Recirculation of solid digestate to enhance energy efficiency of biogas plants: Strategies, conditions and impacts

Ulysse Bremond, Aude Bertrandias, Raphaëlle de Buyer, Eric Latrille, Julie Jimenez, Renaud Escudié, Jean-Philippe Steyer, Nicolas Bernet, Hélène Carrère

Recirculation of solid digestate to enhance energy efficiency of biogas plants: strategies, conditions and impacts

Ulysse Brémond[a,b], Aude Bertrandias[a], Raphaëlle de Buyer[a], Eric Latrille[b], Julie Jimenez[b], Renaud Escudié[b], Jean-Philippe Steyer[b], Nicolas Bernet[b], Hélène Carrere[b,*]

[a] Air Liquide, Innovation Campus Paris, 1 Chemin de la Porte des Loges, 78354 Jouy-en-Josas, France

[b] INRAE, Univ Montpellier, LBE, 102 Avenue des étangs, 11100 Narbonne, France

*Corresponding author.

E-mail address: helene.carrere@inrae.fr (H. Carrere).

Keywords

Anaerobic digestion; Biogas; Solid digestate recirculation; Post-treatments; Solid retention time; Digester total solid content

Abstract

This article aims at providing insight into the recirculation of (post-treated) solid digestate (SD) within digesters. Such a practice has been further identified as promising for anaerobically digesting recalcitrant feedstocks and for improving the energy efficiency of continuously stirred tank reactor (CSTR) agricultural biogas plants. Firstly, implementation strategies and the potential impact of SD direct recirculation on five CSTR biogas plants were investigated. According to the selected strategy, results indicated that plant methane production could rise by 0.6 to 6.3% or that a potential feedstock shortage of 64 to 1431 tons/year could be compensated. Secondly, the relevance of additional post-treatments for improving these initial results was evaluated. Thermo-chemical post-treatments successfully increased SD biodegradability by 30 to 46% although their costs were not compensated by additional methane production. Short-term aerobic post-treatments failed in increasing SD biodegradability (up to 21% loss in biomethane potential). Hence, at full scale, a quick and direct recirculation of SD excluding any post-treatment appears to be the optimal condition to apply. Finally, conditions for the full-scale implementation of direct SD recirculation were theoretically studied. This practice has proved to increase the solid retention time by 11 to 38% and the plant total solid content by 6 to 20%. Thus, the critical point for its implementation should be the capacity of the plant mixing system to handle such an increase in solids. The relevance of SD recirculation needs to be determined on a case-by-case basis. Consequently, for the first time, this article provides a framework where the conditions can be identified for direct SD recirculation to be a relevant digestate management practice. Overall, this article demonstrates how direct SD recirculation can be a simple and low-cost mean for improving agricultural CSTR biogas plant efficiency. It also highlights the importance of achieving efficient digestate management in the biogas sector in order to reduce the costs of biogas production.

1. Introduction

One of the main challenges of the 21st century is to decorrelate growing worldwide energy consumption from the use of fossil fuels. Their substitution with low-carbon/renewable options is crucial for reducing greenhouse gas (GHG) emissions and mitigating global warming. Among these renewable options, anaerobic digestion (AD) displays several advantages since it processes low-cost organic waste via a robust and naturally occurring biological process to produce biogas. It is a versatile energy carrier that can be used for generating power and heat, injected into the gas grid or used as a fuel for vehicles [1]. In Europe, incentive policies have led to the emergence of a biogas industry, mainly based on agricultural feedstocks (agricultural residues, energy crops, catch crops) which represent the largest available deposit of organic matter. Biomethane potential from agricultural feedstocks was estimated at 58.9 billion Nm^3, three times higher than potential from other organic wastes, such as food waste or sewage sludge [2].

In addition to biogas, biogas plants also produce digestate throughout the year. In continuous stirred-tank reactors (CSTR), the main AD technology at full scale, digestate is mostly composed of a mixture of water, residual undigested feedstocks, microorganisms and inorganic matter (minerals…) thus representing a valuable biofertilizer [3]. However, land disposal of digestate in Europe is regulated by the Nitrate directive that aims to reduce ammonia and nitrogen oxide emissions as well as nitrogen brought to the soil [4]. Digestate storage would therefore allow for adequate land spreading in due time, once the digestate is stabilized and the crops have reached a growth stage requiring nutritional input. Digestate storage would also allow for the regulation of quantities, as its application can vary according to the type of soil and to the farm area [5]. To ease storage and handling issues within the farm or its surroundings, digestate is generally separated mechanically into a liquid and solid fraction, using a screw press. This type of phase separator produces a higher total amount of liquid digestate (LD) than solid digestate (SD) (ratio 90/10 in volume) [6]. However, SD presents a higher concentration in organic matter (mainly fibres) while LD is richer in nitrogen and potassium [7].

For SD, the most widespread storage method implies composting before use as a soil amendment. However, according to certain studies: (i) SD can have a biomethane potential as high as 240 Nm^3 CH_4·ton^{-1} volatile solids (VS) [8]; (ii) strong ammonia emissions occur during the composting treatment of agricultural SD since significant amounts of NH_3/NH_4^+ contained in SD are volatilized, while carbon is emitted as CO_2 [9]. To avoid this loss of organic matter during composting, three main strategies have been identified to recover more energy from SD [10]: (i) fermentation of SD to produce bioethanol [11]; (ii) thermal conversion processes of SD such as pyrolysis that produce syngas, biochar and bio-oil [12], hydrothermal and vapothermal carbonisation that produce mainly hydrochar [13] or combustion that provides heat [14]; (iii) recirculation of SD within the biogas plant to recover the residual methane potential [8].

The latter strategy is of particular interest as it may yield more energy than bioethanol, increases biogas plant efficiency, since feedstocks are further degraded and does not need additional investment for pyrolysis or hydrothermal reactors. Besides, it may reduce GHG emissions and air pollution, since less SD would be composted and CO_2 as well as ammonia emissions would be reduced. As recirculation of SD is relatively easy to implement at full-scale, it has already been applied in several agricultural biogas plants. For example, Menardo et al. described biogas plants where recirculated SD corresponded to 4-6% of the mass of total feedstock mix [15]. However, the conditions in which SD recirculation is implemented as well as the resulting impact on methane production and biogas plant operation are, to our knowledge, poorly understood and have never been extensively studied in literature.

Furthermore, several lab scale studies have tested the effect of post-treatment strategies of SD before recirculation on the enhancement of its biomethane potential (BMP). A 13 to

176% increase in SD BMP has been observed for thermal treatment [15], milling [16] and enzymatic treatment [17]. However, in all of these studies, the economic impact of recirculation for agricultural biogas plants and possible post-treatment costs were rarely discussed. Sambusiti *et al.* estimated that direct recirculation of SD could lead to a 14% increase in economic profits, although this was based on very optimistic assumptions: a high solid separation efficiency of the screw press (73%) and a recirculation of the total amount of SD produced [17]. Solid separation efficiency indexes of a screw press vary around 30% for total solids (TS) [6]. Finally, only a partial amount of SD can be recirculated as some solids should exit the system, notably to eliminate the inorganic matter that would otherwise accumulate and cause inhibitions (e.g. heavy metals). Realistic hypotheses need to be applied in order to fully evaluate the economic relevance of such post-treatment practices in a recirculation scheme.

Currently, one of the main challenges in the biogas sector is to reduce the cost of biogas production [18]. To increase knowledge in practices that may improve biogas plant efficiency and therefore the biogas sector economy is a key issue. Since SD recirculation represents one of these practices and according to the problematics highlighted in the two previous paragraphs, the novelty and objective of this paper aim for the first time at providing answers to the following questions:

(i) Which strategies and conditions are relevant for the application of SD recirculation?

(ii) Are additional post-treatments prior to SD recirculation economically viable?

(iii) What are the impacts of SD recirculation on plant methane production, solid retention time and digester TS content?

To fill this knowledge gap, the present study proceeds in several steps. Firstly, three implementation strategies will be defined. Secondly, for five CSTR biogas plants, the impact of direct SD recirculation on the plant methane yield and the significance of performing post-treatment on SD (thermo-chemical and biological methods tested) will be evaluated. Thirdly, the impact of SD recirculation on solid retention time and digester TS content in these biogas plants will be assessed and discussed. Finally, on a case-by-case basis, an innovative procedure is provided to determine the significance of recirculation for a given biogas plant.

2. Materials and methods

2.1. The different strategies for SD recirculation

It is important to define the three possible implementation strategies for SD recirculation. These are represented in **Figure 1**:

(i) SD can be recirculated in addition to the existing ration; additional biogas can thus be produced. Such a strategy will be further called the "addition strategy".

(ii) SD can also be integrated to the ration and replace certain feedstocks that are lacking (shortage/voluntarily non-produced/further stored, in the case of a future anticipated shortage). In this second case, SD recirculation would allow to totally offset ration reduction and maintain a constant biogas production over time. As SD generally generates less methane than feedstocks, this strategy would imply that more SD than replaced feedstocks can be recirculated. Such a strategy will be further called the "replacement strategy".

