

Micropollutant content of Sargassum drifted ashore: arsenic and chlordecone threat assessment and management recommendations for the Caribbean

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Abstract

Massive Sargassum beachings occur since 2011 on Caribbean shores. Sargassum inundation events currently involve two species, namely *S. fluitans* and *S. natans* that are circulating and blooming along the North Atlantic subtropical gyre and in the entire Caribbean region up to the Gulf of Mexico. Alike other brown seaweeds, Sargassum have been shown to bioaccumulate a large number of heavy metals, alongside with some organic compounds including the contamination by historical chlordecone pollution in French West Indies (FWI), an insecticide used against the banana's weevil *Cosmopolites sordidus*. The present study reports, during two successive years, the concentration levels of heavy metals including arsenic in Martinique and Guadeloupe (FWI). We found that Sargassum can also accumulate high concentration of chlordecone. Sargassum contamination by chlordecone is observed in areas that are close to contaminated river mouth. Our results further demonstrate that algae bleaching raises a number of questions about inorganic and organic pollutants bioaccumulation, transport and dissemination.

1. Introduction

Sargassum (Fucales, Phaeophyceae) are present worldwide in tropical and subtropical environments (Niermann, 1986). All the described *Sargassum* species are benthic except two pelagic ones: *Sargassum natans* and *Sargassum fluitans*, initially reported by C. Columbus (Lapointe and Hanisak, 1985), and mainly confined to the Gulf of Mexico and Sargasso Sea where they are found accreted together into lines and mats (Marmorino et al., 2011). Even if climatic events (e.g. hurricanes) could spread them onshore, pelagic *Sargassum* species were never reported elsewhere than the Caribbean area (Loffler and Hoey, 2018). However, since 2011, large beaching events are occurring on both West Africa and Greater Caribbean shores (van Tussenbroek et al., 2017). Firstly episodic, such events tend to be more common in part due to an unexpected growth location in front of Amazonian mouth, and volumes involved are yearly expressed in millions of tons of algae drifted to shores (Wang et al., 2019). *Sargassum* are piled up on shore, then perished, producing foul and corrosive hydrogen sulfate, oxygen depletion in water and colloidal bleed is observed (Brylinsky, 1977), and can also have impacts on human health (Resiere et al., 2018). Such events overwhelmed the public authorities, endangering economical activities, mainly tourism and port ones (Langin, 2018).

Seaweeds are well known to concentrate metals from seawater in the different parts of their thallus, and a wide range of both essential and non-essential metals have been measured (Malea and Kevrekidis, 2014; Bonanno and Orlando-Bonaca, 2018; Garcia-Seoane et al., 2018). Among heavy metals, Arsenic (As) is a notorious and toxic metalloid, ubiquitous in the environments which is accumulated in algae including *Sargassum* spp. and can potentially also be found in algal food or derived products (Devault et al., 2020a; Magura et al., 2019; Milledge et al., 2020 ; Rodriguez-Martinez et al., 2020).

Arsenic chemical behavior in the environment is close to the phosphorus one (Neff et al., 2002). As such, phosphate and As(V) can compete for adsorption sites (Neff et al, 1997). Arsenate (As(V)) is predominant in inorganic 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., 2018 and 2019), including inner iron plaques in *Sargassum*. Arsenite (As(III)) is highly abundant under anoxic environments as oxyanion (Smedley and Kinniburgh 2002) and more toxic than As(V) (Ferguson and Gavis, 1972). As can change in the environment in quickly and repeatedly from one specie to the other –however, methylation preserves the As speciation in organic As compounds.

Along with heavy metals, and especially As, *Sargassum spp.* are known to accumulate organic micropollutants, even if poorly documented, as summarized Devault et al. (2020a): Polycyclic Aromatic Hydrocarbons accumulated into algae driven by aromaticity level (Bertilsson and Widenfalk, 2002; Tobiszewski and Namiesnik, 2012; Seepersaud et al., 2018) and, Stout *et al.* (2018) confirmed for *S. natans* and *S. fluitans* during the offshore Deep Water Horizon MC 252 blowout in the Gulf of Mexico and the subsequent oil spill. For pesticides, sorption behaviour is rather complex. Even if hydrophobicity broadly induces similar behaviour, especially for older formulations, more recent pesticides are partly designed to delude plant metabolism.

Chlordecone (CLD) is also a well-known pesticide impacting French West Indies (Cavelier 1980 ; Bocquené & Franco, 2005; Coat et al., 2006 ; Dubuisson et al., 2007, Gourcy et al., 2009 ; Le Déaut & Procaccia, 2009, Jondreville et al. 2014 ; Devault et al., 2016 amongst many). Notwithstanding the worldwide spread of this pesticide, However, the climactic area where CLD was spread is the West Indies. French West Indies were selected in order to determine the extent of such pollution: French West Indies were selected to study the As and chlordecone content of *Sargassum* drifted to shore. Moreover, French public agency for energy and environmental management (ADEME) supports the attempts for collecting and valorizing the large volumes of *Sargassum* in order to turn this disadvantage into an asset: contamination assessment has to be performed in order to substantiate such perspectives. In this article, authors focused on *Sargassum* beached in Martinique and Guadeloupe shores. Our study aims at quantifying both (i) the organic micropollutant chlordecone and its derived metabolites as well as (ii) the different species of arsenic present in *Sargassum* samples found along the shore of two islands of the French West Indies during two successive years.

2. Material And Methods

2.1. Sampling strategy

Sampling was performed during two campaigns in Martinique and in Guadeloupe. The 2018 campaign was between the 26th and 28th in July for the Guadeloupe Archipelago, and between 30th July 30 and 2rd August in Martinique. The second campaign was early 2019 between 27th February to 4th of March for the Martinique, and 12th March 12 to 4th of April for the Guadeloupe. Most of the samples were collected either on the beach or on the near shore (with a maximum of 500 m from the coast). For the 2019 campaign, most of the time we collected at the same site two onshore samples, one sample of fresh *Sargassum* beached on the sand a one sample corresponding to dry *Sargassum* again lying on the sand.

Samples were collected, during the first rounds, in order to be representative to the low-scale heterogeneity, *i.e.* sampled threefold at few distance on the shore of the same site. Samples collected during the second round were collected at the same place but following the four following conditions: (1) grounded and at least partly dried and browned, being drifted to shore but not yet grounded, (2) recently grounded, *i.e.* still wet, (3) close to be grounded, in the beating of the waves, and (4) still in seawater. Each condition required was filled as possible: sampling was submitted to presence or absence of algae at sampling date and site. Practically, sampling was not significantly impacted and lacking samples were rare.

