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# Long-term pollution by chlordecone of tropical volcanic soils in the French West Indies: A simple leaching model accounts for current residue

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**Abstract**
Chlordecone was applied between 1972 and 1993 in banana fields of the French West Indies. This resulted in long-term pollution of soils and contamination of waters, aquatic biota, and crops. To assess pollution level and duration according to soil type, WISORCH, a leaching model based on first-order desorption kinetics, was developed and run. Its input parameters are soil organic carbon content (*SOC*) and *SOC*/water partitioning coefficient ($K_{oc}$). It accounts for current chlordecone soil contents and drainage water concentrations. The model was valid for andosol, which indicates that neither physico-chemical nor microbial degradation occurred. Dilution by previous deep tillages makes soil scrapping unrealistic. Lixiviation appeared the main way to reduce pollution. Besides the SOC content and rainfall increases, $K_{oc}$ increased from nitisol to ferralsol and then andosol while lixiviation efficiency decreased. Consequently, pollution is bound to last for several decades for nitisol, centuries for ferralsol, and half a millennium for andosol.

**Capsule**: Soil and water contamination by chlordecone will persist for several centuries in the French West Indies, because the only decontamination is through chlordecone leaching by drainage water.

**Keywords**: chlordecone; pollution; persistence; soil; water

## 1. Introduction

Chlordecone (CLD), an organochlorine insecticide, was used in the French West Indies to control the banana weevil *Cosmopolites sordidus* during two periods: (i) 1972 to 1978 under the trade-mark Kepone™ (5% CLD) manufactured by Life Science Products Co. (Hopewell, VA), and (ii) 1982 to 1993 under the trade-mark Curlone™ (5% CLD), manufactured by Calliope S.A. (Port-la-Nouvelle, France). Kepone received a temporary use license from the French Ministry of Agriculture in 1972. Its use ceased in 1978 after its prohibition in the USA. A French paper (Snegaroff, 1977) warned against the hazard of soil and water contamination by CLD. Several years after the beginning of Kepone spreading, the topsoil of some banana fields exhibited CLD content higher than 9 mg $kg^{-1}$. Despite a report intended for the French Environment Ministry (Kermarrec, 1980), which highlighted the contamination of fresh water and terrestrial fauna, the French authorities delivered in late 1981 a license for the use of CLD (Curlone). CLD spreading was restricted to banana fields. Its use was definitively banned in early 1993. Later surveys conducted by the French Department of the Environment (DIREN, 2001) and the French Department of Health (DSDS, 2001) revealed its wide presence in soils, rivers, spring water, but also in drinking water and food crop produce. Authorities immediately asked scientists to investigate pollution duration and trends.

Because chlordecone had been prohibited for a long time in the USA, the scientific literature on chlordecone environmental behavior was sparse and more or less confusing. Given its high peripheral chlorination (Figure 1), CLD hydrophobicity is high. It has a low solubility in water, except at pH > 9 where chlordecone-hydrate appears (Dawson et al., 1979). It is only largely soluble in hydrogenated organic solvents like benzene or hexane, as attested by the high partitioning ratio between octanol and water (log $K_{ow}$ = 4.5). Its partitioning coefficient ($K_{oc}$) between the sorbed part on soil organic matter (estimated by the soil organic carbon, *SOC*) and the dissolved part in water is high, and its value varies widely: 17 500 L $kg^{-1}$ according to Kenaga (1980), log $K_{oc}$ of 3.3 (Bonvallot and Dor, 2004) or between 3.38 and 3.41 (Howard et al., 1981), i.e. $K_{oc}$ between 2 000 and 2 500 L $kg^{-1}$. Those differences could derive from using soils with different physico-chemical properties. Spatial variability of soil pesticide sorptivity has been described for atrazine by Coquet and Barriuso (2002), but the variation did not cover such a wide range of relative values.

The vapor tension of CLD is less than 3 x $10^{-2}$ Pa at 25 °C, and the sublimation temperature is over 350°C (INCHEM, 1984). Solar irradiation of CLD in the presence of ethylene-diamine results in 78% degradation after 10 days, but no study of the degradation products or their toxicity has been undertaken (Dawson, 1978).

There is no evidence of microbial CLD degradation. Using *Pseudomonas aeruginosa* strain KO3 and a bacterial pool isolated from Hopewell plant sludge under aerobic conditions, Orndorff and Colwell (1980) obtained an apparent de-chlorination into mono-hydro-chlordecone and di-hydro-chlordecone. But those authors did not describe the initial mono-hydro- and di-hydro-chlordecone contents of the CLD, so that those values could derive from uncompleted chlorination during the production process. Later, Georges and Claxton (1988) studied CLD degradation by three *Pseudomonas* spp. But again details were lacking. Moreover, the HPLC chromatogram of non-inoculated CLD solution, with only yeast and glucose addition, showed peaks corresponding to hydro- and dihydro-chlordecone. The authors underlined that its highly chlorinated cage-like structure makes CLD a poor carbon source for bacteria: after two weeks, the apparent degradation reached only 15 to 25% with a high relative uncertainty of 10% on chlordecone HPLC concentration measurements.

Only one reference was dedicated to CLD absorption by plants. Topp et al. (1986) studied the uptake of several $^{14}C$ labeled organic molecules by young barley (*Hordeum sativum L.*) and cress (*Nasturtium officinale)*. Measured CLD uptakes were negligible for both species. We therefore speculated that soil leaching was the main factor of decontamination.

To account for inter-annual chlordecone desorption and current level of soil pollution, we chose to build a simple leaching model of CLD, WISORCH, based on first-order desorption kinetics. Because of slow decontamination and long-term soil pollution, existing models of pesticide dissipation were not appropriate: they account for short-term sorption, leaching, and degradation so that applying them over years should result in excessive error. In WISORCH, we assumed that neither degradation, nor plant capture were efficient to remove CLD from the various volcanic tropical soil types of the French West Indies. For validation, because inter-annual CLD leaching experiments were impossible, we used a "space for time" approach: we reconstructed the various schedules of CLD spreading on fields and compared the simulated soil CLD contents to the measured ones.

## 2. Material and methods

### *2.1. Soils*

Table 1 summarizes the distribution pattern of the volcanic soil types and properties according to Colmet-Daage and Lagache (1965). All primary minerals of andesitic rocks are weathered, so that soils have a high content of secondary minerals: halloysite for nitisol, halloysite and Fe-oxihydroxides for ferralsol, and allophane for andosol, the three main soil types contaminated in Guadeloupe. All have a centimeter-size granular structure in the topsoil. Intermediate horizons have polyhedral to massive structure and contain many tubular pores that allow high hydraulic conductivity at saturation. In the "clay" matrix, there are very fine pores smaller than 1 µm, where water and solute transfers are slow. In addition, Martinique has skeletic andosol and young raw soil containing pumice gravels, deriving from recent pyroclasts. All these soil types are acidic, which prevents clay dispersion and sheet erosion. Hydric erosion appears to be due only to bad soil management practices, which concentrate runoff that then forms streams that are able to carry aggregates. Carbon contents are unusually high for tropical soils, in particular for untilled andosols (Table 2).

### *2.2. On-farm parcel survey*

For andosol and andic nitisol, we identified parcels from a previous survey on 36 parcels in the south of Guadeloupe. On these parcels, the diversity of banana cropping systems has been described for the last 15 years (Clermont-Dauphin et al., 2004). Among the 36 parcels, we retained 17 parcels that offered consistent and various CLD spreading dates and tillage depth values, among which 10 parcels had never been tilled. A parcel on andosol was added in Martinique. Two never-treated parcels were sampled downstream from contaminated ones. All together, 18 parcels on andosol and 2 on andic nitisol were well documented. The same approach was applied to ferralsol, ferralic nitisol, and ferralic fluvisol in Guadeloupe, giving 7, 1, and 5 parcels, respectively. In Martinique, the survey was mainly dedicated to the main soils of banana areas: skeletic young andosol (7 parcels) and nitisol (5 parcels). All parcels in Martinique had been deeply tilled during the 70s and 80s. As a whole, 45 parcels were included in the "space for time" survey, in which 43 had been treated more or less recently and more or less frequently, and 2 never treated (Table 2).
For each parcel, we questioned the corresponding farmers about the former applications of Kepone or Curlone. For validation, their statements were compared to the supplies of Curlone to farmers that had been recorded by authorities. Additional questions and observations were focused on spreading location (diffuse or localized), tillage (or not, tool, tractor, tillage depth). During the period of availability, from 1972 to 1978 and 1982 to 1993, CLD was generally applied at a yearly dose of 3 kg $ha^{-1}$.

