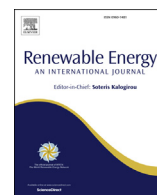




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# Hydrothermal carbonization of sewage digestate at wastewater treatment works: Influence of solid loading on characteristics of hydrochar, process water and plant energetics

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## ABSTRACT

Nowadays the sludge treatment is recognized as a priority challenge to the wastewater industry due to the increasing volumes produced and tighter environmental controls for its safe disposal. The most cost-effective process for sewage sludge is the anaerobic digestion but raw digestate still contains high levels of organic matter that can be transformed into an energy carrier by using processes like Hydrothermal Carbonization (HTC). In this work, the influence of solid loading (2.5, 5.0, 10.0, 15.0, 17.5, 20.0, 25.0 and 30.0% solids w/w) on the composition of hydrochar and process water was studied, together with an evaluation of product yields, solubilisation of organic carbon and biomethane potential of process waters from HTC processing (250 °C, 30- minute reaction time). Hydrochar yields ranged from 64 to 88%wt, whereas the concentration of soluble organic carbon increased from 2.6 g/L in the raw digestate to a maximum of 72.3 g/L in the process water following HTC at the highest solid loading. Furthermore, process modelling with Aspen Plus shows that the integration of AD with HTC to wastewater treatment works provides a significant positive energy balance when process water and hydrochar are considered as fuel sources for cogeneration.

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## 1. Introduction

Sewage sludge (SS) is produced as part of routine operations at wastewater treatment works (WWTWs) and its management is still an important global issue due to the large amounts generated on a daily basis [1]. In the UK, 1.4 million tonnes of sewage sludge (dry weight) are produced annually and around 75% of that undergoes anaerobic digestion (AD) [2]. Despite anaerobic treatment, the resulting sewage digestate is still rich in organic matter and hence, it has the potential to be used as a feedstock for the production of solid energy carriers [1,3–7].

Hydrothermal carbonization (HTC) is considered an alternative

technology to harness energy from sewage digestate, as wet feedstocks are ideally suited to this process – i.e., no reliance on energy intensive dewatering units, as is the case in digestate pyrolysis. HTC is conducted at temperatures ranging from 200 to 250 °C and pressures ranging from 10 to 40 bar [4,8,9]. HTC products include process waters rich in organic compounds suitable for anaerobic digestion and a charcoal like material (hydrochar) that can be used either as a solid fuel or as a soil amender [1,6,8,10]. Hydrochars often have a higher energy density than the feedstock due to deoxygenation [8] and process waters tend to concentrate soluble organic matter and nutrients like nitrogen and phosphorus compounds [11]. The specific hydrochar and process water characteristics however are highly dependent on the choice of feedstock and process conditions [9,12].

The integration of HTC into wastewater systems as a post-treatment step after AD is still under development, but commercial HTC processes are already available – i.e., The Terranova® Ultra-Process [13]. This offers potential energetic and economic

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benefits from the stabilisation of sewage digestate while producing not only a solid fuel product that can be used in a coal fired power plant, but also carbon-rich process water for enhanced biomethane production in existing AD units at WWTWs [11]. The majority of studies reported in published literature on the use of HTC for sewage digestate processing, have been conducted in batch reactors at laboratory scale. Common findings led to conclude that feedstock characteristics, as well as temperature and reaction time are the main operating conditions influencing hydrochar characteristics; in general, the higher the process temperature and the longer the carbonization time, the higher the carbon content and energy density of the resulting hydrochar [1,4,9,11,12,14–18]. For instance, Kim, et al. [5] evaluated HTC process on anaerobically digested sludge at different temperatures (range 180–280 °C) and retention time of 30min. They found that the dewaterability, the carbon densification and heating value, increased as the temperature reaction increased. A similar trend was obtained by Aragón-Briceño et al. [11] in which they evaluated the effect of the HTC reaction temperature (range 180–250 °C) in sewage sludge digestate with a retention time of 30min. Danso-Boateng et al. [4] investigated the effect of process temperature and retention time on the HTC of the sewage sludge digestate and concluded that the carbon densification decreased as the temperature and retention time increased (carbon content from 50 to 77%) but with an increase of the HHV (17–19 MJ/kg). Saetea and Tippayawong [19] studied only the effect of the retention time (1–6 h) in HTC of sewage sludge concluding that there was an increase in the carbon content on the energy properties of the hydrochar for longer process retention time. In general, for sewage digestate in particular, HTC processing at high temperatures and short reaction times however (250 °C, 30min), can still produce a hydrochar with High Heating Values (HHV) in a range suitable to be used as solid energy carriers [11].

In comparison, the influence of solid loading on the characteristics of the resulting hydrochar and process water has received less attention. The very few examples reported in the literature using food waste as feedstock have concluded that higher solid loading contributes to higher hydrochar yields, carbon efficiencies and to improve the energy efficiency of the process [12]. Therefore, the influence of solid loading on HTC used for sewage digestate processing and its effect on the characteristics of the resulting hydrochar and process water have not been reported to date. Most importantly, there are no previous research works studying the influence of solid loading on the anaerobic biodegradability of the process water, its total bio-methane potential, nor the repercussions of varying solid loading in the HTC on the energetics of the whole WWTW. Thus scientific evidence is desperately needed in order to fill this gap and contribute to the better understanding of the overall energy production in an integrated AD + HTC system at sewage treatment works.

Therefore, the main objectives of this study are, firstly, to investigate the influence of solid loading on hydrochar and process water characteristics from HTC used for processing sewage digestate, and secondly explore their effects on the energetics of the WWTW that feature AD coupled with HTC. Production yields and composition of hydrochars and the levels of solubilisation of organic matter and nutrients in process waters are presented. Results from experimental biomethane potential (BMP) tests conducted on process waters are used to present an overall energy balance for the proposed AD + HTC process based on the development of an Aspen Plus plant model. The results reported in this work would inform the potential for implementing a comprehensive treatment process that integrates AD and HTC for sewage sludge management at WWTWs, with the potential to replace current practices for digestate disposal.

## 2. Material and methods

The schematic diagram of the experimental design is presented in Fig. 1.

### 2.1. Sewage digestate sample collection and preparation

A grab sample of sewage digestate was collected from anaerobic digesters processing primary and secondary sludge at the Yorkshire Water's Esholt WWTW in Bradford, UK. A portion of that sample was stored at 4 °C for subsequent characterisation in the laboratory. The remaining sample was centrifuged at 4000 rpm (3220 G) for 30 min and the aqueous fraction (digestate liquor) was separated from the solids and stored at 4 °C before sample preparation. The solid fraction (digestate cake) was dried in an oven at 40 °C for 7 days. The dry digestate and the liquor were used for the preparation of the actual digestate undertaking hydrothermal treatment (HTC) at different solid concentrations (solid loadings).

### 2.2. Hydrothermal carbonization experiments

HTC experiments were conducted in a non-stirred 500 mL stainless steel Parr batch reactor at 250 °C and 40 bar for 30 min, after which the reactor was cooled down to 25 °C before collection of the resulting processed samples (HTC slurries). In each experiment, 220 mL of digestate sample containing a known concentration of solids was loaded into the reactor. The concentrations of solids tested in digestate samples were 2.5, 5.0, 10.0, 15.0, 17.5, 20.0, 25.0 and 30.0% w/w. The resulting HTC slurries were collected and prepared for characterisation of solid and liquid products. Solid (hydrochar) and liquid (process water) products contained in HTC slurries were separated by filtration using pre-weighted Whatman™ glass microfiber filters (Grade GF/C). All HTC experiments were conducted in duplicate.

### 2.3. Feedstock and hydrochar characterisation

Dry digestate cake (feedstock) and hydrochar samples were oven dried for 24 h at 40 °C. Ultimate analysis was performed using a CHNS analyser (CE Instruments, Flash EA 1112 Series). Proximate Analyses were performed using a Thermogravimetric analyser (Shimadzu TGA-50) to determine moisture, ash and volatile matter.

### 2.4. Process waters characterisation

Process waters from the HTC experiments were characterised following Standard Analytical Methods for Chemical Oxygen Demand (COD), Total and Suspended Solids (TS and SS), Volatile Fatty Acids (VFAs), Phosphorus (Total and reactive), Total Kjeldahl Nitrogen (TKN), Ammonia and pH [20]. Ultimate analysis was performed using a CHNS analyser (CE Instruments, Flash EA 1112 Series) for the totally evaporated process water. Total organic carbon analyses were performed using a TOC analyser (HACH Lange, IL550 TOC/TIC Analyser).

