

ASSESSMENT OF THE PESTICIDES POLLUTION OF CORAL REEFS COMMUNITIES  
FROM FRENCH POLYNESIAHélène ROCHE<sup>1</sup>, Bernard SALVAT<sup>2</sup> & François RAMADE<sup>1</sup>

RÉSUMÉ. — *Évaluation de la pollution par les pesticides des communautés coralliennes en Polynésie française.* — Un programme de recherche a été développé dans le cadre de l'Initiative Française pour les Récifs Coralliens (IFRECOR) afin d'évaluer la contamination de la biocénose récifale par les pesticides. Les analyses ont mis en évidence une contamination des organismes majeurs du réseau trophique en particulier par des herbicides des groupes des chloracétamides et des triazines, ainsi que par les principaux insecticides organochlorés. De façon inattendue, des niveaux significatifs de chlordécone (képone®) ont été mis en évidence dans les espèces analysées, dont des poissons consommés localement. Ce fait est d'autant plus préoccupant que l'usage de cet insecticide, dont la rémanence dans les sédiments se chiffre en millénaires, n'aurait jamais été officiellement homologué en Polynésie française.

SUMMARY. — Researches have been carried out in the framework of the French Initiative for Coral Reefs in order to assess the pollution levels of the coral reef trophic webs by pesticides in French Polynesia. An overall contamination with critical organisms from various levels of the trophic web was shown. On one hand, analysis showed a contamination by chloroacetamide and triazine derivatives, two major families of herbicides. On the other hand the occurrence of organochlorine insecticide has proven ubiquitous. Especially worrying is our finding of chlordacone (kepone®) occurrence in all of the organisms investigated, mainly in fishes, which are commonly ingested by the local inhabitants. Moreover, these data are very worrying as the average residence time of chlordacone in sediment numbers in thousand of years.

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The French committee IFRECOR (Initiative Française pour les Récifs Coralliens) is the French section of the *International Coral Reef Initiative* (ICRI). Enacted in 1999 and ruled by the Ministry of Ecology, Energy and Sustainable Development and the one of the French Overseas Territories, his major duty is to set in practice the international conventions and laws addressing the challenge of the coral reef preservation. Indeed, this kind of benthic marine ecosystem is by far the most threatened in the world even before the tropical rain forests. About 20 % of the world reefs are already extinct and over the 50 % remaining might vanish prior to the midst of this century (Wilkinson, 2008). France stands as the 4<sup>th</sup> country in the world regarding the acreage of coral reefs under its rule. For example, the Great Barrier Reef of New Caledonia ranks second after the Australian Great Barrier Reef spelling over 1600 km and recently nominated as an UNESCO World Heritage Site. Therefore, France has to be deeply involved in reefs conservation.

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The IFRECOR Committee, amongst a number of activities, manages several programs said ‘Topics of Transversal Interest’ (=TIT) which involve the whole of the French overseas communities. Amongst a tenth of them, the one entitled Pesticides is intended to assess the pollution of reefs communities by the major compounds in use in agriculture. Various researches have previously demonstrated that pesticides rank among the main stressors of coral reefs and stand as a major threat to coral reefs ecosystems (cf. e.g. Ramade & Roche, 2006).

Pesticides run off and leaching in the agricultural lands from catchment areas is the prevailing source of these compounds to the marine coastal habitats (Lewis *et al.*, 2009) especially during the tropical wet season. Rivers and creeks water plumes lead to large water volume being discharged in waters surrounding inshore coral reefs.

## RESEARCH PROGRAM ON THE STUDY OF THE CONTAMINATION OF FRENCH OVERSEAS REEFS BIOTA BY PESTICIDES

The IFRECOR Committee has launched a survey of pesticides residues<sup>1</sup> in some key organisms of the coral reef trophic webs from 6 French overseas tropical communities, namely: French Polynesia, French West Indies (Guadeloupe and Martinique), Mayotte, La Réunion and Wallis. On each island involved in this program a sampling schedule has been carried out on two inshore coral reef sites: one said ‘agricultural’ as located next from terrestrial agricultural areas and the other ‘traditional rural areas’ as located outside terrestrial areas devoted to intensive agriculture. Two sampling campaigns per year have been achieved (in dry season and in wet season). Samples were preserved in dry ice and sent in France for chemical analysis. Some of them have been processed in the ESE Laboratory from the Orsay Science Faculty (University of Paris-Sud), the rest in the Laboratory of Toxicology of the National Veterinary Institute (Ecole Nationale Vétérinaire de Lyon = ENVL) in Lyon. The compounds investigated were organochlorine pesticides (included Kepone = Chlordecone) and herbicides both from the chloroacetic acid, triazine, and substituted urea families; the last two families having proven experimentally of a highest toxicity for the zooxanthella *Symbiodinium* endosymbiont from scleractinian corals.

