

MDPI

Article

Anaerobic Treatment of Food Waste with Biogas Recirculation under Psychrophilic Temperature

Tafannum Torsha and Catherine N. Mulligan *

Department of Building, Civil and Environmental Engineering, Concordia University, Montreal, QC H3G 1M8, Canada; tafannumtorsha24@gmail.com

* Correspondence: mulligan@civil.concordia.ca

Abstract: Food waste has emerged as a pressing concern, and thus advanced techniques to valorize food waste into nutrition rich materials as well as renewable energy are highly important. The exceptional biodegradability of food waste renders it a highly suitable substrate for anaerobic treatment. This leads to energy production and a reduction in the carbon footprint. Nevertheless, in frigid territories like Canada, the conventional mesophilic anaerobic digestion at 30-40 °C can require substantial amounts of energy. Consequently, this study introduces a new approach to treat food waste at psychrophilic temperatures (1-20 °C). Lower temperatures can negatively impact cellular processes during anaerobic treatment, rendering substrates less accessible to microscopic organisms. To address this challenge associated with lower temperatures, the study introduces an innovative biogas recirculation strategy. The primary objectives of this study are to assess the viability of anaerobic treatment for food waste at psychrophilic temperatures and to investigate the effectiveness of reintroduction of the produced biogas to the anaerobic system in enhancing biomethane generation and stability of the system. Batch experiments were conducted on food waste in various assessments, both with and without biogas recirculation. The outcomes revealed a methane concentration ranging from 68% to 93% when biogas recirculation was employed, whereas without this technique, methane concentration varied between 10% and 45%. Moreover, with biogas recirculation, the reduction in volatile solids reached a maximum of 92%, and there was an 82% decrease in chemical oxygen demand. In conclusion, the utilization of the recirculation of biogas at the psychrophilic temperature range enhanced biomethane production and reduction of volatile solids and chemical oxygen demand. This study underscores the potential of employing anaerobic treatment with reintroduction of produced biogas into the system in cold regions as an economically viable and sustainable choice for treating food waste with nominal energy consumption.

Keywords: anaerobic treatment; food waste; psychrophilic conditions; biogas recirculation



Citation: Torsha, T.; Mulligan, C.N. Anaerobic Treatment of Food Waste with Biogas Recirculation under Psychrophilic Temperature. *Waste* 2024, 2, 58–71.

https://doi.org/10.3390/waste2010003

Academic Editors: Sergey M. Frolov and Vladimir S. Arutyunov

Received: 10 December 2023 Revised: 16 January 2024 Accepted: 19 January 2024 Published: 23 January 2024



Copyright: © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https://creativecommons.org/licenses/by/4.0/).

1. Introduction

In recent times, there has been a substantial rise in food waste (FW) due to population growth and the global economy's expansion. Almost 33% of the global food production for human consumption is discarded along the food supply chain [1,2]. The composition and physicochemical characteristics of FW exhibit variations based on factors such as the country of origin, dietary habits, and cultural and economic influences [3]. FW typically consists of 70–80% water and exhibits high biodegradability [4]. However, the disposal of FW through composting, landfilling, or incineration can have detrimental environmental consequences and contribute to global warming. FW constituted the primary component of solid waste in US municipalities (approximately 21% of all materials) in 2012 [5]. The US Environmental Protection Agency (USEPA) projected that food waste was 12.7% of the total waste disposed of in 2014 [5]. Landfilling of FW leads to a loss of its energy potential, coupled with fugitive releases that result in greenhouse gases (GHG). Research by Clercq et al. demonstrated that significant quantities of the greenhouse gases carbon dioxide (CO₂) and

methane (CH₄) are generated when food waste is discarded in landfills [6]. These emissions exacerbate global warming as methane is an especially potent greenhouse gas, possessing a greenhouse effect 25 times more potent than CO2. It has been estimated that globally food loss and waste collectively contribute to 6.7% of total annual anthropogenic greenhouse gas productions [2]. Another significant consequence of food waste for the environment is related to its disruption of the biogenic cycles of nitrogen and phosphorus, both essential components of agricultural fertilizers [7]. Given that FW represents an inefficient use of limited resources such as land, water, and fertilizers and contributes significantly to environmental degradation, the proper treatment and management of food waste have emerged as a top priority in numerous nations across the globe [8]. Anaerobic treatment of FW is emerging as a highly promising and potentially cost-effective option for producing renewable energy, protecting the environment, and managing waste [9-11]. Numerous research studies have already acknowledged anaerobic treatment as an environmentally friendly and easily implementable technology for transforming organic materials into renewable sources of energy [12,13]. FW, with its high energy value and moisture content, proves to be an excellent substrate for anaerobic treatment [14,15].

Anaerobic treatment is a biochemical process wherein various groups of microorganisms engage in a series of complex reactions and intermediary steps to convert insoluble and intricate organic substances into simpler molecules, including CH₄ and CO₂ [16]. The protein, carbohydrate, and lipid components found in food waste undergo fermentation to yield volatile fatty acids (VFAs) and long-chain fatty acids (LCFAs), which are subsequently transformed into acetate and hydrogen gas—precisely what methanogens require for their metabolism [17]. While anaerobic technology offers numerous advantages, such as reduced greenhouse gas (GHG) releases and the production of high-quality renewable fuels, certain limitations, such as extended retention times, substantial initial investment expenses, and the need for precise control of critical parameters (temperature, pH, alkalinity, feeding rate) impede its widespread application [9].