### 2.5. Biochemical methane potential (BMP) experimental tests

BMP tests were carried out on process water samples following the method described by Aragón-Briceño et al. [11]. The inoculum concentration used in each BMP test was 10 g/L of Volatile Suspended Solids (VSS) and the process water concentration was 2 g/L of COD maintaining a volume ratio of 1:1. Each BMP test was performed at 37 °C for 21 days, in a series of 120 mL bottles sealed with a rubber stopper and aluminium cap. All the BMP tests were performed in duplicate. Distilled water was used for diluting samples

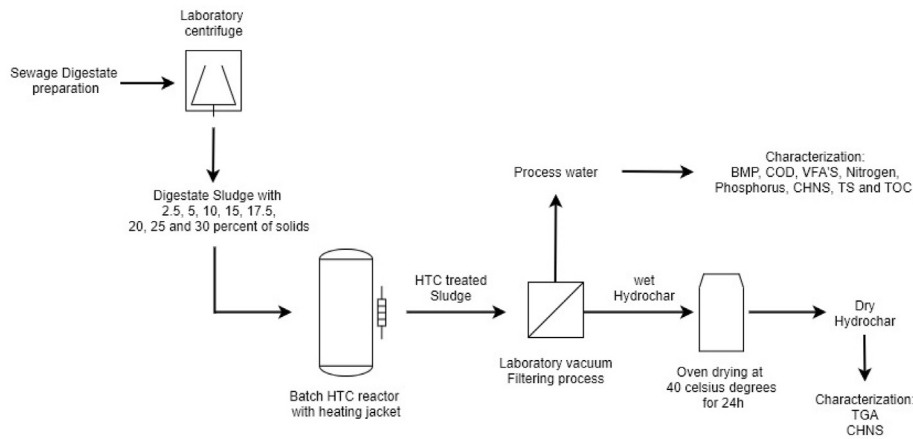


Fig. 1. Schematic Diagram of the experimental design.

to reach the set COD concentration and volume (60 mL for each reactor). The head space of each bottle was filled with nitrogen gas (Grade N4.0, 99.99%) to keep the anaerobic conditions and remove oxygen from inside the bottle. Test bottles were kept undisturbed at all times, except when mixing by hand during biogas production measurements. Methane production was monitored by using a volumetric method with a solution of 1M NaOH. For every measurement, a bottle was sacrificed to perform the analyses. During the experiment, the following parameters were monitored: pH, TS, VS, COD and VFAs. TKN, Ammonium and Phosphorus (total and reactive) were measured at days 0, 3, 5, 7, 11, 14, 17 and 21. All the BMP analyses were carried out in duplicate.

### 2.5.1. Inoculum

The inoculum used for BMP tests was obtained from the outlet of an anaerobic reactor used for sewage sludge digestion at Yorkshire Water's Esholt WWTW in Bradford, UK. The Inoculum was incubated at 37 °C in sealed bottles and fed every week with fresh sewage sludge to keep it active.

## 2.6. Experimental data processing and analysis

Data processing from hydrochar analyses was made using the following equations reported by Aragón-Briceño et al. [11]:

### 2.6.1. Hydrochar yield

Hydrochar yield ( $Y$ ), energy densification ( $E_d$ ) and energy yield ( $E_y$ ) were determined as follows:

$$Y(\%) = \frac{\text{mass of dry hydrochar(g)}}{\text{mass of dry Substrate feedstock(g)}} * 100 \quad (1)$$

$$E_d = \frac{HHV_{char}}{HHV_{feedstock}} \quad (2)$$

$$E_y(\%) = E_d \times Y \quad (3)$$

where  $HHV$  is High Heating Value on a mass basis.

### 2.6.2. Carbon recovery in solid and liquid fractions after HT processing

Carbon recovery in hydrochar ( $Hy_{crec}$ ) and liquid phase ( $Lq_{crec}$ ) were calculated as follows:

$$(\%)Hy_{crec} = \frac{\frac{\%C_{Hydrochar}}{100} \times \text{char mass}}{\frac{\%C_{feedstock}}{100} \times \text{mass of dry Digestate feedstock(g)}} * 100 \quad (4)$$

$$(\%)Lq_{crec} = \frac{\text{Total organic Carbon (g/L)} \times \text{volume of filtrate(L)}}{\frac{\%C_{feedstock}}{100} \times \text{mass of dry Digestate feedstock(g)}} * 100 \quad (5)$$

### 2.6.3. High Heating Value (HHV)

In order to determine the theoretical calorific value of the hydrochar, the Dulong equation reported by Channiwala and Parikh [21] was used.

$$HHV(\text{MJ / kg}) = 0.336(\%C) + 1.433 \left( \%H - \left( \frac{\%O}{8} \right) \right) + 0.0942(\%S) \quad (6)$$

### 2.6.4. Biochemical Methane Production (BMP)

In order to assess the amount of methane production per gram of chemical oxygen demand (COD) added, the following BMP formula was used:

$$BMP = \frac{V_{CH_4} - V_{CH_4,blank}}{(\text{Mass of COD fed in biodigester})} \quad (7)$$

Where:

BMP=Biochemical Methane Potential (mL of CH<sub>4</sub>/g of COD added)

VCH<sub>4</sub> =Volume of methane produced in bottle (mL)

VCH<sub>4, blank</sub> = Volume of methane produced in the blanks (mL)

Mass of COD = Mass of COD of the substrate (g of COD substrate)

### 2.6.5. Theoretical BMP (BMP<sub>th</sub>)

The calculation of theoretical BMP values, which are based on the elemental composition (C, H, N and O) of the samples, was made by using stoichiometric equations for maximum biogas production. Boyle's equation was used to calculate the theoretical BMP values for each tested sample [22,23].

Boyle's equation:

$$BMP_{thBO} = \frac{22400 \left( \frac{n}{2} + \frac{a}{8} - \frac{b}{4} - \frac{3c}{8} \right)}{12n + a + 16b + 14c} \quad (8)$$

where  $n$ ,  $a$ ,  $b$  and  $c$  represent the molar fractions of C, H, O and N, respectively.

#### 2.6.6. Anaerobic biodegradability (BD)

The anaerobic biodegradability of each sample was calculated from the values reported from the experimental BMP ( $BMP_{exp}$ ) and the theoretical BMP ( $BMP_{Th}$ ) and gives an idea of the level of biodegradability of the slurries and process waters under anaerobic conditions [23]:

$$BD_{CH_4}(\%) = \frac{BMP_{exp}}{BMP_{Th}} \times 100 \quad (9)$$

#### 2.7. AD + HTC system energetics analysis

Aspen Plus was used to analyse, in more detail, the potential to integrate AD and HTC processes in a WWTW under the conditions examined experimentally. This allowed the creation of robust mass and energy balances of proposed HTC solid concentration scenarios. HTC, AD and CHP (combined heat and power) systems were all simulated and integrated to form a representative flow sheet. Aspen Plus V8.8 was used throughout with a 'COMMON' method filter and an 'IDEAL' base method. The following assumptions were made: ambient conditions of 1 bar and 23 °C and molar air composition assumed as 79:21 split of  $N_2:O_2$  only.

Experimentally obtained ultimate and proximate results of digestate sludge and associated hydrochar after HTC at 250 °C and 40 bar were used to create 'nonconventional solid' components for their representation in the model. Acetic acid was used as a model representation of COD for the liquid fraction of the digestate liquor and HTC process water, where there is 0.938 g of acetic acid for 1.0 g of COD. Ammonia was used to represent total aqueous nitrogen, and phosphoric acid represented total aqueous phosphorus.

System efficiencies were calculated to highlight the most energetically beneficial solids concentration feedstock for HTC. Electrical efficiency ( $\eta_{P,net}$ ) (based on HHVs) was calculated via Equation (10) and system thermal efficiency ( $\eta_{Q,net}$ ) was determined via Equation (11):

$$\eta_{P,net} = \frac{P_{net}}{\dot{m}_{biogas} \cdot HHV + \dot{m}_{hydrochar} \cdot HHV} \times 100 \quad (10)$$

$$\eta_{Q,net} = \frac{Q_{net}}{\dot{m}_{biogas} \cdot HHV + \dot{m}_{hydrochar} \cdot HHV} \times 100 \quad (11)$$

where  $P_{net}$  and  $Q_{net}$  are the system net electrical and thermal power productions respectively, while  $\dot{m}_{biogas}$  and  $\dot{m}_{hydrochar}$  are the mass flow rates of biogas and char, respectively.

Aspen Plus does not currently have the capability of processing reactions involving 'nonconventional solids', which were used to represent hydrochar in the model. Therefore, the cogeneration potential from hydrochars was inferred from a study by Liao et al. (2013) that analysed the CHP efficiencies of a coal-fired CHP plant in China, by assuming similar efficiencies of conversion to heat and power between hydrochar and coal. In Liao et al. (2013), the system utilised a coal-fired boiler to generate pressurised steam for power generation from turbines. From their analysis, mean electrical and thermal efficiencies of 28% and 43% were calculated respectively on

a LHV-basis. These figures have subsequently been used in the present study to indicate the energy recovery potential from hydrochars generated in the discussed process. Hydrochar LHV values were calculated via the formula presented in Nzihou et al. [24] and shown in Equation (12):

$$LHV(MJ/kg) = 4.18 \times (94.19 \times \%C - 0.5501 - 52.14 \times \%H) \quad (12)$$

where C and H are the mass percentage in dry base from the elemental analysis of carbon and hydrogen respectively.

### 3. Results and discussions

#### 3.1. Mass balance

The distribution of products from sewage digestate before and after HTC at different solid loadings is presented in Fig. 2. The output mass of combined solid and liquid fractions was reduced after HTC treatment by 1.4–3.5%. These values were slightly lower compared with the study carried out by Zabaleta et al. [12], who reported mass losses between 2.3 and 7.1% when food waste was under HTC processing at different solid loadings and different temperatures (180–200 °C); mass losses were attributed to the production of gaseous components, mainly  $CO_2$  [12].

The solid fraction of the feedstock was reduced between 24 and 37% following HTC. That is due to solubilisation of some of the original biomass into the liquid phase during HTC, which includes both soluble inorganic and organic material [11,14]. Nevertheless, as the solid loading increased, there was a slight increase in the yield of solid product following HTC (see Fig. 2b).

#### 3.2. Hydrochar characteristics

##### 3.2.1. Physical characteristics

The yield of hydrochar generally increases with increasing solid loading in agreement with the results reported from food waste by Zabaleta et al. [12]. Hydrochar yields range from 67.9% at 2.5 wt% loading to 75.6% at 25.0% loading (Table 1). The yields obtained in this study are similar to the values reported by Danso-Boateng et al. [4] from primary sewage sludge (60.5–81.1% at 4.5% solid loading) and slightly higher than the findings reported by Aragón-Briceño et al. [11] from sewage digestate (56.8% at 4.5% solid loading).

