Remediation of sedimented fiber originating from pulp and paper industry: laboratory scale anaerobic reactor studies and ideas of scaling up

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Abstract

Anaerobic treatment of sedimented fibers collected from bottom of a bay that had been receiving pulp and paper mill wastewater for about 70 years were studied for the first time in semi-continuously fed continuously stirred tank reactors (CSTR). Anaerobic treatment of the fiber sediment was shown to be feasible, without dilution and with nitrogen and buffer supplement, at organic loading rates (OLR) up to 2.5 kg VS/m^{3}d and hydraulic retention times (HRT) of 60 d resulting in methane yields of 201 ± 18 L CH_4/kg VS. Co-digestion of sedimented fiber with sewage sludge at an OLR of 1.5 kg VS/m^{3}d and HRT of 20 d resulted in a methane production of 246 ± 10 L CH_4/kg VS. The techno-economic feasibility of mono and co-digestion process together with several case dependent factors such as maximum operable OLR, digestate utilization needs to be evaluated before making further conclusions for larger scale remediation applications.

Keywords: Anaerobic digestion, co-digestion, CSTR, methane, pulp and paper industry, sedimented fiber

1. Introduction

Wood utilizing pulp and paper industries have been an important part of the economy in many countries. These industries have been one of the prime consumers of fresh water and at the same time, they list among the top producers of both solid and liquid waste (Ashrafi et al., 2015). Due to the necessity of huge volumes of water for the industry, often they are located near water bodies. Discharge of pulp and paper mill wastewater to the lakes and seas before the implementation of wastewater treatment processes, resulted in discharge and accumulation of solids in the sediments of the water bodies. These solids in general consist of wood fibers (cellulose, hemicellulose and lignin), papermaking fillers like kaolin and calcium carbonate, pitch, lignin by-products, ash, heavy metals, organochlorine compounds, and resin acids (Hoffman et al., 2017; Kähkönen et al., 1998; Leppänen and Oikari, 1999). Today, such fiber-rich sediments originating from past activities of pulp and paper industry can be found in various locations worldwide, including Nordic countries, Canada and China (Guo et al., 2016; Jackson, 2016). For example, the bay area near an old pulp mill at Hiedanranta, in the city of Tampere in Finland received effluents from a sulfite pulp mill from 1910s to 1980s and has recently been estimated to have about 1.5 million m^3 of sedimented fiber that forms a layer up to 10 m height (Kokko et al., 2018). Sedimented fibers can create serious environmental impacts such as oxygen depletion, release of detrimental compounds from the sediment, heavy metal accumulation and toxicity towards aquatic organisms (Guo et al., 2016; Hoffman et al., 2017; Kähkönen et al., 1998; Leppänen and Oikari, 1999).

Though there have been a few studies to characterize sediments accumulated over time in the water bodies discharged from pulp and paper industries (Guo et al., 2016; Hoffman et al., 2017; Kähkönen et al., 1998), very few studies exist that deals with remediation of these sediments (Kokko et al., 2018). Sediments contaminated by long-term industrial activities often require costly remediation, dredging, and/or disposal (Hoffman et al., 2017). The contaminated sediments in Hiedanranta, Tampere will be remediated with the goal of returning the contaminated water bodies to their pre-industrialization state so that the lake can be used for recreational purpose and at the same time the land area surrounding the bay can be put to residential use.