(iii) Finally, SD can also be integrated to the ration and replace certain feedstocks that are lacking although the amount of recirculated SD would not allow for a total offset of ration reduction or maintain a steady biogas production over time. In this case, it would simply limit loss in biogas production. This could be either due to a too high quantity of lacking feedstock that cannot be offset even when all yearly produced SD is recirculated, either due to a quantity of recirculated SD that is lower than the quantity of SD recirculated to reach the replacement strategy ($\dot{m}_{SD\_replacement}$ in tons per year). In this latter case the quantity of recirculated SD ($\dot{m}_{SD\_partial\_replacement}$ in tons per year) would vary within the following range:

$$0 < \dot{m}_{\mathrm{SD\_partial\_replacement}} < \dot{m}_{\mathrm{SD\_replacement}}$$

This strategy will be further called the "partial replacement strategy"

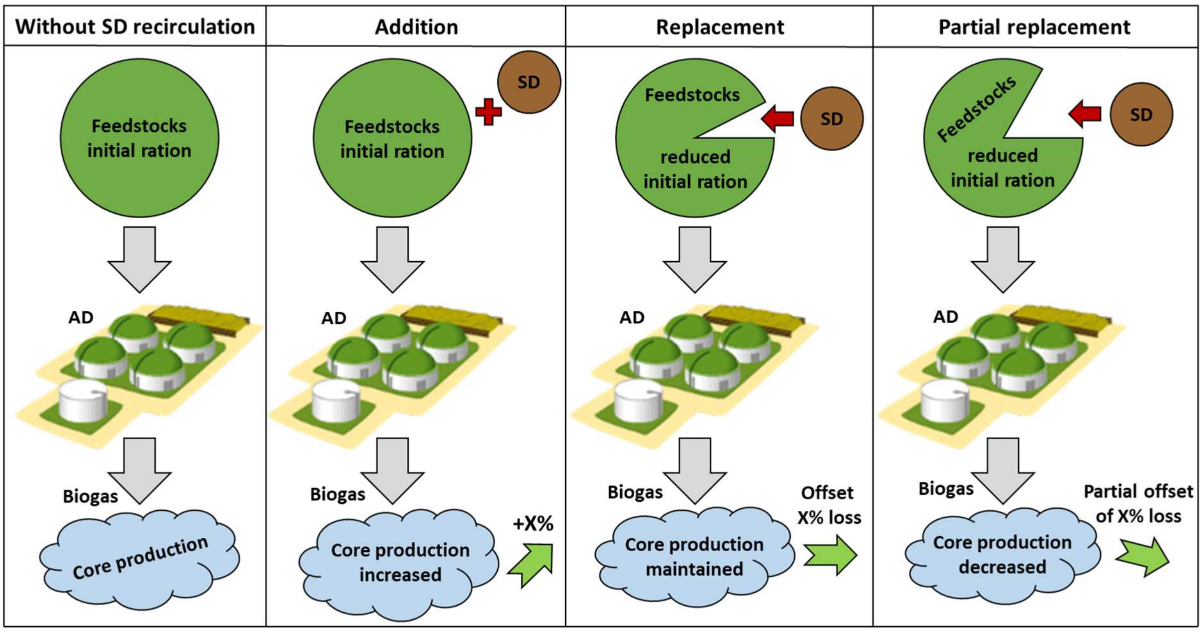


Figure 1: Overview of the three strategies possible for SD recirculation

In the present paper focus is mainly put on the addition and replacement strategies for cases where biogas production increases or, at least, does not decrease.

2.2. Solid digestate sampling and biogas plant features

SD samples were collected from five French full-scale agricultural or territorial CSTR biogas plants. Sampling was performed identically at all plants according to the following procedure: To ensure sample homogeneity, SD was collected from different parts of the fresh SD pile, which was located below the outlet of the phase separator. According to the biogas plant set-up, the phase separator released SD either from the post-digester or from the digester (if there was no existing post-digester). All SD samples were frozen in gas-tight containers in order to preserve their methane potential before subsequent experimental handling.

In addition, each plant operator was interviewed in order to obtain details about the biogas plant features. Information concerning the type and reactor volume (RV in m^3), the type of phase separator, the plant feedstock mix, the percentage of TS in the digester, the quantity of feedstocks per year ($\dot{m}_{feedstock}$ in kg or t/year), the HRT before phase separation, the pH, the temperature, the average percentage of methane in biogas (x_{CH4} in %), the quantity of SD produced yearly ($\dot{m}_{SD}$ in kg or t/year), the type of valorisation and the associated flowrate of biomethane injected (Q_{bioCH4_hour} in Nm^3 biomethane/hour) or combined heat and power valorisation (CHP) engine nominal power (CHP_{power} in kW) were provided by the plant operators. In all calculations, the number of days of operation during the year ($D_{operation}$ in days) was realistically set to 342 days, considering technical maintenance and possible operational contingencies. From these data, a number of additional features were calculated.

The loading rate (LR in kg feedstock/m^3/day) was obtained using the quantity of feedstocks used per year ($\dot{m}_{feedstock}$ in kg/year) and the digester reactor volume ($RV_{Digester}$ in m^3) according to the following equation:

$$LR = \frac{\dot{m}_{feedstock}}{RV_{Digester} * D_{operation}} \quad (1)$$

In absence of biogas flowmeters on plants, calculation of the plant biogas production (V_{biogas_year} in Nm^3/year) was different as a function of the valorisation type. In the case where upgrading and biomethane injection occurred, all produced methane was assumed to be injected and the following equation was used.

$$V_{biogas\_year} = \frac{Q_{bioCH4\_hour} * D_{operation} * 24}{x_{CH4}} \quad (2)$$

In the case of CHP, several hypotheses were made: (i) The CHP engines were sized to have an optimal efficiency, therefore only 85% of the CHP engine nominal power was used; (ii) The electric efficiency of the CHP system was 40% as usually reported in the literature [17]; (iii) Five percent of additional energetic loss to avoid under-supply of the CHP system [19]; (iv) A lower heating value (LHV) of 9.94 kWh/Nm^3 was used for methane; (v) all biogas produced was sent to the CHP system. Total electrical energy produced per year (E_{elec} in kWh$_{el}$/year) and plant biogas production (V_{biogas_year} in Nm^3/year) were calculated using these two equations:

$$E_{elec} = CHP_{power} * 0.85 * D_{operation} * 24 * 0.4 \quad (3)$$

$$V_{biogas\_year} = \frac{CHP_{power} * 0.85 * D_{operation} * 24}{0.95 * LHV * x_{CH4}} \quad (4)$$

It is noteworthy that calculated values are close to the approximate figures provided by plant operators. Finally, the average methane yield from feedstock (Y_{CH4} in Nm^3 CH_4/t feedstock) was calculated from the volume of methane produced by the plant each year (V_{CH4_year} in Nm^3 biomethane/year by multiplying V_{biogas_year} with x_{CH4}):

$$Y_{CH4} = \frac{V_{CH4\_year}}{\dot{m}_{feedstock}} \quad (5)$$

For each biogas plant, given and calculated features in this section, are displayed in **Table 1**. For reasons of confidentiality, a letter was attributed to each plant.

Table 1: Features of the five biogas plants contributing to this study

Plant Name	Plant A	Plant B	Plant C	Plant D	Plant E
Process	CSTR	CSTR	CSTR	CSTR	CSTR
Type of plant	Agricultural	Agricultural	Agricultural	Territorial	Agricultural
Reactors volume (m^3)	Digester – 2,000 Post-dig. – 2,000 Storage tank – 6,000	Digester – 2,200 Post-dig. – 2,200 Storage tank – 5,500	Digester – 2,000 Post-dig. – 2,000 Storage tank – 4,000	Digester – 3,700 Post-dig. – 2,700 Lagoon – 10,000	Digester – 1,200 Post-dig. – 1,200 Lagoon – 16,000
Phase separator	Screw press	Screw press	Screw press	Screw press	Screw press
Feedstocks intake (% of total mass)	Catch crop (30%) – Bovine manure (18%) – Beet pulp (11%) – Cereal dust (8%) – Whey (33%)	Bovine manure (60%) – Energy crop (10%) – Grass silage (10%) – Cereal straw (10%) – Cereal dust (10%)	Catch crop (50%) – Beet pulp (40%) – Cereal dust (10%)	Waste from food industry (43%) – Animal manure (30%) – Cereal residues (12%) – Sludge (12%) – Catch crop (3%)	Manure (65%) – Beet pulp (15%) – Energy crop (10%) – Cereal residues (5%) – Waste from food industry (5%)
%Total solids in digester (%)	12	10.5	10.5	14	7.6
Total quantity of feedstock (t/year)	13,600	18,000	10,500	25,000	20,000
Loading rate (kg feedstock/m^3/day)	19.9	23.9	15.4	19.8	48.73
HRT before phase separation (days)	100	84	130	88	40
pH	7.6-7.9	7.6-7.9	7.6-7.8	7.6-7.8	7.6-7.8
Temperature regime	Mesophilic	Thermophilic	Mesophilic	Mesophilic	Mesophilic
Quantity of produced SD (t/year)	1,800	2,000	1,200	3,300	2,500
Biogas valorization	Upgrading	CHP	Upgrading	Upgrading	CHP
Biomethane injected (Nm^3/hour)	145	/	140	160	/
CHP engine nominal power (kW)	/	1890	/	/	1090
Plant biogas production (Nm^3/year)	2,123,000	2,632,000	2,126,000	2,340,000	1,411,000
Biogas composition (% of methane)	56	53	54	56	57
Plant methane production (Nm^3/year)	1,189,000	1,395,000	1,148,000	1,312,000	805,000
Methane yield (Nm^3 CH_4/t feedstock)	87.4	77.5	109.3	52.5	40.25
Feed-in tariff biomethane (€/MWh HHV)	115.95	/	117.07	102.77	/
Feed-in tariff electricity (€/MWh)	/	144.29	/	/	202.21

2.3. Solid digestate characterisation

Several methods were used to characterise the different SD. The TS and VS contents were obtained following the standard methods of the American Public Health Association [20]. Total carbon (TC) content was measured using a Shimadzu TOC-VCSN Analyser coupled to a Shimadzu solid sample module SSM-5000A. An AutoKjehdahl Unit K-370, BUCHI was used to determine the total Kjeldahl nitrogen. Aqualytic 420721 COD Vario Tube Test MR (0-1.5 $g{\cdot}L^{-1}$) were used to get the chemical oxygen demand (COD). For liquids, 2 ml of sample adequately diluted were pipetted into each tube and oxidation reactions in tubes were performed in a HACH COD reactor at 150 C for 2 hours. Finally, COD concentrations were measured using an Aqualytic MultiDirect spectrophotometer. For solids, in duplicate, 0.25 g of samples were poured into 10 ml of 98% H_2SO_4 and set under strong agitation overnight to solubilize solid particles. Dilution with Milli-Q water up to 250 ml allowed pipetting. The obtained liquid was used as previously in COD tubes.

