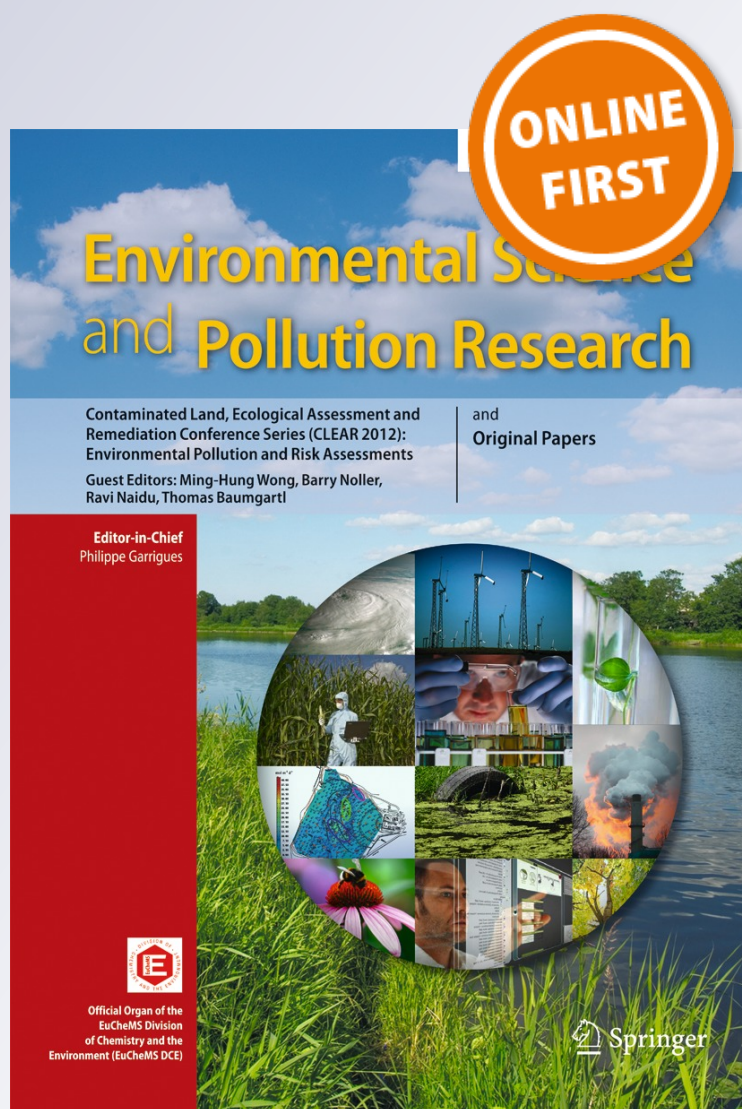# Spatial variability of metallic and organic contamination of anguilliform fish in New Caledonia

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Spatial variability of metallic and organic contamination of anguilliform fish in New Caledonia

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Abstract New Caledonia is one of the main hot spots of biodiversity on the planet. Large amounts of contaminants are discharged into the lagoon as a result of increasing anthropogenic activities such as intense mining, urbanization, and industrialization. Concentrations of 14 trace elements and 26 persistent organic pollutants (POPs: PCBs and pesticides) were measured in the muscles of two anguilliform fish species, over a coast to barrier reef gradient in two lagoon areas differently exposed to anthropic disturbances. This study emphasizes the high trace element contamination status of anguilliform fish and also highlights slight but perceptible organic pollution. The contamination extends throughout the lagoon, from coast to barrier reef, even in areas remote from emission points. High levels of trace elements, especially those linked to mining activities (i.e., Co, Cr, Fe, Mn, and

Ni), were detected in coastal sites. Furthermore, the large dispersion of most POPs throughout the entire lagoon poses the question of their potential toxicity on marine organisms from numerous habitats. Our results underline the need for long-term monitoring of various contaminants over large spatial and time scales.

Introduction

Coral reefs are threatened by a wide range of natural and anthropic disturbances, including chemical pollution (Richmond 1993). Three main contaminant classes are particularly involved: polychlorinated biphenyls (PCBs), pesticides (both classes are part of persistent organic pollutants or POPs), and metallic trace elements. These ubiquitous contaminants can be toxic, even at low concentrations; they are resistant to degradation, transported over long distances, and can bioaccumulate in marine organisms (e.g., Phillips 1995; Robertson and Hansen 2001). The half-life of contaminants in the marine environment is estimated to be at least a decade for the most persistent PCBs (Sinkkonen and Paasivirta 2000; Robertson and Hansen 2001), several years to a decade for trace elements such as Hg (Lodenius 1991) and several months to years for pesticides (Hellawell 1988). After their release into the marine environment, some contaminants can be strongly accumulated by organisms of different trophic levels (Van Ael et al. 2012; Dummee et al. 2012).

Southwest Pacific coral reefs are generally in good health. However, for a few decades, toxic wastes released by human activities threaten some of them (Richmond 1993). New Caledonia's lagoon, the largest in the world, is no exception. This hot spot of biodiversity is subjected to increasing

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contamination pressure resulting from industrialization and urbanization. New Caledonia is the third producer of nickel ore in the world (Dalvi et al. 2004). Since the end of the nineteenth century, many open-cast mines and three metallurgical factories have been opened. Mining activities and natural soil erosion due to rainfall (Ambastian et al. 1997) generate massive sediment deposits and discharge of associated trace elements (Co, Cr, Fe, Mn, and Ni,) into the lagoon. Furthermore, with the expansion of industrial factories and urban development, the lack of efficient treatment of wastewater, and the use of pesticides for agriculture, the contamination by POPs is also an important issue. Nearly 250 pesticides are officially authorized in New Caledonia (http://davar. gouv.nc/). Twenty-six of these are banned but in Europe are still extensively used in New Caledonia.

Several studies focused on trace metal contaminants in coastal waters, in particular around Nouméa (the main urban region) (Fernandez et al. 2006; Metian et al. 2008a, b; Hédouin et al. 2008, 2009, 2011). Bioaccumulation of mining (Co, Cr, Fe, Mn, and Ni) and urban trace elements (Ag, As, Cd, Cu, Hg, Pb, Se, V, and Zn) have been investigated in various taxa (crustaceans, molluscs, ascidians) (Bustamante et al. 2000; Hédouin et al. 2006, 2007, 2009, 2010, 2011; Pernice et al. 2009; Metian et al. 2008a, 2010). One study recently revealed high contamination levels by trace elements in fish from the south-western lagoon, extending from the coast to the barrier reef (Bonnet et al. 2014). However, there is still little information available about trace element contamination of fish across the whole lagoon in New Caledonia (Chouvelon et al. 2009; Fernandez and Breau 2011; Metian et al. 2013). Furthermore, to our knowledge, there is no information available to date on PCB and pesticide concentrations in marine organisms in New Caledonia.

Analyses of spatial patterns of contamination allow the identification of their environmental sources. They also lead to a better understanding of the physical, chemical, and biological processes involved in contaminant accumulation (Robertson and Hansen 2001; Johnson et al. 2005). However, with the exception of the coastal region (especially in Nouméa), contamination by trace elements is insufficiently documented and its impact on marine ecosystems remains unclear. Considering the immense surface of the south-western lagoon of New Caledonia, it is necessary to extend investigations to a large spatial scale and to other contaminants such as POPs.

Marine top predators, mainly long-living species, bioaccumulate high levels of metallic elements (e.g., Cd and Hg) or organochlorine pollutants in their tissues (e.g., Adams and McMichael 1999; Wafo et al. 2012). Fish are particularly useful to assess contamination status and distribution of pollutants, especially if they are site-attached or even territorial, living in benthic habitats and have a high trophic level (carnivorous) (e.g., Kojadinovic et al. 2007; Dierking et al. 2009). The sedentary nature of some fish is a crucial aspect in

spatial variability studies of contamination. Anguilliform fish (morays, congers, and snake eels), which are ubiquitous organisms highly represented in tropical marine waters, fulfill the conditions of good bioindicators in contamination monitoring (Bonnet et al. 2014). These sedentary benthic predators, which mainly live in coral reef matrices of shallow waters, are good candidates for studying the impact and spatial variability of contamination in marine environments. Surprisingly, to date, they have received little interest, which could be due to their cryptic lifestyle and the difficulty to collect them (Ineich et al. 2007).

In this general context, the main purposes of this study were (1) to determine the level of contamination of two anguilliform fish species by trace elements and POPs, (2) to study the spatial variability of these contaminants along two coast-barrier reef gradients which are differently exposed to human activities. As the influence of ecological and biological factors on the concentration of contaminants is important, three biological parameters (body size, trophic position, and lipid content in tissue) and their link to the bioaccumulation processes were also investigated.

Materials and methods

Fish sampling and study areas

Two anguilliform species were studied, *Gymnothorax chilospilus* (Muraenidae) and *Conger* sp. (Congridae). Capture of specimens was carried out using sea kraits (*Laticauda* spp.) because the prey of these predators is almost exclusively anguilliform fish. This method has been successfully used in previous studies (e.g., Reed et al. 2002; Ineich et al. 2007; Brischoux et al. 2011). The prey items were obtained by a gentle massage of the sea krait abdomen. Because the eels consumed are non-spiny, the snake readily regurgitates their prey (Brischoux and Bonnet 2009) without any effect on their survival (Fauvel et al. 2012). The spatial accuracy of sea snake sampling and their use as sentinels was described in previous studies (Brischoux et al. 2007a, 2009; Brischoux and Bonnet 2008; Bonnet 2012). Sea kraits probe the surrounding seafloors within a radius of 10–15 km, and anguilliform fish are extremely sedentary. Considering the large spatial scale of the current study, this accuracy was sufficient to examine possible local influences along a coast-barrier reef gradient.

