Modeling and Analysis

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Life cycle assessment of biomethane from offshore-cultivated seaweed

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Abstract: Algae are a promising source of industrial biomass for the future. In order to assess if aquacultured seaweed (macroalgae) could be considered an environmentally friendly source of biomass for bioenergy, life cycle assessments were performed for European countries, comparing methane as a biofuel from the anaerobic digestion (A) of whole seaweeds, (B) of alginate extraction residues, and (C) natural gas as a fossil fuel reference.

These results clarify that the sources of electricity and energy used to heat the anaerobic digesters have an important impact. Recycling of materials and use of greenhouses at the nursery stage also allow environmental improvements for system (A). Ecodesign can make algal biomethane competitive in several categories compared to natural gas: a decrease of 21.9% and 54.2% in greenhouse gas (GHG) emissions and 58.6% and 68.7% in fossil depletion for systems (A) and (B), respectively, decrease in ozone depletion, and last but not least, improvement in the marine eutrophication index for system (A). For system (B), benefits are more arguable and dependent on the allocation. To conclude, seaweed could become competitive with terrestrial feedstock for biofuel production in the near future. © 2012 Society of Chemical Industry and John Wiley & Sons, Ltd

Keywords biofuel; seaweed; life cycle assessment (LCA); alginate; biogas; *Saccharina latissima*

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Introduction

Since the first concerns about the lack of fossil resources, biofuel production has been increasing worldwide.[1] This expansion leads to many questions concerning the associated social and environmental impacts, especially on land-use competition and on pollution transfers from greenhouse gas (GHG) emissions to other environmental impact categories (e.g. eutrophication,[2,3] resource depletion, ecotoxicity, biodiversity loss,[2] acidification, ozone depletion, and human toxicity[3]). When compared to terrestrial crops traditionally used for food and feed, the alternative of an algal feedstock for biofuel production seems to be very promising.[4–6] In recent years, microalgae have received considerable attention concerning energetic applications,[7] whereas few studies have dealt with the environmental impact of marine macroalgae (seaweed) production as feedstock for bioenergy.[8,9]

Life cycle assessment (LCA) is an efficient tool for quantifying environmental impacts of bio-based materials. LCA studies applied to biofuels from microalgae show that this feedstock leads to environmental impacts in the same range of magnitude as terrestrial biofuels, either through biodiesel[10] or biomethane production.[11] Several ways of improvement could lead to a significant decrease in their environmental impact. Nevertheless, fertilization, harvesting systems, and complex cultivation infrastructures (like raceways or photo-bioreactors) are still bottlenecks that need to be overcome.[12] Because seaweeds are macro-organisms, they can be grown and harvested offshore in a fairly straightforward manner. Compared to microalgae, seaweed aquaculture requires less sophisticated cultivation and harvesting systems. It is also a very interesting means of decreasing eutrophication in coastal areas and, furthermore, does not impact arable land and freshwater resources.[13] Many authors assume that macroalgae could possibly become a new feedstock for bioenergy in the future,[4,5,13] either through bioalcohol, biodiesel, biomethane, or thermochemical treatments. Two LCA studies have been carried out on bioenergy from macroalgae. Aresta *et al.* considered onshore-cultivated macroalgae, under controlled conditions in ponds, with CO_2 and nutrient enrichment.[8] No definite results are provided for anaerobic digestion. Another LCA study dealt with invasive seaweeds in a lake.[9] This studied system is hardly comparable with ours as it considers seaweed feedstock to be waste, whose harvesting avoids the spread of herbicides in water and artificial cultivation systems.

The present study deals with offshore-cultivated macroalgae. The main advantage of this system when compared to that of wild algae is a high biomass density. When compared with onshore facilities under controlled conditions, however, there is no possibility for fertilization offshore, leading to a decrease in biomass production yields. On the other hand, there is no electrical input during cultivation (neither paddlewheel nor pumps) out from the nursery, decreasing energy demand in the production system. Furthermore, competition for land or food is avoided, as biomass from the sea is negligible in world food consumption compared to terrestrial crops (only 15.8 million tonnes of aquatic plants produced in 2008).[14]

The present study is dedicated to the environmental assessment of the production of biogas from offshore-cultivated macroalgal feedstock, as a prospect for European countries. Some LCA studies applied to biomethane production have shown the interest of this biofuel.[15] The goal of this study is to improve production processes and to determine whether offshore-cultivated seaweeds are a more environmentally friendly feedstock for fuel production than natural gas. To this end three systems are considered:

(A) Methane as a fuel from the anaerobic digestion of untransformed whole seaweeds.
(B) Methane as a fuel from the anaerobic digestion of alginate extraction residues.
(C) Natural gas from EcoInvent database as a fossil fuel reference.

At first, biomethane production chains from macroalgal-dedicated feedstock were analyzed (A). As of today, growing seaweed for energy purposes only is not financially profitable.[5] Therefore, at the present time, scenario (A) is only prospective. For this reason we also focused on existing high-value macroalgal biomolecules (alginates) for which industrial extraction residues can be valued. This type of feedstock from biorefinery residues is more commonly used for the production of bioenergy from biomass, referred to as system (B).[16] It is based on future biorefinery requirements,[17] linking production of fuels, energy, and value-added chemicals via the processing of biomass on a single site. In the next section, the systems definition and the associated inventory are described in detail. Results are then presented. The two theoretical scenarios (A) and (B) were analyzed by contribution analysis, and results are discussed. In addition, several assumptions were tested for upgrading scenarios and a comparison of the two systems involving macroalgal biomethane production with natural gas is given. Upgrading scenarios take into account either technical improvements or eco-design choices. The goal of these tests was to determine the margin of improvement that can be expected for macroalgal feedstock, and also to determine the effect of different options on environmental performance.

System definition and inventory

Goal and scope

The objective of this study was to evaluate potential environmental impacts caused by methane production from macroalgae and its combustion in an engine. According to this aim, the functional unit was defined by a 1-km trip with a gas-powered car. The ReCiPe method[18] was used with a hierarchist perspective using the EcoInvent v2.2 database[19] and SimaPro 7.3 software to carry out the impact assessment. In order to conform to the cradle-to-grave approach of LCA,[20] the inventory included all steps of biomass cultivation and harvesting, its transformation to biomethane provided at a gas station, and its combustion. In the case of scenario (B) dealing with macroalgal residues, the inventory also included steps involving extraction and waste-water treatment in the biorefinery. Both the construction and dismantling of facilities and the extraction and transportation of resources were taken into account.

Today, hydrocolloids are still the main commercial seaweed extracts, despite recent attention given to other biologically active compounds (food flavoring, colors or nutrients) and the competitive production of cheap biomolecules from terrestrial crops.[21] We chose to focus on kelp, which belongs to the Laminariacea family, as it is the most abundantly produced seaweed genus in the world,[14] as well as being the main source of alginates.

We concentrated on food-grade alginates, being the most important market used for thickeners, stabilizers, gel formers, or film-forming agents.[21] The species *Saccharina latissima* was chosen for its interesting alginate content. In the present study, kelp was cultivated on long-lines in a coastal environment, following plantlet production in a nursery. Seaweeds were harvested and then transformed into biogas in an anaerobic digestion plant, using either the entire seaweed (A) or solid macroalgal residues from alginate extraction (B). Figure 1 shows an overview of the whole system, from the seaweed cultivation to the use of biomethane as a fuel.

