Enhanced biomethane production by co-digestion of mixed sewage sludge and dephenolised two-phase olive pomace

Rita Fragoso1, Ana Catarina Henriques2, Javier Ochando-Pulido3, Nicole Smozinski2 and Elizabeth Duarte1

Abstract
In this study, co-digestion of mixed sewage sludge from a wastewater treatment plant (WWTP) and partially dephenolised two-phase olive pomace (DOP) as a co-substrate was addressed with the aim of improving the biodigestibility of both substrates. The introduction of DOP into WWTP anaerobic digester facilities could significantly increase biomethane production and enhance the sustainability of both activities. An improvement in the system’s performance was supported by stability parameters: total alkalinity increased and stabilised with the addition of 5% v/v DOP, and the specific energy loading rate was maintained at 0.177 ± 0.03 d−1, which indicated better buffer capacity and stability in the bioreactor, and the possibility of enhancing the organic loading rate. In terms of average daily biogas production rate, an increase of 39% was achieved, up to 0.39 ± 0.11 L L−1 d−1. Moreover, there was a 40% and 37% improvement in specific methane production and methane production rate, respectively, up to 0.28 ± 0.02 L CH4 g TVS−1 and 0.26 ± 0.08 L L−1 d−1. In addition, the proposed strategy leads to an energy saving of 20,328.6 kWh year−1 at the WWTP as a result of the electric energy production surplus, corresponding to an annual saving of €3293.23.

Keywords
Anaerobic co-digestion, biowaste management, dephenolised olive pomace, sewage sludge, phenols

Introduction
At wastewater treatment plants (WWTPs), which are typically located in urban areas and operated and designed on the basis of bio-oxidation processes, a substantial portion of the potential chemical energy in wastewater streams in the form of carbon dioxide is unrecoverable. Moreover, to drive bio-oxidation of organic matter and ammonium and fulfil the legal requirements for effluent discharge, intensive energy input is needed. Furthermore, a huge amount of waste-activated sludge (WAS) is generated (Yang et al., 2019).

Under this framework WWTPs face a new paradigm that will have serious environmental and economic impacts, and anaerobic co-digestion of sewage sludge with other nutrient-rich substrates such as agrofood waste is a potential strategy that could be further explored to enhance energy recovery and nutrient balance (Pellera and Gidarakos, 2017). Such a strategy could improve the management of WWTPs and promote energy self-sufficiency (Zhang et al., 2014, 2017).

European Union (EU-28) policies concerning renewable energy systems set a fixed goal of supplying 32% of the European energy demand by 2030. At least 25% of all bioenergy in the future is expected to come from anaerobic digestion/anaerobic co-digestion (AD/AcoD) of wet organic materials such as sewage sludge, whole crop silage and agrofood waste, among others (Kathiotes, 2016).

On the other hand, olive oil production is expanding worldwide because of the health-giving properties of olive oil. Based
on FAOSTAT data, global olive production (FAO, 2019) was 20.9 m tons, which translates into 3.3 m tons of olive oil produced in the 2017–2018 season. In the EU-28, olive production was 12.9 m tons, which corresponds to 62% of the world’s olive production. This translates into 2.2 m tons of olive oil, which represents 66% of the global olive oil production. It is remarkable that the four leading countries (Spain, Italy, Greece and Portugal) contribute to 99% of olive oil production in the EU-28 (Espadas-Aldana et al., 2019; IOC, 2018). The remaining olive oil production outside the EU-28 (34%) is concentrated mainly in five countries: Morocco, Turkey, Tunisia, Syria and Algeria (FAO, 2019; IOC, 2018).

As a result of the olive oil sector activity, one of the most important and problematic biowastes in Mediterranean countries is generated. A huge amount of this is produced yearly during a short period (November–February) (Muscolo et al., 2019), thus causing serious management problems and causing a negative environmental impact, resource depletion and land degradation (Salome et al., 2015).

Two-phase olive oil milling is currently the most common technology used for olive oil production. It generates two types of olive biowaste: OP, also called olive mill solid waste (OMSW), and olive oil washing wastewater (OOWW) (Dermeche et al., 2013). Olive stones are very rich in lignocellulosic materials, and modern two-phase olive mills recover these from OP. The ratio of tons of OP to tons of olive oil is in the range 4:1 (Lama et al., 2017; Serrano et al., 2019). In the case of Spain, the largest producer, this amount is 2–2.5 m tons annually. The OP by-product is an excellent feedstock for biomass boilers, generating renewable energy for olive oil processing (Rodriguez et al., 2008). However, there are various reasons why the adoption of environmentally friendly approaches for the sustainable waste disposal of OP is difficult: small-scale mills are widely scattered; there is seasonal production only; the low price of the OP; limited storage life; and high transport costs (Caputo et al., 2003; Gunay and Karadag, 2015).

