

# Life cycle assessment of macroalgae cultivation and processing for biofuel production



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## ABSTRACT

There has been a recent resurgence in research investigating bioenergy production from algal biomass due to the potential environmental benefits in comparison to conventional bioenergy crops and conventional fossil fuels. This life cycle assessment (LCA) considered the energy return and environmental impacts of the cultivation and processing of macroalgae (seaweed) to bioethanol and biogas with a particular focus on specific species (*Gracilaria chilensis* and *Macrocystis pyrifera*) and cultivation methods (bottom planting and long-line cultivation). The study was based mainly upon data obtained from research conducted in Chile but the results can be applied to other locations where similar cultivation is feasible. Speculative data were also included to test promising data obtained from research. The results suggested that using base case conditions the production of both bioethanol and biogas from bottom planted *Gracilaria chilensis* was the most sustainable option due to the low input method of cultivation. Using new advances in cultivation and processing methods of long-line cultivated *Macrocystis pyrifera* however resulted in a much more sustainable source of bioenergy. If these methods can be proven on a large scale, the generation of bioenergy from macroalgae could be highly competitive in terms of its sustainability compared to alternative feedstocks. Future research should bear in mind that the results of this study should however be considered highly optimistic given the early stage of research.

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## 1. Introduction

Growing concerns regarding the impact of fossil fuel use upon the environment and the cost of production have led to a growth in the interest of obtaining energy from biomass. The sustainability of recovering energy from biomass however is often criticised due to high energy inputs and adverse environmental impacts (Hall et al., 2009). New biomass feedstocks are being sought which can be sustainably cultivated and converted to an energy source (Antizar-Ladislao and Turrion-Gomez, 2008), algae is an example which is currently gaining much interest (Aitken and Antizar-Ladislao, 2012). One potentially favourable feedstock is marine macroalgae or what is more commonly referred to as seaweed (Hughes et al., 2012). The marine cultivation of seaweed does not generally require arable land or fertiliser, the necessary elements for growth

are typically found in the coastal environment (Langlois et al., 2012). Also the biomass yields of macroalgae over a growing season can be greater than those of most terrestrial crops as a result of high biological productivity (Hughes et al., 2012). Nevertheless, as far as the authors are aware, there are currently no systems which are operating to cultivate and obtain energy from macroalgal biomass on a large scale and the sustainability of such a system is largely untested (Bruton et al., 2009). Research has been conducted in a number of studies dating back to the 1980s to investigate the potential yields of energy recovery from macroalgal biomass (Ghosh et al., 1981; Debusk et al., 1986). These studies have mainly been concerned with the production of methane (Ghosh et al., 1981), however more recently attention has been paid to the recovery of bioethanol (Wargacki et al., 2012). High yields of both methane (Chynoweth et al., 2001) and bioethanol (Wargacki et al., 2012) have been recorded however consequent scaling up has been limited. As a result the overall sustainability of such systems is largely unknown, recently however there have been several life cycle assessment (LCA) studies conducted which have examined

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### List of abbreviations

LCA	Life Cycle Assessment
EROI	Energy Return on Investment
TE <sub>p</sub>	Total Energy Produced
TCED	Total Cumulative Energy Demand
GWP	Global Warming Potential
ODP	Ozone Depletion Potential
CFC-11	Trichlorofluoromethane
POCP	Photochemical Ozone Creation Potential
HTP	Human Toxicity Potential
VS	Volatile solids
w.w.	wet weight
d.w.	dry weight

the sustainability of resource and energy recovery from brown seaweed (Langlois et al., 2012; Alvarado-Morales et al., 2013). Langlois et al. (2012) focussed upon a set of environmental impacts (e.g., climate change, ozone depletion and human toxicity) of the generation of methane from brown seaweed (*Saccharina latissima*) from the whole seaweed and following alginate production. Their results indicated that compared with natural gas the impacts were less favourable although improvements to the design allowed both processes to compare more favourably. Alvarado-Morales et al. (2013) investigated two production scenarios from brown seaweed (*Laminaria digitata*) in Nordic conditions; one producing just biogas and the other bioethanol with the stillage being converted to biogas. The authors compared the energy process balance of the two scenarios and the potential environmental impact on global warming, acidification and terrestrial eutrophication. The production of both bioethanol and biogas facilitated a greater energy export however the environmental impacts were poorer as a result of the extra fermentation and distillation process. These studies suggest bioenergy generation from macroalgae is a promising concept although the focus has been largely on brown seaweed for large scale off-shore sites.

Therefore, the aim of this study was to use a life cycle assessment approach to investigate the sustainability of producing bioenergy from macroalgae using local cultivation techniques and species that are common to a given area, and the production of bioethanol, biogas or both. It should be noted due to the relatively new state of research, there are few studies which provide values that could definitely be replicated on a large scale. Values used in this study were taken from a variety of studies, and chosen because they were deemed the most suitable.

## 2. Materials and methods

### 2.1. The case study

Chile has an extensive coastline and much of the coastal area is suitable for the growth of seaweed. In the past, Chile has been one of the largest producers of alginate, agar and carrageenan, all of which are derived from macroalgae (Alveal et al., 1997), traditionally using wild stocks. Overexploitation, however, has made cultivation necessary (Buschmann et al., 2005). In Chile, macroalgae cultivation has largely been restricted to red algae and more specifically *Gracilaria chilensis*, due to the relatively simple farming methods that have been developed (Buschmann et al., 2005). Brown macroalgae, particularly *Macrocystis pyrifera* or giant kelp, is also becoming an important resource and alongside *G. chilensis*, one of the favoured species for cultivation due to its economic importance (e.g., abalone *Haliotidae* farming, fertiliser) and potentially

high yields (Buschmann et al., 2008). The cultivation of *G. chilensis* is most commonly carried out by a method called 'bottom-planting' in which the thalli of previously cultivated biomass is planted into the seabed sub-tidally, the biomass can then provide several harvests before requiring replanting (Buschmann et al., 1995). 'Long-line cultivation' by tying thalli to ropes and deploying the ropes off-shore has also been successfully demonstrated and can offer cultivation on a larger scale (Buschmann et al., 2008). Long-line cultivation of *M. pyrifera* is the traditional method of culture however lines are first inoculated with spores prior to subsequent deployment offshore. This method can typically deliver very high yields of biomass on a large scale (Gutierrez et al., 2006).

### 2.2. Goal and scope

The goal of this LCA study was to determine the most sustainable method of cultivating and processing macroalgae to bioenergy in Chile. For the purpose of this study, the three above-mentioned cultivation scenarios of macroalgae as potential sources of bioenergy were modelled. These included: bottom planted *G. chilensis* (S1), long-line cultivated *G. chilensis* (S2), and long-line cultivated *M. pyrifera* (S3). Furthermore, following cultivation, the conversion of the biomass to bioenergy was considered. Three different bioenergy pathways were modelled: fermentation and distillation to bioethanol (P1), anaerobic digestion to biogas and subsequent conversion to electricity (P2) or both, with the stillage produced from the bioethanol plant being anaerobically digested and the biogas converted to electricity (P3). Nine different process streams were therefore modelled in total.

The sustainability metrics chosen were the Energy Return on Investment (EROI) and six environmental impact categories (i.e., climate change – GWP 100a (kg CO<sub>2</sub> eq); acidification potential – generic (kg SO<sub>2</sub> eq); eutrophication potential – generic (kg PO<sub>4</sub> eq); stratospheric ozone depletion – ODP steady state (kg CFC-11 eq); photochemical oxidation – High NO<sub>x</sub> POCP (kg ethylene eq); human toxicity – HTP 100a (kg 1,4-DCB eq)).

