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Article *in* Chemosphere · January 2015

DOI: 10.1016/j.chemosphere.2014.06.076 · Source: PubMed

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Chemosphere 118 (2015) 96–102

Contents lists available at ScienceDirect

Chemosphere

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Organochlorine (chlordecone) uptake by root vegetables

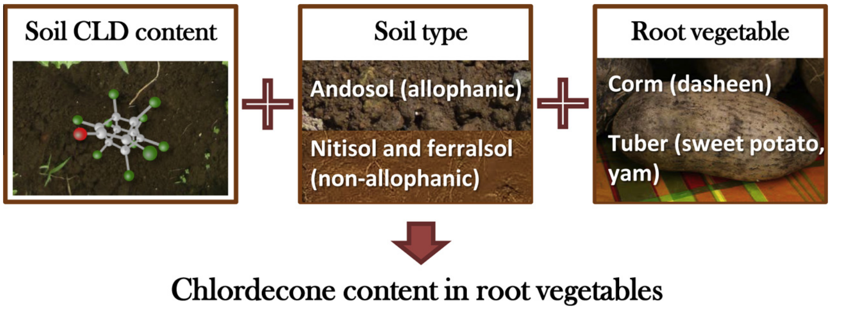
Clostre Florence [a,*], Letourmy Philippe [b], Lesueur-Jannoyer Magalie [a,c]

[a] Cirad/PRAM, UPR fonctionnement agroécologique et performances des systèmes de culture horticoles, B.P. 214 Petit Morne, Martinique, F-97285 Le Lamentin, France
[b] Cirad, UPR Agroécologie et intensification durable des cultures annuelles, Boulevard de la Lironde, F-34398 Montpellier Cedex5, France
[c] Cirad UR HortSys, TA B-103/PS4, Boulevard de la Lironde, F-34398 Montpellier Cedex5, France

HIGHLIGHTS

- Root vegetables can be contaminated over the LMR but not enough for phytoremediation.
- Chlordecone contamination of root vegetables is explained by soil contamination.
- Nitisol and ferralsol are more contaminating than andosol.
- Dasheen corms are more contaminated than yam and sweet potato tubers.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 20 March 2014
Received in revised form 18 June 2014
Accepted 20 June 2014
Available online 17 July 2014

Handling Editor: Myrto Petreas

Keywords:
Soil pollution
Vegetable contamination
Plant uptake
Persistent organic pollutants
Chlordecone

ABSTRACT

Chlordecone, an organochlorine insecticide, continues to pollute soils in the French West Indies. The main source of human exposure to this pollutant is food. Root vegetables, which are staple foods in tropical regions, can be highly contaminated and are thus a very effective lever for action to reduce consumer exposure.

We analyzed chlordecone contamination in three root vegetables, yam, dasheen and sweet potato, which are among the main sources of chlordecone exposure in food in the French West Indies. All soil types do not have the same potential for the contamination of root vegetables, allophanic andosols being two to ten times less contaminating than non-allophanic nitisols and ferralsols. This difference was only partially explained by the higher OC content in allophanic soils. Dasheen corms were shown to accumulate more chlordecone than yam and sweet potato tubers. The physiological nature of the root vegetable may explain this difference. Our results are in good agreement with the hypothesis that chlordecone uptake by root vegetables is based on passive and diffusive processes and limited by transport and dilution during growth.

1. Introduction

Environmental pollution by persistent organic pollutants (POPs) is a worldwide concern. POPs are characterized by a long half-life resulting in their persistence in the environment and often high hydrophobicity with potential bioaccumulation in the food chain. Although POPs were applied decades ago, they continue to pollute the soil, which in turn, releases the pollutants to water, crops, and animals (Miglioranza et al., 1999; Nakata et al., 2002; Gong et al.,

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Abbreviations: POPs, persistent organic pollutants; CLD, chlordecone; DW, dry weight; MRL, maximum residue limit; DS, dry soil; FM, fresh matter; PAHs, polycyclic aromatic hydrocarbons; AIC, Akaike Information Criterion; BIC, Bayesian Information Criterion.

[*] Corresponding author. Address: Cirad, Campus Agro Environnemental Caraïbes, B.P. 214 Petit Morne, Martinique, 97232 Le Lamentin, France. Tel.: +596 596423055; fax: +596 596423001.

E-mail address: florence.clostre@cirad.fr (C. Florence).

http://dx.doi.org/10.1016/j.chemosphere.2014.06.076
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2004). The human body is often the final accumulator of these chemicals, which can lead to health problems. As food consumption is the main pathway for human exposure to environmental contaminants (Nakata et al., 2002; Guldner et al., 2010), public demand for food safety is increasing.