SD matter was characterised via sequential chemical extractions according to the protocol developed by Jimenez et al. [21]. Samples were freeze-dried, then grounded at 1 mm using a Fritsch Pulverisette 19 and associated stainless steel sieve. Subsequent sequential extractions were performed using 0.5 g of samples. Three fractions of decreasing accessibility were then obtained by applying increasingly strong chemical solutions: (i) soluble OM (SPOM) using $CaCl_2$ (10 mM) that corresponds to soluble sugars and proteins; (ii) slowly extractable OM (SEOM) using NaOH (0.1 M), which corresponds to remaining proteins and sugars, some humic substances and lipids; (iii) poorly extractable OM (PEOM) using H_2SO_4 72% w/w that corresponds to hemicellulose and cellulose. At each step, the solubilized VS was recovered in the supernatant by centrifugation (18,750 g for 20 min at 4 °C) and filtered at 0.45 µm. VS of each fraction were then characterized via COD measurement as described before. Finally, the non-extractable OM (NEOM) was calculated by subtracting the four fractions of VS extracted from the sample from the total VS. Total VS were obtained by measuring COD on SD solid sample powder as previously described. As VS correspond to organic matter, the former term will only be used thereafter.

BMP tests were performed on each SD. SD were anaerobically digested in batch mode at 35°C under agitation. Bottles with a working volume of 400 mL, were used. Bottles were filled with: 4mL of trace elements solution ($FeCl_2$, 2 $g \cdot L^{-1}$; $CoCl_2$, 0.5 $g \cdot L^{-1}$; $MnCl_2$, 0.1 $g \cdot L^{-1}$; $NiCl_2$, 0.1 $g \cdot L^{-1}$; $ZnCl_2$, 0.05 $g \cdot L^{-1}$; H_3BO_3, 0.05 $g \cdot L^{-1}$; Na_2SeO_3, 0.05 $g \cdot L^{-1}$; $CuCl_2$, 0.04 $g \cdot L^{-1}$; Na_2MoO_4, 0.01 $g \cdot L^{-1}$), 8.6 ml of macroelement solution (NH_4Cl, 26.6 $g \cdot L^{-1}$; KH_2PO_4, 10 $g \cdot L^{-1}$; $MgCl_2$, 6 $g \cdot L^{-1}$; $CaCl_2$, 3 $g \cdot L^{-1}$), 20.8 mL of bicarbonate buffer ($NaHCO_3$, 50 $g \cdot L^{-1}$), an inoculum at 5 g of $VS \cdot L^{-1}$, the SD at 5 g of $VS \cdot L^{-1}$ and potentially with distilled water to complete to 400 mL. Anaerobic conditions were then obtained by flushing the headspace with nitrogen gas and closing bottles with red butyl rubber septum-type stoppers. A control was made by preparing the same mixture with the inoculum but without the substrate to evaluate the endogenous methane production. The inoculum was the same for all tests; a granular sludge coming from a mesophilic anaerobic digester of a sugar refinery was mixed with water and crushed using a kitchen immersion hand blender. Then, a 25-mesh sieve was used to remove most of the remaining solid/minerals and to obtain a homogenous liquid inoculum. According to standardized practices, triplicates were launched and incubated under agitation at 35°C. Biogas production was frequently monitored by measurement of the headspace pressure and its composition using a PerkinElmer® Clarus 580 gas chromatograph as described in a previous study [22]. Endogenous methane production was evaluated by subtracting the gas produced in the controls and BMP tests were stopped when the daily methane production during three consecutive days was <1% of the accumulated volume of methane [23]. All volumes are expressed in normal conditions.

Besides, anaerobic biodegradability (BD_{ana}), expressed in percentage of total COD, was calculated as the ratio between the BMP and COD_{tot} of the corresponding SD, following the formula [24]:

$$BD_{ana} = \frac{BMP * 100}{0.35 * COD_{tot}} \quad (6)$$

After SD characterisation, decision was made to further work with SD from plants A, B and C as they were representative of agricultural CSTR plant SD.

2.4. Thermo-alkaline post-treatment

SD from plants A, B and C were subjected to a thermo-alkaline post-treatment. Sodium hydroxide (NaOH) was added to SD at a dose of 2% w/w, then alkalized SD was placed in a New Brunswick Scientific™ incubator shaker Innova® 43 at 55°C and 120 revolutions per minute (rpm) for 3 days. BMP tests were then launched according to the previously described method (see 3.2.). Tests involved the post-treated SD, the corresponding untreated SD and the control, without SD, for endogenous production.

2.5. Short-term aerobic post-treatment

Short-term aerobic post-treatment was applied to SD from plants A and B. 300 grams of SD were placed inside 2.5 L bioreactors and aerated according to the set-up displayed in **Appendix A**. SD was contained in a fine polyester net to avoid loss of material. A calibrated peristaltic pump sent air into the system. Airflow first entered a concentrated NaOH (2M) solution to trap atmospheric CO_2. The air was then moisturized by bubbling into water at the bottom of the double jacket 2.5L bioreactor. For all experiments, temperature in the aerobic reactors was set at 30°C. Finally, carbon loss due to respiration was evaluated by trapping the emitted CO_2 in a 0.5 M soda solution set at 30°C, of which conductivity was measured and recorded every five minutes using an internally developed acquisition system. The relationship between conductivity ($mS{\cdot}cm^{-1}$) and trapped carbon ($mg\ C\text{-}CO_2{\cdot}L^{-1}$) was determined according to a calibration line obtained by measuring conductivity of the soda solution with different quantities of Na_2CO_3 dissolved inside and 0.273 as a correction factor to translate the $mg\ CO_2{\cdot}L^{-1}$ trapped into $mg\ C\text{-}CO_2{\cdot}L^{-1}$. The total amount of carbon in the SD placed in the bioreactor was calculated from the mass of SD, its TS and the average TC content. From this set-up, several short durations and low to strong airflow were tested at 30°C. For plant A, 1.75, 3.6 and 6 days with an airflow of 1.5, 10 and 20 $L{\cdot}h^{-1}{\cdot}kg^{-1}$ of TS of SD were tested. For plant B, 2.5 and 5 days with an airflow of 1.65, 16.5 and 33 $L{\cdot}h^{-1}{\cdot}kg^{-1}$ of TS of SD were applied.

Samples from aerated SD were also used as substrates for additional BMP tests. These tests involved aerated SD, the corresponding untreated SD (blank) and the control without SD for endogenous production. For aerated SD, BMP of final post-treated matter (BMP_{FPTM} in $mL\ CH_4{\cdot}g^{-1}$ VS final) was defined as the methane directly measured and produced by the remaining matter without any respiration correction. It can be assimilated to biodegradability of post-treated SD. To determine their BMP, carbon loss was taken into account. It was obtained from raw BMP values corrected according to the hypothesis that all carbon losses as CO_2 during the post-treatment would otherwise have produced biogas (ratio 60% CH_4/40% CO_2).

2.6. Impact of SD recirculation on plant methane production

The impact of SD recirculation on plant methane production (gain or offset) is evaluated as a percentage of the total plant methane production. This percentage ($\%_{total\_plant\_CH4}$) is obtained by dividing the volume of methane produced from recirculation (V_{CH4_SD}) by the plant methane production (V_{CH4_year}). V_{CH4_SD} is calculated using the measured BMP from SD, the quantity of SD recirculated each year ($\dot{m}_{SD\_recirculated}(R)$) and the %VS of SD ($[VS]_{SD}$). For BMP values, in the case of SD from plants A, B and C, the considered value was the average of values obtained during the three different BMP tests (control values in the case of each post-treatment). As an example, in the case of plant A, BMP for each control were 130, 155 and 153 mL $CH_4 \cdot g^{-1}$ VS, respectively. The considered value was therefore their average: 146 ml $CH_4 \cdot g^{-1}$ VS. For SD from plants C, D and E, the BMP values were only obtained from one set of trials (control for simple BMP; no post-treatment tested on these SD). Finally, a 0.8 correction factor was applied to all BMP values from plants A, B, C, D, E and F, in order to simulate a recirculation process at full scale rather than lab scale and to assess realistic values [23]. These new values are called corrected SD BMP ($BMP_{corrected\_SD}$). Besides, it is important to underline that all produced SD cannot be recirculated. Indeed, certain types of recalcitrant organic matter as well as minerals must exit the system to avoid AD inhibitions and be spread on land to recycle carbon and nutrients. Calculations were performed on three amounts of SD that can be recirculated within the biogas plant. They correspond to 30%, 50% and 80% of the total amount of SD produced each year and appear to be realistic in the case of a full-scale application. Therefore, $\dot{m}_{SD\_recirculated}$ varies according to these percentages. All these values were used to calculate the impact of SD recirculation on plant methane production:

$$\%_{total\_plant\_CH4} = \frac{V_{CH4\_SD}}{V_{CH4\_year}} = \frac{BMP_{corrected\_SD} * \dot{m}_{SD\_recirculated} * [VS]_{SD}}{V_{CH4\_year}} \quad (7)$$

In the case of an addition strategy, this percentage of the total plant methane production should represent an additional amount of methane produced. This additional methane is assumed to be bought for the same purchase price or feed-in tariff (FIT) as contractual production, therefore leading to additional economic profits (AEG).

