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Conceptual design of sustainable integrated microalgae biorefineries: Parametric analysis of energy use, greenhouse gas emissions and techno-economics

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ABSTRACT

This study covers four main aspects of the conceptual design of sustainable integrated microalgae-based biorefineries using flue gas from CO_2-intensive industries (*i.e.* 100% CO_2): *i*) screening of technologies (4 options for cultivation, 3 for culture dewatering, 3 for cell disruption, 4 for lipids extraction & purification, and 4 for fractions upgrading); *ii*) analysis of processing variables (parametric study of the main cultivation conditions affecting the global performance of the biorefinery systems); *iii*) combination of final products (10 biorefinery configurations are generated from the combination of 9 final products); and *iv*) assessment of sustainability criteria (*i.e.*, non-renewable energy use (NREU), greenhouse gas (GHG) emissions and economics). The used approach compares processing options and operation conditions to identify those combinations of production processes that minimize NREU and GHG emissions. In a second step, 10 integrated biorefinery concepts are selected and compared with respect to their environmental and economic performances. The impacts of choosing different microalgae species, locations, and nutrient sources were also studied as part of the scenario analysis. The results showed that the biorefinery systems with the best economic and environmental performances are those where microalgae oil-free cake is used as nutrient for substitution of animal feed and where lipids are used for substitution of vegetable oils. The worst economic and environmental performances of biorefineries were obtained when microalgae oil-free cake is anaerobically digested to biogas and lipids are converted to either biodiesel or green diesel. Regarding the cultivation technologies for the biorefinery systems with the best performance, favorable environmental results were obtained for flat panel photobioreactors (FPPBRs), followed by open ponds (OPs), vertical photobioreactors (VPBRs) and horizontal photobioreactors (HPBRs). In contrast, the best economic results were found for FPPBRs followed by VPBRs, HPBRs and OPs.

1. Introduction

Microalgae are emerging as potential feedstock for a number of different industrial sectors such as food commodities, biofuels, biobased chemicals, fine organics, bioplastics, pigments, cosmetics and pharmaceuticals among others [16,19,25,36,43,61]. Additionally, their advantages over conventional forestry, agricultural crops and other aquatic plants have extensively been discussed in literature (*e.g.* [6,43]), some examples are: high aerial biomass productivity; high CO_2 fixation capacity; ability to grow in marginal land, seawater or wastewater; and ability to produce a large variety of valuable products. These advantages create an optimistic environment for their potential commercial exploitation which has been further stimulated by the development of policy frameworks supporting the use of non-food feedstocks. However, despite the large R&D investments for algal biofuels, many challenges have impeded their production and utilization at industrial scale. Brennan and Owende [6]) report six main limiting factors: *i*) microalgae composition; *ii*) photosynthetic efficiencies, *iii*) technological challenges (species cultivation, evaporation reduction and CO_2 diffusion losses); *iv*) high net energy use; *v*) lack of exemplary large-scale plants; and *vi*) usability of flue gases. The combination of these predominantly

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technological factors have resulted in high production costs [20,47,62,63], and in negative energy and CO_2 balances for microalgae biofuels production [7,12,24,31,39,51,56].

An alternative to these stand-alone algae-biofuels production systems is the multi-product biorefinery that uses efficiently all microalgae fractions which may result in improved economic and environmental performances ([19]; and [16]). In this line, the aim of this study is twofold: *i*) to select potential processing pathways and relevant technologies for microalgae valorization at industrial scale, and *ii*) identify the microalgae biorefineries configurations with the best combined techno-economic and environmental performance for future sustainable designs of biorefinery systems.

Section 2 contains the methodological description where the considered microalgae biorefinery systems and related technologies are first discussed, and then the comparative parametric screening method, based on environmental and economic criteria, is presented. The individual results for each technology and processing steps as well as the environmental and economic results for the whole microalgae biorefinery systems are presented and discussed in Section 3. Finally, the conclusions of this study are presented in Section 4.

2. Methodology

This study is based on the sequential application of three methods: process design, environmental life cycle assessment (LCA) and economic evaluation. The integration of these approaches allows to simultaneously comparing multiple processing pathways, technologies, process conditions, and products basket under equivalent conditions. This analysis is conducted from a life cycle perspective having as primary selection criteria the potential environmental impacts and the economics of the whole value chain. This approach ensures that all systems are harmonized and comparable by using consistent process design stages, data-quality, system boundaries and functional units. The three used methods are described below.

2.1. Process design and data sources

The biorefinery systems here considered are composed of four processing stages as shown in Fig. 1: *i*) microalgae cultivation, *ii*) culture dewatering, *iii*) lipids extraction and purification, and *iv*) fractions upgrading. For each stage, the most representative technologies were selected from literature, and their respective process models for mass and energy balances were developed accordingly. The flue gas stream could be obtained from CO_2-intensive industries like oil refineries, natural gas processing, and chemical industries dedicated to the production of cement, ammonia, ethylene oxide and hydrogen. The CO_2 content of these streams varies from 4 to 19% (dry weight) for power plants and cement production to nearly 100% for some selected processes within chemical industries [37]. For this study only flue gas streams containing pure CO_2 (free of contaminants) are here considered. That is the case for industrial processes like natural gas sweetening, ammonia production, and coal and oil gasification, where CO_2 is separated in an economical way and where the separated CO_2 is vented [37].

For microalgae cultivation, four technologies were analyzed: open ponds (OPs), horizontal photobioreactors (HPBRs), vertical stacked PBRs (VPBRs) and flat panel PBRs (FPPBRs); while for downstream processing, five sequential steps were considered: medium dewatering, microalgae cell disruption, concentrate drying, oil extraction and final conversion to energy carriers. For each of these five steps, different options were studied: three sequential dewatering stages (*flocculation*, *centrifugation* and *filtration*), three cell disruption technologies (*high pressure homogenization*, *sulphuric acid treatment* and *ultrasonication*), three lipid extraction systems (*dry organic-*, *supercritical CO_2–* and *wet- extraction*) and five conversion systems to energy carriers (*sequential esterification/transesterification*, *simultaneous extraction and transesterification*, *catalytic hydrotreating* and *anaerobic digestion*). These process alternatives lead to 9 potential final products: *dry microalgae biomass* (*MABM*), *microalgae oil for vegetable oil substitution* (*MAO-VO*), *microalgae oil for fish oil substitution* (*MAO-FO*), *microalgae oil-free cake for fishmeal substitution* (*MAC-FM*), *microalgae oil-free cake for soybean meal substitution* (*MAC-SBM*), *green diesel* (*GD*), *biodiesel* (*BD*), *glycerol* (*Gly*), and *biogas* (*BG*). Furthermore, based on these 9 final products, a total of 10 biorefinery configurations (*i.e.* combination of final products) can be generated as shown in Table 1 (mass flows are also included in the table using a calculation base of 100 kg dry microalgae/h).

2.1.1. Cultivation technologies and microalgae species

Despite significant progress on multiple reactor designs for microalgae cultivation, there is no consensus on which cultivation technology is preferable due to trade-offs between technical, economic and environmental aspects [53]. In general terms, OPs are of lower capital and operation costs (*i.e.* simpler construction and operation) compared to PBRs. On the other hand, PBRs offer advantages on culture density, microalgae productivity, nutrients uptake efficiency, and controllability. For this study, four of the most extensively analyzed cultivation technologies are considered: OPs, HPBRs, VPBRs and FPPBRs. The basic designs for these technologies were adapted from literature [42,47,58] as described in the Supplementary material (Section S1). Furthermore, appropriate operating conditions and arrangement of reactors were selected based on a parametric analysis of the length and number of units *vs.* power requirements and electricity consumption for different gas flow rates in order to minimize the energy requirements (Supplementary material, Figs. S.1.1 to S.1.3). For example, for OPs the power consumption per cultivation unit was found to exponentially increase with the length of the pond while the number of cultivation units (for specific conditions of production scale, productivity and culture density) was exponentially decreased with the length of the pond. Consequently, there is an optimal length (and in consequence width) in which the total power requirements are the lowest. In this case, the selected length was 650 m, which is in line with the system studied by [42]. Similar findings (*i.e.*, optimal dimensions) were obtained for HPBRs and VPBRs for which the optimal length of the tubes are around 2000–2500 m and 1500–2500 m, respectively, depending on the productivity and culture concentration conditions. However, the number of units required significantly decreases at around 4000 m (for HPBRs) and 3500 m (for VPBRs). For VPBRs an arrangement of 8 vertical tubes with 14-L junctions was considered as one cultivation unit. In the case of FPPBRs, an arrangement of 35 sequential flat panels of 0.03 m thickness was considered as one cultivation unit. A ratio of 1.0/2.0/0.5 for height/width/distance-between-panels was kept [47], and the height is selected as the independent variable to calculate the power requirements. Similarly to the previous cases, the number of units exponentially decreases with height of the panels while the power requirements per cultivation unit exponentially increase with the height of the panels.

As part of the analysis of the power consumption in the cultivation stage, an additional operation mode is considered, namely: energy savings mode. In this case, two actions are considered to occur simultaneously: the mixing velocity is slowed down by 25% and the flue gas sparging stops during 10 h/day (nights and periods of low productivity).

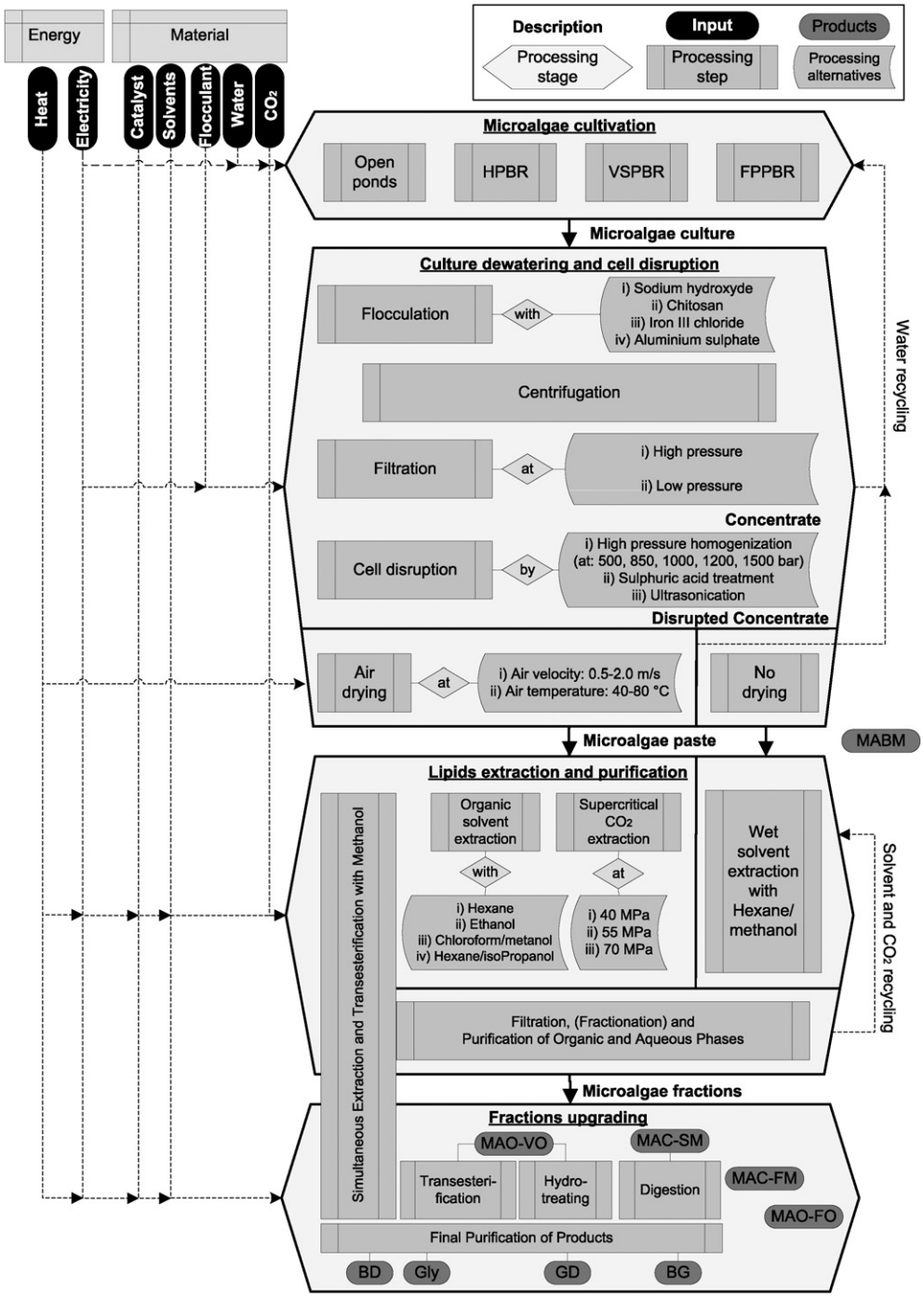

Fig. 1. Systems overview of microalgae biorefineries including technologies, processing conditions, and most representative mass and energy flows. MABM: dry microalgae biomass; MAC-FM: microalgae oil-free cake for substitution of fishmeal; MAC-SBM: microalgae oil-free cake for substitution of soybean meal; MAO-VO: microalgae oil for substitution of vegetable oil; MAO-FO: microalgae oil for substitution of fish oil; BG: biogas; GD: green diesel; BD: biodiesel, Gly: glycerol.

