

Enhancing Stable Anaerobic Digestion of Foodwaste: Co-digestion with Liquid Dairy Manure or Manure Digestate?

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Abstract

Process instability has been a challenge to long-term, continuous anaerobic digestion of foodwaste. Co-digestion is one of the measures to solve instability issue. However, overdosing of a co-substrate may result in inhibition to foodwaste digestion. This study experimentally compared liquid dairy manure and manure digestate as co-substrate for mesophilic digestion of foodwaste and used four substrate limitation – inhibition kinetic models to identify the optimal co-substrate fraction. The dynamics of cumulative biogas production in batch co-digestion experiments could be highly simulated with the modified Gompertz function. Lag phase varied between 0 and 3.3 d, being generally longer at a higher fraction of a co-substrate. Generally, the manure digestate exerted greater inhibition than the liquid dairy manure at the co-substrate fractions tested, 5-50% in terms of substrate volatile solids (VS). The models, however, showed that manure digestate had higher half-inhibition concentrations (88-195% VS) than liquid dairy manure (43-104% VS). Biogas yield reaches the highest when co-digesting with 16.3% manure digestate or 12.0% liquid dairy manure, which are lower than the ranges experimentally compared in earlier studies. At the optimal co-substrate fractions, the maximum biogas yield is 0.66 L/g VS with manure digestate as co-substrate and 0.97 L/g VS with liquid dairy manure as co-substrate. Both liquid dairy manure and manure digestate are attractive co-substrates for anaerobic digestion of foodwaste. Semi-continuous co-digestion of 88% foodwaste and 12% liquid dairy manure at hydraulic retention time of only 14 d attained 81% of the simulated maximum biogas yield.

Keywords: Anaerobic co-digestion, biogas production, dairy manure, foodwaste, instability, kinetic model

1 INTRODUCTION

Laboratory anaerobic digestion studies have demonstrated high biochemical methane potential of foodwaste. Process instability, however, has been a challenge to long-term, continuous anaerobic digestion of foodwaste, especially at higher organic loading rates and under thermophilic conditions [1-4]. The process instability and

failure have been mostly attributed to fast acidification, ammonia inhibition to acetoclastic methanogens, and inadequate availability of trace elements. The high contents of non-fiber carbohydrate and fat of foodwaste may contribute to fast acidification (Table 1). Ammonia accumulation is attributed to its high protein content. Subsequently, successful long-term mono-digestion of food waste has been limited to daily organic loading rates typically less than 4.5 g VS/L unless enhancement measures such as co-digestion are taken [1, 5].

Table 1. Characteristics of substrates used in this study^a.

	Foodwaste	Liquid dairy manure	Digested dairy manure	Digested sludge
pH	4.8	7.7	8.0	7.7
Total solids, TS (g/L)	272.5	123.0	45.5	26.1
VS/TS (%)	92.1	84.6	68.2	55.4
Biochemical composition (% TS)				
Crude protein	27.8	16.9	41.7	31.8
Crude fat	22.4	6.1	4.8	3.3
Non-fiber carbohydrate	35.1	6.5	0.0	3.9
Lignin	2.8	10.6	16.2	5.6
Cellulose	0.9	23.4	7.6	1.6
Hemicellulose	3.2	21.9	5.7	14.3
Elemental macronutrient (% TS)				
Nitrogen	4.6	2.9	7.0	7.3
Phosphorus	0.42	0.80	1.37	2.29
Calcium	0.28	1.74	2.87	4.05
Magnesium	0.06	0.79	1.59	0.65
Potassium	0.67	2.26	6.09	0.67
Sodium	1.15	0.63	1.82	0.82
Sulfur	0.27	0.54	0.71	1.46
Elemental micronutrient (mg/kg TS)				
Iron	43	1041	1220	113350
Zinc	31	194	383	491
Copper	4	122	94	382
Manganese	8	155	308	310
Molybdenum	0.5	2.7	4.6	17.4
Cobalt	0.19	2.18	3.28	7.72

^a Average of two samples.

In an effort to enhance stable digestion of foodwaste, this study investigated the effects of two co-substrates for anaerobic digestion of foodwaste, i.e., liquid dairy manure (LDM) and anaerobically digested dairy manure (ADDM). Earlier studies [6-8] on co-digestion of foodwaste and LDM reported stable operation and synergistic effects at organic loading rates as high as 6.2-12 g VS/L/d, which were attributed to balanced biochemical composition or C/N ratio, neutralized pH, and supplementation of micronutrients from co-substrates. Synergistic effects are manifested with the increased co-substrate fraction, whereas overdosing of a co-substrate may result in disappearance of synergies and even antagonistic effects. The optimum substrate combination is typically determined through laboratory experiments at discrete

substrate combinations or by modeling without consideration of inhibition [6, 9, 10]. This study explored substrate-limitation and inhibition kinetic models for determination of optimal substrate combination.

