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Research article

Anaerobic treatment of propylene glycol-contaminated domestic wastewater and microbial community profile at threshold ratio

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ABSTRACT

An up-flow anaerobic sludge bed (UASB) reactor was operated for five months to treat domestic wastewater contaminated by propylene glycol -main component of Type 2 aircraft deicing fluid (ADF)- at threshold ratio based on optimization study with batch reactors [i.e., operated at externally included ADF = 0.83–4.20%]. Biogas yields up to ~0.4 m^3/kg $COD_{influent}$ was achieved, however drastically reduced along with total chemical oxygen demand (tCOD) removal at ADF>1.20% and indicated strong inhibition. Hence, the UASB reactor was fed at ADF = 1.05% [$tCOD_{influent}$ = 8930 ± 2100 mg/L] and yielded >80% tCOD removal on average. Next generation sequencing (NGS) findings also revealed that *Firmicutes* and *Bacteroidetes* had the highest ratios of relative abundances in biomass sample taken at the last operating day; hence co-existence of these phyla played significant role in glycol removal with *Synergistetes* and *Thermotogae* bacteria whereas *Methanobacterium* and *Methanoculleus* archaea increased remarkably.

1. Introduction

Air transportation is the most preferable alternative due to the increase in world's population. In Turkey, the airline transportation has been highly preferred by the passengers and annual flight number has increased remarkably compared to previous decade (e.g., total number of domestic and international flights was approximately 790,000 in 2009 and nearly doubled to ca. 1,550,000 in 2019) (TUIK, 2020). With respect to this increasing interest in air transportation; aircraft deicing fluids (ADFs) have been have been applied intensely at the special places namely the 'deicing pads' to remove ice, frost and snow accumulated on the outer surfaces of the aircrafts in order to ensure flight safety in winter conditions when heavy snowfall occurs at the airports around the world. Since deicing operations involve the application of chemicals for preventing initial icing or further icing (anti-icing) and for the removal of (and preventing) ice from airfield pavement (runways, taxiways, aprons and ramps); these centralized deicing pads are generally located near terminals and gates, along taxiways serving departure runways, or near the departure end of runways. Hence, one or a combination of all of these locations might be used by the airport for deicing/anti-icing purpose (Switzenbaum et al., 2001; Delasanta, 2010). Four types of ADFs with the main component namely glycol (i.e., ethylene or propylene), have been used during deicing applications. The propylene glycol substances contain a similar family of synthetic organic molecules -consist of the 1, 2-propanediol substance (monopropylene glycol, MPG) and its dimer (dipropylene glycol, DPG), trimer (tripropylene glycol, TPG) and tetramer (tetrapropylene glycol, TePG) forms-that have widespread application and huge production volumes all over the world. ADFs are produced in accordance with the standards set by the International Organization for Standardization (ISO) and aircraft size is the crucial factor while selecting the ADF type. Among the ADFs, Type 1 ADF consists of 8% water, 90% glycol and less than 2% chemical substances whereas Type 2 ADF consists of 33% water, 65% glycol and 2% alcohol-based chemicals with a structure preventing ice formation. The main difference between these fluids is their viscosity (EPA, 2000; West et al., 2014; Elreedy et al., 2017).

During ADF application, the fluid is sprayed on the surface of the airplane after diluted with hot water and mixes with precipitation on the ground as well as with other chemicals (i.e., including fuels, lubricants, solvents, and metals) found on airport pavements and eventually enters the airport's storm drain system. It is known that the storm drains are discharged directly to waters with no treatment or to the nearest municipal wastewater treatment plant (WWTP) at certain ADF ratios (i.e., runoff from deicing activity at airports consists of 1–40% ADF-contaminated water next to grit and sand). However, ADF-contaminated wastewater leads to environmental pollution (e.g., decreased amount of

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dissolved oxygen in the receiving waters etc.) and/or human health risks (e.g., respiratory health problems, skin and eye irritation etc.) in the case of its direct discharge (EPA, 2000; Vasilyeva, 2009; Delasanta, 2010). Moreover, additive chemicals have been identified (e.g., surfactants, viscosity enhancers, buffers and corrosion inhibitors) during toxicity assays (Cornell et al., 2000; Pillard et al., 2001). Among the corrosion inhibitors, benzotriazole derivatives have been widely added into aircraft deicing fluids and decreased biogas production was reported during anaerobic treatment while feeding the system with ADF having more than 300 mg/L 4-, and 5-, methyl benzotriazole (MeBT) (Gruden et al., 2001). However, determination of an approximate ratio of acute:chronic toxicity threshold concentrations from the available ecotoxicological datasets for these substances have been reported as impossible (West et al., 2014). Although the empirical data on these substances clearly showed that acute and chronic effects are not expected to occur for typical and recommended use and disposal of the products containing them; Delasanta (2010) reported that runoff from deicing activity at airports might include up to 40% ADF-containing water and discharge of this pollutant without proper treatment is of serious environmental concern given its persistency in the aqueous environment (Elreedy et al., 2017).