Microalgae productivity and culture density are two process variables with direct effects on environmental and economic performances of cultivation technologies and downstream processing. These two parameters vary significantly due to several factors, like microalgae species, geographical location (*i.e.*, light intensity), cultivation technology, carbon dioxide availability (*e.g.* source, type of industry generating flue gas) and purity (*i.e.* concentration and presence of contaminants), and cultivation conditions (pH and temperature) among others. Most techno-economic and environmental studies on microalgae-based products consider single values for these two variables. Here, two performance conditions are studied, namely: *high performance* (aimed to represent favorable conditions) and *low performance* (aimed to represent unfavorable conditions),[1] as shown in Table 2. Additionally, nutrient requirements (carbon, nitrogen, phosphorus, *etc.*) and presence of contaminants might also affect the global economic and environmental performance of biorefinery systems. Here nutrient requirements are considered as a function of the biochemical and elemental composition of the microalgae species. Two species are considered, *i.e. Nannochloropsis* sp. and *Scenedesmus* sp. The former is used as base-case and therefore discussed through the paper, while

[1] Favorable and unfavorable conditions are expected to be different for each cultivation system when operated at similar conditions. Hence, different values of aerial productivity and culture density are used [58].

Table 1
Biorefinery configurations and mass flows of products (in kg/h)[a] for the 10 biorefinery systems[b].

	BR1	BR2	BR3	BR4	BR5	BR6	BR7	BR8	BR9	BR10
Dry microalgae biomass	100	–	–	–	–	–	–	–	–	–
Microalgae oil-free cake for substitution of fishmeal	–	74.3	–	–	74.3	–	–	74.3	–	–
Microalgae oil-free cake for substitution of soybean meal	–	–	67.8	–	–	67.8	–	–	67.8	–
Microalgae oil for substi-tution of vegetable oil	–	23.1[c]	23.1[c]	23.1[c]	–	–	–	–		–
Microalgae oil for substi-tution of fish oil	–	–	6.5	–	–	6.5	–	–	6.5	–
Green diesel	–	–	–	–	18.8[c]	18.8[c]	18.8[c]	–	–	–
Biodiesel	–	–	–	–	–	–	–	19.1[c]	19.1[c]	19.1[c]
Glycerol	–	–	–	–	–	–	–	2.4	2.4	2.4
Biogas[a] (in Nm^3/h)	–	–	–	14.9	–	–	14.9	–	–	14.9

[a] All flows are in kg/h except for biogas (in Nm^3/h).
[b] Calculation base: 100 kg dry microalgae/h.
[c] Main (energy) product of the biorefinery system.

the latter is strictly considered in the scenario analysis (see Section 2.4). Representative compositions for *Nannochloropsis* sp. and *Scenedesmus* sp. are based on literature as shown in Table 3 [10,24]. Detailed biochemical and elemental compositions are available in the Supplementary material, Section S2. The presence of contaminants in the CO_2 stream might have negative effects on the cultivation stage, leading to potential lower productivities and concentrations of biomass, therefore the low performance conditions are also included as part of the parametric study.

2.1.2. Supply of carbon dioxide

Large-scale microalgae biorefineries will require constant and sufficient supply of CO_2 which may be provided by flue gas streams from CO_2-intensive industries as described in Section 2.1. Depending on the source of CO_2, these flue gas streams might contain CO, NOx, SO_2 and trace metals which may be toxic for microalgae growth [49]. For example, a SO_2 concentration exceeding 60 ppm may cause adverse effects for microalgae cultivation [38]. The inhibitory effect of these contaminants could be avoided by three possible actions: *i*) using carbon capture technologies [38], or *ii*) selecting sources with low levels of impurities, or *iii*) using microalgae species resistant to contaminants [38,49]. The latter cases are preferable due to the high energy requirements of a CO_2 purification system and their associated greenhouse gas (GHG) emissions. These requirements would represent around 30–35% of both the microalgae energy content[2] and of the CO_2 fixed by the microalgae[3] (Chisti, 2007; [34,35,39]) when compared to the non-renewable energy use (NREU) and GHG emissions of associated to the cultivation stage. Furthermore, species like *Nannochloris*, *Nannochloropsis*, *Chlorella*, *Dunaliella tertiolecta*, *Scenedesmus obliquus*, *Tetraselmis* and *Cyanidium caldarium* have been proven to tolerate typical concentrations of SO_2 and NO_X from simulated flue gas streams [38,49]. However, for this analysis only flue gas streams containing pure CO_2 are considered as described in Section 2.1. Thus, the costs, NREU and GHG emissions from CO_2 purification (which is out of the scope of this analysis) is not included in the model. In consequence, the supply system is considered to consists of compressors, pipelines, junctions and spargers which are specific for each cultivation technology (*i.e.*, OPs, HPBRs, VPBRs and FPPBRs). The inner diameter of the pipeline depends on three parameters: *i*) production scale (the calculation base is 100 kg dry microalgae/h), *ii*) CO_2 requirements by the microalgae specie (affected by the elemental composition), and *iii*) CO_2 usage efficiency (75% for OPs and 98% for PBRs, see Supplementary material, Section S3 (eq. S1–S10)). Based on these conditions, an inner pipeline diameter of 0.3 m was selected to ensure low pressure losses and a flue gas velocity of 1.5–2.5 m/s.

This approach can be further extended to analyze flue gas streams from other carbon-intensive industries (*e.g.* petroleum refineries, coal fired plants, and cement where the CO_2 concentration ranges from 4 to 30 vol.% [37,38]) by recalculating the power requirements and the carbon supply conditions (*e.g.* including a CO_2 purification system). However, these type of streams could also contain contaminants that might be harmful for the microalgae species. For example, the efficiency removal of pollutants from flue gas streams may be as low as 6% and as high as 99.4% depending on the type of pollutants and used technologies [60]. Furthermore, the presence of contaminants in the flue-gas stream may affect not only the microalgae cultivation performance, but also the quality of the final products. In the latter case, additional purification steps of the microalgae-based products might be needed according to their expected application. Here, it is assumed that all microalgae-based products, especially those used as substituents of animal feed and vegetable oils (*i.e.* MAO-VO, MAO-FO, MAC-FM, MAC-SBM –see Table 1) are free of contaminants, which is in line with the above made assumption that only flue gas streams containing pure CO_2 are considered.

2.1.3. Dewatering technologies

The culture concentrations achieved during the cultivation stage are 0.2–0.4 g/l (for OPs) and 1.5–2.5 g/l (for all PBRs), as defined for the *low performance* and *high performance* conditions, respectively (see Table 2). Microalgae is dewatered in three sequential steps: flocculation (up to 2.0% total suspended solids (TSS)), centrifugation (to 16.0% TSS) and filtration (to 40.0% TSS). For the first dewatering step, four flocculants are considered, *i.e.* sodium hydroxide [66], chitosan [3,46], iron III chloride [33], and aluminum phosphate [67]. The flocculants requirements and the process efficiency are calculated based on experimental data (correlations are provided in Supplementary material, in Section S3, eq. S11–S15). Furthermore, the power requirements for centrifugation and filtration depend directly on the concentration factor, volumetric flow and specific energy requirements for each dewatering equipment [64]. Average values of energy requirements for pressure filtration, vacuum filtration, and centrifugation of algae biomass dewatering are 1692, 7245, and 11,880 kJ/m^3 respectively [45].

2.1.4. Cell disruption

This process facilitates the release of intracellular products. Three technologies are considered:

i) High-pressure homogenization: In this mechanical process the cells suspension is pumped at high pressure through a narrow orifice of a valve and is then released into a lower pressure chamber. This process requires high pressure conditions and repeated loops to achieve high efficiencies per volumetric unit of processed microalgae culture [22]. The total power requirement is a function of three process variables: number of loops (to achieve at least a process efficiency of 96%), volumetric flow and operation pressure. For the latter, five operation pressures are

[2] Lower heating values (LHV) are calculated based on the microalgae composition [2], *e.g.*, 20–25 MJ/kg dry microalgae.
[3] Assuming a theoretical consumption of 1.83 kg CO_2/kg dry microalgae for an average microalgae specie composed of 50% carbon.

Table 2
Microalgae productivity and culture concentration for high and low performance conditions for *Nannochloropsis* sp.

	OPs		HPBRs		VPBRs		FPPBRs	
	Low	High	Low	High	Low	High	Low	High
Parameter	Performance	Performance	Perf.	Perf.	Perf.	Perf.	Perf.	Perf.
Productivity (g/m^2-day)	10	30	18	45	20	50	22	55
Culture density (g/l)	0.2	0.4	1.5	2.5	1.5	2.5	1.5	2.5

considered: 500, 850, 1000, 1200, 1500 bar. Used correlations for power consumption are provided in Supplementary material, Section S3, eq. S16–S60.

ii) Sulphuric acid treatment: This chemical method has been applied to various types of biomass (esp. for lingocellulosic materials) as preparatory step for bioethanol production. The process efficiency was reported to depend on three main variables [23]: temperature, sulphuric acid concentration and operation time. The adjusted function relating operation time and efficiency (at 160 °C and 8 vol.%. from Halim et al. [22])) is included in Supplementary material, Section S3, eq. S21. The power requirements for stirring are calculated using an electricity consumption factor of 0.4 kW/m^3 [48].

iii) Ultrasonication: In this process, the microalgae are subjected to sonic waves which create cavitation microbubbles and impart kinetic energy into the surface of the cells eventually leading to cells rupture. The process efficiency depends mainly on the operation time, while the power consumption depends on the volumetric flow and operation time [22]. The adjusted function for power consumption is included in Supplementary material, Section S3, eq. S22–S23.

2.1.5. Air drying

This step is done prior to lipids extraction, however it is not the case when wet extraction is used. Here, a convective hot-air drier with natural gas air heaters is considered. The heat losses are estimated as 5% for the whole process, *i.e.* for air heating and heat transfer from the air to the concentrated microalgae paste. The final biomass concentration, in all cases, is set at 80 wt.%. The air drying process is based on the Weibull's probabilistic model for a convective hot air dryer [59]. The power requirement depends on (initial, final and equilibrium: 98 wt.%) biomass moisture, operation time, drying temperature, and air flow rate. The used model for power consumption is included as Supplementary material, Section S3, eq. S24–S29. The model is analyzed within the process conditions of 0.5–2.0 m/s (air flow rate), 40–70 °C (temperature), and 60-90 wt.% (microalgae moisture).

2.1.6. Lipids extraction

Six different methods are considered for lipids extraction, of which four are organic solvent extractions of dry biomass: *i*) hexane [18,21,23], *ii*) ethanol [18,21,23], *iii*) chloroform/methanol (2:1 v/v) [21,23,40], *iv*) hexane/isopropanol (3:2 v/v [21,22], *v*) dry biomass supercritical CO_2 extraction under three different operating pressures (40, 55, and 70 MPa) [1,22], and *vi*) a wet solvent extraction using hexane/methanol (3:1 v/v) [11].

For the four first cases, the efficiency factors as well as the solvent usage and solvent losses factors were calculated based on data reported by [23]. Missing information was completed from [18,21,57]. The used extraction efficiencies were: 96, 91, 98 and 94% respectively; while the losses factors were: 2.4, 5.0, 3.6 and 2.0 g solvent/kg dry microalgae. The power consumption for solvent extraction is based on an electric stirring system with an energy agitation factor of 2.8 kW/m^3 [48].

For wet extraction, the solvent use ratio and the solvent loss ratio were correlated based on the operation temperature and the process efficiency [11] which define the number of cycles. Here, a recovery efficiency of 90% (reported values are around 75–95%) was used with a temperature of 90 °C and a pressure of 1.4 MPa. For these conditions, the solvent loss ratio is 1.6 g solvent/kg dry microalgae. The power consumption for wet extraction is based on an electric stirring system with an energy agitation factor of 2.8 kW/m^3 similar to the dry solvent extraction process [48].

The wet and dry solvent extraction processes generate three phases that must be further separated, *i.e.* microalgae cells, aqueous phase and organic phase. For each solvent case (or mix of solvents), a different downstream process configuration is required. For example, microalgae cells can, in all cases, be withdrawn from the extraction medium by using a filtration unit (either at high pressure or vacuum pressure) whose efficiency was set as 95%. For purifying the aqueous phase, at least one or two distillation columns are required depending on the solubility of the solvent(s) in water. For the organic phase, one distillation column may be sufficient since the distillation product can be either pure solvent (when only one solvent is used) or a mix of used solvents (when two solvents are simultaneously used). Thus a high purity lipids stream is obtained at the bottom from the last distillation stage in the purification of the organic phase; and the solvent(s) are recovered from the top of the distillation columns of the purification of both phases (aqueous and organic). Hence, the downstream process configuration may include up to 4 distillation columns according to each case. Since the energy requirements of the downstream process of the lipids extraction step are inherent to each solvent (or mix of solvents)

Table 3
Typical biochemical composition of microalgae expressed as dry matter basis (%).
Adapted from [10]; and [24].

