**Thermophilic digestion of food waste by dilution: ammonia limit values and energy considerations**

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**Abstract**

Source segregated domestic food waste (FW) collected by Veolia Environmental Services was used as a test substrate for thermophilic anaerobic digestion in a laboratory-scale trial with semi-continuous feeding at an organic loading rate of 3 g volatile solids (VS) L-1 day-1 reached after an acclimatization period of 60 days. The FW had a Total Kjeldahl Nitrogen content of 0.73% on a wet weight basis which without dilution gave a digestate total ammonia nitrogen (TAN) concentration of >5.0 g N L-1. This led to the accumulation of propionic and other longer-chain volatile fatty acids (VFA), followed by a sharp increase in acetic acid which was sufficient to overcome the ammonia buffering capacity, causing the pH to fall and gas production to cease. In digesters run in parallel, TAN concentrations were regulated by diluting the FW with water. This allowed determination of critical threshold concentrations for TAN, and for free ammonia nitrogen (FAN) by calculation, and monitoring of the pattern of VFA production. Below 2.5 g N L-1 TAN there was no evidence of digestion instability. Between 2.5 and 3.5 g N L-1 TAN transient peaks in VFA could be seen, but without long-term accumulation; above 3.5 g N L-1 TAN continuous accumulation of VFA occurred which eventually led to failure. Stable digestion could be maintained by a dilution of 0.5:1 water to FW. The energy implications of using a dilution strategy were evaluated using the ADAT modelling tool for a range of scenarios at dilution ratios of 0.5-3:1, ambient temperatures of 5-35 oC and different plant sizes. As expected dilution had the largest effect on the net energy demand, but at the lowest dilution required to overcome toxicity this only equated to 2-6% loss of the raw energy potential of the biogas produced from the FW when compared to mesophilic digestion with pasteurization.

**Keywords**: ammonia, anaerobic digestion, thermophilic, toxicity, inhibition, dilution

**1 Introduction**

Anaerobic digestion (AD) of food waste has grown in popularity as the energy value of this digestion substrate is more widely recognised 1 and the use of these materials into animal feeds becomes increasingly regulated in many parts of the world due to concerns over disease transmission. The potential for nutrient recovery and the benefits of using digestion to close the loop between urban food waste generation and the fertiliser requirements of agriculture are also important drivers for the promotion and adoption of AD 2.

Food residues of many different types arise in the food production chain from land or sea to the market place, each having specific characteristics that may or may not make them suitable substrates for AD. In the US food waste is estimated to make up 15% or more of the municipal waste stream 3, while in European countries this figure is typically 20-25% and in China the proportion may be as high as 50% 4: this material is therefore available in substantial quantities. While post-consumer food waste from different countries differs in appearance, from the perspective of its biochemical composition it has some similarities, as human dietary inputs tend to be balanced in terms of their protein, carbohydrate and fat contents, particularly in the developed regions of the world 1, 5.

In the UK the first demonstration-scale digester treating source segregated food waste from domestic and commercial properties was built in 2006 6, and formed the basis of an industry that processed 1.6 million tonnes of waste per year in 400 plants generating 480 MW of energy in 2016 in the UK alone 7. The path of commercialization was not without difficulty, however, and one of the major problems was high ammonia concentrations in the digester that could lead to inhibition and ultimately failure of the process.

The issue of ammonia toxicity has been recognised for many years, as detailed in two recent review papers 8, 9. It is now generally accepted that in mesophilic digestion the acetoclastic methanogens are more sensitive to ammonia than the hydrogenotrophs 10-13. There have been numerous reports in the literature of acclimatization of digesters to ammonia 8, 14. It is likely, however, that under mesophilic conditions this is adaptive acclimatization resulting from community structure changes in which the balance between acetoclastic methanogens and hydrogenotrophic methanogens coupled to acetate oxidisers shifts in favour of the later 15-18. In food waste digestion this switch in population was seen to be stimulated by addition of Selenium which unlocked the acetate oxidation route and prevented propionic acid accumulation 16. Digestion at high ammonia concentrations is associated with the predominance of a hydrogenotrophic population, and in particular *Methanomicrobiales* 19: this was also found to be the dominant group in an analysis of full-scale digesters operating at total ammonia nitrogen (TAN) concentrations above 2.8 g-1 N L-1 or 0.44 g-1 N L-1 of free ammonia nitrogen (FAN) 20.

