**Continuous operation of thermophilic food waste digestion with side-stream ammonia stripping**

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**Abstract**

Digesters fed on food waste (high nitrogen content) were operated successfully over an extended period using sidestream biogas stripping to control total ammonia nitrogen (TAN) below inhibitory concentrations. This is the first time biogas stripping has been used to achieve stable thermophilic operation with undiluted substrate of this type. Stripping columns operated batch-wise treated the equivalent of 1.7-4.1% of digester contents daily at pH >10 and 70 oC, with no detrimental effect on digestion. TKN removal was 54%, with potential to recover 3.5 kg N tonne-1 substrate. When stripping

was stopped in one digester TAN increased, accompanied by rising propionic acid concentrations with progressive instability observed from 2.5 g N L-1. Eventual failure as TAN approached 5 g N L-1 was due to rapid acetic acid accumulation, resulting in a fall in pH to below 6.5. The pattern of VFA accumulationindicated failure of both acetoclastic methanogenesis and acetate oxidation.

**Keywords:** Anaerobic digestion, ammonia, food waste, biogas stripping, side-stream

**1 Introduction**

Anaerobic digestion is often considered a more sustainable route to resource recovery than other waste treatment technologies particularly when dealing with high moisture content materials (McKendry, 2002). It normally offers a net energy gain, as the parasitic energy requirements are lower than the energy value contained in the biogas produced (Burnley et al., 2011). It has therefore been used for a number of years for treatment and recovery of energy and nutrients from a wide range of organic waste resources, including food processing wastes (Digman and Kim, 2008). More recently it has become popular for treating source segregated food wastes from municipal sources (Bernstad et al., 2013; ADBA, 2016). Anaerobic digestion of this type of waste is not without difficulties, however, mainly associated with its high protein content. This is potentially problematic, as during digestion the protein is rapidly hydrolysed to release ammonia which is inhibitory or toxic to the process (Chen et al., 2008; Rajagopal et al., 2013). Food waste that has been source segregated in the home or collected from restaurants, cafeterias and supermarkets has been shown to have fairly similar properties in a number of studies (Capson-Tojo et al., 2016) and typically has a C:N ratio of ~15 and a TKN of 3-4% on a total solids basis. If used directly into a digester the total ammonia nitrogen (TAN) concentration will typically equilibrate between 4.5-5.5 g N L-1 (Banks et al., 2008), which can be tolerated in mesophilic systems but is inhibitory under thermophilic conditions.

Yet there may be some advantages in using thermophilic systems for this type of material. Thermophilic treatment offers a higher degree of pathogen destruction, potentially allowing operation without a separate pasteurisation step (Al Seadi et al., 2013). Digestate from thermophilic operation may in some cases be easier to dewater and less prone to foaming (Coelho et al., 2011; Suhartini et al, 2014). Higher specific methane yields and solids destruction have been reported (Labatut et al., 2014), while increased rates of reaction may allow the process to operate at shorter retention times and higher loadings (Cavinato et al., 2013). Inhibition is a particular problem in these conditions, however, as the equilibrium between ammonia in its ionic form and the more toxic free ammonia shifts in favour of the latter at higher temperatures. The high TAN also leads to a high pH environment which again favours a shift in the equilibrium to free ammonia nitrogen (FAN). There have therefore been no successful long-term examples of thermophilic anaerobic digestion of undiluted source segregated food waste.

It is difficult to quote absolute values for the threshold of ammonia inhibition/toxicity as this is dependent on a number of factors, including temperature and pH. It has been widely reported (Chen et al., 2008; Yenigun and Demirel, 2013) that microbial populations can acclimatise to higher ammonia concentrations, but the mechanism by which this occurs is less clear. Inhibition studies specifically related to food waste digestion (Yirong et al., 2013) suggest, however, that under thermophilic conditions (55 oC, pH ~8) stable operation at very low VFA concentrations was possible at up to ~2.5 g N L-1 while the onset of irreversible VFA accumulation is likely occur when the TAN reaches 3.5 g N L-1, corresponding to a FAN concentration of around 0.8 g N L-1.

Where thermophilic anaerobic digestion of food waste has been practiced at a large scale, one approach taken to avoid toxicity has been to reduce the TAN concentration in the digester by dilution (Neiva Correia et al., 2008). This has both resource and energy implications, but if the advantages of thermophilic operation can be shown to outweigh these it may offer an effective strategy. Co-digestion to increase the C/N ratio is also possible, and its benefits have been widely demonstrated (Mata-Alvarez et al., 2011, 2014); but the practicality of this approach depends on the availability of a suitable low nitrogen co-substrate. Reducing the ammonia in the digester or its feed are also potential solutions. Approaches considered include the development of ion exchange or zeolite bed reactors (Milán et al., 1997; Zheng et al., 2015), and the use of membrane contactors for *in situ* removal or as a post-treatment step with stripped liquor recycle (Lauterbock et al., 2014; Du Preez et al., 2005). Gas stripping has been most widely used, both as a pre-treatment to reduce the substrate nitrogen content (Zhang et al., 2012; Markou, 2015; Yabu et al., 2011); and to remove ammonia from the digester mixed liquor using air (Pedezzi et al., 2017), nitrogen (Nielsen et al., 2013), steam (Resch et al., 2011), biogas (De la Rubia et al., 2010; Abouelenien et al., 2010), and flue or exhaust gases (Bousek at al., 2016) as the stripping agent. Many of these studies were based on short-term laboratory-scale batch testing, but Pintucci et al. (2017) operated a 3 m3 pilot-scale system with side-stream air stripping for co-digestion of swine manure and vegetable wastes, while Pedezzi et al. (2017) ran digesters coupled to a side-stream air stripping column on the same feedstock for over 700 days. Sun et al. (2014) operated digesters for a 90-day period, on stillage derived from ethanol fermentation of food wastes, both with and without biogas stripping of ammonia.