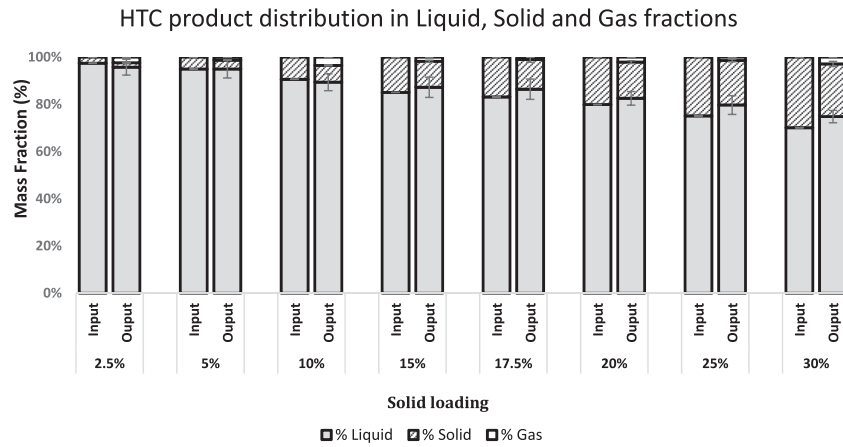
The ash content of the resulting hydrochar reduced as the solid loading increased from 51.2% at 2.5% loading to 48.5% at 30% loading (Table 1). This suggests that less carbon was solubilised as the solid loading increased and correlated with a slight increase in hydrochar yield. The ash content of hydrochars were similar to those reported for hydrochar produced from sewage sludge [4,11,14,25].

The volatile matter content of the resulting hydrochars was similar at all solid loadings and ranges between 40.4 and 42.4%; however, the volatile matter content of the hydrochars was lower than the feedstock's – i.e., 51.8% for the feedstock, while hydrochars had a minimum of 40.3% after HTC.

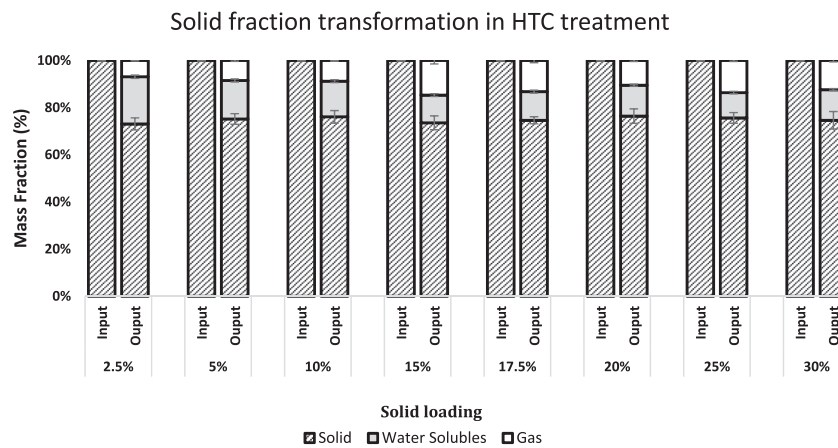
##### 3.2.2. Elemental composition of the hydrochar

The elemental compositions of hydrochars following HTC are shown in Table 1. The carbon content of the hydrochars increased with increasing solid loading (from 32.1% at 2.5% solid loading to 34.4% at 30.0% solid loading), but there only a slight increase in carbon content compared to the original feedstock was achieved at the highest solid loading tested ( $\geq 20\%$  solid loading). Previous reports have observed that the carbon content in hydrochars produced from sewage digestate via HTC processing ranged from 10 to

a)



b)



**Fig. 2.** Changes in the feedstock after HTC at different solid loadings. a) Product distribution in Liquid, Solid and Gas fractions and b) Fate of solids from the feedstock.

**Table 1**

Proximate and ultimate analyses of the feedstock (digestate cake) and hydrochar.

Sample	Proximate Analyses				Ultimate analysis (db)					Yield (%)
	Moisture (%)	Ash (%db)	Volatile matter (%db)	Fixed carbon <sup>a</sup> (% db)	C (%)	H (%)	N (%)	O <sup>b</sup> (%)	S (%)	
Feedstock	2.1	36.7	51.8	9.34	33.3	4.6	4.0	20.3	1.2	
Hydrochar										
2.5%Hy	2.0	51.2	41.9	4.9	32.1	4.2	1.9	9.4	1.2	68%
5.0%Hy	1.7	50.0	41.7	6.5	32.3	4.2	2.1	10.3	0.8	72%
10.0%Hy	0.9	48.4	42.4	8.4	33.1	4.4	2.3	10.5	1.2	75%
15.0%Hy	1.7	49.4	40.4	8.5	33.0	4.3	2.3	9.8	1.3	74%
17.5%Hy	1.7	49.7	40.7	7.9	33.1	4.2	2.3	9.4	1.3	75%
20.0%Hy	1.6	48.6	41.5	8.4	33.8	4.3	2.4	9.7	1.3	76%
25.0%Hy	1.7	48.0	41.7	8.5	33.8	4.3	2.6	9.9	1.3	76%
30.0%Hy	1.9	48.5	41.2	8.4	34.4	4.4	2.8	8.7	1.2	75%

db = dry base.

<sup>a</sup> 100 - (moisture + ash + volatile matter).

<sup>b</sup> Calculated as difference between sum of C,H,N and S content.

39% [4,11,14,18]. However, carbon yields are highly dependent upon feedstock composition and process conditions (i.e., temperature, pressure, solid loading, etc.) [26]. The carbon content of the

hydrochars was reduced after HTC compared to the feedstock (see Table 1), which is unusual and only observed for certain feedstocks such as sewage digestate [11].

The carbon balance across the solid and liquid products is presented in Table 2. The carbon recovery in the hydrochar ( $H_{yCrec}$ ) increased as the solid loading increased. On the other hand, the carbon recovered in the liquid fraction ( $L_{qCrec}$ ) reduced as the solids loading increased. Funke and Ziegler [26] reported that wet biomass can be almost completely dissolved into the liquid fraction at low solid loading. This suggests that there is a saturation point in which solubility becomes important. The recovery of carbon in the hydrochar is likely to be influenced by the degree of polymerization occurring during HTC and the solubility limits in the water. In this study, the  $H_{yCrec}$  ranged from 65.5 to 77.6% and  $L_{qCrec}$  ranged from 16.9 to 35.8%. The values obtained were similar to those obtained by Aragón-Briceño et al. [11].

Levels of oxygen were reduced significantly following HTC due to the occurrence of dehydration and decarboxylation reactions. Fig. 3 shows Hydrogen-to-Carbon (H/C) and Oxygen-to-Carbon (O/C) ratios of the feedstock and hydrochars. The slight reduction of the H/C ratio in the hydrochars provides evidence for dehydration and decarboxylation during hydrothermal carbonization [4]. Nevertheless, changes in solid loading did not provide a clear correlation with regard to its influence on dehydration and decarboxylation reactions, in agreement with the findings reported by Zabaleta et al. [12]. Nitrogen content in the hydrochar increases along with increments in solid loading (see Table 1); however, all hydrochars had a much lower N content (1.9–2.8%) when compared with the original feedstock (4.0%) as the hydrolysis of N-rich compounds during HTC promoted the accumulation of ammonium in process waters [11].

### 3.2.3. Energy characteristics of hydrochars

The energy density of hydrochars and feedstock are listed in Table 2. The Higher Heating Value (HHV) of the hydrochars were only slightly higher than the original feedstock (14.4 MJ/kg), with a maximum value of 16.5 MJ/kg at 30 wt% solid loading. This low level of energy densification is typical for wet feedstocks such as sewage digestates, which tend to result in larger levels of solubilisation of the organic carbon during HTC processing. There was a slight increase in HHV as solid loading increased and this corresponded to a higher carbon content in the resulting hydrochar. The energy densification values obtained for hydrochars ranged from 0.97 to 1.03 MJ/kg. The HHVs of the hydrochars produced were higher compared with the values reported by Berge et al. [18] for digestate (13.7 MJ/kg) and lower than those reported by Danso-Boateng et al. [4] (17.2–18.4 MJ/kg) and Aragón-Briceño et al. [11] (17.8 MJ/kg) for primary sludge and digestate, respectively. The energy densification recovered within the hydrochar was considerably lower compared with other feedstocks, but indicated that a significant amount of the energy was present in the liquid fraction and potentially available for recovery via anaerobic digestion [11].

**Table 2**  
- Energy characteristics of the feedstock and hydrochars.

Hydrochar	HHV (Mj/kg)	Energy densification (Mj/kg)	Energy Yield (%)	$H_{yCrec}$ (%)	$L_{qCrec}$ (%)
Feedstock	14.4	–	–	–	–
2.5%Hy	15.4	0.97	65.9	65.5	33.4
5%Hy	15.3	0.97	69.8	69.9	35.8
10%Hy	15.8	1.00	74.4	74.3	27.1
15%Hy	15.7	0.99	73.1	73.0	16.9
17.5%Hy	15.6	0.97	72.5	74.3	19.4
20%Hy	15.9	1.00	76.2	77.6	18.6
25%Hy	16.0	1.01	76.6	76.8	17.5
30%Hy	16.5	1.03	76.7	77.1	20.5

HHV: High heating value.

