Element concentrations in pelagic *Sargassum* along the Mexican Caribbean coast

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Massive influx of pelagic *Sargassum* spp. (sargasso) into the Caribbean since 2011 has caused major environmental problems, threatening the tourist industry and livelihoods. Industrial uses of sargasso are essential for a sustainable management and to partially compensate for the cost of harvesting and beach cleaning. In this context, we present the concentrations of 28 different elements in 63 samples containing three different sargasso morphotypes collected at eight localities along the coastal waters of the Mexican Caribbean Sea between August 2018 and June 2019. The sargasso tissue contains different concentrations of Al, As, Ca, Cl, Cu, Fe, K, Mg, Mn, Mo, P, Pb, Rb, S, Si, Sr, Th, U, V, and Zn. High variability in elemental concentration was found across sites and between morphotypes. No temporal patterns and lack of any tendency in the element concentrations were found among samples collected in seven different sampling periods at one location. This variation might be due to the pelagic nature of sargasso and different degrees of exposure to contaminants in the ocean. Total arsenic concentrations (24-172 ppm DW) exceed the established safe limits for agricultural soils in 88% of the samples. Considering the high and unpredictable variability of metals, we recommend the regular analyses of sargasso tissues as a mandatory practice to ensure a safe use and management and to mitigate the risks to coastal environments by leaching of heavy metals to the sea and aquifer.

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Abstract

Massive influx of pelagic *Sargassum* spp. (sargasso) into the Caribbean since 2011 has caused major environmental problems, threatening the tourist industry and livelihoods. Industrial uses of sargasso are essential for a sustainable management and to partially compensate for the cost of harvesting and beach cleaning. In this context, we present the concentrations of 28 different elements in 63 samples containing three different sargasso morphotypes collected at eight localities along the coastal waters of the Mexican Caribbean Sea between August 2018 and June 2019. The sargasso tissue contains different concentrations of Al, As, Ca, Cl, Cu, Fe, K, Mg, Mn, Mo, P, Pb, Rb, S, Si, Sr, Th, U, V, and Zn. High variability in elemental concentration was found across sites and between morphotypes. No temporal patterns and lack of any tendency in the element concentrations were found among samples collected in seven different sampling periods at one location. This variation might be due to the pelagic nature of sargasso and different degrees of exposure to contaminants in the ocean. Total arsenic concentrations (24-172 ppm DW) exceed the established safe limits for agricultural soils in 88% of the samples. Considering the high and unpredictable variability of metals, we recommend the regular analyses of sargasso tissues as a mandatory practice to ensure a safe use and management and to mitigate the risks to coastal environments by leaching of heavy metals to the sea and aquifer.

1. Introduction

Unusual large quantities of pelagic *Sargassum* spp. (*S. fluitans* (Boergesen) Boergesen and *S. natans* (Linnaeus) Gallion; hereafter named sargasso) started to arrive in 2011 at the west coast of Africa and to the Eastern Caribbean islands (Gower, Young & King, 2013). The distribution range and abundance of sargasso has increased since then in the Atlantic Ocean and Caribbean

Sea. Wang et al. (2019) reported over 20 million metric tons of sargasso in the open ocean when the Great Atlantic Sargasso Belt extended for 8,850 km in June 2018. Its beaching has been causing havoc to the Caribbean coastal ecosystems as leachates and particulate organic matter from the stranded decaying algal masses lead to mortality of the nearshore seagrasses and fauna due to depleted oxygen, reduced light and deteriorated water quality (van Tussenbroek et al., 2017; Rodríguez-Martínez et al., 2019). Accumulation of sargasso can also interfere with seaward journeys of juvenile turtles (Maurer, De Neef & Stapleton, 2015) and sea turtle nesting ecology (Maurer, Stapleton & Layman, 2018), enhance the process of beach erosion (van Tussenbroek et al., 2017) and change the trophic dynamics of benthic organisms (Cabanillas-Terán et al., 2019). Major income of the Caribbean countries comes from the coastal ecosystem-based tourist industry (Langin, 2018). The potential socio-economic consequences of ecosystem degradation and their effects on livelihoods in this region, however, are yet to be assessed.

The Mexican Caribbean coast began receiving massive amounts of sargasso in late 2014 and it reached a peak in September 2015. An average ~2,360 m^3 of algae (mixed with sand, other algae or seagrasses) was removed per km of shoreline from tourist beaches by hotels in the northern section of the coast between Cancun and Puerto Morelos (Rodríguez-Martínez, van Tussenbroek & Jordán-Dahlgren, 2016). After a phase of minor decrease in the sargasso influx between 2016-2017, a large influx in May 2018 resulted in the removal of ~8,793 m^3 km^{-1} of algae (mixed with sand, other algae or seagrasses) at the same shore section (Rodríguez-Martínez et al., 2019). Even though there have been efforts to remove these algae from the beach and sea, its continued influx has led to thousands of tons of sargasso accumulating annually along the Mexican Caribbean coast. Furthermore, the removed algae are transported inland and disposed in areas that are not properly prepared to avoid leakage of the leachates into the aquifer.