Walker et al. (2011) discussed the advantages and disadvantage of biogas stripping in various operating modes and from different locations in the plant flowsheet or with additional process stages. Of these options, side-stream stripping appears to offer the greatest promise (Serna-Maza et al., 2015, 2017) and was shown to be able to reduce TAN concentrations in a mesophilic digester without apparent detriment to the process (Serna-Maza et al., 2014). The system described involved a simple 'bolt-on' stripping column in which a proportion of the digestate is stripped of its ammonia under alkaline conditions, with recovery as a concentrated ammonium sulphate solution by dissolution into sulphuric acid. The work concluded that to reduce TAN below threshold toxicity concentrations in a thermophilic food waste digestion process a minimum stripping temperature of 70 oC would be required (Serna-Maza et al., 2014)

The aim of the current study was to demonstrate that a side-stream ammonia stripping process used in conjunction with thermophilic food waste digestion could allow successful long-term operation. The work was therefore carried out over an extended period in order to provide steady state data on digester performance and stability with side-stream stripping, and to compare this with an unstripped control.

**2 Materials and methods**

**2.1 Digester and stripping column design**

Four continuously-stirred tank reactor (CSTR) digesters (T1-4) were used in the study, each with a total capacity of 40 L and a working volume of 35 L. These were constructed from 36 cm ID PVC pipe sealed at the top and bottom by plates incorporating feed and drainage ports. The digesters are sealed from the outside atmosphere by a draught tube through which an offset bar stirrer is inserted to allow low-speed mixing at 26 rpm by geared motors (Parvalux, UK). Digester temperature was controlled at 55°C by recirculating water from a thermostatic bath through an internal heating coil. Biogas production was measured using continuous gas flow meters, and is reported at standard temperature and pressure (STP) of 0 oC and 101.325 kPa (Walker et al., 2009). Biogas was also periodically collected in gas-impermeable bags to check gas counter calibrations and determine the biogas composition.

The digesters were coupled to ammonia stripping columns C1-C4 made from stainless steel tube with a height of 56 cm and 10 cm internal diameter. Temperature in the columns was maintained at 70 oC using externally mounted thermostatically-controlled electrical heating mats (Non Adhesive Wire Wound Heater 104 Dia x 200 P 230V, Holroyd, UK). Ammonia stripping was carried out as a semi-batch process with biogas recirculated around the system by a peristaltic pump (505s, Watson Marlow, UK). The biogas enters the stripping column at a controlled flow-rate through a sintered-glass diffuser. After leaving the column it is then passed through traps to remove ammonia: this was achieved by provision of a condensate trap followed by bubbling through deionised water and then through 0.25 N H2SO4 before recirculation to the stripping columns. The system configuration is shown in Figure 1.

**2.2 Inoculum, feedstock and digester operation**

The inoculum used was digestate from a mesophilic digester operated by Southern Water Plc for treatment of municipal wastewater biosolids (Millbrook, Southampton, UK). This was placed in the digesters and the temperature was raised to 55 oC in a single step, after which the digesters were left without feeding for 27 days to allow acclimatisation to the new operating temperature.

From day 0-20 the digesters were fed on a low-nitrogen food waste that has previously been used in thermophilic digestion experiments (Yirong et al., 2013): this was to avoid any rapid build-up of ammonia during the acclimatisation period. Feeding started on day 0 at an organic loading rate (OLR) of 0.5 kg volatile solids (VS) m-3 day-1 and was increased in equal increments up to 1.5 kg VS m-3 day-1 on day 20.

On day 21 the feed was switched to source separated domestic food waste and the OLR was raised to the final target value of 2.0 kg VS m-3 day-1. The food waste used was collected commercially by Veolia Environmental Services (UK) Plc from households in Eastleigh, UK and was typical of this type of material (Yirong et al., 2015). Samples of 200-300 kg were taken and any obvious contaminants, large bones and stones were removed. The material was homogenised to a pulp by passing it through a macerating grinder (S52/010, IMC Limited, UK), then mixed, subdivided into portions and frozen at -20 oC. Portions were defrosted as needed prior to use. During the course of the experiment three batches of waste were prepared in this way.

Digestate was removed twice per week and sieved to remove any large solids, which were returned to the digester. A proportion of the sieved digestate was wasted, and the remainder was added to the stripping column after pH adjustment by addition of lime (CaO), and removal of the stripped digestate. The stripped digestate was returned to the main digester to maintain its working volume. The digesters were monitored on a regular basis for pH, TAN, alkalinity, total and volatile solids and volatile fatty acid (VFA) concentrations.

A trace element solution was added as noted below to give an additional working concentration in the digestate of the following metals: (mg L-1) Cobalt 1.0, Nickel 1.0, Molybdenum 0.2, Selenium 0.2, Tungsten 0.2.

The experiment was conducted in three stages. In Stage 1 all four digesters were run under the same conditions, to demonstrate effective acclimatisation to thermophilic conditions and confirm the repeatability of the response to stripping. In Stage 2 the quantity of digestate stripped was increased for two digesters, while two continued under the same conditions as before; in Stage 3 the quantity stripped was doubled for one digester and in another stripping was stopped. The purpose of Stages 2 and 3 was to demonstrate stable operation could be achieved by stripping with control over the digestate TAN concentration. Table 1 summaries the key operational and other changes.

