Kinetics of metal and metalloid concentrations in holopelagic *Sargassum* reaching coastal environments

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Abstract :

Since 2011, the Caribbean Islands have experienced unprecedented stranding of a pelagic brown macroalgae Sargassum inducing damages for coastal ecosystems and economy. This study measures the kinetics of metal trace elements (MTE) in Sargassum reaching different coastal environments. In July 2021, over a period of 25 days, fixed experimental floating cages containing the three Sargassum morphotypes (S. fluitans III and S. natans I and VIII) were placed in three different coastal habitats (coral reef, seagrass, and mangrove) in Guadeloupe (French West Indies). Evolution of biomasses and their total phenolic content of Sargassum reveals that environmental conditions of caging were stressful and end up to the death of algae. Concentrations of 19 metal(loid) trace elements were analyzed and three shapes of kinetics were identified with the MTE that either concentrate, depurate, or remains stable. In the mangrove, evolution of MTE was more rapid than the two other habitats a decrease of the As between 70 and 50 µg g-1 in the mangrove. Sargassum natans I presented a different metal composition than the two other morphotypes, with higher contents of As and Zn. All Sargassum morphotype are rapidly releasing the metal(oid)s arsenic (As) when they arrive in studied coastal habitats. In order to avoid the transfer of As from Sargassum to coastal environments, Sargassum stranding should be avoided and their valorization must take into account their As contents.

Keywords : Arsenic, Sargassum, Coral reef, Seagrass, Mangrove, Caribbean, Metals

48 **1 Introduction**

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- 50 The Sargassum genus includes more than 350 species, constituting one of the most diverse
- 51 genera of brown macroalgae (Guiry and Guiry, 2022). Among this genus, only two species are
- 52 holopelagic as they drift during their entire life cycle (Dawes and Mathieson, 2008) constituting
- floating rafts called "the golden floating rainforest of the Atantic Ocean" (Laffoley et al., 2011).
- 54 Morphological and molecular studies differentiated three genotypes: S. fluitans III and S. natans
- 55 I and VIII (Amaral-Zettler et al., 2017).
- 56 Historically holopelagic *Sargassum* spp. were present in the Caribbean Sea, at the edge of the
- 57 Gulf Mexico and the Azores islands (Lapointe, 1995). In summer 2011, unprecedented
- 58 quantities of *Sargassum* started to inundate the Caribbean Islands (Gower and King, 2011). In

some places such as northeastern Brazil, *Sargassum* stranded in locations have also spotted that
were never reported before (Széchy et al., 2012). In 2018, 20 million metric tons wet biomass
of Sargasso in the open Ocean formed a Great Atlantic Sargasso Belt extended for 8,850 km
length, since then, this Great Atlantic Sargasso belt is reported annually in the North Equatorial
Recirculation Region (NERR) (Wang et al., 2019).

64 The origins of the sudden and recurring increased of Sargassum abundance still remains unclear (Ardhuin et al., 2019), and different hypotheses are proposed such as an increase in i) sea 65 surface temperature (Sissini et al., 2017), ii) nutrients released from Amazon and Congo rivers 66 67 (Oviatt et al., 2019) and iii) deposition of dust from African desert (Johns et al., 2020). The stranding of Sargassum spp. on the coast areas have ecological issues threatening marina fauna 68 (Cipolloni et al., n.d.; Rodríguez-Martínez et al., 2019) including endangered species such as 69 70 sea turtles (Maurer et al., 2022, 2015; Ross and Casazza, 2008) and can lead to the disappearance of coastal ecosystems (Gledhiir and Buck, 2012; van Tussenbroek et al., 2017). 71 72 Decomposition of abundant brown algae biomass accumulated in coastal environment liberates 73 toxic hydrogen sulphide (H_2S) (Reiffenstein et al., 1992) provoking important human health issues such as respiratory diseases, neurological problems and cardiovascular lesions (Resiere 74 75 et al., 2018). Sargassum also represent an economic cost deterring tourism and obstructing free 76 circulation of boat impacting marine trade and fisheries (Langin, 2018).

Additionally, to these visible impacts, *Sargassum* can generate pernicious and invisible impacts
due to metal trace elements, contamination as it shows a high capacity of absorption of metals
and metalloids contaminants due to the high metallic affinity of alginate in their cell walls
(Davis et al., 1999; Vieira and Volesky, 2000; Volesky and Holan, 1995). Holopelagic *Sargassum spp.* present high level of the total As with a concentrations fluctuating between
100 ppm and 145 ppm (Cipolloni et al., 2022; Dassié et al., 2021; Devault et al., 2020) and can
release this metalloid in coastal environments contaminating marine species (Cipolloni et al.,

n.d.). Arsenate absorbed by the algae is transformed in arsenite As(III) (Andreae and Klumpp,
1979; Howard et al., 1995). Inorganic arsenic, the most toxic form, represent a consistent and
substantial percentage of the total arsenic present in pelagic *Sargassum* spp. (Alleyne et al.,
2023). To our knowledge, the speciation of As released by *Sargassum* is not known.

