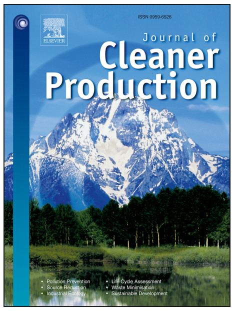

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PII: S0959-6526(16)31848-0
DOI: 10.1016/j.jclepro.2016.11.013
Reference: JCLP 8400

To appear in: *Journal of Cleaner Production*

Received Date: 23 March 2016
Revised Date: 31 October 2016
Accepted Date: 1 November 2016

Please cite this article as: Benavente V, Fullana A, Berge ND, Life cycle analysis of hydrothermal carbonization of olive mill waste: Comparison with current management approaches, *Journal of Cleaner Production* (2016), doi: 10.1016/j.jclepro.2016.11.013.

LIFE CYCLE ANALYSIS OF HYDROTHERMAL CARBONIZATION OF OLIVE MILL WASTE: COMPARISON WITH CURRENT MANAGEMENT APPROACHES

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ABSTRACT

Significant efforts have been direct towards developing environmentally sustainable and economically beneficial treatment of olive mill wastes. Recently, hydrothermal carbonization (HTC) has been shown to be a potentially beneficial approach for the treatment of olive mill wastes. When considering the use of HTC to treat these wastes, however, it is critical that its environmental implications be evaluated and subsequently compared to other commonly used treatment approaches. In this study, the environmental impacts associated with using HTC to treat olive mill wastes were evaluated and compared to aerobic composting, anaerobic digestion, and incineration using life cycle assessment. Results indicate that HTC coupled with subsequent energy recovery from the combustion of the generated hydrochar results in net environmental benefits and that the energy offsets derived from electricity production from hydrochar combustion are critical to achieving these savings. In addition, results indicate that HTC process water discharge significantly influences system environmental impacts, indicating that research investigating treatment alternatives is needed. Changes in carbonization temperature and hydrochar moisture content also influence system environmental impact, suggesting that both are important when considering possible industrial applications. In comparison with current management approaches, alternatives using HTC are more environmental advantageous than composting and anaerobic digestion. However, the use of HTC is not as environmentally advantageous as incineration with energy recovery because 45-35% of the energy contained in the olive mill waste is lost during HTC. However, if the electricity recovery efficiency from incineration increases to greater than 30%, the environmental impacts associated with HTC and subsequent energy generation are equal to or better than direct TPOWM incineration with energy recovery. It is recommended that future research efforts focus on the evaluation of appropriate and environmentally beneficial HTC process water treatment approaches and methods to improve the energetic retention efficiencies of the hydrochar.

Keywords: LCA, hydrothermal carbonization, energy, olive mill waste, disposal treatments, EASETECH

1. Introduction

Current olive oil production processes involve a two-phase centrifugation system that results in the generation of a large mass of a semisolid waste stream referred to as two-phase olive mill waste (TPOMW). TPOMW consists of olive pulp, a considerable amount of water (> 65%), and a variety of organic compounds (e.g., carbohydrates, proteins, lipids, aromatic compounds). This waste stream has been reported to be phytotoxic and has been shown to adversely impact microbial activity (Alburquerque et al., 2004), necessitating treatment prior to its discharge to the environment (MME, 2000). Therefore, significant efforts have been direct towards developing environmentally sustainable and economically beneficial TPOMW treatment/management approaches (e.g., Vlyssides et al., 2004; Tortosa et al., 2012).

In Spain, the largest olive oil producing country, both biological (e.g., aerobic composting (AC), anaerobic digestion (AD)) and thermal treatment processes (e.g., incineration) are used to manage this waste, as illustrated in Figure 1. Each of these processes results in the generation of value-added products (e.g., biogas, heat, power, or pomace oil) that increase system sustainability and/or economic viability. However, these processes are plagued with significant operational challenges. Biological degradation of TPOMW by microorganisms is complicated by its acidic pH and high polyphenol concentrations (Siciliano et al., 2016), and thermal treatment approaches are generally inefficient because of the high moisture content and low energy density of the waste (Van Loo and Koppejan, 2008).

An innovative wet thermal treatment approach that may alleviate many of these challenges is hydrothermal carbonization (HTC). HTC occurs at relatively low temperatures and under autogenous pressures in closed systems (Libra et al., 2011). Because HTC requires moisture, TPOMWs are better suited for conversion via HTC than other dry thermal conversion techniques. Benavente et al. (2015) report that HTC of TPOMW requires significantly less energy than that associated with dry thermal conversion approaches. In addition, HTC of TPOMW results in the generation of energy-rich solids that have properties equivalent to subbituminous/bituminous coals. Additional information associated with the HTC process and the HTC of TPOMWs can be found elsewhere (Funke and Ziegler, 2010; Libra et al., 2011; Benavente et al., 2015).

When considering the use of HTC to treat TPOMW, it is critical that its environmental implications be evaluated and subsequently compared to other commonly used treatment approaches. Such information is currently unknown, but necessary to ensure environmentally responsible treatment process selection. The purpose of this work is to use life cycle assessment (LCA) to determine the environmental impacts associated with TPOMW treatment using HTC, and to compare these impacts with those associated with currently used biological and thermal treatment approaches. The specific objectives of this work are to: (1) evaluate the environmental impacts of the current TPOMW management approaches, (2) evaluate the environmental impacts associated with HTC of TPOMW combined with the subsequent combustion of the hydrochar for energy production, (3) understand how key parameters associated with each treatment approach (e.g., energy recovery efficiencies, hydrochar moisture content) influence system environmental impacts, and (4) provide recommendations for process selection from an environmental perspective.

2. Materials and methods

2.1. Goal and scope definition

The goal of this study is to use results from an LCA to compare environmental impacts associated with the HTC of TPOMW with those associated with AC, AD, and incineration of TPOMW. This study considers the consumption and/or production of materials and energy, as well as pollutant emissions generated over the entire life cycle of each treatment approach. Avoided production and combustion of primary fuels (coal and natural gas) due to energy generation from TPOMW are also included. Upstream processes, such as waste collection and transport, are not considered in this study because it is assumed that these values are the same for all management alternatives. The functional unit of this study is defined as the treatment of 1 kg of fresh TPOMW. The physico-chemical properties of the TPOMW modeled in this work taken from Benavente et al. (2015) and are described in Table SI-1 in the supplementary information (SI).

2.2. Life Cycle Assessment (LCA)

2.2.1. Modelling approach

LCA modeling was performed using the Environmental Assessment System for Environmental Technologies software (EASETECH, version 2.0.0), a mass-flow based LCA tool developed by researchers at the Technical University of Denmark to evaluate the environmental impact of waste management processes (Clavreul et al., 2014). EASETECH was chosen for use in this study because it has been used extensively for modeling waste-related processes, similar to those commonly used as TPOMW management approaches (e.g., incineration, AC, AD, HTC). All input waste material fractions are specified in terms of elemental composition (e.g., carbon, hydrogen, etc.) and fraction-specific properties (e.g., moisture and energy content, etc.), and are tracked through the system. Additional details associated with EASTECH and its use in modeling waste management systems can be found elsewhere (e.g., Clavreul et al., 2014).

2.2.2. Description of scenarios and data inventory

Six TPOMW treatment approaches were modeled and evaluated, as illustrated in Figures 2 and 3. These scenarios encompass different biological and thermal treatment approaches currently practiced in Spain, as well as a scenario in which HTC is used. Three different biological scenarios were modeled (Figure 2), including: (1) co-composting of TPOMW with other agricultural wastes (B.1), (2) anaerobic digestion of the TPOMW with subsequent aerobic composting of the digestate (B.2), and (3) anaerobic digestion of TPOMW pre-treated using catalytic oxidation following the Fenton-like process to maximize anaerobic degradation, with subsequent composting of the digestate (B.3). Specific details associated with each process, including process material and energy needs and operational parameters, can be found in the supporting information (see section 1.1).

Three thermal TPOMW treatment scenarios were also modeled (Figure 3), including: (1) TPOMW incineration with energy recovery (T.1), (2) extraction of pomace oil from the TPOMW, followed waste incineration with energy recovery (T.2), and (3) HTC of TPOMW with subsequent incineration of hydrochar with energy recovery (T.3). Specific details associated with each process, including process material and energy needs and operational parameters, can be found in the supporting information (see section 1.2).

Inventory data associated with these scenarios were either calculated from experimental results or collected from previously published data sources, including life cycle inventory studies, scientific literature describing experimental studies, and/or Ecoinvent databases. Specific details associated with each management strategy and relevant individual assumptions are described in the supporting information (see discussion in section 1).

2.2.3. Impact categories

Nine of the International Reference Life Cycle Data System (ILCD)-recommended impact categories were evaluated and compared in this work, as shown in Table 1. Each method is described in Hauschild et al. (2012). These categories were chosen because they are environmentally relevant and internationally accepted in accordance with ISO 14040:2006 (ISO 2006). Normalization and weighting of the impacts were not performed in this study.

