

Environmental fate of chlordecone in coastal habitats: recent studies conducted in Guadeloupe and Martinique (Lesser Antilles)

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Charlotte Dromard, Damien Devault, Yolande Bouchon-Navaro, Jean-Pierre Allenou, H el ene Budzinski, et al.. Environmental fate of chlordecone in coastal habitats: recent studies conducted in Guadeloupe and Martinique (Lesser Antilles). Environmental Science and Pollution Research, Springer Verlag, In press, 10.1007/s11356-019-04661-w . hal-02061180

HAL Id: hal-02061180

<https://hal.archives-ouvertes.fr/hal-02061180>

Submitted on 7 Mar 2019

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36 **Introduction**

37 The organochlorine pollution by chlordecone, an insecticide spread in the past in banana plantations, is now
38 recognized as a major ecological, economic and social crisis in the French West Indies. Used between 1972 and
39 1993 in fields of Guadeloupe and Martinique, chlordecone (commercialized as Kepone® then Curlone®) has
40 been globally banned by the Stockholm Convention on Persistent Pollutant since 2009. This molecule is
41 responsible for toxicological consequences such as an increased risk of prostate cancer, motor skills delays, an
42 increased risk of preterm birth, endocrine disruptions and reproduction impairments (Epstein 1978; Multigner *et*
43 *al.* 2010; Boucher *et al.* 2013; Kadhel *et al.* 2014; Multigner *et al.* 2016).

44 Due to its physical and chemical properties, this hydrophobic molecule has an affinity with soil organic matter
45 (partition coefficient $K_{oc} = 2500 \text{ l.kg}^{-1}$, Howard *et al.* 1981). Chlordecone has a low volatility (vapour pressure =
46 $2.5 \times 10^{-5} \text{ mm Hg}$), presents high thermodynamic stability and resistance to chemical or biological degradation,
47 involving a high persistence in the environment (Cabidoche *et al.* 2009). Consequently, it remains in the soil
48 before degradation into at least 5b-hydrochlordecone (Devault *et al.* 2016). The time to reach the depollution
49 level (at the threshold of quantification of $10 \mu\text{g.kg}^{-1}$) depends on the nature of the soils: a few decades for
50 nitisol, two to three centuries for ferralsol, five to six for andosol (Cabidoche *et al.* 2009).

51 Andosols contain amorphous clays, called allophanes, issued from the transformation of volcanic materials with
52 very specific properties. Allophanes present drastically different structures and physical properties compared to
53 usual clays, that is a very high poral volume and an important pores surface. Hygroscopic water contributed to
54 allophanes spherical form, leading them to stack in a three-dimensional fractal labyrinth in which chlordecone
55 could be trapped (Woignier *et al.* 2007). Due to its trapping ability, allophanes allow chlordecone to reaches
56 runoff and ground waters, and finally end up in the sea. Nitisols are tropical and subtropical deep (iron oxides
57 dominate), red, well-drained soils with clay content of more than 30%. Clays are more aged than andosols, *i.e.*
58 water content is less important and clays present flatten structure compared to andosols. Nitisols are well-drained
59 type of ferralsol, which are commonly low-draining and muddy due to the high clay content, and more aged than
60 andosols. Andosols and nitisols are particularly abundant in the south of Basse Terre (Guadeloupe) and in the
61 north of Martinique, in the areas where banana plantations occur (Sierra and Desfontaines 2018).

62 Indeed, in Guadeloupe, approximately 11,400 ha present a high risk of contamination that is 90% of probability
63 to find chlordecone in these areas. This area represents 25% of the land surface used for agriculture. In
64 Martinique, this value reaches 40% of agricultural lands (Direction de l'Alimentation, de l'Agriculture et de la
65 Forêt, data 2018).

66 Since 2003, several studies conducted in the Lesser Antilles have highlighted the presence of a contamination by
67 chlordecone in soil (Cabidoche *et al.* 2009), vegetables (Clostre *et al.* 2015), aquatic and marine organisms (Coat
68 *et al.* 2006; Bertrand *et al.* 2013; Coat *et al.* 2011; Bouchon *et al.* 2016; Dromard *et al.* 2016; Monti *et al.* 2016,
69 Méndez-Fernandez *et al.* 2018). However, few studies have been conducted on the fate of chlordecone between
70 agricultural fields and marine ecosystems, especially when it arrives in estuarine environments. The studies on
71 Kepone cycling in aquatic environments has been principally conducted in Virginia, because the original
72 production of Kepone began in Hopewell. In 1975, it was discovered that the Kepone factory had not only
73 exposed workers, but also severely contaminated the James River estuary (Nichols 1990; Luellen *et al.* 2006).

74 Chlordecone desorption in the James River estuary has been studied by Nichols (1990) who showed that
75 chlordecone stayed sorbed to organic particles when arriving in estuarine environment in the range of pH (7-8)
76 and salinity (0.006 to 19.5). To our knowledge, chlordecone desorption has never been studied in the Lesser
77 Antilles that present a particular context due to their tropical pedoclimatic conditions. Our first hypothesis is that
78 chlordecone could be trapped by allophanes, due to their physical structure, and stayed sorbed during its transfer
79 to the marine environment.

