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Sargassum contamination and consequences for downstream uses: a review

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Abstract

Sargassum spp. are brown algae present in the tropical and subtropical waters of the world's oceans. Benthic or pelagic, *Sargassum* spp. represent substantial amounts of biomass when drifted and massive beaching events also named "golden tides" are challenging for environmental and tourism managers. The *Sargassum* spp. biomass can be regarded as offering economic opportunities; however, micropollutant contamination counteracts this wealth. This review describes the contaminant uptake processes and the concentrations reached by various *Sargassum* species in regard to national and international norms for a variety of applications. Amongst the heavy metals, perhaps total arsenic content is the most salient: phytoaccumulation of arsenic is due to confusion in the phosphate transporter between arsenic and phosphate, thus leading *Sargassum* spp. to bioaccumulate arsenic actively. The levels reached could well give cause for concern across all potential applications, in particular for human and animal use. Similarly, there are concerns for the widespread applications or disposal of this biomass due to environmental impacts in the case of storage, landfill or composting. Organic micropollutants are too rarely studied to assert that there is global contamination. However, research studies focused on *Sargassum* spp. contamination confirmed environmental threats. But, without exhaustive analysis of routine algae contamination, the standard contamination level cannot be defined. Treating beach-cast or drifting *Sargassum* spp. as a waste is a Gordian knot, in that it involves investment, work and disposal surfaces, whilst not necessarily providing environmental and economic gains. However, *Sargassum* spp. are a raw material, a resource rich in a broad variety of constituents that could be processed for commercial applications or which could be used for production of energy. In this paper the authors review the diverse applications that can be considered and describe the relevant regulations and norms related to contaminants and effluents that need to be taken into account. However, if valorizing *Sargassum* spp. is currently valuable and promising, the quality of the raw material is a key starting point because fresh and sand-free algae are required: offshore gathering solutions need to be developed in order to limit *Sargassum* sp. contamination by pollutants from land so as to hinder algae decay.

Keywords *Sargassum* · Phaeophyceae · Valorization · Regulatory · Standards · Heavy metals · Arsenic · Extracts

Introduction

In the global context of environmental disruption, proliferative species can induce loss of habitat-forming species and may lead to significant changes in the structure and functioning of ecosystems and to species extinctions. Not only do macroalgae make up a large component of proliferative and/or invasive species, but they also cause significant economic and environmental damage, over which there are only limited post-invasion control and management options (Ribera and Boudouresque 1995; Thresher 1999; Schaffelke et al. 2006; Anderson 2007; Schaffelke and Hewitt 2007; Leong et al. 2018). Amongst the macroalgae, *Sargassum* spp. have been used in Asia for decades and sometimes cultivated for their large spectrum of potential applications, including food, agriculture, medical applications or energy (Holdt and Kraan 2011; Milledge and Harvey 2016; Janarthanan and Senthil Kumar 2018; Latifah and Warganegara 2018; Taghavi et al. 2018, amongst many). Nonetheless, concerns persist because of the increasing reports of invasion in different regions worldwide.

Sargassum horneri and *Sargassum muticum* from Japanese and Korean shores have been shown to be invasive

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(Schaffelke et al. 2006; Valentine et al. 2007; Milledge and Harvey 2016; Belattmania et al. 2018a; Marks et al. 2018). *Sargassum horneri* is invasive in Taiwan (Sung et al. 2018), in Morocco (Belattmania et al. 2018a, b) and on Algeria's shores (Aouissi et al. 2018). *Sargassum polycystum*, from the South China Sea, became established in Oceania (Andréfouët et al. 2017) and *S. muticum* is reported on the Great Barrier Reef (Vye et al. 2018) and is broadly distributed across Europe (Harries et al. 2007; Josefsson and Jansson 2011). Moreover, global changes might also promote *Sargassum* sp. proliferation (Széchy et al. 2017; Graba-Landry et al. 2018; Olabarria et al. 2018).

The pelagic *Sargassum fluitans* and *Sargassum natans* were reported in the North Atlantic Ocean gyre since Christopher Columbus on his first trip (1492). Later on, another "Sargasso sea" was encountered in the South Atlantic by Dutch/Portuguese sailors but has been little documented in this early description (Lodewijcksz and Claesz 1609). Over the last decade, very large amounts of pelagic *Sargassum* spp. (expected to be millions of tonnes in a decade) have been beached on Caribbean, Gulf of Mexico and West African coasts, with increasing frequency and intensity. Besides their massive impacts on local ecosystems and economies and major health concerns, the valorization of the "golden tide seaweeds" could generate revenues contributing to clean-up operations, and develop new business opportunities worldwide for proliferating pelagic and benthic *Sargassum* spp. (Pérez-López et al. 2014; Van Hal et al. 2014; Milledge and Harvey 2016).

However, such opportunities have to be developed within the local regulatory framework relevant for each of these applications (i.e. biochemicals, feed, food, fertilizers, materials and energy). Whilst *Sargassum* sp. use is often not regulated per se, products developed from the biomass have to comply with regulations and norms of use. Whilst a review published on the use of various seaweeds in functional food by Holdt and Kraan (2011) remains relevant for *Sargassum* spp., a broader review across applications, taking into account the specificities of *Sargassum* spp., is still missing. This review focuses on members of the genus *Sargassum* and particularly the species *S. fluitans* and *S. natans*, in order to (i) access current knowledge on the content of potential pollutants, (ii) describe the proportion of valuable compounds, (iii) identify the current or putative applications and (iv) address compatibility with those regulations in force, in the countries affected.

Pollutants of *Sargassum* spp.

This section details the concentrations (in the context of pollutants) of possible micropollutants detected in *Sargassum* spp. and, when available, the sorption processes proposed for pollutant intake and/or accumulation (Pérez-Larrán et al. 2019).

Bioaccumulation of a pollutant involves both intake and adsorption to retain it within the biota. Severe bioaccumulation is observed when adsorption protects from degradation or metabolization. The following section identifies the remarkable intake processes and retention capacities of particular types of seaweed biomass.

Organic micropollutants

Organic micropollutants in *Sargassum* spp. are poorly documented. Organic micropollutants could be adsorbed by organisms because of their physical properties, for instance the surface/volume ratio (Torresi et al. 2017), or a 3D structure which can limit exchanges (Woignier et al. 2011), or due to the chemical composition of the algae, i.e. organic carbon content (Karickhoff et al. 1979).

Polycyclic aromatic hydrocarbons Polycyclic aromatic hydrocarbons (PAHs) are produced by human activities (fuel combustion within a closed system: the petrogenic PAHs) or due to combustion. Fires are due to human action or natural events (the pyrogenic PAHs): human ignition seems to predominate as a source of pyrogenic PAHs also (Marr et al. 1999; Larsen and Baker 2003; Stark et al. 2003). Whatever their source, both petrogenic and pyrogenic sources contribute to PAHs in seaweed samples (Neff et al. 2005; Morillo et al. 2008).

Adsorption of PAHs is driven by their chemical affinity for organic carbon and hydrophobic content: the PAH aromatic level is correlated with PAH hydrophobicity, and the latter has been shown to be correlated to adsorption strength (Madenjian et al. 1999). Since labile molecules, i.e. freely dissolved molecules, are more bioavailable than adsorbed ones, PAH aromaticity is also correlated with PAH persistence in the environment (Bertilsson and Widenfalk 2002; Tobiszewski and Namiesnik 2012).

Stout et al. (2018) reported that the offshore Deepwater Horizon MC 252 oil well blowout in the Gulf of Mexico and the subsequent oil spill were correlated to beachings of pelagic *S. fluitans* and *S. natans*. Data on *Sargassum* sp. contamination from the Gulf of Mexico showed that in an oil spill situation (so too a non-oil spill situation), the measured variability in concentrations was high (Table 1). Stout et al. (2018) revealed lower PAH contamination in *S. natans* and *S. fluitans* in offshore drifting thalli, as compared to onshore samples of the same species, despite a recent massive oil spill that had occurred a few months before their experiments. In another study, Seepersaud et al. (2018) studied PAHs in *S. filipendula* and *S. vulgare* from contrasted locations in Trinidad and Tobago, with oil extracted from Trinidad shallows and Tobago being considered as a relatively preserved island. They demonstrated that the biogeographical effect was dominant and dependent on human activity on the shore.

Table 1 Contamination of *Sargassum* spp. by polycyclic aromatic hydrocarbons

	Naphthalene	Acenaphthylene	Acenaphthene	Fluorene	Phenanthrene	Anthracene	Dibenzothiophene	Fluoranthene	Pyrene	Naphthobenzothiophenes	Benzo(a) anthracene	Chrysene
Seepersaud et al. (2018)	5.95–25.53	18.06–59.35	14.57–112.42	5.73–53.74	n.d.–42.93	9.12–59.9		4.29–62.12	8.33–181.23		n.d.	n.d.
Stout et al. (2018)												
Oil spilled	n.d.–35.33	n.d.	n.d.	n.d.–149.6	2.41–57.12	n.d.	n.d.–237.6	0.49–2.6	0.34–11.9	n.d.–194.4	n.d.–0.98	n.d.–421.6
Non-oil spilled	n.d.–10.17	n.d.–0.07	n.d.–0.31	n.d.–0.21	0.29–0.65	n.d.–0.06	n.d.–0.37	0.14–0.57	0.13–0.46	n.d.–0.06	n.d.–0.11	n.d.–.04

	Benzo[a] fluoranthene	Benzo(b) fluoranthene	Benzo(k) fluoranthene	Benzo(a) pyrene	Benzo(e) pyrene	Perylene	Indeno[1,2,3-c,d]pyrene	Dibenz(ah) anthracene	Benzo(ghi) perylene	Hopane	∑PAHs
Seepersaud et al. (2018)		n.d.	n.d.–42.66	11.6–289.35			n.d.–45.84	n.d.	n.d.–54.69		287.4–605.1
Stout et al. (2018)											
Oil spilled	n.d.	n.d.–26	n.d.	n.d.	n.d.–30.69	n.d.	n.d.–1.15	n.d.–0.56	n.d.–4.25	5.78–70.37	84–5162
Non-oil spilled	n.d.	n.d.–0.3	n.d.–0.22	n.d.–0.19	n.d.–0.28	n.d.	n.d.–0.13	n.d.	n.d.	n.d.–3.1	0.56–17.3

Individual PAH concentration data obtained (courtesy of the authors of Stout et al. (2018)) for Deepwater Horizon consequences)

Polychlorobiphenyls Polychlorobiphenyls (PCBs) have a hydrophobicity that increases with chlorination (Madenjian et al. 1999). James et al. (1987) found no lasting toxic effect on *S. agardhianum* and *S. muticum* using 2 μg L^{-1} PCB in seawater. Such concentration in seawater is high, considering the dilution effect due to the oceanic context. However, PCB contamination impacts *Sargassum* edibility. Indeed, it is known that hijiki, as *S. fusiforme* is named in Japan, where it is used for food, can be contaminated by PCB, and blood serum results for Japanese women indicate PCB contamination with hijiki consumption (Haraguchi et al. 2016).

Pesticides The sorption behaviour of pesticides is rather complex. Even if hydrophobicity broadly induces similar behaviour, especially for older formulations, more recent pesticides are partly designed to delude plant metabolism. In Martinique, French West Indies, studies have been carried out on the chlordecone threat. Chlordecone, a persistent organic pollutant used decades ago as insecticide by banana plantation, accumulated in *S. natans* and *S. fluitans* reaches equilibrium after 48 h of deployment on the surface of contaminated bays (Martial and Tirolien 2018; Devault et al. 2019, 2020). Projects on *Sargassum* sp. contamination by chlordecone in the French West Indies are currently under way.

Hydrazine Hydrazine effects were reported on *S. agardhianum* and *S. muticum* sporophytes, with mortality in 1 to 3 weeks (James et al. 1987). Mitochondria which appeared to be abnormal were also reported by North and James (1987) after a 4-day exposure to 0.25 μg L^{-1}, suggesting that hydrazine might induce membrane injuries and arbitrary reorganization of cell content.

Micropollutant sorption on to *Sargassum* sp. thalli

As mentioned by Li et al. (2017a), the biosorption potential of *Sargassum*-derived products was extensively studied and well demonstrated for metals, but data are scarce for organic compounds. Amongst the few studies, most of them used non-living *Sargassum* sp. biomass and sometimes harsh conditions. For example, Rubín et al. (2006) studied the biosorption of phenolic compounds (phenol, 2-chlorophenol and 4-chlorophenol) on dried *S. muticum* at 60 °C, Chung et al. (2007) worked on phenanthrene sorption on dried *S. hemiphyllum* at 50 °C, Li et al. (2017b) worked on tetracycline adsorption using carbon particles from seaweeds, Tabaraki and Nateghi (2016) worked on hydrazine sorption on dried and sieved *S. ilicifolium* and Liang et al. (2017) worked on the sorption of methylene blue by non-living *S. hemiphyllum*. The processes of sorption of hydrophobic organic compounds have mainly been studied from the point of view of their industrial removal, but the equivalent processes in natural conditions are essentially unknown. Meanwhile, the available results are very useful for understanding a number of sorption behaviours.

Rubín et al. (2006) indicated that the presence of –OH, –COOH, C=O and C–O groups supported the adsorption of phenolic compounds: such groups activate the aromatic ring of monochlorophenols by chlorine, which favours the existence of donor-acceptor interactions between the phenolic compounds and the groups of the biosorbent surface.

Chung et al. (2007) worked on phenanthrene sorption, showing that it fits with a pseudo-second-order kinetic. Phenanthrene is a polycyclic aromatic hydrocarbon with a low hydrophobicity for this family of about 4.57. Optimal hydrazine sorption on *S. ilicifolium* was shown to have a pH of 7.5, i.e. an in situ compatible pH.

Methylene Blue is a specific pollutant from the dye industries. Liang et al. (2017) demonstrated that the Langmuir isotherm model provided the best correlation with experimental data on *S. hemiphyllum*, and that the monolayer biosorption capacity was 729.93 mg g^{-1} within 2 h but at acidic pH (pH = 5) mainly due to surface hydroxyl, amine and carboxyl functional groups.

Li (2015) and Li et al. (2017a, b) noted tetracycline sorption onto *S. hemiphyllum*, fitted with the pseudo-first-order model (Langmuir) up to 278.4 mg g^{-1}. Tetracycline is not hydrophobic ($\log K_{OW} = -1.37$) but interacts strongly with carbon particles: equilibrium is reached after a 2-h exposure. Solution pH and background ionic strength are important controlling parameters in the adsorption process as they affect the tetracycline speciation because, under acidic conditions, the amphoteric form promotes sorption. In such conditions, the surface binding sites of the layered carbon particles promote adsorption ability whilst outer-sphere association suppresses adsorption whilst increasing ionic strength.

