A multi-objective optimization-extended techno-economic assessment : exploring the optimal microalgal-based value chain

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The use of fossil-based products induces a large environmental burden. To lighten this burden, green technologies are required that can replace their fossil-based counterparts. To enable the development of economically viable green technologies, an optimization towards both economic and environmental objectives is required. To perform this multi-objective optimization (MOO), the environmental techno-economic assessment (ETEA) methodology is extended towards a MOO-extended ETEA. The development of this MOO-extended ETEA is the main objective of this manuscript. As an example of a green technology, the concept of microalgae biorefineries is used as a case study to illustrate the MOO-extended ETEA. According to the results, all optimal value chains include open pond cultivation, a membrane for medium recycling and spray drying. The optimal economic value chain uses *Nannochloropsis* sp. in a one-stage cultivation to produce fish larvae feed, while the optimal environmental design uses *Dunaliella salina* or *Haematococcus pluvialis* to produce carotenoids and fertilizer or energy products, by means of anaerobic digestion or gasification. The crucial parameters for both environmental and economic feasibility are the content, price and reference impact of the main end product, the growth parameters and the biomass and carotenoid recovery efficiency alongside the different process steps. By identifying the economic and environmentally optimal algal-based value chain and the crucial drivers, the MOO-extended ETEA provides insights on how algae-based value chains can be developed in the most economic and environmentally-friendly way. For example, the inclusion of a medium recycling step to lower the water and salt consumption is required in all Pareto-optimal scenarios. Another major insight is the requirement of high-value products such as carotenoids or specialty food to obtain and economically and environmentally feasible algal-based value chain. Due to the modular nature of the MOO-extended ETEA, multiple processes can be included or excluded from the superstructure. Although this case study is limited to current microalgae biorefinery technologies, the MOO-extended ETEA can also be used to assess the economic and enviromental effect of more innovative technologies. This way, the MOO-extended ETEA provides a methodology to assess the economic and environmental potential of innovative green technologies and shorten their time-to-market.

Introduction

Technological research is often directed by an economic motivation. When improving the performance of an existing technology or introducing a new technology to the market, the main objective is indeed in general to obtain maximal profits. However, this focus on economic optimization has led to technologies and processes that have a large impact on the environment. The consequences of climate change and other environmental problems have urged researchers to include a new objective during technology development. Besides maximizing profits, minimalizing the environmental impact needs to be a main objective as well. As technology development has become a process with multiple objectives, a multiple criteria decision method is required. The aim of this paper is therefore to develop a multi-objective optimization methodology that takes into account both economic and environmental objectives and that can be easily applied to a large range of green technologies.

A large variety in potential algae species exists, each with their own characteristics and end products.[1] These end products can vary from high-value products, such as food supplements, and pharmaceuticals to low-value products, such as energy.[2] The process value chain of an algae biorefinery contains multiple different process steps, starting from cultivation towards purification of the end products. For each step, different options exist.[3] Due to this variety in species, end products and process steps, multiple algae biorefinery value chains can be formulated.[4] Currently, two main microalgae applications exist. The first application is the sale of high-value carotenoids as food supplements or colorants.[5] The carotenoids, such as β-carotene or astaxanthin, have a limited market volume but a high market

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price.[6] Another commercially viable application is the sale of the whole microalgal-biomass for food or feed.[5] The biomass, which can contain carotenoids and multiple valuable nutrients, has a larger market volume, but a lower market price. Despite these two commercially viable applications, most economic and environmental assessments of microalgal-based value chains have focused on the production of bioenergy as the main product. An example of these assessments is the analysis of the life cycle energy and greenhouse gas analysis for algae-derived biodiesel.[7] However, the production of microalgal-based bioenergy is currently not economically viable and also no consensus exist over the environmental impact of this application.[8] By comparing all these value chains according to economic and environmental objectives using an uniform and transparent method, the potential of the microalgae biorefinery concept can be assessed and the different applications can be compared.

The multi-objective environmental techno-economic optimization that will be developed in this paper, will therefore be applied to explore the economic and environmental potential of microalgae biorefineries. In previous studies, multiple algal-based biorefinery value chains have been assessed on their technological, economic and/or environmental impact.[9] For these individual assessments, the environmental techno-economic assessment (ETEA) methodology, which integrates aspects of process design, techno-economic assessment and life cycle assessment, was developed.[10] However, the ETEA methodology assesses different scenarios one by one. In each scenario a potential microalgae value chain is selected. However, in practice, for each step of the value chain, multiple process options are available. Not only the process options can differ, but also the selected algae species and the production scale can vary, as economies-of-scale can have an influence as well. The number of potential scenarios can therefore become very large. In this paper, 1188 potential value chains were included, which each can have multiple optimal production scales as the economies-of-scale can have different effects on different objectives. Due to this large number of potential scenarios, the ETEA methodology needs to be extended with an optimization step. As this optimization follows both economic and environmental objectives, a multi-objective optimization (MOO) method is used. The ETEA methodology will therefore be adapted towards a MOO-extended ETEA methodology.

The MOO methodology as used for the MOO-extended ETEA was based on the work of Gong and You (2014). In their study, an algal-biorefinery value chain was optimized, producing energy products out of a *Chlorella vulgaris* feedstock, with the objective to minimize unit carbon sequestration and utilization cost.[11] The study adopted multiple strategies, such as the successive piecewise linear approximation algorithm, to reduce the complexity of the non-convex optimization problem. These strategies will also be adopted in the MOO-extended ETEA and will be further discussed in the methodology section. However, Gong and You (2014) considered only one objective. The same authors used their methodological framework as well to optimize towards multiple objectives, those being unit cost and unit global warming potential.[12] To deal with the fractional term in the objective, a parametric algorithm, based on Newton's method was introduced. This way, the unit cost and unit global warming potential could be optimized. However, in their superstructure, always one main end product, being renewable diesel or biodiesel was produced. This way, only the unit global warming potential for this end product will be minimized. The authors used the ε-constraint method to handle the multiple objectives. As this method is the most used method for similar problems, it will also be used in the current model.[13] A third paper by the same authors optimized towards total annual costs and global warming potential and included routes for medium-value byproducts, such as poly-3-hydroxybutyrate. Multiple end products were included in this paper, but only the global warming potential for one product was optimized instead of the entire biorefinery. Economic allocation was used to obtain the global warming potential of the main end product.[14] In a similar study, multiple end products were included as well without the use of the fractional term.[15] Instead of the unit global warming potential, the total global warming potential was minimized. However, the optimal value chain in this case for the environment would be to produce nothing at all. These previous papers provide a good base for the MOO, providing strategies to deal with non-linearities. However, these studies mostly focus on one end product at a time and do not take into account the fossil-based reference products for all end products. Moreover, they also do not take into account the entire life cycle of the end products. Lastly, they only take into account GWP as an environmental indicator. Another MOO study on renewable energy-driven desalination systems does take into account multiple environmental indicators, using the ReCiPe method.[16] However, they aggregate the different indicators into one end indicator, which also does not allow to see the impact on different indicators. An aggregated indicator has also been used by other studies, for example for the MOO of single-effect water/Lithium Bromide absorption cooling cycles; for a medical supply chain design problem and for liquid-liquid extraction operations.[17, 18 19] Studies which do take into account multiple environmental indicators usually use genetic algorithms.[20] However, these algorithms cannot guarantee to find the global optimal solution and are therefore not used in the proposed MOO-extended ETEA.