The study was realized in the South Western lagoon of New Caledonia. Both fish species were caught in two main areas: Grand Nouméa (GN) and Grand Lagon Sud (GS) (Fig. 1). Grand Nouméa, which is close to the main city of Nouméa, is subject to a variety of anthropic pollution such as industrial activities, farming industries, and wastewater. In particular, a nickel factory has functioned since 1880 (SLN,
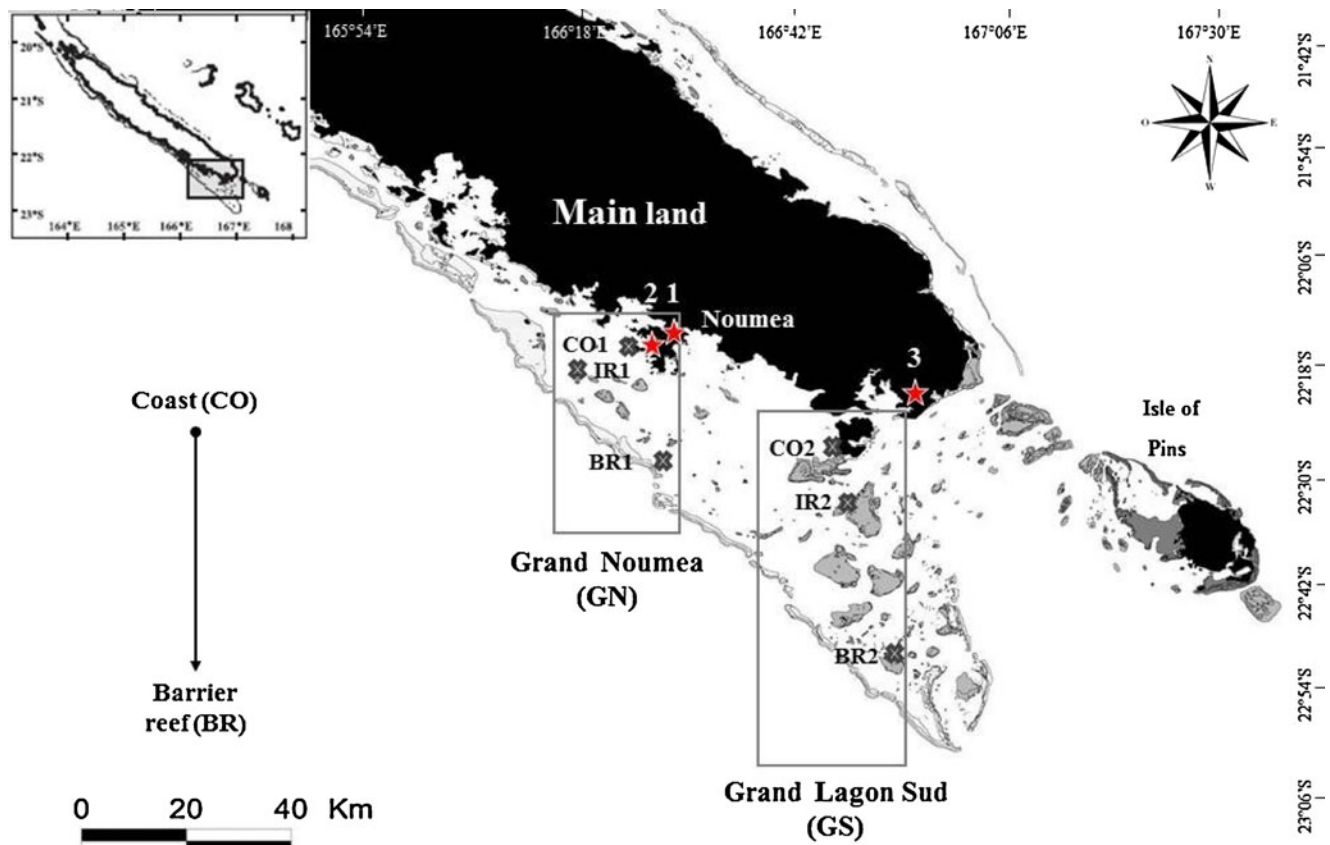
Fig. 1 Location of the sampled sites distributed on two coast-barrier reef gradients in New Caledonia: Kuendu (*CO1*), Signal (*IR1*), and Amédée (*BR1*) in Grand Nouméa (*GN*) and Ouen (*CO2*), Grand Mato (*IR2*) and N'Da (*BR2*) in Grand Lagon Sud. *CO*=coastal sites; *IR*=intermediate reef sites; *BR*=barrier reef sites. *Stars* indicate areas of human activities, which are the industrial area of Ducos (*1*), the Nickel factory « SLN » (*2*), and the Nickel factory « Goro-Nickel » (*3*). The emerged land is indicated in *black*; *grey* areas represent coral reefs; the *dark grey line* represents the slope of the barrier reef

Société Le Nickel, Fig. 1). Grand Lagon Sud is less influenced by industrial and urban pollution. Nevertheless, since 1950, mining activity has developed near the Bay of Prony (Goro-Nickel, Vale Inco, Fig. 1), increasing the erosion of lateritic soil in this region. For each area, three stations located on a coast-barrier reef gradient inside the lagoon were sampled: Kuendu beach (coast; CO1), Signal islet (intermediate-reef; IR1), and Amédée islet (barrier-reef; BR1) inside GN and Ouen island (coast; CO2), Mato islet (intermediate-reef; IR2), and N'da islet (barrier-reef; BR2) inside GS (Fig. 1). Sampling was carried out from January to April 2011 and from August to September 2011, corresponding respectively to the hot and wet versus cool and dry seasons in New Caledonia.

Each fish was identified, measured, and weighed prior to dissection (Table 1). As some fish collected were partially digested, their total length (TL) was estimated applying allometric equations using either the snout vent length or the tail length (Brischoux et al. 2007b). For each individual of both species, a piece of white muscle was sampled on the non-digested part and immediately frozen at −30 °C for subsequent analyses. Muscle tissues were freeze-dried and ground to powder with a porcelain mortar and pestle.

Trace element analyses

The total Hg concentration in the powder obtained from the tissues was determined by analyzing the Hg, with an advanced mercury analyzer (ALTEC AMA 254), directly on aliquots ranging from 5 to 50 mg of dry sample weighed to the nearest 0.01 mg (Bustamante et al. 2006). From 150 to 300 mg of each sample were digested using a 3:1 *v/v* nitric–hydrochloric acid mixture with 65 % ultrapure HNO_3 and ultrapure 37 % HCl. The acidic digestion was performed overnight under ambient temperature and then heated in a microwave for 30 min, increasing the temperature up to 105 °C, and 15 min at 105 °C (1200 W). After the mineralization process, each sample was diluted to 30 or 50 ml with milli-Q quality water, according to the volume of acid added to the mineralization (3 and 4.5 ml, respectively).

The analysis of Ag, As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Se, V, and Zn required an extra step in the preparation protocol. These elements were analyzed by inductively coupled plasma

Table 1 Number (N) of fish analyzed from the coastal (CO), intermediate (IR), and barrier reef (BR) sites in Grand Nouméa (GN) and Grand Lagon Sud (GS)

Species	Site	N	N (GN)	N (GS)	TL (cm) (min–max)	δ^{15}N (‰) (min–max)	Lp (%) (min–max)
G. chilospilus	CO	8	3	5	21.2±4.3 (14.7–28.4)	8.0±0.8 (7.1–9.3)	1.2±1.3 (0.2–3.6)
	IR	20	10	10	25.3±4.8 (14.0–37.2)	7.9±0.7 (6.7–9.5)	0.8±0.8 (0.2–3.3)
	BR	20	10	10	25.1±2.3 (21.4–30.8)	7.8±0.5 (7.1–8.8)	0.7±0.6 (0.2–1.8)
	Total	48	23	25	24.5±4.1 (21.7–31.2)	7.9±0.6 (6.7–9.5)	0.8±0.8 (0.2–3.6)
Conger sp.	CO	14	9	5	29.0±4.6 (23.2–38.3)	8.6±1.4 (6.6–10.5)	1.0±0.7 (0.2–1.9)
	IR	10	0	10	34.4±8.9 (20.3–48.7)	8.5±0.8 (7.7–9.9)	1.5±1.6 (0.3–5.5)
	BR	13	8	5	35.7±6.3 (27.7–51.7)	8.5±0.9 (7.1–10.3)	1.0±1.2 (0.3–3.6)
	Total	37	17	20	32.7±7.1 (20.3–51.7)	8.6±1.0 (6.6–10.5)	1.2±1.2 (0.2–5.5)

Mean (±SD) fish total length (TL, centimeters), trophic position (express with δ^{15}N, ‰), and lipid content (Lp, %) are given, with minimal and maximal values (min–max) indicated in brackets. No variation of TL, δ^{15}N, and Lp between sites was observed ($p_{ANOVA} > 0.05$)

atomic emission spectrometry (Varian Vista-Pro ICP-OES) and mass spectrometry (ICP-MS II Series Thermo Fisher Scientific). Reference tissues—dogfish liver DOLT-4 (NRCC), lobster hepatopancreas TORT-2 (NRCC)—were treated and analyzed in the same way as the samples. Results were in line with the certified values, and the standard deviations were low, proving good repeatability of the method. The results for standard reference materials displayed recovery of the elements ranging from 73 % to 116 %. For each set of analyses, blanks were included in each analytical batch. The detection limits (micrograms per gram dry weight) were 0.005 (Hg), 0.015 (Cd), 0.017 (Ag), 0.02 (Cr, Co, Pb), 0.03 (Ni), 0.08 (Mn), 0.1 (Cu, Se), 0.2 (As), 0.3 (V), and 3.3 (Fe and Zn). All trace element concentrations are given on a dry weight basis (micrograms per gram dry weight).

POP analyses

Choice of PCB congeners and pesticides

The concentrations of 15 individual congeners (IUPAC Nos. 18, 20, 28, 31, 44, 52, 101, 105, 118, 138, 149, 153, 170, 180, 194; Ballschmiter and Zell 1980) were determined in each sample. This list contains the seven target congeners (PCBs 28, 52, 101, 118, 138, 153, 180) proposed by the International Council for the Exploration of the Sea (ICES) as indicators of PCB contamination (Valoppi et al. 2000).

The samples and blank were analyzed for the following pesticides: aldrin, diazinon, dieldrin, endosulfan I, endosulfan II, endrin, heptachlor, heptachlor-epoxide A, heptachlor-epoxide B, lindane, and, pp'-DDD, pp'-DDE, pp'-DDT.

Sample extraction and quantification

Compounds were extracted and quantified following the procedures described by Dierking et al. (2009). About 1 g of lyophilized sample was extracted and concentrated to 2 ml.

About 100 μL of this extract was reserved to determine the content of lipid (Lp, %) of each sample (see next paragraph). The remaining fraction was purified with concentrated sulfuric acid, followed by additional purification by liquid chromatography on a silica-alumina column (Murphy 1972; Wells et al. 1985). Four fractions were eluted: fraction Ia (aldrin, pp'-DDT, heptachlor (50 %)), fraction Ib (heptachlor (50 %), pp'-DDT), fraction II (endosulfan I, endosulfan II, lindane, pp'-DDD) and fraction III (dieldrin, endrin, heptachlor-epox A and B).