Because of the high fiber content of LDM, ADDM from digesters without an extended hydraulic retention time can have higher residual methane potential. Based on biochemical composition of the LDM and ADDM at the same farm, it was estimated with an equation developed by Angelidaki and Sanders [11] that the ADDM would have even higher biomethane potential (0.38 L CH₄/g VS) than the LDM (0.20 L CH₄/g VS). Moreover, the ADDM had higher pH value and nutrient concentrations than the LDM on a dry matter basis (Table 1). Furthermore, it might be less concerned about odor and health issues to transport digestate than undigested LDM from a farm to a foodwaste digestion plant. Therefore, side-by-side comparison of LDM and ADDM as co-substrate could potentially discover a new enhancement measure for anaerobic digestion of foodwaste.

2 MATERIALS AND METHODS

2.1 Batch Experiments on Anaerobic Co-digestion

Batch experiments on co-digestion of foodwaste were conducted in 2-L continuously-stirred mesophilic digesters [8]. For each co-substrate (LDM and ADDM), batch co-digestion experiments were conducted at 7 substrate combinations, i.e., co-substrate VS : foodwaste VS in 5% : 95%, 10% : 90%, 15% : 85%, 20% : 80%, 30% : 70%, 40% : 60%, and 50% : 50%, plus two blanks (each inoculum only). The foodwaste was collected over two weeks from buffet leftovers at the Turning Stone Casino in Verona, New York, USA, ground through a 2.5-mm aperture, and stored at -21 °C. The ADDM was collected from the effluent of mesophilic manure digesters at Twin Birch Dairy Farm in Skaneateles, New York. The LDM was collected from the same farm at the same time as the ADDM. Each digester was initially added 21.345 g VS of inoculum with 1.09-1.46 L of ADDM sieved through 1-mm mesh and anaerobically digested sludge from Syracuse Metropolitan Wastewater Treatment Plant located in Syracuse, New York in the same VS ratios as the targeted co-substrate to foodwaste ratios. The inoculum and headspace were flushed with nitrogen gas and sealed immediately. Substrate to inoculum ratio was set at 0.82 with 17.551 g of combined substrate VS. The substrates and inocula were characterized by Dairy One Forage Laboratory in Ithaca, New York, USA and results are given in Table 1.

After 2 d of acclimation, the digesters were fed with substrates and deionized water to a working volume of 1.8 L and controlled at temperature 36 °C. Biogas production was recorded every 15 min with Model 1615A digital mass flow gas meters (Omega Engineering, Norwalk, Connecticut, USA), displaying biogas flow rate at temperature 25 °C and pressure 14.7 psi. Based on the dynamics of biogas production rate, the batch experiments lasted for 25-30 d. Biogas samples (0.1 mL each) were collected weekly with a gas tight syringe and diluted with air in 11.3-mL Restek serum vials for determination of methane content using a Shimadzu GC-2014 gas chromatograph with a flame ionization detector. Helium was used as carrier gas. The temperatures of oven, injector port and detector were 100, 140 and 100 °C, respectively. Measurements were taken for pH with a Hach HI60 meter connected to an ISFET NMR tube pH probe before feeding and at the end of each batch. Initial and final

concentrations of TS and VS were determined for each batch according to Standard Methods 2540 B and E [12]. Samples collected at the end of each batch were centrifuged at 2500 g for 30 min and centrate samples were diluted for determination of total ammonia nitrogen (TAN) concentration with a QuickChem 8500 series automatic flow injection analyzer (LaChat Instruments, Loveland, Colorado, USA), following Standard Method flow injection analysis [12].

Cumulative biogas production from each batch at a specific substrate combination ratio minus cumulative biogas production from inoculum was fitted over time with the modified Gompertz function [10] as given in Equation 1. The three kinetic constants Y_m , R_m and λ were estimated simultaneously for each batch experiment using the Microsoft Excel Solver tool.

$$Y_t = Y_m \exp \left\{ -\exp \left[\frac{R_m \times e}{Y_m} (\lambda - t) + 1 \right] \right\} \quad (1)$$

where Y_m = the maximum biogas yield (L/g VS); t = time of biogas production in batch anaerobic digestion (d); Y_t = biogas yield at t (L/g VS); λ = length of lag phase (d); and R_m = maximum specific biogas production rate (L/d/g VS).