The current study extends these papers in four different ways. First, a full set of environmental indicators, using the ReCiPe endpoint indicators, is included. Second, a methodological framework for the integrated use of ETEA and MOO is provided. Third, a large range of algae biorefinery value chains containing different sorts of microalgae species and products, ranging from high value antioxidants to low-value energy applications, is assessed. Finally, a cradle-to-grave LCA perspective is used by including the conventional reference processes for all end products. The inclusion of these conventional reference processes enables the use of substitution as an allocation strategy, which is preferred by the ISO guidelines over the generally applied partitioning strategies.[21] To the best of our knowledge, this study is the first one that provides a methodological framework to use a multi-objective optimization in an integrated environmental and techno-economic assessment, including a cradle-to-grave perspective and a full range of environmental indicators. Moreover, this study is also the first one that assesses the economic and environmental potential of algal-based biorefineries, including both high-value applications and low-value applications and including an optimization of the production scale. As most existing algae-based biorefinery studies focus on energy applications, this study aims to broaden this perspective focusing more on the sustainable design of green chemicals. The main objectives of this paper are therefore: 1) integrate a MOO method in the ETEA methodology and provide a generally applicable strategy to perform a MOO-extended ETEA; 2) applying this MOO-extended ETEA to a case study of multiple algal-based value chains to enable the identification of the optimal process routes and to guide further technology development of algal-based biorefineries.

Data and Methodology

Superstructure

The potential microalgae biorefinery value chains are grouped in a superstructure, containing the different options for each process step of the value chain. The included options were selected based on their Technological Readiness Level (TRL).[22] Technologies that are already technologically mature (TRL 9) can be modelled with less uncertainty than technologies on a low TRL. Multiple technologies for bioenergy production were included as well. Although the application for algal-based bioenergy is not yet commercially available, the technologies required for this production are on a high TRL. The model did not include a constraint for multiple products, as this would exclude optimal value chains. Therefore, not all optimal value chains have to be biorefineries. The superstructure is illustrated in Fig. 1. The hypothetical microalgae biorefinery is situated in Belgium and a total project lifetime of ten years is considered. The production scale is limited to 3% of the global market of each end product. The market volumes of the different potential end products can be found in the electronic supplementary information.

The superstructure includes three potential microalgae species: *Dunaliella salina*, *Haematococcus pluvialis* and *Nannochloropsis* sp. The first two species are currently used for anti-oxidant production on a commercial scale.[23] This commercial process can be extended to produce multiple products, such as fertilizer or bioenergy, in a biorefinery concept. *Nannochloropsis* sp. is included as it has been proposed as a potential species for a microalgae biorefinery by multiple studies.[24-26]

In the first cultivation stage the biomass was accumulated in optimal growth conditions. The three process options for this cultivation stage are an open pond, a tubular photobioreactor (PBR) and the Proviron Advanced Photobioreactor Technology (ProviApt), which has been assessed before for microalgae cultivation.[27] The ProviApt reactor consists of multiple reactor chambers, contained by a plastic bag.[28] The water in the plastic bag acts as a buffer against outside contamination and temperature variations.[29]

The algae could be cultivated in one stage or in two stages. If a two-stage cultivation is selected, the second stage is a stress stage. In this stress stage, the nutrients are limited and the salinity is increased. Under these conditions the algae accumulate specific components such as β-carotene, astaxanthin and triacylglycerols (TAG).[30, 31] For *Dunaliella salina*, a maximum β-carotene content after the stress stage of 8.8% was included.[32-34] The astaxanthin content of *Haematococcus pluvialis* reached 3% after the stress stage.[35] The growth parameters of the different cultivation options for the different algae species are summarized in Table 1. To model the cultivation stage, a logistic growth curve was used. The maximum specific growth rate *r* was corrected for a lower irradiation rate in Belgian conditions for open pond cultivation. The algae were transported to the stress stage as the concentration c_1 reached 67% of their maximum concentration. The algae were harvested from the stress stage when the concentration c_2 reached 77% of their maximum concentration.

The same amount of nutrients was provided in each cultivation option. These nutrients included KNO_3, KH_2PO_4, $NaHCO_3$, $MgSO_4$ and $FeCl_3.6H_2O$. The CO_2 consumption was calculated based on the carbon composition of the microalgae, taking into account an uptake efficiency of 59% for open ponds and 71% for the PBR and the ProviApt.[36-40] Besides CO_2, also N_2O, NH_3 and O_2 emissions were included. The bioreactors were both heated to obtain the optimal cultivation temperature. The electricity consumption for cultivation was caused by mixing and the CO_2, water, salt and nutrient supply.

After the cultivation step, the medium, containing water and salt, can be recycled using a membrane. The Integrated Permeate Channel (IPC®) membrane is used for this step.[41] This preharvesting step can be repeated if a two-stage cultivation process is used.

After cultivation and preharvesting, the biomass is harvested in a centrifuge until a biomass concentration of 12% is reached.[42] The electricity consumption for this centrifuge equaled 1.40 $kWh \cdot m^{-3}$.[43] In case of a marine algae species, a washing step was included to reduce the salt content under 4 $g \cdot l^{-1}$. The washing step included a mixer and a centrifuge.

In the next step, two process options are included for the drying step: a spray dryer and a freeze dryer. The atomization energy for the spray dryer came from electricity. In the spray dryer the biomass was dried until an end solid concentration of 5%.[44] For the freeze dryer, the end solid concentration was 6%.[45] An energy consumption of 1,445 $kWh \cdot ton^{-1}$ was calculated for the spray dryer. For the freeze dryer, an energy consumption of 2,000 kWh·ton^{-1} was used, based on the technical properties of a commercial freeze dryer.

If the microalgae have a thick cell wall, a disruption step needs to be included after the drying step. Bead milling is included as the process option as it is one of the technologies generally preferred by the industry.[46] For the beadmilling, an energy consumption of 2.82 kWh·kg dry weight^{-1} was modelled.[47]

After cell disruption, the desired fractions can be extracted. The different desired products, being carotenoids and TAGs, reside in the lipid fraction of the algae biomass. This lipid fraction is extracted using hexane as a solvent.[48] A recovery rate of 95% was assumed. Six extraction steps were included, requiring each time 1 l hexane·l biomass fraction^{-1}.[49] The hexane emissions are 5.20 g·kg hexane^{-1}.[50, 51] The electricity consumption of this extraction step was 1.70 kWh·kg lipid fraction^{-1}.[50-52]

After the extraction step, the two fractions are separated using a filtration step. The hexane in the lipid fraction is recycled in a vacuum distillation step, while the hexane in the residue is recycled in an evaporator.

The lipid fraction can be sold as such or can be further processed into fuel and energy, using a transesterification or a hydrotreating step. In the transesterification step, the triglycerides react with an alcohol to form esters and glycerol in the presence of an acid or base catalyst. The main end product resulting from this process is biodiesel.[53] To model this process, the data parameters from the GREET soybean oil transesterification process were used.[54] GREET is a software tool that calculates the emissions resulting from multiple fuel and vehicle life cycles. It contains data on a variety of conversion processes and feedstocks.[55] Although the process for soybean oil is not identical to the microalgae oil process, this process has been used before as a good proxy for the microalgae process.[56] For the modelling of the equipment, the process design of a previous study was used.[57] Hydrotreating consists of multiple processes where hydrogen reacts with the lipid fraction to produce renewable diesel and naptha.[58] Before the hydrotreating process, a three-step purification process consisting of degumming, demetallization and bleaching was included to remove gums, metals and other impurities that could cause problems for the subsequent catalytic upgrading step.[58] For the equipment modelling, the process design of a previous study was used.[59]

The residue can be sold as fertilizer or can be further processed into energy products. If the carotenoids have not been extracted, the biomass can be sold as fish larvae feed. The included process options for the processing of the residue into energy are pyrolysis, gasification, torrefaction, hydrothermal liquefaction (HTL), and anaerobic digestion. Pyrolysis, gasification, torrefaction and HTL are thermochemical processes that produce a mixture of biochar, biogas and bio-oil.[60] However, the composition differs over these different processes. In the pyrolysis process, the carbon fraction of the algae biomass is decomposed at high heating rates in the absence of oxygen. The main end product of pyrolysis is bio-oil. In gasification, mainly biogas is produced by reacting the biomass fraction under high temperatures with a controlled amount of oxygen and/or steam. Biochar is the main end product in the torrefaction process. This process occurs at a relatively lower temperature (<300°C). The biomass is partly decomposed and can be further pelletized to achieve high densification.[61] The HTL process produces an aqueous phase as well, which can be recycled for the nutrients. The HTL process is comparable to pyrolysis, however, the biomass does not need to be dried.[62] The drying costs can therefore be reduced when the HTL process is selected. The bio-oil resulting from the four thermochemical conversion processes is upgraded and refined into gasoline and diesel before it can be sold. In the anaerobic digestion process, biogas is produced. The digestate, containing nutrients, is recycled to the cultivation stage.[63]

All technological, economic and environmental input data for the different processes can be found in the electronic supplementary information.

