Journal of Environmental Management 269 (2020) 110720

Contents lists available at ScienceDirect

Journal of Environmental Management

journal homepage: http://www.elsevier.com/locate/jenvman

Research article

Life cycle assessment and economic analysis of anaerobic membrane bioreactor whole-plant configurations for resource recovery from domestic wastewater

Melissa Harclerode[a,*], Alexandra Doody[b], Andrew Brower[c], Paloma Vila[d], Jaeho Ho[e], Patrick J. Evans[e]

[a] CDM Smith, 110 Fieldcrest Avenue, PO 8 Fl 6, Edison, NJ, 08837, USA
[b] CDM Smith, 9430 Research Blvd, Suite 1-200, Austin, TX, 79759, USA
[c] CDM Smith, 11490 Westheimer Rd, Suite 700, Houston, TX, 77077, USA
[d] CDM Smith, 993 Old Eagle School Rd, Suite 408, Wayne, PA, 19087, USA
[e] CDM Smith, 14432 SE Eastgate Way, Suite 100, Bellevue, WA, 98007, USA

A R T I C L E I N F O

Keywords:
Anaerobic membrane bioreactor (AnMBR)
Life cycle assessment
Domestic wastewater treatment
Nutrient and sulfide removal
Dissolved methane
Life cycle cost analysis

A B S T R A C T

The use of the anaerobic membrane bioreactor (AnMBR) process for domestic wastewater treatment presents an opportunity to mitigate environmental, social, and economic impacts currently incurred from energy-intensive conventional aerobic activated sludge processes. Previous studies have performed detailed evaluations on improving AnMBR process subcomponents to maximize energy recovery and dissolved methane recovery. Few studies have broadly evaluated the role of chemical use, membrane fouling management, and dissolved methane removal technologies. A life cycle assessment was conducted to holistically compare multiple AnMBR-based domestic wastewater treatment trains to conventional activated sludge (CAS) treatment. These treatment trains included different scouring methods to mitigate membrane fouling (gas-sparging and granular activated carbon-fluidizing) with consideration of upstream treatment (primary sedimentation vs. screening only), downstream treatment (dissolved methane removal and nutrient removal) and sludge management (anaerobic digestion and lime stabilization). This study determined two process subcomponents (sulfide and phosphorus removal and sludge management) that drove chemical use and residuals generation, and in turn the environmental and cost impacts. Furthermore, integrating primary sedimentation and a vacuum degassing tank for dissolved methane removal maximized net energy recovery. Sustainability impacts were further mitigated by operating at a higher flux and temperature, as well as by substituting biological sulfide removal for chemical coagulation.

1. Introduction

Growing environmental awareness and public health concerns have led to more stringent environmental regulations and widespread implementation of advanced systems for wastewater treatment, which has resulted in a substantial increase in energy consumption for wastewater treatment (Pabi et al., 2013). Addressing the question of how to reduce energy consumption and recover resources during wastewater treatment is important for sustainable development. Efficient wastewater treatment capability coupled with a potential for resource recovery has stimulated interest in anaerobic technologies for domestic wastewater in recent years (Watanabea et al., 2014). Anaerobic treatment offers many inherent benefits compared to aerobic treatment, such as low energy consumption and reduced sludge production. It plays a dual role of converting wastewater into methane-rich biogas and minimizing environmental-social impacts by sufficiently treating wastewater prior to discharge or reuse. However, long hydraulic and solids retention times can be required to achieve sufficient treatment.

Anaerobic membrane bioreactors (AnMBR) have the potential to address these treatment challenges. AnMBRs retain the biomass in the reactor and allow the hydraulic retention time (HRT) and solids retention time (SRT) to be independent of each other. AnMBR wastewater

* Corresponding author.
E-mail address: harclerodema@cdmsmith.com (M. Harclerode).

https://doi.org/10.1016/j.jenvman.2020.110720
Received 13 August 2019; Received in revised form 4 May 2020; Accepted 6 May 2020
Available online 17 May 2020
0301-4797/

treatment has been recognized as a promising low-energy alternative to energy-intensive conventional activated sludge (CAS) processes (McCarty, 2018; Salveson et al., 2013). Several studies have evaluated the cost associated with AnMBR-based domestic wastewater treatment systems with different process configurations (Pretel et al., 2015). evaluated the total costs for three different treatment schemes including AnMBR, AnMBR + anaerobic digester (AD), and primary settler (PS) + AnMBR + AD. Although AnMBRs operated at ambient temperature were concluded to be a sustainable technology compared to CAS and aerobic MBRs (Cashman et al., 2018; Pretel et al., 2016), AnMBR could have higher environmental impacts than other technologies if downstream polishing steps to remove dissolved methane and nutrients (Smith et al., 2014) are not incorporated.

Improvements recommended for AnMBRs have mainly focused on membrane performance, biogas production, dissolved methane removal, and biological efficiency (Cashman and Mosely, 2016; Crone et al., 2016; Ozgun et al., 2013; Smith et al., 2014; Watanabea et al., 2014). Few studies, if any, have evaluated the costs and environmental sustainability impacts of sulfide removal from AnMBR permeate; which can be present in high concentrations, thus precluding discharge or reuse. Sulfide can produce toxic and unpleasant odors, corrode and foul downstream equipment, and deleteriously impede discharge or reuse (Pokorna and Zabranska, 2015). Also, costs associated with gas sparging for membrane fouling mitigation are a limiting factor for widespread use of AnMBRs (Pretel et al., 2015). Several investigators (Bae et al., 2013, 2014; Kim et al., 2011; McCarty et al., 2011; Smith et al., 2014) have hypothesized that use of granulated active carbon (GAC) in a fluidized bed to control membrane fouling may significantly reduce the energy consumption and operational cost compared to gas sparging. However, limited direct comparisons have been conducted (Evans et al., 2019). Other methods for membrane fouling mitigation include pretreatment of feed, optimization of operating conditions, addition of substances to the membrane feed, modification of membrane properties, and physical or chemical cleaning (Lin et al., 2013). This study presents life cycle cost (LCC) and life cycle assessment (LCA) evaluations of eight AnMBR process scenarios for domestic wastewater treatment. This study is the first that has holistically evaluated cost and life cycle impacts of the entire treatment process including primary treatment, alternative membrane fouling management methods, sulfide and phosphorus removal, dissolved methane removal, residuals management, as well as net energy consumption.

2. Methods

2.1. Treatment scenarios and design parameters

Two pilot-scale AnMBR processes with different methods of membrane fouling management –biogas sparging and GAC fluidization – were tested on screened domestic wastewater for over one year (Evans et al., 2018, 2019; Lim et al., 2019). The results of these studies provided valuable long-term operational data on treatment performance and cost. A hybrid process was envisioned that had the potential to leverage the advantages of each of these systems. In addition, supplemental treatment processes were conceptualized to improve overall performance or decrease cost. The study summarized herein was conducted to understand and compare the lifecycle impacts and costs of these processes and compare them to conventional sewage treatment. The operating and design conditions are based on experimental results from the pilot studies described previously (Evans et al., 2018, 2019; Lim et al., 2019) and engineering best professional judgement as described further below. Conditions based on engineering best professional judgement will need to be validated with experimental data or calibrated models.