In operational terms tolerance to ammonia has been variously reported in the literature a wide range of values quoted 8, 9. This is not surprising when TAN (NH3 plus NH4+) concentrations are reported, as the toxicity is due to free ammonia nitrogen (FAN, NH3) with the equilibrium concentration depending at least upon pH and temperature. Under mesophilic conditions threshold FAN values of around 1.0 g N L-1 have been reported in well-controlled experiments where ammonia concentrations have been raised using an external nitrogen source such as urea 19. In a typical mesophilic digester treating source segregated food waste from households or from the retail sector the TAN concentration will equilibrate at between 5-6 g N L-1 6 equivalent to FAN of ~0.75 g N L-1, and below the mesophilic inhibition threshold provided that adequate trace element supplementation is supplied 16. The same food waste added to a thermophilic digester at 55 oC may cause severe inhibition as the FAN will rise above 1 g N L-1, in excess of threshold inhibitory values 21.

Yet the thermophilic digestion of food waste may offer some advantages if the limitation of ammonia toxicity could be overcome. These may include achieving higher rates of digestion; greater conversion of waste organics to gas; improved solid liquid separation; and minimization of bacterial and viral pathogen accumulation 8. Acclimatization to increasing ammonia concentrations appears not to be possible for this substrate in thermophilic conditions, despite long-term exposure 22, 23. The alternative is to reduce the ammonia concentration, and the two main approaches are based either on stripping ammonia from the liquor 24-26; or more commonly on dilution to below the toxicity threshold 27. The latter approach, however, results in an increase in the volume of material to be treated, which in turn can make the process economically unattractive 28 and energetically less favourable. As an alternative to water addition, co-digestion with a low nitrogen material also serves to ‘dilute’ the TAN: this strategy was also demonstrated by Nielson and Angelidaki 29 as a means of recovery from ammonia inhibition in the thermophilic digestion of cattle manure. Co-digestion has the benefit of gaining further energy from the added carbon, and is therefore likely to be preferable in terms of the overall energy balance. In the EU the additional energy inputs required for thermophilic digestion of diluted food waste materials may be partially offset by the potential for omitting the 70 oC pasteurization stage which is required for compliance with Animal By-Products Regulations (EC 1069/2009) when operating under mesophilic conditions. Thermophilic digestion of a more dilute mixture may also offer improvements in mixing behaviour and solids separation, and increase the potential for heat recovery from the effluent. Both of these gains, however, are only compensatory and further consideration of energy balances is needed to elucidate the value of this approach.

The current study had two main aims: firstly, to test the threshold inhibition concentration for TAN in food waste digestion at 55 oC using diluted municipally-collected source segregated food waste and to assess the performance of the digestion process under these conditions; and secondly, to make a preliminary assessment of the energy implications of adopting this type of strategy.

**2 Materials and methods**

2.1 Feedstock

The feedstock was source segregated domestic food waste (FW) collected from households in Eastleigh, Hampshire, UK by Veolia Environmental Services (UK) Ltd. A sample of around 300 kg was removed from the biodegradable bags in which it is collected. Any obvious non-food contamination was removed along with any large bones and seeds. The sample was then ground (S52/010 Waste Disposer, IMC Limited, UK) to a homogeneous pulp, mixed thoroughly and frozen at -18 °C in ~3 kg portions. When needed, the feedstock was thawed and stored at 4 °C and used within a period of a few days. This source of feedstock has been used in a number of trials 21, and the key characteristics of the current batch are shown in Table 1.

2.2 Digester construction, start-up and operation

Three pairs of 4-litre working volume anaerobic digesters were used, of a continuously-stirred tank reactor (CSTR) design. These were constructed of PVC tube with gas-tight top and bottom plates. The top plate was fitted with a gas outlet, a feed port sealed with a rubber bung, and a draught tube liquid seal through which an asymmetric bar stirrer was inserted with a 40 rpm motor mounted directly on the top plate. Digester temperature was maintained at 55 ± 1 oC by circulating water from a thermostatically-controlled bath through heating coils around the digesters. Biogas was measured using tipping-bucket gas counters with continuous data logging. Gas counter calibration was checked regularly by collecting the gas produced over a one-day period in a gas-impermeable bag and measuring its volume in a weight-type gasometer 30. All gas volumes reported are corrected to standard temperature and pressure (STP) of 0 oC, 101.325 kPa.

