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Mercury biomagnification and trophic structure patterns in neotropical coastal estuaries impacted by a Chlor-alkali plant in northeast Brazil

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Abstract :

The present study reports the concentration of HgT and MeHg in sediments and food webs in estuarine and coastal areas of Northeast Brazil, which have been subject to different impacts, mainly by chlorine and alkali companies. The results showed that the HgT in the sediment varied from 1.7 to 1186 ng.g⁻¹ and from 3.3 to 2163 ng.g⁻¹ in the organisms. A strong positive relationship between the levels of Hg in the sediment and the organisms was detected. Methylmercury represented 42 to 99% in the muscles of fish and invertebrates. Biomagnification processes were identified for all study areas evaluated according the trophic magnification factor (TMF), which ranged from 3.1 to 12.3. We found significant negative correlations with $\delta^{13}\text{C}$ values and mercury concentrations and positive correlations between $\delta^{15}\text{N}$ values and mercury, typical of Hg trophodynamics. This suggests a direct impact of the level of site-specific contamination on local food web and the ecosystem, making continuous monitoring studies necessary to assess food security and the risks that the riverine community is exposed as a result of ingesting mercury contaminated fish.

56 2. Introduction

57 Bleach (sodium hypochlorite) and sodium hydroxide are consumer chemical
58 products widely used. These products are industrially fabricated from sodium chloride
59 solutions by different electrolytic processes, including those employing a mercury (Hg)
60 cell (Crook and Mousavi, 2016; Mihaiescu et al., 2012), which is considered the oldest
61 and the most polluting processes for the environment and are still being used. In Brazil,
62 nine companies produce Chlor-alkali products and their subproducts. Among these
63 companies, four of them employ the Hg cell process, accounting for 14% of the annual
64 production, and are located in northeast Brazil (ABICLOR, 2012).

65 Brazil has one of the longest coastlines of the world and an enormous mangrove
66 area (13.400 km²). Due to their multiple uses and the ecosystem services they provide,
67 coastal and marine resources are fundamental for the development of the country (Burke
68 et al., 2001). The northeast is one of the most densely populated coastal regions of Brazil,
69 with the Pernambuco State standing out as the epicenter of this concentration.
70 Urbanization and degradation of coastal ecosystems have had multiple impacts, mainly
71 due to domestic pollution, industrial activity, and habitat degradation and loss, all of
72 which are most severe around the main urban zones (Mérigot et al., 2017).

73 Anthropogenic impacts related to elevated Hg levels in sediments of the drainage
74 basin of the North coast of Pernambuco (Northeastern Brazil) have already been
75 documented (Gondim, 2015; Lima et al., 2009; Lima, 2008). Early reports started in 1963,
76 right after the installation of a chlor-alkali plant. In 1982, new regulations were imposed
77 in Brazil (e.g. Decree 87.561/1982 and Law 9.976/2000) prohibiting the development of
78 new installations and/or the expansion of already existing sites employing the Hg cell
79 technique. Nevertheless, as already observed in other countries (Birkett et al., 2002;
80 Fitzgerald and Lamborg, 2013; Levin, 2015), the contamination problem has persisted

1 81 because Hg is highly stable in the environment and bioavailable through its
2 82 biogeochemical cycle for several years and decades, even after the source of
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4 83 contamination has disappeared.
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7 84 Worldwide studies on Hg contamination by Chlor-alkali plants evidence local
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9 85 impacts on coastal ecosystems (Adams and Paperno, 2012; Onsanit et al., 2012), with Hg
10 86 leading to possible pathological alterations in the organisms through trophic pathways
11
12 87 (Bravo et al., 2010; Soto et al., 2011). Since fish occupy a high trophic level and are
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14 88 consumed by the human population, the toxic organic form, methylmercury (MeHg), is
15
16 89 likely to be transferred to humans (Condini et al., 2017; Hosseini et al., 2013; Wu et al.,
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18 90 2019). The degree of human dependency on coastal ecosystems is very high in northeast
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20 91 Brazil. The Pernambuco coast has 34 fishing communities with around 12 000 fishermen
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22 92 (Lessa et al., 2006), which are dependent on estuarine and marine resources for food
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24 93 supply and commercial trade (Lira et al., 2017; Viana et al., 2015). Understanding the
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26 94 concentration and speciation of Hg at different trophic levels from this region is essential
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28 95 to assess the risk of exposure of Hg for the local biota and humans.
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36 96 The objectives of this study are to i) document total mercury (HgT) and MeHg
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38 97 concentrations in sediments, fish, and in the invertebrate community of four tropical
39
40 98 estuarine coastal zones of Pernambuco, northeast Brazil; and (ii) compare and model Hg
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42 99 biomagnification and bioaccumulation in aquatic biota at each site using complementary
43
44 100 carbon ($\delta^{13}\text{C}$) and nitrogen ($\delta^{15}\text{N}$) stable isotope data to diagnose Hg contamination in the
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46 101 study areas and its potential impact on food security of fish consumed by the riverside
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106 **3. Materials and methods**

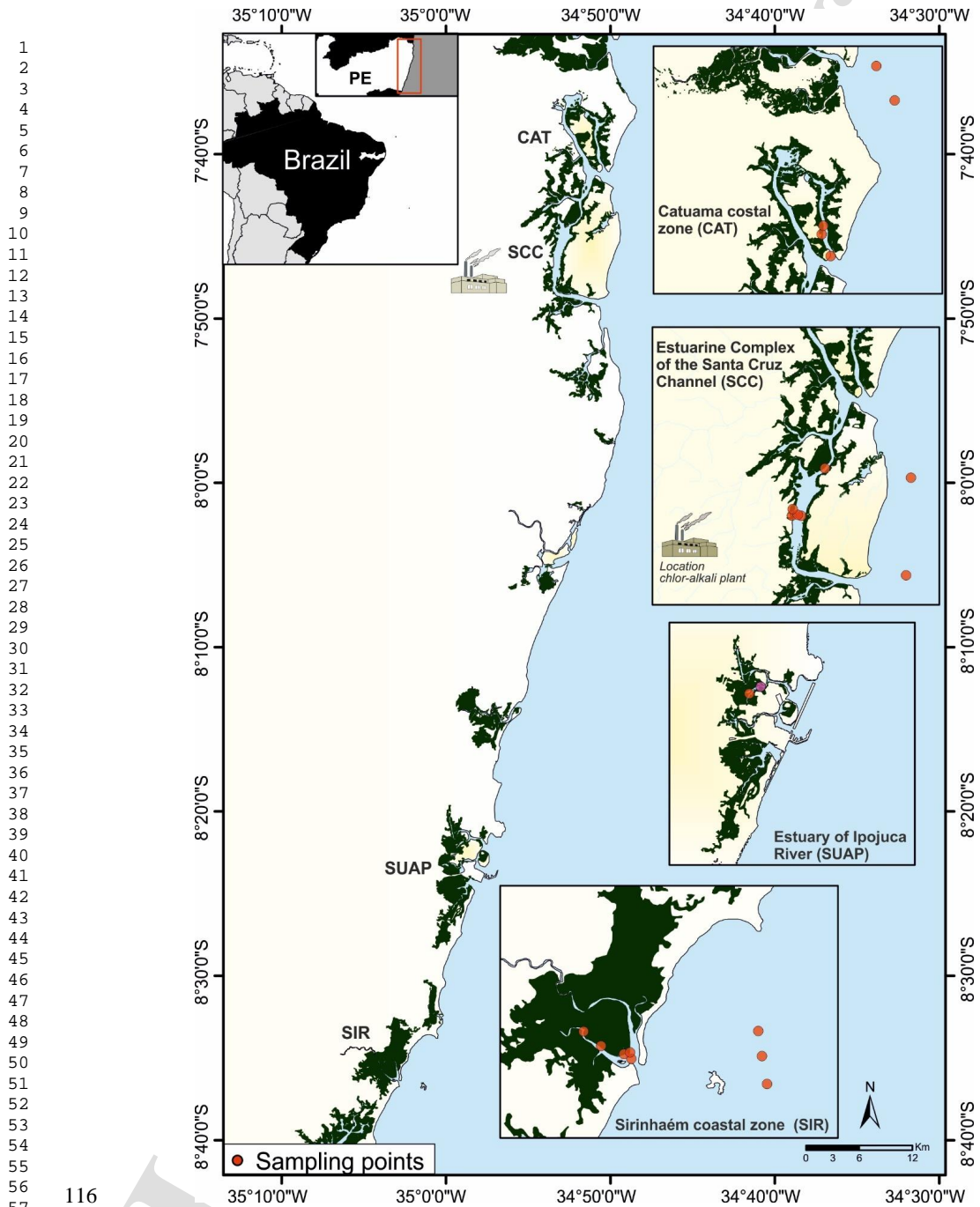
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108 **3.1. Study area and sample collection**