$H_{yCrec}$ : Carbon recovered in the solid fraction.

$L_{qCrec}$ : Carbon recovered in the liquid fraction.

The energy yield provides useful information about the amount of energy remaining within the hydrochar from the original feedstock. The energy yield showed a similar trend as HHV with ranges from 65.9 to 76.7%, but it was observed that upward of 20% solid loading in HTC, the energy yield plateaued.

### 3.2.4. Nutrient balance

The fate of phosphorus and nitrogen after HTC is shown in Fig. 4a and b. Fig. 4a indicates that there was a solubilisation of phosphorus into the liquid fraction (up to 25%) as reported by Aragón-Briceño et al. [11]. However, the majority of the phosphorus therefore remained in the hydrochar (66.8–75.7%).

Fig. 4b shows that a significant proportion of nitrogen from the feedstock was solubilised into the liquid fraction after HTC treatment. The amount of nitrogen transferred from the feedstock into the water increased up to 48% of the total nitrogen content into the liquid, leaving the nitrogen content in the hydrochar ranging between 15 and 50%. Solid loading also significantly influenced the level of nitrogen solubilisation in the process water following HTC. The levels of soluble N decreased as the solid loading increased. The change in feedstock N was due to the liquor containing much higher levels of soluble N than the press cake.

### 3.3. Characteristics of the process waters

The composition of the process waters following HTC at different solid loadings are listed in Table 3. Properties such as pH and soluble hydrocarbons were measured by total organic carbon (TOC) and chemical oxygen demand (COD). The level of soluble inorganic species containing nitrogen were measured by Total Kjeldahl Nitrogen (TKN) and soluble ammonia, total and reactive phosphorus (TP and RP), and total solids (TS) and total volatile solids (TVS) were also measured. The levels of volatile fatty acids (VFA) and the elemental composition of the process waters (CHNOS) were also determined on the evaporated process waters. All of these properties were shown to change with solid loading.

#### 3.3.1. pH

The pH values of process waters are listed in Table 3. The results indicate that the pH of the process waters after HTC treatment increases with solid loadings for all sewage digestate samples from 7.7 to 8.3. Changes in pH are mainly related to the presence of organic and inorganic compounds [27]. The increasing pH is linked to the formation and solubilisation of ammonium and solubilisation of alkaline salts [6,11,14]. Furthermore, according to Berge et al. [18], the pH of an anaerobically treated waste can remain basic depending on its buffering capacity, which may hinder the initial hydrolysis step during the thermal process.

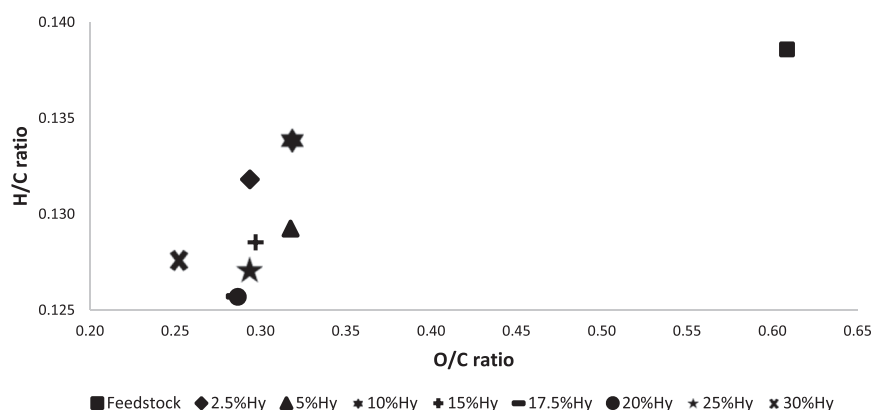


Fig. 3. Atomic H/C and O/C ratios of feedstock and hydrochars following HTC (250 °C and 30 min retention time) at different solid loadings.

### 3.3.2. Total solids and total volatile solids

Table 3 lists the total solid (TS) and total volatile solid (TVS) concentration of process waters at different solid loadings. As expected, the TS concentration was directly related to the amount of solid loading. HTC results in the solubilisation of organic material following hydrolysis [14,28]. TS concentration in process waters increased from 2.4 g/L initially present in the digestate liquor to a maximum of 39 g/L in the process water at 30% solids loading.

The solubilisation of total and volatile solids (TS and VS) into the process waters at different solid loadings are reported in Fig. 5d and demonstrates a significant effect of the solid loading on the solubilisation of organic compounds. At high solid loadings, the concentration of TS and TVS were higher, but the solubilisation was lower due to saturation in the liquid fraction. The highest solubilisation was observed at 2.5% solid loading, which corresponded to 0.17g of TVS solubilised per gram of feedstock processed. As the solid loadings increased beyond 15.0%, the solubilisation became constant having values between 0.10 and 0.12g of TVS solubilised per gram of feedstock processed.

### 3.3.3. Chemical oxygen demand (COD) and total organic carbon (TOC)

The levels of water soluble products generally increased with reaction severity due to the solubilisation of inorganics and the increased production of soluble organics from hydrolysis [14,27,29]. However, the composition of carbon and nutrient rich compounds depends mainly on the nature of the feedstock being treated and process temperature [14,30].

The solubilisation of carbon compounds is due to hydrolysis, which releases organic compounds such as acetic acid, butanoic acid, alkenes, phenols, etc. [4]. Therefore, it is reflected in the increasing amount of COD and TOC measured in process waters.

The COD concentration of the filtered sewage digestate (liquor) was 2100 mg of COD/L. After the HTC process, the COD concentration increased significantly between 9500 and 72,300 mg of COD/L depending on the solid loading during HTC processing. The solubilisation of organic compounds into process waters raises the possibility of recycling some of the carbon embedded in sewage digestate back into the anaerobic digester to boost methane yields, and of reducing fugitive emissions of methane from the digestate cake when the latter is dispersed on arable land. However, process waters need to be carefully added to anaerobic digesters as operational organic loading rates should not exceed design criteria, as higher concentrations of COD may affect the balance between hydrolysis, acetogenesis and methanogenesis in anaerobic digesters. According to the study carried out by Hübner and Mumme [10],

concentrations of organic matter in the feedstock exceeding 30g of COD per L can permanently inhibit methanogenesis due to over-acidification. Therefore, the right recirculation rate of process waters is a factor that must be considered when enhanced methane production in anaerobic digesters is selected as the preferred route for the valorisation of process waters.

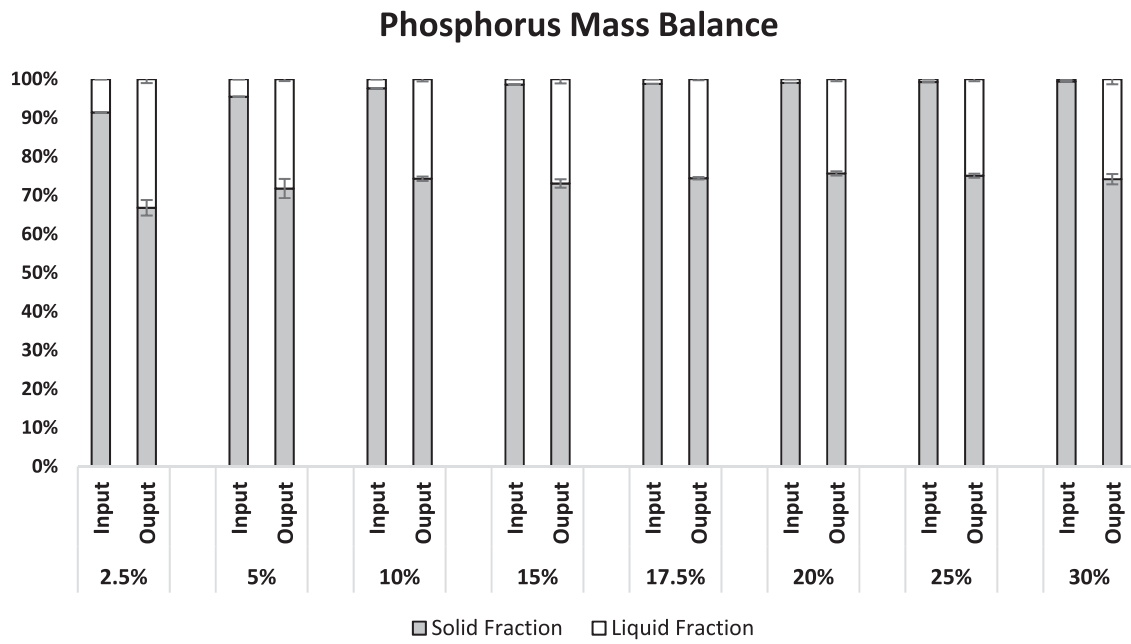
TOC concentration in process waters followed the similar trend found for COD. The concentration of TOC in the digestate liquor was 800 mg of Carbon/L but after HTC, TOC concentration in process waters increased to a maximum of 27,900 mg of Carbon/L at the highest solid loading tested. Both COD and TOC concentrations, increased with respect to the amount of solids in the mix (see Table 3). In the study conducted by Stemann et al. [31], COD and TOC concentrations increased similarly. An increase in the percentage of elemental carbon in the evaporated process waters was observed and ranged from 43.9 to 54.0%. Comparable results were reported by Aragón-Briceño et al. [11], who used a similar sewage derived digestate and reported elemental carbon content in process waters ranging from 46 to 68%.

