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Life-cycle assessment of marine biofuels from thermochemical liquefaction of different olive residues in Spain

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Abstract

Advanced biofuels from thermochemical liquefaction, such as pyrolysis (PY) and Hydrothermal liquefaction (HTL), of olive residues in the Andalusian region of Spain (specifically in the province of Jaen) can potentially play a crucial role in the reduction of greenhouse gas (GHG) emissions in the maritime sector. In this study, an attributional life-cycle assessment (ALCA) was performed to estimate and compare the GHG emissions for producing marine biofuels via pyrolysis and HTL from olive pomace (COP) and pruning biomass (OTPB), to provide 1 megajoule (MJ) of marine biofuel, as a functional unit. For convenience, the different technology-feedstock combination scenarios are represented as scenario 1 (PY_COP), scenario 2 (PY_OTPB), scenario 3 (HTL_COP), and scenario 4 (HTL_OTPB). The life-cycle GHG emissions of the biofuels were 42.0, 44.1, 22.1, and 32.1 g CO₂-eq/MJ for PY_COP, PY_OTPB, HTL_COP and HTL_OTPB scenarios, respectively, corresponding to 47–73% GHG emissions reduction compared with petroleum fuels. The scenarios were also evaluated based on other impact categories such as Sulphur dioxide in the air, Nitrogen oxides in the air, Particulates in the air, and Non-methane volatile organic compounds (NMVOCs) in the air. The scenarios reduced the SO₂ emissions, Nitrogen emissions, NMVOCs, and particulates in the air by at least 50%, 90%, 20%, and 25% respectively in comparison to fossil fuels. A contribution analysis revealed that olive cultivation and upgrading as hot spots for emission in pyrolysis-based systems. Likewise, HTL conversion and upgrading steps were emitting more emissions for an HTL-based system. Therefore, marine biofuel obtained through the thermochemical conversion of olive residues has better environmental performance on a life cycle basis, with a preference for HTL based system over pyrolysis.

Keywords: Marine biofuels, hydrothermal liquefaction, Pyrolysis, Life-cycle assessment, Greenhouse gas emissions.

1. Introduction

Ships play a central role in the global supply chain of any commodity. About 80% of the world's products are transported by the marine sector. This makes it one of the largest consumers of fossil-based fuels. The total consumption is estimated to be around 330 million metric tons of heavy fuel annually (Hsieh & Felby, 2017). In 2019, Spain consumed approximately 5 million tons of in terms of international maritime bunkering (Eurostat, 2022). Even though maritime transportation emits fewer pollutants per ton kilometer, it contributes to 15% of global carbon emissions. If left unregulated, the emissions from this sector are expected to grow by between 250% in 2050 (Rutherford & Comer, 2018). Decreasing these emissions can therefore greatly contribute to the current

world's environmental challenges. Lignocellulosic biomass-based marine biofuels are seen as a must in reaching these goals because the CO₂ emitted during combustion is in balance with the CO₂ absorbed during the cultivation of biomass. Also, the lower Sulphur and nitrogen content in this type of biomass make them suitable to attain IMO regulations. The residues from the olive sector are one such potential candidate for producing biofuel for the maritime sector. In terms of availability, Spain devotes 2.5 million hectares with more than 180 million trees to this industry. In the province of Jaen, over 91% of the land is occupied by olive trees, making it one of the highest occupants in the entire EU. With accounting for 80% production capacity of olive oil in Spain, the Andalusian region is one of the largest agricultural sectors in the olive- and olive oil industry (UNESCO, 2017). Consequently, the sector produces a large amount of lignocellulosic biomass waste such as olive tree pruning biomass (OTPB), olive pits, leaves, and crude olive pomace (COP). For example, pruning biomass alone has an estimated waste of between 1.5 and 3 tons per hectare in a year (Ruiz et al., 2017). Thereby valorizing these residues will provide an opportunity for developing a bio-economy.

