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Life cycle assessment of a novel strategy based on hydrothermal carbonization for nutrient and energy recovery from food waste

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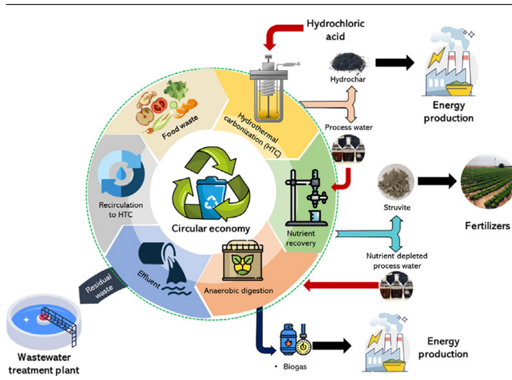
HIGHLIGHTS

- Hydrothermal treatment (HTC) + nutrient recovery + anaerobic digestion (AD) of food waste
- Comparative analysis of AD and HTC based process for food waste management
- Combined process was configured as energy self-sufficient system.
- Substitution of conventional fossil fuels by hydrochar environmentally favors the combined process.
- Struvite substitution for digestate in soil applications reduces ecological impacts.

GRAPHICAL ABSTRACT



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ABSTRACT

In this work, a novel strategy for food waste valorization was evaluated from an environmental life-cycle perspective. A system based on acid-assisted hydrothermal carbonization of food waste combined with the exploitation of hydrochar by combustion and process water through nutrient recovery stage and subsequent anaerobic digestion, was assessed and compared with stand-alone anaerobic digestion as the reference system. This combination of processes aims to recover both nutrients in a stage of struvite precipitation from process water and energy through hydrochar and biogas combustion. Both systems were modeled in Aspen Plus® to identify and quantify their most relevant input and output flows and subsequently evaluate their environmental performance through the life cycle assessment methodology. The novel combined system was found to generally involve a more favorable environmental performance than the reference stand-alone configuration, which would be closely linked to the substitution of hydrochar for fossil fuels. In addition, the impacts associated with soil application of the struvite produced in the integrated process would also be reduced compared to the use of the digestate generated in the stand-alone anaerobic digestion process. Following these results and the evolving regulatory framework for biomass waste management, mainly in the field of nutrient recovery, combined process based on acid-assisted hydrothermal treatment plus nutrient recovery stage and anaerobic digestion is concluded to be a promising circular economy concept for food waste valorization.

1. Introduction

Waste management has become a major social concern and a relevant issue for the European Commission to work within a circular economy

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framework. Food waste (FW) is a type of organic waste from any stage of the food supply chain, which contributes significantly to the fraction of municipal waste (World bank, 2018). In 2019, more than 2000 million tonnes of municipal waste were globally generated, with a FW share exceeding 50 % in many parts of the world (Kaza et al., 2018).

In the European Union (EU), current legislation in terms of biowaste management points out some actions to minimize the environmental impact of approximately 14 million tonnes of generated FW through a low-carbon and circular economy policy with reduced greenhouse gas emissions and biowaste generation (Commission, 2020). The current tendency in many EU states and in other developed countries is to reduce the amount of FW disposed in landfills, supporting an effective source separation (Browne and Murphy, 2013; Campuzano and González-Martínez, 2016).

FW management may follow different routes, including biological technologies and thermochemical pathways (Haldar et al., 2022). Anaerobic digestion (AD) is a widely established biological process that converts organic matter into two valuable products: a nutrient-rich digestate, which is increasingly restricted by current regulation but can be used in agriculture, and a methane-rich biogas that can be used to generate electricity and/or heat or upgraded to replace natural gas (Ipiales et al., 2021). With approximately 8000 biogas plants which operate with agricultural FW in Germany, 1200 in Italy, and 600 in France, biogas production plays a key role in some European energy systems to minimize the dependence on fossil fuels given the high annual availability of FW (EBA, 2021). However, AD requires a long treatment time (up to 30–40 days), and a high concentration of free ammonia (NH_3) and cations can inhibit the process (Pham et al., 2015). Anaerobic digestion of FW can present some instability due to challenges in microbial metabolism, which is associated in many cases with the accumulation of volatile fatty acids, leading to a decrease in pH if no sufficient buffering capacity is present and even process failure (Capson-Tojo et al., 2017). In addition, the low C/N ratio of FW because of its high protein and lipid content can also lead to inhibitory levels of ammonia and hydrogen sulfide, as well as foaming in the digester (Casallas-Ojeda et al., 2021). Co-digestion with sewage sludge, crop residues or lignocellulosic biomass can favor the methanogenic/acidogenic processes by balancing the nutrient and carbon content, diluting inhibitory compounds, adjusting the moisture content or increasing the buffering capacity of the system (Chiu and Lo, 2016).

Besides biological processes, thermochemical conversion represents a feasible option for FW handling. Thermochemical processes such as incineration, gasification, liquefaction, pyrolysis, torrefaction, and hydrothermal carbonization (HTC) could be more beneficial to valorize FW than AD. Nevertheless, most of these processes usually require feedstock pre-treatment (Okolie et al., 2022), and the need for a gas cleaning unit to prevent emissions such as NO_x, SO_x, particulate matter and heavy metals (Saqib et al., 2019). Among these technologies, HTC emerges as a cost-effective technology for thermochemical processing of high-moisture biomass, including FW, to obtain a product with attractive properties as a biofuel (Sarrion et al., 2021).

HTC is a thermochemical process for the treatment of high-moisture content biomass at a wide range of temperatures (170–250 °C), autogenous pressures (2–6 MPa), and time (from a few minutes to several hours). Through hydrolysis, condensation, aromatization, dehydration and decarboxylation reactions, a slurry product containing a solid and a liquid fraction is generated. The solid product, named hydrochar, contains around 40–90 % of the initial carbon of the feedstock, and involves energy values in the range 15–30 MJ/kg (Ipiales et al., 2021). The liquid product or process water (PW) is rich in mineral salts, nutrients and hydrolyzable organic compounds (Aragón-Briceño et al., 2021). This makes PW a potential substrate for AD, which has been widely investigated for different types of biomass waste (e.g. animal manure, FW, garden and park waste, microalgae, and sewage sludge) (Ipiales et al., 2022; Mannarino et al., 2022; Marin-Batista et al., 2020, 2019; Villamil et al., 2020). Being a by-product with interesting valorization potential, particular attention has been paid to phosphorus recovery from PW, which can be precipitated, together with NH_4-N and Mg, into struvite for use as fertilizer (Becker et al., 2019; Zhang et al., 2020). Furthermore, the use of acid reagents during HTC has been proven to promote the nutrient release from different feedstocks, such as sewage sludge, animal manure, and FW, which leads to increased nutrient concentration in the PW (Dai et al., 2017; Ekpo et al., 2016; Qaramaleki et al., 2020; Sarrion et al., 2022, 2021). Acid-assisted HTC (HCl, H_2SO_4) of animal manure allowed the release of more than 95 % of P and 60 % of N at a lower acid concentration than using organic acids (citric acid, acetic acid) at temperatures below 200 °C (Dai et al., 2017; Ekpo et al., 2016; Qaramaleki et al., 2020). Particularly, Sarrion et al. (2021) reported that the use of HCl during HTC is beneficial for nutrient recovery from FW, achieving almost 100 % and up to 98 % N and P solubilization in the PW, respectively, after 60 min HTC mediated by 0.5 M HCl at 170 °C.