### *2.3. Sampling and soil analysis*

In the perennial banana cropping systems, located on heavy slopes, 3 boreholes 8 cm in diameter at 0, 1/4, 1/2 of the distance between banana trees were sampled at the top and the bottom of the slope. The depths of sampling in each borehole were 0-10 cm, 10-30 cm, and 50-70 cm. Composite samples of 0-30 cm were obtained by mixing (1/2 in volume) the first two layers in each topographic position.
To account for the effect of tillage on spatial variability of soil contamination, 20 samples of 0-30 cm layer were taken on each parcel of tilled banana cropping system. On each composite sample, CLD and carbon (SOC) contents were determined.
In addition, bulk density was determined in profiles for each soil type and related to the carbon content. The resulting relations were used to calculate bulk density from SOC for each composite sample, which allowed converting gravimetric into volumetric contents for SOC and CLD. We focused measurements on the 0-30 cm depth layer for two reasons: (i) this depth is most commonly used to estimate C sequestration and (ii) the same depth was sampled during the same period by the crop protection service as part of its program survey of soil pollution. Some repetitions allowed us to establish that there was no significant change in CLD or SOC contents from one year to the other. Soils were sampled between 2001 and 2005.

### *2.4. Chlordecone analysis*

All soil composite samples were analyzed in the LDA26 at Valence (France), which works under the French accreditation committee "COFRAC" and norm NF17025.

The samples successively underwent (i) air drying, (ii) crushing, (iii) homogenizing, and (iv) CLD extraction using dichloromethane + acetone (vol. ratio 50/50). CLD was finally determined by GC-MS-MS "triple quadrupole" (Varian, MS1200), which was calibrated using the standard addition method and two internal standards: hexabromobenzene and triphenylphosphate. Two transitions were systematically applied: 272 > 237 (-20eV) and 274 > 239 (-15eV). The resulting average extraction coefficient was 0.85, with a confidence interval of [0.70; 1.05]. Thus, each raw result was divided by 0.85.

*2.5. Chlordecone leaching model: "WISORCH"*

This acronym summarizes the key words "West-Indies", "Soil", "ORganoCHlorine"

Despite a time lag of 10 to 30 years since the last spreading, the soils exhibited high CLD contamination, without any change from one year to another. We assumed that only lixiviation by percolation water was able to slowly reduce soil contamination. We chose to apply a simple elution model that follows a first order kinetics. For each elementary percolation volume (d$D$), the amount of lixiviated chlordecone (-d$S$) is proportional to the remaining stock ($S$) in the soil:

$$-\mathrm{d}S/\mathrm{d}D = C \times S \quad (1)$$

where C is the ratio between the CLD concentration in water (d$S$/d$D$) and the stock S in the soil, i.e. the inverse of the partition coefficient, at equilibrium, between soil sorbed CLD and dissolved CLD which is equal to $(K_{oc} \times S_{soc})^{-1}$ where $S_{soc}$ is the organic carbon stock of soil (Mg ha$^{-1}$) and $K_{oc}$ (L kg$^{-1}$) the partitioning coefficient of CLD between *SOC* and water. After integrating, it becomes:

$$\ln(S/S_0) = -C \times D \quad (2)$$

or

$$S = S_0 \times \exp(-C \times D). \quad (3)$$

With $S_0$ the initial storage after spreading, i.e. with $D$=0.

The WISORCH model is intended for studies of the long-term leaching of chlordecone. It has thus been discretized at the yearly time step. In each year-step, this model implicitly alternates (i) an open system resulting in a displacement of the dissolved CLD fraction by drainage and (ii) a closed system allowing renewal of this dissolved fraction by desorption of the stored CLD. This is generally true at a daily time step, where heavy rainfall amounts alternate with a few dry days. The remaining stocks resulting from the different CLD applications over time are eluted simultaneously, which supposes that the soil storage capacity for CLD remained unsaturated: the ratio between the maximal cumulated CLD spreading and the minimal carbon content of the 0-30 cm layer never exceeded 2%.

In the year $j$, the remaining stock $S_{ij}$ (kg ha$^{-1}$) of a CLD spreading $S_i$ (kg ha$^{-1}$) that was applied during the year $i$ depends on the amount of cumulated annual drainages $D_n$, $\sum_{n=i}^{j} D_n$ (mm), between the years $i$ and $j$, following the equation:

$$S_{ij} = S_i \times \exp\left(\frac{-10}{K_{oc} \times S_{soc}}\left(\sum_{n=i}^{j} D_n\right)\right) \quad (4)$$

In the first approach, $K_{oc}$ was taken as 17 500 L kg$^{-1}$ according to Kenaga (1980). The factor 10 comes from the units chosen.

The cropping system is accounted for at two levels. First, a tillage depth ($z_{til}$) exceeding the storage layer depth z results in CLD mechanical dilution; the CLD storage accounted for in year $i$ is $S_i \times z/z_{til}$ if $z_{til}$ exceeded $z$. If not, $z_{til}=z$. Secondly, we accounted for the rainfall redistribution by plant canopy, which induces forced drainage ($D_f$) at the foot of the banana stem downstream from the stemflow (Cattan et al., 2007b), which crosses the foot spreading of CLD. The different cycles of banana harvest come from successive suckers from a mother plant, which do not grow exactly at the same place. Indeed, we assumed that, only during the $a$ first years following the spreading of the year $i$, $D$ was the forced drainage ($D_f$) at the banana feet where CLD had been applied. The value of $a$ was 3 years for tilled banana fields where the plants are renewed, and 5 for untilled fields, that is the delay before the successive suckers migrated out of the initial spreading area. $D_f$ was taken as $1.2 \times R$ , where $R$ was the annual rainfall amount (mm), for single-row or staggered-row banana fields (Sansoulet et al., 2007), and $1.5 \times R$ for double-row banana fields (Thieuleux, 2006). After this period of forced drainage, the drainage $D_b$

was considered homogeneous and estimated according to water balance. The total CLD storage $S_j$ (kg ha$^{-1}$) that remains in the soil in the year *j* is then:

$$S_j = \sum_{i=1971}^{j} \left[ S_i \times (z / z_{til}) \times \exp\left( \frac{-10}{K_{oc} \times S_{soc}} \left( \sum_{n=i}^{i+a} D_{fn} + \sum_{n=i+a+1}^{j} D_{bn} \right) \right) \right] \quad (5)$$

The water budget was established as follows:

$$D_b = R - PET - Rf \quad (6)$$

where *R*, *PET,* and *Rf* were the annual amounts (mm) respectively of rainfall, potential evapo-transpiration, and runoff .

