

1 **Title:**

2 **Impact of low loading on digestion of the mechanically-separated organic fraction of**  
3 **municipal solid waste**

4

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14 **Abstract**

15 Changing waste management practice, introduction of new technologies, and population  
16 demographics and behaviour will impact on both quantity and composition of future waste  
17 streams. Laboratory-scale anaerobic digestion of the mechanically-separated organic fraction  
18 of municipal solid waste (ms-OFMSW) was carried out at relatively low organic loading  
19 rates (OLR), and results analysed using an energy modelling tool. Thermophilic operation  
20 with water addition and liquor recycle was compared to co-digestion with dilution water  
21 replaced by sewage sludge digestate (SSD); thermophilic and mesophilic mono-digestion  
22 were also tested at low OLR. All thermophilic conditions showed stable operation, with  
23 specific methane production (SMP) from 0.203-0.296 m<sup>3</sup> CH<sub>4</sub> kg<sup>-1</sup> volatile solids (VS). SSD  
24 addition increased biogas production by ~20% and there was evidence of further hydrolysis  
25 and degradation of the SSD. Long-term operation at 1 kg VS m<sup>-3</sup> day<sup>-1</sup> had no adverse effect  
26 except in mesophilic conditions where SMP was lower at 0.256 m<sup>3</sup> CH<sub>4</sub> kg<sup>-1</sup> VS and stability  
27 was reduced, especially during OLR increases. This was probably due to low total ammonia  
28 nitrogen, which stabilised at ~0.2 g N kg<sup>-1</sup> and limited the buffering capacity. Energy analysis  
29 showed thermophilic operation at OLR 2 g VS L<sup>-1</sup> day<sup>-1</sup> gave 42% of the theoretical methane  
30 potential and 38% of the higher heating value, reducing to 37% and 34% respectively in  
31 mesophilic conditions. Scenario modelling indicated that under low ms-OFMSW load even  
32 an energy-depleted co-substrate such as SSD could contribute to the energy balance, and  
33 would be a better diluent than water due to its nutrient and buffering capacity.

34

35 **Keywords:** OFMSW, mesophilic, thermophilic, sewage sludge digestate, ammonia, energy  
36 modelling

37

38

## 39 1 Introduction

40

41 Anaerobic digestion (AD) of the organic fraction of municipal solid waste (OFMSW) after  
42 mechanical separation is a well-established technology, and its performance characteristics  
43 and limitations are known both from long-term laboratory and pilot-scale research (Mata  
44 Alvarez, 2002; Hartmann and Ahring, 2006); and from studies of full-scale plant (Montejo et  
45 al., 2013; Romero-Güiza et al., 2014; Colón et al., 2017; Barati et al., 2017). By 2014 there  
46 were an estimated 244 AD plants in operation or under construction in Europe for which  
47 OFMSW made up a significant proportion of the feedstock (De Baere and Mattheeuws,  
48 2013). These plants are of many different types, and have evolved together with technical and  
49 engineering developments to accommodate the range of collection and separation systems  
50 used in preparation of this waste feedstock. Despite this extensive experience, there are  
51 circumstances in which the process may operate under loading conditions that lower than  
52 those it was designed for, or otherwise non-optimal. Lower loadings of course occur as a  
53 transient state during start-up, and studies have looked at the best strategies to address this  
54 (Bolzonella et al., 2003). A more important case is where the plant is oversized and the  
55 organic loading rate (OLR) is low due to a shortfall in the anticipated sources of waste: while  
56 such information is often commercially confidential, examination of the available data on  
57 digester sizes, input waste tonnages and combined heat and power (CHP) generation capacity  
58 suggests that not all plants are working at full capacity. This may be due to initial over-sizing,  
59 or to successful initiatives to increase recycling, of the type currently being adopted  
60 worldwide in a drive towards greater sustainability. While separation of recyclable materials  
61 may change the proportion of non-degradable components in the waste, however, there is  
62 evidence that it does not greatly alter the suitability of this residual stream as an AD  
63 feedstock (Waite, 2013; Fonoll et al, 2016).

64

65 A further factor that is likely to have an increasing impact in future is the adoption of  
66 alternative technologies for recovery of materials and energy from the general waste stream,  
67 which may leave reduced volumes of residual waste for digestion. These include waste-based  
68 biorefinery processes to recover selected fractions for conversion into a range of downstream  
69 building blocks, including alcohols (Farmanbordar, 2018; Mahmoodi et al., 2018), volatile  
70 fatty acids (Cavinato et al., 2017; Aguilar et al., 2013), and sugars and high value products  
71 (Vaurs et al., 2018; Sadhukhan et al., 2016). Further sources of competition for wastes may  
72 include thermal processing technologies for recovery of energy and products through  
73 gasification, pyrolysis and hydrothermal carbonisation (Yang et al., 2018; Dong et al., 2018).  
74 While many of these processes are still under development, and not all will make the  
75 transition to full-scale operation, it is clear there is potential for significant future competition  
76 for wastes. AD technologies are likely to retain a place at the core of such biorefineries for  
77 post-processing of residues; but, in addition to reduced tonnages, the characteristics of these  
78 residues will be significantly different from those of traditional mechanically-separated  
79 OFMSW (ms-OFMSW).

80

81 In some cases, a shortfall in ms-OFMSW may be addressed by seeking opportunities for co-  
82 digestion with complementary feedstocks such as wet wastes. Municipal wastewater  
83 biosolids have long been recognised as a promising co-substrate, and have been extensively  
84 studied (Hamzani et al., 1998; Sosnowski et al., 2003; Bolzonella et al., 2006; Silvestre et al.,  
85 2015). Few or no studies appear to have looked at co-digestion with sewage sludge digestate  
86 (SSD), however, probably because the majority of the readily degradable fraction is already  
87 likely to have been converted into biogas. This co-substrate may, however, be useful in

88 complementing the nutrient profile of the original feedstock, which is often cited as a reason  
89 for co-digestion (Tyagi et al., 2018).  
90

91 The current study aimed to consider a number of scenarios where ms-OFMSW digestion is  
92 carried out at relatively low OLR. One objective was to provide a baseline for thermophilic  
93 digestion of a particular ms-OFMSW feedstock, for comparison with alternative energy  
94 recovery options including integrated pyrolysis and anaerobic digestion (Yang et al., 2018b).  
95 Another was to simulate the effect of a lower OLR, such as might occur in an existing plant  
96 with a long-term shortfall in waste feedstock. The digester operating mode was based upon  
97 that used in a number of full-scale UK plants which run at a moderate OLR of around 2 kg  
98 volatile solids (VS)  $\text{m}^{-3} \text{day}^{-1}$ . Solids and liquid retention times in these plants are uncoupled  
99 by digestate separation and solids wasting, with solids typically retained for around 20-30  
100 days. Input to the digesters normally consists of around 14-15% OFMSW, 35-36% fresh  
101 water and 50% recycled digestate on a wet weight basis. This baseline scenario was  
102 compared with one where the waste input is reduced to 1 kg VS  $\text{m}^{-3} \text{day}^{-1}$  but the total input  
103 tonnage and hydraulic retention time (HRT) is maintained by addition of more water. For the  
104 lower OLR, the study also looked at substitution of the added fresh water with SSD to  
105 provide additional buffering and input of inoculum, plus some residual biogas potential. The  
106 trials were run under thermophilic conditions, but the lower OLR without SSD was also run  
107 at a mesophilic temperature to provide comparative data on the energy production potential.  
108 The impact of the different scenarios on the energy balance was considered using the ADAT  
109 modelling tool (BORRG, N.D). The results provide useful insights for operators of full-scale  
110 plant facing current or potential changes in ms-OFMSW feedstock availability, and on  
111 benefits that might arise from substrate blending with digestate from municipal wastewater  
112 biosolids treatment.

113

## 114 **2 Materials and methods**

### 115 2.1 Feedstock

116 Mechanically-separated OFMSW was obtained from the Bursom Recycling Centre, Leicester  
117 (Biffa Plc, UK). The residual mixed waste entering this plant is processed in a ball mill and  
118 then separated by a drum screen into two size fractions of 0-40 mm and 40-80 mm. The 0-40  
119 mm fraction goes through a slotted flip-flop screen to remove excess water and then through  
120 a 5 mm grid. The material is then placed in closed containers for transport to the Wanlip AD  
121 plant (Biffa Plc, UK). Two batches, each of ~300 kg, of this bulk material were passed  
122 through a 10 mm screen to remove any large non-organic contaminants that could damage the  
123 smaller-scale digestion equipment. The <10 mm fraction of the processed OFMSW was  
124 stored in 2-kg plastic bags at approximately -20 °C, and thawed for 24 hours at room  
125 temperature before use. Once defrosted, the OFMSW was maintained at 4 °C and used within  
126 5 days.

127

### 128 2.2 Digestion experiments

129 The trial used four continuous stirred-tank reactor (CSTR) digesters, each with an internal  
130 diameter of 0.32 m, a height of 0.55 m and a working volume of 35 L. Each digester was  
131 fitted with top and bottom flange plates, and was mixed by a mechanical stirrer connected  
132 through a gas-seal draught tube in the top plate connected to a geared motor operating at 35  
133 rpm. Fresh feed was added via a port in the top plate, and digestate removed from the bottom  
134 via a drain tube. Digesters were maintained at  $36 \pm 1$  °C (mesophilic) or  $55 \pm 1$  °C  
135 (thermophilic) by an internal heating coil. Biogas production from each digester was  
136 measured continuously using a tipping-bucket gas flow meter as described in Walker et al.  
137 (2009).