For the extracted by-products in system (B), a proper substitution was not possible due to lack of data: to our knowledge alginate extraction has never been described in a published LCA study. Because the main function of the system was to produce alginate for its high market value (97.9% of total gains), a financial allocation was calculated. The substitution method was used to account for anaerobic digestion by-products used as fertilizers (phosphate, ammonium, and

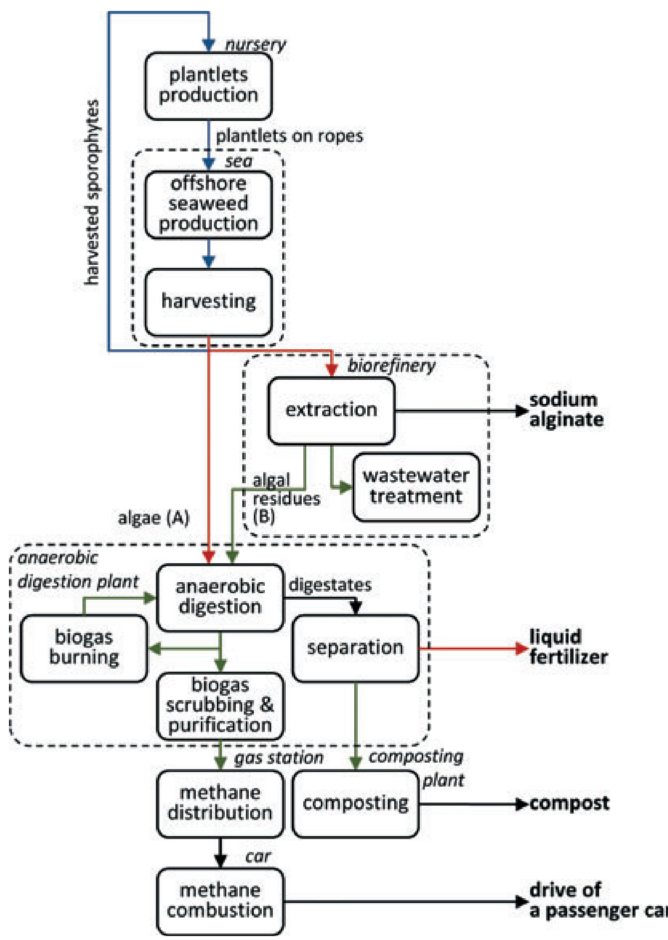

Figure 1. Overview of the biomethane production system (A) from whole seaweeds cultivated in the open ocean and (B) from residues of alginate extraction. Blue, red and green arrows stand for transportation by boat, truck and pipe respectively.

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Table 1. Hypotheses used for the financial allocation (dm=dry matter).

	Product	Product price		Quantity produced		Percent of impact allocated
		Amount	Unit	Amount	Unit	
(A)	Macroalgal biomethane	55.5[22]	$USD.MWh^{-1}$	1.43	$kWh.kg_{dm\ algae}^{-1}$	99.1%
	Compost	5.9[23]	$USD.t^{-1}$	0.13	$kg.\ kg_{dm\ algae}^{-1}$	0.9%
(B)	Sodium alginate powder (90% purity)	12.0[21]	$USD.kg^{-1}$	0.42	$kg.\ kg_{dm\ algae}^{-1}$	97.9%
	Macroalgal biomethane	55.5[22]	$USD.MWh^{-1}$	1.41	$kWh.\ kg_{dm\ algae}^{-1}$	1.5%
	Compost	5.9[23]	$USD.t^{-1}$	4.91	$kg.\ kg_{dm\ algae}^{-1}$	0.6%

potassium dissolved in the leachates). However, substitution of compost produced from the solid part of the digestates was not considered. Compost can be regarded both as a product and as a result of waste treatment. The production of waste which is usually composted would not be avoided using the production we considered.

Hypotheses used for the allocation are detailed in Table 1. For scenario (A), impacts were allocated to macroalgal biomethane (99.1%), and to the compost (0.9%) from the cultivation step to the biomethane distribution and to the composting stage of the solid digestates. Only biomethane combustion impacts were totally dedicated to the biomethane produced. For scenario (B), impacts were mainly allocated to alginate powder (97.9% *versus* 1.5 and 0.6% for macroalgal biomethane and compost, respectively). As for scenario (A), only the biomethane combustion impacts were totally allocated to the macroalgal biomethane.

Process inventory of the initial scenario

The analyzed process chain referred to a hypothetical system, based on basic and present-day knowledge and techniques as well as on extrapolation from semi-industrial scale. In the case of biomass cultivation, data came from a semi-industrial macroalgae production site. Data for alginate extractions were gathered at pilot-scale. Anaerobic digestion was sized up to industrial scale by an anaerobic digestion plant designer on the basis of laboratory experiments. Standard rules were considered for material transportation[19] and substructure replacement (30-year lifespan for plants and replacement of electrical facilities every 10 years). After building, dismantling, and replacing facilities, land-filling was chosen for concrete, mineral wool, polypropylene, polyethylene, polyethylene terephthalate, polyvinylchloride, bricks, cement fibers, steel, and iron. Electricity from the European production mix and waste water treated in a class 5 waste-water treatment plant were considered in the EcoInvent database.[19]

Data used in the inventory for biomass production were measured under a temperate climate. Two cycles of seaweed production per year were considered, assuming some improvements in seasonality management, according to the possible anticipation in the seaweed outplanting time.[24] In the main seaweed-producing countries, the tropical climate enables the biomass to be sundried to 30% moisture content before using an air-forced dryer to reach 90% dry matter (dm). Under a temperate climate, such as the one considered in this study, the pre-drying process under the sun is not feasible at industrial scale. No drying was considered, and consequently storage was not possible. The use of facilities therefore lasted for half of the year concerning the nursery, the biorefinery, and the digestion plant (harvesting duration), and for 75% of the year concerning the offshore facilities (offshore growth duration). There would be no constraint of seasonality under a tropical climate since cultivation could occur all year round and biomass could easily be sun-dried for storage.

Values for the inventory are summarized in Table 2. Matter and energy consumption are described for all steps of the process. As results were expressed per dry matter of algae, more anaerobic digestion facilities were required in scenario (B), due to the addition of cellulose during the process.

Plantlet production onshore

Seaweeds were cultivated on ropes to limit harvesting constraints. A step of cultivation in the nursery ensured seaweed fixing on the ropes. As *saccharina latissima* cannot be grown by vegetative propagation, there needed to be an alternation of generations in the nursery.[27] Two main steps occurred: spores

Table 2. Matter and energy consumption for macroalgal biomethane production (per kg of dry mater of macroalgae).

