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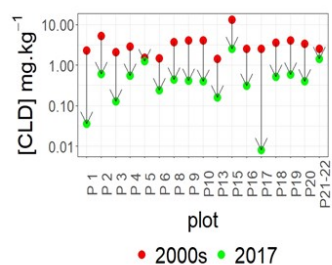
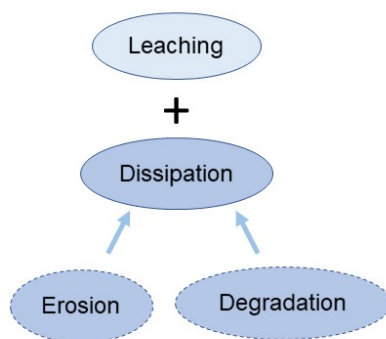
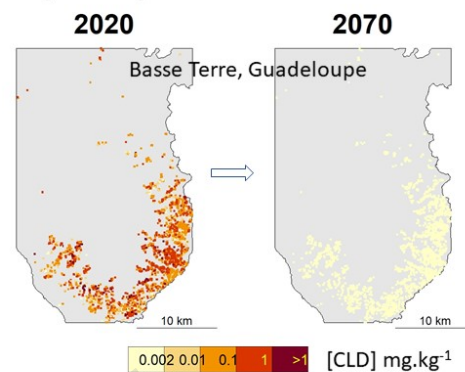
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1 **Author contributions**

- 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 Cattani: 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**

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

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Abstract

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

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

1 INTRODUCTION

Organochlorine compounds have been used all over the world (Loganathan and Lam, 2012). Despite being banned or restricted, some molecules continue to contaminate soils, water bodies and food chains due to their persistence in the environment (Benoit et al., 2020; Falandysz et al., 2022; Jones and de Voogt, 1999; Nakata et al., 2002). This is the case in the French West Indies, where chlordecone (CLD, C₁₀Cl₁₀O; CAS N° 143-50-0; 491 g.mol⁻¹), a toxic organochlorine insecticide was extensively applied in banana fields between 1972 and 1993 (Lesueur Jannoyer et al., 2016).

Although it is known to be particularly stable, the long-term fate of these molecules remains poorly understood.

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

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

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

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

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

The objective of this study was to improve CLD modeling at a large scale. Starting from data on $[CLD]_s$ available from 2001 to 2020, we tested other processes that could be involved in changes in $[CLD]_s$ and thus improve the structure of the model of Cabidoche et al. (2009). We first proceeded with a statistical analysis of the $[CLD]_s$ database that contains all CLD soil analyses performed since 2001 in southern Basse Terre, Guadeloupe, as part of the monitoring program set up by public authorities. Second, we characterized in-situ changes in $[CLD]_s$ between two periods using field measurements. A new model, WISORCH_V1, was then derived from the comparison between field and simulated values. A final simulation, based on updated assumptions, was performed in the thousands of plots in the CLD database to provide updated estimates of the persistence of soil pollution by CLD. This work combines research and public monitoring in the medium-term and should help improve the management of the risk of CLD pollution.

2 MATERIAL AND METHODS

2.1 STUDY SITE

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

(saturated hydraulic conductivity $> 60 \text{ mm.h}^{-1}$) and andosols also have high carbon content (Levillain et al., 2012). In our study, we considered plots at two scales:

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

2.2 RAINFALL DATA

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

At the scale of the Pérou hydrosystem, the annual rainfall for each experimental plot from the beginning of the chlordecone use in 1972 to the present was derived from data recorded by the reference station of Neufchâteau, by applying a correction depending on the altitude of the plot. Neufchâteau station (160403800 N, 613600400 W, 250 m amsl) is located 2.6 km from the Pérou river (SM_Figure2), with an acquisition rate of 97.67%. Missing data were estimated using a rainfall typology (SM Appendix A).

