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Spatio-temporal variability of water pollution by chlordecone at the watershed scale: what insights for the management of polluted territories?

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Abstract

Chlordecone, applied on soils until 1993 to control banana weevil, has polluted waters resources in the French West Indies for more than 40 years. At the watershed scale, chlordecone applications were not homogenous, generating a spatial heterogeneity of the pollution. The roles of climate, hydrology, soil, agronomy and geology on watershed functioning generate a temporal heterogeneity of the pollution. This study question the interactions between practices and the environment that induce such variability. We analyzed hydrological and water pollution datasets from a two years monitoring program on the Galion watershed in Martinique (French West Indies). We conjointly analyzed: i) weekly CLD concentration monitored on 3 river sampling sites, ii) aquifer piezometric dynamics and pollutions, and iii) agricultural practices on polluted soils. Our results showed that chlordecone pollution in surface waters are characterized by annual trends and infra-annual variations. Aquifers showed CLD concentration 10 times higher than surface water, with CLD concentration peaks during recharge events. We showed strong interactions between rainfall events and practices on CLD pollution requiring a systemic management approach, in particular during post cyclonic periods. Small sub-watershed with high CLD pollution, appeared to be substantial contributor to CLD mass transfers to the marine environment via rivers, and should therefore receive priority management. We suggest increasing stable organic matter return to soil as well as external input of organic matter to reduce CLD transfers to water. We identified hydrological conditions – notably drying periods – and tillage as the most influential factors on CLD leaching. In particular, tillage acts on 3 processes that increases CLD leaching: organic matter degradation, modification of water paths in soil and allophane clay degradation.

Keywords

Chlordecone, pollution, observatory, hydrology, environment, management, practices

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46 **1 Introduction**

47 The protection of surface and groundwater resources from pesticide pollutions is a remaining challenge.
48 Such question is a live issue in tropical volcanic island and especially in the French West Indies (FWI)
49 where organochlorine pesticide, namely chlordecone (CLD; $C_{10}Cl_{10}O$; CAS number 143-50-0; 491
50 $g \cdot mol^{-1}$) was used in agriculture in Guadeloupe and Martinique islands. CLD was applied from 1972 to
51 1993 to control the black banana weevils (*Cosmopolites sordidus*). Since a few decades, it pollutes soils,
52 surface waters and groundwaters in a chronic manner. CLD is a very stable pesticide that shows a very
53 slow degradation rate that is not evenly quantifiable (Cabidoche et al. 2009; Clostre et al. 2015; Devault
54 et al. 2016). Because of the very high affinity of chlordecone with soil organic matter -Koc ranging from
55 1200 to 17500 $L \cdot kg^{-1}$ (Cabidoche et al. 2009; Fernandez-Bayo et al. 2013a)-, only small fractions of the
56 soil CLD amounts are drained by infiltrated water. However, despite its low mobility, the drained
57 fraction of CLD was enough to highly contaminate other environmental compartments, such as aquifers
58 and rivers (Cabidoche et al. 2009; Cattan et al. 2019; Charlier et al. 2015; Crabit et al. 2016; Della Rossa
59 et al. 2017; Gourcy et al. 2009; Mottes et al. 2015). Several crops grown on polluted soils might as well
60 be contaminated depending on their nature (Clostre et al. 2017). Such pollution of food and water
61 exposed population to sanitary issues (Multigner et al. 2016). In order to face the problem, strategies
62 were developed to limit the exposure of the population: drinking water is treated using active carbon
63 (Ranguin et al. 2017) and decision rules were developed for cropping crops depending on their
64 sensitivity to CLD pollution from soil (Clostre et al. 2017). The development of such management
65 strategies were helpful to reduce the exposure of the population to the pesticide.

66 Chlordecone was found into aquatic coastal fauna (Dromard et al. 2018) and until pelagic cetacean
67 (Méndez-Fernandez et al. 2018). Such results suggest subsequent amount of CLD transferred into the
68 sea and raises the question of the sources, questioning the past or the present provenance of such
69 contamination. It is known that soils are still the main CLD storage (Lesueur-Jannoyer et al. 2012;
70 Levillain et al. 2012) and that CLD transfers from soils to the other environmental compartments is
71 governed by water flows (Mottes et al. 2016). To that point, CLD behavior in soils has been investigated
72 (Cabidoche et al. 2009; Clostre et al. 2015; Fernandez-Bayo et al. 2013a; Fernandez-Bayo et al. 2013b),
73 especially, the effect of the allophanic clay of andosols favoring CLD retention in soil (Woignier et al.
74 2012a), or the effect of added organic matter on CLD transfers (Woignier et al. 2012b).

75 All of these characteristics draw a complex system whose evolution is difficult to assess. On that topic,
76 Cattan et al. (2019) analyzed the spatio-temporal variability of CLD and its main metabolite (CLD-5b-
77 hydro) in FWI. Authors showed significant decreasing trends of CLD concentrations in waters on some
78 large watersheds. This is consistent with general literature on pesticides: because CLD is not applied
79 anymore, one may expect slowly decreasing concentration in the terrestrial environment. However,
80 because of the very low degradability of CLD, authors highlighted that the pluri-annual CLD decrease
81 in leaching water was likely mostly generated by CLD transfer to another compartment rather than its
82 degradation. This forces us analyzing jointly CLD concentration in the environmental compartments
83 that are related to each other in order to understand the CLD exportation from the watershed system.
84 According to the literature, several factors modify pesticides transfers at the watershed scale: climate,
85 hydrogeological context, cropping practice, soil properties as well as the interaction between these
86 factors (Lewis et al. 2016; Mottes et al. 2014; Mottes et al. 2017). Evolution of CLD concentrations in
87 rivers may also show different variabilities depending on the time and space scales considered. Previous
88 studies showed that CLD seasonal dynamics varied with the strong contributions of unpolluted water
89 from a forested upstream sub-watershed (Crabit et al. 2016). Gourcy et al. (2009) showed that
90 groundwater residence times in aquifers last from years to decades in FWI. We expect that such long
91 residence times may result in long-term evolution of CLD in rivers.

92 In this study, we aimed to characterized factors that determine CLD transfer outside the watershed
93 system. Given that CLD is mainly exported through rivers, we focused on the conjoint analysis of
94 agricultural practices and the evolution of CLD into surface and groundwater. We considered the case

95 of the Galion watershed in Martinique (FWI). This watershed is monitored to survey CLD pollutions in
96 surface and underground water compartments. First, we identified trends and variabilities of CLD
97 concentration in the different environmental compartments monitored. Second, we discussed factors that
98 may have influenced the modification of CLD concentrations in both, rivers and aquifers. Third, we
99 assess the contribution of different sub-watersheds to CLD transfers into the sea (Galion bay). Finally,
100 we propose strategies to prioritize actions and to help reduce CLD transfers into the environment.

