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## Is a dissipation half-life of 5 years for chlordecone in soils of the French West Indies relevant?

Pierre-Loïc Saaidi, Olivier Grünberger, Anatja Samouëlian, Yves Le Roux, Antoine Richard, Damien Devault, Cyril Feidt, Olivier Evrard, Pierre Benoit, Gwenaël Imfeld, et al.

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1 **Is a dissipation half-life of 5 years for chlordecone in soils of the French West Indies relevant?**

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5

6

7 **Highlights**

8

9 - The marked apparent decrease of chlordecone in Guadeloupe soils is questionable

10

11 - The dataset by Comte et al (2022) is not suitable for optimizing a predictive model

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13 - A dissipation half-life of 5 years for chlordecone in FWI soils is not realistic

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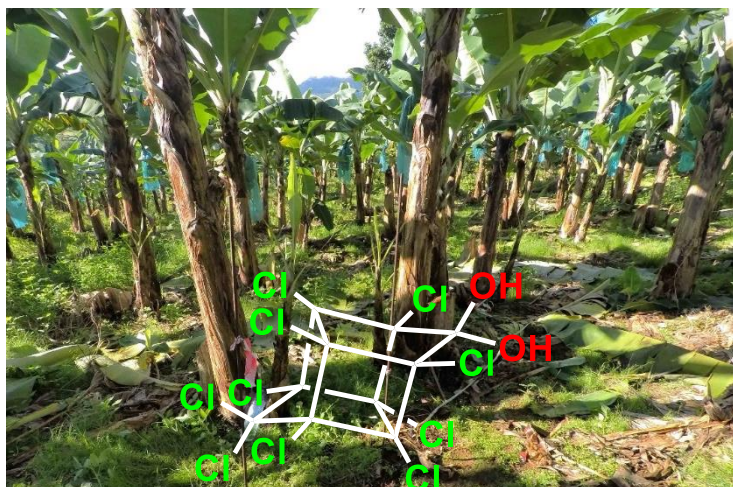
15 - The long-term persistence of Chlordecone should remain the most likely hypothesis

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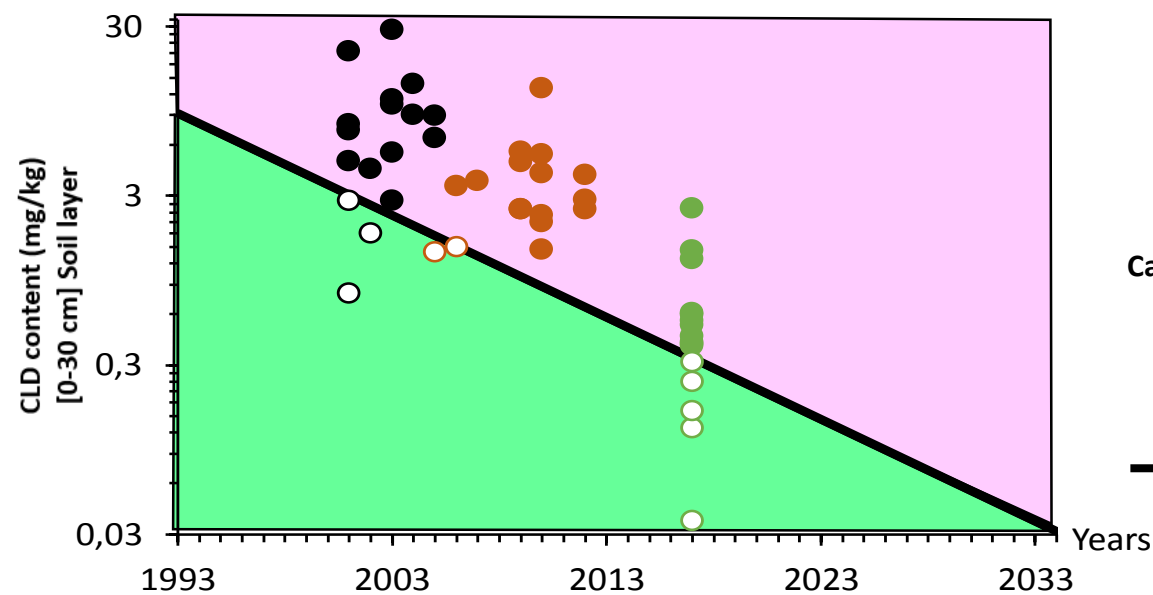
# Is a dissipation half-life of 5 years for chlordecone in soils relevant ?



