



# 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(loïd) 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  $\mu\text{g g}^{-1}$  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

## Introduction

The *Sargassum* genus includes more than 350 species, constituting one of the most diverse genera of brown macroalgae (Guiry and Guiry 2022). Among this genus, only two species are

holopelagic as they drift during their entire life cycle (Dawes and Mathieson 2008) constituting floating rafts called “the golden floating rainforest of the Atlantic Ocean” (Laffoley et al. 2011). Morphological and molecular studies differentiated three genotypes: *S. fluitans* III and *S. natans* I and VIII (Amaral-Zettler et al. 2017).

Historically, holopelagic *Sargassum* spp. were present in the Caribbean Sea, at the edge of the Gulf Mexico and the Azores islands (Lapointe 1995). In summer 2011, unprecedented 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 8850-km length, since then, this Great Atlantic Sargasso belt is reported annually in the North Equatorial Recirculation Region (NERR) (Wang et al. 2019).

The origins of the sudden and recurring increased of *Sargassum* abundance still remain unclear (Arduin

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et al. 2019), and different hypotheses are proposed such as an increase in (i) sea surface temperature (Sissini et al. 2017), (ii) nutrients released from Amazon and Congo rivers (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 (Cipolloni et al. n.d.; Rodríguez-Martínez et al. 2019) including endangered species such as 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). Decomposition of abundant brown algae biomass accumulated in coastal environment liberates toxic hydrogen sulfide (H<sub>2</sub>S) (Reiffenstein et al. 1992) provoking important human health issues such as respiratory diseases, neurological problems, and cardiovascular lesions (Resiere et al. 2018). *Sargassum* also represent an economic cost deterring tourism and obstructing free 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 metal and metalloid 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 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, represents 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.

The aim of the present study was thus to experimentally determine the kinetics of accumulation or depuration of 19 MTE during 25 days in three morphotypes of holopelagic *Sargassum* arriving in three different coastal environments: (i) coral reef, (ii) seagrass meadow, and (iii) mangrove.

## Materials and methods

### Study sites and experimental setting-up construction

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

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 (PCSM) were rapidly (less than one hour) placed in experimental device. Control sample ( $n=3$ ) at the beginning of incubation ( $t=0$ ) was collected, of each species. A fixed fresh weight of approximately 60g of *Sargassum* of the mixed three morphotypes (*S. fluitans* III and *S. natans* I and *S. natans* VIII) was separated morphologically at the experimental device stations and placed in each cage (Fig. 1B). At different temporal intervals (days 1, 4, 11, 18, and 25), macroalgae contained in each cage were simultaneously sampled in each habitat (coral reef, 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 during the experiment and one sample was missing due to disappearance of *S. natans* VIII in the last sampling cage in mangrove.

### Laboratory analyses

#### 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 3 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.

#### Phenolic compound analysis

Phenolic compounds were extracted twice using 15 mg DW of algal powder in 1 mL of 70% ethanol according to a



**Fig. 1** Sampling site and experiments setting up. **A** Location of the sampling sites experimentation: CR (coral reef) (8°02'N; -47°07'W), H (seagrass meadow) (6°87'N; -34°42'W), M (mangrove) (6°98'N;

-31°80'W) of the prototype experimentation during July 2020. **B** Floating cages containing *Sargassum* recovered at day=1, 4, 11, 18, and 25)

modified method from Zubia et al. (2009). The extractions were carried out with an ultrasonic bath (Sonicator 88155, Fisher 160 Bioblock Scientific, France) during 15 min at 4 °C followed by 2 h at 40 °C under magnetic stirring. Then, samples were centrifuged for 10 min at 8000 rpm (Eppendorf Centrifuge 162 5810, Germany) and supernatants were pooled and evaporated at 40 °C using a centrifugal concentrator (miVac, Genevac, France). Total phenolic content (TPC) was determined using the Folin-Ciocalteu colorimetric assay modified from Zubia et al. (2009). Thus, 20  $\mu\text{L}$  of sample was added to 130  $\mu\text{L}$  of distilled water, 10  $\mu\text{L}$  of Folin-Ciocalteu reagent, and 40  $\mu\text{L}$  of sodium carbonate ( $\text{Na}_2\text{CO}_3$ , 200  $\text{g L}^{-1}$ ). Then microplates

were incubated for 10 min at 70 °C before absorbance reading in triplicate at 620 nm (Multiskan FC, Thermo Scientific, USA). TPC was determined using a standard curve of phloroglucinol (1,3,5-trihydroxybenzene) and expressed in milligrams per gram of the dried seaweed powder ( $\text{mg g}^{-1}$  DW) and in percentage of TPC against day 0 level to see the evolution of *Sargassum* phenolic content during the experiment.