Recent studies have addressed the integration of HTC, nutrient recovery, and AD to provide an energy-efficient valorization process for biomass waste with high moisture content. However, there is a lack of knowledge about the environmental feasibility of this integrated process compared to the more widespread and studied AD technology, which could lead to this novel technology being considered a solid route for FW valorization.

Within this context, the purpose of this work is to conduct a system-level analysis to benchmark the environmental impacts of a combined HTC, nutrient recovery, and AD process for FW valorization against the conventional stand-alone AD configuration. Process simulation models were developed to collect inventory data from mass and energy balances, followed by a comparative life cycle assessment (LCA).

2. Material and methods

2.1. Description of the case studies

Two technological scenarios for FW management were modeled and evaluated, as illustrated in Fig. 1. The FW feedstock was collected from a local management plant operating on a food distribution platform (Madrid, Spain). The characterization of the raw FW and the main process specifications implemented in process simulation are summarized in Table 1. Below is a description of both scenarios, which have been developed as energy self-sufficient processes:

- Stand-alone anaerobic digestion scenario (Sc_AD): reference system consisting of feeding FW into a low organic loading rate mesophilic anaerobic digester to produce biogas, which is cleaned in a water scrubbing unit to remove NH_3 and H_2S prior to combustion to generate steam and electricity.
- Hydrothermal carbonization, nutrient recovery and anaerobic digestion combined scenario (Sc_HTC + NR + AD): novel system consisting of treating FW by an acid-mediated HTC to produce hydrochar while promoting solubilization of nutrients (mainly P and N) from the feedstock to the resulting PW. The PW rich in nutrients is then derived to a nutrient recovery stage consisting of a stirred reactor also fed with $MgCl_2$ and NaOH solution. Subsequently, nutrient-depleted secondary PW (PW-S) is subjected to AD to produce biogas, which is cleaned and combusted with air. Exhaust gases from combustion are used to dry the hydrochar up to the required final moisture content. Additionally, part of the hydrochar is combusted to meet the energy needs of the process.

In Sc_AD, the FW is fed into a mesophilic AD stage (1 bar, 35 °C) (Mannarino et al., 2022), where biogas is produced. Since the feedstock involves a moisture content of 92.7 %, there is no need for an extra addition of water for pumping and AD stages (Lucian et al., 2020). A digestate stream containing non-digested material (5 % w/w) together with water and a small amount of dissolved gases leaves the stage and is destined for direct use on the soil. The utilization of biogas requires previous conditioning to avoid corrosion and emission issues in a combined heat and power unit (Golmakani et al., 2022). Hence, biogas is cleaned in a water scrubber unit to eliminate NH_3 and H_2S (both 99 % retention). Biogas scrubbing

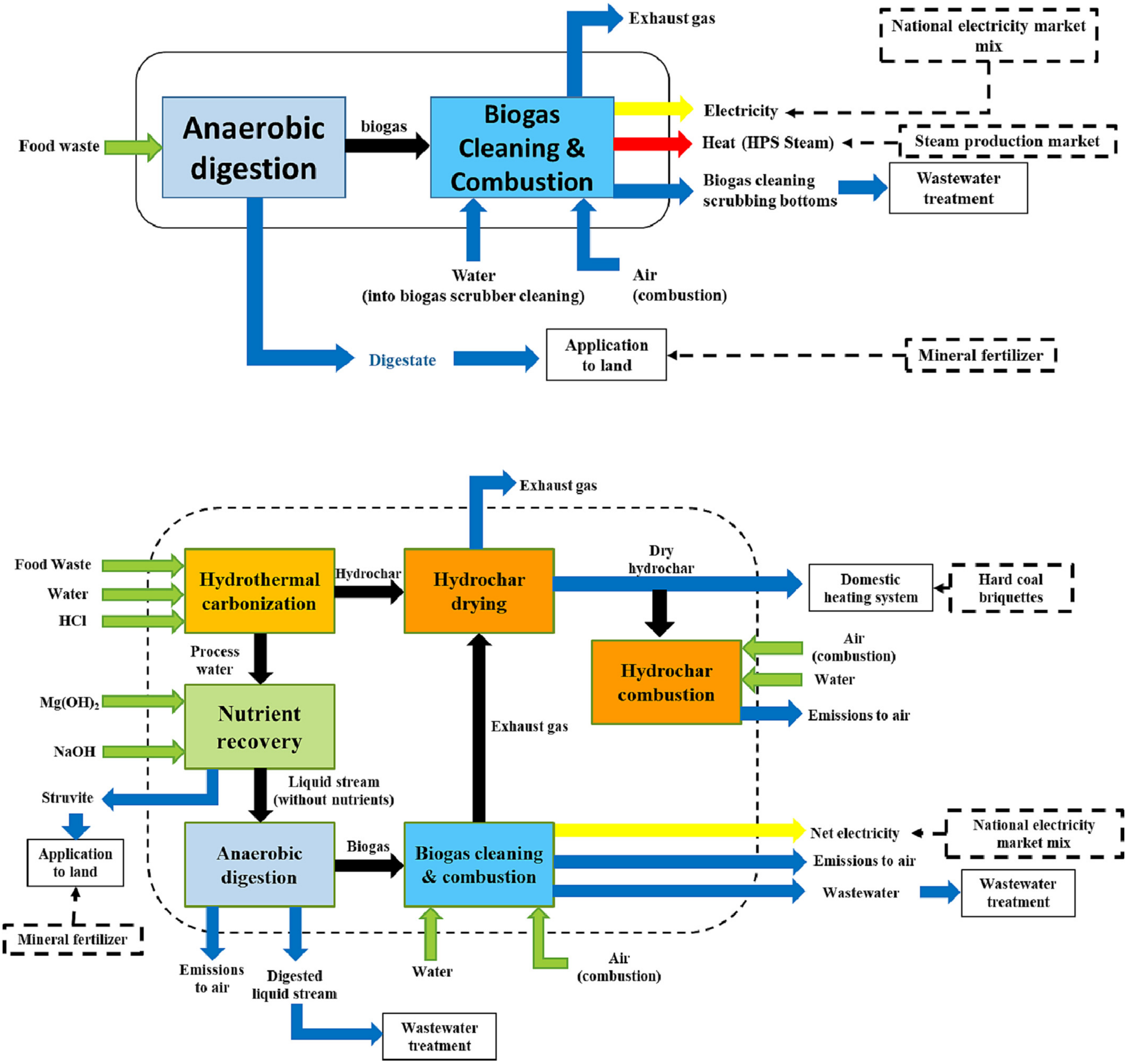

Fig. 1. Block diagram of the two technology scenarios for food waste valorization. Dashed lines represent avoided products. Top: reference system based on anaerobic digestion (Sc_AD); Bottom: alternative system based on hydrothermal carbonization, nutrient recovery and anaerobic digestion (Sc_HTC + NR + AD).

