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

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Highlights

- The marked apparent decrease of chlordecone in Guadeloupe soils is questionable
- The dataset by Comte et al (2022) is not suitable for optimizing a predictive model
- A dissipation half-life of 5 years for chlordecone in FWI soils is not realistic
- The long-term persistence of Chlordecone should remain the most likely hypothesis

Graphical Abstract

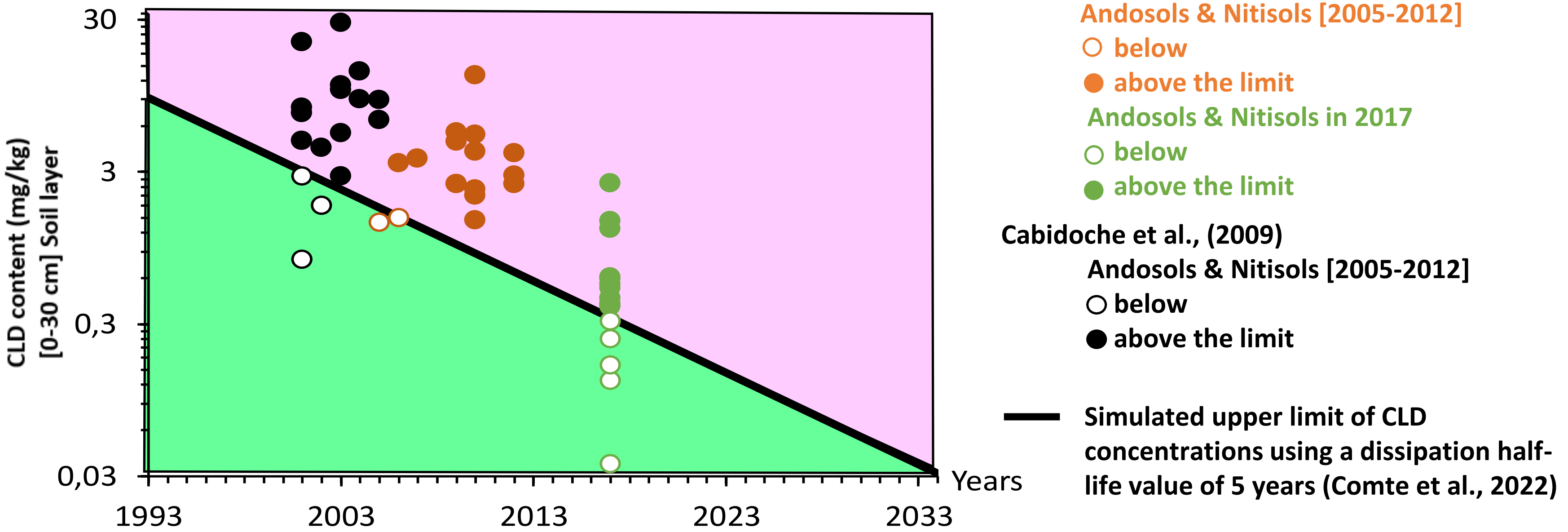
Is a dissipation half-life of 5 years for chlordecone in soils relevant ?

?