Eight AnMBR process scenarios and one CAS scenario were evaluated for a conceptual 5-million gallon per day (MGD) (790 m^3/h) treatment plant design. A 5-MGD plant was chosen because this is the minimum size for which cogeneration units are considered to be economically viable (Naik-Dhungel, 2010). Each system was designed to treat medium strength wastewater to EPA's technology-based effluent limitations summarized in Tables S1 and S2 (Tchobanoglous et al., 2014). Scenarios 1 and 2 represent the gas-sparged and GAC-fluidized AnMBRs that were pilot tested on screened domestic wastewater (Evans et al., 2018, 2019). Scenario 3 is a hybrid of Scenarios 1 and 2 based on recommendations of the pilot study (Evans et al., 2018, 2019), which consists of the GAC-fluidized bed bioreactor followed by gas-sparged ultrafiltration membranes. Scenarios 4 through 6 are equivalent to Scenarios 1 through 3 but include primary sedimentation and anaerobic digestion of the primary sludge in addition to screening. Scenarios 1 through 6 include a hollow-fiber gas-liquid contactor for dissolved methane removal and coagulation-flocculation-sedimentation with ferric chloride and cationic polymer for sulfide and phosphorus removal. A 90% recovery of dissolved methane from AnMBR permeate was assumed and is considered achievable based on previous studies (Velasco et al., 2018). Design effluent phosphorus and sulfide concentrations were 1 mg/L and 0.1 mg/L, respectively. Scenario 7 is identical to Scenario 6 but integrates a vacuum flash tank for dissolved methane removal instead of a hollow-fiber contactor. A biological sulfide removal process in conjunction with Scenario 7 (Scenario 7 A) was also evaluated for comparison with chemical sulfide removal. Biological sulfide oxidation was designed with an up-flow biological aerated filter (BAF) using floating packed media. Under oxygen limiting conditions, sulfur oxidizing bacteria are able to partially oxidize sulfide to elemental sulfur (Cai et al., 2017; de Sousa et al., 2017). Scenario 8 is CAS treatment with anaerobic digestion, also referred to herein as "conventional treatment". The main process units included in each treatment scenario are shown in Table S3. Process flow diagrams for each of these scenarios are provided in Figs. S1–S8.

Two temperature conditions (<20 °C, >25 °C) and three operational flux values were compared in this study. Low (7.5 L m^{-2} h^{-1} [LMH]) and moderate flux (15 LMH) represent performance observed in the pilot study (Evans et al., 2019). High flux (30 LMH) has not been demonstrated but might be achievable in the future with additional development. Each scenario was divided into the following eight subcomponents to further evaluate the extent specific design components contribute towards cost and environmental impacts: primary treatment, secondary treatment, sulfide and phosphorus removal, sludge management, biogas handling and treatment, dissolved methane removal, disinfection, and waste handling.

2.2. Life cycle cost methodology

LCC were developed for the eight AnMBR scenarios and the CAS scenario as 20-year net present worth in 2019 U.S. dollars using a discount rate of 7%. Direct capital costs for the conventional treatment facilities were developed based on scaling of costs from comparable projects implemented by the authors, while costs for less common processes (AnMBR, dissolved methane contactors, etc.) were estimated using equipment vendor pricing and estimated quantities for materials, such as concrete, tank covers, and pre-engineered buildings. Table S4 summarizes the direct costs for each major process unit including the cost of structures, mechanical equipment and installation, and electrical and automation allowances. Indirect costs for taxes, bonds and insurance, contractor's overhead and profit, construction contingency, and engineering design fee were estimated as percentages of the direct cost. The capital costs represent a Class 5 estimate as defined by AACE International (Bredehoeft et al., 2019), which can be used for concept screening purposes. Annual operations and maintenance (O&M) costs were developed for the average daily flow and load conditions in Table S1, using the unit prices and membrane replacement frequencies indicated in Table S5 and standard motor efficiencies and duty cycles for each major piece of equipment. All scenarios include energy recovery, and thus power costs were included only for the power required beyond that produced on-site. Chemical costs included hypochlorite for

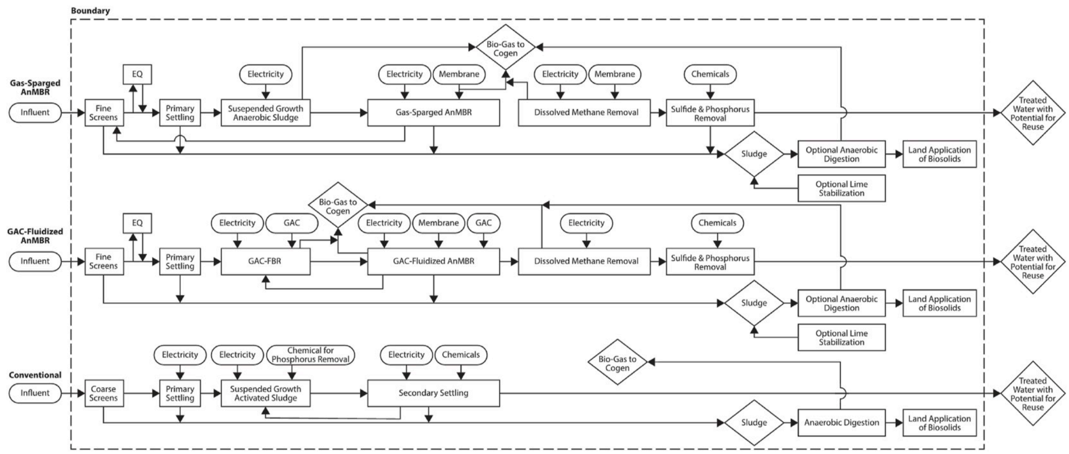

Fig. 1. Life cycle boundary of treatment scenarios, specifically Scenario 1 gas-sparged AnMBR, Scenario 2 GAC-fluidized AnMBR, and Scenario 8 conventional treatment.

disinfection, hypochlorite and citric acid for membrane cleaning, coagulants for phosphorus and sulfide removal, polymer for sludge dewatering, lime for stabilization of biosolids (for certain scenarios), and GAC replacement. Costs for influent equalization were also included for the AnMBR scenarios to reduce the flow peaking factor from three to two, which is generally a more cost-effective approach than sizing the membrane area for peak flow conditions.

To highlight the importance of evaluating contributions to global warming impacts, the costs borne by society from the total carbon dioxide equivalent (CO2eq) footprint were separately quantified by integrating the social cost of carbon (U.S. Government, 2013). The costs borne by society represents financial implications to society for mitigation of climate change impacts.

2.3. Life cycle assessment methodology

2.3.1. LCA boundary and functional unit

A process flow diagram showing the LCA boundary is shown in Fig. 1. The functional unit is treatment of 5 million gallons of medium strength wastewater to the same effluent characteristics. The LCA considered consumption of raw materials and natural resources during materials acquisition, production, use stages, and end-of-life processes during plant operation. The water quality of the effluent was assumed to be the same for all processes and, therefore, was not considered in the LCA boundary. In addition, the construction of the treatment plant infrastructure was not included in the LCA boundary, as previous studies have shown the operation stage of the plant as the primary environmental impact contributor (Smith et al., 2014). The ISO 14040 framework was used to compare the environmental impacts of the AnMBR and CAS scenarios.

2.3.2. Life cycle inventory

Primary data regarding energy and material consumption during each treatment process (Table S3) were compiled from pilot test results (Evans et al., 2018), full-scale treatment plants designed by the authors, engineering calculations, and vendor specifications. Life cycle inventory (LCI) data for treatment system operation (e.g., production of chemicals, membrane materials) were based on average technology data from the EcoInvent life cycle unit process database Version 3 allocation at the point of substitution (APOS) (Wernet et al., 2016). EcoInvent LCI geography was primarily global (GLO), with minor uses of Europe (RER) and Rest-of-World (Row) if GLO unit process was not available. The treatment system operational electricity mix was based on U.S. EcoInvent unit process. Environmental impact offsets designated as “avoided products” were associated with energy production from biogas and methane recovery, elemental sulfur byproduct recovered during biogas treatment prior to cogeneration, and excess heat from biogas cogeneration. All membrane materials were assumed to be recycled at the end of their useful life. Waste scenarios for membrane recycling was developed under the Processes function of SimaPro 8.3 to indicate percentage of materials and/or waste types separated from the waste stream. The life cycle inventory for recycling of polyvinyl chloride (PVC), polyethylene (PE), and polypropylene (PP) was created by Pré Consultants and available in the SimaPro 8.3 program (PReConsultants, 2018). Grit generated during primary treatment was assumed to be disposed of as inert material at a sanitary landfill. Biological and chemical sludge generated in secondary and tertiary treatment were assumed to be beneficially used through land application (Smith et al., 2014).

