





Article

Environmental Assessment of Olive Mill Solid Waste Valorization via Anaerobic Digestion Versus Olive Pomace Oil Extraction

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Abstract: Anaerobic digestion is a promising alternative to valorize agrifood wastes, which is gaining interest under an environmental sustainability overview. The present research aimed to compare anaerobic digestion with olive pomace oil extraction, by using life cycle assessment, as alternatives for the valorization of the olive mill solid waste generated in the centrifugation process with a two-outlet decanter from oil mills. In the case of olive pomace oil extraction, two cases were defined depending on the type of fuel used for drying the wet pomace before the extraction: natural gas or a fraction of the generated extracted pomace. The anaerobic digestion alternative consisted of the production of biogas from the olive mill solid waste, heat and electricity cogeneration by the combustion of the generated biogas, and composting of the anaerobic digestate. The life cycle assessment showed that anaerobic digestion was the best alternative, with a global environmental impact reduction of 88.1 and 85.9% respect to crude olive pomace oil extraction using natural gas and extracted pomace, respectively, as fuel.

Keywords: biogas; environmental impact; life cycle assessment; olive pomace; sustainability

1. Introduction

The olive oil industry represents one of the fastest-growing industrial sectors worldwide, being of great importance in the economy of countries, such as Spain, Greece, and Italy, and becoming an important industry in countries, such as Chile, South Africa, or Argentina. The volume of processed olives in the main olive oil producer countries, such as Spain, leads to the generation of circa 4–5 million metric tons of annual waste. The olive mill solid waste is the main waste produced in olive mills that

uses the two-outlet decanter, the most used system for olive oil extraction. Olive mill solid waste is a semi-solid with a high degree of humidity and high organic load [1].

In general, the olive mill solid waste obtained from the two-outlet decanters is transported to the pomace extraction plants to extract the crude pomace oil from them, mainly by extraction with organic solvents (technical hexane) [2]. Before extraction, a drying phase is necessary to reduce the moisture and volatile matter of the olive mill solid waste (between 65 and 75%) to less than 8%. This drying phase involves the highest energy consumption of the whole process of pomace oil extraction, and it is normally fed by natural gas or by the resulting extracted pomace from pomace oil extraction. Of note is that this extracted pomace, once dried, is regarded as an excellent solid biofuel that is currently used in industrial boilers and electric power generation industries [3], thus decreasing the demand for natural gas. In Spain, this energy production at an industrial scale has been possible, thanks to government incentives for the production of electricity from biomass. Notwithstanding, such incentives have been drastically reduced for running plants and have been canceled for new plants. As a result, the economic feasibility of the plants that use extracted pomace as the thermal source for the generation of electrical energy has decreased. Other challenges that pomace oil producers are facing are the fluctuation of olive mill solid waste generation and, mainly, the low commercial value of crude pomace oil [3].

In this context, anaerobic digestion (AD) has been shown to offer a possible solution for the management of the olive mill solid waste [4,5]. AD of olive mill solid waste produces mainly two streams, i.e., methane, as a source of bioenergy, and a stabilized digestate for use in agriculture as fertilizer, avoiding the need to resort to a drying process.

This study aimed to compare the environmental impacts of the two alternatives considered for olive pomace valorization, i.e., (a) AD of the olive mill solid waste, combustion of the generated biogas for heat and electricity production, and dewatering of the digestate for subsequent composting, and (b) extraction of crude pomace oil after drying with natural gas or extracted pomace.

2. Materials and Methods

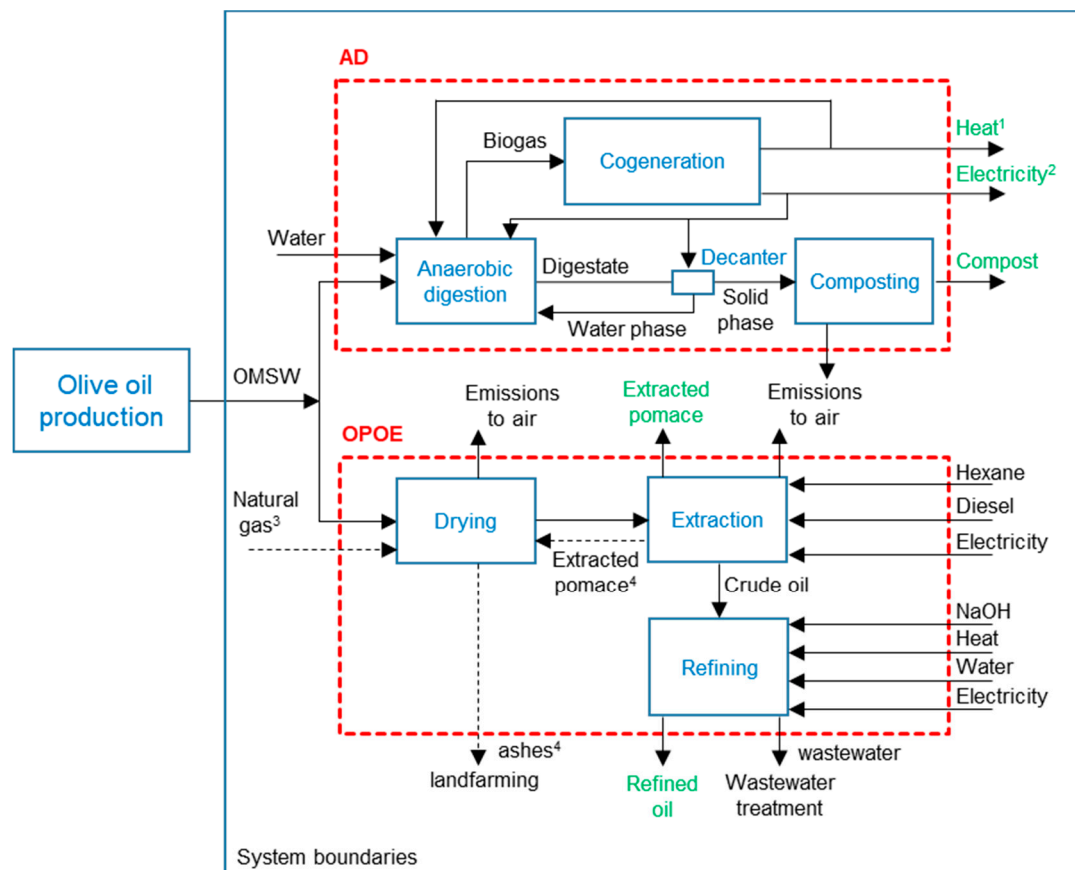
In this study, a comparative attributional life cycle assessment (LCA) was carried out according to the ISO 14040/44 standards [6,7]. The goal and scope, the inventory data, and the impact assessment method used in this study are described in the following sections.

2.1. Goal and Scope of the Study

The main goal was to estimate and compare the life cycle environmental impacts of two alternatives for olive mill solid waste valorization: AD and crude olive pomace oil extraction (OPOE).