Temperature significantly influences the overall performance of acid-forming as well as methanogenic microbes and exerts significant influence in anaerobic treatment. Anaerobic treatment is typically classified into three categories depending on operating temperatures: psychrophilic (≤ 20 °C), mesophilic (30–40 °C), and thermophilic (≤ 50 °C) [18]. The climatic condition in Canada is more suitable for psychrophilic anaerobic treatment over mesophilic alternatives. Despite the majority of anaerobic treatment research being conducted within thermophilic (\leq 50 °C) or mesophilic (30–40 °C) treatment conditions, evidence has also been found indicating that biomethane generation takes place at a lower operating temperature (<20 °C), facilitated by psychrophilic archaebacteria. Several studies have indicated a higher diversity of methanogenic archaebacteria at psychrophilic temperatures compared to mesophilic conditions [19]. Therefore, opting for anaerobic treatment within the mesophilic temperature condition would need a substantial quantity of energy to sustain the anaerobic digester at elevated operating temperatures, which would reduce the net energy yield and escalate operational costs [20]. Ample experimental evidence has demonstrated that anaerobic systems designed for low-temperature treatment yield substantial methane production and excellent chemical oxygen demand (COD) removal efficiency, contrary to the conventional notion that higher methane generation occurs within the mesophilic temperature range [21]. Considering Canada's climate, psychrophilic anaerobic treatment emerges as an efficient approach for food waste treatment with the least external energy demand.

This research introduces an innovative approach involving biogas recirculation to employ anaerobic technology for food waste treatment under psychrophilic temperatures. Thus, the fundamental aim of this work is to determine the efficacy of the biogas recirculation technique in facilitating prolonged psychrophilic conversion of food waste and methane production.

2. Experimental Materials and Methodology

2.1. Characteristics of Food Waste and Inoculum

The food waste sample was generated from weekly collections of food scraps in the residential district of downtown Montreal for four consecutive months (from January to April). The predominant components of this food waste included rice, bread, peas, onions, potatoes, salt fruits and vegetable peels, coffee grounds, tea bags, eggs, eggshells, and non-degradable materials. No significant variation in food waste composition was observed due to seasonality. Figure 1 presents an approximate breakdown of the composition of the collected food waste sample.

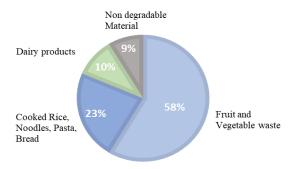


Figure 1. Analysis of the composition of a sample of food waste.

Food waste was collected using a door-to-door collection method. After the waste collection process, the gathered waste underwent a thorough sorting and segregation procedure. Additionally, non-degradable waste was manually separated from the collected waste sample. To begin, the sample food waste was initially created by manually blending assorted food scraps, after which the food scraps were further processed with 100 mL of water in a food processor until they formed a paste-like consistency. Subsequently, the food waste was stored in a refrigerator at a temperature of 1 degree Celsius until it was ready for use. Both the food waste and anaerobic inoculum underwent moisture content, pH, total solids (TS), volatile solids (VS), and chemical oxygen demand (COD) analysis in duplicate before the start of the experiments and upon completion of the treatment period. The initial and final measurements of TS, VS, and COD were utilized to calculate the reduction in solids and the efficiency of COD removal following anaerobic treatment. All assessments were conducted in accordance with established standard methods [22]. The physicochemical properties of the food waste sample utilized in this investigation can be found in Table 1.

Table 1. Physicochemica	l attributes of three different food waste samples.
-------------------------	---

Attribute	Value
Total Solid%	28.2 ± 1.3
Volatile Solid%	27.13 ± 0.94
рН	5.93 ± 0.24
VFA, Volatile Fatty Acid (g/L)	9.73 ± 0.65
(VS/TS) %	97.8 ± 0.39
Total Nitrogen (mg/L)	1026 ± 6.5
Moisture Content (%)	72.3 ± 1.4
Total Carbon (%TS)	53.35 ± 0.77
Alkalinity (g/L CaCO ₃)	1.231 ± 0.0164
C/N ratio	26.07 ± 0.66
Total COD (TCOD) (mg/L)	$180,\!362.5\pm962.5$
Soluble COD (SCOD) (mg/L)	$123,203 \pm 6253$

Inoculum

The inoculum utilized in this work was sourced from a benchtop anaerobic digester that processes waste from dairy products. After collection, the inoculum was primarily placed in an incubator to undergo additional acclimatization within an anaerobic digester until methane production ceased entirely. Subsequently, the inoculum was stored at a temperature of 4 $^{\circ}$ C prior to its utilization. Table 2 provides detailed information regarding the characteristics of this inoculum.

Table 2. Physicochemica	l attributes of thre	e different inoculur	n samples.
-------------------------	----------------------	----------------------	------------

Parameter	Value
Total Solid%	0.75 ± 0.25
Volatile Solid%	98 ± 1.1
рН	6.1 ± 0.2
VFA (Volatile Fatty Acids) (g/L)	0.46 ± 0.04
TN (Total Nitrogen) (mg/L)	340 ± 2.5
TCOD (Total COD) (mg/L)	5250 ± 250
Moisture Content (%)	99.25 ± 0.25
Alkalinity (g/L CaCO ₃)	0.861 ± 0.0164

2.2. Experimental Setup and Operation

The experiments were conducted using a batch operation mode. Pyrex solution containers from Fisher Scientific Ltd. located in Montreal, QC, Canada were utilized as batch anaerobic digesters. These bottles had a capacity of 500 mL each, and they were closed with butyl rubber septa and crimped with aluminum caps. The caps served the dual purpose of sealing the bottles securely to prevent any biogas produced from escaping and allowing for the collection of liquid and gas samples using syringes without the need to open the caps. pH adjustments in the batch reactors were made using 1 M NaOH (sodium hydroxide) and 1 M HCl (hydrochloric acid solution). The batch anaerobic reactors were maintained at ambient temperatures to create psychrophilic conditions within the range of 1–20 °C. Figure 2 illustrates the batch setup.



Figure 2. Batch experiment setup.