**2.3 Analytical methods**

Total solids (TS) and volatile solids (VS) were determined according to Standard Method 2540 G (APHA, 2005) using a Heraeus Function Line Series oven and a 201/301 Carbolite muffle furnace. pH was measured using a Jenway 3010 meter (Bibby Scientific Ltd, UK) with a combination glass electrode calibrated in buffers at pH 4, 7 and 9.2 (Fisher Scientific, UK). Alkalinity was measured by titration with 0.25N H2SO4 to endpoints of pH 5.75 and 4.3 using an automatic digital titration burette system (SCHOTT titroline easy) to allow calculation of total (TA), partial (PA) and intermediate alkalinity (IA) (Ripley et al., 1986). Total Kjeldahl Nitrogen (TKN) was determined after acid digestion by steam distillation and titration. This used a BÜCHI K-435 Digestion Unit with H2SO4 and K2SO4 as the reactants and CuSO4 as the catalyst to convert the amino-nitrogen and free ammonia (NH3) to ammonium (NH4+). TAN was measured using a BÜCHI Distillation Unit K-350 with NaOH addition followed by collection of the distillate in boric acid indicator and titration with 0.25 N H2SO4. FAN concentrations were calculated from TAN, pH and temperature as in Hansen et al. (1998). Volatile fatty acid (VFA) concentrations were determined by gas chromatography (Shimadzu GC-2010), with a flame ionization detector and a capillary column (SGE BP-21) and helium as carrier gas. Samples were acidified to 10% with formic acid and measured against mixed standards of 50, 250 and 500 mg L-1 of acetic, propionic, iso-butyric, n-butyric, iso-valeric, valeric, hexanoic and heptanoic acids (APHA, 2005). Biogas composition (CH4 and CO2) was determined using a Varian star 3400 CX Gas Chromatograph fitted with a packed stainless steel SUPELCO 80/100 mesh porapack-Q column and a TCD detector. The GC was calibrated with a standard gas of 65% CH4 and 35% CO2 (v/v) (BOC Ltd, UK).

**3 Results and discussion**

**3.1 Feedstock characteristics**

Results of a detailed characterisation of the first batch of feedstock are given in Table 2, and confirm that its properties are closely similar to those reported elsewhere for source separated domestic food wastes (e.g. Yirong et al., 2015). Between days 120-244 and 245-345 new batches of source segregated domestic food waste were used, with characteristics similar to those of the original batch but slightly different moisture contents. This led to small changes in the hydraulic retention time (HRT), as the OLR was maintained at 2 kg VS m-3 day-1.

**3.2 Acclimatisation and start of stripping (Stage 1, day 0-107)**

It was necessary initially to acclimate the inoculum to thermophilic conditions, then to avoid toxicity it was necessary to strip ammonia from the digesters almost from the outset to remain below inhibitory concentrations. During stage 1 all digesters were operated under the same conditions. Feeding began on day 0 with low-nitrogen food waste at an OLR of 0.5 kg VS m-3 day-1 and was incrementally raised to 1.5 kg VS m-3 day-1 by day 20. On day 21 the feed was switched to source segregated domestic food waste at 2.0 kg VS m-3 day-1 and ammonia stripping commenced. For this purpose 2.1 kg of digestate was taken from each digester twice per week, and stripped at a gas flow rate of 0.15 m3 m-3digestate min-1 or 0.04 m3 m-2 min-1, corresponding to violent mixing (Perry and Green, 1999). These conditions continued until the end of this stage on day 107.

Figure 2 shows the process monitoring and stability parameters during stage 1. The pH remained fairly steady, averaging around 7.8 in all digesters from day 21 onwards (Figure 2a). TAN concentrations rose after the switch from low-nitrogen feedstock to normal source segregated domestic food waste on day 21, but remained below the lower stability threshold of 2.5 g N L-1 (Yirong et al., 2013) in all digesters apart from T4, where TAN was slightly higher from day 94 onwards (Figure 2b). The reason for this small difference is not known but may be due to minor differences in stripping efficiency, which then affects other parameters. Total, partial and intermediate alkalinity (Figure 2c, d and e) rose throughout the period, reflecting the addition of lime for pH control in the stripping columns as well as the initial rise in ammonia concentration. The ratio of intermediate to partial alkalinity (IA/PA) fluctuated but showed a downward trend (Figure 2f): this parameter is considered a good indicator of digestion stability (Ripley et al., 1986). Digestate volatile solids content had stabilised by day ~100 (Figure 2g), but total solids (Figure 2h) continued to rise while the VS/TS ratio fell (Figure 2i) as a result of the lime addition. Total VFA concentrations (Figure 2j) remained low until day 50 but then increased slightly as TAN concentrations rose towards 2.5 g N L-1: the higher VFA concentration in T4 mirrored the higher TAN in this digester. Figure 3 shows individual VFA species for each digester with total VFA and TAN. The appearance of occasional peaks of propionic acid of around ~1 g L-1 is similar to that previously observed in reactors running on synthetic low-nitrogen food waste spiked with urea to achieve this TAN concentration (Yirong et al., 2013). The slightly higher propionic acid concentration in T4 reflects the higher TAN concentration, as well as the range of natural variability seen in biological systems of this type.

Digester performance was assessed based on the volumetric biogas production (VBP) and the specific methane production (SMP) for each digester. Differences between the digesters were negligible (Figure 3e and f). The average SMP for the first batch of food waste from day 50-107 was 0.480 m3 CH4 kg-1 VS added. This is at the higher end of the expected range for typical source segregated domestic foodwaste, but may reflect both the lipid content of this batch, which was slightly above average, and possibly an enhanced degree of degradation due to the thermophilic conditions and/or high-temperature alkaline hydrolysis occurring in the stripping column.

**3.3 Increasing ammonia removal by increased stripping (Stage 2, day 108-209)**

In stage 2 the quantity of digestate stripped was increased in digesters T1 and T2 to 2.5 kg twice per week. The gas flow rate remained the same, giving gas mixing rates of 0.126 m3 m-3digestate min-1 or 0.04 m3 m-2 min-1 (violent mixing). Feeding with the second batch of food waste began from day 120, and a one-off dose of trace element solution was added to T4 on day 178. From day 108 TAN concentrations in T1 and T2 began to fall more rapidly than in T3 and T4 as a result of the increased stripping (Figure 4a). This was accompanied by an increase in TA and PA (Figure 4b and c) as proportionately more lime was added. The extra lime addition also increased the TS content in T1 and T2 above that in T3 and T4 (Figure 4d), but VS content during this period was stable at around 3.4 % of wet weight (WW) (Figure 4e) leading to a fall in the VS/TS ratios (Figure 4f). In T1 and T2 the TAN was gradually reduced to around 2.0 g N L-1 and the previous signs of mild instability, as indicated by slightly elevated VFA concentrations at the end of stage 1, gradually decreased. VFA concentrations in T3 and T4, however, continued to show fluctuations up to ~1 g L-1 as the TAN concentration in these digesters remained slightly higher. In all 4 digesters there was no apparent loss in performance as both VBP and SMP remained more or less constant during this period (Figure 4g and h), with average values of 1.72 L L-1 day-1 and 0.479 L CH4 g-1 VS, respectively.