These various pesticides have been detected in muscle, tegument and liver by use of Gas Chromatography coupled to mass spectrometry or equipped of an electron capture detector depending on the compounds studied.

The species selected for the basic monitoring program are fishes as they stand as good bioindicators of pollution in aquatic habitats. Two kinds of species have been sorted out. The first one is an herbivorous species from the *Scaridae* Family, a parrot-fish which is ubiquitous in coral reefs elsewhere in the world and which displays an algophagous and detritivorous diet. The second species is carnivorous, from the *Serranidae* Family, a grouper (*Epinephelus* sp.). These fishes are superpredators (= tertiary consumers) and rank next from the top of the trophic web (top predators).

A more detailed sampling program has been completed in French Polynesia where samples have been caught amongst various key species from the reef trophic web. It has implied a green alga (*Halimeda crassata*, Family *Halimedaceae*), a mollusk (*Tridacna maxima*), a scleractinian coral species (*Fungia*), an holothurian (*Halodeima atra*) which is a detritiphagous, and two fishes *Chlorurus sordidus* (*Scaridae*) and *Epinephelus merra* (*Serranidae*). Species were collected in two sites in each of the two islands of the Society archipelago (Tahiti and its sister island Moorea). Even if no very important agriculture occurs in these two islands (as compared with Martinique), we selected two littoral sites as different as possible, related to market gardening. Sites 1 are with some agriculture in the watershed (“Port Phaeton” on Tahiti – “Entre deux baies” on Moorea) and sites 2 are without modern farms but with some local - and of tiny scale - food-producing crops (‘Vairao’ on Tahiti - ‘Tiahura’ on Moorea).

## RESULTS

### KEPONE (= CHLORDECONE) CONCENTRATIONS

Amongst the most unexpected findings is the detection of the ubiquitous occurrence of Kepone<sup>2</sup> which has proven existing at significant concentrations in all the analysed organisms (Tab. I & Fig. 1). Surprisingly, this pesticide would have officially never been in use in French Polynesia.

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<sup>1</sup> One of us, François Ramade, is the Supervisor of this IFRECOR Programm ( TIT 3, Pesticides).

<sup>2</sup> Kepone® is the current name of this compound in North America. In Continental Europe, it is known as Chlordecone, which is its official common name according the IUPAC nomenclature and sometimes Curlone® name under which it was in use in the former French African colonies and in French tropical overseas territory as French West Indies. Another derivative of close chemical structure, the Perchlordecone or Mirex® has been widely in use in the United States, even more than Chlordecone, and various countries of tropical America from the 1950s to the 1970s. Over 100 000 km<sup>2</sup> were for example sprayed in the 1960s by air with this insecticide in the southeastern United States for the fire ant control!

TABLE I

Concentration (geometric mean of analysed samples) in muscle and tegument of chloroacetamine herbicides (alachlore and metolachlor) and Kepone in samples from Moorea and Tahiti. Values are expressed in  $\text{ng}\cdot\text{g}^{-1}$  (wet weight).  $n$  (number of analysed samples) = 4 to 6,  $nd$  = not detected,  $nm$  = not measured)

Sites	Organisms	Alachlor	Metolachlore	Kepone
Moorea site 1 'Between two bays' some agriculture	<i>Halodeima</i>	0.004	0.004	1.00
	<i>Epinephelus</i>	0.003	0.090	0.32
	<i>Chlorurus</i>	0.002	nd	1.19
Moorea site 2 'Tiahura' no agriculture	<i>Halodeima</i>	0.005	nd	0.95
	<i>Epinephelus</i>	0.002	nd	0.89
	<i>Chlorurus</i>	nm	nm	nm
Tahiti site 1 'Port Phaéton' some agriculture	<i>Halodeima</i>	0.004	0.004	0.44
	<i>Epinephelus</i>	0.007	nd	0.26
	<i>Chlorurus</i>	0.004	nd	0.43
Tahiti site 2 'Vairao' no agriculture	<i>Halodeima</i>	0.005	0.008	1.47
	<i>Epinephelus</i>	0.013	nd	0.94
	<i>Chlorurus</i>	nd	nd	0.87

