

Review

The Red Seaweed Giant Gelidium (*Gelidium corneum*) for New Bio-Based Materials in a Circular Economy Framework

Teresa Mouga ^{1,*}  and Isabel Barreto Fernandes ² 

¹ MARE—Marine and Environmental Sciences Centre, CETEMARES Building, Polytechnic of Leiria, Av. Porto de Pesca, 2520-641 Peniche, Portugal

² LIDA—Research Laboratory in Design and Arts, Polytechnic of Leiria, Rua Isidoro Inácio Alves de Carvalho, 2500-321 Caldas da Rainha, Portugal; ibarreto@ipleiria.pt

* Correspondence: mougat@ipleiria.pt; Tel.: +351-262-783-607

Abstract: *Gelidium corneum* (Giant Gelidium or Atlantic agar) is a well-known red seaweed harvested for its high-quality agar content. Agar is a mixture of the polysaccharides used in the food industry as a gelling, thickener, clarifying, and stabilizer agent. The best agar quality is also used in the laboratory as bacteriological agar. Yet, in recent years, the species has been studied for many other applications. Examples of uses are pharmaceuticals, cosmetics, food supplements, bioremediation, biofuels, biofertilizers and biostimulants, biomaterials, and nanocrystals, among others. The use of this biomass, though, raises concerns about the sustainability of the resource, since this is not a cultivated species, being harvested in the wild. Thus, other uses of *G. corneum* biomass increase pressure on wild stocks already stressed due to climate change. However, in a biorefinery approach, a new trend is emerging, using waste biomass rather than harvested biomass to produce new bio-based materials. These are smart solutions that transform waste into innovative products, useful for various sectors of society while reducing the impact of biomass exploitation. The aim of this review paper, thus, is to address the current state of *G. corneum* biology, ecology, threats, its current uses and market, and the ongoing research on innovative proposals in a circular economy framework.

Keywords: harvested biomass; waste biomass; *Gelidium corneum* applications; biorefinery; circular design; bio-based materials



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1. Introduction

Seaweeds are marine macroscopic photosynthetic organisms classified into three taxa, according to their main accessory pigment: red seaweeds (Rhodophyta) present phycoerythrin, green seaweeds (Chlorophyta) contain chlorophyll *b*, and brown seaweeds (Phaeophyceae) exhibit fucoxanthin [1]. These organisms occur mainly in coastal areas, whether rocky or sandy shores, salt marshes, swamps, lagoons, estuaries, or coral reefs, but also in deep-water and open water floating communities, distributed in tropical, temperate, and polar regions. Zonation and community distribution depend on different physical and chemical parameters such as temperature, substrate, salinity, pH, nutrients availability, hydrodynamics, light, tides, wind, and pollution level. Biological factors such as epiphytism, herbivory, and disease also shape the distribution of seaweeds [2].

Seaweeds are known for their healthy primary and secondary metabolite content, such as a high protein content with a balanced essential amino acid content, low lipid content, and high fibre and mineral content. They also contain hydrocolloids, pigments, fatty acids, polyphenols, vitamins, minerals, and terpenoids, and other biomolecules [3–7]. These bioactive compounds show a beneficial effect on human health and well-being, such as antioxidant, antimicrobial, anti-cancer, anti-inflammatory, and antithrombotic effects, and immune-system improvement, among others [8–13]. However, besides human welfare, there are many new biotechnological seaweeds applications, including in the cosmetics

industry, as a moisturizer, anti-ageing, and UV protector [14–17], in agriculture, as a biostimulant or biofertilizer [18–21], and in bioremediation and biosorption, due to the seaweeds' ability to remove contaminants from the surrounding water [22–24]. Seaweed extracts are also gaining research interest as a bioplastic product due to the nature of their polymers, producing resistant durable biofilms [25,26]. Another current approach is the use of seaweed waste biomass to produce biofuels (biogas, methane, or bioethanol) since this process does not increase the atmospheric net CO₂ [27,28]. First-generation biofuels are derived from vegetable oil, starch, or sucrose, usually derived from crops. Second-generation biofuels are derived from lignocellulosic waste biomass. Third-generation biofuels are produced from algae. The second- and third-generation biofuels are much more land-use and environmentally efficient than first-generation biofuels [29,30]. These applications depend on the quantity and nature of the biomolecules present in algal biomass. Thus, one of the key challenges is to be able to efficiently extract these bioactive compounds. Making use of all the constituents of the seaweeds, while keeping their characteristics and bioactivities intact, requires sequential processing of all the material, and often the purification of the algal by-products [31]. The concept of a biorefinery has been proposed allowing the extraction and, therefore, use of all useful components of seaweed in a cascade process, cost-effectively, adopting a zero-waste approach while reducing the impact on climate change [32]. Furthermore, in a circular economy framework, any economic activity must have a positive impact on the environment, and the market encourages the recycling of products rather than extracting new resources. In this production and consumption model, all forms of waste are returned to the economy and/or used more efficiently, allowing the life cycle of products to be significantly extended [33]. Therefore, in a circular economy context, seaweed biomass waste should be processed into new products, and many authors are addressing this opportunity.

For many decades, one of the most industrially sought-after seaweeds was *Gelidium*. *Gelidium* sp. is a canopy-forming red seaweed (Rhodophyta) known for its high quality and content in agar [34–38]. Agar is a phycocolloid existing in the seaweed cell wall, consisting of a heterogeneous mixture of two polysaccharides, agarose, and agarpectin, the first with gelling properties and the second with thickening properties [39,40]. It is a semi-transparent, shiny, tasteless, odourless, and very hydrophilic colloid. Due to the formation of coiled helices, it forms very strong gels retaining water molecules when the agar solution is heated [41]. *Gelidium* sp. is the primary source of high-quality agar (high gelling strength and low sulphate content) and bacteriological grade agar, which is obtained only from this genus [2]. Although agar can be extracted from different species, such as *Pterocladia*, *Pterocliadiella*, *Ahnfeltia*, *Acanthopeltis*, and *Gelidiella*, the world agar market depends almost on wild-harvested *Gelidium* sp. and on cultivated *Gracilaria* sp., which produces a lower quality and lower price agar [42,43]. Currently, the most harvested agarophyte is the *Gelidium corneum*, being harvested in France, Italy, Portugal, Spain, and Morocco [44]. This wild harvest along with other human impacts, such as climate change, raises concern about the sustainability of the resource [45] and a global *Gelidium* landing shortage was recently diagnosed in 2018 [46]. Besides, the cultivation of *Gelidium*, although viable, did not reach enough yields to be economically profitable [47]. Hence, the management of the resource should be approached with attention.

Despite the importance of the agar market and the conservation concern, *Gelidium* sp. has been studied in the last two decades to evaluate other biomass properties such as antioxidant [48–50], antimicrobial [6,14,51,52], anti-inflammatory [53], antiproliferative or cytotoxic [54,55], biosorption of contaminants [56–63], and phyto-stimulant [64]. Besides, the use of agarophytic biomass has been proposed by several authors for biorefinery, including in the energy sector [65–67]. These properties disclose the potential use of *G. corneum* in several applications for which different types of biomasses may be used: harvested, stranded, and waste. The use of harvested biomass for other purposes than as an agarophyte competes with the agar industry and raises serious management and conservation concerns. Yet, the stranded biomass and waste may also be suitable to in-

corporate into new bio-based materials. The agar industry, e.g., produces annually many tonnes of residual *Gelidium* biomass, which are treated as waste [68] or are used in the fertilizer industry with a very low commercial value [6,69]. However, this waste biomass can have other uses, and, thus, a better valorisation, adding value to this biomass already used industrially, and diversifying its use. This may create new business opportunities for coastal populations, who are economically dependent on this valuable natural resource.