	Nature		Quantity			Unit per kg of algae (dm)	Lifespan (years)
	Process	Compound	Raw algae (A)		Extraction residues (B)		
Offshore cultivation facilities	Chain cable	Steel		1.40 x10^{-2}		kg	20
	Buoys	Moulded polypropylene		4.23 x10^{-3}		kg	10
		Rigid foam polyurethane		1.36 x10^{-5}		kg	10
	Ropes (20mm)	Weaved polyamid		1.61 x10^{-2}		kg	10
	Concrete blocks	Concrete		0.50		kg	20
		Glass Fibers		1.02 x10^{-2}		kg	–
Nursery facilities	Ponds	Concrete, for foundations		4.07 x10^{-6}		m^3	20
		Cement		2.17 x10^{-7}		m^3	20
		Concrete blocks		1.76 x10^{-3}		kg	20
	Building	Agricultural shed		2.89 x10^{-5}		m^2	50
Biorefinery facilities	Building and facilities	Chemical plants, organics		1.51 x10^{-14}		p	50
Anaerobic digestion facilities	Building	Concrete blocks	3.62 x10^{-3}		7.26 x10^{-3}	kg	30
		Concrete	4.47 x10^{-7}		8.99 x10^{-7}	m^3	30
		Concrete, for foundations	8.93 x10^{-7}		1.78 x10^{-6}	m^3	30
		Extruded polyvinylchloride	7.79 x10^{-5}		1.57 x10^{-4}	kg	10
	Pump, boiler, agitator	Unalloyed steel	3.17 x10^{-5}		4.82 x10^{-5}	kg	10
	Agitator	Chromium steel	2.40 x10^{-5}		4.82 x10^{-5}	kg	10
	Pump, pipe	Extruded polyethylene high density	4.92 x10^{-6}		6.92 x10^{-6}	kg	10
Plantlet cultivation in the nursery	Fertilizer	Ammonium nitrate		8.03 x10^{-2}		g N	–
		Sodium phosphate		3.24 x10^{-2}		g Na_3O_4P	–
	Mineral solution (Provasoli)	EDTA		1.77 x10^{-2}		g	–
		$FeCl_3$ (40%)		2.68 x10^{-3}		g	–
		Chemical inorganics		2.66 x10^{-3}		g	–
		Anhydrous boric acid		1.55 x10^{-2}		g	–
	Small ropes (3mm)	Weaved polyamid		7.17 x10^{-3}		kg	–
	Circulation pump	Electricity		38.5		Wh	–
	Fluorescent lamps	Electricity		199.4		Wh	–
	Sparger	Electricity		65.9		Wh	–
Water treatment in the nursery	Filtered seawater	Water		4.6		L filtered seawater	–
	Lamp UV	Electricity		8.7		Wh	–
	Circulation pump	Electricity		1.4		Wh	–
	Sand filter pump	Electricity		27.6		Wh	–
Harvesting	Operating barge	Diesel		1.52 x10^{-2}		kg	–
Alcoholic pretreatment step (B)	Crusher	Electricity	–		293	Wh	–
	Strainer	Electricity	–		98	Wh	–
	Still[25]	Electricity	–		339	Wh	–
		Steam	–		5821	Wh	–
	Water	Freshwater	–		73.3	L	–
Alginate extraction (B) – acid lixiviation	Acid	HCl 0.1 M	–		22.29	Kg	–
	Strainer	Electricity	–		391	Wh	–
	Water	Freshwater	–		9.78	L	–

Table 2. Continued.

	Nature		Quantity		Unit per kg of algae (dm)	Lifespan (years)
	Process	Compound	Raw algae (A)	Extraction residues (B)		
Alginate extraction (B) – alkaline extraction	Blender	Electricity	–	1389	Wh	–
	Heating	50–60°C	–	2811	Wh	–
	Alkalin	Na_2CO_3 (1.5%)	–	58.2	kg	–
	Filter press [26]	Electricity	–	1315	Wh	–
	Cooling	Room temperature	–	3762	Wh	–
	Filter aid	Cellulose	–	2.44	kg	–
	Water	Freshwater	–	44.0	L	–
Alginate extraction (B) – rectification	Blender	Electricity	–	66	Wh	–
	Acid	HCl 2M	–	1601.6	kg	–
	Strainer	Electricity	–	1027	Wh	–
	Cooling	0°C–10°C	–	166	Wh	–
	Water	Freshwater	–	16.7	L	–
Alginate extraction (B) – conversion to sodium alginate	Alkalin	Na_2CO_3	–	0.12	kg	–
	Convective dryer [26]	Electricity	–	6432	Wh	–
Anaerobic digestion	Oil	Lubricating oil	8.98×10^{-5}	1.19×10^{-4}	kg	–
	Global electrical consumption	Electricity	33.3	42.2	Wh	–
	Digester's heating	Home consumed biomethane	393.3	629.5	Wh	–
Biomethane purification	Facilities	Facilities, chemical production	1.49×10^{-1}	2.54×10^{-1}	kg	–
		Electricity	5.93	5.35	Wh	–
	Water losses	Water	7.98×10^{-2}	8.84×10^{-2}	m^3	–

were collected from wild, harvested sporophytes and the plantlets resulting from the collected spores were cultivated in ponds. All data concerning plantlet production in the nursery were based on a facility producing algae for food, in accordance with techniques described in the literature.[28]

Spore production lasted for one day, and only required a few inputs to be carried out. After cutting fertile zones on the sporophytes, along with three repeated washings, fertile pieces of algae were subjected to a hydric stress. Following this, a solution was recovered from the stressed pieces, allowing the insemination of small ropes in cultivation ponds.

Plantlet production lasted for one month and many inputs were required. To enable growth in concrete ponds, mineral fertilizers, fluorescent lamps, spargers for bubbling, and booster and circulation pumps were required. The control of water temperature in the ponds was not taken into account, as it does not go up to 15°C during the winter and starting spring times in most European countries. In order to control the photoperiod (18 h per day on average), the nursery was in a closed building (agricultural shed). Ponds were built in concrete. Pumped seawater was filtered and then treated under ultraviolet lamps before being used for plantlet cultivation in ponds. Pipes, filters, and tanks for water filtration were also included in the inventory. Spore production is particularly sensitive to bacterial contamination, so the filtered seawater was also treated in an autoclave before being used to induce sporulation.

Open-ocean cultivation and harvesting

Macroalgae were cultivated by being tied to anchored floating lines in a coastal environment. One long-line raft unit is described in Figure 2, and details are provided in Table 2. The system consisted of 150-m long culture ropes which were tied to 10-m long structural ropes. These were anchored to the bottom using concrete blocks at each corner as well as at 50-m intervals. The culture ropes were maintained 2 m below the surface. Chain cables and polyurethane buoys were used to absorb swell effects.

The length between two culture ropes was kept relatively low (2 m), corresponding to a site with low streams. Ropes

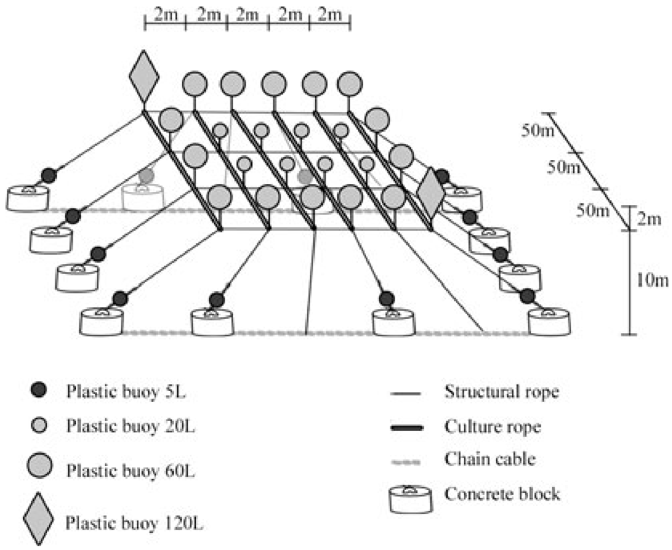

Figure 2. Schematic representation of a long-line raft.

were made from polyamide, chain cables from steel, buoys from polypropylene (with expanded polyurethane inside for the smallest ones), and blocks from fibrous concrete. Macroalgae were able to attach to the longline raft with the help of small polyamide ropes (1.25 m of small ropes wound around every meter of culture rope).

During its growth, seaweed absorbs nutrients. In the case of *Saccharina latissima*, uptake reached 21 g $N.kg^{-1}$ dm, 4.5 g $P.kg^{-1}$ dm. Due to the eutrophication context of coastal ecosystems,[29] this uptake has a positive effect by removing pollution.[13] This could be a potential solution against excess anthropogenic nutrients. CO_2 was fixed though photosynthesis, but the CO_2 net balance was assumed to be equal to zero, since it was not stored but released into the atmosphere when the algal biomethane was burnt. However, loss of biomethane in the anaerobic digestion plant and in the gas station was taken into account.

According to Peteiro and Freire, in the Atlantic Ocean under a temperate climate, the productivity of wet biomass in spring cultivation on long lines reaches an average of 8.95 $kg.m^{-1}$ after four months.[24] This value is largely dependent on the time of outplanting, ranging from 6.2 to 11.7 $kg.m^{-1}$.

Offshore substructure transportation and biomass harvesting for breeding or for industrial treatments were carried out using the same boat. This was based on data for a barge from the EcoInvent database, but with a fuel consumption of 1.1×10^{-1} $L_{diesel}.km^{-1}.t^{-1}_{transported}$.

