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Anaerobic co-digestion of condensate produced from drying of Household Food Waste and Waste Activated Sludge

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A R T I C L E I N F O

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A B S T R A C T

Drying and shedding of Household Food Waste has proved to be an effective method for its valorization. During this process a significant amount of wastewater is produced through condensation of the generated water vapors (condensate). This study investigated the possibility of valorizing the produced condensate through anaerobic co-digestion with the Waste Activated Sludge (WAS) that is produced in large quantities during the aerobic treatment of municipal wastewater. In particular, the biomethane potentials of condensate, WAS and of a mixture of condensate with WAS were calculated. It was proved that the co-digestion of condensate and WAS can increase the methane yield of WAS by 72.5%. Moreover, anaerobic co-digestion of condensate and WAS was conducted in an anaerobic digester that operated in batch and in fed-batch mode. Almost 323 mL CH4/g tCODconsumed and 350 ml CH4/g tCODconsumed were produced during the batch and the fed-batch operation of the digester,respectively.

1. Introduction

In the last 60 years the world population grew rapidly from approximately 3 billion in 1960 to 7 billion in 2011 and it is expected to reach 9.1 billion by 2050. This dramatic increase in global population in combination with economic development has led to rapid urbanization. The urbanization trends of this rapidly growing population are expected to lead to a dramatic increase in the Municipal Solid Waste (MSW) generation [1]. Recently, the generation of MSW due to urbanization is increasing at a rate surpassing that of urbanization itself [2].

In 2012, the annual global MSW generation rate was 1.3 billion tons with an average per capita generation rate of 1.2 kg/d [3]. The annual global generation of MSW is expected to reach 2.2 billion tons per year by 2025 and further increase to 4.2 billion tons by 2050. In the European Union, the average amount of MSW generated by approximately 512 million inhabitants was 477 kg per capita per year in 2015 [4].

MSW can be divided into five major categories -paper, Household Fermentable Waste (HFW) (containing kitchen and yard waste), plastic, metal, and glass [5,6]. The quantity of the HFW corresponds to 30-50% of the total MSW quantities generated [7]. Food waste (FW) occupies the highest proportion of HFW [8,9]. Conventional methods of treatment for FW include landfilling and incineration. Up to 95% of FW is ultimately landfilled. Landfilling of FW has serious environmental consequences including aquifer pollution from landfill leachate, emission of greenhouse gases and odor generation [10].

Incineration of FW leads to energy loss due to water evaporation, as the moisture of FW reaches up to 86% [11] and is often accompanied by release of dioxins. In addition, incineration leads to the loss of valuable compounds and nutrients contained in FW [12].

FW is rich in carbon and nitrogen sources such as carbohydrates, proteins and lipids. These compounds are excellent feedstocks for bioconversion to high value bioproducts such as biofuels, enzymes, probiotics, bioactive compounds or even biodegradable plastics through many different biological processes [12]. Recently, FW valorization through its conversion into rich in proteins and minerals insect larvae biomass has been proposed [13]. All these processes are proposed as alternatives to conventional methods of FW treatment.

At municipal level, apart from HFW, another important organic

Abbreviations: FORBI, Food Residue Biomass; HFW, Household Fermentable Waste; MSW, Municipal Solid Waste; FW, Food Waste; sCOD, Soluble Chemical Oxygen Demand; tCOD, Total Chemical Oxygen Demand; TSS, Total Suspended Solids; VFAs, Volatile Fatty Acids; VSS, Volatile Suspended Solids; WAS, Waste activated Sludge; St, Dev.Standard Deviation

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Table 1
Methane yield of co-digestion of WAS with different co-substrates. Modified from Yang et al. 2019 [34]

Co-substrates	Optimal mixed ratio[a]	Running condition	OLR	Biogas yield	Methane yield	Reference
Swine manure	30:70 (w/w)	Semi-continuous mesophilic	1.91 g VS/L/d	402 ml biogas/ g VS_{added}	192.5 ml CH_4/ g COD_{added}*	Borowski et al. 2014 [40]
Wheat straw	19:11 (VS)	Batch mesophilic	-		345.5 ml CH_4/ g VS_{added} 243.3 ml CH_4/ g COD_{added}*	Elsayed et al. 2016 [41]
Coffee grounds	85:15 (dry solid)	CSTR thermophilic	7.54 g COD/L/d	185 ml/g COD_{added}	107.9 ml CH_4/ g COD_{added}*	Qiao et al. 2015 [42]
Microalgae	25:75 (VS)	Batch mesophilic	-		442.5 ml CH_4/ g VS_{added} 311.6 ml CH_4/g COD_{added}*	Beltran et al. 2016 [43]
Cheese whey	5:95 (v/v)	Batch mesophilic	-		301.2 ml CH_4/g VS_{added} 212.1 ml CH_4/g COD_{added}*	Fernández et al. 2014 [44]
	5:95 (v/v)	Batch thermophilic	-		250.6 ml CH_4/g VS_{added} 176.5 ml CH_4/g COD_{added}*	
Olive mill wastewater	5:95 (v/v)	Continuous mesophilic	0.9 g VS/L/d	304 ml biogas/g VS_{added}	157.1 ml CH_4/g COD_{added}*	Maragkaki et al. 2017 [45]
Brewery sludge	75:25 (w/w)	CSTR mesophilic	1.37 g VS/L/d	650 ml biogas/g VS_{added}	220 ml CH_4/g $COD_{consumed}$	Pecharaply et al. 2007 [46]
Meat Processing sludge	46:54 (VS)	CSTR mesophilic	3.46 g VS/L/d		463 ml CH_4/g VS_{added} 326.1 ml CH_4/g COD_{added}*	Luostarinen et al. 2009 [47]
Co-substrates	Optimal mixed ratio[a]	Running condition	OLR	Biogas yield	Methane yield	Reference
Fat, oil and grease	64:36 (VS)	Semi-continuous mesophilic	2.34 g VS/L/d		598.4 ml CH_4/g VS_{added} 421.4 ml CH_4/g COD_{added}*	Wan et al. 2011[48]
Trapped Grease waste	23:77 (VS)	CSTR mesophilic	1.6 g VS/L/d		369 ml CH_4/g VS_{added} 259.9 ml CH_4/g COD_{added}*	Silvestre et al. 2011 [49]
	27:73 (VS)	CSTR thermophilic	2.1 g VS/L/d-		277 ml CH_4/g VS_{added} 195.1 ml CH_4/g COD_{added}*	Silvestre et al. 2014 [50]
Food wastewater	75:25 (v/v)	Semi-continuous thermophilic	6.88 g COD/L/d		316 ml CH_4/g $COD_{consumed}$	Jang et al. 2015 [51]
	75:25 (v/v)	Semi-continuous mesophilic	6.88 g COD/L/d		268 ml CH_4/g $COD_{consumed}$	Jang et al. 2016 [52]
Food waste	50:50 (VS)	Semi-continuous mesophilic	2.43 g VS/L/d		321 ml CH_4/g VS_{added} 226 ml CH_4/g COD_{added}*	Heo et al. 2004 [53]
Fruit waste	21:79 (w/w)	Semi-continuous mesophilic	3 g VS/L/d		300 ml CH_4/g VS_{added} 211 ml CH_4/g COD_{added}*	Fonoll et al. 2015 [54]

[a] The mixed ratio is Co-substrate:WAS
* Calculated based on origin data of the publication and regarding a VS:COD ratio of 1:1.42.

stream that is generated in large quantities is the Waste Activated Sludge (WAS). The WAS is produced during the treatment of municipal wastewater using the activated sludge method. Considering a total COD production of 120 g per person per day, a sludge production of 50-60 g dry matter per capita per day is expected [14,15].

Anaerobic Digestion (AD) has been proposed as an environmentally friendly and cost- effective alternative for the treatment of both HFW and WAS [16–21]. It is a complex microbially mediated process. in which the organic carbon is converted to its most oxidized state (carbon dioxide), and to its most reduced form (methane) in the form of biogas. [22]. Biogas consists of methane (50–70%), carbon dioxide (30–50%) and minor amounts of other compounds, such as nitrogen, oxygen, hydrogen sulfide, ammonia and water vapor. The Calorific Value of methane is 50.4 MJ/kg. For biogas with a methane content in the range of 60–65% the Lower Calorific Value (LCV) is approximately 20–25 MJ/m^3-biogas [23]. Biogas is a versatile energy carrier that can be used directly for combined heat and electricity (CHP) generation or can be upgraded into biomethane through removal of CO_2 by processes such as water/amine scrubbing, pressure swing adsorption or the Sabatier reaction. It can then be fed to the natural gas grid or used after compression as a fuel (bioCNG) in the automotive sector [24]. Apart from biogas, digestate is also produced. The digestate is the stabilized nutrient-rich effluent of the AD process, which can be used either as a soil conditioner or as compost after being properly processed [25].

