Chlordecone in basal trunk wood of native trees growing in abandoned banana plantations in Guadeloupe, France

Éric, André Nicolini[1]
Jacques Beauchêne[2]
Vincent Bonnal[3]
Tom Hattermann[1]

[1] AMAP
Univ Montpellier
CIRAD, CNRS, INRAE, IRD
Montpellier
France

[2] CIRAD
UMR Écologie des forêts de Guyane (EcoFoG)
AgroParisTech, CNRS, INRA, Université des Antilles, Université de Guyane
97310 Kourou
France

[3] CIRAD
UMR TETIS
Centro de Desenvolvimento Sustentave - Universidade de Brasília - Campus Universitário Darcy Ribeiro
Brasília CEP 70910-900
Brésil

Auteur correspondant / Corresponding author:
Éric-André NICOLINI - eric-andre.nicolini@cirad.fr

Photo 1.
Appearance of an adventitious forest grove in an abandoned former banana plantation in Guadeloupe. This polluted site with Chlordecone was listed as having hosted a pure banana plantation in 1980, and was still a banana plantation in 2003. The two trees in the foreground (*Cecropia schreberiana* Miq. on the left, *Cordia sulcata* DC. on the right) are at most 19 years old and have also their wood consistently polluted with Chlordecone.
Photo E.,A. Nicolini.

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RÉSUMÉ

Chlordécone à la base du tronc d'arbres indigènes dans les bananeraies abandonnées en Guadeloupe, France

Le Chlordécone (CLD), utilisé pour lutter contre le charançon du bananier *Cosmopolites sordidus* et libéré par des bananeraies polluées, continue de contaminer les écosystèmes des Antilles françaises. Les plantes comestibles ont été activement étudiées pour la prévention des risques, mais pas les arbres, alors même qu'ils pourraient jouer un rôle important dans les stratégies de dépollution. Les teneurs en CLD ont été analysées sur 24 arbres pionniers appartenant à 13 essences présentes dans des bananeraies abandonnées en Guadeloupe, sur trois sites contaminés sur Nitisols (site 1) et Andosols (sites 2 et 3). Des échantillons ont été prélevés sur chaque arbre : du bois dans la partie basale du tronc et du sol à son pied. Leur teneur en CLD a été mesurée par le laboratoire d'analyses départemental de la Drôme (26) à Valence, France. Les teneurs moyennes en CLD dans la couche supérieure de 30 centimètres du sol des sites 1, 2 et 3 étaient respectivement de 2 543 ± 702, 5 251 ± 1 102 et 875 ± 865 µg/kg de sol sec. Parmi les arbres, 96 % étaient contaminés. La teneur en CLD des arbres poussant sur Nitisols (3 406 ± 1 658 µg/kg de bois sec) était au moins cinq fois plus élevée que celle des arbres sur andosols (299 ± 314 et 226 ± 378 µg/kg), mais aucune relation nette n'a été établie avec la teneur en CLD du sol. Le calcul du CLD disponible dissous en phase liquide dans le sol, à l'aide d'équations et de séries de données pédologiques de la littérature, a montré des teneurs en CLD disponible plus élevées dans les Nitisols que dans les andosols et une relation linéaire entre le CLD disponible dans le sol et les concentrations de CLD dans le bois, quel que soit le type de sol. Les arbres poussant sur Nitisols sont les organismes végétaux les plus fortement contaminés par le CLD parmi tous ceux dans lesquels ce composé a été étudié jusqu'à présent. Avec un rapport de bioconcentration plante-sol d'environ 150 l/kg, l'efficacité soutenue d'absorption de CLD par les arbres doit être prise en compte dans les recherches futures sur la dépollution des milieux contaminés par le chlordécone.

Mots-clés : bois, Chlordécone, essences forestières, indigène, Guadeloupe, France.

ABSTRACT

Chlordecone in basal trunk wood of native trees growing in abandoned banana plantations in Guadeloupe, France

Chlordecone (CLD), used to control the *Cosmopolites sordidus* banana weevil and released from polluted banana plantations, continues to contaminate ecosystems in the French Caribbean. Edible plants have been actively studied for risk prevention, but trees have not, even though they could play a significant role in future remediation strategies. CLD contents were analysed in 24 pioneer trees belonging to 13 species found in abandoned banana plantations in Guadeloupe, at three contaminated sites on Nitisols (Site 1) and Andosols (Sites 2 and 3). Samples were taken from each tree: wood in the basal part of the trunk and soil at its foot and their CLD content was measured by the analytical laboratory for the Drôme *département* (26) in Valence, France. Mean CLD contents in the top 30-centimetre soil layer from sites 1, 2 and 3 were 2,543 ± 702, 5,251 ± 1,102 and 875 ± 865 µg/kg dry soil respectively. Of the trees, 96% were contaminated. The CLD content in trees growing on Nitisols (3,406 ± 1,658 µg/kg dry wood) was at least 5 times higher than in trees growing on Andosols (299 ± 314 and 226 ± 378 µg/kg), but no clear relationships were found with soil CLD contents. Calculations of available CLD dissolved in the soil liquid phase using equations and soil datasets in the literature showed higher available CLD contents in Nitisols than in Andosols and a linear relationship between CLD available in soil and concentrations of CLD in wood, regardless of the type of soil. Trees growing on Nitisols are the plants most highly contaminated by CLD of all the plants in which this compound has been studied so far. With a plant-to-soil bioconcentration ratio around 150 l/kg, the consistent CLD uptake efficiency of the trees needs to be taken into account in further research for CLD remediation.

Keywords: Wood, Chlordecone, forest trees, native, Guadeloupe, France.

RESUMEN

Clordecona en la madera del tronco basal de árboles autóctonos que crecen en plantaciones de plátanos abandonadas en Guadalupe, en Francia

La clordecona (CLD), utilizada para controlar el gorgojo del plátano *Cosmopolites sordidus* y liberada por las plantaciones de plátanos contaminadas, sigue contaminando los ecosistemas del Caribe francés. Las plantas comestibles se han estudiado activamente para la prevención de riesgos, pero los árboles no, a pesar de que podrían desempeñar un papel importante en las futuras estrategias de descontaminación. Se analizó el contenido de CLD en 24 árboles pioneros pertenecientes a 13 especies encontradas en plantaciones de plátanos abandonadas en Guadalupe, en tres lugares contaminados en Nitisoles (sitio 1) y Andosoles (sitios 2 y 3). Se tomaron muestras de madera de cada árbol en la parte basal del tronco y en el suelo a su pie y su contenido en CLD fue medido por el laboratorio de análisis del departamento de Drôme (26) en Valence, Francia. El contenido medio de CLD en la capa superior de 30 centímetros del suelo de los sitios 1, 2 y 3 fue de 2 543 ± 702, 5 251 ± 1 102 y 875 ± 865 µg/kg de suelo seco respectivamente. El 96 % de los árboles estaban contaminados. El contenido de CLD en los árboles que crecían en Nitisoles (3 406 ± 1 658 µg/kg de madera seca) era al menos 5 veces mayor que en los árboles que crecían en Andosoles (299 ± 314 y 226 ± 378 µg/kg), pero no se encontraron relaciones claras con los contenidos de CLD en el suelo. Los cálculos de la CLD disponible disuelta en la fase líquida del suelo utilizando ecuaciones y conjuntos de datos de suelos de la literatura mostraron mayores contenidos de CLD disponible en los Nitisoles que en los Andosoles y una relación lineal entre la CLD disponible en el suelo y las concentraciones de CLD en la madera, independientemente del tipo de suelo. Los árboles que crecen en Nitisoles son las plantas más contaminadas por CLD de todas las plantas en las que se ha estudiado este compuesto hasta ahora. Con un ratio de bioconcentración planta-suelo de alrededor de 150 l/kg, la eficiencia de absorción de CLD consistente de los árboles debe ser tenida en cuenta en futuras investigaciones para la descontaminación de CLD.

