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Analysis of Biogas Recovery from Liquid Dairy Manure Waste by Anaerobic Digestion

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ABSTRACT

In this paper, an industrial dairy farm unit was taken as a case study to carry out the applicable technical assessment for the construction of a biogas plant using a combined heat and power (CHP) unit. A comprehensive sensitivity analysis was applied to examine the effectiveness of the operational parameters and feed composition in the purity and production rate of biogas. Aspen Plus was used to implement the anaerobic digestion process. The results showed that any increase in the digester's operational performance and mass rate of feedstock water led to the modification of biomethane content, but dropped in biogas mass flow rate. Moreover, an increase in the mass rate of carbohydrates, protein, and organic load rate (OLR) of feedstock reduces methane composition. Besides, increasing the rate of lipids has raised the rate of methane production and its composition.

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1. INTRODUCTION

Nowadays, dairy products include more than 30 % of the gross volume of agricultural wastes in developing countries [1]. This value has an ever-increasing growth due to income and population growth and changes in lifestyle and diet. Dairy products create around 60 million tons of waste annually, about 20 % of the total globally produced wastes [2]. Such an enormous quantity of wastes accumulated without suitable management is quite hazardous to the environment. They would cause a series of irreversible damages to water and soil, environmental contaminations like pollution of aquifers, groundwater eutrophication, accumulation of nutrients in soil, dispersion pathogens, accumulation of toxic ingredients, and greenhouse gas emissions (methane, oxide nitrogen, and ammonia) [3, 4]. Farm-based management approaches could reduce these issues in dairies. Anaerobic digestion is one of the most attractive and economical ways based on environmental regulations [5, 6]. By producing heat and power and remaining digestate as organic compost, this method can decrease the organic waste volume and increase the total efficiency of resources [7]. Anaerobic digestion has four stages where in each stage, a group of microorganisms degrades primary substrates and leaves them to the next stage [8]:

- Hydrolysis: In this stage, complex organic molecules are converted into monomeric compounds by hydrolyzing bacteria, and all non-dissolved particles change into aqueous solutions that would become usable for microorganisms [9]. During this stage, carbohydrate, lipid, protein, lignin, and nonorganic materials break down into simple molecules such as sugar, alcohol, short-chain fatty acids, amino acid, aromatic compounds, non-degradable organic materials, and nonorganic materials. It should be noted that lignin degradation is complicated due to its weak solvability, molecular largeness, and complicated structure.
- Acidogenesis (fermentation): After the fermentation of primary molecules and their transformation into monomeric compounds, all monomers are converted into short-chain organic acids, acetic acids, propionic acids, butyric acids, pentanoic acids, hydrogen, CO_2, and other compounds by acidogenic bacteria. Also, alcohols like ethanol and methanol can be extracted from the remaining substrates. Also, aldehydes and oxygen are produced in this stage. Finally, all produced products are left to the methanogenic bacteria [10-13].
- Acetogenic phase: In the third stage, the decomposed substrates are converted into CO_2, H_2, and acetate by acetogenic bacteria. The hydrogen released during this stage has toxic effects on the process [13].
- Methanogenic phase: The last step of the anaerobic digestion process is the methanogenic phase. In this stage, methanogens produce methane by utilizing the previously produced products (CO_2, hydrogen, and acetate). This phase is carried out by two groups of microorganisms. The first one converts acetate into methane and CO_2 and the second group uses hydrogen as a donor and CO_2 as an

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acceptor to produce methane. The methane production process takes a long time via methanogenic bacteria in the range of 3-50 days [14, 15].

Any degradable and organic material can be used as the feedstock of anaerobic digestion that can be transformed into methane (CH_4), carbon dioxide (CO_2), and other gases through biological processes [16-18]. CH_4 usually comprises about 45-75 % of the total biogas volume. Based on the CH_4 volume percentage, the lower heating value (LHV) of biogas varies between 16-28 MJ/m^3 [7]. The composition and ingredients of feedstocks significantly affect the efficiency of producing biogas [7, 19]. Due to the physical properties, organic content, and abundancy, dairy manures are known as the best biomass and feed for biogas units, especially in developing countries [20]. Carbohydrates, proteins, and lipids are the main compositions of dairy manures that would be very attractive due to their anaerobic bacteria, high water contents (acting as a solvent), and low price [6, 7]. Dairy manure is one of the richest sources of carbohydrates (cellulose, hemicellulose, lignin, sugar, and starch), accounting for over 80 % of organic materials. According to a report published by International Energy Agency (IEA), the agriculture sector will be the biggest biogas producer in the world up to 2040, and animal and agricultural wastes will account for 40 % and 35 % of the total globally produced biogas, respectively [7]. Also, this agency predicts a 2.5 % annual growth in animal waste. The European Union and China are the biggest producers of biogas in this sector [7].