2.2 Modeling Substrate Limitation – Inhibition Kinetics

The variations of biogas yield and lag-phase length with co-substrate fraction were fitted with four kinetic models which consider substrate inhibition effect, i.e., the Andrews inhibition function in Equations 2, Edwards model in Equation 3, Haldane function in Equation 4, and Aiba model in Equation 5 [13, 14]. The kinetic constants were estimated using the Microsoft Excel Solver tool. The similarity between the experimental data and the model predictions was assessed with correlation coefficient, r . K_s and K_i represent the lowest and highest co-substrate concentration, respectively, at which the biogas yield equals to one half the maximum biogas yield.

$$q = q_{max} \frac{S}{K_s + S + \frac{S^2}{K_i}} \quad (2)$$

$$q = q_{max} \left(e^{-\frac{S}{K_i}} - e^{-\frac{S}{K_s}} \right) \quad (3)$$

$$q = q_{max} \frac{S}{(K_s + S) \left(1 + \frac{S}{K_i} \right)} \quad (4)$$

$$q = q_{max} \frac{S e^{-S/K_i}}{K_s + S} \quad (5)$$

where q = the maximum biogas yield Y_m observed at a given substrate combination (L/g VS); q_{max} = the biogas yield potential attainable in the presence of inhibition (L/g VS); S = the VS fraction of a co-substrate in the total substrate VS (%); K_s = the half saturation percentage of the co-substrate (%); and K_i = the inhibition constant (%).

2.3 Semi-Continuous Co-digestion of Foodwaste and Liquid Dairy Manure

Based on results of the batch co-digestion experiments, semi-continuous mesophilic digestion was performed with 88% foodwaste and 12% LDM in terms of substrate

VS. Four digesters were operated in parallel under the same conditions. Inoculum (1.8 L each digester) was made of the sludge digestate and ADDM in the same VS amount (21.345 g) as the batch digestion experiments and the same VS ratio of sludge digestate : ADDM as the substrates (88% foodwaste and 12% ADDM). The inoculum and headspace were flushed with nitrogen gas and sealed immediately. After 2 d of acclimation, it started to feed daily through a chute mounted on the digester cap and submerged in digestate. The digestate temperature was set at 36 °C. Hydraulic retention time was set at 14 d by discharging 129 mL of digestate daily and diluting the feed with tap water to 129 mL for each digester. Organic loading rate was increased by 0.5 g VS/L/d every 14 d from 1.0 to 3.0 g VS/L/d.

Biogas flow rate, pressure and temperature were recorded every 15 min with the digital gas meters. Biogas samples (0.1 mL each) were collected weekly for determination of methane content using the same methods as for batch experiments. Digester effluent was determined weekly for pH, temperature, TS, VS and TAN, using the same methods as described in Section 2.1.

3 RESULTS AND DISCUSSION

The operational conditions in the batch anaerobic co-digestion experiments are given in Table 2. The inoculum sources had similar pH values, 7.51 in the sludge digestate and 7.61 in the sieved ADDM. The measured initial and final pH values were similar across all the batch experiments. The pH values were in the range for stable anaerobic digestion, pH 6.8-8.1 [1, 9]. The slight pH decreases through the batch digestion experiments were attributed to the high pH buffering capacity of dairy manure [15]. The inoculum sources had TAN concentrations at 1170 mg/L in the sludge digestate and 1675 mg/L in the sieved ADDM. Correspondingly, the initial TAN concentrations were estimated for the batch experiments at the specified substrate combination ratios (Table 2). Over the batch digestion, TAN concentration increased by 44-138%. Both initial and final TAN concentrations were in the range for stable anaerobic digestion, less than 5000 mg/L [1], and generally below the inhibition levels as reviewed by Yenigun and Demirel [16] and Rajagopal et al. [17]. Upon feeding, the initial TS and VS concentrations were 2.8-3.9% and 1.9-2.7%, respectively. TS and VS concentrations decreased mostly by 50-76% and 56-82%, respectively. There was an insignificant difference in TS and VS reduction efficiencies between ADDM and LDM ($P = 0.07$ and 0.16).

As shown in Fig. 1, after a lag phase biogas production in all the substrate combinations increased rapidly and began to diverge after approximately 10 d. In the first 4 d there were plateaus of biogas production, especially with ADDM as co-substrate, indicating anaerobic oxidation of readily available organic compounds followed by hydrolysis of particulate substrates. The cumulative biogas production could be well simulated with the modified Gompertz function ($R^2 = 0.977-0.998$). Biogas in all the batches had methane content varied between 58% and 68%, which increased over the first 10 d.

Table 2. Performance and operational conditions of batch co-digestion of foodwaste with different co-substrates.