As all plants are in France, FIT for injected biomethane and electricity, set by the French government, were obtained by using the calculator provided by the Association Technique Energie Environnement (ATEE) Club biogaz [25]. Tariffs are given in **Table 1**. For all plants, the purchase agreement of biomethane or electricity was assumed to be signed on the 1st January 2019. For biomethane injection, a higher heating value (HHV) of biomethane was set at 10.8 kWh/Nm^3 and bonuses for the presence of catch crops, sludge or municipal waste inside the feedstock mix were taken into account to obtain the feed-in tariff. In the case of electricity, similarly bonuses due to the use of livestock manure were considered.

In French biogas legislation, as a standard rule, methane energy equivalence is calculated with the HHV coefficient in the case of biomethane injection, while the lower heating value coefficient is used for CHP valorisation. According to this rule, additional economic profits were obtained using two formulas according to the type of valorisation. For upgrading purposes, the HHV value (kWh/Nm^3), the additional amount of produced methane ($V_{additional\_CH4}$ in Nm^3 CH_4/year) and the corresponding feed-in tariff for the biogas plant ($FIT_{biomethane}$ in €/MWh) were used as displayed in **Eq. 8**. For CHP, the LHV (9.94 kWh/Nm^3), the electrical conversion efficiency set to 40% and 5% of additional energetic loss to avoid the under-supply of the CHP system and the corresponding feed-in tariff for the biogas plant ($FIT_{electrical}$ in €/MWh) were used as displayed in **Eq. 9**.

$$AEG_{biomethane} = \frac{V_{CH4\_SD} * HHV * FIT_{\text{biomethane}}}{1000} \quad (8)$$

$$AEG_{electrical} = \frac{V_{CH4\_SD} * LHV * 0.95 * 0.4 * FIT_{electrical}}{1000} \quad (9)$$

In the case of a recirculation replacement strategy, this percentage of the total plant methane production shoud allow to calculate the tons of feedstocks ($\Delta_{\text{feedstock}}$) that could be replaced by the amount of recirculated SD ($\dot{m}_{SD\_recirculated}$) per year. This amount of feedstock can be calculated using the methane yield (Y_{CH4} – obtained via **Eq. 5**) and according to the following equation:

$$\Delta_{\text{feedstock}} = \frac{V_{CH4\_SD}}{Y_{CH4}} \Leftrightarrow \frac{[VS]_{SD} * BMP_{SD} * \dot{m}_{SD} * R}{Y_{CH4}} \quad (10)$$

2.7. Impact of SD recirculation on CSTR solid retention time and TS content

For agricultural CSTR biogas plants, hydraulic retention time (HRT in days) represents the retention time of liquids in the system and can be calculated in two different manners. The most common and simplest way uses digester and post-digester volumes ($RV_{digester}$ and $RV_{post\text{-}digester}$ in m^3) as well as the feedstock intake ($\dot{m}_{feedstock}$ in t/year divided by the days of operation during the year $D_{Operation}$ in days/year) to calculate HRT ($HRT_{Classical}$ in days) according to the **Eq. 11**. It is generally assumed that feedstock densities are equal to one, therefore tons can be converted to cubic meters.

$$HRT_{Classical} = \frac{RV_{digester} + RV_{post-digester}}{\frac{\dot{m}_{feedstock}}{D_{operation}}} \quad (11)$$

HRT displayed in **Table 1** are calculated in this way and are similar to values provided by operators. However, with this approach, calculations do not take into account the gas production from the feedstock, thus leading to inaccurate results. However, in these biogas plants, the amount of biogas produced (V_{biogas_year} in Nm^3/year) as well as its average composition is known (x_{CH4} defined as the methane percentage in the biogas). The mass of biogas produced ($\dot{m}_{biogas}$ in t/year) can therefore be approximated using average methane and carbon dioxide densities (d_{CH4} - 0.67 and d_{CO2} - 1.87 kg/Nm^3 at 15°C and atmospheric pressure), according to the following equation:

$$\dot{m}_{biogas} = \frac{V_{biogas\_year} * x_{CH4} * d_{CH4} + V_{biogas} * (1 - x_{CH4}) * d_{CO2}}{1000} \quad (12)$$

This new value can be used to perform a more precise HRT calculation (**Eq. 13**), which takes into account the biogas production and realistically reflects the time spent in the digester of the outgoing undigested matter. For this reason, $HRT_{accurate}$ will be used instead of $HRT_{Classical}$ in the rest of this paper.

$$HRT_{accurate} = \frac{RV_{digester} + RV_{post-digester}}{\frac{\dot{m}_{feedstock} - \dot{m}_{biogas}}{D_{operation}}} \quad (13)$$

Without any SD recirculation, $HRT_{accurate}$ corresponds to the solid retention time (SRT), also expressed in days. However, when SD recirculation is performed, HRT and SRT can increase although the extent depends on the selected strategy (addition or replacement).

2.7.1. Direct SD recirculation - Addition strategy

In the case of an addition strategy, HRT should slightly increase due to biogas production from recirculated SD. The new mass of biogas produced ($\dot{m}_{biogas}(R)$ in t/year) can be calculated as a function of the percentage of total SD produced that is recirculated (R), according to **Eq. 14.** This equation is based on **Eq. 12**, the amount of SD recirculated ($\dot{m}_{SD\_recirculated}$ that depends on R, in t/year), the VS content of SD ($[VS]_{SD}$) as well as the SD methane potential (BMP_{SD} in Nm^3 CH_4/t VS). Therefore, $HRT_{accurate}(R)$ can be recalculated using **Eq. 15**.

$$\dot{m}_{biogas}(R) = \dot{m}_{biogas}(0) + \frac{\dot{m}_{SD\_recirculated}(R) * [VS]_{SD} * BMP_{SD} * (d_{CH4} + \frac{(1 - x_{CH4})}{x_{CH4}} * d_{CO2})}{1000}$$

(14)

$$HRT_{accurate}(R) = \frac{RV_{digester} + RV_{post-digester}}{\frac{\dot{m}_{feedstock} - \dot{m}_{biogas}(R)}{D_{operation}}} \quad (15)$$

Eq. 16 was obtained according to the calculations and hypotheses developed in **Appendix B**; it allows for the impact of SD recirculation on SRT to be calculated as a function of the percentage of recirculated digestate (R dimensionless), of the efficiency of the separation unit (SI dimensionless), of the distribution factor of mass flow between SD and LD (α dimensionless), of digester and post-digester volumes ($RV_{digester}$ and $RV_{post\text{-}digester}$ in m^3), of the feedstock intake ($\dot{m}_{feedstock}$ in t/year), and of the mass of biogas produced ($\dot{m}_{biogas}(R)$ in t/year). Based on a similar framework to **Eq. 16,** the impact of recirculation on digester TS content ($[TS]_{digester}$ in t TS/t raw digestate) could be calculated according to **Eq. 17** that notably uses the initial TS content of feedstocks ($[TS]_{feedstock}$ in t TS/t of feedstock). Further details concerning the development of **Eq. 16** and **Eq. 17** can be found in **Appendix B**.

$$SRT = \frac{(1 + \alpha - R)}{(1 + \alpha) * (1 - R * SI)} * \frac{(RV_{digester} + RV_{post-digester})}{\frac{(\dot{m}_{feedstock} - \dot{m}_{biogas}(R))}{D_{operation}}} \Leftrightarrow SRT = \frac{(1 + \alpha - R)}{(1 + \alpha) * (1 - R * SI)} * HRT_{accurate}(R) \quad (16)$$

$$[TS]_{digester} = \frac{(1 + \alpha - R)}{(1 + \alpha) * (1 - R * SI)} * \frac{(\dot{m}_{feedstock} * [TS]_{feedstock} - \dot{m}_{biogas}(R))}{(\dot{m}_{feedstock} - \dot{m}_{biogas}(R))} \quad (17)$$

In the rest of this article, SI and α will be constant but further details concerning their range of validity for the general formula can be found in **Appendix B**. Since the study concerns agricultural biogas plants using screw presses that are low-efficiency phase separators, the average mass distribution profiles provided from a recent study on digestate mechanical separation [6] can be used. Hence, SI can be set to 38% and α to 9 (90% of flow weight goes to the LD and 10% to the SD). Previous equations can thus be simplified as follows:

$$SRT = \frac{(1 - \frac{R}{10})}{(1 - 0.38 * R)} * HRT_{accurate}(R) \quad (18)$$

$$[TS]_{digester} = \frac{(1 - \frac{R}{10})}{(1 - 0.38 * R)} * \frac{(\dot{m}_{feedstock} * [TS]_{feedstock} - \dot{m}_{biogas}(R))}{(\dot{m}_{feedstock} - \dot{m}_{biogas}(R))} \quad (19)$$

In these equations, R will be set to 30%, 50% and 80% of total produced SD. In addition, to determine $[TS]_{feedstock}$, first **Eq. 19** and $[TS]_{digester}$ provided by plant operators and obtained without recirculation will be used. Finally, it is noteworthy that SRT and $[TS]_{digester}$ should increase to a similar extent as the same coefficient is applied for both equations.

2.7.2. Direct SD recirculation - Replacement strategy

In the case of a replacement strategy, $\dot{m}_{feedstock}$ has to be replaced by the new amount of feedstock ($\dot{m}1_{feedstock}$) in **Eq. 15**, **Eq. 16**, **Eq. 17**, **Eq. 18** and **Eq. 19**. This new amount of feedstock can be determined using **Eq. 20** and the feedstock offset ($\Delta_{feedstock}$ in t/year) calculated using **Eq. 19**, that depends on the amount of digestate recirculated. Besides, as feedstock intake is decreased, TS of the initial feedstock ($[TS]_{feedstock}$) and initial biogas produced ($\dot{m}_{biogas}(0)$) is also reduced. It was assumed that TS of the produced feedstocks and biogas decreased linearly as a function of the amount of feedstock intake (homogenous repartition of the impact of feedstocks on TS and biogas production). New calculated values of $\dot{m}_{biogas}(0)$ and $[TS]_{feedstock}$ were used in **Eq. 18** and in **Eq. 19** respectively.

$$\dot{m}1_{feedstock} = \dot{m}_{feedstock} - \Delta_{feedstock}(R) \quad (20)$$

3. Results and discussion

3.1. Characterization and classification of solid digestates

Table 2 summarizes the physico-chemical and biological properties of the five SD. TS were comprised between 22.5% and 25.7% and their VS between 82.5% and 87.3% of TS. It is in the range of values found in literature for SD coming from a screw press comprised between 20 and 30% for TS and 80-90% of TS for VS [24]. Average C/N ratio for the five SD is 21, slightly lower than the optimal C/N ratio for AD often found between 25 and 30 [26].

