

BIANCA BARROS MARANGON

**BIOTECNOLOGIA DE MICROALGAS: AVALIAÇÃO DO CICLO DE VIDA DE
DIFERENTES ROTAS DE VALORIZAÇÃO DA BIOMASSA PRODUZIDA EM
ÁGUAS RESIDUÁRIAS**

Dissertação apresentada à Universidade Federal de Viçosa, como parte das exigências do Programa de Pós-Graduação em Engenharia Civil, para obtenção do título de Magister Scientiae.

Orientadora: Maria Lúcia Calijuri

**VIÇOSA - MINAS GERAIS
2021**

**Ficha catalográfica elaborada pela Biblioteca Central da Universidade
Federal de Viçosa - Campus Viçosa**

T

M311b
2021

Marangon, Bianca Barros, 1995-
Biotecnologia de microalgas : avaliação do ciclo de vida de
diferentes rotas de valorização da biomassa produzida em águas
residuárias / Bianca Barros Marangon. – Viçosa, MG, 2021.
103 f. : il. (algumas color.) ; 29 cm.

Inclui apêndices.

Orientador: Maria Lúcia Calijuri.

Dissertação (mestrado) - Universidade Federal de Viçosa.

Inclui bibliografia.

1. Microalgas. 2. Biocombustíveis. 3. Ciclo de vida -
Avaliação. 4. Briquetes (Combustível). I. Universidade Federal
de Viçosa. Departamento de Engenharia Civil. Programa de
Pós-Graduação em Engenharia Civil. II. Título.

CDD 22. ed. 662.88

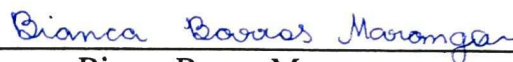
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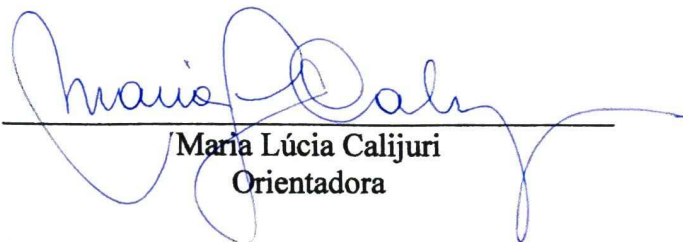
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APROVADA: 01 de março de 2021.

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Maria Lúcia Calijuri
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Aos meus pais Claudia e Elder.

Dedico.

AGRADECIMENTOS

Agradeço, primeiramente, a Deus, por mais essa conquista.

Agradeço à professora Maria Lúcia Calijuri, pela orientação e suporte durante todo o mestrado e por ter me recebido tão bem como membro da família SIGEOnPA.

Aos professores Paula Peixoto Assemany e Eduardo de Aguiar do Couto por terem acompanhado e ajudado no desenvolvimento deste projeto. Agradeço também aos professores Paula e Eduardo e à professora Maria do Carmo Calijuri, por terem aceitado participar da banca examinadora e darem suas valiosas contribuições.

Agradeço à Jaqueline de Siqueira Castro por todo o apoio antes e durante a elaboração deste trabalho. Agradeço também aos demais colegas de laboratório pela convivência prazerosa, em especial à Sabrina do Carmo Alves pela amizade.

À CAPES, pela concessão de bolsa de auxílio aos estudos (o presente trabalho foi realizado com apoio da Coordenação de Aperfeiçoamento de Pessoal de Nível Superior – Brasil (CAPES) – Código de Financiamento 001). Agradeço também aos professores e funcionários da UFV e ao Programa de Pós-Graduação em Engenharia Civil, pela oportunidade de realização deste curso;

Agradeço aos meus pais Cláudia e Elder por tornarem possível a realização deste sonho. Às minhas irmãs Bárbara e Marlene pelo carinho.

Por fim, agradeço a todos que não foram citados, nominalmente, mas que contribuíram para que este propósito fosse alcançado.

“Se você tem medo não o faça,
se você o está fazendo não tenha medo.”

Genghis Khan

RESUMO

MARANGON, Bianca Barros, M.Sc., Universidade Federal de Viçosa, março de 2021. **Biotecnologia de microalgas: avaliação do ciclo de vida de diferentes rotas de valorização da biomassa produzida em águas residuárias.** Orientador: Maria Lúcia Calijuri.

A biomassa de microalgas é uma fonte promissora de energia renovável, principalmente quando o seu crescimento está associado ao tratamento de efluentes. No entanto, a biotecnologia de microalgas ainda tem muito a melhorar em termos de produtividade, custos e impactos ambientais. Além disso, as rotas de valorização da biomassa algal precisam ser otimizadas para serem uma fonte sustentável e viável de bioenergia. Nesta pesquisa, o desempenho ambiental de duas rotas de valorização da biomassa (briquetes e bio-óleo) foi obtido por meio da avaliação do ciclo de vida com modelagem no software SimaPro®. No primeiro capítulo, dois reatores de tratamento de efluentes e crescimento de microalgas (um disperso e um aderido) foram comparados. Foram encontrados os potenciais impactos ambientais tanto da produção de biomassa quanto da valorização em briquetes. Com a avaliação do ciclo de vida, foi possível identificar que a secagem da biomassa, para fazer os briquetes, precisa consumir menos energia para compensar seu rendimento. As categorias de impacto que mais pressionaram o meio ambiente foram ecotoxicidade terrestre, eutrofização de água doce, escassez de recursos fósseis e mudança climática. Apesar disso, os cenários modelados ofereceram mais benefícios do que impactos ao meio ambiente, devido ao aproveitamento de águas residuárias para o crescimento das microalgas. No segundo capítulo, a valorização da biomassa em bio-óleo e o seu beneficiamento em diesel renovável foi estudado. O método de Monte Carlo foi aplicado para verificar as incertezas e a sensibilidade do modelo. Na avaliação do ciclo de vida dessa rota foi evidenciado que os parâmetros operacionais da liquefação hidrotérmica precisam ser otimizados para diminuir a demanda de energia do processo. Além disso, a fase aquosa gerada deve ser aproveitada para reduzir os potenciais impactos ambientais do ciclo de vida do bio-óleo. As categorias de impacto que mais pressionaram o meio ambiente foram eutrofização marinha, mudanças climáticas, ecotoxicidade marinha, depleção de fósseis, toxicidade humana e acidificação terrestre, nessa ordem de importância. A rota úmida (liquefação hidrotérmica) causou menos impactos que a rota seca (briquetes).

Palavras-chave: Microalgas. Bioenergia. Avaliação do ciclo de vida. Briquetes. Bio-óleo.

ABSTRACT

MARANGON, Bianca Barros, M.Sc., Universidade Federal de Viçosa, March, 2021. **Microalgae biotechnology: life cycle assessment of different valorization routes for wastewater grown biomass.** Advisor: Maria Lúcia Calijuri.

Microalgae biomass is a promising source of renewable energy, especially when its growth is associated with wastewater treatment. However, microalgae biotechnology still has much to improve in terms of productivity, costs and environmental impacts. In addition, algal biomass recovery routes need to be optimized to be a sustainable and viable source of bioenergy. In this research, the environmental performance of two biomass routes (briquettes and bio-oil) was obtained through life cycle assessment modeled on SimaPro® software. In the first chapter, two wastewater treatment reactors and microalgae growth (a dispersed and an adhered) were compared. The potential environmental impacts of both biomass production and recovery in briquettes were evaluated. With life cycle assessment, it was possible to identify that biomass drying, to make the briquettes, needs to consume less energy to offset its yield. The impact categories that put the greatest pressure on the environment were terrestrial ecotoxicity, freshwater eutrophication, fossil resource scarcity and climate change. Despite this, the modeled scenarios offered more benefits than impacts to the environment, due to the use of wastewater for the growth of microalgae. In the second chapter, the valorization of biomass in bio-oil and its processing in renewable diesel was studied. The Monte Carlo method was applied to check the model's uncertainties and sensitivity. In the life cycle assessment of this route, it was evidenced that the operational parameters of the hydrothermal liquefaction need to be optimized to decrease the energy demand of the process. In addition, the aqueous phase generated must be used to reduce the potential environmental impacts of the bio-oil life cycle. The impact categories that put the greatest pressure on the environment were marine eutrophication, climate change, marine ecotoxicity, depletion of fossils, human toxicity and terrestrial acidification, in that order of importance. The wet route (hydrothermal liquefaction) caused less impact than the dry route (briquettes).

Keywords: Microalgae. Bioenergy. Life cycle assessment. Briquettes. Bio-oil.

LISTA DE ILUSTRAÇÕES

Figure 4.1. System boundaries (a) scenario 1 – HRAP MB briquette; (b) scenario 2 – Hybrid reactor MB briquette.....	24
Figure 4.2. Potential environmental impacts of scenarios 1 – HRAP MB briquette and 2 – Hybrid reactor MB briquette, at each stage of the life cycle and percentage comparison between scenarios.	32
Figure 4.3. Percentage comparison of the potential environmental impacts of scenarios 1 – HRAP MB briquette and 2 – Hybrid reactor MB briquette, associated with known inputs from the technosphere.	35
Figure 4.4. Relative comparison between impact categories and between scenarios 1 – HRAP MB briquette and 2 – Hybrid reactor MB briquette (a) in the 10 impact categories and (b) in the 4 categories that most affected the environment.	40
Figure 4.5. The potential electricity yield of MB briquettes and demand for electricity in drying and in the complete life cycles of base scenarios 1 and 2, depending on the humidity of the MB.	42
Figure 4.6. Normalized environmental impacts for the sensitivity analyzes and relative comparison between (a) coal briquettes and MB briquettes with natural humidity reduced to 83% - scenario 1a and 85% - scenario 2a; (b) scenarios 1b and 2b, with coal briquettes.	44
Figure 5.1. Stages included in (a) the gate-to-gate system boundary of the microalgae bio-oil life cycle via HTL and (b) the bio-oil upgrading (base scenario)	60
Figure 5.2. Relative comparison between the potential environmental impacts of the life cycle of microalgae bio-oil via HTL (base scenario) (a) in the 18 categories evaluated and (b) in the 6 categories that most exerted pressure on the environment.	70
Figure 5.3. Percentage contribution (a) of the stages and (b) of the inputs and outputs modeled in the life cycle of the base scenario in the 6 impact categories in which the environment was most affected.....	72
Figure 5.4. Absolute uncertainties in the potential average environmental impacts of the bio-oil life cycle and the VC of the results.	75
Figure 5.5. Variation of potential normalized environmental impacts of the bio-oil life cycle (base scenario) caused by the sensitivity analyzes.	77
Figure 5.6. Comparison, using the Monte Carlo method, between the potential environmental impacts of the base scenario and the new scenario.	78

LISTA DE TABELAS

Table 4.1. Process systems and input data for LCIs of the modeled scenarios.	28
Table 4.2. LCI of scenarios 1 – HRAP MB briquette and 2 – Hybrid reactor MB briquette, to reach the FU of 1 MJ of gross energy production.	31
Table 5.1. Average, minimum, and maximum values of operational parameters of HTL and reaction yields, found in the literature.	62
Table 5.2. Elements of the microalgae bio-oil life cycle inventory via LHT.	67
Table 5.3. Changes in the LCI of the base scenario for calculating the model’s sensitivity by varying $\pm 10\%$ in the HTL heat of three different input parameters.	76

LISTA DE SIGLAS E ABREVIATURAS

ACV	Avaliação do ciclo de vida
BM	Biomassa de microalgas
CO ₂	Dióxido de carbono
eq	Equivalente
ETAR	Estação de tratamento de águas residuárias
FA	Fase aquosa
FG	Fase gasosa
FS	Fase sólida
FU	Functional unit
GEE	Gás de efeito estufa
HRAP	High-rate algal pond
HTL	Hydrothermal liquefaction
ICV	Inventário do ciclo de vida
LAT	Lagoa de alta taxa
LCA	Life cycle assessment
LCI	Life cycle inventory
LHT	Liquefação hidrotérmica
MB	Microalgae biomass
N	Nitrogênio
NTK	Nitrogênio total Kjeldahl
P	Fósforo
RB	Reator de biofilme
UF	Unidade funcional

SUMÁRIO

APRESENTAÇÃO.....	13
1. INTRODUÇÃO GERAL	14
2. HIPÓTESES.....	15
3. OBJETIVO GERAL	16
3.1. Objetivos específicos	16
4. A LIFE CYCLE ASSESSMENT OF ENERGY RECOVERY USING BRIQUETTE FROM WASTEWATER GROWN MICROALGAE BIOMASS	17
Abstract:.....	17
Keywords:.....	18
Abbreviation	18
4.1. Introduction.....	19
4.2. Material and methods.....	22
4.2.1. Goal and scope definition	23
4.2.2. Life cycle inventory (LCI), assumptions and limitations of the study	25
4.2.3. Life cycle impact assessment (LCIA)	29
4.2.4. Sensitivity analysis	29
4.3. Results and discussion	30
4.3.1. Classification and characterization	31
4.3.2. Normalization	39
4.3.3. Improvements to the life cycle of microalgae briquettes	41
4.4. Conclusion	46
4.5. References.....	47
5. ENVIRONMENTAL PERFORMANCE OF MICROALGAE HYDROTHERMAL LIQUEFACTION: LIFE CYCLE ASSESSMENT AND IMPROVEMENT INSIGHTS FOR A SUSTAINABLE RENEWABLE DIESEL	56
Abstract:.....	56
Keywords:.....	56
Abbreviations.....	56
5.1. Introduction.....	57
5.2. Material and methods.....	59
5.2.1. Goal and scope definition	59
5.2.2. Assumptions made for the life cycle inventory (LCI) and research limitations.....	61
5.2.3. Life cycle impact assessment (LCIA)	64
5.2.4. Interpretation of results.....	65
5.2.5. Obtaining a scenario with less environmental impact.....	66
5.3. Results and discussion	66

5.3.1.	Potential environmental impacts of the bio-oil life cycle.....	69
5.3.2.	Contribution analysis.....	71
5.3.3.	Uncertainty analysis	74
5.3.4.	Sensitivity analysis	75
5.3.5.	Improvements in the life cycle of microalgae bio-oil via HTL.....	78
5.4.	Conclusion	80
	References.....	81
6.	CONCLUSÃO GERAL	90
7.	SUGESTÃO PARA PESQUISAS FUTURAS.....	90
8.	REFERÊNCIAS GERAIS	92
	APÊNDICES	97

APRESENTAÇÃO

No presente estudo deu-se ênfase na avaliação do ciclo de vida (ACV) de rotas de valorização da biomassa de microalgas (BM) cultivada em esgoto doméstico. A proposta teve como ponto de partida as pesquisas de ASSIS (2016), COSTA (2016) e COUTO (2016), desenvolvidas no Núcleo de Pesquisas Ambientais Avançadas (nPA), na Universidade Federal de Viçosa (UFV).

ASSIS (2016) utilizou esgoto doméstico, coletado após o tratamento em reator Upflow Anaerobic Sludge Blanket (UASB), como meio de cultivo para as microalgas em um reator híbrido formado por lagoa de alta taxa (LAT) e reator de biofilme (RB). Nesta pesquisa foram comparadas as formas de tratamento do efluente e cultivo de microalgas. COSTA (2016) utilizou esgoto doméstico e agroindustrial para a produção de briquetes juntamente a epicarpo de pinhão manso. Esta pesquisa caracterizou a biomassa de microalgas para produzir os briquetes e obteve o seu potencial energético. COUTO (2016) utilizou esgoto doméstico, coletado após o tratamento em reator UASB, como meio de cultivo para as microalgas em LAT e realizou a liquefação hidrotérmica (LHT) da biomassa. Essa pesquisa caracterizou o bio-óleo, a fase aquosa e a fase sólida produzidos. Esses três trabalhos avaliaram, sob a perspectiva técnica, a aplicação da BM nos sistemas de cultivo e rotas de valorização supracitadas. A presente pesquisa visou agregar a ACV das configurações experimentais propostas pelos autores mencionados e dar continuidade no desenvolvimento das tecnologias estudadas.

Este documento foi organizado, além de uma introdução geral, hipóteses, objetivo geral e específicos e as considerações finais, em dois artigos científicos. O primeiro tem como foco a ACV da rota de briquetagem da BM, avaliando-se, principalmente, dois diferentes reatores de crescimento das microalgas (LAT e reator híbrido com LAT e RB). Este artigo foi aceito para publicação no periódico internacional *Journal of Environmental Management*. O segundo artigo, tem o objetivo de estudar a rota de bio-óleo, por meio da LHT da BM, buscando, com a ACV, meios para otimizar este processo. Foram apresentadas também, ao final do documento, as perspectivas e sugestões para pesquisas futuras, que surgiram da experiência adquirida durante a execução dos trabalhos.

1. INTRODUÇÃO GERAL

Diante da elevada demanda energética e das mudanças climáticas, fontes alternativas de energia são importantes opções a serem buscadas e desenvolvidas (SANKARAN et al., 2018; SHUBA; KIFLE, 2018). Cerca de 81% da energia primária utilizada no globo é proveniente de fontes não renováveis, ocasionando impactos ambientais, como a emissão de dióxido de carbono (CO₂), o gás de efeito estufa (GEE) mais encontrado na atmosfera (RITCHIE; ROSER, 2017). Completando as preocupações atuais, espera-se que as estações de tratamento de águas residuárias (ETARs) busquem sustentabilidade, eficiência energética e recuperação de recursos, além de tratar efluentes (MO; ZHANG, 2013). Isso porque o tratamento de águas residuárias é uma atividade impactante, mas que não pode ser evitada. Ademais recuperação de água, nutrientes e energia permite que uma ETAR convencional, que emite poluentes e dissipa energia, se aproxime de uma estação que beneficia o ambiente (HAO et al., 2019).

Nesse sentido, a ACV é uma ferramenta imprescindível para auxiliar na tomada de decisão acerca das tecnologias de tratamento de águas residuárias voltadas para a recuperação de recursos e para o seu desenvolvimento e melhoria (FANG et al., 2016). Com a ACV é possível obter-se o desempenho ambiental de produtos e serviços. Uma ACV pode abordar desde a extração da matéria prima à disposição final do produto e reciclagem de material, ou apenas uma etapa da produção. Na ACV, o produto é avaliado e sistemas de energia, produção e transporte alternativos são pensados, visando a diminuição dos impactos ambientais daquele produto ou serviço (GUINÉE et al., 2002; ISO, 2006a, 2006b; PRÉ, 2016).

Em vista da recuperação de recursos, tem-se a oportunidade para o desenvolvimento de tecnologias que envolvem microalgas (YADAV et al., 2020). Pesquisas com ACV reportaram que soluções alternativas de tratamento de águas residuárias, como a de microalgas, tem melhor desempenho ambiental quando comparadas aos sistemas convencionais de lodos ativados, por consumir menos energia e insumos químicos (ARASHIRO et al., 2018; COLZI LOPES et al., 2018; GARFÍ; FLORES; FERRER, 2017). Ademais, o uso de águas residuárias como meio de cultivo para as microalgas, evitando o uso de recursos (água e fertilizantes contendo N, P e K) causa menos impactos ambientais, quando comparado com o cultivo em meio sintético, em que água e fertilizantes são requeridos (SCHNEIDER et al., 2018).

Durante o tratamento de águas residuárias, as microalgas se desenvolvem e capturam nutrientes como nitrogênio (N) e fósforo (P), que podem ser recuperados pela biomassa produzida

(BATTEN et al., 2013; BEUCKELS; SMOLDERS; MUYLEAERT, 2015). Além disso, as microalgas tem células repletas de lipídios, proteínas e carboidratos e podem ser transformadas em bioenergia e em outros bioprodutos (FERNÁNDEZ et al., 2021). Sabe-se que as rotas energéticas da biomassa algal representam um aspecto favorável à redução dos impactos ambientais do tratamento de águas residuárias (COLZI LOPES et al., 2018). Entretanto, os processos para a conversão da biomassa em bioenergia, são onerosos e impactantes. Contudo, o uso de biocombustíveis vem sendo impulsionado, principalmente por políticas de segurança energética e mitigação de emissões de GEE. Contudo, CARNEIRO et al. (2017) acreditam que o desempenho energético e ambiental da bioenergia de microalgas pode ser melhorado com investigação das vias de produção, utilizando a ACV.

Portanto, o objetivo deste trabalho foi avaliar, por meio da ACV, diferentes rotas de valorização da biomassa produzida via biotecnologia de microalgas, visando aprimorá-las. Isso pois a BM pode ser convertida em biocombustíveis por meio de diversos processos. Porém, para que o seu uso seja viável, as microalgas devem alcançar um melhor desempenho ambiental. Assim, apesar do potencial e vantagens das microalgas, alguns desafios ainda precisam ser vencidos. Dessa forma, espera-se contribuir para o aprimoramento e desenvolvimento das rotas de conversão da biomassa de microalgas em produtos mais sustentáveis, causando menos pressão ao meio ambiente. E, além disso, contribuir para preencher uma lacuna na literatura, discutindo métodos e tecnologias a serem aprimoradas para tornar a biotecnologia das microalgas ambientalmente viável e uma fonte potencial de energia renovável alternativa.

2. HIPÓTESES

- A maior produtividade de biomassa do painel híbrido, aliada à facilidade de colheita, torna esse tipo de cultivo mais favorável ambientalmente se comparado com o cultivo suspenso em lagoas de alta taxa.
- A energia produzida pelos briquetes, produzidos a partir da biomassa de microalgas cultivada em efluentes, compensa os impactos ambientais da sua rota de produção, principalmente a secagem da biomassa.
- A etapa de beneficiamento do bio-óleo de microalgas em combustível renovável impacta negativamente mais o meio ambiente que a obtenção do bio-óleo por meio da liquefação hidrotérmica da biomassa, visto à sua quantidade de entradas e saídas.

