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1 **Mangrove microbiota along the urban-to-rural gradient of the Cayenne**
2 **estuary (French Guiana, South America): drivers and potential**
3 **bioindicators**

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29

30 **Keywords:** Estuarine mangrove sediments, French Guiana, low organic matter enrichment,
31 organochlorine contaminants, PAHs contamination

32

33 **Abstract:**

34 The microbial communities inhabiting the Atlantic-East Pacific (AEP) mangroves have been
35 poorly studied, and mostly comprise chronically polluted mangroves. In this study, we
36 characterized changes in the structure and diversity of microbial communities of mangroves
37 along the urban-to-rural gradient of the Cayenne estuary (French Guiana, South America) that
38 experience low human impact. The microbial communities were assigned into 50 phyla.
39 Proteobacteria, Chloroflexi, Acidobacteria, Bacteroidetes, and Planctomycetes were the most
40 abundant taxa. The environmental determinants found to significantly correlated to the
41 microbial communities at these mangroves were granulometry, dieldrin concentration, pH,
42 and total carbon (TC) content. Furthermore, a precise analysis of the sediment highlights the
43 existence of three types of anthropogenic pressure among the stations: (i) organic matter
44 (OM) enrichment due to the proximity to the city and its wastewater treatment plant, (ii)
45 dieldrin contamination, and (iii) naphthalene contamination. These forms of weak
46 anthropogenic pressure seemed to impact the bacterial population size and microbial
47 assemblages. A decrease in Bathyarchaeota, "*Candidatus Nitrosopumilus*", and *Nitrospira*
48 genera was observed in mangroves subjected to OM enrichment. Mangroves polluted with
49 organic contaminants were enriched in Desulfobacteraceae, Desulfarculaceae, and
50 Acanthopleuribacteraceae (with dieldrin or polychlorobiphenyl contamination), and
51 Chitinophagaceae and Geobacteraceae (with naphthalene contamination). These findings
52 provide insights into the main environmental factors shaping microbial communities of
53 mangroves in the AEP that experience low human impact and allow for the identification of
54 several potential microbial bioindicators of weak anthropogenic pressure.

55 **Significance and impact of the study:**

- 56 • The spatial variation of mangrove microbiota is studied in relation to environmental
57 factors and contaminants in the Cayenne estuary (French Guiana).
- 58 • Cayenne estuary mangroves are globally pristine, but low-level human pressures were
59 detected (OM enrichment and NOAA ERL thresholds exceeded for naphthalene and
60 dieldrin).
- 61 • pH, dieldrin, TC, and granulometry are the main drivers influencing the profile of the
62 microbial community.
- 63 • Bathyarchaeota, “*Candidatus Nitrosopumilus*”, and *Nitrospira* were identified as
64 potential indicators of OM enrichment in mangroves.
- 65 • Polluted mangroves were enriched in Deltaproteobacteria, Acidobacteria,
66 Chitinophagaceae, and Geobacteraceae.

67

68 **1. Introduction**

69 Tropical and subtropical intertidal zones are occupied by iconic mangrove forests
70 ecosystems, extending approximately 137,000 km² along the shores of 123 countries (Bunting
71 et al., 2018). Within latitudes of around 32°N and 38°S, mangroves harbor significant
72 biodiversity distributed in two distinct floristic realms (Spalding, 2010), the AEP and the
73 Indo–West Pacific (IWP) (Duke, 1992). These forests are assemblages of salt-tolerant trees
74 and shrubs that grow on soft sediments in places where freshwater mixes with seawater (e.g.,
75 estuarine margins and coastal marine environments). Mangroves are well adapted to dynamic
76 environments (salinity fluctuations, periodic flooding, and anoxia of sediment) and are among
77 the most productive ecosystems in tropical coastal areas (Donato et al., 2011). They carry out
78 functions of great ecological importance (e.g., carbon and nutrient recycling, provision of
79 breeding and feeding areas), and they also provide valuable ecosystem services such as
80 coastal protection (Massel et al., 1999) and climate change mitigation (Laffoley and

81 Grimsditch, 2009; Walcker et al., 2018). Despite the numerous services they provide,
82 mangroves are being degraded by anthropogenic activities and lost at an alarming rate in
83 some areas (Carugati et al., 2018).

84 The benthic microbiota plays a fundamental role in mangrove functioning because it
85 processes most of the energy flow and nutrients. It is responsible for OM degradation and
86 nutrient recycling (Alongi, 1994). Mangrove microorganism biodiversity has been the subject
87 of numerous studies, which have deepened our understanding of the microbial communities
88 inhabiting the mangrove rhizosphere (Gomes et al., 2014) and mangrove sediments (Jiang et
89 al., 2013; Mendes et al., 2012; Nogueira et al., 2015; Rocha et al., 2016; Santana et al., 2019).
90 However, most of the studies were mainly focused on mangroves in the IWP region. Most of
91 the studies conducted in the AEP region have been within the Eastern American subregion
92 and dealt with coastal mangroves in southern Brazil (Dias et al., 2010, 2011; Peixoto et al.,
93 2011; Rigonato et al., 2013) and Colombia (Torres et al., 2019). In these countries, the
94 anthropogenic pressure on mangroves is very strong in some areas due to the proximity of big
95 industrial cities and sites of offshore oil exploitation. Because microorganisms are stress-
96 sensitive, several microbial bioindicators of pollution were identified in Brazilian and
97 Colombian mangroves (Peixoto et al., 2011; Torres et al., 2019).

98 Apart from chemical contaminants (Maturro et al., 2016; Sheng et al., 2016, Zhang et
99 al 2014), the microbial structure and composition of prokaryotic communities in soils and
100 sediments are known to be shaped by several physicochemical factors such as carbon and
101 nitrogen concentration (Liu et al 2012), redox potential (DeAngelis et al 2010), pH (Aciego
102 Pietri and Brookes, 2008; Wardle, 1992), and salinity (Chambers et al 2016). Compared to the
103 IWP mangroves and the human-impacted AEP mangroves, relatively little is known about the
104 factors shaping the distribution of the microbial communities in low-human impacted AEP
105 mangroves. Within the Eastern American subregion of the AEP zone, the mangrove coastline

106 of the Guianas (between the Oyapock and Orinoco Rivers estuaries) appears to be an
107 exception, as the mangroves here experience low-level human impact (Diop et al., 2016;
108 Michelet et al., 2021). In French Guiana (FG), mangroves occupy about 80% of the coastal
109 line with a total extent of about 70,000 ha (Fromard, Vega, and Proisy 2004). Due to their
110 localization, near the Amazon's mouth (Brazil), FG mangroves are very dynamic and are
111 under the influence of one of the world's largest sediment discharge systems (Anthony et al.,
112 2010; Marchand et al., 2006).

113 In order to better understand which environmental drivers are important to explain the
114 compositional changes of mangrove microbiota in the AEP, an analysis of the microbial
115 diversity in Guiana's mangroves was carried out.

116 Based on the same sedimentary cores used by Michelet et al. (2021) and considering
117 the different sedimentary layers, we assessed the microorganism response to low human
118 pressure along the Cayenne estuary and defined the main environmental drivers shaping the
119 structure and diversity of communities.

120 Finally, an exploration of the changes in the relationship between mangrove
121 microbiota diversity and the abiotic environment, according to sediment depth and the urban-
122 to-natural gradient, allowed us to identify potential microbial bioindicators of mangrove
123 health.

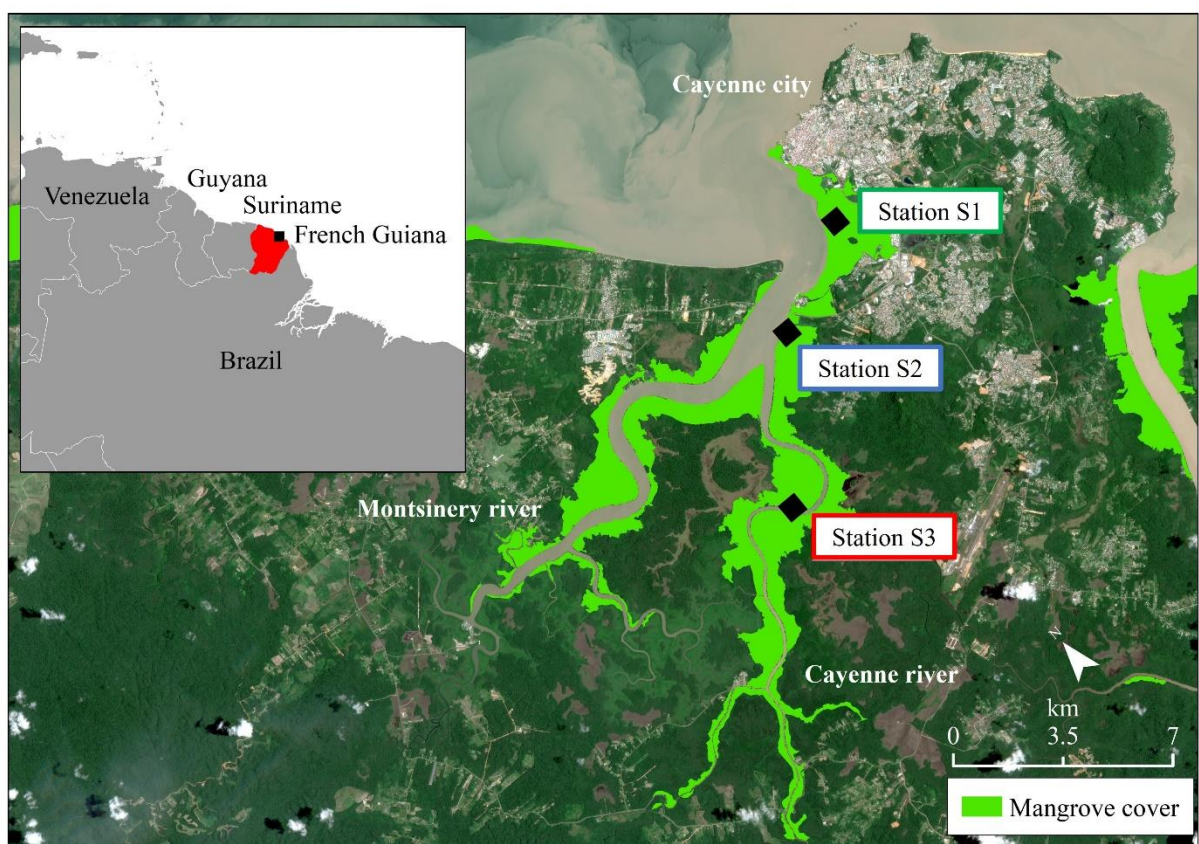
124

125 **2. Materials and Methods**

126 **2.1. Study area and sampling strategy**

127 The location of the study area was the Cayenne estuary in FG (South America; Figure 1).
128 Three stations (S1, S2, and S3) were selected on the edge of the estuary (<5 m) and at an
129 increasing distance from Cayenne city, which is the largest city and the capital of FG (61,268
130 inhabitants; INSEE 2017), along an urban-to-natural gradient. Station 1 (S1) is located at the
131 Crique Fouillée tributary creek (4°54'53.208" N, 52°20'15.9324" W; Figure 1), draining

132 urban waters and the effluents of the wastewater treatment plant of Cayenne city (urban,
133 commercial, and industrial waters). Station 2 (S2) is located near the confluence of the
134 Cayenne and Montsinery Rivers, which flow in the Cayenne estuary ($4^{\circ}53'49.2288''$ N,
135 $52^{\circ}22'27.714''$ W; Figure 1). This station receives water from the suburban Cayenne drainage
136 basin. Station 3 (S3) is located southeast of Cayenne city, 11.7 km from S1 and 4.7 km from
137 S2, along the Cayenne River ($4^{\circ}51'31.9716''$ N, $52^{\circ}23'59.5248''$ W; Figure 1) in a natural
138 area farther from human activities.



139

140 **Figure 1.** Location of mangrove sampling stations in FG (South America). S1 is located near
141 Cayenne city, S2 at the confluence of Cayenne and Montsinery Rivers, and S3 along the
142 Cayenne River. Mangrove cover is represented in green. (modified from Michelet et al.,
143 2021).

144

145 At each station, three sediment cores (A, B and C) were collected at low tide, within a 10
146 m^2 plot and located 2 m away from each other, with plexiglass tubes (internal diameter 10.4
147 cm, height 20 cm) between November 19 and 21, 2017 (spring tides, dry season). The

148 sediment cores were sliced in 0.5 cm thick layers around the two first centimeters, then 1 cm
149 thick layers until the end of the core. The obtained layers were randomly subsampled to create
150 3 pools of sediments: 0 to 2 cm, 2 to 10 cm, and 10 cm to the end of the core.

151

152 During subsampling, the pH and redox potential (Eh) were immediately measured with a
153 multiparameter probe (WTW Multi 3500i). The slices were then subsampled for
154 characterization of environmental parameters: granulometry, TC, total nitrogen (TN), trace
155 metals and metalloids (TMM), organic contaminants, and microbial analysis. Subsamples
156 were immediately frozen and stored at $-80\text{ }^{\circ}\text{C}$ (for TC, TN, TMM, organic contaminants, and
157 microbial analyses) or $4\text{ }^{\circ}\text{C}$ (for granulometry analysis).

158

159 **2.2. Treatment**

160 **2.2.1. Measurement of TC and TN and determination of granulometry**

161 Sediments were freeze-dried over 24 h, crushed to powder, and homogenized for
162 sediment analysis. TC and TN were measured by combustion at $930\text{ }^{\circ}\text{C}$ using a CHN carbon
163 analyzer (Flash-2000; Thermo Fisher Scientific Inc., Milan, Italy). In FG, the total organic
164 carbon and TC appeared to be very well correlated, due to the lack of carbonates (Marchand
165 et al., 2004). Thus, TC can be considered as a proxy for carbon organic content in the
166 sediments. Granulometry analysis was realized using a laser beam diffraction analyzer
167 (Partica LA-950V2; Horiba Instruments, Inc.).