80 Along the James River and at the mouth of this river, contamination of aquatic fauna by chlordecone has been
81 highlighted, starting with plankton, the first link in the trophic food web. Zooplankton is broadly highly impacted
82 by this organochlorine pollution (Bahner *et al.* 1977; Luellen *et al.* 2006). Our second hypothesis is that
83 zooplankton could represent a major way of transfer between chlordecone from the water column (adsorbed on
84 terrestrial particles or dissolved) and others organisms from the trophic food-web.

85 Finally, some studies demonstrated that chlordecone passes through the different levels of the food chain and
86 shows accumulation phenomena, like many other organochlorine pollutants (Bahner *et al.* 1977). In Guadeloupe
87 and Martinique, the degree of contamination of marine organisms depends mostly on their location around the
88 two islands (Dromard *et al.* 2016). Fish and crustacean are generally more contaminated when they are located
89 downstream the contaminated watersheds and when they are close to the coast (Dromard *et al.* 2017). At the
90 same time, detritivorous and carnivorous organisms generally display higher levels of chlordecone contamination
91 than other trophic groups (Luellen *et al.* 2006; Dromard *et al.* 2017). Our third hypothesis is that chlordecone
92 concentrates in marine organisms, depending on their location (close or far from the source of pollution) and on
93 their feeding ecology.

94 The principal objective of the present study is to compile the results of recent studies on the environmental fate
95 of chlordecone from estuaries to the marine ecosystems in the Lesser Antilles, with two main objectives: 1) to
96 study chlordecone desorption as its arrival in the marine environment, 2) to evaluate the transfer of chlordecone
97 in marine trophic food web, from plants to top predators. To do so, three studies recently conducted in
98 Guadeloupe and Martinique are presented in the present work.

99

100 **Material and Methods**

101 **Experimental protocol to measure chlordecone desorption on allophanes**

102 A selected allophanic soil (6.1% w.w.) historically contaminated was sampled in Martinique and sieved on 2
103 mm-mesh stainless steel sieve. Samples of 1.5 g of soil has been incubated at dark and moderately stirred in
104 glass vial during 24h in artificial sea salted water at 0, 10, 20 and 35 psu. Artificial seawater results from a
105 mixture of Vittel® mineral water and coral reef “Instant ocean” sea salt from Aquarium Systems®. Each
106 experiment was performed in triplicates.

107 In order to estimate chlordecone adsorption on the glass of the vials, three positive controls were implemented
108 with Vittel water mixed with chlordecone up to a concentration of 0.1 µg.l⁻¹.

109 After incubation, suspended soils were filtered on Solid Phase Micro-Extraction (SPME) glass fiber filter
110 (PDMS 100 µm Merlin): soil was lyophilized, then extracted in a microwave extractor (4 ml of dichloromethane,
111 30 watts, 10 minutes) and filtered again on a glass fiber filter in order to collect the organic extract.

112 The latter was partially evaporated under 70 % agitation and 51 % vacuum (900 mb), using a Rapidvap provided
113 by Labocongo®, then reconditioned and fully evaporated under nitrogen stream. Extracts were reconstituted in 1
114 ml of acetonitrile and kept frozen until analysis.

115 Internal ¹³C chlordecone was added to filtered water (about 150 ml) then liquid-liquid extracted with 3 x 10 ml
116 of dichloromethane. Extracts, dried on Na₂SO₄, were partially evaporated under 70 % agitation and 51 %
117 vacuum (900 mb) then reconditioned and fully evaporated under nitrogen stream. Extracts were reconstituted in
118 1 ml of acetonitrile and kept frozen until analysis. Extraction recovery was respectively of 106 ± 6 % and 111 %.
119 For this experiment, solvents (dichloromethane, acetonitrile) were at least of analytical grade and provided by
120 ICS (Belin-Beliet®, France). Certified solid standards for chlordecone were obtained from Cluzeau Info Labo.
121 Concentrations of chlordecone in water and soil were measured by liquid chromatography (Agilent series 1200)
122 using in tandem a mass spectrometer detector (Agilent® 6410a) that was provided by Agilent (Santa Clara,
123 California, USA).

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125 **Samplings protocol to evaluate zooplankton contamination**

126 Samples of zooplankton and seawater were collected at the mouth of two rivers in Guadeloupe (Rivière Grande
127 Anse and Rivière du Grand Carbet) and two rivers in Martinique (Rivière Monsieur and Rivière Rouge) in
128 December 2010. A second sampling campaign was done in Guadeloupe in May 2011. These four rivers have
129 been chosen due to their high level of contamination by chlordecone. Samplings were conducted along three
130 transects oriented from the coast to the open sea, using a 500 µm-meshed net for the plankton and glass bottles
131 for seawater. On each site, nine stations were chosen to sample zooplankton and seawater (n = 3 replicates at
132 each station for each compartment). Samples were freeze-dried before analysis.