Species of *Sargassum* have a high capacity to biosorb metals and other elements, similar to other brown algae (Kuyucak & Volesky, 1988; Davis, Volesky & Vieira, 2000). This high absorption capacity is attributed to the unique mixture of polysaccharides, mainly alginate and fucoidan, in their cell walls (Fourest & Volesky, 1997). At present, *Sargassum* spp. are used for different commercial end products, such as fertilizers (Milledge & Harvey, 2016), textiles, paper and drugs (Oyesiku & Egunyomi, 2014) and the production of biogas (Wang et al., 2018). Macroalgae are increasingly used as a food source for animals and humans, and therefore potentially high concentrations of contaminants, including heavy metals amongst others, may pose health risks (Reis & Duarte, 2018). Thus, it is mandatory to evaluate concentrations of all the elements in order to recommend the potential uses of sargasso and to ensure that health regulations of acceptable levels are respected (Fourest & Volesky, 1997). Until present, there have been limited studies on metal contents in sargasso and all of them are based on limited number of samples collected mostly from a single locality (e.g. Nigeria - Oyesiku & Egunyomi, 2014; Dominican Republic - Fernández et al., 2017) or in a single season (Addico & deGraft-Johnson, 2016), and therefore the possible variability in the contents of metals in the algal tissues remains unclear.

In this study we estimate the concentrations of 28 different elements in sargasso collected from the Mexican Caribbean coast, covering a linear north-south distance of 370 km. We hypothesize that the elemental concentrations in sargasso are highly variable in time and space, because they depend on drifting variations at the surface of the ocean. The accurate determinations of metals and other elements presented in this study will provide an essential baseline data for adequate management and potential use of sargasso, as well as to modify the disposal strategies of decomposing sargasso to mitigate the environmental risks in near future.

2. Survey methodology

2.1 Study sites

We collected 63 samples of sargasso along the Mexican Caribbean coast, from the Contoy Island, at northern extreme, to Xcalak in the south (Fig. 1). This region receives an average precipitation of ca.1061 mm y^{-1} and the sea-surface temperature (SST) ranges from 25.1-29.9 °C (Rodríguez-Martínez et al., 2010). Easterly trade-winds dominate this region during the summer and mild cold fronts occur during the winter season. The trade-winds facilitate the transportation of superficial waters towards the shore, importing the pelagic masses of sargasso to the Mexican Caribbean coast. The Yucatán current, a major branch of the Caribbean Current, also transports the algal mass to the coast. The freshwater aquifer of this region and the seawater are constantly interacting with each other as basement geology of the Yucatan Peninsula is karstic and porous (Hernández-Terrones et al., 2011; Hernández-Terrones & Null, 2015).

The coastal environments in this region consist of beaches, rocky shores, seagrass beds, coral reefs, mangroves, jungle and underground rivers (Hernández-Arana et al., 2015). These ecosystems provide services that are crucial for the economy of the population which depends mainly on tourism (Spalding et al., 2017). Today, this coast has the highest number of hotel rooms in Mexico. The number of hotel rooms increased from 3,206 in 1975 to 100,986 in 2017 (SEDETUR, 2019), while the resident population grew from less than 100,000 in 1970 to 1,501,785 in 2015 (INEGI, 2015). This urban development, associated with the tourist industry, has resulted in coastal pollution through nutrients (Carruthers, van Tussenbroek & Dennison, 2005; Hernández-Terrones et al., 2011; Baker, Rodríguez-Martínez & Fogel, 2013; van Tussenbroek et al., 2017), sewage (Metcalfe et al., 2011), and some metals (e.g. Lead, see

Whelan, van Tussenbroek & Santos, 2011). This region has no other industries besides the tourism.

2.2. Methodology

2.2.1. Field collection

Samples were collected between August 2018 and June 2019 at eight sites along the Mexican Caribbean coast (from north to south): 1) Contoy Island, 2) Blue waters, 3) Puerto Morelos, 4) Cozumel, 5) Mahahual, 6) Chinchorro, 7) Xahuayxol and 8) Xcalak (Fig. 1, Table 1). We collected manually the fresh sargasso (golden color) thalli floating near the shore (2-20 m) and in the ocean (>5 km from shore) and subsequently separated them by species and morphotypes (*S. fluitans* III, *S. natans* I and *S. natans* VIII) in the laboratory following Schell, Goodwin & Siuda (2015), except for Contoy Island (CI). The samples collected from CI were frozen before separation of morphotypes and thus, we classified them as *Sargassum* spp. All the samples were placed in an oven for at least 48h at 60°C until completely dry. Special caution was taken to avoid contact between the algal samples and any metal object. Samples were shipped to the Institute of Geology of the National Autonomous University of Mexico for the analysis of element concentrations.