Overall, organic micropollutants tend to show rapid and massive adsorption on modified *Sargassum* spp., driven by pH, in accordance with adsorption at in situ pH, at least for hydrophobic ones. In situ, *Sargassum* spp. could be expected to have a high capability of bioaccumulating organic compounds but probably not because of the seaweed organic contents (Gramatica et al. 2007). In general, the absence of strict correlation between K_{OW} and adsorption suggests that more studies are urgent to better understand the nature and the localization of sorption sites for seaweeds.

Heavy metals

The heavy metal content, as reported in selected references, in *Sargassum* spp. is summarized in Table 2.

In brief, metals mainly contaminate biota via cationic species. At the pH under consideration, heavy metal forms are summarized in Table 3. In the light of the many experiments performed to determine if *Sargassum* spp. could be an effective material for heavy metal removal, heavy metal speciation has to be examined at seawater pH. For marine biota, it has been proposed that heavy metal speciation intrinsically drives bioaccumulation potential. At seawater pH, even if Cd is almost fully available, Zn or Cu are not present in their most bioavailable species form, and the other species mentioned are only present in low-bioavailability forms. Bioavailability is therefore an important criterion for understanding bioaccumulation in *Sargassum* spp. and in parallel with biosorption, active transfer mechanisms need to be considered.

Indeed, metal sorption on *Sargassum* spp. is not homogeneous, as explained by Davis et al. (2000) and Schiewer and Wong (2000). Metal selectivity for sorption is based on their physico-chemical properties, i.e. conformation and ion strength, and on α-L-guluronic acid content. Miao et al. (2014) observed the histological impact of heavy metal on *S. pallidum* ultrastructure in samples exposed to in situ contamination They showed that cell structures and walls are deformed and even broken, extra-numbered vacuoles appear, chloroplast grana are distorted or even disintegrated and stroma are disrupted and even leak (Miao et al. 2014). Losses of selective permeability can also induce alteration of metabolic pathways such as photosynthesis or oxidative mechanisms (Cabral 2003; Carreras and Pignata 2007; Zou et al. 2015). A domino-like effect for metals was proposed by Patra et al. (2004) with (1) weakening of cell membrane permeability, (2) reactions of sulphydryl groups with cations, (3) affinity for reacting with phosphate groups and active groups of ADP or ATP and (4) replacement of essential ions.

In *Sargassum* spp., heavy metal accumulation in cell walls and the role of alginate-rich (20–40%) materials have been reported in many studies (Figueira et al. 1997; Kratochvil and Volesky 1998; Davis et al. 2000, 2003a, b, c; Sheng et al. 2004; Naja and Volesky 2006). Algal cell walls allow the free passage of molecules and ions in aqueous solutions (Hope and Walker 1975). Algal cell walls have metal-binding groups, which can explain divalent cation binding (Puranik et al. 1999; Khoo and Ting 2001; Chen et al. 2002, amongst many). In addition, the cell walls of brown algae contain metal-sequestering alginate and fucoidan (Figueira et al. 1999; Davis et al. 2000). *Sargassum* species present a two-stage kinetic behaviour: very rapid initial sorption over a few minutes, followed by a long period of much slower uptake until equilibrium is reached after 1 to 3 h. In general, about 90% of total metal ion sorption was achieved within the first 60 min. Based on these results, in equilibrium experiments, 6 h was often deemed sufficient to establish equilibrium (Sheng et al. 2004).

Many experiments with different *Sargassum* species have used pseudo-second-order Freundlich, Langmuir and Dubinin-Radushkevich isotherms to fit sorption kinetics: see Fan et al. (2018a) for *S. hemiphyllum*, Foroutan et al. (2018) for *S. oligocystum*, Negm et al. (2018) for *S. vulgare*, Perez et al. (2018) for *S. tenerrimum*, Tukaram Bai and

Table 2 Heavy metal concentration in members of the genus *Sargassum* by species

Species	Al	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn	V	As	Se	Reference
S. binderi		0.76	0.6	3.7	10.2	30.6	3.55	2.8	2.4	2.8	0.9			Al-Shwafi and Rushdi (2008)
S. cinctum		0.07	2.1	0.9	8.3	224	14.8	0.4	0.9	23.2		78.8		Chakraborty and Owens (2014)
S. crassifolium		0.38				1.4		1.5	1.1	1.7				Denton and Burdon-Jones (1986)
S. dentifolium		0.98– 2.1	1.8–3.5	0.6– 1.2	1.5–5.2		6.7 –27.1	2.1–8.7	4.5– 25.8	4.5– 55.3				Abdallah et al. (2005)
S. dentifolium		n.d.-0.72	n.d.-28.39		0.84-9.36	85-1178.5			n.d. -129.64	49.43-7.25				Mohamed (2005)
S. elegans			0.2-1.5	0.41- 3.4	7.43-43.2	111.5-189.77	7.45 -10.27	1.37-3.03		7.16-23.33			0.3-1.47	Magura et al. (2016)
S. filipendula and S. vulgare		1.77-3.65			4.33-13.30	65.79-1237.37	20.2 -134.46	7.07-12.08	4.61 -12.11	12.61-194.15				Seepersaud et al. (2018)
S. fluitans and natans		0.18-10.63	0.25-11.7	0.29-167.1	0.9-68.5			1.53-7.64	0.08-4.37	>1-44.8		5.4-156.1		Devault et al. (2020)
S. fusiforme	147 ±7	0.79 ±0.02		3.4 ±0.02	1.55 ±0.07	311±11			0.43 ±0.03	13.4 ±0.5				Ismail et al. (2017)
S. fusiforme		1.71 ±0.17		0.85 ±0.11	7.93 ±0.89			2.87±0.38	1.5 ±0.62	20.87 ±6.69		57.71 ±13.44		Pan et al. (2018)
S. hemiphyllum		0.6 ±0.1				4.4 ±1.3	62 ±86	10.2 ±1.7	6±2.7	21 ±6.9				Ho (1988)
S. kjellmanianum					1.27-5.66		15.4 -83.8			11.8 -25.4				Hou and Yan (1998)
S. latifolium								24.7-36.2						Al-Homaidan (2007)
S. latifolium								31.6 ±6.4						Al-Homaidan (2008)
S. latifolium		0.643-0.93	0.89-36.54		4.65-8.75	115-1190.3			2.92 -161.43	15.18-46.52				Mohamed (2005)
S. linifolium		0.161± 0.037	12.5±1.5	134.0 ±11	29.9 ±3	39.2 ±3.2		39.2±3.5		99 ±3.5				El-Said and El-Sikaily (2013)
S. muticum		0.26 ± 0.01			0.96 ±0.15	44.45 ±4.32	188 ±7.86	0.86 ±0.2	1.1±0.01	311 ±11.6				Favero and Frigo (2002)
S. pallidum					3.06-4.81				2.36 -18.41			50.16-261.73		Miao et al. (2014)
S. sinicola					36.2					30				Sanchez-Rodriguez et al. (2001)
S. stenophyllum		2.01± 0.33								580 ±68				Amado Filho et al. (1999)
S. stenophyllum		0.3-2.01												Amado Filho et al. (1999)
S. stenophyllum		0.37-0.25			2.2-3.4	2.5-3.8			2.9-4.4	23.7 -108				Karez et al. (1994)
S. subrepandum		0.1-5			2-85			0.5-50	2-51					Alahverdi and Savabieasfahani (2012)
S. swartzii		n.d.-0.311		0.006- 0.334	0.03-0.291		0.118 -7.736		n.d.-0.165	0.019-5.537				Kaviarasan et al. (2018)
S. tenerrimum		<10			<20	110±2	<10			<5				Jadeja and Tewari (2007)
S. thunbergii					3.39		92.5			19.4				Hou and Yan (1998)
S. thunbergii		5.63 ±0.12		3.84 ±0.47	12.81 ±0.83			6.04±0.53	2 ±0.24	40.69 ±4.67		49.08 ±2.46		Pan et al. (2018)
S. vachellianum		1.1 ±0.3				5 ±1	129 ±83	5.4 ±0.9	10.6 ±1.4	63 ±34				Ho (1988)
S. vachellianum		8.48 ±0.29		1.21 ±0.12	8.59 ±0.23			3.11±0.38	1.9 ±0.39	81 ±12.9		23.77 ±3.88		Pan et al. (2018)
S. vulgare		0.16 ±0.03			23.1 ±0.1	4.7 5 ±0.18			1.04 ±0.05	23.74 ±1.19				Al-Masri et al. (2003)
S. vulgare									5.03– 18.6				1.08– 1.47	Lozano et al. (2003)
S. thunbergii					1-2000					20 -1000		20- 2000		Li (2015)
Sargassum sp		0.40– 1.45		1.5– 9.0	6.0– 16.8		93.7– 334.9	8.5–9.7	6.2– 11.1	13.5– 27.1				Amado Filho et al. (2008)
Sargassum sp.		1.25-2.38		1.2-2.6	2.8-7	45-550	12 -89.5	2.2 -9.9	3.9 -6.5	34-41				Horowitz and Presley (1977)
Sargassum sp.		2.4 ±0.38		88 ±0.44	8.8 ±0.44	50.19±3.83	97±7.4	3.1±1.24	69.51 ±10.34	200 ±10.14				Roberts et al. (2008)

Table 2 (continued)

Species	Al	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn	V	As	Se	Reference
Sargassum sp.		0.07				2.1	38	13.7	0.02	18.7				Sawidis et al. (2001)
Sargassum spp. Brito et al. (2012)		0.381-0.719	178-189	4.41- 6.94	4.75-15.5		81.2- 83.2	7.47-9.93	3.02 -3.12	25.1 -26.6	14.1	-25.1	16.1 -19.7	
Sargassum spp.		0.264– 0.91	168– 191	2.77– 7.77	4.33– 16.1		42.9– 123	6.11– 10.1	1.70– 4.33	25.0– 26.9	11.1 – 30.-2		9.95– 29.5	Brito et al. (2012)
Sargassum spp.		0.39				2.4		3.6	2	8.1				Denton and Burdon-Jones (1986)
Sargassum spp.			0.238– 2.07	0.699- 6.05			15.4– 124			11.8 – 38.7		65.3 -182.8		Hou and Yan (1998)
Sargassum spp.	334.95-10129.4	0.027-0.212	0.072-11.857	1.576-37.973	2.188-23.092	638.582-22013.5	4.623-248.84	0.757-21.524		3.812-94.376				Okuku and Peter (2012)

Venkateswarlu (2018) for *S. crassifolium*, Wang et al. (2018a) for *S. horneri* and Jalali et al. (2002), Lee and Volesky (1999) and Yang and Volesky (1999) for *S. fluitans* and *S. natans*. The Freundlich isotherm involves one layer of strong chemical-dominant adsorption pattern, the Langmuir isotherm involves two layers of weaker physical-dominant adsorption pattern and the Dubinin-Radushkevich isotherm involves physical adsorption. The two-stage behaviours could be explained by a combination of adsorption types due to the heterogeneity of the cell walls rather than the size variation of the biomass (Moino et al. 2020) and are in agreement with the second Fick's law concerning adsorption. Various functional groups might serve as sorption sites, differing with respect to the strength of the metal sorptive bond, as well as the rate of adsorption on these sites (Schiewer and Volesky 2000).

Nano-metal particles were shown to be scattered through the cytoplasm, and alginate is proposed to favour nanoparticle formation (Fig. 3), which, in turn, might induce wall damage. Miao et al. (2014) suggested that nano-metal particles could have a sequestration effect and may limit deleterious impacts. Such high-electron density deposits are rich in C, O, Cl, Cu, Pb, As and Hg. In contrast, phenol beads are 4–6 μm in diameter in normal conditions; in reported contaminated conditions, phenol masses are irregularly shaped and undersized (< 1 μm) (Miao et al. 2014).

The carboxyl groups of alginate play a major role in the complexation. Overall, it was shown that different species of algae and conspecific algae may present different adsorption capacities, but unfortunately, in situ experiments of heavy metal sorption on living *Sargassum* spp. have never been reported and are urgently needed for the benthic species *S. fluitans* and *S. natans*.

Arsenic

Arsenic concentrations found in *Sargassum* spp. are summarized in Table 4. Arsenic is a notorious and toxic metalloid, ubiquitous in the environment. Arsenate (As(V)) is the predominant inorganic form in aqueous and aerobic environments and is strongly adsorbed onto the surface of several aquatic organisms and oxidized minerals of Fe, Mn and Al (Al Mamun et al. 2019). Arsenite (As(III)) is highly abundant under anoxic environments and is adsorbed weakly onto fewer minerals, making it a more mobile oxyanion (Smedley and Kinniburgh 2002). As(III) is expected to be 20 to 60 times more toxic than As(V) due to its tendency to react as a soft metal with thiols (Ferguson and Gavis 1972). Arsenic forms can change rapidly in the environment from one species to the other. However, if organic arsenic compounds are formed by As methylation, As species changes are no longer possible. *Sargassum* spp. are known for their massive arsenic content. Several studies on *Sargassum* spp. led to the proposal that As(V) is mistaken for phosphate, owing to physico-chemical similarities between H_2PO^{4-} and H_2AsO^{4-}, and is transported

Table 3 Heavy metal speciation at similar pH

Metal	Species	References
Lead (Pb)	$Pb(CO_3)_2$ [100%]	Powell et al. (2009)
Zinc (Zn)	Zn^{2+} [55%]; $ZnOH^+$ [35%]; $Zn(OH)_2$ [10%]	Sjöberg et al. (2013)
Chromium (Cr)	$Cr(OH)_2^+$ [45%]; $Cr(OH)_3$ [45%]	Dos Santos et al. (2011)
Cadmium (Cd)	Cd^{2+} [97%]; $Cd(OH)^+$ [3%]	Powell et al. (2011)
Copper (Cu)	$CuOH^+$ [60%]; Cu^{2+} [17.5%]; $Cu(OH)_2$ (aq) [17.5%]; $CuSO_4^-$ [5%]	Smith et al. (2015)
Arsenic (As)	As(III): $H_2AsO_3^-$ [90%]; H_3AsO [10%]–As(V): $HAsO_4^{2-}$ [95%]; $HAsO_4^-$ [5%]	Sø 2011
Nickel (Ni)	$NiCO_3$ (aq) [3%]; $Ni(CO_3)_2^{2-}$ [97%]	Pyle et al. (2002)
Mercury (Hg)	Hg(II): $Hg(OH)_2$ (aq) [100%]	Powell et al. (2005)

by phosphate transporters (Taylor and Jackson 2016). The competition between P and As(V) for absorption sites may induce a drop in the concentration of P within algal tissues and an increase in As(V) concentrations. In contrast, Benson (1984) hypothesized that *Sargassum* spp. present higher levels of As(V) when a lower phosphate level is available. On the other hand, the interference of As(III) over uptake might not be inhibited by variations in uptake pathways: P absorption was found to be independent in the presence of As(III). As(III) is thought to be transported through an aquaglyceroporin channel as neutral $As(OH)_3$ (Meharg 2004). To explain low phosphate accumulation in biota, Benson et al. (1988) hypothesized that this was because arsenate is much less stable than phosphate, and biological reducing agents can reduce arsenate, whilst reduction of phosphate is not possible.