Methodology

Environmental Techno-Economic Assessment. The ETEA methodology consists of five steps: The first step is the market study, where the prices, market volumes and process options are identified. In the second step, the process flow diagram (PFD) and mass and energy balance are constructed. The process value chain is formulated and the input, output and equipment size of the entire process is calculated. The third step is the economic analysis, where the Net Present Value (NPV) is determined. All prices have been harmonized to €2016 values using the CEPCI index. In the fourth step, the environmental analysis, the environmental impact indicators are calculated. For this purpose, the midpoint and endpoint indicators of the Recipe 2016 indicator set are used.[64] The last step is the interpretation step, where the contribution of the different process steps and parameters to the output indicators is assessed. A Monte Carlo analysis is used with 10,000 iterations and a triangular distribution on all parameters (-10%; +10%) to identify the parameters that have the highest impact on the output indicators. The triangular distribution was assumed as this is the most commonly used distribution from modeling expert opinion.[65] The fraction of 10% has to be the same for all variables. It does not represent the uncertainty of that parameter.[66]

The ETEA methodology follows an iterative approach where new technologies are guided throughout their development, following a stage-gate approach.[67] The TRLs are used to classify the different stages.[22] To identify the most important parameters, the whole lifecycle needs to be included.[10] The MOO extension will optimize the ETEA results for all process designs, and is therefore an extension of the second, third and fourth step of the ETEA.

The ETEA model is built in Excel. The upstream environmental impact factors of the material and utilities used in the process are extracted from SimaPro and added in a separate sheet in the Excel-model. In this way, the entire model remains in Excel and dynamic linkages between the different parts exist. This dynamic linkage means that a change in one input parameter is automatically translated into all the different output parameters.

MOO-extended ETEA. The result of the optimization algorithm is the Pareto frontier, which consists of all Pareto-optimal value chains. A Pareto-optimal value chain is a value chain that cannot be improved in one objective, without deteriorating another objective.[68]

The MOO–extended ETEA has four objectives, including one economic and three environmental objectives. The environmental objectives are calculated as environmental savings compared to a reference scenario using the substitution method. In the reference scenario the same products are produced, based on a conventional feedstock, such as a fossil feedstock, instead of a microalgae feedstock. If the environmental impact of the microalgal-based product is lower than the environmental impact of the reference product, the environmental savings, indicated by Δ, are positive. The four objectives of the MOO problem are therefore formulated as follows: 1) Maximization of the NPV; 2) Maximization of the Human health environmental savings (ΔHH); 3) Maximization of the Ecosystem quality environmental saving (ΔEQ); and 4) Maximization of the Resource scarcity environmental saving (ΔRS).

The decision variables that are optimized are: 1) the binary variables $b_{g,h}$, which select which process option h is included in step g; and 2) a continuous variable a for the production scale. Based on these decision variables, the four objectives can be calculated for each possible value chain. An overview of all notations used in the MOO problem is provided in Table 2.

To calculate the objective functions based on these decision variables, non-linear functions are required for two reasons. First, the mass and energy balance contains non-linear equations. For example, the total electricity consumption of the open pond process is calculated using a binary variable selecting the open pond process and a continuous variable covering the scale of the process. The multiplication of these two variables, which both need to be optimized, leads to a non-linear equation. The second non-linearity is situated in the cost and environmental impact calculation of the equipment. For example, the cost of equipment unit k in process option h of process step g is calculated following the six-tenth rule.

The optimization problem can therefore be classified as a Multi-Objective Mixed Integer Non-Linear Problem (MOMINLP). However, this type of problem is usually non-convex and it is very challenging to obtain a global maximum solution within a reasonable amount of time. Therefore, we will relax this problem using three strategies in the same model. The first strategy is to remove the non-linearity in the mass and energy balance. In the second strategy, the non-linearity in the cost and environmental impact calculation of the equipment is removed. With the third strategy, the multi-objective problem is transformed into multiple single objective problems.

In the first strategy, new continuous decision variables are introduced for each process option of each process step, following the big-M method.[69] New constraints are added to set the variable at zero, if the corresponding process option has not been selected: $0 \leq a_{g,h} \leq M * b_{g,h}$. If the binary variable is one, the M indicates the upper bound of the variable. In this way, the binary variables are removed from the mass and energy balance equations and added in these additional constraints. The variables a are also divided into input and output variables for each process option. New variables a are created for each component j of the mass throughput as well. The meaning of the different continuous decision variables $a_{g,h,j}$ is illustrated in Fig. 2.

In the second strategy, a piecewise linear approximation is added, which estimates the linear function that results in the same cost for the corresponding scale.[70] Fig. 3 illustrates the concept, by dividing the cost curve into two partition parts, using two partition points (PP). As the scale is situated in the second part of the curve, $Cost(c_{g,h})$ will be approximated by $Cost(c_{g,h,p=2})$. The cost will therefore always be underestimated.[11]

If the non-linear cost function is written as $Cost = \delta_{g,h,k} * c_{g,h,k}{}^{\beta_{g,h,k}}$, with $\delta_{g,h,k}$ being a constant, dependent on the cost of the reference capacity, and $\beta_{g,h,k}$ being the power exponent referring to the economics of scale for the equipment k of option h of step g, then the corresponding linear approximated function will be: $Cost = \sum_{p \in P} e_{g,h,k,p} c_{g,h,k,p} + f_{g,h,k,p} d_{g,h,k,p}$. The binary variable $d_{g,h,k,p}$ will ensure that only one part of the curve is selected. The continuous variable $c_{g,h,k,p}$ equals the appropriate capacity and ensures that the part of the curve is selected that contains this capacity. The continuous variables $e_{g,h,k,p}$ and $f_{g,h,k,p}$ calculate the cost for the selected partition point $PP_{g,h,k,p}$.

In the third strategy, the ε-constraint method is used to transform the multi-objective problem into four single-objective problems.[71] In each single-objective problem, one objective is kept as the main objective and the other objectives are added as an additional constraint. In this additional constraint the transformed objective needs to be larger than the ε-value for that objective. By varying the ε–value between the nadir and utopian value of that objective, a discontinuous Pareto frontier is obtained. This concept is illustrated in Fig. 4 for five ε-value iterations and two objectives.

Using these strategies, the resulting single optimization problem has been transformed into a Mixed-Integer Linear

Problem (MILP). Multiple iterations of this MILP were run, each varying the objective function and the different values of the ε-constraint.

The MOO step is directly linked to the other steps of the ETEA. During the previous steps, technological, economic and environmental data for each process option of each process step was stored in the Excel model. This data is then transferred into two matrices (*Aq* and *Aeq*) and seven vectors (*Bq*, *Beq*, x_0, *npv*, *hh*, *eq*, *rs*) in order to solve the problem.

The matrix *Aeq* contains the equality constraints, where each column stands for one decision variable. Here, all correlations between the different variables of *x* are stated. The potential combinations of process options and parts of the cost curve are also specified in this matrix. The matrix *Aq* contains the inequality constraints and has the same columns as *Aeq*. The last lines of matrix *Aq* contain the objective vectors *npv*, *hh*, *eq* and *rs*. The corresponding parameters of vector *Bq* contain the ε-value for the corresponding iteration. The values of x_0 equal the decision variables of the selected ETEA value chain. Following this approach, the matrices can be checked in the ETEA model in Excel.

The optimization problem was solved using the global SCIP solver in Matlab, by using the OPTI-tool, which provides an interface for a broad range of solvers in Matlab.[72, 73] In the electronic supplementary information, the formulations of the original MOMINLP, the transformed MILP and the composition structure of the matrices are provided.