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110 Samples were collected in four estuarine and coastal areas of the Pernambuco
111 state, northeast Brazil: 1) The Estuarine Complex of the Santa Cruz Channel - SCC
112 (including the estuarine area and Itamaracá Island) and 2) the Catuama coastal zone–
113 CAT, located in the northern zone of Pernambuco; 3) The Sirinhaém coastal zone - SIR
114 and and 4) the estuary of Ipojuca River - SUAP (including the Suape Port area) both are
115 located in the southern zone of Pernambuco (Fig. 1).

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117 **Figure 1** –Study sites in the coastal zone and the mangrove area (green surface) of
 118 Pernambuco, Brazil, chlor-alkali plant (represented by illustration in SCC), CAT-

119 Catuama coastal zone, SCC- Estuarine Complex of de Santa Cruz Channel, SUAP-
120 Estuary of Ipojuca River, SIR - Sirinhaém coastal zone.

121
122 The Sirinhaém coastal zone (SIR) has 9.5 km long, 350 m wide, and 1.2–4.5 m in
123 depth. It is located in the Marine Protected Area of Guadalupe, which covers 450 km²,
124 encompassing Atlantic Forest, rivers, estuaries, mangroves, and coral reefs. Sirinhaém is
125 classified as a coastal plain estuary (Lira et al., 2010; Silva et al., 2011). The estuary of
126 Ipojuca River (SUAP) has a hydrographic basin of 3,800 km² and an approximate range
127 of 15 km. The mangrove suffered degradation following the construction of a large
128 industrial port complex between 1973 and 1976, involving chemical, shipping, and
129 logistics companies. The port resulted in a change in the tidal cycle, strong sedimentation
130 and high deposition of suspended sediments, increasing water turbidity and turning the
131 mouth of the estuary into a coastal lagoon; these effects led to a decrease in local depth
132 that increased salinity (Neumann et al., 1998; Paiva and Araújo, 2010).

133 The estuarine complex of the Santa Cruz Channel (SCC) has a U-shape, is about
134 22 km long, and has variable width that can reach up to 1.5 km. Depth ranges from 4 to
135 8 m, with higher values (10–17 m) close to the ocean discharges, which consist of
136 different rivers. The chlor-alkali plant was established in this complex in 1963 (Fig. 1).
137 The SCC is considered the largest estuarine ecosystem of Pernambuco state, with
138 irregular morphology where the tributaries run off to adjacent regions (Lira et al., 2017;
139 Silva et al., 2011). The SCC is connected to the Catuama coastal zone (CAT), via a *ria*-
140 type estuary (Silva et al., 2011). River discharge is the principal source of nutrients,
141 followed by sediment resuspension, mangrove litter, waste input, terrestrial runoff, and
142 atmospheric input (Medeiros et al., 2001).

143 The sediment, fish, and invertebrates were collected in the estuarine and an
144 adjacent coastal region from January to April and August to September 2015,

145 representing the dry and rainy seasons, respectively. As the estuaries and the coastal area
146 show distinct dynamics, different sampling protocols were used (according to Lira et al.,
147 2017; Mériqot et al., 2017; Silva et al., 2015). Block net and beach seines were adapted
148 for each estuary to cover most of the studied areas: shallow coastal areas and sand/mud
149 banks (beach seine), and flooded mangrove and channels (block net). The seine net was
150 used in the Santa Cruz Channel and the beach seine in the Sirinhaém estuary. The two
151 operations lasted no longer than 20 min and were repeated three times. Along the
152 mangrove forest, channel block nets were set, measuring 70 and 90 m long, 2.5 m high,
153 with a mesh size of 50 mm. At low tide, the net was anchored to the bottom. At slack
154 high-water, the net was deployed and attached to stakes and pulled taut so that it was
155 above the water, enclosing the mangrove area. Blocking was initiated at the end of high
156 tide and continued throughout the entire ebb tide cycle (approximately 6 h). In the coastal
157 environment, the gill net and the double trawler were used. Double trawler consisted of a
158 network 10 m in length with a mouth of 6.10 m, with 30 mm and 25 mm mesh size in the
159 net body and cod end, respectively. Data collection consisted of three hauls lasting two
160 hours each. The gill net was 690 m long and had mesh sizes of 50, 70, and 80 mm.

161 In the laboratory, the biological specimens, consisting of fish and invertebrates, were
162 identified following specific taxonomic keys (Allen, 1985; Carpenter, 2002; Figueiredo
163 and Menezes 2000; Whitehead, 1985, Tavares 2002). A total of 16 species of fish of
164 different trophic guilds and invertebrates were analyzed (Table S1).

165 The classification of the diet of each fish species was based on local studies of
166 stomach content or, when not available, on the literature (Freitas et al., 2011; Leão et al.,
167 2018; Lira et al., 2017, 2021; Mendoza-carranza, 2003; Monteiro et al., 2009;
168 Vasconcelos Filho, 1979; Vasconcelos Filho et al., 2003). The trophic guild was classified
169 according to Elliott et al. (2007) and Mourão et al. (2014): Filter feeder (FL);

170 Zooplanktivore (ZP); Detritivore (DV); Zoobenthivore (ZB); Omnivore (OV);
171 Piscivore/Zoobenthivore (PVZB) and Piscivore (PV). Individuals were measured (total
172 length for fish and shrimp, and carapace width for crab, in cm) and weighed (g). A
173 minimum of two individuals of each species was analyzed for each season (dry and rainy),
174 areas (CAT, SCC, SUAP, and SIR), and environment (coastal and estuary). For each
175 specimen, the muscle of the dorsal region (fish), a cephalothorax (shrimp), or the cheliped
176 muscle (crab) were removed, cleaned with distilled water, and dried at 60°C for 48 hours.

177 For the intertidal zone and the coast, about 5 g of sediments were collected
178 manually using a spatula during the low tide. For the main channel of the estuary, a core
179 of 20 cm length and 10 cm diameter was taken. For each season, area, and environment,
180 samples were collected in triplicate, using the three first mm of the surface layer,
181 representing the SOM (sediment organic material). The sediment was then fractionated
182 on meshes of 600 µm (coarse fraction) and 63 µm (fine fraction), to account for the effect
183 of changes in sediment granulometry based on the contrasted hydrological/accumulation
184 conditions present at the different sampling sites.