The EROI is defined very generally as the ratio of the output energy of a system to the input energy to the system (Murphy and Hall, 2010). In this study, the EROI was calculated as the ratio of the lower heating value of energy carriers (TE<sub>p</sub>) to the total cumulative non-renewable fossil energy demand (TCED) (Eq. (1)).

$$\text{EROI} = \frac{\text{TE}_p}{\text{TCED}} \quad (1)$$

Alongside the EROI analysis, the six environmental impact categories provided a wide scope for comparison. The impacts were determined by combining the emissions from each unit process and calculating the contribution to each impact category using the Centrum voor Millikunde Leiden method (CML 2001). The results of each scenario were compared to the same metrics for energy in the form of petroleum, bioethanol from corn and electricity from biogas obtained from a mix of biowaste and sewage sludge, all of which were calculated using data from Ecoinvent v2.2 (Ecoinvent, 2010).

The functional unit used for comparison was 1 MJ of energy, as the lower heating value of the energy carriers produced. The inputs to the inventory were detailed for 1 ha of cultivation area for clarity. For each scenario, inputs to model a base case were used however as part of the sensitivity analysis several inputs were adjusted to more favourable values to investigate the impact upon the results of the model. The study considered the cultivation of species that are common to Chile but which can also be found in comparable environments, the results are therefore also applicable to these environments.

2.3. System model

2.3.1. Cultivation

Three cultivation scenarios (S1–S3) were modelled (Fig. 1). Scenario 1 (S1) was the bottom planting of *G. chilensis*. The first step of the process stream was preparation of the thalli followed by planting of the thalli 1 km from the landing point and processing facilities. The biomass was then assumed to be harvested by a diver and fishing vessel. The total cultivation area was considered to be 20 ha (based on information provided by local farmers: a diver requires 3 days to harvest 1ha using 30 L/d of diesel). Scenario 2 (S2), the long-line cultivation of *G. chilensis*, was initiated by tying previously cultivated biomass thalli to ropes. The ropes were then deployed 10 km from the landing point using a barge vessel, following a culture period of 6 months they were assumed to be harvested by the same vessel. Current studies use sites of only 1 or 2 ha, a reasonable next step for a commercial development was

assumed to be 100 ha as a manageable size. Thus, the total cultivation area was considered to be 100 ha. Scenario 3 (S3), the long-line cultivated culture of *M. pyrifera*, required first the inoculation of the lines with spores and subsequent development in tanks as part of the hatchery process. The lines were then assumed to be transported 10 km from the landing point to the cultivation site by barge and harvested by the barge at the end of the 9 month culture period. As with S2, the area of cultivation in one development was assumed to be 100 ha.

2.3.2. Processing

The processing of the biomass was common to each scenario. Pre-processing of the biomass included the transport of the biomass from the landing site on a conveyor belt and grinding using a wet biomass attritor (no washing step with freshwater was considered based on discussions with Bio Architecture Lab Inc). The biomass was then either processed using one of the three

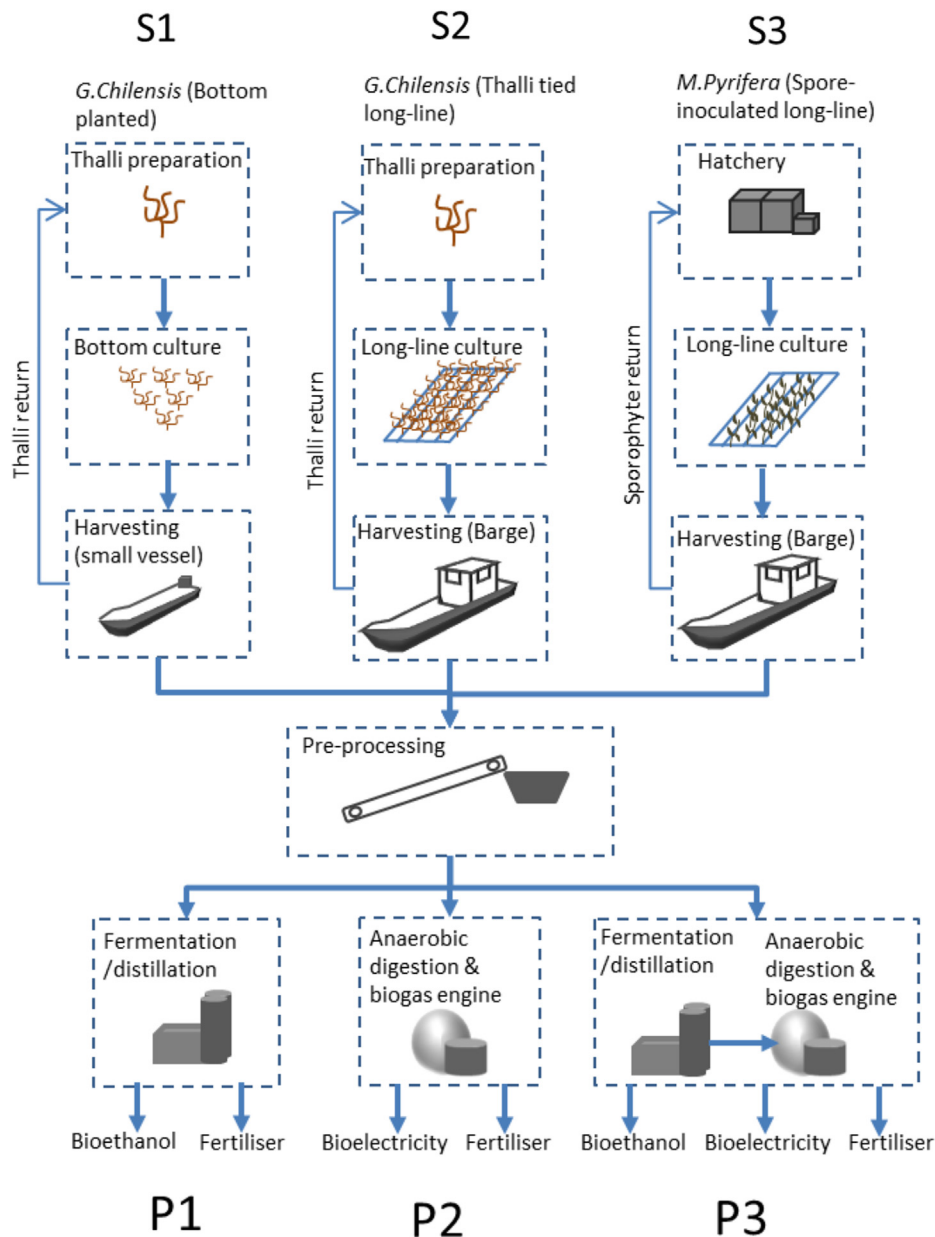


Fig. 1. Flow diagram with each unit process considered in the LCA for the three cultivation scenarios and three processing streams.

production alternatives (P1, P2 or P3). P1, the generation of bioethanol was modelled using data from Ecoinvent for the production of bioethanol from corn (Jungbluth et al., 2007) as data for macroalgal biomass are currently lacking. The model includes data for pre-treatment, saccharification, fermentation and distillation. P2, the generation of biogas was modelled using the method for anaerobic digestion of sludge in wastewater treatment (Tchobanoglous et al., 2003) considering the mixing and heating of the biomass. P3, the generation of bioethanol followed by the processing of the residual biomass to biogas using anaerobic digestion, both processes were modelled using the same methods employed for P1 and P2 respectively.