101 **2 Materials and Methods**

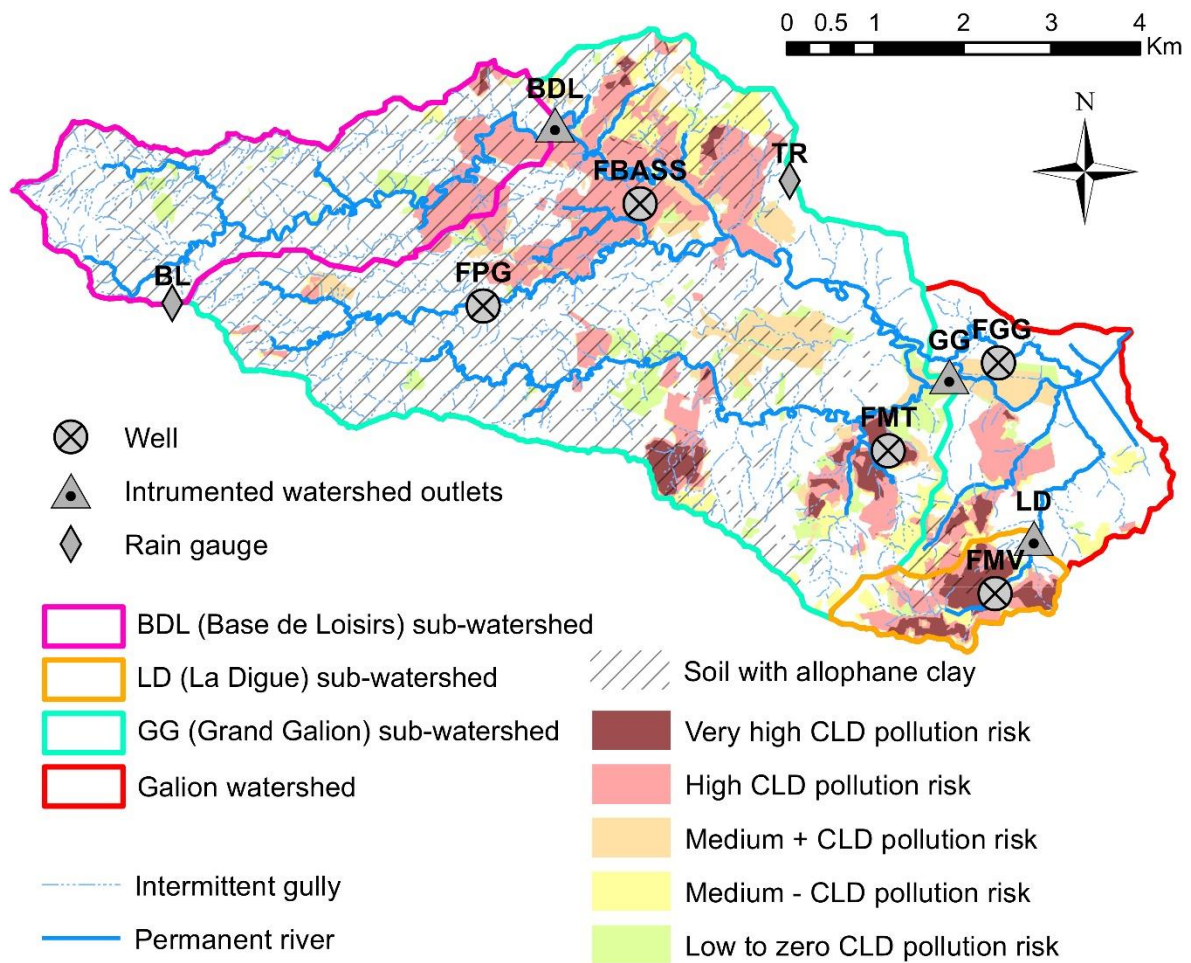
102 **2.1 Study site**

103 The Galion river watershed (61°4.4004'W/14°36.5352' N) is located on the East coast of the Martinique,
104 a volcanic island in the French West Indies. This watershed is part of the OPALE observatory
105 (Observatoire des Pollutions Agricoles aux Antilles - Observatory for agricultural Pollutions in
106 Antilles). It was selected for its representativeness of the various agropedoclimatic conditions of
107 Martinique.

108 The watershed covers 44.5 km² ranging from altitudes 0 to 694 m above sea level. The climate is humid
109 tropical with oceanic influence. Rainfall range from 1500 mm.y⁻¹ downstream up to 4000 mm.y⁻¹
110 upstream. The hydrosystem is composed of 4 major permanent watercourses and a very dense network
111 of intermittent gullies (Figure 1).

112 Steep slopes (>80%) with tropical forest and agricultural land for livestock and traditional food
113 production characterize the watershed upstream. The soils are volcanic ash soils (andosol) characterized
114 by high infiltration rates (Cattan et al. 2006) and high organic matter concentration (Dorel et al. 2000).
115 Soft slopes (~35%) cropped with banana and mixed farming on intergrade soils between andosol and
116 compact ferralsol characterize the intermediate zone. A flood plain characterize the downstream zone
117 where industrial crop productions, including banana and sugar cane, take place on ferralsols (Figure 1).

118 The bedrock is composed of volcanic formations of andesitic type (Germa et al. 2011). Geological
119 formations are thus composed by ash flows, lava flows, and reworked formations as well as atmospheric
120 fallout on a larger scale. Such geology generates a high spatial variability of lithology strata and
121 contrasting weathering levels between geological units. Consequently, volcanic aquifer's units have
122 small dimensions of a few square kilometres at most, generating complex groundwater flows according
123 to more or less permeable materials (Charlier et al. 2011; Vittecoq et al. 2015).



125

126 Figure 1: Map of the Galion watershed, positions of the 3 sub-watersheds, wells, rain gauges, risk of CLD pollution risk
 127 Pollution risks following the method described in Desprats et al. (2004) and applied by Della Rossa et al.
 128 (2017).

129 We monitored the two sub-watersheds BDL (Base de Loisirs - 8.5 km²) and LD (La Digue – 1.9 km²)
 130 in addition to the whole GG (Grand Galion) sub-watershed that covers 35.9 km² (Figure 1). The
 131 monitoring of the GG sub-watershed aims at representing the functioning of the whole Galion watershed
 132 with widely integrated flow from upstream to downstream. The BDL sub-watershed corresponds to the
 133 upstream watershed and its monitoring aims at representing the functioning of the upstream zone. The
 134 LD watershed is a small sub-watershed located on extremely polluted soils and its confluence with the
 135 Galion River is located downstream the GG outlet. The monitoring of LD aims at representing the
 136 functioning of small tributaries. LD has no direct hydrological connection with BDL or GG.

137 **2.2 Spatial variability of pollution**

138 The 3 monitored sub-watershed (GG, BDL and LD) have different characteristics in terms of risk of
 139 pollution (section 2.4.2). Two sub-watersheds have a majority of surface with low risk of pollution, with
 140 BDL being the less polluted: 86% with no risk, followed by GG (75%). LD with 29% of low risks area
 141 has a majority of its area polluted (**Erreur ! Source du renvoi introuvable.**). Figure 1 shows that on
 142 GG, the polluted areas are mainly located in the intermediate area of the sub-watershed. They are located
 143 downstream on BDL and all over the watershed on the small LD sub-watershed because of its very large
 144 rate of polluted area.

	Very high risk 1970+1980+1992	High risk (1970or1980)+1992	Medium 1992	Medium 1970	No historical banana
BDL	0.5%	11.3%	0.9%	1.1%	86.2%
GG	2.2%	14.6%	4.9%	3.4%	74.9%
LD	29.7%	27.6%	3.5%	10.5%	28.7%

145 Table 1 : Percentage of risk of contamination for each watershed (percentage of total area)

146 **2.3 Hydrological monitoring and water sampling**

147 **2.3.1 Rainfall, hydrological and hydrogeological monitoring**

148 Daily rainfall for station « Bois Lézard » (BL) and « Trinité Réservoir » (TR) were acquired from Météo
149 France. « Bois Lézard » is located upstream and « Trinité Réservoir » midstream (see Figure 1 for
150 locations). This combination of stations provided an overview of the rainfall gradient from up to
151 downstream.

152 Discharges at the watershed outlets of BDL and LD were measured at a 3 minutes time step using a
153 MS5 sensor (HYDROLAB[®]) and recorded on DuoSens stations (OTT[®]). We gauged BDL and LD using
154 current meter and Salinomadd (MADD[®]). We used the Manning-Strickler relationship based on
155 measurement of sections of the stream to estimate discharge for high water levels.

156 The direction of the environment, management and housing (DEAL) from Martinique performed the
157 GG watershed gauging and hydrological monitoring. Data are accessible through the HYDRO
158 EauFrance website (<http://www.hydro.eaufrance.fr/>).

