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Article

Life Cycle Assessment of the Mesophilic, Thermophilic, and Temperature-Phased Anaerobic Digestion of Sewage Sludge

Iryna Lanko [1,2,*], **Laura Flores** [1], **Marianna Garfí** [1], **Vladimir Todt** [3], **John A. Posada** [4], **Pavel Jenicek** [2] **and Ivet Ferrer** [1,*]

1   GEMMA—Group of Environmental Engineering and Microbiology, Department of Civil and Environmental Engineering, Universitat Politècnica de Catalunya·BarcelonaTech, c/Jordi Girona 1–3, Building D1, 08034 Barcelona, Spain; laura.flores.rosell@upc.edu (L.F.); marianna.garfi@upc.edu (M.G.)
2   Department of Water Technology and Environmental Engineering, University of Chemistry and Technology, Technicka, 5, 166 28 Prague, Czech Republic; jenicekp@vscht.cz
3   Veolia Česká Republika, a.s., Na Florenci 2116/15, Reception D, 110 00 Prague, Czech Republic; vladimir.todt@veolia.com
4   Department of Biotechnology, Delft University of Technology, van der Maasweg 9, 2629 HZ Delft, The Netherlands; J.A.PosadaDuque@tudelft.nl
*   Correspondence: irynalanko@gmail.com (I.L.); ivet.ferrer@upc.edu (I.F.)

Abstract: In this study the environmental impact of the anaerobic digestion (AD) of sewage sludge within an activated sludge wastewater treatment plant (WWTP) was investigated. Three alternative AD systems (mesophilic, thermophilic, and temperature-phased anaerobic digestion (TPAD)) were compared to determine which system may have the best environmental performance. Two life cycle assessments (LCA) were performed considering: (i) the whole WWTP (for a functional unit (FU) of 1 m^3 of treated wastewater), and (ii) the sludge line (SL) alone (for FU of 1 m^3 of produced methane). The data for the LCA were obtained from previous laboratory experimental work in combination with full-scale WWTP and literature. According to the results, the WWTP with TPAD outperforms those with mesophilic and thermophilic AD in most analyzed impact categories (i.e., Human toxicity, Ionizing radiation, Metal and Fossil depletion, Agricultural land occupation, Terrestrial acidification, Freshwater eutrophication, and Ozone depletion), except for Climate change where the WWTP with mesophilic AD performed better than with TPAD by 7%. In the case of the SL alone, the production of heat and electricity (here accounted for as avoided environmental impacts) led to credits in most of the analyzed impact categories except for Human toxicity where credits did not balance out the impacts caused by the wastewater treatment system. The best AD alternative was thermophilic concerning all environmental impact categories, besides Climate change and Human toxicity. Differences between both LCA results may be attributed to the FU.

1. Introduction

In conventional activated sludge wastewater treatment plants (WWTP), excess sludge is continuously formed at the biological reactor of the wastewater treatment line. WWTP operational expenses to handle the produced excess biological sludge, namely waste activated sludge (WAS), together with primary sludge may go up to 50% [1–3]. It has been a long time since AD was adopted as one of the most effective solutions of sewage sludge treatment in terms of sludge reduction, stabilization,

and resource recovery [4–6]. Previously, sewage sludge was considered only as waste, and its disposal on a regular basis was quite challenging [7,8]. Population growth and natural resources exhaustion made it crucial to find a way out of this situation. Nowadays, sludge is considered as a source of substances that can be recovered and reused [9–11].

The zero-waste approach and circular economy paradigm change the angle at which nutrients, metals, organic matter, and other substances from WAS can be converted into valuable materials like biofuels and biofertilizers. Resource recovery processes are being widely adopted, even though recovery of certain substances, for instance, phosphorus still can be nonprofitable from the perspective of economics [12] or environmental impacts [13]. This is mostly because of the low concentration of valuable substances in the influent wastewater. However, as the world prices for phosphorus as for irreplaceable fertilizer are increasing, its recovery from sludge has become extremely important in a long-term perspective [14]. Currently, the attention is drawn to the reject water obtained after the AD process where the concentration of nutrients is significantly higher than in the influent wastewater [15]. However, the temperature regime and configuration of AD may significantly influence the nutrient concentration in the reject water and its volume [1,16,17].

AD is a biological process where organic matter is being biodegraded under anaerobic conditions, leading to the production of biogas, a gaseous biofuel mostly composed of methane, along with the digestate, which may be reused as biofertilizer. AD can be implemented with some variations in temperature (mesophilic (M), 35–40 °C and thermophilic (T), 55–70 °C) [18] and configuration (one- and two-stage reactors) [1,19]. Normally, the AD process consists of four main stages, namely hydrolysis, acidogenesis, acetogenesis and methanogenesis, which take place in the same reactor in the case of one-stage AD. Separate functioning of the first thermophilic (hydrolytic and acidogenic) and second mesophilic (acetogenic and methanogenic) stages in two-stage AD is accomplished to overcome the drawbacks of one-stage systems [20,21].

The most widely used anaerobic digester is a mesophilic one-stage, since its operation is known as the most simple and stable [22,23]. Nevertheless, the tendency changes towards more metabolically efficient and pathogenically safe digesters such as thermophilic one-stage or temperature-phased two-stage reactors [24]. Temperature phased anaerobic digestion (TPAD) implies a combination of thermophilic and mesophilic one-stage digesters and performs with better stability than thermophilic and higher organic matter degradation rate than mesophilic, which means increased energy efficiency and better control of process parameters [21,25].

Despite the increasing interest in thermophilic and TPAD systems' application, there are few studies comparing the environmental impact of full-scale WWTP with different AD systems [26] and none concerning the comparison of the environmental impact of mesophilic, thermophilic, and TPAD systems. The environmental impact assessment would allow for defining which AD system is the most beneficial in terms of environmental protection. Thus, the objective of this study is to evaluate and compare the environmental impacts of WWTP with different AD systems (M, T, TPAD) using the life cycle assessment (LCA) methodology.

One methodological challenge that leads to variability in LCA results for multiproduct systems, such as WWTPs, is the choice of a functional unit (FU) and its effect on the ecosystem. In this study, two FUs are selected in order to demonstrate a more comprehensive picture of environmental influence of AD implemented in the sludge line alone and at the whole WWTP [27].

The LCA approach is here used not only as a standard practice to estimate the environmental burden of the technological process [22,28,29] on a micro level to compare the three AD options, but also as an alternative to build up a regulatory planning system on a meso level to increase the efficiency of project-level decision-making and to provide advice on potential improvements for the sector's management, and also to ensure the realization of strategic environmental goals [30].

2. Materials and Methods

2.1. Anaerobic Digestion Systems

The LCA compared three different AD systems, namely, mesophilic, thermophilic, and TPAD. Data on the operation and performance of these systems treating the same sewage sludge was gathered from three lab-scale digesters which were run during five months [31]. The substrate was thickened WAS, disintegrated through centrifugation (total solids (TS) = 71.8 ± 3.4 g/L, volatile solids (VS) = 42.3 ± 4.1 g/L, chemical oxygen demand (COD) = 64.1 ± 4.1 g/L). The main features of the mesophilic, thermophilic, and TPAD lab-scale systems are shown in Table 1.

Table 1. Performance of the mesophilic, thermophilic and temperature-phased anaerobic digestion lab-scale systems. Values given as mean ± standard deviation.