Although AD is currently used for FW treatment at industrial scale, its use is limited and still often faces several technical challenges, such as the need for feedstock pretreatment, VFA accumulation and process instability, foaming, low buffer capacity, and especially high cost of transportation and operation [17]. AD has been traditionally used for the stabilization and reduction of the solid content of WAS. During the AD of WAS energy in the form of biogas is produced [26] but due to the low biodegradability of WAS, the process is not cost effective [27]. Biogas production from AD of WAS can be improved by several pre-treatment methods. Thermal, chemical, biological, enzymatic [28] and mechanical processes, as well as combinations of these, have been studied as possible pre-treatment methods of WAS. These methods

Table 2
Main characteristics of condensate, anaerobic sludge and WAS.

	Condensate		WAS		Anaerobic sludge	
Parameter	Average*	St.Dev.	Average*	St.Dev.	Average**	St.Dev.
tCOD (g O_2/L)	11.7	3.07	35.45	5.07	17.09	0.22
sCOD (g O_2/L)	11.7	0.00	0.60	0.28	0.15	0.02
TSS (g/L)	0.05	0.02	39.15	9.97	21.7	1.20
VSS (g/L)	-	-	21.70	1.98	12.7	1.10
pH	4.45	0.40	6.77	0.35	6.98	0.00
Acetate (mg/L)	1340.00	251.40	95.30	10.89	0.00	0.00
Propionate (mg/L)	49.08	13.23	85.60	14.71	0.00	0.00
Iso-butyrate (mg/L)	53.38	16.26	23.62	11.06	0.00	0.00
Butyrate (mg/L)	73.14	40.27	19.95	13.51	0.00	0.00
Iso-valerate (mg/L)	14.99	8.35	14.36	10.10	0.00	0.00
Valerate (mg/L)	0.00	0.00	2.85	1.82	0.00	0.00
Ethanol (mg/L)	3.09	0.85	0.00	0.00	0.00	0.00
TKN (mg/L)	10.90	4.44	1440.00	282.84	1200	120.20
Total alkalinity (mg $CaCO_3$/L)	***		1200.00	150.00	3750.00	250.00

* Average values of the characteristics of three different samples are depicted.
** Average values of the characteristics of three independent measurements of the same sample.
*** Cannot be measured due to low pH.

Table 3
Composition of the feedstocks used in the batch experiments in bench scale bioreactors. Different ratios of condensate to WAS were used in each experiment to investigate the applicability of co-digestion.

Composition (% v/v)			Nomenclature
Condensate	WAS	Anaerobic Sludge (Inoculum)	
19	76.25	4.75	CWA
-	95.25	4.75	WA
95.25	-	4.75	CA
-	-	100	CONTROL

cause disintegration of the cells contained in the WAS, permitting the release of their intracellular matter that becomes more accessible to anaerobic microorganisms. This improves the overall digestion process rate and the extent of sludge degradation, thus reducing the required retention time of the anaerobic digester and increasing the methane production rates [29]. However, these methods of pretreatment require extensive use of chemicals, heat, electricity, or some combination of these, so their application is limited for economic reasons. An alternative method for enhancement of AD of WAS is its co-digestion with other streams of organic waste [30].

It has been proved that for several feedstocks, co-digestion systems perform better than mono-digestion ones [31]. Several studies showed the benefits of co-digestion through mechanisms such as dilution of potential toxic compounds, improvement of nutrient balance, synergistic effects of microorganisms, increased load of biodegradable organic matter leading to higher biogas yields [32]. In particular, co-digestion of WAS with other organic wastes could increase the amount of biodegradable organic matter and at the same time provide a feedstock with an optimum C:N ratio [26]. The optimal C:N ratio for AD ranges from 20 to 30, depending on the feedstock used [33]. A lack of nitrogen has negative effects on the methane yield as it constitutes a structural element of many intracellular components (eg. DNA, RNA and proteins). Conversely, a high nitrogen concentration can imply an excess in the formation of ammonia which is toxic for the process of AD when present at high levels. Many different agricultural, industrial and municipal organic wastes have been used as co-substrates for the anaerobic co-digestion of WAS. The co-digestion of WAS with most of these substrates lead to higher biogas production, compared to the mono-digestion of WAS (Table 1) [9,34].

At municipal level, FW and WAS are currently being treated as separate waste streams, defined by the main phase in each case, solid and liquid, respectively. In the municipality of Halandri [35], in Attica, Greece an innovative FW valorization approach was developed and implemented at pilot-scale within the framework of the Horizon 2020 project WASTE4think [36]. The implemented waste management scheme included the source-separated collection of the household food waste from 250 households. The collected FW was then led to a drying/shredding facility of the Municipality. The drying/shredding process of the food waste results in a homogenized solid biomass product named FORBI (Food Residue Biomass). The mean moisture of FORBI is 10%, as almost 75-80% of the initial moisture of raw material is removed. Moisture is removed in the form of water vapors that are collected by a condenser. FORBI, rich in carbon and nitrogen with optimal C:N ratio is an ideal substrate for many biological processes, such as anaerobic digestion, dark fermentation [37], composting [38] and electricity production through microbial fuel cells [39]. The produced condensate is rich in organic carbon but poor in nitrogen which limits its biological treatment.

Based on the results of the project and the characteristics of condensate, an alternative scenario in which the condensate can be combined and co-managed with the WAS is proposed. In this research work, the feasibility of co-digesting the condensate with WAS is assessed as a novel approach for the valorization of these waste streams.

Through this novel approach for simultaneous treatment of WAS and condensate, the drying and shredding of the FW could take place nearby the existing anaerobic digesters in Wastewater Treatment Plants in order to increase their biogas yield and render the procedure of municipal wastewater treatment less energy demanding and more economical, reducing at the same time the transportation cost of FW treatment.

2. Material and methods

2.1. Analytical methods

The measurements of tCOD and sCOD, TSS and VSS and temperature were carried out according to Standard Methods [55]. The pH was measured using a digital pH-meter (WTW INOLAB PH720). For the quantification of VFAs, 1 ml of sample acidified with 30 μL of 20% H_2SO_4 was analyzed via a gas chromatograph (SHIMADZU GC-2010 plus) equipped with a flame ionization detector and a capillary column (Agilent technologies, 30 m x0.53 mm ID x1 μm film, HP-FFAP) using an autosampler (SHIMADZU AOC-20 s). The oven was programmed from 105 °C to 160 °C at a rate of 15 °C·min^{-1} and subsequently to 225 °C (held for 3 min) at a rate of 20 °C·min^{-1}. Helium was used as the carrier gas at 30 ml·min^{-1}, the injector temperature was set at 230 °C and the detector at 230 °C. For the quantification of the methane content of the biogas, a GC-TCD with Helium as carrier gas was used (SHIMADZU GC-2014). The separation column's (Supelco Carboxen 1000) length was 5 m and the interior diameter 2.1 mm. The initial temperature of the GC-TCD was 40 °C. For the estimation of the methane content a temperature program was used (total duration: 25 min.) during which the temperature was increasing 10 °C·min^{-1} until reaching 185 °C and staying stable at this temperature for 5 minutes. The methane content then was calculated using a standard calibration curve. The biogas production rate was measured using an oil displacement technique [56,57].

