ABSTRACT

Sludge from textile effluent treatment plants (ETP) remains a challenge for many industries due to inefficient and limited waste management strategies. This study explores the potential of using anaerobic digestion (AD) to improve the environmental quality of textile ETP sludge. The AD of ETP sludge is affected by the low C/N ratio (3.7), heavy metal content, and toxicity. To improve the process, co-digestion of ETP sludge with different substrates (sewage sludge, cow dung, and sawdust) under mesophilic conditions (37 °C), followed by a thermochemical pretreatment was assessed. The results showed that anaerobic co-digestion of the textile sludge with the co-substrates is effective in reducing pollution load. It was found that organic matters degraded during the 30-day AD process. The chemical oxygen demand and biological oxygen demand reduction was in the range of 33.1–88.5% and 48.1–67.1%, respectively. Also, heavy metal (cadmium, lead, iron, and, mercury) concentration was slightly reduced after digestion. Maximal biogas yield was achieved from co-digestion of textile sludge and sewage sludge at a mixing ratio of 3:1, 1:1, and 1:3, and methane content was respectively 87.9%, 68.9%, and 69.5% of the gas composition. The results from this study show that co-digestion will not only reduce the environmental pollution and health risks from the textile industry but also recover useful energy.

Key words | anaerobic digestion, biogas, sludge management, textile sludge, wastewater treatment plant

HIGHLIGHTS

- The use of anaerobic co-digestion for textile sludge treatment and energy recovery were investigated.
- Textile effluent treatment plant sludge composition and toxicity affects its biodegradability.
- The addition of co-substrates with high C/N to the textile sludge improved the biodegradability and yielded highest biogas volume.
- Anaerobic co-digestion was able to improve the environmental quality of the sludge. A decrease in chemical oxygen demand, biological oxygen demand, and heavy metals concentration was observed.

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doi: 10.2166/wst.2020.360
INTRODUCTION

The management of industrial wastewater remains a major challenge in sub-Saharan Africa. For most countries, only a small fraction of municipal wastewater is being treated through waste stabilization ponds, which comprise a series of ponds stabilizing organic matter through biological processes. However, most of these ponds are not designed to receive industrial wastewater because of their toxicity to important microorganisms in the biological treatment process. As a result, most industries are required to construct their own industrial effluent treatment plants (ETP). For textile industries, this is a challenge because of the complexity of the type of wastewater they produce. The effluent characteristics of textile wastewater often vary with the production demands and type of product used. Textile processing industries often use dyes and bleaching agents that require a huge amount of freshwater, resulting in the production of toxic wastewaters (Jahagirdar et al. 2013). There is no single treatment method capable of efficiently eliminating the pollutants in the wastewater (Lee et al. 2001). Generally, every treatment technique has its advantages and limitations. Various methods including filtration, coagulation, and flocculation techniques only convey organic pollutants as well as inorganic pollutants from one stage to another. However, an adequate treatment technique requires comprehensive and adapted treatment methods, as well as skilled technicians for the operation and maintenance of the treatment system (Jegatheesan et al. 2016). The textile wastewater treatment, through combined traditional treatment methods, such as physical, chemical, or biological, are found to be suitable but very expensive and leads to the production of bio-sludge (Nguyen & Juang 2013). For instance, the use of chemical coagulation and flocculation techniques requires considerable financial means due to the high cost of chemical reagents. Moreover, this technique results in the production of a high volume of sludge, causing handling and discharge issues (Pang & Abdullah 2013). In places where the local authorities’ capacity to enforce environmental regulation is weak, most industries often discharge their effluent untreated into the surrounding lands and water bodies. However, due to increased public awareness on this matter, most African countries have adopted strict environmental pollution control measures and laws to prevent the discharge of untreated wastewater.

In Tanzania, many industries have installed their ETP for treating generated wastewater prior to disposal. For textile industries, the sludge generated from the treatment process may still be toxic for direct discharge into the environment. Textile effluent sludge, a by-product of the treatment process, often contains organic and inorganic matters, including heavy metals such as iron (Fe), copper (Cu), cadmium (Cd), zinc (Zn) and chromium (Cr), etc., which are toxic to both human health and the environment. Previous studies have reported that cow dung (CD; C/N ratio 16–25) and sawdust (C/N ratio 200–500) can both improve the AD process, add nutrients for microorganisms, and enhance the biogas production. A study conducted by Zhen et al. (2017) has revealed that optimum conditions for thermal hydrolysis
was a temperature of 170 °C for a treatment time ranging between 30 and 60 min. Such conditions were found to reduce the retention time by 5 days and increase biogas production. Thermal hydrolysis is known for its advantages of odor removal, sludge quantity, and pathogens reduction. It also contributes to improving sludge dewaterability. Jain et al. (2015) have reported that a pH between 6.5 and 7.5 is optimal for the microbial activities and enhances a better biodegradability of the substrate. However, pH values above this range are known to adversely affect the methanogens.

In this study, we assessed the performance of ETP being used by A to Z Textile Mills Ltd. to treat its agro-textile wastewater in Arusha. Based on the performance assessment, we explore ways to co-digest the final sludge to realize waste reduction and also improve the environmental quality of the industrial process.

Application of AD for industrial wastewater treatment

Several studies have explored various AD technologies for textile wastewater treatment and reuse. Gnanapragasam et al. (2011) conducted batch reactor experiments using the AD process for treating textile dye effluent to remove color and reduce chemical oxygen demand (COD). The setup was composed of a reactor with 5-L capacity, for the combined treatment of synthetic textile dye and starch wastewater at different mixing ratios of 20:80, 30:70, 40:60, 50:50, and 60:40. They reported that the optimum ratio was 30:70 and the percentage reduction of COD and color were 81.0% and 87.3%, respectively. The authors concluded that the results from the batch studies can be used to design a large-scale continuous reactor for the treatment of combined textile dye and starch wastewater under the same conditions.