Results

Optimization: Pareto frontier

The Pareto frontier consists of the four value chains that are optimal in each dimension and of seven intermediate value chains that cannot be improved in one dimension without deteriorating in another dimension. The names of the optimal value chains refer to the main differences that distinguish them from each other. The first main difference is the algae species, where Ds signifies *Dunaliella salina*, Hp stands for *Haematococcus pluvialis* and Ns refers to *Nannochloropsis* species. The second main difference is the processing option for the residual biomass: FLF means the residual biomass is sold without processing as fish larvae feed; F means the residual biomass is sold without processing as fertilizer; G means that the residual biomass undergoes a gasification step; P refers to a pyrolysis step for the residual biomass processing; AD signifies the residual biomass processing step is an anaerobic digestion step; and T means that the residual biomass goes to a torrefaction process. In all eleven optimal value chains, the algae are cultivated in an open pond and the medium, containing water and salt in case of a marine algae species, is recycled. The algae are harvested in a centrifuge, dried using a spray dryer and no lipid processing step is included. In the value chain with the highest NPV, value chain Ns FLF, *Nannochloropsis* sp. is cultivated in one stage and fish larvae feed is sold. No disruption, extraction, separation, residue processing, residue purification or lipid purification step was included in this value chain. The fish larvae feed is used for larvae in aquaculture. The optimal value chain for human health savings is value chain Ds AD. In this value chain, *Dunaliella salina* is cultivated in two stages. The biomass residue goes through an anaerobic digestion step. The lipid fraction, containing the β-carotene, is purified but not further processed. β-carotene is sold as an end product. In the optimal value chain for ecosystem quality saving, value chain Ds F, *Dunaliella salina* is cultivated as well. The only difference from value chain Ds AD is that the residual biomass is not further processed but sold as fertilizer. The optimal value chain for resource scarcity savings is value chain Hp G. In this value chain *Haematococcus pluvialis* is cultivated. No washing step is required, but a bead mill is included for cell disruption. The lipid fraction is purified and sold for the astaxanthin. The residual biomass is processed in a gasification step. The intermediate value chains are value chains Hp F, Hp AD, Ds G, Hp T, Ds T, Hp P and Ds P. Value chains Hp F and Hp AD are similar to value chain Ds F and Ds AD with some differences. Specifically, *Haematococcus pluvialis* is cultivated instead of *Dunaliella salina*, no washing step is required, a bead mill is included and astaxanthin is sold. Value chains Ds G, Ds T and Ds P are similar to value chain Ds AD. The only difference is that the processing of the residual biomass is either a gasification, a torrefaction or a pyrolysis step instead of anaerobic digestion. In the same way value chains Hp T and Hp P resemble value chain Hp AD. A summary of the results of the eleven Pareto-optimal value chains, which constitute the Pareto-frontier, is provided in Table 3.

Process flow diagram (PFD) and mass and energy balance

The PFDs of the optimal value chains are provided in the electronic supplementary information. The summary of the mass and energy balances of the optimal value chains is provided in Table 4.

Haematococcus pluvialis is a freshwater alga and does not require salt addition or a washing step, which reduces the water consumption for this species compared to the other species. The dry weight biomass content of carotenoids is lower for *Haematococcus pluvialis* than for *Dunaliella salina*, which leads to a larger production scale for *Haematococcus pluvialis*. This is only partially compensated by the lower market volume for astaxanthin compared to the market volume of β-carotene. In the anaerobic digestion value chains, fertilizer and CO_2 are generated in an aqueous phase, which is assumed to be recycled to the cultivation stage. The syngas production in the gasification value chains is larger than in the other design options. In the pyrolysis design options, the most diesel and gasoline is produced.

Economic analysis results

The economic results for the optimal value chains are provided in Table 5. The NPV for the fish larvae feed value chain is higher due to the lower operational and investment costs. These lower costs can be explained by the single cultivation stage. In the ten value chains that produce carotenoids, the *Haematococcus pluvialis* value chains have higher NPVs than the *Dunaliella salina* value chains. Although the larger scale corresponds to higher investment and operational costs, astaxanthin has a higher price. The higher revenues are sufficient to offset the higher costs. The anaerobic digestion value chains have a higher NPV than the gasification, torrefaction and pyrolysis value chains, which is mainly explained by the higher investment costs and the hydrogen cost for the biocrude refining.

Environmental analysis results

The environmental results for the optimal value chains are provided in Table 6, including both the endpoint and the underlying midpoint environmental impact categories. The fish larvae feed value chain only has negative environmental impacts, except for the ΔTETP impact category. The ΔIRP category is negative for all value chains due to the upstream impact of electricity. The fossil-based reference value chain for astaxanthin has a higher environmental impact than the β-carotene reference value chain. However, due to the larger production scale, the *Haematococcus pluvialis* value chains have lower environmental savings on most environmental impact categories.

In Fig. 5, the contribution of the midpoint indicators to the endpoint indicators is analyzed to identify the most relevant midpoint indicators. In the endpoint impact category ΔHH, ΔGWP and ΔPMFP have the highest contribution. In the endpoint impact category, ΔEQ, ΔGWP and ΔTAP have the highest contribution. The last endpoint impact category, ΔRS, is mainly determined by ΔFFP. Therefore, the main midpoint categories that will be further assessed are ΔGWP, ΔPMFP, ΔTAP and ΔFFP.

Interpretation

The contribution of the different process steps to the economic investment and operational costs for the Pareto-optimal value chains is illustrated in Fig. 6. The highest investment costs are caused by the spray dryer, pond liner, land costs and the preharvesting membrane. The main operational costs are the indirect costs, which include the personnel costs and insurance and repair costs of the equipment.

Fig. 7 provides the contribution analysis for the main midpoint indicators. The main contributor to all environmental impacts is the upstream impact of the nutrients and CO_2 in the cultivation stage. The electricity consumption during the drying stage has a significant contribution to all environmental impact categories as well.

In the sensitivity analysis the most crucial parameters for each objective in each value chain are identified. A first iteration of the sensitivity analysis indicated that the carotenoid content, carotenoid price, fish larvae feed price, carotenoid reference impact, fish larvae feed reference impact and weighted average cost of capital shared approximately ninety percent of all variation for all impact categories and all value chains. As the cost and environmental impacts of the algal-based production value chains is compensated by the price and reference impact of the carotenoids and fish larvae feed, these parameters are important and their uncertainty should be systematically analyzed.

However, this first iteration only provides limited insights on the importance of underlying process parameters that differentiate the value chains. Therefore, the sensitivity analysis has been iterated for a second time without these crucial parameters to identify other important parameters as well. The results of the second iteration of the sensitivity analysis are provided in Table 7. A positive value means that an increase in this parameter will ensure an increase in the corresponding indicator. The value indicates the percentage of change in the indicator explained by this parameter. In the second iteration, the most important parameters for most impact categories are either the process parameters that induce a loss of biomass or carotenoids in the process, during drying, extraction or harvesting; and/or the growth-related parameters, such as the correction factor for the lower solar irradiation in Belgium. Also, the upstream environmental impact of electricity is an important parameter. For the ΔGWP and ΔPMFP indicators, the CO_2 requirement and fixation efficiency of the algae in open pond cultivation are crucial as well. The KNO_3 requirement and NH_3 emissions are important for the ΔTAP impact category for the *Haematococcus pluvialis* value chains. The material requirement for the liner of the open ponds is also important for the ΔFFP in the Ns FLF value chain, explaining 8% of the total variation for this impact category.