**Dissipation includes:**

- ✓ transport
- ✓ (bio)degradation

**Chlordecone concentrations in soils: on-field measurements versus simulated upper limit based on a dissipation half-life of 5 years**



- Comte et al., (2022)**
- Andosols & Nitisols [2005-2012]
  - below
  - above the limit
  - Andosols & Nitisols in 2017
  - below
  - above the limit
- Cabidoche et al., (2009)**
- Andosols & Nitisols [2005-2012]
  - below
  - above the limit
- Simulated upper limit of CLD concentrations using a dissipation half-life value of 5 years (Comte et al., 2022)

40 **Abstract**

41 Recently, Comte et al. (2022) re-examined the natural degradation of chlordecone (CLD) in the soils of the French  
42 West Indies (FWI) by introducing an additional ‘dissipation parameter’ into the WISORCH model developed by  
43 Cabidoche et al. (2009). Recent data sets of CLD concentrations in FWI soils obtained by Comte et al. enabled  
44 them optimizing the model parameters, resulting in significantly shorter estimates of pollution persistence than  
45 in the original model. Their conclusions jeopardize the paradigm of a very limited degradation of CLD in FWI soils,  
46 which may lead to the entire revision of the management of CLD contamination. However, we believe that their  
47 study is questionable on several important aspects. This includes potential biases in the data sets and in the  
48 modeling approach. It results in an inconsistency between the estimated dissipation half-life time (DT50) value  
49 of five years determined by the authors and the fate of CLD in soils from the application period 1972-1993 until  
50 nowadays. Most importantly, rapid dissipation of CLD in the field as stated by Comte et al. is not sufficiently  
51 supported by data and estimates. Hence, the paradigm of long-term persistence of CLD in FWI soils is still to be  
52 considered.

53  
54 **Keywords:** chlordecone, persistent organic pollutant, soils, environmental fate, modeling, dissipation

55  
56 **Introduction**

57 Chlordecone (CLD) is a synthetic organochlorine insecticide extensively used in the French West Indies (FWI)  
58 between 1972 and 1993. CLD persistence in the environment and its biomagnification across the food chain  
59 remain major issues nowadays. The high impregnation rate (>90%) of the population (Anses, 2017) and the  
60 deleterious effects of CLD on human health (Multigner, 2018, Maudouit, 2019) have fostered strong local  
61 regulations over the last two decades. In this context, prediction models on the environmental fate of CLD may  
62 contribute to decision tools. In 2009, Cabidoche and colleagues proposed the parametric model WISORCH  
63 (Cabidoche, 2009). Among possible pathways of CLD dissipation, including leaching, (bio)degradation, transfer  
64 through erosion, the authors tested a simple elution model, which led to assigning CLD dissipation to leaching  
65 only. Interestingly, the authors reconstructed the history of CLD use based on farmer surveys and records from  
66 local Authorities, estimated dissipation parameters from a lysimetric experiment, and compared calculated and  
67 concentrations of CLD observed in an Andosol soil. Based on the model validated for Andosol, the authors  
68 concluded that physico-chemical or microbial degradation was insignificant. The calibration of  $K_{oc}$  (soil organic  
69 carbon/water partition coefficient) values allowed them to adapt the model for the other tropical volcanic soils

70 (i.e., Nitisol and Ferralsol) contaminated with CLD. The model estimates indicate that CLD pollution would last 6  
71 to 10 decades, 3 to 4 centuries, and 5 to 7 centuries in Nitisol, Ferralsol and Andosol, respectively. These  
72 conclusions, together with the relationship between CLD chronic exposure and prostate cancer over-incidence,  
73 eventually contributed to convince the French Authorities to launch the first Chlordecone National Action Plan  
74 in 2010.

75 Over the last decade, several studies have examined CLD degradation in laboratory conditions and in the FWI  
76 volcanic soils. The occurrence of CLD degradation, especially under aerobic conditions, would have significant  
77 implications. For instance, the duration of the soil pollution by CLD may be much shorter than the predictions  
78 made by Cabidoche et al, which would call for a complete revision of the exposure management of the population  
79 and ecosystems. Under aerobic conditions, apart from a few studies (Orndorff and Colwell, 1980, George and  
80 Claxton, 1988, Sakakibara, 2011, Amba Esegnet, 2019), for which the analytical quality can be questioned,  
81 incubation studies that used freshly added <sup>14</sup>C-CLD showed no or very limited degradation (Fernandez-Bayo,  
82 2013, Merlin, 2014, Francis and Metcalf, 1984). In contrast, recent laboratory studies evidenced biodegradation  
83 of CLD in liquid medium under anaerobic conditions (Chaussonnerie, 2016, Chevallier, 2019, Della-Negra, 2020,  
84 Lomheim, 2020, Lomheim, 2021, Hellal, 2021). It is worth mentioning that anaerobic conditions are rare in  
85 volcanic soils, mainly due to their high intrinsic permeabilities (eg. Saison, 2008) and location on steep slopes  
86 allowing efficient drainage and avoiding water logging. However, CLD transformation products were detected in  
87 significant concentrations in volcanic soils (Devault, 2016, Chevallier, 2019, Lomheim, 2020). Whether the  
88 transformation products are inherited from past or on-going degradation processes has still to be identified.