2.3.3. Life cycle impact assessment

The life cycle impact assessment (LCIA) method used was the Tool for the Reduction and Assessment of Chemical and other Environmental Impacts (TRACI) Version 2.1 (Bare, 2011), to facilitate comparison to previous studies (Cashman et al., 2018; Cashman and Mosely, 2016; Pretel et al., 2015; Smith et al., 2014). The TRACI environmental impact categories evaluated included: ozone depletion, global warming, smog, acidification, eutrophication, carcinogenics, non-carcinogenics, respiratory effects, ecotoxicity, and fossil fuel depletion. Two normalization analyses were performed: 1) one using the EPA’s TRACI 2.1 LCIA method (Bare, 2011) for the United States region, which uses normalization factors (NF) to relate the impact scores to the average impact of a U.S. citizen per year; and 2) the second using NFs relating impact scores to conventional treatment LCA outputs, as conducted in similar studies (Smith et al., 2014). Sankey Diagrams (Schmidt, 2008) were used to achieve a better understanding of the components of the treatment process that are major contributors to environmental impacts and thus resulting in greater contributions to impact categories in comparison to conventional treatment. Each treatment scenario was evaluated individually to examine which temperature and flux conditions had the greatest contribution to each impact category. Additionally, each temperature and flux condition was evaluated individually.

2.4. Sensitivity analysis

Sensitivity analyses were performed on the LCC and LCA to evaluate cost and environmental impact drivers associated with sulfide and phosphorus removal. Sensitivity analyses specifically evaluated the effects of eliminating the sulfide and phosphorus removal process entirely; substituting the coagulant used for sulfide and phosphorus removal with aluminum sulfate (i.e., alum); and substituting biological sulfide removal for chemical coagulation (Scenario 7 A).

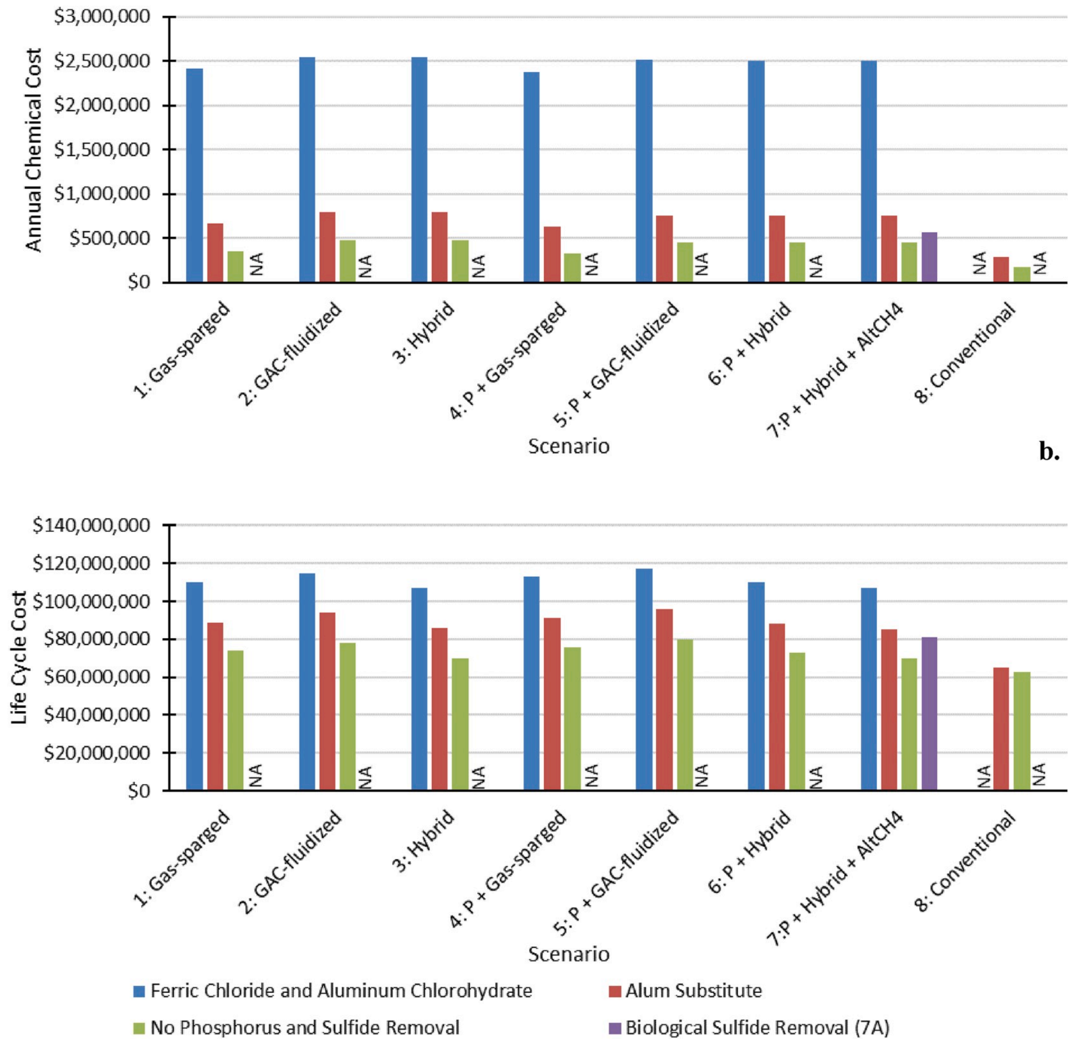

Fig. 2. Comparison of total annual chemical costs (a) and life cycle costs (b) associated with different approaches for phosphorus and sulfide removal at 15 LMH and >25 °C. Total costs include chemicals for membrane cleaning and sludge management. P=Primary sedimentation. AltCH4 = Vacuum flash tank for dissolved methane removal.

3. Results and discussion

In general, the findings discussed herein are representative of all evaluated flux and temperature conditions. Results associated with the 15 LMH, > 25 °C condition are presented to illustrate the findings of this study.

3.1. *Nutrient removal and sludge management drives chemical consumption and residual generation*

Sulfide and phosphorous removal with coagulation using ferric chloride and aluminum chlorohydrate (ACH) required approximately 82% more chemicals compared to conventional treatment (Fig. S9a). Membrane cleaning chemicals and GAC replacement were minor, totaling no more than 3.1% of total chemical demand for AnMBR treatment (Fig. S9). Disinfection chemical demand was 20% higher for AnMBR than for CAS due to greater chlorine demand of anaerobically treated wastewater, however the overall impact of disinfection chemicals on total AnMBR chemical use was relatively small (4% AnMBR treatment, 18% CAS).

Sludge management chemicals (i.e. dewatering polymer addition and for some scenarios, lime for sludge stabilization) made up varying fractions of overall chemical use (4% AnMBR plus primary sedimentation, 15%–16% AnMBR without primary sedimentation, 18% CAS), but in general were required in lesser quantities compared to those for sulfide and phosphorus removal (Fig. S9). The largest amount of sludge management chemical used was during lime stabilization (Scenarios 1–3) as opposed to anaerobic digestion (Scenarios 4–8) to produce Class B biosolids suitable for land application.