168

169 **2.2.2. Contaminants and TMM analysis**

170 Five groups of organic contaminants—15 polycyclic aromatic hydrocarbons (PAHs),
171 some of which are frequently monitored according to recommendations by the European
172 Union and the US Environmental Protection Agency (acenaphthene, acenaphthylene,
173 anthracene, benzo[ghi]perylene, benzo[a]anthracene, benzo[b]fluoranthene,

174 benzo[b+k]fluoranthene, benzothiophene, biphenyl, chrysene, dibenzothiophene,
175 fluoranthene, fluorene, naphthalene, phenanthrene, and pyrene), 11 polychlorinated biphenyls
176 (PCB-7, PCB-28, PCB-52, PCB-101, PCB-105, PCB-118, PCB-138, PCB-153, PCB-156,
177 PCB-169, and PCB-180), 12 organochlorine pesticides (2,4-DDT, 2,4-DDD, 2,4-DDE, 4,4-
178 DDD, 4,4-DDE, 4,4-DDT, aldrin, chlordecone, dieldrin, endrin, hexachlorobenzene (HCB),
179 and isodrin), 6 phthalates (DMP, DEP, DBP, BBP, DEHA, and DEHP), and 7
180 polybromodiphenylethers (BDE 28, BDE 47, BDE 99, BDE 100, BDE 153, BDE 154, and
181 BDE 183) were extracted from the sediments and analyzed (Michelet et al., 2021). For PAHs,
182 PCB, and pesticides quantification, naphthalene d₈, biphenyl d₁₀, phenanthrene d₁₀, pyrene
183 d₁₀, chrysene d₁₂, benzo(a)pyrene d₁₂, benzo(g,h,i)perylene d₁₂ were used as standards. For the
184 plastic additives, di (2-ethylhexyl) phthalate- d₄ and BDE 77 were respectively used as
185 standards. All standards were obtained from LGC Standard (Wesel, Germany) and Interchim
186 (Montluçon, France). In order to discriminate the sources of PAHs, the ratios Flt/(Flt + Pyr)
187 and Ant/(Ant/Phe) (Flt, fluoranthene; Pyr, pyrene; Ant, anthracene; Phe, phenanthrene) were
188 calculated and compared to the published cut-off values (at > 0.5, PAHs are likely to occur
189 from biomass burning) (Brändli et al., 2007; Katsoyiannis et al., 2007).

190 Twenty-seven TMM were quantified in each sample such as lead (Pb), chromium
191 (Cr), nickel (Ni), copper (Cu), zinc (Zn), manganese (Mn), mercury (Hg), arsenic (As) and
192 molybdenum (⁹⁵Mo). Extraction and analysis protocols are presented in detail in Michelet et
193 al. (2021). To check the accuracy of the TMM measurements, the certified reference material
194 MESS-4 (National Research Council Canada) was run before starting the measurements and
195 many times per analytical batch (Supplementary Data A). The analytical recovery was within
196 10% compared to certified concentration. Freeze-dried sediment (Christ Gamma 1-16
197 LSCplus) were analyzed for particulate Hg using a CV-AAS (LECO AMA 254) equipped
198 with a low Hg optical cell. The method detection limit was 2 pg (3 times the standard

199 deviation of the blank samples). The measured values were always within $\pm 5\%$ of the
200 recommended values.

201

202 **2.2.3. Total DNA extraction and quantification**

203 Sediments were freeze-dried for 48 h and crushed to powder using a sterilized mortar
204 and pestle. Total DNA was then extracted from 0.25-0.30 g dry sediment with the DNeasy
205 PowerSoil Kit (Qiagen) according to the manufacturer's protocol. The DNA extracts were
206 quantified by fluorometric dosage with a Quantifluor dsDNA system kit (Promega) according
207 to the supplier's recommendations. The total DNA measurement (molecular microbial
208 biomass) was used as a proxy for the microbial biomass.

209

210 **2.2.4. Quantitative polymerase chain reaction (qPCR)**

211 To determine the bacterial and archaeal abundances in the sediments, 16S rRNA genes
212 (*rrs*) were quantified by qPCR. The GoTaq qPCR Master Mix (Promega) was used following
213 the supplier's recommendations, using specific archaeal (931F: 5'-
214 AGGAATTGGCGGGGAGCA-3' and m1100R: 5'-BTGGGTCTCGCTCGTTRCC-3') and
215 bacterial (300F: 5'-GCCTACGGGAGGCAGCAG-3' and univ516R: 5'-
216 GTDTTACCGCGGCKGCTGRCA-3') primer sets (for a review, see Klindworth et al.,
217 2013). The qPCR cycles for archaea consisted of initial denaturation (3 min at 98 °C),
218 followed by 35 cycles of denaturation for 10 s at 98 °C, primer hybridization for 10 s at
219 62 °C, and elongation for 20 s at 72 °C. For bacteria, the first step of initial denaturation
220 lasted 2 min at 98 °C, followed by 30 cycles of initial denaturation for 5 s at 58 °C, primer
221 hybridization for 10 s at 55 °C, and elongation for 12 s at 72 °C.

222

223 **2.2.5. 16S rRNA gene amplification and sequencing**

224 To characterize the structure and composition of the bacterial and archaeal
225 communities, hypervariable regions V4–V5 of the 16S rRNA genes were amplified by PCR
226 using the 515f (5'-TGT GYC AGC MCG CGC GGT A-3') and 928r (5'-CCG YCA ATT
227 CMT TTR AGT-3') primer sets (Parada et al., 2016). Each 25 μ L reaction mix contained 2
228 μ L of DNA (\sim 0.5–5 $\text{ng } \mu\text{L}^{-1}$), 10 μM of 515f forward primer, 10 μM of 928r reverse primer,
229 10 mM of dNTPs, 0.4 μL of Pfu DNA polymerase (Promega), and 2.5 μL of Pfu buffer
230 (Promega). The PCR cycles consisted of initial denaturing for 2 min at 94 $^{\circ}\text{C}$, followed by 35
231 cycles composed of denaturation for 20 s at 94 $^{\circ}\text{C}$, primer hybridization for 20 s at 50 $^{\circ}\text{C}$, and
232 elongation for 25 s at 72 $^{\circ}\text{C}$ in a T100 thermal cycler (Bio-Rad). PCR products were verified
233 by agarose gel electrophoresis (1.5% w/v). Amplicon purification was performed using the
234 Agencourt AMPure XP system, and quantification, with QuantIT PicoGreen. Primers
235 containing the Illumina adapters flanked by floating tails complementing those on amplicons
236 and 5 μL of purified amplicons were added for the second round of amplification. The PCR
237 cycling started at 94 $^{\circ}\text{C}$ for 2 min, followed by 12 cycles of amplification (94 $^{\circ}\text{C}$ for 1 min, 55
238 $^{\circ}\text{C}$ for 1 min, 68 $^{\circ}\text{C}$ for 1 min), and a final extension step at 68 $^{\circ}\text{C}$ for 10 min. The purified
239 amplicons were sequenced on an Illumina MiSeq platform (Genotoul, Toulouse, France).

240

241 **2.2.6. Sequence analysis**

242 The raw data were processed using the dada2 package (v.3.9) in R studio interface
243 (v3.2.3) following the workflow described by Callahan et al. (2016). The raw sequence
244 datasets are available in the National Center for Biotechnology Information database under
245 the PRJNA735070 BioProject. For taxonomic assignment, amplicon sequence variants
246 (ASVs) were compared with the Silva database (Silva v_132) (Quast et al., 2013). All non-
247 assigned reads at the phylum level and reads belonging to chloroplasts and mitochondria were
248 removed from the dataset. To compare the diversity index (specific richness and Shannon

249 index), the samples were rarefied at an even number of sequences (4180 reads) with the
250 phyloseq package (v3.9) (McMurdie and Holmes, 2013).

251

252 **2.2.7. Statistical analysis**

253 Statistical analyses were performed with R studio software (v3.2.3). As described by
254 Michelet et al. (2021), a multiple factor analysis (MFA) (Escofier and Pagès, 1994)
255 constituted by four variable groups (physicochemical parameters, OM, organic contaminants,
256 and TMM) was performed using FactoMineR (Lê et al., 2008) and factoextra (Kassambara
257 and Mundt, 2020) packages to compare microbiota and environmental parameters in the three
258 stations and depths (statistical individuals). Only variables with a large contribution to one of
259 the axes ($\cos^2 > 0.5$) were plotted (Figure 2). Differences in environmental parameters and
260 relative abundance of each taxonomic group were tested and displayed by the Scheirer–Ray–
261 Hare test and Wilcoxon pairwise test in the R companion package (Mangiafico, 2021). A
262 forward selection and a permutation test (vegan package) (Oksanen et al., 2020) were
263 performed on the noncollinear variables (lattice package) (Sarkar, 2008) to determine the
264 environmental drivers of the microbial community composition. This selection was followed
265 by permutational multivariate analysis of variance (PERMANOVA) in the vegan package in
266 order to test the differences in prokaryotic composition between samples. The top 30 phyla Z-
267 scores were calculated in the ComplexHeatmap package (Gu et al., 2016), and these analyses
268 were completed with pairwise Spearman correlation (r_s) to determine the correlations between
269 taxa and environmental factors.

270

271 **3. Results and discussion**

272 **3.1. Sampling stations characteristics and specificities**

273 The physicochemical parameters (pH, Eh, granulometry), organic matter characteristics
274 (TC, TN, TC:TN), TMM levels, and organic contaminant concentrations of sediments from

275 three sampling sites (S1, S2, and S3) and three depths (0-2, 2-10, and > 10 cm) are recorded
276 in Table 1 and in Supplementary Tables B. In addition, multivariate analysis (MFA) was used
277 to determine the sediment characteristics and specificities (Figure 2).

278 Axis 1 shows a separation between stations, mainly explained by variations in
279 granulometry, TC, TN, TC:TN, and TMM concentrations (Figure 2B). All sediments were
280 dominated by the silt fraction (74-89%), but a higher sand value is shown for S1 (18-21%;
281 $p > 0.05$), situated closer to the ocean (Figure 2B, Supplementary Data B). At this station,
282 higher TC (2-6%) was also measured ($p < 0.001$; Figure 2B; Supplementary Data B), while
283 the TC levels in the sediment at S2 and S3 (1-2%) were lower and similar to those of nearby
284 mangroves (Marchand et al., 2006). A slight enrichment in nitrogen was also observed in the
285 surficial sediment at S1 (TN ~0.3%; Figure 2B, Supplementary Data B). The slight OM
286 enrichment observed was likely due to the proximity with Cayenne city (urban runoff) and its
287 wastewater treatment plant. Higher TC:TN ratios were also observed at S1 at all depths (15.1
288 to 17.9; $p < 0.001$; Figure 2B, Supplementary Data B). This ratio is usually used as a proxy to
289 determine the source and fate of OM in water (e.g., Gordon and Goni, 2003; Ramanathan et
290 al., 2008; Wu et al., 2007). The measured values in S1 are in line with TC:TN ratio generally
291 observed for terrigenous OM (e.g., mangrove tree leaves and roots), which is known to be
292 poorly degradable by microorganisms (Marchand et al., 2003). The OM characteristics (TN,
293 TC, and TC:TN ratio) were very similar at S2 (10.3 ± 1.2) and S3 (9.5 ± 0.4). S1 was also
294 characterized by 1.2 times higher Hg concentration than the two other stations ($p < 0.01$;
295 Table 1, Figure 2B). A positive correlation was found between Hg and TC ($r_s = 0.76$; $p <$
296 0.001). Previous studies have shown that OM content probably promotes Hg storage in
297 mangrove sediments (Marchand et al., 2006; Lei et al., 2019).

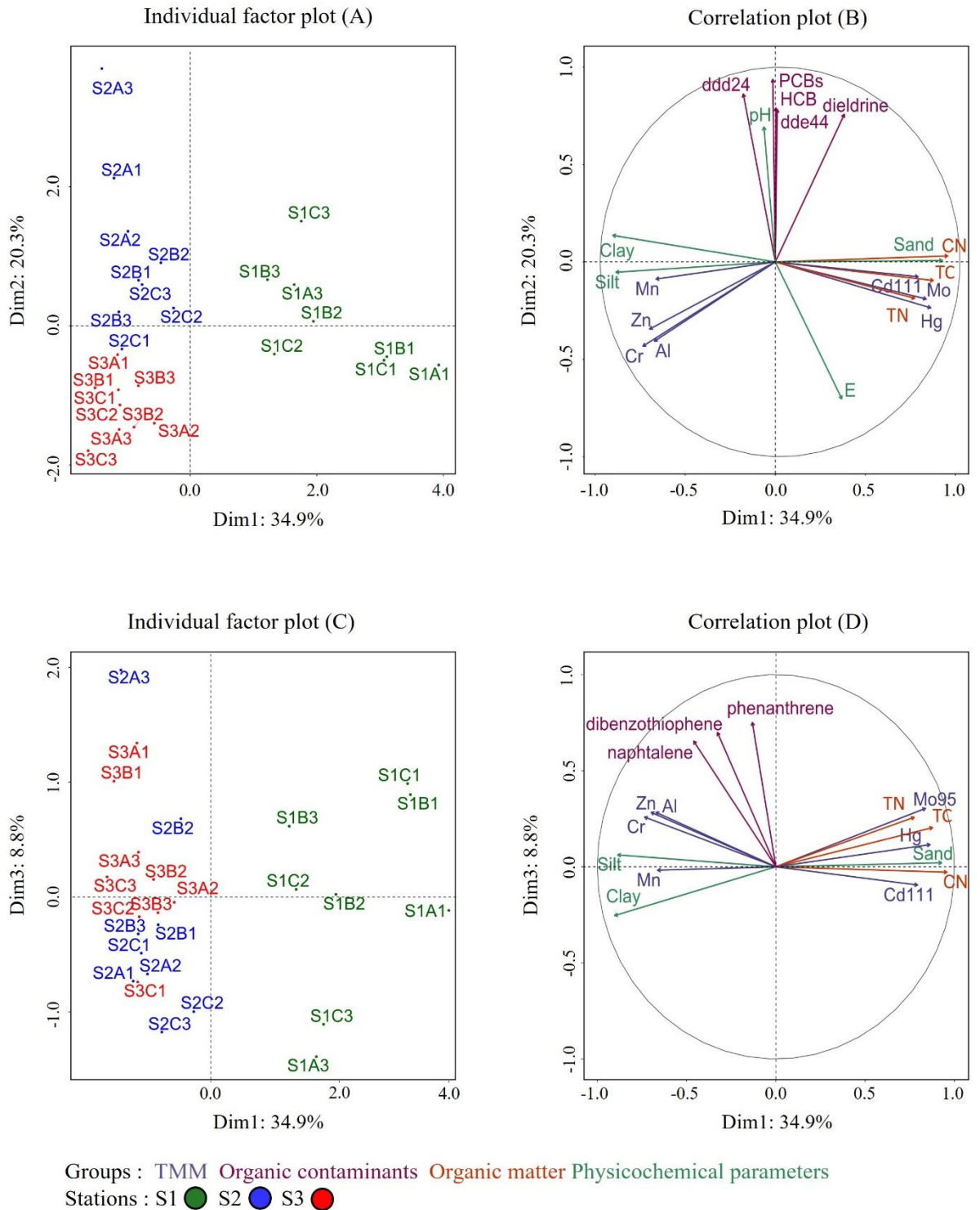
298 **Table 1.** Levels of organic contaminants and metals in sediments at three depths (0-2, 2-10, > 10 cm) at three sampling stations (mean \pm SD, n =
 299 3). Bold numbers correspond to values superior to OSPAR ERL or NOAA threshold.
 300