Extraction of macroalgal high-value biomolecules: alginate

In system (B), biomass was treated in a biorefinery straight after being harvested for food-grade extract of sodium alginate production. All reactants and energy inputs used at every step of the extraction are described in Table 2.

The biomass was first washed and crushed. Next it was treated with alcohol as an alternative to the use of formalin, avoiding the production of a colored alginate powder. Formaldehyde has been classified as 'carcinogen for humans' by the International Agency for Research on Cancer (IARC, belonging to the World Health Organization). Thus we modeled a process avoiding its use, as it was technically feasible, and as dangerous substances tend to be replaced with time. The alcohol was used assuming a high recycling rate.

After acid lixiviation with HCl and a first dewatering on a vibrating sieve, an alkaline extraction was carried out with an Na_2CO_3 solution after 3 h of blending. Alginates were thus solubilized with the sodium cations. After a second dewatering, using a filter press and cellulose powder as a filter aid (considered as thermo-chemical pulp from a paper production plant), extraction residues were recovered for the anaerobic digestion step, and the solution was cooled at ambient temperature. From that point, acid precipitation by HCl with blending was operated in order to obtain a gel of precipitated alginic acid. A final dewatering was performed on a vibrating sieve, at a cold temperature (4°C). A last addition of sodium carbonate allowed the conversion to sodium alginate, which was then dried in a convective dryer without recovery of heating (the most common drying technique used in the agro-food industry).[26] This process led to an alginate content of 37.2% of fresh matter.

Natural gas burnt in a large-scale industrial furnace was chosen for the heating process. An absorption chiller operated by heat from natural gas was chosen for the cooling process. Neither energy transfer coupling nor loss of calories was considered.

Electricity consumption was obtained at pilot-scale, by measuring apparent power consumed, except for the still, the filter-press, and the dryer. Bibliographic data used for the still electrical consumption was 33 $kWh.t^{-1}_{alcohol}$ and 567 $kWh.t^{-1}_{alcohol}$ for electricity and heat requirements respectively.[25] Electricity used for the filter press was 15 $kWh.t^{-1}_{removed\ water}$ and 920 $kWh.t^{-1}_{removed\ water}$ in case of the convective dryer without heat recovery: the most common drying process.[26] These data correspond with the lowest values of usual practices.

Substructures were considered as an organic chemical plant, given that 666.7 $t_{dm}.yr^{-1}$ can be treated in a 1000 m² chemical plant.

Biomethane and fertilizer production by anaerobic digestion

Anaerobic digestion and biogas purification were sized up based on state-of-the-art engineering and expert knowledge involving urban sludge treatment applications. In accordance with current industrial practices, we aimed for a total production capacity of 2MW. According to this sizing, completely stirred tank reactors with a volume of $8.17x10^3$ utile were designed, with replications in order to reach this capacity. Assuming a cultivation on 50% of the total area, the corresponding cultivation sites measured 9524 ha and 4777 ha for scenarios (A) and (B), respectively. This amount is almost twice as low for system (B) due to the addition of cellulose to the process. The hypotheses used to size up the plant and the results of this modeling are written in Table 3. They are expressed per mass of substrate with the exception of fertilizer production which is expressed per mass of dry algae.

The biomass characteristics and anaerobic digestion performance were based on experimental data (Jard *et al.*, unpublished). The biomasses studied were wild fresh *Saccharina latissima* harvested in July (except for the methane yield, measured on dried macroalgae), and fresh alginate extraction residues. The anaerobic digestion performances were based on the biochemical methane potential (BMP), except for the methane yield from untransformed macroalgae, where data from a semi-continuous reactor fed with dried macroalgae was available. The value of 241 L CH_4. kg^{-1}_{vm} (volatile matter) for untransformed macroalgae was consistent with literature values: biogas production yield in

Table 3. Macroalgal biomass characteristics, anaerobic digestion performance and sizing of the biomethane production plant (dm, fm, cod, vm, omd, and omi stand for dry matter, fresh mater, chemical oxygen demand, volatile matter, organic matter degraded and organic matter introduced respectively).

	Parameter	Unit	Value: Untransformed macroalgae	Value: Alginate extraction residues
Biomass characteristics	% Dry Matter	$t_{dm}.t^{-1}_{fm}$	9.74%	13.8%
	% Organic Matter	$t_{om}.t^{-1}_{dm}$	60.60%	96.0%
	COD/VM	$kg_{cod}.kg^{-1}_{vm}$	1.07	not used
	Density	$t_{fm}.m^{-3}$	0.55	0.96
	Nitrogen content	$kg_N.t^{-1}_{dm}$	21.3	3.2
	including NH_4^+	$\%_{total\ N}$	77.8	77.8
	Phosphore content	$kg_P.t^{-1}_{dm}$	4.5	0.01
	including PO_4^{3-}	$\%_{total\ P}$	44.4	5.2
	Potassium content	$kg_K.t^{-1}_{dm}$	110.0	0.5
Anaerobic digestion performance	Degradation rate	$t_{omd}.t^{-1}_{omi}$	87%	13%
	Methane yield	$Nm^3_{CH4}.t^{-1}_{dm}$	146	51
	Methane content	$\%_{CH4}$	55%	49%
Fertilizer production	Ammonium sulfate	$g_N.\ kg^{-1}_{dm\ algae}$	16.3	1.3
	Single superphosphate	$g_{P2O5}.\ kg^{-1}_{dm\ algae}$	8.0	0.0
	Potassium chloride	$g_{K2O}.\ kg^{-1}_{dm\ algae}$	116.2	0.6
	Compost	$kg.kg^{-1}_{dm\ algae}$	0.1	4.9
Digesters characteristics	Retention time	day	43	43
	Loading rate	$kg_{dm}.m^{-3}.day^{-1}$	2.3	3.2
	Biogas home consumption	%	26.7	40.3

semi-continuous reactors for *Saccharina latissima* ranged from 220 to 271 L $CH_4.kg^{-1}{}_{vm}$ depending on the season.[30]

For the residues, the organic matter introduced into the system enabling biomethane production was firstly macroalgae, but also the organic filter aid used in the biorefinery for alginate extraction: powder of cellulose, explaining the differences in methane potential between the two substrates. NH_4^+/N ratio in the alginate extraction residues could not been measured, thus it has been approximated by the NH_4^+/N ratio from the untransformed macroalgae.

The electricity consumption represented 8% of the energy produced within the plant. Digesters' heating to a mesophilic range of temperature came from biogas home consumption.

We considered that all the ammonium, phosphate, and potassium oxide contained in the liquid phase of the digestates had the fertilizing value of the equivalent mineral fertilizer: ammonium sulfate in the case of nitrogen, single superphosphate in the case of phosphorus and potassium chloride in the case of potassium. Rates of mineralization were defined using the degradation rate of carbon organic matter.

The solid fraction of the digestates was composted. The compost production was considered equivalent to terrestrial feedstock composting, avoiding pick-up of waste biomass. We considered a mineralization rate of 0.149 kg $C.kg^{-1}{}_{compost}$ according to the EcoInvent documentation.

The loading rate of the digesters was 2.3 $kg_{dm}.m^{-3}$ per day for the untransformed macroalgae.[30] A low loading rate was selected to allow a more effective degradation rate in the organic matter. The corresponding retention time was 43 days. Due to lack of data this retention time was also applied in the case of extraction residues.

Biomethane distribution and use

The purification system was sized up including gas compression, gas upgrading, and gas drying operations. After raw biogas compression, a scrubber vessel was designed for water scrubbing (absorption of CO_2 and other soluble gases into water). A flashing tank and a stripper vessel were designed for regeneration of the process water. Facilities were based on chemical production facilities from the EcoInvent database. Electricity consumption included a compressor, water pump, chiller, chilled water recirculation pump, stripping air blower, and drier. Odour reduction filters, odorization equipment, gas flare, and gas vent were not included.

