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## Abstract

Metallic and organic pollutants constitute a serious threat for coral reef ecosystems, potentially affecting a great number of species interacting within complex trophodynamic processes. Pesticides, PCBs and trace elements were measured on coral reef communities of three Pacific islands (Moorea, Wallis and New Caledonia) in relation with $\delta^{15}N$ values, a proxy of trophic level. Several potential sources of organic matter, benthic invertebrates and fish belonging to various trophic strategies were sampled at each island. Wallis and New Caledonia displayed, respectively, the highest concentrations of pesticides and trace elements. In the three islands, most trace element concentrations (Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, and V) decreased when $\delta^{15}N$ was rising (i.e. bioreduction), whereas Hg and Se biomagnified with increasing $\delta^{15}N$ values. Only few trace elements in some islands did not show any significant trend in relation with $\delta^{15}N$ (i.e., Ag in New Caledonia, Zn in Wallis and As plus Zn in Moorea). PCBs concentrations showed a significant bioreduction in New Caledonia and in Moorea, but a significant biomagnification in Wallis. Aldrin and heptachlor were the only pesticides to show a similar significant bioreduction in the three islands. Other pesticides, such as chlordecone, diazinon, endosulfan I and II, heptachlor-epoxide A and B, lindane and *pp'*-DDE displayed contrasted patterns (e.g. chlordecone significantly biomagnified in New Caledonia, significantly bioreduced in Wallis and did not displayed any significant trend in Moorea). Finally, for unclear reasons, Moorea displayed only negative significant correlations between $\delta^{15}N$ and all pesticides (except *pp'*-DDT). Our results highlight that trophic level, here assessed through $\delta^{15}N$ values, is a good predictor of metallic trace elements biomagnification or bioreduction in coral reef organisms. However, at large spatial scale, trophic level relevance to predict pesticides and PCBs biomagnification or bioreduction should be considered with caution and studied in close relation with local characteristics.

**Keywords:** nitrogen stable isotope, PCBs, pesticides, trace elements, bioreduction, biomagnification, Serranidae

## Introduction

At the global scale, coral reef ecosystems are increasingly subject to natural and anthropogenic threats (Graham et al., 2008, 2013, 2014; Riegl and Purkis, 2015; Hoey et al., 2016; Mumby et al., 2016). Among human-induced disturbances, chemical pollution is likely one of the most widespread. On-going climate changes might generate an increase of terrigenous runoffs onto coral reefs due to higher rainfalls during cyclonic events for instance. A recent work evidenced a significant trend to a slowdown of tropical-cyclone translation speed, thus generating increases in local rainfall totals in combination with increased tropical-cyclone rain rates (Kossin, 2018). Because runoff constitute a major pathway of contaminants transfer toward marine waters, there is a critical need to understand current levels of contamination of coral reef organisms. Polychlorinated biphenyls (PCBs), pesticides (both belong to the persistent organic pollutants: POPs) and trace elements are among the most potentially pernicious pollutants (Richardson, 1995; Burke et al., 2011). Several studies described the effects of various contaminants on the biology or physiology of some coral reef organisms. Biscéré et al. (2017) evidenced the impact of nickel affecting scleractinian growth, whereas Nystrom et al. (2001) quantified the role of cooper on the coral metabolism. Mercury negatively affect fish body condition (Baumann et al., 2017). Several pesticides may inhibit or limit photosynthesis processes and can favour coral bleaching through the coral symbionts expulsion (Jones and Kerswell, 2003; Negri et al., 2005; Shaw et al., 2012). In addition, the physicochemical properties of these contaminants, such as lipophilicity (that mostly concern organic contaminants), resistance to physical, chemical and biological degradation allow them to be transported over long distances and to bioaccumulate in marine organisms (Phillips, 1995), and/or biomagnify along the food webs (Porte and Albaiges, 1993; Rainbow, 2007; Kelly et al., 2007). These pollutants, transported by oceanic currents and/or river inputs to coral reefs, are likely to integrate into food webs and to bioaccumulate in exploited edible fish and

shellfish (e.g., Hédouin et al., 2009; Metian et al., 2013). Burke et al. (2011) estimated that various types of pollution negatively impact ~20-25% of the coral reefs worldwide. As protein inputs for human populations widely depend on coastal fisheries in many coral reefs, a better assessment of the concentrations in POPs and trace elements in these complex ecosystems is essential. Food webs are the main pathways of organic matter transfer within coral reefs (Arias-Gonzalez et al., 1997; Wyatt et al., 2012; Dromard et al., 2013; Letourneur et al., 2013; McMahon et al., 2016; Briand et al., 2016). However, studying trophic relationships on coral reefs is facing the complexity of interactions related to a great diversity of feeding strategies and multiple primary producers that characterize coral reef ecosystems (Briand et al., 2015, 2016). Thus, it remains challenging to evaluate such complex trophodynamics processes, and consequently the relationships between pathways of organic matter transfer and then contaminations by various pollutants through food webs.

Regarding contaminants in coral reef ecosystems, most studies focus on one (or few) type(s) of contaminant(s) or specie(s) (Rodriguez-Sierra and Jimenez, 2002; Chouvelon et al., 2009; Métian et al., 2009; Hédouin et al., 2009, 2010, 2011; Ikonomopoulou et al., 2012; Bonnet et al., 2014; Dromard et al., 2016; Ritger et al., 2018), preventing a wider interpretation. Only a few coral environment studies have tried to link the level of metallic and organic contaminations with isotopic signatures of several species from different trophic levels and/or based on contrasted sources of organic matter (Briand et al., 2014, 2018, Dromard et al., 2018). Therefore, a more comprehensive assessment of the contamination of coral reef food webs by chemical contaminants is needed. Due to the high number of coral reef taxa, the only realistic way to assess the food webs' contaminations consists in evaluation of selected species that are representing the major feeding strategies within coral reefs.

The present study focused on three Pacific islands: New Caledonia, Wallis and Moorea (French Polynesia) coral food webs, islands that are contrasted in terms of human population

and potential sources of contaminations. For a better understanding of the incorporation processes of contaminants within coral reef species from different islands, our work aimed at (i) assessing the organic and metallic contamination levels for different species and/or trophic groups, (ii) their spatial variation among islands and (iii) assessing the degree of biomagnification or bioreduction along the food chain, from organic matter sources to predator fish, and their specificity: island-specific or contaminant-specific. To answer these questions, this work focused on several trophic compartments constituting a simplified food web, i.e. from several potential sources of organic matter, various potential primary and secondary consumers and high trophic level predators' end-members, such as groupers (Serranidae). This fish family was selected for both their important ecological role as well as for their economic importance for local populations. For this approach, we have studied fourteen trace elements (i.e. metallic contaminants), fifteen PCB's congeners and sixteen pesticides.

## Material & Methods

### *Study sites*

This study focuses on three Pacific islands having contrasted human population and potential sources of contaminants: Moorea (French Polynesia, SE Pacific), Wallis (Central Pacific) and New Caledonia (SW Pacific) (Figure 1). Moorea is inhabited by ~17,000 persons. The island is crossed by perennial rivers, their watersheds are widely used for agricultural practices, mostly pineapple culture, and waste waters from inhabited areas are still poorly treated. Although sparsely populated (~ 9,000 inhabitants), Wallis undergoes anthropogenic stress due to the absence of waste management and domestic releases. In addition, many phytosanitary products are still used with little control for agricultural and domestic activities. Finally, New Caledonia has a higher population (~280,000 inhabitants) and its coral reefs are

subjected to anthropogenic inputs in relation with nickel open sky mining activities, plus inputs linked to urban development (Fichez et al., 2005; Hédouin et al., 2009). Some studies on possible impacts of contaminations on New Caledonian coastal marine species have already been conducted (Hédouin et al., 2011; Metian et al., 2009, 2013; Bonnet et al., 2014; Biscéré et al., 2017) while only scarce information exist for Moorea (Roche et al., 2011; Salvat et al., 2016), and none for Wallis. Apart from the specific case of the mining activities specific to New Caledonia, the coral reefs of Wallis and Moorea are subject to terrigenous inputs, natural and related to human activities that potentially can carry phytosanitary products and/or trace elements of different origins.

***Sampling***

Sampling was carried out during winter 2012 in Wallis (Mata'utu / Matala'a area) and New Caledonia (La Foa / Ouano area) and during summer 2013 at Moorea (Cook / Opunohu area). Seasonal differences in contaminant concentrations are most often non-significant (Briand et al., 2014, 2018) and were thus not investigated here. For each island, several potential sources of organic matter (OM) were collected, i.e. sedimentary organic matter (SOM), algal turf, seagrass and macroalgae. Sediment samples were collected with plastic gear (spoons and tweezers) firstly rinsed in an acid bath (35 mL L-1 nitric acid, 50 mL L-1 hydrochloric acid), then further rinsed with Milli-Q water. All macrophytes were rinsed with seawater on the field to avoid accumulation of detrital matter within thallus and epibionts were eliminated. ( Mainly detrital, the sedimentary organic matter (SOM) is a mixture of phytoplankton, bacteria, invertebrates and fish fecal pellets, detrital particles plus the micro-phytobenthos and the meiofauna (Volkman and Tanoue, 2002; Cresson et al., 2012).

Benthic invertebrates were also collected because they represent a trophic link between the OM sources and secondary consumers in the food web, and are also known for their ability

to accumulate trace elements (Metian et al., 2008; Hédouin et al., 2009). Sampling of invertebrates targeted molluscs (bivalves and gastropods) and crustaceans (crabs and hermit crabs) (See Table S1 for detail). Various fish species were sampled opportunistically but with an attempt towards species having contrasted feeding strategies, including those with supposed high trophic levels such as Serranidae (Table S1). As predators of commercial interest, this family constituted a potential source of contamination for local human populations.

Once collected, the samples (sediments and organisms) dedicated to trace elements were stored in hermetically sealed plastic bags, frozen at -20°C and kept frozen until being processes at the laboratory. For organic contaminant analyses, the samples were stored in clean glass containers (previously combusted at 450 ºC for 4 hours) and frozen at -20 ºC (see also below section dedicated to organic contaminants analyses). Primary producers and invertebrates were collected while snorkelling, and then stored at -20°C. The first cm of marine sediment were collected for SOM. The marine sediment was oven dried (60 °C) for a few days and the large particles (shells, stones, etc.) were subsequently removed. Most fish were captured with a small amount of non-selective ichtyocide, facilitating the collection of small species, whereas larger individuals were caught with a spear gun.

### *$\delta^{15}N$ analysis*

All sediment, animal and vegetal samples were freeze-dried. For fish as well as for invertebrates, nitrogen isotopic ratios were measured on muscle due to its low lipid content (Pinnegar and Polunin, 1999). Freeze-dried sediment, vegetal, invertebrate and fish samples were reduced to a fine powder (< 60 µm) using a mortar and pestle. For each sample, approximately 1 mg of powder was weighed into a tin microcapsule (8 mm x 5 mm) using a precision balance (ADAM PW124®, d= 0.1 mg) previously tared. To avoid sample

contamination, all glass and plastic utensils used were washed with ethanol, rinsed 3 times in deionized (Milli-Q quality) water and dried in an oven at 50°C before use. $^{15}N/^{14}N$ ratios were measured by mass spectrometry in continuous flow Delta V Advantage (Thermo Scientific®, Bremen, Germany) associated with an elemental analyser Flash EA-1112 (Thermo Scientific®, Milan, Italy). Analytical precision was less than 0.15 ‰, thanks to parallel analysis of standard reference material (atmospheric $N_2$). Isotopic values are expressed in ‰ compared to the international standard reference materials such as: $\delta^{15}N = [(R_{sample} / R_{standard} - 1)] \times 1000$, where R is the ratio corresponding to $^{15}N/^{14}N$.

***Organic contaminants analysis (PCBs and pesticides)***

Organic pollutant concentrations were measured for all primary producers and SOM, except for New Caledonia where only algal turf and SOM were analysed. The tissues analysed were muscles for all fish and invertebrates. For fish, analyses were mostly run on Serranidae, plus several additional species depending on sites (Table S1). The samples were frozen and stored at -20°C until the analysis. Glassware was cleaned before use with the detergent TFD4 dec FT30, dried at 200 °C for at least 24 h, and rinsed at least twice with the solvent (hexane) before use. All traces of organochlorinated compounds were removed from the extraction cartridges (22 × 80 mm, Schleicher & Schull) by conducting a blank pre-extraction for 12 h under normal conditions.

Approximately 1 g of freeze-dried samples were analysed, except for sediment samples for which between 3 g and 6 g of material were used. Freeze-dried samples were soxhlet extracted with hexane and the extracts underwent liquid chromatography on a column containing silica gel and alumina following the procedures described by Dierking et al., 2009.

Precisions on extraction, quantitative analyses and control of quality are given in Table S2 and associated text.