Root vegetables accumulate weathered organochlorine pollutants to a greater extent than other vegetables (Adeyeye and Osibanjo, 1999; Cabidoche and Lesueur-Jannoyer, 2012). The soil is the recipient of different types of pollutants to which root vegetables are directly exposed, thus contributing to dietary exposure to pesticides (Jensen et al., 2003; Caldas et al., 2006). In the French West Indies, root vegetables are among the main sources of chlordecone (CLD) exposure, as they account for more than 65% of total CLD intake by the highly exposed populations (Dubuisson et al., 2007). CLD is an organochlorine pesticide formerly used in agriculture and responsible for long term pollution of soils and ecosystems (Hung et al., 2006; Cabidoche et al., 2009; Coat et al., 2011). Exposure to CLD through food is linked with the incidence of prostate cancer and altered child development (Multigner et al., 2010; Boucher et al., 2013).

A better understanding of the factors that control the transfer of pollutants from soil to plants is needed to assess the risk associated with consumption of foods contaminated by pesticides. Uptake by plants may be influenced by different factors linked to both soil and plant characteristics. The aim of this study was to characterize the contamination of three species of tropical root vegetables by organochlorine pesticide, using CLD as a case study. The lack of chlordecone in airborne particles close to a former production site and its characteristic low vapor pressure and volatility but high affinity for organic matter (Dawson et al., 1979; Cabidoche et al., 2009), led us to hypothesize that the soil–air–plant pathway is much less important than uptake through the soil. This hypothesis is in accordance with reports in the literature that downward translocation of organochlorine pesticides with the same characteristics as CLD, via the phloem assimilation stream is negligible, and that the main uptake route for root vegetables is thus through the soil (Schroll et al., 1994; Fujisawa et al., 2002; Juraske et al., 2011).

Field experiments were conducted to investigate uptake in the main types of soil cultivated in the French West Indies, which have a different potential for CLD transfer. We first measured CLD content in the soil and in the root vegetables and then quantified CLD uptake by the root vegetables. We analyzed within field, between field and interspecific variability of these variables. Here we report the differences between soil types and between crops in light of soil organic carbon contents and of the lipid and hemicellulose contents of root vegetables, as they are known to influence pesticide uptake. We discuss our results with respect to contamination processes.

2. Materials and methods

2.1. Experimental setup and sample preparation

We performed our experiments in fields that had been contaminated in the past to account for reduced bioavailability of the pollutant due to aging (Gevao et al., 2000). At the time scale of a crop, CLD content in soils is considered to be constant as degradation is at best very slow, as is true for leaching (Cabidoche et al., 2009). Our field experiments were conducted in three andosols plots (A1 to A3) located in Morne-Rouge, one nitisol plot (N1) located in Sainte-Marie and one ferralsol plot (N2) in Ducos, Martinique. Andosols are allophanic soils whereas ferralsols and nitisols are non-allophanic soils. Volcanic soils like andosols contain an amorphous clay, allophane, which has completely different structures and physical properties than crystalline clays (kaolinite, halloysite). Organic carbon (OC) content is also higher in allophanic soils than in non-allophanic soils. Because of its strong affinity for allophane organic matter, CLD stored in allophane clay is trapped and protected against extraction (Woignier et al., 2012).

In each plot, we cultivated three crops: dasheen [*Colocasia esculenta*], yam [*Dioscorea spp.*] and sweet potato [*Ipomea batatas*]. A vegetable sample and a corresponding soil sample were taken at each sampling point in the plot. Ten samples of each crop were taken in each plot, except in plot A3 where only sweet potato plants were harvested, and in plot N1, where only five replicates of dasheen were obtained. A vegetable sample corresponds to the corm or to all the tubers produced by one plant. Root vegetables were harvested at commercial maturity and thoroughly washed twice with nanopure water to eliminate soil residue and to avoid CLD contamination by tap water. Samples of unpeeled root vegetables were shipped frozen to the laboratory for analysis. Soil was sampled using a 6–7 cm diameter Edelman hand auger, (SDEC, France). Soil samples were air dried, manually crushed, sieved to 2 mm and finally mechanically crushed in a rotor beater mill (model SR200 by Retsch).

