

## Chlordecone in the marine environment around the French West Indies: from measurement to pollution management decisions

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### Abstract

Chlordecone is a very persistent insecticide used in banana plantations of the French West Indies between 1972 and 1993. Chlordecone residues were found in inland water, in agricultural and freshwater products, and in marine organisms. This pollution has become of great concern in 2007. Therefore, a governmental action Plan was launched to better assess the pollution and to improve the consumer's protection. Within this plan, 1048 samples from 69 different species of marine fishes and crustaceans were collected all around both the Guadeloupe and the Martinique Islands and analyzed. The results confirm the presence of chlordecone in marine organisms, with highly variable concentrations (from the detection limit to 1000  $\mu\text{g.kg}^{-1}$ ). In 17.9 % of the samples, concentrations exceeded 20  $\mu\text{g.kg}^{-1}$ , the maximum acceptable level in fish according to the French regulation (Anon, 2008). Two main features of this contamination were characterized. 1) Because of the sedimentation of contaminated suspended soil particles, the sheltered bays are more exposed to chlordecone than the open coast where terrigenous flux are dispersed. 2) Species biology, particularly their lifestyle and diet, appear to influence contamination levels. Thus, the more contaminated species live in relation with sediment or are at high trophic level. These results have directly supported political decisions in order to prevent too high exposure of consumers to chlordecone. Fishing activities in sheltered bays have been forbidden for potentially highly contaminated species like benthic crustaceans and top predators. Further studies are under way to assess the importance of the trophic transport of chlordecone within the foodweb, and to evaluate the historical deposition of this insecticide in sediment and its further bioavailability.

**Keywords:** chlordecone (Kepone), French West Indies, fishes, crustaceans, organochlorinated contaminant, risk assessment, pollution management

### Introduction

Chlordecone (also known as Kepone) is an active organo-chlorinated substance which had been patented in 1952, and included in different pesticide formulations. Chlordecone ( $\text{C}_{10}\text{Cl}_{10}\text{O}$ ) has a very low aqueous solubility and conversely a high hydrophobicity ( $\log K_{\text{OW}} = 4.5$ ; UNEP, 2007). This substance resists to chemical and biological degradation processes. It has a high potential for bioaccumulation and biomagnification. Based on its physico-chemical properties and modelling data, it has been demonstrated that chlordecone can be transported for long distances (UNEP, 2007). For these reasons, it is now included in the more recent list of POPs according to the Stockholm Convention (<http://chm.pops.int/>).

Chlordecone is toxic to aquatic organisms. Crustaceans are the more sensitive species (UNEP, 2007). In small laboratory rodents, lethality occurs at exposure around 100  $\text{mg.kg}^{-1}$  body weight. The biological effects which have been mainly observed concern the reproductive system, the nervous system, the hepatic functions and musculo-skeletal disorders. In humans, similar effects have been observed in

exposed workers which presented also high chlordecone concentrations in blood. Chlordecone is an endocrine disruptor and is also classified as potentially carcinogen to human (Group 2B). In France, a PTDI (Provisional Tolerable Daily Intake) is fixed for chlordecone at  $0.5 \mu\text{g.kg}^{-1}$  body weight in the case of chronic exposure and it should not exceed  $10 \mu\text{g.kg}^{-1}$  b.w. (acute Reference Dose) in the case of accidental exposure (Afssa, 2005).

Historically, the first large accidental pollution by chlordecone in aquatic systems happened in the James River from a plant located in Hopewell, VA (USA), which produced the molecule from 1966 to 1975 when the production was halted after health problems were detected in workers. The contamination of sediments and shellfish in the river downstream the plant has been observed since 1967, very soon after the beginning of chlordecone production (Huggett and Bender 1980). The level of contamination regarding human health was considered worrying enough so as to close the fishing activity in the river for 13 years. The recovering of the river took about 30 years after the source of contamination was removed (Luellen *et al.* 2006). It should be noted that in the U.S. and at that time the safety limit in fish was established at  $300 \mu\text{g.kg}^{-1}$  fresh weight.

