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Life Cycle Assessment of an Integrated Biorefinery for Polyhydroxyalkanoates and PHA-Coated Fertilizer Production in Sewage Sludge and Organic Waste Treatment Plants

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Abstract

This study assesses the prospective integration of mixed microbial culture-based polyhydroxyalkanoate (PHA) recovery and fertilizer coating into sewage sludge (SS) and organic fraction of municipal solid waste (OFMSW) treatment. One baseline and one integration configuration are compared for each feedstock per ton of incoming wet waste. The foreground inventory is reconstructed through a shared wet mass, solids, COD and energy model. Incoming raw OFMSW is distinguished from the diluted pretreated stream; fixed solids are conserved, separated polymer is calculated based on biomass capture, and coating is defined as 2% of final product mass. Missing operating information is represented by explicit prospective assumptions rather than new plant measurements. In the reference configuration, modeled GWP is -40.29 and 228.45 kg CO_2 eq/t wet SS for baseline and integration, and -81.13 and 363.59 kg CO_2 eq/t wet OFMSW. Electricity exports account for baseline net benefits; the integration configurations incur additional cultivation, separation, drying and coating burdens. Thirty coupled cases give integration-minus-baseline GWP ranges of 200.32 to 392.48 kg CO_2 eq/t SS and 369.84 to 594.47 kg CO_2 eq/t OFMSW. These findings concern the stated model and inherited characterization vectors. Unmeasured process coefficients, residual treatment proxies, incomplete solvent emission characterization and unavailable background dataset identifiers limit their generality. No intrinsic feedstock preference or agronomic equivalence of the candidate coated fertilizer is established.

Keywords: life cycle assessment; polyhydroxyalkanoates; sewage sludge; organic fraction of municipal solid waste; coated fertilizer

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1. Introduction

Polyhydroxyalkanoates (PHAs) are fully biodegradable polymers synthesized and accumulated intracellularly as polymeric granules upon cultivating different types of microorganisms in nutrient-limiting conditions [1]. Depending on the types of constituent monomeric units and the length of their polymer chains, PHAs demonstrate appealing properties, including similar tensile strength and crystallinity to petroleum-based plastic. They

break down more rapidly in the environment without leaving toxic residues, offering versatile applications [2]. Accordingly, PHAs represent a promising alternative to conventional petroleum-based plastics, contributing to the transition toward more sustainable materials for the circular economy. However, pure-culture PHA production is associated with high production costs due to costly feedstocks, low conversion efficiency, low productivity, massive freshwater consumption, and complex downstream processes [2,3]. These challenges result in a high price of 4–6 €/kg-PHAs, compared to 1 €/kg threshold of petroleum-based plastics, thereby limiting attraction for further mass production of PHAs [1,3]. The lack of cost competitiveness has driven researchers to develop PHA value chains based on waste-derived carbon sources, enabling potentially simultaneous compliance with regulatory discharge limits while obtaining value-added products as a low-cost bioplastic [4].

The sectors of sewage sludge (SS) and organic fraction of municipal solid waste (OFMSW) treatment are undergoing a shift in perception and are increasingly recognized as valuable resource streams within the circular economy model [4]. The integration of well-established mixed microbial culture (MMC)-derived PHA production technology into existing waste treatment plants is gaining increasing attention [5]. The most commonly applied scheme for this advanced technology typically includes a sequence of elements consisting of three independent phases. The first phase involves anaerobic fermentation of organic waste into a stream rich in volatile fatty acids (VFAs). In the second phase, the produced VFA-rich stream serves as readily biodegradable COD for the PHA-storing MMC selection and enrichment stage, typically conducted under a feast/famine regime and dual substrate limitation in a sequential batch reactor (SBR) [5]. The third phase utilizes a fed-batch reactor for the PHA accumulation stage, where the biomass is fed with the VFA-rich stream under optimized conditions to maximize PHA yield [1,6].

Over the past few years, the implementation of PHA production within waste treatment systems has highlighted the potential of utilizing various types of biowaste as raw materials for PHA production. This includes scale-up of the process from the laboratory to the pilot scale, using substrates such as SS [7–9], OFMSW [10–12], food and feed residues [13], dairy industrial waste [14,15], and combined waste treatment systems [16,17].

Anaerobic digestion stabilizes sludge and enables energy recovery as biogas, while producing digestate requires further management [17–19]. Digestate may provide nutrients and organic matter, but its suitability for agricultural use depends on feedstock quality, treatment and applicable requirements. Reference to Regulation (EU) 2019/1009 [20] does not establish that sewage-sludge-derived digestate or compost is eligible for a particular EU fertilizing product category. Regulatory eligibility and agronomic suitability of the modeled product require separate verification.

Fertilizers play a key role in agricultural production by improving land resource utilization and increasing crop yields. However, conventional fertilizers have low utilization efficiency, typically less than 50%, leading to wasted resources and environmental issues, including soil acidification and water eutrophication [21]. In addition, concerns related to nutrient losses, greenhouse gas emissions, and potential contamination from impurities further challenge the sustainability of fertilizer use. To address these challenges, slow-release fertilizer (SRF) has emerged as a promising solution to enhance nutrient utilization efficiency [22]. Under the Farm to Fork Strategy, the EU aims to reduce chemical fertilizer use by 20% by 2030 to promote sustainable agriculture [23]. A previous study reported that a PHA-based SRF composed of poly(3-hydroxybutyrate-co-3-hydroxyhexanoate), or PHBH, combined with commercial fertilizers enhanced plant growth and stimulated microbial activity [24]. Therefore, this study models a candidate PHA-coated fertilizer application without presuming demonstrated slow-release performance.

From an environmental perspective, Life Cycle Assessment (LCA) has emerged as a key tool for promoting the sustainable development of integrated waste and wastewater treatment processes [4]. As a quantitative environmental assessment method, LCA provides essential support for decision-making during strategic planning in the waste and wastewater treatment sectors. Specifically, it enables the evaluation of potential environmental burdens and resource consumption across the system, identifies environmental hotspots, facilitates systematic comparison of alternative technologies and future operational scenarios, and serves as a benchmark for emerging technologies [25,26].

To date, LCA studies of the integration of PHA production processes into SS and OFMSW treatment plants remain limited. A previous study reported that integrating PHA production into a municipal wastewater treatment plant (WWTP) improved environmental performance compared to the reference scenario [27]. Recently, research found that the integration of PHA production into WWTP enhanced overall environmental performance, except for the ozone depletion potential category, which was negatively affected due to intensive hypochlorite consumption [28]. Similarly, Bassi et al. conducted a comparative LCA of PHA production from urban biowaste against fossil-based plastics and first-generation biomass-derived PHA and concluded that biowaste-based PHA had lower impacts on climate change and fossil fuel depletion but exhibited higher eutrophication and abiotic resource depletion impacts, mainly due to chemical use in downstream processing [29]. The environmental burdens associated with the PHA recovery stage were further addressed by Rossi et al. [30], revealing that PHA extraction using ethyl acetate demonstrated the best environmental performance among the four most commonly used solvents in the downstream processes. The above studies highlight the benefits of integrating PHA production into waste and WWTP on a case-by-case basis, offering valuable insights into the environmental implications in comparison to conventional references.

This study investigates the prospective environmental implications of integrating PHA recovery and fertilizer coating into SS and OFMSW treatment plants. The modeled sequence comprises fermentation, microbial selection, PHA accumulation and extraction, followed by pelletization and coating of a compost-derived core. Each integration configuration is compared with its own waste treatment baseline. The novelty remains the integration of polymer recovery with a candidate-coated fertilizer application in two treatment contexts. The revised analysis propagates shared material and COD balances into energy, residual management and product allocation, and evaluates the resulting conditional environmental trade-offs. The coating is not assumed to provide demonstrated slow-release behavior or agronomic equivalence.