Thermochemical liquefaction, such as pyrolysis, hydrothermal liquefaction (HTL) and gasification, of biomass to bio-crude along with sequential upgrading to biofuel is one of the potential conversion routes for marine biofuel production. Although various studies have been conducted to investigate the thermochemical conversion of olive residues, very few studies have been conducted for biofuel production via HTL based on residues from the olive sector (De Filippis et al., 2016; Evcil et al., 2021). Based on this literature the thermochemical liquefaction of olive residues to produce biofuel is quite promising. With the amount of residues available in the region of Jaen, almost 10% of Spain's national HFO demand can be satisfied. Therefore, apart from techno-economical assessment, analysis based on the environmental aspect is necessary to evaluate the potential of these novel alternatives. Some of the literature also go further beyond the experimental work to evaluate the environmental performance of these methods (Hani & Hailat, 2016; Parascanu et al., 2018). As result indicating better performance in terms of the potential for carbon emission reduction along their life cycle. However, a comparative attributional life cycle analysis is needed to understand and select a suitable conversion pathway for marine biofuel production.

This study's objective is to provide a deeper understanding of the environmental impacts of utilizing olive industry residues, such as COP and OTPB, to biofuels for the maritime sector. To evaluate the environmental performance by quantification of the GHG emissions of these designs by identifying key performance indicators and performing life cycle analysis (LCA). Lastly, this work will serve as a basis for future studies by providing recommendations on improving GHG emission performance. To reach the objective, this thesis is structured to answer the following question: "What are the environmental life cycle impacts of utilizing lignocellulosic residues of the olive sector in Spain to produce marine biofuels via HTL or Pyrolysis?"

2. Methodology

This life cycle analysis (LCA) aims to evaluate the environmental impacts and hotspots of the thermochemical conversion of olive industry residues to produce marine biofuels in Spain. In this LCA, the entire proposed supply chain from cradle to grave is considered, from olive cultivation to the combustion of marine biofuel in ships. In this study, an Attributional LCA (ALCA) is performed. The ALCA is to find information on the global

burdens that can be associated with a product life cycle, it uses cut-off rules and allocation to isolate the production system from the larger system to assess solely the route to marine biofuel. As for geographical coverage, the LCA is focused on the Jaen in the Andalusian region of Spain. The entire chain will be modeled in Spain or Europe. If necessary, global data was used. Figure 1 shows the overall system boundary encompassing all the scenarios. The system includes olive cultivation, primary Virgin olive oil (VOO) extraction, secondary extraction, thermochemical conversions, biofuel upgrading using hydrogen, and final combustion of marine biofuels.

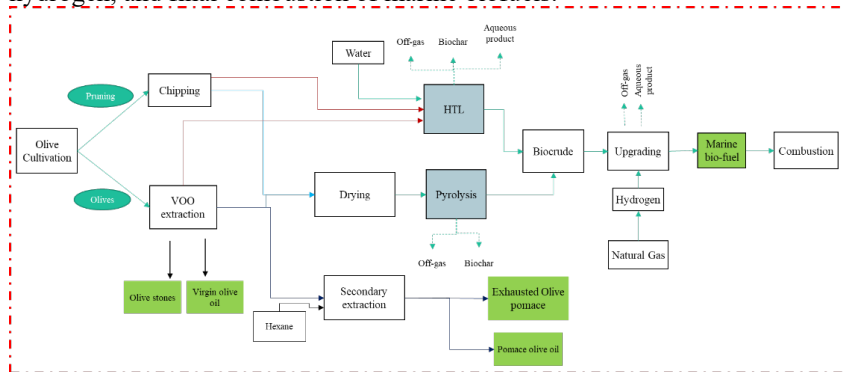


Figure 1: System boundary of scenarios to produce marine biofuels from olive residues in Spain. The green box indicates the products and co-products of the systems, and the blue-grey box indicates the thermochemical pathways. Dotted lines indicate the waste streams.

The analysis follows ISO 14040, the international standard for LCA, and the functional unit is set to be 1 megajoule (MJ) of biofuel produced. All emissions from each process and its associated upstream supply chain were accounted for in this study. But, the emission originating due to the construction of infrastructure, manufacture of equipment, wastewater treatment, biochar, and off-gas utilization were not included within the scope. Data quality and specificity are widely considered key criteria for any LCA studies. Local-specific data such as environmental uniqueness and spatial variation are one of the concerns that require special attention. Hence, the data consistency and accuracy are enhanced by utilizing local data of olive residues obtained from a field visit, e.g., the feedstock availability, the locations of biofuel supply chain nodes, the transportation, and the electricity mix. For the processes modeling and development, e.g., biofuel production and delivery including catalysts, hydro treating and hydrogen production, and nitrogen fertilizer, the GREET 2015 (Greenhouse gases, regulated emissions, and energy use in transportation) or Activity browser coupled with Eco invent v3.7 database was used to model the process emissions with modifications. When the data could not be found in the software database described above, they are collected from peer-reviewed journal articles or reports issued by the government and widely recognized scientific organizations (e.g., IPCC) or laboratories (e.g., PNNL and NREL).