Table 2: Solid digestate physico-chemical and biological properties. Main molecules present in the different OM fractions: SPOM – Soluble sugars & proteins; SEOM – Proteins; PEOM – Hollocellulose; NEOM – Lignin & complex humic acids

SD origin		Plant A	Plant B	Plant C	Plant D	Plant E
$[TS]_{SD}$ (%FM)		22.7±0.3	22.9±0.3	25.7±0.4	22.5±0.3	24.1±0.2
[VS] (%TS)		86±0.2	87.3±0.2	83.6±1	82.5±0.2	84.2±0.4
$[VS]_{SD}$ (%FM)		19.5	20.0	21.5	18.6	20.3
%C/g TS		36.3±0.9	36.0±0.5	33.9±0.4	34.4±0.7	33.3±0.8
%N/g TS		1.8±0.1	1.5±0.1	1.6±0.1	1.5±0.1	1.9±0.1
C/N		20.0	23.8	21.4	23.6	17.2
BMP test (Nm^3 CH_4/t VS)		130±4	145±3	99±6	191±3	155±6
COD_{tot} (gO_2/kg VS)		1,360±50	1,288±76	1,282±10	1,200±62	1,236±30
$\%BD_{ana}$ ($\%COD_{tot}$)		27.3	32.2	22.3	45.5	35.8
Chemical sequential extraction ($\%COD_{tot}$)	**SPOM**	5.0±0.1	3.5±0.1	2.7±0.1	2.7±0.1	3.2±0.1
	SEOM	10.8±0.1	12.9±1.1	12.7±0.1	12.7±0.3	11.2±0.2
	PEOM	39.8±0.1	52.9±0.2	43.6±3.0	50.3±1.9	46.2±1.6
	NEOM	44.3±0.3	30.7±1.4	41.0±4.4	34.3±2.4	39.4±2.5

BMP values are in accordance with values found in the literature for SD (60 to 240 Nm^3 CH_4/t VS) [10]. High BMP (> 145 Nm^3 CH_4/t VS) and anaerobic biodegradability (32-45% COD_{tot}), that were obtained for plants B, D and E, can be explained by the low to intermediate HRT of these plants (84, 88 and 40 days respectively). Intermediate to low BMP values were obtained for plants A and C (99-130 Nm^3 CH_4/t VS) leading to a lower anaerobic biodegradability (< 30% COD_{tot}). For plant A and C, whey, catch crop and beet pulp are the main feedstocks. They are easy to degrade, and applied HRT of 100 and 130 days, respectively, allow for most of the biogas from the feedstocks to be recovered, thus lowering the BMP value.

Chemical sequential extraction of the five SD, displayed in **Table 2,** gave insights into VS accessibility. For all SD, the sums of SPOM and SEOM fractions were close to 15% COD_{tot}, showing that most of the remaining VS is poorly bioaccessible. Two categories of SD can be distinguished: (i) SD with high hollocellulose content (PEOM higher than 45% COD_{tot}) and low lignin-like fraction (NEOM lower than 40% COD_{tot}), which corresponds to SD from plants B, D and E; (ii) SD with low hollocellulose content (PEOM lower than 45% COD_{tot}) and high lignin-like fraction (NEOM higher than 40% COD_{tot}), which corresponds to SD from plants A and C. Finally, considering the recent digestate typology made by Guilayn et al. [7], all SD in this study can be classified as fibrous material originating from low performance separation regarding the calculated TS, VS and C/N ratio.

Decision was made to: (i) further work on SD from plants A and C as they are representative of the agricultural CSTR biogas plant sector with long HRT, mainly easy to degrade feedstocks (> 40% total ration) and SD with high lignin-like content; (ii) eliminate among the last subgroup SD from plants D and E since they have too short HRT compared to the amount of feedstock they treat each year and to their mix (manure rich); (iii) also select SD from plant B as it displays an interesting profile with intermediate BMP and high hollocellulose content that is due, despite a long HRT and thermophilic conditions, to the high amount of manure used as feedstock (60%). The application of post-treatments on SD from plants A, B and C aims to increase their BMP. This can be obtained notably by a specific degradation of the lignin-like fraction that enhances the bioaccessibility of the organic matter.

3.2. Effect of thermo-alkaline and short-term aerobic post-treatments on solid digestate biomethane yield

Previous observations have demonstrated how separated manure fibres, after 3 days at 22°C soaking in an aqueous ammonia solution (32% w/w), entail a BMP increase up to 80% due to a strong lignin breakdown [27]. Similar effects, using sodium hydroxide (2% w/w) and thermophilic conditions (55°C for 3 days), were targeted on SD from plants A, B and C. Values of BMP and BD_{ana} following this thermo-alkaline post-treatment are given in **Table 3**. BMP of all treated SD increased by 30 to 46%.

Table 3: Effect of thermo-alkaline and short-term aerobic post-treatment on SD on the BMP of final post-treated matter

Sample Name	Airflow (L/h/kg TS)	Duration (days)	C loss (% TC)	BMP_{FPTM} (Nm^3 CH_4/t VS_{final})	BMP_{FPTM} variation
Thermo-alkaline post-treatment					
Plant A control	/	/	/	155±3	/
Plant A SD treated	/	/	/	209±7	+35%
Plant B control	/	/	/	150±4	/
Plant B SD treated	/	/	/	195±3	+30%
Plant C control	/	/	/	129±4	/
Plant C SD treated	/	/	/	188±5	+46%
Short term aerobic post-treatment					
Plant A control	/	/	/	153±8	/
Plant A SD1	1.65	6	7.1	154±5	0%
Plant A SD2	10	3.6	4.5	126±3	-21%
Plant A SD3	20	1.75	2.8	142±4	-8%
Plant A SD4	20	3.6	4.8	141±2	-8%
Plant A SD5	20	3.6	6.7	140±3	-9%
Plant B control	/	/	/	171±4	/
Plant B SD1	1.65	5	3.1	149±3	-15%
Plant B SD2	16.5	5	8.6	159±4	-8%
Plant B SD3	33	5	10.5	152±4	-13%

In a study on the effect of composting storage on BMP SD, Menardo *et al.* showed that composting between two and four weeks led to an approximate 30 % decrease in BMP [15]. During the composting process, mesophilic and thermophilic phases favour the removal of easily degradable matter, cellulose and hemicellulose. Lignin components are mainly degraded during the maturation phase [28]. SD VS evolution was not studied by Menardo *et* *al*., but their results seem to indicate that over a medium-term, easily biodegradable fractions are better degraded than complex ones, thus explaining the lower BMP.

In this study, the effect of shorter and controlled aeration on SD was evaluated for its potential as a low-cost biological post-treatment practice at full scale [29]. It was hypothesized that short-term forced aeration can favour lignin degradation over easily degradable fractions and therefore increase biodegradability and BMP. SD from plants A and B were selected from the two previously described subgroups, as the lignin-like fraction (NEOM) content was high (44% COD_{tot}) for the former and low (31% COD_{tot}) for the latter. **Table 3** presents anaerobic biodegradation results. They suggest that the BMP of final post-treated matter (BMP_{FPTM}) was not improved in comparison to the control for both plant SDs. For plant A SD, the BMP_{FPTM} ranged from 126 to 154 Nm^3 CH_4/t VS_{final} with values either significantly lower or similar to the control. For plant B, raw BMP_{FPTM} values were also always significantly lower than the control. These lower methane potentials are due to carbon losses during the short-term aerobic post-treatment.

Additional analyses on treated plant B SD are given in **Appendix C**. It provides supplementary information on evolution of the different carbon fractions during these short-term aerobic post-treatments.

3.3. Impact of various strategies of solid digestate recirculation on plant methane production and evaluation of potential economic gains or capacity offset.

Table 4 displays calculation results that evaluate the effect of direct recirculation of 30%, 50% and 80% of SD produced each year on plant methane production. Moreover, the potential additional economic gains (AEG) in an addition strategy case and the amount of feedstock that can be offset ($\Delta_{feedstock}$) in a replacement strategy case are also provided.

Firstly, the higher the amount of SD directly recirculated, the higher the impact on plant methane production. Direct recirculation of 30% of SD represents between 0.6% and 2.3% of total plant methane production and when recirculating 80% of SD reaches between 1.6% and 6.3%. Plants D and E that have a high amount of feedstock, a short to medium HRT and mesophilic conditions display the highest impact of recirculation on plant methane production that can be explained by the relatively high BMP as well as SD amount produced yearly. Interestingly, the lowest impact of recirculation on plant methane production (only 1.6% at 80% SD recirculation) is obtained for plant C. It can mainly be explained by the long HRT and the use of easy to degrade feedstocks that leads to low SD BMP.

Secondly, as a function of the recirculation strategy applied, impact on plant methane production differs:

(i) If recirculation is performed following an addition strategy, the percentage of plant methane production calculated corresponds to a potential increase of the plant methane production. In this case, additional economic gains (AEG) were calculated and range from 8k€ to 83.2k€ per year as a function of the plant, the FIT and the percentage of SD recirculated. Such economic gains are not negligible for farmers. Besides, direct recirculation strategy does not require a high cost expenditure to be implemented, as part of the produced SD is simply sent back into the hopper. It requires only a low operational expenditure (OPEX) involving a short daily labour time (few minutes) and associated tractor operations (gasoline…).