- A fase aquosa gerada como subproduto da liquefação hidrotérmica da biomassa de microalgas para a produção de bio-óleo causa mais impacto prejudicial ao meio ambiente que as demais saídas (a fase sólida e a gasosa), devido à sua composição.
- A obtenção de biocombustível sólido (briquetes) de biomassa de microalgas causa mais impactos ambientais adversos, se comparado ao biocombustível líquido (bio-óleo via liquefação hidrotérmica), visto a etapa de secagem da biomassa.

3. OBJETIVO GERAL

Avaliar, por meio da ferramenta de ciclo de vida, a sustentabilidade ambiental de rotas de valorização em bioenergia da biomassa produzida via biotecnologia de microalgas durante o tratamento de efluente.

3.1. Objetivos específicos

- Identificar os potenciais impactos ambientais no ciclo de produção dos briquetes, ocasionados pelo tipo de reator de cultivo da biomassa, comparando a lagoa de alta taxa e reator híbrido;
- Avaliar o quanto a secagem da biomassa, para obtenção dos briquetes, afeta o ciclo de vida e o ambiente;
- Propor melhorias para a mitigação dos potenciais impactos ambientais para a rota dos briquetes;
- Identificar os potenciais impactos ambientais do bio-óleo de biomassa, ocasionados, principalmente, pelos parâmetros operacionais da liquefação hidrotérmica;
- Avaliar o quanto as saídas (fase aquosa, sólida e gasosa) da liquefação hidrotérmica afetam nos potenciais impactos ambientais do ciclo de vida do bio-óleo;
- Propor melhorias para a mitigação dos potenciais impactos ambientais para a rota do bio-óleo;
- Comparar os potenciais impactos causados pela rota dos briquetes e do bio-óleo.

4. A LIFE CYCLE ASSESSMENT OF ENERGY RECOVERY USING BRIQUETTE FROM WASTEWATER GROWN MICROALGAE BIOMASS¹

Abstract: Microalgae biomass (MB) is a promising source of renewable energy, especially when the cultivation is associated with wastewater treatment. However, microalgae wastewater technologies still have much to improve. Additionally, microalgae biomass valorization routes need to be optimized to be a sustainable and feasible source of green bioenergy. Thus, this paper aimed to evaluate the environmental impacts of the production of briquettes from MB, cultivated during domestic wastewater treatment. Also, it was evaluated how much the drying of the MB affected the life cycle and the environment. Improvements in the life cycle to mitigate the environmental impacts of this energy route were proposed. Cradle-to-gate modeling was applied to obtain a life cycle assessment (LCA) from cultivation to the valorization of MB, through its transformation into a solid biofuel. With LCA, it was possible to identify which technical aspect of the process needs to be optimized so that environmental sustainability can be achieved. Two scenarios were compared, one with the microalgae growth in a high-rate algal pond (HRAP) (scenario 1) and the other in a hybrid reactor, formed by a HRAP and a biofilm reactor (BR) (scenario 2). LCA highlighted the electric power mix, representing, on average, 60% of the total environmental impacts in both scenarios. The valorization of MB in briquettes needs to consume less energy to offset its yield. The environment suffered pressure in freshwater eutrophication, due to the release of 3.1E-05 and 3.9E-05 kg of phosphorus equivalent; in fossil resources scarcity, with the extraction of 1.4E-02 and 4.5E-02 kg of oil equivalent; and in climate change, by the emission of 1.0E-01 and 1.9E-01 kg of carbon dioxide (CO₂) equivalent, in scenarios 1 and 2, respectively. Scenario 1 was highly damaging to terrestrial ecotoxicity, with the release of 3.5E-01 kg of 1,4 Dichlorobenzene, coming from the CO₂ used in MB growth. This category was the one that most negatively pressured the environment, differing from scenario 2, in which this input was not required. This was the only impact category in which scenario 2 had a better environmental performance when compared to scenario 1. Cotton, required in scenario 2, represented up to 87% of emissions in some of the evaluated categories. Despite the impacts that occurred in the two modeled scenarios, the environmental gains due to the use of wastewater for microalgae growth, replacing the synthetic

¹ Adaptado de MARANGON, B. B. et al. A life cycle assessment of energy recovery using briquette from wastewater grown microalgae biomass. **Journal of Environmental Management**, v. 285, January, n. 112171, maio 2021. <https://doi.org/10.1016/j.jenvman.2021.112171>.

cultivation medium, stood out. In the sensitivity analysis, two alternative scenarios were proposed: (i) electricity consumption for drying has been reduced, due to the natural decrease of MB humidity, and (ii) MB briquettes were considered a substitute for coal briquettes. Results indicated that pressures on climate change and fossil resource scarcity were eliminated in both scenarios and this also occurred for freshwater eutrophication in scenario 2. This paper contributes to the improvement and development of converting MB routes into more sustainable products, causing less pressure on the environment. Also, the study contributes to filling a gap in the literature, discussing methods and technologies to be improved, and consequently making microalgae biotechnology environmentally feasible and a potential renewable energy alternative.

Keywords: Bioenergy; Nutrient recycling; Biorefinery; Solid biofuel; LCA.

Abbreviation

1,4-DCB	1,4-dichlorobenzene
APOS – S	Allocation at the point of substitution – System
BR	Biofilm reactor
CFC11	Trichloromonofluoromethane
CHP	Combined heat and power
CO ₂	Carbon dioxide
Cu	Copper
{DE}	Germany
eq	Equivalent
FU	Functional unit
GHG	Greenhouse gas
{GLO}	Global
HHV	High heating value
HRAP	High-rate algal pond
IEA	International Energy Agency
ISO	International Organization for Standardization
K	Potassium
LCA	Life cycle assessment
LCI	Life cycle inventory

LCIA	Life cycle impact assessment
MB	Microalgae biomass
N	Nitrogen
P	Phosphorus
P ₂ O ₅	Phosphorus pentoxide (fertilizer)
PM _{2.5}	Particles smaller than 2.5 mm
SO ₂	Sulfur dioxide
{RoW}	Rest-of-the-World
SimaPro	System for Integrated Environmental Assessment of Products
WTP	Water treatment plant
WWTPs	Wastewater treatment plants

4.1. Introduction

The global energy matrix is composed mainly of fossil fuels, such as oil (32%), coal (27%), and natural gas (22%) (IEA, 2019a). The fact that most (81%) of the primary energy used in the globe comes from non-renewable sources, which can be depleted, creates insecurity (Yadav et al., 2020). Besides, the use of these fuels causes negative environmental impacts, such as the emission of carbon dioxide (CO₂), the most common greenhouse gas (GHG) in the atmosphere (Ritchie and Roser, 2017).

Thus, investment in renewable energy is essential to reduce CO₂ emissions and move towards policies to minimize the effects of climate change (Garrett-Peltier, 2017). In this context, a renewable and highly available resource, which is being increasingly used, is biomass (Cherubini, 2010; Hughes and Qureshi, 2014; Pandey et al., 2015; Yamakawa et al., 2018). Among the existing biomasses, microalgae biomass (MB) stands out in several aspects, such as high productivity throughout the year, non-competition with agricultural areas, and CO₂ absorption during its growth (Javed et al., 2019; Laurens et al., 2017; Peng et al., 2015). Also, wastewater can be used as a culture medium for microalgae, promoting the recovery of water bodies, by eutrophication prevention through nutrients assimilation during MB growth (Li et al., 2019; Park et al., 2018). It is important to highlight that wastewater treatment is an impacting activity, but it cannot be avoided. Therefore, the reduction of negative impacts, water reuse, energy production, and the recovery of nutrients in wastewater treatment plants (WWTPs) should be sought (Hao et al., 2019).

Despite the benefits of MB, one difficulty in implementing renewable energy projects is the financial subsidies designed for fossil fuels by governments of high-income countries (Monasterolo and Raberto, 2019). However, researches aimed to use MB biofuels as an alternative to fossil fuels are currently being developed (Khan et al., 2018) and can be considered a strategy to increase the attractiveness of these non-alternative sources. The MB can be converted into biofuels through chemical, biochemical, and thermochemical processes. However, for a wide and real scale utilization, microalgae must technically and economically compete with traditional fuels. The performance of some conversion routes is highly dependent on the MB lipid content, which is usually low in wastewater grown microalgae biomass, among other biochemical characteristics of this biomass. Therefore, several bioenergy routes have been evaluated, to find the best options.

Chew et al. (2017) pointed out that a biorefinery with direct combustion of MB to generate electricity is promising in terms of efficiency and can be combined with the co-combustion techniques of coal plants. According to Choi et al. (2019), the co-combustion of MB and coal is a process that may contribute to the reduction of CO₂ emissions. In this context, the briquetting process is applied to several biomasses, to transform them into a regular high-density solid and thus increase their performance as a fuel (Alanya-Rosenbaum and Bergman, 2019; Avelar et al., 2016; Ji et al., 2018; Saba et al., 2020; Wang et al., 2017). Although briquetting is rarely applied to microalgae due to the drying step, which is indispensable for this type of biomass, this process can be a good option, especially when other biomasses are available on-site; and, when biochemical characteristics of the MB are not suitable for wet biomass conversion routes, such as anaerobic digestion. It should be noticed that the drying step of the MB is not exclusive to the briquetting, as it is required in other dry routes, such as lipid extraction for biodiesel production. Briquettes made with mixtures of MB and forest biomass were evaluated by Costa et al. (2017), who found less moisture absorption by the briquettes, reducing their costs and improving their energy efficiency. Briquettes are used for heating, cooling, and cooking, in domestic boilers, cooling devices, and ovens. Moreover, they can be used for obtaining energy, in combined heat and power (CHP) systems, gasification, co-combustion, and direct combustion equipment. They can also be used in diesel production, replacing fossil fuels (Hu et al., 2014; Saba et al., 2020; Wang et al., 2017).

Despite MB's potential for energy purposes (Rajesh Banu et al., 2020), such as its transformation into solid biofuel, some challenges still need to be overcome, mainly concerning the biomass separation stage (Carneiro et al., 2017; Enamala et al., 2018). The density of MB

is very close to that of water (Xiao et al., 2020), requiring physical and/or chemical processes for a more efficient harvesting (Soares et al., 2019), and increasing the environmental impact of the route, when suspended reactors are used, such as high-rate algal pond (HRAP). In this context, biofilm reactors (BRs) for MB growth may be an interesting option, as they provide the attached growth of microalgae on a support material, separated from the liquid (Wang et al., 2018). BR can be used in association with a HRAP, forming a hybrid reactor. Literature shows that this type of cultivation increases MB productivity, in addition to simplifying the harvesting step, due to the higher concentration of microalgae (Assis et al., 2020; Li et al., 2019; Vo et al., 2019). Therefore, the amount of energy, the negative environmental impacts, and the costs associated with microalgae biotechnology may be minimized (Dasan et al., 2019). Another very relevant aspect when it comes to MB dry energy conversion routes is the drying step. This process consumes a large amount of electricity, and may even make microalgae biofuel unfeasible, in some cases (Soares et al., 2019). Therefore, the search for techniques that can reduce the humidity content of the MB using natural methods, with less or no energy demand is of great importance (Agbede et al., 2020).

When combining the reduction of impacts with the integration between WWTPs (Hao et al., 2019) and renewable energy (Chandra et al., 2019), life cycle assessment (LCA) can be an important tool in the quantification and identification of the most critical steps. Among microalgae context, LCA studies were carried out for microalgae cultivation reactors and their various forms of energy recovery, such as bioethanol (Hossain et al., 2019), biogas (Arashiro et al., 2018; Colzi Lopes et al., 2018; Xiao et al., 2020), biodiesel, and biobutanol (Wu et al., 2019, 2018). These researches showed the environmental impacts associated with MB, but also the benefits brought by microalgae, such as CO₂ sequestration (Porcelli et al., 2020; Yadav et al., 2020). In a comparative LCA between wastewater treatment systems, Arashiro et al. (2018) reported that an application of a HRAP, replacing the conventional activated sludge system, can increase the environmental performance of the WWTP, especially when associated with the MB utilization route. Microalgae-based wastewater treatment technologies allow the recovery of nutrients from the effluent in the biomass, enhancing WWTP economic and environmental sustainability. Regarding briquettes, the impact on the environment was also evaluated for briquettes from forest and agricultural biomass (Ji et al., 2018; Rodzkin et al., 2017). Briquettes were reported to be more environmentally friendly than coal (Wang et al., 2017) and favored GHG emissions reduction when used to replace fossil fuels (Alanya-Rosenbaum and Bergman, 2019), besides decreasing the occurrence of burns in the field (Saba

et al., 2020). However, there is a lack of LCA research addressing the environmental impact of wastewater grown MB solid biofuel, and evaluating the categories of most current worrying impacts, such as climate change and fossil scarcity.

Therefore, the objectives of this study are to use the LCA to (i) identify the potential environmental impacts of the briquettes, varying the type of MB growth reactor and comparing HRAP and hybrid reactor, (ii) assess how much the MB drying step for briquettes obtaining affects the life cycle and the environment, and (iii) proposing improvements to mitigate the potential environmental impacts of this energy route. The innovation of the study comprises the LCA modeling of briquettes for energy valorization of MB grown during the treatment of domestic wastewater. Experimental data of MB productivity and briquette properties were used. Also, the use of briquette as a solid biofuel for electricity generation was evaluated, reducing operating costs and mitigating the environmental impacts that occurred in its life cycle by avoiding the use of fossil fuels. The use of a hybrid reactor and biomass drying were evaluated as strategies to improve the performance of wastewater grown MB valorization. Therefore, improvements of the briquettes production process from MB, contributing to this type of bioenergy development, and the path towards sustainability in the WWTPs, are expected contributions.

4.2. Material and methods

For the modeling of LCAs, foreground or primary data (data not present in the LCA available databases), such as data on wastewater treatment using microalgae, involving growth, harvesting, drying and, the valorization of this biomass by briquetting, as well as the demand for electricity in these stages, were collected in the literature. Background or secondary data, with environmental impacts and emissions, called known inputs from the technosphere, such as electricity, heat, materials, fuels, and avoided products, came from the Ecoinvent version 3.5, allocation at the point of substitution - system (APOS - S) database (Ecoinvent, 2018). These data, foreground, and background, were applied to the modeling of LCAs, using the System for Integrated Environmental Assessment of Products (SimaPro), version 9 software (PRÉ, 2019), to obtain the environmental performance of MB briquettes.

The applied methodology consisted of modeling two LCA scenarios for the MB briquettes production, cultivated during the treatment of domestic wastewater. The modeling followed the guidelines defined by the International Organization for Standardization (ISO) described in ISO 14040/2006 - Environmental management - Life cycle assessment - Principles and framework,

and ISO 14044/2006 - Environmental management - Life cycle assessment - Requirements and guidelines (ISO, 2006a, 2006b). Four stages of the LCA were developed: goal and scope definition; life cycle inventory (LCI); life cycle impact assessment (LCIA); and results interpretation (Guinée et al., 2002; ISO, 2006a, 2006b). Some examples of the application of these four stages proposed by the ISO are: Schneider et al. (2018), Souza et al. (2019), Castro et al. (2020a) and Ferreira et al. (2020), all studies of microalgae; Shimako et al. (2016), Carneiro et al. (2017), Arashiro et al. (2018), Colzi Lopes et al. (2018), Sun et al. (2019), Wu et al. (2019) and Mediboyina et al. (2020), studies of microalgae biofuels; and Wang et al. (2017), Alanya-Rosenbaum and Bergman (2019) and Saba et al. (2020) of other types of biomass briquettes.

4.2.1. Goal and scope definition

The steps selected for modeling the life cycle impacts of the briquettes (the growth, harvesting, drying, and briquetting of MB) were previously studied by the research group that the authors are part of (Federal University of Viçosa, Brazil). Thus, the results and procedures detailed in Assis et al. (2017) and Costa et al. (2017) were used as a basis for modeling and can be consulted in the original studies. In addition, other references were used to complete the parameters necessary for modeling, as explained in Section 4.2.2. This study focused on describing methodological aspects of the LCA.

The objective of the LCA was to quantify the potential environmental impacts of the briquettes, as a valorization route of a resource produced during the treatment of wastewater with microalgae, coming from different growth reactors. Therefore, 2 scenarios were compared:

- Scenario 1 – HRAP MB briquette. MB was grown in a HRAP with CO₂ supplementation and harvested by gravitational settling and dried in an electric oven.
- Scenario 2 – Hybrid reactor MB briquette. MB was grown in a hybrid reactor (composed of a HRAP without CO₂ and with a BR). The MB used for the briquettes production was harvested from the BR's adhered growth panel and by gravitational settling. The BR material was cotton woven. In this scenario, the MB drying was also done in an electric oven.

In the two modeled scenarios, the briquetting was carried out with a proportion of 50% of MB and 50% of forest biomass (Costa et al., 2017), and the briquette drying was carried out without electricity. The environmental performances of the scenario using the HRAP (Figure 4.1a) and

the hybrid reactor (Figure 4.1b) were compared. Figure 4.1 shows the system boundary, inputs, outputs, and avoided products, for the two modeled scenarios.

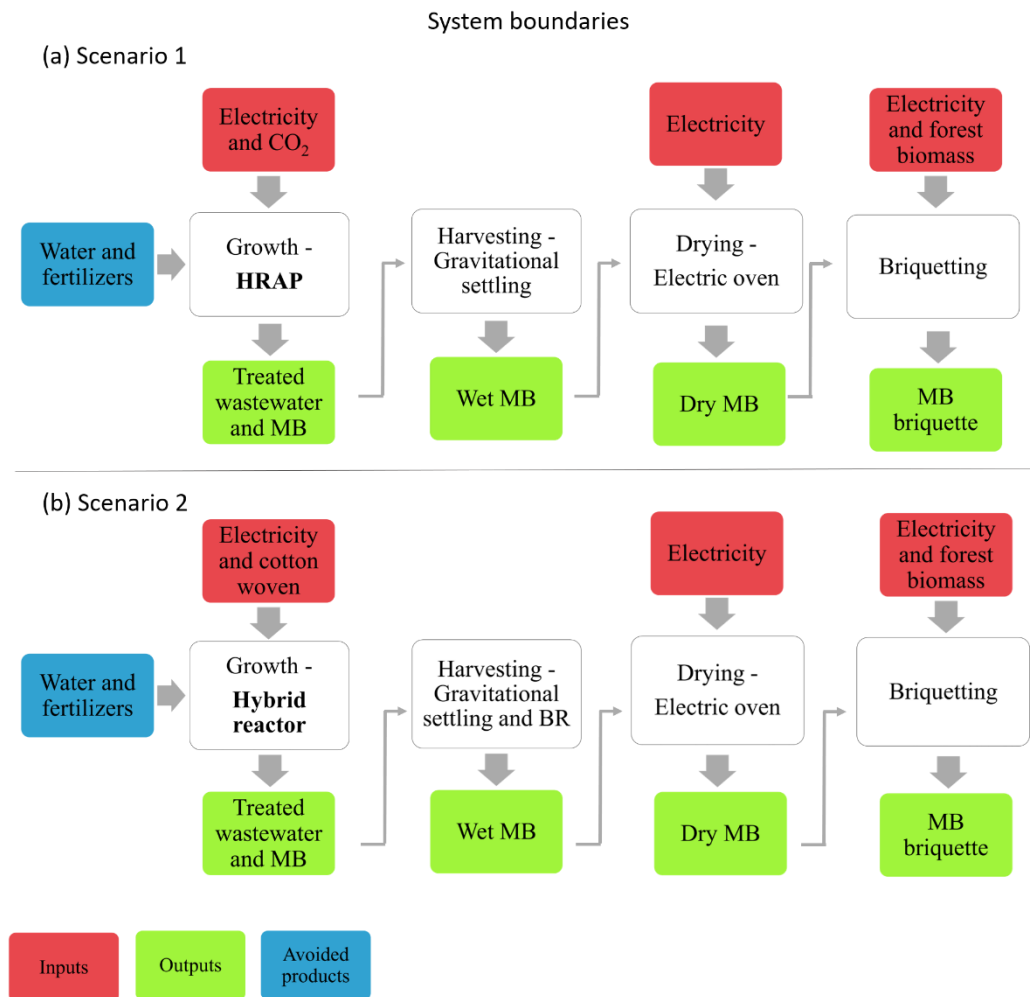


Figure 4.1. System boundaries (a) scenario 1 – HRAP MB briquette; (b) scenario 2 – Hybrid reactor MB briquette.

The cradle-to-gate boundary modeling was applied to the growth, harvesting, drying, and briquetting of MB, obtained during the wastewater treatment. Inputs and outputs of chemical and energy resources were included inside the system frontier. On the other hand, the impacts of capital goods, the end of the useful life of infrastructure and equipment, and the briquettes use after production were not included. According to ISO 14040 and 14044, disregarding these phases is acceptable, as the system boundary remained compatible with the objective of the study (ISO, 2006b, 2006a).

The functional unit (FU) was 1 MJ of gross energy obtained from the briquettes. Similar FU was also used in other studies of MB biofuels (Carneiro et al., 2017; Mediboyina et al., 2020; Shimako et al., 2016; Sun et al., 2019) and forest and agricultural biomass briquettes (Alanya-

Rosenbaum and Bergman, 2019; Saba et al., 2020). The high heating value (HHV) of the briquettes, with 50% MB, was 16.95 MJ/kg (dry basis) (Costa et al., 2017).

During the treatment of wastewater with microalgae, treated sewage and MB are obtained, which can be seen as co-products, each having responsibility for the environmental impacts that have occurred. However, to avoid the allocation of these impacts, the system's frontier expansion was carried out, as well as applied by Arashiro et al. (2018) and Colzi Lopes et al. (2018). With this procedure, provided by the ISO 14040 standard (ISO, 2006b), the multifunctional system is treated as monofunctional. Briquetting was pointed out as the objective of the process so that the treatment of wastewater was approached as a way of reducing the environmental impact of the briquettes. Without using wastewater, tap water and fertilizers containing nitrogen (N) and phosphorus (P) would be needed to supply microalgae growth in synthetic media and obtain MB (raw material for briquettes). So, these were taken as avoided products, as proposed by Castro et al. (2020a) and Souza et al. (2019), since water and nutrients are present in wastewater.