Whilst there are proposals regarding the mechanisms involved, arsenic retention in *Sargassum* spp. is not fully understood. Studies have been conducted to understand the fate of arsenic in algae, including *Sargassum* spp. In marine environments, inorganic arsenic present in seawater was shown to be transformed into organo-arsenic species, mainly arsenoribosides in algae and arsenobetaine in animals (Maher et al. 2009). In macroalgae species, degradation of such less toxic forms leads to thio-arsenoribosides, dimethylarsinoylethanol (DMAE), dimethylarsenate (DMA) and methylarsenate (MA). As(V) can be recycled in seawater due to microbial communities (Duncan et al. 2014), in contrast to the fate of arsenobetaine, which is known to bioaccumulate in biota (Duncan et al. 2015).

In studies of arsenate metabolism, it has been proposed that a primary reaction with adenosine triphosphate (ATP) yields the unstable phosphoric arsenic anhydride (APAs), whose brief lifetime restricts its metabolic capabilities. Benson et al. (1988) interpreted the latter as a reduction of this fugacious intermediate by adenylyl sulphate reductase, or possibly a more specific enzyme. Reductions of arsenate and methyl arsenate to arsenous acid or methylarsenous acid (arsine oxide or methylarsine oxide) proceed in the presence of chloroplasts in light (Hill reaction) (Benson et al. 1988). The arsenous acids or their dehydrated forms, the arsine oxides, seem to react easily with accessible –SH groups to form arsenous thioesters. The nature of the thiol determines the stability and exchange rates of such esters. Monothioesters of arsenous acid seem to be able to further react and, with a second thiol, form arsenous dithiol esters which are also exchangeable with free thiols. Dithiols with proper spatial relationships, such as lipoic acid, 2,3-dimercaptopropan-1-o1 (BAL) or dithiothreitol, form much more stable and difficulty-exchangeable arsenous dithiol esters. Thus, dithiothreitol removes bound arsenous acids from monothiol ester linkages on algal proteins to produce a soluble cyclic arsenous dithiol ester. This metabolic pathway was proposed to predominate in *Sargassum* spp. but considered as marginal in other algae (Benson et al. 1988).

As indicated by García-Sartal (2012), brown seaweeds contain higher amounts of total arsenic than other seaweed classes (Dawczynski et al. 2007). Arsenosugars, i.e. the main arsenic compounds found in seaweed, are due to complex biosynthesis pathways which are still to be explored. As detailed elsewhere, arsenate absorption metabolism by seaweeds is similar to that reported for phosphate. Arsenosugars could

Table 4 Arsenic concentration diversity in *Sargassum* by species

Reference	Species	Arsenic (μg g^{-1})
Edmonds et al. (2000)	*Sargassum lacerifolium*	2.0–40
Han et al. (2009)	*Sargassum fusiforme*	65.3–84.1
Pichler et al. (2006)	*Sargassum horneri*	114.3 ± 27
Leal-Acosta (2013)	*Sargassum sinicola*	62–640
Leal-Acosta (2013)	*Sargassum sinicola*	32–80
Maher 1983	*Sargassum fallax*	64–123
Ruiz Chancho (2010)	*Sargassum ramifolium*	11–66.3
Ruiz Chancho (2010)	*Sargassum vulgare*	16.1–58.8
Ruiz Chancho (2010)	*Sargassum fulvellum*	115 ± 9
Rose et al. (2007)	*Sargassum fusiforme*	18–124
	Sargassum fusiforme	37–146

be the end products of a detoxifying mechanism, due to reduction into DMA(V) with the *S*-adenosylmethionine acting as a methyl group donor (Craig 2003), then reduced glycosylation to DMA(III) due to the adenosyl group of *S*-adenosylmethionine and a glycosylation, leading to dimethylarsinoylribosides (Craig 2003; Murray et al. 2003) (Fig. 2). Arsenosugarphospholipids could be the precursor of arsenosugars in the event of excess regulation of arsenosugarphospholipids in phosphate-rich environments (Raber et al. 2000). Cyclic arsenous dithiol esters in various species of *Sargassum* are soluble. In the event of leaching, arsenic forms will be mobile, in contrast to other cell wall–bound metals, as Ender et al. (2019) suggest for *Laminaria*. However, is such a soluble form adequate for storage?

As(V) adsorbed on Fe has consequences because iron plaques are naturally agglomerated in intercellular spaces (Fig. 1). They are non-specifically adsorbed and tightly complexed or scavenged on plaque surfaces in the presence of strong natural chelates that can sequester a number of nutrients and trace elements including arsenic (Rue and Bruland 1995; David et al. 1999; Khan et al. 2016). However, little is known about the accumulation potential of As species in terms of Fe plaque. Al Mamun et al. (2019), who studied this phenomenon, proposed that the P and Fe in the medium significantly inhibited the intracellular uptake of As(V), leading to heterogeneity in As accumulation inside/outside of tissues. Such adsorption could limit arsenic leaching but in an unknown range.

Sargassum spp. composition and compounds of interest for valorization

Sargassum composition

When looking at the potential use of various members of the genus *Sargassum* in various applications, understanding their biochemical composition is essential (Fig. 2). However, data relative to their composition are usually only partial. Whilst many authors have analysed specific components, there are relatively few complete studies on the global composition of *Sargassum* samples. Moreover, the broad diversity of the analytical methods used, especially for fibres and polysaccharides, or contents calculated by difference, may introduce biases and thus limit the possibilities for obtaining comparative information on various species. In addition, *Sargassum* spp. composition is highly dependent on various factors such as geographical origin of the seaweed, season or even the plant part analysed (Prince and Daly 1981; Bird et al. 1990). Consequently, intraspecific variation measured is high, and no interspecific variations can be assessed. Tables 5 and 6 summarize the best overview we could construct on the biochemical composition of pelagic and benthic *Sargassum* species.

Specific compounds of interest

Minerals

Seaweeds are known for their high mineral content, usually containing 20–40% of ash, and *Sargassum* spp. are no exception (Tables 5 and 6). Whilst the anionic fraction is usually composed mostly of chloride (50%) as well as sulphates and carbonates (Hou and Yan 1998), other compounds within the seaweed, including cell wall anionic and sulphated polysaccharides, are known to interact with cations strongly and selectively. These interactions combined with physiological and environmental factors lead to a great diversity of minerals and trace elements (Hou and Yan 1998; Circuncisão et al. 2018). Whilst this may lead to accumulation of undesirable minerals and heavy metals as exemplified above, it is also the source of many nutrients of interest for food and feed applications: sodium/potassium with an interesting Na/K ratio (Circuncisão et al. 2018), calcium, magnesium, sulphur, iron, iodine, copper, zinc or selenium.

Amongst those minerals, iodine can be present at particularly high levels, especially in brown seaweeds. Mairh et al. (1989) describe concentrations ranging from 500 to 1500 ppm iodine in *S. tenerrimum*, *S. johnstonii* and *S. filipendula*, whilst Hou and Yan (1998) measure values ranging from 100 ppm (*S. thunbergii*) to 6000 ppm (*S. vachellianum* and *S. henslowianum*). These latter values are amongst the highest ever measured values for brown seaweed. Whilst these high values may create concerns with recommended limits for humans (see “Regulatory constraints”) or feed applications (see “Regulatory constraints”), brown seaweed has been used for 2000 years in traditional Chinese medicine, especially for thyroid diseases and goitre (Liu et al. 2012) and may be relevant for specific populations such as pregnant women and young children (Andersson et al. 2012). This high iodine content may also play a role as an anti-bacterial agent and present benefits for health applications or to improve seaweed preservation. Lastly, this high mineral content, especially potassium, may be of agronomic interest, even if the sodium chloride content can limit direct uses in agricultural applications (see “Direct spreading”).

Carbohydrates

Whilst total carbohydrate content in seaweeds can be relatively high, carbohydrates contain mostly polysaccharides. The main carbohydrate families found in *Sargassum* spp. are alginates, fucose-containing sulphated polysaccharides (usually described as fucoidans), laminarin and mannitol, as well as cellulose, which, in most studies, is not quantified.

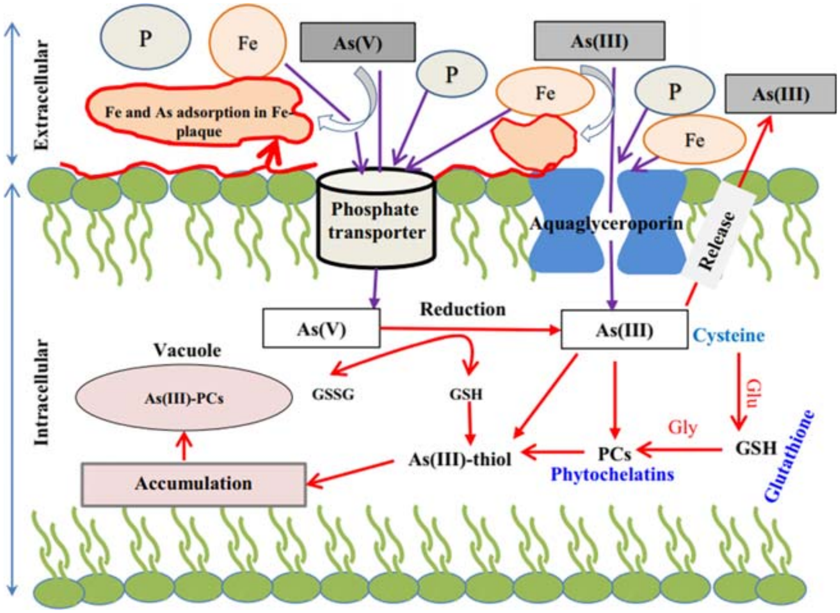

Fig. 1 Conceptual model for the extracellular and intracellular accumulation of arsenic from arsenic treatments modified with iron and phosphate (Al Mamun et al. 2019)

Except for some storage polysaccharides, the digestibility of seaweed polysaccharides by humans and animal species is generally low and they are usually considered as dietary fibres. Nevertheless, their specific structures and solubility characteristics can impair measurements with some traditional fibre methods developed for feed uses of land-based plants, which may not be relevant for seaweed, even if they are sometimes applied (e.g. neutral/acid detergent fibres). Moreover, total carbohydrates in food and feed applications are often calculated by difference (100 − (proteins + lipids + minerals + fibres)). With uncertainties regarding fibre determination, as well as additional categories which can represent a significant part of the seaweed (e.g. 5–15% polyphenols in some brown algae), this formula may also turn out to be very approximate. Knowing this, the contents reported for fibres, digestible carbohydrates and total carbohydrates should be considered with care, as illustrated by the variations in fibres and total carbohydrates compiled in Tables 5 and 6.

Alginates Alginates are the main constituents of the cell walls of brown seaweeds (Bird et al. 1990; Davis et al. 2003c). They usually represent 10–40% of algal dry weight, but their content depends not only on species, season and climate but also on local conditions (e.g. wave intensity). The alginate content in *S. natans* and *S. fluitans* is within 15–25% (Table 5), but values ranging from 3 to 40% have been reported (Bird et al. 1990; Zubia et al. 2008). The highest values should be taken with care as some authors express the alginate content after algae demineralization of the algal tissues (Davis et al. 2004); nevertheless, Bird et al. (1990) measured a content of 28% for *S. fluitans*.

Alginates, mostly extracted from kelp species (Laminariales) but also sometimes from Fucales (*Ascophyllum* and *Durvillaea* spp.), are processed at industrial scale for the production of texturants (Fig. 3). Beyond extraction yield, their molecular weight and chemical composition (balance and distribution of guluronic and mannuronic acids) are a key for their properties and market value. Alginates from *Sargassum* spp. are usually considered to be of poor quality and more difficult to extract than alginates from kelp species, and production yields are low compared to other species used industrially, for which alginate content is usually above 30–40%. Consequently, their processing remains very limited and does not stand out in global reports (Porse and Rudolph 2017). Nevertheless, only a limited number of studies on *Sargassum* spp. alginates are available, and some elements tend to demonstrate a high variability in composition depending on species, sites and harvesting period (Berteau and Mulloy 2003; Davis et al., 2003a, 2004; Torres et al. 2007; Bertagnolli et al. 2014; Mohammed et al. 2020).

