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TREND EDITORIAL

The silent spring of *Sargassum*

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Sargassum is a genus of brown macroalgae (Phaeophyceae, Fucales) spread worldwide in tropical, subtropical, and temperate environments. All *Sargassum* species are benthic except for two pelagic ones: *Sargassum natans* and *S. fluitans*, previously confined to the Gulf of Mexico and Sargasso Sea and now drifting on the surface of the tropical Atlantic Ocean where they accrete together into windrows of floating thalli, ranging in size from large patches to rafts of several hundred metres in length (Ody et al. 2019). Climate events (hurricanes, rises in seawater temperature) and sea current events (anomalies in surface currents) could spread them onshore, but those algae have never been reported significantly elsewhere. However, since 2011, large beaching events have been occurring on the shores of West Africa (Sankare et al. 2016) and the Greater Caribbean (Langin 2018). Initially episodic, such beachings are tending to become increasingly frequent and massive, due to an unexpected location of growth in front of the Amazonian Mouth (Johns et al. 2020). The hypotheses proposed to explain this are a link with some cause within the raft (Lapointe et al. 2014), the deforestation of inner Amazonia, and/or African coastal mangroves or the expansion of the Sahara (Sissini et al. 2017; Oviatt et al. 2019; Wang et al. 2019). The large volumes involved show a trend of yearly increase and can be expressed in millions of tons of algae that have drifted to shores, leading to severe impacts on the environment and human health: *Sargassum* pile up on the shore, then decay, producing foul and corrosive hydrogen sulphide, and depleting oxygen in water; colloidal bleed is also observed (Perry et al. 2018). Such events have overwhelmed public authorities and endangered economic activities, mainly in the tourism sector and port industries and infrastructures. *Sargassum* is also described as a "health issue" by France's regional health agencies, because it now represents a real threat to human health. Several French doctors have already sent out alerts on this problem through a publication (Resiere et al. 2018). They confirm that there is a pressing need to discuss this matter at an international level with a view to boosting marine research, pooling resources, and consolidating local political priorities. Between January and August 2018, health professionals reported 3341 cases in Guadeloupe (West Indies) and 8061 cases in Martinique (West Indies), of which three patients were admitted to intensive care. Otherwise, beaching consequences are currently under intensive study as macroscopic pollution. However, *Sargassum* beachings pose another more insidious threat. Until now, arsenic concentration in geochemical backgrounds of the Caribbean islands has generally been low, but Michel (1985), Muse et al. (1989), Neff (2002), Pell et al. (2013), and Devault et al. (2019) have highlighted that the *Sargassum* content of arsenic gives cause for concern: arsenic (As) is a notorious toxic metalloid. Naturally present in soils and water, arsenic is commonly found in sulphide-bearing mineral deposits mainly associated with gold, antimony, silver, lead, copper, tin, zinc, and iron minerals. Amongst the three other valency states of As (As-3, As-III, and As-V), As-3 is found only at extremely low redox values, i.e. in highly-reducing environments (Moore and Ramamoorthy 1984). Arsenite

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As-III is dominant in reducing environments, and arsenate As-V prevails in oxidising conditions: As-III and As-V are observed in marine conditions, with As-V being more commonly reported because the oxidising environment is more common than the reducing one (Sadiq 1990; Anderson and Bruland 1991a, b; Francesconi and Edmonds 1998; etc.). Above all, arsenic is a Va group element and is related to other Va group elements (e.g. antimony and phosphorus): its behaviour in the environment is close to that of phosphorus (Neff 2002), leading As to compete with phosphorus for adsorption sites (Neff 1997). Arsenate can be assimilated by algae in the As-V oxidation state through phosphate transporters, as described by Bouain et al. (2014), owing to its physicochemical similarities with P ($H_2PO_4^-$ versus H_2AsO4^-) (Taylor and Jackson 2016), leading to a subsequent decrease of phosphorus in the tissues of algae receiving rising concentrations of As(V). Arsenate is predominant in organic aqueous and aerobic environments and is strongly adsorbed on to the surface of several aquatic organisms and oxidised minerals of Fe, Mn, and Al (Al Mamun et al. 2018 and 2019), including inner iron plaques in *Sargassum*. But mobilisation via reduction processes can occur in the water column or sediments. Previous studies have highlighted that the average concentrations recorded for floating *Sargassum* thalli range from 80 to 140 mg/kg of the dry weight (dw) of the alga (Michel 1985; Devault et al. 2019): no explanation has yet been found for such variations. Given the mean concentration in arsenic, assessing the total amount of stranded *Sargassum* on a given territory requires considerable resources that have not yet been utilised for a whole episode. However, a one-shot complete assessment performed on September 27, 2014, in Martinique (West Indies) during an airborne survey concluded that there was a total amount of 34,000 m^3 of fresh floating *Sargassum* contiguous to the shoreline. Considering an average of 250 kg/m^3, 31% dry weight rate, and an arsenic concentration of 80 mg/kg (dw), the total influx of arsenic was about 210 kg in only a few days. Despite variations in time and intensity, the Martinique shoreline has experienced a total of 37 months of heavy stranding episodes since 2011. In theory, this has led to several tons of arsenic spread along the exposed coastline of the island due to *Sargassum* beaching. Occurring year after year, is it possible for this influx to contaminate biota? In other words, is this arsenic bioavailable?