Punzi et al. (2015) used a setup composed of an anaerobic biofilm reactor followed by ozone treatment and revealed that the anaerobic treatment was able to achieve removal efficiency of 70% for COD (hydraulic retention time (HRT) was 3 days for AD). Treating the effluent using the ozonation method for 6 min was able to further reduce organic matter in the effluents. Toxicity was also found to be 20 times lower than that of the initial level. Moreover, aromatic matters of effluent were reported to be degraded by ozone. It was recommended to use the ozonation method as a post-treatment system after AD because it enhances the reduction of refractory compounds and toxicity.

Lin et al. (2017) investigated the performance of combining granular activated carbon (GAC) adsorption, AD under mesophilic conditions, and microalgae scenedesmus species cultivation. Experiments were conducted using laboratory-scale reactors for textile wastewater treatment, as well as regenerating algae biomass and biogas. The wastewater was pre-treated using the GAC to limit AD process inhibition and the microalgae was used to further treat digester effluent. The combined system was able to produce methane, total hydrogen, and ethanol energy at a rate of 16.9 kJ/(L per d). High pollutants removal efficiencies were also obtained for COD 89.5%, color 92.4%, organic acids 94.7%, and carbohydrates 97.4%. However, the authors have found that reducing COD, volatile fatty acids (VFAs) use, and color removal efficiencies depend on the effluent initial concentrations. Thus, for the treatment of textile dye effluent, a flow rate of 1,000 m³/d is recommended by the authors as a conceptual treatment process. Methane production and biomass are predicted to be 2.07 × 10⁷ kJ/d and 9,800 kg/7 d (7 days), respectively.

The major challenge associated with the anaerobic treatment of textile wastewaters is its toxicity to essential microorganisms. AD is suitable for textile wastewater treatment, but the inhibitory compounds existing in the wastewater and the sludge may lead to failure of the AD process (Kumar & Mudhoo 2013). However, the presence of heavy metals in textile wastewater and sludge has been found to have an antagonistic effect on the overall process of AD (Abdel-shafy & Mansour 2014). Heavy metals removal appears to be necessary for a good performance of the AD process. To efficiently remove metals, it is essential to know metal distribution in sludge. This useful information for removing metals is provided by sequential chemical extraction (SCE), which is widely used to describe the chemical distribution of metals in sludge. The SCE process is aimed at fractionating metals in the sludge sample by using chemical extracting agents. Information about metal forms in sludge samples allows the use of a suitable method for metal removal, generally by solubilization. This method mainly consists of separating the solid phase from the liquid phase, followed by precipitation from the liquid phase (Marchioretto 2005; Du 2015).

Although trace elements such as iron, manganese, and molybdenum are important for the AD process, some metals such as antimony, lead, mercury, arsenic, and uranium are very toxic and their biological role is limited (Kumar & Mudhoo 2015). Various methods are widely applied to improve and avoid failure of the microbial activities during AD treatment textile wastewater. Studies have reported that co-digestion with substrates rich in carbohydrates can enhance biogas yield compared to
mono-digestion of single substrates (Kumar et al. 2020). Substrate decomposition can also be improved through pre-treatment methods such as thermal, chemical, enzymatic, and mechanical (Zhang et al. 2017; Bharathiraja et al. 2018). Overall, pollution level reduction in the AD is dependent on the retention time because the degradation of the organic load is effective for a longer retention time in the digester (Vögeli et al. 2014).

Extensive research on the application of AD for industrial wastewater treatment outcomes has been proven to be satisfactory. However, the secondary pollutant (sludge) from this treatment process remains a concern for textile wastewater post-treatment. There is a lack of systematic studies on textile sludge treatment, especially the application of AD technology. This study was oriented toward the application of AD on textile sludge. Additionally, the process was optimized through co-digestion with substrates such as sewage sludge, sawdust, and CD followed by a thermo-chemical treatment of the mixtures. The developed treatment scheme was tested in the laboratory.

MATERIALS AND METHODS

Case study textile ETP

The study was conducted at the A to Z Textile Mills Ltd. in Arusha, Tanzania. The company has installed its ETP where wastewater from the factory undergoes a traditional treatment process (pre-treatment, primary treatment, secondary and tertiary treatment) (Figure 1). The characteristics of the raw effluent from the ETP was determined as reported in Table 1. Moreover, various textile effluent characteristics were collected from the literature to compare the typical composition of the raw effluents.

The system is composed of an equalization tank, a biological tank, regulation tanks, secondary tanks, a pressure sand filter (PSF), and a final discharge tank:

1) Equalization tank: wastewater from the textile dying process is collected in the equalization tank. A blower is used at this step of the treatment process to inject air into the tank. The addition of air enhances the aeration and complete mixing of the influent wastewater. The pH is checked every 2 h and is maintained between 7.5 and 8 using hydrochloride acid (3.01). A pH lower than 6.5 will prevent the sludge from settling. In extreme cases, caustic is added to maintain the pH.

2) Biological tank: the biological treatment of the wastewater is performed under aerobic conditions. The tank is also aerated and mixed using a blower. Color removal is done using microplus, which is a color removal coagulant (50 g/1,000 L if the color is light and 100 g/1,000 L if the color is dark). For microorganism enhancement, CD urea, and fertilizer such as diammonium phosphate (DAP) are used.