	Fraction of co-substrate VS in total substrate VS (%)						
	5	10	15	20	30	40	50
<i>Liquid dairy manure as co-substrate</i>							
TS reduction (%)	69	67	74	75	72	74	51
VS reduction (%)	75	73	77	81	77	77	56
Initial pH	7.57	7.60	7.52	7.61	7.58	7.70	7.63
Final pH	7.51	7.42	7.36	7.52	7.48	7.51	7.35
Initial TAN (mg/L)	958	941	924	907	873	838	804
Final TAN (mg/L)	1650	1510	1580	1425	1555	1600	1700
<i>Anaerobically digested dairy manure as co-substrate</i>							
TS reduction (%)	27	76	55	52	63	50	55
VS reduction (%)	43	82	68	66	67	59	59
Initial pH	7.54	7.53	7.73	7.76	7.75	7.80	7.67
Final pH	7.48	7.41	7.51	7.50	7.45	7.57	7.51
Initial TAN (mg/L)	958	941	924	907	873	838	804
Final TAN (mg/L)	1380	1785	1380	1325	1410	1585	1910

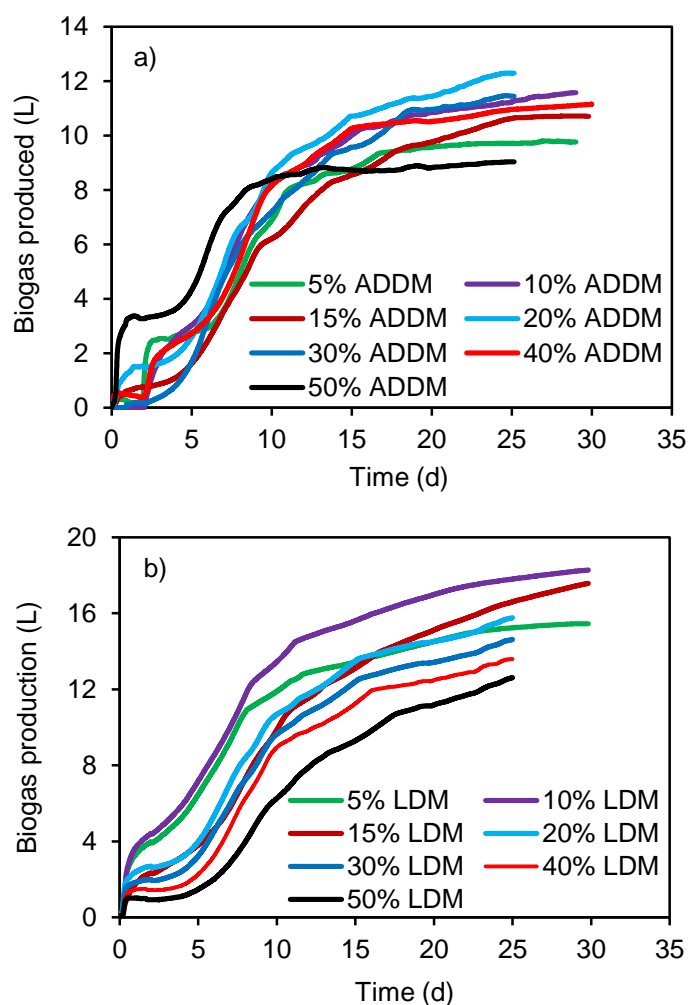


Fig. 1 Cumulative biogas production in batch co-digestion of foodwaste at different VS fractions of a) manure digestate (ADDM), and b) liquid dairy manure (LDM)

3.1 Kinetics of Anaerobic Co-digestion in the Presence of Inhibition

At a small fraction of a co-substrate, the co-substrate serves as a supplementary source of nutrients for microorganisms. At a large fraction, it may behave as an inhibitor to microbial growth. The values of maximum biogas yield and lag phase length estimated with the Gompertz function are presented in Fig. 2 and Fig. 3. As shown in Fig. 2, the length of lag phase increased with increasing fraction of co-substrate, despite the absence of a lag phase in co-digestion of foodwaste with ADDM at 50% substrate VS fraction. Increased lag time is the primary result of inhibition [14]. Possibly, it was inhibition of co-substrate to the microorganisms originated from the seeded sludge digestate since more ADDM was added to the inoculum mixture for the batches with a greater fraction of co-substrate. The unstable biogas production in the first a few days of the batch experiments (Fig. 1) demonstrated an acclimation stage. In continuously fed anaerobic digestion, this type of inhibition can be avoided when acclimated inoculum is used. At most of the co-substrate fractions, ADDM exerted greater inhibition or had a longer lag phase than the LDM. The longer lag phase at higher LDM fractions may also be attributed to the recalcitrance of more lignin and cellulose in the LDM than ADDM (Table 1).