Palabras clave: madera, clordecona, árboles forestales, autóctono, Guadalupe, France.

Introduction

Chlordecone (CLD; $C_{10}Cl_{10}O$; CAS 143-50-0) is an organochlorine previously used to control the black weevil (*Cosmopolites sordidus* Germar) in banana plantations. Classified as a persistent organic pollutant (UNEP, 2007), it was banned worldwide in 1992. With a strong affinity for organic matter in soil (Cabidoche *et al.*, 2009; Cattan *et al.*, 2019), CLD is very stable and not very mobile in soils and contaminated plants. However, CLD is still being released from polluted fields, and is still contaminating aquatic ecosystems, groundwaters and rivers in different parts of the world including in the French West Indies (Cattan *et al.*, 2019) and affects human health.

Different alternatives have been considered to solve CLD pollution. Soil clean-up using different processes (biodegradation by fungi or bacteria, chemical processes) has been considered but no effective clean-up technique has yet been identified due to very low CLD degradation rates (Merlin, 2015) and the need for anaerobic conditions (Mouvet *et al.*, 2017), which are adverse conditions in agricultural environments. Phytoremediation has also been considered, but unfortunately, to date, no plants with these characteristics have been identified for CLD, and phytoremediation by crop plants is limited by the very low soil-to-plant CLD transfer rates (Mouvet *et al.*, 2020).

Inversely, strengthening the ability of soils to retain CLD has also been explored to prevent it contaminating (1) agricultural products, and (2) other sites. The addition of organic matter or activated carbons (ACs) to the soil has been shown to significantly reduce the bioavailability of CLD and its transfer to crops (see respectively, Clostre *et al.*, 2014a; Ranguin *et al.*, 2020; see also Delannoy *et al.*, 2018). Moreover, adding compost to the soil has the advantage of being easy to implement and simultaneously improves the agronomic quality of the soils (Clostre *et al.*, 2014b). However, its effectiveness is limited to between 6 months and one year and inputs of compost must be renewed periodically.

Reducing tillage and herbicides can also be part of this approach since deep tillage and widespread use of Glyphosate are suspected of having allowed CLD to spread into the environment (see respectively, Cabidoche *et al.*, 2004; Sabatier *et al.*, 2021). What is more, tillage is known to increase the mineralization rate of organic matter stored in the soil (Balesdent *et al.*, 2006; Saptoka *et al.*, 2012) and consequently, the release of the stable CLD stored in the deeper soils layers of polluted fields and groundwater. While clean-up pathways have not yet provided the expected solutions, strengthening the soil's capacity to retain CLD seems to be the most realistic.

In this particular context of limiting pollution inside the plot, trees do have a role to play. Trees are guarantors of the integrity of soils through their root systems, and are major purveyors of organic matter. In addition, they could also capture significant quantities of CLD in the medium and long term. To our knowledge, with the exception of tree fruits (citrus, mango; Cabidoche *et al.*, 2006), no studies have yet been conducted on the concentration of CLD in the different tree compartments. While root systems and hemicellulose are the preferred storage structures of CLD in many herbaceous and monocotyledonous species (Clostre *et al.*, 2014b; Clostre *et al.*, 2015), trees, large organisms mainly made up of wood and therefore lignin, most probably capture substantial amounts of CLD. This hypothesis is plausible, even if it is now accepted that the fruits of polluted trees and of many herbaceous plants, mainly filled by phloem streams (elaborate sap), are not affected by pollution and can still be eaten (i.e., *Citrus*; Cabidoche *et al.*, 2006).

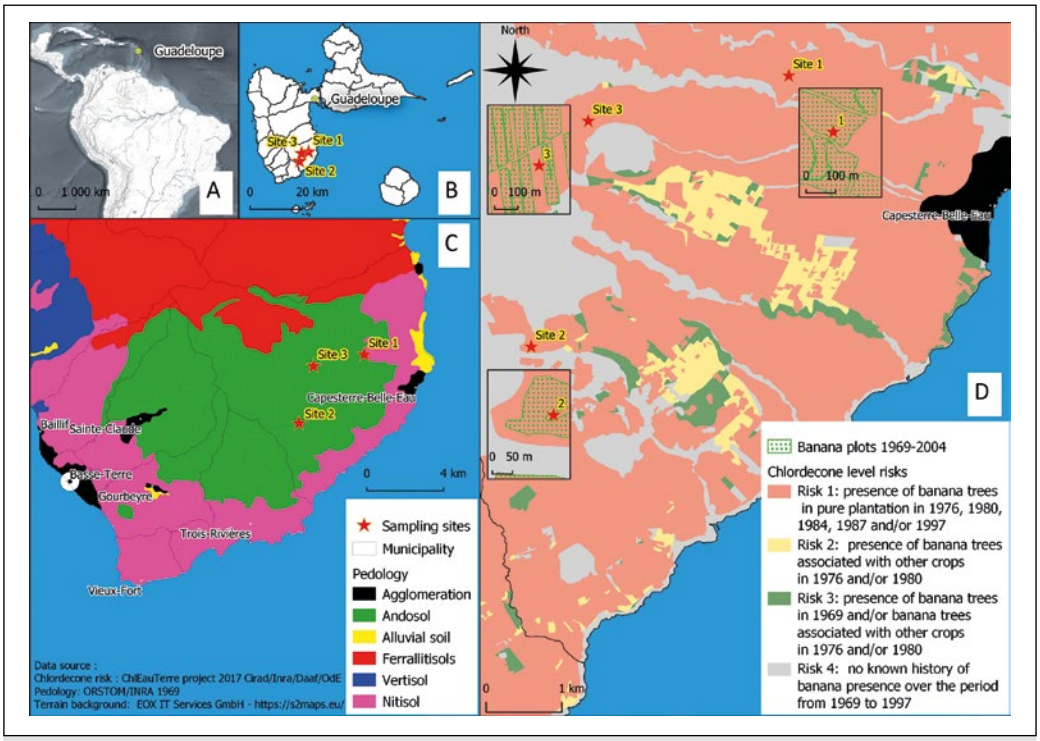

Figure 1.
Location of the three study sites in Caribbean islands (A), Guadeloupe (B), in the south part of Basse-Terre (C). In the left panel, the different soil types encountered in the south part of Basse-Terre. In the right panel (D), a detailed view of the study area showing the different levels of risk relative to plot history: "Risk 1" is the highest level of risk. Each site (1, 2 and 3) is indicated by a red star. The three panels (scale 0-100m) in the right panel (D) show that Sites 1 and 2 are both old pure banana plots but Site 3 is not. However, this site was listed in 1980 as having hosted a banana plantation mixed with a vegetable garden.

The future of CLD in soils is inseparable from that of organic matter. To contain pollution within contaminated areas, and before incorporating any external organic matter at industrial scale, it is thus necessary to quantify the amount of living and dead organic matter originating from different growing systems and to establish their intrinsic abilities to trap CLD. We also need to know the levels of CLD in the organic matter produced by these systems before establishing complete balance sheets. Agroforestry systems are known for their ability to produce organic matter (Guenet *et al.*, 2020), and trees structure these growing systems. Consequently, we need to know more about CLD content in trees growing in contaminated areas.