Anaerobic digestion (AD) is traditionally used to stabilize different types of municipal sewage sludge with simultaneous production of methane. There are several published laboratory studies, using batch assays and/or semi continuously fed reactors, on AD of primary and secondary sludge generated in activated sludge treatment plants of pulp and paper industries (Bayr and Rintala, 2012; Karlsson et al., 2011; Meyer and Edwards, 2014; Sawatdeenarunat et al., 2015; Veluchamy and Kalamdhad, 2017a). Several studies have also focused on biogas production from pretreated pulp and paper sludge (Lin et al., 2017, 2009; Veluchamy and Kalamdhad, 2017b) and on co-digestion with other substrates (Lin et al., 2013, 2011). Previous experimental studies indicate that the AD of biosludge from pulp and paper mills, without any pretreatment, varies widely, with volatile solids (VS) degradation rates of 21–55% and specific methane yields ranging between 40 and 200 mL g^{-1} VS (values are per added VS unless otherwise stated) (Karlsson et al., 2011; Meyer and Edwards, 2014; Veluchamy and Kalamdhad, 2017a). In batch experiments, methane potentials of 210 L CH_4/kg VS were reported for primary pulp and paper industry sludge (Bayr and Rintala, 2012). While primary sludge from pulp and paper mill consists of wood fibers (cellulose, hemicellulose and lignin), papermaking fillers like kaolin and calcium carbonate, pitch, lignin by-products and ash, secondary sludge would majorly consist of microbial biomass. Thus, the sedimented fiber is expected to be more comparable to primary sludge,however the impacts of long term exposure to conditions prevailing in boreal sediments on fiber characteristics are not known. The only published AD study on the fiber sediments reported average methane yields of 250 L CH_4/kg VS in batch experiments (Kokko et al., 2018), which is in the same range or even higher than methane production from primary or secondary paper mill sludge. The study of Kokko et al. (2018) also showed high methane yields for the solid fiber fraction (270 L CH_4/kg VS) as well as reasonably high methane yield for the liquid fraction of the fibers (240 L CH_4/kg chemical oxygen demand (COD)).

Due to the high methane potential of the sedimented pulp mill fibers (Kokko et al., 2018), its stabilization and methane production using AD appears interesting. Considering the complex nature and the huge volume of sedimented fibers to be treated, research effort is needed to evaluate the feasibility of anaerobic treatment of sedimented fibers in reactors, required capacity of the reactors and time for remediation. The optimization of the treatment rate of sedimented fibers can be achieved through a decrease in the hydraulic retention time (HRT) and increase of the organic loading rate (OLR) (Nges and Liu, 2010). However, increasing OLR can lead to a process imbalance, accumulation of acids and a decrease in methane production, if the retention times are not sufficient for microbial growth (Regueiro et al., 2015). With laboratory-scale experiments, the optimization of the digesters at full scale can be studied, without jeopardizing the actual full-scale process and plant economics.

For remediation of the sedimented fibers, within a limited time span, existing AD capacity, which is currently being used for digestion of sewage sludge, would reduce the investment costs for building new digesters as was used initially in early 1990s for AD of municipal biowaste. As the sedimented fibers in Hiedanranta (Tampere, Finland) were found to contain low concentrations of nitrogen (below 6 g/kg total solids (TS)) (Lindroos et al., 2017), co-digestion of the sedimented fibers with another waste rich in nitrogen and alkalinity, like sewage sludge, can furthermore be an optimal option (Syaichurrozi, 2017). In addition, co-digestion can enhance the methane volume, but at the same time it might alter the digestate quality depending on the substrates and thus affecting further treatability and reusability (Budych-Gorzna et al., 2016; Tsapekos et al., 2017).

The objective of the present study was to assess the feasibility of AD of sedimented fiber in laboratory scale completely stirred tank reactors (CSTR) under four different conditions in order to provide design parameters for planning the full-scale remediation. The fiber sludge was studied as such, which was considered the simplest solution for remediation. In order to assess the potential inhibition of the fiber sediments, diluted fiber sediments were studied in two of the reactors with (feed pH adjusted to 7) and without pH adjustment (pH between 4 and 5). Furthermore, co-digestion of the fiber sediment with sewage sludge was studied as it was speculated that some digestion capacity would be available in local sewage digesters. It was also hypothized that in co-digestion, sewage sludge can act as a source of buffer, trace elements and nutrients and hence for future scale up co-digestion with sewage sludge could be considered. Based on the ultimate goal of remediating the bay area this study is a continuation of a previous study by Kokko et al., (2018), where the methane production potential of the sedimented fibers was determined in batch assays.

2. Materials and methods

2.1. Sediment fiber samples, reactor feeds and inoculum

The sediment samples were obtained from the bottom of a bay in Lake Näsijärvi (Tampere, Finland) located near an old pulp and paper industry (Kokko et al. 2018). Sedimented fiber samples were collected in May 2016 using an excavator bucket from a sampling ferry (Ramboll Finland Oy) from three different sampling points and at three different depths (between 0 and 6 m, depending on the sediment height). The samples were stored anaerobically in sealed plastic buckets at 6 °C for about 4 months. Subsequently when starting the reactor studies, 10 L of each of the eight samples were mixed and homogenized with a concrete mixer attached to a power drill. The mixture was stored at 6 ºC until used in feed preparation. After mixing, the density of the sedimented fiber was 1200 kg/m^3. The pH of the mixed sedimented fiber was low, between 4-5, and TS and VS contents were 13.3 ± 0.8% and 12.6 ± 0.8%, respectively (Table 1).

