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Summary

In Guadeloupe, the use between 1972 and 1993 of chlordecone, an organochlorine insecticide, has permanently contaminated the island's soil, thus contaminating the food chain at its very beginning. There is today a strong societal requirement for an improved mapping of the contaminated zones. Given the extent of the areas to be covered, carrying out soil tests on each plot of the territory would be a long and expensive process. In this article, we explore a method of demarcating polluted areas. The approach adopted consists in carrying out, using surface water analyses, a hydrological delimitation that makes it possible to distinguish contaminated watersheds from uncontaminated ones. The selection of sampling points was based on the spatial analysis of the actual and potential contamination data existing at the beginning of the study. The approach was validated by soil analyses, after having compared the contamination data of the watersheds with the soil contamination data of the plots within them. The study thus made it possible to highlight new contaminated areas and also those at risk of contamination, and to identify the plots to be targeted as a priority during future analysis campaigns by State services.

Keywords

Organochlorine pesticide, Watersheds, Contamination, Water, Soil

1. Introduction

The health safety of food produced by agriculture is a major societal concern. Agricultural products can become contaminated by pesticides, either by their direct application to crops or indirectly by their transfer from the soil, to which they had become fixed after application. The latter is especially the case with persistent molecules which were used in the past and still persist in the environment (Barron et al. 2017; Wu and Zhu 2019). When originating from old practices, this contamination is hard to predict, making it difficult to control the risk of exposure of human populations. An obvious prerequisite is the identification and mapping of contaminated areas.

This problem is particularly relevant to the French West Indies where an organochlorine insecticide, chlordecone, was used between 1972 and 1993 on banana crops to control the banana weevil (*Cosmopolites sordidus*). This persistent pesticide is still present in soils (Cattan et al. 2016) and continues to contaminate crops grown there, rendering them unfit for consumption (Cabidoche and Lesueur-Jannoyer 2012; Clostre et al. 2015). Following the discovery of this health problem, maps of soil contamination were drawn up. In Guadeloupe, a first mapping of plots likely to be contaminated was carried out in 2006 (Tillieut 2006) and was based on a classification of areas according to their banana cultivation history. Since chlordecone was used mainly in banana plantations, and in a quasi-systematic manner (Levillain et al. 2012), this hypothesis was based on the assumption that the historical use of the land for banana cultivation should account for the level of soil contamination.

However, historical information on land use was not comprehensive or accurate (unregistered or unmapped banana plots; diverted use of chlordecone on other crops, etc.). There was therefore uncertainty about the state of contamination of the plots: uncontaminated plots could be located in areas identified as being at risk (based on land use history), and, more importantly, contaminated plots could be located in areas identified as being at zero to negligible risk. To reduce these uncertainties, many soil analyses have been undertaken in the territory by different entities (Department of Food, Agriculture and Forestry; Chamber of Agriculture; Regional Health Agency; National Institute for Agronomic Research). The results were collected in 2009 in a spatialized database within a geographical information system (GIS) for an effective mapping of contamination of plots. However, these analyses were not based on systematic campaigns to examine the environment, which were too expensive to undertake. The plots that were analysed were limited mainly to within areas defined as being at risk and thus did not cover the entire territory in a systematic manner. In fact, there is

today a strong need to undertake a new mapping to better determine the areas that are contaminated by chlordecone, and also those that are not, so that their use for agricultural purposes can be allowed.

To draw up contamination maps, it is customary to conduct soil analyses (e.g. (Zhao et al. 2013)), and then, to keep costs down, use spatial interpolation between them to represent potentially polluted areas (e.g. (Frangi and Richard 1997)). However, in the case of chlordecone, soil concentrations are dependent on farmer categories and are not spatially structured (highly contaminated plots may be adjacent to uncontaminated plots), making spatial interpolation unsuitable for this molecule (Levillain et al. 2012). However, other methods can be envisaged based on chlordecone's intrinsic characteristics: it is particularly stable and strongly sequestered in soil organic matter (Cabidoche et al. 2009; Woignier et al. 2013). Although it is not very mobile, some of it is leachable by water drainage, in more or less significant quantities depending on soil type. The knowledge acquired on the transfer of this molecule (Cabidoche et al. 2009; Crabit et al. 2016; Della Rossa et al. 2017) shows that permanent contamination of watercourses can be linked to the state of soil contamination at the watershed scale. On this basis, we can hypothesize that if river water is contaminated, some of the land in its watershed is contaminated. Conversely, the absence of water contamination at the point of sampling should imply that no or very low soil contamination of the associated watershed. So it can be surmised that water analyses can account for the state of contamination of the lands of the watershed associated with the sampling point.

The purpose of this paper is to explore a method to improve chlordecone pollution maps based on this hypothesis. The case of Guadeloupe was selected for the study. The approach adopted consisted of analysing, for different watersheds, both the contamination of water and of the soil. To this end, we carried out a hydrological division of the Guadeloupean territory into watersheds and sub-watersheds, making it possible to distinguish, by means of water analyses at the outlet of each of them, those contaminated by chlordecone from those that are not. This method makes it possible to better manage the health risk associated with the use of the soil for cultivation.