The main islands of the French West Indies are the archipelago of Guadeloupe (1630 km<sup>2</sup> and 400000 inhab.) and the Martinique Island (1128 km<sup>2</sup> and also 400000 inhab.). Their tropical climate is suitable for banana culture, but the warm and wet weather conditions are also favourable for many various pests that explain the important use of biocides. Approximately 180 tonnes of chlordecone have been extensively used in various technical formulations in banana plantations, and probably in a few other cultures, from 1972 to its ban in 1993, to wrestle with the weevil *Cosmopolites sordidus* (Cabidoche *et al.* 2009; Le Déault and Proccacia 2009). This insecticide persists in soils for a long time, and now an important soil surface is contaminated. Approximately 8300 ha in Guadeloupe and 10700 ha in Martinique are considered to be moderately or heavily contaminated by chlordecone (Le Déault and Proccacia 2009), which represent about 25 % of the land surface used for agriculture in each island. The areas at risks, where banana culture is the main activity, are located in the South-West of Guadeloupe (Basse-Terre) and in the Nord-East of Martinique.

After rainy episodes, rain water washed out chlordecone from soil, to the surface waters (Cattan *et al.* 2008) and then to the marine coastal waters. Transport of the substance from soil plantations to the sea, either dissolved or bound to soil particles may be important and rapid because of the importance and the violence of precipitations, because of the mountainous relief and of the relative narrowness of the territory of the islands. It is hypothesized that the eroded contaminated material is transported by rivers to the marine environment and settles close to the seashore, particularly in sheltered bays, where it becomes a source of contamination for marine organisms, and a potential hazard to marine life and finally to human health through seafood consumption.

A few of the characteristics of the local situation are relevant to the impact of chlordecone on fishing activities and seafood safety. First, the high diversity of fish of commercial interest: around the West Indies islands the fish fauna includes many species (Gobert, 1989). Among the demersal species landed, there is no predominant species. Indeed, about 180 demersal species are fished around the Martinique island and none represent more than 5 % of the landings (Gobert and Reynal, 2002). It means that the sampling should cover many various species. Secondly, fish and seafood constitute an important part of the human diet in Martinique and Guadeloupe, around  $55\text{-}110 \text{ g.j}^{-1}$ , approximately 2-3 times higher than the mean consumption of the French population of the mainland (Dubuisson *et al.* 2007). Thirdly, a more or less undefined part of the consumed seafood comes from local sources and follows very short distribution channels that could contribute to excessive exposure of sub populations to chlordecone.

In the French West Indies, the first concerns about the persistence of chlordecone in the banana plantations as well as its presence in the rivers and the wild fauna have been reported in 1977 (Snegaroff, 1977) and 1980 (Kermarrec, 1980). The first studies on contamination in marine fauna were carried out in the early 2000s with systematic investigations on the coastal marine fauna around Guadeloupe (Bouchon and Lemoine 2003, 2007); in Martinique, the studies focused on water and sediment in river plumes along the coastline (Bocquené and Franco 2005) and on fishery species from samples collected in harbours (Coat *et al.* 2006). These studies confirmed the contamination of the coastal marine system by chlordecone. The values recorded in the marine species revealed a much lower contamination in the marine environment than in the freshwater ecosystem, nevertheless without any information related to the possible extension of this contamination in the marine system. The concentrations encountered in the

marine species through these studies were generally under the maximal value recommended at that time for marine products by the French sanitary authorities ( $200 \mu\text{g.kg}^{-1}$  wet weight, Afssa 2005). At that time, two facts justified an increase of the research at sea. One was that the first series of studies were limited; indeed, they might be considered mainly as a warning on the possible extension of the contamination in the marine system, but did not inform on the extend of the contamination and on the factors acting on the contamination process. The second event was the decision of the French food safety authorities which drastically lowered the maximal residue limit (MRL) to  $20 \mu\text{g.kg}^{-1}$  (w.w.) in marine products for human consumption (Anon. 2008). Furthermore, this pollution became of great concern in the islands in 2007. Therefore a governmental Action Plan was launched to better assess the pollution and to ensure consumer's protection. Within this plan, various studies have been carried out since 2008 to characterize the marine fish fauna contamination around the islands.

