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Macroalgae valorization for the production of polymers, chemicals, and energy

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ABSTRACT

The increasing awareness of man-made environmental impact of our modern highly industrialized societies has led to an increasing demand of natural products representing additionally a key element within the transition towards a circular bioeconomy. In this sense, macroalgae constitute a renewable and abundant natural resource with high usage potentials in a wide range of very diverse applications. In addition to the respective useable bioactive compounds varying between species and more significantly between the three taxonomic groups, macroalgae are characterized by additional promising properties such as high growth rates, simple cultivation and harvesting strategies, and no competition with fertile lands. These are only some reasons why macroalgae have been used for centuries as a raw material in very different application fields; the food sector is only one prominent example. Furthermore, new macroalgae-based products have been developed over the last decades; this is true for hydrocolloids, nutra- and pharmaceuticals as well as biomaterials. Additionally, nowadays macroalgae are also a potential feedstock for green energy production in the form of gaseous and liquid biofuels such as biogas, bio-oil, or bioethanol. Within this context this review provides an analysis of the main compounds with possible economic potential obtained from green, red, and brown macroalgae as well as an evaluation of the global macroalgae market and the existing obstacles. Additionally, it also provides an overview of the macroalgae potential in new applications and as a source of green energy.

1. Introduction

Algae biomass has been used for centuries as a source of fertilizers, cattle feed, and human food. Furthermore, new applications have been later developed such as feed and food supplements, hydrocolloids, nutraceuticals, pharmaceuticals, biofuels and biomaterials. Such new applications have led to an increase and diversification of macroalgae products in the market in recent years [1].

These markets, led by especially Asian countries, are currently gaining growing interest also in Europe due to the wide number of possible macroalgae applications, their potential health benefits and the increasing tendency towards natural products [2–4]. In this sense, the feasibility of a macroalgae production within the North Sea have extensively been studied [5] concluding that macroalgae cultivated here show several applications with possible economic potential. Nevertheless, still further research is required in certain areas such as the assessment of the nutritive value in animal diets, the protein digestibility, the extraction of valuable components for fermentation and chemicals, etc. Furthermore, sustainable cultivation systems must be based on a mono- and co-culture nutrient management model or on Integrated Multi Trophic Aquaculture (IMTA) cultivation systems so that nutrient inputs and outputs are balanced and no need is given to additionally introduce specific nutrients into the natural environment. In this sense, a much better understanding of the existing nutrient flows in the open sea is required [3,5]. Industrial macroalgae production, especially when used for food/feed applications, is undesirably influenced by the uptake by macroalgae of possible marine toxins (e.g., cyanotoxins produced by blue-green microalgae) or heavy metals such as arsenic or mercury. Additionally, microplastics play an important role in the safety of macroalgae products since they might be accumulated in denser macroalgae vegetation. Therefore, potential risks associated with the use of macroalgae as a resource for food, feed, and energy need to be

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further studied in order to achieve a sustainable and safe macroalgae biorefinery approach [5].

Despite the fact that macroalgae production in Europe is currently limited at least by regulatory constraints and market challenges, this sector is characterized by a significant potential for an important contribution towards sustainable development on the background of the ongoing efforts to transform our industry step by step towards a bio-economy [6]. This is true because a macroalgae based biorefinery aligns with several of the 17 Sustainability Development Goals (SDG) defined by the United Nations (UN) like promoting a sustainable agriculture (No. 2), ensuring sustainable consumption and production techniques (No. 12) and protection and sustainable use of the oceans (No. 14) [7].

This review presents important potentially economically valuable macroalgae species, with a clear focus on the North Sea as well as the Baltic Sea, and identifies existing product markets. For doing so macroalgae that are endemic to the North and Baltic Sea region are identified and their promising components or properties are introduced together with their biochemical composition. Based on these properties, product markets are examined and presented that could be of importance for the application and use of macroalgae as a resource. With the results of this study the overall potential of macroalgae is evaluated.

2. Macroalgae species endemic to the North and Baltic seas

Even though 70 % of the earth's surface is covered by sea water, the biomass demand of humankind is predominantly supplied from terrestrial resources. The use of the marine environment as a possibility to obtain biomass resources untapped so far – and in this respect mainly plant biomass – beside the "classical" use as a resource for harvesting of fish – is new and currently gaining more and more attention as fertile land declines and biomass demand increases.

Macroalgae grow in the euphotic zone being the upper marine zone that meets the photosynthetic light requirements of phytoplankton. The maximum depth of the euphotic zone is 200 m depending on the turbidity of the water. Marine macroalgae can grow on stony or hard substratum and mainly occur on nutrient rich areas and coasts [8,9]. For sufficient growth rates, a high nutrient level of the marine environment is necessary taking commercial constraints into consideration.

Coasts close to countries/cities with high population densities are eutrophic (excessive nutrients) or even hypoxic (depleted oxygen levels). In these areas, nutrients are introduced into the marine environment either directly or via rivers mainly by industrial and agricultural activities as well as human and other biological wastes [10]. In general, low oxygen concentrations can increase photosynthetic rates for species that are able to photorespire, nevertheless, macroalgae might be significantly impacted by other factors like ocean acidification, fluctuating pH and temperature. They do not necessarily grow better or excessively in eutrophic waters [11].

To establish an industrial utilization of macroalgae, different criteria need to be considered such as feedstock provision (e.g., cultivation, wild harvesting, beach wrack collection), and, depending on the area, available/suitable macroalgae species [12]. Therefore, Table 1 shows indigenous species in the Baltic and the North Sea with potential economic potential.

Macroalgae are classified into three major groups: brown (Phaeophyta), red (Rhodophyta) and green (Chlorophyta) macroalgae. The chemical composition of each group varies considerably in terms of the lipids, proteins and carbohydrates content. Furthermore, each group is also differentiated by the synthesis and metabolism of carbohydrates and therefore, in the composition of the cell wall and the intracellular matrix. Pigments are also characteristic for each group: fucoxanthin and phycobilins are contained in Phaeophyta and Rhodophyta, respectively, and chlorophyll a and b, carotenes and xanthophylls in Chlorophyta. Within each group, the species composition varies notably with harvest season, habitat, environmental conditions, geographic area, etc. [16].

Their biochemical composition and the main compounds with a potential economic value are described below. Fig. 1 shows an overview of the main macroalgae species and derived products.

2.1. Brown macroalgae

Brown algae (Phaeophyceae) grow almost exclusively within marine habitats of cold to temperate zones. Their characteristic color and thus their name is caused by the pigment fucoxanthin. Brown algae are mainly used for food (Wakame and Kombu) and for the production of the hydrocolloid alginate applied as gelling agent in the food, cosmetic and pharmaceutic sector. The globally given demand is fulfilled mainly by harvesting of wild population. Additionally, also aquaculture methods are established for highly used species under promising local conditions.

Some brown algae species can reach enormous sizes and form typical kelp forests. The overall largest macroalgae species *Macrocystis pyrifera,* also known as giant kelp, produces thalli of up to 25–45 m length.

Here examples for brown algae species with potential economic potential are introduced growing in European coastal waters. Additionally, their chemical composition is summarized (Table 2).

- ***Laminaria* spp.** is a macroalgae genus found in temperate to polar marine habitats of the Northern hemisphere with many different species and subspecies [17]. These algae are mostly used as a food precursor and play a very important role in alginate production due to its high guluronic acid content [18].
- ***Undaria pinnatifida***, also known as Wakame, is originally native to the cold-temperate costal lines of Eastern Asia where a large-scale production in aquaculture is established. But this species has spread as an invasive plant to many other areas like North-Eastern Atlantic and Mediterranean Sea [19]. *U. pinnatifida* is mainly used as food ingredient within the Asian cuisine and contains various bioactive compounds such as polysaccharides, polyphenols, and peptides [20].
- ***Fucus vesiculosus*** is a well-known species of the genus *Fucus* which is comprised of 66 species [21]. They frequently occur in cold to

Table 1
Macroalgae species with potential economic potential endemic in the North Sea and the Baltic Sea.

