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Waste-to-Energy innovative system: assessment of integrating anaerobic digestion and pyrolysis technologies

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Abstract

Anaerobic digestion (AD) of sewage sludge is an already well-developed technology to convert waste to bioenergy and digestate. Despite its deployment, AD process still raises environmental, technical and economic issues and many researches have been done to improve the process yield, reduce environmental burdens from digestate application and allow a better distribution of energy. The integration of thermal processes with anaerobic digestion is explored to overcome some of the anaerobic digestion limitations. In this study, a cradle-to-grave life cycle assessment (LCA) is performed to compare a system integrating anaerobic digestion and pyrolysis (PY) with single anaerobic digestion, for the co-digestion of sewage sludge and quinoa residue. The results suggest that the environmental impacts of the two pathways are quite similar except for three impact categories: single anaerobic digestion is the best scenario in terms of global warming potential (-769 vs. -604 kg CO_2 eq. /t substrate), ozone depletion (-33.7 vs -2.11 mg CFC-11 eq. /t substrate) and fossil resources use (-9,900 vs -6,900 MJ/t substrate). The multifunctional process could be a viable competitor to single anaerobic digestion by improving pyrolysis products upgrading. The application of liquid digestate to crop field revealed to have significant burdens to particulate matter, acidification and terrestrial eutrophication impact categories and must be carefully monitored in both pathways.

Keywords:
Sewage Sludge, Quinoa, Life Cycle Assessment, Energy, Multifunctional process

Abbreviations[1]

1. Introduction

According to the last report of the Intergovernmental Panel on Climate Change (IPCC, 2021), the increase of greenhouse gases (GHG) concentrations since 1750, as well as the warming of the atmosphere, water, and land, are unequivocally caused by human activities. To tackle climate change, 196 parties adopted the Paris Agreement in 2015 to limit global warming to well below 2, preferably to 1.5 degrees Celsius, compared to pre-industrial levels (UNFCCC, United Nations Framework Convention on Climate Change, 2015). To limit global warming, one option is the replacement of fossil-based resources by waste or biomass. The development of a circular economy based on waste recovery has already been studied and embodies a major issue for future researches (Belaud et al., 2021; Li et al., 2022). In this study, the focus is on sewage sludge from wastewater treatment plant

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[1] Abbreviations
AD, Anaerobic Digestion; CHP, Combined Heat and Power; DM, Dry Matter; EF, Environmental Footprint; FU, Functional Unit; GHG, Greenhouse Gas; GWP, Global Warming Potential; HRT, Hydraulic Retention Time; IPCC, International Panel on Climate Change; LCA, Life Cycle Assessment; LCC, Life Cycle Costing; LCI, Life Cycle Inventory; LCIA, Life Cycle Impact Assessment; PY, Pyrolysis; VFAs, Volatile Fatty Acids; WWTP, Wastewater Treatment Plant

(WWTP). Depending on country's regulations and sludge type (industrial vs urban), the fate of sludge either represents a load, with economic costs, or a recoverable product for its agronomic properties. Even when land spreading is allowed, new legislation is becoming increasingly strict regarding the presence of heavy metals, pathogens and pharmaceutical products (Legifrance, 2020). Some alternatives for sewage sludge treatment exist in order to make them compliant to new legislation: these are stabilization processes (Yoshida et al., 2013). An already known option for stabilization is to send sewage sludge to anaerobic digestion (AD) unit. This waste-to-energy technology is based on the degradation of organic matter by microorganisms in the absence of oxygen, leading to the formation of biogas, a gas mixture saturated with water, composed mainly of methane (CH_4) and carbon dioxide (CO_2) and of digestate, a nutrient-rich co-product (Monlau et al., 2015a). Degradation mechanism in AD occurs in four steps: hydrolysis, acidogenisis, acetogenesis and methanogenesis (Caruso et al., 2019).

The methane - contained in the biogas – can be upgraded in different ways. The most common valuation method is cogeneration to produce heat and electricity, but other alternatives are gaining ground: injection into the natural gas network after purification, utilisation in boiler or biofuel production. As for the digestate, it generally follows a mechanical phase separation to recover both liquid and solid phases that have complementary agronomic properties: a fertilizing fraction, rich in ammoniacal nitrogen and potassium and an amending fraction, rich in organic matter. In France, there were already 76 units of AD for the treatment of sewage sludge from WWTP in 2020 (ADEME, 2021). In Morocco, about ten units of AD for sludge treatment were counted (Ollivier, 2017).

Although AD has greatly developed for decades as an answer to produce energy without consuming fossil resources, its implementation still raises some limitations. First, it is recognized that the process has operational instability and low carbon conversion, as microorganisms struggle to degrade some biomass components, leading to a digestate still containing a significant amount of energy (Pecchi and Baratieri, 2019). It is also highlighted that the digestate produced is often of insufficient quality, due to a low retention capacity of its nutrients, occurring in groundwater leaching and air emissions, especially during storage period (Monlau et al., 2015a; Pecchi and Baratieri, 2019). Lastly, although some of the energy produced in biogas cogeneration could be reused for the AD system, a major part of the heat can be lost if the digester has a restrictive localisation (without an outlet nearby). It is therefore fundamental for project holder to resolve these different issues before implementing the AD.

Improvements are under study to obtain a better methane yield. For instance, co-digestion of sewage sludge with lignocellulosic component can overcome the constraints of a recalcitrant substrate. It has been reported that co-digestion of manure, or livestock, with agricultural residues increases biogas production and improves the stability of the process (González et al., 2020). In Morocco, quinoa cultivation is developing ("A new culture that is revolutionizing Moroccan agriculture [in French]," 2020), it therefore seems possible to use the waste of this pseudo-cereal in co-digestion with sewage sludge. Besides, it has been reported that quinoa could be a suitable biomass for AD (Pabón-Pereira et al., 2020).

The fact that digestate still contains an interesting amount of energy and that land spreading is not always possible has led to research on how to enhance its potential and reduce the drawbacks of AD. Thermal processes have notably been investigated as pre or post treatments for AD, such as incineration (Rajaeifar et al., 2015), gasification (Guo et al., 2021), hydrothermal carbonization, and pyrolysis (Mills et al., 2014; Monlau et al., 2015a; Opatokun et al., 2017; Pecchi and Baratieri, 2019). Pyrolysis (PY) is receiving increasing attention (Cusenza et al., 2021) as an approach for the conversion of biomass into useful bioenergy (bio-oil and syngas) and pyrolytic residue (called biochar). It occurs in an inert atmosphere, in which organic matter is decomposed under the effect of heat (between 350 °C and 650 °C). Among the outputs of pyrolysis, bio-oil has various applications : it can be combusted in boilers and furnaces to recover energy, it can be used for chemicals production as it contains value added constituents and it could be upgraded into biodiesel (Xiu and Shahbazi, 2012). Biochar is also of particular interest due to its ability to act as a carbon sink. Indeed, biochar is composed of highly stable carbon and this stability determines its long term C-sequestration capacity (Monlau et al., 2016). Biochar could also be used as soil amendment to improve soil quality and crop yield, and its potential would be enhanced if combined with a fertilizer (Hossain et al., 2010).

The co-digestion of sewage sludge with quinoa residue integrated with the pyrolysis of solid digestate is at the core of this study. The aim is to compare single AD with integration of AD and PY from an environmental point of view, using the LCA method. LCA is a standardized method for measuring the environmental impacts associated with each life cycle stage of products or systems (International Organization for Standardization (ISO, 2006a, 2006b). Thanks to a systemic approach, an international effort of standardization and the scientific maturity resulting from a large volume of research and the development of applications and databases, the LCA method has become a powerful tool for impact assessment.

2. Literature review

AD has already been the subject of many LCA studies: Mayer et al. (2019) reviewed 315 LCA of waste-to-energy systems and found that 45% of them evaluated AD. Among these AD studies, 38 considered sludge or slurry as entering waste. As for pyrolysis, 10% of the analysed studies were concerned with this technology (Mayer et al., 2019). In addition, it has been reported that LCA is commonly used to evaluate the potential environmental impact of WWTP, including the treatment of sewage sludge by different technologies among which AD appears to be environmentally interesting (Corominas et al., 2013; Pradel et al., 2016; Yoshida et al., 2013). Thus, there are many publications on LCA of sludge AD but comparison of these studies is not always possible. Indeed, Tarpani et al. (2020) revealed that although several LCA studies compare the impacts of different sludge treatment pathways, they widely vary in scope, methodology, the choice of functional unit and the impacts considered (Tarpani et al., 2020).

