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Uptake and distribution of chlordecone in radish: Different contamination routes in edible roots

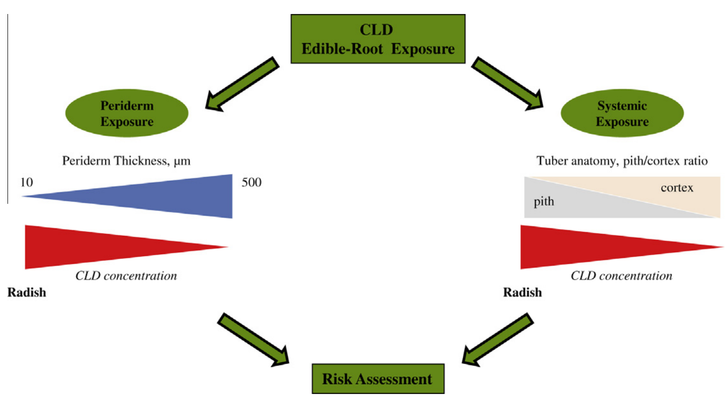
Clarisse Létondor [a,b,c,d], Sophie Pascal-Lorber [a,b,c], François Laurent [a,b,c,*]

[a] Université de Toulouse, INP, UPS, EcoLab (Laboratoire Ecologie Fonctionnelle et Environment), ENSAT, Avenue de l'Agrobiopole, F-31326 Castanet Tolosan, France
[b] CNRS, EcoLab, 31326 Castanet Tolosan, France
[c] INRA, EcoLab, 180 Chemin de Tournefeuille, F-31027 Toulouse, France
[d] ADEME, 20 Avenue du Grésillé, F-49004 Angers, France

HIGHLIGHTS

- Two routes of radish tuber contamination by chlordecone (CLD) were highlighted.
- A systemic way by CLD root absorption then translocation by transpiration stream.
- A non systemic way by CLD periderm adsorption then radial diffusion into flesh.
- CLD exposition of generic tubers depends on tuber location in transpiration stream.
- Exposition depends also on periderm thickness and properties of storage tissues.

GRAPHICAL ABSTRACT



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ABSTRACT

Chlordecone (CLD) was an organochlorine insecticide mainly used to struggle against banana weevils in the French West Indies. Forbidden since 1993, it has been a long-term contaminant of soils and aquatic environments. Crops growing in contaminated soils lead to human exposure by food consumption. We used radiolabeled [^{14}C]-CLD to investigate the contamination ways into radish, a model of edible roots. Radish plants were able to accumulate CLD in both roots (RCF_{35d} 647) and tubers (edible parts, CF_{35d} 6.3). CLD was also translocated to leaves (CF_{35d} 1.7). The contamination of tuber was mainly due to peridermic adsorption or CLD systemic translocation to the pith. TSCF was 3.44×10^{-3}. CLD diffused across periderm to internal tissues. We calculated a mean flux of diffusion J through periderm about 5.71×10^{-14} g cm^{-2} s^{-1}.

We highlighted different contamination routes of the tuber, (i) adsorption on periderm followed by diffusion of CLD towards underlying tissues, cortex, xylem, and pith (ii) adsorption by roots and translocation by the transpiration stream followed by diffusion from xylem vessels towards inner tissues, pith, and peripheral tissues, cortex and periderm. Concerning chemical risk assessment for other tubers, contamination would depend on various parameters, the thickness of periderm and CLD periderm permeance, the origin of secondary tissues – from cortex and/or pith – , the importance of xylem flow in tuber, and the lipid amount within tuber.

Abbreviations: CF, concentration factor; CLD, chlordecone (1,1a,3,3a,4,5,5,5a,5b,6-decachlorooctahydro-2H-1,3,4-(methanetriyl)cyclobuta[cd]pentalen-2-one); DW, dry weight; FW, fresh weight; FWI, French West Indies; RCF, root concentration factor; TF, translocation factor.