138

139 Experimental conditions are summarised in Table 1. The experimental period was divided  
140 into two stages as described below.

141

142 *Stage 1.* The trial used 2 digesters (T1 and T2) which had previously been operated  
143 thermophilically (55 °C) for a period of 345 days treating source segregated domestic food  
144 waste with side-stream ammonia removal at an OLR of 2 g VS L<sup>-1</sup> (Zhang et al., 2017). On  
145 day 0 the digestates from T1 and T2 were removed, mixed to ensure homogeneity and  
146 replaced in the digesters, then left without feeding for 12 days to allow degradation of  
147 residual feedstock. Feeding of T1 and T2 on OFMSW as the sole substrate started on day 13  
148 at an OLR of 0.5 kg VS m<sup>-3</sup> day<sup>-1</sup> and was increased to 1 kg VS m<sup>-3</sup> day<sup>-1</sup> on day 16. From  
149 day 71 onwards the operating mode was changed to include liquid addition, in accordance  
150 with typical operational practice at a number of large-scale plants in the UK. To simulate this,  
151 each day 1.666 kg of digestate was removed and the liquid and solids fractions were  
152 separated using a 1 mm nylon mesh sieve. The separated solids were discarded, giving a  
153 digester solids retention time (SRT) of 21 days. The digester received daily a wet weight of  
154 fresh OFMSW sufficient to give an OLR of 1 g VS L<sup>-1</sup> day<sup>-1</sup>, and 715 g of SSD from a  
155 mesophilic AD plant treating municipal wastewater biosolids (Millbrook, Southampton UK).  
156 Approximately 833 g of the separated digestate liquor was then returned to the digester to  
157 maintain the working volume, giving a nominal HRT of 42 days. T1 and T2 both received the  
158 same feed, and operated at a combined OLR (OFMSW + SSD) of ~1.7 kg VS m<sup>-3</sup> day<sup>-1</sup>.

159

160 *Stage 2.* In this part of the work the digesters were operated to simulate low loadings without  
161 co-digestion with SSD. On day 236 the addition of SSD to T1 and T2 as part of the feed was  
162 discontinued and 715 g of tap water substituted, reducing the OLR to 1 kg VS m<sup>-3</sup> day<sup>-1</sup>.

163 From day 385 the OLR was gradually increased over a 10-day period to  $2.0 \text{ g VS L}^{-1} \text{ day}^{-1}$ ,  
164 by increasing the amount of OFMSW added and decreasing the quantity of tap water  
165 proportionally. Feeding continued at this loading until the end of the experimental period on  
166 day 525.

167

168 A second pair of digesters (M1 and M2) was brought into operation on day 243 and  
169 inoculated with digestate taken from the Millbrook AD plant. These were operated  
170 mesophilically ( $37 \text{ }^\circ\text{C}$ ) at a HRT of 42 days and SRT of 21 days using the same solids/liquor  
171 separation method as for T1 and T2, with the aim of providing comparative data on  
172 thermophilic and mesophilic operation with the same feedstock and operating mode. From  
173 day 244 onwards M1 and M2 were fed on OFMSW, recycled liquor and tap water at an OLR  
174 of  $1.0 \text{ kg VS m}^{-3} \text{ day}^{-1}$ . Attempts to increase the OLR after day 385, as in T1 and T2, led to  
175 signs of instability. The loading on M1 and M2 thus had to be reduced, then was gradually  
176 restored to  $1.0 \text{ kg VS m}^{-3} \text{ day}^{-1}$  from day 479 until the end of the experimental period.

177

178 Trace element (TE) solutions were added to the digesters on two separate occasions to give the  
179 following additional concentrations in the digester ( $\text{mg L}^{-1}$ ): Al 0.1, B 1.0, Co 1.0, Cu 0.1, Fe  
180 5.0, Mn 1.0, Mo 0.2, Ni 1.0, Se 0.2, W 0.2, Zn 0.2 (Banks et al., 2012).

181

### 182 2.3 Analytical methods

183 Total solids (TS) and VS were measured according to Standard Methods 2540 G (APHA,  
184 2005). pH was determined using a Jenway 3010 meter (Bibby Scientific Ltd, UK) calibrated  
185 in buffers at pH 4.0, 7.0 and 9.2 (Fisher Scientific, UK). Alkalinity was measured by titration  
186 with  $0.25\text{N H}_2\text{SO}_4$  to endpoints of pH 5.75 and 4.3 (Ripley et al., 1986). Total Kjeldahl  
187 nitrogen (TKN) and total ammoniacal nitrogen (TAN) were determined using a Kjeldtech



188 block digestion and Büchi steam distillation unit according to the manufacturers' instructions  
189 (Foss Ltd, Warrington, UK). Crude protein content was calculated by multiplying the  
190 difference between TKN and TAN by 6.25 (Hansen et al., 1998). Samples for volatile fatty  
191 acid (VFA) analysis were prepared by centrifuging at 13600 g for 10 min and acidifying the  
192 centrifugate with 10% (v/v) formic acid. VFA in the supernatant was measured by gas  
193 chromatography (model GC-2010, Shimadzu, Tokyo, Japan), using a flame ionization  
194 detector and an FFAP capillary column (SGE Europe Ltd, UK) with helium as the carrier gas.  
195 Biogas composition was analysed using a Varian CP 3800 gas chromatograph with a gas  
196 sampling loop, with argon as the carrier gas. The GC was calibrated using a standard gas  
197 containing 35% CO<sub>2</sub> and 65% CH<sub>4</sub> (BOC, Guildford, UK). Elemental composition and  
198 higher heating values (HHV) were analysed as reported by Yang et al. (2018).

199

200 Digestate parameters such as TS, VS, VFA, TAN and alkalinity, as well as biogas  
201 composition, were analysed once per week. Gas volumes were measured using a weight-  
202 based gasometer and are reported corrected to standard temperature and pressure (STP) of 0  
203 °C, 101.325 kPa as described in Walker et al. (2009).

204

205 The theoretical HHV was calculated using the Dulong equation according to IFRF (2017).  
206 Theoretical methane potential (TMP) was calculated using the Buswell equation (Symons  
207 and Buswell, 1933) and the substrate elemental composition. VS destruction was estimated  
208 by assuming the weight of VS destroyed was equal to that of the biogas produced. Total VFA  
209 concentrations are expressed in terms of chemical oxygen demand (COD) based on the  
210 theoretical COD of individual VFA species measured.

211

212 2.4 Modelling

213 Selected scenarios were modelled using ADAT, a mass and energy balance modelling tool  
214 developed with support from various sources including the FP6 CROPGEN project (SES6-  
215 CT-2004-502824), RCUK RELU (RES-229-25-0022), FP7 VALORGAS (241334), BBSRC  
216 ADNet (BB/L013835/1) and IEA Bioenergy Task 37 (UK), which is freely available for  
217 download from <http://borrg.soton.ac.uk/resources/adat>. The modelling tool calculates the  
218 energy requirement for heating of digester inputs, any pre or post-pasteurisation requirements  
219 (as specified by the user), and heat losses through the digester floor, walls and roof. It takes  
220 into account the efficiency of the CHP, includes estimated parasitic energy demands for  
221 mixing and pumping, and allows for fugitive methane losses in the process. It can also  
222 include energy requirements for transport and pre and post-processing of feedstock and  
223 digestate fractions, energy offset savings from fertiliser substitution and embodied energy in  
224 the major plant components, but these were not considered in the current work. The model is  
225 written in C# and compiled with a user interface allowing input values to be modified.  
226 Default values in the modelling tool were used unless otherwise noted.

227

## 228 **3 Results**

229

### 230 3.1 Stage 1: thermophilic co-digestion with SSD

231 *Operating parameters:* The applied OLR and HRT over the 235 days of operation during  
232 stage 1 are shown in Figure 1a and b. In general there were no disturbances to the planned  
233 operating conditions.

234

235 *Gas production.* Volumetric and specific biogas and methane production and biogas methane  
236 content during stage 1 are shown in Figure 1c-f. Volumetric biogas production (VBP)  
237 stabilised at around  $0.56 \text{ L L}^{-1} \text{ day}^{-1}$  (Figure 1c), and gas composition remained steady at

238 around 59% CH<sub>4</sub> (Figure 1d). Over the first 70 days where there was mono-digestion of  
239 OFMSW the specific methane production (SMP) appeared to stabilise at 0.307 m<sup>3</sup> CH<sub>4</sub> kg<sup>-1</sup>  
240 VS. During the period of co-digestion with SSD from day 71 onwards the SMP fell to 0.203  
241 m<sup>3</sup> CH<sub>4</sub> kg<sup>-1</sup> VS (Figure 1e), while the total SMP based on the OFMSW input alone (Figure  
242 1f) showed only a slight increase to 0.345 m<sup>3</sup> CH<sub>4</sub> kg<sup>-1</sup> VS. This was mainly due to the low  
243 SMP of the SSD, although there may also have been some reduction in methane yield due to  
244 the shorter SRT and HRT.