The aim of this preliminary study was to check and quantify the CLD contents in the basal trunk wood of several native tree species growing spontaneously in contaminated soils in abandoned banana plantations on the island of Guadeloupe. We aimed to test two hypotheses: native forest trees are significantly contaminated as they grow in all highly contaminated soils (hypothesis 1) whatever the type of soils (hypothesis 2). The CLD accumulative property of the different trees we studied is then discussed.

Materials and methods

Our study was conducted in the south part of Basse-Terre, near the city of Capesterre-Belle-Eau (FR-97130), on the island of Guadeloupe (figure 1).

A list of old, abandoned banana plantations was extracted from the ChlEauTerre spatialized data base (Rochette *et al.*, 2017). Using different terms (surface area, vegetation, CLD concentration in soil), we established a list of plots (i) whose soils are significantly contaminated by CLD (level 1: area at maximum risk of contamination) and (ii) area currently abandoned and invaded by native secondary forests. The type of vegetation was checked using aerial photographs. We located three sites using their geographical X and Y coordinates (WGS 84 / UTM zone 20N), each site contained one or several plots.

- Site 1 (plots 31-32); Nitisols; (x = 650,892 m, y = 1,775,921 m), altitude 170 m.
- Site 2 (plots 59-60-61); Andosols: (x = 647,519 m, y = 1,772,203 m), altitude 296 m.
- Site 3 (plot 44); Andosols: (x = 648,251 m, y = 1,775,285 m), altitude 308 m.

Between 1976 and 1980, Site 1 was listed as having hosted a pure banana plantation and was still a banana plantation in 2003. Site 2 was also listed as having hosted a pure banana plantation between 1976 and 1980, but in 2004, it was already abandoned and was still abandoned in 2019. Site 3 is located between two banana plots, but was not listed as having hosted pure banana plantations (figure 1), in 1980, was listed as having hosted a mixed banana plantation and vegetable garden. The location of this abandoned site in a maximum risk zone and the presence of forest cover today made us decide to include it.

At each site, we (i) first selected several trees and (ii) collected a botanical sample from each tree, which was subsequently examined in the Duclos herbarium (INRAE research station) for identification to genus or species level. (iii) Third, we collected around 200 g of fresh wood at the base of the trunk of each selected tree using an electric auger. Finally, (iv) , we collected samples of soil around each selected tree using a hand auger.

Twenty-four trees (8 trees per site) were sampled, belonging to 11 botanic families and 13 species (table I).

Spatial distribution of the CLD in the soil of a plot is extremely uneven for different reasons (Cabidoche *et al.*, 2006): (1) the spatial variability of the organic matter in the soil which determines the retention of CLD, (2) the mode of application of the product, which was only applied at the foot of each banana tree, where the CLD was subsequently found to be concentrated, (3) tillage which could have redistributed the CLD to different depths depending on the tillage practice used. In our sampling campaign, we took four soil samples per tree. We then pooled these four soil samples to form one composite soil sample per tree. The samples were collected between one and two metres from the trunk in the 0-30 cm soil layer to be in line with the existing references. Composite soil samples and wood samples were placed in individual plastic bags, and sent to the Departmental Analytical Laboratory 26 (Valence, France) for analysis of CLD contents.

Table I.
Families and species sampled at the 3 sites.

Family	Species	Abbreviation	Number	Site
Cecropiaceae	*Cecropia schreberiana* Miq.	Cecr	4	1, 3
Cordiaceae	*Cordia cf. sulcata* DC.	Cord	3	1, 3
Euphorbiaceae	Sapium caribaeum Urb.	Sap	1	3
Lauraceae	*Ocotea cf. krugii (Mez)* R.A.	Ocot	1	1
Melastomataceae	sp1	Mela1	1	2
Melastomataceae	sp2	Mela2	2	2
Meliaceae	*Swietenia mahagony* (L.) Jacq.	Swie	2	2
Mimosaceae	*Inga ingoïdes* (Rich.) Willd.	Inga	4	1, 2, 3
Moraceae	*Artocarpus altilis* (Parkinson) Fosberg	Arto	1	3
Rubiaceae	sp1	Rub	1	2
Rubiaceae	*Chimarrhis cymosa* Jacq.	Chim	1	2
Simaroubaceae	*Simarouba amara* Aubl.	Sima	2	1, 3
Sterculiaceae	*Sterculia caribea* R. Br.	Sterc	1	3

Extraction and measurement of CLD contents in soils (Rochette *et al.*, 2020)

All composite soil samples were analysed in the Departmental Analytical Laboratory (LDA26) in Valence, France, which works under the French Accreditation Committee (COFRAC), according to the NF EN ISO/CEI 17025 standard. For the analysis of CLD in soils, 10 g of the sample were placed in an extraction cell with regenerated hydromatrix and tracers were added (HBB/TPP; 100 µL). Accelerated solvent extraction (ASE) was carried out with a 50/50 dichloromethane/acetone mixture at 100 °C under 120 bar pressure. The resulting extract was concentrated in a vacuum centrifuge (GENEVAC EZ2) which greatly reduces the loss of volatile compounds. The extract was concentrated to 10 ml and a 1 mL aliquot was removed for analysis. A drop of pentanol was added to the extract, and the solvent was evaporated in a GENEVAC miVac system to preserve the volatile compounds. The extract was then taken up by a mixture of acetonitrile and water with the Chlordecone 13C internal standard. Analysis was performed by HPLC-MS/MS, with an analytical uncertainty of 40%, a detection threshold of 2 µg/kg of dry soil, and a quantification threshold of 5 µg/kg of dry soil.

Extraction and measurement of CLD contents in wood

After the wood samples were ground, they were all also analysed in the LDA26 laboratory in Valence. The method used was the method for the determination of CLD in food products of plant origin (ANSES PBM Pest LSA-INS-0161; version 02; 14th of September 2015). CLD was extracted from the matrix with strong wash solvent H2O/acetonitrile & 0.1% formic acid including tracers Atrazine D5 and 24D–D3. After filtration, extraction was performed by liquid/liquid partitioning in the presence of sodium chloride, water, and dichloromethane. The resulting extract was purified in a silica cartridge and analysis was performed by high performance liquid chromatography-tandem mass spectrometry (HPLC-MS/MS). It should be noted that (1) Atrazine D5 and 24D–D3 present in the sample recovery solvent but were not used for analysis, and (2) 150 µl Chlordecone ^{13}C was used as the external standard.

Table II.
Measured and calculated variables. DM: dry matter; CLD: Chlordecone.