2.2 Investigated sites

Sampled sites were selected because they are on the windward coast of Guadeloupe and Martinique. The Basse-Terre windward coast in Guadeloupe, *i.e.* the mountain part, and the north of Martinique windward coast are known to have surface water and groundwater discharge contaminated by chlordecone. Moreover, *Sargassum* mats being drifted to the shore by wind and currents, windward coast is the most impacted by *Sargassum* groundings, whatever the island –so authors sampled only windward coast, *i.e.* eastward coast. Sampling sites are spaced of about 10 km of Guadeloupe coastline and 5 km of Martinique one.

Over the 14 sampled sites in Guadeloupe, 7 were in Basse-Terre shores and 7 in Grande-Terre shores. Over the 20 sampled sites in Martinique, 9 were in an area expected to be contaminated by chlordecone (restricted area for fishing and gathering), 8 were apart from the restricted area, and 3 were piled-up area, *i.e.* sampled in *Sargassum* piles, formed by drifted algae brought together in order to de-clog the ports and beaches. The 3 piles were only sampled during the first campaign. The sampling locations are presented in Figure 1.

2.3. Analytical methods

2.3.1. General sample preparation

After material sampling in Guadeloupe and Martinique, samples were frozen in dry ice and dispatched to La Drôme Laboratoire in metropolitan France in the 48h following sampling. The overall sample quantity was grinded with a Grindomix GM 200 Retsch apparatus till ($< 63 \mu\text{m}$). After this first grinding, dry matter and mineral matter were determined thanks to NF EN 12880 (Chiffre et al., 2015) and NF EN 15169 standard (Dia et al., 2019), respectively.

2.3.2. TOC and N-Kjeldahl analysis

Total Organic Content (TOC) and N-Kjeldahl were carried out after freeze-drying of previous grinded samples on a CHRIST Alpha 1-2 LD Plus. TOC was determined following the NF ISO 14235 (Remon et al., 2005) operating procedure including hot sulfochromic oxidation and spectrophotometry quantification (LQ = 1 000 mg(C) per kg of dried sample (DS)). N-Kjeldahl analysis followed the NF EN 13342 standard

involving hot sulphuric acid mineralisation with selenium catalyst (Guilayn *et al.*, 2020). Titrimetry analysis allowed to determine N-Kjeldahl content (LQ = 200 mg(N) per kg of dried sample (DS)).

2.3.3. Metal and metal speciation analysis

The list of analyzed metals included As, Cd, Cr, Co, Cu, Ni, Pb, Zn, Hg, P and S. For the overall metal content and metal speciation, previous grinded samples were mineralized with nitric acid in micro-waves following the NF EN 13805 procedure (Chevallier *et al.*, 2015). Quantitative metal analysis was achieved thanks to ICP-MS X-serie apparatus from Thermofisher. Metal determination followed the NF EN 17294-2 standard (Glorennec *et al.*, 2010). For total metal content, limit of quantification in mg(metal) per kg of dried sample (DS) are listed in Table 1.

Table 1
Limit of quantification for metal compounds
(dried sample, DS).

| Metal compound | LQ | unit |
|-----------------------|-----------|--------------|
| As (total) | 0.05 | mg(As)/kg DS |
| Cd (total) | 0.05 | mg(Cd)/kg DS |
| Cr (total) | 0.10 | mg(Cr)/kg DS |
| Co (total) | 0.05 | mg(Co)/kg DS |
| Cu (total) | 0.10 | mg(Cu)/kg DS |
| Hg (total) | 0.05 | mg(Hg)/kg DS |
| Ni (total) | 0.10 | mg(Ni)/kg DS |
| Zn (total) | 1.00 | mg(Zn)/kg DS |

Metal speciation allowed to determine As and Hg species. As compounds were As (total), AsIII (arsenite), AsV (arsenate), MMA (monomethylarsonic acid), DMA (dimethylarsinic acid), AsB (arsenobetain) and AsC (arsenocholin). Hg compounds were Hg (total), iHg (inorganic Hg) and MMeHg (monomethylHg). Arsenic speciation analysis was carried out on a ICP-MS Nexion 300 apparatus from Perkin Elmer coupled with a liquid chromatography supplied by Flexar. Hg speciation was carried out on the same ICP-MS Nexion 300 apparatus from Perkin Elmer but coupled with a gas chromatography 680 supplied by Clarus. For all metal species, the limit of quantification are summarized in Table 2.

Table 2
Limit of quantification for As and Hg compounds (dried sample, DS).

| Metal specie | LQ | unit |
|--------------------------------|------|----------|
| As (total) | 0.20 | mg/kg DS |
| AsIII (Arsenite) | 0.05 | mg/kg DS |
| AsV (Arseniate) | 0.05 | mg/kg DS |
| MMA (Monomethylarsonique acid) | 0.10 | mg/kg DS |
| DMA (Ddimethylarsinique acid) | 0.10 | mg/kg DS |
| AsB (Arsenobetain) | 0.10 | mg/kg DS |
| AsC (Arsenocholin) | 0.10 | mg/kg DS |
| Hg (total) | 0.50 | µg/kg DS |
| iHg (Inorganic Hg) | 0.50 | µg/kg DS |
| MMeHg (MonomethylHg) | 0.50 | µg/kg DS |

2.3.4. Chlordecone and chlordecone metabolites analysis

Following the protocol detailed in Devault et al. (2016), chlordecone and chlordecone metabolites (5b-hydro and chlordecol) analysis was carried out on the previous grinded samples. 150 µL of ¹³C chlordecone tracer was added to 10 g of grinded sample. 20 mL of acetone was mixed and stirred with the previous mixing for 24 h. After filtration sample was washed two times with 5 mL of acetone. Chlordecone compounds were extracted after stirring with a dichloromethane solution (20 mL of CH₂Cl₂ in 175 mL of H₂O saturated with NaCl salt). After two extractions, the CH₂Cl₂ recovered was concentrated to 1 mL by dried evaporation (N₂). 1 mL of hexane was added to the previous CH₂Cl₂ concentrated solution. For liquid chromatography analysis the sample had to be changed. Chlordecone compounds were extracted from 200 µL of CH₂Cl₂ and hexane solution thanks to 1 mL of H₂O / acetonitrile solution. Chlordecone compounds were analysed by Acquity UPLC System with XevoTQ-S Mass spectrometer (LC-MS-MS). LC conditions and MS-MS results exploitation are presented in Table 3. The LQ in µg (chlordecone compound) per kg of fresh sample (FS) are listed in Table 4.

