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Trace element accumulation in the muscles of reef fish collected from southern new Caledonian lagoon: Risk assessment for consumers and grouper *Plectropomus leopardus* as a possible bioindicator of mining contamination

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ABSTRACT

Flesh of 141 fish specimens collected along the southern coast of New Caledonia, close to the mining industry Prony Resources New Caledonia, were analyzed for 10 elements (As, Cd, Co, Cr, Cu, Fe, Hg, Mn, Ni and Zn). The leopard coral grouper *Plectopomus leopardus* revealed significant spatial variations for Cr, Fe, Mn and Zn and size-dependent accumulation of Hg. Sanitary risk assessment suggests that Hg and Me-Hg could potentially be a concern for heavy fish consumers. A previous study in New Caledonia had demonstrated the capacity of *P. leopardus* to differentially accumulate Ag, Cd, Cu, Hg and Zn and as such its potential as bioindicator specie to monitor contamination status in urban areas (Metian et al., 2013). Our results demonstrate that this specie can also to be used as a bioindicator to monitor the contamination status of Cr, Fe and Mn in New Caledonian lagoon in relation to mining activities.

1. Introduction

Seafood, particularly fish, is of primary importance in the Pacific islands as it represents, for many island countries of the region, a prime source of protein (Labrosse et al., 2006) and is a major contributor to its economy (Charlton et al., 2016). Fish consumption in the Pacific on a *per capita* basis is among the highest in the world, particularly in the Polynesian and Micronesian atoll countries, and in most of Melanesia (SPC, 2011). However, consumption of fish is subject to concern as it is a known important pathway of human exposure to a variety of chemical contaminants (Marti-Cad et al., 2007; Storelli, 2008). Numerous health benefits provided by fish consumption may thereby be compromised by the presence of contaminants, which can have harmful effects on human body if consumed in toxic quantities (Bosch et al., 2016).

Among the contaminants, the heavy metals, mercury (Hg), cadmium (Cd) and lead (Pb) pose real threats to human health as these elements tend to accumulate in marine organisms. Extensively studied, these heavy metals are known to cause acute and chronic disorders in human (Järup, 2003). Increasing anthropogenic activities such as urbanization, agriculture, industrialization and the development of mining activities lead to environmental pollution through effluents and emanations (Ashraf et al., 2012) and to serious illness such as Minamata diseases linked with methylmercury (Me-Hg) bioaccumulation (Hachiya, 2006) and Itai-itai disease, a severe form of chronic Cd poisoning (Inaba et al., 2005).

Located in the Southwest Pacific Ocean, New Caledonia is surrounded by a barrier reef of 1600 km, which makes it the second largest lagoon in the world (Labrosse et al., 2000). New Caledonia has also the longest history of mining in the Pacific islands. The New Caledonian soils are naturally rich in cobalt (Co), chromium (Cr), iron (Fe), manganese (Mn) and nickel (Ni). According to estimates, New Caledonia disposes of around 25 % of the global nickel resource and is the third largest producer of this metal in the world (Savy et al., 2011).

From intense mining activities and from natural erosion of soils

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associated with tropical rainfall, New Caledonian coastal waters are subjected to large inputs of metals (Ambatsian et al., 1997; Fernandez et al., 2006; Debenay and Fernandez, 2009). Thereby, it constitutes a threat to the local coastal ecosystems and the quality of local seafood. Since 2000, a handful of toxicological studies have been conducted in New Caledonia on marine species, ranging from consumed bivalves to non-consumed ascidians, brown alga, fish, nautilus and whales. These studies have largely focused on the bioaccumulation of trace metals into biological tissues and the validation of certain organisms as bio-indicators (Bonnet et al., 2014; Briand et al., 2014; Bustamante et al., 2000, 2003; Hédouin et al., 2006, 2007, 2008, 2009, 2010a, 2010b, 2010c, 2011, 2016, 2018; Metian et al., 2005, 2008a; Metian and Warnau, 2008; Monniot et al., 1994; Pernice et al., 2009). Inversely, little attention has been given to health risk assessment for human consumers. Indeed, so far, only 4 studies have considered the risk for New Caledonian consumers (Chouvelon et al., 2009; Metian et al., 2008b, 2010, 2013). Information on the contamination status of the New Caledonian lagoon and its edible products remains therefore largely incomplete (Labrosse et al., 2000). Given the growing intensity of mining activities in New Caledonia (opening of two news mining complexes in the last twenty years) and the importance of seafood as nutrition, such information is crucial for risk management.

The main objective of the present study is therefore to complement the baseline information on concentration levels of elements in commercial edible coastal fish species and to establish a risk assessment for the consumers. From five fishing grounds located off the southern tip of New Caledonia, a large population of comestible coral reef fish was analyzed for the following ten trace elements: As, Cd, Co, Cr, copper (Cu), Fe, Hg, Mn, Ni and zinc (Zn). These fishing sites are situated in the vicinity of the nickel hydrometallurgical extraction plant of Prony Resources New Caledonia (PRNC; formerly known as Vale-NC) which started construction in 2000. The current study was undertaken in 2012 as part of their environmental impact monitoring programs.

2. Material and method

2.1. Sampling sites and sampling population

Situated between 300 m to 10 km off the coast, 5 sampling sites, Bonne Anse (BA), Baie Kwe (BK), Canal de la Havannah (CH), Ile Ouen (IO), and Port de Goro (BG) were selected as sampling zones (Fig. 1). These sites contour the southern tip of the mainland of New Caledonia from southwest to southeast.

Of these 5 sites, BA, BK and CH have been identified, *via* prior hydrodynamics prediction model, as potentially impacted from current mining activities of PRNC (Douillet and Fernandez, 2009). BK is a drainage basin located in an enclosed zone with very weak tidal water mass exchange and is subjected to significant soil runoff due to the opencast mines of PRNC in its catchment basin. BA, with a higher tidal water mass exchange, is subjected to both natural terrigenous influences from former mines and from the currently operating refinery complex and the industrial port of PRNC. BG has also a higher tidal water mass exchange rate than that of the BK and can temporarily be subjected to soil runoffs from former opencast mines. IO and particularly CH are zones situated farthest from the industrial complex and are directly influenced by the tide, which allows oceanic water masses to enter through the Havannah pass. However, in CH, treated industrial liquid effluents are discharged *via* a 1 km long diffuser; this site can therefore be potentially impacted by industrial activity (Fig. 1).

These 5 sites are also consistent with local fishing habits as per the data collected *via* a small fishing survey carried in 2009 where professional and non-professional fishermen of nearby tribes were briefly questioned on their fishing habits (grounds fished, fishing techniques and equipment used, and fish species targeted).--

From 23 to 27 January 2012, 141 fish belonging to 29 species from three trophic groups (Table 1; see appendix X) were gathered by free-diving with spear guns regardless of size, length and sex. The three

Fig. 1. Location of the fishing sites. Situated off the Southern coast of mainland New Caledonia, Bonne Anse (BA) and island of Ile Ouen (IO) is located in the southwest; Port de Goro (BG) in the southeast and in the south Baie Kwe (BK) and the Canal de la Havannah (CH).

Table 1
Characteristics of fish specimen collected from the 5 fishing sites of the Southern lagoon of New Caledonia (n = 141).