Independent researchers are revealing new smart and sustainable solutions using bio-based materials from *G. corneum*. Soon, these innovative bio-based materials, many using waste biomass, should be industrially applied for different purposes from a circular economy perspective. Thus, there is a growing interest in the use of *G. corneum* biomass beyond the agar industry, with emphasis on its residues, information that deserves to be compiled and discussed. This paper thereby covers the aspects of the biology, distribution, life cycle, harvesting, market, and applications of *G. corneum*, including innovative solutions that could constitute new industrial products in the future.

2. Methods

Papers indexed to Science Direct were analysed, using the following queries:

“*Gelidium corneum*”

“*Gelidium corneum*” AND “agar”

The former accepted name of *G. corneum* “*Gelidium sesquipedale*”

“*Gelidium sesquipedale*” AND “agar”

“*Gelidium corneum*” NOT “*Gelidium sesquipedale*”

All retrieved references were analysed, whenever available (Table 1). Many of these references were secondary citations of original research. Thus, they were not cited in this paper. Other low-impact references, not indexed on the Web of Science, are important regional references concerning the studied species. Therefore, Google Scholar was also analysed, for the same queries. Those available and that were found most relevant for the present study were analysed and cited.

Table 1. References retrieved from Science Direct and Google Scholar for *Gelidium corneum* and the synonym *Gelidium sesquipedale*.

References Retrieved	Science Direct	Google Scholar
<i>Gelidium corneum</i>	192	1320
<i>Gelidium corneum</i> AND Agar	98	584
<i>Gelidium sesquipedale</i>	222	1900
<i>Gelidium sesquipedale</i> AND Agar	123	963
Shared references <i>G. corneum</i> and <i>G. sesquipedale</i>	25	196
Total references	399	3220

FAO FishStatJ 2022 database [44] was used to gather data on the harvesting and cultivation of *Gelidium* sp. and other agarophytes (Supplementary Materials, Table S1).

Pre-set criteria:

1. Country.
2. Time frame: 1960 to 2020.
3. Production source: capture (harvest) or aquaculture production.
4. Species name: *Gelidium* seaweeds (*Gelidium* spp.), Giant *Gelidium* (*Gelidium corneum*), *Gracilaria* seaweeds (*Gracilaria* spp.), Warty *Gracilaria* (*Gracilaria gracilis*). The data on “Red Seaweeds” were added, for all the countries where *Gelidium* harvesting was not reported for all the period, and for which there were references of *Gelidium* harvesting; these include, e.g., Canada, France, India, Indonesia, Ireland, Japan, Mexico, Morocco, Portugal, and New Zealand. “Red seaweeds” from Spain, South Korea, and South Africa were also incorporated, whenever “red seaweeds” were counted in a one-time frame and “*Gelidium* seaweeds” in another. Other red seaweed species besides *Gelidium* spp. may have been included and, therefore, the data reported may be overestimated.

3. *Gelidium corneum* Biology, Distribution, and Ecology

Gelidium corneum common names: Atlantic agar, Giant Gelidium (English), Gelidium imperial (French), Ágar, Limo-encarnado, Cabelo-de-cão (Portuguese), Ocle, Caloca (Spanish) [70,71].

The Genus *Gelidium* includes, currently, 144 taxonomically accepted marine species [72] distributed worldwide. Among these species, *Gelidium corneum* (Hudson) J.V. Lamouroux (formerly *Gelidium sesquipedale*) is one of the best-known species. It is a cartilaginous dark-red seaweed (division Rhodophyta), with flattened branches with spoon-shaped branchlets, and creeping stolons at the base, up to 20 (30) centimetres tall, forming large tufts. Erect thalli grow from a system of creeping axes attached to rocky substrates through rhizoids (Figure 1) [69,70,73–75].



Figure 1. Macroscopic image of *Gelidium corneum* collected in Centre Portugal.

Gelidium is a clonal-modular seaweed [2]; consequently, it spreads laterally and vegetatively over the soil surface via creeping axes. It also produces erect thalli (fronds). Storms and grazing remove the fronds but not the creeping axes, which remain attached to the substrate. Regeneration and growth of erect fronds from the creeping axes are common and fast [76]. An interesting feature of *Gelidium* populations is the ability to grow from vegetative reproduction. These erect fronds can have an autonomous life when fragmented and can reattach to the substrate, so vegetative propagation, through fragmentation, is a frequent method of colonisation [76]. The species has a wide distribution, occurring in Atlantic Europe [6,73,77–82], Mediterranean Sea [83], Atlantic islands [84–86], Atlantic Africa [87–90], and Atlantic America [91]. Guiry also mentions populations in the Indic and Pacific oceans, namely in India, Indonesia, Korea, Vietnam, and Australia, but no published information on these locations could be found [72] (Figure 2).

G. corneum grows in temperate to tropical areas, with seasonal temperatures ranging between 10 and 25 °C, in partly shaded habitats, with strong tides and sea currents. In Europe, *G. corneum* forms widespread beds, usually subtidal zones up to about 25 m in depth. As to the substrate, the species prefers growing on slightly sloping regular bedrock, with little to moderate sand sedimentation [77,92]. In these temperate waters, *Gelidium* species reach high abundances and frequencies. Regardless of the species or the latitude, these are slow-growing organisms, up to 100 mm y⁻¹ [76,93,94]. *G. corneum* is a canopy-forming seaweed, which is to say, it is a habitat-forming seaweed, creating a stable and complex community providing food, shelter, nursery, and habitat for many other species, such as invertebrates, fish, and other smaller algae [95,96]. *G. corneum* is sensitive to environmental parameters such as temperature, light, nutrients, and water movement [97]. Santelices [76] also states several biological factors affecting productivity,

comprising morphology, age of the fronds, thallus part, reproductive state, seasonality, crop density, life history phase, and geographic and ecological origin of the species. Additional events of importance affecting *Gelidium* populations include extreme low tides, storms, and grazing [36,76].

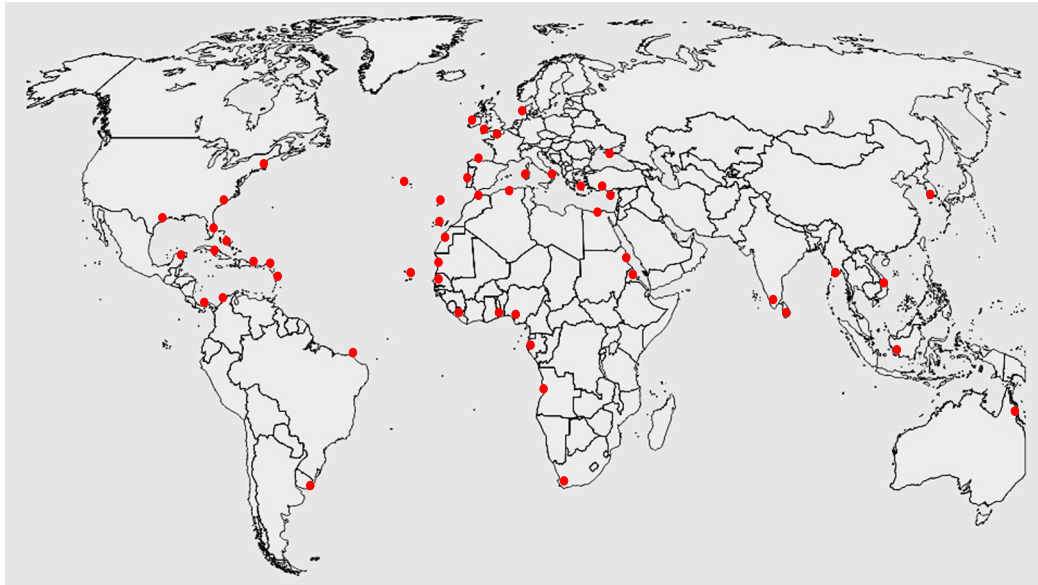


Figure 2. World distribution of *Gelidium corneum* (data from Guiry [72]).

