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Research article

Strategies for the sustainable management of orange peel waste through anaerobic digestion

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ABSTRACT

The processing of oranges is a major industry worldwide and leads to the production of large amounts of orange peel waste (OPW). Energy production through anaerobic digestion of OPW is a promising option; however, the high content of essential oil, mainly composed of D-limonene, a well-known antioxidant, can cause the inhibition of the biological activity. In this paper, different pretreatment methods were tested (e.g. ensiling, aeration, thermal and alkaline treatments) to optimize the anaerobic digestion of OPW focusing on D-limonene removal. The raw and pretreated substrates were characterized and their biochemical methane production was measured. The results demonstrated the ability of some of the treatments to reduce D-limonene content up to 80%. A relatively high biomethane potential production of OPW (up to about 500 NmL $CH_4\ g^{-1}VS$) was measured. The importance of the acclimation of inoculum and the risk connected to the accumulation of inhibiting substances in the reactor is discussed.

1. Introduction

Orange juice is an important agro-industrial product at global level (production of 1.6 Tg was estimated during harvesting season 2015–2016 according to the United States Department of Agriculture (2016). Its production leads to the generation of a large amount of orange peel waste (OPW) that represents about 50–60% (wb) of the processed fruit (Martín et al., 2010). OPW includes rotten/discarded fruits, peel, seeds and membrane residues (Wilkins et al., 2007) and contains water (75–85% wb), simple sugars (glucose, fructose, sucrose; 6–8% db), polysaccharides (pectin, cellulose and hemicellulose; 1.5–3% db). It is characterized by a very low pH (3–5) and a significant presence of essential oil (EO), which is composed mainly by D-limonene (83–97%) (Bicas et al., 2008).

OPW presents a very high potential for bio-refining. Several valorisation routes have been proposed (e.g. production of fertilizers, pectin, ethanol, EO, chemicals, cattle feed, absorbent material (Ángel Siles López et al., Ángel Siles López et al., 2010; Boukroufa et al., 2015; Farhat et al., 2011; Tripodo et al., 2004, Ángel Siles López et al., 2010; Boukroufa et al., 2015; Farhat et al., 2011; Tripodo et al., 2004). Another possible valorisation option could be via the use of solid state fermentation (SSF) that until now has never been applied to OPW but has fully demonstrated its valorisation potential on other organic substrates (Ballardo et al., 2016; El-Bakry et al., 2015; Jiménez-Peñalver et al., 2016). Unfortunately, some of the aforementioned technologies are not mature or economically sustainable to be implemented in full scale and, therefore, OPW is typically used as a fodder after ensiling or drying or it is landfilled uncontrollably near the production sites (Ángel Siles López et al., Ángel Siles López et al., 2010; Tripodo et al., 2004; Ángel Siles López et al., 2010; Tripodo et al., 2004).

Energy production through anaerobic digestion is a promising and sustainable option for OPW; however, the high content of EO which mainly comprises D-limonene, a well-known antioxidant, can cause the inhibition of biomass activity (Forgács et al., 2012; Martín et al., 2010; Ruiz and Flotats, 2014). There are two possible options to overcome this limitation:

- The codigestion of OPW (fresh or ensiled) with other substrates (Anjum et al., 2017; Forgács et al., 2012; Martín et al., 2013) to dilute the D-limonene load;
- The OPW pretreatment to reduce the D-limonene content (Martín et al., 2010; Ruiz et al., 2016).

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Due to the seasonality of orange processing (mainly from December to March in South Europe), OPW is often ensiled (stored) allowing its use throughout the year. Ensiling is a process that was originally used for the preservation of cattle fodder and is nowadays used also for the substrates used in biofuels production. During ensiling, the materials is placed in a silo, it is then compressed to reduce porosity and to remove as much air as possible, and is then covered with a plastic sheet, or wrapped in a plastic film and baled. During ensiling, a spontaneous lactic acid fermentation occurs due to anaerobic conditions. That is, bacteria ferment the water-soluble carbohydrates mainly to lactic acid. Due to the pH drop, further microbial activity, and the consequent spoilage, is inhibited. The effect of OPW ensiling on the anaerobic digestion that is done using the ensiled wastes is not documented in the scientific literature.

Occasional measurements of methane generation from raw OPW appear in the literature. For example, Kaparaju and Rintala (2006) obtained from raw OPW about 450 NL $CH_4\,kg^{-1}$ VS in 30-day batch assays under thermophilic conditions. Gunaseelan (2004) also measured 450 NL $CH_4\,kg^{-1}$ VS from raw orange peels under mesophilic conditions (35 °C).