PCBs and pesticides were analyzed by gas chromatography coupled to mass spectrometry. Calibration was done using fifteen individual standard solutions for PCBs and a standard mixture containing all pesticides was used for pesticides. All organic pollutant concentrations are given on a dry weight basis (nanograms per gram dry weight). Detection limits were 0.01 ng g^{-1} for PBC congeners, 0.1 ng g^{-1} for DDT, and its metabolites, 0.01 ng.g^{-1} for heptachlor-epoxide A and B, 0.1 ng.g^{-1} for lindane, diazinon, heptachlor, aldrin, endosulfan II, endrin, and 0.2 ng g^{-1} for endosulfan I and dieldrin.

δ^{15}N and lipid quantification

The nitrogen stable-isotope ratio (δ^{15}N) was used to give an approximate trophic position. Analyses were carried out on dorsal white muscle, as this tissue gives the most reliable values (Pinnegar and Polunin 1999). Then 1±0.1 mg of powdered freeze-dried samples was weighed out and sealed in tin capsules for analyses. ^{15}N/^{14}N ratios were determined by continuous-flow isotope-ratio mass spectrometry with a Thermo Scientific Delta V Advantage mass spectrometer coupled to a Thermo Scientific Flash EA1112 elemental analyzer (Chouvelon et al. 2011). Results are expressed as isotope ratios δ^{15}N (‰) relative to international standard (atmospheric N$_2$ for nitrogen), according to the formula:

$$\delta^{15}N = \left[(R_{sample}/R_{standard}) - 1\right]^* 10^3$$

where $R = {}^{15}N/{}^{14}N$ (Peterson and Fry 1987). Replicate measurements of internal laboratory standards (acetanilide) indicated a precision of approximately 0.2‰ for $\delta^{15}N$ values.

To evaluate the lipid content (Lp, %) in each fish, 100 μL of the evaporated extraction residue was introduced in an initially tarred (tablet) container; then the whole was deposited into a desiccator. After drying to constant mass, the lipid content is determined by gravimetry.

Statistical analysis

Several indices were used to characterize the POP contamination. Our PCB profiles revealed that PCBs detected in the environment mainly came from capacitors (pyralene: DP3, DP4 or Arochlor1230, 1242). Consequently, it was decided to express the global footprint of PCB contamination ($PCBTot$) using the estimation proposed by Annema et al. (1995): $PCBTot= \Sigma \ (28+52+101+138+153+180)*5$. $\Sigma ICES$ (sum of concentrations of the seven ICES congeners), and the proportional contribution of each PCB class (tri- to octa-chlorinated) was also calculated using all of the PCBs analyzed. The total DDT concentration, ΣDDT, was calculated as the sum of pp'-DDD, pp'-DDE, and pp'-DDT. An indication of the timing of PCB inputs was estimated through the ratio of the two congeners CB153/CB138 (Monod et al. 1995; Wafo et al. 2005: A high ratio (>1) indicates continuous inputs or persistence of PCBs in the environment, whereas a low ratio (<1) reflects a decrease or sporadicity of PCB inputs or non-persistence. Finally, the ratio of the total concentration of pesticides to total concentration of PCBs analyzed ($\Sigma Pest/\Sigma PCB$) was used as an approximate indication of the relative importance of agricultural *versus* industrial sources of organic pollutants in the environment (de Mora et al. 2004); a ratio higher than 1 indicates prevalence of agricultural sources, whereas a ratio less than 1 indicates prevalence of industrial sources.

Data were log-transformed to satisfy conditions of normality (Shapiro-Wilk test) and homogeneity of variance (Bartlett test). However, for some compounds (aldrin, diazinon, dieldrin, endrin, pp'-DDD, and pp'-DDT), transformation was not sufficient. When conditions were satisfied, parametric tests were used. In other cases, non-parametric analogues were applied.

Principal component analysis (PCA) was used to obtain an overview of the contribution of each contaminant according to sites. Trace elements and POP concentrations were analyzed in separate PCAs, and as no clear patterns were revealed from POP analysis, only trace element results were presented. Spatial variability of contaminant concentrations in each species was also tested using two-way ANOVA and Tukey's post hoc multiple comparisons. For all contaminants, Pearson or

Spearman correlations were used to explore the relationship between both contaminant concentration and fish size (TL) and contaminant concentration and trophic level ($\delta^{15}N$). For organic pollutants, the relationship between contaminant concentration and the percentage of lipids in muscle (Lp) was also examined. When a correlation existed, ANCOVA, instead of ANOVA was applied, using TL, $\delta^{15}N$ or Lp as a covariate. R project (R version 2.13.2) was the statistical program used for all analyses. The levels of significance for statistical analyses was always set at $\alpha =0.05$.

Results

Accumulation levels in anguilliform fish

All trace elements measured were detected in anguilliform fish, except Ag and V, which were below the detection limit. Mean concentrations and ranges of detected values of contaminants (trace elements and POPs) in anguilliform fish muscle are presented in Table 2. Several elements showed a high inter-individual variability (i.e., Cd, Cr, Ni, Pb; coefficient of variation CV>100 %). However, it was most often the same individuals that accumulated high concentrations of several trace elements linked to their origin (e.g., a *G. chilospilus* specimen from the coastal site of Grand Nouméa; 35.7 μg g^{-1} dw of Cr and 17.4 μg g^{-1} dw of Ni).

Overall, few differences in mean contaminant concentrations were found between species. However, significantly higher concentrations of As, Hg, and Se were measured in *Conger* sp., whereas *G. chilospilus* exhibited higher concentrations of Cd and Zn ($p_{ANOVA}<0.001$). PCBs had been similarly accumulated by both species, albeit weak interspecific differences were detected for pesticides: *Conger* sp. presented significantly higher concentrations of heptachlor, heptachlor-epoxy B ($p_{ANOVA}<0.01$), while slightly higher concentrations of endosulfan I and aldrin were measured in *G. chilospilus* ($p_{ANOVA}<0.05$).

Spatial patterns in contaminants

Trace elements

A significant spatial variability of several trace elements, in particular, those linked with mining activity (Co, Cr, Fe, Mn, and Ni), was detected along the coast-barrier reef gradient (Fig. 2 and Table 3). The PCAs in *G. chilospilus* indicated that the coastal sites were characterized by Co, Mn, and Ni elements, in contrast to the barrier reef sites which were characterized by As and Cd concentrations (the plot of the first two PCA dimensions explained respectively 51.1 % and 59.0 % of data variability in *G. chilospilus* and *Conger* sp.,

Table 2 Trace element and organic pollutant concentrations (mean±SD and min-max values; micrograms per gram dry weight) measured in muscle of *G. chilospilus* and *Conger* sp., all sites confounded

		G. chilospilus	*Conger* sp.
Trace elements	As	11±7.4 (0.9–31.9)	25.7±25.6 (3.0–117.9)
	Cd	0.41±0.47 (0.01–2.91)	0.04±0.04 (0.01–0.16)
	Co	0.11±0.08 (0.02–0.45)	0.13±0.15 (0.02–0.68)
	Cr	4.53±7.15 (0.86–35.71)	4.03±5.36 (0.45–23.60)
	Cu	1.6±0.7 (0.7–3.4)	1.9±0.9 (0.7–4.5)
	Fe	40.2±36.9 (12.2–196.8)	34.3±31.1 (7.9–134.4)
	Hg	0.049±0.028 (0.02–0.13)	0.117±0.112 (0.03–0.6)
	Mn	2.3±1.4 (0.6–7.2)	2.7±2.3 (0.4–10.1)
	Ni	1.95±3.19 (0.30–17.38)	1.44±1.77 (0.10–7.74)
	Pb	0.05±0.04 (0.01–0.2)	0.04±0.09 (12.2–196.8)
	Se	1.2±0.4 (0.7–2.6)	1.8±0.8 (0.9–4.6)
	Zn	56.5±16.4 (26.5–112.3)	45.4±23.8 (21.6–112.1)
Organic pollutants	*PCBTot*	59.1±52.7 (9.3–271.5)	63.2±54.8 (9.1–211.4)
	ΣICES	14.0±12.2 (2.0–64.3)	16.5±14.3 (2.1–60.9)
	Aldrin	0.1±0.06 (0.08–0.3)	0.1±0.05 (0.04–0.2)
	Diazinon	1.8±1.9 (<0.1–10.2)	2.4±3.3 (<0.1–15.3)
	Dieldrin	0.4±0.3 (<0.2–1.8)	0.5±0.4 (<0.2–2.2)
	Endosulfan I	1.3±1.3 (<0.2–6.1)	1.0±1.7 (0.09–9.7)
	Endosulfan II	2.1±1.7 (<0.1–7.3)	2.9±2.3 (<0.1–8.9)
	Endrin	0.3±0.8 (<0.1–4.5)	0.8±2.0 (0.07–9.9)
	Heptachlor	0.4±0.4 (<0.1–1.9)	0.5±0.4 (<0.1–1.9)
	Heptachlor-epox. A	1.5±1.2 (0.03–5.6)	1.8±1.9 (0.1–9.1)
	Heptachlor-epox. B	0.3±0.5 (<0.01–2.6)	0.5±0.6 (<0.01–2.7)
	Lindane	1.3±0.6 (0.3–3.1)	1.7±1.2 (0.2–5.1)
	ΣDDT	1.6±1.5 (0.3–6.9)	1.6±1.5 (0.3–7.2)

Fig. 2). In *Conger* sp., the difference between sites was even stronger, with most trace elements (As, Co, Cr, Fe, Ni, Se, and Zn) characterizing coastal sites. These general trends were confirmed by the significantly higher concentrations of mine elements at coastal sites for both species (p_{ANOVA} or p_{ANCOVA} <0.05, Table 3).

The same spatial pattern (coast vs. barrier reef) was revealed in the two studied areas (GN and GS, Table 3 and S1). The coastal site of Grand Nouméa showed significantly higher concentrations of Ni in *G. chilospilus* (p_{ANOVA}=0.02) and Cr and Fe in *Conger* sp. (p_{ANOVA}<0.05, Table 3 and S1 of the Electronic supplementary material), with the same trends followed by several other trace elements (Cr, Fe in *G. chilospilus*, As, Co, Cu, Ni, Se, and Zn in *Conger* sp., results of PCA not shown). However, in Grand Lagon Sud, the spatial gradient was less pronounced. The coastal site was characterized by higher Co and Mn concentrations in both species (results of PCA not shown), but spatial analyses only suggested significantly lower concentrations of As and Cd in *G. chilospilus* (p_{ANCOVA}=0.001, Tables 3 and S1 of the Electronic supplementary material).