Accordingly, the Environmental Protection Agency (EPA) has proposed technology-based effluent guidelines and new source performance standards which were specific to the runoff resulting from airport deicing operations, under the protection of the Clean Water Act for more than a decade. Besides, according to the EPA regulations; large airports need to gather, store, treat (with biological processes or distillation), and either dispose of the consumed ADF properly to a sewer system or recover for reuse. Hence, in order to fulfill the requirement of the best available technology; one of the following two approaches should be considered in order to decrease the discharges of ADF wastewater without treatment: capturing and collecting a specified proportion (20–60%) of available ADF or treating the gathered ADF according to end-of-pipe discharge limitations (EPA, 2000; Delasanta, 2010). For example in Sweden; Lönnqvist et al. (2015) reported that there was an attractive potential in the residues deriving from the use of propylene glycol for preventing icing at some airports where more than 80% of 1442 tons were collected by a drainage system in 2012 in order to meet environmental requirements.

With the risk of the spent ADFs reach to the municipal WWTPs via surface runoff through raining and snow melting; lower biological treatment efficiencies could be obtained due to the increased tCOD values as well as higher organic loading rates (OLRs) which are not compatible with design parameters. Therefore, ADF ratio should be at optimum/appropriate rate inside the wastewater in order not to reduce the performance of the biological treatment processes. Preliminary studies reported that anaerobic systems provide more effective and feasible treatment performance for the wastewaters like ADF-contaminated waters at high OLRs especially when applied at the source of the pollution (i.e., on-site or decentralized treatment) (Lopez et al., 2009; Switzenbaum et al., 2001; Schoenberg et al., 2001; Goktas et al., 2019). Among the anaerobic systems, up-flow anaerobic sludge bed (UASB) reactor occupies an important place with the high removal efficiency, less space requirement, biogas potential as a renewable energy source, and relative resistance to inhibitory compounds (Gomec et al., 2005; Gomec, 2010; Elreedy et al., 2017; Goktas et al., 2019).

Despite of widespread presence in the environment, microorganisms involved in biodegradation of propylene glycol substances have not been fully specialized (West et al., 2014). Hence, the co-existence of different microbial groups and the predominant species should be investigated in such biological reactors using high-throughput analytical methods such as next generation sequencing (NGS) (Gomec et al., 2008; Chen et al., 2016; Yangin-Gomec et al., 2017; Pekyavas and Yangin-Gomec, 2019). Accordingly, the predominant microbial communities at high glycol ratio would help to enhance organic removals and biogas recovery during on-site treatment especially at the airports. However, within our knowledge, no study is available focusing on predominant microorganisms at a UASB reactor operated at threshold propylene glycol concentration.

This study aimed to investigate anaerobic treatability of domestic wastewaters contaminated with propylene glycol (i.e., main component of Type 2 ADF) at certain ratios by an up-flow anaerobic sludge bed (UASB) reactor based on the findings of a batch optimization study. Within the scope of this study, bacterial and archaeal communities in the biomass samples [i.e., taken when organic loading arising from ADF content in domestic wastewater was increased] were also identified in order to understand the impact of propylene glycol especially at threshold concentration.

2. Materials and methods

2.1. Aircraft de-icing fluid, domestic wastewater, and inoculum sources

Concentrated Type 2 ADF [propylene glycol: 88%; viscosity: 20 cp; pH: 8.7–9.7; tCOD: 615000 mg/L] was taken from an airport in Istanbul. The used ADF was a propylene glycol ($C_3H_8O_2$)-based fluid consisting of water, corrosion inhibitors, wetting agents, and dye.

Raw domestic wastewater was obtained (i.e., samples were stored at +4 °C) from an airport at which several package wastewater treatment units were available during the experimental study. Domestic wastewater samples were taken from two certain points in order to get relatively stable influent wastewater characteristics [i.e., 7.34 ± 0.11, pH; 551 ± 202 mg $CaCO_3$/L, alkalinity; 653 ± 324 mg/L, tCOD, 290 ± 95 mg/L, sCOD; and 519 mg/L, TSS; 94 mg/L, VSS; 64 mg/L, TAN; and 10 mg/L, Ortho-PO_4^{3-}]. On the other hand in order to inoculate the batch reactors; the granular anaerobic sludge from an industry treating paper wastewater was used as the seed after crushed by a kitchen grinder. However, the UASB reactor was seeded with some amount of this original anaerobic granular sludge (800 mL) and also with the sludge already adapted to Type 2 ADF in the batch study (500 mL) yielded 52% volatile content of total solids (TS) of about 8.5 g/L.