Municipal sewage sludge (thickened combined primary and secondary sludge, Table 1) from Viinikanlahti wastewater treatment plant (Tampere, Finland) was used in co-digestion experiments (R4). The inoculum was digestate from mesophilic anaerobic digester treating the sewage sludge from the same wastewater treatment plant (Table 1). The sewage sludge and inoculum were used within two weeks of collection.

Trace elements and nitrogen were added to the reactors fed with fibers (R1, R2 and R3), weekly from days 53 and 60 onwards, respectively. 10 mL of 40 g NH_4^+/L solution was added to the reactors (liquid volume 5-5.3 L) each week. 5 mL of the trace element solution was added weekly and it contained (g/L): 0.05 $CoCl_2{\cdot}6H_2O$, 0.1 $Na_2SeO_3{\cdot}5H_2O$, 0.05 $Na_2WO_4{\cdot}2H_2O$, 0.05 $(NH_4)_6Mo_7O_{24}{\cdot}4H_2O$, 0.092 $NiCl_2{\cdot}6H_2O$, 2.0 $FeCl_2{\cdot}4H_2O$, 0.05 H_3BO_3, 0.05 $ZnCl_2$, 0.038 $CuCl_2{\cdot}2H_2O$, and 0.05 $MnCl_2{\cdot}4H_2O$ (modified from Angelidaki and Sanders, (2004)). Buffer was added in the form of $NaHCO_3$ (3 - 4 g/L reactor liquid volume) on day 77 in R1 and day 63 in R2 and R3.

Table 1

2.2. Experimental set up

AD studies were done in four CSTR with liquid volumes of 5.0 - 5.3 L (Fig. 1). The reactor contents were mixed with a mechanical mixer that was on/off at 15 rpm for 30 min at a time. The gas generated during the AD were collected in 10 L aluminum gas bags (Supel™ Inert Foil Gas Sampling Bags, Supelco, USA). The reactors were run in fed-batch mode and feeding was done five times a week from the top of the reactor with a tube going under the liquid level (Fig. 1), after the digestate was removed from the bottom of the reactor. Due to some technical problems in reactor operation, feeding as stopped for some individual days (shown in section 3.1). The reactors were operated at 37 ± 1°C and the heating was realized with a water jacket.

Three of the four CSTRs were fed with sedimented fiber as such (R1) or diluted (R2, and R3) (Table 2). The first reactor (R1) was fed with sedimented fiber with a longer HRT of 60 d to have an OLR of 2.5 kg VS/m^{3}d. The second (R2) and third (R3) reactors were fed with sedimented fiber that was diluted with tap water to an OLR of 1.5 kg VS/m^{3}d with an HRT of 30 d. In R3, the pH of the feed was adjusted from 4 to 5 to around 7.0 with 1 M NaOH. The feed of the fourth reactor, used for co-digestion (R4), was a mixture of sedimented fiber (25% by VS) and sewage sludge (75% by VS) with a mass ratio of 1:15, which resulted in a carbon and nitrogen ratio (VS:N) in the range of 18-22. The co-digestion reactor (R4) was initially operated with HRT of 30 d at initial OLR of 1.0 kgVS/m^{3}d. On day 45, OLR was increased to 1.5 kgVS/m^{3}d by decreasing the HRT to 20 d.

Figure 1

Table 2

2.3. Sampling, analyses and calculations

The TS and VS content of the feed materials were analyzed every alternate week to account for the change in solids content due to storage. From the reactor digestates, pH was analyzed every weekday, soluble COD (sCOD) and volatile fatty acids (VFA) were analyzed twice a week, and TS and VS were analyzed every alternate week. Biogas volume and methane and carbon dioxide content in the biogas were measured 3-5 times a week.