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



Abstract

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

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

Introduction

Chlordecone (CLD) is a synthetic organochlorine insecticide extensively used in the French West Indies (FWI) between 1972 and 1993. CLD persistence in the environment and its biomagnification across the food chain remain major issues nowadays. The high impregnation rate (>90%) of the population (Anses, 2017) and the deleterious effects of CLD on human health (Multigner, 2018, Maudouit, 2019) have fostered strong local regulations over the last two decades. In this context, prediction models on the environmental fate of CLD may contribute to decision tools. In 2009, Cabidoche and colleagues proposed the parametric model WISORCH (Cabidoche, 2009). Among possible pathways of CLD dissipation, including leaching, (bio)degradation, transfer through erosion, the authors tested a simple elution model, which led to assigning CLD dissipation to leaching only. Interestingly, the authors reconstructed the history of CLD use based on farmer surveys and records from local Authorities, estimated dissipation parameters from a lysimetric experiment, and compared calculated and concentrations of CLD observed in an Andosol soil. Based on the model validated for Andosol, the authors concluded that physico-chemical or microbial degradation was insignificant. The calibration of K_{oc} (soil organic carbon/water partition coefficient) values allowed them to adapt the model for the other tropical volcanic soils (i.e., Nitisol and Ferralsol) contaminated with CLD. The model estimates indicate that CLD pollution would last 6 to 10 decades, 3 to 4 centuries, and 5 to 7 centuries in Nitisol, Ferralsol and Andosol, respectively. These conclusions, together with the relationship between CLD chronic exposure and prostate cancer over-incidence, eventually contributed to convince the French Authorities to launch the first Chlordecone National Action Plan in 2010.

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

Recently, Comte et al. (2022) detected unexpected marked decreases of CLD concentration in FWI soils over time (Table 2 in Comte, 2022). They compared the original WISORCH model that accounts for CLD dissipation using only leaching with a new version 'WISORCH_V1' that also includes a 1st-order temporal dissipation process to best simulate the observed variations of CLD concentrations over two sampling campaigns conducted on the same plots. Based on OECD recommendations (OECD, 2016), Comte et al. defined 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". After discussing the possible drivers of CLD dissipation, the authors concluded that CLD degradation was the only mechanism responsible for the temporal dissipation process as implemented in WISORCH_V1. The observed variations of CLD concentrations were best simulated using an estimated average degradation half-life time (DT50) of five years.

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

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

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

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

- CLD spatial distribution and sampling protocol in the 17 plots: CLD concentrations can vary by a factor of 10 in the same plot as illustrated in Clostre et al. (2014, Fig 1). Clostre et al. concluded that CLD horizontal heterogeneity is higher under no-till or shallow-till practices and may vary with the applied cropping system and the field characteristics. Tillage also tends to homogenize CLD contamination at depth (>30 cm). To reduce the plot sampling fluctuation, Clostre et al. proposed in 2014 a decision tool support for soil sampling to achieve a precision of 30% in CLD assessment (Clostre, 2014, Fig 3). In Comte (2022), it is not fully clear whether the sampling protocol applied follows these recommendations, in particular for the samples taken before 2014 included in Table 2. Indeed, the strong decrease in CLD concentrations observed between the two sampling periods (2005-2012 and 2017) could at least partially be explained by a lack of reproducibility or spatial representativity of the applied sampling protocol. Deep tillage practices (>30 cm) performed between the two sampling periods, thus diluting CLD in the soil depth, could also contribute to the observed decreasing trend as only the 0-30 cm layer has been analyzed. Hence, for such a study, we recommend to: i) provide additional details on the plots, including till practices over time, ii) clearly specify whether the sampling protocol matches the published recommendations, iii) indicate the quality controls used to ensure the reproducibility of the sampling protocol repeated over the 12-year period, and iv) discuss possible biases with their consequences.

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

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

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

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

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

- About the data set based on the two sampling campaigns (2005-2017) (Comte, 2022, Table 2): We agree with Comte et al. that the data set from Table 2, if not biased, demonstrates a decrease in soil CLD content between the two sampling campaigns. However, the marked amplitude of the decrease (-82% over 8 years on average) is not at all compatible with the observed evolution of CLD concentrations in FWI soils from its period of application (1972-1993) to nowadays. Indeed, the cumulative loading of CLD in FWI soils according to recommended practices (3 kg/year/ha) corresponds to theoretical maximum concentrations of CLD in the range of 10-15 mg/kg in 1993 as documented by Cabidoche et al. (2009, Table 2). More than one-third of the CLD concentrations (i.e., 35 of 96) reported in Andosol soils for the 2017-2020 period by Comte et al. 2022 (Fig S3) indicate concentrations between 1 mg/kg and 10 mg/kg. This corresponds to decreasing rates lower by a factor of 3 to 10 compared to the 12 Andosol plots (Table 2) used by Comte et al. to calibrate the WISORCH_V1 model. Hence, these 12 plots may not be representative enough of the overall CLD fate in FWI Andosol soils.