2. Materials and methods

2.1 Study site

Guadeloupe (16°15'N, 61°32'W) is an archipelago located on the arc of the Lesser Antilles, which marks the boundary between the Atlantic Ocean and the Caribbean Sea (Figure 1). The study focuses on 'continental' Guadeloupe, which is composed of two main islands: Basse-Terre, a mountainous volcanic island with an area of 848 km², which rises to an altitude of 1467 m; and Grande-Terre, limestone plateau of 587 km², whose altitude does not exceed 136 m.

The study site is subject to an oceanic tropical climate, regulated by a steady flow of hot and humid trade winds. The year is divided into two clearly distinct seasons: a so-called dry season (*carême*) from December to May and a wet

season (*hivernage*) from June to November, which is marked by the occurrence of tropical waves. Rainfall shows great variation over the territory, with annual cumulative amounts ranging from 950 mm on the east of Grande-Terre and on the west coast of Basse-Terre, to more than 10,000 mm at the highest altitudes of Basse-Terre. The hydrographic networks of the two islands are also very different. In Basse-Terre, where rainfall is high, the rivers are perennial, sustained during periods of low precipitation by groundwater (Charlier et al. 2011; Crabit et al. 2016). In Grande-Terre, the absence of marked relief and the much lower rainfall make surface flows much more limited and very irregular, giving rise to numerous phenomena of endorheism (when the surface flows do not reach the sea and get lost in closed depressions) (Morell and Jérémie 1994).

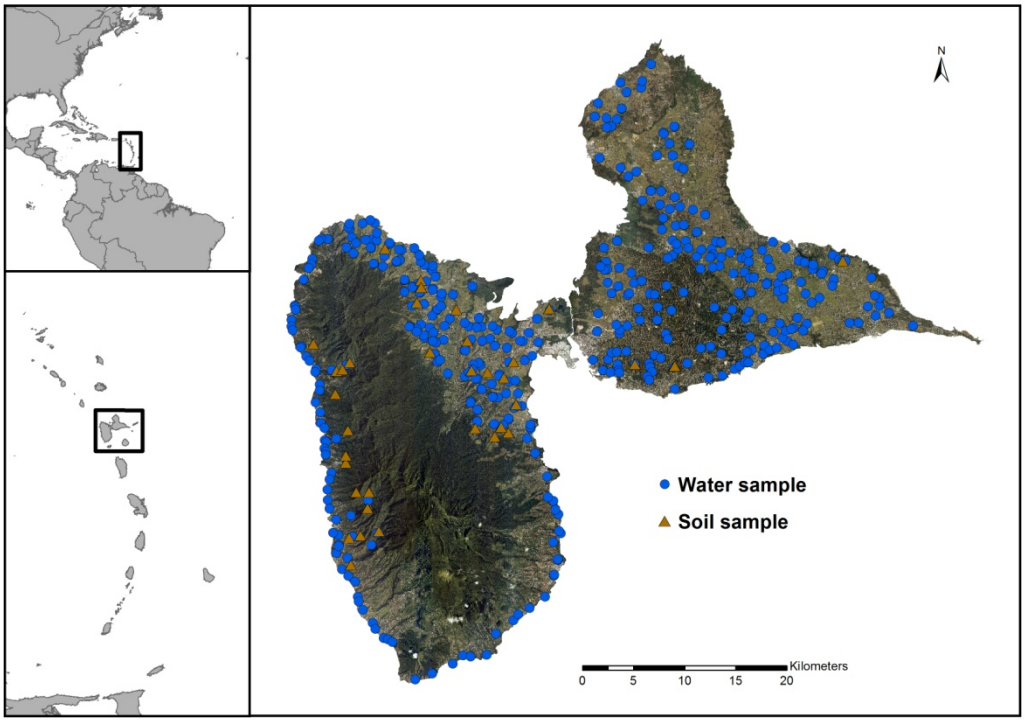

Figure 1 Geographical location of Guadeloupe and water and soil sampling points

2.2 Approach

The approach we adopted was to link the water contamination at the outlet of a watershed with that of its soils. On the basis of the spatial analysis of the actual and potential contamination data existing at the beginning of the study, we carried out a hydrological delimitation of the territory into watersheds and sub-watersheds to distinguish, by means of water analyses at the outlet of each of them, those that are contaminated by chlordecone from those that are not. If the water is contaminated, then the watershed is considered to be at high risk and will require soil tests before plots can be used for cultivation and increased monitoring of the agricultural production originating from the watershed. If the water is not contaminated, the watershed is considered low-risk and will not require such increased monitoring. In this approach, we did not take into account the soil type. In fact, although the soil type is obviously a determinant of the

level of river contamination, we considered that the fact that the soils were contaminated, regardless of their type, was sufficient to explain the binary state of the river: to be or not to be contaminated.