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

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

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 weather station

2.3 COLLECTION OF DATA ON CLD CONCENTRATIONS IN THE SOIL

2.3.1 Soil CLD database in southern Basse Terre

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

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

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

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

2.3.2 Soil sampling campaign in the Pérou hydrosystem

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

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

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

In the 2017 campaign, at each sub-sampling point, bulk density (BD) was measured at the soil surface after removing the first centimeters of dead plant cover using a 100 cm³ stainless-steel cylinder. The

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

2.3.3 Soil profile campaign in 2010: CLD distribution with depth

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

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

2.4 PESTICIDE ANALYSIS

The selected LDA26 is accredited by Cofrac, the French Accreditation Committee for pesticide analyses thereby guaranteeing their technical skills and reliability as well as good management practices. LDA26 complies with ISO 17025 standards for testing and calibration. The concentration of CLD, CLD-5b-hydro and chlordécol were determined according to the French standard AFSSA/ LERQAP/TOP POP04 based on Blanke et al. (Blanke et al., 1977), the routine procedure (internal procedure CMO_MT06: quantification of organochlorine and organophosphorous pesticides based on the norm XP X 33-012)(IPCS, 1990).

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

spectrometry ensured that analytes did not interfere in the analysis of chlordecone. Internal injection standards were Chlordecone C13 and 2.4D d3. The limit of quantification was 0.01mg.kg⁻¹.

2.5 CALCULATION.

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

The relative decrease in [CLD]_s between two dates was computed according to Eq. 2:

$$\text{Eq. 2: } \text{relative decrease} = ([CLD]_{si} - [CLD]_{sj}) / [CLD]_{si} = 1 - 1 / \exp(a * (T_i - T_j))$$

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

Annual rate of [CLD]_s decrease in the 17 plots was calculated from [CLD]_s measured at two dates according to the following sequence:

- Calculation of x, the annual rate of [CLD]_s decrease from Eq. 3;
- then, knowing x, calculation of the relative decrease at the desired date tx with Eq. 4

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

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

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

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

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

2.6 WISORCH MODEL

In the initial version of the WISORCH model, called WISORCH_V0, and described by Cabidoche et al. (2009), variations in the concentrations of the pesticide in the soil dS (kg.ha⁻¹) were a function of the remaining stock of that pesticide in the soil, S (kg.ha⁻¹), and of the amount of percolating water dD (mm), as described by Eq. 5:

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

where C1 is the percolation coefficient and equals 10 / (Koc × SSOC); Koc (mL.g⁻¹) is the partitioning coefficient between the sorbed part of the pesticide on soil organic carbon and the dissolved part in water; SSOC (t.ha⁻¹) is the stock of organic carbon calculated with Eq. 6.

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

where SOC (g/100g) is the soil organic carbon, BD is bulk density in T.m⁻³, and Vol_s is the soil volume (m³) of 1 ha = 0.3 m × 10 000 m² (0.3 m being the sampling depth).

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

First, we ran the WISORCH_V0 model on each plot in our dataset using the initial parameter values provided by Cabidoche et al. (2009) or in-situ measurements (Table 1). The initial CLD stock S_0 was calculated with Eq. 7 using the [CLD]_s analyzed during the first campaign: In this case, S_0 is equivalent to the initial, unique, and uniform CLD application at the sampling date.

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

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

Table 1: parameter values in simulations

Parameters	Initial values	Calibration
Koc nitisol (L.kg^{-1})	2 000	2 000; 3 000
Koc andosol (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) %

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

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

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

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

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

3 RESULTS AND DISCUSSION

3.1 UNEXPECTED DECREASE IN THE OBSERVED CONCENTRATIONS OF CLD IN THE SOIL OVER TIME

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

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

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

3.1.2 Substantial decrease in soil CLD concentrations between the two soil campaigns at the local scale

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

The [CLD]_s was higher in andosols than in nitisols (respective median of 3.45 and 2.3 mg.kg⁻¹, p value of 0.11 in Campaign 1; median of 0.532 and 0.24 mg.kg⁻¹, p value of 0.048 in Campaign 2 according to a Wilcoxon test). In agreement with Cabidoche et al. (2009), this result was expected and may be partially linked to the high soil carbon content of andosols compare to nitisols (mean SOC of 4.28% for andosol vs 2.2% for nitisol, see Table 2). However, there was no significant effect of the type of soil on the decrease in [CLD]_s 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 Chlordocol (p value >
282 0.2).