Fifteen PCB's congeners were analysed: IUPAC (International Union of Pure and Applied Chemistry) numbers 18, 20, 28, 31, 44, 52, 101, 105, 118, 138, 149, 153, 170, 180 and 194. The International Council for the Exploration of the Sea (ICES) designated seven of these congeners (28, 52, 101, 118, 138, 153 and 180) as marine PCB contamination indicators. Quantification was performed by gas chromatography (Agilent Technologies HP6890®) equipped with an electron capture detector at 300 °C and an automatic injector in the column (DB5 J&W column 60 x 0.32 i.d x 0.25 µm), with helium as carrier gas. During injection, the temperature is 60 °C, then it increases by 10 °C per minute until 160 °C, then 25 °C per minute until 280 °C. The detection limit is 0.01 ng. $g^{-1}$dry mass.

Sixteen pesticides were also analysed (aldrin, atrazine, chlordecone, diazinon, dieldrin, endosulfan I and II, endrin, glyphosate, heptachlor, heptachlor epoxide A and B, lindane, linuron, malathion, *pp'*-DDT, *pp'*-DDE and *pp'*-DDD ; the confirmation of the presence of *pp'*-DDT was made by dehydrochlorination with alcoholic potassium hydroxide of selected samples). These pesticides were selected for their environmental persistence and toxicity, inducing a potential threat to ecosystems as well as for human health. Atrazine, linuron and malathion were analysed as described by Pang et al. (2006), while glyphosate was quantified by liquid chromatography. The other compounds were quantified by gas chromatography (Agilent Technologies 6890N®) equipped with an injector without division, coupled to mass spectrometry. The column used was the same as the PCB (helium as carrier gas), but programmed to increase by 25 °C/min between 50 °C and 100 °C, then 5 °C/min between 100 °C and 280 °C. The detection limits (DL, in ng. $g^{-1}$ dry mass) are 0.01 ng. $g^{-1}$ for chlordecone and heptachlor epoxide A and B, 0.02 ng. $g^{-1}$ for linuron and malathion. For aldrin, diazinon, endosulfan II, endrin, heptachlor, lindane, *pp'*-DDT, *pp'*-DDE and *pp'*-DDD, the DL is 0.1

ng.g$^{-1}$, 0.2 ng.g$^{-1}$ for atrazine, dieldrin and endosulfan I. Finally, the DL for glyphosate is 1 ng.g$^{-1}$. Overall, 218 analysis (60 samples of sources of OM, 39 invertebrates and 119 fish; Table S1) of PCBs and pesticides were processed.

***Metallic contaminants analysis***

Fourteen trace elements were analysed: silver (Ag), arsenic (As), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), iron (Fe), mercury (Hg), manganese (Mn), nickel (Ni), lead (Pb), selenium (Se), vanadium (V) and zinc (Zn). Their concentrations were measured in the various OM sources of each island, as well as for several invertebrates and fish (muscle tissues, as for organic contaminants). Except for Hg, analysis of these elements was performed by Induced Coupled Plasma Atomic Emission Spectrometry (ICP-AES Vista-Pro Varian®) or coupled with mass spectrometry (ICP-MS II Series Thermo Fisher Scientific®) as described in Kojadinovic et al. (2011). Total Hg concentrations were quantified on samples ranging from 3 to 100 mg of dry mass. Analyses were performed by atomic absorption spectrometry using an Advanced Mercury Analyzer (AMA 254 ALTEC®) according to the procedure described by Bustamante et al. (2006). A quality control program was performed, including treatment and analysis of Certified Reference Material (CRMs were dogfish liver DOLT-4 and lobster hepatopancreas TORT-2; National Research Council, Canada) and blanks along with the samples. Recoveries for theCRM varied from 83% to 115%. The concentrations of trace elements are given in $\mu g.\, g^{-1}$ dry mass. The detection limit (in $\mu g.\, g^{-1}$ dry mass) are 0.005 $\mu g.\, g^{-1}$ (Hg), 0.015 $\mu g.\, g^{-1}$ (Ag, Cd), 0.02 $\mu g.\, g^{-1}$ (Cr, Co, Pb), 0.03 $\mu g.\, g^{-1}$ (Ni), 0.08 $\mu g.\, g^{-1}$ (Mn), 0.1 $\mu g.\, g^{-1}$ (Cu, Se), 0.2 $\mu g.\, g^{-1}$ (As), 0.3 $\mu g.\, g^{-1}$ (V) and 3.3 $\mu g.\, g^{-1}$ (Fe, Zn). Overall, 354 measures of metallic contaminants (66 samples of sources of OM, 60 invertebrates and 228 fish; Table S1) were processed.

***Data processing***

Fish species have been assigned into four trophic groups: herbivores (eating mainly macrophytes), omnivores (eating both animal and plant material), micro-carnivores (eating small prey) and macro-carnivores (eating larger prey), according to data on the diets of coral reef fish (Kulbicki et al., 2005) and taking into account the size of the individuals. Biomagnification potential of both POPs and trace elements was estimated for the various organisms sampled by a trophic magnification factor (TMF), quantified with the equation relating contaminant concentrations and trophic position (assessed through $\delta^{15}N$ proxy) (Nfon et al., 2009):

$$Log_{10}[contaminant] = a + b \times \delta^{15}N$$

the linear regression slope (b) represents the concentration variation per changing unit of trophic position over the trophic network, and (a) is a constant depending on the background contaminant concentration (Rolff et al., 1993). Given that this process generally involves at least three trophic levels (Wang, 2002), accumulation levels measured in species belonging to the three sampled categories of organisms trophic compartments, i.e. primary producers, invertebrates and fish, were taken into account. The slope (b), also called 'biomagnification power' (Nfon et al., 2009), can be used to calculate the trophic magnification factor (TMF) of an element via the formula:

$$TMF = 10^{b}$$

A TMF value higher than 1 indicates an accumulation of contaminants with increasing $\delta^{15}N$ values (proxy of trophic level), i.e. biomagnification, while a value lower than 1 implies decreasing concentration through food chain, i.e. bioreduction or biodiminution (Nfon et al., 2009).

To characterize the PCB contamination, several indexes were used. First, the concentrations of all analysed congeners have been summed, taking the ∑PCB notation. The seven ICES congeners (C28, C52, C101, C118, C138, C153 and C180) have also been summed (∑ICES). The global PCB contamination ($PCB_{tot}$) was calculated in two ways. The first, noted $PCB_{tot1}$, was proposed by Annema et al. (1995) considering that congeners C28, C52, C101, C138, C153 and C180 represent about 20 % of the total concentration of PCB in commercial mixtures:

$$PCB_{tot1} = (C28+C52+C101+C138+C153+C180) / 0.20$$

The second formula, noted $PCB_{tot2}$, is based on the components of the commercial mixture DP6 (or Arochlor 1260), this product being the major contamination source in most environments. Congeners predominantly present in this mixture are C118, C138, C153 and C180. These represent 41 % of the total amount of PCBs (Wafo et al., 2012).

$$PCB_{tot2} = (C118+ C138+C153+C180) / 0.41$$

For the pesticides, contamination can be characterized by the total concentration in DDT (∑DDT), the sum of *pp'*-DDD, *pp'*-DDE and *pp'*-DDT. Calculating the ratio DDT / ΣDDT is

an indicator of the seniority of the intake DDT in the environment (a ratio close to 0 indicates an ancient contribution; a ratio close to 1 indicates a recent contamination, Wafo et al. 2012). The sum of pesticides analyzed for each organism (∑Pest) was also calculated. The ratio of the sum of pesticides on the sum of PCBs (ΣPest / ΣPCB), proposed by de Mora *et al.* (2004), gives an indication of the relative importance of organic contamination from agricultural or industrial origin. A ratio higher than 1 indicates prevalence of agricultural sources whereas a ratio less than 1 indicates prevalence of industrial sources.

For trace elements, only the average concentrations of each element were compared. When concentrations were below the detection limit, half of the detection limit was used.

***Statistical analyses***

To compare mean concentrations of contaminants between islands, a normality test Shapiro-Wilk was first applied to determine the normality of distribution of data. To test the homogeneity of variance, the Bartlett's test was used when more than two groups normally distributed were compared. If normality was not respected, a Bartlett permutation test was performed. In the case where only two normally distributed groups were compared, Fisher-Snedecor's test was used. We performed ANOVAs by permutation, when the variances were homogeneous, or the Kruskal-Wallis test in the other cases. When the test was significant, a pairwise permutation t-test was performed after the ANOVA by permutation, or a Mann-Whitney pairwise permutation test for the Kruskal-Wallis test. All statistical analyzes were performed using R (R development team, 2018) and significance thresholds set at $\alpha = 0.05$.

# Results

### *Inter-islands distribution of contaminants*

*Trace elements*

All metallic contaminants measured were found in all categories of sources of OM, invertebrates and fish trophic groups and have shown numerous significant inter-island differences (Table 1, Table S3). However, some trace elements displayed relatively few significant inter-island differences such as Ag ("only" for bivalves, omnivores fish and macrocarnivore fish), As ("only" for SOM, algal turf and seagrass, Figure 2) and Ni ("only" for macroalgae, seagrass and microcarnivores fish) (Table 1, Table S3). On the contrary, several other trace elements displayed much more significant inter-island differences such as Co, Cr, Fe, Hg and Mn (see Figure 2 for the example of Fe). Mean concentration also strongly varied according to trace elements; for instance Fe was, overall, the most concentrated trace element at all islands for all categories of OM sources and invertebrates, followed by Mn and to a lower extent Ni (Table 1) whereas Zn was the most concentrated trace element in the different fish trophic groups and followed to a lower extent by As.

Categories of sources of OM and organisms displayed different patterns in trace elements concentrations, with all sources of OM having numerous significant inter-island differences, bivalves and herbivores fish having few significant inter-island differences (Table 1, Table S3, Figure 2). Wallis displayed the lowest values in trace element concentrations, except for a few cases as among which Pb for SOM, As for algal turf and Mn for macrocarnivore fish (Table 1, Table S3). Conversely, Moorea had significantly higher concentrations of several elements (Cr, Fe, Mn or Zn) for SOM, algal turf, micro- and macrocarnivore fish than the two other islands. New Caledonia had significantly higher concentrations for Co and Ni for macroalgae, seagrass, invertebrates and omnivore fish (Table 1, Table S3).

*Polychlorinated biphenyls*

As for metallic contaminants, PCBs were found in all categories of sources of OM, invertebrates and fish trophic groups and have shown almost systematically significant inter-island differences (Table 2, Table S4, see also Figure 3 for the example of ∑PCBs). The different indexes used for assessing PCB concentrations gave similar results, but in two cases (bivalves and macrocarnivore fish), their complementarity appeared necessary to detect an inter-islands difference (Table 2, Table S4). In New Caledonia, the highest PCB concentrations were found in algal turf and, to a lower extent, in bivalves and macrocarnivore fish (Table 2). In Wallis, the highest values were found in invertebrates and microcarnivore fish whereas the highest PCB concentrations concerned algal turf, macroalgae and seagrass in Moorea (Table 2). In all islands, PCB concentrations in SOM were low. For all sources of OM, mean PCB concentrations were significantly higher in Moorea, except for algal turf that did not significantly differ from New Caledonia (Table 2, Table S4). For invertebrates and fish trophic groups, the trend clearly changed as mean PCB concentrations were significantly higher in Wallis than in the other islands, except for omnivore fish that did not significantly differ from Moorea (Table 2, Table S4).

*Pesticides*

As for PCBs, pesticides were found in all categories of sources of OM, invertebrates and fish trophic groups and have shown numerous significant inter-island differences (Table 2, Table S4). Overall, the sum of pesticides (except glyphosate) was higher in Moorea for two sources of OM (SOM and seagrass) and higher in Wallis for invertebrates and fish trophic groups (Table 2, Table S4). However, one can notice several other trends for each pesticide. For instance, chlordecone was significantly more concentrated in omnivore and macrocarnivore fish in New Caledonia than those of Wallis and Moorea. Heptachlor is another example, when

significant difference were recorded between islands, this pesticide was more concentrated in New Caledonia (SOM, bivalves) (Table 2, Table S4). Some pesticides are characterised by few inter-island differences, such as diazinon and endosulfan I (Figure 3), whereas other have shown numerous inter-island significant differences (chlordecone, lindane, *pp'*-DDD) (Table 2, Table S4, Figure 3).

The current EU regulation on pesticides imposes maximal limits of residues (MLR) for the most part of food items (vegetables and animals) with the notable exception of fishes. However, in our study we can consider as MLR the one who applies to most of the animal-derived food items taken into account by the regulations, i.e. 10 $\mu g.kg^{-1}$ by pesticide (Commission Européenne 2005). Thus for most pesticides, the mean concentrations remained relatively low, although some cases displayed moderate/high values such as atrazine in invertebrates and fish in Wallis, malathion in sources of OM in Moorea, dieldrin in invertebrates in New Caledonia (Table 2). The most atypical case was the glyphosate in New Caledonia which was highly concentrated but as it was measured in only four compartments (algal turf, seagrass, omnivore and macrocarnivore fish) (Table 2), it remains difficult to raise a wider comparison.