159 Groundwater level was recorded in 5 wells (see Figure 1 for location), equipped with automatic
160 piezometric level recorders (OTT[®]): Malgré Tout (FMT) – depth = 45 m, Mont-Vert (FMV) – 15 m,
161 Bassignac (FBASS) – 50 m, Grand Galion (FGG) – 50 m and Petit Galion (FPG) – 43 m.

162 **2.3.2 Water sampling in rivers and wells**

163 We sampled water in rivers using two different methods. A first campaign was performed in 2013 by
164 sampling all river confluences within 5 days with punctual (one off) sampling. For more detail, the
165 protocol is described in Della Rossa et al. (2017).

166 The second campaign started in February 2016. We sampled water at the 3 sub-watershed outlets (BDL,
167 LD and GG) with a time dependent sampling protocol. To that purpose, each pollution measurement
168 station was equipped with an automatic sampler: ISCO 6712 (ISCO Incorporation[®]) sampler on BDL;
169 Sigma SD900 (Sigma[®]) sampler on GG and LD. Throughout each week, that lasted from Monday to the
170 next Monday unless exception, the sampling frequency of each sampler was set to sample twice 100 mL
171 every 1h15. One sample was stored in a plastic container while the other in a glass container. Both
172 containers were collected once a week meaning that each week the composite sample in each bottle
173 integrated on average 135 samples. At the end of each week, we collected the two containers containing
174 the composite samples and filled the bottles provided by the laboratory (3 glass bottles: 2 x 1 L +100
175 mL and 2 plastic bottles : 2 x 150 mL totaling 2.4 L) with the composite samples stored in the plastic
176 and glass containers. Samples were collected every weeks from 01/02/2016 to 01/02/2018 and sent in
177 ice coolers.

178 From October 2016 to March 2017, GG station was destroyed by a large storm (Matthew). During that
179 period, we performed punctual sampling each week.

180 During our sampling Campaign, French Water Office ODE also sampled water monthly at the GG
181 outlet. Punctual sampling was performed once a month and analyzed by LDA26 for pesticide
182 concentration.

183 For groundwater, sampling was based on standard NF EN ISO 5667-3, and the FD T 90-523-3 and FD
184 X31-615 guidelines. Before sampling in wells, at least three purge volumes were pumped with a

185 submersible pump until stabilization of the chemical groundwater parameters. Samples were stored at
186 5°C and shipped in ice coolers before expedition for analysis.

187 **2.3.3 Chlordecone analysis**

188 We send the aliquots to laboratory for pesticide analysis. The selected LDA26 laboratory has been
189 accredited by Cofrac, the French Accreditation Committee for pesticide analyzes providing guarantees
190 for their technical skills and reliability as well as good management practices. LDA26 complies with
191 ISO 17025 standards for testing and calibration.

192 Laboratory performed liquid/liquid extractions on samples. One liter of water sample was submitted to
193 3 extractions by using 50 mL of a mix of dichloromethane (80%) and ethyl acetate (20%). One cycle
194 was performed at pH 7 and two cycles at pH 2. Extract was concentrated to 1mL in ethyl acetate. The
195 extract was then split into twice 500 µL. 500 µL were use for GC MS and 500 µL for HPLC MS MS.
196 The HPLCC MS-MS was done on two transitions: 506.7/424.5 and 506.7/426.5 with a Sciex API 4000
197 QTRAP. Internal standards used as tracers were hexabromobenzene, tributylphosphate (added at
198 extraction step), 2,4D3, Atrazine D5 (added at injection step) in multiple residual analysis.

199 The validation was done according to NFT 90210 standards. The limit of quantification was determined
200 using six times two repetitions, the reproducibility using at least eight additions of CLD at LOQ at
201 different dates. The trueness, recovery were estimated by using six times two additions of CLD at 3
202 concentrations. Uncertainty was estimated using six times two additions of CLD.

203 A GC quality control is performed every 20 samples at LOQ. Laboratory performed an internal
204 calibration on 5 points before each analysis sequence. CLD addition on matrix and participation to inter
205 laboratory comparison were also performed to control the quality of results.

206 CLD concentrations are given by the laboratory with a $\pm 35\%$ error interval, a limit of quantification
207 (LOQ) of $0.01 \mu\text{g}\cdot\text{L}^{-1}$, a limit of detection (LOD) of $0.0033 \mu\text{g}\cdot\text{L}^{-1}$. The extraction efficiency is 89%.

208 **2.4 Agricultural practices and risks of soil pollution**

209 **2.4.1 Actual agricultural practices**

210 Surveys on farms located on the watersheds were performed between November 2016 and January 2016.
211 During surveys, decision rules for practices application and plantation dates were inquired. We analyzed
212 the evolution of fallow and crop plantations at the watershed scale using the field geographical register
213 (RPG) from 2016 and 2017 (Direction of Agriculture And Forest of Martinique - DAAF 972). These
214 georeferenced data describe agricultural land uses in 2016 and 2017 as declared by farmers.

215 **2.4.2 Mapping the risk of soil pollution**

216 The risk of soil pollution by CLD was estimated following the method described in Desprats et al. (2004)
217 and applied by Della Rossa et al. (2017) on the Galion watershed. The method rely on a retrospective
218 analysis of banana land use during the period of CLD application in Martinique (1972–1993). Soil
219 pollution risk was classified according following rule: i) Very high risk for banana plantations present
220 in 1970, 1980 and 1992, ii) High risk for banana plantations present for two out of the three years, iii)
221 Medium + for banana plantations present in 1992 only, iv) Medium - for banana plantations present in
222 1970 only, and v) No historical banana.

223 **2.5 Data analysis**

224 We compared distributions of CLD concentrations between the two years using the non-parametric
225 Wilcoxon Mann-Whitney test. In order to visualize CLD concentration trends, we applied a Hodrick-
226 Prescott filter on the data. This filter was designed to separate long-term trends to short-term variations,
227 suiting our need for pollutants dynamic analysis as previously applied by Sun et al. (2013). The best
228 captures of trend were obtained with lambda parameter: 1600×3^4 . The baseflow component of the
229 discharge time series was calculated using the one-parameter recursive digital filter proposed by Lyne

230 and Hollick (1979) implemented in the HydRun package (Tang and Carey 2017). The best capture of
 231 baseflow component of the discharge time series were acquired with parameter value of 0.8.

232 2.6 Transfers estimation

233 Chlordecone mass transferred at the outlet of each instrumented watersheds was computed for each day
 234 using the formula.

$$235 Q_{cld} = WeeklyAVGCLD * dailyDischarge * \frac{3600 \times 24}{1000} \quad (1)$$

236 With Q_{cld} the daily mass of CLD transferred at outlet (g), $WeeklyAVGCLD$, the weekly average CLD
 237 concentration in water samples ($\mu\text{g.L}^{-1}$) and $dailyDischarge$ the average daily discharge at sampling
 238 point ($\text{m}^3.\text{s}^{-1}$). The other term is a unit conversion factor.