185

186 3.2. Total mercury (HgT) and methylmercury (MeHg) concentration analysis

187

188 The analysis of HgT in biological materials and sediments (ng Hg.g^{-1} , dry weight)
189 was performed with a DMA-80 (Milestone, USA) and AMA-254 (Leco, Europe) for the
190 sediments and the biological samples, respectively.

191 To investigate the robustness, accuracy, and precision of measurements, three
192 biological and one sediment certified reference materials (CRM), with varying
193 proportions of MeHg and HgT levels, were tested: TORT-3 (lobster hepatopancreas),

194 IAEA-436 (tuna tissue), SRM2976 (mussel tissue) and Mess-3 (marine sediment). For
195 each batch of ten samples, three blanks, and three CRMs were analyzed.

196 Methylmercury (MeHg) in biological samples were extracted and separated
197 according to Masbou et al. (2013). The analysis of the final purified MeHg fractions was
198 performed by cold vapor atomic fluorescence spectroscopy (CV-AFS). HgT
199 concentrations in both sediment and biological samples were determined on dry material.
200 HgT and MeHg concentrations for biological samples were converted to wet weight (wet
201 $\text{ng}\cdot\text{g}^{-1}$) with applying a moisture factor, previously determined for each sample (Table
202 S1).

203 To assess the potential impact of Hg levels, HgT concentrations recorded in
204 sediments were first compared to the Hg guidelines proposed by the “Canadian Sediment
205 Quality Guidelines for the Protection of Aquatic Life” (CCME, 1995). These guidelines
206 were developed to evaluate the degree of contamination and its likely impact on the
207 environment, and consist of Threshold Effect Levels (TELs = $130 \text{ ng Hg}\cdot\text{g}^{-1}$) and
208 Probable Effect Levels (PELs = $700 \text{ ng Hg}\cdot\text{g}^{-1}$): below the TEL- the minimal effect range
209 within which adverse effects rarely occur; between the TEL and PEL - the possible effect
210 range within which adverse effects occasionally occur; above the PEL - the probable
211 effect range within which adverse effects frequently occur (Macdonald et al., 1996).
212 Afterward, HgT and MeHg levels recorded in biological samples were compared to the
213 World Health Organization (WHO) guidelines (Canady et al., 2001; World Health
214 Organization, 1976; $1000 \text{ ng Hg}\cdot\text{g}^{-1}$ for predator and $500 \text{ ngHg}\cdot\text{g}^{-1}$ for the other fish).

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216 3.3. Stable isotope analysis and trophic level position

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218 To determine food web structure, carbon ($\delta^{13}\text{C}$) and nitrogen ($\delta^{15}\text{N}$) stable
219 isotopes were determined for each sample. The isotopic measurements were performed
220 by continuous flow Thermo Delta V+ mass spectrometer coupled via a Thermo ConFlo
221 IV interface to a Thermo Flash 2000 Elementary Analyzer at the Pôle de Spectrométrie
222 Océan (PSO, Plouzané, France). Standards used for $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ were Sigma-Peptide-
223 Home STD, IVA- casein, IAEA-Sucrose, IAEA-N1, IAEA-N2, and IAEA-Caffeine. The
224 home standard Thermo-Acetanilide was used for analytical precision of the analysis for
225 every six samples, indicating the precision of $\pm 0.11\text{‰}$ for carbon and $\pm 0.07\text{‰}$ for
226 nitrogen.

227 The trophic level (TL) of each species was estimated from $\delta^{15}\text{N}$ values using the
228 oyster species *Crassostrea rhizophorae* as a baseline. Except, for *Mugil* spp. in SCC and
229 CAT, where *G. stomatus* of near N value was considered as the most appropriate baseline.
230 TL of one consumer was computed as:

$$231 \quad \text{TL}_{\text{consumer}} = [(\delta^{15}\text{N}_{\text{consumer}} - \delta^{15}\text{N}_{\text{baseline}}) \times \Delta^{-1}] + \text{TL}_{\text{baseline}}.$$

232 with $\text{TL}_{\text{baseline}} = 2$ for both baselines, and $\Delta = 3.4\text{‰}$ as the isotopic discrimination factor
233 for $\delta^{15}\text{N}$ (Post, 2002).

234
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236 3.4. Methylmercury biomagnification model

237
238 To predict accumulation through the trophic web, we applied a biomagnification
239 model based on trophic magnification factors (TMF) and nitrogen stable isotopes,
240 according to Borgå et al. (2012) and McLeod et al. (2015). $\delta^{15}\text{N}$ values can be used to
241 estimate relative trophic level, and TMF was estimated as the slope of the linear
242 regression of log-transformed MeHg concentrations versus $\delta^{15}\text{N}$ values:

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$$243 \quad \log_{10}(MeHg) = b \cdot \delta^{15}N + a \quad \text{and} \quad TMF = 10^b$$

244 where the intercept a is the concentration of MeHg incorporated at the base of the
245 food web and b is the slope from the linear regression. A $TMF > 0$ indicates that
246 biomagnification has occurred (Lavoie et al., 2013). To investigate whether the slope b
247 was significantly different from 0 (i.e., if there was disequilibrium), a Student's t -test was
248 performed ($H_0 = 0$), with a confidence level of 95% (Zar, 2010). The differences between
249 areas by environment were also compared with t -tests.

250

251 3.5. Data analysis and statistics

252

253 Differences in HgT and MeHg were tested for the sediment and the organisms in
254 different areas (SIR, SUAP, SCC, and CAT), during both seasons (dry and rainy), two
255 sediment fractions (63 and 600 μm) and environment (coastal and estuary). We used two-
256 way ANOVA and Tukey's *post hoc* tests with data square-root transformed if required
257 (Zar, 2010) or Kruskal-Wallis non-parametric tests including multiple comparisons Z
258 scores. Pearson's correlation coefficient was used to test for correlations between HgT in
259 sediments and organisms, and between HgT and the isotopes $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$, by season
260 and area (Zar, 2010). All data were log-transformed.

261 A heat map visualization was performed to investigate the relationships between
262 HgT concentrations in organisms about study area and seasonality. Data were square-root
263 transformed and clustering was done using the Bray-Curtis dissimilarity and the
264 hierarchical complete linkage method. The color scale mosaic represents the HgT
265 concentration intensities (Wilkinson and Friendly, 2009). We used the *gplot* package from
266 the R program (Warnes et al., 2015).

267

268 4. Results and discussion

269

270 4.1. Mercury concentrations and distribution in estuarine sediments

271

272 Total mercury concentrations (HgT) in the sediment compartment ranged from
273 1.7 to 1186 ng.g⁻¹(Table 1), with a mean value of 288 ± 54 ng.g⁻¹. HgT concentrations in
274 the fine sediment fraction (< 63 μ m; Table 1) are significantly higher than that of the
275 coarse fraction (> 63 μ m and < 600 μ m) at all sites and for the two seasons (Kruskal-
276 Wallis $p < 0.001$), due to the strong affinity of Hg for small particles classes, with
277 enhanced specific surface and organic matter contents (García-Rico et al., 2006). Overall,
278 HgT concentrations in the North area (CAT and SCC) are 4-5 times higher than those
279 found in the south coast (SIR and SUAP) (Kruskal-Wallis $p < 0.001$). When size fractions
280 were considered, the fine fraction from CAT exhibited the highest concentrations during
281 the dry season and was significantly different from SCC. During the rainy season, the
282 coarse fraction from CAT had a lower concentration than SCC in the North area (two-
283 way ANOVA, $p < 0.05$, Table 1). HgT concentrations in southern areas (SIR and SUAP)
284 were similar, except during the dry season, with lower HgT concentration in SUAP for
285 both the fine and coarse fractions (two-way ANOVA, $p < 0.05$, Table 1). The lower HgT
286 concentrations in SUAP may be associated with the similarity of an anthropic source with
287 a low potential for mercury input in both areas, such as industrial and domestic waste
288 with low mercury enrichment (CPRH, 2001). HgT concentrations in CAT (mesh 600 μ m:
289 736 ± 158 and mesh 63 μ m: 1024 ± 58 ng.g⁻¹) and SCC (mesh 600 μ m: 547.5 ± 126.4 and
290 mesh 63 μ m: 520.2 ± 154.4 ng.g⁻¹) were above the TEL (130 ng.g⁻¹) and PEL (700 ng.g⁻¹)
291 guidelines, mainly during the dry season.