2.2. Drying and shredding

The drying and shredding of HFW took place in the commercially available Dryer-shredder GAIA GC-300. In each operation cycle, 130 kg

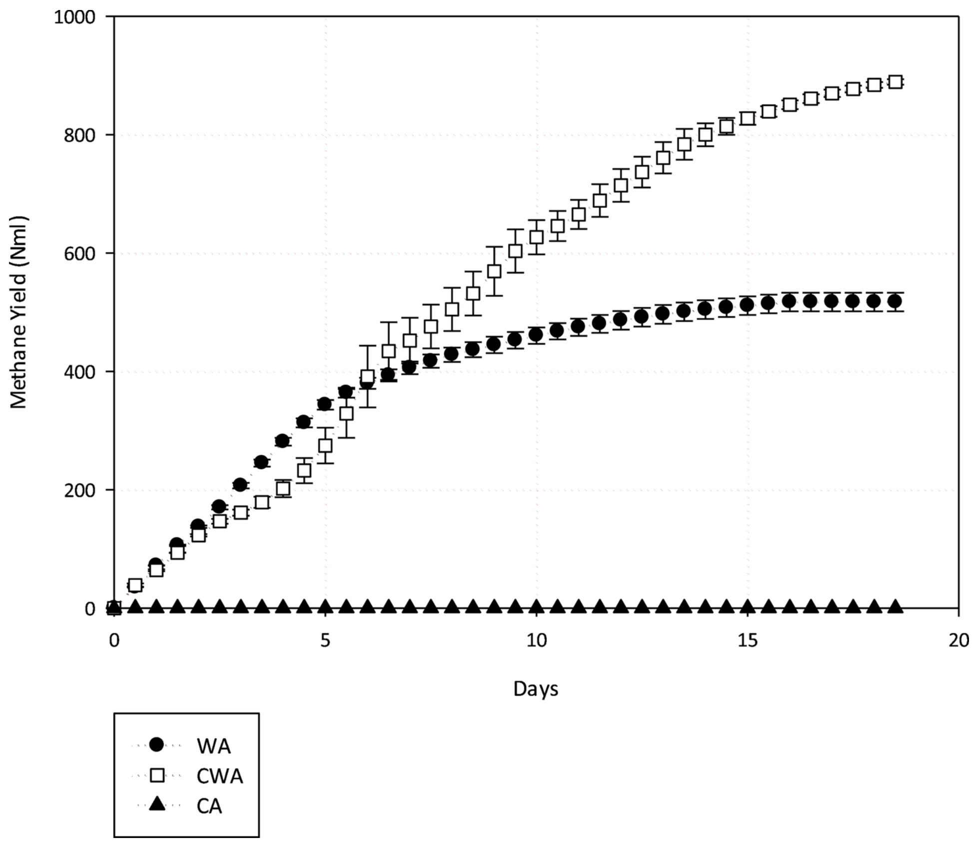

Fig. 1. Cumulative methane production from anaerobic digestion of WAS (WA), condensate (CA) and co-digestion of WAS and condensate (CWA). Each point is the mean of three different replicate experiments. Error bars indicate standard deviation.

Table 4
Main characteristics of each bench scale bioreactor during the start-up and after the end of the batch experiment. Data represent the mean (± standard deviation) of three independent experiments, each performed in triplicate. Total VFAs represent the cumulative concentration of acetate, propionate, isobutyrate, butyrate, isovalerate and valerate.

Sample	Parameter	Initial concentration	Final concentration
CWA	tCOD (g/L)	26.68 ± 1.04	20.02 ± 0.72
WA		28.43 ± 2.05	21.74 ± 1.19
CA		19.64 ± 0.03	20.00 ± 1.79
CWA	sCOD (g/L)	4.08 ± 0.08	0.15 ± 0.03
WA		0.42 ± 0.19	0.13 ± 0.04
CA		18.77 ± 0.99	18.35 ± 1.00
CWA	TKN (mg/L)	1083.69 ± 125	-
WA		1335.25 ± 143	
CA		74.15 ± 15.12	
CWA	TSS (g/L)	28.45 ± 1.74	19.78 ± 2.08
WA		35.12 ± 5.95	23.19 ± 3.94
CA		1.69 ± 0.25	0.29 ± 0.36
CWA	VSS (g/L)	13.68 ± 1.05	9.06 ± 3.24
WA		16.89 ± 2.39	10.13 ± 2.58
CA		0.80 ± 0.15	0.15 ± 0.08
CWA	Total VFAs (mg/L)	671.582 ± 20.05	0 ± 0.01
WA		159.69 ± 15.75	0 ± 0.01
CA		2905.92 ± 124	2878.176 ± 135
CWA	pH	6.4 ± 0.3	7.45 ± 0.2
WA		6.8 ± 0.2	7.52 ± 0.15
CA		4.5 ± 0.2	4.28 ± 0.2

of FW are used, producing 30 kg of FORBI with 10% humidity and 100 kg of condensate. The collected FW contains mainly kitchen waste including fruits, vegetables and cooked food. The food waste is placed inside the chamber of the dryer- shredder and the temperature of the chamber is increased up to 94 °C using an electrical resistance. The temperature is maintained at 94 °C for 9 hours until the drying procedure is complete. A shredder inside the machine is used for grinding. During the process the vapors generated from the chamber are passed through a condenser, generating a liquid condensate.

2.3. Substrates and inoculum

Anaerobic sludge obtained from the mesophilic anaerobic digester of the Municipal Wastewater Treatment Plant of Lycovrisi, Attica, Greece was used as inoculum for both bench and lab scale bioreactors.

The substrates for the co-digestion were condensate from the drying and shredding of FW and WAS from the abovementioned Municipal Wastewater Treatment Plant. Table 2 gives the main characteristics of anaerobic sludge, condensate and WAS.

The condensate: WAS ratio used throughout the experiments was 1:4. This ratio was determined considering the typical production of 0.04 kg of WAS in dry base per capita [58]. The average TSS of the samples taken from WAS and examined in this study was 39.15 ± 9.97 g/L, which is common for WAS, therefore almost 0.8 L of WAS are produced per capita per day. Regarding the production of HFW, almost 0.05- 0.06 kg of HFW on a dry basis are produced per capita per day [14] corresponding to 0.22 kg of raw HFW per capita per day. As mentioned before, 100 kg of condensate are produced from drying and shredding of 130 kilograms of raw HFW, which occupy a volume of 110 L. Therefore, almost 0.2 L of condensate are produced per capita per day.

2.4. Methane potential assessment

Batch experiments were performed using the Automated Methane Potential Test System II (AMPTS; Bioprocess Control AB, Lund, Sweden). Each of the AMPTS' bottles (500 ml total volume; 400 ml

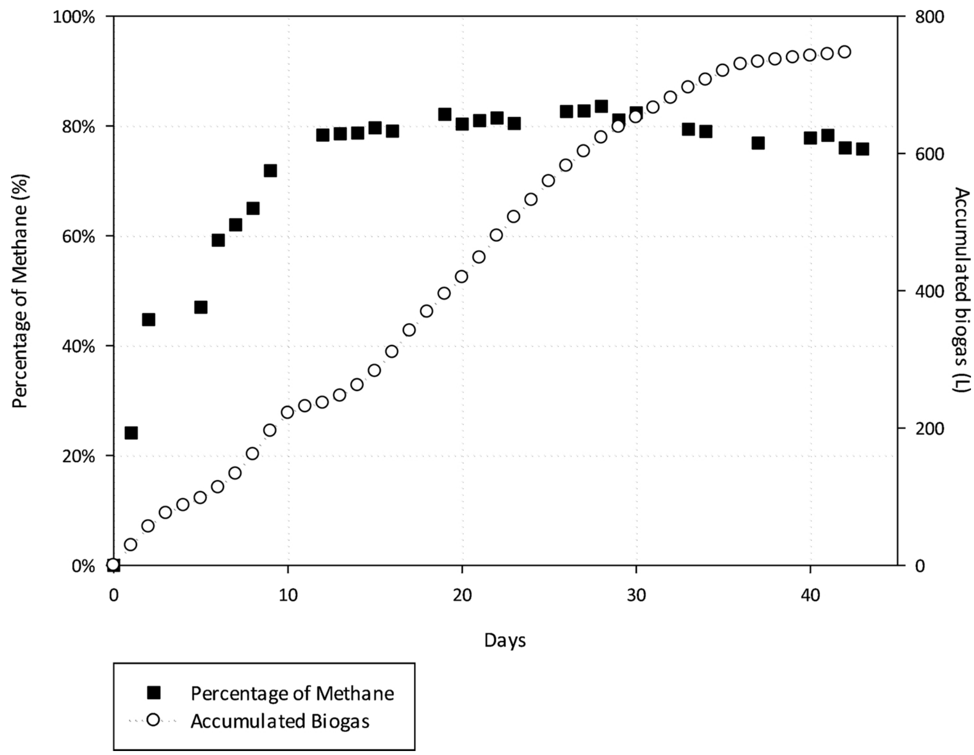

Fig. 2. Cumulative biogas production and percentage of methane during the batch operation of the lab scale anaerobic digester. For the batch operation of the bioreactor a mixture of WAS and condensate in a ratio 4:1 and the inoculum were added at once during start-up.