The digesters were initially filled with inoculum from a mesophilic digester treating municipal wastewater biosolids (Millbrook, Southampton UK). The inoculum was supplemented with trace elements to give an added concentration of 0.8 mg L-1 Selenium and 4 mg L-1 Cobalt; these values are higher than typically used in mesophilic digestion as it has been suggested that thermophilic systems have higher TE requirements 31. These concentrations were maintained throughout the run by weekly addition of trace element solutions in proportion to the total volume of daily feed (i.e. including water). The temperature was adjusted to 55 oC in a single step and the digesters were left for 5 days without feeding. The organic loading rate (OLR) was then raised gradually from 0.5 g VS L-1 day-1 to the target of 3 g VS L-1 day-1 over a period of 51 days, using FW diluted with tap water at a ratio of 2:1 (water : FW) on a wet weight (WW) basis. In all cases feed was added once per day and seven days per week.

After day 62 the feeding regime was modified so that in one pair of digesters (T1 and 2) the feed was added without water (dilution 0:1); in one pair (T3 and 4) the dilution was reduced to 0.5:1; and in one pair (T5 and 6) the dilution remained at 2:1 as before. On day 164 after T5&6 had completed more than 4 HRT at a 2:1 dilution, the dilution in these digesters was reduced to 1:1 to provide a further set of operating conditions for assessment. The experiment ran for a total of 238 days, and the OLR and HRT in each set of digesters during this period is shown in Figure 1.

2.3 Analytical methods

Total and volatile solids (TS and VS) were measured according to Standard Method 2540 G 32 using a Heraeus Function Line Series oven and a 201/301 Carbolite muffle furnace. pH was determined 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) 33. Total ammonia nitrogen (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. 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% using 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. 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 containing 65% CH4 and 35% CO2 (v/v) (BOC, UK).

2.4 Modelling

Dilution scenarios were modelled using ADAT, a mass and energy balance modelling tool that was developed with support from various sources including the FP6 CROPGEN project (SES6-CT-2004-502824), RCUK RELU (RES-229-25-0022), FP7 VALORGAS (241334), BBSRC ADNet (BB/L013835/1) and IEA Task 37 (UK), and which is freely available for download from www.bioenergy.soton.ac.uk/resources.htm. The modelling tool takes account of energy requirements for feedstock heating, pre or post-pasteurisation and heat losses through the floor, walls and roof of the digester. It includes the estimated parasitic energy demand for mixing and pumping, and from fugitive methane losses in the process. It also takes into account the efficiency of the CHP plant or boiler, and/or the energy use in upgrading and compressing biogas (as appropriate). The modelling tool can also include energy requirements for transport and pre and post-processing of feedstock and digestate fractions, energy offset savings from fertiliser substitution and embodied energy in the major plant components but these were not included in the current work unless noted in the text. The model is written in C# and compiled with a user interface allowing input values to be modified. Unless noted, the default values in the modelling tool were used.

Modelling was carried out for a user-defined feedstock was with a specific methane production (SMP) of 0.45 L CH4 g-1 VS added, 60% CH4 in the biogas, TS 23.9%, VS 90.5% of TS, nutrient values 31 g N kg-1 TS, 4 g P kg-1 TS, 13 g K kg-1 TS, and 10 kWh tonne-1 parasitic energy for feedstock (not including dilution). These properties were based on the current feedstock, but are closely similar to default values for source segregated domestic food waste in the ADAT modelling tool. The SMP value used for modelling mesophilic digestion was the same as that for thermophilic, as previously reported for this FW feedstock 23. Digester design was based on an OLR of 3 g VS L-1 day-1, now regarded as fairly moderate for food waste digestion. The digester was assumed to be a steel tank with 50 mm of high performance insulation (typical U-value 0.0245 W m-1 K-1, giving a composite heat transfer coefficient of 0.49 W m-2 K-1), and a double membrane roof with a composite heat transfer coefficient of 1 W m-2 K-1 34. Digester temperature was set at 55 oC for thermophilic operation and 37 oC for mesophilic. Modelling was carried out for default conditions in Southampton, UK (annual average air and soil temperatures 10.6 oC), and also at annual average temperatures from 5-35 oC. Unless noted, it was assumed there was no separate pasteurization step, as at 55 oC compliance with the Animal By-product Regulations **(EC 1069/2009**) may be achieved by providing a guaranteed HRT, e.g. through intermittent feeding and discharge. It was assumed that the biogas produced was used on site in a Combined Heat and Power (CHP) plant.