89 Recently, Comte et al. (2022) detected unexpected marked decreases of CLD concentration in FWI soils over  
90 time (Table 2 in Comte, 2022). They compared the original WISORCH model that accounts for CLD dissipation  
91 using only leaching with a new version 'WISORCH\_V1' that also includes a 1st-order temporal dissipation process  
92 to best simulate the observed variations of CLD concentrations over two sampling campaigns conducted on the  
93 same plots. Based on OECD recommendations (OECD, 2016), Comte et al. defined dissipation as "the overall  
94 process leading to the eventual disappearance of substances from the site at which it was applied or an  
95 environmental compartment. It includes transport and transformation processes". After discussing the possible  
96 drivers of CLD dissipation, the authors concluded that CLD degradation was the only mechanism responsible for  
97 the temporal dissipation process as implemented in WISORCH\_V1. The observed variations of CLD  
98 concentrations were best simulated using an estimated average degradation half-life time (DT50) of five years.

99 Comte et al. thus estimated that CLD pollution would end in the 2050-2070's. According to the authors, these  
100 results were also in agreement with the apparent decrease of CLD concentrations in FWI soils over time,  
101 extrapolated from a public database of 2545 spatially distinct plots analyzed each once over the 2001-2020  
102 period (Fig S3 in Comte, 2022).

103 These conclusions challenge previous findings highlighting the very low degradation of CLD in the FWI volcanic  
104 soils. Moreover, if confirmed, they may significantly modify the perception and management of CLD pollution in  
105 the FWI. Most importantly, it may give scientifically unfounded hope to local populations, already strongly  
106 affected by the food, health and environmental effects of the CLD pollution. In this respect, the conclusions of  
107 Comte et al. (2022) are formulated too firmly since, in our opinion, several aspects of their work should be  
108 reconsidered, thereby challenging the predicted outcomes and conclusions of their study.

109  
110 **1. About the data set of temporal variation of CLD concentrations in soil**

111 Comte et al. (2022) conclusions rely on the soundness and the relevance of CLD concentrations in soils from  
112 a public database (2001-2020; Fig S3 in Comte, 2022) and in a data set of 34 values they obtained from 17 distinct  
113 plots sampled twice between 2005 and 2017 (Table 2 in Comte, 2022). However, little information is provided  
114 about possible data biases and analytical controls related to CLD quantification in soil. There are two major  
115 sources of bias which, in our view, need to be verified before further use of the databases.

116 **- CLD spatial distribution and sampling protocol in the 17 plots:** CLD concentrations can vary by a factor of  
117 10 in the same plot as illustrated in Clostre et al. (2014, Fig 1). Clostre et al. concluded that CLD horizontal  
118 heterogeneity is higher under no-till or shallow-till practices and may vary with the applied cropping system and  
119 the field characteristics. Tillage also tends to homogenize CLD contamination at depth (>30 cm). To reduce the  
120 plot sampling fluctuation, Clostre et al. proposed in 2014 a decision tool support for soil sampling to achieve a  
121 precision of 30% in CLD assessment (Clostre, 2014, Fig 3). In Comte (2022), it is not fully clear whether the  
122 sampling protocol applied follows these recommendations, in particular for the samples taken before 2014  
123 included in Table 2. Indeed, the strong decrease in CLD concentrations observed between the two sampling  
124 periods (2005-2012 and 2017) could at least partially be explained by a lack of reproducibility or spatial  
125 representativity of the applied sampling protocol. Deep tillage practices (>30 cm) performed between the two  
126 sampling periods, thus diluting CLD in the soil depth, could also contribute to the observed decreasing trend as  
127 only the 0-30 cm layer has been analyzed. Hence, for such a study, we recommend to: i) provide additional details

128 on the plots, including till practices over time, ii) clearly specify whether the sampling protocol matches the  
129 published recommendations, iii) indicate the quality controls used to ensure the reproducibility of the sampling  
130 protocol repeated over the 12-year period, and iv) discuss possible biases with their consequences.