	Process Variable (Units)	Thermophilic	Mesophilic	TPAD	
				1st Stage	2nd Stage
Operational conditions	Temperature (°C)	57 ± 1.5	38 ± 1.5	57 ± 1.5	38 ± 1.5
	HRT (days)	19	19	2	17
	V (L)	8.45	8.45	1.0	8.45
	OLR (g VS/L·day)	2.25	2.25	2.24	
Biogas production	Methane production rate (L CH$_4$/L·day)	7.8 ± 0.5	7.1 ± 0.6	2.5 ± 0.4	9.9 ± 0.2
	Methane yield (L CH$_4$/g COD)	0.17 ± 0.02	0.16 ± 0.02	0.05 ± 0.01	0.19 ± 0.02
	Methane content in biogas (% CH$_4$)	61.7 ± 3.1	66.1 ± 1.8	58.9 ± 5.3	70.9 ± 2.7
Removal efficiency	TS removal (%)	20.5 ± 2.4	18.5 ± 1.7	24.5 ± 2.2	
	VS removal (%)	27.6 ± 1.9	26.0 ± 2.6.	35.5 ± 1.2	
Effluent characteristics	pH	7.4 ± 0.06	7.5 ± 0.02	6.7 ± 0.13	7.6 ± 0.02
	TS (g/L)	57.1 ± 2.4	58.5 ± 1.7	55.3 ± 2.2	
	VS (g/L)	30.7 ± 1.9	31.3 ± 2.6	28.6 ± 1.2	
	VS/TS (%)	53.8	53.5	51.7	
	COD (g/L)	43.2 ± 4.8	44.6 ± 3.7	48.8 ± 5.7	42.1 ± 1.2

2.2. Life Cycle Assessment

LCA is an analytical tool which allows to assess the environmental impacts of a product, technology, or process according to the "cradle-to-grave" approach. This time-tested technique allows not only to evaluate the potential environmental risks, but also define the life cycle stage and type of environmental impacts. The application of this method may help to improve the studied product, technology, or process by making its life cycle more friendly to the environment. LCA consists of four main steps according to ISO 14040 (2006), and ISO 14042 (2006): (i) goal and scope definition; (ii) inventory analysis; (iii) impact assessment; (iv) result interpretation.

2.2.1. Goal and Scope Definition

The goal of this study was to compare the potential environmental impacts of three types of AD processes: (i) mesophilic; (ii) thermophilic; (iii) TPAD systems.

To this end, two LCA cases with two different functional units (FU) were conducted: first for an activated sludge WWTP with the three different AD systems (here named as WWTP-LCA); second for the sludge line alone with the three different AD systems (here named as SL-LCA).

Since it has been reported in literature that the choice of the FU may change the overall balance of environmental impacts from harmful to beneficial and vice-versa [28], these two FUs were chosen considering the major outputs and functions of a WWTP. For the first case, the FU was 1 m^3 of treated

wastewater, as the main function of the WWTP is to treat the wastewater stream. For the second case, the FU was 1 m^3 of produced methane, as one of the main functions in AD systems (also the secondary one within WWTP) is to produce energy out of the methane contained in the biogas. Methane was taken instead of biogas, as biogas might have a different content of methane depending on the AD system.

The two FUs were coupled with the system expansion approach (the alternative production of energy and fertilizers) and adopted to evaluate the environmental burden of AD at both scales, the whole WWTP (to assess the contribution of the sludge line to the whole WWTP) and the sludge line alone (to highlight the potential environmental benefits from methane production as an additional function of the system beyond the wastewater treatment). The application of two FUs would demonstrate a deeper, more comprehensive and more transparent picture of AD implementation at the sludge line and at the WWTP. Thus, applying the two FUs helps to present the environmental profile of each AD from different points of view [4,27].

For both LCAs, a period of one-year operation was considered, as it is a timescale that is long enough to assess the averaged operational parameters of any WWTP, including the fact that the construction part was not estimated in the impacts' analysis. The impacts of the construction phase were not accounted for, since the dimensioned WWTP is the same for all three scenarios and LCAs, and it would make the difference among three AD systems' exploitation less evident [4,32].

To sum up, the system's boundaries—for the first and second LCAs—consider the year-around operation of the whole WWTP with three different AD technologies (Figure 1a), and the sludge line alone with three different AD technologies (Figure 1b), respectively. Thus, three scenarios were considered in each LCA; namely mesophilic (M), thermophilic (T), and temperature-phased anaerobic digestion (TPAD).

2.2.2. Inventory Analysis

Inventory data for the WWTP-LCA and SL-LCA are summarized in Tables 2 and 3. Among the inputs and outputs, the flows of materials and energy resources, gaseous emissions, and solid wastes were considered. When possible, data from a full-scale facility was used, which was complemented by data gathered in the lab-scale set-up described previously (Section 2.1) [33].

Full-scale data were taken from a WWTP with thermophilic AD operated by Veolia Česká Republica, a.s. The full-scale operation was used to scale-up and validate the laboratory reactor results. The rest of the information for the mesophilic one-stage and TPAD systems was calculated based on different literature sources and benchmarking data of Veolia, a.s., as described below.

The input parameters included energy consumption, anti-bulking, anti-foaming and dewatering agent dosage, gaseous emissions from the disposed digestates, digestate amounts. Calculated parameters were energy consumption according to the literature [34], specific biogas production based on benchmarking data of Veolia, a.s., as well as anti-bulking, anti-foaming and dewatering agents' dosages.

The final digestate amount for each AD system was estimated in correspondence with the VS destruction observed in the lab-scale digesters (Table 1), which was consistent with other AD systems studied in the literature [19,35,36].

Since the lab-scale set-up treated WAS and the full-scale plant used sewage sludge with 32.5% WAS, the annual biogas production was recalculated for the thermophilic scenario based on the specific biogas production of the corresponding laboratory reactor.

The heat and electricity production was calculated based on the annual biogas production and technical equipment data (efficiency of combined heat and power unit) given by Veolia, a.s.

Annual gas emissions (CH_4 and N_2O) from the wastewater treatment process and disposed digestates were estimated according to the literature [37,38].

Background data (chemicals, avoided fertilizers—that are contained in the digestate and can be used in agriculture, transportation, wastewater treatment in a municipal wastewater treatment plant, solid wastes, energy provider) were taken from Ecoivent 3.1 database [33]. For all electricity