Class	Species	Growth rate, wt%/d	Parameter	Optimal temperature, °C	Source
Phaeophyta (brown macroalgae)	*Saccharina latissima* (formerly *Laminaria saccharina*), (Sugar kelp)	Up to 20	dry weight	<18	[5]
		Up to 10	length	10–15	[13]
	Laminaria digitata (Finger kelp)	Up to 20	dry weight	September–May/<18	[5]
	Fucus vesiculosus (Bladder wrack)	Up to 15	fresh weight	15–20	[14]
		Up to 3	length	10–20	[13]
Rhodophyta (red macroalgae)	*Palmaria palmata* (Dulse)	Up to 35	dry weight	Summer/15-20	[5]
	Furcellaria lumbricalis	Up to 3	dry weight	Summer/10-20	[15]
	Chondrus crispus	Up to 3	area	10–20	[13]
Chlorophyta (green macroalgae)	*Ulva lactuca* (Sea lettuce)	Up to 50	dry weight	Summer/15-20	[5]
		Up to 24	area	10–20	[13]
	Ulva sp. (formerly *Enteromorpha* sp.)	Up to 7	length	10–20	[13]

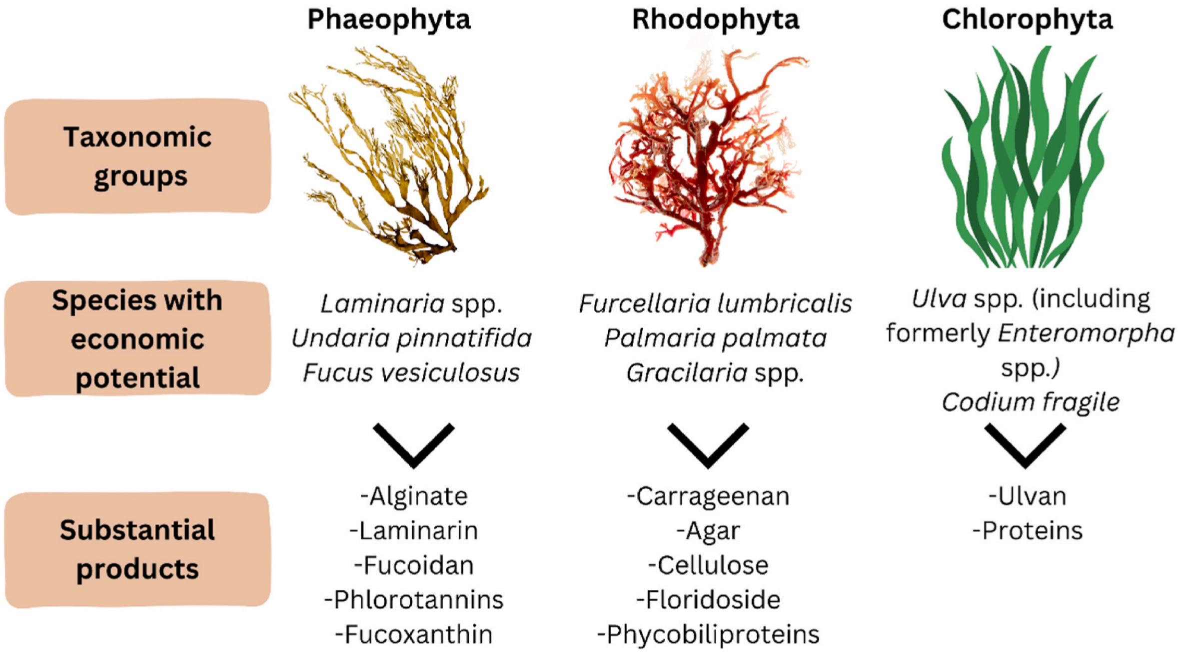

Fig. 1. Macroalgae taxonomic groups with the main species with economic potential and substantial products.

Table 2
Biochemical composition of the brown algae species *Laminaria* spp., *Undaria pinnatifida* and *Fucus vesiculosus*.

	Carbohydrates, %	Proteins, %	Lipids, %	Ash, %	Reference
Laminaria spp.	54–82	7–8	–	22–42	[24]
Undaria pinnatifida	9–52	3–21	≤1	27–28	[20,25,26]
Fucus vesiculosus	34–66	1–11	1–4	23–36	[21]

temperate waters along rocky shorelines of the Northern hemisphere. Due to their macro and micro nutrients they are used as food in Asian and European countries [22]. They contain fucoidans, phlorotannins, and fucoxanthins as bioactive compounds with potential applications in the food, cosmetic and pharmaceutic sector [23].

2.2. Red macroalgae

Red algae (Rhodophyta) represent one of the oldest and largest group of eukaryotic algae with more than 10,000 registered species distributed worldwide. These are the most widely cultivated and commercialized algae in the world and used as source of food ingredients, hydrocolloids, fertilizers, and animal feed, among many other applications. In addition, their unique polysaccharide composition makes them a suitable raw material for several industrial areas such as the food and pharmaceutic sector [27]. Rhodophyta are photosynthetic, lack flagella and contain chlorophylls (a and b), carotenoids and phycobiliproteins. They are found in all latitudes (being more abundant in equatorial regions) and can live at high depths of up to 200 m thanks to their content in accessory pigments such as β-carotene and phycobilins [28].

Below, red macroalgae species with a potential economic value that can be found in European coastal waters are presented (Table 3).

Table 3
Biochemical composition of the red algae species *Furcellaria lumbricalis*, *Kappaphycus alvarezii* and *Gracilariopsis longissima* (formerly *Gracilaria verrucosa*).

	Carbohydrates, %	Proteins, %	Lipids, %	Ash, %	Reference
Furcellaria lumbricalis	30–57	16–28	≤1	33	[34–36]
Palmaria palmata	41–66	17–27	≤1–2	15–21	[37,38]
Gracilariopsis longissima (formerly *Gracilaria verrucosa*)	32–41	16–24	≤1	3–13	[39–41]

- ***Furcellaria lumbricalis*** is a particularly important alga in Europe due to its already realized long time use by the food industry. This algae is used as a source of agar, carrageenan and sulfated galactan being widely used in dietary products to improve their structure and sensation in mouth [29]. Some other compounds with biotechnological applications are pigments (phycoerythrin), oxylipins, phenolic compounds and minerals [15].
- ***Palmaria palmata*** is a red algae species, also known as "dulse", being common on stony coasts of the Northern hemisphere. It is traditionally harvested for human consumption and is recently gaining more awareness due to its nutritional benefits as well since it contains high amounts of vitamins, iron, minerals, and fibers. This species is characterized by an umami taste and a comparatively high protein content of up to 35 % (DW) [5]. Proteins of this species are known to have a high quality, with up to 50 % of the total amino acids being essential amino acids, as well as bioactive and health-promoting properties [30].
- ***Gracilaria* spp.** are extensively used in industrial applications; more than 80 % of the global produced agar comes from *Gracilaria* species. This genera also contains numerous bioactive metabolites used in different industries such as mycosporine-like amino acids (active ingredient in sunscreens), phycobiliproteins (color additives), prostaglandins (pharmaceutical ingredient) or carotenoids (antioxidant and color additive) [31,32]. Within the *Gracilaria* genus, *Gracilariopsis longissima* (formerly *Gracilaria verrucosa*) is the most commonly used species in Europe [33].

2.3. Green macroalgae

Green algae (Chlorophyta) consist of around 7000 known species distributed globally from polar to tropical regions. Compared with red and brown algae, green macroalgae are less sensitive to climate change and have a relative advantage in warm climates. Despite being the least commercially exploited group, several components are derived from Chlorophyta such as proteins (lectin and taurine), polysaccharides (ulvan), vitamins (tocols), and antioxidants (carotenoids, chlorophylls, bromophenol, etc.). In addition, their polysaccharide fraction represents a promising ingredient to form new biomaterials. Nowadays, the most widespread application of green macroalgae is as animal and fish feed and as a fertilizer [27,42].

Green macroalgae species found in European coastal waters with a potential economic potential are discussed below (Table 4).

- ***Ulva* spp.** is a well-studied genus of macroalgae species that can be found along the western parts of the Baltic Sea, among other areas. Its rich composition in micro- and macronutrients and bioactive compounds makes *Ulva* species a valuable source of food and pharmaceutical ingredients [43]. Especially, the sulfated polysaccharide ulvan displays biological and physicochemical properties of interest in food, medicine and chemical applications. Among *Ulva* species, *Ulva lactuca* is one of the most extensively used species as food ingredient due to its high nutritional value derived from its high content in proteins, minerals and vitamins [44,45].
- ***Ulva* (formerly *Enteromorpha*)** species grow abundantly in coastal zones of the Southern Baltic Sea [46]. The potential economic potential of this algae is due to the rich content of essential amino acids and minerals (mainly Ca and P), which makes it a potential product for human consumption. Furthermore, Ulva sp. (formerly *Enteromorpha* spp.) are rich in bioactive compounds, being sulfated carbohydrates the most abundant. Polysaccharides from this genus exert physiological and biological properties such as antioxidant, anti-microbial, anti-cancer and anti-coagulant, among many others [47,48]. Currently, all species of the genus *Enteromorpha* have been reclassified to the genus *Ulva*.
- ***Codium fragile*** is an invasive species that can be found along European coastlines as well as in lots of other parts of the world. It has been studied for its dietary and nutritional value as food and feed, but also for specific compounds such as bioactive phenolic compounds. Furthermore, *C. fragile* extracts exert anticancer, anticoagulant and anti-obesity effects [49]. Since *C. fragile* is an invasive species, it represents a promising source of natural compounds and its exploitation by wild-harvesting for industrial applications can help to preserve natural habitats [50].