Microalgae fraction	Nannochloropsis	Scenedesmus
Lipids	40.0	25.0
Carbohydrates	10.0	17.0
Proteins	35.0	40.0
Phenolics	2.0	2.0
Other organics	4.5	5.0
Minerals	8.5	11.0

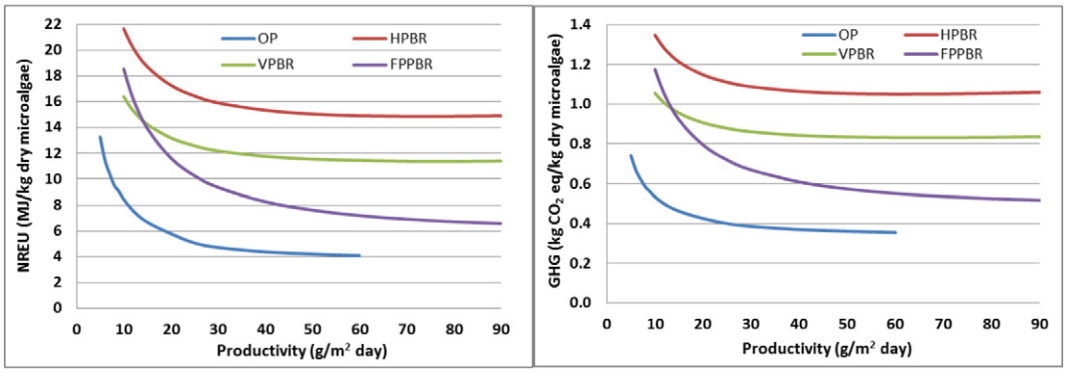

Fig. 2. NREU and GHG emissions for microalgae cultivation technologies (including nutrient requirements and construction materials) as function of productivity (at high concentration conditions and saving operation mode).

used, the energy supply as well as the purification efficiency are separately calculated using the process simulator Aspen Plus (Aspen Technologies, Inc., USA) based on the mass balances obtained from the extraction process.

For supercritical extraction, the process efficiency was reported to depend on the pressure operation and time. These variables were correlated based on experimental data [23], and the adjusted functions are included in Supplementary material, Section S3, eq. S30–S32. The minimum extraction efficiency was here set at 95%. Then, the power consumption for supercritical extraction is calculated based on adiabatic compression of CO_2. Lipids are finally recovered by decompression. Here, the only considered process inputs are the power requirements (calculated as an adiabatic compression of CO_2) and the CO_2 losses that occur during the process. Furthermore, if a flue gas stream containing contaminants is used for the cultivation stage, those contaminants might be transferred to the different microalgae fractions resulting potentially in additional purification steps which would be case dependent according to the type of contaminants.

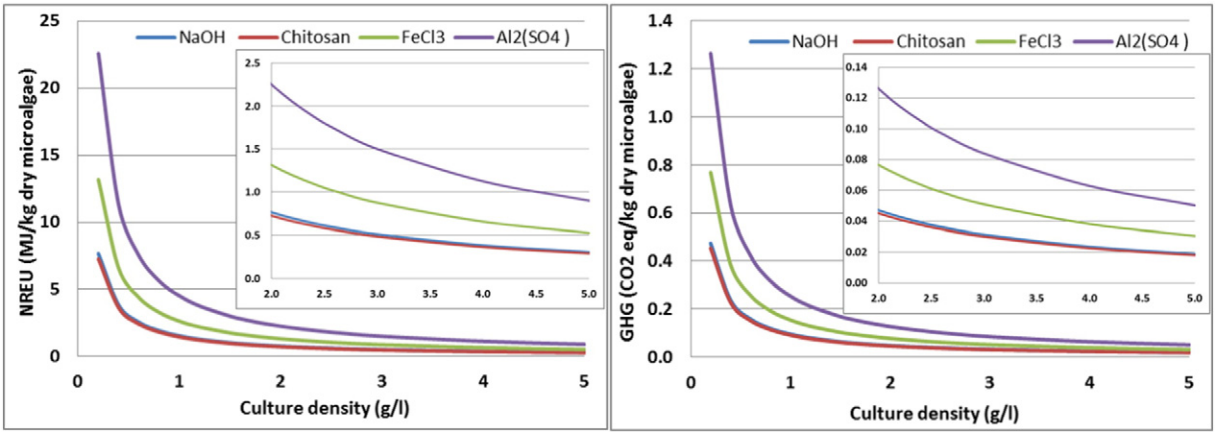

Fig. 3. NREU and GHG emissions for microalgae dewatering (flocculants + power for: flocculation, centrifugation and high pressure filtration). The inset shows a zoom in of the y-axis at higher culture density.

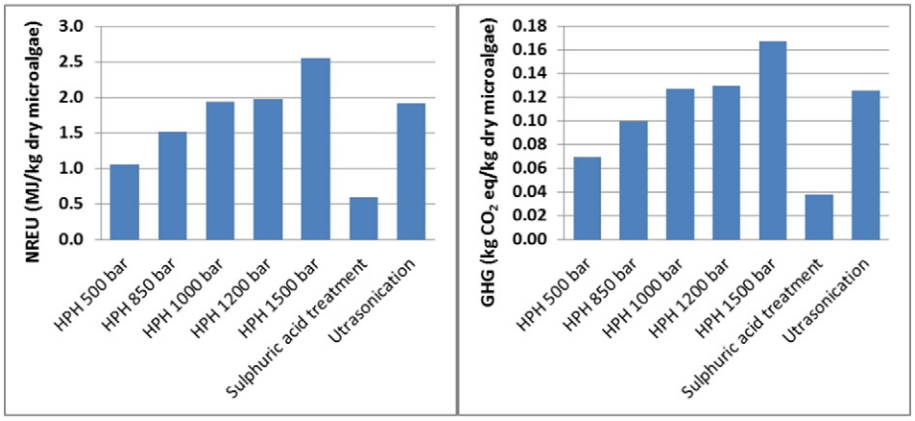

Fig. 4. NREU and GHG emissions for cell disruption technologies. *HPH: high pressure homogenization.

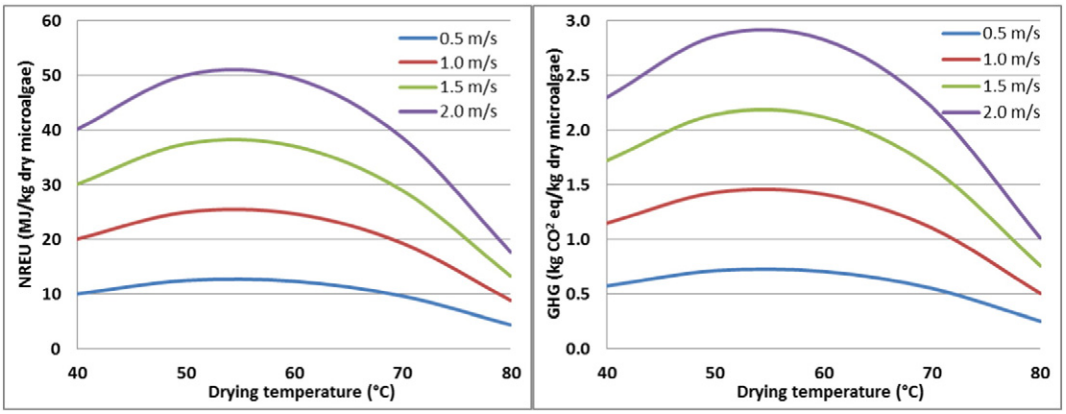

Fig. 5. NREU and GHG emissions for air drying. Parametric lines are for air velocity flows.

2.1.7. Fractions upgrading

After extraction, the lipids fraction can be subjected to: *i*) fractionation to separate higher value fatty acids as fish oil substitute, *ii*) transesterification into biodiesel, or *iii*) upgrading into green diesel by hydrotreatment.

Here, lipids fractionation with urea is analyzed considering four different mass ratios of urea/lipids (1/4, 1/3, 1/2 and 1/1), in order to separate the polyunsaturated fatty acids where different efficiencies are achieved for each urea/lipids mass ratio (93.1, 91.3, 86.2, and 47.1% respectively) [54].

The lipids fraction can further be directly converted into biodiesel. Here, a two-step esterification/transesterification reactor system with heat integration was adapted from [55] and simulated using Aspen Plus, for three catalytic conditions (H_2SO_4, NaOH, KOH).

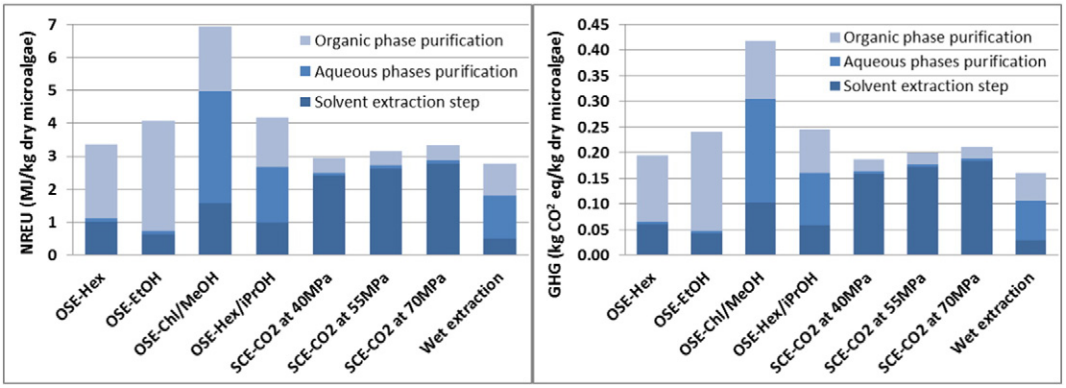

Fig. 6. NREU and GHG emissions for lipids extraction technologies. OSE: organic solvent extraction; SCE: super critical extraction; Hex: hexano; EtOH: ethanol; Chl: chloroform; MeOH: methanol; PrOH: propanol.

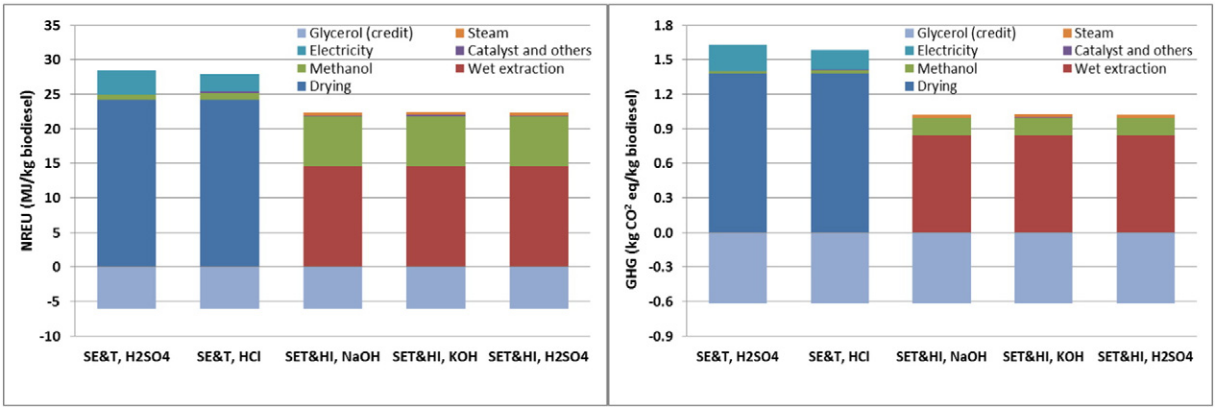

Fig. 7. NREU and GHG emissions for biodiesel production technologies. SE&T: simultaneous extraction and transesterification process; SET&HI: sequential esterification/transesterification system with heat integration.

An alternative route for biodiesel production is the simultaneous extraction and transesterification with methanol on dry biomass [41]. In this case drying is still needed, but during the extraction process methanol is used as both solvent and reactant. However, an acid is added as catalytic agent. Here, two catalysts (*i.e.* H_2SO_4 at 0.2 M and HCl at 0.4 M) are considered. The operation temperature and required operation time were fit as functions of the selectivity based on experimental data [41]; the adjusted functions are included in Supplementary material, Section S3, eq. S33–S35. The energy and material balances of the simultaneous extraction-reaction process are separately calculated by using Aspen Plus simulation.

For green diesel production, the so-called NExBTL process (Next Generation Biomass to Liquid, developed by Neste Oil, Porvoo, Finland) is considered [52]. This is a catalytic hydrotreating process producing three separate branched chain paraffins, fuel gas and biogasoline. Here, the green diesel production is considered as a black box for which the mass and energy balances are directly obtained from ([52], based on the case "European (as Porvoo)").