2.2. Sample analysis

2.2.1. Soil characteristics

The physicochemical properties of the soil samples were assessed in one composite sample per plot per crop (Table 1). Organic carbon and the C:N ratio were determined at the CIRAD-Amis-laboratory (Montpellier, France) by dry combustion using a Flash EA – 1112 Series (Thermo Electron Corporation, Waltham, MA) elemental analyzer according to NF ISO 10694.

2.2.2. CLD analysis

Unpeeled vegetable samples were analyzed at the Drôme County analytical laboratory (LDA26) in Valence, France. After extraction and purification, CLD content was measured in a high performance liquid chromatograph-mass spectrometer. The dry matter content of each vegetable sample was also measured.

Soil samples were analyzed at the French analytical laboratory in Martinique (LDA972). After concentration and purification, CLD content was measured by gas chromatography with an electron capture detector for highly contaminated samples and with a gas chromatographer coupled with a mass spectrometer for less contaminated samples.

Both methods are described in detail in Woignier et al. (2012).

As this study was conducted in the framework of public health protection measures, the CLD content we refer to here is the sum of CLD and its main metabolite, 5b-hydroCLD, in the soil. Acute toxicity of 5b-hydro CLD is known to be close to that of CLD (Soileau and Moreland, 1983).

2.2.3. Lipid and hemicellulose analyses

Lipids and fibers were analyzed in a single composite sample of the peel and pulp compartments of each crop (Table 2). Samples

Table 1
Organic carbon content (OC) and C/N ratio in soil composites.

Plot	OC (%)			C/N		
	Dasheen	Sweet potato	Yam	Dasheen	Sweet potato	Yam
A1	3.61	2.69	3.08	11.57	10.12	11.14
A2	4.12	3.12	3.81	11.58	10.09	11.03
A3	/	2.90	/	/	12.24	/
N1	1.58	1.59	1.38	9.7	9.07	9.54
N2	2.08	1.78	1.98	10.19	9.73	10.54

were dried in oven at low temperature (40 °C) to avoid the Maillard reaction and finely ground. Lipid and hemicellulose contents of the unpeeled root vegetables were then calculated using the relative mass contents of the peel and pulp compartments.

Lipids were analyzed at the IDAC laboratory in Nantes, France, according to the NF V04-402 standard (AFNOR, 1968). After hydrolysis with hydrochloric acid, lipids were extracted with petroleum ether. Results are given with a relative error of 5%.

Fiber contents were measured at the CIRAD laboratory (Reunion Island, France) by sequential analysis with Termamyl (heat-stable alpha amylase) to remove the starch, followed by extraction, according to the French standard XP U 44-162 (AFNOR, 2009) after Van Soest (Van Soest et al., 1991). The method is described in detail in Clostre et al. (2014b).

2.3. Statistical analysis

We calculated CLD content in the dry weight of the root vegetables with dry matter content of the vegetable and CLD content in the fresh matter.

To ensure the distribution of ANOVA model residues followed the assumptions of equal variances and normality, we used the log transformed (natural log) data. We analyzed CLD contents in the soil (dry soil, DS), CLD contents in the soil normalized to OC content, and CLD contents in root vegetables (fresh matter, FM, and dry weight, DW). Uptake ratios are the ratios of the concentration in the vegetable to the concentration in the soil, both in mg kg^{-1} DW, as only dry matter content affects contamination capacity. Ratios were calculated as the difference of the log transformed values using both normalized soil CLD contents and non-normalized soil CLD contents. For these six independent variables, the same mixed model was used in SAS software (SAS Institute Inc., 2002-2010).

$$Y_{\mathrm{ijkl}} = \mu + \alpha_i + \beta_j + (\alpha\beta)_{\mathrm{ij}} + \gamma_{\mathrm{jk}} + D_{\mathrm{ijk}} + \varepsilon_{\mathrm{ijkl}} \qquad (1)$$

where Y_{ijkl} is the (log) observed CLD value, i is the crop, j the soil type, k the plot, l the sample; μ the intercept; α_i the effect of crop i, β_j the effect of soil j; $(\alpha\beta)_{\mathrm{ij}}$ interaction between crop i and soil j; γ_{jk} the effect of plot k nested in soil j; D_{ijk} the random effect of crop i and plot k nested in soil j; and $\varepsilon_{\mathrm{ijkl}}$ the residual error.

As our dataset was log transformed, a dispersion index was calculated as half the difference between the limits of the confidence interval (confidence coefficient: 0.68) in Table 3. To analyze within plot variability, we compared models with and without the random effect D_{ijk} and computed the likelihood ratio chi-squared, AIC (Akaike Information Criterion) and BIC (Bayesian Information Criterion).