2.2. Anaerobic reactors and operating conditions

For the batch study, N_2-flushed, 1 L glass flasks were used. In the reactors, domestic wastewater (600 mL) and the seed sludge (100 mL) (v/v = 1/6) were added with the following ADF ratios; 0.83%, 1.01%, 1.2%, 2.24%, 2.83%, 3.08%, 3.45%, and 4.20% (i.e., the measured ADF ratios inside the bioreactors based on propylene glycol analysis as explained in the below section 2.3 at start-up). In addition to these reactors, a control flask was also run including only domestic wastewater and inoculum sludge. Accordingly, OLR varied in the range from ca. 0.2–2.0 kg/m^3.d. Determination of the initial ADF ratios in the flasks was done according to the results of a previous batch optimization study. Moreover, a sample was taken from the ADF storage tank of the investigated airport and tCOD concentration was determined more than 90000 mg/L. This value corresponded to ADF-contaminated waters having about 10% Type 2 ADF (Goktas et al., 2019). However, initial propylene glycol ratios inside the batch reactors were determined slightly higher than the targeted values which might be due to already available propylene glycol in raw domestic wastewater sample originating from the detergents used at the airport.

On the other hand, a laboratory scale UASB reactor [$V_{effective}$, 6.45 L; H, 1.0 m; D, 90 mm] was operated at 35°C in semi-continuous mode with the daily prepared substrate (i.e., externally included Type 2 ADF into raw domestic wastewater to provide targeted influent propylene glycol ratio). In accordance with the aim of this particular study, the UASB reactor was operated in two periods by Type 2 ADF-contaminated wastewater which included propylene glycol contents at 0.45 ± 0.22%

for about two months and 1.05% on average for about three additional months (i.e., between 68-154 d). At start-up; the UASB reactor was operated at low OLR (i.e., 0.2 kg/m^3.d at ADF = 0.45%) corresponded to a HRT of 13 d due to possible inhibitory impact of the ADF in domestic wastewater. However, according to the results of the optimization study with the batch system; ADF concentration was almost doubled in domestic wastewater (ADF = 1.05%) after a 67-d of operation that also led to a sudden increase in the influent tCOD concentration and indicated an increase in the applied OLR (i.e., 0.6 kg/m^3.d) as well. The steady-state condition was observed between 70 and 154 days and the UASB reactor was operated with the same HRT in order to keep the stability of the system. Respective influent substrate characteristics for two periods were as follows: pH, 7.23 ± 0.15 and 7.33 ± 0.15; Alkalinity, 558 ± 273 and 923 ± 235 mg $CaCO_3$/L; TAN, 54 ± 7 and 81 ± 11 mg/L; TSS, 382 and 944 mg/L; sCOD, 2355 ± 982 and 7148 ± 1322 mg/L, and tCOD, 2898 ± 879 and 8931 ± 2101 mg/L according to the fluctuation in the domestic wastewater composition (Engiz, 2018).

2.3. Experimental procedure

The performance of the UASB reactor was monitored with the following parameters; alkalinity, total COD, soluble COD, and total suspended solids (TSS) concentrations (APHA, 2005). HI 2211-02 HANNA Model pH meter was used for the pH measurements (Sigma-Aldrich Chemie GmbH, Germany). Dichromate closed-reflux method was used and COD concentrations were measured by HACH DR/2010 spectrophotometer (Hach Company, U.S.). Daily biogas generation in batch flasks was monitored by a manometer (i.e., Lutron PM-9107) in the headspaces of the reactors before the produced gas pressures were released by injection needles. Then the biogas volume (mL) in each reactor was calculated under the standard conditions. Daily biogas production in the UASB reactor was measured using the Ritter MilliGas Counter 770991000 model gas meter (Ritter, Germany). The Bellingham + Stanley Abbe type refractometer (Model 60/70) (Bellingham + Stanley Ltd., United Kingdom) was used for propylene glycol measurements. For microbial community analysis, the isolation of total DNAs were done and 16S rRNA genes were sequenced according to the Illumina MiSeq NGS method of the V4–V5 hypervariable region (Illumina, Inc., San Diego, CA, USA) as reported by Yangin-Gomec et al. (2017).

