A MULTI-LEVEL BIOGAS MODEL TO OPTIMISE THE ENERGY BALANCE OF FULL-SCALE SEWAGE SLUDGE CONVENTIONAL AND THP ANAEROBIC DIGESTION

Jin Liu and Stephen R. Smith

Department of Civil and Environmental Engineering, Imperial College London, South Kensington Campus, London, SW7 2AZ, UK

Corresponding Author Email s.r.smith@imperial.ac.uk

Abstract

Anaerobic digestion (AD) is a long-established method for treating wastewater sludge and has been extensively researched, but there remains a lack of generic or practical modelling tools to guide operators and maximise the energy output. Detailed kinetic models have been developed, but are too complex as practical tools for industrial level application. A multi-level model of biogas yield (BY) was therefore developed based on operational data from 72 full-scale sites in the UK showing a wide range of AD performance. The model focused on the controllable operational parameters that are currently monitored at full-scale, including: temperature, hydraulic retention time and dry solids content in the feed sludge. The model effectively described performance variations in BY of full-scale processes, and provides a practical management tool to aid decision support to improve AD efficiency and net energy balance.

Keywords

Anaerobic digestion; biogas yield; digestion conditions; energy balance; process optimisation; sewage sludge

1. Introduction

The water industry consumes up to 3% of the total energy used (Howe, 2009), is the fourth most energy intensive sector (POST, 2007) and contributes approximately 1% of national greenhouse gas (GHG) emissions in the UK (Water UK, 2009); specifically, wastewater treatment contributes almost 60% of overall GHGs emitted by the industry (Ainger *et al*., 2009). However, the water industry is also a significant producer of renewable energy, for example, 20% of the energy consumed in Thames Water is from renewable sources supplied through anaerobic digestion (AD) of sewage sludge (Thames Water, 2019). Therefore, improving energy output is one of the key drivers for full-scale AD process management.

Anaerobic digestion is well established as a process for the stabilisation and treatment of residual sewage sludge from wastewater treatment. Scientific models of the AD process have been developed for almost 40 years, motivated by the need for more efficient operation (Donoso-Bravo *et al.*, 2011). The complexity of the system requires a modelling approach to balance the various influencing operational parameters, and specific models have been developed for different purposes (Kythreotou *et al.*, 2014). The simple stoichiometric equation first proposed by Buswell and Muller (1952) calculates the maximum biogas potential of the digestible substrates in sludge. The next generation of models focussed on the rate limiting step of the biochemistry and were based, for example, on the rates of conversion of fatty acids, methanogenesis or the hydrolysis of suspended solids (Eastman and Ferguson, 1981). These models were simple and easy to use, but did not adequately capture the overall process performance (Donoso-Bravo *et al.*, 2011). More complex models incorporate additional process steps, microbial species and detailed kinetics, including inhibitory mechanisms, based on improved microbiological understanding. For example, Hill (1982) used the volatile fatty acid (VFA) concentration as a key parameter and separated the kinetics of acidogenesis and acetogenesis into individual stages. More recently, the Anaerobic Digestion Model No.1

(ADM1), developed by Batstone *et al.* (2002), describes the dynamics of 24 species and includes 19 bioconversion processes, and aims to provide a generic model of fundamental AD mechanisms. Whilst valuable in research, a constant-volume and completely-mixed system is assumed by ADM1 and this is often difficult to achieve at full-scale (Kythreotou *et al.*, 2014). Moreover, the complexity and large number of input parameters restricts the application of dynamic models at a practical level for optimisation of full-scale industrial plant. Several authors have modified ADM1 for full-scale application to individual sites (Otuzalti and Perendeci, 2018; Ozgun, 2019) by reducing the number of input parameters. Nevertheless, a considerable amount of additional chemical information is still required, such as chemical oxygen demand, VFA and alkalinity, which are not routinely measured at full-scale sewage sludge AD plant.

The parameters that are typically available for process control are usually relatively limited and include: digestion temperature, hydraulic retention time (HRT), and the dry solids (DS) content of the digester feed sludge. However, sites with advanced mesophilic anaerobic digestion (MAD) often record additional sludge chemical properties: volatile solids (VS), VFA, and pH. The effects of these principal operational parameters on the AD process have been extensively studied individually in controlled laboratory experiments (Boušková *et al*., 2005; Alepu *et al*., 2016; Nielsen *et al*., 2017). However, it is less clear how digestion conditions affect the performance of full-scale, industrial AD plants, when interactive effects of multiple process variables exist. The development of artificial intelligence and deep learning algorithms, linked to artificial neural networks (ANN) has enabled the simulation of such complex non-linear systems. Indeed, several authors have developed ANN models of digester performance at full-scale sites (Qdais *et al*., 2010; Güçlü *et al*., 2011). For example, Güçlü *et al*. (2011) accurately predicted (R^2 = 0.71) the daily methane (CH_4) production volume using temperature, pH, sludge feed volume, VS, VFA and alkalinity as input variables, based on full-scale data collected over a 245 day period at Ankara Central wastewater treatment plant (WWTP). ANN models undoubtedly provide a major advancement in industrial systems control, however, they represent a 'black box' approach to process modelling and do not provide defined parameters to interpret the relationships between input and output variables (Dumitru and Maria, 2013). So far, ANN models have been successfully applied, but only to single, full-scale sewage sludge AD treatment sites and, consequently, reflect local operational performance conditions and may not be readily transferrable to other sites.

Modelling full-scale AD performance with limited operational parameters presents major challenges due to the complexity of the full-scale MAD process and relative differences in operational conditions, sludge composition and data recording between sites. However, it is possible to overcome these problems by extending conventional regression analysis techniques through the multi-level modelling of data from multiple sites with a hierarchical or clustered structure (Harrison *et al*., 2018).

The aim of this research, therefore, is to develop a statistically based, decision support tool, to predict and optimise AD performance using operational parameters available at full-scale sites, that can be applied by plant operators to optimise the biogas yield (BY) and energy balance of full-scale sewage sludge digestion processes. This was achieved by developing a multi-level model of BY based on operational data from 72 full-scale conventional and advanced, thermal hydrolysis process (THP) AD sites in the UK. Three calibration strategies were developed for the model to account for local site conditions, based on recorded BY and electricity yield (EY) data and the composition of major sludge organic constituents (protein, fat, carbohydrate and fibre). Finally, we applied the model to devise optimisation strategies to achieve the maximum net energy output from full-scale AD.

2. Material and Methods

2.1. Site and data information

Data were provided by 66 conventional and six THP MAD sludge treatment facilities in the UK. Operational data were recorded on a daily frequency for periods of 2 to 7 years, between 2009 and 2017. However, the information was collected and reported differently between the sites and the first stage was to consolidate the numerical information into a consistent format with equivalent units in a central database. A description of the different types of data recorded at the sites relating to the AD process is presented in Figure 1. The critical operational information available at all AD sites included: digestion temperature (^{o}C), HRT (d), and the DS content of the sludge feed (%). Gas volume was recorded as normal cubic metre (Nm^3) and was combined with sludge volume and DS data to obtain BY (m^3/t DS). The majority of sites also reported the volume of biogas distributed between combined heat and power (CHP), boiler and flare.

2.2. Sludge composition analysis

Three conventional MAD sites, representing high (Site 1), moderate (Site 31) and low (Site 38) BY performance (Figure 2), and one THP site (THP site 4), representing average THP performance, based on BY, were selected for sampling and sewage sludge composition analysis. Sludge samples were collected on six occasions from each site at intervals of 6-8 weeks from May 2018 to February 2019 for examination of the fibre, carbohydrate, protein, fat, DS and VS content. Total nitrogen (TN), ammonium-nitrogen (NH_4-N) and nitrate-nitrogen (NO_3-N) were determined by a standard Dumas method and EPA-600/4-79-020 method 350.1, respectively (USEPA, 1983). Protein was estimated by multiplying the organic nitrogen fraction (TN minus NH_4-N and NO_3-N) by 6.25 (Mariotti and Mirand, 2008). The total fat content was determined by standard procedure, 5520E (APHA, 2005) and the Van

Soest (1991) method was used to determine the proportion of cellulose, hemicellulose, and lignin in the fibre fraction. The difference in VS content and the sum of the various organic fractions (fibre, protein and fat) was assumed to represent the total carbohydrate concentration (Astals *et al.*, 2013). Sludge samples were collected at the digester feed, and post digestion.

2.3. Statistical analysis and model development

The overall approach to data merger and statistical analysis is shown in Figure 1. The IBM SPSS Statistics 21 programme was used to complete the statistical analysis calculations. A descriptive analysis and screening process was applied initially to the conventional and THP MAD datasets to remove extreme outliers larger than 3 times the interquartile range, using the boxplot method (Frigge *et al.*, 1989). The agreement with statistical assumptions was tested at three main levels: (1) significant outliers, high leverage points or highly influential points (leverage value >0.2 and Cook's Distance values >1); (2) independence of observations, linearity and multicollinearity; and (3) normality of residuals and homoscedasticity. The screened variables were converted into a consistent format based on monthly average values; this approach allowed the maximum data capture and provided a representative performance for each site by removing short-term fluctuations in the process variables.

An operational AD modelling strategy requires a large number of sites with different levels of process performance represented, to capture the complete envelope of conditions as comprehensively as possible, which cannot be achieved by studying single or small numbers of sites. Indeed, the observed BY of full-scale, conventional AD sites varied considerably (Figure 2) due to differences in actual performance, in response to the main process control variables, and also the influence of local site data measurement, as well as other operational reasons. Consequently, BY data from specific sites are strongly clustered (Figure S1) and it is not possible to derive a continuous, absolute statistical regression model with this type of numerical distribution pattern. Therefore, we used a multi-level regression method (Gelman and Hill, 2006; Gries, 2015) to identify the statistically significant (P<0.05), controllable operational parameters that impact BY, and to assess their individual and interactive effects, independently of other site-specific conditions. The multi-level model incorporated a varying intercept to identify the optimum slope coefficients for the significant AD process control parameters, for the combined data from all sites. The intercept value represents a categorical factor in the multi-level AD model, and is determined for each specific site data cluster by the model calibration procedure described below.

Backward elimination and forward selection methods (Leech *et al*., 2015) were used to identify statistically significant (P≤0.05), continuous predictor variables in multi-level regression models of BY. The data was examined in the following sequence of increasing model complexity: (1) linear regression, (2) curvilinear and non-linear regression (quadratic/cubic and log transformed), and (3) testing the interactive effects of the significant predictors (all combinations of interaction terms were tested). The predictors were centred before fitting into the model, to evaluate interaction effects, by subtracting the overall mean value (for the combined site data) from each variable (Aiken *et al.*, 1991). The general structures of the multi-level models tested are summarised in the Supplementary Material.