* Corresponding author at: INRA – USC 5245 EcoLab (Laboratoire Ecologie Fonctionnelle et Environment), 180 Chemin de Tournefeuille, Bâtiment A – BP 93173, F-31027 Toulouse Cedex 3, France. Tel.: +33 561285392; fax: +33 561285244.

E-mail address: francois.laurent@toulouse.inra.fr (F. Laurent).

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1. Introduction

Persistent Organic Pollutants (POPs) are widespread organic chemicals that accumulate in tissues of animals due to their persistence in the environment. Chlordecone (CLD, molecular formula $C_{10}Cl_{10}O$, CAS registry number 143-50-0) is an organochlorine insecticide classified as a POP in 2009 and its use and production were prohibited in 2011 by the Stockholm Convention. Stability of organochlorine molecules increases with the number of chloride atoms whereas the solubility in water decreases. These compounds are strongly hydrophobic and weakly degraded by microorganisms (Aislabie et al., 1997). However some anaerobic bacteria are able to partially dechlorinate these molecules into soils (Orndorff and Colwell, 1980; George and Claxton, 1988).

CLD is strongly apolar and its cage structure makes it highly stable into soils. Moreover, there is not any evidence of CLD degradation in the natural environment. Cabidoche et al. (2009) have estimated a half-life of 30 years for CLD whereas that of dichlorodiphenyltrichloroethane (DDT) is 15 years (Mackay et al., 1997). CLD has a relatively high lipophilicity with an octanol–water partition coefficient (K_{ow}) value of 4.50 (Cabidoche et al., 2006). It has a strong affinity for organic matter (Dawson et al., 1979) but different K_{oc} values between the equivalent fraction sorbed on the soil carbon and the fraction soluble in water were reported in literature, from 2000 L kg^{-1} (Bonvallot and Dor, 2004) to 17 500 L kg^{-1} (Kenaga, 1980).

CLD was used as insecticide on tobacco plants, ornamental shrubs, and lemon trees and to trap ants and cockroaches. It was mainly used in the French West Indies (FWI) from 1972 to 1993 to struggle against banana black weevils (*Cosmopolites sordidus*), which larva cause damages in shoots. It was applied at a dose of 60 kg ha^{-1} (5% active ingredient), advantageously replacing application (180–270 kg ha^{-1}, 25–50% active ingredient) of hexachlorocyclohexane, an organochlorine pesticide used between 1960 and 1972. Production of CLD at Hopewell (Virginia, USA) was stopped in 1976, following an accident at the production factory that caused serious neurological troubles to the workers and local people. This accident evidenced the acute toxicity of CLD. Multigner et al. (2010) have pointed out the chronic toxicity of CLD by correlation between the increased rate of prostate cancer and CLD exposure in FWI. A recent study has shown that pre- and postnatal chronic exposure to environmental levels of CLD is correlated to negative effects on cognitive and motor functions during infancy (Dallaire et al., 2012; Boucher et al., 2013). CLD use was forbidden in France in 1990 but was continued until 1993 in FWI, due to weevil proliferation after two cyclones in the early nineties.