2. Materials and Methods

This assessment follows the goal, scope, inventory, impact assessment and interpretation framework of ISO 14040 and ISO 14044 [31,32]. It is a prospective process comparison based on literature coefficients and declared engineering assumptions. Conservation within the modeled foreground is checked computationally; this does not constitute validation against plant measurements or a complete elemental characterization. The foreground inventories are summarized in Tables 1–4; introduced assumptions and sensitivity cases are described in this section and Supplementary Tables S5 and S6.

Table 1. Revised SS-B foreground inventory. Annual values are model outputs; source inputs and introduced assumptions are described in Section 2 and Supplementary Tables S5 and S6. Water, wet mass and dry-solid quantities are not interchangeable.

Flow	Annual Quantity	Basis
Incoming wet waste	4,050,456.48	kg wet/y
Incoming TS	1,012,614.12	kg TS/y
Incoming VS	798,952.54	kg VS/y
Incoming FS	213,661.58	kg FS/y
Incoming COD	1,134,512.61	kg COD/y
Digester wet inlet	20,252,282.40	kg wet/y
Digester inlet VS	798,952.54	kg VS/y
Digester inlet particulate FS	213,661.58	kg FS/y
Digester inlet dissolved salts	0.00	kg FS/y
Digester inlet COD	1,134,512.61	kg COD/y
Biogas production	389,565.57	Nm3/y
Digestate wet outlet	19,761,841.52	kg wet/y
Digestate VS	351,539.12	kg VS/y
Digestate particulate FS	213,661.58	kg FS/y
Wet cake to composting	1,697,862.89	kg wet/y
Dewatering liquid to residual treatment	18,070,195.83	kg wet/y
FS in dewatering liquid	55,552.01	kg FS/y
COD in dewatering liquid	129,788.24	kg COD/y
Dry dewatering polymer	6217.21	kg/y
Fresh yard waste	203,743.55	kg wet/y
Final wet compost	806,398.51	kg wet/y
FS in final compost	168,296.75	kg FS/y

SS-PHA scenario.

Table 2. Revised SS-PHA inventory including explicit liquid returns and coating. All relevant material streams are explicitly accounted for in the inventory balances.

Flow	Annual Quantity	Basis
Incoming wet waste	4,050,456.48	kg wet/y
Incoming TS	1,012,614.12	kg TS/y
Incoming VS	798,952.54	kg VS/y
Incoming FS	213,661.58	kg FS/y
Incoming COD	1,134,512.61	kg COD/y
Fermentation wet inlet	23,549,165.58	kg wet/y
Fermentation gas mass	715.06	kg/y
VFA COD produced	167,780.03	kg COD/y
Culture dilution	40,845,458.70	kg water/y
Cultivation oxygen demand	46,843.12	kg O_2/y
Harvest wet inlet	51,390,768.65	kg wet/y
Captured organic solids	72,415.64	kg VS/y
Dry PHA recovered	4254.11	kg/y
Harvest liquid return to AD	51,018,889.10	kg wet/y
Extraction residue return to AD	840,156.30	kg wet/y
Digester wet inlet	64,854,439.77	kg wet/y
Digester inlet VS	675,294.06	kg VS/y
Digester inlet particulate FS	213,661.58	kg FS/y
Digester inlet dissolved salts	474.70	kg FS/y
Digester inlet COD	1,077,366.45	kg COD/y
Biogas production	370,948.62	Nm3/y
Digestate wet outlet	64,386,714.39	kg wet/y
Digestate VS	297,129.39	kg VS/y
Digestate particulate FS	213,661.58	kg FS/y

Table 2. *Cont.*

Flow	Annual Quantity	Basis
Wet cake to composting	1,534,471.16	kg wet/y
Dewatering liquid to residual treatment	62,857,867.15	kg wet/y
FS in dewatering liquid	56,018.16	kg FS/y
COD in dewatering liquid	123,250.72	kg COD/y
Dry dewatering polymer	5623.92	kg/y
Fresh yard waste	184,136.54	kg wet/y
Final wet compost	739,456.09	kg wet/y
FS in final compost	167,324.95	kg FS/y
Final candidate coated product	212,705.54	kg wet/y
Predried core allocated	208,451.43	kg/y
PHA used in coating	4254.11	kg/y
Physical surplus PHA	0.00	kg/y
Remaining wet compost	364,243.52	kg wet/y
Chloroform circulation and supply	80,828.10	kg/y
Captured solvent output	80,019.82	kg/y
Fugitive solvent air stream	808.28	kg/y

OFMSW-B scenario.

Table 3. Revised OFMSW-B inventory. Raw feed, rejects and the diluted accepted stream are explicitly distinguished. Original source concentration coefficients are retained on their correct wet-stream basis.

Flow	Annual Quantity	Basis
Incoming wet waste	145,000,000.00	kg wet/y
Incoming TS	57,565,000.00	kg TS/y
Incoming VS	51,328,791.67	kg VS/y
Incoming FS	6,236,208.33	kg FS/y
Incoming COD	49,333,205.00	kg COD/y
Wet rejects	11,020,000.00	kg wet/y
Pretreatment dilution water	309,270,500.00	kg water/y
Accepted diluted feed	443,250,500.00	kg wet/y
Accepted feed TS	53,190,060.00	kg TS/y
Digester wet inlet	886,501,000.00	kg wet/y
Digester inlet VS	47,427,803.50	kg VS/y
Digester inlet particulate FS	5,762,256.50	kg FS/y
Digester inlet dissolved salts	0.00	kg FS/y
Digester inlet COD	45,583,881.42	kg COD/y
Biogas production	22,914,972.65	Nm3/y
Digestate wet outlet	854,946,239.08	kg wet/y
Digestate VS	14,939,758.10	kg VS/y
Digestate particulate FS	5,762,256.50	kg FS/y
Wet cake to composting	62,188,851.87	kg wet/y
Dewatering liquid to residual treatment	792,985,109.38	kg wet/y
FS in dewatering liquid	1,498,186.69	kg FS/y
COD in dewatering liquid	3,733,319.89	kg COD/y
Dry dewatering polymer	227,722.16	kg/y
Fresh yard waste	7,462,662.22	kg wet/y
Final wet compost	28,467,554.86	kg wet/y
FS in final compost	4,637,202.92	kg FS/y

OFMSW-PHA scenario.

Table 4. Revised OFMSW-PHA foreground inventory. Recovered polymer, coated product, remaining compost and equivalent displaced products are distinct quantities. Reference substitution credits are zero.