This communication reports on the work done to improve the knowledge on the contamination distribution in the marine fishery species, and the new measures taken by the French authorities to prevent fish consumers from too high exposure to chlordecone. It is presented chronologically in two parts, referring to the evaluation of the contamination by chlordecone of the seafood at sea and to the pollution management.

### **Action step 1 (2008-2009). Preliminary characterization of the contamination and first pollution management decisions**

In the same time, various research initiatives have been scheduled at Ifremer through an incitement by the authorities in charge of environment and by the veterinary authorities through control and monitoring plans. All these investigations focused on the contamination of the marine living resources, on the consequence of this contamination on food safety, and on a potential social and economical impacts of prevention measures. Very shortly, the efforts of these bodies have been joined in a unique strategy in order to improve our knowledge of the contamination and, more precisely, to get a better coverage of the situation both in terms of space and of biological species.

#### *Sampling strategy*

The organisation of a sampling of marine fauna at a small geographical scale has to face with a lot of difficulties. Some are related to the access to wild specimens of species more or less abundant and scattered, others are linked to the very wide variety of species of interest in the area. The design of the sampling plan was based on two main hypotheses. The first one was that the contamination of the marine species might be related with the level of contamination of their biotope. The second one considered that contamination of the individuals might be related to their biology (lifestyle, feeding habits, etc.), with previous evidence of a strong relationship between contamination and feeding habits (Connolly and Tonelli 1985).

At the beginning, in both islands the spatial design was based on the definition of the water masses established according to various criteria (morphology of the coast, sediment, hydrology, hydrodynamical conditions) already considered for the implementation of the European Water Framework Directive (Diren 2005; Pareto *et al.* 2007). The objective was to distribute the samples equitably within the different areas, without prejudging on their potential contamination. Concerning the species diversity, it was evident that only a part of the existing species could be sampled. A typology of the species has been established, based on their feeding habits (combination of diet and trophic level), their mobility scheme (sedentary against mobile and migrant), as well as their habitat, all this information being collected from literature. Finally, four trophic types were defined by their trophic level and their main diet type: detritivorous, herbivorous or carnivorous.

The fish specimens were collected by professional fishermen with appropriate technical guidelines and each sampling position was reported. Finally, 69 different species were included in the 1048 samples collected in both islands in 2008 and 2009, with only very few specimen for some of them. The table gives the distribution of the samples collected according to their origin (zones) and to their trophic-feeding habit groupings.

Table 1. Distribution of the samples according the origin, the sampling period, the geographical zone and their feeding habit-trophic levels.

Z o n e	GUADELOUPE								MARTINIQUE							
	GDP-2008				GDP-2009				MTQ-2008				MTQ-2009			
	2- DN	2- Aut	3- Aut	4- Aut	2- DN	2- Aut	3- Aut	4- Aut	2- DN	2- Aut	3- Aut	4- Aut	2- DN	2- Aut	3- Aut	4- Aut
1	2	1	4	0	6	5	6	0	3	4	20	0	0	3	0	
2	2	2	13	4	22	21	65	19	15	17	16	0	90	41	23	4
3	3	10	21	2	8	9	30	5	6	16	5	0	16	12	15	8
4	0	3	1	2	6	1	8	0	4	7	5	0	5	2	2	1
5	1	5	6	2	6	5	15	7	0	0	0	0	0	0	0	0
6	3	9	15	2	11	27	27	6	0	12	20	0	0	4	11	3
7	2	2	4	0	6	0	5	0	2	7	9	0	22	7	3	0
8	2	1	2	0	4	0	5	0	8	0	1	0	8	0	0	0
9	3	3	8	0	7	5	0	0	0	9	6	0	0	1	2	1
10	2	8	11	0	4	5	0	0	0	3	11	8	0			2
11	0	0	0	15	0	0	0	0								

With 2-DN: detritivorous, 2-Aut: herbivorous, 3-Aut: carnivorous 1 (secondary consumers), 4-Aut : top carnivorous.