The scope of the study was from 'gate to gate'. Figure 1 shows all the foreground and background processes included in the system boundaries for each alternative—AD and OPOE. For the sake of clarity, foreground processes were framed with a dashed line box per each valorization alternative, including (1) for AD: biogas generation in an AD reactor, combustion of the biogas in a cogeneration engine for the production of heat and electricity, dewatering of the digestate in a decanter, and composting of the solid phase from the decanter, and (2) for OPOE: drying of the olive mill solid waste, extraction of the oil from the dried waste, and refining of the crude olive pomace oil. Hence, in the function of the energy pricing policies, producers might prefer burning natural gas and selling the extracted pomace from an economic point of view; two cases for OPOE were considered depending on the source of energy employed for olive mill solid waste drying: natural gas (OPOE-A) and extracted pomace (OPOE-B). The construction and decommissioning of the treatment plants were excluded under the hypothesis that the lifespan of these infrastructures is long enough to assume that the impacts of these stages per functional unit can be considered negligible.

The functional unit was defined as the valorization of 1 metric ton of olive pomace.



¹ Net heat production after subtracting the total heat consumed from the heat produced.

² Net electricity production after subtracting the total electricity consumed from the electricity produced.

³ Only for case OPOE-A (olive pomace oil extraction, drying with natural gas).

⁴ Only for case OPOE-B (olive pomace oil extraction, drying with extracted pomace).

Figure 1. System boundaries.

2.2. Description of the Systems

As shown in Figure 1, the system under study consisted of two alternative pathways for olive mill solid waste valorization. Each of the processes included in the foreground is described below. The reasons for choosing each background process are justified in Section 2.3, concerning inventory data. As a common practice, it was assumed that the olive husks were removed from the olive mill solid waste in the olive mill. The main characteristics of the olive mill solid waste are summarized in Table 1 [1].

Table 1. Olive mill solid waste characterization.

Total solids (g/L)	266 ± 4
Volatile solids (g/L)	250 ± 4
pH	4.97 ± 0.01
Alkalinity (mg CaCO ₃ /L)	6559 ± 5

2.2.1. Anaerobic Digestion (AD)

The first stage of this alternative was the production of biogas via AD of the olive mill solid waste stream in the anaerobic reactor. The AD conditions for olive mill solid waste were based on experimental results obtained in previous research works [1,8]. Water was consumed for dilution

in AD as to reduce the total solids concentration until 9%wt before feeding the reactor [9]. The heat was necessary for keeping the temperature of the digester around 30 °C for mesophilic conditions, and electricity was used for pumping and stirring.

The digestate was dewatered using a decanter. The solid phase was valorized by composting, while the liquid phase was recirculated to the AD reactor to reduce the water consumption for diluting the olive mill solid waste. During composting, moisture was reduced from 65% from the solid phase to 35% in the compost, and NH₃, N₂O, and CH₄ were emitted to air. The compost was sold for its use as organic fertilizer. Direct application of digestate to the soil was not considered since it is a practice that is increasingly limited in the legislation, forcing the implementation of stabilization processes, such as composting, before the reuse of the anaerobic digestate [10].

The generated biogas was combusted in a cogeneration engine in which heat and electricity were simultaneously generated. Both heat and electricity were enough to cover the energy requirement of the rest of the stages of the system—AD (heat and electricity) and decanter (electricity). The surplus energy was sold and fed to the grid.

2.2.2. Olive Pomace Oil Extraction (OPOE)

Firstly, the olive mill solid waste was dried to reduce its humidity in a rotary dryer. As aforementioned, two options were considered depending on the fuel selected to supply the energy required for drying: natural gas (OPOE-A) or extracted pomace resulting from the crude pomace oil extraction stage (OPOE-B). Flue gas was emitted due to fuel combustion. When extracted pomace was used as fuel, ashes were generated and used in landfarming.

The dried olive mill solid waste, or olive pomace, was then subjected to the oil extraction phase. The extraction with an organic solvent, namely, technical hexane, was chosen for this study as it is widely used at industrial scale in the extraction plants. The extracted phase was distilled to remove the solvent from the pomace oil. The recovered solvent was then recirculated to the extraction process. The main intakes for this process were considered in the study: technical hexane, electricity, and diesel for heat production [11]. The emission from diesel combustion and hexane losses were also included within the boundary limits.

Due to its high acidity, the crude pomace oil must be refined. The most employed method for olive pomace oil refining is the chemical refining, in which the crude pomace oil reacts with an alkali solution to neutralize the free fatty acids [12]. Caustic soda was added, forming soap stock by reacting with the fatty acids. A centrifuge was used for separating the oil/soap mixture and, subsequently, the oil from the soap was clarified by filtration. The generated wastewater stream was sent to appropriate treatment. Heat, electricity, water, and sodium hydroxide were considered in the study as needed supplies. Soap stock is not considered as a by-product according to the results reported in [13].

2.3. Inventory Data

The life cycle inventory (LCI) data for both alternatives for olive mill solid waste valorization are detailed in Table 2. All background data were sourced from Ecoinvent v3.3 [14]. The figures shown in Table 2 were calculated based on the data and assumptions summarized below.

Table 2. Inventory data referred to 1 metric ton of olive mill solid waste. na; not applied.

Category	Unit Per Metric Ton of Olive Mill Solid Waste	Anaerobic Digestion (AD)	Olive Pomace Oil Extraction (OPOE-A) ³	Olive Pomace Oil Extraction (OPOE-B) ⁴
Electricity	kWh	−215.06 ¹	2.42	2.42
Heat	MJ	−850.97 ²	0.06	0.06
Diesel	kg	na	1.07	1.07
Natural gas	m ³	na	61.9	na
Compost	kg	221.03	na	na
Refined olive pomace oil	kg	na	22.17	22.17
Extracted olive pomace	kg	na	224.39	82.46
Water	kg	157.75	3652	3652
Technical hexane	kg	na	0.03	0.03
NaOH solution	kg	na	0.11	0.11
Emissions to air				
CO ₂ (fossil)	kg	na	143.07	3.37
CH ₄	kg	1.15	0.91	0.91
N ₂ O	kg	0.05	0.05	0.05
NH ₃	kg	0.25	na	na
Technical hexane		na	0.02	0.02
Olive mill solid waste transportation	tkm	na	100.00	100.00
Wastewater to treatment	m ³	na	3.10	3.10
Ashes to landfarming	kg	na	na	3.78

¹ Net electricity production after subtracting the total electricity consumed from the electricity produced. ² Net heat production after subtracting the total heat consumed from the heat produced. ³ Natural gas employed as fuel for drying. ⁴ A fraction of the extracted olive pomace is employed as fuel for drying.

2.3.1. Anaerobic Digestion (AD)

Anaerobic digester. The ultimate methane production (G_{\max}) obtained from olive mill solid waste in previous work by using biomethane potential tests was 216 cm³ CH₄/g volatile solids (VS) [1]. The biomethane production was obtained then by applying a scale-up factor of 0.85 to this experimental G_{\max} value (216 cm³ CH₄/g VS) [9].

Decanter. The electricity consumed by the decanter for dewatering was 3.5 kW/h per metric ton of digestate [15].

Composting. Emission to air during composting was calculated according to average reported values [16].

Cogeneration engine and energy integration. The energy generation efficiency in a cogeneration biogas engine was considered 33% for electricity and 55% for thermal energy (30% in hot water and 25% in exhausted gas) [17]. The 200 kJ of thermal energy per kg of waste fed to the AD was consumed to keep the operating temperature of the reactor [18]. The electricity consumption in the AD section reached 15% of the electricity generated by the co-generation biogas [19].