The microalgae oil-free cake is considered to be upgraded by anaerobic digestion to produce biogas. The models for the anaerobic digestion plant and for the biogas purification plant are based on results reported by [12]. The original model uses industrial data and represents the engineering state-of-the-art for wastewater treatment applications. The methanization yield exceeds 75% of its maximal biological potential, producing 0.201 m^3 of biogas at 96% purity, by each kg of microalgae oil-free cake, and with electricity requirements of 0.2162 kWh. Liquid digestates (composed of organic and mineralized matter), obtained during the anaerobic digestion process, have a concentration per cubic meter of: 120.01 kg of carbon (C), 4.5 kg of nitrogen (N), 0.607 kg of phosphorous (P) and 0.495 kg of potassium (K), respectively. The model also gives environmental credits to the digestates since they can be recycled to the microalgae cultivation media thereby reducing the global nutrient requirements [12].

2.2. Life cycle assessment (LCA)

The LCA follows the ISO-14040 and ISO-14044 guidelines [28,29]. The goal of this part of the study is twofold: *i*) identify the technologies and operation conditions that lead to the best environmental performance for each processing step, and *ii*) evaluate the environmental performance of different microalgae biorefinery configurations, in order to identify promising pathways for microalgae processing. To this end, two impact categories are here studied in a cradle-to-gate analysis: primary energy use (calculated as non-renewable energy use, NREU) and global warming potential (determined as greenhouse gases emissions, GHG). For this analysis the "gate" depends on the biorefinery configuration analyzed in each case

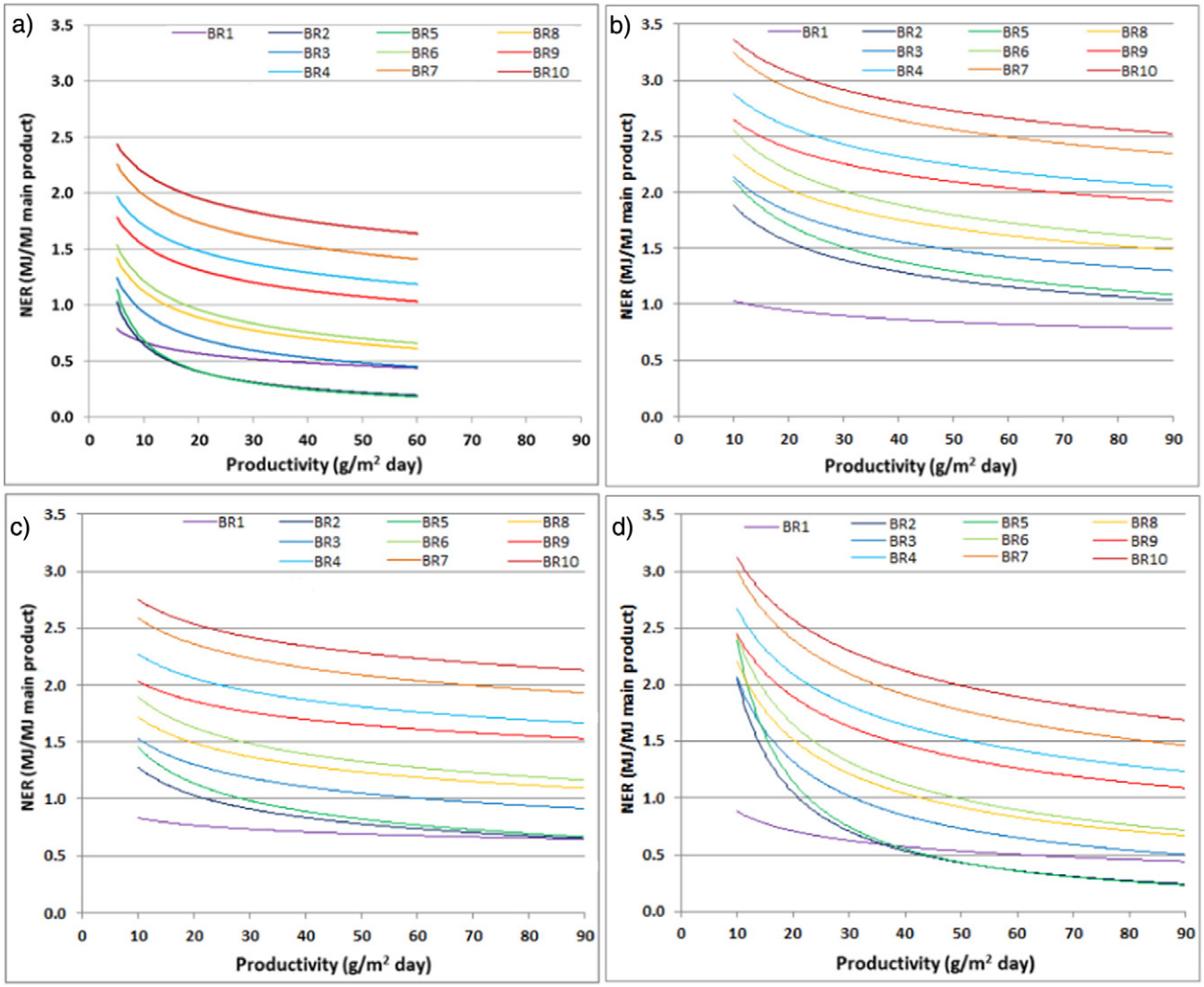

Fig. 8. NER of microalgae biorefinery systems as function of productivity: a) OPs, b) HPBRs, c) VPBRs and d) FPPBRs.

(see Fig. 1). The functional unit is defined as 1 MJ of the main (energy) product (*i.e.* microalgae biomass, microalgae oil, biodiesel or green diesel). For all biorefinery configurations, the environmental impacts include inputs from: nutrients, PBRs design materials, power consumption for culture circulation, gas distribution, and downstream processes. Where appropriate, the system expansion approach is used to assign credits to by-products (see Table 1). Credits are given to the mix of microalgae oil-free cake and omega-3 fatty acids as a substitute for fishmeal, and an 80% substitution is assumed for microalgae oil-free cake, *i.e.* 1 kg of microalgae oil-free cake is equivalent to 0.8 kg of fishmeal [16]. Additionally, credits are given to microalgae oil-free cake and to omega-3 as substitutes for soybean meal (from Brazil) and for fish oil respectively [16]. Similarly, credits are given to the obtained biogas and glycerol.

Microalgae biorefineries can achieve a double purpose in terms of reduction of environmental impacts, *i.e.* production of renewable energy and/or materials, and mitigation of GHG emissions by making direct use of industrial CO_2 as carbon source. Given this characteristic of the microalgae system, the NREU and the GHG emissions are here expressed as "net energy ratio" and "net GHG ratio" to estimate the potential reduction of environmental impacts of a determined biorefinery configuration. These two ratios are expressed with respect to the functional unit, *i.e.* per 1 MJ of the main (energy) product.

The Net Energy Ratio (NER) is here defined as the ratio of the NREU associated to the microalgae biorefinery life cycle (*i.e.* the primary energy used in the whole value chain of a specific biorefinery) with respect to the total energy output generated in the microalgae biorefinery as shown by Eq. (1). Thus, a NER < 1.0 means that more energy is embodied in the total output of products than the non-renewable energy that is invested in the entire biorefinery system. In this case lower values are preferable.

The Net GHG Ratio (NGHGR) is defined as the ratio of GHG emitted associated to the entire microalgae biorefinery life cycle (expressed as equivalents of fossil CO_2) with respect to the CO_2 fixed by the microalgae during the cultivation stage as shown by Eq. (2). Since credits are not given to the industrial CO_2 emissions avoided, values of NGHGR < 1.0 mean that the GHG emissions generated by the biorefinery configuration are lower than the total CO_2 consumed by the microalgae during the cultivation stage (1.96 kg CO_2 eq./kg dry microalgae in the case of *Nannochloropsis*). Thus, both assessment parameters, NER and NGHGR, allow to easily comparing not only the environmental performance but also the potential reduction of environmental impacts of different biorefinery configurations. Here lower values also reflect a higher potential reduction of environmental impacts.

$$NER = \frac{NREU\ associated\ to\ the\ microalgae\ biorefinery\ life\ cycle}{total\ energy\ output\ generated\ in\ the\ microalgae\ biorefinery} \quad (1)$$

$$NGHGR = \frac{GHG\ emitted(as\ CO_2\ equivalents)\ associated\ to\ the\ microalgae\ biorefinery\ life\ cycle}{CO_2\ fixed\ by\ the\ microalgae} \quad (2)$$

A comprehensive life cycle inventory (LCI) was assembled for each processing option based on the models described in Section 2.1 (see Table 1 for global outputs), and the NREU and GHG emissions were compared among the different biorefinery configurations developed. These two environmental impact categories were compared in three sequential steps: *i*) cultivation systems were analyzed as function of the biomass productivity for high and low culture densities (see Table 2), *ii*) the dewatering system was studied as function of the culture density, and *iii*) the different technological alternatives and processing conditions were directly compared for each processing step (*i.e.* cell disruption, drying, lipids extraction and fractions upgrading). The parametric analysis of technologies was initially performed considering 1 kg of dry microalgae since this is a common point (or intermediate product) for all systems, however the final comparisons are made using 1 MJ of main (energy) product as functional unit. The LCI data were derived from the Ecoinvent v2.2 database using specific processes for the Netherlands, where possible. Additionally, specific regional data for France, Brazil and China were also used for the scenarios analysis (see Section 2.4). The LCA was compiled using a spreadsheet model and SimaPro LCA software. Inputs and outputs in the LCI were assigned to the main (energy) product of each biorefinery configuration while the by-products (see Table 1) were credited, thereby reducing the total impacts.

2.3. Economic analysis

By analogy with the methodology for environmental assessment, the total production costs for the 10 microalgae biorefinery systems presented in Table 1 were calculated per MJ of main (energy) product. All economic data used was converted to 2012 Euros by using the *Chemical Engineering Plant Cost Index* (CEPCI) for processing technologies [9] and the *Consumer Price Index* (CPI) [17] for the commercial price of raw materials.

The capital costs for the cultivation technologies, dewatering systems and cell disruption equipment were adapted from [30,42,47]. The capital costs for the wet extraction and transesterification processes were adapted from ([15] and Sanchez et al., 2011) and normalized per kg biomass/h and per kg biodiesel/h respectively; while the capital costs for hydrotreating and anaerobic digestion were adapted from [14] and normalized per kg of lipid hydrotreated/h and per kg total solids/h, respectively. All capital costs are available in the Supplementary material (Table S.4.1., Section S4).

The power and natural gas costs were taken from (Key world energy statistics, 2012) while the steam costs were calculated assuming an energy conversion efficiency of 85% and 3% of depreciation for steam boilers. The land and nutrient costs were taken from ([14,47], respectively. The raw materials costs were obtained from [26] and the prices of all products were taken from [65]. The cost of utilities, raw materials and products costs are shown in Table S.4.2 in the Supplementary material (Section S4).

The estimation of the total production costs (TPC) is based on the method applied by [15]. The TPC is the total operating cost (TOC) over a year per functional unit (*i.e.* MJ main (energy) product/year); the resulting unit is €/MJ of main product.

The TOC is the sum of the operating cost (OC) (*i.e.*, utilities, labor, plant overhead, general & administrative, initial expenses, process start-up and others) and the fixed cost (FC). The FC is calculated from the depreciable capital (DC) with an annuity factor of 14.9%, based on an interest rate of 8% and a lifetime of 10 years [30]. The DC consists of the direct capital costs (DCC) (*i.e.*, equipment, installation, instrumentation & control, piping and buildings), indirect capital costs (ICC) (*i.e.*, maintenance & storage, engineering & supervision, spare parts and license fees) and initial expenses. The system of equations and used parameters are presented in Table S.4.3 in the Supplementary material. The production costs estimation includes the credits from the by-products (*i.e.*, microalgae oil for substitution of fish oil, microalgae oil-free cake for substitution of fishmeal, microalgae oil-free cake for substitution of soybean meal and glycerol) but does not consider the credits for CO_2 fixation neither for wastewater treatment; however these credits are further studied in the scenario analysis in two different cases, one for prices of the by-products (given the price volatility of oil) and one for the CO_2 fixation taxes (see Section 2.4).