*Synthetic ratios*

The mean DDT / ∑DDT ratios displayed several differences between trophic categories and/or islands, varying from 0.01 for seagrass in Moorea to 0.38 for omnivores and macrocarnivores fish in New Caledonia (Table 3). Overall, Moorea has lower mean values than Wallis and New Caledonia, except for SOM, whereas New Caledonia displayed the highest values. In all islands, DDT / ∑DDT ratios were clearly closer to 0 rather than close to 1

The mean ∑Pest / ∑PCB ratios also displayed strong differences between trophic categories and/or islands, ranging from 0.34 for macrocarnivores in Moorea to 2.34 for SOM in Wallis (Table 4). Overall, Moorea had the lowest mean values, whereas Wallis displayed the highest values. ∑Pest / ∑PCB ratios were higher than 1 in Wallis and New Caledonia and was lower than 1 in Moorea.

### *Relationships between contaminants and $\delta^{15}N$ values*

*Trace elements*

Most trace elements concentrations (Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, and V) decreased significantly with the increase of $\delta^{15}N$ values, therefore showing a bioreduction in the three islands. In contrast, Hg and Se have shown significant increases in concentrations with increased $\delta^{15}N$ values, i.e. a biomagnification, in the three islands (Table 4). The other trace elements tended to bioreduce with increasing $\delta^{15}N$ values but not in all islands: Ag did not shown any significant trend in New Caledonia, neither did Zn in Wallis and neither did As and Zn in Moorea (Table 4).

*Polychlorinated biphenyls*

The responses of PCBs concentrations to increase of $\delta^{15}N$ values depended from the islands. The four descriptors of PCBs concentrations (i.e., $PCB_{tot1}$, $PCB_{tot2}$, ΣPCB and ΣICES) have all shown a significant bioreduction in New Caledonia and in Moorea (except for $PCB_{tot1}$) (Table 4). Conversely, significant biomagnification was found in Wallis for all these PCBs descriptors.

*Pesticides*

The different pesticides displayed various patterns in relation to $\delta^{15}N$ values. Only two pesticides displayed a similar trend in the three islands, i.e. aldrin and heptachlor that have shown a significant bioreduction with increasing $\delta^{15}N$ values (Table 4). All other pesticides have shown contrasted trends. Although several pesticides did not shown any significant trend in two of the islands, there was significant one in the third island: in Wallis, atrazine significantly biomagnified; in Moorea, dieldrin, endrin, linuron and malathion significantly bioreduced, and in New Caledonia *pp'*-DDD and *pp'*-DDT significantly biomagnified (Table 4). Other pesticides have even displayed opposite trends depending on islands, such as chlordecone which displayed a significant biomagnification in New Caledonia, no significant trend in Moorea, and a slight but significant bioreduction in Wallis (Table 4). Heptachlor epoxide A and B, lindane and *pp'*-DDE also displayed opposite trends in relation to $\delta^{15}N$ values.

At island scale, patterns of response of pesticides' concentrations with $\delta^{15}N$ values also varied: Wallis displayed the most numerous non-significant trends (9 cases on 18 tested, Table 4), New Caledonia recorded the most numerous cases of biomagnification (8) whereas Moorea displayed the most numerous cases of bioreduction (13). It should be noted that only one significant case of biomagnification was found in Moorea (*pp'*-DDT) (Table 4).

## Discussion

Our study constitutes an important baseline work on organic and metallic contamination on Pacific coral reef communities. It also evidenced a generalized contamination by several pollutants among all studied compartments / trophic groups involved in coral reef food webs from OM to macrocarnivore fish (Serranidae) across three islands over a Pacific wide scale. This work has also shown that trace elements have, overall, a similar "behavior" among islands

in relation to organisms' trophic levels (i.e. biomagnification or bioreduction). In contrast POPs displayed contrasted and sometimes opposite responses according to compartments / trophic groups and/or insular systems. This is essential because it highlighted that trends of contamination may strongly differ among islands (irrespective of the concentrations themselves) and/or between trophic groups, potentially obscuring patterns if data are not investigated at local scales.

***Inter-island' variations of the chemical contaminations***

Although depending on the food web's compartments considered, it was to some extent surprising that the typical trace elements associated with the nickel mining exploitation (i.e., Co, Cr, Mn, and Ni) were not all higher in New Caledonian OM sources, invertebrates or fish. This finding remain however similar to past analysis on New Caledonian nautiluses (Bustamante et al. 2000; Pernice et al., 2009) and marine mammals (Bustamante et al., 2003, Garrigue et al. 2016). It is therefore possible that the lack of difference for these mining elements, with the exception of Co, is not uniquely related to mining activity, but to other complementary characteristics such as the geology or hydrology of the island (soil composition, erosion, etc.) (Bonnet et al., 2014). Indeed, the studied site is not directly close to watershed impacted by mining activities, suggesting that metals linked to mining activities may generate a contamination at a local coral reef scale rather than at a larger New Caledonian coral reef lagoon scale. Apart from the mining elements, some elements such as Ag, Hg or Se were more concentrated in fish and Cd in invertebrates from New Caledonia compared to Wallis and Moorea. These elements are most often linked to human activities like agriculture, industries or urban sewages (Nriagu, 1994; Callender et al., 2000; Briand et al., 2018). As New Caledonia is populated by ~ 280,000 inhabitants, hosts three large metallurgic factories as well as small

numerous industrial areas and devotes large land areas to agriculture, our results can be interpreted as a general consequence of this situation.

For organic contaminants, the highest total PCB and ∑PCB concentrations in OM sources were detected in Moorea, whereas invertebrates and fish most often reached their peak values in Wallis. A similar pattern was found for the total pesticide concentration (∑Pest), with the exception of some given pesticides which reached their highest concentration in New Caledonia. To some degree, these results confirm those of Briand et al. (2014) where marine organisms from New Caledonia appeared to have generally lower PCB and pesticide contamination than other tropical regions (see examples in Briand et al., 2014). Both ∑PCB and ∑Pest have shown highly variable values within and between fish trophic groups as well as for OM sources and invertebrates. Bioavailability of contaminants is influenced by the nature of the food ingested (Luoma et al., 2002) and the species detoxification strategies. Thus, the contaminants variability between fish trophic groups composed of different species is not surprising, especially when considering the biomagnification properties of some compounds (see below) (Kelly et al., 2007; Dromard et al., 2018). For most pesticides, the highest concentrations were found in invertebrates, without any clear pattern for filtering species *versus* other invertebrates. Surprisingly, sources of OM in Moorea had higher concentrations of pesticides than fish contrasting with data obtained in Wallis and New Caledonia where OM sources recorded the lowest concentrations.

Chlordecone levels found on all islands reveal a wide use of this insecticide although it is officially forbidden in the three islands. The remanence of this product can reach several centuries in sediments (Roche et al., 2011; Dromard et al. 2018), a situation that is potentially very worrying for marine systems because resuspension of sediments can be a continuous source of contamination rather than a source of contamination resulting from an active illegal use. It is therefore difficult to assess if it was introduced to the system before or after the

legislation forbidding its use was in place. Several other POPs might be in a similar situation. The case of PCBs and several pesticides in Wallis raise questions because the island is sparsely populated (~9,000 people) and agricultural practices do not concern large surface areas. Despite this, our results suggest that local practices could be strongly related to the use of phytosanitary products. The Wallis lagoon is much smaller than in New Caledonia and the reef lacks passes in the East and the North. Consequently, and even in windward zones, lagoonal water masses could be only weakly submitted to wind-driven currents, limiting dispersion of pollutants towards the open sea resulting in higher concentrations within the lagoon.

The few glyphosate samples analyzed (in Wallis and New Caledonia) showed an important contamination of this herbicide, likely linked to phytosanitary habits in local agricultural practices. However, the sanitary risk assessed through the $CE_{50}$ is ~240 $mg.kg^{-1}$ for most marine organisms (US-EPA, 2013). Thus our results are well under a sanitary risk threshold. Overall, concentrations of other pesticides and PCBs appeared to be relatively low and reinforce similar results found in other New Caledonian coral reefs (Briand et al. 2014). However, given the toxicity of some pollutants even at low-dose (for instance ~75-300 $ng.g^{-1}$ is a range of thresholds for ∑ICES depending on species/organ, US-EPA, 2013), these contaminations should not be neglected in particular because synergetic effects remain plausible ("cocktail effect") and their possible consequences unknown. According to the DDT / ∑DDT ratio, the last DDT inputs in the marine environment could be more recent for New Caledonia (average ratio of 0.45) than for Wallis and Moorea (a ratio of about 0.09 for these two islands) although we do not have reliable data from local authorities or importers to support this hypothesis. The mean ∑Pest / ∑PCB ratios suggest that Wallis and New Caledonia would be more influenced by organic contaminants of agricultural and/or domestic origin -a hypothesis consistent with local activities in the vicinity / watershed of the studied reef- whereas Moorea would be more subject to industrial inputs. The latter remains difficult to explain as

Moorea is not a densely urbanized island and does not host large industries except a fruit juice factory. As concentrations of most contaminants *per se* were globally low at Moorea, this ratio might reflect 'equilibrium' between two low values. This uncertainty highlights the limits of such indexes to determine precisely the sources of contaminants when they are at relatively low concentrations.

***Relationships between contaminants and trophic level***

Any organic compounds analysed in this study biomagnified along the food webs all islands pooled, but at island scale the trends were slightly different. For instance, in Moorea only decreasing concentrations of pesticides were found with trophic level rise. This remains surprising because several POPs most often biomagnify along food webs (Jarman et al., 1996; Fisk et al., 2001; Kidd et al., 2001; Bayen et al., 2005; Dromard et al., 2018). One possible explanation could be related to the potential of metabolization of poorly hydrophobic pollutants that usually do not biomagnify in fish (Martin et al., 2003; Kelly et al., 2007).

The survey results have also shown that most trace elements did not transfer up to high trophic levels' fish (Table 4), irrespective of the islands. Thus, it suggests a bioreduction process along food webs, either due to a low assimilation efficiency for these elements and/or a decrease of their bioavailability along the food chain (Autman et al., 2012; Merciai et al., 2014), that can generate a low or modest concentrations for some mining trace elements at high trophic levels (Briand et al., 2014, 2018). In general, OM sources accumulate specific trace elements (Cr, Fe, V, and Pb) without any clear distinction between macrophytes and SOM. The highest concentrations of most trace elements studied (Ag, As, Cd, Cu, Ni, Se, and Zn) were detected in invertebrates. Zn was the only element to show a disposition to accumulate preferentially in filtering organisms, suggesting either an affinity to adsorb to suspended particles in the water column or a major incorporation of this element through the dissolved pathway as reported for

tropical oysters (Hédouin et al., 2009, 2010). Since invertebrates are known for their ability to bioaccumulate and retain contaminants, especially bivalves (Metian et al., 2008; Hédouin et al., 2009), our results reinforce those mentioned by the previous publications. Hg and Se are the only trace elements showing a clear biomagnification between the OM sources and the high trophic level fish. This observation is not surprising for Hg because the biomagnification process of methylmercury being the dominant chemical form of Hg in organisms when trophic levels increase, is well known in various marine food webs (Zizek et al., 2007; Trudel and Rasmussen, 2006; Cresson et al., 2014; Baumann et al., 2017; Ritger et al., 2018; Chouvelon et al., 2018). Results of this survey confirm the importance of organic matter transfer for the accumulation of these two elements through trophic networks.

Most of the other trace elements concentration decreased with increasing trophic level. This dilution of the concentrations through trophic networks had already been observed in another food webs (ending to anguilliforms) of New Caledonia for several trace elements (Ag, Cd, Co, Cu, Mn, and Pb) (Briand et al., 2014, 2018). Because trace elements such as Co, Cr, Cu, Mn, and Ni accumulated more in macrophytes and invertebrates, it suggests that they were poorly transferred through the food web (Eisler, 2010; Metian et al., 2013) possibly due to a low assimilation efficiency of these elements by predators. This preferential concentration in macrophytes and invertebrates, rather than fish of higher trophic levels, could be linked to biological and physiological characteristics (exposure pathways, ingestion rate, excretion rate, assimilation efficiency, animal size, etc.) but also to different detoxification capacities of these organisms. Furthermore, a dilution effect of trace element concentrations can occur during growth (Phillips, 1995; Briand et al., 2014, 2018). This global analysis required the grouping of different species, yet biological factors specific to each species are known to influence the process of contaminants' bioaccumulation in marine organisms (Zizek et al., 2007). Thus, further research is needed to take into account the ecology (habitat, feeding) and the biology of

some selected species (growth rate, longevity, age (size/mass), mobility, etc.). This would allow better interpretation of the concentrations levels and the spatial variations. On the other hand, even if the contaminants were at relatively low levels and thus limiting the sanitary risk for humans, their interaction can potentially increase their dangerousness ("cocktail effect"). This aspect should be explored further in the future bearing in mind that predicted increasing climatic changes might alter a given pattern.

## Conclusions

This study provides a substantial data set on the contamination status of several sources of organic matter, invertebrates and different categories of fish constituting (simplified) food webs of Wallis, Moorea and New Caledonia coral reefs. The vast majority of organic compounds and metallic elements did not appear to biomagnify and even displayed a significant bioreduction along the food webs, with the notable exceptions of Hg and Se, and several pesticides which biomagnified locally (i.e. at island scale). Overall, contaminant concentration levels remain under safety thresholds, preventing any serious sanitary risk for human populations relying on fish. However, it remains preoccupying that a large variety of metallic and organic contaminants were so widely distributed on these fragile coral reefs ecosystems at such large scale. In addition, the possible synergetic and/or additive effects of the contaminants clearly remain an important question that merit further investigations.