133 Quantification of chlordecone in zooplankton and seawater was conducted by the Center for Analytical Research
134 and Technology at Liege University (CART, Belgium). Lipids and chlordecone were extracted with an
135 accelerated solvent extractor using n-hexane-dichloromethane (90:10; v:v). The extracts were then dried under
136 nitrogen flow until a constant weight is obtained. 3 ml n-hexane and a surrogate marker (PCB congener 112), at
137 a concentration of 50 pg.µl⁻¹, were added. Then, the extracts underwent an acid clean-up carried out with 2 ml of
138 sulphuric acid (98-100 %) in order to eliminate organic compounds (lipids, lipoproteins,...). Finally, the eluates
139 were evaporated under an almost dry nitrogen stream. 100 µl n-hexane and 100 µl PCB 209 congener, used as an
140 internal standard at a concentration of 50 pg.µl⁻¹, were added to the samples before injection. The analyses were
141 performed by GCMS-MS (ThermoQuest ITQ 1100 ion trap). With this method, the lower limit of quantification
142 (LOQ) was 0.010 µg.kg⁻¹ w.w. for zooplankton and 0.010 µg.l⁻¹ for seawater (Monti *et al.* 2017).

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144 **Samplings protocol to evaluate trophic-food web contamination**

145 To study the contamination of marine food webs by chlordecone, samplings were conducted at Petit-Bourg in
 146 Guadeloupe, in three habitats: mangrove, seagrass beds (located at 500 m from the coast) and coral reefs (4 km
 147 from the coast), between January 2014 and February 2015. On each habitat, sediment, Superficial Sediment
 148 Layer (SSL), Suspended Organic Matter (SOM), vegetal matter (macroalgae and seagrass) and consumers from
 149 different trophic levels (primary consumers, secondary consumers, top predators) were collected. The
 150 classification of species into trophic group was done according to the literature (Randall 1967, Froese and Pauly
 151 2017). The full list of collected species is given in S1 Table. Macroalgae, fishes and crustaceans were collected
 152 by hand, spearfishing or using nets in seagrass beds and mangroves. Whenever possible, three replicates were
 153 made for each species. Each individual was rinsed, measured (total length (TL) in centimeters) and weighed
 154 (w.w. in grams). For each organism, a piece of flesh was collected and frozen (-18°C) until analyses.
 155 To sample the suspended organic matter (SOM), seawater was collected in the three habitats in acid-cleaned
 156 plastic drums. Water was then filtered on Whatman® GF/F 47 mm filters. Sediments were recovered using a
 157 corer to a depth of about 20 cm. Superficial sediment layer (SSL) was sampled on the first 2 cm of sediment.
 158 Concentrations in chlordecone were quantified by LABOCEA (Plouzané, France) with liquid chromatography
 159 coupled to mass spectrometry in tandem (UPLC-MS/MS). The lower LOQ with this method was 1 µg.kg⁻¹
 160 (w.w.) for organic material and 10 µg.kg⁻¹ (w.w.) for sediment.

161

162 **Results**

163 **Chlordecone desorption on allophanes**

164 No adsorption on the glass of the vials has been observed on controls.

165 Initial concentration of chlordecone measured in soil was 957 ± 13.2 µg.kg⁻¹ that is about 1.145 µg of
 166 chlordecone in each sample of 1.5 g of soil. After 24h of incubation, the maximal desorption of chlordecone was
 167 observed in the minimal salinity conditions (0 psu) while the minimal desorption was observed in the maximal
 168 salinity condition (35 psu) (Table 1).

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170 **Table 1** Concentrations of chlordecone ([CDC]) measured in soil after 24h of incubation. Initial concentration of
 171 chlordecone in soil was 957 ± 13.2 µg.kg⁻¹. SD: Standard Deviation; RSD: Relative Standard Deviation.

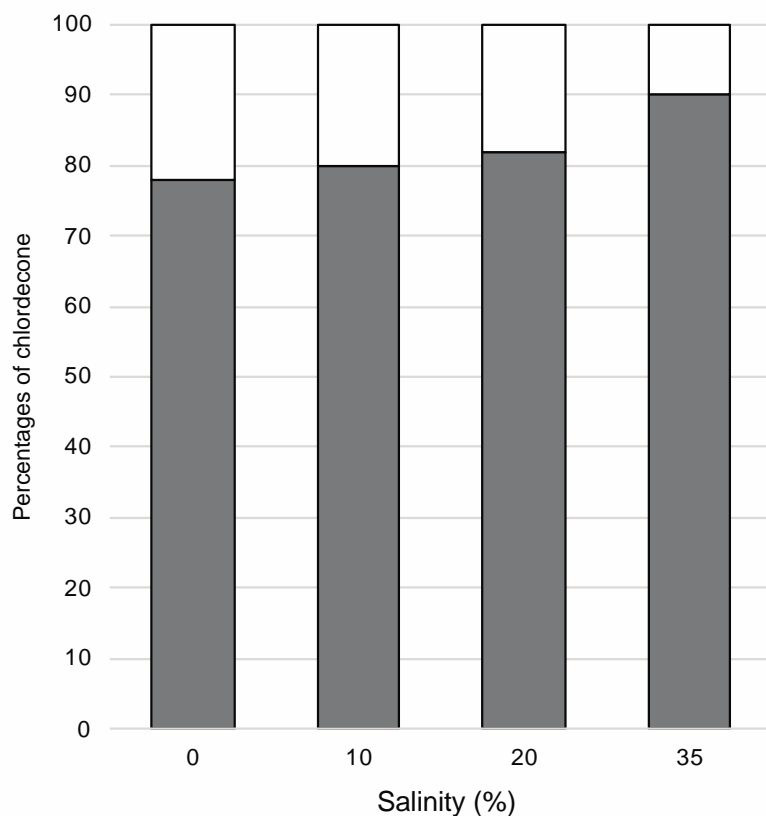
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Salinity (psu)	Replicates	[CDC]	Mean [CDC] ± SD	RSD
		µg.kg ⁻¹	µg.kg ⁻¹	%
0	1	729	788 ± 173.1	22.0
	2	983		
	3	653		
10	1	905	799 ± 92.6	11.6
	2	733		
	3	759		
20	1	821	837 ± 57.8	6.9
	2	902		
	3	790		
35	1	674	814 ± 169.2	20.8
	2	1 002		
	3	766		