Suitable and sustainable treatment of OP is imperative because of its high organic load and humidity, low pH and the presence of inhibitory compounds such as polyphenols. Some solutions already proposed for the management of OP suggest it could be used for the absorption of heavy metals (Baccar et al., 2009; Bouzid et al., 2008; Malkoc et al., 2006), dyes (Akar et al., 2009) and phenols (Stasinakis et al., 2008), as well as composting (Stasinakis et al., 2008), as well as composting (Haddadin et al., 2009) and biogas production (Tekin et al., 2000), among others.

One of the most promising technologies for the treatment of the biowaste generated from the olive oil extraction process is anaerobic digestion (AD), because of its potential to recover bioenergy (Battista et al., 2013). However, anaerobic degradation of olive mill wastes (OP and OOOW) presents some drawbacks in relation to the high amount of barely degradable cellulosic materials contained therein, low pH and the presence of toxic substances, mainly phenols, long-chain fatty acids and ethanol. These substances lead to a decrease in pH in the anaerobic reactor and, thus, can inhibit the activity of methanogenic archaea (Camarillo and Rincón, 2012). Properly optimised co-digestion is necessary to increase microbial activity inside the digestor, enhancing the stability and performance of the process by supplying a carbon source and essential micronutrients (Gunay and Karadag, 2015). In this area, a number of studies have attempted the co-digestion of OP resulting from three-phase and two-phase production processes with several substrates. The following research should be highlighted: co-digestion with pig slurry (20%) (Orive et al., 2016); with cattle manure digestate (30% v/v) pre-treated with hydrogen peroxide (Siciliano et al., 2016); with cotton gin, wine and juice industry wastes (Pellera and Gidarakos, 2017); with OOOW (10% w/w) (Fezzani and Cheikh, 2010); and monodigestion of thermally pre-treated (180 min at 120°C) OP (Rincón et al., 2013).

In the present research work, a suitable management process for both OP generated in olive mills and mixed sewage sludge (MSS) from WWTPs is proposed. To this end, a series of bench-scale experiments were conducted to compare the system response in terms of biogas and biomethane production yield with co-digestion of MSS and DOP, as compared with the monodigestion of MSS. Prior to this, the OP utilised has been pre-treated for the extraction of high added-value polyphenols, which helps to revalorise the OP by-product from a material point of view and, subsequently, this process will facilitate its energy valorisation through the proposed anaerobic co-digestion process. The introduction of this by-product as a co-substrate in the anaerobic digestion of MSS may help enhance the circular bioeconomy target in both WWTPs and olive mills. This objective would be favoured in olive oil producing regions where small-scale mills are situated in the vicinity of WWTPs. To the authors’ knowledge, there is no previous research work that has focused on the use of destined and partially DOP as a co-substrate in the anaerobic co-digestion of MSS produced by urban WWTPs.

**Material and methods**

**Source of samples and pre-treatments**

Sewage sludge samples were collected from a WWTP located in Lisbon (Portugal) with a total treatment capacity of 50,000 m³/day of wastewater mainly derived from municipal sources, which corresponds to 210,000 in terms of population equivalent (unit per capita loading). The sewage entering the plant undergoes grit removal before primary sedimentation (PS), followed by an activated sludge treatment process. The sludge from PS is thickened and then homogenised with WAS that has been previously thickened by dissolved air flotation. The MSS has an average proportion of 40% PS:60% WAS (v/v).

OP samples were taken from an olive oil mill in the Andalusian province of Granada (Spain), one of the main olive and olive oil production regions worldwide. The mill from which the samples were taken operates with the most up-to-date two-phase olive oil production technology. Raw OP was taken in-situ directly from the exit of the horizontal centrifuges during the production process.