2.2.4. Sensitivity analysis and model simulations

Sensitivity analyses (SA) were conducted to ascertain how parameters associated with the HTC of TPOMW that represent information that is either currently unknown or may change from application to application influence overall system environmental impact. Tables 2 and 3 present a summary of the model simulations conducted and the parameters modified in each simulation. Several model simulations varying the hydrochar moisture content (%) (SA1) and the energy yield (%) (SA2) of the hydrochar were performed to understand their influence on the environmental impacts caused by scenarios in which HTC is practiced. To quantify results from these scenarios, sensitivity ratios (SRs), defined as the percent change of the result divided by the percent change of the parameter, were subsequently calculated for each parameter varied. SRs were used to quantify how the uncertainty associated with those parameters may contribute to each impact category.

Simulations were also conducted to understand how increases in energy recovery efficiencies may influence the LCA of the TPOMW thermal management treatments. Data associated with a potential future biowaste incinerator reported by Jungbluth et al. (2007) were used to model and simulate potential future scenarios (labeled as T.X.F). Table 4 summarizes the energy recovery efficiencies and the electricity consumption of the biowaste incineration in the base case (reference value) and the future perspective (new value). Since simulating the future perspective involves the variation of several parameters, the results obtained were only compared with those obtained in the base case, and no sensitivity ratios were determined. Additional model simulations were performed to illustrate how changing the heat and electricity production efficiencies of the future biowaste incinerator (SA3 and SA4) may affect overall results to understand the impact of each process.

3. Results and discussion

3.1. LCA analysis of currently used technologies

LCA results for each evaluated impact category associated with all modeled waste treatment approaches are reported in Figures 4-6. A positive impact potential indicates a burden to the environment (negative environmental effects), while a negative potential indicates environmental emissions savings (positive environmental effects).

3.1.1. Biological treatment approaches

Results indicate that all biologically based treatment scenarios (B.1, B.2, and B.3) result in a net cost to the environment for all evaluated impact categories, except for FEP (Figures 4-6). The net environmental savings associated with the FEP impact category results almost entirely (99-100%) from benefits associated with using the compost as a fertilizer (Figure 5). Using composted organics in this manner ultimately reduces the need for mineral fertilizer production, and thus the need for mining virgin phosphorus, which also imparts a positive impact on the GWP and POF categories (Figure 4 and 5). Conversely, using composted organics as a fertilizer results in ammonia emissions to the atmosphere, negatively contributing to the AP, TEP and MEP environmental impacts. Furthermore, heavy metal emissions (mainly zinc and copper, which are present in the TPOMW, see Table SI-1 in the SI) resulting from land application of the compost ultimately increase the HT-NC and ET environmental potentials. These results suggest that when utilizing biological processes to treat TPOMW, additional treatment of the composted digestates may be required before land application. Investigating such treatment appears advantageous because of the high potential for environmental savings associated with reducing the requirement for virgin mineral fertilizer.

Energy recovery from generated biogas also represents a significant environmental benefit (scenarios B.2 and B.3, Figures 2 and 4-6). The energy recovered from biogas conversion in a CHP engine offsets emissions associated with the need for non-renewable energy production. When recovering energy in scenario B.2 (Figure 2b), reductions in HT-NC (94%), HT-C (93%), ET (89%), MEP (58%), TEP (16%) and GWP (11%) impact potentials result when compared with scenario B.1. These reductions resulting from energy recovery represent the greatest benefit for all impact potentials, except for GWP and FEP. Using the composted digestate in place of mineral fertilizers results in the greatest reduction of the GWP and FEP impact potentials. Not surprisingly, all energy-related benefits significantly increase when the TPOMW is catalytically oxidized through the Fenton process (scenario B.3) to improve its biodegradability. This increase in energy-related benefits is illustrated by greater reductions in the HT-C (95%), MEP (76%), TEP (58%), AP (56%), POF (43%) and GWP (29%) impact potentials associated with scenario B.3 when compared to scenario B.1. Scenario B.3 exhibits the lowest burden among the biological treatments because of the higher electricity production from the biogas combustion, although a net cost to the environment remains. Overall, these results indicate that when selecting biological treatment approaches, it is critical that the biogas be collected and used for energy to decrease system environmental burden.

A significant cost to the environment associated with the anaerobic digestion of TPOMW and subsequent aerobic composting of the digestate is related to process energy needs. Each process requires the use of diesel and/or coal-based electricity, resulting in NO_x and SO_x emissions which increase process environmental burden. The biostabilization stage during the composting process is another source of emissions (e.g., ammonia, methane, and nitrous oxide) that contributes to process environmental burden. NO_x and SO_x emissions contribute to the total POF impact potential of scenarios B.2 and B.3. These NO_x emissions also contribute to the AP and TEP impact potentials. Ammonia emissions are the main component of the TEP impact category, and methane and nitrous oxide ultimately contribute to the environmental costs in the GWP and POF categories.

3.1.2 Thermal Treatment Approaches

When using traditional thermal treatment approaches (scenarios T.1 and T.2), drying of fresh TPOMW in a rotary dryer and pomace oil extraction result in costs to the environment (Figures 4-6). TPOMW drying occurs in both scenarios and imparts an environmental cost to all impact categories, with GWP most affected due to the large amount of energy required to dry the waste stream. The environmental costs associated with pomace oil extraction (scenario T.2) also largely result from energy consumption. In addition, hexane emissions as a result of the pomace oil extraction process contribute to the GWP, AP, TEP, ET and POF impact potentials. The majority of this impact is due to actual hexane emissions, which result from residual hexane found in the exhausted TPOMW. The majority of the POF impact potential is caused by these hexane emissions. Hexane manufacturing (e.g., heptane, hexane and aliphatic, alkane and cyclic hydrocarbons) also contributes to the environmental impact of this process. Only 1% of the POF impact potential is associated with gas emissions resulting from hexane manufacturing. Hexane manufacturing also contributes to the GWP, AP, ET and TEP impact potentials. The GWP category is increased because of the energy required to manufacture hexane, while the process specific emissions associated with the manufacturing process (e.g., heptane, hexane, aliphatic, alkane, and cyclic hydrocarbons) influence the AP, ET and TEP potentials.

A net environmental savings associated with all impact categories, except for FEP, results when incinerating TPOMW with energy recovery (scenario T.1). Environmental costs associated with incineration are mainly attributed to the pollutant emissions (e.g., non-methane volatile organic compounds, NMVOC), particulates, sulfur dioxide, and nitrogen oxides) and material requirements (e.g., natural resources and chemicals used in the gas cleaning). However, the environmental benefits associated with substituting coal-based electricity with energy recovered from the incineration of the TPOMW (Figures 4-6) outweigh these costs. The environmental costs associated with the FEP impact are related to the phosphorus content in the TPOMW, which is emitted to the water compartment. Since the phosphorus content is not degraded during waste drying or pomace oil extraction, the influence of phosphorus to FEP category is the same in scenarios T.1 and T.2.

It should be noted that although pomace oil extraction contributes to system environmental costs, recovery of this value-added product represents a potential economic benefit that is not accounted for in this LCA. In addition, it is important to note that the environmental costs associated with pomace oil extraction are offset by the benefits associated with energy recovery. Therefore, if pomace oil is extracted, it is critical that energy recovery occurs.

3.1.3. Comparison between currently used biological and thermal treatment approaches

Overall, thermal treatment of TPOMW results in reducing the climate change (GWP), ecosystem quality (except for FEP), and human toxicity (Figures 4-6) impact potentials. Conversely, biological treatment approaches result in environmental costs to these impact potentials, except for FEP (Figures 4-6). Environmentally favorable results are achieved when using a thermal treatment approach because of the significant environmental credits obtained from electricity production and its subsequent replacement of coal-based electricity. These results suggest that energy generation and subsequent recovery is critical to ensure environmental savings associated with the climate change, the ecosystem toxicity and the human toxicity potentials of the scenarios studied occur.

3.2. LCA analysis of Hydrothermal Carbonization

3.2.1. Environmental impact of hydrothermal carbonization and influence of carbonization reaction temperature

When using HTC to treat TPOMW, an overall environmental savings results, except for the ET and FEP impact categories (Figures 4-6). Environmental costs associated with the HTC process itself are a result of electricity requirements and liquid and gas-phase emissions. The HTC process at all evaluated temperatures (200, 225, and 250^{o}C) contributes most significantly to the FEP and ET impact categories because of liquid-phase discharge to the environment (Figure 6). Liquid-phase nutrient emissions to the surface water represent 100% of the FEP impact potential, while liquid-phase metal emissions (zinc and copper, present in the TPOMW, see Table SI-1 in the SI) represent the largest influence (99%) on the ET impact category. Released liquid-phase zinc also imparts a contribution to the HT-NC impact potential (24-33%). It should be noted that although in these scenarios the HTC-liquid was not treated before its discharge, such treatment will be required to meet regulatory discharge limits. Berge et al. (2015) reported that if 90% of the liquid-phase contaminants are removed, the environmental impact associated with HTC can be significantly reduced. The gas-phase emissions associated with HTC are mostly composed of biogenic carbon dioxide (88-93%, vol.), and thus do not represent a significant environmental impact. However, a more extensive evaluation of gas-phase composition associated with HTC is needed.