requirements, the Czech electricity mix was considered since the full-scale WWTP is located in the Czech Republic [39] (which is composed of fossil 58.5%, nuclear 35.3%, solar 2.8%, hydro 2.7%, wind −0.7%).
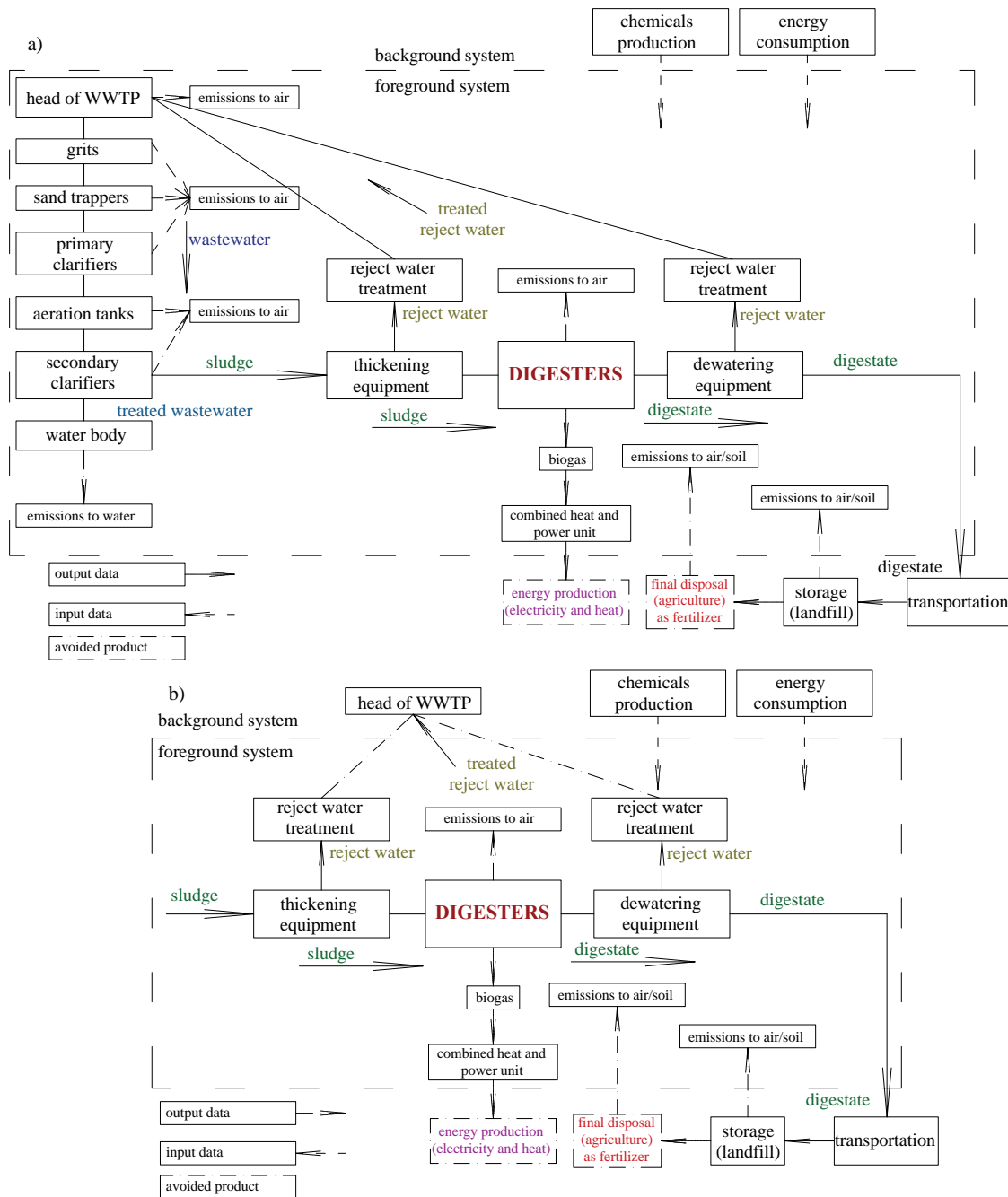
Figure 1. Flowcharts: (**a**) for life cycle assessment of the whole wastewater treatment plant (WWTP-LCA) and (**b**) for life cycle assessment of the sludge line alone (SL-LCA).

Table 2. Inventory data for the whole WWTP LCA (FU: 1 m^3 of treated wastewater).

Type of Data	WW/SL	Item	Referred to FU			Unit
			T	M	TPAD	
Input data						
Energy consumption	WW + SL	Total energy consumption	0.42	0.39	0.39	kWh/m^3
	WW	Only for WW	0.32	0.31	0.3	kWh/m^3
	SL	Only for SL	0.1	0.08	0.09	kWh/m^3
Reagents	WW	Phosphorus precipitation agent	0.07	0.07	0.03	kg/m^3
		Sludge anti-bulking agent	0.05	0.05	0.05	kg/m^3
	SL	Thickening agent	1.23×10^{-4}	1.23×10^{-4}	1.23×10^{-4}	kg/m^3
		Anti-foaming agent	2.63×10^{-4}	5.27×10^{-4}	5.27×10^{-4}	kg/m^3
		Dewatering agent	0.002	0.001	0.002	kg/m^3
		Chemo dezodoration agent	1.0×10^{-5}	1.0×10^{-5}	1.0×10^{-5}	kg/m^3
		Bio dezodoration agent	2.2×10^{-5}	2.2×10^{-5}	2.2×10^{-5}	kg/m^3
Output data						
Waste		Sand	0.02	0.02	0.02	kg/m^3
		Sand	0.001	0.001	0.001	t*km/m^3
		Coarse waste	0.04	0.04	0.04	kg/m^3
		Coarse waste	0.002	0.002	0.002	t*km/m^3
Wastewater air emissions	WW	CO	9	9	9	g/m^3
		SO$_2$	0.03	0.03	0.03	g/m^3
		Ozone	0.08	0.08	0.08	g/m^3
		N$_2$O	0.05	0.05	0.05	g/m^3
		H$_2$S	0.0008	0.0008	0.0008	g/m^3
		NH$_3$	2.8	2.8	2.8	g/m^3
		N$_2$O	1.8	1.81	1.81	g/m^3
		CO$_2$	81	81	81	g/m^3
		N$_2$	41	41	41	g/m^3
Wastewater contaminants		COD	0.04	0.04	0.04	kg/m^3
		SS	0.009	0.009	0.009	kg/m^3
		TN	0.02	0.02	0.02	kg/m^3
		TP	6.7×10^{-4}	6.7×10^{-4}	6.7×10^{-4}	kg/m^3
		As	9.6×10^{-6}	9.6×10^{-6}	9.6×10^{-6}	kg/m^3
		Pb	1.5×10^{-5}	1.5×10^{-5}	1.5×10^{-5}	kg/m^3
		Cd	1.2×10^{-6}	1.2×10^{-6}	1.2×10^{-6}	kg/m^3
		Cr	2.2×10^{-6}	2.2×10^{-6}	2.2×10^{-6}	kg/m^3
		Cu	2.9×10^{-5}	2.9×10^{-5}	2.9×10^{-5}	kg/m^3
		Ni	6.9×10^{-6}	6.9×10^{-6}	6.9×10^{-6}	kg/m^3
		Zn	3.2×10^{-5}	3.2×10^{-5}	3.2×10^{-5}	kg/m^3
		Hg	0.2×10^{-7}	0.2×10^{-7}	0.2×10^{-7}	kg/m^3
Digestate contaminants	SL	Dewatered digested sludge (wet)	0.82	0.86	0.78	kg/m^3
		Dewatered digested sludge (wet)	0.032	0.034	0.031	t*km/m^3
Avoided products		Electricity	0.23	0.22	0.35	kWh/m^3
		Heat	0.26	0.25	0.39	kWh/m^3
Air emissions		H$_2$S	2.0×10^{-4}	2.0×10^{-4}	2.0×10^{-4}	g/m^3
		CH$_4$	0.02	0.02	0.02	g/m^3
		CO$_2$, biogenic	28	28	28	g/m^3
		CO$_2$, not biogenic	44	44	44	g/m^3

Note: For T and TPAD, the transportation distance was 40 km to the sludge handling plant. According to the microbiological analyses and other sources [40], TPAD and T digestates meet the requirements of Class A biosolids. In case of M, due to pathogenic unsafety, the digestate was additionally treated by composting before its agricultural application. Therefore, it was transported over 2 × 40 km to the composting plant and back, so an additional energy consumption of 16 kWh/t of digestate for the purposes of composting was included [41].

Table 3. Inventory data for the sludge line LCA (FU: 1 m^3 of produced methane).