Journal Pre-proof

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

Plot ID	Soil type	Land use in 2017	Altitude (m)	Bulk density (g.cm ⁻³)	SOC 2017 (%)	1st Campaign (2005-2012)			2nd Campaign (2017)			chlordecol (µg.kg ⁻¹)	CLD decrease (%)
						Sampling year	CLD (µg.kg ⁻¹)	5b hydro chlordecone (µg.kg ⁻¹)	Sampling date	CLD (µg.kg ⁻¹)	5b hydro chlordecone (µg.kg ⁻¹)		
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

3.2 INCONSISTENCY BETWEEN OBSERVED AND VALUES PREDICTED BY

WISORCH_V0

First, the observed decrease in $[CLD]_s$ was much bigger than expected according to the simulations performed with the first version of WISORCH model (WISORCH_V0). Indeed, for the 17 plots, simulated values of $[CLD]_s$ for 2017 were, on average, 4-fold higher than the observed values (slope of 4.21, r^2 of 0.75, p value less than 0.001).

Secondly, the results of the WISORCH_V0 calibration (4200 simulations; SM_Figure 4), show that the leaching process alone cannot lead to decreases as large as those observed in 2017. In fact, the case that maximized r^2 with a slope of 1 was obtained for a rainfall coefficient of 1.2 and a SOC equal to half the observed value. These results suggest that, in order to obtain good results, the parameters must be distorted in such a way that they no longer correspond to what is observed in the field in terms of climate conditions and field properties: systematic overestimation of SOC by a factor 2 (measured SOC / SOC simulation = 2), and systematic underestimation of the annual rainfall by 17% (measured rainfall / rainfall simulation = 83%). Additionally, the Koc for andosol has to be set to 2,000, i.e., the minimum value for andosols and the same value as for nitisols.

Few references exist concerning the mobility of CLD in the soil profile. Ollivier et al. (Ollivier et al., 2020) found that CLD percolation ranged from 13.9% to 15.1% of the stock in experimental conditions equivalent to 3.2 years of percolation and 5,340 mm of rainfall on a nitisol. With the same percolation flux, WISORCH_V0 yielded higher losses (37%) with the initial parameters presented in section 2.6 for a nitisol. On the other hand, the losses calculated over the same period from data measured in the 17 plots (see Eq. 3 and Eq. 4 in section 2.5) yield a mean decrease of 60% on nitisol and 50% on andosol. The gap between these and previous decreases suggests other processes than leaching are involved.

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

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

A new version WISORCH_V1 was then derived from WISORCH_V0. Learning from data and model tests, we added a new process of CLD dissipation in WISORCH_V1. Here, we define dissipation as the overall process leading to the eventual disappearance of substances from the site at which it was applied or an environmental compartment. It includes transport and transformation processes (OECD, 2016). We

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

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

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

After integration this gives:

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

where $S(0, T_a)$ is the quantity of pesticide applied at application a . $T-T_a$ is the time elapsed (in days) between 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 of percolating water from T_a to T (Eq. 10).

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

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

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

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

WISORCH_V1 was calibrated with the parameter values described in section 2.6 to which we added DT50 values ranging from 2 to 50, according to the literature (ATSDR, 1995; National Center for Biotechnology Information, 2021). By running the resulting 33,600 simulations (SM Figure5), we identified the 738 best simulations characterized by: $0.8 < \text{slope} < 1.2$ and $r^2 > 0.975 \times \max(r^2)$. In this group, 99% of simulations present a DT50 of 5 years (SM_Table1). Additionally, Koc value for andosol is less than $17,000 \text{ mL.g}^{-1}$ with a maximum frequency for the value of 5,000 (54%). The distribution of the other parameters does not show such heterogeneity in distribution frequency, which means that they are less sensitive and therefore have less impact on the quality of the simulation.

3.4 ASSESSMENT OF TWO MAIN PROCESSES INVOLVED IN CLD DISSIPATION

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

3.4.1 Erosion

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

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

The soil profiles carried out in 2010 in the Pérou watershed showed that $[\text{CLD}]_s$ decreased rapidly with depth (SM_Figure6). Although the mobilization of CLD in deeper horizons may be slightly increased because of tillage, most CLD remained in the topsoil. To evaluate the effect of different soil stripping intensities due to erosion on the CLD concentration measured, using these profiles, we calculated the concentration of CLD in a 30-cm soil column taken from different depths compared to the current measurement in the 0-30 cm soil layer. According to Figure 1, it would require a soil stripping of at least 20 cm to reduce the $[\text{CLD}]_s$ by 80% (i.e. the average decrease observed in the 17 plots) in the case of a no-till andosol, and of 50 cm or more for other soils. Retaining the above values 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 initial concentration prior to stripping in a no-till andosol and more than 95% in other soils. Thus, the surface erosion process alone is unlikely to be responsible for the decrease in $[\text{CLD}]_s$ in the 17 experimental plots.