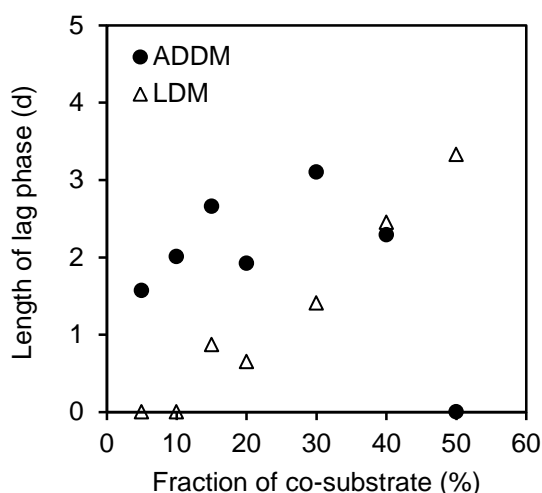


Fig. 2 Variation of lag phase length with the fraction of co-substrate in anaerobic co-digestion of foodwaste with anaerobically digested dairy manure (ADDM) and liquid dairy manure (LDM)

Substrate-limiting anaerobic digestion kinetics has been largely simulated with the Michaelas-Menten equation or its modifications [10]. However, substrate inhibition due to over-dosing of a co-substrate in anaerobic digestion has rarely been simulated [6]. Inhibition often involves multiple, rather than specific mechanisms, such as reduced activity of an enzyme by complexing with excess substrate, modification of physiochemical variables, and imbalanced metabolism of the cell [13]. As Edwards [13] concluded upon comparison of five kinetic models that significant case-to-case variations may occur in the functional form of substrate inhibition, this study selected four inhibition kinetic models to fit the results from the batch digestion experiments. The Haldane and Andrews models were derived with the assumption of multiple inactive enzyme-substrate complexes. The semi-empirical Edwards model assumes a protective diffusion-limitation of high and inhibitory substrate concentrations. The

Aiba model is an empirical equation.