After purification to 96% biomethane yield, biogas was compressed within a gas station. This step was described using the EcoInvent database, considering the macroalgal biomethane equivalent to natural gas. The data from the EcoInvent database was also used for natural gas and biogas combustion.

Main results of LCA applied to macroalgal biomethane production and discussion

Contribution analysis in the initial scenario

The results of LCA applied to the initial scenario for the production of biomethane from macroalgae in order to drive a passenger car for 1 km are shown in Figure 3(a) regarding untransformed macroalgae and in Figure 3(b) regarding macroalgal residues.

The graph in Figure 3(a), highlights the importance of macroalgae cultivation techniques in order to ensure the environmental performance of the production system. Within the cultivation impacts, the analysis shows that the operations which occur in the nursery play an important role (28.4% of the impacts on average). This is mostly due to electricity consumption. The main facilities accounting for these impacts are the fluorescent lamps used to grow the plantlets. The small polyamide ropes also play an important role.

Facilities and substructures also induce strong impacts, especially offshore facilities (27.1% of the impacts on average). This is principally due to the concrete blocks anchoring the cultivation system, as well as the steel used for the chain cable. The nursery substructure has important impacts on land-use occupation (58.4% and 56.5% for agricultural and urban land, respectively). Nevertheless this impact should be considered less important than the others because land occupation is very limited in this kind of system when compared to terrestrial biomass production systems. Moreover the average impact of the nursery substructure is only 9.5%.

Operations during offshore cultivation would represent 9.5% of impacts on average if only considering the harvesting step. Because seaweeds absorb nutrients during their growth, a strong positive impact on the environment is accounted for marine and freshwater eutrophication.

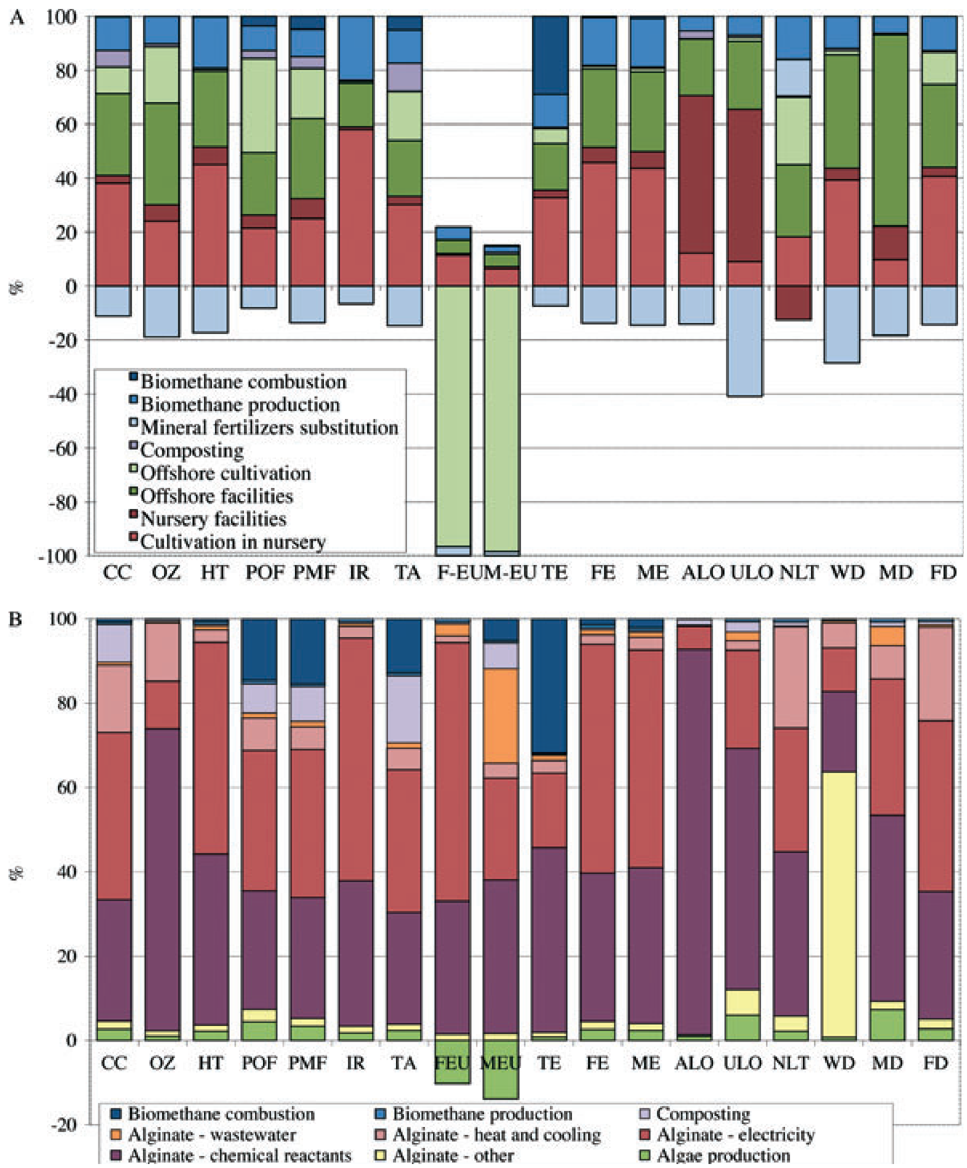

Figure 3. Environmental impacts of biomethane production from (a) untransformed macroalgae and (b) macroalgal residues from alginate production. [CC: climate change, OZ: ozone depletion, HT: human toxicity, POF: photochemical oxidant formation, PMF: particulate matter formation, IR: ionizing radiation, TA: terrestrial acidification, F-EU: freshwater eutrophication, M-EU: marine eutrophication, TE: terrestrial ecotoxicity, FE: freshwater ecotoxicity, ME: marine ecotoxicity, ALO: agricultural land occupation, ULO: urban land occupation, NLT: natural land transformation, WD: water depletion, MD: metal depletion, FD: fossil depletion]

Thus offshore cultivation operations become beneficial for the environment (-2.4% of the impacts on average). A methodological limitation in this analysis is that phosphate catchment is only taken into account in freshwaters impacts within the ReCiPe method. Thus the positive impacts of the phosphate removed offshore are accounted for the 'freshwater eutrophication' impact category instead of 'marine eutrophication'.

The impacts due to biomethane production itself are relatively low (11.7% of the impacts on average). Nevertheless it is important to note that home consumption of biogas is not represented on the graph. This heating corresponds to 26.7% of the algae production. Therefore the same proportion of pollution due to cultivation techniques is in reality indirectly due to anaerobic digestion. The substitution method used to account for anaerobic digestion by-products (phosphate, nitrate and potassium dissolved in the leachates) also allows positive impacts in avoiding the production of mineral fertilizer.

The impacts due to biomethane combustion are low (2.5% of the total impacts on average). The main negative impacts occur for terrestrial ecotoxicity (28.9% of the impacts in this category), but is limited for the rest, with less than 1% of the impacts in 14 impact categories.

The graph in Figure 3(b) highlights the importance of the macroalgal transformation steps within the biorefinery for alginate production. The most important pollution factor is energy consumption: electricity represents 34.0% of the impacts on average. This is principally due to the convective dryer and the filter press, as well as the blenders used for several hours (alkaline extraction). The chemical reactants are important parts of the environmental impacts too (40.2% of the impacts on average), especially cellulose powder used as a filter aid and hydrochloric acid, used in large amounts in the extraction process.

Biomass production accounts for only 1.1% of the impacts on average, ranging from -13.9% (beneficial for the environment) to 7.3% of the total impacts. Biomethane production is almost negligible, too, with less than 1% of the total impacts for each impact category. As for scenario (A), it is again important to note that the impacts of biomethane production are partly shown as impacts of biomolecules and biomass production due to home consumption of biogas. In this case, home consumption is a lot higher (40.3%) than for scenario (A).

