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METHANE PRODUCTION IN THE CO-DIGESTION OF LANDFILL LEACHATE WITH DOMESTIC SEWAGE AND THE METHANOGENIC ACTIVITY OF FULL-SCALE UASB REACTORS

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Abstract

The study aimed to individually assess the specific methanogenic activity of the sludge of 10 UASB reactors from a full-scale domestic sewage treatment plant (STP) and the sludge behavior under conditions of anaerobic co-digestion of leachate with domestic sewage in dilutions (v/v) of 0.1%, 1.0%, 2.5%, 5.0%, 10.0%, 25.0%, 50.0%, 75.0% and 100%. In order to compare the influence of the activity on co-digestion sludge, a parallel test was performed with sludge coming from the treatment of swine wastewater under the same conditions as the test with STP sludge. Specific methanogenic activity results showed the difference between STP sludge conversion capacities at each reactor, as well as preponderant conversion routes, which can have several origins within the mesh of factors that affect the units. Co-digestion analyses pointed out a better adaptation of piggery sludge to higher organic loads, such as the viability of up to 50% of leachate in relation to treated sewage volume, while the STP sludge showed a more delayed response in methane production. The best co-digestion condition was 10% leachate. The study highlights the possibility of increased leachate fractions in the co-digestion with domestic sewage with methane production potential, with incorporation of sludge from agro-industrial wastewater treatment.

Keywords: agro-industrial sludge, SMA, co-treatment, wastewater, biogas.

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Introduction

Current sanitation models opt to manage the treatment and disposition of solid waste and domestic sewage by centralized processes (Chernicharo *et al.*, 2015; Stazi and Tomei, 2018). This has happened in conjunction with the spatial "peripherization" of these units, which are normally close to one another. On the other hand, it provides a combined treatment of effluents generated by domestic sewage in landfills (De Albuquerque *et al.*, 2018; Felipe *et al.*, 2018; Nascentes *et al.*, 2015). In Brazil, the use of Sewage Treatment Plants (STPs) by anaerobic processes has grown significantly and has currently been one of the most used biological processes (Chernicharo *et al.*, 2015; Stazi and Tomei, 2018), especially after the emergence of upflow anaerobic reactors known as UASB (Chernicharo, 2007; Van Haandel and Lettinga, 1994). This is because this process shows several advantages, such as low sludge production, low energy consumption, biogas production and lower area requirement, especially when compared with stabilization ponds.

The combined treatment of domestic sewage with landfill leachate is an interesting alternative to minimize the harmful effects of leachate on the environment and to enable the use of technologies in pollution control (Costa *et al.*, 2019; Nascentes *et al.*, 2015; Wu *et al.*, 2015), especially in developing countries with low access to services of sewage collection and treatment, and to adequate disposal of solid waste. However, according with Çeçen and Çakiroğlu (2001), some economic aspects should be considered, such as the viability of leachate transport to STP; the plant capacity to assimilate this effluent; the compatibility of the process with characteristics of this material, given that landfill leachate has high organic load, heavy metals and ammoniacal nitrogen, factors which can inhibit the sludge present in the treatment, diminishing the STPs' treatment capacity (Mojiri *et al.*, 2012; Renou *et al.*, 2008; Wu *et al.*, 2015). For these reasons, there is a great concern in regard to the complexity involving the adoption of biological treatment for landfill leachate, since its physical and chemical characteristics can compromise the metabolism of microorganisms (Mojiri *et al.*, 2012; Neczaj *et al.*, 2007; Renou *et al.*, 2008; Stazi and Tomei, 2018). The adoption of systems of co-digestion of the leachate coming from landfill and domestic sewage should be adjusted according to their characteristics, since the increment of leachate provides an increase of organic load and other substances that can be harmful to the treatment process. Thus, studies are needed to find fractions to be co-digested under optimum conditions of dilution and concentration for both effluents (Chernicharo *et al.*, 2015; Esposito *et al.*, 2012; Kawai *et al.*, 2012).