### 2.3.3. Products

The high polysaccharide and low oil content of macroalgae favours the production of bioethanol and biogas (Bruton et al., 2009), therefore these were the two products considered in this study. Using assumptions consistent with Ecoinvent (Jungbluth et al., 2007), the bioethanol was assumed to be upgraded to 99.7% ethanol and biogas (63% CH<sub>4</sub>) was assumed to be converted to electricity and heat in a Combined Heat and Power (CHP) engine. A proportion of the electricity and heat produced from the biogas was assumed to be used as the energy source for the system (the proportion was dependent upon the scenario), the remaining electricity was exported to the national grid as the produced energy and the heat was not used although the environmental impacts of the heat produced were allocated to the exported electricity. The lower heating value of bioethanol (28.1 MJ/kg) (Jungbluth et al., 2007) and the energy value of electricity produced from the biogas (as the lower heating value) were used for comparison. Following the generation of either or both, bioethanol and biogas, the stillage and excess digestate were assumed to be used as fertiliser offsetting the impacts of the production of conventional fertilisers (i.e., ammonium sulphate, superphosphate and potassium chloride). The values were based on the nutrient contents of the biomass and their bioavailability (See Supplementary data A). As the produced fertiliser contains a proportion of the total carbon

contained within the cultivated seaweed this was considered as the mass of carbon biofixed from the atmosphere during the cultivation process. The calculated value was negated against the CO<sub>2</sub> eq emissions of the system to determine the global warming potential (See Supplementary data B).

### 2.3.4. Energy supply

Where biogas was produced, a proportion of the electricity and heat was assumed to provide the necessary energy for each process as required. The values of electrical energy and heat generated from the biogas were calculated using data from the Ecoinvent inventories for bioenergy (Jungbluth et al., 2007). The Ecoinvent report detailed that 1 MJ of biogas (63% CH<sub>4</sub> content, and lower heating value of biogas of 22.73 MJ/m<sup>3</sup>) produces 0.55 MJ of heat and 0.32 MJ of electricity. The environmental impacts were allocated based on exergy values of 1 for electricity and 0.17 for heat (Jungbluth et al., 2007). The impacts were allocated to the processes which use the energy based on the proportion of use, the impacts of the co-generation of biogas converted to electricity for export were allocated to “biogas cogeneration (export)” which included the impacts allocated to the waste heat.

Where biogas was not produced or insufficient biogas was produced to supply enough energy, the required electricity was assumed to be provided by the Chilean national grid which was modelled on available data (Supplementary data B). Heat was assumed to be provided by a natural gas boiler which was modelled on the data for heat, natural gas, at boiler condensing modulating >100 kW from the Ecoinvent database (Ecoinvent, 2010).

## 2.4. Data acquisition and modelling

Data were acquired from a variety of sources but mainly from published literature and personal communication with seaweed farmers during 2012. The data for material and energy use necessary to calculate the cumulative energy demand and environmental impacts for each unit process were obtained from the Ecoinvent

**Table 1**  
Input values assumed for the productivity, characteristics and biofuel yields from each scenario.

	S1		S2		S3	
	Value	Ref.	Value	Ref.	Value	Ref.
<b>Productivity t/(ha·y) (d.w.)</b>						
Lower	12.6	Buschmann et al. (1995)	7.6	Abreu et al. (2009)	16.5	Macchiavello et al. (2010)
Upper	19.2	Buschmann et al. (1995)	12.6	Buschmann et al. (2008)	18.8	Buschmann et al. (2008)
Base case	15.9	–	10.1	–	17.6	–
<b>Characteristics</b>						
Carbon content (%)	30.0	Chung et al. (2011)	30.0	Chung et al. (2011)	30.0	Chung et al. (2011)
VS content (%)	58.9	Habig et al. (1984)	58.9	Habig et al. (1984)	58.9	Roesijadi et al. (2010)
N content (%)	2.80 <sup>a</sup>	Troell et al. (1997)	2.80 <sup>a</sup>	Troell et al. (1997)	1.90	Roesijadi et al. (2010)
P content (%)	0.96 <sup>a</sup>	Troell et al. (1997)	0.96 <sup>a</sup>	Troell et al. (1997)	0.33	Roesijadi et al. (2010)
K content (%)	11.40 <sup>b</sup>	Tabarsa et al. (2012)	11.40 <sup>b</sup>	Tabarsa et al. (2012)	9.34	Castro et al. (2009)
<b>Bioethanol production</b>						
Lower yield (kg/kg biomass)	0.038	Kumar et al. (2013)	0.038	Kumar et al. (2013)	0.109	Lee and Lee (2012)
Upper yield (kg/kg biomass)	0.079	Wang et al. (2011)	0.079	Wang et al. (2011)	0.132	Adams et al. (2011)
Base case (kg/kg biomass)	0.059	–	0.059	–	0.120	–
<b>Biogas production</b>						
Lower yield (L CH <sub>4</sub> /g VS)	0.18 <sup>c</sup>	Costa et al. (2012)	0.18 <sup>c</sup>	Costa et al. (2012)	0.20 <sup>e</sup>	Vergara-Fernández et al. (2008)
Upper yield (L CH <sub>4</sub> /g VS)	0.23 <sup>d</sup>	Habig et al. (1984)	0.23 <sup>d</sup>	Habig et al. (1984)	0.41 <sup>f</sup>	Chynoweth et al. (2001)
Base case (L CH <sub>4</sub> /g VS)	0.21	–	0.21	–	0.30	–

<sup>a</sup> Calculated using mean value of N and P content and molecular weight of N and P.

<sup>b</sup> Value for *Gracilaria salicornia*.

<sup>c</sup> Retention time of 82 days and temperature of 37 °C.

<sup>d</sup> Retention time of 58 days and temperature of 32 °C.

<sup>e</sup> Retention time of 37 days and temperature of 37 °C and assumes a biogas methane content of 65%.

<sup>f</sup> Retention time of 46 days and temperature of 35 °C.

database (Ecoinvent, 2010) and were calculated using OpenLCA (GreenDelta TC, 2012) and compiled in Microsoft Excel.

### 2.5. Life cycle inventory

The assumptions of each scenario related to the biomass characteristics, productivity rates and bioenergy yields are detailed in Table 1. The inputs to the unit process and methods of attainment are provided in Tables 2 and 3 (For detailed information see Supplementary data A).

## 3. Results and discussion

### 3.1. Energy return on investment

Calculating the energy return on investment is a useful metric for understanding and comparing the sustainability of an energy product in terms of its energy gain. It is desirable that the energy product generates a net gain in energy, which corresponds to an EROI value above one. According to Hall et al. (2009) for an energy product to be considered sustainable, a minimum EROI of three is necessary. The greater the value of EROI, the more sustainable the source of energy is with a lower depletion of finite fossil energy (Murphy and Hall, 2011). Fig. 2 displays the EROI values for the processing streams of bioethanol production (P1), electricity from biogas production (P2) and bioethanol plus electricity from biogas production (P3) together respectively for the base case conditions for each cultivation scenario. The calculated EROI values for petroleum, bioethanol from corn and electricity from biogas are also

**Table 2**  
Life cycle inventory inputs for each scenario.

S1	S2	S3
Preparation Shed: 0.67m <sup>2</sup> Lighting: 5.6 kWh	Preparation Shed: 1.33 m <sup>2</sup> Lighting: 22.4 kWh Polyamide rope: 111.9 kg	Hatchery Shed: 0.67 m <sup>2</sup> Lamps: 960 kWh Pumping: 0.62 kWh Aeration: 8.05 kWh Water treatment: 3.02 kWh Ammonium nitrate, as N: 0.25 kg Sodium phosphate: 0.38 kg Polyamide rope: 50.4 kg
Cultivation Diesel: 23.7 kg Steel: 0.71 kg Aluminium: 0.98 kg	Cultivation Polyamide rope: 21.5 kg Concrete: 952 kg Steel: 38.4 kg Polyethylene: 52.5 kg Diesel (Barge): 0.045 kg Barge: $1.67 \times 10^{-4}$ Diesel (observation): 0.68 kg Aluminium: 0.36 kg	Cultivation Polyamide rope: 111.0 kg Concrete: 952 kg Steel: 38.4 kg Polyethylene: 52.5 kg Diesel (Barge): 0.072 kg Barge: $1.67 \times 10^{-4}$ Diesel (observation): 1.02 kg Aluminium: 0.36 kg
Harvesting Diesel: 71.0 kg Steel: 4.94 kg Aluminium: 6.83 kg	Harvesting Diesel: 4.78 kg Barge <sup>a</sup> : $1.67 \times 10^{-4}$	Harvesting Diesel: 8.35 kg Barge <sup>a</sup> : $1.67 \times 10^{-4}$
Pre-processing Conveyor belt Electricity: 5.51 kWh Steel: 2.16 kg Rubber: 3.45 kg Attritor Electricity: 289.3 kWh Steel: 22.5 kg	Pre-processing Conveyor belt Electricity: 3.58 kWh Steel: 0.43 kg Rubber: 0.69 kg Attritor Electricity: 188.0 kWh Steel: 4.49 kg	Pre-processing Conveyor belt Electricity: 6.38 kWh Steel: 0.43 kg Rubber: 0.69 kg Attritor Electricity: 334.9 kWh Steel: 4.49 kg