Type of Data	Item	Referred to FU			Unit
		T	M	TPAD	
Input data					
Energy consumption	AD energy consumption	1.32	1.12	0.79	kWh/m^3
	Energy consumption for composting	-	0.2	-	kWh/m^3
Reagents	Thickening agent	0.002	0.002	0.001	kg/m^3
	Anti-foaming agent	0.004	0.008	0.005	kg/m^3
	Dewatering agent	0.027	0.019	0.014	kg/m^3
	Chemo dezodoration agent	1.3×10^{-4}	1.4×10^{-4}	8.7×10^{-5}	kg/m^3
	Bio dezodoration agent	3.0×10^{-4}	3.1×10^{-4}	2.0×10^{-4}	kg/m^3
Output data					
Wastewater	Reject water	0.26	0.26	0.17	m^3/m^3
Wastes	Dewatered digested sludge (wet)	11.3	12.3	7.1	kgDM/m^3
	Dewatered digested sludge (wet)	0.45	0.49	0.28	t*km/m^3
Avoided products	Electricity	3.14	3.14	3.12	kWh/m^3
	Heat	3.51	3.51	3.49	kWh/m^3
Emissions	H$_2$S	0.003	0.003	0.002	kg/m^3
	CH$_4$	0.28	0.29	0.18	kg/m^3
	CO$_2$, biogenic	386	402	253	kg/m^3
	CO$_2$, not biogenic	607	633	398	kg/m^3

2.2.3. Impact Assessment

The LCA was performed with the software of *SimaPro® 8*. The potential environmental impacts were calculated by the ReCiPe midpoint method V1.12/Europe Recipe H [33].

Characterization was conducted for the following environmental impact categories: Climate change, Ozone depletion, Terrestrial acidification, Freshwater eutrophication, Human toxicity, Ionizing radiation, Agricultural land occupation, Metal depletion, and Fossil depletion [4]. The above mentioned nine environmental impact categories were selected and assessed considering their close connection with processes that take place in activated sludge WWTP with AD and that have been used in previous LCA studies [33,42].

Classification and characterization were performed as the only compulsory steps of impact assessment in terms of standards—ISO 14040 (2006) and ISO 14042 (2006).

2.2.4. Sensitivity Analysis

A sensitivity analysis on the digestate volume obtained from the TPAD system for both cases WWTP-LCA and SL-LCA was performed in order to take into account the influence that this parameter may have on the environmental impacts associated to digestate transport, treatment, and reuse.

The sensitivity analysis allowed to evaluate if and how the uncertainty of the assumed value in the inventory table could influence the final results. A variation of ±5% of the digestate volume was set for the TPAD scenario only, according to the variability of lab data obtained by the previous studies [19,26,35], and shortage of the reported data from full-scale WWTPs, since TPAD is the least spread AD system worldwide among others [43].

This analysis is done through the sensitivity coefficient (S) which indicates the sensitivity of a particular model output to the changes in the variable being considered. The S is calculated according to the following equation [44]:

$$\text{Sensitivity coefficient (S)} = \left(\frac{\text{Output}_{high} - \text{Output}_{low}}{\text{Output}_{baseline}}\right) / \left(\frac{\text{Input}_{high} - \text{Input}_{low}}{\text{Input}_{baseline}}\right) \quad (1)$$

where Input is the value of the input variable (i.e., digestate amount), and Output is the value of indicator according to the correspondent impact category.

3. Results and Discussion

3.1. Life Cycle Assessment

3.1.1. WWTP-LCA with Mesophilic, Thermophilic, or Temperature-Phased Anaerobic Digestion

The results of the LCA for the whole WWTP (WWTP-LCA) are shown in the Figure 2. This figure includes all environmental impact categories considered in this study, and within each impact category there are three scenarios: mesophilic, thermophilic, and TPAD. For each scenario, results are shown for the whole WWTP, and separately for the wastewater treatment line and the sludge treatment line; this disaggregation of results was done to better identify the contributions of each process stage to the overall impacts. Positive values represent the environmental impacts, while negative values refer to the avoided environmental impacts.

According to the results (Figure 2), the differences among the three AD scenarios are not significantly large, however there are some trends that are discussed here. First, the wastewater treatment line would lead to larger environmental impacts than those cases where the sludge line is incorporated into the AD system. Thus, any implemented AD improves the environmental status of the WWTP mainly due to the credits obtained from the substitution of electricity generation from the fossil fuels [45]. Similar results have been reported for other LCA studies on full-scale AD plants [4,26].