Fishing site and Specie collected	N	Local name	Common name	Mean wet weight (g)	Mean caudal length (cm)	Trophic group (index)
BA	27					
Acanthurus mata	1	Chirurgien	Elongate surgeonfish	395	27	H (2.5)
Chlorurus sordidus	3	Perroquet	Daisy parrotfish	552 ± 133	29.3 ± 1.5	H (2)
Choerodon graphicus	1	Perroquet Wallis	Graphic tuskfish	1920	46	C (3.5)
Naso hexacanthus	1	Ume	Sleek unicornfish	420	30	H (3.3)
Naso unicornis	7	Dawa	Bluespine unicornfish	1955 ± 732	45.9 ± 6.5	H (2.2)
Plectropomus leopardus	10	Loche saumonée	Leopard coralgrouper	891 ± 326	40.6 ± 4.5	P (4.5)
Scarus frenatus	1	Perroquet	Bridled parrotfish	910	35.5	H (2)
Scarus niger	1	Perroquet	Dusky parrotfish	375	27.5	H (2)
Scarus rivulatus	2	Perroquet	Rivulated parrotfish	468 ± 117	28.0 ± 1.4	H (2)
BG	26					
Aprion virescens	1	Mekoua	Green jobfish	3995	65	P (4)
Cetoscarus bicolor	2	Perroquet	Bicolour parrotfish	1170 ± 127	39.3 ± 1.8	H (2)
Chlorurus microrhinos	4	Perroquet bleu	Steephead parrots	1515 ± 1458		H (2)
Chlorurus sordidus	1	Perroquet	Daisy parrotfish	570	30.5	H (2)
Choerodon graphicus	2	Perroquet Wallis	Graphic tuskfish	1188 ± 385	37.5 ± 3.5	C (3.5)
Naso unicornis	2	Dawa	Bluespine unicornfish	1398 ± 1213	40.0 ± 14.1	H (2.2)
Plectorhinchus chaetodonoides	1	Loche castex	Harlequin sweetlips	2050	47.5	C (3.8)
Plectropomus laevis	2	Loche saumonée	Blacksaddled coralgrouper	3590 ± 2673	60.5 ± 13.4	P (4.1)
Plectropomus leopardus	7	Loche saumonée	Leopard coralgrouper	786 ± 305	38.7 ± 4.9	P (4.5)
Scarus frenatus	2	Perroquet	Bridled parrotfish	718 ± 187	33.0 ± 2.8	H (2)
Scarus ghobban	1	Perroquet	Blue-barred parrotfish	370	29	H (2)
Scarus prianatus	1	Perroquet	–	775	35	H (2)
BK	24					
Chlorurus microrhinos	1	Perroquet bleu	Steephead parrots	1120	36	H (2)
Chlorurus sordidus	1	Perroquet	Daisy parrotfish	715	32	H (2)
Kyphosus sp	2	Wi Wa	–	1150 ± 665	36.3 ± 6.7	H (2)
Naso unicornis	1	Dawa	Bluespine unicornfish	610	31	H (2.2)
Plectorhinchus chaetodonoides	1	Loche castex	Harlequin sweetlips	5450	4	C (3.8)
Plectropomus laevis	1	Loche saumonée	Blacksaddled coralgrouper	580	37	P (4.1)
Plectropomus leopardus	8	Loche saumonée	Leopard coralgrouper	1521 ± 830	45.7 ± 9.0	P (4.5)
Plectropomus maculatus	1	Loche saumonée	Spotted coralgrouper	2340	56	P (4.1)
Scarus altipinnis	1	Perroquet	Filament-finned parrotfish	835	35	H (2)
Scarus rivulatus	5	Perroquet	Rivulated parrotfish	563 ± 158	30.3 ± 2.7	H (2)
Scarus schlegeli	2	Perroquet	Yellowband parrotfish	350 ± 177	29.4 ± 2.0	H (2)
CH	29					
Acanthurus dussumieri	1	Picot kanak	Eyestripe surgeonfish	640	30	H (2)
Acanthurus mata	1	Chirurgien	Elongate surgeonfish	750	33	H (2.5)
Chlorurus microrhinos	1	Perroquet bleu	Steephead parrots	2760	52	H (2)
Epinephelus maculatus	1	Loche de sable	Highfin grouper	910	35	C (3.9)
Naso unicornis	5	Dawa	Bluespine unicornfish	2184 ± 316	45.7 ± 1.6	H (2.2)
Plectorhinchus picus	2	Loche castex	Painted sweetlip	2658 ± 534	57.0 ± 2.8	C (3.9)
Plectropomus leopardus	9	Loche saumonée	Leopard coralgrouper	2453 ± 426	53.3 ± 3.6	P (4.5)
Scarus forstenu	1	Perroquet	–	760	30	H (2)
Scarus frenatus	3	Perroquet	Bridled parrotfish	713 ± 81	31.7 ± 2.5	H (2)
Scarus ghobban	1	Perroquet	Blue-barred parrotfish	1570	43	H (2)
Scarus niger	1	Perroquet	Dusky parrotfish	565	29	H (2)
Scarus rivulatus	1	Perroquet	Rivulated parrotfish	880	34	H (2)
Scarus rubroviolaceus	2	Perroquet	Ember parrotfish	2410 ± 587	49.5 ± 4.9	H (2)
IO	35					
Acanthurus dussumieri	3	Picot kanak	Eyestripe surgeonfish	492 ± 232	26.2 ± 2.9	H (2)
Chlorurus microrhinos	1	Perroquet bleu	Steephead parrots	565	29	H (2)
Chlorurus sordidus	3	Perroquet	Daisy parrotfish	777 ± 266	32.2 ± 3.2	H (2)
Cromileptes altivelis	2	Loche truite	Humpback grouper	1285 ± 127	43.5 ± 0.7	P (4.5)
Lethrinus atkinsoni	1	Petit bec de cane	Pacific yellowtail emperor	605	31	C (3.5)
Naso unicornis	3	Dawa	Bluespine unicornfish	845 ± 640	33.2 ± 7.3	H (2.2)
Parupeneus cyclostomus	1	Barbet à selle d'or	Gold-saddle goatfish	1280	43	P (4.2)
Plectropomus laevis	1	Loche saumonée	Blacksaddled coralgrouper	1880	55	P (4.1)
Plectropomus leopardus	15	Loche saumonée	Leopard coralgrouper	1431 ± 936	44.9 ± 11.0	P (4.5)
Scarus frenatus	1	Perroquet	Bridled parrotfish	830	35	H (2)
Scarus ghobban	1	Perroquet	Blue-barred parrotfish	1205	38	H (2)
Scarus rivulatus	2	Perroquet	Rivulated parrotfish	595 ± 64	32.3 ± 1.1	H (2)
Scomberomorus commerson	1	Tazar	Narrow-barred Spanish mackerel	5345	86	P (4.5)

H: herbivore; C: Carnivore; P: Piscivore.

trophic groups collected are:

- 17 herbivorous species with trophic index values between 2 and 2.5 (n = 73);
- 5 carnivorous species with trophic index values between 3.5 and 3.9 (n = 9); and
- 7 piscivorous species with trophic index values above 4 (n = 59).

Among the 29 species, the piscivore *Plectropomus leopardus* is well represented in the sampled population with 49 individuals in total. Moreover, this specie is also reasonable well dispatched over the 5 studied sites with 7–15 individuals per site (Table 1; see appendix X).

2.2. Sample preparation and trace element analysis

Once caught, the fish were first identified, then weighed and caudal fork lengths measured. Due to the implications, it carries for human consumption and health risk, the muscle (flesh) was chosen. Dissected into filets, the tissues were transferred into individual referenced bags and stored at −20 °C. Around 5 g of each sample were then freeze-dried. Freeze-dried tissues were then weighed, grounded and stored in a desiccator.

The analysis of As, Cd, Co, Cr, Cu, Fe, Hg, Mn, Ni and Zn in biological tissues required the mineralization of the samples in a microwave oven (Milestone, Ethos1). Aliquots of around 500 mg of the grounded tissues were taken and digested with a solution of 4 mL nitric acid (67 %) and 1 mL hydrogen peroxide (30 %) in hyperbaric conditions (180 °C, 100 b). The reagents used were of Suprapur™ quality.

Once mineralized, the samples are transferred into 20 mL pre-cleaned HDPE vials and diluted to 20 mL with ultrapure quality water (R = 18.2 MΩ•cm). All elements were analyzed by ICP-OES (Perkin Elmer 730ES Varian), except Hg which was analyzed by ICP-MS (Agilent Technologies 7500CX series).