<sup>a</sup> The fraction of an average barge tanker operating on inland waterways, data from Ecoinvent v2.2.

**Table 3**  
Life cycle inventory inputs for each process stream.

P1	P1a	P1
Electricity: 515 kWh Heat: 14,601 MJ Ammonium sulphate, as N: 36.5 kg Diammonium phosphate, as N: 36.5 kg	Electricity: 335 kWh Heat: 9488 MJ Ammonium sulphate, as N: 23.7 kg Diammonium phosphate, as N: 23.7 kg	Electricity: 606 kWh Heat: 18,000 MJ Ammonium sulphate, as N: 42.2 kg Diammonium phosphate, as N: 42.2 kg Soda powder: 157.6 kg Sulphuric acid: 105.1 kg
Soda powder: 136.2 kg	Soda powder: 88.5 kg	Soda powder: 157.6 kg Sulphuric acid: 105.1 kg
Sulphuric acid: 90.8 kg	Sulphuric acid: 59.0 kg	Sulphuric acid: 105.1 kg
Ethanol fermentation plant <sup>a</sup> : $7.99 \times 10^{-6}$	Ethanol fermentation plant <sup>a</sup> : $5.19 \times 10^{-6}$	Ethanol fermentation plant <sup>a</sup> : $9.31 \times 10^{-6}$
P2	P2	P2
Electricity: 602 kWh Heat: 14,783 MJ Concrete: 2777 kg	Electricity: 391 kWh Heat: 11,319 MJ Concrete: 2184 kg	Electricity: 697 kWh Heat: 16,342 MJ Concrete: 3043 kg
P3 (+P1 inputs)	P3 (+P1 inputs)	P3 (+P1 inputs)
Electricity: 531 kWh Heat: 13,617 MJ Concrete: 2577 kg	Electricity: 345 kWh Heat: 10,561 MJ Concrete: 2055 kg	Electricity: 528 kWh Heat: 13,565 MJ Concrete: 2568 kg

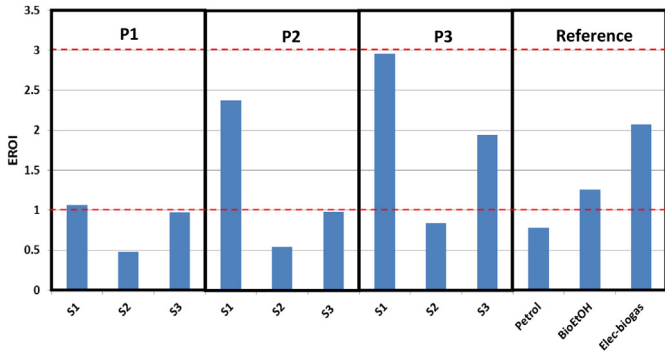
<sup>a</sup> Fraction of an ethanol fermentation plant with a throughput of 90,000 tons per year, data from Ecoinvent v2.2.

presented in Fig. 2 for reference, these were calculated using data from Ecoinvent (Ecoinvent, 2010). The EROI values were calculated by dividing the sum of 1 MJ of the total energy produced (as the lower heating value of the energy carriers) and the corresponding energy credit of the co-product by the total cumulative non-renewable fossil energy demand to produce 1 MJ. Fig. 2 includes horizontal lines displaying EROI values for the minimum values for a net energy gain (EROI = 1) and for a sustainable energy product (EROI = 3) (Hall et al., 2009).

Fig. 3 displays the contribution to the cumulative energy demand (CED) and energy produced for each scenario for the production of 1 MJ of bioethanol (P1), electricity from biogas (P2) and both bioethanol plus electricity from biogas (P3), respectively. The contribution of fertiliser as an energy credit is also displayed.

None of the scenarios and process streams reached an EROI value of 3 although bioethanol and electricity from biogas for scenario 1 was only slightly lower (2.95). No process stream allowed for an EROI value above 1 for scenario 2 due to the high CED contributions of the materials for the preparation and cultivation processes (Fig. 3). This method of cultivation was therefore discounted as a possible method of bioenergy generation. The generation of only bioethanol (Fig. 3a) produced low values of EROI with only scenario 1 providing an EROI value just over 1, which was lower than bioethanol produced from corn feedstock (1.26) when data from Ecoinvent were used (Jungbluth et al., 2007). Such low values suggest the production of bioethanol alone is not worth considering. The high CED of energy generation from the national grid and natural gas led to the low EROI values.

When only electricity from biogas was produced and used for energy generation the EROI value was higher for scenario 1 (2.38) but the value for scenario 3 remained low (0.98) because the value of energy generated was lower and the high CED of material inputs remained. For scenario 1, the production of fertiliser accounted for a large proportion of the energy produced (72%), without this allocation the EROI would be reduced to 1.39. In terms of the cumulative energy demand, the harvesting process accounted for the greatest demand at 40%. For scenario 3, the greatest contributors to the cumulative energy demand were the hatchery and cultivation processes due to the production of the polyamide rope accounting for 56% of the CED.



**Fig. 2.** The EROI values for the base case conditions of S1, S2 and S3 for bioethanol production, P1, electricity production from biogas, P2, and bioethanol and electricity production from biogas, P3 and reference energy carriers, petrol, bioethanol (BioEtOH) and electricity from biogas (Elec-biogas). (Note – Reference values use data from Ecoinvent (Ecoinvent, 2010), where bioethanol is from corn and the electricity generated from biogas is from the production mix in Switzerland using the allocation factors determined by Ecoinvent).

When both bioethanol and electricity from biogas were produced, the EROI value did increase for scenario 3 but not to the same level of scenario 1. Despite not reaching a value of 3, the production of bioethanol and electricity from biogas for scenario 1 compared favourably to the alternative energy carriers tested with electricity from biogas produced (production mix) having the

closest value of 2.07. The greatest contributors for scenario 1 were the harvesting process (30%) mainly due to diesel consumption and the fermentation and distillation process (28%) due to the chemical consumption which accounted for 84% of the fermentation and distillation CED input. As the energy was supplied from the biogas produced, the heat and electricity requirements of both fermentation and distillation and anaerobic digestion were low. Fertiliser accounted for 33% of the energy produced, without including fertiliser the EROI was 2.22. The EROI value for scenario 3 was 1.94, despite a high energy output the cumulative energy demand of scenario 3 was also high mainly from the hatchery and cultivation processes which contributed 19% and 56% respectively to the CED.