The coastal site of Grand Nouméa was characterized by significantly higher levels of several trace elements compared with Grand Lagon Sud (e.g., As, Cd, and Ni in *G. chilospilus*, and Cr, Fe in *Conger* sp.; p_{ANCOVA}<0.05, Tables 3 and S1 of the Electronic supplementary material).

Organic pollutants

The PCAs did not reveal any clear spatial pattern for POPs in either species (results not shown). Analyses suggested a low spatial variability along the coast-barrier reef gradient (Table 4).

The PCB contamination in *G. chilospilus* did not vary significantly between sites (see *PCBTot* index; p_{ANOVA}> 0.05), in spite of a slight increase from the coast to the barrier reef. However, in *Conger* sp., the intermediate sites were characterized by high concentrations (p_{ANCOVA}<0.05, Table 4). The *CB*153/*CB*138 ratio, which was generally below 1, showed few spatial variations and only *G. chilospilus* from coastal sites showed a significantly higher ratio (p_{ANCOVA}<0.05, Tables 4 and S2 of the Electronic supplementary material). The PCB chlorination classes differed slightly along the coast-barrier reef gradient and between studied areas, with a low variability detected in each case (<10 %, p_{ANCOVA} or p_{ANOVA}<0.05, Table 4). However, the
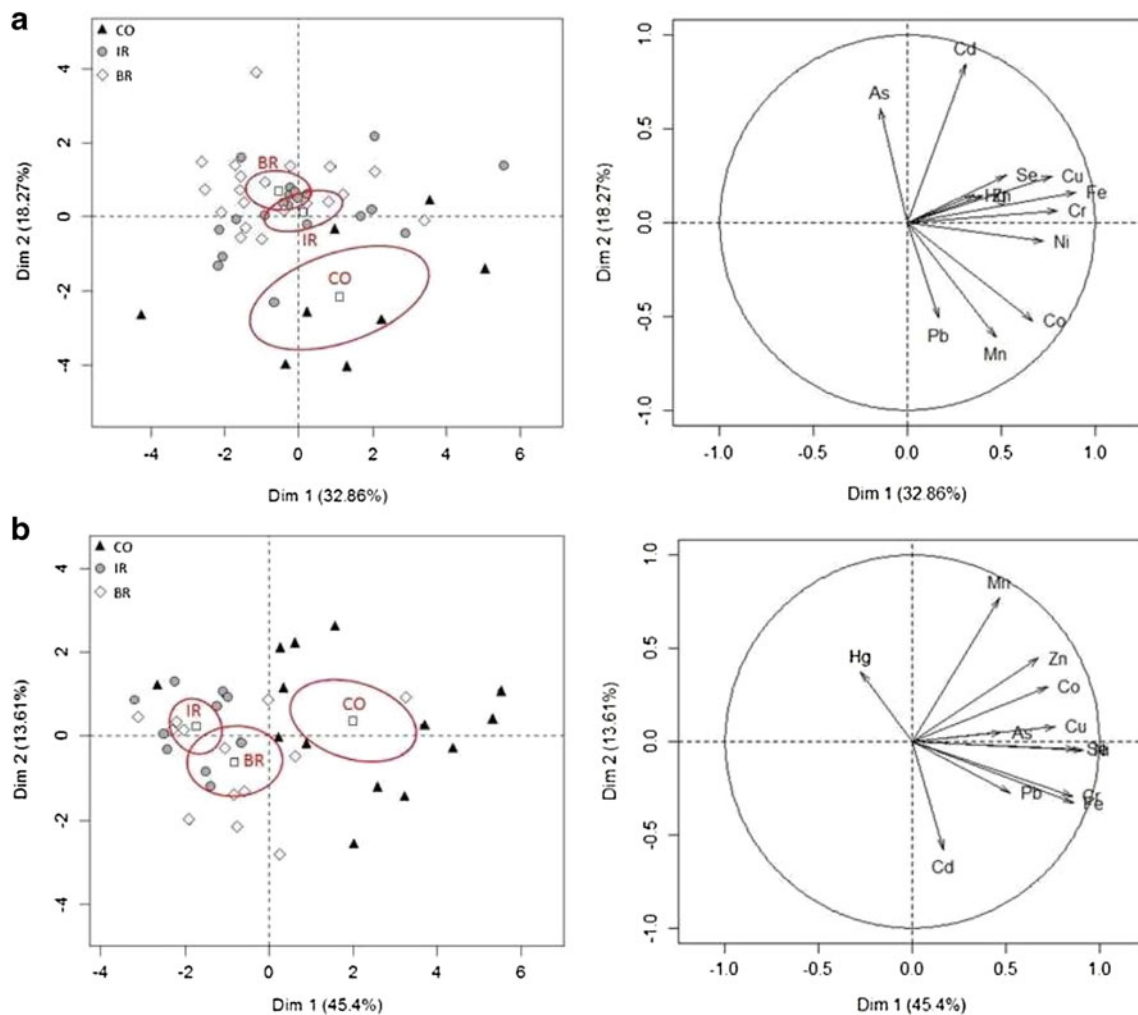
Fig. 2 Plot of the principal component analysis (PCA) assessing spatial patterns of anguilliform fish contamination by trace elements on the coast-barrier reef gradient (*left panel*) and the contribution of the contaminants analyzed to site differentiation (*right panel*) in *G. chilospilus* (**a**) and *Conger* sp. (**b**). *CO*=coastal sites; *IR*=intermediate reef sites;

BR=barrier reef sites. The most contributing variables in both species were Cr, Cu, Fe, and Ni concentrations for the first axis and Cd and Mn concentrations for the second axis. The significant differences between sites are illustrated by confidence ellipses of centroids at 95 %

results highlighted a remarkable increase in the proportion of the most toxic chlorinated classes in coastal sites (i.e., 8Cl in GN for *G. chilospilus* and 7Cl in GS for *Conger* sp., $p_{ANOVA}<0.05$).

The results for pesticides indicated few spatial variations (Tables 4 and S2 of the Electronic supplementary material). The coastal sites highlighted significantly higher concentrations of some pesticides in *G. chilospilus* (aldrin and heptachlor, p_{ANCOVA} or $p_{KW}<0.05$), whereas the intermediate reef sites revealed significantly higher rates of several other pesticides in *Conger* sp. (e.g., diazinon, heptachlor epoxide A and pp'-DDT; p_{ANCOVA} or $p_{KW}<0.05$).

Finally, spatial analyses of POP contamination according to sources of pollution (agriculture vs. industrial) reinforce previous results (see *ΣPest/ΣPCBs;* Table 4). Along the gradient, *G. chilospilus* from the coastal sites were significantly different from other sites, with quite homogeneous sources of contamination (ratio≥1). In comparison industrial

sources dominated in other sites (ratio≅0.50, $p_{ANOVA}<0.05$, Table 4). A strong influence of industrial pollution in intermediate sites (ratio≅0.50, $p_{ANCOVA}<0.05$) was also highlighted in *Conger* sp.

Influence of size, trophic position, and lipid content on contamination level

Several trace elements (Co, Cr, Ni, Pb, Se, and Zn in *G. chilospilus*, Cd and Mn in *Conger* sp.) were not correlated with either the size or the trophic position of fish ($p_{Pearson}>0.05$), but some trace elements were significantly correlated with fish size (Table 5). Only As and Hg highlighted a positive relationship with the trophic position of fish (δ^{15}N) in *Conger* sp., whereas no correlation was found for *G. chilospilus*.

Organic pollutant concentrations were linked to size, trophic position, or lipid content, depending on the contaminant.

Table 3 Trace element concentrations (mean (SD); micrograms per gram dry weight) in *G. chilospilus* and *Conger* sp. along the coastal-barrier reef gradient

		Gradient					Spatial patterns			
		CO	IR	BR	Stat test	p	Gradient	GN	GS	GN vs. GS
G. chilospilus	As	6.77 (10.27)	7.84 (5.75)	14.33 (5.85)	ACV (δ^{15} N)	***	BR>CO/IR	BR1>IR1	IR2/BR2>CO2	CO1>CO2
	Cd	0.16 (0.23)	0.44 (0.63)	0.43 (0.31)	ACV (TL)	***	IR/BR>CO		IR2/BR2>CO2	CO1>CO2
	Co	0.19 (0.12)	0.10 (0.05)	0.07 (0.05)	ANV	***	CO>IR>BR			
	Cr	6.59 (11.93)	3.57 (5.98)	3.97 (5.81)	ANV	NS				
	Cu	1.62 (0.76)	1.66 (0.78)	1.37 (0.40)	ACV (TL)	NS				
	Fe	46.5 (50.5)	38.2 (39.9)	33.8 (27.4)	ACV (TL)	NS				
	Hg	0.05 (0.02)	0.05 (0.03)	0.04 (0.03)	ACV (TL)	NS				
	Mn	3.77 (2.18)	1.84 (0.91)	1.76 (0.76)	ACV (TL)	***	CO>IR/BR			
	Ni	3.72 (5.65)	1.51 (2.33)	1.34 (2.32)	ANV	*	CO>BR	CO1>IR1/BR1		CO1>CO2
	Pb	0.06 (0.05)	0.04 (0.03)	0.04 (0.03)	ANV	NS				
	Se	0.98 (0.27)	1.20 (0.38)	1.07 (0.42)	ANV	NS				
	Zn	42.9 (9.9)	55.8 (17.3)	55.1 (17.1)	ANV	NS				
Conger sp.	As	37.96 (31.44)	20.57 (21.08)	16.45 (16.60)	ACV (δ^{15} N)	*	CO>BR			
	Cd	0.04 (0.04)	0.03 (0.04)	0.06 (0.05)	ANV	ns				
	Co	0.23 (0.19)	0.10 (0.06)	0.05 (0.03)	ACV (TL)	***	CO/IR>BR			
	Cr	7.88 (7.09)	0.95 (0.31)	2.26 (1.68)	ACV (TL)	**	CO/IR>BR	CO1>BR1		CO1>CO2
	Cu	2.37 (1.05)	1.32 (0.43)	1.88 (0.79)	ACV (TL)	ns				
	Fe	57.4 (40.2)	15.3 (4.0)	24.2 (9.7)	ACV (TL)	**	CO>IR/BR	CO1>BR1		CO1>CO2
	Hg	0.11 (0.07)	0.07 (0.03)	0.15 (0.17)	ACV (δ^{15} N)	**	CO/BR>IR			
	Mn	3.7 (2.30)	1.76 (0.95)	2.28 (2.66)	ANV	ns				
	Ni	2.78 (2.25)	0.35 (0.14)	0.83 (0.67)	ACV (TL)	***	CO>IR /BR			
	Pb	0.07 (0.05)	0.03 (0.01)	0.09 (0.14)	ACV (TL)	ns				
	Se	2.31 (1.06)	1.37 (0.21)	1.51 (0.43)	ACV (TL)	**	CO>BR			
	Zn	59.5 (26.2)	33.8 (6.3)	39.0 (23.10)	ACV (TL)	*	CO>IR/BR			

ANOVA (ANV) or ANCOVA (ACV) with Tukey's pairwise comparisons were used to analyze spatial patterns

GN Grand Nouméa, *GS* Grand Lagon Sud, *CO* coastal sites, *IR* intermediate reef sites, *BR* barrier reef sites, *ns* not significant ($p > 0.05$)

*$p < 0.05$, **$p < 0.01$, ***$p < 0.001$

Most of them were mainly significantly correlated with the lipid contents in fish (Table 5). For example, PCBs were only correlated with lipids in both species. Pesticides were also correlated with Lp, and to a lesser extent with δ^{15} N and TL (Table 5).

