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# Journal Pre-proof

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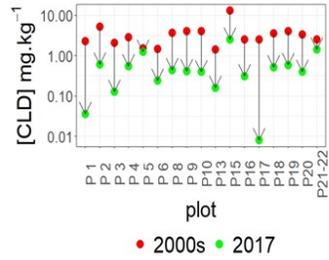
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1 **Author contributions**

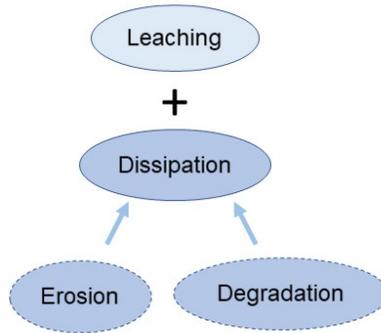
- 2 Comte: Conceptualization; Data curation; Formal analysis; Investigation; Methodology; Project  
3 administration; Resources; Software; Supervision; Validation; Visualization; Roles/Writing - original  
4 draft; Writing - review & editing.
- 5 Pradel: Conceptualization, Data curation, Investigation; Methodology; Writing - review & editing
- 6 Crabit: Data curation; Investigation; Methodology; Writing - review & editing
- 7 Mottes: Writing - review & editing
- 8 Pak: Writing - review & editing
- 9 Cattan: Conceptualization, Data curation, Formal analysis; Funding acquisition, Methodology,  
10 Software, Visualization, Roles/Writing - original draft, Writing - review & editing.

### 1. In situ observation

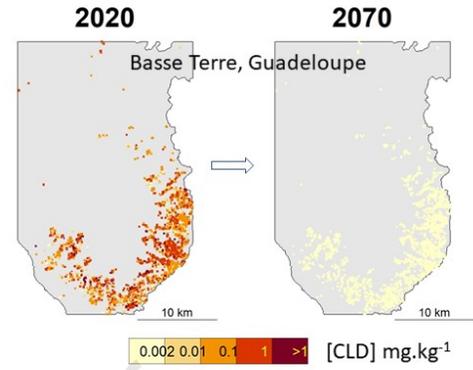
Decrease of soil chlordecone (CLD) concentration



### 2. Model Improvement



### 3. Updated pollution duration forecast



Journal Pre-proof

1 **Title: Long-term pollution by chlordecone of tropical volcanic soils in the French West Indies: new**  
2 **insights and improvement of previous predictions**

3 Irina Comte<sup>1,2</sup>, Alice Pradel<sup>3</sup>, Armand Crabit<sup>4</sup>, Charles Mottes<sup>2,5</sup>, Lai Ting Pak<sup>2,6</sup>, Philippe Cattan<sup>2,7</sup>

4

5 <sup>1</sup>Cirad, UPR GECCO, Capesterre-Belle-Eau, Guadeloupe F-97130, France

6 <sup>2</sup>GECCO, HortSys, TETIS, Univ Montpellier, CIRAD, Montpellier, France

7 <sup>3</sup>Univ Rennes, CNRS, Geosciences Rennes, UMR 6118, F-35000 Rennes, France

8 <sup>4</sup>Montpellier SupAgro, UMR G-EAU, University of Montpellier, INRAE, Avenue Jean-François Breton,  
9 34000 Montpellier, France

10 <sup>5</sup>Cirad, UPR HortSys, F-97455 Saint-Pierre, Réunion, France

11 <sup>6</sup>Cirad, UPR HortSys, Le Lamentin, Martinique F-97285, France

12 <sup>7</sup>Cirad, UMR TETIS, F-34398 Montpellier, France

13

14 Corresponding author: Philippe Cattan, [philippe.cattan@cirad.fr](mailto:philippe.cattan@cirad.fr), Dpt ES du CIRAD – UMR TETIS  
15 TA C-91 / MTD - Maison de la Télédétection - 500 rue Jean-François Breton - 34398 Montpellier  
16 Cedex 5 France

## 17 Abstract

18 Chlordecone (CLD), was widely applied in banana fields in the French West Indies from 1972 to 1993.  
19 The WISORCH model was constructed to assess soil contamination by CLD and estimated that it lasts  
20 from 100 to 600 years, depending on leaching intensity and assuming no degradation. However,  
21 recent studies demonstrated that CLD is degraded in the environment, hence questioning the  
22 reliability of previous estimations. This paper shows how to improve the model and provides insights  
23 into the long-term dissipation of CLD. In-situ observations were made in nearly 2,545 plots between  
24 2001 and 2020, and 17 plots were sampled at two dates. Results of soil analyses showed an  
25 unexpected 4-fold decrease in CLD concentrations in the soil, in contrast to simulations made using  
26 the first version of WISORCH at the time. Neither erosion, nor CLD leaching explained these  
27 discrepancies. In a top-down modeling approach, these new observations of CLD concentrations led  
28 us to implement a new dissipation process in the WISORCH model that corresponds to a DT50  
29 dissipation half-life of 5 years. The new version of the improved model allowed us to update the  
30 prediction of the persistence of soil pollution, with soil decontamination estimated for the 2070s.  
31 This development calls for re-evaluation of soil pollution status. Further validation of the new version  
32 of WISORCH is needed so it can contribute to crop management on contaminated soil.

33 Keywords: organochlorine, soil pollution, pollution decrease, modelling, Guadeloupe.

## 34 **1 INTRODUCTION**

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35 Organochlorine compounds have been used all over the world (Loganathan and Lam, 2012). Despite  
36 being banned or restricted, some molecules continue to contaminate soils, water bodies and food  
37 chains due to their persistence in the environment (Benoit et al., 2020; Falandysz et al., 2022; Jones  
38 and de Voogt, 1999; Nakata et al., 2002). This is the case in the French West Indies, where  
39 chlordecone (CLD, C<sub>10</sub>Cl<sub>10</sub>O; CAS N° 143-50-0; 491 g.mol<sup>-1</sup>), a toxic organochlorine insecticide was  
40 extensively applied in banana fields between 1972 and 1993 (Lesueur Jannoyer et al., 2016).  
41 Although it is known to be particularly stable, the long-term fate of these molecules remains poorly  
42 understood.

43 Modelling is needed to predict the duration of soil contamination by the pesticide at the scale of the  
44 area of application. While physically-based models can provide a detailed representation of the  
45 degradation and transfer of molecules and associated dynamics, the main difficulty with such models  
46 is their parameterization. In the case of large areas, the difficulty lies in obtaining sufficient  
47 information to include the spatial heterogeneity of the environments studied, along with problems of  
48 scale and of the representativeness of field measurements (Beven, 1989; Grayson et al., 1992).