After collection, raw OP was readily subjected to phenol extraction with OOWW from the same olive mill, as shown in...
The following parameters were determined in accordance with standard methods (Baird and Bridgewater, 2017) for MSS, DOP, feed mixtures and digestate samples: pH; electrical conductivity (EC); mass density (ρ); total solids (TS); total suspended solids and volatile suspended solids (TSS, VSS); total chemical oxygen demand and soluble chemical oxygen demand (TCOD and SCOD); total alkalinity (TA); and total Kjeldahl nitrogen (TKN).

**Table 1.** Physico-chemical characterisation of raw mixed sewage sludge and partially dephenolised two-phase olive pomace used in the trials (Average ± SD; n = 10).

| Parameters          | MSS          | DOP          |
|---------------------|--------------|--------------|
| pH                  | 5.70 ± 0.15  | 5.35 ± 0.12  |
| EC [mS cm⁻¹]        | 1.89 ± 0.16  | 1.84 ± 0.14  |
| ρ [kg m⁻³]          | 1014         | 1785         |
| TS [g L⁻¹]          | 17.43 ± 1.05 | 217.70 ± 11  |
| TVS [g L⁻¹]         | 15.04 ± 0.85 | 209.39 ± 21  |
| TVS/TS (%)          | 86.30        | 96.18        |
| TCOD [g L⁻¹]        | 23.06 ± 0.98 | 256.22 ± 23.22 |
| SCOD [g L⁻¹]        | 2.36 ± 0.14  | 34.10 ± 3.30 |
| SCOD/TCOD (%)       | 10.20        | 13.30        |
| TOC [g L⁻¹]         | 8.72 ± 0.34  | 121.46       |
| TKN [g L⁻¹]         | 1.23 ± 0.16  | 2.95         |
| C/N                 | 7.0          | 42.0         |
| TPhs [mg GAE L⁻¹]   | 54.17        | 4220.77      |

MSS: mixed sewage sludge; DOP: partially dephenolised two-phase olive pomace; EC: electrical conductivity; ρ: mass density; TS: total solids; TVS: total volatile solids; TCOD: total chemical oxygen demand; SCOD: soluble chemical oxygen demand; TOC: total organic carbon; TKN: total Kjeldahl nitrogen; C/N: carbon–nitrogen ratio; TPhs: total phenols.

As described in Figure 1(b), to obtain a homogeneous feed stream, the MSS (for AD trials) or a mixture of 5% v/v of DOP and MSS (for AcoD trials) was grinded with a mechanical blender (P = 150 W, t = 2 min), followed by sieving (mesh size of 2 mm). This procedure aimed to avoid problems with clogging and prevent the formation of floating layers inside the digester.

**Physico-chemical characterisation**

The following parameters were determined in accordance with standard methods (Baird and Bridgewater, 2017) for MSS, DOP, feed mixtures and digestate samples: pH; electrical conductivity (EC); mass density (ρ); total solids and total volatile solids (TS, TVS); total suspended solids and volatile suspended solids (TSS, VSS); total chemical oxygen demand and soluble chemical oxygen demand (TCOD and SCOD); total alkalinity (TA); and total Kjeldahl nitrogen (TKN).
high-temperature thermally dephenolised OP by Serrano et al. (2019), who attained TCOD and SCOD values only about 7% lower as well as slightly lower pH. In our case, as the extraction procedure of OP with OOWW is performed at ambient temperature, there is no additional energy consumption. TS and TVS values obtained for DOP are also in accordance with the data reported by Lama et al. (2017) for thermally pre-treated two-phase OP.

With regard to total phenols (TPhs), DOP presented 4220.77 mg L\(^{-1}\), a value lower than those reported by other authors (Alagoz et al., 2015; Serrano et al., 2017, 2019). This value is 44.6% below that of fresh OP, which was 7613.57 mg L\(^{-1}\), thus 3392.8 mg L\(^{-1}\) total phenols were extracted from the pomace during pre-treatment. This value is significantly lower than the 10,000 mg L\(^{-1}\) reported by Maragkaki et al. (2017) as the limit above which TPhs concentration is considered highly inhibitory for microorganisms.