In many areas of the globe, the cumulative impacts of human pressure, such as habitat destruction, pollution, over-harvesting, invasive species, and ocean warming, decrease the resilience of the seaweeds and promote the loss of the seaweed's biomass. *G. corneum* is no exception, showing a decline in the past decades. In response to these disruptions, shifts in the distribution patterns of canopy species occur. Notably, these shifts are observed with a decline in canopy-forming species, the increase in morphologically simpler warm-water species and coralline algae, and the progressive introduction and expansion of non-indigenous species [45,78,79,98–105].

4. *Gelidium corneum* Life Cycle

The genus has a complex life cycle, representative of the most evolved red seaweeds (class Florideophyceae), with a triphasic isomorphic life cycle [74]. This life cycle is portrayed by a haploid independent gametophyte, producing gametes through mitosis (either male spermatangia or female carpogonia). The male gametes (spermatia) are released and pass to the trichogyne of the female gamete (carpogonium) where fertilization occurs (Figure 3). The mitotic division of the zygote produces gonimoblast filaments, the first diploid generation (carposporophyte phase), which grow within the female gametophytes. These carposporophytes produce carposporangia inside which diploid carpospores are formed by mitosis. Carpospores are released and each matures into an independent diploid tetrasporophyte, the second diploid generation. The tetrasporangial mother cell divides by meiosis to produce four haploid tetraspores, each becoming a new gametophyte [106]. Although isomorphic, the tetrasporangial are more common than the gametangial thalli. This finding suggests that tetrasporophytes are more robust and competitive and the gametophytes are more sensitive to environmental conditions and less viable [76].

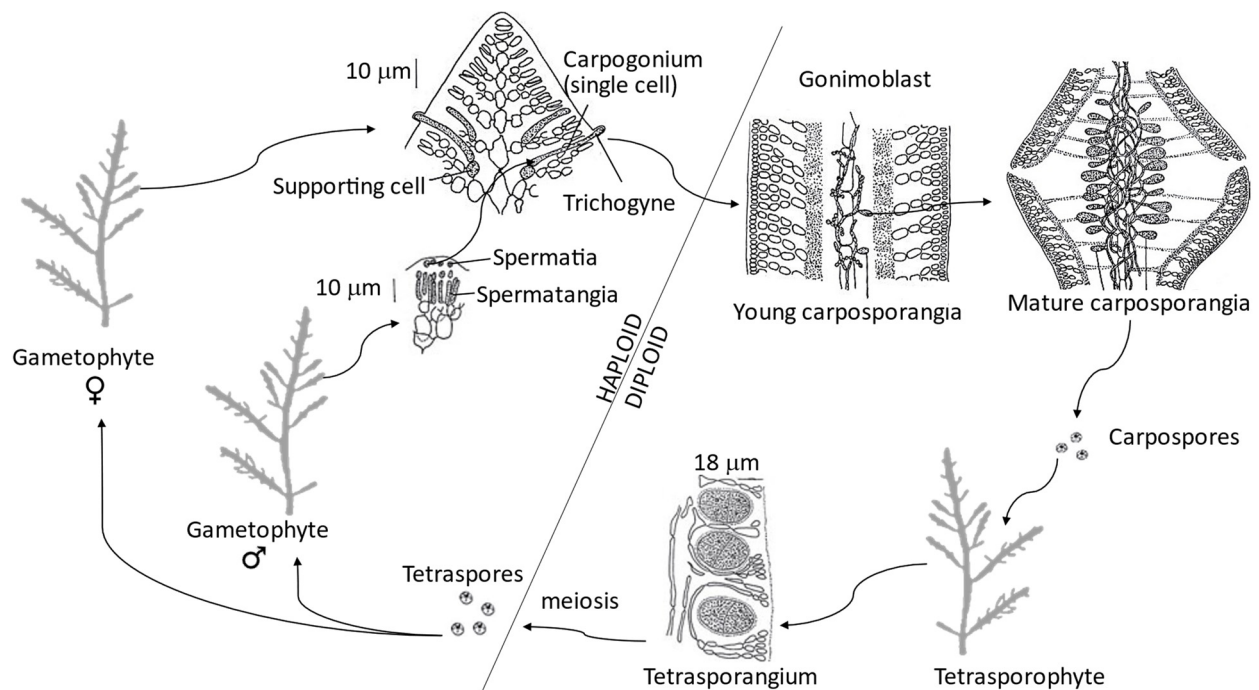


Figure 3. Triphasic isomorphic haplodiploid life cycle of *Gelidium corneum*, adapted from Lee [106].

The species is reported to have a certain inability to attach itself to a substrate through spore colonisation. So, sexual reproduction is often replaced by vegetative reproduction, in which the creeping thalli produce a disk of rhizoids that can attach and penetrate suitable substrates [107].

5. *Gelidium* Harvesting

Different species of *Gelidium* are harvested in the world to produce agar. *G. corneum* is harvested in Portugal, Spain, France, and Morocco, *G. amansii* and *G. latifolium* in Japan, Taiwan, Korea, and Indonesia, and *G. robustum* in Mexico. Smaller quantities of *G. linguatum*, *G. chilense*, and *G. rex* are harvested in Chile, whereas *G. pristoides*, *G. abbotiorum*, *G. pteridifolium*, *G. capense* are harvested in China, Namibia, and South Africa [46,92,108,109]. The biomass may be gathered from beach-cast seaweeds, using nets or rakes, a method common in countries such as South Africa, Australia, and New Zealand [110]. The attached thalli may also be cut by hand. More recently, seaweeds are harvested mechanically with the biomass being plucked off by divers, stowing the seaweeds in bags or baskets which are then lifted onto a boat [95,111]. When the creeping axes are removed, regeneration of the vertical thalli may take several years to regenerate, allowing the area to be invaded by other algae [94]. Harvesting, therefore, has a direct impact on the biomass and structure of seaweed beds and marine biodiversity. Harvesting canopy-forming seaweeds affects the morphology, canopy structure, thalli growth and regeneration, standing stock, and species composition of the foundation species. In turn, these changes affect the ecological roles of the canopy-forming seaweeds in marine ecosystems [110].

No doubt wild seaweed harvesting is facing the challenge of balancing the socio-economic and environmental sustainability of the activity. The impact on the wild stocks depends on the methods used, the mechanical clear-cutting being more severe than the hand-harvesting. For *G. corneum*, the creeping taxa must be left unharmed, allowing *Gelidium* populations to recover rapidly [94]. Over the past decades, management of natural resources has tackled the need to protect species and ecosystems, enabling habitat protection to sustain species diversity and abundance, whilst simultaneously granting sustainable exploitation of the marine resources. There is a need to evaluate permanent stocks of *Gelidium* where they are still harvested, to define harvest effort, to study the

possibility of restocking, to establish and respect the harvest season, and to apply local resource management regulations [110].

6. Gelidium Cultivation

The demand for algal biomass for industrial purposes has far outstripped the capacity that traditional harvesting of wild stocks can provide. Trying to meet the demand, different attempts to cultivate *Gelidium* species have been performed for the past decades with some biological success but without economical relevance due to the slow growth of the genus [93]. These attempts were carried out in the laboratory, inland (tanks or ponds), or in the sea (net bag method, rafts, and net pouch), forming spores, fragments, and grown from reattachment thalli [74,107,112–119]. Different cultivation conditions were tested, assessing the influence of temperature, season, irradiance, nutrients, and water movement productivity of the biomass [94,117,120–123]. Although the experiments succeeded to a greater or lesser extent, they resulted in low yields. Yields vary among species, but cultivation points to a maximum yield of 25 kg FW m⁻², which cannot compete economically with wild harvesting [47]. The only species that has been industrially produced is *G. amansii* in ponds, recorded for North Korea, but with very little data available [46,124].