3. Results and discussion

3.1. Reactor performances at increased propylene glycol concentrations

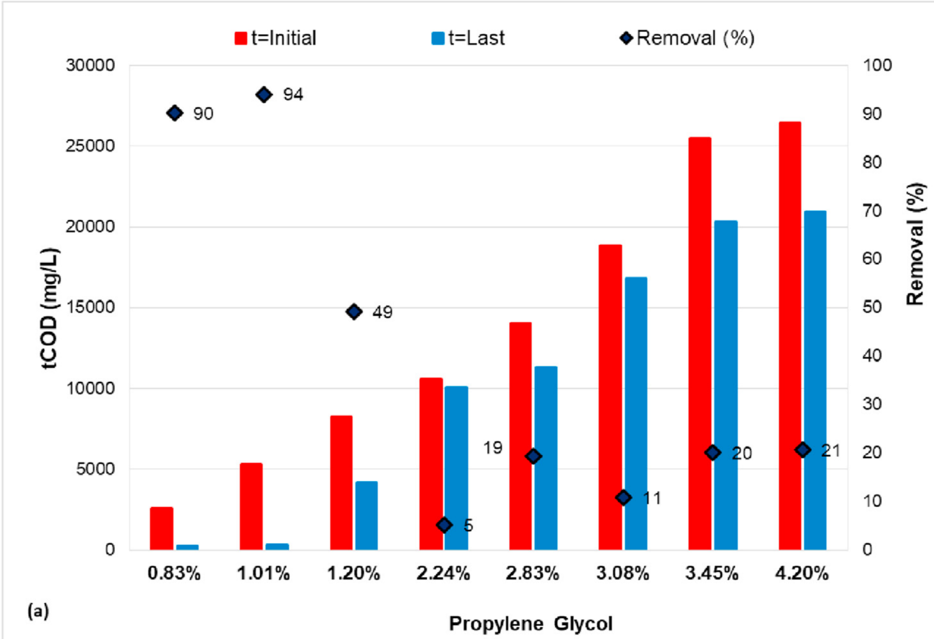
Depending on the inoculum type and incubation condition; the propylene glycol substances are rapidly biodegraded in air, water, soil, and sediment. Moreover, due to lack of bioaccumulation potential; these substances yield low environmental harm (West et al., 2014). Schoenberg et al. (2001) also reported complete anaerobic degradation of propylene glycol- and ethylene glycol-based ADFs. In this study, anaerobic treatability of Type 2 ADF-contaminated domestic wastewater at different ratios was investigated using anaerobic systems operated as batch (i.e., ADF = 0.83–4.20%; OLR = 0.2–2.0 kg/m^3.d) and semi-continuous (i.e., ADF = 0.45% and ADF = 1.05%; OLR = 0.2 and OLR = 0.6 kg/m^3.d) modes both at mesophilic temperature (35 °C). Although Delasanta (2010) reported that runoff from deicing activity at airports might include up to 40% ADF-containing water with grit and sand; ADF ratio was below 4.5% in this particular study based on a previous optimization study (Goktas et al., 2019).

In the batch study, inhibition of the threshold ADF ratio on the microbial cultures was determined based on the biogas yields during operation (74 d). The maximum biogas yield was determined as ca. 0.40 m^3/kg $sCOD_{influent}$ in the flasks having 1.20% ADF (Table 1). However, when sCOD removals were also taken into account; 1.01% ADF represented the highest removal (97%) including the control reactor. This might be due to the fact that the reactors with more than 1.20% ADF showed remarkable inhibition on bacterial and archaeal cultures with substantial reductions in biogas yields and COD removals. Therefore, it was observed that high biogas productions occurred between 56% and 97% COD removals at propylene glycol ratios in the range from 0.83% to 1.20% during anaerobic batch treatment of Type 2 ADF-contaminated domestic wastewater (Figure 1a). However, biogas yields decreased dramatically in the reactors where ADF ratios were ≥1.20% which might be due to substantial increase in volatile fatty acids (VFAs) caused inhibitory effect on methanogenic archaea at pH values lower than the optimum range (i.e., between 6.7 and 7.4) (Gomec et al., 2002). Total VFA concentrations increased up to more than 5500 mg/L at the final operating day when Type 2 ADF was 2.7% in a similar study by Goktas et al. (2019). Although Watari et al. (2015) reported much higher OLRs (i.e., up to 13 kg/m^3.d) are applicable for high-rate anaerobic reactors; high propylene glycol ratios caused significant reduction in biogas production as well as in soluble organic matter removal (i.e., down to ca. 0.03 m^3/kg $sCOD_{influent}$ and 11%, respectively) in batch systems. This finding was also proven by low pH values in the flasks having more than 1.20% propylene glycol so that pH could not be recovered following the acidogenesis phase at initial incubation times (i.e., 7th, 14th, and 21st days of operation) and the measured pH values were all lower than 6.0 indicating inhibition of the methanogenic archaea. However, in the reactor having lower than 1.20% ADF; although pH dropped from 7.36 to 6.13; it was measured as 7.39 at the last operating day indicating sufficient consumption of the produced VFAs by the methanogens. Goktas et al. (2019) also reported similar VFA results with reduced pH values in the batch reactors treating propylene glycol-containing wastewater and no total VFA removal could be obtained at more than 1.8% however total VFAs were mostly consumed under this ADF content. Besides, as for the methane content of the biogas; approximately 60% methane was measured which was lower than the methane content of a similar study by Jin et al. (2016). Here, biogas yield was reported as approximately 0.3 m^3/kg $sCOD_{feed}$ with high methane content (i.e., 77%) during anaerobic treatment of propylene glycol-contaminated wastewater at high concentrations.