### Analytical techniques

Fish were wrapped in aluminium foil and kept in deep freezer. Fish samples to be analyzed for chlordecone were prepared according to precise recommendations (Afssa, 2007): composite samples of fish flesh were prepared taking out the fillet with the skin from at least 3 fish specimens having the same characteristics (species, size and origin). Then the meat that constituted the composite sample was homogenized before analysis.

For the different surveys the analyses have been done in three laboratories and basically two methods were applied. The protocols include the extraction of the contaminants from the homogenized fish tissue and a further clean up of the extract. In these analytical methods, the instrumental determination step differs, either by GC-ECD (Gas Chromatography with Electron Capture Detection) with a quantification limit at 5  $\mu\text{g.kg}^{-1}$  (wet weight) for a sample amount of 2 g or by GC – MS (Gas chromatography and mass spectrometry). One other laboratory used HPLC-MS-MS detection (High Pressure Liquid Chromatography and double mass spectrometry detection) with a better specificity and a lower quantification limit, below 0.5  $\mu\text{g.kg}^{-1}$  w.w.

### Main results

The results of the investigations carried out in 2008 confirmed the presence of chlordecone in marine organisms, with highly variable concentrations (from below the quantification limit to 1000  $\mu\text{g.kg}^{-1}$  w.w.). In 17.9 % of the samples, concentrations exceeded 20  $\mu\text{g.kg}^{-1}$ , the maximum residue level in fish (Afssa, 2008). Two main features of this contamination were characterized.

1) Most of the highly contaminated fishes and crustaceans were caught in areas located close to contaminated basins. More precisely, the specimens living in sheltered bays (zones 1 and 5 in Guadeloupe; zones 2 and 7 in Martinique; fig. 1 and 2) were more exposed to chlordecone than those living outside where terrigenous flux are more dispersed. This has been related to the sedimentation dynamics of contaminated suspended soil particles. The results have shown that the spatial distribution according to the WFD zonation was useful to guide the sampling campaign around the islands, but it was not fully appropriate to characterize the type of contamination encountered, particularly the type of bottom (sandy/muddy). The use of precise individual positioning of each sample was required.

2) The species biology, particularly their lifestyle and diet, appeared to strongly influence contamination levels. Thus, the more contaminated species, detritic feeders and carnivorous, were living in relation with sediment or were at high trophic level.