4.2.2. Life cycle inventory (LCI), assumptions and limitations of the study

The stages of growth, harvesting, drying, and briquetting were modeled in SimaPro v9, with the reference flow of 0.06 kg of biomass, with 0.03 kg of MB, enough to obtain 1 MJ of energy gross with briquettes, the analyzed FU.

Growth: The operating time of the cultivation reactors (HRAP with CO₂ and hybrid reactor, formed by a HRAP without CO₂ and a BR) was fixed at 5 days, as being the hydraulic retention time used by Assis et al. (2017). Thus, the productivity of microalgae in that period, in g/m² of HRAP area, was obtained in Assis et al. (2017) and converted to g/m³ of wastewater, since HRAPs usually have a useful depth of around 0.30 m (Albarelli et al., 2018b; Assis et al., 2017; Park and Craggs, 2010). It should be observed that, in the case of the hybrid reactor, the attached growth panel has additional productivity, in g/m² of the panel, with the recirculation of the sewage from the HRAP. The amount of CO₂ required for carbon supplementation of microalgae in the HRAP, in kg CO₂/kg microalgae, was found in Albarelli et al. (2018a, 2018b). The electrical energy consumed by the paddlewheels, which operated for 24 hours during the 5 days, came from Albarelli et al. (2018a, 2018b) and Mian et al. (2015), in kW/ha of HRAP area. For the CO₂ supplementation system, which operates for 10 hours for 5 days, the amount of energy consumption was obtained in Mian et al. (2015). The electricity required for the recirculation of HRAP wastewater in the hybrid reactor panel was not found in the literature and, therefore,

it was assumed that this procedure consumes the same amount of electricity as the CO₂ supplementation system, in kW/ha, with 10-hour activity during the 5 days (Assis et al., 2017). Another input, required only in scenario 2, was the material of the support panel for the attached growth of microalgae, in the hybrid reactor. This material was considered for the LCI, as it has low durability (Assis et al., 2019), unlike the other equipment used in the modeled processes. The weight of this material was estimated through the weight of the 100% cotton fabric, in g/m². The amount of material added in the LCI was proportional to the 5-day operation since the same cotton material could continue to be used for about 40 days (Assis et al., 2019), a period that more wastewater would be treated and more MB would be produced.

Harvesting: The humidity of the MB, harvested with gravitational settling and from the BR attached to the hybrid reactor, was considered equal to 93%, an average value found by Ferreira et al. (2020) for both types of harvesting. Harvesting efficiency of the hybrid reactor panel was 100% (Assis et al., 2017) and 60% of the settler (Park and Craggs, 2010). It is important to notice that the modeled harvesting methods have not been inputted chemical products or electricity.

Drying: Electric power, for the operation of the drying oven, in kWh/kg of evaporated water, was obtained in Xu et al. (2011). The density of MB was considered to be very close to that of water (Xiao et al., 2020) and, so, it was estimated the amount of water evaporated during drying and consequently, the total of energy consumed in the process. The MB was dried until it varied from initial humidity of 93% (Ferreira et al., 2020) to 12%, enough for briquetting (Costa et al., 2017).

Briquetting: The electricity demand, for the operation of the briquetter, in kWh/ton of biomass, was originated from Hu et al. (2014). Forest biomass, also used in briquetting, was not considered in LCI calculations, as it is a pruning residue.

Returning to what was discussed at the end of Section 4.2.1, tap water and fertilizers that would be needed to grow microalgae in synthetic medium, and that were saved due to the use of domestic wastewater as a growth medium, had avoided quantities equal to the wastewater composition. The volume of wastewater, in m³, was calculated through the microalgae production required to obtain 0.03 kg of MB and the nutrients were calculated using the characterization of the wastewater, in mg/L, performed by Assis et al. (2017). The treated wastewater was not considered an emission to water, as it meets the Brazilian guideline for the discharge of effluents into water bodies (COPAM, 2008). Furthermore, evaluating the

efficiency of pollutant removal by the HRAP and the hybrid reactor was not the objective of this study, since this discussion was presented in Assis et al. (2017). The present study focused on the assessment of the environmental performance of MB briquettes, which could be obtained from the two mentioned growth reactors. Thus, the emissions to the environment were mostly coming from the process systems from the APOS - S library of Ecoinvent v3.5, used to compose the known inputs from the technosphere of the LCI of scenarios 1 and 2. These processes are briefly listed and described in Table 4.1.

Table 4.1. Process systems and input data for LCIs of the modeled scenarios.

Known inputs from the technosphere	Process description (Ecoinvent v3.5)	
Electricity		
Electricity – Electricity, high voltage {DE} production mix	Paddlewheels, 24h for 5 days: 2.7 kW/ha ^(a, d) ; CO ₂ supplementation: 1.5 kW/ha ^(d) ; Recirculation of wastewater in the BR panel: 1.5 kW/ha; Drying the MB in an oven: 0.92 kWh/kg evaporated water ^(e) ; Briquetting: 70 kWh/ton of biomass ^(c) .	Production, transmission, and distribution of network electricity, meeting the demands for high, medium, and low voltage.
Materials		
Cotton - Textile, woven cotton {GLO} production	100% cotton fabric of the attached growth panel: 160 g/m ² (40-day durability) ^(b) .	Manufacture of cotton fabric in a specialized industry.
CO ₂ - Carbon dioxide, liquid {RoW} production	CO ₂ supplementation: 3.30 kg CO ₂ /kg microalgae ^(a) .	Production of liquid carbon dioxide from waste gases from different production processes.
Avoided products		
N - Nitrogen fertilizer, as N {RoW} urea ammonium nitrate production	Equivalent to N present in wastewater: 37.30 mg/L ^(b) .	Production of ammonium nitrate, urea, from ammonia and nitric acid.
P - Phosphate fertilizer, as P ₂ O ₅ {RoW} single superphosphate production	Equivalent to P present in wastewater: 5.20 mg/L ^(b) .	Production of simple superphosphate from sulfuric acid and rock phosphate.
Water - Tap water {RoW} tap water production, conventional treatment	Equivalent to the amount of wastewater to produce MB with a productivity of 104 g/m ³ for the HRAP and 97 g/m ³ for the hybrid reactor ^(b) .	Tap water under pressure at the gate of the water treatment plant (WTP), ready for distribution in the network.

References in parentheses: ^aAlbarelli et al. (2018a, 2018b); ^bAssis et al. (2017); ^cHu et al. (2014); ^dMian et al. (2015); ^eXu et al. (2011).

The code {DE}, mentioned in the name of the Ecoinvent v3.5 process, indicates that the inventory was built taking into account average values of systems in Germany; {GLO} represents the activities with an average valid for all countries in the world; and {RoW} means that inventory took into account the average data of the “Rest-of-the-World”, reflecting global data, but Europe is excluded (Ecoinvent, 2018). Table 4.1 shows that only one of the inputs had global data (cotton). The others were modeled with “RoW”, which is an alternative very similar to global data, since it is linked to geographic activities, adjusting to uncertainties. In the case of electricity, the German mix was used, which is based on data from the International Energy

Agency (IEA) (Ecoinvent, 2018). This adaptation was necessary since there is no global electricity mix in the database. However, the chosen mix has about 51% of non-renewable sources, being very similar to the world reality (IEA, 2019b), so that the environmental impacts found in this research had a global characteristic. With this in mind, Brazil's mix was not used, as its composition is very different from that found worldwide (IEA, 2019b).

4.2.3. Life cycle impact assessment (LCIA)

In the LCIA stage, to understand the potential environmental impacts of the modeled scenarios, the classification, characterization, and normalization of the results of the LCIs were carried out in SimaPro v9. With the classification and characterization, the elements of the LCI were assigned to the impact categories, in the form of equivalent emissions. Posteriorly, normalization was applied so that impact categories can be compared with each other. Thus, the impact categories were expressed on the same scale through Ecopoints, according to the reference value of the selected LCIA method (Guinée et al., 2002; ISO, 2006a, 2006b; PRé, 2019, 2016). The ReCiPe 2016 version 1.12 method was used, which has 18 impact categories at the midpoint, hierarchical, and uses global impact mechanisms (ReCiPe Midpoint H v1.12 / World) (RIVM, 2017). The potential environmental impacts were assessed using the following categories: climate change, stratospheric ozone depletion, particulate matter formation, terrestrial acidification, freshwater eutrophication, marine eutrophication, terrestrial ecotoxicity, mineral resource scarcity, fossil resource scarcity, and water consumption (RIVM, 2017). These categories were selected due to their environmental relevance, their relationship with the subject of this study, and their use in other studies, such as Schneider et al. (2018) from the microalgae context; Arashiro et al. (2018), and Wu et al. (2019) that modeled MB biofuels; and Saba et al. (2020) and Wang et al. (2017) that studied LCA from biomass briquettes.

4.2.4. Sensitivity analysis

Sensitivity analysis was performed to assess how much the results and conclusions of the LCAs were affected by the methods selected for the modeled process steps (PRé, 2016). For this, scenarios were modified, and the model was recalculated for the following situations:

- a) The humidity of the MB, after the concentration stage, was varied to evaluate its effect on the energy demand of the drying process.

b) Coal briquette was considered an avoided product, as the possibility of MB briquettes being used for electricity generation in a CHP system was evaluated, replacing coal briquettes.

As a result, strategies were identified to improve the modeled scenarios and to reduce the environmental impacts of MB briquettes.

4.3. Results and discussion

The LCI of scenarios 1 and 2, modeled using the reference flow of 0.06 kg of biomass, containing 0.03 kg of MB, necessary to comply with the adopted FU, is shown in Table 4.2.

Table 4.2. LCI of scenarios 1 – HRAP MB briquette and 2 – Hybrid reactor MB briquette, to reach the FU of 1 MJ of gross energy production.

Stage			Value		Unity	
			Scenario 1	Scenario 2		
Growth	Input	Municipal wastewater	0.470	0.270	m ³	
		CO ₂	0.162	-	kg	
		Cotton	-	0.0005	kg	
		Electricity	0.063	0.036	kWh	
	Output	Treated wastewater and wet MB	0.470	0.270	m ³	
		Avoided products	Tap water	470.0	270.0	kg
			N	0.018	0.010	kg
			P	0.002	0.001	kg
Harvesting	Input	Treated wastewater and wet MB	0.470	0.270	m ³	
	Output	Wet MB	0.421	0.421	kg	
Drying	Input	Wet MB	0.421	0.421	kg	
		Electricity	0.357	0.357	kWh	
	Output	Dry MB	0.030	0.030	kg	
Briquetting	Input	Dry MB	0.030	0.030	kg	
		Forest biomass	0.030	0.030	kg	
		Electricity	0.004	0.004	kWh	
	Output	Briquette	0.060	0.060	kg	

4.3.1. Classification and characterization

The elements of the LCI were assigned to the impact categories through classification and each result was multiplied by characterization factors, determined by the ReCiPe Midpoint H v1.12 / World method, composing the LCIA.

4.3.1.1. Comparison of scenarios by modeled stage

Figure 4.2 presents the results of the classification and characterization and the total potential environmental impacts of LCIA in scenarios 1 and 2, in each category. They were obtained from the sum of emissions of growth, drying, and briquetting stages for scenarios 1 and 2. Thus, although there were positive and negative impacts in all categories, they were discounted and resulted in an environmental benefit or harm. The impacts, through the equivalent emissions, are written in the vertical bars and indicate the maximum value found, which represents 100% (when the environment was harmed) or -100% (when the environment was favored) of the impact in that category. Values used for making Figure 4.2, are found in full in Supplementary Tables S1 and S2 (Appendix A).

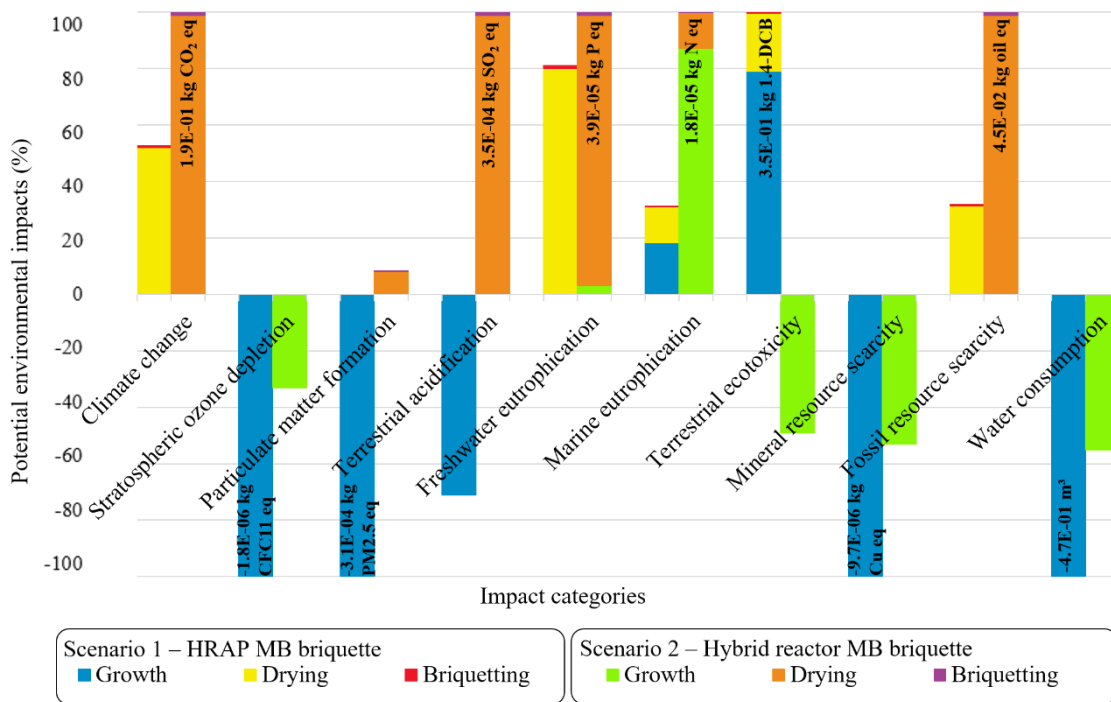


Figure 4.2. Potential environmental impacts of scenarios 1 – HRAP MB briquette and 2 – Hybrid reactor MB briquette, at each stage of the life cycle and percentage comparison between scenarios.

The harmful environmental impacts, shown in Figure 4.2, occurred, in both scenarios, in the categories of climate change, freshwater eutrophication, marine eutrophication, and fossil resources scarcity. Scenario 1 had a better environmental performance in almost all impact categories when compared to scenario 2, except for terrestrial ecotoxicity, in which only scenario 1 had an adverse environmental impact. Scenario 2, on the other hand, caused negative

impacts in the particulate matter formation and terrestrial acidification, categories in which scenario 1 benefited the environment.

The categories of stratospheric ozone depletion, mineral resource scarcity, and water consumption were favored in both scenarios. This occurred due to the microalgae growth stage, using wastewater, and avoiding tap water and fertilizers, and being sufficient to reduce the impacts of other stages of the life cycle. However, this stage has caused environmental damage, due to the required inputs, in marine eutrophication in scenarios 1 and 2 and terrestrial ecotoxicity in scenario 1. Schneider et al. (2018) compared the use of wastewater in the growth of microalgae with the use of water and nutrient solution (NPK), where the wastewater scenario was less impactful in 17 of the 18 impact categories. The use of wastewater as a culture medium for microalgae growth also acts in environmental compensation by promoting its treatment and avoiding the use of fertilizers. Raghuvanshi et al. (2018) also compared the growth of microalgae in clean water and wastewater, finding fewer environmental impacts in the stages from growth to the valuation of MB in biodiesel when wastewater was used. Thus, it is understood that, without the association with wastewater treatment, the briquettes would have detrimental environmental performance in these categories, as well. Also, the impacts would be greater in these and other categories that have already suffered.

The harvesting stage did not participate in the impact categories and therefore does not appear in Figure 4.2, since the modeled method, gravitational settlement of microalgae, did not require chemical inputs or electricity. Schneider et al. (2018) pointed out the importance of this step to maximize environmental gains. Despite their efficiency, coagulation-flocculation techniques, with the use of chemical products, contribute to the increase in environmental impacts. According to Jorquera et al. (2010) and Show et al. (2013), the coagulation-flocculation process helps in the separation of microalgae, increasing the efficiency of gravitational sedimentation 30 to 50x and reducing energy consumption in drying. In the present study, there were no environmental impacts from the harvesting stage, but the efficiency of suspended microalgae separation was reduced.

About the most impacting stages, drying contributed to the greatest adverse environmental impacts in several categories in both scenarios. In scenario 1, there were climate change, freshwater eutrophication, and fossil resource scarcity, corresponding to 99% of 1.0E-01 kg CO₂ eq, 3.1E-05 kg P eq, and 1.4E-02 kg oil eq, in each category, respectively. In scenario 2, emissions were 1.9E-01 kg CO₂ eq in climate change, 3.9E-05 kg P eq in freshwater

eutrophication, and $4.5E-02$ kg oil eq contributing to the fossil resources scarcity, where drying also represented 99 % of these impacts. Still, in scenario 2, the drying stage reflected 99% of $3.5E-04$ kg SO_2 eq in terrestrial acidification and 96% of $2.6E-05$ kg $PM_{2.5}$ (particles smaller than 2.5 mm) eq in the particulate matter formation. In the four categories, in which drying was responsible for 99% of emissions, the briquetting emitted the remaining 1%. In particulate matter formation, in scenario 2, briquetting represented an additional 1% and growth, 3%. It is interesting to note that, although the same amount of MB was dried in the two scenarios, the resulting emissions were different. This fact can be justified by the resulting environmental impact in each of the categories that consider the emissions that occurred and those that were avoided in all stages of the modeled life cycle, with the growth stage responsible for these divergences. In the LCA carried out by Sun et al. (2019), to obtain 1 MJ using different biofuels from MB, the drying stage had high impacts on climate change, due to biomass drying, explained by its high consumption of electricity. Thus, Schneider et al. (2018) pointed out the importance of naturally reducing the amount of water in which the microalgae were diluted to reduce the consumption of electricity during drying. Therefore, in this study, as a sensitivity analysis (discussed in Section 4.3.3), the variation of MB humidity was evaluated by partial drying in the sun inside a greenhouse.

In the modeled scenarios, the briquetting stage represented low electricity consumption and fewer environmental impacts compared to the other stages, i.e., growth and drying. In other LCAs, such as biomass briquettes from corn stalks, carried out by Hu et al. (2014), the growth of biomass did not require inputs or electricity and drying was not necessary, with briquetting being the step that most demanded electricity. In this way, the briquetting also needs to increase its energy efficiency and, thus, improve the environmental performance of the briquettes, since it is a step that cannot be avoided in the life cycle of the briquettes. Besides, briquetting needs to consume less electricity to improve the energy balance of biomass briquettes with low energy yield, such as corn straw, used by Ji et al. (2018).

One stage that was not included in the modeled life cycles, but is found in the LCAs of biomass briquettes, is the briquette use. Saba et al. (2020) modeled, with cradle-to-grave boundary, using FU of 1 MJ, the entire briquette production chain, from the obtaining of raw material until the use of pruning waste briquettes, including transportation, emissions from briquettes burning, and alternatives for the disposal of the materials used in the packaging of briquettes. The use of the briquettes contributed to emissions in climate change, although the resulting contribution was beneficial to the environment. Also, when compared to diesel, to obtain thermal energy,

the briquettes showed better environmental performance in 10 of the 15 evaluated impact categories, mainly in climate change and scarcity of non-renewable resources.

4.3.1.2. Comparison of scenarios by known entry into the technosphere

Figure 4.3 shows the potential environmental impacts resulting from the LCIA in scenarios 1 and 2, in each of the evaluated categories. Impacts were associated with required known inputs from the technosphere (electricity, water, CO₂, cotton and fertilizers, nitrogen, and phosphate). Data used for Figure 4.3 construction are detailed in Tables S1 and S2 (Appendix A).

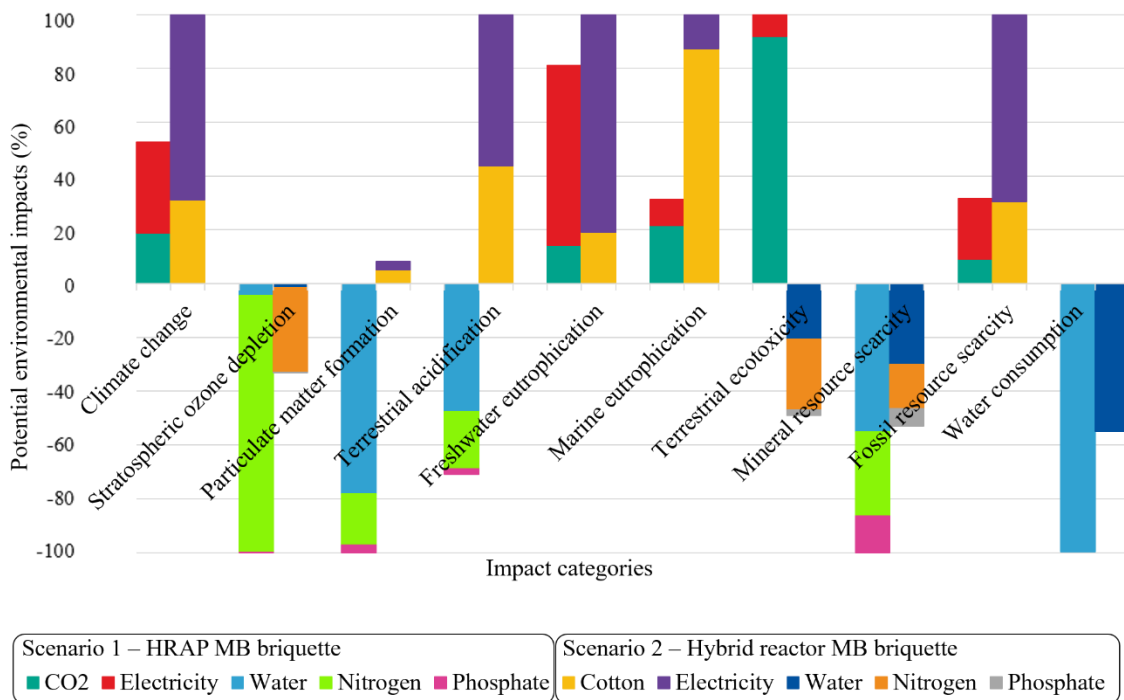


Figure 4.3. Percentage comparison of the potential environmental impacts of scenarios 1 – HRAP MB briquette and 2 – Hybrid reactor MB briquette, associated with known inputs from the technosphere.