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After 24h of incubation, 78 % of the initial quantity of chlordecone was still adsorbed on soil in the freshwater condition (0 psu) and 90 % for the marine condition (35 psu) (Fig. 1). About 20 % of the initial quantity of chlordecone was desorbed from soil in estuarine conditions (10 and 20 psu).



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Fig. 1 Percentages of chlordecone in water (white) and in soil (grey) related to salinity after 24h of incubation.

183 Contamination of the planktonic compartment

184 In Martinique, the minimal concentration of chlordecone found in zooplankton was 22 $\mu\text{g.kg}^{-1}$ (Rivière Rouge)
185 while the maximal concentration was measured at the mouth of Rivière Monsieur (272 $\mu\text{g.kg}^{-1}$). In Guadeloupe,
186 concentrations in chlordecone varied between 40 and 306 $\mu\text{g.kg}^{-1}$ (Fig. 2). Concentrations of chlordecone in
187 seawater were under the LOQ in 53% of the studied stations. In the other stations, concentrations varied from
188 0.01 to 0.053 $\mu\text{g.L}^{-1}$. No correlation was found between the concentration of chlordecone measured in seawater
189 and zooplankton.

190 Temporal variations in the level of contamination of zooplankton were tested in Guadeloupe. No statistical
191 significant difference was found on the level of contamination in zooplankton between samples collected at the
192 end of the dry season (May) and those collected at the end of the wet season (December), according to the two
193 sites in Guadeloupe.

194 Spatial variations in the level of contamination of zooplankton were tested between the four sites (two in
195 Martinique and two in Guadeloupe) sampled in December 2010. Mean concentrations of chlordecone in
196 zooplankton show significant spatial differences (Kruskal-Wallis, $X^2 = 13.9$, $p = 0.002$). These spatial

197 differences were globally due to the lower concentrations of chlordecone measured in zooplankton at the mouth
 198 of Rivière Rouge compared to the other sites.

199 The bioconcentration factor (BCFw), calculated as the ratio between concentrations of chlordecone measured in
 200 zooplankton and those measured in seawater, varied from 440 and 27200 (Table 2).

201
 202 **Table 2** Mean concentrations of chlordecone (min - max) measured in zooplankton (in $\mu\text{g.kg}^{-1}$ w.w.), seawater
 203 (in $\mu\text{g.l}^{-1}$) and mean bioconcentration factors (BCFw) at the mouth of four rivers. WS: end of the wet season
 204 (December), DS: end of the dry season (may), n: number of samples, “-“ indicates absence of measures.

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Islands	Rivers	Seasons	n	Zooplankton	Seawater	BCFw
Martinique	Rivière Rouge	WS	9	62.3 (22 - 99)	0.028 (0.01 - 0.05)	4320 (440 - 9900)
Martinique	Rivière Monsieur	WS	9	164.3 (79 - 272)	0.01	27 200
Guadeloupe	Grande Anse	WS	9	104 (40 - 294)	0.023 (0.02 – 0.027)	5338 (2095 - 10889)
Guadeloupe	Grande Anse	DS	9	167.8 (79.5 - 271.8)	-	-
Guadeloupe	Grand Carbet	WS	9	133.0 (50.9 - 231.7)	-	-
Guadeloupe	Grand Carbet	DS	9	164.7 (100 - 306)	0.027 (0.017 – 0.053)	7 422 (2509 - 14647)