	Station 1			Station 2			Station 3		
Depth	0-2 cm	2-10 cm	>10 cm	0-2 cm	2-10 cm	>10 cm	0-2 cm	2-10 cm	>10 cm
	Organic contaminants concentrations (ng g⁻¹ d.w.)								
ddd24	0.1 \pm 0.1	0	0.3 \pm 0.3	0.3 \pm 0.5	0.2 \pm 0.1	0.5 \pm 0.5	0	0	0
dde44	0.1 \pm 0.1	0	0.3 \pm 0.3	0.1 \pm 0.2	0.2 \pm 0.2	0.2 \pm 0.3	0	0	0
DBT	0	2.8 \pm 3.9	2.4 \pm 4.2	4.4 \pm 7.6	2.4 \pm 4.1	10.1 \pm 17.4	11.2 \pm 12.6	1.4 \pm 2.4	6.4 \pm 1.6
Dieldrin	1.6 \pm 0.3	1.7 \pm 0.3	1.8 \pm 0.3	1.9 \pm 0.2	1.9 \pm 0.3	2.0 \pm 0.1	0	0	0
HCB	1.1 \pm 1.0	0.7 \pm 0.1	1.4 \pm 0.9	1.9 \pm 2.2	0.6 \pm 0.4	2.8 \pm 2.6	0	0	0
Naphthalene	0	0	0	0	28.1 \pm 48.6	227.5 \pm 394.1	274.1 \pm 191.6	110.5 \pm 33.6	232.2 \pm 33.4
PCBs	1.2 \pm 0.5	1.1 \pm 0.1	2.2 \pm 0.9	1.4 \pm 4.3	3.2 \pm 0.9	3.4 \pm 3.0	0.1 \pm 0.1	0	0
Phenanthrene	8.5 \pm 1.2	12.5 \pm 5.0	6.7 \pm 6.9	7.3 \pm 4.8	8.1 \pm 5.1	14.1 \pm 13.5	17.0 \pm 18.9	6.0 \pm 1.6	9.3 \pm 1.3
	TMM concentrations (μg g⁻¹ d.w.)								
Al (mg g ⁻¹)	109.3 \pm 5.8	113.2 \pm 1.5	107.8 \pm 5.0	115.8 \pm 3.7	115.0 \pm 3.7	114.7 \pm 6.0	118.3 \pm 1.5	117.9 \pm 0.5	116.9 \pm 6.0
¹¹¹ Cd	0.09 \pm 0.02	0.08 \pm 0.01	0.08 \pm 0	0.07 \pm 0	0.07 \pm 0.01	0.07 \pm 0	0.07 \pm 0	0.06 \pm 0	0.07 \pm 0
Cr	78.1 \pm 5.0	82.5 \pm 3.5	77.2 \pm 4.5	84.4 \pm 3.6	84.3 \pm 2.3	84.9 \pm 4.4	87.5 \pm 1.4	87.7 \pm 0.5	87.2 \pm 4.5
Hg (ng g ⁻¹)	64.2 \pm 5.0	56.3 \pm 2.3	57.5 \pm 4.4	44.7 \pm 0.8	48.4 \pm 1.2	47.9 \pm 0.3	48.8 \pm 1.3	53.2 \pm 0.6	50.9 \pm 3.4
Mn	436.7 \pm 44.0	434.4 \pm 225.6	424.3 \pm 68.4	1328.4 \pm 404.2	696.7 \pm 291.3	560.0 \pm 80.0	1136.6 \pm 100.5	999.2 \pm 227.2	840.0 \pm 224.3
⁹⁵ Mo	2.6 \pm 0.2	1.8 \pm 0.1	1.9 \pm 0.6	1.2 \pm 0.2	1.5 \pm 0.3	1.2 \pm 0.1	1.3 \pm 0.1	1.6 \pm 0.1	1.4 \pm 0.1
Zn	131.7 \pm 13.3	149.20 \pm 4.18	134.5 \pm 14.2	149.4 \pm 5.2	147.8 \pm 6.8	146.6 \pm 8.0	160.2 \pm 4.2	154.0 \pm 2.6	152.4 \pm 6.8

301 The Hg concentrations measured in the Cayenne estuary correspond to natural background
302 levels observed in mangrove estuarine sediments in FG (Marchand et al., 2006) and were
303 below the NOAA ERL sediment quality guidelines (low effects range) (Burton, 2002). The
304 concentrations of ^{95}Mo ($2.6\text{-}1.8\ \mu\text{g g}^{-1}$) and ^{111}Cd ($0.07\text{-}0.08\ \mu\text{g g}^{-1}$) were also higher at S1 (p
305 < 0.001 ; Table 1, Figure 2B). On the contrary, Cr, Zn, Al, and Mn concentrations were lower
306 at S1 ($p < 0.01$). At S2 and S3, the Cr content ($84.5\ \mu\text{g g}^{-1}$) was slightly higher than the
307 NOAA ERL values ($80\ \mu\text{g g}^{-1}$) (Burton, 2002) and higher than the range already observed in
308 FG (Marchand et al., 2006). At S3, the Zn concentration ($155.5\ \mu\text{g g}^{-1}$) in the sediments was
309 also slightly higher than the ERL value ($150\ \mu\text{g g}^{-1}$). As concluded by Marchand et al. (2006),
310 even if the origin of metals is difficult to clearly define, metals in FG mangrove sediments
311 most probably result from diagenetic processes rather than anthropogenic inputs. Observed
312 differences between stations are probably related to the differences in granulometry (e.g., sand
313 content) and TC contents.

314 The second axis of the MFA discriminated the samples based on pH, redox potential, and
315 organic contaminant concentration (Figure 2B). The pH of sediment pore water was 6.2 on
316 average (min 5.7, max 6.5), but clear differences in pH profiles were observed between
317 stations. Notably, pH gradients existed within the sedimentary columns at S1 and S3, where
318 increases and decreases greater than 0.5 pH units were observed, respectively. The sediments
319 were dominated by aerobic redox conditions in S1 and S3 ($E_h \sim 100\text{-}200\ \text{mV}$) and were more
320 reduced in S2 ($E_h \sim -100\ \text{mV}$) (Figure 2B, Supplementary Data B). Significant differences in
321 pesticide, PCB, and PAH concentrations were also observed between stations ($p < 0.05$;
322 Figures 2B, 2D). Although organochlorine pesticides (HCB, ddd24, dieldrin) and PCBs were
323 detected at S1, the highest concentrations were observed at S2 (Table 1, Figure 2B). These
324 compounds are among the 12 most persistent organic pollutants (POPs) according to the

325 Stockholm Convention on POPs (2001) and are also listed as priority chemicals in the EU
 326 Water Framework Directive 2013/39/EC (European Union, 2013).



327

328 **Figure 2.** Multiple factor analysis of data obtained from different stations with four groups of
 329 variables ($\cos^2 > 0.5$): physicochemical parameters (green), OM (orange), organic
 330 contaminants (purple), and TMM (blue). Graphs represent (A,C) individual factor plots and

331 (B,D) correlation plots on axes 1 and 2 (A,B) and 1 and 3 (C,D). Dots correspond to 26
332 samples from three sampling stations: S1 (green), S2 (blue), S3 (red). A, B, C indicate
333 replicates; 1: 0-2 cm; 2: 2-10 cm; 3: > 10 cm.
334

335 Among these compounds, dieldrin was the only one to show values close to or slightly
336 higher than the ERL (2 ng g^{-1}) for about half of the samples at S2 (Table 1). This pesticide
337 was used extensively in agriculture as an alternative to DDT until its use was forbidden by the
338 French government in 1972. Measured HCB concentrations (up to 5.7 ng g^{-1}) were under the
339 no-effect concentration value of 20 ng g^{-1} for chronic exposure of benthic organisms (OSPAR
340 ERL value) (Long et al. 1995). However, the quality of the sediment could be poor (class 3)
341 according to the environmental quality classification of sediment in Norway (Bakke et al.,
342 2010). Following this classification, considering the sum of DDT, the sediments would be
343 considered as good (class 2) or even, for some samples, as background (class 1) (Long et al.,
344 1995). The maximum value of the sum of PCBs (6.8 ng g^{-1} ; Table 1) was far from the
345 sediment ERL guideline value of 23 ng g^{-1} (Long et al., 1995). The maximum value of the
346 sum of PCBs (6.8 ng g^{-1} ; Table 1) was far from the sediment ERL guideline value of 23 ng
347 g^{-1} (Long et al., 1995). By contrast, compared to the other two stations, S3 was characterized
348 by elevated concentrations of naphthalene (96.4 to 468.9 ng g^{-1} ; Table 1), one of the 16 PAHs
349 considered to be priority pollutants by the EPA (Bojes and Pope, 2007). According to the
350 values of the Flt/(Flt + Pyr) ratio, usually used to discriminate the sources of PAHs,
351 hydrocarbons detected in Cayenne River sediments are from diesel and wood combustion
352 (Pereira et al., 2019; Pichler et al., 2021). When present in the sediments, naphthalene levels
353 were always higher than the NOAA threshold effect level (34.57 ng g^{-1}), and for some
354 samples, were above the NOAA ERL value (160 ng g^{-1}) (MacDonald et al., 2000).

355 The study of Michelet et al. (2021) on the same cores showed low averaged
356 concentrations of the sum of the different organic contaminants measured (PAHs,
357 organochlorine pesticides) in the whole sedimentary cores that were under the regulatory

358 thresholds. By considering the mean individual concentrations of compounds at each depth
359 layers (0-2 cm; 2-10 cm; > 10 cm), it clearly appears however that some compounds were
360 above the regulatory thresholds in some layers at stations 2 et 3 (i.e. dieldrin and naphthalene,
361 respectively).

362

363 **3.2. Prokaryotic community of mangrove sediments of Cayenne estuary**

364 **3.2.1. Estimations of community size**

365 The total DNA extracted from the sediments (the molecular microbial biomass) was
366 used to estimate the size of the microbial pool at the sampling stations (Dequiedt et al., 2011;
367 Terrat et al., 2012). This proxy includes the prokaryotic communities and micro eukaryotic
368 inhabitants of mangrove sediments (e.g., fungi and meiofauna). At stations 1 and 3, molecular
369 microbial biomass was $3.1 \pm 0.4 \mu\text{g g}^{-1}$ d.w. and $3.98 \pm 0.52 \text{ g}^{-1}$ d.w., respectively
370 (Supplementary Data C). Higher values were observed in the surface layer at station 2 ($p =$
371 0.05 ; $7.3 \pm 0.8 \mu\text{g g}^{-1}$ d.w). These molecular microbial biomass values correspond well to
372 values reported in other mangrove sediments ($\sim 11\text{-}20 \mu\text{g g}^{-1}$) (Fernandes et al., 2014; Jiang et
373 al., 2011).

374 Within the microbial communities, differences were only observed in the size of the
375 bacterial populations in the surface layer ($p < 0.001$). Indeed, the number of bacterial 16S
376 rRNA gene copies was higher at station 1 than at stations 2 and 3 ($p < 0.01$) on the surface
377 and reached 7.9×10^9 copies g^{-1} d.w. vs. 1.4×10^9 and 4.9×10^5 copies g^{-1} d.w., respectively
378 (Supplementary Data C). The environmental conditions at station 1 and, notably, the higher
379 OM content (1.5- to 4-fold more than other stations). seemed to promote bacterial growth (TC
380 and TN $r_s = 0.7$, $p < 0.01$) (Kaiser and Guggenberger, 2003; Rocha et al., 2016). By contrast,
381 the archaeal abundance was not significantly different between the three stations at any depth
382 ($p > 0.05$; 5.8×10^6 copies g^{-1} sediment d.w.; Supplementary Data C) and corresponded to
383 values earlier observed for other mangrove sediments (Li et al., 2019; Zhou et al., 2017).