To the authors' knowledge, there are few LCA studies that address the coupling of anaerobic digestion and pyrolysis for biomass treatment. Mayer et al. (2019) reviewed LCA of waste-to-energy technologies and concluded that there were too little studies on cascaded technologies (Mayer et al., 2019). However, the very recent appearance of new papers demonstrates the growing interested for this kind of integrated system for waste treatment (Havukainen et al., 2022; Li et al., 2022). The following results have been found in the literature for the combination of AD and PY. It appeared that integrated system had higher apparent energy efficiency (71.4 %) than single pyrolysis (60.4%) (Cao and Pawłowski, 2012) and that both system achieved energy and GHG emissions benefits from a Life Cycle Assessment (LCA) perspective, with better performance for the coupling of AD with PY for sewage sludge treatment (Cao and Pawłowski, 2013). Similarly, it was established that the hybrid system of AD combined with PY led to a net 42% increase in electricity production compared to single AD and that the heat surplus from AD could cover solid digestate drying (Monlau et al., 2015a). Wang et al. (2021) used the LCA method to evaluate four treatment pathways for organic fraction of municipal solid waste, and found that the AD+PY combination was the most environmentally friendly compared to AD, PY and PY+AD. The same result was obtained using food waste as feedstock (Opatokun et al., 2017). However, some authors obtained different results when comparing AD, PY and integrated system. It was found that AD had advantages over PY and combined processes, except for sludge with a high organic content for which AD + PY pathway is a good alternative (Li and Feng, 2018). These integrated systems are recent and there is an obvious need to assess their technical, economic and environmental sustainability.

Some LCA studies of AD and PY integration have already been carried out, but the novelty of this study is twofold: the substrate of AD is a mixture of sludge and quinoa residue (co-digestion) and several impact categories are evaluated, not only climate change issue.

3. Methods

3.1 Description of the systems

Most of the experimental data used in this study were obtained from pilot scale experiments (Tayibi, 2021) and extrapolated to industrial scale for a digester of 1500 m^3. Quinoa residue have been collected in a private farm in Ben Guerir, Morocco, while sewage sludge have been taken from WWTP located in Lescar, France. Pre-treatment for quinoa stalks consists in grinding, while sludge is centrifuged secondary sludge and does not require pre-treatment. In both scenarios, sewage sludge and ground quinoa residue are mixed with water before entering AD to achieve a dry matter (DM) content of 10%. This DM content is characteristic of wet AD. The process temperature is of 39°C (mesophilic AD), with a hydraulic retention time (HRT) of 45.2 days.

After 45.2 days of fermentation, biogas and digestate are recovered from AD. The biogas, composed of 57.8% of CH_4 and 42.2 % of CO_2, is delivered to a combined heat and power unit (CHP), assumed to have an electrical efficiency of 38% and a thermal efficiency of 50%, while digestate is separated by screw press, to obtain liquid (3.65 % DM) and solid phases (17 % DM). In the first scenario, liquid and solid digestate are respectively used as fertiliser and soil amending products.

In the second scenario, the solid digestate is dried to reach a DM content of 92% before entering pyrolysis. The pyrolysis heating rate is of 10°C / min, which is characteristic of a slow pyrolysis. Slow pyrolysis is more favourable to biochar, while fast pyrolysis generates a greater amount of bio-oil (Monlau et al., 2015b). Residence time is of one hour, with a maximal temperature equal to 500°C. To ensure an oxygen free environment, the furnace is purged with N_2 before the experiment. The collected products are biochar, bio-oil and syngas, with yields of 40%, 36% and 24%, respectively. It is supposed that the syngas is delivered to the same CHP unit as the biogas. The bio-oil is also sent to a CHP unit, with a lower electrical efficiency (30%). Finally, the biochar is transported over 40 km and spread on land, along with the liquid digestate. The two scenarios are shown in figure 1.

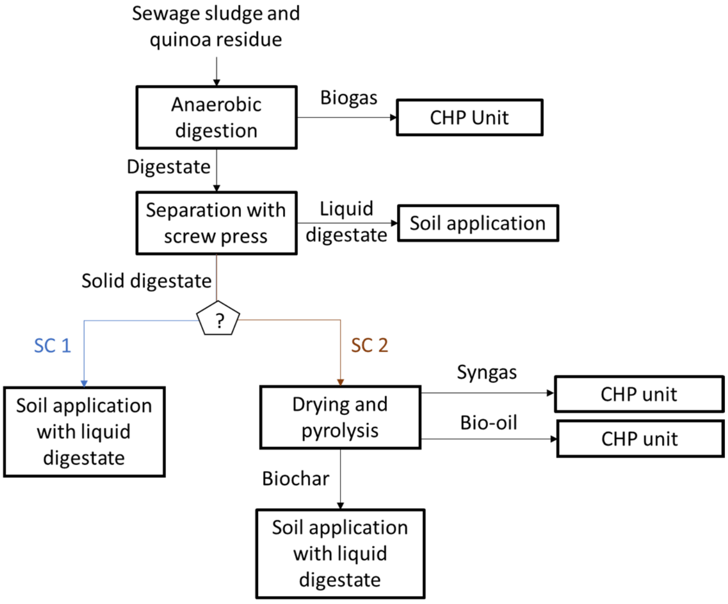

Figure 1 : Life cycle diagram for two different treatment of solid digestate corresponding to scenario 1 (SC1) and scenario 2 (SC2)

3.2 Life Cycle Assessment

The aim of this attributional LCA study is to compare two sludge-to-energy pathways based on their environmental impacts. The study is performed from a cradle-to-grave perspective. The LCA method is standardized by ISO 14040 and 14044 and is synthetized in four steps: (1) Goal and Scope Definition; (2) Life Cycle Inventory (LCI); (3) Life Cycle Impact Assessment (LCIA); (4) Results and Interpretation. The following subsections relate the realization of these four stages.

3.2.1 Goal and Scope Definition

This study is part of a large biorefinery project in Morocco, aiming at developing new waste treatment channels for energy and fertilizer production while implementing circular economy concept. AD units for sewage sludge treatment are already in operation in Morocco; hence, the objective is to evaluate the interest of adding pyrolysis of digestate. The results are first intended to Moroccan leaders of the project but also to scientific community.

Methodological choices in LCA of biorefineries are often complicated issues. Indeed, the choice of the functional unit (FU) and of coproducts management method is difficult when the system includes several products of interest (Sills et al., 2020). In order to fairly represent all the functions of the system, it is sometimes proposed to choose a FU that combines several functions (Ahlgren et al., 2015; Sills et al., 2020). However, the results can be difficult to understand and compare with other studies. Another suggestion is to choose a FU based on one of the products, but then it is necessary to define the main product, which can be discriminating. That is why a FU based on entering biomass is often chosen, even if it is not perfectly adapted to the objective of the study. It avoids indeed the choice of a main product and allows the comparison with others studies (Ahlgren et al., 2015). Following these considerations, the FU was set as the treatment of one ton of dried biomass (t DM), which is a mix of sewage sludge (80%) and quinoa residue (20%), in Morocco. All the inputs and outputs of the system are then expressed per FU.

The system boundaries include sludge and quinoa acquisition and pre-processing, treatment of mixture by AD (and PY in the second scenario), distribution and storage of the products, end-of-life treatment of the products. The system boundaries are illustrated in figures 2 and 3. The previous stages related to the quinoa harvest and the WWTP are not included in this analysis, as they can be considered independent of the evaluated scenarios. The infrastructure, maintenance and dismantling steps are excluded, only the operational phase is evaluated. The system expansion approach is adopted in this study, which means that the excess electricity and heat produced can avoid the production of the same amount of electricity from the Moroccan national grid and heat from natural gas boiler. Similarly, as the liquid digestate contains a significant amount of nitrogen N, phosphorus P and potassium K, it is supposed to replace a part of chemical fertilizer, which therefore does not have to be produced. On the contrary, solid digestate and biochar have been excluded from the system expansion approach. Indeed, the authors considered that the application of these two carbon-rich products is an added-value for lands but it does not compensate for the use of other amending products. However, CO_2 emissions and nutrients lixiviation have been measured and compared for the application of both products along with liquid digestate.