Impacts of the substitution to mineral fertilizers becomes negligible too compared with scenario (A) not only due to the allocation, but also because concentration of nitrogen, phosphorus and potassium are very low. They are leached during the extraction process.

Substructure and facility impacts stay negligible despite the fact that macroalgae production is only taken into account for half of the year.

To conclude, in order to improve the environmental performance of macroalgal biomethane, it is necessary to focus on the following main points:

- Improving the efficiency of processes and facilities where electricity is used.
- Changing the nature of the energy used for a clean and efficient one. This is particularly important regarding electrical consumption and heating of the digesters. The high level of home consumption means that a high amount of biomass is required. Furthermore, burning causes pollutants to be released into the atmosphere.
- In scenario (A), another important point is the quantity of steel and concrete used offshore. This can be reduced, depending on the environmental conditions on site.
- In scenario (B), reactant consumption and especially that of filter aid and HCl used for biomolecule extraction are important bottlenecks which need to be overcome.

Accounting for high-value co-products in scenario (B)

Financial allocation is particularly beneficial to biomethane production (1.5% of total impacts) rather than to high-value biomolecules. In the case of biofuels issued from corn, rapeseed, and soybean, 16%, 23%, and 53% of the plant's revenue, respectively, comes from co-products, used as protein and energy source for the livestock industry.[31] Thus, the economic value of co-products in the first-generation biofuels industry is not as high as that of the system described with alginate production. Consequently, the relevance of considering the production of bioenergy as the main function of system (B) is debatable: from a financial point of view, bioenergy production is simply a way of giving value to wastes, and possibly of limiting the impacts of high-value biomolecules.

If an existing alginate producer decides to produce energy for its waste management, this would justify financial allocation and environmental performances of the resulting biomethane would be very high. If an industrial company decides to produce bioproducts in order to complement energy production, then an energetic allocation would be justified and environmental performances of this biomethane would no longer compete with other biofuels.

Furthermore, prices change with time, particularly as biofuel by-products have a double function as both shock absorber and price adjuster. The production of by-products on the conventional biofuel market could increase due to government subsidies or positive oil price shocks. As a result, their prices fall relative to other feed products.[31] If the quantity of macroalgae treated for the production of biomethane from extraction residues became significant, this phenomenon would appear. Then the market prices would strive for equilibrium between the profitability of the biofuel and of the byproduct, leading to an increase of environmental impacts allocated to the biofuel. Thus, as far as financial allocation is concerned, it is hard to come to a conclusion concerning the environmental benefits of biomethane from extraction residues.

Efficiency of the anaerobic digestion process and seasonality

The seasonality parameters used within this paper play secondary roles in the results from an environmental point of view. Nevertheless, in real biorefineries this question usually needs to be studied carefully.[17] For the industrial scaling, we considered that it was possible to use the digesters only half of the year. Nevertheless they would take time to become stable and efficient after being set up. This could be a hard point to manage in industrial conditions, unless the digestion of terrestrial feedstock is possible for the rest of the year, and seasonality could be more important for the anaerobic digester management than a simple question of wear-out of the facilities.

This is the next challenge which needs to be overcome before tackling the following step of eco-design in macroalgal bioenergy production chains. Furthermore, biomolecule content is highly variable throughout the year.[32] With the potential methane yield, these seasonal variations are even more important than variations between different macroalgal species (Jard *et al.,* unpublished work). For these reasons it is also important to focus on cultivation methods in order to optimize macroalgal composition and degradability.

The nature of the digested waste needs to be focused on in scenario (B). In this study, only the anaerobic digestion of solid alginate extraction residues is taken into account. From an industrial point of view, this digestion could not occur without co-digestion as they are not sufficiently biodegradable and their productivity in CH_4 is too low. Experiments carried out on the digestion of alginate extraction residues from *Laminaria digitata* show that digestion of the liquid effluents plays an important role in optimizing the process performance.[33] Terrestrial feedstock could also be used to optimize digestion of this waste and to solve seasonality problems.[34] In this study, with a view to improving digestion performance, the use of cellulosic filtration aids was considered rather than that of the diatomaceous earth (traditionally used).

Data collection from pilot-scale to industrial scale

Because data were collected from a pilot-scale for biomass cultivation and extraction, industrial realities can sometimes be different compared with the system described. Lab-scale extraction experiments do not lead to energy efficiency maximization, contrary to industrial level.[35] Nevertheless, data for water consumption in the pilot were consistent with industrial data: 0.670 $m^3.kg^{-1}$ of sodium alginate (not including washing water) versus 1 to 1.5 m^3 all water included in the alginate industries.[36] In spite of relatively good results compared with this mean water consumption value, the quantity of water used is still very high, and many efforts need to be made to decrease this. Different options exist.[37] One of them is to reuse some of the water flows between operations, depending on the water quality requirements within the different production steps. Another option is to recycle water reclaimed from waste-water treatment for the same or for other operations.

Values were corrected by bibliographic industrial data regarding electricity consumption. High energy consumption is typical within the food and drink industry.[38] Nevertheless, due to the possible coupling with a wind farm, the remaining point to focus on after ecodesign and replacement of the source of energy is the quantity of reactants used, notably the filter aid and the hydrochloric acid. Cellulose powder could easily be replaced by diatomaceous earth, commonly used in alginate extraction. However, its mineral matter content would increase the volume treated in the digester, and decrease the anaerobic digestion performance. Furthermore, this is a non-renewable matter. This question still needs to be studied in more detail.

Upgrading scenarios and comparative study

In this part of the study, the influence of some technical parameters and of some ecodesign choices is tested on both scenarios (A) and (B). These hypotheses are compared to the initial scenario detailed in the inventory and analyzed using contribution analysis. Following this, a comparison between macroalgal biomethane and natural gas is performed.

Influence of ecodesign and technical improvements on environmental performance

Improvements can be summarized within three categories.

1. **The source of energy used**. Because of the possible links between offshore wind farms and seaweed cultivation, the coupling of these two activities within an integrative framework was tested (improvements 1 and 2).
2. **External improvements, which can be chosen between existing solutions through ecodesign** (improvements 3 to 6). In this study, the aspects treated for scenarios (A) and (B) were: first, the recycling of materials used for buildings and facilities and second, the replacement of nursery substructures with greenhouses to decrease the use of fluorescent lamps. For macroalgal residues from scenario (B), two more hypotheses were tested, concerning a decrease in electricity consumption during the drying process and a decrease in energy consumption during heat transfers within the biorefinery.
3. **Technical improvements** (improvements 7 and 8). The hypotheses tested were biomass productivity enhancement and a decrease in the fuel consumed by the harvesting boat.

Improvement hypotheses tested

Improvement 1: Electricity from offshore wind farms

In order to exploit full use of the offshore cultivated area, it is biologically and technically feasible to couple seaweed and electricity by way of offshore wind turbine production.[39] Since electricity from wind farms is a renewable source of energy and is produced locally, tests were carried out in order to replace the European electrical mix with an offshore wind farm to feed the nursery, the biorefinery, the anaerobic digestion plant and the gas station.

Improvement 2: Heating of digesters using offshore wind farms

As the use of biogas to heat the digesters is not necessarily the most efficient option, a test was carried out to replace its home consumption by heating within an electrical boiler. Home-consumed biogas was produced after several energy conversions: solar energy to raw biomass through photosynthesis, biomass transportation, a possible transformation into extraction residues, anaerobic digestion into biogas, biogas burnt to produce heat, and finally, heat transfer to maintain the digesters at a mesophilic range of temperature. At each step, loss coefficients reduced the energetic potential initially available in the whole system. Therefore offshore wind power was considered as an energy source for digester heating.

