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

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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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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

12

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

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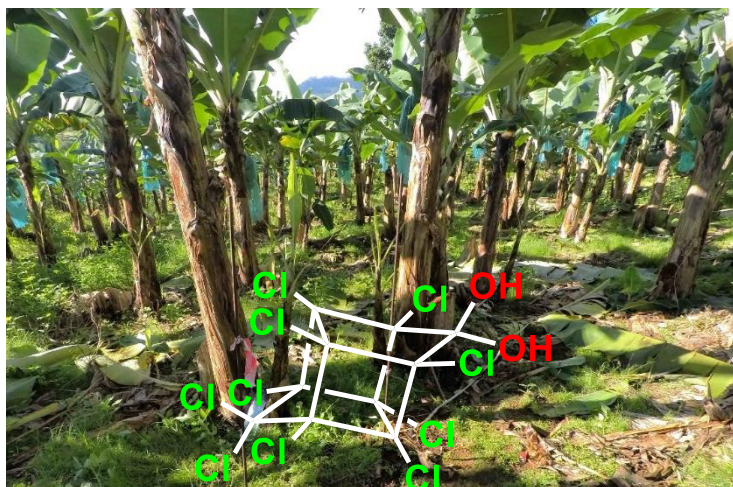
15 - The long-term persistence of Chlordecone should remain the most likely hypothesis

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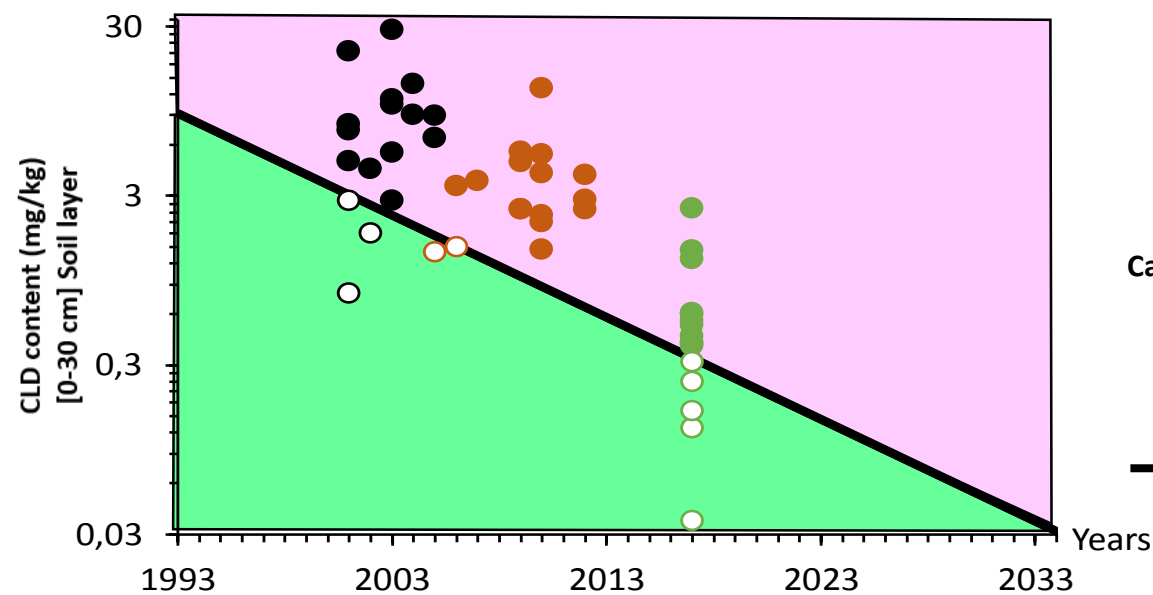
Is a dissipation half-life of 5 years for chlordecone in soils relevant ?



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 ¹⁴C-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 ¹³C-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.