The emissions from each process are obtained based on the physical allocation method. Specifically, mass and energy balances were initially calculated for each process through the Aspen Plus v12 process simulator and then multiplied by the corresponding emission factors. The aspen process model developed includes biomass pretreatment, hydrothermal reactor system, product separation and purification, and byproduct valorization. The off-gas and aqueous stream were recycled within the system. The collected raw data from the various sources described above were first compiled in Microsoft Excel to build the

lifecycle inventory of marine biofuels, and then, IPCC 2007 Global warming potential factors were used to convert CO₂, CH₄, and N₂O into CO₂-eq for a time horizon of 100 years.

3. Results

3.1. GHG emissions from product allocation

The GHG emissions obtained as a result of producing marine biofuel to reach the reference flow of 1 MJ is presented in Figure 2 as g CO₂-eq physically allocated to the fuel production system. Scenarios 1 and 2 emit 42.0 and 44.1 g CO₂-eq/MJ, respectively compared to 83.6 g CO₂-eq/MJ for HFO as calculated for 1 MJ by using the Eco invent database. This is approximately a 50% decrease in emissions. Likewise, Scenarios 3 and 4 even decreased the GHG emissions to approximately 25% of that of HFO with 22.1 and 32.1 g CO₂-eq/MJ. A similar reduction can be observed in terms of kg CO₂-eq/kg fuel where scenarios 1-4 releases 1.6, 1.2, 0.7, and 0.8 kg CO₂-eq/kg fuel compared against 3.42 kg CO₂-eq/kg fuel for HFO. These values are in the range of data reported in the literature (Capaz et al., 2020, 2021; Nie & Bi, 2018; Parascanu et al., 2018; Santos et al., 2018; Tanzer et al., 2019),

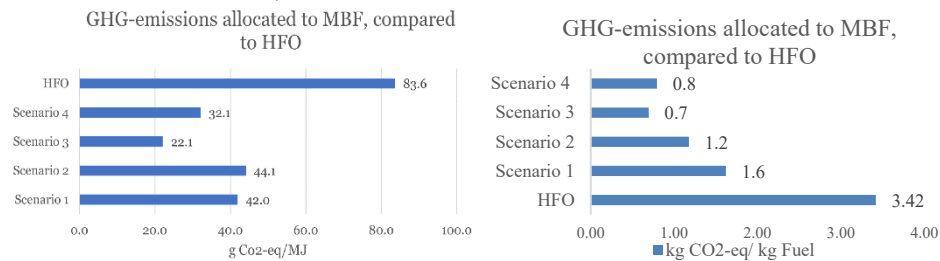


Figure 2: The physically allocated g CO₂-eq/MJ fuel (left) and kg CO₂-eq/kg fuel (right) per scenario compared to HFO fromecoinvent.

3.2. Other impact categories

The environmental performance of the four scenarios considering the other environmental impact criteria is discussed in this section. Figure 3, shows the LCI results for emissions SO₂, NO_x, NMVOC, and particulates <2.5 am, in centigrams per megajoule. This is

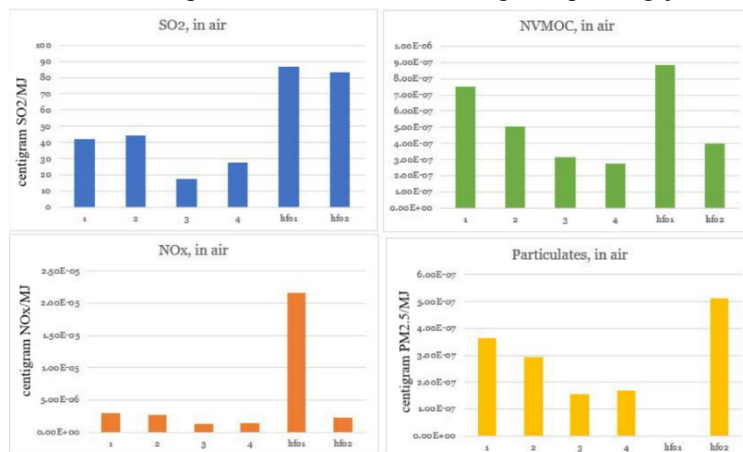


Figure 3: LCI results for emissions to air, compared to HFO from Eco invent and HFO from (Andersson et al., 2016) based on physical allocation

compared to HFO LCA results from the Eco invent database (hfo1) and the LCA results from literature (hfo2) (Andersson et al., 2016). The scenarios reduced the SO₂ emissions, Nitrogen emissions, NMVOCs, and particulates in the air by at least 50%, 90%, 20%, and 25% respectively in comparison to fossil fuels.