The electricity required to run the HTC process represents only a small contribution to the overall environmental impacts, contributing to the GWP (8.3%), HT-C (2.1%), POF (3.0%), AP (4.4%) and TEP (2.0%) impact categories. It is important to note that the environmental impacts caused by the HTC process itself may change when scaling-up the HTC and mechanical dewatering processes due to changes in requirements for auxiliary equipment requiring diesel oil, electricity, and/or chemicals, all of which were not considered in this study.

As carbonization temperature increases, the environmental costs of the HTC process increases (Figures 4-6). Changes in reaction temperature influence energy requirements for reactor heating and volume of discharged HTC process water (see Table SI-2). When increasing the reaction temperature from 200ºC to 250ºC, impact potentials associated with the HTC process increase, although they remain insignificant on overall environmental impact.

Hydrochar drying, which is represented in the rotary dryer category in Figures 4-6, following carbonization represents a small environmental impact to each scenario and, importantly, presents a significantly lower contribution to the impact categories than that of TPOMW drying in scenarios T.1 and T.2. This reduction in impact results because the moisture content of the solid (hydrochar) following HTC is lower than that associated with raw TPOMW (Tables SI-1 and SI-2). As HTC reaction temperature increases, the contribution of hydrochar drying decreases because hydrochar hydrophobicity increases and consequently less moisture is retained in the solids. The environmental impact associated with hydrochar drying decreases by almost 80% in all impact categories when increasing the HTC reaction temperature from 200ºC to 250ºC.

Similar to energy recovery in the biological and thermal treatment approaches discussed previously, the energy recovered from hydrochar combustion represents an important and significant environmental savings; the GWP (-81.5%), HT-C (-96.0%), HT-NC (-76.6%), POF (-92.9%), AP (-91.0%), TEP (-96.3%), and MEP (-96.2%) impact categories (Figures 4-6) are influenced. These savings are significant enough to overcome the aforementioned burdens attributed to the HTC process (e.g., energy consumption and gas emissions, excluding liquid emissions) and hydrochar drying, indicating that the energy offsets associated with electricity production from hydrochar combustion play an important role in the environmental implications of the HTC process. The percent of energy initially present in the TPOMW that is recovered in the hydrochar, which is dependent on both hydrochar energy content and mass yield, reaches a maximum at 225ºC (Table 2 in the SI, Benavente et al., 2015) and, as a consequence, greater environmental benefits are obtained at this temperature. Approximately 4% and 18% more electricity are produced from the hydrochar generated at 225°C than that at 200ºC and 250ºC, respectively. Therefore, scenario T.3.225 exhibits the lowest overall impact potentials in all categories, except for ET. The large impact associated with the ET category remains because liquid emissions remain significant.

3.2.2. Comparison between scenarios with HTC and other currently used treatment approaches

A ranked comparison of all evaluated scenarios for all evaluated impact categories is presented in Table 5. Results from scenarios in which HTC is used to overcome the technical challenges biological and thermal treatment approaches pose indicate that environmental savings in the GWP impact potential occur; however, such savings are not as significant as those obtained with traditional thermal treatment approaches (scenario T.1 and T.2). This finding results because the energy content of the hydrochar is approximately 45-35% smaller than that associated with dried TPOMW, which results in the recovery of less energy. It should be noted, however, that the HTC of TPOMW and subsequent incineration of the hydrochar does result in a net environmental benefit, and is shown to be more environmental advantageous than any of the biological treatments analyzed from a climate change perspective. No significant differences are observed between HTC of TPOMW and subsequent incineration of the hydrochar (scenarios T.1 and T.2) regarding the impact categories associated with human toxicity (HT-C, HT-NC, and POF).

When comparing the ecosystem quality impact potentials, scenarios T.1 and T.2 remain more environmentally advantageous (Figures 4 and 5) than the scenarios with HTC. Greater FEP, MEP, and ET emissions savings result in scenarios T.1 and T.2 because of greater electricity production. In addition, HTC of TPOMW represents a burden to the ecosystem quality due to the HTC liquid-phase discharge to surface waters. When compared with biological treatment approaches, it is found that the ET impact potential of the HTC scenarios is lower than that determined for scenarios B.1, B.2, and B.3.

Most environmental costs associated with the integration of the HTC process within a TPOMW treatment scheme are related to the liquid phase discharge to the environment. As previously stated, adequate treatment of the HTC liquid before its discharge may significantly decrease the magnitude of these impact potentials. Wirth and Mumme (2013) showed that anaerobic digestion was a suitable process to treat HTC wastewaters. The properties of the HTC process water obtained from TPOMW carbonization are similar to the properties of olive mill wastewaters (OMWW). Therefore, it is expected that anaerobic digestion may also be effective for the treatment of wastewaters produced from TPOMW carbonization. Treatment processes that have been found to be useful in treating OMWW may also be used to treat this liquid stream (e.g., Aly et al., 2014; Sponza and Oztekin et al., 2014). It is important to note that emissions associated with HTC process water treatment will also contribute to the system environmental impact. However, results from previous studies suggest that these processes may cause significantly less environmental impacts than the direct discharge of HTC wastewaters (Hong et al., 2010).

The reduction in energy contained in the solid during HTC is another other key factor affecting the global energy efficiency, and consequently the environmental savings associated with scenarios T.3.200-250. Although the HTC technology is more energetically advantageous and ultimately alleviates some operational and economic challenges associated with current thermal treatment processes, the amount of energy produced from the combustion of the hydrochar is lower than that produced from the direct combustion of the TPOMW. Research focusing on maximizing solids yields and hydrochar energy contents is needed to potentially reduce this difference.

Overall, these results suggest that thermal treatment approaches (traditional and with HTC) out-perform biologically based approaches. It should be noted that several LCA simulations were conducted to evaluate the importance of various key parameters associated with biological treatment approaches, as described in the supplementary information (Section 2, Table SI-4, Figures SI-1 and SI-4). However, even when values for these key parameters were chosen to minimize system environmental impacts, scenarios B.1, B.2, and B.3 remain less favorable than thermal treatment approaches with and without HTC.

3.3. Sensitivity analyses

3.3.1. Influence of the energy recovery efficiency during thermal treatment

Simulations were conducted to assess how changes in energy-related parameters associated with incineration may influence system environmental impact. First, gross electric and thermal energy efficiencies were modified according to values corresponding with a potential future biowaste incinerator (Jungbluth et al., 2007). In these future scenarios, the thermal energy efficiency is projected to greatly increase because some additional heat energy can be generated from the flue gas by means of a heat pump and lowering of the flue gas temperature after condensation. However, electric efficiencies are projected to only slightly increase.

The individual impact potentials resulting from this future scenario analysis are presented in Figure 7, and indicate that changes in the efficiencies associated with electricity and heat generation impart a significant influence on all impact categories associated with these thermal management approaches. As the total energy efficiencies increase, the overall impact potentials decrease because greater emission offsets are obtained from the larger electrical and thermal energy production. The GWP impact potentials are reduced by 70-83% in all scenarios studied. This change in energy efficiency also results in corresponding reductions in environmental impact potentials related to human toxicity and ecosystem quality.

When comparing the results from the future scenarios (T.1.F, T.2.F, T.3.200.F, T.3.225.F, T.3.250.F) with their corresponding base case scenarios (T.1, T.2, T.3.200, T.3.225, T.3.250), interesting observations associated with electricity and heat production/requirements result (Table 4). For each base case scenario, the heat produced is less than total heat consumption (as reflected by negative net heat production values); therefore, an industrial furnace operated with natural gas is required. When HTC is implemented, less heat is required for TPOMW drying and as a consequence, heating energy savings results, even when the heat produced from the hydrochar is lower than that produced from the TPOMW (as seen in Table 4). This ultimately reduces the environmental impacts associated with the auxiliary natural gas-based heat supply. When considering the future perspective, however, all scenarios result in net heat production that can be used for beneficial purposes at/nearby the facilities; 28% and 10% of the heat produced could be exported for other uses if practicing scenarios T.1.F and T.2.F, respectively, while 14%, 21% and 29% of excess heat is produced in scenarios T.3.200, T.3.225 and T.3.250, respectively.