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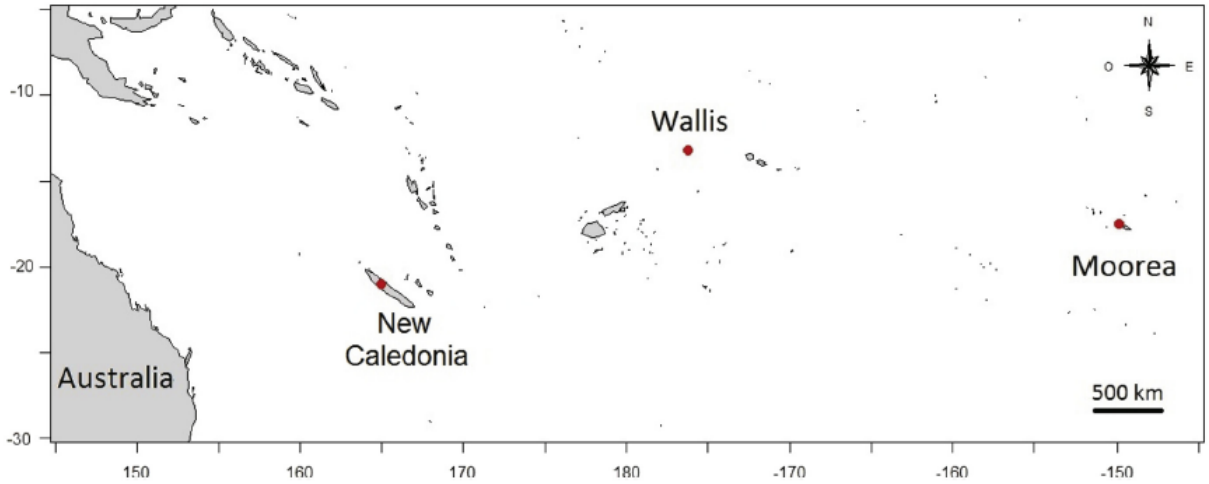


Figure 1. Location of the three studied island in the Pacific Ocean

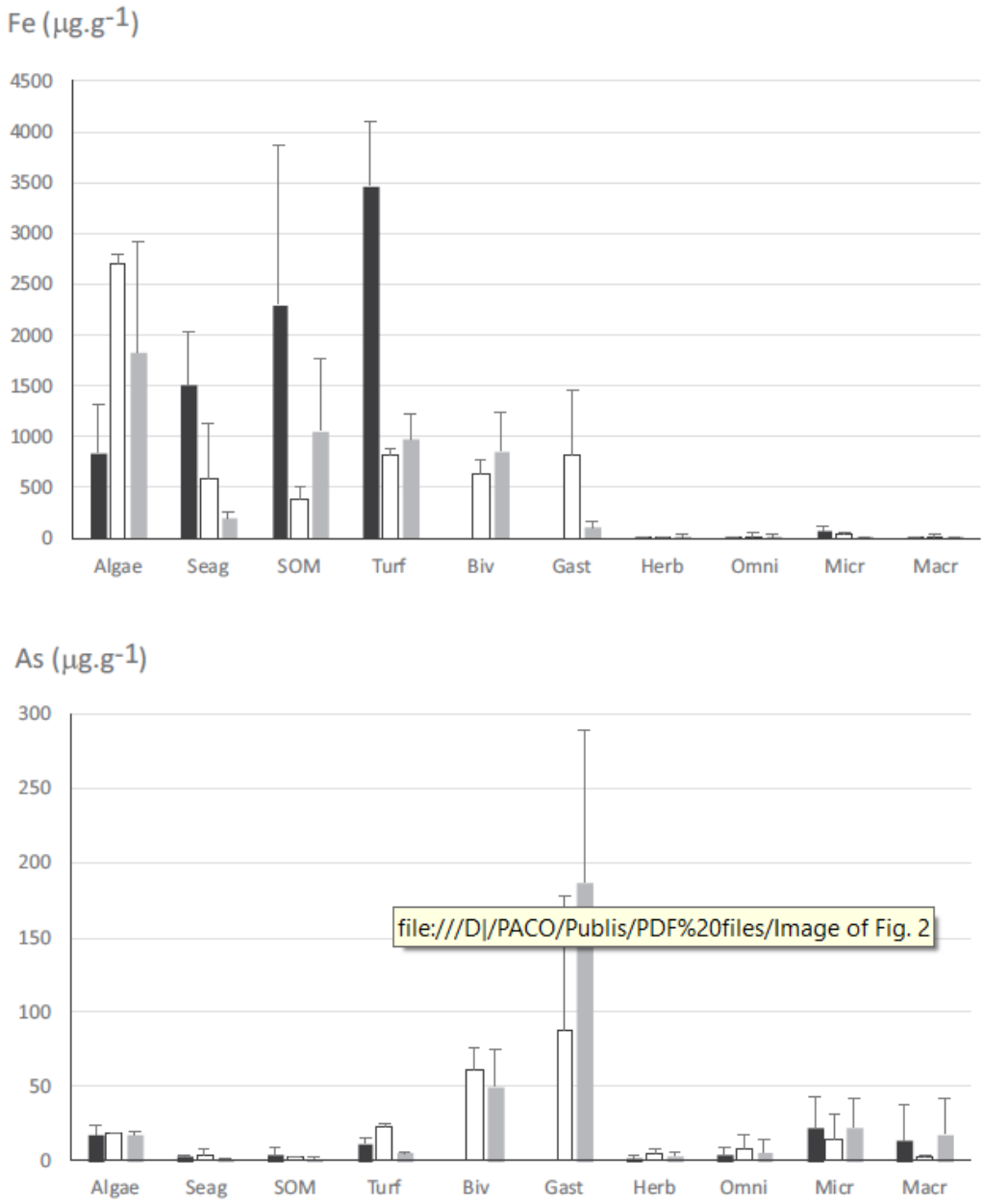


Figure 2. Example of numerous (A: Fe) and few (B: As) inter-islands differences in mean concentrations (± SD) of trace elements for various compartments, algae, seagrass (Seag), sedimentary organic matter (SOM), algal turf, bivalves (Biv), gastropods (Gast), herbivorous (Herb), omnivorous (Omni), micro-carnivorous (micr) and macro-carnivorous (Macr) fish. Black bars represent Moorea, white bars represent New Caledonia and grey bars represent Wallis. See Table 2 and Table S3 for other trace elements.

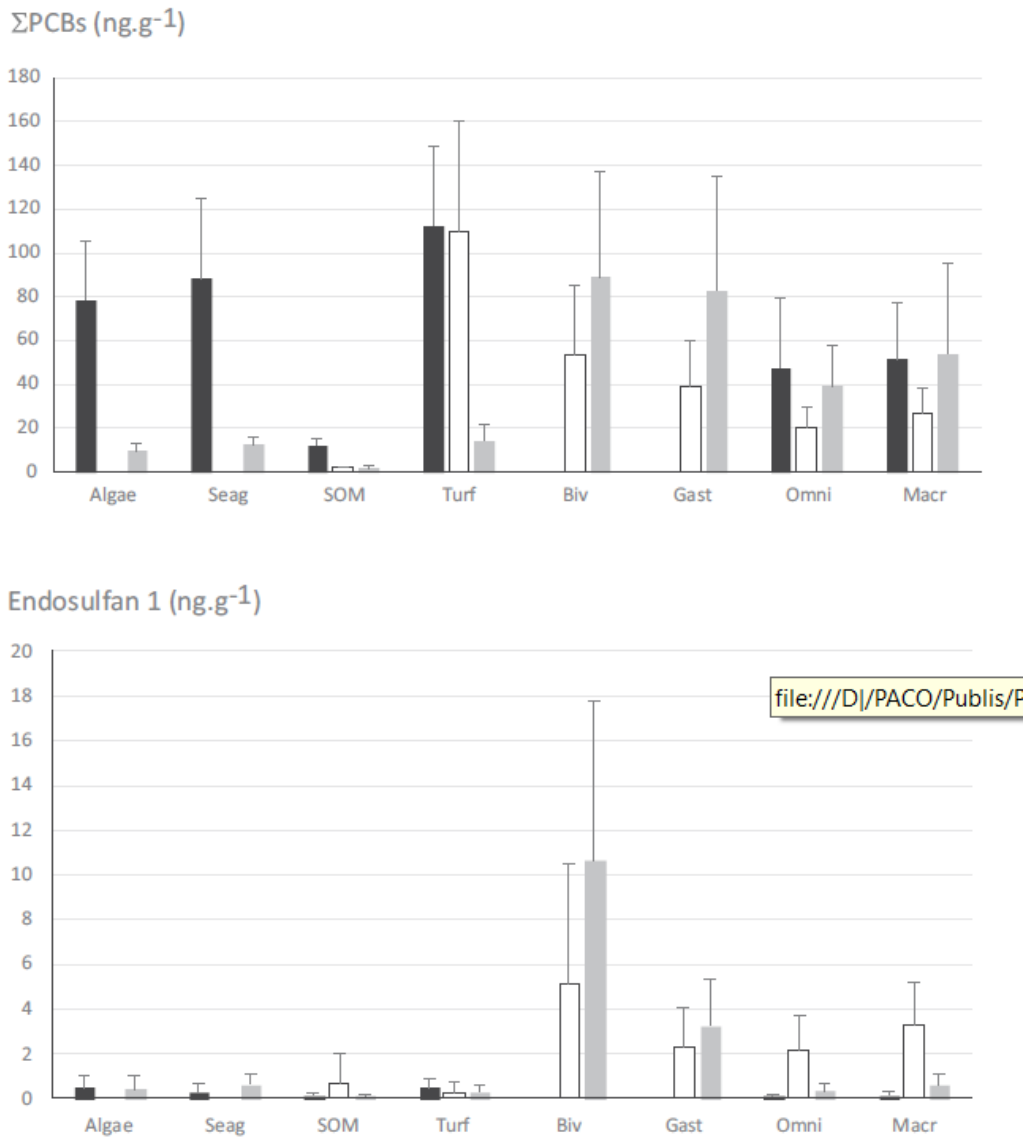


Figure 3. Example of numerous (A: ∑PCBs) and few (B: Endosulfan 1) inter-islands differences in mean concentrations (± SD) of organic contaminants for various compartments, algae, seagrass (Seag), sedimentary organic matter (SOM), algal turf, bivalves (Biv), gastropods (Gast), omnivorous (Omni) and macro-carnivorous (Macr) fish. Black bars represent Moorea, white bars represent New Caledonia and grey bars represent Wallis. See Table 3 and Table S4 for other organic contaminants.

**Table 1.** Mean concentrations in trace elements in $\mu g \cdot g^{-1}$ (±SD) in sources of organic matter, invertebrates and fish trophic groups from New Caledonia (A)Wallis (B) and Moorea (C). Standard deviations <0.001 are indicated by the sign +, nd=no data. Number of samples is given in Supplementary Table 1. Detail by species is available on request to the corresponding author.