bottoms containing cleaning water plus pollutants are derived to conventional wastewater treatment. The clean biogas (65 % CH_4, 34 % CO_2 v/v) is compressed and combusted with air (10 % volumetric excess) in a gas turbine, which operates at 10 bar and 1200 °C, to generate electricity. Additionally, heat from the exhaust gas leaving the turbine (1.5 bar and 852 °C) is used to produce high-pressure steam (HPS; 40 bar and 340 °C). Finally, the exhaust gas stream is released to the atmosphere at 56 °C.

In Sc_HTC + NR + AD, the FW is fed into the HTC reactor, which operates at 20 bar and 170 °C for a retention time of 1 h (Sarrion et al., 2021). According to previous work, the FW feedstock with 92.7 % moisture is suitable to carry out the HTC reaction without the need for additional water (Mannarino et al., 2022; Sarrion et al., 2021). In order to improve the nutrient release to the PW, 0.5 M HCl is added. These operating conditions were selected according to the results reported by Sarrion et al. (2021) on nutrient recovery from FW by acid-mediated HTC process. They reported maximum P and N solubilization in the PW (up to 98 % P, as PO_4-P, and almost 100 % N, mainly as organic N and NH_4-N (16 %)) achieved after 60 min HTC at 170 °C in the presence of 0.5 M HCl. The resulting slurry (mixture of hydrochar and PW) from HTC is subsequently depressurized in two flash tanks (6.24 bar and 1.98 bar). Both head (process water) and bottom (wet hydrochar) streams from depressurization are used for energy integration in the process, helping to preheat the FW inlet to the HTC reactor up to 120 °C. After that, the solid hydrochar is separated from the PW by filtration in a filter press. PW flows to a stirred reactor to chemically precipitate P and NH_4-N by addition of a magnesium agent ($MgCl_2$) to promote struvite formation according to a molar ratio of NH_4:Mg:PO_4 of 1:1.3:1 (Pastor et al., 2008). The mixture pH is increased with 2 M NaOH up to pH 9, and maintained under stirring for 20 min. The precipitated solid is then separated by filtration and PW-S is derived to an AD stage similar to that in SC_AD. The produced biogas is cleaned and combusted, while the digested liquid stream is sent to wastewater treatment. In this case, the hot exhaust gas from biogas combustion is used to

Table 1
Main data implemented in process simulation for the AD stand-alone (food waste composition together with anaerobic digestion plus biogas cleaning and combustion conditions) and HTC + NR + AD scenarios.

Item	Unit	Value		Comments
Solids composition		Food waste	Hydrochar	Hydrochar before drying
Moisture	% w/w	92.7	30.0	
Proximate analysis	% w/w			
Fixed carbon		20.6	40.2	Dry basis
Volatile matter		67.6	53.4	Dry basis
Ash		11.8	6.4	Dry basis
Ultimate analysis	% w/w			
Carbon		44.5	56.1	Dry basis, ash free
Hydrogen		6.1	6.0	Dry basis, ash free
Oxygen		34.3	30.9	Dry basis, ash free
Nitrogen		3.1	0.3	Dry basis, ash free
Sulfur		0.2	0.3	Dry basis, ash free
Higher heating value	MJ/kg	18.9	23.6	Dry basis
Hydrothermal carbonization				
Temperature	˚C	170		
Pressure	bar	20.0		
Residence time	h	1.00		
Hydrochar yield	% w/w	49.5		Referred to dry input
Anaerobic digestion				
Temperature	˚C	35.0		
Pressure	bar	1.00		
Biogas cleaning and combustion				
Scrubber pressure	bar	5.00		Top stage
Scrubber pressure drop	bar	0.10		
Scrubber NH_3 retention efficiency	%	98.0		
Gas turbine inlet pressure	bar	10.0		
Gas turbine discharge pressure	bar	1.50		
Exhaust gas temperature	˚C	852		Out of gas turbine
Hydrochar drying				
Dry hydrochar moisture	% w/w	8.00		
Exhaust gas temperature	˚C	538		After steam generation
Exhaust gas emission temperature	˚C	80.0		
Hydrochar combustion (at plant)				It meets the thermal energy demand gap which is not satisfied by biogas combustion
Temperature	˚C	1200		
Pressure	bar	1.30		
Exhaust gas emission temperature	˚C	80.0		After steam generation

dry the wet hydrochar (30 % moisture) to a final 8 % moisture content. Finally, the exhaust gas stream is released to the atmosphere at 80 ˚C.

2.2. Process modeling and simulation

Life-cycle inventory (LCI) data for the analyzed scenarios were mainly retrieved from process simulation models implemented in Aspen Plus® V12, treating 150 kt y^{-1} of FW, which is considered a medium-high scale according to some industrial processes related to HTC found in literature, such as those described by de Mena Pardo et al. (2016) to treat 35 kt y^{-1} of organic fraction of municipal waste or by Saba et al. (2019) for processing around 1500 kt y^{-1} of coal and miscanthus mixture. A complete description of such models can be found in Medina-Martos et al. (2020), which were adapted to suit specific aspects of the current work (Table 1). LCI data for nutrient recovery stage were directly derived from experimental work. Besides this, the most relevant difference refers to the inclusion of a module for on-site combustion of a fraction of the generated hydrochar, as a backup to cover the internal heat demand, which cannot be fully satisfied by biogas. The utilized thermodynamic property methods were: Peng-Robinson with Boston-Mathias correction (PR-BM) equation of state for the HTC section, steam table functions (STEAMBS) in the steam generation section, and NRTL for the rest of the sections. Both FW and hydrochar were introduced as nonconventional components, meaning they were defined based on their ultimate and proximate analyses, which Aspen Plus® utilizes to estimate enthalpy and density by means of empirical correlations. Other compounds were directly retrieved from the Aspen Plus® database. In particular, acetic acid was utilized as a proxy to simulate the thermodynamic behavior of the organic load in PW.