Studies of anaerobic co-digestion present satisfactory results as to the efficiency of biogas treatment and recovery with ratios of 1:9 of leachate in relation to domestic sewage volume. (Brennan *et al.*, 2017; Neczaj *et al.*, 2007). However, these values are variable in relation to leachate age, for older leachates tend to have higher concentrations of ammoniacal nitrogen and reduced organic matter (COD), which reduces the C/N ratio. As a consequence of these variations, Çeçen and Çakiroğlu (2001) establish that the leachate ratio in relation to domestic sewage cannot be higher than 20% when substrate volume is used as a basis for co-treatment, and that the COD concentration in the leachate do not surpass 50% of total COD applied to the treatment system. Nonetheless, it is important to highlight that there is a need for full-scale evaluations, because in including matters of the operationality of STPs, of seasonality and hydrodynamics, variations can be observed in the process and influence negatively on biogas production, or worsen the quality of the final effluent (Bakonyi *et al.*, 2019; Çeçen and Çakiroğlu, 2001; Hagos *et al.*, 2017).

As a response to these variations, the sludge present in treatment systems shows oscillations, which are reflected in the treatment conditions, and in the biological assimilation and bioavailability of the different effluents being treated, since the leachates commonly show a significant amount of recalcitrant compounds and inert fractions, which makes the microbial action difficult. (Bakonyi *et al.*, 2019; Nielfa *et al.*, 2014) The introduction of new sludge or its acclimatization are alternatives to attenuate these discrepancies around the behavior of the own sludge under anaerobic digestion processes (Khilyas *et al.*, 2017; Rodriguez-verde *et al.*, 2014).

In this way, the aim of the study was to evaluate (i) the methanogenic activity of the UASB reactors of a full-scale STP, as well as the (ii) methane production potential under conditions of anaerobic co-digestion of domestic sewage with landfill leachate and the (iii) introduction of piggery sludge (agro-industry) into the process.

Materials and methods

Full-scale UASB reactors

The STP operates with a flow rate of 76204 $m^3.d^{-1}$, possesses conventional preliminary treatment, screening followed by grit chamber and has 10 anaerobic reactors as biological treatment. Each reactor possesses dimensions of 6x4x4m of length, width and height, respectively. In addition to domestic sewage, the STP receives (in semi-continuous flow) the leachate from a landfill that has been in operation for 4 years, including the leachate of a dump in the process of being deactivated, which has operated for 30 years. Leachate in relation to affluent flow is approximately 1%. Sludge samples were collected in each reactor separately and collection point maximum height was 2 meters.

Specific methanogenic activity (SMA)

The specific methanogenic activity was evaluated, by means of batch tests, of the sludge present in 10 UASB (Upflow Anaerobic Sludge Blanket) reactors, which are part of a full-scale domestic Sewage Treatment Plant (STP).

Domestic sewage, acetate synthetic solution and glucose synthetic solution were used as substrate. The sewage was collected in the own STP and its characteristics are described in Table 1. The synthetic solutions were prepared using sodium acetate and glucose, with an equal concentration of 1 gCOD.L^{-1}, both with micro- and macronutrient solutions, following the protocol described by Leitão *et al.* (2009). Sodium bicarbonate was used as buffer (1.0 gCOD.g^{-1}) in the glucose synthetic solution.

Table 1. Physical and chemical characterization of leachate and domestic sewage.

Parameters	Sewage	Leachate
COD (mg.L^{-1})	506.67 ± 130.64	4693.33 ± 349.32
Alkalinity (mgCaCO3.L^{-1})	28.00 ± 11.31	425.00 ± 190.91
Total acidity (mgCaCO3.L^{-1})	30.00 ± 8.48	96.00 ± 130.10
TKN (mg.L^{-1})	37.82 ± 11.44	193.59 ± 39.83
Ammoniacal Nitrogen (mg.L^{-1})	14.18 ± 1.99	81.15 ± 1.74
Phosphorus (mg.L^{-1})	20.10 ± 6.61	30.94 ± 16.73

** ± standard deviation*.

Experiments were carried out by following protocols suggested by Angelidaki *et al.* (2009) using 500 mL capacity glass flasks, with 80% of the usable volume of each flask being filled, and in triplicate. In all conditions, a 1:1 ratio of organic load of substrate (gCOD) and sludge (gTVS) was maintained. After filled and sealed, the flasks were maintained at a temperature of 30º±2 ºC. Methane production was quantified daily using the liquid displacement method described in Aquino *et al.* (2007).