Operational condition including temperature, pressure, pH, retention time, carbon-to-nitrogen ratio, ammonia concentration, and long-chain fatty acid concentration affect the digestion process in terms of its performance and stability. Hence, optimal conditions can bring about the health and growth of microorganisms which would be necessary for continuous digestion process [17, 21, 22]. Accordingly, implementation and simulation of anaerobic digestion is a scientific and complicated process. Many studies have been carried out on modeling. Among them, Anaerobic Digestion Model NO. 1 (ADM1), a mathematical model developed by the International Water Association (IWA) task group [23], is a precise and complete one over its information, reactions, and kinetic calculations. Angelidaki et al. introduced a model covering most of inhibiting factors like free ammonia, volatile fatty acids (VFAs), and long-chain fatty acids [24]. However, this model is characterized by some defects including not considering hydrogen and temperature parameters. Al-Rabiei et al. [25] developed a process simulation model (PSM) and implemented sensitivity analyses considering the effect of hydrogen injection, substrate concentration, and operational parameters on raising the production efficiency of biogas, quantitatively and qualitatively. However, only limited attention has been dedicated to the simulation of anaerobic digestion via physical-chemical processes and parameters affecting biogas production efficiency [13, 25-27]. Hence, this paper aims to conduct a process study of dairy waste produced in livestock farms and production of value-added streams, including biogas and digestate. Moreover, the effect of operational parameters and some inhibitors on CH_4 production was studied.

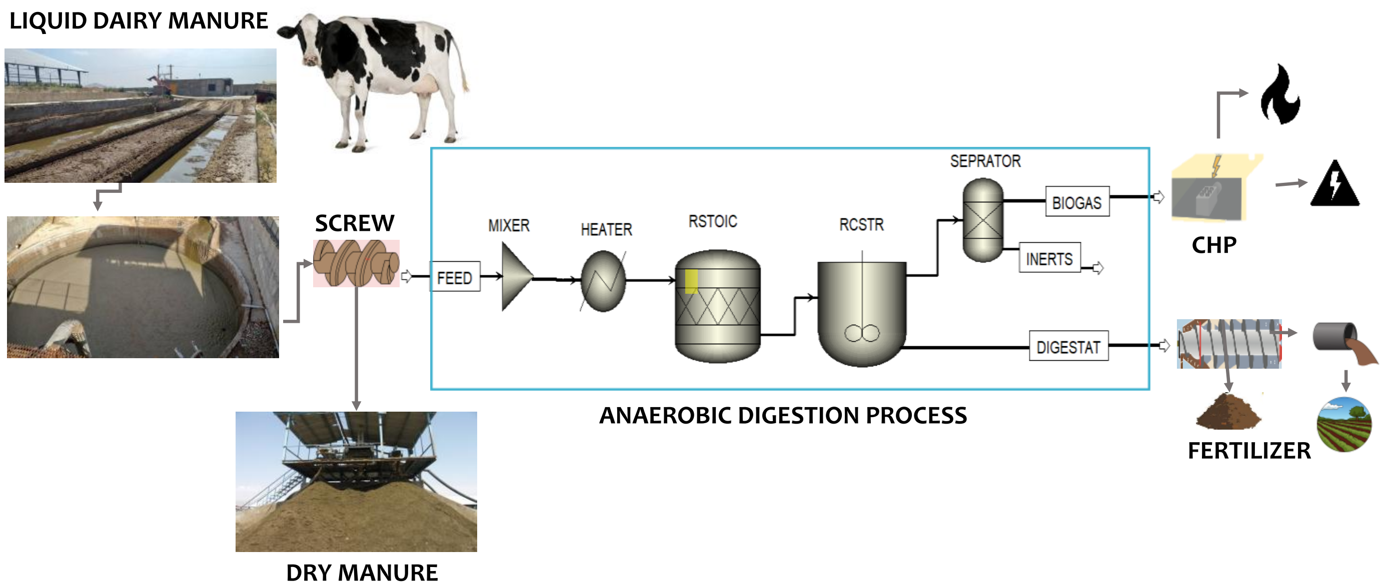

Figure 1. Schematic of anaerobic digestion process from row material to end products