Figure 1 | Flow diagram of textile ETP.
Table 1 | Characteristics of A to Z Textile Mills Ltd. wastewater and typical values

<table>
<thead>
<tr>
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<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>–</td>
<td>9.49 ± 0.4</td>
<td>7.5 ± 0.58</td>
<td>8 ± 0.2</td>
<td>9.5 ± 0.6</td>
<td>8.2 ± 0.1</td>
<td>9.2 ± 0.2</td>
</tr>
<tr>
<td>Temperature</td>
<td>°C</td>
<td>49 ± 5.09</td>
<td>38 ± 3.5</td>
<td>25 ± 4</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Conductivity</td>
<td>mS/cm</td>
<td>8.25 ± 2.60</td>
<td>5.2 ± 0.63</td>
<td>4.056 ± 0.003</td>
<td>4.01 ± 0.32</td>
<td></td>
<td></td>
</tr>
<tr>
<td>TDS</td>
<td>ppm</td>
<td>912.67 ± 55.18</td>
<td>3,567 ± 470</td>
<td>5,116 ± 558</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>COD</td>
<td>mg/L</td>
<td>1,231 ± 43.84</td>
<td>689 ± 48</td>
<td>2,210 ± 3.21</td>
<td>760 ± 102</td>
<td>699 ± 236</td>
<td>832 ± 52</td>
</tr>
<tr>
<td>BOD</td>
<td>mg/L</td>
<td>565 ± 21.21</td>
<td>248 ± 31</td>
<td>1,162.9 ± 4.4</td>
<td>215 ± 50</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cadmium (Cd)</td>
<td>mg/L</td>
<td>6.67 ± 0.89</td>
<td>1.05 ± 0.14</td>
<td>0.0007 ± 0.0</td>
<td></td>
<td></td>
<td>&lt;1</td>
</tr>
<tr>
<td>Chromium (Cr)</td>
<td>mg/L</td>
<td>12.78 ± 0.02</td>
<td>0.19 ± 0.03</td>
<td></td>
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<td></td>
<td>&lt;1</td>
</tr>
<tr>
<td>Copper (Cu)</td>
<td>mg/L</td>
<td>36.29 ± 0.19</td>
<td></td>
<td></td>
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</tr>
<tr>
<td>Lead (Pb)</td>
<td>mg/L</td>
<td>9.58 ± 0.18</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Nickel (Ni)</td>
<td>mg/L</td>
<td>39.88 ± 0.69</td>
<td>1.03 ± 0.05</td>
<td>0.002 ± 0.0</td>
<td></td>
<td></td>
<td>&lt;1</td>
</tr>
<tr>
<td>Zinc (Zn)</td>
<td>mg/L</td>
<td>45.26 ± 16.93</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Iron (Fe)</td>
<td>mg/L</td>
<td>128.60 ± 156.68</td>
<td>2.35 ± 1.1</td>
<td>0.016 ± 0.0</td>
<td>18.04 ± 1.3</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mercury</td>
<td>mg/L</td>
<td>0.27 ± 0.33</td>
<td></td>
<td></td>
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<td></td>
</tr>
</tbody>
</table>

3) Regulation tanks: composed of a chemical dosage tank and HRT regulating tank. Hydrated lime is dosed at this step to enhance the reduction of oxidizable organic pollutants but also for clarification purposes through coagulation and flocculation of particles.

4) Secondary tanks: comprises the primary clarifier and the secondary clarifier. In the secondary clarifier, coagulation and flocculation method is used to enhance decolorization. However, this method has some limitations due to low decolorization efficiency, but also the production of bio-sludge (Holkar et al. 2016). Further color removal is also performed, using microplus at around 10 g/1,000 L for light shade and 20–25 g/1,000 L for a dark shade. Along with this, a polyelectrolyte solution animole 2030 is dosed at around 2 g/1,000 L in the flocculator tank to help in particle bridging and compaction of the sludge. An electrical coagulation machine is used to generate heat that will enhance the color removal from the water. A polymer is also dosed to enhance the flocculation of the sludge. The produced sludge from the secondary clarifier is transferred to the filter press for dewatering purposes and discharge.

5) PSF: the effluent is directed to the PSF for final treatment, and then conveyed in the final discharge tank for reuse.

Approximately 23,400 L (234 m³) of effluent is being treated at A to Z Textile Mills Ltd. daily. One of the major challenges with the current treatment process is the large quantity of sludge produced, which is being dried and stored in bags onsite. Due to its pollution risks, A to Z Textile Mills Ltd. currently does not have a suitable solution for the disposal of the sludge.

The company has also installed a sewage treatment plant (STP) (Figure 2) for the treatment of wastewater generated by households (approximately 8,000 inhabitants) living within the company’s residential houses. The wastewater comes from flushing toilets, bathing, washing sinks, etc.

The STP is composed of the sewage collection tank, the biological tank, the clarifier, the filtering tanks, and the final discharge tank:

1) The sewage collection tank: the raw water is collected in this tank as a first step of the treatment process. A mechanical screen for heavy particles and plastics removal is installed in the collection tank and is cleaned regularly.

2) The biological tank: blowers are installed in this tank to generate air for aeration. Microplus is dosed at around 30 g/1,000 L. In this tank, microorganisms are grown to enhance the biological process. CD and urea are added (molasses bacteria) to enhance the bacteria’s growth, and therefore increase the consumption of organic matter.

3) The clarifier: the wastewater clarification process is operated in this tank. Dosing of aluminum sulfate is done in this step at 25–30 g/1,000 L continuously as per the flow. The addition of aluminum sulfate allows the coagulation and flocculation process and enhances...
the treatment to reduce suspended solids and organic loads from primary clarifiers.