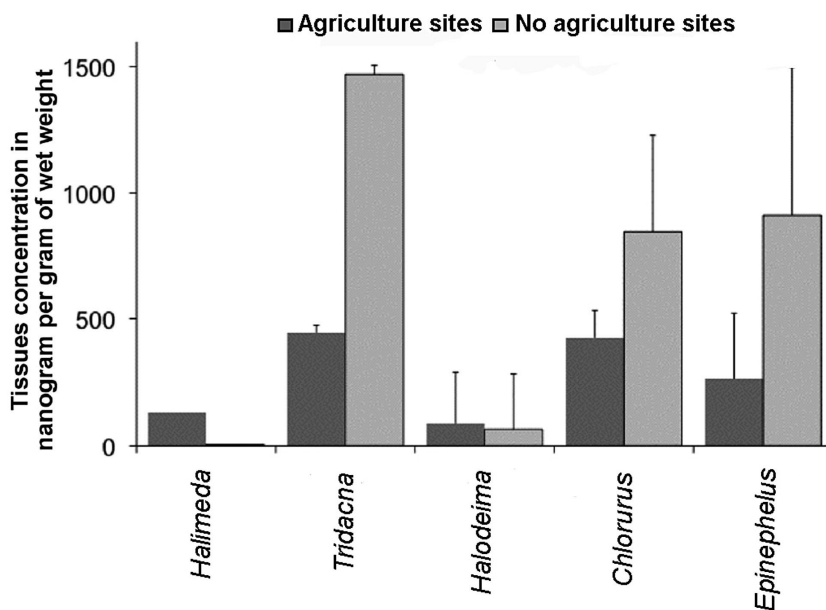


Figure 1. — Contamination by Kepone of some key species from the coral reefs in sites located off areas of “modern” agriculture or off “no agriculture” areas. Where analysed: whole tissue from *Halimeda*, mantle from *Tridacna*, digestive tract from *Halodeima*, filets from *Chlorurus* and *Epinephelus*.

Another unexpected result is that the average concentrations of Kepone — as well as the ones of herbicides — are higher in organisms from reefs sites located off rural ‘traditional’ areas than in those off areas of intensive agriculture. Nevertheless it must be considered that the agricultural activities are still traditional in these islands and fields are loosely distributed despite the existence of some ‘modern’ farms. Much alike what has occurred in French West Indies, one may assume that the use of Kepone and that of a vast array of other pesticides by this self-sufficient agriculture has been both widespread and in a way more or less regulatory regarding the compounds involved in this study.

## HERBICIDES CONCENTRATIONS

The major herbicides detected in Polynesian reef organisms are atrazine, simazine, alachlor, and to a lesser extent terbutylazine (Tab. I & II). Analysis has moreover shown up trifluraline, a toluidine derivative herbicide, in some samples. Paradoxically, substituted urea herbicides have been undetected, even diuron, though the use of this herbicide in tropical area is prevailing, and ranks frequently at the highest concentrations in the waters of inshore coral reefs from the Australian GBR (Shaw & Müller, 2005; Lewis *et al.*, 2009).

TABLE II

Concentrations (geometric mean of analysed samples) of herbicides (trifluraline, atrazine, simazine, terbutylazine and diuron). Values are expressed in  $\text{ng.g}^{-1}$  wet weight. n (number of analysed samples) = 4 to 6, nd = not detected, nm = not measured

Sites	Organisms	Trifluraline	Atrazine	Simazine	Terbutylazine	Substituted urea (diuron)
Moorea site 1 'Between two bays' some agriculture	<i>Halodeima</i>	0.002	0.012	nd	nd	nd
	<i>Epinephelus</i>	nd	0.005	0.023	nd	nd
	<i>Chlorurus</i>	nd	0.016	0.010	nd	nd
Moorea site 2 'Tiahura' no agriculture	<i>Halodeima</i>	nd	0.013	nd	nd	nd
	<i>Epinephelus</i>	0.002	0.010	nd	nd	nd
	<i>Chlorurus</i>	nm	Nm	nm	nm	nd
Tahiti site 1 'Port Phaéton' some agriculture	<i>Halodeima</i>	nd	0.017	0.007	0.014	nd
	<i>Epinephelus</i>	nd	0.013	0.004	0.044	nd
	<i>Chlorurus</i>	nd	0.006	nd	0.018	nd
Tahiti site 2 'Vairao' no agriculture	<i>Halodeima</i>	nd	0.008	0.005	0.023	nd
	<i>Epinephelus</i>	nd	0.008	nd	0.033	nd
	<i>Chlorurus</i>	nd	0.008	0.002	nd	nd