### Isotope analysis and calculation

The  $\delta^{15}\text{N}$  and the  $\delta^{13}\text{C}$  isotopic compositions of each *Sargassum* samples (*S. fluitans* III and *S. natans* I and *S. natans*

VIII) from the experimental devices were measured by EA-IRMS (elemental analysis–isotope ratio mass spectrometry) (Narancic et al. 2017). The isotope compositions were expressed as  $\delta$  – values relative to reference standard in per mil (‰) such as nitrogen composition is expressed in delta notation as:

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

### Metal(loid) trace element analysis

A series of 19 elements (Ag, Al, As, Ba, Cd, Co, Ca, Cr, Cu, Fe, Gd, Mn, Mo, Ni, Pb, Se, Sr, V, and Zn) were analyzed using an inductively coupled plasma optical emission spectrometer (Spectrometer ICP-OES 700®, Agilent Technologies). Certified reference materials DOLT-5 (dogfish (*Squalus acanthias*) liver), TORT-3 (Lobster Hepatopancreas), and IAEA-413 (Algae) were analyzed using ICP-OES, and their recovery rates vary between  $84.1 \pm 3.33$  and  $111.6 \pm 0.21$  (Table 1). For the values below the instrument detection limit, theoretical minimum concentration

values are calculated (the detection limit of the instrument (in  $\mu\text{g g}^{-1}$ ) multiplied by 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 ( $\text{HNO}_3$  67%). The powdered sample was then mineralized for 3 h at  $100^\circ\text{C}$  (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  $\mu\text{g g}^{-1}$  (ppm) dry weight.

### Data analysis

The variance and the homogeneity of metals and metalloid concentration values were verified 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 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 RCrn, using the following

**Table 1** Element concentration (ppm =  $\mu\text{g g}^{-1}$ ) 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)

		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±24.2	74.9±0.3	581.5±17.6	2.5±0.1	21.03±0.06
	Day=4	1279±77.9	67.8±0.7	1424.5±86.5	4.6±0.1	32.2±0.4
	Day=11	1089.5±20	53±0.7	1068.9±15.9	4.5±0.05	47.8±0.5
	Day=18	1180±22.5	50.9±0.0	869.5±13.5	4.5±0.06	60.9±0.7
	Day=25	NA	48±0.08	1518.7±76.1	6.03±0.1	98.1±0.2
Mean		<b>784.6</b>	<b>61.47</b>	<b>1112.8</b>	<b>4.9</b>	<b>53.09</b>
Seagrass	Day=1	440.6±19.4	70.5±1.07	680.4±9.01	3.2±0.12	21.7±0.2
	Day=4	501.1±25.5	64.6±1.1	438.1±28.1	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
	Day=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±0.09	711.80±5.8	2.6±0.03	34.8±0.40
Mean		<b>653.2</b>	<b>52.2</b>	<b>598.8</b>	<b>2.84</b>	<b>25.3</b>
Coral reef	Day=1	384.8±20.5	74.5±0.2	309.6±15.6	2.1±0.03	9.3±0.1
	Day=4	640.3±16.07	64.7±0.5	492.3±9.5	2.8±0.02	2.5±0.02
	Day=11	763.7±14.6	44.9±0.5	486.1±12.8	2.9±0.06	23.2±0.3
	Day=18	1072.4±28.1	40.00±0.1	656.5±18.7	2.9±0.08	35.5±0.2
	Day=25	NA	37.08±0.3	998.9±4.3	2.8±0.13	31.4±2.2
Mean		<b>715.3</b>	<b>52.2</b>	<b>588.7</b>	<b>2.7</b>	<b>20.4</b>
IAEA413 recovery rate (%SD)			101.29 ±4.66	96.76±44.39		97.66±7.33
DOLT-5 recovery rate (%SD)			99.93±0.58	91.21±24.59	100.12±0.61	99.91±2.36
TORT-3 recovery rate (%SD)			111.67±0.21	84.15±3.33		96.36±0.91

packages: FactoMiner (Husson et al., 2020), factoextra (Kassambara and 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, 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.