2. EXPERIMENTAL

2.1. Case study description

An industrial dairy farm is a case study for the integrated management of livestock-made dairy manures. This unit is located in the southeastern province of Khorasan Razavi and has 1620 head of livestock, including cattle and dairy cows. The average weight of cows is around 670 kg, and each one produces approximately 0.048 m^3 manure daily. The keeping system is based on freestalls, and the flushed-water system is used for waste collection, washing, and sand scratching. It should be noted that before storing the wastes, solid-liquid separation is needed because the solid particles derived from livestock beds can cause many problems during the pumping of waste and dislocation, and their concentration over time decreases the capacity of the waste storage system (see Fig. 1). Moreover, mechanical separators can facilitate recycling wastes and land applications. More information about this case is shown in Table 1.

Table 1. Animal husbandry, liquid dairy manure, and biogas components information [28]

Animal husbandry	
Total number of mature animals (head)	1620
Type of keeping and washing system	Free stall-flushed water system
Bed type	Sand
The volume of manure produced per animal (m^3/day)	0.048
The volume of consumed water by each animal (m^3/day)	0.01
Yearly produced waste (m^3/day)	34295
Total solid (%) [5]	9.5
Volatile solid (%) [29]	4.355
Liquid dairy manure	
Main components of liquid dairy manure [26]	(% TS)
Carbohydrates (cellulose, hemicellulose, lignin, and starch)	90.2
Fat	1.5
Protein	8.3
Elemental composition of manure (typically dry)	See [30]
Biogas	
Biogas yield (m^3/t ODM) [5]	300
Main components of biogas:	**Composition (%)**
CH_4	50-75
CO_2	25-50
N_2	0-10
H_2	0-1
H_2S	0-3

2.2. Process design

In this process, the degradable materials were decomposed in the absence of oxygen and converted into CH_4, CO_2, and other gases [5, 13, 31]. Table 1 shows the biogas compounds from the anaerobic digestion process. The PSM was chosen to model the anaerobic digestion process. The model, developed by Karthik Rajendran [26], is a library model estimating the productions of the anaerobic digestion process from any organic feedstock in different process conditions. This model includes 46 reactions (13 reactions of hydrolysis and 33 reactions related to acidogenic, acetogenic, and methanogenic stages) and inhibitions such as pH, ammonia concentration, load rate, retention time, and ten calculator blocks. The model has investigated biogas reactors working in 55 °C thermophilic conditions. Due to the comprehensiveness of the model (in terms of reactions, kinetic, stoichiometry, and inhibition factors), it was selected as the implemented model. Aspen Plus software was used to simulate the process. Making the right choice of the thermodynamic model is crucial since its inappropriate selection affects the calculations and can yield results ranging from non-optimal to catastrophic. The thermodynamic model NRTL is chosen as the property package. The reason for this selection is its ability to compute the activity coefficient and the molar fraction [13]. The heater was also used to increase the feed temperature up to 55 °C. All the kinetic reactions are followed by the power-law model and are represented by first-order kinetics. Acid-base reactions are carried out as equilibrium reactions.

The Aspen Process model is shown in Figure 1. Apparently, the digestion process takes place within two phases: the first phase is hydrolysis in which all of the reactions were carried out by stoichiometric and based on the extent of the reaction inside a stoichiometric reactor. The second stage comprised acidogenic, acetogenic, and methanogenic steps. The output of the first reactor is carbohydrate, lipid, and protein monomers added to the continuous stirred tank reactor (CSTR). At real biogas power plants, the same reactor model is used. In this reactor, the feedstock is continuously mixed, entered, and exited. Acidogenic, acetogenic, and methanogenic reactions were operated based on kinetic reactions. Residence time was chosen as the design and reactor simulation parameter. The CSTR reactor has one source stream and two product streams: biogas and digestate. The flow of biogas contains inert gases which must be separated by the splitter component during the separation process. Feedstock characteristics and process parameters such as temperature, pressure, load rate, and retention time are shown in Tables 2 [13, 20, 32]. Since the degradation of lignin is highly complicated and time-consuming, it is considered an inert component.