Methane production potential by the co-digestion of domestic sewage with landfill leachate

In order to evaluate the biodegradability of domestic sewage in the co-digestion with landfill leachate, as well the performance of the sludge against different conditions (tested dilutions), batch experiments were carried out by following protocols suggested by Angelidaki *et al.* (2009). The domestic sewage, as well as the leachate used were collected in the STP evaluated in the previous test. After collection, we carried out analyses of COD (Chemical Oxygen Demand) and the series of suspended solids (total, fixed and volatile), alkalinity, sulfide, pH, phosphorus and ammoniacal nitrogen for characterization (Table 1), following the protocol described in Standard Methods for the Examination of Water and Wastewater (APHA, 2012).

For this experiment, two distinct and separate biomasses were used so as to compare the influence of sludge on co-digestion. This first, stemming from the previously cited STP, was made using a blend of sludge collected in reactors with the best methanogenic activity results by using domestic sewage. The second sludge came from a swine wastewater biodigester (agro-industry). Analyses of solids were carried out to characterize the biomasses (Table 2) by following the protocol described in Standard Methods for the Examination of Water and Wastewater (APHA, 2012).

Table 2. Characterization of the sludge from a STP treating domestic sewage (STP sludge) and of sludge coming from swine wastewater biodigester (piggery sludge).

Parameters	STP Sludge	Piggery sludge (agro-industry)
TSS ($g.L^{-1}$)	44.96 ± 36.85	46.61 ± 1.94
VSS ($g.L^{-1}$)	28.96 ± 31.84	23.99 ± 9.00
FSS ($g.L^{-1}$)	16.00 ± 4.96	22.62 ± 11.12

TSS: total suspended solids; VSS: volatile suspended solids; FSS: fixed suspended solids.

Experiments were carried out in two steps. The first comprised dilutions of 0.1%, 1.0%, 2.5%, 5.0% and 10.0% of leachate in relation to domestic sewage, which were established in relation to the volume used. In this step, tests were carried out with the STP sludge only.

In the second step, dilutions of 5.0%, 10.0%, 25.0%, 50.0% and 75.0% of leachate in relation to domestic sewage were tested, as well as 100% leachate and 100% domestic sewage as standards. All dilutions were tested using biomasses coming from the STP's domestic sewage (STP sludge) and from the swine wastewater biodigester (piggery sludge originating from agro-industry) in parallel.

All conditions (dilutions) were evaluated in triplicate, where the 1:1 ratio was maintained for the organic loads of substrate (gCOD) and sludge (gTVS). Glass flasks with 300 and 500 mL capacity were used, which were filled by 80% of their usable volume. After filled and sealed, the flasks remained at ambient temperature (32±4ºC) and they were manually shaken twice a day. Methane production was quantified daily using the liquid displacement method described in Aquino *et al.* (2007).

Results and discussion

Specific Methanogenic Activity (SMA) of full-scale UASB reactors

The methanogenic activity of sludge from reactors of a Sewage Treatment Plant (STP) receiving approximately 1% of leachate (Fig. 1) showed different values for each evaluated reactor. In using domestic sewage as substrate, the conversion mean was 0.032 $gCOD\text{-}CH_4.gTVS.d^{-1}$, where 7 from 10 reactors evaluated presented an average of 0.019 $gCOD\text{-}CH_4.gTVS.d^{-1}$. When acetate was used as substrate, conversion mean for glucose was 0.054 $gCOD\text{-}CH_4.gTVS.d^{-1}$ and 0.085 $gCOD\text{-}CH_4.gTVS.d^{-1}$.

There is a very large variability of SMA values in relation to some factors, such as type of reactor, temperature of operation and test performance, type of substrate used and time of acclimatization or activation of sludge (Hussain and Dubey, 2017).