4) The filtering tanks: the system is composed of one activated carbon tank and one PSF tank for clarified water filtration. The objective is to remove the small particles remaining in the clarified water. A maximum of 1 g/1,000 L of sodium hypochlorite is dosed after the filtration for smell removal. The effluent is transferred to the final discharge tank for reuse purposes.

During this study, sludge and water samples were collected at different stages of the treatment units. Wet sludge and dewatered sludge were collected from the final discharge tank and the filter press, respectively (Figure 1). The sewage sludge was collected at the same site from the STP. The sludge was packed and stored in polyethylene bags for various laboratory analyses. Water samples were collected in polyethylene plastic bottles from the inlet and outlet of the treatment plant.

Characterization of the textile industry wastewaters

The physical parameters of the wastewater and sludge samples, namely pH, temperature, total dissolved solids (TDS), and electrical conductivity (EC) were measured onsite and in the laboratory using HANNA Instruments Multiparameter (RI, USA). Selected physicochemical parameters were also measured, according to Standard Methods: Method 5220D for COD and Method 2540G for total solids (TS) and volatile solids (VS), which were dried at 105 and 505 °C, respectively (APHA 1999). For biological oxygen demand (BOD), samples were incubated for 5 days at 20 °C in the incubator (model OxiTop Box) and the C/N ratio was determined using the Thermo Scientific FLASH 2000 HT Elemental Analyzer. Selected heavy metals such as cadmium (Cd), lead (Pb), zinc (Zn), copper (Cu), chromium (Cr), nickel (Ni), iron (Fe), and mercury (Hg) were analyzed using an atomic absorption spectrophotometer (model WFX-210). VFAs were determined using a gas chromatographic method. Pollutants removal efficiency for each sample was determined using the following formula (Equation (1)):

$$\frac{C_i - C_f}{C_i} \times 100 \quad (1)$$

where $C_i$ and $C_f$ represent is the initial and final concentration of pollutants in the samples, respectively.

Design of laboratory-scale batch reactor

A laboratory-scale biogas batch reactor (Figure 3(c)) was assembled and used to perform experiments under mesophilic conditions (37 °C). Temperature control was done using the Memmert water bath method (type ONE 7). The reactor was constructed using an Erlenmeyer flask bottle (0.5 L) connected to a measuring cylinder (1 L). A plastic pipe was used to connect the reactor to the gas collector and parafilm was used to seal the Erlenmeyer flask’s outlet to prevent gas leakages. The water displacement method was used to test the biogas production. The HRT was chosen to be 30 days and the series of experiments lasted for 122 days. Biogas volume was monitored daily and a biogas analyzer (model Biogas 5000, UK) was used to determine the methane content of the gas.
Co-digestion of ETP sludge and STP sludge

To realize waste reduction and improve the environmental quality of the industrial process, a series of co-digestion experiments were conducted using the textile effluent sludge and/or wastewater as a strategy for textile waste management. Co-digestion is known for improving biochemical conditions, therefore increasing the biogas yield (Wang et al. 2014). Selected parameters, including pH, TS, VS, COD, and BOD were analyzed for the different mixtures. In the first experimental setup, sewage sludge was selected as the main substrate to increase the organic content of ETP sludge, but also due to its availability. Karlsson et al. (2014) have reported that sewage sludge is a good base that can enable better nutrient and trace element balances. ETP sludge and STP sludge were mixed at different ratios 4:0, 3:1, 1:1, 1:3, 0:4 by volume, and 100 mL of raw water was added to sludge mixtures to have a total working volume of 500 mL.

Pre-treatment conditions

The second experimental setup explored various ways of optimizing biogas and methane production. Parameters such as pH, substrate type, alkali, and thermal pre-treatment were selected for the optimization. Thus, two series of experiments were set as follow:

1) ETP sludge + STP sludge at different mixing ratios (3:1, 1:1, and 1:3) + 50 g of CD,
2) ETP sludge + STP sludge at different mixing ratios (3:1, 1:1, and 1:3) + 50 g sawdust.

Thermal hydrolysis conditions depend on the treatment temperature and time. In the current study, high thermal hydrolysis of 170 °C was used for about 60 min. A 1-L beaker and a hot plate were used for the heating process. The beaker was covered with aluminum paper to avoid water evaporation. The alkali treatment consisted of adding 3 g/L of sodium hydroxide to the sludge mixtures to maintain the pH between 6.5 and 7.5. The pretreated sludge was then used in the batch digestion described earlier. Nitrogen gas was flushed into the reactors to ensure better anaerobic conditions of the digesters.

Statistical analysis

One-way analysis of variance (ANOVA) in Excel software was used to compare the differences in biogas yield among data obtained from anaerobic co-digestion of ETP sludge with different substrates at different mixing ratios. Statistical significance was set at a p-value <0.05. All the graphs were plotted in Origin software version 9.5.

RESULTS AND DISCUSSION

Textile ETP process performance assessment

The A to Z Textile Mills Ltd. ETP performance was evaluated and the percentage reduction of each parameter is presented in Table 2. The removal efficiency of organic matters such as BOD and COD was 26.74% and 8.18%, respectively. Measured BOD concentration was 550 mg/l, 510 mg/l, and 505 mg/l at the inlet, biological tank, and the outlet, respectively. The COD concentrations were 1,262 mg/l, 1,010 mg/l, and 924.5 mg/l at the inlet, biological tank, and the outlet, respectively. For both COD and BOD, the concentrations were above the Tanzanian Bureau of Standards (TBS) recommendation values for effluent discharge (60 mg/L for COD and 30 mg/L for BOD). Metal concentration was found to be high in the inlet of the ETP and slightly decreased after treatment for chromium, copper, nickel, zinc, and iron. All the
concentrations were also above the TBS guideline values ranging between 0.005 to 5 mg/L.