The method was validated by comparing the results of the water analyses with the initial data of soil contamination of watersheds: risk map drawn up in 2006 on the basis of historical land use (Tillieut 2006) and GIS database of chlordecone soil analyses. For the watersheds for which there were inconsistencies, new soil analyses were carried out on some plots to:

- verify the contamination of plots previously analysed for chlordecone;
- verify the contamination – or not – of plots identified as at risk;
- if no information was available, find a terrestrial origin for the detected water contamination.

2.3 Delimitation of watersheds

Watersheds and their drainage systems have been previously defined, delimited and generated through spatial analysis tools under Esri ArcMap©. We used the RGE ALTI® digital elevation model (DEM) from the French National Institute of Geographic and Forest Information (IGN), which covers the whole of continental Guadeloupe at a resolution of 1 m. However, this DEM had to be modified because the elevated elements of the road network were not open, i.e., they formed barriers to water flow. It was therefore decided to resample it at a resolution of 25 m to minimize this problem. We then used the AGREE method (Hellweger 1997) to force the DEM by IGN's BD TOPO® hydrographic network, considered as a reference, by verifying 1) the correspondence between the generated hydrographic network and that of the reference network, 2) the coherence between the generated watersheds and the reference hydrographic network, and 3) the non-modification/conservation of ridges, by comparing the watersheds generated by the forcing with those that did not undergo forcing.

2.4 Water sampling

2.4.1 Water sampling plan

The water sampling points were located at the outlets of watersheds with a minimum area of 50 ha, which represented a good compromise between the desire to cover the territory as widely as possible and the budgetary constraints which allowed only a limited number of water analyses. In cases in which the outlets were not accessible (mangroves, private access, steep relief, etc.), the sampling points were moved upstream.

Additional sampling points were then distributed over certain watersheds to obtain information on the state of soil contamination at the sub-watershed scale and therefore to be able to distinguish contaminated sub-watersheds from uncontaminated ones, especially in areas that have not been extensively studied so far. These sampling points were

selected on the basis of the spatial analysis of actual and potential contamination data existing at the beginning of the study (chlordecone soil contamination risk map and spatialized water and soil contamination data for Guadeloupe).

In Basse-Terre, in watersheds with an available agricultural area (AAA – (SIRS 2015)) greater than 25 ha, a size we considered sufficiently big to warrant further analysis, it was possible to identify sub-watersheds bigger than 50 ha that did not have known chlordecone contamination. Sampling points were identified along their watercourses at the frequency of one point every 1.2 km or so. This allowed us to obtain a grid sufficiently dense to identify possible contamination as well as to follow its evolution along the watershed's hydrological slope, i.e., upstream to downstream. For watersheds with an AAA of less than 25 ha, it was considered that the analysis conducted at their outlet was sufficient to assess their chlordecone contamination.

In Grande-Terre, flows are not perennial and chlordecone contamination data were sparse. As a result, we mapped a grid across the territory, but this time with the objective of obtaining information on the contamination of areas that are hydrologically disconnected from one another for most of the year. Samples were thus planned on the flow axes of all the sub-watersheds with an area greater than 50 ha, with an average sampling frequency of 1.2 km. Since flows are rare, the majority of the sampling points were in ponds/reservoirs along the flow axes.

On the basis of this sampling plan, 219 different points were sampled in Basse-Terre, spread over 97 watersheds emptying into the sea and representing 83% of the island's surface area. In Grande-Terre, samples were collected from 198 different points, spread over 33 watersheds emptying into the sea and representing 71% of the island's surface area. All the water sampling points are shown in Figure 1.

2.4.2 Periods of water sampling

Water samples were collected during different hydrological periods depending on the sampling location.

In Basse-Terre, previous measurements, made on different soil types, have shown that runoff is far less significant than infiltration. Indeed, runoff rates are only 5 to 10% for low-intensity rainfall, and rise to a maximum of 30% for intense events (Cattan et al. 2006). Surface runoff is therefore not very effective in transporting chlordecone under the conditions prevailing in the French West Indies (Cabidoche et al. 2009). Contamination of rivers is mainly via groundwater, which provides flows during low-water periods (Charlier et al. 2009; Crabit et al. 2016). This period also represents the most stable state of the hydrological system. For Basse-Terre, water sampling was therefore scheduled during low-water periods.

In Grande-Terre, too, infiltration is much more significant than surface runoff, with the latter (estimated at 1% of annual rainfall) being almost negligible (Cottez 1972). Moreover, since rainfall is much lower than in Basse-Terre, the flows in the ravines are very limited and very irregular, giving rise to a number of endorheism phenomena. Surface water is

therefore not present throughout the year, and depends heavily on rainfall during the rainy season. For Grande-Terre, the samples were therefore planned during the rainy season.

As far as possible, samples from each watershed were made on the same day and/or in similar hydrological conditions. If a stream was dry on the day planned for the sampling, the sample was rescheduled, if possible, after a rain event sufficient to cause runoff.