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293 **Table 1** – Total mercury concentrations (ng Hg.g⁻¹, dry weight) in sediments from the
 294 coastal zone of the Pernambuco region, Brazil. North zone: CAT- Catuama coastal zone
 295 and SCC – Estuarine Complex of the Santa Cruz Channel; South zone: SIR – Sirinhaém
 296 coastal zone and SUAP – Estuary of Ipojuca River; Mesh sizes: 600 and 63 μm; SE:
 297 Standard Error. Bold results correspond to locations where Hg concentrations exceed the
 298 threshold effect levels (TEL: 130 ng Hg.g⁻¹, dry weight) and probable effect levels (PEL:
 299 700 ng Hg.g⁻¹, dry weight).

Area	Season	Samples	63μm		600μm		
			Min-Max	Mean ±SE	Samples	Min-Max	Mean ±SE
CAT	Dry	4	863 – 1138	1.0248 ± 58.1	4	511 – 1186	735 ± 158
	Rain	4	364 – 514	456 ± 33.9	4	74.8 – 229	147 ± 36.6
SCC	Dry	5	242 – 955.7	547 ± 126	7	1.7 – 794	520 ± 154
	Rain	4	403 – 525	468 ± 29.4	7	3.4 – 355	293 ± 39.8
SIR	Dry	6	103 – 306	149 ± 31.5	4	31.6 – 135	89.2 ± 21.9
	Rain	4	96.7 – 134	113* ± 7.8	4	54.1 – 109	85.3 ± 14.3
SUAP	Dry	5	74.1 – 109	83.7* ± 6.4	4	2.7 – 92.4	33.5 ± 20
	Rain	2	87.2 – 90	88.6* ± 1.4	2	30.9 – 56.8	43.8 ± 12.9

300

301 HgT values in the sediment exceeded the acceptable limit for maintenance of the
 302 biota according to the areas and season (Table 1) in CAT and SCC. Lima (2008) evaluated
 303 the evolution of the sediment contamination by comparing sediment cores over
 304 approximately 150 years (1851 to 2004) in the same area. This author observed that the
 305 concentration of Hg at the base of the core was close to 150 ng.g⁻¹ (Table 2) and increased
 306 toward the top, exceeding 8 times the value established by PEL. The authors attributed
 307 this substantial increase to a single pollution source, which was the industrial production
 308 of chlor-alkali plant in the region. Studies conducted in the same region from 1981
 309 showed no subsequent reduction in Hg contamination in the sediment, with values
 310 ranging between 110 and 3700 ng.g⁻¹ (Table 2) (Gondim, 2015; Lima et al., 2009; Lima,
 311 2008). Meyer et al. (1998) observed HgT between 300 and up to 20500 ng.g⁻¹ in the grain
 312 size fraction < 63 μm (Table 2), and between 430 and 5560 ng.g⁻¹ in the suspended matter.

313 The HgT concentrations observed in this study are considerably high and similar
 314 to other areas also impacted by chlor-alkali plants worldwide (Table 2). Specifically, in
 315 the Northern area (CAT and SCC), the concentration of Hg ranged from 147 to 1024 ng.g-

1, evidencing the persistent contamination by this trace metal in relation to previous studies. Hg adsorption occurs in both granulometries (63 and 600) fractions evaluated, but the higher clay and organic matter content of the smaller fraction (63) results in greater adsorption and metabolization of mercury into its organic species, and longer persistence in the environment. It is important to consider that the study area, located in the intertidal zone of the estuary, is a zone of high hydrological dynamics influencing the sedimentation of mercury. Moreover, the decrease in the concentration of Hg in the soil may be linked to the sedimentation rate of non-enriched geological material as reported by Albuquerque et al.(2019).

Gondim (2015) and Lima (2008) concluded that Hg transfer occurred from the site where the metal was discharged (via the river) into the estuary and Santa Cruz Channel (SCC), so the metal in the sediment was deposited into the river flow. In addition, Lima et al. (2009) showed that the deposition occurs mainly on the banks of the channel, because with the less interference of the currents occurs the greatest accumulation of fine particles, justifying an influence that is still measurable today.

Table 2- Total mercury concentrations (ng Hg.g⁻¹, dry weight) in the sediment of this study area and other coastal areas also contaminated by chlor-alkali industrial activities.

Location	THg (ng.g ⁻¹)	Sampled year	Authors
Study area – Brazil			
Botafogo River	71–222 ^a	1851–1961 ^b	Lima (2008)
Botafogo River	398–5948 ^a	1963 ^c –2004	Lima (2008)
Botafogo River	110–3700	1981 ^d	Lima et al. (2009)
Santa Cruz Channel	300–20500	1993–1994	Meyer et al. (1998)
Botafogo River	130–240	2004–2006	Lima et al. (2009)
Botafogo River	480–6900	2011	Gondim (2015)
Botafogo River	130–10440	2017	Araújo et al (2019)
Botafogo River	100–14400	2015	Araújo et al. (2021)
Itapessoca River	40–1290 ^a	2017	Albuquerque et al. (2019)
Santa Cruz Channel	147–1024.3	2015	This Study
Other regions - Brazil			
Guanabara Bay	42–7506	2005	Machado et al. (2008)
Portugal			

1	Estarreja	117–49233	1998	Inácio et al. (1998)
2	France			
3	Thur River basin	430–13000	2001–2002	Hissler and Probst (2006)
4	Spain			
5	Cinca River	15–400	2002	Raldúa et al. (2007)
6	Cuba			
7	Sagua River	160–9700	Before 2015	Bolaños-Álvarez et al. (2016)
8	USA			
9	Androscoggin River	33–2045	2010–2011	Buckman et al. (2015)

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12 333 ^a Sediment core.

13 334 ^b Without contamination before the establishment of the chlorine industry.

14 335 ^c Establishment of the chlorine industry.

15 336 ^d First monitoring in the area by CETESB - Environmental Company of the State of São
16 337 Paulo.

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20 339 Hg contamination was higher in sediments during the dry season than during the
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22 340 rainy season (Table 1). In tropical latitudes, one of the main factors influencing the
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24 341 concentration or dilution of Hg is the volume of river discharge (Costa, et al., 2009;
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26 342 Barletta et al., 2012; Di Benedetto et al., 2013).. In areas adjacent to those of the present
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28 343 study, Costa et al. (2009) and Lima (2008) showed that when the rainfall increases during
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30 344 the rainy season, river drainage dilutes Hg.