**3 Results and discussion**

3.1 Start-up and acclimatization (day 0- 62)

Following the single-step increase in operating temperature there were some signs of process disturbance. Total VFA concentrations in all digesters rose to a peak of around 2.6 g L-1 by day 3. This was accompanied by IA/PA ratios of between 0.9-1.1 (Figure 2a), although pH remained unaffected at around 7.5 (Figure 2b). When feeding began on day 6 the pH and TAN concentrations rose to between 7.9-8.1 and 2.1-2.2 g N L-1 respectively, indicating that effective hydrolysis of the feedstock was occurring. Biogas production began, and volumes increased in line with the increasing OLR (Figure 2c). Over the following 6 weeks TAN gradually declined to around 1.6 g N L-1, probably as a result of uptake by microbial biomass in response to the increasing OLR and reducing HRT. TA, PA and IA fell slightly in all digesters reaching values of around 7.4, 5.2 and 2.2 g CaCO3 L-1 respectively by the end of the acclimatization period, with the IA/PA ratio dropping to around 0.4. The nature of the digestate solids also changed in this period, with a slight fall in TS and VS concentrations (Figure 2d) and an increase in VS/TS ratio (Figure 2e), reflecting the change from the previous wastewater biosolids feed to a dilute feedstock with a high VS content.

VFA profiles in this period showed an interesting transition, which was almost identical in all digesters (Figure 3) and probably reflected changes in the microbial community structure as the slower-growing methanogens came into balance with the acidogenic and acetogenic populations. There was an initial peak in acetic acid of around 1.9 g L-1, suggesting that the temperature shock had disturbed the acetoclastic methanogenic population. This fell to less than 0.1 g L-1 by day 22-27 indicating that acetate consumption rates, either through the acetoclastic route or via acetate oxidation, had recovered to match the rate of production. Propionic acid concentrations increased to around 1 g L-1 up to day 27, indicating firstly that the capacity of the syntrophic hydrogenotrophic methanogenic community was overloaded at this point; and secondly that the accumulation of longer chain VFA seen between days 0-15 (Figure 3a-f) was beginning to reduce through conversion to propionate. Concentrations of all VFA had reached very low values by days 30-35 indicating that populations had stabilised in all digesters. A similar initial response was seen in previous work on acclimatization of inoculum from the same source to thermophilic digestion of a similar but undiluted food waste feedstock 21; but in that case after 40 days ammonia concentrations had reached inhibitory values, causing a second increase in propionic acid which eventually led to digester failure.

By day 62 at the end of the acclimatization period and after the OLR had reached its target value the process was operating stably with a pH of around 7.6, TAN 1.6 g N L-1, and VFA around 0.1 g L-1. The biogas methane content was steady at around 60% and SMP had reached 0.400 L CH4 g-1 VS added (Figure 2f), similar to the start-up value reported for this feedstock in other work 21 and within 10% of the long-term steady-state value. During the start-up period values for all monitoring parameters showed very similar behaviour in all 6 digesters and by day 62 all digesters were considered to be successfully acclimated to the new feedstock and thermophilic conditions.

**3.2 Digester performance and operational stability (days 63 - 238)**

On day 62 the dilution regime was changed with the aim of allowing TAN concentrations in one pair of reactors (T1&2) to increase to the point of failure. The feedstock to another pair of reactors (T3&4) was diluted to a level at which some instability was expected, while one pair (T5&6) was maintained at the previous ‘safe’ dilution until day 164.

In T1&2 (dilution 0:1) TAN rose steadily and exceeded 5.0 g N L-1 by day 215 (Figure 4a), equivalent to 2 HRT in this operating mode. In T3&4 (dilution 0.5:1) TAN concentrations began to stabilise by day 160, and remained steady at around 3.1 g N L-1 until the digesters reached 3 HRT on day 215. T5&6, which remained at dilution 2:1, reached 3 HRT at day 130 with TAN concentrations stabilising at around 1.5 g N L-1. On day 164 the dilution in these digesters was changed to 1:1 and the TAN concentration rose, reaching 2.3 g N L-1 by day 238 after 2 HRT in this operating mode.

Total VFA (TVFA) showed a clear response to the changes in TAN concentrations (Figure 4b). In T1&2 minor fluctuations in TVFA appeared from day 80 onward, when the TAN reached 2.2 g N L-1; from day 120 when TAN reached 3.5 g N L-1 there was a steady rise until the end of the experiment. In T3&4 during the period between day 160-215 TVFA increased from its previous baseline value to around 0.5 g L-1, with short-term fluctuations up to 1.6 g L-1. Between day 75 and 164 the average TVFA concentration in T5&6 was < 0.15 g L-1. The TVFA profiles indicated that T3&4 were showing signs of stress at TAN concentrations of 3.1 g N L-1, but without VFA accumulation; whereas there were clear signs of metabolic inhibition in T1&2 once the TAN concentration exceeded 3.5 g N L-1.