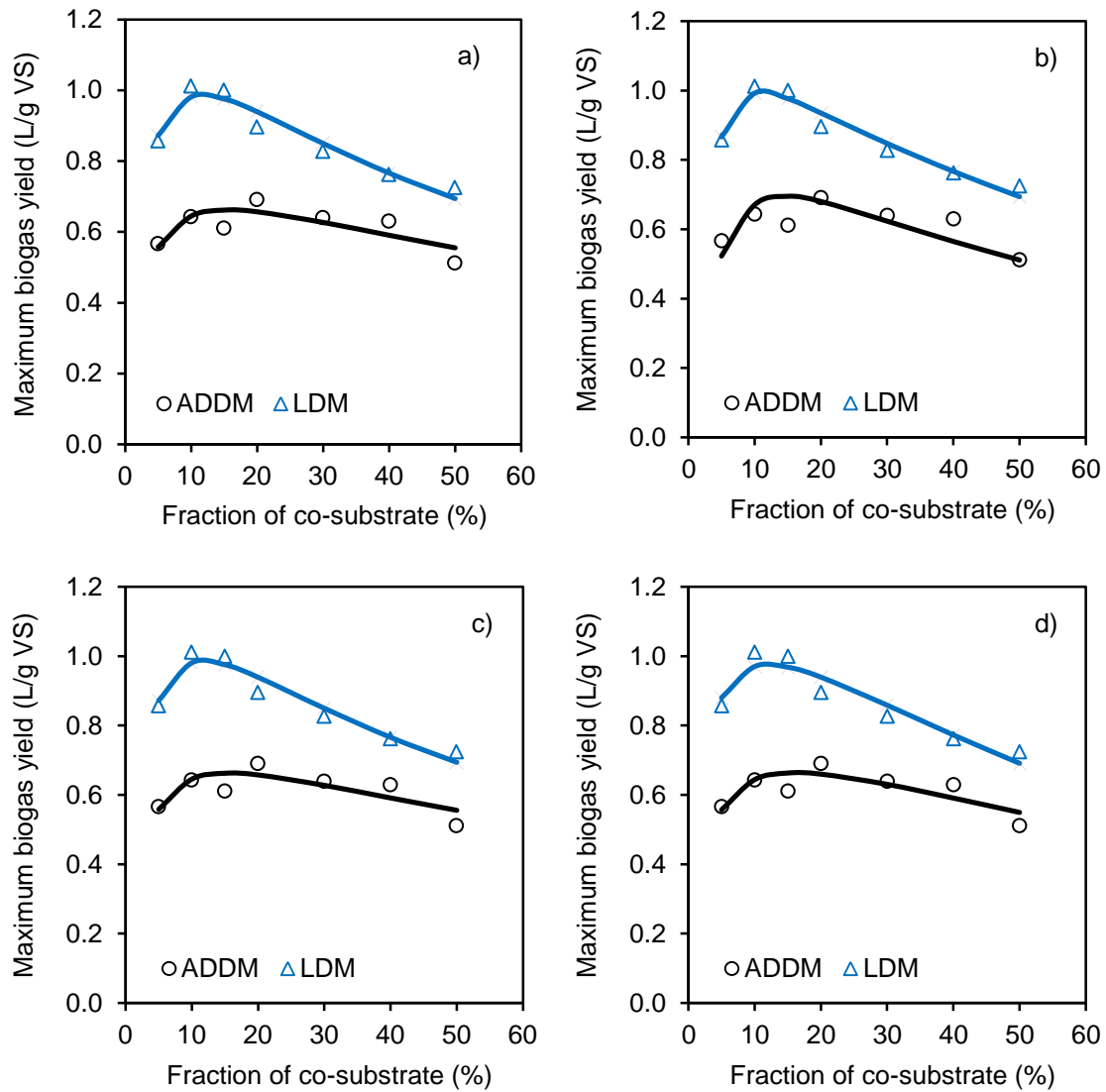


Fig. 3 Non-linear curve fitting of substrate limitation-inhibition kinetics using a) Andrews; b) Edwards; c) Haldane; and d) Aiba models for anaerobic co-digestion of foodwaste with anaerobically digested dairy manure (ADDM) and liquid dairy manure (LDM)

As shown in Table 3, all the models fitted well to the experimental data when LDM was used as co-substrate while the Edwards model fitted poorly relative to the other three models when ADDM was used as a co-substrate. Overall, the Aiba model is the best to simulate the inhibition of foodwaste digestion by ADDM and LDM. Both co-substrates had small K_s values and large K_i values. Fig. 3d shows that the maximum biogas yield increased as ADDM and LDM VS fractions were increased up to 16.3% and 12.0% respectively, then decreased due to inhibition. At the optimal co-substrate fractions, the maximum yield at standard temperature of 0 °C and pressure of 1 atm (stp) was 0.66 L (stp) biogas/g VS or 0.45 L (stp) CH₄/g VS with ADDM as co-substrate and approximately 0.97 L biogas (stp)/g VS or 0.66 L (stp) CH₄/g VS with LDM as co-substrate.

Table 3. Parameter values obtained from curve fitting of inhibition models to maximum biogas yield against fraction of co-substrate VS.

Parameter	Inhibition kinetic model			
	Andrews	Edwards	Haldane	Aiba
<i>Anaerobically digested dairy manure</i>				
q_m (L (stp)/g VS)	0.89	0.72	0.92	0.88
K_s (%)	2.7	3.2	2.8	2.6
K_i (%)	91.0	195	88.2	118
Correlation coefficient, r	0.80	0.75	0.80	0.82
<i>Liquid dairy manure</i>				
q_m (L (stp)/g VS)	1.48	1.13	1.59	1.32
K_s (%)	3.0	3.0	3.2	2.1
K_i (%)	46.4	104	43.2	82.2
Correlation coefficient, r	0.96	0.97	0.96	0.95

3.2 Digested versus Undigested Dairy Manure as Co-substrate

Although co-digestion of foodwaste with LDM yields more biogas and needs less co-substrate than co-digestion with LDM (Fig. 3), co-digestion with ADDM has an attractive biogas yield as well. Besides, it has less health and odor concerns when transporting ADDM instead of LDM for co-digestion at a foodwaste processing site. Moreover, the lower TS concentration makes ADDM a better co-substrate to dilute the thick foodwaste. Furthermore, farms are often more willing to provide ADDM than LDM as LDM has traditionally been land-applied as a liquid fertilizer.

Digestate is typically low in residual biomethane potential. As Table 1 shows, however, proteins are enriched after anaerobic digestion of LDM, generating relatively higher residual methane potential. Besides, ADDM has higher nutrient content compared with LDM and foodwaste (Table 1), supplementing macro- and micro-nutrients for co-digestion of foodwaste. Recent studies on anaerobic digestion of foodwaste have verified that the supplementation of trace elements such as selenium, cobalt and iron improves process stability and methane production [2, 18]. The inhibition at higher fractions of co-substrate, however, may be attributed to over-supplementation of elements such as Fe, Mn, Na and K. The batch experiments showed that ADDM had higher inhibition concentrations than LDM (Table 3), possibly because more trace elements are transformed to bioavailable forms by anaerobic digestion and enhance anaerobic digestion of foodwaste. For example, the initial concentrations of 55.1 mg/L Fe, 9.7 mg/L Mn, 1.11 g/L Na and 1.9 g/L K in the batch experiments at 50% ADDM were close to the upper limits suggested by Chen et al. [19] to prevent from inhibition.

3.3 Optimal Combination of Foodwaste and Dairy Manure

Earlier studies have compared biogas and methane yield in co-digestion of foodwaste and LDM at higher LDM fractions and the highest methane yields were attained at the lowest LDM fractions. Usack and Angenent [6] found the optimum VS ratio of LDM to be 51% among the tested fractions of 51-100%. Zarkadas et al. [7] found the optimum VS ratio of LDM to be 29.2% among the tested LDM fractions of 29.2-

100%. El-Mashad and Zhang [20] found a higher methane yield in batch co-digestion of foodwaste with 52% LDM than with 68% LDM. Through both batch digestion experiments and modeling, this study found that only a small fraction of LDM (12%) is needed for high-yield digestion of foodwaste.