Flow	Annual Quantity	Basis
Incoming wet waste	145,000,000.00	kg wet/y
Incoming TS	57,565,000.00	kg TS/y
Incoming VS	51,328,791.67	kg VS/y
Incoming FS	6,236,208.33	kg FS/y
Incoming COD	49,333,205.00	kg COD/y
Wet rejects	11,020,000.00	kg wet/y
Pretreatment dilution water	309,270,500.00	kg water/y
Accepted diluted feed	443,250,500.00	kg wet/y
Accepted feed TS	53,190,060.00	kg TS/y
Fermentation wet inlet	709,200,800.00	kg wet/y
Fermentation gas mass	12,967,149.64	kg/y
VFA COD produced	22,336,101.90	kg COD/y
Culture dilution	3,694,783,300.12	kg water/y
Cultivation oxygen demand	3,319,831.69	kg O_2/y
Harvest wet inlet	3,996,691,960.71	kg wet/y
Captured organic solids	5,493,001.89	kg VS/y
Dry PHA recovered	849,272.45	kg/y
Harvest liquid return to AD	3,972,502,636.87	kg wet/y
Extraction residue return to AD	53,632,767.41	kg wet/y
Digester wet inlet	4,420,030,591.99	kg wet/y
Digester inlet VS	34,849,011.64	kg VS/y
Digester inlet particulate FS	5,762,256.50	kg FS/y
Digester inlet dissolved salts	36,008.09	kg FS/y
Digester inlet COD	38,297,240.20	kg COD/y
Biogas production	16,120,408.47	Nm3/y
Digestate wet outlet	4,399,711,026.85	kg wet/y
Digestate VS	10,977,438.67	kg VS/y
Digestate particulate FS	5,762,256.50	kg FS/y
Wet cake to composting	50,288,868.08	kg wet/y
Dewatering liquid to residual treatment	4,349,606,691.51	kg wet/y
FS in dewatering liquid	1,533,884.92	kg FS/y
COD in dewatering liquid	3,136,543.97	kg COD/y
Dry dewatering polymer	184,532.74	kg/y
Fresh yard waste	6,034,664.17	kg wet/y
Final wet compost	23,591,865.77	kg wet/y
FS in final compost	4,566,112.88	kg FS/y
Final candidate coated product	9,094,370.02	kg wet/y
Predried core allocated	8,912,482.62	kg/y
PHA used in coating	181,887.40	kg/y
Physical surplus PHA	667,385.05	kg/y
Remaining wet compost	7,549,397.05	kg wet/y
Chloroform circulation and supply	3,455,860.61	kg/y
Captured solvent output	3,421,302.00	kg/y
Fugitive solvent air stream	34,558.61	kg/y

2.1. Goal, Scope, and Functional Unit Definition

The goal is to quantify the change associated with prospective PHA recovery and candidate fertilizer coating within separate SS and OFMSW treatment systems. SS-B and OFMSW-B are the respective baselines; SS-PHA and OFMSW-PHA add the integration stages. Comparisons are made within each feedstock pair. Capacities, incoming material and existing operations differ between the systems, so the comparison does not establish intrinsic feedstock suitability.

The functional unit is treatment of 1 t of incoming wet SS or incoming wet OFMSW at the stated entry point [28,33,34]. SS enters as dewatered sludge: 0.72 kg/m^3 wastewater multiplied by 5,625,634 m^3/y gives 4,050,456.48 kg wet SS/y. The common upstream wastewater line is excluded from the SS comparison. The OFMSW input is 145,000 t raw wet waste/y. Annual characterized exchanges are divided by these feedstock-specific wet tonnages. Physical outputs are reported separately; impact ratios per kilogram of nominal product are not interpreted as allocated product footprints.

2.2. System Boundaries

The foreground includes pretreatment and dilution where required, fermentation, cultivation, recovery, digestion, CHP, separation, composting, core preparation and coating. Harvest liquid and extraction residues return to digestion; the final digestate liquid fraction is assigned a residual treatment proxy. Figure 1 retains the original conceptual boundaries; the revised routing is specified here and in the inventory methods. Common reject disposal beyond transport is excluded from both members of a feedstock pair because identical quantities and properties are assumed; absolute totals therefore describe the modeled subsystem. Agricultural application, crop response and product end of life are outside the quantified comparison. Unverified changes in field emissions are not credited. In the original conceptual diagrams, cultivation/recovery liquid arrows are interpreted in the revised inventory as returns to digestion before final liquid treatment, not as direct discharges. Dashed substitution boxes are conditional: reference peat, nutrient and PET credits and external heat export are zero. The coating heater is included in equipment electricity without a duplicate external duty.