The solubilisation of organic matter in process waters was found to range between 240 and 360 mg of COD solubilised per gram of feedstock processed (Fig. 5a) and between 100 and 140 mg of Carbon per gram of feedstock processed. That corresponded to an increased solubilisation 3 to 4.5 times higher compared with the digestate liquor based on COD, and between 4 and 6 times higher based on TOC (80 mg of COD per gram of feedstock and 20 mg of Carbon per gram of feedstock). The solubilisation of organic matter from the feedstock's solid fraction increased until a maximum was found at 15% solid loading; however, carbon solubilisation became constant beyond this threshold (see Fig. 5a), as the aqueous phase saturated and any excess hydrolysed material could concentrate on the hydrochar. The saturation concentration of hydrolysed organic compounds is important to consider as it is possible that additional washing of hydrochars may liberate additional soluble organic compounds, this in turn may improve the properties of the hydrochars for further applications.

### 3.3.4. Volatile Fatty Acids (VFAs)

Table 3 presents the concentration of VFAs in process waters, which indicates an increase in VFAs with increasing solid loading. In this context, VFA analysis refers to the presence of C1–C6 organic acids and includes acetic acid, propanoic acid, isobutyric acid, butyric acid, isovaleric acid and valeric acid. VFAs concentration ranges from 909 to 4606 mg of COD/L (2.5 and 30% solids concentration, respectively). VFAs can be attributed to the decomposition of hydrolysis products during the HTC process [18]. Berge

a)



b)

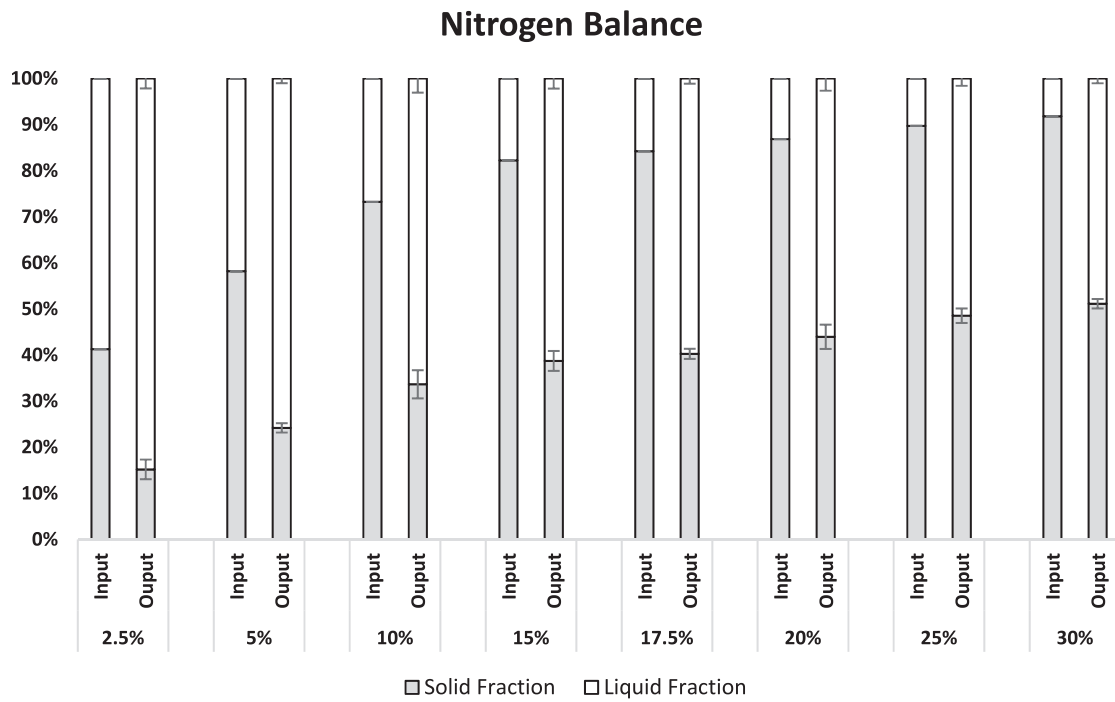


Fig. 4. Mass balance distribution of Phosphorus (a) and Nitrogen (b) before and after HTC treatment at different solid loadings.



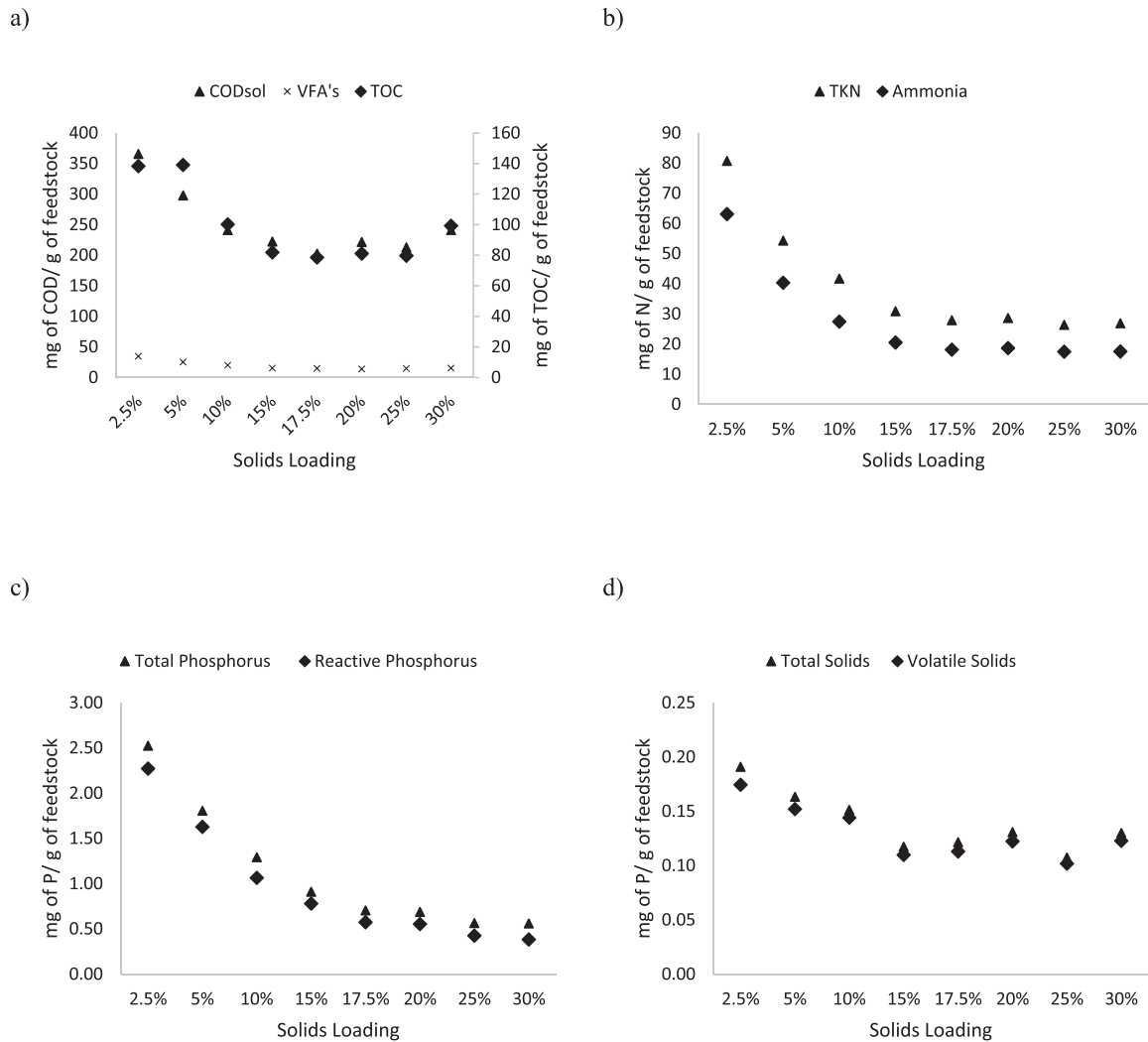
**Table 3**  
Characteristics of digestate liquor and process waters from HTC at different solids loading.

Parameter	Process waters from HTC at different loadings								
	Liquor	2.5%	5.0%	10.0%	15.0%	17.5%	20.0%	25.0%	30.0%
pH	7.7	7.9	7.8	8.1	8.1	8.1	8.2	8.3	8.3
CODsol (mg/L)	2103	9575	14894	24114	33333	35461	44326	53192	72340
TOC (mgC/L)	657	3623	6953	10016	12277	13737	16215	19922	29778
TKNsol (mgN/L)	1463	2114	2716	4172	4634	4886	5726	6594	8064
Ammonia (mgN/L)	1316	1652	2016	2744	3080	3173	3733	4368	5264
TP <sup>b</sup> sol(mgP/L)	48.4	66.1	90.3	129.2	136.6	123.2	137.7	141.3	167.6
RP <sup>c</sup> (mgP/L)	38.1	59.5	81.4	106.4	117.0	100.5	110.8	106.4	114.7
TS (g/L)	2.4	5.0	8.2	15.1	17.6	21.3	26.2	26.9	39.0
TVS (g/L)	1.7	4.6	7.6	14.4	16.5	19.8	24.5	25.4	36.8
VFAs (mg of COD/L)	350	909	1265	2009	2317	2587	2814	3705	4606
C (wt%)	43.9	44.1	48.0	52.2	51.9	51.8	52.7	53.1	53.1
H (wt%)	5.4	4.8	5.6	6.8	6.4	6.5	7.0	6.8	6.4
N (wt%)	3.5	5.6	6.8	7.8	7.5	7.5	6.7	7.5	6.9
O <sup>a</sup> (wt%)	47.1	44.1	38.2	32.0	33.0	32.9	32.7	31.4	32.4
S (wt%)	0.1	1.4	1.4	1.2	1.3	1.3	1.0	1.2	1.1

<sup>a</sup> Calculated as difference between sum of C,H,N,S.

