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New findings on the anaerobic co-digestion of thermally pretreated sludge and food waste: laboratory and pilot-scale studies

Andrés Donoso-Bravo IWA [a,b,*], Javiera Toledo-Alarcón[a,c], Valentina Ortega[a], Valeria Barría[a], Yves Lesty[d], Jordi Fontana[e] and Hugues Vanden Bossche [e]

[a] Cetaqua, Centro Tecnológico del Agua, Los Pozos 7340 Santiago, Chile
[b] Departamento de Ingeniería Química y Ambiental, Universidad Técnica Federico Santa María, Avenida Vicuña, Mackenna 3939 Santiago, Chile
[c] Escuela de Ingeniería Bioquímica, Pontificia Universidad Católica de Valparaíso. Av. Brasil 2085 Valparaíso, Chile
[d] Aguas Andinas, Gerencia Economía Circular, Presidente Balmaceda 1398 Santiago, Chile
[e] EDAM Limitada, Alonso de Córdova 5151 Oficina 901, Santiago, Chile
*Corresponding author. E-mail: andreseduardo.donoso@cetaqua.com

AD-B, 0000-0002-7246-4416; HVB, 0000-0002-2176-1322

ABSTRACT

Co-digestion of thermally pretreated sewage sludge with food waste is an innovative strategy that could improve the balance and availability of nutrients needed to increase the efficiency of anaerobic digestion in terms of biogas production. In this context, the aim of this research was to evaluate the impact of different proportions of sewage sludge/food waste in laboratory- and pilot-scale reactors. Special focus was placed on the impact of the variability of food waste composition on the behaviour of the pilot digester. Our results show that by adding 40% of co-substrate, a higher biogas production was possible during laboratory operation. Interestingly, using a co-substrate of variable composition had no negative impact on the reactor's stability at pilot-scale, promoting an increase in biogas production through a more efficient use of organic matter. In both the lab and pilot experiences there was an impact on the amount of nitrogen in the digestate compared to digester operating in monodigestion. This impact is more significant as the proportion of co-substrate rises. Overall, our results show that co-digestion of thermally pretreated sewage sludge with food waste allows better management of food waste, especially when their composition is variable.

Key words: anaerobic co-digestion, food waste, pilot-scale, sewage sludge, thermal treatment

HIGHLIGHTS

- Thermally pretreated sewage sludge is being co-digested with industrial waste.
- At laboratory-scale, the highest biogas yield was obtained using 40% FW as co-substrate.
- ACoD with FPW of variable composition has no negative impact on digester stability.
- ACoD with FPW led to more biogas and a more efficient use of the organic matter.

1. INTRODUCTION

Wastewater treatment involves the simultaneous production of a high quantity of sewage sludge (SS), which must be properly managed and stabilized to minimize any impact on the environment. Anaerobic digestion (AD) has proved to be the most appropriate alternative, since at the same time the SS is revalued through the energy generation (Han *et al.* 2017; Al bkoor Alrawashdeh 2019). Thermal hydrolysis process (THP) is the most widely used pre-treatment in the wastewater treatment plants prior AD. This pre-treatment disintegrates the sludge and solubilizes the complex molecules, making them more accessible for microbial degradation. Furthermore, the viscosity decreases and dewatering increases, which allows us to work with higher organic loads without losing efficiency. In addition, the level of pathogens in the digestate is reduced, so further treatment could be unnecessary (Han *et al.* 2017; Farhat *et al.* 2018; Westerholm *et al.* 2019).

To maximize biogas production, anaerobic co-digestion (ACoD) of two or more wastes has been widely studied. The successful application of ACoD in wastewater treatment plants has great potential, as the necessary infrastructure is already in place for SS digestion (Labatut *et al.* 2011). In this context, food waste (FW) represents the main co-substrate used for resource recovery from wastewater, through its ACoD with SS (Nghiem *et al.* 2017; Tyagi *et al.* 2018; Xu *et al.* 2018;

Donoso-Bravo *et al.* 2020). Another important source of FW is the food processing industry, which can vary depending on the food products, so specific processing conditions must be considered for proper and efficient waste management. The literature shows that ACoD of SS and FW is efficient in increasing biogas production. For instance, during a batch operation, Yun *et al.* (2015) tested the ACoD of FW and waste activated sludge (WAS), resulting in an increase in methane yield and productivity by 50 and 100%, respectively. In a semi-continuous operation, Heo *et al.* (2003) reported that methane percentage in biogas and methane production rate depended on the FW:WAS ratio and hydraulic retention time (HRT) for which the optimum was using a 50:50 (FW:WAS) ratio and HRT of 10 day. Kim *et al.* (2017) assessed the FW:SS feed ratio in five laboratory-scale reactors, reporting that the addition of FW increased biogas production and chemical oxygen demand (COD) removal, while the ammonia concentration in the digestate remained low. In a pilot-scale study, Cabbai *et al.* (2016) reported that as they increased the organic load rate (OLR), up to 3.2 $kg_{VS}\,m^{-3}d^{-1}$, the amount of specific and total biogas accumulated also increased, without significant accumulation of inhibitors, such as VFAs.

With respect to ACoD using thermally pretreated SS, limited reporting has been done. For instance, Aichinger *et al.* (2015) tested ACoD using a pretreated SS with THP and an organic co-substrate, showing an increase of 35% in biogas production rate compared to the control. Donoso-Bravo *et al.* (2019b) evaluated the ACoD during a semi-continuous operation of thermally pretreated mixed sludge with beverage wastewater at laboratory-scale, obtaining better results compared with the control reactor. This study aimed to evaluate the ACoD of thermally pretreated sewage sludge and FW as co-substrate. Different substrate/co-substrate ratios were evaluated at laboratory and pilot scale. Also, in this last one, the robustness and stability of the ACoD was evaluated through the variation of the co-substrate composition during the operation time. In addition, further analysis of the digestate was included to evaluate the efficiency of organic matter use and the impact of ACoD in the nitrogen presence.