49 In the context of the CLD pollution crisis in the French West Indies, Cabidoche et al. (Cabidoche et al.,  
50 2009) created the West-Indies-Soil-ORGanoCHlorine (WISORCH) model to forecast the persistence of  
51 contamination of soils. The main advantage of the model was its robustness (evaluated on a local  
52 scale) and the fact it only required a few parameters. This type of model was also called  
53 “parametrically parsimonious conceptual model” by [Littlewood 2003], and, according to Jakeman  
54 (Jakeman and Hornberger, 1993), can be defined as not needing more than six parameters. Based on  
55 the persistent character of the molecule, the originality of the model was that the dissipation of CLD  
56 in soils was mainly driven by leaching with no CLD degradation and with a minor contribution of  
57 erosion. Based on these assumptions, Cabidoche et al. (2009) predicted CLD contamination would  
58 last from 100 years to several centuries. However, these assumptions remained uncertain since the  
59 projections made by the model in 2009 could not be compared with in-situ measurements of the  
60 concentration of CLD in soils ([CLD]s) at different dates.

61 Recent studies indeed question these modeling results. Cattan et al. (Cattan et al., 2019) observed a  
62 reduction in the concentration of CLD in rivers over time, pointing to reduced presence of the  
63 molecule in the environment with medium-term persistence of pollution predicted rather than long-  
64 term. Cattan et al.'s observations are in agreement with those of Crabit et al. (Crabit et al., 2016),  
65 who estimated the persistence of pollution in the Pérou River that may not exceed around 50 years.

66 In addition, CLD degradation has been measured directly in laboratory studies (Fernández-Bayo et al.,  
67 2013; Lomheim et al., 2020; Mouvet et al., 2017), or through the detection of CLD metabolites in the  
68 environment (Chevallier et al., 2019; Devault et al., 2016). However, none of these studies quantified  
69 CLD degradation or estimated its dissipation rate based on repeated in-situ sampling over time.

70 Assessing the decrease of  $[CLD]_s$  is crucial since currently, public authorities think in terms of  
71 pollution lasting decades or even centuries, eliminating any possibility of cultivation in contaminated  
72 areas. Today, 10 years after Cabidoche et al.'s (2009) prediction, we have sufficient hindsight to  
73 assess the accuracy and the robustness of the model, which may be being used elsewhere. The first  
74 version of the WISORCH model thus needed to be improved. To this end, we used a top-down  
75 modeling approach that consisted in identifying the most appropriate model structure through a  
76 process of hypothesis testing (Littlewood et al., 2003; Sivapalan et al., 2003; Young, 2006). According  
77 to Sivapalan (Sivapalan et al., 2003), interpreting the observed data, or pattern in the data, makes it  
78 possible to build appropriate causal models and then to test these model predictions against the data  
79 in an iterative manner.

80 The objective of this study was to improve CLD modeling at a large scale. Starting from data on  $[CLD]_s$ ,  
81 available from 2001 to 2020, we tested other processes that could be involved in changes in  $[CLD]_s$   
82 and thus improve the structure of the model of Cabidoche et al. (2009). We first proceeded with a  
83 statistical analysis of the  $[CLD]_s$  database that contains all CLD soil analyses performed since 2001 in  
84 southern Basse Terre, Guadeloupe, as part of the monitoring program set up by public authorities.  
85 Second, we characterized in-situ changes in  $[CLD]_s$  between two periods using field measurements. A  
86 new model, WISORCH\_V1, was then derived from the comparison between field and simulated  
87 values. A final simulation, based on updated assumptions, was performed in the thousands of plots  
88 in the CLD database to provide updated estimates of the persistence of soil pollution by CLD. This  
89 work combines research and public monitoring in the medium-term and should help improve the  
90 management of the risk of CLD pollution.

## 91 **2 MATERIAL AND METHODS**

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### 92 **2.1 STUDY SITE**

93 The study was conducted in Guadeloupe in the southern part of the volcanic island of Basse Terre.  
94 The study area is characterized by a marked rainfall gradient with altitude ranging from 2,400  
95 mm.yr<sup>-1</sup> downstream to 7,000 mm.yr<sup>-1</sup> upstream (Morell and Jérémie, 1994). The main soil types are  
96 andosols, nitisols and ferralsols (Levillain et al., 2012). All three soils have a high infiltration capacity

97 (saturated hydraulic conductivity > 60 mm.h<sup>-1</sup>) and andosols also have high carbon content (Levillain  
98 et al., 2012). In our study, we considered plots at two scales:

- 99 1. Basse Terre island is the historical banana cultivation area where CLD was used in the past.  
100 Based on this land use, risk areas were mapped (DAAF, 2018) in three classes (low, medium  
101 and high risk) (Rochette et al., 2017). These areas were subject to extensive soil testing for  
102 CLD (Supplementary material - SM Figure 1).
- 103 2. The Pérou hydrosystem (SM Figure 2) in Basse Terre, which is a typical basin where banana  
104 has been and is still intensively cropped. All three main soil types (andosols, nitisols and  
105 ferralsols) are present in the basin. The experimental plots were distributed over the entire  
106 cultivated area, mainly on banana fields (12 fields, two on sugarcane, three on fallow) and  
107 andosol (12 fields, five on nitisols), between the isohyets 2,500 mm and 4,500 mm.

## 108 2.2 RAINFALL DATA

109 At the scale of Basse Terre island, rainfall was assessed from a vector layer of the isohyet of mean  
110 annual rainfall between 1929 and 1978 (L'Hôte, 1981).

111 At the scale of the Pérou hydrosystem, the annual rainfall for each experimental plot from the  
112 beginning of the chlordecone use in 1972 to the present was derived from data recorded by the  
113 reference station of Neufchâteau, by applying a correction depending on the altitude of the plot.  
114 Neufchâteau station (160403800 N, 613600400 W, 250 m amsl) is located 2.6 km from the Pérou  
115 river (SM\_Figure2), with an acquisition rate of 97.67%. Missing data were estimated using a rainfall  
116 typology (SM Appendix A).

117 To correct for altitude, we used the empirical equation based on rainfall series from 1929 to 1978  
118 and related rainfall to altitude on the eastern windward coast of the island of Guadeloupe with a  
119 slope of 5.3684 (Chaperon et al., 1983). The annual rainfall of each plot in each year was calculated  
120 according to Eq. 1.

121 Eq. 1: 
$$R_{i,j} = 5.3684 * (Z_i - Z_{NCT}) + R_{NCT,j}$$

122 where  $R_{i,j}$  is annual rainfall at site  $i$  in year  $j$ ;  $Z$  is the elevation (m) of site  $i$ ;  $NCT$  is the site of Neufchâteau  
123 weather station

## 124 2.3 COLLECTION OF DATA ON CLD CONCENTRATIONS IN THE SOIL

### 125 2.3.1 Soil CLD database in southern Basse Terre

126 We used data from the soil sampling campaign performed by various agencies using the same  
127 sampling protocol (notably a sampling depth of 0-30 cm; samples were sent to the *Laboratoire*

128 *Départementale d'Analyses de la Drôme* (LDA26, Valence, France) in charge of CLD soil analysis), as  
129 part of environmental or agricultural monitoring networks.