| | Sources of organic matter | | | | Invertebrates | | | Fish | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | SOM | Algal turf | Macro-algae | Seagrass | Bivalves | Gastropods | Hermit crabs | Herbivores | Omnivores | Micro-carnivores | Macro-carnivores |
| (A) | | | | | | | | | | | |
| Ag | 0.005 (+) | 0.005 (+) | 0.015 (0.004) | 0.043 (0.042) | 0.016 (0.017) | 1.29 (2.09) | 0.058 (0.007) | 0.005 (+) | 0.22 (0.10) | 0.025 (0.042) | 0.32 (0.06) |
| As | 1.98 (1.01) | 22.98 (2.31) | 17.90 (0.24) | 3.94 (2.59) | 60.47 (14.55) | 87.11 (91.31) | 13.58 (5.10) | 4.99 (2.68) | 6.92 (9.54) | 14.39 (17.34) | 2.43 (1.50) |
| Cd | 0.007 (+) | 0.22 (0.01) | 0.21 (0.04) | 0.13 (0.04) | 5.35 (2.22) | 5.71 (4.94) | 0.63 (0.64) | 0.008 (+) | 0.019 (0.016) | 0.021 (0.04) | 0.011 (+) |
| Co | 0.82 (0.05) | 0.53 (0.01) | 2.35 (0.04) | 1.03 (0.38) | 0.49 (0.09) | 0.78 (0.43) | 1.18 (0.63) | 0.23 (0.16) | 0.08 (0.04) | 0.05 (0.06) | 0.011 (+) |
| Cr | 4.28 (0.67) | 10.92 (1.57) | 9.54 (2.19) | 2.63 (2.05) | 3.84 (1.58) | 7.15 (6.13) | 1.33 (0.09) | 0.10 (0.07) | 0.16 (0.09) | 0.07 (0.02) | 0.31 (0.16) |
| Cu | 0.16 (0.01) | 1.47 (0.02) | 2.77 (0.07) | 1.98 (1.14) | 9.46 (5.01) | 60.67 (70.59) | 36.35 (10.33) | 0.39 (0.15) | 0.87 (0.34) | 0.80 (0.57) | 0.45 (0.09) |
| Fe | 389 (123) | 807 (82) | 2690 (109) | 581 (540) | 625 (152) | 802 (651) | 140 (79.3) | 4.55 (3.60) | 15.44 (20.83) | 19.80 (24.65) | 9.87 (10.91) |
| Hg | 0.005 (+) | 0.010 (0.001) | 0.007 (+) | 0.007 (0.004) | 0.14 (0.04) | 0.30 (0.31) | 0.020 (0.005) | 0.010 (+) | 0.035 (0.011) | 0.14 (0.09) | 0.48 (0.11) |
| Mn | 13.32 (4.94) | 26.08 (0.52) | 103.06 (11.37) | 54.29 (35.58) | 11.17 (5.94) | 50.32 (44.67) | 51.30 (49.26) | 0.29 (0.14) | 0.43 (0.30) | 0.46 (0.51) | 0.28 (0.11) |
| Ni | 3.59 (0.76) | 10.69 (0.23) | 11.04 (0.36) | 8.18 (2.70) | 4.66 (1.03) | 10.42 (5.94) | 4.88 (1.42) | 0.13 (0.07) | 0.11 (0.07) | 0.097 (0.041) | 0.11 (0.03) |
| Pb | 0.09 (+) | 0.56 (0.01) | 0.89 (0.04) | 0.23 (0.16) | 0.15 (0.04) | 0.15 (0.09) | 0.04 (0.02) | 0.01 (+) | 0.022 (0.029) | 0.077 (0.029) | 0.083 (0.14) |
| Se | 0.19 (+) | 0.20 (+) | 0.19 (0.01) | 0.34 (0.04) | 4.79 (0.50) | 3.38 (2.84) | 1.06 (0.21) | 0.97 (0.32) | 1.12 (0.31) | 1.65 (0.48) | 2.20 (0.31) |
| V | 1.35 (0.95) | 10.86 (1.37) | 6.47 (0.40) | 3.25 (2.01) | 1.76 (0.57) | 5.42 (3.02) | 0.96 (0.43) | 0.52 (0.03) | 0.30 (0.28) | 0.63 (0.16) | 0.16 (0.01) |
| Zn | 1.94 (0.03) | 7.67 (0.62) | 9.19 (0.55) | 12.60 (2.85) | 968 (383) | 60.58 (36.22) | 71.14 (25.31) | 13.25 (3.62) | 15.42 (5.96) | 24.36 (20.28) | 14.02 (2.03) |
| (B) | | | | | | | | | | | |
| Ag | 0.01 (+) | 0.16 (0.17) | 0.077 (0.073) | 0.13 (0.14) | 0.52 (0.34) | 0.81 (0.69) | nd | 0.029 (0.048) | 0.013 (0.008) | 0.021 (0.022) | 0.011 (0.005) |
| As | 1.24 (0.67) | 5.42 (1.11) | 17.15 (2.00) | 0.74 (0.35) | 50.11 (24.10) | 187.45 (101.84) | nd | 2.87 (1.72) | 5.70 (9.08) | 21.56 (18.91) | 17.87 (23.59) |
| Cd | 0.01 (+) | 0.23 (0.09) | 0.15 (0.05) | 0.02 (0.01) | 2.67 (1.53) | 0.14 (0.10) | nd | 0.042 (0.056) | 0.023 (0.029) | 0.022 (0.021) | 0.011 (0.007) |
| Co | 0.43 (0.18) | 0.54 (0.08) | 1.28 (0.74) | 0.63 (0.14) | 8.33 (10.76) | 0.33 (0.02) | nd | 0.062 (0.047) | 0.030 (0.023) | 0.026 (0.022) | 0.010 (0.002) |
| Cr | 4.70 (3.13) | 6.52 (1.60) | 6.14 (4.48) | 0.78 (0.19) | 2.48 (1.23) | 0.56 (0.27) | nd | 0.26 (0.13) | 0.32 (0.27) | 0.40 (0.27) | 0.38 (0.26) |
| Cu | 0.65 (0.77) | 1.73 (0.50) | 2.03 (1.50) | 1.51 (0.61) | 12.89 (15.86) | 34.24 (35.42) | nd | 0.54 (0.29) | 0.75 (0.26) | 0.60 (0.22) | 0.38 (0.11) |
| Fe | 1051 (727) | 977 (239) | 1827 (1096) | 189 (65.9) | 862 (374) | 104 (46.8) | nd | 10.03 (8.70) | 11.78 (7.03) | 10.19 (6.02) | 6.63 (5.79) |
| Hg | 0.003 (+) | 0.014 (0.004) | 0.004 (0.002) | 0.003 (0.001) | 0.14 (0.08) | 0.03 (0.02) | nd | 0.009 (+) | 0.025 (0.01) | 0.084 (0.062) | 0.26 (0.19) |
| Mn | 15.78 (10.87) | 27.19 (3.65) | 104.46 (90.12) | 50.52 (44.19) | 856.93 (1176.78) | 2.10 (0.40) | nd | 0.63 (0.39) | 0.69 (0.38) | 0.63 (0.36) | 0.34 (0.13) |
| Ni | 3.93 (1.45) | 9.79 (2.25) | 4.71 (3.25) | 0.71 (0.35) | 12.65 (12.68) | 5.47 (10.47) | nd | 0.21 (0.18) | 0.18 (0.10) | 0.21 (0.16) | 0.059 (0.032) |
| Pb | 0.21 (0.19) | 0.52 (0.18) | 0.53 (0.44) | 0.21 (0.07) | 0.20 (0.12) | 0.07 (0.04) | nd | 0.050 (0.070) | 0.019 (0.015) | 0.022 (0.023) | 0.022 (0.030) |
| Se | 0.07 (0.04) | 0.55 (0.20) | 1.19 (1.10) | 0.06 (0.02) | 5.58 (3.08) | 5.25 (3.13) | nd | 0.85 (0.28) | 0.89 (0.32) | 1.71 (0.48) | 2.62 (0.44) |
| V | 3.99 (3.08) | 10.03 (4.95) | 4.79 (3.80) | 0.22 (0.05) | 3.23 (3.53) | 0.19 (0.01) | nd | 0.57 (0.96) | 0.27 (0.17) | 0.43 (0.45) | 0.19 (0.006) |
| Zn | 3.04 (1.83) | 12.76 (6.67) | 10.98 (8.90) | 12.77 (6.39) | 480 (339) | 48.71 (9.12) | nd | 14.79 (7.36) | 16.41 (5.51) | 17.22 (7.61) | 17.71 (4.58) |
| (C) | | | | | | | | | | | |
| Ag | 0.015 (0.012) | 0.05 (0.02) | 0.02 (0.01) | 0.014 (0.004) | nd | nd | nd | 0.010 (+) | 0.010 (+) | 0.021 (0.015) | 0.010 (+) |
| As | 4.66 (3.20) | 11.59 (3.93) | 17.18 (6.30) | 2.65 (0.74) | nd | nd | nd | 1.83 (1.43) | 3.57 (3.08) | 21.17 (20.44) | 13.57 (23.43) |
| Cd | 0.75 (1.81) | 0.14 (0.04) | 0.17 (0.15) | 0.03 (0.02) | nd | nd | nd | 0.010 (+) | 0.014 (0.011) | 0.072 (0.089) | 0.019 (0.025) |
| Co | 0.79 (0.39) | 0.99 (0.25) | 0.68 (0.25) | 0.38 (0.13) | nd | nd | nd | 0.031 (0.036) | 0.025 (0.13) | 0.077 (0.071) | 0.010 (+) |
| Cr | 7.60 (5.04) | 12.86 (1.70) | 2.67 (1.46) | 5.06 (1.74) | nd | nd | nd | 0.15 (0.05) | 0.41 (0.31) | 0.56 (1.18) | 0.26 (0.28) |
| Cu | 2.79 (5.78) | 2.95 (0.48) | 0.63 (0.07) | 0.64 (0.30) | nd | nd | nd | 0.37 (0.05) | 0.72 (0.19) | 2.01 (2.17) | 0.36 (0.14) |
| Fe | 2296 (1572) | 3468 (625) | 843 (470) | 1510 (524) | nd | nd | nd | 7.87 (2.07) | 13.78 (4.81) | 63.50 (51.07) | 6.23 (3.47) |
| Hg | 0.004 (0.001) | 0.008 (0.001) | 0.003 (0.001) | 0.004 (0.001) | nd | nd | nd | 0.008 (0.005) | 0.018 (0.008) | 0.072 (0.040) | 0.60 (0.43) |
| Mn | 76.68 (34.51) | 75.40 (26.51) | 34.04 (5.29) | 27.81 (12.99) | nd | nd | nd | 0.55 (0.16) | 1.25 (2.21) | 5.80 (6.16) | 0.16 (0.08) |

**Table 1.** Continued.

| | Sources of organic matter | | | | Invertebrates | | | Fish | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | SOM | Algal turf | Macro-algae | Seagrass | Bivalves | Gastropods | Hermit crabs | Herbivores | Omnivores | Micro-carnivores | Macro-carnivores |
| Ni | 5.54 (2.60) | 9.38 (0.69) | 2.59 (1.06) | 3.27 (1.10) | nd | nd | nd | 0.043 (0.039) | 0.12 (0.11) | 0.55 (0.62) | 0.078 (0.14) |
| Pb | 0.51 (0.44) | 1.27 (0.22) | 0.20 (0.10) | 0.43 (0.21) | nd | nd | nd | 0.022 (0.011) | 0.025 (0.024) | 0.039 (0.038) | 0.013 (0.009) |
| Se | 1.07 (2.11) | 0.92 (0.20) | 0.18 (0.03) | 0.21 (0.05) | nd | nd | nd | 0.44 (0.24) | 0.74 (0.33) | 2.19 (0.53) | 2.69 (0.54) |
| V | 5.18 (3.30) | 22.24 (3.70) | 2.29 (1.42) | 6.97 (1.77) | nd | nd | nd | 0.21 (0.02) | 0.19 (0.01) | 4.75 (12.00) | 0.19 (0.01) |
| Zn | 41.20 (85.82) | 17.96 (2.39) | 8.49 (1.17) | 17.14 (9.46) | nd | nd | nd | 13.82 (2.42) | 17.77 (4.46) | 36.74 (24.03) | 15.25 (3.34) |

**Table 2.** Mean concentrations in PCB and pesticides in $ng \cdot g^{-1}$ (±SD) in sources of organic matter, invertebrates and fish trophic groups fromNew Caledonia (A)Wallis (B) and Moorea (C). Standard deviations <0.001 are indicated by the sign +, nd= no data. Number of samples is given in Supplementary Table 1. Detail by species is available on request to the corresponding author.