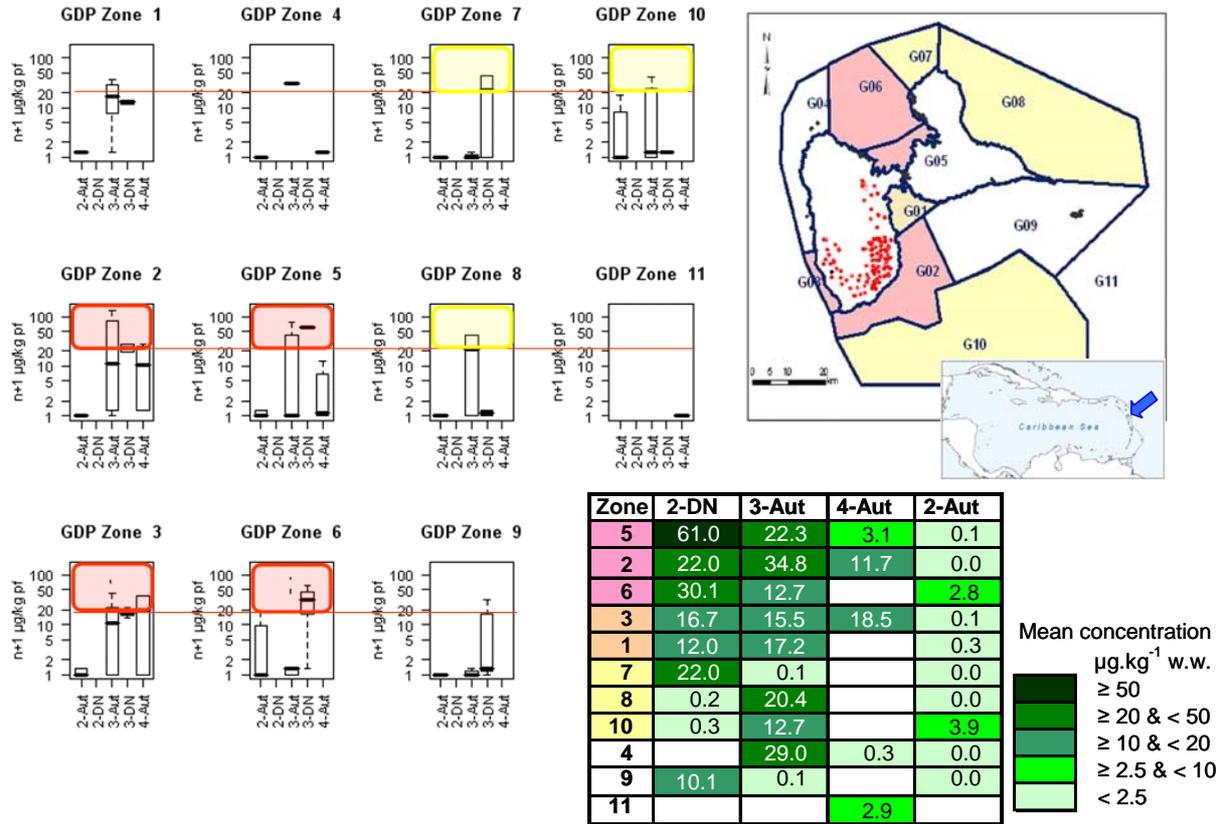


Fig. 1. Main trends of the chlordecone contamination in the fished marine fauna from Guadeloupe.