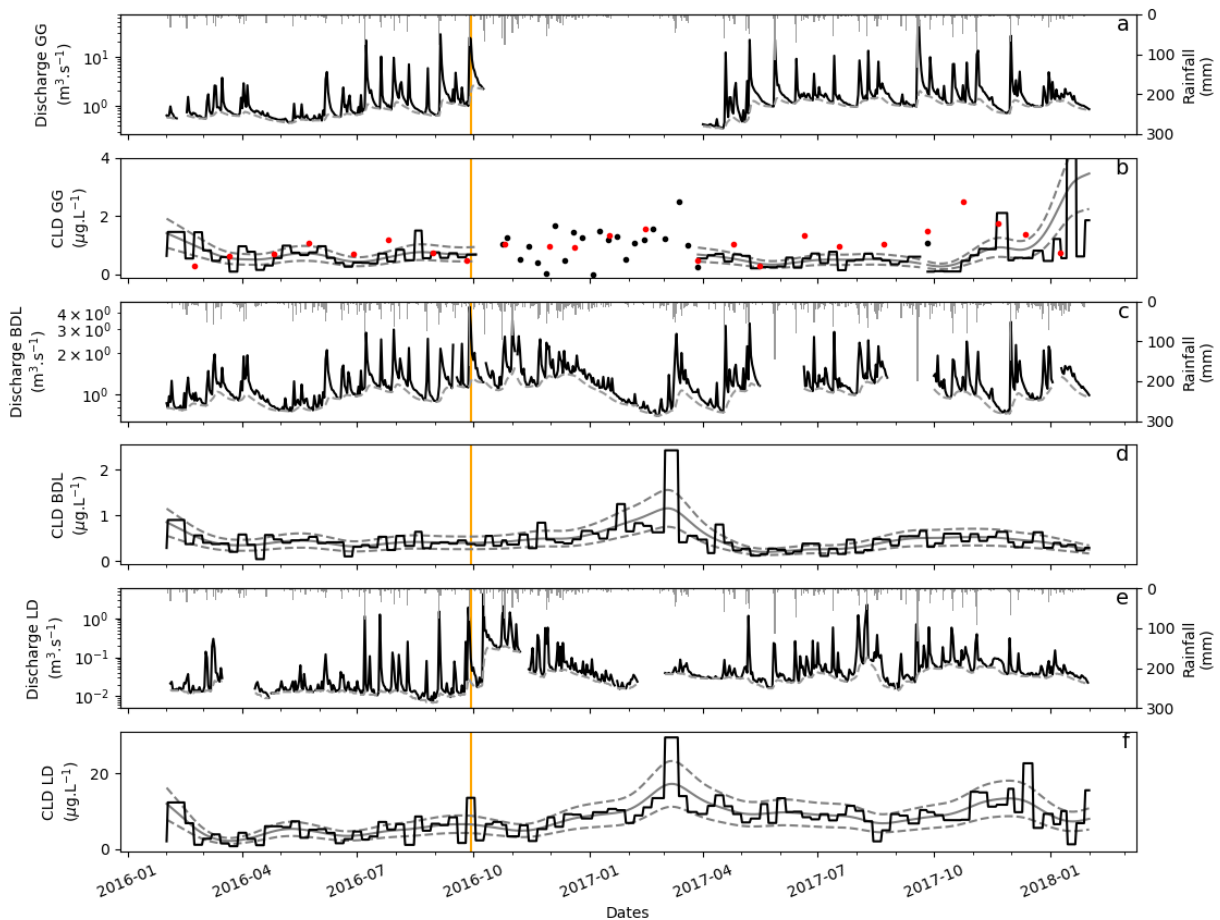
239 We calculated the daily mass of CLD transferred at outlet per unit of polluted area (classified in low,
 240 medium-, medium+, high or very high risks) by dividing Q_{cld} by the polluted surface of each sub-
 241 watershed estimated with the methodology described in 2.4.2.

242 3 Results

243 3.1 Evolutions of CLD concentration in rivers

244 3.1.1 Infra-annual evolution

245 Rainfall, discharge and CLD concentrations recorded on the 3 monitoring stations are presented in
 246 Figure 2. The proximity of rainfall and flood peaks for the 3 stations on a daily basis, illustrated that the
 247 watershed quickly reacted to rainfall events at the sub-watershed as well as the watershed scales.
 248 Reaction time of each watershed or sub-watershed was less than one day.



249

250 Figure 2 : Hydrology and chlordecone dynamics on the 3 sampling sites of the watershed. Red dots: ODE data.
251 Grey lines on CLD concentration graphics: Hodrick-Prescott filter and $\pm 35\%$ error interval. Grey dashed lines on
252 discharge graphics: baseflow. Orange vertical line: Matthew storm.

253 Figure 2d and 2f shows that the trend of the CLD concentrations were correctly assessed by the Hodrick-
254 Prescott filter at BDL and LD stations because most measurements fell within the $\pm 35\%$ interval around
255 the filtered values which correspond to the laboratory analytical uncertainty on CLD concentration
256 measurements. Deviations from the trend may also results from shorter wavelength processes, such as
257 flooding, sampling uncertainty, organic matter or sediments in samples. The dynamics highlighted by
258 the filter showed patterns with period of increasing and decreasing concentrations. The up and down
259 trends highlighted by the filter were within the same periods on BDL and LD stations. According to this
260 result, it is likely that the dynamics we observed had a common causality. We observed that short-term
261 increase of baseflow is associated with CLD concentration decrease on all stations, while short-term
262 baseflow decrease are associated with CLD increase (see between 02/2016 and 09/2016 - Figure 2).
263 During that period, CLD concentrations decrease on all station between 02/2016 and 04/2016, then it
264 increased between 04/2016 and 05/2016, and decreased again between 05/2016 and 07/2016. This
265 behavior corresponds to the hydrological functioning of the watershed and subwatersheds subjected to
266 small hydrological variations where the baseflow in not subjected to changes in regimes that last more
267 than few months. A finer analysis on the trends showed that an uptrend started few time after the
268 “Matthew” storm (29/09/2016) when baseflow decreased and ended in March 2017. The trends were
269 more difficult to assess on the GG station (Figure 2b) due to the lack of integrated data between 10/2016
270 and 03/2017 after debris transported in water few time after the Matthew storm destroyed our station.
271 In fact, punctual sampling at that time showed high values: from 12/2016 to 04/2017 in particular, 12
272 punctual samples out of 15 had CLD concentrations higher than $1.1 \mu\text{g.L}^{-1}$ while the average
273 concentration from 02/2016 to 02/2018 was $0.75 \mu\text{g.L}^{-1}$. The ODE follow up also suggested high CLD
274 concentrations during that period. Starting from June, ODE data followed a bell shaped dynamic while
275 our integrated sampling did not. In fact, concentrations measured by ODE decreased while the values
276 of our integrated measurements increased (see 10/2017 in Figure 2b). Punctual sampling results diverged
277 from the ones we obtained with integrated sampling indicating that CLD trends should not be easily
278 deduced from punctual sampling at the infra-annual temporal scale.

279 To summarize, we identified general trends in the dynamics of CLD according to our integrated
280 sampling. It showed a good consistency between stations LD and BDL that have no direct hydrological
281 connection. However, we also identified that punctual weekly samples led to record high variations of
282 concentrations. It prevents from assessing trends of CLD concentration on punctual sampling and justify
283 assessing CLD concentrations on a weekly basis.

284 3.1.2 Inter-annual evolution

285 The inter-annual dynamics on LD (Figure 2f), highlighted an increase of CLD concentration in early
286 2017. Then, until late 2017, these concentrations remained higher than in 2016 (Table 2). Comparing
287 average concentration between 2016 and 2017, we observed i) a slight increase of CLD average
288 concentration on GG watershed (+15%), ii) average concentrations on BDL that remained stable over
289 the two years, and iii) that CLD average concentration almost doubled (+82%) between 2016 and 2017
290 on the LD watershed. However, average concentrations did not reflect distribution of pollution between
291 the two years The distributions of concentrations recorded in 2016 and 2017 were significantly different
292 for all sub-watersheds, being highly significant for GG and LD sub-watersheds ($p\text{-value} < 0.001$), but in
293 a lower extent for BDL sub-watershed ($p\text{-value} < 0.05$). This result shows that, on BDL, similar average
294 concentration ($0.46 \mu\text{g.L}^{-1}$) resulted from different river behavior in 2016 and in 2017 (significantly
295 different distributions). Indeed, the trend analysis of BDL station showed that 2016 was characterized
296 by a stable CLD concentration surrounding $0.46 \mu\text{g.L}^{-1}$, while concentrations recorded during 2017 were
297 more variable with a CLD peak ($> 2 \mu\text{g.L}^{-1}$ - 03/2017) followed by an overall decrease of the
298 concentration ($< 0.46 \mu\text{g.L}^{-1}$) before returning to the initial value in 10/2017 ($0.46 \mu\text{g.L}^{-1}$) (Figure 2d).