Discussion

This study confirms, reinforces, and extends the high trace element contamination status of anguilliform fish in New Caledonia (Bonnet et al. 2014). It also constitutes the first substantial baseline on organic pollutant contamination of fish in New Caledonia. Importantly, our results show that large scale contamination reaches remote parts of the lagoon, more than 30 km offshore (SW lagoon), and thus well beyond the urbanized Nouméa region.

Contaminant levels and spatial variability

Trace element patterns

Anguilliform fish concentrate both significant levels of trace elements associated with mining exploitation (Cr, Fe, Mn, and Ni for both species), and also some elements linked to urban activities (e.g., Cd, Cu, and Zn, especially in *G. chilospilus*) (Table 2). This elevated typical mining signature detected in sedentary benthic species contrasts with other studies which highlight low contaminant levels in some more mobile fish in both the lagoons of New Caledonia and other tropical regions (Eilser 2010; Bonnet et al. 2014; Table 6).

Although coastal sites in New Caledonia are highly contaminated by trace elements, directly, or indirectly linked to mining activity (Co, Cr, Fe, Mn, and Ni), the contamination

Table 4 PCBs and pesticide concentrations (mean (SD); nanograms per gram dry weight, except percent of classes) in *G. chilospilus* (A) and *Conger* sp. (B) along the coast-barrier reef gradient

		Gradient			Stat test	p	Spatial patterns			
		CO	IR	BR			Gradient	GN	GS	GN *vs.* GS
A)	*G. chilospilus*									
	PCB index (ng g⁻¹ dw)									
	PCBTot	26.96 (9.21)	68.64 (67.30)	60.62 (43.55)	ANV	ns				
	$\Sigma ICES$	6.02 (2.16)	17.00 (16.02)	13.74 (9.04)	ANV	ns				
	153/138	1.17 (0.52)	0.76 (0.13)	0.80 (0.10)	ACV (Lp)	***	CO>IR/BR		CO2>IR2/BR2	CO2>CO1
	PCB chlorination classes (% of CTotal PCBs)									
	3 Cl	20.39 (7.67)	16.64 (5.30)	14.12 (3.22)	ANV	*	CO>BR		CO2/IR2>BR2	CO2>CO1
	4 Cl	20.09 (9.93)	14.84 (7.94)	18.12 (5.31)	ACV (Lp)	ns				
	5 Cl	25.65 (4.87)	34.32 (6.53)	30.48 (5.85)	ANV	*	IR>CO			
	6 Cl	28.07 (6.89)	29.56 (8.73)	32.46 (7.44)	ANV	ns				
	7 Cl	3.55 (1.15)	3.82 (1.99)	4.33 (2.72)	ACV (Lp)	ns				
	8 Cl	2.25 (3.12)	0.82 (1.17)	0.50 (0.75)	ANV	**		CO1>BR1		GN>GS
	Pesticides (ng g⁻¹ dw)									
	Ald.	0.16 (0.11)	0.11 (0.03)	0.06 (0.04)	KW	**	CO>BR			
	Dia.	3.91 (3.41)	1.47 (1.68)	1.35 (0.92)	KW	ns				
	Diel.	0.41 (0.26)	0.36 (0.24)	0.37 (0.39)	KW	ns				
	Endo.I	1.09 (1.06)	1.27 (1.04)	1.36 (1.66)	ACV (δ^{15} N)	ns				
	Endo. II	1.53 (0.86)	2.38 (2.16)	2.00 (1.54)	ANV	ns				
	Endr.	0.16 (0.09)	0.28 (0.62)	0.34 (0.97)	KW	ns				
	Hept.	0.83 (0.51)	0.43 (0.48)	0.20 (0.17)	ACV (Lp)	***	CO>IR/BR			
	Hept. A	0.66 (0.22)	1.69 (1.49)	1.58 (1.09)	ANV	ns				
	Hept. B	0.30 (0.23)	0.23 (0.33)	0.27 (0.60)	ANV	*			CO2>IR2/BR2	IR1>IR2
	Lind.	1.32 (0.47)	1.50 (0.85)	1.22 (0.46)	ANV	ns				
	DDE	0.23 (0.23)	0.34 (0.36)	0.50 (0.77)	ANV	ns				
	DDD	0.29 (0.17)	0.24 (0.21)	0.41 (0.52)	KW	ns				
	DDT	1.88 (1.58)	0.86 (1.19)	0.47 (0.63)	KW	ns				
	ΣDDT	2.40 (1.70)	1.44 (1.58)	1.38 (1.36)	ANV	ns				
	$\Sigma Pest/ \Sigma PCB$	1.13 (0.30)	0.5 (0.12)	0.51 (0.12)	ANV	***	CO>IR/BR			

Table 4 (continued)

B)

Conger sp.

		Gradient					Spatial patterns			
		CO	IR	BR	Stat test	p	Gradient	GN	GS	GN vs. GS
PCBs (ng g^{-1} dw)										
	PCBTot	67.66 (54.60)	99.47 (67.68)	31.91 (16.39)	ACV (Lp), **	IR>BR				
	ΣICES	17.17 (12.78)	26.32 (18.94)	8.58 (4.55)	ACV (Lp), **	IR>BR				
	153/138	0.82 (0.19)	0.88 (0.36)	1.0 (0.47)	ACV (Lp), ns					
PCB chlorination classes (% of ΣTotal PCBs)										
	3 Cl	15.75 (7.20)	19.94 (6.08)	16.09 (7.62)	ACV (Lp), ns					
	4 Cl	17.71 (6.99)	11.07 (4.59)	21.74 (8.23)	ACV (Lp), *	IR<BR				
	5 Cl	32.02 (8.23)	32.74 (7.86)	34.84 (5.25)	ACV (Lp), ns					
	6 Cl	26.5 (5.15)	32.37 (7.73)	22.28 (4.51)	ACV (Lp), ***	IR>CO>BR		CO2>BR2		
	7 Cl	6.50 (5.97)	3.62 (1.38)	3.70 (2.12)	ANV, *					
	8 Cl	1.53 (1.38)	0.26 (0.24)	1.36 (1.03)	ACV (Lp), ns					
Pesticides (ng g^{-1} dw)										
	Ald.	0.13 (0.06)	0.12 (0.04)	0.10 (0.03)	KW, ns					
	Dia.	2.02 (4.74)	3.99 (2.31)	1.54 (1.74)	KW, ***	IR>CO				
	Diel.	0.77 (0.64)	0.25 (0.09)	0.36 (0.17)	KW, *	CO>IR			CO2>CO1	
	Endo. I	1.06 (0.89)	0.61 (0.73)	1.30 (2.69)	ACV (Lp), ns					
	Endo. II	3.78 (2.46)	2.61 (2.60)	2.26 (1.83)	ACV (Lp), ns					
	Endr.	0.24 (0.31)	1.69 (3.19)	0.56 (1.55)	KW, ns					
	Hept.	0.59 (0.40)	0.51 (0.55)	0.52 (0.31)	ACV (TL), ns					
	Hept. A	2.37 (2.58)	2.47 (1.77)	0.88 (0.56)	ACV (Lp), *	IR>BR				
	Hept. B	0.79 (0.80)	0.30 (0.48)	0.33 (0.19)	ACV (Lp), ns					
	Lind.	2.56 (1.51)	1.25 (0.93)	1.23 (0.62)	ACV (Lp), ns					
	DDE	0.44 (0.61)	0.37 (0.21)	0.36 (0.29)	ANV, ns					
	DDD	0.29 (0.30)	0.65 (0.22)	0.15 (0.09)	KW, ***	IR>CO/BR		IR2>BR2		
	DDT	0.82 (1.07)	0.40 (0.61)	1.19 (1.90)	KW, ns					
	ΣDDT	1.56 (1.19)	1.42 (0.71)	1.70 (2.12)	ANV, ns					
	ΣPest/ΣPCB	0.92 (1.29)	0.51 (0.21)	0.94 (1.02)	ACV (Lp), **	CO/BR>IR				

ANOVA or ANCOVA with Tukey's pairwise comparisons were used to analyze spatial patterns

GN Grand Nouméa; *GS* Grand Lagon Sud, *CO* coastal sites, *IR* intermediate reef sites, *BR* barrier reef sites, *ns* not significant ($p>0.05$)

[a] Only one value

*$p<0.05$, **$p<0.01$, ***$p<0.001$/(+) positive and (−) negative correlation

Table 5 Summary of correlations performed on trace elements and organic pollutants (PCBs and pesticides) concentrations versus total length (TL), trophic position (δ^{15}N), and lipid content (Lp) in *G. chilospilus* and *Conger* sp

	Trace elements	PCBs	Pesticides
G. chilospilus			
TL	As*, Cd**, Cu*, Fe*, Hg*, Mn*		Ald.**, Dia.**
δ^{15}N			Endo.*
Lp		*CB153/138***, 4 Cl**, 8 Cl*	Hept.**
Conger sp.			
TL	Co**, Cr*, Cu**, Fe*, Ni*, Pb*, Se*, Zn*		Hept.**
δ^{15}N	As***, Hg***		Ald.*
Lp		CTotal PCBs**, *SICES* *, *CB153/138**, 3 Cl*, 4 Cl***, 5 Cl**, 6 Cl*, 8 Cl**	Dia.*, Diel.*, Endo.I**, Endo.II**, Endr.**, Hept.A*, Hep.B*, Lind.**, DDT**

Positive correlations are underlined

3 Cl to 8 Cl classes of chlorination, *Lind.* lindane, *Dia.* diazinon, *Hept.* heptachlor, *Ald.* Aldrin, *Hept. B* heptachlor-epox. B, *Hept. A* heptachlor-epox. A, *Endo. I* endosulfan I, *Diel* dieldrin, *Endr.* endrin, *DDT* pp'-DDT

*$p < 0.05$, **$p < 0.01$, ***$p < 0.001$, significance levels

has spread throughout the entire lagoon as far as the remote barrier reef of the extremely vast south-western lagoon. This is probably due to dispersal by local currents (Fichez et al. 2008). However, it should be noted that concentrations decrease significantly from the coast to the barrier reef.