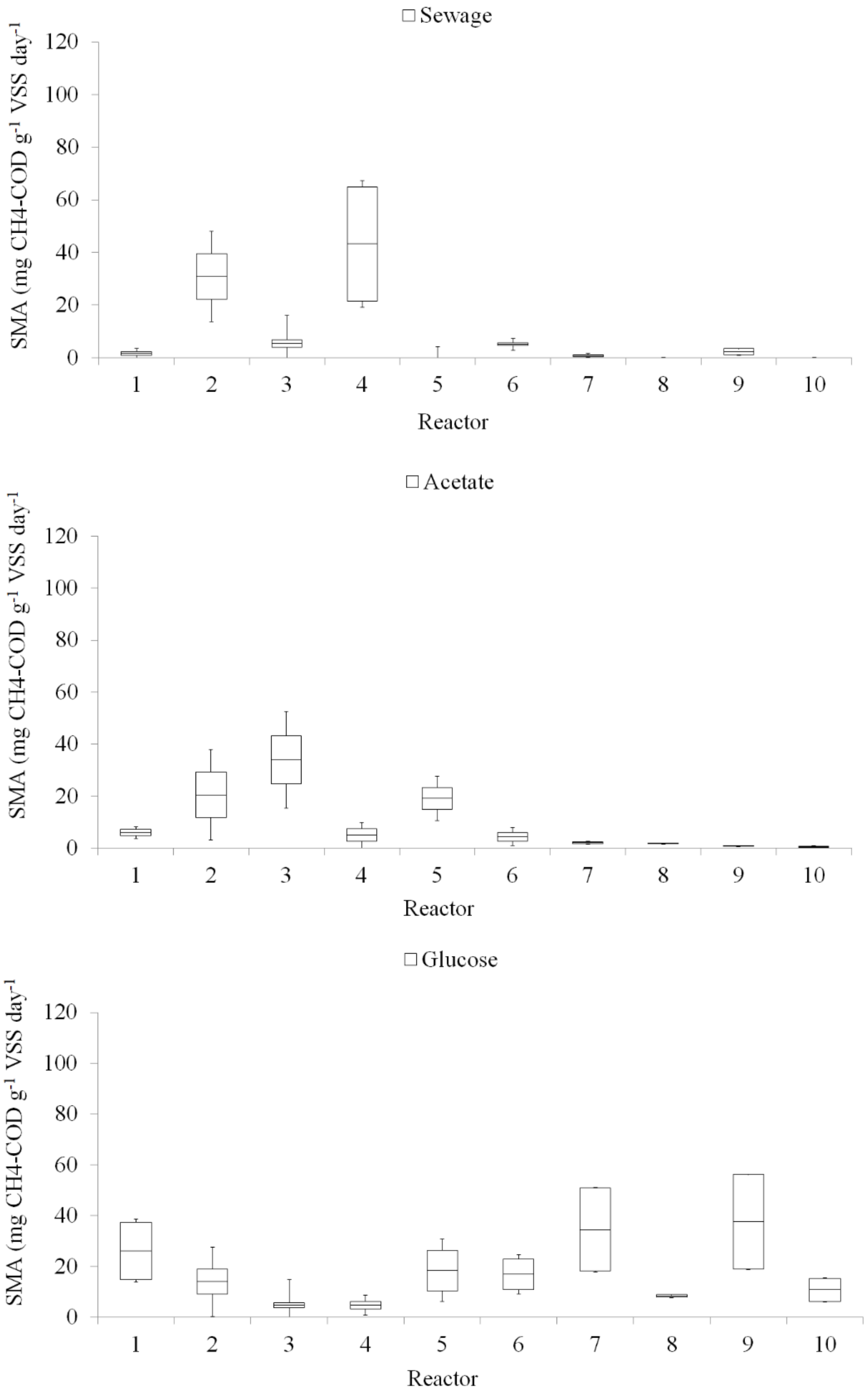

Figure 1. Specific methanogenic activity (SMA) of the sludge from reactors of the sewage treatment plant.

The sludge from full-scale anaerobic systems treating domestic sewage has values between 0.01 to 0.40 gCOD-CH_4.gVS.d^{-1}. Where lower activities are generally found in digesters and reactors that are simpler and with lower operational control (Hussain and Dubey, 2017; Van Haandel and Lettinga, 1994). Results found in the evaluation of reactors are consistent with simpler systems with low operational control, even when only considering the best conversion rates, which comprise values of 0.043 gCOD-CH_4.gTVS.d^{-1} (reactor 4, with sludge), 0.037 gCOD-CH_4.gTVS.d^{-1} (reactor 9, with glucose) and 0.034 gCOD-CH_4 .gTVS.d^{-1} (reactor 3, with acetate).

More robust systems, with greater operational control, usually present values closer to 0.40 gCOD-CH_4.gTVS.d^{-1}, as in results obtained by De Lucena *et al.* (2011), which evaluated sludge at different heights in anaerobic reactors in full-scale domestic sewage treatment, using a mixture of acetic, propionic and butyric acids as substrate, and obtained values between 0.29 and 0.37 gCOD-CH_4.gTVS.d^{-1}.