A major problem generated by the reef water contamination by herbicides (Jones *et al.*, 1997; Owen *et al.*, 2002; Hill *et al.*, 2005) is related to their potential risk for the photosynthetic activity of the zooxantella *Symbiodinium* sp., especially Photosystem II herbicides (Jones & Kerswell, 2003). These endosymbiotic *Dinoflagellata* are common to the whole species of scleractinian hard coral and others hermatypic organisms from reef biota such as Molluscs of the *Tridacnidae* Family.

Usually the concentrations inhibiting photosynthesis of *Symbiodinium* observed in the waters of inshore reefs are under or at the lower level to which currently occurs photosynthesis inhibition. Here, the herbicide concentrations in reef organisms were in some instances of the value to which a significant photosynthesis inhibition may be detected.

It could be noted that Negri *et al.* (2005) showed a growth inhibition of young colonies of scleractinian corals exposed to triazine herbicides.

## OTHER INSECTICIDES CONCENTRATIONS

Organochlorine insecticides have been investigated in all of the reef organisms sampled (Tab. III). They have been investigated in the hepatic tissues of fishes and giant clam and in the gut of holothurians. Though overall levels of concentration are rather low, they may reach elevated concentrations in some samples. For example,  $813 \text{ ng.g}^{-1}$  (= 0.8 ppm or  $\text{mg.Kg}^{-1}$ ) of  $\Sigma$ DDT were found in the liver from a grouper (*Epinephelus merra*) and  $1080 \text{ ng.g}^{-1}$  ( $\approx 1 \text{ ppm}$ ) were detected in parrot-fish (*Chlorurus sordidus*) livers from Tahiti fringing reef and  $999 \text{ ng.g}^{-1}$  in samples of the same species from Moorea. Similarly,  $311 \text{ ng.g}^{-1}$  of Endosulfan were found in the liver of a *Chlorurus sordidus* from Moorea Tiahura.

TABLE III

Concentration (geometric mean of analysed samples) of organochlorine insecticides. Values are expressed in  $\text{ng.g}^{-1}$  (dry weight).  $n$  (number of analysed samples) = 4 to 6, nd = not detected

Sites	Organisms	$\Sigma\text{HCH}$	$\Sigma\text{hepta-chlor}$	$\Sigma\text{endrin}$	$\Sigma\text{endo-sulfan}$	Aldrin + dieldrin	$\Sigma\text{DDT}$
Moorea site 1 'Between two bays' some agriculture	<i>Halodeima</i>	nd	6.5	nd	1.3	10.0	44.0
	<i>Epinephelus</i>	7.0	179	1.9	3.0	54.7	64.2
	<i>Chlorurus</i>	149	nd	0.5	6.6	11.9	172
Moorea site 2 'Tiahura' no agriculture	<i>Halodeima</i>	25.2	193	0.8	4.3	21.7	393
	<i>Epinephelus</i>	114	121	nd	2.9	67.7	220
	<i>Chlorurus</i>	26.4	195	54.6	311	21.1	999
Tahiti site 1 'Port Phaéton' some agriculture	<i>Epinephelus</i>	nd	nd	38.4	236	0.8	813
Tahiti site 2 'Vairao' no agriculture	<i>Halodeima</i>	34.4	8.9	nd	9.1	29.7	13.5
	<i>Epinephelus</i>	84.0	4.7	3.0	31.1	2.8	147
	<i>Chlorurus</i>	8.6	nd	19.7	304	5.9	1088

Samples from *Halodeima* digestive tissue and from parrot-fish liver have proven much contaminated despite their lower position into reef trophic web: *Holothuria* sample from Moorea Tahiuara held up to  $193 \text{ ng.g}^{-1}$  of  $\Sigma\text{heptachlor}$  and  $393 \text{ ng.g}^{-1}$  of  $\Sigma\text{DDT}$ .