**3.4 Comparison of stripped and unstripped digesters (Stage 3, day 210 on).**

This stage was designed to assess the effectiveness of the ammonia stripping system as a means of producing different conditions in the digesters. In digester T4 stripping was stopped and the ammonia concentration allowed to increase above the critical threshold at which instability and VFA accumulation were expected to occur. In digester T2 the ammonia concentration was further reduced by doubling the volume stripped to 5.0 kg twice per week (using stripping columns C2 and C4 in parallel). The other two digesters continued to operate under the same stripping conditions as in stage 2 (2.5 kg in T1 and 2.1 kg in T3 stripped twice per week). Feeding with the third batch of food waste started from day 245.

At the beginning of stage 3 TAN in digesters T1-3 was close to 2.0 g N L-1 while that in T4 was slightly higher at 2.5 g N L-1. For the rest of the experimental period the TAN concentration in T4 increased steadily (Figure 4a), reaching 5.2 g N L-1 by day 345. In T2, where the amount of digestate stripped was doubled, TAN reduced over the next 70 days and stabilised at an average value of 1.5 g N L-1, well below any inhibitory threshold (Yirong et al., 2013). In T1 and T3, where the stripping regimes remained the same as in the previous stage, TAN concentrations rose slightly reflecting a slight increase in the TKN of the new batch of feedstock, and were close to the threshold value of 2.5 g N L-1 where early signs of instability may be observed. Process stability parameters during this period can be seen in Figure 4. The pH in all of the digesters except T4 remained stable during the whole stage (Figure 4i), averaging around 7.8. In T4 the pH remained close to that in the other digesters until day 322 after which it fell sharply to below 6.5 on day 345, at which time feeding was stopped. This sharp fall in pH can be attributed to the buffering capacity in the digester being overcome by the increasing VFA concentration (Figure 4j). Changes in total alkalinity (Figure 4b) reflected the degree of alkali addition for stripping purposes. In T4 the TA dropped after stripping ceased; whereas in T1 and T3 it increased slightly, and in T2 there was a sharper rise due to the double stripping. There were some problems in obtaining consistent alkalinity measurements for T2, especially after double stripping was introduced, probably due to insufficient mixing of samples; but these were resolved by the end of the run with stable values achieved. PA (Figure 4c) followed a similar trend to TA until around day 325 when the PA value in T4 began to fall dramatically, from ~18 to ~8 g CaCO3 L-1 by day 345. At the same time the IA in T4 rose from day 300 onwards, leading to a peak in IA/PA ratio of 2.3 by day 345. TS concentrations in all four digesters (Figure 4d) reflected the degree of alkali addition for stripping purposes. VS concentrations both as a proportion of wet weight and of TS content (Figure 4e and f) clearly indicated, however, that VS conversion in T4 decreased after stripping ceased; while conversion in T2 (double stripping) appeared to be slightly higher than in T1 and T3 (single stripping).

The changes in pH and alkalinity were linked to changes in VFA concentrations in the digesters (Figure 4j). In T2 where the TAN concentration was reduced to 1.5 g N L-1, the total VFA concentration between days 220-345 was very low, averaging 0.15 g L-1. In T1 (2.5 kg stripped) total VFA showed a slight increase when TAN concentrations reached ~2.2 g N L-1, averaging around 0.7 g L-1. In T3 (2.1 kg stripped) total VFA concentrations were similar to those in T2, apart from during a brief peak around day 330 which may have been due to a short-term temperature rise in T3 to 57 oC.

The VFA species are shown in Figure 5 together with calculated FAN concentrations. In T1 and T3 the main acids present were acetic and propionic, with elevated levels appearing when FAN concentrations approached 0.5 g N L-1. In T4 immediately after stripping ceased there was a gradual but steady increase in propionic acid concentration which reached around 11 g L-1 by day 345. Acetic acid also rose to around 2.2 g L-1 by day 241, leading to a short-term reduction in FAN, but then fell over the next 10 days. From day 300 onwards other VFA species began to appear; and from day 329 there was a rapid linear increase in acetic acid of around 0.95 g L-1 day-1 until feeding ceased on day 345, by which time the concentration had reached 17 g L-1.

SMP of the stripped digesters was very close to that in the previous period (Figure 4h), with average values of 0.472, 0.474 and 0.472 L CH4 g-1 VS for T1, T2 and T3, respectively. VBP fell by around 7-10% in comparison with stage 1 (Figure 4g), possibly due to the effect of lime addition on the carbonate equilibrium leading to an increase in CO2 dissolution: this was reflected in a small increase in biogas methane content from around 55% to 59%, resulting in the SMP remaining constant. In T4 gas production declined slowly from day ~250 onwards, and sharply from day ~300, reaching 0.16 L CH4 g-1 VS by day 345. Biogas methane content in T4 had fallen to 53.8% by the end of the run, well below the average of 59.2% for the other digesters.

After feeding and stripping stopped on day 345, monitoring of selected digestion parameters continued at a reduced frequency until day 382. TAN concentrations rose slightly in all reactors, probably indicating a reduction in and hydrolysis of the active microbial biomass. In T4 the pH rose, IA/PA ratio fell and VFA concentrations reduced slightly, reflecting a fall in acetic acid concentration to around 15 g L-1. Concentrations of other VFA species were unaffected, however, while the FAN concentration rose in response to these changes.