Table 2. Liquid dairy manure composition used as feedstock and presumed operational parameters in the Aspen Plus model

Component (dry composition)	
Cellulose	0.204
Hemicellulose	0.086
Glucose	0.257
Protein (glycine)	0.083
Triolein	0.015
Lignin	0.20
Inert	0.155
Assumed operational parameters of the process	
Reactor temperature (°C)	55
Reactor pressure (atm)	1
Hydraulic retention time (day)	15
Volumetric flow rate* (m^3/day)	93.96
* volume of manure and consumed water	

3. RESULTS AND DISCUSSION

3.1. Biogas production and compounds

Table 3 shows the biogas composition from the simulation. CH_4 and CO_2 make up 54 % and 37 % of biogas, respectively. The left 9 % includes other gasses: water vapor, hydrogen sulfide (H_2S), nitrogen, ethanol, and ammonia. About 95 % of feed is converted into digestate, and its compound is shown in Table 3. To investigate the effect of operational parameters and the composition of the liquid dairy manure on the composition of CH_4, sensitivity analysis is performed.

Table 3. Biogas compounds and characteristics resulting from the anaerobic digestion process

Biogas	
Biogas flow (m^3/day)	1306
Enthalpy flow (kW)	300
Density (kg/m^3)	1.12
Component	Composition (%)
CH_4	0.54
CO_2	0.37
H_2O	0.02
H_2S	0.009
NH_3	0.003
C_2H_5OH	0.004
H_2	0.002
Digestate	
Biogas flow (m^3/day)	105.3
Enthalpy flow (KW)	-16710
Density (kg/m^3)	1.003
Component	Composition (%)
Water	0.84
CO_2	0.01
Glucose	0.04
Cellulose	0.01
Hemicellulose	0.006
Ethanol	0.008
Protein	0.001
Ammonia	0.001
Inert	0.06

3.1.1. Effect of the hydraulic retention time and operational pressure

The Hydraulic Retention Time (HRT) indicates the time it takes to exchange the entire feedstock volume in the reactor and this is dependent on feed composition, reactor temperature and volume, and reaction kinetic rates. Due to longer HRT, the volume of the reactor and investment cost will also increase. On the other hand, lower HRT inhibits the growth and activity of microorganisms and increases the accumulation of VFAs [33]. HRT must be long enough so that the number of microorganisms removed with digestate would not exceed the number of regenerated microorganisms [34]. The residence time required for duplicating anaerobic bacteria is at least 10 days [6]. Besides, the operating pressure is one of the most critical process parameters affecting anaerobic digestion. The solubility of some components in the liquid phase depends on the pressure of the digester [25]. For example, since compounds such as CO_2 and triglycerides have acid-based reactions, they affect the pH level and inhibit the toxic effect of ammonia and non-ionized hydrogen sulfide [21, 35, 36].

As shown in Figure 2, the volume percentage of CH_4 increased by increasing digester pressure, but a higher HTR did not significantly affect the composition of CH_4 product. The reason for this trend is the availability of the substrates that have been reduced and decomposed. In addition, the increase in retention time increases the risk of VFA accumulation. This increase leads to a decrease in the pH level and inhibits the activity of microorganisms, especially methanogenic microorganisms. However, accumulation of VFAs does not always reduce pH levels, and it depends on the capacity of the digester and alkalinity of feedstock. It can be concluded that increasing the operating pressure will reduce the content of CH_4 product and cause the formation of the inhibitor effects on the process.