In both scenarios, emissions associated with energy consumption stood out, representing, on average, 60% of the total environmental impacts. Most of the impacts were reflected in climate change, terrestrial acidification, freshwater eutrophication, and fossil resource scarcity categories. German electricity mix (Electricity, high voltage {DE} | production mix | APOS – S), used in LCA, contributed, in scenarios 1 and 2, respectively, to 2.8E-01 and 2.6E-01 kg CO₂ eq, 6.3E-04 and 5.9E-04 kg SO₂ eq, 4.4E-05 and 4.1E-05 kg P eq and 6.7E-02 and 6.3E-02 kg oil eq. Taking into account the impacts caused by electricity and the fact that drying demanded

more from this input, it is understandable why this stage was the most impactful in both scenarios.

According to the mix of electricity, it can be noticed that emissions in scenario 1 were higher than those in scenario 2. This result is justified, since scenario 1 demanded more electricity, in the growth stage, in comparison to scenario 2, as shown in the LCI of the scenarios (Table 4.2). However, it is evident that, when comparing the two scenarios all categories (except for terrestrial ecotoxicity), scenario 1 presented better environmental performance. By avoiding more water, N, and P, the environmental impacts were reduced in scenario 1. The database representing water and the inventories of the N and P processes (see Table 4.1), from the APOS - S library by Ecoinvent v3.5, includes, in addition to obtaining products, the transport of raw materials, intermediate products, and fertilizer from the factory to the local warehouse (Althaus et al., 2007). Therefore, in addition to water, N, and P avoided in scenario 1 compared to scenario 2, several chemicals, electricity, and fuel were considered, contributing to improving the environmental performance of the first scenario.

Technically, considering the development of microalgae technologies, this result can be treated as a limitation of the study. In the case of avoided water, for example, there is the water consumption impact category, expressed in terms of m^3 of tap water, which accounts for the consumed or avoided volume. In this category, scenario 1 showed a greater balance, benefiting the environment, as 0.470 m^3 of wastewater was used in the growth of microalgae, compared to 0.270 m^3 in scenario 2. However, it is justified that, in scenario 2, less volume of water was avoided because less wastewater was used to produce the same amount of MB. The hybrid reactor, modeled in scenario 2, has higher MB productivity and includes the BR panel with a harvesting efficiency of 100%. In other words, scenario 2 needed less sewage because it is more productive and has an additional harvesting mechanism (in addition to the settler, present in both scenarios). Therefore, if, in scenario 2, 0.470 m^3 of wastewater were admitted, more MB would be produced, surpassing the studied FU. However, it is important to highlight the non-agreement of this result in a real scale operation of these reactors. In a microalgae-based WWTP, ways are sought to increase biomass productivity and harvesting efficiency, besides, to treat a greater volume of wastewater per unit of reactor area, benefits achieved when using the hybrid reactor.

The fact that the category water consumption is expressed in m^3 of water and the modeled system avoids water, facilitated this observation, which also can be extended to the case of

avoided fertilizers. Again, scenario 1 avoided more fertilizers than scenario 2, due to the higher amount of required wastewater. However, it is noteworthy that this result does not represent a benefit of the technology, since larger amounts of sewage could be treated on a real scale when using the hybrid reactor. Hypothetically, if water and fertilizers were used to produce a synthetic growth medium for microalgae growth, the situation would be reversed. Therefore, scenario 1, with greater demand for both, would have worse environmental performance.

The importance of avoiding the consumption of tap water is emphasized, and the considerations of the water consumption category are correct and consistent, due to the lack of this resource with desirable quality and/or quantity for countless people in the world (UN-Water, n.d.). The relevance of microalgae for the wastewater treatment is also evident, by avoiding contamination of water resources, especially in low-income and developing countries, in which the universalization of sanitation is still a goal to be achieved (UN-Water, n.d.). In this context, WWTPs using microalgae may contribute to avoid chemical inputs and recover resources (Cai et al., 2013), being a technology of simple and low-cost operation and maintenance.

Not only do the avoided products make up the emissions from the modeled scenarios, but there were also the emissions from the other known inputs of the technosphere, CO₂ in scenario 1 and cotton in scenario 2. The database of CO₂ product system used in the modeling (Carbon dioxide, liquid {RoW} | production | APOS – S), represents its production from waste gases of different production processes. Despite being considered a residual gas from other production processes, such as ammonia and hydrogen production, and obtained free of environmental charges (Althaus et al., 2007; Ecoinvent, 2018), it was nevertheless a very impacting input in the modeling. In scenario 1, CO₂ used to supplement microalgae growth and to control the pH, contributed with 1.6E-01 kg CO₂ eq in climate change, 9.3E-06 kg P eq in freshwater eutrophication, and 2.7E-02 kg oil eq in fossil resource scarcity. This input was also primarily responsible for the impacts on marine eutrophication and terrestrial ecotoxicity, for releasing 6.1E-06 kg N eq and 9.8E-01 kg 1,4-dichlorobenzene (1,4-DCB), respectively, representing around 68% and 92% of emissions in these categories. With this, it is possible to explain why the impacts in these 2 categories were caused during the growth stage, as shown in Figure 4.2, and not by drying, as in the other categories. Also, it is understood why terrestrial ecotoxicity was more impacted in scenario 1, since in scenario 2 there was no CO₂ supplementation. Therefore, to reduce the environmental impacts associated with the use of this input in the growth stage, one possibility is to use exhaust gases from the combustion of gasoline as a CO₂ source, despite the high investment (de Assis et al., 2019). Porcelli et al. (2020) found that the

environmental impacts of using residual CO₂ from biogas refining were lower compared to when synthetic CO₂ is used. In these cases, CO₂ could be considered an avoided emission to the environment. Therefore, using residual CO₂ from one process could reduce the impact in the terrestrial ecotoxicity category and perhaps even benefit the environment in other categories.

In scenario 2, the use of cotton (Textile, woven cotton {GLO} | production | APOS – S), as a support material for microalgae growth, in the hybrid reactor, had significant participation in the environmental impacts. Therefore, an aspect that could improve the performance of this scenario is the use of less impacting material in the hybrid reactor. Assis et al. (2019) found superior productivity and adhesion of microalgae in the material support with cotton woven in comparison to other tested materials, even though this material presented less durability than the others. The life cycle of cotton fabric, available in Ecoinvent v3.5 and used in modeling scenario 2, addresses cotton planting, harvesting by ginning, and processing. In the planting phase, carbon sequestration by cotton cultivation is included, reducing the impacts on climate change in this phase, and its release is included at the end of its useful life. Emissions associated with nitrogen obtention and fertilizer production also contribute to impacts. Still, in planting, the use of water for irrigation of cotton culture is also included, being the main contributor to the impact category of water consumption, in the life cycle of the final product (Cotton Incorporated, 2012; Ecoinvent, 2018). The use of 5.0E-04 kg of woven cotton in scenario 2 (Table 4.2) consumed 1.1E-02 m³ of water, which is one more reason why scenario 2 presented a lower performance in this category when compared to scenario 1. The industry process (cotton processing) includes spinning, thorough preparation, dyeing, and finishing. The impacts of this phase are mainly attributed to the use of energy. Along with the energy consumption of the processing, there is also the use of irrigation and the ginning of cotton (Cotton Incorporated, 2012). Thus, the use of cotton emitted 1.2E-01 kg CO₂ eq in climate change, 2.5E-04 kg PM_{2.5} eq in particulate matter formation, 4.6E-04 kg SO₂ eq in terrestrial acidification, 9.7E-06 kg P eq in freshwater eutrophication and contributed 2.8E-02 kg oil eq to the fossil resource scarcity. Moreover, cotton released 1.7E-05 kg N eq in marine eutrophication, representing 87% of scenario 2 impacts in this category. Thus, the cotton used during the microalgae growth stands out as being responsible for raising the environmental impacts in scenario 2. In some categories, such as those mentioned above, their emissions have outweighed the benefits of using wastewater for microalgae growth. However, there is a possibility, which is not considered in the inventory of the cotton fabric or in the life cycle modeled in this paper, which is the use of

the cotton bark, residue from the fiber separation process, to compose the briquettes with the MB, benefiting both life cycles.

As stated, the combination of released and avoided emissions resulted in the values shown in Figure 4.2 and the scenario comparison shown in Figures 4.2 and 4.3. In climate change, for example, emissions from CO₂ and electricity, in scenario 1, were higher than cotton and electricity in scenario 2, but the resulting one, taking the benefits of avoided products, made scenario 2 to have a greater environmental impact. Comparing cotton with CO₂, as they are the inputs that differ between the two scenarios, cotton polluted more than CO₂, implying terrestrial acidification, freshwater eutrophication, marine eutrophication, and fossil resource scarcity. In terrestrial ecotoxicity, CO₂ was about 6x more impactful than cotton.

4.3.2. Normalization

Normalization was used to identify the extent to which an impact category had a relatively high or low value, based on the average global pressure applied on the environment by an individual in 2010 (RIVM, 2016; Sleeswijk et al., 2008). As a result, emissions in the impact categories were expressed as relative contributions that the modeled life cycle has on the environment. This procedure was carried out to clarify which of the scenarios had the best environmental potential. Both scenarios have considerably impacting inputs in different categories, and it is important to know which one causes the greatest pressure on the environment. Besides, it was possible to identify the environmental importance of the only impact category that scenario 1 had a worse environmental performance, compared to scenario 2, which was terrestrial ecotoxicity. Figure 4.4a shows the normalized environmental impacts of each scenario in the 10 evaluated impact categories, with the relative comparison between these categories and between the two scenarios. In Figure 4.4b, the four categories in which the environment was most negatively pressured are highlighted. The normalized environmental impact values, used to produce Figure 4.4, can be detailed found in Table S3 (Appendix A).

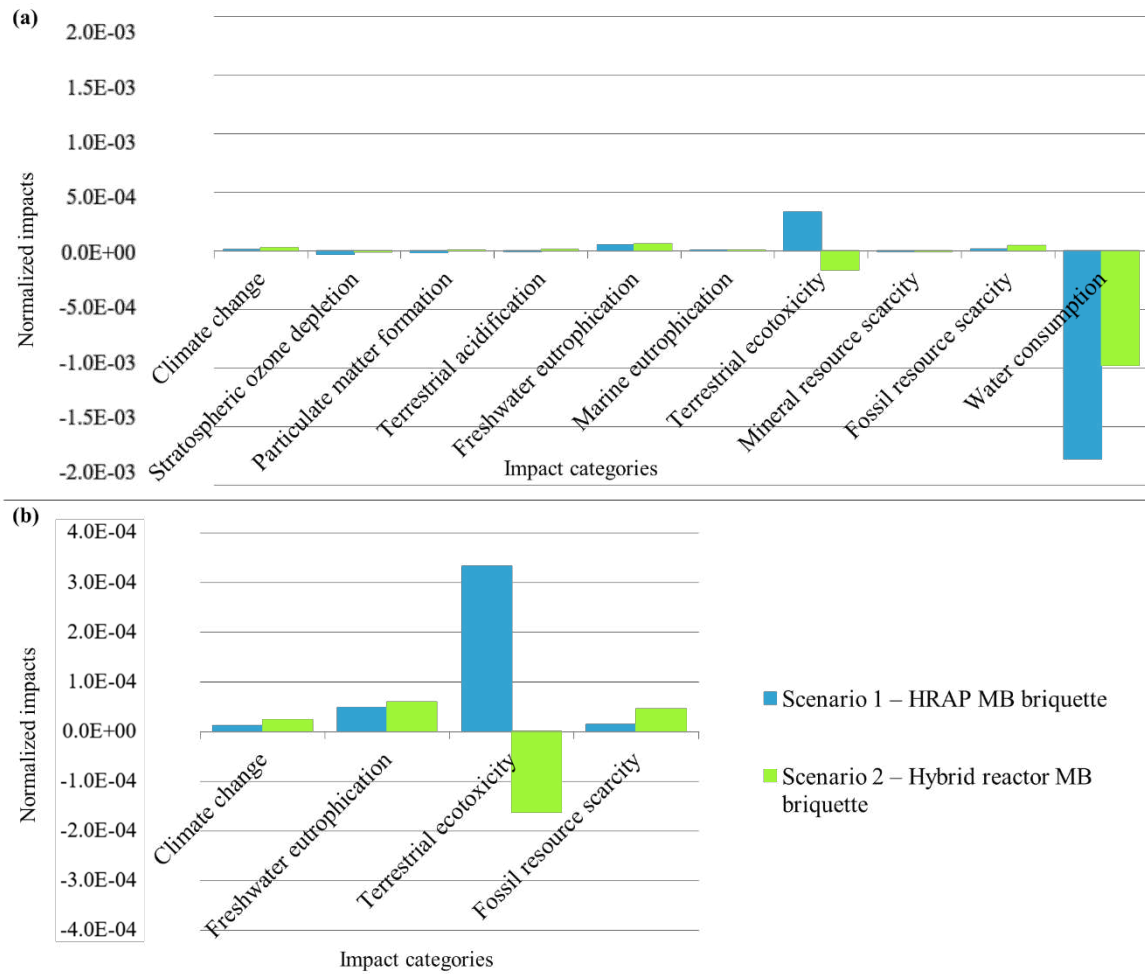


Figure 4.4. Relative comparison between impact categories and between scenarios 1 – HRAP MB briquette and 2 – Hybrid reactor MB briquette (a) in the 10 impact categories and (b) in the 4 categories that most affected the environment.

The most significant impact category was water consumption, as can be seen in Figure 4.4a, in which the two modeled scenarios benefited the environment. This category was already expected to be so representative, due to the importance of saving water, already discussed in Section 4.3.1.2. Furthermore, as reported by Raghuvanshi et al. (2018) and Schneider et al. (2018), and explained in Section 4.3.1.1, the environmental benefits of microalgae and their products are increased when wastewater is used, instead of synthetic growth medium.

Terrestrial ecotoxicity was the cause of great influence in the environment, being negative in the case of scenario 1 and positive in scenario 2. This category presented high relevance due to the impacts caused by the release of toxic substances, such as heavy metals, in terrestrial ecosystems. Thus, scenario 1 harmed the environment much more, due to the emissions resulting from $3.5E-01$ kg 1,4-DCB eq, mainly due to the CO_2 supplementation in the growth

stage, in comparison to the emissions in other categories and scenario 2. Standardization highlighted a very interesting result because, only with the classification and characterization (Figures 4.1 and 4.2), scenario 1 showed the best environmental performance. However, terrestrial ecotoxicity was the one that most caused harmful impacts on the environment, being solely caused by scenario 1.

The environment also suffered pressure in freshwater eutrophication, fossil resource scarcity, and climate change, as shown in Figure 4.4b, being negative in the two modeled scenarios. In these three categories of impacts, emissions occurred mainly in the drying stage (in both scenarios), as they came mostly from the electricity mix, which was very demanding in this part of the modeled life cycle. Although emissions were reduced by saving water and mainly, fertilizers, in the case of freshwater eutrophication, emissions were $3.1\text{E-}05$ kg P eq in scenario 1 and $3.9\text{E-}05$ kg P eq in scenario 2.

Contributing to the fossil resource scarcity, oil extraction corresponded to $1.4\text{E-}02$ kg oil eq in scenario 1 and $4.5\text{E-}02$ kg oil eq in scenario 2 and, in climate change emissions were $1.0\text{E-}01$ kg CO₂ eq in scenario 1 and $1.9\text{E-}01$ kg CO₂ eq in scenario 2. Emissions of CO₂ eq were originated, to a greater extent, from the electricity mix, but also from the CO₂ supplementation in scenario 1 and the cotton in scenario 2, both inputs used in MB's growth stage. The category of climate change, although it was not expressive in the life cycle of the modeled scenarios, is widely mentioned, mainly due to emissions during the phase of briquettes use (not modeled in this study). However, in LCAs that encompassed the use phase, the briquettes from several biomasses presented significantly lower GHG emissions, when compared to fossil fuels (Alanya-Rosenbaum and Bergman, 2019; Hu et al., 2014; Ji et al., 2018).

The category of greatest expression, harming the environment, was terrestrial ecotoxicity, being caused by the CO₂ used in the growth of MB, in scenario 1. Other categories that negatively impacted the environment were mainly influenced by the drying stage due to its high electricity consumption. However, in a relative comparison between the impact categories, $-1.8\text{E-}03$ Ecopoint contributed to the environment in scenario 1, by saving water, versus $3.3\text{E-}04$ Ecopoint damaging the environment in terrestrial ecotoxicity. In scenario 2, the environment was favored by $-9.8\text{E-}04$ Ecopoint in water consumption versus $6.0\text{E-}05$ in freshwater eutrophication. Therefore, it can be observed that the modeled life cycles benefited the environment more than they have harmed it.

4.3.3. Improvements to the life cycle of microalgae briquettes

Figure 4.5 shows the electricity demand in the drying phase and the modeled base life cycles, depending on the humidity of the MB taken for drying. Electricity demand for drying, in the base scenarios 1 and 2, was 0.357 kWh. When including growth, drying, and briquetting stages, demand was 0.423 kWh for scenario 1 and 0.397 for scenario 2 (as shown in Table 4.2). The potential electricity yield that can be obtained with MB briquettes is also shown in Figure 4.5 and is equivalent to 0.196 kWh. This value represents a 70% efficiency in the conversion of the briquette's gross energy, 1 MJ, which is equivalent to 0.280 kWh.

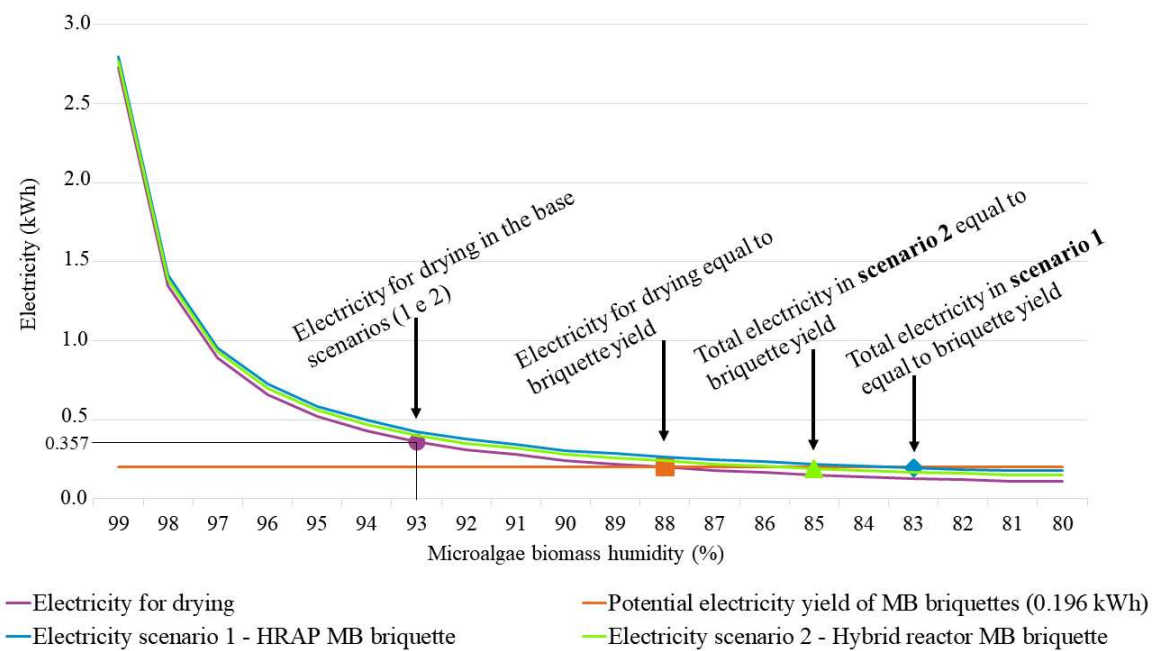


Figure 4.5. The potential electricity yield of MB briquettes and demand for electricity in drying and in the complete life cycles of base scenarios 1 and 2, depending on the humidity of the MB.

In the base scenarios, 1 and 2, 0.357 kWh was used to dry the MB, with the possibility of obtaining 0.196 kWh through the briquettes. Thus, the process showed an unfavorable energy balance, with a negative net energy. By varying the humidity content of the MB, shown in Figure 4.5, it was determined that the humidity, after the harvesting step, should be reduced, by natural means, until it reaches at least 88%, to then be dried in an electric oven. In this way, the potential electricity yield of the briquettes is equal to the energy required for drying. In scenario 1, the humidity should be reduced up to 83% and, up to 85% in scenario 2, to the electric potential of the briquettes be equivalent to the electricity demand of all the processes of the modeled life cycles. This drying was simulated in the sun and inside a greenhouse in order not to consume electricity, although it requires more time, space, and depending on the climate, possible changes in the texture and color of the microalgae (Show et al., 2015).