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35 345 These results indicate that the Hg concentrations in the sediments at the four
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37 346 investigated sites are a function of: (i) the distance of the source of Hg pollution (chlór-
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39 347 alkali plant), e.g., SCC and CAT are close to important sources of Hg and presented high
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41 348 Hg values whereas in SUAP and SIR, away from strong mercury sources, the
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43 349 concentration is low; and (ii) the hydrological control of the estuarine system between
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45 350 wet and dry seasons.

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52 352 4.2. Mercury concentrations, speciation, and distribution in the estuarine food webs

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56 353 Similar to what was observed for sediments, the HgT concentrations in estuarine
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58 354 organisms of CAT and SCC were significantly higher than those reported in SIR and

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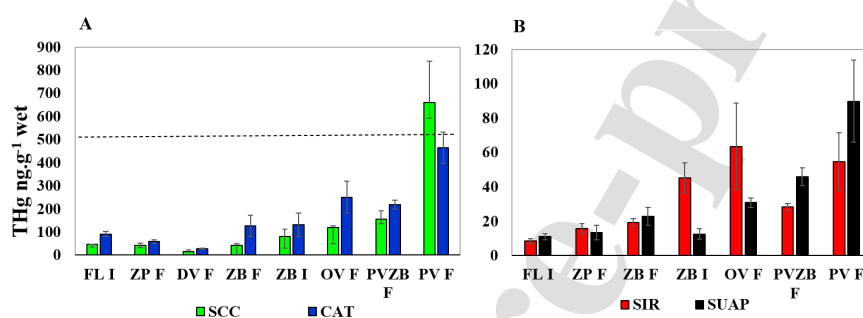
355 SUAP (Kruskal-Wallis, $p < 0.001$) (Table S2 to S5 in supplementary material). When the
356 areas were compared between the coastal and estuarine environments, no significant
357 differences were found for CAT and SCC (Kruskal-Wallis, $p > 0.05$). However, significant
358 differences were found between seasons, with higher concentrations observed in the dry
359 season for all areas (Kruskal-Wallis, $p < 0.05$). This may be linked to a reduction of river
360 discharge due to lower freshwater input from natural sources, such as rainfall. In contrast,
361 the increased river discharge during the rainy season causes the dilution of contaminants
362 and/or their displacement within the estuary. A strong relationship was observed between
363 the HgT concentrations in sediments and the corresponding Hg levels in organisms for
364 CAT and SCC (north area) (Pearson, $r = 0.82$, $p < 0.05$) suggesting direct site-specific
365 contamination of the local food webs and the close ecosystem. In contrast, SIR and SUAP
366 (south area) did not exhibited a significant relationship between HgT concentrations in
367 the sediment and the corresponding Hg levels in organisms (Pearson, $r = 0.70$, $p > 0.05$).

368 The total mercury concentration observed in organisms ranged from 3 to 2163 ng.g⁻¹
369 (wet weight) (Table S2 to S5). We recorded high concentrations above WHO guidelines
370 (Fig.2) for some species, such as *Centropomus undecimalis* and *Bardiella ronchus* for
371 CAT as well as *Bardiella ronchus* and *Caranx hippos* for SCC.

372 Considering that some fish species are more generalists than others, the
373 contamination by Hg can occur indirectly via consumption of different resident
374 invertebrates and fishes that have fed on other contaminated sources; this can lead to the
375 greater the intake of Hg. However, further studies focused on prey and predators need to
376 be developed to investigate this issue. The increase in the mercury concentration can also
377 be influenced by the variability in the consumption of small fish by larger fish (Hosseini
378 et al., 2013), such as *Caranx hippos*. Methylmercury represented 42 to 92% of HgT
379 concentrations in invertebrates and 56 to 99% in fish (Tables S2 to S5). This means that

380 the coastal area of Pernambuco offers favourable conditions for the methylation of
 381 inorganic mercury and the bioavailability of *in situ* produced MeHg to these organisms.
 382 Piscivore and piscivore/zoobenthivore fish showed higher MeHg concentrations than
 383 invertebrates, with methylmercury accounting for 70 to 90% of HgT.

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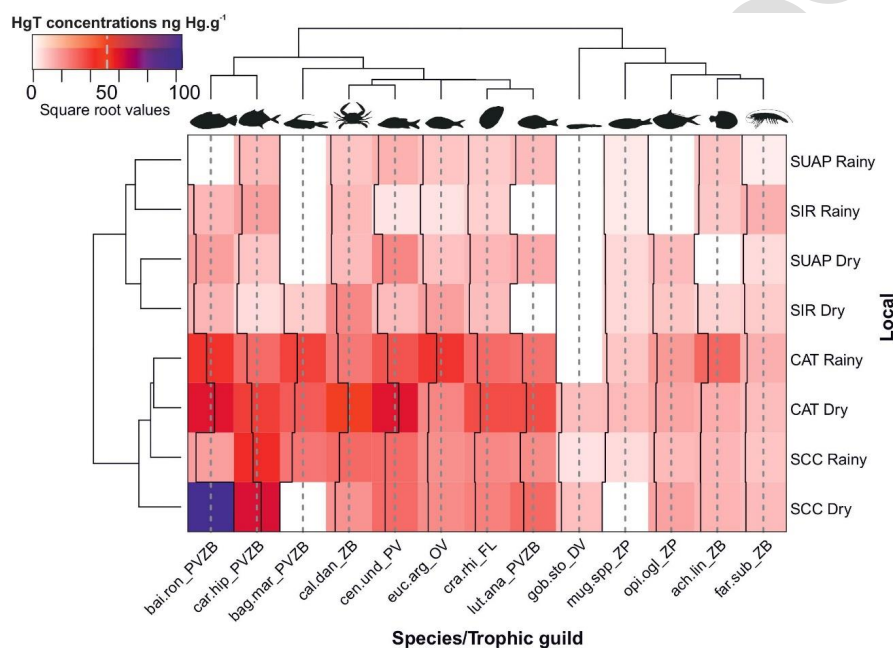
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 386 **Figure 2** –Total mercury for organisms by area and trophic guild. A) CAT – Catuama
 387 coastal zone and SCC – estuarine complex of Santa Cruz Channel; B) SIR – Sirinhaém
 388 coastal zone and SUAP – estuary of Ipojuca River. Filter feeder (FL); Zooplanktivore
 389 (ZP); Detritivore (DV); Zoobenthivore (ZB); Omnivore (OV); Piscivore/Zoobenthivore
 390 (PVZB); Piscivore (PV); I – Invertebrate and F – Fish. The horizontal dashed line at 500
 391 ng.g⁻¹ indicates the limit recommended by the WHO for non-piscivores.

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393 Two groups were observed considering the distribution of the organisms
 394 according to the relationship between trophic guild and HgT: (i) one representing the area
 395 contaminated by anthropogenic mercury (northern zone) and (ii) the area reflecting more
 396 natural Hg inputs (southern zone) (Table1, Fig. 3). The dendrogram also showed the
 397 formation of two groups segregated by trophic position, and the formation of four
 398 subgroups differentiated by accumulated Hg concentrations.