2. MATERIALS AND METHODS

2.1. Substrate, co-substrate and inoculum

For the laboratory- and pilot-scale evaluation of ACoD, thermally pretreated secondary sludge (hydrolyzed in a full-scale CAMBI® THP, at 6 bars and 170 °C for 30 min) mixed with primary sludge, from the wastewater resource recovery factory (WRRF) Mapocho-Trebal in Santiago – Chile, was used as substrate (called HMS hereafter). The substrate was taken daily from the WRRF and the composition was of 63–69% of primary sludge and 31–37% of thermally pretreated secondary sludge. As co-substrate, FW from the WRRF cafeteria and processed food waste from a food industry were used in laboratory- and pilot-scale experiments, respectively. Table 1 specifies the physical–chemical characteristics of the substrates and co-substrates used in this study. The FW was collected, separated and weighed by the cafeteria staff and delivered to the laboratory each week. FW was mixed after classification into four categories, as follows: (1) 20% protein (*e.g.* meat, fish, eggs), (2) 55% vegetables (*e.g.* greens, fruit, pulses) (3) 20% processed (*e.g.* rice, bread, mush potatoes, pasta and others) and (4) 5% paper (*e.g.* napkin, paper towel, packaging). The mixture was diluted by 10% w/w water and then milled in a laboratory-scale knife mill Waring model HGB250 15 at 24,500 rpm, before being fed into the reactor. FW from food processing industry (FPW) was obtained weekly and was stored in a high-density polyethylene tank. For both experiments, inoculum from a full-scale biodigester treating HMS was used.

2.2. Analytical methods

For laboratory-scale experimentation, the pH, total solid (TS) and volatile solid (VS) value were determined daily. While the total and soluble COD (TCOD and SCOD), total alkalinity (TAC), volatile fatty acids (VFA) and total ammonium nitrogen (TAN) were determined once a week. For the pilot-scale experimentation, the FW was characterized weekly, determining the value of pH, ST, SV, TCOD, SCOD, TAC, VFA and TAN. While the HMS, the digestate and the feeding tank of each pilot digester was characterized twice a week. The analytical methodology used is specified next: TS were measured by gravimetric method and drying at 103–105 °C (method 2540 B, Standard Methods) and VS were measured by gravimetric method and incineration at 550 °C (method 2540 E, Standard Methods). The TCOD and SCOD was determined with commercial vials and HACH photometer, by the reactor digestion method. VFA and TAC were measured by boiling and titration with NaOH and HCl 0.1 N. The pH was determined with a pH meter (method 4500-H^+, Standard Methods) and TAN were measured by selective electrodes (method 4500-NH_3, Standard Methods). The content of TKN was measured through method 4500-N, Standard Methods). The total phosphorus was determined with the vanadomolybdophosphoric acid method (NCh2313/15, Chilean Regulations). The solid and liquid phase of the digestates of the pilot digesters, after the

Table 1 | Characterization of the substrate and co-substrate used in this study

Laboratory scale							Physicochemical parameters								
							TS (gL^{-1})	VS (gL^{-1})	TCOD (gL^{-1})	SCOD (gL^{-1})	TAN (mgL^{-1})	VFA (gL^{-1})	pH	TAC (mgL^{-1})	TKN (mgL^{-1})
Co-substrate FW (averaged values ±SD)[a]							215 ± 40	204 ± 39	197 ± 56	n.d	122 ± 74	2.8 ± 1.0	5.0 ± 0.6	925 ± 239	155
Substrate HMS (averaged values ±SD)[a]							53 ± 7	41 ± 6	95 ± 20	9 ± 4	595 ± 224	2.3 ± 0.7	5.8 ± 0.1	2,094 ± 579	4,000
Pilot scale							Physicochemical parameters								
Co-substrate FPW															
Week	M[b]	Y[b]	Ju[b]	Bu[b]	Je[b]	W[b]									
1	100	0	0	0	0	0	50	43	78	42	35	4.5	3.7	n.d	n.d
2	100	0	0	0	0	0	35	30	n.d	n.d	35	3.8	4.1	400	n.d
3	40	40	15	5	0	0	80	76	156	36	50	2.6	4.2	300	n.d
4	70	20	5	0	5	0	75	68	119	43	49	3.2	4.7	533	n.d
5	70	25	2.5	0	0	2.5	115	108	224	63	107	3.2	5.9	2,433	n.d
6	65	20	15	0	0	0	104	98	186	53	71	3.2	5.4	1,200	n.d
7	80	15	5	0	0	0	84	79	144	41	11	1.8	5.0	933	n.d
8	60	25	15	0	0	0	85	80	155	22	n.d	3.6	3.7	n.d	n.d
Substrate HMS (averaged values ±SD)[c]							49 ± 6	39 ± 5	81 ± 16	8 ± 3	0.68 ± 0.31	2.4 ± 1.1	5.9 ± 0.2	n.d	n.d

[a]Estimated from the semi-continuous operation data.
[b]% volume.
[c]Estimated from the continuous operation data – M: Milk, Y: Yogurt, Ju: Juice, Bu: Butter, Je: Jelly, W: Water – TS, Total Solids; VS, Volatile Solids; TCOD, Total Chemical Oxygen Demand; SCOD, Soluble Chemical Oxygen Demand; TAN. Total Ammonium Nitrogen; VFA, Volatile Fatty Acids; TAC, Total Alkalinity; TKN, Total Kjeldahl Nitrogen; n.d, not determined.

co-digestion period, was characterized by determining TS, VS, TKN, total phosphorus, TAC, VFA, TCOD, SCOD and lipids (method 5520, Standard Methods).