Plant interception loss was neglected as Cattan et al. (2007a) showed its value does not significantly differ from zero because of the low wettability of banana plants. In addition, using a yearly time step allowed us to neglect the soil water content variation, which amounted to less than 70 mm while cumulative drainage represented more than 15 m from leaching simulations for the 10 years or more, i.e. since the last spreading. *R* was calculated by interpolation from local linear functions of elevation (Figure 2), which were established from average rainfall data of Meteo-France (1972 to 2005) on 3 transects following the slope in Guadeloupe. According to the results of Robin (1990), *PET* depends on global radiation and temperature, which decrease as elevation increases. Like *R*, *PET* was calculated from elevation using a linear function (Figure 2). Then, combining the two linear models, *PET* was expressed as a function of *R*: $PET = 1.18 \times R - 1905$. Since we could not measure runoff for each parcel, we used the results from two experimental sites: (i) the first one at Neufchâteau (soil NFC, Guadeloupe) under banana cropped on andosol where runoff and rainfall were measured from 2001 to 2002 on a half hectare plot (Cattan et al., 2006) and from 2003 to 2004 at the banana 8 m² mesh scale (Ruy et al., 2005; Sansoulet et al., 2007), and (ii) the second one at Rivière Lézarde (soil BRH5, Martinique) where runoff and rainfall were measured from 1999 to 2000 under banana cropped on nitisol on a 200 m² parcel (Khamsouk and Roose, 2003). On andosol, 2001 and 2002 gave low runoff coefficients (*RfC*), between 0.035 and 0.07, depending on cropping system (perennial or tilled banana, with or without leaf and stem mulch), with an average *RfC* of 0.06 (Figure 3). For these years, the average of monthly rainfall calculated for the periods of runoff measurement was close to the average of monthly rainfall calculated from 1972 to 2002 at the same site. Only abnormal rainy periods in the last few months of 2004, which included particularly high intensity rainfall events, gave *RfC* between 0.3 and 0.6. To take into account the variability of annual runoff, we retained a range of *RfC* of 0.06±0.06. The two extreme values correspond to the interception of (i) the empirical logarithmic curve describing *RfC* as a function of the average of monthly rainfall during periods of runoff measurement and (ii) the 95% confidence interval of the average of monthly rainfall over 30 years (Figure 3). For nitisol, Khamsouk and Roose (2003) found *RfC* of 0.025 on average in 1999 and 2000 for banana cropping and between 0.045 and 0.072 on bare nitisol, i.e. the same order of magnitude as for andosol. In addition, the soil surface states did not vary significantly since CLD spreadings: 38 over 45 parcels remained cropped in banana and the 7 others were under recent grass fallow after banana. For these reasons the same range of *RfC* as for andosol, i.e. 0.06±0.06, was finally retained for all parcels of our survey.

For the upper layer of soil whose depth is z, Eq. (5) has the following detailed expression according to elementary input variables:

$$S_j = \sum_{i=1971}^{j} \left[ S_i \times (z / z_{til}) \times \exp\left( \frac{-10}{K_{oc} \times SOC \times BD} \left\{ \sum_{n=i}^{i+a} R_n \times (D_f / R) + \sum_{n=i+a+1}^{j} (R_n \times (1.18 - RfC) - 1905) \right\} \right) \right] \quad (7)$$

WISORCH outputs include also:

(i) the corresponding CLD gravimetric content $[CLD]_c$ (mg kg$^{-1}$)of the z cm layer, following the equation:

$$[CLD]_c = S_j / (10 \times z \times BD) \quad (8)$$

(ii) the concentration of drainage water $[CLD]_w$ (µg L$^{-1}$) at the bottom of the storage layer, at the end of the year *j*, following the equation:

$$[CLD]_w = 10^5 \times (S_j - S_{j-1}) / D_{bj} \quad (9)$$

*2.6. Error and sensitivity analysis*

Figure 4 shows the contribution of the variables to the error on $[CLD]_c$ for the 0-30 cm layer. CLD application and tillage depth ($z/z_{til}$) errors have the main impacts, followed by soil gravimetric carbon content (*SOC*), in contrast with bulk density (*BD*). Average annual rainfall ($R_n$) error has less impact but potential evapo-transpiration has a lot ($PET = 1.18 \times R - 1905$). The runoff/rainfall coefficient (*RfC*) error seems to have a low contribution, but its relative error increased by 100%. If the model is applied more than 10 years after the last CLD spreading, the forced drainage due to rainfall concentration at the banana foot ($D_f/R$) and its duration ($a$) have the lowest impact.
Finally, we applied the following errors to the calculated CLD storage $S_j$ or its corresponding gravimetric content $[CLD]_c$. For $S_{ij}$, ± 10%, because the plant density varied between 1800 and 2200 plant/ha even if each banana foot received a well defined dose. For $z_{til}$, ± 2 cm for perennial banana parcels (uncertainty of borehole sampling considered as a tillage error), and ± 7 cm for tilled banana parcels. For *R*, ± 10% represents the sum of the inter-annual average confidence interval and of the interpolation uncertainty. For *RfC*, ± 100% of relative uncertainty.

### *2.7. Drainage and runoff waters*

Drainage leachates were collected using fiberglass wick lysimeters at 65 cm depth. The length of the wick was calculated to apply the same suction as the average of the surrounding soil material. We verified that the lysimeter cumulated flow corresponded to the natural soil cumulated flow by using the HYDRUS 1D or 2D models (Šimůnek et al, 2006). The detailed method and experimental validation were described in Sansoulet et al. (2007). No solids transfer was observed on filters.
Lysimeters were buried under and between banana plants for single-row banana field NFC on andosol. For the double-row banana field BRH5 on nitisol, three positions were used: under banana tree, under the small 1.2 m inter-row, and under the large 2.4 m inter-row. Dates of leachate measuring and sampling are given in Fig. 7.
As the WISORCH model allows calculating CLD concentration of leachates $[CLD]_w$ outing an upper layer, we iterated its application to the lower layer, considering that leachates of the first one were a contamination source for the second one, whose lower boundary corresponded to the lysimeter depth. Two flow patterns were applied in WISORCH to calculate $[CLD]_w$ at 65-cm depth: matrix drainage (MD) and dual drainage (DD). MD applied $D_b$ to the area without stemflow influence. DD was used at the banana feet receiving stemflow. At this place, the drainage flow was split into (i) a matrix flow $D_b$, whose $[CLD]_w$ resulted from sorption and desorption in the second layer and (ii) a drainage over- flow equal to ($D_f$-$D_b$) that lixiviated the upper layer and crossed the lower layer through macropores, without interaction.
Runoff was collected and sampled downstream from lysimeter parcels on andosol and nitisol, at the same dates as lysimeter sampling.
$[CLD]_w$ were measured using GPC-MS by Institut Pasteur Guadeloupe, at Pointe-à-Pitre.

## 3. Results

### *3.1. Soil pollution levels*

No parcel was contaminated without CLD spreading, even if it was downstream from contaminated parcel.
The $[CLD]_m$ measured contents and WISORCH parameters and variables are listed in Table 2 for the 45 parcels. Untilled andosol showed the highest $[CLD]_m$ in the 0-30 cm layer. This is in accordance with the persistence of CLD mainly in the first 0-10 cm layer, which reached more than 50 mg kg$^{-1}$ of CLD. The 50-70 cm layer never reached more than 1 mg kg$^{-1}$, and it generally contained less than 0.3 mg kg$^{-1}$. The high $[CLD]_m$ of the upper 0-30 cm layer was related to high SOC content, between 35 and 85 g kg$^{-1}$. For tilled andosol, the 0-30 cm layer had lower $[CLD]_m$, even though the SOC content was equal. This finding derived from the mechanical dilution of CLD by former deep tillage. Opposite to andosol, nitisol had very low $[CLD]_m$ content, even if spreading occurred during a long time. This could be explained by the lower *SOC* content or a lower $K_{oc}$.

### *3.2. Modeling chlordecone persistence and losses by leaching with WISORCH*

The *SOC* content is accounted for by the WISORCH model. For each parcel, we compared $[CLD]_c$ values, calculated using 0.3 m for depth and 17.5 m$^3$ kg$^{-1}$for $K_{oc}$, and $[CLD]_m$ measured values on 0-30 cm soil samples taken in parcels (Table 2, Figure 5). Each soil type shows a linear relation between $[CLD]_c$ and $[CLD]_m$ (Figure 5). The linear regressions and variance explanation ratios $R^2$ are given in Table 3. For andosol, the model gives nearly the same calculated $[CLD]_c$ as measured $[CLD]_m$ contents in soil, with an $R^2$ exceeding 0.99. Conversely, for nitisol, $[CLD]_c$ was on average 10-fold higher than $[CLD]_m$. The regression slopes were higher for ferralsol and nitisol. Inverting the WISORCH model allowed us to fit the apparent $K_{oc}^*$ for each soil type. Average apparent $K_{oc}^*$ were significantly different and decreased from tilled andosol (12 to 25) to ferralsol (7.5 to 12) and then nitisol (2 to 3 m$^3$ kg$^{-1}$) (Table 3).
We applied the iteration of WISORCH to the lower layer of NFC andosol above the lysimeters. The tilled layer was 32 cm thick, and the lower B layer was between 32 and 65 cm, which is the depth of the lysimeters. Figure 6 show the accordance between $[CLD]_c$ and $[CLD]_m$ in 2003, using the same $K_{oc}^*$ of 19.8 m$^3$ kg$^{-1}$ for each layer.