In general terms, TPAD has the lowest environmental impacts, in comparison to T and M, for eight out of the nine impact categories presented in Figure 2, except for Climate change. Furthermore, a one-to-one comparison between T and M shows that their calculated environmental impacts are virtually the same for five out the nine compared categories (i.e., Ionizing radiation, Agricultural land occupation, Metal depletion, Fossil depletion, and Freshwater eutrophication), and with a slightly better environmental performance (meaning lower environmental impacts) for T over M in two impact categories (i.e., Terrestrial acidification and Ozone depletion). T outperforms both M and TPAD in one category (i.e., Climate change), and has a slightly better performance than M in only one category (i.e., Human toxicity).
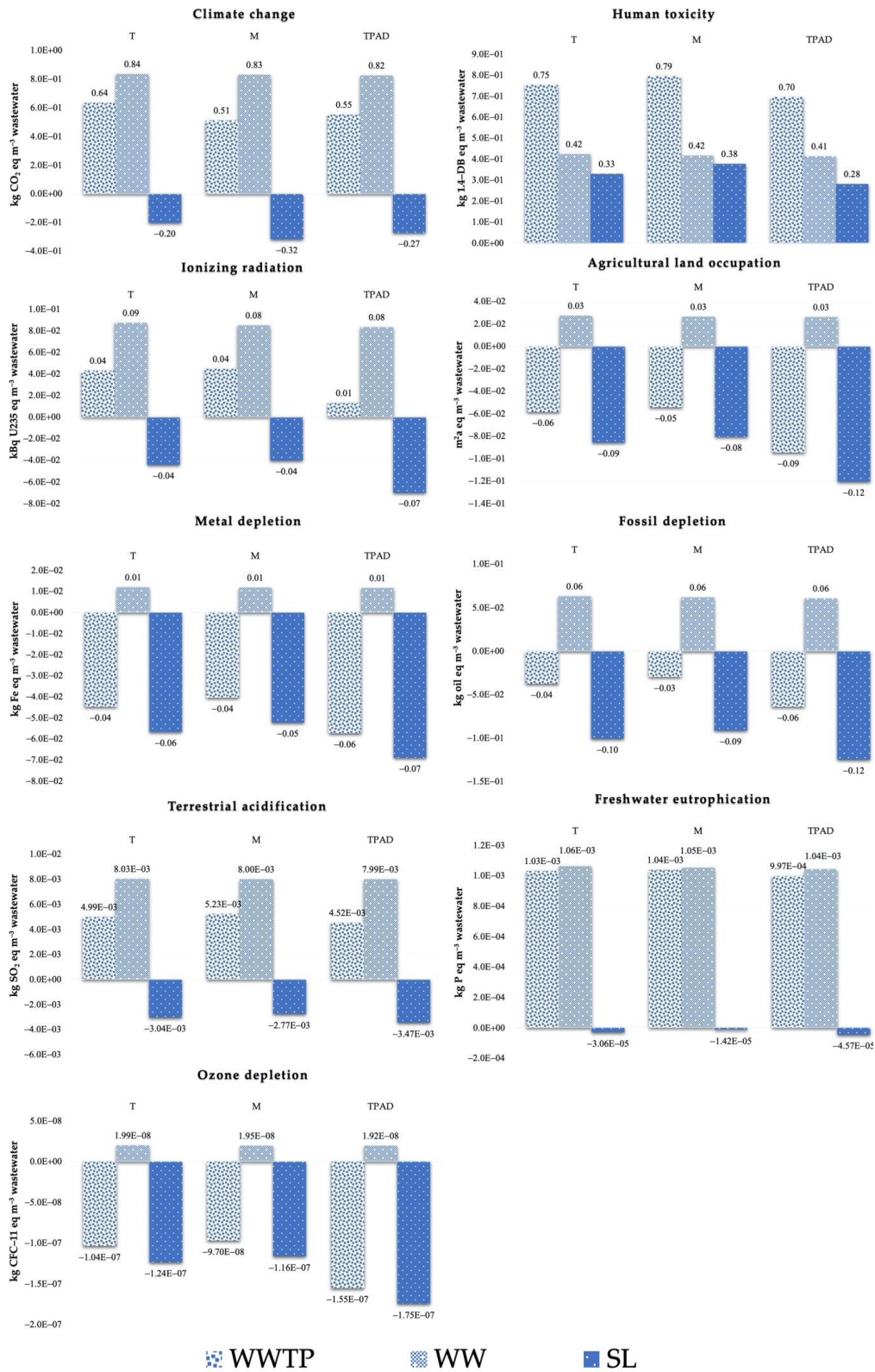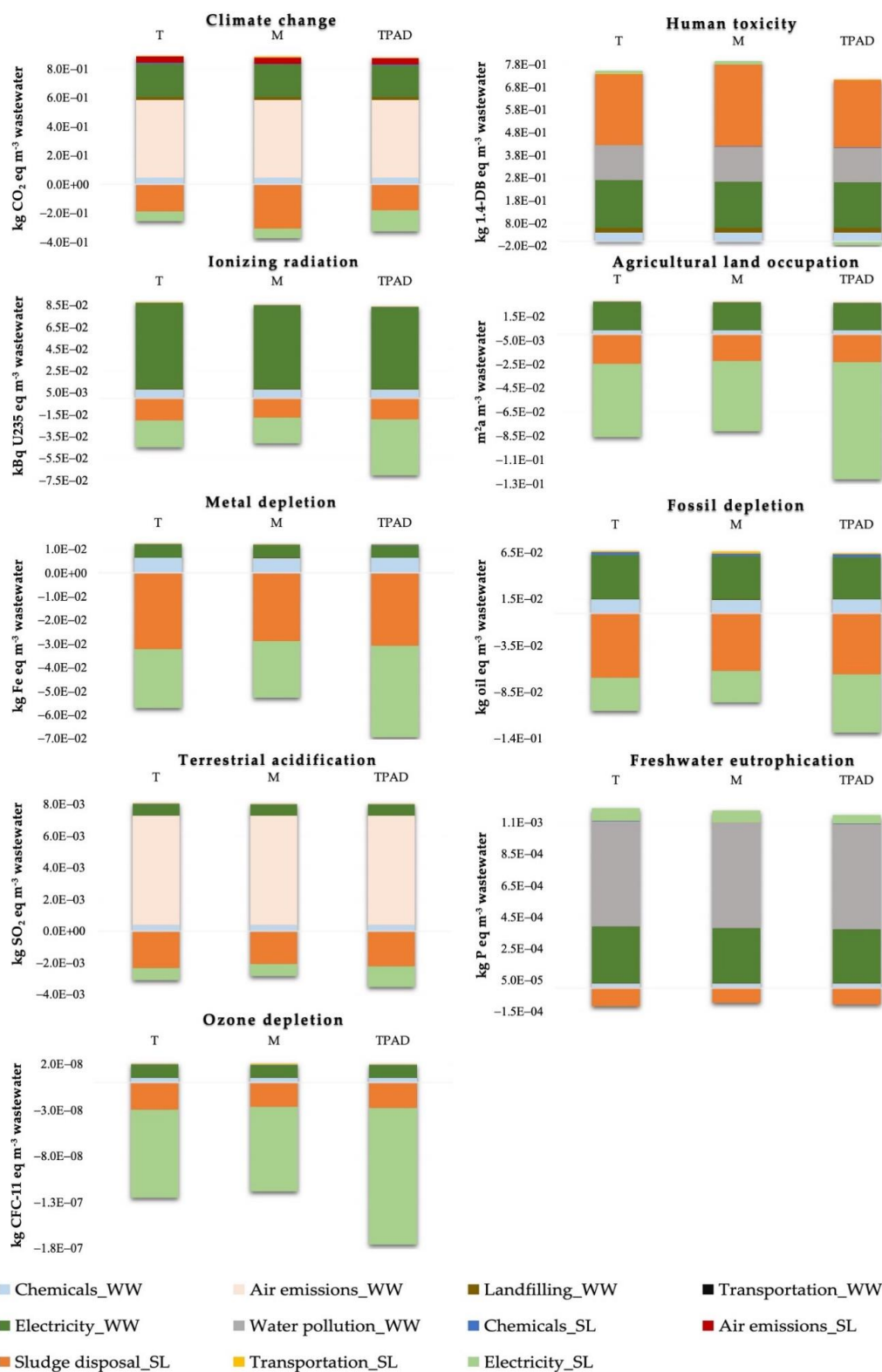
Figure 2. Potential environmental impacts for the three scenarios of the whole WWTP (WWTP-LCA): Mesophilic (M), Thermophilic (T), and Temperature-Phased Anaerobic Digestion (TPAD). WWTP: wastewater treatment plant; WW: wastewater line; SL: sludge line. Results shown for the FU: 1 m^3 of treated wastewater.

In the case of Climate change, the biggest impacts are caused by the wastewater treatment line (Figure 2). Conversely, the sludge line decreases the Climate change impacts up to 38% for M, around 35% for TPAD, and 24% for T. Climate change is related to nonrenewable energy consumption, which is especially high in the biological reactor of activated sludge WWTP, accounting for more than 50% of the total energy consumption—Table 2 and [46]. The positive influence of sludge line mainly comes from AD which supplies with the fertilizer obtained after WAS is digested (Sludge disposal_SL) that substituted the industrial production of the fertilizer with its harmful effect through Climate change. Additionally, AD generates renewable energy out of the biogas produced as a result of the organic matter biodegradation, and counterbalances nonrenewable energy consumption that would otherwise be required to fuel the process. The highest avoided impacts on Climate change are obtained with the mesophilic digestion (M) which is 40% and 42% better than T and TPAD, respectively. These avoided impacts occur due to a type of digestate disposal (which is composting and consequent agricultural land application for M, agricultural land application alone—for T and TPAD) and to its larger amount in comparison to T and TPAD (Table 1). In terms of the energy balance, TPAD is more beneficial than T and M by more than 50% within the sludge line. The lowest total avoided impact on Climate change is obtained with the thermophilic digestion, as a consequence of the energy balance of the process, i.e., energy produced vs. energy consumed by each AD system—Figure 3.

The results of WWTP-LCA regarding all constituents are depicted in Figure 3.

With regard to the factors that contributed the most to the environmental impact of Climate change, those emissions to the air (Air emissions_WW) and from the energy demand (Electricity_WW) in the wastewater treatment line are the most significant ones (i.e., around 60% and 25%, respectively, of all contributors from the wastewater treatment line). The contribution from the emissions to the air in the sludge line (Air emissions_SL) are only 5% (Figure 3). Hence, the total environmental impact was partly compensated by land application as the final sludge disposal (Sludge disposal_SL) and energy produced from the methane (Electricity_SL) obtained during AD with the following percentage of these two factor contributions, respectively: 72% and 28% for T, 82% and 18% for M, and 54% and 46% for TPAD. The balance of these two factors for TPAD shows better long-term performance of this AD technology.

For Human toxicity (Figure 2) the wastewater line constituents are quite similar in all scenarios, however, the absolute value of the sludge line varies: the larger negative effect to the environment is for M, 0.377 kg 1.4-DB eq, and the smaller one is for TPAD, 0.282 kg 1.4-DB eq, which is almost 25% less than that of M, and 15% less than T. This happens due to the higher total amount of digestate produced at M conditions rather than at T or TPAD. In particular for the Human toxicity category, less than 3% of the avoided environmental impacts are given by the energy production at TPAD conditions. The main contributor to this impact category is land application (Sludge disposal_SL) due to the heavy metals and other toxic substances that are still present in the digestate (41–45%)—Figure 3. The other major contributors are the energy consumption in the wastewater treatment line (26–29%), followed by the water body pollution (Water pollution_WW) made by treated wastewater discharge (19–22%) and, finally, by the different chemicals' consumption (Chemicals_WW) used at different stages of the wastewater treatment processes such as phosphorus precipitation and coagulation (all around 5%).