In this context, after the determination of the threshold propylene glycol ratio in the domestic wastewater in the batch study, an UASB reactor was operated at semi-continuous mode for about 5 months in the second part of this study. During the start-up of the UASB reactor, some of the sludge taken from the batch reactors was used as the inoculum in order to operate the bioreactor with the sludge already adapted to Type 2 ADF. Nevertheless, the same original granular methanogens were also added into the bioreactor at start-up. The reactor was operated at the ADF ratio of 0.45 ± 0.22% in the domestic wastewater in the first period between 0-67 d while ADF increased to 1.05% in the second period between 68-154 d. Although high biogas productions occurred during the aforementioned anaerobic batch study with similar Type 2 ADF ratios; biogas production from the UASB reactor indicated relatively lower

Table 1. Biogas yields in the batch reactors at increased propylene glycol ratios.

Parameter	Unit	Propylene Glycol (%)								
		Control	0.83	1.01	1.20	2.24	2.83	3.08	3.45	4.20
Influent sCOD	mg/L	240	2404	5267	7483	10308	13675	18354	23708	26125
Biogas Yield	m^3/kg $sCOD_{inf.}$	0.65	0.40	0.28	0.37	0.09	0.09	0.03	0.08	0.09



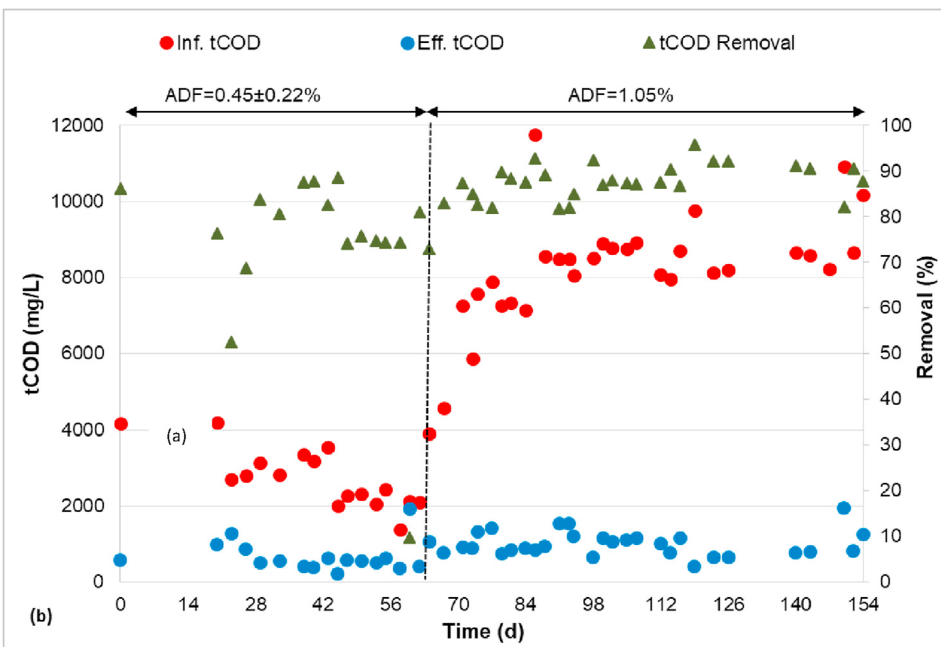
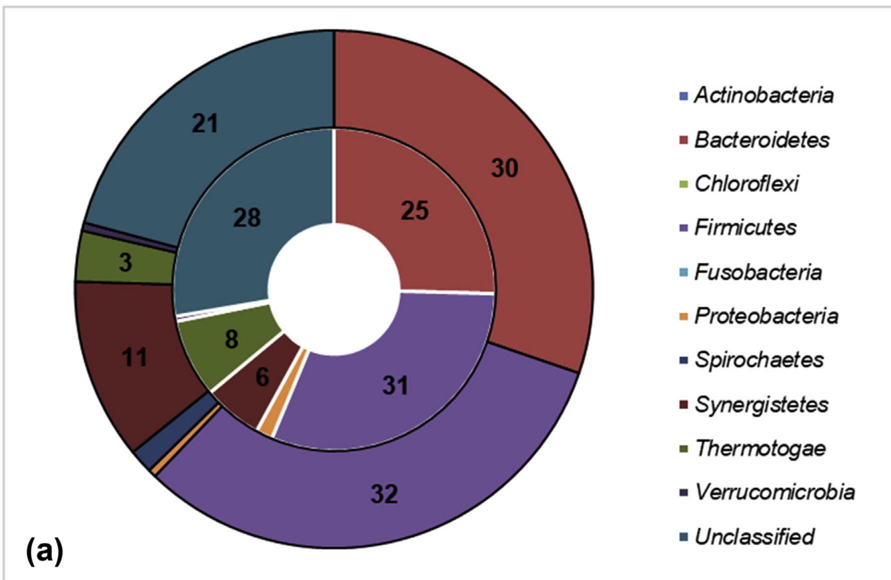