Since spreadings, the model showed contamination of the lower layer increased while that of the upper one decreased.

*3.3. CLD concentrations of leachates*

The $[CLD]_w$ of runoff without solid charge were more than 3-fold lower than in drainage, while runoff volume was 10-fold lower than drainage volume. Consequently, CLD loads in runoff represented less than 1/30 of those in drainage, and were neglected.

That the apparent $K_{oc}^*$ decreased from andosol to nitisol could be due to a lower extraction during analysis. But no abnormal CLD recovery was observed after standard addition. The CLD loads of drainage water were measured during a period of moderate to intense drainage under the NFC andosol (Figure 7a). Both cumulated drainage and lixiviation of CLD were lesser under the bare soil between banana feet and during the first phase of moderate drainage under banana foot than under banana foot during the last phase of intense drainage. The slopes of the regression lines, i.e. the average $[CLD]_w$ in drainage water, were respectively 0.8 and 2.2 µg $L^{-1}$. For the BRH5 nitisol period, only intense drainage was collected (Figure 7b). The differences between the three lysimeter positions were low, for both CLD lixiviation and concentration: the large inter-row lysimeter collected drainage resulting from dropping points at the extremity of the banana leaves (Bassette, 2005), which increased water fluxes at this place. Overall, although $[CLD]_m$ was only 0.26 mg $kg^{-1}$ in the top nitisol (against 4.6 for andosol), the concentrations of drainage water of similar volumes were 3- to 4-fold higher for nitisol than for andosol. This finding shows that the desorption capacity of nitisol was higher than that of andosol.

*3.4. Applying WISORCH to calculate $[CLD]_w$*

The apparent $K_{oc}^*$ were used in the WISORCH model to estimate the inter-annual evolution of $[CLD]_w$ for the different soil types (Figure 8). For NFC andosol, the DD pattern applied to the water flow below banana foot and the MD pattern applied under bare soil accounted for the respective $[CLD]_w$. For BRH5 nitisol, Figure 8 shows that both DD and MD applied to the water flow below banana gave the same simulated $[CLD]_w$, which was similar to the ones measured at the sampling date. The different $K_{oc}^*$ did not result from experimental bias. $K_{oc}^*$ was lower for nitisol than for andosol, which has higher CLD sorption and lower desorption capacity.

**4. Discussion**

Some of our results were expected. Notably, the soils were polluted only where chlordecone had been spread, except at emergence of contaminated water tables (data not shown). Indeed, the very low volatility of chlordecone makes atmosphere unable to carry any measurable contamination. Moreover, the CLD loads in runoff flow accounted for less than 3% of those in drainage. Consequently, the inter-parcel contamination remained rare. The soil pollution was spatially delimited. We observed only current redistribution of chlordecone from upper to lower layers and very slow decontamination by leaching. Drainage was the main transfer vector. As drainage feeds water tables, and downstream emergence areas and rivers, diffuse pollution affects the water resources for a long time. This diffuse pollution could not be reduced by diminishing surface runoff and favoring infiltration.

WISORCH, unfortunately, corroborated the hypothesis of long-term persistence of CLD in the soils of the French West Indies. Despite the high rainfall and the high associated drainage, which could ensure efficient chlordecone leaching, the high $K_{oc}$ and high carbon content of the soils, which allows a high secondary "clay" mineral content (Feller et al., 2001), make soil decontamination very slow. The extrapolation of soil decontamination from WISORCH is given in Figure 9, after spreading of 60 kg $ha^{-1}$ $year^{-1}$ of "Kepone" and then "Curlone" between 1972 and 1993. This simulation shows that it will take 6 to 10 decades, 3 to 4 centuries, and 5 to 7 centuries before nitisol, ferralsol, and andosol, respectively, are depolluted by leaching. The earlier decontamination of the top soil of nitisol will not be an advantage until it is completed: its lower $K_{oc}$ results in a higher water contamination capacity. As a consequence, both water-tables and below-ground harvested crops will undergo sustainable contamination by all soil types. This points need to be studied.

Even though it is known that $K_{oc}$ has a wide spatial variability, we need to understand why such large differences have been highlighted in this research: the roles of the organic matter types and organo-mineral relations need further investigation.

Applying WISORCH requires having sufficiently long and spatially dense climatic and hydrologic data, both rare under tropical conditions. In particular, runoff/rainfall coefficients, which are low on the acidic soils and under the covering crops we studied, can be so high or variable for other soil types and crops that WISORCH would not operate. This situation would occur if the vertisols of the driest areas were contaminated.

**Conclusion**

Chlordecone, an organochlorine insecticide in the POPs prohibition list of the Stockholm Convention (UNEP, 2005), was regularly applied in the banana crops of the French West Indies at the end of the last century. Our results show that the simple WISORCH model, which applies a first-order kinetics of lixiviation based on *SOC*

content, soil carbon / water partitioning coefficient $K_{oc}$, and drainage to the soil chlordecone stock, accounts for the current chlordecone residues of volcanic soils in Guadeloupe and Martinique.
Given the low volatility and solubility of this molecule, neither atmospheric nor runoff inter-parcel contamination was observed. Chlordecone remained where it had been applied. The unusually high *SOC* content of the volcanic soils and $K_{oc}$ values allow their strong and persistent contamination by hydrophobic chlordecone, reaching tens of mg $kg^{-1}$ in the top layer of andosol. Remediation by scrapping remains unrealistic: most of the parcels were tilled at 60 or 70 cm depth since CLD spreading. Even though it was applied to soils receiving chlordecone 30 years before, the lixiviation model remains valid. It shows indirectly that no microflora selection allowing chlordecone degradation occurred.
The persistence of chlordecone differs between soil types. The $K_{oc}$ of nitisol is lower than that of ferralsol or andosol. As a consequence, the soil decontamination will take some decades for nitisol, several centuries for ferralsol, and more than half a millennium for andosol. Facilitating SOC mineralization would be a way to accelerate decontamination, but it would lead to agronomical and environmental problems.
Both model and experiment values verified that the lower the $K_{oc}$, the higher the chlordecone concentration in water. This poses problems in terms of contamination hazard mapping because there is not a univocal relation between soil pollution and its contamination capacity for water and plants. Further research is needed to understand why the different organo-mineral matrixes have different behaviors in chlordecone sorption.