The HTC reactor was simulated as an RYield block, reproducing mass distribution from experimental data. A Buswell model was utilized to estimate biogas production and composition based on the inlet composition of PW. A combination of two stoichiometric reactor blocks (RStoic) coordinated by a calculator block was used to that end. The scrubber for biogas cleaning was simulated by means of a RadFrac column block with no reboiler and condenser. Finally, both the combustion of clean biogas and hydrochar were simulated in RGibbs reactor blocks, with 20 % stoichiometric air excess. The steam generation and hydrochar combustion sections were interlinked with the rest of the flowsheet to automatically calculate the required amount of steam and the fraction of combusted hydrochar to fulfill the internal heat demand. Following a conservative approach, thermal losses of 40 % for the HTC reactor and 20 % for the digester were considered.

2.3. Life cycle assessment framework

2.3.1. Goal and scope

The goal of this study is to benchmark the potential environmental impacts of the combined HTC, nutrient recovery, and AD system (Sc_HTC + NR + AD) against those of the stand-alone AD scenario (Sc_AD). This study considers the consumption and production of materials and energy, as well as waste and emissions generated over the life cycle of each FW treatment system. As also shown in Fig. 1, avoided production and use of conventional energy products (grid electricity, industrial heat, and hard coal) and mineral fertilizers because of energy and material recovery in both scenarios were also included. The functional unit of this study was defined as the treatment of 1 kg of wet FW, with the composition presented in Table 1. Therefore, both scenarios were defined as waste management systems whose main function is the treatment of (impact-free) food waste. An attributional modeling framework with averaged data was used.

2.3.2. Life cycle inventory

Inventory data associated with Sc_AD and Sc_HTC + NR + AD were based on process simulation (Section 2.2) and previous work by Medina-Martos et al. (2020). Background data were retrieved from the ecoinvent database. Specific details relevant to each system are provided below.

2.3.2.1. Sc_AD reference system. Table 2 presents the inventory data of the reference system. It was assumed that fugitive emissions in the AD stage account for 2.5 % of the biogas produced (Parravicini et al., 2022). Then, biogas stream is derived to the scrubber unit, where 2 % of the biogas was considered to be lost as fugitive emission (Kapoor et al., 2019). Information from Tong et al. (2018) was used to estimate NH_3 emissions.

The amount and type of material left over after anaerobic digestion was calculated by subtracting the amount of C, N, and S broken down during the process from the initial feedstock composition, assuming that metals remain within the digestate during AD (Chiu and Lo, 2016). It should be noted that the metal content in the Sc_AD (and Sc_HTC + NR + AD) digestates is below the Spanish limits for metals in materials to be used on agricultural land. Therefore, it was assumed that, after being transported 50 km, the digestate could be used on agricultural land.

The amount of C, N, and P emitted after digestate application was modeled by using emission factors. N emission factors were based on data for a sandy loam soil located in a low-precipitation area (2.6 % of the N applied is emitted as N_2O, 6.8 % is emitted as NH_3, 22.5 % is emitted to groundwater as nitrate, and 14 % is emitted to surface water bodies as nitrate) (Herrera et al., 2022). The C balance was assumed to be 0.05 % emitted as methane, 93.5 % released as biogenic CO_2, and the remaining part sequestered in the soil after 100 years (Yoshida et al., 2018). Herrera et al. (2022) found that 1.5 % of the P contained in the digestate was lost by surface water run-off. Metal elements were applied to the land.

Land application of digestate avoids the production and application of industrial mineral fertilizers of N, P and K. The amount of mineral fertilizer avoided is related to the nutrient content in the digestate (N, P and K) and its availability to plants compared to that achieved by mineral fertilizers (Bala et al., 2021). According to the results in Chiew et al. (2015), 50 % of the N contained in a digestate or in a mineral fertilizer can be absorbed by plants, while all P and K were assumed to be plant available. Regarding avoided environmental impacts associated with mineral fertilizer application to land, according to Yoshida et al. (2018), 2 % of the N contained in a fertilizer is emitted as N_2O, while 10 % and 4 % are released as nitrate into groundwater and surface water, respectively.

Table 2
Main inventory data of the reference system Sc_AD (values per kg of wet food waste managed via anaerobic digestion).

Item	Unit	Value	Comments
Inputs			
FW	kg	1.00	Wet; functional unit
Digestate transport	t·km	$4.45 \cdot 10^{-2}$	By truck
Digestate spreading	m^3	$8.56 \cdot 10^{-4}$	Application to land
Water	kg	$8.13 \cdot 10^{-2}$	For biogas cleaning and combustion
Outputs			
Heat	MJ	$1.21 \cdot 10^{-1}$	Avoided product (heat from steam in chemical industry)
Electricity	kWh	$1.35 \cdot 10^{-2}$	Avoided product (Spanish grid electricity according to production mix in 2020)
N mineral fertilizer	kg N	$7.30 \cdot 10^{-4}$	Avoided product
P mineral fertilizer	kg P_2O_5	$6.33 \cdot 10^{-4}$	Avoided product
K mineral fertilizer	kg K_2O	$3.32 \cdot 10^{-3}$	Avoided product
Biogas fugitive emissions	kg	See comment	From anaerobic digestion and biogas cleaning. CO_2 (biogenic): $1.30 \cdot 10^{-3}$; CH_4 (biogenic): $6.43 \cdot 10^{-4}$; NH_3: $1.28 \cdot 10^{-5}$; H_2S: $5.73 \cdot 10^{-6}$
Digestate use: emissions to air	kg	See comment	CO_2 (biogenic): $4.19 \cdot 10^{-2}$; CH_4 (biogenic): $8.10 \cdot 10^{-6}$; NH_3: $1.66 \cdot 10^{-4}$; N_2O: $9.41 \cdot 10^{-5}$; NO_x: $1.76 \cdot 10^{-5}$
Digestate use: emissions to water	kg	See comment	NO_3^- (river): $1.24 \cdot 10^{-3}$; P (river): $1.29 \cdot 10^{-5}$; NO_3^- (groundwater): $3.32 \cdot 10^{-3}$
Digestate use: emissions to soil	kg	See comment	CO_2: $2.67 \cdot 10^{-3}$; N: $3.13 \cdot 10^{-4}$; Na: $1.61 \cdot 10^{-4}$; Mg: $3.14 \cdot 10^{-4}$; Al: $7.15 \cdot 10^{-5}$; As: $2.91 \cdot 10^{-7}$; P: $8.57 \cdot 10^{-5}$; Ca: $8.54 \cdot 10^{-4}$; Cd: $7.30 \cdot 10^{-9}$; Ti: $2.27 \cdot 10^{-6}$; Fe: $2.20 \cdot 10^{-6}$; Cr: $1.31 \cdot 10^{-6}$; Ni: $5.59 \cdot 10^{-7}$; Pb: $2.58 \cdot 10^{-7}$; Si: $7.10 \cdot 10^{-5}$; Sb: $7.30 \cdot 10^{-11}$; Cu: $1.30 \cdot 10^{-6}$; Zn: $3.93 \cdot 10^{-6}$; Sr: $3.13 \cdot 10^{-6}$; Mn: $3.92 \cdot 10^{-6}$; Mo: $9.56 \cdot 10^{-8}$; Li: $1.96 \cdot 10^{-7}$; Co: $7.15 \cdot 10^{-8}$
Biogas combustion emissions to air	kg	See comment	CO_2 (biogenic): $7.13 \cdot 10^{-2}$; CH_4 (biogenic): $4.43 \cdot 10^{-4}$; CO (biogenic): $3.16 \cdot 10^{-4}$; SO_2: $4.30 \cdot 10^{-5}$; NO_x: $9.73 \cdot 10^{-6}$; N_2O: $5.73 \cdot 10^{-7}$
Wastewater	m^3	$7.81 \cdot 10^{-5}$	Waste to treatment
Other avoided items			
Fertilizer transport	t·km	$4.45 \cdot 10^{-2}$	Associated with the avoided mineral fertilizers
Fertilizer spreading	ha	$2.38 \cdot 10^{-4}$	Associated with the avoided mineral fertilizers
Fertilizer use: emissions to air	kg	See comment	Associated with the avoided mineral fertilizers. N_2O: $2.29 \cdot 10^{-5}$; NO_x: $4.29 \cdot 10^{-6}$
Fertilizer use: emissions to water	kg	See comment	Associated with the avoided mineral fertilizers. NO_3^- (river): $1.29 \cdot 10^{-4}$; P (river): $9.74 \cdot 10^{-6}$; NO_3^- (groundwater): $3.23 \cdot 10^{-4}$
Fertilizer use: emissions to soil	kg	See comment	Associated with the avoided mineral fertilizers. N: $1.23 \cdot 10^{-4}$; As: $3.25 \cdot 10^{-8}$; Cd: $1.04 \cdot 10^{-8}$; Cr: $1.11 \cdot 10^{-7}$; Cu: $4.76 \cdot 10^{-8}$; Hg: $2.65 \cdot 10^{-10}$; Mo: $1.49 \cdot 10^{-8}$; Ni: $4.80 \cdot 10^{-8}$; Pb: $3.07 \cdot 10^{-8}$; Se: $2.34 \cdot 10^{-8}$; Zn: $2.62 \cdot 10^{-7}$