In instances in which the excess heat produced cannot be used, it would be more beneficial to produce larger amounts of electricity than heat. Two model simulations were conducted to evaluate how decreasing heat production while increasing electricity production influence overall impact potentials. In the first analysis (SA3), the heat recovery efficiency corresponding to the future biowaste incinerator was decreased so that only the amount of heat required in each scenario is generated, as illustrated in Tables 3 and 4. In the second analysis (SA4), the heat recovery efficiency used in analysis SA3 was coupled with the electricity recovery efficiency equivalent to the future biowaste incinerator, as shown in Table 3. Results from these analyses are illustrated in Figures 7 and 8 and indicate that changes in heat and electricity recovery efficiencies do not have the same effect on the environmental impact results. A reduction in the heat recovery efficiency imparts a smaller influence on the environmental impact of the thermal treatments than the same increase in the electricity recovery efficiency. Decreases in the heat generation efficiency (SA3) lead to a slight reduction of all the environmental impacts, with GWP being the most affected impact potential (Figures 7 and 8). However, when increasing the electricity recovery efficiency (SA4), the associated environmental impacts are greatly reduced, indicating that these environmental savings are more significant. These results are not surprising, as offsetting emissions associated with coal-based electricity are likely greater than the offsetting emissions associated with natural gas and heat. In addition, it is also observed that the environmental impacts associated with scenario T.1 and scenarios with HTC are equivalent when the electricity production efficiency is increased, with the exception of the ET impact potential, which is associated with the HTC-liquid emissions. This result is significant and suggests that when electricity efficiencies are high, HTC is likely the preferred approach, noting that using HTC to treat TPOMW alleviates some of the operational challenges associated with TPOMW incineration.

3.3.2. Influence of hydrochar properties: moisture and energy content

There is a significant amount of uncertainty associated with hydrochar moisture content following liquid drainage. This is an important parameter because it impacts the amount of energy required for hydrochar drying and can vary greatly depending on the mechanical dewatering technology employed (i.e., filter presses or centrifuges). Therefore, a sensitivity analysis was performed to understand the influence of hydrochar moisture content on the overall environmental impacts of scenarios T.3.200, T.3.225 and T.3.250. Results from this analysis are illustrated in Figure 8a and indicate that increases in hydrochar moisture content negatively affect system environmental impacts for all impact categories that are influenced by liquid-phase emissions (ET, MEP, and HT-NC). Increases in hydrochar moisture content result in lower liquid-phase discharges, imparting a positive effect on the environmental impacts associated with the ET, MEP, and HT-NC categories. Additionally, results from this analysis indicate that system environmental impact is not very sensitive to changes in the electricity required for liquid evaporation. These results are important when considering possible industrial applications, as the dewatering process should consider both energy and environmental implications.

Another analysis was conducted to determine the percentage of energy initially present in the TPOMW that needs to be recovered in the hydrochar to match the GWP impact potential results obtained when incinerating the TPOMW. Results from this analysis indicate that the minimum amount of energy recovered from the initial TPOMW in the hydrochar needs to be 91% for scenarios T.3.200 and T.3.225 and 86% for scenario T.3.250. These differences result because of the improvement of the dewatering properties of the hydrochar with increasing temperature. If achieving these energy recoveries during HTC is possible, HTC of TPOMW and subsequent incineration of the hydrochar would be as environmental beneficial as direct incineration of TPOMW, with the added advantage of overcoming the operational and economic limitations associated with the currently used processes. These results suggest that research focusing on improving hydrochar energy content and solids yields from HTC would be advantageous.

4. Conclusions

LCA was used to conduct a system level analysis to determine the environmental impacts associated with currently used TPOMW management approaches and to compare them with the impacts associated with the use of HTC to treat the TPOMW. Results indicate that the largest environmental impacts arise from the biological treatment of TPOMW. The most environmentally friendly TPOMW management option is scenario T.1, which includes waste drying and incineration, followed by scenario T.2, which involves drying, pomace oil extraction, and incineration. It is important to note, however, that despite having greater environmental benefits, these alternatives pose some operational and economic challenges caused by the high moisture content of the TPOMW. The benefit of using HTC to overcome these challenges is not reflected in this LCA. Additional analyses in which benefits associated with these operational and economic challenges are considered also need to be conducted.

Results from this work also indicate that the hydrothermal carbonization of TPOMW followed by hydrochar combustion with energy recovery results in net environmental savings for all impact categories with the exception of FEP and ET, which are mainly attributed to the untreated HTC process water discharge to the environment. Improving the fraction of energy initially present in the TPOMW that remains in the hydrochar and treating the liquid emissions from the HTC process will result in reducing system environmental impact, resulting in scenarios with HTC being as environmentally beneficial as current thermal treatment approaches. Thus, it is recommended that future research efforts focus on the evaluation of appropriate and environmentally beneficial HTC process water treatment approaches and methods to improve the energy retention efficiencies of the hydrochar.

Acknowledgements

Author V. Benavente kindly wishes to thank the Conselleria d’Educación, Cultura i Esport, for a Ph.D. grant (contract grant number ACIF/2014/275) and a pre-doctoral employee stays grant (grant number BEFPI/2015/062).

Appendix A. Supplementary data

Abbreviation list

TPOMW – Two Phase Olive Mill Waste

AC – Aerobic Composting

AD – Anaerobic Composting

HTC – Hydrothermal carbonization

LCA – Life Cycle Analysis

B – Biological treatment

T – Thermal Treatment

GWP – Global Warming Potential

AP – Terrestrial Acidification

TEP – Terrestrial eutrophication

HT-C – Human Toxicity, Carcinogenic

HT-NC – Human Toxicity, Non-Carcinogenic

POF – Photochemical Oxidant Formation

MEP – Marine Eutrophication

FEP – Fresh water Eutrophication

ET – Ecotoxicity

SA – Sensitivity Analysis

SR – Sensitivity Ratio

CHP – Combine Heat and Power engine

COD – Chemical Oxygen Demand

TOC – Total Organic Carbon

OMWW – Olive Mill Waste Water

NMVOC – Non-Methane Volatile Organic Compounds

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Table 1. Impact categories used in the impact assessment.

Impact category	Method	Abbreviation	Unit/kg
Climate change	IPCC 2007 (Forster et al., 2007))	GWP	kg CO2-eq.
Human toxicity, cancer effect[a]	USEtox (Rosembaum et al., 2008)	HT-C	CTU_h
Human toxicity, non-cancer effect[a]	USEtox (Rosembaum et al., 2008)	HT-NC	CTU_h
Photochemical ozone formation	ReCiPe midpoint (Van Zelm et al., 2008)	POF	kg NMVOC-eq.
Terrestrial acidification[c]	Accumulated exceedance (Seppala et al., 2006; Posch et al., 2008)	AP	AE
Terrestrial eutrophication	Accumulated exceedance (Seppala et al., 2006; Posch et al., 2008)	TEP	AE
Freshwater eutrophication	ReCiPe midpoint (Struijs et al., 2009)	FEP	kg P-eq.
Marine eutrophication	ReCiPe midpoint (Struijs et al., 2009)	MEP	kg N-eq.
Freshwater ecotoxicity	USEtox (Rosembaum et al., 2008)	ET	CTU_e

[a] CTU_h: comparative toxic unit for humans.

[b] CTU_e: comparative toxic unit for ecosystem.

[c] AE: accumulated exceedance (keq).

Table 2. Summary of model simulations conducted in EASETECH.

Scenario	Label	Description
COMPOSTING	B.1	Base case
ANAEROBIC DIGESTION	B.2	Base case
	B.3	Base case with pretreatment of TPOMW
INCINERATION WITH ENERGY RECOVERY	T.1	Base case
	T.1.F	Future perspective
	T.1.F.SA3	Lower heat production
	T.1.F.SA4	Larger electricity production
SECOND EXTRACTION OF POMACE OIL AND INCINERATION WITH ENERGY RECOVERY	T.2	Base case
	T.2.F	Future perspective
	T.1.F.SA3	Lower heat production
	T.1.F.SA4	Larger electricity production
HTC FOR SUBSEQUENT ENERGY PRODUCTION	T.3.200	Base case (HTC 200ºC)
	T.3.225	Base case (HTC 225ºC)
	T.3.250	Base case (HTC 250ºC)
	T.3.200.SA1	+25% hydrochar moisture (HTC 200ºC)
	T.3.225.SA1	+25% hydrochar moisture (HTC 225ºC)
	T.3.250.SA1	+25% hydrochar moisture (HTC 250ºC)
	T.3.200.SA2	Change in HTC-char energy yield (HTC 200ºC)
	T.3.225.SA2	Change in HTC-char energy yield (HTC 225ºC)
	T.3.250.SA2	Change in HTC-char energy yield (HTC 250ºC)
	T.3.200.F	Future perspective (HTC 200ºC)
	T.3.225.F.SA3	Change Heat eff. (HTC 225ºC)
	T.3.250.F.SA3	Change Heat eff. (HTC 250ºC)
	T.3.200.F.SA4	Change Electricity eff. (HTC 200ºC)
	T.3.225.F.SA4	Change Electricity eff. (HTC 225ºC)
	T.3.250.F.SA4	Change Electricity eff. (HTC 250ºC)

Table 3. Parameters modified in the models simulations conducted.