131 **- Analytical procedure:** In the articles from Cabidoche (2009), Clostre (2014), and Crabit (2016), CLD analyses  
132 in FWI soils were carried out by the LDA26, the same laboratory as the one cited in Comte et al. (2022). The  
133 protocol described in the three former articles mentioned the use of a GC-MS instrument and a series of non  
134 isotopic standards. The relative error and the limit of quantification were estimated to be 30% and 0.01 mg/kg,  
135 respectively (Clostre, 2014). The analytical protocol mentioned in the materials and methods section in Comte  
136 et al. describes the use of a LC-MS instrumentation and a <sup>13</sup>C-CLD internal standard. A quantification limit of 0.01  
137 mg/kg for CLD is given in the material and method section, whereas the authors indicate that they “focused on  
138 the plots located in the high-risk area (...), with [CLD]s higher than the detection threshold of 0.002 mg/kg.” The  
139 2001-2020 database (Comte, 2022; Figure S3) and the data set presented in Table 2 show the presence of CLD  
140 and 5b-monohydroCLD concentrations below the limit of quantification of 0.01 mg/kg from 2017 onwards. The  
141 first campaign (Comte, 2022; Table 2) was sampled in the Pérou River area from 2005 to 2012. It exactly  
142 corresponds to the sampling area studied by Crabit et al. 2016 during the period 2003-2012. We thus believe  
143 that the first campaign (Comte et al, 2022; Table 2) is part of the database compiled in Crabit et al. (2016).

144 We wonder whether or not two analytical protocols were used over the years: the oldest one previously  
145 described in Cabidoche (2009), Clostre (2014) and Crabit (2016), with a limit of quantification of 0.01 mg/kg using  
146 GC-MS instrumentation and the new one described in Comte et al. (2022) with a lower limit of quantification and  
147 the use of a LC-MS instrument. Additionally, the latter method allows to quantify chlordecol (CLDOH) (Comte,  
148 2022, Table 2), although the relative error of this method has not been reported. Even if this new analytical  
149 protocol probably offers lower relative error, an instrumental systematic bias/deviation may exist. The only inter-  
150 laboratory tests for CLD determination in soil reported so far mentions variations among laboratories (for both  
151 GC-MS and LC-MS methods) by a factor or two or more, depending on the soil type (Amalric, 2014). Before  
152 comparing data from the two distinct time periods (2001-2016 and 2017-2020), we strongly suggest to clearly  
153 relate the data to the analytical procedure applied and to discuss the controls used to limit analytical bias.

154 In addition, the presence of CLDOH in samples of the second campaign (Comte, 2022, Table 2) is proposed as  
155 a direct insight of CLD degradation. This may mislead the readers since this molecule has not been targeted in  
156 the initial analytical procedure used before 2017. Furthermore, CLDOH is systematically found as contaminant in

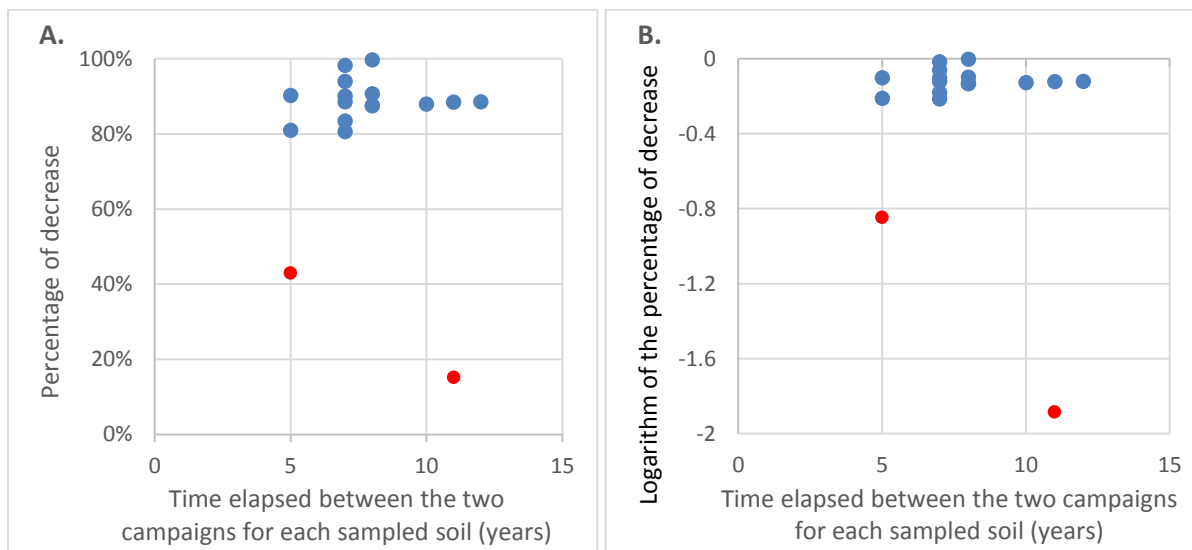


157 CLD commercial formulations (Kepone®, Curlone®) and known to be present in FWI soils at low levels (Chevallier,  
158 2019, Amalric, 2014). Lastly, to our knowledge, the formation of CLDOH has never been mentioned in any  
159 laboratory degradation of CLD reported so far. For all these reasons, the presence of CLDOH may not be a  
160 relevant indicator of CLD degradation in the environment.