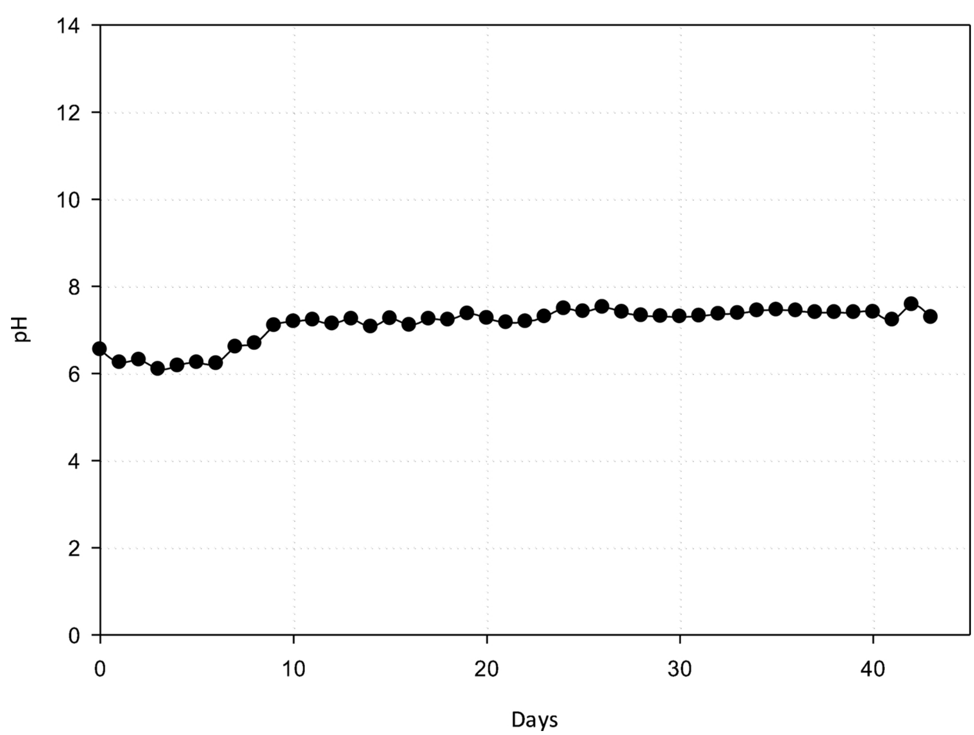

Fig. 3. pH of the bioreactor during the batch operation of the lab scale anaerobic digester. For the batch operation of the bioreactor the feedstocks and the inoculum were added at once during start-up.

working volume and 100 ml headspace) was equipped with an individual mechanical mixer and operated as a bench scale anaerobic bioreactor. The produced biogas from each of the bottles passed through a 3 M NaOH solution which retained CO_2 and H_2S, while allowing methane to pass though. Finally, the upgraded biogas passed through a flow meter device (one for each incubation bottle) which measured gas productivity through water displacement. The results of bench scale experiments are expressed as normalized mL [59]. All experiments started at the same time using the same inoculum and continued until no further biogas was produced. During start-up, flushing with N_2 took place and all samples were incubated at 35 °C throughout the experiment.

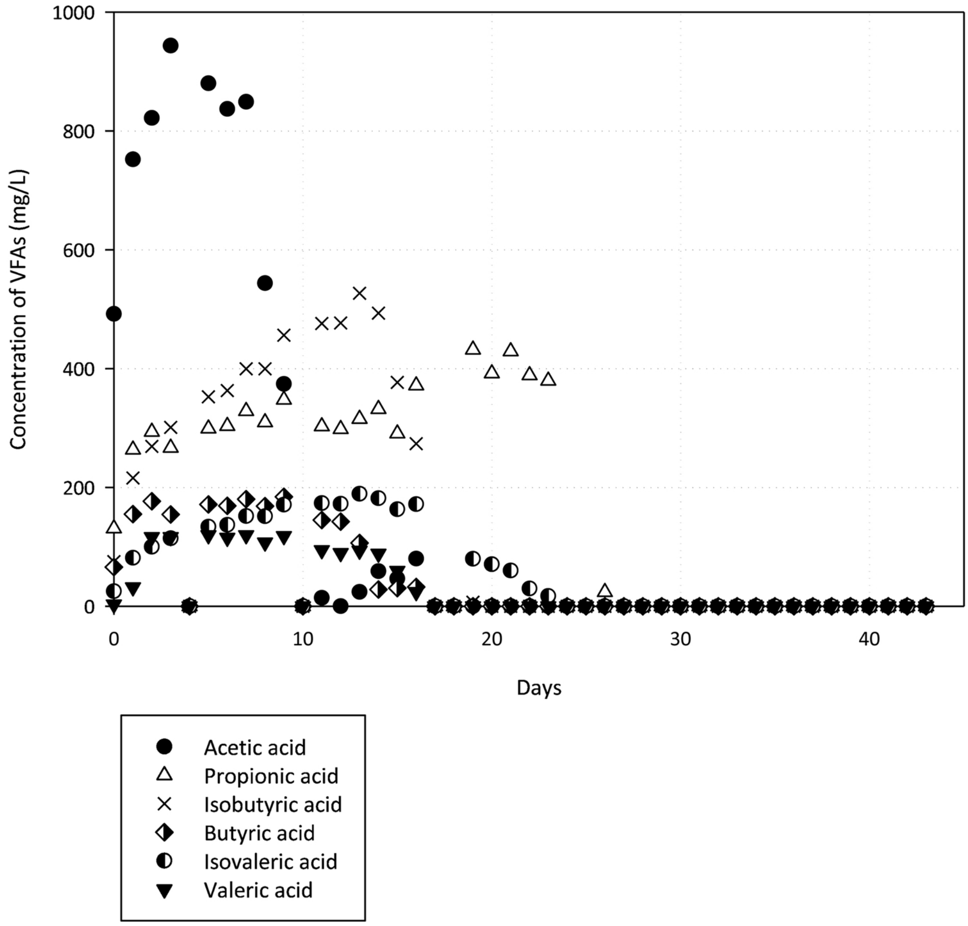

Fig. 4. Concentration of VFAs during the batch operation of the lab scale anaerobic digester. For the batch operation of the bioreactor the feedstocks and the inoculum were added at once during start-up.

Table 5
Main characteristics of the of the lab scale bioreactor during the start-up and after the end of the batch experiment. Total VFAs represent the cumulative concentration of acetate, propionate, isobutyrate, butyrate, isovalerate and valerate.

Parameter	Initial concentration	Final concentration
tCOD (g/L)	33.75	17.9
sCOD (g/L)	3.82	0.28
TSS (g/L)	42.8	25.1
VSS (g/L)	18.8	13.0
pH	6.55	7.29
Total VFAs (mg/L)	791.64	0
TKN (mg/L)	1100	-

These experiments were conducted to determine the methane yield of the individual substrates (condensate and WAS) and of the mixture of the condensate and WAS (ratio 1:4). Anaerobic mono-digestions of WAS and condensate were performed to assess the biomethane potential of each substrate. Anaerobic co-digestion was performed to assess the feasibility of co-digesting the two streams. The experimental design is shown in Table 3. All batch tests were performed in triplicate.

2.5. Batch and fed-batch experiments in bioreactor

A bioreactor made of stainless steel with a working volume of 100 L was used for the conduction of batch and fed-batch experiments. The content of the bioreactor was continuously stirred by a propeller agitator. The temperature was kept constant at 35 °C by circulating water from a thermostated bath through the bioreactor's jacket. In both cases, during start-up, 5 L of anaerobic sludge were used as inoculum. For the batch mode operation 20 L of condensate and 80 L of WAS were added at once during start-up. For the fed batch operation, 80 L of WAS were added at the start-up of the bioreactor and 1 L of condensate was added once a day for a time period of 20 days.

