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Mercury accumulation and transfer in hydrothermal coastal environment: the case of the geothermal plant of Bouillante

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Abstract

Geothermal vents can constitute local significant sources of mercury (Hg) in the environment. The geothermal power plant of Bouillante (Guadeloupe, Lesser Antilles) artificially enhances the release of hydrothermal water in shallow waters of the Bay. To assess the impact of this release on the Hg transfer in the environment, Hg concentrations were assessed in sediments, sulphur oxidising bacteria, and six animal species (urchin, sponges and fish) with various diets and trophic levels from the Bouillante Bay and a distant Control Site. Concentrations of Hg in all samples from Bouillante were greater than those from the Control Site (2 to 627 times higher). A comparison with the Hg concentrations reported in the literature for similar sample types reveals that they are abnormally high in most Bouillante samples suggesting a local Hg contamination imputable to the release of Hg hydrothermal water. Rocky pebbles of the shallow discharge channel are covered by a mat of sulphur oxidising bacteria presenting high concentration of Hg (13 $\mu\text{g}\cdot\text{g}^{-1}$). Through analysis of species with various diet, two pathways of Hg integration into Bouillante's food webs were emphasised. First, greater Hg concentrations

Abbreviations: C = Control sampling site; B = Bouillante sampling site; THg = total mercury; Hg_{inorg} = inorganic mercury; MeHg = methylmercury; CI = Confidence Interval; FDR = False Discovery Rate correction; PTWI = Provisional Tolerable Weekly Intake; MSWC = Maximum Safe Weekly Consumption

in filter-feeders (sponges X3 and bivalves X10) not consuming bacterial mats and liver/muscle ratios in fishes suggest that discharge waters enhance the exposure of marine organisms to dissolved inorganic Hg (*i.e.* waterborne pathway). Second, greater Hg concentrations in opportunistic animals such as the fish *Acanthurus bahianus* (X49) and the sea urchin *Diadema antillarum* (X50) known to ingest sulphur-bacterial mats from the discharge channel suggest that these chemosynthetic bacteria play a role in the integration (*i.e.* dietary pathway) and transfer of Hg in local biota, which enlightens the consideration of vent fields as natural sources of Hg.

Keywords

Total Mercury Concentrations – Bacterial Biofilms – Geothermal Power Plant – Shallow Hydrothermal Vents – Dietary Pathway

1. Introduction

Mercury (Hg) is one of the most bothering contaminants as it is highly toxic for all living organisms (Liu et al., 2012). The organic form of Hg is the methylmercury (MeHg) which is the most easily accumulated and retained form of Hg in organisms. MeHg raises major concerns since it biomagnifies along the food web and can reach dangerous levels in top predators including humans (Baeyens et al., 2003; Bosch et al., 2015).

The primary Hg emissions in the air decreased by 30% after the 1970s but then global anthropogenic releases to the atmosphere have remained stable since the 1980s and even increased slightly by $1.8\% \cdot \text{year}^{-1}$ between 2010 and 2015 (Streets et al., 2019). In addition, this decrease has not been passed on the Hg mass in the ocean, making stable the Hg levels in key marine species (Medieu et al., 2024). The main natural processes controlling Hg concentration in surface oceans is the atmospheric deposition uptake (Jiskra et al., 2021) and natural sources only account for a limited proportion ($\sim 17\%$) of all Hg released in surface oceans (Amos et al., 2013). However geothermal vents can constitute significant local sources through inorganic Hg degassing (Amos et al., 2013). As a result, they can shape the surrounding ecosystems at local scales by enhancing marine organisms' exposure to Hg (Martins et al., 2001; Martins et al., 2006) justifying the need of additional knowledge about Hg contamination associated with geothermal activity.

Very shallow vent fields are only found in 28 known locations worldwide, representing 4% of all known (*i.e.* active or inferred) active vent fields (Beaulieu & Szafranski, 2020). Among these sites, the west coast of Basse-Terre in Guadeloupe of Lesser Antilles constitutes an active volcanic area and presents several hydrothermal sources occurring at very shallow depths (Sanjuan & Bach, 1997), but also in streams pouring into the sea (Bagnato et al., 2009). This region is also characterized by high levels of natural Hg in sediments, and it was therefore hypothesised that shallow vent fields occurring there could be sources of Hg (Fabriol & Hazan,

1984; Morrison et al., 2015). Along this coast, since 1986, the “Bouillante” geothermal plant built close to the sea greatly enhanced the release of hydrothermal waters into the shallow Bouillante Bay (depth lower than 25 m). This site represents thus an opportunity to study a potential recent increase in Hg concentration in these peculiar coastal environments.

To assess the impact of enhanced releases of hydrothermal waters in the Bouillante Bay on Hg accumulation and transfer in coastal organisms, total Hg (THg) concentrations were measured in sediments, bacterial mats and different organisms of various trophic levels (sponges, bivalves, sea urchins and fishes). These data were compared with Hg concentrations in the same samples collected in a Control station located far from the hydrothermal vents of Bouillante.

2. Materials and Methods

2.1. Geothermal plant

The study area is located on the west coast of Guadeloupe, which belongs to the active Lesser Antilles volcanic arc (**Figure 1**). The multiple hot springs and fumaroles characterising this region explain the name of the town there: Bouillante, which means “boiling” in French. Similar to the well-characterised Champagne Hot Springs of Dominica (Kleint et al., 2017), Bouillante Bay and the surrounding coastline are known for their numerous very shallow springs that produce numerous strings of gas bubbles and hydrothermal fluid (Sanjuan & Brach, 1997).

To exploit this green source of energy, one of the two France’s geothermal plants was put into service in 1986 in Bouillante, and now generates 6-7% (100-110 GWh per year) of the whole Guadeloupe Island electricity consumption (Demarcq et al., 2014). High-pressure steam is exploited in two turbines (Unit 1 & 2) and to maintain the pressure in the aquifer, around

10% of the liquid water is reinjected while the rest is cooled down with pumped seawater ($\sim 8380 \text{ m}^3 \cdot \text{h}^{-1}$), and then evacuated into the sea at a temperature of 40-45°C by a discharge channel ($\sim 9000 \text{ m}^3 \cdot \text{h}^{-1}$; Géothermie Bouillante, 2018) (Fig. 1).