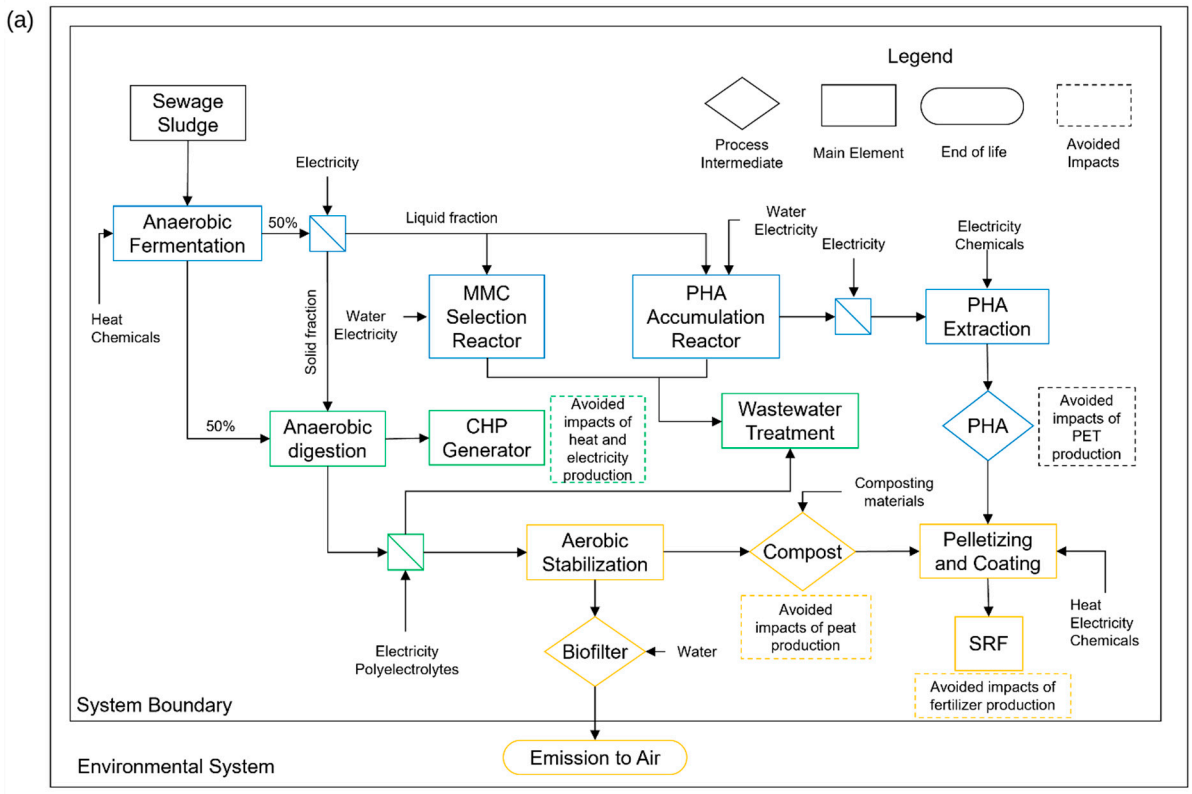


Figure 1. *Cont.*

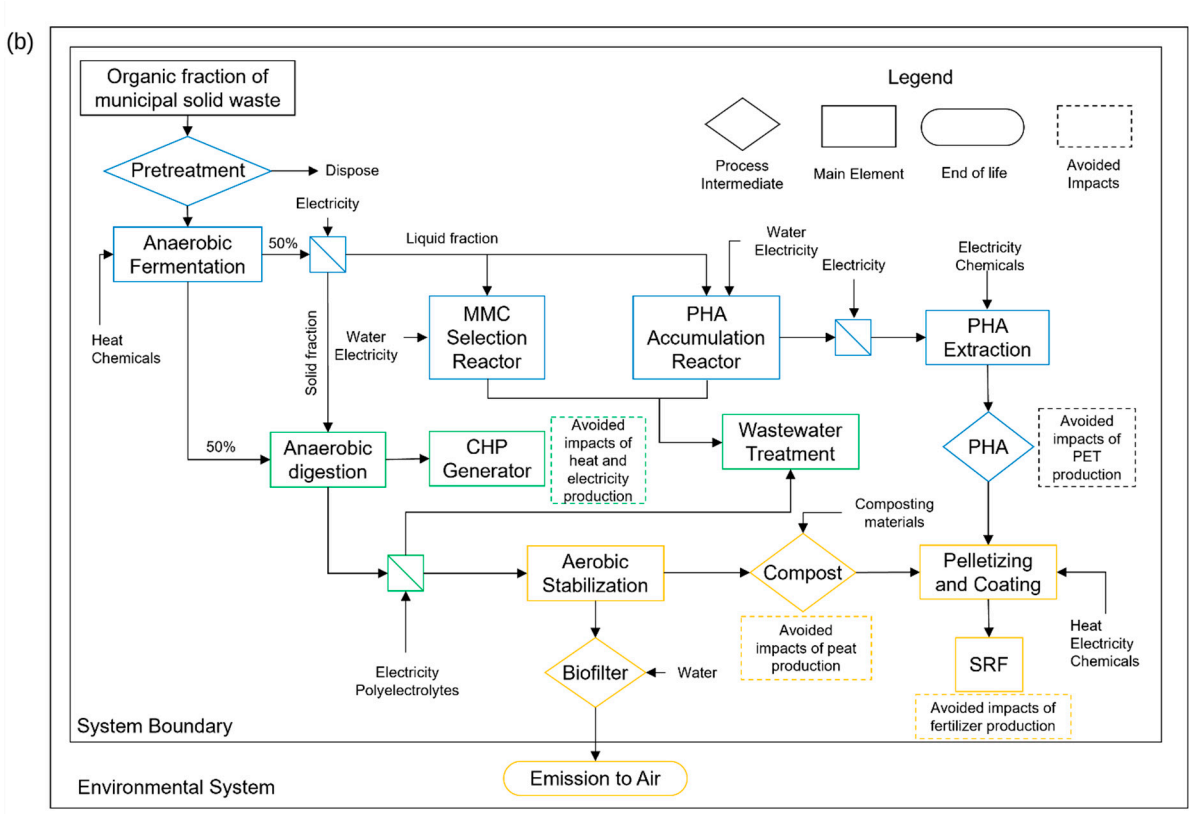


Figure 1. LCA system boundaries of (**a**) SS-PHA and (**b**) OFMSW-PHA. Original diagrams retained; revised residual routing and conditional credits follow the inventory methods. SRF labels denote candidate coated fertilizer, without demonstrated slow-release performance.

System expansion treats displaced energy and materials as separate avoided exchanges. Internal CHP electricity and heat meet process demand before any export is considered. No additional credit is assigned to self-consumed energy. Reference cases assign no fertilizer, peat or PET credit because functional equivalence is unverified. Sensitivity cases retain the earlier compost-use fractions and equivalent-product factors as conditional upper cases [30,35,36] A nutrient equivalent is not a physical fertilizer core, and PHA used in coating is excluded from surplus polymer.

2.3. Scenario Descriptions

Four configurations retained the original study design. The two baselines digested their accepted feed and composted the separated solid residue. The two integration configurations added acidogenic fermentation and diverted a clarified liquid stream to culture selection and PHA accumulation. All non-recovered organic material was assigned to an explicitly defined residual stream. The revised calculation did not apply an independent 26% biogas enhancement to the fixed digestion load.

Mass was tracked through water, VS, and particulate or dissolved FS; TS was calculated as their dry-solids sum. COD was tracked as an oxygen-equivalent quantity and was not added to wet mass. A carbon surrogate was also incorporated to constrain methane and CO_2 partitioning. Reagent carriers, wash water, polymer addition, reaction water, evaporation, and gas outlets were explicitly accounted for. Organic composition was represented by lumped CHO surrogates constrained by dry mass, COD, and carbon; nitrogen, sulfur and chlorine speciation were not independently validated.

2.4. SS-B Scenario

SS-B retained the Viareggio case-study entry flow and source composition, consisting of 25% TS and 78.9% VS/TS [37]. COD was represented by 1.42 kg COD/kg VS, a declared surrogate based on the source biomass conversion [38,39] Water was added to meet the source 5% digestion TS target [40]. The source 56% organic conversion was applied jointly to VS, COD and surrogate carbon. FS were conserved rather than independently reducing total solids by 50%. Digestate wet mass was calculated following gas removal and reaction water; its composition determined the dewatering requirements, polymer dose and composting input.

SS-PHA retained fermentation at the 4.3% TS target, the source VFA yield of 0.21 kg COD/kg feed VS and the source separation concept [9,38,41–43] Half the fermentate was separated; captured particles and entrained soluble matter returned to digestion, while the clarified liquid entered cultivation. The first separator applied a 74% capture rate to particles, not to dissolved VFA. Cake moisture content and solution concentration were used to determine liquid entrainment. Source selection yield was retained. The 32% PHA content was consistently interpreted as a fraction of final cell dry mass, including stored polymer; the corresponding accumulation COD yield was derived from nonpolymeric biomass and the source monomer factors.

OFMSW-B retained 145,000 t raw wet waste/y and 7.6% wet rejects. The 120 g TS/kg and 107 g VS/kg values in the cited study describe mechanically pretreated wet OFMSW [44] rather than dry-solid yields per ton of raw waste. The reference therefore retained 39.7% incoming raw TS, assumed rejects had the same TS and VS/TS composition as incoming material, and added process water to obtain 12% TS in the accepted stream. This was an explicit prospective pretreatment design, not a new observation from the original plant. Accepted TS equaled incoming TS minus reject TS; it was not fixed at 17,400 t/y. The source COD factor was 0.857 kg COD/kg accepted TS. Equal composition of raw and accepted organic matter was a transfer assumption tested through pretreatment sensitivity cases.

OFMSW-PHA shared the same pretreatment definition as OFMSW-B and retained the source acidification, selection and accumulation yield coefficients [16,44]. Fermentation dilution was set to 7.5% TS; selection liquid concentration and reactor volumes were calculated based on the source loading/retention assumptions. The 0.44 kg PHA COD/kg accumulation VFA COD yield was used with the 0.24 biomass growth yield, and oxygen demand was calculated based on the remaining COD. A separate 45% cell-content constraint was not imposed simultaneously. A 74% particle and polymer capture was followed by 90% extraction in the reference; 70–100% extraction was tested. Carbon and COD remaining after recovery and hypochlorite oxidation determined the digestion load.

2.5. Relevant Assumptions of Scenarios

The corrected reference replaced loss-free CHP conversion with 40% electrical and 50% recoverable thermal efficiency, consistent with the energy assumptions in [44]. Methane production was calculated based on COD conversion and the carbon surrogate; CO_2 was calculated based on the remaining converted carbon. Thus, gas volume and heating value were outputs of the same balance, not independent yield multipliers. Fermentation gas retained the source volumetric assumption and was represented as a methane/CO_2 surrogate with explicit heating value. Methane leakage was assumed at 1%, deducted from useful fuel and characterized as an emission. Useful heat exports were zero without a documented external sink.

2.6. Coupled Inventory Reconstruction and Annual Energy Accounting

The four foreground configurations were recalculated using a shared, coupled inventory model. Wet mass, water, TS, VS, FS, COD and a carbon surrogate were balanced across the linked process stages; annual material and energy flows, gas generation and residual treatment exchanges were derived from these balances. Source values and prospective design assumptions are defined in the Materials and Methods Section. Thirty sensitivity cases were recalculated using the same dependencies. The calculations are prospective model results and have not been validated against new plant measurements.

Incoming, intermediate and outgoing wet mass, water, TS, VS, FS and COD were tracked separately. First-stage separator soluble matter was calculated based on water partition; captured particles were calculated based on the assumed efficiency. The final digestate separator additionally accounted for dissolved salts in its liquid water and retained dry polyelectrolyte in the cake. Complete foreground identities were checked for wet mass, water (including net reaction water), dry solids (including organic conversion), FS, COD, surrogate carbon and recovered-polymer allocation. These were conservation checks on the declared lumped model, not proof of measured process behavior.