The VFA profiles, VBP and SMP results provided a clear indication of the stability and performance of the digesters at each of the targeted TAN concentrations. As the TAN was allowed to increase in T4 the digester became unstable, resulting in loss of methane production, a sharp drop in pH and a rapid rise in IA/PA ratio as the TAN approached 5 g N L-1 (FAN 0.8 g N L-1); but increasing propionic acid concentrations indicated the onset of progressive instability as the TAN exceeded 2.5 g N L-1. Digesters T1 and T3 showed signs of the onset of mild instability in the form of fluctuating VFA concentrations at a threshold TAN concentration of 2.2 g N L-1, or FAN of around 0.5 g N L-1. It is therefore difficult to specify a precise TAN (or FAN) concentration where ammonia toxicity occurs, as this manifests itself in different forms. Fluctuating VFA concentrations with the appearance of VFA of predominantly C3 and longer chain length indicate some mild interference with the flow of carbon through to methane once the TAN exceeds a threshold of 2.5 g N L-1. This could directly affect the activities of the propionate-degrading acetogens, which may be more sensitive to ammonia than methanogenic *Archaea* (Calli et al., 2005). It is also possible that ammonia inhibition of acetoclastic methanogenesis results in the channelling of carbon to CH4 via acetate oxidation and hydrogenotrophic methanogenesis. This, however, may lead to an increase in the partial pressure of H2 and a consequent reduction in propionate degradation as changes in the thermodynamic equilibrium make it a less desirable substrate (Muller et al., 2010). This could explain the consistently low concentrations of acetic acid observed until the TAN concentration approached 5.0 g N L-1, at which point a rapid increase in acetic acid was observed, indicating that both the acetoclastic and acetate oxidation routes to methane formation had been compromised. The fall in pH resulting from the increased acid load takes this to below an optimum value for methanogenesis and the digester becomes ‘stuck’ (McCarty and McKinney, 1961), a condition from which recovery is difficult.

**3.5 Stripping parameters and performance**

The purpose of sidestream stripping is to allow an increase in the temperature and pH of a proportion of the digestate to facilitate removal of ammonia in the gaseous phase: in this case the aim was to increase the temperature to 70 oC and the pH to above 10 by lime (CaO) addition, whilst recirculating biogas at a fixed flow rate. The lime dose required was around 14 g CaO kg-1 WW of digestate, which was lower than the 18.6-21.4 g CaO kg-1 WW needed by Serna et al. (2014) in a previous study with food waste of the same type digested under mesophilic conditions. The lime dose used was sufficient to raise the initial pH above 10, and on some occasions even above 11. The pH of the stripped sample declined during the stripping process to final values which were generally between 8.5-9.5.

Figure 6a shows the TAN concentration in the stripped digestate at the end of each 3.5-day stripping period, while Figure 6b shows the removal efficiency. During the first ~50 days of operation, when TAN concentrations were increasing as a result of the introduction of a normal food waste feedstock, removal efficiencies decreased (Figure 6b). It can be seen that during stage 2 the final TAN concentration was slightly lower in C1 and C2 (2.5 kg stripped) than in C3 and C4 (2.1 kg stripped); while the removal efficiency in C1 and C2 was slightly higher, at 79% compared to 73% in C3 and C4, perhaps due to the greater depth of digestate in stripping columns C1 and C2. In stage 3 the final TAN concentrations for the two columns (C2 and C4, 2.5 kg stripped) used for digestate from digester T2 were closely similar, indicating that both were performing equally well. A fall in stripping efficiency in C1 during stage 3 could have been due to partial blockage of the gas sparger, after a temporary interruption to gas recirculation on day 213.

Figure 6c shows TAN removal kinetics over the 3.5-day stripping period on day 315. These results allow comparison of stripping performance for initial TAN concentrations of 2.5 and 1.5 g N L-1, and for duplicate columns C2 and C4 used to strip digestate from T4 under the same conditions. As expected, TAN removal was non-linear with most removal occurring on the first day. The results also showed, however, that there were slight differences in performance between different columns even when operated under conditions as similar as experimentally possible. This can be seen in C1 and C4 which both achieved a final removal rate of over 75% but showed a difference in kinetics, with C4 reaching a plateau sooner. Small differences were also visible between C1 and C3, with the poorer performance of C1 possibly associated with fouling of the diffuser as noted above. The final concentration at the end of the stripping period appeared to depend on the initial concentration. In all cases the final pH was 9 or below, and it would be interesting to see whether sequential lime addition could enable further removal in C1 and C3 which had the higher initial and final TAN concentrations. The small number of data points made reliable estimation of time constants for ammonia removal difficult, but values appeared to be at the lower end of those reported by Serna-Maza et al. (2015) for fresh FW digestates, indicating stripping was relatively easy. C2 and C4 (2.5 kg stripped) had stripped ~70% of TAN by the second day, while C3 (2.1 kg) had removed 61%.

Figure 4d shows TKN and organic N (i.e. TKN minus TAN) in the digesters. There is a clear reduction in TKN while the organic N is relatively unaffected, indicating that removal was almost entirely in the form of TAN. No evidence of additional breakdown of organic material in the stripper was seen in the current experiment, as organic N in the digester remained almost constant throughout the experiment. This differs from the results of Serna-Maza et al. (2015) who saw conversion of organic nitrogen to TAN in the stripping process for ammonia removal from mesophilic food waste digestates, and speculated that thermophilic digestion could give a slight improvement in overall biogas yield through improved degradation. The current result may indicate that the thermophilic conditions already have the capacity for additional hydrolysis of organic N, leaving little scope for further physico-chemical degradation in the stripping column. As there was no mesophilic control digester it was not possible to identify or quantify any differences in specific gas production at different operating temperatures.

Overall TKN removal rates for T2 were calculated from the feedstock TKN value (6.67 g N kg-1 WW) and the digestate TKN, taking into account the reduction in digestate volume associated with VS destruction. The digestate TKN concentration without stripping was estimated as 8.3 g N kg-1 WW on a mass balance basis based on VS measurements, or 8.5 g N kg-1 WW based on weight of dry biogas produced. The measured TKN in T2 at the end of stage 3 was around 3.9 g N kg-1 WW, corresponding to a removal rate of 53% or 54% respectively for these two methods of estimating VS destruction. Serna-Maza et al. (2014) noted that for mesophilic digestion with stripping at high temperature (≥70⁰C) and pH ~10, 48% of the TAN was removed over a 138-day period without any detrimental effects on digester performance. The removal in the current study equates to a potential recovery of around 3.6 kg of N per tonne of feedstock treated.