The pretreatment of OPW prior to anaerobic digestion aims primarily to reduce its D-limonene content. For example, Martín et al. (2010) pretreated OPW by steam distillation and obtained about 0.33 and 0.23 $Nm^3CH_4\,kg^{-1}VS$ under thermophilic and mesophilic conditions, respectively, in continuous laboratory scale experiments. Wikandari et al. (2015) obtained a maximum of 0.22 $Nm^3CH_4\,kg^{-1}VS$ after chemical leaching of D-limonene by hexane in batch reactors under thermophilic conditions. A similar procedure had been used by Negro et al. (2017). Forgács (2012) obtained a methane potential of 0.54 $Nm^3CH_4\,kg^{-1}VS$ after steam explosion in batch experiments. Ruiz et al. (2016) assessed the effect of three pretreatments (i.e. biological pretreatment by fungi, steam distillation and ethanol extraction). Methane production of untreated OPW was assessed to be about 0.35 $Nm^3CH_4\,kg^{-1}VS$, while no significant differences were observed after biological treatment. On the other hand, an increment up to 36% (0.47 $Nm^3CH_4\,kg^{-1}VS$) was observed after steam distillation and up to 34% (0.45 $Nm^3CH_4\,kg^{-1}VS$) after extraction of D-limonene with hexane. Calabrò et al. (2016) obtained 0.37 $Nm^3CH_4\,kg^{-1}VS$ from OPW treated by microwave assisted steam distillation.

Often pretreatment does not lead to a dramatic increase of methane production by raw OPW, while in some cases (Wikandari et al., 2015) there can be a significant reduction probably due to the generation of inhibitory compounds during pretreatment.

All the pretreatments mentioned above require a significant amount of energy and/or chemicals and therefore cost and energy balance for the entire process are important issues. Moreover, this balance should include the eventual further reduction of methane potential due to the loss of VS occurring during pretreatment that is not typically considered in the literature.

In this paper, several different pretreatment methods were applied to OPW following a fractional factorial experimental design. The methods were: ensiling, aeration, thermal and chemical. The ensiling lasted 37 days at room temperature; aeration was provided at a rate of 400 $L\,h^{-1}$ per kg of wet biomass; thermal treatment was performed at 70 °C and chemical (alkaline) treatment was achieved by adding $CaOH_2$ at 5 g 100 g^{-1} TS_{OPW}. All the chemical and physical pretreatments applied here had a duration of 24 h.

The goal of the pretreatment methods selected were to favour the disruption of oil sacs that contain the essential oils and/or to strip the D-limonene from the matrix. The pretreatment processes presented in this paper do not aim in the recovery of the essential oils but on the removal of D-limonene prior to anaerobic digestion so that to prevent its well documented inhibitory effects in anaerobic environments. Nevertheless, in future stages of the research, those same pretreatment techniques could be likely used to recover the EO for further utilization.

All the pretreatment methods tested do not require special equipment or an excessive amount of energy or chemicals. For example, ensiling is already routinely used in many agro-industrial installations, the $CaOH_2$ cost is low (around 1–2 €/MgOPW at this dosage) and also dosage addition and mixing can be performed with already existing equipment commonly found in biogas plants. The heat necessary for thermal treatment can be produced during biogas utilization via a combined heat and power scheme or after recovering residual heat from flue gas. The only exception is aeration, in which the energy needed to achieve the aeration rate used in the experiments, has been estimated to be around 15–20 kWh/Mg_{OPW}; however, the rate of aeration could be optimized in future studies to reduce those energy requirements. In addition, the effect of different types of inoculums were studied using a blocked design.

The novelty in this research is that the above four mentioned pretreatment methods were applied for the first time to OPW with the goal to reduce its EO content. This was done using an appropriate experimental design (factorial design).

2. Materials and methods

2.1. Sampling and pretreatment techniques

The orange peel waste used in the experiments was obtained from a real orange processing industry located near the city of Catania in Sicily; a total of about 7 kg of OPW, sufficient for the whole experimental activity, was collected and frozen (−20 °C for 1–4 months) in batches of about 1 kg. The aforementioned OPW sample was obtained after combining smaller grab samples from several locations of the original material pile (which was already homogeneous). It is noted that the aforementioned storage period was not expected to affect the biological activity of the samples. According to Pognani et al. (2012), freezing does not affect the biological activity of certain organic solid wastes for storage periods less than 20 weeks (and in some cases for periods up to 1 year). Prior to the experiments, the necessary amount of OPW was always thawed at room temperature.

The ensiling of OPW (duration 37 days) was carried out using two batches hermetically sealed. The first contained the material to be used as substrate to measure BMP; the biogas formed in this batch was removed three times per week. The other batch was a sacrificial one used to withdraw weekly the amount of OPW necessary for basic characterization, namely pH, TS, VS according to standard methods (APHA et al., 2012) and for the measurement of biogas production during ensiling.

Thermal pretreatment was carried out by keeping the sample at 70 °C for 24 h in an open vessel. Aeration was carried out by placing the OPW on a layer of quartz gravel used to improve the diffusion of the air (flow 400 $L\,kg^{-1}\,h^{-1}$) coming from a pipe immersed in the gravel; the duration of aeration was 24 h. Chemical treatment was carried out by dosing 5 g of $CaOH_2$ in 100 kg TS, while the material was hand stirred during and after the dosage to ensure homogenization. The contact time was set at 24 h.

Immediately after the end of all pretreatment periods, the material was tested for BMP and characterized as previously (pH, TS, VS).

2.2. Quantification of D-limonene

D-limonene extraction from orange peels was achieved by using