3. Sources

For commercial and large-scale applications, macroalgae biomass is produced by aquaculture or by wild harvesting. In Europe, the most extensively cultivated species are *Alaria esculenta, Palmaria palmata, Saccharina latissima and Ulva* spp. and the most harvested species are *Laminaria hyperborea* and *Ascophyllum nodosum* [1]. There are different ways to make these and other macroalgae available for an industrial use from the oceans. These options can be differentiated into wild harvesting, beach wrack collection and aquaculture and are explained below. Beach wrack is defined as organic material constituted by macroalgae and seagrass that has become detached from the place where it grows at the sea and accumulates on the beach.

3.1. Wild harvesting

This system consists of harvesting and gathering macroalgae from wild stocks. The main requirement for this approach is the availability of a harvest technology and labor during the seasonal occurrence of the species or genera of interest [56]. Mechanical harvesting is the preferred option for large-scale applications and is performed by boats using different technologies such as a macroalgae trawl, a paddle wheel cutter or a vacuum-sucker. Alternatively, some species are also harvested manually [57].

Macroalgae feedstock might strongly vary in quality due to the natural heterogeneity of physicochemical parameters within marine environments. Strong fluctuations in algae communities can be observed due to changes, among others, in light and nutrient levels, temperature, salinity or presence of contaminants. In addition, food safety can represent a concern since it depends on the water quality of the area [58].

Sustainable and effective management of algae and application of sustainable harvesting practices is especially important for wild harvesting to avoid over-exploitation of natural environments with commercial purposes. A critical assessment of harvesting regimes and a profound knowledge about the dynamics of natural local populations is crucial for the sustainable exploitation of wild harvested macroalgae [56].

3.2. Beach wrack

Beach wrack represents a low cost and abundant source of marine macroalgae. Nevertheless, the composition of this biomass to be washed ashore is very heterogeneous and influenced by numerous factors such as species, season, environmental conditions within the respective ocean, storage time on the beach, environmental/meteorological conditions at the beach, degree of natural biomass decomposition/degradation etc. In addition, changes in macroalgal communities as a consequence of opportunistic and invasive species have an effect on the composition and amount of beach wrack [59]. The fluctuating and hardly predictable availability of beach wrack challenges a reliable biomass supply chain for basically any kind of application processes. For these reasons, the most studied applications of beach wrack with potential economic prospects include its use as soil improvement ingredient, biofertilizer, for coastal protection as material to avoid accelerated dune vegetation succession and as an additional substrate for biogas production [22,60].

The suitability of beach wrack for these applications depends directly on the development of cost-efficient technologies for its collection on beaches as well as for the separation of the organic fraction from sand and other natural as well as anthropogenic residue and waste components [60].

3.3. Aquaculture mass cultivation

Aquaculture systems can be performed in land-based tanks and ponds or in offshore and coastal production facilities. Offshore algal cultivation is characterized by its simple installation and maintenance is traditionally performed by using ropes, lines, rafts, cages, or nets where

Table 4
Chemical composition of selected green algae found in Europe.

	Carbohydrates, %	Proteins, %	Lipids, %	Ash, %	Reference
Ulva lactuca	41–60	10–23	1–4	14–29	[51]
Ulva sp. (formerly Enteromorpha spp.)	14–65	9–14	2	33–36	[52,53]
Codium fragile	42–67	11–12	1–2	21–50	[50,54,55]

macroalgae are attached [27]. More advanced methods have been developed lately as a mooring system anchored to the seabed consisting of a grid of ropes where the macroalgae plant to be cultivated is connected [61]. Within this group, long-line systems including anchors, buoys, and tope ropes are the most extensively used methods [62]. There are different ways to cultivate macroalgae on ropes and lines. On the one hand, the algae can settle naturally on lines laid out in the sea, but this way an inhomogeneous and uncontrolled culture develops. On the other hand, desired types of algae can also be cultivated in a targeted manner indoors and are immobilized on the ropes in indoor hatcheries [63]. After that the ropes are brought to the sea and the algae sprouts can grow. To implement a biorefinery based on macroalgae, offshore cultivation seems to be the preferred option, as near-shore cultivation might compete with alternative coastal uses (e.g., tourism) [64]. Generally it is important only to cultivate species that are also endemic to the specific cultivation area in order to avoid disturbances of the ecosystem like the uncontrolled proliferation, e.g. due to a lack of predators.

In addition to large-scale open water cultivation, some macroalgae species are cultured in on-land tanks and ponds. This method represents a sustainable alternative which enables demand-based production of macroalgae biomass [56,65]. Nevertheless, tank cultivation requires expensive construction, operation, and maintenance efforts limiting the production of functional ingredients from macroalgae at industrial scale. In addition, this system is influenced by several factors such as the site selection, tank design and construction, reproductive biology of the species, strain selection and control of environmental conditions, among others [66].

Further improvement of aquaculture systems should be focused on the development of product-specific cultivation technologies which are cost and energy efficient [67].

4. Products

Macroalgae extracts like carrageenan, alginate, and agar make up 40 % of the hydrocolloid market in the food industry [68]. The growing interest in macroalgae as a substrate to be used for such applications is based on the trend towards healthy and natural products in nutraceuticals, cosmetics and bio-based materials as well as the need for renewable resources for sustainable energy production (e.g., biofuels) [33]. This section highlights the potential of macroalgae biorefineries as source of chemicals and energy carriers.

4.1. Substantial products

Macroalgae contain high valuable components such as polysaccharides, proteins, polyphenols and pigments, the main components of macroalgae and their applications are presented in Fig. 2.

4.1.1. Polysaccharides

Polysaccharides from marine plants vary a lot from those of terrestrial plants due to the different environmental conditions such as light intensity, salinity, temperature and nutrient dynamics. Table 5 shows the main interesting polysaccharides, their content in the biomass and their characteristics.

Applications of alginate. Alginate can be extracted by different techniques depending on the intended application. A typical way is to form sodium alginate by adding acid, ethanol and sodium carbonate so that the insoluble calcium and magnesium alginates that are located in the cell wall are transformed to soluble sodium alginate [79]. Apart from possible changes of the physico-chemical characteristics of the alginates related to the chemicals used, there are also difficulties in its purification due to the high viscosity of the sodium alginate solution [76].

Alginate forms viscous solutions in water and is, due to its gelling, stabilizing and thickening properties, an important additive within the food industry (e.g., for sauces, ice creams, syrups, jellies, etc.). It also prevents water-in-oil emulsions like mayonnaise from separating or the formation of ice crystals in ice cream after defrosting and refreezing. In combination with calcium ions, it forms a water insoluble matrix, which is often used to create liquid spheres in molecular cooking. Droplets of a liquid mixed with e.g., calcium chloride are transferred into a sodium alginate solution and develop a sodium-calcium alginate skin [80]. With the growing trend and demand for meat alternatives and substitutes, alginate is gaining more attention in the food industry for its use in meat analogs, where alginate is used together with protein fibers to imitate the known meat structure with the desired chewability, elasticity, and stretchability [74]. Apart from that, alginates are also widely used in the cosmetic industry as thickening and gelling agents to stabilize and improve the consistency of creams, lotions, toothpaste and foams [76,81,82].