3. Results

3.1. CLD content in soils

CLD contents in the soil ranged from 160 to 20,300 µg kg^{-1} DS in allophanic plots (andosol plots, A1 to A3), and from 820 to 4330 µg kg^{-1} DS in non-allophanic plots, N1 (nitisol) and N2 (ferralsol). The mean CLD content of the soil varied greatly from on plot to another, especially in the andosol plots (Table 3).

We did not use the data from plot A3 to measure variability of CLD content in the soil, as we only had results for one crop in this plot. We compared the difference in soil CLD contents using model 1. No significant difference was found between soil types ($p = 0.0540$), because for a better comparison of the transfer, we worked with ranges of CLD soil contents that overlapped for the two soil types. Neither the crop effect nor the interaction between crop and soil type was significant, at $p = 0.5459$ and $p = 0.6422$, respectively. On the other hand, there was a significant difference in the level of soil contamination between farms ($F = 42.72$, $p = 0.0020$).

The results showed the same trend after normalization of soil CLD content by OC content, with no significant differences except between farms with $p = 0.0021$.

Table 2
Lipid and hemicellulose content of vegetables (g 100 g^{-1} on a fresh matter basis).

Crop	Lipids	Hemicelluloses
Dasheen	0.16	2.31
Yam	0.04	1.56
Sweet potato	0.12	1.14

3.2. CLD content in root vegetables

All the vegetable samples were contaminated; 86% of them exceeded 20 µg kg^{-1} fresh matter (FM). Irrespective of the type of soil, CLD content in the vegetables ranged from 18 to 336 µg kg^{-1} FM in dasheen, from 2 to 439 µg kg^{-1} FM in yam and from 2 to 209 µg kg^{-1} FM in sweet potato, with mean contents of 157, 77 and 54 µg kg^{-1} FM respectively. In the vegetables grown on allophanic soils, mean CLD contents calculated with model 1 were 244, 109 and 89 µg kg^{-1} DM in dasheen, yam, and sweet potato respectively, and in the vegetables grown on non-allophanic soils, 1005, 234 and 144 µg kg^{-1} DM in dasheen, yam, and sweet potato, respectively. Mean contents per plot on a DM basis are summarized in Table 3.

We also compared CLD contents (DW) in vegetables grown in the same four plots as those described in Section 3.1 using model 1. Crop and soil type had significant effects on the contamination of the vegetables at respectively $p = 0.0193$ and $p = 0.0241$, whereas their interaction was not significant ($p = 0.3945$). There was a significant difference ($F = 16.28$, $p = 0.0120$) in CLD contents in the vegetable between farms, but less than the difference in CLD contents in the soil ($F = 42.72$, $p = 0.0020$).

3.3. Uptake ratios

We used uptake ratios because soil contamination varied widely between and within plots (Levillain et al., 2012; Clostre et al., 2014a), so direct comparison of CLD contents in vegetables would not have been relevant. For sweet potato (the crop for which no data was missing), we plotted mean CLD contents in vegetables (FM) versus mean CLD content in the soil for the five farms as a function of soil type (allophanic or non-allophanic). We hypothesized a simple positive linear relationship between CLD contents in the soil and vegetables, based on the assumption that unpolluted soil cannot contaminate plants (Fig. 1). The slope of the regression for allophanic soils was twice lower (1%) than the slope for non-allophanic soils (2%).

On a dry weight basis, mean CLD contents in root vegetables were lower than in the soil in all the plots sampled regardless of the crop and the soil type (Table 3). To quantify and characterize CLD uptake by root vegetables, we analyzed uptake ratios using model 1. The most significant effect was soil type ($p = 0.0031$) followed by crop ($p = 0.0100$). The farm and the interaction between the crop and the type of soil had no significant effect on CLD transfer ($p = 0.6651$ and $p = 0.1428$, respectively).

We performed contrast analysis to assess differences between crops and soil types. The results of the analysis showed that transfer to dasheen was significantly higher than transfer to yam and sweet potato ($p = 0.0076$ and $p = 0.0055$, respectively), while there was no significant difference in transfer to yam and sweet potato ($p = 0.6549$). There was no significant difference in transfer in the non-allophanic plots (nitisol and ferralsol, N1 and N2; $p = 0.4006$) nor in the allophanic plots (A1 and A2; $p = 0.8897$). All pairwise

Table 3
Mean chlordecone contents in the soil and in 3 root vegetables, and mean uptake ratios calculated with a fixed effects model; dispersion index in parentheses and $n = 10$ for all treatments except $n = 5$ for dasheen in the N1 plot.