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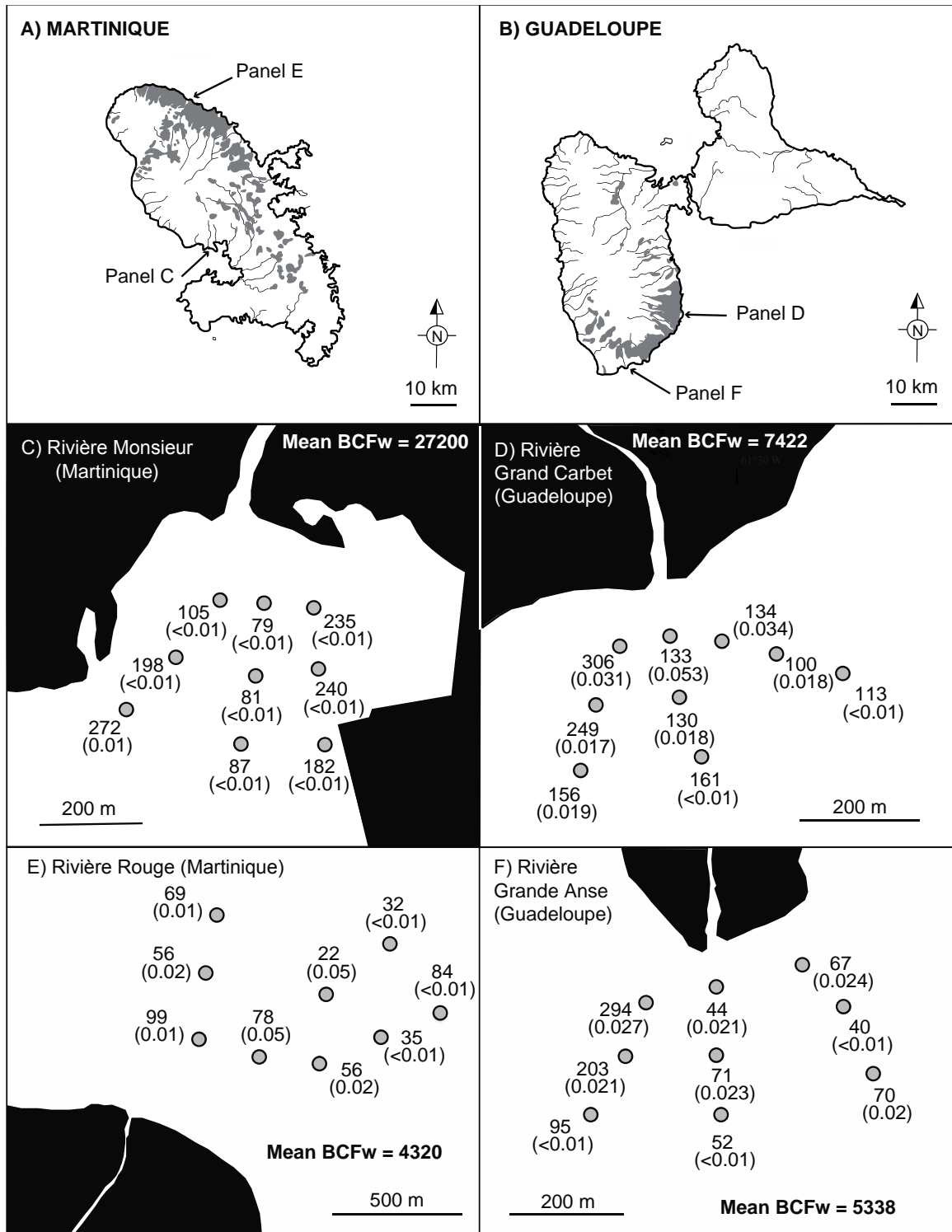
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210 Mean BCFw was maximal at the mouth of Rivière Monsieur in Martinique. The three other sites reached similar
 211 order of magnitude: 4320 at Rivière Rouge, 7422 at Grand Carbet and 5338 at Grande Anse (Fig. 2).