Depending on the treatment, alginates can be modified to desired materials and form sponges, gels, porous scaffolds, microcapsules, fibers and microspheres [73,83] (e.g., it is pharmaceutically applied in tissue healing; due to the high water content in the gels, it generates a cooling function and water emission to the wound which facilitates faster wound healing [84,85]). Alginate-based wound dressings are non-toxic,

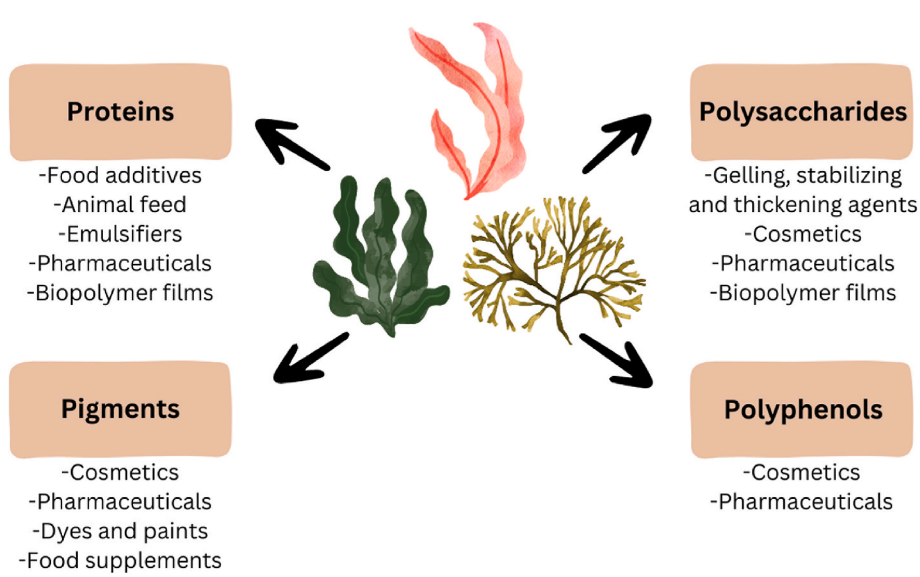

Fig. 2. Main applications of the different compounds produced by macroalgae.

Table 5
Important polysaccharides of the three different macroalgae groups.

Biomass	Polysaccharide	Amount, wt.-% DW	Characteristics	Source
Brown Macroalgae	Laminarin	Up to 30	Not viscous, water-soluble, no gelling properties, anti-tumor properties, prebiotic effects, anti-coagulant, anti-inflammatory	[69–72]
	Fucoidan	Up to 40	Viscous, water-soluble, prebiotic effects, anti-coagulant, anti-tumor properties, anti-viral properties, natural antioxidant, anti-inflammatory agent	[70,71]
	Alginate	Up to 50	Viscous, water-soluble, gelling properties, mucosal protection, anti-bacterial effects, prevention of gastric ulcers, wound healing	[5,69–71,73,74]
Red Macroalgae	Carrageenan	Up to 50	Viscous, water-soluble, gelling properties, badly fermentable, anti-coagulant, anti-tumor and anti-viral properties, hypoglycaemic/anti-diabetic properties	[70,75,76]
	Agar	Up to 30	Water-soluble, gelling properties, anti-tumor properties	[5,69,70]
Green macroalgae	Ulvan	Up to 55	Viscous, water-soluble, badly fermentable, binding of heavy metals, anti-influenza, treatment of gastric ulcers, antioxidant, antiviral, anticancer, anti-aging	[70,77,78]

biocompatible, water absorbing and they have a good water vapor transmission rate, antiseptic properties and conformability. Alginate hydrogels are also applied in cartilage repair/regeneration or alginate porous scaffolds with implanted stem cell for bone regeneration. The scaffold is then injected to promote bone tissue formation [73]. Another pharmaceutical application is the development of drug delivery systems using alginate-based hydrogels, polyelectrolyte complexes and colloidal particles. Alginate microcapsules can be modified by adding PEG (for higher stability at low pH) or varying wall thickness and composition so that a controlled release of drugs can be achieved [73].

With the ability to form strong polymeric networks, alginate is also used for the production of biobased packaging materials. Since it is very hydrophilic, it is not suitable for applications that require water repellent characteristics. However, it can be utilized as a protective barrier e.g., to prevent that covered food does not lose its moisture [86].

Film formation using alginate can be performed by different mechanisms such as coacervation, gelation or thermal coagulation. The most common method to produce biopolymer films is the solvent casting method, where a solution is poured on a surface and the solvent (usually water or water-ethanol) is evaporated. Alginate can also be combined with other polymers such as proteins to produce materials with enhanced properties since the proteins and polysaccharides can form stable complexes by binding covalently or electrostatically together [86–88].

First experiments on the production of macroalgae packaging materials from whole biomass using extrusion techniques has also been conducted [145]. Since the extraction of pure natural polymers from macroalgae is expensive, the use of whole biomass is favorable in this respect regarding production applicability [89].

Applications of carrageenan. Carrageenan can be obtained in a refined or semi-refined grade. For the semi-refined grade, the whole macroalgae biomass is heated in a potassium hydroxide solution. This way, μ- and ν-carrageenan (precursors) are transformed to κ- and ι-carrageenan showing better gelling properties [76]. The whole biomass is then dried and pulverized. For the refined carrageenan, biomass is heated up to ~100 °C and afterwards, carrageenan is obtained by alcoholic precipitation [79].

Carrageenan also has good gelling properties and forms thermo-reversible gels, so that it is used in the food industry as a thickener and stabilizer as well as alginate. ι- and κ-carrageenan have gelling properties, while λ-carrageenan is applied as a thickener or viscosity enhancer. In contrary to alginate, carrageenan forms thermally reversible gels by ionic and hydrogen bonds [3,80]. Since it has good water binding properties, it is also used as a fat substitute in processed meat with low fat content to keep the softness. Apart from that, carrageenan is also used as a gas barrier to prevent oxidation reactions e.g., on sliced fruit by applying it as a coating [83].

Additionally, carrageenan has various applications in the pharmaceutical industry due to its anticoagulant, antithrombotic, antiviral, antitumor and cholesterol lowering effects [83]. It is also used for controlled drug delivery in microcapsules and microspheres [76].

Applications of agar. Agars are usually extracted like carrageenan with an alkaline treatment to induce a chemical conversion of the precursor L-galactose-6-sulfate to 3,6-anhydro-galactopyranose for better gelling properties. This is followed by an extraction with water at ~100 °C for ~3 h, filtration of residues and an alcoholic precipitation of the agar [76].

Agar forms thermally reversible gels as well as carrageenan and is therefore also used as a gelling and thickening agent. Some agars from specific macroalgae species (e.g., *Gracilaria chilensis*) interact well with sugar and the combination result in a higher gel strength. Hence, it is used, among other applications, in candies with high amounts of sugar [83].

Agar also shows thermo-reversible properties and is therefore used as dental impression material. In laboratories, agars are used in different grades as culture media ingredient because they are hardly digestible and metabolizable. Additionally, they form strong, transparent gels [83]. Agarose purified from agar is used for gel electrophoresis or chromatography [76].

Applications of sulfated polysaccharides. Sulfated polysaccharides such as fucoidan, laminarin and porphyran from brown algae, ulvan from green algae or carrageenan from red algae have a high value due to their bioactivity (Table 5). They do not all have gelling or viscous properties, but still have promising potential in the biomedical or cosmetic industry due to, among others, their skin whitening, anticoagulant, anti-inflammatory and antiulcer activities. Fucoidan has influence on the activity of human skin enzymes such as metalloproteinase-1 as well as moisture retention capacity and is therefore used in anti-aging products [80,82]. Still further research is necessary regarding the topical applicability of these substances, since it was shown that, for example, fucoidan can penetrate the skin and reach into muscles and plasma and be distributed [81]. Ulvans are not commercially used so far but are currently researched for pharmaceutical and cosmetic applications due to its bioactive properties (Table 5) and metal binding abilities as well as for biomedical applications such as tissue engineering and structures like nano-fibers, particles, hydrogels and membranes [78].

Another polysaccharide of interest for biomedical applications is cellulose from macroalgae. Compared to cellulose from terrestrial plants, macroalgae cellulose has several benefits, some of them related to the absence of lignin which results in pure cellulose fractions that are more suitable for biomedical applications. Furthermore, the lack of lignin allows the extraction of cellulose under less severe conditions, resulting in less degraded cellulose fractions [90].

4.1.2. Proteins

Protein content in macroalgae has a wide range from ~3 % (DW) in brown algae up to ~47 % (DW) in green and red algae [91–93]. Proteins from aquatic biomass are predominantly structural and enzymatic, for example, for photosynthetic reactions. They vary significantly regarding charge, complexation and hydrophobicity and they can only be classified by soluble and insoluble proteins and not by Osborne-fractionation like storage proteins (e.g., in crops). Soluble proteins from aquatic biomass consist mainly of RuBisCo (ribulose 1,5-bisphosphate carboxylase/oxygenase) [94,95], while insoluble proteins are dominated by membrane proteins in association with lipids, pigments and carbohydrates [91,96–99]. Their size varies in a wide range from <5 kDa to >60 kDa. The heterogeneous nature of these proteins leads to low extraction yields and hampers the industrial usage of algal proteins [95]. Further research in this field is therefore necessary to unlock the great potential that lies in proteins from macroalgae since they offer a lot of opportunities as food additives, nutraceuticals, pharmaceuticals, cosmetics and biobased materials. Fig. 3 shows the main steps for the extraction and purification of macroalgae proteins.