Table 3
LC-MS-MS operating conditions.

| Apparatus | Operating conditions |
|-----------|---|
| LC | <p>Solvent A : H₂O and 0.1% formic acid</p> <p>Solvent B : Acetonitrile</p> <p>Flow rate : 0.4 mL/min (0.5 min 80% of solvent A and 4 min 100% of solvent B)</p> <p>Injection : 10 µL</p> |
| MS-MS | <p>Negatif mode sampling</p> <p>Chlordecone :</p> <ul style="list-style-type: none"> • First transition : 506.9/426.78 • Second transition : 506.9/424.8 <p>5-b-hydro :</p> <ul style="list-style-type: none"> • First transition : 472.96/392.88 • Second transition : 472.96/390.8 <p>Chlordecol :</p> <ul style="list-style-type: none"> • Transition 490.2/490.2 <p>Internal standard :</p> <ul style="list-style-type: none"> • Transition Chlordécone C13: 516.9/435.87 <p>Injection label :</p> <ul style="list-style-type: none"> • Transition 24D-D3 : 221.9/163.82 |

Table 4
Limit of quantification for chlordecone compounds (fresh sample, FS).

| Chlordecone compound | LQ | unit |
|----------------------|-------|----------|
| Chlordecone | 0.50 | µg/kg FS |
| 5-b-hydro | 10.00 | µg/kg FS |
| Chlordecol | 0.50 | µg/kg FS |

2.4. Data analysis

Statistical analyses were performed with R version 4.0.3 (R Core Team (2020). R: A language and environment for statistical computing. R Foundation for Statistical Computing, Vienna, Austria. URL <https://www.R-project.org/>). Data were visualized using the *ggplot2* package (Wickham, 2016). Differences in chemical concentrations or ratios between treatments were evaluated a Kruskal-Wallis rank sum test followed by a Dunn test with Benjamini-Hochberg corrections.

3. Results And Discussion

Contamination pattern, in Guadeloupe and in Martinique, involves both (1) *Sargassum* offshore contaminated by oceanic As, mainly resulting from the human activities at global or local scale, and (2) *Sargassum* contamination by chlordecone in coastal environment due to water resource and soil pollution by banana weevil control during 70's and 80's (Bocquené and Franco, 2005). All the results are detailed in the Appendix 1.

3.1. Arsenic

As content offshore has been ensured by Atlantic Ocean samplings in front of Santa Lucia Channel (identified as "Transect" in Table A1 of Appendices): As(V) was the dominant form of arsenic (mean \pm standard deviation, $58.2 \pm 18.0\%$ of total As), corroborating international literature (Michel, 1985). Climactic As content (125.7 mg/kg dw) is observed since pelagic sampling sites and higher As concentrations in *Sargassum* ashore, whatever island or sampling sites, are related to drifting algae in opposition to dried ones (Figure 2). We found significantly less total As, As(V), MMA and AsBet in the dried samples from the beach than in the floating algae ($P < 0.05$). For the As(III) and DMA species, no significant differences were observed (Figure 2). *Sargassum* are floating algae and beaching often involve a first step of compression of massive *Sargassum* biomass along the shoreline, during which *Sargassum* density will exceed the density in the mat. At this step, which have to be more studied to properly understand the inducing parameter(s), *Sargassum* will be stressed and will transudate. Transudation content is poorly known but authors identified that chlordecone and As are significantly transudate in few hours for floating and sank algae (Devault et al., 2021). Due to tide, gravity waves and wind, a part of *Sargassum* could beach and be dried in the sun: due to rain and tissue degradation, *Sargassum* will leach: this leach can reach $22,000 \text{ }\mu\text{g/L}$ (Chevalier, pers. com.). Another part can sink and, in case of cup-shaped dent from which *Sargassum* could not drift away, *Sargassum* biomass could form a pseudo-vase by sedimentation in shallows. In the present study, authors did not focus in such environments, but limited them to floating *Sargassum*, beached and fresh ones, beached and dried ones, and piled ones, but two sampling sites correspond to such extreme accretion situation.

Comparing the results of our study with the current literature is delicate because the analysis proposed elsewhere about As were mainly performed on total As content, even including inorganic As species (Maher et al., 1983), focuses on the protocol enhancement (Han et al., 2009), or on the metabolic pathways (Edmonds et al., 2009 ; Pichler et al., 2006 ; Leal-Acosta et al., 2013) not about detailed environmental monitoring (Devault et al., 2020a) or did not include As speciation detail Rodríguez-

Martínez *et al.* (2019). As such, the detailed protocol proposed in the present article provided new insights into As dynamics in algae. Total As concentrations into *Sargassum* exceed the concentration known for the other algal species but are in agreement with previous results for those species (Michel, 1985; van Tussenbroek *et al.*, 2017). This upper (at least double) As concentration could be due to pelagic conditions: the other *Sargassum* species are benthic ones and phosphate concentration in shallows is higher than in deep-sea so the phosphate need is higher.

Benson (1984) and fellows hypothesized that orthophosphate and As are competitive for phosphate transporter and that *S. natans* and *fluitans* overuse of phosphate transporter to compensate the phosphate lack in the open ocean, increasing As accumulation. Total As content heterogeneity is limited: if piled *Sargassum* presents a significantly lower As content than other ($26.6 \pm 20.9 \mu\text{g/g}$ and $80.9 \pm 29.8 \mu\text{g/g}$ dw, respectively), piled *Sargassum* sampling sites, located in Martinique, are scarce. Total concentration, at $80.9 \pm 29.8 \mu\text{g/g}$ dw (minimum 28.8 and maximum 127 $\mu\text{g/g}$ dw) is very in agreement with Rodríguez-Martínez *et al.* (2019) who reported a median of 80 and a minimum 24 and a maximum of 172 $\mu\text{g/g}$ dw. -This statement has to be limited to total As but, based on the 20 million tons estimation per year of floating *Sargassum* drifted to Caribbean shores (Wang *et al.*, 2019), even in considering a minimized 50 $\mu\text{g/g}$ dw concentration, the total flux of As is yearly about 1 000 tons. Considering the sites where algae were not piled, a significant distinction could be made between the two places where “pseudo-vase” (defined as densely accreted, *Sargassum* amounts decay in shallow waters) were sampled: At surface of the amounts, algae will be parched by sun, forming a few centimeters-thick compact crust (able to support a crawling adult but not a walking one) covering decimeters-thick wet rotten algae. They are stuck in place due to the limited tidal range (about 40 cm) and permanent trade winds and currents. Such sites have total As content not significantly different to piled ones but significantly different with other non-piled *Sargassum* (respectively $22.6 \pm 13.5 \mu\text{g/g}$ and $83.4 \pm 27.7 \mu\text{g/g}$); in such clogged places, interstitial water into the pseudo-vase reaches more than 1000 $\mu\text{g/L}$ due to As transudate from *Sargassum* but also due to limited water circulation. For such concentrations, sanitary impact of As is unknown: dermic way contamination has only been studied until 200 $\mu\text{g/L}$ (Mink *et al.*, 2008; Tsuji *et al.*, 2014) for intact skin when *Sargassum* sp. lugs which harsh the skin: impact at 200 $\mu\text{g/L}$ are controversial for intact skin and have to be comforted for scratched one. Progressive accumulation of *Sargassum*, sank or accumulated as pseudo-vase, is a threat for coastal environment because of this As input at each *Sargassum* event.