TS and VS were analyzed according to standards SFS-EN 14346 and SFS-EN 15169, respectively. Digestate pH was measured with WTW ProfiLine pH 3210 meter and SenTix 41 electrode. sCOD and VFA were analyzed after filtration (0.45 µm, Chromafil Xtra PET) according to standard SFS 5504 and protocol presented by Kokko et al., (2018), respectively.

Methane content in the produced biogas was measured with Shimadzu GC-2014 gas chromatograph with a thermal conductivity detector (TCD) and Porapak N80-100 mesh column. Detector and injector temperatures were 110 °C and oven temperature was 80 °C. Carrier gas was nitrogen with a flow rate of 20 mL/min. The gas volume in the gas bags was measured using water replacement method. Air temperature and pressure were monitored throughout the experiment and methane production results were converted to STP conditions (0 °C, 1 bar).

The total kjeldahl nitrogen in the samples was analyzed with Kjeldahl nitrogen method with the instructions from FOSS. NH_4^+-N was measured with an ammonium electrode (Orion 9512HPBNWP) from the liquid phase separated by centrifugation (4000 rpm, 17 min). PO_4^{3-}-P was analyzed with Hach Lange kits (LCK349) according to the instructions.

The OLR and HRT was calculated for seven days, although the reactors were fed on five working days a week except three individual days (Fig. 2), hence the actual OLR during the weekdays was 1.4 times the average while the HRT was shorter. After addition of nitrogen VS:N ratio was calculated on the weekly average of VS and nitrogen added. Theoretical calculations were made to get an idea of total time and reactor volumes or number of reactors needed to treat the entire volume of the sedimented fiber using mono-digestion or co-digestion at different OLR. Single reactor volume of 5000 m^3 was assumed to make the calculations.

3. Results and discussion

3.1. Overall performance of the reactors

The operational pH during reactor operation and average weekly methane yield along with methane content is shown in Fig. 2, and digestate sCOD and VFA in Fig. 3. The summary of overall reactor performance is reported in Table 3. The pH (Fig. 2B) of the digestate in the reactor treating sedimented fiber as such without dilution (R1) showed a decreasing trend from an initial value above 7 to 6.3 during the first 50 days of operation. Most of the time the pH was 0.1 - 0.3 units lower in reactor treating diluted fiber without pH adjustment (R2) than in reactors treating non-diluted fiber (R1) and pH adjusted diluted fiber (R3). When the pH of the digestate dropped below 6 around day 63 in reactor fed with diluted fiber (R2 and R3), buffer was added to both the reactors in the form of $NaHCO_3$ (3 - 4 g/L reactor volume), which increased the pH up to around 6.7. Buffer was added to R1 on day 77 when its pH dropped below 6, which again brought the pH to around 7. The pH was around 7.1 for the co-digestion experiment (R4) during the whole operation time showing the buffering ability of sewage sludge co-digestion in AD systems (Sosnowski et al., 2008).

Figure 2

The daily methane production typically followed the weekly feeding cycle with slightly increasing yield along the weekdays (data not shown). Similarly, methane content followed weekly feeding cycle in the mono-digestion reactors (R1, R2 and R3) ranging from 45 to 55%, while methane content in the co-digestion reactor (R4) varied less and was around 61% (Fig. 2C). The weekly average methane yield after one month of operation was around 160 L/kg VS for the reactors fed with sedimented fiber (Fig. 2B). The methane yields were observed to follow a decreasing trend with time and reached below 130 L/kg VS for the non-diluted fiber (R1) on day 70 and below 100 L/kg VS for the diluted fiber reactors (R2 and R3) on day 53. Weekly trace element addition from day 53 did not have any impact on methane yield. The ammonium nitrogen content was less than 10 mg/l in all the fiber treating reactors, as measured on day 58, and subsequently weekly nitrogen addition was started on day 60, after which pH dropped below 6 and VFA accumulated and reached up to almost 1 g/L. After addition of buffer on day 63, the methane yield improved again in the two reactors fed with diluted fiber (R2 and R3) and regained the former values of around 170 L/kg VS. Similar effect of weekly buffer addition from day 77 in the reactor treated non-diluted fiber (R1) could be observed where the methane yield increased to around 200 L/kg VS. On contrary to the mono-digestion, co-digestion showed less fluctuating methane yield during the whole run ranging from 230 to 270 L/kg VS without clear impact of the OLR (1.0 - 1.5 kgVS/m^{3}d) nor HRT (30 and 20 d). Low nitrogen content in the fiber was limiting biodegradation of the organic matter present in the first 60 days of operation, i.e. before addition of nitrogen (Table 3).