2.2.2. Elemental analysis

Concentrations of 28 different elements were measured in dry samples using a Niton FXL 950 energy dispersive X-ray fluorescence (XRF) containing a 50kV X-ray tube of Ag and equipped with a geometrically optimized large area drift defector following Quiroz-Jimenez & Roy

(2017). Table S1 shows the limit of detection of these elements in the Niton FXL. In the laboratory, the dried samples were processed using a non-destructive sample preparation technique and approximately 5-7 g of each sample was placed in a plastic capsule that has a 4 μm thick polypropylene X-ray film on one side and the other side of the capsule was packed with synthetic flexible gauze. Samples were measured in the mining Cu/Zn mode and three different filters using the internal calibration curves previously generated by comparing the results of Niton FXL with a conventional XRF (e.g. Quiroz-Jimenez & Roy, 2017). The results are expressed in parts per million dry weight (ppm DW) after carrying out the analysis in five repetitions in each sample. We used two different geological reference materials (Es-2, organic rich argillite and Es-4, dolostone) for estimation of precision (Kiipli et al., 2000). Except for Mg, all other elements have relative standard deviation (RSD) between <1 and 5%. Mg concentrations show RSD of 26% and it is the least precise among all the analyzed elements.

2.3. Data Analyses

We calculated the median of the five readings per element of each sample and used it for further analysis. For each element, the readings below the limit of detection ($<$LOD; Table S1) were substituted with LOD/$\sqrt{2}$ for calculation of summary statistics (Celo & Dabek-Zlotorzynska, 2010). Distributions (spread of data and the median values) of the fourteen most commonly found elements (e.g. Al, As, Ca, Cl, K, Mg, Mn, P, Rb, S, Si, Sr, Th and U) in sargasso tissue for each sampling locality are illustrated by dot plots. Statistical comparison in metals concentration among localities was not done due to small sample sizes per site. Differences in the concentration of elements among species and morphotypes were tested using non-parametric ANOVAs based on the Kruskal-Wallis rank procedure. We constructed a heatmap using the data from fourteen elements from Puerto Morelos (location 3, see Fig.1) to visualize temporal differences in concentration of metals in seven different sampling periods between August 2018 and April 2019. Element concentration values were Z-score-transformed across sampling times and their values above and below the mean were used to generate the heatmap. The Z-value is a dimensionless quantity which is defined by the following equation (Larsen & Marx, 1986):

$$Z = (X - \mu) / \sigma$$

Where X represents an individual raw score that is to be standardized, σ is the standard deviation of the population, and μ is the mean of the population.

All analyses were done in R (R Core Team, 2019) using packages: dplyr (Wickham et al., 2019), ggplot2 (Wickham, 2009), gplots (Warnes et al., 2009), pgirmess (Giraudoux, 2013), reshape (Wickham, 2018), tidyr (Wickham & Henry, 2016), and RColorBrewer (Neuwirth, 2011). A reproducible record of all statistical analyses is available on GitHub (https://github.com/rerodriguezmtz/ElementsSar). This includes all underlying data and R code for all analyses.

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3. Results

The most frequent elements in sargasso tissue, detected in 100% of the samples, were As, Ca, Cl, K, Mn, P, Rb, S, Si, Sr, Th, and U. They were followed in frequency by Mg (92.1% of samples) and Al (58.7% of samples) (Table 2). Other elements were found in fewer samples and had overall median concentrations below LOD: V (28.6% of samples), Zn (12.7% of samples), and

Cu, Fe, Mo and Pb, present in 7.9% of samples (Table 2). Concentrations of Ba, Cd, Co, Cr, Ni, Ti, Y, and Zr remained below the LOD in all samples measured (See Table S1 for LOD values). The range of concentrations (ppm DW) in some elements was considerably large (i.e. > 5-fold difference between minimal and maximal concentrations). For example, Cl showed 71.1-fold difference, K had a 23.1-fold difference, As had a 7.2-fold difference, Si showed a 6.5-fold difference and Ca exhibited a 5.7-fold difference (Table 2).

Inter-site variability in element concentrations was high, with P, S and Sr being the least variable elements and Al, As, Cl and K the most (Fig. 2). Of the potentially toxic elements, only As (median contents range among sites: 37-121 ppm DW) and Mn (40-139 ppm DW) were present in all the samples. About 88% of the total sample size, collected at seven sites (Fig. 2), had concentrations of As above the maximum allowable concentration under European regulations for animal feed (40 ppm DW; Monagail et al., 2018) and 100% of the samples were above the maximum allowable concentration for agricultural soils in Mexico (22 ppm DW; NOM-147-SEMARNAT-SSA1-2004). Among the total samples, 22% had Mn above 100 ppm DW and 5% showed Cu above 100 ppm DW. Other potentially toxic elements (e.g. Mo, Pb and Zn) were detected in only 8-13% of the samples and they had median concentrations (Table 2) below the toxic limits (see Supplementary Table 2).