As speciation has been detailed for arsenate, arsenite, methylarsonic acid (MMA), dimethylarsinic acid (DMA), arsenobetaine and arsenocholine. All the organic forms are based on pentavalent As. As(V) prevails in oxic conditions (Michel, 1985; Muse *et al.*, 1989; Nekk, 2002; Pell *et al.*, 2013) ($41.8 \pm 25.2 \text{ mg/kg}$ dw for whole samples: $55.7 \pm 18 \mu\text{g/kg}$ dw in Guadeloupe and $33.0 \pm 26.04 \mu\text{g/kg}$ dw in Martinique (but $39.7 \pm 25 \mu\text{g/kg}$ dw without piled *Sargassum*) minimum 0.025 $\mu\text{g/kg}$ dw (0.8 $\mu\text{g/kg}$ dw without piled *Sargassum*) and maximum 106.5 $\mu\text{g/kg}$ dw). Considering the other species of As, they are formed under anoxic conditions (Brown *et al.*, 2010) by reduction of As(V) into As(III), inorganic both but in a reversible process, and microbial pathways to form arseno-organic molecules which lead to a non-reversible speciation (Muse *et al.*, 1989). Comparing Martinique and Guadeloupe results, the presence and

abundance of such non-As(V) species are the more segregating: non-As(V) are, in Martinique, more present than in Guadeloupe: As(III) concentration was about $3.8 \pm 5.2 \mu\text{g/kg dw}$ for whole samples: $2.4 \pm 2.2 \mu\text{g/kg dw}$ in Guadeloupe and $4.9 \pm 6.4 \mu\text{g/kg dw}$ in Martinique (but $4.3 \pm 6.3 \mu\text{g/kg dw}$ without piled *Sargassum*), minimum $0.03 \mu\text{g/kg dw}$ and maximum $29.9 \mu\text{g/kg dw}$, to compare to concentration in MMA ($8.5 \pm 10.8 \mu\text{g/kg dw}$ for whole samples: $3.7 \pm 4.6 \mu\text{g/kg dw}$ in Guadeloupe and $12.2 \pm 12.4 \mu\text{g/kg dw}$ in Martinique - but $14.8 \pm 12.5 \mu\text{g/kg dw}$ without piled *Sargassum*), minimum $0.1 \mu\text{g/kg dw}$ and maximum $42.9 \mu\text{g/kg dw}$ ($40.3 \mu\text{g/kg dw}$ without off-sampled *Sargassum*) and concentration in DMA ($8.5 \pm 10.8 \mu\text{g/kg dw}$ for whole samples: $3.7 \pm 4.6 \mu\text{g/kg dw}$ in Guadeloupe and $12.2 \pm 12.4 \mu\text{g/kg dw}$ in Martinique - but $14.8 \pm 12.5 \mu\text{g/kg dw}$ without piled *Sargassum*), minimum $0.01 \mu\text{g/kg dw}$ and maximum $28.1 \mu\text{g/kg dw}$. In other words, non-As(V) species concentration in Martinique are at least twofold of the corresponding concentration in Guadeloupe, and this ratio often reaches fourfold for the organoarsenic species, even if they are based on As(V). Concentration of arsenite, MMA, DMA are in the same order, and arsenobetaine and arsenocholine in an order less, but with the same higher concentrations in Martinique samples (respectively. $1 \pm 1 \mu\text{g/kg dw}$ in Guadeloupe and $2.9 \pm 3.5 \mu\text{g/kg dw}$ in Martinique and $0.5 \pm 0.4 \mu\text{g/kg dw}$ in Guadeloupe and $1.3 \pm 2.2 \mu\text{g/kg dw}$ in Martinique). Those differences are not significant due to wide standard deviation associated to the spatial heterogeneity and suggest that the fate of As speciation is related to sites. The wide standard deviation is related to spatial heterogeneity, about which the first campaign focused on, but this result is not satisfying and led the authors to sample depending to location on the beach: in water, grounded but still wet, and grounded and desiccated (Figure 2). However, if the ratios between As(V) and the other as form is island-dependent, the considered ratios from the same island are comparable, whatever the location on the shore, *i.e.* all the As forms are transuding with the same intensity whatever the island. Such inter-island difference might be explained by the presence of the genes driving the organoarsenial metabolism (Héry et al., 2008) but a dedicated study to state is needed. This inter-island difference might also be related to geomorphologic patterns; Guadeloupe shoreline being straight but Martinique shoreline being rugged in front of *Sargassum* inputs, authors could hypothesize that *Sargassum* trend to be more often under anaerobic condition in Martinique than in Guadeloupe. Further studies about current and water column oxygenation have to test this hypothesis, including the frequency of the genes driving the organoarsenial metabolism.

Considering As fate for drifted *Sargassum*, authors observed that As content decreases dramatically between floating *Sargassum* to dried ones (Figure 2). This phenomenon cannot be explained by volatilization (As and organoarsenial compounds studied having a volatilization temperature upper than 350°C) nor degradation – not pertinent for elements. If photolysis of organic forms of arsenic is reported (authors only found Howard and Comber, 1992 as reference), this aspect is too weakly studied to decide, and, whatever, it cannot justify the total As decrease. Figure 2 informs about the temporal process because dried *Sargassum* are the oldest, but is this arsenic leaching due to rain, due to daily dipping by tide, or both? The dataset does not allow to determine but further studies have to be performed to understand. Nevertheless, Devault et al. (2021) highlighted that *Sargassum* transudate the main part of their As content in few hours in water column when stressed, addressing the environmental managers reactivity. Considering the second campaign, during which authors distinguished dried and wet algae, the

ratios between As species are not modified whatever the concentration decrease level but As species sums still represented $105.1 \pm 10.1\%$ of total As measurement, discarding the hypothesis of an apparent decrease due to formation of no-analyzed As species. The homogeneous decrease for most of the measured As species (except DMA and As(III)) is in favor with leaching in opposition to a species-dependant metabolisation. As consequence, As leaching, as leachate on beach sand as transudate in the water column, is an As input which can affect the coastal environment in a Caribbean environment where As were never reported as critical in geochemical background.

3.2 C, N, P

We also performed an elemental analysis of the C, N and P contents (Figure 3). No significant difference was observed for the C/N ratio of the sample from different drifted status (Figure 3A). It is noteworthy that this C/N ratio (mean 24.85) would make *Sargassum* potentially compatible with its use as organic amendment. Regarding the N/P ratio, we found a significant increase ($P < 0.001$) in the dried samples from the beach compared to the wet samples and those collected in the sea (Figure 3B). Such increase in the N/P ratio indicates a decrease of P, which could be explained by a phosphorous lixiviation from *Sargassum*. In oligotrophic environments, such input would have to be quantified to determine how this flux can contribute to eutrophication, but bearing in mind the massive scale of *Sargassum* beaching.