Moreover, the following sentence raised our attention: "there were no significant effect of the length of the period between the first and second campaign on the decrease". While the authors observed that the CLD decrease cannot be explained by the time elapsed between the sampling campaigns, this data set was used to establish the temporal predictive model WISORCH_V1. The absence of relationships between the time elapsed between the two campaigns elapsed time and the decrease of CLD is illustrated in Figure 1.

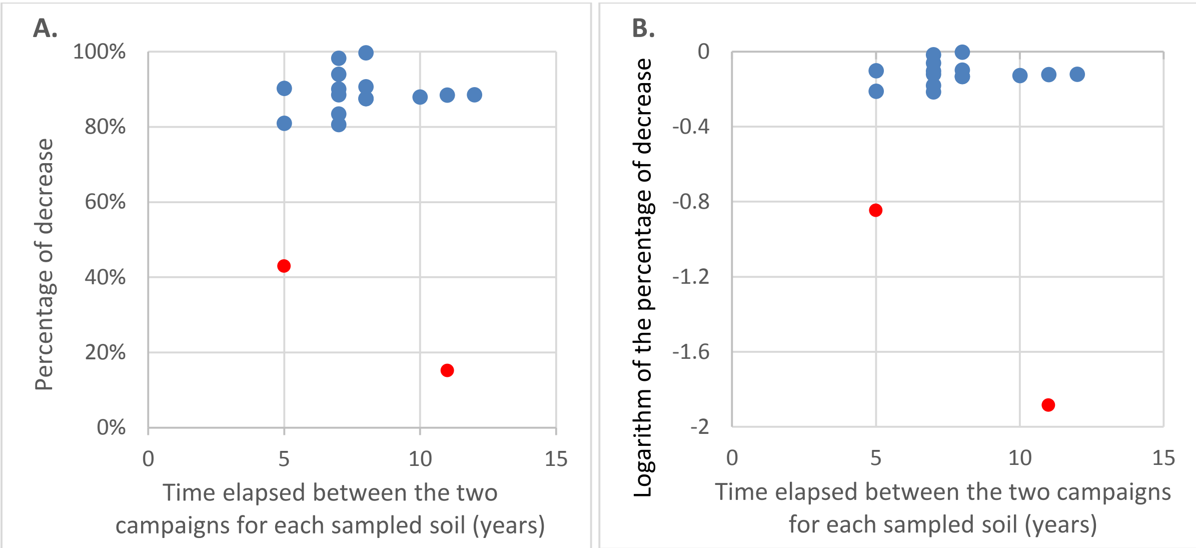


Figure 1: A. Evolution of the CLD decrease as a function of the time elapsed between the two sampling campaigns (n=17); B. Evolution of the logarithm of the CLD decrease as a function of the time elapsed between the two sampling campaigns (n=17); the apparent 'outliers' are marked in red.

Using the Spearman test (most suitable for data not following the Normal distribution) no significant relationship is observed between these series of values. Removing the apparent 'outliers' (2 Andosol plots among the 12 Andosol plots used to set the WISORCH_V1 model), it is possible to use the Bravais-Pearson test, which requires data that follow the Normal distribution. Under these conditions, as with the Spearman test, no significant correlation is observed. An experimental bias between the two campaigns, e.g. the change in the analytical protocol, can explain both the global decrease of CLD concentrations between the two sampling campaigns and the absence of relationship between the CLD decrease and the time elapsed between the two campaigns. Overall, we believe that the robustness and accuracy of the data set is not sufficient to optimize a parametric predictive model.