In observing results according with the substrate used, synthetics presented better SMA values when compared to tests with domestic sewage. This difference can be attributed to the characteristics of the own substrates - as when they are applied to the sludge, there is a difference as regards bioavailability - as well as to each substrate's relative homogeneity, which is higher in synthetic substrates, which are, in turn, ready for biomethanization (Hussain and Dubey, 2017). As to wastewater (sewage), it will demand from the whole anaerobic digestion process which precedes the specific activity of methanogenic groups. According to Khan *et al.* (2015), the methanogenic activity decreases when lower organic loads are applied, loads which they undergo in this system.

This oscillation of SMA in the STP sludge can also be attributed to hydraulic, operational and microbiological conditions and their preponderant conversion paths, as well as the characteristics of biodegradability and biological assimilation, which can be compromised by the composition of the effluents present in treatment (Leitão *et al.*, 2011; Zhen *et al.*, 2015). The configuration of anaerobic reactors provides a selective environment for the development of microorganisms, which are compiled from the reactors' physical structures until their operational dynamics, which dictates the spatial distribution of biomass. Gulhane *et al.* (2017) and Leitão *et al.* (2009) concluded that operational and environmental variations exist and will always exert an effect on anaerobic biological systems.

For Chernicharo *et al.* (2015), there are diverse factors to which STPs are subjected to keep a regular operational regime, and flaws are accumulated since the design period, in which they are not yet considered responsible for the hydraulic regime (daily and annual variations) for which they are designed. Thus, sometimes STPs will operate in overload conditions occasioned by undersized flow hydrograph. Another noteworthy aspect is the non-uniform distribution of effluent in the reactors, which can provoke detritus accumulation and sludge stagnation, create dead zones and form preferential flows, directly affecting the methanogenic activity of the sludge and treatment levels.

In addition to these mentioned factors, sludge sampling height in the reactor and time and stability of operation influence directly the SMA results (De Lucena *et al.*, 2011; Souto *et al.*, 2010). In reactors operated under stable conditions and with low concentration of VFA (volatile fatty acids), acetoclastic methanogens are predominant (McHugh *et al.*, 2003; Sun *et al.*, 2014; van Haandel *et al.*, 2014). Overall, it is observed greater archaea diversity in the start of processes in anaerobic systems, with diversity reduction when the system reaches a stable performance (McHugh *et al.*, 2003). Considering that some STPs carry out partial or total sludge exchanges from the reactor during their operation (Chernicharo *et al.*, 2015), it is possible to relate the results obtained in the present study to the sludge lifespan in the reactor. The best results for conversion via acetoclastic methanogenesis using acetate as substrate, were obtained in reactors with lower results when glucose was used as substrate (Fig. 1), while the highest values obtained with glucose were in reactors with lower results using acetate and domestic sewage as substrates. Possibly the reactors with the best activity with acetate and domestic sewage possess the most stable sludge and low archaea diversity, with acetate as the preferred route for methane production.

Methane production potential by the co-digestion of domestic sewage with landfill leachate

Results of accumulated methane production (Fig. 2) indicate that the methane production potential is directly proportional to the increase in the concentration of leachate in sewage, since the higher the concentration of leachate in the substrate composition, the higher will be its organic load (COD). This potential increase in the production of biogas is justified by the increase in biodegradable load during the process, given that the concentration of organic matter in the leachate varies from 0.5 to 40 g.L^{-1} of COD, while domestic sewage presents mean COD values around 0.6 g. L^{-1} (Bakonyi *et al.*, 2019; Naveen *et al.*, 2016).

In the first days of experiment, piggery sludge had a better methane production when compared with STP (Sewage Treatment Plant) sludge results, with an increasing methane production proportional to the increase of leachate in relation to domestic sewage until the 50% condition. Results of STP sludge presented low methane production in the first days and needed a period of adaption. This can be related to its low activity, given that the STP sludge had low SMA even when its own effluent of origin was used (Fig. 1). From the thirtieth day, the STP sludge had, under a 50% condition of leachate, an increase in production arriving at 0.20 gCOD-CH_4.L^{-1}.