| | Sources of organic matter | | | | Invertebrates | | Fish | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | SOM | Algal turf | Macro-algae | Seagrass | Bivalves | Gastropods | Herbivores | Omnivores | Micro-carnivores | Macro-carnivores |
| (A) | | | | | | | | | | |
| PCB ($ng \cdot g^{-1}$ dry weight) | | | | | | | | | | |
| $PCB_{tot1}$ | 4.74 (0.97) | 343.45 (91.25) | nd | nd | 111.74 (68.38) | 77.64 (46.44) | nd | 64.22 (25.80) | nd | 80.13 (41.64) |
| $PCB_{tot2}$ | 2.45 (0.50) | 140.47 (80.45) | nd | nd | 50.03 (30.75) | 36.58 (24.01) | nd | 26.81 (13.29) | nd | 32.01 (17.05) |
| ΣPCB | 2.24 (0.28) | 109.56 (50.61) | nd | nd | 53.67 (31.73) | 39.27 (20.96) | nd | 20.20 (9.11) | nd | 24.82 (11.89) |
| ΣICES | 1.33 (0.24) | 80.48 (44.32) | nd | Nd | 28.94 (16.94) | 21.31 (13.07) | nd | 15.36 (7.14) | nd | 19.69 (9.97) |
| Pesticides ($ng \cdot g^{-1}$ dry weight) | | | | | | | | | | |
| Aldrin | 0.05 (+) | 0.33 (0.69) | nd | nd | 1.29 (0.81) | 2.14 (1.33) | nd | 0.16 (0.14) | nd | 0.19 (0.12) |
| Atrazine | Nd | nd | nd | nd | nd | nd | nd | 1.55 (0.71) | nd | 1.95 (0.61) |
| Chlordecone | 0.053 (0.012) | 1.20 (0.51) | nd | nd | 0.62 (0.30) | 0.86 (0.65) | nd | 9.52 (3.05) | nd | 11.37 (4.27) |
| Diazinon | 0.11 (0.08) | 1.49 (1.60) | nd | nd | 5.88 (3.72) | 5.97 (3.19) | nd | 2.70 (1.59) | nd | 4.11 (2.15) |
| Dieldrin | 0.055 (0.057) | 1.43 (3.11) | nd | nd | 8.04 (3.46) | 8.20 (4.71) | nd | 0.58 (0.40) | nd | 0.79 (0.55) |
| Endosulfan I | 0.65 (1.38) | 0.28 (0.43) | nd | nd | 5.14 (5.29) | 2.31 (1.76) | nd | 2.14 (1.48) | nd | 3.01 (1.92) |
| Endosulfan II | 0.092 (0.050) | 1.67 (2.09) | nd | nd | 0.83 (1.22) | 10.80 (10.94) | nd | 2.32 (1.41) | nd | 2.31 (1.17) |
| Endrin | 0.60 (0.50) | 2.15 (2.28) | nd | nd | 2.98 (2.68) | 4.44 (3.55) | nd | 0.55 (0.58) | nd | 0.40 (0.31) |
| Glyphosate | Nd | 926.6 (809.9) | nd | 696.2 (612.8) | nd | nd | nd | 629.3 (318.8) | nd | 710.9 (678.4) |
| Heptachlor | 0.26 (0.10) | 1.03 (0.57) | nd | nd | 7.12 (3.96) | 4.35 (2.62) | nd | 0.90 (0.80) | nd | 0.97 (0.66) |
| Heptachlor-epoxide A | 0.011 (0.014) | 0.50 (0.68) | nd | nd | 0.005 (+) | 0.37 (1.09) | nd | 1.35 (0.91) | nd | 1.41 (1.01) |
| Heptachlor-epoxide B | 0.011 (0.013) | 0.01 (+) | nd | nd | 0.71 (1.72) | 0.51 (1.07) | nd | 0.33 (0.24) | nd | 0.51 (0.30) |
| Lindane | 0.015 (0.05) | 0.24 (0.14) | nd | nd | 0.63 (0.33) | 0.72 (0.30) | nd | 0.19 (0.12) | nd | 0.36 (0.21) |
| Linuron | Nd | nd | nd | nd | nd | nd | nd | 0.87 (0.60) | nd | 0.95 (0.50) |
| Malathion | Nd | nd | nd | nd | nd | nd | nd | 1.93 (0.75) | nd | 2.38 (0.38) |
| *pp'*-DDE | 0.05 (+) | 0.05 (+) | nd | nd | 0.05 (+) | 0.05 (+) | nd | 0.78 (0.65) | nd | 0.58 (0.31) |
| *pp'*-DDD | 0.05 (+) | 0.05 (+) | nd | nd | 0.05 (+) | 0.05 (+) | nd | 0.56 (0.40) | nd | 0.76 (0.50) |
| *pp'*-DDT | 0.05 (+) | 0.05 (+) | nd | nd | 0.05 (+) | 0.05 (+) | nd | 1.01 (0.90) | nd | 1.05 (0.78) |
| ΣPest[4] | 2.06 (1.88) | 10.46 (7.93) | nd | nd | 33.40 (11.11) | 40.82 (20.14) | nd | 27.44 (8.43) | nd | 31.11 (10.51) |
| (B) | | | | | | | | | | |
| PCB ($ng \cdot g^{-1}$ dry weight) | | | | | | | | | | |
| $PCB_{tot1}$ | 3.15 (2.51) | 47.33 (29.37) | 27.41 (9.27) | 31.31 (10.65) | 281.75 (172.59) | 255.83 (139.39) | 109.70 (14.78) | 107.40 (45.62) | 304.09 (172.14) | 154.51 (104.82) |
| $PCB_{tot2}$ | 2.14 (1.39) | 21.13 (12.93) | 9.61 (4.54) | 13.49 (3.94) | 111.95 (69.89) | 103.34 (74.80) | 31.39 (11.55) | 44.12 (20.82) | 135.50 (122.76) | 64.17 (66.19) |
| ΣPCB | 1.64 1.03) | 14.29 (7.36) | 9.67 (3.07) | 12.62 (3.00) | 89.04 (48.21) | 83.00 (52.18) | 35.13 (3.74) | 39.34 (18.13) | 92.96 (76.04) | 53.98 (41.49) |
| ΣICES | 1.23 (0.87) | 10.96 (5.32) | 6.88 (2.55) | 8.72 (2.48) | 63.36 (36.34) | 58.50 (38.57) | 24.21 (3.24) | 25.57 (11.31) | 68.71 (59.64) | 36.51 (32.78) |
| Pesticides ($ng \cdot g^{-1}$ dry weight) | | | | | | | | | | |
| Aldrin | 0.06 (0.03) | 0.29 (0.08) | 0.31 (0.27) | 0.50 (0.44) | 0.096 (0.055) | 0.09 (0.03) | 0.07 (0.03) | 0.07 (0.03) | 0.09 (0.02) | 0.09 (0.11) |
| Atrazine | 0.23 (0.19) | 2.41 (1.68) | 2.86 (1.12) | 2.93 (1.16) | 38.99 (18.07) | 47.82 (20.25) | 35.47 (6.58) | 32.47 (21.89) | 44.01 (44.38) | 31.93 (20.30) |
| Chlordecone | 0.37 (0.14) | 13.69 (10.69) | 5.06 (1.13) | 5.48 (1.09) | 9.78 (4.78) | 5.79 (1.18) | 4.06 (1.83) | 5.86 (2.31) | 6.80 (4.57) | 5.70 (2.82) |
| Diazinon | 0.84 (0.61) | 2.32 (1.37) | 3.61 (0.93) | 2.49 (1.07) | 7.36 (4.95) | 5.56 (2.63) | 9.26 (3.56) | 4.04 (3.77) | 4.31 (3.08) | 3.59 (2.49) |
| Dieldrin | 0.15 (0.11) | 0.18 (0.11) | 0.13 (0.05) | 0.10 (+) | 1.00 (0.82) | 0.25 (0.13) | 0.32 (0.31) | 0.32 (0.18) | 1.25 (1.65) | 0.37 (0.24) |
| Endosulfan I | 0.12 (0.04) | 0.31 (0.23) | 0.43 (0.51) | 0.63 (0.51) | 10.65 (7.06) | 3.25 (2.09) | 0.18 (0.11) | 0.35 (0.27) | 0.40 (0.44) | 0.57 (0.52) |
| Endosulfan II | 0.12 (0.07) | 0.64 (0.38) | 1.05 (0.63) | 2.08 (1.62) | 3.71 (2.95) | 3.73 (2.06) | 0.96 (0.82) | 1.37 (0.61) | 1.71 (1.95) | 2.24 (2.54) |
| Endrin | 0.11 (0.13) | 0.15 (0.04) | 0.21 (0.09) | 0.17 (0.06) | 0.57 (1.13) | 0.18 (0.11) | 0.10 (0.07) | 0.44 (1.04) | 0.65 (1.05) | 0.19 (0.34) |
| Glyphosate | 1.67 (0.62) | 37.02 (4.16) | 60.10 (6.22) | nd | nd | nd | nd | nd | nd | nd |
| Heptachlor | 0.073 | 1.52 (0.36) | 1.45 (1.04) | 1.80 (1.35) | 0.65 (0.41) | 2.98 (2.45) | 0.63 (0.82) | 0.51 (0.72) | 0.89 (0.41) | 0.73 (0.96) |

**Table 2.** Continued.

| | Sources of organic matter | | | | Invertebrates | | Fish | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | SOM | Algal turf | Macro-algae | Seagrass | Bivalves | Gastropods | Herbivores | Omnivores | Micro-carnivores | Macro-carnivores |
| | (0.038) | | | | | | | | | |
| Heptachlor-epoxide A | 0.078 (0.091) | 0.41 (0.11) | 0.34 (0.33) | 0.35 (0.23) | 2.67 (1.79) | 2.26 (2.01) | 1.12 (0.41) | 0.84 (0.38) | 1.87 (2.71) | 1.38 (1.41) |
| Heptachlor-epoxide B | 0.09 (0.13) | 0.13 (0.09) | 0.29 (0.20) | 0.17 (0.10) | 0.51 (0.32) | 0.40 (0.49) | 0.18 (0.01) | 0.49 (0.70) | 0.37 (0.51) | 0.29 (0.30) |
| Lindane | 0.17 (0.20) | 0.07 (0.04) | 0.10 (0.07) | 0.14 (0.08) | 4.67 (2.48) | 3.07 (0.60) | 2.99 (0.88) | 3.51 (2.01) | 4.58 (5.03) | 3.69 (1.48) |
| Linuron | 0.18 (0.28) | 0.71 (0.38) | 1.18 (0.80) | 1.61 (0.46) | 14.09 (11.08) | 19.65 (12.35) | 2.22 (0.78) | 3.38 (2.48) | 13.92 (4.89) | 4.47 (3.89) |
| Malathion | 0.26 (0.08) | 3.00 (0.69) | 2.91 (1.07) | 2.77 (1.51) | 7.32 (4.84) | 4.18 (1.49) | 4.44 (1.58) | 3.55 (3.09) | 3.77 (0.79) | 3.02 (1.16) |
| *pp'*-DDE | 0.37 (0.31) | 0.33 (0.20) | 0.72 (0.09) | 0.54 (0.31) | 0.87 (0.45) | 0.84 (0.34) | 0.63 (0.32) | 0.51 (0.27) | 0.72 (1.02) | 1.03 (2.10) |
| *pp'*-DDD | 0.06 (0.02) | 0.09 (0.07) | 0.13 (0.16) | 0.17 (0.10) | 0.63 (0.60) | 0.34 (0.56) | 0.05 (+) | 0.15 (0.18) | 0.54 (0.74) | 0.44 (0.59) |
| *pp'*-DDT | 0.06 (0.02) | 0.25 (0.37) | 0.31 (0.39) | 0.14 (0.21) | 0.05 (+) | 0.05 (+) | 0.05 (+) | 0.05 (+) | 0.05 (+) | 0.14 (0.46) |
| ΣPest[b] | 3.34 (1.78) | 26.48 (8.55) | 21.08 (3.83) | 22.04 (3.94) | 103.62 (33.66) | 100.42 (32.49) | 62.75 (7.84) | 57.91 (34.90) | 85.94 (66.86) | 59.87 (24.63) |
| (C) | | | | | | | | | | |
| PCB (ng·g$^{-1}$ dry weight) | | | | | | | | | | |
| $PCB_{tot1}$ | 17.74 (5.19) | 268.66 (108.61) | 166.40 (58.16) | 156.55 (84.85) | nd | nd | nd | 102.69 (98.10) | nd | 111.05 (84.78) |
| $PCB_{tot2}$ | 14.78 (4.39) | 118.69 (42.45) | 71.91 (31.27) | 81.12 (38.92) | nd | nd | nd | 38.85 (37.77) | nd | 45.06 (36.36) |
| ΣPCB | 12.03 (3.38) | 112.60 (36.34) | 78.52 (26.89) | 88.30 (36.98) | nd | nd | nd | 33.97 (31.28) | nd | 35.97 (25.37) |
| ΣICES | 8.43 (2.04) | 66.39 (23.32) | 41.29 (14.02) | 42.02 (20.35) | nd | nd | nd | 25.61 (23.43) | nd | 27.77 (19.28) |
| Pesticides (ng·g$^{-1}$ dry weight) | | | | | | | | | | |
| Aldrin | 0.05 (+) | 0.32 (0.40) | 0.07 (0.03) | 0.83 (0.22) | nd | nd | nd | 0.05 (+) | nd | 0.07 (0.07) |
| Atrazine | 2.21 (2.21) | 9.66 (7.70) | 3.16 (3.45) | 1.35 (1.64) | nd | nd | nd | 1.85 (2.25) | nd | 1.28 (2.25) |
| Chlordecone | 0.98 (0.58) | 16.74 (7.05) | 13.58 (10.09) | 12.43 (2.93) | nd | nd | nd | 1.84 (2.19) | nd | 4.96 (3.23) |
| Diazinon | 2.56 (3.93) | 3.74 (4.37) | 3.96 (2.57) | 4.26 (4.16) | nd | nd | nd | 0.97 (0.48) | nd | 0.73 (0.75) |
| Dieldrin | 0.14 (0.05) | 0.87 (0.46) | 0.86 (1.08) | 1.70 (0.85) | nd | nd | nd | 0.24 (0.12) | nd | 0.19 (0.15) |
| Endosulfan I | 0.14 (0.09) | 0.49 (0.39) | 0.51 (0.54) | 0.26 (0.35) | nd | nd | nd | 0.11 (0.04) | nd | 0.15 (0.14) |
| Endosulfan II | 0.29 (0.20) | 3.28 (1.68) | 3.72 (3.22) | 3.09 (1.97) | nd | nd | nd | 3.05 (3.50) | nd | 1.13 (0.78) |
| Endrin | 0.11 (0.09) | 0.13 (0.04) | 0.19 (0.14) | 0.40 (0.33) | nd | nd | nd | 0.06 (0.02) | nd | 0.08 (0.07) |
| Heptachlor | 0.097 (0.056) | 1.17 (1.23) | 1.23 (1.07) | 1.89 (0.74) | nd | nd | nd | 0.13 (0.15) | nd | 0.26 (0.38) |
| Heptachlor-epoxide A | 0.33 (0.25) | 0.50 (0.48) | 0.78 (0.38) | 0.12 (0.09) | nd | nd | nd | 0.01 (0.005) | nd | 0.01 (0.01) |
| Heptachlor-epoxide B | 0.037 (0.052) | 1.23 (0.46) | 1.18 (0.60) | 1.46 (0.46) | nd | nd | nd | 0.43 (0.45) | nd | 0.40 (0.38) |
| Lindane | 0.08 (0.02) | 1.76 (1.91) | 1.23 (1.06) | 1.62 (0.58) | nd | nd | nd | 0.89 (0.89) | nd | 0.73 (0.55) |
| Linuron | 4.43 (4.07) | 7.43 (5.89) | 6.67 (5.39) | 7.95 (5.55) | nd | nd | nd | 0.97 (1.01) | nd | 1.00 (2.03) |
| Malathion | 5.52 (5.66) | 7.37 (4.68) | 2.83 (1.54) | 5.50 (2.55) | nd | nd | nd | 0.79 (0.96) | nd | 1.16 (1.64) |
| *pp'*-DDE | 0.69 (0.40) | 4.36 (3.44) | 4.68 (2.35) | 7.51 (1.25) | nd | nd | nd | 0.81 (0.80) | nd | 1.09 (0.80) |
| *pp'*-DDD | 0.07 (0.02) | 0.13 (0.20) | 0.36 (0.41) | 0.80 (0.83) | nd | nd | nd | 0.21 (0.27) | nd | 0.32 (0.38) |
| *pp'*-DDT | 0.05 (+) | 0.05 (+) | 0.05 (+) | 0.05 (+) | nd | nd | nd | 0.05 (+) | nd | 0.28 (0.44) |
| ΣPest | 17.77 (11.68) | 59.20 (20.17) | 45.03 (16.69) | 51.22 (12.41) | nd | nd | nd | 12.44 (5.97) | nd | 13.85 (8.90) |