2.3.2. Olive Pomace Oil Extraction (OPOE)

Drying and extraction. Data from the literature [11] were adapted to consider both fuel options for drying. Olive mill solid waste needed to be dried until 10%wt humidity. The energy requirement for drying was 2176 MJ/t wet olive mill solid waste [20]. Lower heating value (LHV) of natural gas and extracted oil pomace was 42.4 MJ/kg and 15.33 MJ/kg, respectively [20]. Emissions to air from fuel combustion were calculated using emission factors from the Intergovernmental Panel on Climate Change [21]. Background data from Ecoinvent was used for the use of the ashes in landfarming.

Refining. Inventory data from the Ecoinvent database for the chemical refining of crude vegetable oil were adapted by using an average acidity of 10% for the crude olive pomace oil [12]. Background inventory data for a specific treatment for wastewater from vegetable oil refinery were also included.

2.3.3. Transport

Transport background from the Ecoinvent database was assumed for all materials except for the olive mill solid waste. The AD facility was considered to be located in the same area as the olive mill. In this sense, the transport of the olive mill solid waste to the AD reactor could be despised. The distance from the olive mill to the extraction and refining plant was assumed to be 100 km.

2.3.4. System Expansion Approach

To compare both alternatives for the valorization of olive mill solid waste from the 2-outlet decanter, for which the obtained products were different, system expansion was applied. Each alternative was credited for avoiding the production of products that could be substituted by the different valuable outcomes. The credits were equal to the impacts of the production, by current production processes, of the substituted products. The data for these avoided production systems were sourced from Ecoinvent. Table 3 summarizes the credits associated with substituted products for each valorization alternative.

Table 3. Credits associated with avoided products for each valorization alternative.

Anaerobic digestion		
Outcomes	Credits for avoided products	Equivalence ratio
Electricity	Medium voltage-Spanish mix	1:1 (kwh)
Heat	Industrial heat from natural gas	1:1 (MJ)
Compost	Peat ¹	1:1 (kg) ¹
Olive pomace oil extraction		
Outcomes	Credits for avoided products	Equivalence ratio
Refined olive pomace oil	Refined vegetable oil	1:1 (kg)
Extracted olive pomace	Natural gas	1:1 (MJ) ²

¹ According to [16]. ² LHV (lower heating value): natural gas = 42.4 MJ/kg; extracted olive pomace = 15.33 MJ/kg [20].

2.4. Impact Assessment

SimaPro v.8.3. software from Pré Consultants B.V. (Amersfoort, The Netherlands) was used to model the life cycle. The latest available version of CML 2001 (Centrum voor Milieuwetenschappen, January 2016 version) impact assessment method was used to calculate the environmental impacts [22]. The eleven impact categories included in the CML 2 method were assessed: abiotic depletion potential of elements (ADe), abiotic depletion potential of fossil fuel resources (ADf), global warming potential (GWP), ozone depletion potential (ODP), human toxicity potential (HTP), freshwater aquatic ecotoxicity potential (FWEP), marine aquatic ecotoxicity potential (MWEP), terrestrial ecotoxicity potential (TEP), photochemical oxidants creation potential (POP), acidification potential (AP), and eutrophication potential (EP).

Despite ISO standards do not require normalization and weighting, they are frequently applied in practice to identify important impact categories or to solve tradeoffs between results [23]. In this study, normalization was included to obtain a single score per alternative by using the reference values included in the CML 2001 method, as well as a default weighting factor of one.

3. Results

The environmental impacts of the alternatives considered for olive mill solid waste valorization are shown in Figure 2. A thorough discussion determining the main contributors to each impact category has been addressed in the following section for a better understanding of the environmental differences between both alternatives. To illustrate the origin of the environmental impacts, Figure 3, Figure 4, and Figure 5 for AD, OPOE-A, and OPOE-B, respectively, show the percentage contribution of the different concepts included in the inventory to each impact category, distinguishing between

positive and negative (credits) impacts. Additionally, Figure 6 shows the percentage of contributions to the impacts for the refining of the crude pomace oil (OPOE-A and OPOE-B). In general terms, it is worth noting that most of the credits for AD came from avoiding the external production of electricity. This was in contrast to OPOE, in which the main contributors to the credits depended on the impact category.