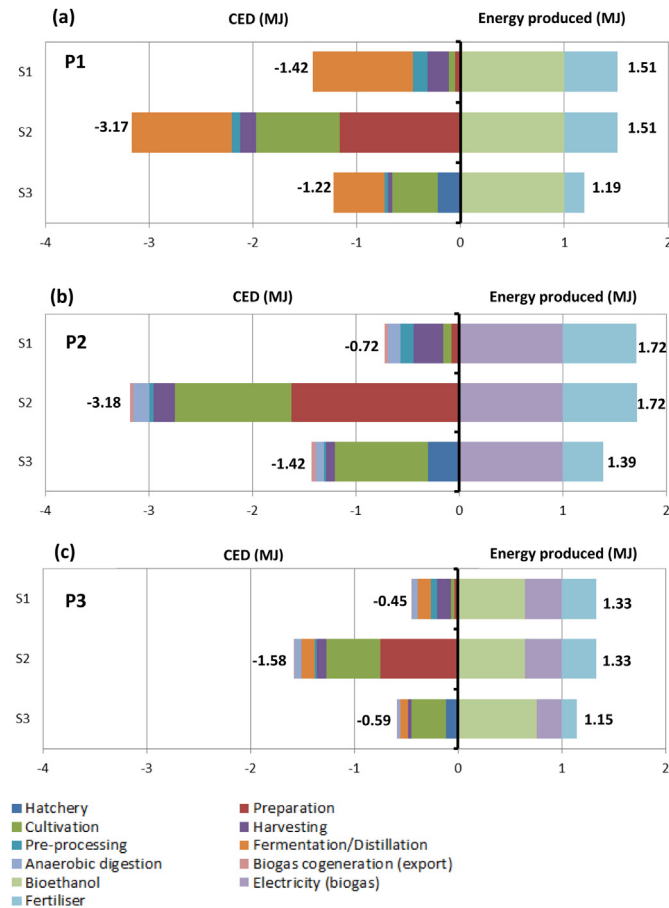
For an alternative comparison, Clarens et al. (2011) calculated that the EROI value for biodiesel production combined with electricity generated from biogas produced from microalgal biomass ranged from 0.65 to 1.13 under different cultivation conditions. They also tested other processing methods finding direct combustion favourable providing EROI values from 1.53 to 4.09. The values are quite similar to this study although the highest value of direct combustion is higher and suggests that direct combustion of the biomass should also potentially be considered for future research.

3.2. Environmental impacts

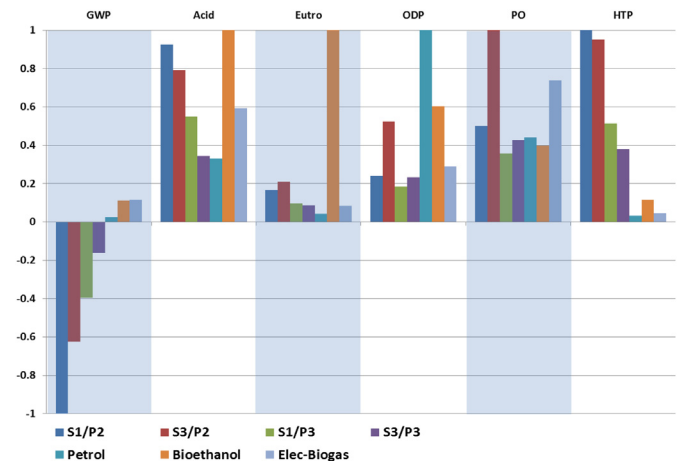
As the energy analysis favoured the generation of electricity from biogas (P2) and both bioethanol and electricity from biogas (P3) for scenarios 1 and 3, these scenarios and process streams were analysed for their environmental impacts. A wide range of environmental impacts were considered, the results for each category from each scenario were normalised to the highest impact and displayed in Fig. 4. The impacts for the generation of petrol, bioethanol from corn and electricity from biogas were also included. The contribution to each impact category for all scenarios is displayed in Fig. 5.

3.2.1. Global warming potential

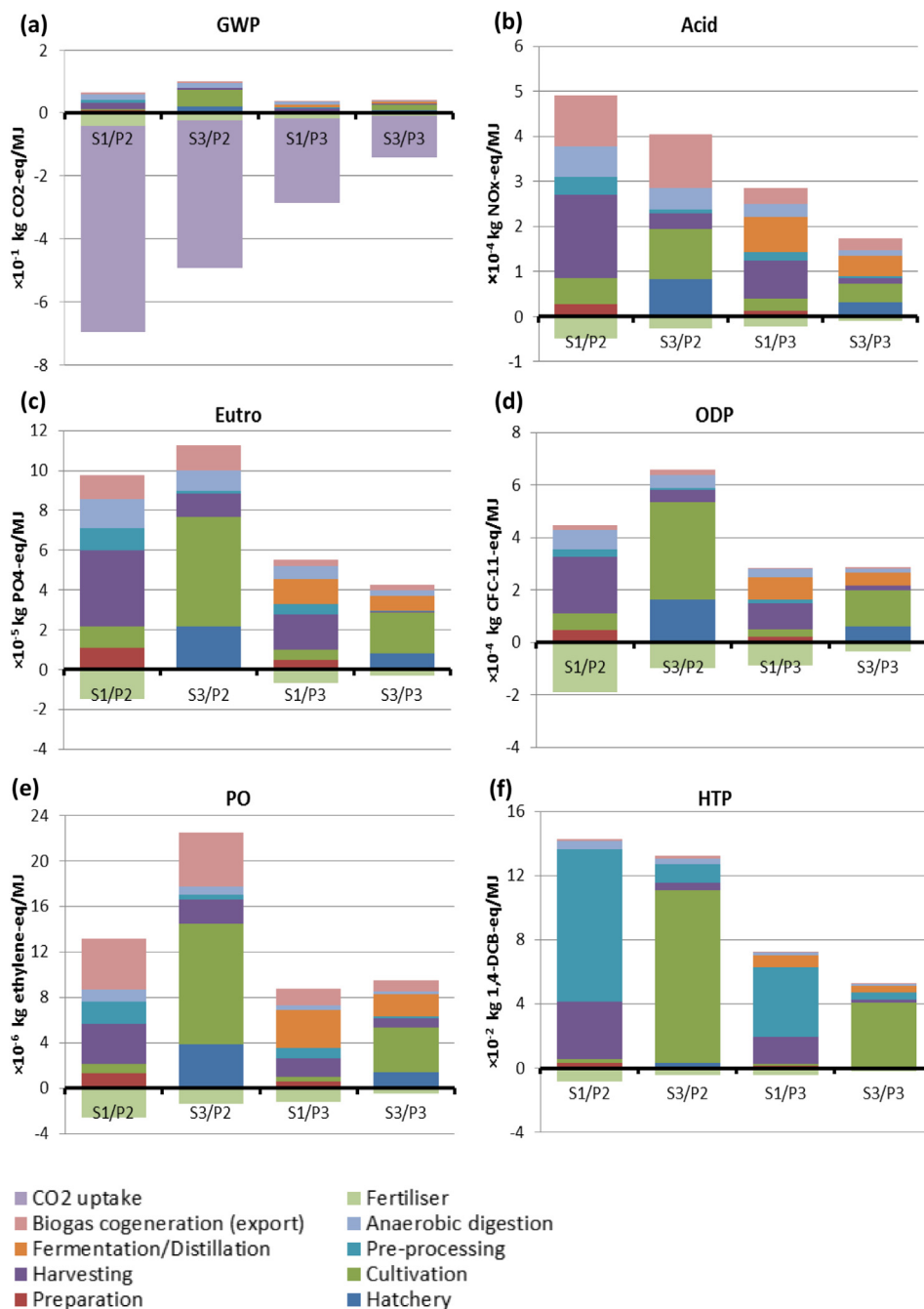
When considering the global warming potential, both scenarios provided highly promising results. For both process streams the global warming potential was negative as a result of carbon uptake by using a proportion of the biomass for fertiliser. This was in contrast to the alternative fuels tested which produced a net



**Fig. 3.** Contribution of each unit process to the cumulative energy demand and for the production of 1 MJ of (a) bioethanol, P1, (b) electricity from biogas, P2 and (c) bioethanol and electricity from biogas, P3, for each cultivation scenario, S1, S2 and S3.