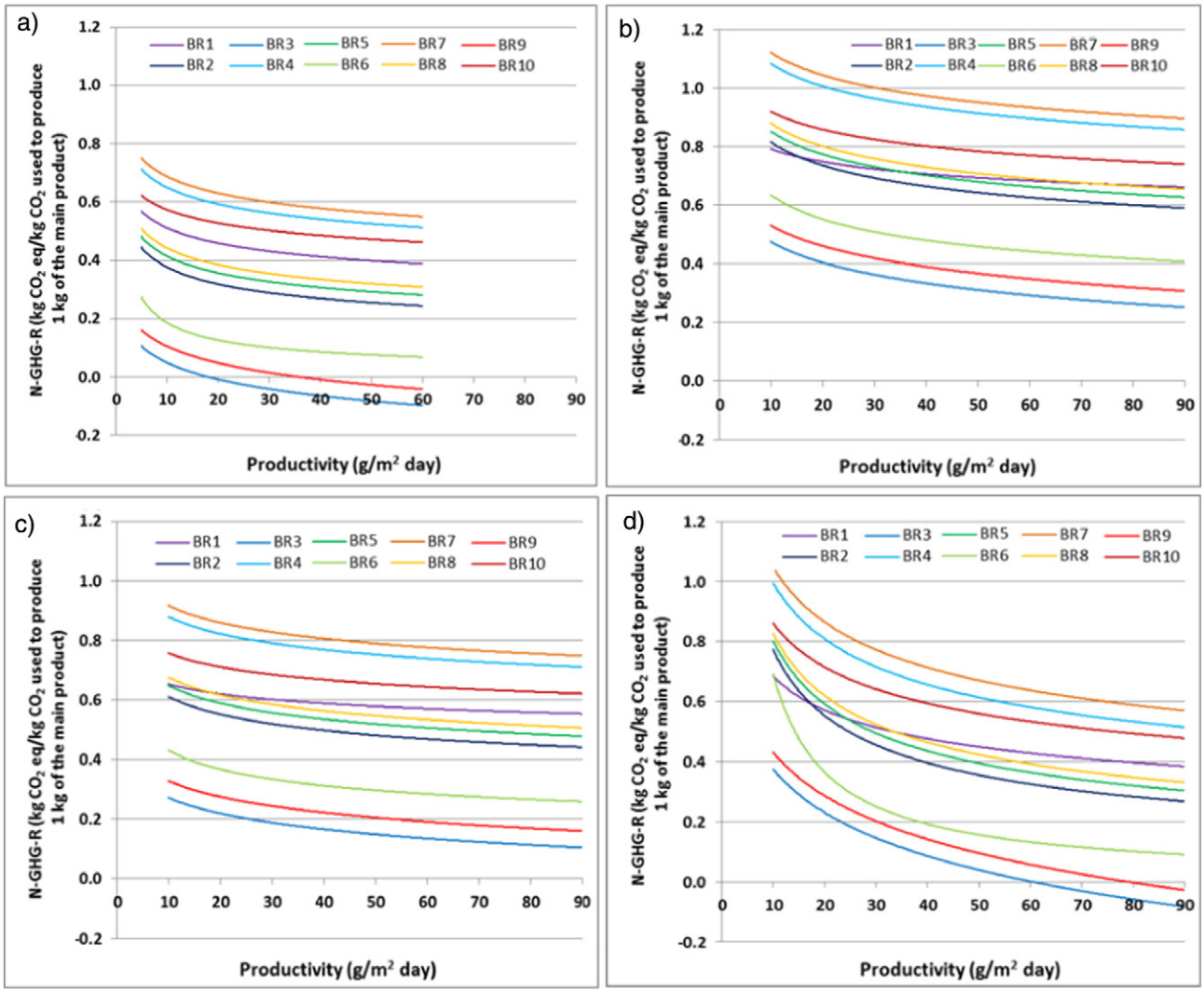

Fig. 9. NGHGR of the microalgae biorefinery systems as function of the productivity for each cultivation technology: a) OPs, b) HPBRs, c) VPBRs and d) FPPBRs.

2.4. Scenario analysis

Scenario analysis was conducted to determine the influence of four main factors on the environmental and economic performance of the ten biorefinery configurations:

- Microalgae specie: a second species, *Scenedesmus* sp., was considered by using specific biochemical and elemental compositions (see Tables S.2.1 and S.2.2 in the Supplementary material), with their respective cultivation performance (see Table S.2.3 in the Supplementary material).
- Geographical location: specific geographical conditions are usually considered by using different values of productivities and/or culture densities [30,47] since these parameters are affected by light intensity and light absorption efficiency; but these parameters are also affected by the cultivation technology and microalgae specie. Since the variation of these two parameters, productivity and culture density, is already integrated as a fundamental part of the methodology (see Section 2.1.), an additional consideration, *i.e.* the electricity mix (and its price), is here included. In this case, electricity mix of three countries with a significantly different combination of primary energy sources, and in consequence with different costs, are considered. Base case: the Netherlands; scenario cases: France, China and Brazil. For example, France has an electricity mix mainly based on nuclear power, while in the Netherlands and China the main primary energy sources are fossil fuels, and finally in Brazil the electricity mix is mostly composed by hydropower and fossil fuels [27].
- Wastewater as alternative source of nutrients: the average concentration of nitrogen and phosphorus in municipal wastewater for the Netherlands (42 mgN/l and 6.7 mgP/l) and France (52 mgN/l and 9.3 mgP/l) [50] were considered to determine the potential substitution of nutrients and the effect of the composition of wastewater in the environmental and economic performance of the biorefinery systems. Although, wastewater may contain contaminants (which change by types and concentration levels from place to place, *e.g.* NL *vs.* FR in this scenario) that can negatively affect the microalgae cultivation stage (and the quality of the microalgae based products), these effects have only been considered by the *low performance conditions* (*i.e.* low concentration and low productivity) as shown in Table 2. On the other hand, microalgae biomass could even be a beneficial alternative to efficiently remove a wide range organic contaminants from urban wastewater (including pharmaceuticals and personal care products, fire retardants, surfactants, anticorrosive agents, pesticides and plasticizers, among others) [44]. These potential positive effects have not either been considered in this study. It is important to acknowledge that those potential contaminants

Table 4
Qualitative comparison of NREU and GHG emissions for the 10 microalgae biorefinery configurations. "+" represents a positive environmental performance).

System	NREU (as NER)				GHG emissions[a] (as NGHGR)				Total
	OPs	HPBRs	VPBRs	FPPBRs	OPs	HPBRs	VPBRs	FPPBRs	
BR1	+	+	+	+	−	−	−	−	4
BR2	+	−	+	+	+	−	−	+	5
BR3	+	−	−	+	+	+	+	+	**6**
BR4	−	−	−	−	−	−	−	−	0
BR5	+	−	+	+	+	−	−	+	5
BR6	+	−	−	+	+	−	+	+	5
BR7	−	−	−	−	−	−	−	−	0
BR8	+	−	−	+	+	−	−	+	4
BR9	−	−	−	−	+	+	+	+	4
BR10	−	−	−	−	−	−	−	−	0

[a] For NGHGR, favorable values were considered to be lower than 0.4 given that most of the configurations and technologies lead to NGHGR < 1.0. Although 0.5 was initially considered as the upper value, the differences among the systems was not completely clear (as for 0.4) from a practical point of view. Bold "+" is indicated for systems with favorable results for both indicators, i.e. NREU and GHG. The total value in bold ("**6**") is for the highest number of favorable results for each BR system.

in the wastewater might also be transferred to the microalgae-based product which could lead to additional requirements in the downstream process for purification of the final products according to the intended application.

- Price of by-products: the total production costs of the microalgae biorefinery systems depend not only on the capital and operational costs but also on the used sale price of by-products (Section 2.3). Furthermore, since every biorefinery concepts has a particular distribution of products (see Table 1), this analysis is performed for each biorefinery system (BR1 is excluded because no by-product is generated) considering a price drop of 50% for each by-product (prices for base-cases available in Table S.4.2. in the supplementary material) as a function of the microalgae cultivation productivity for a selected technology based on the techno-economic and environmental screening described in Sections 2.2 and 2.3. This

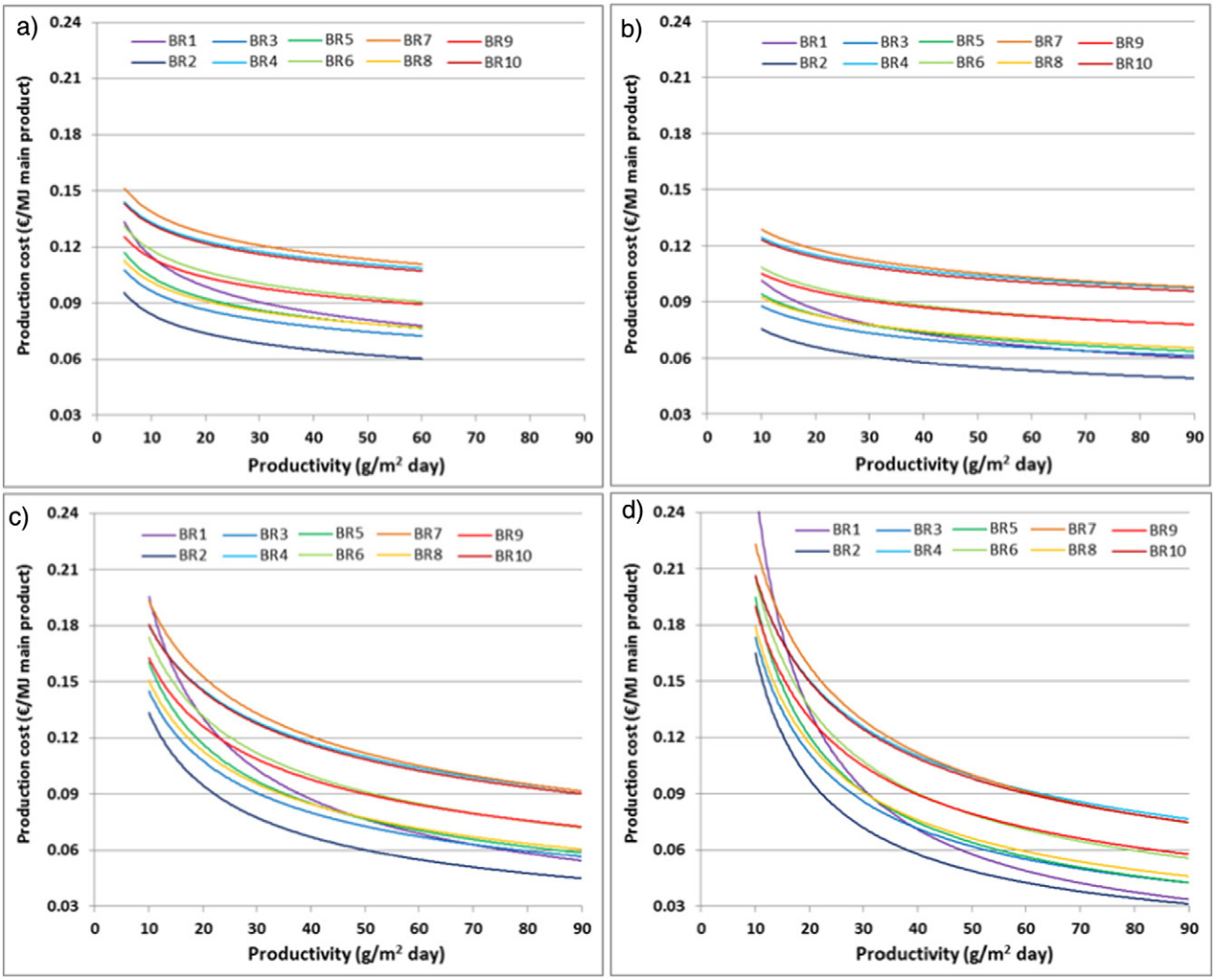

Fig. 10. Production costs of the microalgae biorefinery configurations as function of the productivity for each cultivation technology: a) OPs, b) HPBRs, c) VPBRs and d) FPPBRs.

Table 5
Economic ranking of biorefinery systems and categorization by groups.

Ranking	OPs	HBPRs	VPBRs	FPPBRs	Group
1	BR2	BR2	BR2	BR2	
2	BR3	BR3	BR3	BR1	I
3	BR8	(BR1)[a]	(BR8)[a]	BR3	
4	BR5	BR5	BR5	BR5	II
5	BR1	(BR8)[a]	(BR1)[a]	BR8	
6	BR9	BR9	BR9	BR6	III
7	BR6	BR6	BR6	BR9	
8	BR10	BR10	BR10	BR10	
9	BR4	BR4	BR4	BR4	IV
10	BR7	BR7	BR7	BR7	

[a] Numbers in brackets are for biorefinery configurations that vary slightly within the ranking.

drop of 50% aims to represent the price volatility on the total production costs of the main energy products. As a descriptive example, BR9 produces biodiesel as a main energy product and glycerol, MAO-FO, and MAC-SB as by-products. In this case, the commercial sale prices of glycerol, MAO-FO, and MAC-SB are reduced by 50% (one at the time) to determine the increment in the total production costs of biodiesel for BR9.

- CO_2 credits: economic credits are given to the CO_2 fixation that occurs in the microalgae cultivation stage since the base cases do not consider any economic credits for CO_2 fixation. The analyzed carbon credits are in the range 10–50 €/t of CO_2 fixed [15].