Figure 1. tCOD profile and removal rates at increased propylene glycol ratio; (a) batch reactors (b) UASB reactor.

yields (data not shown) however substantial COD removals could be achieved during the study (i.e., average tCOD and sCOD removals were in the range from 84% to 90% at ADF = 1.05%) (Figure 1b). Following the termination of the UASB reactor's operation; it was understood that the special gas-solids-liquid separator at the top of the UASB reactor was slightly lopsided and unfortunately much lower biogas generation could be achieved due to this incident. Darlington and Kennedy (1998) also studied the removal of ADF-included wastewater in a UASB reactor system and they reported from 85% to 95% COD removals at an OLR of ca. 10 g COD/L/d; however COD removal rates decreased to 70% at increased OLR up to ca. 40 g COD/L/d. Moreover, compatible with the results of this particular study; Watari et al. (2015) also reported ca. 91% COD removal with 82% methane recovery at a high-rate anaerobic reactor fed by industrial wastewater including 8% ethylene- and 2% propylene-glycol. Besides, Marin et al. (2010) demonstrated 75% sCOD removal rate and 0.30 $L/gCOD_{removed}$ methane yield at mesophilic anaerobic baffled reactor fed with synthetic wastewater contaminated with ADF. In order to indicate the importance of glycol ratio in the treated wastewater; Tham and Kennedy (2004) also operated a high rate UASB reactor with a synthetic wastewater having ADF ratios in the range from 0.8% to 1.6% and reported more than 97% COD removals. According to pH measurement, on the other hand; average pH value in the influent was 7.23 ± 0.15 whereas it was 8.08 ± 0.26 in the effluent in the first period (at ADF of 0.45 ± 0.22%) whereas respective average pH values in the influent and in the effluent were 7.33 ± 0.15 and 8.04 ± 0.19 in the 2nd period (at ADF of 1.05%). On the other hand, alkalinity results indicated that respective average values in the influent and effluent were 558 ± 273 and 929 ± 297 mg $CaCO_3$/L in the first period whereas 923 ± 235 and 1115 ± 253 mg $CaCO_3$/L in the second period. Liu et al. (2009) also called attention to alkalinity measurement such that VFA/alkalinity ratio played significant role on the microbial cultures in the bioreactors especially when operated at high OLRs.

On the other hand, next to initial samples of the batch reactors; propylene glycol ratios were also measured from the final samples (t = 74 d) and refractometer results indicated up to 50% glycol removals (i.e., in the flasks having ADF ratios ≤1.20%). This finding was also proved by the refractometer results of the UASB reactor study so that ca. 46% glycol removal could be obtained on average when the influent ADF ratio was 1.05% between 68 d and 154 d of operation. However, almost no glycol removal could be achieved when the ADF ratio was above threshold level which was compatible with sharp COD removals as well as with the unrecoverable pH values (e.g., pH reduced to 5.71 at t = 74 d from 7.12 at t = 0 d when ADF ratio was ≥2.83%) in the batch reactors.