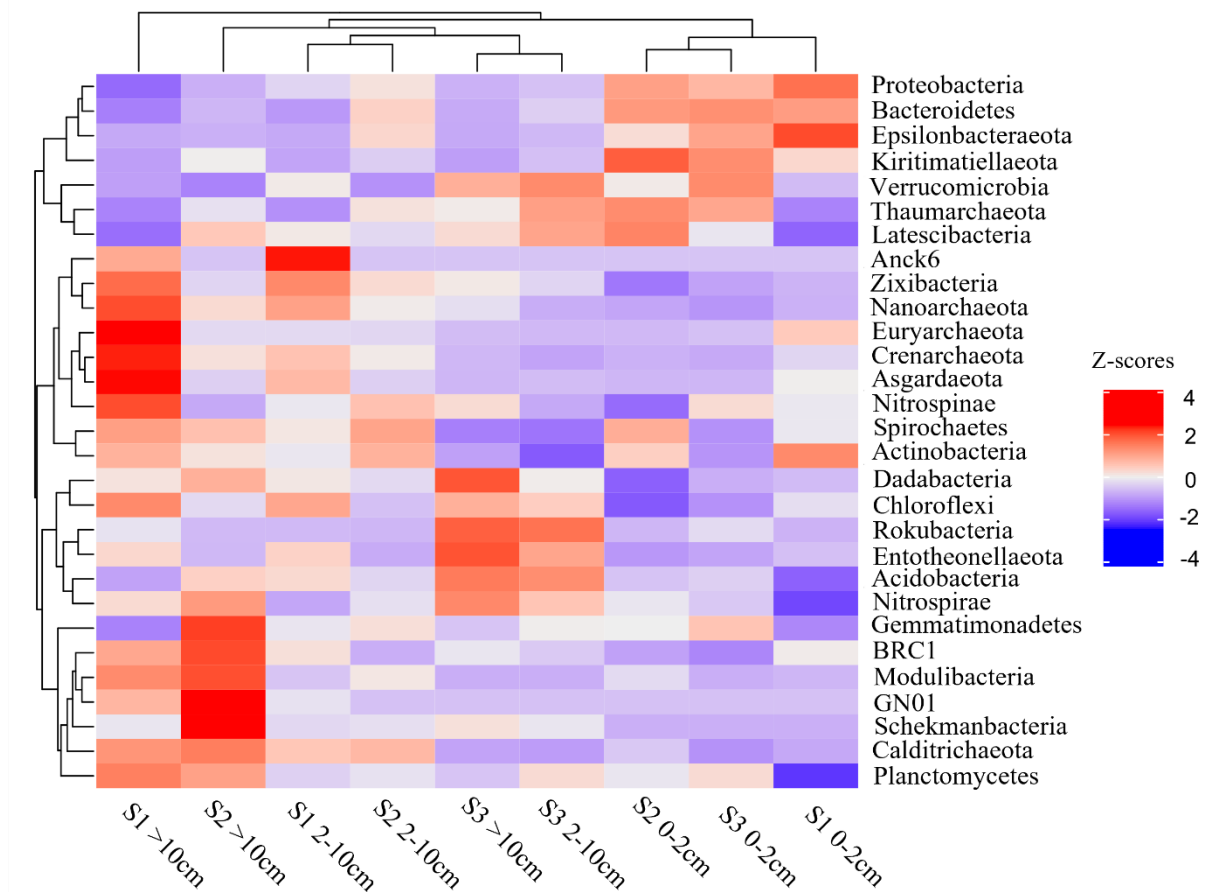
384 **3.2.2. Structure and composition of mangrove microbiota**

385 After the filtering and trimming processes, 7759 ASVs were assigned (465 ± 167
386 ASVs by sample; Supplementary Data C), and 650 ASVs that were not assigned, representing
387 0.6% of the community, were removed. There was no significant difference in alpha diversity
388 between stations ($p > 0.05$); specific richness and Shannon index fluctuated between 220 and
389 817 ASVs and 5.1 to 6.3, respectively (Supplementary Data C). By contrast to the soil
390 microbiomes in Brazilian mangroves, microbiota associated with the Cayenne estuary's
391 mangroves seems less diverse (Tavares et al., 2021).

392 The taxonomic composition of the mangrove microbiota encompassed 50 phyla
393 (Supplementary Data D). These communities were predominantly composed of members of
394 Proteobacteria (40-58%), Chloroflexi (9.0-22.3%), Acidobacteria (4.7-13.1%), Bacteroidetes
395 (3.5-13.1%), Planctomycetes (3.7-6.9%), Crenarchaeota (1.0-9.4%), Thaumarchaeota (0.7-
396 4.7%), Gemmatimonadetes (2.3-4.7%), and Nitrospirae (0.65-3.6%) (Figure 3). The observed
397 prevalence of the most abundant phyla ($> 3\%$ of relative abundance) is in accordance with the
398 literature (Andreote et al., 2012; Huergo et al., 2018; Tavares et al., 2021) suggesting that
399 Proteobacteria, Chloroflexi, Acidobacteria, Bacteroidetes, and Planctomycetes constitute the
400 core microbiome of mangrove sediments in the AEP region. Differences in community
401 composition were observed between depths and stations (PERMANOVA, $p < 0.05$). At the
402 phylum level, the heatmap clustering analysis highlighted several microbial patterns
403 (Figure 3). In the surface mangrove sediment of the three stations, the relative abundance of
404 the major phyla Proteobacteria, Bacteroidetes, Epsilonbacteraeota, and Kiritimatiellaeota was
405 higher. Bacteroidetes and Kiritimatiellaeota are regarded as specialists in the degradation of
406 macromolecules, such as complex polysaccharides and proteins, to obtain carbon, amino
407 acids, and sulfur (McBride, 2014; Sackett et al., 2019). Both phyla could have a pivotal role in
408 initiating the mineralization of high-molecular-weight organic matter in mangrove sediments.
409 Within the Proteobacteria, Desulfobacteraceae (1.1-6.4%), Desulfobulbaceae (1.8-4.4%), and

410 Desulfarculaceae (0.4-2.2%) were dominant (Supplementary Data E). These three families are
411 strictly anaerobic and chemoorganoheterotrophic, and they belong to the group of sulfate-
412 reducing bacteria (Kuever, 2014). They are commonly observed in mangroves (Huergo et al.,
413 2018; Gong et al., 2019). Mangrove sediments contain high levels of OM and sulfates, and
414 frequently experience anaerobic conditions (Correia and Guimarães, 2017), creating favorable
415 conditions for sulfate reduction (Gros et al., 2018; Fernández-Cadena et al., 2020). Within the
416 Epsilonbacteraeota, sulfur-oxidizing bacteria *Sulfurimonas* (Thiovulaceae) and *Sulfurovum*
417 (*Sulfurovaceae*) were the most abundant. These aerobic bacteria oxidize the sulfide produced
418 by anaerobic sulfate-reducing bacteria, suggesting transitions between oxic and anoxic
419 conditions in the surface sediment, probably related to tidal cycles and benthic infaunal
420 activities. Changes of the prokaryotic communities were observed between the surficial
421 sediment and the other sedimentary layers. Amongst the most abundant phyla,
422 Planctomycetes and Calditrichaeota were found to be enriched in deeper sediments at S1 and
423 S2 (Figure 3 and Supplementary Data D). Moreover, a significant increase in the relative
424 abundance of Crenarchaeota and Gemmatimonadetes was observed at S1 and S2,
425 respectively. Planctomycetes phylum comprises members related to Planctomycetia and
426 Phycisphaerae, but surprisingly no known anaerobic ammonia-oxidizing microorganisms
427 belonging to “*Candidatus* Brocardiales” were observed. Calditrichaeota was mostly
428 represented by the genus *Calorithrix*, but the genus *Caldithrix* is specifically observed in
429 deeper sediments at S1 and S2. The cultured Calditrichaeota are anaerobic,
430 chemoorganoheterotroph, and are able to degrade detrital proteins through the use of
431 extracellular peptidases (Marshall et al., 2017). Members of the phylum Gemmatimonadetes
432 are characteristically detected in the soil and mangrove microbiomes (Takayasu et al., 2019;
433 Zhang et al., 2019). Despite their widespread distribution, their physiology and ecology are
434 poorly described (Zeng, et al., 2021). Finally, a strong vertical stratification was observed for

435 Crenarchaeota at S1 with higher relative abundances in the deeper sediments than in surficial
 436 sediments. Identified members of Crenarchaeota belong exclusively to Bathyarchaeota, a
 437 taxon of global generalists that are widespread in anoxic sediments (Zhou et al., 2018).
 438 Sediment depth is known to be one of the determinants in the structuration of the archaeal
 439 communities in mangrove sediment, principally due to the biogeochemical zonation along
 440 with depth profile (Zhou et al., 2017). Crenarchaeota and Lokiarchaeota usually dominate the
 441 archaeal communities in subsurface sediments, while Thaumarchaeota and Euryarchaeota
 442 dominate in surface sediments.



443

444 **Figure 3.** Z-score heatmap of top 30 phyla (representing 94.3% of total community) at three
 445 sampling stations (S1, S2, S3). Red indicates higher Z-scores (higher relative abundance
 446 compared to mean of abundance of that phylum) and blue indicates lower Z-scores (lower
 447 relative abundance).

448

449 A different trend in the vertical stratification of the microbial communities was
450 observed at S3. In this station, Nitrospirae, Acidobacteria, and Chloroflexi were found to be
451 enriched in the subsurface layers. Within the phylum Nitrospirae, *Nitrospira* and
452 *Thermodesulfovibrio* were particularly abundant at S3. *Nitrospira* plays a pivotal role in the
453 second step of nitrification as nitrite-oxidizing bacteria (NOB) in diverse natural ecosystems
454 (Daebeler et al., 2014) or in wastewater treatment plants (Daims et al., 2011). They are often
455 associated with ammonia oxidizing archaea (AOA) for the two-step process of nitrification.
456 *Nitrospira* members seem also to be able to achieve complete ammonia oxidation
457 (comammox; Daims et al., 2015). This genus is adapted to oligotrophic conditions (Daims
458 and Wagner, 2018; Koch et al., 2019) but little is known about potential factors driving niche
459 specialization between comammox and canonical ammonia oxidizers. *Thermodesulfovibrio*
460 representatives occur in anaerobic environments, where they contribute to the degradation of
461 organic compounds and indirectly to the production of methane (Daims, 2014). Among the
462 less abundant phyla, Latescibacteria (family Latescibacteriaceae), Enttheonellaeota (family
463 Enttheonellaceae) Dadabacteria (order Dadabacteriales), Rokubacteria (order Rokubectiales)
464 were also more abundant in the deepest layer at S3 (Figure 3; Supplementary Data D).

465 Some studies have shown that factors such as pH, TC, TN, and granulometry could
466 modulate the observed microbial patterns in mangrove sediments (Colares and Melo, 2013;
467 Zhang et al., 2019; Li et al., 2021). The complex distributions that can be observed are
468 notably the results of the microbial lifestyles (*e.g.*, heterotroph or oligotroph, anaerobe or
469 aerobe) with respect to the local environmental conditions.

470

471 **3.2.3. Drivers of mangrove microbiota in the Cayenne estuary**

472 Differences in microbial community composition among the sediment samples were
473 found to have significant correlations with four environmental factors (granulometry, dieldrin,

474 pH, and TC), together explaining $R^2_{adj} = 31.2\%$: sand percentage ($p < 0.01$, $R^2_{adj} = 0.12$),
475 dieldrin ($p < 0.01$, $R^2_{adj} = 0.11$), pH ($p < 0.01$, $R^2_{adj} = 0.04$), and TC ($p < 0.01$, $R^2_{adj} =$
476 0.04).

477 Several studies reported the effects of clay, sand, and silt contents on the microbial
478 communities in soils (e.g., Dequiedt et al., 2011) and mangrove sediments (Colares and Melo,
479 2013). Sessitsch et al. (2001) studied the effect of soil particle size fractions on microbial
480 population structures. These authors demonstrated that the microbial structure was
481 significantly affected by particle size, with higher diversity observed for microbiota inhabiting
482 soil composed of smaller size fractions. The authors hypothesized that the low nutrient
483 availability, protozoan grazing, and competition with microbes (i.e., fungi) could explain the
484 reduced diversity in larger size fractions. Colares and Melo (2003) studied the relationship
485 between microbial community structure and environmental factors in Brazilian mangrove
486 sediments. Their redundancy analysis revealed that the silt–clay percentage was the most
487 important factor controlling the microbial profiles. Particle size fractions (sand, silt, and clay)
488 differ in mineralogical composition (Acosta et al., 2011). They have specific surface reactivity
489 yielding the formation of organomineral complexes with different concentration, composition,
490 and availability of associated OM (Christensen, 2001). As a consequence, these fractions
491 represent various microenvironments in terms of organic substrates, but also oxygenation,
492 accessible water, and nutrients, promoting the development of specific communities.

493 An unexpected outcome in this study was the identification of dieldrin as one of the
494 main drivers of the distribution of microbial communities in the Cayenne estuary. To date,
495 studies showing a shift in microbial communities as a result of organochlorine pesticides are
496 scarce (Sangwan et al., 2012; Tejada et al., 2015; Wu et al., 2018). The effect of dieldrin on
497 microbial communities was observed in the gastrointestinal system of zebrafish (Hua et al.,

498 2020), with an increased abundance of members belonging to Verrucomicrobia and a decrease
499 in Clostridia and Betaproteobacteria. Organochlorines can also induce shifts in microbial
500 functions in soils, such as through suppression of nitrogen-fixing bacteria that have a pivotal
501 role in replenishing natural nitrogen fertilizer in soil (Poltera, 2007).

502 In biogeography studies, pH was recognized as another factor determining the
503 microbiota structure of soils in terms of richness, diversity, and composition (Fierer and
504 Jackson, 2007; Lauber et al., 2009; Dequiedt et al 2011). Chen et al. (2016) and Li et al.
505 (2021) showed that in mangroves, pH is also one of the main factors explaining the microbial
506 community composition in sediments regardless of the season or developmental stage of the
507 mangrove forest. pH was also shown to be the best predictor of community structure and the
508 relative abundance of major dominant lineages at the continental scale in North and South
509 America (Lauber et al., 2009). Most microorganisms have a low tolerance to pH variations
510 that affect biogeochemical processes and their activities and related biogeochemical processes
511 (Nicol et al., 2008).