In the beginning of the reactor operations, VFAs of the digestates were below detection limit except for a short period in the reactor treating diluted and pH adjusted fiber (R3). Some VFAs started to accumulate in the mono-digestion reactors R2 and R3 after initiation of weekly nitrogen addition (day 60) (Fig. 3), probably due to enhanced rate of hydrolysis and/or acidogenesis of complex organic matter followed by organic acid production. Although nitrogen addition immediately improved hydrolysis, the effects on methanogenesis followed after three days. Accumulation of VFA was also accompanied by dropping of pH (below 6) in the reactors. Then methane production started and VFAs were consumed with weekly buffer addition after day 63. No VFAs could be detected in the digestate of co-digestion reactor (R4) throughout the study. The sCOD in the digestates after 1-2 months operation, when VFAs were below detection limit, was highest in reactor fed with non-diluted fibers (R1) (sCOD, 1.04 ± 0.27 g/L) as compared to the ones fed with diluted fibers with digestate sCODs 0.7 ± 0.3 g/L and 0.5 ± 0.3 g/L for R2 and R3, respectively). Due to the dilution of the feed the sCOD values of digestate from R2 and R3 were around the expected values of almost half than that of R1. The co-digestion digestate (R4) had similar or slightly higher sCOD (0.7 ± 0.1 g/L) than the reactors with diluted fiber suggesting higher non degradable sCOD of the sewage sludge (Fig. 3). The digestate sCODs increased in parallel when VFA was detected in the digestates.

Figure 3

Maximum VS destruction of 54 ± 7% could be attained with non-diluted sedimented fiber (R1) at an OLR of 2.5 kg VS/m^3.d and HRT of 60 d (Table 3). VS destruction was around 45% with diluted sediments (R2, R3), suggesting that similar or even higher VS destruction is feasible with non-diluted fiber than with diluted one. The VS destruction was lowest in co-digestion experiments (31%) operated at an OLR of 1 kg VS/m^3.d possibly due to low biodegradability of microbial biomass present in sewage sludge (Appels et al., 2008), which formed the bulk (75% VS) of the substrate in R4.

The digestates from mono-digestion of sedimented fiber as such (R1) and co-digestation process (R4) had 700 - 900 mg/L nitrogen of which ammonium contributed more than 70 to 95%, respectively. Phosphate concentration of the two digestates was

160 - 170 mg/L, of which less than 4 mg/L was in soluble fraction. The fiber sediment had originally low nitrogen content (240 mg/L of N) and thus its value as fertilizer in agriculture directly is apparently limited. While sewage sludge would have heavy metal and organic trace contaminants in its digestate, the sedimented fiber would contain traces of chemicals originating from the pulping process (Lindroos et al., 2017). Hence, while considering the use of the digestates from these processes due attention to these different contaminants should be given.

Table 3

3.2. Performance summary and comparison to other studies

As there are no previous studies available on the anaerobic digestion of sedimented fiber originating from pulp and paper mill in long-term reactor experiments, the results of the present study are compared to results from the anaerobic digestion of primary pulp and paper mill sludge as well as with mixed sludge. Methane yields of the sedimented fiber as such were slightly lower (201 L/kg VS) than that reported by Bayr and Rintala, (2012) using primary sludge (240 L/kg VS) (Table 3). During the reactor runs, the maximum weekly average methane yield of 223 L/kg VS (day 43 to 49) was obtained from the non-diluted fiber reactor (R1) after addition of buffer. This methane yield is slightly lesser than the methane yield of 250 ± 80 L CH_4/kg VS of the sedimented fibers previously obtained in batch assays by Kokko et al., (2018). The present results suggest that for sedimented fiber, OLRs of 2.5 kg VS/m^3d with pH control and nitrogen supplement can result in a methane yield close to the batch studies reported by Kokko et al., (2018). However, to sustain this methane yield regular control of pH and addition of nutrients, especially nitrogen, is necessary as observed from the results of the present study. Maximum weekly average methane production was 201

L/kg VS and 179 L/kg VS for the reactors fed with non-diluted and diluted sedimented fiber, respectively.