2.4.3 Water sampling method

The water samples were collected in 1 litre brown-coloured glass bottles. On return from the field, these bottles were stored in a refrigerator at 4 °C before being shipped in refrigerated coolers the next day to metropolitan France for analysis. The shipments were made through an express delivery service (Chronopost) and took 2 to 3 days to reach the laboratory.

2.5 Soil sampling

2.5.1 Soil sampling plan

A typology of all the watersheds was carried out according to the contamination data available for each of them, namely: (1) the state of contamination of the river waters at the watershed's outlet, according to the analyses performed (WC); (2) the presence of contaminated soils in the watersheds, according to the GIS database of already existing soil tests (SC); (3) the presence of plots at risk, according to the risk map drawn up in 2006 (PC). One of two values was assigned to each of these three variables (WC, SC and PC): uncontaminated = 0; contaminated = 1. Four watersheds with plots known to be contaminated (SC 1) presented a particular context, with contaminated water but with only a few number of contaminated plots, of small areas or which were far from the main flow axes, i.e. contributing not much to water pollution. These watersheds could therefore contain other contaminated plots not referenced in the GIS database. We then created a new type denoted $SC1^{-}$ for these 4 watersheds. Soil analyses were then conducted on watersheds with inconsistencies: (case 1) presence of contaminated soils or plots at risk of contamination while the rivers were not contaminated, (case 2) absence of contaminated soils or plots at risk of contamination while the rivers are contaminated, and (case 3) presence of contaminated rivers while the contribution of actual contaminated plots to water pollution is low.

The decision rules concerning the plots to be targeted for soil analyses were thus formalized as follows:

- for watersheds of type [WC 0, SC 1, PC 0] and [WC 0, SC 1, PC 1] (case 1): verify the contamination of plots indicated as contaminated. For Grande-Terre, only plots located in the immediate vicinity of a water sampling point were considered, as it was difficult to define the catchment area of a given water sampling point with accuracy (for reasons already mentioned above);

- for watersheds of type [WC 1, SC 1$^-$, PC 1] (case 3), [WC 1, SC 0, PC 1] (case 2) and [WC 0, SC 0, PC 1] (case 1): verify the contamination status of plots said to be at risk;
- for watersheds of type [WC 1, SC 1$^-$, PC 0] (case 3) and [WC 1, SC 0, PC 0] (case 2): locate an as yet unknown terrestrial contribution to the water contamination. In this case, the plots to be analysed were selected from the analysis of old aerial photos and historical land use maps (analysis of the documents used to produce the 2006 risk map, as well as two additional maps), and were then validated by field investigations.

In Basse-Terre, of the 219 watersheds analysed through water analyses, 60 presented inconsistencies. For Grande-Terre, only 3 watersheds out of the 199 analysed presented inconsistencies. Because of cost considerations, additional analyses were conducted on only 36 plots: 32 on Basse-Terre and 4 on Grande-Terre (Figure 1). The targeted watersheds were primarily those that each had an unknown source of contamination and those that had only one doubtful plot to verify. Watersheds with several doubtful plots were also analysed. In these, the suspect plot that was closest to the river and was easiest to access was selected for analysis.

2.5.2 Soil sampling method

Chlordecone exhibits strong spatial heterogeneity at the plot scale (Clostre et al. 2014). The scientific literature recommends 20 sampling points per plot for soil analyses [21]. However, this high number of samples is difficult to implement on a large number of plots. For our study, we decided on a density of 15 samples per hectare, with a minimum of 8 samples for plots with areas less than 1 ha. These sampling points were located on transects on each plot, spaced 10 to 30 m apart and at least 7 m distant from the plot's boundaries to limit edge effects. The samples were collected only from the 0-30 cm horizon, to be in line with the existing reference base.

Samples from the same plot were collected in a thick plastic bag, which was closed once filled. The auger was cleaned with brush and water after sampling each plot. On return from the field, each sample was manually homogenised after removal of extraneous material and reduction of aggregates. Then, a sub-sample of 500 g was formed by quartering method. It was placed in a polypropylene sampling pot. The samples were stored in the refrigerator at 4 °C before shipment to metropolitan France at the beginning of each week for analysis. The shipments were made through an express delivery service (Chronopost) and took 2 to 3 days to reach the laboratory.

2.6 Analyses of samples

The water and soil samples were analysed by the La Drôme Departmental Laboratory (LDA 26).

For the analysis of chlordecone in water, 1 L of sample was extracted in liquid/liquid partition with a dichloromethane/ethyl acetate mixture (80/20) at several pH values. Extraction tracers HBB/TPP/DIA-DS where added prior to extraction. The organic phases were collected and frozen at -18 °C to remove traces of water. The extract was

then concentrated under nitrogen flow in a 35 °C bath (Turbo-Vap) and a volume tracer, propazine D6, was added. A drop of pentanol was brought to the extract and the solvent was evaporated. 0.5 mL of the extract were introduced in a water/acetonitrile mixture (50/50) and internal standards, 24-D D3 and atrazine D5, were then added. The analysis was performed by HPLC-MS/MS, with an analytical uncertainty of 30% and detection and quantification thresholds of 0.01 µg/L.