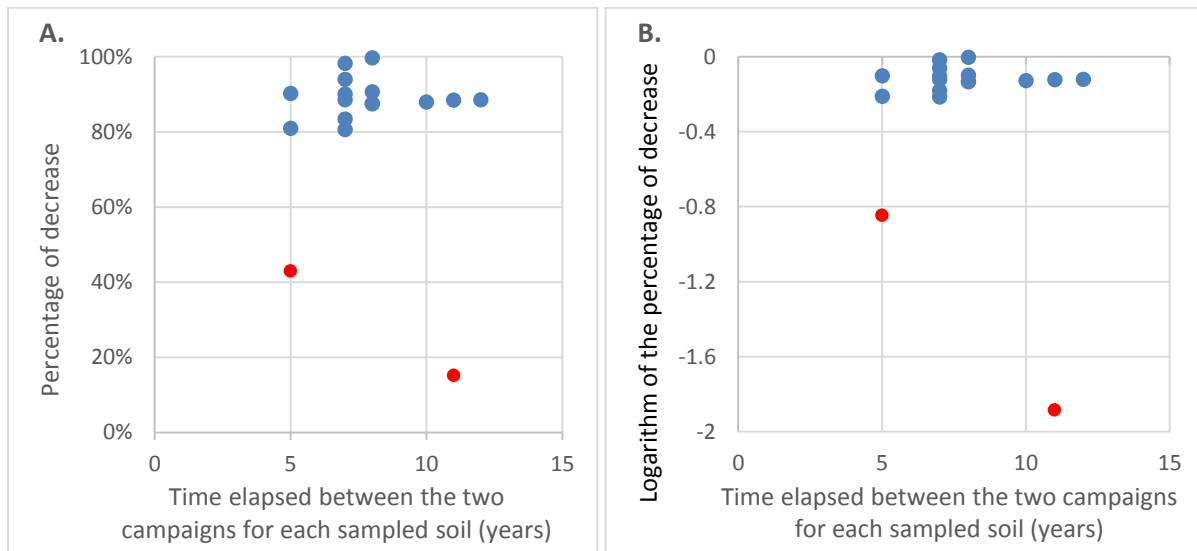
154 In addition, the presence of CLDOH in samples of the second campaign (Comte, 2022, Table 2) is proposed as
155 a direct insight of CLD degradation. This may mislead the readers since this molecule has not been targeted in
156 the initial analytical procedure used before 2017. Furthermore, CLDOH is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

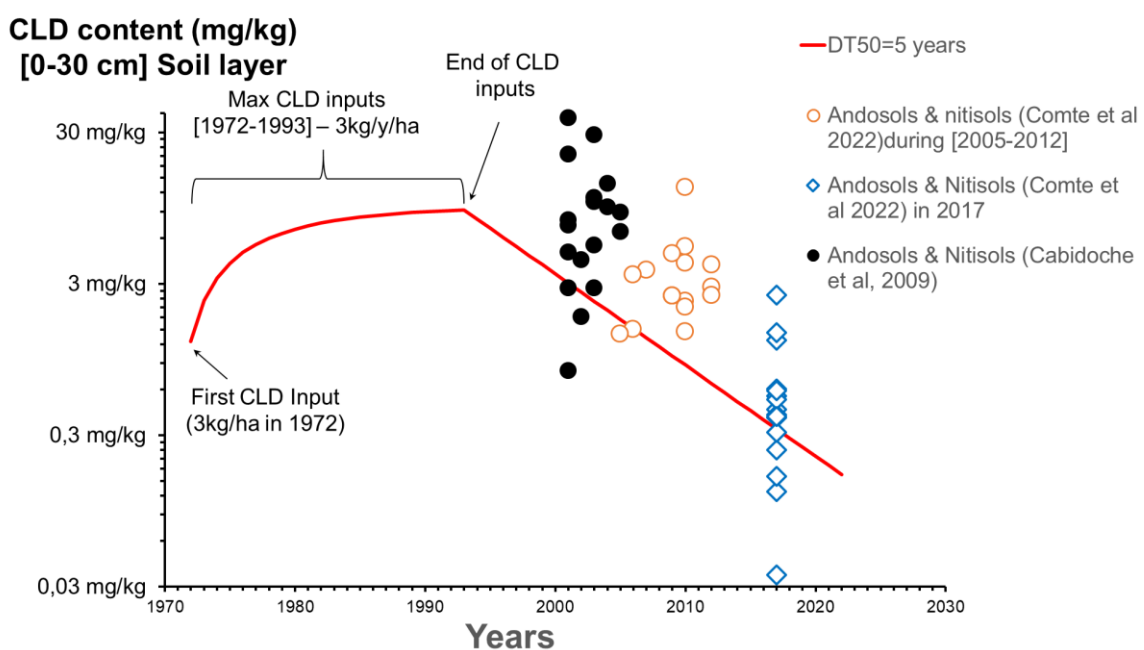
237 presumed 'outliers' presented in Figure 1a-b) for each of the seven parameters to be set (Comte, 2022; Figure
238 S7), selecting the best fits. However, the WISORCH_V1 model itself was not designed to optimize the best DT50
239 for a given set of experimental data and no uncertainty was determined. 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 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.

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