3.3. Contribution analysis

To determine the hotspots in the biofuel product systems, a contribution analysis is run through activity-browser, on the allocated model. The figure shows the life-cycle stage-wise GHG emissions of the four biofuel production scenarios. The contribution from main unit processes: Olive cultivation, virgin olive oil extraction, drying and crushing of biomass, HTL or pyrolysis conversion and upgrading, are analyzed.

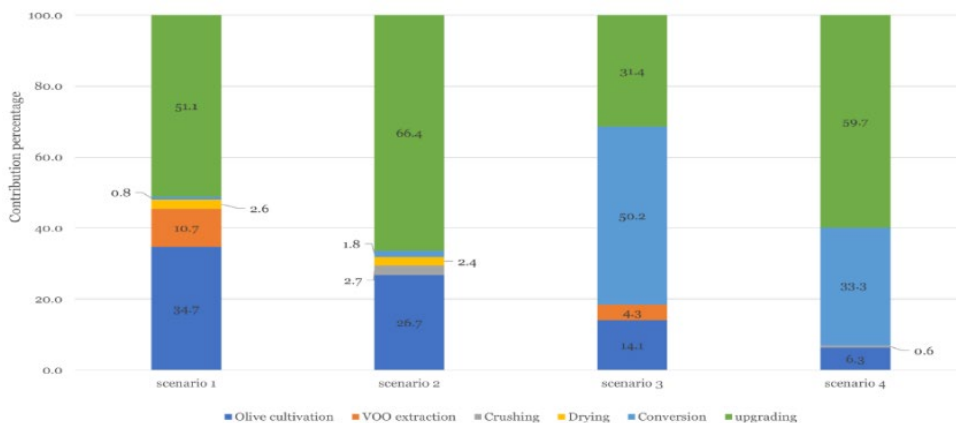


Figure 4: Contribution percentage of the unit processes that contribute to the scenarios

The most dominant contributor of Pyrolysis systems to GHG emissions is bio-crude upgrading, which makes up around 50-60% followed by olive cultivation. The most dominant for HTL_OP is the biomass conversion process, which counts for almost 50%, followed by upgrading with 31.5%. However, an opposite trend was observed for HTL_OTPB with upgrading causing 60% emission followed by conversion. This is might be due to the assumption of similar fuel characteristics of pyrolysis OTPB biofuel and HTL_OTPB biofuel which was made due to data unavailability.

4. Conclusion

This study shows that the product system towards marine biofuel (MBF) compared to HFO has lower allocated GHG emissions. The final GHG emissions from scenarios 1-4 are modeled to be: 42, 44.1, 22.1, and 32.1 g CO₂-eq/MJ, respectively. Which is all a decrease compared to 84 g CO₂-eq/MJ from HFO. Also, the use of MBF reduced the amount of SO₂, particulates, and PMVOC emissions into the air, specifically from 83 g SO₂/MJ (HFO) to 20-40g SO₂/MJ. Therefore, implementing the proposed systems could reduce two of the major environmental impacts of using HFO in marine shipping. Based on the analysis of bio-crude obtained from the thermochemical conversion pathways the nitrogen content was negligible hence corresponding NO_x emissions while combusting the MBF are expected to be meager. Finally, the contribution analysis indicated that the conversion pathways were the hotspots for GHG emission in the scenarios. Thereby optimizing the process will lead to a further reduction in emissions to the environment.

However, sensitivity analysis and system expansion studies are recommended to effectively understand the impact of the scenarios.