However, this transfer is still poorly documented. Information on the kinetics and intensity of
those transfers in different coastal environments constitute an important information for the
implementation of coherent stranding management policy.

In addition of the metallic trace elements, the conditions of the experiments were also analyzed
in order to evaluate the physiological condition of the brown algae using their stable isotope
(Gager et al., 2021) composition and their phenolic compounds.

94 The aim of the present study was thus to experimentally determine the kinetics of accumulation 95 or depuration of 19 MTE during twenty-five days in three morphotypes of holopelagic 96 *Sargassum* arriving in three different coastal environments: *i*) coral reef *ii*) seagrass meadow 97 and *iii*) mangrove.

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99 2 Materials and methods

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2.1 Study sites and experimental setting-up construction

The Grand Cul-de-Sac marin (GCSM) in Guadeloupe (French West Indies) presents shallow 102 103 waters (less than 20m depth) bordered by a coral reef at the North and a mangrove at the South 104 (Guilcher and Marec, 1978). Over a period of 25 days during the month of July 2021, three 105 fixed experimental devices were placed in GCSM in the three different habitats (coral reef, seagrass and mangrove) (Fig.1 A). Different habitats are localized at a distance at least 200m. 106 The coral reef is a natural bio constructed structure mainly composed of corals, followed by 107 108 seagrass forming dense underwater meadows and mangrove forest are closer to terrestrial 109 environment.

110 Each experimental device was composed of five floating plastic cages with dimension of 30 cm in diameter and 20 cm in height. Sargassum freshly collected in the Petit Cul-de-Sac marin 111 (PCSM) were rapidly (less than one hour) placed in experimental device. Control sample (n=3) 112 113 at the beginning of incubation (t=0) were collected, of each species. A fixed fresh weight of approximatively 60g of Sargassum of the mixed three morphotypes (S. fluitans III and S. natans 114 115 I and S. natans VIII) was separated morphologically at the experimental devices stations and placed in each cage (Fig.1) BAt different temporal intervals (days 1, 4, 11, 18 and 25) 116 117 macroalgae contained in each cage were simultaneously sampled in each habitat (coral reef, 118 seagrass meadow, mangrove). After collection, each sample was separated by genotypes, placed in paper wraps and oven-dried during 48 h at 50°C. In total, 47 samples were collected 119 120 during the experiment and one sample was missing due to disappearance of S. natans VIII in 121 the last sampling cage in mangrove.

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- 123 **2.2** Laboratory analyses
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125 Biomass analysis and laboratory preparation

After the drying step, each *Sargassum* morphotype sample was weighed. The samples were then ground and homogenized using a vibro-grinder with zirconium balls of 10 mm for three min with a frequency of 30 beat/s (Retsch® MM 400). Grounded samples were used to carry all the following measurements: stable isotope, phenolic compounds and MTE levels.

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131 Phenolic compounds analysis

Phenolic compounds were extracted twice using 15 mg DW of algal powder in 1 mL of 70 %
ethanol according to a modified method from (Zubia et al., 2009). The extractions were carried
out with an ultrasonic bath (Sonicator 88155, Fisher 160 Bioblock Scientific, France) during

135 15 min at 4 °C followed by 2 h at 40 °C under magnetic stirring. Then, samples were centrifuged for 10 min at 8,000 rpm (Eppendorf Centrifuge 162 5810, Germany) and supernatants were 136 pooled and evaporated at 40 °C using a centrifugal concentrator (miVac, Genevac, France). 137 138 Total phenolic content (TPC) was determined using the Folin-Ciocalteu colorimetric assay modified from (Zubia et al., 2009). Thus, 20 µL of sample was added to 130 µL of distilled 139 water, 10 µL of Folin-Ciocalteu reagent and 40 µL of sodium carbonate (Na₂CO₃, 200 g.L⁻¹). 140 Then microplates were incubated for 10 min at 70 °C before absorbance reading in triplicate at 141 620 nm (Multiskan FC, Thermo Scientific, USA). TPC was determined using a standard curve 142 143 of phloroglucinol (1,3,5-trihydroxybenzene) and expressed in milligrams per gram of the dried seaweed powder (mg.g⁻¹DW) and in percentage of TPC against day 0 level to see the evolution 144 145 of Sargassum phenolic content during the experiment.