Concentrations of As, Ca, Cl, K, Mn, Rb and Si varied significantly among sargasso species/morphotypes (Fig. 2, Kruskal-Wallis test, $p < 0.05$; Table 3). As, Cl, K and Rb were significantly higher in *S. natans* VIII compared to *S. natans* I. The concentrations of Ca and Si were significantly lower in *S. natans* VIII than in *S. fluitans* III and *S. natans* I. Similarly, the concentration of Mn was higher in *S. natans* I than in *S. fluitans* III and *S. natans* VIII (Table 3). Contents of Al, Mg, P, S, Sr, Th and U did not vary significantly among species and morphotypes (KW, $p > 0.05$; Table 3). We did not compare the concentrations of Cu, Fe, Mo, Pb and Zn statistically among the species/morphotypes as their medians remained <LOD.

The concentrations of fourteen different elements (i.e. Al, As, Ca, Cl, K, Mg, Mn, P, Rb, S, Si, Sr, Th and U) in sargasso collected at Puerto Morelos in seven different sampling periods, between August 2018 to April 2019, showed considerable variability in time (Fig. 3). This inconsistent pattern and lack of any tendency indicates absence of seasonal tendencies in elemental concentrations.

4. Discussion

Tissue contents of As, Cu and Mn in sargasso from Mexico were higher than those observed in previous studies in Nigeria, Ghana and Dominican Republic, while those of Cd, Cr, Pb and Zn were lower (Table 4). Most striking was the high variability of element concentrations detected both in the space (along the coast) and time (different sampling months). This variability may be partially attributed to pelagic nature of sargasso. In our study, it is unlikely that heavy metals were adsorbed nearshore, because 1) the residence time of these algae in near-shore waters before beaching is in the order hours, and 2) these elements are virtually absent in the near-shore waters of the Mexican Caribbean, as there are no industrial, heavy agricultural or mining activities in the region. Thus, the heavy and trace elements were likely acquired before entering the Mexican coastal waters. In different parts throughout the geographical region of the North Equatorial Recirculation Region of the Atlantic Ocean (NERR) and the Wider Caribbean Region, different contaminants are released into the ocean, some as point sources and others more continuous (as a result of long-range transport). Fernandez, Singh & Jaffe (2007) recognized discharge of sewage and inputs of mineral extraction, fertilizer and pesticide used in the agricultural and industrial sector as principal pollution sources of metals in the oceans. Whether sargasso was exposed to these contaminants depended on its trajectory in the ocean and whether (or not) the pelagic masses passed near areas with high natural (i.e. upwelling) or human-induced concentrations of metals; the biosorption of metals in turn may have depended on the pH of seawater (Davis et al., 2000). In addition, metal sequestration involves complex mechanisms of ion exchange, chelation, adsorption, and ion entrapment in polysaccharide networks of the algae (Volesky & Holan 1995). This ion entrapment, in turn, depends on the affinity of some divalent metals to alginates (Haug, 1961). Alginates are often characterized by the proportion of mannuronic (M) and guluronic (G) acids present in the polymer (M:G ratio), which may vary among species and with samples of the same species. For example, we found higher concentrations of Mn in *S. natans* I, of Ca and Si in *S. fluitans III* and *S. natans* I, and of As, Cl, K and Rb in *Sargassum natans* VIII than in *S. natans* I. Variable concentrations of Ca, Mg, Sr and U may be attributed to changes in calcifying epifauna associated with the sargasso, such as bryozoans, tube polychaeta, and crustose coralline algae (Weiss, 1968; Huffard et al., 2014). Similarly, the variable amounts of Si (447-2,922 ppm DW) were possibly contributed by different abundances of marine diatoms and silicoflagellates present in the sargasso samples (Takahashi & Blackwelder 1992).

Seaweeds, including various *Sargassum* species, are rich in essential nutrients and are often used as complementary fertilizers that enhance growth, seed germination and photosynthesis of crop plants on mineral-depleted soils (Sathya et al., 2010; Kumari, Kaur & Bhatnagar, 2013; El-Din, 2015). These essentials nutrients also contribute the value of sargasso as animal fodder (Marín et al., 2009; Carrillo et al., 2012). Amongst all the detected elements in sargasso samples along the Mexican Caribbean coast, Ca (23,723-136,146 ppm DW), K (1,990-46,002 ppm DW), Mg

(<2915-13,662 ppm DW), P (228-401 ppm DW) and S (9,462-24,773 ppm DW) are essential macronutrients for algae and higher plants. Other elements like Cu, Mn, Mo and Zn are essential micronutrients in low concentrations, but potentially toxic when present in high concentrations. Cu (<8-540 ppm DW) and Mo (<1-7 ppm DW) were present in 7.9% of the samples, Zn (<2-17 ppm DW) in 12.9 % of the samples and Mn (40-139 ppm DW) in all the samples. Of these, the concentrations of Cu exceeded safely limits recommended for agricultural soils by several countries (see Supplementary Table 2S) in 5% of the samples. For Mo, 8% of the samples contained above the maximum level established by Canada (i.e. 2 ppm DW) but all the samples had below the limits established by Austria and Poland (i.e. 10 ppm DW). However, 22% of the samples contained above 100 ppm DW of Mn. This value could be considered as toxic for some plant species, but others have tolerance up to 5000 ppm DW of Mn (Howe et al., 2004). Pb (<2-3 ppm DW) was present in 7.9% of the samples but it remained below the toxic levels in all of them.