130 This soil CLD database is hosted at the *Direction de l'Alimentation, de l'Agriculture et de la Forêt*  
131 (DAAF). It contains 6,497 georeferenced entries made from 2001 to 2020, with an average analysis  
132 frequency of 401 analyses per year since 2004; from 2001 to 2003, the number of analyses was less  
133 than 10. Soil analyses were carried out throughout Guadeloupe, but most in the high-risk area in  
134 southern Basse Terre (SM Figure 1).

135 To assess the changes in [CLD]<sub>s</sub> over time, we focused on the plots located in the high-risk area (SM  
136 Figure 1), with [CLD]<sub>s</sub> higher than the detection threshold of 0.002mg.kg<sup>-1</sup>. This included 1,038 plots  
137 on andosol, 380 plots on ferralsol, and 1,127 plots on nitisol. The concentrations of [CLD]<sub>s</sub> in plots  
138 located outside the high-risk area were too low or below the detection threshold, thereby preventing  
139 us from correctly assessing variations in [CLD]<sub>s</sub> over time.

### 140 **2.3.2 Soil sampling campaign in the Pérou hydrosystem**

141 The study of in situ variation of [CLD]<sub>s</sub> between two sampling dates was performed on 17 plots  
142 located in the Pérou hydrosystem . The first measurements of [CLD]<sub>s</sub> on these plots were carried out  
143 from 2005 to 2012. This period is called the first soil sampling campaign. These same plots were  
144 sampled a second time using the same sampling protocol between July and August 2017, called the  
145 second soil sampling campaign. The duration between the two sampling dates is therefore different  
146 for each plot and varies from 5 to 12 years.

147 A uniform and regular sampling grid was designed for each plot with three transects 10-30 m apart  
148 and at least 7 m from the edge of the field. A minimum of 15 points per hectare were sampled in the  
149 0-30 cm soil layer and a minimum of eight points in smaller plots. Subsequently, all the soil sub-  
150 samples of a given plot were pooled and manually homogenized to make a single composite soil  
151 sample from which an aliquot was extracted. Leaves, roots, and pebbles were removed before the  
152 aliquot was extracted. After each plot was sampled, all the sampling equipment was thoroughly  
153 washed with distilled water to avoid contamination between plots.

154 All the composite soil aliquots were cooled to 4 °C prior to being sent to laboratories within one  
155 month maximum. Aliquots for CLD analysis were sent to LDA26. Aliquots for physico-chemical  
156 parameters were sent to the Service Unit (US49) of Cirad, Montpellier, France.

157 In the 2017 campaign, at each sub-sampling point, bulk density (BD) was measured at the soil surface  
158 after removing the first centimeters of dead plant cover using a 100 cm<sup>3</sup> stainless-steel cylinder. The

159 average value of bulk density was then calculated at the plot scale. Soil organic carbon (SOC) was  
160 measured using the dry combustion (Dumas) method.

### 161 **2.3.3 Soil profile campaign in 2010: CLD distribution with depth**

162 In 2010, four plots were selected in the Pérou hydrosystem to study [CLD]<sub>s</sub> along soil profiles in  
163 different types of soil and under different soil tillage: one andosol at an elevation of 347 meters with  
164 no tillage; one andosol at an elevation of 128 meters with soil tillage; two nitisols with soil tillage at  
165 elevations of 60 and 76 meters respectively, the first with high CLD contamination, the second with  
166 low CLD contamination (Cattan, 2013)

167 On each plot, a soil profile was excavated and samples were taken at 5 depths: 0-10 cm; 10-30 cm;  
168 30-50 cm; 50-70 cm; 70-90 cm. The sampling equipment was carefully cleaned with distilled water  
169 before sampling at each depth to avoid contamination between soil horizons. Like in the soil  
170 sampling campaign in 2017, all the samples of soil from the four profiles were cooled prior to being  
171 sent to the LDA26 laboratory.

## 172 **2.4 PESTICIDE ANALYSIS**

173 The selected LDA26 is accredited by Cofrac, the French Accreditation Committee for pesticide  
174 analyses thereby guaranteeing their technical skills and reliability as well as good management  
175 practices. LDA26 complies with ISO 17025 standards for testing and calibration. The concentration of  
176 CLD, CLD-5b-hydro and chlordecol were determined according to the French standard AFSSA/  
177 LERQAP/TOP POP04 based on Blanke et al. (Blanke et al., 1977), the routine procedure (internal  
178 procedure CMO\_MT06: quantification of organochlorine and organophosphorous pesticides based  
179 on the norm XP X 33-012)(IPCS, 1990).

180 The soil samples were homogenized, lyophilized, and screened (2 mm) before extraction. Next, 10 g  
181 of the lyophilized soil sample was mixed with Fontainebleau sand (purified at 400 °C). The extraction  
182 was carried out using dionex ASE 200 (Accelerated Solvent Extractor) under heavy pressure (200  
183 bars) and high temperature (100 °C) in three 5-min cycles. This step was performed once at neutral  
184 pH and once at acidic pH. The extraction solvent was a mix of acetone/dichloromethane (50/50). A  
185 solution containing known concentrations of tracer HBB-TPP (Hexabromobenzene and  
186 Triphenylphosphate) was added to guarantee proper extraction: the recovery rates of HBB-TPP had  
187 to be between 80 and 120% ; If not, a second extraction was performed to check the correctness of  
188 the process. The extract residue was transferred into a Turbovap tube with 1 mL of isooctane and  
189 concentrated to 1 mL before addition of ethyl acetate to obtain a final volume of 10 mL. Then 1 mL of  
190 this volume was analyzed by HPLC MS. The coupling of HPLC with two-dimensional mass

191 spectrometry ensured that analytes did not interfere in the analysis of chlordecone. Internal injection  
 192 standards were Chlordecone C13 and 2.4D d3. The limit of quantification was 0.01mg.kg<sup>-1</sup>.

## 193 2.5 CALCULATION.

194 **Trend in [CLD]<sub>s</sub> on the DAAF database:** for each soil type, log regression analysis (equation of the  
 195 form  $\text{Log}([CLD]_s) = a * \text{Time} + b$ ) was performed using R freeware (R Core Team., 2020). We retained  
 196 plots whose [CLD]<sub>s</sub> was above the detection threshold of 0.002mg.kg<sup>-1</sup>; [CLD]<sub>s</sub> ranging between the  
 197 detection and the quantification threshold of 0.01 mg.kg<sup>-1</sup> were set to 0.005 mg.kg<sup>-1</sup>.

198 The relative decrease in [CLD]<sub>s</sub> between two dates was computed according to Eq. 2:

$$199 \text{ Eq. 2: } \text{relative decrease} = ([CLD]_{s,i} - [CLD]_{s,j}) / [CLD]_{s,i} = 1 - 1 / \exp(a * (T_i - T_j))$$

200 where i is the sample during the first campaign and date j for the sample during the second campaign (i.e.  
 201 2017); a is the slope computed by the log regression analysis. Difference between dates is in days.