Annual aeration was calculated using the COD consumed minus COD retained in nonpolymeric biomass and PHA, multiplied by the source 0.5 kWh/kg O_2 coefficient [39]. Mixing was estimated based on working volumes, an assumed 6.6 W/m^3 and 8760 h/y. Pumping was calculated using calculated wet transfers, assuming 10 m equivalent head and 65% efficiency. Every separator was charged at 2.2 kWh/t actual wet inlet [45]. These values represent declared scale-up assumptions. Reactor working volumes followed annual flow divided by 365 days and the specified retention time; the selection concentrations imply explicit dilution rather than an oxygen-stock calculation.

Reactor heating was calculated using wet throughput, 4.18 kJ/(kg K), 20 °C incoming temperature, 37 °C operation, 50% sensible heat recuperation and an assumed standing loss of 0.5 kWh/(m^3 reactor day). Previously heated fermentate and returned ambient-temperature cultivation/recovery streams were determined through digestion heating. Composting input was calculated using annual cake throughput, evaporation to the target moisture and a declared usable fraction of biological oxidation heat. The previously assumed heating duty of 1584 kW over an 18-day period was not extrapolated to annual heat demand. Cultivation was assumed at ambient 20 °C; cooling and detailed thermal integration require further design validation.

All harvest liquid and extraction residues returned to digestion. Recovered polymer therefore reduced residual COD, whereas unrecovered polymer remained in the residual load. A source 5% hypochlorite solution dose was applied to captured organic solids; its available oxygen was assigned to limited oxidation of residual organic matter, with NaCl accounted for as dissolved FS. Wash water and water evaporated during PHA drying were explicitly defined. The interpretation of the supply coefficient of the source solution mass was conservatively retained and tested against an active-mass interpretation. Detailed oxidation byproducts, polymer purity and salt inhibition remain uncertain.

Composting retained the source 12% yard waste and 63% mineral bulking ratios [30]. Yard waste was assumed to comprise 50% TS and 90% VS/TS, with transport included under a waste cut-off convention. Mineral bulking was explicitly screened and returned as an internal circulating stream; it was not accounted for as a net compost ingredient or an avoided product. Compost mass followed conserved FS, retained organic solids and a 50% final TS target rather than a fixed 30% wet yield. Biogenic organic conversion was assumed at 35% and tested at 20–50%. Source CH_4, N_2O and CO factors were applied to actual wet cake; NH_3 was represented as a declared 0.5 kg/t wet-cake allowance, not a regulatory concentration limit.

The product was defined as 0.98 kg predried core plus 0.02 kg PHA per kilogram of final coated product. The core was specified at 90% TS; its preparation removed water from the allocated wet compost and was assigned a separate drying requirement. Coated output was limited by the availability of both the core and recovered polymer. Remaining wet compost and surplus polymer were considered separate physical outputs. A 5 wt% PHA coating solution was assumed, resulting in 19 kg chloroform per kilogram of coating PHA. The earlier equal solvent/polymer quantities implied an unverified 50 wt% solution. The 0.8919 kWh/kg equipment requirement included the coating heater; therefore, no additional external coating heat was assigned. Solvent capture/recovery and its added electricity were explicitly defined; the uncharacterized fugitive solvent stream is reported separately.

2.7. Acquisition of LCI Data

Foreground inputs and eleven exchange-specific characterization vectors are based on the source study and the original inventory structure, with unit and formula corrections described here. Electricity imports and exports use the same inherited grid vector. Specific background dataset identifiers, complete geography and system model metadata, and the documented transition between database versions are unavailable. Accordingly, neither ecoinvent 3.3 nor 3.8 is asserted as a fully verified exchange-level configuration. This limitation is separate from conservation of the modeled foreground.

2.8. Life Cycle Impact Assessment (LCIA)

Eleven midpoint categories were calculated using the inherited CML-labeled coefficient vectors [46]. The characterized result is the sum of revised exchanges multiplied by their category-specific vectors. No cross-category sum or person-equivalent interpretation was used. CH_4 and N_2O retained source GWP factors of 28 and 265. Gross burdens and energy credits are reported separately; reference material credits are zero. The fugitive chloroform air flow and detailed residual chemical speciation were not characterized by the inherited factors. Toxicity-related results therefore exclude direct characterization of these releases and may understate their contribution.

Thirty cases were recalculated using the same material, COD and energy model. These cases examined variations in polymer formation and recovery, aeration, equipment duty, digestion and composting conversion, CHP efficiencies, heat recuperation, coating solution concentration, methane leakage, residual treatment, chemical factor interpretation and substitution assumptions. Changes in PHA yield and recovery were propagated through respiration, residual digestion COD, gas generation, separator demand and compost output. These results represent coupled scenario calculations, not statistical uncertainty intervals. Reference assumptions and executed ranges are summarized in Supplementary Tables S5 and S6.

3. Results and Discussion

3.1. LCA Results

Table 5 reports characterized results from the recalculated reference inventory. Reference GWP was -40.29 and 228.45 kg CO_2 eq/t wet SS for baseline and PHA integration, and -81.13 and 363.59 kg CO_2 eq/t wet OFMSW. Integration-minus-baseline differences were 268.74 and 444.72 kg CO_2 eq/t, respectively. These values concern the stated subsystem and design assumptions; common reject disposal and agricultural use are outside the quantified boundary. The results do not demonstrate plant performance or intrinsic product footprints.

Table 5. Revised characterized impacts per ton of incoming wet waste. Reference case excludes unverified material credits and external useful heat export. Totals retain the declared residual treatment and background data limitations.

Category	Unit/t Wet Waste	SS-B	SS-PHA	OFMSW-B	OFMSW-PHA
AD-elements	kg Sb eq	1.21×10^{-4}	7.15×10^{-4}	9.42×10^{-5}	9.97×10^{-4}
AD-fossil	MJ	-7.51×10^{2}	2.30×10^{3}	-1.20×10^{3}	4.04×10^{3}
GWP	kg CO_2 eq	-4.03×10^{1}	2.28×10^{2}	-8.11×10^{1}	3.64×10^{2}
ODP	kg CFC-11 eq	9.62×10^{-8}	1.46×10^{-2}	-1.71×10^{-7}	1.74×10^{-2}
HTP	kg 1,4-DB eq	-2.01×10^{1}	4.35×10^{2}	-6.63×10^{1}	7.24×10^{2}
FAETP	kg 1,4-DB eq	-3.20×10^{1}	8.55×10^{1}	-5.48×10^{1}	1.45×10^{2}
MAETP	kg 1,4-DB eq	-1.21×10^{5}	2.33×10^{5}	-1.96×10^{5}	4.17×10^{5}
TETP	kg 1,4-DB eq	-1.40×10^{-1}	2.93×10^{0}	-4.56×10^{-1}	4.89×10^{0}
PCOP	kg C_2H_4 eq	1.48×10^{-3}	3.94×10^{-2}	1.94×10^{-3}	6.57×10^{-2}
AP	kg SO_2 eq	3.33×10^{-1}	9.58×10^{-1}	3.85×10^{-1}	1.49×10^{0}
EP	kg PO_4 eq	1.36×10^{0}	1.60×10^{0}	1.16×10^{0}	1.52×10^{0}

SS: sewage sludge; OFMSW: organic fraction of municipal solid waste; AD-elements: abiotic depletion potential (elements); AD-fossil: abiotic depletion potential (fossil fuels); GWP: global warming potential; ODP: ozone depletion potential; HTP: human toxicity potential; FAETP: freshwater aquatic ecotoxicity potential; MAETP: marine aquatic ecotoxicity potential; TETP: terrestrial ecotoxicity potential; PCOP: photochemical ozone creation potential; AP: acidification potential; EP: eutrophication potential; CFC-11: trichlorofluoromethane; 1,4-DB: 1,4-dichlorobenzene.