Despite these setbacks, *G. corneum* is an important biological resource with multiple applications, so the feedstock cannot depend exclusively on wild resources. Further scientific research is, therefore, required to develop more efficient cultivation techniques that will produce abundant quality biomass at a competitive price compared to wild biomass. Therefore, to introduce economically profitable cultivation of *Gelidium*, it will be required to work on genetic improvement of the genus, through the selection of the more productive strains or genetic engineering [47]. Genetic engineering, together with further studies on cultivation and reproduction techniques, will hopefully allow full domestication of *Gelidium*, as has already been achieved for other seaweed species in the past years.

7. Gelidium Market

The *Gelidium* market is largely agar extraction and selling. Marketable *Gelidium* biomass primarily relies on wild-harvested specimens (Figure 4). Until 1967, *Gelidium* harvesting was an increasing activity on four continents (America, Africa, Europe, and Asia), with a maximum of 276,409 tonnes of fresh weight biomass being brought onto the market. Then a decrease occurred, until 1977, with mainly Africa and Europe showing a reduction in harvesting. Between 1978 and 1996, Asia significantly increased the harvesting of red seaweeds, with Indonesia being the main harvester. All the other countries, from Europe, America, and Africa, considerably reduced *Gelidium* harvesting. Between 1989 and 2010, FAO also recorded a small amount of *Gelidium amansii* produced in aquaculture in North Korea (about 1000 tonnes y⁻¹). Since 1998, Asian countries have reduced harvests, joined by Europe and America. Contrary to this trend, African countries have recovered the *Gelidium* market. Currently, the main player in the market is Morocco, which had over 64% of the world's share with over 22,218 tonnes of *Gelidium* harvested in 2020 (FAO, 2022b).

Due to the increasing difficulty in providing enough wild *Gelidium* for the food industry, the use of wild *Gelidium* is progressively being replaced by other agarophytes. The most widely used seaweed belongs to the genus *Gracilaria*, the only agarophyte cultivated worldwide, especially since 2002 (Figure 5).

Although the agar produced by *Gracilaria* spp. is of inferior quality and cannot be used as bacteriological agar, *Gracilaria* agar is profusely used in the food industry [125]. *Gelidium*, hence, is used almost exclusively for the bacteriological agar industry.

The *Gelidium* agar market, hence, has severely declined from over 92% in 1967, with more than 276 thousand tonnes harvested, to less than 1% in 2020 (34,500 tonnes), while cultivated *Gracilaria* agar has increased remarkably from less than 1% in 1977 (1741 tonnes) to over 98% in 2020 with more than 5 million tonnes. Harvested *Gracilaria* currently has a minor contribution to the market, with around 54 thousand tonnes (Figure 6).

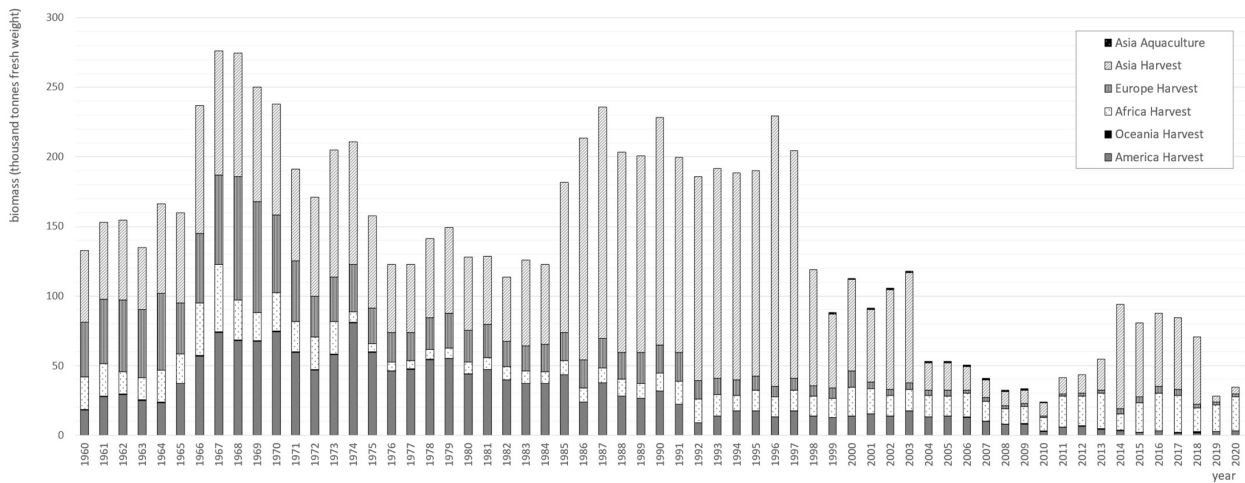


Figure 4. Wild harvested red seaweeds (*Gelidium* sp., *Gracilaria* and unspecified species) and cultivated *Gelidium amansii* marketed between 1960 and 2020 (in thousand tonnes of fresh weight). Data from FAO [44].

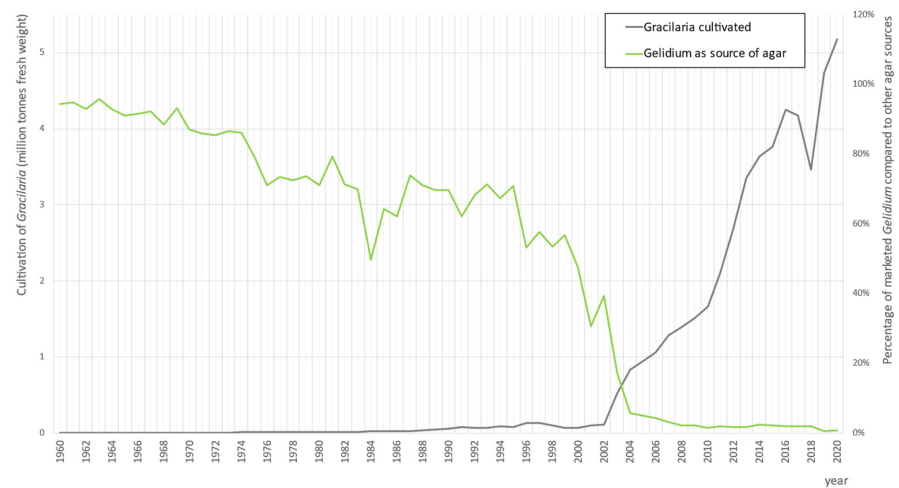


Figure 5. Percentage use of *Gelidium* as agar-bearing biomass source (green line) compared to the cultivation of *Gracilaria* (grey line), between 1960 and 2020. Data from FAO [44].

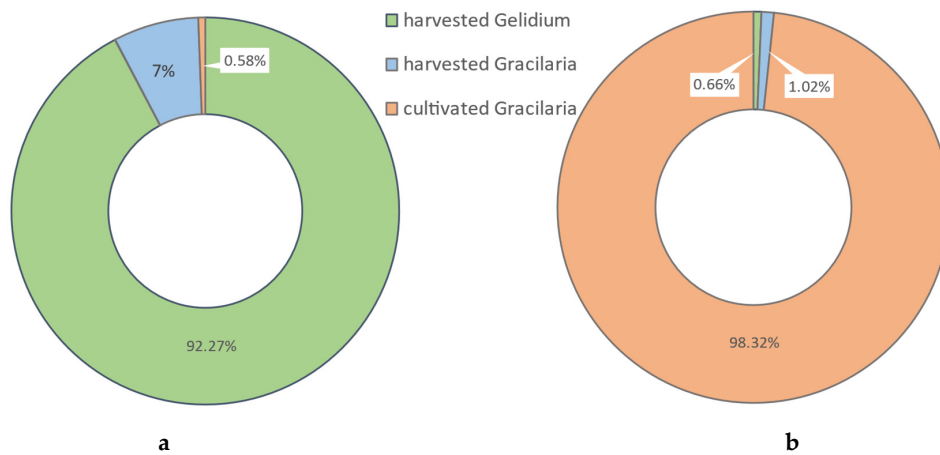


Figure 6. Percentage of agar market use in 1996 (a) and 2020 (b) of wild *Gelidium* biomass, wild *Gracilaria* biomass, and cultivated *Gracilaria* biomass. Cultivated *Gelidium* biomass was not in the market in these years. Data from FAO [44].