	Allophanic soils (andosol)		Non-allophanic soils (nitisol and ferralsol)	
	Plot A1	Plot A2	Plot N1	Plot N2
Dasheen				
Soil (μg kg^{-1} DS)	1352 (163)	7762 (934)	1425 (243)	2210 (266)
Root vegetable (μg kg^{-1} DW)	130.07 (26.59)	457.69 (93.55)	860.32 (250.40)	1105.33 (225.93)
Uptake ratio (DW/DS)	0.096 (0.020)	0.059 (0.012)	0.604 (0.178)	0.500 (0.104)
Sweet potato				
Soil (μg kg^{-1} DS)	744 (90)	8051 (969)	2468 (297)	1595 (192)
Root vegetable (μg kg^{-1} DW)	28.86 (5.90)	272.82 (55.76)	165.47 (33.82)	125.89 (25.73)
Uptake ratio (DW/DS)	0.039 (0.008)	0.034 (0.007)	0.067 (0.014)	0.079 (0.016)
Yam				
Soil (μg kg^{-1} DS)	1326 (160)	9247 (1113)	2355 (283)	2008 (242)
Root vegetable (μg kg^{-1} DW)	32.21 (6.58)	369.33 (75.49)	376.49 (76.95)	145.40 (29.72)
Uptake ratio (DW/DS)	0.024 (0.005)	0.040 (0.008)	0.160 (0.033)	0.072 (0.015)

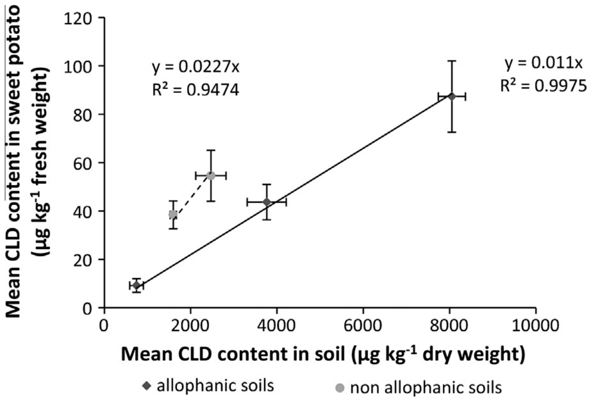

Fig. 1. Mean CLD content in sweet potato (FM) as a function of mean CLD content in soil (DS) in the five plots, with the dispersion index for each plot and linear regressions for each soil type (allophanic and non-allophanic); calculated with log transformed data.

differences in transfer between allophanic and non-allophanic plots were significantly different.

Uptake ratios calculated with soil CLD contents normalized to OC contents were analyzed using model 1 to check if OC content could account for the observed differences. Results followed the same general trend as that of the uptake ratios without normalization, i.e. with the same two significant effects but the crop effect ($p = 0.0055$) was more significant than the effect of the type of soil ($p = 0.0306$).

3.4. Within field variability

In the model comparison, regardless of the variables (CLD contents in the soil, in the vegetables, or uptake ratios), AIC and BIC were systematically lower when the random effect in the model was taken into account.

The random effect was significant for CLD content both in the soil ($p = 0.009$) and in the root vegetables ($p = 0.011$) underlining the high variability at plot scale. For uptake ratios, within field variability was lower, as the random effect was not significant ($p = 0.078$) but almost; and AIC and BIC values showed that the random effect improved the model. Thus, there was additional uptake variability at the within field scale.

4. Discussion

First, we measured CLD content in the soil and its variability at within and between plot scale and analyzed the level of contamination in root vegetables. Second, we confirmed that contamination of vegetable was related to soil contamination and compared uptake ratios between soil types and crops. Below, we discuss our results in light of the contamination processes.

In our study, the plot had a significant effect on soil CLD content whereas the crop did not. The range of soil contamination was wider in the andosol plots than in the two non-allophanic plots. We also showed that within plot variability of soil CLD content was relatively high. These findings are in agreement with previous results on CLD pollution of soils in the French West Indies (Levillain et al., 2012; Clostre et al., 2014a).

It is worth noting that the CLD contents in our plots were relatively high compared to the usual levels of organochlorine found in soils, even though they are representative of the contamination levels found in the French West Indies (Cabidoche et al., 2009; Levillain et al., 2012; Clostre et al., 2014a). They were higher than total or individual organochlorine pesticide contents measured in agricultural soils in Argentina, Mexico, England and the USA in other works (Miglioranza et al., 1999; White et al., 2005; Zohair et al., 2006; Waliszewski et al., 2008) but in the same range as chlordane levels found in experiments conducted by Mattina et al. (2000) in a field previously used for herbicide trials.