3.3. Chlordecone

Sargassum contamination by organic micropollutants has already be reported by Yasmeen et al. (2018) for 48 different molecules in/on *S. wightii*, Stout et al. (2017) and Torralba et al. (2017) for petroleum hydrocarbons due the Deepwater Horizon oil spill, but the contamination by organic micropollutant has not been reported with the same intensity that inorganic ones, particularly As, reported by Devault et al. (2020a). For the present study, results for the chlordecone concentration are presented in Figures 4A and 4B and highlight that organic micropollutants can contaminate *Sargassum* to concerning concentrations.

Chlordecone content is due to terrestrial pollution, leading to a coastal contamination. In this way, concentration level of chlordecone could be directly related to chlordecone-contaminated watersheds. In Guadeloupe, chlordecone has been used in the Southern part of Basse-Terre, *i.e.* the place where *Sargassum* are the most highly contaminated (Figures 4A and 4B, Figure 5). Pérou river, already studied intensively (Coat et al., 2011; Crabit et al., 2016; Mendez-Fernandez *et al.*, 2018), is known to be a major source of chlordecone to the surrounding shell.

Chlordecone is only observed for sites of Basse-Terre of Guadeloupe, where average concentration is 127.0 ± 169.7 $\mu\text{g}/\text{kg dw}$ (minimum: 0.8 $\mu\text{g}/\text{kg dw}$, maximum 616.4 $\mu\text{g}/\text{kg dw}$) for the first sampling campaign and 495 ± 828.5 $\mu\text{g}/\text{kg dw}$ for the five sites concerned for the second campaign (minimum 15.9 $\mu\text{g}/\text{kg dw}$, maximum 2697 $\mu\text{g}/\text{kg dw}$). Concerning all the 7 sampling sites of Grande-Terre of Guadeloupe, chlordecone was never observed. Concerning Guadeloupe, chlordecone was only observed in Basse-Terre of Guadeloupe at places where contaminated watershed are known to discharge at sea (Dromard et al., 2019). In the concerned area fishing and collecting seafood is restricted due to the chlordecone threat. However, chlordecone was observed in Saintes archipelagos (first campaign), at 58.8 ± 13.2 $\mu\text{g}/\text{kg dw}$ and

in Marie-Galante Island, at 7.7 µg/kg dw. These concentration highlight the risk of chlordecone marginal spread due to mats first landed on contaminated shores then drifted to chlordecone-free coastal areas, where Devault et al. (2021) suggested than chlordecone and other micropollutants can transudate because Marie-Galante Island and Saintes archipelagos are chlordecone-free.

5b-hydrochlordecone was detected and quantified only twice, *i.e.* where chlordecone concentrations were maximal. Average concentration was 7.3 ± 4.7 µg/kg dw, ranging from 2.4 to 15.26 µg/kg dw.

As for Guadeloupe, campaigns performed in Martinique sites discriminated by chlordecone are included into restricted area due to chlordecone contamination. *Sargassum* samples contaminated by chlordecone have an average chlordecone concentration of 137.9 ± 90.9 µg/kg dw for the first campaign and 246.6 ± 271.1 µg/kg dw (minimum 4.4 µg/kg dw, maximum 798.9 µg/kg dw) for the second campaign. In opposition to Guadeloupe results, where all the samples from the same sites were contaminated, samples from Martinique show a heterogeneity in the samples, with undetectable concentration and notable contamination on the same place among the replicates. 5b-hydrochlordecone was only observed in high-CLD concentration samples, *i.e.* a ratio about 0.06 ± 0.01 with chlordecone concentration in agreement with Devault et al. (2016).

In the *Sargassum* piled sampled in Martinique, no chlordecone was observed but algae collected were sampled on chlordecone-free shores despite few chlordecone vestigial concentrations. Notwithstanding, other pollutions are reported in the Caribbean, due to crop as house contamination (Devault et al., 2020b) and have to be considered as well.

3.3. Consequences for *Sargassum* issue management

As contamination of *Sargassum* mat is inevitable because As is deep-sea accumulated by algae. Concerning As, the threat imposes to limit beaching and shallows. Boom cover could be a preliminary action : (1) they are efficient to avoid that sand be picked up with beached algae, sand limiting industrial valorization, (2) *Sargassum* decay induces leaks and anaerobiosis, worsening the *Sargassum* effect on biota by oxygen-deprivation, (3) As leaks will be more easily diluted into deeper water column than ashore, (4) chlordecone concentration will be limited due to seawater concentration dilution for estuaries to booms, (5) ease the transport from gathering places to stock even valorization plants.

The As content involves that *Sargassum* have to be picked up as soon as possible, ideally before a day, in order to limit the leak –and before sank, which occurs in about 3 days, as authors observed. *Sargassum* collected have to be gathered in dedicated landfill limiting the groundwater pollution and leaching of *Sargassum* piles have to be treated. As content limits the use of picked up algae: contamination level exceed animal even nutrition, neither crude *Sargassum* deposition on ground, as it occurs illegally. Shore piles on sand ground, the actual way of stock, have to be avoided. Large amounts of *Sargassum*, as observed in Pointe rouge (Martinique) and Porte d'Enfer (Guadeloupe) have to be monitored because population could have to walk the “pseudo-vase” in order to reach fishing device, as observed in the field.

Abrasive for the human skin, this “pseudo-vase” can increase the risk of As contamination due to the interstitial water concentration.

Chlordecone is adsorbed on *Sargassum* in polluted bays: polluted and chlordecone-free *Sargassum* have to be piled separately in order to allow distinct valorization. Drift along the shoreline have to be obstructed in order to limit contaminated mat transfer to chlordecone-free areas, particularly in Guadeloupe where the coastline is less cup-shaped. Even more than for chlordecone-free *Sargassum*, chlordecone content induces that non-authorized *Sargassum* valorization has to be tracked.

Leaching from *Sargassum* occurs in dissolved phase, *i.e.* under labile and/or colloidal phase, even if the present study does not allow to conclude the prevalence of each. But dissolved phase is the phase in which micropollutants are bioavailable, as for inorganic the authoritative free-ion activity model (Morel, 1983) as for organic ones (Bosma et al., 1997; Leppänen et al., 2000, 2003, 2006; Kraaij et al., 2002, 2003; Kukkonen et al., 2004; Landrum et al., 2007; De Weert et al., 2008; Cui et al., 2013). Progressive contamination by As of the biota is a concern for the whole food web, firstly filtering species, and especially bivalves, which are eaten by local populations.