For the analysis of chlordecone in soils, 10 g of sample were introduced into an extraction cell and tracers were added (HBB/TPP). Accelerated solvent extraction (ASE) was carried out with a dichloromethane/acetone mixture at 100°C and under pressure (120 bar). The resulting extract was concentrated with a vacuum centrifuge (GENEVAC EZ2) which greatly reduces the loss of volatile compounds. The extract was concentrated to 10 mL and an aliquot of 1 mL was taken. A drop of pentanol was added to the extract and the solvent was evaporated in a GENEVAC MiVac system to preserve the volatile compounds. The extract was then taken up by a mixture of acetonitrile and water with the Chlordecone C13 internal standard. The analysis was performed by HPLC-MS/MS, with an analytical uncertainty of 40%, a detection threshold of 2 µg/kg of dry soil and a quantification threshold of 5 µg/kg of dry soil.

3. Results

3.1 Contamination of watersheds

The level of water contamination by chlordecone was assessed against thresholds defined in French law by the decree of January 11, 2007, concerning the limits and references of quality of raw water and water intended for human consumption, in application of European Directives 75/440/EEC and 98/83/EC. The first threshold, of 0.1 µg/L, corresponds to the limit of the concentration of chlordecone that is allowed in tap water intended for human consumption. The second, of 2 µg/L, is the chlordecone concentration threshold above which raw water can no longer be made safe for drinking even after treatment.

3.1.1 Highly contaminated water in Basse-Terre

In Basse-Terre, chlordecone was found in 36% of 277 analyses, with concentrations ranging from 0.01 to 42.9 µg L^{-1} (Figure 2). 24.1% of rivers have levels above the French consumption threshold for this pesticide (0.1 µg L^{-1}), and 7.9% of them have water contamination above the higher threshold of 2 µg L^{-1}, i.e., water that is considered unfit for consumption even after treatment. Furthermore, of the 110 watersheds analysed at their outlets, 43 (39%) were identified as discharging contaminated water into the sea. In Grande-Terre, 5 points of contamination out of 198 were detected, each of them at low levels, lower than the consumption norm.

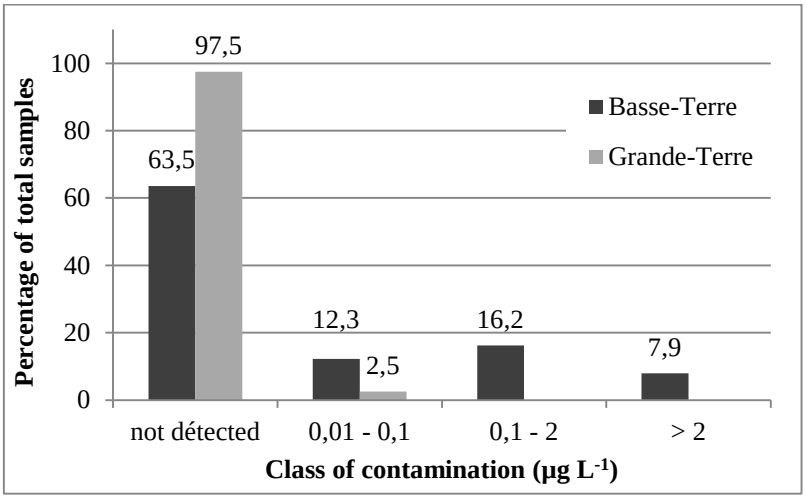

Figure 2 Distribution in different classes of contamination of the results of analyses of chlordecone in the waters of Basse-Terre and Grande-Terre

3.1.2 A contamination structured by banana cultivation

A map of watershed contamination by chlordecone was drawn up (Figure 3).

In Basse-Terre, no chlordecone contamination was detected in the watercourses in the northern half of the west coast or the northern part of the east coast, except in an upstream tributary of a small stream, where this contamination was measured at 0.011 µg L (slightly higher than the quantification threshold of 0.01 µg L^{-1}). In the centre of the east coast, contamination was recorded in various rivers and some of their tributaries, with concentrations ranging from 0.016 µg L^{-1} to 0.41 µg L^{-1}. Finally, in the island's southern part, where chlordecone had seen massive use, it was not possible to identify uncontaminated sub-watersheds belonging to contaminated watersheds.

In Grande-Terre, 5 points of contamination were recorded, with relatively low levels for each (close to the quantification threshold). Field investigations that were undertaken allowed 3 of these points to be eliminated since the source was not land contamination but a contaminated water supply (water of the agricultural irrigation network coming from Basse-Terre).