8. Innovative Uses for *Gelidium corneum*

As stated, *Gelidium corneum* biomass is sought after for its use in the agar industry. This industry produces tons of macroalgae waste annually, which is currently disposed of or used in the fertiliser industry [53,68,126], resulting in a loss of valuable bioactive compounds. This biomass, therefore, may have other uses and certainly deserves a better valorisation. In recent decades, the harvested biomass, extracts, and residues of *G. corneum* have been studied for other applications, with interesting results. Examples are the food industry, pharmaceuticals, agriculture, biofuel, biomaterials, and bioremediation, among others. Table 2 reviews relevant published papers seeking biomass valorisation.

Table 2. Properties and applications of *Gelidium corneum* biomass, extracts, and waste.

Type of Resource	Properties and Applications	References
<i>Gelidium corneum</i> wild biomass or extracts applications		
Raw biomass chemical composition		
Raw biomass elements' bioavailability		
Raw dry biomass and storage	Agar industry	[26,34,35,37,38,127–136]
Raw biomass yield improvement		
Raw biomass Process optimization		
EtOH/Aq extracts		
Chf extracts	Antioxidant	[48,49,54,137–139]
MAA		
MeOH extracts		
DCM-MeOH extracts	Antimicrobial	[6,14,51,140]
DCM-EtOH extracts		
Ag nanoparticles	Antimicrobial	[52]
	Antifouling	[53]
EtOH extracts	anti-inflammatory	[53]
Aq extracts		
DCM-methanol extracts	Anti-proliferative	[53–55]
MeOH/Aq extract	Cytotoxicity	[53–55]
Elemental analysis	Food, food supplement	[69]
Waste biomass hydrolysates	Source of biochemicals for biomaterials, biofuels, and fine chemicals, such as poly-3 hydroxybutyrate and D-tagatose	[141–143]
Biomass mechano-enzymatic deconstruction of sugar and bioethanol	Biofuels	[144]
Agarose and agar-based matrices	Probiotics encapsulation	[145]
Oligosaccharides	Phyto-stimulant	[64]
Agar-based hydrogels and aerogels		
Enzyme immobilization in nanoflowers for lactose breakdown	UV protection	[50]
Agar, agar-gelatines	Antimicrobial/Antioxidant Edible	
Polysaccharides, fibres	biofilms for food packaging	[146–157]
Nanocellulose biofilms	Biodegradable biocomposites	
Agar/clay nanocomposite films	Biodegradable packaging	[153,158]
Heated mucilaginous carbohydrates	Paper	[159–161]
Biomass fermentation for ethanol production	Biofuel	[159,162]
Cellulosic ethanol		
<i>Gelidium corneum</i> waste biomass applications		
Waste chemical characterization	Antioxidant	[48]
Cellulose nanocrystals	Polymer composites	[163]
	Bioplastics	
Biodegradable biofilms	Bioplastics for packaging	[164–167]
	Biofuel	
Biochar	Adsorbent	[168–172]

Table 2. Cont.

Type of Resource	Properties and Applications	References
Full waste biomass	Antifungal soap for cosmetics	[173]
Full waste biomass	Biofertilizer	[126]
Full waste biomass	Bioremediation Biosorption	[56–59,61–63,174–179]

Abbreviations: DCM—Dichloromethane, MeOH—methanol, EtOH—Ethanol, Aq—Aqueous, Chf —chloroform, MAA—mycosporine-like amino acids, Ag—silver.

8.1. Primary Metabolites

Gelidium corneum is known to have a high content of polysaccharides, very low content of lipids, and a fairly high content of protein [180,181]. Further, a close analysis of *G. corneum* shows that there are significant seasonal variations in the moisture, ash, protein, lipid, and carbohydrate content of the harvested wild biomass [6]. This biomass seems to have interesting nutritional properties that should not be overlooked as food, feed supplement, or feed. Once the shortage of the resource is fully addressed, through efficient management or cultivation, *G. corneum* biomass may become an important source of nutritious and healthy food.

Remarkably, agar waste biomass maintains most of the chemical properties, namely protein and carbohydrate content, and, as Faraj noted, amino acids [180]. The protein, lipid, and carbohydrate content, as well as the antioxidant capacity of the residue obtained from the agar industry (provided by IberAgar Lda.), were also analysed. The antioxidant capacity was measured using DPPH (1,1-diphenyl-2-picrylhydrazyl radical scavenging assay) or ABTS (2,2'-azino-bis (3-ethylbenzothiazoline-6-sulfonic acid) diammonium radical scavenging assay). The methodologies used to measure protein, lipid, and carbohydrate content, as well as DPPH and ABTS assays, were the same as those used in our previous work [6]. The results are shown in Table 3.

Table 3. Comparative analysis of wild-harvested and agar residue of *Gelidium corneum*, regarding proximate analysis and antioxidant capacity (data from [6] and personal unpublished data).

<i>Gelidium corneum</i> Chemical Composition and Antioxidant Activity	Wild Harvested Biomass	Agar Industry Residue
Proximate Analysis (% dry weight)		
Protein	14.19 ± 0.33	20.59 ± 0.79
Lipids	2.10 ± 0.11	0.95 ± 0.04
Carbohydrates	33.30 ± 1.25	36.40 ± 3.96
Antioxidant Analysis (% Inhibition)		
ABTS	12.31 ± 1.39	9.79 ± 0.15
DPPH	8.50 ± 1.32	7.53 ± 0.51

The amount of carbohydrates obtained in the residue is smaller than that obtained by Tuma et al. [141], who obtained a value of 44.8%, but similar to that obtained by Trigueros et al. (37%) [182]. As these authors underline, the main carbohydrate present is cellulose, which is not removed by the agar extraction process, and, thus, can be applied in other industries, in a biorefinery process. The low amount of lipids is also consistent with the findings of these authors. The protein content remained high, meaning that the process of agar extraction used did not affect these primary metabolites, as Trigueros et al. [139,182] also obtained. This result is not surprising since the residue biomass used by Trigueros et al. and the residue biomass currently studied were obtained from Hispanagar and Iberagar, respectively, both from the same group. Thus, the agar extraction process was probably the same. Conversely, the biomass analysed by Tuma et al. [141] obtained much lower protein values (0.68%), indicating that the pre-treatment carried out severely damaged the proteins.

As to the antioxidant capacity, *G. corneum* is considered a species with low antioxidant capacity, and the values in our previous study for wild harvested biomass are within the range found by Matos et al. [53]. As to the agar residue, although there was a decrease, there is still a noticeable antioxidant capacity. The same finding was also obtained for total phenolic content by Trigueros et al. [139]. Therefore, depending on previous extraction processes performed during the industrial processing, a cascade biorefinery may be applied granting the full use of the biomass and delivering a broad range of bio-based products.