## Results

### Indicators of the physiological condition of *Sargassum*: biomass, phenolic contents, and isotopic signatures

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

In the three habitats, phenolic contents of each morphotype also follow similar kinetics with two distinct phases: (i) a first phase of decrease in phenolic content compounds until the 11th days and (ii) a second phase with an increase in phenolic content from the 18th day to the end of the experiment (Fig. 2). The phenolic content in  $\mu\text{g g}^{-1}$  was higher in the morphotype *S. fluitans* III (25 and  $30 \mu\text{g g}^{-1}$ ) than in *S. natans* I (10 and  $20 \mu\text{g g}^{-1}$ ) and *S. natans* VIII (10 and  $12 \mu\text{g g}^{-1}$ ).

Isotopic compositions ( $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$ ) of *Sargassum* were not clearly different between all habitats and morphotypes. The values of the isotopic signature in the three habitats, for the three genotypes, remain around 3–4‰ for the  $\delta^{15}\text{N}$  and 14–18‰ for the  $\delta^{13}\text{C}$ .

### Metal and metalloid content

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 of the samples analyzed.

Kinetics analysis was 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).

## Principal component analysis (PCA)

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. 4A). F1 distinctly discriminates the variables Al (13.37%), Fe (14.45%), Zn (14.08%), and Cu (15.25%), whereas F2 clearly discriminates As (11.65%).

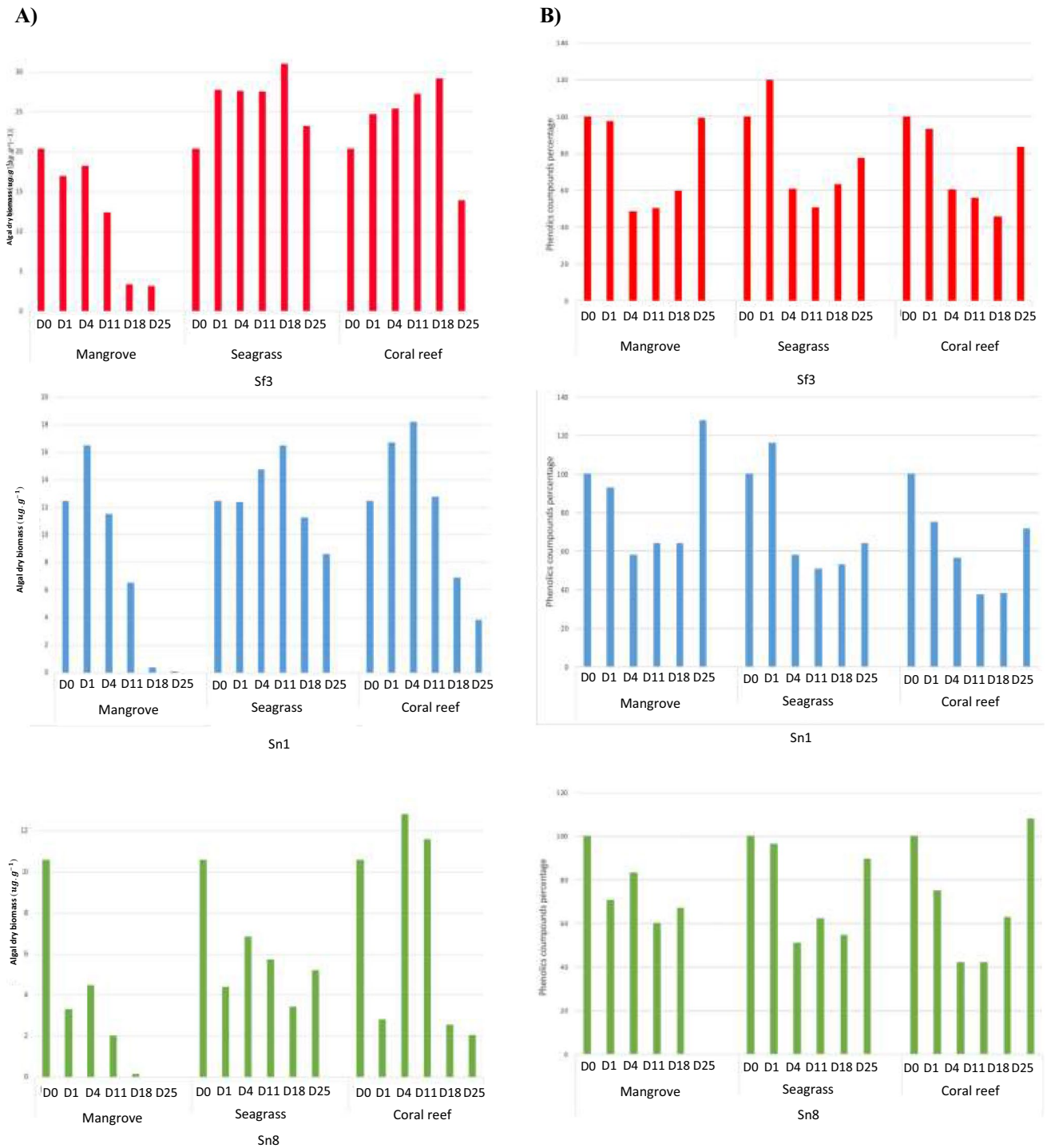
The PCA analysis of the sample discriminates the mangrove habitat characterized by high concentrations in Fe, Al, Zn, Cu, and As in *Sargassum* (Fig. 4B) whereas samples from seagrass meadow and coral reef were similar with high concentrations of Ni, V, Cd, and Cr. PCA discrimination according to *Sargassum* by the day (control, day=1, day=4, day=11, day=18, 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 (Fig. 4B). On the PCA analysis, whatever the habitat and the experiment duration, each of the three morphotypes followed similar trend (Fig. 4B).