The highest methane yield of 280 L/kg VS (average 262 ± 19 L/kg VS) at an OLR of 1 kg VS/m^3.day in R4 (co-digestion with sewage sludge) indicates the possibility of co-digesting sedimented fiber with sewage sludge. Co-digestion with a complementary substrate like sewage sludge has been reported to improve methane yield from kitchen waste (De Vrieze et al., 2015; Ratanatamskul et al., 2014), fruit waste (Fonoll et al., 2015), fatty waste (Li et al., 2015; Tandukar and Pavlostathis, 2015) etc. The major reasons speculated by researchers for improvement in methane production while using sewage sludge as a co-substrate could be dilution and buffering ability, presence of micro-nutrients and constant source of an inoculum (De Vrieze et al., 2015). Higher methane yield in R1 with higher OLRs rules out the role of sewage sludge as a diluting agent. Also, addition of trace elements, nitrogen and pH control in the reactors with sedimented fiber only (R1, R2 and R3) could not result in such high methane yield as R4 with sewage sludge as co-substrate. Lower VS destruction coupled with higher methane production indicates presence of high methane containing compounds, probably fatty and greasy materials in the readily biodegradable material of sewage sludge (De Vrieze et al., 2015). However, with co-digestion there is a lot more digestate in the end that has to be dealt with and the digestate is totally different from the digestate from AD of sewage sludge or sedimented fiber only.

VFAs are intermediary products formed during the fermentation of complex organic materials in the acidogenesis stage of anaerobic digestion. Under typical operation in anaerobic digestion, VFA accumulation beyond 50 to 250 mg/L as acetic acid resulting from some type of microbial species imbalance, caused by factors such as overloading, toxicity and nutrient deficiency leads to making acetogenesis and methanogenesis a rate limiting step (Chatterjee et al., 2017; Mussoline et al., 2012). Accumulation of VFAs in the reactors after nitrogen supplementation indicates that nitrogen addition improved the hydrolysis and/or acidogenesis step. This resulted in lowering of pH below 6, due to low buffering ability of the sedimented fiber. Future experiments designed to study the role of pH adjustment as well as addition of ammonium and trace compounds could be useful to determine the loading potential and long term operation of the process.

Pulp and paper mill sludge contains high amount of lignin, cellulose and hemicellulose, of which cellulose is reported to be degraded well by anaerobic micro-organisms (Bayr and Rintala, 2012). The previous results suggest a relatively high biodegradability of the sedimented fibers with VS removal of 61 - 65% (Kokko et al., 2018) which is also reflected in the CSTRs operated with non-diluted fiber in this study with a VS removal of around 54%. For pulp and paper mill sludge VS removal in the range of 13 to 40% has been reported previously (Bayr and Rintala, 2012; Veluchamy and Kalamdhad, 2017a).

3.3. Scaling up the digesters

In this study, anaerobic treatment of sedimented fibers collected from bay that had received pulp and paper mill wastewater for decades was shown to be feasible in semi-continuous anaerobic digesters (CSTR). Based on the results (R1) it is assumed that OLR of 2.5 kg VS/m^{3}d and HRT of 60 d would be feasible, even though the run lasted only 1.5 HRTs, as the highest weekly average methane yield of 223 L CH_4/kg VS was obtained with this feedstock. It can be speculated that for sedimented fiber, OLRs above 2.5 kg VS/m^3.d would be feasible with a shorter HRT and nutrient and buffer addition, because no process imbalance was observed in the performance of the reactors and no

VFA was detected in the digestate. In addition, dilution of sedimented fiber did not improve process performance. During co-digestion of sedimented fiber with sewage sludge higher methane production and stable performance at an OLR of 1.5 kg VS/m^3.d suggests feasibility of the process and also indicates the possibility of increasing the OLR. However, while scaling up these reactors, consideration needs to be given to both waste treatment and methane production to reach to an economic solution for the remediation of sedimented fiber and the usage of digestate.