3. Results and discussion

3.1. Methane potential experiments

The cumulative methane yields during the anaerobic digestion of each substrate and co-digestion of both substrates are shown in Fig. 1. The batch experiments lasted 20 days until little or no biogas production was observed. The results presented are the net methane yield after subtracting the control yield.

According to Fig. 1, no methane was produced from anaerobic digestion of the condensate alone. Almost 518 Nml of methane were produced through anaerobic digestion of WAS, which corresponds to a methane yield of 193.6 Nml CH_4/g $tCOD_{consumed}$.

Co-digestion of WAS and condensate led to a higher methane yield of 334.1 Nml CH_4/g $tCOD_{consumed}$ producing almost 890 Nml of methane. Therefore, the co-digestion of WAS with condensate enhanced the methane yield from 193.6 to 334.1 Nml CH_4/g $tCOD_{consumed}$ that is an almost 72.5% increase in the methane yield compared with the anaerobic digestion of WAS.

The C:N ratio of the feedstock used in each experiment is an important factor that determines the methane yield. The C:N ratio of condensate is almost 1070:1 therefore the amount of nitrogen is a limiting factor for its anaerobic digestion. Through the addition of WAS

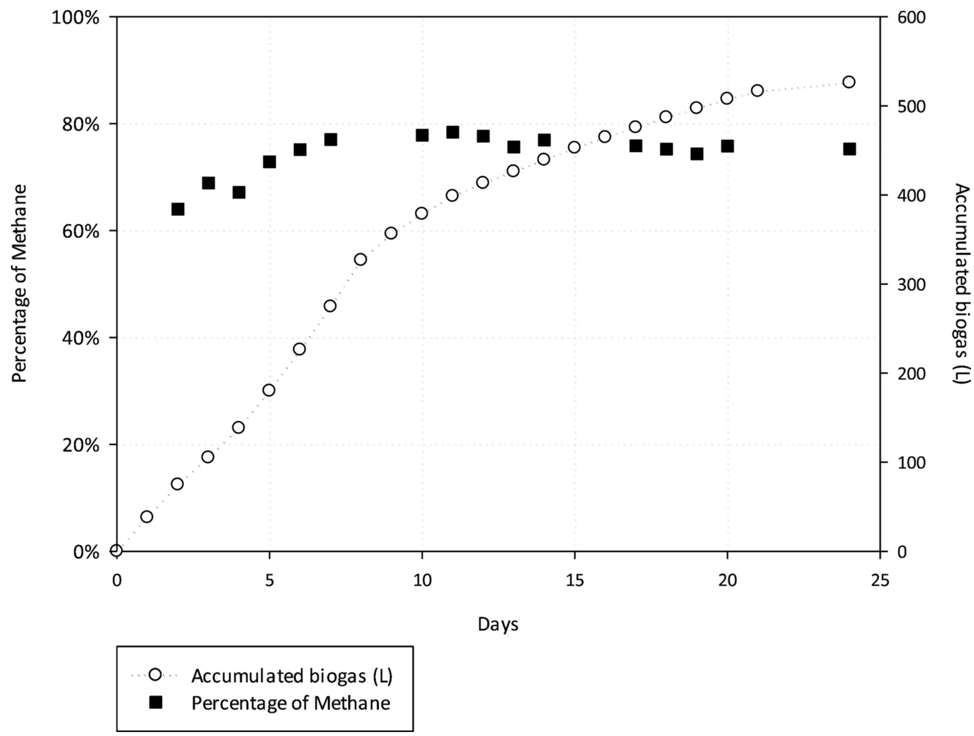

Fig. 5. Cumulative biogas production and percentage of methane during the fed batch operation of the lab scale anaerobic digester. For the fed batch operation, WAS and the inoculum were added at the start-up of the bioreactor and 1 L of condensate was added once a day for a time period of 20 days.

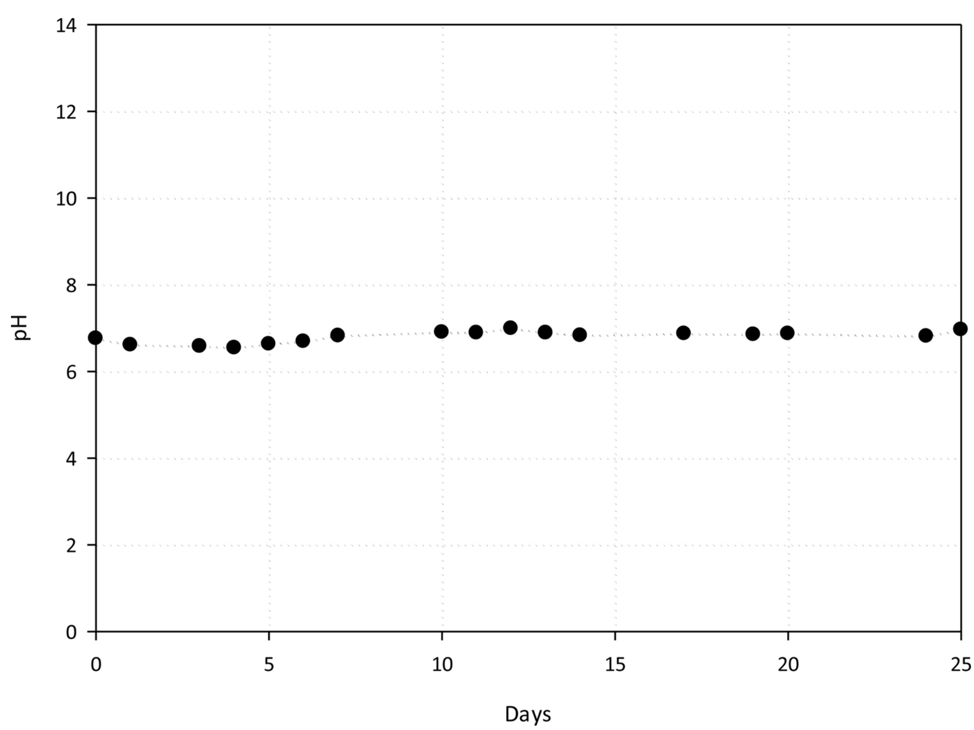

Fig. 6. pH of the bioreactor during the fed batch operation of the lab scale anaerobic digester.. For the fed batch operation, WAS and the inoculum were added at the start-up of the bioreactor and 1 L of condensate was added once a day for a time period of 20 days.

to condensate, the C:N ratio of the feedstock used for anaerobic digestion increased from 21.3 (WA) to 24.6 (CWA) (Table 4).

3.2. Batch experiment in bioreactor

A batch experiment in a bioreactor with a working volume of 100 L was also conducted. The batch experiment lasted for 42 days until no further biogas was produced. As seen in Fig. 2, almost 750 L biogas were produced during the operation of the bioreactor. The mean methane percentage was 71.6% and it remained above 59% after the sixth day.

During start-up of the bioreactor, the pH decreased sharply reaching 6.1, which is inhibitory for anaerobic digestion. Nevertheless, after the 7th day of operation the pH of bioreactor (Fig. 3) increased up to 7.