In this study, a conceptual design of one module UASB reactor [according to the findings of this study while using the reported aircraft numbers and de-icing application data of previous years by TUIK (2020)] was also done and it was calculated that ADF-contaminated domestic wastewater could be treated if feeding and operating conditions are carefully monitored with reasonable energy production [i.e., ~920 m^3/d biogas production equivalent to ca. 4340 kWh total energy (electricity and heat) generation at about 0.5% ADF-contaminated domestic wastewater] reflecting successful on-site management of de-icing fluids at airports. In fact, such systems could be operated at much higher OLRs allowing lower HRTs that would provide better ADF management at full-scale plants. In relation to energy requirement; Lönnqvist et al. (2015) also reported for countries where high amounts of deicing fluids are used (e.g., Sweden); the technical development of and policy support for other renewable transport fuels will be the main foci. Accordingly, although the vehicle gas potential could meet the demand by 2020, it



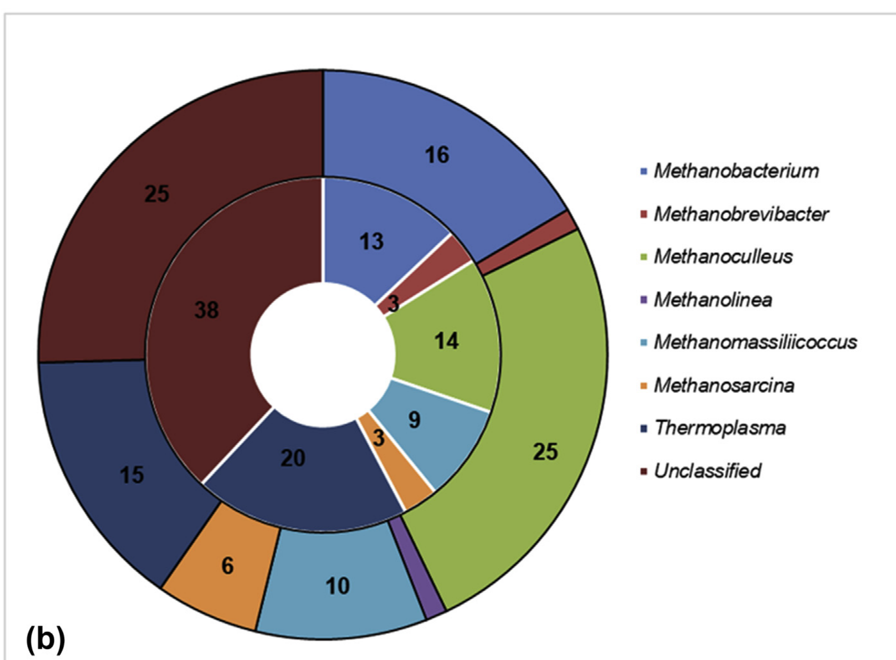


Figure 2. Comparative relative abundance (%) of microbial cultures; 1st period [t = 0–67 d; ADF = 0.45 ± 0.22%] (inner doughnut) and 2nd period [t = 68–154 d; ADF = 1.05%] (outer doughnut). (a) bacteria at phylum level (b) archaea at genus level.

might cover only 50% of the demand by 2030 (i.e., the vehicle gas potential estimation is 597 GWh, of which 72% derives from residues and the rest from energy crops) in Stockholm County and biogas from neighboring regions would hence be required to fulfill this demand (Lönnqvist et al., 2015).