While the human consumption in Asian cultures involves mainly untreated macroalgae (e.g., Nori for sushi wrapping), the Western countries are also expanding their research in protein extraction from macroalgae [93,101]. The quality of macroalgae proteins has mainly been studied for species that are generally consumed as human food. The protein quality is considered high since 40 % of the total amino acids are essential amino acids [92]. Cytoprotective, anti-aging, anti-inflammatory and anti-tumor properties are attributed to macroalgae amino acids, which is why potential applications in the food, pharmaceutical and cosmetic industry are being researched [81].

4.1.3. Pigments

Pigments in macroalgae differ due to photosynthesis in different depths of the sea. Chlorophyll, which occurs almost in all macroalgae, exists in different kinds of similar structures (porphyrin ring with varying substituents). Other pigments are carotenoids and phycobiliproteins, that enlarge the absorbed light spectrum. In red macroalgae, the major pigments are phycobiliproteins, in brown macroalgae it is chlorophyll c (chl c) and fucoxanthin and in green macroalgae it is chlorophyll b (chl b) and lutein. The main function of pigments is the capture of the sunlight energy for photosynthesis by light absorption, transfer of excitation energy to reaction centers and the degradation of excess energy to avoid a damage of the photosynthesis apparatus [92,102].

Due to the hydrophobic nature of most of the macroalgal pigments (e.g., chlorophylls, carotenoids), they are typically extracted with nonpolar solvents such as acetone, dimethylformamide, diethylether and methanol [92]. Phycobiliproteins are water soluble and can be obtained after cell disruption by water extraction or in buffers. Macroalgae pigments contain antioxidants and have anti-inflammatory, antiviral, neuroprotective, anti-obesity, anti-angiogenic and anti-cancer activities [102,103]. Applications of algal pigments already exist in the pharmaceutical or nutraceutical industry such as the use of fucoxanthin as an expensive weight loss supplement, β-carotene as a pigment or health supplement and phycocyanin as a fluorescent probe in laboratories for analytical purposes [104].

4.1.4. Polyphenols

Another component mainly found in brown macroalgae and in lower amounts in red macroalgae are phlorotannins, which are polyphenols with valuable properties such as hyaluronidase inhibitory capacity, antioxidant, anti-inflammatory, neuroprotective and anti-microbial activities. Macroalgae produce these polyphenols to protect themselves from feeding herbivores, formation of free radicals due to high oxygen concentrations and bacteria. The cosmetic industry applies phlorotannins as anti-aging agent for the skin, since the aging process of the human skin is characterized by free radical damage and the reduction of hyaluronic acid concentration [105,106].

4.2. Energetic products

Macroalgae generally contain low amounts of lipids but high amounts of carbohydrates, which is why their energetic use is practically limited to the production of gaseous fuels or fermentative pathways to produce liquid biofuels.

Depending on several factors such as age, season, species and environment, macroalgae can contain up to 70 wt-% (DW) of polysaccharides [107]. The main difference between macroalgae and terrestrial plants is that algae offer various special polysaccharides like alginate, ulvan, carrageenan, mannitol and laminarin while terrestrial plants are mainly constituted by starch and cellulose. This complicates conventional processes as corresponding enzymes do not exist yet or are too expensive. However, they do not contain lignin and only little amounts of hemicellulose so that the hydrolysis does not need to be as harsh, which in turn facilitates the pretreatment [108,109]. One major limitation in the energy conversion of macroalgae is the high water content of 70–90 wt-%. Thus, methods that require dry biomass for further processing like gasification and pyrolysis are not suitable for this type of biomass [110,111]. Therefore only methods that can be applied to wet biomass are considered in this review: fermentation, hydrothermal liquefaction and anaerobic digestion for the production if bioethanol, bio-oil and biogas respectively [112–114].

4.2.1. Bioethanol

The production of bioethanol by fermentation based on a starch-containing feedstock is usually preceded by a physical, chemical or biological pretreatment for cell disruption of the biomass and a subsequent hydrolysis/saccharification step. Related to macroalgae, complex sugars (polysaccharides) such as alginate, laminarin, carrageenan, ulvan or agar need to be hydrolyzed to sugars (monosaccharides) to be

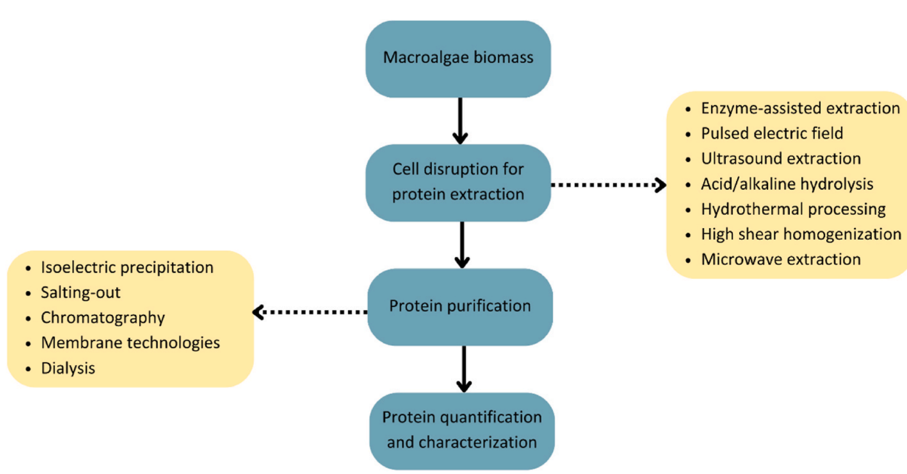

Fig. 3. Overview of a common process for cell disruption and protein extraction from macroalgae biomass. Adapted from de Souza et al. (2023) [100].

afterwards fermented to ethanol by yeast or bacteria. For lignocellulosic biomass, the use of diluted sulfuric acid (H_2SO_4) or hydrochloric acid (HCl) at a temperature of 121 °C for 45 min is a widely applied method which often is also applied for macroalgae biomass under less harsh conditions [110,115].

Enzymatic hydrolysis of algal biomass usually leads to significantly higher yields than chemical hydrolysis and is also more sustainable since less toxic by-products are produced [108,114,116]. For lignocellulosic photosynthetic biomass, enzymes such as cellulases, amylases, lysozymes or glucosidases are applied to break the glycosidic bonds of the polysaccharides and thereby produce reducing sugars that can be fermented to bioethanol [108,111]. These are also applied for macroalgae biomass, but not all of the macroalgae polysaccharides (e.g., alginate, carrageenan, fucoidan, ulvan) can be hydrolyzed using these common enzymes. The hydrolysis of those polysaccharides needs specific enzymes that are not yet commercially available.

During fermentation, these monosaccharides are converted to ethanol and CO_2 as a by-product. The most widely used microorganism for this purpose is the yeast *Saccharomyces cerevisae* [108,117]. Nevertheless, the composition of monosaccharides or fermentable sugars can vary between macroalgae groups and species, therefore, the most suitable microorganism for this purpose has to be chosen depending on specific conditions and prerequisites [110]. Table 6 shows results of several studies on the fermentation of macroalgae for bioethanol production.

Overall, the potential of bioethanol production by fermentation from macroalgae is promising, but there are still some obstacles regarding the hydrolysis process. Sustainable hydrolysis methods (e.g., enzymatic) are not yet economic viable. Moreover, the depolymerization of marine biomass polysaccharides is not yet established and needs further research due to their special composition. Finally, a cascade approach should be realized due to high amounts of by-products such as proteins, lipids and unfermentable sugars that are necessarily generated [107, 108].

4.2.2. Bio-oil

The principle of hydrothermal liquefaction (HTL) is the elevation of pressure to keep water in a liquid state, also called subcritical water (5–20 MPa, 250–350 °C). In this state, water has a lower dielectric constant and therefore a lower polarity. Also, under these conditions, water dissociates to hydronium (H_3O^+) and hydroxide ions (OH^-) due to the increased self-dissociation constant and therefore allows acid-base catalysis. This way, water becomes a solvent that is able to react with nonpolar and organic substances. Hydrogenation reactions break down larger molecules, such as proteins, lipids and carbohydrates, and these fragments are converted into even smaller molecules, for example through dehydration. Through condensation and polymerization reactions, these are then recombined to form oil-like substances [122]. As a result, bio-oil is obtained usually composed of carbon (71–73 wt-%), hydrogen (7–8 wt-%), oxygen (10–11 wt-%), nitrogen (6–7 wt-%) and sulfur (<1 wt-%). The content of organic carbon in the biomass is decisive for the bio-oil yield. High amounts of protein increase the bio-oil yield, but also result in nitrogen containing compounds. These in turn lead to emission of harmful and legally limited nitrogen oxides (NO_x) during fuel combustion [110].