The digestion method and analysis were verified by inserting into each series a reagent blank and samples of certified reference materials of marine origin (TORT-2 corresponding to NRC lobster hepatopancreas) treated identically to natural sample. The recoveries ranged from 80 % to 108 % for all elements and the relative standard deviations were below 8 %, thus proving a good accuracy and repeatability of the method. The limits of quantification (LQ expressed as μg/g dry weight) for each element are as follows: 1.00 (As); 0.050 (Cd and Hg); 0.200 (Co and Ni); 0.100 (Cr, Cu and Mn); 2.00 (Fe) and 10.0 (Zn).

2.3. Data analysis

The concentration obtained in μg/g dry weight (dry wt) were converted to μg/g wet weight (wet wt), using the dry wt/wet wt ratios measured. These ratios indicated a mean moisture content of 80 % for fish flesh.

For certain elements and samples, values below the LQ were observed. The elimination of these non-quantified values from the statistical processing reduces significantly the number of samples to be treated, with the consequence on the interpretation of data. In order to reduce statistical bias as much as possible, the data of the entire fish population collected was considered; it was therefore agreed that all values below the LQ would be replaced by LQ/3 (=limit of detection; LD).

Numerous studies suggested that bioaccumulation patterns in aquatic organisms are related to their diet and thus to their trophic level and taxonomic affiliation. For example, larger apex predators have been reported (Medieu et al., 2021; Gentes et al., 2021) to bio-magnify certain contaminants like Hg. Another factor reported is the age of individuals (Vieira et al., 2020).

The 5 sampling sites characterized by their proper hydrodynamic and anthropogenic conditions can also influence the availability of different metals. Therefore, the spatial distribution can also be a structuring factor for bioaccumulation patterns. As a result, in the present study, the factors, trophic groups, fishing sites and species, were considered in order to examine the possible bioaccumulation patterns in all collected fishes.

The representation of the specie *P. leopardus* in this study, in terms of number of specimens, allowed further the exploration of spatial effects. The effect of fish physiological stage on the accumulation of different trace elements was also explored with this specie. As the growth-size relationship in *P. leopardus* is locally well-documented, caudal length of *P. leopardus* was used as a proxy to calculate age with von Bertalanfy growth equation (Loubens, 1980; Preuss, 2012).

All variations in metals concentrations were investigated using one-way statistical tests. As all datasets showed significant departures from ANOVA's assumptions, non-parametric tests (Kruskal-Wallis tests) were used. ANCOVA were also performed. Bar and scatter graph representations of metal concentrations among these factors were generated to complement the interpretation of the statistical results.

2.4. Risk assessment for human consumers

To evaluate the health risks derived from metal exposure through consumption of fish from these sites, toxicological data provided by the Joint Expert Committee on Food Additives (JECFA) and European Food Safety Authority (EFSA) for different metals were considered. These include the Provisional Maximum Tolerable Daily Intake (PMTDI) for Cu, Fe, and Zn, Provisional Tolerable Weekly Intake (PTWI) for Hg and Provisional Tolerable Monthly Intake (PTMI) for Cd established by JECFA and the Tolerable Daily Intake (TDI) provided by EFSA for Cr and Ni. These data and the mean concentrations measured for these 7 trace elements in the present study were then used to calculate the different Maximum Safe Consumption (MSC) thresholds that a 70 kg adult should not exceed while consuming fish caught in the studied zone.

In order to determine if these MSC limits are indeed being reached, knowledge on the average consumption of seafood by the local population is required. While no such specific data exist for the southern region of the main island of New Caledonia, two studies in the north (Labrosse et al., 2006) and on island of Ouvea (Leopold et al., 2004), have been considered. They showed that the average annual consumption (52 weeks) ranges from 28.0 ± 2.0 kg *per capita* to 63 ± 9.7 kg */ per capita*, respectively. Based on these two consumption rates, daily, weekly and monthly fish quantity ingestion have been estimated and compared with the calculated MSC thresholds for each element of interest.

Furthermore, based on the mean concentration of total Hg (T-Hg) measured in *P. leopardus* fish, we have also evaluated in this fish specie the MSC threshold for Me-Hg by taking a conversion ratio of 100 % for Me-Hg/T-Hg as per Chouvelon et al., 2009 and the JECFA-established PTWI data.

3. Results

3.1. Accumulation patterns in all species

The trace element concentrations measured in the muscles of 141 fish are given in appendix X. All species included, the mean concentrations and the ranges of values measured are presented in Table 2. Of the 10 elements, the values of Cd remained mainly under the LD and as such were not further analyzed. The mean extremes ranged from 0.020 for Ni to 2.12 μg/g wet wt for Zn.

The trace element concentrations measured in the fish muscle were combined in a PCA which indicated that Cr and Fe concentrations and, to a lesser extent, that of Ni were the variables determining the variations in the first axis whereas that of Co and Hg and, to a lesser extent, that of Zn concentrations contributed to the variations of the second axis (Fig. 2).

While the rich lateritic ores of the New Caledonia present high correlations between the elements Co, Cr, Fe, Mn and Ni (Fernandez et al., 2017; Quantin et al., 2002), in fish flesh, we found only a single strong correlation with Fe and Cr (r = 0.734) (Table 3).

With exception for Cr, Ni and Zn, the influence of trophic groups was detected with significant differences in the levels of bioaccumulation for other metals between the herbivores, carnivores and piscivores. For Cu, Fe and Mn, these effects corresponded either to minor variations between groups (in terms of relative magnitude) or to the punctual effect of a few high values. On the other hand, the other elements (As, Co and Hg) showed differences that were both significant and of large magnitude between groups (Table 4; Fig. 3).

Of worth noting, Hg appeared remarkably higher in carnivores and piscivores compared to herbivores where all concentrations remained below the LQ. As for As, the concentrations are remarkably higher in carnivores while those of Co were higher in herbivores compared to two other groups (Fig. 3).

Table 2
Mean concentrations ± confidence interval of trace elements in the fish flesh and lower and higher range values (n = 141).

Element (µg/g wet wt)	As	Cd	Co	Cr	Cu	Fe	Hg	Mn	Ni	Zn
Minimal value	<0.067	<0.003	<0.013	<0.007	0.051	<0.133	<0.003	<0.007	<0.013	<0.667
Maximal value	24.5	0.015	0.654	0.726	0.489	14.5	0.384	0.465	0.196	10.5
Mean	1.41	0.003	0.065	0.043	0.207	1.63	0.053	0.072	0.020	2.12
Confidence interval	0.50	0.000	0.017	0.011	0.019	0.31	0.012	0.010	0.004	0.30

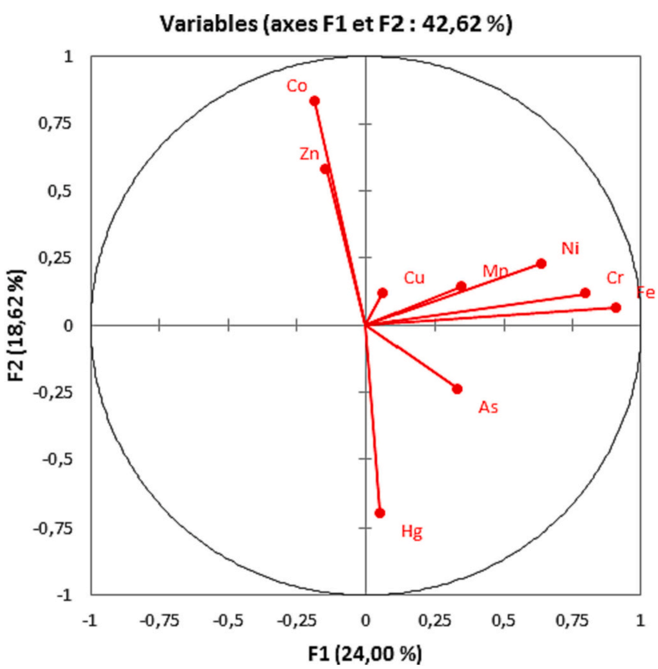

Fig. 2. Plot of the principal component analysis (PCA) showing global distribution of variables in all species (n = 141).