Label	Parameter	Reference Value	New Value	Percent change (%)
SA1	HTC-char moisture at 200 °C (%)[*a]	60	75	+25
	HTC-char moisture at 225 °C (%)[*a]	55	69	+25
	HTC-char moisture at 250 °C (%)[*a]	27	34	+25
SA2	HTC-char energy yield at 200 °C (%)[*b]	65	91	+40
	HTC-char energy yield at 225 °C (%)[*b]	68	91	+35
	HTC-char energy yield at 250 °C (%)[*b]	55	86	+55
F	Future perspective*[c]			
	Gross electric energy efficiency (%)	13	17	+31
	Gross thermal energy efficiency (%)	26	56	+115
	Gross total efficiency (%)	39	73	+87
	Electricity consumption (kWh/kg)	0.144	0.100	-31
SA3	Heat recovery efficiency, Heat eff (%)[*d]			
	T.1.F (%)	56	40	-29
	T.2.F (%)	56	50	-11
	T.3.200.F (%)	56	43	-23
	T.3.225.F (%)	56	40	-29
	T.3.250.F (%)	56	36	-36
SA4	Electricity recovery efficiency, Elect eff (%)[*e]			
	T.1.F (%)	17	33	+94
	T.2.F (%)	17	23	+35
	T.3.200.F (%)	17	30	+76
	T.3.225.F (%)	17	33	+94
	T.3.250.F (%)	17	37	+118

New values from:

[a*] Arbitrary values.

[b*] Energy recovery determined to make HTC as beneficial as the best case scenario.

[c]* Jungbluth et al., 2007

[d*] Heat recovery efficiency determined to cover the heat requirements of the scenario, defined as *breakeven heat requirement (%)* (see Table 9).

[e]*Electricity recovery efficiency determined: (Elect eff)$_{\text{Ref. value}}$ (%) + (ΔHeat eff)$_{\text{SA5}}$ (%).

Table 4. Summary of energy requirements and energy production in thermal treatment approaches (with and without HTC).

Current Perspective	T.1	T.2	T.3.200	T.3.225	T.3.250
Energy available from the combustion of the waste (MJ/kg OMWin)	7.44	6.31	4.85	5.03	4.12
Electricity production (kWh/kg OMWin)	0.27	0.82	0.63	0.65	0.54
Total electricity consumption (kWh/kg OMWin)	0.05	0.05	0.03	0.03	0.02
Net electricity production (kWh/kg OMWin)	0.22	0.78	0.6	0.63	0.51
Heat production (MJ/kg OMWin)	1.9	1.61	1.24	1.29	1.05
Total heat consumption (MJ/kg OMWin)	3	3.17	2.1	2.01	1.47
Net heat production (MJ/kg OMWin)	-1.1	-1.56	-0.86	-0.72	-0.42
Breakeven heat requirement (%)*	40	50	43	40	36
Future Perspective	T.1.F	T.2.F	T.3.200.F	T.3.225.F	T.3.250.F
Energy available from the combustion of the waste (MJ/kg OMWin)	7.44	6.31	4.85	5.03	4.12
Electricity production (kWh/kg OMWin)	0.35	1.07	1.1	1.14	0.93
Total electricity consumption (kWh/kg OMWin)	0.03	0.04	0.02	0.02	0.02
Net electricity production (kWh/kg OMWin)	0.31	1.04	1.08	1.12	0.92
Heat production (MJ/kg OMWin)	4.19	3.55	2.44	2.53	2.08
Total heat consumption (MJ/kg OMWin)	3	3.17	2.1	2.01	1.47
Net heat production (MJ/kg OMWin)	1.19	0.38	0.35	0.53	0.61
Breakeven heat requirement (%)*	40	50	43	40	36

* Breakeven heat requirement (%) = (Total heat consumption/energy available from the combustion of the waste)*100

Table 5. Ranking of the alternatives.

Order of Env. Impact		GWP	HT-C	HT-NC	POF	AP	TEP	FEP	MEP	ET
Best	1	T.1 (-)	T.1 (-)	T.1 (-)	T.1 (-)	T.1 (-)	T.1 (-)	B.2 (-)	T.1 (-)	T.1 (-)
	2	T.2 (-)	T.2 (-)	T.2 (-)	T.3.225 (-)	T.2 (-)	T.2 (-)	B.3 (-)	T.2 (-)	T.2 (-)
	3	T.3.225 (-)	T.3.225 (-)	T.3.225 (-)	T.3.200 (-)	T.3.225 (-)	T.3.225 (-)	B.1 (-)	T.3.225 (-)	T.3.200 (+)
	4	T.3.200 (-)	T.3.200 (-)	T.3.200 (-)	T.2.250 (-)	T.3.200 (-)	T.3.200 (-)	T.1 (-)	T.3.200 (-)	T.3.225 (+)
	5	T.2.250 (-)	T.2.250 (-)	T.2.250 (-)	T.2 (-)	T.2.250 (-)	T.2.250 (-)	T.2 (+)	T.2.250 (-)	T.3.250 (+)
	6	B.3 (+)	B.3 (+)	B.3 (+)	B.3 (+)	B.3 (+)	B.3 (+)	T.3.225 (+)	B.3 (+)	B.1 (+)
	7	B.2 (+)	B.2 (+)	B.2 (+)	B.1 (+)	B.2 (+)	B.2 (+)	T.3.200 (+)	B.2 (+)	B.3 (+)
Worst	8	B.1 (+)	B.1 (+)	B.1 (+)	B.2 (+)	B.1 (+)	B.1 (+)	T.3.250 (+)	B.1 (+)	B.2 (+)

The annotation in parentheses indicates whether the contribution associated with the scenario is positive or negative

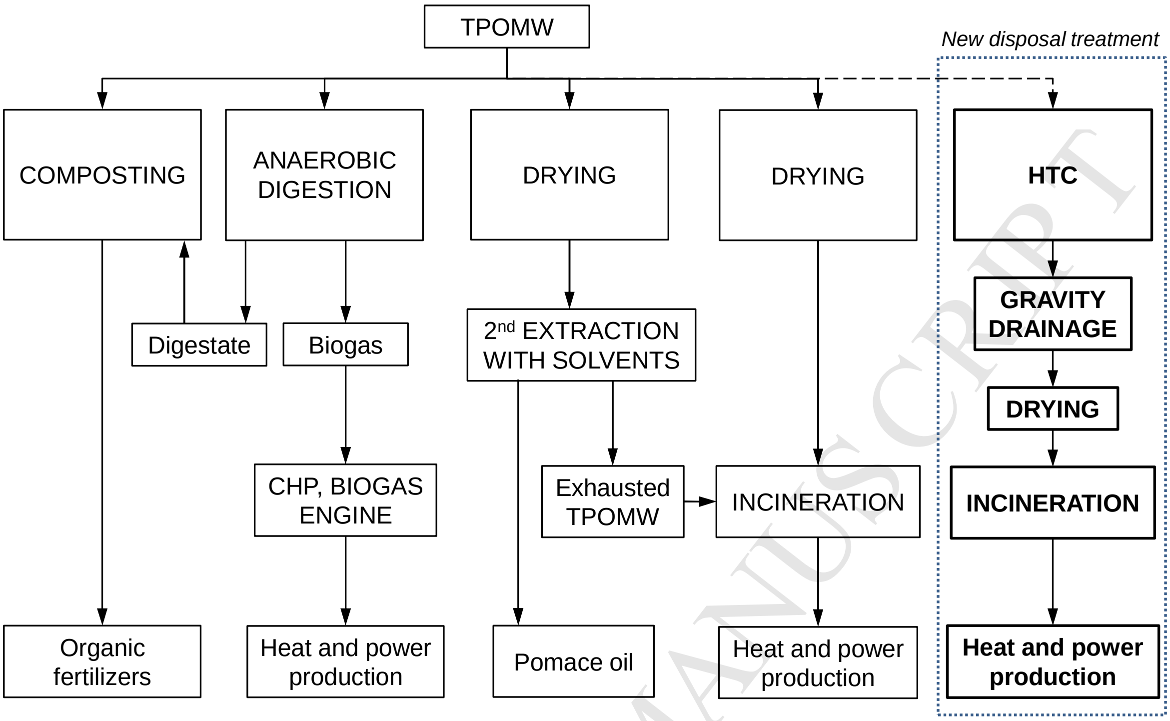

Figure 1. Currently used approaches for management of TPOMW in Spain (adapted from MME, 2000), including the proposed use of HTC as a pretreatment technique.