Sulfide and phosphorous removal with coagulation using ferric chloride and ACH produced approximately twice the total residuals (1.45–1.48 kg/m^3 AnMBR treatment) compared to conventional treatment (0.65 kg/m^3) (Fig. S10a). Residuals generated include screenings and grit, biosolids, and chemical solids associated with phosphorus and sulfide removal. Biosolids quantities were lower for all AnMBR technologies (Scenarios 1–7, 0.34–0.37 kg/m^3) compared with conventional treatment (Scenario 8, 0.61 kg/m^3).

3.2. *Alternative chemical coagulants and biological processes minimize chemical consumption and residuals generation*

Using alum as a substitute for ferric chloride with ACH reduced chemical consumption for sulfide and phosphorus removal by 49%–57% for the AnMBR treatment scenarios (Fig. S9b); however, chemical consumption (0.18–0.25 kg/m^3) was still greater than conventional treatment (0.08 kg/m^3). Using alum coagulant resulted in total residual quantities (0.57–0.60 kg/m^3) similar to conventional treatment (0.65 kg/m^3) (Fig. S10b).

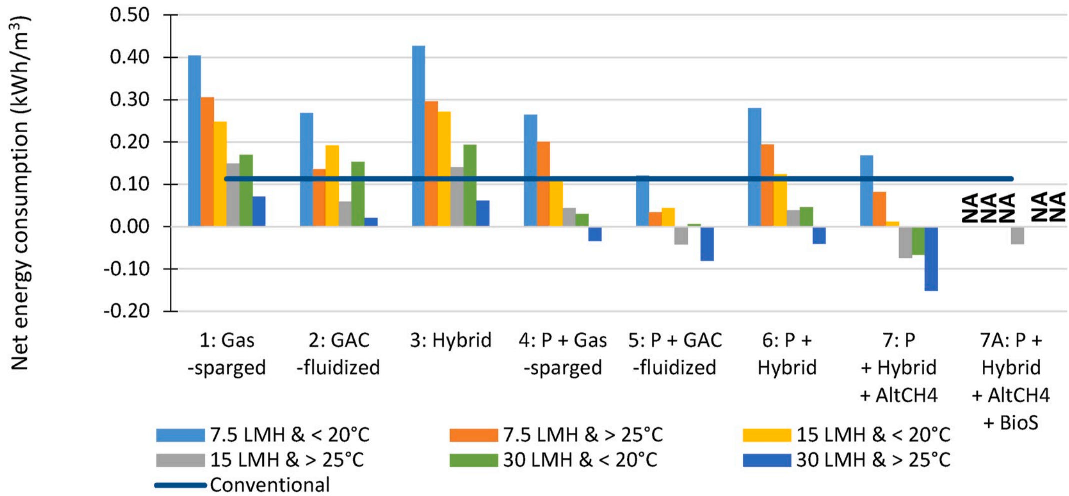

Fig. 3. Net energy consumption for different AnMBR scenarios compared to conventional treatment including comparison of biological sulfide oxidation for 15 LMH, >25 °C. P=Primary sedimentation. AltCH4 = Vacuum flash tank for dissolved methane removal. BioS = Biological sulfide oxidation.

Alternative methods for sulfide removal, such as biological oxidation, have potential to further reduce AnMBR chemical use and residuals production relative to CAS. Biological sulfide oxidation (Scenario 7 A) was evaluated for comparison with chemical-based treatment technologies for Scenario 7. Coagulation chemicals were still used to remove phosphorus, but sulfide was oxidized to elemental sulfur using a packed-tower (Cai et al., 2017; de Sousa et al., 2017). Biological sulfide oxidation reduced total chemical usage (0.10 kg/m^3) to a quantity similar to conventional CAS treatment (0.08 kg/m^3), with that quantity being used primarily for phosphorus removal (Fig. S11). Biological sulfide oxidation also produced fewer residuals (0.46 kg/m^3) than all other alternatives (0.57–1.48 kg/m^3 for AnMBR scenarios with and without alum substitute; 0.65 kg/m^3 for conventional) (Fig. S12).

3.3. Chemical consumption required for nutrient removal drive cost impacts

Chemical costs for sulfide and phosphorous removal represented between 64% and 77% of overall AnMBR operating costs, compared with membrane cleaning costs which represented 2%–7% of total operating costs (Tables S13 and S14). Membrane replacement costs varied widely from 3% of operating cost for high-flux gas sparged reactors, to 22% for low-flux GAC-fluidized reactors. Power costs were relatively low (ranging from net −3%, to +6% of total operating costs) because power generated on-site offset the power required for the process operations.

Using biological sulfur oxidation with a vacuum flash tank for dissolved methane removal (Scenario 7 A) reduced the chemical and operating costs and, as a result, total project LCC from $100-$120 million for Scenarios 1–7 to $81 million (Fig. 2 and Fig. S15b). The life cycle cost of Scenario 7 A was within 25% of that for CAS Scenario 8, which is within the range of accuracy for a Class 5 cost estimate. The 20-year LCC for Scenario 7 A which incorporates biological sulfur oxidation ($81 M net present worth) is slightly lower (approximately 8%) than the Scenario 7 alum alternative ($85 M net present worth) (Fig. 2). This reduction in cost of only 8% was attributable to a higher overall capital cost and lower operating cost for biological sulfide oxidation compared to alum coagulation. Biological phosphorus removal could potentially reduce lifecycle costs further but was not evaluated.

Even with chemical substitute (alum) and biological removal design improvements, the estimated life cycle costs for each AnMBR scenario operating at all three fluxes remain greater than conventional treatment because of several factors, as demonstrated in Figs. S15 and S16. First, capital costs are higher due to the cost of the membranes as well as the more stringent preliminary treatment requirements (i.e., fine screening) upstream of membranes. Additionally, operating costs are higher due to the membrane replacement costs, GAC media replacement (for Scenarios 2–3 and 5–7), membrane cleaning chemical costs, and to some degree the cost of dissolved methane removal from the permeate. Similar findings were identified by Smith et al. (2014) and Pretel et al. (2015). Although advancements in membrane technology, including hypothetical operation at a flux rate of 30 LMH and cost of membrane manufacturing, may reduce cost over time, these costs are not incurred for conventional treatment.

3.4. 3.4 specific process scenarios and operating conditions strongly determine potential for net positive energy operation

Net energy consumption (or in some cases, production) was dependent on specific AnMBR treatment scenarios and operational flux/temperature (Fig. 3). Scenarios 2 and 5 using GAC-fluidized AnMBR consumed less energy (−0.08 to 0.27 kWh/m^3) than gas-sparged AnMBR Scenarios 1 and 4 (−0.03 to 0.40 kWh/m^3) and the hybrid AnMBR Scenarios 3 and 6 (−0.04 to 0.43 kWh/m^3) because energy was not required for membrane gas sparging. However, polymeric ultrafiltration (UF) membranes have been previously demonstrated to be abraded in fluidized GAC systems so Scenarios 2 and 5 are considered impractical (Evans et al., 2019).