This time difference in the response of methane production between piggery sludge and STP sludge, shows that the sludges are different as for capacity and adaptability. Piggery sludge has higher biological assimilation with high organic load effluents (Zhang *et al.*, 2014). Furthermore, after acclimatization, it is possible to increase the applied load even more. The STP sludge shows a late response (it needs acclimatization) and has a restricted conversion capacity. In this way, piggery sludge shows to be an alternative for a high conversion capacity in the face of the applied load amplitude.

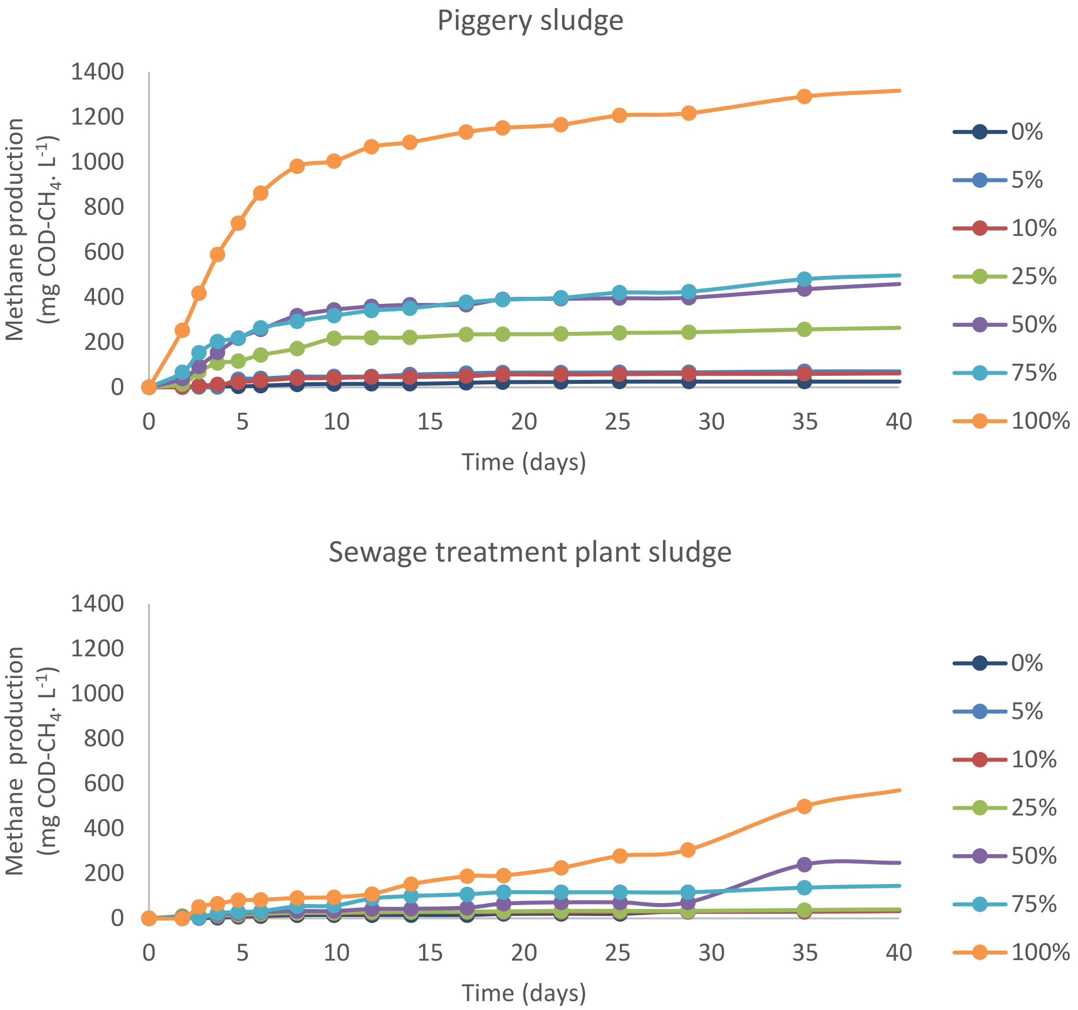

Figure 2. Production of biogas in the 40 days of tests of dilution conditions (v/v) at 0%, 5%, 10%, 25%, 50%, 75% and 100% of leachate for piggery sludge and STP (Sewage Treatment Plant) sludge.