**Fig. 4.** Environmental impacts per MJ of energy produced from S1 and S3 for the production of biogas (P2) and bioethanol and biogas (P3) normalised to the greatest impact. (Note – GWP – Global warming potential, Acid – Acidification, Eutro – Eutrophication, ODP – Ozone layer depletion, PO – Photochemical oxidation (summer smog), HTP – Human toxicity potential).



**Fig. 5.** The contribution of each unit process to the environmental impacts for S1 and S2 for the production of biogas (P2) and bioethanol plus biogas (P3). (a) GWP – Global warming potential, (b) Acid – Acidification, (c) Eutro – Eutrophication, (d) ODP – Ozone layer depletion, (e) PO – Photochemical oxidation (summer smog), (f) HTP – Human toxicity potential.

positive GWP value. The highest GWP avoidance was for electricity from biogas for scenario 1, largely as a result of the relatively low energy and high fertiliser yield but also because of low greenhouse gas emissions. The greatest contributors were the diesel consumption for harvesting (23%) and the concrete production of the anaerobic digestion tank (29%). For scenario 3, the high material use, largely a result of rope production for both the hatchery and cultivation processes led to a higher CO<sub>2</sub> eq output, the avoided emissions from fertiliser production were also less than for scenario 1. The models did not take into account potential greenhouse gas leakages from the anaerobic digestion facility which can potentially negatively impact the global warming potential

particularly when CH<sub>4</sub> is released (Bolin et al., 2009). Additionally, the emissions associated with fertiliser use were not considered which can also greatly impact the global warming potential particularly due to N<sub>2</sub>O emissions (Snyder et al., 2009). These emissions can be reduced by good farming practice such as ensuring no more fertiliser is applied than is sufficient for crop growth and using efficient methods of application (Snyder et al., 2009). Another source of greenhouse gases could be from biomass decomposition during storage which occurs with other biomass sources (Wihersaari, 2005). Nevertheless as the results suggest, the cultivation and processing of macroalgal biomass could provide a good method of carbon uptake and may be of interest for

businesses looking to invest in carbon credits providing an additional source of economic revenue.

### 3.2.2. Acidification

For acidification, each scenario and process stream performed favourably against bioethanol produced from corn but all were worse than petrol. The production of only electricity from biogas performed poorly particularly for scenario 1. Electricity from biogas from scenario 1 compared least favourably due to the release of NO<sub>x</sub> eq from diesel consumption during the harvesting process and biogas combustion for exported electricity production which accounted for 33% and 23% of emissions respectively. Electricity from biogas generated from scenario 3 was the second least favourable result, due to the emissions from biogas combustion for export electricity generation (30%), polyamide rope production (21%) and the barge production and diesel consumption of harvesting (9%). Methods to reduce diesel use could cut the emissions for scenario 1, a simple method could be finding the optimal cruising speed for distance travelled to fuel usage. For scenario 3, alternative materials for rope production could be sought.

### 3.2.3. Eutrophication

Each scenario compared well to corn based bioethanol for eutrophication although the emissions were higher than electricity from biogas (production mix) and petrol. Bioethanol production plus electricity from biogas performed best with similar values for both scenarios due to the higher yields of energy. The material requirements of scenario 3 produced high emissions, particularly from rope production (40%) however the high energy yield of bioethanol and electricity from biogas production lessened the impact. For scenario 1 the main source of emission was diesel consumption which similarly to acidification could be reduced by optimising fuel consumption when harvesting. The production of fertiliser has a beneficial impact through avoiding PO<sub>4</sub> eq emissions from conventional fertiliser production however the eutrophication potential of using the fertiliser was not considered, PO<sub>4</sub> releases could occur as a result of run-off from agricultural land (Smith et al., 1999). It is important that leaching is minimised by good agricultural practice such as only applying as much fertiliser as the crop can utilise and the use of efficient application methods (Smith et al., 1999).

The cultivation of macroalgae in coastal waters offers strong benefits in terms of eutrophication due to the uptake of eutrophication-causing nutrients, mainly nitrogen and phosphorous. Some studies have considered the benefit of macroalgal cultivation for nutrient removal particularly when cultivation areas are located beside fish farms where the nutrient run-off can be reduced (Buschmann et al., 2008, 2009). Macroalgae and particularly, *M. pyrifera* and *G. chilensis*, have been found to be effective biofilters (Buschmann et al., 2008). The long-line cultivation method is more applicable for nutrient removal because lines can be deployed in most coastal areas whereas bottom cultivation is restricted due to depth requirements (Abreu et al., 2009). Further work needs to quantify this benefit but it should be noted that the potential benefits are great.

### 3.2.4. Ozone layer depletion

Each of the scenarios compared well against petrol and bioethanol from corn for ozone layer depletion. In general, the values were similar to those for electricity from biogas (production mix) with the exception of electricity from biogas for scenario 3 which was higher. As with eutrophication, the main CFC-11 eq emissions for scenario 3 were a result of the polyamide ropes used, accounting for 64% of emissions, the manufacture of the barge and the diesel consumption also had a high impact. S1 provided the lowest

emissions due to the low material inputs, the emissions were mainly a result of diesel consumption. The avoided emissions from fertiliser production also greatly benefited scenario 1.

### 3.2.5. Photochemical oxidation

The values of photochemical oxidation were similar to the alternative fuels tested apart from the production of electricity from biogas for scenario 3 which produced higher emissions. The greatest contribution of kg-ethylene eq emissions was from the materials for cultivation, particularly rope production (36%). The combustion of biogas for electricity export also had a high impact (21%), this process also contributed the greatest amount to electricity from biogas for scenario 1. For bioethanol and electricity from biogas, the fermentation/distillation process also contributed greatly due to energy and chemical requirements.

### 3.2.6. Human toxicity potential

Each of the scenarios and process streams performed poorly in comparison to the alternative fuels tested for human toxicity potential. The values were greatly influenced by the production of metals for different processes. In the case of scenario 1, the high value was largely a result of the steel used for the production of the wet biomass attritor accounting for 66% of 1,4-DCB eq emissions. For scenario 3, it was the steel chains required for securing the off-shore lines that had the greatest impact, accounting for 76% of emissions. The production of both bioethanol and biogas from each scenario reduced the emissions as a result of higher yields of energy although the comparison with petrol, bioethanol from corn and electricity from biogas remained poor. Lowering the use of steel would obviously reduce the human toxicity values which could be done by using an alternative material for securing the lines and using the smallest size of attritor for biomass grinding.

## 3.3. Sensitivity analysis

Sensitivity analysis was included to test the impact upon the results of using different specific inputs to the model based on data from alternative sources or using different conditions. The alternative inputs are detailed below and the results are displayed in Table 4. The analysis was not conducted for all cultivation scenarios but only for scenarios 1 and 3 for the production of electricity from biogas (P2) and bioethanol plus electricity from biogas (P3). The best case combined all of the input changes.

### 3.3.1. Higher biomass productivity

For the production of *M. pyrifera*, recent studies performed in the South of Chile where selective breeding of biomass gametophytes have been developed have led to very high biomass yields using attachment of the developed gametophytes to long-lines. A value of 80 kg/m (w.w.) was obtained over a 12 month period by Westermeier et al. (2006). The corresponding value of 60 t/(ha·y) (d.w.) was input to the model instead of the base case. For bottom planted *G. chilensis*, a higher productivity value of 145 t/(ha·y) (w.w.) was tested which was obtained as a result of tri-monthly harvesting in the South of Chile (Buschmann et al., 1995).