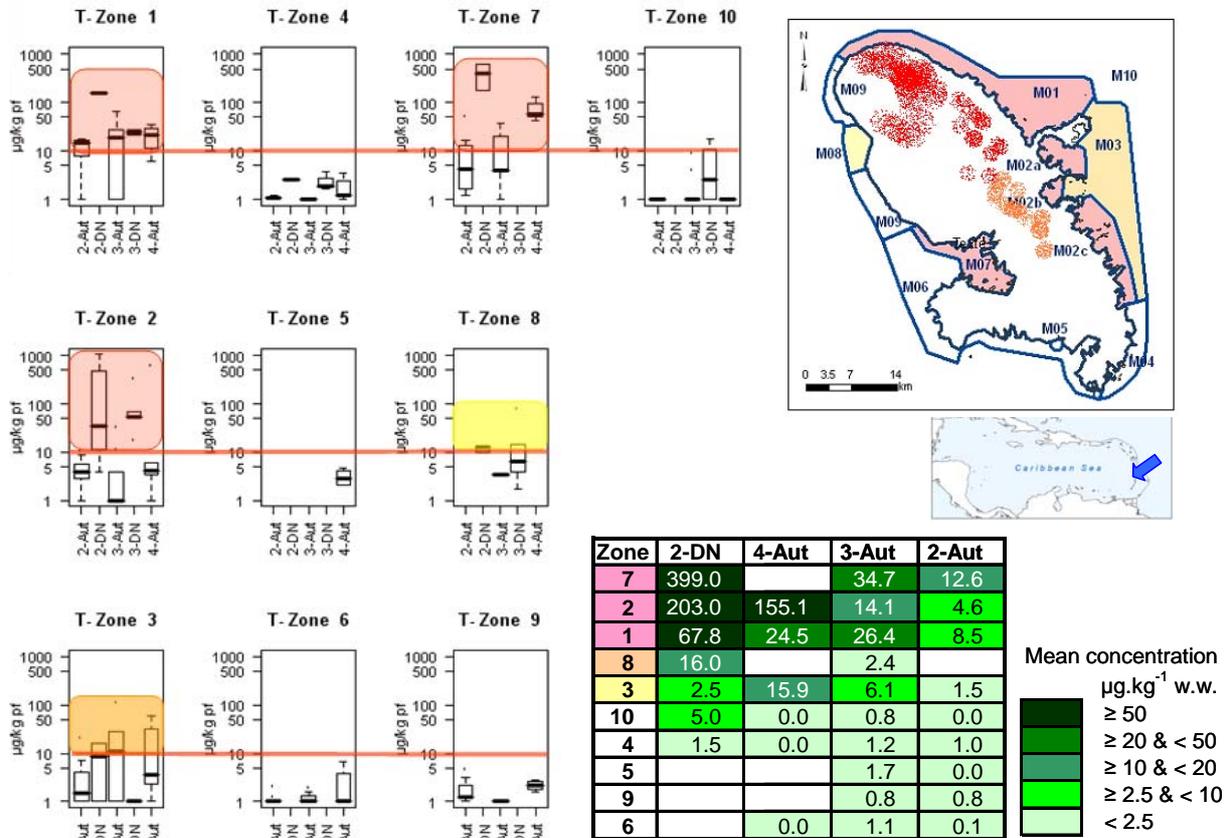


Fig. 2. Main trends of the chlordecone contamination in the fished marine fauna around Martinique.

### *Management decisions*

All along the research process, a strong partnership was maintained with the local authorities acting within the chlordecone action plan (from veterinary services, marine affairs, environmental department) as well as with fishermen representatives (regional fishery councils) through ad hoc advisory committees. It allowed these stakeholders to share a same knowledge on the research progress and to give their inputs all through the course of the studies.

The results presented above have directly supported political decisions in order to prevent fish consumer from too high exposure to chlordecone. Indeed, since 2009, a first set of measures of conservation have been taken in each island taking into account the available data (Anon. 2009). These limiting measures on fishing activity were based on two observations: the geographical distribution and the biological species. The main principles of these measures were to introduce restrictions targetted on the species/areas for which results above the MRL had been encountered during the surveys.

Due to the particular sensitivity of this problem, the local authorities have made special efforts to explain the measures both to the fishermen and to the population in order to ensure their full application. In the communication on risk management it is also important and maybe difficult to account for potential social and economical impacts of such regulations that could perturb the fish market well beyond the strict scope of fishing limitations. The main actions done were interviews in local medias (radio and TV), meetings with fishermen in the harbours and spreading a leaflet explaining the measures easy to use by the fishermen, particularly those who usually sell themselves their fish.

### **Action step 2 (2009-2010). Confirmation investigations and further management decisions**

The first step of action was considered as precautionary. Indeed, the knowledge available at that time was considered sufficient by the sanitary authorities to justify the development of one set of precautionary measures. Nevertheless, the extent of the contamination of marine fish species was not so fully characterised as to allow the definition of all the suitable measures. For this reason, a second step of actions has been organized in 2009-2010 in order to complement the knowledge and to prepare eventual further management measures.

### *Sampling strategy*

For the 2009 surveys, the sampling scheme was designed to improve the information on the contamination in the different areas and in the various species which were insufficiently described during the first survey, with different main targets in the two islands.

In Martinique, a general schedule of contamination was available from the first step; thus the priority was to identify areas more or less free from contamination beside highly contaminated areas, the last been closely related to the proximity of contaminated catchment basins and to the geography of the coast (sheltered bays vs. open sea coasts). Around this island, a special care was directed towards the most potentially exposed species in the most sensitive areas. Finally three objectives guided the sampling operations during this 2009 campaign in Martinique:

- a better coverage of the areas and of the various trophic groups, notably the detritic feeders from the zone 1, and all other trophic groups from the zones 1, 2 and 3 (fig. 2),
- an attempt to improve the information on the contamination of very costal species at higher risk like the blue land crab *Cardisoma guananhumi*,
- an estimation of the variability of the contamination, focusing on detritic feeders like the Caribbean spiny lobster *Panulirus argus* from the zone 2b and on carnivorous pelagic fish.

In Guadeloupe, two main objectives were aimed, the first targetting the most sensitive areas and species (like in Martinique), the second one devoted to complement the general investigation on the contamination all around the island.

Finally, in 2009, 280 crustacean and 626 fish samples were collected and analyzed both by Ifremer and by the veterinary services within various monitoring plans around the Martinique and the Guadeloupe islands.