2.3.2.2. Sc_HTC + NR + AD system. Table 3 presents the inventory data of the novel system for FW management. The composition of the digested PW was obtained by a mass balance between the chemical elements of the feedstock and PW and the mass of N and P precipitated with the struvite. Approximately 100 % of the N and 98 % of the P were transferred to the PW, of which 5 % and 100 % of N and P precipitated as struvite. In this case, 21 % of the N and 71 % of the P contained in the feedstock were considered to be taken by plants upon struvite application to land.

Table 3
Main inventory data of the novel system Sc_HTC + NR + AD (values per kg of wet food waste managed via hydrothermal carbonization followed by nutrient recovery and anaerobic digestion).

Item	Unit	Value	Comments
Inputs			
FW	kg	1.00	Wet; functional unit
Water	kg	$5.10 \cdot 10^{-1}$	For hydrothermal carbonization, biogas cleaning and combustion, and hydrochar combustion at plant
HCl	kg	$4.59 \cdot 10^{-2}$	For hydrothermal carbonization
NaOH	kg	$3.00 \cdot 10^{-3}$	For nutrient recovery
$Mg(OH)_2$	kg	$4.02 \cdot 10^{-4}$	For nutrient recovery
Hydrochar transport	t·km	$8.88 \cdot 10^{-4}$	Net hydrochar (surplus) transported by truck
Struvite transport	t·km	$3.30 \cdot 10^{-3}$	By truck
Struvite spreading	m^3	$3.18 \cdot 10^{-6}$	Application to land
Outputs			
Heat	MJ	$7.11 \cdot 10^{-1}$	Avoided product (heat from hard coal briquettes)
Electricity	MJ	$2.11 \cdot 10^{-2}$	Avoided product (Spanish grid electricity according to production mix in 2020)
N mineral fertilizer	kg N	$1.26 \cdot 10^{-4}$	Avoided product
P mineral fertilizer	kg P_2O_5	$4.02 \cdot 10^{-4}$	Avoided product
Biogas fugitive emissions	kg	See comment	From anaerobic digestion and biogas cleaning. CO_2 (biogenic): $8.90 \cdot 10^{-4}$; CH_4 (biogenic): $3.07 \cdot 10^{-4}$; NH_3: $2.48 \cdot 10^{-6}$; H_2S: $5.63 \cdot 10^{-7}$
Struvite use: emissions to soil	kg	See comment	Na: $3.53 \cdot 10^{-5}$; Cl: $5.45 \cdot 10^{-5}$
Biogas combustion emissions to air	kg	See comment	Released after hydrochar drying. CO_2 (biogenic): $4.15 \cdot 10^{-2}$; CH_4 (biogenic): $2.95 \cdot 10^{-4}$; CO (biogenic): $2.10 \cdot 10^{-4}$; SO_x: $2.22 \cdot 10^{-5}$; NO_x: $4.98 \cdot 10^{-6}$; N_2O: $1.31 \cdot 10^{-9}$; H_2O: $2.90 \cdot 10^{-2}$
Hydrochar combustion emissions to air (plant)	kg	See comment	CO_2 (biogenic): $1.97 \cdot 10^{-3}$; N: $6.29 \cdot 10^{-3}$; O_2: $3.16 \cdot 10^{-4}$; CO (biogenic): $1.99 \cdot 10^{-8}$; SO_x: $5.75 \cdot 10^{-6}$; NO_x: $4.04 \cdot 10^{-6}$; H_2O: $5.96 \cdot 10^{-4}$
Hydrochar combustion emissions to air (households)	kg	See comment	CO_2 (biogenic): $5.61 \cdot 10^{-2}$; CH_4 (biogenic): $8.81 \cdot 10^{-8}$; NH_3: $1.79 \cdot 10^{-8}$; N_2O: $1.88 \cdot 10^{-6}$; CO (biogenic): $1.63 \cdot 10^{-6}$; SO_2: $3.17 \cdot 10^{-7}$; NO_x: $1.29 \cdot 10^{-4}$; Al: $3.15 \cdot 10^{-11}$; P: $6.04 \cdot 10^{-8}$; Ca: $1.51 \cdot 10^{-6}$; Fe: $1.79 \cdot 10^{-13}$; Cr: $9.04 \cdot 10^{-10}$; Ni: $5.57 \cdot 10^{-10}$; Mg: $4.49 \cdot 10^{-8}$; Mn: $5.93 \cdot 10^{-10}$; particulates: $5.16 \cdot 10^{-6}$ (PM10–2.5) and $3.14 \cdot 10^{-5}$ (PM2.5)
Wastewater	m^3	$3.83 \cdot 10^{-2}$	Waste to treatment
Ash from hydrochar combustion at plant	kg	$2.50 \cdot 10^{-4}$	Waste to treatment. Inputs required: landfill infrastructure ($5.26 \cdot 10^{-13}$ p) and process-specific burdens ($2.50 \cdot 10^{-4}$ kg). Emissions to river (kg): Al ($5.51 \cdot 10^{-9}$), As ($2.75 \cdot 10^{-10}$), B ($1.55 \cdot 10^{-11}$), Cd ($1.95 \cdot 10^{-13}$), Ca ($1.55 \cdot 10^{-11}$), Cr VI ($1.00 \cdot 10^{-9}$), Co ($9.01 \cdot 10^{-11}$), Cu ($1.38 \cdot 10^{-12}$), Fe ($1.10 \cdot 10^{-10}$), Pb ($5.26 \cdot 10^{-13}$), Mg ($2.13 \cdot 10^{-11}$), Mn ($1.58 \cdot 10^{-12}$), Mo ($3.50 \cdot 10^{-9}$), Ni ($1.78 \cdot 10^{-11}$), K ($1.08 \cdot 10^{-6}$), Se ($3.25 \cdot 10^{-9}$), Si ($6.26 \cdot 10^{-9}$), Na ($7.26 \cdot 10^{-7}$), Ti ($4.51 \cdot 10^{-9}$), Zn ($1.70 \cdot 10^{-12}$). Emissions to groundwater (kg): Al ($3.25 \cdot 10^{-6}$), As ($2.75 \cdot 10^{-19}$), B ($1.95 \cdot 10^{-9}$), Cd ($1.18 \cdot 10^{-10}$), Ca ($9.26 \cdot 10^{-9}$), Cr VI ($3.25 \cdot 10^{-9}$), Co ($5.26 \cdot 10^{-8}$), Cu ($8.26 \cdot 10^{-10}$), Fe ($6.76 \cdot 10^{-8}$), Pb ($3.25 \cdot 10^{-10}$), Mg ($1.28 \cdot 10^{-8}$), Mn ($9.51 \cdot 10^{-12}$), Mo ($1.63 \cdot 10^{-11}$), Ni ($1.05 \cdot 10^{-8}$), K ($2.75 \cdot 10^{-6}$), Se ($6.01 \cdot 10^{-9}$), Si ($2.00 \cdot 10^{-7}$), Na ($1.20 \cdot 10^{-6}$), Ti ($2.76 \cdot 10^{-6}$), Zn ($1.08 \cdot 10^{-9}$)