Discussion

The crucial parameters that defined the economic and environmental potential of the optimal value chains were mainly the price content and reference impact of the carotenoids and biomass. Therefore, research should focus on limiting losses of these components throughout the chain. Another important focus is the inclusion of a medium recycling step to lower the consumption of water and salt. A third important focus are the growth parameters. As algae differ in productivity and composition, the selection of the optimal algae is crucial to define an optimal algae-based biorefinery. Metabolic engineering can also play a role in obtaining the optimal algae strand.[74]

The superstructure as optimized in this paper contained both high-value and low-value applications. The production scale was restricted by the smallest market volume of the products. According to the results, the optimal biorefinery has a limited scale and bioenergy is only produced as a byproduct from the residual biomass. The lipid fraction of the biomass contains the carotenoids. However, it can also be converted to biofuel. All optimal value chains that refined the biomass into multiple products, preferred the purification of the carotenoids. In this way, the production scale remained limited. In the fish larvae feed value chain, the biomass was not refined but sold as whole. As the environmental impact savings were negative for these value chains, all production scales correspond to a Pareto-optimal scenario until a production scale of 114 ton biomass·year^{-1} is reached. If the production scale is between 114 ton biomass·year^{-1} and 132 ton biomass·year^{-1}, the environmental performance can be improved by reducing the production scale. At the production scale of 114 ton biomass·year^{-1}, the value chain cultivating *Haematococcus pluvialis* for carotenoids and fertilizer has a higher NPV than the value chain cultivating *Nannochloropsis* sp. for fish larvae feed.

A major outcome from the model is that food or feed-based microalgae biorefineries with a limited production scale were identified as more economically and environmentally optimal than fuel-based microalgae biorefineries on a large scale. A lot of current microalgae biorefinery studies still focus on energy as the main end product.[75, 76] According to our results, it would be more appropriate to focus on food or feed as main end products, and consider energy as a side product.

The fish larvae feed that is produced in the optimal economic value chain is used for the early life stages of fish larvae. As a specialty feed, the price for this product is high compared to other feed. The algae that are currently sold for this purpose have been grown in the ProviApt reactor and have been freeze dried instead of spray dried. This value chain is also economically profitable, although less profitable than the optimal fish larvae feed value chain as identified in this study. Quality considerations can have additional influences and affect the final price of the product. However, as no reliable estimate for this relation was available, it was not included in the model.

As the larvae feed scenario only sells larvae feed, it is strictly speaking no biorefinery as only one product is valorized. However, this scenario has been included as the larvae feed is currently commercially produced. This way, this scenario acts as a benchmark scenario to compare with other biorefinery scenarios. According to our results and given the current market situation, none of the included biorefinery scenarios has higher profits than the larvae feed scenario.

The lipid fraction, containing the carotenoids, contributes much more to the revenues than the residue fraction. However, multiple opportunities exist to further valorize this residue fraction.[77] Current research increasingly focusses on the valorization of the protein or polysaccharide fraction, which can also be extracted from the residual fraction.[78-80] However, a clear market for these applications does not exist yet. If multiple fractions can be valorized, the biorefinery concept can be further elaborated. Although the current microalgae market mainly exists for the whole biomass fraction or the lipid fractions, a biorefinery with a full range of end products may be able to economically compete with these applications. This way, also medium-value applications such as chemicals or plastics might become economically and environmentally feasible. A recommendation towards the chemical researchers, based on our results would therefore be to more focus on residual fractions and their applications instead of the lipid fractions, which already have applications. The production of biofuels from the lipid fraction, which still receives a lot of attention, can only become economically viable if the residual fraction is valorized to a large extent. In addition, the further valorization of this residual fraction can also reduce the environmental impact.

The fish larvae feed value chain has a higher environmental impact than the reference scenario. The reference scenario was based on conventional fishmeal.[81] However, there is no environmental impact category included in the ReCiPe 2016 indicator set that includes the environmental impact of overfishing or biotic resource use in general. The additions of a biotic resource indicator can overcome this gap.[82]

The land used in the optimal value chains varies between 28 and 114 hectares. Although algae can grow on degraded land and the cultivation can be done on different locations, finding this large land area in a small and densely populated country such as Belgium will be difficult. However, the algae cultivation does not have to be restricted to one location. An interesting extension to the model would be to find the optimal distribution of locations and to optimize the logistics of algae value chains in the model, based on existing logistic analyses.[83, 84]

The use of equipment is often neglected in LCA studies. However, the upstream impact of the liner was identified as an important parameter for the FFP of the fish larvae feed value chain. Neglecting the upstream impact of the pond manufacture would have had a significant impact on the outcome result. Therefore, the environmental impact of equipment cannot be excluded per se.[85]

The superstructure contains a wide range of different microalgae biorefinery value chains. However, the aim is not to capture all potential value chains. The preference was given to processes with a higher TRL to minimize the uncertainty. New processes and improved data can be added to extend and update the superstructure. The superstructure can also be extended to include other applications. A review by Laurens *et al*. discussed a broad range of valorization possibilities and their market potential.[26] For example, different fractions of microalgae have cosmetic attributes and can therefore be used as moisturizers, sunscreen, whitening, hair care or anti-aging products.[86] Also, other fractions, such as the protein fraction have an important commodity value and can be sold separately.[26, 87] Although clear market possibilities can be identified, the technological process steps to obtain these different fractions are still under development.

The optimization model was restricted to 3% of the end product market volume. This constraint was included to avoid oversupply of the market, which could decrease the price of the end products, given the price elasticity of supply. This constraint also allowed high-volume products such as biofuels to benefit from the economies-of-scale that are induced by their higher production scale. The inclusion of this constraint enables the comparison of low-volume and high-volume products, which would not be feasible otherwise.

The environmental indicators were optimized relative to a reference scenario, using the conventional production processes, as environmental savings. This means that a value chain that has a lower environmental impact than the reference scenario will be optimal if it produces at the maximal scale. By selecting the environmental savings as an objective, all objectives will be maximized in the optimization problem. If one of the objectives would be minimized instead, the production scale would function as a trade-off, and a continuous Pareto frontier would be assumed. However, this also means that the optimal value of this objective would be zero and no biomass would be produced. Although all these processes still have an absolute environmental impact on the environment and a negative absolute environmental impact should remain the end goal, an environmental saving compared to the reference scenario can already be considered an improvement and was therefore selected as the objective in the optimization problem.

The sensitivity analysis assumed a triangular distribution on all parameters to identify the most sensitive parameters. To obtain an uncertainty range on the output indicators, a more accurate distribution needs to be added to all parameters. However, a large number of parameters in different dimensions were included and some parameters, such as the productivity of the algae, were highly uncertain. the main result of this MOO-extended ETEA is not the value of the output indicators, but the identification of the optimal value chains and the most sensitive parameters, underlying the output indicators. More specific uncertainty distributions on all parameters would lead to a large range of the output indicators, which would not provide any useful information for the objectives of this paper.

In this study, the MOO analysis was performed using the ε-constraint method to transform the problem with four objectives into multiple iterations of a single-objective problem. Another possibility to deal with the multiple objectives would be the use of evolutionary algorithms, such as the NSGA III algorithm.[88] The advantage of these algorithms is that they can better handle non-linear problems and can include more objectives. The use of these algorithms to integrate MOO, LCA, process design and economic costs has also previously been used.[89] However, evolutionary algorithms are not able to confirm a solution as the global optimal. As the differences between the different process options in a process step that does not have a large contribution can be very small, evolutionary algorithms may have a hard time to identify the global optimal value chain. The ε-constraint method, as implemented in this study, is the generic version. An augmented version was developed in order to accelerate the process.[90] The implementation of this augmented version could be an interesting addition to the MOO-extended ETEA model.

The indicators used for the optimization are the three endpoint indicators of the ReCiPe 2016 method. Optimizing the seventeen midpoint indicators instead would provide more information. However, using the ε-constraint method, this would lead to a large amount of iterations and the optimization problem would become complex. The use of evolutionary algorithms can better handle multiple objectives. The NSGA III algorithm was developed to handle a large number of objectives. However, this algorithm has also not been tested for more than 15 objectives.[91] Another solution would be the development of a specific indicator set that selects the most important environmental indicators for the corresponding case study.[92] To enable the incorporation of a large amount of objectives, objective reduction methods have been developed as well, which could also be used to solve this problem.[93-95]

The results of the MOO-extended ETEA include the Pareto frontier with all Pareto-optimal value chains. No weights or subjective preferences have been included to choose between the different value chains. If one optimal value chain needs to be selected, the decision maker should decide on the weighting method after the Pareto-frontier has been calculated. This weighting method can for example use goal programming or be based on a multiple-criteria decision analysis.