Fucose-containing sulphated polysaccharides Fucose-containing sulphated polysaccharides (FCSPs), often referred to as fucoidans, are the second main constituents of brown seaweed cell walls (after alginates), which are further reinforced by cellulose microfibrils. They are characteristic of brown algae but present a very broad diversity of structures. Whilst *true* fucans or fucoidans, presenting a fucose backbone, can be found in Laminariales and some species of Fucales (*Fucus* spp., *Himanthalia elongata*), numerous FCSPs contain only a fraction of fucose and numerous other monosaccharides and uronic acids (glucose,

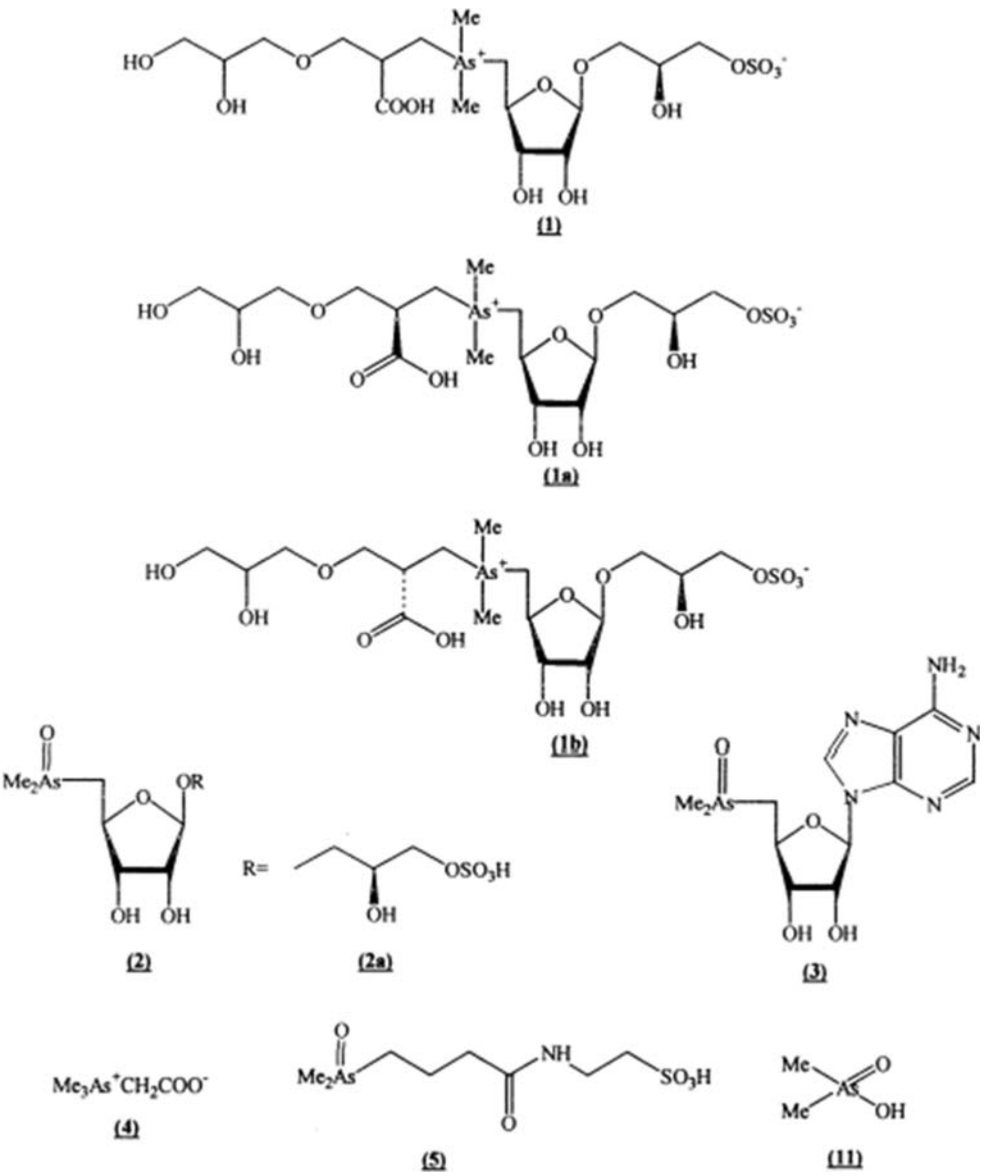

Fig. 2 From Edmonds (2000): compounds **1a** and **1b** from *Sargassum*, however, contain a substituent on arsenic that seems unrelated to any that could be provided by *S*-adenosylmethionine. This is also true of compound **5** isolated from *Tridacna* kidney (Francesconi et al. 1992) which has been identified by NMR spectroscopy and its structure confirmed by synthesis (Eichenberger 1993). So, the origin of these substituents can now be considered. Compounds **1a** and **1b** are similar to the polar portion of lipid **6**, a significant membrane component of several species of algae (Sato 1988), except that whilst **6** has the trimethylammonio group, **1a** and **1b** have an arsenic group that carries two methyl groups and a 5′-deoxyribose-5′-yl group as substituents. This latter group is related to the major dimethylarsinoylribose derivative **2a** of *Sargassum lacerifolium*. The similarities between compounds **1a** and **1b** of *S. lacerifolium*. The similarities between compounds **1a** and **1b** and **6** suggest that they might originate in parallel biosynthetic processes

galactose, mannose, xylose, glucuronic acid), with various patterns of branching and substitution (sulphates, acetates). *Sargassum* FCSPs fall in the latter case, and the most recent publications describe a backbone essentially composed of sulphated mannose, galactose and glucuronic acid, with sulphated fucose mostly present in side chains (Berteau and Mulloy 2003; Ale and Meyer 2013; Deniaud-Bouët et al. 2017).

These FCSPs are credited with a large part of the health benefits attributed to *Sargassum* spp. in traditional Chinese medicine (Liu et al. 2012). Extensive work has been performed to demonstrate their anti-viral, anti-cancer, anti-thrombotic or anti-inflammatory properties, as well as their potential uses in cosmetic applications (Berteau and Mulloy 2003; Holdt and Kraan 2011; Wijesekara et al. 2011; Liu et al. 2012; Deniaud-Bouët et al. 2017). It should also be noted that many publications are focused on the testing of FCSP extracts, but relatively few actually provide detailed information on their structure, or even their content in the seaweed, which limits perspectives for industrial uses and understanding of structure-function relationships. Analyses performed over recent years on various pelagic and benthic *Sargassum* spp. (Li et al. 2018a; CEVA, unpub.) show FCSP contents in the 3–12% range.

Laminarin Laminarin is a storage polysaccharide exclusively observed in brown seaweed species, including *Sargassum* spp. (Davis et al. 2003b; Holdt and Kraan 2011; Kadam et al. 2015). It is a branched (soluble) or unbranched (insoluble) glucose polysaccharide, mostly in the form of β-(1,3)-glucan, with some 6-O branching in the main chain and some β-(1,6)-intrachain links, as well as a potential terminal mannitol group (Kadam et al. 2015).

Table 5 Composition of pelagic *Sargassum* species

Reference	Species	Region	Minerals	Sulphur (%)	Alginates (%)	Laminarin (%)	Fucoidans (%)	Mannitol (%)	Total carbohydrates (%)	Fibres (%)	Lipids (%)	Polyphenols (%)	Proteins
Davis et al. (2004)	*S. fluitans*	Cuba			25								
Aponte de Otaola et al. (1983)	*S. fluitans*	Puerto Rico			18.8								
Aponte de Otaola et al. (1983)	*S. natans*	Puerto Rico			19.9								
Diaz-Vazquez et al. (2015)	*S. fluitans* + *S. natans* + *S. filipendula*	Puerto Rico	27.5%						10[1]		0.3		11.3%
Wang et al. (2008)	*S. natans*								64.0		1.4		9.6%
	S. natans/*S. fluitans*	Nigeria	8.7%						57.3	7.1[2]	2.5	0.1	15.4%
CEVA (unpublished)	*S. natans* + *S. fluitans*	French Caribbean	34.7%	1.2	13.4	< 0.5	~ 3	5.5				0.3	5.5%
Bird et al. (1990)	*S. fluitans*	Florida	32.9% (bladders)		13.4			3.4	7.9[1]	34.6			8.9%
			34.5% (blades)		22.7			3.7	7.0[1]	31.7			7.6%
			36.8% (stipe)		45.2			3.2	7.8[1]	32.4			6.4%
			39.6% (whole)		28.3			4.5	8.3[1]	36.5			8.6%
	S. natans	China	29%	1.2									22.2% (calc.)

[1] Soluble

[2] Crude fibre

Table 6 Composition of benthic *Sargassum* species

Reference	Species	Region	Minerals	Sulphur (%)	Alginates (%)	Laminarin	Fucoidans (%)	Mannitol	Total carbohydrates (%)	Fibres (%)	Lipids (%)	Polyphenols (%)	Proteins
Marinho-Soriano et al. (2006)	*S. vulgare*	Brazil	13.0–30.3%						52.6–68.5	4.8–10.5	1.5–7.9		9.2–19.9%
Torres et al. (2007)	*S. vulgare*	Brazil			17								
Marín et al. (2009); Robledo and Pelegrin (1997)	*Sargassum* spp.	Mexico	33.3%							6.4^{1} 47.1^{3}	0.5		6.3%
Robledo and Pelegrin (1997)	*S. filipendula*	Mexico	44.3%							6.6^{1}			8.7%
Carrillo et al. (2012)	*Sargassum* spp.	Mexico	38.4%							6.5^{1}	1.9		6.6%
	S. pteropleuron (autumn, morphology)	Florida	25–30%			3–5% (perennial) 3–4% (stem) 2–3% (leaves) 1–2% (bladders)		7% (perennial) 9% (stem) 15% (leaves) 26% (bladders)					11–12%
Prince and Daly (1981)	*S. pteropleuron* (whole plant, seasonal variation)	Florida	28–33%			1–5%		6–9%	55–65				10–12%
Bird et al. (1990)	*S. pteropleuron*	Florida	27% (bladders)		27.4			3.7%	8.6^{3}	18.7			6.0%
			28.9% (blade)		32.8			3.4%	6.5^{3}	33.0			6.2%
			21.8% (stipe)		21.0			3.1%	8.3^{3}	34.7			8.1%
			23.5% (whole)		24.5			3.5%	9.2^{3}	40.6			5.1%
Zubia et al. (2008)	*S. pacificum*	Tahiti			7.7–11.3			13–14%				2.1–4.5	
Lahaye (1991); Lahaye and Kaeffer (1997)	*S. fusiforme*	Japan								49–70			
Dawczynski et al. (2007)	*S. fusiforme*	Japan								62.3	1.4		10.9%
Chapman and Chapman (1980)	*Sargassum* spp.	Europe			17–35								
Herbreteau et al. (1997)	*S. muticum*	Europe									3.9		
Jard et al. (2013) and CEVA (unpub.)	*S. muticum*	Europe	36.6%	1.3	13.6	< 0.5%	~ 5	4.9%	16.6	51.2	1.9		8.4%
Jayasankar (1993)	*S. wightii* (seasonal variation)	India			12–26				$6–16^{4}$		0.2–1.6		3.1–7.2%
Kumar et al. (2015)	*S. wightii* (seasonal variation)	India	15–22%					1–2%	49–57		2–3	4.5–5.7	8.0–12.2%
Wong and Cheung (2001)	*S. hemiphyllum*	Hong Kong	21.3%						17.9	49.5	4.7		5%
	S. henslowianum		20.7%						2.3	60.7	4.3		11.9%
	S. patens		26.3%						4.3^{5}	55.5	5.7		8.2%
Borines et al. (2011)	*S. kushimotense*	Philippines	26.7%						42.9^{5}	11.5	0.1		6.4%
	S. ilicifolium		24.9%						46.8^{5}	16.1^{1}	0.2		5.1%
Borines et al. (2013)	*Sargassum* spp.	Philippines	26.2%						41.8^{5}	9.8^{1}	0.8		10.3%
Li et al. (2012a, b)	*S. patens*	China	17.8%	1.0									12.5% (calc.)
Li et al. (2018a, b)		China	27–38%		15–24		5–12				3–4.5	2–5	8–12%

Table 6 (continued)

Reference	Species	Region	Minerals	Sulphur (%)	Alginates (%)	Laminarin	Fucoidans (%)	Mannitol	Total carbohydrates (%)	Fibres (%)	Lipids (%)	Polyphenols (%)	Proteins
	S. fusiforme (seasonal variation)												
Di Filippo-Herrera et al. (2018)	*S. horridum* (seasonal variation)	Mexico	28–40%						44–58	6–8.5	0.1–0.4		5–8.5%

[1] Crude fibre

[2] Neutral detergent fibre (NDF)

[3] Soluble

[4] Alginates not included

[5] Fibres not included

Its content varies strongly, depending on season and on the seaweed reproduction cycle, but it can reach over 30% in members of the Laminariales in autumn. The laminarin content reported in various species of *Sargassum* appears to be lower, even if it has rarely been determined. Only one study provides a detailed analysis of the laminarin content in *S. pteropleuron* from Florida (Prince and Daly 1981), which varies between 2% in spring/early summer and 4% in autumn. The laminarin content is high in the perennial part of the seaweed (2.5–5%), whilst its content in the annual and reproductive portions remained much lower. Other authors have also demonstrated the presence of laminarin in *S. pallidum* (Ye et al. 2013) or *S. fusiforme* but neither accurately determined their content, even if Jin et al. (2017) showed that glucose proportion reaches its maximum in August. It should also be noted that these variations, related to seaweed morphology and to the reproduction cycle of benthic *Sargassum* species, should not necessarily be extrapolated to pelagic species, and that different patterns might be observed.

Laminarin is processed industrially for its capacity to stimulate the immune defences of plants and is also studied for its biological properties. It is also usually well metabolized by various organisms or can be hydrolyzed to glucose for further fermentation, making it an interesting starting material for energy applications such as biofuels or biogas.

Mannitol Mannitol is another storage carbohydrate. It is a polyol representing the first storage step in the photosynthetic chain, and which also plays an important role as an osmoprotectant in seaweed. Mannitol contents ranging from 3 to 14% have been described for *Sargassum* spp. (Zubia et al. 2008; Saha and Racine 2011) depending on season, geographies and species. As for laminarin, mannitol distribution throughout the seaweed thallus also exhibits strong variations depending on part and growth/reproduction cycle (Prince and Daly 1981).

Mannitol is sometimes co-extracted commercially by alginate producers (mostly in China) for food and pharmaceutical applications, when its content is sufficiently high to make it viable. However, it suffers from competition with the chemical route (hydrogenation) or biotechnological route from starch or sugar (Saha and Racine 2011; Varzakas et al. 2016). It is also one of the main constituents proposed as a bioactive by producers of plant biostimulants derived from brown seaweed (Sharma et al. 2014).

Proteins

The protein content of various seaweeds is usually calculated by determining nitrogen content using the Kjeldahl method (or sometimes Dumas) and applying the standard factor of 6.25 used for terrestrial plants. However, this factor might be slightly overestimated for those seaweeds which contain a

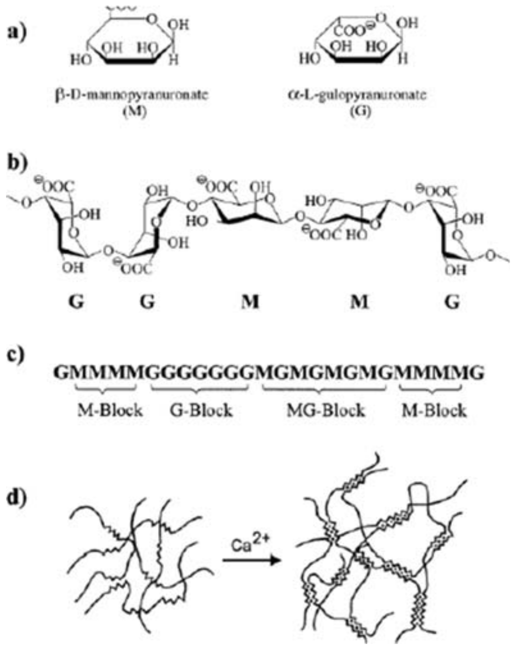

Fig. 3 Alginate molecular structure. **a** Alginate monomers (uronic acids: β-D-mannopyranuronate M vs. α-L-gulopyrunuronate G). **b** Macromolecular conformation of the alginate polymer. **c** Chain sequences; block co-polymer structure (Draget et al. 1996). **d** Calcium-induced gelation of alginate: schematic representation in accordance with the "egg-box" structure (Christensen et al. 1990)

relatively high content of acidic amino acids (Schiener et al. 2015) and for which non-proteic nitrogen represents 10–20% of nitrogen (Lourenco et al. 2002). Therefore, the nitrogen-to-protein conversion factor varies depending on species and season (Schiener et al. 2015; Angell et al. 2016) and a more realistic universal factor of 5 was recently proposed by Angell et al. (2016). Nevertheless, the traditional 6.25 factor is still often used. It might be slightly overestimated but facilitates comparisons with older studies. As for many brown seaweeds, the protein content of some *Sargassum* species is relatively low, usually ranging from 5 to 2% (Table 5), even if higher values have occasionally been reported.