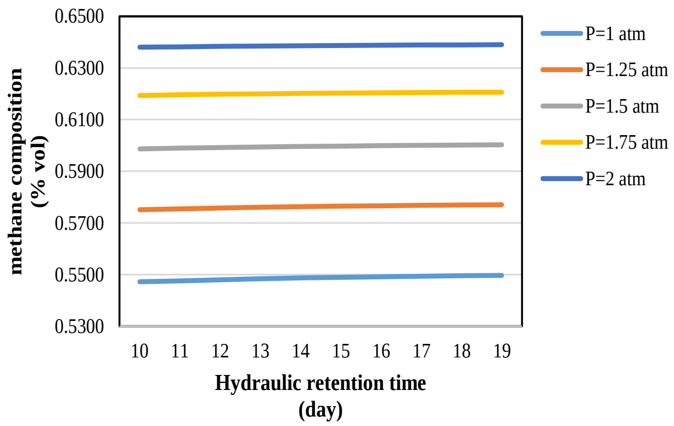

Figure 2. Effect of retention time on composition of biogas at different digestion pressures

3.1.2. Effect of carbon-to-nitrogen ratio

The C/N ratio shows the nutrient content of substrates [37]. Too low or high C/N ratios can inhibit or even stop the digestion process. At high C/N ratios, methanogenic microorganisms rapidly consume nitrogen in the substrates, and the lack of nitrogen disrupts the growth and duplication of microorganisms and reduces biogas efficiency [38, 39]. On the other hand, at a low C/N ratio, ammonia is produced from the decomposition of excess ammonia nitrogen, which is toxic to the health and growth of microbes and increases the risk of ammonia inhibition [13]. Since carbon is the main element of carbohydrates to study the effect of carbon-to-nitrogen ratio on the anaerobic digestion process, carbohydrate is considered a representative of carbon [27]. Carbohydrates are substances resisting decomposition; therefore, pretreatment is needed to hydrolyze them [40]. The addition of enzymes is one of the best ways to increase the efficiency of biogas production from carbohydrate-rich organic substrates [19]. Because of their smaller size, higher solubility, and greater dynamism of enzymes, they have more access to substrates and the digestion process occurs faster [41]. According to Figure 3, increasing the mass rate of carbohydrates and digestion retention time decreases the biomethane content. This declining trend results from the resistance of carbohydrates to degradation and decomposition by hydrolyzing microorganisms, reducing the rate of hydrolysis, and process efficiency. Also, increasing the mass rate of carbohydrates leads to the reduction and deficiency of nutrients required for the activity of microorganisms and also, it increases the production rate of VFAs and long-chain fatty acids resulting from the acidification process [42].

3.1.3. Effect of fatty acids

The concentration of fatty acids has a direct effect on digester stability [37]. Fatty acids and glycerol are intermediate components produced by fat (lipids) hydrolysis. First, Glycerol is decomposed into propionic acid during the acidification process and then, decomposed into hydrogen, CO_2, and acetic acid during the acetogenic process along with long-chain fatty acids [19]. Finally, hydrogen and CO_2 are converted into CH_4 during the hydrogen utilizing step reactions and acetic acid during acetoacetic reaction [12]. Due to the high degradability of fat-rich substrates, the yield of biogas production from them is high [42]. The potential for CH_4 production from this biomass is 170 % and 111 % higher than feedstocks, which are rich in carbohydrates and proteins, respectively [43]. However, excessive concentrations of fatty acids reduce the pH level [6, 42]. Figure 4 shows the effect of fat mass flow rate on CH_4 composition and volume flow at different HRTs. As can be seen, by increasing the amount of lipid, the CH_4 composition would be modified. The upward trend is due to more volatile organic substrates, which are available to microorganisms, and the high alkalinity of dairy manure preventing excessive accumulation of VFAs.

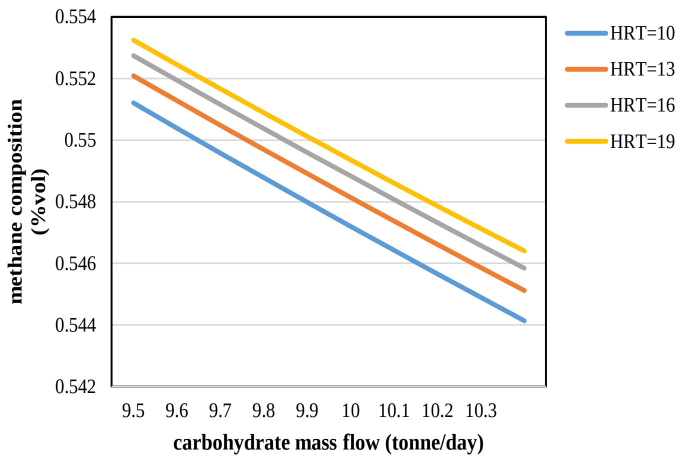

Figure 3. Effect of carbohydrate rate on the composition of biogas at different retention times