202 **Annual rate of [CLD]<sub>s</sub> decrease in the 17 plots** was calculated from [CLD]<sub>s</sub> measured at two dates  
 203 according to the following sequence:

- 204 - Calculation of x, the annual rate of [CLD]<sub>s</sub> decrease from Eq. 3;
- 205 - then, knowing x, calculation of the relative decrease at the desired date tx with Eq. 4

$$206 \text{ Eq. 3: } \text{for one unit of time, we have } [CLD]_{s,t1+1} = [CLD]_{s,t1} (1-x)^{(t1+1-t1)} = [CLD]_{s,t1} (1-x)$$

$$207 \text{ then for dates } t2 \text{ and } t1 \quad [CLD]_{s,t2} = [CLD]_{s,t1} (1-x)^{(t2-t1)} \Leftrightarrow$$

$$208 \quad x = 1 - ([CLD]_{s,t2} / [CLD]_{s,t1})^{1/(t2-t1)}$$

$$209 \text{ Eq. 4: } ([CLD]_{s,tx} - [CLD]_{s,t1}) / [CLD]_{s,t1} = 1 - (1-x)^{(tx-t1)}$$

210 where t1 and t2 are the measurement dates in the first and second campaign, respectively

## 211 2.6 WISORCH MODEL

212 In the initial version of the WISORCH model, called WISORCH\_V0, and described by Cabidoche et al.  
 213 (2009), variations in the concentrations of the pesticide in the soil dS (kg.ha<sup>-1</sup>) were a function of the  
 214 remaining stock of that pesticide in the soil, S (kg.ha<sup>-1</sup>), and of the amount of percolating water dD  
 215 (mm), as described by Eq. 5:

$$216 \text{ Eq. 5: } -dS = C1 \times S \times dD$$

217 where C1 is the percolation coefficient and equals  $10 / (Koc \times SSOC)$ ; Koc (mL.g<sup>-1</sup>) is the partitioning  
 218 coefficient between the sorbed part of the pesticide on soil organic carbon and the dissolved part in water;  
 219 SSOC (t.ha<sup>-1</sup>) is the stock of organic carbon calculated with Eq. 6.

$$220 \text{ Eq. 6: } SSOC = (10^{-2} \times SOC) \times BD \times Vol_s$$

221 where SOC (g/100g) is the soil organic carbon, BD is bulk density in T.m<sup>-3</sup>, and Vol<sub>s</sub> is the soil volume (m<sup>3</sup>) of 1  
 222 ha = 0.3 m x 10 000 m<sup>2</sup> (0.3 m being the sampling depth).

223 The stemflow was considered in the previous version of the model because of the need to consider  
 224 the redistribution of rainfall through the plant canopy and preferential fluxes along the banana stem  
 225 that could have enhanced CLD leaching during the three to five years after CLD application. In this  
 226 paper, the stemflow function was removed since there were no further applications of CLD.

227 First, we ran the WISORCH\_V0 model on each plot in our dataset using the initial parameter values  
 228 provided by Cabidoche et al. (2009) or in-situ measurements (Table 1). The initial CLD stock  $S_0$  was  
 229 calculated with Eq. 7 using the [CLD]<sub>s</sub> analyzed during the first campaign: In this case,  $S_0$  is  
 230 equivalent to the initial, unique, and uniform CLD application at the sampling date.

231 
$$\text{Eq. 7: } S_0 = [\text{CLD}]_s \times \text{BD} \times \text{Vol}_s \times 10^{-3}$$

232 with  $S_0$  in  $\text{kg.ha}^{-1}$ ,  $[\text{CLD}]_s$  in  $\text{mg.kg}^{-1}$

233

234 *Table 1: parameter values in simulations*

Parameters	Initial values	Calibration
Koc nitisol ( $\text{L.kg}^{-1}$ )	2 000	2 000; 3 000
Koc andosol ( $\text{L.kg}^{-1}$ )	17 500	2 000 to 20,000, step of 3,000
Bulk density	In-situ measurement	Sim0 + (0, 10, 15, 25, 50) %
SOC ( $\text{g}/100 \text{ g}$ )	In-situ measurement	Sim0 x (0, 90, 75, 50) %
Runoff coefficient (%)	0.06	0.03; 0.06
Rainfall (mm)	Calculation (see section 2.2)	Sim0 + (0, 25, 50) %

235 SOC: soil organic carbon; Koc, the partitioning coefficient between the sorbed part of the pesticide on soil  
 236 organic carbon

237

238 Second, a calibration was performed with the range of parameter values described in Table 1.

239 Simulations were run with Koc values ranging from 2,000 to 20,000 with lower Koc for nitisols than  
 240 for andosols (ATSDR, 1995; Baran and Arnaud, 2013; Bonvallot and Dor, 2004; INERIS, 2021). The  
 241 runoff coefficient was that reported in the Cabidoche paper (Cabidoche et al., 2009). Other  
 242 parameter values were attributed to favor a reduction in the concentration of CLD in the soil, under  
 243 the hypothesis of systematic bias in field measurements. Thus, simulations were performed with:  
 244 increasing values of BD ([CLD]<sub>s</sub> inversely proportional to BD - Eq. 7); decreasing values of SOC (the  
 245 lower SOC, the lower soil retention of CLD); decreasing values of RC (the lower RC, the higher the  
 246 leaching of CLD); increasing values of Rainfall coefficient (the higher the rainfall, the higher the  
 247 leaching of CLD).

248 All combinations of parameter values were simulated (n=4200) using R freeware (R Core Team.,  
 249 2020). The  $r^2$  and the slope (intercept = 0) of the regression between the simulated and observed  
 250 value on the 17 plots were used to analyze the effect of parameter variations on the quality of the

251 relation between observed and simulated [CLD] values. We considered a good result to be when  
 252 both  $r^2$  and the slope were close to 1.

## 253 **3 RESULTS AND DISCUSSION**

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### 254 **3.1 UNEXPECTED DECREASE IN THE OBSERVED CONCENTRATIONS OF CLD IN THE** 255 **SOIL OVER TIME**

#### 256 **3.1.1 Overall decrease in the CLD concentrations in the soil between 2001 and 2020 at a** 257 **large scale.**

258 Although the data show marked dispersion, the regressions of the log of [CLD]<sub>s</sub> according to time  
 259 from samples collected from 2001 to 2020 in southern Basse Terre (see section 2.3.1) revealed a  
 260 significant decrease in [CLD]<sub>s</sub> (SM Figure 3). Slopes and their confidence interval were  $-4.2 \cdot 10^{-4}$  [ $-$   
 261  $4.2 \cdot 10^{-4}$ ,  $-3.5 \cdot 10^{-4}$ ],  $-2.8 \cdot 10^{-4}$  [ $-3.6 \cdot 10^{-4}$ ,  $-2.0 \cdot 10^{-4}$ ],  $-3.7 \cdot 10^{-4}$  [ $-4.3 \cdot 10^{-4}$ ,  $-3.1 \cdot 10^{-4}$ ] for andosol, ferralsol  
 262 and nitisol, respectively. However, these slopes did not significantly differ from each other and no  
 263 effect of soil was evidenced from these data.