Ash from hydrochar combustion at households	kg	$6.14 \cdot 10^{-3}$	Conventional MSW landfilling
Other avoided items			
Fertilizer transport	t·km	$1.65 \cdot 10^{-4}$	Associated with the avoided mineral fertilizers
Fertilizer spreading	ha	$3.75 \cdot 10^{-5}$	Associated with the avoided mineral fertilizers
Fertilizer use: emissions to air	kg	See comment	Associated with the avoided mineral fertilizers. N_2O: $1.95 \cdot 10^{-6}$; NO_x: $4.10 \cdot 10^{-7}$
Fertilizer use: emissions to water	kg	See comment	Associated with the avoided mineral fertilizers. NO_3^- (river): $1.10 \cdot 10^{-5}$; P (river): $5.19 \cdot 10^{-7}$; NO_3^- (groundwater): $2.75 \cdot 10^{-5}$
Fertilizer use: emissions to soil	kg	See comment	Associated with the avoided mineral fertilizers. N: $2.13 \cdot 10^{-5}$; As: $1.53 \cdot 10^{-8}$; Cd: $6.11 \cdot 10^{-9}$; Cr: $6.47 \cdot 10^{-8}$; Cu: $1.99 \cdot 10^{-8}$; Hg: $8.77 \cdot 10^{-11}$; Mo: $8.60 \cdot 10^{-9}$; Ni: $2.23 \cdot 10^{-8}$; Pb: $1.75 \cdot 10^{-8}$; Se: $5.97 \cdot 10^{-9}$; Zn: $1.40 \cdot 10^{-7}$

Table 4
Environmental life-cycle profile of the reference and novel systems for food waste management (values per kg of wet feedstock; green shaded cells denote a comparatively favorable performance under a specific indicator).

Indicator [a] / System	GWP (kg CO_2 eq)	AP (mol H^+ eq)	EP_f (kg P eq)	EP_m (kg N eq)	EP_t (mol N eq)	ADP_f (MJ)	ADP_m (kg Sb eq)
Reference (AD)	$1.76 \cdot 10^{-2}$	$-1.95 \cdot 10^{-4}$	$-3.01 \cdot 10^{-5}$	$9.03 \cdot 10^{-4}$	$-2.84 \cdot 10^{-7}$	$-5.96 \cdot 10^{-1}$	$-4.09 \cdot 10^{-7}$
Novel (HTC+NR+AD)	$-8.28 \cdot 10^{-2}$	$-5.42 \cdot 10^{-4}$	$-2.16 \cdot 10^{-5}$	$6.32 \cdot 10^{-5}$	$-3.74 \cdot 10^{-4}$	$-6.15 \cdot 10^{-1}$	$8.08 \cdot 10^{-7}$

[a] GWP: global warming impact potential; AP: acidification impact potential; EP_f/EP_m/EP_t: freshwater/marine/terrestrial eutrophication impact potential; ADP_f/ADP_m: abiotic depletion impact potential for fossil and nuclear resources/minerals and metals.

In order to achieve an energy self-sufficient process, 12.5 % of the dry hydrochar produced was combusted in an industrial heating stove with a heat conversion efficiency of 85 % on a higher heating value (HHV) basis (managing to produce 3.9 MJ per kg of FW), while the rest was transported 20 km and combusted in a domestic heating stove with a heat conversion efficiency of 70 %. Heat production was 23.3 MJ per kg of dry hydrochar. Owing to the lack of measured data associated with emissions from hydrochar combustion, the inventory was adapted from Owsianiak et al. (2016), who observed emissions (per kg of dry biochar) of 38 mg CO, 3 g NO_x, 0.12 g $PM_{10–2.5}$ and 73 mg $PM_{2.5}$ at pilot plant scale. For other waste-specific emissions, such as heavy metals, transfer coefficients for municipal solid waste incineration were retrieved from the ecoinvent database as previously used in the literature (Berge et al., 2015). It was also used information from Glover et al. (2022) to model the impacts associated with hydrochar ash disposal. Finally, the use of hydrochar for heat production was considered to avoid the production, combustion, and disposal of ash associated with hard coal briquettes. The substitution ratio in MJ is 1:1.

2.3.3. Life cycle impact assessment

Inventory data were implemented in specific LCA software (SimaPro 9) to subsequently evaluate the following environmental indicators using the Environmental Footprint (EF 3) method: global warming (GWP), acidification (AP), freshwater eutrophication (EP_f), marine eutrophication (EP_m), terrestrial eutrophication (EP_t), abiotic depletion for fossil resources (ADP_f), and abiotic depletion for mineral and metal resources (ADP_m).