All microalgae biorefinery value chains are situated at the same location in Belgium. As the solar irradiation was an important parameter in the model, an interesting extension of this model would be to optimize this location. This can have an impact on the cultivation characteristics, resource availability, prices, financial parameters and the upstream environmental impact factors. However, the current MOO method might need improvement, for example by using the augmented ε-constraint method to keep the computational efficiency at a satisfactory level.

The current multi-objective optimization takes economic and environmental objectives into account. However, to perform a full techno-sustainability analysis, social objectives need to be included as well.[96] The integration of such a social analysis into the MOO-extended ETEA framework would be an interesting path for further research.

The methodological framework of ETEA-MOO is used in this study for a microalgae-biorefinery case study, but can be used in other applications as well. The structure of the matrices used in the optimization problem is constructed in such a way that it can be generally applied for MOO problems including a superstructure with different process options and steps.

Conclusions

The MOO-extended ETEA as developed in this study provides useful insights into the broad range of potential microalgae biorefinery designs by the identification of the most promising value chains from different perspectives and the identification of the most sensitive parameters. The optimal value chain for a microalgal-based biorefinery consists of an open pond cultivation, a medium recycling step and a spray dryer. In the optimal economic value chain, *Nannochloropsis* sp. is cultivated in a one-stage cultivation process and the biomass is sold as fish larvae feed for early life phases of aquaculture. In the optimal environmental value chains, *Dunaliella salina* and *Haematococcus pluvialis* are cultivated in a two-stage process and β-carotene, fertilizer or energy, are produced using gasification or anaerobic digestion. Intermediate value chains include the two-stage cultivation of *Haematococcus pluvialis* and *Dunaliella salina* for astaxanthin, β-carotene and fertilizer production and energy products using anaerobic digestion, gasification, torrefaction or pyrolysis. The crucial parameters for economic and environmentally feasible value chains are the content, price and reference impact of the main end product, growth parameters and the loss of biomass and carotenoids alongside the value chain. The developed methodology is not only limited to microalgae biorefinery applications, but can assist in optimizing a broad range of new technologies towards economic and environmental perspectives and therefore accelerate the market introduction of green products and technologies.

Conflicts of interest

There are no conflicts to declare.

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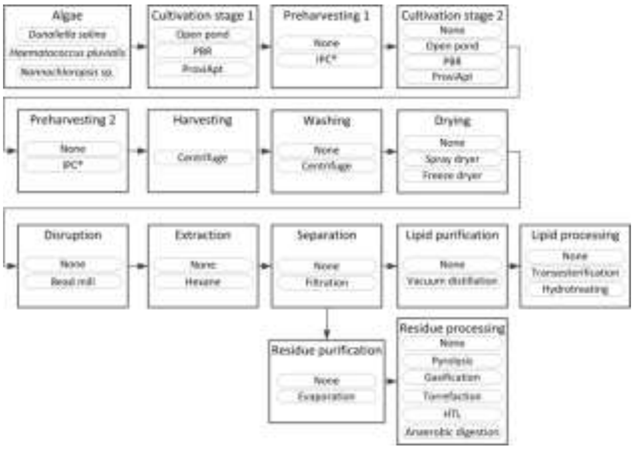

Fig. 1 Superstructure of all algae biorefinery process options included in the model, PBR=photobioreactor, IPC®=integrated permeate channel, HTL=hydrothermal liquefaction.

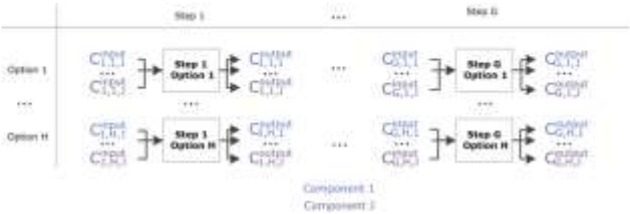

Fig. 2 Continuous decision variables used in the optimization model where component *j* is an input or output to process option *h* of process step *g*

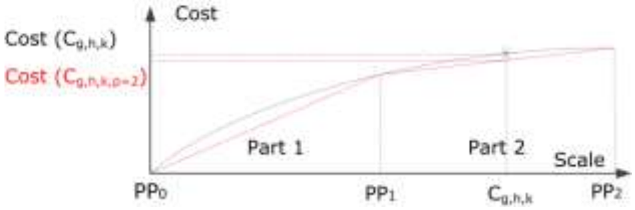

Fig. 3 Piecewise linear approximation with two partition points PP_1 and PP_2, dividing the cost curve in two parts, Part 1 and Part 2. The cost $C_{g,h,k}$ is the real cost, while the cost $C_{g,h,k,p=2}$ is the cost approximated by the linear approximation curve that is situated in Part 2 of the cost curve.

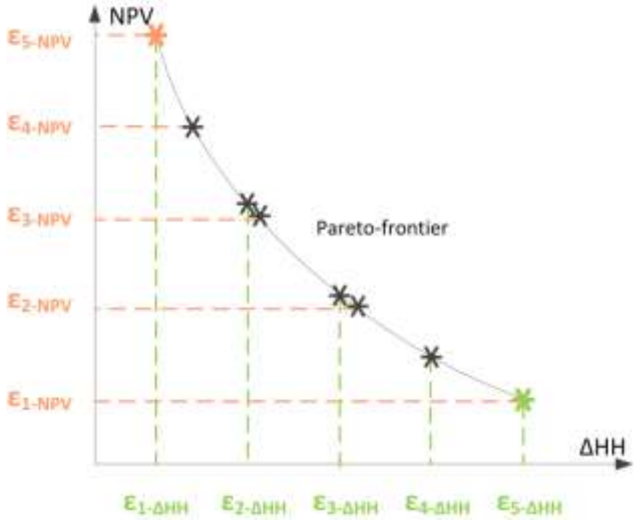

Fig. 4 Example of the ε-constraint method to obtain the Pareto-frontier with two objectives, ΔHH and NPV, and five ε-iterations for both objectives, ΔHH =human health savings, NPV=Net Present Value.

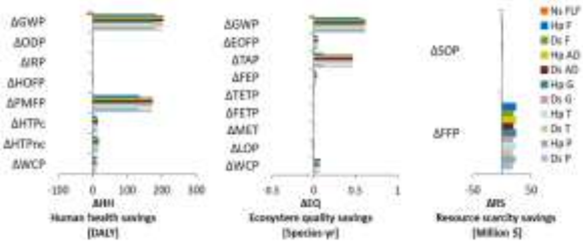

Fig. 5 Contribution of the midpoint indicators to the endpoint indicators for all Pareto-optimal scenarios, ΔHH = Human health savings; ΔEQ = Ecosystem quality savings; ΔRS = Resource scarcity savings; ΔGWP = Global warming potential; ΔODP = Ozone depletion potential; ΔIRP = Ionizing radiation potential; ΔPMFP = Particulate matter formation potential; ΔEOFP = Photochemical oxidant formation potential for ecosystems; ΔHOFP = Photochemical oxidant formation potential for humans; ΔTAP = Terrestrial acidification potential; ΔFEP = Freshwater eutrophication potential; ΔHTPc = Human toxicity potential cancer; ΔHTPnc = Human toxicity potential non-cancer; ΔTETP = Terrestrial ecotoxicity potential; ΔFETP = Freshwater ecotoxicity potential; ΔMETP = Marine ecotoxicity potential; ΔLOP = Agricultural land occupation potential; ΔWCP = Water consumption potential; ΔSOP = Surplus ore potential; ΔFFP = Fossil fuel potential.

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Fig. 6 Contribution analysis of the different process steps to the investment and operational costs of all Pareto-optimal scenarios

Fig. 7 Contribution analysis of the different process steps to the main environmental output indicators for all Pareto-optimal scenarios, GWP=global warming potential, PMFP=particulate matter formation potential, TAP=terrestrial acidification potential, FFP=fossil fuel potential.