2.3. Laboratory-scale ACoD studies

2.3.1. Batch test

Biochemical Methane Potential (BHP) batch test were performed in 120 mL glass bottles with a 100 mL working volume. A substrate/inoculum (S/I) ratio of 0.5 $g_{VS}g_{VS}^{-1}$ and an inoculum concentration of 10 g_{VS} L^{-1} were established. Sodium bicarbonate was added at a concentration of 0.5 $gNaHCO_3$ gVS^{-1} to ensure neutral pH values during the assay. Blank trials with only inoculum were used to quantify the amount of biogas produced by endogenous respiration. Each test BMP was performed in triplicate. The temperature of the assay was 35 °C, and the bottles were stirred at 40 rpm in a thermoregulated shaker system. The biogas production was measured using a plug syringe. The net biogas yield was obtained by subtracting the endogenous production of the blank bottle and converted to normal conditions of temperature (0 °C) and pressure (1 atm). The substrate/co-substrate proportions assessed in the batch test were as follow: 100/0, 90/10, 80/20, 60/40, 40/60, 20/80, 0/100 (in VS basis). A summary of the experimental design studied in this article is shown in the Figure 1.

2.3.2. Semi-continuous operation

Six laboratory-scale digesters (BRS, BioReactor Simulator, Bioprocess control®) with a 1.6 L working volume were set up in a thermoregulated bath at 35 °C with mechanical stirring. The digesters were operated in semi-continuous mode for 162 days, fed once a day manually. The biogas flow was continuously monitored by a liquid displacement/floatability method and the methane content was measured once a week offline by NaOH solution (40 g L^{-1}) displacement. During the Mono-AD stage (day 1 to 52), all digesters were fed equally with a stepwise increase in the OLR of 1.0, 1.5 and 2.0 kg_{VS} m^3d^{-1}. The ACoD period started on day 53 and the inlet conditions, which were chosen considering potential scenarios that can take place at full scale, were as follows (HMS/FW): 100/0, 90/10, 80/20, 60/40, 40/60 and 20/80 for R1, R2, R3, R4, R5 and R6,

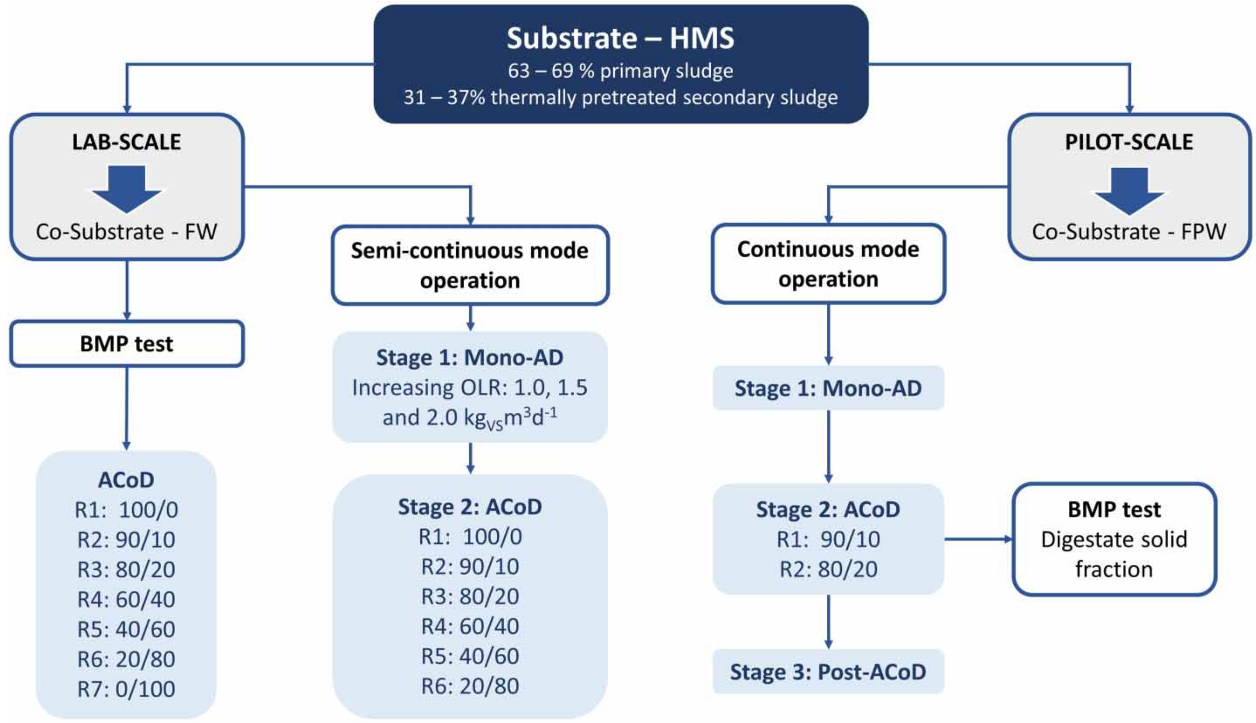

Figure 1 | Summary of experimental design for laboratory- and pilot-scale experiments.

respectively. During the full ACoD period the OLR was maintained at 2.0 $kg_{VS}\ m^3d^{-1}$ in all six reactors, while HRT was 24 ± 2, 25 ± 2, 28 ± 2, 34 ± 2, 44 ± 3 and 61 ± 4 days for R1, R2, R3, R4, R5 and R6, respectively.