2.2. Sampling sites and method

The Bouillante sampling site (GPS: 16.127964, -61.770353) is located near the discharge channel (<1m depth) characterised by a strong current ($2.5 \text{ m}^3 \cdot \text{s}^{-1}$). At the mouth of the channel a $\sim 45^\circ$ pebble slope leads to a sedimentary bottom at a depth of 2m. The hot (40-45°C) discharge water flowing between the surface and the depth of the channel creates a strong thermocline. Geothermal water released in the sea is composed by high amounts of sulphur compounds (Fabriol & Ouzounian, 1985), allowing the growth of chemosynthetic bacteria using sulphur compounds (*e.g.* S^0 , H_2S and $\text{S}_2\text{O}_3^{2-}$) to create organic molecules in aerobic conditions thanks to their sulphur-oxidising activity (Takai et al., 2004). Sulphur-bacteria create a thick (6-7mm) white bacterial mat identified as *Plectonema sp.*, covered with white filamentous bacteria of the genus *Thiomicrospira* (Pascal et al., 2017). This white bacterial mat covering pebbles is limited in the discharge channel and fragments are continuously ripped off by the current and spread into the bay (**Figure 1**).

The Control sampling site is located on the southern side of the creek “Anse à la Barque” close to “Pointe Diburque” (GPS: 16.087326, -61.768897), 4.5km away from Bouillante. This site contains many diffuse hot springs scattered in very shallow water and receives occasionally hydrothermal water from the multiple hot springs around the intermittent stream named “Ravine Renoir” (Sanjuan & Bach, 1997; Figure 1).

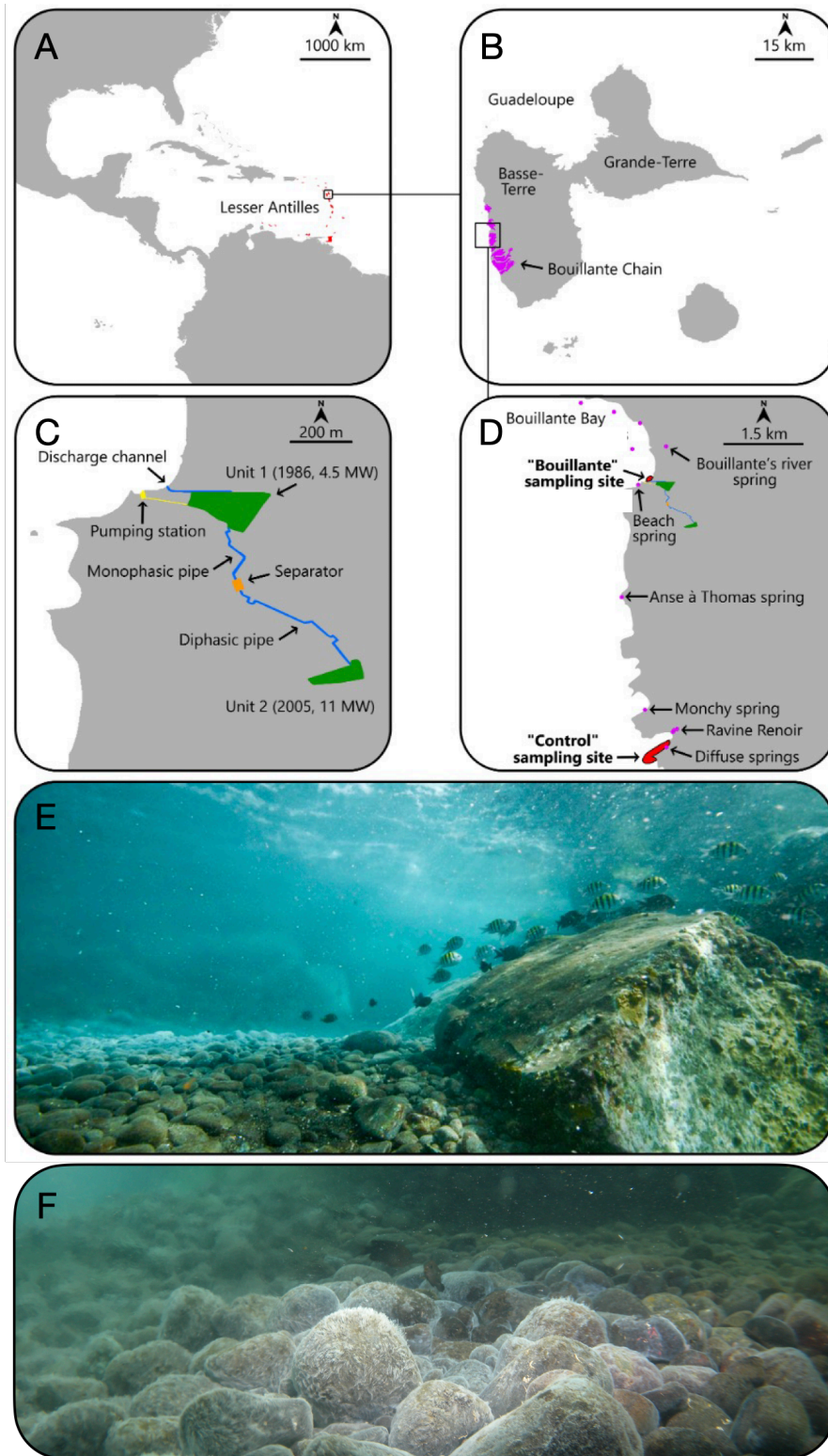


Figure 1: Description of sampling sites – Map of the location of two sampling sites and geothermal plant (A, B, C, D). The Bouillante Chain in purple on the upper right panel was represented according to Gadalia et al. (2019). The locations of all known springs pouring geothermal fluid in seawater represented in purple on the lower right panel were retrieved from Sanjuan & Bach (1997). Picture of the outlet of the discharge channel (E) presenting a bottom composed by rocky pebbles covered by bacterial white (F) producing many clearly visible fragments ripped off by the strong current, which are eaten by two visible fish species *A. saxatilis* and *A. bahianus* (E).

Previous studies realised in leeward coast of Guadeloupe in nearby Pigeon Islets (4.8 kms from Bouillante Bay) described benthic invertebrates (Malahel et al, 2022) and herbivorous fish communities (Koop et al., 2012). In Bouillante Bay, most abundant fish species Bouillante Bay associated with discharge channel are *A. saxatilis* and *A. bahianus* (Pascal et al. 2017).