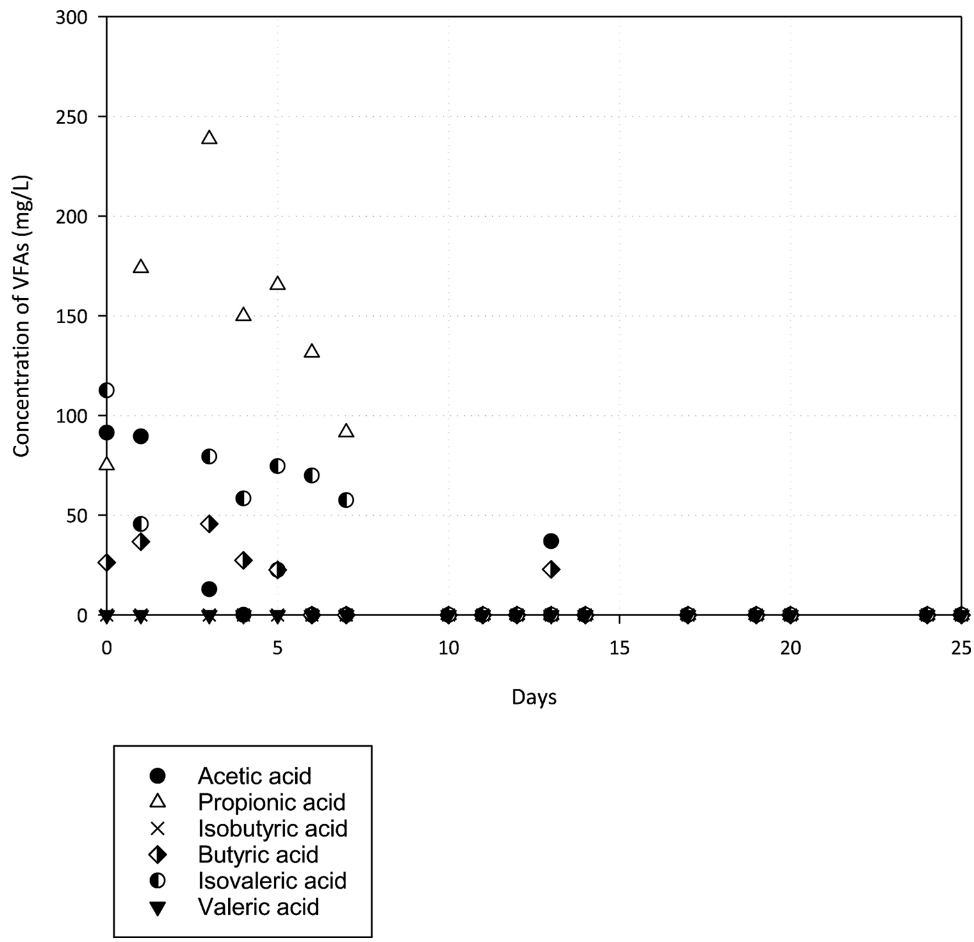

Fig. 7. Concentration of VFAs during the fed batch operation of the lab scale anaerobic digester. For the fed batch operation, WAS and the inoculum were added at the start-up of the bioreactor and 1 L of condensate was added once a day for a time period of 20 days.

Table 6
Main characteristics of the of the lab scale bioreactor during the start-up and after the end of the fed batch experiment. Total VFAs represent the cumulative concentration of acetate, propionate, isobutyrate, butyrate, isovalerate and valerate.

Parameter	Initial concentration	Final concentration
tCOD (g/L)	32.04	16.69
sCOD (g/L)	1.21	0.15
TSS (g/L)	33.22	21.73
VSS (g/L)	16.47	12.74
pH	6.76	6.96
Total VFAs (mg/L)	305.1	0
TKN (mg/L)	1250	-

Then, it remained stable and close to the optimum for anaerobic digestion pH range (6.8 to7.2) [60].

The significant decrease of pH during the start-up of the bioreactor coincided with a significant increase in the concentration of VFAs of bioreactor (Fig. 4). The concentration of acetate increased significantly during the start-up of the bioreactor and reached up to 945 mg/L. The accumulation of acetate is indicative of inhibition of acetotrophic methanogenesis. After a period of acclimation, the concentration of VFAs decreased significantly until no VFAs were detected.

Co-digestion of WAS and condensate in bioreactor led to a methane yield of 322.67 mL CH_4/ g $tCOD_{consumed}$ (Table 5). Therefore, the results obtained from the batch experiment in the lab scale bioreactor are in agreement with the findings of the methane potential experiments.

3.3. Fed batch experiment in bioreactor

According to Fig. 1 the cumulative methane production curve during the batch experiment appears to be sigmoid. The lag phase is indicative of inhibition. During the lag phase the pH of the bioreactor remained below the optimum for methanogens range of pH resulting in a low percentage of methane in the produced biogas. This inhibition could be attributed to the low pH and high concentration of VFAs of the condensate. In order to alleviate the possible inhibition due to addition of a high quantity of condensate, we also operated the bioreactor in a fed-batch mode.

The fed batch experiment lasted for 24 days until no further biogas was produced. Almost 525 L of biogas were produced during the fed-batch operation of the lab scale bioreactor (Fig. 5). The mean methane percentage was 74.3% and remained above 64% throughout the experiment.

The pH remained above 6.5 throughout the fed-batch experiment (Fig. 6) which is within the optimum pH range for anaerobic digestion. In addition, the concentration of VFAs remained below 310 mg/L throughout the experiment (Fig. 7). Therefore, VFAs did not accumulate in the bioreactor.

The fed-batch operation of the lab scale bioreactor led to a methane yield of 342.81 mL CH_4/ g $tCOD_{consumed}$ (Table 6), which is higher than the one obtained though the batch operation of the lab scale digester. The methane yield is close to the theoretical yield of methane which is equal to 350 ml/ g $tCOD_{consumed.}$ Therefore, the co-digestion of WAS with condensate proved to be a really promising alternative for the valorization of both streams.

3.4. Discussion

Co-digestion of WAS and condensate proved to be an effective way not only to enhance the methane yield of WAS but also to treat the condensate.

The condensate could not be used as feedstock for anaerobic microorganisms due to low concentration of TKN. The low C:N ratio of condensate hinders the biological treatment of this stream, as there is not enough nitrogen for the microorganisms to build up important biological molecules like proteins and nucleic acids. Apart from nitrogen, the condensate is also poor in other minerals and phosphate, which are important for the biological processes. Nevertheless, the condensate contains easily degradable carbon that can easily be consumed by microorganisms whenever sufficient amounts of nitrogen, phosphorus and minerals are secured.

During the batch operation of the 100 L bioreactor almost 320 mL CH_4/ g $tCOD_{consumed.}$ which is close to the yield obtained from anaerobic co-digestion of FW with food wastewater. (Table 1). Even higher methane yield was achieved during the fed-batch operation of the bioreactor, reaching up to 342.81 mL CH_4/ g $tCOD_{consumed}$.. In this research work the ratio of condensate: WAS used in the co-digestion process was calculated based on real data regarding the production of condensate and WAS at municipal level, so we did not attempt to optimize these ratios to get a better methane yield but to investigate the effect of several operational parameters on the methane yield.

This research work represents part of an innovative method for the combined treatment of the overall biodegradable organic wastes (HFW and wastewater) that are produced at Municipal level which is totally different from the current waste management scheme

4. Conclusion

In this study, WAS and condensate, produced through drying and shredding of source-separated collected FW, were co-digested in mesophilic conditions. Almost 322.67 mL CH_4/ g $tCOD_{consumed}$ and 342.81 mL CH_4/ g $tCOD_{consumed}$ were produced during the batch and the fed-batch operation of a lab scale anaerobic digester. The co-digestion of WAS and condensate enhanced the methane yield of WAS by 40% (batch operation) and by 43.5% (fed batch operation). This finding is important for the re-design of the current waste management scheme. Based on the obtained results, units for the drying and shredding of FW could be installed in existing wastewater treatment plants. The produced condensate could be co-digested with WAS, limiting the cost of treatment of wastewater treatment and the produced FORBI could be an ideal substrate for many biological processes.

Declaration of Competing Interest

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

CRediT authorship contribution statement

G. Lytras: Methodology, Validation, Writing - review & editing. **E. Koutroumanou:** Investigation, Writing - original draft. **G. Lyberatos:** Conceptualization, Writing - review & editing, Supervision.