2.4. Pilot scale ACoD studies

2.4.1. Start-up of pilot-scale digesters

Two stainless steel pilot-scale digesters (2.6 m^3) operated at 35 °C and mixed by mechanical stirring were used. Biogas flow was continuously monitored by a drum-type gas meter (Ritter TG1). The substrate and the co-substrate were accumulated in a glass fibre storage tank which was refilled with fresh waste every 3 days. Additionally, each digester has a tank where the substrate and co-substrate are mixed (feed tank) and a tank of digested sludge (outlet of digesters). The substrate is pumped into the reactor and a same amount is pumped out of the reactor, both by centrifuge pumps. The amount of waste fed into the digester was daily estimated by the level change of the storage tank. The data of biogas flow, pH and level tanks and temperature were collected daily except the weekends. The biogas composition was estimated once a week by a portable gas analyzer Geotech (GA2000 model).

2.4.2. Digester's operation

The pilot digesters were operated in continuous mode approximately for 5 months. The operation was divided into three periods: Mono-AD fed with only HMS (0–42 days), ACoD fed with HMS and FPW (43–97 days) and post co-digestion (Post-ACoD) fed with only HMS (98–142 days). The digesters were operated with an OLR close to 2.5 $kg_{VS}\ m^{-3}\ d^{-1}$, similar to the one the full-scale digesters were being operated. During the ACoD period, the reactors were fed with FPW as a fraction of the OLR of 10 and 20% for digester A and B, respectively. These values were chosen as possible proportions that can be applied at full-scale. By the end of the operation of the digesters in ACoD mode (day 97), samples of digestate from both pilot digesters were taken for further analysis. The digestate fractions were separated in a solid and liquid fraction by centrifugation for 30 min at 13,500 rpm. The solid fraction of the digestate was used as substrate in a BMP test. Digestate from a full-scale digester treating mixed sewage sludge was used as inoculum and a substrate/inoculum ratio of 0.6 $g_{VS}\ g_{VSS}^{-1}$ was selected (VSS: volatile suspended solids).

2.5. Data analysis

The biogas production data from the BMP test were fitted with the Gompertz modified equation and processed as proposed in (Donoso-Bravo *et al.* 2019a):

$$B = P\,exp\left(-exp\left(\frac{R_m\,e}{P}(\lambda - t) + 1\right)\right) \quad (1)$$

where B: biogas production a time t (d), P: Maximum biogas production (ml g_{VS}^{-1}), R_m: Maximum biogas production rate or biogas productivity (ml g_{VS}^{-1} d^{-1}), and λ: Lag-phase time (d). To estimate the parameters, single least squares criteria were used as the minimization procedure. The estimated values of these parameters were defined as the observed values (OV) from the experiments.

For the data generated in continuous mode, a one-way analysis of variance (ANOVA) was performed, after checking normal data distribution, to evaluate significant differences in both biogas yield and productivity. Then, Tukey's test was performed as a post hoc analysis to find out which pairs of samples were statistically different. The analyses were carried out with PAST v4.03 software.

3. RESULTS AND DISCUSSION

3.1. Laboratory scale – BMP results

The biogas production curves using different substrate/co-substrate ratios can be found in the Supplementary material (Figure S1). The biogas yield increased significantly (ANOVA, F (7,16) = 7.087, $p = 0.0006$) with the FW proportion from 512.4 ± 5.5 ml·g_{SV}-s^{-1} (control – no FW) to 629.2 ± 67.1 ml·$_{gSV}s^{-1}$ (FW only) (Ara *et al.* 2015). While biogas productivity showed no statistically significant differences (ANOVA, F (6,14) = 2.40, $p = 0.08$) between the conditions studied (Table S1, Supplementary material). In addition, the methane content in the biogas decreased as the proportion of FW increased from 64.3 ± 3.3% (control – no FW) to 59.4 ± 3.9% (FW only). Therefore, the increase in total biogas production was offset by the reduction in the methane content of the biogas due to FW proportion. The parameters values obtained from the fitting of the biogas production curves with the modified Gompertz equation are shown in Table 2. Furthermore, the synergy and antagonism determination were added, which were estimated by the equation proposed and described by Donoso-Bravo *et al.* (2019a).

In general, the synergy attained on the P was negligible, which means that the obtained values are aligned with the expected ones associated to the substrate/co-substrate proportion. Regarding the Rm, a slight negative effect or antagonism was observed as the FW proportion increases, which can be explained by the fact that the soluble and more readily biodegradable material is less accessible, and it is found in a less proportion as the FW content increases. The solid content of FW is higher and more heterogeneous than the content for HMS. When Yun *et al.* (2015) used WAS as a substrate, an increase in methane yield and productivity of about 50 and 100%, respectively, was observed. This study shows a large difference with our results, probably because difference in the samples preparation (FW dilution). In addition, WAS has very

Table 2 | Parameters from the Gompertz equation fitting with the biogas production data for Mono-AD and ACoD BMP test with HMS (substrate) and FW (co-substrate)

		Biogas yield (*P*)			Maximum productivity (*Rm*)		
Substrate	**Co-substrate**	**EV**	**OV**	**Sinergy**	**EV**	**OV**	**Sinergy**
100%	0%	–	512	–	–	53	–
88%	12%	526	526	0%	53	53	0%
79%	21%	537	547	2%	53	52	−2%
59%	41%	560	567	1%	53	51	−4%
40%	60%	582	592	2%	53	50	−6%
20%	80%	606	626	3%	53	51	−4%
0%	100%	-	629	–	–	53	–

EV, Expected Value; OV, Observed Value. The standard deviation of the triplicates average was less than 10% of the average values in all cases.

different properties compared to HMS, especially in terms of soluble content and biodegradability, so the base scenario compared to WAS is different.