309 Since we noticed in section 3.1.1 that LD and BDL have CLD trends showing similarities, the high
 310 increase in mean CLD concentration in 2017 for LD stemmed, on the one hand, from two CLD peaks
 311 instead of one for BDL and, on the other hand, from higher CLD contents after the first peak relative to
 312 Matthew storm. This may be explained by the fact that the stabilized increase of CLD concentration in
 313 LD in 2017 is related to a higher baseflow level than in 2016 (Figure 2e and f). The fact that this small
 314 watershed is mainly drained by subsurface flows make it more responsive to modifications of land uses
 315 for instance. Such relationship with baseflow was not observed on BDL or GG. However, it might be
 316 that the increase of CLD concentration we observed on GG between 10/2017 and 01/2018 has the same
 317 origin but delayed in time because it is 19 times larger than LD.

Sampling site	CLD Mean (2016) ($\mu\text{g}\cdot\text{L}^{-1}$)	CLD Mean (2017) ($\mu\text{g}\cdot\text{L}^{-1}$)
GG	0.70	0.81
BDL	0.46	0.46
LD	5.54	10.08

308 Table 2 : Average chlordecone concentration on the 3 sampling points for the 3 sampling campaigns (2013, 2016
 309 and 2017).

310 Monitoring CLD concentrations on the 3 stations showed that CLD is a relatively stable and chronic
 311 pollution in rivers. We showed that the CLD dynamics observed in river has two components: a trend
 312 of inter-annual evolution that may be driven by baseflow as well as infra-annual variations surrounding
 313 that trended evolution whose intensities vary according to stations and may be driven by specific event
 314 like storms in the present case.

315 3.1.3 Mass transfers

316 The average daily CLD mass transferred at the outlet of each sub-watershed, as well as the average daily
 317 CLD mass transferred per unit of area polluted by CLD are presented in Table 3. According to **Erreur !**
 318 **Source du renvoi introuvable.**, we deduced potentially polluted area on the 3 sub-watersheds: 116 ha
 319 for BDL, 901 ha for GG and 136 ha for LD, meaning that the area between GG and BDL holds 785
 320 potentially polluted hectares. The highest values of daily average CLD transferred at outlet were
 321 recorded on GG, while lowest values were recorded on BDL (Table 3).

	2016			2017		
	Daily average ($\text{g}\cdot\text{day}^{-1}$)	Daily average per unit of polluted area ($\text{g}\cdot\text{day}^{-1}\cdot\text{ha}^{-1}$)	Specific discharge ($\text{m}^3\cdot\text{s}^{-1}\cdot\text{km}^{-2}$)	Daily average ($\text{g}\cdot\text{day}^{-1}$)	Daily average per unit of polluted area ($\text{g}\cdot\text{day}^{-1}\cdot\text{ha}^{-1}$)	Specific discharge ($\text{m}^3\cdot\text{s}^{-1}\cdot\text{km}^{-2}$)
GG	91±171	0.10	0.047	137±303	0.15	0.065
LD	52±169	0.38	0.052	68±124	0.5	0.042
BDL	49±24	0.42	0.147	51±53	0.44	0.144
GG-BDL	49±159	0.06	0.018	97±266	0.12	0.027

322 Table 3 : Chlordecone mass transferred by each watershed monitored

323 Table 3 shows that sub-watersheds with larger polluted area (GG and GG-BDL) are not necessarily the
 324 one that transfers the most CLD toward our monitored outlet. BDL and LD having transfers rates by
 325 polluted area 4 times higher than GG or GG-BDL (Table 3). It means that in terms of mass, the area
 326 between BDL and GG, in spite of large potentially polluted area had a lower transfer's rate to outlet.
 327 The upstream zone (represented by BDL) is humid with large rainfall and characterized by a larger
 328 specific discharge than the other sub-watersheds (Table 3). Large water volumes at relatively low CLD
 329 concentrations generated stabilized transfers at outlet as shown by the lower standard deviation than on
 330 GG or LD (Table 3). This also means that GG and LD are more subject to large variations of daily CLD
 331 mass transferred. The small specific discharge of GG-BDL also indicate that the area between GG and
 332 BDL has a small contribution to the discharge. Our results also shows that LD, in spite of its small size

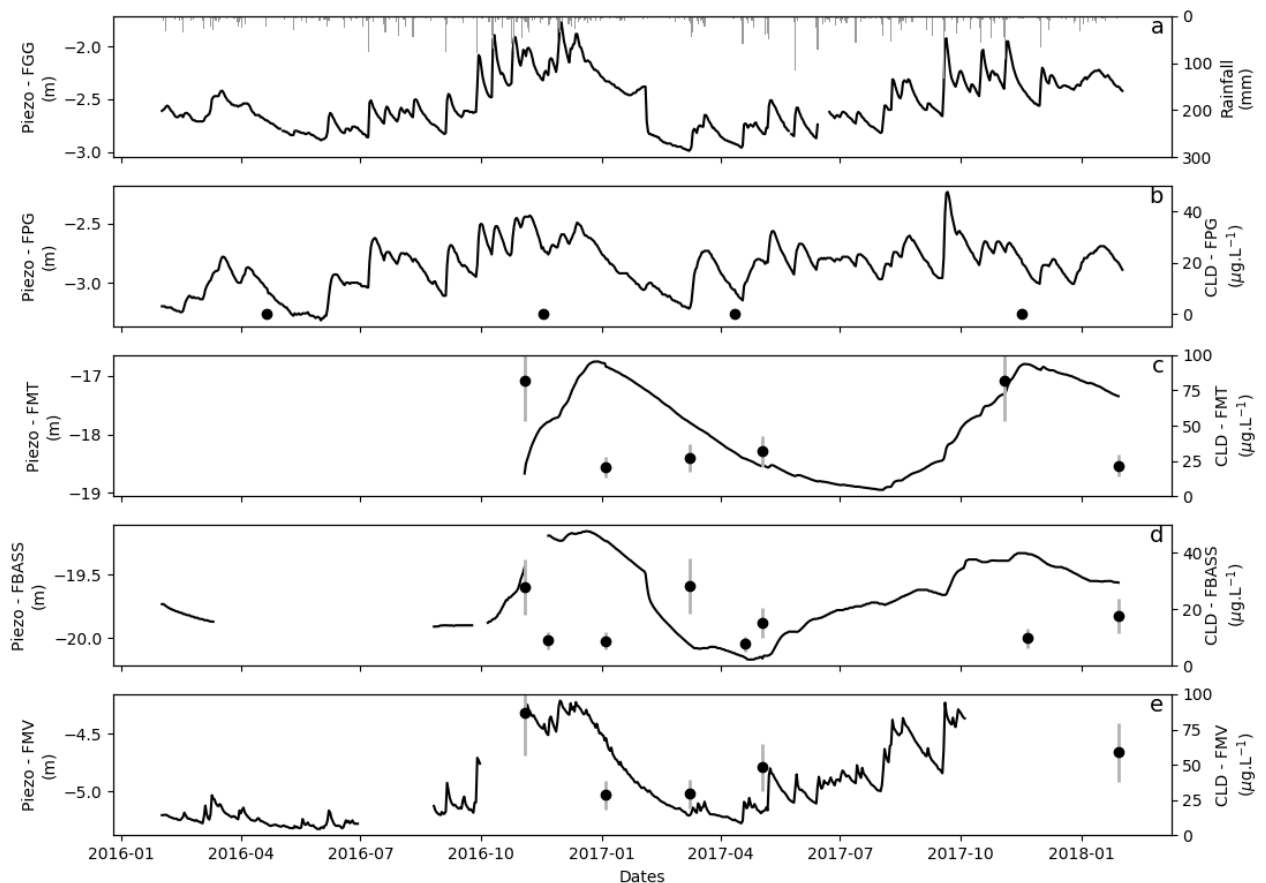
333 (191 ha) and low discharge compared to GG or BDL, transfers unneglectable mass of CLD because of
334 very high water CLD concentrations.