a Except glyphosate (and except atrazine, linuron and malathion for groups not measured).

b Except glyphosate.

**Table 3.** Mean values of DDT/ΣDDT and ΣPest/ΣPCB ratios (±SD) for the various sources of organic matter, invertebrates and fish trophic groups in the three studied islands. nd=no data, a sign '+' indicates a standard deviation lower than 0.01.

| | | Sources of organic matter | | | | Invertebrates | | Fish | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | SOM | Algal turf | Macro-algae | Seagrass | Bivalves | Gastropods | Herbivores | Omnivores | Micro-carnivores | Macro-carnivores |
| DDT/ΣDDT | New Caledonia | 0.33 (+) | 0.33 (+) | nd | nd | 0.20 (0.14) | 0.33 (0.02) | nd | 0.38 (0.20) | nd | 0.38 (0.16) |
| | Wallis | 0.19 (0.14) | 0.27 (0.19) | 0.19 (0.21) | 0.14 (0.11) | 0.05 (0.03) | 0.05 (0.02) | 0.08 (0.03) | 0.09 (0.04) | 0.21 (0.17) | 0.14 (0.10) |
| | Moorea | 0.33 (+) | 0.02 (0.01) | 0.02 (+) | 0.01 (+) | nd | nd | nd | 0.09 (0.05) | nd | 0.18 (0.24) |
| ΣPest/ΣPCB | New Caledonia | 0.75 (0.29) | 1.21 (0.46) | nd | nd | 1.26 (1.07) | 1.32 (0.80) | nd | 1.45 (0.37) | nd | 1.14 (0.38) |
| | Wallis | 2.34 (0.81) | 2.07 (0.68) | 2.29 (0.59) | 1.80 (0.35) | 1.58 (0.89) | 1.37 (0.47) | 1.81 (0.42) | 1.69 (1.05) | 1.80 (1.07) | 1.42 (0.61) |
| | Moorea | 0.52 (0.49) | 0.61 (0.37) | 0.58 (0.12) | 0.70 (0.38) | nd | nd | nd | 0.38 (0.20) | nd | 0.34 (0.25) |

**Table 4.** Linear regressions between contaminant concentrations (in $Log_{10}$) and $\delta^{15}N$ ratios of reef organisms. Regression parameters (p and r), the trophic magnification factor (TMF) and number of samples (N) are indicated. Significance: * = p < 0.05, ** = p < 0.01 and *** = p < 0.001. ns= non-significant (p > 0.05).

| Contaminant | New Caledonia | | Wallis | | Moorea | |
|---|---|---|---|---|---|---|
| | r | TMF | r | TMF | r | TMF |
| Ag | 0.12 ns | 1.10 | 0.52*** | 0.72 | 0.54*** | 0.93 |
| As | 0.39*** | 0.80 | 0.27* | 1.12 | 0.03 ns | 0.99 |
| Cd | 0.71*** | 0.48 | 0.55*** | 0.66 | 0.55*** | 0.86 |
| Co | 0.90*** | 0.54 | 0.77*** | 0.50 | 0.87*** | 0.70 |
| Cr | 0.74*** | 0.57 | 0.55*** | 0.76 | 0.77*** | 0.71 |
| Cu | 0.59*** | 0.64 | 0.49*** | 0.77 | 0.51*** | 0.91 |
| Fe | 0.83*** | 0.47 | 0.73*** | 0.53 | 0.86*** | 0.61 |
| Hg | 0.42*** | 1.26 | 0.68*** | 6.95 | 0.92*** | 1.52 |
| Mn | 0.85*** | 0.47 | 0.72*** | 0.48 | 0.87*** | 0.62 |
| Ni | 0.85*** | 0.46 | 0.68*** | 0.58 | 0.79*** | 0.66 |
| Pb | 0.55*** | 0.67 | 0.72*** | 0.66 | 0.83*** | 0.72 |
| Se | 0.30** | 1.10 | 0.44*** | 1.22 | 0`.81*** | 1.21 |
| V | 0.80*** | 0.63 | 0.52*** | 0.77 | 0.75*** | 0.74 |
| Zn | 0.28** | 0.87 | 0.13 ns | 0.94 | 0.10 ns | 1.01 |
| *N* | *104* | | *150* | | *57* | |
| $PCB_{tot1}$ | 0.36** | 0.40 | 0.31** | 1.11 | 0.31 ns | 0.96 |
| $PCB_{tot2}$ | 0.39** | 0.90 | 0.30** | 1.12 | 0.36* | 0.95 |
| ΣPCB | 0.54*** | 0.86 | 0.36*** | 1.12 | 0.35* | 0.96 |
| ΣICES | 0.38** | 0.90 | 0.31** | 1.10 | 0.33* | 0.96 |
| Aldrin | 0.29* | 0.85 | 0.47*** | 0.87 | 0.36* | 0.92 |
| Atrazin | 0.07 ns | 1.10 | 0.53*** | 1.32 | 0.20 ns | 0.92 |
| Chlordecone | 0.83*** | 1.57 | 0.29** | 0.95 | 0.05 ns | 0.98 |
| Diazinon | 0.06 ns | 1.04 | 0.32** | 1.01 | 0.45** | 0.89 |
| Dieldrin | 0.09 ns | 1.02 | 0.19 ns | 1.07 | 0.59*** | 0.88 |
| Endosulfan I | 0.43*** | 1.26 | 0.25* | 0.86 | 0.27 ns | 0.96 |
| Endosulfan II | 0.29* | 1.22 | 0.02 ns | 1.01 | 0.54*** | 0.89 |
| Endrin | 0.23 ns | 0.86 | 0.20 ns | 0.92 | 0.40** | 0.94 |
| Heptachlor | 0.44*** | 0.90 | 0.34** | 0.85 | 0.45** | 0.88 |
| Heptachlor-epoxide A | 0.73*** | 2.11 | 0.12 ns | 1.06 | 0.60*** | 0.80 |
| Heptachlor-epoxide B | 0.78*** | 1.98 | 0.07 ns | 1.03 | 0.42** | 0.89 |
| Lindane | 0.21 ns | 0.93 | 0.67*** | 1.50 | 0.34* | 0.93 |
| Linuron | 0.16 ns | 1.18 | 0.20 ns | 1.09 | 0.57*** | 0.74 |
| Malathion | 0.23 ns | 1.25 | 0.05 ns | 1.01 | 0.52*** | 0.81 |
| *pp′*-DDD | 0.84*** | 1.52 | 0.04 ns | 1.02 | 0.06 ns | 1.02 |
| *pp′*-DDE | 0.83*** | 1.54 | 0.04 ns | 1.01 | 0.43** | 0.90 |
| *pp′*-DDT | 0.62*** | 0.95 | 0.27* | 0.93 | 0.39* | 1.01 |
| ΣPesticides | 0.36** | 1.10 | 0.31** | 1.08 | 0.62*** | 0.90 |
| *N* | *68* | | *88* | | *41* | |

Supplementary Table 1. Numbers of samples analyzed for metallic (N1) and organic (N2) contaminants analyses.

| Type of sample | Category or family | Group or species | Size (min-max), TL in cm | N1 | N2 |
|---|---|---|---|---|---|
| *Sources of organic matter* | | | | | |
| | Sedimentary organic matter | | | 18 | 18 |
| | Algal turf | | | 18 | 18 |
| | Macro-algae | *Padina australis* | | 15 | 12 |
| | Seagrass | *Cymodocea serrulata* | | 3 | |
| | | *Halophila decipiens* | | 6 | 6 |
| | | *Syringodium isoetifolium* | | 6 | 6 |
| *Invertebrates* | | | | | |
| | Bivalves | *Pinna rugosa* | | 11 | 11 |
| | | *Isognomon isognomon* | | 6 | 6 |
| | | Unidentified oyster | | 4 | 4 |
| | Gastropods | *Conomurex luhuanus* | | 8 | 6 |
| | | *Conus marchionatus* | | 4 | 3 |
| | | *Conus marmoreus* | | 3 | |
| | | *Cyprea tigris* | | 3 | 3 |
| | | *Tectus niloticus* | | 5 | |
| | | *Turbo* sp. | | | 6 |
| | | Unidentified | | 2 | |
| | Hermit crabs | *Dardanus* sp. | | 6 | |
| | Others crustaceans | *Palinurus ornatus* | | 3 | |
| | | *Palinurus versicolor* | | 3 | |
| | | Unidentified shrimps | | 2 | |
| *Fish* | | | | | |
| | Acanthuridae | *Acanthurus nigrofuscus* | 10.5 - 12.6 | 4 | |
| | | *Ctenochaetus striatus* | 6.5 - 19.3 | 11 | |
| | Apogonidae | *Apogon* sp. | nm * | 2 | |
| | | *Cheilodipterus quinquelineatus* | 7.0 - 7.4 | 3 | |
| | | *Fowleria variegata* | 4.8 - 8.7 | 5 | |
| | Caesionidae | *Caesio caerulaurea* | 10.1 - 14.4 | 6 | |

| | | | | |
|---|---|---|---|---|
| Chaetodontidae | *Chaetodon vagabundus* | 4.5 - 5.0 | 2 | |
| Gobiidae | *Amblygobius phalaena* | 7.0 - 8.1 | 5 | |
| Holocentridae | *Myripristis kuntee* | 4.0 - 8.1 | 8 | 5 |
| | *Neoniphon sammara* | 6.5 - 15.4 | 4 | |
| | *Sargocentron diadema* | 6.4 - 11.3 | 4 | |
| | *Sargocentron tiere* | 19.2 - 21.7 | 3 | |
| | *Sargocentron violaceum* | 6.4 - 14.4 | 2 | |
| Labridae | *Cheilinus chlorurus* | 4.9 - 8.7 | 7 | |
| | *Halichoeres trimaculatus* | 6.5 - 10.2 | 5 | |
| | *Thalassoma hardwicke* | 5.6 - 7.5 | 3 | |
| | *Thalassoma lunare* | 7.0 - 13.3 | 3 | |
| Muraenidae | *Gymonthorax* sp. | 24.5 - 58.0 | 4 | |
| Pomacentridae | *Amblyglyphidodon curacao* | 4.8 - 6.5 | 4 | |
| | *Chromis iomelas* | 2.5 - 6.8 | 4 | |
| | *Chrysiptera biocellata* | 5.5 - 8.6 | 3 | |
| | *Chrysiptera taupou* | 4.5 - 6.0 | 10 | |
| | *Pomacentrus pavo* | 6.0 - 6.5 | 2 | |
| | *Stegastes lividus* | 12.4 - 16.2 | 10 | 10 |
| | *Stegastes nigricans* | 4.6 - 14.7 | 43 | 43 |
| | *Chlorurus sordidus* | 5.3 - 8.7 | 7 | |
| Scorpaenidae | *Scorpaenodes guamensis* | 4.6 - 8.6 | 6 | |
| Serranidae | *Cephalopholis argus* | 16.2 - 37.5 | 9 | 9 |
| | *Cephalopholis sexmaculata* | 18.8 - 41.0 | 16 | 16 |
| | *Epinephelus coeruleopunctatus* | 32.1 - 47.0 | | 2 |
| | *Epinephelus howlandi* | 24.5 - 32.0 | 2 | 2 |
| | *Epinephelus macrospilos* | 25.5 - 36.0 | 2 | 2 |
| | *Epinephelus merra* | 14.0 - 20.5 | 11 | 12 |
| | *Epinephelus polyphekadion* | 24.3 - 52.0 | 2 | 3 |
| | *Plectropomus laevis* | 44.0 - 95.0 ** | 7 | 7 |
| | *Plectropomus leopardus* | 37.9 - 48.8 ** | 8 | 8 |
| Tetraodontidae | *Canthigaster solandri* | nm * | 1 | |