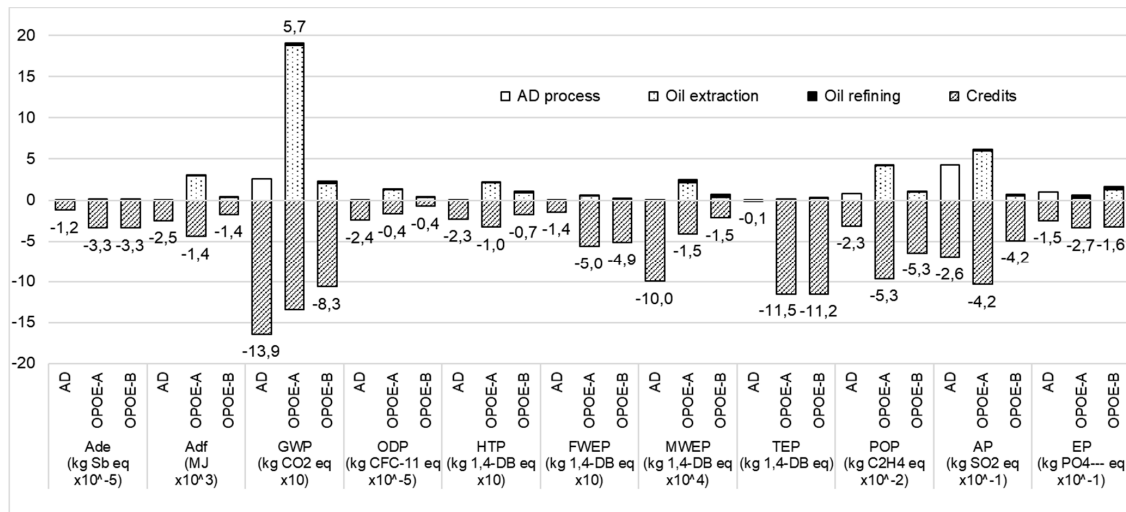
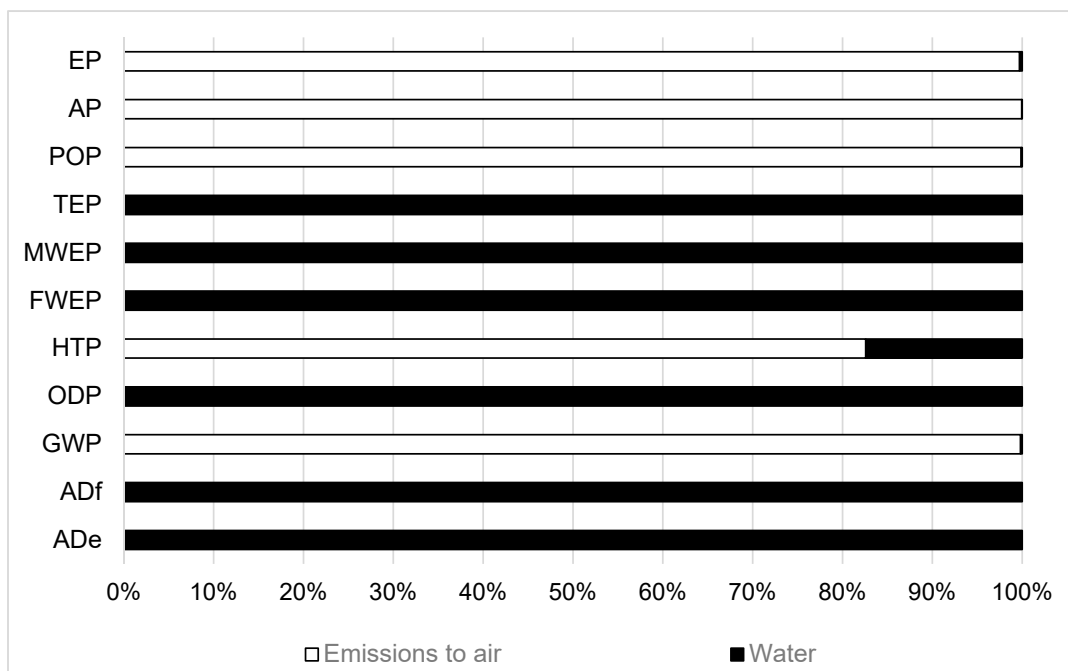
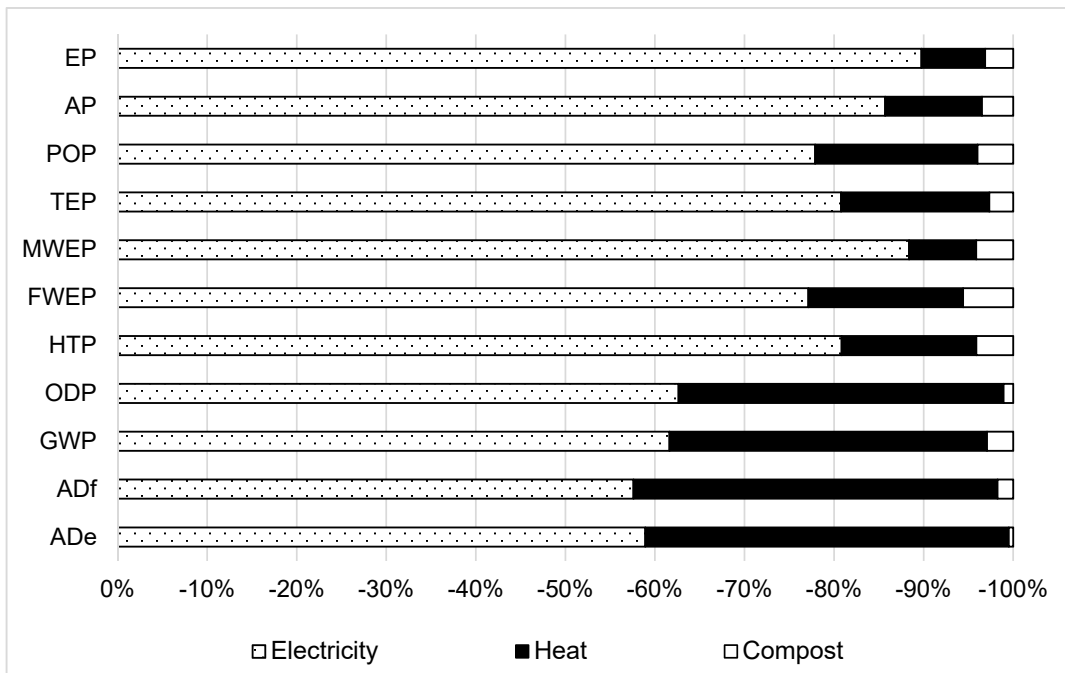


Figure 2. Life cycle environmental impacts of olive mill solid waste valorization via anaerobic digestion and olive pomace oil extraction. AD: anaerobic digestion; OPOE-A: crude olive pomace oil extraction with natural gas as fuel for olive pomace drying; OPOE-B: crude olive pomace oil extraction with extracted pomace as fuel for olive pomace drying. The values shown on top of each bar represent the total impact after the system credits have been applied. Some impacts have been scaled to fit. To obtain the original values, multiply by the factor shown on the x-axis for the relevant impacts.



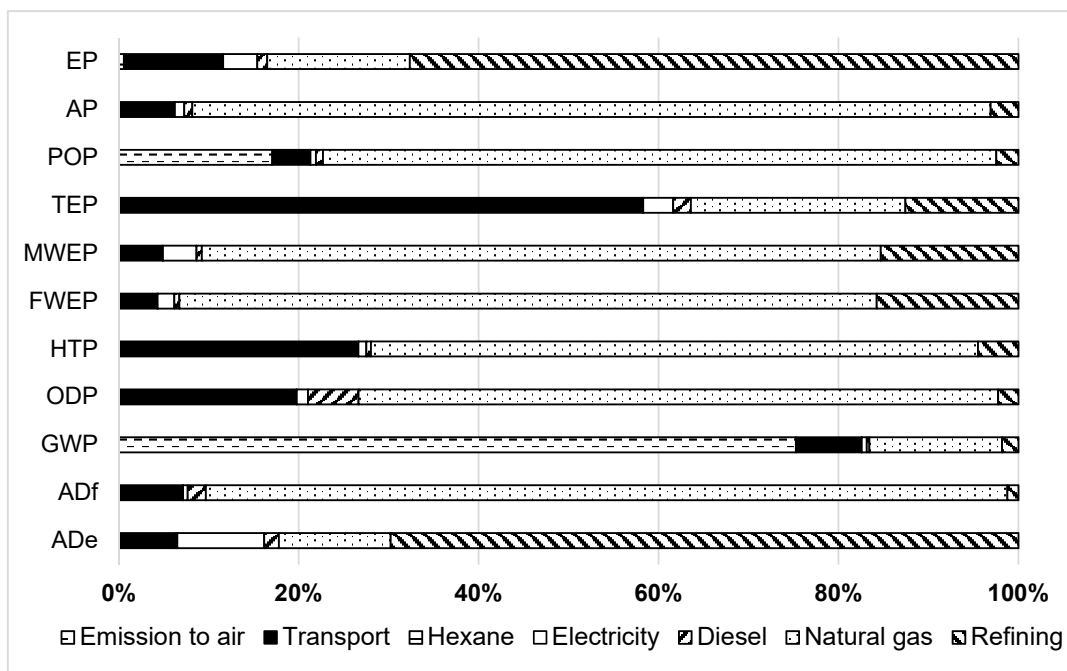
(a)

Figure 3. Cont.