When *Sargassum* thalli are massively accreted in shallows or on floating dams in stress conditions (which have to be defined more precisely), they transudate—a phenomenon that is visible to the naked eye. Such transudation has scarcely been studied (de Carvalho et al. 1994; Figuiera et al. 2000; Davis et al. 2004; Veit et al. 2014; and more recently, Perry et al. 2018), but has been identified as colloidal leaks. The phenomenon has never been evaluated quantitatively, and indirect data show a range from 1.6 to 49% of dried biomass (Veit et al. 2014 and Figuiera et al. 2000, respectively) depending on the species studied and how the biomass is handled. Ender et al. (2019) propose that the bulk of the arsenic is adsorbed on the cell walls due to the polysaccharides that structure the cell wall of brown algae, therefore due to the alginate content. If oceanic streams do not drift the raft westward into the Caribbean Sea, stressed *Sargassum* sink and their decaying close to the sediment produces a mud close in density to fluid mud (as defined by Thouvenin et al. 1994; Abril et al. 2003, 2004); the fate of arsenic content in fluid mud is unknown, including its speciation in the more toxic As-III, but arsenic input may be expected to be mostly bioavailable.

Moreover, the Caribbean population traditionally eats bivalves, which are known for filtering high volumes of seawater and for bioaccumulating micropollutants. The average fish and other seafood consumption is apparently well below the world average (8.96 versus 18.98 kg/capita/year) (Faostat 2013), probably also because shellfish and especially bivalves are not a traditional element in the diet of Caribbean peoples (Josupeit 2011). However, bivalves are widely targeted by the subsistence coastal fisheries in the Caribbean. Local artisanal fisheries play a significant role in the livelihoods and food security of more than two million people (de Oliveira Leis et al. 2019). Many healthcare professionals are seeing an increase in consultations with local populations due to the effects of chronic exposure to hydrogen sulphide (Resiere et al. 2018). However, the health consequences of long-term exposure to arsenic from bivalves remain poorly documented and impede hospitals from understanding this reality. This value can reach more than 5 mg/kg (dw) (Sloth et al. 2005), but as Francesconi (2010) and Zmozinski et al. (2015) have demonstrated, trimethylated arsenobetaine dominates quantitatively in the multiple arsenic species in seafood due to arsenic speciation. Amongst the tri- and tetramethyl As forms, including arsenobetaine, arsenic toxicity varies according to the degree of methylation: in decreasing order, MMA(III) and DMA(III) > arsenous acid (As(III)) > As acid or arsenate (As(V)) >> monomethylarsonic acid (MMA(V)) ≈ dimethylarsinic acid (DMA(V)) > trimethylated species (arsenobetaine (AsB), etc.) (Geng et al. 2009).

Arsenobetaine is known to be slightly toxic ($LD_{50} \approx$ 10 g/kg), but the other forms have acute LD_{50} of 5–100 mg/kg_{body}: with chronic tolerance daily intake (TDI) for inorganic arsenic of 0.45 $\mu g_{inorganic\ As}/kg_{body}$/day, 27 μg/day is a sanitary threshold following the international standard for an adult (60 kg). TDI for organic species is not known, partly because data for MMA(III) and DMA(III) are scarce, and partly because the other organic species of As-V are slightly toxic. During beaching events, bioavailable arsenic accumulation is not studied, nor the conversion of the seaweed arsenosugars, and the time needed to turn input of inorganic arsenic into

arsenobetaine needs to be determined, at least to estimate the duration between the *Sargassum* beaching and re-entry to the harvesting site.

The challenge is compounded by the lack of knowledge about arsenic speciation and bioavailability and about coastal streams inducing dilution and drift. However, year after year, arsenic contamination will be maintained by the increasing beached volumes. Such a concern has to be solved at an international scale due to the extent of the problem, but collaboration at the scale of beaching events, i.e. along West Africa, the USA, and the Caribbean basin, is in its infancy. At the local scale, arsenic concern is ignored by most stakeholders and the governance is not aware of it, except for the French West Indies; yet even for the FWI, there is room for improvement and scientific means must be mobilised. In particular, caution must be exercised with regard to the use of stranded *Sargassum* biomass by local populations.

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