** nm= individuals not measured ; ** Fork length*

Supplementary Table 2. Quality of analytical results: recoveries were obtained by spiking a fish devoid of PCB and pesticides, mean ± standard deviation (SD) from certified material reference (IAEA 406 fish homogenate), CIM (95% confidence interval of the median; IAEA 406 fish homogenate) and detection limits (DL, in µg $kg^{-1}$). See also text below.

| | Recovery (%) | Mean ± SD | CIM | DL |
|---|---|---|---|---|
| | N= 13 | N= 6 | | |
| *pp'*-DDT | | 3.58 ± 0.49 | 1.8-5.6 | 0.1 |
| *pp'*-DDD | | 2.67 ± 0.74 | 2.0-3.7 | 0.1 |
| *pp'*-DDE | | 6.15 ± 1.78 | 6.2-11.0 | 0.1 |
| Diazinon | 84.28 ± 9.84 | | | 0.1 |
| γ-lindan | 97.85 ± 9.28 | 0.15 ± 0.27 | 0.11-0.80 | 0.1 |
| Heptachlor | 115.36 ± 11.34 | 0.56 ± 0.26 | 0.23-0.46 | 0.1 |
| Heptachlor epoxide A | 94.78 ± 9.07 | 0.46 ± 0.14 | 0.37-1.60 | 0.01 |
| Heptachlor epoxide B | 85.34 ± 7.10 | 0.65 ± 0.17 | | 0.01 |
| Aldrin | 90.87 ± 9.95 | 0.67 ± 0.21 | 0.61-1.20 | 0.1 |
| Dieldrin | 101.22 ± 10.18 | 4.92 ± 1.30 | 1.4-7.0 | 0.2 |
| Endrin | | 5.30 ± 0.67 | 0.86-5.1 | 0.1 |
| Chlodecon | 87.56 ± 8.45 | | | 0.01 |
| Glyphosate | 70.87 ± 13.76 | | | 1 |
| Endosulfan 1 | | 3.04 ± 0.86 | 0.94-4.7 | 0.2 |
| Endosulfan 2 | | 0.94 ± 0.47 | 11.6 | 0.2 |
| C28 | 87.53 ± 0.97 | 0.61 ± 0.11 | 0.43-1.3 | 0.01 |
| C52 | 114.44 ± 0.51 | 1.15 ± 0.34 | 1.0-2.2 | 0.01 |
| C101 | 90.06 ± 0.52 | 3.71 ± 0.78 | 2.2-3.4 | 0.01 |
| C118 | 90.06 ± 0.52 | 3.19 ± 0.76 | 1.9-3.7 | 0.01 |
| C138 | 94.20 ± 0.35 | 4.28 ± 0.77 | 2.5-6.3 | 0.01 |
| C153 | 113.37 ± 0.49 | 4.13 ± 0.70 | 2.9-6.0 | 0.01 |
| C180 | 91.47 ± 0.62 | 1.52 ± 0.19 | 1.0-1.2 | 0.01 |

*Extraction and quantitative analysis*

For PCBs and Chlorinated pesticides, the procedure used is described by Dierking et al., 2009. Briefly, approximately 1 g of freeze-dried sample (tissue) was analysed, except sedimentary samples for which between 3 to 6 g of material were used. Freeze-dried samples were extracted with hexane in the thimbles in the Soxhlet apparatus for 16 hours minimum. Each extract was then concentrated to 2 mL using Rotavapor. About 100 µL of the extract was used to gravimetrically determine the lipid content of each sample. The remaining fraction was purified with concentrated sulfuric acid, followed by additional purification by liquid chromatography on a silica–alumina column, and a final purification (only fraction I) on a silica column. Four fractions were eluted, with fraction Ia containing the PCBs and the pesticides Aldrin, pp'-DDE, chlordecone and Heptachlor (50%); fraction Ib, Heptachlor (50%) and pp'-DDT; fraction II, lindane (γ-HCH), and pp'-DDD; and fraction III, Dieldrin and Heptachlor epoxide.

For PCB compounds, analyses were performed with a HP 6890 series gas chromatograph equipped with a 63Ni electron capture detector (ECD) at 300°C and an automatic injector on-column, HP 6890 series. The column used was a DB5 J&W (60 m x 0.32 i.d. x 0.25 µm). The carrier gas was helium. The temperature of injection was 60°C and was programmed to increase

up to 250°C at 100°C/min. The column temperature was 60°C and programmed to increase first up to 160°C (10°C/min) and then up to 280°C at 2°C/min.

The chlorinated pesticides, (fractions Ib, II, and III) were analyzed by gas chromatography (Agilent Technology 6890 N, equipped with a splitless injector) coupled to mass spectrometry (MSD) with SIM mode. The same column as for PCBs was used (program: from 50 to 100°C at 25°C/min and from 100 to 280°C at 5°C/min, keeping the first temperature for 1 min and the final temperature for 10 min). Injector and detector temperature were 250 and 280°C. The carrier gas was helium. A standard mixture containing all pesticides in this study was used for calibration. In all case, the variation in the response of the detector was corrected by a daily calibration with a standard solution of PCBs congeners, and for pesticides. The sample response was matched to that of the standard solution by dilution or concentration of the sample.

*Quality control*

The analytical precision was checked by analyzing the organochlorinated compounds in fish homogenate sample six times (IAEA 406), which was distributed to worldwide laboratories in January 2000 (Villeneuve et al., 2000). The means results with standard deviation were reported in Table S2. Furthermore, recoveries were obtained by spiking with 1 mL of a standard solution at 0.05 mg $L^{-1}$ for each standard of PCBs and pesticides, a homogenized powder of a sample of fish devoid of these compounds. Thirty replicates were conducted on the same day. Table S2 shows that the percentage recoveries and their standard deviations range from 87.53±0.97 to 114.44±0.51 % for PCBs and from 84.28±9.84 to 115.36±11.34 % for pesticides, depending on the compounds. Subsequently, in each batch, a blank and this Certified Reference Material (IAEA 406) were systematically introduced in order to check glassware and lab contamination. The results obtained for the CRM were used to plot control charts and to decide upon acceptance or rejection of the data produced for each sample batch. Rejected batches were reanalyzed. Calibration curves were constructed for each compound and checked by inspection of at least two standards at the beginning of the analyses and one every ten samples.

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Supplementary Table 3. Summary of statistical significances (Kruskal-Wallis test or ANOVA) of comparisons in mean metallic concentrations of organic matter sources, invertebrates (no data from Moorea) and fish trophic groups from Wallis (W), Moorea (M) and New Caledonia (NC). ns : not significant, p < 0.05 : normal characters, p < 0.01 : italic characters, p < 0.001 : bold characters.

| | **Sources of organic matter** | | | | **Invertebrates** | | **Fish** | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | SOM | Algal turf | Macroalgae | Seagrass | Bivalves | Gastropods | Herbivores | Omnivores | Micro-carnivores | Macro-carnivores |
| Ag | ns | ns | ns | ns | *W>NC* | ns | ns | **NC>W=M** | ns | *NC>W=M* |
| As | M>W=NC | **W>M>NC** | ns | NC=W>M | ns | ns | ns | ns | ns | ns |
| Cd | ns | ns | NC>W=M | **NC=M>W** | NC>W | NC>W | ns | ns | *M>W=NC* | ns |
| Co | NC=M>W | *M>W=NC* | NC>W>M | *NC=W>M* | ns | NC>W | **NC>W=M** | *NC>W=M* | ns | ns |
| Cr | ns | *M=NC>W* | *NC=W>M* | *M>NC>W* | NC>W | NC>W | ns | *M=W>NC* | **M=W>NC** | ns |
| Cu | ns | *M>W=NC* | NC=W>M | NC=W>M | ns | ns | ns | ns | **M>W=NC** | ns |
| Fe | M>W>NC | *M>W=NC* | NC>W=M | **M>NC>W** | ns | NC>W | ns | ns | M>W=NC | ns |
| Hg | ns | W>NC=M | *NC>W=M* | NC>W=M | ns | ns | ns | *NC>W=M* | NC>W=NC | M=NC>W |
| Mn | **M>W=NC** | *M>W=NC* | ns | ns | ns | NC>W | ns | *M=W>NC* | **M>W=NC** | *W=NC>M* |
| Ni | ns | ns | **NC>W=M** | **NC>M>W** | ns | ns | ns | ns | *M=W>NC* | ns |
| Pb | W=M>NC | M>W=NC | NC>W=M | ns | ns | NC>W | ns | ns | ns | ns |
| Se | ns | *M>W>NC* | ns | **NC=M>W** | ns | ns | ns | NC>W=M | ns | M=W>NC |
| V | ns | **M>W=NC** | ns | **M>NC>W** | ns | **NC>W** | ns | ns | M>W=NC | **M=W>NC** |
| Zn | ns | M=W>NC | ns | ns | *NC>W* | ns | ns | ns | M=NC>W | W>NC=M |

Supplementary Table 4. Summary of statistical significances (Kruskal-Wallis test or ANOVA by permutation) of comparisons in mean PCBs and pesticides concentrations of organic matter sources, invertebrates (no data from Moorea) and fish trophic groups from Wallis (W), Moorea (M) and New Caledonia (NC). '-': not tested, ns : not significant, $p < 0.05$ : normal characters, $p < 0.01$ : italic characters, $p < 0.001$ : bold characters (ΣPest *: sum of pesticides, except atrazine, linuron and malathion for groups not measured in New Caledonia).

| | **Sources of organic matter** | | | | **Invertebrates** | | **Fish** | |
|---|---|---|---|---|---|---|---|---|
| | SOM | Algal turf | Macroalgae | Seagrass | Bivalves | Gastropods | Omnivores | Macro-carnivores |
| $PCB_{tot1}$ | **M>NC=W** | *NC=M>W* | **M>W** | *M>W* | W>NC | *W>NC* | W=M>NC | ns |
| $PCB_{tot2}$ | **M>NC=W** | *NC=M>W* | **M>W** | *M>W* | W>NC | W>NC | W=M>NC | ns |
| ΣPCB | **M>NC=W** | **NC=M>W** | **M>W** | M>W | Ns | W>NC | *W=M>NC* | W>M=NC |
| ΣICES | **M>NC=W** | *NC=M>W* | **M>W** | *M>W* | W>NC | *W>NC* | W=M>NC | ns |
| Aldrin | ns | ns | ns | ns | **NC>W** | NC>W | *NC>W=M* | *NC>W=M* |
| Atrazine | **M>W** | **M>W** | ns | ns | - | - | **W>NC=M** | **W>NC=M** |
| Chlordecone | *M=W>NC* | *M=W>NC* | ns | **M>W** | **W>NC** | **W>NC** | **NC>W>M** | **NC>W=M** |
| Diazinon | ns | ns | ns | ns | Ns | ns | W>NC>M | **W=NC>M** |
| Dieldrin | ns | ns | ns | **M>W** | **NC>W** | **NC>W** | NC>W=M | **NC>W=M** |
| Endosulfan I | ns | ns | ns | ns | Ns | ns | **NC>W=M** | **NC>W=M** |
| Endosulfan II | W=M>NC | M=NC>W | ns | ns | Ns | ns | ns | ns |
| Endrin | NC>W=M | NC>W=M | ns | ns | *NC>W* | NC>W | | *NC>W=M* |
| Heptachlor | **NC>M=W** | ns | ns | ns | **NC>W** | ns | NC=W>M | ns |
| Heptachlor-epoxide A | *M>W>NC* | ns | ns | W>M | **W>NC** | W>NC | **NC=W>M** | **W=NC>M** |
| Heptachlor-epoxide B | ns | **M>W>NC** | *M>W* | **M>W** | Ns | ns | ns | ns |
| Lindane | ns | M>NC>W | M>W | **M>W** | **W>NC** | **W>NC** | **W>M=NC** | **W>M=NC** |
| Linuron | **M>W** | **M>W** | M>W | ns | - | - | **W>NC=M** | **W>NC=M** |
| Malathion | **M>W** | **M>W** | ns | M>W | - | - | *W>NC>M* | **W=NC>M** |
| *pp'*-DDE | **M>W** | ns | ns | ns | Ns | W>NC | M=NC>W | |
| *pp'*-DDD | M>W | ns | M>W | *M>W* | Ns | **W>NC** | NC>W=M | *NC>W=M* |
| *pp'*-DDT | ns | *W=M>NC* | ns | ns | Ns | ns | ns | ns |
| ΣPest * | **M>W=NC** | ns | ns | **M>W** | **W>NC** | ns | *W>NC>M* | *W>NC>M* |