Table 1 Summary of the main growth parameters, C_0=initial concentration, c_1=end concentration 1st stage, c_2=end concentration 2nd stage, $time_1$=cultivation period 1st stage, $time_2$=cultivation period 2nd stage, r_1=maximum specific growth rate 1st stage, r_2=maximum specific growth rate 2nd stage, $productivity_1$=productivity 1st stage, $productivity_2$=productivity 2nd stage, PBR=photobioreactor.

Species	*Dunaliella salina*			*Haematococcus pluvialis*			*Nannochloropsis* sp.		
Cultivation option	Open pond	PBR	ProviApt	Open pond	PBR	ProviApt	Open pond	PBR	ProviApt
C_0 [$g \cdot l^{-1}$]	0.06	0.23	0.23	0.06	0.50	0.50	0.2	0.25	0.25
C_1 [$g \cdot l^{-1}$]	0.35	1.41	1.41	0.35	2.90	2.90	0.73	1.75	1.75
C_2 [$g \cdot l^{-1}$]	0.40	1.59	1.59	0.40	3.27	3.27	0.82	1.98	1.98
$Time_1$ [days]	23	8	3	23	14	5	29	7	3
$Time_2$ [days]	6	2	1	6	4	1	9	2	1
r_1 [day^{-1}]	0.12	0.35	0.90	0.12	0.20	0.52	0.08	0.41	1.05
r_2 [day^{-1}]	0.08	0.23	0.60	0.08	0.13	0.35	0.06	0.27	0.70
$Productivity_1$ [$g \cdot l^{-1} \cdot day^{-1}$]	0.013	0.142	0.363	0.013	0.168	0.441	0.018	0.202	0.517
$Productivity_2$ [$g \cdot l^{-1} \cdot day^{-1}$]	0.007	0.081	0.057	0.008	0.101	0.069	0.009	0.127	0.081

Table 2 Overview of the notations used in the MOO problem

	Set of indices
G	set of process steps, indexed by g
H	set of process options of step g, indexed by h
J	set of mass components of option h of step g, indexed by j
K	set of equipment units of option h of step g, indexed by k
P	set of partition parts, indexed by p
IT_{npv}	set of iterations for the ε-constraint of NPV, indexed by it-npv
IT_{hh}	set of iterations for the ε-constraint of HH, indexed by it-hh
IT_{eq}	set of iterations for the ε-constraint of EQ, indexed by it-eq
IT_{rs}	set of iterations for the ε-constraint of RS, indexed by it-rs
	Variables
$a_{g,h,j}$	component j of option h of step g
$b_{g,h}$	binary variable of step g, option h
$c_{g,h,k,p}$	capacity at part p of the equipment k of option h of step g
$d_{g,h,k,p}$	binary variable selecting part p of the cost curve of equipment k of option h of step g
$x_{g,h,j,k,p}$	decision variable, consisting of $a_{g,h,j}$, $b_{g,h}$, $c_{g,h,k,p}$, $d_{g,h,k,p}$
$e_{g,h,k,p}$	continuous variable to calculate the cost at part p of equipment k of option h of step g
$f_{g,h,k,p}$	continuous variable to calculate the cost at part p of equipment k of option h of step g
$PP_{g,h,k,p}$	Partition point of part p of equipment k of option h of step g
	Parameters
$\alpha_{g,h,k}$	reference capacity of equipment k of option h of step g
$\beta_{g,h,k}$	power exponent of equipment k of option h of step g
$M_{g,h,j}$	upper bound of continuous decision variable $a_{g,h,j}$
$\gamma_{g,h,j}$	component j from input to output of option h of step g
$\delta_{g,h,k}$	reference price of equipment k of option h of step g
$\varepsilon_{it\text{-}npv}$	epsilon-constraint for NPV indexed by amount of iterations
$\varepsilon_{it\text{-}hh}$	epsilon-constraint for HH indexed by amount of iterations
$\varepsilon_{it\text{-}eq}$	epsilon-constraint for EQ indexed by amount of iterations
$\varepsilon_{it\text{-}rs}$	epsilon-constraint for RS indexed by amount of iterations
$npv_{g,h,j,k,p}$	parameter to multiply with $x_{g,h,j,k,p}$ to calculate the NPV savings
$hh_{g,h,j,k,p}$	parameter to multiply with $x_{g,h,j,k,p}$ to calculate the HH savings
$eq_{g,h,j,k,p}$	parameter to multiply with $x_{g,h,j,k,p}$ to calculate the EQ savings
$rs_{g,h,j,k,p}$	parameter to multiply with $x_{g,h,j,k,p}$ to calculate the RS savings

Table 3 Main results for the value chains of the Pareto frontier, NPV=net present value, ΔHH=human health savings, ΔEQ=ecosystem quality savings, ΔRS=resource scarcity savings, bm=biomass.

	NPV [10^6 €]	ΔHH [DALY]	ΔEQ [species·yr]	ΔRS [10^6 $]	Scale [ton bm·yr^{-1}]
Ns FLF	180	-20	-0.07	-1.1	132
Hp F	155	353	0.80	23.8	331
Ds F	17.6	415	1.21	18.7	149
Hp AD	154	353	0.80	23.8	331
Ds AD	17.1	415	1.21	18.7	149
Hp G	151	349	0.79	24.0	331
Ds G	15.1	414	1.21	18.8	149
Hp T	150.6	349	0.79	24.0	331
Ds T	15.0	414	1.21	18.7	149
Hp P	150.4	351	0.79	24.0	331
Ds P	14.9	415	1.21	18.7	149

Table 4 Summary of the mass and energy balance of the Pareto-optimal value chains

Parameter	Ns FLF	Hp F	Ds F	Hp AD	Ds AD	Hp G	Ds G	Hp T	Ds T	Hp P	Ds P
Water [dam^3]	224	434	550	598	609	434	550	434	550	434	550
Salt [kton]	3.48	0.00	26.1	0.00	26.1	0.00	26.1	0.00	26.1	0.00	26.1
CO_2 [kton]	5.84	14.7	6.62	14.7	6.62	14.7	6.62	14.7	6.62	14.7	6.62
Nutrients [kton]	3.32	11.8	5.34	11.8	5.34	11.8	5.34	11.8	5.34	11.8	5.34
Hexane [ton]	0.00	96.4	43.6	96.4	43.6	96.4	43.6	96.4	43.6	96.4	43.6
Hydrogen [kton]	0.00	0.00	0.00	0.00	0.00	34.8	12.7	46.7	17.1	60.87	22.2
Electricity [GWh]	17.3	79.5	32.5	80.0	32.7	79.8	32.6	79.6	32.6	076.7	32.6
Heat [GWh]	0.00	0.00	0.00	1.66	0.60	0.03	0.01	0.04	0.02	0.06	0.02
Fertilizer [kton]	0.00	2.45	0.88	0.19	0.07	0.00	0.00	0.00	0.00	0.00	0.00
Fish larvae feed [kton]	1.2	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Carotenoids [ton]	0.00	84.0	111	84.0	111	84.0	111	84.0	111	84.0	111
Biogas [GWh]	0.00	0.00	0.00	6.73	2.42	0.00	0.00	0.00	0.00	0.00	0.00
Syngas [GWh]	0.00	0.00	0.00	0.00	0.00	14.0	5.10	9.27	3.39	7.22	2.64
Diesel [m^3]	0.00	0.00	0.00	0.00	0.00	122	44.6	164	59.8	213	77.9
Gasoline [m^3]	0.00	0.00	0.00	0.00	0.00	113	41.3	152	55.4	197	72.1
CO_2 [dam^3]	0.00	0.00	0.00	275	99.1	0.00	0.00	0.00	0.00	0.00	0.00
Wastewater [dam^3]	227	440	565	440	565	440	565	440	565	440	565
Emissions [kton]	4.85	12.3	5.56	12.3	5.56	12.3	5.56	12.3	5.56	12.3	5.56
Land [ha]	28	114	52.1	114	52.1	114	52.1	114	52.1	114	52.1