3.2. Laboratory scale – semi-continuous digesters performance

The biogas production and the digester characterization during the whole operation time is shown in Figure 2. According to the obtained results based on biogas production, the whole evaluation period was divided into three stages: Mono-AD (day 0–52), ACoD I (day 53–132) and ACoD II (day 133–162).

3.2.1. Mono-AD

During this period, the OLR was progressively increased simultaneously in all six digesters. As expected, biogas production was similar and there were no significant statistical differences between the digesters when an OLR of 1.0 (ANOVA, $F\ (5,108) = 0.047$, $p = 0.99$), 1.5 (ANOVA, $F\ (5,84) = 0.027$, $p = 0.99$), and 2.0 $kg_{VS}\ m^3 d^{-1}$ (ANOVA, $F\ (5,60) = 0.066$, $p = 0.99$) was applied, as observed in Figure 2. The biogas production was about 0.28 ± 0.11, 0.41 ± 0.15 and 0.59 ± 0.19 $L_B\ L_R^{-1}\ d^{-1}$, respectively. This increase on the biogas production was proportional to the increase in the OLR during this period. Our results are consistent with those obtained in the literature under similar operational conditions and using the same substrate (Donoso-Bravo *et al.* 2019b). These results provide baseline data to compare the results that were obtained during the ACoD periods.

3.2.2. ACoD I – chaotic period

In this period, a significant variability of the digesters behaviour was observed, thus, no stable results were attained in any reactor. R1 (control), R2 and R4 followed a similar trend, with an early increase in the biogas production (between days 56 and 63), followed by a steady decrease up until day 112, where values as low as 100 $L_B\ kg_{SV}^{-1}$ were achieved. After this period, the biogas production rebounded again on day 132, reaching similar values with those reported at the beginning of the ACoD I period. R3 showed two down-and-up periods, reaching similar values to the previous described digesters by the end of the ACoD I period. R5 and R6, which had the highest FW proportion, showed a similar behaviour with an early biogas production drop followed by an unstable production until the end of the ACoD I period. The methane content of the biogas in the reactor remained between 60 and 65%. These behaviours were also reflected in the VFA and pH values. In R4, VFA and pH values as high as 7,000 $mg_{AC\text{-}eq}\ L^{-1}$ and as low as 5.7 (day 105) were recorded, respectively. Despite this, R4 recovered a stable operation at the end of this period. Total ammonia nitrogen showed values between 1,750 up to 3,000 mg L^{-1}. The reactor's behaviour during this period cannot be associated with the impact of the presence of FW. Particularly, the behaviour of R1 (control)

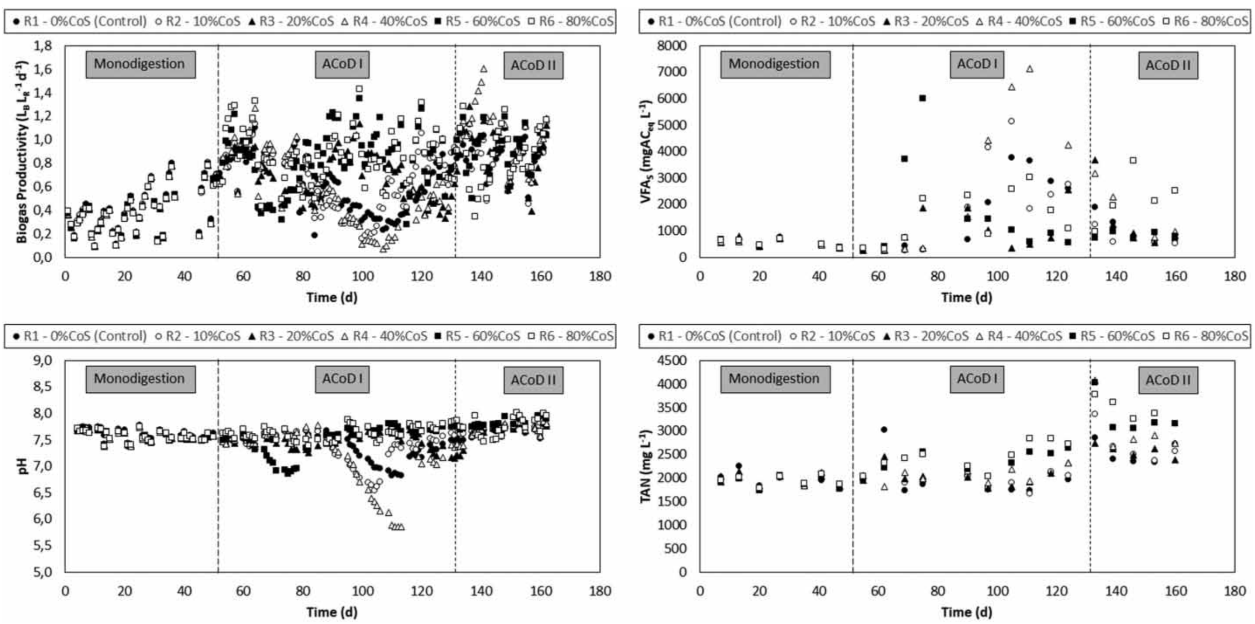

Figure 2 | Dynamic of biogas production and digestate properties during the whole operation time.

led us to conclude that there was something else than the FW effect. At the same time as this experiment was being conducted, some of the large-scale digesters operating at the WRRF also showed unstable behaviour, in terms of VFA concentration and biogas production. Those reactors were being fed with same substrate (HMS) as the laboratory-scale digesters. Because of previous similar experience at the WRRF, this anormal behaviour may be explained by the presence of VFA tolerant acetoclastic methanogens such as *Methanosarcinas*, which tend to outgrow, also known as population shift, the *Methanosaeta* group when the ammonia concentration increases (McMahon *et al.* 2001; Mehariya *et al.* 2018). This situation makes us believe that some changes in the microbial population were also taking place in the laboratory digesters.