This point has to be correlated to the diet of these reef species. Holothurians are detritivorous and currently ingest vast amounts of sediment for feeding. Parrot-fishes scrap the surface of dead corals in order to collect algae on which they feed. In both cases, particular matter from terrigenous origin, loaded with pesticides, is slowly deposited on the substrate so that the considered species are overexposed to pesticides.

## OVERALL DISCUSSION

Coral reefs contamination by land use of pesticide in the intensive - so-called "modern" - agriculture is considered as a major threat for the preservation of coral reefs ecosystems (Fabricius *et al.*, 2005; Ramade & Roche, 2006).

Researches we carried out in the reefs biotas from the Society Islands in French Polynesia have shown up a diffuse and ubiquitous contamination of coral reefs communities by organochlorine insecticides and by some major groups of herbicides especially triazine and chloroacetamide derivatives. This contamination has proven uncorrelated with any level of intensity of agriculture in the cultivated coastal areas in the immediate vicinity to the reefs sampled.

Organochlorine insecticides showed the highest concentration comparatively to what has been observed elsewhere in reef communities from others biogeographical areas. The average level of contamination we found out has proven of the same magnitude and even higher in comparison to that detected in fishes and invertebrates of Florida reefs biotas and rather higher than that from the Australian Great Barrier Reef. Formerly, Dichmann *et al.* detected  $0.11 \text{ ppm}$  of  $\Sigma\text{DDT}$  ( $=1100 \text{ ng.g}^{-1}$ ) in the fats (reported to the whole body weight) in *Sphyræna barracuda* caught in Florida reef waters. Glynn *et al.* (1995) have found  $1.8 \text{ ng.g}^{-1}$  of DDT in fillets from Grunts (*Haemulon plumeri*, a carnivorous fish of the Family *Haemulidae*) and later (Glynn *et al.*, 1995)  $1.2 \text{ ng.g}^{-1}$  of aldrin in fillets from the same fish species from Key Largo Marine sanctuary and Pennekamp Coral Reef State Park, a rather unpolluted reef area of Florida. Available data regarding the organochlorine contamination of fishes from the Australian GBR display similar or even lower contamination levels than in Florida. For example Von Westerhagen & Klumpp (1995) detected in average from  $0.04$  to  $0.1 \text{ ng.g}^{-1}$  (assessed in equivalent dry weight) of DDE in samples from *Scomberomorus commersoni* (a carnivorous

fish, Family *Scombridae*) caught in the reefs from the Cairns area and 2.3 ng.g<sup>-1</sup> were found in *Lates calcarifer* (*Latidae* from the Perciform Order) sampled in the McKay area, despite intensive cash crop cultivations occurring in this coastal area from the GBR. Comparatively, our average ΣDDT concentration is over 100 ng.g<sup>-1</sup> and exceeds at its highest 1000 ng.g<sup>-1</sup> in a grouper sample from Tahiti. Regarding dieldrin, 0.6 ng.g<sup>-1</sup> were found in samples from *Scomberomorus commersoni* caught in the reefs from the Cairns area and peaked at 9.7 ng.g<sup>-1</sup> in one sample of *Lates calcarifer* from the McKay area. At the opposite our findings for the dieldrin concentrations are in the range from 0.8 ng.g<sup>-1</sup> to 67.7 ng.g<sup>-1</sup>, values which exceed usually the ones detected in the GBR fishes.

One of the major findings of our work is that surprisingly Kepone has been detected everywhere in the sites sampled, and in all of the organisms analysed. Such ubiquity was as well observed with herbicides, though detected concentrations were at much lower levels than the ones of these previous pesticides.

Paradoxically at first sight, this widespread contamination of water from inshore reefs by pesticides is usually loosely related – if not even uncorrelated - to their vicinity to inland areas of intensive agriculture, a fact that has been previously quoted in several research works. For example, Shaw *et al.* (2010), observed that the concentrations of herbicides in Hanah Island fringing reef waters, in a remote area from the far North Queensland stand at the same level of the ones detected in coastal waters in areas where river discharge waters overloaded in pesticides from watershed with intensive agriculture land use, in more southern areas.