Sampling took place on 07/14/17 and 07/18/17 and was carried out freediving. The white bacterial biofilm was collected through pebbles scraping. One sediment sample from Bouillante and three sediment samples from the control site were collected in the first centimetres of the substrate at ~2m depth. Sponges (*Aplysina fistularis* and *Iotrochota birotulata*), as well as sea urchins (*Diadema antillarum*) and bivalves (*Spondylus tenuis*) were collected by hand. Fishes (*Acanthurus bahianus* and *Abudefduf saxatilis*) were speared in the head, avoiding damaging the visceral mass and muscles. Ten specimens of each species were taken from each site.

2.3. Mercury concentration

Fishes and sea urchins were measured (fishes: total length, urchins: test size) and weighed (wet weight). The whole branches were examined for sponges. Ten samples of muscle were extracted for all other species, as well as ten liver samples for fish species and ten gonad samples for sea urchins. Muscles extracted were protractor, retractor and compass elevator muscles for *D. antillarum*, adductor muscles for *S. tenuis* and fillets for fishes. The 183 solid samples were then freeze-dried. Therefore, all concentrations of solid materials are based on dry weights.

Dry samples were reduced to a fine powder using a mortar and a pestle in order to homogenize them. The total Hg concentrations (THg) were measured (using Sartorius Cubis™ II MicroBalance) on a subsample of 0.05–50 mg dry powder (depending on the Hg concentrations) using an Advanced Mercury Analyser spectrophotometer (Altec AMA 254)

able to detect 0.005 mg, as described in Chouvelon et al. (2009). Briefly, the samples were combusted under oxygen and the liberated Hg was analysed by atomic absorption spectrophotometry (Roos-Barracough et al., 2002). For each sample, analyses were repeated at least two times, until the relative standard deviation between measures was <10%. Subsequently, the mean of the repeated Hg measurements was used for statistical analysis. To ensure the accuracy of measurements, a certified reference material, NRCC DOLT-5 (dogfish liver = $0.44 \pm 0.18 \mu\text{g}\cdot\text{g}^{-1}$), was analysed every 15 samples for recalibration of the analyser. Results indicated an average Hg concentration of $0.43 \pm 0.01 \mu\text{g}\cdot\text{g}^{-1}$ dw, in good agreement with certified values.

2.4. Data analyses

All statistical analyses were performed using R version 4.0.2. Since samples were not following a gaussian distribution, mean confidence intervals were calculated using 1 million iterations Bootstrap with the BCA (Biased-Corrected & Accelerated) method because it is considered the most conservative (Chou et al., 2006). The confidence intervals around mean ratios (*i.e.* Bouillante / Control) were computed with the same method, using a modified version of the function `ci_mean_diff()` from the *confintr* package to adapt it to ratios.

According to the validation or not of the conditions of use, means were compared using Student, Student with Welch correction or Wilcoxon tests. Differences between means with confidence interval and standardised effect size (*i.e.* Cohen's d and Wilcoxon r) were calculated in case of significant p-values. Pairwise Wilcoxon tests were used to test for differences between Hg concentration in tissues for each species and among species. The Benjamini-Hochberg adjustment (1995) method was chosen to control the False Discovery Rate (FDR).

2.5. Risk assessment for Human consumers

A Maximum Safe Weekly Consumption (MSWC) was evaluated only for the tissues which are usually consumed by the local population. Thus, only *S. tenuis* muscle, *A. bahianus* muscle and *D. antillarum* gonads (Gobert & Reynal, 2002, Grisolia et al., 2012) were considered, since sponges, as well as livers, are not edible, and *A. saxatilis* is never used as food by local consumers due to its bad taste (Silvano & Begossi, 2012).

The MSWC was calculated on the basis of the Provisional Tolerable Weekly Intake (PTWI) given by the Joint Expert Committee on Food Additives (JECFA). The PTWI is 4 $\mu\text{g.kg}^{-1}$ bw for inorganic mercury (Feeley et al., 2011) and 1.6 $\mu\text{g.kg}^{-1}$ bw for methylmercury (Barlow et al., 2007). The average body weight (BW_{ind}) in the Caribbean is 67.9 kg (Walpole et al., 2012) and was used in the following formula:

$$MSWC_{(g.week^{-1})} = \frac{BW_{\text{ind}} (kg) \times PTWI (\mu\text{g.kg BW}^{-1}.week^{-1})}{[Hg] (\mu\text{g.g}^{-1} ww)}$$

The wet weight (ww) Hg concentrations were converted to dry weight (dw) based concentrations using this formula:

$$[Hg]_{ww} = [Hg]_{dw} \times \frac{100 - \text{Moisture content } (\%)}{100}$$

The conventional moisture content of 75% was used because it fitted well with the values measured for close species, ranging from 73% to 77% (e.g. Elliott et al., 2015; Keshavarz & Jahromi, 2017; Szkoda et al., 2015).

3. Results

Among all 191 samples retrieved from the field, the Hg concentration could be successfully assessed for 189 of them. Indeed, the small livers of one *A. bahianus* (10 cm long) and one *A. saxatilis* individuals did not allow to reach the Hg threshold limit of the AMA. Otherwise, the mean RSD for replicates in each sample was kept under 4%, which indicates a good accuracy of measurement.

3.1. Differences of Hg concentration among organisms

The pairwise comparisons (Wilcoxon's tests with FDR correction) between the Hg concentrations measured in the samples from the Control Sites delineate 5 statistical groups, from the highest to the lowest total THg concentrations as: *A. bahianus* liver > *D. antillarum* gonad ~ *A. fistularis* branch > *I. birotulata* branch > *A. saxatilis* liver ~ *D. antillarum* muscle ~ *S. tenuis* muscle ~ *A. saxatilis* muscle > *A. bahianus* muscle (**Figure 2**). The mean concentrations ranged from 0.013 $\mu\text{g}\cdot\text{g}^{-1}$ (95% CI [0.010, 0.024]) for *A. bahianus* muscle to 1.879 $\mu\text{g}\cdot\text{g}^{-1}$ (95% CI [0.866, 3.885]) for *A. bahianus* liver (*i.e.* 145-fold higher). Excepted for sponges, *A. bahianus* livers and *D. antillarum* livers, THg concentrations measured in all samples were similar or inferior to the sediment concentrations of the Control Site. The concentration in the *A. bahianus* liver was 78 times higher than this in the sediment, although there was large variability among measurements for this sample type.