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147 Isotope analysis and calculation

148 The δ^{15} N and the δ^{13} C isotopic compositions of each *Sargassum* samples (*S. fluitans* III and 149 *S. natans* I and *S. natans* VIII) from the experimental devices were measured by EA-IRMS 150 (Elemental Analysis – Isotope Ratio Mass Spectrometry) (Narancic et al., 2017). The isotopes 151 compositions were expressed as δ – *values* relative to reference standard in per mil (%) such 152 as nitrogen composition is expressed in delta notation as:

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$$(\delta^{15}N) = \left[\frac{\binom{1^5N}{_{14_N}}sample}{\binom{1^5N}{_{14_N}}reference}} - 1\right] \times 100$$

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156 *Metal(loid)s trace elements analysis*

157 A series of 19 elements (Ag, Al, As, Ba, Cd, Co, Ca, Cr, Cu, Fe, Gd, Mn, Mo, Ni, Pb, Se, Sr,

158 V, and Zn) were analyzed using an Inductively Coupled Plasma Optical Emission Spectrometer

159 (Spectrometer ICP-OES 700®, Agilent Technologies). Certified reference materials DOLT-5

160 (dogfish (*Squalus acanthias*) liver), TORT-3 (Lobester Hepatopancreas), and IAEA-413 161 (Alagae) were analyzed using ICP-OES, their recovery rates vary between $84,1\pm3,33$ and 162 $111,6\pm0,21$ (Table 1). For the values below the instrument detection limit, theoretical minimum 163 concentration values are calculated (the detection limit of the instrument (in $ug. g^{-1}$) multiplied by 164 the volume of the sample (in L) divided by the sample *Sargassum* weight (in g)).

For each sample, a fixed amount of algal powder (70-80mg) was placed in a plastic tube and acidified by the addition of 1 mL of nitric acid (HNO_3 67%). The powdered sample was then mineralized for 3 h at 100°C (Environmental – EXPRESS HotBlock® - 54). After mineralization, 5 mL of deionized water was added to each sample. With identical process, certified reference materials (DOLT-5, TORT-3, IAEA-413) were analyzed and were systematically in the concentration range. The metal concentrations in *Sargassum* samples were expressed in $\mu g.g^{-1}(ppm)$ dry weight.

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- **2.3 Data analysis**

174 The variance and the homogeneity of metals and metalloid concentration values were verified 175 by the Shapiro's and Levene's tests, respectively (with significance at the 95% confidence level). As normality was not observed, the differences between means concentrations in 176 177 genotypes per habitat and in all habitats were tested using the non-parametric test Kruskal-Wallis test. Principal Component Analyses (PCA) were executed on RStudio® and RCran, 178 using the following packages: FactoMiner (Husson et al., 2020), factoextra (Kassambara and 179 180 Mundt, 2020), ggplot (Wickham et al., 2020) and corrplot (Wei et al., 2021) to select the MTE with higher influence in data structuration between the 19 elements (Al, As, Cd, Co, Cr, Cu, 181 182 Fe, Mn, Ni, Pb, Se, Sr, V, and Zn). Metallic elements (Co, Pb, Se, and Sr) below the limit of detection (LOD), were not considered. 183

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185 **3** Results

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3.1 Indicators of the physiological condition of *Sargassum*: biomass, phenolics contents and isotopic signatures.

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190 There were two distinct phases in the physiological state of the algae. The biomass of each 191 morphotype of *Sargassum (S. fluitans III, S. natans I and VIII)* in the three habitats (coral reef, 192 seagrass meadow and mangrove) regularly increased after the beginning of the experiment and 193 started to decrease after the day 18 (Fig.2).

194 In the three habitats, phenolic contents of each morphotype also follows similar kinetics with

195 two distinct phases, *i*) a first phase of decrease in phenolic content compounds until the 11th 196 days, and *ii*) a second phase with an increase in phenolic content from the 18th day to the end 197 of the experiment (Fig.2). The phenolic content in $ug.g^{-1}$ was higher in the morphotype

198 S.fluitans III, (25 and 30 $ug.g^{-1}$) than in S.natans I (10 and 20 $ug.g^{-1}$) and S.natans VIII

199 (10 and 12 $ug.g^{-1}$).

200 Isotopic compositions (δ^{15} N and δ^{13} C) of *Sargassum* were not clearly differ

rent between all habitats and morphotypes. The values of the isotopic signature in the three habitats, for the three genotypes remains around (3%-4%) for the δ^{15} N and 14%-18% for the δ^{13} C.

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205 **3.2 Metals and metalloids content**

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Ten elements (Al, As, Cd, Cu, Fe, Mn, V, Ni, Cr and Zn) were the most abundant and were
detected in all samples above the limit of detection (LOD). The elements Ag, Co, Sr, Se, Mo,
Gd, Ca, Pb and Ba were below the LOD. (Table 1)

The variability of the data analyzed, has been verified by tri-replicates on the measurements ofthe samples analyzed.