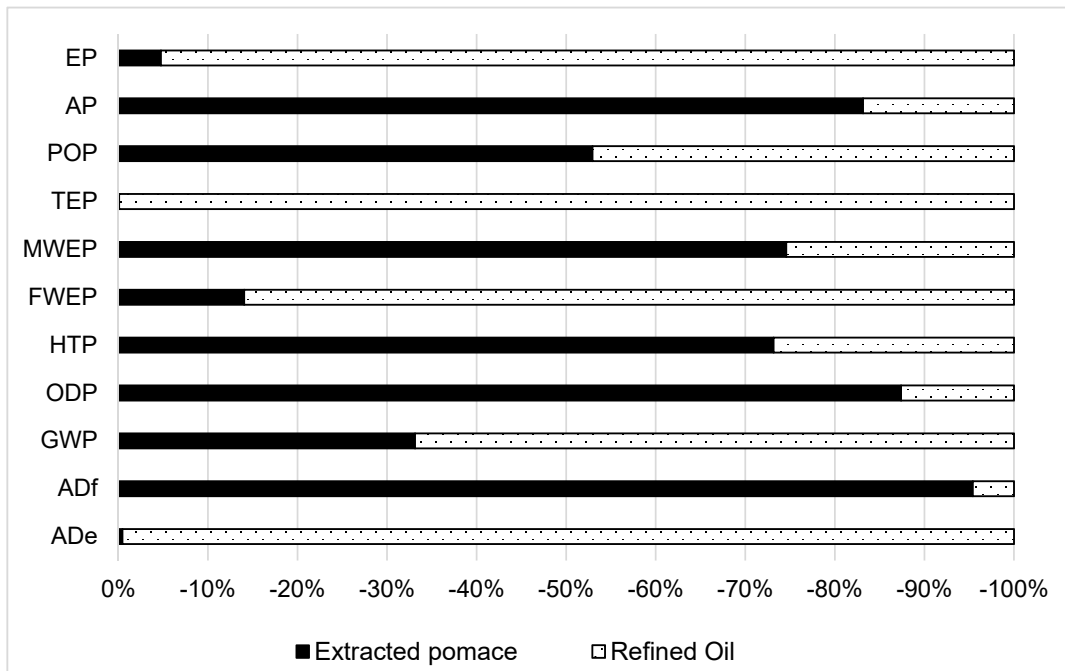
(b)

Figure 3. Percentage contribution to the impacts for anaerobic digestion scheme (AD): (a) positive contribution; (b) credits.



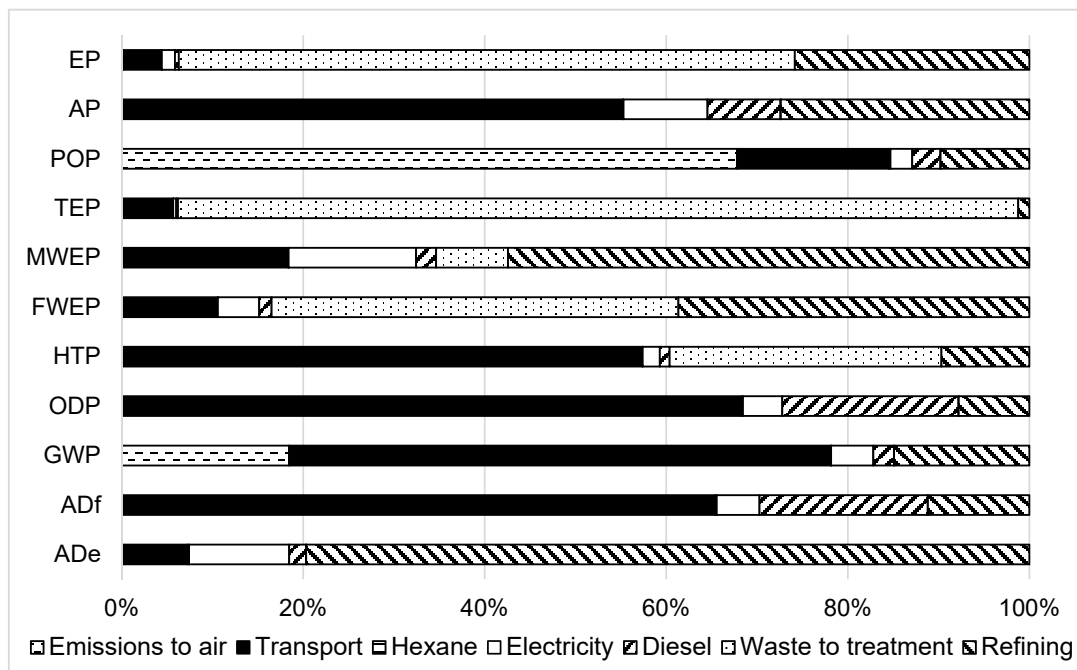
(a)

Figure 4. Cont.



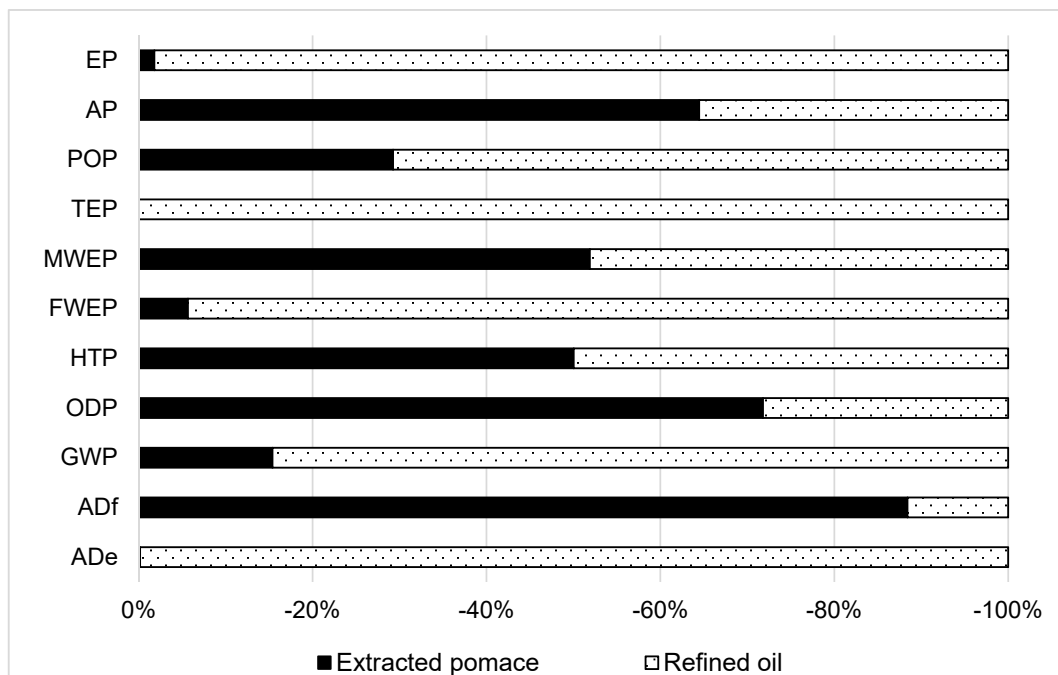
(b)

Figure 4. Percentage contribution to the impacts for crude olive pomace oil extraction, burning natural gas (OPOE-A): (a) positive contribution; (b) credits.



(a)

Figure 5. Cont.



(b)

Figure 5. Percentage contribution to the impacts for crude olive pomace oil extraction, burning a fraction of the extracted olive pomace (OPOE-B): (a) positive contribution; (b) credits.