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Figures

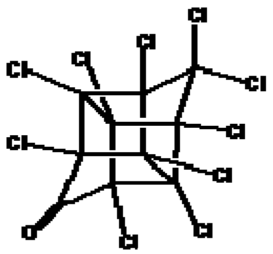


**Figure 1: The molecule of chlordecone**

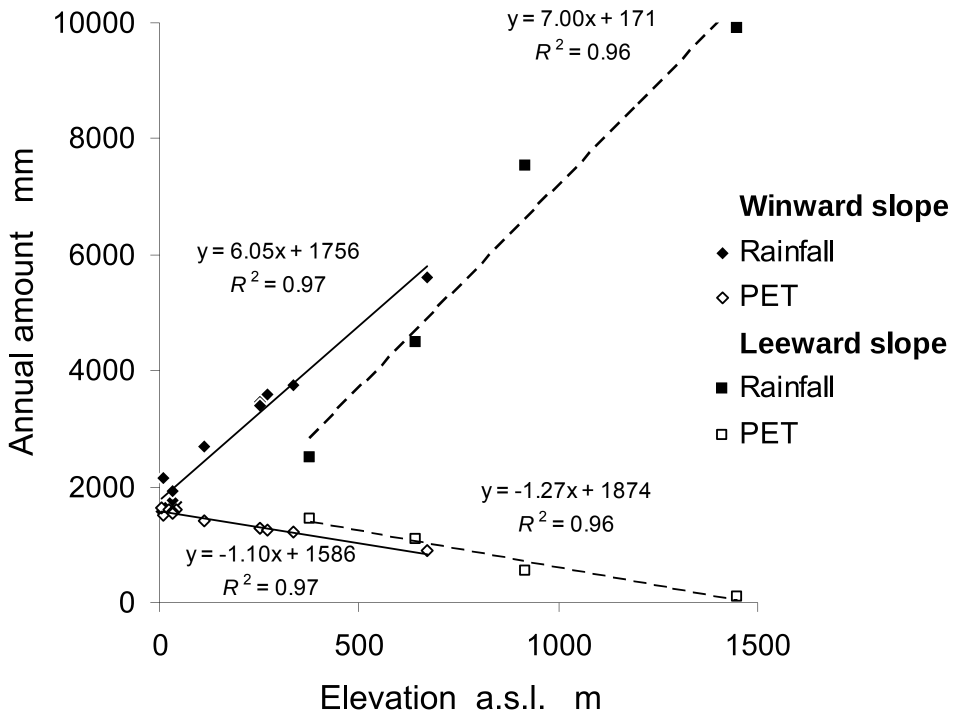


**Figure 2: Relations between rainfall or potential evapo-transpiration (PET) and elevation above sea level (a.s.l.) on the two sides of the volcano chain of Guadeloupe.** Rainfall is the average of the annual rainfall data from 1972 to 2003 (Meteo-France, 1972 to 2003); PET is the potential evapo-transpiration according to Robin (1990). Continuous lines are regression lines for windward slope values, dashed lines those for leeward slope ones.

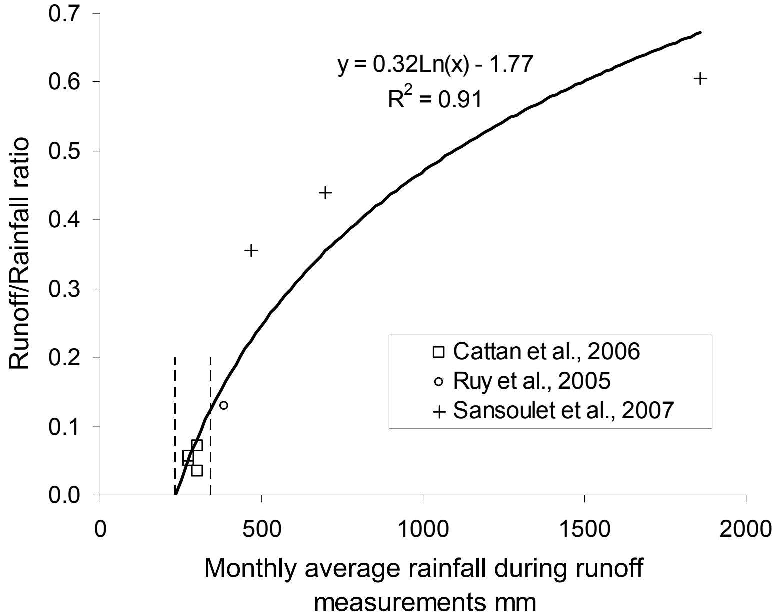


**Figure 3: Rainfall/Runoff ratios as a function of relative monthly rainfall amounts measured on andosol cropped with banana at Neufchâteau (Guadeloupe), at the 0.6 ha parcel scale between September 2001 and December 2002 (Cattan et al., 2006) and at the banana mesh plot between August and October 2003 (Ruy et al., 2005) and between June and November 2004 (Sansoulet et al., 2007).** Vertical dotted lines indicate the confidence interval of the relative average monthly rainfall between 1972 and 2003.

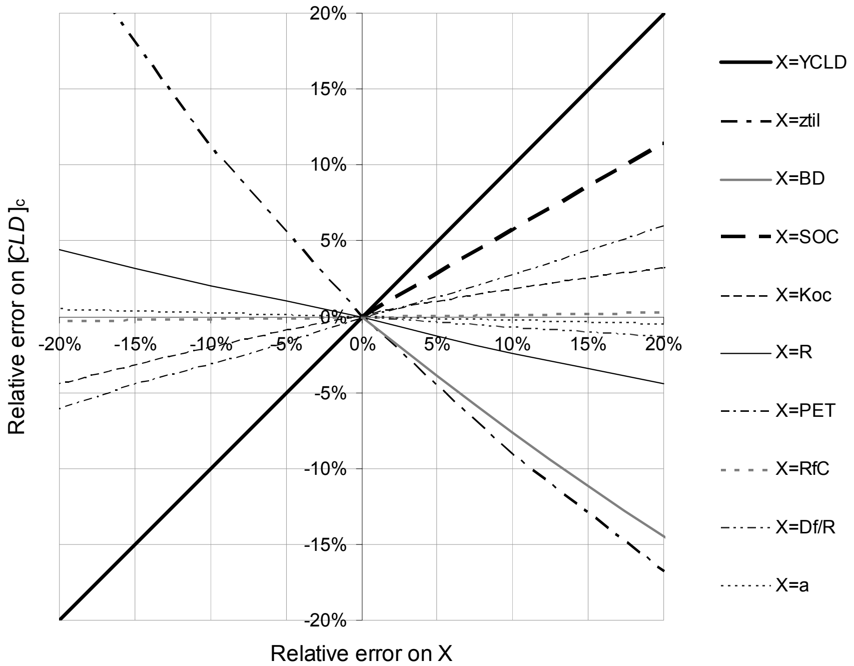


**Figure 4: Impact of relative errors of each variable accounted for in the WISORCH model on the errors on calculated values of the chlordecone gravimetric content $[CLD]_c$ in the 0-30 cm layer of soils.** The most impacting variables are chlordecone spreading schedule (*YCLD*) and depth of tillage ($z_{til}$). Then soil gravimetric carbon content (*SOC*), in contrast with bulk density (*BD*). Then average annual rainfall (*R*), in contrast with potential evapo-transpiration (*PET*). Runoff/rainfall coefficient (*RfC*), forced drainage due to rainfall concentration at the banana tree foot (*Df/R*), and its duration (*a*) have a lower impact.