264 Based on these regressions, the relative decreases in the non-log CLD concentrations were computed  
 265 according to Eq. 2 (see section 2.5) over the periods 2005-2017 and 2012-2017, i.e. the periods that  
 266 overlap the sampling periods of the 17 plots in the Pérou watershed. This yielded decreases of up to  
 267 78% between 2005 and 2017, and of up to 47% between 2012 and 2017.

#### 268 **3.1.2 Substantial decrease in soil CLD concentrations between the two soil campaigns at** 269 **the local scale**

270 A substantial decrease in [CLD]<sub>s</sub>, ranging from 15.2% to 99.7% (average 82%), was observed between  
 271 the first sampling period (2005-2012) and the second sampling period in 2017 in the same plots  
 272 (Table 2). This decrease was slightly higher than the tendency observed in the DAAF database  
 273 mentioned above (47-78% decrease). In this case, there were no significant effect of the length of the  
 274 period between the first and second campaign on the decrease.

275 The [CLD]<sub>s</sub> was higher in andosols than in nitisols (respective median of 3.45 and 2.3 mg.kg<sup>-1</sup>, p value  
 276 of 0.11 in Campaign 1; median of 0.532 and 0.24 mg.kg<sup>-1</sup>, p value of 0.048 in Campaign 2 according to  
 277 a Wilcoxon test). In agreement with Cabidoche et al. (2009), this result was expected and may be  
 278 partially linked to the high soil carbon content of andosols compare to nitisols (mean SOC of 4.28%  
 279 for andosol vs 2.2% for nitisol, see Table 2). However, there was no significant effect of the type of  
 280 soil on the decrease in [CLD]<sub>s</sub> although according to Cabidoche et al. (2009), a bigger decrease was

281 expected in nitisol. The effect of soil was not significant for CLD-5b-hydro or for Chlordecol (p value >  
282 0.2).

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283 *Table 2: Characteristics of the sampled plots and concentration of CLD, CLD-5b-hydri and Chlordecol in the two soils campaigns*

Plot ID	Soil type	Land use in 2017	Altitude (m)	Bulk density (g.cm <sup>-3</sup> )	SOC 2017 (%)	1st Campaign (2005-2012)			2 <sup>nd</sup> Campaign (2017)				
						Sampling year	CLD (µg.kg <sup>-1</sup> )	5b hydrochlordecone (µg.kg <sup>-1</sup> )	Sampling date	CLD (µg.kg <sup>-1</sup> )	5b hydrochlordecone (µg.kg <sup>-1</sup> )	chlordecol (µg.kg <sup>-1</sup> )	CLD decrease (%)
1	nitisol	Fallow	166	0,80	2,29	2010	2,300	50	2017-07-17	36	<LoD	28	-98%
2	andosol	Banana	179	0,79	2,79	2010	5,276	181	2017-07-31	606	<LoD	186	-89%
3	nitisol	Banana	156	0,85	2,28	2010	2,100	26	2017-08-01	127	<LoD	84	-94%
4	andosol	Sugarcane	196	0,66	4,41	2012	2,862	50	2017-08-03	546	<LoD	252	-81%
5	andosol P.	Banana	322	0,67	4,91	2006	1,500	NA	2017-04-08	1,272	16.7	784	-15%
6	nitisol	Fallow	52	1,01	2,00	2010	1,445	42	2017-08-04	240	<LoD	80	-83%
8	nitisol	Sugarcane	48	1,01	2,21	2007	3,700	NA	2017-08-08	446	7.7	403	-88%
9	andosol P.	Banana	248	0,74	5,59	2010	4,100	NA	2017-08-08	409	<LoD	166	-90%
10	andosol	Banana	208	0,86	4,23	2012	4,010	40	2017-08-09	393	<LoD	192	-90%
13	andosol	Banana	113	0,72	3,55	2005	1,400	NA	2017-08-11	161	<LoD	95	-89%
15	andosol	Banana	120	0,67	4,64	2010	12,952	NA	2017-08-14	2,518	18.5	436	-81%
16	nitisol	Banana	70	1,04	2,23	2009	2,500	NA	2017-08-16	314	<LoD	239	-87%
17	andosol	Banana	135	0,70	4,13	2009	2,500	NA	2017-08-17	8	<LoD	<LoD	-100%
18	andosol P.	Banana	336	0,68	3,78	2009	5,500	NA	2017-08-18	517	9.8	230	-91%
19	andosol P.	Banana	425	0,60	5,94	2009	4,740	10	2017-08-21	593	5.9	215	-87%
20	andosol P.	Banana	271	0,71	4,42	2006	3,450	NA	2017-08-21	400	7.5	150	-88%
21	andosol	Fallow/ Sugarcane	171	0,69	4,22	2012	2,507	50	2017-08-22	1431	15.2	390	-43%

284

285 cc: concentration; andosol P.: Perhydrated andosol; SOC: soil organic carbon; LoD: Limit of Detection

## 286 **3.2 INCONSISTENCY BETWEEN OBSERVED AND VALUES PREDICTED BY**

### 287 **WISORCH\_V0**

288 First, the observed decrease in  $[CLD]_s$ , was much bigger than expected according to the simulations  
289 performed with the first version of WISORCH model (WISORCH\_V0). Indeed, for the 17 plots,  
290 simulated values of  $[CLD]_s$  for 2017 were, on average, 4-fold higher than the observed values (slope  
291 of 4.21,  $r^2$  of 0.75, p value less than 0.001).

292 Secondly, the results of the WISORCH\_V0 calibration (4200 simulations; SM\_Figure 4), show that the  
293 leaching process alone cannot lead to decreases as large as those observed in 2017. In fact, the case  
294 that maximized  $r^2$  with a slope of 1 was obtained for a rainfall coefficient of 1.2 and a SOC equal to  
295 half the observed value. These results suggest that, in order to obtain good results, the parameters  
296 must be distorted in such a way that they no longer correspond to what is observed in the field in  
297 terms of climate conditions and field properties: systematic overestimation of SOC by a factor 2  
298 (measured SOC / SOC simulation = 2), and systematic underestimation of the annual rainfall by 17%  
299 (measured rainfall / rainfall simulation = 83%). Additionally, the Koc for andosol has to be set to  
300 2,000, i.e., the minimum value for andosols and the same value as for nitisols.

301 Few references exist concerning the mobility of CLD in the soil profile. Ollivier et al. (Ollivier et al.,  
302 2020) found that CLD percolation ranged from 13.9% to 15.1% of the stock in experimental  
303 conditions equivalent to 3.2 years of percolation and 5,340 mm of rainfall on a nitisol. With the same  
304 percolation flux, WISORCH\_V0 yielded higher losses (37%) with the initial parameters presented in  
305 section 2.6 for a nitisol. On the other hand, the losses calculated over the same period from data  
306 measured in the 17 plots (see Eq. 3 and Eq. 4 in section 2.5) yield a mean decrease of 60% on nitisol  
307 and 50% on andosol. The gap between these and previous decreases suggests other processes than  
308 leaching are involved.