(ii) If recirculation is performed following a replacement strategy, the percentage of plant methane production calculated corresponds to an offset capacity of the plant methane production in the event, for instance, of a feedstock shortage. It does not generate any additional income from biogas but it can allow the maintenance of a steady methane production as well as its associated revenues. If the TS and methane yield from feedstock are assumed to be evenly distributed, such a strategy can replace between 64 and 1431 tons of feedstock according to the plant (plant size, SD BMP) and to the percentage of recirculated SD. As SD is a free resource produced on site, direct recirculation would be be a low-cost alternative to buying feedstock when the plant ration is diminished.

Finally, it is noteworthy that for both recirculation strategies, the quantities incorporated on a daily basis in the digester (SD + feedstock) are increased. Indeed, even in the case of a replacement strategy, for the 6 studied biogas plants, on average 3.3 times more quantities of SD than feedstock are needed to offset methane production loss. This is due to the fact that SD has a lower methane potential than feedstocks on average as it has already been digested once. Therefore, conditions of implementation at full-scale will need to be further discussed and notably their impact on plant TS.

Table 5 presents for plant A, B and C the effect of recirculation of post-treated SD on plant methane production. Additional thermo-alkaline post-treatment increases SD BMP between 30 and 46%. In comparison to direct recirculation, this allows an additional increase of total plant methane production between 0.2% (plant B – 30% SD recirculated) and 1.2% (plant A – 80% SD recirculated). Consequences vary as a function of the type of recirculation but conclusions remain similar:

(i) In an addition strategy, recirculation of post-treated SD leads to AEG between 10.1k€ and 59k€ per year. In comparison to direct recirculation, AEG after a thermo-alkaline post-treatment are increased (+2.1k€ to +17.7k€). However, these economic gains will hardly compensate for additional post-treatment expenditures. Purchase of soda (540€/t at 2% w/w), on average already accounts for 88% of the additional gains of plant A, 300% of the additional gains of plant B and 67% of the additional gains of plant C. Besides, other expenditures can be added to the cost of soda. OPEX has to be increased due to additional labour and heating (thermophilic conditions) and the potential capital expenditure (CAPEX), such as the cost for a post-treatment tank (at least 30k€). Therefore, such a strategy does not prove to be economically viable.

(ii) Similar conclusions can be drawn in the case of a replacement strategy. Soda post-treated SD allow to offset an additional amount of feedstock comprised between 49 t (plant B – 30% SD recirculated) and 163 t (plant A – 80% SD recirculated) per year (in this specific case, regarding the loading rate, it represents 8 days of feedstocks). However, investment costs are again too high and buying feedstocks should represent a less expensive strategy. As an example, in the case of plant A with 80% SD recirculation, cost for soda (15.5k€ per year) could be allocated instead to buy at least 346 t of catch crops at a high price of 45€ per ton (cost ranges normally between 15 and 45€/t) [30]. This amount is more than two times higher than the additional 163 t of feedstocks offset by recirculated post-treated SD. Consequently, in this case, buying feedstock largely outcompetes the thermo-alkaline post-treatment of SD.

Short-term aeration post-treatments lead to lower SD BMP for plant A and B SD. Calculations based on the recirculation of 50% of the total produced SD show that the recirculation impact on total plant methane production is lower than for direct recirculation. Thus, in the case of an addition strategy, slightly lower AEG are obtained in comparison to direct recirculation (-800€ to -1,800€). Similarly, for a replacement strategy case, the amount of feedstock that can be offset is slightly lower (-16 to -20 t/year). These results suggest that short-term aeration is not a significant strategy either, despite its low cost. Besides, they also indicate that SD should not be aerated nor stored in open air for too long before being recirculated, to avoid methane losses in the form of CO_2 respiration.

Finally due to the low impact of SD direct recirculation on the total plant methane production (mainly originating from low SD BMP), a post-treatment, however effective it may be in tested scenarios, is unlikely to be profitable. Indeed, additional CAPEX and OPEX may always be greater than the income or offset capacity related to additional methane.

Table 4: Impact of 30%-50% and 80% SD direct recirculation on plant methane production and its additional economic gain in an addition strategy case or the amount of feedstock that can be offset in a replacement strategy case

Ratio of total SD produced that is recirculated		Plant A	Plant B	Plant C	Plant D	Plant E
	BMP at lab scale (Nm^3 CH_4/t VS)	146	155	114	191	155
	$BMP_{corrected\_SD}$ (Nm^3 CH_4/t VS)	117	124	91	153	124
30%	$\dot{m}_{SD\_recirculated}$ (t/year)	540	600	360	990	750
	Mass of organic solids of $\dot{m}_{\%SD\_recirculated}$ (t VS/year)	106	118	77	184	152
	V_{CH4_SD} (Nm^3 CH_4/year)	12,402	14,632	7,007	28,152	18,848
	$\%_{total\_plant\_CH4}$ (%)	1.04	1.05	0.61	2.15	2.34
	AEG – addition strategy case (k€/year)	15.5	8	8.9	31.2	14.4
	$\Delta_{feedstock}$ – replacement strategy case (t/year)	141	189	64	537	468
50%	$\dot{m}_{SD\_recirculated}$ (t/year)	900	1,000	600	1,650	1,250
	Mass of organic solids of $\dot{m}_{\%SD\_recirculated}$ (t VS/year)	176	197	129	306	254
	V_{CH4_SD} (Nm^3 CH_4/year)	20,592	24,428	11,739	46,818	31,496
	$\%_{total\_plant\_CH4}$ (%)	1.73	1.75	1.02	3.56	3.91
	AEG – addition strategy case (k€/year)	25.8	13.3	14.8	52.0	24
	$\Delta_{feedstock}$ – replacement strategy case (t/year)	235	315	107	894	783
80%	$\dot{m}_{SD\_recirculated}$ (tons/year)	1,440	1,600	960	2,640	2,000
	Mass of organic solids of $\dot{m}_{SD\_recirculated}$ (t VS/year)	282	315	206	490	406
	V_{CH4_SD} (Nm^3 CH_4/year)	32,994	39,060	18,746	74,970	50,344
	$\%_{total\_plant\_CH4}$ (%)	2.77	2.8	1.63	5.71	6.25
	AEG – addition strategy case (k€/year)	41.3	21.3	23.7	83.2	38.5
	$\Delta_{feedstock}$ – replacement strategy case (t/year)	376	504	172	1,431	1251
Similar for 30%-50%-80%	Ratio $\dot{m}_{SD\_recirculated}/\Delta_{feedstock}$	3.83	3.17	5.59	1.84	1.60

Table 5 Impact of various SD post-treatments followed by recirculation on biogas plant methane production and its economic impact in comparison to direct SD recirculation

Post-treatment type	Ratio of total SD produced that is recirculated		Plant A	Plant B	Plant C
Thermo-alkaline		**$BMP_{corrected\_SD}$ (Nm^3 CH_4/t VS)**	167	156	150
	30%	**V_{CH4_SD} (Nm^3 CH_4/year)**	17,702	18,408	11,550
		$\%_{total\_plant\_CH4}$ (%)	1.49	1.32	1.00
		AEG – addition strategy case (k€/year)	22.2	10.1	14.6
		AEG variation compared to direct recirculation (k€/year)	+6.7	+2.1	+5.7
		$\Delta_{feedstock}$ – replacement strategy case (t/year)	203	238	106
		$\Delta_{feedstock}$ variation compared to direct recirculation (t/year)	+62	+49	+42
	50%	**V_{CH4_SD} (Nm^3 CH_4/year)**	29,392	30,732	19,350
		$\%_{total\_plant\_CH4}$ (%)	2.47	2.20	1.69
		AEG – addition strategy case (k€/year)	36.8	16.7	24.5
		AEG variation compared to direct recirculation (k€/year)	+11	+3.4	+9.7
		$\Delta_{feedstock}$ – replacement strategy case (t/year)	336	397	177
		$\Delta_{feedstock}$ variation compared to direct recirculation (t/year)	+101	+82	+70
	80%	**V_{CH4_SD} (Nm^3 CH_4/year)**	47,094	49,140	30,900
		$\%_{total\_plant\_CH4}$ (%)	3.96	3.52	2.69
		AEG – addition strategy case (k€/year)	59	26.8	39.1
		AEG variation compared to direct recirculation (k€/year)	+17.7	+5.5	+15.4
		$\Delta_{feedstock}$ – replacement strategy case (t/year)	539	635	283
		$\Delta_{feedstock}$ variation compared to direct recirculation (t/year)	+163	+131	+112
Short-term aeration		**$BMP_{corrected\_SD}$ (Nm^3 CH_4/t VS)**	109	116	/
	50%	**V_{CH4_SD} (Nm^3 CH_4/year)**	19,184	22,852	/
		$\%_{total\_plant\_CH4}$ (%)	1.61	1.63	/
		AEG – addition strategy case (k€/year)	24.0	12.5	/
		AEG variation compared to direct recirculation (k€/year)	-1.8	-0.8	/
		$\Delta_{feedstock}$ – replacement strategy case (t/year)	219	295	/
		$\Delta_{feedstock}$ variation compared to direct recirculation (t/year)	-16	-20	/

3.4. Assessment of the impact of SD recirculation on HRT, SRT, digester TS and of mixing costs on CSTR biogas plant

The evolution of CSTR plant $HRT_{accurate}$ and SRT as a function of the applied recirculation strategy and for three different percentages of recirculated SD (30-50 and 80% of $\dot{m}_{SD}$) are presented in **Table 6**. For the five CSTR biogas plants studied and in the case of an addition strategy, $HRT_{accurate}$ only slightly increases (less than one percent on average) since the additional methane production from SD is relatively low in comparison to total methane production. However, for SRT, the increase is more significant. SRT are on average 10%, 18% and 33% higher, comparatively to SRT without recirculation, when 30%, 50% and 80% SD are recirculated, respectively. For a replacement strategy, $HRT_{accurate}$ and SRT increases are slightly better, compared to an addition strategy because the initial feedstock supply was reduced. Thus, comparatively to $HRT_{accurate}$ and SRT without recirculation, $HRT_{accurate}$ are on average 2%, 3% and 4% higher and SRT are on average 11%, 21% and 38% higher, when 30%, 50% and 80% SD are recirculated, respectively.