3. Results

3.1. Environmental evaluation

Table 4 presents the life-cycle profile computed for each scenario. The combined system (Sc_HTC + NR + AD) shows a more favorable performance than the reference one (Sc_AD) under five out of seven environmental impact indicators, including the carbon (GWP) and non-renewable energy (ADP_f) footprints. Moreover, it involves a similar freshwater eutrophication footprint (EP_f), but a worse performance in terms of mineral and metal depletion (ADP_m).

Regarding Sc_HTC + NR + AD, the main effect on the carbon footprint indicator comes from the substitution of hydrochar for coal as an energy source. It was considered that hydrochar is of biogenic origin, thus leading to biogenic carbon emissions when combusted. In contrast, the combustion of coal pellets generates a large amount of greenhouse gas emissions. The application of the digestate obtained after AD is the main contributor to the global warming impact in Sc_AD. The main emission from the digestate is caused by the emission of N_2O, via microbial (nitrification-denitrification pathways) and nonmicrobial (photodegradation, thermal degradation, oxidation by reactive oxidation species, extracellular oxidative metabolism, and/or inorganic chemical reactions) processes (Wang et al., 2017); a compound that is avoided during the application of struvite in the novel scenario. The subprocesses shared in both scenarios have similar contributions, caused by the fugitive methane emissions in the AD and biogas cleaning stages and the direct emissions of methane during biogas combustion. As shown in Table 4, the net carbon footprint of Sc_HTC + NR + AD –unlike that of the reference scenario– is negative, meaning that the combined HTC-based process generates a desirable impact thanks to coal avoidance.

The reduced acidification impact in Sc_HTC + NR + AD compared to Sc_AD was also found to be driven by the beneficial impacts associated with coal substitution by hydrochar. It should also be noted that struvite application to land eludes the release of H_2S, NH_3 and N_2O to the atmosphere associated with the use of digestate in Sc_AD, and the recovered heat in Sc_HTC + NR + AD is crucial to not purchase heat from market.

Regarding eutrophication impact categories, the spreading of digestate and its associated transport arose as relevant impact sources. Terrestrial eutrophication was found to be mainly related to N_2O and NH_3 emissions to air, which is eluded in Sc_HTC + NR + AD because of the struvite production. Furthermore, this struvite also contributes to reduce terrestrial eutrophication by replacing commercial fertilizers. In Sc_AD, nitrate and P release from digestate to groundwater and surface water involve a negative impact on marine and freshwater eutrophication, respectively, which could also be skipped by the application of struvite in the novel scenario.

Finally, the combined scenario would allow reducing the non-renewable energy footprint (ADP_f) with respect to the reference one (Sc_AD, mainly associated with the transport of the large amount of liquid FW digestate generated after AD). However, the infrastructures required for the novel scenario are larger and more complex than for Sc_AD, in addition to the need to use reagents related to nutrient recovery. The latter is reflected in an unfavorable mineral and metal depletion impact for Sc_HTC + NR + AD.

As regards the comprehensive analysis of each scenario separately, Fig. 2 presents a breakdown of the contribution of the different subsections involved in each scenario to identify the subsections that contribute most to the selected environmental impact indicators. In Sc_AD (Fig. 2a), the transport and use of digestate arose as the main source of damaging impact in all of the indicators. Specifically, application of digestate to land unfavorably affects GWP because of the emission of a large amount of N_2O after the nitrification-denitrification processes. Biogas fugitive emissions from the AD stage plus the biogas scrubbing and those emissions from biogas combustion also have a relevant contribution to the carbon footprint. The emission of NH_3 resulting from digestate application mainly contributes to AP, EP_f and EP_t, while nitrate leaching negatively affects EP_m. In relation to ADP_f and ADP_m, the transport and use of the digestate represents the only significant unfavorable impact because of the necessities of transport (50 km by truck), pumping and spreading.

These unfavorable impacts are sufficient to make Sc_AD environmentally harmful in terms of GWP and EP_m, as presented in Table 4. However, for the remaining impact categories, the avoidance of artificial fertilizers has a much more relevant desirable effect. In this sense, the substitution of fertilizers plays a leading role in the achievement of net favorable AP, EP_t, ADP_f and ADP_m results. Additionally, the beneficial impact associated with avoiding conventional heat and electricity also plays a key role, especially in terms of EP_f and ADP_f.

