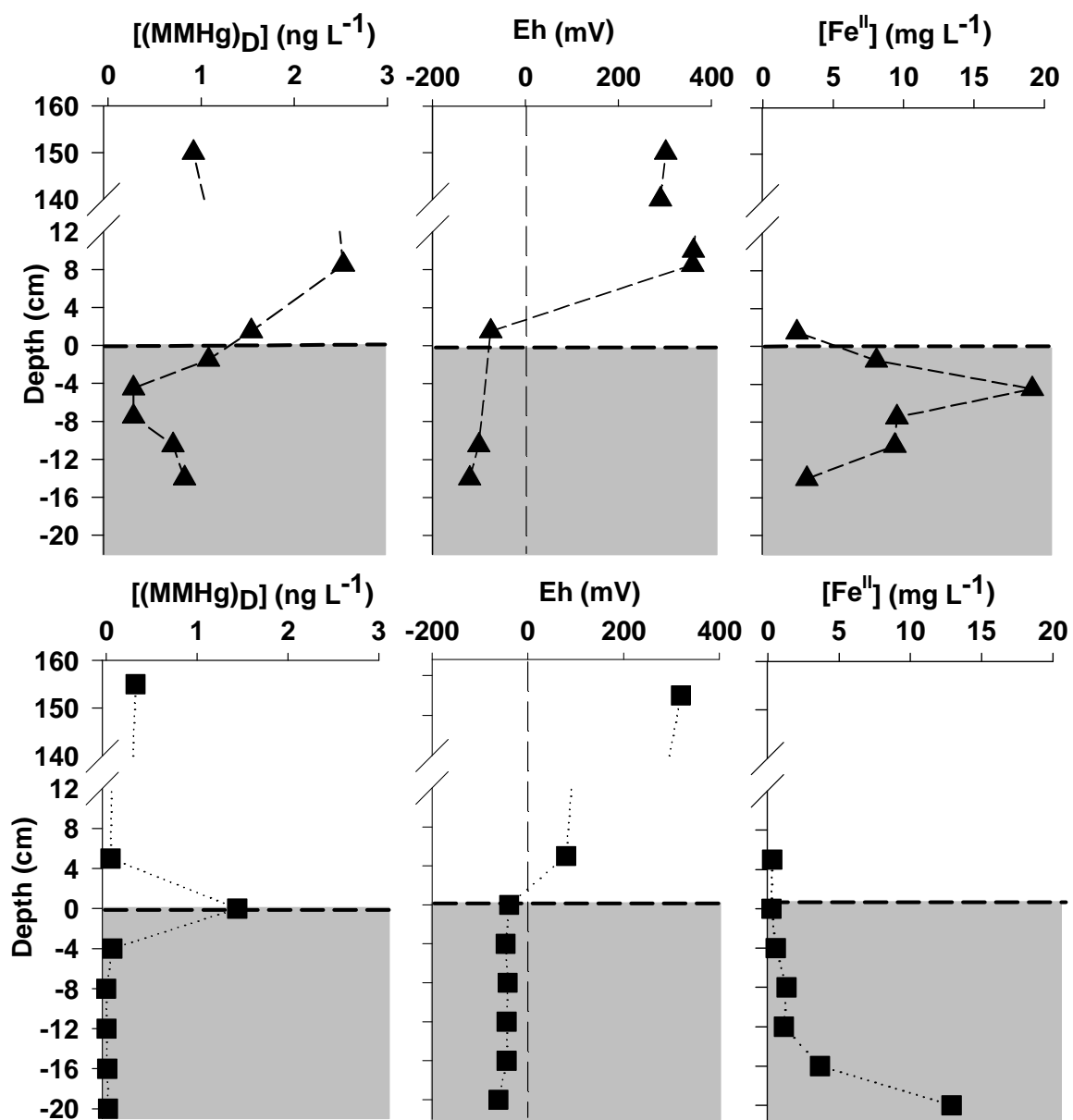
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