**ETP sludge and STP sludge characteristics**

The results of ETP sludge and STP sludge characterization are shown in Table 3. The ETP sludge is found to be slightly alkaline, with pH values between 8.4 and 9.6. The VS and TS were 56.5 and 51.5 g/L, respectively. In the STP sludge, VS and TS were 75.5 g/L and 51.35 g/L, respectively. The C/N ratio was found to be higher in STP sludge (10.46–11.27) than in ETP (3.55 to 3.81) and it could balance the C/N ratio of the sludge mixture. BOD values ranged from 850 mg/L to 2,250 mg/L for ETP sludge and from 1,900 mg/L to 4,950 mg/L for STP sludge. COD values ranged from 2,600 mg/L to 3,980 mg/L for ETP sludge and from 2,980 mg/L to 9,867 mg/L for STP sludge. In the ETP sludge, electrical conductivity was higher and ranged between 6,830 and 16,560 $\mu$S/cm. This indicates that ions are present in the sludge (Pandey et al. 2011). The current studies revealed that heavy metals were present in considerable quantities in both ETP and STP sludge. However, metal concentration was higher in ETP sludge and was composed of Fe, Zn, Cd, Cu, Ni and Pb in high concentrations, and Cr, and Hg in low concentrations. ETP sludge characteristics from the current study are comparable to the values found in the literature (Zhan & Poon 2015; Anwar et al. 2018). The ETP sludge characterization studies conducted by Pandey et al. (2011) found that the sludge was mostly composed of Cu (39.81–389.83 mg/kg), Ni (23.72–88.75 mg/kg), Cd (4.25–5.41 mg/kg), total Cr (32.00–316.33 mg/kg), hexavalent chromium (below detection limit), Pb (20.31–52.04 mg/kg), cobalt (Co) (12.12–13.46 mg/kg), and Zn (73.48–386.94 mg/kg).

The sources of heavy metals in the STP sludge are mainly industrial contributions, which represent approximately 50% of the metals load of the sludge. Heavy metals in sewage sludge can emanate from the use of detergents containing trace metals, such as Cd, Cu, and Zn, and leachates from plumbing materials (Dewil et al. 2007).

Several studies have reported that heavy metals from influent are concentrated in the bio-sludge produced during wastewater treatment (Van de Velden et al. 2008). Heavy metals removal from wastewater is performed using chemical precipitation followed by coagulation. This process leads to the accumulation of a huge volume of sludge containing a considerable amount of heavy metals. Heavy metals are usually adsorbed onto sludge flocs through the rapid ion-exchange process. Biopolymers are present in significant concentration in sludge flocs and play a key role of exchangers, leading to the formation of anionic and cationic binding surfaces (Dewil et al. 2007; Ahmed & Ahmaruzzaman 2016).

**Mixed sludge characteristics**

Characteristics of the mixed sludge (ETP sludge: STP sludge) are shown in Table 4. The pH values ranged from 6.68 to 9.49. VS were higher in raw STP sludge than in raw ETP sludge.
Moreover, results showed that VS of the different sludge mixtures were slightly similar to one in the raw ETP sludge and the values ranged from 38.6 to 110.8. VS are indicators of the degradability potential of sludge (Bo et al. 2012).

Biogas volume at different mixing ratios (ETP sludge: STP sludge)

During the study, biogas testing was performed in laboratory-scale reactors, and experiments were conducted for 30 days, under mesophilic (37 °C) conditions. The cumulative and day-wise biogas yield at different mixing ratios (ETP sludge: STP sludge) are presented in Figure 4. Zhen et al. (2018) reported that the mono-digestion of textile sludge is less efficient for biogas production. In the current study, the mono-digestion of ETP sludge was slow and resulted in low biogas production. The produced biogas volume was only 65 mL after 30 days of the experiment. These results confirm that ETP sludge has a poor biodegradability. However, the addition of STP sludge to ETP sludge significantly improved the biogas volume generated, as well as the methane yield compared to the mono-digestion of ETP sludge. High biogas production was achieved at mixing ratios of 3:1, 1:1, and 1:3, and the highest volume was from the ratio of 1:3. The greater the volume of STP sludge, the greater the volume of biogas generated. Biogas yield was enhanced due to the high organic content of STP sludge. The C/N ratio was 3.7 and 10.9 for ETP sludge and STP sludge, respectively. ANOVA analysis of cumulative biogas production at different mixing ratios of ETP sludge to STP sludge showed a significant difference ($p < 0.05$). We can therefore conclude that the digestion of ETP sludge with STP sludge contributed to biogas yield improvement at different mixing ratios.