161 **- About the interpretation of the public database (2001-2020):** The temporal variation of CLD concentrations  
162 in spatially distinct plots is actually quite irregular (Comte, 2022; Figure S3), in contradiction with the global  
163 decreasing trend stated by Comte et al. Whatever the type of volcanic soil considered, no significant change  
164 could be observed before 2017, although CLD concentrations markedly decreased after 2017 following a data  
165 gap (only six CLD concentrations measured during the 2014-2016 period according to Fig S3). This sudden  
166 decrease in CLD concentrations again suggests a change in the analytical protocol. Moreover, in Figure S3, we  
167 noticed the presence of values below the quantification limit mentioned by the authors for the 2017-2020 period.  
168 It contradicts the decision protocol applied by the authors (“[CLD]s ranging between the detection and the  
169 quantification threshold of 0.01 mg/kg were set to 0.05 mg/kg.”) Taking into account these lower values for 2017-  
170 2020 creates an artificial decreasing trend in CLD concentrations. Hence, we recommend discarding all data  
171 below 0.01 mg/kg from the second period (2017-2020) before testing any quantitative relationship. In addition,  
172 the possibility that sampling may have been oriented from the very beginning towards plots showing the highest  
173 risk of contamination needs to be addressed. Indeed, this bias, if confirmed, would also contribute to an  
174 ‘apparent’ decrease of CLD concentrations from 2001 to 2020.

175 **- About the data set based on the two sampling campaigns (2005-2017) (Comte, 2022, Table 2):** We agree  
176 with Comte et al. that the data set from Table 2, if not biased, demonstrates a decrease in soil CLD content  
177 between the two sampling campaigns. However, the marked amplitude of the decrease (-82% over 8 years on  
178 average) is not at all compatible with the observed evolution of CLD concentrations in FWI soils from its period  
179 of application (1972-1993) to nowadays. Indeed, the cumulative loading of CLD in FWI soils according to  
180 recommended practices (3 kg/year/ha) corresponds to theoretical maximum concentrations of CLD in the range  
181 of 10-15 mg/kg in 1993 as documented by Cabidoche et al. (2009, Table 2). More than one-third of the CLD  
182 concentrations (i.e., 35 of 96) reported in Andosol soils for the 2017-2020 period by Comte et al. 2022 (Fig S3)  
183 indicate concentrations between 1 mg/kg and 10 mg/kg. This corresponds to decreasing rates lower by a factor  
184 of 3 to 10 compared to the 12 Andosol plots (Table 2) used by Comte et al. to calibrate the WISORCH\_V1 model.  
185 Hence, these 12 plots may not be representative enough of the overall CLD fate in FWI Andosol soils.

186 Moreover, the following sentence raised our attention: “there were no significant effect of the length of the  
 187 period between the first and second campaign on the decrease”. While the authors observed that the CLD  
 188 decrease cannot be explained by the time elapsed between the sampling campaigns, this data set was used to  
 189 establish the temporal predictive model WISORCH\_V1. The absence of relationships between the time elapsed  
 190 between the two campaigns elapsed time and the decrease of CLD is illustrated in Figure 1.



191  
 192 *Figure 1: A. Evolution of the CLD decrease as a function of the time elapsed between the two sampling*  
 193 *campaigns (n=17); B. Evolution of the logarithm of the CLD decrease as a function of the time elapsed between*  
 194 *the two sampling campaigns (n=17); the apparent ‘outliers’ are marked in red.*

195 Using the Spearman test (most suitable for data not following the Normal distribution) no significant  
 196 relationship is observed between these series of values. Removing the apparent ‘outliers’ (2 Andosol plots among  
 197 the 12 Andosol plots used to set the WISORCH\_V1 model), it is possible to use the Bravais-Pearson test, which  
 198 requires data that follow the Normal distribution. Under these conditions, as with the Spearman test, no  
 199 significant correlation is observed. An experimental bias between the two campaigns, e.g. the change in the  
 200 analytical protocol, can explain both the global decrease of CLD concentrations between the two sampling  
 201 campaigns and the absence of relationship between the CLD decrease and the time elapsed between the two  
 202 campaigns. Overall, we believe that the robustness and accuracy of the data set is not sufficient to optimize a  
 203 parametric predictive model.