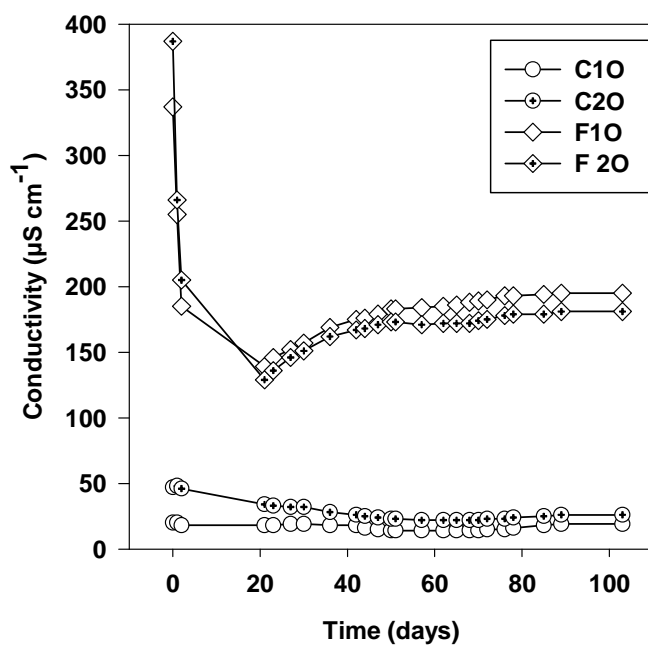
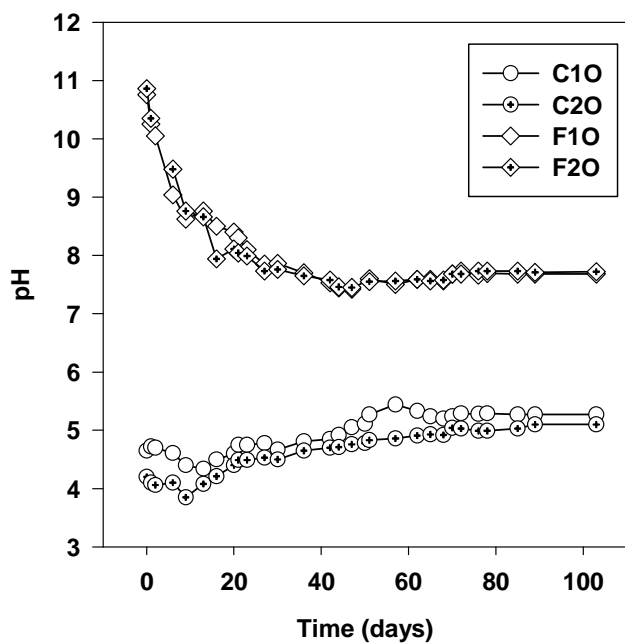