The THg concentrations between samples from Bouillante ranged from 0.176 $\mu\text{g}\cdot\text{g}^{-1}$ (95% CI [0.163, 0.188]) in the branches of *I. birotulata* to 77.22 $\mu\text{g}\cdot\text{g}^{-1}$ (95% CI [45.70, 109.9]) in the liver of *A. bahianus* (*i.e.* 438-fold higher) (**Figure 2**). The bacterial mats found in the Bouillante site showed a THg of 13.01 $\mu\text{g}\cdot\text{g}^{-1}$. The muscles of *S. tenuis*, *A. saxatilis* and *A. bahianus*, as well as the branches of *I. birotulata*, contained similar Hg concentrations to those found in the surface sediments of Bouillante. Mean THg concentrations in *A. bahianus* and *A.*

saxatilis liver were 5.94 times and 4.10 times higher, respectively, than those measured in the bacterial mat. The concentrations values for the other samples were comprised between those found in the sediments and in the bacterial mat (**Table S1**).

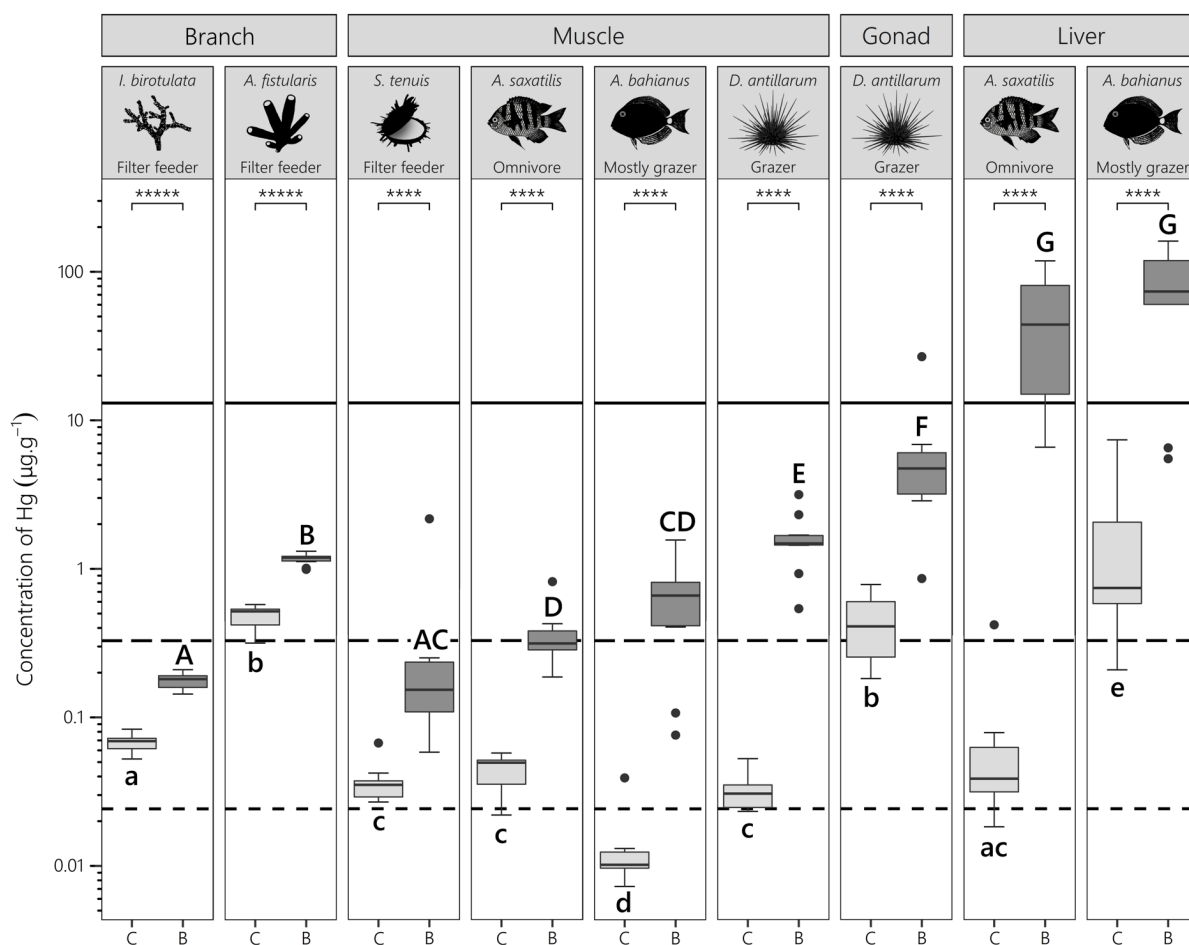


Figure 2: Comparisons of Hg concentrations – Hg concentrations ($\mu\text{g}\cdot\text{g}^{-1}$; dry weight) in marine organisms from the Control site (C, light grey) and Bouillante (B, dark grey) sites. The solid horizontal line represents the Hg concentration in the bacteria meadow ($[\text{Hg}] = 13.01 \mu\text{g}\cdot\text{g}^{-1}$). The dashed lines represent the Hg concentration in surface sediments (long-dash = Bouillante: $[\text{Hg}] = 0.326 \mu\text{g}\cdot\text{g}^{-1}$; short-dash = Control Site: $[\text{Hg}] = 0.024 \mu\text{g}\cdot\text{g}^{-1}$). The asterisks indicate a statistically significant difference (Student or Wilcoxon tests; $p\text{-value} < 0.0001 = ****$ & $p\text{-value} < 0.00001 = *****$) between the two sampling sites. Letters regroup non-statistically different groups (Pairwise Wilcoxon's Tests with FDR correction), the lower cases being used for the comparisons between Control samples and the upper cases for the comparisons between Bouillante samples. Concentrations were calculated based on dry weight.

3.2. Differences in Hg concentrations between sites

The THg concentrations were drastically higher at Bouillante than at the Control Site, whatever the sample type. All statistical tests (*i.e.* Student or Wilcoxon tests) were significant and standardised effect sizes (*i.e.* Cohen's D or Wilcoxon's R) indicated a large magnitude of effect (**Table S2**). Indeed, the predicted differences between both sites (Bouillante – Control) by Student or Wilcoxon tests were always positive, ranging from + 0.11 $\mu\text{g}\cdot\text{g}^{-1}$ (95% CI [0.09, 0.13]) for *Iotrochota birotulata* branches, to + 70.9 $\mu\text{g}\cdot\text{g}^{-1}$ (95% CI [52.6, 118]) for *Acanthurus bahianus* liver (**Table S2**).