Validation of the conventional MAD model was performed using datasets collected from selected, specific WWTPs that were: (a) used in model development, and (b) obtained subsequent to, and independent of, model development. Sites 37, 38 and 42 (Figure 2), with typical average observed BY values of approximately 400 m^3/t DS (Table 1; CIWEM, 1996; Bachmann *et al.* 2015), were selected as good examples for model calibration and validation with information already used in model development, and Site 31, 38 and 1 were selected as representative, independent datasets. Conventional and THP MAD datasets were pooled and a combined Conventional+THP-MAD model was also developed and tested.

The default approach to model calibration, to account for site specific conditions for calendar year periods, estimated the deviation in mean predicted BY values relative to the observed mean recorded BY for the site, following Equation 2.1 (an example is shown in Figure S2):

$$\text{Site factor} = \text{Yearly average observed BY} - \left[\sum_{i=1}^{n} (\text{BY predicted by fixed coefficients})_i\right]/n \qquad (2.1)$$

Where n is the number of observed BY values each calendar year.

Biogas predictions were compared and cross-referenced with electricity generation data from AD biogas used by CHP plant. The mean annual observed EY also provided an alternative approach to model calibration for the selected sites by substituting the observed BY in Equation 2.1 with an electricity derived BY_e. This was calculated based on a conversion factor of 2.14 for electricity generation from biogas (assuming electrical conversion efficiency = 35% and 1 m^3 biogas = 2.14 kWh electricity; Banks, 2009). The BY equation was modified using the biogas-to-electricity conversion factor to predict EY and this alternative form of the model was also validated for the selected sites used in the conventional MAD BY validation. Finally, the results from the sludge composition analysis were used to calculate a theoretical BY_c value for the examined sites, based on the destruction of major organic fractions and their associated CH_4 yield values (Figure 1b; see Supplementary Material for further details). This provided an independent approach to model calibration by substituting BY_c for the observed BY in Equation 2.1 to obtain a sludge-composition derived site factor.

Response surface plots of the relative changes in BY were generated based on Model 4b, using representative combinations of values for two of the continuous variables within the 5 to 95 percentile operational data range, and setting the third factor (temperature, or DS) to their overall mean values. Thus, when the explanatory variables are set to their mean values, BY is equivalent to zero. This enabled a generic representation of the overall BY response to the main operational factors controlling the AD process.

3. Results

3.1. Overview of the conventional and THP MAD dataset and AD model

Overall average values of the main operational variables for conventional and THP MAD were calculated from monthly mean data and are summarised in Table 1, and specific mean data for individual conventional sites included in model development are shown in Figure 2. The overall mean BY for conventional treatment sites was approximately 400 m^3/t DS, which is typical for sewage sludge MAD (Bachmann *et al.* 2015). The overall mean values for conventional operational variables were: DS of raw feed sludge, 4.5%; VS of raw feed sludge, 76.1%; HRT, 21.2 days and temperature, approximately 36.0 ^oC. Volatile solids data is not collected routinely by all WWTP, and was therefore not included in the AD model development, but is presented in Table 1 as a parameter used widely in the literature to interpret AD performance. As would be expected (Barber, 2016), the overall mean BY, DS feed, VS feed, HRT and digestion temperature for THP MAD sites were all greater compared to the conventional process and were approximately equivalent to: 440 m^3/t DS, 7.9%, 78.9%, 22.4 days and 38.0 ^oC, respectively, albeit for a much smaller subset of 6 sites compared to the conventional process, which included 66 sites. Nevertheless, the site characteristics were consistent with the expected operational criteria and performance range of THP MAD (Barber, 2016).

The effects of temperature, HRT and DS feed on BY of conventional MAD were all highly statistically significant (P<0.001) and together with the categorical site factor, explained >50% of the total variation in BY data, which is extremely important to site operators (Table 2).

The natural logarithm model (Model 3) of the continuous predictors explained the largest overall proportion of total variation in BY (11.9%) and was selected for further analysis, as all the operational predictors in the model also had interpretable coefficients. This included the statistical analysis of interaction effects, which showed a significant (P=0.029) interaction between HRT and DS feed that was formulated into Model 4a (Table 2). The influence of the interaction between HRT and DS on the overall, relative response of the BY is shown in Figure S3 of the Supplementary Material.

3.2. Model validation

3.2.1. Conventional MAD sites

Model 4a was validated against specific, selected WWTP datasets. The results showed the patterns in digester performance were effectively captured, and demonstrated the important influence of the principal, controllable operating conditions (namely, digestion temperature, HRT and DS feed) on BY (Figure 3a), especially for Site 37 and 42. Note that the R^2 values presented in Figure 3a only provide a guide to the model description of operational data, since the site-specific, predicted BY (BY_p) values depend on the calibration frequency. Nevertheless, an R^2 value of 0.65 was obtained for Site 42, suggesting relatively good data agreement and recording at this site. A smaller R^2, such as that obtained for Site 38, indicates that other factors, not represented in the model, and/or poor data recording, have a stronger influence on the apparent BY than the main operational, controllable process variables, which could be a trigger for further site performance investigation.

Additional data were collected for conventional sites 1, 31 and 38, between 2016 – 2019, as part of the sludge composition study, and were also used for model validation as independent datasets, collected subsequent to the main data pool used to produce the model (Figure 3b). The results showed that the BY_p values generally followed the overall patterns in measured operational monthly average data for Site 38 (P<0.001, R^2= 0.59) and 31 (P<0.001, R^2= 0.57). However, the BY data supplied by Site 1 was significantly above the range considered representative of conventional AD: 300 – 440 m^3/t DS (CIWEM, 1996; Bachmann *et al.*, 2015). The default approach to internal site calibration using local BY data will thus track the site information, irrespective of whether this is representative of the actual process BY, as shown for Site 1 (P=0.003, R^2= 0.32) (Figure 3b).

3.3. Combined Conventional +THP-MAD model

To test whether THP MAD could be explained by the same operational parameters as conventional MAD, Model 4a was applied to the THP dataset to predict BY. The results showed the conventional MAD Model 4a gave a good overall description of BY at THP MAD sites with the default calibration method (P<0.001, R^2=0.72). The results demonstrated that the response of these processes to the major operational variables is fundamentally similar, although THP typically performs at a higher range. Therefore, THP and conventional data were pooled to generate a combined Conventional-THP model (Model 4b) following the procedure described in Section 3.1. Model 4a and 4b have equivalent significant predicting parameters and similar regression coefficients (Table 2; the general form of the equation is given in the Supplementary Material). Model 4b provided an effective description of the observed BY for THP (THP sites 1 – 6) and conventional (sites 37, 38 and 42) processes; P values were <0.001 in both cases, and R^2 was equivalent to 0.73 and 0.59, respectively (Figure S4). Consequently, Model 4b has practical application in monitoring the full-scale performance of both conventional and THP MAD.

3.4. Electricity yield

Electricity was generated by CHP plant from AD biogas at all sites and, accounting for other uses (for example, combustion in a boiler and flaring), is related to the amount of biogas produced. As would be expected, a strong linear relationship (R^2 = 0.90, P<0.001) was found between the electricity and biogas yields for the period 2016 to 2019 for selected sites (Conventional: 1, 31 and 38, and THP site 4; data not shown). However, Site 1 showed a very different gas-electricity profile compared to the other sites and both gas and electricity yields were significantly above the expected AD operational range (the mean BY of the data collected between May 2018 – Feb 2019 was >1000 m^3/t DS, whereas previously it was approximately 640 m^3/t DS, see Figure 2). Poor quality of gas data recording is a possible reason for the misalignment of gas and electricity data. However, a strong relationship between the biogas and electricity yields for Site 1 (R^2 = 0.90, P<0.001; data not shown) suggested reliable gas measurement in this case. Therefore, the large biogas (and electricity) yields observed here were attributed to factors other than gas measurement and we suspect that unreliable (underestimated) sludge feed volume recording was the most likely explanation. Site 1 was therefore removed from the correlation of biogas and electricity yield, nevertheless, the relationship remained highly statistically significant (R^2=0.60, P<0.001; data not shown).

The electricity modified site factor generated a similar BY compared to observed and default predicted values for the sites selected for detailed validation assessment (Figure 4). The detailed operational data used in the validation and site factor calculations are summarised in Table S1. Small differences between the observed BY and BY_e may be attributed to the quality of recording biogas use by CHP, and the efficiency of electricity conversion at specific sites. Thus, biogas-electricity generation conversion factors provide an effective alternative to the default biogas approach to site specific calibration of the model.

An adjusted form of the BY Model 4a was used to predict electricity yield (EY_p) (Model 4c in the Supplementary Material), by applying the standard biogas-to-electricity conversion factor (2.14; Banks, 2009), and validated for the selected sites (Figure S5) considered in Figure 3. An improved R^2 was observed for most of the selected sites compared to BY_p, and this suggested that electricity recording may be more reliable than biogas measurement at sites where the proportion of biogas used by CHP is known, and the electricity conversion efficiency is similar to the standard value. For example, a small increase in R^2, from 0.65 to 0.77, was observed for Site 42, where 94% of biogas was used by CHP and the average biogas-to-electricity conversion ratio was 1.8.

3.5. Sludge composition

The effects of temperature, HRT and DS feed on full-scale AD performance are represented by fixed coefficients in the multi-level regression model (Section 3.3). However, sites with similar operating conditions may have different BY performance, and this may be partly attributed to the variation in feed sludge composition (Weiland, 2010; Li *et al.*, 2018). The effect of sludge composition on the AD process was therefore investigated by measuring the major organic fractions (protein, fat, carbohydrate and fibre contents) in feed and digested sludge from the four selected MAD sites representing different levels of operational performance (Conventional: high, Site 1; moderate, Site 31; and low, Site 38; THP Site 4).

No significant relation (P>0.05) was detected between BY and sludge feed composition. However, a compositional BY_c value was derived for each sampling event based on the substrates destroyed during AD, and the theoretical CH_4 yield and content in the biogas (See Figure 1 and S6). A positive and highly statistically significant correlation (P = 0.007) was found between BY_c and the observed BY for the sites with reasonable biogas recording (Figure S7). Therefore, observed BY could be confidently described by the BY_c based on measurements of energy substrates in sludge pre and post digestion and the fractions destroyed. This provided a further approach to model calibration that was independent of the standard operational data recording of biogas and electricity. The results (Figure 4) showed that sludge composition generally underestimated the BY to some an extent compared to observed, predicted and electricity derived values. This could be explained because minor energy substrates may not be determined in the chemical analysis and/or that published gas yield values for one or more substrates may be slightly underestimated. However, the difference in BY_c compared to operationally observed or predicted values was particularly distinct for Site 1 (BY = 1362 m^3/t DS, BY_e = 1466 m^3/t DS and BY_c = 381 m^3/t DS). The BY_c value for the site was representative of typical AD performance and confirmed our suspicion of local problems with recording total sludge volumes fed to the digesters at the site. Therefore, where there are issues with the local measurement of absolute gas flow or feed volumes, for instance, quantifying the sludge composition is valuable to: (a) cross-reference the site data and, (b) independently calibrate the model.