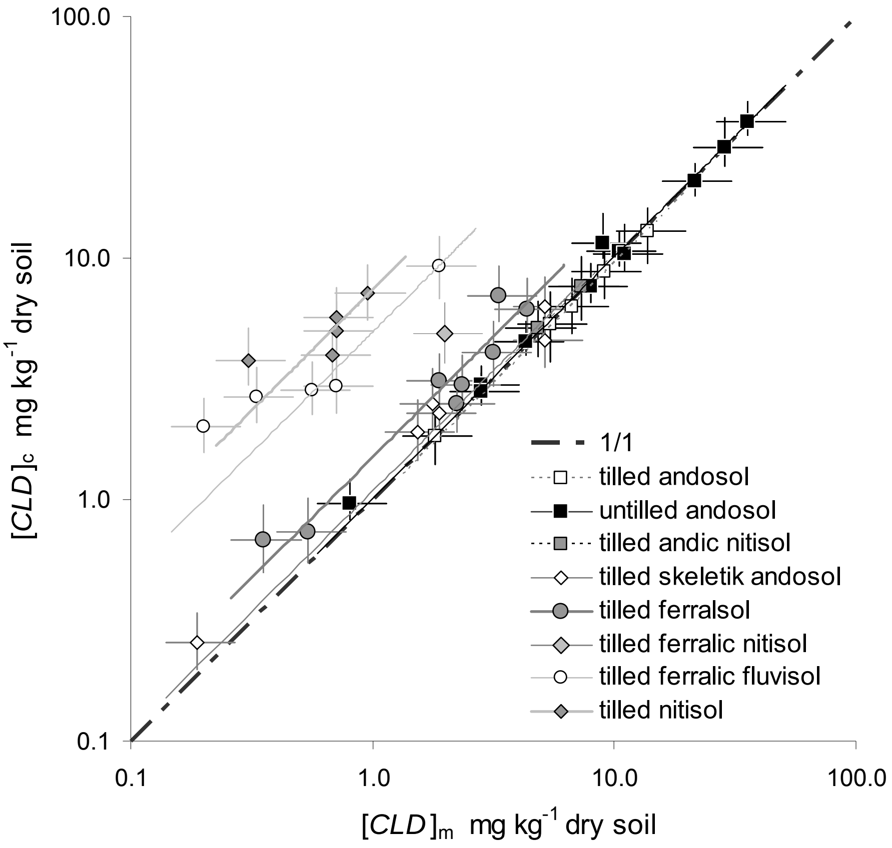


**Figure 5: Relations between calculated [*CLD*]$_c$ and measured [*CLD*]$_m$ chlordecone contents of the 0 to 30 cm layer of several soil types deriving from volcanic ashes in the French West Indies**
The calculated values are outputs of the leaching model "WISORCH", using a $K_{oc}$ of 17.5 $m^3\ kg^{-1}$.
Each point abscissa is the corrected value of chlordecone content, which was the measured one, divided by 0.85, i.e. the average recovery coefficient of standard additions.
Straight lines are the linear regression lines for each soil type.
Horizontal bars represent the uncertainties on measured contents, which are mainly affected by the dispersion of apparent recovery coefficients (between 0.7 and 1.05 for chlordecone additions) and by sampling uncertainty.
Vertical bars represent the uncertainties on calculated contents, which are mainly affected by uncertainty on tillage depth estimation (± 7 cm) or sampling depth for untilled soils (± 2 cm), cumulated CLD spreading (± 10%) and interpolated mean annual rainfall between spreading and measurement year (± 10 %).

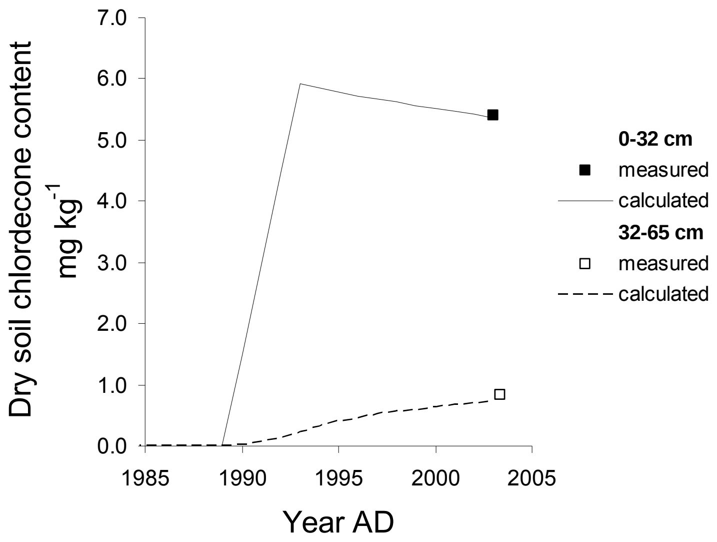


**Figure 6: Applying the WISORCH model to deeper layer contamination by chlordecone lixiviation from the top layer.**
NFC site, andosol, $z_{til}$ = 32 cm, $K_{oc}$= 19.8 m$^3$ kg$^{-1}$, *SOC* = 65 for top (0-32 cm) and 32 g kg$^{-1}$ for bottom (32-65 cm) layer.

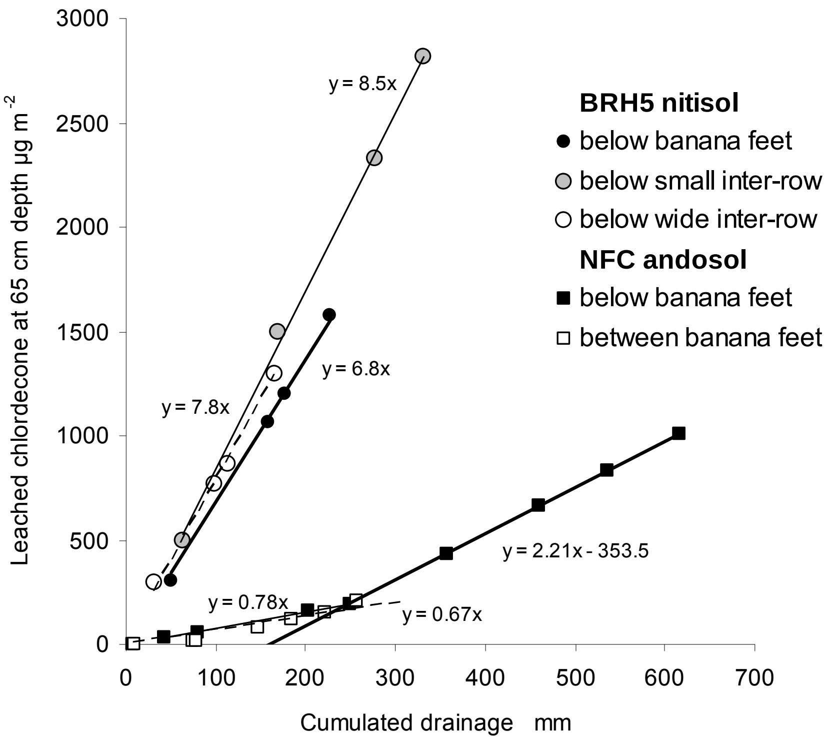


**Figure 7: Cumulated chlordecone collected by wick lysimeters, at 65 cm depth, during two drainage periods at two sites. The slopes of the linear regression represent the average CLD concentrations of water, in µg L$^{-1}$.**
a: below NFC andosol, $z_{til}$ = 0.32 m, from 08 September to 14 October 2003, showing two phases/types of lixiviation. b: below BRH5 nitisol, from 06 to 13 June 2005, $z_{til}$ = 0.6 m, where lixiviation appeared to be higher and less dependent on the lysimeter position.

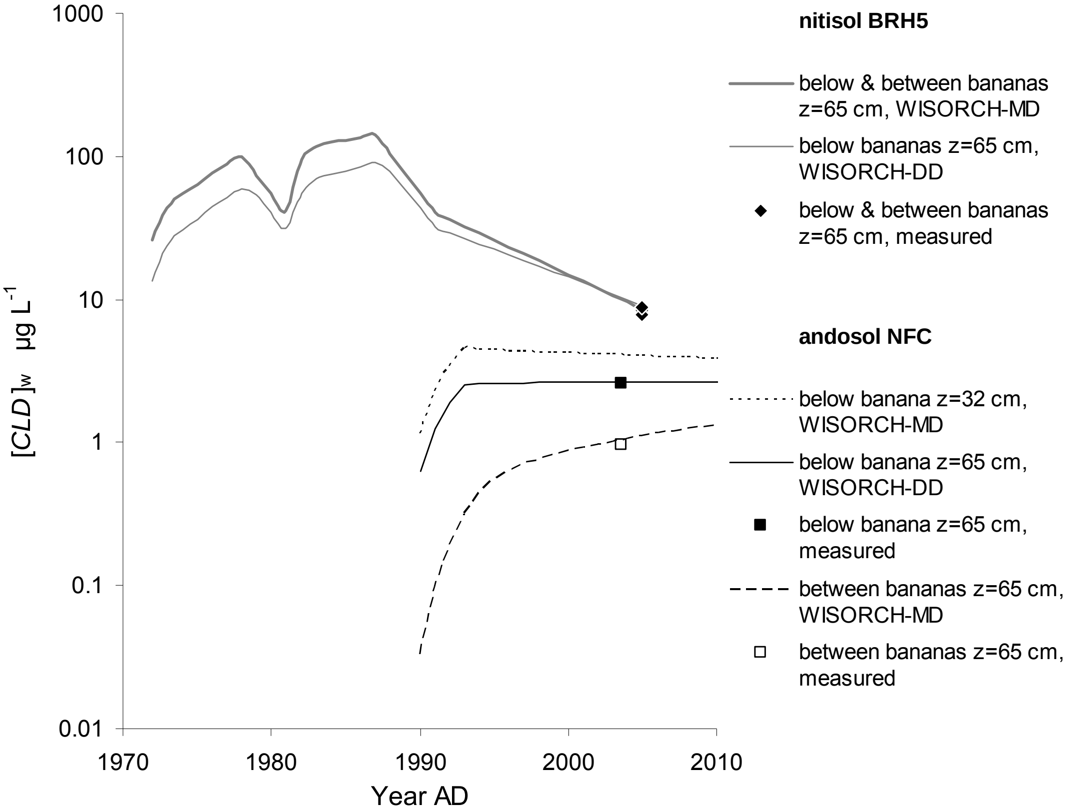


**Figure 8: Using WISORCH to calculate chlordecone concentration in drainage water [*CLD*]$_w$ according to two sub-models:**

- **DD is a dual drainage pattern applied below banana feet, which splits the drainage flow into (i) a bypass subflow, equal to the excess of drainage ($D_f$ -$D_b$) resulting from stem flow, whose CLD concentration derives from the lixiviation of the first layer, and crossing the second layer without interaction, and (ii) a matrix subflow, equal to the normal drainage flow $D_b$, whose CLD concentration results from the sorption/desorption balance in the second layer.**
- **MD is an ordinary matrix drainage applied to the bare soil between banana feet, where the stemflow does not increase the drainage.**

Both DD and MD drainage patterns were applied with the apparent $K_{oc}^*$ of each soil, 2.7 m$^3$ kg$^{-1}$ for BRH5 nitisol and 19.8 m$^3$ kg$^{-1}$ for NFC andosol.