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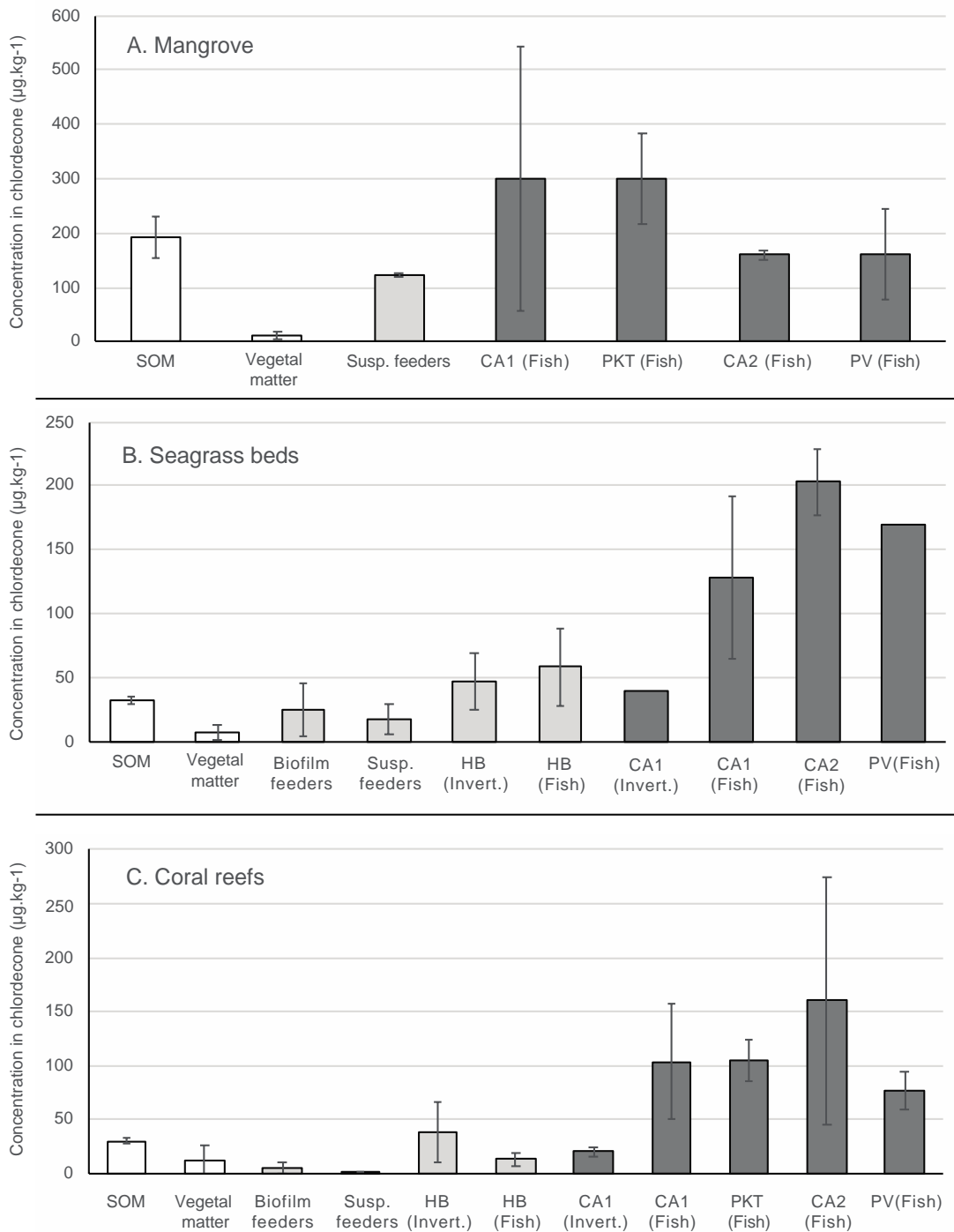
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 214 **Fig. 2** Mean concentration of chlordecone measured in zooplankton (in $\mu\text{g.kg}^{-1}$ w.w.) and seawater (into
 215 brackets, in $\mu\text{g.l}^{-1}$ w.w.) in Martinique (A) and in Guadeloupe (B) at the mouth of four rivers (C to F). BCFw:
 216 Bioconcentration factor (data from Monti *et al.* 2012); grey surfaces in A and B panels indicate soils
 217 contaminated by chlordecone.

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220 **Contamination of the trophic food web**

221 Concentrations in chlordecone were measured in different sources of matter and marine organisms in three
 222 marine habitats: mangrove, seagrass beds and coral reefs (Fig. 3).

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225 **Fig. 3** Mean concentration of chlordecone \pm SD (in $\mu\text{g.kg}^{-1}$) measured in sources of carbon (in white, SOM:
 226 Suspended Organic Matter; Vegetal matter: algal turf, macroalgae, seagrass), primary consumers (in light grey,
 227 Susp. Feeders: suspension feeders, HB: herbivores) and secondary consumers (in dark grey, CA1: invertebrate
 228 feeders, PKT: planktivores, CA2: fish and invertebrate feeders, PV: piscivores), in mangrove (A), seagrass beds
 229 (B) and coral reefs (C).

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Mean concentrations per trophic category were calculated in pooling all individuals from similar trophic group. Concentrations in chlordecone in sediment and superficial sediment layer (SL) were found under the limit of quantification ($10 \mu\text{g.kg}^{-1}$). However, chlordecone was found in SOM and vegetal matter in the different habitat. SOM exhibited the highest values among the different sources of carbon. Primary consumers, including biofilm feeders, suspension feeders and herbivorous organisms, presented intermediate mean concentrations of chlordecone from $122.3 \pm 3.8 \mu\text{g.Kg}^{-1}$ in mangrove to $17.0 \pm 22.0 \mu\text{g.Kg}^{-1}$ in coral reefs. Secondary consumers, that are carnivorous organisms (invertebrates feeders, invertebrates and fish feeders and piscivorous), showed the highest mean concentrations of chlordecone among the studied trophic categories from $232.6 \pm 172.3 \mu\text{g.Kg}^{-1}$ in mangrove to $91.9 \pm 57.6 \mu\text{g.Kg}^{-1}$ in coral reefs. In each habitat, significant differences in the mean concentrations of chlordecone were found between the food sources (SOM, vegetal matter) and the trophic categories (primary consumers and secondary consumers) (Table 3). In mangrove, multiple post-hoc comparisons tests indicated that differences of chlordecone concentrations between trophic categories are due to the low value in vegetal matter. In this habitat, SOM, primary and secondary consumers showed similar levels of contamination but were statistically different from the level in vegetal matter. In seagrass beds and coral reefs, the patterns of contamination were similar: SOM, vegetal matter and primary consumers were not statistically different according to their level of contamination. However, these three trophic categories significantly displayed lower concentrations of chlordecone than secondary consumers (Table 3).

Table 3 Mean concentrations of chlordecone (\pm SD) in $\mu\text{g.kg}^{-1}$ measured in food sources (SOM and vegetal matter), primary consumers and secondary consumers. Differences in chlordecone concentrations between the trophic categories were tested with a Kruskal-Wallis test. Letters indicate the results of multiple comparisons conducted with post-hoc tests.