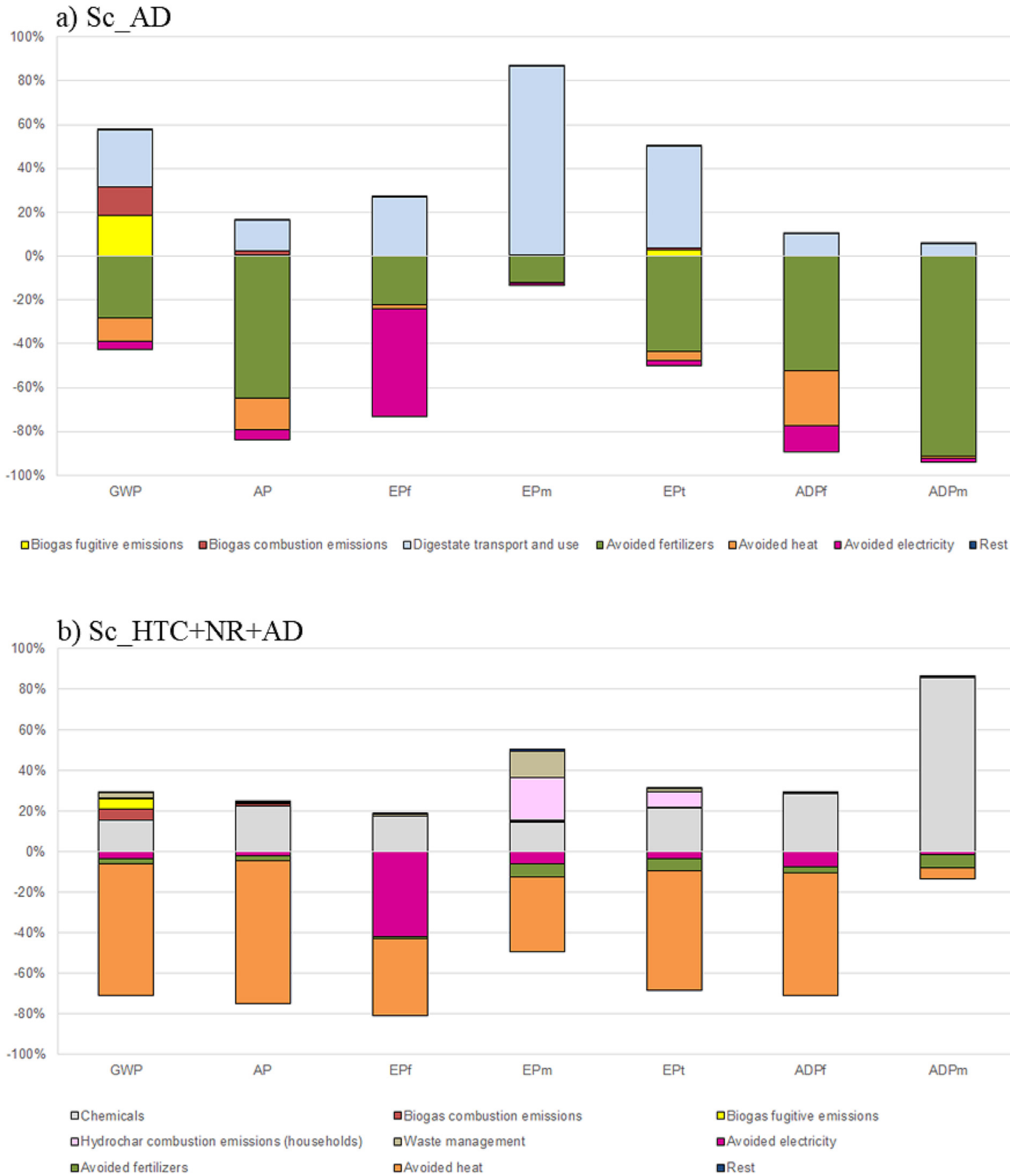

Fig. 2. Process contribution to life-cycle impacts for a) Sc_AD and b) Sc_HTC + NR + AD. Values above zero represent environmental impacts produced. Values below zero represent environmental impacts avoided.

In the novel scenario (Fig. 2b), the use of chemicals was found to be a general environmental hotspot, which is mainly associated with the use of HCl for the acid-mediated HTC process and both $MgCl_2$ and NaOH for the nutrient recovery stage. In particular, it was responsible for the unfavorable ADP_m performance of the system. In a future bioeconomy context, the potential use of chemical reagents obtained as by-products from biorefineries could reduce the environmental impacts linked to chemicals (Karka et al., 2015). Regarding EP_m and EP_t, the emissions from hydrochar combustion in households would also account for a relevant contribution to these impact categories.

The main favorable impact in Sc_HTC + NR + AD is associated with the avoidance of production and combustion of hard coal briquettes, which is promoted by the role of hydrochar as feedstock in stoves for heat production. This has a beneficial impact on all the selected indicators (except for ADP_m), being enough to achieve a negative net impact for these categories (except for EP_m). Further beneficial impact was achieved because of the avoided grid electricity and the substitution of commercial fertilizers by the recovered struvite.

3.2. Additional remarks

Overall, the combination of HTC, nutrient recovery and anaerobic digestion was found to involve a favorable environmental performance as an FW management system, also in comparison with the conventional anaerobic digestion system (Section 3.1). Besides potential environmental benefits, combining these technologies could also be cost-effective. In this sense, regarding the HCl-mediated HTC process, the continuous mode of the HTC reactor, the energy integration of the streams for heat recovery and the use of both biogas and hydrochar as biofuels minimize energy requirements and the associated costs. Moreover, a low HCl concentration is required during the HTC process to significantly solubilize nutrients in PW, offering the possibility of recovering them as value-added struvite (Becker et al., 2019). Then, considering negligible energy and HCl requirements, the cost of the HTC process was estimated to be approximately 3 € per ton of FW (Mannarino et al., 2022), i.e. inexpensive compared to other options such as anaerobic fermentation plus sequential extraction of P, reported by Vardanyan et al. (2018), and that HCl plus H_2O_2-assisted hydrothermal carbonization with struvite crystallization stage, reported by Zhang et al. (2020).

Regarding struvite precipitation, the cost of the required reagents ($MgCl_2$ and NaOH) can be calculated according to previous studies (Becker et al., 2019; Munir et al., 2017; Zhang et al., 2020), resulting in approximately 1.3 € per ton of FW. This value is similar to that in other studies on struvite precipitation (Huang et al., 2015), even lower (Zhang et al., 2020). In addition to the economic feasibility of the struvite precipitation step, this product also brings other advantages. Obtaining struvite through the combined system allows nutrient recovery in an easily reusable form, which can be used directly as a fertilizer in accordance with current regulations and provides economic benefits from the sale of the product (Bouzas et al., 2019). Struvite has a good nutritional content of P_2O_5, N-NH_4^+ and MgO and a low heavy metal content for use as a fertilizer. Furthermore, its low solubility makes it a slow-releasing fertilizer, being possible to use it in single doses without risk of damaging plant growth, and prevents problems associated with filtration into groundwater (Yesigat et al., 2022). Ahmed et al. (2018) compared the efficiency of struvite and di-calcium phosphate as phosphate fertilizers, concluding that struvite is comparable to di-calcium phosphate as a phosphate source but has the additional advantage of containing available nitrogen.

Finally, both the LCA and the economic outlook reported in this work could be considered for processes of similar dimension to the studied case, since the results could be affected by the scale-up of the system (Cornejo et al., 2016). According to Finzi et al. (2020), scale-up of a livestock waste treatment system is essential to achieve a favorable environmental, economic and energy balance, reporting that implementing an industrial-scale facility or a regional-scale collective for energy and nutrient valorization of livestock waste reduced both emissions and input costs (energy and infrastructure requirements) compared to single-farm facilities.

4. Conclusions

In this work, a novel combined technology (HTC of FW, nutrient recovery from the PW and subsequent AD of PW-S) for FW management was comprehensively evaluated and benchmarked against a reference AD process, both configurated to be energetically self-sufficient, by using the LCA methodology. The combined process represents an opportunity to improve the energy recovery from FW because of the production of hydrochar as a biofuel. Additionally, the HTC reaction was mediated by acid (HCl) to promote nutrient solubilization in the PW, subsequently recovered as struvite by chemical precipitation. This enhanced energy and resource recovery within the novel FW management system generally translates into favorable environmental impacts, as well as into a better environmental life-cycle environmental than the reference AD system.

CRediT authorship contribution statement

Andres Sarrion: Conceptualization, Methodology, Formal analysis, Investigation, Formal analysis, Writing – original draft. **Enrique Medina-Martos:** Investigation, Writing – review & editing. **Diego Iribarren:** Methodology, Formal analysis, Writing – review & editing, Supervision. **Elena Diaz:** Conceptualization, Methodology, Writing – review & editing, Supervision. **Angel F. Mohedano:** Conceptualization, Funding acquisition, Methodology, Resources, Writing – review & editing, Supervision, Project administration. **Javier Dufour:** Conceptualization, Funding acquisition, Resources, Writing – review & editing, Supervision, Project administration.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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