When organic loading rate was increased stepwise from 1.0 to 2.0 g VS/L/d in the semi-continuous co-digestion at a substrate VS ratio of 88% foodwaste to 12% LDM, biogas production rate increased steadily to 1.78 L/L/d (Fig. 4a) with 68.0% methane in the biogas. Unfortunately, biogas production dropped to approximately 0.48 L/L/d due to an unexpected room temperature increase and subsequent biogas temperature jump on days 29-32 (Fig. 4b). In the last 6 d before this mishap, nevertheless, biogas yield reached 0.79 L (stp)/g VS and methane yield 0.54 L (stp)/g VS, which were already 81% of the simulated maximum yields despite the shorter hydraulic retention time. This methane yield estimate is higher than those reported by earlier co-digestion studies at higher LDM fractions. Usack and Angenent [6] reported a methane yield of 0.30 L/g VS at a foodwaste : LDM of 49% : 51%. El-Mashad and Zhang [20] reported a methane yield of 0.31 L/g VS at a foodwaste : LDM of 48% : 52%. Zarkadas et al. [7] reported a methane yield of 0.41 L/g VS at foodwaste : LDM of 70.8% : 29.2%.

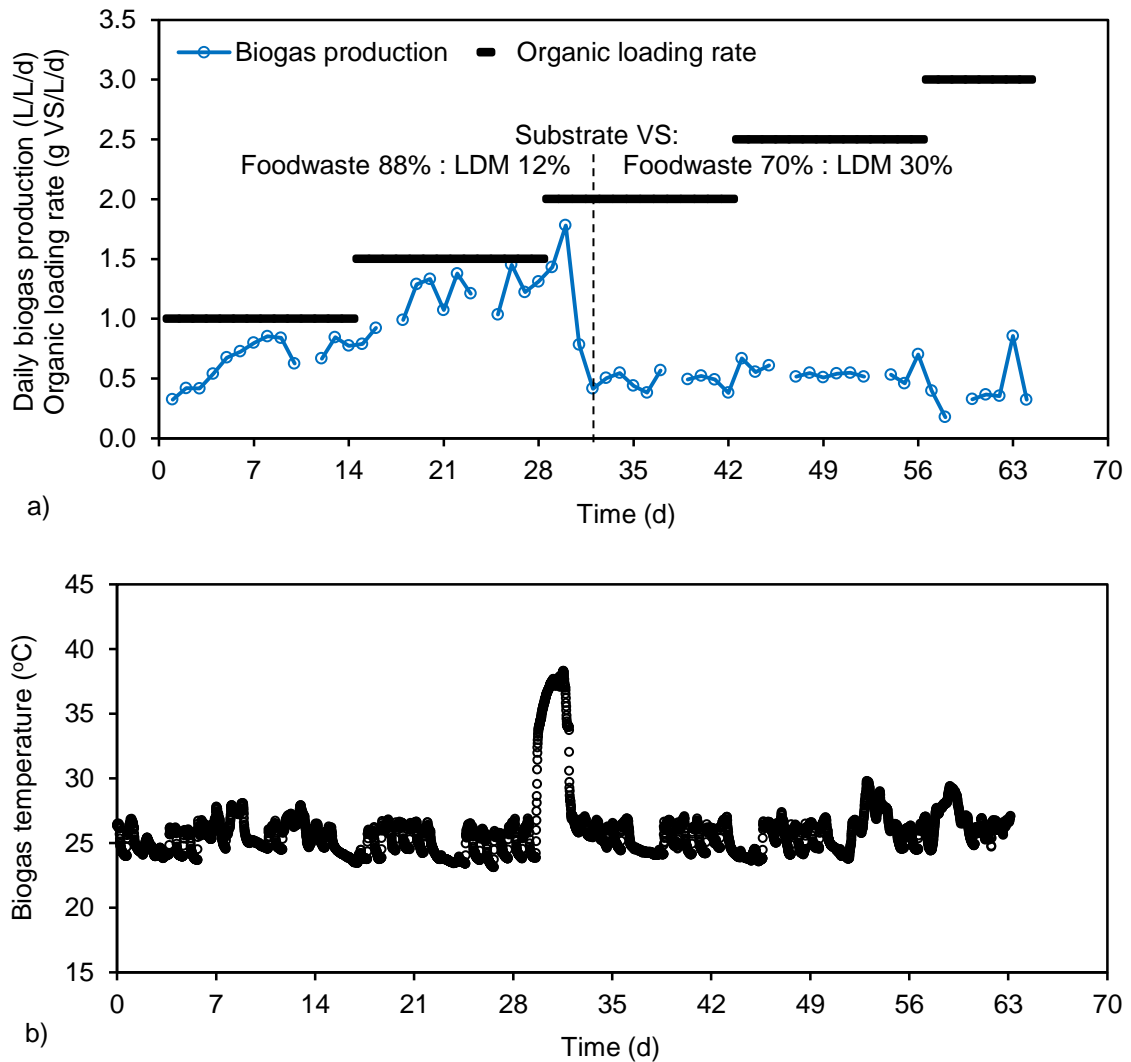


Fig. 4 Irreversible impact of a temperature pulse (b) on biogas production by anaerobic co-digestion of foodwaste and liquid dairy manure

This 2-d temperature shock resulted in increases in digestate TS and VS concentrations and a sharp decrease of digestate pH (Fig. 5). On day 32 calcium bicarbonate was added to adjust pH to above 7 and the substrate was changed to have more LDM (30%). Except for the temperature jump period, weekly measurements of digester effluent showed a small standard deviation ($0.6\text{ }^{\circ}\text{C}$) at a mean digestate temperature of $35.4\text{ }^{\circ}\text{C}$. Digestate TAN concentration was between 629 and 1226 mg/L, which were probably non-inhibitory. Although pH was