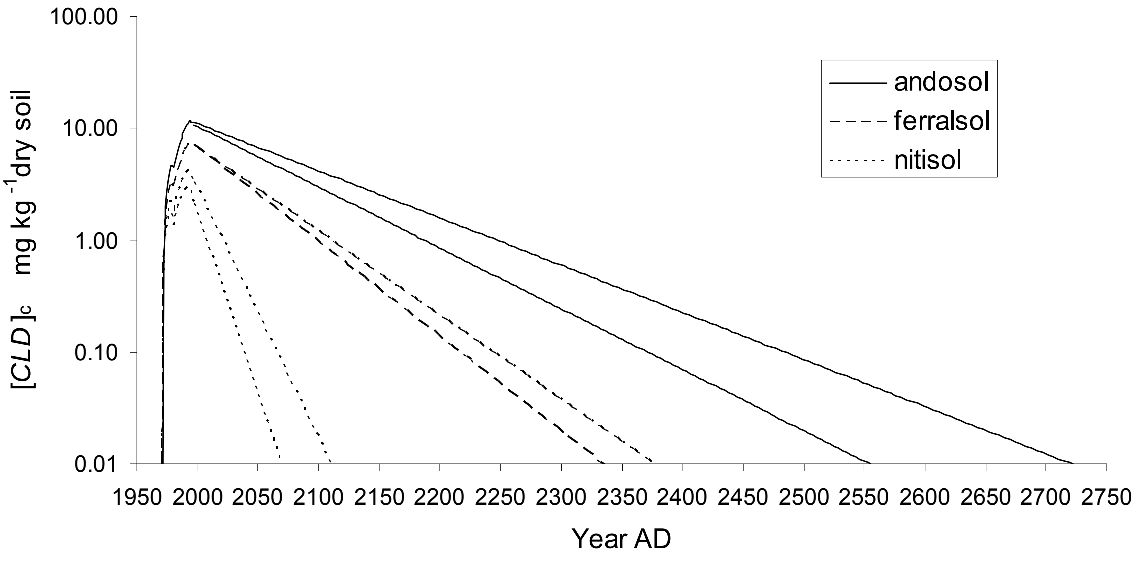


**Figure 9: Extrapolating chlordecone content of soils $[CLD]_c$ using the WISORCH model, after spreading during the whole period of chlordecone availability (1972-78 and 1982-93).**

For each soil type the two curves delimit the confidence interval at $P = 0.05$. Continuous lines represent andosol ($SOC = 48$ g kg$^{-1}$, $R = 4$ m y$^{-1}$), dashed lines ferralsol ($SOC = 21$ g kg$^{-1}$, $R = 3$ m y$^{-1}$), and dotted lines nitisol ($SOC = 19$ g kg$^{-1}$, $R = 2.5$ m y$^{-1}$). All soil types are considered to have been tilled at 0.6 m depth.

Tables

**Table 1**: **Distribution pattern of soils derived from andesitic pyroclast weathering in the French West Indies** (summarizing Colmet-Daage and Lagache, 1965). Italic characters indicate soil types sampled. PET is the potential evapo-transpiration. $K_{sat}$ is the hydraulic conductivity measured at saturation with a Müntz double ring.

| | **Soil type (FAO-WRB, 1998)** | **Secondary « clay » minerals** | **Clay %** | **$pH_{H2O\ 1/5}$** | **$K_{sat}$** mm $h^{-1}$ |
|---|---|---|---|---|---|
| **Young soils ($10^3$-$10^4$ years)** Contain primary sandy minerals (feldspars, amphibole, pyroxene) | | | | | |
| Rainfall < PET ≈ 1.6 m $year^{-1}$ | Vertic Cambisol | smectite | 60-80 | >6 | 1-10 |
| PET < Rainfall < 2 PET | *Nitisol* | halloysite | 60-80 | 5.5-6 | 10-50 |
| Rainfall > 2 PET | *Andosol* | allophane | 60-80 | 5-5.5 | 50-200 |
| **Old soils ($10^5$-$10^6$ years)** Primary weathering completed | | | | | |
| Rainfall < PET ≈ 1.6 m $year^{-1}$ | Vertisol | smectite | > 90 | >6 | <1-10 |
| PET < Rainfall < 2 PET | *Ferralic Nitisol* | halloysite + ε Fe-Al- oxihydroxides | > 90 | 5.5-6 | 10-50 |
| Rainfall > 2 PET | *Ferralsol* | halloysite + Fe- & Al- oxihydroxides | > 90 | <5.5 | 50-150 |
| **Fluvial terraces** | | | | | |
| | *Fluvisol* | halloysite + ε Fe- & Al- oxihydroxides | 50-90 | 5.5-6 | 10-50 |

**Table 2: Input variables of the chlordecone leaching model WISORCH, measured $[CLD]_m$ and calculated $[CLD]_c$ soil chlordecone contents at the sampling year (*SPL*) of each parcel whose former chlordecone spreading had been established.**

Soil types of Guadeloupe and Martinique (specified) are named according to the World Reference Base for Soil Classification (FAO, 1998). The number of subsamples pooled into composite 0-30 cm samples for analysis was >= 9. ***SOC*** was the soil organic carbon content, and $S_{soc}$ the corresponding storage. ***R*** was the average annual rainfall amount and $z_{til}$ the observed maximal tillage depth. **$[CLD]_m$** was the corrected value of soil CLD content, i.e. the measured one, divided by the average recovery coefficient of standard additions. $K_{oc}$ was taken as 17.5 $m^3\ kg^{-1}$ to calculate **$[CLD]_c$** by WISORCH.