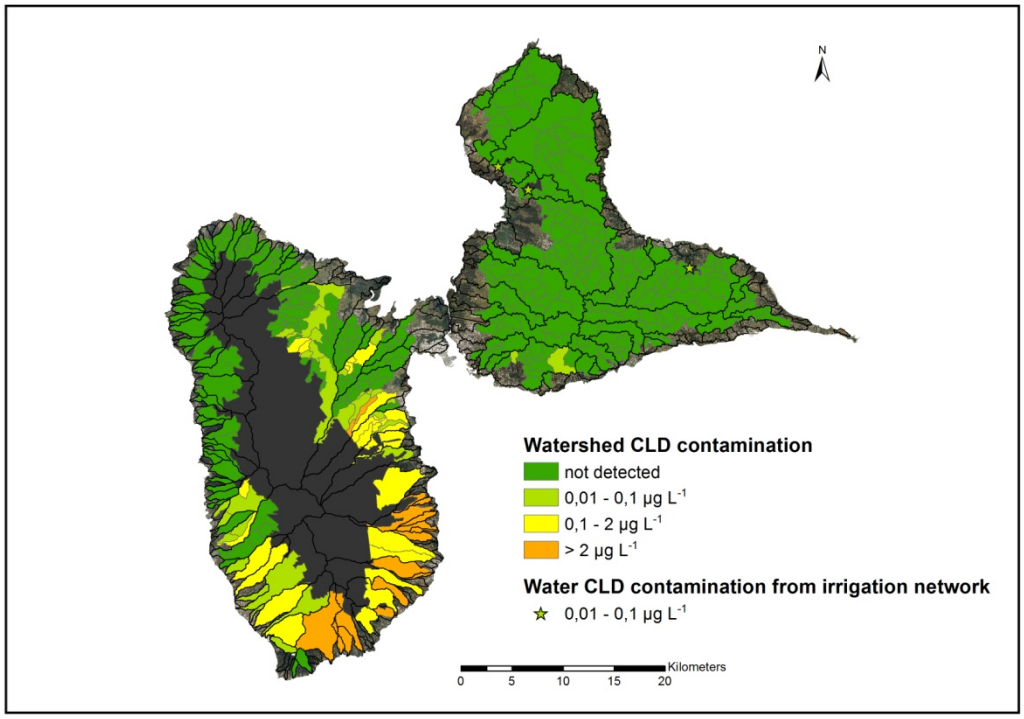

Figure 3 Mapping of chlordecone contamination of watersheds in continental Guadeloupe

3.2 Comparisons between chlordecone databases

These comparisons were made only for Basse-Terre, for which the three variables WC, SC and PC were available. Indeed, the risk of soil contamination by the chlordecone on Grande-Terre appears to range from nil to negligible on the entirety of its territory. In addition, as already noted, it is impossible to define the supply watershed of a given water sampling point with any accuracy, which makes it difficult to establish a correspondence between soil data and water data.

3.2.1 Soil analyses consistent with the plot contamination risk map

Table 1 shows that for 81% of the watersheds analysed in Basse-Terre there exists a correlation between the presence of contaminated soils and plots at risk of contamination (i.e. [PC 0, SC 0] and [PC 1, SC 1]). The absence of contaminated soils, while plots at risk are present in the watersheds ([PC 1, SC 0], 9% of cases), can be explained by the fact that no plot has yet been analysed in these watersheds. On the other hand, the presence of contaminated soils in watersheds without plots at risk of contamination [PC 0, SC 1] reflects either the lack of exhaustiveness of the historical analysis of the plots on which banana was cultivated, or shows the existence of historical uses of the pollutant diverted to plots other than those of banana, or is due to the uncertainty associated with the contamination data. In the end, the risk of having contaminated soils (SC 1) in watersheds without a plot at risk (PC 0) appears to be significant and amounts to 23/(120 + 23) = 16% (percentage calculated on the number of watersheds without a plot risk only).

Table 1 Number of watersheds with/without the presence of plots at risk of contamination and contaminated soils in Basse-Terre. The percentages are calculated on the total number of watersheds

	PC 0	PC 1
SC 0	120	19
	55%	9%
SC 1	23	57
	11%	26%

3.2.2 Correspondence between water contamination and soil contamination

Table 2 shows that for 80% of the watersheds in Basse-Terre, there is a correspondence between the existence or the absence of soil contamination and the existence or the absence of water contamination ([SC 0, WC 0] and [SC 1, WC 1]).

Table 2 Number of watersheds according to soil and water contamination status in Basse-Terre. The percentages are calculated on the total number of watersheds

	WC 0	WC 1
SC 0	124	15
	57%	7%
SC 1	29	51
	13%	23%

For the remaining 20%, 7% is due to a lack of information on soil contamination. In contrast, for the other 13%, contamination of plots is not detectable in watercourses. This problem reflects either the uncertainty related to the measurement of soil contamination, or a very large dilution effect of the soil contamination in the water feeding the river.

3.3 Verification of contamination of plots

3.3.1 Identification of watersheds with inconsistencies between WC, SC and PC

In Basse-Terre, of the 153 watersheds whose water was found to be uncontaminated, 39 (25%) contained plots that had to be verified by new soil analyses (Table 3a):

- 29 watersheds with plots known to be contaminated, to confirm that they were not ‘false positives’;
- 10 watersheds with plots at risk, to verify that these plots are uncontaminated.