3.6. Impacts of digestion conditions on process performance

The relative changes in BY described by Model 4b in response to DS feed and HRT, are shown in Figure 5a (omitting the site factor) for the average temperature recorded at conventional MAD sites, equivalent to 35.9 ^{o}C (Table 1). Biogas yield declined with increasing DS, but the magnitude of the response diminished with increasing HRT. The effect of HRT also depended on the DS concentration and BY increased to a greater extent with longer HRT as DS increased. For example, increasing HRT from 15 to 20 days raised the relative BY at 2.5% and 4.0 % DS by 28.1 m^3/t DS and 36.4 m^3/t DS, respectively. However, the magnitude of the response in BY to extending the HRT further from 20 to 25 days diminished at equivalent DS contents in the feed sludge to 21.8 to 28.3 m^3/t DS, respectively, due to the logarithmic relationship between HRT and BY. Thus, the model indicated the overall increases in BY possible with 10 days more HRT were equivalent to approximately 50 and 65 m^3/t DS for these DS contents in feed sludge, respectively. The relative changes in BY for THP MAD sites followed similar patterns to conventional treatment, but in this case, the operational conditions represented larger DS values and a wider range of HRT (see Table S2 in the Supplementary Material).

The effect of temperature and HRT on the relative changes in BY at the overall mean conventional DS in feed sludge of 4.5% DS (Table 1) is shown in Figure 5b. Biogas yield increased following a diminishing response to rising temperature. For example, an incremental rise of 3 ^{o}C from baseline temperatures of 33 ^{o}C and 36 ^{o}C increased BY by 23.1 and 21.2 m^3/t DS, respectively. THP MAD processes operated at higher temperature and DS feed, compared to conventional sites (Table 1). However, multi-level Model 4b showed that the effect of temperature on BY was independent of DS, therefore, equivalent relative increases in BY with temperature may be expected, irrespective of the differences in DS feed to these processes. Thus, a 3 ºC lift in temperature increased BY by 23.1 m^3/t DS relative to 33 ºC, at both 4.5 and 7.9% DS feed (the average DS values in feed sludge to conventional and THP digesters, respectively).

3.7. Net energy generation

Digester performance is controlled by the combined and interactive effects of the process parameters (Figure 5). Therefore, selecting a combination of appropriate and corresponding HRT, DS feed and digestion temperature conditions is necessary to maximise the BY, but this does not necessarily equate to the optimum performance of full-scale AD in terms of the overall maximum biogas volume and the net energy balance. The net energy calculations are summarised in the Supplementary Material. For example, according to Equation S1, if it is assumed that a 2000 m^3 digester is operated with a 20 day HRT, and daily feed volume of 100 m^3/day at 5% DS, and produces an optimum BY of 400 m^3/t DS and total biogas volume equivalent to 2000 m^3 biogas/d, the results show that increasing the daily feed volume by 33.3% reduces the HRT from 20 to 15 days and BY from 400 to 358 m^3/t DS, respectively. However, the total sludge feed is increased from 5.0 to 6.7 t DS/d. Therefore, although HRT and BY were reduced, the daily biogas flow increased by 19.4 % to 2387 m^3.

Table 3 shows the energy required to heat 1 t of wet sludge to increase the digestion temperature by 2 ºC, and the net energy generated from biogas produced from 1 t of sludge at DS concentrations in the feed equivalent to: 2.7%, 3.0%, 4.5% and 7.9%, respectively, according to Equation S2. A sludge DS of 2.7% was selected to illustrate the effects of a lower range value on the process energy balance, as this represented the lower 5% percentile of monthly average operational DS data for conventional MAD sites. Under these circumstances, increasing the digestion temperature to >39 ºC produced a negative net energy balance (the heating efficiency of the system was not considered). This is explained by the greater energy demand necessary to heat larger volumes of water and the reduced calorific output per wet t of sludge treated at low DS concentrations compared to larger DS contents. Therefore, it is recommended that the temperature of conventional MAD operated with low feed DS (<2.7%) is controlled in the low to medium mesophilic range (<39 ^{o}C) to achieve a positive energy balance. The multi-level model showed that a 2^{o}C lift in temperature can generate an additional 1.3 and 4.1 kWh of energy per t of wet sludge at the average DS concentrations of 4.5 and 7.9% found in sludge feed to conventional and THP MAD processes, respectively. Thus, the results demonstrate the potential advantage of increasing the digestion temperature of conventional MAD processes operated at typical DS feed concentrations, and also supported current operational strategies of adopting high mesophilic digestion temperatures at full-scale THP sites (Table 1), to gain the overall maximum quantitative energy benefit. Careful consideration is also necessary to balance the other main process conditions of sludge DS content and HRT when selecting the operating temperature for conventional MAD treatment.

4. Discussion

4.1. Multi-level modelling of full-scale AD processes

Modelling the performance of full-scale AD processes with a limited number of operational parameters is challenging due to the complexity of the system and the differences in data recording quality between sites. However, digestion temperature, DS of feed sludge and HRT are the main parameters available to operators to control and optimise sludge AD. Therefore, it would be a considerable advantage if a quantitative, modelling based decision support tool were available to adjust these conditions on an informed basis to guide and improve process operation. Here, we used up to 7 years of operational data recorded at 72 full-scale MAD sites to construct a widely applicable, operationally based, multi-level MAD biogas model to describe the changes in BY that are influenced by the major controllable AD parameters. The

BY was in the range of 150 to 700 m^3/t DS (Figure 2) and provided a comprehensive range of performance conditions to develop the multi-level, statistical model. Although, part of this variation may be attributed to poor measurements at some of the full-scale sites, the mean BY for conventional MAD was approximately 400 m^3/t DS, and was entirely consistent with typical values reported for sewage sludge MAD (Bachmann *et al.* 2015). Furthermore, the BY was highly statistically significantly correlated to EY ($P < 0.001$, $R^2 = 0.78$), and cross-validation provided confidence in the overall reliability of biogas recording.

Multi-level modelling is an appropriate method of analysis when raw data has a clustered structure; here, for example, data clustering was caused by inter-site differences that were not captured by the recorded operational parameters. Thus, the effects of operational parameters on the relative changes in BY were determined by accounting for the variance attributed to unknown differences between sites.

4.2. Factors affecting digestion performance

To a large extent, AD research has focussed on maximising biogas or CH_4 yield at laboratory scale by evaluating individual operational parameters (Nixon, 2016). For example, Dokulilová *et al*. (2018) showed BY increased with temperature in lab-scale digestion of sewage sludge at 20 days HRT, and suggested the maximum BY occurred in the upper mesophilic range (42 ^oC). In another example, Alepu *et al*. (2016) reported the CH_4 yield in lab-scale MAD of sludge increased with HRT and recommended 30 days HRT as the optimum. However, digester performance is determined by the combined and interactive effects of multiple process parameters and the multi-level model of full-scale MAD showed statistically significant positive effects of temperature and HRT, a negative effect of DS, with a positive, statistically significant interaction between HRT and DS.

The optimum performance of full-scale MAD processes requires the adoption of a suitable HRT depending upon the feed DS concentration (Figure 5a). This is necessary to ensure adequate contact time with the biodegradable substrates for metabolism as DS increases, and to prevent bacterial washout to maintain a sufficient population in the digester for the efficient conversion of complex organic matter to CH_4 and CO_2 (Parkin and Owen, 1986). Pretreatments for sludge are designed to increase the capacity of conventional MAD systems, and also provide other possible benefits, such as better sludge digestibility and pathogen reduction. Higher solids inputs and quicker reaction rates are reported (Wilson *et al*., 2008; Xue *et al*., 2015) for thermally hydrolysed sludge in the MAD process, compared to conventional digestion, and this is attributed to changes in sludge rheology and soluble organic contents after THP (Barber, 2016; Gurieff *et al.*, 2011; Xue *et al*. 2015). This behaviour was captured by Model 4b, by the statistically significant (P<0.001) main, positive effect of HRT on BY at full-scale conventional MAD sites, and the statistically significant (P=0.038) interaction between DS and HRT. Therefore, the magnitude of the response in BY to changes in the HRT was greater for THP MAD processes, due to the larger DS concentrations in THP feed sludge (4.2 – 10.5%), compared to conventional MAD, which operates with a lower range of DS feed contents (2.7 – 6.3%), over an equivalent range of HRT. Consequently, THP MAD may achieve a similar BY compared to conventional MAD, but with shorter HRT.

In general, the changes in BY observed at operational sites were effectively captured by the modelled parameters (Section 3.2). However, the extent of the variation explained may vary between different sites and time periods (Figure 3). This is because the model uses the three key, operationally monitored, parameters available at and used by all WWTP to control the AD process, as predictors of the total variation in BY, which, inevitably, also includes variability potentially attributable to other factors not directly considered in the model. These factors, amongst others, could include, for example, the sludge composition and primary:secondary sludge ratio, which determine the theoretical CH_4 yield and biodegradability of the feed sludge. Different substrates have varying CH_4 potentials, as shown in Figure 1b, and, under optimum digestion conditions, the typical, specific CH_4 yield for surplus activated sludge is 190 - 240 Nm^3/t VS, which is much smaller compared to primary sludge at 315 - 400 Nm^3/t VS, under equivalent conditions (Bachmann *et al.* 2015). This is because activated sludge is of biological origin and the microbial cells are embedded within the floc structure and extracellular polymeric substances, which provide a relatively resistant environment to lysis under AD conditions (Mottet *et al.*, 2010). Digester mixing is also an important factor defining AD performance, but is difficult to measure and quantify at full-scale, and models of the AD process generally assume a perfectly mixed system (Kythreotou *et al.*, 2014). The effect of mixing is linked strongly to the DS in the feed sludge and poorer mixing efficiency at high solids concentrations has been confirmed using tracer tests (Kamarád *et al.*, 2013). Therefore, our approach was to combine and capture all such 'known-unknown' variables within the categorical site factor of the multi-level MAD model.

Sludge composition is one such source of potential inter-site variability and routine analysis of the AD energy substrates is a feasible and practicable option to understand the impact on BY. Other major sources of variation are still to be identified and explored as a considerable amount of uncertainty in predicting AD performance and BY remains. For example, the microbial community involved in AD is dynamic and complex, however, it is well known that a stable population with the correct balance of the major groups responsible for the various stages of organic matter conversion by AD is essential (Narihiro *et al.*, 2015). This requires knowledge of the AD ecosystem to develop a fundamental understanding of how microbial community dynamics, interactions and functionality influence digester efficiency and stability (Vanwonterghem *et al.*, 2014). There have been several attempts to define the core AD microbiome, to identify the critical population responsible for the AD process, for example, Mei *et al.* (2017) examined 90 full-scale digesters from 5 countries and concluded that AD microbiomes were influenced by the operating conditions (for example, pretreatment, temperature range, and salinity) and could be classified into eight clusters on this basis. Therefore, greater fundamental insight to the AD process may be possible from a combination of engineering and meta-omics analysis.