**Anaerobic mono-digestion vs co-digestion experiments**

The AD bench-scale unit used in this study consisted of a semi-continuously stirred tank reactor (CSTR) with 16 L total volume and 11.3 L working volume. The system was provided with a feed pump (Watson Marlow – 60 rpm), an electric blade stirrer (Velp Scientifica ES overhead stirrer) and an external jacket coupled with a thermostat and thermal blanket with an accuracy of ±0.5°C, which served to keep the temperature range at 35.5 ± 1.8°C. In addition, the biogas volume production rate was measured by a gas flow meter (Ritter Milligas counter), whereas the composition of the biogas stream obtained −%\(\text{v/v}\) of methane (CH\(_2\)), carbon dioxide (CO\(_2\)) and nitrogen (N\(_2\)), as well as hydrogen sulphide (H\(_2\)S) in ppm – was provided by a portable biogas quality analyser (LMSxi multifunction landfill gas analyser), with an accuracy of ±3 % and a detection range for H\(_2\)S of between 200 and 1500 ppm.

To compare the effect of the addition of DOP on the anaerobic bioreactor performance in terms of biogas and biomethane production, the experimental procedure comprised two main phases that were performed consecutively: first, AD with 100% MSS as feedstock (reference scenario); thereafter, AcoD using a feed mixture of 95:5 v/v of MSS:DOP.

The AD phase, after achieving steady state conditions, comprised two complete operating cycles. Once this phase was completed, AcoD with DOP was initiated and conducted for three consecutive runs. The first AcoD cycle served to observe the effect of the addition of DOP on the performance of the bioreactor through the gas production rate. Following the kinetic curve profile, the steady state condition was assumed when the daily average gas production rate varied less than 5%. Two additional AcoD cycles were carried out to observe the evolution of the system’s performance, thus allowing the comparison with the AD trial.

The total monitoring period of the study was 85 days, corresponding to 5 cycles, with a hydraulic retention time (HRT) of 17 days per cycle. During all the cycles, the temperature of the process was controlled and corrected by the heating system, which allowed the maintenance of appropriate mesophilic conditions (35.5 ± 1.8°C).

During the experimental period, inlet and outlet flow rates, reactor temperature and biogas production were measured daily (working days), whereas biogas quality was analysed once a week. The feed mixtures were fully monitored during the trials to control process performance, in terms of pH, EC, TS, TVS, TCOD, SCOD, TKN, C/N and TPhs. The digestates were also characterised to determine the removal efficiencies and the digester stability. The operational parameters – organic loading rate (OLR), biogas production rate (GPR) and methane production rate (MPR), specific methane production (SMP), total alkalinity (TA) and specific energy loading rate (SELR) – were determined twice per cycle.

The SELR (\(\frac{\text{QT COD}}{\text{V working}}\)) refers to the ratio between the daily average fed organic load (expressed in TCOD) and the active biomass inside the reactor (expressed in VSS), according to equation (1) (Pinto et al., 2016), and can be considered as an indicator of food to mass ratio (F/M). It expresses the rate of the methanogenic bioconversion into biogas/biomethane.

\[
\text{SELR} = \frac{Q \times [\text{TCOD}]_{\text{inlet}}}{[\text{VSS}] \times V_{\text{working}}} \quad (1)
\]

where:

\(Q\) = inlet flow rate (L d\(^{-1}\))
\([\text{TCOD}]\) = feed total COD concentration (g L\(^{-1}\))
\([\text{VSS}]\) = digestate volatile solids concentration (g L\(^{-1}\))
\(V_{\text{working}}\) = working volume of the reactor (L)

**Benefits of changing from mono-digestion to co-digestion for electric energy production**

An estimation of electric energy production surplus (\(\Delta E_p\)) resulting from the introduction of DOP as co-substrate as compared with MSS as a mono-substrate was performed based on a simplified approach. The energy consumption was considered to be the same in both scenarios.

The electric energy produced in each trial was determined based on the total biogas obtained and its methane content (v/v percentage). According to Singh and Basak (2018), one cubic metre of biogas containing 60% methane (v/v) on average, produces around 2 kWh of electric energy. Given that the biogas produced in the developed mono- and co-digestion sets of experimental runs provided different methane concentrations, it was necessary to adjust the lower calorific value for each of the different trials. The calculations of \(LCV_{\text{biogas}}\), based on the equations referenced in the literature by Von Mitzlaff (1988), were the following:

\[
LCV_{\text{biogas}} = \times \rho_{\text{CH}_4} \times LCV_{\text{CH}_4} \quad (2)
\]
For each trial, the amount of specific electric energy potential (EP) was calculated according to the following equation

$$ E_P = \frac{LCV_{\text{Biogas}} \times \eta \times \sigma}{\tau} $$

where

- $LCV_{\text{Biogas}}$: biogas low calorific value (MJ/Nm$^3$)
- $C_{\text{CH}_4}$: methane concentration (% v/v)
- $\rho_{\text{CH}_4}$: methane specific weight (0.72 kg/m$^3$)
- $LCV_{\text{CH}_4}$: methane low calorific value (50 MJ/kg)