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References

[1] D. Hoornweg, P. Bhada-Tata, What a Waste: A Global Review of Solid Waste Management, Urban Dev. Ser. Knowl. Pap. No.15, World Bank, 2012, p. 116, https://doi.org/10.1111/febs.13058.
[2] The World Bank, What a waste: a global review of solid waste management, (2012), https://doi.org/10.1111/febs.13058.
[3] A. Kumar, S.R. Samadder, A review on technological options of waste to energy for effective management of municipal solid waste, Waste Manag. 69 (2017) 407–422, https://doi.org/10.1016/j.wasman.2017.08.046.
[4] J. Malinauskaite, H. Jouhara, D. Czajczyńska, P. Stanchev, E. Katsou, P. Rostkowski, R.J. Thorne, J. Colón, S. Ponsá, F. Al-Mansour, L. Anguilano, R. Krzyżyńska, I.C. López, A. Vlasopoulos, N. Spencer, Municipal solid waste management and waste-to-energy in the context of a circular economy and energy recycling in Europe, Energy. 141 (2017) 2013–2044, https://doi.org/10.1016/j.energy.2017.11.128.
[5] W.S. Ho, H. Hashim, J.S. Lim, C.T. Lee, K.C. Sam, S.T. Tan, Waste Management Pinch Analysis (WAMPA): Application of Pinch Analysis for greenhouse gas (GHG) emission reduction in municipal solid waste management, Appl. Energy. 185 (2017) 1481–1489, https://doi.org/10.1016/j.apenergy.2016.01.044.
[6] Y.-C. Chen, Effects of urbanization on municipal solid waste composition, Waste Manag. 79 (2018) 828–836, https://doi.org/10.1016/j.wasman.2018.04.017.
[7] I. Mιchalopoulos, G.M. Lytras, K. Papadopoulou, A. Goumenos, I. Zacharopoulos, C. Lytras, G. Lyberatos, Hydrogen and Methane Production from Food Residue Biomass Product (FORBI), 15th Int. Conf. Environ. Sci. Technol (2017) 1–5.
[8] K. Paritosh, M. Yadav, S. Mathur, V. Balan, W. Liao, N. Pareek, V. Vivekanand, Organic fraction of municipal solid waste: Overview of treatment methodologies to enhance anaerobic biodegradability, Front. Energy Res. (2018), https://doi.org/10.3389/fenrg.2018.00075.
[9] W.L. Chow, S. Chong, J.W. Lim, Y.J. Chan, M.F. Chong, T.J. Tiong, J.K. Chin, G.T. Pan, Anaerobic co-digestion of wastewater sludge: A review of potential co-substrates and operating factors for improved methane yield, Processes. (2020), https://doi.org/10.3390/pr8010039.
[10] G. Capson-Tojo, M. Rouez, M. Crest, J.-P. Steyer, J.-P. Delgenès, R. Escudié, Food waste valorization via anaerobic processes: a review, Rev. Environ. Sci. Bio/Technology. 15 (2016) 499–547, https://doi.org/10.1007/s11157-016-9405-y.
[11] V. Cabbai, M. Ballico, E. Aneggi, D. Goi, BMP tests of source selected OFMSW to evaluate anaerobic codigestion with sewage sludge, Waste Manag. (2013), https://doi.org/10.1016/j.wasman.2013.03.020.
[12] H.S. Ng, P.E. Kee, H.S. Yim, P.-T. Chen, Y.-H. Wei, J. Chi-W. Lan, Recent advances on the sustainable approaches for conversion and reutilization of food wastes to valuable bioproducts, Bioresour. Technol. 302 (2020) 122889, , https://doi.org/10.1016/j.biortech.2020.122889.
[13] J.-W. Lim, S.-N. Mohd-Noor, C.-Y. Wong, M.-K. Lam, P.-S. Goh, J.J.A. Beniers, W.-D. Oh, K. Jumbri, N.A. Ghani, Palatability of black soldier fly larvae in valorizing mixed waste coconut endosperm and soybean curd residue into larval lipid and protein sources, J. Environ. Manage. 231 (2019) 129–136, https://doi.org/10.1016/j.jenvman.2018.10.022.
[14] L.D. Nghiem, K. Koch, D. Bolzonella, J.E. Drewes, Full scale co-digestion of wastewater sludge and food waste: Bottlenecks and possibilities, Renew. Sustain. Energy Rev. 72 (2017) 354–362, https://doi.org/10.1016/j.rser.2017.01.062.
[15] D. Bolzonella, F. M\icolucc\i, F. Batt\ista, C. Cav\inato, M. Gottardo, S. P\iovesan, P. Pavan, Producing Biohythane from Urban Organic Wastes, Waste and Biomass Valorization. (2019), https://doi.org/10.1007/s12649-018-00569-7.
[16] J.-I. Oh, J. Lee, K.-Y.A. Lin, E.E. Kwon, Y.F. Tsang, Biogas production from food waste via anaerobic digestion with wood chips, Energy Environ. 29 (2018) 1365–1372, https://doi.org/10.1177/0958305X18777234.
[17] F. Xu, Y. Li, X. Ge, L. Yang, Y. Li, Anaerobic digestion of food waste – Challenges and opportunities, Bioresour. Technol. 247 (2018) 1047–1058, https://doi.org/10.1016/j.biortech.2017.09.020.
[18] S.K. Pramanik, F.B. Suja, S.M. Zain, B.K. Pramanik, The anaerobic digestion process of biogas production from food waste: Prospects and constraints, Bioresour. Technol. Reports. 8 (2019) 100310, , https://doi.org/10.1016/j.biteb.2019.100310.
[19] F. Kader, A.H. Baky, M.N.H. Khan, H.A. Chowdhury, Production of Biogas by Anaerobic Digestion of Food Waste and Process Simulation, Am. J. Mech. Eng. 3 (2015) 79–83, https://doi.org/10.12691/ajme-3-3-2.
[20] H. Fisgativa, A. Tremier, Influence of food waste characteristics variations on treatability through anaerobic digestion, (2015).
[21] Y. Xu, Y. Lu, L. Zheng, Z. Wang, X. Dai, Perspective on enhancing the anaerobic digestion of waste activated sludge, J. Hazard. Mater. 389 (2020) 121847, , https://doi.org/10.1016/j.jhazmat.2019.121847.
[22] P.G. Kougias, I. Angelidaki, Biogas and its opportunities—A review, Front. Environ. Sci. Eng. 12 (2018) 14, https://doi.org/10.1007/s11783-018-1037-8.
[23] I. Angelidaki, L. Treu, P. Tsapekos, G. Luo, S. Campanaro, H. Wenzel, P.G. Kougias, Biogas upgrading and utilization: Current status and perspectives, Biotechnol. Adv. 36 (2018) 452–466, https://doi.org/10.1016/j.biotechadv.2018.01.011.
[24] I. Bassani, P.G. Kougias, L. Treu, I. Angelidaki, Biogas Upgrading via Hydrogenotrophic Methanogenesis in Two-Stage Continuous Stirred Tank Reactors at Mesophilic and Thermophilic Conditions, Environ. Sci. Technol. 49 (2015) 12585–12593, https://doi.org/10.1021/acs.est.5b03451.
[25] M. Logan, C. Visvanathan, Management strategies for anaerobic digestate of organic fraction of municipal solid waste: Current status and future prospects, Waste Manag. Res. 37 (2019) 27–39, https://doi.org/10.1177/0734242X18816793.
[26] Z. Zahan, M.Z. Othman, W. Rajendram, Anaerobic Codigestion of Municipal