Primary sedimentation with mesophilic digestion of primary and secondary solids in Scenarios 4–7 resulted in lower net energy consumption (−0.08 to 0.28 kWh/m^3) compared with Scenarios 1–3 without primary sedimentation (0.02–0.43 kWh/m^3) (Fig. 3). Organics in solids removed by primary sedimentation were more efficiently transformed to methane in an anaerobic digester (Scenarios 4–6) compared to being loaded directly into an AnMBR (Scenarios 1–3). Additional energy required to operate primary clarifiers and mesophilic digesters was small (0.01 kWh/m^3) compared with the additional energy produced by the mesophilic digesters (0.20–0.24 kWh/m^3). Therefore, from an energy perspective, scenarios with primary sedimentation prior to AnMBR treatment are preferable though the life cycle cost may be slightly greater (Fig. 2).

Scenarios 1 through 6 removed dissolved methane from AnMBR permeate with vacuum-assisted, hollow-fiber membranes. High pumping head loss (14.3 m of water) in the membrane contactors negates the energy benefit of methane recovery. A vacuum flash tank was evaluated as an alternative gas removal technology in Scenario 7. Net energy consumption (−0.15 to 0.11 kWh/m^3) under all temperature and flux conditions (except for 7.5 LMH, <20 °C at 0.17 kWh/m^3) was similar or

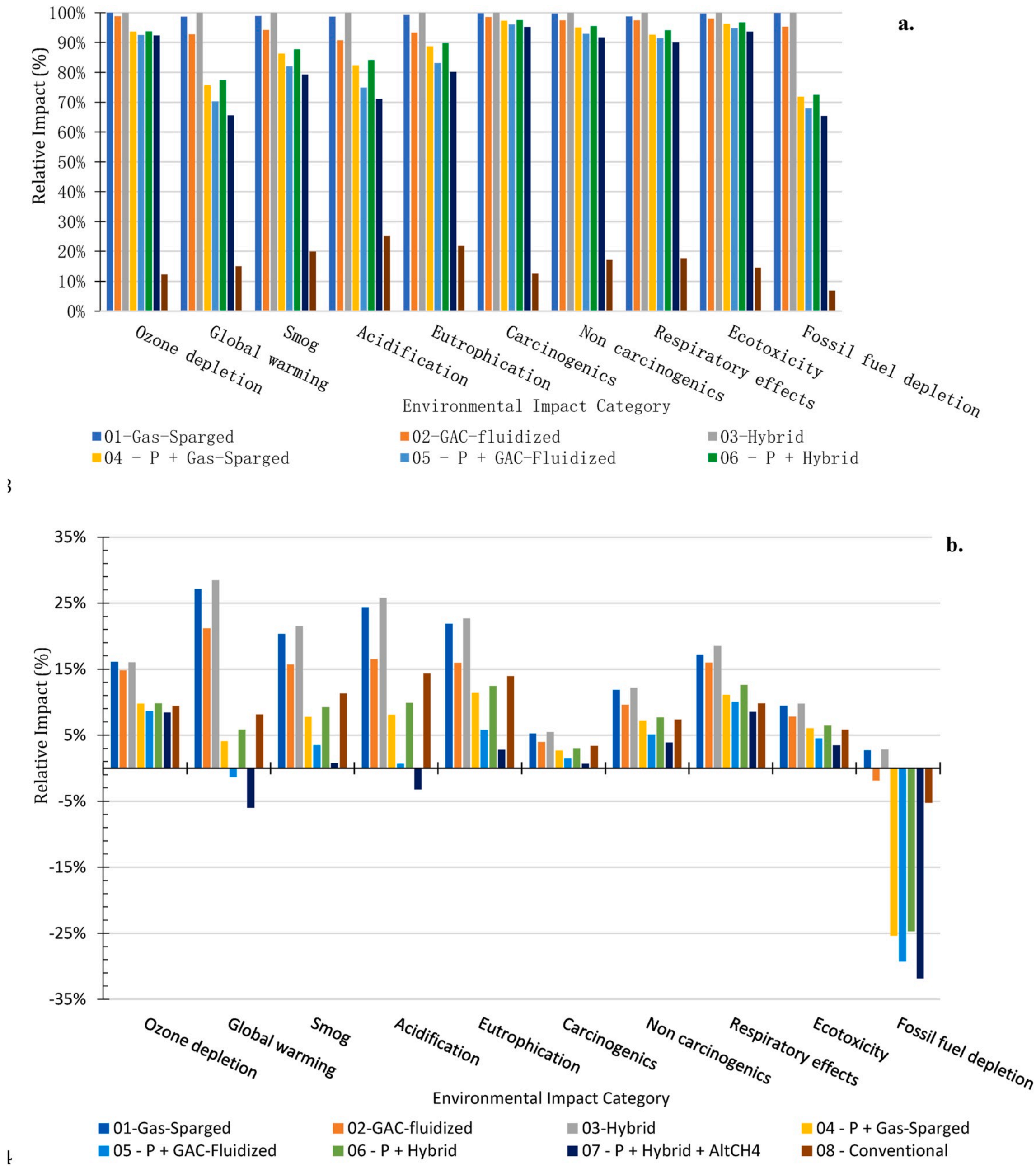

Fig. 4. Relative impacts of the AnMBR and conventional treatment scenarios at 15 LMH (F2) and >25 °C (T1) with (a) and without (b) sulfide and phosphorus removal by chemical coagulation using ferric chloride and ACH. Impacts in *b* are relative to those for Scenarios 1 and 3 in *a*.

less than conventional treatment (0.11 kWh/m^3). Thus the potential for energy positive operation was highly dependent on the process used for dissolved methane removal in addition to use of primary sedimentation.

UF membrane flux and operating temperature also affected the potential for energy-positive operation. Membrane flux had the greatest impact on net energy consumption (Fig. 3). AnMBR pumping and gas-sparging processes were the largest energy consumers for all scenarios at 7.5 LMH (60–76% of total energy consumption) compared to 15 LMH (46–64%) and 30 LMH (35–52%). This energy demand included recycle pumping, permeate pumping, and gas sparging. Higher operating temperatures in the anaerobic systems resulted in higher energy efficiency due to increased hydrolysis and net methane yield (Evans et al., 2018, 2019; Lim et al., 2019). Therefore, high membrane flux and high temperature were the most favorable from an energy perspective, consistent with previous research (Pretel et al., 2016; Smith et al., 2014). Flux maximization for energy efficiency is desirable but will be limited by increasing energy demands for fouling management by gas-sparging. Newer gas-sparging strategies such as intermittent sparging can increase overall energy efficiency while simultaneously minimizing membrane fouling (McAdam et al., 2010; Meng et al., 2009). Temperature in unheated AnMBR designs (as studied herein) is not controllable, but is influenced by geographic location and season. Thus maximization of energy efficiency should consider these factors.

AnMBR treatment was more energy efficient than conventional treatment depending on the process scenario, membrane flux, and operating temperature (Fig. 3). For the medium strength municipal wastewater evaluated for this study, none of the AnMBR alternatives without primary sedimentation were energy-positive. All primary sedimentation scenarios were energy-neutral or -positive when operated at a flux of 15 LMH and at either temperature. Using the vacuum flash tank for dissolved methane removal increased the potential for energy-positive operation. The AnMBR scenario with biological sulfide