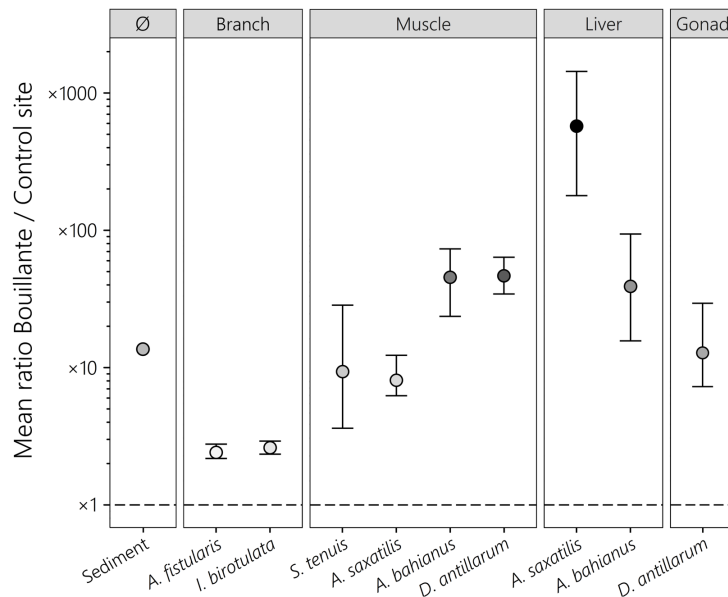


Figure 3: Ratios of increase in Hg concentrations in Bouillante – Mean ratio between the Hg concentration of Bouillante and the Control Site, enabling to evaluate the effect of the geothermal plant independently of initial differences between types, species and tissues. The vertical bars represent the 95% bootstrapped confidence interval around the mean ratio (iterations = 1,000,000). Ratios were established using concentrations in $\mu\text{g}\cdot\text{g}^{-1}$ (dry weight). The different shades of grey are proportional to the mean ratio. The dashed line corresponds to equal concentrations in Bouillante and the Control Site. Concentrations were calculated based on dry weight.

This site difference was expressed as the ratio between concentrations in Bouillante Bay and those found in Control Site (**Figure 3**) allowing to assess the magnitude of the

contamination increase. The THg concentrations in surficial sediment at 2 m depth was much higher close to the geothermal plant discharge channel, with concentrations 13.6-fold higher in Bouillante than in the Control site. Both *I. birotulata* and *A. fistularis* displayed the lowest Hg ratio with sponge branch with values of 2.64 (95% CI [2.37, 2.96]) and 2.44 (95% CI [2.20, 2.81]), respectively (**Table S1**). Interestingly, muscle samples, *A. bahianus* ($\times 48.51$, 95% CI [24.96, 78.77]) and *D. antillarum* ($\times 49.75$, 95% CI [36.50, 68.34]) displayed the highest increase of contamination in Bouillante, compared to *S. tenuis* ($\times 9.71$, 95% CI [3.69, 30.02]) and *A. saxatilis* ($\times 8.38$, 95% CI [6.43, 12.78]). Contrastingly, *A. saxatilis* liver ($\times 626.82$, 95% CI [191.24, 1589.50]) showed the highest magnitude of increase compared to *A. bahianus* one ($\times 41.09$, 95% CI [16.22, 99.77]), suggesting that both species were not equally exposed to the Hg contamination of Bouillante.

3.4. Risk assessment for human consumers

Although the Hg speciation in the respective tissues remained unknown in this study, the recorded Hg concentrations in those species might be of concern for seafood consumers. We unrealistically assume that 100% of THg measured in edible tissue was either inorganic Hg (Hg_{inorg}) or MeHg, to calculate the range of the the Maximum Safe Weekly Consumption. These values were thus calculated considering both Provisional Tolerable Weekly Intake (PTWI) for Hg_{inorg} and MeHg. Considering THg as inorganic, the MSWC ranged from 2.43 kg.week⁻¹ (95% CI [1.85, 3.35]) for *D. antillarum* gonads to 83.6 kg.week⁻¹ (95% CI [45.3, 108.7]) for *A. bahianus* muscle, when sampled at the Control Site (**Figure 4**). Consuming seafood from Bouillante, the MSWC dropped and ranged from 167 g.week⁻¹ (95% CI [77.9, 282.9]) of *D. antillarum* gonad to 3.1 kg.week⁻¹ (95% CI [0.95, 7.8]) of *S. tenuis* muscle.

Considering that THg was found under MeHg, all MSWC values were approximately divided by 2.5 (**Table S3**).

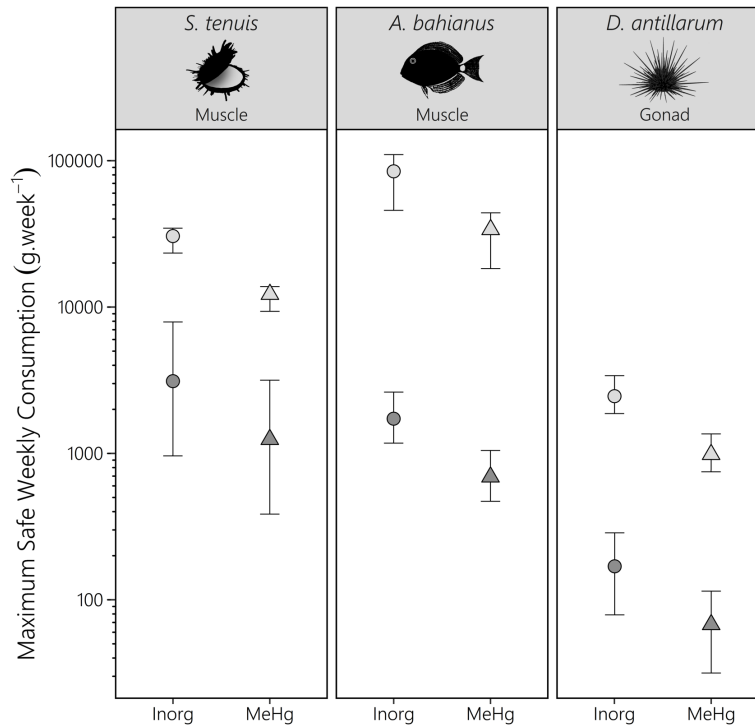


Figure 4: Risk assessment for Human consumers – Maximum Safe Weekly Consumption (MSWC) quantities according to the FAO guidelines for the three species potentially eaten by humans. The different colours represent the different sites (light grey = Control Site, dark grey = Bouillante) while the different shapes represent the different hypotheses concerning the type of mercury bioavailable in the Bouillante Bay (dot = inorganic Hg; triangle = organic Hg). The vertical bars represent the values obtained after calculation of the MSCW on the limits of the 95% bootstrapped confidence interval of the Hg concentration (iterations = 1,000,000).