Kinetics analysis were focused on the five metallic elements standing out in PCA analysis (Al,
As, Fe, Cu and Zn). Three kinetics profiles are observed: *i*) a significant decrease contamination
(As), *ii*) an increase in contamination (Zn) and (Cu) *iii*) a bell-shaped profile (Al and Fe) (Fig.3).

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3.3 Principal Component Analysis (PCA)

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Principal Component Analysis (PCA) was used to evaluate the influence of habitat and *Sargassum* morphotype on metallic elemental concentrations. The first two dimensions of PCA
representing respectively 38.83% (F1) and 13.17% (F2) of the total variance (Fig 4.A). F1
distinctly discriminates the variables Al (13.37%), Fe (14.45%), Zn (14.08%), and Cu (15.25%), whereas and F2 clearly discriminates As (11.65%).

The PCA analysis of the sample discriminates the mangrove habitat characterized by high 225 226 concentrations in Fe, Al, Zn, Cu and As in Sargassum (Fig.4 B) whereas samples from seagrass meadow and coral reef were similar with high concentrations of Ni, V, Cd and Cr. PCA 227 228 discrimination according to Sargassum by the day (Control; Day=1; Day=4, Day=11, Day=18 229 and Day=25) showed that the evolution of the variability of MTE concentrations increased between inter-habitats. Except, for the Day 25th with lower concentration in metallic elements 230 (Fig.4 B). On the PCA analysis whatever the habitat, and the experiment duration, each of the 231 232 three morphotypes followed similar trend (Fig. 4 B).

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- 235 4 Discussion
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The present study measured the kinetics of metal(oid)s trace elements contaminations of holopelagic *Sargassum (S.fluitans* III and *S.natans* I and *S.natans* VIII) remaining floating in three different tropical coastal environments. Different kinetics patterns were observed according to *i*) MTE *ii*) coastal environments with highest fluctuations in mangrove *iii*) and morphotype due to the singularity of the genotype *S. natans* I.

In our sample entire macroalgal thalli were dried and ground before the analyzes and each
morphological character (leaves, stems, bladders) was not separately analyzed masking
potential specificities of metal concentrations in each algal tissue (Sadeghi et al., 2014).
However, in our samples were homogeneous, and triplicates of the measures were realized,
which removes the bias of the measures.

Environmental parameters were not measured during the experiments as each habitat presented many specificities making the identification of most structuring variable complex. As a result, the present experimental approach in *situ* did not allow to identify the specific role played by each variable but provided results realistically transposable to *in situ* field conditions. Those field conditions are representative of each habitat with a classical decreased gradient of terrestrial influence from mangrove to coral reef. Mangrove are consequently classically more influenced by freshwater and organic matter inputs than the two other habitats.

254 A similar evolution of algal biomasses was generally observed in all samples whatever the 255 morphotype and habitat with a first increase in biomass followed by a decrease. This decrease 256 is likely due to the degradation of seaweeds indicating unsuitable conditions. Physiological or 257 stress conditions of algae were also evaluated measuring phenolics contents (Plouguerné et al., 258 2006). Phenolic compounds of brown seaweeds or phlorotannins are present sometimes in high 259 level, between (10 % and 120%) in the cells walls and the physodes and as there are produced in response to any changes in abiotic (temperature, light) and biotic (grazing, fouling) factor, 260 their content may be used to evaluate algal stress (Arnold and Targett, 2002; Ragan and Craigie, 261

262 1976; Schoenwaelder and Clayton, 1999). Seaweed samples were oven-dried at 50°C and this process can alter phenolics compounds (Gager et al., 2021), however a similar drying method 263 was applied for all samples allowing inter-samples comparisons in the evolution of the phenolic 264 265 content compared to the beginning of the experiment. The evolution of phenolics contents was generally observed in all samples whatever the morphotype and habitat with a first decrease of 266 267 phenolic compounds between the day 0 (Day=D0) to the day four (Day=D4). During this initial phase, the stress of algae would be limited and their biomass would increase. During the second 268 269 phase, the content of phenolic compounds in algae increased, between the day four (Day=D4) 270 and the day tweety-five (Day=D25) potentially due to their production by algae and/or the degradation of the algal thallus with increasing proportion of phenolic compounds from less 271 272 degraded cell walls were still within the thalli and in proportion, their content expressed in $ug.g^{-1}$ algal dw increased (Koivikko et al., 2005; Schoenwaelder and Clayton, 1999). The 273 274 temporal variation in phenolic contents therefore suggest that the algae are first in good condition, then stressed, leading to the degradation of the algae and its death. Evolution of 275 isotopic composition (δ^{15} N and δ^{13} C) of algae during the experiments can reflect the uptake 276 of C and N from the environment and/or the preferential disappearance of isotope form during 277 278 the degradation process. However, no clear trend was observed during the experiment suggesting that stable isotope would not be adapted to evaluate physiological state of 279 Sargassum in this type of experiment. The evolution of biomass and phenolic compounds, with 280 281 the used of trireplicates, both suggest that environmental conditions in cages are stressful for Sargassum and conduct to the death of algae during the experiments. A similar laboratory 282 experience with Sargassum in mesocosm bags during 26h showed a rapid degradation of the 283 284 macroalgae (Devault et al., 2021) which is in agreement with our study.