Wastewater Treatment Plant Sludge with Food Waste: A Case Study, Biomed Res. Int. (2016), https://doi.org/10.1155/2016/8462928.
[27] N.D. Park, R.W. Thring, S.S. Helle, Comparison of methane production by co-digesting fruit and vegetable waste with first stage and second stage anaerobic digester sludge from a two stage digester, Water Sci. Technol. 65 (2012) 1252–1257, https://doi.org/10.2166/wst.2012.004.
[28] Y.X. Liew, Y.J. Chan, S. Manickam, M.F. Chong, S. Chong, T.J. Tiong, J.W. Lim, G.T. Pan, Enzymatic pretreatment to enhance anaerobic bioconversion of high strength wastewater to biogas: A review, Sci. Total Environ (2020), https://doi.org/10.1016/j.scitotenv.2019.136373.
[29] J.A. Müller, Pretreatment processes for the recycling and reuse of sewage sludge, Water Sci. Technol. 42 (2000) 167–174, https://doi.org/10.2166/wst.2000.0197.
[30] M. Al-Addous, M.N. Saidan, M. Bdour, M. Alnaief, Evaluation of biogas production from the co-digestion of municipal food waste and wastewater sludge at refugee camps using an automated methane potential test system, Energies. (2019), https://doi.org/10.3390/en12010032.
[31] J.A.V. Piñas, O.J. Venturini, E.E.S. Lora, O.D.C. Roalcaba, Technical assessment of mono-digestion and co-digestion systems for the production of biogas from anaerobic digestion in Brazil, Renew. Energy. 117 (2018) 447–458, https://doi.org/10.1016/j.renene.2017.10.085.
[32] D. Bolzonella, F. Battista, C. Cavinato, M. Gottardo, F. Micolucci, G. Lyberatos, P. Pavan, Recent developments in biohythane production from household food wastes: A review, Bioresour. Technol. (2018), https://doi.org/10.1016/j.biortech.2018.02.092.
[33] X. Wang, X. Lu, F. Li, G. Yang, Effects of Temperature and Carbon-Nitrogen (C/N) Ratio on the Performance of Anaerobic Co-Digestion of Dairy Manure, Chicken Manure and Rice Straw: Focusing on Ammonia Inhibition, PLoS One. 9 (2014) 1–7, https://doi.org/10.1371/journal.pone.0097265.
[34] Q. Yang, B. Wu, F. Yao, L. He, F. Chen, Y. Ma, X. Shu, K. Hou, D. Wang, X. Li, Biogas production from anaerobic co-digestion of waste activated sludge: co-substrates and influencing parameters, Rev. Environ. Sci. Bio/Technology. 18 (2019) 771–793, https://doi.org/10.1007/s11157-019-09515-y.
[35] Municipality of Halandri, Municipality of Halandri, Munic. Halandri Off. Website, (2014).
[36] WASTE4Think, Moving towards Life cycle Thinking by integrating Advanced Waste Management Systems, (2015).
[37] I. Michalopoulos, G.M. Lytras, D. Mathioudakis, C. Lytras, A. Goumenos, I. Zacharopoulos, K. Papadopoulou, G. Lyberatos, Hydrogen and Methane Production from Food Residue Biomass Product (FORBI), Waste and Biomass Valorization. (2019), https://doi.org/10.1007/s12649-018-00550-4.
[38] I. Michalopoulos, G. Lytras, S. Michalakidi, S. Zgouri, K. Papadopoulou, G. Lyberatos, Evaluation of in-vessel and pilot scale composting as an alternative for food waste valorization, 7th Int. Conf. Eng. Waste Biomass Valoris, Prague, 2018.
[39] A. Tremouli, I. Karydogiannis, P.K. Pandis, K. Papadopoulou, C. Argirusis, V.N. Stathopoulos, G. Lyberatos, Bioelectricity production from fermentable household waste extract using a single chamber microbial fuel cell, Energy Procedia. 161 (2019) 2–9, https://doi.org/10.1016/j.egypro.2019.02.051.
[40] S. Borowski, J. Domański, L. Weatherley, Anaerobic co-digestion of swine and poultry manure with municipal sewage sludge, Waste Manag. (2014), https://doi.org/10.1016/j.wasman.2013.10.022.
[41] M. Elsayed, Y. Andres, W. Blel, A. Gad, A. Ahmed, Effect of VS organic loads and buckwheat husk on methane production by anaerobic co-digestion of primary sludge and wheat straw, Energy Convers. Manag. (2016), https://doi.org/10.1016/j.enconman.2016.03.064.
[42] W. Qiao, S. Mohammad, K. Takayanagi, Y.Y. Li, Thermophilic anaerobic co-digestion of coffee grounds and excess sludge: Long term process stability and energy production, RSC Adv. (2015), https://doi.org/10.1039/c4ra15581e.
[43] C. Beltrán, D. Jeison, F.G. Fermoso, R. Borja, Batch anaerobic co-digestion of waste activated sludge and microalgae (Chlorella sorokiniana) at mesophilic temperature, J. Environ. Sci. Heal. - Part A Toxic/Hazardous Subst. Environ. Eng. (2016), https://doi.org/10.1080/10934529.2016.1181456.
[44] C. Fernández, D. Blanco, J. Fierro, E.J. Martínez, X. Gómez, Anaerobic Co-digestion of Sewage Sludge with Cheese Whey under Thermophilic and Mesophilic Conditions, Int. J. Energy Eng. 4 (2014) 26–31, https://doi.org/10.5923/j.ijee.20140402.02.
[45] A.E. Maragkaki, M. Fountoulakis, A. Gypakis, A. Kyriakou, K. Lasaridi, T. Manios, Pilot-scale anaerobic co-digestion of sewage sludge with agro-industrial by-products for increased biogas production of existing digesters at wastewater treatment plants, Waste Manag. (2017), https://doi.org/10.1016/j.wasman.2016.10.043.
[46] A. Pecharaply, P. Parkpian, A.P. Annachhatre, A. Jugsujinda, Influence of anaerobic co-digestion of sewage and brewery sludges on biogas production and sludge quality, J. Environ. Sci. Heal. - Part A Toxic/Hazardous Subst. Environ. Eng. (2007), https://doi.org/10.1080/10934520701369818.
[47] S. Luostarinen, S. Luste, M. Sillanpää, Increased biogas production at wastewater treatment plants through co-digestion of sewage sludge with grease trap sludge from a meat processing plant, Bioresour. Technol. (2009), https://doi.org/10.1016/j.biortech.2008.06.029.
[48] C. Wan, Q. Zhou, G. Fu, Y. Li, Semi-continuous anaerobic co-digestion of thickened waste activated sludge and fat, oil and grease, Waste Manag. (2011), https://doi.org/10.1016/j.wasman.2011.03.025.
[49] G. Silvestre, A. Rodríguez-Abalde, B. Fernández, X. Flotats, A. Bonmatí, Biomass adaptation over anaerobic co-digestion of sewage sludge and trapped grease waste, Bioresour. Technol. (2011), https://doi.org/10.1016/j.biortech.2011.04.019.
[50] G. Silvestre, J. Illa, B. Fernández, A. Bonmatí, Thermophilic anaerobic co-digestion of sewage sludge with grease waste: Effect of long chain fatty acids in the methane yield and its dewatering properties, Appl. Energy. (2014), https://doi.org/10.1016/j.apenergy.2013.11.075.
[51] H.M. Jang, M.S. Kim, J.H. Ha, J.M. Park, Reactor performance and methanogenic archaea species in thermophilic anaerobic co-digestion of waste activated sludge mixed with food wastewater, Chem. Eng. J. (2015), https://doi.org/10.1016/j.cej.2015.04.072.
[52] H.M. Jang, J.H. Ha, M.S. Kim, J.O. Kim, Y.M. Kim, J.M. Park, Effect of increased load of high-strength food wastewater in thermophilic and mesophilic anaerobic co-digestion of waste activated sludge on bacterial community structure, Water Res. (2016), https://doi.org/10.1016/j.watres.2016.04.051.
[53] N.H. Heo, S.C. Park, H. Kang, Effects of mixture ratio and hydraulic retention time on single-stage anaerobic co-digestion of food waste and waste activated sludge, J. Environ. Sci. Heal. - Part A Toxic/Hazardous Subst. Environ. Eng. (2004), https://doi.org/10.1081/ESE-120037874.
[54] X. Fonoll, S. Astals, J. Dosta, J. Mata-Alvarez, Anaerobic co-digestion of sewage sludge and fruit wastes: Evaluation of the transitory states when the co-substrate is changed, Chem. Eng. J. (2015), https://doi.org/10.1016/j.cej.2014.10.045.
[55] APHA/AWWA/WEF, Standard Methods for the Examination of Water and Wastewater, Stand. Methods. (2012) 541 https://doi.org/ISBN 9780875532356.
[56] I.V. Skiadas, H.N. Gavala, G. Lyberatos, Modelling of the periodic anaerobic baffled reactor (PABR) based on the retaining factor concept, Water Res. 34 (2000) 3725–3736, https://doi.org/10.1016/S0043-1354(00)00137-8.
[57] I.V. Skiadas, G. Lyberatos, The periodic anaerobic baffled reactor, Water Sci. Technol. 38 (1998) 401–408, https://doi.org/10.1016/S0273-1223(98)00717-3.
[58] A. Karagiannidis, P. Samaras, T. Kasampalis, G. Perkoulidis, P. Ziogas, A. Zorpas, Evaluation of sewage sludge production and utilization in Greece in the frame of integrated energy recovery, Desalin. Water Treat. 33 (2011) 185–193, https://doi.org/10.5004/dwt.2011.2613.
[59] H. Himanshu, M.A. Voelklein, J.D. Murphy, J. Grant, P. O'Kiely, Factors controlling headspace pressure in a manual manometric BMP method can be used to produce a methane output comparable to AMPTS, Bioresour. Technol. 238 (2017) 633–642, https://doi.org/10.1016/j.biortech.2017.04.088.
[60] A.E. Cioabla, I. Ionel, G.A. Dumitrel, F. Popescu, Comparative study on factors affecting anaerobic digestion of agricultural vegetal residues, Biotechnol. Biofuels. (2012), https://doi.org/10.1186/1754-6834-5-39.