245

246 *Operational stability.* pH in digesters T1 and T2 showed a small initial decline as the system  
247 acclimated to the change in feedstock followed by the increase in OLR (Figure 2a). By day  
248 50 the pH had stabilised, however, and remained close to 8.0 for the rest of stage 1, despite  
249 the change in operating mode from day 71. TAN concentrations also remained relatively  
250 steady at around 2.3 g N kg<sup>-1</sup> wet weight (WW) (Figure 2b). This is below the threshold for  
251 progressive VFA accumulation in thermophilic conditions, but close to the value where the  
252 first signs of mild instability may appear in the form of slightly elevated VFA concentrations  
253 (Yirong et al., 2017; Zhang et al., 2017a).

254

255 Total alkalinity (TA) in T1 and T2 dropped slightly with the initial change of feedstock, then  
256 fell more sharply as a result of the reduction in HRT from day 71 (Figure 2c). TA and partial  
257 alkalinity (PA) decreased to around 13.6 and 9.5 g CaCO<sub>3</sub> kg<sup>-1</sup> WW respectively (Figure 2d  
258 and e): the reduction in intermediate alkalinity (IA) was proportionally less, with the result  
259 that the IA/PA ratio in both digesters rose from an initial value of around 0.3 to an average of  
260 around 0.4-0.5 (Figure 2f). The absence of any sudden changes in IA/PA ratio indicates,  
261 however, that these were still stable operating conditions for this feedstock (Ripley et al.,  
262 1986). Total solids content in T1 and T2 rose after the new feedstock was introduced, then

263 fell from the introduction of solids wasting on day 71, and stabilised at around 6 % WW  
264 (Figure 2g). VS as a proportion of TS rose in the first 70 days (Figure 2h), reflecting the  
265 much lower biodegradability of the OFMSW feedstock compared to the source segregated  
266 food waste on which the digesters had previously been fed. By the end of stage 1 VS had  
267 stabilised at around 56% of TS, with an estimated overall VS destruction of ~42% based on  
268 biogas production.

269

270 Figure 2i and j show the total VFA concentrations in both digesters, and the individual VFA  
271 species in digester T2. In both T1 and T2 there was a brief peak in VFA associated with the  
272 initial change in feedstock, which reached around 6.5 g COD L<sup>-1</sup> and consisted mainly of  
273 acetic acid. This fell rapidly and for the rest of stage 1 VFA in T1 remained below 1 g COD  
274 L<sup>-1</sup>, though with continuing small fluctuations. VFA concentrations in T2 were slightly higher  
275 than in T1 and included 0.1-0.2 g L<sup>-1</sup> of propionic acid. A one-off dose of TE solution was  
276 added to both digesters on day 80, but did not appear to have any immediate effect, and  
277 minor fluctuations in VFA continued. Total VFA in T2 started to fall from around day 175  
278 and stabilised at below 1 g COD L<sup>-1</sup> by the end of stage 1 (Figure 2j). This small difference  
279 between the digesters was also reflected in the TAN and TA concentrations and the IA/PA  
280 ratio, which were slightly higher in T2 than T1 for most of this period.

281

282 Average values for gas production and stability parameters over the last 20 days of stage 1,  
283 after 3.9 HRT and 7.8 SRT in the same operating mode, are shown in Table 2 below.

284

### 285 3.2 Stage 2: Thermophilic and mesophilic digestion without SSD addition

286 *Operating parameters:* Figure 3a and b show the OLR and HRT applied during stage 2. For  
287 the thermophilic digesters there were no disturbances to the planned conditions, apart from an

288 accidental spill of around 3 kg of digestate from T1 on day 447. Over the next 8 days the  
289 daily OFMSW feed to T1 was reduced slightly in order to maintain an approximately  
290 constant OLR, and digestate removal was decreased until the digester reached its previous  
291 working volume, leading to a small temporary increase in HRT.

292

293 Feeding of the mesophilic digesters was adjusted in response to changes in the monitoring  
294 parameter values. Feeding of M1 followed the same pattern as in the thermophilic digesters  
295 until day 389, shortly after the start of the incremental rise in OLR. Feeding of M1 was  
296 stopped for 3 days, then an attempt was made to resume the OLR increase, but this was  
297 abandoned on day 398. M1 was left without feeding for 8 days, then the OLR was gradually  
298 stepped up between days 407-479 when it reached the previous value of 1 g VS L<sup>-1</sup> day. M2  
299 showed earlier and more severe signs of stress than M1, and reduced or intermittent feeding  
300 began on day 364 after 3 HRT under the new operating conditions. An attempt was made to  
301 increase the OLR in parallel with the other digesters between days 385-398, but this was  
302 abandoned and M2 was left without feeding for 11 days before the same stepped return to  
303 OLR 1 g VS L<sup>-1</sup> day was applied as in M1. These changes in OLR were reflected in the HRT,  
304 which rose to infinity in the periods without feeding (Figure 3b).

305

306 *Gas production.* VBP, SMP and biogas methane content during stage 2 are shown in Figure  
307 3c-e. In T1 and T2 following the switch from SSD to water addition the VBP fell, but  
308 stabilised after 5 days and reached an average of 0.490 L L<sup>-1</sup> day<sup>-1</sup> in the last 20 days before  
309 the OLR was increased from 1 to 2 g VS L<sup>-1</sup> day<sup>-1</sup>. VBP then increased in parallel with the  
310 rise in OLR between day 385-394. There was a small fall in VBP around day 450 for T1 and  
311 day 462 for T2: the reason for the former is unknown, while the latter was due to the  
312 reduction in feed associated with the digestate spill. Apart from day-to-day fluctuations VBP

313 in T1 and T2 then remained stable, reaching an average of  $0.999 \text{ L L}^{-1} \text{ day}^{-1}$  at the end of the  
314 experimental period. Gas composition in T1 and T2 remained steady at around 59%  $\text{CH}_4$   
315 (Figure 3d). SMP was closely similar in both digesters and averaged  $0.290 \text{ L CH}_4 \text{ g}^{-1} \text{ VS}$  in  
316 the last 20 days before the OLR increase, and  $0.296 \text{ L CH}_4 \text{ g}^{-1} \text{ VS}$  at the end of the run at  
317 OLR  $2 \text{ g VS L}^{-1} \text{ day}^{-1}$ .

318

319 VBP in the mesophilic digesters at OLR  $1 \text{ g VS L}^{-1} \text{ day}^{-1}$  was slightly lower than in the  
320 thermophilic (Figure 3c), with an average value of  $0.452 \text{ L L}^{-1} \text{ day}^{-1}$  between day 343-362.  
321 From day 363 the OLR on M2 was reduced, with a corresponding fall in VBP, while M1  
322 showed a slight downward trend in VBP despite the constant OLR. Subsequent variations in  
323 VBP reflected the applied loading rates, with VBP finally recovering to around its previous  
324 value once the OLR was restored to  $1 \text{ g VS L}^{-1} \text{ day}^{-1}$ . Gas composition was relatively stable  
325 apart from a dip in  $\text{CH}_4$  content around day 370 for M2 and a peak around day 407 in both  
326 M1 and M2 (Figure 3d). SMP averaged  $0.267 \text{ L CH}_4 \text{ g}^{-1} \text{ VS}$  for the two digesters between  
327 day 344-363 and  $0.263 \text{ L CH}_4 \text{ g}^{-1} \text{ VS}$  in the last 20 days of the run.

328

329 *Operational stability.* Figure 4 presents the values for monitoring parameters during stage 2.  
330 TAN concentrations in both sets of digesters initially fell as ammonia from the inoculum in  
331 M1 and M2 and from the SSD addition in T1 and T2 was progressively washed out (Figure  
332 4a). After 3 HRT under the new operating conditions, TAN had stabilised and was slightly  
333 higher in the thermophilic digesters at an average of  $0.42 \text{ g N kg}^{-1} \text{ WW}$  for day 365-384 than  
334 in M1 at  $0.29 \text{ g N kg}^{-1} \text{ WW}$ . Feeding of M2 had altered after day 363 due to signs of  
335 instability, but until then it showed a closely similar trend to M1, while the slight increase in  
336 TAN in M2 after day 363 may indicate some associated biomass die-off and lysis.

337

338 TA and PA values mirrored the fall in TAN (Figure 4b-d), with the TA stabilising at around  
339 3.8 and 3.6 and PA at 2.8 and 2.3 g CaCO<sub>3</sub> kg<sup>-1</sup> WW in the thermophilic and mesophilic  
340 digesters respectively. After some minor fluctuations the IA/PA ratio settled at around 0.3-0.4  
341 in all digesters (Figure 4e). The pH in both sets of digesters also gradually fell in response to  
342 the changing TAN, but from different start points and to different degrees (Figure 4f). In T1  
343 and T2 the initial pH was just below 8.0, reflecting the higher initial TAN content of around  
344 2.2 g N kg<sup>-1</sup> WW. After 3 HRT, this appeared to be stabilising at pH 7.4. The initial pH in  
345 M1 and M2 was around 7.4 with a TAN content of 1.8 g N kg<sup>-1</sup> WW and this stabilised at just  
346 above 7.0.