335 Daily mass transferred by GG gave annual values from 33 kg.y⁻¹ to 50 kg.y⁻¹ of CLD transferred to the
336 Galion Bay. For LD watershed, annual value transferred to the Galion bay ranged from 19 kg.y⁻¹ to 25
337 kg.y⁻¹. This means that the total yearly contribution of that two outlets to the Galion bay may be
338 estimated from 50 kgCLD.y⁻¹ to 75 kgCLD.y⁻¹. Using a simple assumption of constant transfer rate, and
339 without considering years of applications, we estimated total transfers to the bay from 1993 to 2018 to
340 be between 2500 kg and 3750 kg. This might be even more because we did not took into account
341 transfers from 1970 to 1993. These amounts represent between 2.4 and 3.6 kgCLD per ha of polluted
342 area. This is low compared to the amount of CLD applied ≈ 3 kg.ha⁻¹ at each application (Cabidoche et
343 al. 2009; Levillain et al. 2012) during more than 20 years. Calculations of CLD stocks in soils according
344 to Levillain et al. (2012) gave values of few kg of CLD stocked in soils in the 0-60 cm depth. This
345 suggests that, it is very likely that large amounts of CLD already transferred to other environmental
346 compartments, meaning that substantial amount of CLD could be in the marine environment and in
347 aquifers. Because CLD was applied around banana stems and banana fields were mainly cropped on
348 bare soils with stemflow at that period (Charlier et al. 2009) that conjointly favors surfaces transfers. It
349 is likely that the larger fraction of CLD that lacks in our CLD balance went mainly to the marine
350 environment during storms towards fast surface transfer processes such as runoff or subsurface flows,
351 both concentrated in surface drains that were a very common on banana field at that time.

352 We showed that small watershed can nowadays have significant chlordecone transfers contributions to
353 the ocean. In spite of the potentially large amounts of CLD that transferred in the past, we suggest that
354 such areas with fast hydrological response should be managed with care and prioritized for pesticides
355 reduction strategies as well as decontamination. We expect that the results of mitigation measures could
356 be observable in the river in a couple of years for these areas.

357 **3.2 Evolutions of CLD in groundwaters**

358 Piezometric levels on the 5 wells located on the watershed and CLD concentrations recorded in 4 of
359 these aquifers are presented in Figure 3: FPG (Fig.3b), FMT (Fig.3c), FBASS (Fig.3d), FMV (Fig.3e).
360 FBASS and FMT are deep aquifers while FGG, FPG, and FMV have piezometric levels around few
361 meters. CLD concentrations in groundwaters located in contaminated areas were 10 to 50-fold higher
362 than concentrations measured in rivers (Figure 2 and 3). No chlordecone was found in FPG waters that
363 is consistent with its location upstream the polluted areas (Figure 1). Based on the punctual data set, we
364 can say that the CLD concentrations in aquifers varied along the year, but we could not define a clear
365 relationship between CLD concentrations and piezometric levels due to the scarcity of sampling. All
366 aquifers sampled on the 03-11-2016, at the beginning of the high water level period, had particularly
367 high CLD concentrations. Then, CLD concentrations decreased in the following samples (Figure
368 3b,d,e), but remained always in a high range of values. Besides, each aquifer had its own piezometric
369 dynamic as shown in Figure 3. For instance, FMT had more inertial dynamic than FBASS or FMV, and
370 even more than FGG or FPG. According to Charlier et al. (2015), the dynamic between recharge and
371 discharge to rivers can be very long for deep aquifers with residence times from 4 years (FMV), 6 to 8
372 years (FMT) to more than 20 years (FBASS).



373

374
375

Figure 3 : Piezometric levels and CLD concentrations in aquifers of the Galion watershed. Black dots: chlordecone concentrations. Grey lines: analytical uncertainty on chlordecone concentrations.

376 3.3 Evolutions of agricultural practices

377 CLD polluted soil of the watershed are still mainly cropped with banana tree. This is because CLD is
 378 not quantified in banana fruit even when cropped on highly polluted soils (Clostre et al. 2017). CLD
 379 being stored into soil, several grower practices might affect CLD transfers. According to our banana
 380 grower survey, we could draw a general cropping system practiced by banana growers. Banana cropping
 381 system usually last around 7 years. During those 7 years, 5 or 6 years are indeed cropped with banana
 382 and the remaining 1 or 2 years consist in a fallow period. The fallow period allows for the soil to rest
 383 and eliminates diseases that developed in soil (nematodes and weevils) during the repeated banana
 384 cropping cycles. The fallow is established using herbicide injected into banana trees to kill them. Then,
 385 soil is tilled (Figure 4b). A fallow cover crop may either be planted, for instance with *C. spectabilis* or
 386 *B. decumbens*, or results from spontaneous species (such as *C. ruditosperma*). Depending on farms, at
 387 the end of the fallow period, either soil is tilled again before plantation, or banana tree are planted in a
 388 mulch of plants killed with herbicide. On the farms of the Galion watershed, tillage is usually carried on
 389 before or during the fallow period.

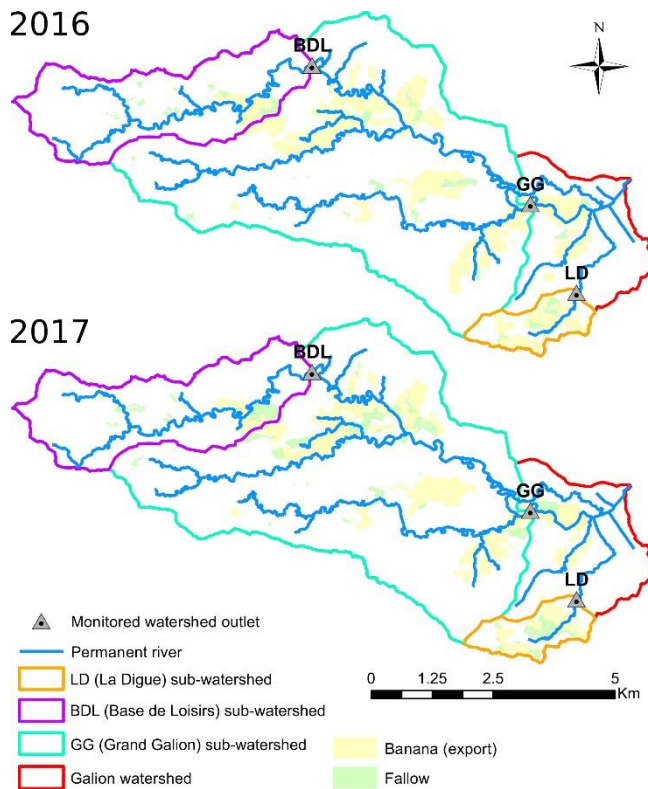
390 Figure 4a shows the field of one farmer of the LD watershed right after the Matthew storm. This farmer
 391 cut all developed banana tree in its farm (25 ha out of the 79 ha of cropped area by all farmers on the
 392 watershed) because most developed trees were laid down by the storm.



393

394 Figure 4 : a) Cycloned field just after the Matthew storm on the La Digue (LD) sub-watershed (C. Mottes
 395 Oct-2016). b) Fallow (grass with dead banana tree on the left) been tilled with excavator (bare soil) on
 396 the La Digue (LD) sub-watershed (C. Mottes Apr-2017).

397 Figure 5 shows fallows surface evolution between 2016 and 2017. We can see that 2017 was
 398 characterized by large increase of fallow area on the watershed that almost doubled: in 2016 fallow
 399 covered 79 ha whereas it covered 139 ha in 2017. This means that larger area than usual were tilled on
 400 the watershed between 2016 and 2017. This practice of large fallow integration after a cyclone or a
 401 violent tropical storm is a common practice among banana growers in the FWI resulting in large-scale
 402 modification of agricultural practices.