Subsequently, scenarios 1 and 2 were remodeled, considering that the MB had initial humidity of 83 and 85%, respectively, before being taken to dry in an electric oven. Thus, all the electricity consumed from the MB growth to the briquetting could be supplied by the briquettes. It was considered that the briquettes would be used in CHP generation plants, through co-combustion with coal briquettes, similar to the one suggested by Alanya-Rosenbaum and Bergman (2019) for biomass briquettes and Chew et al. (2017) and Choi et al. (2019) for MB. Considering that the MB briquette can be used as a substitute for coal, the latter was treated as an avoided product, avoiding the extraction of coal, its preparation, and briquetting to be used to generate electricity. Although coal can be used without prior transformation into briquettes, the briquetting may increase the performance of the fuel (Alanya-Rosenbaum and Bergman, 2019; Avelar et al., 2016; Ji et al., 2018; Saba et al., 2020; Wang et al., 2017). Therefore, value of 1 MJ of hard coal briquettes {RoW} | production | APOS – S, from Ecoinvent v3.5 was used, with a HHV equal to 31.4 MJ/kg. This database represents the average global production of hard coal briquettes, with the requirements for electricity and basic materials, as well as emissions during the production process. The extraction of hard coal is modeled for the eight most important mining regions in the world and the coal washing step is analyzed together with the extraction (Dones et al., 2007; Ecoinvent, 2018).

Figure 4.6a shows the normalized environmental impacts of climate change, freshwater eutrophication, terrestrial ecotoxicity, and fossil resource scarcity. Impacts were presented in the form of relative comparison between the categories and between the two remodeled scenarios (scenarios 1a and 2a), considering the natural reduction of MB humidity to 83% in scenario 1a and 85% in scenario 2a. For the comparison between scenarios 1a and 2a, the life cycle of 1 MJ (gross energy) of coal briquettes was also placed. Figure 4.6b shows the normalized environmental impacts and the relative comparison, in the four categories in which the briquettes caused more pressure on the environment, for the two remodeled scenarios (scenarios 1b and 2b). Scenarios 1b and 2b were built taking advantage of the reduction in the humidity of the MB, modeled in scenarios 1a and 2a, adding the impacts avoided when replacing the coal briquettes with those of MB, thus encompassing the two improvements proposed for the life cycle. Normalized environmental impacts, life cycles of scenarios 1a, 1b, 2a, and 2b, and hard coal briquettes data, used in Figure 4.6, can be consulted in Table S3 (Appendix A).

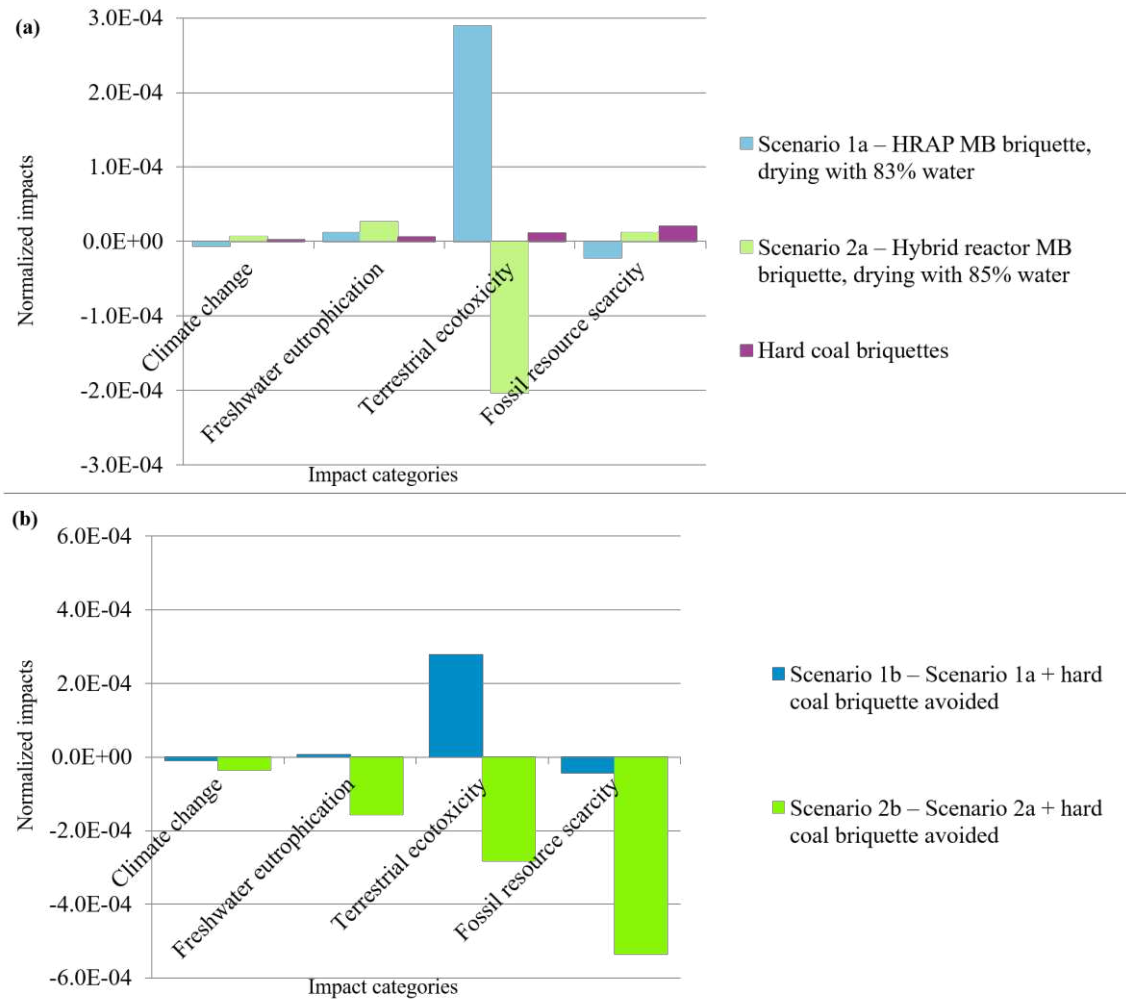


Figure 4.6. Normalized environmental impacts for the sensitivity analyzes and relative comparison between (a) coal briquettes and MB briquettes with natural humidity reduced to 83% - scenario 1a and 85% - scenario 2a; (b) scenarios 1b and 2b, with coal briquettes.

From Figure 4.6a, it is possible to notice that MB briquettes continue to cause more damaging impacts compared to coal. According to Carneiro et al. (2017), much still needs to be improved so that microalgae biofuels can compete with fossil fuels, due to their low energy yield, compared to the demand for growth and biomass recovery routes. Also, these optimizations will contribute to improving the environmental performance of microalgae (Shimako et al., 2016). However, when comparing Figures 4.6a and 4.6b with Figure 4.4b, which shows the normalized impacts of the base scenarios, there was a reduction in the impacts in the four presented categories. In the base scenarios, climate change, freshwater eutrophication, and fossil resource scarcity were negatively affected by the briquettes in scenarios 1 and 2. In the sensitivity analysis of Figure 4.6a, two of these categories have been benefited from the new life cycle of scenario 1a. In the sensitivity analysis presented in Figure 4.6b, scenario 2b starts

to benefit the environment in all impact categories, but scenario 1b continues to negatively impact terrestrial ecotoxicity, due to the CO₂ supplementation for microalgae growth. It should be noticed that the normalized environmental benefits gradually increased from Figure 4.4b to Figure 4.6a and 4.6b, while environmental losses decreased. So, after reducing the consumption of electricity for drying and considering MB briquette as a substitute for coal, it was possible to obtain an attractive source of bioenergy, which does not emit GHG nor does it contribute to fossil resource scarcity. This result is similar to that found by Alanya-Rosenbaum and Bergman (2019), who considered, for the life cycle of the modeled forest waste briquettes, the generation of 1 MJ of thermal energy for domestic heating and the generation of 1 kWh electricity through co-combustion with coal. In these two scenarios, the replacement of propane gas and coal with biomass briquettes provided a reduction in GHG emissions.

It is important to highlight that the sensitivity analysis performed in this study is only one among the several possibilities that can be changed so that the model is recalculated, and the impacts compared. In both microalgae and briquette LCAs, it is usual to vary the mix of electricity, given that this parameter is responsible for varying the environmental impacts of the evaluated life cycles. Passell et al. (2013) replaced the German electricity mix with the United States mix and found an additional 55% eq CO₂ emission caused by the exchange of electricity from one country to another. This occurred because the U.S. mix has 15% more fossil fuels than the German mix (IEA, 2019b), highlighting the need to diversify the electric energy mix with renewable sources. Saba et al. (2020) evaluated the differences in the LCA of forest biomass briquettes, caused by the inclusion of 12% more renewable sources in the Lebanese mix. In this case, there was a reduction in emissions in several of the impact categories, again emphasizing the importance of increasing the contribution of renewable energies.

In the case of microalgae modeling, Porcelli et al. (2020) simulated different operational strategies during microalgae growth and concluded that by increasing biomass productivity, environmental impacts were reversed to environmental benefits. Mediboyina et al. (2020) found, in sensitivity analysis, a reduction in the contribution to the category of climate change and a more favorable energy balance by increasing the productivity of microalgae and reducing the consumption of electricity in the growth stage. In this research, the increase in microalgae productivity occurred through the use of the hybrid reactor and this proved to be environmentally feasible since the life cycle with only HRAP had a considerable impact on terrestrial ecotoxicity and this category caused a strong pressure on the environment. The

association of HRAP with BR, forming the hybrid reactor, showed to be an interesting option to avoid this impact.

4.4. Conclusion

The environmental performance of MB briquettes was obtained through LCA via modeling in the SimaPro® software. The cradle-to-gate boundary modeling was applied to the growth, harvesting, drying, and briquetting of MB, obtained during the treatment of domestic wastewater. Two wastewater treatment and microalgae growth reactors (a HRAP and a hybrid reactor) were evaluated. Thus, the potential environmental impacts were obtained in 10 impact categories for each growth reactor, both for the biomass and briquettes production.

When comparing the microalgae growth reactors, the HRAP had lower emissions than the hybrid reactor in 9 of the 10 categories. However, through impacts normalization, it was possible to observe that the only category (terrestrial ecotoxicity) in which the HRAP had the most damaging impact, compared to the hybrid reactor, was the one that most caused negative pressure on the environment. Emission of $3.5E-01$ kg 1,4-DCB eq has severely harmed the environment when compared to other impact categories and the hybrid reactor. Terrestrial ecotoxicity impact was mainly caused by the production of synthetic CO₂ used for microalgae growth supplementation in the HRAP.

The major obstacle of the MB valorization route through briquettes was the drying stage. In some of the impact categories, drying represented up to 99% of the potential environmental impacts, due to the significant electricity consumption. Therefore, LCA was optimized with the natural reduction of MB humidity to minimize electricity demand in the biomass drying stage. This scenario favored the energy balance of the process, positively contributing to the environment. Also, when microalgae biomass briquettes were considered substitutes for coal briquettes, in CHP plants, besides wastewater nutrients, coal extraction was avoided, benefiting the environment.

Thus, to enhance the feasibility and sustainability of MB briquettes, efforts must be made to reduce environmental impacts. According to this study, impacts can be minimized in the microalgae growth stage, by reducing or replacing inputs; and, mainly, in the drying stage, by reducing the electricity demand through solar drying. Therefore, critical aspects of the process were highlighted, guiding the next research. Contributions were proposed to develop the energy

conversion route of microalgae biomass through briquetting, aiming at more sustainable products, which cause less pressure on the environment. Moreover, contributions should help to make microalgae biotechnology more environmentally attractive and a potential alternative to renewable energy.

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5. ENVIRONMENTAL PERFORMANCE OF MICROALGAE HYDROTHERMAL LIQUEFACTION: LIFE CYCLE ASSESSMENT AND IMPROVEMENT INSIGHTS FOR A SUSTAINABLE RENEWABLE DIESEL

Abstract: The potential environmental impacts of bio-oil production from algal biomass via hydrothermal liquefaction (HTL) and its upgrading in renewable diesel were obtained. In this life cycle assessment, the gate-to-gate boundary was applied with a functional unit to obtain 1 MJ of energy. The operational parameters of the HTL were evaluated, and the effects of the outputs (solid, aqueous, and gaseous phases) were also studied. With the normalization of the results, 6 impact categories stood out due to the negative pressure that the environment suffered (marine eutrophication, climate change, marine ecotoxicity, fossil fuels depletion, human toxicity, and terrestrial acidification). The environment suffered from marine eutrophication due to emissions of 0.0017 kg of nitrogen equivalent to water from the aqueous phase. HTL was more harmful to the environment due to the heat demand. The bio-oil upgrading was responsible for about 79% of the environmental impacts in the climate change category. Uncertainties of the model, calculated with Monte Carlo analysis, varied from 20% in climate change to 60% in the results of marine eutrophication. The sensitivity analysis highlighted the importance of the residual heat recovery in the HTL reactor. The 10% increase in this parameter can reduce up to 52% of impacts on marine ecotoxicity. Finally, a new scenario was proposed to reduce the heat input into the HTL reactor and recirculate the aqueous phase after dilution. Results demonstrated a 45% reduction in the potential environmental impacts of the bio-oil life cycle, with the environment no longer suffering from marine eutrophication.

Keywords: Bioenergy; Sustainability; wastewater grown microalgae biomass; bio-oil.

Abbreviations

1,4-DCB	1,4- diclobenzene
AD	Anaerobic digestion
Al	Aluminum
Al ₂ O ₃	Aluminum oxide
AP	Aqueous phase
C	Carbon
CH ₄	Methane
CHP	Combined heat and power

CI	Confidence interval
CO ₂	Carbon dioxide
DOC	Dissolved organic carbon
eq	Equivalent
FU	Functional unit
GP	Gaseous phase
H ₂	Hydrogen
HHV	Higher heating value
HTL	Hydrothermal liquefaction
LCA	Life cycle assessment
LCI	Life cycle inventory
LCIA	Life cycle impact assessment
MB	Microalgae biomass
MoO ₃	Molybdenum trioxide
N	Nitrogen
NH ₃	Ammonia
NO _x	Nitrogen oxides
PM _{2.5}	Particles smaller than 2.5 μm
SO ₂	Sulfur dioxide
SP	Solid phase
SS	Suspended solids
TKN	Total Kjeldahl nitrogen
VC	Variation coefficient
WWTP	Wastewater treatment plant

5.1. Introduction

In the context of high energy demand and climate change, alternative energy sources are important options to be sought [1,2]. Biotechnologies based on microalgae may represent vital solutions to replace fossil fuels [3]. Characteristics such as rapid growth throughout the entire year and cellular content make them a potential substrate for third-generation biofuels generation [4]. Microalgae biomass (MB) can also be obtained through wastewater treatment, protecting water bodies from eutrophication [4,5].

A promising way of valuing MB is hydrothermal liquefaction (HTL) for bio-oil production [6,7]. This thermochemical conversion requires subcritical conditions of temperature (ranging from 200 to 374 ° C) and pressure (in the range of 5 to 20 MPa), with a reaction time of 5 to 120 minutes [8]. Another critical parameter is the biomass/water ratio [9,10]. Despite being a wet route, higher bio-oil yields were obtained when performing HTL with less humid biomass, indicating the need for prior biomass dewatering [11–13]. The use of wet biomass is the main advantage of HTL compared to other energy thermochemical routes since the drying stage is avoided [14,15]. Related to other technologies for converting MB into liquid biofuels, such as biodiesel or bioethanol, HTL is interesting because it converts the entire organic content of biomass into bio-oil and not just the lipid or carbohydrate fraction [16]. However, despite these advantages, the aforementioned operational parameters still need to be improved [9], bearing in mind that bio-oil quality also depends on the biomass characteristics [16].

The MB bio-oil produced from HTL has a calorific value varying between 22 and 39 MJ/kg [10], close to that of crude oil [14,17]. It can be upgraded as an oil substitute, aiming to produce chemical products, or be applied as a renewable transport fuel and heat and energy generation. In addition to bio-oil, other subproducts from HTL are the solid, aqueous, and gaseous phases [9,14,15,18]. However, there are still no integrated ways of recovering energy and nutrients by enhancing the outputs of HTL, indicating MB as a source of unsustainable renewable energy due to the technological challenges that need to be overcome [15].

Within this context, life cycle assessment (LCA) is a powerful method to analyze the environmental performance of products and services. It has been applied to various MB valorization routes to detect the most impacting stages and processes [16]. For example, in a MB biofuel LCA, the drying and lipid extraction stages were identified as the ones that most require electricity, generating a negative energy balance and not favoring the environment [4,19]. Again, biomass drying was decisive for increasing the environmental impacts in pyrolysis compared to the LCA of HTL [20,21].

Despite the expressive potential of HTL and the ability of LCA to help understand the potential environmental impact of processes, analyzes focused on HTL from MB, especially with the use of wastewater, are scarce. Therefore, much still needs to be investigated to improve HTL so that MB biofuels are established. Through LCA, this study aims to assess the potential environmental impacts of MB bio-oil via HTL. It seeks to improve the environmental performance of this energy route, covering from the reaction operational parameters to the bio-

oil upgrading. Thus, it hopes to contribute with information on the bottlenecks of the processes to achieve the sustainability of renewable microalgae fuel, mainly when domestic wastewater is used for biomass growth.

5.2. Material and methods

Modeling was performed in the software System for Integrated Environmental Assessment of Products (SimaPro®), version 8.2.3, PhD [22], based in the International Standards “Environmental Management - Life Cycle Assessment”, ISO 14040 - Principles and structure and ISO 14044 - Requirements and guidelines [23,24]. Four steps were adopted: goal and scope definition; life cycle inventory (LCI); life cycle impact assessment (LCIA); and the interpretation of results [23–25], as found in the literature [21,26–30].

5.2.1. Goal and scope definition

The functional unit (FU) was to obtain 1 MJ of renewable fuel, also adopted in other studies [19,21,31,32]. The gate-to-gate system boundary modeling was applied to the biomass dewatering, HTL, phase separation, and bio-oil upgrading stages (Figure 5.1).

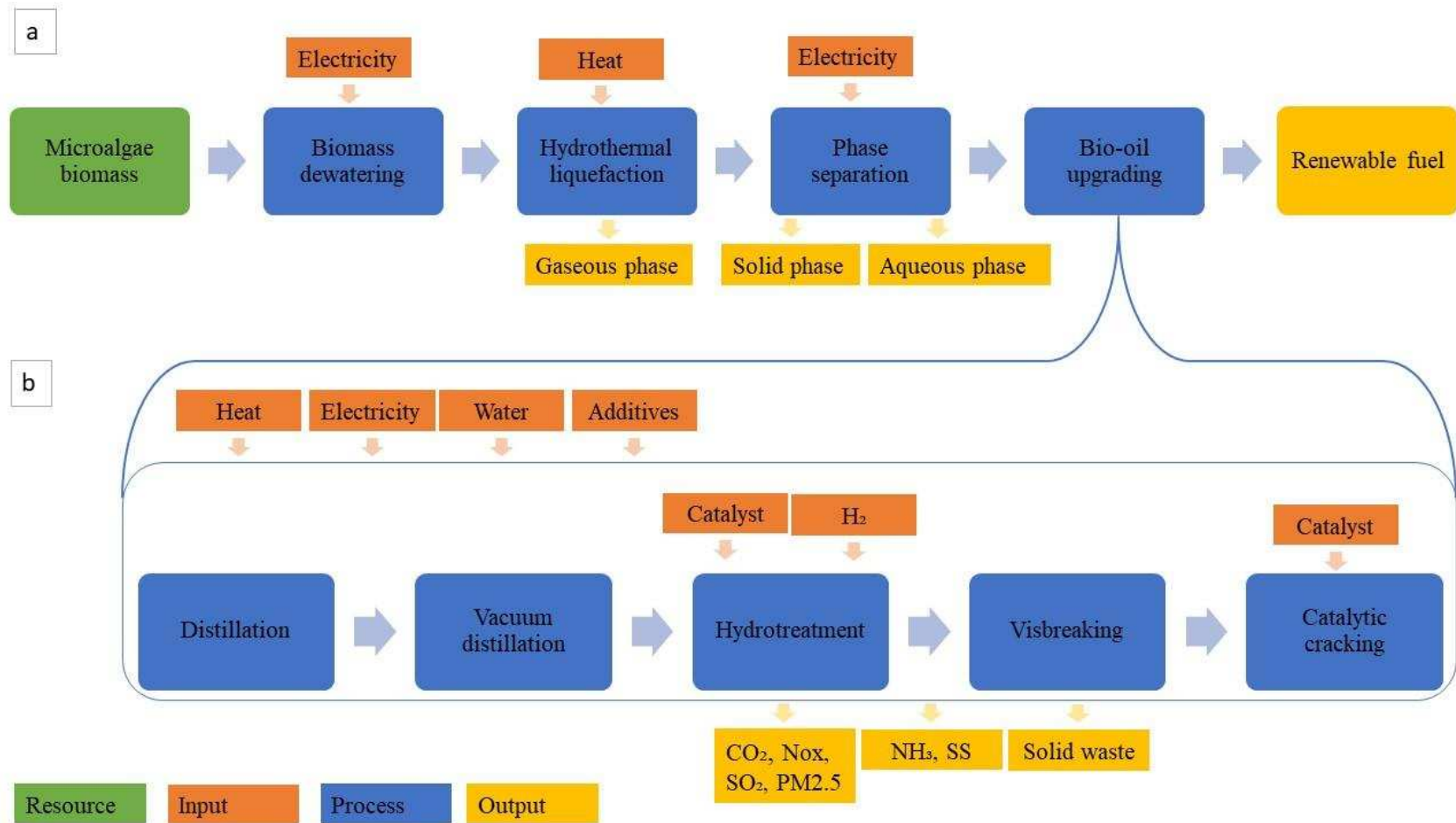


Figure 5.1. Stages included in (a) the gate-to-gate system boundary of the microalgae bio-oil life cycle via HTL and (b) the bio-oil upgrading (base scenario)

The stages of obtaining biomass, such as microalgae growth and harvesting, were not included in the system boundary. Castro et al. [26] stated that these stages are part of the operation of the wastewater treatment plant (WWTP) that is obligatory to occur. Thus, the impacts of the wastewater treatment and, consequently, microalgae production were not loaded into the bio-oil.