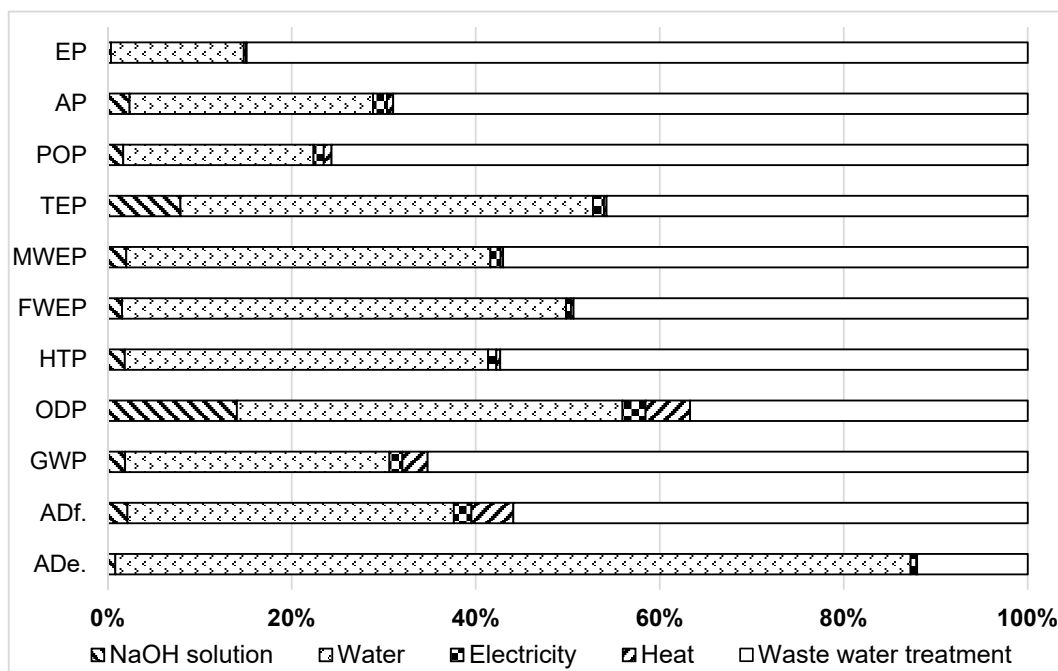
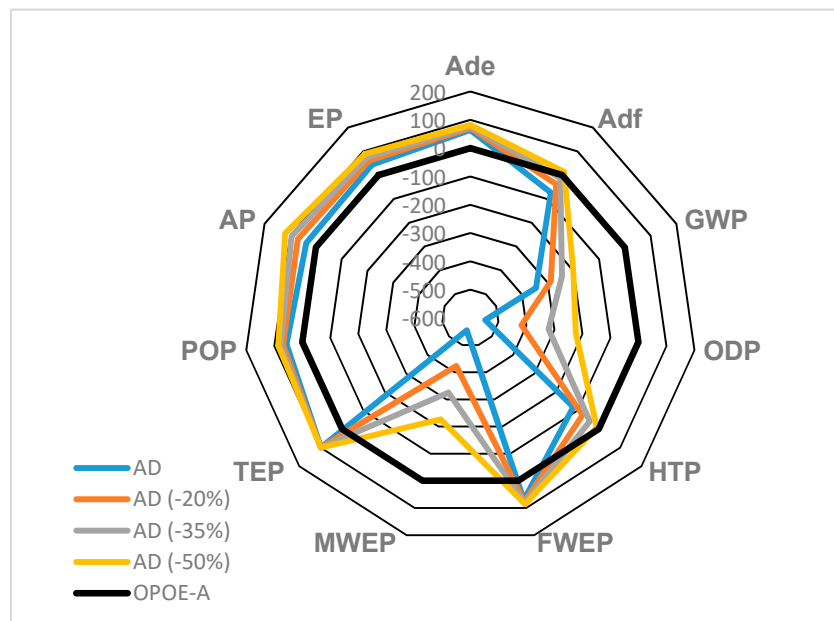


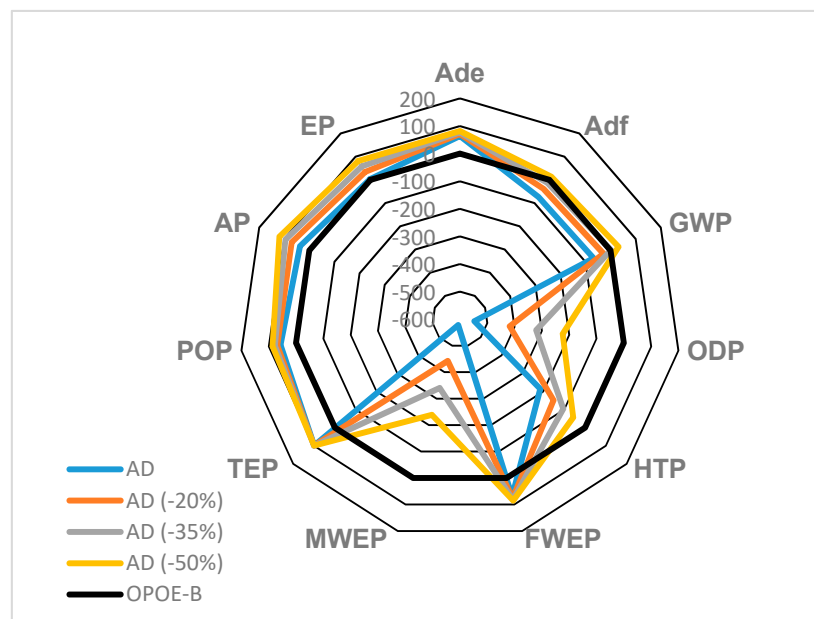
Figure 6. Percentage contribution to the impacts of the crude pomace oil refining (OPOE-A and OPOE-B).

Since AD of olive mill solid waste was still a process at the developing stage and due to the relevance of the credits obtained for this alternative by avoiding electricity production, a sensitivity analysis was performed, varying the amount of biogas generated per kg of treated olive mill solid waste. Relative results obtained for AD, related to OPOE-A and OPOE-B environmental impacts and expressed as percentage increment (+ values) or decrement (– values) are represented in Figure 7,

including cases with a reduction on biogas production of 20, 35, and 50% respect to the experimental value used as a reference.



(a)



(b)

Figure 7. Influence of the reduction of the biogas production from olive mill solid waste in the anaerobic digestion (AD) on the environmental LCA (life cycle assessment) comparison with OPOE employing natural gas (a) and a fraction of the extracted olive pomace (b) as fuel for drying.

Finally, Table 4 illustrates the normalized environmental results and the simple scores, these latter ones by applying a weighting factor of 1.

Table 4. Normalized environmental impacts and single scores per kg of olive mill solid waste, where ADe, abiotic depletion potential of elements; ADf, abiotic depletion potential of fossil fuel resources; GWP, global warming potential; ODP, ozone depletion potential; HTP, human toxicity potential; FWEP, freshwater aquatic ecotoxicity potential; MWEF, marine aquatic ecotoxicity potential; TEP, terrestrial ecotoxicity potential; POP, photochemical oxidants creation potential; AP, acidification potential; and EP, eutrophication potential (EP).