The environmental impacts of the two pathways are assessed with the Environmental Footprint (EF) 3.0 method, developed by the European Commission, as it is the most up-to-date European method. The EF impact assessment method includes the 16 following impact categories: climate change, ozone depletion, ionizing radiation, photochemical ozone formation, particulate matter, human toxicity non cancer, human toxicity cancer, acidification, freshwater eutrophication, marine eutrophication, terrestrial eutrophication, freshwater ecotoxicity, land use, water use, fossils resource use, and mineral and metals resource. To assess the reliability of the results, uncertainty and sensitivity are performed. First the sensitivity of the impact method is evaluated by performing the LCA with the ReCiPe 2016 method. Then the sensitivity analysis is carried out concerning factors which are highly uncertain in this study: the distance between the digester and the field, the amount of desulfurizing agent and the source of water for AD. To evaluate data quality, the Monte Carlo analysis is realized in SimaPro to measure the uncertainty propagation between the input parameters and the results. Each foreground data is evaluated according to the pedigree matrix (Weidema, 1998).

3.2.2 Life Cycle Inventory

The life cycle inventory (LCI) step consists in the collection, description and quantification of the input flows (resource and energy consumption) and output flows (emissions to air, water and soil) of the whole system. Foreground data were obtained from the project experiments at pilot scale (20 L digester) which were extrapolated (linear scaling) for an industrial unit of 1500 m^3. The LCI database Ecoinvent v3.4 was used to describe background data. Details are given in Table 1.

3.2.2.1 Feedstock transportation and storage

The AD plant is assumed to be adjacent to the WWTP, so there is no need to transport sewage sludge. As for quinoa, the residues are supposed to be transported over 40 km (distance from the quinoa field to AD unit) using truck. Emissions and consumptions associated to transportation steps are determined within the Ecoinvent database. Emissions linked to a potential storage step of the quinoa residues are neglected because the quinoa stalks are composed of 90% DM which results in very few emissions (eventually some dust). The substrates characteristics are detailed in supplementary information.

3.2.2.2 Pre-treatment and mixing

After transportation, the pre-treatment of quinoa stalks consists in grinding and require electricity consumption. Sludge and quinoa are then mixed with rainwater to control DM content (10%). To model the impacts of electricity, the Ecoinvent process data corresponding to medium voltage electricity in

Morocco is chosen. This process data represent Moroccan electric mix of 2016, consisting of 81% of fossil fuels (coal, natural gas and oil) (IEA- International Energy Agency, 2016).

3.2.2.3 Anaerobic digestion and CHP plant

After pre-treatment, the mixture (sludge, quinoa residue and water) is fed into the AD plant. The energy requirements for AD consists in electricity to allow a permanent mixing of compounds and heat to maintain temperature to 39°C. An average value of 1% of the methane produced is considered to leak in the atmosphere (Poeschl et al., 2012).

Biogas always contains an amount of hydrogen sulphide H_2S, with concentrations ranging from 100 ppm to 1000 ppm depending on substrate characteristics.

The presence of H_2S in biogas raises technical, environmental and health issues. First, this compound is highly corrosive for most of the equipment (pipes, motors, gas tank etc.). In addition, its combustion leads to sulphur dioxide emissions SO_2 that is harmful for the environment. Finally, in case of H_2S leaks, there are risks of pulmonary oedema for concentrations above 400 mg/m^3 (Okoro and Sun, 2019).

Due to various problems caused by hydrogen sulphide, a desulfurization step is necessary. Numerous methods exist to remove H_2S and the choice of a method depends on the desired end use of biogas (use for cogeneration or boilers, injection into the natural gas network etc.)(Andriani et al., 2020). It also depends on biogas volume, H_2S concentration and digester size. In this study, H_2S concentration in the biogas is equal to 794 ppm and the biogas enters CHP unit. The selected desulfurization method consists in adding ferric chloride $FeCl_3$ directly to the digester, with the incoming substrate. This method allows to drastically reduce the concentration of H_2S but could not be used for the transformation of biogas into biomethane. Ferric chloride then reacts with hydrogen sulfide to form iron sulfide salts, according to equation 1 (Eq 1). The iron sulfide salts are found in the digestate and are assumed not to modify digestate properties.

$$2\mathrm{FeCl_3} + 3\mathrm{H_2S} \rightarrow 2\mathrm{FeS}\ (s) + \mathrm{S}(s) + 6\mathrm{HCl}(g) \qquad \text{(Eq 1)}$$

The quantity of $FeCl_3$ has been taken from the literature, although the values of $FeCl_3$ consumption are very heterogeneous depending on the publication (Al-Imarah et al., 2017, Zhuan et al., 2018, Bailon Allegue and Hinge, 2014), as underlined in the sensitivity analysis.

In order not to mitigate the environmental impacts, the greatest value has been retained and is equal of 160 g $FeCl_3$ / Nm^3 of biogas (Bailon Allegue and Hinge, 2014). The biogas is sent to CHP unit, and the energy requirement for CHP is supposed to be compiled in AD energy consumption. The energy requirement data for the pre-treatment and AD are presented in Table 1.

3.2.2.4 Treatment of digestate

Digestate is separated into liquid and solid phases after digestion by means of a screw press. This solid-liquid separation requires electricity. In the first scenario, liquid and solid digestate are stored in covered pits before being transported and spread on land together for their respective fertilizing and amending properties. During the storage phase, it is revealed that GHG emissions can occur due to the high content of degradable organic matter (volatile solid) in digestate. Indeed Zeshan and Visvanathan underlined that digestate can lead to emissions of methane, ammonia, carbon dioxide and nitrous oxide (Zeshan and Visvanathan, 2014). However, the GHG emissions could be reduced if the storage phase is realised in covered pits. In this study, the emissions occurring during storage are not taken into account due to a lack of data.

The liquid digestate is composed of valuable nutrients (nitrogen N, phosphorus P and potassium K) and has proven to be a potential fertilizer (Monlau et al., 2015b). Thereby, where liquid digestate is spread, there is no need for additional chemical fertilizer. In LCA, it is then considered that some chemical fertilizer production is avoided by the application of liquid digestate. According to experimental tests (Tayibi, 2021), one kilogram of liquid digestate contains 3.61 g of N, 2.45 g of P_2O_5 and 2.7 g of K_2O.

An application of 7.4 tons of liquid digestate (value corresponding to FU) provides 27 kg of nitrogen as N, 18 kg of phosphorus as P_2O_5 and 20 kg of potassium as K_2O. The CO_2 emissions and nutrients lixiviation associated with liquid digestate spreading have also been measured (Table 1).

As for solid digestate, experimental tests were realised to measure CO_2 emissions related to its spreading (Table 1).

In the second scenario, the solid digestate is dried to 92% DM using a boiler, and then fed to the pyrolysis unit. The energy requirement for digestate separation and drying of digestate are presented in the Table 1.

3.2.2.5 Pyrolysis and products treatment

Pyrolysis of solid digestate is performed on pilot scale in a steel reactor. For the experiment, the basket of feedstock and the furnace are purged with N_2 for 30 minutes. At industrial scale, we suppose that the amount of N_2 needed to ensure inert conditions at the beginning is negligible as the device is operated continuously. Pyrolysis requires electricity to heat the furnace and to cool the condensable gases. The energy requirements are expressed in Table 1.

Biochar is recovered from the furnace, transported over 40 km and spread along with liquid digestate on crop field. The co-application of biochar and liquid digestate result in a slightly better nitrogen retention compared to solid and liquid digestate spreading (Table 1, soil nitrogen emissions). The real benefit of applying biochar is a better carbon storage, which significantly reduces CO_2 emissions (-99.7%) compared to solid digestate application (Table 1).

The bio-oil is sent to CHP unit and the syngas is sent to the same CHP unit as the biogas.