Lipids

From a qualitative perspective, seaweed lipids are different from terrestrial plant lipids. Seaweeds possess elongase-desaturase complexes with the ability to synthesize long-chain unsaturated fatty acids (e.g. C20 and C22 ω-3 fatty acids). Another peculiarity of various seaweeds is a relatively low proportion of triglycerides (the most common lipids in plants) and significant fractions of phospholipids and glycolipids. Yet, the overall lipid content in seaweed is low, usually 1–3% of dry weight, even if some species such as *Ascophyllum nodosum* can contain up to 5% lipids (Dawczynski et al. 2007; Holdt and Kraan 2011).

In *Sargassum* spp., lipid contents are usually in the range of 0.5–5% (Table 5), even if higher values have been observed (Wong and Cheung 2001; Marinho-Soriano et al. 2006). It should also be noted that analytical methods differ between studies, which may impact results (e.g. methanol extractions of lipids which may co-extract polyphenols). Beyond fatty acid derivatives, the unsaponifiable fraction of *Sargassum* spp. tissues also contains sterols (Aknin et al. 1992; Guerra Dore et al. 2013), terpenoids (Balboa et al. 2013; Oyesiku and Egunyomi 2014), tocopherols and carotenoids such as fucoxanthin (Le Lann 2009; Heo et al. 2010; Yoon et al. 2012; Balboa et al. 2013).

Polyphenols

Brown seaweeds, and in particular Fucales (to which *Sargassum* species belong), are known for their high contents of polyphenols. These polyphenols are mostly composed of phlorotannins, oligomers and polymers of phloroglucinol.

Numerous studies have explored the anti-oxidant activity of various *Sargassum* extracts (Budhiyanti et al. 2012; Balboa et al. 2013, 2014) or even the structures of the polyphenols present in these extracts (Keusgen et al. 1997; Li et al. 2017b). However, even if the extraction processes are described, most authors focus on activity testing but do not determine the extraction yields, thus limiting the ability to determine the polyphenolic content in the seaweed. Polyphenol contents of up to 5% and more have been reported for *Sargassum* species (Connan et al. 2006; Balboa et al. 2013; Tanniou et al. 2013; Li et al. 2018a). Nevertheless, the content seems to be generally lower in pelagic species such as *S. natans* and *S. fluitans*, with contents usually below 1% (Table 6), which is consistent with their "yellow" colour, whilst seaweed rich in polyphenols are much darker.

Polyphenol extracts from brown seaweed are mostly used for cosmetic applications and food supplements. Whilst their relatively low content in pelagic *Sargassum* spp. might limit their potential extraction, it can also positively affect the extraction and purification of other seaweed components.

Current and potential applications for *Sargassum* spp.: technical and regulatory constraints

Food and food supplements

Sargassum consumption

Of the multiple *Sargassum* species present throughout the world, only one is consumed to a significant extent. As previously mentioned, *S. fusiforme*, also known as *Hizikia fusiforme*, hiziki or hijiki, is a traditional food from

Southeast Asia, exported throughout the world. Although it can be sold in its basic dry form, it is usually not directly consumed but rather used as raw material for further processing. *S. fusiforme* is traditionally boiled for several hours to remove astringent compounds (Nisizawa et al. 1987). This process also significantly impacts water-soluble compounds and helps reduce heavy metals and arsenic content (Hanaoka et al. 2001; Ichikawa et al. 2006).

Nevertheless, other *Sargassum* species, including *S. fluitans* and *S. natans*, are described as being consumed in the Caribbean as well as on the Pacific coast, e.g. *S. vulgare*, *S. fluitans*, *S. natans*, *S. platycarpum*, *S. cymosum*, *S. filipendula*, *S. liebmannii*, *S. polycystum* and *S. polyceratium* (Radulovich et al. 2013; Pereira 2016), but their peculiar taste and bitterness are mentioned. They are consumed fresh or rehydrated, fried, possibly after boiling or even as flour in tortillas. Other *Sargassum* species have also been considered as a potential food source, such as *S. wightii* in India (Kumar et al. 2015).

Regulatory constraints

There are no harmonized seaweed/algal regulations in Europe. The suitability of an algae for human food consumption is governed by the so-called “Novel Food” Regulation (Regulation (EU) 2015/2283), which applies to food and ingredients which were not consumed to a significant degree in Europe before May 15, 1997. Whilst *S. fusiforme* was consumed in Europe before 1997 and is listed as “not novel”, *S. fluitans* and *S. natans* are not listed and would require evaluation before being placed on the market in Europe (including some Caribbean territories where pelagic *Sargassum* species are present).

There are also no specific regulations in the USA for edible seaweed. Some species of brown (21CFR184.1120) and red (21CFR184.1121) algae are considered generally recognized as safe (GRAS) and are allowed as food additives. However, this covers only a limited amount of species, which have to be dried and ground or chopped and used as a flavour enhancer or flavour adjuvant. Some kelp species are also authorized as a source of iodine with maximum daily iodine intake (21CFR172.365). These additives have to fulfil the criteria for kelp as defined in the Food Chemical Codex. Some provisions can also be found in regulations relating to organic food (7CFR205.606). Nevertheless, uncertainty remains high as to which species can actually be consumed (Janasie 2018).

Whether or not the seaweed species is specifically allowed for human consumption, it also has to fulfil the relevant food product regulations, especially with respect to heavy metals and arsenic (Table 7). Maximum levels of contaminants are governed by general food laws, unless stated otherwise in specific regulations. Recommendations have also been published in France relative to the presence of heavy metals, arsenic and iodine, and serve as a reference across Europe in the absence of European regulations. Also, even if *S. fusiforme* consumption is authorized worldwide, a number of food safety agencies (UK, Ireland, Canada, Hong Kong, New Zealand, Australia), have issued warnings regarding the risks presented by its high arsenic content (Food Standards Australia New Zealand 2016).

Quality and supply

Heavy metals, arsenic and iodine in *Sargassum spp.*

Arsenic *Sargassum* species are known to present high contents of arsenic and inorganic arsenic, even in pristine waters, which can limit their use in food applications (Hanaoka et al. 2001; Ichikawa et al. 2006; Dawczynski et al. 2007; Grinham et al. 2014). *Sargassum fusiforme*, traditionally consumed in Asia, has been the most thoroughly studied. Ichikawa et al. (2006) describe contents of 40–80 ppm total arsenic in seaweed from China, Korea and Japan, whilst Almela et al. (2006) found contents ranging from 70 to 150 ppm total arsenic (40–120 ppm inorganic) in Japanese hijiki imported into Spain. Hanaoka et al. (2001) even describe total arsenic contents exceeding 200 ppm, mostly present in the most toxic inorganic form. Data relative to other species are more scarce but exhibit similar levels (Table 3). Arsenic levels in the order of 80–150 ppm total arsenic are also regularly observed in *Sargassum* spp. from the Caribbean (Michel 1985; Dawczynski et al. 2007; García-Sartal 2012), including a significant proportion of inorganic arsenic (40–70 ppm). This elevated arsenic content also led ANSES, France’s Food Safety Agency, to recommend exclusion of any food/feed uses of *Sargassum* spp. from the Caribbean (ANSES 2017).

Traditional processes of soaking and boiling applied to *S. fusiforme* present the advantage of significantly reducing their arsenic content. Nonetheless, Hanaoka et al. (2001) have shown that washing and soaking could reduce total arsenic by 30 to 60%. It was also shown that cooking can decrease total arsenic content by up to 90% depending on temperature and pH (Ichikawa et al. 2006; Katayama 2015). Nevertheless, arsenic concentrations in the seaweed after processing can still exceed regulatory levels.

Cadmium Whilst cadmium content in *Sargassum* spp. is of lower concern, when compared to arsenic, it still tends to exceed regulatory thresholds or recommendations (e.g. 0.5 ppm in France), with contents measured in the 0.5–2.0 ppm range (Almela et al. 2006; Dawczynski et al. 2007;

Table 7 Maximum levels of arsenic, heavy metals and iodine in algae authorized for human consumption (mg kg^{-1})

Compound	France Food	Europe Supplements	USA Additives	China
Inorganic arsenic (As$_i$)	3	–	3	1
Cadmium (Cd)	0.5	3		1
Mercury (Hg)	0.1	0.01		
Lead (Pb)	5	3	10	
Tin (Sn)	5			
Total heavy metals (as Pb)			40	
Iodine (I)	2000		1000–5000	
PAH-4 (polycyclic aromatic hydrocarbons)		0.05		
Chlordecone	0.01	0.01		
References	(CSHPF 1990; AFSSA 2009) Regulation (EC) 396-2005	Regulation (EC) 629/2008 Regulation (EU) 2015/1933 Regulation (EC) 396-2005	*Food Chemical Codex* 3rd Ed	(Cherry et al. 2019)

Holdt and Kraan 2011), and sometimes exceeding this level (Table 2).

Lead As illustrated in Table 2, the content in *Sargassum* spp. around the globe regularly exceeds 10 ppm, which is above the limits set in most food regulations.

Iodine Whilst the iodine content in *Sargassum* spp. can occasionally exceed regulatory thresholds with levels of up to 6000 ppm (Hou and Yan 1998), this is not uncommon for brown seaweed. It is also to be expected that industrial washing/soaking processes significantly decrease the iodine content, as already demonstrated for other species (Stévant et al. 2018), although this was not specifically considered in soaking/cooking studies related to arsenic.

Biomass availability and quality

Beyond the strictly regulatory aspects mentioned previously, edible seaweeds are generally cultivated or harvested from the wild as attached, benthic species. Both collection of beach-cast and possibly partially degraded *Sargassum* spp. and even harvesting of floating *Sargassum* spp. rafts raise numerous quality concerns: uncertain quality and food safety, lack of traceability, unknown history of growth and potential contaminations and so on.

Feed

Use of *Sargassum* spp. in animal feed

Various studies covered the inclusion of *Sargassum* spp. in animal nutrition, covering both animal performance and immunity aspects as well as product quality. Nevertheless, it should be noted that longer-term studies might be required to evaluate the potential impact of contaminants, and in particular arsenic, which was not usually quantified in the seaweed meals.

Poultry Carillo et al. (2012) described the use of 4–8% of *Sargassum* spp. in the diets for hens, which reduced the egg cholesterol content, whilst not affecting productive variables and the egg's physical quality. However, egg productivity was slightly reduced. Choi et al. (2014) also observed improved growth parameters and reduced mortality of broiler chickens with a diet enriched with 0.5% of *S. fusiforme* processing by-products (with or without fermentation).

Ruminants *Sargassum* spp. could be incorporated at up to 30% in the diets of sheep (Marín et al. 2009) and 25% for goats (Casas-Valdez et al. 2006) with no significant difference in feed intake, feed conversion ratio, growth performance and diet digestibility. Palatability remained good, but water consumption was increased, likely due to the elevated mineral content. More recently, 4% inclusion of *S. latifolium* was shown to improve stress resistance of sheep by regulating thermo-respiratory and inflammatory response (Ellamie et al. 2020).

Fish and shrimp Multiple studies evaluated the impacts of *Sargassum* spp. on growth performance and immunity of various aquatic species. For example, 6% of *S. fusiforme* in olive flounder diet (Pham et al. 2006; Kim et al. 2014) increased growth rate (although not significant vs. control) as well as disease resistance during challenge tests. Inclusion of 2–4% of *S. wightii* in the diet of Asian seabass fingerlings also brought major improvements in disease prevalence when challenged with *Aeromonas hydrophila* (Kodithuwakku et al. 2017). Up to 7.5% of

S. ilicifolium was also fed to rainbow trout without negative impact (Zamannejad et al. 2016). Ma et al. (2005) investigated high inclusion levels of *S. siliquastrum* (30%) in silver seabream production to enhance their marine taste by increasing bromophenol content. In shrimp aquaculture, low-level inclusion (0.5%) of *S. filipendula* improved resistance to thermal shock (Schleder et al. 2017), whilst 0.5% *S. horneri* upregulated immunity genes and growth performance (Lee et al. 2020). Cruz-Suárez et al. (2008) have also reviewed multiple studies with 2–10% inclusion of various *Sargassum* species and their impacts on growth performance, survival rates and immunity. The use of various *Sargassum* spp. extracts, rich in fucoidans, is also reported for anti-bacterial and anti-viral activity (Immanuel et al. 2012), which might be correlated with the capacity of FCSPs to reduce oxidative stress and modulate inflammation. Wang et al. (2018b) demonstrated that FCSPs extracted from *S. fusiforme* had a strong protective effect against H_2O_2-stimulated oxidative stress in vivo, in zebrafish. Other authors evaluated the effects of FCSPs from *S. horneri* and *S. fusiforme* in LPS-stimulated or virus-infected RAW264.7 mouse macrophage cells or zebrafish embryo models, demonstrating the downregulation of toxicity, modulation of NF-κB signalling cascade and reduction of pro-inflammatory cytokine production (Wen et al. 2016; Chen et al. 2018a, b; Sanjeewa et al. 2018).

Regulatory constraints

In Europe, Commission Regulation (EU) n° 68/2013 established the catalogue of feed materials and allowed for the inclusion of various algal forms (fresh, dried, algal meal, algae extracts, etc.) without specifying any restrictions on species which could be incorporated (Table 8).

In the USA, the Food, Drug, & Cosmetic Act requires that any substance that is added to or is expected to become a component of animal food, either directly or indirectly, must be used in accordance with a food additive regulation unless it is GRAS for that intended use, which brings us back to the same uncertainty on species authorized for food use. The Association of American Feed Control Officials (AAFCO) has specifically approved dried kelp from the families Laminariaceae and Fucaceae for use as ingredients in livestock feed, but not Sargassaceae (USDA National Organic Program 2013).