In Table 7, several studies using macroalgae for HTL and their reaction conditions are summarized as well as the resulting Higher Heating Value (HHV) of the obtained bio-oil.

Zhou et al. (2010) analyzed the water soluble fractions and concluded that in addition to the recovered bio-oil, value-added products like acetic acid and glycerol can also be obtained by HTL [124].

HTL is characterized by some limitations due to the high equipment investments and the costs for the catalyst; due to the latter, HTL processes at larger scale are only feasible when solutions regarding catalyst recovery and optimization are established. Sometimes it is even concluded that the application of HTL on macroalgae biomass is not feasible and that conversion techniques such as fermentation or extraction processes of valuable compounds (hydrocolloids, fucoidans, etc.) should be further researched rather than HTL of whole biomass [123]. Most likely, a cascade use where first high value products are removed from the macroalgae biomass and the rest is treated within a HTL process might be the most promising approach in terms of the overall product yield.

4.2.3. Biogas

The production of biogas by anaerobic digestion (AD) has, compared with the production of other products e.g., bio-oil by HTL or bio-ethanol by fermentation, the lowest investment cost, an easier process handling and a better net energy gain due to the use of all components that can be degraded. AD processes are typically based on the digestion of carbohydrates, proteins and lipids to methane (CH_4) and carbon dioxide (CO_2) in absence of oxygen. However, there are complex microorganism interactions involved not yet adapted to the molecular composition of macroalgae biomass [110].

For the AD of terrestrial biomass, operating conditions are already well-established. But marine biomass is characterized by a higher salt and sulfate content as well as different polysaccharides, which is why the conventional operating methods used in biogas digesters could not yet be successfully applied. Especially adapted microorganisms are necessary to digest these polysaccharides that are also different for each algae phylum (brown, green and red algae). Table 8 shows that methane yields in different studies vary significantly even for the same species due to different reaction conditions or pretreatment methods [125].

5. Commercial aspects

Macroalgae have been used as food and fertilizer for hundreds of years. Due to the trend towards healthy and natural products [33] and a still growing world population, the global macroalgae production tripled from ca. 10 Mt wet weight in the year 2000 to roughly 33 Mt in the year 2019 characterized by a value of ca. 13.3 billion US-$ [1,134]. This growth is mainly attributed to macroalgae from the aquaculture

Table 6
Process conditions and ethanol yields obtained from different macroalgae species and fermenting microorganisms.

Macroalgae species	Class	Microorganism	Ethanol Yield, g/g reducing sugar	Fermentation Time, h	Fermentation Temperature, °C	Source
Saccharina japonica (formerly *Laminaria japonica*)	Phaeophyta	*S. cerevisae* *E. coli K011*	0.41	24	30	[118]
Dictyota fasciola (formerly *Dilophus fasciola*)	Phaeophyta	*S. cerevisae*	0.32	72	20	[107]
Ulva lactuca (formerly *Ulva fasciata*)	Chlorophyta	*S. cerevisae*	0.40	48	30	[117]
		Pseudomonas sp.	0.22	48	30	[117]
Gelidium amansii	Rhodophyta	*B. custersii (CTC18154P)*	0.38	39	30	[119]
Sargassum spp.	Phaeophyta	*S. cerevisae*	0.16	48	30	[120]
Kappaphycus alvarezii	Chlorophyta	*S. cerevisae*	0.53	48	30	[121]

Table 7
HTL Reaction conditions and results for different macroalgae species.

Species	Bio-oil Yield, wt-%	Temperature, °C	Reaction Time, min	Bio-oil HHV, MJ/kg	Catalyst	Source
Ulva prolifera (formerly *Enteromorpha prolifera*), (green algae)	23.0	220–320	30	28–30	Na_2CO_3	[186]
	34.7	370	60	29.8	K_2CO_3	[187]
Saccharina latissima (formerly *Laminaria saccharina*), (brown algae)	19.3	350	15	36.5	None	[188]
	20.9	350	15	35.2	None	[123]
Fucus vesiculosus (brown algae)	22.0	350	15	33.4	None	[123]

Table 8
Different methane yields depending on pretreatment and process conditions in anaerobic digestion.

Species	Pretreatment	Residence Time (d), Temperature (°C)	Yield	Source
F. vesiculosus	Bead beating	21, 37	231 mL CH_4/g TS	[126]
	Hydrothermal (80 °C, 24 h)	20, 37	71 mL CH_4/g VS	[127]
	Hydrothermal (80 °C, 2 h)	22, 37	80 mL CH_4/g VS	[128]
	Enzymatic (50 °C, 5 h; Mix: cellulase, hemicellulose, pectinase, protease)	52, 37	49 mL_N CH_4/g VS	[129]
Sargassum sp.	Untreated	21, 35	48 mL CH_4/g VS	[130]
	Hydrothermal	21, 35	102 mL CH_4/g VS	
	Autoclave (121 °C, 30 min, 1 bar)	42, 37	541 mL CH_4/g VS	[131]
Laminaria sp.	Microwave (50 Hz, 560 W, 30 s)	38, 25	244 mL_N CH_4/g VS	[132]
	Enzyme (Cellulase; 37 °C, 24 h)	32, 35	225 mL biogas/g VS	[133]

sector, while harvesting macroalgae from wild resources stayed constant at around 1 Mt over the years contributing only 3.5 % to the global macroalgae production [1,68]. The main reason for this limited use are uncertainties regarding food safety of wild harvested macroalgae from areas where the coastal zone management is not as strictly enforced. Here the macroalgae are often contaminated with heavy metals like cadmium, mercury or arsenic [68]. In addition, there are often difficulties regarding ownership due to complicated laws and the protection and prevention of destruction of marine ecosystems [135].

5.1. Current use

Species that are the most cultivated on a global scale are the brown macroalgae *Saccharina japonica, Undaria pinnatifida*, and the red macroalgae *Porphyra* spp., *Pyropia tenera* (formerly *Porphyra tenera*), *Eucheuma* spp., *Kappaphycus alvarezii, Gracilaria* spp. and *Gracilariopsis longissima* (formerly *Gracilaria verrucosa*) [136]. Red (ca. 53 % of the total macroalgae aquaculture) and brown algae (ca. 46 %) are the most cultivated macroalgae worldwide while green macroalgae (ca. 1 %) are not extensively exploited yet [68].

The global macroalgae production by aquaculture is dominated by Asian countries with China, Indonesia and the Philippines at the top, producing around 14, 12 and 1.5 Mt/a (FW), respectively (Fig. 4) [1,68, 134]. The global distribution of macroalgae farming shows that the major producer are Asian countries with more than 99 % (based on fresh weight) of the global market [68].

The fast growth of the macroalgae market in Southeast Asia is mainly attributed to the rising demand of the hydrocolloid industry growing about 2–3 %/a [68]. A major driver in the production of macroalgae for carrageenan extraction is Indonesia, with a macroalgae farming output of around 12 Mt/a (FW) and the species *Kappaphycus alvarezii* and *Eucheuma* spp. being the most farmed for this purpose [134].

In contrast to the rest of the global market, macroalgae in Europe mainly come from wild harvesting (99 % of the total European macroalgae production). The macroalgae production (wild harvesting and aquaculture) goes up to around 330,000 t/a (FW) with a market value of 1.02 billion US-$, where European aquaculture, with roughly 5000 t/a (FW), only represents <1 % of the global aquaculture production. European wild harvesting on the other hand brings 18 % to global wild harvesting production [1,3,137].

In the European macroalgae market, France, Spain and Portugal are the leading producers of macroalgae-based hydrocolloids. Main importers of hydrocolloids are Germany, Spain, UK, France, Netherlands and Belgium [68]. The increasing demand for vegan food products where hydrocolloids are often used as a replacement for gelatin, is leading to higher imports of hydrocolloids and also a growing interest in hydrocolloid production. In Europe, Hydrocolloid production is mainly focused on the species *Gracilaria* spp. and *Gelidium* spp. for agar, *Ascophyllum nodosum* and *Laminaria* spp. for alginate and *Chondrus crispus* for carrageenan [138,139].

Globally, ca. 77 % of the produced algae biomass is used for human food and hydrocolloid production [140]. In contrast, European companies also going towards new process technologies and more innovative applications such as cosmetics, pharmaceuticals and biomaterials (Fig. 5). The biggest share of companies applies their biomass in human food (36 %), followed by cosmetics (17 %) and food supplements/hydrocolloid production (15 %), but also new process technologies such as bioremediation and production of biomaterials are applied [6].