Measured	[CLD]wood	Total CLD content in wood dry matter	µg/kg DM
	[CLD]soil	Total CLD content in soil dry matter	µg/kg DM
	Tsoil	Depth of soil considered	m
Calculated	BTR	Bulk transfer ratio	µg/µg or unit less
	[CLD]stock	Volumetric soil CLD content	µg/l
	[CLD]available	CLD dissolved in soil liquid phase	µg/l
	WCR	Wood CLD bioconcentration ratio	µg/µg or unit less

We studied several measured and calculated variables (table II). The variables measured were the gravimetric CLD contents in the dry matter (DM) of tree wood ($[CLD]_{wood}$) and soil ($[CLD]_{soil}$). We then calculated an initial soil-plant bioconcentration ratio (McKone and Maddalena, 2009) also called bulk transfer ratio (BTR; Cabidoche and Lesueur-Jannoyer, 2012):

$$BTR = [CLD]_{wood} / [CLD]_{soil}$$

Organic matter *plus* CLD trapping by different clays strongly affect the bioavailability of CLD. Hence $[CLD]_{soil}$ does not provide any information on the volumetric content of CLD in the soil ($[CLD]_{stock}$) nor on bioavailable CLD content ($[CLD]_{available}$), i.e. the actual CLD dissolved in the soil liquid phase, which is a determining factor (Cabidoche and Lesueur-Jannoyer, 2012). As we were unable to measure the soil physical properties (BD, W_{fc}, K_{oc} and SOC) needed to calculate these two variables at our three study sites, we used values (table III) in the literature (Levillain *et al.*, 2012; Cabidoche *et al.*, 2009) measured in many previously observed situations in both Andosols and Nitisols in the same area study.

Table III.
Physicochemical characteristics and retention capacities of the main soil types in the study area. Koc: Partitioning coefficient between the Chlordecone fraction absorbed by the soil organic matter (estimated by soil organic carbon) and the Chlordecone fraction dissolved in water.

	Parameter	Unit of measurement	Andosols	Nitisols	References
Wfc	Gravimetric water content in soil at field capacity in kg of water per kg of soil dry matter	(kg/kg DM)	0.8	0.35	Colmet-Daage (1969)
BD	Bulk density	(kg/dm³ DM) or (kg/l DM)	0.55 0.6	1.1 0.9	Levillain *et al.* (2012) Cabidoche *et al.* (2009)
Koc	Soil/water partition coefficient relative to the organic carbon content	(dm³/kg) or (l/kg)	20,000 17,900	2,000 2,500	Levillain *et al.* (2012) Cabidoche *et al.* (2009)
SOC (150 m)	Soil carbon content in kg per kg of dry soil according to elevation (m)	(kg/kg)	/	0.02	Dorel *et al.* (2005) Levillain *et al.* (2012)
SOC (300 m)			0.09 0.067	/	Cabidoche *et al.* (2009)

For each composite soil sample, we calculated $[CLD]_{stock}$ and $[CLD]_{available}$ using the equations reported by, respectively, Levillain *et al.* (2012) and Cabidoche and Lesueur-Jannoyer (2012), (the parameters measured are listed in table II, and soil property values are listed in table III.

$[CLD]_{stock} = T_{soil} \times 10 \times [CLD]_{soil} \times BD$, where T_{soil} is the depth of soil considered = 0.3 m.

$[CLD]_{available} = [CLD]_{soil} \times W_{fc} \times BD / K_{oc} / SOC$

However, these 2 datasets (table III) have noticeably different values, especially for SOC or K_{oc} on Andosols. We kept them all to have a wider range of soil conditions that could be encountered in the area. After calculating $[CLD]_{available}$ values from each of these datasets, we also calculated the $[CLD]_{available}$ values using all possible combinations using data from the two sets (table III). In this way, we found four possible combinations on Nitisols and eight possible combinations on Andosols. Table IV lists mean $[CLD]_{available}$ in soil of each tree and for each site.

Finally, we calculated a second soil-plant bioconcentration ratio also cited in the critical review of McKone and Maddalena (2009). This is the ratio of the concentration of CLD in fresh plant tissue (µg/l) to the concentration of CLD in the soil solution (µg/l). However, like Cabidoche and Lesueur-Jannoyer (2012), we considered dry matter rather than fresh matter to calculate the wood bioconcentration Ratio (WCR).

$WCR = [CLD]_{wood} / [CLD]_{available}$

Table IV.
Soil and wood Chlordecone contents and uptake ratios. Ind.: individual; Spec.: species abbreviation (see table II); CLD: Chlordecone; BTR: Bulk transfer ratio; Nit: Nitisols; And: Andosols; for the name of the species (Spec.), see table I; SE: Standard error; SD: Standard deviation; *: mean, median and coefficient of variation calculated with exclusion of the highest WCR values in bold in the column WCR "Individual"; Coef. Var.: Coefficient of variation.

Soils Sites	Ind.	Spec.	$[CLD]_{Wood}$	$[CLD]_{Soil}$	BTR	$[CLD]_{Avail.}$	WCR	
						Individual mean ± SD	Individual	Mean ± SE **Median** Coef. Var.
			(µg/kg dry matter DM)		(µg/µg) or unit less	(µg/dm³) or (µg/l)	(µg/kg/µg/l) or (µg/µg; McKone et Maddalena, 2009) or unit less	
Nit 1	1	Cecr	3,595	2,968	1.21	23.4 ± 4	153	
	2	Cecr	1,422	1,803	0.79	14.2 ± 2.4	100	
	3	Ocot	2,406	3,148	0.76	24.8 ± 4.3	97	176 ± 31
	4	Inga	5,265	2,183	2.41	17.2 ± 3	306	**161**
	5	Sima	2,106	3,283	0.64	25.9 ± 4.5	81	49
	6	Inga	5,243	3,195	1.64	25.2 ± 4.3	208	
	7	Cord	5,290	2,320	2.28	18.3 ± 3.2	290	
	8	Cord	1,921	1,446	1.33	11.4 ± 2	169	
And 2	9	Mela1	289	6,519	0.04	2.1 ± 0.4	140	
	10	Mela2	112	4,120	0.03	1.3 ± 0.2	86	236 ± 112
	11	Rub	452	4,120	0.11	1.3 ± 0.2	346	**113**
	12	Inga	610	6,052	0.10	1.9 ± 0.3	318	134
	13	Swie	3	3,886	0.00	1.9 ± 0.3	2.14	
	14	Swie	2	3,886	0.00	1.2 ± 0.2	1.6	*134 ± 54
	15	Chim	855	6,005	0.14	1.2 ± 0.2	**950**	***86**
	16	Mela2	72	4,924	0.01	1.6 ± 0.3	46	*107
And 3	17	Cecr	138	2,398	0.06	0.86 ± 0.15	215	
	18	Arto	55	2,398	0.02	0.86 ± 0.15	86	2,219 ± 1,721
	19	Sterc	91	247	0.37	0.08 ± 0.01	**1 155**	**190**
	20	Inga	1,114	247	4.51	0.08 ± 0.01	**14 146**	219
	21	Sima	12	616	0.02	0.2 ± 0.04	62	
	22	Cord	375	616	0.61	0.2 ± 0.04	**1 923**	*106 ± 38
	23	Sap	0	642	0.00	0.2 ± 0.04	0	***86**
	24	Cecr	25	474	0.05	0.15 ± 0.02	165	*81

Results

All the plots were contaminated with CLD (figure 2A). The concentrations of CLD in the soil ($[CLD]_{soil}$) measured at the three sites differed significantly (Kruskal-Wallis test for equal medians; *p* (same): 0.0001). Mean $[CLD]_{soil}$ at Site 1 with Nitisols (2,543 ± 702 µg/kg dry matter DM) was between the mean at Site 2 (5,251 ± 1,102 µg/kg DM) and the mean at Site 3 (875 ± 865 µg/kg DM) both on Andosols. Note that Site 3 had one relatively high value (2,398 µg/kg DM) leading to the highest variation coefficient (99 *versus* 27 and 21 for Sites 1 and 2 respectively).