309 These results thus question the processes involved in the observed decreasing tendency in  $[CLD]_s$ ,  
310 and challenge the assumptions used in the first version of WISORCH model: negligible CLD  
311 degradation and erosion, and preponderant effect of leaching.

## 312 **3.3 NEW DISSIPATION FUNCTION AND CLD HALF-LIFE VALUE OF 5 YEARS**

313 A new version WISORCH\_V1 was then derived from WISORCH\_V0. Learning from data and model tests,  
314 we added a new process of CLD dissipation in WISORCH\_V1. Here, we define dissipation as the overall  
315 process leading to the eventual disappearance of substances from the site at which it was applied or  
316 an environmental compartment. It includes transport and transformation processes (OECD, 2016). We

317 then added a dissipation function depending on time  $T$  (in days) and following a first order kinetics (Eq.  
 318 8), i.e. the form  $-dA/dT=kA$  describing the disappearance of a product  $A$  according to  $T$ . As a result of  
 319 this added process, a new parameter was introduced in the model:  $DT50$  is the 50% dissipation time,  
 320 i.e., the amount of time required for 50% of the initial pesticide concentration to dissipate.

321 Eq. 8:  $-dS = C1 \times S \times dD + C2 \times S \times dT$

322 where  $C2$  is the dissipation coefficient and equals  $\text{Log}(2) / DT50$  (days). Note that in our case,  $C2$  excludes  
 323 leaching which is already accounted in the first term of Eq. 8 ( $C1 \times S \times dD$ )

324 After integration this gives:

325 Eq. 9:  $S(D_a, T-T_a) = S(0, T_a) \times \exp(-C1 \times D_a) \times \exp(-C2 \times (T-T_a))$

326 where  $S(0, T_a)$  is the quantity of pesticide applied at application  $a$ .  $T-T_a$  is the time elapsed (in days) between  
 327 the date  $T_a$  of application  $a$  to the valuation date  $T$  of the remaining stock in the soil.  $D_a$  (mm) is the amount  
 328 of percolating water from  $T_a$  to  $T$  (Eq. 10).

329 Eq. 10:  $D = \text{Rainfall} - \text{PET} - \text{Runoff}$

330 with  $\text{Runoff} = RC \times \text{Rainfall}$  (mm),  $RC$  being the Runoff coefficient (%);  $PET$  the potential evapotranspiration  
 331 (mm).

332 Then for  $na$  applications during the period of time  $T-T_a$ , the remaining stock in soil is assessed by (Eq.  
 333 11)

334 Eq. 11:  $S = \sum_{a=1}^{na} S(0, T_a) \times \exp(-C1 \times D_a) \times \exp(-C2 \times (T-T_a))$

335 WISORCH\_V1 was calibrated with the parameter values described in section 2.6 to which we added  
 336  $DT50$  values ranging from 2 to 50, according to the literature (ATSDR, 1995; National Center for  
 337 Biotechnology Information, 2021). By running the resulting 33,600 simulations (SM Figure5), we  
 338 identified the 738 best simulations characterized by:  $0.8 < \text{slope} < 1.2$  and  $r^2 > 0.975 \times \max(r^2)$ . In this  
 339 group, 99% of simulations present a  $DT50$  of 5 years (SM\_Table1). Additionally,  $Koc$  value for andosol  
 340 is less than  $17,000 \text{ mL.g}^{-1}$  with a maximum frequency for the value of 5,000 (54%). The distribution of  
 341 the other parameters does not show such heterogeneity in distribution frequency, which means that  
 342 they are less sensitive and therefore have less impact on the quality of the simulation.

### 343 **3.4 ASSESSMENT OF TWO MAIN PROCESSES INVOLVED IN CLD DISSIPATION**

344 Basically, the decrease in  $[CLD]_s$  can be ascribed either to the mobility of the compound moving from  
 345 the contaminated plots and/or in deeper horizons, or to its degradation over time. Different  
 346 dissipation pathways may be involved (volatilization, sorption, leaching, degradation, plant uptake,  
 347 etc.) (OECD, 2016). We discuss the two main processes, erosion and degradation (notably,  $CLD$  is a  
 348 non-systemic and non-volatile insecticide).

### 349 3.4.1 Erosion

350 Soils in the French West Indies (andosols, nitisols, ferralsols) present a highly stable micro-aggregated  
351 structure due to the nature of their minerals (iron oxyde, allophans). This may limit the erosion of  
352 fine particles (Cabidoche et al., 2009). A study on nitisol in Martinique reported low erosion rates in  
353 banana fields ( $0.4$  to  $0.6 \text{ t}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ ) and in sugar cane fields ( $< 0.2 \text{ t}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ ), but higher erosion rates  
354 in mechanized pineapple fields ( $5$  to  $29 \text{ t}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ ) and bare soils ( $85$  to  $165 \text{ t}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ ) (Khamsouk,  
355 2001). Saffache (Saffache, 2000) extrapolated sediment losses to equal  $39.6 \text{ t}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  on farmed soils  
356 in an experiment using a mini rain simulator on  $1\text{m}^2$  fields. Rad et al. (Rad et al., 2006) proposed an  
357 approximate mechanical weathering rate of  $8\text{--}40 \text{ t}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  for Guadeloupe based on a geochemical  
358 study of dissolved and solid matter in rivers.

359 Based on these data, we speculated that long-term erosion (i.e. 15 years, consistent with our data) of  
360 soil from banana fields could range from  $157$  to  $690 \text{ t}\cdot\text{ha}^{-1}$ . This value is based on the assumption of  
361 three cycles of banana production with an erosion rate of  $0.5$  to  $40 \text{ t}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$  alternating with two  
362 periods of bare soil (9 months each) with an erosion rate of  $100 \text{ t}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ . Assuming a BD of  $1 \text{ g}\cdot\text{cm}^{-3}$ ,  
363 this represents an eroded soil layer ranging from  $1.6$  to  $6.9 \text{ cm}$  over the 15-year period.

364 The soil profiles carried out in 2010 in the Pérou watershed showed that  $[\text{CLD}]_s$  decreased rapidly  
365 with depth (SM\_Figure6). Although the mobilization of CLD in deeper horizons may be slightly  
366 increased because of tillage, most CLD remained in the topsoil. To evaluate the effect of different soil  
367 stripping intensities due to erosion on the CLD concentration measured, using these profiles, we  
368 calculated the concentration of CLD in a 30-cm soil column taken from different depths compared to  
369 the current measurement in the 0-30 cm soil layer. According to Figure 1, it would require a soil  
370 stripping of at least 20 cm to reduce the  $[\text{CLD}]_s$  by 80% (i.e. the average decrease observed in the 17  
371 plots) in the case of a no-till andosol, and of 50 cm or more for other soils. Retaining the above values  
372 of 2-7 cm for soil stripping, the  $[\text{CLD}]_s$  in the top 30-cm soil layer would still be about 88% of the  
373 initial concentration prior to stripping in a no-till andosol and more than 95% in other soils. Thus, the  
374 surface erosion process alone is unlikely to be responsible for the decrease in  $[\text{CLD}]_s$  in the 17  
375 experimental plots.