Unsurprisingly, fish from the coastal site in Grand Nouméa present important concentrations in trace elements, especially Cr, Fe, and Ni (Table 3). This urbanized and industrialized region is subjected to clear impact from the metallurgic industry (Hédouin et al. 2009, 2011; Metian et al. 2008, 2013). In contrast, the Grand Lagon Sud is less impacted by trace element inputs. Even if coastal sites in this area seem to be less threatened, trends still reveal important concentrations of Co, Mn, and Ni (Table 3). This strong contamination level likely results from severe soil erosion, due to deforestation and mining exploitation in this region over several decades. Given the numerous mines in New Caledonia and the intensive development of mining activities, assessing to what extent other areas of the New Caledonian lagoon have been affected is timely.

We expected a strong impact of urban pollution at least in Grand Nouméa sites, given the intense activity in the coastal industrial area of Ducos. However, while spatial patterns are obvious for trace elements associated with mining activities, those associated with urban pollution (As, Cd, Cu, Pb, and Zn) are less clear. Whereas Se and Zn are mostly concentrated in coastal sites for *Conger* sp., Cu and Pb are homogeneously distributed along the coast-barrier reef gradient. Furthermore, the cases of As and Cd are more complex with sometimes higher concentrations (e.g., As in *Conger* sp.) and sometimes lower concentrations in coastal sites (e.g., As and Cd in *G. chilospilus*). Finally, elements issued from urban pollution concern the entire lagoon, maybe due to their wide dispersal by local currents (Fichez et al. 2008). Another possible explanation is that in the past some islets were used as rubbish tips for

decades (e.g., Amédée islet in Nouméa region, Bonnet et al. 2014). Further information on these issues is thus needed.

Organic pollutant patterns

Contamination by POPs detected in anguilliform fish was rather low. However, considering the high toxicity of some POPs, even at very low doses, this contamination should be considered as important. Levels of PCB concentrations recorded in New Caledonia are generally lower than in fish from other tropical or temperate regions (e.g., *ΣICES* index in New Caledonia, 14.0 ± 12.2 ng g^{-1} dw (*G. chilospilus*) and 16.5 ± 14.3 ng g^{-1} dw (*Conger* sp.); in Wallis, Central Pacific, 31.3 ± 9.6 ng g^{-1} dw (*Cephalopholis argus*), and 49.7 ± 45.3 ng g^{-1} dw (*Epinephelus merra*) (Letourneur et al., unpublished data); in the Mediterranean Sea, 100.6 ng g^{-1} dw (*Solea solea*) (Dierking et al. 2009)). Exposure profiles of PCBs showed a contribution of chlorination classes in both species, with the prevalence of moderately chlorinated classes (5Cl and 6Cl) and the scarcity of highly chlorinated classes (7Cl and 8Cl). These results emphasize a recent or even current use of PCBs in New Caledonia, otherwise only the more persistent classes (highly chlorinated) should be detected. Further studies are needed to confirm this hypothesis.

In New Caledonian fish, pesticide concentrations are equivalent or lower than in other regions (e.g., lindane in New Caledonia, 1.3 ± 0.6 ng g^{-1} dw (*G. chilospilus*) and 1.7 ± 1.6 (*Conger* sp.); in Wallis, Central Pacific, 3.5 ± 1.03 ng g^{-1} dw (*C. argus*) and 3.6 ± 2.07 ng g^{-1} dw (*E. merra*) (Letourneur et al., unpublished data); in French Polynesia, 73.3 ± 34.5 ng g^{-1} dw (*Epinephelus hexagonatus*) (Salvat et al. 2012); in the Mediterranean Sea, 35.4 ng g^{-1} dw (*S. solea*) (Dierking et al. 2009)). Despite the quantities and diversity of pesticides imported into New Caledonia, these low levels detected reflect

Table 6 Comparison of several trace element concentrations (mean±SD and range of values; micrograms per gram dry weight) measured in muscle of *G. chilospilus*, *Conger* sp., and other lagoon fish species in New Caledonia

Trace element	Concentration (μg.g^{-1}dw)	Species	Reference
As	11±7.4 (0.9–31.9)	*G. chilospilus*	This study
As	25.7±25.6 (3.0–117.9)	*Conger* sp.	This study
As	13.7±7.57 (<7.87–19.1)	*Lethrinus laticaudis*	Metian et al. (2013)
As	9.47±1.47 (<7.85–10.7)	*Priacanthus hamrur*	Metian et al. (2013)
As	<6.01–7.77	*Cymbacephalus beauforti*	Metian et al. (2013)
As	1.2–52.5	Mix	Fernandez and Breau (2011)
Cd	0.41±0.47 (0.01–2.91)	*G. chilospilus*	This study
Cd	0.04±0.04 (0.01–0.16)	*Conger* sp.	This study
Cd	<0.06	Mix	Fernandez and Breau (2011)
Co	0.11±0.08 (0.02–0.45)	*G. chilospilus*	This study
Co	0.13±0.15 (0.02–0.68)	*Conger* sp.	This study
Co	<0.03	*L. laticaudis*	Metian et al. (2013)
Co	<0.03	*P. hamrur*	Metian et al. (2013)
Co	<0.02	*C. beauforti*	Metian et al. (2013)
Co	<0.1–1.1	Mix	Fernandez and Breau (2011)
Cr	4.53±7.15 (0.86–35.71)	*G. chilospilus*	This study
Cr	4.03±5.36 (0.45–23.60)	*Conger* sp.	This study
Cr	<0.79	*L. laticaudis*	Metian et al. (2013)
Cr	1.39 (<0.70–1.39)	*P. hamrur*	Metian et al. (2013)
Cr	<0.60	*C. beauforti*	Metian et al. (2013)
Cr	<0.1–5.7	Mix	Fernandez and Breau (2011)
Cu	1.6±0.7 (0.7–3.4)	*G. chilospilus*	This study
Cu	1.9±0.9 (0.7–4.5)	*Conger* sp.	This study
Cu	<0.79	*L. laticaudis*	Metian et al. (2013)
Cu	<0.83	*P. hamrur*	Metian et al. (2013)
Cu	<0.53–0.64	*C. beauforti*	Metian et al. (2013)
Cu	0.25–3.0	Mix	Fernandez and Breau (2011)
Fe	40.2±36.9 (12.2–196.8)	*G. chilospilus*	This study
Fe	34.3±31.1 (7.9–134.4)	*Conger* sp.	This study
Fe	9.19±1.42 (7.56–10.1)	*L. laticaudis*	Metian et al. (2013)
Fe	10.4±3.56 (7.82–16.9)	*P. hamrur*	Metian et al. (2013)
Fe	7.01–7.60	*C. beauforti*	Metian et al. (2013)
Fe	4.0–211	Mix	Fernandez and Breau (2011)
Mn	2.3±1.4 (0.6–7.2)	*G. chilospilus*	This study
Mn	2.7±2.3 (0.4–10.1)	*Conger* sp.	This study
Mn	<0.79	*L. laticaudis*	Metian et al. (2013)
Mn	<0.83	*P. hamrur*	Metian et al. (2013)
Mn	<0.60	*C. beauforti*	Metian et al. (2013)
Mn	<0.03–3.3	Mix	Fernandez and Breau (2011)
Ni	1.95±3.19 (0.30–17.38)	*G. chilospilus*	This study
Ni	1.44±1.77 (0.10–7.74)	*Conger* sp.	This study
Ni	<0.06–1.35	Mix	Fernandez and Breau (2011)

Mix=144 fish from 27 species and 10 families (Acanthuridae, Carangidae, Haemulidae, Labridae, Lethrinidae, Lutjanidae, Mugilidae, Scaridae, Serranidae, and Siganidae)

their relatively moderate use, which probably comes from local and non-professional activities rather than from the agricultural industry. In addition, Nouméa is located far from the most important agricultural areas. This type of non-professional use could also explain a non-negligible detection of pesticides such as DDT and its metabolites (i.e., *ΣDDT*, 1.55±1.50 ng g^{-1} dw in *G. chilospilus* and 1.57±1.48 in *Conger* sp.), which are supposed to be prohibited by international regulations and not homologated in New Caledonia (http://davar.gouv.nc/).

Overall, POP concentrations are homogeneous throughout the lagoon, without any clear spatial pattern along the coast-barrier reef gradient, or between studied areas. Nevertheless,

some high pesticide concentrations in *G. chilospilus* were observed in several coastal sites and in intermediate reefs, with high PCB and some high pesticide levels in *Conger* sp. (Tables 4 and S2). Storage organs (i.e., liver, kidney, or digestive gland) usually accumulate higher concentrations than muscle (e.g., Bustamante et al. 2003; Chouvelon et al. 2009; Metian et al. 2013; Eilser 2010). Further research is required to test this notion in anguilliforms and thus to better assess spatial contamination gradients, especially for POPs.

The results underlined the complexity in identifying the agricultural and/or industrial origin of organic contamination for each site (see *ΣPest/ΣPCBs* ratio, Table 4). Trends seem to emphasize a slight prevalence of agricultural contamination sources at coastal sites, in comparison with other sites dominated by industrial pollution. Furthermore, the Nouméa area generally seems to be more exposed to industrial contamination, whereas Grand Lagon Sud has revealed strong local agricultural pollution (except at intermediate reefs, Table S2). In Grand Nouméa, anguilliform fish have not been exposed to PCB inputs in recent times in view of their low *CB153/CB138* ratio (i.e., ratio<0.8, ≈0.75 in *G. chilospilus* and ≈0.78 in *Conger* sp., Table S2). On the other hand, the PCB inputs in Grand Lagon Sud seem to be more recent mainly in coastal sites, as detected in both species (ratio >1, 1.64 in *G. chilospilus* and 1.04 in *Conger* sp., Table S2). Industrial activities in this area, including the recent construction of a mining factory, could explain in part the continuing inputs of PCBs in this area.