3.4.2 Degradation

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

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

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

3.5 PREDICTION OF THE DURATION OF POLLUTION IN SOUTHERN BASSE-TERRE, GUADELOUPE

The same dataset as that used in section 3.1.1 was used to predict [CLD]_s at different dates using WISORCH_V1. For each plot, the dataset was completed with mean annual rainfall by spatial intersection of the plot layer with the isohyet layer of the mean annual rainfall between 1929 and 1978 (L'Hôte, 1981). This mean annual rainfall value was used for all the simulated years. The values of the soil parameters SOC and BD were not available in the DAAF dataset, and were consequently set according to Levillain et al. (2012): 2% SOC for nitisols, 3% for ferralsols and 7% for andosols; BD of 1.10 T.m⁻³ for nitisols and ferralsols and 0.55 T.m⁻³ for andosols. Koc values were set according to our own simulation results: 2,000 L.kg⁻¹ for nitisols and ferralsols, and 5,000 L.kg⁻¹ for andosols. The runoff coefficient was set at 0.06 (Cabidoche et al., 2009) and the DT50 at 5 years.

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

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

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

These speculations are important for farmers who are currently highly constrained in their land use, and calls for setting up a new system to monitor and reassess the state of soil contamination, which is not provided for in the current monitoring systems. With this aim in view, WISORCH_V1 could be used to select the plots to be analyzed according to the foreseeable date at which CLD content dropped below the different thresholds stated above.

4 CONCLUSION

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

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

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

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

5 DECLARATION

6 ETHICS APPROVAL AND CONSENT TO PARTICIPATE

Not applicable

7 CONSENT FOR PUBLICATION

Not applicable

8 AVAILABILITY OF DATA AND MATERIALS

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

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

9 COMPETING INTERESTS

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

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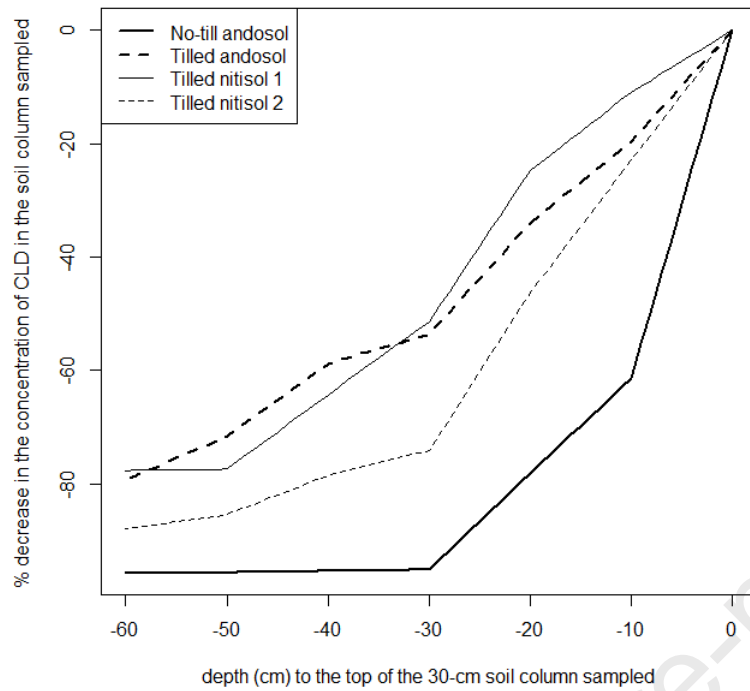


Figure 1: percentage decrease in [CLD]_s at different depth, relative to the [CLD]_s in the topsoil (0-30 cm).
 Reading: a 30-cm soil column sampled at a depth of 10 cm (i.e. between 10 and 40 cm) has a mean [CLD]_s
 20% lower than the [CLD]_s of a soil column sampled at the soil surface (i.e. in the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