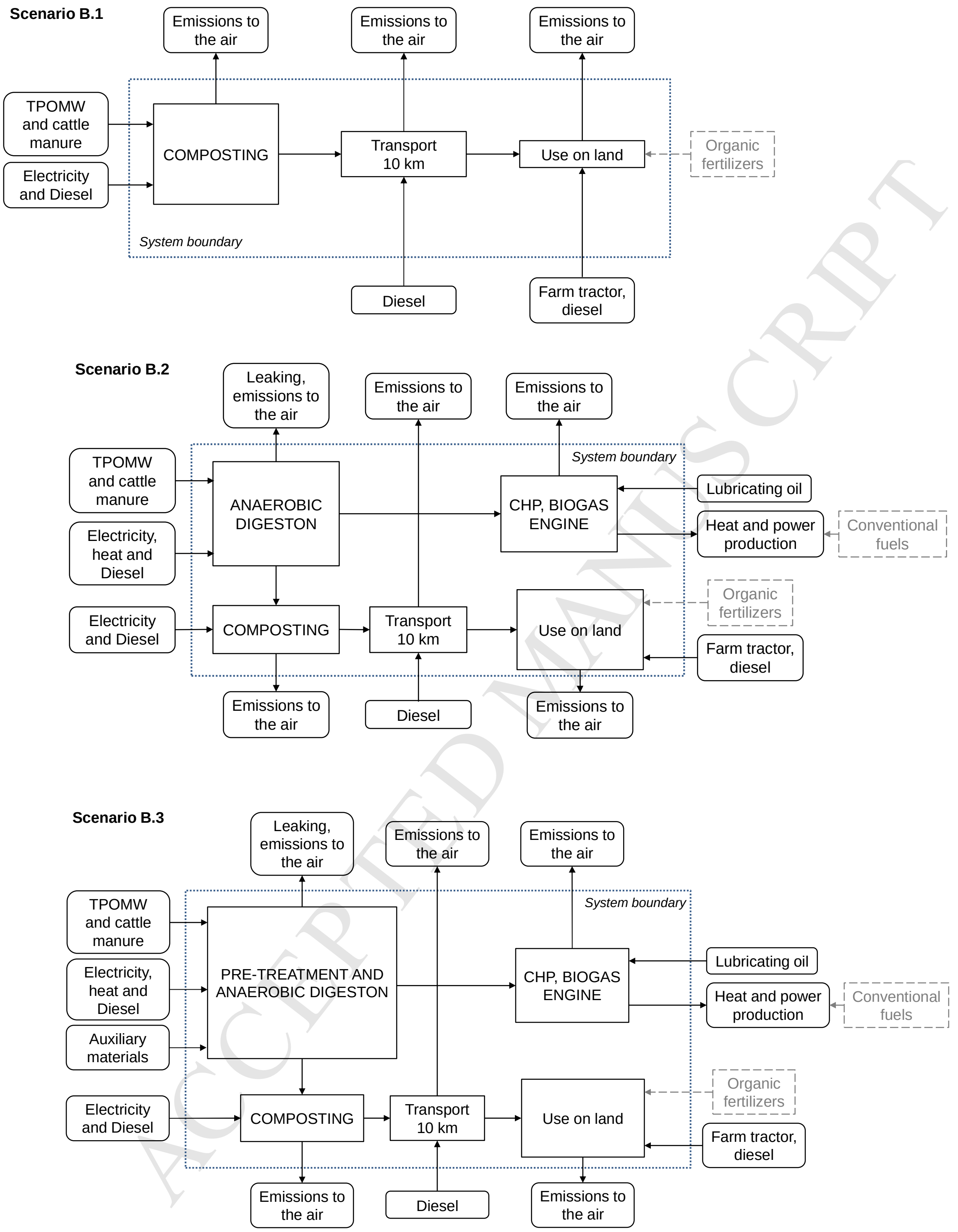

Figure 2. Modeled biological alternatives and overview of system flows: (a) Scenario B.1: Co-composting with other agricultural wastes; (b) Scenario B.2: Anaerobic digestion and composting of the digestate; and (c) Scenario B.3: Anaerobic digestion of TPOMW pre-treated using catalytic oxidation following the Fenton-like process to maximize anaerobic degradation, with subsequent composting of the digestate.

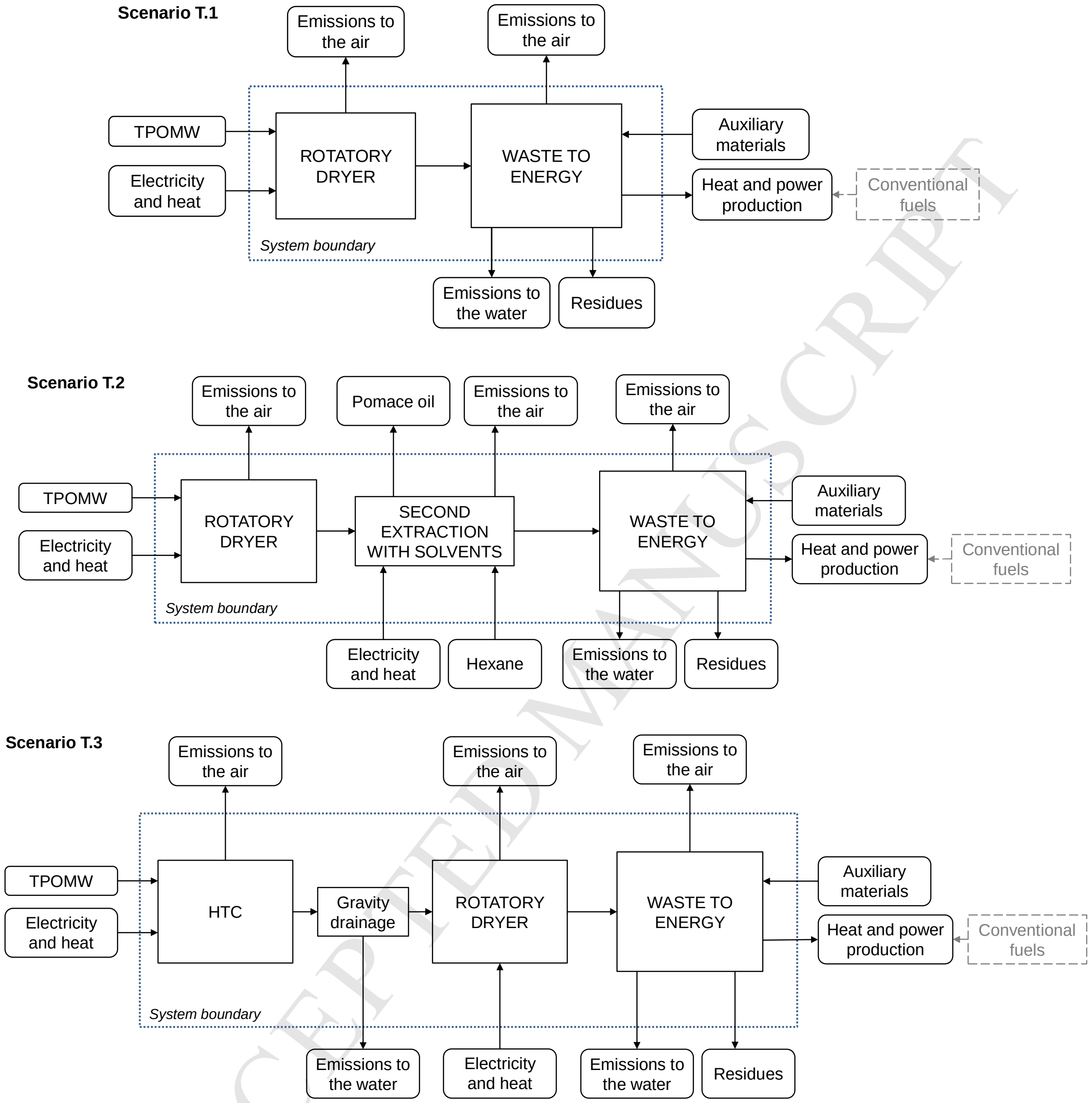

Figure 3. Modeled thermal alternatives and overview of the flows: (a) Scenario T.1: Incineration of TPOMW with energy recovery; (b) Scenario T.2: Second extraction with solvents of TPOMW and incineration with energy recovery; and (c) Scenario T.3: Hydrothermal carbonization of TPOMW and incineration of the hydrochar with energy recovery.