### Table 3 | Characteristics of raw ETP sludge and STP sludge before digestion

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Units</th>
<th>ETP sludge</th>
<th>STP sludge</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>–</td>
<td>9 ± 0.6</td>
<td>6.5 ± 0.20</td>
</tr>
<tr>
<td>Temperature</td>
<td>°C</td>
<td>31.9 ± 0.78</td>
<td>30.6 ± 2.51</td>
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<tr>
<td>Conductivity</td>
<td>µS/cm</td>
<td>4.5 ± 0.92</td>
<td>10.13 ± 2</td>
</tr>
<tr>
<td>TDS</td>
<td>ppm</td>
<td>1,711.5 ± 2,375.17</td>
<td>6,500 ± 1,322.88</td>
</tr>
<tr>
<td>Total alkalinity</td>
<td>mg/L</td>
<td>246.4 ± 161.49</td>
<td>56.8 ± 4.81</td>
</tr>
<tr>
<td>Ammonia nitrogen</td>
<td>mg/L</td>
<td>85 ± 34.03</td>
<td>188 ± 63.09</td>
</tr>
<tr>
<td>COD</td>
<td>mg/L</td>
<td>3,386.7 ± 710.02</td>
<td>5,655.67 ± 3,691.39</td>
</tr>
<tr>
<td>BOD</td>
<td>mg/L</td>
<td>1,816.7 ± 838.65</td>
<td>2,916.67 ± 1,760.91</td>
</tr>
<tr>
<td>VS</td>
<td>g/l</td>
<td>56.5 ± 7.75</td>
<td>75.5 ± 49.92</td>
</tr>
<tr>
<td>TS</td>
<td>g/l</td>
<td>51.5 ± 15.38</td>
<td>51.35 ± 4.17</td>
</tr>
<tr>
<td>C/N</td>
<td>–</td>
<td>3.7 ± 0.18</td>
<td>10.9 ± 0.57</td>
</tr>
<tr>
<td>Cadmium (Cd)</td>
<td>mg/kg</td>
<td>56.9 ± 7.22</td>
<td>42.34 ± 1.66</td>
</tr>
<tr>
<td>Chromium (Cr)</td>
<td>mg/kg</td>
<td>4.15 ± 0.30</td>
<td>21.31 ± 0.71</td>
</tr>
<tr>
<td>Copper (Cu)</td>
<td>mg/kg</td>
<td>42.16 ± 2.52</td>
<td>16.78 ± 0.53</td>
</tr>
<tr>
<td>Lead (Pb)</td>
<td>mg/kg</td>
<td>31.59 ± 10.92</td>
<td>9.32 ± 0.29</td>
</tr>
<tr>
<td>Nickel (Ni)</td>
<td>mg/kg</td>
<td>48.43 ± 2.52</td>
<td>5.35 ± 0.94</td>
</tr>
<tr>
<td>Zinc (Zn)</td>
<td>mg/kg</td>
<td>111.42 ± 7.79</td>
<td>77.93 ± 2.29</td>
</tr>
<tr>
<td>Iron (Fe)</td>
<td>mg/kg</td>
<td>434.31 ± 590.49</td>
<td>24.17 ± 2.33</td>
</tr>
<tr>
<td>Mercury (Hg)</td>
<td>mg/kg</td>
<td>0.68 ± 0.12</td>
<td>0.04 ± 0.02</td>
</tr>
</tbody>
</table>

### Table 4 | Characteristics of the mixture at different ratios

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Units</th>
<th>Characteristics of the mixture at different ratios (ETP sludge: STP sludge)</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>–</td>
<td>4.0 3.1 1.1 1.3 0.4</td>
</tr>
<tr>
<td>TS</td>
<td>g/L</td>
<td>94.99 94.98 92.7 7.84 6.68</td>
</tr>
<tr>
<td>VS</td>
<td>g/L</td>
<td>68.6 90.6 60 100 48.4</td>
</tr>
<tr>
<td>COD</td>
<td>mg/L</td>
<td>47.8 40 62.2 38.6 110.8</td>
</tr>
<tr>
<td>BOD</td>
<td>mg/L</td>
<td>2,600 1,800 2,800 2,300 2,400</td>
</tr>
<tr>
<td>MCa</td>
<td>%</td>
<td>93.1 89.9 93.3 88.9 94.6</td>
</tr>
</tbody>
</table>

*a Moisture content.*

Moreover, results showed that VS of the different sludge mixtures were slightly similar to one in the raw ETP sludge and the values ranged from 38.6 to 110.8. VS are indicators of the degradability potential of sludge (Bo et al. 2012).
Guha & Mohammad (2015) conducted similar experiments by co-digesting textile ETP sludge with CD, and the results showed that 525 cc (525 mL) of biogas was obtained after 18 days using 1.5 kg of ETP sludge, 200 g of CD and 1 L of sludge liquor. To maintain the pH at 8.5, 1 g of sodium bicarbonate (NaHCO₃) was added. In another experiment, 350 cc (350 mL) biogas was generated after 3 days using 500 g of ETP sludge, 50 g of CD, 1.5 L of sludge liquor, and 4 g of NaHCO₃.

Methane content at different mixing ratios

The methane content of the biogas was analyzed several times for the different mixing ratios during the experiments and the results are shown in Table 5. The results revealed an upward trend in methane production, although mono-digestion of ETP sludge yielded the lowest (39.6%). In the current investigation, there was no methane production during the first stages of AD but this slowly increased progressively during the process. Methane production was effective in all the mixing ratios, except for ETP sludge only (1:0). Oxygen was found in the biogas composition (3.2–20.1%) and can be considered as one of the factors affecting biogas yield. Babel et al. (2009) have reported that high oxygen and heavy metals concentration affect the methanogenesis step during the AD process. Methane production was similar from co-digestion of ETP sludge and STP

Table 5 | Methane content of biogas at different mixing ratios (ETP sludge: STP sludge)

<table>
<thead>
<tr>
<th>Day</th>
<th>Cumulative methane content (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>4:0</td>
</tr>
<tr>
<td>1</td>
<td>0</td>
</tr>
<tr>
<td>5</td>
<td>3.8</td>
</tr>
<tr>
<td>10</td>
<td>39.6</td>
</tr>
<tr>
<td>15</td>
<td>15.8</td>
</tr>
<tr>
<td>20</td>
<td>7</td>
</tr>
<tr>
<td>25</td>
<td>1.3</td>
</tr>
<tr>
<td>30</td>
<td>3.6</td>
</tr>
</tbody>
</table>

Figure 4 | Day-wise biogas volume (a) and cumulative biogas volume (b) at different mixing ratios (ETP sludge: STP sludge).
sludge at ratios of 3:1, 1:1, 1:3, and the highest yields were 87.9%, 68.9%, and 69.5% of the gas composition, respectively. The addition of STP sludge had a positive effect on methane yield.