For each trial, the amount of specific electric energy potential ($E_P$) was calculated according to the following equation

$$ E_P = \frac{LCV_{\text{Biogas}} \times \eta \times \sigma}{\tau} $$

where

- $E_P$: specific electric energy potential (kWh/kgTVS)
- $LCV_{\text{Biogas}}$: biogas low calorific value (MJ/Nm$^3$)
- $\eta$: biogas yield (m$^3$/kgTVS)
- $\sigma$: conversion factor of MJ to kWh (0.28)

The specific $\Delta E_P$ (kWh/kgTVS) was calculated using equation (4), whereas the $E_P$ for AD and AcoD was calculated as described above using the biogas and biomethane yield obtained (Table 3).

$$ \Delta E_P \text{(kWh/kgTVS)} = E_P \text{(AcoD)} - E_P \text{(AD)} $$

### Results and discussion

#### Bioreactor performance and stability during AD vs AcoD trials

The physico-chemical characteristics of the feed streams used in AD and AcoD, and of the digestates obtained are summarised in Table 2.

As can be observed in Table 2, the addition of 5% (v/v) DOP per litre of MSS led to a slight increase (4.5%) in the OLR of the blend. Specifically, the OLR was set at $0.90 \pm 0.02 \text{ gTVS L}^{-1} \text{ reactor d}^{-1}$ during anaerobic mono-digestion experiments with MSS (lasting two complete cycles), whereas it was increased to $0.94 \pm 0.06 \text{ gTVS L}^{-1} \text{ reactor d}^{-1}$ during AcoD runs when DOP was added. In any case, the established OLR value complied with the recommendation reported by various researchers that feeding the anaerobic digester at OLR higher than $1.00 \text{ gTVS L}^{-1} \text{ reactor d}^{-1}$ should be avoided (Battista et al., 2015; Serrano et al., 2019; Stoyanova et al., 2017).

As explained in section Material and Methods, the last two AcoD cycles were performed to examine the subsequent response
of the system under the established conditions. Bioreactor performance was assessed based on biogas production and quality, along with digestate control parameters.

As a result of the addition of 5% (v/v) DOP co-substrate to the MSS substrate, there was a 57.1% improvement in the C/N ratio, reaching a more adequate balance (C/N = 11) that was mainly due to the DOP contribution in terms of carbon content, whereas a decrease (22.0%) in TKN was measured. The TVS/TS and SCOD/TCOD ratios also increased by 3% and 30%, respectively, indicating significant availability of organic matter for the anaerobic co-digestion process. The blend of 5% (v/v) DOP and MSS ensured 89% TVS/TS content in the feed stream. On the other hand, as expected, there was a higher amount of TPhs (around 4.5 times) due to the addition of DOP to the MSS.

As far as AD runs were concerned, the digestates obtained showed a reduction of 18% in TVS if compared with the values of the feed stream (MSS); it is important to note that this value was enhanced up to 27% in AcoD runs with DOP as the co-substrate. This implies that almost twice the TVS content is biodegraded when DOP is added to the feed blend.

Another important aspect is that TCOD biodegradability was enhanced by 37.3% when the process was performed under the AcoD regime. The reduction of TCOD in AD experiments was 13%, whereas it achieved up to 43% in AcoD, representing a twofold increase in this best scenario. Moreover, with regard to TPhs concentration, there was an increase of 86% in removal under the AcoD regime.

The effect on the performance and stability of the MSS anaerobic co-digestion process when DOP is incorporated as a co-substrate is summarised in Table 3.

As can be observed from Table 3, the average daily GPR during the proposed anaerobic co-digestion regime rose as high as 0.39 ± 0.11 L L⁻¹ d⁻¹, which implied a significant increase, equal to 39.3%, when compared with the AD operational phase fed with MSS as a mono-substrate (GPRAD = 0.28 ± 0.10 L L⁻¹ d⁻¹). Furthermore, in terms of SMP (L CH₄ gTVS⁻¹) and MPR (LCH₄ L⁻¹ d⁻¹), the blend of MSS and 5% (v/v) DOP provided considerable enhancements, equivalent to 40% and 37%, respectively.