### 3.3.2. High bioethanol yield from *M. pyrifera*

Recent research conducted by Bio Architecture Lab using DNA from *Vibrio splendidus* to allow the metabolism of alginate in brown macroalgae achieved high yields (0.281 kg ethanol/kg biomass) of bioethanol from *Saccharina japonica* (Wargacki et al., 2012). Such a yield has not been proven for *M. pyrifera* but from personal correspondence with Bio Architecture Lab it was understood that such yields are considered possible and therefore this yield was tested.

**Table 4**

EROI and environmental impact values resulting from the sensitivity analysis for each scenario for biogas production (P2) and bioethanol and biogas production (P3). (Note: GWP-Global Warming Potential, Acid – Acidification, Eutro – Eutrophication, ODP – Ozone Layer Depletion, PO – Photochemical Oxidation (summer smog), HTP – Human Toxicity Potential).

	EROI	GWP kg CO <sub>2</sub> eq	Acid kg NO <sub>x</sub> eq	Eutro kg PO <sub>4</sub> eq	ODP kg CFC-11 eq	PO kg ethylene eq	HTP kg 1,4-DCB eq
Base case							
S1 P2	2.38	-0.63	$4.41 \times 10^{-4}$	$8.28 \times 10^{-5}$	$2.58 \times 10^{-9}$	$1.06 \times 10^{-5}$	$1.35 \times 10^{-1}$
S3 P2	0.98	-0.39	$3.78 \times 10^{-4}$	$1.05 \times 10^{-4}$	$5.58 \times 10^{-9}$	$2.12 \times 10^{-5}$	$1.28 \times 10^{-1}$
S1 P3	2.96	-0.25	$2.49 \times 10^{-4}$	$4.86 \times 10^{-5}$	$1.97 \times 10^{-9}$	$7.55 \times 10^{-6}$	$6.90 \times 10^{-2}$
S3 P3	1.94	-0.10	$1.64 \times 10^{-4}$	$4.36 \times 10^{-5}$	$2.49 \times 10^{-9}$	$9.02 \times 10^{-6}$	$5.12 \times 10^{-2}$
Higher productivity							
S1 P2	3.10	-0.64	$3.52 \times 10^{-4}$	$6.19 \times 10^{-5}$	$1.52 \times 10^{-9}$	$8.56 \times 10^{-6}$	$9.62 \times 10^{-2}$
S3 P2	2.97	-0.42	$2.05 \times 10^{-4}$	$3.86 \times 10^{-5}$	$1.32 \times 10^{-9}$	$8.92 \times 10^{-6}$	$3.46 \times 10^{-2}$
S1 P3	3.58	-0.26	$2.21 \times 10^{-4}$	$3.89 \times 10^{-5}$	$1.48 \times 10^{-9}$	$6.60 \times 10^{-6}$	$5.13 \times 10^{-2}$
S3 P3	4.77	-0.12	$1.02 \times 10^{-4}$	$1.96 \times 10^{-5}$	$9.22 \times 10^{-10}$	$4.60 \times 10^{-6}$	$1.71 \times 10^{-2}$
High bioethanol yield ( <i>M. pyrifera</i> )							
S3 P3	3.19	-0.23	$7.78 \times 10^{-5}$	$2.22 \times 10^{-5}$	$1.57 \times 10^{-9}$	$4.61 \times 10^{-6}$	$2.72 \times 10^{-2}$
Fewer buoys and longer life span of rope							
S3 P2	1.29	-0.41	$3.56 \times 10^{-4}$	$8.89 \times 10^{-5}$	$4.14 \times 10^{-9}$	$1.80 \times 10^{-5}$	$1.27 \times 10^{-1}$
S3 P3	2.48	-0.11	$1.55 \times 10^{-4}$	$3.77 \times 10^{-5}$	$1.95 \times 10^{-9}$	$7.84 \times 10^{-6}$	$5.08 \times 10^{-2}$
Alternative fermentation/distillation data							
S1 P3	3.74	-0.27	$2.15 \times 10^{-4}$	$4.25 \times 10^{-5}$	$1.30 \times 10^{-9}$	$5.27 \times 10^{-6}$	$6.98 \times 10^{-2}$
S3 P3	1.98	-0.12	$1.43 \times 10^{-4}$	$4.31 \times 10^{-5}$	$2.30 \times 10^{-9}$	$8.38 \times 10^{-6}$	$5.48 \times 10^{-2}$
Best case							
S1 P2	3.10	-0.64	$3.52 \times 10^{-4}$	$6.19 \times 10^{-5}$	$1.52 \times 10^{-9}$	$8.56 \times 10^{-6}$	$9.62 \times 10^{-2}$
S3 P2	3.73	-0.42	$1.99 \times 10^{-5}$	$3.43 \times 10^{-5}$	$9.30 \times 10^{-10}$	$8.06 \times 10^{-6}$	$3.43 \times 10^{-2}$
S1 P3	4.85	-0.28	$1.71 \times 10^{-4}$	$3.22 \times 10^{-5}$	$7.75 \times 10^{-10}$	$4.25 \times 10^{-6}$	$5.09 \times 10^{-2}$
S3 P3	10.26	-0.04	$4.35 \times 10^{-5}$	$8.38 \times 10^{-6}$	$3.94 \times 10^{-10}$	$1.93 \times 10^{-6}$	$8.95 \times 10^{-3}$

### 3.3.3. Fewer buoys and longer life span of support ropes

Where biomass was cultivated on long lines off-shore (S2 and S3) the number of buoys was reduced to a half of the initial number, 125 buoys per hectare. Additionally, the life spans of the support ropes were increased from 5 to 10 years.

### 3.3.4. Fermentation and distillation data

An area of the study where the data used were potentially unreliable was the fermentation/distillation process. As there are no data for ethanol production from macroalgae, the base case used data from Ecoinvent (Jungbluth et al., 2007) for the production of bioethanol from corn. Alternative data which were used in the LCA study by Alvarado-Morales et al. (2013) was tested. Their LCA study used data from research where the energy input for the production of ethanol from blue-green microalgae was determined (Luo et al., 2010).

### 3.3.5. Discussion of sensitivity analysis

The higher values of productivity increased the EROI value for each scenario and process stream but the greatest increase was for *M. pyrifera* production using the selective gametophyte method (Westermeier et al., 2006). The EROI value for bioethanol and electricity from biogas increased by 246% to a value of 4.77, a higher value than that of scenario 1. This level of productivity has been proven in another study (Westermeier et al., 2011) and should be possible on a large scale. Such an EROI value suggests this cultivation method and process stream can provide a sustainable source of bioenergy. The higher yield of biomass and energy production led to lower environmental impacts, particularly ozone layer depletion and human toxicity potential.

Increasing the bioethanol yield of scenario 3 to the value that has been reported for *Saccharina japonica* (28.1 g/g) by Wargacki et al. (2012) increased the EROI to 3.19 and improved each of the environmental impacts over the base case. The values of acidification potential and eutrophication were reduced by 52% and 49%, respectively. The improvement meant the scenario compared well

in terms of both the EROI and environmental impacts to scenario 1 for base case conditions.

Reducing the number of buoys used and assuming a longer life span of rope had a relatively small impact upon the EROI and did not produce values greater than scenario 1. The environmental impacts for bioethanol and electricity from biogas production were however preferable, apart from photochemical oxidation. As *M. pyrifera* floats it is likely that in practice less buoys could be used.

When the alternative data for the fermentation and distillation process from Alvarado-Morales et al. (2013) were used there was a 26% increase to the EROI for scenario 1 but little impact upon the EROI for scenario 3. The electricity and heat consumption values for the alternative data were higher than those used for the base case but chemical consumption was not included. The higher energy values made little difference to the EROI and environmental impacts because biogas was used as the energy source but not including the chemicals greatly improved the EROI and environmental impacts for the processing of biomass from scenario 1.