347

348 The main difference in digester operational stability parameters was in the VFA  
349 concentrations. T1 and T2 showed initial peaks in total VFA of up to 1.3 g COD L<sup>-1</sup>  
350 immediately after the cessation of SSD addition. These declined to < 0.1 g COD L<sup>-1</sup> by day  
351 300 and remained very low throughout the rest of the trial, apart from a brief increase around  
352 day 363. These initial peaks were primarily acetic acid but also contained up to 0.5 g COD L<sup>-1</sup>  
353 of propionic acid and small amounts of longer chain VFA. In contrast the mesophilic  
354 digesters had total VFA of < 0.1 g COD L<sup>-1</sup> until day 363 when concentrations rose,  
355 especially in M2. It was thought this could be caused by washout of essential TE after 3  
356 HRT; and as a precautionary measure on day 368 all four digesters were given a one-off dose  
357 of TE solution as described above. Total VFA concentrations in M2 fell to < 0.1 g COD L<sup>-1</sup>  
358 by day 377 and values in the other digesters reduced even further. The attempt at raising the  
359 OLR in M1 and M2 led to another increase in VFA concentrations, however, reaching 1.5 g  
360 COD L<sup>-1</sup> in M2 and consisting almost entirely of acetic acid. VFA concentrations finally  
361 stabilised at very low levels after day 475 once the OLR was returned to 1 g VS L<sup>-1</sup> day<sup>-1</sup>.

362 Although the trial continued for a further 3.8 HRT after the one-off TE dosing there were no  
363 further TE additions and no sign of reappearance of VFA.

364

365 TS in both sets of digesters fell from the start of stage 2 (Figure 4g), stabilising at around  
366 2.1 % in T1 and T2 and 2.2 % in M1 and M2 after 3 HRT, with VS/TS ratios of 57.9 and  
367 59.7% respectively. After the increase in OLR in T1 and T2 there was a clear rise in TS  
368 content but a decline in VS/TS ratio to around 52%. Estimated VS destructions based on gas  
369 production were around 60% for the thermophilic digesters and 53% for mesophilic.

370

371 Table 2 shows average values for gas production and operational stability parameters over the  
372 last 20 days before the OLR increase from 1 to 2 g VS L<sup>-1</sup> day<sup>-1</sup>, and before the end of the  
373 experimental period. These periods correspond to 3.5 HRT and 7.0 SRT and to 3.1 HRT and  
374 6.2 SRT respectively in each operating mode.

375

### 376 3.3 Energy and performance considerations

377 Characteristics for Batch 1 of the feedstock shown in Table 3, with typical values for  
378 mechanically-sorted OFMSW from a 'medium complex' plant (Cecchi et al., 2003) and those  
379 previously reported for waste from the same source (Zhang et al., 2012). The TS and VS  
380 values given in Table 3 are the average from the experimental period, with a relative standard  
381 deviation of approximately 5% in each case.

382

383 Table 3 also shows the calculated TMP and theoretical and measured HHV values for the  
384 OFMSW feedstock based on elemental composition, with values from Zhang et al. (2012) for  
385 comparison. Measured and theoretical HHV show good agreement, providing support for the  
386 elemental analysis results. The biogas methane content of 60.0% predicted by the Buswell



387 equation is close to the observed average value of around 59.3%. The SMP of 0.290 L CH<sub>4</sub> g<sup>-1</sup>  
388 VS achieved in thermophilic operation at OLR 1 g VS L<sup>-1</sup> day<sup>-1</sup> represents conversion of  
389 42% of the TMP and 38% of the measured HHV. Equivalent conversion values for the SMP  
390 of 0.256 L CH<sub>4</sub> g<sup>-1</sup> VS in mesophilic conditions are 37% and 34%. The methane yield of the  
391 OFMSW on a wet weight basis was 86 and 77 m<sup>3</sup> CH<sub>4</sub> tonne<sup>-1</sup> in thermophilic and mesophilic  
392 conditions respectively.

393

394 Zhang et al. (2012) reported a lower TMP and HHV, a higher mesophilic SMP of 0.304 L  
395 CH<sub>4</sub> g<sup>-1</sup> VS, and thus higher conversion rates of 55% TMP and 55% HHV for material from  
396 the same plant. There were no known changes in on-site processing between the two trials;  
397 but Zhang et al. (2012) included an additional in-house pre-processing step in which small  
398 pieces of plastic and glass were manually picked out before feeding to the digesters. In  
399 addition to natural variation between batches and over time, this may account for the lower  
400 VS content and VS/TS ratio in the current feedstock (Table 3). Removal of small fragments  
401 of plastic will lead to an apparent increase in biogas and methane yield on a VS basis, which  
402 may partially account for the slightly higher SMP. On the other hand removal of plastics will  
403 reduce the measured and theoretical TMP and HHV, and may account for the lower values of  
404 these parameters compared to the current feedstock. Removal of inert materials (glass and  
405 metal fragments) will also slightly increase the SMP on a TS basis; while removal of both  
406 inerts and plastics may increase or reduce the HHV on a TS basis depending on the relative  
407 proportions removed. It is likely that at least part of the difference in conversion is due to the  
408 additional pre-processing by Zhang et al. (2012). Specific methane production on a wet  
409 weight basis was 20% lower in the current study, suggesting that there may have been some  
410 reduction in feedstock VS content and biodegradability between the two trials, perhaps due to  
411 increasing popularity of source separated collections for domestic food waste. This is also

412 supported by the differences in measured and predicted biogas methane content in each case.  
413 The SRT of 21 days used in the current study was shorter than the 30 days in Zhang et al.  
414 (2012), and this may also have accounted for some of the difference. Zhang et al. (2012)  
415 measured the BMP of the feedstock at inoculum-to-substrate (i/s) ratios of 2:1 and 4:1 on a  
416 VS basis. At i/s 2:1 the methane production at 21 days was only 73% of the value at 28 days  
417 and 69% of the final BMP value, but at 4:1 it was respectively 97% and 95%.

418

419 In the current trial the SMP in thermophilic conditions was higher than in mesophilic (0.290  
420 and 0.256 L CH<sub>4</sub> g<sup>-1</sup> VS respectively at OLR 1 g VS L<sup>-1</sup> day<sup>-1</sup>). It is often stated that  
421 thermophilic digestion can give higher gas production, especially for feedstocks with  
422 relatively high lignocellulosic content. In some cases this is simply an assertion without  
423 supporting data, while in others insufficient detail is given on how or whether gas volumes  
424 have been normalised to STP: measurement at 35 or 55 °C gives a 6% difference in gas  
425 volume. In some technical trials where appropriate datasets are provided, significant  
426 improvements have been demonstrated (Cecchi et al., 1991; Fernández-Rodríguez et al.,  
427 2013). In the current work there is reasonable confidence in the data, as the gas counters used  
428 were calibrated approximately weekly against the collected volume of gas at a measured  
429 room temperature and pressure. Moisture content can also account for 5-6% of gas volume at  
430 35 °C and 15-16 % at 55 °C; but in the method used any associated errors should be small as  
431 the gas bags were allowed to equilibrate to room temperature before measurement. It is  
432 therefore likely that there was a real difference in SMP at the two temperatures: while the  
433 nature of the digested material makes it difficult to obtain very accurate and consistent VS  
434 concentrations, a small difference in the VS/TS ratios (Table 2) may also support this. To  
435 further reduce small day-to-day differences in gas volume measurement, such as those seen

436 on days 322 and 331, direct correction of gas counter data by continuous logging of ambient  
437 temperature and pressure would be beneficial.

438

439 The SMP in thermophilic conditions at OLR 2 g VS L<sup>-1</sup> day<sup>-1</sup> was closely similar to that at 1  
440 g VS L<sup>-1</sup> day<sup>-1</sup>, at 0.296 and 0.290 L CH<sub>4</sub> g<sup>-1</sup> VS respectively, indicating that the thermophilic  
441 digesters operated well at the lower OLR despite the lower TAN and alkalinity (Table 2). In  
442 mesophilic conditions, however, there were signs of reduced stability at the lower OLR, with  
443 IA/PA ratios of around 1 by the end of the experimental period (Table 2), and the appearance  
444 of VFA during periods of moderate loading increase (Figure 4h). This was probably linked to  
445 the low TAN concentration and associated reduction in buffering capacity in the mesophilic  
446 digesters (Figure 4a). The low TAN may possibly indicate a slightly lower degree of  
447 hydrolysis in mesophilic conditions. TAN concentrations in the thermophilic digesters rose  
448 from 0.42 to 0.75 g N kg<sup>-1</sup> WW when the OLR was raised from 1 to 2 g VS L<sup>-1</sup> day<sup>-1</sup>, with  
449 the resultant increases in pH, TA and IA/PA ratio all suggesting improved operational  
450 stability. Zhang et al. (2012) reported stable operation at a TAN concentration of 1.4 g N kg<sup>-1</sup>  
451 WW with total VFA concentrations < 0.1 g L<sup>-1</sup> and a pH of 7.5 in mesophilic conditions. The  
452 higher TAN concentration is due to the higher OLR of 2 g VS L<sup>-1</sup> day<sup>-1</sup> but also to recycling  
453 of digestate liquor only, unlike the current trial where a proportion of water was added to  
454 simulate the operation of a number of full-scale UK AD plants working on this type of  
455 feedstock. Where digester TAN concentrations are low in this mode of operation, for  
456 example due to a reduction in OLR, increasing the digestate recycle and reducing water input  
457 may offer a simple means of improving stability and performance.