Anaerobic treatment of food waste was performed through multiple trials using six distinct total solid (TS) concentrations: 5%, 10%, 12%, 15%, 18%, and 20%, all conducted under psychrophilic temperature range ($1-20^{\circ}$ C). The anaerobic bioreactors were designed with a capacity of 500 mL, and the operational working volume of the reactors was considered to be 75% of this total capacity, equivalent to 375 mL. The biomass and tap water were added

according to the total solid percentages of each reactor up to the working volume of the reactor. At the top of each reactor, a pressure gauge was incorporated to monitor both pressure levels and accumulation of biogas. The pressure within the headspace fluctuated within the range of 3.5 to 20.7 kPa (0.5 to 3 psi). The initial batch experiments were carried out without any recirculation technique while in the later set of experiments, the psychrophilic anaerobic treatment was conducted with the reintroduction of the produced biogas into the anaerobic batch system. For the recirculation of biogas, the initial step involved collecting the accumulated biogas from the headspace using a 20 mL syringe. Subsequently, this collected gas was reintroduced into the feedstock through sparging, followed by anaerobic batch digesters that were manually mixed for a minimum of two to three minutes for further conversion. This recirculation process helped further reduce the CO₂ in the biogas through hydrogen or other electron donors and ensured more opportunity to covert CO_2 into methane. This manual mixing of biogas with feedstock stimulates methanogenesis but also enhances the interaction between gases and microorganisms, potentially leading to an augmented production of methane. Each group of batch reactors was operated under similar experimental conditions, featuring a food waste-to-inoculum ratio of 1.0.

Tap water was added to reach a final working volume of 500 mL in each anaerobic reactor. To establish anaerobic conditions, the digesters' headspaces were purged with ultra-high purity argon gas for a duration of 5 min. Throughout the treatment process, all digesters were manually stirred once for at least 2–3 min each day. Prior to every experiment, blank containers containing only inoculum and tap water were also set up to evaluate the biogas generation solely from the inoculum. Each batch experiment was carried out in duplicate, and the experiments were continued over a 30-day duration until the daily biogas generation fell below 1% of the total cumulative biogas production.

2.3. Biogas Withdrawal and Analysis

For the collection of gas and liquid samples, two different syringes were utilized: a 60 mL gas-tight syringe made of glass and a 10 mL plastic syringe, respectively. These syringes were procured from Fisher Scientific Ltd. in Montreal, QC, Canada. During the batch experiment, a liquid sample was extracted on a daily basis. Simultaneously, biogas that had accumulated in the headspace of the anaerobic reactors was sampled at five-day intervals throughout the entire treatment process (Figure 3). This sampling regimen allowed monitoring of the liquid samples and the composition of generated gas. The concentrations of CH₄ and CO₂ in the produced biogas were measured by a gas chromatograph (GC, Agilent 7890B with a thermal conductivity detector), Agilent Technologies (Santa Clara, CA, USA) utilizing a CARBOXEN 1010 PLOT (30 m × 0.32 mm) capillary column from SUPELCO (Merck KGaA, Darmstadt, Germany) with helium as the carrier gas at a 250 °C inlet temperature. A thermal conductivity detector (TCD) was utilized with a column oven temperature of 250 °C which was incrementally increased at a rate of 5 °C per minute. The injection flow rate was maintained at 6.5 mL/min, and the entire experimental duration spanned 5 min. In the experimental setup, a Tedlar bag was attached to a needle to extract the gas generated within the anaerobic digesters. Subsequently, the water displacement method was utilized to measure the volume of biogas produced in each bag. Several crucial process parameters were monitored throughout the course of the experiment. These included the measurement of Chemical Oxygen Demand (COD), tracking the production of biogas, and assessing the methane content in generated biogas. Continually observing these factors provided a better understanding of the dynamics of the treatment process and its outcomes.

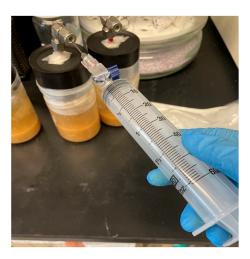


Figure 3. Biogas withdrawal from anaerobic digester.

2.4. Analytical Parameters

Chemical Oxygen Demand (COD) Measurement

The USEPA reactor digestion method was employed to assess the chemical oxygen demand (COD) within a range of 20 to 1500 mg/L, as outlined in Standard Method 5220 D [22]. This analysis involved the utilization of COD test ampoules provided by Hach Inc., Loveland, CO, USA alongside a spectrophotometer, specifically the Cole Parmer model DR 2800, and a DRB200 Digital Reactor Block. This comprehensive setup ensured precise measurements of COD within the specified range. The sample was diluted with distilled water when the sample's COD concentration exceeded the measurement capabilities.

Alkalinity Analysis

Alkalinity serves as a critical parameter that represents a solution's buffering capacity and its capacity to neutralize acidic influences. The titration method outlined in NO. 2320B was utilized for the measurement of alkalinity of the food waste samples and inocula [23]. The materials used for this analysis included distilled water, Bromocresol green, and a 0.1 N solution of sulfuric acid.

Alkalinity
$$(mg \frac{CaCO_3}{l}) = \frac{A \times N \times 50,000}{mL \text{ sample}}$$
 (1)

A = volume (mL) of acid used for titration; N = normality of standard acid.

Total Nitrogen (TN) Measurement

Total nitrogen encompasses every type of nitrogen present within a sample. We quantified Total Nitrogen (TN) and evaluated it using the persulfate digestion method, which involved the use of Total TNT plus Reagent purchased from Hach Inc. and a spectrophotometer, specifically the Cole Parmer model DR 2800, in conjunction with a DRB200 Digital Reactor Block.