Regarding Kepone, it seems to us that its distribution in reef organisms, paradoxical at first sight, may be explained not only by coastal currents, which drive it far from a coastal point source pollution, but additionally through the fact that traditional agriculture, which is widely dispersed, can supply its odd distribution disregarding the occurrence of intensive cultivations. A similar distribution has been observed in Martinique where the concentrations of Kepone display analogous values in the waters from river mouths in intensively cultivated watersheds and from those of dispersed ‘traditional’ agriculture (Bocquene & Franco, 2005).

Indeed, the Kepone contamination of reef organisms from French Polynesia stands as a worrying risk for public health as it has been previously experienced in French West Indies. It stands particularly as a major problem in Martinique where this insecticide has been widely in use on bananas and others tropical exportation cultivations, and, last but not least, on vegetable crops due to its stealthy use in the traditional agriculture. Subsequently, Kepone has been the source of a pervasive contamination of soils, inland and coastal waters, fishes and others edible marine animals.

Another major concern related to Kepone contamination of reef biota stands into the extreme remanency of these pesticides in the aquatic environments as its average time of residence is assumed exceeding centuries, may be even millenium, in marine sediments. Researches have been carried out in Virginia, where was located, in Hopewell, the only American factory of Kepone, whose facilities were closed in 1975. A survey program was initiated in the late 1975 on the Kepone pollution of the fish community from the James River and of its estuary in the Chesapeake Bay, contaminated by the plant release of wastewaters (Luellen *et al.*, 2006). During the first years after the plant closure, was observed a fast decline of the Kepone content in fishes. Later this decrease of the fish contamination levelled and the Kepone content still remain at a constant value since two decades, the concentrations detected in fishes tissues having displayed a ‘plateau’ with time. Therefore, a fish consumption advisory is still in effect over thirty years after the source of contamination was removed, the concentrations still standing at an average level of 50 to 100 ng.g<sup>-1</sup> some of them exceeding the limit values admitted in human food (0.3 µg.g<sup>-1</sup> fresh weight).

Up to now, the concentrations of Kepone detected in tropical fish from French Polynesia are happily well below this dietary limit value (about a hundredfold below in average). With an average value of 0.9 ng.g<sup>-1</sup> in Moorea and 0.7 ng.g<sup>-1</sup> in Tahiti for the reef animals we analysed, the contamination in these island reef biotas stands somewhat lower than the average levels found in French West Indies. In Martinique this average value has been assessed at 1.4 ng.g<sup>-1</sup> for the whole sampling of coral reef animals by IFREMER scientists who found a

lower average of 0.3 ng.g<sup>-1</sup> for those from Guadeloupe (Bertrand *et al.*, 2009). Nevertheless, though the value of the vast majority of fishes analysed stayed under 20 ng.g<sup>-1</sup>, some West Indies samples from marine fishes muscles have proven far more contaminated. Bertrand *et al.* (2009) have found 50.5 ng.g<sup>-1</sup> in a Martinique Parrot-fish (*Sparisoma chrysopterygum*) and the concentrations they recorded peaked at 126 ng.g<sup>-1</sup> in a grunt (*Haemulon carbonarum*). In Guadeloupe, it was found up to 42 ng.g<sup>-1</sup> in a grouper species (*Epinephelus guttatus*) and concentration peaked at 133 ng.g<sup>-1</sup> in an Emperor fish (*Lutjanus synagris*).

Coat *et al.* (2005) had previously found in Martinique high concentrations (386 ng.g<sup>-1</sup>) in wild red tilapia (*Oreochromis* sp.) from a small river and 132 ng.g<sup>-1</sup> in the same tilapia species from fish farming.

## CONCLUSION

The investigations that we have carried up in the Society Islands from French Polynesia have demonstrated a widespread contamination of reef organisms by herbicides derivatives from chloroacetic acid and triazine, as well as on ubiquitous occurrence of organochlorine insecticides, particularly Chlordecone. This contamination was observed in all the sampling sites, and in all the species we have investigated.

Some concern has arisen regarding public health as the fishes and giant clams where pesticides residues were always detected are currently used as food species among the local populations. Though the average level of Kepone in coral reefs fishes from Polynesia looks at first sight quite close to that found in samples from French West Indies, the residues level seems well lower for the most contaminated samples. Nevertheless, some serious concerns are still pending regarding the health risks related to long-term exposures to this level of residues in fish food from Polynesia. Accordingly, it seems to us that a more in depth survey is requested and needs to be urgently launched in order to keep more detailed data on the accurate level of contamination of reef organisms in French Polynesia, particularly those which could be ingested by the local populations.

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