Table 1 : Input from technosphere and output to technosphere for both scenarios (values reported to UF, 1 ton of dried biomass) with equivalent processes from Ecoinvent database v3.4 (Cut-off, U for each data)

Biomass preparation	**Units**	**Scenario 1 AD**	**Scenario 2 AD + PY**	**Data quality score**	**Database process**
Input					
Electricity (quinoa grinding)	kWh	3.5		(2,na,1,5,na)	Electricity, medium voltage {MA} | market for electricity, medium voltage
Transport (quinoa stalks)	tkm	24.3		(3,na,1,1,na)	Transport, freight, lorry 16-32 metric ton, EURO3 {RoW}|market for transport, freight, lorry 16-32 metric ton, EURO3
Water	tons	6.97		(1,na,1,3,na)	Water, harvested from rainwater {GLO} | market for water, harvested from rainwater
Anaerobic digestion and CHP unit					
Input					
Iron chloride (desulfurization)	kg	53		(3,na,2,3,na)	Iron (III) chloride, without water, in 40% solution state {GLO} | market for
Heat	kWh	141		(2,na,1,1,na)	Heat, central or small-scale, natural gas {RoW} | market for heat, central or small-scale, natural gas
Electricity	kWh	72		(2,na,1,1,na)	Electricity, medium voltage {MA} | market for electricity, medium voltage
Output		Biogas	Biogas and Syngas		
Methane (emissions to air)	kg	1.26		(2,na,2,2,na)	Methane
Heat	kWh	942	1052 (942+110)	(2,na,1,1,na)	Heat, central or small-scale, natural gas {RoW} | market for heat, central or small-scale, natural gas
Electricity	kWh	716	799 (716+83)	(2,na,1,1,na)	Electricity, medium voltage {MA} | market for electricity, medium voltage
Heat, waste	kWh	226	252 (226+26)	(2,na,1,1,na)	Heat, waste
Digestate separation					
Input					
Electricity	kWh	4.5		(2,na,2,5,na)	Electricity, medium voltage {MA} | market for electricity, medium voltage
Liquid digestate spreading					
Input					
Transport	tkm	296		(3,na,1,1,na)	Transport, freight, lorry 16-32 metric ton, EURO3 {RoW}|market for transport, freight, lorry 16-32 metric ton, EURO3
Output		With solid digestate	With biochar		
Nitrogen fertilizer	kg	27		(2,na,1,3,na)	Nitrogen fertilizer, as N {GLO}| market for
Phosphate fertilizer	kg	18		(2,na,1,3,na)	Phosphate fertilizer, as P_2O_5 {GLO}| market for
Potassium fertilizer	kg	20		(2,na,1,3,na)	Potassium fertilizer, as K_2O {GLO}| market for
Carbon dioxide (emissions to air)	kg	102		(2,na,1,3,na)	Carbon dioxide
Ammonia (emissions to air)	kg	1.33	1.32	(2,na,1,3,na)	Ammonia
Nitrogen (emissions to soil)	kg	6.33	4.81	(2,na,1,3,na)	Nitrogen
Phosphorus pentoxide (soil)	kg	0.46	0.51	(2,na,1,3,na)	Phosphorus pentoxide
Solid digestate spreading					
Input					
Transport	tkm	66	/	(3,na,1,1,na)	Transport, freight, lorry 16-32 metric ton, EURO3 {RoW}|market for transport, freight, lorry 16-32 metric ton, EURO3
Output					
Carbon dioxide (emissions to air)	kg	31	/	(2,na,1,3,na)	Carbon dioxide
Solid digestate drying					
Input					
Heat	kWh	/	1140	(2,na,1,1,na)	Heat, central or small-scale, natural gas {RoW} | market for heat, central or small-scale, natural gas
Pyrolysis					
Input					
Electricity	kWh	/	76	(2,na,1,1,na)	Electricity, medium voltage {MA} | market for electricity, medium voltage
Bio-oil cogeneration					
Output					
Heat	kWh	/	106	(3,na,1,1,na)	Heat, central or small-scale, natural gas {RoW} | market for heat, central or small-scale, natural gas
Electricity	kWh	/	64	(3,na,1,1,na)	Electricity, medium voltage {MA} | market for electricity, medium voltage
Heat, waste	kWh	/	43	(3,na,1,1,na)	Heat, waste
Biochar spreading					

Input					
Transport	tkm	/	4.9	(3,na,1,1,na)	Transport, freight, lorry 16-32 metric ton, EURO3 {RoW}|market for transport, freight, lorry 16-32 metric ton, EURO3
Output					
Carbon dioxide (emissions to air)	g	/	79	(2,na,1,3,na)	Carbon dioxide

The overall tree processes for scenarios 1 and 2 are shown in figures 2 and 3.

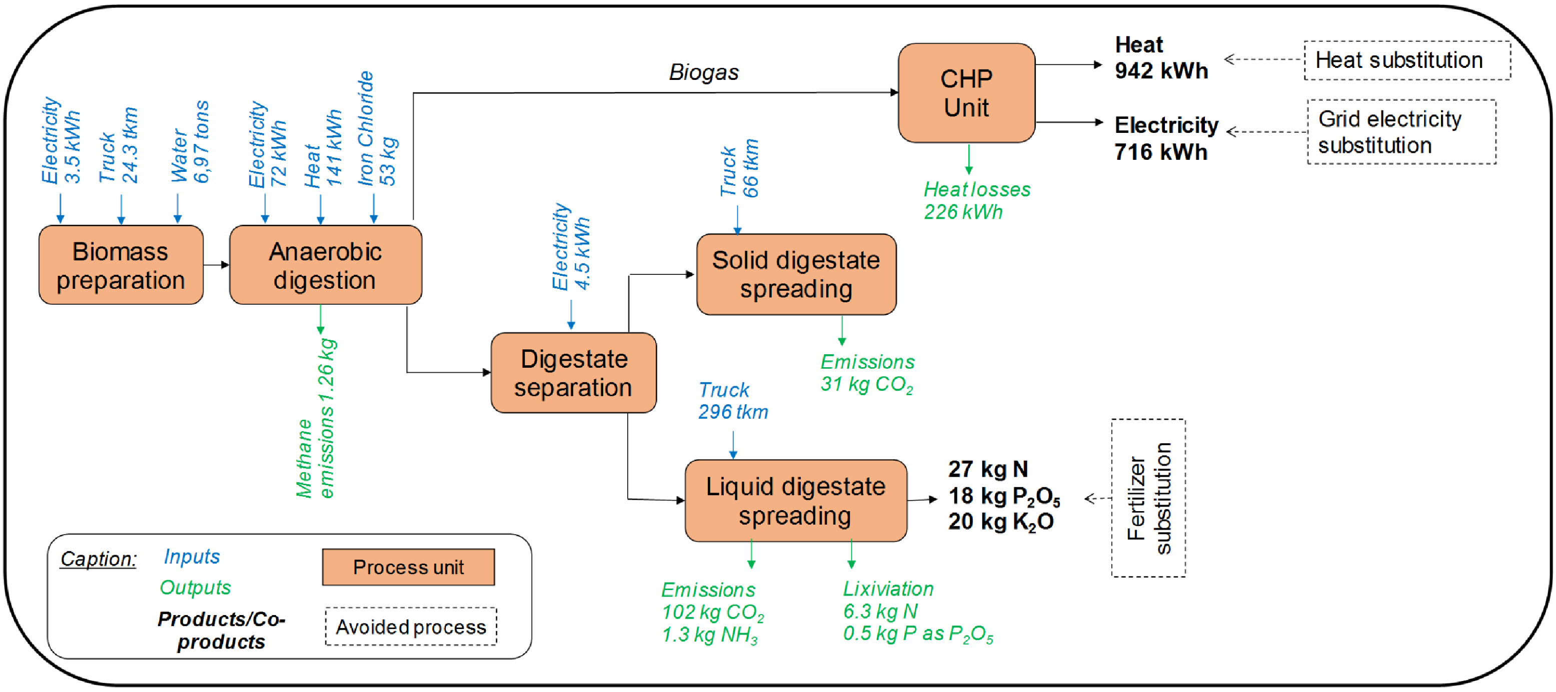

Figure 2 : tree process for anaerobic digestion scenario – scenario 1 (values reported to UF, 1 ton of dried biomass)