3. Results and discussion

3.1. NREU and GHG emissions of individual processing steps

The electricity consumption for culture circulation and gas distribution for the four cultivation systems (OPs, HPBRs, VPBRs, and FPPBRs) was calculated at low performance conditions (representing unfavorable conditions in the cultivation stage, *e.g.* presence of contaminants in the flue gas stream, or low light intensity, or inadequate pH or temperature) and high performance conditions (representing favorable conditions for microalgae cultivation), see Table 2. Hence two operation modes were considered: normal operation and energy saving operation as discussed in Section 2.1.1. (detailed results for power consumption are available in Figs. S.1.1–S.1.3 in the Supplementary material). The power requirements of OPs at low productivity conditions, in the normal operation mode, are 4 times higher than those at high productivity conditions. Microalgae cultivation in OPs consumes between 3 and 13% of their energy content (assuming a LHV of 24.2 MJ/kg for *Nannochloropsis* [2]). However, when the energy saving operation mode at high culture density and at high productivity conditions is considered, the power requirements can be reduced by 50%. Therefore, the subsequent cases discussed in this paper are analyzed considering in all cases the energy saving operation mode and high concentration conditions. The NREU and GHG emissions for the cultivation stage are shown in Fig. 2. In all cases, OPs have the lowest NREU and GHG emissions among the four cultivation systems considered, at both low and high productivities conditions, followed by FPPBRs, VSPBRs and HPBRs respectively.

The performance of the dewatering stage depends particularly on the flocculation step since the microalgae culture must be concentrated in this step up to 6% TSS regardless of its initial concentration. Typical values of the culture density for OPs are below 1 g/l while for closed systems these values are above 1 g/l. The NREU and GHG emissions of this stage are shown in Fig. 3 for the four flocculants as function of the culture density.

For cell disruption, three technologies are considered: ultrasonication, sulphuric acid treatment and high pressure homogenization (at 5 pressure conditions). The NREU and GHG emissions for each technology are shown in Fig. 4. The best results were obtained for sulphuric acid treatment with a significant difference compared to the other options. The NREU and GHG emission are both lower by 50% and 70% than those for high pressure homogenization (at 500 bar) and ultrasonication, respectively.

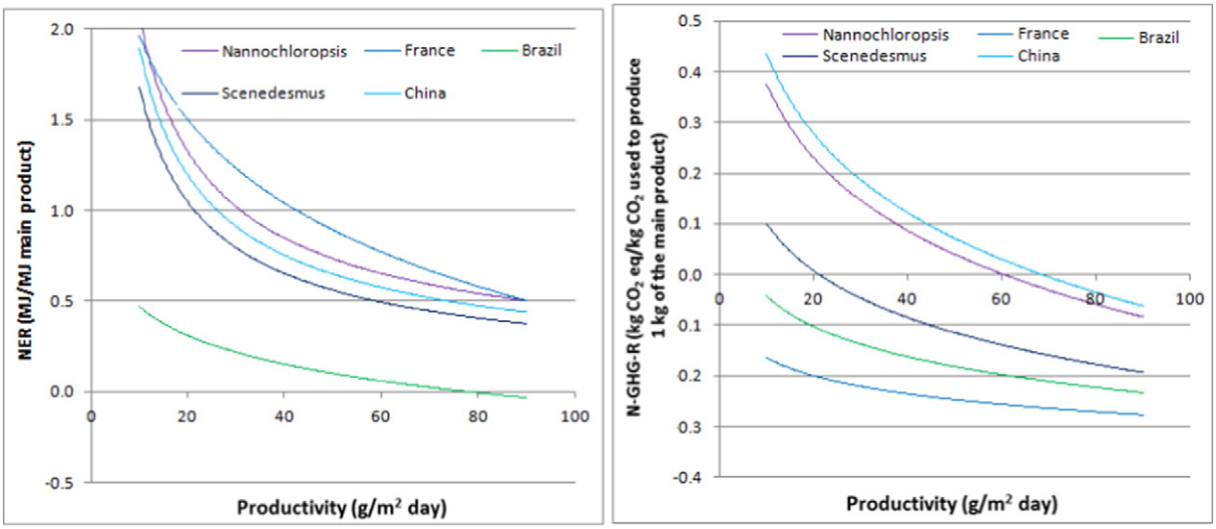

Fig. 11. Scenario analysis. NER and NGHGR values of the BR3 with FPPBRs using different microalgae species and at different locations.

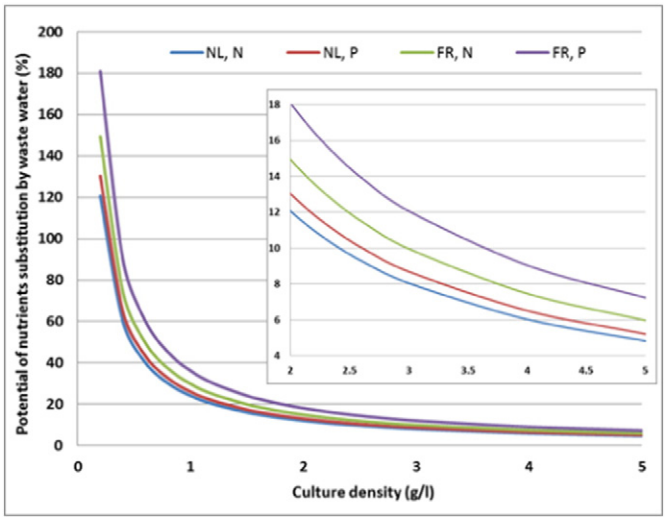

Fig. 12. Percentages of potential nutrients substitution with municipal wastewater, based on an averaged N and P content, as a function of the culture density. (The inset shows the trend at higher culture densities).

Microalgae lipids can be extracted by two alternative technologies: dry extraction or wet extraction (see Fig. 1). In the first case, drying is necessary. Fig. 5 shows the NREU and GHG emissions for the drying process at different air velocity flows and temperatures. Lower values of NREU and GHG emissions are obtained when low air flow rates are used at either low (<50 °C) or high (>70 °C) temperatures. For low temperatures, longer operation times are required leading to a better heat transfer efficiency; on the other hand, at high temperatures, the required mass flow of air decreases resulting in a reduction of the heating requirements. Here, the selected operation conditions are 75 °C and 0.5 m/s since higher temperatures may lead to microalgae metabolites degradation [13].

For lipids extraction, six alternatives are considered as described in Section 2.1.6. Fig. 6 shows the NREU and GHG emissions for the lipids extraction processes, where wet extraction has the lowest impacts among these technology options. It is important to notice that wet extraction does not require a previous drying step as the other cases.

Once the lipids are extracted, they can further be purified to obtain omega-3 by fractionation. Thus, three main possible products are identified after extraction (see Table 1 and Fig. 1): microalgae oil-free cake, microalgae oil and omega-3. Here two possible uses are considered for microalgae oil-free cake (as described in Section 2.2): *i*) as fishmeal substituent (but since fish have certain requirements of essential amino acids, omega-3 is not considered as an extra product in this case) and *ii*) as soybean meal substitution (given its high content of proteins, *i.e.* 50.4–51.4 wt.%, which is in line with the protein content of soybean meal 50–55% [4]) while omega-3 is used as replacement for fish oil. In the case of microalgae oil-free cake as substituent of fish meal, it has been described that the replacement levels can vary from 5 to 10% up to 25–40% with no significant differences in the growth or feed performances [32]. Thus, credits are given to the microalgae oil-free cake as potential substituent of fishmeal and soybean meal. It is important to notice that if the flue gas used for microalgae cultivation contains contaminants, these contaminants can potentially be transferred to the different fractions of microalgae biomass which could result in either additional purification steps (and in consequence in higher energy consumption and GHG emissions) or in lower replacements capacity (*i.e.* replacement of lower quality products).

Microalgae oil can further be upgraded to either biodiesel or green diesel as shown in Fig. 1.

For biodiesel production, two main processes are considered: *i*) a sequential esterification/transesterification system with heat integration (adapted from Sanchez et al., 2011) using either acid (H_2SO_4) or basic (NaOH or KOH) catalyst and *ii*) a simultaneous extraction and transesterification process. In the latter case, a drying step is required. The NREU and GHG emissions for biodiesel production were calculated considering the whole process: esterification/transesterification reactions (including methanol as raw material and the acid/base homogeneous catalyst), and downstream processes to recover biodiesel, glycerol and methanol in excess. These impacts consider the credits from glycerol obtained as a by-product. The NREU and GHG emissions results for biodiesel production are presented in Fig. 7. In this case the SET&HI process leads to lower NREU and GHG emissions while the SE&T process has an additional major contribution from the drying step.

Microalgae oil can alternatively be hydrotreated to green diesel as described in Section 2.1.7. The environmental impacts of the NExBTL process are directly calculated from the global inputs and outputs given in the IFEU report [52]. Here, the used conditions are based on the Porvo scenario, meaning that the NREU is 5.5 MJ/kg green diesel and the GHG emissions are 0.29 kg CO_2 eq/kg green diesel.

The above discussed comparisons of technologies and processing conditions allow to select the best options, for each process stage and for each biorefinery configuration (as defined in Table 1), from an environmental perspective. For example, the best environmental performance for dewatering and cell disruption was for flocculation with chitosan (followed by centrifugation and filtration, where the achieved biomass concentration is 40% TSS) and sulphuric acid treatment respectively. However, if dry-algae biomass is the target product (Biorefinery 1, BR1 in Table 1) the cell disruption process must be skipped and drying must be included. Otherwise, lipids are obtained by using the wet extraction process (and fractionation if necessary). Biodiesel is obtained by the SET&HI process with NaOH. The other main (energy) products, *i.e.* green diesel and biogas, are obtained by hydrotreating and anaerobic digestion as described in Section 2.1.7.

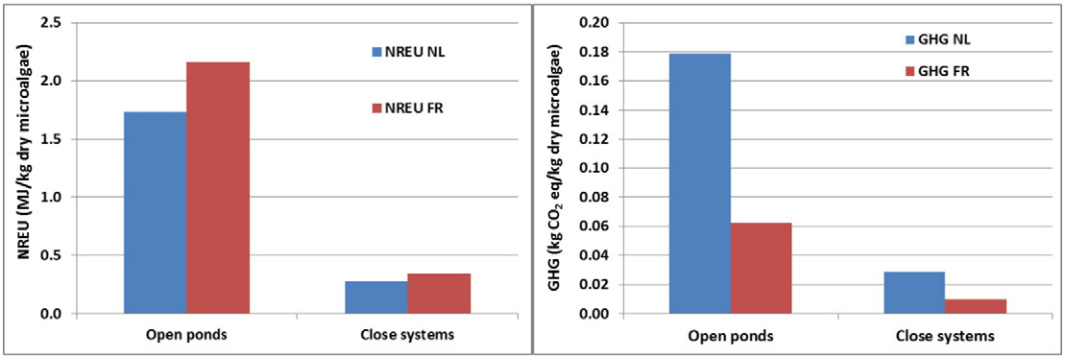

Fig. 13. Reduction of NREU and GHG emissions from N and P nutrients substitution from urban wastewater in OPs @0.4 g/l, and closed systems (HPBRs, VPBRs and FPPBRs) @2.5 g/l.

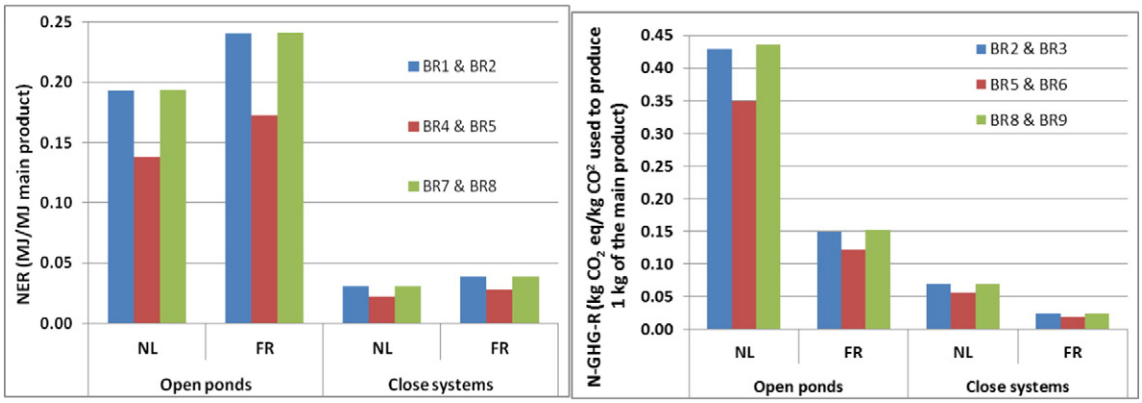

Fig. 14. Potential reduction of NREU and GHG emissions from N and P nutrients substitution from urban wastewater in OPs @0.4 g/l and PBRs (HPBRs, VPBRs and FPPBRs) @2.5 g/l.