Initial metals and metalloids concentrations of *Sargassum* used in the present experiment were similar to values previously measured in seaweeds collected in coastal areas (García-Sartal, 2012; Rodríguez-Martínez et al., 2020). However *S.natans* I presented metal(oid)s
concentrations standing out from values of the two others morphotypes whereas this outsider
role was played by *S.natans* VIII in previous studies (Cipolloni et al., 2022; Dassié et al., 2021).
Kinetics of *Sargassum* metal(loid) concentrations followed three different kinetics: *i*) a
significant decrease in contamination (As), *ii*) a significant increase in contamination (Zn and
Cu) *iii*) a bell-shaped profile (Al and Fe).

Marine organisms incidentally take up As through different transporters like the phosphate 293 294 transporter (Garbinski et al., 2019; Saberzadeh Sarvestani et al., 2016). Arsenic can derive in arsenate pentavalent AsO_4^{3-} and this form is similar to the phosphate ion and can consequently 295 296 enter in algae using phosphate transporter pathway (Gobert et al., 2022). In order to tolerate 297 such cellular absorption, algae limit As entrance in cytosol (Garbinski et al., 2019) and accumulate the majority of As as hydrophilic compounds in the cells (Ender et al., 2019). This 298 specific distribution of As could be an explanation of the rapid release of As counterbalanced 299 300 by an increase of other metallic elements like the Fe, Cu, Zn and Al (Delshab et al., 2016; Gobert et al., 2022). This mechanism would explain antagonism between As and Fe 301 302 concentrations in Sargassum previously observed in experimental (Mamun et al., 2019) and in 303 situ conditions (Cipolloni et al., 2022).

304 Due to the proximity with land, coastal environments are more enriched in organic matter than offshore ones. Degradation of this organic matter in coastal area sediment induce hypoxic 305 306 conditions resulting in high contents of metallic elements like Fe, Cu, Zn and Mn (Holloway et al., 2016; Rezaei et al., 2021). The increase in metal elements in Sargassum can be explained 307 by the carboxylate group within alginates (cell wall polysaccharides) of the algae presenting an 308 309 extremely high affinity with divalent metals like Cu, and Zn (He and Chen, 2014). This increased fixation of metallic elements by Sargassum would induce the release of As as 310 previously suggested in the present study. 311

In all the study in the three different coastal environments, metallic elements present hightemporal fluctuations with higher fluctuations in mangrove habitat.

Due to higher proximity with terrestrial environment and high primary production, the 314 315 mangrove is characterized by higher amount of OM, than the two other habitats. The OM can potentially influence the metal availability. Suspended OM present high affinity with metal 316 317 elements and form different complexes (Doig and Liber, 2006). Chelation and sequestration of pollutants in mangrove (Bastakoti et al., 2019) would consequently reduce their bioavailability 318 319 implying a release of this compounds by Sargassum. In accordance with this hypothesis Sargassum were previously observed depurating As due to competitive exchange with 320 terrigenous metals (Gobert et al., 2022). Salinity variations are more important in the mangrove 321 322 than in other habitat due to mainland proximity and decreased salinity specific physiological 323 and morphological processes of mangrove organisms (Clough et al., 1989; Feller et al., 2010). 324 As OM, salinity could leads to the formation of stable metal-chloride complexes decreasing the 325 availability of metallics elements (Mader et al., 1995). In mangrove, the decreased salinity and 326 increased content of organic matter have opposite effects on metallic elements complexation. 327 The observed releasing activity of metalloids (As) by Sargassum in mangrove suggests that OM is more structuring than salinity and reduce metals availability in this environment. 328

329 One morphotype, S.natans I stand out of PCA analyzes in the mangrove. In this environment 330 kinetics of As and Zn contents were faster in S. natans I than for the two other morphotypes (S. 331 natans VIII and S. fluitans III). This specificity of metallic concentration of the morphotype S. natans I was previously observed (Davis et al., 2021; Gobert et al., 2022). This difference could 332 be due to its morphology particularity as S. natans I presenting a more complex structure with 333 higher exchange surface favoring fixation or release of pollutants (Khotimchenko et al., 2001). 334 Compared to other morphotypes S. natans I also present a specific chemical composition, 335 S.natans I appears to be significantly more enriched in P compared to the others morphotypes 336

337 (Gobert et al., 2022). The ability of *S.natans* I to absorb pollutants may notably be due to its
338 alginates which may have a different structure compared to the two other *Sargassum* genotypes
339 as length alginates limit the retention of some cations such as metals (Rhein-Knudsen et al.,
340 2017).