It is important to highlight that the GPR value attained in the proposed co-digestion process of mixed MSS and DOP is three times higher than that obtained by Serrano et al. (2017, 2019) for high-temperature thermally treated OP. Moreover, research developed by Pellera and Gidarakos (2017) using three-phase OP and inoculum from mesophilic anaerobic sludge digestion achieved SMP in a range 1.7 times lower than the yield obtained in this research.

Figure 2 shows the GPR and MPR for AD (a) and AcoD (b). Figure 2(b) includes the AcoD acclimatisation period (see section Material and Methods). It can be seen that the system responds...
well to famine periods, because during non-feeding days (data points with a yellow solid line around the marker) there is still biogas production, and production increases when feeding takes place. It should be mentioned that during the first HRT of AD there was a clogging event on the 11th day and that days 12–16 correspond to a weekend followed by bank holidays. Testing reactor performance under these circumstances can give an idea of how the process will perform under real scale conditions.

As can be observed in Figure 2(b), the microbial consortia in the bioreactor were not only able to tolerate the addition of DOP to the MSS, they also responded positively and were able to adapt in a stable way to this new co-substrate. The system was not affected by the phenol concentration introduced by the addition of DOP. This can be seen by the enhancement of bioconversion observed when the shift to the co-digestion process took place (Figure 2(b)), corroborated by the increase in GPR and MPR yields (~ 40%) during various subsequent HRTs if compared with the mono-digestion phase (up to day 34).

In addition, the stability patterns of the AD and AcoD processes are supported by the data shown in Figure 3, in which key physico-chemical parameters with regard to the bioreactor performance during operation are reported: the pH of the feed stream and of the digestate, the TA of the digestate, the SMP (LCH₄ gTVS⁻¹) and the SELR of the system.

As can be noted in Figure 3, a major buffer capacity was attained in the bioreactor when DOP was added as a co-substrate, supported by an increase in and stabilisation of TA values when the shift from AD of MSS to AcoD with DOP was made (Athanasoulia et al., 2012; Maragkaki et al., 2017). TA values for the AcoD trial were very similar to those reported by Serrano et al. (2019) for the same OLR range in a mesophilic CSTR, indicating process stability during the AcoD process. This was also supported by the narrow and stable pH values measured in the digestate during the whole AcoD phase (7.02 ± 0.07).

Furthermore, calculation of SELR during AD and AcoD trials allowed comparison of system stability. As mentioned in Materials and Methods section SELR expresses the rate of the methanogenic bioconversion into biogas/biomethane. According to Evans et al. (2016), SELR should be kept below 0.4 d⁻¹. If this capacity is exceeded, the digester might become unstable due to the rate of acidogenesis outpacing the rate of methanogenesis. Values higher than 0.4 d⁻¹ indicate instability among the microbial consortia biomass and with regard to loading the feed mixture. As can be seen from Table 3 and Figure 3, system SELR was maintained at 0.122 ± 0.02 d⁻¹ during AD of MSS as a mono-substrate (reference scenario), and 0.177 ± 0.03 d⁻¹ during the AcoD phase of MSS with the addition of 5% (v/v) DOP, indicating better buffer capacity and stability in the bioreactor. The fact that during AcoD the SELR was less than half of the advisable maximum value (0.4 d⁻¹) suggests that the DOP quantity in the feed mixture may be increased to 10% without risk of digester instability or failure.

The results achieved in this study can contribute to overcome the main challenges posed by the sewage sludge anaerobic digestion process, that is, improvement in biodegradability and enhancement of the methane yield. In fact, finding suitable co-substrates and optimum operating conditions are among the major challenges for biogas plants (Orfanoudaki et al., 2019; Siddique and Wahid, 2018). The type and structure of substrates together with their biodegradability are the key factors for methane production (Hagos et al., 2017). To overcome these barriers, the introduction of a co-substrate such as DOP with a C/N and a
SCOD/TCOD in the order of 6 and 1.3 times higher than that of MSS, respectively, would help enhance biomethane production. In the present work, co-digestion of DOP improved CSTR performance and enabled it to operate at a higher OLR. The co-digestion regime utilising MSS with added DOP ensured improved nutrient balance and process stabilisation, thus increasing biomethane yield (Hagos et al., 2017; Shrestha et al., 2017).