Scaling up of AD treating sedimented pulp and paper fiber would probably need shorter HRT and higher OLR without compromising the reactor stability. The process stability of an anaerobic digester is dependent on the balance between microorganisms, which are known to be vulnerable, e.g., to inhibition by high VFA or ammonia or changes in the process variables like OLR (Chatterjee et al., 2017; McLeod et al., 2015; Tampio et al., 2016). Thus, increasing OLR can lead to a process imbalance, accumulation of VFAs and a decrease in methane production if the retention times are not sufficient for microbial growth (Chatterjee et al., 2017; Regueiro et al., 2015). Increasing OLR up to 2.5 kg VS/m^3.d in the reactor fed with non-diluted sedimented fiber (R1) did not result in any of the process imbalances mentioned above, suggesting the possibility of further increasing the OLR in CSTRs treating sedimented fibers.

There is approximately 1.5 million m^3 sedimented fibers accumulated in the studied bay area in Lake Näsijärvi, Finland. In Figure 4, the time required to treat the entire quantity of sedimented fiber is estimated. The estimations are done based on the two best results obtained in this study: 1) anaerobic treatment of non-diluted sedimented fibers at an OLR of 2.5 kg VS/m^3.d and 2) anaerobic treatment of sedimented fibers with co-digestion of sewage sludge. Assuming a reactor volume of 5000 m^3, to treat the entire volume at an OLR of 2.5 kg VS/m^3.d in eleven years, four reactors would be needed, while the same number of reactors can treat the waste in less than 9 years if operated at an OLR of 5 kg VS/m^3.d. Stable operation of AD at such high OLRs of around 5 kg VS/m^3.d has been reported for treating high solid containing waste like dewatered sewage sludge (Nges and Liu, 2010). Also it is interesting to note that the volume or number of digesters required to treat the entire mass of sedimented fiber does not follow a linear trend with the time required for treatment, rather it is an asymptotic curve. Hence, it is possible to decide on a break-even point to minimize costs by treating the fibers at the shortest time as possible and with the lowest capital costs. Assuming that nine digesters are used for co-digestion, operated at an OLR of 2.5 kg VS/m^3.d and HRT of 30 d, all of the sedimented fibers could be treated in 5.5 years when using a 1:1 mass ratio (5:1 VS ratio) with sewage sludge, which would increase to 17.5 years if a mass ratio of 1:15 (1:3 VS ratio) is used. A major reduction in required reactor numbers could be achieved by increasing the treatment time to at least 5 years (Fig. 4). Depending on budget and available facilities decisions on OLR, co-digestion and actual treatment time can be decided. The existing facilities could be utilized to treat a part of the fibers.

Decreasing pH and VFA accumulation with time has been commonly reported in anaerobic digesters (Appels et al., 2008). Thus, two-stage anaerobic digestion instead of one stage process is often recommended to have production of VFAs in the first reactor, which prevents accumulation of acids in the methanogenic phase under, e.g., increased OLRs (Banks and Humphreys, 1998). Future experiments can be designed to explore the possibility of using such two-stage treatment procedure for sedimented fibers. Alternative reactor designs and set-ups could be considered, e.g. for solid and liquid fractions produced from sedimented fiber using mechanical dewatering equipment.

Figure 4

4. Conclusions

Anaerobic treatment of decades old sedimented fiber, from pulp and paper mill, collected from the bottom of a bay was studied in this paper. Process operation at an OLR of 2.5 kg VS/m^3.d treating non-diluted sedimented fibers with nitrogen and buffer supplement achieving around 60% VS destruction and methane yield of around 200 L/kg VS was shown feasible. The reactor studies showed that the fibers can be used as such without any form of dilution. High methane yield of around 250 L/kg VS and stability of process during co-digestion with sewage sludge opens the option of using already existing sewage sludge digesters for remediating these fibers. Stable operation and good quality digestate indicates possibility of operating digesters at even higher organic loading, which would decrease the required digester volume from 20000 m^3 to 13000 m^3, to remediate the 1.5 million m^3 of sediments in 10 years.

