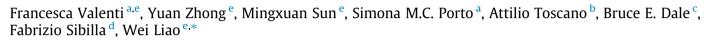
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Anaerobic co-digestion of multiple agricultural residues to enhance biogas production in southern Italy



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ABSTRACT

To valorize agricultural wastes and byproducts in southern Italy, anaerobic co-digestion of six feedstocks (citrus pulp, olive pomace, cattle manure, poultry litter, whey, and corn silage) was studied to produce biogas for renewable energy generation. Both batch and semi-continuous co-digestion approaches were adopted to carry out the investigation. The feedstocks were mixed at different percentages according to their availabilities in southern Italy. The batch anaerobic co-digestion demonstrated that six studied feedstock mixtures generated an average of 239 mL CH₄/g VS loading without significant difference between each other, which concluded that the feedstock mixtures can be used for biogas production. Considering the feedstock availability of citrus pulp and olive pomace in Sicily, three feedstock mixtures with the highest volatile solids concentration of citrus pulp (42% citrus pulp, 17% corn silage, 4% cattle manure, 8% poultry litter, and 18% whey; 34% citrus pulp, 8% olive pomace, 17% corn silage, 4% cattle manure, 8% poultry litter, and 18% whey; and 25% citrus pulp, 16% olive pomace, 17% corn silage, 4% cattle manure, 8% poultry litter, and 18% whey, respectively) were selected to run the semi-continuous anaerobic digestion. Under the stabilized culture condition, the feed mixture with 42% citrus pulp, 17% corn silage, 4% cattle manure, 8% poultry litter, and 18% whey presented the best biogas production (231 L methane/ kg VS loading/day). The corresponding mass and energy balance concluded that all three tested feedstock mixtures have positive net energy outputs (1.5, 0.9, and 1.2 kWh-e/kg dry feedstock mixture, respectively).

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1. Introduction

Biogas as one of renewable energy sources attracts increasing attention due to its capabilities of waste treatment and energy recovery. In Italy, biogas industry is steadily growing and more than 1300 biogas plants with the capacity of 8 GWh-e/year have been established on farm in last twenty years, making Italy the third largest biogas producer in the world after China and Germany (Fabbri et al., 2010; Torrijos, 2016). The feedstocks for Italian biogas plants are very much limited to animal manure and one or two dedicated energy crops (e.g., corn and wheat silages, beetroot, sugar cane, and sorghum) (Giuliano et al., 2013). However, the

https://doi.org/10.1016/j.wasman.2018.05.037 0956-053X/© 2018 Elsevier Ltd. All rights reserved. agriculture in Italy is diverse, particularly in southern Italy. For instance, Sicily is the largest agricultural land in Italy with more than 6 million tons per year agricultural commodities, in which more than 55% are from permanent crops, about 35% from vegetables and fruits, and 6% from greenhouse products (ISTAT, 2015). Corresponding, multiple agricultural residues are generated with different quantities. Citrus pulp, olive pomace, cattle manure, poultry litter, whey have been identified as the main residues available in Sicily as well as other Mediterranean regions (Valenti et al., 2018). Moreover, particularly in Sicily, the disposal of the large amount of wastes (agri-industrial by-products) coming from both citrus juice and olive oil industries (Valenti et al., 2017a, 2016) actually represents a crucial problem in terms of environmental sustainability and costs (Valenti et al., 2017b). Therefore, anaerobic digestion (AD) of multiple feedstocks is urgently needed to utilize the wastes for bioenergy production and reduce the environmental impacts of waste disposal in southern Italy.





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Anaerobic co-digestion of different organic residues has been widely investigated to enhance digestion performance of biogas production and solids reduction (Gou et al., 2014; Liu et al., 2009; Mata-Alvarez et al., 2014). The most common co-digestion scenario is that a main basic feedstock (e.g., animal manure or sewage sludge) is mixed with a minor amount of a secondary feedstock (e.g., crop residues, silage, and food wastes) to feed the digester (Aboudi et al., 2017; Kurahashi et al., 2017; Lehtomaki et al., 2007; Zhang et al., 2017). Only a few studies have reported that multiple feedstocks were used to carry out co-digestion. Tasnim et al (2017) ran a co-digestion on mixed cow manure, sewage sludge, and water hyacinth that had better gas production than the co-digestion of cow manure and kitchen wastes. Callaghan et al. (2002) optimized a co-digestion process using three feedstocks of cattle manure, chicken manure, and fruit/vegetable wastes, in which the effect of adding the three feedstocks to a digester with organic loading rates (OLRs) ranging from 3.2 to 5.2 kg VS/m³/day under mesophilic conditions (35 °C) for a HRT of 21 days, and the improvement in methane yield was demonstrated by increasing the proportion of fruit/vegetable waste from 20% to 50%. Muradin and Foltynowicz (2014) studied the economic performance of a commercial biogas plant receiving nine organic residues (corn silage, potato pulp, spent vinessa waste, fruit and vegetable pomace, cereals, plat tissue waste, municipal sludge, and soya oil). These studies demonstrated successful biogas production from multiple organic residues. Considering diversity and availability of agricultural residues and biomass, more and more biogas plants intend to use multiple feedstocks to improve their digestion performance, and request lab-scale testing approaches to determine the feasibility of such operations, as well as appropriate logistic and territorial studies to optimize the different feedstocks supply chain (Valenti et al., 2018).