4.3. Model validation

The R^2 of the validation results (Figure 3) depended on two factors: (1) the extent to which the coefficients of the principal operational parameter explained the BY, and (2) the frequency of model calibration to adjust the site factor (intercept) to determine the absolute BY value. The model was calibrated yearly in the model validation (Section 3.2), and therefore assumed that the influence of unmeasured factors was consistent for each annual period. The results showed that the model could capture and track the changing patterns in BY in time series data, and, for sites with good data quality recording, an R^2 value of 60% or better was achieved (Figure 3). The most reliable prediction of BY will be obtained for sites with characteristically stable operating conditions and where the site factor is consistent and captures the effects of other relevant management factors (for example, sludge composition, mixing efficiency, primary sludge ratio) that are not explained by the continuous variables in the model.

4.4. Model calibration

Three model calibration methods were applied using the local BY, electricity generation and sludge composition to calculate the specific site factor to represent the effects of unmeasured parameters on digestion performance. The default, internal calibration method using local, site BY data is generally recommended for routine model calibration, however, it relies on the availability of reliable gas flow measurements. Two additional calibration methods, using EY or sludge composition analysis, are also proposed, and can be useful when local data issues exist with the absolute measurement of gas flow or sludge feed volume, for instance.

Electricity yield is an effective method to cross-check historical and contemporary biogas data, and for model calibration, as electricity recording is reliable and routinely available at most

AD sites operating CHP. In addition, EY can provide an alternative to modelling BY as an indicator of AD performance where there is effective site recording of CHP biogas consumption. For example, in some cases, we found that the multi-level model of EY (which was based on the BY model, adjusted by a factor for electricity generation) had an improved R^2 value, compared to the corresponding BY prediction for the same site (Figure 3 and S5).

Measuring the destruction of the major groups of organic substrate in sludge by AD requires additional sample collection and analysis and is not routinely conducted at WWTP. The results presented here indicate the value of this approach to characterise the potential performance of sludge AD systems; it can provide an independent approach to industry recorded operational data for model calibration and can be used as a cross-reference, for instance, when there are multiple data recording issues at a particular site. For example, the BY_e was consistent with the observed and predicted BY results for Site 1, but both values were larger than would appear reasonable for conventional MAD of sewage sludge (Bachmann *et al.* 2015) based on the DS loading rates to the digesters calculated by the DS concentrations and feed volume reported by the operator for this site. However, a BY_c based on sludge composition analysis and the DS concentration at Site 1 was consistent with the typical performance of conventional MAD of sewage sludge. Therefore, unreliable recording of sludge feed volume to the AD process was the suspected cause of the unrepresentative biogas and electricity yield values obtained in this case.

4.5. Optimisation strategies

The multi-level BY model is based on currently monitored operational parameters and can provide decision support to increase the efficiency of sewage sludge AD. The model can be used to determine the appropriate temperature, HRT and DS control metrics to optimise the BY within the operating boundaries of full-scale sludge AD processes. However, only focussing on increasing the BY as the main objective of process control does not necessarily maximise the overall process energy balance. For example, reducing the HRT to potentially a sub-optimal BY can increase the total amount of sludge treated and the overall amount of biogas produced, thus improving the overall energy balance of the process, compared to digestion at higher BY (Table 3). Raising the digestion temperature also increases BY, but requires additional energy input. A positive energy benefit is possible by increasing the operating temperature in the high mesophilic range >39 ^{o}C, provided that a minimum DS of 3.0% is supplied in the feed sludge, and a greater energy surplus is obtained with increasing feed DS content.

5. Conclusions

A multi-level regression modelling technique was applied to large, sewage sludge AD, process monitoring data sets to quantify the significance of operational parameters controlling BY. The model effectively predicted the digester BY performance using basic operational parameters (temperature, HRT and DS in feed sludge) that are routinely recorded and used for process control at full-scale sewage sludge AD plant. The model shows that focussing on increasing the BY of MAD alone does not necessarily lead to an improvement in overall process performance in terms of energy balance. The importance of the integrated adjustment of DS feed, HRT and temperature is emphasised to optimise the overall energy balance of the AD process.

E-supplementary material of this work can be found in the online version of the paper.

6. Acknowledgement

This research is sponsored by Anglian Water Services Ltd, Severn Trent Plc, Thames Water Utilities Limited, United Utilities Group Plc, Yorkshire Water Services Ltd. The authors would like to thank the project steering group for their expert advice and comments on this project.

The views expressed in the paper are those of the authors and do not necessarily represent the companies supporting the research.

7. References

1. Alepu, O.E., Li, Z., Ikhumhen, H.O., Kalakodio, L. and Wang, K., 2016. Effect of hydraulic retention time on anaerobic digestion of Xiao Jiahe municipal sludge. International Journal of Waste Resources, 6(231), 231.

2. Astals, S., Esteban-Gutiérrez, M., Fernández-Arévalo, T., Aymerich, E., García-Heras, J. L. and Mata-Alvarez, J., 2013. Anaerobic digestion of seven different sewage sludges: a biodegradability and modelling study. Water Research, 47(16), 6033-6043.

3. Ainger, C., Butler, D., Caffor, I., Crawford-Brown, D., Helm, D. and Stephenson, T., 2009. A Low Carbon Water Industry in 2050. Report: SC070010/R3. Environment Agency, Bristol.

4. APHA, 2005. Standard Methods for the Examination of Water and Wastewater. Twenty-first edition, American Public Health Association, Washington.

5. Aiken, L.S., West, S.G. and Reno, R.R., 1991. Multiple regression: Testing and Interpreting Interactions. Sage Publications, Inc. London.

6. Barber, W.P.F., 2016. Thermal hydrolysis for sewage treatment: a critical review. Water Research, 104, 53-71.

7. Banks, C., 2009. Anaerobic Digestion and Energy. University of Southampton, School of Civil Engineering and the Environment. http://www.valorgas.soton.ac.uk/Pub_docs/JyU%20SS%202011/CB%204.pdf. [accessed: 03 May 2019]

8. Batstone, D.J., Keller, J., Angelidaki, I., Kalyuzhnyi, S.V., Pavlostathis, S.G., Rozzi, A., Sanders, W.T.M., Siegrist, H. and Vavilin, V.A., 2002. The IWA anaerobic digestion model no 1 (ADM1). Water Science and Technology, 45(10), 65-73.

9. Buswell, A. M.; Mueller, H. F., 1952. Mechanism of methane fermentation. Industrial and Engineering Chemistry, 44 (3), 550 – 552.

10. Bachmann, N., la Cour Jansen, J., Bochmann, G. and Montpart, N., 2015. Sustainable biogas production in municipal wastewater treatment plants. IEA Bioenergy, 20, 5-10.

11. Boušková, A., Dohanyos, M., Schmidt, J.E. and Angelidaki, I., 2005. Strategies for changing temperature from mesophilic to thermophilic conditions in anaerobic CSTR reactors treating sewage sludge. Water Research, 39(8), 1481-1488.

12. Chartered Institution of Water and Environmental Management (CIWEM), 1996. Sewage Sludge: Stabilisation and Disinfection. Handbooks of UK Wastewater Practice. CIWEM, London.

13. Donoso-Bravo, A., Mailier, J., Martin, C., Rodríguez, J., Aceves-Lara, C.A. and Wouwer, A.V., 2011. Model selection, identification and validation in anaerobic digestion: A review. Water Research, 45(17), 5347-5364.

14. Dumitru, C. and Maria, V., 2013. Advantages and disadvantages of using neural networks for predictions. Ovidius University Annals, Series Economic Sciences, 13(1), 444-449.

15. Dokulilová, T., Vítěz, T., Chovanec, J., Rouš, R., Vítězová, M. and Kushkevych, I., 2018. Primary and activated sludge biogas production: Effect of temperature. Acta Universitatis Agriculturae et Silviculturae Mendelianae Brunensis, 66(1), 23-28.

16. Eastman, J.A. and Ferguson, J.F., 1981. Solubilisation of particulate organic carbon during the acid phase of anaerobic digestion. Water Pollution Control Federation, 53(3), 352-366.

17. Frigge, M., Hoaglin, D.C. and Iglewicz, B., 1989. Some implementations of the boxplot. The American Statistician, 43(1), 50-54.

18. Güçlü, D., Yılmaz, N. and Ozkan-Yucel, U.G., 2011. Application of neural network prediction model to full-scale anaerobic sludge digestion. Journal of Chemical Technology & Biotechnology, 86(5), 691-698.

19. Gurieff, N., Bruus, J., Hoejsgaard, S., Boyd, J. and Kline, M., 2011. Maximizing energy efficiency and biogas production: Exelys™–continuous thermal hydrolysis. Proceedings of the Water Environment Federation, 2011(4), 1103-1116.

20. Gries, S. Th., 2015. The most under-used statistical method in corpus linguistics: Multi-level (and mixed-effects) models. Corpora, 10(1), 95-125.

21. Gelman, A. and Hill, J., 2006. Data Analysis Using Regression and Multilevel/hierarchical models. Cambridge University Press, Cambridge.

22. Hill, D.T., 1982. A comprehensive dynamic model for animal waste methanogenesis. Transactions of the ASAE, 25(5),1374-1380.

23. Howe, A., 2009. Renewable Energy Potential for the Water Industry. Report: SC070010/R5. Environment Agency, Bristol. http://www.environment-agency.gov.uk/research/library/publications/114393.aspx [accessed: 29 Jan 2014]

24. Harrison, X.A., Donaldson, L., Correa-Cano, M.E., Evans, J., Fisher, D.N., Goodwin, C.E., Robinson, B.S., Hodgson, D.J. and Inger, R., 2018. A brief introduction to mixed effects modelling and multi-model inference in ecology. Journal of Life and Environmental Sciences, 6, 4794.

25. Kythreotou, N., Florides, G. and Tassou, S.A., 2014. A review of simple to scientific models for anaerobic digestion. Renewable Energy, 71, 701-714.

26. Kamarád, L., Pohn, S., Bochmann, G. and Harasek, M., 2013. Determination of mixing quality in biogas plant digesters using tracer tests and computational fluid dynamics. Acta Universitatis Agriculturae et Silviculturae Mendelianae Brunensis, 61(5), 1269-1278.

27. Leech, N., Barrett, K. and Morgan, G.A., 2015. SPSS for Intermediate Statistics: Use and Interpretation. Fifth edition. Routledge, New York.

28. Li, W., Khalid, H., Zhu, Z., Zhang, R., Liu, G., Chen, C. and Thorin, E., 2018. Methane production through anaerobic digestion: Participation and digestion characteristics of cellulose, hemicellulose and lignin. Applied Energy, 226, 1219-1228.

29. Mei, R., Nobu, M.K., Narihiro, T., Kuroda, K., Sierra, J.M., Wu, Z., Ye, L., Lee, P.K., Lee, P.H., van Lier, J.B. and McInerney, M.J., 2017. Operation-driven heterogeneity and overlooked feed-associated populations in global anaerobic digester microbiome. Water Research, 124, 77-84.