458

459 At an OFMSW OLR of 1 g VS L<sup>-1</sup> day<sup>-1</sup>, the addition of SSD reduced the SMP but gave a  
460 higher VBP (Table 2). If it is assumed that the SMP of the OFMSW fraction at this OLR was

461 constant with or without SSD, the estimated SMP of the SSD is around  $0.078 \text{ L CH}_4 \text{ g}^{-1} \text{ VS}$ .  
462 This is slightly higher than the value of  $0.04\text{-}0.06 \text{ L CH}_4 \text{ g}^{-1} \text{ VS}$  typically found for this  
463 digestate when used as inoculum in long-term mesophilic BMP tests (unpublished data,  
464 University of Southampton), indicating that some additional hydrolysis may be occurring at  
465 the higher temperature. Alternatively the increased buffering capacity and addition of  
466 inoculum and trace elements in the SSD may lead to a slight increase in the OFMSW SMP;  
467 in either case, the overall methane productivity of the system is increased. The proportion of  
468 SSD added was sufficient to maintain the TAN concentration just below that associated with  
469 the onset of instability in thermophilic conditions. SSD addition might also have been  
470 beneficial to stability in mesophilic digestion, especially at the lower OLR, but this was not  
471 tested in the current work.

472

### 473 3.4 Modelling

474 The scenarios considered were those tested in the laboratory trials, i.e. thermophilic operation  
475 at OLR  $1 \text{ kg VS m}^{-3} \text{ day}^{-1}$  with and without SSD addition (TH1 and TH1+SSD), thermophilic  
476 operation at OLR  $2 \text{ kg VS m}^{-3} \text{ day}^{-1}$  without SSD addition (TH2), and mesophilic operation at  
477 OLR  $1 \text{ kg VS m}^{-3} \text{ day}^{-1}$  without SSD addition (ME1). User-defined feedstock properties were  
478 based on average properties for the OFMSW used (Table 3) and on steady state gas  
479 production and composition data in Table 2. Feedstock parasitic energy demand was taken as  
480  $40$  and  $10 \text{ kWh tonne}^{-1}$  for OFMSW and SSD respectively, based on the model's default  
481 values for mechanically-recovered biodegradable municipal waste and sewage sludge,  
482 respectively.

483

484 The working volume of the digester was set at  $3439 \text{ m}^3$  to give an OLR of  $2 \text{ kg VS m}^{-3} \text{ day}^{-1}$   
485 for an assumed OFMSW input of  $8500 \text{ tonnes WW year}^{-1}$ . Digester construction was taken as

486 a steel tank with 75 mm of high performance insulation (typical U-value  $0.0245 \text{ W m}^{-1} \text{ K}^{-1}$ ,  
487 giving a composite heat transfer coefficient of  $0.35 \text{ W m}^{-2} \text{ K}^{-1}$ ), with gas stored in a separate  
488 gas holder. Digestion temperatures were set at  $55 \text{ }^\circ\text{C}$  for thermophilic operation and  $37 \text{ }^\circ\text{C}$  for  
489 mesophilic, with ambient conditions based on Southampton, UK (annual average air and soil  
490 temperatures  $10.6 \text{ }^\circ\text{C}$ ). In thermophilic conditions no separate pasteurisation step was  
491 included, since compliance with the Animal By-product Regulations (EC 1069/2009) may be  
492 achieved by providing a guaranteed HRT, e.g. through intermittent feeding and discharge. In  
493 mesophilic conditions pre-pasteurisation of the OFMSW at  $70 \text{ }^\circ\text{C}$  for one hour was specified;  
494 based on earlier work this was assumed not to affect the methane yield of the OFMSW  
495 (Banks and Zhang, 2010). It was assumed that the biogas produced was used on site in a CHP  
496 plant with an electrical conversion efficiency of 38% and heat recovery of 47% of input  
497 biogas energy. Fugitive emissions were set at 0.5%.

498

499 Table 4 shows the output from the modelling. Total biogas and methane production are  
500 determined by the experimental SMP values: as expected, total energy output followed the  
501 same sequence, being highest for TH2, TH1+SSD, TH1 then ME1. All scenarios show a  
502 positive energy balance, even when waste input is halved in this operating mode.

503

504 The heat demand of the AD process is the same for all three thermophilic scenarios, but  
505 considerably higher for the ME1 scenario, due to the requirement for pre-pasteurisation. The  
506 relative proportions of the raw biogas energy required for heating thus show a wide range,  
507 from 3.6% for TH2 to 13.7% for ME1. Very small differences in the AD plant parasitic  
508 electricity demand are due to the degree of solids breakdown in each scenario. The model  
509 calculates the VS destruction based on biogas production and adjusts the amount of digestate

510 accordingly; since the energy demand for centrifugation is based on the tonnage of digestate,  
511 this leads to a small reduction in scenarios with higher biogas yield.

512

513 Reducing the OLR from 2 to 1 kg VS m<sup>-3</sup> day<sup>-1</sup> halves the required CHP generation capacity,  
514 but adding SSD recovers some of this. For larger AD plants with multiple digesters and CHP  
515 generators the simplest response to a major fall in feedstock tonnages is to close one or more  
516 units down: but where there is a single line this option may not be practicable. The  
517 experimental and modelling results both suggest that under thermophilic conditions the plant  
518 could still operate with a positive energy balance at a substantially reduced loading; although  
519 this may not be economically attractive, particularly if a proportion of the income stream is  
520 obtained from gate fees. The results show clearly that the worst option would be to drop the  
521 operating temperature of the digester as this would reduce the biogas yield, and make the  
522 plant more susceptible to pH fluctuations as a result of loss of buffering capacity and a  
523 tendency for VFA accumulation in response to load variations. The scenario of mesophilic  
524 co-digestion with SSD was not tested experimentally, but could provide a solution for  
525 instability resulting from the loss of buffering, although the additional biogas production in  
526 mesophilic conditions is likely to be lower. Addition of SSD in thermophilic conditions gives  
527 a 48% increase in electricity available for export and a 22% increase in heat compared to the  
528 equivalent output for water addition.

529

530 The results for the scenarios tested were as expected, confirming the accuracy of the  
531 modelling with respect to the growing knowledge base gained from operation of OFMSW  
532 digesters over a number of years. The model does, however, provide a flexible tool for testing  
533 different scenarios at scale and can be used both to provide baseline values for comparison  
534 with alternative process options; and to assess the potential for integration of AD with other

535 emerging waste management options and technologies. An example where further process  
536 integration could be applied is in the use of waste heat in drying OFMSW as pyrolysis  
537 feedstock. The model cannot, however, predict some of the unforeseen outcomes of such  
538 integration: as noted by Yang et al. (2018) the pyrolysis of OFMSW at higher feedstock  
539 solids concentrations is associated with greater toxicity of the aqueous fraction when used as  
540 an AD feedstock.

541

542 One potential benefit of using co-digestates such as SSD is the increased nutrient content and  
543 fertiliser value of the resulting digestate. In countries such as the UK, where digestate from  
544 municipal solid waste (MSW) can still be applied to industrial crops, the enhanced nitrogen  
545 content from the addition of SSD could in theory be beneficial. This route, however, is very  
546 likely to be closed off in the near future due to growing environmental concerns over issues  
547 such as microplastics, endocrine disrupters and other priority pollutants; in many countries  
548 land application of this material is already specifically prohibited. Thermal processing of the  
549 digestate is therefore the most likely option for final disposal of residues from MSW  
550 digestion in future. Where SSD and residual OFMSW both contain heavy metals and other  
551 contaminants making them unsuitable for land application, the case for co-digestion is  
552 potentially attractive, as the increase in total energy output from the CHP plant can provide a  
553 heat source for pre-drying the centrifugate solids. This would reduce the energy input  
554 required for incineration or gasification of digestate from these mixed contaminated sources.  
555 The model allows inclusion of a value for post-processing energy per tonne of digestate, and  
556 this could be explored further using experimental data from dewatering; but this was not  
557 included in the current work.

558

559 **4 Conclusions**

560

561 Thermophilic CSTR digesters fed on mechanically-separated OFMSW at a moderate OLR of  
562  $2 \text{ kg VS m}^{-3} \text{ day}^{-1}$  and operated to simulate a full-scale plant with combined digestate  
563 recycling and water addition showed stable operation with a specific methane production of  
564  $0.296 \text{ m}^3 \text{ CH}_4 \text{ kg}^{-1} \text{ VS}$ , equivalent to  $87 \text{ m}^3 \text{ CH}_4$  per tonne of waste input. VS destruction was  
565 estimated at 82% based on the weight of biogas produced. Operating in the same mode but at  
566 a lower OLR of  $1 \text{ kg VS m}^{-3} \text{ day}^{-1}$  and constant HRT had no adverse effect on performance,  
567 with specific methane production of  $0.290 \text{ m}^3 \text{ CH}_4 \text{ kg}^{-1} \text{ VS}$  ( $86 \text{ m}^3 \text{ CH}_4 \text{ tonne}^{-1}$  OFMSW).  
568 Under mesophilic conditions at OLR  $1 \text{ kg VS m}^{-3} \text{ day}^{-1}$  with the same operating mode,  
569 however, the specific methane yield was lower at  $0.256 \text{ m}^3 \text{ CH}_4 \text{ kg}^{-1} \text{ VS}$  ( $77 \text{ m}^3 \text{ CH}_4 \text{ tonne}^{-1}$   
570 OFMSW); and signs of reduced operational stability were seen especially when at  
571 incremental increases in OLR were attempted. This was probably due to the low TAN  
572 concentrations, which stabilised at around  $0.2 \text{ g N kg}^{-1} \text{ WW}$  and led to limited digester  
573 buffering capacity. In practice if the OLR in such a plant is to be reduced steps should be  
574 taken to maintain TAN and buffering capacity, e.g. by reducing water addition and allowing  
575 an increase in HRT.