Market prices for European marine macroalgae produced for food consumption can vary significantly depending on species and type of cultivation ranging from 22 US-$/kg DW e.g., for *Palmaria palmata* up to 296 US-$/kg DW e.g., for *Porphyra* spp. [6,141].

The highest product values of biomass applications are attributed to pharmaceutical and cosmetic products (>1000 US-$/kg) followed by food and feed applications (500–1000 US-$/kg). Energy and heat applications represent the lowest value products (<1 US-$/kg) [142–144]. To make macroalgae-based products competitive against fossil-based or conventional products, the product values need to be in these ranges.

Table 9 shows the market and product values as well as the expected compound annual growth rate (CAGR) of the potential industries for macroalgae applications and end uses as well as already commercially available macroalgae products. The highest economic value for hydrocolloids is in the pharmaceutical industry, where several hydrocolloid applications are already in use or being researched [145]. Another important driver is the growing demand for high-nutrition food and food-products e.g., due to an increasing health awareness. Still, macroalgae products for human consumption (whole biomass, hydrocolloids for gelling/texture properties) have by far the biggest share of the global market for macroalgae cultivation. In Europe, there are already running programs and initiatives to increase, improve and support macroalgae

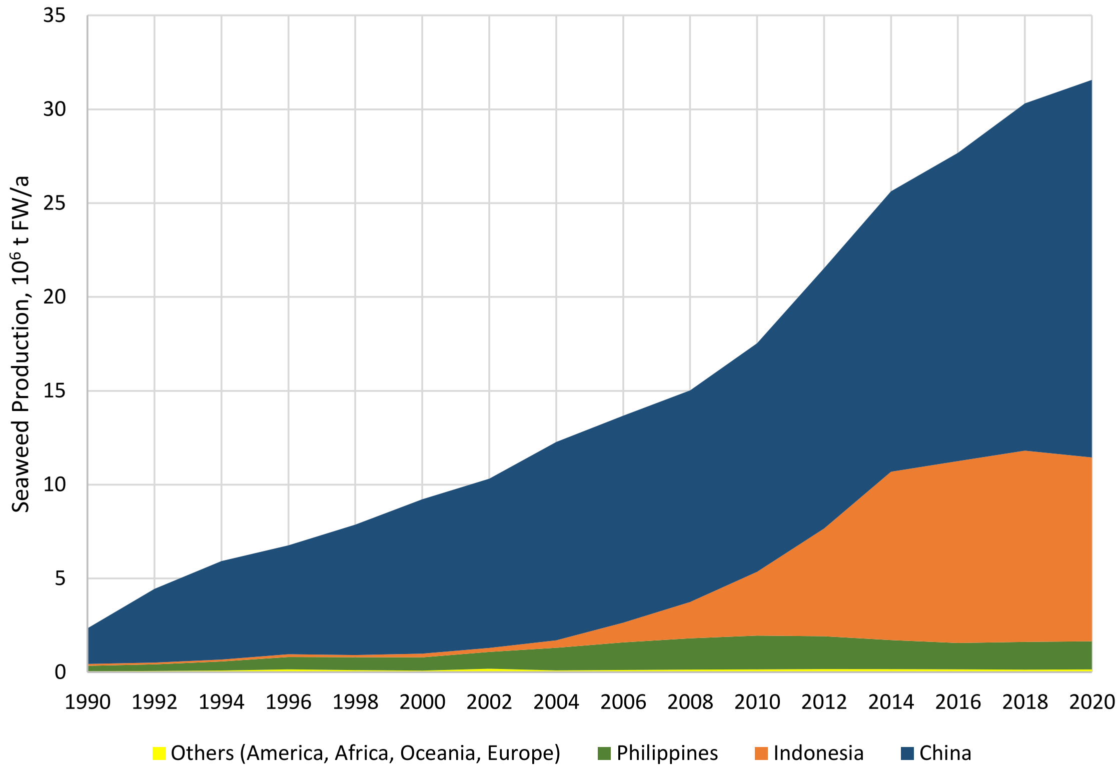

Fig. 4. Global macroalgae production of marine macroalgae [137].

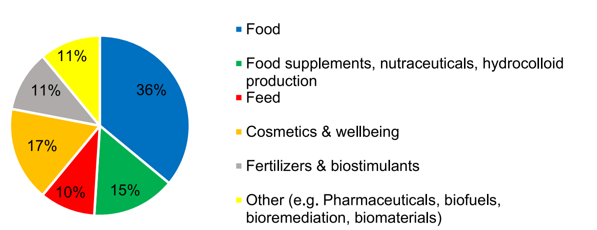

Fig. 5. Commercial macroalgae biomass applications in Europe by companies adapted from Araújo et al. (2021) [6].

farming and cultivation by implementing cultivation guidelines. The European Commission also published 23 actions to boost the algae sector as the demand for macroalgae is expected to be increase to 9.9 billion US-$ in the year 2030 [146].

5.2. Cost consideration

The economics of algae production is a relevant issue addressed so far only to a limited degree. But the few studies indicate that feedstock supply is the main bottleneck for a macroalgae based biorefinery [3, 162]. The value and yield of the final products to be sold potentially on the market constitute the main factors influencing the economic feasibility of algae production. Especially for the provision of biobased materials and other low cost products, an economic feasible cultivation strategy is necessary to develop processes that lead to biomaterials that can compete in the market, for example, with single use plastics. In this sense, one reason for the lower algae production in Europe compared to Asia are higher production costs due to higher labor costs [3,163]. For example, the economic feasibility of macroalgae production in six developing countries (Tanzania, Indonesia, Philippines, India, Solomon Islands and Mexico) has been investigated by comparing different cultivation systems. Since macroalgae farming is very labor intensive, it was shown that labor costs constitute here the biggest part of the variable costs. With 0.13 to 0.27 US-$/kg DW (depending on country, yield and size of production site), the labor expenditures are still very low in these developing countries in comparison with the European Union, where this value (based on a yield of 20 t DW and without harvesting) sums up around 1.77 US-$/kg DW [4,6,163].

The offshore macroalgae production is not yet extensively studied related to a reliable cost analysis. In terms of process/implementation feasibility, the sea lettuce *Ulva fenestrata* is suitable for a large-scale offshore cultivation on long lines in the Northern European hemisphere and is able to adjust adequately to the environmental conditions (storms, cold temperatures and wave action) given in this area [164]. A detailed economic analysis of such a multi-use offshore macroalgae production in the North Sea in combination with the use of offshore wind showed that such a multi-use linkage does not provide sufficient benefits to make a production of macroalgae more economically viable. Still, high personnel costs in particular avoid an economic viable production. Moreover, legal issues due to laws which are not clear on the applicability of offshore areas as well as property rights that are not well defined in these areas, represent major obstacles [165].

To reduce the high costs of macroalgae production, their nutrient bio-mitigation is an additional beneficial factor maybe considered while estimating macroalgae production costs. A model has been designed to calculate the monetary value of nutrient recovery by macroalgae cultivation that leads to an estimated value of 1.2–3.5 billion US-$. The introduction of nutrient trading credits (NTC), next to carbon trading credits (CTC), has been proposed as a strategy to regulate macroalgae prices and bring them closer to the low prices of non-natural resources or resources where only carbon credits are accounted for. This could be true since NTCs have a higher monetary value (recovery costs in wastewater treatment plants 10 to 30 US-$/kg N and 4 US-$/kg P) than CTCs (assumed carbon tax 0.03 US-$/kg) [142].

Table 9
Market value and expected growth rate of potential target markets and commercial macroalgae products.