Figure 3. Contribution analysis of the potential environmental impacts for the three scenarios of both wastewater and sludge lines (WWTP-LCA): Mesophilic (M), Thermophilic (T), and Temperature-Phased Anaerobic Digestion (TPAD). Results shown for the FU: 1 m^3 of wastewater treated.

In terms of the Ionizing radiation impact category, even though the absolute values for both lines are lower than ±0.1 kBq U235 eq/m^3 of treated wastewater, the avoided environmental impacts given

by the sludge line of WWTP compensates the negative influence of wastewater treatment line for more than 40% for both T and M, and around 90% for TPAD (Figure 2). The latter leads to a better balance of both avoided and overall environmental impacts for TPAD. The rest of the contributions are given by different chemicals' consumption (Chemicals_WW) used for wastewater treatment processes such as phosphorus precipitation and coagulation (all less than 9%)—Figure 3. The factors that represent the avoided environmental impact are land application as the final sludge disposal (Sludge disposal_SL) and the energy production (Electricity_SL), both are from the sludge line of WWTP. The percentage contributions of them, respectively, are 45% and 55% for T, 43% and 57% for M, 27% and 73% for TPAD. For TPAD, the distribution is significantly different from T and M due to the better energy balance after AD.

In the context of other impact categories such as Agricultural land occupation, and Metal and Fossil depletion, the avoided impacts of the sludge line made mainly by land application as the final sludge disposal (Sludge disposal_SL) and energy production (Electricity_SL) completely captures the negative influence of wastewater treatment line given by energy consumption (Electricity_WW) and chemicals' consumption (Chemicals_WW)—Figure 3.

The environmental impact represented through the rest of the assessed impact categories show relatively low absolute values: <0.0052 kg SO_2 eq/m^3 of treated wastewater for Terrestrial acidification, <0.0011 kg P eq/m^3 of treated wastewater for Freshwater acidification, and $<-1.0 \times 10^{-7}$ kg CFC-11 eq/m^3 of treated wastewater for Ozone depletion.

Terrestrial acidification impacts are built up due to the gaseous emissions (Air emissions_WW) from the wastewater treatment line, $<10\%$ from energy demand (Electricity), and $<5\%$ from chemicals (Chemicals_WW) used at wastewater treatment line—Figure 3.

Freshwater eutrophication is mostly affected by water body pollution (Water pollution_WW) with a 45–50% contribution—Figure 3—and by energy consumption (Electricity_WW) with a 25% contribution from the wastewater treatment line.

Ozone depletion results are driven by the avoided environmental impacts of both the energy produced (Electricity_SL)—around 60% for T and M, and more than 65% for TPAD; and the land application (Sludge disposal_SL)—around 20% for T and M, and around 15% for TPAD (see Figure 3). These avoided impacts are significantly bigger than those caused by Electricity_WW and Chemicals_WW consumption.

Concerning the factors mainly contributing to the different environmental impact categories negatively, there are certain ones confirming their prevailing parts in the total environmental burden. In the case of WWTP-LCA, the major contributors are the gaseous emissions from the open biological step reservoirs to the air, the energy consumption for aeration tanks [47], and the water body secondary pollution given by treated wastewater discharge—see Figure 3. All of them are related to the wastewater treatment line.

For the Climate change impact category, both the gaseous emissions to the air and the energy consumption—again related to the wastewater treatment line—are the biggest contributors to the environmental burden, followed by Chemicals consumption—related to the wastewater treatment line—and the gaseous emissions to air—from the sludge line—with around 10–15% all together.

3.1.2. SL-LCA with Mesophilic, Thermophilic, or Temperature-Phased Anaerobic Digestion

The LCA results of the sludge line (SL-LCA) including methane production are presented in Figure 4 using the second FU: 1 m^3 of methane produced (unlike Figures 2 and 3 which use the FU: 1 m^3 of wastewater treated). Figure 4 includes all environmental impact categories considered in this study, and within each impact category there are three scenarios: mesophilic, thermophilic, and TPAD. Furthermore, the different contributions from all process inputs and outputs are included for each of the three scenarios and for all categories. Positive values represent the environmental impacts, while negative values refer to the avoided environmental impacts (here considered as environmental credits).
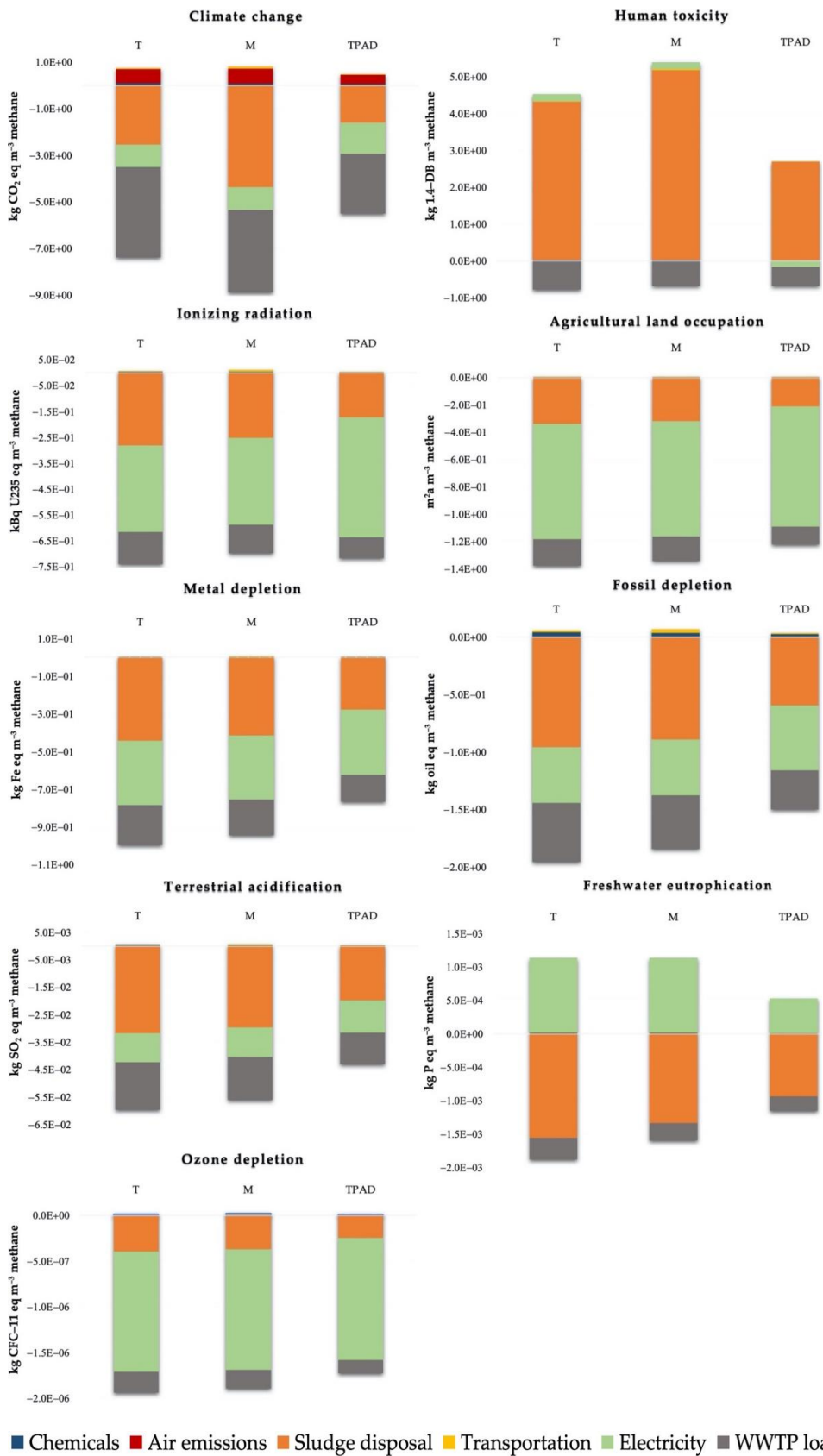
Figure 4. Contribution analysis of the potential environmental impacts for the three scenarios of the sludge line (SL-LCA): Mesophilic (M), Thermophilic (T), and Temperature-Phased Anaerobic Digestion (TPAD). Results shown for the FU: 1 m^3 of methane produced.