576

577 When the water was replaced by addition of municipal wastewater biosolids digestate (SSD)  
578 at OLR  $1 \text{ kg VS m}^{-3} \text{ day}^{-1}$  in thermophilic conditions, volumetric biogas and methane yields  
579 increased by around 20%. This may have been due in part to the addition of supplementary  
580 inoculum and trace elements, but a more likely explanation is the further hydrolysis and  
581 degradation of the mesophilically-digested biosolids. Modelling indicated that addition of  
582 SSD in thermophilic conditions gave a 48% increase in electricity available for export (from  
583  $0.45$  to  $0.67 \text{ GJ tonne}^{-1}$  OFMSW) and a 22% increase in heat (from  $1.21$  to  $1.48 \text{ GJ tonne}^{-1}$   
584 OFMSW) compared to the equivalent output for water addition.



585

586 If transport and handling costs permit, addition of SSD may be an alternative to fresh water  
587 inputs, but it is important to maintain the digester TAN concentration below the threshold  
588 value of around 2.5 g N kg<sup>-1</sup> WW of digestate to avoid the onset of inhibition and intermittent  
589 or progressive VFA accumulation.

590

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596

### 597 **Data accessibility statement**

598 The datasets generated during and/or analysed during the current study are available from the  
599 corresponding author on reasonable request.

600

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719

720 **Table 1** Experimental conditions

		Days	OLR kg VS m <sup>-3</sup> day <sup>-1</sup>	HRT days	SRT days	Scenario name
Thermophilic	day 0-15	16	Variable	variable	equal to HRT	-
T1 & T2	day 16-70	55	1.0 OFMSW	approx 297	equal to HRT	-
	day 71-235	165	1.0 OFMSW, 0.7 SSD	42	21	TH1+SSD
	day 236-384	149	1.0 OFMSW	42	21	TH1
	day 385-393	9	Increasing	Falling	21	-
	day 394-525	132	2.0 OFMSW	42	21	TH2
Mesophilic	day 244-384	141	1.0 OFMSW	42	21	ME1
M1 & M2	day 385-478	94	Variable - increasing	42	21	-
	day 479-525	47	1.0 OFMSW	42	21	-

721 Note: Stage 1 refers to days 0-235, Stage 2 to days 236-525. OLR is shown as value in kg VS m<sup>-3</sup> day<sup>-1</sup> followed

722 by feedstock type: with two feedstock the total OLR is the sum of both values.

723

724 **Table 2** Average steady-state values for digestion monitoring parameters

	Unit	TH1+SS <sup>a, b</sup>	TH1 <sup>a, c</sup>	TH2 <sup>a, d</sup>	ME1 <sup>c, e</sup>	ME1 <sup>a, d, f</sup>
Temp	°C	55	55	55	37	37
MSW	kg VS m <sup>-3</sup> day <sup>-1</sup>	1.0	1.0	2.0	1.0	1.0
SS dig	kg VS m <sup>-3</sup> day <sup>-1</sup>	0.7	0.0	0.0	0.0	0.0
VBP	L L <sup>-1</sup> day <sup>-1</sup>	0.583 ± 0.005	0.490 ± 0.003	0.999 ± 0.000	0.432	0.457 ± 0.007
CH4	% vol	59.3 ± 0.2	59.3 ± 0.3	59.3 ± 0.0	59.2	59.1 ± 0.1
SMP	L CH <sub>4</sub> g <sup>-1</sup> VS	0.203 ± 0.002 <sup>g</sup>	0.290 ± 0.000	0.296 ± 0.000	0.256	0.263 ± 0.002
pH	-	7.98 ± 0.02	7.44 ± 0.01	7.57 ± 0.01	7.04	6.94 ± 0.00
TA	g CaCO <sub>3</sub> L <sup>-1</sup>	13.9 ± 0.0	3.8 ± 0.0	8.2 ± 0.04	3.6	2.9 ± 0.2
PA	g CaCO <sub>3</sub> L <sup>-1</sup>	9.6 ± 0.2	2.8 ± 0.0	5.1 ± 0.5	2.6	1.5 ± 0.0
IA	g CaCO <sub>3</sub> L <sup>-1</sup>	4.3 ± 0.2	1.0 ± 0.0	3.0 ± 0.1	1.1	1.4 ± 0.2
IA/PA	-	2.8 ± 0.0	0.4 ± 0.0	0.4 ± 0.0	0.4	0.9 ± 0.1
TAN	g N kg <sup>-1</sup> WW	2.27 ± 0.0	0.42 ± 0.0	0.75 ± 0.0	0.29	0.19 ± 0.0
VFA	mg COD L <sup>-1</sup>	532 ± 82	20 ± 0.3	21 ± 0.8	92	22 ± 0.3
TS	%WW	5.8 ± 0.1	2.1 ± 0.1	4.3 ± 0.0	2.2	2.2 ± 0.0
VS	%WW	3.2 ± 0.1	1.2 ± 0.1	2.2 ± 0.0	1.3	1.3 ± 0.0
VS	%TS	55.8 ± 0.9	57.9 ± 0.7	52.4 ± 0.6	59.7	57.7 ± 1.9
VS destruction <sup>g</sup>	%VS	42%	60%	61%	53%	56%

725 <sup>a</sup> Values shown as ± are range of averages for 2 digesters; <sup>b</sup> average day 215-234, i.e. end stage 1; <sup>c</sup> average day  
726 365-384, i.e. before OLR step; <sup>d</sup> average day 505-524, i.e. end stage 2; <sup>e</sup> values for M1 only; <sup>f</sup> not full steady  
727 state, < 3 HRT at OLR 1 g VS L<sup>-1</sup> day<sup>-1</sup>; <sup>g</sup> based on combined organic load; <sup>h</sup> based on gas production  
728  
729



730 **Table 3** Physico-chemical characteristics and energy values for ms-OFMSW substrate

Parameter	Units	This study	Zhang et al. (2012) <sup>a</sup>	Cecchi et al. (2003) <sup>b</sup>
<i>Characteristics</i>				
Total solids (TS)	g TS kg <sup>-1</sup> wet weight (WW)	536	528	~540
Volatile solids (VS)	g VS kg <sup>-1</sup> WW	296	336	~270
	% TS	55.1	63.5	47
Total Kjeldahl Nitrogen (TKN)	g N kg <sup>-1</sup> WW	7.3	7.3	~6
C	%TS	28.8 <sup>c</sup>	33.0	-
H	%TS	4.7 <sup>c</sup>	3.8	-
N	%TS	1.4 <sup>c</sup>	1.3	-
S	%TS	0.2 <sup>c</sup>	0.3	-
O	%TS	16.1 <sup>c</sup>	24.2	-
Measured HHV	MJ kg <sup>-1</sup> TS	15.4 <sup>c</sup>	13.9	-
<i>Energy values</i>				
TMP <sup>d</sup>	L CH <sub>4</sub> g <sup>-1</sup> VS	0.684	0.548	-
Calculated biogas composition <sup>d</sup>	%CH <sub>4</sub>	60.0	56.6	-
HHV of CH <sub>4</sub> in TMP <sup>d</sup>	MJ kg <sup>-1</sup> TS	15.7	13.9	-
Theoretical HHV <sup>d</sup>	MJ kg <sup>-1</sup> TS	15.3	13.9	-
Measured HHV	MJ kg <sup>-1</sup> TS	15.4	13.9	-
Thermo SMP	L CH <sub>4</sub> g <sup>-1</sup> VS	0.290	-	-
	% TMP	42%	-	-
	% measured HHV	38%	-	-
	m <sup>3</sup> CH <sub>4</sub> tonne <sup>-1</sup> WW	86	-	-
Meso SMP	L CH <sub>4</sub> g <sup>-1</sup> VS	0.256	0.304	-
	% TMP	37%	55%	-
	% measured HHV	34%	55%	-
	m <sup>3</sup> CH <sub>4</sub> tonne <sup>-1</sup> WW	77	102	-

731 <sup>a</sup> Zhang et al. (2012); <sup>b</sup> Cecchi et al. (2003); <sup>c</sup> analysed by Yang et al. (2018); <sup>d</sup> calculated from elemental  
 732 composition  
 733