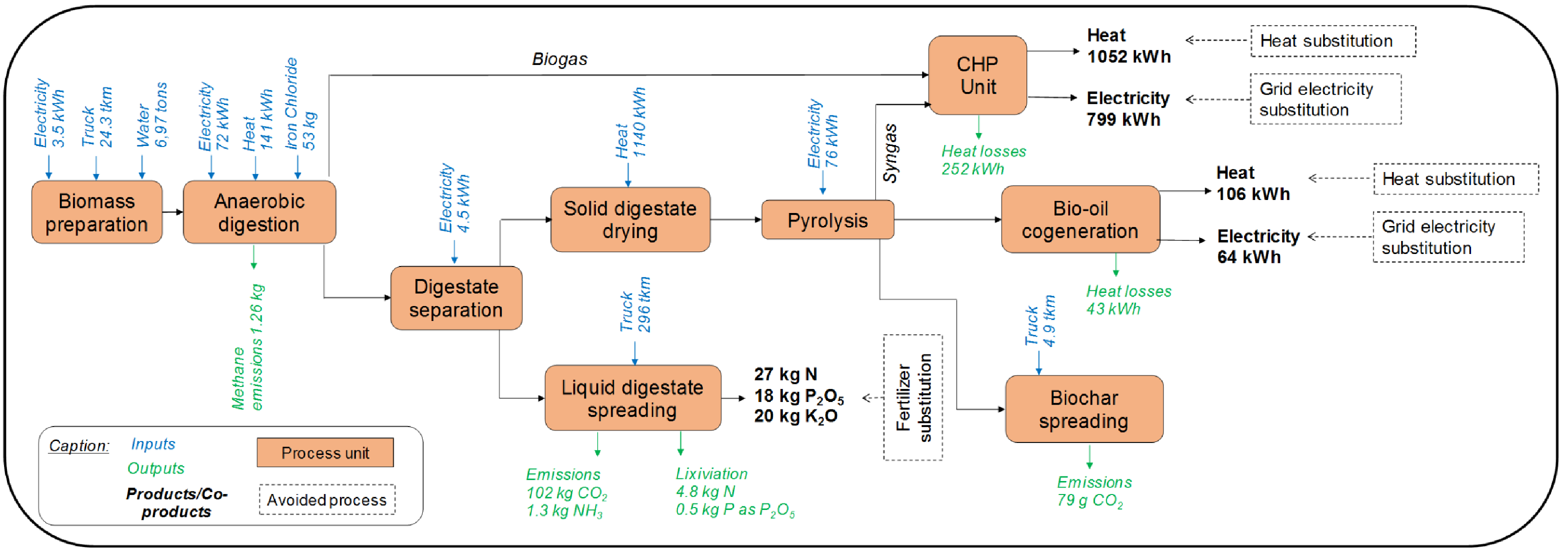

Figure 3 : Tree process for integration of anaerobic digestion and pyrolysis – scenario 2 (values reported to UF, 1 ton of dried biomass)

3.2.3 Life Cycle Impact Assessment

The Life Cycle Inventory step made it possible to determine the quantities of materials extracted and energy required, as well as emissions to air, soil and water, related to the different stages of the processes under study. After data collection, the third step, called life cycle impact assessment (LCIA), consists of estimating environmental impacts of the system by relating the inventory flows from the LCI to the respective environmental impacts. Thus, each emitted or extracted substance is linked to a number of impacts categories (classification step) then the values are converted into common units and aggregated within each category (characterization step). The LCIA is done using SimaPro software (V8.5.2) and the midpoint EF 3.0 method. According to the European Commission (European Commission, PEFCR Guidance document, 2017), not all categories in this method have the same robustness, ranging from I to III. The robustness indicators for each category are presented in Table 2 and in all results figures.

Following the characterization step, ISO 14044 standard proposes two optional steps, normalization and weighting. Normalization is useful to identify the most relevant impacts categories and process stages in the system. Normalization is done using normalization factors, which represent the total impact of a reference region for a certain impact category (e.g. climate change, eutrophication, etc.) over the course of a reference year. For the EF method, the normalization factors represent the total worldwide impact for each category in 2010. These factors are then divided by the number of inhabitants, to obtain one normalization factor per year per person.

4. Results

4.1. Results of scenario 1

The LCIA results of the first scenario are illustrated in figure 4; positive values represent environmental burdens while negative values represent environmental benefits. For each impact category, the contribution of each process unit to the total impact is highlighted in these figures.

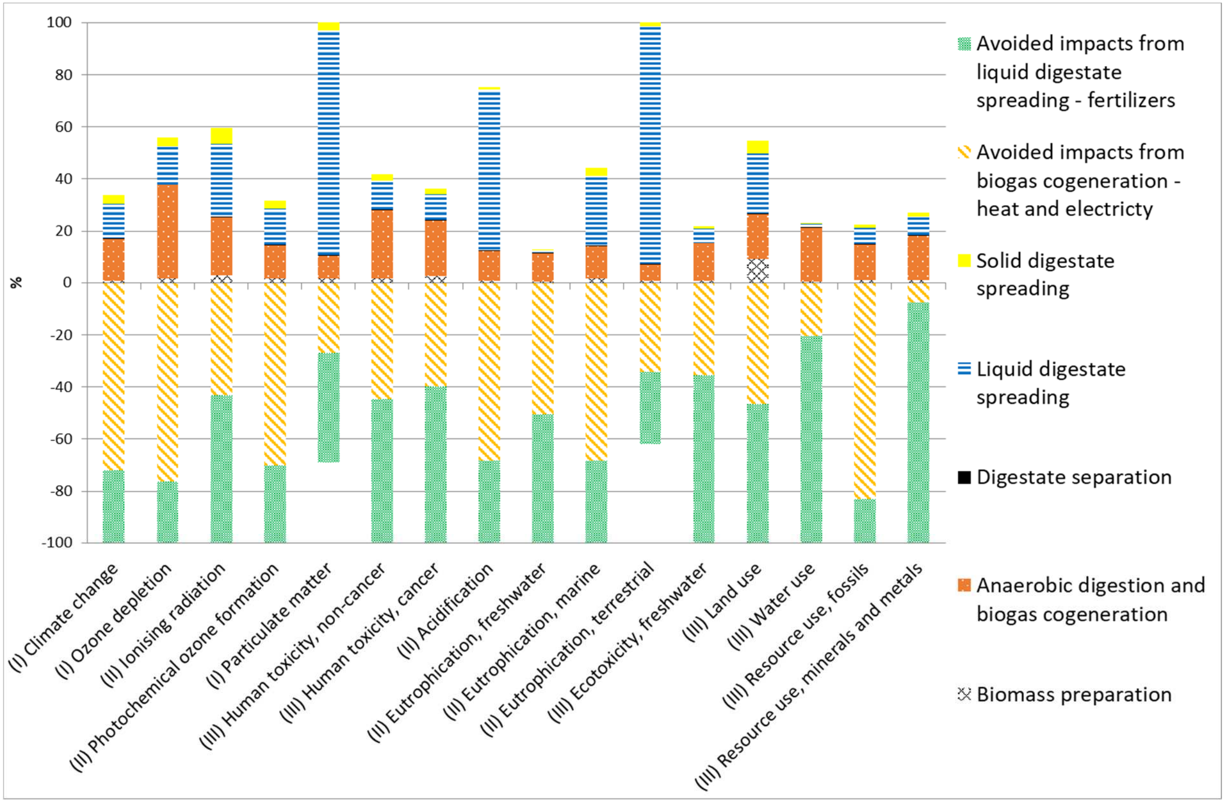

Figure 4 : Processes contribution to environmental impacts for anaerobic digestion process – scenario 1

In the first scenario, the main contributor to nine out of sixteen categories is the anaerobic digestion/biogas cogeneration step, while for the other seven categories, major impacts are induced by liquid digestate spreading. The impacts of liquid digestate application are mainly due to ammonia and carbon dioxide emissions, leading to major impacts on particulate matter, acidification and terrestrial eutrophication, but the impacts are due also to nutrients leaching. For anaerobic digestion/biogas cogeneration, environmental burdens are explained by the requirement of a consequent amount of ferric chloride for desulfurization and the consumption of a lot of electricity. The digestate separation and solid digestate spreading stages do not contribute much to the impacts compared to the other life cycle steps. Except for terrestrial eutrophication and particulate matter categories, the abatement of the emissions are greater than the emissions, as highlighted in the Table 2. Although scenario 1 generates impacts, these impacts are compensated by the energy generated by the biogas cogeneration that substitutes an equivalent amount of energy from national grid. The avoided impacts due to avoided electricity production are very important because the Moroccan electricity mix is strongly based on coal. The impacts are also compensated trough liquid digestate application, which substitutes a large amount of synthetic fertilizer and limits resources depletion. As different LCIA methods are used in the literature, the comparison of the results is mainly done regarding the climate change impact as it is estimated according to the same approach developed by the IPCC (IPCC, 2013). Highly-variable results have been found for the anaerobic digestion of sludge with agricultural application of digestate. Tarpani et al. (2020) determined net-negative GHG emissions of -174 kg CO_2 eq. / t DM, after the credits for electricity and fertilizer. They stated that their value falls in the lower range of results from other studies that they found (ranging from -280 to 650 kg CO_2 eq./ t DM) (Tarpani et al., 2020). The potential climate change impact determined here is very low compared to these results (- 769 kg CO_2 eq. / t DM). The difference may be explained by the avoided impacts considered. The electricity produced thanks to CHP units allows to replace electricity from the Moroccan grid, which is strongly based on coal: the impacts of electricity consumption are higher, so as the avoided impacts which counterbalance the environmental burdens. Opatokun et al. (2017) obtained a net value of – 757 g CO_2 eq. for the treatment of one kilogram of food waste thanks to AD (Opatokun et al., 2017). As stated in the introduction, the studies can be hard to compare with each other due to differences in scope, feedstock, geographical area, technologies etc. (Eriksson et al., 2016; Tarpani et al., 2020).