### 376 3.4.2 Degradation

377 Pesticide analysis (Table 2) revealed the presence of CLD metabolites CLD-5b-hydro and chlordecol,  
378 in sampled plots. Their presence attests to in-situ degradation. Notably, the mean CLD-5b-hydro/CLD  
379 ratios in fields where CLD-5b-hydro was detected was 0.018 (minimum 0.0025) during the first  
380 campaign and 0.013 (minimum 0.0073) in the second campaign, i.e. far higher than the mean of  
381 0.00077 reported by Devault et al., (Devault et al., 2016) and suggesting CLD-5b-hydro was produced

382 in the soil after application of CLD. However, in all plots where both data were available, CLD-5b-  
383 hydro decreased between campaign 1 and 2 (Table 2). Due to the fate of metabolites in the  
384 environment, their concentration in the soil cannot accurately account for the extent of the CLD  
385 degradation.

386 These results are consistent with those of recent studies providing evidence that the CLD  
387 degradation process is likely underway in the French West Indies: the existence of chlordecone  
388 degrading organisms in a tropical andosol (Fernández-Bayo et al., 2013); identification of CLD  
389 metabolites in soils and sediment in Martinique, account for the capacity of CLD to degrade in these  
390 tropical soils (Chevallier et al., 2019). CLD degradation is proven but its contribution to global CLD  
391 dissipation remains unknown.

### 392 **3.5 PREDICTION OF THE DURATION OF POLLUTION IN SOUTHERN BASSE-TERRE,** 393 **GADELOUPE**

394 The same dataset as that used in section 3.1.1 was used to predict [CLD]<sub>s</sub> at different dates using  
395 WISORCH\_V1. For each plot, the dataset was completed with mean annual rainfall by spatial  
396 intersection of the plot layer with the isohyet layer of the mean annual rainfall between 1929 and  
397 1978 (L'Hôte, 1981). This mean annual rainfall value was used for all the simulated years. The values  
398 of the soil parameters SOC and BD were not available in the DAAF dataset, and were consequently  
399 set according to Levillain et al. (2012): 2% SOC for nitisols, 3% for ferralsols and 7% for andosols; BD  
400 of 1.10 T.m<sup>-3</sup> for nitisols and ferralsols and 0.55 T.m<sup>-3</sup> for andosols. Koc values were set according to  
401 our own simulation results: 2,000 L.kg<sup>-1</sup> for nitisols and ferralsols, and 5,000 L.kg<sup>-1</sup> for andosols. The  
402 runoff coefficient was set at 0.06 (Cabidoche et al., 2009) and the DT50 at 5 years.

403 Figure 2 shows that [CLD]<sub>s</sub> in plots located on nitisols and ferralsols would drop below the detection  
404 limit of 0.002 mg.kg<sup>-1</sup> by the 2050s, and in plots located on andosols, by the 2070s. The crops that  
405 can be grown could be assessed referring to threshold defined by Jannoyer ((Jannoyer-Lesueur,  
406 2018), i.e. 1 and 0.1 mg.kg<sup>-1</sup> the thresholds below which it is possible to grow plants with  
407 intermediate or high sensitivity to CLD, respectively. According to these thresholds, from 2050 on,  
408 there would be no restriction on cultivating plants on all soils, and from 2030 on, on nitisol ([CLD]<sub>s</sub>  
409 below 0.01 mg.kg<sup>-1</sup>).

410 Consistent results were obtained with the regression analysis described in section 3.1.1 with the  
411 same dataset. The [CLD] would drop below the detection limit of 0.002 mg.kg<sup>-1</sup> by the 2050s in all  
412 soils (2048 [2026-2071] in andosols, 2059 [2030-2097] in ferralsols, 2047 [2025-2072] in nitisols – the

413 confidence intervals of the dates were calculated according to the confidence intervals of the  
414 regression slopes).

415 These speculations are important for farmers who are currently highly constrained in their land use,  
416 and calls for setting up a new system to monitor and reassess the state of soil contamination, which  
417 is not provided for in the current monitoring systems. With this aim in view, WISORCH\_V1 could be  
418 used to select the plots to be analyzed according to the foreseeable date at which CLD content  
419 dropped below the different thresholds stated above.

## 420 **4 CONCLUSION**

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421 Our study aimed to improve CLD modeling at a large scale based on field observations to provide an  
422 updated forecast of the persistence of pollution in southern Basse Terre, Guadeloupe. Our results  
423 showed that observed soil [CLD]<sub>s</sub> decreased faster than expected and simulation results revealed  
424 that, if this trend continues, CLD pollution would be reduced to 50 years in southern Basse Terre  
425 Guadeloupe.

426 Top-down modeling led us to implement a dissipation half-life DT50 of 5 years. The improved model  
427 makes it possible to reassess CLD contamination at a large scale with parsimonious data (carbon  
428 content and mean annual rainfall). Despite its lack of accuracy, estimations are consistent with a  
429 trend in the analysis of CLD in about 2545 plots in southern Basse Terre, confirming the interest of  
430 such an approach.

431 These results considerably modify our perception of pollution in the future, as CLD contamination  
432 was considered by land managers to be stable. Such a model can help envisage an end to CLD  
433 contamination. Model [CLD]<sub>s</sub> assessment remains a convenient and cost-efficient way to organize  
434 and prioritize additional soil analyses in order to check their contamination status, which is a  
435 prerequisite for increasing the range of possible land uses.

436 Still, given the high uncertainty of predictions, preliminary tests are required. To confirm our  
437 observations, new soil campaigns should be carried out on plots that have already been sampled for  
438 CLD analysis in the past. Further research is also needed to better understand the processes  
439 underlying CLD degradation and mobility.

## 440 **5 DECLARATION**

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441

## 442 **6 ETHICS APPROVAL AND CONSENT TO PARTICIPATE**

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443 Not applicable

## 444 **7 CONSENT FOR PUBLICATION**

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445 Not applicable

## 446 **8 AVAILABILITY OF DATA AND MATERIALS**

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447 The datasets used and/or analyzed in the present study are available from the corresponding author  
448 on reasonable request.

449 All data generated or analyzed during this study are included in this published article [and its  
450 supplementary information files].

## 451 **9 COMPETING INTERESTS**

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452 All the authors declare they have no actual or potential conflict of interest including financial,  
453 personal, or other relationships with other people or organizations that could inappropriately  
454 influence (bias) our work.