Negative values indicate that modeled avoided burdens exceed characterized process burdens. They do not indicate that the treatment process itself has negative emissions. In the reference, baseline GWP benefits originate from net electricity exports. Both PHA integration configurations had higher characterized totals in all eleven categories in the executed reference. This statement is restricted to the current characterizations and assumptions, including incomplete solvent emission and residual treatment representation; it is not a general claim about PHA integration.

3.2. Contribution Analysis

Figures 2 and 3 separate characterized positive burdens from energy/material credits for all eleven categories. Each category is scaled by the sum of absolute stage contributions within that configuration, without summing across categories. Electricity demand is shown alongside generation through the net exchange; annual energy flows are summarized in Table 1. Figure 4 reports coupled GWP differences for the thirty cases. All figures were generated from the same revised numerical model.

3.3. Contribution Analysis of the SS Treatment Plant Case Study

PHA integration adds liquid conditioning, culture volumes, aeration, separation and downstream processing. For SS, interpreting 32% as a fraction of final cell dry mass changes the relationship between biomass and polymer formation. Uniform capture followed by extraction replaces selective loss-free PHA recovery. The non-recovered polymer and biomass remain in named digestion returns, so a polymer-yield change cannot be combined with an unchanged residual COD load.

In the SS reference, recovered dry PHA was 4254.11 kg/y. Available polymer limited coated output to 212,705.54 kg/y, with 4254.11 kg/y PHA in coating and 0.00 kg/y physical surplus. Remaining compost was 364,243.52 kg wet/y. These are conditional process outputs; the candidate core composition, polymer quality, coating feasibility and agricultural function have not been demonstrated.

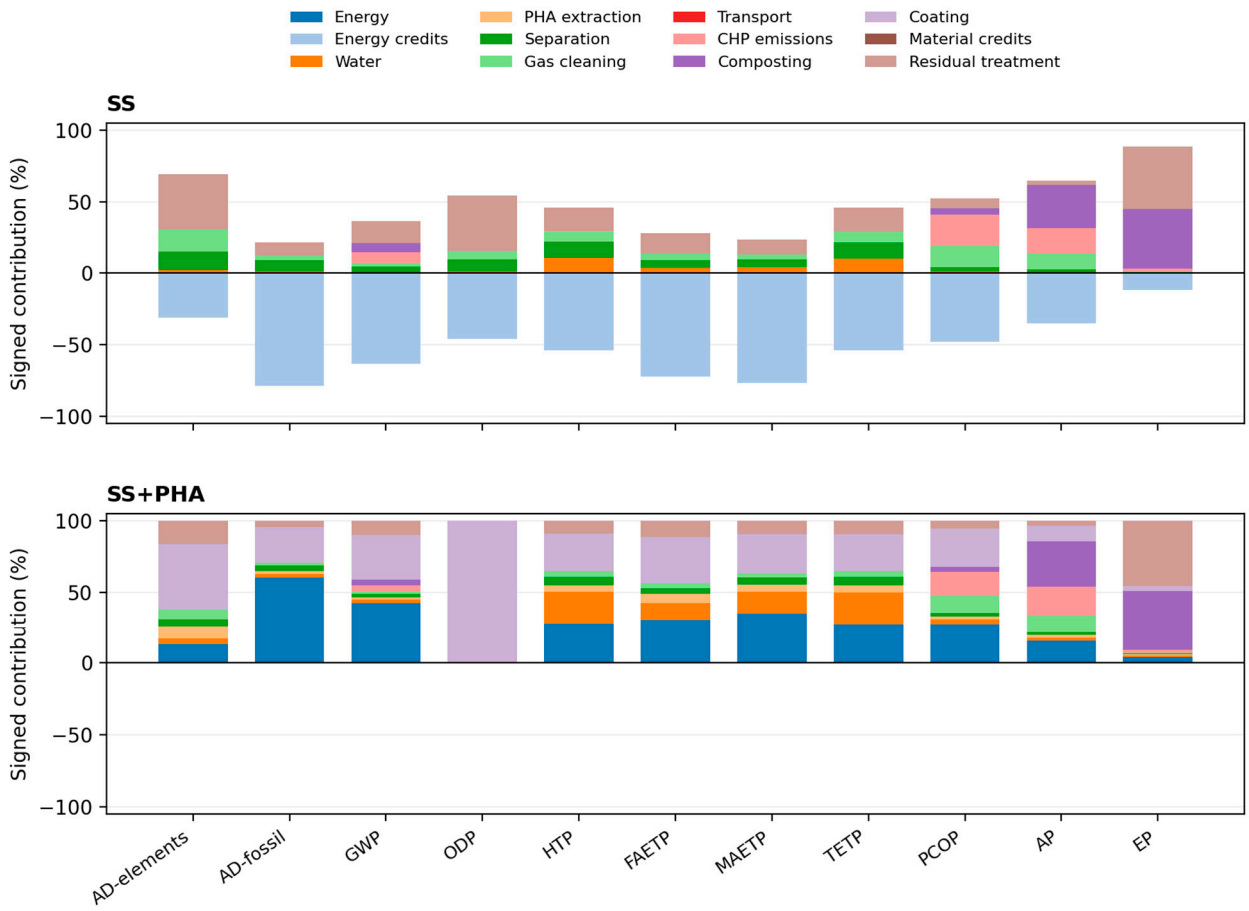


Figure 2. Revised characterized contributions for SS-B and SS-PHA, excluding common upstream wastewater treatment. Signed stage contributions are scaled within each impact category. The figure describes the reference design and characterized exchanges only.

For SS, the revised annual electricity demand was 335,854 kWh/y in the baseline and 981,343 kWh/y with PHA integration, compared with CHP generation of 873,575 and 833,505 kWh/y. Useful heat purchases were 0 and 1,024,032 kWh/y. Therefore, the corrected comparison includes differences in annual water handling and drying services that the earlier oxygen stock and fixed composting heat formulas did not represent.

Chloroform supply is a large characterized contribution to the integration configurations under the conservative reference scenario based on a 5 wt% PHA solution, without a material credit for recovered solvent. With 99% capture, the model reports a 1% fugitive air stream of 808.28 kg/y for SS-PHA and 34,558.61 kg/y for OFMSW-PHA. The inherited vectors characterize solvent supply but do not assign a characterization factor to this direct air release, so its contribution to toxicity-related categories cannot be quantified. The releases could add to toxicity-related impacts, and the reported categories may therefore be incomplete. Recovery electricity is included without assigning a material substitution credit. More concentrated solutions or a verified internal solvent loop require a defensible process design rather than removal of solvent burdens alone.

The recovered polymer displaces digestion substrates and changes both methane and CO_2 production through the shared COD/carbon calculation. The revised model therefore does not retain an independent gas-yield enhancement or a fixed source heating value where composition has changed. Methane leakage reduces useful fuel and adds a direct emission. These corrections link energy recovery to the residual inventory instead of using an environmental credit to compensate for an unbalanced material flow.

Conserving FS increases the solids that must be discharged in the cake or liquid relative to the source independent-removal formulas. The final liquid fraction is an explicitly quantified outlet: SS-PHA sends 62,857,867 kg/y of dewatering liquid to treatment (123,251 kg COD/y and 56,018 kg fixed solids/y), while OFMSW-PHA sends 4,349,606,692 kg/y (3,136,544 kg COD/y and 1,533,885 kg fixed solids/y). Harvest liquid and extraction residues return to digestion before final dewatering; the reported outlet is the post-digestion flow, not the gross internal return flow. The reference residual treatment model applies the wastewater treatment exchange vector from the source inventory in proportion to residual COD. It contributes 22.68 kg CO_2 eq, 39.05 kg 1,4-DB eq (HTP) and 0.73 kg PO_4 eq (EP) per ton of wet SS-PHA, and 16.12, 27.76 and 0.52 kg equivalents per ton of wet OFMSW-PHA, respectively. A twofold treatment burden case was included in the sensitivity analysis. This is a screening proxy rather than a validated plant design; sludge handling, nutrient partition, salinity and chemical speciation remain uncertain.