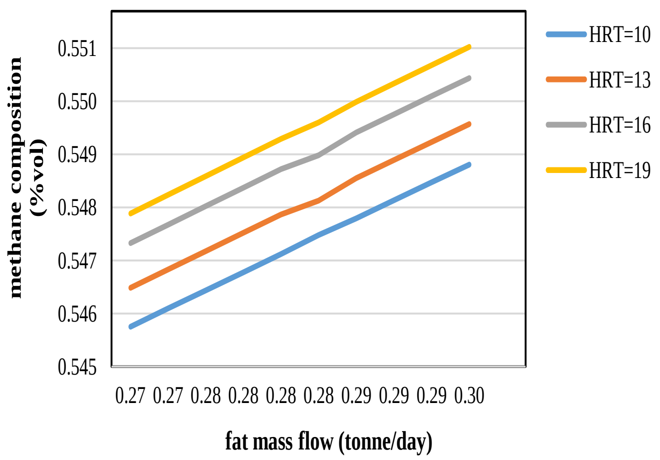

Figure 4. Effect of lipid on biogas composition of at different retention times

3.1.4. Effect of ammonia

The amino acids produced during the process of protein hydrolysis are converted into ammonia by Stickland reactions. Ammonia is essential for the growth and nutrition of bacteria, but its excessive concentration interferes with the activity of methanogenic bacteria and can be perceived as an inhibitory parameter for the process [6, 44]. According to findings, protein-rich biomass is not a viable resource for biogas production, and even it can stop the anaerobic digestion process [45]. According to Figure 5, the addition of protein increases the biomethane content. Due to the high concentration of dairy manure ammonia (derived from livestock urine), the ammonia obtained from protein breakdown intensifies the ammonia inhibition effect and reduces the biomethane purity. One way to reduce the concentration of ammonia is the addition of calcium hydroxide ($Ca(OH)_2$), magnesium hydroxide ($Mg(OH)_2$), and sodium hydroxide (NaOH) [46-48]. The concentration of free ammonia is directly related to temperature, and temperature control affects the dissolution rate of the ammonia component. Increasing the temperature increases the solubility of the ammonia and increases the risk of ammonia inhibition [46].

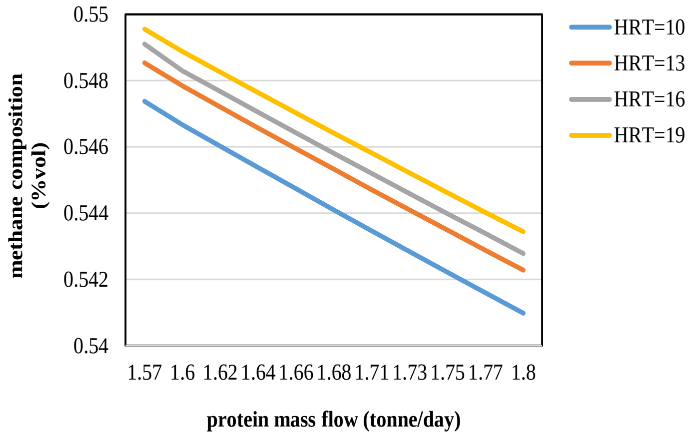

Figure 5. Methane composition as a function of mass protein rate at different retention times

3.1.5. Effect of water

The balanced amount of dry matter and water in the feedstock are vital factors in the design of the anaerobic digestion process. The water content of feedstock affects the growth rate of cells. If the water content is too high, less dry matter will be available to microorganisms, and the process will not be economical. Also, if the amount of water is too low, cell growth will be delayed and the material conversion becomes a limiting factor [49]. Figure 6 show the decline in biomethane content as the water rate increases.