Trophic categories	Mangrove	Seagrass beds	Coral reefs
SOM	191.3 ± 38.5^b	31.7 ± 2.9^a	30.3 ± 2.1^a
Vegetal matter	12.3 ± 6.4^a	6.8 ± 5.6^a	12.3 ± 13.5^a
Primary consumers	122.3 ± 3.79^b	34.6 ± 25.9^a	17.0 ± 22.0^a
Secondary consumers	232.6 ± 172.3^b	147.5 ± 66.0^b	91.9 ± 57.6^b
X ²	15.1	44.1	35.6
p values	0.002	<0.0001	<0.0001

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Discussion

In the Lesser Antilles, all the environmental compartments that have been in contact with chlordecone present a signal of contamination: agricultural soil, rivers, seawater, zooplankton and fauna from the marine food webs. In the present work, the experiments conducted to study the behavior of chlordecone during its arrival in estuarine and marine environments show that molecules stay strongly linked to the particles of terrestrial organic matter, whatever the salinity (0 to 35 psu). About 20 % of the initial quantity of chlordecone was desorbed from soil in estuarine conditions (10 and 20 psu) and 10 % was released in marine conditions (35 psu). Thus, even if furthers

265 investigations should be conducted to verify this affirmation, marine conditions seem to be the least conducive to
266 desorption. These results are in accordance with those of Nichols (1990), who demonstrated that once
267 chlordecone is sorbed it stays adsorbed in the range of pH (7-8) and salinity (0.006 to 19.5). Bakir *et al.* (2014)
268 demonstrated that salinity generally does not affect desorption rates of persistent organic pollutants (POP),
269 however their study was conducted on sorption of POP on microplastics. The capacity of POP sorption and
270 desorption on allophanes could be different. In the present study, salinity appears to limit chlordecone desorption
271 because of a process which needs further investigations. The present experiments could explain the high
272 concentrations of chlordecone measured in the suspended organic matter (SOM), collected after filtration of
273 seawater in the coastal areas (from 28 to 233 $\mu\text{g.kg}^{-1}$). Chlordecone was not encountered in superficial sediment
274 layers and marine sediments cores (all samples presented concentrations inferior to the limit of quantification: 10
275 $\mu\text{g.kg}^{-1}$). These results could be due to the fact that some samples of sediments collected in the present study
276 were poor in organic matter (for example, less of 2 % of OM in sediments collected in coral reefs). Indeed, the
277 level of contamination of sediments is generally associated with the nature of sediment: organic-rich sediment
278 has much greater capacity to sorb chlordecone than organic-poor sand or kaolinite (O'Connor and Connolly
279 1980). Bodiguel *et al.* (2011) and Robert (2012) also found a very low level of contamination for these two
280 compartments in a bay in Martinique (22 samples, all were $< 0.5 \mu\text{g.kg}^{-1}$). In 2005, Bocquené and Franco found
281 a disparity between the concentrations of chlordecone measured in sediment ($< 10 \mu\text{g.kg}^{-1}$) and SOM (52 and 22
282 $\mu\text{g.kg}^{-1}$). Chlordecone can be transferred to estuaries and the marine environment in particulate form (Crabit *et*
283 *al.* 2016) *via* SOM to which it is firmly attached. This suspended matter does not seem to settle on the bottom
284 directly when it arrives at sea, since the sediment compartment is very little affected by the contamination.
285 Several hypothesis could explain these observations: 1) contaminated SOM is constituted by very fine and low
286 density particles which may settle far from estuarine and coastal areas (but this explanation cannot be plausible
287 in semi-enclosed bays), 2) contaminated SOM may be directly consumed in the water column by planktonic
288 compartment upon arrival in the marine environment, 3) contaminated SOM may settle in estuaries and coastal
289 areas but may be rapidly degraded by microorganisms in the sedimentary compartment, 4) samples were
290 collected during the wet season, which can induce intense rainfall and disturbances of estuarine and coastal
291 waters, preventing sedimentation of contaminated SOM (Eggleton and Thomas 2004; Noegrohati *et al.* 2008).
292 Levels of contamination of zooplankton compartment were investigated to verify the second hypothesis on
293 chlordecone pathway (see above). Zooplankton is closely linked and in direct contact to SOM in estuarine and
294 coastal waters. Indeed, previous studies have highlighted the sensibility to this compartment towards
295 organochlorine pollutants, probably due to its lipid content (Jordan 1979; Day 1990; Nichols 1990). Nichols
296 (1990) measured a mean concentration equal to 4 800 $\mu\text{g.kg}^{-1}$ in zooplankton from the James River estuary. Coat
297 *et al.* (2011) reported a mean concentration equal to 5 100 $\mu\text{g.kg}^{-1}$ in the river mouth and 3 500 $\mu\text{g.kg}^{-1}$ in the
298 coastal waters of Guadeloupe. In the present study, concentrations of chlordecone measured in zooplankton were
299 considerably lower and varied between 22 and 306 $\mu\text{g.kg}^{-1}$. Concentrations of chlordecone in zooplankton also
300 spatially varied. The lowest concentrations were observed in front of Rivière Rouge, probably due to the
301 exposure of the site to swell on this Atlantic coast. No temporal variations of chlordecone concentrations were
302 observed between zooplankton collected at the end of the wet season and those collected at the end of the dry
303 season. However, the number of samples was relatively low and samples were taken in the same day during each
304 campaign. Because the planktonic compartment displays very short turn-over, further sampling effort should