204  
 205 **2. About the construction and results of the WISORCH\_V1 model:**

206 Comte et al. modeled the dissipation as an overall process including a leaching process and a time-dependent  
 207 dissipation process mainly resulting from in situ CLD transformation. However, the DT50 values that were

208 estimated by the authors using their modeling approach refer only to the half-life values of the time-dependent  
209 dissipation process. Obviously, the overall DT50 of the whole dissipation kinetic will be even shorter since it must  
210 also include the leaching process that was thought to bring the main contribution to CLD elimination according  
211 to Cabidoche et al. (2009). In our opinion, eq. (8) and (9) in Comte et al. (2022) can be used to build a  
212 phenomenological multi-parameter model but the resulting 'apparent' dissipation coefficient should not be over-  
213 interpreted for the following reasons.

214 **- Erosion:** Based on a set of calculations and environmental measurements, Comte et al. (2022) concluded  
215 that "the surface erosion process alone is unlikely to be responsible for the decrease in CLD concentrations in  
216 the 17 experimental plots". However, their calculation assumed implicitly that CLD content is homogeneous over  
217 the 0-30 cm topsoil layer. This assumption is strong and its relevance depends whether the topsoil of each  
218 sampled plot was subjected or not to strong mixing by tillage practices. This is illustrated by Comte et al. (Fig S6),  
219 where a very marked difference (3-fold decrease in CLD concentrations) is observed between the 0-10 cm and  
220 the 10-30 cm topsoil layers in the case of no-till Andosol. Unfortunately, Table 2 did not provide details about  
221 the land use history including the tillage practices during the entire period for the 17 plots. If the topsoil had not  
222 been strongly mixed by tillage, the possibility that the CLD content is highly heterogeneous within the 0-30 cm  
223 soil layer with very high contents in the upper centimeters of soil ought to be considered. In that case, there is  
224 no need to have a large erosion rate and a deep eroded soil layer to explain a large decrease in CLD storage in  
225 the 0-30 cm soil layer. Accordingly, it would have been useful to provide a sensitivity analysis of the model and a  
226 test of the model optimization results assuming, for example, different rates of CLD removal through erosion to  
227 provide uncertainty ranges for predictions. The occurrence of a marked CLD dissipation pathway through soil  
228 erosion and sediment transfer would be consistent with recent results by Sabatier et al. (2021). The authors  
229 observed an increase in soil erosion rates along with CLD contamination levels since the 1990s by studying  
230 sediment cores collected in coastal waters off the Pérou River (Guadeloupe) and the Galion River catchments  
231 (Martinique). Moreover, this study demonstrates the occurrence of significant sediment transfer and CLD along  
232 the land-to-sea continuum in response to changes in agricultural practices.

233 **- Optimization of the WISORCH V1 model:** Comte et al. explained how they determined the DT50 of 5 years:  
234 "By running the resulting 33,600 simulations, we identified the 738 best simulations (...). In this group, 99% of  
235 simulations present a DT50 of 5 years". In other words, Comte et al. used the model WISORCH\_V1 with a series  
236 of 12 couples of concentration values (12 plots analyzed at two distinct periods of time that include the two