| Soil type | Chlordecone spreading | | *SPL* | *SOC* | Bulk density | $S_{soc}$ | *R* | $z_{til}$ | $[CLD]_m$ | $[CLD]_c$ |
|---|---|---|---|---|---|---|---|---|---|---|
| / Parcel | Period (19..) | kg $year^{-1}$ | year | g $kg^{-1}$ | kg $dm^{-3}$ | Mg $ha^{-1}$ | mm $year^{-1}$ | m | mg $kg^{-1}$ | mg $kg^{-1}$ |
| **Andosol** | | | | | | | | | | |
| LGT | never | 0 | 2004 | | | | | 0 | 0 | |
| MTB | never | 0 | 2001 | | | | | 0 | 0 | |
| NDL | 71 | 3 | 2001 | 67 | 0.61 | 123 | 4720 | 0 | 0.80 | 0.96 |
| BAL | 87-88 | 3 | 2001 | 81 | 0.55 | 134 | 3910 | 0 | 2.82 | 2.99 |
| MOS | 88-90 | 3 | 2002 | 50 | 0.69 | 104 | 4170 | 0 | 4.33 | 4.50 |
| LGR | 72-76&½ 82-87 | 3 | 2001 | 61 | 0.64 | 117 | 4720 | 0 | 7.96 | 7.62 |
| NRN | 84-93 | 3 | 2003 | 50 | 0.69 | 104 | 4170 | 0 | 10.4 | 10.7 |
| PMX | 82-93 | 6 | 2001 | 85 | 0.54 | 138 | 4120 | 0 | 37.4 | 36.8 |
| TIL | 87-93 | 3 | 2001 | 80 | 0.56 | 134 | 4330 | 0 | 21.5 | 20.9 |
| NEY | 71-79 | 3 | 2005 | 67 | 0.61 | 123 | 3530 | 0 | 8.90 | 11.5 |
| DUF | 72-78 & 82-93 | 3 | 2003 | 47 | 0.71 | 100 | 4700 | 0 | 11.1 | 10.4 |
| BUR | 72-78 & 82-93 | 3 | 2003 | 136 | 0.4 | 163 | *7700*[a] | 0 | 28.8 | 28.7 |
| NRZ2 | 92-93 | 3 | 2003 | 67 | 0.61 | 123 | 3650 | 0 | 2.82 | 2.80 |
| NFC | 90-93 | 3 | 2003 | 67 | 0.61 | 123 | 3540 | 0.32 | 5.41 | 5.32 |
| NRZ1 | 92-93 | 3 | 2002 | 88 | 0.5 | 132 | 3650 | 0.55 | 1.81 | 1.82 |
| BBN | 82-93 | 3 | 2004 | 62 | 0.63 | 117 | 3070 | 0.6 | 9.06 | 8.79 |
| DMY1 | 82-93 | 3 | 2005 | 35 | 0.79 | 83 | 3300 | 0.6 | 6.60 | 6.32 |
| ACP12 | 71-78&83-93 | 4.5 | 2004 | 42 | 0.74 | 93 | 4100 | 0.6 | 13.8 | 12.3 |
| **Andic nitisol** | | | | | | | | | | |
| NNS1 | 85-93 | 3 | 2001 | 44 | 0.73 | 96 | 3600 | 0.45 | 7.29 | 7.59 |
| NNS2 | 85-93 | 3 | 2001 | 31 | 0.83 | 77 | 3600 | 0.5 | 4.84 | 5.11 |
| **Skeletic Andosol[b](Martinique)** | | | | | | | | | | |
| ACP3 | 92 | 1.5 | 2004 | 13 | 1.01 | 39 | 4210 | 0.6 | 0.19 | 0.26 |
| *ACP7* | 71-75 | 3 | 2004 | 45 | 0.73 | 97 | 4220 | 0.6 | 0.74 | 1.70 |
| ACP8 | 71-75 | 3 | 2004 | 55 | 0.67 | 111 | 4200 | 0.6 | 1.88 | 2.27 |
| ACP9 | 82-90 | 3 | 2004 | 13 | 1.00 | 40 | 3550 | 0.6 | 1.76 | 2.50 |
| ACP10 | 82-90 | 3 | 2004 | 12 | 1.02 | 36 | 3550 | 0.6 | 1.53 | 1.90 |
| ACPM05 | 71-93 | 3 | 2005 | 19 | 0.93 | 54 | 3450 | 0.6 | 5.18 | 4.58 |
| ACP11 | 72-78&82-94 | 3 | 2004 | 25 | 0.92 | 58 | 4000 | 0.6 | 5.18 | 6.28 |
| **Ferralsol** | | | | | | | | | | |
| *RCL1*[c] | 76 & 83 | 3 | 2005 | 16 | 0.96 | 48 | 3200 | 0.5 | 0.35 | 0.73 |
| *RCL2* | 76 & 83 | 3 | 2001 | 17 | 0.98 | 47 | 3200 | 0.5 | 0.54 | 0.68 |
| *REL* | 82-93 | 3 | 2004 | 23 | 0.9 | 61 | 3500 | 0.45 | 4.35 | 6.10 |
| MDG | 86-91 | 3 | 2005 | 29 | 1 | 88 | 3480 | 0.4 | 3.18 | 4.05 |
| FNT | 72-93 | 3 | 2005 | 21 | 0.92 | 58 | 3590 | 0.6 | 3.33 | 7.00 |
| *DLD2* | 82-89 | 3 | 2005 | 21 | 0.92 | 57 | 2990 | 0.6 | 2.35 | 3.00 |
| MTG 3 | 82-87 | 3 | 2005 | 22 | 0.91 | 60 | 2800 | 0.5 | 2.24 | 2.50 |
| **Ferralic fluvisol** | | | | | | | | | | |
| VRN | 72-93 | 3 | 2005 | 32 | 0.82 | 78 | 3500 | 0.51 | 1.88 | 9.20 |
| MTG2 | 82-87 | 3 | 2005 | 21 | 1 | 62 | 2800 | 0.5 | 0.33 | 2.67 |
| MTG1 | 82-87 | 3 | 2005 | 22 | 0.93 | 62 | 2800 | 0.55 | 0.20 | 2.00 |
| *DLD1* | 82-89 | 3 | 2003 | 21 | 1 | 63 | 2950 | 0.6 | 0.71 | 2.83 |
| *DLD3* | 82-89 | 3 | 2005 | 21 | 1 | 63 | 2960 | 0.6 | 0.56 | 2.93 |
| **Ferralic nitisol** | | | | | | | | | | |
| DMY2 | 82-93 | 3 | 2005 | 13 | 1.05 | 40 | 2700 | 0.6 | 2.00 | 4.85 |
| **Nitisol (Martinique)** | | | | | | | | | | |
| BRH2 | 72-78&82-85&94 | 3 | 2004 | 19 | 0.9 | 52 | 2510 | 0.5 | 0.71 | 5.67 |
| *BRH3* | 83-93 | 3 | 2004 | 19 | 0.9 | 52 | 2520 | 0.7 | 0.68 | 3.94 |
| BRH5 | 72-79&82-87 | 3 | 2004 | 15 | 0.95 | 40 | 2520 | 0.6 | 0.31 | 3.78 |
| BRH6 | 72-79&82-87 | 3 | 2004 | 19 | 0.9 | 52 | 2520 | 0.6 | 0.71 | 5.00 |
| BRH7 | 72-79&82-94 | 3 | 2004 | 19 | 0.9 | 51 | 2500 | 0.6 | 0.95 | 7.20 |

[a] *corrected from gravel and stone content*

[b] ***$[CLD]_m$** and **SOC** were analyzed on the whole sample, without gravel sieving*

[c] *italic codes indicate parcels which were not continuously cropped with banana since CLD applications, and had grass fallow at sampling dates*

**Table 3: Ratio between calculated $[CLD]_c$ and measured $[CLD]_m$ chlordecone contents using $K_{oc}$=17.5 m$^3$ kg$^{-1}$, and apparent soil organic carbon / water sharing coefficient of chlordecone ($K_{oc}^*$) calculated by inverting the WISORCH model for each soil type.**

*n* is the number of sampled parcels. The average $K_{oc}^*$ values associated with the same exponent letter are not significantly different at *P*=0.05 according to a Newman-Keuls test; max and min are respectively the maximum and the minimum of the confidence interval (*P*=0.05) of average $K_{oc}^*$.

| Soil type | Tillage > 0.3 m | *n* | $[CLD]_c$ / $[CLD]_m$ slope of linear regressions of fig.5 | $R^2$ | Calculated $K_{oc}^*$ m$^3$ kg$^{-1}$ Average | | max | min |
|---|---|---|---|---|---|---|---|---|
| andosol | + | 5 | 0.95 | 1.00 | **20.1** | a | 24.4 | 15.8 |
| andosol | - | 11 | 1.01 | 0.99 | **17.9** | ab | 23.9 | 11.9 |
| skeletic andosol | + | 7 | 1.11 | 0.94 | **14.4** | ab | 16.6 | 12.3 |
| andic nitisol | -/+ | 2 | 1.05 | 0.99 | **13.8** | b | 14.2 | 13.5 |
| ferralsol | + | 7 | 1.50 | 0.84 | **9.3** | c | 11.9 | 6.7 |
| ferralic nitisol | + | 1 | 2.41 | - | **3.6** | | - | - |
| ferralic fluvisol | + | 5 | 4.94 | 0.93 | **2.8** | d | 4.6 | 1.0 |
| nitisol | + | 5 | 7.92 | 0.65 | **2.5** | d | 2.8 | 2.2 |