The effect of the sampling location on element concentrations is studied in the Table 4 and represented in the Figs. 4, 5 and 6. For all species, significant spatial effects were detected for Cr, Fe, Ni and Zn (Table 4). Regarding Cr, Fe and Ni, the effects detected corresponded to higher concentrations in BK and in BA. For Zn, the variation corresponded to a higher concentration in BA and a lower concentration in IO (Fig. 4).

The detailed effects by trophic groups showed no spatial influence in carnivores, whereas herbivores and piscivores displayed similar patterns of significant variations for Cr, Fe and Zn. Metal concentrations in piscivores appeared particularly sensitive to sampling location, with additional effects on Mn and Ni concentrations (Table 4). Indeed, in both herbivores and piscivores (Figs. 5 and 6), the concentrations of the lateritic metals Cr, Fe and Mn measured, are the highest in individuals caught from BK. The BA area also shows higher concentrations for Cr, Fe and Mn but with a greater variability. The CH presents the lowest concentrations while the BG and OI show transitional concentrations levels. The concentrations of Ni, equally distributed between BK and CH, were only quantified (>LQ) in piscivores caught from these two sites. As observed earlier, Zn concentrations in herbivores and especially in piscivores are the lowest in IO, with important variations for the other sites (Figs. 5 and 6).

3.2. *Accumulation patterns in P. leopardus*

As the grouper *P. leopardus* is well-represented, the accumulation patterns and the effects of sampling location and of size on elements are closely studied with this specie. The trace element concentrations measured in the muscles of 49 samples of *P. leopardus* are given in appendix X and the mean concentrations and the ranges of values detected are presented in Table 5; among the 10 elements, all the values of Cd remained under the LD. The mean extremes ranged from 0.016 for Co to 1.72 µg/g wet wt for Zn.

The trace element concentrations measured in the *P. leopardus* muscle were combined in a PCA which indicated that Cr and Fe concentrations were the variables determining the variations in the first axis and that of Co, Hg and Mn concentrations contributed to the variations of the second axis (Fig. 7).

The correlation observed for Fe and Cr in all sampling population is amplified in *P. leopardus* species with a robust r value of 0.949 (Table 6).

Though differing only in magnitude, the spatial distribution patterns of metal concentration observed in *P. leopardus,* are basically identical to those observed in piscivores; significant spatial variations were also revealed for Cr, Fe, Mn and Zn (Table 7). As in piscivores, the concentration of Cr, Fe and Mn are highest in BK and lowest in CH with intermediary concentrations in BG and IO. BA presents also higher concentrations, however, with a greater standard deviation. With regards to Zn, the concentrations are the lowest in IO with important variations for the other sites as was observed in piscivores. Ni is the only element that presents an exception; while the variations observed in piscivores were significant, those observed in *P. leopardus* are non-significant (Fig. 8).

The statistical studies on *P. leopardus* have also shown variations for Hg, Mn and As concentrations in relation to size (Table 7). While this variation of minor amplitude for the metalloid As, it is highly significative for Hg and Mn. An ANCOVA was performed to assess a possible interaction between spatial distribution (sampling location) and size for all trace elements and showed no significant effect of this interaction except for Co (data not shown).

Using von Bertalanfy growth equation (Loubens, 1980; Preuss,

Table 3
Pearson correlation matrix of trace element concentrations (n = 141).

Variables	As	Co	Cr	Cu	Fe	Hg	Mn	Ni	Zn
As	**1**								
Co	−0.096	**1**							
Cr	−0.032	−0.066	**1**						
Cu	0.176	0.092	−0.044	**1**					
Fe	0.435	−0.034	**0.734**	0.038	**1**				
Hg	0.241	−0.375	−0.014	−0.032	0.084	**1**			
Mn	−0.025	−0.069	0.100	0.156	0.147	−0.254	**1**		
Ni	−0.010	0.013	0.345	−0.027	0.419	−0.111	0.250	**1**	
Zn	0.082	0.491	−0.032	0.016	0.045	−0.065	−0.214	−0.062	**1**

In bold: r > 0.600.

Table 4
Results of the Kruskal-Wallis tests analyzing the effect of trophic groups (herbivores, carnivores and piscivores), the effect of sampling location, all species included and according to trophic groups, and the effect related to the taxonomy on element concentrations (n = 141).

Variables	Trophic group effect (n = 141)	Spatial (sampling location) effect				Taxonomic group effect (n = 141)
		All species (n = 141)	Carnivores (n = 9)	Herbivores (n = 73)	Piscivores (n = 59)	
As	***	ns	ns	ns	ns	***
Co	***	ns	ns	ns	ns	***
Cr	ns	***	ns	***	**	*
Cu	*	ns	ns	ns	ns	***
Fe	***	***	ns	**	*	***
Hg	***	ns	ns	ns	ns	***
Mn	***	ns	ns	ns	***	***
Ni	ns	*	ns	ns	*	ns
Zn	ns	***	ns	*	***	***

ns: not significant
* P < 0.05
** P < 0.01
*** P < 0.001

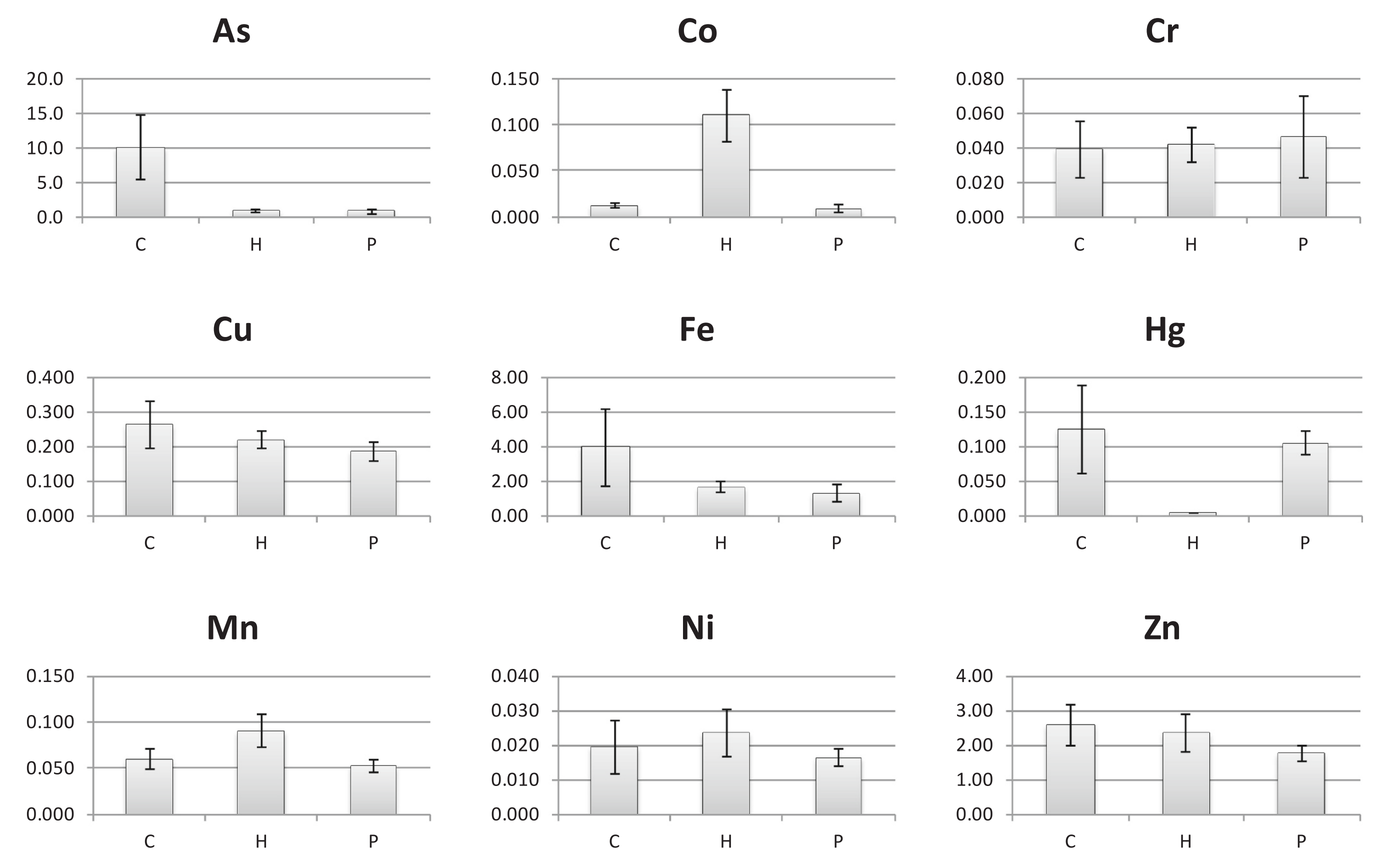

Fig. 3. Trophic variability of trace element concentrations (mean ± confidence interval expressed in μg/g of wet wt) measured in fish muscle (n = 141).