Alkalinity parameters confirmed the above observations. As expected the TA concentration closely mirrored the TAN (Figure 4c). IA remained relatively stable in digesters T5&6 and T3&4 throughout the experimental period (Figure 4d), reflecting the absence of VFA accumulation, while in T1&2 there was a rapid increase in IA clearly related to the rise in VFA. The IA/PA ratio was below 0.4 for T3&4 and T5&6 (Figure 4e), a typical value for stable operation of food waste digesters; whereas in T1&2 this ratio showed a rising trend, with a very rapid increase where VFA accumulation finally broke the buffering capacity of the digester. This led to a rapid drop in pH (Figure 4f) and PA (not shown). The available alkalinity was also reflected in the pH values, which were lower in T5&6 than in T1&2 or T3&4 until the dilution in T5&6 was reduced and the TAN and PA increased.

After day 100 volumetric biogas production (VBP) in T3&4 and T5&6 fluctuated around 2.2 L L-1 day-1 (Figure 4g) with SMP of around 0.44 L CH4 g-1 VS (Figure 4h). Although T1&2 continued to produce biogas, the VBP gradually decreased to around 2.0 L L-1 day-1 in both digesters by day 150, and SMP fell to 0.39 L g-1 VS. After day 220 biogas production in T1 fell rapidly, corresponding to the period when the pH dropped from 7.9 to 6.8 and the IA/PA ratio rose from 0.98 to 3.1. By day 233 the biogas methane content for T1 had fallen below 50 %. These results demonstrated that an anaerobic digester can appear to operating normally even at relatively high TAN concentration under thermophilic conditions, as long as there is sufficient alkalinity to buffer pH changes. Examination of the data, however, clearly shows metabolic instability as indicated by VFA accumulation. The higher threshold of inhibition for the process can thus be taken as the point at which there is an irreversible increase in VFA concentration. In this particular study this occurred at a TAN concentration of around 3.4 g N L-1, yet signs of instability were obvious at 3.1 g N L-1. These values are reported as TAN concentrations as this was the parameter measured, but correspond to a calculated FAN of 0.82 and 0.57 based on the measured operating temperature and pH 35.

At the end of the acclimatization period the digestate TS content was 2.3 % of wet weight (WW) and this continued to fall in T5&6, reaching around 1.8 % WW by the end of 3 HRT. TS concentrations in these digesters began to rise again after the feedstock dilution was decreased to 1:1, reaching a final value of 2.0%. The TS content rose in T3&4 and in the period between day 160-215 stabilised at 3.0 %WW. T1&2 showed a rising trend in TS content, reaching 5.9% by the end of the experiment without achieving a steady state. This corresponded to the period of falling gas production and indicates that the degree of solids destruction was also declining. VS content in T1&2 followed similar trends to TS and the VS/TS ratio was slightly higher than in T3&4 and T6&6, again indicating less efficient conversion of VS.

# VFA profiles for T1&2 (Figure 5a and b) showed that the increase in TVFA concentration from day 120 onwards was due mainly to accumulation of propionic acid, which reached around 12 g L-1 by the end of the experiment; although iso-butyric and iso-valeric concentrations also rose to between 1-2 g L-1. At the beginning of this period acetic acid concentrations showed no consistent upward trend in T1 or T2. After day 215, however, the acetic acid concentration in T1 rose very sharply reaching 14 g L-1 by day 233. VFA concentrations in T2 lagged slightly behind T1 throughout the preceding period (Figure 4d), but by the end of the experiment also showed signs of an increase in acetic acid. The appearance of acetic acid clearly indicates severe disruption of the conversion to methane by either the acetoclastic route or through acetate oxidation. The sequence of VFA accumulation suggests that the syntrophy between acetogens and methanogens had become imbalanced. The mechanism for this was not further investigated, but it is likely that under high TAN concentrations methane formation was primarily through acetate oxidation; and any increase in hydrogen concentrations may lead to feedback inhibition of the acetogenic propionate-degrading bacteria as the thermodynamics of the reaction become less favourable. VFA profiles in T3&4 showed no accumulation of individual VFA (Figure 5c and d) but there were elevated levels of acetic acid with occasional transient peaks of propionic and a small increase in iso-valeric concentrations, all indicating mild instability. Although analysis of dynamic community structure was not carried out, this could provide a useful insight into whether this instability is related to the transition between acetoclastic and hydrogenotrophic pathways, as has been noted in mesophilic conditions at the point where ammonia becomes toxic; or to the onset of inhibition of acetate oxidation, as recent evidence suggests some syntrophic acetate oxidisers may be more sensitive than hydrogenotrophic methanogens to high TAN concentrations 36. After the initial acclimatization period T5&6 showed consistently low concentrations of all VFA species up to day 164 (Figure 5e and f) apart from a transient spike in acetic acid on day 78, the reason for which is unknown. When the dilution in this pair of digesters was reduced to 1:1 the acetic acid concentration rose slightly from day 200, indicating the onset of mild instability at a TAN concentration corresponding to 1.8-1.9 g N L-1 (FAN 0.5 g N L-1)