Influence of biological parameters on contamination level

As mentioned in several studies, concentrations of contaminants can also vary according to ecological and biological factors such as fish size (or age), trophic position, and feeding habits or lipid content (Monteiro et al. 1991; Geyer et al. 2000; Penedo de Pinho et al. 2002). These complex relationships can induce interspecific differences in bioaccumulation processes and so emphasize the relevance in detecting the contamination rate at a specific level.

Our results indicated the influence of the three factors tested (body size, trophic position, and lipid content) on several contaminants in both species. The link between body size and bioaccumulation is complex, depending on both the contaminants and the fish species considered. In some cases, trace elements accumulated with increasing fish size (e.g., As, Cd, and Hg in *G. chilospilus*, Table 5), which is consistent with continuous bioaccumulation in muscle during life span (Braune 1987; Burger et al. 2007; Bloom 1992). In some other cases, trace element concentration is negatively correlated with size (e.g., Cu and Fe in both species, or Co, Cr, Ni, Pb, Se, and Zn in *Conger* sp.). This trend could be explained by a decrease in assimilation or by more efficient elimination processes with increasing fish size (Braune 1987; Swaileh and

Adelung 1995; Warnau et al. 1995). Such a relationship can also suggest that an ontogenetic diet shift might occur, implying a variation in the exposure to contaminants through the food pathway, as found in other species (Chouvelon et al. 2011, 2014). However, with the exception of Hg, and to a lesser extent Se, biomagnification of trace elements in trophic webs is still unclear and difficult to identify (Wang 2002). For anguilliform fish from New Caledonia, the trophic position influenced As and Hg concentrations only in *Conger* sp. The POP results are in accordance with their hydrophobic nature, which allows their sequestration in fat tissues during the process of organochlorine accumulation (Robertson and Hansen 2001). Most of them were indeed positively linked to the lipid content of fish, in particular in *Conger* sp., where lipid variation ranged between 0.2 and 5.5 % (Table 1).

Measures of $\delta^{15}N$ in fish confirm the high trophic level of anguilliform fish (Brischoux et al. 2011). When this is associated with other life history traits (size, age, longevity, philopatry, etc.), the detection of high trace element levels appears consistent. These benthic predators are also probably good candidates for monitoring organic pollution because of the strong relationship of POPs with lipid content (even if the flesh of these fish is not very fatty). The specific biological characteristics of each species may well explain interspecific differences in bioaccumulation of contaminants.

Moreover, contamination of anguilliform fish (high trophic level predators) suggests that trophic networks in general are contaminated (Monniot et al. 1994; Hédouin et al. 2006, 2007; Metian et al. 2008b). As a consequence, it is important to carry out contaminant analysis in prey, top predators (e.g., sea snakes, Bonnet 2012), and in the global trophic web. This will provide a more complete assessment of contamination of the environment and lead to better understanding of the mechanisms and processes involved (e.g., bioaccumulation, biomagnification, Bryan 1984; Rainbow and Phillips 1993). A good understanding of both the organisms (i.e., biology, ecology and metabolic capacities) and the trophic network structures, plus environmental conditions, is necessary to understand the dynamics of bioaccumulation and biomagnification throughout the trophic webs (Harmelin-Vivien et al. 2009).

Conclusion

This study strongly confirms a clear accumulation of trace metal contaminants in the muscles of anguilliform fish and the obvious coastal contamination by trace elements linked to mining activities. It is also shown that contamination by POPs occurs in the whole lagoon.

These results raise an alert about complex contaminations throughout the lagoon on a large spatial scale. Various contaminants may show different behavior in abiotic and biotic

environments. Due to their lipophilic nature, POPs have different targets compared with trace elements. This characteristic can explain in part their divergences in mobility in comparison to trace elements. Thus, the more widespread repartition of POPs within the lagoon is probably due to their great capacity of dispersion, through atmospheric transport and current-driven dilution during tropical rain events. This highlights the importance of considering multiple sources of contamination. Moreover, the possibility of a cocktail effect of these various contaminants is not known and could increase the threat. Therefore, in the future, long-term monitoring studies and experiments on toxicity are necessary.

References

Ambastian P, Fernex F, Bernat M, Parron C, Lecolle J (1997) High metal inputs to closed seas: the New Caledonia lagoon. J Geochem Explor 59:59–74

Annema JA, Beurskens JEM, Bodar CWM (1995) Evaluation of PCB fluxes in the environment. The Netherlands: National Institute of Public Health and Environmental Protection Bilthoven, Report N°6010144011. 87 pp

Adams DH, McMichael RH (1999) Mercury levels in four species of sharks from the Atlantic coast of Florida. Fish Bull 97(2):372–379

Ballschmiter K, Zell M (1980) Analysis of polychlorinated biphenyls (PCB) by glass capillary gas chromatography. Fresenius' Zeitschrift für analytische Chemie 302(1):20–31

Bloom NS (1992) On the chemical form of mercury in edible fish and marine invertebrate tissue. Can J Fish Aquat Sci 49(5):1010–1017

Bonnet X (2012) Long-term field study of sea kraits in New Caledonia: fundamental issues and conservation. Integr Comp Biol 52(2):281–295

Bonnet X, Briand MJ, Brischoux F, Letourneur Y, Fauvel T, Bustamante P (2014) Anguilliform fish reveal large scale contamination by trace elements in the coral reefs of New Caledonia. Sci Total Environ 470: 876–882

Braune B (1987) Mercury accumulation in relation to size and age of Atlantic herring (*Clupea harengus harengus*) from the southwestern Bay of Fundy, Canada. Arch Environ Contam Toxicol 16(3):311–320

Brischoux F, Bonnet X, Shine R (2007a) Foraging ecology of sea kraits (*Laticauda* spp.) in the Neo-Caledonian lagoon. Mar Ecol Prog Ser 350:145–151

Brischoux F, Bonnet X, De Crignis M (2007b) A method to reconstruct anguilliform fishes from partially digested items. Mar Biol 151(5): 1893–1897

Brischoux F, Bonnet X (2008) Estimating the impact of sea kraits on the anguilliform fish community (Muraenidae, Congridae, Ophichthidae) of New Caledonia. Aquat Living Resour 21:395–399

Brischoux F, Bonnet X (2009) Life history of sea kraits in New Caledonia. Mémoires du Muséum d'Histoire naturelle 198:133–147

Brischoux F, Bonnet X, Legagneux P (2009) Are sea snakes pertinent bio-indicators for coral reefs? A comparison between species and sites. Mar Biol 156:1985–1992

Brischoux F, Bonnet X, Cherel Y, Shine R (2011) Isotopic signatures, foraging habitats and trophic relationships between fish and seasnakes on the coral reefs of New Caledonia. Coral Reefs 30(1): 155–165

Bryan GW (1984) Pollution due to heavy metals and their compounds. Mar Ecol 5:1289–1431

Burger J, Gochfeld M, Jeitner C, Burke S, Stamm T, Snigaroff R, Snigaroff D, Patrick R, Weston J (2007) Mercury levels and potential risk from subsistence foods from the Aleutians. Sci Total Environ 384(1–3):93–105

Bustamante P, Grigioni S, Boucher-Rodoni R, Caurant F, Miramand P (2000) Bioaccumulation of 12 trace elements in the tissues of the nautilus *Nautilus macromphalus* from New Caledonia. Mar Pollut Bull 40(8):688–696

Bustamante P, Bocher P, Chérel Y, Miramand P, Caurant F (2003) Distribution of trace elements in the tissues of benthic and pelagic fish from the Kerguelen Islands. Sci Total Environ 313(1–3):25–39

Bustamante P, Lahaye V, Durnez C, Churlaud C, Caurant F (2006) Total and organic Hg concentrations in cephalopods from the North Eastern Atlantic waters: influence of geographical origin and feeding ecology. Sci Total Environ 368(2–3):585–596

Chouvelon T, Warnau M, Churlaud C, Bustamante P (2009) Hg concentrations and related risk assessment in coral reef crustaceans, molluscs and fish from New Caledonia. Environ Pollut 157(1):331–340

Chouvelon T, Spitz J, Cherel Y, Caurant F, Sirmel R, Mèndez-Fernandez P, Bustamante P (2011) Species and ontogenic-related differences in $\delta^{13}C$ and $\delta^{15}N$ values and Hg and Cd concentrations of cephalopods. Mar Ecol Prog Ser 433:107–120

Chouvelon T, Caurant F, Cherel Y, Simon-Bouhet B, Spitz J, Bustamante P (2014) Size-related patterns in stable isotopes and mercury concentrations in fish help refining marine ecosystem indicators and evidence distinct management units for hake in the north-eastern Atlantic. ICES J Marine Sci. doi:10.1093/icesjms/fst199

Dalvi AD, Bacon WG, Osborne RC (2004) The past and the future of nickel laterites. In: *PDAC International Convention, Trade Show & Investors Exchange*. Toronto: The prospectors and Developers Association of Canada, 2004. p. 7–10.

de Mora S, Villeneuve J-P, Reza Sheikholeslami M, Cattini C, Tolosa I (2004) Organochlorinated compounds in Caspian Sea sediments. Mar Pollut Bull 48(1–2):30–43

Dierking J, Wafo E, Schembri T, Lagadec V, Nicolas C, Letourneur Y, Harmelin-Vivien ML (2009) Spatial patterns in PCBs, pesticides, mercury and cadmium in the common sole in the NW Mediterranean Sea, and a novel use of contaminants as biomarkers. Mar Pollut Bull 58:1605–1614

Dummee V, Kruatrachue M, Trinachartvanit W, Tanhan P, Pokethitiyook P, Damrongphol P (2012) Bioaccumulation of heavy metals in water, sediments, aquatic plant and histopathological effects on the golden apple snail in Beung Boraphet reservoir, Thailand. Ecotoxicol Environ Saf 86:204–212

Eilser R (2010) Compendium of trace metals and marine biota 2: vertebrates. Elsevier, 522 p