The use of catalysts in the HTL reactor was not considered. The HTL outputs - solid phase (SP), aqueous phase (AP), and gaseous phase (GP) - were treated as emissions to the soil, water, and air, as it was also sought to evaluate and quantify the potential environmental impacts of these launches. Therefore, the allocation of environmental responsibilities was unnecessary, following ISO 14040 [23].

The upgrading to transform bio-oil into renewable fuel was included at the system boundary, but the use of fuel was not, similar to Bennion et al. [20]. The impacts of transporting inputs and materials, capital goods, and the end of the useful life of infrastructure and equipment were not considered, as also done by Colzi Lopes et al. [33] and Passell et al. [34].

5.2.2. Assumptions made for the life cycle inventory (LCI) and research limitations

Researches that used MB produced in domestic sewage for HTL were consulted to obtain yields (bio-oil, SP, AP, and GP) and the bio-oil higher heating value (HHV) [11,35–40]. The operational parameters (reaction time and temperature, pressure in the reactor, residual heat recovery, and solids content in the biomass) were obtained in the abovementioned research, and also in others, in which the biomass was produced in the synthetic or marine growth medium [12,31,41–44]. Literature information was compiled, units were made compatible, and, subsequently, the minimum, average and maximum values were obtained (Table 5.1), composing part of the bio-oil LCI. The average values were assumed to model the base scenario for obtaining bio-oil (Figure 5.1), as described below. Meanwhile, the minimum and maximum values were applied in the analyzes explained in section 5.2.4.

Table 5.1. Average, minimum, and maximum values of operational parameters of HTL and reaction yields, found in the literature.

Parameter	Unit	Average value ¹	Minimum value	Maximum value
Reaction time	min	35	10	90
Reaction temperature	°C	300	260	350
Pressure in the reactor	MPa	8.2	0.7	17.0
Recovery of residual heat in the reactor	%	85	70	90
Solids content in biomass ²	wt% ³	16.5	1.5	25.0
Bio-oil HHV	MJ/kg	30.4	15.0	58.7
Bio-oil yield	% ³	32.5	25.8	39.0
SP yield	% ³	41.4	5.0	60.0
AP yield	% ³	17.6	5.0	65.0
GP yield	% ³	8.5	5.0	15.4

¹ Values used in modeling the base scenario;

² In some of the references consulted, the biomass humidity was presented and not the solids content. Therefore, the solids content was obtained by the difference between 100% and humidity of biomass (in%);

³ Dry base.

References consulted to obtain the average, minimum and maximum values: [11,12,31,35–44].

Starting the calculations of the LCI of the base scenario, the reference flow was obtained using the average values of HHV (30.4 MJ / kg) and bio-oil yield (32.5% based on dry biomass). It was considered that in the upgrading, about 30% of the raw material was lost as waste [45]. Thus, in the base scenario, the reference flow was 0.05 kg of bio-oil, obtained by 0.14 kg of MB (dry basis), necessary to obtain the FU with the aforementioned upgrading efficiency.

Biomass dewatering: MB from the WWTP with approximately 93% humidity after the gravitational settling [27] was concentrated to an average solids content of 16.5%. Electricity expenditure by the centrifuge (Electricity for dewatering) in kWh was estimated according to Equation 1 [31].

$$\text{Electricity for dewatering (kWh)} = E * W_s / S_i * d_s \quad (1)$$

Where, E is the electricity requirement of the centrifuge (1.20 kWh.m⁻³ [31]); W_s is the mass of dry biomass (0.14 kg); S_i is the average initial solids content of the biomass (0.007) and d_s is the specific mass of the biomass (1 kg.m⁻³).

HTL reaction: Equation 2 [31] was used to estimate the heat consumption for HTL (Heat for HTL) in kJ.

$$\text{Heat for HTL (kJ)} = [W_w * C_{pw} * \Delta T + W_s * C_{ps} * \Delta T] * (1 - R_c) \quad (2)$$

Where, W_w is the mass of water in the reactor (0.71 kg); C_{pw} is the specific heat of the water ($4.18 \text{ kJ.kg}^{-1}.\text{°C}^{-1}$ [46]); ΔT is the temperature increase required for the reaction, equal to 275 °C (considering the average temperature of 300 °C and that the wet biomass was at room temperature of 25 °C); W_s is the mass of solids in the reactor (0.14 kg); C_{ps} is the specific heat of the biomass ($1.25 \text{ kJ.kg}^{-1}.\text{°C}^{-1}$ [46]); and R_c is the residual heat recovery in the reactor, adopted as 0.85 [20].

After HTL, an average of 32.5% of the biomass (mass on a dry basis) was converted into bio-oil (yielding about 0.05 kg), and, on average, 41.4% became the SP, 17.6% formed the AP, and 8.5% generated the GP (Table 5.1). Emissions to the soil, air, and water were quantified by relating the performance of the phases to their characterization. It should be noted that predominant components represent the SP, AP, and GP composition. In the SP, the components considered were ash, carbon (C), and nitrogen (N). In the AP, dissolved organic carbon (DOC) and total Kjeldahl nitrogen (TKN). Finally, in the GP, carbon dioxide (CO_2) and methane (CH_4) gases were considered for emission into the air. The chemical composition of these phases was obtained in Couto et al. [11].

Phase separation: It was considered that the entire GP emitted during the operation of the HTL reactor was collected, and gases (CO_2 and CH_4) were quantified as emissions to air. The separation of bio-oil from the other phases (SP and AP) was done in a disk centrifuge, whose consumption of electricity (Electricity for phases separation) was through Equation 3 [31].

$$\text{Electricity for phases separation (kWh)} = El * W_s / C * ds \quad (3)$$

In which El is the electricity requirement of the disk centrifuge (130 kW [31]); W_s is the mass of dry biomass, equal to 0.13 kg (weight of biomass on a dry basis minus 8.5% GP); C is the capacity of the equipment ($190 \text{ m}^3.\text{h}^{-1}$ [31]), and ds is the specific mass of the biomass ($1\text{k}.\text{m}^{-3}$).

Considering the separation with 100% of efficiency, 0.06 kg of GP was obtained and released into the soil (ashes, C and N). In the AP, 0.02 kg was obtained in emissions to water (DOC and TKN).

Bio-oil upgrading: According to Baloch et al. [47], Kumar and Strezov [48] and the processes implemented in an oil refinery, described in the report “Erdöl”, ecoinvent report No. 6 - Part IV [45], hydrotreatment and visbreaking were considered, as being the most important for bio-

oil and crude oil upgrading. Moreover, distillation, vacuum distillation, and catalytic cracking were part of the bio-oil upgrading in renewable diesel [45] (Figure 5.1b).

At the refinery, the need for net electricity of 0.037 kWh per kg of oil was considered for the operation of pumps and other equipment and, for the upgrading, the heat demand was 1 MJ per kg of diesel produced. It was also considered the input of 0.1 m³ of water per ton of fuel produced. And the total requirement of 10 g of catalysts for hydrotreating and catalytic cracking per ton of oil. The main components of the catalyst for catalytic cracking were 30% aluminum (Al) and 1% sulfur (S), and for hydrotreatment were 77% aluminum oxide (Al₂O₃) and 19% molybdenum trioxide (MoO₃) [45]. In this last stage, an average amount of 15 g of hydrogen (H₂) per kg of bio-oil was added [49]. As additives, a flow property enhancer, the ethylene-vinyl acetate copolymer, was used in a concentration of 400 ppm (weight). Emissions to water were 6.0 mg of ammonia (NH₃) per L of water and 10 mg of suspended solids (SS) per L. The primary emissions to air were 3,120 kg of CO₂ per ton of fuel; 206 g of nitrogen oxides (NO_x) per ton of oil; 2.4 g of particles smaller than 2.5 μm (PM_{2.5}) per kg of oil and 158 g sulfur dioxide (SO₂) per ton of oil [45]. These are some of the inputs and outputs of a refinery, representing a suggestion of bio-oil upgrading and a limitation of this modeling.

Each of the inputs (heat, electricity, chemical inputs) was modeled using a “known input from the technosphere”, represented by process systems from the ecoinvent v3.1 library (Supplementary material Table S1 (Appendix B)), composing the background data of the modeling.

5.2.3. Life cycle impact assessment (LCIA)

In this step, ISO 14044 recommendations were followed to carry out the classification, characterization, and normalization of LCI results [24]. Thus, the elements of the LCI were assigned to the impact categories through classification. With the characterization, each result was multiplied by characterization factors, forming the equivalent (eq) emissions. Subsequently, through normalization, the impact categories were expressed on the same scale (ecopoints) and compared.

ReCiPe 2016 midpoint, hierarchical, version 1.12, was the LCIA method which has 18 impact categories and uses global mechanisms (ReCiPe Midpoint H v1.12 / World). As a reference for normalization, this method considers the average global pressure applied on the environment by an individual in 2010 [50,51]. The potential environmental impacts were assessed using the

18 categories: climate change, ozone depletion, terrestrial acidification, freshwater eutrophication, marine eutrophication, human toxicity, photochemical oxidants formation, particulate matter formation, terrestrial ecotoxicity, freshwater ecotoxicity, marine ecotoxicity, ionizing radiation, occupation of agricultural land, occupation of urban land, transformation of natural land, water depletion, metal depletion, and fossil fuels depletion.

Several complex process systems from the ecoinvent v3.1 library were used in the modeling, and any of the categories mentioned above could be strongly impacted. So, it was decided to obtain the potential environmental impacts in all available impact categories [27,28,30,52,53]. However, after obtaining the results of the LCIA, only the impact categories in which the environment was most under pressure were highlighted and considered for the sequential analyzes.

5.2.4. Interpretation of results

After identifying the most relevant impact categories, contributions, uncertainties, and sensitivity analyzes were carried out, following ISO standards [23,24], to verify and better understand the results.

Contribution analysis: Was carried out to identify which elements contributed most to the potential environmental impacts of the bio-oil life cycle. The contribution percentage of the modeled stages, the inputs, and the outputs of the life cycle were obtained.

Uncertainty analysis: Was applied considering that the bio-oil yield and HHV were the main results of the HTL and the range of values found in the literature. Also, as these two parameters were used to find the reference flow of the LCI elements, the minimum and maximum values of bio-oil yield (min.: 15.0% and max.: 58.7%, dry basis) and HHV (min.: 25.8 MJ/kg and max.: 39.0 MJ/kg) were tested. These values were used to define two new reference flows (both considering upgrading 70% of bio-oil in renewable fuel). Thus, for the minimum yield and HHV, the reference flow was 0.06 kg of bio-oil through HTL of 0.37 kg of MB. For maximum yield and HHV, a reference flow of 0.04 kg of bio-oil and 0.06 kg of MB was applied.

Subsequently, the calculations described in section 5.2.2 have been redone, and the LCI recalculated. The new inputs and outputs values were inserted in the base scenario model with the minimum and maximum data variation. Monte Carlo statistical method was used in SimaPro® to calculate the absolute uncertainty of the results. The value of each parameter was varied (between the minimum and the maximum), and the potential environmental impacts

were recalculated. The procedure was repeated 10,000 times to obtain the range of uncertainties, with a 95% confidence interval (CI). The probability distribution of the occurrence of the results of each impact category was found. The variation coefficient (VC) was determined to express the variability of the results, excluding the influence of the variables' magnitude order so that the ranges of uncertainties could be compared [54].

Sensitivity analysis: After identifying the inputs that most affected the environment, variations were made in its parameters, increasing and decreasing the fixed value of 10%, similar to what was performed by Sun et al. [21] and Xiao et al. [55]. Sensitivity analysis was done to verify the degree of influence of these inputs on the result.

5.2.5. Obtaining a scenario with less environmental impact

After identifying the critical stages, changes were proposed in the model assumptions, aiming for the better environmental performance of the bio-oil life cycle. Subsequently, a new scenario was modeled, considering the points highlighted through the results of previous analyzes, and compared to the base scenario. For this comparison, using the Monte Carlo method, variations in the reference flow and the absolute uncertainties of the model were maintained.

5.3. Results and discussion

The elements included in the LCI of the base scenario are shown in Table 5.2, containing all inputs and outputs. The values present in the column "Base scenario" represent the elements calculated for the LCI of the scenario modeled with the reference flow of 0.05 kg of bio-oil. The values of the columns "Minimum yield and HHV" and "Maximum yield and HHV" were obtained through the reference flow of 0.06 and 0.04 kg of bio-oil, respectively.

Table 5.2. Elements of the microalgae bio-oil life cycle inventory via LHT.

Life cycle elements	Unit	Base scenario	Minimum yield and HHV	Maximum yield and HHV
Biomass dewatering				
Inputs				
Wet biomass (7% solids + 93% water)	kg	2.00	5.27	0.89
Electricity	kWh	0.0024	0.0063	0.0011
Outputs				
Dewatered wet biomass (16.5% solids + 83.5% water)	kg	0.85	2.24	0.38
HTL reaction				
Inputs				
Dewatered wet biomass (16.5% solids + 83.5% water)	kg	0.85	2.24	0.38
Heat	kJ	129.64	341.14	57.67
Outputs				
Bio-oil with impurities	kg	0.05	0.06	0.04
Emission to air: CO ₂	kg	0.0096	0.0177	0.0030
Emission to air: CH ₄	kg	0.0002	0.0004	0.0001
Phase separation				
Inputs				
Bio-oil with impurities	kg	0.05	0.06	0.04
Electricity	kWh	0.0001	0.0002	0.00004
Outputs				
Crude bio-oil	kg	0.05	0.06	0.04
Emission to soil: C	kg	0.0044	0.0178	0.0011
Emission to soil: N	kg	0.0003	0.0011	0.0001
Waste: Ash	kg	0.0517	0.2069	0.0125
Emission to water: DOC	kg	0.0108	0.0299	0.0044
Emission to water: TKN	kg	0.0017	0.0048	0.0011
Bio-oil upgrading				

Life cycle elements	Unit	Base scenario	Minimum yield and HHV	Maximum yield and HHV
Inputs				
Crude bio-oil	kg	0.05	0.06	0.04
Heat	kJ	0.0350	0.0388	0.0256
Electricity	kWh	0.0019	0.0020	0.0014
Water	L	0.0035	0.0039	0.0026
Catalyst (30% Al + 1% S)	g	0.0003	0.0003	0.0003
Catalyst (77% Al ₂ O ₃ + 19% MoO ₃)	g	0.0003	0.0003	0.0003
Hydrogen	g	0.75	0.83	0.55
Additives (ethylene-vinyl acetate)	g	0.014	0.016	0.010
Outputs				
Renewable diesel (1 MJ)	kg	0.035	0.039	0.026
Emission to air: CO ₂	kg	0.1092	0.1209	0.0800
Emission to air: NO _x	g	0.0103	0.0114	0.0075
Emission to air: SO ₂	g	0.0079	0.0087	0.0058
Emission to air: PM _{2.5}	g	0.0001	0.0001	0.0001
Emission to water: NH ₃	mg	0.021	0.023	0.015
Emission to water: SS	mg	0.035	0.039	0.026
Waste: Ash	kg	0.015	0.017	0.011

5.3.1. Potential environmental impacts of the bio-oil life cycle

Using the LCIA method, ReCiPe Midpoint H v1.12 / World, the potential environmental impacts of the life cycle of MB bio-oil via HTL were obtained in the 18 impact categories (the result of classification and characterization) (Figure 5.2a). However, with normalization, 6 of them were highlighted: marine eutrophication, climate change, marine ecotoxicity, depletion of fossil fuels, human toxicity, and terrestrial acidification, in that order of importance (Figure 5.2b).

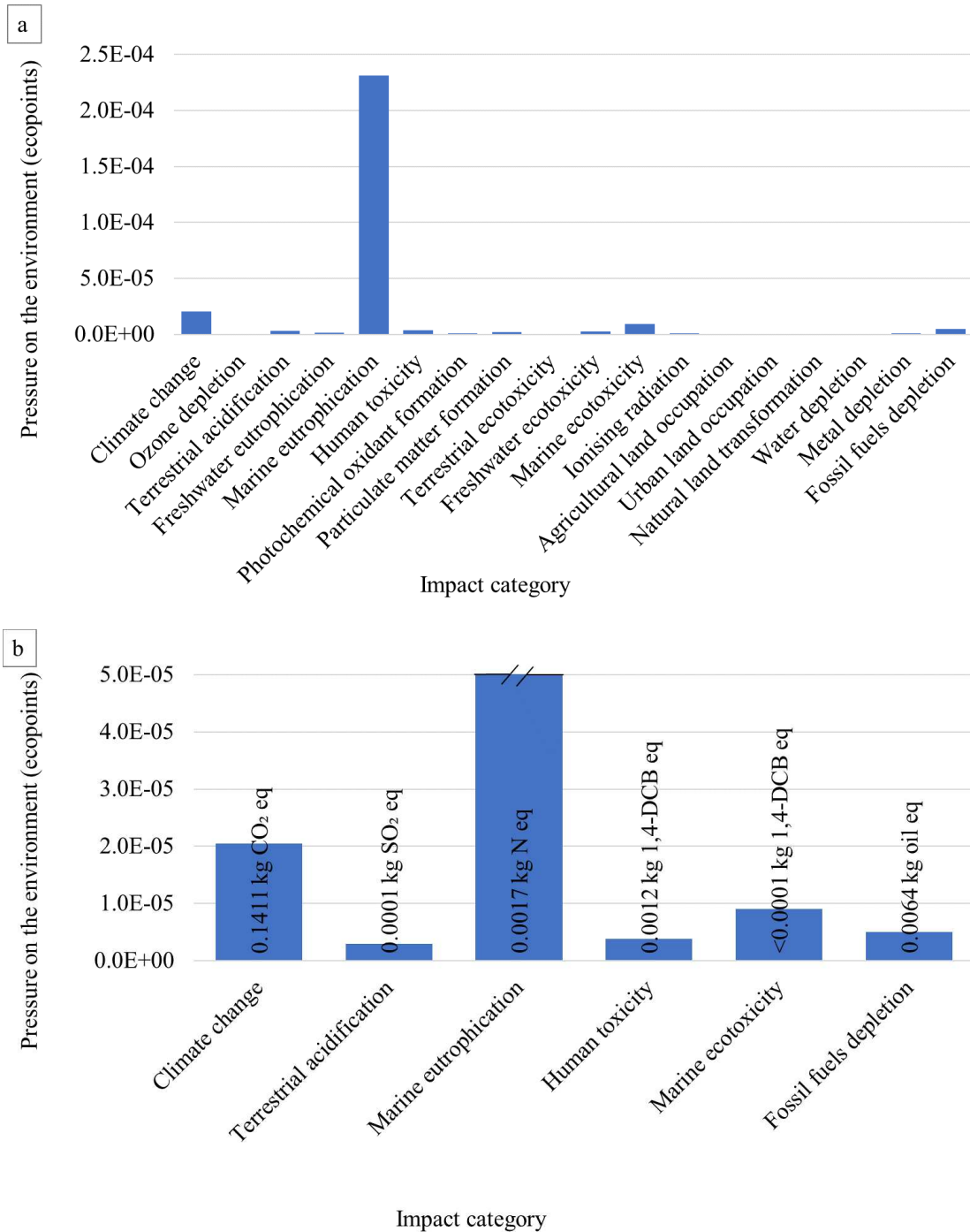


Figure 5.2. Relative comparison between the potential environmental impacts of the life cycle of microalgae bio-oil via HTL (base scenario) (a) in the 18 categories evaluated and (b) in the 6 categories that most exerted pressure on the environment.

The environment suffered intense pressure with marine eutrophication due to emissions of 0.0017 kg of N eq to the water. Marine eutrophication was followed by climate change due to emissions of 0.1411 kg of CO₂ eq to the air, marine ecotoxicity (emission less than 0.0001 kg of 1,4-dichlorobenzene (1,4-DCB) eq to water), fossil fuels depletion (extraction of 0.0064 kg

of oil eq), human toxicity (emission of 0.0012 kg of 1,4-DCB eq to air) and terrestrial acidification (emission of 0.0001 kg of SO₂ eq). These 6 categories, identified as the most relevant for the modeled base scenario, were selected for later evaluations. They were also essential representatives of the potential environmental impacts in other LCAs of microalgae biotechnology [27,30,56,57].

Comparing the potential environmental impacts (Figure 5.2.b) with the outputs accounted for in the base scenario LCI (Table 5.2), it is noted that the TKN outflow into the water, in the phase separation, corresponds precisely to the amount of N eq causing marine eutrophication (0.0017 kg). However, in climate change, other LCI input or output, besides the CO₂ emitted into the air during bio-oil upgrading, contributed to the impacts in this category. In terrestrial acidification, the potential environmental impacts (0.0001 kg of SO₂ eq) were also greater than the direct output in the LCI (SO₂ in the upgrading (0.0079 g of SO₂)), since SO₂ is only one of the components considered in this category. NH₃ emissions in upgrading (0.021 mg of NH₃) were also considered and are even more harmful to this impact category [51,58]. Therefore, in addition to the SO₂ and NH₃ upgrading outputs, other elements also contributed to the environmental impacts of terrestrial acidification. Emissions of 1,4-DCB eq and the extraction of oil eq occurred because they were incorporated in some process of the ecoinvent library v3.1, used in the known inputs from the technosphere. Then, with the contribution analysis (item 3.2), the elements of the LCI that contributed to the potential environmental impacts in each category were detailed, and these questions could be clarified.

5.3.2. Contribution analysis

Figure 5.3a shows the percentage contributions of the stages modeled in the life cycle of the base scenario in the 6 impact categories in which the environment was most affected. Figure 5.3b shows the percentage contributions of inputs and outputs present in the LCI of the base scenario.