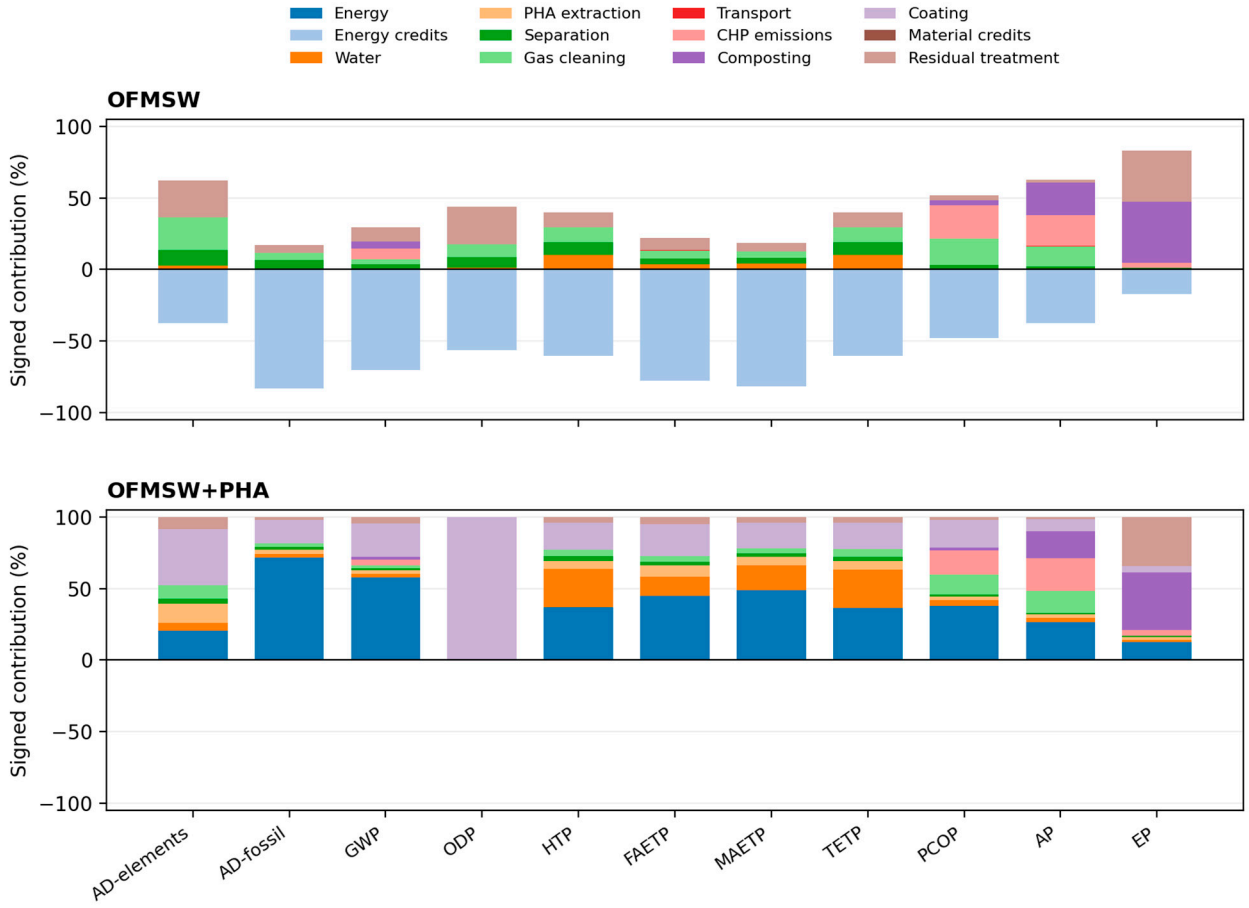


Figure 3. Revised characterized contributions for OFMSW-B and OFMSW-PHA. Pretreatment, water, energy and residual management exchanges use the same revised reference configuration summarized in Table 1. Credit signs and denominators follow Figure 2.

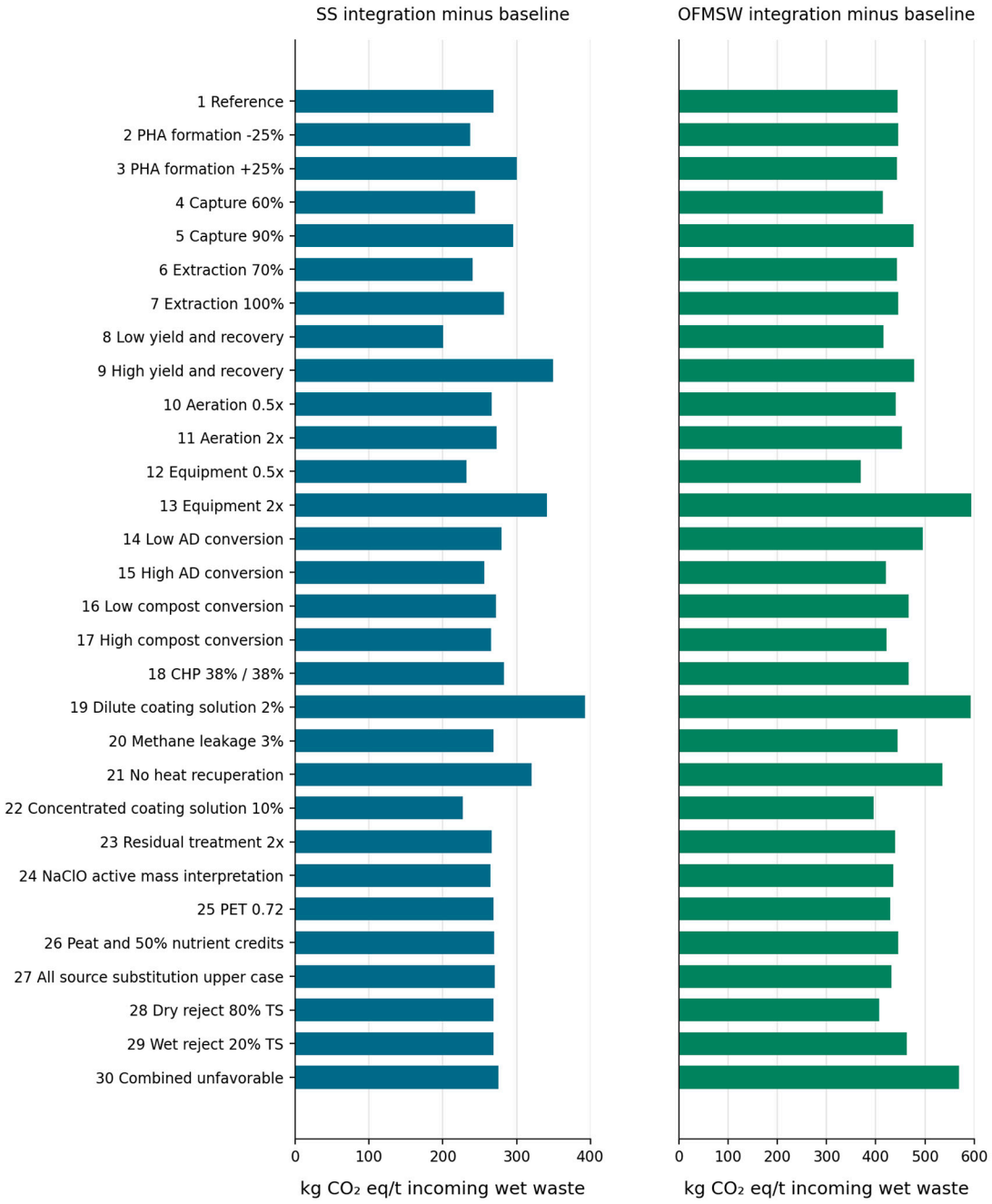


Figure 4. Thirty coupled sensitivity cases. Bars show integration minus the corresponding baseline in kg CO_2 eq/t incoming wet waste. Every case recalculated the mass, COD and energy model; the depicted range is not a statistical confidence interval.