<sup>b</sup> Total Phosphorus.

<sup>c</sup> Reactive Phosphorus.



**Fig. 5.** Solubilisation of (a) carbon rich compounds (Chemical Oxygen Demand (COD), VFAs (Volatile Fatty Acids) and Total organic carbon (TOC)); (b) nitrogen rich compounds (Total Kjeldahl Nitrogen (TKN) and Ammonium; (c) phosphorus rich compounds (Total Phosphorus (TP) and Reactive Phosphorus (RP)); and (d) solids (Total Solids (TS) and Volatile Solids (VS)).

et al. [18] and Danso-Boateng et al. [4] detected acetic, propanoic, and butanoic acids together with many other organic and inorganic compounds like aromatics, aldehydes and alkenes. The solubilisation of VFAs (Fig. 5a) followed the same trend as the other parameters measured where higher levels of solubilisation were achieved for the lower solid loadings (2.5, 5.0 and 10%) and became constant beyond 15% of solids loading.

### 3.3.5. Phosphorus

The solubilisation of phosphorus in process waters following HTC is due to decomposition of complex organic phosphorus containing compounds (e.g., phospholipids, DNA and phosphates monoesters), which results in a combination of reactive ( $\text{PO}_4^{3-}$ ) and organic phosphorus compounds in solution [14,32]. Table 3 shows the total and reactive phosphorus concentrations in the digestate liquor and process waters at different solid loadings. The results indicate that the concentration of phosphorus (total and reactive) increased as the solid loading increased. The concentration of total phosphorus ranged from 66 to 167 mg P/L and for reactive phosphorus from 59 to 114 mg P/L for process waters derived from mixes containing 2.5 to 30 wt% solid loading (i.e., the difference between total and reactive phosphorus gives an estimate of the concentration of organic phosphorus compounds in solution). The results indicate that the total and reactive phosphorus concentrations increased with increasing solid loading. However, once again, a saturation point was reached, with the reactive phosphorus remaining relatively constant beyond a solid loading of 15%. Despite the increase of TP and RP in process waters, these only represented a small proportion of the total phosphorus originally present in the feedstock. The solubilisation of phosphorus in mg/g of feedstock (TP and RP) is shown in Fig. 5c. The overall phosphorus solubilisation from the feedstock decreased as the solid loading increased. This is typical of HTC feedstocks containing counter ions such as  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$  that are capable of promoting P precipitation as  $\text{PO}_4^{3-}$  on the hydrochar surface.

In Fig. 6b it is possible to observe that the percentage of phosphorus solubilised from the solid fraction ranged from 24 to 27%. This shows that the phosphorus transferred from the solid fraction to the liquid fraction remained constant independently of the solid loading.

### 3.3.6. Nitrogen

Sewage sludge contains large concentrations of organic matter from faecal material (primary sludge) and bacterial biomass (surplus activate sludge), that largely contribute to the presence of nitrogen compounds in anaerobic digesters processing sewage sludge. During anaerobic digestion, nitrogen compounds are taken up by anaerobic bacteria that mainly constitute the solid fraction of the digestate. For that reason, when hydrothermal treatment is performed, proteins are hydrolysed resulting in the release of soluble ammonium in process waters [29,33].

Table 3 shows the concentration of TKN and ammonium in process waters from HTC. As expected, there was an increase in TKN and ammonium concentrations as the solid loading increased, with figures ranging from 2114 to 8064 mg N/L of TKN and from 1652 to 5,264 mg N/L of ammonium, at 2.5 and 30 wt% solid loading respectively.

The effect of solid loading on the solubilisation of nitrogen compounds followed a similar trend found with other organic and inorganic species (phosphorus and carbon containing compounds – i.e., TP, RP, COD, TOC, TS, VS, etc.), and resulted in an increase in the concentration of nitrogen in process waters (soluble TKN and ammonium). The solubilisation of nitrogen compounds in mg of N/g feedstock (N reported herein using soluble TKN and ammonium analysis) is shown in Fig. 5b. Nitrogen solubilisation ranged from

80.72 to 26.38 mg of N-TKN/g feedstock and from 63.8 to 17.47 mg of N-Ammonium/g feedstock within the solid loading tested (2.5–30 wt%); however, it seems that N solubilisation reached a maximum at 15 wt% solid loading, which then became relatively constant at higher solid loadings (see Fig. 5b).

In Fig. 6a shows the percentage of nitrogen extracted from the solid fraction exclusively into the process water. Nitrogen compounds present in the solid fraction of the anaerobic digestate were hydrolysed and solubilised into process water with efficiencies ranging between 43 and 66%. However, it was observed that the percentage of nitrogen solubilised into the process waters was higher at low solid loadings, which could infer some dependency on process conditions (temperature, pressure, contact time, etc.).

### 3.4. Anaerobic biodegradability and biomethane potential (BMP) of process waters

According to Aragón-Briceño et al. [11], process waters derived from sewage digestate are proven to be a suitable substrate for biomethane production via anaerobic digestion. Anaerobic biodegradability of HTC process waters should not be limited by hydrolysis as most of the complex organic matter has been already hydrolysed during thermal processing [15]; however there are some organic inhibitors (e.g., phenols and PAHs) that can affect the anaerobic digestion process as a whole but mainly the methanogenesis step [10].

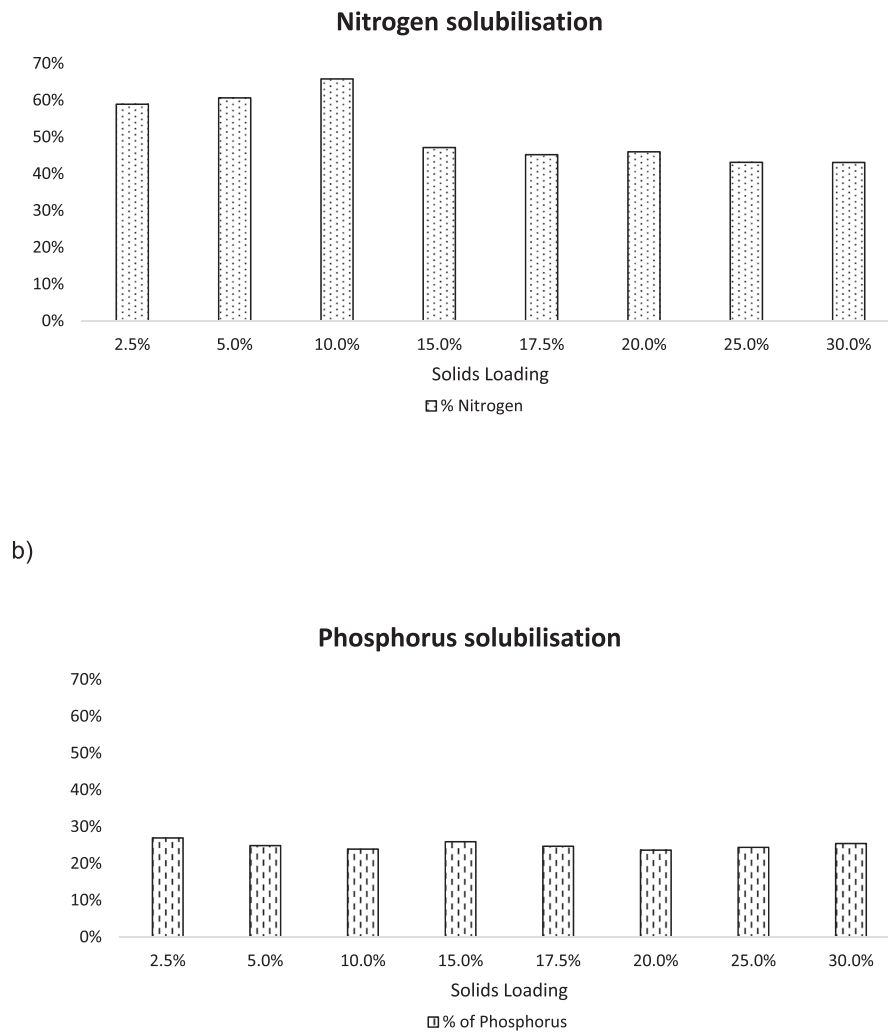
Fig. 7a shows the results from BMP tests for process waters generated at the different solid loading rates tested. A significant increment in methane yields was observed when digestate liquor used as a control (131 mL  $\text{CH}_4/\text{g}$  COD on average) was compared with process waters (228–301 mL  $\text{CH}_4/\text{g}$  COD). It is worth mentioning that methane yields increased with solid loadings until a maximum was reached at 10% before decreasing. This may be due to process waters generated from higher solid loadings having higher levels of phenols as found by Berge et al. [18] Previous studies investigating the anaerobic digestion of HTC and pyrolysis derived process waters from digestate report methane yields ranging from 220 to 227 mL of  $\text{CH}_4$  per g of COD [11,15].

COD consumption during anaerobic processing is presented in Fig. 7b. According to Becker et al. [34], the anaerobic degradation of HTC process waters should not be limited by hydrolysis as only small concentrations of complex organic matter are in the aqueous phase following thermal treatment and hence, organic matter removal is expected to be higher from process waters. COD removal was found to range from 55 to 81%, with the process water from 2.5% of solid loading resulting in the highest COD removal, while the lowest COD removal was obtained from process water from 15% of solid loading. VFAs were entirely consumed, with the exception of the control (Digestate Liquor) that showed no additional biogas production after day 9th (see Fig. 7c). These results were similar to the values of COD removal (63.8%) reported at similar thermal conditions (250 °C, 30 min and 40 bar) with sewage digestate [11] and match reported data from other studies with HTC and Pyrolysis process waters treated anaerobically (32–75% COD removal) [10,15].