399 The first group was represented by organisms that occupy lower levels
 400 compartments of the food web and present lower HgT concentrations, such as
 401 zooplanktivores (ZP), zoobenthivores (ZB), and detritivores (DV). The second group of
 402 organisms is formed by the carnivorous generalist and specialist, such as omnivores (OV),
 403 piscivores/ zoobenthivores (PVZB), and piscivores (PV), with higher Hg concentrations

404 (Fig. 3). The mean HgT concentrations by trophic guild revealed that, piscivores had
 405 higher Hg concentrations compared to the other trophic guilds (Fig. 2) (Kruskal-Wallis,
 406 $p < 0.001$) for all areas, except to SIR where omnivorous Hg concentrations were higher.
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408
 409 **Figure 3** – Cluster heatmap of total mercury concentrations in organisms from study areas
 410 using a color scale ranging from light pink (low concentration) to purple (high
 411 concentration). CAT – Catuama coastal zone and SCC – estuarine complex of Santa Cruz
 412 Channel; SIR – Sirinhaém coastal zone and SUAP – estuary of Ipojuca River. Filter feeder
 413 (FL); Zooplanktivore (ZP); Detritivore (DV); Zoobenthivore (ZB); Omnivore (OV);
 414 Piscivore/Zoobenthivore (PVZB); Piscivore (PV); ach.lin – *A. lineatus*; opi.ogl – *O.*
 415 *oglium*; far.sub – *F. subtilis*; gob.sto – *G. stomatus*; mug.spp - *Mugil spp.*; cal.dan – *C.*
 416 *danae*; cen.und – *C. undecimalis*; euc.arg – *E. argenteus*; bag.mar – *B. marinus*; cra.rhi –
 417 *C. rhizophorae*; lut.ana – *L. analis*; bai.ron – *B. ronchus*; car.hip – *C. hippos*.

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 422 Among the species at the base of the food web (first group), the zooplanktivore
 423 *O. oglium* and the zoobenthivore fish *A. lineatus* presented the highest mercury
 424 concentrations during the rainy season, mainly in CAT coastal areas. On the other hand,
 425 the detritivore *G. stomatus* and zooplanktivore *Mugil spp.* showed the highest Hg
 426 concentrations during the dry season (Fig. 3, Tables S2 to S5). In the second group, the

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427 piscivore/zoobenthivore *B. marinus* and the omnivore *E. argenteus* showed the highest
428 Hg concentrations during the rainy season, while the other species showed enriched Hg
429 concentrations during the dry season. The predators *B. ronchus* and *C. hippos* displayed
430 the highest HgT levels relative to all other organisms and therefore they were grouped as
431 similar in the dendrogram, mainly in SCC and CAT, in dry season (Fig. 3, Tables S2 to
432 S5). High Hg concentrations were also reported for the filter feeder *C. rhizophorae* and
433 the zoobenthivore *C. danae* of SCC, displaying similar Hg concentrations to organisms
434 of higher trophic levels (Fig.3, Tables S2 to S5).

435 Our results demonstrate the occurrence of biomagnification along the trophic
436 chain, in the order of detritivore < zooplanktivore < filter feeder < zoobenthivore <
437 piscivore/zoobenthivore < piscivore, increasing from the lower trophic position,
438 represented by invertebrates and primary consumers up to the fish predators. It is well
439 documented that mercury, mainly in the form of methylmercury, is more concentrated at
440 higher trophic levels, where predators feed on contaminated prey (Di Benedetto et al.,
441 2013; Hosseini et al., 2013; Scudder Eikenberry et al., 2015).

442 443 4.3. Mercury biomagnification trends in the estuarine food webs of the Pernambuco 444 region

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446 For CAT and SCC areas, correlations between HgT and MeHg with $\delta^{13}\text{C}$ were
447 negative and significant: the more enriched the samples in ^{13}C , the lower their HgT
448 concentration (Fig. 4, Table S2-S3, Pearson = Dry: -0.69 (CAT), -0.78 (SCC), $p < 0.05$
449 and Rainy: -0.62, $p < 0.05$ and -0.52, $p > 0.05$, respectively CAT and SCC). Enriched
450 carbon values (-17.05 to -13.27) were found in the low trophic guilds' species, such as
451 *Mugil spp.*, *O. oglium*, *G. stomatus*, *E. argenteus*, *C. danae*, *F. subtilis*, and juveniles of

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452 *C. hippos*. However, in SIR and SUAP areas (south zone), the results showed weaker
453 correlations for both seasons, but tended to be negative in the dry season and slightly
454 positive during the rainy season (Fig. 4, for SIR Pearson = Dry: -0.08, $p > 0.05$; Rainy:
455 0.47, $p > 0.05$; for SUAP Pearson= Dry: -0.28; Rainy: 0.43, $p > 0.05$).

456 Correlations between HgT and MeHg with $\delta^{15}\text{N}$ were higher during the dry season
457 than the rainy season, in both areas (Fig.5). In CAT, the Pearson correlation for the dry
458 season was 0.80 for HgT and 0.77 for MeHg and, in the rainy season, 0.73 for HgT and
459 0.78 for MeHg ($p < 0.05$, Fig. 4-5). Finally in SCC, the correlation between HgT and
460 MeHg with $\delta^{15}\text{N}$ were 0.80 and 0.90 respectively. This result indicates that Hg levels
461 increase with trophic level (Fig. 5).

462 The negative correlation between HgT and $\delta^{13}\text{C}$ observed in the northern zone can
463 be related to feeding behaviour (trophic guild), considering the nutritional source used by
464 the species. Studies show that primary consumers can assimilate nutrients derived from
465 detritus, macroalgae, microalgae, and seagrass that are the source of carbon and energy
466 at the base of the food web in estuaries (Adams and Paperno, 2012; Claudino et al., 2015).
467 Several authors reported that consumers with lower $\delta^{13}\text{C}$ values combined with reduced
468 $\delta^{15}\text{N}$ values, have benthic-based prey as their consumption preference (Adams and
469 Paperno, 2012; Fry et al., 2008; Peterson et al., 1985). This corroborates with the increase
470 in exposure and greater accumulation of Hg into biota because most of the Hg
471 contamination is concentrated in the sediment. In an estuary of northeast Brazil, Claudino
472 et al. (2015) discussed that the higher $\delta^{13}\text{C}$ values found in fishes probably were a result
473 of the values of this $\delta^{13}\text{C}$ in the seagrass and macroalgae which were incorporated by the
474 fishes directly or indirectly via consumption of invertebrates that feed upon mangrove
475 detritus.