Volatile Fatty Acids (VFAs) Measurement

Volatile Fatty Acid concentration was assessed by using the esterification method and employing the Volatile Acids TNT plus Reagent, which was purchased from Hach Inc. (Ames, LA, USA). This analytical process was carried out using the Cole Parmer model DR 2800 spectrophotometer and a DRB200 Digital Reactor Block [14].

Microbial Parameters (TS, VS) Analysis

Analyzing the solids content plays a pivotal role in managing both biological and physical treatment procedures, as well as ensuring adherence to regulatory effluent standards analysis. This analytical assessment plays a crucial role in evaluating whether effluents

are in compliance with the stipulated regulatory effluent standards. These experiments were conducted to determine the biomass concentration in the anaerobic digester and to quantify the reduction in volatile solids content in the effluent when compared to the influent, thereby assessing the system's overall efficiency. These analyses were conducted as per the Standard Methods following the Standard Methods for measuring total Solids (TS) and volatile Solids (VS) [24,25]. It is important to note that each test underwent a minimum of three repetitions to ensure the accuracy and reliability of the results.

$$TS = \frac{Wd - Wc}{Ws - Wc} \times 100 \tag{2}$$

$$VS = \frac{Wd - Wash}{Wd - Wc} \times 100 \tag{3}$$

TS: total solid concentration; VS: volatile solid concentration

Wd: dried sample weight, g Ws: fresh sample weight, g Wc: blank crucible weight, g

Wash: ash weight, g.

3. Results and Discussion

Each biological reaction in anaerobic treatment is inherently sensitive to temperature fluctuations. A reduction in treatment temperature not only impacts bacterial metabolism but also alters bacterial reaction kinetics. The mesophilic temperature range (30–35°C) is generally considered optimal for bacterial growth in anaerobic treatment. However, in regions like Canada and other countries where winter temperatures drop to sub-zero temperature levels, maintaining mesophilic conditions demands a substantial energy input. Given these challenging climatic conditions, psychrophilic anaerobic treatment emerges as a viable alternative with the potential to maximize net energy production. Nevertheless, operating at psychrophilic temperatures can adversely affect the hydrolysis rate of lipids and proteins, potentially leading to the accumulation of volatile fatty acids (VFAs) and ultimately resulting in process failure. To address these challenges and improve system efficiency while operating at lower temperatures, this research investigates the influence of biogas recirculation on the performance of anaerobic food waste treatment, aiming to boost methane concentration in the biogas.

3.1. Biogas Composition during Psychrophilic Anaerobic Treatment with or without Recirculation

To investigate the potential of psychrophilic treatment for food waste and explore the influence of the recirculation of biogas on biomethane generation, a series of experiments were conducted utilizing two sets of six batch digesters. These digesters were subjected to varying total solid percentages, specifically 5%, 10%, 12%, 15%, 18%, and 20%, over 30-day periods. The primary objective was to evaluate the feasibility of psychrophilic treatment while examining the impact of the recirculation of biogas.

The initial trial of psychrophilic batch anaerobic reactors was performed without any biogas recirculation, while in the subsequent trial of experiments, biogas recirculation was introduced into the system. The experimental findings demonstrated a remarkable enhancement in methane production and methane concentration within the generated biogas due to the implementation of biogas recirculation under psychrophilic conditions without biogas recirculation. The methane concentration exhibited fluctuations between 10% and 15% within the first seven days of the treatment period. Conversely, with biogas recirculation, the purity of the generated methane experienced a substantial increase during the initial phase of treatment, reaching approximately $50 \pm 10\%$. Without biogas recirculation, methane production became noticeable after 25 days of the treatment duration. However, with biogas recirculation, the detection of methane occurred much earlier, after only 15 days of operation. Subsequently, as volatile fatty acids (VFAs) were gradually

consumed over time, the high alkalinity in the medium counteracted acidification, resulting in an elevation of pH levels. This, in turn, led to the degradation of VFAs. The data derived from both sets of experiments clearly demonstrated that the recirculation of biogas within the anaerobic digester had a profound impact, increasing the methane content in the biogas by more than 50%.

In the initial series of experiments, the biogas composition displayed a high concentration of CO₂, reaching up to 90% in the early stages of the treatment process. This contrasted with the typical range of carbon dioxide content in biogas, which usually falls between 30% and 40% [26]. As the anaerobic treatment progressed, the CO₂ percentage gradually decreased over time, reaching its lowest point after 25 days of operation. Specifically, the lowest CO₂ content levels for various total solid concentrations of 5%, 10%, 12%, 15%, 18%, and 20% were recorded at 61.1%, 58.7%, 54.1%, 61.7%, 74.1%, and 74.2%, respectively, when no biogas recirculation was applied. In the absence of biogas recirculation, findings from the psychrophilic anaerobic treatment demonstrated that CO2 was the predominant constituent within the generated biogas. Nonetheless, the introduction of biogas recirculation had a significant impact on reducing CO₂ levels in the generated biogas. Specifically, the lowest CO₂ content values were measured at 18%, 6.5%, 19.3%, 32.9%, 26.9%, and 30% for TS concentrations of 5%, 10%, 12%, 15%, 18%, and 20%, respectively, when biogas recirculation was employed. Consequently, the experimental outcomes clearly demonstrated that biogas recirculation not only increased methane production but also reduced the carbon dioxide concentration in the generated biogas. As a result, this led to a system that was more dependable and exhibited higher energy efficiency.