All three root crops were shown to take up organochlorine. Most of the samples were contaminated over the maximum residue limit (MRL) of 20 μg kg^{-1} FM (European Union, 2008), which they exceeded up to 10–20 times depending on the crop. CLD contamination has also been studied in radish and turnip (Cabidoche and Lesueur-Jannoyer, 2012; Clostre et al., 2014c), and results showed that the levels in their tubers sometimes also exceeded the MRL. Previous studies in carrot, potato, radish, turnip and beet, showed that root crops can accumulate other weathered hydrophobic organochlorines, e.g. DDT, lindane, heptachlor, PCDD/PCDF, chlordane, endrin, from soils (Müller et al., 1994; Miglioranza et al., 1999; Mattina et al., 2000; Poulsen and Andersen, 2003; Zohair et al., 2006). Our results confirm the general ability of root vegetables to be contaminated by CLD at levels that raise concerns for consumer health.

The concentration of organochlorines in roots depends to a great extent on the concentration in the soil (Mikes et al., 2009). In their study on the uptake of organic pollutants from soil, Schroll et al. (1994) found clear relationships between soil and plant contamination only in root crops. Carrots were also shown to accumulate lipophilic organochlorine pesticides as a function of soil content (Zohair et al., 2006; Waliszewski et al., 2008). However, Zohair et al. (2006) found no positive correlation in potato. Cabidoche and Lesueur-Jannoyer (2012) found a linear relationship between CLD content in soil and in yam tubers. For these reasons, and because regulations are based on pesticide content in the fresh product, and soil content is usually expressed on a dry weight basis, especially by laboratories, we studied the relationship between mean CLD contents in sweet potato (FM basis) and soil (DW basis). We then quantified uptake by calculating the uptake ratio (DW basis) for each crop and each plot to be able to compare the results with those of other studies on hydrophobic organochlorine uptake by root crops.

CLD contents were lower in root vegetables than in the corresponding soil regardless of the crop or the plot concerned. In sweet potato, the mean CLD content of the soil explained the mean CLD content of the root vegetable (FM) in both allophanic and non-allophanic soils, thus supporting the hypothesis of the soil being a major uptake pathway for CLD. Transfers were low with a regression slope in the range 1–2% in both soil types. Depending on the type of soil, mean uptake ratios (DW:DS) ranged from 0.06 to 0.60 for dasheen, 0.03–0.08 for sweet potato and from 0.02 to 0.16 for yam. By way of comparison, in studies on organochlorines, uptake ratios (including DDT and HCH) in carrots ranged from 0.15 to 0.3 (Zohair et al., 2006); while in another study, uptake ratios for DDT and HCH in carrots were close to 1 (Waliszewski et al., 2008). In radish, organochlorine uptake ranged from 0.03 to 0.74 depending on the compound and on the growth conditions (Mikes et al., 2009). For chlordane, mean uptake ratios were 0.06 in red potato, 0.08 in white potato and 0.04 in beet (Mattina et al., 2000). The uptake ratios of CLD we found for yam and sweet potato were thus in the same range as those observed for uptake of other hydrophobic organochlorines by potato and beet. In a nitisol field, Cabidoche and Lesueur-Jannoyer (2012) found CLD uptake ratios (bulk transfer ratios) for yam of between 0.058 and 0.161 $\mu g\,\mu g^{-1}$ and for sweet potato, of between 0.037 and 0.120 $\mu g\,\mu g^{-1}$. Our results are thus in agreement with these authors in comparable conditions. The good correlation between CLD contents in the soil and root vegetables and the low uptake ratios confirm the hypothesis of a passive transfer from soil to root crops.

No significant difference in uptake was found between plots. Nevertheless, the heterogeneity of soil content in addition to between plot variability of uptake account for the high variability of CLD contents in root vegetables at plot scale. Variability of uptake between plots was due to variability of soil conditions and plant response (between individual variability).