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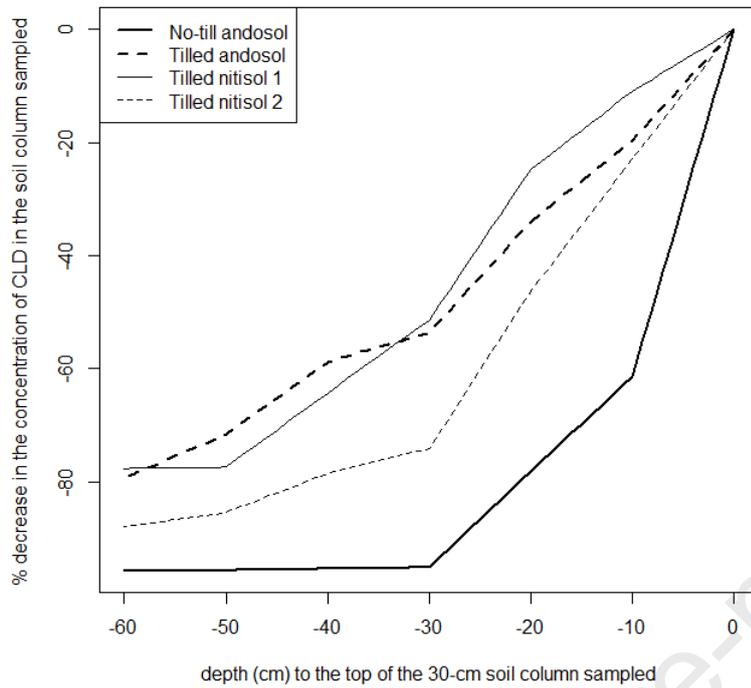
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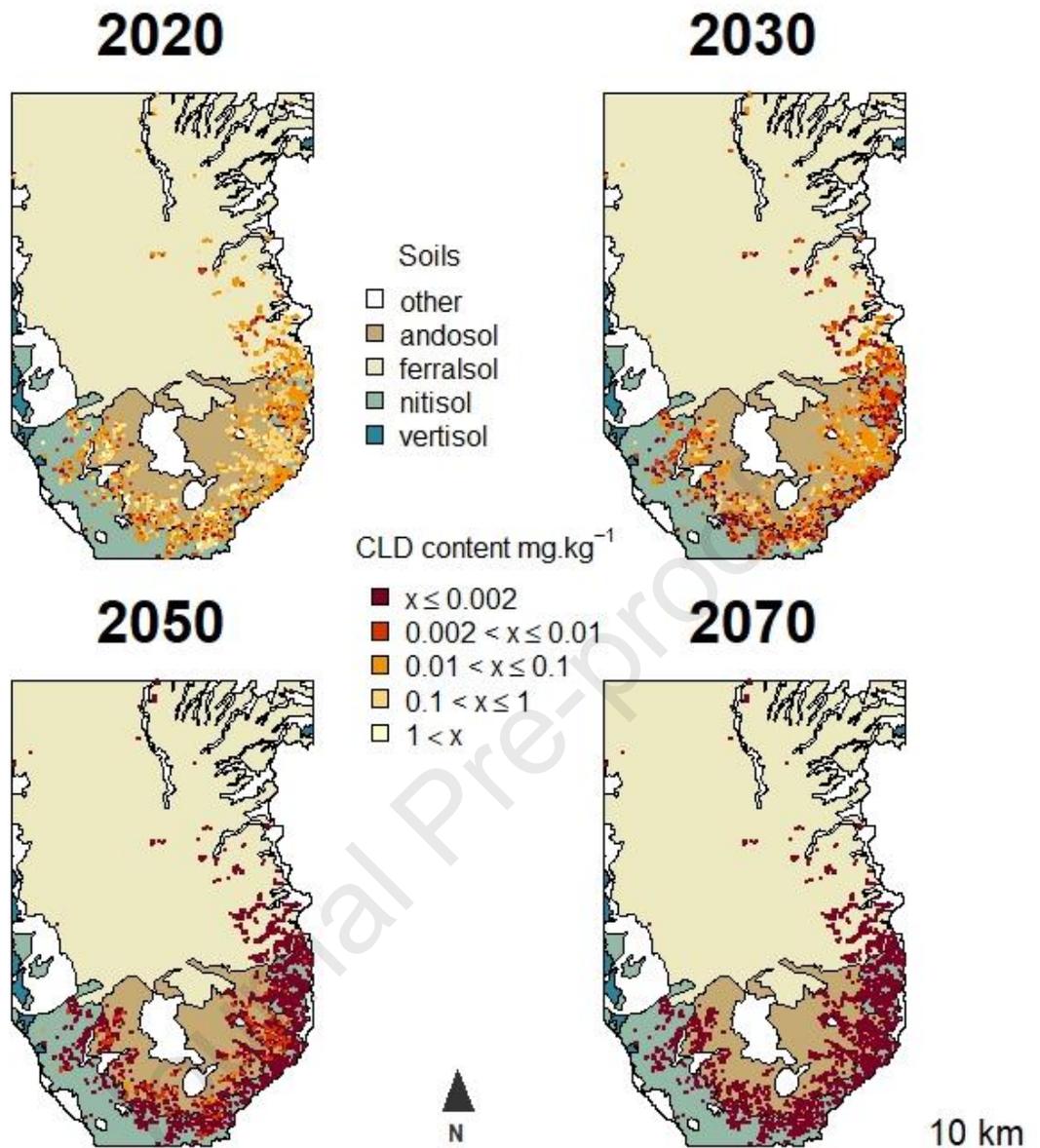


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594 *Figure 1: percentage decrease in [CLD]s at different depth, relative to the [CLD]s in the topsoil (0-30 cm).*  
 595 *Reading: a 30-cm soil column sampled at a depth of 10 cm (i.e. between 10 and 40 cm) has a mean [CLD]<sub>s</sub>*  
 596 *20% lower than the [CLD]<sub>s</sub> of a soil column sampled at the soil surface (i.e. in the 0-30 cm layer) for a tilled*  
 597 *andosol (about 60% for a no-till andosol).*

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600 *Figure 2: Prediction of [CLD]s for plots in high risk areas in 2020, 2030, 2050 and 2070 according to four*  
 601 *thresholds: 1 and 0.1  $\text{mg.kg}^{-1}$  the thresholds below which it is possible to grow plants with intermediate or*  
 602 *high sensitivity to CLD, respectively; 0.01  $\text{mg.kg}^{-1}$  the limit of quantification, 0.002  $\text{mg.kg}^{-1}$  the limit of*  
 603 *detection. Above 1  $\text{mg.kg}^{-1}$  only plants with low sensitivity to CLD can be grown.*

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## Highlights

- Chlordecone (CLD) pollution in FWI was previously estimated to last for centuries
- Field observations improved CLD modeling in a top-down modeling approach
- a DT50 of 5 years allowed us to simulate the observed decrease of CLD in soils
- With this unexpected decrease rate, soil decontamination is forecast for the 2070s
- This calls for re-evaluation of soil pollution status, not envisaged until now

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**Declaration of interests**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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