620 Fig. 2.  
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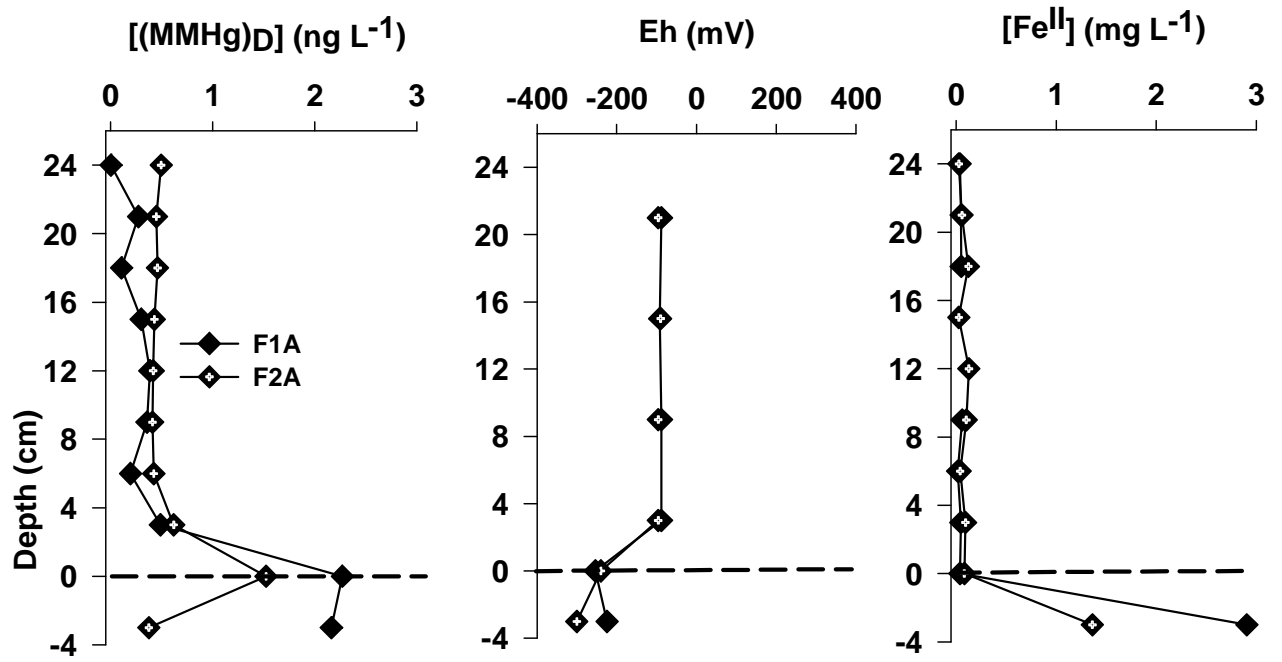
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626 Fig. 3.  
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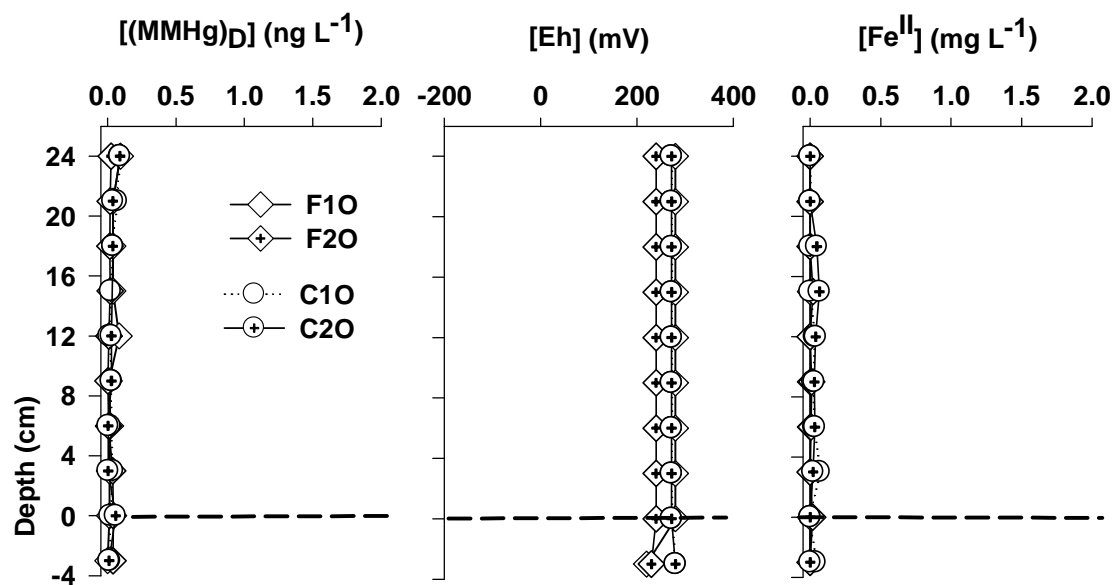
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632 Fig. 4.



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636 Fig. 5.  
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