## Results

Globally the results obtained in this second series of fish and crustacean samples collected in Martinique in 2009 confirmed and validated the previous observations. Again the higher concentrations were found in detritic feeders (2-DN group) and in higher carnivorous species (4-Aut group); for the spatial distribution, a same trend as during the step 1 was observed which confirmed a higher contamination in the sheltered bays. The land crab, which feeds on leaves and fruits on land was virtually not contaminated. Oppositely, the swimming crabs *Callinectes spp.* from the zones 2 and 4 presented very high chlordecone concentrations. The contamination of Caribbean spiny lobster *Panulirus argus* samples obtained in the zone 2 is high and varies within a large range. This variability was not explained by the size of the specimen, but the number of samples was too limited to allow the estimation of any significant relationship between size and contamination level. In higher predators like the King mackerel *Scomberomorus cavalla* the very high contamination in a sample from the zone 2 was not confirmed by the analyses of samples from the zones 4 and 9.

One more general observation concerns the distribution of the concentration in the set of data: most of the results data are very low values, frequently around the quantification levels and few significant or high concentrations are measured. An increase of the sampling might lead to the measurements of more very high levels which move upwards the average calculated concentration.

The samples collected in 2009 around Guadeloupe confirmed those obtained in 2008. The chlordecone contamination is very present in the south coast of Basse Terre. These new results allowed to identify few species particularly sensitive to the chlordecone contamination, such as spiny lobsters (*Panulirus argus* and *P. guttatus*), certain coastal pelagic fishes (*Caranx latus*) and more generally most of the carnivorous species. Again the low contamination of herbivorous species has been observed, even in potentially exposed areas.

Last and importantly, high sea pelagic fishes are not contaminated by chlordecone; those fishes, such as tunas, wahoo *Acanthocybium solandri*, dolphin fish *Coryphaena hippurus*, flying fishes represent a major part of the landings in the French West Indies.

## Management decisions

Taking into account the results of analyses and the conclusions of the monitoring plans, in 2010 the authorities of Guadeloupe have modified the fishing restrictions around the island (Anon. 2010a). Shortly, the main principles of this new decision were to ban the fishing of all the species in the areas deemed as the most contaminated, with the exception of a short list of clearly defined species, mainly herbivorous, which are presumed not being contaminated. Additionally, in other areas some species considered more at risk are also banned to fishing.

At this time (30 July 2010) fishing restrictions have not yet been promulgated in Martinique. New management decisions are expected very soon; discussions between the professional organizations and the sanitary authorities are still in progress to define the best measures with the aim to fully apply the sanitary regulations and to minimize, as far as possible, their consequences on the fishing activity.

## Conclusion

The activity done during the last years in the French West Indies has allowed effective progress both in knowledge of the chlordecone contamination of the marine living resources and the risk management. Two main driving factors of contamination have been identified: the characteristics of the environment, principally the location of the areas with regard to the contamination of the catchment basins and the local hydrodynamics in one hand, and some biological features of the species, principally their diet and habitat preference in the other hand. All this information has been used to define the management of fishing in order to reduce the risk of fishing animals contaminated at level exceeding the MRL. The management measures rely upon a recent and sound knowledge of the contamination around Guadeloupe and Martinique. At each critical step of progress, the communication with the professional fishermen and to the citizens was a central strategy implemented by the authorities. At each stage of this course, the researchers have given a support by explaining their main new results. This process has been considered useful to improve the meaning of the measures by all the stakeholders, to promote its application and, as much as possible, to reduce the risks of misinterpretation, in a particularly sensitive context in the islands.

Further studies are under way. Their first objectives are to assess the importance of the trophic transport of chlordecone within the foodweb, and to evaluate the historical deposition of this insecticide in sediment and its further bioavailability. Nevertheless, this pollution is foreseen to exist for a very long time in the French West Indies. As explained in by the chairman of the scientific council of the governmental action plan on chlordecone "the Caribbean populations will have to live with this pollution for a long time, probably several generations" (Anon. 2010b). In this context, efforts have to be maintained in the near future to better know the characteristics of this pollution in the marine ecosystems such as to take better into account its impact on ecosystems, on human health as well as on economical activities, and such as to adjust mitigation measures at the best, by integrating these objectives comprehensively.

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