3.2.3. ACoD II

In the last month of operation, a more stable behaviour was attained in all the digesters, hence these data were used for comparison purposes. The main results (average and quartiles) regarding the biogas production are presented in Table 3. Biogas yields in all the reactors were between 403 ± 106 and 490 ± 105 $L_B Kg_{SV}^{-1}$, in which at least one of them was statistically different (ANOVA, F (5,160) = 3.52, $p = 0.005$). After Tukey's post hoc analysis, only R4 was statistically different with R1 and R6 (more details in the Supplementary material Table S4). Similarly, the biogas productivity was between 0.85 ± 0.15 and 1.01 ± 0.24 $L_B L_R^{-1} d^{-1}$, where at least one was statically different (ANOVA, F (5,190) = 3.62, $p = 0.004$). After Tukey's post hoc analysis, only R4 was statistically different compared with R1, R2 and R6. In addition, the highest content of methane in the biogas was attained in R4. These results demonstrated that there was a synergistic effect caused by the addition of FW as co-substrate in semi-continuous operation. Despite this, a high variability in the results was observed, probably associated with the FW properties and its variable composition, along with the manual pulse-type feeding procedure. Our results are consistent with those reported by Heo *et al.* (2003), who determined that ACoD at a 50/50 FW/WAS ratio was optimal in terms of biogas production. Using the same SS/OFMSW (Organic Fraction of Municipal Solid Waste) ratio, Ara *et al.* (2015) achieved similar improvements in biogas productivity, although lower HRTs were applied. Caffaz

Table 3 | Biogas and digestate properties of the digesters during ACoD II

	R1	R2	R3	R4	R5	R6
Biogas yield ($L_B Kg_{VS}^{-1}$)						
Average	419 ± 59	427 ± 56	443 ± 85	490 ± 105	435 ± 77	403 ± 106
Q25	375.7	382.2	372.3	403.9	392.0	389.2
Q50*	415.7	445.5	447.7	447.6	439.8	414.7
Q75	442.1	495.8	468.3	490.7	495.6	509.7
Biogas productivity ($L_B L_R^{-1} d^{-1}$)						
Average	0.85 ± 0.15	0.86 ± 0.17	0.91 ± 0.20	1.01 ± 0.24	0.89 ± 0.18	0.85 ± 0.22
Q25	0.755	0.789	0.786	0.891	0.759	0.746
Q50*	0.842	0.888	0.946	0.992	0.908	0.841
Q75	0.901	0.958	1.06	1.16	1.004	1.008
CH_4 content (%)	64 ± 12	77 ± 6	63 ± 6	70 ± 1	64 ± 12	71 ± 22
Digestate						
TCOD (mg L^{-1})	55,240 ± 4,204	45,850 ± 3,270	48,460 ± 3,991	47,230 ± 2,448	43,920 ± 2,191	45,780 ± 926
SCOD (mg L^{-1})	3,202 ± 492	2,968 ± 239	3,494 ± 692	3,966 ± 1,663	2,858 ± 976	4,156 ± 525
TAN (mg L^{-1})	2,526 ± 231	2,559 ± 125	2,609 ± 204	2,822 ± 119	3,218 ± 230	3,603 ± 295
VFA (mgAC L^{-1})	778 ± 322	610 ± 106	817 ± 279	801 ± 113	866 ± 99	2,346 ± 819
pH	7.8 ± 0.2	7.8 ± 0.1	7.8 ± 0.1	7.7 ± 0.1	7.7 ± 0.1	7.7 ± 0.1
TAC (mg L^{-1})	7,467 ± 353	7,720 ± 387	8,080 ± 493	8,867 ± 846	10,293 ± 661	11,427 ± 886
NTK (mg L^{-1})	4,035	n.d	n.d	4,430	n.d	8,300
P total (mg L^{-1})	618	n.d	n.d	458	n.d	678

n.d, not determined.

et al. (2008) worked on pilot-scale two 200 L digesters for the ACoD of WAS and OFMSW at an increasing OLR which led to improvements in the yield and biogas productivity.

With regards to the digestate properties (Table 3), in general, no significant differences were observed in organic matter expressed as either total (ANOVA, F $(5,24) = 3.467$, $p = 0.017$) or soluble COD (ANOVA, F $(5,24) = 1.335$, $p = 0.284$). With regards to the VFAs, a spike of 2.5 gL^{-1} in the concentration was observed at a 80% proportion of FW. The high VFA concentration in the ACoD of FW and SS has been reported to be one of the main drawbacks for the ACoD of these wastes (Mehariya *et al.* 2018). Regarding the TAN, overall, the concentration increased for all the digesters from the monodigestion at the end of the ACoD II. During monodigestion, the TAN concentration was between 1,700–2,200 mg L^{-1} whereas during ACoD II the concentration was between 2,500–4,000 mg L^{-1}. Conversely, a steady increasing trend was observed as the proportion of FW increased. Similar results were found by Kim *et al.* (2017) who ran five laboratory-scale reactors under different conditions related to the proportions of FW and SS. The addition of co-substrate improved the biogas production and the COD removal as well as the concentration of ammonia in the digestate. Nonetheless, the values reported were lower than values observed in the present study, which shows the influence of THP on the TAN concentration in the digestate. In Kim *et al.* (2017), one of the reactors was only fed with SS (as the control test), but with an adjusted C:N ratio, equal to the reactor with the highest fraction of co-substrate. Results from this reactor demonstrated that C:N, which is usually seen as the main reason for the positive effect of adding co-substrate, has a low impact on the actual performance of the digester; thus, synergy must be a combination of factors instead of just a single one. Furthermore, some studies have neglected the effect of the C/N ratio on the synergy observed during ACoD (Yun *et al.* 2015; Ohemeng-Ntiamoah & Datta 2018). Moreover, a lower concentration of total P was observed in the digestate of R4, which may indicate that a more balanced uptake of this nutrient took place.