8.2. UV Protection

Gelidium corneum biomass presents other interesting compounds that deserve further scrutiny. An example is the presence of mycosporine-like amino acids (MAAs) shinorine, P334, palythine, asterina, and palythiol [16,183]. These compounds absorb ultraviolet radiation and have been in the spotlight in recent years due to their photoprotective role. MAAs disperse the harmful UV radiation by converting it into heat energy and dissipating it into the surroundings within the cell [184]. MAAs also have a strong antioxidant capacity, by quenching reactive oxygen species [137]. Thus, the growing importance of these compounds is increasing as the incidence of skin cancer rises due to overexposure to UV radiation [185]. The direct application of algae to obtain derivatives of organic and inorganic compounds that act as UV radiation blockers has been the subject of several research works, mostly to be used in cosmetic applications as sunscreen [138,184,186]. The incorporation of UV-protective ZnO nanoparticles into cotton, wool, or polyester fabric to produce clothing is another trend [187–189]. Less frequently, algal extracts or algal biomass have been studied to be applied in the textile industry, incorporated into fabrics that, therefore, confer UV protection to them [190,191].

Work from the University of Minho (2C2T—Textile Science and Technology Centre, unpublished work), in Portugal, used ZnO nanoparticles with *G. corneum* extracts on cotton and on polyester, to test the effectiveness of seaweed as a UV protector. In brief, ZnO nanoparticles were prepared as follows: Zinc nitrate was dissolved in distilled water and added to freeze-dried seaweed extract (25:1). The mixture was heated and after cooling, a sodium hydroxide solution (25%) was added. The supernatant containing the ZnO nanoparticles was applied to the textile substrates. The UV radiation blocking of the initial untreated textile substrate sample and the cotton and polyester samples treated with the nanoparticle solution was measured by the percentage transmittance in a reflectance spectrophotometer (Datacolor 550) at 200–400 nm, the UV region. Figure 7a,b show the transmittance results for cotton and polyester, respectively. The effectiveness of blocking UV radiation in cotton and polyester samples treated with the nanoparticle solution is visible in Figure 7a for cotton and Figure 7b for polyester. Therefore, this technique allowed obtaining textile substrates of interest for the manufacture of outdoor clothing. The use of this feature in technical textiles is also of great interest, as it avoids the ageing of materials, either by loss of mechanical properties or aesthetic properties, increasing their life cycle.

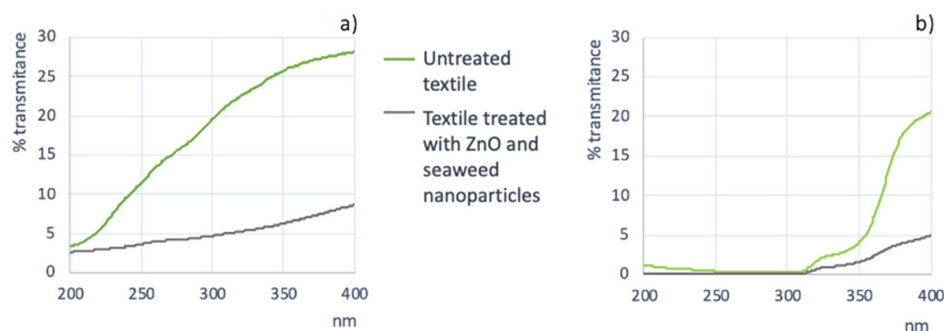


Figure 7. Percentage of transmittance spectra of the textiles tested: (a) cotton; (b) polyester. The green lines represent the control (untreated) textiles, and the grey lines represent the textiles treated by coating with the zinc oxide nanoparticles solution with *Gelidium corneum* extracts.

As follows, UV-treated fabrics with algae extract provide high UV protection and can be used to produce garments and accessories for individuals with sensitive skin (babies, children) or for individuals with special needs who have photosensitive skin reactions. Certainty shortly, new products with this concept of bio-based solutions may come to the market.

8.3. Biofertilizer and Biostimulant

Another area of growing interest is in the agricultural field, notably the harnessing of low-quality biomass that is unsuitable for medical, pharmaceutical, or food applications. Seaweeds are excellent plant biostimulants due to their richness in compounds that promote plant growth and yield, seed germination, root development, and resistance to abiotic stresses [20,21,192–194]. The biodegradability of seaweed, its non-polluting nature, and the absence of toxic effects on humans and animals are important factors contributing to the interest in this waste as a biofertilizer or biostimulant. The biofertilizing effect, using the complete seaweed biomass, both macronutrients and micronutrients, helps foster the growth of plants and fruits. The key nutrients held in the seaweed biomass valuable for crops are polysaccharides, such as starch and cellulose, as well as other primary metabolites, such as proteins and lipids [195]. Biostimulants are substances or materials, other than nutrients and pesticides, applied in small amounts (including seaweed extracts) to seeds or plants, having the potential to modulate plant physiological processes and enhance their growth, development, or stress response [196]. These compounds also boost plants' tolerance to abiotic stresses, such as salt, drought, and heat. Biostimulant compounds are often secondary metabolites, such as vitamins, phytohormones (auxins, cytokinins, gibberellins, abscisic acid, and ethylene), and other growth promoters (e.g., betaines) and elicitors (e.g., peptides, glycoproteins, fatty acids, oligosaccharides, phycocolloids), which prompt defence response [197–199]. Seaweed biomass, either full or extracted, is accordingly an interesting alternative to commercially available chemicals, which can be used for agricultural purposes by enhancing growth, while ameliorating stress and disease response.

Among the possible biostimulant applications, a bio-based material has been developed, using seaweed harvested biomass fibres and seaweed waste fibres. Small pots were manufactured, in which lettuces (*Lactuca sativa*) were cultivated. The experiment was conducted for 22 days at 21 ± 1 °C, and the pots were filled with commercial garden soil. Plastic cups were used as controls. By the end of the experiment, the fresh weight was measured (Figure 8). The pot with algal waste allowed the best vegetative development of the leafy plants (lettuce). In comparison, the pots with biological waste-based material showed slightly lighter plants. In both cases, the fibrous pots were penetrated by the plant roots and cracked after 10 days. In contrast, the plastic pots yielded the smallest and the most lightweight plants. Although not statistically significant (one-way ANOVA, p value = 0.470), it seems that the pots formed by the algal fibres, both harvested biomass and waste, exhibit some biostimulant activity. This will be the first approach to the production of biological materials from seaweed waste applied to agriculture, through the production of biodegradable pots with biostimulant properties. The results achieved, although very preliminary, provide us with some guidelines for the uses of this biomass in the agricultural sector.

The market target for the waste fibres, therefore, is plant nurseries requiring small containers for seedlings, which are introduced into the soil directly. Being biodegradable and biostimulating, the pots not only boost the plant's growth but also enhance the qualities of the soil. Further studies are, however, required to validate this concept, namely by comparing it with other alternatives such as coconut pots. But this is a promising solution that decreases waste and reduces the agricultural pollution load whilst increasing its efficiency.

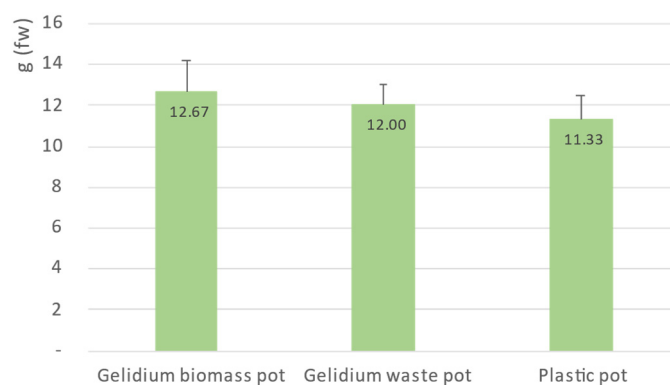


Figure 8. Fresh weight (in g) of *Lactuca sativa* (lettuce) grown in pots made with *Gelidium* biomass fibres, *Gelidium* waste fibres, and plastic pots, for 22 days (n = 3).