30. Mottet, A., François, E., Latrille, E., Steyer, J. P., Déléris, S., Vedrenne, F. and Carrère, H., 2010. Estimating anaerobic biodegradability indicators for waste activated sludge. Chemical Engineering Journal, 160(2), 488-496.

31. Mariotti, F., Tomé, D. and Mirand, P.P., 2008. Converting nitrogen into protein—beyond 6.25 and Jones' factors. Critical Reviews in Food Science and Nutrition, 48(2), 177-184.

32. Narihiro, T., Nobu, M.K., Kim, N.K., Kamagata, Y. and Liu, W.T., 2015. The nexus of syntrophy-associated microbiota in anaerobic digestion revealed by long-term enrichment and community survey. Environmental Microbiology, 17(5), 1707-1720.

33. Nixon, J.D., 2016. Designing and optimising anaerobic digestion systems: A multi-objective non-linear goal programming approach. Energy, 114, 814-822.

34. Nielsen, M., Holst-Fischer, C., Malmgren-Hansen, B., Bjerg-Nielsen, M., Kragelund, C., Møller, H.B. and Ottosen, L.D.M., 2017. Small temperature differences can improve the performance of mesophilic sludge-based digesters. Biotechnology Letters, 39(11), 1689-1698.

35. Otuzalti, M.M. and Perendeci, N.A., 2018. Modeling of real scale waste activated sludge anaerobic digestion process by Anaerobic Digestion Model 1 (ADM1). International Journal of Green Energy, 15(7), 454-464.

36. Ozgun, H., 2019. Anaerobic Digestion Model No. 1 (ADM1) for mathematical modeling of full-scale sludge digester performance in a municipal wastewater treatment plant. Biodegradation, 30(1), 27-36.

37. Parkin, G.F. and Owen, W.F., 1986. Fundamentals of anaerobic digestion of wastewater sludges. Journal of Environmental Engineering, 112(5), 867-920.

38. POST; Parliamentary Office of Science and Technology, 2007. Energy and Sewage. Number 282. http://www.parliament.uk/business/publications/research/briefing-papers/POST-PN-282/energy-and-sewage-april-2007 [accessed: 29 Jan 2014]

39. Qdais, H.A., Hani, K.B. and Shatnawi, N., 2010. Modeling and optimization of biogas production from a waste digester using artificial neural network and genetic algorithm. Resources, Conservation and Recycling, 54(6), 359-363.

40. Thames Water Utilities Limited, 2019. Annual Report and Annual Performance Report 2018/2019.https://corporate.thameswater.co.uk/-/media/Site-Content/Thames-Water/Corporate/AboutUs/Investors/Annual-report/2019/2018-19-Annual-Report-and-Annual-Performance-Report.pdf [accessed: 02 Sept 2019]

41. USEPA., 1983. Methods for Chemical Analysis of Water and Wastes. EPA-600/4–79-020. US Environmental Protection Agency, Environmental Monitoring and Support Laboratory. Cincinnati.

42. Vanwonterghem, I., Jensen, P.D., Ho, D.P., Batstone, D.J. and Tyson, G.W., 2014. Linking microbial community structure, interactions and function in anaerobic digesters using new molecular techniques. Current Opinion in Biotechnology, 27, 55-64.

43. Van Soest, P.V., Robertson, J.B. and Lewis, B.A., 1991. Methods for dietary fiber, neutral detergent fiber, and nonstarch polysaccharides in relation to animal nutrition. Journal of Dairy Science, 74, 3583-3597.

44. Wilson, C.A., Murthy, S.M., Fang, Y. and Novak, J.T., 2008. The effect of temperature on the performance and stability of thermophilic anaerobic digestion. Water Science and Technology, 57(2), 297-304.

45. Ward, A. J., Hobbs, P. J., Holliman, P. J., & Jones, D. L., 2008. Optimisation of the anaerobic digestion of agricultural resources. Bioresource Technology, 99, 7928-7940.

46. Water UK, 2009. How the Water Industry is Managing its Contribution to Climate Change. http://www.water.org.uk/home/policy/climate- change/mitigation [accessed: 25 Feb 2014]

47. Weiland, P., 2010. Biogas production: current state and perspectives. Applied Microbiology and Biotechnology, 85(4), 849–860.

48. Xue, Y., Liu, H., Chen, S., Dichtl, N., Dai, X. and Li, N., 2015. Effects of thermal hydrolysis on organic matter solubilization and anaerobic digestion of high solid sludge. Chemical Engineering Journal, 264,174-180.

Table 1 Summary of overall mean values of principal parameters used for conventional and THP MAD process control

Parameter		Anaerobic digestion type	
		Conventional	THP
DS feed (%)	Mean	4.5	7.9
	SD	1.1	2.1
VS feed (%)	Mean	76.1	78.9
	SD	4.0	6.3
HRT (days)	Mean	21.2	22.4
	SD	6.9	10.0
Temperature (^{o}C)	Mean	35.9	38.2
	SD	2.2	3.7
BY (m^3/t DS)	Mean	398.7	438.6
	SD	176.3	148.0

BY- biogas yield; DS - dry solids; VS - volatile solids; HRT - hydraulic retention time; SD - standard deviation.

Note that VS is not collected at all sites and therefore was not included in model development.

Table 2 Multi-level biogas models of full-scale anaerobic digestion of sewage sludge

Model	R^2	Significant predictors (*P* value)	Predictor coefficient	Total variation explained by predictors
1. Linear	0.51	HRT_c ($P < 0.001$)	5.54	10.7%
		Temperature_c ($P < 0.001$)	6.80	
		DS input_c ($P < 0.001$)	-47.6	
		Site factor ($P < 0.001$)		
2. Quadratic	0.53	HRT_c ($P < 0.001$)	7.17	11.8%
		Temperature_c ($P < 0.001$)	6.62	
		DS input_c ($P < 0.001$)	-53.2	
		$(HRT\_c)^2$ ($P < 0.001$)	-0.20	
		$(DS\ input\_c)^2$ ($P < 0.001$)	7.46	
		Site factor ($P < 0.001$)		
3. Natural Log	0.53	Ln(HRT)_c ($P < 0.001$)	137.1	11.9%
		Ln(Temperature)_c ($P < 0.001$)	232.5	
		Ln(DS input)_c ($P < 0.001$)	-221.6	
		Site factor ($P < 0.001$)		
4a. Interaction	0.53	Ln(HRT)_c ($P < 0.001$)	136.2	**12.1%**
		Ln(Temperature)_c ($P < 0.001$)	231.0	
		Ln(DS input)_c ($P < 0.001$)	-224.8	
		Ln(HRT)c* Ln(DS input)_c (0.029)	75.5	
		Site factor ($P < 0.001$)		
4b. Interaction	0.52	Ln(HRT)_c ($P < 0.001$)	133.7	**8.5%**
		Ln(Temperature)_c ($P < 0.001$)	265.3	
		Ln(DS input)_c ($P < 0.001$)	-216.4	
		Ln(HRT)c* Ln(DS input)_c (0.037)	61.7	
		Site factor ($P < 0.001$)		

HRT_c: hydraulic retention time centred with mean; Temperature_c: digestion temperature centred with mean; DS input_c: digester feed dry solids concentration centred with mean; *Site factor*: is a categorical variable for site. Models: 1, 2, 3, and 4a are conventional MAD and 4b is combined conventional + THP MAD.

Table 3 Energy balance of conventional and THP MAD with increasing digestion temperature under different dry solids (DS) feed concentration regimes

Temperature increase (°C)	31 to 33	33 to 35	35 to 37	37 to 39	39 to 41
Energy required to heat 1 t wet sludge(kWh)	2.3	2.3	2.3	2.3	2.3
Net energy out from 1 t wet sludge in kWh (2.7% DS)	**0.4**	**0.3**	**0.1**	**0.0**	*-0.1*
Net energy out from 1 t wet sludge in kWh (3.0% DS)	**0.7**	**0.6**	**0.4**	**0.3**	*0.1*
Net energy out from 1 t wet sludge in kWh (4.5% DS)	**2.3**	**2.0**	**1.8**	**1.5**	1.3
Net energy out from 1 t wet sludge in kWh (7.9% DS)	*5.7*	5.2	4.8	4.4	4.1

Note that bold and underlined values reflect conditions that may overlap in operational practice for conventional and THP MAD; The italic values reflect extreme cases that fall outside the 90% operational data range (i.e. <5 and >95 percentile). 2.7% DS is the lower 5% percentile range value of monthly average operational data for conventional MAD sites; 3.0% is the break point sludge feed DS for a positive net energy balance for MAD; 4.5% and 7.9% DS are the mean values of monthly average operational data for conventional and THP MAD sites, respectively.

Figure Captions

Figure 1 The overall approach to data collection, model development, validation, calibration and optimisation. Operational parameter data available at all sites and used in conventional MAD model development are marked in blue. Theoretical CH_4 yield and CH_4 contents of biogas produced from different specific organic substrates were from Weiland (2010) and Li *et al.* (2018); Electricity conversion factor by combined heat and power (CHP) was from Banks (2009). (solid line: conventional MAD model development pathway; dashed line: conventional + THP-MAD model development pathway). DS – dry solids; HRT – hydraulic retention time; VS – volatile solids; VFA – volatile fatty acids; BY – biogas yield; BY_c – composition derived biogas yield; BY_e – electricity derived biogas yield

Figure 2 Mean biogas yield (BY) , temperature, hydraulic retention time (HRT) and dry solids (DS) feed for individual conventional MAD sites, the period of data collection for each Company is also shown

Figure 3 Conventional MAD model validation showing the observed and predicted monthly average biogas yield (Model 4a) for: (a) Site 37, 38 and 42 for the data involved in model generation and (b) Site 31, 38 and 1 for independent datasets

Figure 4 Observed and predicted (Model 4b) average biogas yield (BY) for conventional Site 1, 31, 38 and THP site 4, based on default (local biogas data), electricity generation and sludge composition calibration methods and data collected between May 2018 to February 2019 (error bar: standard deviation); model input data are given in Table S1

Figure 5 Relative changes in biogas yield in relation to: (a) hydraulic retention time (HRT) and dry solids (DS) feed at the overall mean conventional digestion temperature of 35.9 ^oC, and (b) HRT and temperature at the overall mean DS feed concentration for

conventional digestion of 4.5 %. (Temperature, HRT and DS data ranges limited to the 5 and 95 percentiles of monthly average operational values)

Supplementary Material to "A Multi-level Biogas Model to Optimise the Energy Balance of Full-Scale Sewage Sludge Conventional and THP Anaerobic Digestion"

Jin Liu and Stephen R. Smith

Department of Civil and Environmental Engineering, Imperial College London, South Kensington Campus, London SW7 2AZ, UK