Improvement 3: Material recycling

After dismantling building and replacing facilities, the following materials were recycled instead of land-filled: concrete (reinforced or not), mineral wool, polypropylene, polyethylene, polyethylene terephthalate, polyvinylchloride, bricks, cement fibers, steel, and iron.

Improvement 4: Greenhouses

A roof with double-glazing was added to the nursery building to allow direct sunlight to reach the plantlets. Daily artificial photoperiod was then decreased from 18 h per day to 10 h per day for zoospores collected in autumn. In China, where most of the kelp is cultivated, greenhouses are used in the nurseries to ensure plantlet cultivation.[40] This technique can only be applied for zoospores grown during wintertime under a temperate climate. During the summertime superheat should be compensated with an air-conditioning system.

Improvement 5: Drying process

In the biorefinery, the most energy-consuming steps are the drying process and heat transfer. The breathing space in this field is especially high depending on the chosen technology. In the initial scenario convective drying was chosen without heat recovery (920 $kWh.t^{-1}_{water\ removed}$) for alginate extracts. With heat recovery and/or evaporation by mechanical compression of steam, energy consumption could be consequently reduced:[26] half of the initial value is assumed.

Improvement 6: Heat transfer process

The same hypothesis was applied to the heat transfer as for the drying process, halving the energy consumption.

Improvement 7: Productivity 11.7 $kg.m^{-1}$

It would be possible to improve the average productivity of 8.95 kg.m of wet biomass. Under a temperate climate, the date of outplanting determines productivity values, ranging from 6.7 to 11.7 $kg.m^{-1}$ in Spain (even reaching 13 $kg.m^{-1}$ in the northern part of Spain).[24] The influence of productivity values on environmental performance was tested for 11.7 $kg.m^{-1}$ of rope, corresponding to an increase of 23.5% in biomass productivity.

Improvement 8: Fuel consumption -12%

The fuel consumed by the boat was decreased by 12%, i.e. from 1.17×10^{-1} to $1.04\times10^{-1} L_{diesel}.t^{-1}.km^{-1}$.

Results of improvement solutions for macroalgal biomethane production

All results are presented in Figure 4. For each hypothesis, the result is shown according to the initial scenario (e.g. the value of 0.57 for climate change in Hypothesis 1 means that the environmental impact represents 57% of the initial value when electricity from the grid is replaced by electricity from offshore wind farms). Thus for each impact category the value of 1 on the radar corresponds to the initial value, and the bigger the area on the radar, the higher the environmental impacts are.

The most important influence is due to the use of electricity from offshore wind turbines. Compared to the initial scenarios, this change allowed environmental improvements ranging from 5.6 to 86.0% (with an average improvement of 38.9%) and from 5.7 to 70.8% (with an average improvement of 34.3%) for scenarios (A) and (B), respectively.

The use of wind power to heat the digesters also led to significant environmental benefits. Environmental improvements ranged from -18.5 to 17.9% (with an average improvement of 10.9%) and from 19.5 to 28.7% (with an average improvement of 27.3%) for scenarios (A) and (B), respectively. A negative influence for scenario (A) corresponded to the marine and freshwater eutrophication impact categories: less macroalgae needed to be harvested to produce a given amount of biogas when biogas is not home consumed. Thus the positive impacts of offshore nutrient uptake in the initial scenario were limited when biogas home consumption was avoided.

Material recycling revealed interesting results in scenario (A). The average improvement calculated on every impact category reached 9.0% compared to the initial scenario. It was especially beneficial for metal depletion, with a 47.6% decrease in environmental impacts. Only ionizing radiation and natural land transformation were not reduced by material recycling, since energy was required for waste recycling and because re-vegetation of land-fills was avoided.

In scenario (A), the use of double glazing without recycling for the greenhouses in the nursery decreased the system's environmental performance, but glass recycling led to a better performance with improvement ranging from 1.0 to 15.6%. This was due to lower electrical requirements to light the plantlet cultivation ponds.

The influence of the material recycling performance on scenario (B) and the use of greenhouses in the nursery was very low (an improvement of 0.4 and 0.2% respectively, averaged on every impact category). This is due to the high quantities of reactants and energy consumed.

Ecodesign solutions concerning both the convective dryer and heat transfer processes averaged improvements at 8.9% and 2.1% compared with the initial scenario.

In scenario (A), the hypotheses in which biomass productivity increased (+23.5%) and fuel consumption decreased (-12%) led to a decrease in the environmental impact of 19.7% and 1.3%, respectively. With an increase in biomass productivity, the results were once again distinctive in the case of freshwater and marine eutrophication: the uptake of nitrogen and phosphorus was proportional to the quantity of biomass produced; therefore the environmental performance of the system remains stable and does not improve with an increase in productivity. These technical improvements had a very low impact on the environmental performance of scenario (B): 0.04% and 0.6% in the case of biomass productivity and fuel consumption respectively. It should be noticed that more important productivities (as 25 $kg.m^{-1}$ in an Asiatic context) would decrease even more these environmental impacts at the cultivation stage.

Comparison between the three production scenarios after improvement

A comparison of the impacts of scenarios (A), (B), and (C) are shown on Figure 5, in blue, green, and grey, respectively. Both the initial scenario and an ecodesigned scenario were tested in (A) and (B). Material recycling and the use of offshore wind farms to feed all the facilities and to heat the digesters were tested in the ecodesigned scenarios. These three hypotheses were chosen since technical implementation was easier.

The results emphasized the fact that ecodesign and change in the source of energy could make macroalgal biomethane competitive with natural gas in terms of environmental