2. About the construction and results of the WISORCH_V1 model:

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

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

- Optimization of the WISORCH_V1 model: Comte et al. explained how they determined the DT50 of 5 years: "By running the resulting 33,600 simulations, we identified the 738 best simulations (…). In this group, 99% of simulations present a DT50 of 5 years". In other words, Comte et al. used the model WISORCH_V1 with a series of 12 couples of concentration values (12 plots analyzed at two distinct periods of time that include the two presumed 'outliers' presented in Figure 1a-b) for each of the seven parameters to be set (Comte, 2022; Figure S7), selecting the best fits. However, the WISORCH_V1 model itself was not designed to optimize the best DT50 for a given set of experimental data and no uncertainty was determined. Contrary to what has been done in Cabidoche et al., the sensitivity of the parameters (e.g. the DT50 value in WISORCH_V1) on the model predictions has not been studied. Eventually, it is worth noting that the criteria used for identifying the best fit of the WISORCH models were the slope and the r^2 of the regression between the simulated and measured soil CLD concentrations during the period between 2005 and 2017. Indeed these two criteria are useful to characterize model performance, although they do not inform on model bias. The mean error and the root mean square error would be more appropriate to evaluate prediction bias and variance. Hence, we question the robustness of the optimal model parameter values based on the calibration criteria chosen.

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

$C_{y+1} = C_y * e^{-\frac{ln(2)}{DT_{50}}} + T_y$ where y stands for the year; T_y is the CLD treatment for year y with T_y = 3 kg/ha 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.

The applied CLD amounts were converted in mg/kg of soil based on the assumption of a soil bulk density of 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. Figure 2 shows the predicted variation of CLD concentration in soil with time. When confronting these results to the measured soil concentrations given in Cabidoche et al. (2009) (Table 2) and in Comte et al. (2022) (Table 2) for Andosols and Nitisols of Guadeloupe, the vast majority of observations before 2017 (15 of the 17 values available for the 2005-2012 period and 15 of the 18 values for the 2001-2005 period) had significantly greater concentrations than the predicted values although the model was already meant to largely overestimate the actual values. Only 6 of the 17 data observed in 2017 seem consistent with the predictions. This thus questions the dissipation rate calculated by Comte et al. (2022), computed from samples taken from identical plots between 2005 and 2017. At least, it appears that the estimated dissipation rate should not be extrapolated to other periods since it is not suitable for retrospectively simulating soil CLD content changes since the start of CLD application.