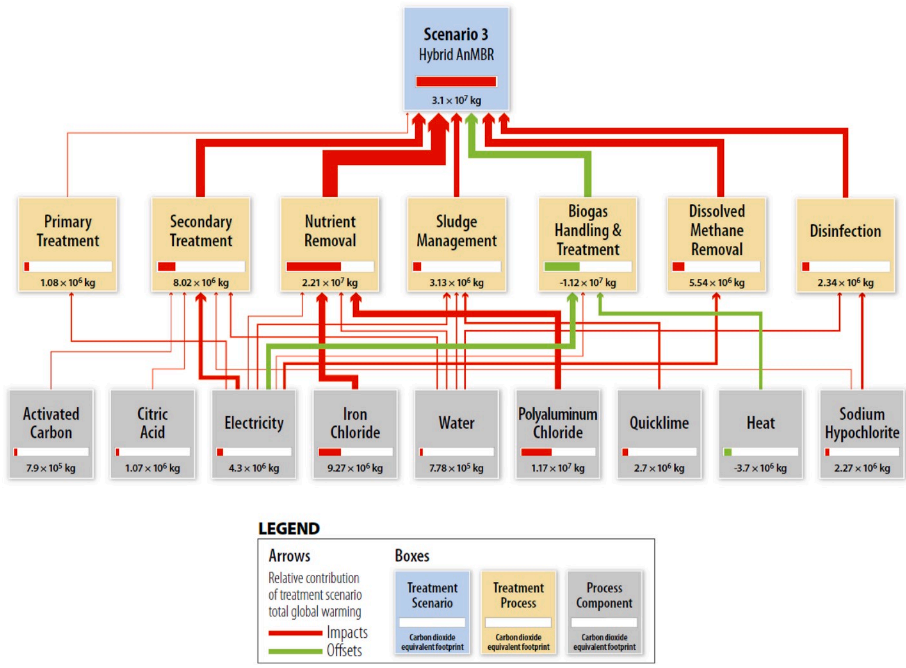

Fig. 5. Global warming (kg CO2 eq) impact assessment normalized to conventional treatment method. Sankey diagram for Scenario 3 – Hybrid AnMBR at 15 LMH and >25 °C.

oxidation (Scenario 7 A) at 15 LMH, >25 °C was less energy efficient than Scenario 7, but was still energy positive (Fig. 3). Thus energy-positive AnMBR operation has good potential provided that attention is paid to specific process selection and operating conditions. Improvements in membrane design and fouling control are also necessary to enable operation at a flux of 15 LMH.

3.5. *Chemical use drives environmental impacts*

AnMBR scenarios 1–7 with chemical coagulation for sulfide and phosphorus removal had contributions to human health carcinogenic hazards (carcinogenics), ecotoxicity, and human health non-carcinogenic hazards (non-carcinogenics) that were approximately 70, 20, and 7 times higher than the average impact of a U.S. citizen per year (i.e., the TRACI 2.1 LCIA normalization factors), respectively; contributions to fossil fuel depletion, ozone depletion, global warming, and ecotoxicity were approximately 16, 8, 7, and 7 times higher than conventional treatment scenario impacts, respectively (Figures S17 through S23). The results suggest that all of the AnMBR scenarios were not capable of reducing environmental impacts more than conventional treatment. Therefore, further evaluation was conducted to identify the sources of these impacts as described below.

Adding primary sedimentation (Scenarios 4–6) reduced overall contributions to environmental impact categories (Fig. 4a, Figs. S24–S28), but still resulted in AnMBR environmental impacts greater than for conventional treatment. Eliminating sulfide and phosphorus removal resulted in more than a 70% reduction in all environmental impact categories for the seven AnMBR scenarios evaluated and in more than 24% reduction for conventional treatment (Fig. 4b). Most significantly, eliminating sulfide and phosphorus removal resulted in environmental impacts for AnMBR with primary sedimentation (Scenarios 4–7) being similar to or less than those for conventional treatment. Sankey diagrams (Fig. 5, S29-S32) show the sulfide and phosphorus removal component as the major contributor to environmental impacts; with ferric chloride and polyaluminum chloride (PACl - representative of aluminum chlorohydrate [ACH]) as the main impact drivers for AnMBR treatment and aluminum sulfate for conventional treatment. Net electricity consumption was also a significant environmental impact driver **(Fig. 5) but not as much as chemical use. Increased flux and temperature also decreased environmental impacts but not as much as chemical use and primary sedimentation (Figs. S17–23)**.

Smith et al. (2014) identified unrecovered dissolved methane as the primary driver (75%) for global warming impacts by AnMBR treatment, followed by electricity (Smith et al., 2014). Sulfide removal was assumed unnecessary in that study. In this study, vacuum-operated membrane contactors were assumed to have potential of removing 90% of dissolved methane from AnMBR permeate, however the pressure loss through the contactors can result in high energy consumption. Alternative dissolved methane removal methods, such as vacuum flash tanks, were shown to decrease environmental impacts (cf. Scenarios 6 and 7, Fig. 4a). The major environmental impacts were associated with sulfide and phosphorus removal and not with releases of unrecovered dissolved methane. Under Scenario 7, sulfide and phosphorus removal was identified as the primary driver (approximately 75%) for global warming impacts, followed by secondary treatment (approximately 48%), disinfection (approximately 8%), and then dissolved methane removal (approximately 6%) (Fig. S33). Approximately 2% of global warming impacts are due to treatment system outputs, including dissolved methane emissions to the atmosphere. Chemical use for sulfide removal was the primary driver of global warming impacts. These results indicate that more attention is required to minimize the environmental impact of sulfide removal rather than dissolved methane removal.

3.6. *Aluminum sulfate as a chemical substitute partially mitigates impacts*

Replacing ferric chloride and PACl in the coagulation process with aluminum sulfate (alum) and reducing the use of polymer in the sludge thickening process resulted in nearly a 20%–50% reduction across all environmental impact categories for the seven AnMBR scenarios (Fig. S34). Fossil fuel impact was nearly eliminated in Scenario 7 P + Hybrid + AltCH4 (includes vacuum degassing tank subcomponent) when alum was used due to offsets from biogas recovery and net energy consumption back to the grid. Fossil fuel impacts for Scenarios 4–6 were less than or similar to conventional treatment. Thus, optimization of

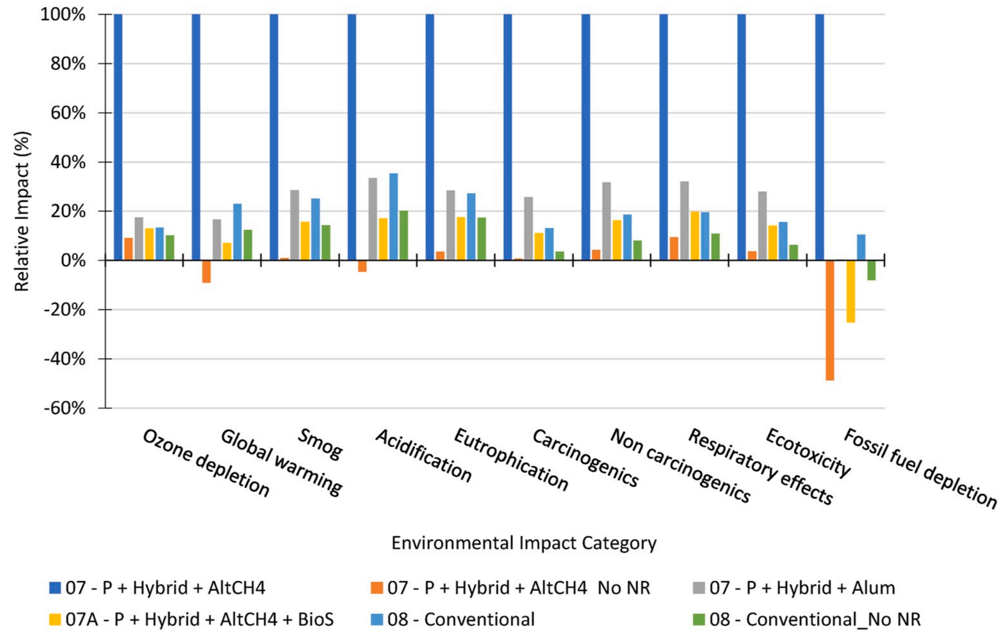

Fig. 6. Characterization impact sensitivity for Process 7 – P + Hybrid + AltCH4 with 15 LMH (F2) and >25 °C (T1) with respect to sulfide and phosphorus removal using biological treatment processes. NR = nutrient removal.