The stranding of *Sargassum* causes visible impacts on environment, economy and public health
(Resiere et al., 2018; van Tussenbroek et al., 2017). Several solutions have consequently been
considered to limit the impacts of *Sargassum* in coastal environments (Robledo et al., 2021).
Less visible impacts such as As contamination of algae must be considered in those strategic
choices of *Sargassum* management.

The total Arsenic is the most widely distributed element in the marine environment with a 346 347 complex biogeochemistry (Fattorini et al., 2006; Neff, 1997). Arsenic concentrations obtained 348 in the present study are in the range of the values previously observed in Sargassum collected in coastal, with values between 80 and 150 ppm (Cipolloni et al., 2022; García-Sartal, 2012; 349 350 Rodríguez-Martínez et al., 2020) and off-shore environments with values a mean of 140 ppm 351 (Cipolloni et al., 2022; Dassié et al., 2021). Those values are above European norms for products intended for human consumption (European Commission, 2019). Our study revealed 352 that, once arrived in coastal environment, Sargassum rapidly release their As and this 353 354 characteristic is observed for all morphotype and coastal ecosystem studied.

Marine algae accumulating As usually biotransform it once in the cells (Alleyne et al., 2023). Brown algae plant have set up a regulation mechanism in order to reduce the toxicity of As (Howard et al., 1995; Sanders and Windom, 1980). The major part of the arsenate absorbed by the algae is transformed in arsenite As(III) (Andreae and Klumpp, 1979; Howard et al., 1995)(Andreae and Klumpp, 1979; Howard et al., 1995; Sanders and Windom, 1980) and then stocked in the brown algae in the form of nontoxic arsenosugars (Francesconi and Edmonds, 1996). Experiments conducted with caged *Sargassum* suggest a rapid release of As (Chapitre III). However, the speciation of As released by *Sargassum* is not known and this form could be
 non-bioavailable explaining the absence of increased As in organisms adjacent to *Sargassum* accumulations.

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366 Conclusion and perspectives.

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To avoid this transfer of As from *Sargassum* spp. to coastal environment, dams can be used to deviate macroalgae or stop them before stranding. Dams must be placed as far away from coast as possible and trapped algae must be collected rapidly. In our study, the As is the metalloid the more fastest released element with a decreasing contamination. Among the element above LOD, Al and Fe have a bell-shaped kinetics contamination whereas element (Zn) present an increasing contamination. Phenolics compounds and reveals the algal stress during the experiment"

Limited release of As by Sargassum implies that collected algae will presents a high As 375 376 concentration that must be considerate for their further valorization. The use of Sargassum as fertilizers (Milledge and Harvey, 2016) represent a potential risk of contamination of 377 378 agricultural lands. Public health could potentially be impacted when Sargassum are used as food for cattle or as drugs (Velasco-González et al., 2013) and as textiles and papers (Oyesiku 379 and Egunyomi, 2014). High content of As would present a limited risk when Sargassum spp. 380 381 are used to produce biogas (López Miranda et al., 2021) constituting the less risk valorization solutions. 382

The release of As by stranded *Sargassum* has already been shown to increase As contamination of coastal organisms representing a risk for seafood consumers (Cipolloni et al., n.d.). The present study reveals that this transfer from *Sargassum* is rapid in all coastal zone and must be considered when managing *Sargassum* inundation event.

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B)



Figure 1 Sampling site and experiments setting up. **Figure 1.A** Location of the sampling sites experimentation: CR (coral reef) (8°02'N;-47°07W), H (seagrass meadow) (6°87'N;-34°42'W), M (mangrove) (6°98'N; -31°80'W) of the prototype experimentation during July 2020. **Figure 1.B** Floating cages containing *Sargassum* recovered at Day=1, 4, 11, 18 and 25).