The basal trunk wood of 96% of the sampled trees was also contaminated by CLD (figure 3). The mean concentrations of CLD in wood ($[CLD]_{wood}$) at Site 1 (3,406 ± 1,658 µg/kg DM) were 10 -times higher than the means at Sites 2 and 3 (respectively 299 ± 314 and 226 ± 378 µg/kg DM) even though the soils at Site 1 were not the most heavily contaminated. Trees at Sites 2 and 3 had relatively low $[CLD]_{wood}$ means whatever the $[CLD]_{soil}$ at the respective sites. Again, at Site 3, it should be noted that one value (1,114 µg/kg DM) was an outlier resulting in a high coefficient of variation (167 *versus* 49 and 105 for Sites 1 and 2 respectively).

No relationship was found between $[CLD]_{wood}$ and $[CLD]_{soil}$. On the other hand, the $[CLD]_{wood}$ of five out of the eight trees sampled at Site 1 (Nitisols) was much higher than the $[CLD]_{soil}$ of the soil sampled around their base (table IV; BTR > 1; $mean_{BTR}$: 1.38 ± 0.7) whereas only one tree at Site 3 (Andosol) showed a BTR > 1 ($mean_{BTR}$: 0.7 ± 1.6). All trees sampled at Site 2 had a BTR < 1 ($mean_{BTR}$: 0.06 ± 0.08).

Like Cabidoche and Lesueur-Jannoyer (2012), we hypothesized that the $[CLD]_{wood}$ depended more on the soil CLD stock ($[CLD]_{stock}$) or on the actual CLD dissolved in soil liquid phase and therefore actually available ($[CLD]_{available}$).

The $[CLD]_{soil}$ reduced to the soil bulk density makes it possible to account for the real $[CLD]_{stock}$ in the soil volume. Thus, Site 2 whose $[CLD]_{soil}$ values were almost double those of Site 1, ultimately had a $[CLD]_{stock}$ similar to that of Site 1 (figure 2B) because its soil bulk density was half that of Site 1 (table III; Andosols 0.55 *versus* Nitisols 1.1). Site 3 again had the lowest $[CLD]_{stock}$. Despite this change, no relationship was found between soil $[CLD]_{stock}$ and $[CLD]_{wood}$.

Results for $[CLD]_{available}$ differed significantly. Given that we disposed of two sets of soil parameter values taken from the literature for the calculation of $[CLD]_{available}$, we were able to obtain three sets of possible results (figure 4, A, B and C). $[CLD]_{available}$ values in A and B were obtained from data sets taken from Cabidoche *et al.* (2009) and of Levillain *et al* (2012) respectively, whereas the values in C were obtained from the combination of the two previous data sets (see "Materials and methods"). Box plots in A and B (figure 4) show that means and medians differed significantly at site level. In C (figure 4), it will be recalled that the number of values is more than eight trees per site due to all the possible combinations of the two datasets (see "Materials and methods"): 4 per tree in Nitisols, and 8 per tree in Andosols. This combination provided the intermediate $[CLD]_{available}$ values we finally retained: Site 2 which

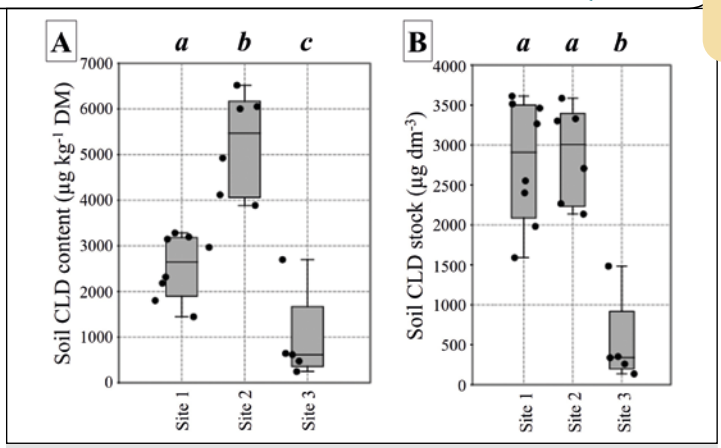

Figure 2.
Box plots of (A) Chlordecone content (µg/kg soil dry matter DM) and (B) Chlordecone stock (µg/dm) in the soils at the 3 sites. Box plots with the same letter do not have significantly different medians (Kruskal-Wallis test for equal medians; in A, *p* (same) = 0.0007; in B, *p* (same) = 0.005).

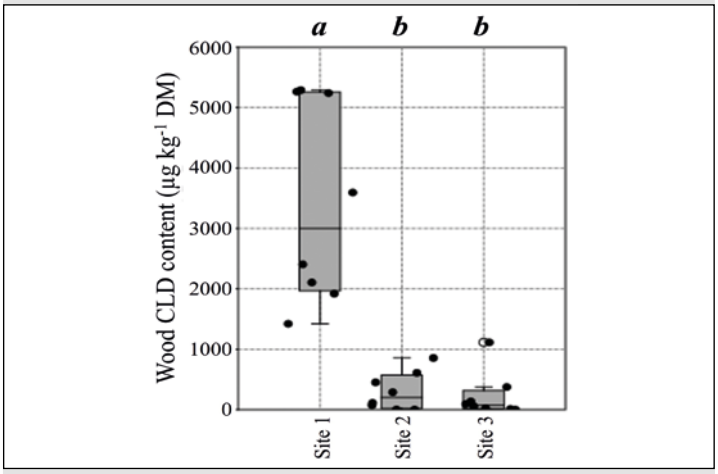

Figure 3.
Box plots of Chlordecone content (µg/kg wood dry matter DM) in the wood collected from the basal part of the trees sampled at the three sites. Box plots with the same letter do not have significantly different medians (Kruskal-Wallis test for equal medians; *p* (same) = 0.0004).

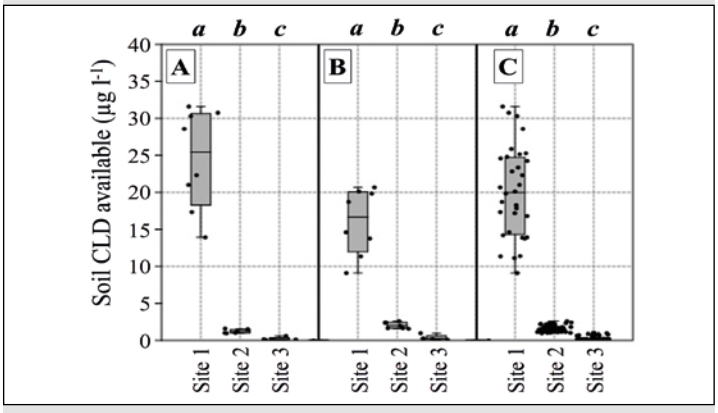

Figure 4.
Box plots of the volumetric content of Chlordecone (CLD) dissolved in the soil solution of the soil samples collected at the three study sites. To calculate the CLD dissolved and available in the soil solution ($[CLD]_{available}$), we first used soil property values taken from two different studies (see also table III): (A) from Levillain *et al.* (2012), and (B) Cabidoche *et al.* (2009). Box plots in C present the $[CLD]_{available}$ values using all the possible combinations of the 2 datasets: 4 per tree in Nitisols and 8 per tree in Andosols. Box plots with different letters have significantly different medians (Kruskal-Wallis test for equal medians; in A and B, *p* (same) = 0.0003; in C: *p* (same) = 0.000).

had the highest $[CLD]_{soil}$ presented a mean $[CLD]_{available}$ (1.66 ± 0.43 µg/l) that was significantly lower than that at Site 1 (20 ± 6.13 µg/l), and Site 3 presented the lowest mean $[CLD]_{available}$ (0.3 ± 0.29 µg/l).