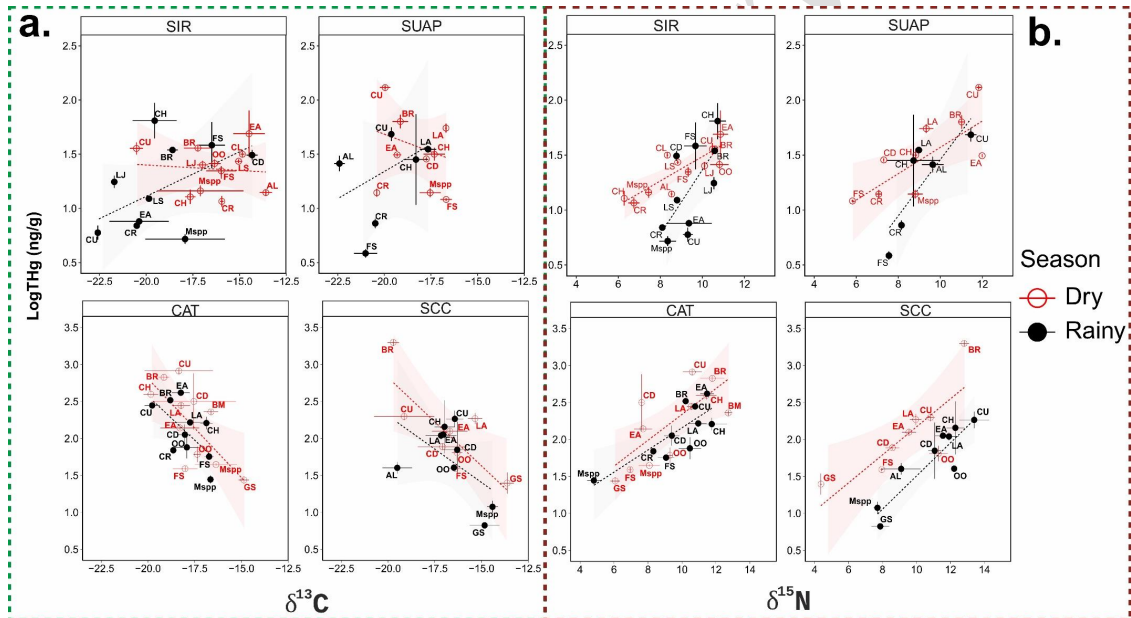


Figure 4 - Scatterplots between nitrogen isotope ($\delta^{15}\text{N}$), carbon ($\delta^{13}\text{C}$), and the logarithm of total mercury (Log HgT) for the coastal of Pernambuco, Brazil. CAT – Catuama coastal zone and SCC – estuarine complex of Santa Cruz Channel; SIR – Sirinhaém coastal zone and SUAP – estuary of Ipojuca River (statistics on table S6 in supplementary material) Species code: AL: *Achirus lineatus*; BM: *Bagre marinus*; BR: *Bardiella ronchus*; CD: *Callinectes danae*; CH: *Caranx hippos*; CL: *Caranx latus*; CU: *Centropomus undecimalis*; CR: *Crassostrea rhizophorae*; EA: *Eucinostomus argenteus*; FS: *Farfantepenaeus subtilis*; GS: *Gobionellus stomatus*; LA: *Lutjanus analis*; LJ: *Lutjanus jocu*; LS: *Lutjanus synagris*; Msp: *Mugil spp.*; OO: *Opistonema oglium*.

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476 The proportion of HgT as methylmercury with HgT concentrations varied from
477 the mean value of $41 \pm 3\%$ (*F. subtilis*) to $92 \pm 5\%$ (*C. danae*) for invertebrates, and from
478 $56 \pm 3\%$ (*L. analis*) to $99 \pm 19\%$ (*B. ronchus*) for fish, with higher proportions observed
479 in the coastal area of SCC and the estuarine area of SUAP (Tables S3 and S5). There were
480 no significant differences in the proportion of HgT as methylmercury between the dry
481 and rainy seasons when all areas were gathered (two-way ANOVA, $p > 0.05$). The biplot
482 between %MeHg and $\delta^{15}\text{N}$ showed a positive correlation, with the top predator presenting
483 a higher accumulation MeHg than those at the base of the trophic chain (Fig. 5).

484 For all studied areas, these results were confirmed by the methylmercury
485 biomagnification model. All TMFs were higher than 1 ($b \neq 1$, t-test: $p < 0.05$) (Table 3)
486 and there were significant differences in slope in different zones of the same environment
487 (t-test: $p < 0.05$), ranging between 3.1 and 12.3 in SUAP estuary and SCC coastal area
488 respectively, except for SIR and SUAP which were similar. The HgT and, in particular
489 MeHg concentration are magnified up to 12 times for each unit of increase in trophic level
490 as found in SCC. Other documented environments with TMF values higher than 4, such
491 as the Gulf of St. Lawrence in Canada and the northern coast of Rio de Janeiro, are also
492 known to be highly influenced by anthropogenic processes and mercury discharges into
493 the water (Di Benedetto et al., 2012; Lavoie et al., 2010).

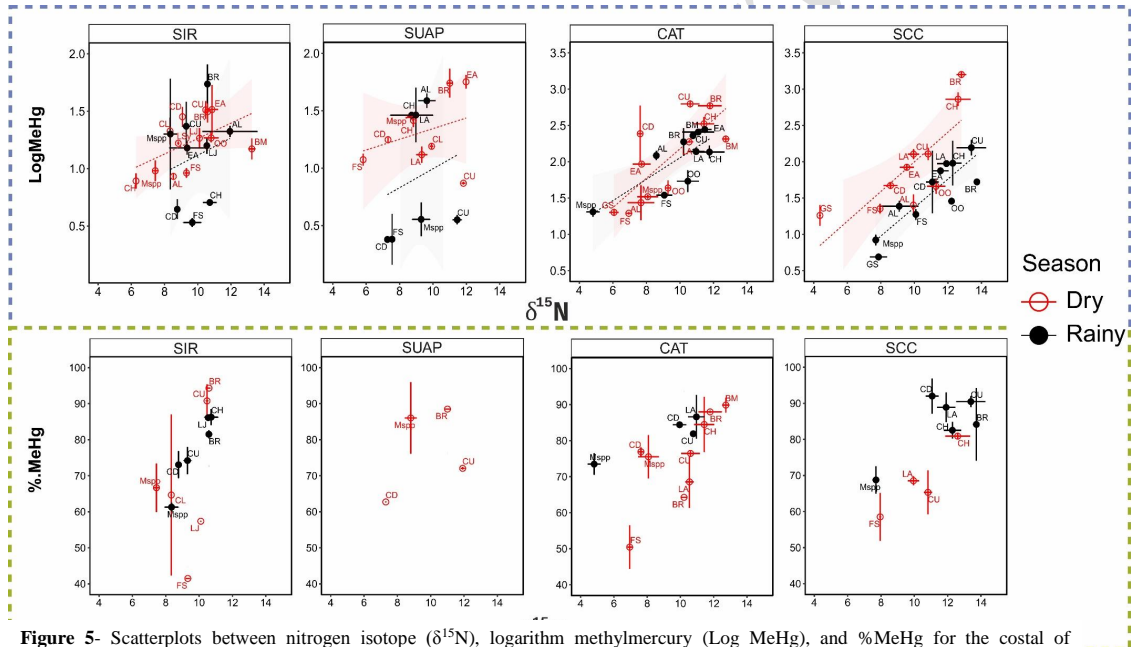


Figure 5- Scatterplots between nitrogen isotope ($\delta^{15}\text{N}$), logarithm methylmercury (Log MeHg), and %MeHg for the coastal of Pernambuco, Brazil. CAT – Catuama coastal zone and SCC – estuarine complex of Santa Cruz Channel; SIR – Sirinhaém coastal zone and SUAP – estuary of Ipojuca River (statistics on table S7 in supplementary material). Species code: AL: *Achirus lineatus*; BM: *Bagre marinus*; BR: *Bardiella ronchus*; CD: *Callinectes danae*; CH: *Caranx hippos*; CL: *Caranx latus*; CU: *Centropomus undecimalis*; CR: *Crassostrea rhizophorae*, EA: *Eucinostomus argenteus*, FS: *Farfantepenaeus subtilis*; GS: *Gobionellus stomatus*; LA: *Lutjanus analis*; LJ: *Lutjanus joca*; LS: *Lutjanus synagris*; Msp: *Mugil spp.*; OO: *Opistonema ogtium*.

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Table 3- Methylmercury biomagnification model for the coastal zone of Pernambuco, Brazil. N – Number of individuals; S – Number of Species evaluated; b = slope of the regression curve; a = intercept of the concentration of MeHg in the baseline; CI – Confidence interval; R^2 = Coefficient of Pearson r -squared; TL – Trophic level (Min – minimum and Max – maximum); TMFs - Trophic magnification factors.