Corresponding Author Email s.r.smith@imperial.ac.uk

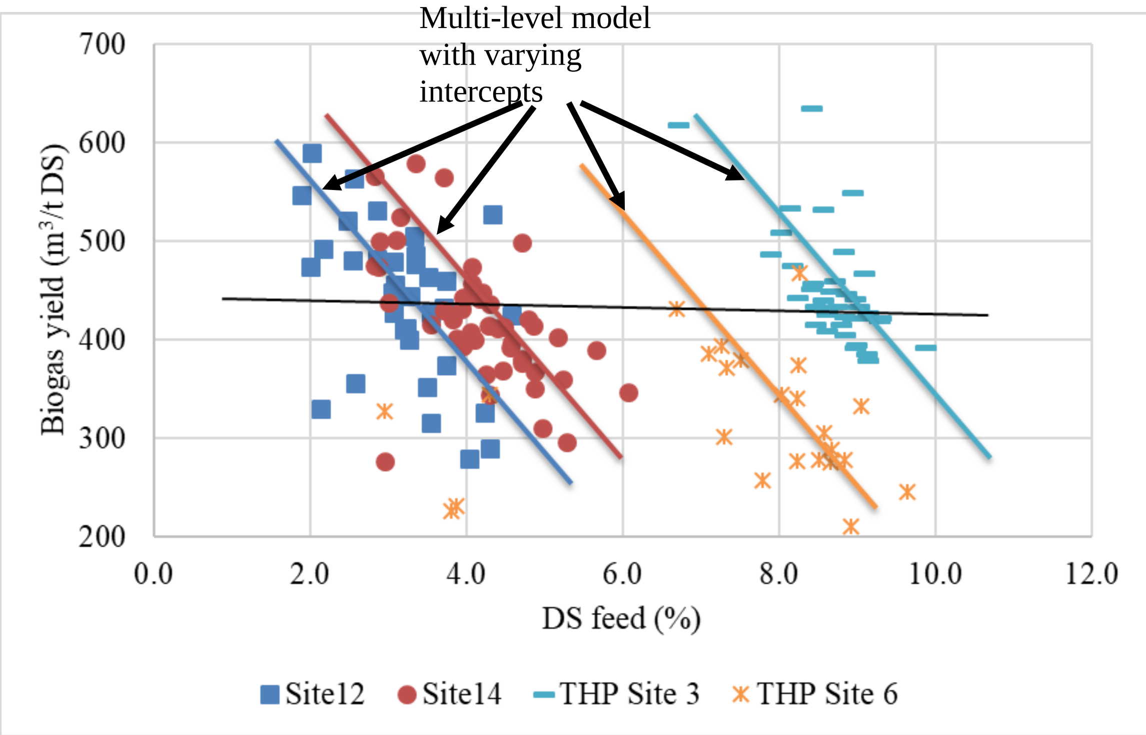

Figure S1 Clustered data structure of biogas yield relative to dry solids (DS) feed content in feed sludge for full-scale AD sites (Site 12 and 14 are conventional MAD and THP Site 3 and 6 include thermal hydrolysis pretreatment), showing examples of data modelling by standard linear regression wi~~th fi~~xed intercept ( ) and multi-level regression with varying intercepts

Description of the general model structures:

Random intercept, multi-level models were constructed by examining the operational parameters dry solids (DS) feed, temperature and hydraulic retention time (HRT)) in the following sequence: (1) linear regression, (2) curvilinear and non-linear regression (quadratic/cubic and log transformed), and (3) testing the interactive effects of the significant predictors. The predictors were centred before fitting into the model, by subtracting the mean value from each variable. The equations had the following general forms:

Linear:

$$BY_{ij} = \alpha_j + \beta_1 \mathrm{HRT}_{\mathrm{centred\ ij}} + \beta_2 \mathrm{Temperature}_{\mathrm{centred\ ij}} + \beta_3 \mathrm{DS\ input}_{\mathrm{centred\ ij}} + e_{ij}$$

Quadratic and cubic:

$$\begin{aligned} BY_{ij} = \alpha_j &+ (\gamma_1 \mathrm{HRT}_{\mathrm{centred\ ij}} + \gamma_2 \mathrm{HRT}^2_{\mathrm{centred\ ij}} [+ \gamma_3 \mathrm{HRT}^3_{\mathrm{centred\ ij}}]) \\ &+ (\gamma_4 \mathrm{Temperature}_{\mathrm{centred\ ij}} \\ &+ \gamma_5 \mathrm{Temperature}^2_{\mathrm{centred\ ij}} [+ \gamma_6 \mathrm{Temperature}^3_{\mathrm{centred\ ij}}]) \\ &+ (\gamma_7 \mathrm{DS\ input}_{\mathrm{centred\ ij}} + \gamma_8 \mathrm{DS\ input}^2_{\mathrm{centred\ ij}} [+ \gamma_9 \mathrm{DS\ input}^3_{\mathrm{centred\ ij}}]) + e_{ij} \end{aligned}$$

Log transformed:

$$\begin{aligned} BY_{ij} = \alpha_j &+ \delta_1 \mathrm{Ln(HRT)}_{\mathrm{centred\ ij}} \\ &+ \delta_2 \mathrm{Ln(Temperature)}_{\mathrm{centred\ ij}} + \delta_3 \mathrm{Ln(DS\ input)}_{\mathrm{centred\ ij}} + e_{ij} \end{aligned}$$

Where BY_{ij} is the biogas yield (BY) for observation *i* in Site *j*, α_j is the Level-2 random effect (site specific factor), β_n are the coefficients of operational parameters for the linear equation, γ_n are the coefficients for the quadratic/cubic operational parameters (cubic terms are represented in square brackets), δ_n are the coefficients for log transformed operational parameters and e_{ij} is the Level-1 random effect. For the quadratic and cubic models, the cubic terms are shown in square brackets. Backward elimination and forward selection methods were applied to identify statistically significant predictors from the total number of independent predictors available. Following this stage, the log-transformed model (Model 3, see Table 2) gave marginally the largest overall R^2 value and was of a simpler form than the quadratic relation (Model 2, Table 2) and was selected for testing interaction terms (using Ln transformed data): DS input*temperature, DS input*HRT, temperature*HRT and DS input*temperature*HRT; the interaction term: HRT*DS input was statistically significant (P<0.05) and included in the final Model 4a.

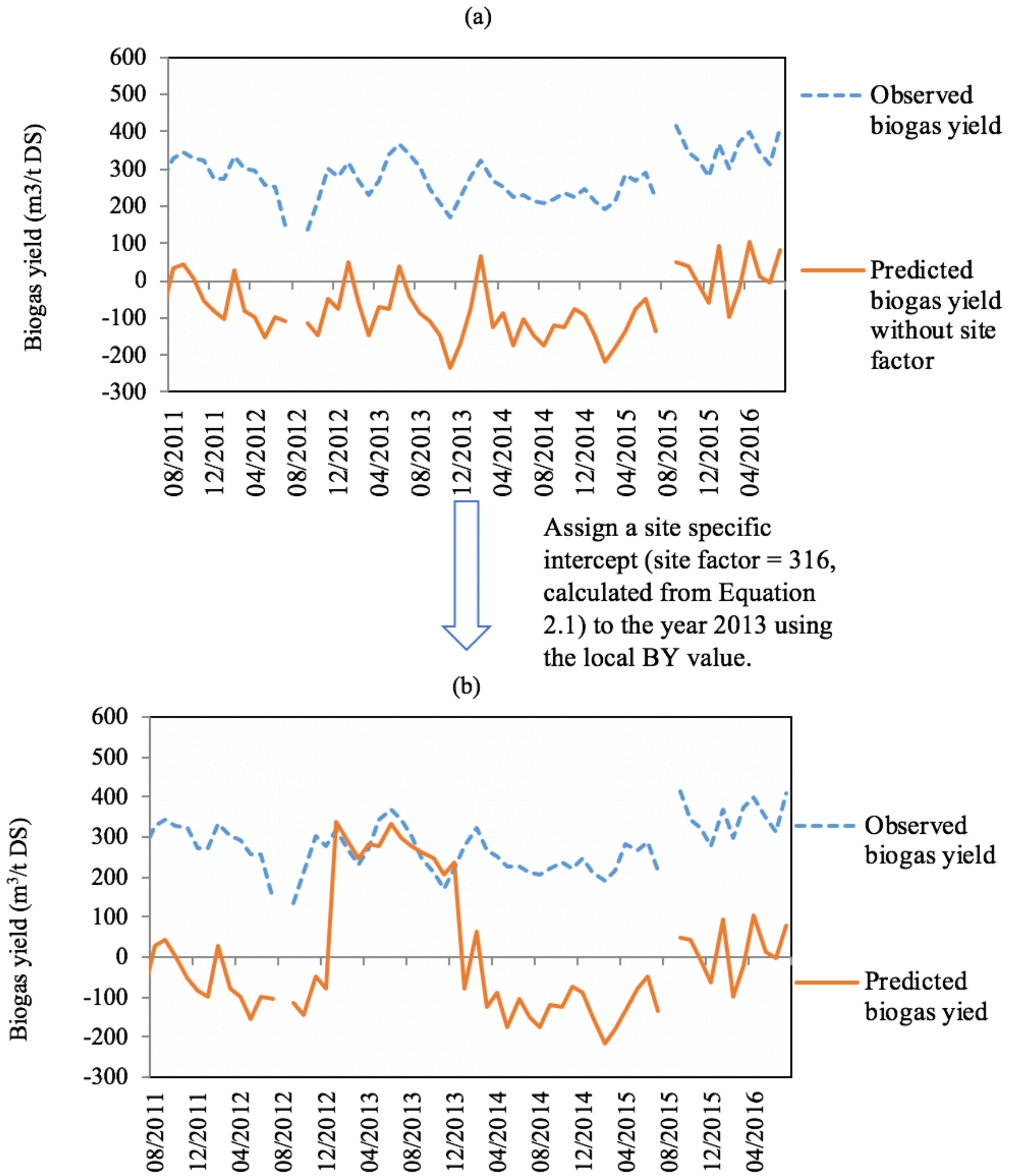

Figure S2 Example of the model calibration showing the observed and predicted (Model 4a) biogas yield for Site 42: (a) with predictor variables centred by subtracting the mean value from each variable and (b) with the specific intercept (site factor) determined from the local biogas data for year 2013