Table 2 : Results of the environmental assessment of both sludge-to-energy systems with EF method: single AD (scenario 1) and integration of AD with pyrolysis (scenario 2)

Impact category	Unit	Result for scenario 1 AD	Result for scenario 2 AD + PY	Robustness indicator
Climate change	kg CO_2 eq	-7.69E+02	-6.04E+02	I
Ozone depletion	kg CFC-11 eq	-3.37E-05	-2.11E-06	I
Ionising radiation	kBq U235 eq	-4.59	-4.79	II
Photochemical ozone formation	kg NMVOC eq	-2.00	-2.00	II
Particulate matter	disease inc.	1.18E-05	1.12E-05	I
Human toxicity, non cancer	CTUh	-4.27E-06	-4.14E-06	III
Human toxicity, cancer	CTUh	-1.84E-07	-1.55E-07	III
Acidification	molc H+ eq	-1.74	-1.98	II
Eutrophication, freshwater	kg P eq	-4.24E-02	-4.40E-02	II
Eutrophication, marine	kg N eq	-5.35E-01	-5.58E-01	II
Eutrophication, terrestrial	mol N eq	8.04	7.78	II
Ecotoxicity, freshwater	CTUe	-1.23E+04	-1.22E+04	III
Land use	Pt	-1.01E+03	-1.11E+03	III
Water use	m^3 depriv.	-1.39E+02	-1.40E+02	III
Resource use, fossils	MJ	-9.90E+03	-6.90E+03	III
Resource use, minerals and metals	kg Sb eq	-1.36E-02	-1.35E-02	III

4.2. Results of scenario 2

As for the second scenario, the results are slightly different. Liquid digestate spreading remains the main contributor for seven out of sixteen categories, but anaerobic digestion/gas cogeneration is the main contributor to only six categories (Figure 5). Drying of solid digestate brings new impacts because of a large amount of heat from natural gas consumed to achieve 92 % of DM. Thus, this step is the most impactful for climate change, ozone depletion and resource use (fossils). The impacts are particularly meaningful for ozone depletion category, as the natural gas transport process is responsible for emissions of halogenated chemical compounds (halon 1211 and halon 1301) that deplete the ozone layer. The pyrolysis process is also responsible of impacts because of electricity consumption. As in scenario 1, the avoided impacts from liquid digestate spreading and biogas cogeneration are meaningful and allow to reduce the net impact value for each category. The avoided impacts from syngas and bio-oil cogeneration participate to a lesser extent in reducing impacts. The impacts associated with biochar application are very insignificant compared to the other life cycle stages. However, it is interesting to compare the impacts of biochar application and solid digestate application, as one of the objectives of the study is to compare these two amending products. For the climate change category, the spreading of solid digestate results in the emissions of 41 kg CO_2 eq per ton of biomass, while the biochar spreading emits 0.91 kg CO_2 eq. This result underlines the carbon sequestration capacity of the biochar that allow to reduce GHG emissions by 97.8 %. The application of biochar is also beneficial for the other impact categories, as all the impacts are reduced by 92.5% compared to solid digestate application. This is due to the fact that a much smaller amount of biochar has to be transported and therefore the impacts associated with transportation are decreased.

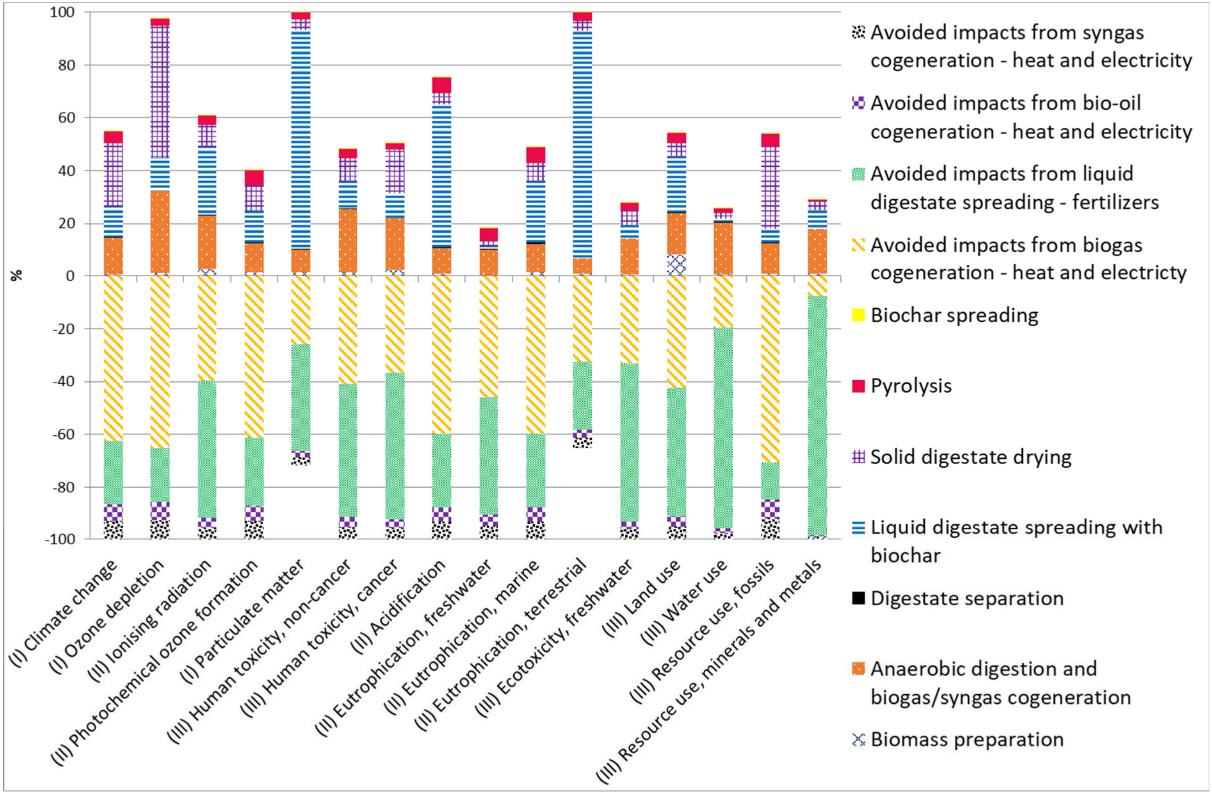

Figure 5 : Processes contribution to environmental impacts for the integration of anaerobic digestion and pyrolysis – scenario 2

4.3. Comparison of the two analysed scenarios

The comparison between the two scenarios is presented in figure 6. The results of the LCIA highlighted that both scenarios are not so different from an environmental point of views, except for three categories, for which there is a difference greater than 20% - climate change, ozone depletion and resource use (fossils). Two of these three categories have the best robustness indicator (from I to III). The gaps between the two scenarios are mainly due to heat consumption for solid digestate drying in the integrating system, wherein the heat produced cannot fulfil the heat requirement. The most significant difference (>90%) between the two scenarios concerns the ozone depletion category.

In both scenarios, only two categories have a positive net impact value: particulate matter and terrestrial eutrophication. These categories are deeply impacted by liquid digestate spreading.

In addition, the results of the normalization step (figure in the supplementary information) confirms that these two categories should receive more attention.

The overall results could make believe that the scenarios are almost environmentally beneficial. However, the particulate matter and terrestrial eutrophication categories should not be underestimated. The integration of AD and PY avoids land spreading of solid digestate and produces a preferable carbon component as the GHG emissions are significantly reduced.