2012), age data was plotted against Hg and Mn concentrations measured in each specimen of *P. leopardus* collected. This showed a positive linear relation between age and concentrations of Hg accumulated providing confirmation that the contamination levels of this heavy metal increase with the age. Conversely, a negative correlation, between age and Mn was found. This relation, though much weaker than that for Hg, shows that the concentrations of Mn may tend to decrease in this specie with age (Fig. 9).

3.3. Risk assessment for human consumer

The Table 8 presents the results of risk assessment to humans from consumption of fish caught from the study zone. For the elements Cd, Cr, Cu, Fe, Ni and Zn, the risk of attaining the MSC thresholds is either inexistent for Cd (as the concentrations remained largely under the LD) or very low for Cr, Cu, Fe, Ni and Zn (around 200 to 6000 times less). Indeed, an adult of 70 kg will have to eat 33 to 488 kg of fish flesh on daily basis to attain the limits set for Cr, Cu, Fe, Ni and Zn whereas the daily consumption in New Caledonia is estimated to be inferior to 173 g of fish flesh per day (Leopold et al., 2004).

For Hg, this scenario could be more likely; especially in households where the weekly consumption of fish is higher. With the mean concentration measured (0.053 ± 0.010 μg/g wet wt), an adult consuming >5.3 kg of fish flesh per week will exceed the tolerable limits for Hg. If carnivorous (0.124 ± 0.097 μg/g wet wt) and piscivorous fish (0.105 ± 0.068 μg/g wet wt) are privileged in meals, this limit drops to 2.3 and 2.7 kg of fish flesh per week, respectively. This MSC threshold reduces furthermore if the sedentary piscivore *P. leopardus* from the CH is

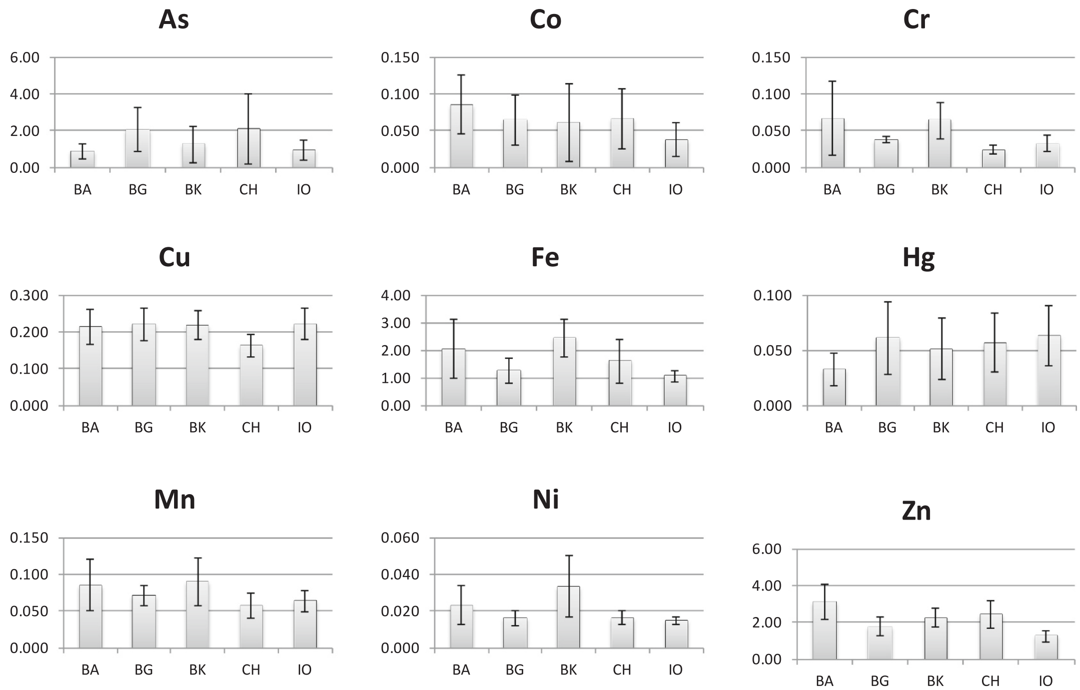

Fig. 4. Spatial variability of trace element concentrations (mean ± confidence interval expressed in μg/g of wet wt) measured in fish muscle (n = 141).

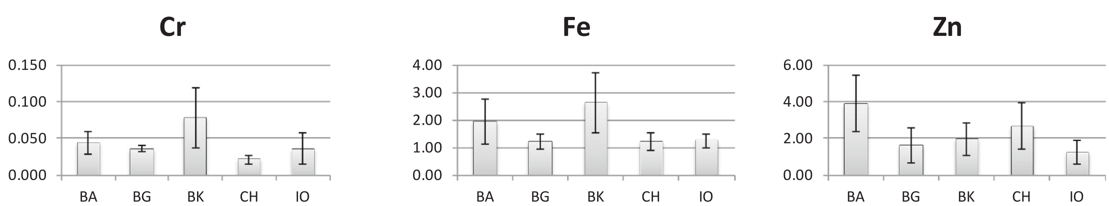

Fig. 5. Spatial variability of Cr, Fe and Zn concentrations (mean ± confidence interval expressed in μg/g of wet wt) measured in herbivorous fish muscle (n = 73).

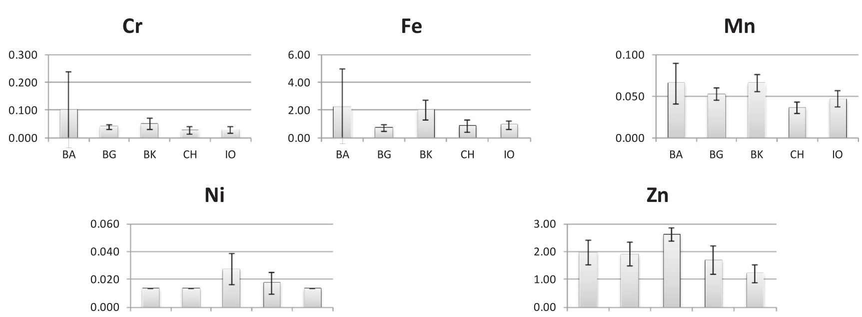

Fig. 6. Spatial variability of Cr, Fe, Mn, Ni and Zn concentrations (mean ± confidence interval expressed in μg/g of wet wt) measured in piscivorous fish muscle (n = 59).

Table 5
Mean concentrations ± confidence interval of trace elements in the *P. leopardus* muscle and lower and higher range values (n = 49).

Element (μg/g wet wt)	As	Cd	Co	Cr	Cu	Fe	Hg	Mn	Ni	Zn
Minimal value	<0.067	<0.003	<0.013	<0.007	0.084	<0.133	<0.003	<0.007	<0.013	<0.667
Maximal value	11.08	<0.003	0.106	0.726	0.453	14.5	0.384	0.175	0.052	3.23
Mean	0.796	<0.003	0.016	0.049	0.175	1.37	0.103	0.053	0.016	1.72
Confidence interval	0.435	–	0.004	0.029	0.028	0.58	0.019	0.007	0.003	0.24

privileged; with a mean Hg concentration of 0.139 ± 0.033 μg/g wet wt (n = 9), only 2.0 kg of flesh per week will lead the consumers to exceed the PTWI.