305 highlight temporal variations in the level of chlordecone contamination of zooplankton. Bioconcentration factors
306 (BCF_w) were calculated as the ratio between concentrations of chlordecone in zooplankton and those in
307 seawater. The highest BCF_w was found at the mouth of Rivière Monsieur, probably due to its high degree of
308 confinement (semi-enclosed bay). Zooplankton is a keystone component of both marine and rivers systems. It
309 represents a link between invertebrates and fish by providing food and by recycling essential nutrients through
310 feeding on living and detrital material. The accumulation of organochlorine pollutants in zooplankton is a severe
311 threat to ecosystems and a potential way of transfer of the molecule along marine and freshwater food webs.

312 Bioaccumulation phenomena of chlordecone have been demonstrated experimentally (Bahner 1977) and in the
313 field (Dromard *et al.* 2018). Two processes of bioaccumulation were demonstrated in the present study. In
314 mangrove, the ambient level of contamination is high (SOM: 191.3 µg.kg⁻¹) and this value from the basis of the
315 trophic food web is not statistically different from the other trophic categories (excepted with vegetal matter).
316 Indeed, in mangrove, the entire trophic food web exhibits high concentrations of chlordecone, reflecting the
317 contamination of the baseline. This first way of contamination indicates a prevalence of a contamination “by
318 bath”, *i.e.* by contact between organisms (*via* teguments and gills) and contaminated surrounding waters. In
319 seagrass beds and coral reefs, food sources and primary consumers shared similar level of contamination that
320 were significantly different from those of the secondary consumers. In these two habitats, a phenomenon of
321 bioamplification, *i.e.* a contamination by trophic way, is highlighted. The introduction of carnivory in fish diet is
322 linked to a significant increase in the chlordecone concentrations of marine organisms, probably due to the
323 higher lipid content in the preys. In the Lesser Antilles, the degree of contamination of marine systems by
324 chlordecone was drastically lower than that measured in the James River estuary, due to the difference of
325 pollution discharge between the two sites. However, the majority of the studied species were impacted by
326 chlordecone and showed concentrations above the maximal residue limit (LMR) authorized by the French food
327 and safety authorities for the consumption and the commercialization of seafood products (LMR = 20 µg.kg⁻¹).

328 The variations among species from similar trophic category were sometimes very high, suggesting that other
329 processes could influence the level of contamination of marine fishes, such as their physiology, their size or their
330 movement among the different habitats. Indeed, Luellen *et al.* (2006) indicated that fish ecology could
331 considerably influence the level of contamination of marine fishes, especially for migratory species that spend a
332 period of the year in an estuarine environment.

333 To conclude, molecules of chlordecone can reach marine ecosystems with two principal processes. Firstly,
334 dissolved molecules of chlordecone can infiltrate ground waters during percolation process and join marine
335 environment with resurgences inside rivers or in the sea (Crabit *et al.* 2016). In the present study, this pollution
336 was highlighted by the concentrations of chlordecone measured in seawater samples. Secondly, due to its strong
337 affinity with soil particle (allophane or others terrestrial particles), molecules of chlordecone can reach runoff
338 waters, rivers and estuaries by leaching of the contaminated soil throughout the erosion process of the catchment
339 (Crabit *et al.* 2016). When chlordecone arrive in marine environment, it seems to stay sorbed on allophanes and
340 this fact could be similar with other organic compounds, as salinity generally does not affect desorption rates
341 (Bakir *et al.* 2014). Chlordecone, adsorbed or dissolved, is then integrated in SOM and zooplankton. These two
342 compartments are located at the base of trophic food-web and contribute to the contamination of the entire food
343 chain, *via* contact or trophic way.

344 In perspectives, several aspects of chlordecone transfer between terrestrial, rivers and marine ecosystems should
345 be developed in future studies. The low proportion of chlordecone desorbed from allophanes requires further
346 studies on the physical and chemical behavior of allophanes when arriving in marine environments. The
347 potential ways of chlordecone degradation in marine sediment could also be investigated in order to understand
348 the low concentrations of chlordecone found in marine sediment. Measurements of chlordecone metabolites in
349 the sediment could help with the understanding of a potential bacterial degradation in this compartment. Finally,
350 studies on the kinetic of contamination *versus* decontamination in marine fauna could be useful to explain the
351 high variations in the concentrations of chlordecone between species and individuals.

352

353 **Acknowledgements**

354 The authors express thanks to the institutions that gave financial supports to realize this study: Office de l'Eau of
355 Guadeloupe and Martinique, ONEMA (AFB), DEAL Guadeloupe and Martinique (for the program CLIPPER
356 and the study on the plankton contamination by chlordecone), Prefecture of Martinique (ChloHal program).

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