8.4. Production of Biochar

Biochar is the solid product of pyrolysis of dried algal biomass, under oxygen-restricted conditions at a given heating rate [200]. Biochar from varied sources has been appraised for its ability to sequester carbon and improve soil fertility. Seaweeds are among the biomasses screened to produce biochar for soil improvement and bioremediation, with highly promising results. These are attributed to the low carbon content of the biochar produced and the high yield of nitrogen, phosphorus, and potassium, as well as exchangeable ions, and its ability to remove different contaminants from soil [22,201]. In addition to these interesting applications, biochar produced from agriculture waste has also been tested for the construction industry, namely being combined in cementitious applications as a CO₂ sequester [202]. The use of biochar in the cementation material diminishes the greenhouse gases that the agricultural waste would produce when disposed of. Conversely, biochar processing into biochar enables the sequestration of CO₂. Ferrera-Lorenzo and co-workers [169,172] developed activated carbons from *G. corneum* agar industrial waste (algal biomass and carbonized biomass) to use as biosorbents. The authors proved that the high content in carbon and nitrogen of the algal biomass renders it highly suitable for the preparation of activated carbons, exhibiting very good CO₂ and CH₄ capture capacities. In a world with rising carbon emissions, this application is also encouraging, moreover when waste can be turned into a recycling product.

8.5. Biosorption Capacity

The biosorption of metal ions using different biomasses has been reported to be an alternative technology to conventional metal effluent treatment. Between 2007 and 2009, Vilar and collaborators published several papers on the metal ion biosorption capacity of *Gelidium corneum* [56–63,175,176]. The authors demonstrated that *G. corneum* biomass and a granulated algal residue exhibited the ability to effectively remove metal ions under different experimental conditions. Although the results obtained are noteworthy, no further developments could be found in this regard.

8.6. Biomaterials for Packaging and Coatings

One of the world's major concerns not yet addressed is petroleum-based plastic pollution. A possible innovative solution to tackle this problem is the development of bioplastics. The bio-based materials are made from natural resources, biodegradable, and non-toxic. Their production also reduces the waste generated, decreases fossil fuel consumption, and the number of greenhouse gases emitted. Several organisms have been studied to develop bioplastics. Due to the ability to produce biofilms, seaweeds are among them with *Gelidium* as one of the most popular [165]. As stated, red algae are composed of a high concentration of polysaccharides, many of which are fibres (cellulose) and phycocolloids, which in the case of *G. corneum* is agar. These natural polysaccharides are valuable to the plastic industries as

they contain carbon that can be converted into biofilms [203]. Polyhydroxyalkanoates are among the most encouraging potential bioplastics that can be obtained by fermentation, using *Gelidium* biomass as feedstock [149]. Biofilms from these polysaccharides usually have good mechanical strength, moderate gas barrier properties, and, most importantly, are edible and easily degradable in most cases. Edible food packages have also been tested for their antioxidant and antimicrobial properties [35,155,157]. The development of packaging with these properties expands the spectrum of uses of these biomaterials, as they increase the shelf life and the quality of the food products. Yet, the quality of the biofilm seems to vary with the physiological state of the algae, and it is very hydrophilic and brittle, which are some negative features [204]. To improve the biofilm quality and decrease the impact of production, green methods have been applied for seaweed bioplastics, developing better, cheaper, and more eco-friendly options [205]. Red seaweed fibres are also suitable for paper or board productions. However, because these fibres are immersed in a phycocolloid matrix, they need to be removed before processing as pulp [160]. The benefit of working with waste from the agar industry, thus, is that the waste biomass has undergone this process, leaving mainly cell wall endofibers. This material can be used for various purposes, including the development of packaging or other containers for various industries [206]. Cast-off seaweed blooms biomass can be used in this industry, providing an opportunity to reduce deforestation and global warming [161]. This biomass may also be used to manufacture non-woven fabrics, made with pressed fibres. The materials could be interesting for construction or architectural applications for protection, drainage, containment of land, and floor or wall coating. Both resistance [148] and thermal insulation [149] of *G. corneum* fibres have been previously tested in biocomposites, with encouraging results. In this last case, these coatings may also be moisture or acoustic insulating, but this still needs to be validated.

8.7. Seaweeds for Biofuels

The most abundant biofuels produced on an industrial scale are bioethanol, mostly from sugarcane and biodiesel from soy and palm oil, all edible plants or used in the food industry, being thus first-generation fuels. The wide use of these crops relies on the simplicity and affordability of growing these plants [162,207]. However, increasing demand for biofuel crops directly competes with food production, resulting in food shortages and rising food prices [39]. Seaweeds are an interesting alternative to land crops because they do not compete for arable land, can be cultivated year-round, use wastewater and CO₂ for the cultivation process, exhibit high yields, and different products can be produced along with biofuels [208]. Besides, seaweeds have an adequate composition in carbohydrates, which can be exploited for biofuel production through dehydration, hydrolysis, and fermentation to produce bioethanol and other derivatives [207]. Seaweeds do not exhibit lignin as in terrestrial plants. Removing lignin before hydrolysis is a major challenge increasing the costs of bioethanol production and generating chemical effluents [68]. Thus, the vigorous pre-treatment required to release fermentable sugars from lignocellulosic biomass is not necessary for seaweeds, representing a major advantage of using seaweeds [209]. The current challenge, thus, is to establish a successful and efficient strategy to achieve the full hydrolysis of the seaweed polysaccharides, as to release the fermentable sugars. Many studies are being developed to optimize the biofuel production from the different red, green, and brown seaweed species, through fermentation to produce bioethanol [66,67,159,210–212]. Frequently, the optimization process is species-specific [213] since in the production of biofuels based on macroalgae different species produce various forms of polysaccharides, which need to be subsequently hydrolysed into fermentable sugars [162]. Among the seaweeds, *Gelidium* has been appointed as a promising feedstock for the production of bioethanol [214]. In this context, *Gelidium* species have been tested as feedstock to produce bioethanol [215]. Amamou et al. [144] also describe a robust enzymatic process to efficiently hydrolyse *G. corneum* polysaccharides combined with mechanical fractioning,

which proved to be an efficient methodological approach for sugar production, required for the fermentation process.

Conventional acid and enzymatic hydrolysis methods are efficient, but less energy-intensive, non-toxic, and cost-effective methods are required. Moreover, a more efficient fermentation process is needed to turn this into a profitable economic activity. However, bioethanol production may not become a profitable activity by itself, requiring the addition of other extractive processes to algal biomass. Accordingly, in a biorefinery context, biofuels from seaweeds must be a cascade extraction process harnessing lipids, hydrocolloids, and other important biomolecules, thus adding value to the seaweed's biomass. In this context, Yoon et al. [159] also suggest the use of *G. corneum* biomass residues from pulp extraction of cellulose to manufacture paper for bioethanol production, through a sequential process of saccharification, purification, and then fermentation. Furthermore, waste biomass from the agar industry could be used alternatively to harvested/cultivated biomass to produce bioethanol, because, as stated earlier, the polysaccharides remain largely intact in the waste biomass. Another method to convert seaweed biomass into biofuels is anaerobic digestion producing biogas (methane) [28]. Likewise, the thermochemical conversion, using different temperatures and concentrations of oxygen produces biogas through gasification, biochar through pyrolysis, and bio-oils through liquefaction, which are alternative interesting energy sources [208]. The use of macroalgae in energy production could have favourable economic and environmental consequences. However, cost-effective and environmentally friendly conversion technologies are still key challenges to overcome before large-scale deployment of seaweed biorefineries [27].