734 **Table 4** Modelling results

	Unit	TH2	TH1	TH1+SSD	ME1
Temperature	oC	55	55	55	37
OFMSW input	tonnes year <sup>-1</sup>	8500	4250	4250	4250
Water input	tonnes year <sup>-1</sup>	21,415	25,665	0	25,665
SSD input	tonnes year <sup>-1</sup>	0	0	25,665	0
Total digester Input	tonnes year <sup>-1</sup>	29,915	29,915	29,915	29,915
OLR	kg m <sup>-3</sup> day <sup>-1</sup>	2.0	1.0	1.7	1.0
Methane produced	m <sup>3</sup> year <sup>-1</sup>	728,003	364,002	432,065	330,292
	m <sup>3</sup> tonne <sup>-1</sup> OFMSW	86	86	102	77
VS destroyed	tonnes year <sup>-1</sup>	1,516	758	900	688
Digestate for disposal	tonnes year <sup>-1</sup>	28,399	29,157	29,015	29,227
Required CHP electrical capacity	kW	330	165	196	150
Parasitic heat demand	GJ year <sup>-1</sup>	949	949	949	1,629
Parasitic electricity demand	GJ year <sup>-1</sup>	3,450	3,001	2,999	3,002
Electricity for export	GJ year <sup>-1</sup>	6,410	1,929	2,852	1,471
Heat for export	GJ year <sup>-1</sup>	11,246	5,148	6,288	3,904
Total energy for export	GJ year <sup>-1</sup>	17,655	7,077	9,141	5,375
	GJ tonne <sup>-1</sup> OFMSW	2.1	1.7	2.2	1.3

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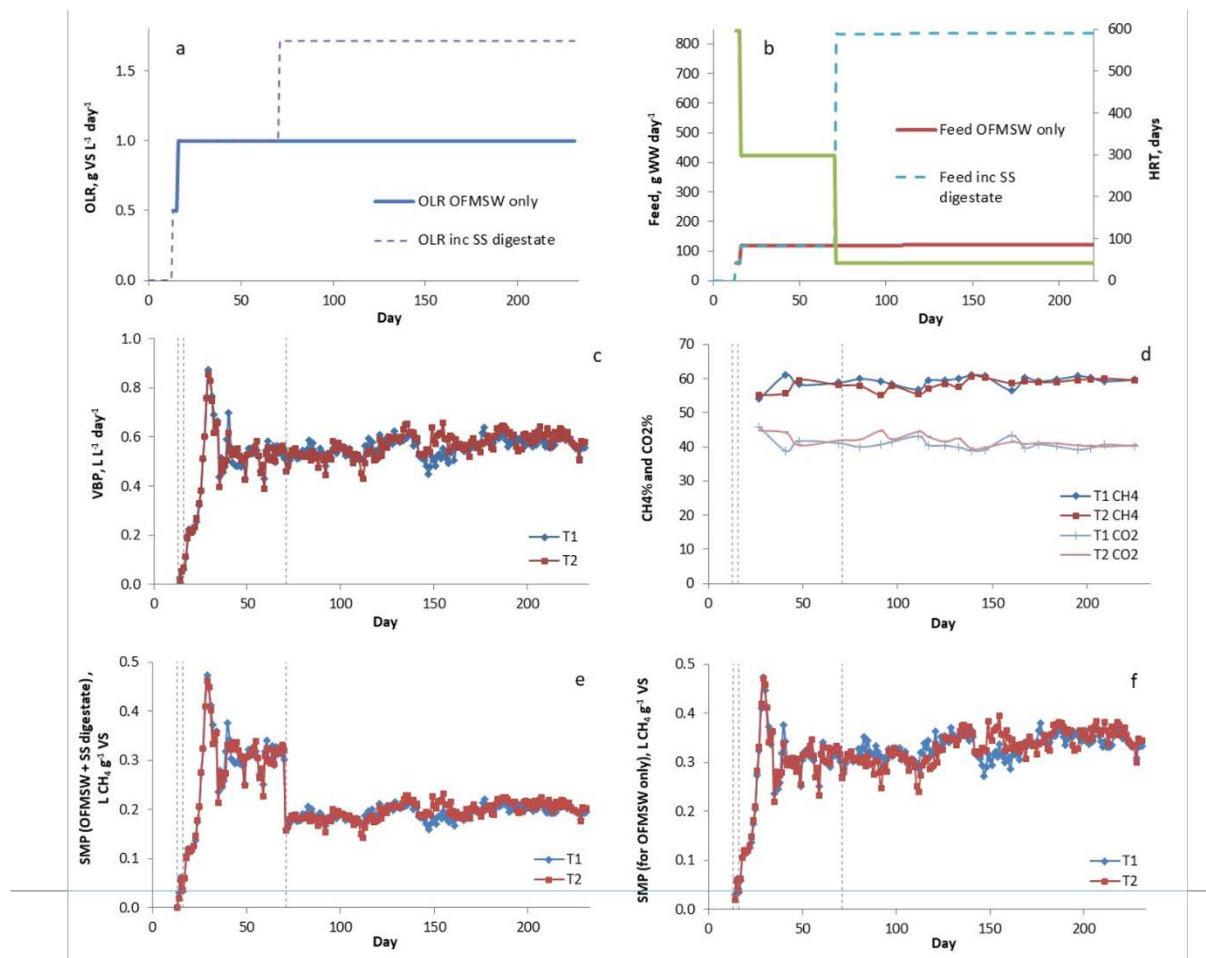
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737 List of Abbreviations

AD	Anaerobic Digestion
CHP	Combined Heat and Power
COD	Chemical Oxygen Demand
CSTR	Continuous Stirred-Tank Reactor
HHV	Higher Heating Value
HRT	Hydraulic Retention Time
IA	Intermediate Alkalinity
MSW	Municipal Solid Waste
ms-OFMSW	Mechanically-Separated Organic Fraction of Municipal Solid Waste
OFMSW	Organic Fraction of Municipal Solid Waste
OLR	Organic Loading Rate
PA	Partial Alkalinity
SMP	Specific Methane Production
SRT	Solids Retention Time
SSD	Sewage Sludge Digestate
TA	Total Alkalinity
TAN	Total Ammoniacal Nitrogen
TE	Trace Elements
TKN	Total Kjeldahl Nitrogen
TMP	Theoretical Methane Potential
TS	Total Solids
VBP	Volumetric Biogas Production
VS	Volatile Solids
WW	Wet Weight

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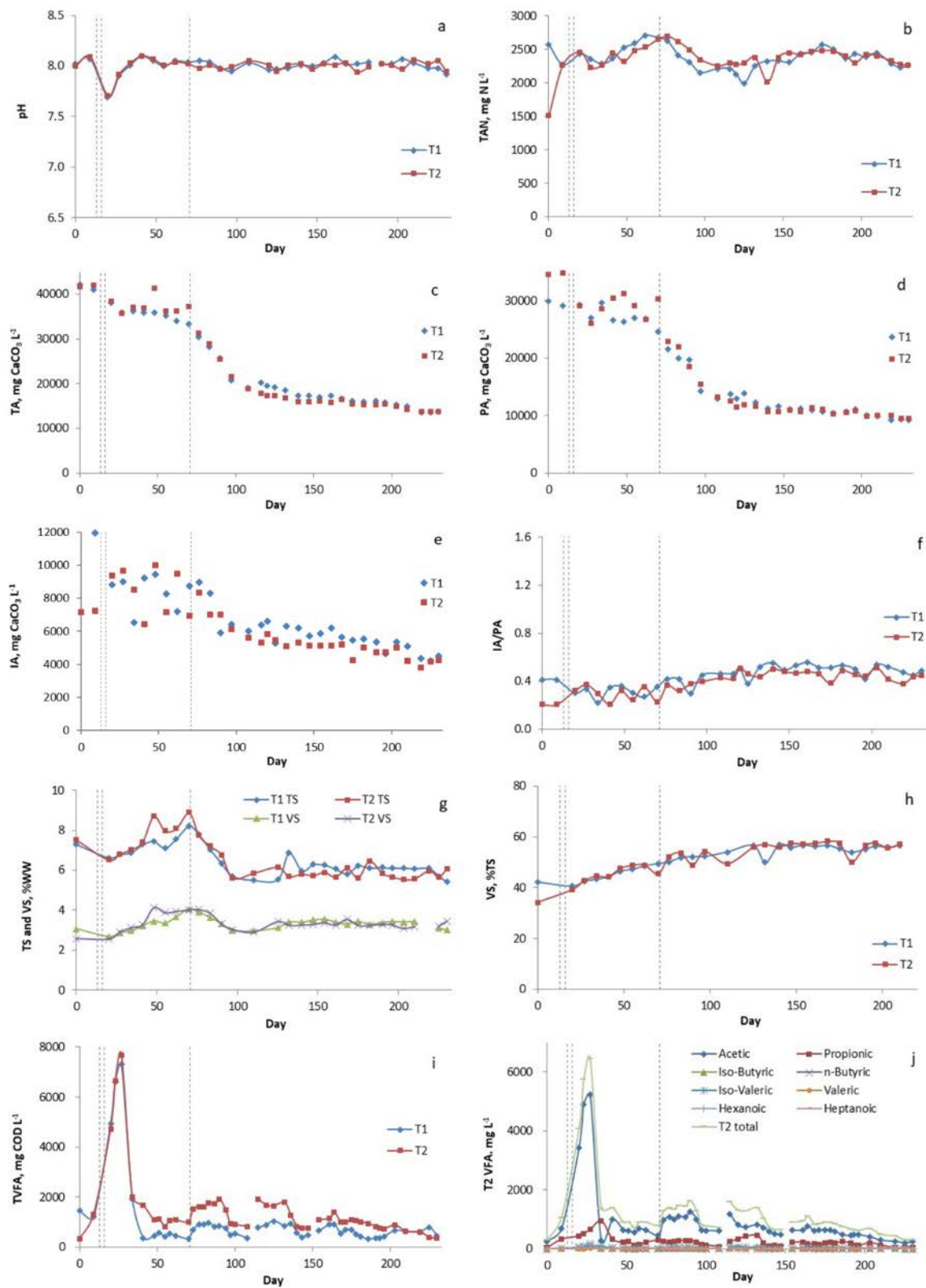
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742 **Figure 1.** Organic loading rate (a), daily feed and hydraulic retention time (b), volumetric  
 743 biogas production (c), biogas composition (d) and specific methane production based on  
 744 OFMSW and SSD (e) and OFMSW only (f) for T1 and T2 during stage 1. Vertical dotted  
 745 lines indicate change in OLR on days 13 and 16 and change in operating mode on day 69.