4. Discussion

Forty years ago, prospection studies preceding the construction of the Bouillante geothermal plant in 1986 revealed that THg in shallow surface sediments near the future discharge channel ($0.065 \mu\text{g}\cdot\text{g}^{-1}$) was already slightly higher in Bouillante than in the Control Site ($0.040 \mu\text{g}\cdot\text{g}^{-1}$; Fabriol & Hazan, 1984). Although being not clearly explained, this could be due to scattered shallow bubbling springs in the sediment bed more abundant near the Bouillante sampling site than in the Control sampling site (P-Y. Pascal pers. com.).

Table 1: Comparative literature analysis – Comparison between Hg concentrations measured in this study and Hg concentrations measured in other sites worldwide differing in Hg contamination levels. Only species with close biology, ecology and trophic levels were compared. The range (min – max) of Hg concentrations between sample types in other articles is reported in the last two columns instead of the mean and their 95% robust CI in the first two columns, since not enough data was available for calculations of these statistics in most cases. All concentrations are based on dry weight and reported in $\mu\text{g}\cdot\text{g}^{-1}$. Concentrations given in wet weight in other articles were converted in dry-weight based concentrations using a conventional 75% moisture content if not given.

Species / Type	Tissue	Control	Bouillante	Probable Hg-poor sites	Hg-rich sites
Sediments	Ø	0.024 [0.023, 0.025]	0.326	0.001 - 0.041 ^a	0.005 - 0.145 ^B 0.035 - 4.802 ^C 130 - 146 ^D
<i>Iotrochota</i> <i>birotulata</i>	Ø	0.066 [0.060, 0.072]	0.176 [0.163, 0.188]	15 sponge species: 0 - 0.6 ^b	6 sponge species: 0 - 1.9 ^E
<i>Aplysina</i> <i>fistularis</i>	Ø	0.477 [0.416, 0.519]	1.164 [1.094, 1.220]	4 sponge species: 0.005 - 0.051 ^c	5 sponge species: 0.008 - 0.087 ^G
<i>Diadema</i> <i>antillarum</i>	Gonad	0.447 [0.324, 0.587]	6.498 [3.84, 13.93]	<i>Stomopneustes variolaris</i> : 0 - 0.752 ^f <i>Paracentrotus lividus</i> : 0 - 0.11 ^g	<i>Paracentrotus lividus</i> : 0.01 - 0.49 ^I
<i>Diadema</i> <i>antillarum</i>	Muscle	0.032 [0.028, 0.039]	1.601 [1.253, 2.108]	Not available	Not available
<i>Spondylus</i> <i>tenuis</i>	Muscle	0.036 [0.032, 0.047]	0.353 [0.139, 1.143]	2 Pectinidae species: 0.03 - 0.15 ^d	<i>S. spinosus</i> : 0 - 0.08 ^F 3 Ostreidae species: 0.109 - 0.211 ^B
<i>Abudefduf</i> <i>saxatilis</i>	Muscle	0.043 [0.034, 0.049]	0.361 [0.293, 0.528]	<i>A. septemfasciatus</i> : 0.044 - 0.056 ^e <i>A. sordidus</i> : 0.112 ^e	<i>A. saxatilis</i> : 0.229 ^B <i>A. sexfasciatus</i> : 0.361 - 0.369 ^C
<i>Acanthurus</i> <i>bahianus</i>	Muscle	0.013 [0.010, 0.024]	0.637 [0.420, 0.935]	<i>A. lineatus</i> : 0.002 - 0.054 ^a <i>A. triostegus</i> : 0.020 - 0.056 ^e <i>A. xanthopterus</i> : 0.014 - 0.037 ^c <i>A. dussumieri</i> : 0.002 - 0.016 ^h	<i>A. coeruleus</i> : 0.120 - 0.163 ^B <i>A. chirurgus</i> : 0.182 - 0.385 ^B <i>A. xanthopterus</i> : 0.017 - 0.165 ^F
<i>Abudefduf</i> <i>saxatilis</i>	Liver	0.084 [0.038, 0.221]	52.88 [30.60, 79.89]	<i>Caesio xanthonotus</i> : 0.031 - 0.591 ^h	Not available
<i>Acanthurus</i> <i>bahianus</i>	Liver	1.879 [0.866, 3.885]	77.22 [45.70, 109.9]	<i>A. xanthopterus</i> : 0.053 - 0.111 ^c <i>A. dussumieri</i> : 0.159 - 1.630 ^h	<i>A. xanthopterus</i> : 0.123 - 1.060 ^F

	a: Tutuila, one of the Samoan Islands (Morrison et al., 2015)
	b: Antarctica (Bargali et al., 1998); Sydney coastal lakes (de Mestre et al., 2012); Red Sea coasts (Pan et al., 2011)
Probable	c: Control site without Hg-rich sediments (Denton et al., 1997) in Guam harbours (data: Denton et al., 2006)
Hg-poor	d: Multiples sites in the Marlborough Sound (New Zealand), a marine reserve (Schlieman, 2020)
sites	e: Cocos Lagoon, near a marine reserve, Guam (Hartwell et al., 2017)
	f: Three different reefs on the south and the south-west coast of Sri Lanka (De Zoysa et al., 2018)
	g: Two different seagrass beds on the French Mediterranean coast (Calvi & Marseille; Warnau et al., 1995)
	h: Multiples sites along the Great Barrier Reef (Denton & Burdon-Jones, 1986)

	A: Six active volcanic areas around the world, including freshwater sources and waterways around the Soufrière volcano in Guadeloupe, with the highest Hg concentrations in the study (Bagnato et al., 2009)
	B: Florida, contaminated with runoff waters charged in HgCl ₂ and MeHg (Frederick et al., 2005). Data from Florida Bay for water and sediments (Rumbold et al., 2011), and from Keys Archipelago for animals (Rumbold et al., 2018).
	C: Kayeli Bay in Indonesia, polluted by an artisanal gold mine using Hg since 2011 (Manullang et al., 2019)
Hg-rich	D: Minamata Bay, contaminated with HgO from a factory (Balogh et al., 2015), killing thousands of people (data: Fujiki & Tajima, 1992)
sites	E: Polluted sites near Marseille (Perez et al., 2004); Polluted site near Rio de Janeiro (Batista et al., 2014)
	F: Sites with Hg-rich sediments (Denton et al., 1997) in Guam harbours (data: Denton et al., 2006)
	G: Polluted sites near large Lebanese towns (Micheline et al., 2019)
	H: Porewaters from Paleochori Bay (Greece), containing more Hg than surrounding pore fluids (Roberts et al., 2021)
	I: Ischia Island (Italy) with many bubbling springs and high levels of Hg (Bagnato et al., 2009; Warnau et al., 1995)