An earlier study (Serna-Maza et al., 2014) showed how ammonia could be removed from a mesophilic digester using a side-stream stripping process: this indicated the critical parameters for time/temperature/pH that were used in the design of these experiments. The major differences here were that the digestion was under thermophilic conditions, and TAN was controlled at the desired values by preventing its accumulation rather than by stripping from a high initial concentration. The current work confirmed the process conditions for ammonia stripping, and adjusted the stripping rate to achieve target TAN concentrations in the digesters. The maximum quantity of digestate stripped from one digester was 10 kg per week, to which 140 g of lime had been added. The average daily proportion of the digester stripped was therefore 4.1%, corresponding to an equivalent retention time of 24.5 days for stripped material. This is a considerably lower fraction that the 21% of digester contents treated 3 or 5 times per week in the air stripping system used by Pedezzi et al. (2017), although the latter required no chemical addition. In the current work the material was held in the stripping column at an initial pH ≥10 for a period of around 84 hours before being returned to the digester, without any apparent detrimental effect. Such treatment, however, is likely to disrupt microbial activity in the treated portion and may cause thermal hydrolysis. The process will therefore increase the overall growth rate of the system, as the dead biomass can be regarded as being ‘washed out’ in terms of its activity, even though the residues were returned to the digester. Evidence for any effect from the alkaline thermal hydrolysis is largely speculative, as it is not possible to run a parallel thermophilic system without ammonia stripping due to the issue of toxicity; but the SMP of ~0.47-48 L CH4 g-1VS of the food waste was slightly higher than when the same batches of FW were digested mesophilically with typical SMP values between 0.45-0.46 L CH4 g-1 VS (Yirong et al., 2013). Further evidence can be seen in the solids data in stage 3 where the rates of stripping varied between the digesters. As expected the total solids in T2 increased with the higher lime dose. The VS content in this digester was lower than in the other digesters, however, suggesting a greater breakdown of solids than in T1 and T3 although these digesters appeared to perform equally well in terms of methane production. The experimental results presented do not allow a firm conclusion to be reached, and a comparative study of specific methane yields between stable mesophilic digestion and a lime stripped thermophilic system would be required to quantify any differences. The organic loading used in the study was also comparatively low in relation to those now used in commercial digestion systems, which are usually in the region of 4 kg VS m3 day-1 in mesophilic conditions: a more marked effect may have been seen if a higher loading had been used. This would also have affected the stripping rate required to maintain the target digestate TAN concentration. There is ample capacity in the system to achieve this, however, as demonstrated by the low TAN concentrations that were achieved in T4. While the current trial successfully demonstrated stable digestion for periods of >3 HRT for the main digester, and >5.5 cycles in terms of the stripping column, there is considerable scope for optimisation of the stripping conditions.

**4 Conclusions**

Thermophilic food waste digestion was possible using a sidestream stripping process at 70 oC with initial pH >10. Ammonia inhibition thresholds were established for this substrate, and the process could control TAN below these without detrimental effects. Stripping achieved 54% TKN removal, potentially allowing recovery of 3.5 g N kg-1 substrate. Stripping kinetics indicated removal could be achieved within 1 day, but there is still scope for process optimisation. The pattern of VFA accumulation without stripping suggested that the acetate oxidation pathway failed as TAN approached 5 g N L-1, although progressive instability was noted at >2.5 g N L-1.