3.4. Contribution Analysis of the OFMSW Treatment Plant Case Study

OFMSW pretreatment dilution was 309,270.50 t water/y in the reference. Accepted TS was 53,190.06 t/y, derived from raw composition and wet rejects. This replaces the incompatible interpretation of 120 kg TS/t incoming raw waste. Cultivation and subsequent liquid returns further changed hydraulic demand. The large annual water and energy requirements are therefore a consequence of transferring the selected wet-process concentrations to this prospective capacity, not an observed operating record; opportunities for validated water reuse and process intensification are important future design questions.

OFMSW-PHA produced 849,272.45 kg/y recovered dry polymer under 74% capture and 90% extraction. Candidate coated output was 9,094,370.02 kg/y, using 181,887.40 kg/y

PHA and leaving 667,385.05 kg/y physical surplus. Predrying removed 7,129,986.10 kg water/y from the actual allocated compost core. The reference assigned no PET credit to surplus polymer. A conditional 0.72 kg PET/kg surplus case was retained for comparison with the earlier substitution assumption [30].

The source 1.25 nitrogen and 1.03 phosphorus enrichment factors were removed from physical product accounting. Coating does not create nutrients. Nutrient-equivalent sensitivity quantities used actual dry compost outputs and conditional availability, with elemental P and K converted to P_2O_5 and K_2O where required. These cases do not demonstrate agronomic equivalence, regulatory eligibility, enhanced uptake or reduced field emissions. Physical coated mass and avoided mineral fertilizer equivalents remained separate in the inventory and impact accounting.

3.5. Sensitivity Analysis

All thirty executed cases retained positive integration-minus-baseline GWP differences, spanning 200.32 to 392.48 kg CO_2 eq/t wet SS and 369.84 to 594.47 kg CO_2 eq/t wet OFMSW. The eleven characterized category totals were also higher for integration in these particular cases. These findings include the tested coupled physical changes and differ from the earlier frozen inventory sensitivities. They remain limited to the specified parameter combinations, inherited vectors and characterization scope, and do not demonstrate all possible process designs or a probabilistic robustness statement.

Yield and recovery changes simultaneously alter oxygen demand, recovered polymer, residual COD, gas generation and compost production. Improved recovery can reduce residual gas potential while increasing a polymer-limited coating load; consequently, higher yield does not necessarily reduce total system impacts. The scenarios provide a computational comparison of the defined multifunctional waste treatment systems. They do not identify an optimized plant or an optimum polymer yield. An extensive validated parameter distribution and matching future grid coefficient vectors are not available.

A 2050 electricity comparison was not calculated because a technology-share table alone does not define a characterized grid vector. Supplementary Table S1 corrects other sources while retaining the historical mix for context. A prospective future grid calculation would require matching technology datasets and a documented system model. Solvent-loop and improved water reuse scenarios likewise require compatible mass and energy data; no unverified future improvement is introduced into the reference.

The reported results separate demonstrated calculation behavior from unverified process assumptions. The equations and coefficient vectors can be reproduced, and the declared lumped foreground conserves mass, solids and COD within numerical tolerance. Feed sampling, reject composition, cell yields, reagent performance, solvent quality, wet separation, thermal duties and residual treatment have not been validated against new process data. Background dataset identifiers are unavailable. These limits constrain interpretation and any claim of compliance or industrial transferability.

4. Conclusions

The original integrated-biorefinary comparison was retained and recalculated on a shared prospective inventory. Conservation of fixed solids, explicit water and residual routing, coupled polymer recovery and consistent final-product coating replace the contradictory source formulations. In the reference model, PHA integration increased GWP by 268.74 kg CO_2 eq/t SS and 444.72 kg CO_2 eq/t OFMSW. This increase persisted in the thirty specified coupled cases. Added hydraulic, energy, drying and solvent services are important constraints in this configuration; zero unverified product credits in the reference prevent assuming environmental benefit from nominal outputs. The findings concern a con-

ditional process design, not measured plant performance, intrinsic feedstock superiority or demonstrated slow-release fertilizer behavior. Further process data and verified background mappings are required before industrial or agronomic conclusions can be generalized.

Additional energy, assumptions, sensitivity, provenance and conservation details are provided in Supplementary Tables S4–S12.

Gross process demand, CHP generation, net imports/exports and useful heat are separate quantities. Heating and drying refer to annual processed mass; reactor losses refer to working volume and 365 operating days. Coating includes its electrical heater, while predrying the compost core and drying recovered PHA are distinct thermal services. The model does not infer an annual duty by multiplying a nameplate power by a single 18-day operating period.

Recovered PHA, coating allocation and physical surplus satisfy a separate polymer identity. The amount of source compost directed to the core is constrained by polymer availability and the predrying specification. The remaining compost is still a physical output; it is not removed when coating is limited by polymer availability. Credit factors were applied only to their corresponding eligible equivalent-product flows in the explicitly labeled sensitivity cases.

Wet mass, water (including net reaction water), dry solids (including conversion), FS, COD, surrogate carbon and polymer allocation satisfy requirements within numerical tolerance across all four reference configurations and the thirty evaluated cases. Soluble salts depend water partition and particulate FS are explicitly defined in the cake/liquid outlets. These identities establish computational conservation of the selected configuration; they do not validate the assumed composition, kinetics, separation coefficients or external residual treatment proxy.

Supplementary Materials: The following supporting information can be downloaded at https://www.mdpi.com/article/10.3390/environments13120656/s1. Sources used to compile the Supplementary Materials are included in the manuscript reference list and cited here [47–67]. The Supplementary Materials contain the following tables: Table S1. Historical European electricity-generation shares for the 2050 scenario, retained for context only. The "Other sources" category replaces the former "Geothermal" label. No future-grid LCIA is executed from these shares alone. Table S2. Revised percentage change, 100 × (integration − baseline) / |baseline|. Positive values indicate higher characterized impacts. Values concern the current characterization scope; near-zero ODP baselines are reported through absolute results in main Table 5 instead of unstable percentages. Table S3. Principal reference assumptions. These are model inputs, not new measurements. Table S4. Revised annual energy balance, kWh/y. Internal useful heat is accounted for once; external heat export is zero in the reference. Equipment and residual-treatment duties are prospective assumptions. Table S5. Coating and downstream assumptions used consistently in the revised reference. Source equipment duties are retained where applicable; new concentration and moisture assumptions are explicit. Table S6. Executed sensitivity ranges. These are scenario bounds, not measured confidence intervals or an exhaustive uncertainty distribution. Table S7. Entry flow and functional-unit conventions. Comparisons remain within each feedstock pair. Table S8. Background characterization provenance and availability. Scalar vectors permit numerical replication; dataset-level reconstruction remains limited. Table S9. Principal changes to the original inventory and their revised implementation. Table S10. Revised physical product and residual outputs. No allocated product footprint, agronomic equivalence or universal PET displacement is inferred. Table S11. Thirty evaluated coupled cases. GWP difference is PHA integration minus its own baseline, kg CO_2 eq/t incoming wet waste. Counts refer to the eleven characterized totals within the stated model scope. Table S12. Reference foreground conservation checks. Residuals are kg/y or kg COD/y; absolute numerical tolerance is 0.001. The checks concern the declared lumped foreground and separate ancillary ledgers, not measured elemental or plant validation.

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