None of the crops concentrated CLD efficiently (uptake ratios < 1). The relatively low levels found in root vegetables on a fresh weight basis compared to CLD in the soil may be partly due to aging of the pollutant and plant growth. Aging of organochlorine pesticides in soil has been shown to reduce their bioavailability (Gevao et al., 2000; White et al., 2005). Moreover, tubers are not directly connected to the root system or to the transpiration stream but are loaded via the phloem from leaves, which is assumed to be free of CLD. The concentration of lipophilic compounds in thick roots and tubers is thus limited by dilution due to growth through filling processes (Trapp, 2002; Trapp et al., 2007; Paraíba and Kataguiri, 2008).

In sweet potato grown on allophanic soils, CLD transfer to the vegetables (FM basis) was twice lower than on non-allophanic soils. Uptake ratios for the three crops confirmed that transfer on andosols was lower than on ferralsols and nitisols, with factors of 5–10 for dasheen, 2 for sweet potato and 2–7 for yam. The type of soil is known to influence the behavior of pesticides in the soil and their release into the environment (Gevao et al., 2000; Kumar and Philip, 2006). Other authors have reported that the different types of soil in the French West Indies have contrasted ability to contaminate different crops but also water: andosol have been shown to be less contaminating than other types of soil (Cabidoche et al., 2009; Cabidoche and Lesueur-Jannoyer, 2012; Woignier et al., 2012). Similarly, a preliminary study reported two to three times lower CLD uptake ratios for sweet potato (vegetable FM:DS) (data not shown) on andosol than on nitisol. Our results confirm that ferralsols and nitisols behave similarly regarding the transfer of CLD from the soil to the plant, probably because their OC contents are close, but their similar clay type could also play a role (Woignier et al., 2012).

The amount of pesticides adsorbed on soils is generally considered to be linked to soil OC content (Kanazawa, 1989; Barriuso et al., 1997; Li et al., 2003). Allophanic soils are usually more contaminated by CLD than non-allophanic soils and have also higher OC contents (Brunet et al., 2009; Cabidoche et al., 2009; Clostre et al., 2014a). Hence CLD content in the soil may depend on the OC content. Concentrations of organic contaminants in the soil normalized to soil organic matter content is a better estimator of soil contamination potential than raw soil content (Chiou et al., 2001; Hung et al., 2009). By normalizing soil CLD content using plot OC content, we tested if the OC content could explain reduced CLD availability for plants growing on allophanic soils compared to non-allophanic soils. After normalization, the difference in uptake between the types of soils was still significant but less so than without normalization. Thus, even if OC content partially explains the lower uptake, alone it cannot account for the difference observed.

Our results are in agreement with those of Woignier et al. 2012, who showed that, even if CLD had a high affinity for organic matter (Chevallier et al., 2010; Woignier et al., 2013), the high organic carbon contents of allophanic soils alone could not explain the reduced transfer of CLD to the crop observed in this type of soil compared to non-allophanic soils. These authors hypothesized that trapping of the pollutant in the microstructure of allophanic soils could contribute to the lower availability of CLD for plants.

The three crops were not contaminated to the same extent. Dasheen was more contaminated than yam and sweet potato, which had similar uptake ratios. Likewise, Cabidoche and Lesueur-Jannoyer (2012), found no significant difference between organ concentration factors calculated for yam and sweet potato. The difference cannot be attributed to the length of the period of contamination, as the crops had the following order of maturity: sweet potato (five months), dasheen (seven months) and yam (ten months). The three root vegetables also differ from a physiological point of view: sweet potato and yam are tuberous roots whereas dasheen is a corm, i.e. a modified stem (Onwueme and Charles, 1994; Onwueme, 1999), which could partially account for the higher contamination of dasheen, although at this stage we do not have a physiological explanation.

The difference in CLD uptake between crops could be linked to a difference in lipid and fiber contents, as these plant components have an affinity for organic pollutants. In plants, lipid contents have been shown to be positively correlated with the level of pesticide residues (Schroll et al., 1994; Simonich and Hites, 1995; Trapp, 2004; Li et al., 2005). According to CLD K_{ow} values, CLD uptake by a crop could mainly be controlled by lipid uptake (Chiou et al., 2001). Lipophilic pesticides may be adsorbed on fibers with different sorptive properties depending on the plant component and the chemical compound concerned (Harms, 1992; Ta et al., 1999; Rodríguez-Cruz et al., 2009). In a previous study (Clostre et al., 2014b), we showed that only lipids and hemicelluloses explained the differences in CLD contamination observed between the peel and the pulp of root vegetables and cucurbits.