Impact Category	Anaerobic Digestion (AD)	Olive Pomace Oil Extraction (OMOE-A)	Olive Pomace Oil Extraction (OMOE-B)
ADe	-1.40×10^{-13}	-3.89×10^{-13}	-3.89×10^{-13}
ADf	-7.86×10^{-11}	-4.53×10^{-11}	-4.53×10^{-11}
GWP	-2.76×10^{-11}	1.13×10^{-11}	-1.65×10^{-11}
ODP	-2.71×10^{-13}	-4.19×10^{-14}	-4.19×10^{-14}
HTP	-2.92×10^{-12}	-1.33×10^{-12}	-9.27×10^{-13}
FWEP	-2.79×10^{-11}	-9.66×10^{-11}	-9.43×10^{-11}
MWEF	-8.55×10^{-10}	-1.31×10^{-10}	-1.26×10^{-10}
TEP	-2.22×10^{-12}	-2.36×10^{-10}	-2.31×10^{-10}
POP	-2.71×10^{-12}	-6.30×10^{-12}	-6.30×10^{-12}
AP	-9.36×10^{-12}	-1.50×10^{-11}	-1.50×10^{-11}
EP	-1.17×10^{-11}	-2.07×10^{-11}	-1.21×10^{-11}
Single score ¹	-1.02×10^{-9}	-5.41×10^{-10}	-5.48×10^{-10}

¹ Weighting factor = 1.

4. Discussion

4.1. Global Warming

As expected, due to the CO₂ of fossil origin emitted with the flue gas during the drying stage, the valorization of olive mill solid waste via AD could involve a great reduction in GWP (345%) with respect to the pomace oil extraction when natural gas is used as fuel (OPOE-A). This reduction was still significant (67%) when a fraction of the extracted pomace was burned (OPOE-B). Without considering the credits, AD and OPOE-B had similar GWP (23.5–25.5 kg CO₂ eq./t). The difference in GWP was mainly due to the greater credits obtained in AD (165 kg CO₂ eq./t) with respect to OPOE-B (106 kg CO₂ eq./t). The main contributors to these credits were the electricity and the olive pomace oil for oil extraction, respectively. The emissions during composting were the main contributors for AD, whereas the transport of olive mill solid waste to the extraction plant was the main contributor for OPOE-B. As expected, most of the GWP came from the combustion of natural gas for OPOE-A. Even though credits in OPOE-A were higher than those obtained in OPOE-B (135 kg CO₂ eq./t), this could not compensate for the emissions from natural gas combustion. As aforementioned, the GWP of OPOE-A was the only positive impact (57 kg CO₂ eq./t) of all the categories under study.

Assuming a production volume of olive oil in Spain of 1,250,000 metric tons in one regular-season [24] and a ratio of olive mill solid waste to an olive oil of 819:176 kg/kg [25], the application of AD to olive mill solid waste could save the emission of 808 kt CO₂ eq. In this sense, AD could be considered environmentally friendlier, in terms of GWP, than OPOE with biomass combustion (saving just 483 kt CO₂ eq.) and OPOE when natural gas is burned (almost 330 kt CO₂ eq. emission).

This meant that if the olive oil production process could release up to 2.5 kg CO₂eq./dm³ olive oil [26], the valorization of the olive mill solid waste by AD could compensate around 28% of the greenhouse gas emissions from olive oil production, which was far higher than the 17% compensated in the same scenario by applying the crude olive pomace oil extraction (OPOE-B).

4.2. Abiotic Depletion of Elements

Regarding the abiotic depletion of elements (ADe), the contribution of the processes to the impact was negligible compared with the credits for all studied cases. The fuel used for drying did not affect

the impact of the pomace olive oil extraction since almost 100% of the credits were coming from avoiding the production of vegetable oil. Concretely, most of these credits were coming from avoiding the manufacturing of pesticides used in vegetable crops for oil production. These credits made OPOE the best option according to this impact category, being the impact value obtained for AD 64% higher than for OPOE.

4.3. Abiotic Depletion of Fossil Resources and Ozone Layer Depletion

Concerning abiotic depletion of fossil resources (ADf), AD was the best option with a low contribution to the process, limited to the water consumption, and the obtained credits due to the avoiding of the production of electricity from fossil fuels. AD had a 74% lower ADf than OPOE. Both cases for OPOE had the same value of ADf due to the energy equivalence applied between the extracted pomace and the avoided natural gas. The amount of extracted pomace consumed in OPOE-B was equivalent to the natural gas consumed in OPOE-A, reducing in the same quantity the credits obtained in OPOE-B due to the sale of extracted pomace to be used as fuel. The results obtained for ozone layer depletion (ODP) were similar to those achieved for ADf. In this case, the impact of AD was 547% lower than for OPOE.

4.4. Human Toxicity

As aforementioned, AD was the best option for this category. Another remarkable result that requires deeper analysis is that using extracted pomace as fuel was a worse alternative than using natural gas in terms of HTP. Without counting the credits, OPOE-A had a higher impact than OPOE-B due to the contribution of natural gas production. However, in this case, the reduction of the credits in OPOE-B was not equivalent to the reduction in the impacts, as shown for ADf, due to the contribution of the treatment in a landfill of the ash from the extracted pomace combustion.

4.5. Ecotoxicity

OPOE-A had the lowest burdens for freshwater ecotoxicity (FWEP) and terrestrial ecotoxicity (TEP), followed by OPOE-B. The credits from avoiding the production of vegetable oil were responsible for these results. Nevertheless, OPOE had the highest impact on marine water ecotoxicity (MWEP) regardless of the fuel used for drying due to the higher credits assigned to AD from electricity production.

4.6. Photochemical Oxidation (POP) and Acidification (AP)

OPOE was the best alternative for photochemical oxidation and acidification. The value of the impact did not depend on the fuel chosen for drying for the same reason explained in Section 4.3 for ADf and ODP.

For POP, without credits, the process for OPOE had always a higher impact than the AD due to the contribution of the production of natural gas (OPOE-A) or the treatment of the ashes from the extracted pomace combustion (OPOE-B). Nevertheless, these impacts were mainly compensated by the credits obtained from avoiding the use of pesticides in vegetable crops for oil production. Conversely, concerning AP, AD had higher burdens than OPOE-B when credits were not considered. The contribution of the emissions to air from composting could be responsible for this fact.

4.7. Eutrophication

OPOE-A showed the lowest burdens for eutrophication potential (EP), 42% lower than OPOE-B, which was the second-best option. The higher impacts of OPOE-B and AD were caused by the ash treatment and the emissions to air during composting, respectively.

4.8. Sensitivity Analysis

As could be seen in Figure 7, for a reduction of 50% in the production of biogas from olive mill solid waste, remarkable changes were observed in ADf and GWP. On the one hand, for ADf, OPOE turned to be a better option than AD regardless of the fuel used for drying. OPOE-B could be regarded as a better option than AD in terms of GWP.

4.9. Normalization

According to the normalized results and applying a weighting factor of 1, AD was the best alternative with a global environmental impact reduction of 85.9–88.1% with respect to both OPOE options. Within a circular economy approach, the use of natural gas (OPOE-A) was the worst option, but the use of a fraction of the extracted pomace as fuel for drying (OPOE-B) offered a reduction of only 1.2% of the environmental impact, expressed as a single score, with respect to the case of using natural gas (Table 4).