3.4. Recommendations for sampling strategy

During the present study, two sampling strategies were performed: sampling along the shoreline or sampling perpendicularly to shoreline in order to propose to environmental managers a way to collect beached *Sargassum* properly. Basing on Table A3 and focusing on As (total) value (*i.e.*: not the sum of the As species) because of the better quantification frequencies, metrological repeatability and lower standard deviation, blind sampling (*i.e.* whatever the position) leads to 81.6 ± 30.8 $\mu\text{g}/\text{kg dw}$ (standard deviation/mean: 37.8%), (1) grounded and at least partly dried and browned, being drifted to shore but not yet grounded (53 ± 30.3 $\mu\text{g}/\text{kg dw}$; standard deviation/mean 57.2%), (2) recently grounded, *i.e.* still wet (91.7 ± 29.3 $\mu\text{g}/\text{kg dw}$; standard deviation/mean/mean 32%), (3) close to be grounded, in the beating of the waves (92.2 ± 21.9 $\mu\text{g}/\text{kg dw}$ standard deviation/mean 23.7%), and (4) still in seawater (92.1 ± 17.7 $\mu\text{g}/\text{kg dw}$; standard deviation/mean 19.2%).

Considering such results, and regarding that sampling in condition (4) is the less ambiguous (floating, fresh and old gold colored algae), has the most homogenous As results and tends to reach the higher values, authors propose to environmental managers to sample them during their campaign. Considering chlordecone, 4-condition sampled *Sargassum* reach also the higher values.

4. Conclusions

The present article highlights that *Sargassum* reaches West Indian shores already contaminated by deep-sea As natural content and, in coastal seawater, is contaminated by chlordecone residues due to 70's-90's anthropic uses in French West Indies. Accumulation kinetics of chlordecone is rapid, algae reaching their *in situ* accumulation level in a day, but is in the range predicted by international literature. However, as for As and for chlordecone, pollutants are able to also leak and pollute the water column, drained out by

intense transudation of dissolved matters –an unexpected behavior for chlordecone. Thus, chlordecone pollution into *Sargassum* is more segregating polluted bays because chlordecone content in *Sargassum* decreases elsewhere but contaminated *Sargassum* mats can drift from polluted to chlordecone-free shores and could pollute them secondarily. It is also a severe concern because micropollutants in dissolved phase are bioavailable and could integrate easily the trophic web. Monitoring it needs a common sampling strategy and authors suggest to use the same protocol based on algae close to beach but not yet beached.

Declarations

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Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Authors' contribution

DAD performed the conception, preparation, sampling, experiment, the analysis of results and the writing of the present article. FM contributed to the preparation, experiment, analysis and writing. CM and FD contributed to the conception and sampling. JL, LD and APA contributed to sampling.

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Competing interests

The authors declare that they have no competing interests

Availability of data and materials

All data generated or analyzed during this study are included in this article (and its supplementary information files).

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Figures

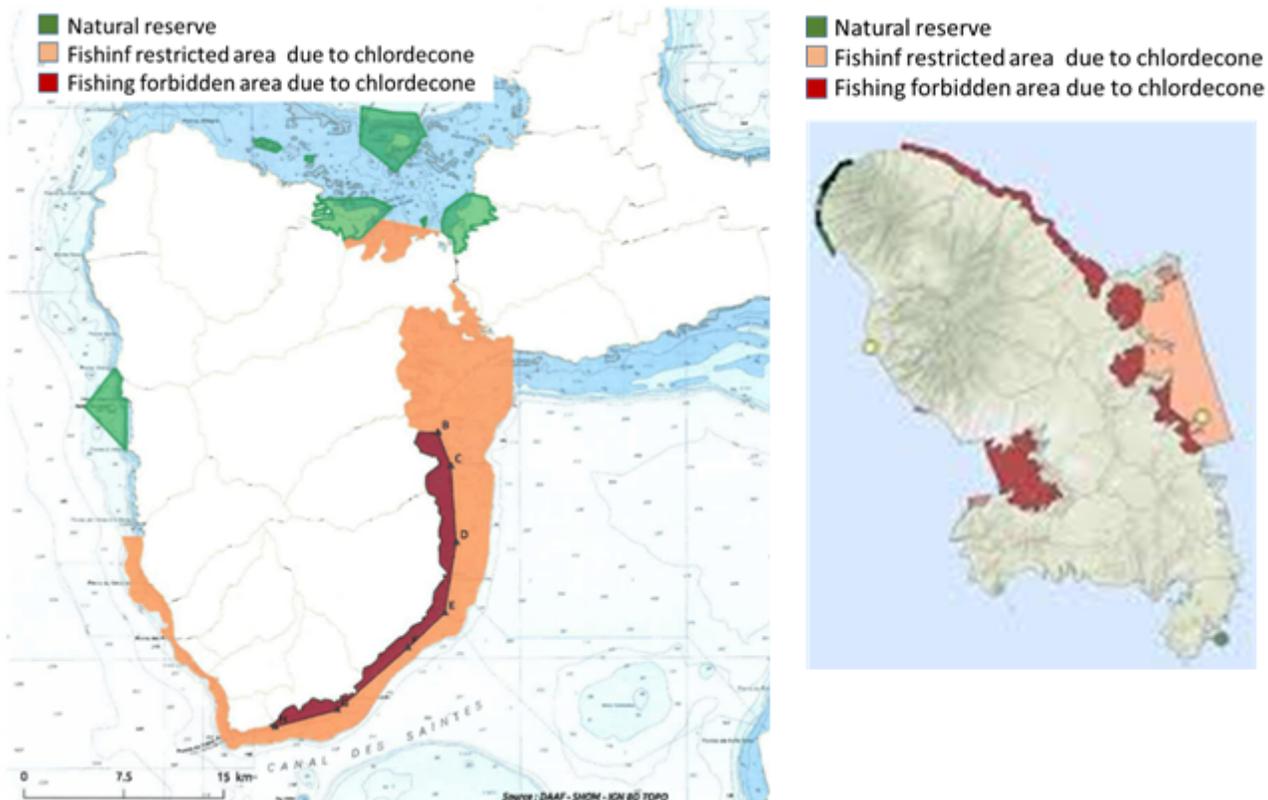


Figure 1

Maps of related Martinique and Guadeloupe (sources: Direction de la Mer). Because of putative contamination by chlordecone, some of the marine areas within which fishing is totally forbidden (red) or

restricted (purple) are presented.