Depending on the origin of the contamination, from banana plantations to watersheds, CLD in soils cover a wide range of concentrations, up to a few mg kg^{-1}. CLD is currently found in all compartments of catchment areas where CLD was applied, water resources with bio-magnification in aquatic food chains, soils, and plants. Concentrations vary from 0.5 to 10 $\mu g\ L^{-1}$ in rivers of Guadeloupe and in shrimps (*Macrobrachium faustinum*), which are at the apex of aquatic chains but concentrations could be higher than the RML of 20 $\mu g\ kg^{-1}$ (Coat et al., 2011). According to the high lipophilicity of the molecule, CLD uptake by plants was considered as very unlikely. Topp et al. (1986) have found that CLD uptake by young seedlings of barley (*Hordeum sativum* L.) and cress (*Nasturtium officinale*) was negligible from soil. However, Cabidoche et al. (2006) have shown that crops cultivated on CLD contaminated soils could accumulate CLD, particularly in tuber plants, which grew in direct contact with soil. Root absorption was evidenced for other POPs such as HAPs in carrot, lettuce, and potato (Wild and Jones, 1992; Fismes et al., 2002). Webber et al. (1994) have shown the uptake of organochlorine contaminants, HAPs, pesticides, and particularly PCBs by corn, carrot, and cabbage. Several studies have shown that some organochlorine compounds, with physicochemical properties close to those of CLD, could be transferred from soil to shoots. DDT and its metabolite, dichlorodiphenyldichloroethylene (DDE), were taken up and translocated to shoots of *Cucurbita pepo* species, graminae, and alfalfa (Lunney et al., 2004). *Cucurbita* species specifically accumulated OCs. Weathered *p,p′*-DDE was taken up and stored in stems, leaves, and fruits of varieties of *Cucumis* and *Cucurbita* (White, 2002). *C. pepo* subspecies *pepo* accumulated a significant concentration of PCBs in plant shoots via root uptake and translocation (Whitfield Åslund et al., 2007, 2008). PCBs were identified in the root, shoot, and for the first time in xylem tissues of pumpkin (Greenwood et al., 2011). These authors showed that PCBs were able to subsequently diffuse into shoot tissues from xylem vessels. In addition, significant concentration of chlordane ($\log K_{ow}$ 4.74) and its residues were found in the edible root tissues of carrots, beets, potatoes and in the aerial tissue of spinach, lettuce and even bioaccumulation in fruits of zucchini (*Cucurbita pepo* L ssp. *pepo*) (Mattina et al., 2000).

A recent study of Cabidoche and Lesueur-Jannoyer (2012) has shown that the contamination rate of aerial parts of plants (shoots, leaves, and fruits) by CLD varied depending on the plant species. For example, tomato fruits remained CLD-free while courgette was largely contaminated due to the root production of organic acids that facilitated the desorption from soil followed by adsorption by plants (White et al., 2003).

Nowadays, human exposure to CLD is mainly due to consumption of contaminated food (Multigner et al., 2010). However, data remain scarce and conflicting concerning contamination pathways of plants by CLD. In this context, the aim of the present study was to understand how edible root contamination occurred. To achieve this goal, we used radiolabeled [^{14}C]-CLD to study the ways of contamination into radish (*Raphanus sativus* cv. Nelson). We highlighted various contamination routes: (i) absorption by roots followed by translocation in whole plants via the evapotranspiration stream and diffusion from xylem vessels and (ii) peridermic adsorption then diffusion to tuber parenchyma. According to these results, contamination of edible roots could be predicted according to their different anatomic characteristics, thickness of periderm, type of parenchyma development (phloem or xylem), physiology and histology.

2. Materials and methods

2.1. Chemicals

[^{14}C]-Chlordecone (specific activity = 1.443 GBq $mmol^{-1}$, radiochemical purity 97% as determined by radio-reverse phase-high performance liquid chromatography analysis) was purchased from Moravek (Brea, CA). [U-^{14}C-imidazolidine]-Imidacloprid (specific activity = 9.916×10^2 MBq $mmol^{-1}$, radiochemical purity 97% as determined by radio-reverse phase-high performance liquid chromatography analysis) was obtained from Institute of Isotopes Co. (Budapest, Hungary). Analytical standard chlordecone (Fluka 45379, PESTANAL®) and all other chemicals were obtained from Sigma–Aldrich (Saint-Quentin Fallavier, France) and were of analytical grade. Solvents used in the experiments were provided by Scharlau Chemie S.A. (Barcelona, Spain).

2.2. Determination of radioactivity

Radioactivity in liquid samples was determined by direct counting on a Packard scintillation analyzer (model Tricarb 2200CA, PerkinElmer Life and Analytical Sciences, Courtabœuf, France)