512 Other studies have shown that mangrove microbiota composition responds strongly to
513 other physicochemical properties of the sediment, such as TC content (Colares and Melo,
514 2003; Zhang et al 2019; Li et al., 2021). Colares and Melo (2003) highlighted a significant
515 correlation between microbial community structure and OM content in Brazilian mangroves.
516 Zhang et al. (2019) characterized the microbiota from six Chinese mangroves and showed that
517 mean annual precipitation and TC were the main factors explaining the distribution of the
518 community. Tidal cycles (Zhang et al., 2018) and variables associated with the vegetation
519 above (e.g., Jiang, et al., 2013; Gomes et al., 2014, Luis et al., 2019) have also been reported
520 to be drivers of the distribution and structure of bacterial communities of mangrove soils and
521 sediments.

522

523 **3.3. Potential microbial bioindicators of anthropogenic pressures**

524 Potential microbial bioindicators of anthropogenic pressure were found based on the
525 correlations between specific taxa and the variables associated with the three types of
526 anthropogenic pressure identified along the urban-to-rural gradient of the Cayenne estuary
527 (OM enrichment and organochlorine and naphthalene contamination).

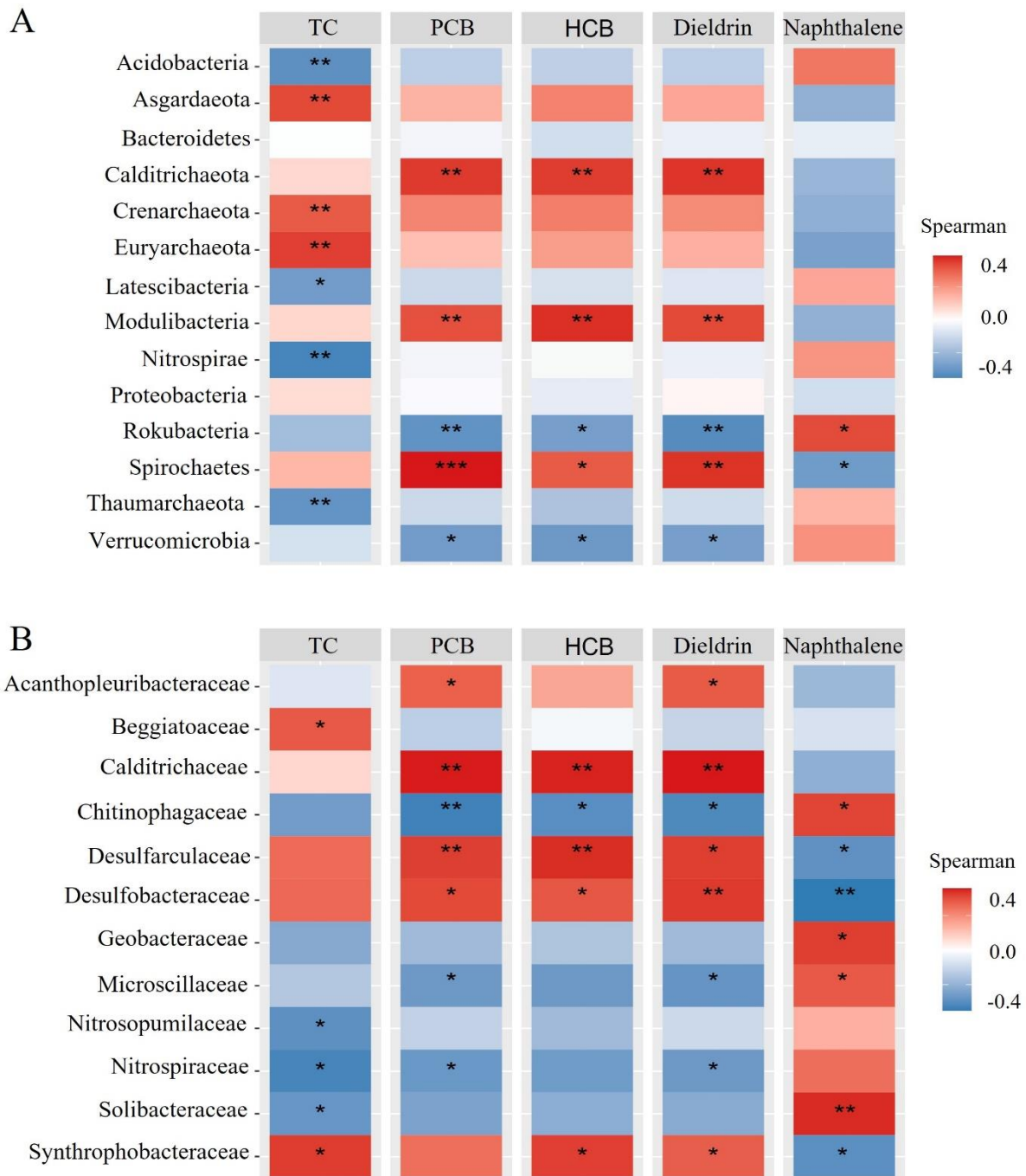
529 **3.3.1. Microbial signature of slight OM enrichment in mangrove**

530 Several microbial taxa exhibited a negative or positive correlation with OM
531 enrichment (TC), making them serious candidates for bioindication purposes (Figure 4).

532 This is the case of the Bathyarchaeota class, formerly known as the Miscellaneous
533 Crenarchaeotal Group (MCG), (0.93-8.1%), which was positively correlated with TC ($r_s = 0.6$,
534 $p < 0.01$; Figure 4A, Supplementary Data F). In mangrove sediments, Bathyarchaeota is the
535 dominant archaeal taxon (Zhou et al., 2017, 2018), its abundance was suggested to be mainly
536 correlated with total organic carbon content (Xiang et al., 2017; Yu et al., 2017). This
537 observation may be associated with their heterotrophic lifestyle and their ability to
538 anaerobically use detrital proteins, fatty acids/aromatic compounds, polymeric carbohydrates,
539 and other OM (Lazar et al., 2016; Meng et al., 2014). Despite the diversity of Bathyarchaeota
540 (over 25 subgroups) (Zhou et al., 2018) and their environmental preferences, the abundance of
541 the overall population seems to be a good indicator of OM enrichment in mangrove.

542 The levels of Euryarchaeota (0-1.6%) and Asgardaeota (0-0.88%), both highly
543 abundant in anaerobic marine sediments (Hoshino et al., 2020), were also correlated with TC
544 (both $r_s = 0.7$, $p < 0.01$; Figure 4A). Almost all the ASVs in the phylum Euryarchaeota were
545 included in the Thermoplasmata sublineage, especially in the anaerobic heterotrophic MBG-D
546 archaeal group (newly named Thermoprofundales). In recent years, single-cell genomic and
547 metagenomic approaches have shown that MBG-D archaea are capable of exogenous protein
548 mineralization and acetogenesis in marine sediment (Lazar et al., 2017; Lloyd et al., 2013). In

549 mangrove sediments, these archaea appear to be capable of transporting and assimilating
 550 peptides and generating acetate and ethanol through fermentation (Zhou et al., 2019). In the
 551 phylum Asgardaeota, the majority of ASVs belong to Lokiarchaeota, which vary in their
 552 metabolic activity, and their in situ activities are still largely unknown (Yin et al., 2021).



553

554 Spearman's rank correlation coefficient heatmap showing relationships between
 555 environmental variables and interesting taxa. A: phylum level; B: family level. Statistically
 556 significant Spearman correlations are highlighted (*p < 0.05, **p < 0.01, ***p < 0.001).

557 By contrast, Nitrospirae ($r_s = -0.7$; $p < 0.01$), Acidobacteria ($r_s = -0.6$; $p < 0.01$),
558 Thaumarchaeota ($r_s = -0.6$; $p < 0.01$), and Latescibacteria ($r_s = -0.6$; $p < 0.01$) were
559 negatively correlated with TC (Figure 4A). The majority of Thaumarchaeota ASVs (38
560 ASVs) belong to “*Candidatus Nitrosopumilus*”, which are mesophilic, autotrophic, and
561 aerobic AOA. Some groups of Thaumarchaeota are recognized as major contributors of
562 ammonia oxidation (the first step in nitrification) in marine oligotrophic ecosystems (Pester et
563 al., 2011; Wang et al., 2019) and could be dominant in some ecosystems, such as in the
564 Amazon River and its plume (Pinto et al., 2020). Ammonia oxidation is known to be inhibited
565 by several parameters, notably organic carbon content (Pester et al., 2012), OM quality
566 (Strauss and Lamberti, 2000), and the TC:TN ratio (Bates et al., 2011). AOA are adapted to
567 low ammonium concentrations and are outcompeted by bacteria when concentrations become
568 higher (Nakagawa and Stahl, 2013). Sensitivity to slight OM enrichment was also observed
569 for the *Nitrospira* genus (19 ASVs; 0-0.9%; $r_s = -0.68$; $p < 0.05$), the NOB and comammox
570 bacteria. Latescibacteria (formerly WS3) represents a ubiquitous phylum found in terrestrial,
571 aquatic, and marine environments (Youssef et al., 2015). Metabolic reconstruction suggests a
572 saprophytic lifestyle for this candidate phylum, with a marked capacity for decomposition of
573 OM (proteins, lipids, and polysaccharides) predominant in plant, bacterial, fungal, crustacean,
574 and eukaryotic algal cell walls (Frag et al., 2017; Youssef et al., 2015). Finally, a negative
575 correlation between Acidobacteria abundance and TC content was observed in our dataset
576 (Figure 4A), as it was also in terrestrial ecosystems, leading to the conclusion that members of
577 this taxon may be oligotrophic bacteria (Fierer et al., 2007).

578

579 **3.3.2. Taxa sensitive to organochlorine compounds**

580 Correlations between several microbial taxa and organochlorine (dieldrin, PCB, and
581 HCB) concentrations were observed (Figure 4; Supplementary Data F).

582 Calditrichaeota (Calditrichaceae family) and Spirochaetes (*Spirochaeta* genus) were
583 significantly correlated ($r_s < 0.7$; $p < 0.05$) with PCB, HCB, and dieldrin content (Figure 4).
584 Spirochaetes are indigenous to sulfide-rich mud or sediments (Tanner et al., 2000; Leschine et
585 al., 2006) and are typically found in PCB-contaminated waters (Dong et al., 2018) or
586 contaminated sediments (Quero et al., 2015). Calditrichaeota and Spirochaeta are known to
587 be very diversified and to have a wide range of ecological niches (Leschine et al., 2006;
588 Marshall et al., 2017). Until now, these organisms have never been associated with
589 dechlorination of organochlorine compounds; however, our results suggest that
590 Calditrichaeota and *Spirochaeta* could have a role in the degradation of PCB, HCB, and
591 dieldrin, or at least could be tolerant to these compounds. Future experimental studies are
592 needed to validate this finding (e.g., biodegradation assays). “*Candidatus* Modulibacteria”
593 phylum (previously named KS3B3 or GN03) was also positively correlated with the three
594 organochlorine compounds. By contrast, “*Candidatus* Rokubacteria” (0–1.5%; previously
595 named SPAM or candidate division NC10) was negatively correlated with PCB ($r_s = -0.6$; $p <$
596 0.05), HCB ($r_s = -0.6$; $p < 0.001$), and dieldrin ($r_s = -0.7$; $p < 0.01$). This phylum is found in
597 diverse terrestrial ecosystems such as alpine soil, crop soil, or copper mine soil (Becraft et al
598 2017), and this is the first time that it is identified in a marine ecosystem. The lack of
599 knowledge on “*Candidatus* Modulibacteria” and “*Candidatus* Rokubacteria”, two phyla
600 without cultivable representatives, does not allow us to explain the significant correlations
601 with organochlorine compounds in this study. The relative abundance of the Verrucomicrobia
602 phylum was significantly higher at S3 than at S1 and S2 (0.49%; $p < 0.05$). This taxon is
603 related to the Planctomycetes–Verrucomicrobia–Chlamydia superphylum, has diverse
604 metabolism capabilities (such as methanogenesis and degradation of sulfated polysaccharides)
605 (Freitas et al., 2012; Van Vliet et al., 2019), and is typically found in mangrove sediments
606 (Liang et al., 2007; Nogueira et al., 2015). It was negatively correlated with PCB ($r_s = -0.6$; p

607 < 0.05), HCB ($r_s = -0.6$; $p < 0.05$), and dieldrin ($r_s = -0.5$; $p < 0.05$), suggesting that
608 Verrucomicrobia is sensitive to the organochlorine compounds in mangrove sediments.

609 At the family level, some taxa seem particularly interesting in their response to PCB
610 pressure, notably in the Desulfobacterota (formerly Deltaproteobacteria) phylum. Two
611 families were significantly correlated with PCB content (Figure 4B): Desulfobacteraceae ($r_s =$
612 0.6 ; $p < 0.05$) and Desulfarculaceae ($r_s = 0.6$; $p < 0.001$). These Desulfobacterota are strictly
613 anaerobic and sulfate-reducing bacteria, and are indirectly implicated in PCB dechlorination
614 by promoting organohalide respiration of PCB dechlorinating bacteria (Praveckova et al.,
615 2016; Matturro et al., 2020). The Acanthopleuribacteraceae family (Acidobacteria) is known
616 to degrade PCB and is considered very useful in remediating polluted environments
617 (Falkiewicz-Dulik et al., 2015). The significant response of Acanthopleuribacteraceae ($r_s =$
618 0.6 ; $p < 0.01$) to the low quantity of PCBs measured makes this taxon a particularly
619 interesting bioindicator of PCB contamination.

620

621 **3.3.3. Bacterial signature of naphthalene contamination in mangrove**

622 Although naphthalene content was above the regulatory threshold, it was not identified
623 as one of the main drivers of mangrove microbiota along the estuary. However, the levels of
624 some taxa were positively correlated with naphthalene content, such as uncultured
625 Rokubacteria phylum ($r_s = 0.5$, $p < 0.05$; Fig. 4A). As seen previously, Rokubacteria are not
626 well described, but their diversified genome reveals a potential implication in the nitrogen,
627 methane, and sulfur cycles (Hug et al., 2013; Becraft et al., 2017). However, a meta-analysis
628 of the rokubacterial MAGS conducted by Kroeger et al. (2018) showed that taxa members
629 have several features involved in metabolizing long and short hydrocarbons in soil via two
630 enzymes, cytochrome P450 and alkane 1-monooxygenase, respectively. Cytochrome P450
631 has been described as one of the enzymes involved in the naphthalene biodegradation
632 (Mohapatra and Prashant, 2021) which may explain the observed positive correlation. These

633 details can explain the positive correlation observed in this study. Within the Acidobacteria,
634 the Solibacteraceae family was also positively correlated with naphthalene ($r_s = 0.7$; $p < 0.01$;
635 Figure 4B). Members of this family possess some functional genes that allow them to degrade
636 branched hydrocarbons such as pristane (Wang et al., 2016), yet it has not been proven if they
637 can degrade PAHs.