| | | Al | As | Fe | Cu | Zn |
|-------------------------------|--------|----------------------|----------------------|----------------------|----------------------|---------------------|
| Control | Day=0 | 123+2.0 | 74,0+0,1 | 101+3.1 | 2.2+0.06 | 5.1+0.06 |
| Mangrove | Day=1 | 251,6 <u>+</u> 24,2 | 74,9±0,3 | 581, 5 <u>+</u> 17,6 | 2,5±0,1 | 21,03±0,06 |
| | Day=4 | 1279 <u>+</u> 77,9 | 67,8 <u>±</u> 0,7 | 1424,5 <u>+</u> 86,5 | 4,6 <u>±</u> 0,1 | 32,2 <u>+</u> 0,4 |
| | Day=11 | 1089,5 <u>+</u> 20 | 53 <u>±</u> 0,7 | 1068,9 <u>+</u> 15,9 | 4,5 <u>±</u> 0,05 | 47,8 <u>±</u> 0,5 |
| | Day=18 | 1180 <u>+</u> 22,5 | 50,9 <u>+</u> 0,0 | 869,5 <u>+</u> 13,5 | 4,5 <u>±</u> 0,06 | 60,9 <u>+</u> 0,7 |
| 14 | Day=25 | NA | 48 <u>+</u> 0,08 | 1518,7±76,1 | $6,03\pm0,1$ | 98,1±0,2 |
| Mean | | 784,6 | 61,47 | 1112,8 | 4,9 | 53,09 |
| Spagrass | Dav=1 | 440 6+19 4 | 70 5+1 07 | 680 4+9 01 | 3 2+0 12 | 217+02 |
| Scugruss | Day=4 | 5011+255 | 64 6+1 1 | 4381+281 | 2,2+0,12 2,6+0,02 | 12.9+0.3 |
| | Day=11 | 781.05 ± 8.9 | 50.7 ± 0.02 | 612.9+4.2 | 2,9+0.02 | 26.6 ± 0.09 |
| | Dav=18 | 890.2+10.00 | 39.02 ± 0.2 | 549.7+3.9 | 2.7 ± 0.08 | 30.3 ± 0.5 |
| | Day=25 | NA | $36,4\pm0,09$ | 711,80 <u>+</u> 5,8 | $2,6\pm0,03$ | $34,8\pm0,40$ |
| Mean | - | 653,2 | 52,2 | 598,8 | 2,84 | 25,3 |
| | | | | | | |
| Coral reef | Day=1 | 384,8 <u>+</u> 20,5 | 74,5 <u>±</u> 0,2 | 309, 6 <u>+</u> 15,6 | 2,1 <u>±</u> 0,03 | 9,3 <u>+</u> 0,1 |
| | Day=4 | 640,3 <u>±</u> 16,07 | 64,7 <u>±</u> 0,5 | 492,3 <u>+</u> 9,5 | 2,8 <u>+</u> 0,02 | 2,5 <u>+</u> 0,02 |
| | Day=11 | 763, 7 <u>+</u> 14,6 | 44,9 <u>+</u> 0,5 | 486,1 <u>+</u> 12,8 | 2,9 <u>+</u> 0,06 | 23,2 <u>+</u> 0,3 |
| | Day=18 | 1072,4 <u>+</u> 28,1 | 40,00 <u>±</u> 0,1 | 656,5 <u>+</u> 18,7 | 2,9 <u>±</u> 0,08 | 35,5 <u>+</u> 0,2 |
| | Day=25 | NA | 37,08±0,3 | 998,9 <u>+</u> 4,3 | $2,8\pm0,13$ | 31,4 <u>+</u> 2,2 |
| Mean | | 715,3 | 52,2 | 588,7 | 2,7 | 20,4 |
| IAE 4412 naconam nata (0/ SD) | | | 101 20 ±4.66 | 06 76±11 20 | | 07 66 17 22 |
| DOLT 5 manual (%SD) | | | 101,23 - 7,00 | 01.21 ± 24.59 | 100.12 ± 0.61 | 00.01 ± 2.26 |
| DULI-5 recovery rate (%SD) | | | 99,93 <u>+</u> 0,58 | 91,21 <u>+</u> 24,59 | 100,12 <u>±</u> 0,61 | 99,91 <u>+</u> 2,36 |
| TORT-3 recovery rate (%SD) | | | 111,67 <u>±</u> 0,21 | 84,15 <u>+</u> 3,33 | | 96,36 <u>+</u> 0,91 |
| | | | | | | |

Table 1. Elements concentration (ppm = $\mu g. g^{-1}$) of pelagic *Sargassum* spp. collected from the three habitats (coral reef, seagrass, mangrove) Ocean to the Lesser Antilles (Guadeloupe – French West Indies) with their respective standard error (standard deviation divided by the squared root of the number of data), and the average in bold. Below the table there are recovery rates obtained from the analyses of certified reference material (TORT-3, DOLT-5, IAEA 413, and IAEA 407).





D0 D1 D4 D11 D18 D25

Seagrass

Sn8

D0 D1 D4 D11 D18 D25

Mangrove



D0 D1 D4 D11 D18 D25

Coral reef

0

D0 D1 D4 D11 D18 D25

Mangrove

D0 D1 D4 D11 D18 D25

Seagrass

Sn8

B)

D0 D1

D4 D11 D18 D25

Coral reef

D0 D1 D4 D11 D18 D25

Coral reef

D0 D1 D4 D11 D18 D25

Coral reef



Fig 3. Temporal variability of metal concentrations. Concentrations in Al, As, Cu, Fe and Zn ($\mu g. g^{-1}$) in *Sargassum* algae collected from the three different habitats (coral reef, seagrass and mangrove) for the three morphotypes of *Sargassum* spp. (*S. fluitans* III (Sf3); *S. natans* I (Sn1) and *S. natans* VIII (Sn8)) during twenty-five days (Days 0, 1, 4, 11, 18 and 25).