403

404 Figure 5: Evolution of fallow and banana between 2016 and 2017 (data from RPG 2016 and 2017 DAAF).

405 4 Discussion

406 4.1 CLD evolutions in rivers and aquifers

407 Our results showed that river and aquifer pollutions by CLD was roughly stable and chronic. Several
 408 authors have reported this (Crabit et al. 2016; Mottes et al. 2015; Mottes et al. 2017). However, CLD
 409 concentrations in rivers vary according to sites. On the small LD subwatershed, we showed that CLD
 410 interannual variability observed in river may be driven by baseflow. In the case of watersheds draining

411 the highest reliefs of volcanic islands, Crabit et al. (2016) found correlation between CLD concentration
412 and discharge because the main process of CLD evolution was the dilution of water drained from lateral
413 cultivated areas by high amounts of unpolluted waters from the forested areas upstream. In the case of
414 watersheds draining mostly cultivated zones, Mottes et al. (2016) did not find such relationship on the
415 Ravine watershed, highlighting complex conditions of CLD transfers. At the watershed scale, Della
416 Rossa et al. (2017) showed that the spatial variability of CLD concentration in water during low flow
417 periods was explained by the potential pollution of the fields located on the watershed of the sampled
418 points. At the regional scale, comparing numerous watersheds in Martinique Island, Cattani et al. (2019)
419 showed that the spatial variability of CLD concentration may be explained by a combination of the
420 location of banana cropping areas, soil and geological factors; these conditions defining a high diversity
421 of situations in terms of the mid- to long-term duration of pollution.

422 Unlike previous studies, our results highlights that, at a weekly time step, CLD concentrations in rivers
423 showed annual evolution trends (Figure 2). Aquifers located in polluted area (Figure 1) showed higher
424 CLD concentrations during recharge (Figure 3) then followed by CLD uptrends in rivers (Figure 2).
425 Figure 2 and Figure 3 analyzed conjointly showed that the uptrends we observed in rivers followed the
426 generalized CLD peak in aquifer observed in 11/2016. Here, we are in the opinion that CLD
427 concentration increased in aquifers and rivers as the consequence of, either translatory flow (Lischeid et
428 al. 2002), or recent leaching of water highly concentrated with CLD. This suggests that surrounding that
429 time, factors generated an increase of CLD leaching to aquifers and subsurface flows.
430

431 **4.2 Factors explaining increased CLD leaching**

432 Increased CLD leaching from soils, that is still a large reservoir of CLD (Lesueur-Jannoyer et al. 2012),
433 to aquifers and subsurface flows is likely to have generated the increases of CLD concentrations in
434 aquifers and rivers we observed. Several factors that increase the availability of CLD into soil to water
435 flows can explain an increase of CLD leaching.

436 **4.2.1 Effect of hydroclimatic conditions**

437 Our results showed higher CLD concentrations on all wells situated in polluted area on the watershed at
438 a specific date: 03-11-2016 but we could not conclude on a relationship with piezometric levels. This
439 pattern is coherent with previous works carried out in similar hydrogeological context in Martinique
440 Island by Arnaud et al. (2016). Based on a higher resolution of sampling (monthly), these authors
441 showed that the intensity of piezometric fluctuations was not correlated with the amplitude of variations
442 in CLD concentration, but that the CLD higher concentrations were often observed during periods of
443 rising groundwater levels. Our results suggest that period of soil drying may increase the availability of
444 CLD to water flow during the first leaching events. This is in accordance with the observations of
445 Arnaud et al. (2016), showing that the main CLD peak in groundwaters from the last decade occurred
446 during the first major recharge event after a several year period of deficient recharge that resulted in a
447 very low water table. This was true for 8 wells located all over the contaminated zones of the Martinique
448 Island, meaning that such regional pattern is likely controlled by hydroclimatic conditions. Following
449 this explanation, we expect that the main CLD higher concentrations observed in rivers of the Galion
450 watershed after the storm of October 2016 is related to the higher groundwater concentrations observed
451 at that time.

452 **4.2.2 Effects of cropping practices**

453 CLD applications were forbidden after 1993, so that theoretically no applications occurred from that
454 date. As a result, the only agricultural practices that could have increased CLD leaching are the one that
455 modified environmental characteristics that affect pesticide transfers (Mottes et al. 2014). Here,
456 modifications of the soil organic matter content, allophane clay content, or both potentially decreased
457 CLD retention by soil (Woignier et al. 2016; Woignier et al. 2012a; Woignier et al. 2012b; Woignier et
458 al. 2013). It has been shown that the peculiar hierarchical (fractal) microstructure of allophane clay traps
459 CLD (Woignier et al. 2012a; Woignier et al. 2013). It is known that tillage activate soil organic matter

460 decomposition (Haddaway et al. 2017) and expose soil to direct sunlight, hence, to strong drying at its
461 surface resulting in destruction of allophane (Quantin 1972) and allophane microstructure. During
462 drying, the fractal microstructure progressively disappears (Woignier et al. 2008). As a result, tillage
463 would act on both degradation of soil organic matter and allophanes trapping properties in andosols. On
464 top of that, tillage destroy the soil structure. The destruction of the soil structure imply a modification
465 of the water paths into soil (Alletto et al. 2010). Indeed, structured soils present preferential infiltration
466 flow that may be washed from extractible CLD after several time. Destroying those paths induce water
467 to access more porosity in soil and thus extract more CLD from soil. This may explain the difference of
468 one order of magnitude found in CLD soil organic carbon coefficients (Koc) between modelling field
469 studies (Cabidoche et al. 2009; Mottes et al. 2015) and laboratory data (Baran and Arnaud 2013;
470 Fernandez-Bayo et al. 2013a). In the last, soils are totally destroyed (Oecd 2000) while in the first one
471 structured soils are considered. Finally, tillage induces modifications of soil organic matter and clay
472 characteristics are modified as well as the soil structure. All these evolutions favor the leaching of CLD.

473 The modification of water path in soil could results from tillage but may also results from modifications
474 of soil covers. According to Lewis et al. (2016) and to Mottes et al. (2014) practices that modify ground
475 cover may modify pesticide leaching. This is particularly true for vegetative ground cover such as
476 banana tree. According to several authors, we know that banana trees concentrate rainfall along their
477 stem by a process called stemflow (Cattan et al. 2007; Charlier et al. 2009; Sansoulet et al. 2008). This
478 means that, in a cropped banana field, water will preferentially infiltrate right under banana tree (Cattan
479 et al. 2007; Sansoulet et al. 2008) generating preferential flow paths under banana tree. As for structured
480 soil, we can expect that such preferential water path are “washed” from CLD after several time of semi-
481 perennial banana cropping. Stemflow was also shown to increase runoff (Charlier et al. 2009), meaning
482 that this process also acts on reducing the overall leaching on banana fields. Consequently, stemflow
483 increases erosion of particulate CLD transfers as well as other surface pollutant transfers. To conclude,
484 it means that any modification that reduce stemflow is likely to increase leaching on the field.

485 Our results showed that between 2016 and 2017 the surface of tilled polluted fields increased (Figure 5)
486 on all monitored sub-watershed. Figure 1 shows that most soils of the watersheds contain allophane
487 clay, meaning that either tillage, dry period or both could have resulted in CLD uptrend. This means that
488 it increased the access of CLD to leaching water in 2017. Such processes could have generated
489 increasing CLD concentration in subsurface flows thus generating uptrends in river CLD concentrations
490 we observed on LD and BDL.