Table 5 Economic results for Pareto-optimal value chains, NPV=net present value

	NPV [10^6 €]	Investment costs [10^6 €]	Operational costs [10^6 €·yr^{-1}]	Revenues [10^6 €·yr^{-1}]
Ns FLF	180	14.9	3.83	38.16
Hp F	155	34.8	9.39	42.11
Ds F	17.6	19.1	5.85	11.18
Hp AD	154	35.3	9.50	42.07
Ds AD	17.1	19.4	5.90	11.17
Hp G	151	38.6	9.73	42.07
Ds G	15.1	21.2	6.04	11.17
Hp T	150	38.7	9.75	42.05
Ds T	15.0	21.3	6.04	11.17
Hp P	150	38.7	9.77	42.05
Ds P	14.9	21.3	6.05	11.16

Table 6 Environmental savings results of all Pareto-optimal value chains

Parameter[a]	Ns FLF	Hp F	Ds F	Hp AD	Ds AD	Hp G	Ds G	Hp T	Ds T	Hp P	Ds P
ΔHH	-19.6	353	415	353	415	349	414	349	414	351	415
ΔEQ	-0.07	0.80	1.22	0.80	1.21	0.79	1.21	0.79	1.21	0.79	1.21
ΔRS	-1.2	23.8	18.7	23.8	18.7	24.0	18.8	24.0	18.7	24.0	18.7
ΔGWP	-17	198	219	197	219	195	218	195	218	195	218
ΔODP	-112	15.1	577	5.51	573	-7.28	569	-7.20	569	-7.07	569
ΔIRP	-10.1	-29.8	-9.79	-30.0	-9.87	-29.9	-9.85	-29.9	-9.83	-29.9	-9.84
ΔHOFP	-12.7	298	357	297	357	296	357	298	357	301	359
ΔPMFP	-2.13	213	275	213	275	210	274	210	274	212	275
ΔEOFP	-13.1	310	371	309	371	309	371	310	371	313	372
ΔTAP	-0.05	0.52	2.19	0.52	2.19	0.51	2.19	0.51	2.19	0.51	2.19
ΔFEP	-2.9	46.1	53.3	47.9	53.9	46.2	53.3	46.2	53.3	46.3	53.3
ΔTETP	74.4	21.5	62.4	21.1	62.3	-625	-174	-760	-223	-519	-135
ΔFETP	-0.4	1.46	2.40	1.63	2.46	1.45	2.40	1.46	2.40	1.47	2.40
ΔMETP	-0.58	2.13	3.42	2.35	3.50	2.13	3.42	2.13	3.42	2.14	3.43
ΔHTPc	-0.01	3.19	4.24	3.28	4.27	3.22	4.26	3.24	4.26	3.26	4.27
ΔHTPnc	-0.27	1.87	2.65	1.99	2.70	1.91	2.67	1.94	2.68	1.97	2.69
ΔLOP	-1.01	-1.14	0.60	-1.21	0.57	-1.24	0.56	-1.21	0.57	-1.19	0.58
ΔSOP	-0.01	2.60	0.48	2.60	0.48	2.60	0.48	2.61	0.48	2.61	0.48
ΔFFP	-3.60	63.7	52.7	63.6	52.7	64.4	53.0	64.3	52.9	64.3	52.9
ΔWCP	-0.07	5.52	3.17	5.35	3.11	5.39	3.12	5.37	3.12	5.40	3.13

[a] ΔHH = Human health savings [DALY]; ΔEQ = Ecosystem quality savings [species·yr]; ΔRS = Resource scarcity savings [10^6 $]; ΔGWP = Global warming potential [10^6 kg CO_2-eq]; ΔODP = Ozone depletion potential [kg CFC_{11}-eq]; ΔIRP = Ionizing radiation potential [10^6 kBq Co-60-eq]; ΔHOFP = Photochemical oxidant formation potential for humans [10^3 kg NOx-eq]; ΔPMFP = Particulate matter formation potential [10^3 kg PM2.5-eq]; ΔEOFP = Photochemical oxidant formation potential for ecosystems [10^3 kg NOx-eq]; ΔTAP = Terrestrial acidification potential [10^6 kg SO_2-eq]; ΔFEP = Freshwater eutrophication potential [10^3 kg P-eq]; ΔTETP = Terrestrial ecotoxicity potential [10^3 kg 1,4-DCB-eq]; ΔFETP = Freshwater ecotoxicity potential [10^6 kg 1,4-DCB-eq]; ΔMETP = Marine ecotoxicity potential [10^6 kg 1,4-DCB-eq]; ΔHTPC = Human toxicity potential cancer [10^6 kg 1,4-DCB-eq]; ΔHTPnc = Human toxicity potential non-cancer [10^9 kg 1,4-DCB-eq]; ΔLOP = Agricultural land occupation potential [10^6 m^2 yr]; ΔSOP = Surplus ore potential [10^6 kg Cu-eq]; ΔFFP = Fossil fuel potential [10^6 kg oil-eq]; ΔWCP = Water consumption potential [10^6 m^3 water-eq].

Table 7 Sensitivity analysis results of the second iteration [%]

	Value chain	Ns FLF	Hp F	Ds F	Hp AD	Ds AD	Hp P	Ds P	Hp G	Ds G	Hp T	Ds T
NPV	Biomass loss in dryer [%]	-11										
	Solar irradiation correction factor [%]	-15	-16	-16	-17	-18	-17	-16	-16	-16	-18	-15
	Maximum biomass concentration cultivation [$g{\cdot}l^{-1}$]	+14	+23	+25	+24	+24	+23	+25	+24	+23	+24	+23
	Maximum specific growth rate [day^{-1}]	+14	+15	+16	+17	+17	+17	+16	+18	+15	+17	+15
ΔGWP	CO_2 fixation efficiency [%]	+27	+15	+10	+16		+16	+10	+16	+11	+16	
	CO_2 requirement [g CO_2 g $biomass^{-1}$]	-16					-10					
	Electricity impact [Impact kWh^{-1}]		-13		-12		-12		-11		-12	
	Carotenoid loss in extraction [%]			-13		-13		-13		-13		-14
	Biomass loss in dryer [%]			-14		-13		-14		-13		-14
ΔTAP	Carotenoid loss in extraction [%]			-28		-28		-28		-28		-28
	Biomass loss in dryer [%]			-29		-29		-28		-28		-29
	Biomass loss in centrifuge [%]			-11		-10		-10		-10		-10
	Biomass loss in washing step [%]					-10		-10		-10		-10
	KNO_3 requirement 1st growth stage [$g{\cdot}l^{-1}$]	-39	-29		-29		-28		-28		-27	
	NH_3 emission [g NH_3 g KNO_3^{-1}]	-15	-11		-12		-11		-11		-11	
	NH_3 emission impact [impact kg^{-1}]	-14	-12		-11		-12		-12		-12	
ΔPMFP	Electricity impact [Impact kWh^{-1}]	-11	-14		-14		-13		-13		-13	
	Biomass loss in dryer [%]		-13	-21	-11	-21	-11	-21	-11	-21	-12	-20
	Solar irradiation correction factor [%]											
	Carotenoid loss in extraction [%]		-13	-21	-13	-20	-12	-21	-12	-21	-11	-20
ΔFFP	Maximum biomass concentration cultivation [$g{\cdot}l^{-1}$]	+13	+13	+11	+13	+11	+12	+12	+14	+10	+14	+12
	Solar irradiation correction factor [%]	-18	-13	-12	-13	-13	-14	-10	-13	-12	-14	-11
	Maximum specific growth rate [day^{-1}]	+15	+12	+11	+13	+12	+12	+11	+13	+12	+13	+11
	Biomass loss in dryer [%]			-10		-10		-11		-11		-11
	Carotenoid loss in extraction [%]			-10		-10		-11		-10		-11
	Electricity impact [Impact kWh^{-1}]	-11	-15		-16		-16		-16		-14	

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