As the lipid contents in dasheen and sweet potato were similar but lipid contents in yam were far lower (Table 2), lipid content cannot account for the difference in CLD contamination between these root crops. Conversely, hemicellulose contents were highest in dasheen followed by yam followed by sweet potato (Table 2), which is similar to the level of contamination of root vegetables by CLD. However, at this stage, we do not have enough data for statistical analysis and consequently cannot draw any firm conclusions, especially because uptake variability was high and there were far fewer differences between hemicellulose contents than between uptake ratios.

When interpreting our results, the processes underlying the uptake of pollutants by plants should be kept in mind, particularly uptake by tuber and corm root crops. CLD is a hydrophobic organic compound, poorly volatile and non-ionized (Cabidoche et al., 2009) and its uptake by root crops can be compared with the uptake of other hydrophobic compounds. The processes that account for the contamination of edible roots of root crops by non-ionized pesticides are (1) incorporation into the root core via the transpiration stream through root hairs, (2) adsorption by root epidermis, (3) diffusion from the root epidermis to the core (Fujisawa et al., 2002).

The translocation of POPs and highly lipophilic compounds from the soil to the shoot tissues in the transpiration stream has been shown to be low (Wild et al., 2005; Trapp, 2007). POP uptake by plant is limited by their high affinity for soil organic matter and for the surface of roots (Wang and Jones, 1994; Trapp, 2002) resulting in a negative correlation between K_{oc} and transfer in potato (Paraíba and Kataguiri, 2008). With a K_{oc} of between 1200 and 15,800 L kg^{-1}, depending on the soil type (Howard, 1991; Schüürmann et al., 2006; Fernandez Bayo et al., 2013) and a log K_{ow} around 5 (United States Environmental Protection Agency, 2012), CLD uptake should thus be limited (Cabidoche et al., 2009), and this hypothesis was confirmed by our results. Once adsorbed by the skin of the root vegetable, lipophilic and organochlorine pesticides are then incorporated very slowly by diffusion through the skin into the inner tissues (Fujisawa et al., 2002; Trapp, 2002; Trapp et al., 2007; Juraske et al., 2011). Previous studies confirmed that the skin of the root vegetable remained the most highly contaminated by CLD compared to flesh (Cabidoche and Lesueur-Jannoyer, 2012; Clostre et al., 2014b). Moreover, as the time scale of diffusion depends largely on the radius, due to the small surface-to-volume ratios of root vegetables, highly lipophilic chemicals with a long half-life are unlikely to reach equilibrium within one vegetation period (usually less than 150 days) in carrot (Trapp, 2002). In the root vegetables studied here, the vegetation period ranged from 150 to 300 days depending on the crop, but even with these longer times for diffusion, the CLD contents in the vegetables were still lower than in the soil.

5. Conclusion

Weathered CLD was transferred from soil to roots. Although CLD uptake by the plant was relatively low, the risk of exceeding the health threshold (MRL) for CLD in root vegetables remains, raising concerns for consumer safety. When assessing risk, it is important to keep in mind that correct preparation of root vegetables by thorough washing and peeling considerably reduces exposure (Cabidoche and Lesueur-Jannoyer, 2012; Clostre et al., 2014b). The variability of soil content and of uptake justifies not only using mean values, but also quantile regressions, in risk assessment.

Based on the possibly deleterious effects of CLD on health, intake should be kept as low as possible. The public should be better informed about the risk of contaminants in foodstuffs and, in particular, how they can reduce their own exposure by washing and peeling root vegetables. As the ability of root vegetables to take up CLD is now confirmed, they should only be grown on soils with the lowest rates of soil contamination. Root vegetables traditionally form a substantial part of the staple diet of people who live in the tropics and normally contribute to a healthy diet. A decision tool should thus be designed to help home gardeners grow healthy products and to support farmers in redesigning their production systems in order to comply with regulations (MRL). Unlike recommendations to consumers on how to prepare these foods that apply regardless of the type of soil (Clostre et al., 2014b), recommendations to farmers should take the type of soil into account as uptake by root vegetables was shown to be lower in allophanic soils.

The contamination of other types of vegetables and fruits now needs to be investigated along with the root-to-shoot translocation of this organochlorine pesticide. Other crops than those studied here are known to be "pesticide uptakers" and thus contribute to exposure via food (Mattina et al., 2000; White, 2002; Cabidoche and Lesueur-Jannoyer, 2012).

Acknowledgments

We thank the staff of the Chamber of Agriculture of Martinique and the farmers. We are grateful to Eliane Marville from CIRAD Martinique for sharing her expertise.

This work was supported by the French Chlordecone National Plan and Regional Health Agency of Martinique through the "JAFA"; Family Garden Health Program.

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