3.3. Pilot scale study

3.3.1. Co-substrate characterization and reactor feed

Chemical composition of FPW was variable during the whole time of experimentation, particularly in terms of solid and organic contents, as shown in Table 1. In general, milk and yogurt constituted the largest fraction of the waste, while small proportions of water, jelly and butter were occasionally added. The FPW has an organic matter content, measured as COD, almost two or three times higher than the substrate. About a quarter of this COD corresponds to the soluble fraction. The ammonia content was negligible, which contributed to balancing the C/N ratio of the mixture used for ACoD. While the VFA values were between 1.8 and 4.5 g L^{-1} during the experimental period.

3.3.2. Pilot digesters performance

The operation of the digesters was divided into three stages: Mono-AD, ACoD and Post-ACoD.

The inlet conditions applied to pilot digesters A and B during 142 days of operation in terms of OLR and HRT are shown in Figure S2 (Supplementary material). In the Mono-AD stage, after seeding, there were four days without feeding, after which the applied OLR was maintained at around 2.5 $kg_{VS}\ m^{-3}\ d^{-1}$ for 43 d. Then, the ACoD stage began, and the feeding of the co-substrate was maintained for 55 days, between day 43 and day 97. After this, during the Post-ACoD stage the reactors were fed only with HMS until the end of the operation. This allowed us to evaluate if the previous stage of ACoD impacted the operation of the reactor when returning to a Mono-AD. The only difference between both reactors was the proportion of co-substrate in the feeding.

A summary of the average values obtained during the pilot digesters operation is shown in Table 4. During Mono-AD similar performances were achieved for both pilot digesters. The biogas yield was 0.37 ± 0.09 and 0.48 ± 0.13 $L_B\ g_{VS\ ad}^{-1}$ for digesters A and B, respectively, and it showed no significant statistical difference (ANOVA, F $(1,12) = 2.839$, $p = 0.118$). Despite this, digester B obtained slightly higher values than digester A. This could be associated with some operational difficulties that took place during the feeding of digester A. In addition, the gas totalizer of digester A presented some problems of gas quantification during this first period of operation, so it was replaced by a new one.

During ACoD, digesters A and B produced 0.61 ± 0.10 $L_BgVS_{ad}^{-1}$ and 0.54 ± 0.10 $L_BgVS_{ad}^{-1}$, respectively. This difference of 12.2% was statistically significant (ANOVA, F $(1,64) = 7.246$, $p = 0.009$). However, the accumulated biogas production was slightly higher by 3.0% in digester B (Table 4). During Post-ACoD, just like during Mono-AD, the biogas yield in both reactors showed no significant statistical difference (ANOVA, F $(1,46) = 0.538$, $p = 0.467$) and was 0.50 ± 0.15 and 0.52 ± 0.09 $L_BgVS_{ad}^{-1}$ for digesters A and B, respectively.

Table 4 | Performance summary of the pilot digesters during each operation stage

		Pilot A			Pilot B		
		Mono-AD (17–42)	ACoD (43–97)	Post-ACoD (98–142)	Mono-AD (17–42)	ACoD (43–97)	Post-ACoD (98–142)
	Inlet conditions						
Co-substrate	%	0	10	0	0	20	0
OLR	$g_{VS}L^{-1}d^{-1}$	2.8 ± 0.6[a]	2.1 ± 0.4	2.2 ± 0.3	2.7 ± 0.6	2.2 ± 0.4	2.1 ± 0.3
HRT	d	14 ± 3	20 ± 4	19 ± 6	15 ± 3	20 ± 4	19 ± 5
	Biogas						
Yield	$L_BgVS_{ad}^{-1}$	0.37 ± 0.09[a]	0.61 ± 0.10	0.50 ± 0.15	0.48 ± 0.13	0.54 ± 0.10	0.52 ± 0.09
CH_4	%	68 ± 2	70 ± 2	71 ± 1	69 ± 1	70 ± 1	72 ± 1
Cumulative	$L_BL_R^{-1}$	177.8			183.1		
	Digestate						
TAN	gL^{-1}	2.1 ± 0.1	1.9 ± 0.1	2.1 ± 0.3	2.2 ± 0.2	1.9 ± 0.1	2.0 ± 0.3
tCOD	gL^{-1}	34.3 ± 0.7	32.7 ± 1.5	34.6 ± 0.8	35.9 ± 0.5	32.1 ± 1.4	34.5 ± 1.2
sCOD	gL^{-1}	1.9 ± 0.3	1.8 ± 0.1	1.8 ± 0.3	1.7 ± 0.3	2.0 ± 1.6	1.8 ± 0.4
VFA	$mg_{HAc}L^{-1}$	484 ± 111	399 ± 82	415 ± 53	511 ± 60	372 ± 45	355 ± 19
TAC	$g_{CaCO3}L^{-1}$	7.1 ± 0.4	7.2 ± 0.4	7.4 ± 0.5	7.7 ± 0.2	7.3 ± 0.5	7.0 ± 0.4
pH	–	7.7 ± 0.2	7.7 ± 0.1	7.7 ± 0.0	7.7 ± 0.2	7.7 ± 0.1	7.6 ± 0.0
$VS_{removal}$	%	48 ± 9	53 ± 6	52 ± 7	46 ± 9	54 ± 6	52 ± 8
TS	gL^{-1}	30.4 ± 0.9	27.7 ± 0.9	28.4 ± 0.7	35.1 ± 1.1	27.6 ± 0.9	28.3 ± 0.5
VS	gL^{-1}	19.9 ± 0.9	17.8 ± 0.8	18.3 ± 0.6	20.9 ± 0.8	18.0 ± 0.8	18.2 ± 0.5

[a]Operational issues occurred during this period which affects the reliability of these values.