The results indicated that digester operation could be carried out without VFA accumulation at TAN concentrations below 3.1 g N L-1, as achieved by a 0.5:1 (water:FW) dilution. At this TAN concentration some signs of incipient instability were observed, indicated by a slight rise in baseline VFA concentrations with transient VFA peaks. It can be argued that this makes a digester more vulnerable to minor shocks in operation, such as short-term temperature shifts or changes in feed composition: a more cautious approach in terms of process stability would therefore involve increasing the dilution to give a TAN concentration below the lower threshold for onset of instability, corresponding in this study to FAN of around 0.5-0.6 g N L-1. The experimental data showed that at the OLR used dilution of the input FW did not affect the specific methane production, despite a reduction in the retention time from 75 days using undiluted material to 25 days at a dilution of 2:1. The dilution strategy adopted would therefore not change the required digester volume or surface area, and is thus unlikely to have a major effect on heat losses (as confirmed with the assumptions used in the modelling work) or on the capital cost associated with the digester, although the required capacity of ancillary equipment such as heat exchangers, pumps and CHP systems may be affected.

3.3 Modelling

Rajagopal et al. 9 commented that in anaerobic digestion dilution is commonly considered as a solution to ammonia toxicity, but this strategy has resource and energy implications that may be economically unfavourable. The actual impact, however, is very dependent on the energy value of the substrate, the availability of water, the potential for recycling of effluent after nutrient removal, and the final markets for the energy products. The most important consideration is the net energy yield, and where the energy potential of the substrate is low the energy requirement to raise the temperature of dilute materials may be greater than that embodied in the substrate. The energy potential in food waste is high, however, and the proportion that is required for digester heating is thus relatively small.

A number of scenarios were modelled to quantify the energy balance for food waste digestion. The first step was to simply assess the impact of dilution itself, and a thermophilic AD plant treating 2500-25000 tonnes year-1 was modelled using 0.5:1 to 3:1 dilutions of the input material. This was compared to a mesophilic plant with the same tonnage feed range but without dilution, and with or without a pre-pasteurisation stage at 70 oC for 1 hour. The final output energy (combined electricity and heat for export, after deduction of parasitic energy requirements) in all cases was expressed as a percentage of the energy available in the raw biogas produced. It can be seen (Figure 6a) that, while operation in thermophilic conditions with dilution does reduce the final energy output, the difference between thermophilic at 0.5:1 dilution and mesophilic digestion with a pre-pasteurization phase for ambient temperatures in Southampton, UK is only about 2% of the energy in the raw biogas. At a 3:1 dilution this difference would increase to around 15%, all of which is a demand for heat energy. Where pasteurization is not needed the difference in energy demand rises to around 6% and 19% at the smaller and larger dilution, respectively, but in the EU the pasteurization step would be required as a condition of the Animal By-Products Regulations (EC 1069/2009).

The heat loss from the plant is affected by the size of the digester as its surface area to volume ratio changes. The size of digester in this case was determined by the feedstock tonnage as a uniform loading rate was applied. For the range of tonnages considered digester volumes from 665 to 6647 m3 would be required. Above this size range it is common practice to use multiple units rather than a single digester, and smaller digesters may have a higher specification for insulation. The results showed that although the heat loss from the smallest plant was higher this was in fact a relatively minor impact (Figure 6a).

The parasitic heat demands taking into account heat loss and feedstock and diluent heating were compared to heat availability from a CHP unit. Under UK climatic conditions a mesophilic digester with pre-pasteurization processing 15000 tonnes year-1 (mid size-range) would consume 16.7% of the available heat output from the CHP unit. The equivalent thermophilic digester with a 0.5:1 dilution and no pasteurizer would consume 20.0%, a relatively small increase in comparison with mesophilic operation. At a 3:1 dilution the heat demand would increase to 46.4% of the CHP output, while for the worst case scenario (highest dilution of 3:1 and smallest plant at 2500 tonnes year-1) the heat demand rises to 49.9% of CHP output, still well within the heat generation capacity of the plant.