## Discussion

The present study measured the kinetics of metal(oid)s trace element 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, and (iii) morphotype due to the singularity of the genotype *S. natans* I.

In our sample, entire macroalgal thalli were dried and ground before the analyses 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 much 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. Mangroves are consequently classically more influenced by freshwater and organic matter inputs than the two other habitats.



**Fig. 2** Algal dry biomass in  $\mu\text{g g}^{-1}$  (A) and phenolic content (%) (B) of *Sargassum* algae collected from the three different habitats (coral reef, seagrass, and mangrove) for the three morphotypes of *Sargas-*

*sum* sp. (*S. fluitans* III (SF3), *S. natans* I (SN1), and *S. natans* VIII (SN8)) during 25 days (0, 1, 4, 11, 18, and 25)

A similar evolution of algal biomasses was generally observed in all samples whatever the morphotype and habitat with a first increase in biomass followed by a decrease. This decrease is likely due to the degradation of

seaweeds indicating unsuitable conditions. Physiological or stress conditions of algae were also evaluated measuring phenolics contents (Plouguerné et al. 2006). Phenolic compounds of brown seaweeds or phlorotannins are

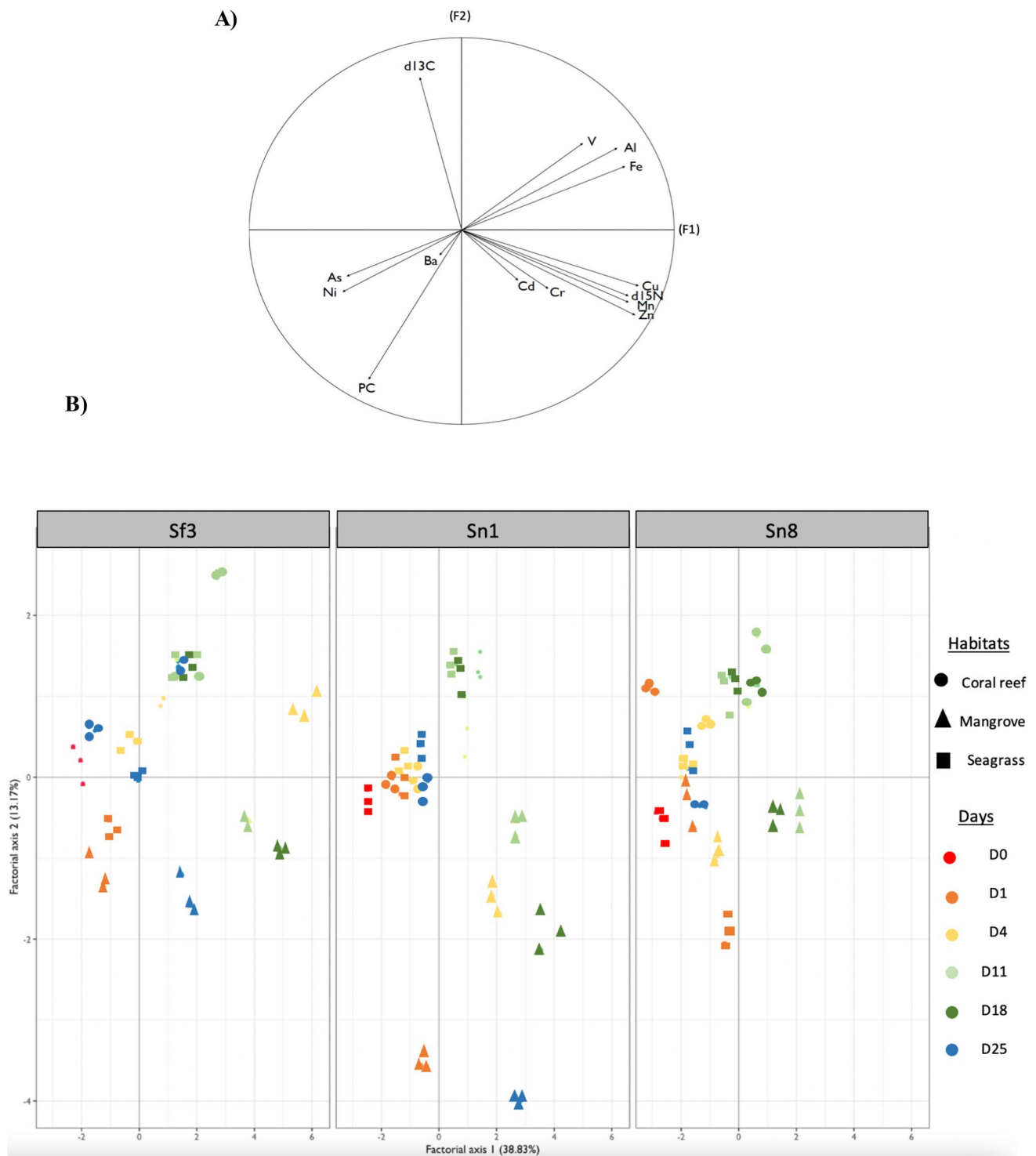


**Fig. 3** Temporal variability of metal concentrations. Concentrations in Al, As, Cu, Fe, and Zn ( $\mu\text{g g}^{-1}$ ) in *Sargassum* algae collected from the three different habitats (coral reef, seagrass, and mangrove)

for the three morphotypes of *Sargassum* spp. (*S. fluitans* III (Sf3), *S. natans* I (Sn1), and *S. natans* VIII (Sn8)) during 25 days (days 0, 1, 4, 11, 18, and 25)