When the best case was modelled, bioethanol plus electricity from biogas production from scenario 3 clearly performed most favourably in terms of the EROI (10.26) and for all of the environmental impacts. Combining the potentially high biomass yields using selective breeding and assuming a yield of 28.1 g ethanol per g of biomass produced a highly sustainable method of bioenergy generation. These assumptions however have only been proven on a small scale and the high bioethanol yield has not yet been recorded for *M. pyrifera*. Nevertheless, if the cultivation method can be scaled up and if the method for high ethanol conversion efficiency can be applied to *M. pyrifera*, a similarly high value of EROI could be possible.

## 3.4. General discussion

This study investigated the sustainability of local cultivation of seaweed in Chile and the subsequent production of bioenergy using

mainly data from studies conducted in Chile, but data from studies conducted in other countries were also used when data from Chile were not available. It should be emphasized that as the state of research is at a relatively early stage, the extrapolation of data from lab scale to full scale will incur in unavoidable inaccuracies that should be contemplated in future studies. When considering the EROI values, the results showed that the use of long-line cultivated *G. chilensis* was an unsustainable method of cultivation regardless of the subsequent processing stream, this scenario was therefore disregarded as a potential cultivation method. In terms of the processing streams, production of only bioethanol using any of the cultivation scenarios was very poor and the least sustainable processing method. The EROI results also demonstrated that electricity from biogas would only be sustainable when bottom cultivated *G. chilensis* was used. When both bioethanol and electricity from biogas were considered the EROI values were higher with a marked increase for long-line cultivated *Macrocystis pyrifera* which produced a net energy gain. In terms of EROI, the most favourable scenarios and process streams in decreasing order are as follows: S1/P3, S1/P2 and S3/P3.

In terms of environmental impacts the production of electricity from biogas for scenario 1 was poorest in comparison to S1/P3 and S3/P3 due mainly to the low energy production and therefore high relative emissions. Conversely the beneficial negative global warming potential appears to be best because of the high CO<sub>2</sub> uptake relative to the low energy yield. The environmental impacts of S1/P3 and S3/P3 are similar, despite a lower energy yield for S1/P3, the environmental impacts for cultivation and processing were also lower.

When considering base conditions it seems that S1/P3 was the best option however the limitations of the study must be considered. The sustainability of this scenario was greatly benefited by the production of fertiliser as the digestate. If the digestate was found to be less effective than assumed in this study or indeed unusable, the sustainability of bioenergy from this scenario would be reduced. Additionally, the cultivation area suitable for producing bottom cultivated *G. chilensis* is likely to be more restricted than the area which could be allocated for long-line cultivation of *M. pyrifera* due to depth requirements and the need for good accessibility. The high labour costs of *G. chilensis* planting and harvesting were not included but would have a high impact upon the economics of the cultivation method.

For these reasons, the long-line cultivation of *M. pyrifera* and processing to bioethanol and electricity from biogas is likely to be preferable as cultivation can be conducted on a large scale on much of the coastline with relatively low labour inputs. When advanced techniques for cultivation and processing of the *M. pyrifera* were considered as part of the sensitivity analysis the sustainability of the scenario with processing to bioethanol and electricity from biogas was by far the most promising.

The two major environmental benefits of development are the uptake of CO<sub>2</sub> and the uptake of nutrients from the coastal waters. The impact of the CO<sub>2</sub> recycling may be limited unless large scale development takes place. Investment in systems may be encouraged through the sale of carbon credits as a result of carbon biofixation providing this impact can be quantified.

For nutrient uptake the most effective method is cultivation near fish farms (Buschmann et al., 2008). The effluents from fish farms can be a cause of local eutrophication leading to anoxic conditions and algal blooms (Zhou et al., 2006) ultimately producing a highly negative impact against the fish farm and the coastline as a whole. As macroalgae have been proven to utilise the nutrients that cause eutrophication to occur (Buschmann et al., 2008; Sanderson et al., 2012), the impact could be greatly lessened improving the coastline for all users but particularly

fisherman and fish farmers. It should also be noted that an increase in marine biomass may lead to greater shoreline biodiversity (Bruton et al., 2009) as refuges are created for local fauna.

In terms of the socio-economic benefits, the cultivation of the macroalgae can potentially be conducted by local fisherman or fish farmers providing the dual benefit of an extra source of income by selling the bioenergy and fertiliser produced as well as treating the nutrient run-off from the fish farming activities (Buschmann et al., 2008). The cultivation phase of the system is not particularly intensive and it can therefore be conducted alongside fish farming or artisanal fishing (Buschmann et al., 2009). Equipment such as boats and sheds could be shared between the activities. When a high volume of labour is required (e.g., planting/deployment, harvesting etc.,) local fishermen could also be employed providing an extra income and a form of wealth distribution. The skills related to cultivation could be developed easily and taught to allow expansion to other local communities.

This study focussed upon the sustainability of generating bioenergy from macroalgae however it should be noted that the economic and technical viability of the concept are also greatly important. When considering the economics of bioenergy generation it is necessary for the bioenergy product to be competitive with conventional bioenergy products. A conventional feedstock for bioethanol production is corn which has a production cost of ca. \$0.16 per kg (d.w.) and an estimated cost of kelp of ca. \$0.50 per kg (d.w.), based on production in China (Roesijadi et al., 2008). Unless production costs can be considerably reduced, it is currently unlikely that bioethanol from macroalgae will be capable of competing with conventional bioenergy feedstocks. More research is necessary to study the economics of bioenergy generation from macroalgal feedstock, which may be dependent on location, and will benefit from including co-products (e.g., pharmaceutical products, fertiliser and biogas). Carbon credits and government subsidies such as feed-in tariffs can provide additional economic support for macroalgal bioenergy systems (Nayar and Froese, 2013), however subsidies can only be short term provisions. It should be emphasized that the cultivation and processing of macroalgae remains at an early stage and advancements in research are likely to improve the economics. Another barrier to full implementation may be that alternative products have a higher value than the proposed bioenergy products from macroalgal biomass. The current market value of bioethanol is ca. \$476–511 per tonne (based on 274–295 \$/USG, Argus( 2012)) whereas align and mannitol can be sold at a greater price of ca. \$6000 per tonne (Roesijadi et al., 2008). It is also a possibility however to combine the production of high value products with the generation of bioenergy which could improve the sustainability and economic viability (Langlois et al., 2012) and certainly merits further research.

Systems to cultivate macroalgae and convert the biomass to bioenergy have remained on a relatively small scale and thus full scale technical viability is largely unproven. Problems are likely as a result of mechanical failure over time, initial field tests have shown that open ocean macroalgae production are strongly affected by storms which can break the infrastructure and damage the plants (Roesijadi et al., 2008). Careful placement of offshore systems in sheltered areas may provide some risk avoidance alongside new and stronger materials being used. Further development of larger scale systems and the testing of various designs and materials will allow these technical issues to be fully identified and resolved. Further technical issues that have been mentioned previously are the risks of disease and epiphytism. Offshore cultivation is likely to lead to some epiphytism which can reduce biomass productivity however optimising growing areas and maintaining clean environmental conditions can greatly reduce the risk (Roesijadi et al., 2008).

#### 4. Conclusions

Using LCA analysis it was found that the production of bioenergy from macroalgae cultivated in Chile can be sustainable. Using current techniques, bioethanol and electricity from biogas produced from bottom cultivated *G. chilensis* was calculated as being the most sustainable cultivation technique and processing method. In the longer term, however, with improved cultivation and processing techniques, the production of bioethanol and electricity from biogas produced from long-line cultivated *M. pyrifera* was determined to provide a much more sustainable method. The flexibility of development location and size would also favour long-line cultivation of *M. pyrifera*.

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#### Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.jclepro.2014.03.080>.

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