Nowadays, the concentrations measured in shallow surface sediments on the Control Site (*i.e.* 0.024 µg.g⁻¹, 95% CI [0.023, 0.025]) were ~1.7 times lower than the values reported in 1984 (Fabriol & Hazan, 1984), being similar to those reported in other probable Hg-poor sites (*i.e.* 0.001-0.041 µg.g⁻¹; Morrison et al., 2015; **Table 1**). Hg concentrations in sediment from Bouillante became contrastingly 5 times higher than 4 decades ago (0.326 µg.g⁻¹ now), suggesting an enhanced release of Hg-rich hydrothermal waters through the discharge channel of the geothermal plant. This is congruent with reported abnormally high concentrations of Hg hydrothermal waters of the Bouillante chain's hot springs compared to other active volcanic areas (*i.e.* 10 – 376 ng.L⁻¹; Bagnato et al., 2009).

Concentrations of Hg in organisms sampled in the Control Site seem consistent with previous values reported for the same species, or close species in terms of biology, ecology and trophic level, in other study zones with probably low levels of Hg (**Table 1**). However, in some cases, they were closer to those reported in Hg enriched sites: for example, gonads of *D. antillarum* contain similar concentrations to those of *Paracentrotus lividus* in Ischia Island (Italy), indicating similarities between both sites (Warnau et al., 1995; Bagnato et al., 2009).

Regarding sponges, Hg concentrations recorded in *I. birotulata* and *A. fistularis* were in the range of values reported in sponges from non-polluted areas (**Table 1**). Interestingly, Hg concentrations in *A. fistularis* were on average 7 times higher than in *I. birotulata*. This difference could be due to their different shapes (Lacoue-Labarthe et al., 2016), their differences of microbial abundance (Pawlik et al., 2015), their differences in microbial communities (*e.g.* symbiotic *Synechococcus* of *A. fistularis* can act on HgCl₂; Erwin & Thacker, 2007; Lefebvre et al., 2007) and filtration rate (Weisz, et al., 2008).

In fish, both liver and muscle exhibit specific concentrations, as a consequence of the accumulation, organotropism, detoxification and elimination mechanisms, driven by the metal speciation, the exposure pathways (waterborne *vs.* diet), food regime and proportion of Hg species in food items (Maury-Brachet et al., 2006; Wang & Wong, 2003). Interestingly, *A. bahianus* showed the lowest and the highest Hg concentrations in the muscle and the liver, respectively, likely because its benthivorous/periphytophagous diet favours the ingestion of inorganic forms of Hg (48% to 72% of Hg in their food) that preferentially accumulates in the liver and kidneys (Maury-Brachet et al., 2006). On the contrary, the planktivorous *A. saxatilis*, which has a higher trophic level ($\delta^{15}\text{N} = 5.70 \pm 0.31\text{‰}$) than *A. bahianus* ($\delta^{15}\text{N} = 4.89 \pm 0.10\text{‰}$; Pascal et al., 2017), likely contains more MeHg, since the concentrations in its muscle and liver are similar (Scheuhammer et al., 2015). The comparison with other probable Hg-poor sites reveals similar tendencies for fishes' liver and muscle (**Table 1**) indicating a classical

accumulation of Hg in the tissues of these fishes, with differences between species probably explained by their different diets.

Present study reveals a large and significant increase of the Hg content in Bouillante organisms whatever the sample (**Figure 3 & Table S2**). Assuming that the studied species reflect the local contamination as their ranges of movement are considered to be less than 400 m (Chapman & Kramer, 2000; Pascal et al., 2017), these higher levels demonstrate a local Hg contamination at this site imputable to the release of Hg enriched discharge hydrothermal water by the Bouillante power plant, as evidenced by the increase in sediment Hg content since 1984. Moreover, these values were generally much higher than those reported in Hg-poor sites (**Table 1**), implying a high amplitude of Hg contamination in Bouillante. For example, Hg in livers of *A. bahianus* is ~80-fold higher than in livers of *Acanthurus xanthopterus* from known Hg-enriched harbours in Guam (Denton et al., 1997; Denton et al., 2006).

Previous isotopic signature analyses (*i.e.* $\delta^{15}\text{N}$, $\delta^{13}\text{C}$ & $\delta^{34}\text{S}$) showed that fish (*A. bahianus* & *A. saxatilis*) and sea urchins (*D. antillarum*) actively seek out (*i.e.* greater abundance) and consume the bacterial mats growing specifically in the channel of Bouillante hydrothermal plant whereas filter feeders (*I. birotulata*, *A. fistularis* & *S. tenuis*) do not ingest those bacteria (**Figure 5**; Pascal et al., 2017). Such results raise the question of a possible role of bacteria in the integration and transfer of Hg for those opportunistic species.

The dense white bacterial mat covering the rocky pebbles of the shallow discharge channel is made of filamentous *Plectonema sp.* cyanobacteria (Oscillatoriaceae) associated with the sulphur-oxidising *Thiomicrospira sp.* gammaproteobacteria (Pascal et al., 2017). Associations between Oscillatoriaceae and sulphur-oxidising bacteria were already reported in mats over sulphur-rich sediments (Stolz, 1985; Mir et al., 1991; Guézennec et al., 2011) since

bacteria benefits from the O₂ released by the cyanobacteria. The development of *Thiomicrospira* in the discharge channel is clearly linked with the Bouillante plant activity, since this obligate sulphur-oxidising bacteria (Takai et al., 2004) quickly disappears when discharge waters stop flowing during maintenance work, cutting off the abundant sulphur supply (Pascal et al., 2017). The high concentration of Hg measured in the bacterial mats (13.01 µg.g⁻¹) (**Figure 2**) supported the probable release of Hg-rich discharge waters and that the bacterial consumption might be a significant source of Hg in *A. bahianus*, *A. saxatilis* and *D. antillarum*.