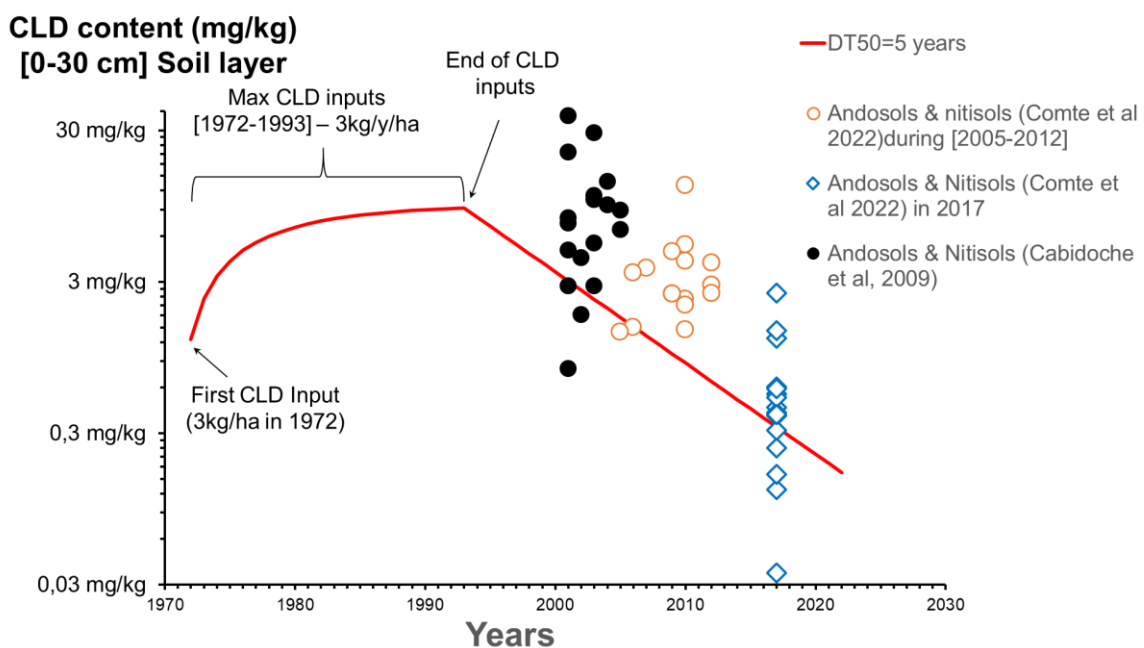
237 presumed 'outliers' presented in Figure 1a-b) for each of the seven parameters to be set (Comte, 2022; Figure  
238 S7), selecting the best fits. However, the WISORCH\_V1 model itself was not designed to optimize the best DT50  
239 for a given set of experimental data and no uncertainty was determined. Contrary to what has been done in  
240 Cabidoche et al., the sensitivity of the parameters (e.g. the DT50 value in WISORCH\_V1) on the model predictions  
241 has not been studied. Eventually, it is worth noting that the criteria used for identifying the best fit of the  
242 WISORCH models were the slope and the  $r^2$  of the regression between the simulated and measured soil CLD  
243 concentrations during the period between 2005 and 2017. Indeed these two criteria are useful to characterize  
244 model performance, although they do not inform on model bias. The mean error and the root mean square error  
245 would be more appropriate to evaluate prediction bias and variance. Hence, we question the robustness of the  
246 optimal model parameter values based on the calibration criteria chosen.

247 **- Relevance of the DT50 estimated for CLD:** Comte et al. used their estimated DT50 values to predict the  
248 potential rates of decrease of soil CLD concentrations in the future. It is thus worth verifying whether a dissipation  
249 half-life of five years also enables a realistic simulation of the temporal variation of CLD concentrations in soil  
250 from its application period between 1972 and 1993 until now. Accordingly, we computed a theoretical evolution  
251 in soil CLD concentration in the upper soil layer (0-30 cm) of a field plot from 1973 to 2020, taking into account  
252 only the first order temporal dissipation kinetics of CLD as calibrated by Comte et al. We expected that the  
253 predictions of CLD concentrations in soil would overestimate the actual data since in the field leaching does also  
254 contribute to the dissipation of CLD in the soil. Moreover, we considered a continuous treatment of 3 kg/ha/year  
255 during the full period of CLD use [1972-1993] (Cabidoche, 2009), which maximizes the amounts of CLD inputs,  
256 and in turn, also maximizes the predicted CLD concentrations in soil. Indeed, the CLD treatments were likely  
257 interrupted a few years in several plots due to re-plantations, turnover of cultivations or other reasons. The  
258 yearly degradation of the soils stocks was computed as follows:

259 
$$C_{y+1} = C_y * e^{-\frac{\ln(2)}{DT50}} + T_y$$
 where  $y$  stands for the year;  $T_y$  is the CLD treatment for year  $y$  with  $T_y = 3$  kg/ha  
260 for  $y \in [1972-1993]$  and  $T_y = 0$  kg/ha for  $y \in [1994-2020]$ ;  $C_y$  is the CLD soil concentration for year  $y$ .

261 The applied CLD amounts were converted in mg/kg of soil based on the assumption of a soil bulk density of  
262  $800 \text{ kg/m}^3$ ; that is an application of 3 kg/ha corresponds to an additional average soil CLD content of 1.25 mg/kg.  
263 Figure 2 shows the predicted variation of CLD concentration in soil with time. When confronting these results to  
264 the measured soil concentrations given in Cabidoche et al. (2009) (Table 2) and in Comte et al. (2022) (Table 2)

265 for Andosols and Nitisols of Guadeloupe, the vast majority of observations before 2017 (15 of the 17 values  
 266 available for the 2005-2012 period and 15 of the 18 values for the 2001-2005 period) had significantly greater  
 267 concentrations than the predicted values although the model was already meant to largely overestimate the  
 268 actual values. Only 6 of the 17 data observed in 2017 seem consistent with the predictions. This thus questions  
 269 the dissipation rate calculated by Comte et al. (2022), computed from samples taken from identical plots  
 270 between 2005 and 2017. At least, it appears that the estimated dissipation rate should not be extrapolated to  
 271 other periods since it is not suitable for retrospectively simulating soil CLD content changes since the start of CLD  
 272 application.