Conversely, of the 66 watersheds whose water was found to be contaminated, 21 (32%) contained plots that had to be verified by soil analyses in order to locate a terrestrial origin to the water contamination (Table 3b):

- 15 watersheds with plots at risk, to check whether these plots contributed to the contamination found in the water;
- 6 watersheds without any indication of contamination and without risk, to try to locate a terrestrial origin for the contamination.

Table 3 Number of watersheds with inconsistencies when: a. no water contamination by chlordecone was detected; b. water contamination by chlordecone was detected

a.	Num. of watersheds	[SC 1, PC 0] [SC 1, PC 1]	[SC 0, PC 1]
WC 0	153	29	10
	100%	19%	7%

b.	Num. of watersheds	[SC 0, PC 1] [SC 1^-, PC 1]	[SC 0, PC 0] [SC 1^-, PC 0]
WC 1	66	15	6
	100%	23%	9%

In Grande-Terre, of the 197 watersheds whose water was found to be uncontaminated, only one contained a plot known to be contaminated immediately upstream from the water sampling point.
Conversely, in 2 watersheds whose waters were found to be contaminated, we carried out soil analyses in order to try to locate a terrestrial origin for the contamination found in the water.

3.3.2 Results of the soil analysis

In Basse-Terre, in the watersheds that did not show water contamination (WC 0), 24 soil analyses were carried out (Table 4a). Of the 14 plots known to be contaminated that were verified, 11 were found to be chlordecone free (79%). Similarly, of the 10 plots at risk of contamination that were analysed, 8 were found to be uncontaminated (80%).
In the watersheds whose waters were found to be contaminated (WC 1), 11 soil analyses were carried out (Table 4b). In 9 of the 11 watersheds analysed (82%), we managed to find terrestrial origins for the chlordecone contamination. Of the 9 plots at risk of contamination that were analysed, 7 were found to be contaminated (78%).

Table 4 Summary of the results of soil analyses carried out in Basse-Terre watersheds where: a. no water contamination by chlordecone was detected; b. water contamination by chlordecone was detected

a.	Type of plots analysed	Num. of plots analysed	Num. of negative analyses	% negatives
	Contaminated plots	14	11	79%
	Plots at risk	10	8	80%

1

b.	Type of plots analysed	Num. of plots analysed	Num. of positive analyses	% positives
	Plots at risk	9	7	78%
	Plots with unknown status	2	2	100%

2 In Grande-Terre, the only plot known to be contaminated that was re-analysed did not contain chlordecone. For
3 watersheds in which the terrestrial origin of water contamination was sought to be located, we could not identify, from
4 the documents of historical land use, any real leads allowing us to identify a possible source of pollution. 3 plots were
5 nevertheless analysed but no contamination was found. This reflects the difficulty, in some watersheds, of finding a
6 source of contamination without having any information beforehand as to where to start looking.

7 **3.3.3 Assessment of the approach**

8 Our approach consisted of characterizing the contamination of the soil of a watershed on the basis of the quality of its
9 watercourse. We assessed this approach on the basis of the correlation between water contamination data (WC) and soil
10 contamination data (SC) or soil contamination risk (PC).
11 In Basse-Terre, the approach proved to be robust in the sense that a watershed identified as uncontaminated by our
12 water analyses (WC 0) had a very good chance of not being contaminated. Indeed, 75% of these watersheds had no
13 contamination or known risk of contamination of their soils ([WC0, PC0, SC0]). In the remaining 25%, 80% of the
14 plots that were verified by soil analyses were actually found to be uncontaminated. Either these were plots at risk that
15 happened to be uncontaminated, or they were plots that were falsely identified as contaminated in the GIS database
16 (because of a methodological or technical error during the sampling, or an error when entering data). Given the time
17 elapsed between the old and new analyses (6 to 11 years), this cannot be due to the degradation process, unless a high
18 degradation rate is retained, which is not consistent with the current state of soil contamination in Guadeloupe. For the
19 other 20% of these plots, the limitation of the approach seems to be the cause and raises the question of the sources of
20 errors in the different methods of assessing contamination (see 4. Discussion). In watersheds identified as contaminated,
21 68% of water contaminations can be explained by pre-existing soil contamination data. For the remaining 32%, for
22 which there is therefore a lack of information, a terrestrial origin of the contamination could be identified in 82% of the
23 watersheds analysed. In Grande-Terre, our approach has shown certain limitations in view of the limited period during

which surface water is present and the low hydrological connectivity between the different points of a watershed (due to the geomorphology of Grande-Terre). However, contaminations could still be detected.

4. Discussion

Our goal was to define a method to determine the state of chlordecone contamination in areas where few analyses had been carried out. This method is based on the assumption that water quality at the outlet of a watershed reflects the state of contamination of its soils. On the whole, our results showed good agreement between the contamination of rivers and the contamination of soils in their watersheds. However, there are some limitations of this method.