Regarding to the biogas composition (Table 4), methane concentration ranged between 74 and 80 vol% showing a good quality biogas coming from process waters at different solids loading. These were slightly higher than the figures obtained by Wirth and Mumme [15] in HTC liquor from corn silage (70% methane).

### 3.5. Maximum potential methane yields

Empirical formulas are widely accepted to estimate methane potential production because they produce fast, economical and



**Fig. 6.** Percentage of Nitrogen (a) and Phosphorus (b) extracted from the original solids into process waters after HTC processing.

representative data, with close matches to experimental results even for process waters [11]. Table 4 shows the results from using Boyle's equation to predict theoretical BMP values from the control samples and process waters, allowing a comparison with the corresponding experimental BMP values.

According to Raposo et al. [23], anaerobic biodegradability of feedstocks can be determined from  $BMP_{exp}$  and  $BMP_{th}$  values. The anaerobic biodegradability is defined as the amount of organic matter that can be degraded during the anaerobic process. The biodegradability (BD) of process waters is listed in Table 4 and shows that the values ranged from 75 to 89%. It is expected that the predicted values should be higher than the experimental counterparts, as the calculations are based on elemental content [11].

The biodegradability of the process waters increased from 36 to 89% compared with the digestate liquor (control). This demonstrates that the digestates still have significant organic content that can be used to produce methane and can be solubilised through HTC. The biodegradability in the process water decreased as the solid loading increased.

### 3.6. AD + HTC system energetics analysis

Fig. 8 details the Aspen Plus flowsheet built for energetic,

scenario and system analysis. The HTC process begins with sludge contained in the stream labelled 'INLET', which was pumped to a pressure of 40 bar and contained a symbolic flow of 1 kg of solids per hour. The quantity and composition of liquid in the 'INLET' stream depended on the solids loading scenarios, which corresponded to those experimentally analysed: 2.5%, 5%, 10%, 15%, 17.5%, 20%, 25% and 30%. The pressurised sludge then exchanged heat with the HTC outlet in the heat exchanger 'HX'. The sludge was then heated to the desired HTC temperature of 250 °C. The HTC reactor was represented by a 'RYield' block, where RYield reactors allow the user to specify desired yields of components at a given temperature and pressure. Experimental data was used accordingly so the reactor outlet held hydrochar and process water in the quantity and composition found experimentally under the 40 bar, and 250 °C conditions.

A separator block was used to represent a centrifuge separator which split the solid and liquid fractions. A heater block was positioned before the AD unit to represent the thermal energy requirements of heating the process liquid to 35 °C. The process water was then sent to an anaerobic digester that was simulated in another RYield block at 35 °C and 1 bar. Here, BMP experimental results were used to determine its outlet yield composition. A Separator block located downstream represented the extraction of

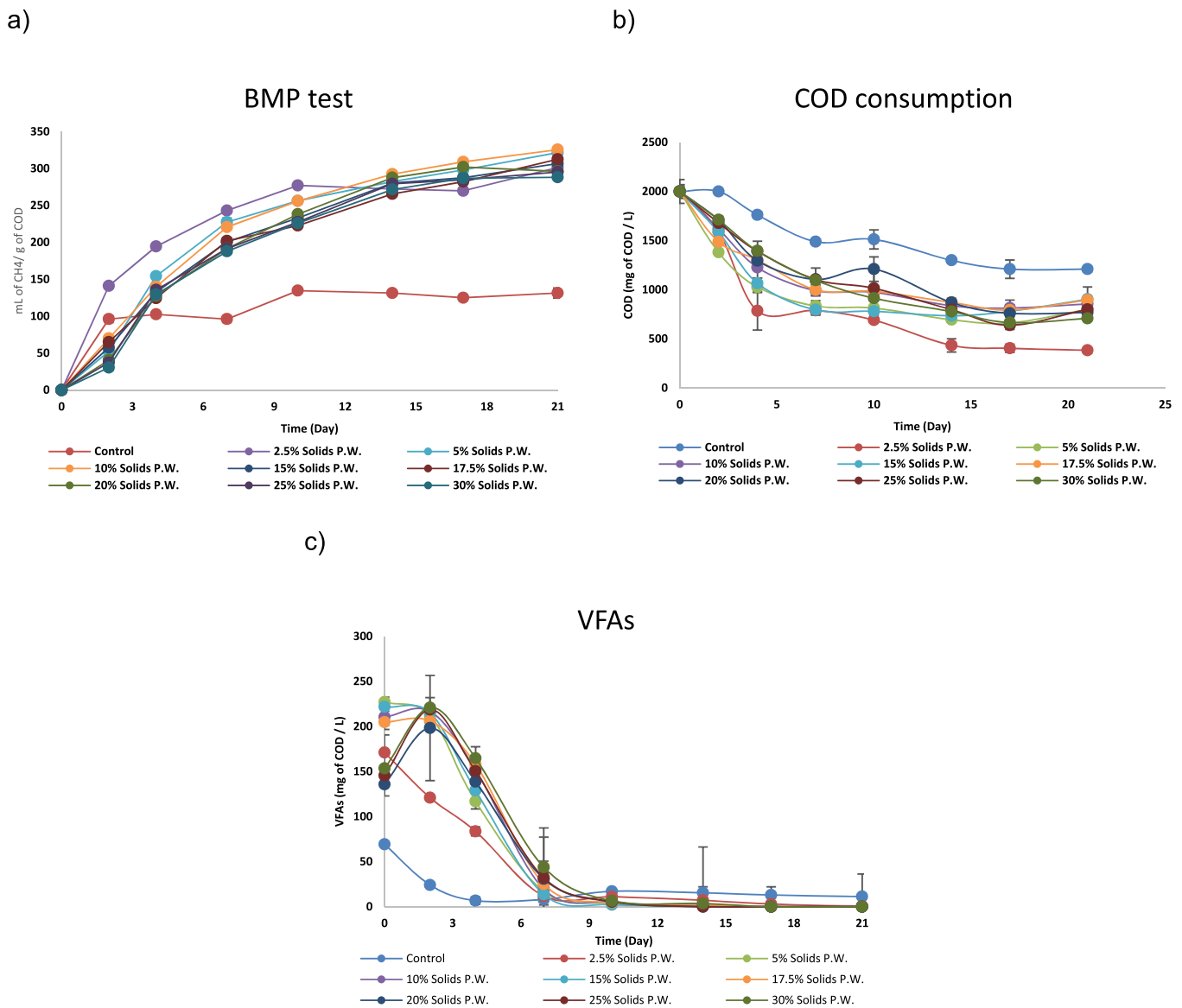


Fig. 7. BMP test results (a) from process waters – Process water 'PW' at different solid loadings and changes in COD (b) and VFA (c) concentration during BMP tests.

**Table 4**  
Comparison of the Experimental BMP v. theoretical BMP.

Sample	BMP <sub>exp</sub> (mL of CH <sub>4</sub> /g of COD added)	BMP <sub>th</sub> Boyle's Eq. (mL of CH <sub>4</sub> /g of COD)	<sup>a</sup> BD Boyle's eq	COD removal	Methane content in Biogas
Control Process Water	134.6	431.5	36%	40%	63%
2.5% P.W.	301.5	337.5	89%	81%	74%
5.0% P.W.	321.7	370.3	87%	60%	77%
10.0% P.W.	325.6	435.4	75%	57%	79%
15.0% P.W.	306.8	360.4	85%	55%	78%
17.5% P.W.	312.7	400.9	78%	55%	79%
20.0% P.W.	302.1	403.1	75%	62%	80%
25.0% P.W.	295.4	351.5	84%	60%	80%
30.0% P.W.	288.2	368.2	78%	62%	80%

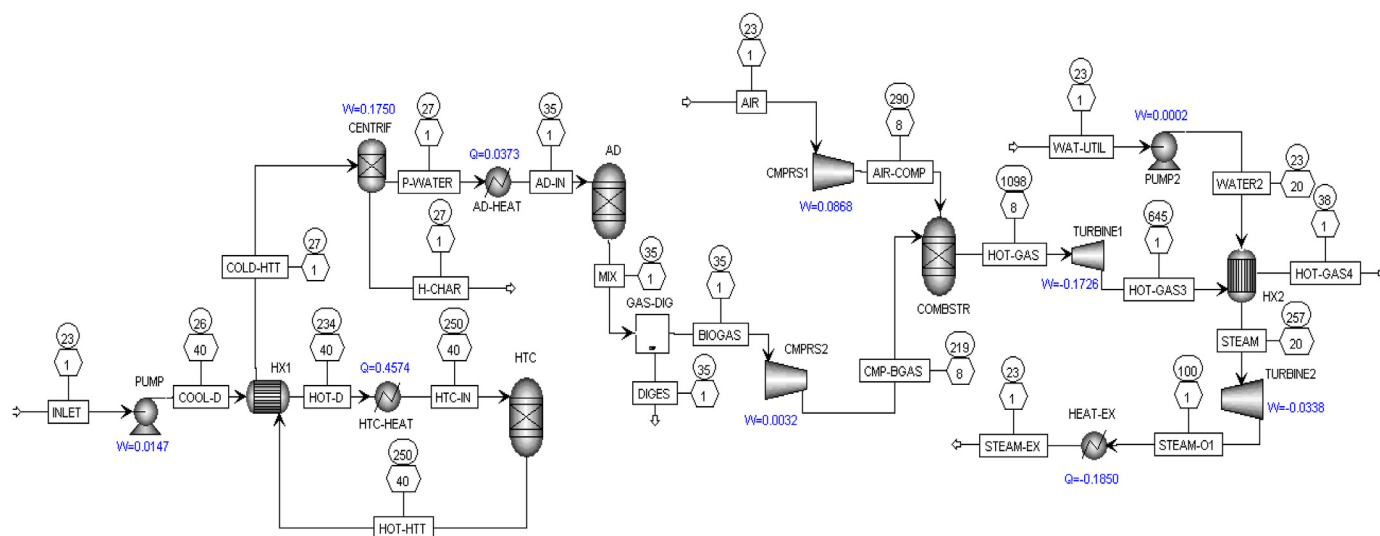
<sup>a</sup> BD: Biodegradability.

biogas from the head space of the digester.