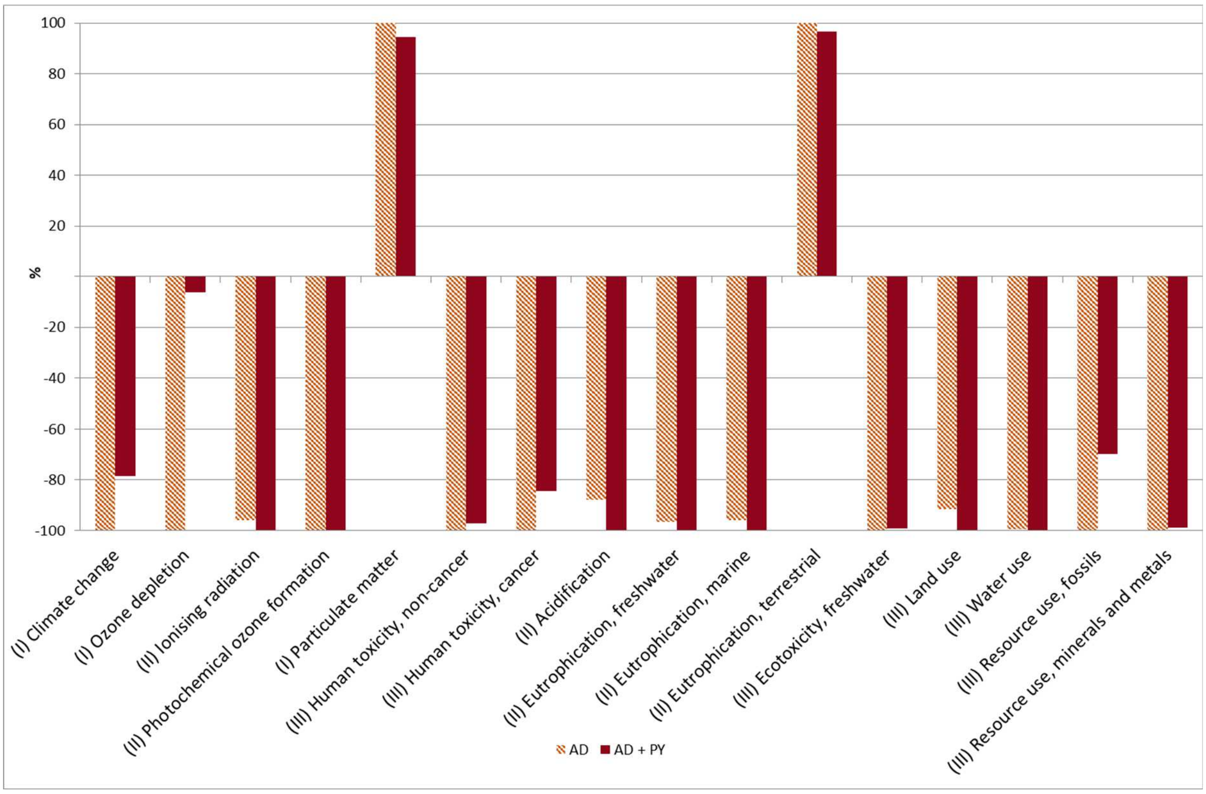

Figure 6 : Comparison between single AD (scenario 1) and integration of AD with pyrolysis (scenario 2)

The results of the integrated system have been compared to other studies. The comparison is made for the climate change category, for which it was found a net value of – 604 kg CO_2 eq / ton of dried biomass (Table 2). Similar trends were reported by Mills et al. (2014) when evaluating several sludge-to-energy technologies using the CML2001 method. They found a net impact value equal to -614 kg CO_2 eq per ton of dry solid sludge, for a scenario combining thermal hydrolysis process, AD, drying and PY (Mills et al., 2014). Likewise, Cao and Pawlowski (2013) found a net value of -630 kg CO_2 eq per dry ton sludge for an integration of AD with pyrolysis, using IPCC GWP 100y (Cao and Pawłowski, 2013). An emissions abatement of -721 g CO_2 eq per kilogram of food waste was also observed with the ReCiPe 2008 method for the assessment of AD integrated with PY (Opatokun et al., 2017). However, some authors found different trends for the digestion of 1 ton of dry paper mill sludge followed by pyrolysis. Indeed, it was calculated a net value for climate change category equal to -0.428 kg CO_2 eq (Mohammadi et al., 2019).

4.4. Uncertainty and sensitivity analyses

In order to validate the reliability of the results, sensitivity and uncertainty analyses are performed. The analyses are presented in this section and the associated figures can be found in the supplementary information. First, the sensitivity of the choice of the impact method is evaluated by performing the LCA with the ReCiPe 2016 method to compare results with EF 3.0. The two methods share 15 impact categories although the units can be different. For both scenarios, the results are rather similar concerning the processes contributions to environmental impacts with each method. There are two major differences between the LCA results obtained with ReCiPe 2016 and those obtained with EF 3.0:

- One difference concerns the ozone depletion category: with EF, AD appears to be more preferable than coupling, while with ReCiPe, the two scenarios have similar trend. This difference is due to the absence of nitrous oxide among the substances that contribute to ozone depletion in the EF model.
- The other difference concerns two impact categories that are evaluated with ReCiPe but not with EF and for which the gap between AD and AD+PY is significant: terrestrial and marine ecotoxicity.

- The last difference is about particulate matter and terrestrial eutrophication, which are the only categories with a positive net impact value with EF, while they result in negative impact value with ReCiPe.

Some of the input data were collected from literature or resulted from hypotheses, it was therefore necessary to control the influence of these parameters. The influence of the ferric chloride amount used for desulfurization was first evaluated : the value retained for the study has been compared to two other values (1.3 kg $FeCl_3$ / FU (Al-Imarah et al., 2017); 10 kg $FeCl_3$ / FU (Zhuan et al., 2018); 53 kg $FeCl_3$ / FU (Bailon Allegue and Hinge, 2014)). The ferric chloride amount appears to be a sensitive data and its value should be adjusted for later research.

The distance between crop field and AD unit was set hypothetically at 40 km. The two scenarios have been analysed with three distance possible: 10, 40 and 100 km. The results highlight that the distance is a sensitive parameter for almost eleven categories in both scenarios. The farther the field is from the unit, the greater are the impacts.

Another interesting parameter is the water consumption for biomass preparation. In this study, the water was modelled as coming from rainwater. At industrial scale, there are other possibilities as using residual water from WWTP or manure, using mains water or recycling liquid digestate. The latter alternative would save up to 89.1 % of the water required, and would avoid digestate spreading (which is complex and lead to environmental burdens).

Lastly, a Monte Carlo analysis was applied on SimaPro to quantify analytical propagation of uncertainty. For this purpose, each foreground data was evaluated according to a data quality matrix proposed in Weidema (Weidema, 1998) and the results were implemented in SimaPro. Five indicators are suggested in this pedigree matrix (in the order of appearance in SimaPro): reliability of the source, completeness, temporal correlation, geographical correlation and further technological correlation. Each indicator can be evaluated by a score ranging from 1 to 5 and the scores attributed to foreground data are presented in Table 1. The completeness and further technological correlation parameters were not used in this study, their corresponding score are accordingly stated as “na”. Once the evaluation of foreground data is performed, the Monte Carlo analysis can be launched. The LCA models were run 1000 times. The Monte Carlo analysis confirms the main results of the study (Figure 7). AD appears to have less environmental impacts for four categories for 100% of the runs, including the three categories – climate change, ozone depletion, fossils resource use – for which the gaps between AD and AD+PY were significant. For the human toxicity (cancer) category, the AD also appears to be more environmentally friendly in 100% of cases.

On the contrary, the integration of AD+PY results to have less impact for the following categories: land use, marine eutrophication and freshwater eutrophication.

For the others categories, neither of the two scenarios appears preferable because of data uncertainties.