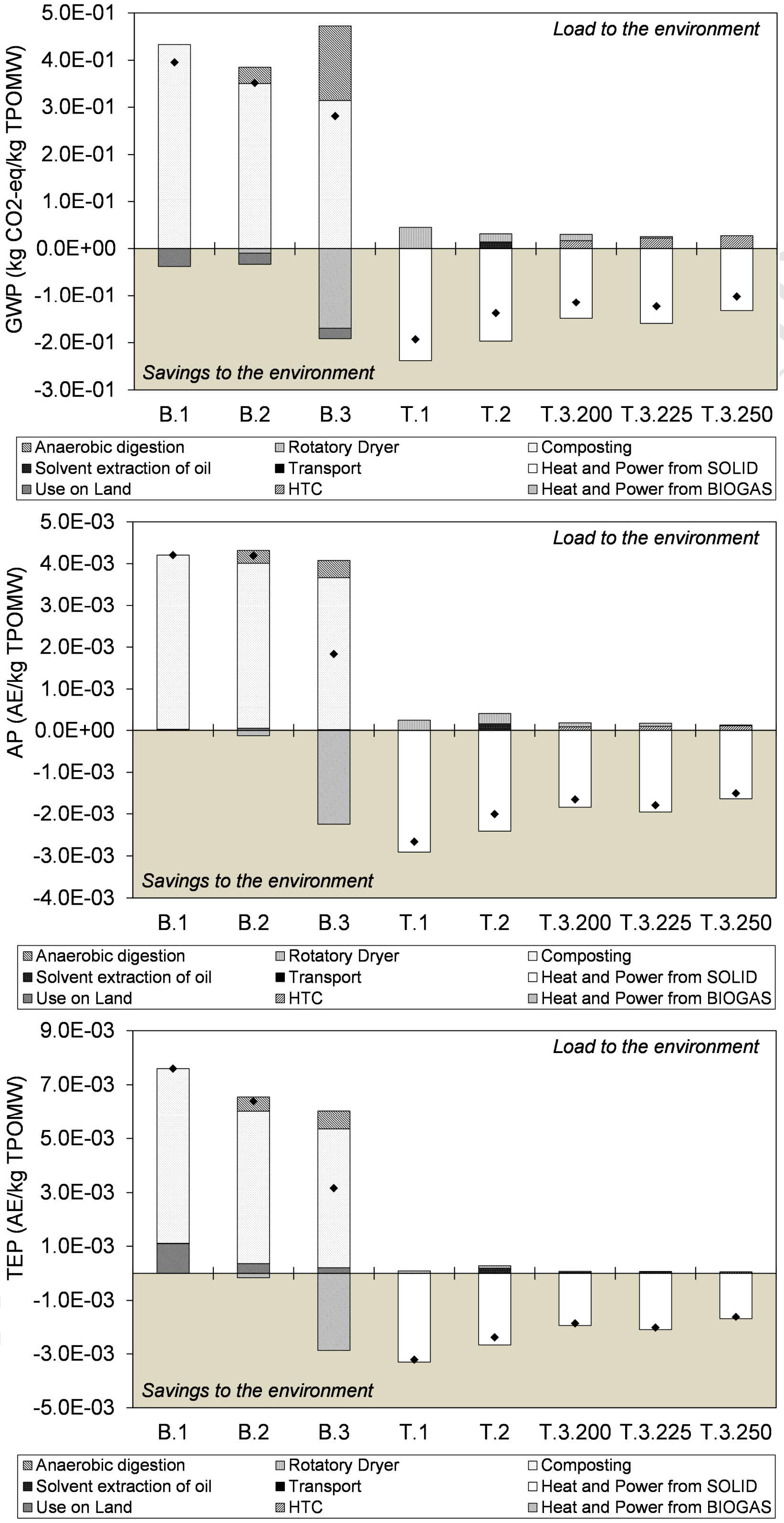

Figure 4. Climate change (GWP), and ecosystem quality (AP, TEP) impact potentials associated with the TPOMW treatment approaches.

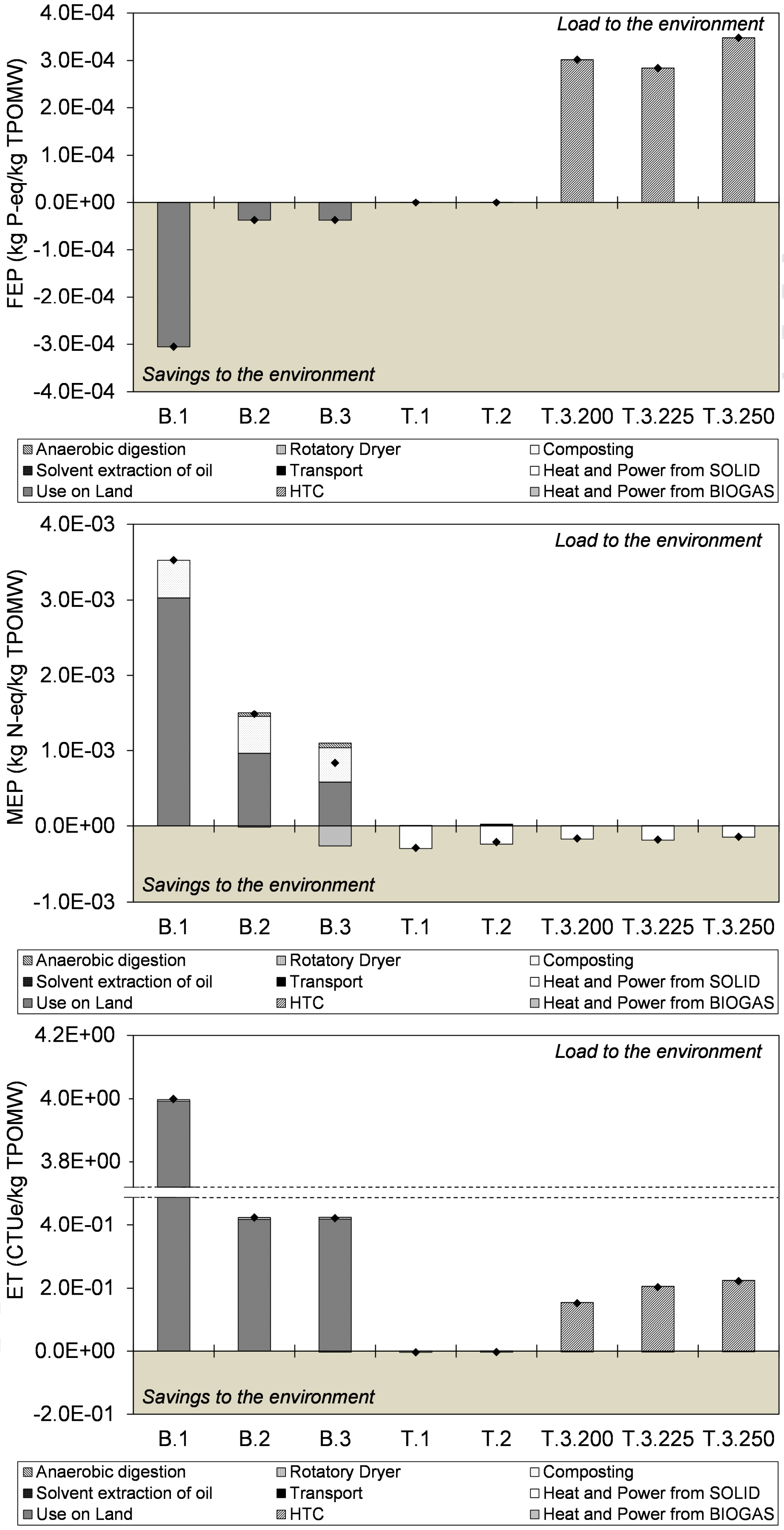

Figure 5. Ecosystem quality (FEP, MEP, ET) impact potentials associated with the TPOMW treatment approaches.

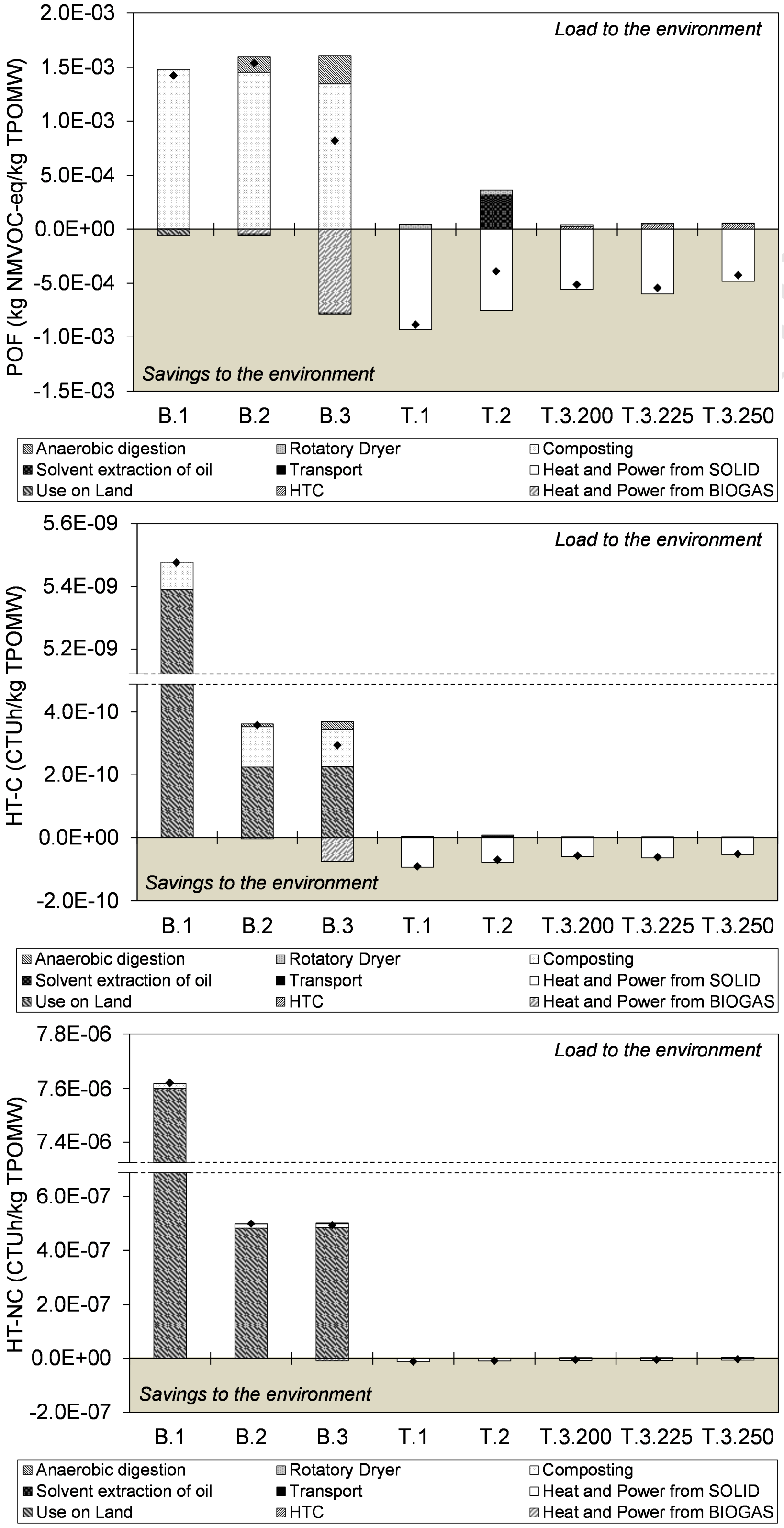

Figure 6. Human toxicity (POF, HT-C, HT-NC) impact potentials associated with the TPOMW treatment approaches.