Although the data point out good results as regards the capacity and adaptability of sludges in high organic load conditions, when the applied COD and converted COD relation is analyzed, it is possible to present the real capacity of sludge biomethanization with regard to substrates constituted for co-digestion (leachate and domestic sewage relation), as per Fig. 3. Results obtained using STP sludge presented methane production potential, with the addition of up to 10% of leachate in relation to the domestic sewage volume. In dilutions higher than 10%, the sludge had a drop in daily methane production. Differently, results using piggery sludge showed promising with above 10% leachate applications in co-digestion with domestic sewage.

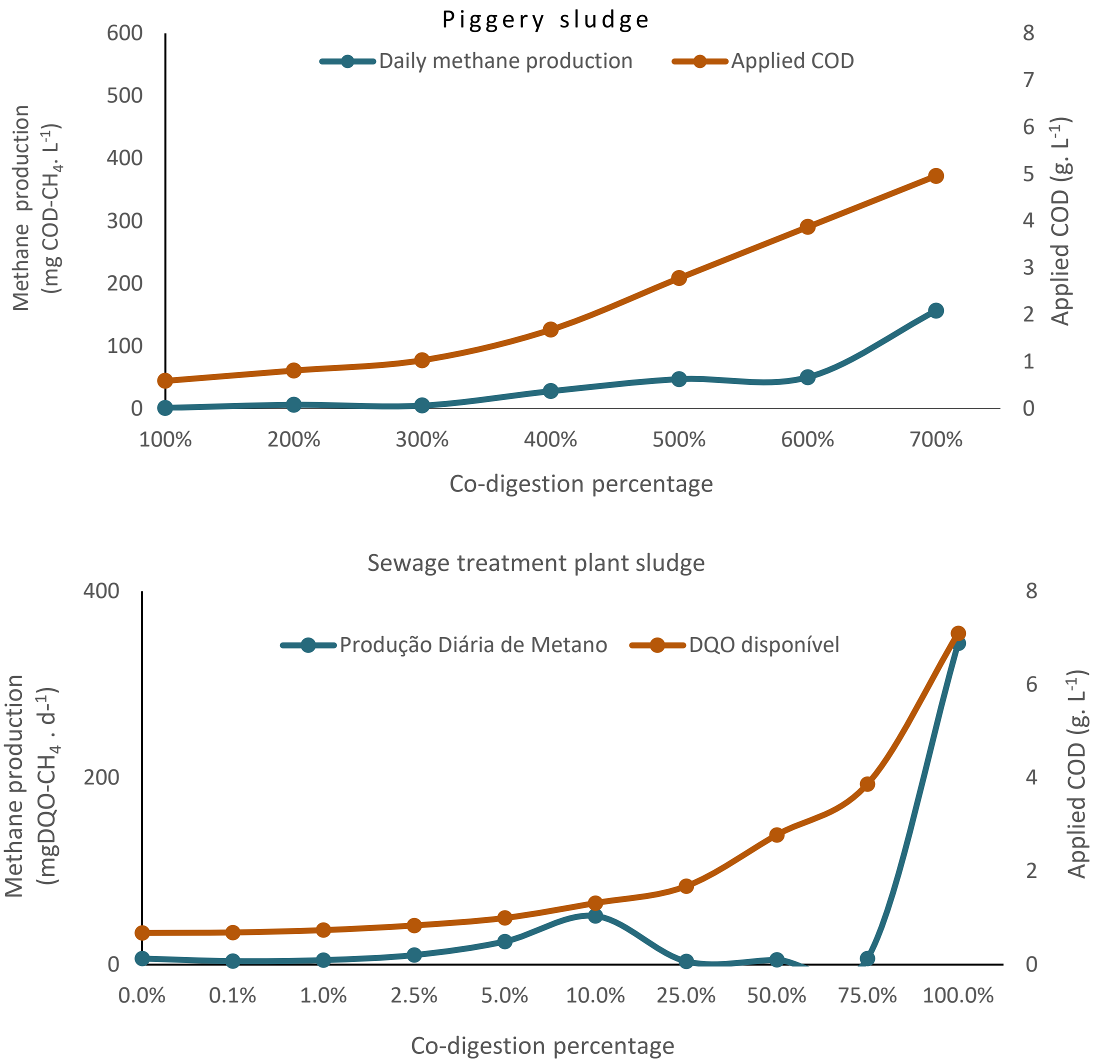

Figure 3. Daily methane production and applied COD relation in each condition according with the volumetric concentration percentage (v/v) of leachate and domestic sewage (100% of leachate).