For both strategies, recirculation of a fraction of total produced SD has a negligible impact on the $HRT_{accurate}$ but allows for the SRT to increase because the SD remains longer within the biogas plant. These higher SRT values lead to a higher methane recovery (as SD BMP between 67 and 191 Nm^3 CH_4/t VS). This can account for the previously determined increase in plant methane production as well as the higher biogas plant efficiency. Finally, 200 days of AD are seemingly required to reach a 95% methane recovery from the most complex feedstocks to degrade, such as manure [31]. For plant SRT to approach such a value via SD recirculation would be noteworthy for biogas plants that treat complex feedstocks (e.g. plant A, B or D) in order to allow a high biogas plant efficiency. However, impact of recirculation on the digester TS content should also be taken into account.

Table 6: Evolution of CSTR plant HRT, SRT and digester TS content as a function of recirculation strategy applied and for different percentages of SD recirculated

Plant Name	Plant A	Plant B	Plant C	Plant D	Plant E
$RV_{digester}$ (m^3)	2,000	2,200	2,000	3,700	1,200
$RV_{post\text{-}digester}$ (m^3)	2,000	2,200	2,000	2,700	1,200
$\dot{m}_{feedstock}$ (t/year)	13,600	18,000	10,500	25,000	20,000
$HRT_{classical}$ (days)	100	84	130	88	40
$\dot{m}_{biogas}(0)$ (t/year)	2,494	3,184	2,547	2,749	1,641
$HRT_{accurate}$ = SRT (days) for R=0%	123	102	172	98	45
Initial $[TS]_{digester}$ (%TS)	12.0	10.5	10.5	14.0	7.6
Calculated Initial $[TS]_{feedstock}$ (%TS)	28.1	26.3	32.2	23.5	15.2
Strategy	**Addition**				
$HRT_{accurate}$ for R=30% (days)	123	102	172	99	45
$HRT_{accurate}$ for R=50% (days)	124	102	173	99	45
$HRT_{accurate}$ for R=80% (days)	124	103	173	99	45
SRT for R=30% (days)	135	111	189	108	49
SRT for R=50% (days)	145	119	202	116	53
SRT for R=80% (days)	164	135	229	131	59
$[TS]_{digester}$ for R=30% (%TS)	12.9	11.3	11.3	15.1	8.1
$[TS]_{digester}$ for R=50% (%TS)	13.7	11.9	12.0	16.0	8.5
$[TS]_{digester}$ for R=80% (%TS)	15.1	13.1	13.2	17.7	9.3
Strategy	**Replacement**				
$HRT_{accurate}$ for R=30% (days)	125	103	173	101	46
$HRT_{accurate}$ for R=50% (days)	126	104	174	102	47
$HRT_{accurate}$ for R=80% (days)	127	105	176	105	48
SRT for R=30% (days)	137	113	190	110	50
SRT for R=50% (days)	148	122	205	120	55
SRT for R=80% (days)	169	139	232	139	63
$[TS]_{digester}$ for R=30% (%TS)	12.8	11.1	11.2	14.7	7.9
$[TS]_{digester}$ for R=50% (%TS)	13.4	11.7	11.9	15.4	8.2
$[TS]_{digester}$ for R=80% (%TS)	14.7	12.7	13.0	16.6	8.7

Indeed, digester TS will be modified when SD recirculation is performed. This is due to the fact that in CSTR biogas plants, the volumes of the digester ($RV_{digester}$) and post digester ($RV_{post\text{-}digester}$) cannot be increased as they are definitively set once constructed. In general, this feature is due to: i) an overflow system involving a pipeline between the digester and post-digester in which digestate is transferred from the former to the latter; ii) a sensor system set in the post-digester that switches the phase separator on when a certain height of digestate is reached (generally corresponding to the height of the overflow system), then switches it off when the level drops to a lower pre-defined digestate height (e.g. switch on at

5.30 m and switch off at 5.20 m). Thanks to this system, digestate can flow from the digester to the post-digester. The position of the overflow pipeline coincides with the maximum height that the digestate can reach and thus sets the volume of the CSTR biogas plant. Hence, if the SRT increases while the volume of the biogas plant is fixed, the TS content of digester should increase.

Table 6 displays the evolution of digester TS according to the applied recirculation strategy and to the different percentages of recirculated SD. For the five studied CSTR biogas plants, results point out that an increase in the amount of recirculated SD also leads to an increase in digester TS for both recirculation strategies. Thus, for an addition strategy, the initial digester TS content rises, on average, by 7%, 14% and 25% (representing an average +0.8, +1.5, +2.8% increase of the initial digester TS value), when 30%, 50% and 80%, respectively, of the total produced SD is recirculated,. In the case of a replacement strategy digester, the initial digester TS content rises on average by 6%, 11% and 20% (representing an average +0.6, +1.2, +2.2% increase of the initial digester TS value), when 30%, 50% and 80%, respectively, of the total produced SD is recirculated. Slightly lower increases in TS values for the replacement strategy than for the addition strategy can be explained by the removal of certain feedstocks from the initial feedstock supply. Such rises in digester TS content are not negligible; furthermore they should modify digestate viscosity and potentially affect digester and post-digester mixing. Digestate rheology and mixing costs need to be further discussed, to better define conditions of application for SD recirculation.

Rheological studies in anaerobic digestion have been historically focused on activated sludge and raw manure. Only recently have several studies have been performed on digestate from agricultural biogas plants [32]. In most of these studies, agricultural digestate originates from a co-digestion process, and consists of a mix of livestock manure and various biomasses that display a non-Newtonian shear-thinning flow behaviour, which is frequently modelled via the power-law model [33]. A study performed in a full-scale plant using an in-line viscometer demonstrated that an increase in the TS content of the digester, entails an increase in the apparent viscosity of digestate. Associated power-law flow-behaviour model for this biogas plant digestate shows that at a low shear rate (10 s^{-1}), a shift from 11.2% TS to 11.7% TS in the digester (+0.5% TS) increases apparent viscosity of digestate by 35% [34]. Similarly, digestates with a higher TS content were also reported to present a higher viscosity than digestates with a lower TS content [35]. However, contrarily to manure or activated sludge, studies have indicated that the TS content only is insufficient for providing a reliable estimation of agricultural digestate rheological properties [36]. Indeed, agricultural digestates have a complex structure, notably comprising a large quantity of particulate matter. Additional parameters need to be taken into account to fully determine the rheological properties of agricultural digestate such as particle size, particle size distribution and gel forming structure (e.g. mucilage) [32]. To illustrate the impact of these additional parameters on viscosity, size reduction of solids via mechanical treatments have been found to reduce digestate viscosity [37]. Hence, for SD recirculation, if only the increase in TS content is known, it is not possible to precisely determine the extent of the increase in digestate viscosity, although a significant rise can be expected.

According to a long-term study performed on a full-scale research biogas plant, an average of 4% of the total electricity produced (CHP unit) is used for mixing [38]. This value corresponds to 1.6% of the total methane produced with an electrical efficiency of 40% and lies within the same range as the potential gains or capacity offset in methane production

(0.61 to 6.25% of total methane produced). If this value is used as an example and is specific to one digester, it implies that the mixing energy consumption is significant and should be taken into account when evaluating the SD recirculation strategy. If the TS content and digestate viscosity increase after SD recirculation, it is likely that the mixing energy consumption would also increase. However, it remains difficult to determine precisely, and even more to generalise, the extent to which the energy consumption can rise. For instance, for 13 full-scale biogas plants, no clear relationship has been observed between the average TS content during operation and mixing energy consumption, since the highest TS content is not associated to the highest mixing energy consumption [39]. This can be explained by the fact that mixing energy consumption is not only dependant on the TS content as well as digestate viscosity but also on the type of mixers, their numbers, the type of impellers, the agitation frequency and the agitation speed [39]. All these parameters can greatly vary from one biogas plant to another; therefore the sensitivity of mixing energy consumption towards an increase in TS can also be likely to vary according to the biogas plant.

Indeed, because of i) the specificity of each biogas plant mixing system and of ii) the current lack of precise correlation between a digestate TS and its viscosity, a turnkey equation allowing for the variation in mixing costs as a function of digester TS cannot be provided. For a given biogas plant, the relationship between mixing costs and digestate TS should rely on the operator’s empirical knowledge. Overall, the impact of SD recirculation on digestate viscosity and mixing costs can vary strongly from one biogas plant to another. The relevance of SD recirculation therefore has to be evaluated on a case-by-case basis; this will be further described in section 3.5. Nevertheless, regarding the potential increase in digester TS content displayed in **Table 6**, recirculation above 50% of total SD produced is likely to be unreasonable and not economically profitable. A noteworthy potential trade-off between additional methane or offset capacity and higher mixing cost may even consist in low to medium recirculation percentage values (R≤0.5).