In the marine environment, Hg is mainly found under its organic forms, Me-Hg. With 80 to 90 % of Me-Hg coming from fish and shellfish intake, seafood consumption is one of the main sources of human

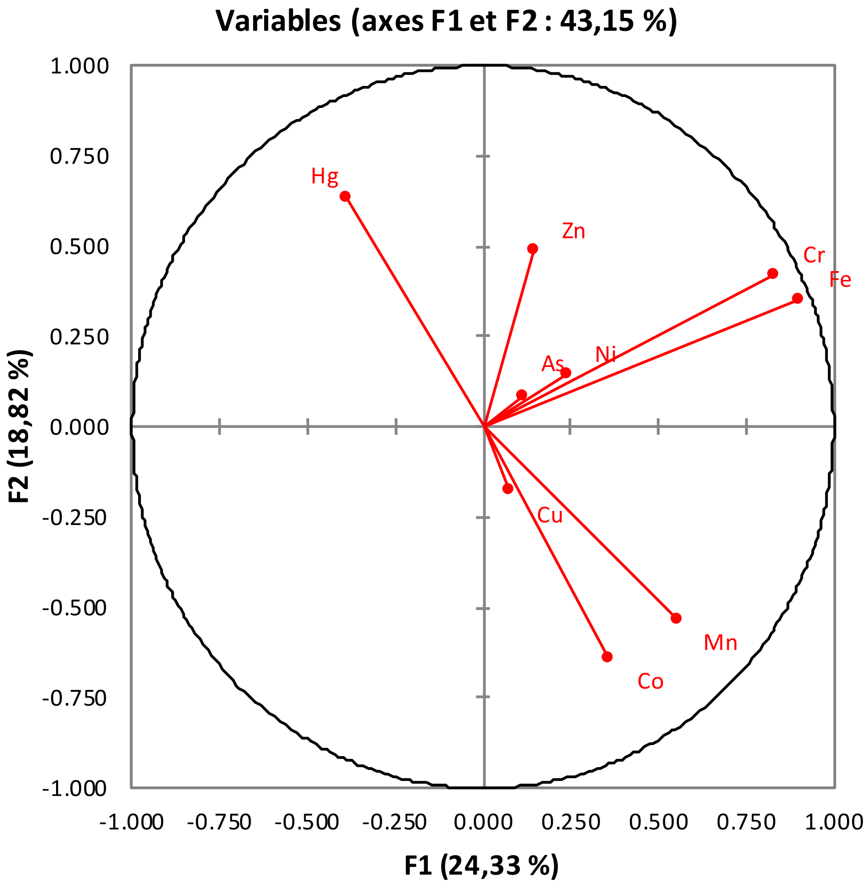

Fig. 7. Plot of the principal component analysis (PCA) showing global distribution of variables in all *P. leopardus* specie (n = 49).

Table 6
Pearson correlation matrix of trace element concentrations in the specie *P. leopardus* (n = 49).

Variables	As	Co	Cr	Cu	Fe	Hg	Mn	Ni	Zn
As	**1**								
Co	0.050	**1**							
Cr	−0.037	0.022	**1**						
Cu	−0.001	0.175	−0.076	**1**					
Fe	0.146	0.145	**0.949**	−0.006	**1**				
Hg	−0.013	−0.336	−0.056	0.044	−0.084	**1**			
Mn	−0.010	0.326	0.143	0.150	0.227	−0.429	**1**		
Ni	−0.029	−0.090	0.069	0.011	0.137	0.001	0.203	**1**	
Zn	0.140	−0.168	0.132	0.158	0.174	0.130	−0.052	0.171	**1**

In bold: r > 0.600.

exposure to Me-Hg (Hong et al., 2012). This organic form in top-level predatory fishes like the *P. leopardus* can contribute to 100 % of T-Hg content (Chouvelon et al., 2009). JEFCA has allocated a PTWI for Me-Hg of 1.6 μg/kg body wt/week. Based on this, an adult can exceed the MSC threshold with only 1.1 kg of edible flesh of *P. leopardus* per week. The risk becomes even greater with this specie if it is caught from CH (800 g per week). According to Leopold et al. (2004), the weekly consumption of fish is estimated to 1.2 kg of edible flesh.

4. Discussion

Despite the intense mining for Ni ores in New Caledonia, very few studies have been undertaken to assess contamination status of the comestible species and the human health issues related to this potential source of contamination. To the best of our knowledge, the current study is to date the widest survey performed (or at least reported) in New Caledonia in terms of number of edible marine fish species and number of contaminants analyzed. Thereby, this study aliments the scarce trace elements contamination dataset of New Caledonia initially constituted by campaigns of fish collect dating back to the year 2007 by Chouvelon et al. (2009) and Metian et al. (2013).

However, unlike these two previous studies whereby only 9 individuals were collected in the Southern lagoon, the 141 fish samples of the present work provide a better appreciation on the contamination status of this region as well as a larger database against which future comparison can be carried out. Furthermore, Chouvelon et al. (2009) and Metian et al. (2013) covered zones in urban and its commuting zones with the Grande Rade being the unique sampling site subjected to dual discharges from urban activities and from mining products from the nearby Ni ore smelter plant. Comparatively, the present study is focused

Table 7
Results of the Kruskal-Wallis tests analyzing the effect of sampling location and of fish size (as a proxy of fish age) of the specie *P. leopardus* on element concentrations (n = 49).

Variables	Spatial (sampling location) effect	Fish size effect
As	ns	*
Co	ns	ns
Cr	**	ns
Cu	ns	ns
Fe	*	ns
Hg	ns	***
Mn	***	***
Ni	ns	ns
Zn	***	ns

NS: not significant
* P < 0.05
** P < 0.01
*** P < 0.001

on the southern coastline exposed almost exclusively to mining activities. Given the exposed lateritic soils of the southern mainland, these waters are naturally influenced by terrigenous input of Co, Cr, Fe, Mn and Ni through numerous rivers and creeks. The numerous abandoned mines and active excavations in this area, notably that of PRNC presently operating in the Kwe drainage basin, has led to intensification of lateritic metals lixiviation and thereby, is of a potential public health concern to local tribes who live and fish in bordering zones.

One of the results of Metian et al. (2013) revealed that *P. leopardus* fish showed geographical variations with significantly higher concentrations of Ag (silver), Cd, Cu, Hg, Pb and Zn measured in the zones under heavy anthropogenic inputs from the surroundings of Noumea. On the other hand, no such difference was revealed for Ni mining ores associated elements (Co, Cr, Fe, Mn and Ni), neither in the muscle nor in the liver of *P. leopardus*. These observations and the physiological specificities of *P. leopardus* (resident species and an apex predator) led them to recommend the use of leopard coral grouper as a potential bioindicator species to evaluate the contamination status of its environment in urban setting but not that related to mining activities.

Our results with *P. leopardus* show significant spatial difference with Cr, Fe and Mn, and, as such, are in contradiction with those of Metian et al. (2013). Indeed, highest concentrations were measured in fish from BK and/or BA and lowest from CH. Regarding the anthropogenic metals, apart from Zn, no such difference was observed with Cd, Cu and Hg. The BK and BA zones, located closest to the PRNC mining activity, are directly subject to natural and anthropogenic terrigenous influences (mine front). As for the CH sampling area, it is located several kilometers from this industrial complex in a sector strongly influenced by ocean water masses. This is also the case for IO and BG. However, these two areas are mainly influenced by runoff water which leaches the superficial lateritic layers poor in Ni but rich in Mn, Fe, and occasionally in Cr (Fritsch et al., 2017), thus explaining the differential spatial response between Ni and its associated metals (Cr, Fe and Mn).

In this manner, our results show that *P. leopardus* can also be useful as a bioindicator to evaluate the contamination level of Cr, Fe and Mn in seawater; this hypothesis is furthermore supported by the coefficient of correlation obtained with Cr and Fe (r = 0,949) which suggests that the two metals originate from a same terrigenous source. Indeed, the soils of the southern New Caledonia are also known for their high chromite deposits (Savy et al., 2011).