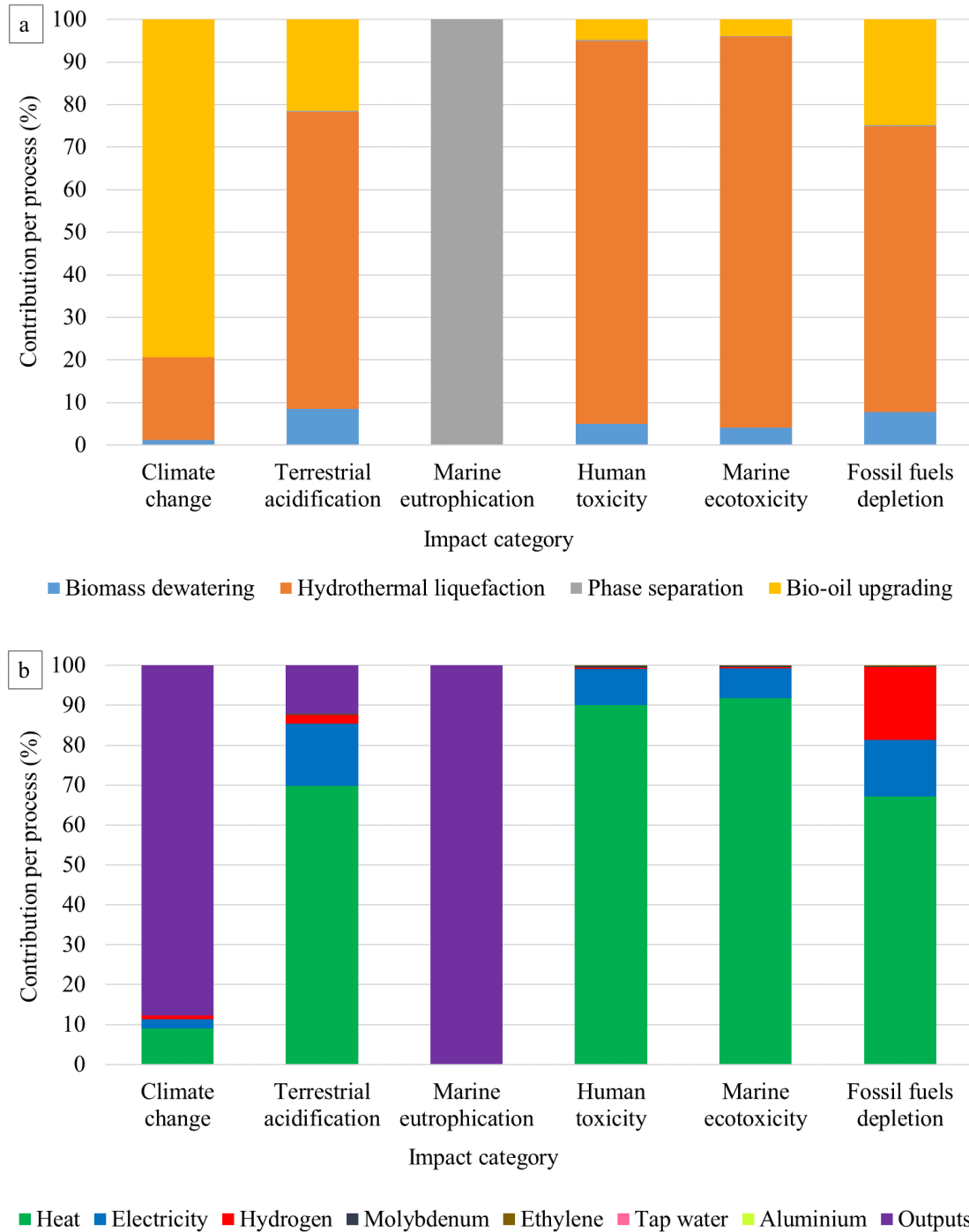


Figure 5.3. Percentage contribution (a) of the stages and (b) of the inputs and outputs modeled in the life cycle of the base scenario in the 6 impact categories in which the environment was most affected.

The phase separation caused fewer negative impacts when compared to the other stages modeled in the bio-oil life cycle, except in the marine eutrophication category. However, it should be noted that it was not the phase separation process that caused the impacts on marine eutrophication but the destination chosen for the AP. The AP was modeled as an emission to

water, which caused 100% of N eq emissions in marine eutrophication. However, in addition to TKN, the AP is also composed of DOC and other non-modeled nutrients, such as potassium, sodium and magnesium, aluminum, calcium, iron, organic, inorganic compounds, and some toxic compounds [18]. So, the impact would be more significant if all these inputs were considered.

Therefore, to improve the bio-oil scenario, there are some ways to take advantage of the AP, with promising recycling in microalgae cultivation. However, AP needs to be diluted to avoid microalgae growth inhibition [7,14,17]. Chen et al. [18] showed that, with dilutions of the AP in more than 100x of water, there was no decrease in the growth of *Chlorella vulgaris*. Also, the authors determined that, with previous activated carbon treatment, the AP can be diluted with less water and still promote microalgae development. Beims et al. [17] discussed the potential of AP recirculation in a new HTL. They reported that the recirculation could be carried out a limited number of times, requiring AP treatment at the end.

Other options for AP are: the gasification of supercritical water, converting the AP into H₂ gas rich in supercritical water, anaerobic digestion (AD) for the production of CH₄, and the fermentation in two stages for the production of CH₄ and H₂ [14,17]. However, anaerobic techniques require further studies due to the AP's toxic compounds, making the process unfeasible [59]. Torri et al. [60] tested the AD of the AP after its continuous treatment in anaerobic-aerobic conditions and, despite the reduction of organic matter, the inhibition represented a challenge to be overcome to increase the CH₄ yield and make the process viable.

The biomass dewatering was, on average, 13x less impacting than HTL in the 6 categories and, on average, 12x less impacting than the upgrading in 4 categories. Only in human toxicity and marine ecotoxicity, the dewatering was about 3% more impactful than the upgrading. Considering that this stage required only one input, electricity, it can be said that the impacts associated with dewatering come from this demand. In this bio-oil life cycle, the sources of heat and electricity were responsible for the greatest number of emissions. Together they caused, on average, 91% of the impacts on terrestrial acidification, human toxicity, marine ecotoxicity, and fossil fuels depletion. Therefore, the potential environmental impacts could be more negligible if an electricity mix with fewer fossil fuels was used or a renewable source was used to heat supply [34]. Fortier et al. [31] considered the supply of heat for HTL through gas from a WWTP sludge and gas collected from landfills. Arashiro et al. [56] assumed that the electricity and heat for the modeled stages were provided through the combined heat and power (CHP)

generation using the biogas produced by the AD of the primary sludge from a WWTP. Souza et al. [29] considered the electricity supply for the modeled stages from photovoltaic panels.

The HTL impacted, on average, 10x more than upgrading, except for climate change, where upgrading was responsible for approximately 79% of CO₂ eq emissions. Therefore, although the upgrading has a more significant number of inputs and outputs than HTL, it was the potential environmental impacts of HTL that stood out in more categories. The demand for heat in this stage may explain, which is substantially concerned compared to other processes. Besides, heat for HTL represented about 80% of the potential environmental impacts on terrestrial acidification, human toxicity, marine ecotoxicity, and fossil fuels depletion (4 of the 6 categories evaluated).

The upgrading was 4x more impactful in climate changes than HTL due to GHG emissions. Therefore, measures to mitigate these impacts need to be adopted, for example, mechanisms to reduce emissions at the refinery [61]. CO₂ and CH₄ emissions from the HTL were considered, which also contributed to climate change impacts, although to a lesser extent. However, these gases can be used as a source of C for microalgae growth [7,9,14].

The contribution of life cycle outputs (emissions to air, water, and soil) caused approximately 88% of the negative impacts on climate change (emission of 0.1238 kg of CO₂ eq) and 100% in marine eutrophication (emission of 0.0017 kg of N eq). It was possible to observe that the emission of 0.1092 kg of CO₂ into the air in the upgrading represented about 77% of climate change damage. The consumption of heat, electricity, and H₂ also contributed to 9, 2, and 1%, respectively, of the emissions that cause climate change. In terrestrial acidification, ecoinvent v 3.1 processes were the major contributors, followed by the upgrading outputs (SO₂ and NH₃, specifically) that contributed together with 12% of impacts. These ecoinvent v 3.1 processes were the supply of heat (responsible for 70% of emissions), electricity (16%), and H₂ (2%).

5.3.3. Uncertainty analysis

Two types of probability distribution of the potential environmental impacts were obtained using the Monte Carlo statistical method, the normal distribution (as in climate change) and the uniform distribution (as in marine eutrophication), as shown in the Supplementary Material (Figure S1) (Appendix B). Model absolute uncertainty, caused by the variation in the LCI of the base scenario around the minimum and maximum value of HHV and bio-oil yield (Table 5.2), is shown in Figure 5.4, as well as the VC of the results.

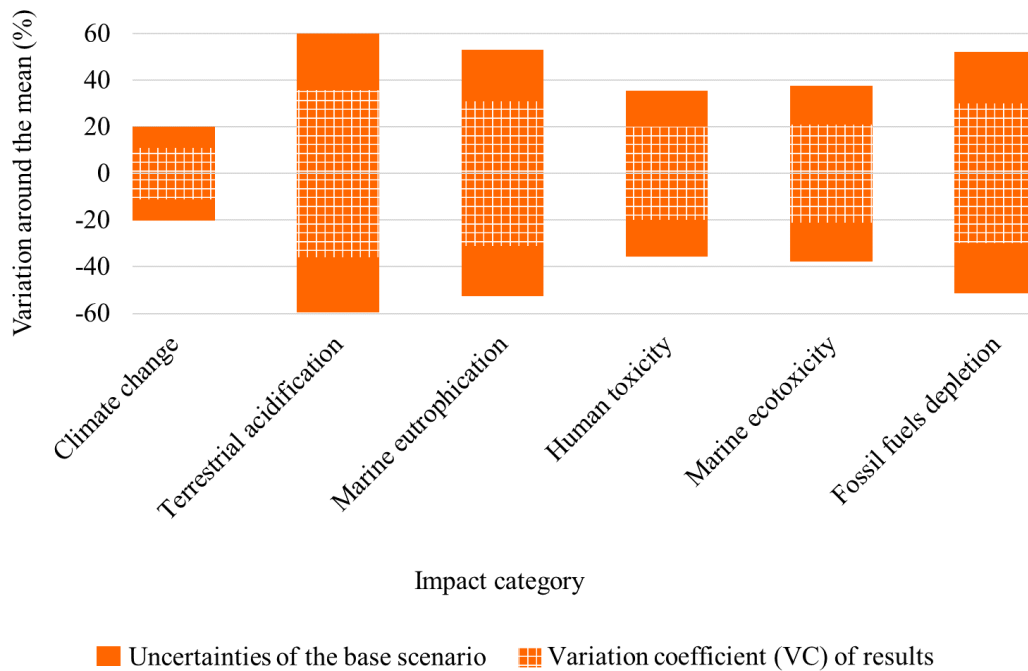


Figure 5.4. Absolute uncertainties in the potential average environmental impacts of the bio-oil life cycle and the VC of the results.

The impact category with the greatest uncertainty was marine eutrophication, with the potential environmental impacts varying about $\pm 60\%$ around the average and a VC of 36%. This result shows how much the variation of the LCI influenced the potential environmental impacts in this category. The lowest uncertainty and more consistent category was climate change, with the potential environmental impacts varying around $\pm 20\%$ around the average and a VC of 11%. When more than one element represents impacts in a category, the variation in that set of values causes uncertainty. Thus, to cause substantial uncertainty in these impact categories, it is necessary to vary all the parameters that contribute to it or that the parameter that contributes the most varies over a vast range. On the other hand, when only one element of the LCI represents 100% of the impacts, i.e., TKN in marine eutrophication, its range of variation dictates the uncertainty of the result.

5.3.4. Sensitivity analysis

The demand for heat to HTL was input that most contributed to the harmful impacts on the environment (disregarding the categories of climate change and marine eutrophication, which were mainly pressured by the output of CO_2 in the upgrading and by the output of TKN from the AP, respectively). Thus, as proposed in section 5.2.4, some parameters values in Equation 2, used to calculate the heat demand in HTL, were varied by $\pm 10\%$:

- **Sensitivity i** –The water mass in the reactor (W_w) was varied from 0.64 to 0.78 kg. For that, it was necessary to vary the biomass humidity from 75.15 to 91.85%;
- **Sensitivity ii** – The temperature increase (ΔT) was varied from 248 to 303 °C. For this, the reaction temperature had to vary from 273 to 328 °C;
- **Sensitivity iii** – The residual heat recovery in the reactor (R_c) was varied from 77 to 94%.

The heat demand for HTL, considering each of these variations (sensitivity i, ii, and iii), is shown in Table 5.3.

Table 5.3. Changes in the LCI of the base scenario for calculating the model’s sensitivity by varying $\pm 10\%$ in the HTL heat of three different input parameters.

Sensitivity	Unit	Base scenario	HTL heat input	
			Variation +10%	Variation -10%
i – Biomass humidity	kJ	129,64	141,88	117,40
ii – Reaction temperature	kJ	129,64	142,62	116,68
iii – Residual heat recovery in the reactor	kJ	129,64	56,18	203,10

The greatest variation in the heat demand for HTL occurred when the residual heat recovery in the reactor was changed by $\pm 10\%$. This recovery can be carried out using several methods that include the transfer of heat between gases and liquids (for example, the preheating of combustion air and the preheating of boiler feed water); the heat transfer to furnaces (such as for ovens preheating); the generation of mechanical and electrical energy [62]. Besides the environmental importance pointed out in this study, the recovery of residual heat during cooling and depressurization of the reactor is also crucial for the economic viability of HTL [63].

The variation in the potential environmental impacts of the bio-oil life cycle (base scenario) caused by the sensitivity analyzes is shown in Figure 5.5. The results are presented in terms of potential normalized environmental impacts so that the relevance of each category can be observed and not only the variation caused by the analyzes.

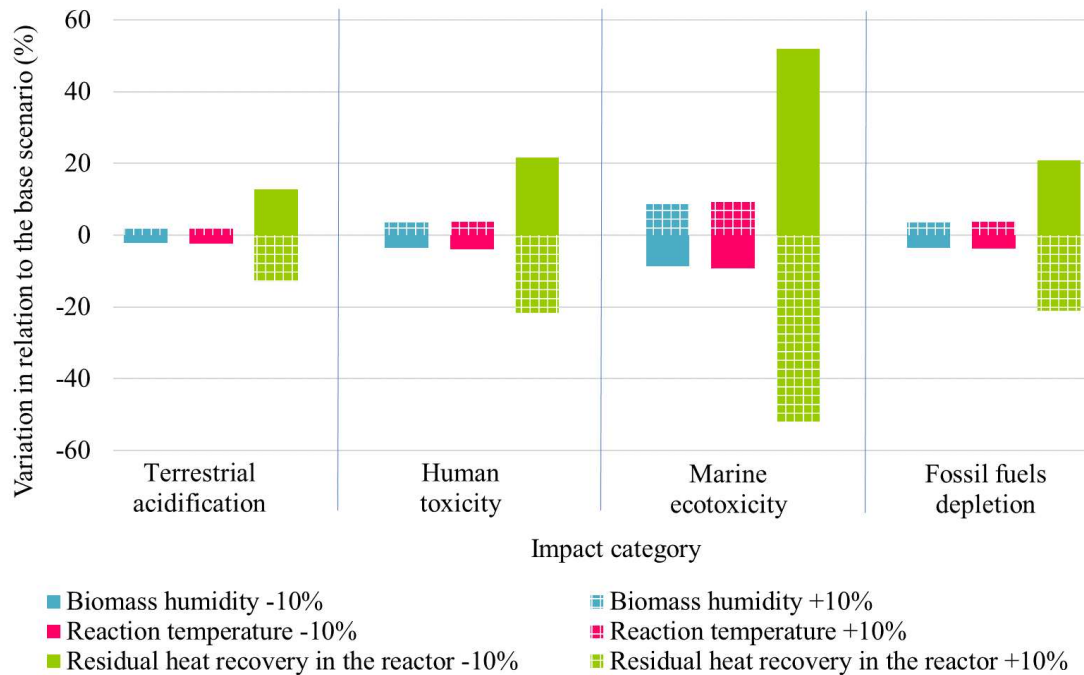


Figure 5.5. Variation of potential normalized environmental impacts of the bio-oil life cycle (base scenario) caused by the sensitivity analyzes.

Each of the four impact categories was more sensitive to the residual heat recovery in the reactor than the other changes. The category that showed the most significant reduction or increase in potential normalized environmental impacts was marine ecotoxicity. The 10% increase in the residual heat recovery in the reactor can reduce up to 52% of the potential environmental impacts in marine ecotoxicity.

The reduction in the reaction temperature can be made as long as yield losses are not observed since the temperature in the HTL reactor dictates the reactions that occur and the compounds that will be degraded [9,64]. Temperatures below 250 °C are not indicated due to the lower production of bio-oil [65]. Chen et al. [35] reported that, at 260 and 280 °C, MB HTL yielded higher proportions of SP and AP instead of bio-oil. On the other hand, Yu et al. [39] obtained a yield of about 65% of bio-oil at a temperature of 280 °C. According to Torri et al. [66], at temperatures above 300 °C, proteins and carbohydrates are converted into bio-oil, providing a greater yield of this product. However, the degradation of amino acids releases N, which can accumulate in the bio-oil. Yu et al. [39] observed an increase in the content of N and C in the bio-oil as the temperature and reaction time were increased. Couto et al. [11], with about 70% of the N present in MB, concluded that temperatures higher than 300 °C were better for bio-oil formation.

Reducing biomass humidity is achievable, and there is no harm to the process. In the modeling, the electric centrifuge was used to reduce MB humidity. However, there are other thickening methods, for example, filtration by gravity or pressure or vacuum filters and membrane filtration [67]. There are also dewatering and drying methods: sun drying, low-pressure shelf drying, spray drying, drum drying, fluid bed drying, rotary drying, flash drying, and freeze drying [67]. Among them, sun drying stands out as it does not consume electricity, presenting a potential to reduce the impacts of the bio-oil life cycle and operational costs.

5.3.5. Improvements in the life cycle of microalgae bio-oil via HTL

The values used in Equation 2 were again changed, aiming to reduce the HTL heat demand. The biomass humidity was changed from 83.5% (base scenario) to 75% [35,36], and the residual heat recovery in the reactor was changed from 0.85 (base scenario) to 0.90 [31] (values shown in Table 5.1). Moreover, to eliminate N eq emissions from AP to water, the model was recalculated, assuming the following premise. A 150x dilution of the AP in water was considered for microalgae growth without harming or inhibiting them [18]. So, the emission of DOC and TKN was no longer considered as now nutrients from AP are recycled. The potential environmental impacts of this new scenario, modeled with the three described changes, were compared to the base scenario by the Monte Carlo method (Figure 5.6).

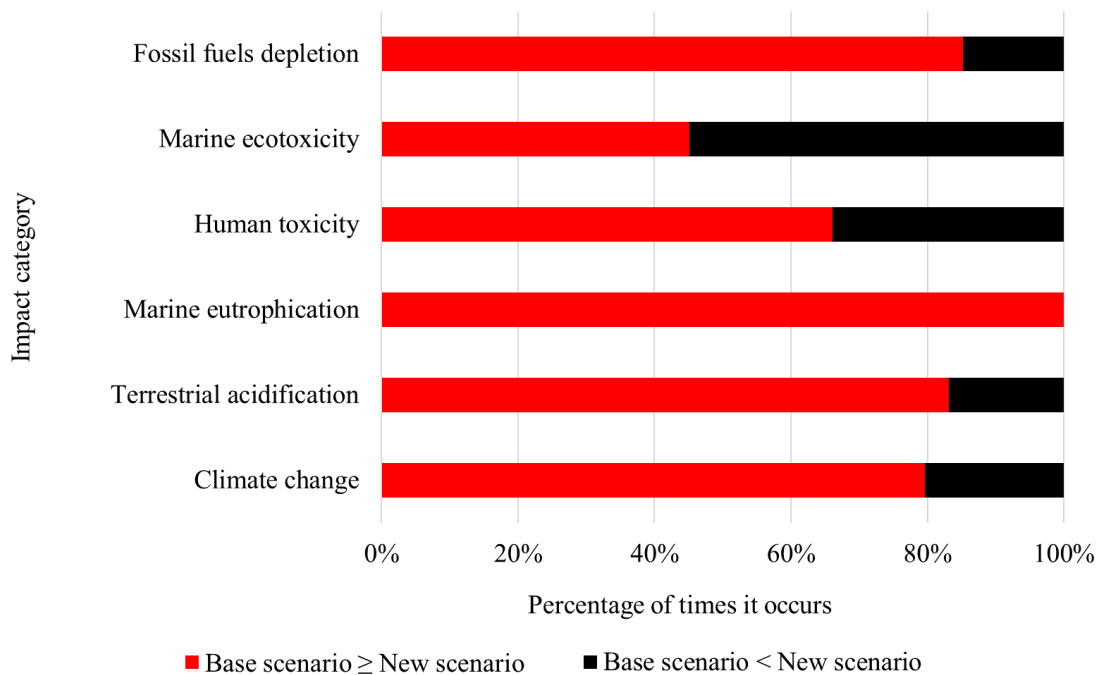


Figure 5.6. Comparison, using the Monte Carlo method, between the potential environmental impacts of the base scenario and the new scenario.

The base scenario presented a more significant environmental impact in marine eutrophication in 100% of Monte Carlo tests when compared to the new scenario. The base scenario was also more impacting than the new scenario in fossil fuels depletion (85%), human toxicity (66%), terrestrial acidification (83%), and climate change (80%). The new scenario was more impactful than the base scenario only in the marine ecotoxicity category (45% of the time) due to freshwater consumption to AP dilution. Thus, for this category, the proposal for recirculating the AP for microalgae growth was not interesting (considering the variations in the LCI).