Different climatic conditions will also affect the final energy output. Figure 6b shows the effect of annual average ambient temperatures (air and soil) from 5-35 oC on a digester processing 15000 tonnes year-1 under mesophilic or thermophilic conditions, with the results expressed as a percentage of energy in the raw biogas. Again, there is an increased energy cost for thermophilic operation compared to the mesophilic/pasteurization combination, which is only between 0.2-2% of the energy at dilution 0.5:1 but rises to between 6-17% for 3:1 dilution.

When the energy output of the digester is electrical power from a CHP plant there is often no economic use for the heat produced, and if this is the case then thermophilic operation with dilution is a feasible alternative as no additional energy inputs are required. Even where CHP heat is used, for example in district central heating systems, the extra heat demand of thermophilic digestion at the lowest dilution is small. There are also other potential advantages of this mode of operation which may offset the additional energy requirement. There is some evidence that thermophilic digestion can improve the solids handling and dewatering properties of digestate 37-39, and dilution itself may also improve dewaterability. This in turn will influence the types of equipment used for mixing and pre and post-processing, and could affect the ancillary parasitic power demand, but little comparative data is available. Some of the additional energy demand for thermophilic operation could be compensated for by more efficient heat recovery, for example from the digestate discharge, as a result of the higher temperature gradients. The use of surplus heat from CHP may therefore be economically acceptable, although it should not be regarded as providing an environmental benefit in terms of fossil fuel replacement or avoided greenhouse gas emissions. Where the energy output is not as electrical power but through upgrading the biogas to biomethane as a fuel or for gas grid injection there is no surplus heat and the parasitic heat requirements of the digester have to be found by importing fuel or burning a proportion of the biogas in a boiler. The modelling tool allows gas upgrading as an alternative to CHP, but the final combined energy balance is the same as that reported here as the heat parasitic heat demands of the digester remain the same.

A number of other factors affect the overall energy balance and could also be taken into consideration in modelling. The modelling tool estimates the embodied energy in the plant, but in the current study the required digester volume is the same for any given input tonnage of FW, since the OLR was kept constant and the HRT allowed to alter at different dilutions. For feed inputs of 2500 to 25000 tonnes year-1 the embodied energy was equivalent to 0.9 to 0.4% respectively of the total energy in the raw biogas produced over the assumed working life of the plant (taken as 30 years for the digester and 15 years for gas utilisation equipment). Digestate transport costs were not included in the overall energy balance analysis, as these are highly site-specific; but it is recognised that transport of unseparated digestate would have some impact on the final energy balance. For example, if a travel distance of 15 km in a rigid 17-tonne vehicle is assumed for a 3:1 diluted digestate, transportation would account for 4.3% of the raw biogas energy. In practice, however, at higher dilutions it is likely that the digestate would be separated and treated to recover water or allow disposal on site or to sewer. Energy gains from fossil fuel displacement through N and P recovery are calculated by the modelling tool, and for the current FW could represent a contribution of 10.3% of the energy in the raw biogas. This figure was also not included in the final balance, as there are also associated energy costs that will differ considerably depending on whether recovery is by direct application of unseparated digestate onto land, or through on-site treatment of separated liquor and solids. The modelling tool allows input of an energy demand for post-treatment of separated liquors, but this option was not used in the present work.

The idea that using dilution to solve the problems of ammonia toxicity is unattractive does not appear to be well founded. There are clearly cases where the energy value of the waste and the final target energy market of the plant will allow thermophilic operation and still produce waste heat, even with small plant, high dilution and unfavourable climatic conditions. The difference in energy output between mesophilic and thermophilic digestion is in fact surprisingly small for food waste. The main reason for this is the requirement under mesophilic conditions to include a pasteurization step which requires heating of the material to 70 oC. FW is a special case, however, and for other high N wastes where a pasteurization step is not required the difference would be larger: for example, the energy differentials reported above of 0.2-2% at a 0.5:1 dilution and 6-17% for 3:1 dilution would rise to 4-10% and 10-24% respectively when thermophilic digestion is compared to mesophilic digestion without pasteurization. In the end it may not be energy alone that is the arbiter between thermophilic and mesophilic operation. Equal importance is now being placed on sustainability of resources, and nutrient capture and recycling is a high priority. Replacing the direct spreading of digestate with methods that allow recovery of nutrients as higher value-added products may be more feasible either as part of, or following, thermophilic digestion. Stripping and recovery of ammonia in the gaseous phase requires high temperatures 24, and process integration would favour thermophilic operation. Where recovery using membranes has been proposed 40, 41 this could be facilitated by working with more dilute material. Thermophilic operation is also well suited to the vacuum thermal stripping-acid absorption process described by Ukwuani and Tao 42, or simply to a direct aeration strategy 43. Work on electrochemical systems for ammonia removal 44, 45 is new and innovative but there are, as yet, no indications as to the temperature regime to which these techniques might be best suited. Nutrient recovery where the bio-derived product is competitive in its application and usage to the wide range of synthetic fertilisers on the market could, however, have resource and economic benefits that could outweigh any energy penalty.