Variation in methane yield can be justified by the addition of biodegradable fraction from the co-substrate organic matter (STP sludge). Moreover, increased methane yield can be associated with the improvement of the C/N ratio (Grosser et al. 2017).

The methane yield observed during our study is comparable to the findings from the work of Kumar et al. (2020). In their lab-scale study, the authors tested methane production from textile sludge, co-digested with CD in 1:1 ratio under controlled conditions (36 ± 1°C; 30-day HRT). They found that methane yield with CD as the added co-substrate was 244.1 mL/g VS, while mono-digestion of textile sludge did not produce any biogas.

**Inhibitory effects of heavy metals on the AD process**

The poor biodegradability of the ETP sludge can be explained by the sludge composition, which contains toxic heavy metals that probably inhibit the AD process during the experiment. Studies have shown that a higher concentration of heavy metals in sludge samples adversely affects microbial activities (Bassan et al. 2016). In this study, ETP sludge was composed of Cd (56.9 mg/kg), Cr (4.15 mg/kg), Cu (42.16 mg/kg), Pb (31.59 mg/kg), Ni (448.43 mg/kg), Zn (111.42 mg/kg), Fe (434.31 mg/kg), and Hg (0.68 mg/kg). The presence of heavy metals can lead to a decrease in biogas production and an increase in intermediate organic complexes. Heavy metals also affect the AD process through physicochemical reactions. They can form compounds with intermediate AD products and precipitate with carbonate, hydroxide, and sulfide (Kumar & Mudhoo 2013; Paulo et al. 2015).

**Effect of substrate composition and thermochemical pretreatment on biogas yield**

The effect of thermochemical pretreatment on substrate biodegradability was investigated. The mixtures were treated under high thermal temperature (170 °C) for 60 min, followed by the addition of 3 g/L of NaOH to the sludge mixtures to maintain the pH between 6.5 and 7.5. The goal of sludge thermal hydrolysis is to increase methane yield and improve sludge dewaterability, at the lowest HRT possible in the reactor. Stuckey & McCarty (1984) reported that thermal hydrolysis improved biodegradability (based on methane yield) of waste activated sludge (WAS). The combined effects of substrate composition and thermochemical pre-treatment are shown in Figure 5.

The first experiment comprised the co-digestion of ETP sludge and STP sludge and 50 g of CD to increase the
biodegradability of the sludge mixtures. Biogas volume from this experiment was observed to be 550 mL, 600 mL, and 1,650 mL for a mixing ratio of 3:1, 1:1, and 1:3, respectively. In this experiment, the monitoring of biogas production stopped after 3 weeks due to a slow and progressive decrease in production. The decrease in biogas production did not affect the methane production, although the carbon dioxide (CO₂) content of the biogas decreased slowly. Methane yield was efficient, and its production increased exponentially during the first 10 days of the experiments. Biogas is typically composed of methane (55–70%), carbon dioxide (35–40%), hydrogen sulfide (20–20,000 ppm), and other gases such as hydrogen and ammonia, in low concentration (Vögeli et al. 2014). Table 6 describes the biogas composition at different mixing ratios with different feedstocks. For ETP sludge to STP sludge at a mixing ratio of 3:1, methane gas was found to be 55.2% of total gas composition. Furthermore, methane gas was 65.5% and 70.8% respectively for ETP sludge to STP sludge mixing ratio of 1:1 and 1:3, respectively. Biogas from ETP sludge co-digested together with ETP sludge and sawdust provided a better biogas yield. Methane content was 54.6, 67.1, and 71.9, for sludge mixtures at ratios of 3:1, 1:1, and 1:3, respectively.

We can conclude from these two experiments that the addition of a third substrate, as well as the thermochemical pretreatment, was efficient in improving sludge biodegradability and enhanced biogas yield compared to the initial experiment. Moreover, the HRT was reduced to 21 days. However, the cumulative biogas yield from the two experiments did not vary significantly, even though they were higher than the initial experiment. The ANOVA analysis of cumulative biogas production at different mixing ratios of ETP sludge to STP sludge with an addition of a third substrate, as well as the thermochemical pretreatment, was efficient in improving sludge biodegradability and enhanced biogas yield compared to the initial experiment. Moreover, the HRT was reduced to 21 days. However, the cumulative biogas yield from the two experiments did not vary significantly, even though they were higher than the initial experiment.

Similar thermal pretreatment conditions (170 °C for 30 min) applied to WAS revealed that the sludge biodegradability increased (in terms of mL CH₄/g VS added or methane yield) for the pretreated WAS compared to the non-pretreated WAS (Pérez-Elvira et al. 2008; Donoso-Bravo et al. 2011; Pérez-Elvira & Fdz-Polanco 2012). Mottet et al. (2009) also reported that treatment at 170 °C of poorly biodegradable WAS showed a 78% increase in methane yield. Methane production increased from 128 to 228 mL CH₄/g VS added.