The aggregated SL-LCA results for the three scenarios lead to overall avoided environmental impacts in all categories with exception of Human toxicity. From the three scenarios, T outperforms M and TPAD in seven out of the nine impact categories here analyzed (except for Climate change and Human toxicity) (Figure 4). M is consistently the second best scenario in six out of the nine categories, except for Climate change (where it performs the best), and both Ionizing radiation and Human health (where it performs worse). Finally, TPAD has the lowest environmental impacts for Human health (by over 50%), while it also has the least avoided environmental impacts for seven categories out of the nine here analyzed (Figure 4).

In all SL-LCA scenarios, the contributing factors with the largest absolute values (i.e., either potential impacts (for Human toxicity) or avoided impacts (for all the other categories)) are the final sludge disposal (starting from 15% for Ozone depletion to almost 80% for Human toxicity), energy balance (from 12% for Climate change to over 75% for Ozone depletion), and water pollution (from 11% for Human toxicity and Ionizing radiation to 22% for Fossil depletion). It is also important to highlight that the factor of gaseous emissions to the air contributes significantly in a harmful way only for Climate change (more than 80% of caused environmental impact and only less than 8% of total environmental impact) due to the digestate accumulated at the landfill [26].

In the case of specific impact categories, the avoided environmental impacts in Climate change for both T and M are larger than those of TPAD by 24% and 38%, respectively—Figure 4. The only factor causing environmental impacts on Climate change for the three scenarios are the gaseous emissions from AD installations (Air emissions). On the contrary, the avoided environmental impacts have been credited by the following factors: additional reject water treatment (WWTP load concerning each scenario: 53% for T, 40% for M, and 47% for TPAD), final sludge disposal (Sludge disposal: 34% for T, 49% for M, and 29% for TPAD) and energy production (Electricity: 13% for T, 11% for M, and 24% for TPAD). Further minor avoided impacts are related to chemical consumption (Chemicals) (around 10% for all AD types) and transportation (Transportation) (around 5% for T and TPAD, and around 11% for M due to longer distance—a round trip—to the composting site).

In terms of Human toxicity, TPAD demonstrates the best results with the lowest environmental impacts at SL-LCA (Figure 4). The TPAD's impacts on Human toxicity are 46% lower than T, and 58% lower than M. The most substantial contribution to Human toxicity is coming from the final sludge disposal (Sludge disposal—95% for T, 96% for M, and 99% for TPAD) which makes sense as it is the agricultural land application for the T and TPAD scenarios and agricultural land application via composting for M scenario—Figure 4 and Table 3. The rest of the impacts on Human toxicity are mostly caused by the energy consumption (Electricity) with 4% and 3%, for T and M, respectively. While for TPAD, the energy balance is slightly positive, meaning that the system produces surplus energy with respect to its total consumption which leads to avoided impacts by almost 6%. Hence, the TPAD scenario for SL can be considered as energy self-sufficient process and an electricity supplier. Furthermore, for Human toxicity there are some minor avoided impacts from additional reject water treatment (WWTP load—100% for T and M and 94% for TPAD) which is highly polluted, meaning that it can be used as an additional source for resource recovery [15,48].

Interestingly, Human toxicity is the only impact category that does not result in overall avoided impacts at the sludge line. This happens due to the sufficient amounts of heavy metals and other toxic pollutants that are not completely removed during AD operation. Knowing that, T's and TPAD's digestates are considered to be pathogenically safe, and their final disposal can be a direct land application as fertilizers [35]. M digestate undergoes an additional step of composting prior to its application in agriculture. However, the gaseous emissions as well as the traces of heavy metals (Table 3) result in certain danger to the human health [9].

Looking at the Freshwater eutrophication impact category, the TPAD scenario shows both the lowest environmental impacts (50% lower than T and M, with energy consumption—Electricity—as the main contributor) and the lowest avoided environmental impacts. In the latter case, the prevailing

contributors are digestate usage for the agricultural land application (Sludge disposal) and water body pollution reduction (WWTP load).

The rest of the impact categories (i.e., Ionizing radiation, Agricultural land occupation, Metal and Fossil depletion, Terrestrial acidification, Freshwater eutrophication, and Ozone depletion) follow a similar pattern. For all SL-LCA scenarios, the overall result can be referred as avoided impacts, with the best results being obtained for T, followed by M, and finally by TPAD. The main contributors are Sludge disposal and Electricity, and the WWTP load to a lower extent (with a maximum of 20% for Terrestrial acidification and lesser for other impact categories).

3.2. Sensitivity Analysis

The sensitivity response (i.e., the sensitivity coefficient "S" as described in Section 2.2.4) of all studied environmental impact categories was analyzed with respect to the assumed values for the digestate volume (with ±5% of the baseline value for the TPAD scenario, i.e., 90,332 t/year > 85,816 t/year > 81,299 t/year). Only the TPAD scenario was considered for sensitivity analysis due to the variability of the experimental data obtained by the previous studies [19,26,35], and shortage of the reported data from full-scale WWTPs, especially considering that TPAD is the least spread AD system worldwide among others [43].

The sensitivity coefficients were analyzed considering the processing conditions of TPAD for both the WWTP and the SL alone as shown in Table 4.

Table 4. Sensitivity coefficients (S) and environmental impacts of the whole WWTP (WWTP-LCA) and SL (SL-LCA) with respect to the TPAD baseline value assumed for the digestate volume.

Case	WWTP			SL		
Impact Category	S Coefficient	+5%	−5%	S Coefficient	+5%	−5%
Climate change, kg CO_2 eq/FU	−0.309	0.543	0.561	0.321	−5.108	−4.938
Human toxicity, kg 1.4-DB eq/FU	0.431	0.711	0.679	**1.325**	**2.162**	**1.880**
Ionising radiation, kBq U235 eq/FU	−1.424	0.012	0.014	0.241	−0.722	−0.704
Agricultural land occupation, m^2a/FU	0.245	−0.096	−0.093	0.173	−1.226	−1.204
Metal depletion, kg Fe eq/FU	0.530	−0.059	−0.056	0.363	−0.777	−0.748
Fossil depletion, kg oil eq/FU	0.980	−0.068	−0.061	0.415	−1.495	−1.431
Terrestrial acidification, kg SO_2 eq/FU	−0.478	4.41×10^{-3}	4.64×10^{-3}	0.467	−0.044	−0.042
Freshwater eutrophication, kg 1.4-DB eq/FU	−0.103	$−9.92 \times 10^{-4}$	$−1.0 \times 10^{-3}$	**1.504**	$\mathbf{−6.76 \times 10^{-4}}$	$\mathbf{−5.77 \times 10^{-4}}$
Ozone depletion, kg CFC-11 eq/FU	0.172	$−1.57 \times 10^{-7}$	$−1.54 \times 10^{-7}$	0.143	$−1.73 \times 10^{-6}$	$−1.71 \times 10^{-6}$

Note: Sensitivity coefficients (S) are unitless; Units of each environmental impact category consider the specific FU for WWTP and SL, i.e., 1 m^3 of treated wastewater and 1 m^3 of produced methane, respectively. Bold numbers are for the most sensitive impact categories (with S > 1.0).