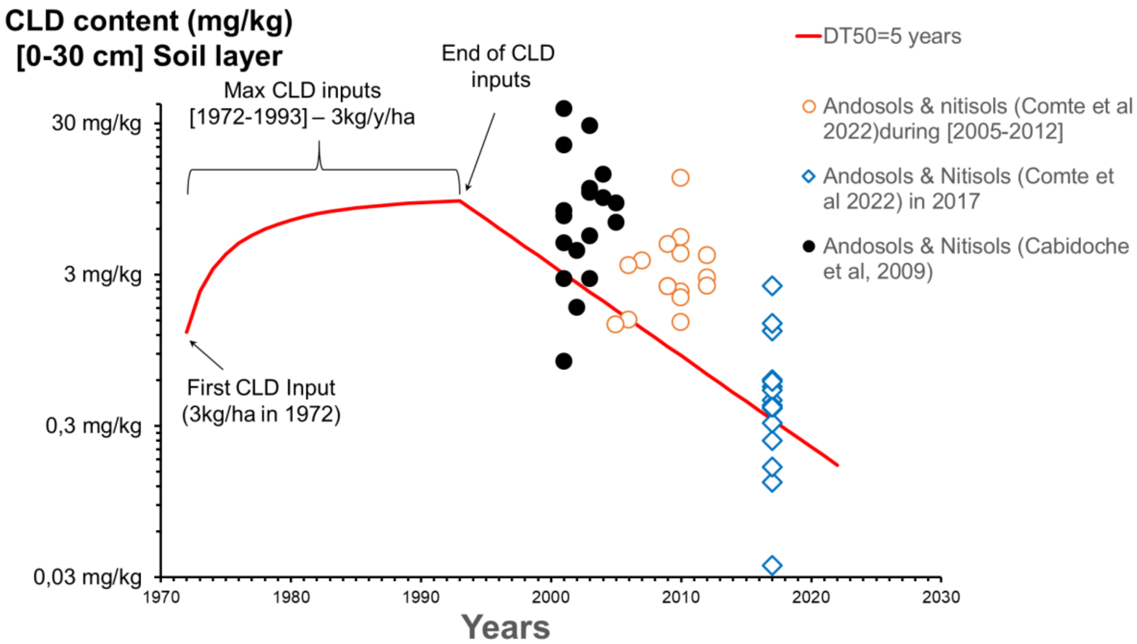


Figure 2: Evolution of the predicted CLD maximum concentration (red curve) in the upper layer soil for a DT50 of 5 years compared to Andosols and Nitisols concentrations in mentioned papers. CLD inputs are maximized by assuming a treatment every year between 1972 and 1993. A logarithmic scale was used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

Although the WISORCH_V1 could not be optimized for the Nitisol nor the Ferralsol due to the lack of data, Comte et al. concluded for these two soils to the same DT50 as for Andosol, and used this value to run the WISORCH_V1 model on the whole data set from 2001 to 2020 to predict the fate of CLD over the next several decades. Comte et al. argued that the comparison of the trends observed for the 2001-2020 between the three types of soils did not significantly differ (Comte, 2022, Fig S3). However, considering the above-mentioned biases about the public database, and the well-documented stronger sorption of CLD in Andosol than in other soil types (Comte, 2022; Cabidoche, 2009; Fernandez-Bayo, 2013), using the same DT50 of 5 years to all soil types may be incorrect. Predictions of CLD fate in Nitisol and Ferrasol soils established by Comte et al. should thus be viewed with even more caution than that obtained for Andosol soils.

Conclusion

The DT50 of 5 years for CLD and other outcome data of WISORCH_V1 clearly diverge from all previous studies and observations made on the fate and degradation of CLD in contaminated soils. Here, we highlighted a number of potential biases in the data and analyses. Although Comte et al. provided an insightful contribution to evaluate CLD dissipation in the environment, we conclude that the data set, the model and their conclusions raise several serious issues. As a result, the estimated dissipation half-life time (DT50) value determined by the authors contradict the observed fate of CLD in soils from the application period 1972-1993 until nowadays. We showed that the conclusion by Comte et al. of a fast dissipation of CLD in soils is not sufficiently supported by field data and model estimates. Hence, the paradigm of long-term persistence of CLD in FWI soil should remain the most likely hypothesis. The issues raised here may help to define in the future the criteria for more robust and reliable predictions of CLD dissipation in soil, starting from the field experimental design and sampling protocols. A reliable experimental design should thus include:

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

- Repetitive samplings should be carried out within the same period of the year and at a higher frequency (i.e. every year or every two years) to generate a statistically relevant dataset for each plot;

- Statistical treatments of these databases would also highlight possible outliers, reflecting irregular temporal or spatial dissipation processes;

- Long time storage of numerous duplicate control soil samples should allow to: (i) validate previous CLD contents and discard the possible erroneous values by re-analyzing; (ii) validate new quantification methods; (iii) apply new analytical methods (e.g. to search for transformation products (Chevallier et al., 2019) and/or to measure stable isotope signatures of CLD to trace *in situ* degradation (Höhener et al., 2022)). This will however require to define the optimal storage conditions to avoid the degradation of CLD and its transformation products in the stored samples.

We also recommend to include for any new predictive model:

- A validation using a retrospective simulation accounting for historical CLD applications;

- A sensitivity study to provide reliable uncertainties associated with any predictive outcomes.

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