638 A correlation between the Chitinophagaceae family and naphthalene was observed
639 ($r_s = 0.6$; $p < 0.05$). Strains of this family have been isolated from freshwater sediments and
640 soil contaminated with petroleum (Blanco-Enríquez et al., 2018). Members of
641 Chitinophagaceae are known to be able to degrade biopolymers (Gomes et al., 2010) and to
642 belong to microbial consortia that remove PAHs. Specifically, some members of
643 Chitinophagaceae have the capacity to degrade pyrene and benzo[a]pyrene in water (Blanco-
644 Enriquez et al., 2018), but no significant correlation was found in this study between
645 Chitinophagaceae and these two compounds, which were barely detected in the samples. No
646 direct link between Chitinophagaceae and naphthalene degradation is reported in the
647 literature. Similarly, Geobacteraceae, a sulfur-reducing and iron-reducing Desulfobacterota
648 bacteria (Kleindienst et al., 2012) were positively correlated with naphthalene ($r_s = 0.6$; $p <$
649 0.01 ; Figure 4B). Some studies have shown that this family has the capacity to degrade
650 aromatic compounds and PAHs (Coates et al., 2001; Holmes et al., 2004; Durante-Rodríguez
651 et al., 2018), and it was used in the bioremediation of organic contaminants in the soil
652 subsurface (Holmes et al., 2004).

653

654 **4. Conclusion**

655 Our study shows that estuarine mangroves of the Cayenne River are globally exposed
656 to low anthropogenic pressures. However, we detected low OM enrichment, and
657 organochlorine and pyrolytic PAH pollution derived from human activities, mainly at the
658 stations closest to the city of Cayenne (S1 and S2). The core microbial taxa are similar to the

659 one observed in Brazil mangroves, which are more impacted by human activities.
660 Granulometry, dieldrin concentration, pH, and TC content appear to drive the mangrove
661 microbiota. Taxa of interest were identified that are correlated to OM enrichment and
662 organochlorine (dieldrin, PCB and HCB) and naphthalene contamination, represent potential
663 bioindicators of the health status of the mangrove ecosystem. However, much more research
664 is needed to assess the structural, spatial, and temporal distributions of microbial communities
665 in mangroves as well as their response to growing anthropogenic pressures in an overall still
666 relatively preserved environment.

667

668 **Data Availability Statement:** The data presented in this study are openly available in
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670

671

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683

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1190

1191 **Figures legends :**

1192 **Figure 1.** Location of mangrove sampling stations in FG (South America). S1 is located near
1193 Cayenne city, S2 at the confluence of Cayenne and Montsinery Rivers, and S3 along the
1194 Cayenne River. Mangrove cover is represented in green. (modified from Michelet et al.,
1195 2021).

1196

1197 **Figure 2.** Multiple factor analysis of data obtained from different stations with four groups of
1198 variables ($\cos^2 > 0.5$): physicochemical parameters (green), OM (orange), organic
1199 contaminants (purple), and TMM (blue). Graphs represent (A,C) individual factor plots and
1200 (B,D) correlation plots on axes 1 and 2 (A,B) and 1 and 3 (C,D). Dots correspond to 26
1201 samples from three sampling stations: S1 (green), S2 (blue), S3 (red). A, B, C indicate
1202 replicates; 1: 0-2 cm; 2: 2-10 cm; 3: > 10 cm.

1203

1204 **Figure 3.** Z-score heatmap of top 30 phyla (representing 94.3% of total community) at three
1205 sampling stations (S1, S2, S3). Red indicates higher Z-scores (higher relative abundance
1206 compared to mean of abundance of that phylum) and blue indicates lower Z-scores (lower
1207 relative abundance).

1208

1209 **Figure 4.** Spearman's rank correlation coefficient heatmap showing relationships between
1210 environmental variables and interesting taxa. A: phylum level; B: family level. Statistically
1211 significant Spearman correlations are highlighted (* $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$).

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1214 **Table legend:**

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1216 **Table 1.** Levels of organic contaminants and metals in sediments at three depths (0-2, 2-10, >
1217 10 cm) at three sampling stations (mean \pm SD, n = 3). Bold numbers correspond to values
1218 superior to OSPAR ERL or NOAA threshold.

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Supplementary Data A: Concentrations of TMM in the blank ($\mu\text{g L}^{-1}$) and calculated limits of detection (LOD) and quantification (LOQ) ($\mu\text{g L}^{-1}$) for water samples.

Element	blank	LOD	LOQ
Ag	0.002	0.001	0.003
Al	0.5	0.3	1
As	0.002	0.003	0.01
Ba	0.02	0.005	0.015
Be	0.001	0.001	0.003
Bi	0.002	0.001	0.003
Cd	0.001	0.001	0.003
Co	0.001	0.002	0.006
Cr	0.005	0.003	0.01
Cs	0.001	0.001	0.003
Cu	0.02	0.01	0.03
Fe	0.02	0.02	0.06
Li	0.005	0.002	0.006
Mn	0.005	0.003	0.01
Mo	0.005	0.003	0.01
Ni	0.001	0.001	0.003
Pb	0.001	0.001	0.003
Rb	0.005	0.003	0.01
Sb	0.001	0.002	0.006
Sn	0.002	0.001	0.003
Sr	0.02	0.01	0.03
Ti	0.05	0.03	0.1
U	0.001	0.001	0.003
V	0.002	0.002	0.006
Zn	0.010	0.005	0.015

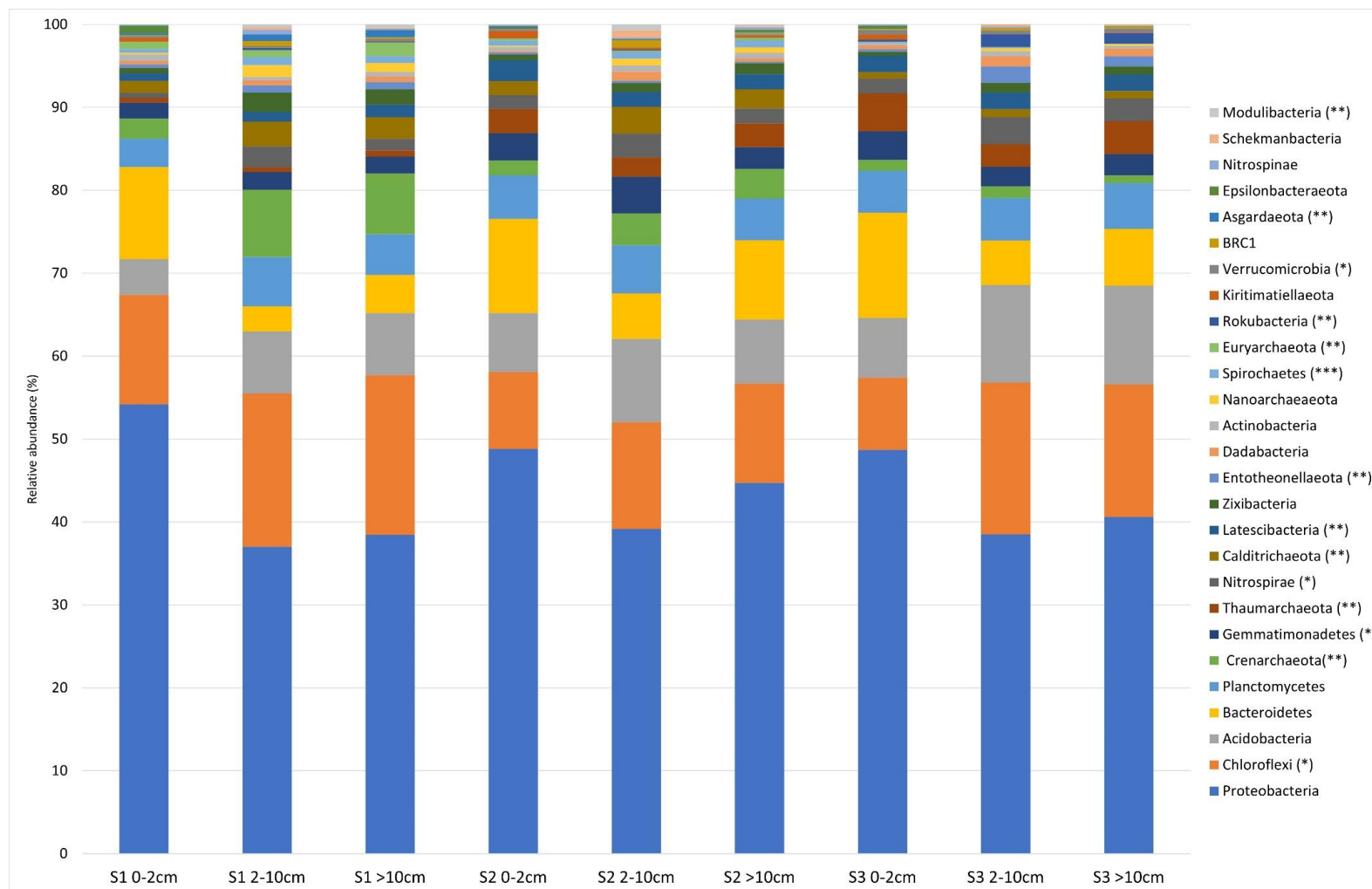
Supplementary data B: Averaged physicochemical characteristics over depth (0-2 cm; 2-10 cm; > 10 cm) from the three sampling stations (mean \pm SD, n=3).

	Station 1			Station 2			Station 3		
Depth	0-2 cm	2-10 cm	> 10 cm	0-2 cm	2-10 cm	> 10 cm	0-2 cm	2-10 cm	> 10 cm
	Physicochemical parameters								
pH	6.0 \pm 0.1	6.3 \pm 0.0	6.5 \pm 0.1	6.4 \pm 0.2	6.2 \pm 0.2	6.5 \pm 0.1	6.4 \pm 0.1	6.0 \pm 0.3	5.7 \pm 0.1
Redox potential (mV)	198 \pm 34.6	177.6 \pm 16.8	26.1 \pm 39.3	-142.5 \pm 55.5	-50.3 \pm 61.5	-77.6 \pm 24.8	165.0 \pm 15.2	152.6 \pm 11.9	164.7 \pm 14.7
	Granulometry (%)								
Clay	1.9 \pm 0.7	3.9 \pm 1.0	4.9 \pm 0.8	8.0 \pm 1.1	8.1 \pm 0.4	8.6 \pm 0.8	7.8 \pm 1.3	6.7 \pm 0.5	7.2 \pm 0.2
Silt	76.1 \pm 4.5	73.7 \pm 3.0	76.1 \pm 0.9	88.7 \pm 2.6	86.4 \pm 1.3	85.8 \pm 2.0	88.9 \pm 1.3	87.8 \pm 2.1	88.2 \pm 1.7
Sand	20.9 \pm 4.6	21.5 \pm 4.0	18.2 \pm 0.8	3.0 \pm 1.5	5.1 \pm 1.1	5.3 \pm 2.1	3.0 \pm 0.3	5.0 \pm 2.5	2.2 \pm 1.7
	Carbon TC and nitrogen TN contents								
TC (%)	5.5 \pm 1.3	3.0 \pm 0.4	2.5 \pm 0.3	1.7 \pm 0.3	1.9 \pm 0.4	1.3 \pm 0.4	1.5 \pm 0.2	1.3 \pm 0.2	1.7 \pm 0.2
TN (%)	0.3 \pm 0.1	0.2 \pm 0.0	0.2 \pm 0.01	0.2 \pm 0.15	0.2 \pm 0.03	0.1 \pm 0.01	0.2 \pm 0.01	0.2 \pm 0.01	0.1 \pm 0.01
TC:TN (mol/mol)	17.9 \pm 2.1	14.3 \pm 1.3	15.1 \pm 1.5	9.8 \pm 0.8	11.5 \pm 1.2	9.7 \pm 0.7	9.7 \pm 0.4	9.7 \pm 0.2	9.1 \pm 0.5

Supplementary data C: Values of the molecular microbial biomass, bacterial and archaeal 16S rRNA gene copies number and alpha diversity indexes (specific richness and Shannon index) at each station and for the different depth layers (mean \pm SD; n=3).