3.2. Methane Concentration and Methane Yield at Different Total Solid Concentrations

With the aim of comprehensively assessing the influence of total solid concentration (TS%) and the introduction of the recirculation of biogas on the methane purity within the biogas, maximum methane concentrations of the produced biogas under psychrophilic temperatures with or without biogas recirculation are presented in Figure 4a. The error bars depicted in Figure 4a represent the standard deviation of the measured methane concentration taken for every individual sample. The findings of the study reveal that the highest methane content, reaching 93.6%, was observed at a total solid (TS) level of 10% when biogas recirculation was employed. Conversely, without biogas recirculation, the highest methane concentration attained was 45.9%, occurring at a TS% of 12%. Previous research by Costa et al. suggested that production of methane tends to increase with rising temperatures [27]. However, the experimental results demonstrated that substantial methane production can occur at psychrophilic conditions when recirculation of biogas is employed. Furthermore, the outcomes suggest that methane concentrations within the biogas were generally higher in both wet and semi-dry digestion processes, whereas the impact of total solid percentages on methane concentration became negligible when considering dry anaerobic treatment conditions.

The methane production findings from the psychrophilic anaerobic treatment without biogas recirculation exhibited a range of 0.07 to 0.12 L CH $_4$ per gram of COD removed, as shown in Figure 4b. Conversely, when biogas recirculation was employed, methane production increased, ranging from 0.18 to 0.24 L CH $_4$ per gram of COD removed. It is important to note that the theoretical methane yield, at 0 $^{\circ}$ C and 1 atm pressure, is 0.35 L per gram of COD removed [28]. However, since the study was conducted under psychrophilic temperature conditions, an adjustment is required to account for this operational temperature. The highest theoretical methane yield achievable at psychrophilic temperatures was calculated to be 0.378 L per gram of COD. It is frequently observed that the practical methane yield is below theoretical expectations. This discrepancy may be attributed to the inherent limitations of a bench-top reactor setup, which might not represent the efficiency potential achievable in more advanced reactor designs, such as commercial anaerobic digesters or expanded bed reactors. Additionally, the lower methane production yield observed in the batch anaerobic digester can be attributed to the quality and state of the

biomass, which had been stored for one month before the treatment process. In contrast, treatment plants typically use biomass that has been adapted to the waste material and is employed directly without the need for prior storage.

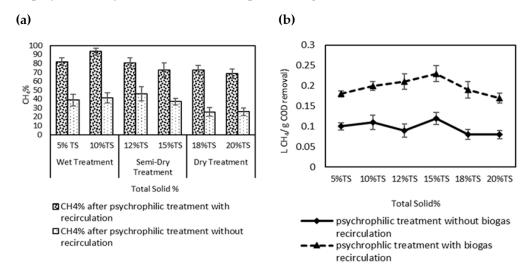


Figure 4. (a) Impact of total solid percentages and recirculation of biogas on methane concentrations in the biogas (b) Variation of methane yield with different total solid percentages.

Methane production yields displayed consistent patterns across all treatment conditions, indicating uniform trends and behaviors (Figure 4b). A research study of the anaerobic treatment of food waste reported that the highest methane yield was obtained under semi-dry conditions [18]. Likewise, in this research, the highest methane yields were recorded under semi-dry treatment conditions, whereas the lowest methane yields were observed in the case of dry treatment conditions. The error bars depicted in Figure 4b serve as indicators of the standard deviation, representing the degree of variability in measured methane yield across different samples.

3.3. Removal Efficiency of VS and COD at Different Total Solid Contents after Anaerobic Treatment of Food Waste

Figure 5 summarizes the impact of total solid percentages and recirculation of biogas on both COD removal efficiency and VS reduction. The error bars in Figure 5 represent the standard deviation of the COD removal and VS reduction for each sample. The outcomes reveal that, regardless of the recirculation of biogas, the highest COD removal occurred at a 10% total solid percentage during psychrophilic anaerobic treatment. Comparatively, COD removal efficiencies were lower in semi-dry conditions than in wet digestion, reaching their lowest point during dry treatment. Previously, a study reported a cumulative total COD removal efficiency of approximately 75% in a hybrid anaerobic digester operating at psychrophilic temperatures [29]. However, in this study, the highest COD removal efficiency was found to be 82% at a 10% total solid percentage in psychrophilic conditions with biogas recirculation. This outcome closely corresponds to the chemical oxygen demand (COD) reduction of 73.7% that was achieved during mesophilic anaerobic treatment of food waste, conducted within a prototype anaerobic digester, with a hydraulic retention time (HRT) of 27 days [13]. Anaerobic digestion relies on microorganisms breaking down organic compounds in the absence of oxygen. Higher COD removal suggests that the microbial community is effectively breaking down complex organic substances into simpler compounds, such as methane and carbon dioxide. Therefore, a higher COD removal indicates that more organic matter has been converted into biogas, leading to increased methane production. This is beneficial for harnessing renewable energy from the treated food waste. Conversely, the lowest COD removal efficiency was observed at 34% for 18% total solid percentage under psychrophilic conditions without biogas recirculation. Similarly, with respect to VS reduction, after 30 days of operation, the addition of biogas recirculation

during psychrophilic anaerobic treatment resulted in a VS reduction of 74.9% at 5% total solid percentage, while volatile solid reduction (VS) reduction was around 53% without recirculation. At 10% total solid percentage, these values changed to approximately 72% with recirculation and 68% without. At 12% total solid percentage, VS reduction percentages were 92.4% with recirculation and 61.6% without. Similarly, for 15%, 18%, and 20% total solid percentages, VS reductions reached almost 88%, 77%, and 78%, respectively, with recirculation of biogas. On the other hand, without any recirculation, volatile solid reductions for these percentages decreased to 74.2%, 73.8%, and 60.7%, respectively. Notably, under psychrophilic conditions with recirculation, VS reductions fell within the range of 74% to 94%, which is similar to the outcomes typical for mesophilic anaerobic treatment of food waste [30]. Higher volatile solids (VS) reduction during the anaerobic treatment of food waste means that a larger proportion of the volatile solids present in the food waste has been successfully degraded or converted into biogas during the anaerobic digestion process. VS reduction is a key parameter used to evaluate the effectiveness of anaerobic digestion, particularly in the context of organic waste treatment.