present sometimes in high level, between 10 and 120% in the cell walls and the physodes and as there are produced in response to any changes in abiotic (temperature, light) and biotic (grazing, fouling) factor, their content may be used to evaluate algal stress (Arnold and Targett 2002; Ragan and Craigie 1976; Schoenwaelder and Clayton 1999). Seaweed samples were oven-dried at  $50^{\circ}\text{C}$  and this process can alter phenolics compounds (Gager et al. 2021); however, a similar drying method was applied for all samples allowing inter-sample comparisons in the evolution of the phenolic 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 phenolic compounds between the day 0 (day=D0) and the day 4 (day=D4). During this initial phase, the stress of algae would be limited and their biomass would increase. During the second phase, the content of phenolic compounds in algae increased, between the day 4 (day=D4) and the day 25 (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 degraded cell walls were still within the thalli, and in proportion, their content expressed in  $\mu\text{g g}^{-1}$  algal dw increased (Koivikko et al. 2005; Schoenwaelder and Clayton 1999). The temporal variation in phenolic contents therefore suggests that the algae are first in good condition, then stressed, leading to the degradation of the algae and its death. Evolution of isotopic composition ( $\delta^{15}\text{N}$  and  $\delta^{13}\text{C}$ ) of algae during the experiments can reflect the uptake of C and N from the environment and/or the preferential disappearance of isotope form during 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 *Sargassum* in this type of experiment. The evolution of biomass and phenolic compounds, with the use of triplicates, both suggest that environmental conditions in cages are stressful for *Sargassum* and conduct to the death of algae during the experiments. A similar laboratory experience with *Sargassum* in mesocosm bags during 26h showed a rapid degradation of the macroalgae (Devault et al. 2021) which is in agreement with our study.