Berenjkar *et al.* (2019), in analyzing the biogas production potential in co-digestion tests with dilutions of up to 40% leachate by using sludge from a STP, obtained better results with the addition of up to 10% leachate, not recommending the application of tests above 20%. For De Albuquerque *et al.* (2018), the combined treatment between leachate and domestic sewage becomes attractive for mixtures of up to 2% leachate. They compare analyses of microbial communities and of organic load removal efficiency precisely because of the characteristics of leachate, which hinders treatability and biological adaptability when in higher dilutions.

One of the main factors for the drop in methane production when leachate is applied in proportions higher than 10% is related to the potential toxicity of the leachate, which can have high levels of recalcitrant compounds (Kewu and Wenqi, 2008). This factor makes anaerobic processes more difficult, when preliminary treatments for leachates become necessary (Neczaj *et al.*, 2007). Another factor that makes the co-digestion of leachate with domestic sewage limiting is the high concentration of ammonia present in the leachate, where concentrations higher than 400 mgN-NH_4+.L^{-1} can be inhibitory or even toxic for methanogenic organisms (Bakonyi *et al.*, 2019). In verifying the concentration of N-NH^{4+} available in leachate, we can rule out this hypothesis, given that its value does not surpass 100 mg.L^{-1}. Besides the low concentration of ammonia, the C/N ratio (based on total COD) is high, which provides better substrate biodegradation (Çeçen and Çakiroğlu, 2001).

An aspect that can justify the better performance of piggery sludge in relation to results with STP sludge is the operational regime. The STPs possess greater operational control, with continuous flow and low organic load, which reduces the methane generation potential (De Lucena *et al.*, 2011; Hussain and Dubey, 2017; Rizvi *et al.*, 2015). Animal waste treatment systems, like the place where piggery sludge is collected, are operated with low control as regards feeding regimes, which can occur daily or seasonally. Moreover, they work with high organic and ammoniac nitrogen loads, which makes the biomass originating from these systems more adaptable to other substrates (Chen *et al.*, 2012).

That being so, for a higher biogas recovery, alternatives such as the complimentary use of sludges with better capacity of biodegradation or activation of the biomass already existing in the rectors become attractive. In addition, standardizing appropriate operational regimes is necessary so that the limiting of mass transfer and the microbial dynamics favor technologies in biogas utilization, since in laboratory they design an anaerobic digestion structure completely different from real full-scale conditions (Gulhane *et al.*, 2017; Zhen *et al.*, 2015).

Conclusion

In general, the methanogenic activity of the full-scale reactors was low using domestic sewage as substrate, with an average value of 3.26 mg COD-CH4.gTVS.d^{-1}. When using the synthetic substrates the reactor sludge showed a slight improvement in SMA, with average values of 5.43 and 8.54 mg COD-CH4.gTVS.d-^{1} for acetate and glucose synthetic solution, respectively. This is probably due to the full-scale hydrodynamics and the operating conditions. Such as, receiving sludge from a Water Treatment Plants, sludge from septic tanks, variation in flow throughout the day and year, receiving leachate at certain times of the year, inadequate handling of sludge and scum, among other examples.

Besides the methanogenic activity, the rate improvement methane production capacity was also evidenced with the inclusion of landfill leachate in co-digestion with domestic sewage, as well as the use of a complementary sludge, such as piggery sludge (agro-industry). Even with higher methane production potential, co-digestion tests point out that there is a better condition for STP sludge when with a concentration of up to 10% of leachate with domestic sewage. For piggery sludge, positive response was found for conditions up to 75%. However, when the applied COD and converted COD relation is evaluated, the viability was up to 50% of leachate inclusion in relation to domestic sewage.

The acclimatization of sludge or its thickening using sludges of other origins, especially from the treatment of agro-industrial effluents, presents itself as a viable alternative to optimize methane production in co-digestion, so as to provide treatment with higher concentrations of leachate and better conversion rates.

This strategic alternative can be adopted in countries and regions that have a thriving agro-industrial sector and even rely on smart solutions to control pollution, such as Latin America, the Caribbean, Africa and Asia. The potential of the relationship between water (sewage) and energy is evident, connecting urban and rural areas. It is recommended that experiments be carried out with different types of sludge from the agroindustry and with different types of leachates, in addition to studies focused on hydrodynamics and taking into account more detailed operational aspects.

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