5. Conclusions

Evaluating each category separately, AD was shown as the best alternative for GWP and the other four categories, including ADf, ODP, HTP, and MWTP. The use of extracted pomace as fuel (OPOE-B) instead of natural gas (OPOE-A) could strongly reduce GWP but, conversely, increase the impact in the other two categories, i.e., HTP and EP, leaving the rest without change or with a negligible increment (FWTP, TEP). More specifically, the refining process of crude oil had a very low contribution compared to the extraction process. After evaluating the three alternatives with normalized results and applying a weighting factor of 1, AD showed a global environmental impact reduction of 88.1 and 85.9% with respect to crude olive pomace oil extraction using natural gas and extracted pomace as fuel, respectively.

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References

- Serrano, A.; Feroso, F.G.; Alonso-Fariñas, B.; Rodríguez-Gutiérrez, G.; Fernandez-Bolaños, J.; Borja, R. Olive mill solid waste biorefinery: High-temperature thermal pre-treatment for phenol recovery and biomethanization. *J. Clean. Prod.* **2017**, *148*, 314–323. [CrossRef]
- García Martín, J.F.; Cuevas Aranda, M.; Feng, C.-H.; Álvarez Mateos, P.; Torres García, M.; Sánchez Villasclaras, S. Energetic valorisation of olive biomass: Olive-tree pruning, olive stones and pomaces. *Processes* **2020**, *8*, 511. [CrossRef]
- Olimerca. Cunde el Pesimismo Entre Los Orujeros. Available online: <https://www.olimerca.com/noticiadet/cunde-el-pesimismo-entre-los-orujeros-/e49e04531bc5c2f12effc6f6c2f30ed8> (accessed on 15 October 2019).
- Rincón, B.; Feroso, F.G.; Borja, R. Olive Oil Mill Waste Treatment: Improving the Sustainability of the Olive Oil Industry with Anaerobic Digestion Technology. In *Olive Oil—Constituents, Quality, Health Properties and Bioconversions*; Boskou, D., Ed.; InTech: London, UK, 2012.
- Serrano, A.; Feroso, F.G.; Alonso-Fariñas, B.; Rodríguez-Gutiérrez, G.; López, S.; Fernandez-Bolaños, J.; Borja, R. Performance evaluation of mesophilic semi-continuous anaerobic digestion of high-temperature thermally pre-treated olive mill solid waste. *Waste Manag.* **2019**, *87*, 250–257. [CrossRef] [PubMed]

6. ISO 14040. *Environmental Management—Life Cycle Assessment: Principles and Framework*; International Organization for Standardization: Geneva, Switzerland, 2006.
7. ISO 14044. *Environmental Management—Life Cycle Assessment: Requirements and Guidelines*; International Standardization Organization: Geneva, Switzerland, 2006.
8. Borja, R.; Rincón, B.; Raposo, F. Anaerobic biodegradation of two-phase olive mill solid wastes and liquid effluents: Kinetic studies and process performance. *J. Chem. Technol. Biotechnol.* **2006**, *81*, 1450–1462. [[CrossRef](#)]
9. Keith, F.; Tchobanoglous, G. *Handbook of Solid Waste Management*, 2nd ed.; McGraw-Hill: New York, NY, USA, 2002; ISBN 0071500340.
10. Hudcová, H.; Vymazal, J.; Rozkošný, M. Present restrictions of sewage sludge application in agriculture within the European Union. *Soil Water Res.* **2019**, *14*, 104–120. [[CrossRef](#)]
11. Figueiredo, F.; Marques, P.; Castanheira, É.G.; Kulay, L.; Freire, F. Greenhouse gas assessment of olive oil in Portugal addressing the valorization of olive mill waste. In Proceedings of the Symbiosis International Conference, Athens, Greece, 19–21 June 2014.
12. Sánchez Moral, P.; Ruiz Méndez, M.V. Production of pomace olive oil. *Grasas y Aceites* **2006**, *57*, 47–55. [[CrossRef](#)]
13. Schneider, L.; Finkbeiner, M. Life Cycle Assessment of EU Oilseed Crushing and Vegetable Oil Refining. *Tech. Rep.* **2013**, *2013*, 1–59.
14. Moreno Ruiz, E.; Lérová, T.; Reinhard, J.; Valsasina, L.; Bourgault, G.; Wernet, G. *Documentation of Changes Implemented in Ecoinvent Database 3.3*; Ecoinvent: Zurich, Switzerland, 2016.
15. Tampio, E.; Marttinen, S.; Rintala, J. Liquid fertilizer products from anaerobic digestion of food waste: Mass, nutrient and energy balance of four digestate liquid treatment systems. *J. Clean. Prod.* **2016**, *125*, 22–32. [[CrossRef](#)]
16. Saer, A.; Lansing, S.; Davitt, N.H.; Graves, R.E. Life cycle assessment of a food waste composting system: Environmental impact hotspots. *J. Clean. Prod.* **2013**, *52*, 234–244. [[CrossRef](#)]
17. Cano, R.; Nielfa, A.; Fdz-Polanco, M. Thermal hydrolysis integration in the anaerobic digestion process of different solid wastes: Energy and economic feasibility study. *Bioresour. Technol.* **2014**, *168*, 14–22. [[CrossRef](#)]
18. Gonzalez, A. Environmental, Energetic and Economic Fesiability of the Biomethanization of Wastes from the Agri-Food Industry in Extremadura. Ph.D. Thesis, Universidad de Extremadura, Badajoz, Spain, 2014.
19. Angelidaki, I.; Schmidt, J.E.; Karakashev, D.B. *A Sustainable Solution for Pig Manure Treatment: Environmental Compliance with the Integrated Pollution Prevention and Control Directive*; CORDIS: Nicosia, Cyprus, 2006.
20. *Potencial Energético de los Subproductos de la Industria Olivarera en Andalucía*; Regional Government of Andalusia: Seville, Spain, 2010.
21. Intergovernmental Panel on Climate Change. Emissions Factors Database. Available online: <https://www.ipcc-nggip.iges.or.jp/EFDB/main.php> (accessed on 7 August 2019).
22. Guinée, J.B.; Gorrée, M.; Heijungs, R.; Huppes, G.; Kleijn, R.; Wegener Sleeswijk, A.; Udo De Haes, H.A.; de Bruijn, J.A.; van Duin, R.; Huijbregts, M.A.J. *Life Cycle Assessment: An Operational Guide to the ISO Standards*; Kluwer Academic Publishers: Dordrecht, The Netherlands, 2001; ISBN 1-4020-0228-9.
23. Pizzol, M.; Laurent, A.; Sala, S.; Weidema, B.; Verones, F.; Koffler, C. Normalisation and weighting in life cycle assessment: Quo vadis? *Int. J. Life Cycle Assess.* **2017**, *22*, 853–866. [[CrossRef](#)]
24. Mercacei. Cunde el Pesimismo Entre Los Orujeros. Available online: <https://www.mercacei.com/noticia/47472/actualidad> (accessed on 5 July 2019).
25. Fernández, J. *Los Residuos de las Agroindustrias Como Biocombustibles Sólidos*; Vida Rural: Madrid, Spain, 2006; Volume 233, pp. 14–18.
26. Olivar del Sur. El Olivar, Importante Aliado del Medio Ambiente. Available online: <https://olivadelsur.com/es/blog/el-olivar-importante-aliado-del-medio-ambiente-b100.html> (accessed on 5 July 2019).