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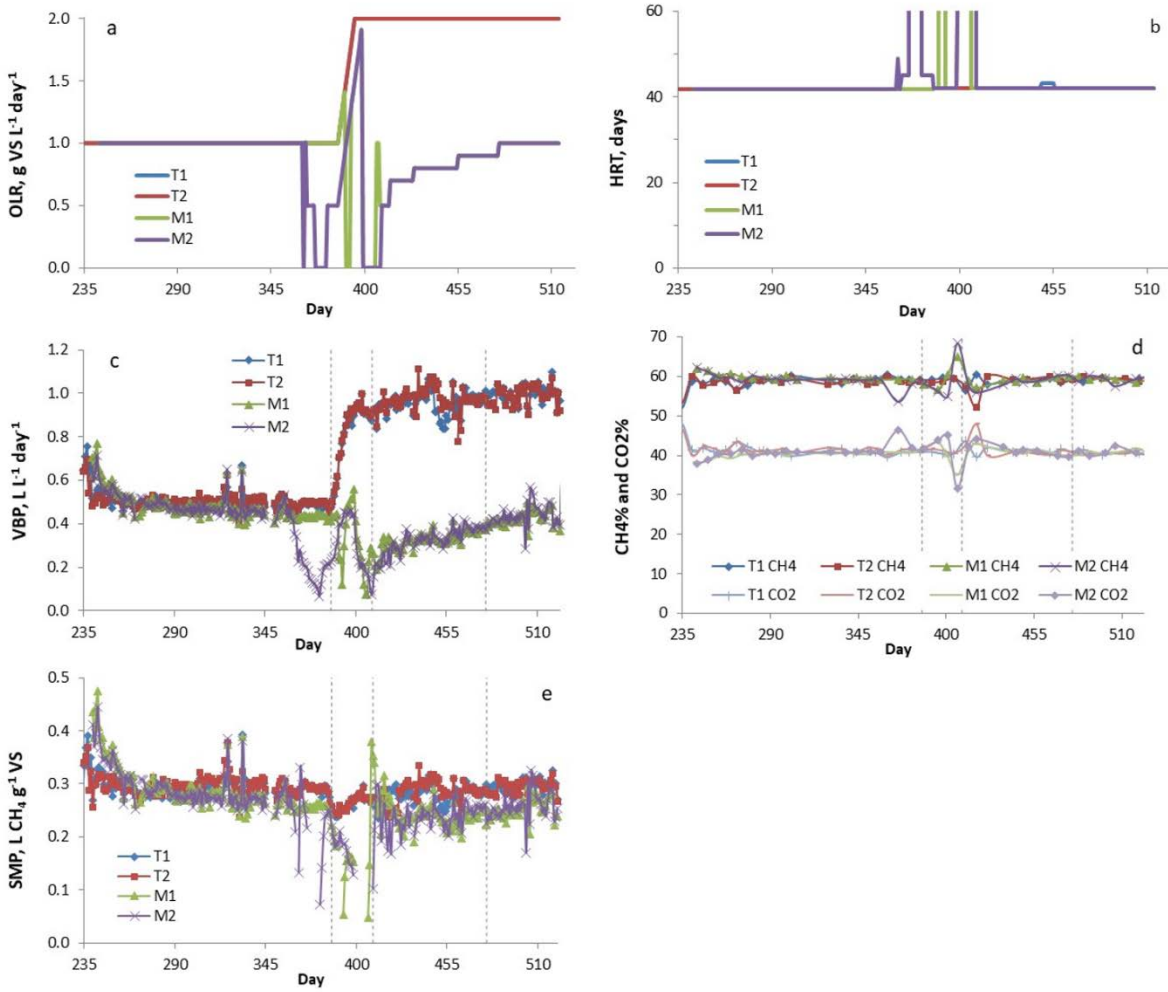


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748 **Figure 2.** pH (a), TAN (b), TA (c), PA (d), IA, (e), IA/PA (f), TS and VS (g), VS as %TS

749 (h), total VFA (i) and VFA profile (j) during stage 1. Vertical dotted lines indicate change in

750 OLR on days 13 and 16 and change in operating mode on day 69.

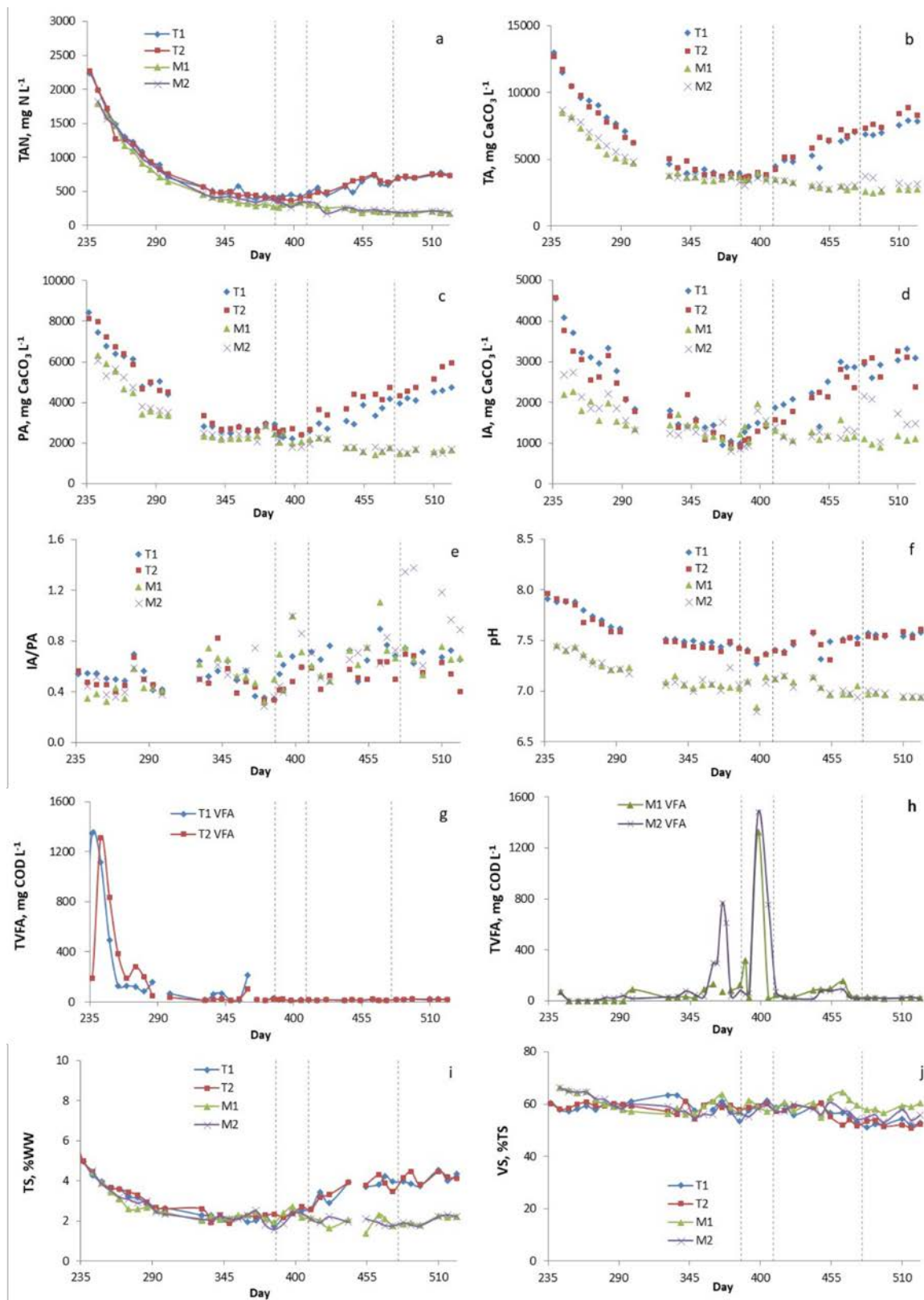


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753 **Figure 3.** OLR (a), HRT (b) VBP (c), biogas composition (d) and SMP (e) in stage 2 for all  
754 digesters. Vertical dotted lines indicate changes in OLR as shown in Table 1.

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756



757

758 **Figure 4.** TAN (a), TA (b), PA (c), IA (d), IA/PA (e), pH (f), VFA (g and h), TS (i) and VS

759 as %TS (j) for all digesters during stage 2. Vertical dotted lines indicate changes in OLR as

760 shown in Table 1.