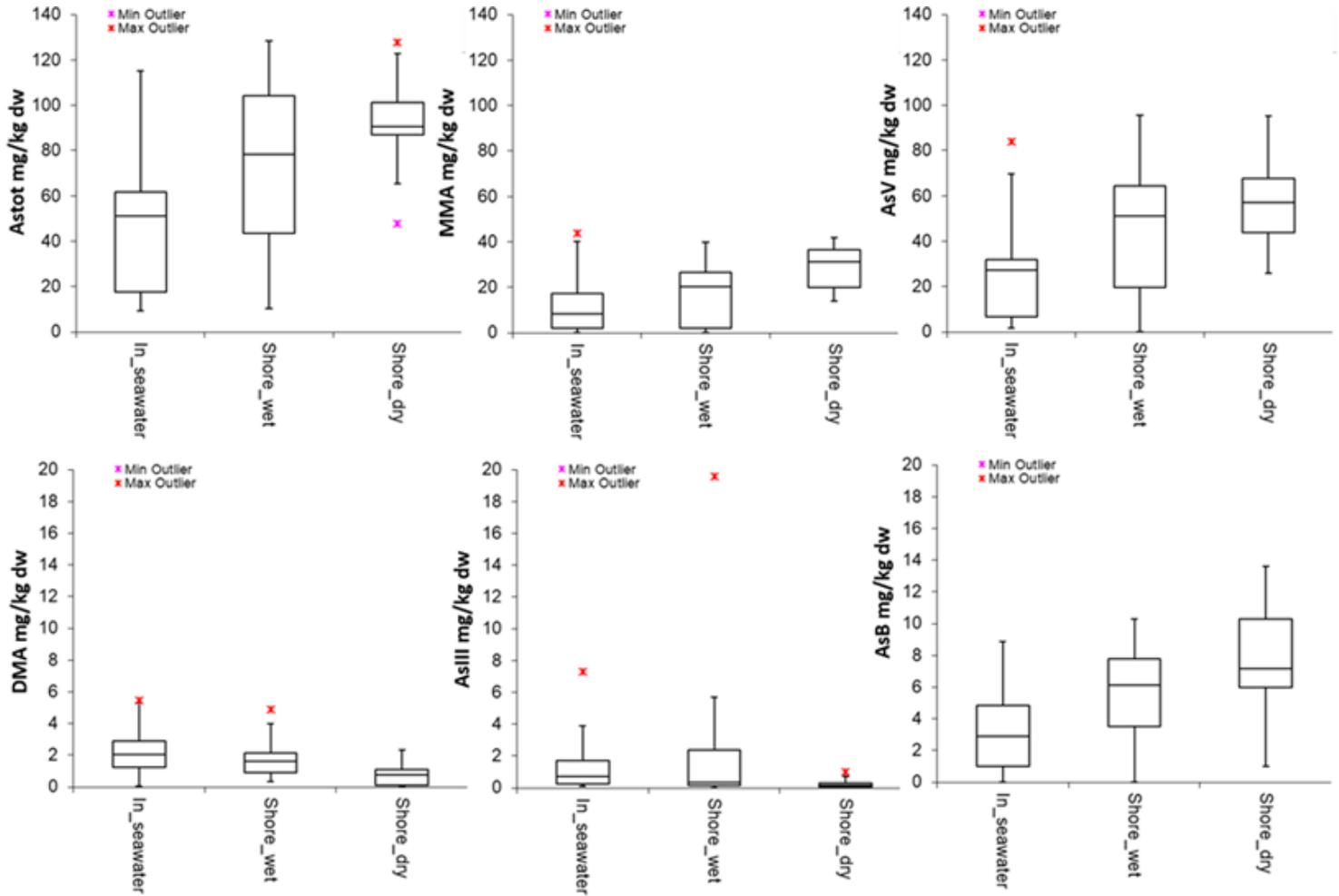


Figure 2

Concentration of arsenic and of the different arsenic species in the *Sargassum* samples according to their drifted status: still floating, fresh algae (In_seawater), recent deposit still humid, algae supple and not purplish (Shore_wet), and dated deposit dry even brittle, algae color turned purplish even greyed (Shore_dry).

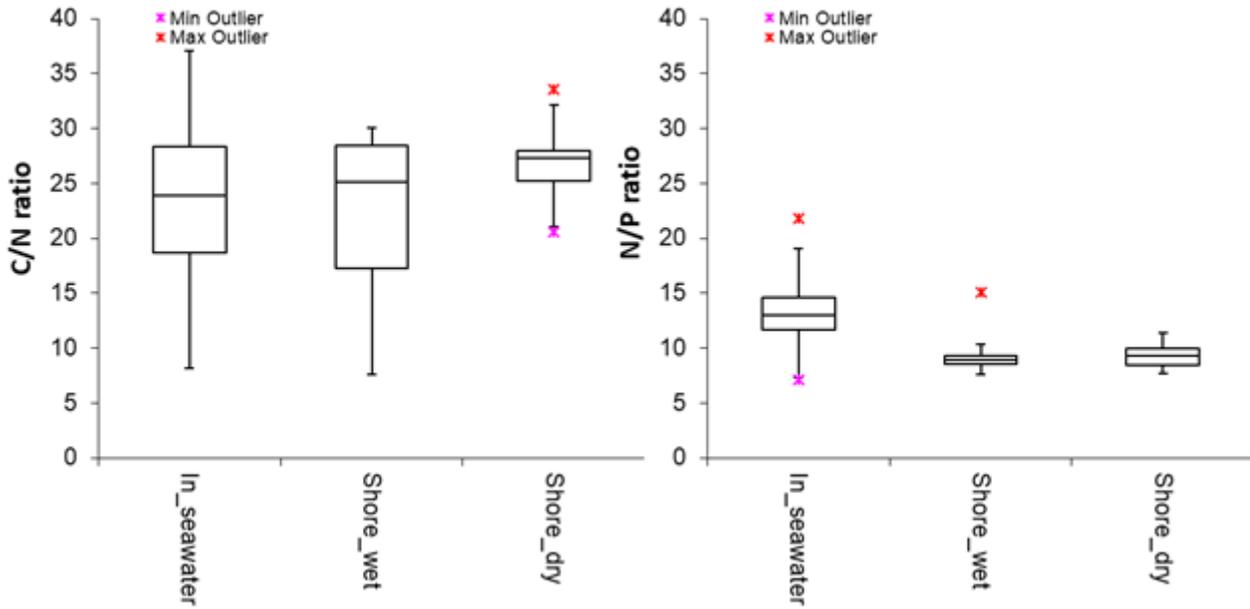


Figure 3

C/N and N/P ratio of the samples considering their drifted status: still floating, fresh algae (In_seawater), recent deposit still humid, algae supple and not purplish (Shore_wet), and dated deposit dry even brittle, algae color turned purplish even greyed (Shore_dry).