Fauvel T, Brischoux F, Briand MJ, Bonnet X (2012) Do researchers impact their study populations? Assessing the effect of field procedures in a long term population monitoring of sea kraits. *Amphibia-Reptilia*:1–8

Fernandez J-M, Ouillon S, Chevillon C, Douillet P, Fichez R, Gendre RL (2006) A combined modelling and geochemical study of the fate of terrigenous inputs from mixed natural and mining sources in a coral reef lagoon (New Caledonia). Mar Pollut Bull 52(3):320–331

Fernandez J–M and Breau L (2011) Impact potentiel de l'effluent de Vale NC sur le milieu marin (2005–2011). Rapport AEL 110328–OE–03, 209p

Fichez R, Breau L, Chevillon C, Chifflet S, Douillet P, Faure V, Fernandez J-M, Gérard P, Hédouin L, Lapetite A, Ouillon S, Pringault O, Torréton J-P (2008) Origine, transport et devenir des apports naturels et anthropiques dans le lagon sud-ouest de Nouvelle-Calédonie. *Le Journal de la Société des Océanistes*, 126–127:41–58

Geyer H, Rimkus G, Scheunert I, Kaune A, Schramm K-W, Kettrup A, Zeeman M, Muir DG, Hansen L, Mackay D (2000) Bioaccumulation and occurrence of endocrine-disrupting chemicals (EDCs), persistent organic pollutants (POPs), and other organic compounds in fish and other organisms including humans. Bioaccumulation, new aspects and developments. Handbook of environmental chemistry 2-J. Springer Verlag, Berlin, pp 1–166

Harmelin-Vivien ML, Cossa D, Crochet S, Banaru D, Letourneur Y, Mellon-Duval C (2009) Difference of mercury bioaccumulation in red mullets from the north-western Mediterranean and Black seas. Mar Pollut Bull 58:679–685

Hédouin L, Metian M, Teyssié J-L, Fowler SW, Fichez R, Warnau M (2006) Allometric relationship in the bioconcentration of heavy metals by the edible tropical clam *Gafrarium tumidum*. Sci Total Environ 336:154–163

Hédouin L, Pringault O, Metian M, Bustamante P, Warnau M (2007) Nickel bioaccumulation in bivalves from the New Caledonia lagoon: seawater and food exposure. Chemosphere 66(8):1449–1457

Hédouin L, Bustamante P, Fichez R, Warnau M (2008) The tropical brown alga *Lobophora variegata* as a bioindicator of mining contamination in the New Caledonia lagoon: a field transplantation study. Mar Environ Res 66(4):438–444

Hédouin L, Bustamante P, Churlaud C, Pringault O, Fichez R, Warnau M (2009) Trends in concentrations of selected metalloid and metals in two bivalves from the coral reefs in the SW lagoon of New Caledonia. Ecotoxicol Environ Saf 72(2):372–381

Hédouin L, Batista MG, Metian M, Buschiazzo E, Warnau M (2010) Metal and metalloid bioconcentration capacity of two tropical bivalves for monitoring the impact of land-based mining activities in the New Caledonia lagoon. Mar Pollut Bull 61(7–12):554–567

Hédouin L, Pringault O, Bustamante P, Fichez R, Warnau M (2011) Validation of two tropical marine bivalves as bioindicators of mining contamination in the New Caledonia lagoon: field transplantation experiments. Water Res 45(2):483–496

Hellawell JM (1988) Toxic substances in rivers and streams. Environ Pollut 50(1–2):61–85

Ineich I, Bonnet X, Brischoux F, Kulbicki M, Séret B, Shine R (2007) Anguilliform fishes and sea kraits: neglected predators in coral-reef ecosystems. Mar Biol 151(2):793–802

Johnson GW, Quensen III JF, Chiarenzelli JR, Hamilton MC (2005) Polychlorinated biphenyls. In: Morrison, R., Murphy, B. (Eds.), *Environmental forensics.* Burlington, MA, Academic Press, pp. 187–225

Kojadinovic J, Potier M, Le Corre M, Cosson RP, Bustamante P (2007) Bioaccumulation of trace elements in pelagic fish from the Western Indian Ocean. Environ Pollut 146(2):548–566

Lodenius M (1991) Mercury concentrations in an aquatic ecosystem during twenty years following abatement the pollution source. Water Air Soil Pollut 56(1):323–332

Metian M, Bustamante P, Hédouin L, Warnau M (2008a) Accumulation of nine metals and one metalloid in the tropical scallop *Comptopallium radula* from coral reefs in New Caledonia. Environ Pollut 152(3):543–552

Metian M, Giron E, Borne V, Hédouin L, Teyssié J-L, Warnau M (2008b) The brown alga *Lobophora variegata*, a bioindicator species for surveying metal contamination in tropical marine environments. J Exp Mar Biol Ecol 362(1):49–54

Metian M, Hédouin L, Eltayeb M, Lacoue-Labarthe T, Teyssié J-L, Mugnier C, Bustamante P, Warnau M (2010) Metal and metalloid bioaccumulation in the Pacific blue shrimp *Litopenaeus stylirostris* (Stimpson) from New Caledonia: laboratory and field studies. Mar Pollut Bull 61(7–12):576–584

Metian M, Warnau M, Chouvelon T, Pedraza F, Rodriguez y Baena A, Bustamante P (2013) Trace element bioaccumulation in reef fish from New Caledonia: influence of trophic groups and risk assessment for consumers. Mar Environ Res 87–88:26–36

Monniot F, Martoja R, Monniot C (1994) Cellular sites of iron and nickel accumulation in ascidians related to the naturally and anthropic enriched New Caledonian environment. Ann Inst Oceanogr 70: 205–216

Monod JL, Arnaud PM, Arnoux A (1995) PCB congeners in the marine biota of Saint Paul and Amsterdam Islands, Southern Indian Ocean. Mar Pollut Bull 30(4):272–274

Monteiro LR, Isidro EJ, Lopes HD (1991) Mercury content in relation to sex, size, age and growth in two scorpionfish (*Helicolenus dactylopterus* and *Pontinus kuhlii*) from Azorean waters. Water Air Soil Pollut 56(1):359–367

Murphy PG (1972) Sulfuric acid for the cleanup of animal tissues for analysis of acid stable chlorinated hydrocarbons residues. J Assoc Off Anal Chem 55:45–53

Penedo de Pinho A, Guimaraes JRD, Martins AS, Costa PAS, Olavo G, Valentin J (2002) Total mercury in muscle tissues of five shark species from Brazilian offshore waters: effects of feeding habit, sex, and length. Environ Res Sec A 89:250–258

Pernice M, Boucher J, Boucher-Rodoni R, Joannot P, Bustamante P (2009) Comparative bioaccumulation of trace elements between *Nautilus pompilius* and *Nautilus macromphalus* (Cephalopoda: Nautiloidea) from Vanuatu and New Caledonia. Ecotoxicol Environ Saf 72(2):365–371

Peterson BJ, Fry B (1987) Stable isotopes in ecosystem studies. Annu Rev Ecol Syst 18:293–320

Phillips DJH (1995) The chemistries and environmental fates of trace metals and organochlorines in aquatic ecosystems. Mar Pollut Bull 31(4–12):193–200

Pinnegar JK, Polunin NVC (1999) Differential fractionation of δ^{13}C and δ^{15}N among fish tissues: implications for the study of trophic interactions. Funct Ecol 13(2):225–231

Rainbow PS, Phillips DJ (1993) Cosmopolitan biomonitors of trace metals. Mar Pollut Bull 26(11):593–601

Reed RN, Shine R, Shetty S, Montgomery WL (2002) Sea kraits (Squamata: *Laticauda* spp.) as a useful bioassay for assessing local diversity of eels (Muraenidae, Congridae) in the Western Pacific Ocean. Copeia 2002(4):1098–1101

Richmond RH (1993) Coral reefs: present problems and future concerns resulting from anthropogenic disturbance. Am Zool 33:524–536

Robertson LW, Hansen LG (2001) PCBs: Recent advances in environmental toxicology and health effects. Lexington, Kentucky, The University Press of Kentucky

Sinkkonen S, Paasivirta J (2000) Degradation half-life times of PCDDs, PCDFs and PCBs for environmental fate modeling. Chemosphere 40(9–11):943–949

Salvat B, Roche H, Berny P, Ramade F (2012) Recherches sur la contamination par les pesticides d'organismes marins des réseaux trophiques récifaux de Polynésie Française. Société nationale de protection de la nature et d'acclimatation de France, Paris, p 19

Swaileh KM, Adelung D (1995) Effect of body size and season on the concentrations of Cu, Cd, Pb and Zn in *Diastylis rathkei* (kröyer) (Crustacea: Cumacea) from Kiel Bay, Western Baltic. Marine Pollution Bulletin 31(1–3):103–107

Valoppi L, Petreas M, Donohoe RM, Sullivan L, Callahan CA (2000) *Use of PCB congener and homologue analysis in ecological risk assessment*. USA: American Society for Testing and Materials. In: Price FT,

Brix KV, Lane NK (eds) Environmental toxicology and risk assessment: recent achievements in environmental fate and transport, vol 9. ASTM International, West Conshohocken, pp 147–161

Van Ael E, Covaci A, Blust R, Bervoets L (2012) Persistent organic pollutants in the Scheldt estuary: environmental distribution and bioaccumulation. Environ Int 48:17–27

Wafo E, Sarrazin L, Diana C, Dhermain F, Schembri T, Lagadec V, Pecchia M, Rebouillon P (2005) Accumulation and distribution of organochlorines (PCBs and DDTs) in various organs of *Stenella coeruleoalba* and a *Tursiops truncatus* from Mediterranean littoral environment. Sci Total Environ 348:115–127

Wafo E, Risoul V, Schembri T, Lagadec V, Dhermain F, Mama C, Portugal H (2012) PCBs and DDTs in *Stenella coeruleoalba* dolphins from the French Mediterranean coastal environment (2007–2009): current state of contamination. Mar Pollut Bull 64(11):2535–2541

Wang WX (2002) Interactions of trace metal and different marine food chains. Mar Ecol Prog Ser 243:295–309

Warnau M, Ledent G, Temara A, Alva V, Jangoux M, Dubois P (1995) Allometry of heavy metal bioconcentration in the echinoid *Paracentrotus lividus*. Arch Environ Contam Toxicol 29(3):393–399

Wells DE, Cowan A, Christie AEG (1985) Separation of organochlorine residue by absorption chromatography prior to capillary gas chromatography. J Chromatogr 328:372–377