The levels of bioaccumulation in marine organism have been shown to vary with environmental and biological factors such as age (size/weight), trophic level and lifestyle. These influences have been strongly revealed in the present study for elements Co, Fe and Mn with As and Hg providing the best examples. Indeed, the highest concentrations of As were measured in carnivorous fish that feed on invertebrates. Previous

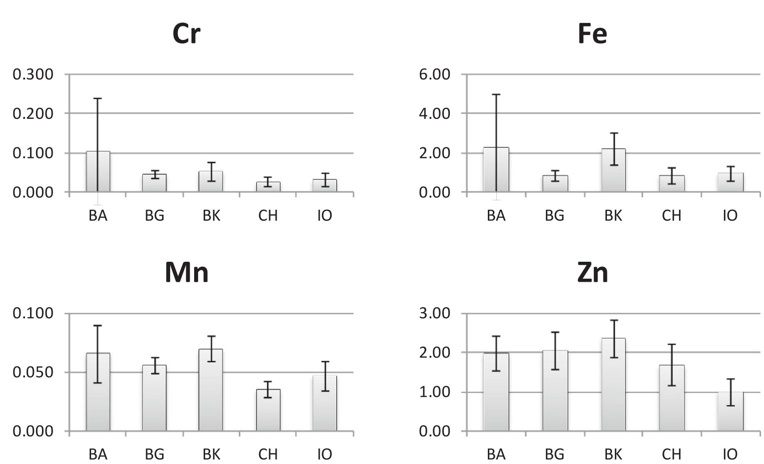

Fig. 8. Spatial variability of Cr, Fe, Mn, and Zn concentrations (mean ± confidence interval expressed in µg/g of wet wt) measured in *P. leopardus* muscle (n = 49).

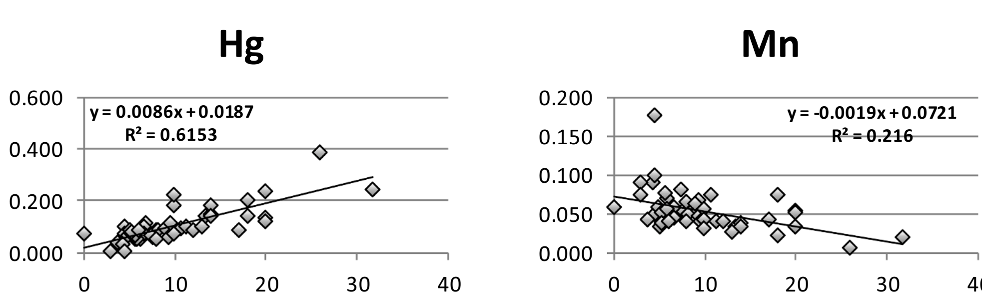

Fig. 9. Correlation between age (size as proxy) and concentrations of Hg and Mn (expressed in µg/g of wet wt) based on individual data in fish species *P. leopardus* (n = 49).

Table 8
Risk assessment for human consumers (70 kg) based on toxicological data established by JECFA and EFSA for Cd, Cr, Cu, Fe, Hg, Ni and Zn and the consumption data of fish in New Caledonia.

Sampling population	All species (n = 141)							P. leopard specie (n = 49)	
Elements	Cd	Cr	Cu	Fe	Hg	Ni	Zn	T-Hg	Me-Hg
Toxicological data established by JECFA[1] and EFSA[2]	25	300	500	800	4	13	1000	4	1.6
	μg/kg bw/m	μg/kg bw/d	μg/kg bw/d	μg/kg bw/d	μg/kg bw/w	μg/kg bw/d	μg/kg bw/d	μg/kg bw/w	μg/kg bw/w
	JECFA 2013 (PTMI)	EFSA 2014 (TDI)	JECFA 1982 (PMTDI)	JECFA 1983 (PMTDI)	JECFA 2011 (PTWI)	EFSA 2020 (TDI)	JECFA 1982 (PMTDI)	JECFA 2011 (PTWI)	JECFA 2007 (PTWI)
Mean concentration ± confidence interval measured in 141 fish muscle caught in the studied zone (μg/g wet wt)	0.003 ± 0.000	0.043 ± 0.011	0.207 ± 0.019	1.63 ± 0.31	0.053 ± 0.012	0.020 ± 0.004	2.12 ± 0.30	0.103 ± 0.019	0.103 ± 0.019
Maximum Safe Consumption (MSC) threshold of fresh fish flesh	583	488	169	34.4	5.30	45.5	33,0	2.70	1.10
	kg/m	kg/d	kg/d	kg/d	kg/w	kg/d	kg/d	kg/w	kg/w
Fresh fish consumption range based on average annual consumption of 28.0 ± 2.0[3] and 63 ± 9.7[4] kg/*per capita*	2.33–5.25	0.077–0.173	0.077–0.173	0.077–0.173	0.538–1.212	0.077–0.173	0.077–0.173	0.538–1.212	0.538–1.212
	kg /m	kg/d	kg/d	kg/d	kg/w	kg/d	kg/d	kg/w	kg/w

bw: body weight of consumer; d: day; w: week; m: month.
[1] https://apps.who.int/food-additives-contaminants-jecfa-database/.
[2] https://www.efsa.europa.eu/en/topics/topic/metals-contaminants-food.
[3] Labrosse et al. (2006).
[4] Leopold et al. (2004).

studies carried out in New Caledonia have shown the capacity of invertebrates to highly accumulate As in their tissues (Metian et al., 2008b; Hédouin et al., 2009). For Hg, the concentrations in this study were only detected in the carnivorous and piscivorous and not at all in herbivorous fish, which is in good agreement with the study of Chouvelon et al. (2009). Furthermore, our age-concentration studies in *P. leopardus* with the Hg confirm the positive correlation that was also reported by Chouvelon et al. (2009). In addition, our study also revealed that the concentrations of Mn may be negatively correlated with age. This negative correlation has previously been reported by Luczynska and Tonska (2006) in the muscles of freshwater piscivorous perches (*Perca fluviatilis*) and pike (*Exos lucius*) species.

Finally, though our toxicological study, very low risk on consumption of flesh of fish has been revealed for Cd, Cr, Cu, Fe, Ni and Zn. This cannot be said for Hg. While the risk with Hg remains checked when a variety of fish from different fishing zones are consumed, it becomes of concern, if certain species and fishing sites are privileged. Overall, 5.3 kg of fish flesh must be ingested by an adult of 70 kg on weekly basis in order to exceed the MSC threshold for Hg. However, if carnivorous and piscivorous fish are privileged in meals, the tolerable limit is attained with only 2.3 and 2.7 kg of fish flesh per week, respectively. And if the piscivore *P. leopardus* from the CH is privileged, then only 2 kg of flesh per week will lead a consumer to exceed the PTWI.

Based on the assumption that 100 % of T-Hg measured in piscivore *P. leopardus* in the present study is in its organic Me-Hg form (Chouvelon et al., 2009), an adult of 70 kg consuming just over 1 kg per week of edible flesh of this species will exceed the prescribed PTWI for Me-Hg. For those fished in the CH, only 800 g per week suffice to attain the MSC threshold. These results show that with a fish consumption rate varying between 28 and 63 kg/inhabitant/year, *i.e.* between 0.5 and 1 kg/week (Labrosse et al., 2006; Leopold et al., 2004), concerns can be raised for human groups for whom seafood constitutes the main resource of protein alimentation.

Top marine predators are known to contain higher concentrations of Hg in their tissues as a result of bioaccumulation up the food web (Martins et al., 2006). Therefore, through their feeding behavior, the higher levels of Hg measured in *P. leopardus* in this study was predictable. Chouvelon et al. (2009), based on their sampling population, found the highest concentration of Hg in the Grande Rade. Furthermore, they reported that eating only 260 g wet wt of edible flesh of *P. leopardus* caught from this site exposed consumers to Me-Hg intoxication. This situation is highly likely as the Grande Rade is one of the most contaminated areas in New Caledonia due to its proximity to the city of Noumea and to the Ni ore smelter plant that has operating since the early 1900's, powered by an on-site heavy fuel electric plant.