Quality and supply

Heavy metals, arsenic and iodine

Arsenic As for food applications, arsenic content appears to be a major limitation for the use of *Sargassum* spp. in feed applications. The arsenic content, and particularly the content of inorganic arsenic, largely exceeds the maximum values permitted in Europe, whilst other countries may not have specific regulations. Moreover, the processing costs related to the treatments for reducing arsenic content would be difficult to bear for feed ingredient manufacturers.

Natural and anthropogenic contents of cadmium may also exceed the maximum values, as well as lead content, whilst iodine content might also be a concern for formulators, limiting inclusion levels (but this is also the case for other brown seaweed species currently available and used in this sector).

Biomass availability and quality

As for food applications, collection of beach-cast *Sargassum* and even harvesting of floating *Sargassum* spp. rafts raise numerous quality concerns: uncertain quality and food safety, lack of traceability, unknown history of growth and potential contaminations and so on. Nevertheless, if quality and safety concerns could be resolved, the large volumes available at limited cost could make sense for feed applications as a novel source of seaweed for nutrition and health.

Cosmetics

Sargassum spp. have been the object of numerous studies related to their potential cosmetic applications and evaluating various types of extracts and fractions. Whilst some authors have investigated the activity of specific compounds, others have explored the bioactive properties of *Sargassum* spp. crude extracts, without establishing the link between chemical composition of the extracts and the effects observed. Some examples are outlined below.

Anti-oxidant/anti-ageing Numerous publications have described the in vitro anti-oxidant properties of *Sargassum* FCSPs, for example for *S. fusiforme* (Jaswir et al. 2014; Chen et al. 2016; Wang et al. 2018c), *S. ilicifolium* (Wang et al. 2015) or *S. filipendula* (Costa et al. 2011, 2015). It should be noted that this may not be directly related to the sulphated polysaccharides, but rather to co-extracted polyphenols, which are also relevant for cosmetic extracts. FCSPs from *S. fusiforme* have been studied for their protective effects against UVB-induced skin damage in vitro, in human dermal fibroblasts and in vivo with hairless mice (Ji et al. 2017; Wang et al. 2018c; Ye et al. 2018). Other lipidic compounds like carotenoids, sterols and meroterpenoids have also been tested. For example, fucoxanthin extracted from *S. siliquastrum* showed a strong in vitro anti-oxidant and protective effect against oxidative stress (Heo et al. 2008) and a similar protective effect against UVB radiation on human fibroblasts (Heo and Jeon 2009). Other studies have

Table 8 Maximum levels of arsenic, heavy metals and iodine authorized in algae for animal feed (mg kg^{-1})

Compound	France Raw materials	Europe	USA Additives
Total arsenic (As$_t$)	40	40	
Inorganic arsenic (As$_i$)	2*	2*	3
Cadmium (Cd)	1	1	
Mercury (Hg)	0.1	0.1	
Lead (Pb)	10	10	10
Total heavy metals (as Pb)			40
Iodine (I)		4–20**	
Dioxins and dioxin-like PCBs (ppt)	1.25	1.25	
PCBs	10		
Chlordecone		0.01	
References	30 October 2013 ruling	Directive 2002/32/EC Regulation (EC) 396-2005	

*Upon request of the competent authorities, the operator must perform an analysis to demonstrate that the content of inorganic arsenic is lower than 2 ppm. This analysis is of particular importance for the brown alga Hizikia fusiforme (sic)

**Added iodine in formulated feed depending on species: equines, 4 mg kg^{-1} (total); ruminants for milk production; laying hens, 5 mg kg^{-1} (total); fish, 20 mg kg^{-1} (total). Other species or categories of animals, 10 mg kg^{-1} (total)

investigated the effects of fucosterol on UV-induced cell damage (Hwang et al. 2014). Plastoquinones and chromene from methanolic extracts of *S. micracanthum* have been shown to possess significant anti-oxidant activities (Iwashima et al. 2005). The inhibitory effect of sargachromanol E extracted from *S. horneri* was investigated on UVA-irradiated dermal fibroblasts. The results indicated that sargachromanol E oxidative stress and expression of collagenases showed interesting anti-ageing properties (Kim et al. 2012, 2013a).

Whitening In a study aiming to find new whitening agents, 43 marine algae were screened for their tyrosinase inhibitory activity. Four seaweed extracts, including *S. siliquastrum*, evidenced potent tyrosinase inhibitory activity similar to that of positive control (kojic acid). *Sargassum siliquastrum* extracts also exhibited excellent inhibitory effects on the pigmentation of zebrafish (Abdel-Fattah et al. 1974; Cha et al. 2011). Hypopigmenting properties of the ethanolic extract from *S. serratifolium* were found to be correlated to the presence of sargaquinoic acid as an active component (Azam et al. 2018). FCSPs are of interest for skin-whitening applications, and a study conducted with depolymerized fucoidans from *S. fusiforme* revealed that low-weight polysaccharide had higher anti-tyrosinase activity than the original polysaccharide (Chen et al. 2016).

Anti-pollution New kinds of tests are being developed for this recent trend in cosmetic products. Ethanolic extract of *S. horneri* was reported to increase in vitro the mouse macrophage cellular resistance to fine–dust-induced inflammation and oxidative stress, showing anti-pollution potential (Jayawardena et al. 2018). Alginic acid extracts from *S. horneri* were also shown to significantly reduce inflammatory response in keratinocytes exposed to urban dust aerosols (Fernando et al. 2018). This interest in *Sargassum* spp. usage in the cosmetic field can also be illustrated by the commercialization of various extracts (Table 9).

Regulatory constraints

Heavy metals Concerning heavy metals, there are no clear international regulations on maximum levels allowable as impurities. For example, in Europe, cosmetic products must not contain any of the substances listed in Annex II to Regulation (EC) No. 1223/2009, which includes heavy metals such as lead, cadmium, arsenic, antimony and mercury. However, traces of these compounds are often present in certain cosmetics and in make-up products, due, amongst other reasons, to the inclusion of mineral pigments. According to most national/international regulations, traces of heavy metals in cosmetic products are allowed if they are technically unavoidable, but explicit limits are not always listed.

Nevertheless, some countries have issued regulations or recommendations on this point, and a number of them have implemented maximum levels that should not be exceeded in final formulated cosmetic products based on "technically avoidable" concentrations. Table 10 summarizes the limit concentrations proposed by several national authorities.

Some countries are also beginning to tighten their limits, as illustrated by the recently revised limits in Germany, where Federal Office of Consumer Protection and Food Safety

Table 9 Commercial and cosmetic ingredients based on *Sargassum* spp.

Ingredient name	Supplier	Description/claims
Fuligo	Clariant	*Sargassum fusiforme* extract; firming, moisturizing, anti-ageing and anti-wrinkle agent
AT MaronWhite S	Athena	*Hizikia fusiforme* extract; skin moisturization, skin lightening
AT MaronUV S	Athena	*Sargassum fulvellum* extract; protective activities against UVB
Gulfweed extract	Dermalab	*Sargassum fulvellum* extract; anti-oxidant, anti-inflammatory agent
LOOKETHIC	Biocosmethic	*Sargassum filipendula* extract; anti-ageing, anti-wrinkle, anti-oxidant, anti-fatigue, protecting and soothing agent
PHYACTYL®	Gelyma	*Sargassum muticum* extract; protective agent, lightening/whitening
Pearl Alga	Odycea	*Sargassum muticum* extract; whitening agent

(BVL) considered that good manufacturing practices would allow lower technically avoidable levels (BVL 2017). If levels of these contaminants exceed those values, the safety report of the cosmetic product has to consider it and provide the evidence that they are safe for the consumer.

Whilst these maximum limits or recommendations do not apply to the raw seaweed material, it is clearly necessary to monitor the content of arsenic and heavy metals in any extracts produced from *Sargassum* spp. As cosmetic active ingredients are usually used at low levels (typically below 1–2% for dry extracts), this reduces the risk of exceeding the thresholds, but the lower limits for arsenic or cadmium in Germany, which might also be applied in other countries, could be a risk. Some companies' manufacturing end products might also be requesting that the ingredients/actives themselves respect those limits.

Organic contaminants It should also be noted that all PAHs with carcinogenic properties are forbidden in cosmetic products in Europe, as well as chlordecone (Regulation (EC) No. 1223/2009—Article 15 and Annex 2).

IECIC list To be sold on the Chinese market, one of the largest markets for cosmetics worldwide and exhibiting significant growth, cosmetic products should only contain ingredients found on the Inventory of Existing Cosmetic Ingredients in China, also known as the IECIC list (CFDA 2015). To date, only four *Sargassum* spp. extracts were listed on the IECIC list, i.e. *S. filipendula*, *S. fusiforme*, *S. muticum* and *S. pallidum*. The absence of *S. natans* and *S. fluitans* from this list means that most international manufacturers of cosmetic ingredients would avoid investing in those species, and that developments from these seaweeds might be limited to regional products. Whilst the registration of new ingredients is still an option, this is a very lengthy and uncertain process that few companies are willing to undergo.

Quality and supply

Beyond the regulatory aspects mentioned previously, quality, traceability and sustainability of the sourcing are key factors for cosmetic applications, as they are often part of the marketing of the product. Despite the significant interest of *Sargassum* spp. extracts for a wide range of cosmetic benefits, using *Sargassum* spp. blooms as raw material might be difficult to reconcile with these important determinants for cosmetic manufacturers. This is particularly true as a large number of benthic *Sargassum* species are available globally, with harvestable stocks largely sufficient for the small volumes required for cosmetic extracts, and some of these species

Table 10 Maximum acceptable levels of arsenic and heavy metals in final formulated cosmetic products (μg g^{-1})

Compound	Germany (1985)	Germany (2007)	Canada	Southeast Asia
Lead (Pb)	20	2*	10	20
Arsenic (As)	5	0.5*	3	5
Cadmium (Cd)	5	0.1	3	5
Mercury (Hg)	1	0.1	1**	1
Antimony (Sb)	10	0.5	5	
References	(BGA 1985)	(BVL 2017)	(Health Canada 2012)	(ASEAN 2017)

*Levels of 5 ppm and 2.5 ppm, respectively, for some make-up products

**The impurity limit for mercury has been amended from 3 to 1 ppm as per the Minamata Convention on Mercury signed by Canada on October 10, 2013 are already listed in China. *Sargassum* spp. blooms might also be perceived negatively by end-users, worried by press and media coverage of pollution, heavy metals and toxic hydrogen sulphide emissions. Lastly, whilst the development of cosmetic ingredients from *Sargassum* spp. blooms might bring economic returns to the companies involved, the small volumes required would not have any significant impact on the volumes to be handled.

Health applications

As for cosmetic applications, *Sargassum* spp. have been the object of numerous studies related to their health properties and bioactive compounds. However, as stated by Liu et al. (2012) in the conclusion of their review, many studies lack proper phytochemical characterization of the extracts used.

Namvar et al. (2013) investigated the anti-proliferative and anti-angiogenesis properties of *S. muticum* polyphenols, whilst ethanolic extracts of *S. serratifolium* were also recently shown to have potential against certain osteoclastic bone diseases and osteoarthritis (Kim et al. 2018; Park et al. 2018). Other research teams focused on lipidic compounds and investigated fucoxanthin (Heo et al. 2010) or extracts of sterols (Buwono et al. 2018) for anti-inflammatory and analgesic properties, or hepato-protective properties of meroterpenoids from *S. serratifolium* (Lim et al. 2018).

However, many studies report the potential of FCSPs from *Sargassum* spp. with regard to their anti-oxidant and anti-microbial (Jaswir et al. 2014), anti-tumour (Costa et al. 2011; Yu et al. 2017; Fan et al. 2018b), immune-modulatory (Jeong et al. 2015; Chen et al. 2018a), anti-inflammatory (Guerra Dore et al. 2013; Chen et al. 2018b), anti-coagulant and anti-thrombotic (Athukorala et al. 2007; Guerra Dore et al. 2013), hepato-protective (Yu et al. 2017), neuro-protective (Huang et al. 2017; Jin et al. 2018) or anti-viral (Preeprame et al. 2001; Thuy et al. 2015) activities.

Other *Sargassum* spp. polysaccharides have also been described for their prebiotic properties, e.g. laminarin or complex polysaccharides (Chamidah 2018; Fu et al. 2018).

Regulatory constraints

Holdt and Kraan (2011) wrote an extensive review in 2011 covering seaweed bioactives and the regulatory requirements around the development of new functional ingredients. Even if the overall regulatory framework remains relatively unchanged, some regulations have evolved, for example the new Novel Food Regulation in Europe and associated changes in the procedures for authorization of novel foods.

An additional difficulty, at least for Europe, would be that *S. fluitans* and *S. natans* are not yet considered as edible species, which may imply more extensive testing to demonstrate the safety of the ingredients. Moreover, heavy metal and arsenic levels in the final products will also be under scrutiny.

We will not cover the regulatory requirements for drugs, which would not be different when prepared from *Sargassum* versus other sources, but again contaminants might be a concern.

Quality and supply

The development of health products requires the highest quality standards (Holdt and Kraan 2011; Gellenbeck 2012; Wells et al. 2017), which can be an issue when starting from raw materials harvested from blooms containing potentially unknown contaminants. This point could possibly be solved by processing steps and extensive purifications if required.

Raw material variability might be even more challenging (Gellenbeck 2012; Wells et al. 2017), particularly for complex compounds like polyphenols or polysaccharides, and in particular for FCSPs. A very high reproducibility of composition and bioactivity is expected from health products, which might be easier to obtain from cultivated seaweed (Gellenbeck 2012), or at least controlled harvesting. The composition of complex molecules in seaweed found in drifting blooms is likely to present higher variability, due to various environmental factors such as geographical origin, nutrients and weather conditions. Specific metabolites like fucoxanthin would remain easier targets as their content may vary but not their composition.

Agriculture

Direct spreading

Beach-cast seaweed (fresh or partially dried) has been used historically in rural coastal areas, in order to amend the soil in fields close enough to the coast (uncharacterized blend of seaweed collected on the beach, especially in autumn after the first storms in Europe). This is the most direct and cheapest use and is still performed locally across the globe. When fresh *Sargassum* spp. are used, safety measures similar to those implemented for *Ulva* spp. should be taken. To prevent anaerobic fermentation and formation of hydrogen sulphide, seaweed should be spread within 24–48 h from collection and ploughed rapidly (ANSES 2011).