Product	Species	Application	Market Value, million US-$	Growth rate (Expected CAGR), %
Markets by industry				
Biofuels	–	–	116,460 [147]	8.3 [147]
Cosmetics	–	–	341,100 [148]	5.1 [148]
Nutraceuticals	–	–	396,290 [149]	5.2 [149]
Pharmaceuticals	–	–	1,420,000 [150]	
Bioplastics	–	–	11,610 [151]	18.8 [151]
Hydrocolloids	–	–	11,230 [152]	6.0 [152]
Macroalgae product markets				
Seaweed (Dried)	*Kombu, Wakame, Dulse, Nori, Kelp*	Human food	14,000 [153]	9.1 [153]
Carrageenan	*Kappaphycopsis cottonii* (formerly *Eucheuma cottonii*), *Chondrus crispus*	Gelling thickening and stabilizing agent	872 [154]	5.4 [154]
Alginate	*Laminaria, Macrocystis, Ascophyllum*	Thickening and gelling agent, water retention	728 [155]	5.0 [155]
Agar	*Gracilaria, Gelidium*	Human food, gelling agent, biotechnology	324 [156]	5.1 [156]
Agarose	Produced from Agar	Biotechnology	105 [157]	4.4 [157]
Proteins	Red and green macroalgae	Animal feed, cosmetics	550 [158]	11.6 [158]
Phycobiliproteins	Cyanobacteria, Red macroalgae	Food colorant, dietary supplement, cosmetics industry, biotechnology, research (fluorescent detection reagent)	91 [159]	21.8 [159]
Fucoxanthin (allenic carotenoid)	Brown macroalgae	Cosmetic industry, nutraceutical, pharmaceutical	190 [160]	5.0 [160]
Fucoidan	Brown macroalgae	Nutraceutical industry	30 [161]	3.8 [161]

6. Conversion and fractionation of macroalgae biomass

To ensure the best possible value creation and industrial production from macroalgae biomass with the lowest possible environmental impact, an integrated, cascade biorefinery approach should be applied in which all components and residual materials are used. Various studies have shown that the generated residue after extraction of high-value polysaccharides like alginate or carrageenan still contains valuable substances such as proteins, other polysaccharides and minerals that are mostly treated as waste. Table 10 presents recent studies regarding macroalgae biorefineries.

Table 10
Biorefinery approaches using marine macroalgae as substrate.

Species	Products	Methodology	Source
Ulva lactuca	Water-soluble proteins and carbohydrates	Osmotic shock, enzyme treatment, pulsed electric field, high shear homogenization	[166]
	Proteins, Mineral rich sap, Lipids, Ulvan, Cellulose	Solid-Liquid-Extraction (alkaline, acidic, aqueous, solvent), heat treatment	[167]
	Ulvan, Mineral rich sap, Proteins, Biogas	Solid-Liquid-Extraction (aqueous, acidic, alkaline)	[168]
Ascophyllum nodosum, Ulva lactuca, Fucus vesiculosus, Laminaria digitata	Biogas, Bio-Crude, Bio-Char, aqueous fertilisers	Hydrothermal liquefaction	[169]
Laminaria digitata, Fucus vesiculosus	Laminarin, Fucoidan, Feed supplements	Solid-Liquid extraction (aqueous, acidic), Filtration	[170]
Laminaria digitata	Succinic acid, Bioenergy, Proteins, Lipids	Enzymatic hydrolysis, fermentation, anaerobic digestion	[171]
	Bioethanol, Proteins	Enzymatic hydrolysis, fermentation	[172]
	Fucoidan, Alginate, Bioethanol	Solid-Liquid-Extraction (acidic), Acid Hydrolysis, Fermentation	[173]
Gracilariopsis longissima (formerly *Gracilaria verrucosa*)	Agar, Bioethanol	Solid-Liquid extraction (aqueous), enzymatic hydrolysis, fermentation	[41]

Due to the many different constituents in macroalgae, the range of possible products is wide. For example, a biorefinery concept for the production of succinic acid by fermentation of *Laminaria digitata* has been studied being an important bulk chemical usually produced by the petrochemical industry. The remaining residues after the fermentation step are rich in proteins and lipids and are therefore applicable in the food and feed industry or for bioenergy production [171]. Especially *Laminaria digitata* has been studied for the simultaneous production of bioethanol with protein extraction from the residues. They achieved a potential ethanol yield of 78 % using enzymatic hydrolysis and fermentation. The residue was rich in proteins with high amounts of glutamic and aspartic acid which are amino acids of especial interest in fish feeding through their action as food attractants for many fish species [172]. Another study achieved promising results investigating *Ulva lactuca* for the production of mineral rich sap, proteins, ulvan and biomethane. Here the biomethane yield was twice as high after the extraction of ulvan and sap than in the initial biomass, showing that these components have an inhibitory effect on the biogas production [168].

Most studies regarding macroalgae products are targeted only into the direction of biobased fuels and bioenergy, but the separation of non-energy products from macroalgae and the subsequent production of bioenergy using the residues is a more promising approach improving efficiency, sustainability and added value of macroalgae processing and products (Table 10). The enrichment of target components with every process step increases product yields and saves chemicals for following process steps which is why only a cascade approach makes sense for the use of macroalgae as a bioresource.

In general, there are several approaches and research studies on biorefinery processes for the production of high-value and other products from macroalgae, but these are always dependent on the feedstock. Hydrothermal systems have demonstrated to be an advanced technology with wide potential to obtain valuable compounds from macroalgae in a sustainable way [174]. Other extraction and fractionation technologies that can be combined to achieve an integrated biorefinery approach include supercritical fluid extraction, ultrasound, microwave, enzymatic or pulsed electric field extraction, etc. [175,176]. Fig. 6 shows a cascade biorefinery process with proteins, biogas and bio-fertilizers/biostimulants as main products.

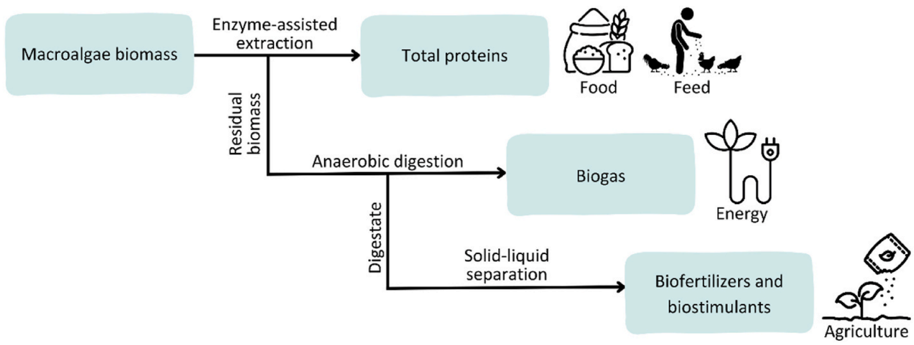

Fig. 6. Overview of a macroalgae biorefinery concept with the utilization of all biomass fractions.

7. Discussion and outlook

In this review, the properties of macroalgae and the individual groups are presented as well as established and potential applications being of commercial interest. In particular, the pharmaceutical and biomedical industry offer a wide range of macroalgae applications. The high potential of macroalgae as a biobased resource for the production of high-value products due to their versatile bioactive properties have been indicated several times. The production of lower value products such as biobased materials or energy from macroalgae is also promising, but the supply of sufficient biomass at reasonable prices is still a major bottleneck. The implementation of new technologies (e.g., process engineering and artificial intelligence) might be at least a part of the solution. Also, the productivity of selected macroalgae species should be further optimized.

As a resource, macroalgae have the advantage that they can be used for a wide range of products (energy, pharmaceuticals, cosmetics, food, feed, bio-based materials, research, etc.) and do not require agricultural land or large amounts of water. But still research in the field of off-shore macroalgae cultivation is needed to understand and assess such production concepts in a much better way.

Recent advancements in macroalgae biorefineries can significantly impact the landscape of sustainable resource utilization. The combination of advanced systems (e.g. hydrothermal, mechanical, artificial intelligence, etc.) allows the efficient extraction, processing, and conversion of macroalgae biomass within integrated biorefinery approaches. These approaches have not only enhanced overall productivity and efficiency but have also contributed to a more sustainable practices which align with environmental and ecological considerations. Therefore, macroalgae biorefineries are key players in the global transition towards a circular bioeconomy, aligning with the 2030 Agenda for Sustainable Development. The integration of macroalgae biorefineries into existing industries (e.g. agriculture, food, energy, etc.) could help expanding and diversifying conventional industries, influencing market dynamics and creating novel opportunities for businesses.

Multidisciplinary collaborations will play a pivotal role in advancing the development and impact of macroalgae biorefineries. Policy and regulatory changes can influence various aspects of the macroalgae industry, including environmental regulations, aquaculture and harvesting licenses, incentives for sustainable practices, and support for research and development. First approaches to pave the way to macroalgae biorefinery in Europe have been initiated by the introduction of guidelines for the cultivation of macroalgae [1,146]. These serve to ensure sustainable aquaculture, taking into account technical, legal, scientific and economic aspects.

CRediT authorship contribution statement

Sinah Kammler: Investigation, Writing – original draft, Writing – review & editing, Conceptualization. **Ana Malvis Romero:** Conceptualization, Investigation, Writing – original draft, Writing – review & editing. **Christin Burkhardt:** Writing – original draft, Conceptualization. **Leon Baruth:** Writing – original draft, Investigation. **Garabed Antranikian:** Supervision. **Andreas Liese:** Supervision. **Martin Kaltschmitt:** Supervision.

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