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**References**

1. Abouelenien, F., Fujiwara, W., Namba, Y., Kosseva, M., Nishio, N., Nakashimada, Y. 2010. Improved methane fermentation of chicken manure via ammonia removal by biogas recycle. *Bioresource technology*, **101**(16), 6368-6373.
2. ADBA, 2016. Anaerobic Digestion Market Report. Anaerobic Digestion and Bioresources Agency Quarterly report July 2016. Available <http://adbioresources.org/adba-market-policy-reports/adba-market-report>, last accessed May 2017.
3. Al Seadi, T., Fuchs, W., Drosg, B., Janssen, R., Rutz, D. 2013. Biogas digestate quality and utilization. *The biogas handbook: Science, production and applications*, 267.
4. APHA, 2005. Standard Methods for the Examination of Water and Wastewater, 21st ed. American Public Health Association, American Water Works Association, Water Environment Federation, Washington, USA.
5. Banks, C.J., Chesshire, M., Stringfellow, A. 2008. A pilot-scale comparison of mesophilic and thermophilic digestion of source segregated domestic food waste. *Water science and technology*, **58**(7), 1475-1481.
6. Bernstad, A., Malmquist, L., Truedsson, C., la Cour Jansen, J. 2013. Need for improvements in physical pretreatment of source-separated household food waste. *Waste management*, **33**(3), 746-754.
7. Bousek, J., Scroccaro, D., Sima, J., Weissenbacher, N., Fuchs, W. 2016. Influence of the gas composition on the efficiency of ammonia stripping of biogas digestate. *Bioresource technology*, **203**, 259-266.
8. Burnley, S., Phillips, R., Coleman, T., Rampling, T. 2011. Energy implications of the thermal recovery of biodegradable municipal waste materials in the United Kingdom. *Waste Management*, **31**(9), 1949-1959.
9. Calli, B., Mertoglu, B., Inanc, B., Yenigun, O. 2005. Effects of high free ammonia concentrations on the performances of anaerobic bioreactors. *Process Biochemistry*, **40**(3), 1285-1292.
10. Capson-Tojo, G., Rouez, M., Crest, M., Steyer, J.-P., Delgenès, J.-P., Escudié, R. 2016. Food waste valorization via anaerobic processes: a review. *Reviews in Environmental Science and Bio/Technology*, **15**(3), 499-547.
11. Cavinato, C., Bolzonella, D., Pavan, P., Fatone, F., Cecchi, F. 2013. Mesophilic and thermophilic anaerobic co-digestion of waste activated sludge and source sorted biowaste in pilot-and full-scale reactors. *Renewable Energy*, **55**, 260-265.
12. Chen, Y., Cheng, J.J., Creamer, K.S. 2008. Inhibition of anaerobic digestion process: a review. *Bioresource technology*, **99**(10), 4044-4064.
13. Coelho, N.M.G., Droste, R.L., Kennedy, K.J. 2011. Evaluation of continuous mesophilic, thermophilic and temperature phased anaerobic digestion of microwaved activated sludge. *Water research*, **45**(9), 2822-2834.
14. De la Rubia, M.Á., Walker, M., Heaven, S., Banks, C.J., Borja, R. 2010. Preliminary trials of in situ ammonia stripping from source segregated domestic food waste digestate using biogas: Effect of temperature and flow rate. *Bioresource technology*, **101**(24), 9486-9492.
15. Digman, B., Kim, D.S. 2008. Review: alternative energy from food processing wastes. *Environmental Progress*, **27**(4), 524-537.
16. Du Preez, J., Norddahl, B., Christensen, K. 2005. The BIOREK® concept: a hybrid membrane bioreactor concept for very strong wastewater. *Desalination*, **183**(1-3), 407-415.
17. Hansen, K.H., Angelidaki, I., Ahring, B.K. 1998. Anaerobic digestion of swine manure: inhibition by ammonia. *Water research*, **32**(1), 5-12.
18. Labatut, R.A., Angenent, L.T., Scott, N.R. 2014. Conventional mesophilic vs. thermophilic anaerobic digestion: a trade-off between performance and stability? *Water research*, **53**, 249-258.
19. Lauterböck, B., Nikolausz, M., Lv, Z., Baumgartner, M., Liebhard, G., Fuchs, W. 2014. Improvement of anaerobic digestion performance by continuous nitrogen removal with a membrane contactor treating a substrate rich in ammonia and sulfide. *Bioresource technology*, **158**, 209-216.
20. Markou, G. 2015. Improved anaerobic digestion performance and biogas production from poultry litter after lowering its nitrogen content. *Bioresource technology*, **196**, 726-730.
21. Mata-Alvarez, J., Dosta, J., Macé, S., Astals, S. 2011. Codigestion of solid wastes: a review of its uses and perspectives including modeling. *Critical Reviews in Biotechnology*, **31**(2), 99-111.
22. Mata-Alvarez, J., Dosta, J., Romero-Güiza, M., Fonoll, X., Peces, M., Astals, S. 2014. A critical review on anaerobic co-digestion achievements between 2010 and 2013. *Renewable and sustainable energy reviews*, **36**, 412-427.
23. McCarty, P.L., McKinney, R.E. 1961. Volatile acid toxicity in anaerobic digestion. *Journal (Water Pollution Control Federation)*, 223-232.
24. McKendry, P. 2002. Energy production from biomass (part 2): conversion technologies. *Bioresource technology*, **83**(1), 47-54.
25. Milan, Z., Sanchez, E., Weiland, P., de Las Pozas, C., Borja, R., Mayari, R., Rovirosa, N. 1997. Ammonia removal from anaerobically treated piggery manure by ion exchange in columns packed with homoionic zeolite. *Chemical Engineering Journal*, **66**(1), 65-71.
26. Müller, N., Worm, P., Schink, B., Stams, A.J. and Plugge, C.M., 2010. Syntrophic butyrate and propionate oxidation processes: from genomes to reaction mechanisms. *Environmental microbiology reports*, **2**(4), 489-499.
27. Neiva Correia, C., Vaz, F., Torres, A. 2008. Anaerobic digestion of biodegradable waste–operational and stability parameters for stability control. *5th IWA International Symposium on AD of Solid Wastes and Energy Crops, Tunisia*.
28. Nielsen, A.M., Christensen, K.V., Møller, H.B. 2013. Inline NH 3 removal from biogas digesters. *Biomass and Bioenergy*, **50**, 10-18.
29. Pedizzi, C., Lema, J.M., Carballa, M. 2017. Enhancing thermophilic co-digestion of nitrogen-rich substrates by air side-stream stripping. *Bioresource Technology*.
30. Perry, R.H., Green, D.W. 1999. Perry's Chemical Engineers' Handbook. 7th ed. McGraw-Hill.
31. Pintucci, C., Carballa, M., Varga, S., Sarli, J., Peng, L., Bousek, J., Pedizzi, C., Ruscalleda, M., Tarragó, E., Prat, D. 2017. The ManureEcoMine pilot installation: advanced integration of technologies for the management of organics and nutrients in livestock waste. *Water Science and Technology*, **75**(6), 1281-1293.
32. Rajagopal, R., Massé, D.I., Singh, G. 2013. A critical review on inhibition of anaerobic digestion process by excess ammonia. *Bioresource technology*, **143**, 632-641.
33. Resch, C., Wörl, A., Waltenberger, R., Braun, R., Kirchmayr, R. 2011. Enhancement options for the utilisation of nitrogen rich animal by-products in anaerobic digestion. *Bioresource technology*, **102**(3), 2503-2510.
34. Ripley, L., Boyle, W., Converse, J. 1986. Improved alkalimetric monitoring for anaerobic digestion of high-strength wastes. *Journal (Water Pollution Control Federation)*, 406-411.
35. Serna-Maza, A., Heaven, S., Banks, C. 2015. Biogas stripping of ammonia from fresh digestate from a food waste digester. *Bioresource technology*, **190**, 66-75.
36. Serna-Maza, A., Heaven, S., Banks, C.J. 2014. Ammonia removal in food waste anaerobic digestion using a side-stream stripping process. *Bioresource technology*, **152**, 307-315.
37. Serna-Maza, A., Heaven, S., Banks, C.J. 2017. In situ biogas stripping of ammonia from a digester using a gas mixing system. *Environmental Technology*, 1-9.
38. Suhartini, S., Heaven, S., Banks, C.J. 2014. Comparison of mesophilic and thermophilic anaerobic digestion of sugar beet pulp: performance, dewaterability and foam control. *Bioresource technology*, **152**, 202-211.
39. Sun, Z.-Y., Yamaji, S., Cheng, Q.-S., Yang, L., Tang, Y.-Q., Kida, K. 2014. Simultaneous decrease in ammonia and hydrogen sulfide inhibition during the thermophilic anaerobic digestion of protein-rich stillage by biogas recirculation and air supply at 60 C. *Process Biochemistry*, **49**(12), 2214-2219.
40. Walker, M., Iyer, K., Heaven, S., Banks, C.J. 2011. Ammonia removal in anaerobic digestion by biogas stripping: an evaluation of process alternatives using a first order rate model based on experimental findings. *Chemical Engineering Journal*, **178**, 138-145.
41. Walker, M., Zhang, Y., Heaven, S., Banks, C. 2009. Potential errors in the quantitative evaluation of biogas production in anaerobic digestion processes. *Bioresource Technology*, **100**(24), 6339-6346.
42. Yabu, H., Sakai, C., Fujiwara, T., Nishio, N., Nakashimada, Y. 2011. Thermophilic two-stage dry anaerobic digestion of model garbage with ammonia stripping. *Journal of bioscience and bioengineering*, **111**(3), 312-319.
43. Yenigün, O., Demirel, B. 2013. Ammonia inhibition in anaerobic digestion: a review. *Process Biochemistry*, **48**(5), 901-911.
44. Yirong, C., Banks, C., Heaven, S. 2013. Effect of ammonia nitrogen on thermophilic anaerobic digestion of food waste. *21st European Biomass Conference and Exhibition Copenhagen (Denmark)*.
45. Yirong, C., Heaven, S., Banks, C. 2015. Effect of a trace element addition strategy on volatile fatty acid accumulation in thermophilic anaerobic digestion of food waste. *Waste and Biomass Valorization*, **6**(1), 1-12.
46. Zhang, L., Lee, Y.-W., Jahng, D. 2012. Ammonia stripping for enhanced biomethanization of piggery wastewater. *Journal of hazardous materials*, **199**, 36-42.
47. Zheng, H., Li, D., Stanislaus, M.S., Zhang, N., Zhu, Q., Hu, X., Yang, Y. 2015. Development of a bio-zeolite fixed-bed bioreactor for mitigating ammonia inhibition of anaerobic digestion with extremely high ammonium concentration livestock waste. *Chemical Engineering Journal*, **280**, 106-114.