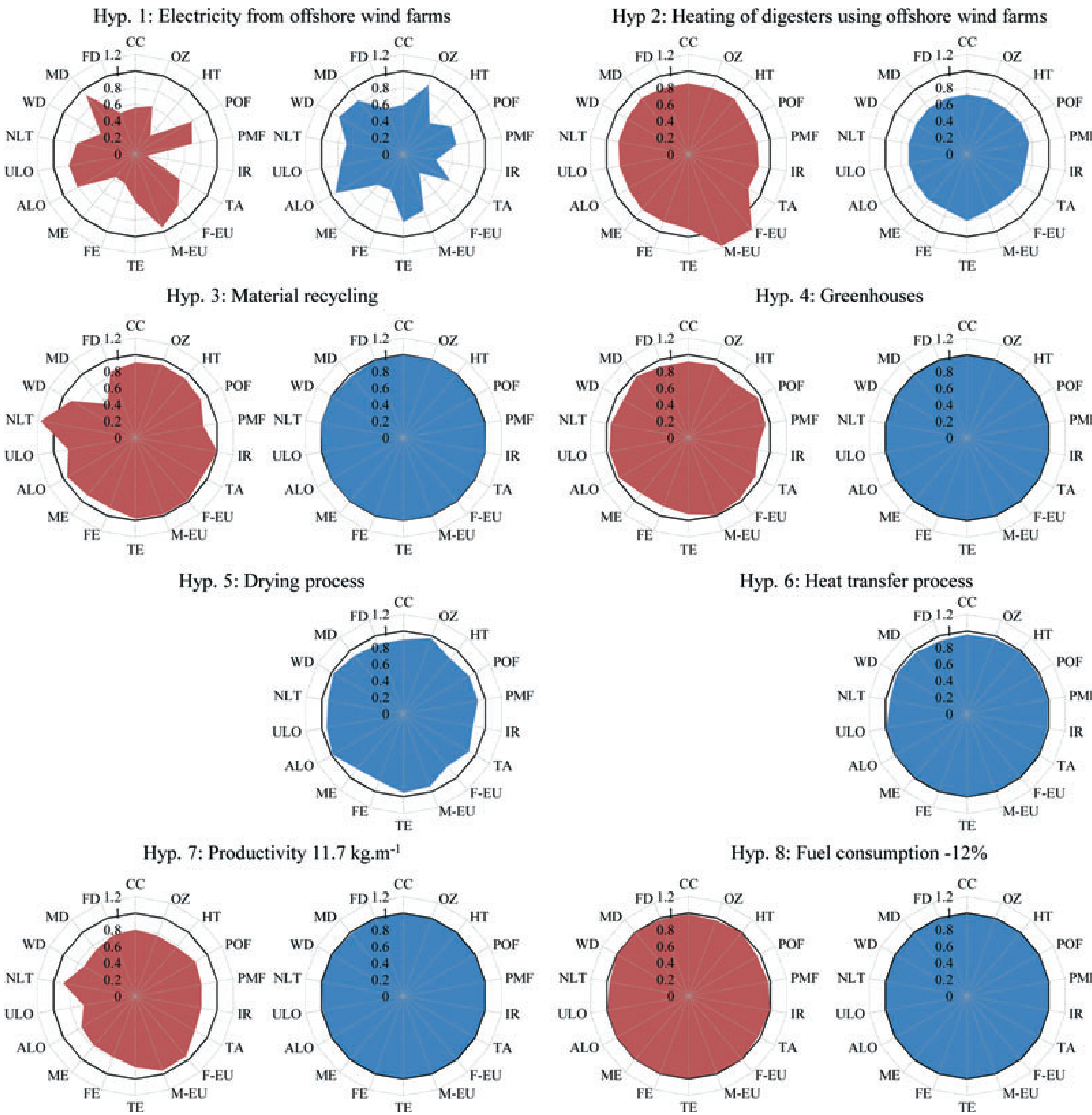

Figure 4. Influence of parameters on the environmental performance of macroalgal biomethane (scenario (A) in red on the left-hand side and scenario (B) in blue on the right-hand side) in proportion to the initial impacts. Abbreviations are detailed in Figure 3.

performance. Significant improvements resulted in climate change (-21.9% and -54.2%), fossil depletion (-58.6% and -68.7%), and ozone depletion (-70.6% and -31.1%). In scenario (A) there were even environmental benefits concerning marine eutrophication. Nevertheless, impacts were significantly higher in relation to human, terrestrial and freshwater toxicity, metal and water depletion, urban and agricultural land occupation.

Environmental impacts of digestion of extraction residues were lower than for untransformed macroalgae (A): in the case of (A), ecodesign results were less efficient concerning 10 impact categories. Nevertheless, considering the variability of the results due to the financial allocation, it is hard to determine which system is the most efficient.

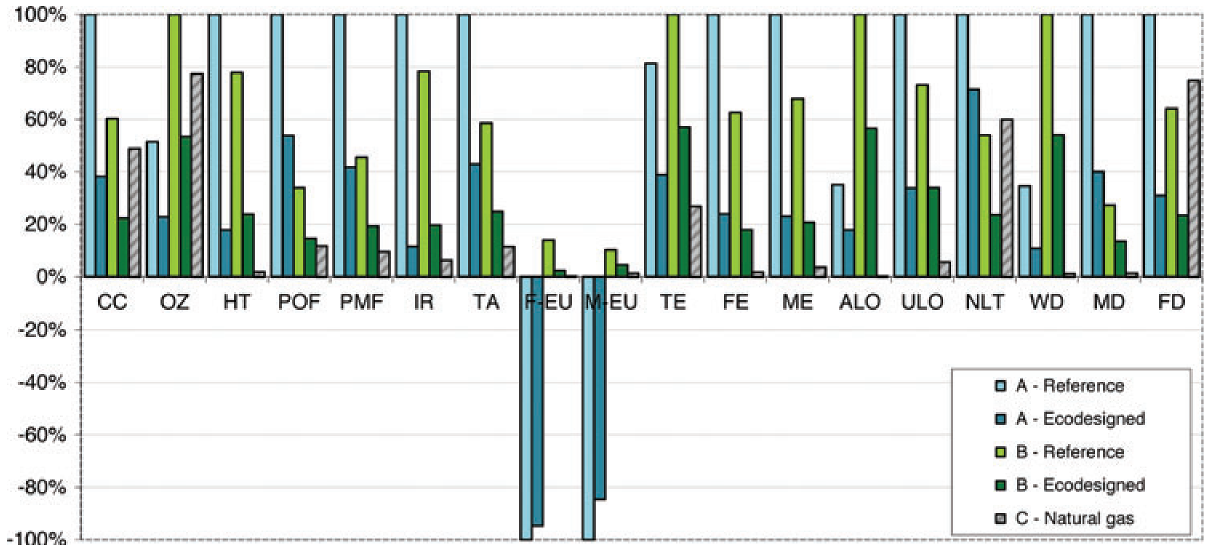

Figure 5. Comparison of the environmental impacts of biomethane from (A) untransformed algae, (B) macroalgal extraction residues and (C) natural gas used to drive a passenger car for 1 km. Abbreviations are detailed in Figure 3.

Conclusion

Macroalgal biomethane from fresh algae appears to be an interesting biofuel from an environmental point of view. With conventional techniques, its impacts are still higher than those of natural gas. Nevertheless, after ecodesign steps and considering technical improvement, its production can present high levels of efficiency, especially in the case of climate change and of fossil depletion. This is possible by designing the systems with a clean and efficient source of electricity (offshore wind farms) on site and to heat the digesters. In scenario (A), using untransformed, whole, macroalgae for anaerobic digestion, the remaining impacts where efforts have to be made are the offshore infrastructures, mainly because of steel and concrete. In scenario (B), using macroalgal residues from alginate extraction, the remaining improvements are linked to the biomolecule extraction process itself.

Choice of financial allocation strongly influences the results, notably depending on the alginate price. This type of allocation depends on the functions given to the biorefinery: producing energy (scenario A) or reducing impacts of waste treatment (scenario B). A realistic scenario is a combination of both kinds of feedstock, giving more flexibility to the production system.

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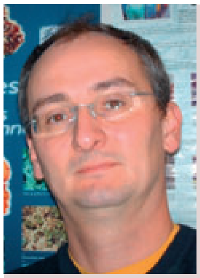
Juliette Langlois

Juliette Langlois is a PhD student at Montpellier SupAgro (France), where she graduated as an agronomist (Master's degree, 2009). She is currently pursuing a PhD on life cycle assessment (LCA) of bioenergy from seaweed cultivated in open ocean (WinSeaFuel project) and more largely on impact assessment of land use and of marine activities in LCA.



Jean-François Sassi

Jean-François Sassi has been Department Manager and Team Leader for the Algae Product Innovation at Centre d'Etude et de Valorisation des Algues (CEVA, France) since 2006, after several researcher positions in chemical companies and academic groups. He holds a PhD (1995) in Chemistry of Natural Polymers.

Gwénaëlle Jard

Gwénaëlle Jard holds a PhD (2009) in Microbiology and Process Engineering and a Master's degree in Biochemistry (2006, INSA, France). After a research engineer position at INRA (2010/2011) on anaerobic digestion of macroalgae, she is now a researcher in an agronomic engineering school (El Purpan). Her teaching is dedicated to process in food industry.

Jean-Philippe Steyer

Jean-Philippe Steyer holds a PhD in Applied Mathematics (1991). He is now the Executive Director of the Laboratory of Environmental Biotechnology (INRA, Narbonne, France), working on modeling and control of bioprocesses (mainly anaerobic digestion).

Jean-Philippe Delgenes

Jean-Philippe Delgenes holds a PhD in Environmental Sciences (1989). He is a researcher on anaerobic digestion of lignocellulosic biomass and organic solid waste at the Laboratory of Environmental Biotechnology (INRA, Narbonne, France), where he has been Executive Director.

Arnaud Hélias

Arnaud Hélias obtained a PhD (2003) in Process Engineering. He is now Associate Professor at Montpellier SupAgro, where he graduated as an agronomist (2000), with teaching on ecotechnology and environmental assessment. His research focuses on life cycle assessment applied to waste, wastewater treatment, and algae cultivation for energy purposes.