As we had access to $[CLD]_{available}$ values, we were able to test the relationship between $[CLD]_{available}$ and $[CLD]_{wood}$, regardless of the soil type and the site. We fitted three positive linear regressions (figure 5), whose intercept proved to be zero for both soil types since we assumed no CLD in plants growing on soil that was never contaminated. The two opposite linear regressions with $[CLD]_{available}$ values calculated from the two sets of parameters (Levillain *et al.*, 2012; Cabidoche *et al.*, 2009) differed significantly (slope = 133, confidence intervals [76, 178]; slope = 203, confidence intervals [121, 269]). The third linear regression corresponding to the two datasets combined had a slope of 160 (confidence intervals [129, 188]). With no more soil indications, we preferred the last one, which represents an average plant-to-soil bioconcentration ratio (WCR) for the trees in our study.

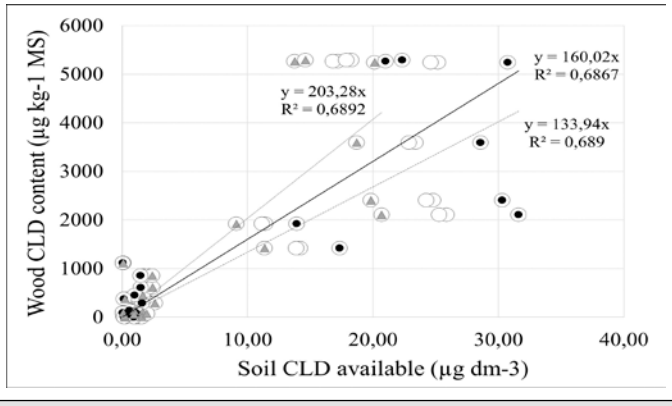

Figure 5.
Linear relationships between volumetric content of Chlordecone (CLD) dissolved in the soil solution ($[CLD]_{available}$) and Chlordecone content in the dry matter (DM) of wood ($[CLD]_{wood}$). In "dataset 1", "dataset 2" and "mixed", soil ($[CLD]_{available}$ values were calculated respectively from soil property values (see also table III) taken from Cabidoche *et al.* (2009), Levillain *et al.* (2012) and a mixed dataset containing data taken from the two studies.

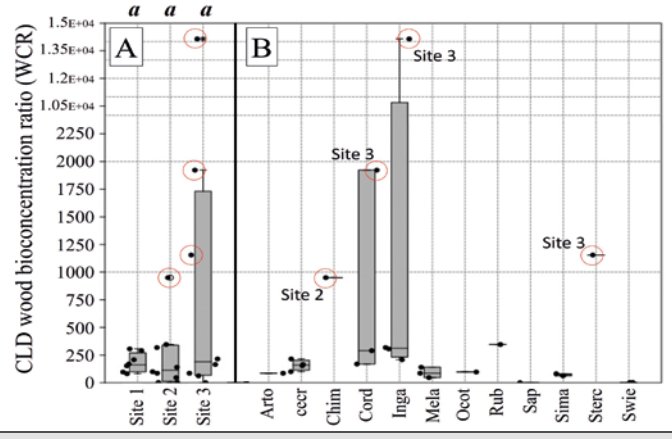

Figure 6.
Box plots of the WCR (Chlordecone wood bioconcentration ratio) for trees (A) in the three study sites and (B) for each collected genus or species. Box plots with the same letter have no significantly different medians (Kruskal-Wallis test for equal medians in A: *p* (same) = 0.778, ns). The red circles highlight extreme values mainly observed at Site 3 (three individual) and Site 2 (one individual). For the name of the species in B, see table I.

However, the mean WCR of each site was checked separately. We first calculated the mean WCR for each sampled tree from the individual value of $[CLD]_{wood}$ measured at the laboratory and the mean $[CLD]_{available}$ calculated from the datasets in the literature. With a mean value of 2,219 ± 1,721 (median: 190), it appears that the average WCR at Site 3 (table IV) did not fit with the slope of the linear model (figure 5, white circle, "mixed"; slope = 160). Moreover, WCR at Site 3 has a strong coefficient of variation, 219 (table IV) mainly due to an outlier (WCR = 14,146) *plus* two other WCR values of more than 1,000 (figure 6). In Site 2, variability was much lower (coefficient of variation: 134) but with one relatively high value of 950 ($mean_{WCR}$ = 236 ± 112; median: 113). Finally, Site 1 has rather homogenous distribution ($mean_{WCR}$ = 176 ± 31; median = 161) with a very low coefficient of variation, 49.

The WCR medians of the three sites (table IV) were not significantly different (Kruskal Wallis test for equal medians; *p* (same): 0.778). Grouping all WCR values except the four highest WCR values gave a mean WCR for trees of 154 ± 119 (median = 129; Coeff. Var. = 78).

Part B of figure 6 shows the WCR values for the different species. For *Inga* and *Cordia* trees, with the exception of the highest values at Site 3, the values at Sites 1 and 2 grouped respectively, around 300 and 250. For *Cecropia* trees, the values at Sites 1 and 3 grouped around 150. No other trees belonging to the other species reach 400.

Discussion

Presence of CLD in trees

In this preliminary study, we looked for trees belonging to the native flora of Guadeloupe able to grow on abandoned banana plots containing different types of soil (Nitisols or Andosols) that were significantly contaminated with CLD. We measured the CLD content in the wood collected at the base of the trunk of each sampled tree. Our results confirm our first hypothesis: the levels of CLD found in the trees at the three sites clearly demonstrate that native forest trees can be significantly contaminated by CLD. To our knowledge, contamination of trees by CLD has never previously been reported, with the exception of one study (Cabidoche *et al.*, 2006) but which only dealt with tree fruits belonging to several species in Andosols in Guadeloupe. That study reported that some fruits may be contaminated but at very low levels (e.g., no more 12 µg/kg for a grapefruit). Nevertheless, even though only a few trees were studied here, the fact that all the trees growing in Site 1 had $[CLD]_{wood}$ values of more than 1,000 µg/kg DM suggests it is not a marginal phenomenon in trees.

Wood CLD content: trees *versus* herbaceous plants

Trees growing at Site 1 (on Nitisols) had the highest CLD contents (3,406 ± 1,658 µg/kg DM; or 1,800 ± 1,000 µg/kg fresh matter FM), and often 10 times (1.8 to 22-times) higher than

the concentrations recorded in the aerial parts of several plants, very often monocotyledons or herbaceous taxa. We found no such high CLD contents reported in the literature. The CLD levels at the base of the stem of sugarcane growing on Andosols ranged from 190 to 450 µg/kg DM (projet Rebecca; Chopart *et al.*, 2012) or < 130 µg/kg FM (Lesueur-Jannoyer *et al.*, 2012). The concentration of CLD in shoots of *Miscanthus* × *giganteus* and *Miscanthus sinensis* Andersson, were respectively 150 ± 28 µg/kg DM and 260 ± 70 µg/kg DM in soil contaminated at a rate of 1,000 µg/kg DM (Liber *et al.*, 2017). However, Cabidoche and Lesueur-Jannoyer (2012) reported relatively high values (more than 3,000 µg/kg DM) for courgette fruits.