The second experiment with sawdust (50 g) as the third substrate was also conducted under the same conditions as the previous one to make a comparative assessment. Biogas was observed during the first 10 days of the experiment and the production described an upward trend. After 21 days, the total biogas volume was 680 mL, 1,130 mL, and 2,020 mL for ETP sludge to STP sludge mixing ratio of 3:1, 1:1, and 1:3, respectively. Biogas from ETP sludge co-digested together with ETP sludge and sawdust provided a better biogas yield. Methane content was 54.6, 67.1, and 71.9, for sludge mixtures at ratios of 3:1, 1:1, and 1:3, respectively.

We can conclude from these two experiments that the addition of a third substrate, as well as the thermochemical pretreatment, was efficient in improving sludge biodegradability and enhanced biogas yield compared to the initial experiment. Moreover, the HRT was reduced to 21 days. However, the cumulative biogas yield from the two experiments did not vary significantly, even though they were higher than the initial experiment. The ANOVA analysis of cumulative biogas production at different mixing ratios of ETP sludge to STP sludge with an addition of a third substrate did not show a significant difference for the two experiments (p > 0.05). The CD and the sawdust can both be considered as good feedstock for biogas yield enhancement.

### Table 6 | Biogas composition at different mixing ratios with different feedstocks

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Formula</th>
<th>Unit</th>
<th>ETP sludge + STP sludge + CD</th>
<th>ETP sludge + STP sludge + sawdust</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>3:1</td>
<td>1:1</td>
</tr>
<tr>
<td>Biogas volume</td>
<td>–</td>
<td>mL</td>
<td>550</td>
<td>600</td>
</tr>
<tr>
<td>Methane</td>
<td>CH₄</td>
<td>%</td>
<td>55.2</td>
<td>65.5</td>
</tr>
<tr>
<td>Carbone dioxide</td>
<td>CO₂</td>
<td>%</td>
<td>26.6</td>
<td>32.5</td>
</tr>
<tr>
<td>Oxygen</td>
<td>O₂</td>
<td>%</td>
<td>8.1</td>
<td>8.8</td>
</tr>
<tr>
<td>Ammoniac</td>
<td>NH₃</td>
<td>ppm</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>Hydrogen sulfide</td>
<td>H₂S</td>
<td>ppm</td>
<td>188</td>
<td>599</td>
</tr>
</tbody>
</table>

Pollution level in the digested sludge

Digested sludges were characterized after the biogas production to determine and assess the pollution level, but also the pollutant reduction efficiency. Selected parameters such as COD, BOD, and heavy metals were analyzed after AD of sludge mixtures following the standards methods and the results are shown in Table 7. In this study, VS was also found to decrease after biogas production. The effectiveness of the AD process relies on VS removal efficiency (Grosser et al. 2017). The concentration of COD and BOD was reduced in the digested sludge. The values ranged from 300 mg/L to 1,100 mg/L and 280 mg/L to 700 mg/L for COD and BOD, respectively. Previous studies found that the AD process was effective in reducing COD and BOD (Jain et al. 2015) as a result of organic matter decomposition by microorganisms (Isni et al. 2015).
Figure 6 presents the percentage reduction of organic and inorganic pollutants in the digested sludge. The results revealed a decrease in heavy metal concentration after biogas production, which may be due to precipitation or a dilution effect. According to the initial concentration of metals presented in Table 7, only Cd, Pb, Fe, and Hg were sufficiently reduced and the percentage reduction ranged from 47.2%–74.3%, 18.1%–55.3%, 31.6%–96.1%, and 14.5%–69.7% for Cd, Pb, Fe, and Hg, respectively. Heavy metals are present in sludge in many forms (Dewil et al. 2013): bound to organic matter, bound to manganese oxides and iron, bound to carbonates, and exchangeable. Soluble metals are released in an oxidizing environment due to organic fraction degradation, whereas the residual
fraction does not dissolve but rather keeps metals in its crystal structure. In an anoxic environment, the iron and manganese oxides are thermodynamically unstable and the carbonate fraction is pH sensitive. The water ionic composition, sorption, and desorption processes probably affect the exchangeable fraction.

CONCLUSION

This study explored the potential of reducing environmental pollution from industrial wastewater sludge through biogas production. The study used effluent sludge from a large industry in Arusha, Tanzania. Results show that biogas production from textile wastewater sludge is very low and is probably affected by the high concentration of heavy metals found in the dye compounds used by the industry. However, biogas production can be improved through co-digestion with domestic wastewater sludge, CD, or sawdust. Moreover, thermochemical treatment of the sludge before digestion was found to be effective in improving biogas yield, as well as methane content of the gas. The pollution level in the residual sludge after biogas production was also found to be lower, confirming that AD is probably a good technique for sludge treatment and waste management strategies for textile processing industries. It is possible to co-digest the industrial wastewater through the AD process, which will not only reduce the environmental pollution and health risks from the industries but also recover useful energy. We conclude from this study that the co-digestion of textile industries wastewater and/or sludge is a suitable approach for achieving waste-reduction and improving the quality of final wastewater plant effluent and sludge. Although the current study provided useful methods to enhance waste-reduction and improve environmental quality, it was limited to laboratory-scale experiments. There is a need to test the current findings using a pilot-scale and to conduct long-term experiments.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

REFERENCES


Grosser, A., Neczaj, E., Singh, B. R., Almás, Á. R., Brattebo, H. & Kacprzak, M. 2017 Anaerobic digestion of sewage sludge with


Marchioretto, M. M. 2005 Heavy Metals Removal From Anaerobically Digested Sludge. Wageningen University, Wageningen.


First received 10 April 2020; accepted in revised form 21 July 2020. Available online 3 August 2020