3.2. NREU and GHG emissions of integrated microalgae biorefineries

The above performed comparison and selection of technologies and operation conditions based on environmental performances led to favorable NER values for six integrated biorefinery configurations when OPs are considered, they are: BR1, BR2, BR3, BR5, BR6, and BR8 (see Table 1). The NER values improve at high productivity conditions as shown in Fig. 8.a. However for HPBRs and VPBRs only one (*i.e.*, BR1) and three (*i.e.*, BR1, BR2, and BR5) biorefinery configurations reach favorable NER values (see Fig. 8b and c). In the case of FPPBRs, six biorefinery configurations (same as for OPs: BR1, BR2, BR3, BR5, BR6, and BR8) show favorable NER values (see Fig. 8.d). In general, OPs lead to the lowest NER vales at low productivities among the four cultivation technologies while FPPBRs generate the best NER values at high productivities. For all cultivation technologies, BR1 has consistently one of the best NER values reaching ratios as low as 0.5–0.7 MJ NREU/MJ dry microalgae at high productivity conditions. BR2, BR5 and BR8 also lead to positive energy balances for most of the cultivation technologies. In these three cases, microalgae oil-free cake is considered as a fish meal substituent, for which, the energy credits have a large contribution (40.8 MJ/kg microalgae oil extracted). BR2 and BR5 are especially interesting, for OPs and FPPBRs due to their NER values which can be lower than 0.5 (see Fig. 8a and d). The systems BR3, BR6, and BR9 (which consider the use of microalgae oil-free cake as substituent of soybean meal), rely on intermediate values of NER due to the total energy credits obtained from the substitution of both soybean meal and fish oil lower than those received from the 80% fishmeal substitution (as described in Section 2.2.), *i.e.* 30.45 *vs.* 40.8 MJ/kg microalgae oil extracted. BR4, BR7 and BR10 are the configurations with the lowest energy performances for all cultivation systems as shown in Fig. 8. In these three biorefinery cases, the microalgae oil-free cake is used to produce biogas. The low energy performance of these biorefineries is due to the lower

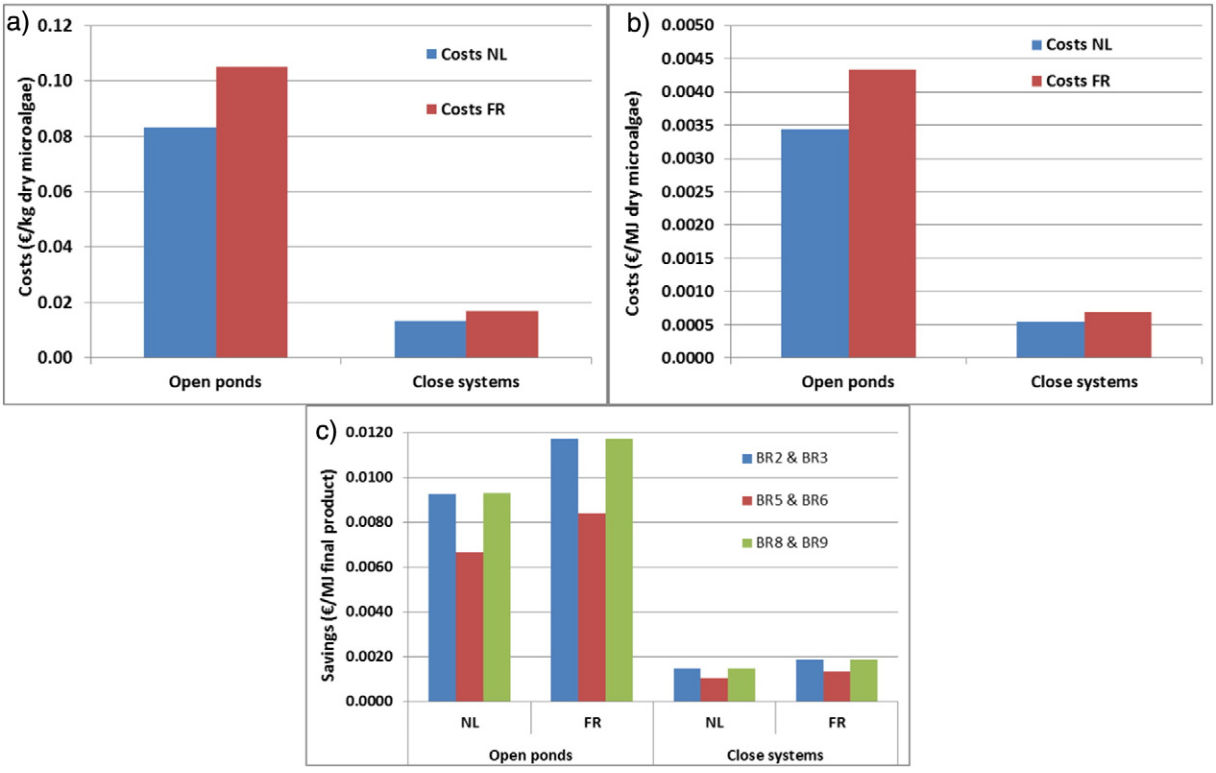

Fig. 15. Potential reduction of production costs from N and P nutrients substitution from urban wastewater: a) in (€/(kg dry biomass), b) in (€/(MJ dry biomass), c) in (€/(MJ main product).

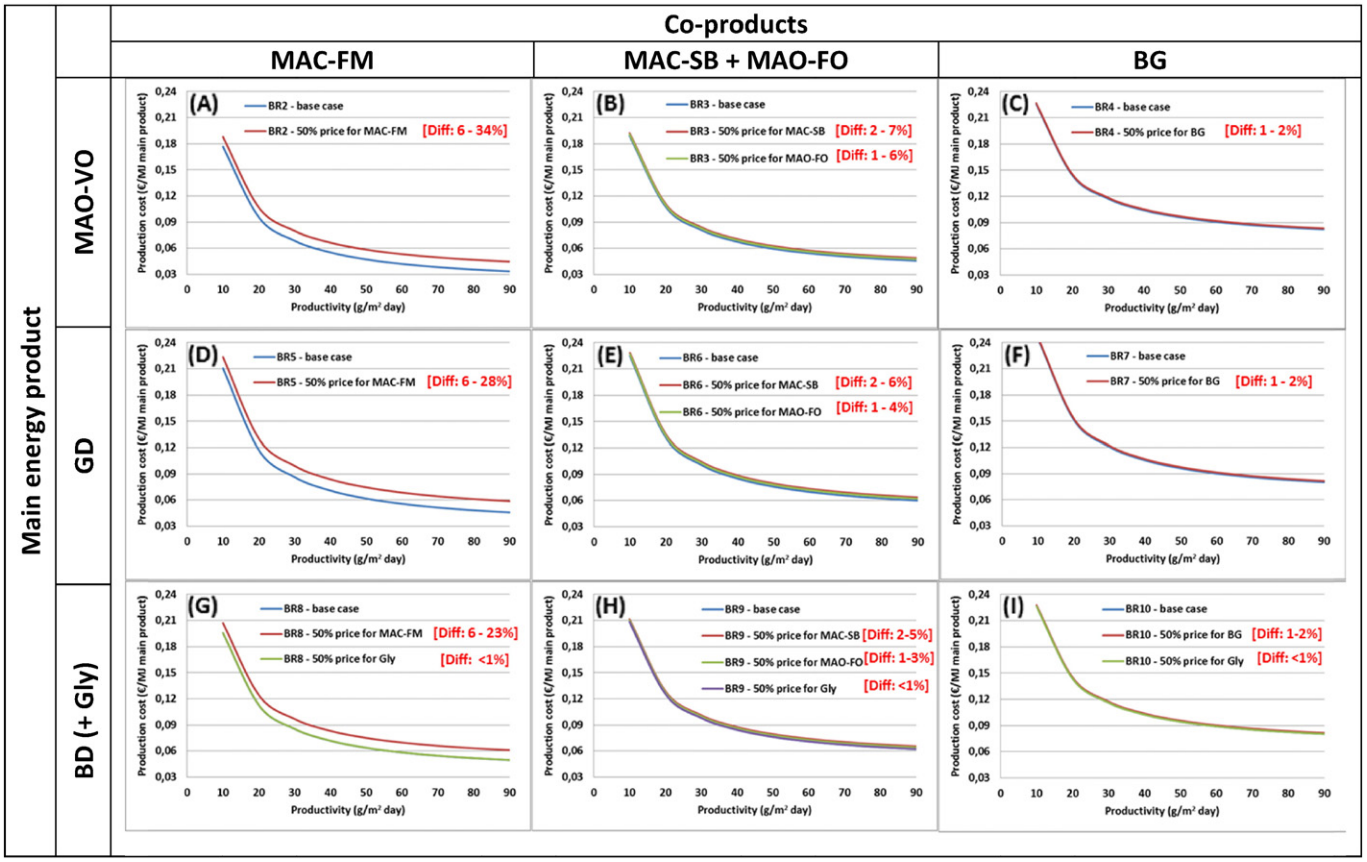

Fig. 16. Sensitivity analysis of by-products prices on the total production costs using FPPBRs. A) BR2; B) BR3; C) BR4; D) BR5; E) BR6; F) BR7; G) BR8; H) BR9; and I) BR10. Diff: relative difference in the total production costs from the lowest productivity considered in the figure to the higher productivity considered in the figure (*i.e.* @10 and @90 g/m^2 day).

energy credits from biogas. They are much lower (*i.e.*, 3.1–3.2 MJ/kg microalgae oil produced) than those from microalgae oil-free cake for replacement of either fishmeal (*i.e.* 40.8 MJ/kg microalgae oil) or soybean meal + fish oil (*i.e.*, 30.45 MJ/kg microalgae oil).

In the case of GHG emissions balances, favorable values (lower than '1') of NGHGR (see Eq. 2) were obtained in most of the biorefinery systems as shown in Fig. 9. Here, the lowest NGHGR values are obtained for OPs (see Fig. 9.a) followed by FPPBRs (see Fig. 9.d). In these two cases, the GHG savings are larger than 50% in at least seven configurations (*i.e.*, BR1, BR2, BR3, BR5, BR6, BR8, and BR9), especially when high productivities are considered. In the case of VPBRs, the GHG savings, achieved by the same seven biorefinery configurations (*i.e.*, BR1, BR2, BR3, BR5, BR6, BR8, and BR9), are around 40% at high productivities

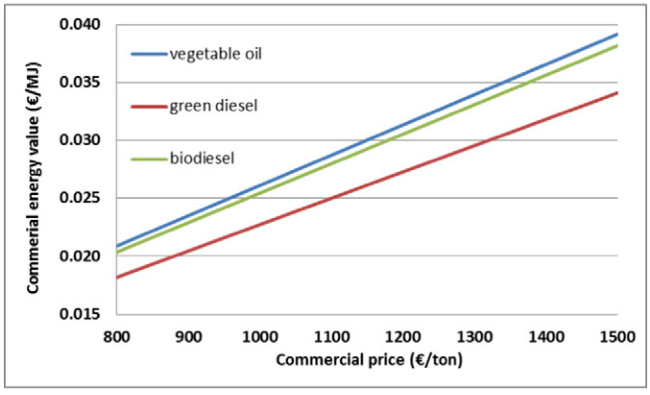

Fig. 17. Commercial energy values for the three main products of the microalgae biorefinery systems.

conditions (see Fig. 9.c). For HPBRs, the GHG savings are around 40% only for three configurations (*i.e.*, BR3, BR6 and BR9; see Fig. 9.b). These three systems consistently reach the best NGHGR values due to the credits from soybean meal + fish oil substitution are larger than those from fishmeal replacement, *i.e.* 3.88 *vs.* 1.77 kg CO_2 eq/kg microalgae oil extracted, respectively. These GHG credits are especially interesting when microalgae is cultivated in OPs or FPPBRs (see Fig. 9a.d) since the global CO_2 balance results in negative NGHGR values. This means that the CO_2 credits obtained from the by-products are larger than the CO_2 emissions generated in the biorefinery configuration.

Table 4 qualifies the NREU (as NER) and GHG emissions (as NGHGR) based on the global balances of energy use and GHG emissions for high productivity conditions. A plus "+" is given when either the energy or GHG ratio is favorable (*i.e.* <1.0) for each cultivation technology. Thus, the best overall environmental performance is for 'BR3' with 'FPPBRs'. This system is selected for the scenario analysis in Section 3.4.

3.3. Economics of integrated microalgae biorefineries

The total production costs, in €/MJ main (energy) product, for each biorefinery concept and for the four cultivation technologies are shown in Fig. 10 as a function of the cultivation productivity. A general comparison among the four cultivation technologies shows that, at low productivity values, the lowest production costs are obtained for HPBRs following, in increasing order, by: OPs, VPBRs, and FPPBRs; however the order of the production costs by technology changes at high productivities, *i.e.* FPPBRs < VPBRs < HPBRs < OPs.

Predominant rankings can be identified for each cultivation technology with only slight differences between low and high productivities conditions. The economic rakings are shown in Table 5. All configurations are categorized in four groups, which are mostly related to the

distribution of final products. In Group IV (composed by BR4, BR7 and BR10), all configurations contain biogas as the final product from microalgae oil-free cake. In Group III (composed by BR6 and BR9), microalgae oil-free cake is considered as substitute of soy bean meal. Finally, Groups I and II consist mainly of biorefinery systems where microalgae oil-free cake is considered as replacement of fish meal. The economic performance is additionally improved when the produced oil is directly used with not further transformation.