The first pertains to the physical environment. Indeed, the hypothesis adopted assumes that the watershed corresponds to the hydrogeological basin supplying the river. However, this hypothesis can be challenged (Crabit et al. 2016), especially in the volcanic context of Basse-Terre where water flow paths are affected by successive eruptive episodes and where the transfer routes are mostly underground (Charlier et al. 2008). Thus a river may receive flows from adjacent watersheds or not receive all the flows from the plots within its watershed. These situations may explain the fact that contaminated plots may be present in watersheds that do not present any water contamination at their outlets.

The risk map was also found to be incomplete. This map is based on only 3 years of land use for banana cultivation (1969, 1985 and 1997) and only concerns plots that were still declared in 2003 as being used for agriculture (Tillieut 2006). It thus incorporates a large amount of uncertainty as to land use during the period of use of chlordecone in Guadeloupe. Furthermore, it cannot account for the diverted use of the chemical on other crops. Although these cases were, no doubt, limited (Levillain et al. 2012), they are nevertheless likely to cause water contamination. These observations lead to the conclusion that the risk map under-represents the state of contamination and explain why certain watersheds can present water contamination without having plots at risk within them (case [WC1, PC0] of table 3b). On the other hand, the results of the soil analysis show that an inverse bias exists: plots are classified at risk but are uncontaminated. This phenomenon can arise from the fact that the risks of contamination of plots used to cultivate banana in 1969 or 1997 were taken into account in the map, but they may not have been actually used to cultivate banana during the period of chlordecone use (1972-1993). It can also arise from the extreme variability of agricultural practices (Levillain et al. 2012; Clostre et al. 2014) from one plot to the next, and the difficulty of interpolating levels of practice to a set of plots. Another explanation is that although the molecule is stable, it is nevertheless carried away by the water and thus the contamination of the plots decreases over time (Cabidoche et al. 2009). Depending on rainfall conditions and soil types, some plots at risk may no longer be sources of contamination. Notice that today, the degradation process cannot be invoked to explain why some plots are no longer contaminated. Recent work (Chevallier et al. 2019) shows the value of a better understanding of chlordecone degradation conditions in order to understand the evolution of soil contamination in watersheds.

Consequently, the map of the contaminated soil raises questions regarding its reliability. The abnormally high rate of false positives found by comparing soil analyses guided by the map and our campaign of analysis highlights the problem of undertaking the analyses. There are many factors that can skew the results of an analysis: minimum number of samples per plot, improper cleaning of tools before collecting each sample, changes in analytical precision, etc.

Finally, the analytical bias can also concern water. To begin with and considering the consistency of river contamination by chlordecone (Crabit et al. 2016; Cattan et al. 2019), collecting a single river sample does not lead to any significant bias in the assessment of pollution: the permanence of contamination suggests that irrespective of the sampling period, the analysis will be positive. A negative sample, on the other hand, may be related to the absence of contamination or correspond to a very low level of contamination, around the detection threshold. Water contamination by the container (glass bottle) seems unlikely given the process of cleaning the flasks by the laboratory and the sampling protocol put in place by our team. Finally, the time taken to send samples to the laboratory, less than 5 days, is sufficiently short to avoid a significant degradation of the intrinsically very stable molecule transported in a refrigerated condition.

All these biases cannot be controlled, irrespective of the environment. Therefore, the delimitation of the areas at risk and those not at risk of contamination must combine several approaches. In our case, these approaches are: 1) a hydrological approach based on the dispersal process of the chemical, 2) a historical approach based on practices, and 3) a pedological approach based on contamination data that leads to the zoning of the environment for risk management (restrictions on cultivation depending on the danger of contamination, for example). It should also be noted that the method has limited applicability in the case of low water-flow conditions (case of Grande-Terre) and of low levels of contamination since it will require a large number of samples to be taken from across the territory. Finally, the difficulty in drawing up contamination maps highlights the need for plot-level information on agricultural practices. The monitoring of practices remains the principal way to effectively manage the risks of contamination.

5. Conclusions

Our study focused on the delimitation of areas contaminated by a persistent organochlorine used more than 30 years ago in banana plantations in the French West Indies. The originality of our approach was to determine terrestrial contamination of a watershed from the quality of its watercourses, by dividing the territory on a hydrological basis. To our knowledge, this is a new approach in the field of pesticide contamination of soil and water that attempts to take advantage of the functional relationship between soil and river contamination. This approach has made it possible to identify areas at high risk of contamination that were as yet unknown, as well as areas where the probability of contamination was low.

The different biases that exist in the acquisition of water data as also in soil analyses and knowledge of practices do not allow for an unequivocal confirmation of the absence of contamination of a geographical area. The saving in costs compared to a systematic soil analysis is however significant (in this case about 219 sampling points led to information on 83% of the surface area of Basse-Terre). This method of analysis of the water at the watershed's outlet thus acquires relevance in the context of exploratory surveys. Furthermore, as it reflects soil pollution trends, this approach may complement monitoring of soil pollution over time. So, when used in combination with all the other information produced (risk map, soil contamination map), this method can become part of an approach for long-term risk management, including the identification of agricultural plots to be targeted as a priority during future analysis campaigns.

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