	Station 1			Station 2			Station 3		
Depth	0-2 cm	2-10 cm	> 10cm	0-2 cm	2-10 cm	> 10cm	0-2 cm	2-10 cm	> 10cm
Molecular microbial biomass ($\mu\text{g g}^{-1}$ d.w.)	3.67 \pm 0.35	2.38 \pm 1.31	2.07 \pm 0.11	7.34 \pm 0.86	2.73 \pm 1.45	2.40 \pm 0.36	3.98 \pm 0.52	3.26 \pm 0.23	2.40 \pm 0.04
	Prokaryotic abundance estimation (16S rRNA copies g^{-1} d.w.)								
Bacterial 16S rRNA genes	7.9 $\times 10^9 \pm$ 4.4 $\times 10^9$	2.3 $\times 10^9 \pm$ 1.4 $\times 10^9$	1.5 $\times 10^9 \pm$ 6.2 $\times 10^8$	1.4 $\times 10^9 \pm$ 1.3 $\times 10^8$	2.4 $\times 10^8 \pm$ 1.7 $\times 10^8$	9.4 $\times 10^7 \pm$ 4.8 $\times 10^7$	4.9 $\times 10^8 \pm$ 1.8 $\times 10^8$	4.0 $\times 10^8 \pm$ 5.6 $\times 10^7$	2.6 $\times 10^8 \pm$ 1.4 $\times 10^8$
Archaeal 16S rRNA genes	5.8 $\times 10^6 \pm$ 3.1 $\times 10^6$	4.1 $\times 10^6 \pm$ 1.6 $\times 10^6$	6.6 $\times 10^6 \pm$ 3.7 $\times 10^6$	2.5 $\times 10^7 \pm$ 1.7 $\times 10^7$	8.0 $\times 10^6 \pm$ 6.1 $\times 10^6$	2.5 $\times 10^6 \pm$ 9.6 $\times 10^5$	5.2 $\times 10^6 \pm$ 3.1 $\times 10^6$	3.1 $\times 10^6 \pm$ 1.2 $\times 10^6$	4.9 $\times 10^6 \pm$ 6.0 $\times 10^6$
	Alpha diversity index								
Specific richness	352 \pm 128	539 \pm 40	643 \pm 213	437 \pm 193	515 \pm 227	458 \pm 220	354 \pm 122	431 \pm 120	480 \pm 144
Shannon index	5.5 \pm 0.3	6.0 \pm 0.1	6.1 \pm 0.4	5.7 \pm 0.5	5.8 \pm 0.5	5.7 \pm 0.4	5.5 \pm 0.3	5.8 \pm 0.3	5.9 \pm 0.3

Supplementary data D: Histogram of the relative abundance of the top 30 phyla at each station and for the different depth layers (mean \pm SD; n =3). The p-values represent the significant differences of taxa relative abundances between station (*p = 0.05, **p= 0.01, ***p=0.001).



Supplementary data E: Relative abundances of the top 50 families at each station over depth (L1: 0-2 cm; L2; 2-10 cm; L3: > 10 cm) (mean \pm SD; n = 3).

Family	S1L1	S1L2	S1L3	S2L1	S2L2	S2L3	S3L1	S3L2	S3L3
Desulfobacteraceae	3.43 \pm 0.18	5 \pm 0.37	6.4 \pm 1.02	3.97 \pm 1.72	4.71 \pm 1.83	5.24 \pm 1.4	1.32 \pm 0.37	1.08 \pm 0.52	1.89 \pm 0.78
Syntrophobacteraceae	0.44 \pm 0.17	1.18 \pm 0.06	1.26 \pm 0.09	0.19 \pm 0.05	0.33 \pm 0.16	0.8 \pm 0.47	0.15 \pm 0.03	0.22 \pm 0.05	0.15 \pm 0.14
Anaerolineaceae	1.99 \pm 0.54	2.64 \pm 0.76	2.11 \pm 0.12	0.97 \pm 0.32	1.72 \pm 0.91	1.79 \pm 0.47	1.71 \pm 0.56	3.48 \pm 1.26	3.74 \pm 1.01
Desulfobulbaceae	3.28 \pm 0.1	1.84 \pm 0.01	2.61 \pm 1.11	3.88 \pm 0.78	4.38 \pm 0.74	2.26 \pm 1.33	2.26 \pm 0.63	2.14 \pm 0.47	2.29 \pm 0.81
Nitrosopumilaceae	0.23 \pm 0.14	0.16 \pm 0.09	0.07 \pm 0.02	2.4 \pm 0.39	1.37 \pm 1.54	1.05 \pm 1.25	2.24 \pm 0.09	2.58 \pm 0.49	1.5 \pm 0.43
Xanthobacteraceae	1.09 \pm 0.34	1.38 \pm 0.59	0.96 \pm 0.45	0 \pm 0	0.06 \pm 0.06	0.13 \pm 0.12	0.32 \pm 0.46	0.9 \pm 0.25	1.08 \pm 0.09
Rhizobiales_IS	0.4 \pm 0.1	0.78 \pm 0.04	0.28 \pm 0.23	0.03 \pm 0.02	0.14 \pm 0.08	0.04 \pm 0.03	0.28 \pm 0.08	0.47 \pm 0.21	0.56 \pm 0.17
TRA3-20	0.05 \pm 0.09	0.09 \pm 0.01	0.01 \pm 0.02	0.13 \pm 0.02	0.08 \pm 0.06	0.09 \pm 0.05	0.19 \pm 0.06	0.51 \pm 0.16	0.33 \pm 0.13
Beggiatoaceae	0.53 \pm 0.08	0.55 \pm 0.22	0.3 \pm 0.17	0 \pm 0	0 \pm 0	0 \pm 0	0.08 \pm 0.14	0.15 \pm 0.14	0.09 \pm 0.05
bacteriap25	0 \pm 0	0.09 \pm 0.13	0.13 \pm 0.07	0 \pm 0	0 \pm 0	0.14 \pm 0.15	0.06 \pm 0.09	0.48 \pm 0.26	0.89 \pm 0.29
Kiloniellaceae	0.4 \pm 0.27	0.72 \pm 0.26	0.51 \pm 0.46	0.84 \pm 0.24	0.56 \pm 0.22	0.76 \pm 0.16	0.93 \pm 0.24	1 \pm 0.11	0.88 \pm 0.04
Sva1033	0.27 \pm 0.12	0 \pm 0	0 \pm 0	0.68 \pm 0.1	0.74 \pm 0.58	0.17 \pm 0.3	0.53 \pm 0.11	0.2 \pm 0.2	0 \pm 0
Mariprofundaceae	1.55 \pm 0.73	0.37 \pm 0.01	0.4 \pm 0.27	0.11 \pm 0.09	0.32 \pm 0.3	0.28 \pm 0.32	0 \pm 0	0 \pm 0	0.17 \pm 0.27
Desulfarculaceae	0.64 \pm 0.06	1.36 \pm 0.05	2.21 \pm 0.54	0.57 \pm 0.07	0.74 \pm 0.31	1.14 \pm 0.36	0.49 \pm 0.09	0.43 \pm 0.13	0.58 \pm 0.05
Calditrichaceae	0.55 \pm 0.19	1.44 \pm 0.18	1.97 \pm 0.24	0.68 \pm 0.14	1.3 \pm 0.35	2.13 \pm 0.9	0.44 \pm 0.19	0.58 \pm 0.23	0.69 \pm 0.12
Spirochaetaceae	0.26 \pm 0.03	0.36 \pm 0.06	0.65 \pm 0.43	0.43 \pm 0.06	0.47 \pm 0.08	0.55 \pm 0.23	0.09 \pm 0.07	0.04 \pm 0.05	0.07 \pm 0.07
Thioalkalspiraceae	1.13 \pm 0.65	0.57 \pm 0.15	1.08 \pm 0.71	0.25 \pm 0.04	0.63 \pm 0.35	0.23 \pm 0.21	0.21 \pm 0.21	0.07 \pm 0.09	0.1 \pm 0.12
Vibrionaceae	0.32 \pm 0.27	0.07 \pm 0.1	0 \pm 0	0.12 \pm 0	0.04 \pm 0.06	0.07 \pm 0.12	0.49 \pm 0.41	0.11 \pm 0.12	0.02 \pm 0.03
Gemmatimonadaceae	0.09 \pm 0.15	0.58 \pm 0	0.48 \pm 0.25	0.2 \pm 0.18	0.42 \pm 0.16	0.98 \pm 0.32	0.26 \pm 0.26	0.63 \pm 0.15	0.74 \pm 0.17
Pirellulaceae	0.62 \pm 0.41	0.99 \pm 0.08	1.13 \pm 0.81	0.29 \pm 0.27	0.66 \pm 0.06	0.78 \pm 0.27	0.35 \pm 0.38	0.74 \pm 0.16	0.79 \pm 0.27
Entotheonellaceae	0.18 \pm 0.1	0.6 \pm 0.14	0.61 \pm 0.62	0.04 \pm 0.06	0.12 \pm 0.03	0.23 \pm 0.14	0.09 \pm 0.08	0.79 \pm 0.15	1.28 \pm 0.38
Nitrosomonadaceae	0.65 \pm 0.25	0.53 \pm 0.2	0.14 \pm 0.08	0.16 \pm 0.02	0.25 \pm 0.01	0.16 \pm 0.03	0.67 \pm 0.46	1.11 \pm 0.31	0.99 \pm 0.29
Ectothiorhodospiraceae	0.27 \pm 0.12	0.31 \pm 0.3	0.19 \pm 0.17	0 \pm 0	0.08 \pm 0.07	0.05 \pm 0.09	0.05 \pm 0.08	0 \pm 0	0.06 \pm 0.06
Desulfuromonadaceae	0.2 \pm 0.2	0.09 \pm 0.12	0.03 \pm 0.06	0.21 \pm 0.07	0.23 \pm 0.07	0.21 \pm 0.08	0.26 \pm 0.19	0.3 \pm 0.1	0.33 \pm 0.14

KF-JG30-B3	0.12 ± 0.04	0.3 ± 0.04	0.07 ± 0.07	0.13 ± 0.11	0.1 ± 0.01	0.07 ± 0.08	0.24 ± 0.06	0.51 ± 0.28	0.25 ± 0.13
SG8-4	0.13 ± 0.12	0.18 ± 0.04	0.52 ± 0.18	0.09 ± 0.16	0.24 ± 0.4	0.38 ± 0.4	0.09 ± 0.08	0.06 ± 0.11	0.12 ± 0.11
Acidiferrobacteraceae	0.59 ± 0.11	0.52 ± 0.16	0.38 ± 0.12	0 ± 0	0.04 ± 0.05	0.03 ± 0.03	0.04 ± 0.07	0.34 ± 0.1	0.47 ± 0.22
Kiritimatiellaceae	0.2 ± 0.15	0.04 ± 0.05	0.03 ± 0.04	0.4 ± 0.08	0.09 ± 0.08	0.19 ± 0.15	0.28 ± 0.07	0.1 ± 0.1	0.03 ± 0.03
Moduliflexaceae	0.01 ± 0.02	0.05 ± 0.04	0.43 ± 0.39	0.07 ± 0.07	0.14 ± 0.13	0.56 ± 0.28	0 ± 0	0 ± 0	0 ± 0
Thermoanaerobaculaceae	0.58 ± 0.47	1.81 ± 0.11	1.23 ± 1.19	0.93 ± 0.15	1.19 ± 0.5	1.9 ± 0.86	0.47 ± 0.23	0.9 ± 0.26	1.21 ± 0.47
Nitrosococcaceae	0.03 ± 0.05	0.13 ± 0.06	0.05 ± 0.05	0.46 ± 0.04	0.27 ± 0.19	0.29 ± 0.07	0.28 ± 0.06	0.12 ± 0.05	0.14 ± 0.06
Thiohalorhabdaceae	0 ± 0	0 ± 0	0 ± 0	0.44 ± 0.16	0.11 ± 0.15	0.08 ± 0.13	0.24 ± 0.01	0.07 ± 0.08	0 ± 0
PHOS-HE36	0.23 ± 0.11	0.48 ± 0.07	0.4 ± 0.11	0.14 ± 0.25	0.59 ± 0.33	0.92 ± 0.38	0 ± 0	0.21 ± 0.06	0.39 ± 0.25
Prolixibacteraceae	0.4 ± 0.23	0.01 ± 0.01	0.01 ± 0.01	0.9 ± 0.15	0.66 ± 0.2	0.34 ± 0.27	0.35 ± 0.15	0.15 ± 0.06	0.23 ± 0.06
Rubinisphaeraceae	0.15 ± 0.09	0.09 ± 0.02	0.19 ± 0.2	0.03 ± 0.04	0 ± 0.01	0 ± 0	0.06 ± 0.1	0.19 ± 0.05	0.09 ± 0.08
Flavobacteriaceae	1.08 ± 0.43	0.05 ± 0.07	0.01 ± 0.01	1.67 ± 0.39	0.7 ± 0.61	0.38 ± 0.51	2.54 ± 1.38	0.33 ± 0.16	0.01 ± 0.01
SB-5	0.45 ± 0.44	0.46 ± 0.11	0.67 ± 0.48	0.21 ± 0.15	0.49 ± 0.1	0.35 ± 0.14	0 ± 0.01	0.06 ± 0.05	0.23 ± 0.07
Woeseiaceae	0.23 ± 0.16	0.22 ± 0.04	0.02 ± 0.02	2.17 ± 0.73	0.85 ± 0.58	0.75 ± 0.54	1.44 ± 0.22	0.48 ± 0.24	0.33 ± 0.13
Nitrospiraceae	0.01 ± 0.02	0 ± 0	0.14 ± 0.2	0.3 ± 0.09	0.15 ± 0.1	0.09 ± 0.12	0.35 ± 0.11	0.53 ± 0.14	0.59 ± 0.19
B1-7BS	0 ± 0	0 ± 0	0 ± 0	0.11 ± 0.03	0.07 ± 0.07	0.24 ± 0.09	0.16 ± 0.02	0.28 ± 0.09	0.3 ± 0.11
Solibacteraceae_Subgroup3	0.08 ± 0.02	0.15 ± 0.02	0.14 ± 0.09	0.03 ± 0.06	0.2 ± 0.1	0.18 ± 0.11	0.32 ± 0.2	0.41 ± 0.07	0.41 ± 0.13
Nitrincolaceae	0.33 ± 0.19	0.05 ± 0.03	0 ± 0	0.1 ± 0.12	0.08 ± 0.07	0 ± 0	0.65 ± 0.14	0.33 ± 0.13	0.1 ± 0.02
Gallionellaceae	0.44 ± 0.18	0.03 ± 0.02	0.12 ± 0.11	0 ± 0	0.05 ± 0.05	0.01 ± 0.01	0.1 ± 0.18	0.03 ± 0.06	0.1 ± 0.15
Phycisphaeraceae	0.08 ± 0.1	0.19 ± 0.09	0.21 ± 0.06	0.44 ± 0.24	0.2 ± 0.02	0.45 ± 0.18	0.49 ± 0.09	0.31 ± 0.03	0.36 ± 0.13
Ignavibacteriaceae	0.64 ± 0.34	0.31 ± 0.04	0.12 ± 0.09	0 ± 0.01	0.18 ± 0.08	0.08 ± 0.14	0 ± 0	0.2 ± 0.09	0.36 ± 0.2
Gimesiaceae	0.55 ± 0.18	0.57 ± 0.11	0.25 ± 0.18	0.15 ± 0.06	0.18 ± 0.12	0.15 ± 0.15	0.55 ± 0.38	0.57 ± 0.22	0.38 ± 0.17
A4b	0.34 ± 0.18	0.66 ± 0.09	0.39 ± 0.1	0.34 ± 0.17	0.2 ± 0.15	0.24 ± 0.07	0.66 ± 0.13	1.06 ± 0.44	0.94 ± 0.32
Sphingomonadaceae	0.4 ± 0.3	0.14 ± 0.04	0.07 ± 0.1	0.26 ± 0.05	0.2 ± 0.22	0.17 ± 0.16	0.63 ± 0.42	0.42 ± 0.19	0.17 ± 0.09
Stappiaceae	0 ± 0	0 ± 0	0 ± 0	0.25 ± 0.16	0.09 ± 0.1	0.12 ± 0.17	0.02 ± 0.04	0 ± 0	0.07 ± 0.12
Rhodobacteraceae	0.97 ± 0.3	0.02 ± 0.02	0 ± 0	0.67 ± 0.05	0.51 ± 0.37	0.24 ± 0.3	1.19 ± 0.4	0.38 ± 0.08	0.18 ± 0.08

- 1 **Supplementary data F:** Values of the Spearman's rank correlation coefficient between the environmental variables and the taxa at the phylum
- 2 levels (*p = 0.05, **p= 0.01, ***p=0.001), HCB = Hexachlorobenzene, Naph = Naphthalene, Phen = Phenanthrene.