The economy results showed to be highly dependent on the final use (and quality) of the microalgae oil-free cake, *i.e.* biogas production < substitution of soybean meal < substitution of fish meal. This result is not surprising since microalgae oil-free cake (free of oils) represents 68 wt.% of the total biomass. Thus, the main energy product has a secondary effect (but still relevant, see Group I in Table 5) on the economic performance, *i.e.* green diesel < biodiesel < oil; lower conversion stages of microalgae oil results in a better economic performance. The best economics were found to be for BR2 which is: microalgae oil-free cake for fish meal substitution and microalgae oil for substitution of vegetable oil. BR1 (microalgae biomass) also showed a positive economic performance, but its actual marketability must further be analyzed. It is however important to mention that if a flue gas stream containing contaminants is used for microalgae cultivation, these contaminants could potentially be transferred into the microalgae oil-free cake which might result in either a lower quality (and commercial value) for this fraction, or in additional purification steps (and processing costs).

The total production costs obtained are, in general terms, in the same order as those reported in literature (see comparative Table S.5.1 in the Supplementary material). However a direct comparison of production costs is not always possible because of the existing methodological differences among the reported studies. Table S.5.1 (Supplementary material) presents a more detailed comparison of the total production costs (of microalgae biomass and bioenergy) and the main methodological differences among the results obtained with respect to those reported in literature.

3.4. Scenario analysis of integrated biorefinery systems

Scenedesmus sp. is considered to analyze possible changes in the environmental performance when using another microalgae specie. The biochemical composition of both species is compared in Table 3 (see also Tables S.2.1–S.2.3 in the Supplementary material). *Scenedesmus* sp. typically have a lower content of lipids and a higher amount of carbohydrates and proteins, leading to a reduced production of oils and an increased production of microalgae oil-free cake (for substitution of soybean meal). This new distribution of mass flows of products results in lower NER and NGHGR values with respect to *Nanochloropsis* sp as shown in Fig. 11. NER values are lower by 20–25%, while NGHGR values change even to negative numbers at high productivities. The main reason for this improved environmental performance is the higher production of microalgae oil-free cake for soybean meal substitution which leads to larger energy and GHG credits, especially for the CO_2 balance. Interestingly, the combined effect of lower lipids content and lower productivity does not implies a worse environmental performance. On the contrary, the mass flow credited microalgae fractions increases improving the overall environmental performance.

A second factor analyzed is the geographical location which mainly accounts for the effects of the specific electricity mix and its related price (see Section 2.4). Fig. 11 compares the NER and NGHGR values for the Netherlands (using *Nanochloropsis* sp. and FPPBRs -base case-) to those obtained for France, China and Brazil, using the same process configuration and operation conditions (as selected from Section 3.4). The NER results for the Netherlands and China are quite similar due to the fact that in both countries the electricity mix is mainly composed of fossil fuels. However, NER values for the Netherlands are around 10% higher due to its higher NREU associated to electricity production. In the case of the NGHGR, these values are lower for the Netherlands due to its lower GHG emissions (as kg CO_2 eq./MJ) associated to electricity production. The NER values for France are the highest of this analysis due to the large NREU requirements for electricity production (especially from nuclear power). By the contrary, the NER values obtained from Brazil are the lowest due to the relatively low NREU requirements for electricity production (especially from hydropower). With respect to the NGHGR values, the GHG emissions for electricity production in both countries is relatively low; but in the case of Brazil, electricity mix contains a significant amount of fossil fuels which leads to a higher GHG emissions. However, in these two cases, the GHG emitted by the microalgae biorefinery processing are larger than the credits obtained from the by-products and therefore negative NGHGR values are obtained at high productivities conditions.

The analysis of wastewater as an alternative source of nutrients for microalgae cultivation is done by considering average concentrations of nitrogen and phosphorous in municipal wastewater for the Netherlands and France as described in Section 2.4. Fig. 12 shows the potential substitution of each nutrient (in percentages) as a function of the culture density. For very low culture densities (around 0.2–0.3 g/l), the substitution potential is even higher than 100% for both micronutrients and no additional supply would be required. On the other hand, additional use of N and P would be needed if high culture densities are aimed for the cultivation stage. At a higher culture density, *e.g.* 0.4 g/l, the potential substitution of N and P are 60 and 65% in the Netherlands, and 75 and 90% in France, respectively. At 2.5 g/l, the substitution potential for N and P decreases to only 9.6 and 10.4% in the Netherlands, and 12 and 15% in France. The potential substitution of micronutrients is higher in France than in the Netherlands due to the higher concentration of heavy metals in urban wastewater. However, it is important to mention that this higher concentration of heavy metals may also have negative effects in the microalgae growth due to a potential increase in cytotoxicity resulting in a lower performance of the microalgae cultivation stage, for example in lower productivities.

The total reduction of the environmental impacts associated to the nutrients substitution from urban wastewater are calculated for *Nanochloropsis* at high culture densities, *i.e.* 0.4 for OPs and 2.5 g/l for PBRs (see Table 2), and for 1 kg of dry microalgae produced as shown in Fig. 13. The potential reduction of environmental impacts (NREU and GHG emissions) is 7 times larger for OPs than for closed systems due to the higher substitution potential of nutrients in OPs, and they are also higher for the FR wastewater due to its higher potential substitution of nutrients. This potential reduction of the environmental impacts can directly be subtracted from the total impacts obtained for each biorefinery system. However, the biorefinery configurations containing anaerobic digestion already consider recycle of macronutrients to the cultivation stage. Therefore, these three systems (*i.e.*, BR4, BR7, and BR10) are left out for this analysis. The potential reduction of NER and NGHGR values for both types of cultivation technologies (OPs and PBRs) and for both locations are shown in Fig. 14. Results for NER and NGHGR have similar trends as those for NREU and GHG emissions, *i.e.* the potential environmental impacts reduction are higher in OPs than in closed PBRs, and are also higher in FR than in NL. For OPs, the NREU savings from wastewater, as nutrients substituent, are between 14 and 25% while the GHG emissions savings can be as high as 44%. In the case of closed PBRs, the energy savings and the GHG emissions reduction is only around 2–7%. This analysis shows the high dependency existing between the culture density, the nutrients requirement and the potential substitution of nutrients from wastewater.

Nutrients substitution from urban wastewater can also improve the economic performance of a microalgae biorefinery. The potential costs reductions are calculated based on the N and P substitution capacity and on their prices, as shown in Fig. 15. The total cost savings for OP vary from 6 to 16%, while for close PBRs these savings are in between 1 and 5%.

Another factor analyzed is the effect of the sale price of by-products on the total production costs. This analysis was done for all biorefinery systems, excluding BR1 which produces only microalgae biomass, considering FPPBRs in the microalgae biomass cultivation stage. Furthermore, the sale price of each by-product (one at the time) was decreased by 50%. Results are shown in Fig. 16 and they indicate that the total production costs are more sensitivity to the type of by-product than to the main energy product. For example, a 50% reduction of the MAC-FM value resulted in an increment of the total production costs of 34, 28, and 23% (at high productivities conditions) when MAO-VO, GD, and BD (+ Gly) are produced as main energy products (see Fig. 15A, D, and G). On the other hand, a 50% reduction of the value of MAC-SB or MAO-FO represent an increase in the total production costs of 1–7% independently of the main energy product (see Fig. 15B, E, and H). In the cases of Gly (when BD is the main energy product) and BG, their reduction in the sale price did not represent any significant change in the total production costs. For Gly this increment was always lower than 1% even for high productivity conditions (see Fig. 15G, H, and I), while for BG the increment was around 1–2% for low to high productivities respectively (see Fig. 15C, F, and I). From this analysis, it can also be noticed that the microalgae biomass productivity in the cultivation stage has a much higher effect of the total production costs than the sale price of the by-products even for those of a high value like MAC-FM. For example, an increment of the microalgae productivity from 20 to 40 g/m^2 day would result in a 40% reduction of the total production costs (see Fig. 15A), while doubling the sale price of MAC-FM (at 20 g/m^2 day) results only in a 20% reduction of the total production costs.

The production costs can be reduced if a carbon credit is considered. In this case, carbon credits of 10–50 €/t of CO_2 are considered as described in Section 2.4. Thus, the production costs could further be lowered by 0.018–0.0054 €/MJ of main product, representing additional saving of 9–12% for OPs and 9–18% for close systems. An important aspect to consider from the use of municipal wastewater for nutrients substitution is that these type of streams might contain contaminants (which vary by types and concentration levels from place to place) that can be transferred first to the microalgae biomass and then to the final products. Thus, the presence of potential contaminants in the wastewater might lead to additional requirements for purification of the microalgae biomass fractions (resulting in more capital costs, primary energy use and GHG emissions) or in products of lower quality and commercial value. Therefore, strict characterization of the microalgae based products would be required before commercialization specially for food and fed applications.

Considering that the prices for vegetable oil, biodiesel and green diesel can vary from 900 to 1500 €/t ([26,65]), the energy values (*i.e.*, commercial price/energy content, €/MJ) of these products was found to be higher than the production costs here obtained as shown in Fig. 17. Thus, the highest potential for economic feasibility of the biorefinery configurations here analyzed is for BR2 (oil as main product). Therefore, the production costs would be competitive for vegetable oil prices above 1150 €/ton (or above 1000 €/ton if wastewater is used and/or if carbon credits are considered).

As a final remark, it is important to mention that the successful implementation of such as biorefinery systems would require to overcome other socio-technical potential barriers like market acceptance of microalgae-based products (*e.g.* bioplastics, oleochemistry, biolubricants, adhesives, food and feed, human nutrition and health, and pharmaceuticals) and their related regulations. For example, the market opportunities and public acceptance are significantly higher for bioplastics and chemical applications than for food and feed applications. Furthermore, markets like human nutrition, health and pharmaceuticals require strict certification standards which in the case of most microalgae-based products would have to be developed taking significant time and economic resources before actual commercialization [5].

4. Conclusions

The combination of methods for process design (conceptual design, mass/energy balances, and parametric analysis) together with the use of tools for systems analysis (energy efficiency, LCA, and economics) has proven to be a useful approach for quick definition and screening of multiple concepts of integrated microalgae-based biorefineries (using pure CO_2) from a sustainability perspective. The biorefinery systems with the lowest net ratios of NREU and GHG emissions are those where microalgae oil-free cake is used as nutrient substituent for animal feed and where lipids are used as substituents of vegetable oils (*i.e.* BR2 and BR3). In these cases no potential contaminants from the flue gas (here pure CO_2) were assumed to be transferred to the final microalgae-based products. Further conversion of oil to either biodiesel or green diesel do not offer any energy or GHG emissions benefits (*i.e.* BR5, BR6, BR8, and BR9). Similarly, anaerobic digestion of microalgae oil-free cake results in reduced environmental performances (*i.e.* BR4, BR7, and BR10). Similarly, the economic performance largely depends on the final use of the microalgae oil-free cake; for example, the best results are obtained when the microalgae oil-free cake is used as fishmeal substituent (independently of the final product from the lipids fraction, *e.g.* oil, biodiesel and green diesel), and the worst economic performance when the microalgae oil-free cake is considered to be digested to biogas.

Overall, microalgae based systems seem to have a promising balance between high-demand energy-oriented and low-demand material-oriented products for future multiproducts biorefineries. In this case, energy products are recommended to be limited only to the lipids fraction, while the microalgae oil-free cake could be used for the replacement of animal feed.

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Appendix A. Supplementary Material

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.algal.2016.04.022.

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Glossary

BD: Biodiesel
BG: Biogas
CEPCI: Chemical Engineering Plant Cost Index
CPI: Consumer Price Index
DC: Depreciable capital
DCC: Direct capital costs
DSP: Downstream process
FC: Fixed cost
FPPBRs: Flat panel photobioreactors
GD: Green diesel
GHG: Greenhouse gases
Gly: Glycerol
HP: High productivity conditions
HPBRs: Horizontal tubular photobioreactors
ICC: Indirect capital costs
LP: Low productivity conditions
LHV: Lower heating value

MABM: Dry microalgae biomass
MAC-FM: Microalgae oil-free cake for substitution of fishmeal
MAC-SM: Microalgae oil-free cake for substitution of soybean meal
MAO-FO: Microalgae oil for substitution of fish oil
MAO-VO: Microalgae oil for substitution of vegetable oil
NER: Net Energy Ratio (=NREU/Energy content of main product or energy product)
NGHGR: Net GHG Ratio (=GHG emitted in CO_2 equivalents/CO_2 fixed per kg of main product or energy product)
NREU: Non-renewable energy use
OC: Operating cost
OSE: Organic solvent extraction
OPs: Open ponds
PBRs.: Photo bioreactors
SCE: Super critical extraction
SE&: Simultaneous extraction and transesterification process
SET&HI: Sequential esterification/transesterification system with heat integration
TOC: Total operating cost
TPC: Total production costs
TSS: Total suspended solids
VPBRs: Vertical stacked tubular photobioreactors