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Figure captions

Figure 1. Schematic representation of the CSTR reactors used in the experiments (All dimensions are in mm)

Figure 2: (A) Weekly OLR, (B, C) weekly methane yield, and (D) pH in the CSTRs (R1: Sedimented fiber, R2: Diluted sedimented fiber, R3: Diluted sedimented fiber pH 7, R4: Co-digestion) (The drop in OLR around day 49 was due to non-feed conditions for technical problems)

Figure 3: (A) sCOD, (B) VFA in the CSTR digestates

Figure 4: (a) Total reactor volume with increase in treatment time at different OLRs, (b) Treatment time required with increasing OLR while co-digesting sedimented fiber with sewage sludge at different dilutionsu using nine reactors, (c) Number of digesters required (assumed volume of 5000 m^3) with increase in treatment time at different OLRs.

Table 1. Characteristics of the feed materials and inoculum

Feed/inoculum	**TS (%)**	**VS (%)**	**VS/TS (%)**	**sCOD (g/L)**	**TN (mg/L)**	**PO_4^{2-}-P (mg/L)**	**VS:TN**
Sedimented fibers	13.3	12.6	94.4	3.58	240	41.7[a]	420
Sewage sludge	3.7	2.5	68.1	1.31	1210	716	17
Inoculum	3.0	1.7	54.3	0.53	1425	431	11

[a] Lindroos et al., 2017

Table 2. The feed compositions and operational parameters of the four reactors (the values reported are average with standard deviations).

Reactor	**Feed**	**TS (%)**	**VS (%)**	**VS:TN**	**HRT (d)**	**OLR (kg VS/m^{3}d)**	**Days of operation**
R1	Sedimented fiber	13.0 ± 0.5	12.3 ± 0.5	184 159 (after N addition)	60	2.5	91
R2	Diluted sedimented fiber	4.5 ± 0.2	4.2 ± 0.2	62 51 (after N addition)	30	1.5	77
R3	Diluted sedimented fiber, pH 7	4.5 ± 0.2	4.2 ± 0.2	62 51 (after N addition)	30	1.5	77
R4	Co-digestion	4.0 ± 0.4	2.8 ± 0.3	20	30	1.04	42
					20	1.5 (days 45-91)	49

Weekly nitrogen and trace element additions in R1-R3 were started on days 53 and 60, respectively.

Table 3: Overall performance of the CSTRs treating fiber sediments and comparison with literature (the values reported are average along with standard deviations)

	Days	**Digestate**			**VS reduction (%)**	**Average Methane yield (L/kg VS)**	**Methane content (%)**	**Reference**
		TS (%)	**VS (%)**	**sCOD (g/L)**				
R1 (Sedimented fiber)	50 - 70	6.9 ± 0.5	5.9 ± 0.4	0.8 ± 0.1	54 ± 7	157 ± 29	52 ± 3	This study
R1 (after nutrient addition and pH correction)	78 - 91	5.9 ± 1.3	5.0 ± 1.2	0.8 ± 0.1	65 ± 5	201 ± 18	55 ± 1	This study
R2 (Diluted sedimented fiber)	22 - 49	2.7 ± 0.4	2.3 ± 0.2	0.6 ± 0.2	46 ± 5	144 ± 27	50 ± 2	This study
R2 (after nutrient addition)	64 - 77			0.8 ± 0.2		167 ± 19	60 ± 4	This study
R3 (Diluted sedimented fiber pH 7)	15 - 49	3.0 ± 0.5	2.3 ± 0.1	0.5 ± 0.1	43 ± 5	161 ± 10	54 ± 4	This study
R3 (after nutrient addition)	64 - 77			0.6 ± 0.1		179 ± 47	50 ± 2	This study
R4 (Co-digestion, OLR 1.0 kg VS/m^3.d)	29 - 44	2.9 ± 0.7	2.4 ± 0.8	0.6 ± 0.1	27 ± 7	262 ± 19	61 ± 2	This study
R4 (Co-digestion, OLR 1.5 kg VS/m^3.d)	50 - 91	2.2 ± 1.2	1.3 ± 0.7	0.7 ± 0.1	37 ± 14	246 ± 10	62 ± 1	This study

Primary sludge	1.9 ± 0.1	2.9 ± 0.4	40	240	(Bayr and Rintala, 2012)
Mixture PS and WAS			41 % VSS	90 L/kg VSS	(Puhakka et al., 1988)
Mixture municipal sludge, PS and WAS (Bench scale)			27	185	(Jokela et al., 1997)

PS – primary sludge, WAS – waste activated sludge