However, in most sargasso samples, the concentrations of total As exceeded the safe limits established for agricultural soils (15-50 ppm DW depending on the country; Belmonte et al., 2010) and in 38% of samples was above the maximum tolerable levels of dietary minerals for domestic animals (100 ppm DW; Supplementary Table 3). Toxicity of As depends on its chemical form. The inorganic forms (trivalent state As III and pentavalent state As V) are considered toxic, while the organic ones (such as arsenosugars, arsenolipids and arsenobetaine) are not (Yuan, 2007; Circuncisão et al., 2018). Due to the higher total As content (24-172 ppm DW) in sargasso of the Mexican Caribbean coast, we suggest that As speciation studies must be conducted before employing sargasso for animal fodder. Arsenic may also be of some concern when sargasso is used as complementary fertilizer for ornamental and crop plants. High concentrations of As in soil may be toxic for the plants, as it interferes with photosynthesis and other metabolic processes (Påhlsson, 1989, Ruiz Huerta & Armienta Hernández, 2012). Higher level of As is a concern and its unpredictable variability poses further setback for the commercial uses of collected sargasso in the region. Presence of As is also of concern as sargasso removed from Mexican Caribbean beaches is deposited at abandoned limestone quarries near the coast without any treatment. The Yucatan Peninsula has a highly porous karst aquifer that is the only source of freshwater in the region. Without proper protection, the pollutants from the near surface deposits infiltrate into the aquifer and accumulation of As and potentially other toxic metals into the aquifer could threaten human health. Considering that water from the aquifer flows into the ocean through belowground rivers, the metals and excessive nutrients infiltrated into the aquifer will eventually reach the marine environment (Carruthers et al. 2005; Metcalfe et al. 2011; Baker et al. 2013).

Even if the concentration of some other potentially toxic elements were low in sargasso, their accumulation over time might be a potential source of contaminant in this region. The Mexican Caribbean coast has received millions of tons of this algae since late 2014. Prevention and mitigation measures are urgently needed to ensure that the massive arrival of sargasso does not continue to harm the coastal ecosystems, the tourism-based economy and general human well-being in countries affected by the Great Atlantic *Sargassum* belt, including the Mexican Caribbean. The high and inconsistent variability of some potentially toxic metals indicates that analyses of different specimens collected over longer periods, and/or in different locations are required in order to obtain reliable information about metal contents in tissues, even though it might need additional logistical and financial efforts.

Conclusion

The Mexican Caribbean coast has received millions of tons of sargasso since late 2014. Buildup of decomposing masses of sargasso on the shores has harmed the coastal ecosystems, the tourism-based economy and general human well-being in countries affected by the Great Atlantic Sargassum belt, including the Mexican Caribbean. In this study, we conclude that potential contamination by some toxic elements is a likely additional impact of the massive influx. Compared to concentrations of toxic elements reported by other studies, we observed relatively higher values of As, Cu and Mn and lower values of Cd, Cr and Pb. Total arsenic exceeded the limits established for usages in agricultural soil in several countries. We recommend further studies of inorganic forms of As to determine if it complies with guidelines of different institutions and organizations (i.e. FAO, WHO) before using sargasso in food or pharmaceutical industries. The determination of the concentrations of different elements in sargasso is also necessary to establish adequate disposal strategies of decomposing sargasso in order to mitigate the environmental risks by toxic elements pollution of coastal waters and aquifer in the near future. Sargasso should not be allowed to decompose on the beach and it should be disposed at properly prepared and managed disposal sites in order to restrict the leaching of nutrients, metals and salt into the underlying aquifer. The high and inconsistent variability of some potentially toxic metals indicates that analyses of different specimens collected over longer periods, and/or in different locations are required in order to obtain reliable information about metal contents in tissues. Governments and industries have a moral, legal and economic responsibility to consider the potential presence of toxic elements above the limits established for humans, animals and plants before approving its use and consumption.

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Table 1(on next page)

Samples information

Number of samples collected at eight sites along the Mexican Caribbean coast during 2018-2019. Habitat refers to distance from coast, shore (2-20 m from coast) or ocean (>5 km from coast).

Locality	Habitat	Sflu III	Snat I	Snat VIII	Sarg sp	Total
1 - Contoy Island	Ocean				4	4
2 - Blue waters	Ocean	1	1			2
3 - Puerto Morelos	Shore	9	9	9		27
4 - Cozumel	Ocean	1	1	2		4
5 - Mahahual	Shore	4	3			7
6 - Chinchorro	Shore		1			1
7 - Xahuayxol	Shore	6	6			12
8 - Xcalak	Shore	3	2			6
Total		**24**	**24**	**11**	**4**	**63**

Sarg sp: *Sargassum* spp., Sflu III: *Sargassum fluitans* III, Snat I: *S. natans* I, Snat VIII: *S. natans* VIII.

Table 2 (on next page)

Element concentrations median and range

Element concentrations (ppm DW) of pelagic *Sargassum* spp. tissue collected from eight localities along the Mexican Caribbean coast between 2018 and 2019. The number of samples with readings above LOD are expressed in % of the total sample size (n=63). LOD: Limit of detection.

Element	LOD	Samples with readings above LOD (%)	Minimum	Maximum	Median
Al	140	58.7	<LOD	500	206
As	4	100	24	172	80
Ca	394	100	23,723	136,146	70,040
Cl	266	100	747	53,101	22,350
Cu	6	7.9	<LOD	540	<LOD
Fe	3	7.9	<LOD	11	<LOD
K	333	100	1,990	46,002	19,666
Mg	2915	92.1	<LOD	13,662	6,537
Mn	13	100	40	139	71
Mo	1	9.5	<LOD	7	<LOD
P	145	100	228	401	327
Pb	2	7.9	<LOD	3	<LOD
Rb	1	100	30	143	56
S	199	100	9,462	24,773	14,363
Si	342	100	447	2,922	1,767
Sr	6	100	1,605	2,564	1,890
Th	1	100	5	23	10
U	4	27	11	48	23
V	3	60.3	<LOD	13	<LOD
Zn	5	12.7	<LOD	17	<LOD

Table 3(on next page)

Elements concentrations in sargasso morphotypes

Median and range (in parenthesis) of elements (ppm DW) in three sargasso species/morphotypes collected from eight localities along the Mexican Caribbean coast in 2018-2019. *P* values show summary of statistical analyses using Kruskal-Wallis *H* test (bold if significant) and the last column shows results of multiple comparison test. LOD: limit of detection.

Element	a) *S. fluitans* III (n=24)	b) *S. natans* I (n=24)	c) *S. natans* VIII (n=11)	*P*	Multiple comparison test
Al	221 (<LOD-392)	198 (<LOD-500)	<LOD (<LOD-327)	0.7341	
As	59 (34-172)	55 (32-172)	123 (24-145)	**0.0213**	c > b
Ca	76,727 (46,599-115,260)	81,965 (37,260-136,146)	43,289 (23,723-75,849)	**0.0013**	(a = b) > c
Cl	21,487 (1,831-46,502)	10,122 (747-46,485)	32,086 (2,279-53,101)	**0.0111**	c > b
K	19,466 (3,620-44,280)	14,309 (1,990-39,642)	32,900 (4,902-46,002)	**0.0121**	c > b
Mg	6,376 (2,062-12,325)	6,385 (2,062-12,949)	6,883 (2,062-13,662)	0.5990	
Mn	70 (51-112)	89 (52-139)	56 (40-135)	**0.0019**	b > (a = c)
P	336 (229-401)	328 (262-394)	300 (228-350)	0.0590	
Rb	60 (32-102)	51 (30-143)	67 (48-120)	**0.0071**	c > b
S	14,341 (11,328-24,773)	12,776 (9,462-21,170)	16,231 (12,449-19,500)	0.1370	
Si	1,861 (927-2,877)	2,095 (696-2,564)	1,049 (447-2,135)	**0.0009**	(a = b) > c
Sr	1,934 (1,641-2,395)	1,876 (1,605-2,564)	1,793 (1,633-2362)	0.4375	
Th	10 (5-17)	8 (6-23)	9 (8-20)	0.3021	
U	22 (11-48)	23 (12-47)	27 (16-45)	0.2321	

1

Table 4(on next page)

Element concentrations in different studies

Comparison of element concentration in sargasso from the Mexican Caribbean coast and other studies in different parts of the world.

Element	**Site (Year)**			
	Nigeria[a] (2012)	Dominican Republic[b] (2015)	Ghana[c] (2015)	Mexican Caribbean[d] (2018-2019)
Al		303-4,188		<140-517
As		14-42	13-54	24-172
Ba		7-17		<36
Ca		96,901-133,400		23,723-136,146
Cd		0.1-0.3	78-119	<2
Cl			61-1353	747-53,101
Co		0.4-1		<11
Cr		2-56		<8
Cu		2-12	24-36	<6-540
Fe	8,700 ± 280	20-655	1,209-5,910	<3-11
K	28,000 ± 740	2,208-33,602		1,990-46,002
Mg	42,750 ± 3,500	10,211-18,241		<2915-13,662
Mn		16-32		40-139
Mo		0.6-3		<1-7
Ni		10-33		<10
P	96,500 ± 21,200	761-1,145		228-401
Pb		1-2	105-335	<2-3
Rb		0.3-10		30-143
Si		23,883-55,776		447-2,922
Sr		1,162-1,437		1,605-2,564
Th		0.04-0.4		5-23
Ti		37-92		<29
U		0.2-0.7		11-48
V		1-3		<3-13
Y	40 ± 0.0	0.1-0.8		<1
Zn	50 ± 0.0	13-21	16-100	<5-17
Zr		8-34		<2

[a] Oyesiku & Egunyomi, 2014 (mean and SD); [b] Fernández et al., 2017 (range); [c] Dzama-Addico & deFraft-Johnson, 2016 (range); [d] This study (range)

Figure 1

Sampling sites

Location of the sampling sites of sargasso along the Mexican Caribbean coast between August 2018 and June 2019.

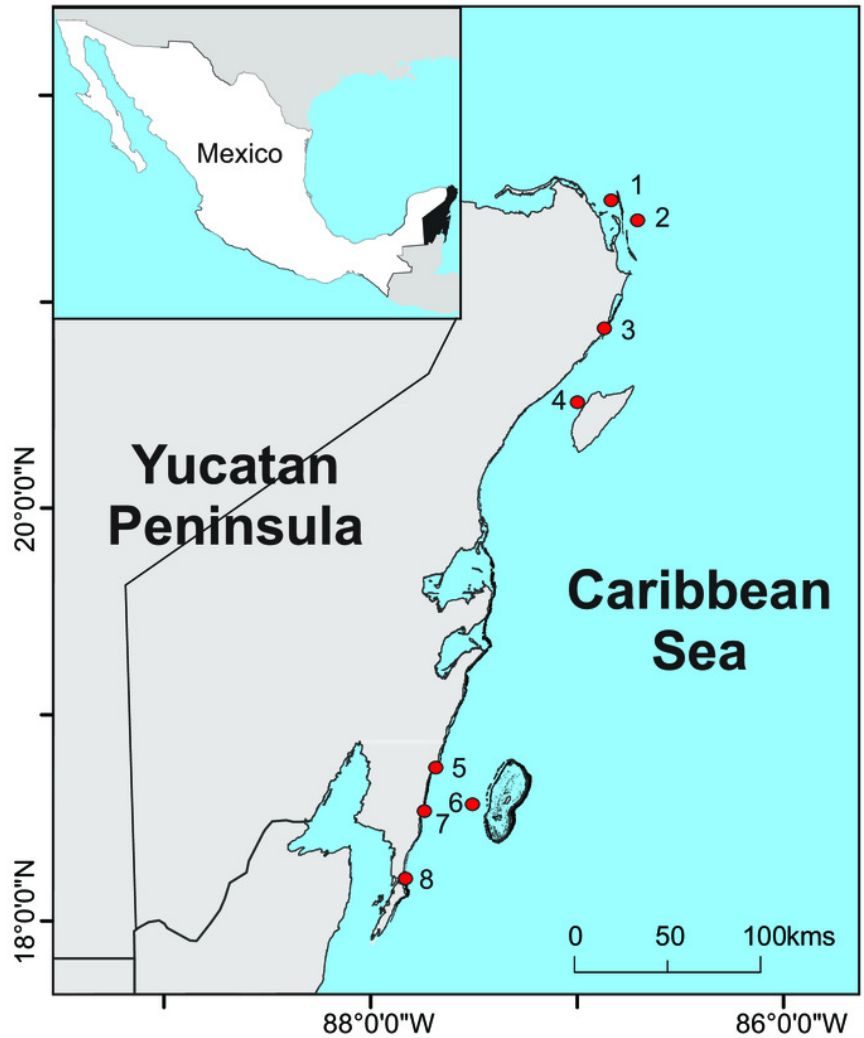

Mexico
Yucatan
Peninsula
Caribbean
Sea
1
2
3
4
5
6
7
8
0
50
100kms
20°0'0"N
18°0'0"N
88°0'0"W
86°0'0"W

Figure 2

Spatial variability in element concentrations

Concentration of fourteen most frequent elements (ppm algal DW) in tissues of sargasso collected at eight sites along the Mexican Caribbean coast in 2018-2019. Note differences in scale of the Y-axis. Each dot corresponds to the median of the five XRF readings per sample. Color of the dot represents the sargasso species/morphotype. The horizontal black lines correspond to the median for each site. The dotted blue line corresponds to the limit of detection of the XRF equipment. Fig. 1 and Table 1 have the site and sample details.