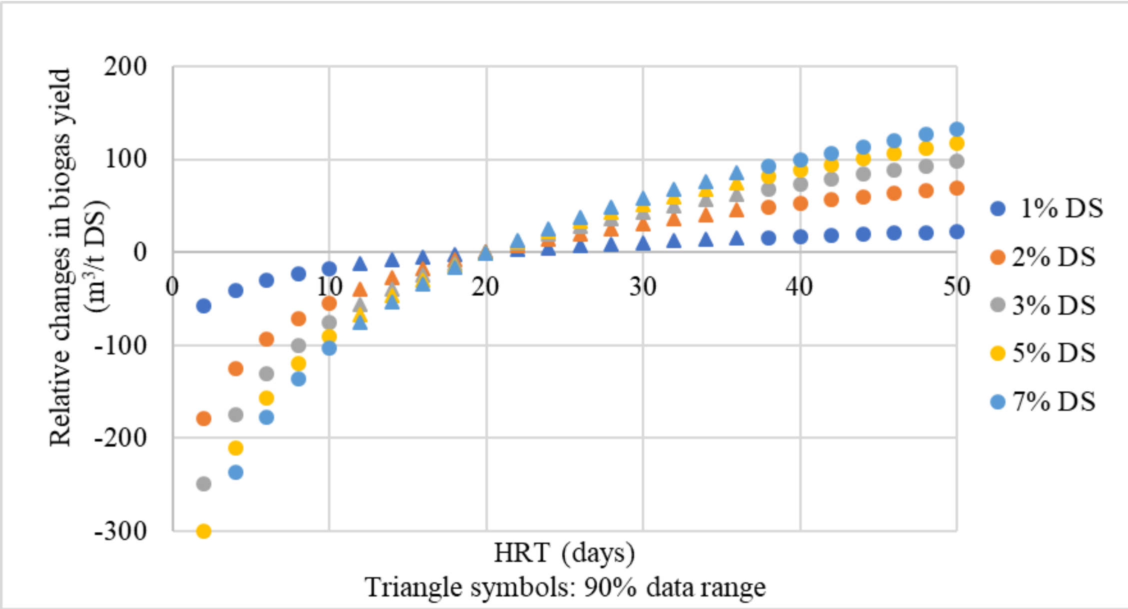

Figure S3 Effect of hydraulic retention time (HRT) on the relative change in biogas yield at different dry solids (DS) feed concentrations for conventional MAD model (Model 4a)

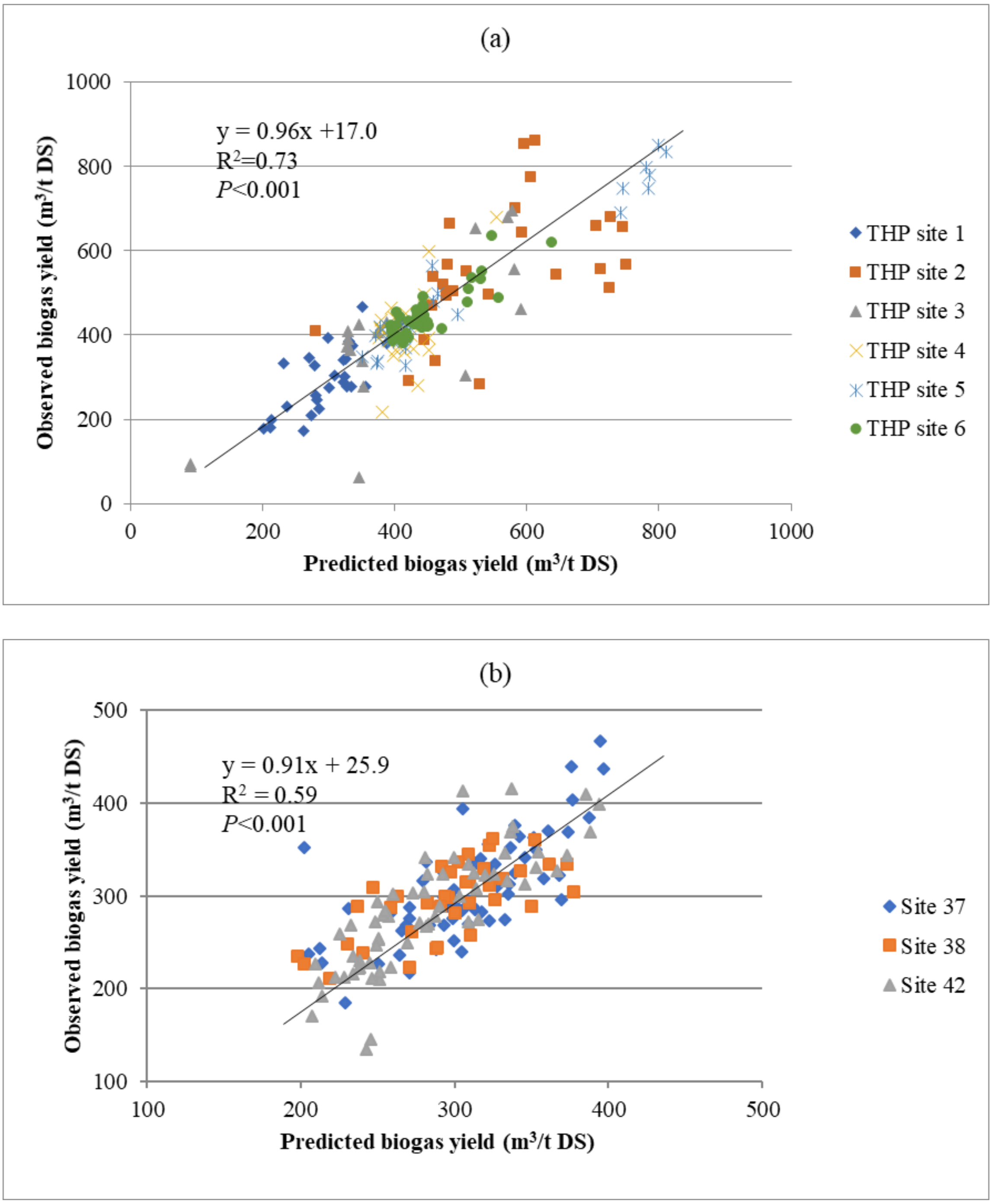

Figure S4 Observed biogas yield relative to values predicted by Model 4b: (a) all THP MAD sites and (b) selected conventional MAD sites

Table S1 Average sludge dry solids (DS) feed, hydraulic retention time (HRT), temperature, observed, electricity and sludge composition derived biogas yield (BY, BY_e and BY_c, respectively), and the estimated site factors used to predict biogas yield using Model 4b and shown in Figure 4 based on default biogas, electricity generation and sludge composition calibration methods (Equation 2.1)

Site	DS feed (%)	HRT (days)	Temperature (ºC)	Observed BY (m^3/t DS)	BY_e (m^3/t DS)	BY_c (m^3/t DS)	Default site factor	Electricity modified site factor	Sludge composition site factor
Site 1	3.7	27.9	36.9	1361.7	1461.2	381.3	1273.4	1378.5	293.0
Site 31	4.1	24.5	37.0	448.4	474.6	412.3	395.3	421.5	359.2
Site 38	6.2	29.0	41.1	379.0	356.4	299.0	355.6	333.0	275.6
THP site 4	9.7	19.4	42.6	491.1	512.5	372.0	619.1	640.5	500.0

Site factors are calculated by substituting the default observed average, electricity or sludge composition derived BY into Equation 2.1 as follows:

$$\text{Default site factor} = \text{Average observed BY} - [\textstyle\sum_{i=1}^{10}(\text{BY predicted by fixed coefficients})_i]/10$$

$$\text{Electricity modified site factor} = \text{Average electricity derived BYe} - [\sum_{i=1}^{10}(\text{BY predicted by fixed coefficients})_i]/10$$

$$\text{Sludge composition site factor} = \text{Average composition BYc} - [\textstyle\sum_{i=1}^{10}(\text{BY predicted by fixed coefficients})_i]/10$$

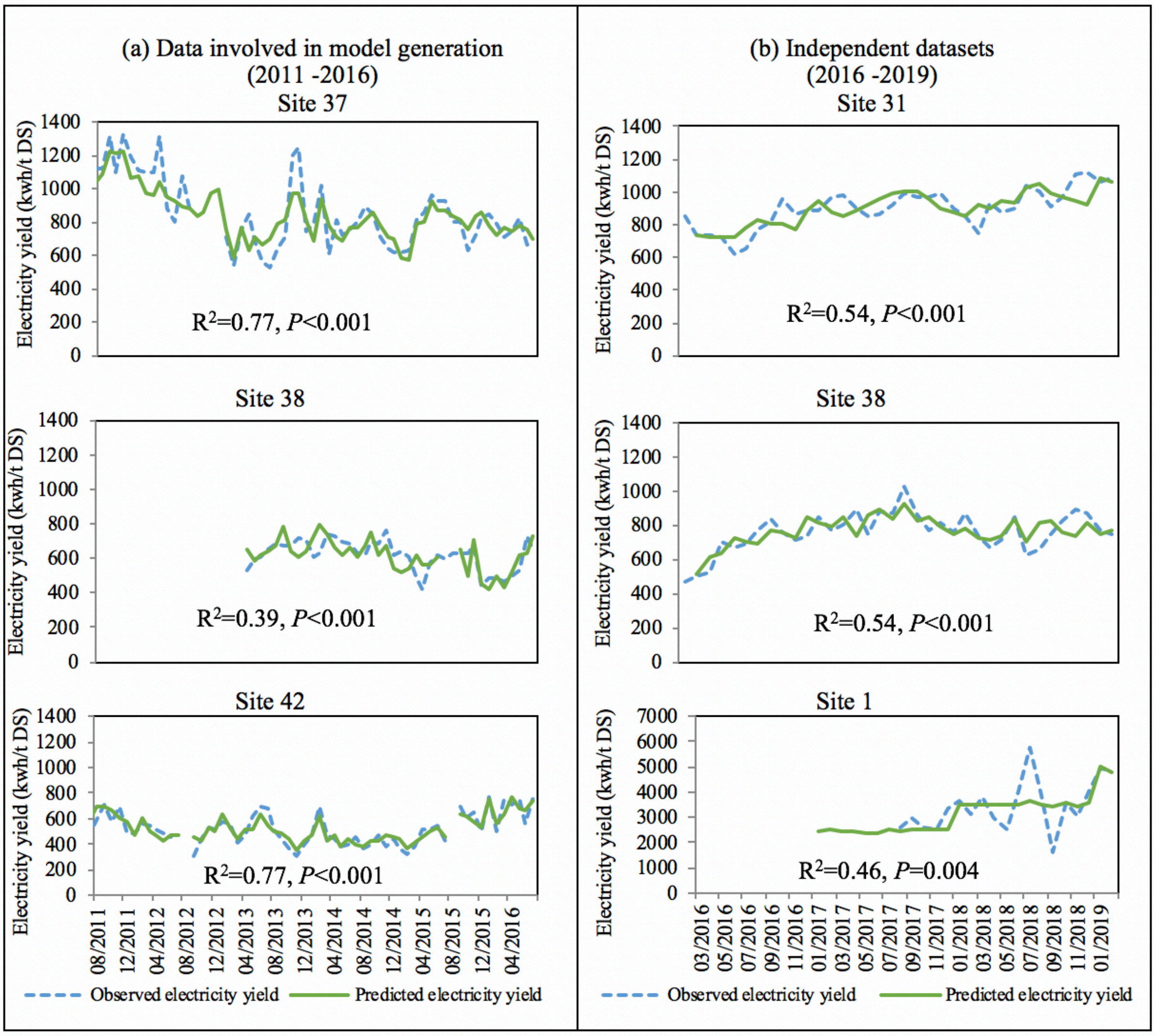

Figure S5 Conventional MAD model validation showing the observed and predicted monthly average electricity yield (Model 4c) for: (a) Site 37, 38 and 42 for the data involved in model generation and (b) Site 31, 38 and 1 for independent datasets

(Model 4c: Electricity yield (kwh/t DS) = 2.14*[230.9*(Ln(Temperature) - 3.6) + 136.2*(Ln(HRT) - 3.0)-224.8*(Ln(DS) - 1.5) + 75.5*((Ln(HRT) – 3.0) *(Ln(DS) - 1.5))] + site factor (calibrated using local electricity yield data))

Sludge composition:

The proportions of each substrate in the feed sludge and the destroyed fractions during AD on a volatile solids (VS) basis were calculated and are summarised in Figure S6. A sludge composition BY_c was derived for each sampling site based on the substrates destroyed during AD, and the theoretical CH_4 yield and content in the biogas (Figure 1). A positive and highly statistically significant correlation (P = 0.005) was found between BY_c and the observed BY for the sites with reasonable biogas recording (Figure S7). Therefore, observed BY could be described by BY_c with a reasonable degree of confidence based on the composition of energy substrates measured in sludge.