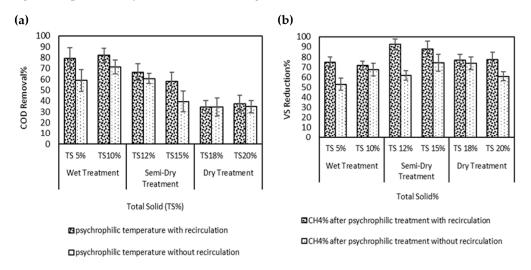


Figure 5. (a) COD removal efficiency and **(b)** VS reduction by anaerobic treatment with or without recirculation of biogas.

The substantial increase in volatile solids (VS) removal efficiency serves as a strong indicator of the rapid consumption of the organic portion within the total solids by methanogenic microorganisms [31]. Notably, the highest volatile solid reduction, reaching 92.4%, was observed at a 12% total solid percentage during psychrophilic anaerobic treatment with the application of biogas recirculation. A prior experiment into the psychrophilic anaerobic treatment of food waste had produced a substantial 87.0% reduction in volatile solids (VS) when employing semi-dry treatment. These results align closely with the outcomes of the current study, providing additional support and consistency regarding the effectiveness of semi-dry conditions in the anaerobic treatment of food waste [32]. These consistent outcomes underscore the notable benefits of introducing biogas recirculation into the anaerobic reactor, enhancing both COD reduction efficiency and VS reduction, without the need for any additional heating.

3.4. Temperature Profile during Anaerobic Treatment of Food Waste

Batch experiments with six total solid percentages were conducted under mesophilic and psychrophilic temperatures. Batch digesters were kept at atmospheric temperature for psychrophilic conditions (1–20 $^{\circ}$ C). Each day, a temperature reading was taken for 30 days to create the temperature profile. The variation in daily temperature during the treatment process is presented in Figure 6. The lowest temperature reading of 18 $^{\circ}$ C was obtained while the highest temperature was recorded at 22 $^{\circ}$ C during psychrophilic treatment. The average temperature of 19.8 $^{\circ}$ C was recorded at the end of the 30-day digestion.

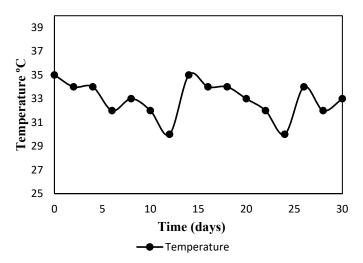


Figure 6. Temperature profile during anaerobic treatment of food waste.

3.5. pH Profile during Psychrophilic Treatment with or without Recirculation Technique

The pH level exerts a significant influence on the hydrolysis stage, which constitutes a rate-limiting stage in anaerobic digestion. Figures 7a and 7b portray the reactor pH throughout the psychrophilic data collection period with and without biogas recirculation, respectively.

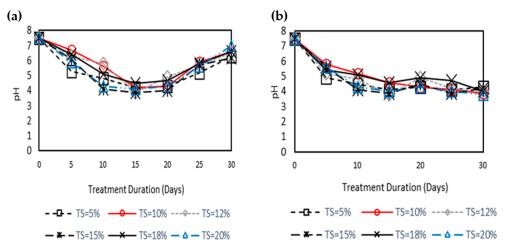


Figure 7. pH profile during anaerobic treatment. (a) with recirculation of biogas, (b) without recirculation of biogas.

In anaerobic treatment, the optimal pH range for methanogenic microorganisms typically falls between 6.5 and 7.5 [33]. To assess the variations in pH levels throughout the anaerobic treatment procedure, the initial pH values were kept the same for all batch experiments. Observing the pH trend in Figure 7a, it becomes evident that psychrophilic anaerobic treatment with recirculation of biogas fosters system stability as well as system efficiency. However, in the subsequent set of experiments depicted in Figure 7b, the final pH levels for varying total solid percentages (TS 5%, 10%, 12%, 15%, 18%, and 20%) were observed at 4.3, 3.9, 4.0, 4.2, 4.0, and 3.8, respectively. This decline in pH can be due to the accumulation of volatile fatty acids (VFAs) within the anaerobic system [34]. Such pH decrease could potentially inhibit methanogenic microorganisms partially or entirely, introducing stress to the system, particularly in terms of methane production [35].

Conversely, when biogas recirculation was applied, the final pH values for the same range of total solid percentages (5%, 10%, 12%, 15%, 18%, and 20% total solid) were 6.3, 6.6, 6.5, 6.2, 6.6, and 7, respectively. These pH levels serve as indicators of efficient reactor

performance [36,37]. Several studies have similarly reported that effluent pH tends to decrease gradually after the first week of anaerobic treatment, signing the formation of volatile fatty acids (VFAs) [38,39]. However, pH levels typically rebound to their standard operating values after volatile fatty acid (VFA) metabolism. The aforementioned pH profiles suggest that reactors undergoing psychrophilic treatment with biogas recirculation appear to operate in a healthy state, as the pH values fall within the stable neutral spectrum of 6.0 to 7.5, which is conducive for methanogens [40].

4. Conclusions

The application of anaerobic treatment technology to mixed food waste remains relatively limited, despite this waste being one of the most energy-rich materials available. This research endeavours to showcase anaerobic treatment technology as a viable approach of collecting methane from food waste and utilizing it as a sustainable source of renewable energy. Furthermore, apart from the conventional anaerobic system, this work introduces an innovative approach to food waste treatment for improved methane production, employing biogas recirculation at lower temperatures. As a result, contrary to the prevailing belief that maximal methane generation in anaerobic treatment occurs within the mesophilic conditions, this research provides experimental evidence demonstrating that significant methane production can also be achieved in the psychrophilic temperature range. The psychrophilic anaerobic treatment of food waste demonstrates a significantly enhanced energy efficiency system compared to the mesophilic process, specifically in frigid climates. Nonetheless, further research is required to achieve a thorough understanding of this proposed method of food waste treatment, encompassing various types of anaerobic digesters and variations in composition of food waste. Additional initiatives should be directed towards clarifying the limitations of bacteria-mediated procedures under psychotropic environment, and the synergistic interactions between different archaeal and bacterial communities should be investigated to recognize the function of the individual community within the diverse consortium.