9. Integrated Approaches and Future Perspectives

In the previous lines, possible applications of seaweeds have been described. Seaweeds are indeed an interesting feedstock for many bio-based materials and are being used as third-generation biofuels [208]. These can be produced in a single production step, or in a more sustainable valorisation of the biomass process, through an integrated biorefinery approach, making new products and producing energy at the same time. A complete algae biorefinery involves the integration of upstream and downstream processing of the biomass into bio-based products [214]. The upstream route depends on high biomass productivities, to ensure enough feedstock for the extraction processes. Thus, efficient, and low-cost cultivation processes are under development for several species of seaweeds. As stated, when it comes to *Gelidium*, further studies are needed to accomplish more productive profitable cultivation [47].

The downstream route includes the processing technologies to obtain biochemical products and biofuels. These include several steps, from the bioproducts recovery refinery to the biological conversion refinery, chemical conversion refinery, thermochemical refinery and, finally, to waste refinery [214]. The biological conversion step includes the fermentation and anaerobic digestion pathways, using different microorganisms to convert biomass into bioethanol and biogas, respectively. Although the production of bioethanol has been tested with *Gelidium*, this constitutes a challenging step when it comes to seaweeds, for each taxon presents specific polysaccharides, which must be hydrolysed into monosaccharides, not all fermentable into bioethanol, thus the optimization process is species-specific [215,216]. *Gelidium* has also been investigated as feedstock for anaerobic digestion to produce biogas (methane) [28,217] along with other seaweed species, with a direct relation between biomethane yield and high carbohydrate content. The anaerobic digestion, however, is influenced by several inhibitors that still need to be overcome, including the high molecular weight of organic compounds, which hampers cell wall disintegration during hydrolysis, among others [214,218]. Several hydrolysis treatments have also been tested, including acid, alkaline, and enzymatic autoclaving, among others [30,215]. Within this approach, Baptista et al. [143] extracted a valuable monosaccharide (D-tagatose) from the residues of *G. corneum* from the agar industry, by enzymatic isomerization, demonstrating the quality of the algal waste and the feasibility of the biorefinery process.

The chemical route includes, e.g., transesterification for high lipid content algae [28,219], which can be coupled with supercritical fluids such as water, CO₂, or alcohol to convert the lipids into biodiesel. After this transesterification process, the downstream process can further process the biomass to extract other valuable products. The thermochemical biorefinery uses the whole biomass for bioenergy conversion, namely through pyrolysis, gasification, and hydrothermal treatment. Pyrolysis-producing biochar, oil and gas have been successfully produced from *G. corneum* waste, presenting high fuel properties [170–172]. Target valuable bioactive compounds can also be extracted within the biorefinery context, using the chemical route, through solvent extraction, oxidation, polymerization, and other processes, often as a coproduct from biofuels production [215]. All of these are valuable compounds with recognized market applications, but these green extraction processes are still rather experimental, dependent on the desired products and have only been used in lab or pilot scales [219]. The residual biomass can also be used to produce energy and, the remaining solid fraction may be used as a fertilizer [220,221]. New extraction techniques are improving the cost-efficiency, sustainability, environmental and industrial-scale feasibility requirements to obtain algal components. New emerging extraction technologies are under development, such as ultrasonication, enzymatic processes, high hydrostatic pressure, microwave-assisted extraction, ultrasound-assisted extraction, radiation, or the use of alternative solvents, such as ionic liquid-based extraction, subcritical water or supercritical fluid extraction with CO₂, or a combination of several methodologies [26,48,127,162,216,222–225].

The effective implementation of these new technologies in the industry, however, has been difficult. Some of the reasons for this lack of growth include the significantly lower prices of traditional products (e.g., oil and plastics), low yields from biotechnological processes, great heterogeneity among seaweed biomass and waste properties, low availability of seaweed feedstock, time-consuming management of seaweed biomass and waste (collection, transport, and processing), among others. To tackle these challenges, it is essential for new technological advances in the extraction and purification of compounds to be incorporated into the industry, as well as to motivate political decisions, creating incentives for the industrial implementation of these processes. These should, as far as possible, be greener, more economical, and efficient, lowering costs and optimising production yields. This will respond to the growing market demand for healthy and environmentally friendly bio-based products and enable the successful industrial up-scaling of new scientific discoveries. Furthermore, the large demand and price increase in fossil fuels is creating an opportunity for the development of biofuels, especially second and third-generation biofuels. The supply has to meet a large fraction of this demand, but biofuels are not yet produced in quantity, with high yields or cost-effectively. As stated, a promising approach to reduce biofuel production costs is the use of biorefineries with the coproduction of high added-value products in a very efficient integrated approach. The added value of co-products will make it possible to produce fuels at more competitive costs and, therefore, achieve a higher market share [226]. To become truly circular and sustainable, the industry is changing from a fossil-fuel energy base to a renewable energy base and transforming waste into products [227]. Both seaweed biomass and waste can benefit from the cascade approach, allowing obtaining safe high-value bioactive compounds in a downstream process, which otherwise could be harder and more expensive to use [218,227].

10. Conclusions

Gelidium corneum is a remarkable species, providing noticeable ecosystem services, including shelter, food, nursery, and habitat for many other species, as well as biomass for human use. The communities they form in the temperate subtidal zone, thus, must be valued and properly managed. Still, climate change, pollution, and over-harvest have paid their toll and severely affected the *G. corneum* populations. This had a major impact on the wild harvesting of the species and the agar industry. Today, the species has been almost entirely replaced by other agarophytes, notably cultivated *Gracilaria*. As a scarce resource,

Gelidium biomass must be exploited with great caution. While it is true that there are coastal communities that depend on this wild resource, the low cost of feedstock and the increasing difficulty in harvesting sufficient biomass make the activity less attractive. Thus, local management plans for the resource are fundamental, guaranteeing sustained exploitation, according to the regenerative capacity of wild populations. On the other hand, to overcome biomass shortage further efforts must be performed to efficiently cultivate *G. corneum*. The sustainability of feedstock supply is a prerequisite for the continued exploitation of the resource.

The biomass of *G. corneum*, nonetheless, has been studied for other applications besides the production of agar. Mainly fibres from harvested biomass or industrial waste have shown potential to be included in various types of biomaterials. The incorporation of algae fibres into cotton or polyester, among others, could be an intelligent formula for incorporating UV protection into these fabrics, intended for photosensitive skins. The fibres are very resistant and can produce robust containers with antioxidant, antimicrobial, and biostimulant effects. Thus, another interesting application could be the use of residual biomass in the production of containers for agriculture purposes. These can be used to grow seedlings and then directly added to the soil. The biostimulant capacity improves the quality of the soil while reducing the consumption of plastic. The potential application as an architectural cladding is a hypothesis that likewise deserves to be investigated. The residues can also have many other applications, be it the production of biochar, bioremediation and CO₂ capture, or the production of biofuels. In a biorefinery approach, *G. corneum* biomass can thus benefit from a downstream process that will allow the recovery of different bioactive molecules that could have several industrial applications, and the remaining biomass can be used for agriculture, energy, or coating industries. This adds value to the algal biomass and makes full use of it, with no waste being produced. In their 2002 book *Cradle to Cradle*, Braungart and McDonough outline the design principles that underpinned the development of the circular economy concept [228]. These include the principles of circular design: a design for longevity, design for service, design for manufacturing, and design for the reuse of materials through recycling when the previous options are no longer possible. Seaweeds' biomasses, and especially their waste, are valuable resources for different industries. It is time now to seize the challenge and creatively develop innovative products through circular design.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/earth3030045/s1>, Table S1: Data retrieved from FishStatJ as *Gelidium*, *Gracilaria* and red seaweeds harvest; *Gelidium* and *Gracilaria* aquaculture, between 1960 and 2020.

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