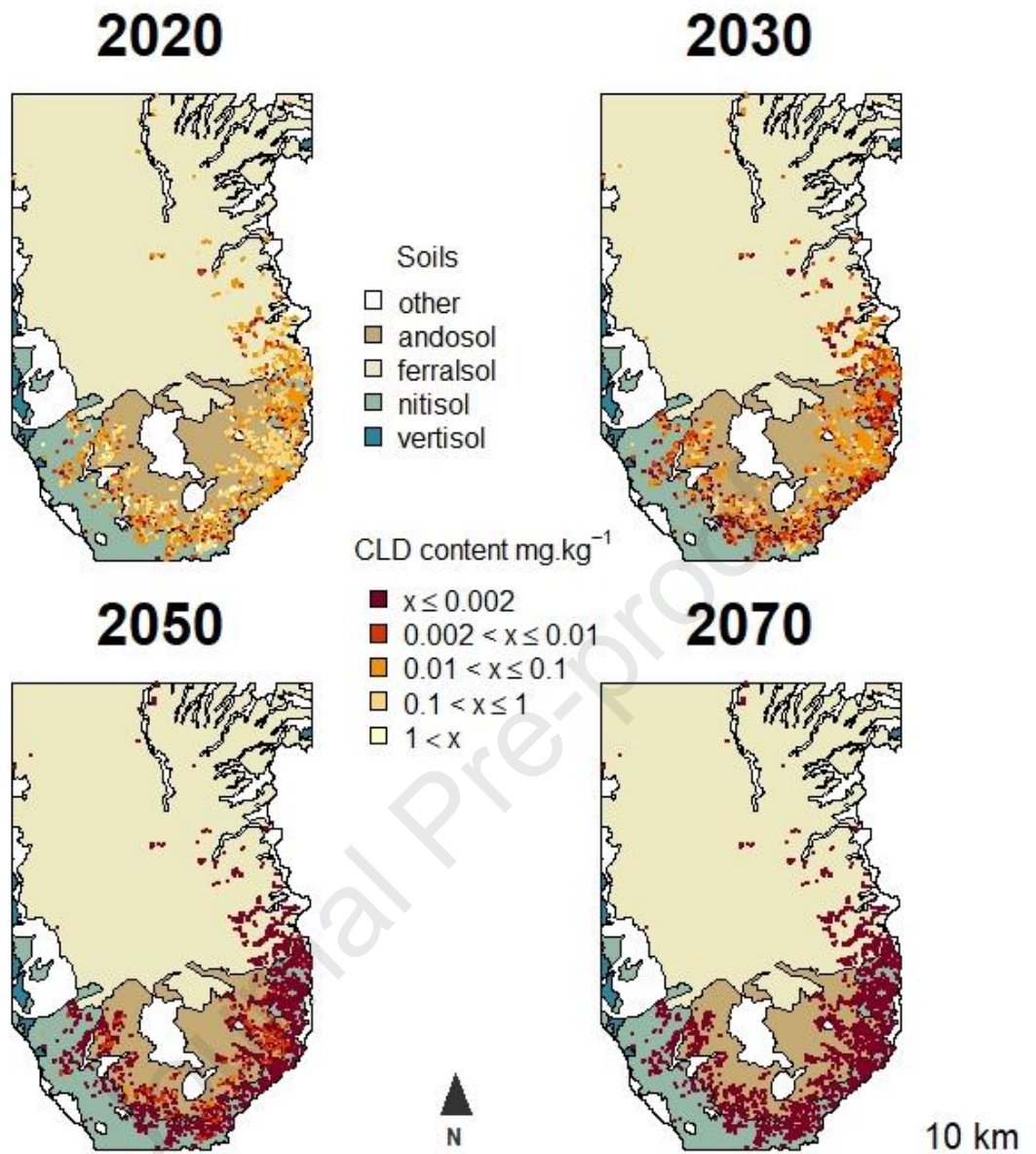


Figure 2: Prediction of [CLD]s for plots in high risk areas in 2020, 2030, 2050 and 2070 according to four thresholds: 1 and 0.1 mg.kg⁻¹ the thresholds below which it is possible to grow plants with intermediate or high sensitivity to CLD, respectively; 0.01 mg.kg⁻¹ the limit of quantification, 0.002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

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

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: