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Pelagic *Sargassum* events in Jamaica: Provenance, morphotype abundance, and influence of sample processing on biochemical composition of the biomass

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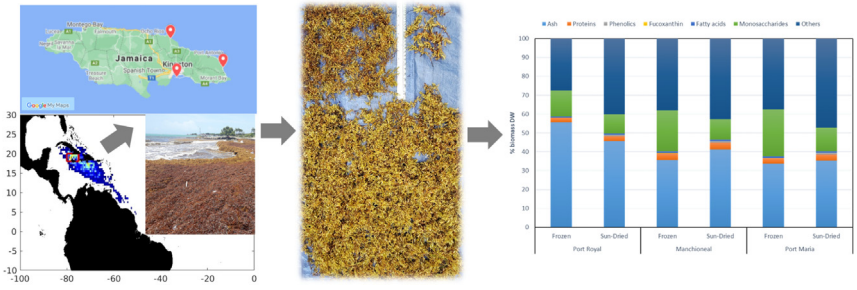
HIGHLIGHTS

- Pelagic *Sargassum* events are the new normal in the Caribbean and Western Africa.
- *Sargassum fluitans* was dominant in summer, but morphotype abundance may be seasonal.
- Seaweeds from the south and the north of Jamaica followed a similar migration route.
- Sample processing affect the biochemical composition of pelagic *Sargassum* biomass.

GRAPHICAL ABSTRACT



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ABSTRACT

Pelagic *Sargassum* species have been known for centuries in the Sargasso Sea of the North Atlantic Ocean. In 2011, a new area concentrating high biomass of these brown algae started developing in the Tropical Atlantic Ocean. Since then, massive and recurrent *Sargassum* influxes have been reported in the Caribbean and off the coast of Western Africa. These *Sargassum* events have a major negative impact on coastal ecosystems and nearshore marine life, and affect socio-economic sectors, including public health, coastal living, tourism, fisheries, and maritime transport. Despite recent advances in the forecasting of *Sargassum* events, and elucidation of the seaweed composition, many knowledge gaps remain, including morphotype abundance during *Sargassum* events, drift of the seaweeds in the months prior to stranding, and influence of sample processing methods on biomass biochemical composition. Using seaweeds harvested on the coasts of Jamaica in summer of 2020, we observed that *S. fluitans* III was the most abundant morphotype at different times and sampling locations. No clear difference in the geographical origin, or provenance, of the *Sargassum* mats was observed. The majority of *Sargassum* backtracked from both north and south of Jamaica experienced ambient temperatures of around 27 °C and salinity in the range of 34–36 psu before stranding. We also showed that cheap (sun) compared to expensive (freeze) drying techniques influence the biochemical composition of biomass. Sun-drying increased the proportion of phenolic compounds, but had a deleterious impact on fucoxanthin content and on the quantities of monosaccharides, except for mannitol. Effects on the content of fucose containing sulfated polysaccharides depended on the method used for their extraction, and limited variation was observed in ash, protein, and

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fatty acid content within most of the sample locations investigated. These observations are important for the storage and transport of the biomass in the context of its valorisation.

1. Introduction

Pelagic *Sargassum* species *S. fluitans* (morphotype III) and *S. natans* (morphotypes I and VIII) form important ecosystems in the Sargasso Sea and in the Atlantic Ocean to support a diverse epifauna and important migratory species (Martin et al., 2021). However, during the last decade, the coasts of the Caribbean and Western Africa have been affected by massive strandings of these brown algae, referred to as *Sargassum* events (Fidai et al., 2020). As recently stated in the "*Sargassum* White Paper – Turning the crisis into an opportunity" published by the United Nations Environmental Programme and the Caribbean Environmental Programme (UNEP-CEP, 2021), these *Sargassum* events have a major negative impact on coastal ecosystems and nearshore marine life, and affect socio-economic sectors, including public health, coastal living, tourism, fisheries, and maritime transport.

Concomitantly to the increase in *Sargassum* events, there has been a surge in publications related to several aspects of pelagic *Sargassum* beyond the Sargasso Sea. These concerned methods and analysis for remote sensing, forecasting, and quantification of biomass (Ody et al., 2019; Wang et al., 2019; Berline et al., 2020; Marsh et al., 2021; Wang and Hu, 2021), as well as investigation of factors driving *Sargassum* events (Johns et al., 2020; Lapointe et al., 2021). Another area of interest is the description of the environmental (Van Tussenbroek et al., 2017; Salter et al., 2020; Devault et al., 2021) and socio-economic impact (Resière et al., 2019; Merle et al., 2021) in regions affected by recurrent influxes of pelagic *Sargassum*. Despite the numerous challenges associated with pelagic *Sargassum* events, it is also clear that many opportunities exist around the valorisation of this biomass (Milledge and Harvey, 2016; Chávez et al., 2020; Thompson et al., 2020, 2021; Amador-Castro et al., 2021; Joniver et al., 2021; Oxenford et al., 2021; Robledo et al., 2021). Indeed, it is well acknowledged that seaweeds are making a significant contribution towards achieving several targets of the United Nations sustainable development goals (Bourgougnon et al., 2021), and will play a key role in the development of a sustainable circular economy in several parts of the World (Barbier et al., 2020), notably as biorefineries (Kostas et al., 2021).

Pelagic *Sargassum* is important in carbon cycling and sequestration at the local scale (Hu et al., 2021). Several directions have been explored to valorise pelagic *Sargassum*, as the seasonal occurrence of this potential feedstock may represent the new norm in the Caribbean and Western Africa. Very recently, a number of studies have investigated the conversion of pelagic *Sargassum* biomass into renewables liquid fuels (Aparicio et al., 2021; Marx et al., 2021), for bioenergy production through anaerobic digestion (Thompson et al., 2020, 2021; López-Aguilar et al., 2021), for production of activated carbon to be used in the context of wastewater treatment (Francoeur et al., 2021) and bioremediation in coastal ecosystems (Saldarriaga-Hernandez et al., 2020). Gray et al. (2021) described a different approach based on development of low-cost machinery for harvesting and sinking of *Sargassum* biomass, as an alternative to onshore mechanical harvesting and disposal at landfills.

Recent progress has also been made on extending the characterization of the biochemical and elemental composition of pelagic *Sargassum*. This is important to understand the biology of these organisms, and also to inform valorisation pathways. Most of the recent results on biomass species and biochemical composition have been obtained from samples collected from the coasts of Mexico (García-Sánchez et al., 2020; Rodríguez-Martínez et al., 2020; Rosado-Espinosa et al., 2020; Hernández-Bolio et al., 2021; Robledo et al., 2021; Saldarriaga-Hernandez et al., 2021; Vázquez-Delfín et al., 2021), with limited insights from other parts of the Caribbean, i.e., Turks and Caicos (Milledge et al., 2020) and Jamaica (Davis et al., 2021). Several parameters potentially influencing the biochemical and elemental composition of the pelagic *Sargassum* have been investigated, including morphotype abundance, sampling conditions and locations, season of sampling, methods for sample stabilization, processing and extraction of the biomass, before implementing a wide range of analytical approaches to quantify several compounds known to be present in *Sargassum*.

In this context, we had several objectives in this new study: (i) assess the most abundant *Sargassum* morphotypes stranding in Jamaica during summer 2020; (ii) analyse the origin, hence provenance, of these seaweeds; and (iii) investigate the influence of sample processing on the biochemical composition of this biomass.

2. Materials and methods

2.1. Sample collection and processing

Sargassum was collected from the surf (in less than 1 m of water) using a 'surf net' in the following sequence: Fort Rocky beach, Port Royal (17°56′13.9"N 76°49′02.8"W) on July 10, 2020; Manchioneal (18°02′39.6"N 76°16′31.5"W) on July 12, 2020; Port Maria (18°21′55.3"N 76°53′32.2"W) on August 9, 2020 (Fig. 1A). Samples (at least 7 kg) were transported to the laboratory, where they were cleaned of non-*Sargassum* debris. Three two-kg portions of the freshly cleaned *Sargassum* had the three morphotypes separated (using their gross morphological features) into three individual piles and spread to dry for ~36 h at temperatures of 34.8 °C (sun), 27.9 °C (shade) and 25 °C (night-time). Three 100-g portions were packaged in separate Ziploc® bags and frozen using liquid nitrogen within two hours of packaging. Frozen samples were stored at −20 °C. All fresh sample processing was complete within six hours of collecting. Each morphotype was weighed separately on an Ohaus AX423N Adventurer top-loading balance, and then recombined and packaged for shipment to the University of York. After arrival, the frozen samples were freeze-dried for 48 h in a Heto Power Dry PL3000 freeze dryer (Thermo Fisher Scientific). Both frozen and sun-dried samples were milled to 1 mm diameter particle size using a Cyclone Mill Twister (Retsh). The frozen samples were stored at 4 °C as suggested by Milledge et al. (2020), and the sun-dried at room temperature.

2.2. Moisture and ash content

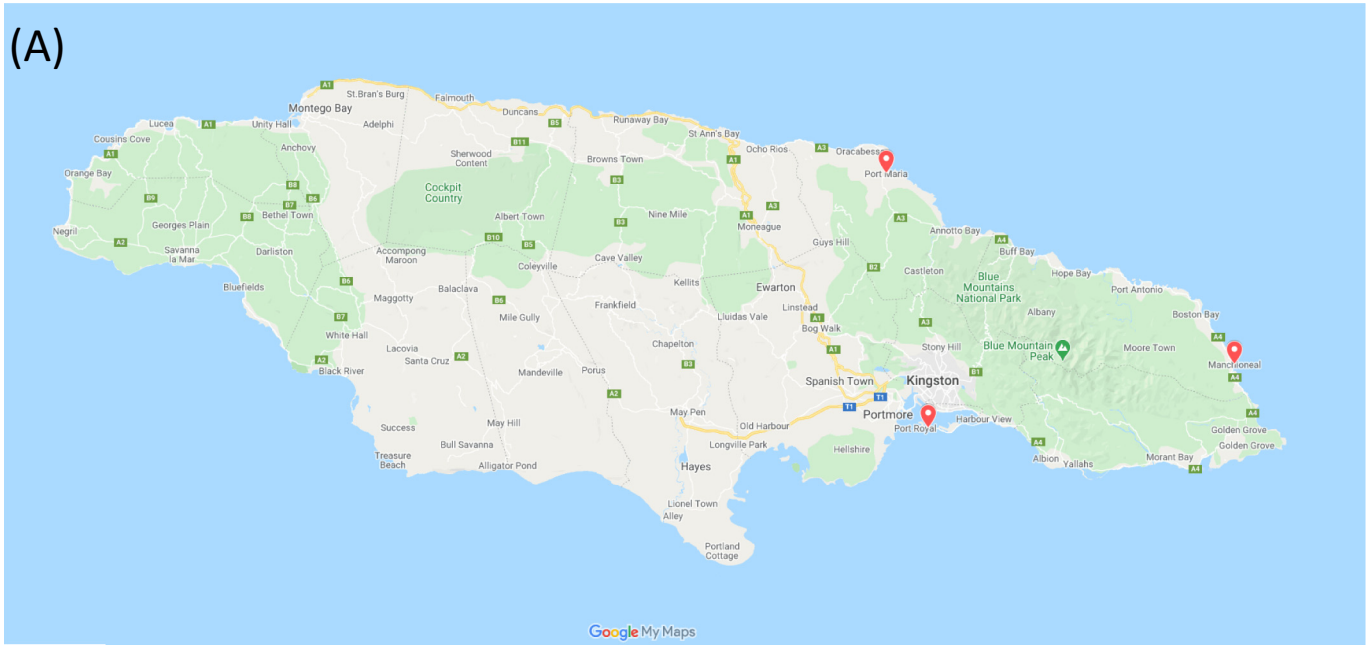
For moisture analysis, one gram of dried algae was weighted in a crucible, and incubated in a Gallenkamp Hot Box Oven at 105 °C for 24 h before monitoring change in the weight of the samples. For ash determination, samples were further incubated for 2 h at 550 °C in a Carbolite AAF ashing furnace, and then monitored again. All the measurements were performed in triplicate.

2.3. Monosaccharide composition of the non-cellulosic fraction of the biomass

Four milligrams of samples were weighed in triplicate in screw-capped tubes, and analysed as described in Davis et al. (2021) and in Supplementary Text.

2.4. Fucoxanthin content

Fifty mg of dried algae powder were loaded into 2-ml plastic tubes and the pigments were extracted by the addition of 1 ml of methanol. After short vortexing, the samples were centrifuged at room temperature at 14,000 rpm for 5 min, and the supernatant was recovered. The fucoxanthin content was determined using a Waters 2695 high-performance liquid chromatography system according to Gupta et al. (2015). Fifty μl of methanolic extract were injected onto a reverse-phase C30, 3 μm column (250 × 4.6 mm; YMC Co., Kyoto, Japan) coupled to a 20 × 4.6 mm C30 guard column (YMC Co., Kyoto, Japan) using a mobile phase consisting of methanol (A) and water (B). The gradient elution used with this column was 60% A/40% B for 30 min, followed by a linear gradient up to 100% A



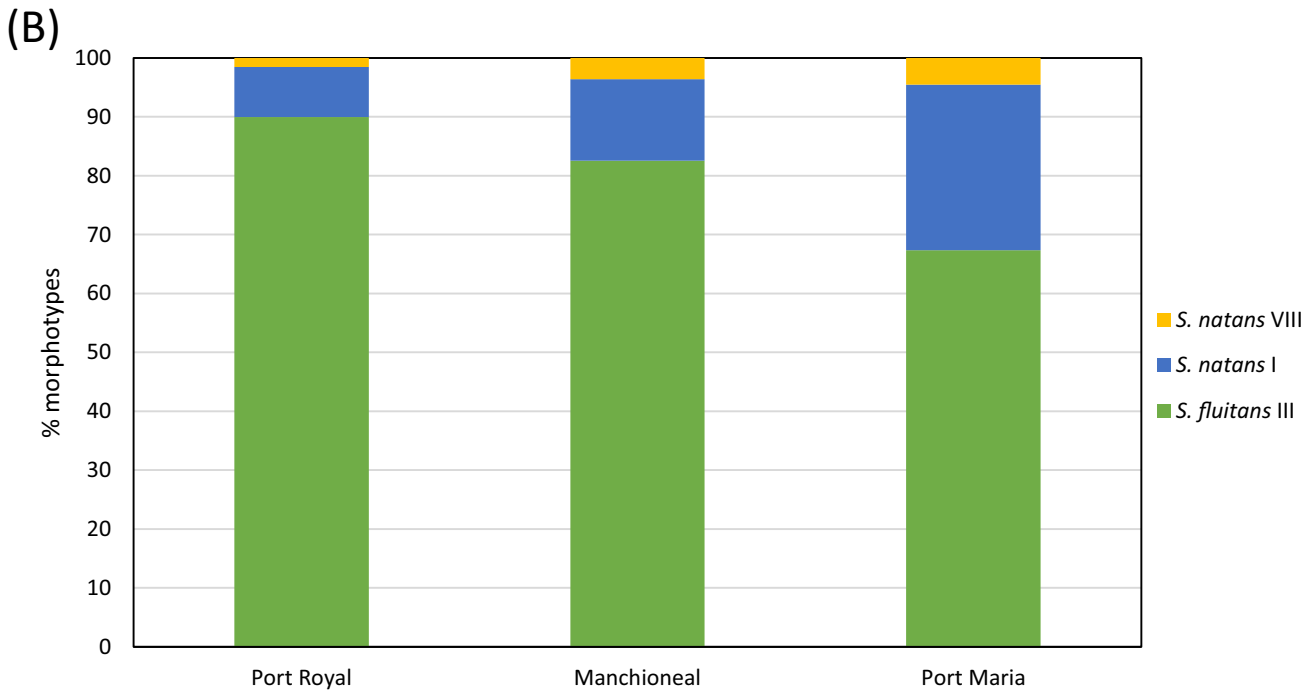


Fig. 1. Sampling location and pelagic *Sargassum* morphotype abundance. A) Map of sampling location. B) Relative abundance of the three morphotypes at different sampling sites. Results (mean ± SD) are expressed as % of total biomass DW.

for 30 min at a flow rate of 1 ml/min, and then 100% A for 10 min. A re-equilibration step of 5 min with 60% A/40% B was included between each sample. The column temperature was maintained at 25 °C. The eluting peaks were monitored using a photodiode array detector (250–700 nm range). For quantification, a stock solution of fucoxanthin (F6932-10MG, Sigma, UK) was prepared at 1 mg/ml final concentration using acetone and a standard curve was obtained by injecting 50 μl of different concentrations (0.05, 0.025, and 0.005 mg/ml) as described above for the algal samples. Comparison of peak area between algae samples and standards concentrations was performed using Empower software.

2.5. Polyphenol and phlorotannin content

Analysis was performed on 0.5 g of dried algae powder by adapting a protocol described by Davis et al. (2021) and in Supplementary Text.

2.6. Extraction of fucose containing sulfated polysaccharides (FCSPs)

These FCSPs are specific components of brown algae cell walls. Two different protocols, using an enzyme-assisted or acid extraction on 0.5 g of dried algae, were adapted from Nguyen et al. (2020), and are described in Supplementary Text.

2.7. Sulfate content and monosaccharide composition of extracted FCSPs

Extracted FCSPs (4–5 mg) were hydrolysed with TFA, and hydrolysates were used for analysis of monosaccharide content as described in Section 2.3. These were also used for determining the sulfate content considering a microplate assay adapted by Nguyen et al. (2020) according to the barium chloride ($BaCl_2$) gelatin method developed by Dodgson and Price (1962), and described in Supplementary Text.

2.8. Determination of fatty acid content

Fatty acid methyl esters (FAMEs) were analysed using the direct transmethylation protocol described by Larson and Graham (2001), with modifications for lyophilised seaweed analysis, as described in Supplementary Text.

2.9. Protein content

Quantities of proteins were determined by employing a modified protocol originally developed for microalgae by Slocombe et al. (2013). Briefly, proteins were extracted from twenty mg of dried algae powder using a sequential hot-TCA and alkaline solution incubation. The hydrolysates were recovered to perform protein quantification using the Lowry assay as previously described (Slocombe et al., 2013).

2.10. Data analysis

All the morphotype and biochemical analyses were performed in triplicate. Potential difference in the morphotype abundance and influence of sampling location on the algae composition were assessed. Potential variations related to sites of collection (related to different proportions of morphotypes) and processing of samples (frozen and sun-dried) on the biochemical composition of the *Sargassum* biomass were also investigated. For these, data were tested for normality and homogeneity of variance using the Shapiro-Wilk test and Brown-Forsythe test, respectively. Two-way ANOVA was performed, followed by a post-hoc Holm-Sidak test for all pairwise multiple comparisons. The significance level was set at p-value ≤ 0.05 for all the data analysis. All the statistical analyses were done using SigmaPlot version 14.0.

2.11. Backtracking of pelagic Sargassum

We use an ensemble approach to determine the statistics of *Sargassum* drift towards Jamaica, subjecting virtual particles to variable surface winds and currents. *Sargassum* detected as floating algae (FA) is provided as daily "FA_density" in satellite images available from the Optical Oceanography Laboratory at the University of South Florida, via the website https://optics.marine.usf.edu. For a given day, an image represents the mean of the seven past days (including that day), based on the method described in Wang and Hu (2016). In proportion to the *Sargassum* detected in July and August of 2020, and allocating one particle per 10 m^2 of *Sargassum*, we initialised virtual particles in the vicinity of Jamaica. These particles were then backtracked for six months, sampling currents and winds from an eddy-resolving (1/12°) ocean hindcast for 1988–2010, based on the Nucleus for European Modelling of the Ocean (NEMO) ocean model (Madec, 2008) in eddy-resolving global configuration (ORCA12), henceforth NEMO-ORCA12.

To calculate individual particle trajectories, we used the off-line Lagrangian ARIANE mass-preserving algorithm (Blanke and Raynaud, 1997) in 'qualitative mode', with the 5-day mean horizontal velocity fields of NEMO-ORCA12. We thus analysed the provenance of *Sargassum* detected south of Jamaica (16–18°N, 75–80°W), backtracked from around 10th of July, and detected north of Jamaica (18–20°N, 75–80°W), backtracked from around 9th of August. Using each year of the hindcast, we obtained a 23-member ensemble that samples a range of ocean variability, in the absence of synoptic current and wind data for 2020. Particle locations and local water properties (in the NEMO-ORCA12 hindcast) are recorded every 5 days. Given the number of particles used to represent the satellite-detected *Sargassum*, of order 10^3–10^4, we average this data on a 0.5° grid to obtain areal fractions of *Sargassum* for selected times (before arrival near Jamaica) and means of 'age' (time prior to arrival near Jamaica), temperature and salinity. This method followed an approach to seasonal forecasting for *Sargassum* outlined in Marsh et al. (2021).

3. Results and discussion

3.1. S. fluitans was the most abundant morphotype in summer 2020 in Jamaica

As seen in Fig. 1B, *S. fluitans* III was the most abundant morphotype observed at the three sampling locations (67–90% of the biomass DW). Differences between proportions of the three morphotypes within each sampling location were statistically supported (all p-values <0.005; Supplementary Table S1). The percentage of *S. natans* VIII did not change significantly between the three locations, in contrast to what was observed for *S. natans I* and *S. fluitans* III. Indeed, while *S. natans* I represented less than 10% of the total biomass in Port Royal, it reached almost 30% in Port Maria, with concomitant decrease in the abundance of *S. fluitans* III. So far, there has been limited reports on accurate determination of the proportions of the different pelagic morphotypes in beach-cast *Sargassum* biomass. However, observations in Jamaica are similar to results described by García-Sánchez et al. (2020) and Vázquez-Delfín et al. (2021) after events affecting the Mexican Caribbean coasts between 2016 and 2020. Both studies reported *S. fluitans* III as the most abundant of the three morphotypes, in similar proportions compared to the Jamaican samples.

Differences in the proportions of morphotypes observed in different locations in Jamaica may be related to the time of sampling and potential changes in the species composition of the pelagic *Sargassum* mats, i.e., the sampling in Port Maria (north coast) happened one month after sampling in Port Royal (south coast). The extent to which *Sargassum* at these two sampling locations and times may have distinct origins was investigated with the backtracking approach outlined in Section 2.11. A summary of ensemble-mean results is presented in Fig. 2. The range of pathways and movement towards Jamaica is evident in the fractional (%) *Sargassum* coverage 50, 100, and 150 days prior to arrival north/south of Jamaica (panels A-B-C, and E-F-G in Fig. 2 respectively), and mean ages to 180 days (prior to July/August) as shown in panels D and H of Fig.2. Note that, as little *Sargassum* was detected in early August, we limited the legend range in Fig. 2 panels A-B-C. Comparing July (south) and August (north) provenance, and mean ages in particular, we note slightly swifter movement from the equatorial Atlantic to south Jamaica for July backtracking, with *Sargassum* from this origin arriving off south Jamaica around 30 days earlier than off north Jamaica. Corresponding in-situ surface temperature and salinity are shown in Supplementary Fig. S1. On this evidence, the majority of *Sargassum* backtracked from both north and south of Jamaica experienced ambient temperatures of around 27 °C and salinity in the range of 34–36 psu, in July and August. Particles that originate in the western equatorial Atlantic 100–150 days prior to arrival off Jamaica experienced higher temperatures of up to 29 °C. Particles adjacent to the Amazon plume may experience salinities substantially below 34 psu. Differences in ambient temperature and salinity may be of consequence for growth rates and mortality vital effects which may vary between *Sargassum* morphotypes. Previous findings based on *Sargassum* morphotypes in the Sargasso Sea (Hanisak and Samuel, 1987) may not apply to blooms in the tropics, where macronutrients are plentiful in comparison to the subtropics. However, more recent analyses of different benthic *Sargassum* species found in subtropical and temperate coastal waters of northeast Asia indicate a strong growth dependency on temperature and salinity (Zou et al., 2018; Li et al., 2019).

3.2. Moisture and ash content

Moisture content accounted for 8.5–13.2% of the biomass DW in frozen samples, and 8.48–11.6% in sun-dried samples (Table 1, Supplementary

Table S2). Differences were statistically significant when comparing the three sampling sites (all p-values ≤ 0.029) and between frozen and sun-dried samples within Manchioneal and Port Maria ($p < 0.001$ and $p = 0.037$ respectively). Ash content comprised between 33.7 and 55.7% of the biomass DW in frozen samples, and 35.3–45.7% in sun-dried samples. Significant differences were observed between frozen and sun-dried samples in Port Royal ($p = 0.002$) and Manchioneal ($p = 0.043$), not in Port Maria. Ash content was significantly different when comparing the three sampling sites (all p-values ≤ 0.047).

The moisture content determined in the current samples was slightly higher compared to values measured in sun-dried samples after harvesting in February 2019 (7–8%) in the Port Royal area of Jamaica (Davis et al., 2021). Ash contents in the 2020 samples were in a range similar to those determined in 2019 Jamaican samples (35–40%). In addition, they were higher compared to those (18–24%) determined in *Sargassum* biomass beach-casted on the coasts of the Mexican Caribbean in September 2018 (biomass in which *S. fluitans* III was the most abundant morphotype; Vázquez-Delfín et al., 2021). Values for ash in the 2020 samples are in the same range as those reported by Milledge et al. (2020) in pelagic *Sargassum* samples harvested in the Turks and Caicos Islands in June 2019. Differences may be due to season and location, as suggested by Milledge and Harvey (2016).

3.3. Fucoxanthin content

Fucoxanthin is the main carotenoid pigment produced by brown algae, and numerous bioactivities have been demonstrated in vitro for this pigment (Pereira et al., 2021). Fucoxanthin content was measured by HPLC, and differences between frozen and sun-dried samples were supported statistically for the three sampling sites (all p-values <0.001) (Table 1, Supplementary Table S3). In the frozen samples, contents ranged between 262 and 504 μg/g biomass DW, while they were 58–136 μg/g in the sun-dried samples. The highest amount of fucoxanthin was measured in the Manchioneal frozen samples, where it represents 0.05% of the biomass DW. Significant variations were observed between Manchioneal and Port Maria, and between Manchioneal and Port Royal (both p-values <0.001), but not

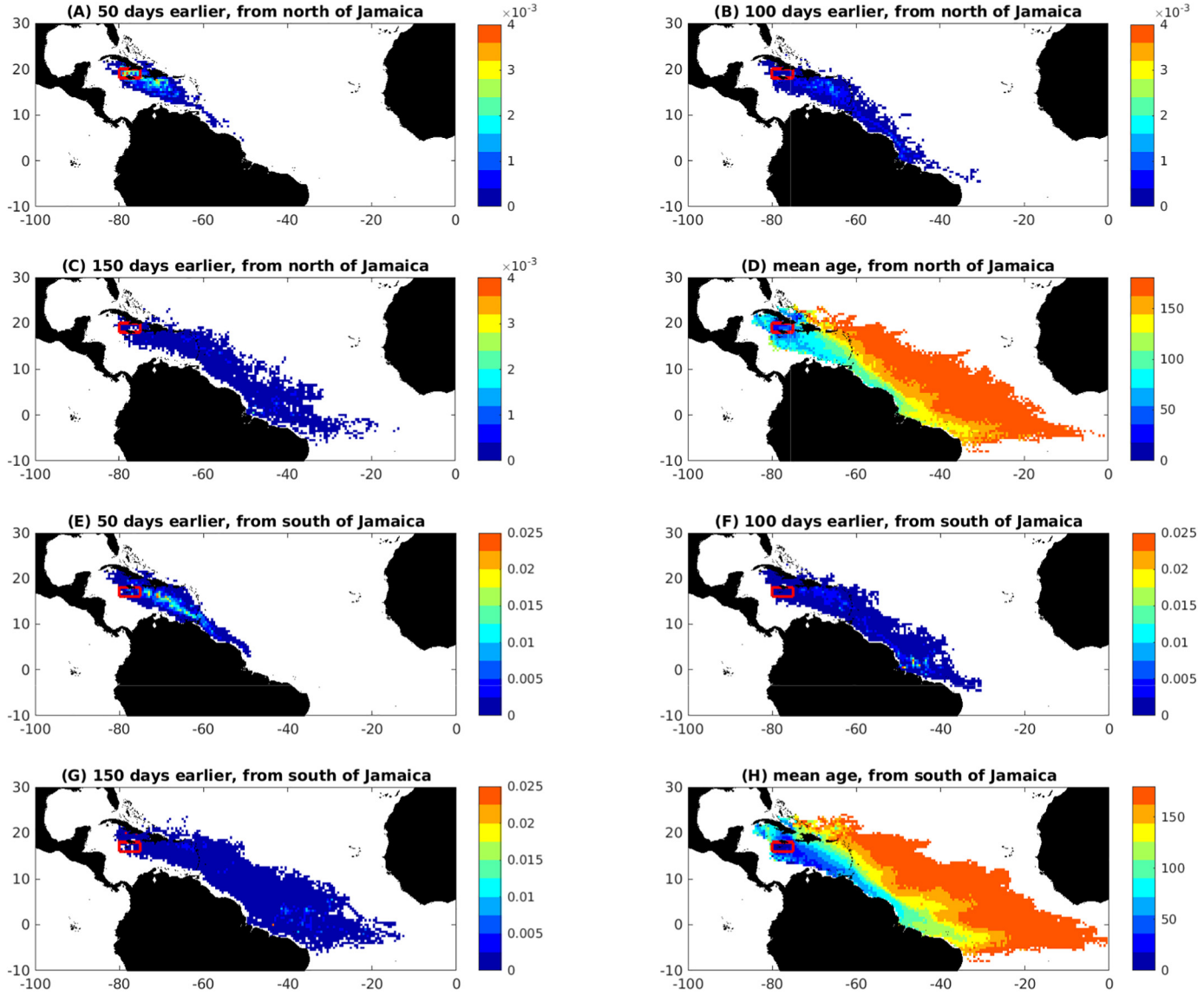


Fig. 2. Backtracking from north (panels A-B-C-D) and south (panels E-F-G-H) of Jamaica, indicating areal fraction of *Sargassum* (colour-coded in the range 0–0.004% in A-B-C, and 0–0.025% in E-F-G) 50 days (A,E), 100 days (B,F), and 150 days (C, G) prior to arrival off Jamaica (within red box); the mean age at all recorded locations is indicated in panels D and H. In proportion to the satellite detection of *Sargassum* areal fractions, backtracking from north and south of Jamaica was represented with 2701 and 9437 particles respectively, for each of the 23 sampled hindcast years (see Section 2.11). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 1
Biochemical composition of frozen and sun-dried samples of pelagic *Sargassum* biomass harvested at three different locations in Jamaica (summer 2020). Results represent mean ± SD of three biological replicates for each site and sample processing method.

Biochemical content	Port Royal		Manchioneal		Port Maria	
	Frozen	Sun-dried	Frozen	Sun-dried	Frozen	Sun-dried
Moisture (% of biomass DW)	8.50 ± 1.18	8.48 ± 0.25	12.87 ± 0.35	9.54 ± 0.18	13.23 ± 0.29	11.60 ± 1.08
Ash (% of biomass DW)	55.69 ± 5.43	45.65 ± 1.79	35.61 ± 0.54	41.29 ± 1.50	33.74 ± 1.34	35.32 ± 0.72
Fucoxanthin (μg/g of biomass DW)	272.26 ± 45.36	107.15 ± 10.72	503.81 ± 42.49	136.32 ± 16.48	261.68 ± 18.19	57.74 ± 22.67
Total phenolics (mg/g of biomass DW)	0.98 ± 0.05	7.66 ± 0.60	1.46 ± 0.03	8.78 ± 2.03	1.45 ± 0.14	10.50 ± 6.21
Phlorotannins (mg/g of biomass DW)	0.35 ± 0.07	2.84 ± 0.13	0.46 ± 0.15	3.20 ± 0.51	0.29 ± 0.10	3.56 ± 2.31
Proteins (mg/g of biomass DW)	23.27 ± 0.95	26.21 ± 1.63	34.27 ± 1.55	34.91 ± 1.51	29.37 ± 1.35	31.80 ± 3.17
Alginate:						
% biomass DW	5.95 ± 1.27	5.12 ± 0.42	13.79 ± 0.45	5.60 ± 0.21	16.34 ± 0.89	6.82 ± 0.52
% total monosaccharides	43.05 ± 4.46	50.14 ± 1.27	63.81 ± 1.05	51.33 ± 0.38	65.78 ± 1.74	54.89 ± 2.88
M/G ratio	3.06 ± 0.11	3.00 ± 0.09	2.42 ± 0.02	2.87 ± 0.04	2.41 ± 0.04	2.83 ± 0.02

between Port Royal and Port Maria. The lower amounts of fucoxanthin measured in sun-dried samples maybe explained by the exposure of algae to light and high temperature during the drying process since both abiotic factors have been shown to be responsible for the degradation of fucoxanthin extracted from the brown alga *Costaria costata* (Zhao et al., 2019).

There is a limited description of carotenoid content in pelagic *Sargassum*. Using a different analytical approach, Vázquez-Delfín et al. (2021) described amounts of carotenoids ranging between 0.08 and 0.13 mg/g biomass DW, so lower than the fucoxanthin content determined in our analysis. Interestingly, quantities of fucoxanthin extracted from pelagic *Sargassum* frozen samples were above those determined in *S. muticum* freeze-dried samples after using different extraction protocols and analysis by HPLC (up to 0.12 mg/g biomass DW; Conde et al., 2015). These values in pelagic and benthic *Sargassum* are in the lower range compared to those determined for other brown algae, i.e., 0.04 mg/g for *Laminaria japonica* to 18.60 mg/g for *Himanthalia elongata* biomass DW (Pereira et al., 2021).

3.4. Contents of phenolics and phlorotannins

Polyphenols are a large and diverse group of bioactive compounds with antioxidant properties, and are used in a number of industries (Cotas et al., 2020). In contrast to fucoxanthin, higher contents of phenolics and of phlorotannins were observed in sun-dried compared to frozen samples, and this was supported statistically (all p-values <0.001) (Table 1, Supporting Table S4). Higher amounts of phenolics and phlorotannins in such samples maybe due to the activation of their biosynthetic pathways during the drying process and long exposure to light, as previously observed by Catarino et al. (2019) for a brown alga closely related to *Sargassum* species. Phenolic contents reach up to 1% biomass DW in Port Maria sun-dried samples. However, this data should be taken carefully because, despite several attempts to quantify phenolics and phlorotannins, one biological replicate from this location showed very low amounts compared to the other two biological replicates, and we do not currently have any explanation for this. The percentage of phlorotannins within the total amounts of phenolics was quite similar in both frozen and sun-dried samples, i.e., 0.2–0.4%. No significant differences were observed between the three sampling sites for both phenolics and phlorotannins. Values determined in the sun-dried samples for the total phenolic content are higher than those described for the same type of samples in Davis et al. (2021) and Milledge et al. (2020) for individual morphotypes, and by Saldarriaga-Hernandez et al. (2021) for *Sargassum* biomass harvested in 2020 from the east coast of Yucatan in Mexico. However, they are in the same range as those described by Vázquez-Delfín et al. (2021). Interestingly, Powers et al. (2019) described pelagic *Sargassum* as the most significant biological source of polyphenols in the ocean recorded to date after sampling in the Sargasso Sea during July 2016, and this warrant further analysis of the fate of these phenolic compounds in the open sea.

3.5. Protein content

Protein content ranged between 23 and 35 mg/g of biomass DW in the samples analysed. (Table 1, Supplementary Table S5). No statistical difference was observed between frozen and sun-dried samples, although slightly higher contents of proteins were measured in sun-dried samples of both Port Royal and Port Maria compared to frozen samples. However, significant differences were overserved between the three sampling sites (all p-values ≤ 0.009), with higher amounts of proteins in biomass harvested at Manchioneal. The content of protein determined in this study (2.3–3.5% of the biomass DW) is low compared to previous observation in other samples of pelagic *Sargassum*, as illustrated in Vázquez-Delfín et al. (2021; 8–10% of the biomass DW), and Saldarriaga-Hernandez et al. (2021; 3–11% of the biomass DW). However, Milledge et al. (2020) determined a total amino acid content of 4.16% biomass DW from mixed *Sargassum* harvested in Turks and Caicos, which is similar to the results in our study. Estimation of protein content using different methods makes the comparison between studies difficult. In addition, one important parameter to consider when assessing amounts of proteins in a targeted biomass is its amino acid profile, in particular the presence and quantities of the essentials amino acids (histidine, isoleucine, leucine, lysine, methionine, phenylalanine, threonine, tryptophan, and valine) that cannot be produced by the human body. The scarce information available shows that the three pelagic *Sargassum* morphotypes produce all these essential amino acids (Milledge et al., 2020). However, and as stated by these authors, more investigations are required to explore pelagic *Sargassum* feedstock as a potential source of amino acids and proteins.

3.6. Fatty acid content

Fatty acid (FA) content ranged between 6000 and 10,000 μg/g biomass DW, so equivalent to 0.6–1% of the biomass DW. Higher amounts were quantified in frozen compared to sun-dried samples for Manchioneal and Port Maria, but differences were statistically significant only for Manchioneal ($p < 0.001$) (Fig. 3, Supplementary Table S6). The most abundant fatty acids were: palmitic acid (16:0, 40–50 wt%), oleic acid (18:n9, 10–12 wt%), arachidonic acid (20:4n6, 8–14 wt%), and myristic acid (14:0, 7–8 wt%). Very limited variations were observed in the percentage of individual FAs between frozen and sun-dried samples. However, changes in the content of 20:4n6, a polyunsaturated fatty acid for which different activities have been demonstrated in human biology, were significant between the three locations after pairwise comparison (all p-values ≤ 0.036), and between frozen and sun-dried samples in each of the three locations (all p-values ≤ 0.038), with 1.5–2 fold more arachidonic acid in frozen samples compared to sun-dried ones. Similarly, but to a lower extent, amounts of the omega 3 fatty acid eicosapentaenoic acid (EPA) were higher in frozen samples, although significantly only for algae harvested in Manchioneal ($p < 0.001$) and Port Maria ($p = 0.007$).

In line with our observations, previous reports by van Ginneken et al. (2011) and Milledge et al. (2020) showed that palmitic acid, oleic acid, and arachidonic acid were the most abundant FAs in pelagic *Sargassum* species harvested from the middle of the North Atlantic Ocean (*S. natans*) or from the coasts of Turks and Caicos (*S. fluitans* and *S. natans*), respectively. A similar pattern was also observed in other species of benthic *Sargassum* by Khotimchenko (1991), Conde et al. (2015), and Schmid et al. (2018), except that the content of EPA, a health beneficial polyunsaturated fatty acid, was higher in the different species of *Sargassum* investigated in the three studies mentioned above compared to the results obtained for pelagic *Sargassum*.

3.7. Monosaccharide composition in the non-cellulosic fraction

The amounts of monosaccharides ranged between 100 and 250 mg/g biomass DW, so equivalent to 10–25% of the biomass (Fig. 4, Supplementary Table S7). Significant differences were observed between the three sampling sites investigated (all p-values ≤ 0.003), and also between frozen and sun-dried samples (all p-values ≤ 0.002), with higher contents in the former type of samples compared to the latter. Values obtained for the sun-dried samples, which ranged between 10 and 15% of the biomass DW, are lower compared to those measured for mix *Sargassum* biomass, also predominantly composed of *S. fluitans*, harvested in Jamaica in February 2019 (Davis et al., 2021).

The extraction of the cell wall polysaccharide alginates, made of mannuronic and guluronic acids, supports the economic value and numerous applications of brown algae. Both uronic acids were the most abundant compounds in the fractions investigated, with alginate representing 43.05 to 65.78% of total sugars measured, and 5.12 to 16.34% of the biomass DW (Table 1). These values, based on the direct quantification of both uronic acids, are lower to those recently described, based on different procedures, in samples harvested in Mexico for individual species: Rosado-Espinosa et al. (2020) reported alginate representing 34.6 ± 2.8% of biomass DW in *S. fluitans*, while Robledo et al. (2021) described alginate content accounting for 18.2–32% and 20–25.2% of biomass DW for *S. fluitans* and *S. natans*, respectively. Vázquez-Delfín et al. (2021) analysed mix biomass samples harvested along 120 km in the north coast of the Mexican Caribbean, and observed only limited variations in the alginate content in these samples since all values were comprised between 29.3 ± 2.0 to 33.9 ± 1.7% of the biomass DW.

Significant differences were observed in the alginate content expressed as % of the biomass DW between the three sampling sites (all p-values ≤ 0.003), and between frozen and sun-dried samples taken from Manchioneal ($p < 0.001$) and from Port Maria ($p < 0.001$). The M/G ratio

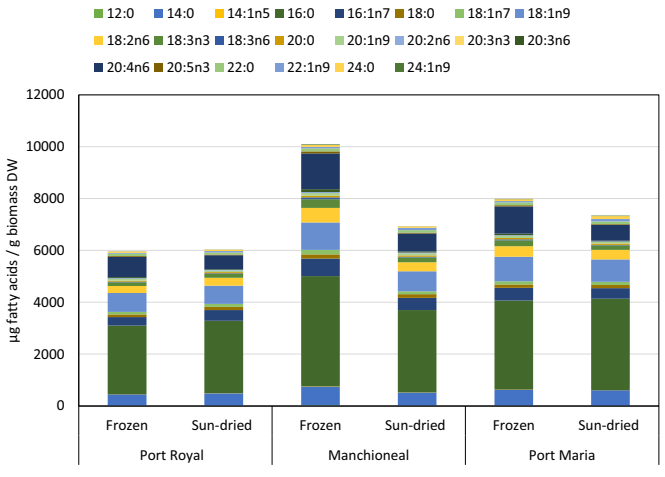


Fig. 3. Fatty acid composition of frozen and sun-dried samples of pelagic *Sargassum* biomass harvested at three different locations. Results are expressed as µg/g of biomass DW.

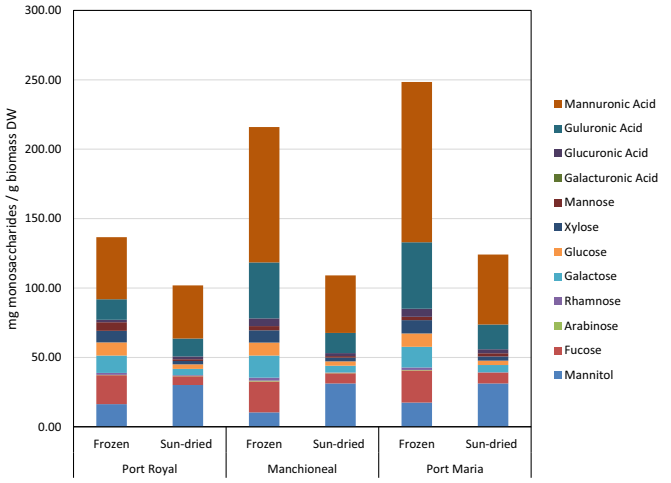


Fig. 4. Monosaccharide composition in the non-cellulosic fraction in frozen and sun-dried samples of pelagic *Sargassum* biomass harvested at three different locations. Results are expressed as mg/g of biomass DW.

ranged between 2.41 and 3.06, higher than values measured for individual morphotype harvested in February 2019 (values less than 2; Davis et al., 2021). This is due to the lower percentage of guluronic acid measured in the current biomass (Supplementary Table S7) compared to the study published earlier on. Differences were statistically supported between Port Royal and the other collection sites (both p-values <0.001), but not between Manchioneal and Port-Maria. Significant differences were also measured between frozen and sun-dried samples in Manchioneal and Port Maria (all p-values <0.001), but not in Port Royal.

When looking at individual monosaccharides other than those of alginates, mannitol, fucose and galactose were the most abundant in the frozen and sun-dried samples. Interestingly, the collection site did not show any influence on their contents. However, clear and significant differences were noticed when comparing frozen and sun-dried samples. Mannitol increased in sun-dried samples significantly (all p-values ≤ 0.012), to represent up to 25–30% of the monosaccharide content in this type of sample. This polyol is a well-known osmolyte in brown algae (Reed et al., 1985), and its production would have been enhanced by osmotic stress caused by sun-drying. In contrast, fucose (all p-values <0.001) and galactose (all p values <0.001) content was 2–3 fold higher in frozen compared to sun-dried samples. Changes in contents of most of the other monosaccharides investigated followed a trend similar to what was observed for fucose and galactose, except for arabinose and glucuronic acid in Port Royal samples.

3.8. Extraction and quantification of FCSPs using acid and enzyme-assisted methods

FCSPs correspond to homopolymers of sulfated L-fucose displaying numerous substitutions (fucans) and heteropolymers of galactose, mannose, xylose, and glucuronic acid containing sulfated L-fucoses mostly as side chains (fucoidans). These heterogeneous biopolymers are key components of the cell wall of brown algae and have many documented bioactivities in vitro (Deniaud-Bouët et al., 2017). Most of the studies reporting extraction of FCSPs from brown algae are based on a mild acid protocol, and therefore there are limited examples of comparing extraction of FCSPs using acid and enzyme-assisted extraction in these organisms. In addition, most of the information on FCSP content in pelagic *Sargassum* is very recent and provided only limited information on the composition of this cell wall polysaccharide (Rosado-Espinosa et al., 2020; Robledo et al., 2021; Vázquez-Delfín et al., 2021). In these studies, using mild acid or microwave assisted-extraction, the authors reported FCSPs representing between 4 and 11% of the biomass DW in individual species of pelagic *Sargassum* and in mix biomass samples.

Based on biomass harvested in Jamaica, and when comparing the different sampling sites and types of samples, at least five times more FCSPs were extracted by the enzyme-assisted method compared to the acid protocol (Fig. 5, supplementary Table S8). Indeed, FCSPs represented 10–20% of the biomass DW after enzymatic extraction, compared to 1–3.5% after acid extraction. Both methods provided higher contents of FCSPs from the sun-dried compared to the frozen samples, except for the acid extraction of Port Royal samples. Pair-wise comparison between frozen and sun-dried samples after acid extraction for each collection site show statistically supported differences (all p-values ≤ 0.012), while this is not the case for the enzyme-assisted extraction. When assessing potential differences in the quantities of FCSPs extracted at different sampling sites, significantly lower amounts were obtained in Manchioneal compared to Port Maria ($p = 0.001$) and Port Royal ($p = 0.014$) after enzyme-assisted extraction. For the acid extraction, the only statistical difference was observed between Port Maria and Manchioneal ($p = 0.041$). Differences in the quantities of FCSPs extracted according to the two methods considered maybe explained by the presence of compounds other than monosaccharides, such as proteins and phenolic compounds (Deniaud-Bouët et al., 2017; Wang et al., 2020). This would require further investigation.

One important characteristic of the FCSPs is their level of sulfation, as it has been shown to vary according to environmental conditions (Kloareg et al., 2021), and to influence their bioactivities (Oliveira et al., 2017). Therefore, the sulfate content was determined in the different FCSP extracts produced. As shown in Fig. 5 and Supplementary Table S8, the amount of sulfate in the acid extracted FCSPs (120–200 mg/g of extracted FCSPs) was approximately three times higher than after the enzyme-assisted extraction (40–60 mg/g of extracted FCSPs). This is in line with results obtained by Nguyen et al. (2020). No significant differences were observed when analysing the potential influence of sampling sites and types of samples on the sulfate content of the enzyme-assisted extracted FCSPs. For the acid extraction, differences were only significant between the frozen and sun-dried samples harvested in Manchioneal ($p < 0.001$). The sulfate contents determined in pelagic *Sargassum* samples are in the range of values reported for other brown algae (January et al., 2019).

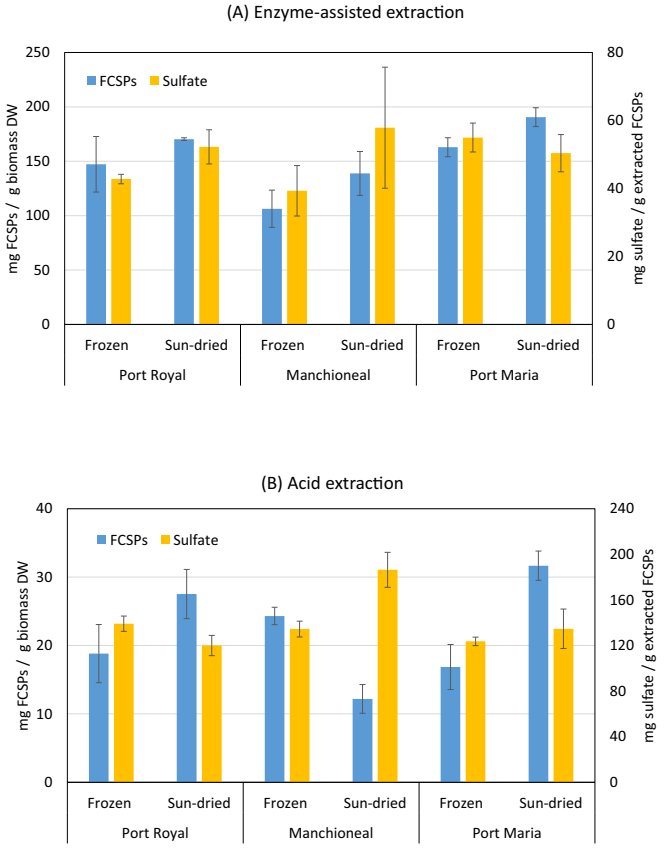


Fig. 5. FCSP content, and sulfate content in FCSPs, of frozen and sun-dried samples of pelagic *Sargassum* biomass harvested at three different locations and after enzyme-assisted (A), and acid (B) extraction. Results (mean ± SD) are expressed as mg FCSPs / g biomass DW, and as mg of sulfate / g of extracted FCSPs.

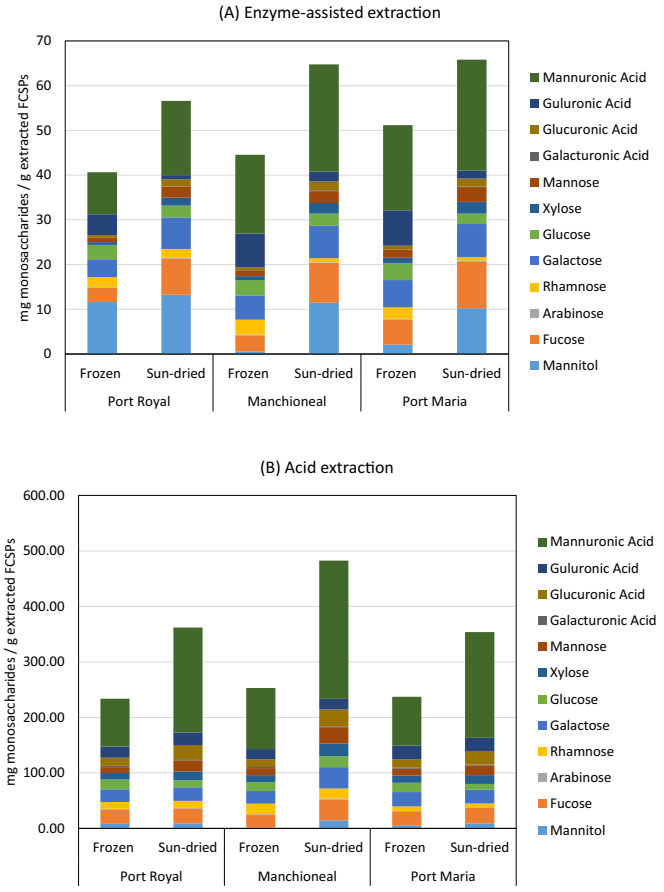


Fig. 6. Monosaccharide composition of FCSPs extracted from frozen and sun-dried samples of pelagic *Sargassum* biomass harvested at three different locations and after enzyme-assisted (A), and acid (B) extraction. Results (mean ± SD) are expressed as mg/g of extracted FCSPs.

The differences observed regarding quantities of FCSPs extracted and their sulfate content prompted the analysis of the monosaccharide composition of these extracts. Higher contents of monosaccharides were observed after acid extraction (234–483 mg/g FCSPs) compared to enzymatic extraction (40–66 mg/g FCSPs). (Fig. 6 and Supplementary Table S9). No statistically supported differences were observed after acid or enzymatic extraction according to sampling sites. However, significant variations were observed within each location between frozen and sun-dried samples after enzyme-assisted extraction (all p values ≤ 0.028), and after acid extraction except for Port Maria samples ($p = 0.062$; other p-values ≤ 0.042). When looking at individual monosaccharides, high levels of alginate were measured in all the FCSP extracts, representing between 30 and 60% of their dry weight. Mannitol was also abundant in some of the extracts obtained by enzymatic extraction, notably in Port Royal and sun-dried samples. The presence of these contaminants in FCSPs has already been noticed after extraction of fucoidans from other brown algae (Nguyen et al., 2020). Except for Manchioneal samples, no significant changes were observed in the content of fucose between frozen and sun-dried samples after acid extraction. In contrast, statistically supported increase of the quantities of fucose in sun-dried compared to frozen samples from the three sampling locations was observed after enzymatic extraction (all p-values ≤ 0.001). Similar trends in the enzyme extracts were observed for other known components of FCSPs galactose, xylose, mannose, and

glucuronic acid. All information gathered on pelagic *Sargassum* FCSPs suggests that different fucoidans were extracted using acid and enzymatic methods, and that sample processing may influence the composition of some of these extracts. Therefore, in-depth analysis is required to better understand the composition of the FCSP extracts, the structure of these polysaccharides, and to explore their bioactivities.

3.9. Pelagic Sargassum biomass macro-composition

Overall composition of this biomass is presented in Fig. 7 and Supplementary Table S10. The compounds analysed in this study covered 62–74% and 53–60% of the total content of biomass in frozen and sun-dried samples, respectively. Differences in the % of biomass DW between the two types of samples were statistically supported within the three sample locations (all p-values ≤ 0.022). Macro-composition of other brown algae has been analysed recently, including several species representing different orders of brown algae, and after sample freeze-drying (Olsson et al., 2020). Reported coverage values ranged between 66% for *Fucus vesiculosus* (a species member of the order Fucales, to which pelagic *Sargassum* species belong) and 88% for *Laminaria digitata* (order Laminariales). Analysis of pelagic *Sargassum* biomass macro-composition showed that sample processing using cheap (sun) or expensive (freeze) drying technique has a clear influence on the quantities of compounds extracted. During sun-drying, seaweeds are exposed to UV radiations and high temperatures that produce dehydration and affect the biology of these organisms. This process had a limited impact on ash, protein, and fatty acid content within most of the sample locations investigated. In contrast, it triggered an increase in the production of phenolic compounds, and it had a deleterious impact on fucoxanthin content and on the quantities of monosaccharides extracted from the non-cellulosic fraction, except for mannitol. Effects on FCSP content and composition depended on the method of extraction.

Influence of sample processing, and in particular comparison between frozen-freeze-dried and sun-dried samples has been previously assessed for several species of brown algae, including several benthic *Sargassum* species. Chan et al. (1997) analysed the composition in nutrients (amino acids, fatty acids, minerals, and vitamin C) of *S. hemiphyllum* after sun-, freeze-, and oven-drying. They concluded that the nutritional composition of this seaweed was greatly affected by the different drying methods. Charles et al. (2020) compared freeze- and sun-dried samples of *S. duplicatum*, and observed significant changes in colour characteristics and antioxidant activities between the two types of samples, with sun-dried samples containing more phenolic compounds, but showing less antioxidant activities compared to freeze-dried samples. In the same vein, Neoh et al. (2021) assessed the influence of six different drying methods, including freeze- and sun-drying, and of solvents, on the antioxidant content and bioactivities of *S. polycystum* extracts. They observed that the total phenolic contents in those prepared from freeze-dried samples were slightly higher than in those prepared from sun-dried samples, in contrast with our observations and those from Charles et al. (2020). However, they recovered at least 50% more carotenoids after freeze-drying compared with after sun-drying, similar to what was observed for pelagic *Sargassum* in our study.

4. Conclusion

Backtracking ensemble statistics indicate no clear differences in the provenance or oceanic exposure of *Sargassum* sampled from different sites and on different dates. Sample processing influences the biochemical composition of harvested biomass, and therefore its potential economic value. When comparing sun-dried and freeze-dried samples, only limited variations were observed in ash, protein, and fatty acid contents. However, sun-drying led to an increase of the quantities of phenolic compounds and of mannitol in the biomass, and to a decrease of its fucoxanthin and monosaccharide content. Influence of drying methods on the yield and composition of the FCSPs was dependent on the extraction procedure used for these polysaccharides. In this context, methods other than those used in our study, e. g., shade-drying and ensilage, should be investigated. In addition, during the last years, an increasing body of literature on different aspects of pelagic *Sargassum* has been published. However, there is still much to understand on the causes of the *Sargassum* influxes and the biology of these organisms to establish processes and policies for the exploitation of this natural feedstock.

Most of the studies published so far on morphotype abundance and biomass composition of pelagic *Sargassum* have been conducted in the Caribbean. There is a need to extend this to Western Africa, the other region highly impacted by pelagic *Sargassum* events. It is important to assess if the most abundant morphotype identified during beaching events in the Caribbean during the last years, i.e., *S. fluitans* III, is also the most abundant in Western Africa. This would provide valuable information on the seasonal and regional evolution of *Sargassum* biomass composition. It will also be interesting to target mats in the sea, and follow them up to the beach, to be able to compare potential changes of morphotype abundance and of biochemical and elemental composition during their drift.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.152761.

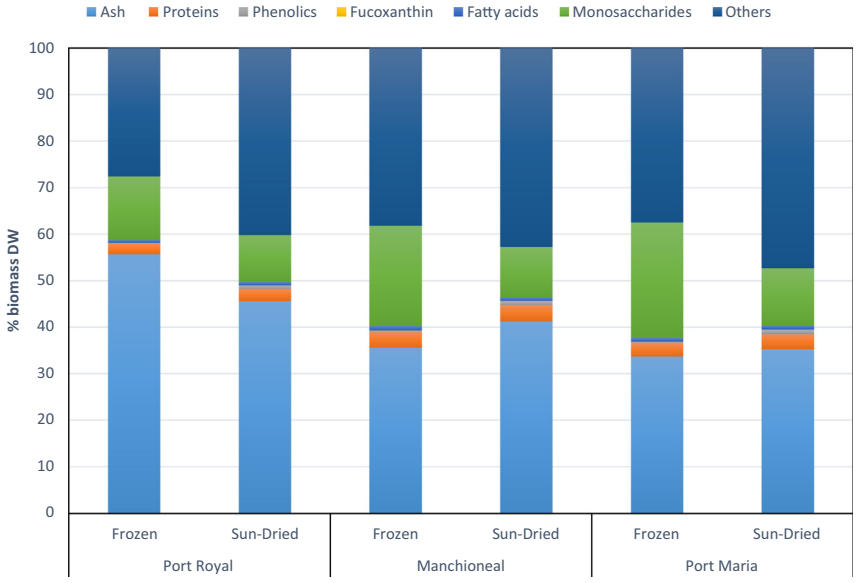


Fig. 7. Major constituents of pelagic *Sargassum* biomass investigated in this study. Results are expressed as weight % biomass DW.

CRediT authorship contribution statement

Carla Botelho Machado: Investigation, Formal analysis, Visualization, Writing – review & editing. **Gina-Marie Maddix:** Investigation, Formal analysis, Writing – review & editing. **Patrice Francis:** Investigation, Formal analysis, Writing – review & editing. **Shanna-Lee Thomas:** Investigation, Formal analysis, Writing – review & editing. **Jodi-Ann Burton:** Investigation, Formal analysis, Writing – review & editing. **Swen Langer:** Investigation, Formal analysis, Writing – review & editing. **Tony R. Larson:** Investigation, Formal analysis, Writing – review & editing. **Robert Marsh:** Conceptualization, Funding acquisition, Investigation, Formal analysis, Visualization, Writing – review & editing. **Mona Webber:** Conceptualization, Funding acquisition, Investigation, Formal analysis, Visualization, Writing – review & editing. **Thierry Tonon:** Conceptualization, Funding acquisition, Investigation, Formal analysis, Visualization, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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