As a final note it is worth mentioning that in practice a cost benefit analysis determines the degree of digester insulation used, and in all of the modelling scenarios used the amount of insulation on the mesophilic and thermophilic digesters was equal. It could, however, be improved to the point where the degree of heat loss from both thermophilic and mesophilic systems was the same, and at this point the energy balance for the thermophilic digester will be better than for a mesophilic digester with pasteurizer.

The suitability of dilution as a technique to solve ammonia toxicity problems is thus dependent on a number of factors that include the source and type of input material, the availability of surplus heat, the availability of a dilution medium, the energy market, and markets for recoverable products and every case should be considered against these criteria.

**4 Conclusions**

The thermophilic digestion of a typical source segregated food waste as collected in the UK in single pass mixed digesters with intermittent daily feeding leads to an increase in digester TAN concentrations to a point where the digestion fails. Diluting the feedstock proved a reliable method for establishing the critical TAN concentration at which instability was first observed (2.5 g N L-1) and also the point where incremental accumulation of propionic and other longer chain VFA began (3.5 g N L-1). Methane production continued until the digestion finally failed as a result of the pH falling to a critical level when the buffering capacity of the digester was overcome. This happened at a TAN concentration of ~5 g N L-1. Stable digestion without loss of specific or volumetric biogas production could be maintained at a 0.5:1 water:FW dilution. Energy modelling compared the net energy yields of thermophilic digestion to those of mesophilic digestion with and without a pasteurization stage at different dilutions, digester sizes, and environmental conditions. The results indicated that the impacts of dilution were relatively small for this energy-rich substrate where mesophilic digestion requires pasteurization and can easily be met from the energy available from use of the biogas in a CHP plant even at the highest dilutions, assuming there is no other economic use for the heat. Considering the other potential advantages of thermophilic systems in terms of improved rheology and dewaterability and the enhanced potential for advanced forms of nutrient recovery dilution may be an acceptable operating strategy.

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**Tables**

**Table 1** Food waste characteristics

|  |  |  |
| --- | --- | --- |
| Parameter | Unit | Value |
| Total solids (TS) | % wet weight (WW) | 23.9 |
| Volatile solids (VS) | % WW | 22.4 |
| VS/TS | % | 93.7 |
| Total Kjeldahl Nitrogen (TKN) | % WW | 0.73 |

**Figure captions**

**Figure 1** Operating conditions for FW digesters with/without dilution (a) OLR), (b) HRT.

**Figure 2** Selected monitoring parameters during acclimatization (day 0-62) (a) IA/PA ratio, (b) pH, (c) volumetric biogas production (VBP), (d) TS VS as % of wet weight (WW), (e) VS as %TS, (f) SMP.

**Figure 3** VFA profiles in all digesters during acclimatization (day 0-62).

**Figure 4** Selected monitoring parameters during experimental period (day 0-238) (a) TAN, (b) TVFA, (c) TA, (d) IA. (e) IA/PA ratio, (f) pH, and rolling 7-day averages of (g) VBP and (h) SMP. Vertical dotted lines indicate change in feed dilution.

**Figure 5** VFA profiles in all digesters during experimental period. Vertical dotted lines indicate a change in feed dilution. Note difference in y-axis scale for T1&2 compared to T3&4 and T5&6.

**Figure 6** Modelling results for final energy output with variations in (a) digester feed input (ambient temperatures Southampton, UK), (b) average ambient temperature, under thermophilic conditions at a range of dilutions (0.5:1. 1:1. 2:1, 3:1 water:FW) and in mesophilic conditions with no dilution with and without pre-pasteurization. Final energy output (electricity + heat) is expressed as % of energy in raw biogas produced