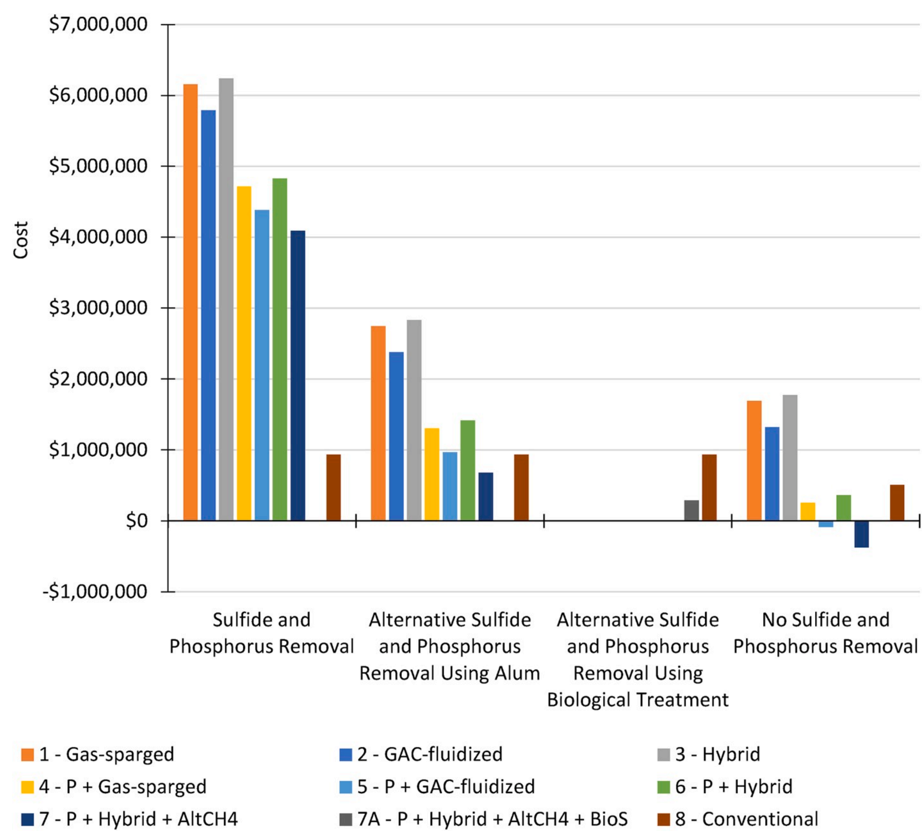

Fig. 7. Costs borne by society (CO2eq) for conventional treatment and AnMBR scenarios at 15 LMH and >25 °C.

chemical coagulation offers an opportunity to mitigate environmental impacts of sulfide generation and removal in AnMBR treatment trains.

3.7. Biological sulfide removal reduces AnMBR impacts below conventional treatment

A biological sulfide removal process in conjunction with Scenario 7 P + Hybrid + AltCH4 (Scenario 7 A) was compared to chemical coagulation (Fig. 6). Biological oxidation resulted in the lowest environmental impacts across all impact categories compared to Scenario 7 even when alum as used as a coagulant. Scenario 7 A also resulted in lower environmental impacts than conventional treatment across all impact categories except respiratory effects, where the impact was similar. Fossil fuel depletion offsets were achieved using Process 7 A. Thus AnMBR treatment of domestic wastewater has the potential to have less environmental impact than conventional treatment provided sulfide removal is conducted using a sustainable process.

3.8. Social cost of carbon

Fig. 7 presents the costs borne by society for all treatment scenarios with a flux of 15 LMH and >25 °C. Financial implications from the CO2eq footprint were significantly reduced by either using an alum substitute or biological treatment for sulfur removal. Under both alternative scenarios, Scenario 7 P + Hybrid + AltCH4 resulted in total social cost of carbon less than conventional treatment (Scenario 8). When no sulfide removal was conducted, all AnMBR scenarios that included primary sedimentation (Scenarios 4–7) have total social costs of carbon less than the conventional scenario. These results support the conclusion that improvements in AnMBR treatment train processes can result in wastewater treatment operations that are less environmentally deleterious than conventional treatment.

4. Conclusions

Sulfide removal from AnMBR effluent using chemical coagulation was determined to have a greater environmental impact than dissolved methane removal and AnMBR energy efficiency. While opportunities exist to optimize coagulation chemistry, oxidative biological sulfide removal has the potential to be more environmentally sustainable and cost-effective. Dissolved methane removal using hollow fiber contactors was determined to be energy-intensive because of high pressure drop. Alternatives such as vacuum-assisted degassing tanks and stripping towers appear to have better potential for sustainable dissolved methane removal (Cai et al., 2017; de Sousa et al., 2017; Evans et al., 2018; Bae, 2015). Upstream of the AnMBR, primary sedimentation combined with anaerobic digestion of the settled solids was determined to be more sustainable than just fine screening. The combination of primary sedimentation with anaerobic digestion, alternative processes for dissolved methane removal, and biological sulfide removal has the potential to render AnMBR treatment of domestic wastewater more energy-efficient and sustainable than conventional aerobic treatment.

Declaration of competing interest

This research was supported by the Department of Defense Environmental Security Program (ESTCP Project number ER-201434; www.serdp-estcp.org/Program-Areas/Environmental-Restoration/Wastewater-and-Drinking-Water/ER-201434) and the U.S. Environmental Protection Agency under U.S. Army Corps of Engineers Humphreys Engineering Service Center Contract W912HQ-15-C-0001. **The authors declare no competing financial interests. Declarations of interest: none.**

CRediT authorship contribution statement

Melissa Harclerode: Methodology, Formal analysis, Writing - original draft. **Alexandra Doody:** Methodology, Formal analysis, Writing - original draft. **Andrew Brower:** Formal analysis, Writing - review & editing. **Paloma Vila:** Software, Formal analysis, Writing - review & editing. **Jaeho Ho:** Writing - review & editing. **Patrick J. Evans:** Conceptualization, Writing - review & editing.

Acknowledgements

This research was supported by the Department of Defense Environmental Security Program (ESTCP Project number ER-201434; www.serdp-estcp.org/Program-Areas/Environmental-Restoration/Wastewater-and-Drinking-Water/ER-201434) and the U.S. Environmental Protection Agency under U.S. Army Corps of Engineers Humphreys Engineering Service Center Contract W912HQ-15-C-0001. The authors declare no competing financial interests. Declarations of interest: none.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvman.2020.110720.

References

Bae, J., 2015, August. Comparison of dissolved methane removal processes: Removal efficiency, energy consumption and application of recovered methane. In: Abstracts of Papers of the American Chemical Society, 250. Amer Chemical SOC, 1155 16th st, NW, Washington, DC 20036 USA.

Bae, J., Shin, C., Lee, E., Kim, J., McCarty, P.L., 2014. Anaerobic treatment of low-strength wastewater: a comparison between single and staged anaerobic fluidized bed membrane bioreactors. Bioresour. Technol. 165, 75–80.

Bae, J., Yoo, R., Lee, E., McCarty, P.L., 2013. Two-stage anaerobic fluidized-bed membrane bioreactor treatment of settled domestic wastewater. Water Sci. Technol. 68 (2), 394–399.

Bare, J., 2011. Traci 2.0: the tool for the reduction and assessment of chemical and other environmental impacts 2.0. Clean Technol. Environ. Policy 13 (5), 687–696.