Therefore, this study focused on applying batch and semicontinuous co-digestion approaches to investigate, for the first time, the effect of mixing six feedstocks (citrus pulp, olive pomace, whey, corn silage, cattle and poultry manure) available in Sicily on methane production for bioenergy generation. The aim of this study was to find environmentally friendly and economically feasible solutions to re-use and valorize majority of agricultural wastes and by-products in Sicily. Moreover, the approach and results could facilitate developing biogas production in other Mediterranean regions with similar sources of organic residues.

2. Materials and methods

2.1. Feedstocks and seed

The cattle manure and corn silage used for this study were taken from the Diary Teaching and Research Center at Michigan State University (MSU). The poultry manure was collected from the chicken farm at MSU. The whey was taken from a milk processing facility in Lansing, MI. The citrus pulp was obtained from processing oranges using a bench-scale orange juice processor (Black & Decker Citrus Juicer, Black & Decker, Beachwood, OH). The oranges were purchased from a local supermarket in East Lansing, MI. Two olive pomace samples (two-phase olive pomace and three-phase olive pomace) from two-phase and three-phase olive processing systems, respectively, were collected in Italy, and shipped to Michigan in coolers. The three-phase system applies a decanter to generate three fractions from olive: olive oil, olive husk, and olive mill wastewater (Alburguergue, 2004; Dermeche et al., 2013). While, the two-phase system just uses the extraction process to extract olive oil and generate a mixture of olive husk and olive mill wastewater as the wet pomace.

A blender (Waring Commercial Laboratory, Model No. 34BL97 (7012)) was used to reduce particle size of individual samples. After size reduction, all samples were stored at -20 °C prior to use. The characteristics of individual feedstocks were listed in Table 1.

The seed was the liquid filtrate after liquid/solid separation of the AD effluent from a commercial anaerobic digester located at MSU south campus ($24^{\circ} 41'N, 84^{\circ} 25'W$). The feeds for the digester were cattle manure and food wastes. The characteristics of seed were also listed in Table 1.

Six feedstock mixtures (FMs) (Table 2) were prepared for the batch co-digestion based on the amounts and availability of the agricultural wastes and by-products available in Catania, Sicily, Italy, with the consideration of seasonal variation of individual feedstocks. Cattle manure, poultry manure, corn silage and whey were fixed at 4.4%, 7.9%, 17.3%, and 17.9% (VS, w/w), respectively. The citrus pulp and olive pomace were varied between FMs. The VS ratio of two-phase olive pomace and three-phase olive pomace in the olive pomace was 1:2. The sum of citrus pulp and olive pomace was kept at $41.4 \pm 0.6\%$ (VS, w/w). The C:N ratios of the six FMs were 17.82 \pm 0.00, 18.74 \pm 0.02, 19.42 \pm 0.04, 20.02 \pm 0.05, 19.76 \pm 0.06, and 19.45 \pm 0.07, respectively.

2.2. Batch anaerobic co-digestion of feed mixtures (FMs)

The experimental apparatus included two 500 mL glass Wheaton bottles and a volumetric cylinder connected by tubes. One of the 500 mL bottles with rubber septa cap served as the reactor containing a 200 mL of the substrate. A needle was inserted into the rubber septa to collect the biogas. After the substrate was introduced, the reactor was purged by nitrogen gas for 10 min at a flow rate of 750 mL/minute to remove oxygen in the headspace. The reactors were then placed on a MaxQ 4000 benchtop orbital shaker (ThermoFisher Scientific Inc. Waltham, MA. U.S.A.), and cultured at 35 ± 1 °C and 150 rpm for 26 days. The other 500 mL bottle was used as the gas holder and initially filled with water. The reactor (through the needle), the gas holder and volumetric cylinder were sequentially connected by tubes. As biogas was produced, the biogas pushed the water from the gas holder into the volumetric cylinder. The volume of the water collected in the volumetric cylinder was recorded every day as the amount of biogas produced.

The substrate for the batch co-digestion was prepared by mixing the FM and seed at a VS ratio of 1:2 for 15 s in a blender (Waring Commercial Laboratory, Model No. 34BL97(7012)). Deionized water was added into the substrate to the targeted DM content of ~2.5%. The seed was used as the control. All tests were ran in duplicates. Methane content in the biogas was analyzed by GC. VS and pH of the substrates before and after the batch co-digestion were monitored as well. The methane production (mL methane/g VS loading) was expressed based on the total VS input.

2.3. Semi-continuous anaerobic digestion on selected FMs

The selected FMs from the batch co-digestion tests were used as feeds to run semi-continuous anaerobic digestion. The digestion was carried out in 750 mL bottles (reactor) with rubber septa caps. The working volume for all reactors was 500 mL with a headspace of 250 mL. Needles were also used to penetrate the rubber septa to release and collect biogas. Duplicate reactors were prepared for individual runs. The reactors were placed on a MaxQ 4000 bench-top orbital shaker (ThermoFisher Scientific Inc. Waltham, MA. U.S. A.) and cultured at 35 ± 1 °C and 150 rpm. The hydraulic retention time (HRT) was set at 25 days. The VS of all reactors was controlled in a range between 6.70 and 6.90 by dosing 30% (w/w) sodium hydroxide (NaOH) solution. The daily biogas production

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