Area: Environment	N	S	a	95%CI a	b	95% CI b	R^2	TLMin- Max	TMFs
Catuama coastal zone									
Coastal	26	5	-0.361	(-0.986) - (0.263)	0.81	(0.596) - (1.022)	0.72	2-3.81	6.45
Estuary	43	7	-0.184	(-0.837) - (0.468)	0.89	(0.613) - (1.157)	0.51	2-3.34	7.76
Estuarine complex of the Santa Cruz Channel									
Coastal	19	5	-1.346	(-2.823) - (0.130)	1.09	(0.594) - (1.593)	0.54	2-4.59	12.30
Estuary	41	7	-0.635	(-1.305) - (0.035)	0.90	(0.655) - (1.154)	0.58	2-3.95	7.94
Sirinhaém coastal zone									
Coastal	22	6	-1.157	(-2.052) - (0.262)	0.88	(0.560) - (1.198)	0.62	2-3.95	7.58
Estuary	34	6	-0.128	(-0.593) - (0.337)	0.51	(0.330) - (0.686)	0.51	2-3.55	3.23
Suape Port									
Estuary	33	5	0.194	(-0.267) - (0.657)	0.49	(0.312) - (0.669)	0.53	2-3.6	3.10

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500 The regression slopes (b) were used to evaluate the biomagnification power of
501 MeHg for the estuaries in the present study (Table 3). Smaller values of b indicate low
502 TMF, and higher values, the opposite. The results clearly showed values much higher
503 than those observed worldwide (CAT: 0.89, SCC: 0.90, SIR: 0.51, SUAP: 0.49), showing
504 that the anthropic inputs transferred to the food webs are higher in the north than in the
505 south zones. This suggests that the Chlor Alkali plant has a direct impact from the site-
506 specific contamination level in SCC and CAT, because of the concentration of mercury
507 in food webs and sediments. In temperate marine ecosystems, b fluctuates between 0.235
508 and 0.260; in tropical waters between 0.140 and 0.833; in polar marine areas from 0.210
509 to 0.941 and tropical freshwater from 0.08 to 0.430 (Di Benedetto et al., 2012; Lavoie et
510 al., 2013, 2010; Poste et al., 2015; Pouilly et al., 2013; Ruus et al., 2015).

511 The other parameter of the mercury biomagnification model, the intercept (a), can
512 be considered as an estimate of the Hg incorporation at the base of the food web (Borgå
513 et al., 2012). Initially, it is possible to observe an association between the trophic
514 interception and amplification. The lower the value of a in SCC (-1.34) the higher the
515 TMF (12.30), and the higher value of a in SUAP (+0.19), presented the lowest TMF
516 (3.10). The trophic level may also influence the intercept value as it was possible to
517 observe in the present study since the values of the intercept on the coast were lower than
518 those obtained in the estuary for all areas, consequently, the coastal environments reached
519 the highest maximum trophic level. Thus, through this parameter, it was observed that a
520 SUAP baseline was the least impacted and SCC the one with the greatest impact among
521 the studied areas, pointing out that regardless of local contamination, the range of
522 variation of trophic levels in the area, trophic guild is one of the main factors that interfere
523 in the increase of biomagnification of mercury in the environment. Studies developed by
524 Di Benedetto et al. (2012) in Rio de Janeiro corroborate our study about the rate of

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525 biomagnification. Lavoie et al. (2013) explained that, in warmer regions, the trophic
526 transfer efficiency of Hg and, therefore, biomagnification would be reduced at each
527 trophic step, which is the opposite of what was observed in this study. Furthermore, these
528 authors and Al-Reasi et al. (2007) observed that high diversity of species, characterized
529 by a complex food web, could potentially reduce the efficiency of trophic Hg transfer.
530 The results of the present study, that high diversity was not associated with reduced
531 biomagnification, also contradict this hypothesis.

532 High diversity of species found in these regions can mean a longer food web,
533 which typically increases relative bioaccumulation at the top (Post, 2002). Doi et al.
534 (2009) suggested that resource availability and ecosystem size can predict the food-chain
535 length. The two characteristics are present in the estuaries and coastal region of
536 Pernambuco: a heterogeneous resource availability and a highly complex system
537 (Gonzalez et al., 2019; Lira et al., 2017, 2021; Mérigot et al., 2017; Silva-Júnior et al.,
538 2017, Ferreira, 2019). However, from the diversity of species evaluated in the region was
539 not associated with reduced biomagnification.

540 The significant correlation between HgT and $\delta^{15}\text{N}$ ($R^2 > 0.50$) showed the
541 importance of mercury transfer through the food web, confirmed by the TMF results
542 (TMF > 1) which demonstrated the occurrence of bioaccumulation. The use of $\delta^{15}\text{N}$
543 verified the accuracy of the trophic position of species (Lavoie et al., 2010; Scudder
544 Eikenberry et al., 2015). This could explain why, in the northern zone, the organisms of
545 higher trophic levels showed a higher concentration of mercury. The TMF assumes that
546 the main source of the exposure of organisms to contaminants in the diet and, this
547 absorption and transfer occurs faster than elimination, resulting in an accumulation that
548 increases with increasing TL (Borgå et al., 2012).

550 5. Conclusions

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2 551 This study is the first to use stable isotopes of carbon ($\delta^{13}\text{C}$) and nitrogen ($\delta^{15}\text{N}$)
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4 552 associated with Hg and MeHg to evaluate the biomagnification and contamination by
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7 553 mercury in food webs in estuarine/coastal zones of Northeast Brazil. The industrial Hg
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9 554 contamination occurring in the Northern Coast of Pernambuco is attributed to a chlór-
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11 555 alkali company installed in 1963, which after about 29 years of direct discharge of
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13 556 mercury-enriched effluents into the estuary, still presents high Hg levels in the sediment,
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16 557 above the acceptable limit for biota in the two areas influenced by this industry (CAT and
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18 558 SCC). The increased concentrations of HgT between sediments and organisms in CAT
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20 559 and SCC areas relative to SIR and SUAP is clear evidence that basal levels of Hg are the
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22 560 main route of Hg concentration transfer along the food web to the upper trophic levels.
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24 561 Among the sources of mercury, the most relevant is the chlór-alkali plant, influencing the
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27 562 biomagnification process that took place in all study areas.
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31 563 Our results also demonstrate the occurrence of Hg biomagnification along the
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33 564 trophic chain was confirmed by a methylmercury biomagnification model. We found a
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35 565 significant correlation of HgT and MeHg with carbon and nitrogen stable isotope ratios
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37 566 for the four areas, indicating that there is an enrichment of the nitrogen isotope with the
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39 567 increase of trophic level but that the opposite occurs for the carbon isotope, with a
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41 568 negative correlation related to the food sources used by these species.
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45 569 The biomagnification of mercury, mainly in the form of methylmercury, in areas
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47 570 where the chlor-alkali industry operates is a reality, so it is necessary to improve existing
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49 571 legislation and mainly to provide the continuous monitoring of the areas already
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51 572 impacted, by obligating a complete change of the existing Hg process, by other less
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53 573 polluting technologies. This work reveals that even if the Hg inputs from the chlór alkali
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55 574 industry are over, Hg is highly stable and accumulated in the sediment compartment
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2 575 leading to legacy contamination of the entire nearby ecosystems for decades and likely
3 576 centuries.

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8
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32 899 **6. Supplementary material**

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1 **Highlight**

- 2 • Hg pollution and C and N stable isotopes were examined in four Brazilian
3 estuaries.
- 4 • Increased concentrations of HgT between sediments and organisms were
5 measured.
- 6 • Methylmercury represented 42 to 99% of HgT in the muscles of fish and
7 invertebrates.

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Declaration of interests

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.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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