**Benefits of changing from mono-digestion to co-digestion for electric energy production**

The specific $\Delta Ep$ (kWh/kg TVS) resulting from changing to an AcoD regime was calculated as described in Materials and Methods section, using the biogas and biomethane yield obtained (Table 3).

$$\Delta Ep = E_p(AcoD) - E_p(AD) = 0.774\text{kWh kg TVS}^{-1}$$

This result can be applied to estimate the impact of changing to the proposed AcoD process at real industrial scale in a WWTP. For this, a standard WWTP serving approximately 100,000 inhabitants was considered in which an AD bioreactor with 1800 m$^3$ of active volume was implemented.

The following assumptions were considered:

1. The sludge from PS is thickened and homogenised with WAS, previously thickened by dissolved air flotation. The MSS has an average proportion of 40% PS:60% WAS (v/v);
2. The average HRT is 17 days;
3. A medium-sized two-phase olive mill processes 2000–3000 mtons of olives per year;
4. The ratio of olive mtons to DOP mtons is 5:4 (Lama et al., 2017; Serrano et al., 2019).

Considering the OLR previously established, that is 0.90 kg TVS m$^{-3}$ reactor d$^{-1}$ for AD and 0.94 kg TVS m$^{-3}$ reactor d$^{-1}$ for AcoD, adopting AcoD leads to an improvement of 0.04 kg TVS m$^{-3}$ reactor d$^{-1}$. Therefore, for the 1800 m$^3$ bioreactor mentioned in the assumptions there is an extra daily loading of 72 kg of TVS. Once the $\Delta Ep$ is 0.774 kWh kg TVS$^{-1}$, this would result in a daily surplus of 55.69 kWh, which corresponds to 20,328.6 kWh year$^{-1}$. If the price of electricity is considered to be €0.162 per kWh, the annual saving would be €3293.23.

As can be observed from the results obtained, the surplus of electric energy production ensured by the introduction of only 5% DOP indicates that its inclusion in the treatment is a promising strategy. The proposed process could result in not only positive environmental impacts but also provide a source of renewable energy that would reduce the specific energy consumption in WWTPs.

A smart and ‘green’ solution is proposed here for the management of both OP produced in olive mills and MSS generated in WWTPs. On the one hand, the age-old olive oil industry is concerned about making the whole process environmentally friendly, and this implies that the treatment and exploitation of the residues produced in the mills should be done in a technically feasible, environmentally clean and economically efficient manner. On the other hand, WWTPs face the challenge of improving their sustainability in terms of their carbon footprint and effecting a transition to greener forms of energy in the framework of the circular economy.

The results obtained here in relation to an improvement in system performance with regard to biomethane yield when MSS is co-digested with DOP, as compared with mono-digestion of MSS, are rather promising. The introduction of this readily available by-product as a co-substrate in the anaerobic digestion of MSS could be a key factor for the sustainability of both sectors in the next few years.

**Conclusions**

In this research work, the impact on anaerobic digestion performance when DOP is added to MSS is considered. The aim was to address the challenge of improving sustainability in terms of carbon footprint and a greener form of energy in the framework of the circular economy for both WWTPs and olive mills, especially in those regions that produce olive oil.

When 5% (v/v) DOP co-substrate is added to MSS, a 4.4% improvement in the OLR as well as a 57% improvement in the C/N ratio was achieved by the system. Moreover, the bioreactor performance was not negatively affected by the concentration of phenols introduced by the addition of DOP. The results obtained confirm not only a significant improvement in terms of biomethane production yield, but also the enhancement of system stability during the co-digestion of MSS and DOP, as compared with mono-digestion of MSS. The shift from a mono-digestion regime to a co-digestion one led to an increase in the CSTR biodegradation of 1.4 times in the same HRT (HRT = 17 days).

Furthermore, the average daily GPR, SMP and MPR of the system could be successfully increased by 39%, 40% and 37%, respectively, and this was ensured during three consecutive HRTs. This could allow a surplus of electric energy in terms of 20,328.6 kWh year$^{-1}$. The proposed process could not only have a positive environmental impact but could also provide a source of renewable energy that would serve to reduce the specific energy consumption in WWTPs.

The overall results obtained, indicating an electric energy surplus from the co-digestion of DOP and MSS, constitute a promising alternative for both OP from olive mills and MSS from WWTPs in terms of biowaste management in the framework of a transition to greener energy and economy decarbonisation, a key factor for the sustainability of both sectors in the near future.

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ORCID iD
Rita Fragoso https://orcid.org/0000-0003-2957-7340

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