For the Southern lagoon, the highest concentrations of Hg were measured in the oceanic influenced area of CH. The source(s) of this input is/are unknown. The wastewaters that are pumped out *via* the diffuser into the CH, are mandatorily analyzed on daily basis by PRNC; these analyses have shown over time nondetectable levels of Hg within the effluent (non-published data). Urbanization and agricultural impacts are also low in this region. In the absence of reel anthropogenic input, we suspect that the geothermal activities of the study zone could probably be one of source of Hg.

Documentation on hydrothermal sources in New Caledonia is very limited, apart from a pinnacle called the Aiguille de Prony located not far from the mining complex of PRNC (Maurizot et al., 2020). Situated on the western side of the Baie de Prony, this natural carbonate formation with stalagmites raises up to 35 m above the seafloor to within 2 m of the surface. Further prospection in this bay using high-resolution bathymetric mapping has revealed the existence of numerous other shallow-water submarine structures like that of the Aiguille de Prony (Monnin et al., 2014). This area is now known as the Prony Hydrothermal Field (PHF). Existence of other vents in areas closer to the CH is also suspected. Though, no study has been undertaken to show if any of these vents naturally release Hg containing fluids into seawater, Hg emissions from the hydrothermal vents have been reported in the nearby region of the Tonga-Kermadec subduction zone (Lee et al., 2015; Zitoun et al., 2019).

Another source suspected is the volcanic activity in the region of the Vanuatu arc. The volcanic plumes are known to be rich in Hg content and the release of significant amounts of Hg from volcanoes Ambrym and Yasur of Vanuatu arc has recently been reported (Allard et al., 2016; Bagnato et al., 2011). A retro-assessment based on sulfur dioxide (SO2) emission data from 2005 to 2015, has seen the volcanos Ambrym et Yasur being placed as the top 20 potential emitters of Hg worldwide. These studies go on to hint at the potential of other volcanoes of the Vanuatu Arc (ex. Aoba) as important emitters (Edwards et al., 2021; Carn et al., 2017). Given the propensity for long-rang atmospheric transport of Hg (Jackson, 1997), it is worth noting that Yasur, situated in one of the southernmost islands of Vanuatu, Tanna, is the nearest neighbor to New Caledonia.

In the Pacific region, barely any information exists on the sources of Hg and Me-Hg whether it be natural or anthropic and even less on how it is transformed and transferred within the coastal ecosystem. Only scare results have been reported for the tuna across the Western and Central Pacific. Due to high commercial value of this oceanic pelagic fish to the fishing industry, large-scale efforts have been employed to model the Hg concentration patterns in three tuna species: bigeye, (*Thunnus obesus*); yellowfin, (*T. albacares*); and albacore, (*T. alalunga*) caught between 2001 and 2015. Houssard et al. (2019) has shown the existence of important geographical patterns of Hg concentrations independent of the size effect. For all three tuna species, individuals captured in the Equatorial region of the Pacific Ocean show depleted values, whereas relatively enriched values can be observed at lower latitudes than 15°S, in the southern part of the Archipelagic Deep Basins modified (ARCHm) area that includes the New Caledonian exclusive economic zone. It is worth noting that higher concentrations of Hg were measured in species foraging at depth (bigeye and albacore) than in species surface foraging predators (yellowfins). Among the drivers listed, the ARCHm region's island arc volcanism and hydrothermalism has been suggested as potential inputs of Hg by Houssard et al. (2019).

Other studies conducted at Mid-Atlantic Ridge hydrothermal vent fields have reported that the Hg levels in deep-sea fish are higher than "other deep-sea fish species with identical diets" caught elsewhere (Martins et al., 2006), thus supporting the hypothesis that hydrothermal vents can be an additional source for Hg for benthic fish species, like the *P. leopardus* in this study.

Furthermore, in a more recent study of Ruiz et al., 2022 carried out near the vents of the Bouillante Bay in Guadeloupe, the authors demonstrated that the marine organisms like the bivalves integrated dissolved inorganic hydrothermal Hg *via* diffusive pathway while opportunistic animals integrated this Hg by feeding on mats of sulfur-oxidizing bacteria that thrive under abundant sulfur supply and entrap elevated levels of Hg *via* dietary pathway. It is thus worth noting that in PHF vents, abundant and diverse presence of bacteria communities involving hydrogen, methane and sulfur compounds (*e.g.* sulfate) has been revealed (Quemeneur et al., 2014). It is therefore our hypothesis that the shallow vent fields occurring in the current study zone could be sources of Hg to the local ecosystem.

Finally, the magnitude of Hg measured in *P. leopardus* in this study in comparison to those analyzed in tuna deserves an attention. In CH, the grouper's size ranged from 50 to 60 cm and the Hg levels measured in muscle averaged to 0.139 ± 0.033 μg/g wet wt (n = 9). For the same size range, in tuna collected in ARCHm area, Hg averages range from 0.087 ± 0.044 (yellowfin; n = 12) to 0.329 ± 0.165 μg/g wet wt (bigeye; n = 16) (Houssard et al., 2019). This shows that the capacity of the leopard coral grouper as apex neritic predators evolving in the coastal waters to bioaccumulate equally high levels of Hg from its environment as does some tuna species in the pelagic context.

Like the tuna, the leopardus coral grouper has a strong commercial importance and is particularly abundant in New Caledonia (Preuss, 2012). Hence, the use of this fish as bioindicator species can proof to be valuable for monitoring the levels of several trace elements (Ag, Cd, Cu, Cr, Fe, Mn, Hg, Pb and Zn) in coastal waters. The commercial fishing and bioaccumulating capacity of this specie can provide combined optimal conditions for large-scale and continuous surveys allowing robust spatial and temporal studies in order to survey contamination levels of New Caledonian lagoons.

5. Conclusion

This study confirms that the bioaccumulation of different elements varies between population depending on their trophic, geographic, taxonomic and size characteristics, thus providing multifaceted results on contamination status of the Southern lagoon. Nonetheless, in herbivores and piscivores, a clear spatial accumulation pattern for Cr, Fe and Zn and to a lesser extent of Mn was observed, regardless of their sizes and species.

The bioaccumulator model *P. leopardus* showed higher concentrations of Cr, Fe and Mn in specimen caught in sites under impact of the excavation (BK) and/or port and refinery activities (BA) of PRNC. Of the non-lateritic elements, only the concentrations of Zn in *P. leopardus* muscle reflected a spatial distribution with lower concentrations in IO.

Regarding trophic group studies, As and Hg showed strong diet-related response with higher concentrations in carnivores (As and Hg) and piscivores (Hg); in *P. leopardus*, Hg showed a strong size-related accumulation with concentrations steadily increasing with aged individuals. The inverse was observed for Mn.

The risk assessments studies showed that there is a potential risk of contamination to mercury (Hg and Me-Hg) for humans consuming fish caught in the studied zone, especially for families who privilege fish as main source of protein. This risk becomes greater if piscivore species like *P. leopardus* are privileged in meals.

Previous studies in New Caledonia had demonstrated the ability of *P. leopardus* to differentially accumulate Ag, Cd, Cu, Hg and Zn and as such its potential as a bioindicator species to be used to assess the contamination status of urban area (Metian et al., 2013). Here, our results demonstrate that this species can also be used as a bioindicator tool to assess the state of Cr, Fe and Mn contamination related to mining activities in the New Caledonian lagoon. Hence, for the future monitoring programs for the PRNC, plans are now underway to use *P. leopardus* to survey the quality of the marine environment.

CRediT authorship contribution statement

Shilpa Kumar-Roine: Conceptualization, Data curation, Writing-Original draft preparation, Investigation. **Nicolas Guillemot**: Formal analysis, Visualization. **Pierre Labrosse**: Supervision.**: Jean-Michel Fernandez**: Writing- Reviewing and Editing, Validation. **Jean-Michel N'Guyen:** Validation, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The authors do not have permission to share data.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2022.114210.

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