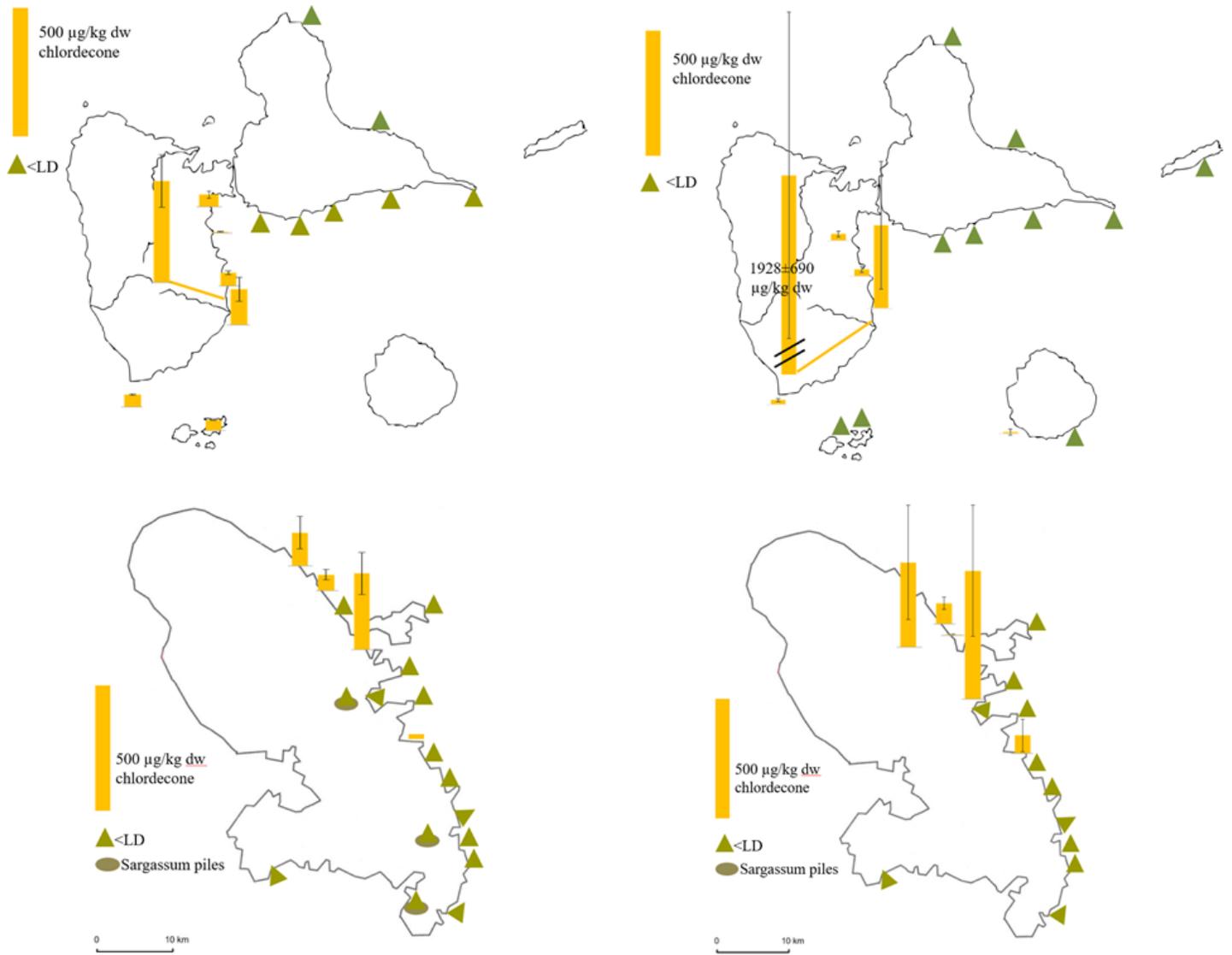


Figure 4

Chlordecone concentration observed in samples from the first (A) and the second (B) campaigns in Martinique and Guadeloupe.

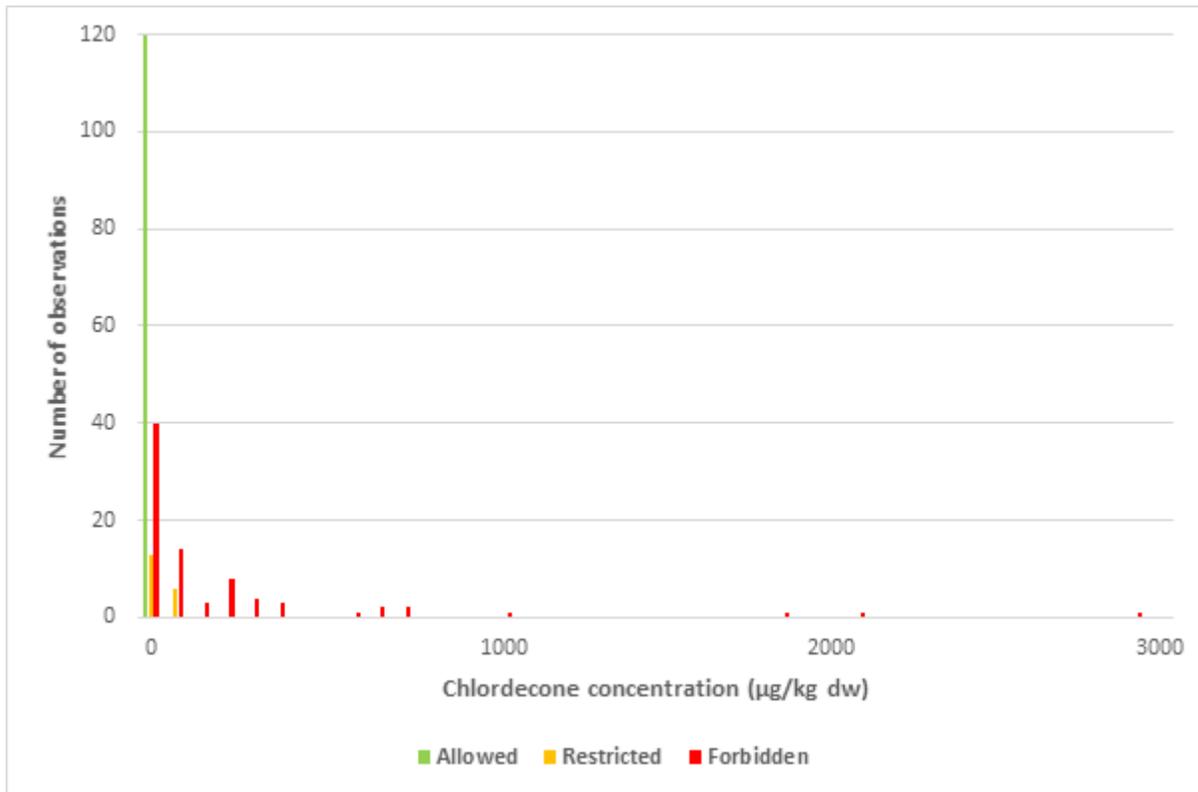


Figure 5

Distribution of the chlordecone concentration in the different official fishing areas. The legal status for fishing limitations in these areas was defined according to the chlordecone hazard level.

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- [Appendix1.docx](#)