Bredehoeft Jr., P.R, Hollmann, J.K., Dysert, L.R., 2019. AACE International Recommended Practice No. 18R-97: Cost Estimate Classification System - As Applied in Engineering, Procurement, and Construction for the Process Industries, TCM Framework 7.3 - Cost Estimating and Budgeting. AACE International, Morgantown, WV.

Cai, J., Zheng, P., Qaisar, M., Zhang, J., 2017. Elemental sulfur recovery of biological sulfide removal process from wastewater: a review. Crit. Rev. Environ. Sci. Technol. 47 (21), 2079–2099.

Cashman, S., Ma, X., Mosley, J., Garland, J., Crone, B., Xue, X., 2018. Energy and greenhouse gas life cycle assessment and cost analysis of aerobic and anaerobic membrane bioreactor systems: influence of scale, population density, climate, and methane recovery. Bioresour. Technol. 254, 56–66.

Cashman, S., Mosely, J., 2016. Life cycle assessment and cost analysis of water and wastewater treatment options for sustainability: influence of scale on membrane bioreactor systems. Environ. Prot. Agency.

Crone, B.C., Garland, J.L., Sorial, G.A., Vane, L.M., 2016. Significance of dissolved methane in effluents of anaerobically treated low strength wastewater and potential for recovery as an energy product: a review. Water Res. 104, 520–531.

de Sousa, J.T., Lima, J.F., da Silva, V.C., Leite, V.D., Lopes, W.S., 2017. Recovery of elemental sulphur from anaerobic effluents through the biological oxidation of sulphides. Environ. Technol. 38 (5), 529–537.

Evans, P.J., Doody, A., Harclerode, M., Brower, A., Vila, P., Parameswaran, P., Lim, K., Bae, J., Shin, J., Lee, P.-H., Tan, A., McCarty, P., Parker, W., Guy, K., Page, M., Maga, S., 2018. Anaerobic Membrane Bioreactor for Sustainable Wastewater Treatment. Project ER-201434. Department of Defense Environmental Security Technology Program Final Report. December. https://www.serdp-estcp.org/Program-Areas/Environmental-Restoration/Wastewater-and-Drinking-Water/ER-201434.

Evans, P.J., Parameswaran, P., Lim, K., Bae, J., Shin, C., Ho, J., McCarty, P.L., 2019. A comparative pilot-scale evaluation of gas-sparged and granular activated carbon-fluidized anaerobic membrane bioreactors for domestic wastewater treatment. Bioresour. Technol. 288, 120949.

Kim, J., Kim, K., Ye, H., Lee, E., Shin, C., McCarty, P.L., Bae, J., 2011. Anaerobic fluidized bed membrane bioreactor for wastewater treatment. Environ. Sci. Technol. 45 (2), 576–581.

Lim, K., Evans, P.J., Parameswaran, P., 2019. Long-term performance of a pilot-scale gas-sparged anaerobic membrane bioreactor under ambient temperatures for holistic wastewater treatment. Environ. Sci. Technol. 53 (13), 7347–7354.

Lin, H., Peng, W., Zhang, M., Chen, J., Hong, H., Zhang, Y., 2013. A review on anaerobic membrane bioreactors: applications, membrane fouling and future perspectives. Desalination 314, 169–188.

McAdam, E., Eusebi, A., Judd, S., 2010. Evaluation of intermittent air sparging in an anoxic denitrification membrane bioreactor. Water Sci. Technol. 61 (9), 2219–2225.

McCarty, P.L., 2018. What is the best biological process for nitrogen removal: when and why? Environ. Sci. Technol. 52 (7), 3835–3841.

McCarty, P.L., Bae, J., Kim, J., 2011. Domestic wastewater treatment as a net energy producer - can this be achieved? Environ. Sci. Technol. 45 (17), 7100–7106.

Meng, F., Chae, S.-R., Drews, A., Kraume, M., Shin, H.-S., Yang, F., 2009. Recent advances in membrane bioreactors (MBRs): membrane fouling and membrane material. Water Res. 43 (6), 1489–1512.

Naik-Dhungel, N., 2010. The opportunities for and benefits of combined heat and power at wastewater treatment facilities. Proceedings of the Water Environment Federation (4), 557–566. **2010**.

Ozgun, H., Dereli, R.K., Ersahin, M.E., Kinaci, C., Spanjers, H., van Lier, J.B., 2013. A review of anaerobic membrane bioreactors for municipal wastewater treatment: integration options, limitations and expectations. Separ. Purif. Technol. 118, 89–104.

Pabi, S., Amarnath, A., Goldstein, R., Reekie, L., 2013. Electricity Use and Management in the Municipal Water Supply and Wastewater Industries. Electric Power Research Institute, Palo Alto. **194**.

Pokorna, D., Zabranska, J., 2015. Sulfur-oxidizing bacteria in environmental technology. Biotechnol. Adv. 33, 1246–1259, 6, Part 2.

PReConsultants, 2018. Life Cycle Assessment Software and Database, 1 ed. SimaPro 8.5. 1.

Pretel, R., Monino, P., Robles, A., Ruano, M.V., Seco, A., Ferrer, J., 2016. Economic and environmental sustainability of an AnMBR treating urban wastewater and organic fraction of municipal solid waste. J. Environ. Manag. 179, 83–92.

Pretel, R., Shoener, B.D., Ferrer, J., Guest, J.S., 2015. Navigating environmental, economic, and technological trade-offs in the design and operation of submerged anaerobic membrane bioreactors (AnMBRs). Water Res. 87, 531–541.

Salveson, A., Mackey, E., Juby, G., Killian, R., Reardon, R., Sandino, J., Debroux, J., McCarty, P., Shenakar, L., O'Callaghan, P., 2013. Challenge Projects on Low Energy Treatment Schemes for Water Reuse, Phase 1. WateReuse Research Foundation, Alexandria, Virginia, USA.

Schmidt, M., 2008. The Sankey diagram in energy and material flow management: Part I: History. J. Ind. Ecol. 12 (1), 82–94.

Smith, A.L., Stadler, L.B., Cao, L., Love, N.G., Raskin, L., Skerlos, S.J., 2014. Navigating wastewater energy recovery strategies: a life cycle comparison of anaerobic membrane bioreactor and conventional treatment systems with anaerobic digestion. Environ. Sci. Technol. 48 (10), 5972–5981.

Tchobanoglous, G., Stensel, H.D., Tsuchihashi, R., Burton, F.L., Abu-Orf, M., Bowden, G., Pfrang, W., 2014. Wastewater Engineering. 2014. *Wastewater Engineering. Treatment, Disposal, and Reuse*, fifth ed. McGraw Hill, New York.

U.S. Government, 2013. Technical Support Document - Technical Update of the Social Cost of Carbon for Regulatory Impact Analysis under Executive Order 12866. Interagency Working Group on Social Cost of Carbon. Washington, D.C. Rvised August 2016.

Velasco, P., Jegatheesan, V., Othman, M., 2018. Recovery of dissolved methane from anaerobic membrane bioreactor using degassing membrane contactors. Frontiers in Environmental Science 6 (151).

Watanabea, R., Qiaoa, W., Nortond, M., Wakaharae, S., Lia, Y.-Y., 2014. Recent developments in municipal wastewater treatment using anaerobic membrane bioreactor: a review. Journal of Water Sustainability 4 (2), 101–122.

Wernet, G., Bauer, C., Steubing, B., Reinhard, J., Moreno-Ruiz, E., Weidema, B., 2016. The ecoinvent database version 3 (part I): overview and methodology. Int. J. Life Cycle Assess. 21 (9), 1218–1230.