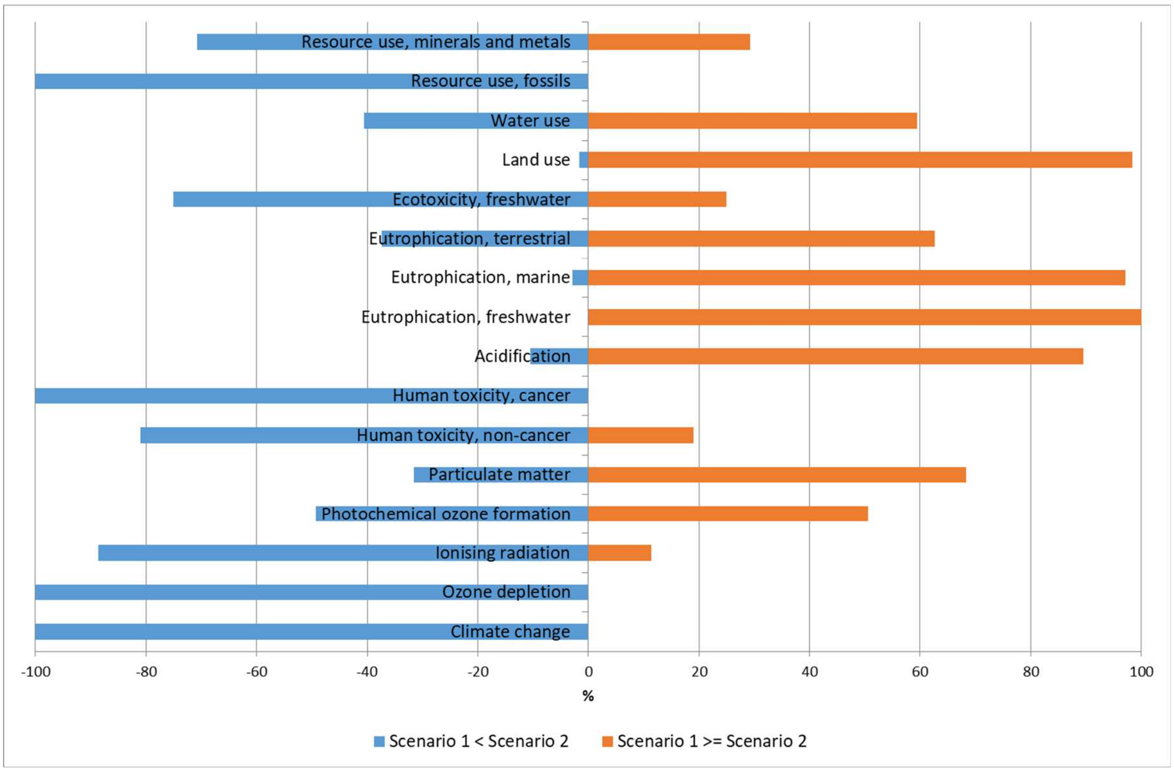

Figure 7 : Results of uncertainty analysis using the Monte Carlo method, for the comparison between scenario 1 and scenario 2

5. Discussion

In this study, a methodological challenge was faced as there was a need to make a comparative LCA between a well-established process (anaerobic digestion) and a prospective one (pyrolysis). The experimental results were extrapolated and it is not uncommon that processes at the industrial scale are significantly different from their laboratory-scale counterpart (Shibasaki et al., 2006). But LCA is particularly useful in identifying and comparing the advantages and weaknesses of innovative processes with conventional ones (Shibasaki et al., 2006; Shibasakia et al., 2007).

The comparison between the two sludge-to-energy systems highlight that both alternatives have relatively similar environmental impacts, but more research should be done according to the following considerations. The benefits or drawbacks of pyrolysis may be accentuated by considering a more detailed treatment of pyrolysis products.

In this study, bio-oil from pyrolysis was considered to enter CHP unit to recover energy content, as the organic bio-oil has a higher heating value (HHV) estimated at 34 MJ / kg. The energy produced during bio-oil combustion was estimated to have beneficial impacts on the environment. However, the impacts of the bio-oil recovery process were probably underestimated. After pyrolysis, gases and vapours need to be cooled to recover the oils on the one hand and non-condensable gases on the other hand. This step may include product and energy consumption that appeared not to be negligible in previous study (Cusenza et al., 2021). Although the combustion route is the most common for bio-oil treatment in industrial units, another possible recovery route is to separate bio-oil by liquid-liquid extraction to obtain organic and aqueous phases. Indeed, both phases contain molecules of interest. The aqueous fraction of the bio-oil contains mainly water, water soluble substances (sugars, volatile fatty acids, oligomers…) and slightly soluble substances (phenols and furans) (Tayibi, 2021). It has been reported for instance that phenolic compounds in the aqueous phase have antifungal potential (Brassard et al., 2020). The application of the aqueous phase as bio-fungicide is coherent with the increased need of bio-pesticide to avoid fossil-based synthetic pesticides. To perform a rigorous evaluation of the extraction and valorisation of bio-oil, it is necessary to qualify and quantify the molecules of interests from both phases, and to know their possible recovery route. To be profitable, the process must produce a sufficient quantity of valuable products, to compensate environmental and economical burdens of the extraction and recovery processes.

Environmental assessment of the integration of AD and pyrolysis highlighted that land application of biochar reduces GHG emissions by 97.8 % compared to solid digestate spreading, revealing the carbon sequestration potential of biochar. According to the literature, biochar has revealed to have many other potentials. It could be used as a soil amendment to improve soil physical and biological properties, increase soil fertility and crop yield (Mohammadi et al., 2019; Monlau et al., 2016). It is also known to prevent nutrients leaching thus preserving water quality and reducing soil erosion (Monlau et al., 2016). Further tests on biochar application are still necessary to carry out a rigorous LCA taking into account all the benefits linked to biochar application. Several authors have also highlighted the possibility to use biochar as an additive in AD to improve biogas production (Kumar et al., 2021).

In addition to the advantages of pyrolysis products, the integration of AD and PY of solid digestate could make it possible to treat all kinds of biomass, even those recalcitrant to degradation by AD alone. Indeed, biomass rich in lignin (wood, olive pomace) are not suitable for AD as they lead to operational instability, but they could be treated by pyrolysis.

Regardless of the scenario, liquid digestate spreading results in consequent impacts. As stated in the introduction, digestate spreading is not without consequences. In this study, solid digestate is evaluated as a substrate for pyrolysis, but liquid digestate is spread on field, for its fertilizing properties. Other studies have evaluated pyrolysis of dewatered digestate, but no spreading of the aqueous residual material (Li and Feng, 2018; Mills et al., 2014; Opatokun et al., 2017). In these studies, the “liquid digestate” is either sent to a WWTP or simply not considered. The point is that digestate should not be applied on any land, and that the quantities should be carefully monitored. Questions about storage duration, season of storage, maturity of digestate are raised and can lead to debate about land application of digestate. A possibility underlined in the literature is to reuse liquid digestate as a substrate for AD (Li et al., 2020), while sending the solid digestate to pyrolysis. In this way, the handling of liquid digestate spreading would be resolved and less water would be needed at the digestion input to decrease the DM. Many researches are still to be undertaken to approach the best compromise for the circular economy and to get closer to the concept of industrial ecology (Belaud et al., 2019).

Another important issue is the desulfurization step during AD. In this study, the amount of ferric chloride needed to remove hydrogen sulphide was found in the literature, and the worst value was chosen. It could be interesting to evaluate several desulfurization technologies, from an environmental and an economic point of views.

In this study, the characteristics of the substrates entering the AD process were assumed to be constant. However, the quality and composition of entering biomass could vary and lead to different LCA results (Li and Feng, 2018). This variability was not included in the analysis. Similarly, the issue of heavy metals has not been addressed, but it would be interesting to compare the amount of heavy metals in the sludge to that in the products applied to land, as underlined in some studies (Tarpani et al., 2020).

To better inform decision makers, this environmental assessment should be completed with an economic analysis. In parallel to LCA, it is possible to develop Life Cycle Costing analysis (LCC), which aims to determine costs and benefits associated with each life cycle step of a system. However, unlike LCA which is standardized, there is no international standard or certification for LCC, resulting in a wide variety of terms being used (Carlsson Reich, 2005; Zhou et al., 2018). The determination of global cost is often requested to apply LCC and consists in the acquisition of investment costs (capital expenditure, CAPEX) and operational costs (operational expenditure, OPEX). The complexity of determining the costs of an innovative process such as the one studied, is to access certain prices and costs. The costs of pyrolysis units, bio-oil and syngas treatment technologies and also the price of biochar can be difficult to obtain. Another complexity that appears when carrying out LCC is the monetization of environmental impacts. It consists in transcribing the results of the environmental assessment to monetary units. So far, there is no consensus in the scientific community on the most appropriate method for application of monetization in LCA (Rajabi Hamedani et al., 2019). Its implementation certainly simplifies the communication of results, but this simplification has significant limits (Rödger et al., 2018, Garcia and Reyes Carillo, 2019).

6. Conclusions

Through a cradle-to-grave LCA, the environmental performance of a process integrating anaerobic digestion and pyrolysis for sewage sludge treatment is compared to a single anaerobic digestion process. The study aims to emphasize an eventual potential of integrated systems compared to stand-alone ones. The overall results show that both systems have similar environmental impacts – whether beneficial or harmful. More research should be conducted to optimise the valorisation of pyrolysis products, as bio-oil could be upgraded for its interesting molecules and biochar has several potentials that were not evaluated here. The fate of liquid digestate should also be re-evaluated: its spreading on land is an alternative that need a real monitoring and other solutions as its reuse in anaerobic digestion should be considered. Otherwise, considering avoided production of energy and synthetic fertilizer play an important role as these avoided productions contribute to decrease the impacts of almost all categories. This innovative multifunctional pathway seems quite promising and could avoid some of the drawbacks of anaerobic digestion. Further evaluation of the enhanced overall process is recommended, along with an economic analysis.

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