Phylum	Al	Cd11 1	CN	Cr	Ctot	ddd24	dde44	DBT	dieldrin	HCB	Hg	Mn	Mo9 5	Naph	Ntot	PCBs	Phen	Zn
Acidobacteria	0,2	-0,7 (**)	-0,7 (**)	0,3	-0,6 (**)	-0,1	-0,1	0,3	-0,3	-0,3	-0,2	0,4	-0,2	0,5	-0,6 (*)	-0,3	0,0	0,3
Actinobacteria	-0,4	0,5	0,4	-0,5	0,3	0,3	0,1	-0,2	0,4	0,2	0,1	-0,4	-0,1	-0,6 (*)	0,1	0,4	-0,1	-0,5
Altiarchaeota	-0,3	0,2	0,2	-0,3	0,1	-0,2	-0,1	-0,2	-0,1	0,2	0,3	-0,3	0,1	-0,2	0,0	0,0	-0,1	-0,3
AncK6	-0,4	0,3	0,3	-0,5	0,2	0,1	0,2	0,0	0,3	0,3	0,4	-0,5 (*)	0,2	-0,2	0,0	0,3	0,1	-0,4
Asgardaeota	-0,5	0,6 (*)	0,7 (***)	-0,6 (*)	0,7 (**)	0,0	0,2	-0,2	0,3	0,5	0,7 (**)	-0,8 (***)	0,6 (*)	-0,4	0,4	0,3	0,2	-0,5
Bacteroidetes	0,3	0,1	-0,1	0,2	0,0	0,0	-0,1	-0,2	-0,1	-0,2	-0,3	0,5	-0,2	-0,1	0,3	-0,1	-0,2	0,2
BRC1	-0,4	0,0	0,2	-0,2	0,2	0,1	0,1	0,2	0,0	0,1	0,3	-0,4	0,3	0,1	-0,1	0,1	0,2	-0,2
Calditrichaeota	-0,4	0,1	0,3	-0,4	0,1	0,5	0,5	0,0	0,7 (**)	0,7 (**)	0,0	-0,5	0,0	-0,4	-0,2	0,7 (**)	0,0	-0,5
Chlamydiae	-0,3	0,4	0,3	-0,4	0,3	0,1	0,2	0,0	0,3	0,0	0,2	-0,4	0,2	-0,2	0,2	0,2	0,0	-0,2
Chloroflexi	-0,3	0,2	0,3	-0,2	0,3	-0,2	0,0	0,1	-0,1	0,0	0,6 (*)	-0,6 (**)	0,5	0,1	0,0	-0,2	0,2	-0,1
CK-2C2-2	-0,2	0,3	0,2	-0,2	0,3	-0,2	-0,1	0,1	0,2	0,0	0,2	-0,3	0,2	-0,2	0,2	0,0	0,3	0,0
Crenarchaeota	-0,6 (*)	0,6 (*)	0,7 (***)	-0,5	0,6 (**)	0,2	0,2	-0,1	0,4	0,5	0,4	-0,9 (***)	0,3	-0,6 (*)	0,2	0,5	0,1	-0,5
Dadabacteria	0,0	-0,1	-0,2	0,2	-0,2	-0,2	-0,4	0,2	-0,3	-0,1	0,2	-0,2	0,1	0,2	-0,5	-0,3	0,2	0,1
Deinococcus-Thermus	-0,2	0,3	0,2	-0,3	0,2	0,2	0,0	0,1	0,1	0,1	0,0	-0,2	0,0	-0,1	0,2	0,2	0,3	-0,3
Dependentiae	-0,5	0,1	0,3	-0,4	0,2	0,1	0,0	-0,1	0,2	0,3	0,1	-0,3	0,0	-0,2	-0,1	0,2	0,1	-0,5
Diapherotrites	-0,2	0,3	0,3	-0,3	0,3	0,0	0,1	0,1	0,0	0,3	0,3	-0,3	0,3	-0,2	0,1	0,1	0,1	-0,2
Elusimicrobia	0,0	-0,2	-0,3	-0,2	-0,2	0,3	0,3	-0,2	0,3	0,3	-0,3	0,3	-0,3	-0,2	0,0	0,3	-0,3	0,0
Entotheonellaeota	0,0	-0,2	-0,2	0,1	-0,1	-0,3	-0,1	0,3	-0,4	-0,1	0,5	-0,2	0,4	0,4	-0,2	-0,4	0,3	0,1
Epsilonbacteraeota	0,0	0,1	0,1	-0,1	0,2	0,0	-0,1	-0,2	-0,1	-0,3	-0,2	0,3	-0,1	-0,1	0,4	-0,1	-0,1	0,0
Euryarchaeota	-0,5	0,7 (***)	0,8 (***)	-0,5	0,7 (**)	0,0	0,1	-0,3	0,3	0,4	0,6 (*)	-0,9 (***)	0,5	-0,5	0,4	0,2	0,1	-0,5
FCPU426	0,0	0,2	0,0	-0,1	0,0	0,0	-0,2	0,1	-0,2	0,0	0,1	0,0	-0,1	0,1	0,1	-0,1	0,2	0,0
Fibrobacteres	0,0	0,2	0,1	-0,1	0,1	0,0	0,0	0,0	0,3	0,3	-0,2	0,0	-0,1	-0,4	0,2	0,3	-0,2	-0,1
Firmicutes	-0,2	-0,1	0,1	-0,2	0,0	0,2	0,2	0,0	0,1	0,0	0,2	0,0	0,2	0,1	0,0	0,1	0,2	-0,2
Fusobacteria	-0,3	0,3	0,3	-0,3	0,2	0,4	0,4	0,0	0,3	0,2	0,2	-0,3	0,2	-0,1	0,3	0,4	0,1	-0,3
Gemmatimonadetes	0,2	-0,5	-0,3	0,2	-0,5	0,2	-0,1	0,0	0,2	0,2	-0,4	0,4	-0,5	0,1	-0,5	0,1	0,0	0,2
GN01	-0,3	0,1	0,2	-0,2	0,0	0,4	0,3	-0,1	0,5	0,5 (*)	0,1	-0,5	-0,1	-0,1	-0,3	0,4	0,0	-0,3

Hydrogenedentes	0,2	-0,4	-0,3	0,2	-0,3	-0,1	-0,1	0,0	-0,2	-0,3	0,0	0,3	-0,1	0,3	-0,1	-0,3	-0,1	0,2
Kiritimatiellaeota	0,4	0,1	-0,1	0,2	0,0	0,1	0,0	-0,2	0,1	0,1	-0,5	0,4	-0,3	-0,1	0,3	0,1	-0,2	0,2
Latescibacteria	0,6 (*)	-0,5	-0,6 (**)	0,5	-0,6 (*)	-0,2	-0,2	0,2	-0,1	-0,2	-0,5	0,6 (*)	-0,4	0,3	-0,3	-0,2	-0,1	0,5
LCP-89	-0,4	0,1	0,3	-0,4	0,1	0,3	0,3	0,0	0,4	0,5	0,2	-0,5 (*)	0,0	-0,1	-0,2	0,4	0,0	-0,4
Lentisphaerae	0,4	-0,3	-0,4	0,3	-0,4	0,2	0,1	0,2	0,1	-0,1	-0,6 (*)	0,6 (**)	-0,6 (*)	0,2	-0,1	0,1	0,0	0,4
Margulisbacteria	0,1	-0,2	-0,1	0,2	0,0	-0,2	-0,1	-0,2	-0,2	-0,2	0,1	0,1	0,2	0,2	0,0	-0,2	-0,3	0,2
Marinimicrobia	-0,2	0,3	0,3	-0,2	0,2	0,1	-0,1	-0,1	0,0	0,0	0,1	-0,4	0,1	-0,1	-0,1	0,1	0,0	-0,3
Modulibacteria	-0,3	0,3	0,3	-0,3	0,2	0,4	0,3	-0,2	0,7 (**)	0,7 (**)	-0,1	-0,4	-0,1	-0,4	-0,1	0,6 (**)	-0,1	-0,4
Nanoarchaeaeota	-0,5	0,3	0,5	-0,4	0,4	0,0	0,1	0,0	0,2	0,2	0,4	-0,7 (**)	0,4	-0,2	0,0	0,2	0,1	-0,4
Nitrospinae	-0,3	-0,1	0,1	-0,3	0,0	0,3	0,4	0,3	0,1	0,1	0,3	-0,2	0,1	0,2	0,0	0,1	0,2	-0,1
Nitrospirae	0,3	-0,7	-0,7 (**)	0,4	-0,7 (**)	0,2	0,1	0,2	-0,1	0,0	-0,3	0,4	-0,4	0,4	-0,7 (***)	-0,1	-0,1	0,3
Ochrophyta	-0,2	0,1	0,1	-0,1	0,1	0,3	-0,1	-0,2	0,0	-0,1	-0,2	0,0	0,0	-0,2	-0,2	0,2	-0,2	-0,2
Omnitrophicaeota	-0,5	0,3	0,4	-0,4	0,3	0,0	0,2	0,1	0,2	0,2	0,5 (*)	-0,6 (**)	0,4	-0,1	0,1	0,1	0,3	-0,4
Patescibacteria	0,0	-0,2	-0,1	0,0	-0,2	0,4	0,2	0,2	0,2	-0,1	-0,1	0,1	-0,3	0,2	-0,4	0,2	0,0	0,0
PAUC34f	0,0	-0,1	0,0	0,1	-0,1	0,3	0,4	0,4	0,3	0,1	0,0	-0,1	-0,2	0,3	-0,2	0,2	0,2	0,1
Planctomycetes	0,0	-0,3	-0,3	-0,1	-0,2	0,2	0,4	0,3	0,2	0,3	-0,1	0,2	-0,1	0,2	-0,1	0,2	0,2	0,1
Proteobacteria	0,3	0,2	0,1	0,1	0,1	-0,1	-0,1	-0,1	0,0	-0,1	-0,2	0,3	-0,1	-0,2	0,5	0,0	0,0	0,1
Rokubacteria	0,1	-0,4	-0,5	0,3	-0,4	-0,4	-0,2	0,4	-0,7 (**)	-0,6 (*)	0,1	0,2	0,0	0,7 (*)	-0,3	-0,6 (**)	0,2	0,3
Schekmanbacteria	0,0	-0,3	-0,2	0,1	-0,3	0,3	0,1	0,1	0,2	0,1	-0,1	-0,2	-0,1	0,2	-0,7 (**)	0,2	0,0	0,0
Spirochaetes	-0,4	0,3	0,5	-0,5	0,3	0,5	0,5	-0,2	0,7 (**)	0,6 (*)	-0,1	-0,3	-0,1	-0,6 (*)	0,1	0,8 (***)	-0,2	-0,6
TA06	-0,3	0,0	0,1	-0,3	-0,1	0,3	0,2	-0,1	0,4	0,5	0,0	-0,4	-0,2	-0,1	-0,4	0,4	-0,2	-0,4
Tenericutes	-0,1	0,2	0,1	-0,1	0,1	-0,2	-0,2	-0,2	-0,2	-0,3	0,3	0,0	0,2	0,0	0,1	-0,2	0,0	-0,1
Thaumarchaeota	0,7 (*)	-0,6 (*)	-0,6 (**)	0,6 (*)	-0,6 (**)	0,0	-0,2	0,1	-0,2	-0,3	-0,7 (**)	0,8 (***)	-0,6 (*)	0,3	-0,4	-0,2	-0,3	0,6
Verrucomicrobia	0,5	-0,2	-0,4	0,4	-0,2	-0,4	-0,3	0,3	-0,5 (*)	-0,6 (*)	-0,2	0,4	-0,1	0,4	0,1	-0,6 (*)	0,1	0,5
WS1	-0,3	-0,4	-0,2	-0,2	-0,2	0,3	0,2	0,1	0,3	0,3	-0,1	-0,1	-0,4	0,1	-0,4	0,3	0,1	-0,3
WS2	-0,2	0,0	0,2	-0,1	0,2	0,0	-0,3	-0,1	0,1	0,1	-0,2	-0,1	-0,2	-0,4	0,0	0,1	0,0	-0,2
WS4	-0,2	0,1	0,1	-0,1	0,1	0,3	-0,1	-0,2	0,0	-0,1	-0,2	0,0	0,0	-0,2	-0,2	0,2	-0,2	-0,2
Zixibacteria	-0,3	0,2	0,3	-0,3	0,3	0,1	0,2	0,1	0,1	0,2	0,5	-0,6 (**)	0,4	0,0	0,0	0,2	0,2	-0,2