If CLD is bioavailable in the soil and is absorbed by the roots, the first factor that explains the higher CLD content of trees is most probably their woody character in contrast to the herbaceous character of most of the plants studied to date. Cellulose, lignin, and hemicellulose are the main materials in trees. Studying on the adsorption of pesticides, Barak *et al.* (1983) concluded that woody stems adsorbed more of the pesticides than herbaceous stems, and that binding of pesticides in the apoplastic pathway of stems is related to their degree of lignification and to the lipophilicity of the pesticides. Clostre *et al.* (2014b, 2015) underlined the positive correlation between hemicellulose content and the concentration of CLD in root vegetables and cucurbits. Hemicellulose is a complex carbohydrate polymer which accounts for 25-30% of the total dry weight of wood (Pérez *et al.*, 2002). Gérard *et al.* (2019) provide a detailed composition of tropical woods. Among tropical hardwoods, the species we sampled are mainly constituted of cellulose (42.3%), hemicellulose (19.6%) and lignin (29.2%). Conversion into a unit of fresh material makes it possible to compare the quantities expected in hardwoods (cellulose: 244 g/kg FM, hemicellulose: 113 g/kg FM; lignin: 169 g/kg FM), the quantities found in the different plants studied by Clostre *et al.* (2014b; cellulose < 21 g/kg FM, hemicellulose < 30 g/kg FM, lignin < 16 g/kg FM) and in sugarcane (Brienzo *et al.*, 2016; hemicellulose: 27.7% DM; cellulose: 42% DM, lignin: 20% DM). Not surprisingly, the amount of hemicellulose is much greater in trees.

However, other factors may be linked to the difference in CLD contents between trees and monocotyledons or herbaceous dicotyledons. Pascal-Lorber *et al.* (2016) highlighted the fact that the distribution of CLD contamination in grasses resulted from a link between the age of the plant and the evapotranspiration rate of tissues. Even if we cannot guess the evapotranspiration rate of the tree species we studied, their long-life cycle is still a major character. Indeed, a wood ring can remain functional for several years (Gebauer *et al.*, 2008), and the vessels can be crossed by rising contaminated raw saps over a period of many years, like their associated parenchyma, which remain partly alive until the wood enters the duraminization stage. In the monocotyledons studied here, the xylem vessels do not function for long. To conclude, we hypothesise that the longer life cycle and the recurrent circulation of contaminated sap probably explain the high CLD contents observed in the wood at the base of the tree trunk.

Significantly lower CLD contents in trees growing on Andosols

Our second hypothesis was that trees are significantly contaminated whatever the type of soil contaminated. Our results clearly invalidate this second hypothesis: trees growing on Andosols at Site 2 had significantly lower concentrations of CLD in their wood than trees growing on Nitisols at Site 1. Yet, the soils at Site 2 contained double the CLD contents at Site 1. Andosols are known to retain CLD better than Nitisols (Levillain *et al.*, 2012) and less CLD is taken up by plants growing on Andosols (Cabidoche and Lesueur-Jannoyer, 2012). Trees are no exception to this rule. Made up of allophane clays, Andosols contain more organic matter than Nitisols (Dorel *et al.*, 2005; Cabidoche *et al.*, 2009). Due to the aggregative and fractal structure of allophane clays, Woignier *et al.* (2012) showed that the pollutant trapped in the microstructure of allophanic soils could reduce its sensitivity to leaching and its availability to plants. Inversely, Nitisols made of halloysite clays (Sierra and Desfontaines, 2018) are known to release CLD by leaching much more easily than Andosols (Levillain *et al.*, 2012) and are thus more contaminating for plants (Clostre *et al.*, 2015). Thus, organic matter *plus* CLD trapping by allophane clays strongly affect the bioavailability of CLD in Andosols, meaning that total CLD content in Andosols provides no information about the bioavailable CLD content, which is a determining factor (Cabidoche and Lesueur-Jannoyer, 2012).

We were unable to measure the soil characteristics (W_{fc}, BD, K_{oc} and SOC) that provide access to soil $[CLD]_{available}$. However, Cabidoche *et al.* (2009) and Levillain *et al.* (2012) provide a calculation model of $[CLD]_{available}$ as well as mean characteristics (W_{fc}, BD, K_{oc}, SOC; table III) for Andosols and Nitisols. These two types of soils have such contrasted characteristics (table III) that a calculation using average data in the literature allowed us to explain – although roughly – the differences in $[CLD]_{wood}$ between Sites 1 and 2. As expected, $[CLD]_{available}$ in Andosols at Site 2 was much lower than that in the Nitisols at Site 1 (~2.5 $\mu g.l^{-1}$ *versus* ~20 $\mu g.l^{-1}$), regardless of the $[CLD]_{soil}$. This difference in $[CLD]_{available}$ leads to a significant difference in $[CLD]_{wood}$ mean values between Sites 2 and 1 (299 µg/kg *versus* 3,400 µg/kg respectively). In other words, since more CLD is available in the soil, trees can absorb more, regardless of the type of soil. This conclusion is supported by the wood bioconcentration ratios (WCR) obtained for all the trees studied even though they belong to different species. Medians of the 3 box plots (figure 6A; 161, 113 and 190) were not significantly different between sites, a fact supporting the idea that trees have similar CLD absorption efficiencies related to the $[CLD]_{available}$ values rather than to the type of soil.

Mean CLD bioconcentration ratio (WCR) in the trees studied

Could the WCR median value of 160 be considered as a first mean CLD bioconcentration ratio for trees in Guadeloupe? The WCR values of 20 trees out of 24 (83%) ranged between 0 and 346 l/kg and constituted a relatively homoge-

neous group for CLD bioconcentration efficiency. On the other hand, the existence of very high WCR values (1,155 and 1,923), and even extreme values (14,146), mainly in Site 3, raises questions about the variability of WCR in the trees studied here. How should we interpret the high WCR values in Site 3 that differ significantly from the majority of values, even if they come from different species?

Our first hypothesis is that these values reflect real hyper accumulation resulting from specific expression in the Andosols at Site 3. However, two WCR extreme values stem from two species *Inga ingoides* and *Cordia sulcata* that are found at Sites 1 and 2. The WCR values at sites 1 and 2 do not exceed 350 for these two species. Could such large individual variability exist for this trait? The number of trees studied is far too small to support such a hypothesis.

Our second hypothesis is that these very high WCR values are most probably due to a combined effect of our soil sampling strategy around trees and the spatial variability of the CLD concentrations in the soil at Site 3. It will be recalled that Site 3 hosted only a banana plantation mixed with market gardening. So, Site 3 had probably hosted a much lower density of banana trees than Sites 1 and 2 which hosted pure banana plantations (around 2,000 ind/ha). From 1970 to 1993, CLD was applied in solid form only at the foot of each banana tree at an "average" dose of 30 g of commercial product/foot/Ha/year, resulting in soil contamination that varied from the metric to the centimetre scale (Lesueur-Jannoyer *et al.*, 2012). Thus, the more banana trees per unit area, the more treatment points per unit area, which tends to reduce the heterogeneity of CLD distribution in the soil. With a supposed lower density of banana trees, we hypothesise that there is less CLD and greater CLD heterogeneity in Site 3 than in Sites 1 and 2. Indeed, Site 3 had (1) the lowest mean $[CLD]_{soil}$ but (2) the highest variation coefficient (99 *versus* 27 and 21 for Sites 1 and 2, respectively). The greater heterogeneity in Site 3 would also be expected to reduce the likelihood of collecting highly contaminated soil samples using our irregularly spaced soil sampling method. Inversely, the root systems of trees explore large areas (> 10 m^2) and could encounter highly contaminated points present in Site 3 much more easily than we did. Therefore, in Site 3, we can more frequently associate high $[CLD]_{wood}$ with low $[CLD]_{available}$. This combination leads to very high WCR values such as those encountered in Site 3. Naturally, we assume the opposite situation may also exist as the two *Swietenia* trees for which relatively significant $[CLD]_{available}$ values (1.2 and 1.9 µg/l) were associated with very low $[CLD]_{wood}$ values (3 and 2 µg/kg). Either can we exclude the possibility that certain species do not accumulate CLD for various reasons? A deeper root system, hence avoiding CLD that is mainly present in soil superficial layers is one possibility. Given the small number of individuals of each species used in this study, further investigation is required to answer these questions.