**Table 1** Stages and changes in key operational conditions

|  |  |  |
| --- | --- | --- |
| Stage | Day | Operational change |
| 1 |  | All digesters operated under the same conditions |
|  | 0 | Start of feeding on low N food waste at 0.5 kg VS m-3 day-1 |
|  | 0-20 | Incremental loading increase to 1.5 kg VS m-3 day-1 |
|  | 21 | Feed switched to source segregated domestic food waste at 2.0 kg VS m-3 day-1Ammonia stripping started at 2.1 kg of digestate twice a week for each digester T1-4 (equivalent to 1.7% of digester volume per day). |
| 2 | 108 | Stripping regime in T1 and T2 changed to 2.5 kg twice a week (equivalent to 2.0% of digester volume per day); T3 and T4 continue as Stage 1. |
|  | 120 | New batch of source segregated domestic food waste |
|  | 178 | One-off dose of trace element solution added to T4 |
| 3 | 210 | Stripping regime changed: T2 increased to 5.0 kg twice a week (using 2 stripping towers; equivalent to 4.1% of digester volume per day). Stripping stopped for T4; T1 and T3 continue as Stage 2. |
|  | 245 | New batch of source segregated domestic food waste |
|  | 346 | Feeding and ammonia stripping stopped in all digesters  |
| 4 | 370 | End of monitoring |

**Table 2** Physico-chemical characteristics of first batch of substrate

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Parameter** | **Units** | **Value** |  | **SD**  |
| Total solids | g TS kg-1 WW | 238.5 | ± | 1.2 a |
| Volatile solids | g VS kg-1 WW | 206.8 | ± | 2.1 a |
| TKN | g N kg-1 WW | 7.0 | ± | 0.12 |
| Elemental C  | % of TS | 50.28 | ± | 0.75 |
| Elemental H  | % of TS | 6.37 | ± | 0.07 |
| Elemental N  | % of TS | 3.68 | ± | 0.09 |
| Carbohydrate | g kg-1 VS | 492.1 | ± | 12.5 |
| Lipid | g kg-1 VS | 197.0 | ± | 2.2  |
| Crude protein | g kg-1 VS | 211.2 | ± | 3.7  |
| Calorific value | MJ kg-1 TS | 22.0 | ± | 0.06 |
|  | MJ kg-1 VS | 25.4 | ± | 0.06 |

SD = standard deviation for triplicate samples unless noted.

a 8 samples

**Figure captions**

**Figure 1** Schematic of the coupled anaerobic digestion and side-stream ammonia stripping process.

**Figure 2** Monitoring parameters during stage 1 of the experimental period: (a) pH, (b) TAN, (c) TA, (d) PA, (e) IA, (f) IA/PA, (g) VS as %WW, (h) TS as % WW, (i) VS as %TS, (h) total VFA concentrations.

**Figure 3** Monitoring parameters during stage 1 of the experimental period: Individual VFA species and TAN concentrations in (a) T1. (b) T2, (c) T3, (d) T4, and (e) VBP and (f) SMP in all 4 digesters.

**Figure 4** Monitoring parameters during the whole experimental period: (a) TAN, (b) TA, (c) PA, (d) TS as % WW, (e) VS as % WW, (f) VS as % WW, (g) VBP, (h) SMP, (i) pH and (j) total VFA concentrations. Vertical dotted lines indicate ends of stages.

**Figure 5** Individual VFA species and FAN concentration in (a) T1. (b) T2, (c) T3, (d) T4, during the whole experimental period (note change of axes for VFA in T4). Vertical dotted lines indicate ends of stages.

**Figure 6** Stripping parameters: (a) TAN concentrations in stripped digestate, (b) TAN removal rates in stripper during experimental period, (c) TAN concentration and (d) % TAN removal over the 3-day stripping period starting on day 315, (e) TKN and organic N in digesters during experimental period. Vertical dotted lines indicate ends of stages.