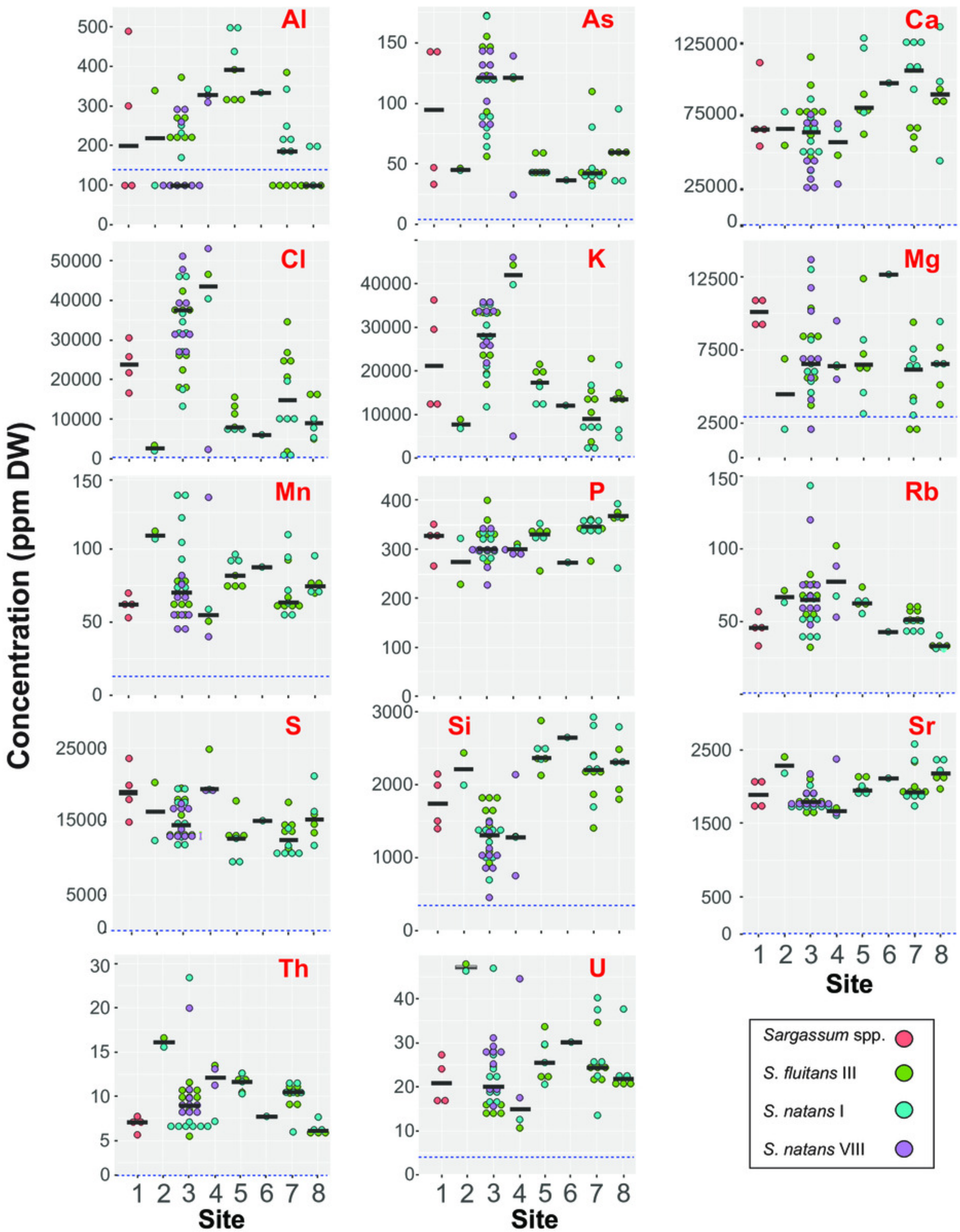
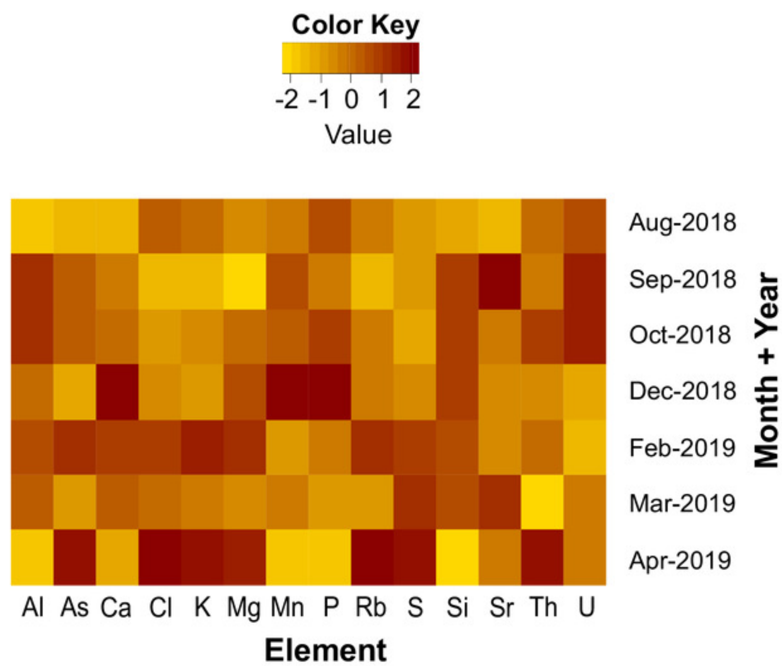

Al
As
Ca
Cl
K
Mg
Mn
P
Rb
S
Si
Sr
Th
U
Concentration (ppm DW)
Site
Sargassum spp.
S. fluitans III
S. natans I
S. natans VIII

Figure 3

Temporal variability in element concentrations

Variability in concentration of fourteen different elements (ppm DW) in sargasso collected at Puerto Morelos between August 2018 and April 2019. Z-score transformations were applied to values of each element across all the sampling periods and their intensities above and below the mean are represented on the heatmap by red and yellow colors respectively, as shown on the color key bar.


Color Key
-2 -1 0 1 2
Value
Aug-2018
Sep-2018
Oct-2018
Dec-2018
Feb-2019
Mar-2019
Apr-2019
Month + Year
Al As Ca Cl K Mg Mn P Rb S Si Sr Th U
Element