Subsequently, the biogas was ready for processing in a combined cycle gas turbine system. It was first compressed to 8 bar before meeting compressed air (also at 8 bar) in the combustor. The

combustor was simulated with an adiabatic 'RGibbs' block. Excess air was used to maintain a temperature below 1100 °C at varying flowrates depending on the biogas composition.

The hot exhaust gas from the combustor was passed through a



**Fig. 8.** Aspen Plus diagram for the integration of the HTC process at the end of a WWTW with 20% solids loading at 1 kg/hr (Stream temps (°C) found in circular labels, pressures (bar) in hexagonal labels above streams).

turbine which recovered energy as electrical power, based on its expansion from 8 bar to 1 bar. The expanded exhaust gas downstream of the gas turbine, still carrying an abundance of thermal energy was made to exchange heat with compressed water (20 bar) to generate superheated steam. The superheated steam passed through a steam turbine, expanding to 1 bar and generating further electrical power. The remaining steam was cooled to 23 °C in a 'Heater' block in order to determine the thermal power output of the system.

The breakdown of energy consumption and production during CHP processing for the 20% solid loading scenario can be seen in Fig. 8. The cogeneration system running from biogas built in Aspen Plus was found to operate with a HHV electrical efficiency ( $P_{eff,CHP}$ ) of 33.3% and an HHV thermal efficiency of 47.6% ( $Q_{eff,CHP}$ ). Note this took into account the electrical losses experienced at 'CMPRS1', 'CMPRS2' and 'PUMP2' as well as power production from 'TURBINE1' and 'TURBINE2' with heat production from the heat-duty of block 'HEAT-EX'.

Results show the heat duties, power consumptions and power productions of various blocks in the CHP part of the process flow. Sign convention: positive electrical and thermal power represent a production of electricity or heat, negative represent consumptions.

As previously mentioned, it was not possible to model a hydrochar-fuelled CHP system in Aspen Plus due to its representation as an 'unconventional solid' in the flowsheet. Thus, the CHP efficiencies of the coal-fired cogeneration process analysed in Liao et al. [35] (28% electrical and 43% thermal on an LHV-basis) have been used to infer electrical and thermal power production potentials from hydrochar, if they were to be used accordingly. These can be found in Table 5 in the rows 'Hydrochar-based  $P$ ' and 'Hydrochar-based  $Q$ '. The system's net electrical power production ( $P_{net}$ ) was calculated via the summation of the net power production during cogeneration processing of hydrochar and biogas combustion with the power terms from 'PUMP' and 'CENTRIF' during HTC and AD processing. The power consumption for the centrifuge was set at 35 kWh/t, as stated in Huber Technology (2018). The system's net thermal power production ( $Q_{net}$ ) was calculated via the sum of heat terms in 'AD-HEAT' and 'HTC-HEAT' with the net heat produced during cogeneration processing of hydrochar and biogas.

Table 5 also highlights that high solid loading scenarios,

compared to low solids loading scenarios, produce more electricity and heat from hydrochar combustion but less from combustion of biogas generated from the AD of HTC process water. The overall result of this is that scenarios with lower solids loading generate more combined heat and power than higher solid loadings. For example, the total electricity produced from hydrochar and biogas combustion from 2.5% to 30% solids loading are 1.53 kW and 1.36 kW per kg of HTC solids inlet respectively. This occurs because the improved BMP potential from increased flows of process water outweighs the impaired energy densification of hydrochar during lower solids loading HTC.

However, higher solids loading scenarios are undoubtedly more favourable when net system electricity and heat production are calculated. This takes into account the energy requirements for HTC and AD processing. It can be seen in Table 5, that the system energy consumption is far greater at lower solid loading. This is because the total flow-rate experienced at lower-solid loading scenarios is higher, thus requiring more power for pumping and heating. Therefore, under a 2.5% solids loading scenario, more thermal and electrical power are consumed than produced, and a net thermal consumption also occurs under the 5% solids loading scenario. Thus, from an energetic perspective and under these conditions, HTC appears to be only worthwhile when using HTC feedstock with a solids loading of 10% and over.

It is worthy of note that under each scenario, if on-site combustion of hydrochar is not carried out for CHP (represented by the 'hydrochar-based  $Q$ ' and 'hydrochar-based  $W$ ' terms in Table 5), then neither net electrical or net thermal power can be produced. For example, if under the 30% solids loading scenario the electricity and heat produced by the combustion of hydrochar for CHP is omitted from the net system power production calculations, then the process would consume 0.01 kW of electricity and 0.06 kW of heat per kg of solids input for HTC. This only highlights the fact that hydrochar from the HTC of digestate could have an important impact on the sustainability performance of wastewater treatment works.

It is clear that the greater solids loading scenarios provide superior electrical and thermal efficiencies, with the 30% solids scenario exhibiting the highest, with a 25.8% electrical efficiency and a 38.9% thermal efficiency. This provides evidence that the proposed AD + HTC system is not only self-sufficient but can help maximise

**Table 5**  
System energy balance for total solids inlet of 1 kg/h. Positive values of P and Q represent a net production (and negative values of P and Q represent net consumption) of electrical power and heat respectively.

	2.5%	5%	10%	15%	17.5%	20%	25%	30%
Hydrochar-based P (kWh)	1.17	1.20	1.19	1.23	1.22	1.22	1.24	1.24
Hydrochar-based Q (kWh)	1.82	1.86	1.85	1.91	1.89	1.89	1.93	1.93
Biogas-based P (kWh)	0.36	0.29	0.21	0.17	0.15	0.15	0.12	0.12
Biogas-based Q (kWh)	0.52	0.41	0.30	0.24	0.21	0.21	0.17	0.17
'PUMP' P requirement (kWh)	-0.14	-0.07	-0.03	-0.02	-0.02	-0.01	-0.01	-0.01
'CENTRIF' Q requirement (kWh)	-1.40	-0.70	-0.35	-0.23	-0.20	-0.18	-0.14	-0.12
HTC-HEAT Q requirement (kWh)	-5.73	-2.72	-1.21	-0.71	-0.57	-0.46	-0.34	-0.27
AD-HEAT Q requirement (kWh)	-0.29	-0.15	-0.07	-0.05	-0.04	-0.04	0.00	0.00
$P_{net}$ (kWh)	-0.01	0.71	1.02	1.14	1.15	1.17	1.21	1.24
$Q_{net}$ (kWh)	-3.69	-0.60	0.87	1.39	1.50	1.61	1.75	1.83
$\eta_{P,net}$ (%)	-0.32	17.70	26.20	30.00	30.60	31.10	32.10	32.90
$\eta_{Q,net}$ (%)	-82.50	-14.80	22.30	36.50	39.80	42.60	46.60	48.70

the energetic potential of wastewater treatment works. If a 30% solids loading was deemed too high for ease of pumping (risks of deposition and blockages), then a HTC feedstock with 20% solid loading would still provide a comparable electrical efficiency of 24.5% and a thermal efficiency of 33.5% in the proposed AD + HTC system. The data associated with this paper is available from University of Leeds at <https://doi.org/10.5518/819>.

#### 4. Conclusions

Solid loadings had a direct influence on hydrochar composition and its energetic properties. The process waters were also influenced by the solid loading increasing the concentration of carbon, nitrogen and phosphorus compounds, which increased the potential for resource recovery from sewage digestate. With regard to the accumulation of soluble organic matter in the process waters, a significant increment in methane yields was observed when digestate liquor used as a control for BMP tests (131 mL CH<sub>4</sub>/g COD on average) was compared with process waters anaerobically digested (228–301 mL CH<sub>4</sub>/g COD). The coupling of anaerobic digestion with hydrothermal carbonization integrated in a wastewater treatment work showed a significant net electrical and thermal power production when process water and hydrochar were considered as fuel sources. The proposed AD + HTC process was a net energy producer beyond 10% of solid loading when the hydrochars were considered as a fuel source in a CHP system. Although further studies are needed in order to better understand the influencing factors controlling process conditions that lead to improvements in the hydrothermal carbonization of sewage digestate, this research work demonstrates the great potential from combining AD and HTC as an alternative to conventional sludge management systems in wastewater treatment works.

#### CRedit authorship contribution statement

**C.I. Aragón-Briceño:** Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing - original draft. **O. Grasham:** Software, Validation, Formal analysis, Data curation, Writing - review & editing, Visualization. **A.B. Ross:** Resources, Supervision, Writing - review & editing, Project administration, Funding acquisition. **V. Dupont:** Resources, Supervision, Writing - review & editing, Project administration, Funding acquisition. **M.A. Camargo-Valero:** Resources, Supervision, Writing - review & editing, Project administration, Funding acquisition.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.renene.2020.05.021>.

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