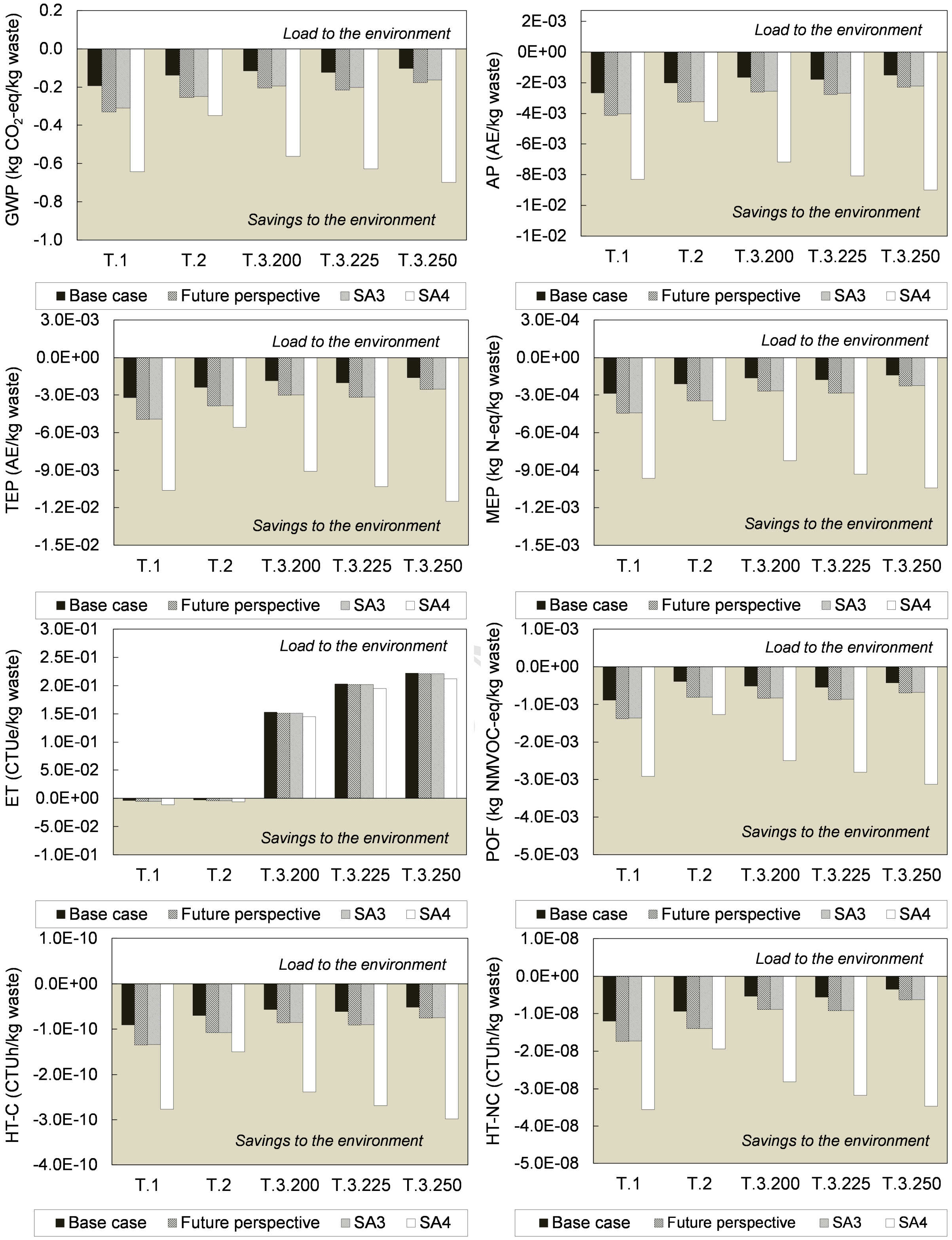

Figure 7. Differences in net climate change (GWP), ecosystem quality (AP, TEP, MEP, ET) and human toxicity (POF, HT-C, HT-NC) impact potentials due to changes in heat (SA3) and electricity (SA4) production efficiencies.

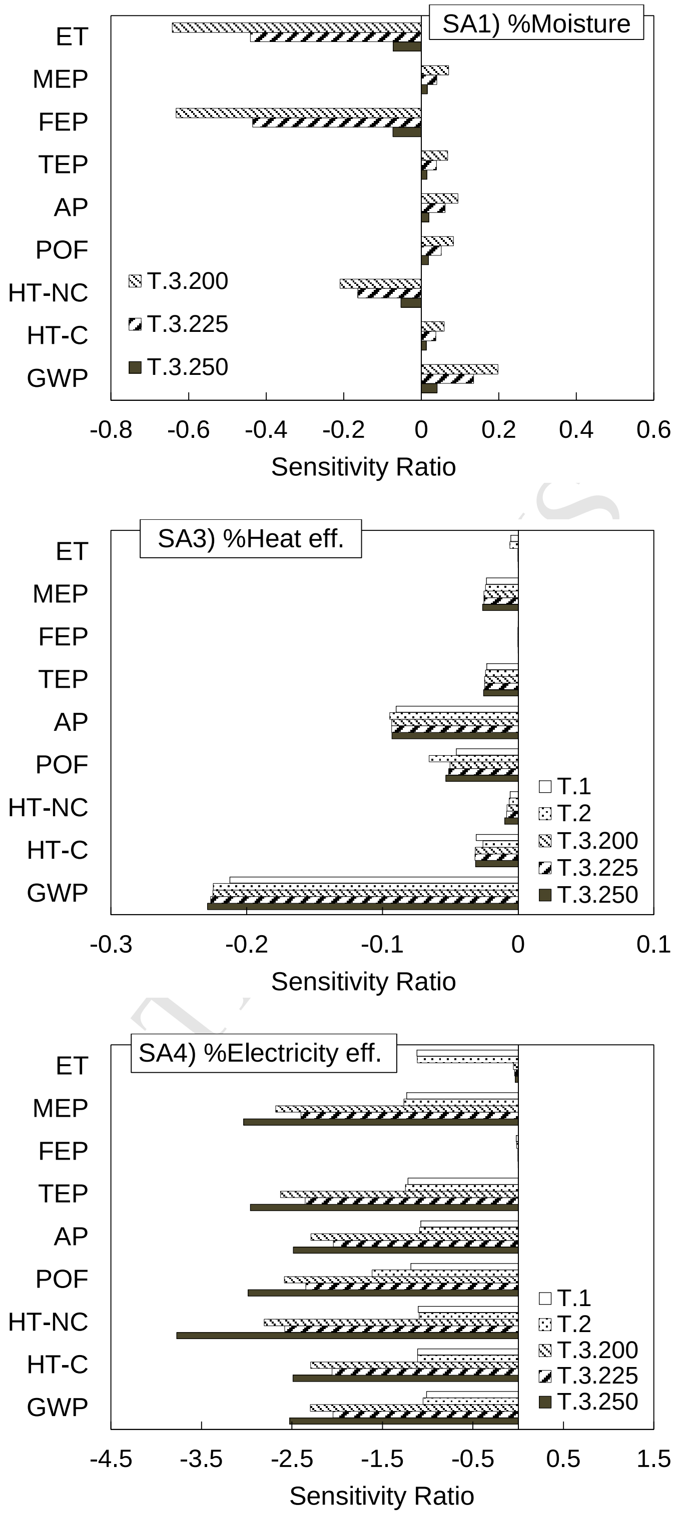

Figure 8. Sensitivity ratios associated with changing (a) % moisture of hydrochar (SA1), (b) %Heat eff (SA3) and (c) %Elect Eff (SA4) in thermal treatment scenarios.

Highlight 1: Environmental impacts associated with TPOMW treatment is evaluated using LCA

Highlight 2: Thermal treatment of TPOMW is more environmentally advantageous than biological treatment

Highlight 3: Energy recovery is critical in reducing system environmental impact

Highlight 4: Incineration with energy recovery has a lower impact than HTC, but operational challenges remain

Highlight 5: Increase hydrochar energy recovery efficiency and treat liquid emissions to reduce impact