Direct spreading of seaweed requires the setting up of a nutrient management plan, which may take different regulatory forms depending on national regulations. It usually describes the farm(s), the zones where seaweeds will be spread and the conditions under which spreading will be performed. It should take into account the nitrogen, phosphorus and potassium contribution from the seaweed, and the plan is usually complemented with an agronomic follow-up to assess efficiency and required adjustments.

There is still a limited amount of scientific data relative to direct use of *S. natans* and *S. fluitans* in this context, but several questions have been raised with regard to their content of nutrients and contaminants. A study performed for DAAF Guadeloupe in 2011 (personal communications) showed adequate nitrogen and phosphorus inputs in beach-cast *Sargassum* spp. (expected to contain *S. natans* and/or *S. fluitans*) but flagged sodium content as a potential contributor to soil salinization. The report recommended an input of 20 t ha^{-1} for sugar cane, with spreading during the rainy season and 2 months before cultivation in order to allow sodium leaching. The same study also reviewed heavy metals, but lead, cadmium and mercury were not a concern from a regulatory perspective. A more recent study performed by IT2 (Tropical Technical Institute) and several partners in the French Caribbean also concluded that the addition of 10 to 20 t ha^{-1} of beach-cast *Sargassum* species (*S. fluitans* and/or *S. natans*) had no agronomic interest for cucumber, lettuce, banana, sweet potato and sugarcane, whilst presenting a risk of salinization and raising concerns with arsenic and, potentially, chlordecone. Consequently, a recommendation was issued to farmers to avoid using *Sargassum* (Sargasses and Agriculture 2018). Indeed, with total arsenic contents reaching 100–200 mg kg^{-1}, of which a large part was inorganic, direct spreading of *Sargassum* spp. cannot be performed under the French regulation for organic amendments (limit of 18 mg kg^{-1} total As) or even the European Regulation (limit of 40 mg kg^{-1} of inorganic arsenic) (see Table 8). In addition, even if this limit is not taken into account, the French annual limit of arsenic of 90 g ha^{-1} year^{-1} for organic amendments would limit the use to less than 1 ton ha^{-1} of seaweed, which presents little agronomic interest.

Composting

Composting is an aerobic method used to decompose fermentable organic materials, eliminating or reducing pathogens, reducing the waste material mass by evaporation and CO_2 emission and transforming organic matter into humus. The compost obtained can be used as organic amendment/soil conditioner. Composting is usually performed on dedicated sites (possibly classified facilities for environment protection), where liquid phases (and sometimes gases) can be collected and treated. It generally consists of two phases: an active phase of aerobic fermentation (4–6 weeks), where the biodegradation is at its most active, with significant leaching of fermentation juices and atmospheric release (water vapour, CO_2, ammonia, aerosols, etc.) and a maturation phase (4–6 months) with reduced aeration, where organic matter reaches a humic state and the compost stabilizes.

Composting of pure seaweed is known to be difficult (inadequate C/N ratio (too low), high mineral content and salinity, etc.) and generally requires the addition of other organic wastes (typically green waste, wood or straw, acting as structuring material and providing organic matter). The proportion of plant waste should be adapted according to the seaweed species to provide adequate organic content and porosity for proper aeration (Han et al. 2014; Cole et al. 2016).

A number of studies have been conducted specifically with *Sargassum* spp. A study was carried out in 1984 with *S. muticum* (Brault et al. 1984) and various types of green waste (75–85% seaweed in the mix). The technical feasibility of using *S. muticum* as the main constituent for compost production conforming to the French NFU 44-051 standard for organic amendments was demonstrated, despite a lack of economic value if the seaweed had to be harvested. Various technical parameters relative to seaweed preparation were also tested. Pressing or partial drying of the seaweed to 20–25% dry matter was a pre-requisite to reach sufficient temperature during the fermentation phase. Washing to reduce the sodium content and conductivity was also efficient, but detrimental to other compounds of interest for fertilization (e.g. potassium). Co-composting of *S. natans* and *S. fluitans* with food waste and wood chips was also investigated at the University of Texas (Sembera 2013), but limited amounts (2%) of seaweed were used and the impact was limited. In a more recent study, the same authors increased incorporation to 4% (which remains relatively low), obtaining good quality compost (Sembera et al. 2018). Washing of the seaweed had no significant impact on salinity, which remained in the lower range accepted by the US Composting Council.

More recently, massive strandings in the Caribbean incited a number of compost facilities to incorporate *Sargassum* spp. in their products. A case in point can be found in Martinique, where three companies (Holdex, Idex and Société Martiniquaise des Eaux) obtained the authorization to incorporate the collected seaweed in their composts. Sand and excess sodium had to be considered, but co-composting with green waste or sugarcane bagasse with up to 10% fresh *Sargassum* spp. turned out to be feasible, whilst maintaining products complying with standards NFU 44-051 and 44-551. This inclusion level may seem relatively small, in order to cope with massive strandings, but it also allows relatively easy integration into existing composting units, which have to run on alternative feedstocks during periods where no strandings occur. New trials performed under strongly hypoxic conditions were also performed by Holdex and exhibited interesting composting efficiency (personal communications). The results will be published in the near future.

Whilst arsenic might be a concern for the direct spreading of *Sargassum* spp., co-composting with a moderate content of *Sargassum* spp. (e.g. $\leq 10\%$) “mathematically” brings it down to levels below or close to the limit set in France, for example

18 mg kg^{-1} dw, and in typical ranges for soils. In addition, a significant fraction of the arsenic is likely to leach out during the composting phase, as shown for other arsenic-rich plants (Cao et al. 2010). Some arsenic may also volatilize as arsine gas (AsH_3) or its volatile methylated forms after conversion by microorganisms, although volatilization in aerobic conditions is expected to remain low (Naidu et al. 2006; Cao et al. 2010; Cullen and Reimer 2016). However, the presence of arsenic in compost leachates should be monitored, and specific remediation treatments might be required. Volatilization, whilst increasingly considered as an arsenic mitigation strategy, will also subsequently lead to oxidation of the released arsine derivatives into non-volatile forms returning to the soil through particle deposition and rain (Punshon et al. 2017).

Formulated extracts (liquid fertilizers, biostimulants, etc.)

Brown seaweed extracts, sold as seaweed concentrates or formulated biostimulants, have a significant history of use for plant growth and health (Craigie 2011; Sangha et al. 2014; Sharma et al. 2014; Battacharyya et al. 2015). Seaweed polysaccharides, including laminarin and FCSPs, can also act as elicitors, triggering plant immune response by stimulating defence enzymes (Zheng et al. 2020). Beyond compounds of interest for biostimulation (phytohormones, polysaccharides, etc.), the species used were also strongly influenced by their availability either as wild harvestable resources or as beach-cast seaweed (Zemke-White et al. 2004; Stirk and van Staden 2006). The most commonly used species in commercial products are Fucales and Laminariales. In the Northern Hemisphere, *Ascophyllum nodosum* is the most frequent (Northwest Europe, North America) and, to a lesser extent, *Laminaria* spp., whereas in the Southern Hemisphere, products are usually based on *Ecklonia* spp. (New Zealand, South Africa) and *Durvillaea potatorum* (Australia).

Sargassum spp. are far less common, although the Chinese company Beijing Leili Marine Bioindustry proposes a range of *Sargassum*-based products, and local products are available in the Caribbean (e.g. Santa Lucia–based Algas Organics) or in Indonesia, for example. They are also less covered in the scientific literature, even if a number of recent studies describe interesting results of *S. wightii* extracts on cluster bean (Vijayanand et al. 2014) or okra (Tensingh Baliah et al. 2017). An optimal concentration may also be necessary, as both studies show that beyond certain levels, the addition of these extracts might be detrimental. *Sargassum vulgare* extract was also shown to improve yield and quality of red radish (Mahmoud et al. 2019), but germination of mung bean was delayed by *S. horridum* extract (Di Filippo-Herrera et al. 2018) and *S. muticum* extracts did not stimulate mung bean and pak choi (Sharma et al. 2012). Another recent study using several *Sargassum* species as solid and liquid extracts for rice growth did not significantly improve harvest yields, even if impacts on vegetative growth were observed (Sunarpi et al. 2019).

Regulation for organic fertilizers, compost and biostimulants

Different limits may apply to *Sargassum*-based agricultural products depending on countries and product types (Table 11). For example, compost in the USA falls within the regulation of biosolids (40 CFR 503) with the same limits as for sewage sludge, whilst in the case of France, specific limits apply in the standards for organic fertilizers and composts.

Fertilizing products and biostimulants placed on the market in Europe must comply with the newly introduced regulation (EU) 2019/1009 (for CE-marked products) or with national regulations/standards (e.g. by complying with specific standards or through specific authorization procedures). In addition, all substances incorporated into the EU fertilizing product, e.g. *Sargassum*-based extracts, need to have been registered pursuant to Regulation (EC) No. 1907/2006 concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) and may require additional testing depending on production volumes.

Bio-based materials

There are a number of uses of *Sargassum* species as raw materials for bio-based materials, although these are not extensively covered in the scientific literature. They are usually driven by individual initiatives and the creation of dedicated companies. To cite just a few, *Sargassum* sp. from the Caribbean region is currently being processed or explored for its use in bio-based or partially bio-based plastics (AlgoPack, Renovare), in papers and boards (Sargánico, Sargasse Project, The Marine Box) or for the production of *Sargassum* bricks in Quintana Roo, Mexico (Sargablock).

Regulatory constraints are too dependent on product types and sectors of applications to be covered here, but migration of contaminants, for example, should be closely monitored for packaging applications or any products coming into contact with food, as well as final disposal for products that would be produced in large volumes.

Energy

Biogas production

Anaerobic digestion presents the advantage of being able to ferment complex organic compounds and can be applied to a broad range of animal and vegetal biomass. Whilst converting potential organic waste, it can be seen as an environment-friendly means of access to renewable energy or heat via the production of biogas (methane and CO_2).

Table 11 Maximum levels (mg kg^{-1} dry weight) and maximum flux (g ha^{-1}) of arsenic, heavy metals and contaminants in fertilizers, organic amendments, composts and plant biostimulants

Compound	France			Europe	USA Compost
	Max level	Max annual flux	Max 10-year flux	Max level	Max level
Total arsenic (As_t)	18	270	900		41
Inorganic arsenic (As_i)				40	
Cadmium (Cd)	3	45	150	1.5	39
Chromium (Cr)	120	1800	600	2 (Cr(VI))	–
Mercury (Hg)	2	30	100	1	17
Lead (Pb)	180	2700	9000	120	300
Nickel (Ni)	60	900	3000	50	420
Total heavy metals (as Pb)					
Fluoranthene	4	6			
Benzo(b)fluoranthene	2.5	4			
Benzo(a)pyrene	1.5	2			
References	Standards NF U 42-001, NF U 44-051 and NF U 44-095			Regulation (EU) 2019/1009	(US Composting Council) Based on 40 CFR 503

In brown seaweeds, polysaccharides and carbohydrates (alginates, fucoidans, laminarin, mannitol) are the main substrate for this fermentation process. But as anaerobic digestion is relatively unspecific, other seaweed constituents such as proteins and lipids can also be degraded. Chynoweth and coworkers (2001) showed that seaweed can be relatively easily converted to biogas. Promising results were also reported for various seaweed species and genera: *S. muticum* (Gellenbeck and Chapman 1986), *Laminaria hyperborea* (Horn et al. 2001), *Ulva* sp. (Morand and Briand 1999; Morand et al. 2006) and *Macrocystis pyrifera* (Vergara Fernandez et al. 2008). Average conversion yield reported in various studies varies between 200 and 300 Nm^3 biogas per tonne of volatile matter (Briand and Morand 1997; Chynoweth 2002; Vergara Fernandez et al. 2008). In comparison with other biomasses commonly used for biogas production, this value is close to that found for manure, pig slurry or lignocellulosic residues.

However, amongst the algal genera studied, *Sargassum* spp. are not the best candidates. Jard et al. (2013) showed that *S. muticum* had the lowest methane yield amongst ten species from the Brittany coast, with 0.13 L CH_4 g^{-1} of organic matter, and similar values were found by Milledge and Harvey (2016). Other studies reviewed the anaerobic digestion of *S. muticum* in Europe (Oliveira et al. 2015; Soto et al. 2015; Milledge and Harvey 2016), but also *S. fluitans* and *S. pteropleuron* in Florida (Habig and Ryther 1983; Bird et al. 1990). This low methane yield might be explained by a relatively large proportion of insoluble fibres (recalcitrant to anaerobic digestion) and polyphenols (inhibitors), with low contents in digestible carbohydrates and proteins, and an inadequate C:N ratio. Although most authors do not report particular problems with the composition of the biogas and product forms, these limited yields remain a major concern for any developments, especially as the high mineral content results in progressive filling of the digester (Jard et al. 2013; Soto et al. 2015). The use of co-substrates such as glycerol might improve conversion (Oliveira et al. 2015) but has not, to our knowledge, been significantly applied to seaweed biomass.

Other components of *Sargassum* spp. might negatively impact biogas production. For example, H_2S formation during anaerobic digestion is known to induce inhibitions, even if these inhibitions are often not observed with seaweed despite high H_2S concentrations (Montingelli et al. 2015), and yield losses are observed due to competition with sulphate-reducing bacteria (Cecchi et al. 1996). The presence of H_2S in the biogas can also trigger corrosion in co-generators, and specific treatments might be required (Peu et al. 2011; McKennedy and Sherlock 2015). Inhibition phenomena related to polyphenols have also been described with brown seaweed (Jard et al. 2013; McKennedy and Sherlock 2015; Soto et al. 2015), although the contents are relatively low in *S. natans* and *S. fluitans*. In a recent study, Milledge et al. (2019) confirmed that alginates were highly recalcitrant, and that the addition of phenolics further decreased conversion ratio, confirming their inhibitory properties. Minerals, especially sodium, might also induce inhibitions, although those usually occur at relatively high levels (Soto et al. 2015). Lastly, the presence of sand and foreign bodies can damage grinding equipment, pumps and digesters. In addition, as digesters require a stable feed to reach a stable methane production regime, irregular supply from strandings is far from ideal. A recent study also explored the possibility to fractionate *S. muticum* to obtain products of interest (biostimulants, FCSPs and polyphenols), with final solid residues which were shown to be more adequate for biogas production than the raw seaweed (Flórez-Fernández et al. 2020).

Taking into account the above-mentioned limitations, *Sargassum* spp. still appear to be a potential co-substrate in biogas production, but we are not aware of any industrial sites currently processing seaweed for biogas.