3.2. Microbial community at increased propylene glycol concentrations

In this study, Illumina Miseq NGS analysis was performed by taking two sludge samples from the UASB reactor at gradually increased ADF ratios during about a five-month operating period. Although *Proteobacteria* (19%), *Actinobacteria* (16%), *Bacteriodetes* (10%), *Firmicutes* (9%), *Synergistetes* (8%), and *Thermotogae* (4%) as well as *Euryarchaeota* (10%) was the predominant microbial consortium in the inoculum sludge (data not shown); *Firmicutes* and *Bacteriodetes* meaningfully increased by the end of a 154-d operation. Therefore, results indicated that the main dominant bacteria during the operation were detected as *Firmicutes* and *Bacteriodetes.* Between these two phyla; *Firmicutes* increased slightly from ca. 31%–32% whereas *Bacteroidetes* increased from about 25% to 30% at average ADF ratios of 0.45% and 1.05%, respectively (Figure 2a) as operation continued. Hence, when influent ADF concentration was 1.05% with respective average tCOD and sCOD values as 8931 $\pm$ 2101 and 7148 $\pm$ 1322 mg/L; the relative abundances of these bacteria almost tripled compared to inoculum and this remarkable shift indicated that glycol reduction was substantially actualized by *Firmicutes* and *Bacteriodetes* in the system. Noteworthy increase in these phyla was also confirmed by the determination of microorganisms that belonged to respective classes of *Clostridia* (30%) and *Bacteroidia* (26%) at the highest level when influent average ADF was 1.05% in domestic wastewater. It was reported that these bacterial phyla have been also detected as the dominant ones at anaerobic reactors and *Bacteroidetes* is especially responsible from biomass granulation at rapidly biodegraded organic carbon rich environments (Lucas et al., 2015; Cao et al., 2016). In accordance with the study by Doloman et al. (2017) who detected similar major phyla as *Bacteroidetes* and *Firmicutes* as well as *Synergistetes* and *Thermotogae* during anaerobic treatment of high organic content waste by an UASB reactor. Similarly; the third abundant phylum was *Thermotogae* (8%) in this particular study which have been also identified during the most likely acidogenic-methanogenic phase of anaerobic processes and have been isolated from several environments such as oil reservoir and wells etc. According to previous studies; the unique order of *Thermotogales* was also found in mesothermic, anoxic, hydrocarbon-rich environments (Nesbo et al., 2010; Bhandari and Gupta, 2014; Doloman et al., 2017). In this context, according to EPA results of the collected samples from a variety of airport wastewater storage facilities; semi-volatile compounds (di-n-butyl phthalate and n-dodecane) and other pollutants such as Polycyclic Aromatic Hydrocarbons (PAHs) and Polychlorinated Biphenyls (PCBs) might be also detected in the raw wastewater from aircraft deicing/anti-icing operations and in direct discharge of ADF-contaminated storm water (EPA, 2000; Sulej et al., 2011). Moreover, in accordance with the microbial findings of this particular study; Hania et al. (2011) also reported that *Thermotogales* reduce the elemental sulphur to sulphide particularly from the anaerobic enriched cultures on polychlorinated biphenyl compounds inoculated with sediments and this order was also detected in microbial communities degrading chlorophenols, polycyclic aromatic hydrocarbons and dimethyl phthalate. Hence, their ecological role within microbial consortia enriched on halogenated aromatic compounds (e.g. polychlorobiphenyl compounds) is very crucial. On the other hand, *Synergistetes* nearly doubled its relative abundance; however *Proteobacteria* -a bacterial phylum reported as denitrifying bacteria-were no longer detected in the biomass sample taken on 154-d of operation which might be due to possible inhibition depending on the concentration of glycol available in domestic wastewater (Chen et al., 2019). Watari et al. (2015) also reported one of the predominant microbial groups as the phylum *Synergistetes* next to the phyla *Proteobacteria, Euryachaeota, Bacteroidetes,* and *Firmicutes*. Another study by Swiatczak et al. (2017) also observed that the acetogenic *Synergistes* sp. closely collaborated with methanogens (i.e., *Methanomicrobia*) by transferring hydrogen between species and this phylum favored biogas yield in anaerobic reactors.

Accordingly, methanogenic archaea under the phylum *Euryarchaeota* which were involved in mesophilic glycol degradation were also investigated in this study. NGS results indicated that three dominant archaeal classes were detected namely *Methanobacteria* (32%), *Methanomicrobia* (30%)*,* and *Thermoplasmata* (16%) (methylotrophic methanogens) were identified at class level for ADF = 1.05%. According to the results of the archaeal sequence of the biomass sample taken at day 154; the dominant genus shifted to *Methanoculleus* from *Thermoplasma*. In this context, *Methanoculleus* increased from about 14% for ADF = 0.45% to ca. 25% for ADF = 1.05% indicated the most abundant genus together with *Methanobacterium* (~17%) as operation continued (Figure 2b). Swiatczak et al. (2017) also reported that *Methanoculleus* and *Methanobacterium* were the predominant genera in anaerobic digesters operated at high loadings. Hence, by the identification of predominant species; individual bacterial and archaeal responses to inhibitory substances at threshold ratios could be better understood in the bioreactors while treating wastewaters contaminated with special pollutants like propylene glycol.

4. Conclusions

The results revealed that propylene glycol-containing domestic wastewater up to a certain ratio had high level of anaerobic biodegradability and a substantial quantity of biogas could be obtained this way. Accordingly, high sCOD removal (97%) with ~0.3 $m^3/kg\ sCOD_{influent}$ biogas yield was obtained at ~1.0% ADF, but strong inhibition was observed when glycol was increased. Hence, although mesophilic anaerobic treatment was a promising approach; bioenergy production was mainly dependent on propylene glycol ratio in wastewater. Moreover, initial glycol had also crucial impact on predominant bacterial and archaeal phyla (respective changes from Proteobacteria to *Firmicutes* and from *Methanocarcina* to *Methanoculleus* at increased ADF).

Declarations

Author contribution statement

Cigdem Yangin-Gomec: Conceived and designed the experiments; Analyzed and interpreted the data; Wrote the paper.

Gizem Engiz: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data.

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Data availability statement

Data associated with this study has been deposited at =https://tez.yok.gov.tr/UlusalTezMerkezi/TezGoster?key=T1mWGp9MngYYkCSgiJvtVrD7fs2ufwfxUR6K-W0Dk0AHhk7OHRMye5RhaMPv0IRd under the accession number 541857.

Declaration of interests statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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