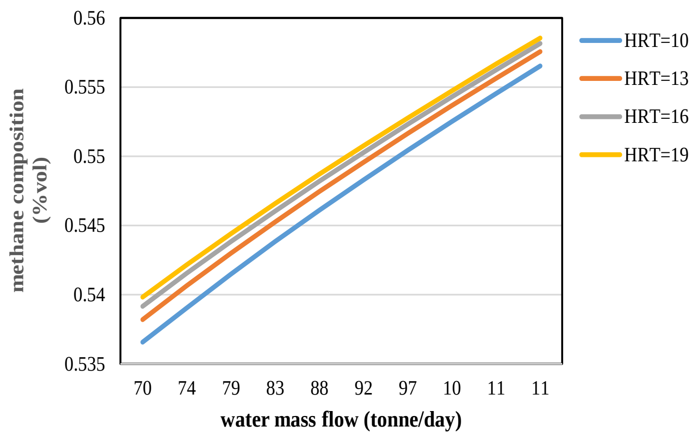

Figure 6. Effect of protein on composition of biogas at different retention times

3.1.6. The effect of organic load rate on CH_4 gas production

The OLR represents the amount of volatile solid mass entered into the digester per unit of time. This parameter is often defined as the mass flow rate of volatile solids (e.g., kg VS per day). According to Fig.7, the addition of the OLR led to a decrease in CH_4 composition. This declining trend is due to the composition of liquid dairy manure and the operating conditions of the process. The major composition of liquid dairy manure is fiber (carbohydrates) and will have a more significant impact on the digestion process than other dairy manure compounds.

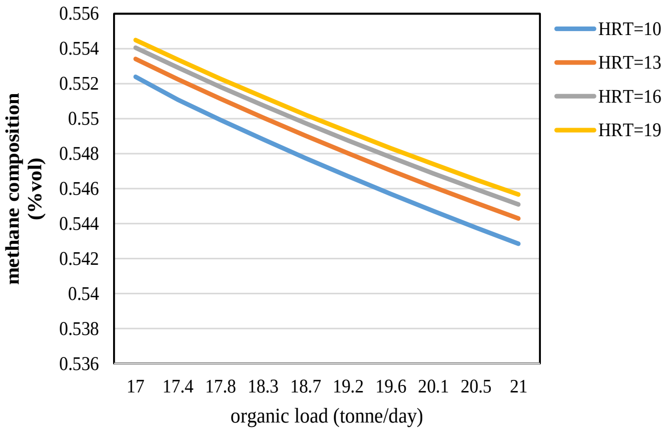

Figure 7. Effect of OLR on composition of biogas at different retention times

As discussed in previous sections, with increasing the mass flow of carbohydrates and proteins, the CH_4 composition decreased. Further, increasing the OLR has increased the methane production rate. The effect of increasing retention time is not significant and even negligible. It is due to the percentage of volatile organic matter in liquid dairy manure, which is lower than other degradable organic matter. As a result, the degradable substrates are consumed quickly, and the increase in retention time does not have a significant impact on the increment of CH_4 production efficiency.

4. CONCLUSIONS

Anaerobic digestion is one of the most appealing waste management systems in developing countries which, in addition to producing value-added materials (biogas and digestate), has significant impacts on reducing adverse environmental problems. The technical performance of this industrial unit was evaluated by selecting a typical industrial livestock farm using the anaerobic digestion system to manage the liquid dairy manure produced on dairy farms. The PSM model is one of the most comprehensive models; thus, it was implemented for anaerobic digestion at a thermophilic temperature of 55 °C and pressure of 1 atm. According to the results, biogas compounds include 54 % biomethane, 36 % CO_2, and around 5 % of trace elements, hence being in good agreement with the results of previous studies. Besides, 1306m^3 of biogas and 105 tons of digestate were produced daily under specified operating conditions. To examine the effect of process operating conditions and liquid dairy manure compounds on the efficiency of biomethane production, sensitivity analysis was performed. According to the results, since the solubility of some compounds at the liquid phase depends on digestion pressure, increasing this parameter reduces the ammonia inhibition and increases the biomethane composition. The increase in carbohydrates mass rate and OLR increased the inhibition of the fatty acids, and increasing the mass rate of protein increased the ammonia inhibition effect and reduced CH_4 composition. As demonstrated by the results, an increase in lipid mass rate increases biomethane composition. The increase in process water led to an increase in the composition of produced biomethane.

5. ACKNOWLEDGEMENT

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NOMENCLATURE

LHV	Lower Heating Value
PSM	Process Simulation Model
CSTR	Continuous Stirred Tank Reactor
HRT	Hydraulic Retention Time
VFA	Volatile Fatty Acids
OLR	Organic Load Rate
CHP	Combined Heat and Power

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