The fate of arsenic during biogas production still raises a number of questions. Various authors studied the anaerobic digestion of arsenic-rich sludge from wastewater treatment (Mohapatra et al. 2008), arsenic-rich plants (da Silva et al. 2019) or arsenic-containing sewage sludge (Weithmann et al. 2019). All authors concluded that there was a limited impact of the arsenic on methanogenic bacteria and digestion performance. Whilst some authors mention potential significant volatilization of arsenic (Mohapatra et al. 2008; Mestrot et al. 2013), most recent studies tend to show that arsenic volatilization remains very limited, often around or below 1% of total arsenic (Mestrot et al. 2013; Clancy 2015; da Silva et al. 2019; Weithmann et al. 2019) and arsenic concentration in the biogas was largely below regulatory limits. Nevertheless, technical limits might be reached with very low levels of arsenic in the gas phase and risks of machinery failure and catalyst poisoning are reported (Weithmann et al. 2019). Also, as most of the arsenic is transferred to the aqueous phase, the digestate might require specific post-treatments, and it could also prevent it being used for fertilization.

Hydrogen production

Hydrogen production is another potential use of vegetal biomass, including algae. The mechanisms involved in this process are well described in a recent review by Xia et al. (2015) even if it targets microalgae more specifically. In the context of hydrogen production, the main limiting factors appear to be inhibition by ammonium, related to the presence of proteins in the algae biomass, but which should remain limited considering the low protein content in *Sargassum* spp. as well as concerns with the purity of the gas produced (especially related to the presence of H_2S, sulphur compounds and ammonia), which need to be cleaned up before use.

Bio-oil and biochar production (hydrothermal liquefaction, pyrolysis)

Hydrothermal liquefaction (HTL) and pyrolysis are two conversion techniques for the production of bio-oils, allowing vegetal biomass to be converted into energetic substrate analogous to unrefined crude oil. Both techniques have been explored in recent years on seaweed biomass and, in particular, on *Sargassum* spp. (Li et al. 2010, 2012a, b; Kim et al. 2013b; Diaz-Vazquez et al. 2015; Ghadiryanfar et al. 2016; Biswas et al. 2018, 2020; Li et al. 2020). Their principles are different, but both techniques lead to relatively similar bio-oils (Saber et al. 2016). Pyrolysis is a thermal treatment at atmospheric pressure in the presence of oxygen. It should be performed on dry seaweed, but the pyrolysis itself can provide energy for the drying stage. HTL is a thermal treatment in aqueous conditions under pressure, which presents the advantage of not requiring preliminary seaweed drying. In this review, we will not cover the advantages and drawbacks of the two techniques on yield and quality of the bio-oil. Both require significant post-processing to cope, amongst others, with oxygen and nitrogen-containing compounds (Wang et al. 2013; Saber et al. 2016). During this refining step, usually via catalytic hydrotreatment, sulphur compounds are converted to H_2S (Biddy et al. 2013). Nevertheless, as these are purely thermo-chemical processes, and not biological ones, inhibition by ammonia and toxicity of sulphur compounds are not a concern as in biogas production, but they still need to be removed from the final products and may present similar risks for equipment (corrosion, deposition and so on) and catalysts. Transfer of contaminants to the gas phase during pyrolysis should also be considered, as it must in the case of combustion (see “Waste”).

Depending on process parameters, various levels of biochars can be produced during these processes. They can be of interest for various applications, such as the production of porous, activated carbon (Xu et al. 2013; Biswas et al. 2018; Li et al. 2018b), which could be further used for electronics, agriculture or bioremediation applications. But, their level of impurities will have significant impact on potential uses. Although the possibility has not been significantly studied yet, these techniques could also be considered as a potential tool to concentrate some of the contaminants present in *Sargassum* spp., such as arsenic and heavy metals, in a concentrated aqueous phase or in dry ashes.

Fate of arsenic and heavy metals in the processing of *Sargassum* spp. biomass: regulatory constraints

Fate of arsenic and heavy metals

As discussed in the previous section, numerous applications are, or can be, considered for *Sargassum* spp. Except for a limited number of direct uses, any industrial processes applied to those seaweeds will generate wastes, co-products, water effluents and possibly gaseous effluents. Moreover, even direct applications may require temporary storage or cleaning/drying steps which will themselves generate similar products/effluents. Determining the fate of contaminants such as arsenic and heavy metals over those processes should be performed on a case-by-case basis. However, a number of regulatory aspects should be taken into account whilst assessing their industrial feasibility.

Effluents and waste regulations

Waste

Spent biomass from *Sargassum* spp. extraction can be considered as plant waste, seaweed or compostable material (depending on national regulations and on specific processes/chemicals that might have been used) and, as such, be used for spreading on land or in fertilizers. In this case, the regulatory constraints already outlined for fertilizers and biostimulants would prevail. However, if they do not comply with these regulations (e.g. due to arsenic content), they might also have to be treated more specifically, or incinerated.

Water

Wastewater from seaweed processing has to be considered with care, especially as most soluble forms of arsenic, and particularly inorganic arsenic, are likely to be solubilized to a significant extent, as described earlier in this review for *S. fusiforme* treatments (Hanaoka et al. 2001; Ichikawa et al. 2006; Katayama 2015). Leachates from *Sargassum* spp. storage sites should also be monitored.

A good example is the case of a French processing plant-producing alginates and processing 50 kT of *Laminaria* spp. per annum. The company improved their wastewater treatment unit in order to improve the quality of the water released into the environment, with a specific focus on arsenic content. The improved process, in turn, increased the arsenic content in the sewage sludge to 159 mg kg^{-1}, which significantly impacted their sewage sludge–spreading plan (Préfecture du Finistère 2014). This example illustrates how large-scale processing of arsenic-rich *Sargassum* spp. species may imply significant work on effluent clean-up. Beyond biological treatment, various technologies for arsenic removal from contaminated waters have been described. They include ultrafiltration (mostly for drinking water due to elevated cost and sensitivity to fouling), precipitation with iron oxides or treatments with magnesium salts (e.g. $MgCl_2$) (da Silva et al. 2019).

However, it is difficult to list general regulatory limits for contaminant content in effluents, as these limits depend on various factors:

- Effluents treated further in a municipal wastewater treatment plant or sent back to the environment
- Volumes of effluents produced, dilution
- Presence of contaminants or chemicals which might impact ecosystems or wastewater treatment efficiency
- Local limits: e.g. technologies in place and treatment capacity of the downstream municipal wastewater treatment plant
- Sectorial limits with norms set depending on the type of industries, and available/affordable technologies, e.g. US National Pretreatment Program and National Pollutant Discharge Elimination System (NPDES), European Best Available Techniques Reference documents (BREFs)

Companies usually have to register their uses and characterize/control their effluents (volumes, physico-chemical parameters, composition, etc.) and apply for specific authorizations, which may come in several forms depending on national regulations, and the volumes/contaminants concerned (e.g. Facilities Classified for Environmental Protection (ICPE) in France, Industrial User Permit in the USA). Additionally, registers may have to be held for specific contaminants emitted or transferred to the environment (Table 12).

Sewage sludge (biosolids)

Disposal of sewage sludge usually depends on several parameters. In Europe they can be directly considered as fertilizers if their content complies with national or European norms. Alternatively, they can be integrated in spreading plans for sludge from wastewater treatment on agricultural land (Table 13), in which case their nutrient content also needs to be integrated in the local/farm nutrient management plan. Spreading on non-agricultural land can also be considered, sometimes with higher limits (e.g. 40 CFR 503.13 in the USA).

It should be noted that the European Union has no specific limits for arsenic in sewage sludge to be used in agriculture. However, although France does not regulate arsenic in sewage sludge either, the values allowed for fertilizers in France could be considered as a reference (90 g ha^{-1} year^{-1} over 10 years). In the example mentioned previously (Préfecture du Finistère 2014), sewage sludge from a seaweed processing plant in France reached 159 mg kg^{-1} of arsenic, which is above the

Table 12 Reporting thresholds (kg year^{-1}) for arsenic and heavy metals released into the environment

Compound	France
Lead (Pb)	200
Arsenic (As)	20
Cadmium (Cd)	10
Chromium (Cr)	100
Mercury (Hg)	10
Nickel (Ni)	50
Antimony (Sb)	10
References	31 January 2008 ruling

limit set in the USA for agricultural use, or for French fertilizers. However, with a spreading plan considering an average sewage input of 500 kg ha^{-1} year^{-1}, the arsenic load is brought down to 80 g ha^{-1} year^{-1}, below the maximum flux allowed in France, and 500 times below the cumulative loading rate allowed in the USA. But at this level, the contribution in nutrients is very low compared to standard sludge/compost input of 5–30 t ha^{-1} and requires significant surfaces (3600 ha in their case). This is more a way of disposing of the waste, rather than a true contributor to the nutrient plan, even if other residues/compounds from the seaweed processing might have benefits for the plant.

Beyond these direct uses, sewage sludge can also be processed further to compost (e.g. complying with the norm NF U 44-095 in France), integrated into biogas production units or incinerated. Long-term arsenic immobilization could be improved further by preliminary treatment with calcium salts (e.g. lime) before sludge incineration (Fujita et al. 2000).

Air

Specific limits for arsenic and heavy metals are set for incineration of sewage sludge and wastes. For example, the US EPA determined limits (expressed in μg m^{-3}) for incineration of sewage sludge (40 CFR 503.43), which allow, depending on the system used, calculation of maximum levels of these compounds in the sewage sludge to be incinerated. Similarly, the European Directive 2010/75/EU establishes air emission limits for waste incineration plants and waste co-incineration plants for cadmium/thallium, mercury and the sum of other heavy metals including arsenic.

During incineration of biomass contaminated with heavy metals, they are mostly recovered as ash (mostly bottom ash and cyclone ash when cyclone is present), even if they are also found in fly ash (partly recovered on filters, although very fine fly ash will be lost with flue gas) (Nzihou and Stanmore 2013). Therefore, their loss is mostly in accordance with the efficiency of gas cleaning for particulates. However, more volatile metals like mercury, selenium and, to a lesser extent, arsenic can also be lost as vapours.

In the case of arsenic, retention in bottom and cyclone ash is relatively good up to 600 °C (Nzihou and Stanmore 2013) or even 800 °C (Gerstle and Albrinck 1982) but significantly decreases at higher temperatures, which should be avoided. However, controlling the combustion parameters (e.g. at 800–950 °C with high O_2 content) can allow for the better capture of arsenic in fly ash through condensation/absorption on particulates and conversion into its less toxic As(V) form (Zhao et al. 2018). Incinerating with co-substrates rich in compounds such as calcium, or iron, is also reported to help reduce the toxicity of the fly ash by increasing oxidation and absorption (Zhao et al. 2017).

However, final arsenic content in the ashes may exceed regulatory limits used in agriculture, for example, or may even lead to the ashes being classified as toxic waste, which are not allowed for disposal in standard landfills. Clean-up of flue gases and filters also generates additional liquid effluents, which may contain non-negligible amounts of contaminants.

Table 13 Maximum levels (mg kg^{-1} dry weight) and maximum input (g ha^{-1}) of arsenic, heavy metals and contaminants for sewage sludge used in agriculture

Compound	France		Europe		USA	
	Max level*	Max 10-year flux*	Max level	Max 10-year flux	Max level	Cumulative loading rate
Total arsenic (As_t)					41	41,000
Inorganic arsenic (As_i)						
Cadmium (Cd)	10	150	20–40	1500	39	39,000
Chromium (Cr)	1000	15,000	–	–		
Mercury (Hg)	10	150	16–25	1000	17	17,000
Lead (Pb)	800	15,000	750–1200	150,000	300	300,000
Nickel (Ni)	200	3000	300–400	30,000	420	420,000
PCB**	0.8	12				
Fluoranthene	5	75				
Benzo(b)fluoranthene	2.5	40				
Benzo(a)pyrene	2	30				
References	17 August 1998 ruling		Directive 86/278/EEC		40 CFR 503.13	

*General case: different limits apply for pastures and soils with pH < 6

**Main PCB: 28, 52, 101, 118, 138, 153 and 180

Conclusion

Various members of the brown algal genus *Sargassum* spp. are reported as contaminated by heavy metals and, above all, (total) arsenic. *Sargassum* spp. valorization is a challenging economic opportunity, but industrial applications in food, in agriculture or for alginate extraction, especially from some Asian species, confirm its feasibility. Reactivity and organization are also a key to exploit such biomass, as most industrial processes require stable supply of well-preserved and sand-free algae, which is difficult to obtain from beach-cast biomass.

The high total arsenic content is an obstacle to food and feed valorization, even if *Sargassum* spp. consumption is traditional in Southeast Asia and other parts of the world, as food regulations drastically limit the arsenic content allowed. Some processes, reducing the arsenic concentration in *Sargassum* spp., have been developed in Asia, especially for *S. fusiforme*. Its raising consumption is a concern from food safety agencies worldwide. Transposition of these processes to other applications could be considered, especially for the extraction of value-added compounds (nutraceuticals, cosmetics, etc.), but may require further research to limit processing costs.

Sargassum valorization in agriculture, especially as compost and organic amendment, remains a challenge because of the volume involved and the need to reduce arsenic and sodium levels to acceptable levels, but incorporation in composts proves to be feasible, and more value-added applications, such as plant biostimulants, should be investigated further.

Lastly, energy production from *Sargassum* or from waste streams from its processing should be further explored. Whilst limited added value can be expected, this process could contribute to dealing with the volumes of seaweed collected, whilst concentrating the contaminants in a form that might be more easily handled. Similarly, the use of *Sargassum* spp. in biomaterials warrants higher academic interest to support industry developments. However, the fate of arsenic, i.e. metabolism, location of adsorption, relationship with transudate and so on, must be elucidated to support public authorities and decision-makers involved in *Sargassum* crisis governance. Moreover, arsenic transformation in industrial processes should be clarified further, at least to underpin regulatory compliance, at best for pre-treatment process optimization and waste management (liquid, gaseous and/or solid ones).

In some studies, organic micropollutant concentrations in *Sargassum* spp. also reach concerning levels for a marine matrix, considering that marine matrices classically exhibit low levels due to dilution. However, organic micropollutants are not routinely monitored. Studies are scarce and related to specific pollution contexts, as the Deepwater Horizon event. Organic micropollutant contents reported have thus to be regarded with caution. Data is lacking to determine whether *Sargassum* spp. present a higher risk of organic micropollutant contamination compared to other marine matrices, and whether this might impact their downstream uses.

Sargassum spp. are a promising matrix triggering renewed interest in response to recent *Sargassum* spp. blooms and the large volumes of biomass available. Promising valorization routes, especially in industrial fields, should be explored further with a focus on processes and their impact on contaminants.

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