adjusted to between 6.68 and 7.02 in the following 30 days, the biogas production rate was never recovered and TS and VS concentrations kept increasing. This type of incidents rarely occurs and has not been addressed, but indicated the influence of headspace temperature fluctuation on digestion stability.

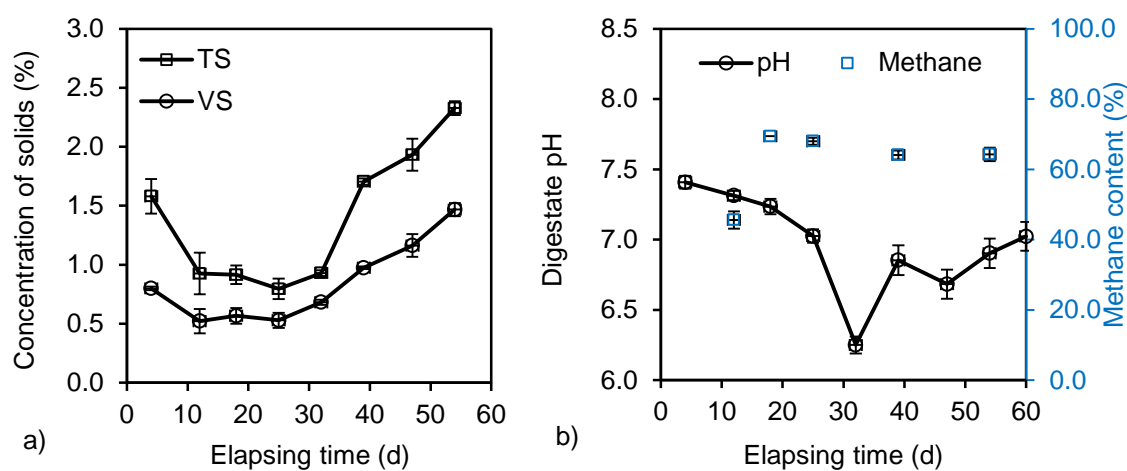


Fig. 5 Influences of a temperature pulse on days 29-32 and following recovery measures (increasing manure from 12% to 30%; pH adjustment) on mesophilic co-digestion of foodwaste and liquid dairy manure

4 CONCLUSIONS

A lower fraction of a co-substrate improves biogas production from anaerobic co-digestion of foodwaste. Overdosing of a co-substrate, however, may result in inhibition. The effect of co-substrate fraction on biogas yield could be simulated similarly with four substrate limitation – inhibition kinetic models. Biogas yield reached the highest when co-digesting with 16.3% ADDM or 12.0% LDM, which were lower than those reported by earlier studies at higher ranges of LDM fractions.

At the low fractions of a co-substrate (5-50%) in the batch co-digestion of foodwaste, the ADDM exerted greater inhibition or had a longer lag phase than LDM. However, the ADDM had higher half-inhibition concentrations (88-195% VS) than the LDM (43-104% VS).

Both LDM and ADDM are attractive co-substrates for anaerobic digestion of foodwaste. Semi-continuous co-digestion of 88% foodwaste and 12% liquid dairy manure at hydraulic retention time of only 14 d attained 81% of the simulated maximum biogas yield.

A short temperature jump in the digester headspace resulted in a sharp pH decrease and substantial decrease in biogas yield. The microbial response to a temperature shock from headspace needs to be investigated in the future.

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