Regarding fish, the specimens from Bouillante displayed a muscle/liver ratio much lower than 1/3, suggesting that Hg was mainly entrapped in the liver likely under inorganic form (Scheuhammer et al., 2015). This ratio of liver/muscle ratios strongly differed between two fish species *A. saxatilis* and *A. bahianus* in the Control site (2 and 146 respectively) likely due to contrasting food sources with different chemical forms predominance. This ratio hugely increased in *A. saxatilis* (**Figure 1**) from Bouillante, slightly surpassing the value reported in *A. bahianus* (145 vs 122 respectively). Combined with 15-fold increase of THg in the liver (**Figure 1**), these results suggest either 1) unusual proportions (and high concentrations) of iHg in food sources as shown by the large Hg increase in the liver of the opportunistic damselfish and/or 2) a dietary shift, between the two sites, from omnivorous (with a more balanced MeHg/iHg containing prey) to a benthivorous/periphytophagous regime. This diet generally causes elevated liver/muscle ratio such as in *Acanthurus* species as this food source remained rich in iHg that is entrapped in the *A. saxatilis* liver (**Table 1**; Cidziel et al., 2002; Maury-Brachet et al., 2006). Basically, this hypothesis was supported by the partial diet shift showed by Pascal et al. (2017) based on stable isotope signatures, from planktonic preys, presumably containing varying MeHg (*i.e.* accumulated in fish muscles) from 20 to 70% (Harding et al.,

2018), to bacterial mats containing mostly inorganic Hg (*i.e.* stored in liver and kidneys; Maury-Brachet et al. 2006).

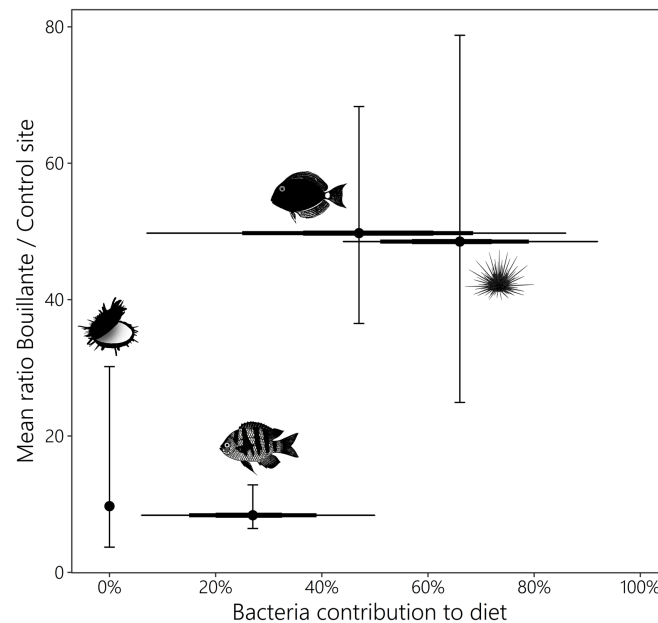


Figure 5: Relation between bacteria consumption and accumulation of Hg in muscles – Mean ratio between the Hg concentration in muscles collected in Bouillante and the Control Site compared with the bacteria contribution to diet found for other individuals of the same species by Pascal et al. (2017) in the Bouillante Bay. The three different thickness represent respectively the 25%, 75% and 95% credibility intervals (SIAR modelling) while the vertical bars represent the 95% confidence interval around the mean ratio of Hg concentration (iterations = 1,000,000).

In addition, it is noteworthy that the fish *A. bahianus* and the sea urchin *D. antillarum* showed the highest increase of THg in muscle between Control and Bouillante site, while both depend most on bacterial mats for food in Bouillante (Pascal et al., 2017). The figure 5 shows the relations between the Bouillante/Control site ratio of Hg concentrations and the contribution of bacteria in the diet of *A. saxatilis*, *A. bahianus*, *D. antillarum* and *S. tenuis*, calculated by SIAR modelling in Pascal et al. (2017). This qualitative representation suggests that bacterial mats are a source of Hg contamination for bacteria grazers but still raises the question of Hg speciation in these tissues and consequently in the bacterial mats, as MeHg usually concentrated

in muscular tissue (e.g. Storelli et al., 2003, Sandheinrich and Wiener, 2011). As none *Thiomicrospira* strains (NCBI, 2022), and generally no Gammaproteobacteria (Podar et al., 2015; Gionfriddo et al., 2016), possess the *hgcAB* gene pair coding for proteins responsible of Hg methylation (Parks et al., 2013), it is unlikely that bacterial mats are a source of MeHg in Bouillante. In addition, filter feeders, which are not known to consume bacterial mats still contain ~2.5 (sponges) to ~10 (*S. tenuis*) times more Hg in Bouillante than in the Control Site, suggesting another route of exposure, such as the waterborne pathway with iHg dissolved in seawater. Further analyses will have to clarify the Hg contamination and transfer processes that occur in front of the Bouillante plant, by investigating simultaneously Hg speciation in water, sediment, and biota and trophic ecology of the main marine species.

From a human health perspective, the MSWC in the three edible tissues surveyed in this study do not indicate a major risk to consumers in either the Hg_{inorg} or MeHg scenarios (**Figure 4**). For example, when considering Hg as inorganic, consuming ~34 fish *A. bahianus* (1.7 kg.week⁻¹, 95% CI [1.2, 2.6]) and ~186 clam *S. tenuis* (3.1 kg.week⁻¹, 95% CI [1.0, 7.8]) from Bouillante per week would be necessary to ingest harmful doses of Hg (**Table S3**). Similarly, the maximal dose of ~21 adult urchins' gonads per week (**Table S3**) when considering Hg as inorganic has poor chances to be crossed since *D. antillarum* is rarely consumed in Guadeloupe, notably due to its very painful sting (Coppard & Campbell, 2004). Nevertheless, if we assume that *A. bahianus* muscle mainly contain MeHg, only 700 g can be consumed a week based on the PTWI of 1.6 µg kg⁻¹ body weight of MeHg (JECFA). More largely, our results raise the question of human health risks associated to the consumption of direct predators of *A. bahianus* and *A. saxatilis* such as *Sphyraena barracuda*, *Caranx sp.* or *Lutjanus sp.* (Miller et al., 1971, Rumbold et al., 2018) which are present near the discharge channel (P-Y. Pascal pers. com.) and that fed on locally Hg contaminated fish.

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