During the digester A operation, significant differences (ANOVA, F (2,52) = 12.6, $p < 0.001$) in biogas yield were observed at each stage. Only the ACoD stage was statistically different from the other two stages given the post hoc Tukey's analysis results (more details in the Supplementary material). Our results show that using 10% co-substrate (digester A) increased biogas yield by 64.7% compared to Mono-AD. While using 20% co-substrate (digester B) the biogas production increased only by 13.8% compared to Mono-AD. During the operation of digester B, the biogas yield at each stage showed no significant difference (ANOVA, F (2,70) = 1.413, $p = 0.250$). Overall, our results demonstrated that the addition of the co-substrate promoted a better performance and operational stability of the digesters, which is evidenced by the decrease in the variability of biogas production data during ACoD, despite the variability in the chemical composition of the FPW throughout the digester's operation. These improvements during continuous ACoD using fat-rich residues as co-substrates have been already reported (Wang *et al.* 2013; Grosser *et al.* 2017).

3.3.3. Digestate properties

To evaluate the impact of co-substrate addition on the digestate properties, samples of the solid and liquid fractions were taken at the end of ACoD period for further analysis (Table 5). As a reference (control) test, a digestate sample from a full-scale reactor treating only HMS was analyzed.

Regarding the solid phase of the digestates, similar characteristics were observed in all cases. However, the sample coming from digester B had a slight lower solid content than those obtained in digester A and in the control. This may indicate that the ACoD at this co-substrate proportion may have a negative impact on the digestate dewaterability. The BMPs conducted with the solid fraction of the digestates as substrate showed that methane yields were lower than the control, which were similar to reported values under similar conditions (Ortega-Martinez *et al.* 2016). These results demonstrated that the ACoD leads to a more efficient use of the available organic matter. In addition, this is consistent with the lower total and soluble COD concentration observed in the liquid phase of the digestate samples obtained from the pilot reactors (Table 5). Regarding the nutrients, a slightly higher concentration of phosphorus was observed in the control digestate, in turn, the nitrogen content of the liquid phase was not affected by the ACoD.

Table 5 | Digestate characteristics of the pilot digesters during the ACoD period

Phase	Parameter	Unit	Control	Pilot A	Pilot B
Solid	TS	g kg^{-1}	120 ± 8	130 ± 5	100 ± 4
	VS	g kg^{-1}	80 ± 1	90 ± 1	70 ± 2
	Lipids (D.W.)	%	4.8 ± 0.2	4.6 ± 0.2	4.6 ± 0.2
	TKN	g L^{-1}	<0.5	0.91 ± 0.08	1.27 ± 0.04
	Phosphorus	mg g^{-1}	9.2 ± 0.1	8.5 ± 0.1	9.0 ± 0.1
	Methane yield (BMP test)	$NmlCH_4$ gVS^{-1}_{added}	64.7 ± 10.8	28.4 ± 4.4	24.8 ± 0.9
Liquid	TS	g L^{-1}	3.20 ± 0.02	2.40 ± 0.10	3.28 ± 0.03
	VS	g L^{-1}	2.29 ± 0.03	1.59 ± 0.07	1.98 ± 0.05
	TAC	mg_{CaCO3} L^{-1}	4,067 ± 38	3,250 ± 50	5,675 ± 43
	VFA	mg_{HAc} L^{-1}	430 ± 9	330 ± 15	600 ± 15
	TCOD	mg L^{-1}	4,300 ± 20	2,480 ± 20	3,370 ± 10
	SCOD	mg L^{-1}	3,620 ± 24	1,770 ± 6	2,760 ± 14
	TKN	g L^{-1}	4.2 ± 0.2	4.6 ± 0.1	3.4 ± 0.2

D.W., Dry Weight.

4. CONCLUSIONS

The co-digestion of thermally pretreated sewage sludge with FW was evaluated in laboratory and pilot-scale reactors.

The BMP test results showed no positive or negative synergistic effect by the incorporation of FW as co-substrate in none of the evaluated proportions regarding the biogas potential. A slight negative effect was observed for the maximum production rate.

Unlike the BMP test, the results obtained in semi-continuous operation demonstrated that there is a synergistic effect caused by the addition of FW as co-substrate. The reactor fed with a 60/40 (SS/FW) proportion yielded the best productivity with 0.490 ± 0.105 L_B g^{-1}_{VS}.

In pilot-scale experiments, the variability of the co-substrate composition had no negative impact on the digester's performance. In contrast, the addition of the co-substrate increased the production of biogas as well as led to a more efficient use of organic matter. The biogas yield was 0.61 ± 0.10 and 0.52 ± 0.10 L_B $g^{-1}_{VS\ ad}$ for the digester fed with 10 and 20% of variable food processing FW, respectively.

In both the laboratory and pilot experiences there was an impact on the amount of nitrogen in the digestate compared to digester operating in monodigestion. This impact is more significant as the proportion of co-substrate rises.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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