A positive value of the sensitivity coefficient (S) refers to a straight influence of the studied parameter on the environmental results: e.g., the more sludge that is considered, the higher the (avoided) environmental impacts are. On the contrary, a negative sensitivity coefficient means an opposite influence of the studied parameter on the environmental results, it is e.g., the more sludge that is considered, the less the (avoided) environmental impacts are.

In this study, negative sensitivity coefficients are obtained only for WWTP-LCA, concerning four impact categories: Climate change, Ionizing radiation, Terrestrial acidification, and Freshwater eutrophication.

In a case of the potential environmental impacts related to the Climate change, Ionizing radiation, and Terrestrial acidification (Table 4), this behavior occurs due to an increased (proportional to the digestate volume) amount of both digestate as fertilizer substituent and energy recovered as biogas. In the case of Freshwater eutrophication, this opposite behavior occurs since an increase in the digestate volume leads to an additional amount of highly polluted reject water (that in turn needs to be further treated) generating a minor amount additional environmental impacts but that overall reduces the total avoided impacts.

On the contrary, the sensitivity coefficients for SL-LCA are positive values in all impact categories indicating a positive relation between the input variable (i.e., the assumed digestate volume) and the

out variable (each environmental impact category). In this case, larger digestate volumes lead to the larger (either potentially caused or avoided) environmental impacts. In particular for the Human toxicity category, impacts are higher with the increase in the digestate volume (due to the proportional increase in the present pollutants in the digestate), while all the other categories included in Table 4 result in larger avoided environmental impacts with the increment in the digestate volume (due to the production of the avoided products such as fertilizers and electricity).

The most sensitive environmental impact categories in terms of ±5% variation in the digestate amount are, at SL-LCA, Human toxicity and Freshwater eutrophication, as S is positive and higher than 1.0.

The digestate amount variation of +5% increases the environmental burden of Human toxicity by 6.5% from the baseline value at SL-LCA. At WWTP-LCA, the digestate amount of ±5% had the influence of around 2.0% referring to the baseline value. It is also important to mention that the Human toxicity impact category is the only one with positive sensitivity coefficients at both LCAs, and for SL-LCA the sensitivity coefficient is higher than 1.0. Hence, it is important to mention that such sensitivity behavior of the Human toxicity category reveals that the major environmental concern based the variability of the digestate amounts would be on this impact category.

For Freshwater eutrophication at SL-LCA, the avoided environmental impacts increase by over 7.0% along with the increment in the digestate amount applied in agriculture as a fertilizer. At WWTP-LCA, the sensitivity coefficient at this impact category is negative and lower than 1.0, and can be neglected.

At the WWTP-LCA, the rest of the impact categories (i.e., apart from the ones with negative sensitivity coefficient) result in S values lower than 1.0. The sensitivity coefficient values higher than 0.5 are for the impact categories Metal and Fossil depletion. These two impact categories are affected by 2.5% to 5%, respectively (Table 4), and they refer to overall avoided environmental impacts. Furthermore, the sensitivity coefficients for Metal and Fossil depletion at WWTP-LCA are higher than those at SL-LCA. A reason for such a difference is the contribution in the energy balance (Electricity_WW) at WW line (Tables 3 and 4). The absolute values of avoided environmental impacts at ±5% of digestate are essentially higher at SL-LCA than at WWTP-LCA for Metal depletion (on over 92%) and for Fossil depletion (on over 95%) due to the energy consumption at the WW line concerning both impact categories.

In general terms, it can be said that the WW line has a higher harmful effect on the environment than SL line itself, and the larger its scale is, the larger the potential environmental impacts will be, contrary to the SL line.

Other general trends from the sensitivity analysis are that the sensitivity gives a clear overview that AD, namely TPAD, affects the environment mainly due to the toxic substances' content and air emissions derived from the digestate, which are proportional to its volume. The digestate production affects the environment negatively by the contribution to Human toxicity due to the final sludge disposal (Sludge disposal) coming from the SL line which relates to both WWTP-LCA and SL-LCA (Figures 3 and 4). At the same time, digestate production has also a positive effect given by resource (fertilizer) and energy (electricity and heat) recovery (Sludge disposal and Electricity, respectively) and also due to the additional reject water treatment (WWTP load) derived from the SL line (Figure 4).

Therefore, the impact categories of Human toxicity, Metal and Fossil depletion which are directly related to the produced digestate amount are of major attention for these types of processes. Considering the case of the TPAD scenario, it can also be said that ±5% of digestate production does not affect most of the (avoided) environmental impacts. Only three environmental impact categories have S > 1.0, namely: Human toxicity (SL-LCA), Ionizing radiation (WWTP-LCA), and Freshwater eutrophication (SL-LCA). These S values, bigger than 1.0, are strongly related to several contributing factors such as energy consumption (WWTP-LCA), final sludge disposal, and reject water treatment (SL-LCA)—Figures 3 and 4, and Table 4. Hence, these findings of the sensitivity analysis should be considered and taken into account for future designs of WWTPs and AD systems.

Based on the sensitivity analysis performed, it can be said that the main factor that contributes to the environmental impact through Human toxicity impact category is digestate quality (pathogenic safety, presence of the toxic substances, and gaseous emissions) and its amount. Therefore, by considering and changing the final digestate disposal, the total environmental impact can be reduced.

4. Conclusions

In this study, a comparative LCA analysis was carried out to evaluate the environmental impacts of three alternative AD processes (mesophilic, thermophilic, and TPAD) used for sludge treatment in activated sludge WWTP. The environmental burden was evaluated at two scales, namely the whole WWTP (to assess the contribution of the sludge line to the whole WWTP)—with a FU of 1 m^3 treated wastewater—and the sludge line alone (to highlight the potential environmental benefits from methane production as an additional function of the system beyond the wastewater treatment)—with a FU of 1 m^3 produced methane.

In the WWTP-LCA, five (Climate change, Human toxicity, Ionizing radiation, Terrestrial acidification, and Freshwater eutrophication) out of the nine environmental impact categories analyzed showed potential environmental impacts. The rest of the environmental impact categories (Agricultural land occupation, Metal and Fossil depletion, Ozone depletion) showed avoided environmental impacts, since the WW line led to potential environmental impacts in all impact categories, while the SL line led to avoided environmental impacts for most environmental impact categories (except for Human toxicity). Among all scenarios, the WWTP with TPAD outperformed those with mesophilic and thermophilic AD in all the environmental impact categories, besides Climate change.

The SL-LCA showed mostly avoided impacts, being the highest for thermophilic AD, followed by mesophilic AD and TPAD, except for Climate change where mesophilic AD was the most beneficial. The only potential environmental impact was Human toxicity, being the lowest for TPAD.

Differences between both LCA results may be attributed to the FU.

In addition, it can be also concluded that such products as nutrients and energy recovered from the AD systems and incorporated into the sludge treatment create an amount of credits that make the whole WWTP more environmentally friendly.

Abbreviations

AD	anaerobic digestion
COD	chemical oxygen demand
HRT	hydraulic retention time
LCA	life cycle assessment
M	mesophilic
OLR	organic loading rate
SL	sludge treatment line
SL-LCA	LCA performed within the boundaries of the sludge line
T	thermophilic
TPAD	temperature-phased anaerobic digestion
TS	total solids
V	reactor working volume
VS	volatile solids
WAS	waste activated sludge
WW	wastewater treatment line
WWTP	wastewater treatment plant
WWTP-LCA	LCA performed within the boundaries of the whole wastewater treatment plant

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