B)



Fig 4. Principal Component Analyses (PCA) showing variables (Fig.4 A) and Temporal Principal Component Analyses (Fig.4 B). F1 (38.82%) and F2 (13.17%) represents the relationship in *Sargassum* sp. between all the metallic elements (As; Ni; Ba; Cd; Cr; Zn; Cu; Mn, Fe; Al and V), the isotopic signature (δ 15*N* and δ 13*C*) and the phenolic content (PC) (Fig.4 A). Symbols shapes represent different coastal environments (coral reef, seagrass and mangrove) and colors represent different days (0,1, 4,11, 18 and 25).











Fig (supplement). Isotopic signature for $\delta 15N$ (A) and for $\delta 13C$ (B) of *Sargassum* algae collected from the three different habitats (coral reef, seagrass, mangrove) for the three morphotypes of *Sargassum* spp. (*S. fluitans* III (SF3); *S. natans* I (SN1) ans *S. natans* VIII (SN8)) during twenty-five days (0, 1, 4, 11, 18 and 25).

A)

| | | Al | As | Fe | Cu | Zn |
|--------------------------------|--------|----------------------|----------------------|----------------------|----------------------|---------------------|
| Control | Day=0 | 123+2.0 | 74,0+0,1 | 101+3.1 | 2.2+0.06 | 5.1+0.06 |
| Mangrove | Day=1 | 251,6 <u>+</u> 24,2 | 74,9±0,3 | 581, 5 <u>+</u> 17,6 | 2,5±0,1 | 21,03±0,06 |
| | Day=4 | 1279 <u>+</u> 77,9 | 67,8 <u>±</u> 0,7 | 1424,5 <u>+</u> 86,5 | 4,6 <u>±</u> 0,1 | 32,2 <u>+</u> 0,4 |
| | Day=11 | 1089,5 <u>+</u> 20 | 53 <u>±</u> 0,7 | 1068,9 <u>+</u> 15,9 | 4,5 <u>±</u> 0,05 | 47,8 <u>±</u> 0,5 |
| | Day=18 | 1180 <u>+</u> 22,5 | 50,9 <u>+</u> 0,0 | 869,5 <u>+</u> 13,5 | 4,5 <u>+</u> 0,06 | 60,9 <u>+</u> 0,7 |
| 14 | Day=25 | NA | 48 <u>+</u> 0,08 | 1518,7±76,1 | $6,03\pm0,1$ | 98,1±0,2 |
| Mean | | 784,6 | 61,47 | 1112,8 | 4,9 | 53,09 |
| Spagrass | Dav=1 | 440 6+19 4 | 70 5+1 07 | 680 4+9 01 | 3 2+0 12 | 217+02 |
| Scugruss | Day=4 | 5011+255 | 64 6+1 1 | 4381+281 | 2,2+0,12 2,6+0,02 | 12.9+0.3 |
| | Day=11 | 781.05 ± 8.9 | 50.7 ± 0.02 | 612.9+4.2 | 2,9+0.02 | 26.6 ± 0.09 |
| | Dav=18 | 890.2+10.00 | 39.02 ± 0.2 | 549.7+3.9 | 2.7 ± 0.08 | 30.3 ± 0.5 |
| | Day=25 | NA | $36,4\pm0,09$ | 711,80 <u>+</u> 5,8 | $2,6\pm0,03$ | $34,8\pm0,40$ |
| Mean | - | 653,2 | 52,2 | 598,8 | 2,84 | 25,3 |
| | | | | | | |
| Coral reef | Day=1 | 384,8 <u>+</u> 20,5 | 74,5 <u>±</u> 0,2 | 309, 6 <u>+</u> 15,6 | 2,1 <u>±</u> 0,03 | 9,3 <u>+</u> 0,1 |
| | Day=4 | 640,3 <u>±</u> 16,07 | 64,7 <u>±</u> 0,5 | 492,3 <u>+</u> 9,5 | 2,8 <u>+</u> 0,02 | 2,5 <u>+</u> 0,02 |
| | Day=11 | 763, 7 <u>+</u> 14,6 | 44,9 <u>+</u> 0,5 | 486,1 <u>+</u> 12,8 | 2,9 <u>+</u> 0,06 | 23,2 <u>+</u> 0,3 |
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| Mean | | 715,3 | 52,2 | 588,7 | 2,7 | 20,4 |
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