**Fig. 4** Principal component analyses (PCA) showing variables (**A**) and temporal principal component analyses (**B**). F1 (38.82%) and F2 (13.17%) represent 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 ( $\delta^{15}N$  and  $\delta^{13}C$ ), and the phenolic content (PC) (**A**). Symbol shapes represent different coastal environments (coral reef, seagrass, and mangrove) and colors represent different days (0, 1, 4, 11, 18, and 25)

Initial metal and metalloid 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 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 enter in algae using phosphate transporter pathway (Gobert et al. 2022). In order to tolerate 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 specific distribution of As could be an explanation of the rapid release of As counterbalanced 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 concentrations in *Sargassum* previously observed in experimental (Al Mamun et al. 2019) and in situ conditions (Cipolloni et al. 2022).

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 induces hypoxic 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 by the carboxylate group within alginates (cell wall polysaccharides) of the algae presenting an 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 previously suggested in the present study.

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

Due to higher proximity with terrestrial environment and high primary production, the 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 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 implying a release of this compounds by *Sargassum*. In accordance with this hypothesis, *Sargassum* were previously observed depurating As due to competitive exchange with terrigenous metals (Gobert et al. 2022). Salinity variations are more important in the mangrove than in other habitat due to mainland proximity and decreased salinity-specific physiological and morphological processes of mangrove

organisms (Clough et al. 1989; Feller et al. 2010). As OM, salinity could leads to the formation of stable metal-chloride complexes decreasing the availability of metallic elements (Mader et al. 1995). In mangrove, the decreased salinity and increased content of organic matter have opposite effects on metallic element complexation. The observed releasing activity of metalloids (As) by *Sargassum* in mangrove suggests that OM is more structuring than salinity and reduces metal availability in this environment.

One morphotype, *S. natans* I, stands out of PCA analyses in the mangrove. In this environment, kinetics of As and Zn contents were faster in *S. natans* I than for the two other morphotypes (*S. 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 be due to its morphology particularity as *S. natans* I presenting a more complex structure with higher exchange surface favoring fixation or release of pollutants (Khotimchenko et al. 2001). Compared to other morphotypes, *S. natans* I also present a specific chemical composition, and *S. natans* I appears to be significantly more enriched in P compared to the other morphotypes (Gobert et al. 2022). The ability of *S. natans* I to absorb pollutants may notably be due to its alginates which may have a different structure compared to the two other *Sargassum* genotypes as length alginates limit the retention of some cations such as metals (Rhein-Knudsen et al. 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 complex biogeochemistry (Fattorini et al. 2006; Neff 1997). Arsenic concentrations obtained 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; Rodríguez-Martínez et al. 2020) and off-shore environments with values a mean of 140 ppm (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 that, once arrived in coastal environment, *Sargassum* rapidly release their As and this 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.

## Conclusion and perspectives

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 the element Zn presents an increasing contamination. Phenolic compounds reveal the algal stress during the experiment.

Limited release of As by *Sargassum* implies that collected algae will presents a high As concentration that must be considerate for their further valorization. The use of *Sargassum* as fertilizers (Milledge and Harvey 2016) represents a potential risk of contamination of 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 and Egunyomi 2014). High content of As would present a limited risk when *Sargassum* spp. are used to produce biogas (López Miranda et al. 2021) constituting the less risk valorization solutions.

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 zones and must be considered when managing *Sargassum* inundation event.

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## Declarations

**Ethical approval** This manuscript is an original work and has not been previously published somewhere else not submitted to more than one journal for simultaneous consideration.

**Consent to participate** Not applicable

**Consent for publication** The authors consented.

**Competing interests** The authors declare no competing interests.

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