Author Contributions: Conceptualization, C.N.M. and T.T.; methodology, C.N.M. and T.T.; software, T.T.; validation, C.N.M., formal analysis, T.T.; investigation, T.T.; resources, T.T.; data curation, T.T., writing—original draft preparation, T.T.; writing—review and editing C.N.M.; visualization, C.N.M.; supervision, C.N.M.; project administration, C.N.M.; funding acquisition, C.N.M. All authors have read and agreed to the published version of the manuscript.

Funding: Funding for this research was provided by NSERC PURE and Concordia University.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The original contributions presented in the study are included in the article, further inquiries can be directed to the corresponding author.

Acknowledgments: The authors sincerely acknowledge the support of the staff of the Environmental Lab of the Department of Building, Civil and Environmental Engineering at Concordia University for their assistance in carrying out the experiments.

Conflicts of Interest: The authors declare no conflict of interest.

References

- 1. Ma, Y.; Liu, Y. Turning food waste to energy and resources towards a great environmental and economic sustainability: An innovative integrated biological approach. *Biotechnol. Adv. Manag.* **2019**, *34*, 2627–2633. [CrossRef]
- 2. Slorach, P.C.; Jeswani, H.K.; Cuéllar-Franca, R.; Azapagic, A. Environmental sustainability of anaerobic digestion of household food waste. *J. Environ. Manag.* **2019**, 236, 798814. [CrossRef]
- 3. Lin, C.S.K.; Pfaltzgraff, L.A.; Herrero-Davila, L.; Mubofu, E.B.; Abderrahim, S.; Clark, J.H.; Luque, R. Food waste as a valuable resource for the production of chemicals, materials, and fuels. Current situation and global perspective. *Energy Environ. Sci.* **2013**, *6*, 426–464. [CrossRef]

4. Zamanzadeh, M.; Hagen, L.H.; Svensson, K.; Linjordet, R.; Horn, S.J. Biogas production from food waste via co-digestion and digestion-effects on performance and microbial ecology. *Sci. Rep.* **2017**, *7*, 17664. [CrossRef]