However, a more environmentally attractive possibility, not yet explored, is the dilution of the AP in wastewater with a lower N load to balance the C, N, and P ratio, in addition to diluting the AP, avoiding inhibition of microalgae growth due to toxicity effects. Arun et al. [68] performed AP dilution in the wastewater used for microalgae growth and subjected this mixture to pollutants removal by adsorption before reuse. N must be kept in a cycle to improve the environmental performance of microalgae bioproducts [55]. Watson et al. [14] showed that about 40% of the N present in the AP could be converted back into biomass when assimilated by microalgae. In terms of mass, approximately 30% of the AP can be transferred to the new MB.

Another possibility is removing proteins from biomass before HTL to reduce NH_3 and total N formation. Barreiro et al. [69] reported a reduction of N in the bio-oil (this reduction varied by 18 and 33% in the different microalgae species evaluated) after protein removal by the enzymatic hydrolysis method developed by Romero García et al. [70]. There are many advances in this area, considering that MB can be valued to provide food supplements and animal feed [71]. Callejo-López et al. [72] developed an alkaline-enzymatic sequential method that achieves 80% protein extraction in *Chlorella vulgaris* and other species. However, as Couto et al. [11] raised, the effect of protein extraction on decreasing the MB organic content to be converted into energy should be evaluated, decreasing the bio-oil yield.

Ash is another component that can negatively affect HTL performance. A high ash content increases SP production, difficulting bio-oil formation [36] and impairing its quality [7]. Given this, ash removal procedures were experimented with MB [35,36], such as techniques of water washing and alkaline extraction tested by Hess et al. [73]. The costs of biofuel production via HTL were reduced with ash removal. However, such strategies involve chemical inputs and/or energy, and their feasibility must be assessed from a technical and environmental perspective.

As climate change was the category of impact in which the environment was most pressured and had the slightest uncertainty in the results, it was used for literature results comparison. In the life cycle of the new scenario, 0.1349 kg of CO₂ eq was emitted per MJ of renewable diesel. The renewable diesel by Bennion et al. [20], which also went through upgrading, released about 0.1060 kg of CO₂ eq per MJ when the microalgae growth was disregarded. Thus, due to the stages included in the system's boundary, the authors found negative net emissions. In other words, the renewable microalgae diesel obtained via HTL has benefited the environment by removing CO₂ from the atmosphere. A similar result was found by Sun et al. [21], in which the environment was favored due to CO₂ absorption by microalgae. Fortier et al. [31] reported that the renewable aviation fuel obtained through HTL released 0.021 kg of CO₂ eq per MJ, including MB growth at the system's border. These authors modeled the transport of renewable fuel, and its use and results were better than the average CO₂ emission values when petroleum fuel is used.

5.4. Conclusion

In the bio-oil life cycle modeled, the most impacting stage was the phase separation due to the release of TKN into the water coming from the AP. This output from the bio-oil LCI caused marine eutrophication, the impact category in which the environment suffered the most. Another damaging stage was the HTL due to the heat demand in the reactor. Among the parameters used to calculate this demand, the residual heat recovery in the reactor was the most sensitive input for this model. Thus, it was highlighted how the performance of the reactor is important for a reduction in energy consumption and environmental impacts of bio-oil production. In bio-oil upgrading in renewable diesel, despite the various inputs at this stage, it were the outputs that harmed the environment. The bio-oil upgrading caused significant emissions contributing to the increase of climate change. It is noteworthy that the analysis of uncertainties by the Monte Carlo method identified climate change as the most consistent impact category for the model of potential environmental impacts of modeled bio-oil production.

With the results obtained, improvements were proposed for the life cycle of bio-oil. These proposals involved reducing heat demand in HTL and the reuse of the AP. A new scenario was obtained with the potential environmental impacts reduced by 45%, on average. However, it is emphasized that to make the new scenario even less impactful, strategies to improve the quality of the final fuel can be adopted, such as removing ash and N from biomass.

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6. CONCLUSÃO GERAL

De acordo com este estudo, os potenciais impactos ambientais da produção dos briquetes foram reduzidos devido à fase de crescimento das microalgas, modelada por meio da substituição dos insumos (água e fertilizantes) por esgoto. Entretanto, o grande obstáculo da rota de valorização da biomassa em briquetes foi a etapa de secagem. Em algumas das categorias de impacto avaliadas, a secagem representou até 99% dos potenciais impactos ambientais, devido ao consumo significativo de energia elétrica.

O desempenho ambiental do bio-óleo foi prejudicado devido à demanda de energia térmica para a LHT. A fase aquosa (FA) foi modelada como emissão para a água, o que provocou 100% das emissões de N equivalente (eq) em eutrofização marinha, a categoria em que o ambiente mais foi pressionado negativamente. Enquanto, as demais saídas da LHT (fase sólida (FS) e fase gasosa (FG)), também modeladas como emissões ao solo e ao ar respectivamente, não se destacaram.

Considerando-se que as ACV realizadas (briquetes e bio-óleo) tiveram a mesma UF, elas foram comparadas, ainda que as fronteiras adotadas tenham sido diferentes. Para os briquetes, a fronteira do sistema abrange o cultivo da biomassa, etapa desconsiderada no ciclo do bio-óleo. A obtenção de 1 MJ de energia de briquetes de microalgas cultivadas em reator híbrido emitiu 0,1894 kg de CO₂ eq e, para obter essa mesma quantidade de energia vinda de briquetes feitos com BM cultivadas em LAT, se teve emissão de 0,0998 kg de CO₂ eq. Ressalta-se que, essas emissões resultantes tiveram uma redução de quantidade devido à forma que o cultivo das microalgas foi modelado. Portanto, desconsiderando-se o cultivo, para que as etapas modeladas para os briquetes correspondessem às etapas do bio-óleo, os briquetes de BM, tanto do reator híbrido quanto da LAT, emitiram 0,2380 kg de CO₂ eq. A obtenção de 1 MJ de energia vinda do bio-óleo de microalgas, beneficiado em diesel renovável, emitiu 0,1411 kg de CO₂ eq. Portanto, em termos de emissão de CO₂ eq, causadores de mudanças climáticas/ aquecimento global, a rota úmida da BM (LHT) foi menos impactante que a rota seca (briquetagem).

7. SUGESTÃO PARA PESQUISAS FUTURAS

Os resultados desta pesquisa evidenciaram quantitativamente o quanto as rotas secas da BM resultam em mais impactos ambientais em comparação às rotas úmidas. Entretanto, foi ressaltada a importância do desenvolvimento de técnicas de secagem da biomassa que demandem menor quantidade de energia para diminuir o impacto das rotas secas. Portanto,

sugere-se avaliar técnica-econômica e ambientalmente os diferentes meios de se secar a BM. Ademais, sugere-se, posteriormente à avaliação técnica das formas de secagem, avaliar as características da BM e a sua adequação para a briquetagem. Esse processo é muito interessante para ser aplicado à BM, principalmente pela possibilidade de misturar diferentes biomassas. Dessa forma, a briquetagem pode integrar o gerenciamento de resíduos sólidos (florestais e agrícolas) com a sustentabilidade nas ETARs.

Relacionado à LHT, fontes alternativas precisam ser buscadas para o fornecimento de calor para o reator. Uma possibilidade que precisa ser avaliada é, por exemplo, o uso de biogás gerado em processos anaeróbios em uma ETAR como o combustível para o aquecimento do reator de LHT. Sugere-se avaliar as rotas de aproveitamento da FA resultante da LHT, principalmente a rota modelada (recirculação da FA no cultivo das microalgas). Entretanto, sugere-se avaliar a aplicação de efluente com menor conteúdo de N para a diluição da FA para não ocorrer a inibição do crescimento das microalgas. Dessa forma, evita-se o uso de água para a diluição e se promove o tratamento do efluente utilizado na diluição, além de proporcionar a manutenção de nutrientes em ciclo. Portanto, devem ser feitos testes com a mistura dos efluentes para avaliar a composição química da mistura e também o crescimento das microalgas. Posteriormente, realizar a LHT e verificar a diluição necessária para que a FA possa ser recirculada no cultivo das microalgas. Recomenda-se também integrar o aproveitamento da FG e da FS à LHT e ao tratamento de esgoto. Ademais, questões como o alto teor de cinzas e de N na biomassa precisam ser aprofundadas buscando soluções que possam melhorar a qualidade do bio-óleo para torna-lo mais sustentável.

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APÊNDICES

APÊNDICE A (APPENDIX A).

Supplementary material

Table S1. Potential environmental impacts of briquettes in scenario 1 – Microalgae biomass grown in a high-rate algae pond, by modeled stage and known inputs from the technosphere.

Impact category	Unity	Total	Stage			Input				
			Growth	Drying	Briquetting	CO ₂	Electricity	Water	Nitrogen	Phosphate
Climate change	kg CO ₂ eq	1.0E-01	-1.4E-01	2.4E-01	2.6E-03	1.6E-01	2.8E-01	-2.2E-01	-1.1E-01	-4.8E-03
Stratospheric ozone depletion	kg CFC11 eq	-1.8E-06	-1.9E-06	1.3E-07	1.5E-09	3.7E-08	1.6E-07	-8.8E-08	-1.9E-06	-2.2E-09
Particulate matter formation	kg PM2.5 eq	-3.1E-04	-4.5E-04	1.4E-04	1.6E-06	1.9E-04	1.7E-04	-5.2E-04	-1.3E-04	-1.9E-05
Terrestrial acidification	kg SO ₂ eq	-2.5E-04	-7.9E-04	5.3E-04	6.0E-06	3.4E-04	6.3E-04	-8.2E-04	-3.7E-04	-3.8E-05
Freshwater eutrophication	kg P eq	3.1E-05	-6.1E-06	3.7E-05	4.2E-07	9.3E-06	4.4E-05	-1.4E-05	-4.4E-06	-3.8E-06
Marine eutrophication	kg N eq	5.6E-06	3.3E-06	2.3E-06	2.5E-08	6.1E-06	2.7E-06	-6.6E-07	-2.5E-06	-5.0E-08
Terrestrial ecotoxicity	kg 1,4-DCB	3.5E-01	2.7E-01	7.2E-02	8.0E-04	9.8E-01	8.5E-02	-3.0E-01	-3.9E-01	-3.4E-02
Mineral resource scarcity	kg Cu eq	-8.0E-04	-5.6E-05	4.6E-05	5.2E-07	4.2E-04	2.2E-04	-7.9E-04	-4.5E-04	-2.0E-04
Fossil resource scarcity	kg oil eq	1.4E-02	-4.3E-02	5.7E-02	6.4E-04	2.7E-02	6.7E-02	-5.6E-02	-2.2E-02	-1.5E-03
Water consumption	m ³	-4.7E-01	-4.7E-01	1.3E-03	1.5E-05	7.8E-04	1.6E-03	-4.7E-01	-1.4E-03	-1.0E-04

Table S2. Potential environmental impacts of briquettes in scenario 2 – Microalgae biomass grown in a hybrid reactor, by modeled stage and known inputs from the technosphere.

Impact category	Unity	Total	Stage			Input				
			Growth	Drying	Briquetting	Cotton	Electricity	Water	Nitrogen	Phosphate
Climate change	kg CO ₂ eq	1.9E-01	-4.9E-02	2.4E-01	2.6E-03	1.2E-01	2.6E-01	-1.3E-01	-6.0E-02	-2.4E-03
Stratospheric ozone depletion	kg CFC11 eq	-5.9E-07	-7.2E-07	1.3E-07	1.5E-09	3.8E-07	1.5E-07	-5.0E-08	-1.1E-06	-1.1E-09
Particulate matter formation	kg PM2.5 eq	2.6E-05	-1.2E-04	1.4E-04	1.6E-06	2.5E-04	1.6E-04	-3.0E-04	-7.1E-05	-9.4E-06
Terrestrial acidification	kg SO ₂ eq	3.5E-04	-1.8E-04	5.3E-04	6.0E-06	4.6E-04	5.9E-04	-4.7E-04	-2.0E-04	-1.9E-05
Freshwater eutrophication	kg P eq	3.9E-05	1.3E-06	3.7E-05	4.2E-07	9.7E-06	4.1E-05	-7.9E-06	-2.4E-06	-1.9E-06
Marine eutrophication	kg N eq	1.8E-05	1.6E-05	2.3E-06	2.5E-08	1.7E-05	2.5E-06	-3.8E-07	-1.4E-06	-2.5E-08
Terrestrial ecotoxicity	kg 1,4-DCB	-1.7E-01	-2.4E-01	7.2E-02	8.0E-04	1.5E-01	8.0E-02	-1.7E-01	-2.1E-01	-1.7E-02
Mineral resource scarcity	kg Cu eq	-4.3E-04	-6.1E-04	1.8E-04	2.0E-06	1.7E-04	2.0E-04	-4.5E-04	-2.5E-04	-9.8E-05
Fossil resource scarcity	kg oil eq	4.5E-02	-1.2E-02	5.7E-02	6.4E-04	2.8E-02	6.3E-02	-3.2E-02	-1.2E-02	-7.7E-04
Water consumption	m ³	-2.6E-01	-2.6E-01	1.3E-03	1.5E-05	1.1E-02	1.5E-03	-2.7E-01	-7.6E-04	-5.0E-05

Table S3. Normalized environmental impacts (Ecopoints), which occurred in the life cycle of the modeled base scenarios and the sensitivity analyzes.

Impact category	Normalization factor	Base scenario		Sensitivity analysis (a) reduction of biomass moisture			Sensitivity analysis (b) scenario 1a and 2a with avoided coal briquette	
		Scenario 1	Scenario 2	Scenario 1a	Scenario 2a	Hard coal briquettes (Ecoinvent v3.5)	Scenario 1b	Scenario 2b
Climate change	7.99E+03 kg CO ₂	1.3E-05	2.4E-05	-6,2E-06	6,6E-06	2,1E-06	-8,3E-06	-3,5E-05
Stratospheric ozone depletion	5.99E-02 kg CFC11	-3.0E-05	-9.8E-06	-3,1E-05	-1,1E-05	1,0E-07	-3,1E-05	-1,2E-05
Particulate matter formation	2.56E+01 kg PM2.5	-1.2E-05	1.0E-06	-1,6E-05	-2,3E-06	2,1E-06	-1,8E-05	-2,3E-05
Terrestrial acidification	4.10E+01 kg SO ₂	-6.2E-06	8.7E-06	-1,4E-05	1,1E-06	1,8E-06	-1,6E-05	-4,1E-05
Freshwater eutrophication	6.49E-01 kg P	4.8E-05	6.0E-05	1,2E-05	2,7E-05	6,1E-06	5,9E-06	-1,6E-04
Marine eutrophication	4.61E+00 kg N	1.2E-06	3.9E-06	9,1E-07	3,6E-06	1,3E-08	9,0E-07	3,3E-06
Terrestrial ecotoxicity	1.04E+03 kg 1.4-DCB	3.3E-04	-1.6E-04	2,9E-04	-2,0E-04	1,1E-05	2,8E-04	-2,8E-04
Mineral resource scarcity	1.20E+05 kg Cu	-6.7E-09	-3.6E-09	-7,7E-09	-4,5E-09	1,2E-10	-7,8E-09	-5,7E-09
Fossil resource scarcity	9.80E+02 kg oil	1.5E-05	4.6E-05	-2,2E-05	1,2E-05	2,1E-05	-4,3E-05	-5,4E-04
Water consumption	2.67E+02 m ³	-1.8E-03	-9.8E-04	-1,8E-03	-9,8E-04	2,8E-07	-1,8E-03	-9,8E-04

APÊNDICE B (APPENDIX B).

Supplementary material

Process systems from the ecoinvent library v3.1 (Table S1), used to model the inputs (heat, electricity, chemical inputs), which are the “known input from the technosphere”.

For the choice of the ecoinvent v3.1 process system, global data (code {GLO}) was prioritized for modeling. When the database did not have this inventory, it was decided to use the {RoW} system, an alternative very similar to global data, as it is linked to geographical activities, adjusting uncertainties. Finally, in the absence of both, authors opted for the North American database (code {US}). Specifically, the electricity supply was modeled considering a {US} process system, as there is no production mix in the database {GLO}. Thus, aiming for the potential environmental impacts found in this LCA to have global representation, the North American production mix was adopted. The supply of electricity in this country has about 63% of non-renewable sources, being very similar to the global reality, in which these sources represent approximately 64% of the generated electric energy. For heat supply, fuel oil was considered since this is the most used energy source worldwide [5].

Table S1. Background data included in the life cycle of the modeled bio-oil.

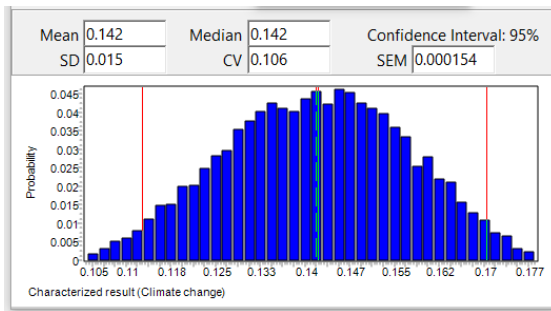
Known input from the technosphere	Documentation
Electricity – Electricity, high voltage {US} production mix.	Life cycle inventories of energy systems, ecoinvent report No. 5 [1].
Heat – Heat, district or industrial, other than natural gas {RoW} heat production, heavy fuel oil, at industrial furnace 1 MW.	Life cycle inventories of energy systems, ecoinvent report No. 5 [1].
Water – Tap water {RoW} tap water production, conventional treatment.	Documentation of changes implemented in ecoinvent v3.1 [2].
Catalyst – Molybdenum trioxide {GLO} production.	Life cycle inventories of chemicals, ecoinvent report No. 8 [3].
Catalyst – Aluminium oxide {GLO} aluminum oxide production.	Life cycle inventories of chemicals, ecoinvent report No. 8 [3].
Hydrotreatment input – Hydrogen, liquid {RoW} hydrogen cracking, Association of Plastics Manufacturers in Europe (APME).	Life cycle inventories of chemicals, ecoinvent report No. 8 [3].
Additive – Ethylene-vinyl acetate copolymer {RoW} production	Life cycle inventories of chemicals, ecoinvent report No. 8 [3].

Note: The {US} code indicates that the inventory was constructed taking into account average values for systems in the United States of America; {RoW} means that inventory has taken into account the “Rest-of-the-World” average data, which are global data excluding Europe; and {GLO} means data with a global average [4].

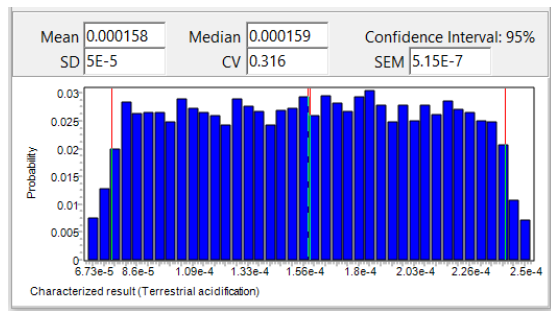
Figure S1 shows the distribution of possibilities of potential environmental impacts in marine eutrophication, climate change, marine ecotoxicity, fossil fossils depletion, human toxicity and terrestrial acidification. An area bounded by the vertical red lines indicates the percentile ranges from 2.5% to 97.5% and the vertical line indicates the average result in the 10,000 with a 95% confidence interval (CI).

Figure S1. Distribution of possibilities of potential environmental impacts.

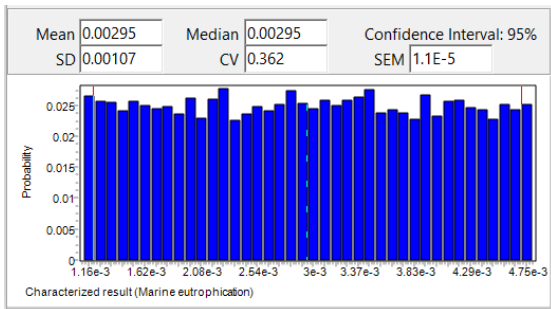
Climate change



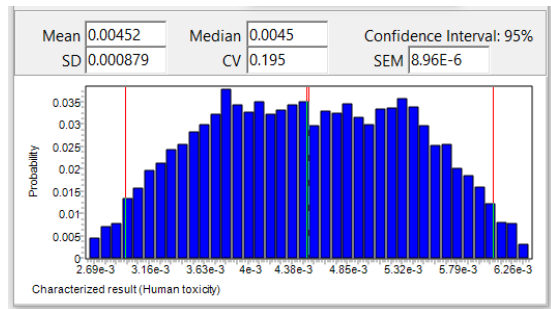
Terrestrial acidification



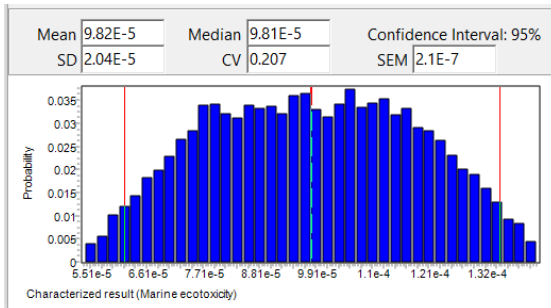
Marine eutrophication



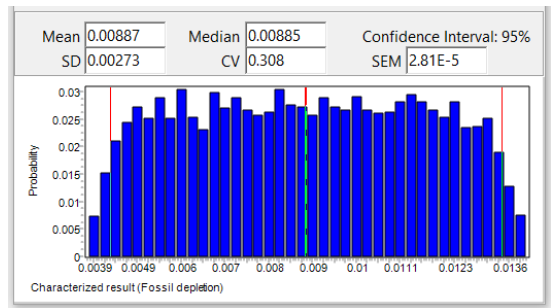
Human toxicity



Marine ecotoxicity



Fossil fossils depletion



References

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