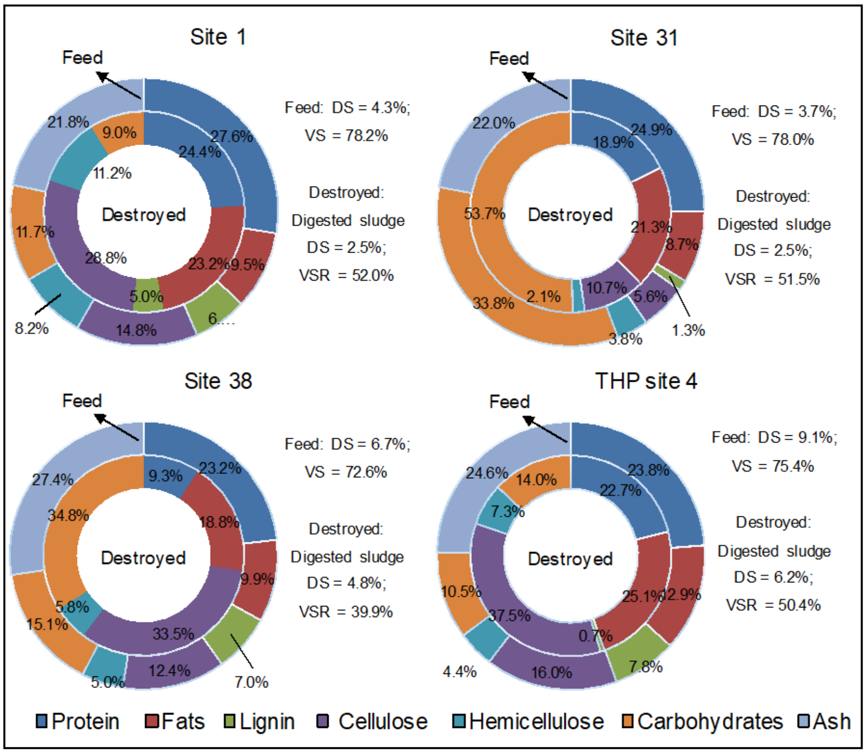

Figure S6 Composition of organic and ash fractions in sludge feed (% dry solids basis) and corresponding destroyed fractions (% volatile solids basis) by anaerobic digestion for conventional Site 1, 31, 38 and THP site 4 (outer circle: sludge feed; inner circle: fraction destroyed by digestion); VSR, volatile solids reduction; DS, dry solids; VS, volatile solids

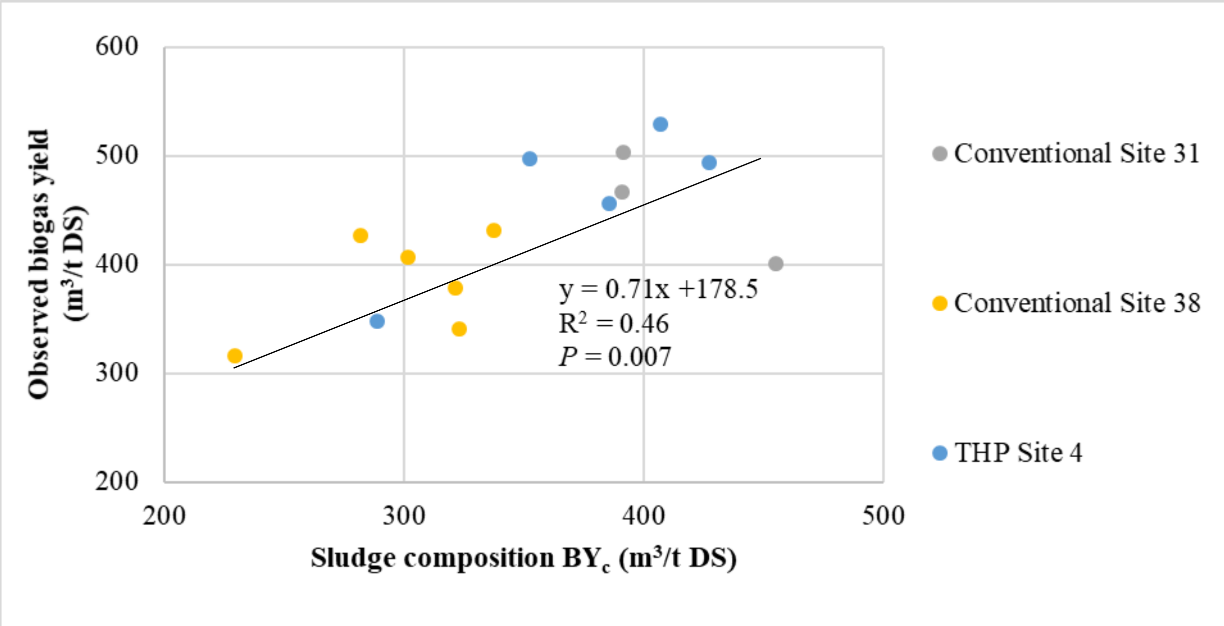

Figure S7 Relationship between observed biogas yield (BY) and sludge composition BY_c for selected conventional and THP sites

Table S2 Relative changes in biogas yield (BY) for different combinations of hydraulic retention time (HRT) and dry solids (DS) feed concentrations, reflecting conventional and

THP MAD operating ranges (conventional range: bold values; THP range: underlined values)

HRT changes \ DS feed	2.5%	4.0%	5.0%	6.5%	8.0%	10.0%
	Relative changes in BY (m^3/t DS)					
10 to15 days	**39.6**	**51.4**	**57.0**	**63.5**	68.7	74.3
15 to 20 days	**28.1**	**36.4**	**40.4**	**45.1**	48.8	52.7
20 to 25 days	**21.8**	**28.3**	**31.3**	**35.0**	37.8	40.9
25 to 30 days	**17.8**	**23.1**	**25.6**	**28.6**	30.9	33.4
30 to 35 days	NA	19.5	21.7	24.1	26.1	28.2
35 to 40 days	NA	16.9	18.8	20.9	22.6	24.5

Note that bold and underlined values reflect conditions that may overlap in operational practice for conventional and THP MAD; NA, not applicable

Net energy calculation

Biogas yield is a widely used performance indicator to evaluate the anaerobic digestion (AD) process and research on AD optimisation has tended to focus on manipulating individual operational parameters to maximise BY or CH_4 yield (Alepu *et al*. 2016; Dokulilová *et al*. 2018). However, a major quantitative benefit of the full-scale AD model is to understand the simultaneous effects of all the main process control factors on the overall net energy output. For example, a key strategic focus of AD operation is optimising the balance between the rate of sludge treatment and the energy output, primarily by manipulating the feed DS and/or HRT. Thus, extending HRT may increase the BY, but reduces the total sludge throughput at equivalent DS feed. The effect of HRT on the net biogas output can be calculated using Equation S1:

$$\text{Net daily biogas gas } (\mathrm{m^3/day}) = \text{Biogas volume}_1 - \text{Biogas volume}_2$$

$$= \frac{\mathrm{DS}}{100} \times \left[\frac{\text{Digester volume}}{\mathrm{HRT_1}} \times \mathrm{BY} - \frac{\text{Digester volume}}{\mathrm{HRT_2}} \times (\mathrm{BY} + x)\right]$$

$$= \frac{\mathrm{DS}}{100} \times \text{Digester volume } \times \left[\frac{\mathrm{BY}\times\mathrm{HRT_2} - \mathrm{HRT_1}\times(\mathrm{BY}+x)}{\mathrm{HRT_1}\times\mathrm{HRT_2}}\right] \qquad \text{(Equation S1)}$$

Where:

Biogas volume$_1$ (m^3/day) = the volume of biogas produced when HRT is equal to HRT_1

Biogas volume$_1$ (m^3/day) = the volume of biogas produced when HRT is equal to HRT_2

BY (m^3/t DS) = the biogas yield when HRT is equal to HRT_1

x (m^3/t DS) = the relative change of biogas yield when HRT is changed from HRT_1 to HRT_2

Increasing the digestion temperature may be considered as an option to raise the BY (Dokulilová *et al*., 2018), but requires additional energy input; thus, increasing the temperature of 1 t of wet sludge by 1^{o}C requires 4.18 kJ/kg = 1.16 kWh energy (assuming sludge has the same specific heat capacity as water), and the energy in 1 m^{3} biogas (60% CH_4 content) is equivalent to 22 MJ = 6.1 kWh (Banks, 2009). Therefore, the net energy required to increase the digestion temperature of 1 t of wet sludge may be calculated using Equation S2:

Net energy (kWh) = Additional energy generated – Additional energy required for heating = $6.1 * DS * z - 1.16 * (Temperature_1 - Temperature_2)$ (Equation S2)

Where:

z (m^{3}/t DS) = the relative change of BY when the temperature is adjusted from $Temperature_1$ to $Temperature_2$

Supplementary material references

1. Alepu, O.E., Li, Z., Ikhumhen, H.O., Kalakodio, L. and Wang, K., 2016. Effect of hydraulic retention time on anaerobic digestion of Xiao Jiahe municipal sludge. International Journal of Waste Resources, 6(231), 231.

2. Banks, C., 2009. Anaerobic Digestion and Energy. University of Southampton, School of Civil Engineering and the Environment. http://www.valorgas.soton.ac.uk/Pub_docs/JyU%20SS%202011/CB%204.pdf. [accessed: 03 May 2019]

3. Dokulilová, T., Vítěz, T., Chovanec, J., Rouš, R., Vítězová, M. and Kushkevych, I., 2018. Primary and activated sludge biogas production: Effect of temperature. Acta Universitatis Agriculturae et Silviculturae Mendelianae Brunensis, 66(1), 23-28.