Anyway, our study did allow us to get an idea of the ability of trees to absorb CLD. Excluding the supposedly aberrant WCR values, the CLD absorption efficiency of the studied trees (mean: 144 ± 24; median: 120; Var. Coeff.: 75) is very similar to that reported for fruits of *Cucurbita pepo* ssp. *Pepo* cv. Floridor (Organ Concentration Factor: 177-181; Cabidoche and Lesueur-Jannoyer, 2012). Among plants, only a few species, for example, *Cucurbita*, exhibited distinctive capacity for the uptake of notable quantities of persistent organic pollutants (POPs) from the soil (White, 2010) and are considered to be POP hyperaccumulators (Malik *et al.*, 2022). Unfortunately, we were not able to locate any results concerning the relationships between trees and CLD in the literature to compare with our results except for one a study on orchard tree fruits (Cabidoche *et al.*, 2006). Consequently, we were obliged to look for experiments that reported the tree uptake efficiency of other POPs. In a successful experiment of phytoremediation of *Hexachlorocyclohexane* (HCH) in Argentina, Gotelli *et al.* (2020) reported similar POP concentrations (mean $[HCH]_{wood}$: 2,730 µg/kg DM; 300-12,700 µg/kg DM) in wood collected in the first three metres of the trunk of *Eucalyptus dunnii* growing in a highly contaminated soil ($[HCH]_{soil}$: 10-6,000 mg/kg DM). The authors did not provide wood bioconcentration ratio (WCR) values for HCH, but we were able to calculate a mean BTR (see "material and method") from "supplemental material" for *E. dunnii*: 0.01 ± 0.01. This value is significantly below the BTR means calculated for our trees (1.38 ± 0.68, 0.05 ± 0.06 and 0.03 ± 0.02 for Sites 1, 2 and 3 respectively). Although the sensitivity of plants to different POPs varies greatly (White, 2010), our trees growing on much less polluted Nitisols (< 7 mg/kg DM) had similar POP contents (here $[CLD]_{wood}$: 3,406 ± 1,658 µg/kg DM) and consistent CLD uptake efficiency. Thus, we can claim that most of the trees used in our study have a consistent CLD phytoextraction potential.

Conclusion and perspectives

In this study, we found Chlordecone in 100% of our soil samples and in 96% of our tree wood samples. The concentrations of Chlordecone ($[CLD]_{soil}$) ranged from 1,446 to 3,283 µg/kg DM in Nitisols and from 247 to 6,519 µg/kg DM in Andosols. In wood samples, the concentrations of Chlordecone ($[CLD]_{wood}$) ranged from 1,422 to 5,290 µg/kg DM in trees growing on Nitisols and from 0 to 1,114 µg/kg DM for trees growing on Andosols. These are among the highest CLD concentrations recorded in the aerial parts of plants for this POP. Wood contamination capacity was higher for Nitisols than for Andosols, this is explained by the K_{oc} value, which is known to be lower for Nitisols. Applying generic soil property values to both Nitisols and Andosols reported in the literature, we were able to access the volumetric dissolved Chlordecone content ($[CLD]_{available}$) of our soil samples. The volumetric dissolved Chlordecone content explained the contamination of the trees by a satisfactory linear relation (slope: 160) regardless of the tree species. We then calculated a soil-plant bioconcentration ratio (WCR), which is the ratio of the concentration of CLD in dry plant tissue (µg/kg DM) to the concentration of CLD in the soil solution (µg/l). Despite four aberrant values mainly due to the spatial soil sampling method used in Site 3, we found that around 83% of the WCR values of the trees were between 0 and 346 and provide a first mean CLD bioconcentration efficiency for trees (144 ± 24) with no clear distinction between Nitisols and Andosols. We need

further research to explore the variability of CLD uptake efficiency by tree species and the [CLD] contents in the different compartments of the trees, from roots and wood to leaves and fruits.

As part of a future pollution remediation strategy whose contours are not yet known, cultivating trees offers different commercial opportunities and provides interesting services. These services include (1) soil protection against erosion, (2) the production of timber (exported), fruit (citrus fruits, etc.) and organic matter (feeding the litter of the plot), or (3) the sequestration of carbon and CLD that will have been extracted from polluted soils, not to mention many other services (biodiversity, landscape, etc.). By providing sustainable protection of soils against erosion as well as the regular production of living or dead organic matter, occupying the edaphic space, the cultivation of multispecies forest and fruit trees combined with other herbaceous crops could be an effective CLD trap, preventing this POP from escaping to other fragile environments and from contaminating other more fragile crops. But, will this trap be sufficient to sustainably trap most CLD pollution? In this perspective, many questions emerge. What amounts of CLD can we expect trees to sequester, both in their architecture and in the organic matter they release and which "feeds" the soil? Will the products (fruits, leaves, bark) of these agro-forests grown on polluted site be fit for consumption? Will the wood be usable, marketable? Will additional contributions of organic matter and/or activated carbons that have been shown to be good CLD traps still be necessary? Future trials should be implemented with different fast-growing tree species planted on Nitisols to study the within and between-species variability in CLD uptake efficiency. Additionally, new samplings (roots, shoot, wood, rings, bark, leaves and fruits) from adult trees already growing in abandoned contaminated banana plantations could reveal (1) [CLD] gradients in the different compartments (from roots to leaves) of the trees and (2) total CLD contents in adult trees. Finally, leaching experiments should be conducted under different types of plant cover (tree, herbaceous, mixed) to determine which types best retain CLD in the soil.

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This study was carried out thanks to CIRAD's own funds.

Access to data

The raw data are recorded in an Excel file that has been uploaded to the CIRAD-AMAP Dataverse.

The data are of public interest and can be accessed without any particular restrictions by using the following link: https://doi.org/10.18167/DVN1/4AZBLK

Nevertheless, the authors would like to be informed of their possible use, whether by an organisation or a person interested or concerned by the subject of the study.

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Nicolini *et al.* – Author's contributions

Contributor role	Contributor names
Conceptualization	E. Nicolini, J. Beauchêne
Data Curation	E. Nicolini, J. Beauchêne
Formal Analysis	E. Nicolini, T. Hattermann
Funding Acquisition	E. Nicolini, J. Beauchêne
Investigation	E. Nicolini, J Beauchêne, V. Bonnal
Methodology	E. Nicolini, J Beauchêne, V. Bonnal
Project Administration	E. Nicolini, J. Beauchêne
Resources	V. Bonnal
Supervision	E. Nicolini,
Validation	E. Nicolini, J. Beauchêne
Visualization	E. Nicolini, V. Bonnal
Writing – Original Draft Preparation	E. Nicolini
Writing – Review & Editing	E. Nicolini, J. Beauchêne, V. Bonnal

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