References

- Andersson, K., Brynolf, S., Lindgren, J. F., & Wilewska-Bien, M. (2016). Shipping and the Environment: Improving Environmental Performance in Marine Transportation. *Shipping and the Environment: Improving Environmental Performance in Marine Transportation*, 1–426. <https://doi.org/10.1007/978-3-662-49045-7>
- Capaz, R. S., de Medeiros, E. M., Falco, D. G., Seabra, J. E. A., Osseweijer, P., & Posada, J. A. (2020). Environmental trade-offs of renewable jet fuels in Brazil: Beyond the carbon footprint. *Science of the Total Environment*, 714, 136696. <https://doi.org/10.1016/j.scitotenv.2020.136696>
- Capaz, R. S., Guida, E., Seabra, J. E. A., Osseweijer, P., & Posada, J. A. (2021). Mitigating carbon emissions through sustainable aviation fuels: costs and potential. *Biofuels, Bioproducts and Biorefining*, 15(2), 502–524. <https://doi.org/10.1002/bbb.2168>
- De Filippis, P., De Caprariis, B., Scarsella, M., Petruccio, A., & Verdone, N. (2016). Biocrude production by hydrothermal liquefaction of olive residue. *International Journal of Sustainable Development and Planning*, 11(5), 700–707. <https://doi.org/10.2495/SDP-V11-N5-700-707>
- Eurostat. (2022). *Supply, transformation and consumption of oil and petroleum products*. https://ec.europa.eu/eurostat/databrowser/view/NRG_CB_OIL_custom_1359468/bookmark/?lang=en&bookmarkId=57d36982-0e9a-4226-9634-3ee6ead50431
- Evciil, T., Tekin, K., Ucar, S., & Karagoz, S. (2021). Hydrothermal liquefaction of olive oil residues. *Sustainable Chemistry and Pharmacy*, 22(June), 100476. <https://doi.org/10.1016/j.scp.2021.100476>
- Hani, F. F. B., & Hailat, M. M. (2016). Production of Bio-Oil from Pyrolysis of Olive Biomass with/without Catalyst. *Advances in Chemical Engineering and Science*, 06(04), 488–499. <https://doi.org/10.4236/aces.2016.64043>
- Hsieh, C.-W. C., & Felby, C. (2017). *Biofuels for the marine shipping sector*. 86. <http://task39.sites.olt.ubc.ca/files/2013/05/Marine-biofuel-report-final-Oct-2017.pdf%0Ahttps://www.ieabioenergy.com/wp-content/uploads/2018/02/Marine-biofuel-report-final-Oct-2017.pdf>
- Nie, Y., & Bi, X. (2018). Life-cycle assessment of transportation biofuels from hydrothermal liquefaction of forest residues in British Columbia. *Biotechnology for Biofuels*, 11(1), 1–14. <https://doi.org/10.1186/s13068-018-1019-x>
- Parascanu, M. M., Puig Gamero, M., Sánchez, P., Soreanu, G., Valverde, J. L., & Sanchez-Silva, L. (2018). Life cycle assessment of olive pomace valorisation through pyrolysis. *Renewable Energy*, 122, 589–601. <https://doi.org/10.1016/j.renene.2018.02.027>
- Ruiz, E., Romero-García, J. M., Romero, I., Manzanares, P., Negro, M. J., & Castro, E. (2017). Olive-derived biomass as a source of energy and chemicals. *Biofuels, Bioproducts and Biorefining*, 11, 1077–1094. <https://doi.org/10.1002/bbb.1812>
- Rutherford, D., & Comer, B. (2018). *THE INTERNATIONAL MARITIME ORGANIZATION'S INITIAL GREENHOUSE GAS STRATEGY*. https://theicct.org/sites/default/files/publications/IMO_GHG_StrategyFinalPolicyUpdate042318.pdf
- Santos, C. I., Silva, C. C., Mussatto, S. I., Osseweijer, P., van der Wielen, L. A. M., & Posada, J. A. (2018). Integrated 1st and 2nd generation sugarcane bio-refinery for jet fuel production in Brazil: Techno-economic and greenhouse gas emissions assessment. *Renewable Energy*, 129, 733–747. <https://doi.org/10.1016/j.renene.2017.05.011>
- Tanzer, S. E., Posada, J., Geraedts, S., & Ramirez, A. (2019). Lignocellulosic marine biofuel: Technoeconomic and environmental assessment for production in Brazil and Sweden. *Journal of Cleaner Production*, 239, 117845. <https://doi.org/10.1016/j.jclepro.2019.117845>
- UNESCO. (2017). *The Olive Grove Landscapes of Andalusia*. <https://whc.unesco.org/en/tentativelists/6169/>