273  
 274 *Figure 2: Evolution of the predicted CLD maximum concentration (red curve) in the upper layer soil for a DT50*  
 275 *of 5 years compared to Andosols and Nitisols concentrations in mentioned papers. CLD inputs are maximized by*  
 276 *assuming a treatment every year between 1972 and 1993. A logarithmic scale was used for the CLD soil*  
 277 *concentrations.*

278 Although the WISORCH\_V1 could not be optimized for the Nitisol nor the Ferralsol due to the lack of data,  
 279 Comte et al. concluded for these two soils to the same DT50 as for Andosol, and used this value to run the  
 280 WISORCH\_V1 model on the whole data set from 2001 to 2020 to predict the fate of CLD over the next several  
 281 decades. Comte et al. argued that the comparison of the trends observed for the 2001-2020 between the three  
 282 types of soils did not significantly differ (Comte, 2022, Fig S3). However, considering the above-mentioned biases  
 283 about the public database, and the well-documented stronger sorption of CLD in Andosol than in other soil types  
 284 (Comte, 2022; Cabidoche, 2009; Fernandez-Bayo, 2013), using the same DT50 of 5 years to all soil types may be

285 incorrect. Predictions of CLD fate in Nitisol and Ferrasol soils established by Comte et al. should thus be viewed  
286 with even more caution than that obtained for Andosol soils.

287  
288 **Conclusion**

289 The DT50 of 5 years for CLD and other outcome data of WISORCH\_V1 clearly diverge from all previous studies  
290 and observations made on the fate and degradation of CLD in contaminated soils. Here, we highlighted a number  
291 of potential biases in the data and analyses. Although Comte et al. provided an insightful contribution to evaluate  
292 CLD dissipation in the environment, we conclude that the data set, the model and their conclusions raise several  
293 serious issues. As a result, the estimated dissipation half-life time (DT50) value determined by the authors  
294 contradict the observed fate of CLD in soils from the application period 1972-1993 until nowadays. We showed  
295 that the conclusion by Comte et al. of a fast dissipation of CLD in soils is not sufficiently supported by field data  
296 and model estimates. Hence, the paradigm of long-term persistence of CLD in FWI soil should remain the most  
297 likely hypothesis. The issues raised here may help to define in the future the criteria for more robust and reliable  
298 predictions of CLD dissipation in soil, starting from the field experimental design and sampling protocols. A  
299 reliable experimental design should thus include:

- 300 - A spatial sampling design that specifically aims to detect the temporal changes of mean soil CLD content with  
301 an expected accuracy (see Papritz and Webster, 1995a and b for theoretical aspects in designing best spatial  
302 sampling designs for estimating changes in soil properties). Such a design differs from the one used to determine  
303 with an expected accuracy the mean CLD content at a single given time;
- 304 - The analysis of several individual or bulked samples across each sampled plot in order to estimate the actual  
305 variance of the mean CLD concentration and the temporal variation of the mean CLD concentrations for each  
306 field;
- 307 - An analysis of CLD distribution across depth (0 - 100 cm) for a representative number of plots repeated over  
308 time to improve contribution of percolation in CLD dissipation in soil;
- 309 - A dedicated study to improve the estimations on erosion processes in the FWI soils;
- 310 - A sufficient duration of the monitoring (e.g. 20 years, assuming a DT50 higher than 5 years);
- 311 - Detailed information regarding the history of tillage practices, amendments and changes of soil physico-  
312 chemical properties (e.g. pH, organic matter and allophane content);

- 313 - Repetitive samplings should be carried out within the same period of the year and at a higher frequency (i.e.  
314 every year or every two years) to generate a statistically relevant dataset for each plot;
- 315 - Statistical treatments of these databases would also highlight possible outliers, reflecting irregular temporal or  
316 spatial dissipation processes;
- 317 - Long time storage of numerous duplicate control soil samples should allow to: (i) validate previous CLD contents  
318 and discard the possible erroneous values by re-analyzing; (ii) validate new quantification methods; (iii) apply  
319 new analytical methods (e.g. to search for transformation products (Chevallier et al., 2019) and/or to measure  
320 stable isotope signatures of CLD to trace *in situ* degradation (Höhener et al., 2022)). This will however require to  
321 define the optimal storage conditions to avoid the degradation of CLD and its transformation products in the  
322 stored samples.
- 323 We also recommend to include for any new predictive model:
- 324 - A validation using a retrospective simulation accounting for historical CLD applications;
- 325 - A sensitivity study to provide reliable uncertainties associated with any predictive outcomes.

326

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