491 **4.3 Integration of processes at the watershed scale**

492 Our results showed that the different aquifers have contrasted piezometric inertia (Figure 3). In the
493 Galion watershed, Charlier et al. (2015) showed variable mean residence times varying from 4 years
494 (FMV), 6 to 8 years (FMT) to more than 20 years (FBASS). They also reported smaller residence times
495 of one year for sources or during high water levels. Because, rivers in FWI are mainly governed by
496 underground flows (Charlier et al. 2008; Crabit et al. 2016; Mottes et al. 2015), this means that, at the
497 watershed scale the river is likely to be fed by different aquifer’s units having different inertia. The
498 combination of residence times and the multiplicity of aquifers contribute to the stability of CLD
499 concentration at the weekly time step. According to our results along with the ones of Charlier et al.
500 2015, there is every likelihood that only underground major contributors changing conjointly may
501 modify the trend of CLD concentration in rivers. The increased CLD concentration at LD confirms this.
502 LD watershed is mainly governed by subsurface flow and the monitored aquifer (FMV) has a short mean
503 residence time of 4 years, allowing it to be sensitive, among others, to strong cyclonic events. Our results
504 showed that the mid-term CLD concentration increase in 2017 is related to the increase of the baseflow
505 on this river (Figure 2e). According to our results, we are in the opinion that increasing CLD
506 concentrations rely on the activation of new water paths on polluted areas on the sub-watershed.

507
508 Our results showed that in rivers, CLD concentration in punctual and integrated sampling diverged
509 (Figure 2b). This behavior was unexpected, because we would expect to see both monitoring programs

510 showing the same dynamics for a chronic pollution such as CLD. One explanation could be that CLD
511 concentrations show large variations with time at the event scale as it was shown for CLD (Mottes et
512 al. 2016) and for other pesticides (Liger et al. 2012). The variability of CLD concentrations we obtained
513 when we performed weekly punctual sampling instead of integrated sampling (between Oct 2016 and
514 April 2017) supports that hypothesis because CLD concentration variations are very high at that period.
515 This could mean that integrated sampling integrates the contributions of the different zones of the
516 watersheds taking into account the delayed transfers' times according to positioning on the watershed.
517 The CLD concentration trend we measured corresponds to the evolution of average concentrations that
518 integrates such variability. A variable transfer time according to positioning on the watershed was
519 suggested by Mottes et al. (2015) who included a delay in modelling CLD transfers into river according
520 to the distance of contributing areas. On large watershed, this short-term effect might be buffered at the
521 weekly time step or induce larger dynamics on CLD transfers. On that point, our results showed that the
522 spatial organization of polluted fields differed in the 3 sub-watersheds, with LD being polluted on almost
523 all its area, BDL downstream and GG mid-stream. It could be that the spatial organization of polluted
524 area affected the CLD dynamics observed at the watershed scale (Figure 2): BDL showed uptrend
525 followed by downtrend that could correspond to downstream contributions followed by upstream
526 contributions. As a result, the variation of CLD concentration of punctual and integrated sampling might
527 results from the contributions of the different transfer times of surface and subsurface flow generating a
528 dynamics at the short to mid-time scale depending on the size of the watershed.

529 CLD short terms variation could also come from the fact that sediments interact with CLD in the
530 dissolved phase, especially in the sampling containers of our sampling stations. With automatic
531 integrated samplers, erosion events are sampled along the week with water. As a result, sediments enters
532 into the sampling container. They could increase CLD concentration in the sample because of CLD
533 bound to sediments. According to ODE data, sediments analyzed at the GG station between 2008 and
534 2015 revealed CLD concentration varying between $<10 \mu\text{g.kg}^{-1}$ and $30 \mu\text{g.kg}^{-1}$. These values are low
535 compared to soil CLD concentrations ($0.1 - 5 \text{ mg.kg}^{-1}$), but could be sufficient to modify CLD
536 concentrations in sampling containers according to the concentration value measured (Table 2) (Katagi
537 2006). Finally, higher variability of punctual sampling may be related to the difficulty to follow the
538 same protocol at each sampling date. Notably sampling site may differ according to the possibilities of
539 access to the river.

540 Our results suggest either that there are variability of CLD concentrations at the instant time scale or a
541 strong interaction between dissolved and particulate phases. This means that the average concentration
542 we obtain in weekly samples results from CLD trends, variations of CLD concentrations all over the
543 week and from sediment dynamics in the river.

544 **4.4 How to manage CLD pollution ?**

545 Our results showed that causes and processes that modify CLD concentration in water, are interrelated,
546 making it difficult to isolate single effects. A complexity exists at the watershed scale where different
547 organization levels and processes interact. Agricultural practices triggered according to climatic
548 windows stand for an example of such interaction. Banana is still mainly cropped on polluted soils
549 because it is not sensitive to soil pollution by CLD. Banana farming practices follow decision rules
550 based on climate (cyclonic season, dry season), market.... Banana plantations increase in July to reach
551 good price marked at the first harvest. Fallow, are largely practiced post-cyclonic after banana trees are
552 laid down by high speed winds. Tillage is easier and recommended at medium humidity conditions
553 corresponding to conditions of the beginning or of the end of the dry season. This means that monitoring
554 of CLD in river will always show systemic response to event such as storm or cyclones. After them,
555 large recharge occurs, stemflow disappear, tillage and fallow are practiced during dryer periods, all these
556 potential causes of CLD increase have a similar triggering event, which is the storm. In a counterpart, it
557 means that we can act on several leeways at the watershed scale. In a mid-term objective, we
558 recommend to integrate knowledge on pesticides transfers at different scale into modelling tools such

559 as WATPPASS (Mottes et al. 2015) and to adapt their formalism to help farmers along with extension
560 workers and decision makers to test and assess the effect of different scenarios on CLD transfers. Test
561 the collective response of farmers at the watershed scale to specific climatic events such as storm that
562 actually induce practice response with deleterious effects on CLD concentrations.

563 Our analysis showed that processes at different scale interact. Because, we cannot act on the aquifers-
564 river relationship, we recommend focusing on limiting transfers from polluted fields. To that point, we
565 showed that hot spots, such as LD watershed are heavy polluted by CLD resulting in large CLD
566 concentrations in water. Such high concentration results in large amount transferred at outlet and
567 reaching the marine environment. At the watershed scale, we recommend to focus management of
568 transfers, on that kind of area for 3 major reasons. First, in spite of their small surface, they are large
569 contributor to pollution transfers. Second, according to our results, such small watershed react quickly
570 to practices and hydroclimatic conditions so that monitoring will be able to assess the positive impact
571 of practices change on water quality. Third, they are managed by less farmers, making it easier to
572 consistently manage all fields from the zone.

573 Decontamination of soils remains the final objective (Chaussonnerie et al. 2016; Mouvet et al. 2017). In
574 the meanwhile, in terms of actions, we showed that tillage that active different processes favoring CLD
575 transfers should be avoided as much as possible on polluted soils, or at least positioned on specific area
576 where required such as plantation lines. To avoid pollution peaks, we recommend to avoid long drying
577 of soils, in particular soils with allophanic clays. Maintaining a ground cover and a minimal controlled
578 irrigation to maintain minimal hydratation could help reduce CLD peak at the end of the dry season.

579 Sequestration method using organic matter could be applied (Clostre et al. 2014). Using farm available
580 organic matter, returning large amount of crop residues, the use of grass or covercrop to produce large
581 amount of OM returned to soil, and bringing external sources of OM from unpolluted fields could help
582 increase OM concentration that stabilize CLD into soils.

583 **5 Conclusion**

584 We showed that CLD s concentrations vary at different spatial and time scale. The large variability of
585 biophysical and decisional processes as well as their interactions at that scale require a systemic
586 approach for their understanding and management. Such systemic analysis requires a local
587 characterization of the hydrogeological functioning and of the agricultural practices to better understand
588 losses in rivers. In the French West Indies, these involves historical CLD application, soil characteristics
589 and the modifications by agricultural practices, and the reload conditions of aquifers defined by climatic
590 events and the permeability of underground geological formations modulating transfers times.

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