- 5. U.S. EPA. Municipal Solid Waste Generation, Recycling, and Disposal in the United States: 2014. Available online: https://www.epa.gov/sites/default/files/2015-09/documents/2012_msw_fs.pdf (accessed on 22 January 2024).
- 6. Clercq, D.D.; Wen, Z.; Gottfried, O.; Schmidt, F.; Fei, F. A review of global strategies promoting the conversion of food waste to bioenergy via anaerobic digestion. *Renew. Sustain. Energy Rev.* **2017**, *79*, 204–221. [CrossRef]
- 7. Papargyropoulou, E.; Lozano, R.; Steinberger, K.; Wright, J.; Ujang, N.; Bin, Z. The food waste hierarchy as a framework for the management of food surplus and food waste. *J. Clean. Prod.* **2014**, *76*, 106–115. [CrossRef]
- 8. Gokarn, S.; Kuthambalayan, T.S. Analysis of challenges inhibiting the reduction of waste in food supply chain. *J. Clean. Prod.* **2017**, *168*, 595–604. [CrossRef]
- 9. Ariunbaatar, J.; Panico, A.; Esposito, G.; Pirozzi, F.; Lens, P.N.L. Pre-treatment methods to enhance anaerobic digestion of organic solid waste. *Appl. Energy* **2014**, *123*, 143–156. [CrossRef]
- 10. Morales-polo, C.; Cledera, M.; Moratilla Soria, B.Y. Reviewing the Anaerobic Digestion of Food Waste: From Waste Generation and Anaerobic Process to Its Perspectives. *Appl. Sci.* **2018**, *8*, 1804. [CrossRef]
- 11. Posmanik, R.; Labatut, R.A.; Kim, A.H.; Usack, J.G.; Tester, J.W.; Angenent, L.T. Coupling hydrothermal liquefaction and anaerobic digestion for energy valorization from model biomass feedstocks. *Bioresour. Technol.* **2017**, 233, 134143. [CrossRef]
- 12. Bachmaier, J.; Effenberger, M.; Gronauer, A. Greenhouse gas balance and resource demand of biogas plants in agriculture. *Eng. Life Sci.* **2010**, *10*, 560–569. [CrossRef]
- 13. Ratanatamskul, C.; Onnum, G.; Yamamoto, K. A prototype single-stage anaerobic digester for co-digestion of food waste and sewage sludge from high rise building for on-site biogas production. *Int. Biodeterior. Biodegrad.* **2014**, *95*, 176–180. [CrossRef]
- 14. Cirne, D.G.; Paloumet, X.; Björnsson, L.; Alves, M.M.; Mattiasson, B. Anaerobic digestion of lipid-rich waste—Effects of lipid concentration. *Renew. Energy* **2007**, *32*, 965–975. [CrossRef]
- 15. Moriarty, K. Feasibility Study of Anaerobic Digestion of Food Waste in St. Bernard, Louisiana: A study prepared in partnership with the Environmental Protection Agency for the RE-Powering America's Land Initiative: Siting Renewable Energy on potentially Contaminated land and Mine Sites. 2013. Available online: https://www.nrel.gov/docs/fy13osti/57082.pdf (accessed on 22 January 2024).
- 16. Mulligan, C.N. *Environmental Biotreatment Technologies for Air, Water, Soil, and Wastes*; ABS Consulting, Government Institutes: Rockville, MD, USA, 2002.
- 17. Rivas-García, P.; Botello-Álvarez, J.; Miramontes-Martínez, L.; Cano-Gómez, J.; Rico Martínez, R. New model of hydro lysis in the anaerobic co-digestion of bovine manure with vegetable waste: Modification of anaerobic digestion model No. 1. *Rev. Mex. Ing. Química* 2020, 19, 109–122. [CrossRef]
- 18. Zhang, C.; Su, H.; Baeyens, J.; Tan, T. Reviewing the anaerobic digestion of food waste for biogas production. *Renew. Sustain. Energy Rev.* **2014**, *38*, 383–392. [CrossRef]
- 19. Dhaked, R.K.; Singh, P.; Singh, L. Bio methanation under psychrophilic conditions. Waste Manag. 2010, 30, 2490–2496. [CrossRef]
- 20. Lettinga, G.; Rebac, S.; Zeeman, G. Challenge of psychrophilic anaerobic wastewater treatment. *Trends Biotechnol.* **2001**, *19*, 363–370. [CrossRef]
- 21. Rusín, J.; Chamrádová, K.; Basinas, P. Two-stage psychrophilic anaerobic digestion of food waste: Comparison to conventional single-stage mesophilic process. *Waste Manag. J.* **2021**, *119*, 172–182. [CrossRef]
- 22. APHA. Standard Methods for the Examination of Water and Wastewater, 21st ed.; American Public Health Association, American Water Works Association, Water Environment Federation: Washington, DC, USA, 2005.
- 23. Hach Company. Hach Procedure Manual; Hach Company: Loveland, CO, USA, 2009.
- 24. Clesceri, L.S.; Greenberg, A.E.; Eaton, A.D. (Eds.) *Standard Methods for the Examination of Water and Wastewater*; United Book Press: Baltimore, MD, USA, 1998.
- 25. U.S. EPA. Wastewater Technology Fact Sheet. Chemical Precipitation; Office of Water: Washington, DC, USA, 2000.
- 26. Environmental and Energy Study Institute (EESI). Biogas: Converting Waste to Energy. 2017. Available online: http://www.eesi.org/papers (accessed on 1 October 2017).
- 27. Costa, C.; Rodríguez, J.; Márquez, M. A simplified dynamic model for the activated sludge process with high strength wastewaters. *Environ. Model Assess* **2009**, *14*, 739–747. [CrossRef]
- 28. Michaud, S.; Bernet, N.; Buere, P.; Roustan, M.; Moletta, R. Methane yield as a monitoring parameter for the start-up of anaerobic fixed film reactors. *Water Res.* **2002**, *36*, 1385–1391. [CrossRef]
- 29. Tiwari, B.R.; Rouissi, T.; Brar, S.K.; Surampalli, R.Y. Critical insights into psychrophilic anaerobic digestion: Novel strategies for improving biogas production. *Waste Manag.* **2021**, *131*, 513–526. [CrossRef]
- 30. Kayhanian, M. Biodegradability of the organic fraction of municipal solid waste in a high-solids anaerobic digester. *Waste Manag. Res.* **1995**, *13*, 123–136. [CrossRef]
- 31. Minale, M.; Worku, T. Anaerobic co-digestion of sanitary wastewater and kitchen solid waste for biogas and fertilizer production under ambient temperature: Waste generated from condominium house. *Int. J. Environ. Sci. Technol.* **2014**, *11*, 509–516. [CrossRef]
- 32. Rajagopal, R.; Bellavance, D.; Rahman, S. Psychrophilic anaerobic digestion of semi-dry mixed municipal food waste: ForNorth American context. *Process Saf. Environ. Prot.* **2017**, *105*, 101–108. [CrossRef]

33. Zupančič, G.D.; Grilc, V. Anaerobic Treatment and Biogas Production from Organic Waste, Management of Organic Waste; Kumar, S., Ed.; InTech: Berlin, Germany, 2012; ISBN 978-953-307-925-7.

- 34. Maurus, K.; Kremmeter, N.; Ahmed, S.; Kazda, M. Biomass Conversion and Biorefinery High-resolution monitoring of VFA dynamics reveals process failure and exponential decrease of biogas production. *Biomass Convers. Biorefinery* **2023**, *13*, 10653–10663. [CrossRef]
- 35. Amani, T.; Nosrati, M.; Sreekrishnan, T.R. Anaerobic digestion from the viewpoint of microbiological, chemical, and operational aspects—A review. *Environ. Rev.* **2010**, *18*, 225–278. [CrossRef]
- 36. Wheatley, A. *Anaerobic Digestion: A Waste Treatment Technology for Developing Countries*, 2nd ed.; Elsevier Science Publisher Ltd.: London, UK, 1991.
- 37. Kondusamy, D.; Kalamdhad, A.S. Pre-treatment and anaerobic digestion of food waste for high-rate methane production—A review. *J. Environ. Chem. Eng.* **2012**, *2*, 1821–1830. [CrossRef]
- 38. Anderson, G.K.; Yang, G. pH control in anaerobic treatment of industrial wastewater. *J. Environ. Eng.* **1992**, *118*, 551–567. [CrossRef]
- 39. Rao, M.S.; Singh, S.P.; Singh, A.K.; Sodha, M.S. Bioenergy conversion studies of the organic fraction of MSW: Assessment of ultimate bioenergy production potential of municipal garbage. *Appl. Energy* **2000**, *66*, 75–78. [CrossRef]
- 40. Gunaseelan, V.N. Biochemical methane potential of fruit and vegetable solid waste feedstocks. *Biomass Bioenergy* **2004**, *26*, 389–399. [CrossRef]

Disclaimer/Publisher's Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.