3.5. A potential approach to determine the significance of SD recirculation for a given CSTR biogas plant

In order to evaluate the significance of SD recirculation for a given CSTR biogas plant, the following approach can be applied. Firstly, the operator must determine the VS content and BMP of its SD as well as the annual quantity of SD produced from the phase separation system ($\dot{m}_{SDyear}$). This information would allow for the potential gains/offset capacity to be calculated using **Eq. 7**, **8**, **9** and **10**. The potential increase in TS could then be calculated according to the % of SD recirculated (R) using **Eq. 14** as well as **Eq. 17** based on average $[TS]_{digester}$, annual quantity of feedstock incorporated ($\dot{m}_{feedstock}$), annual quantity of biogas produced ($\dot{m}_{biogas}$) and phase separator properties (α and SI). The new calculated digester TS values should then be compared with the working range of installed mixing equipment. **Appendix D** provides an overview of the estimated upper TS content range that a certain type of mixer can handle. It shows that except for submersible motor mixers equipped with a propeller, most existing mixing technologies can handle TS contents above 10%. If the new calculated digester TS lies within the working range, the operator should be able to determine (from a historical monitoring of the plant) the potential impact of a given increase in TS on the mixing costs. When applicable, observations dating from periods with a similar TS content might be useful for assessing the extent of the increase in the electrical consumption or maintenance rate. Besides, the presence of an on-site solid size-reduction device would have to be taken into account since it would reduce the impact of SD recirculation on digestate viscosity. If the plant operator should consider that the potential increase on mixing costs would be sufficiently low in comparison to the potential gains/offset capacity, then the implementation of SD recirculation can be initiated.

During the implementation of SD recirculation, it is likely that, while the digester TS rises, SD production from the screw press should also increase. Thus, if SD recirculation should be performed over a long-term basis, it may be expected that the total produced SD during the initial year ($\dot{m}_{SDyear}$) will be lower than the total produced SD of the following years ($\dot{m}_{SDyear+n}$). Therefore, over a long-term, the biogas plant operator should maintain the initial amount of recirculated SD (determined during the first year) instead of recalculating it every year as a percentage of the new total SD produced ($\dot{m}_{SDyear+n}$). In this way, a progressive increase over time in the amount of recirculated SD would be avoided and the recirculation process would remain stable.

Finally, if the digester already has a high TS content in comparison with its mixing equipment, a partial replacement strategy can be an alternative to the replacement strategy, since its impact on the TS content is lower. For instance, the quantity of missing feedstocks can be replaced by the same quantity of SD. In such a specific case the impact on digester TS content would be on average 45% lower than for a replacement strategy. Applying such a strategy would limit loss in biogas production and increase the biogas plant efficiency.

Figure 2 summarizes the findings from this study in the form of a decision diagram destined to biogas plant operators that wish to implement SD recirculation strategies. This decision diagram can be applied to all CSTR biogas plants located across various countries. The potential economic gains should be recalculated using local biomethane/electrical tariffs.

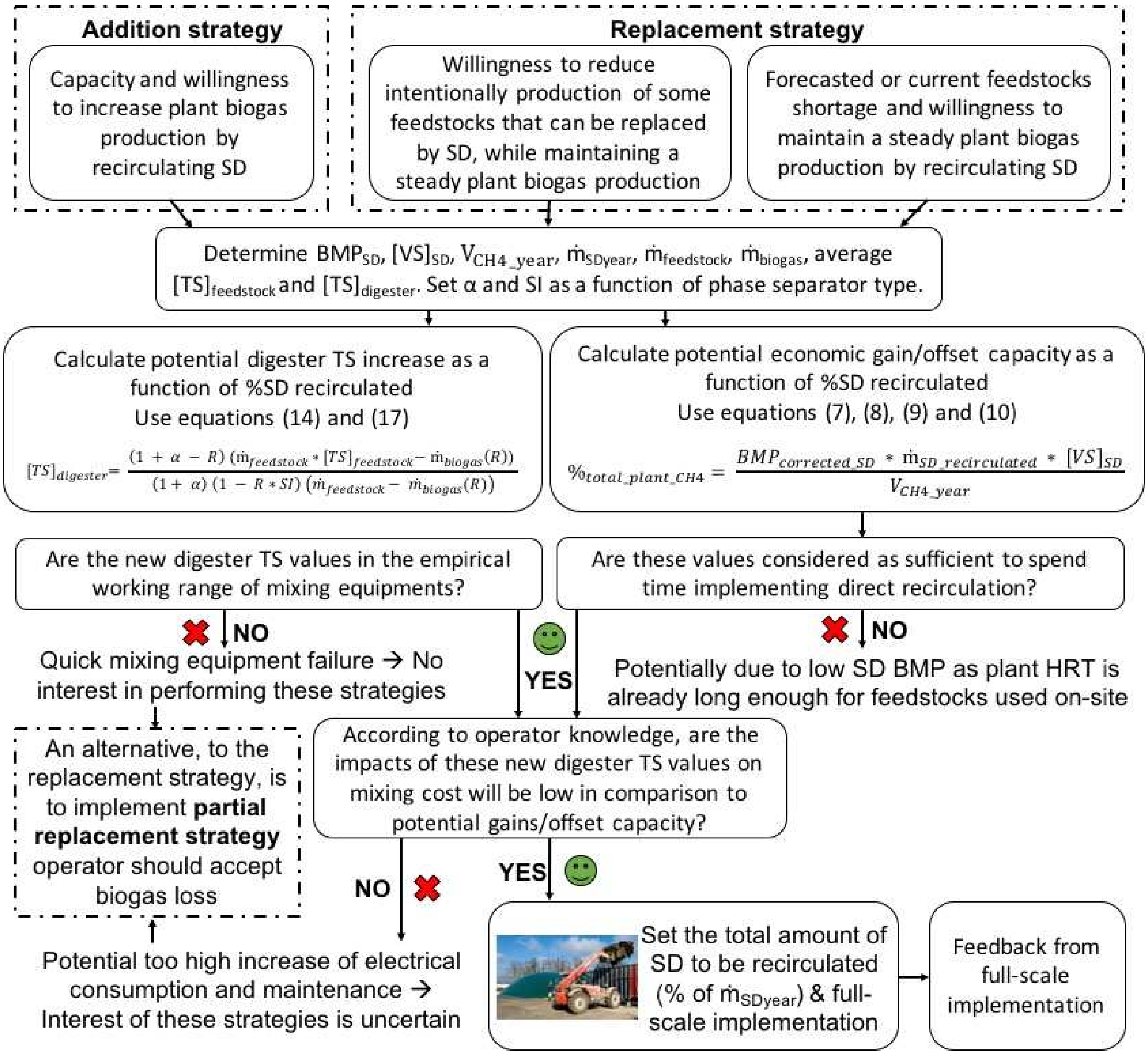


Figure 2: Decision diagram for plant operators from all countries to determine whether SD recirculation following an addition or a replacement strategy can be relevant and viable.

Two examples of the procedure to be adopted regarding the potential TS increase and installed mixing system can be provided after discussion with plant A and B operators. For plant A, the digester is equipped with 3 Amaprop® submersible motor mixer with large-blade rotors (KSB, Frankenthal, Germany). The average TS content in the digester without recirculation (12%) is in the high range of mixing equipment working capacity. At this TS, maintenance operations are already twice more frequent than for the post-digester that has the same mixing equipment but with a lower TS due to biogas production (on average close to 10.5% TS). Therefore, recirculation does not appear for the plant operator to be a relevant strategy unless it should remain at low levels (R≤0.1). A partial replacement strategy could also be envisioned if ever feedstock shortage occurs. For plant B, the digester is equipped with one Flyght® 4430 submersible motor mixer with large-blade rotors (Xylem, New York, United-States) and two Biogator® HPR 1 paddle mixers (REMA GmbH, Hausen, Germany). For this plant, the average TS content in the digester without recirculation (10.5%) is in the low range of mixing equipment working capacity (13 to 14% TS). Besides, the presence of a PreMix incorporator, equipped with a RCX® Rotacut shredder (Vogelsang, Essen, Germany) may potentially reduce the impact of SD recirculation on digestate viscosity. Here, recirculation can be envisioned as a potential strategy; indeed, even at a high percentage of recirculation (R=0.8), the TS in the digester would remain within the working range. The relevance of this strategy remains to be assessed by the plant operator according to the potential gain/offset capacity and to the estimated increase in mixing costs.

Finally, different full-scale implementation cases for solid and liquid digestate recirculation can be identified. LD recirculation can be used to dilute high TS feedstocks. In cases, where storage tank is not covered and heated, additional methane might be obtained from LD. However, particular attention should be paid to ammonia inhibition, especially for plants treating nitrogen rich feedstocks, as it may accumulate in digesters with LD recirculation. Intermediate air stripping devices for nitrogen removal [40] or adaptation of the recirculation rates [41] might represent potential solutions to avoid inhibitions. In comparison, SD recirculation may serve to increase the TS content for plants treating large amounts of liquid feedstocks. Besides, as indicated for composted solid digestate, it may have an additional buffering impact in the digester and allow for stabilisation In addition, according to the recirculation rate, both LD and SD recirculation are likely to allow for an increase or at least sustain anaerobic microbial load and community in the digester. The application of these recirculation strategies should depend on the features of the biogas plant as well as the different stages of its operation (onset of acidogenesis, seasonal high loading of TS feedstocks...). Further full-scale studies to increase our understanding of such practices (for instance on potential heavy metals accumulation in the digester) are required as they are likely to be part of the occasional/permanent tools to reach excellence in biogas plant operation.

4. Conclusions

In this work, three possible recirculation strategies have been defined for the first time: "addition", "replacement" and "partial replacement". Direct recirculation was then proposed as the best strategy to adopt, since additional tested post-treatments (thermo-alkaline, aerobic) did not prove to be economically viable. The impact of SD direct recirculation was estimated to enable the plant to produce an additional 0.6 to 5.7% of biogas or to compensate for 64 to 1431 tons/year in case of feedstock shortage.

Moreover, this article provides practical tools for assisting operators from all countries in setting up the required conditions for the implementation of such practices in their CSTR biogas plants. Focus was put on a trade-off to be found between potential economic gains and increase in agitation costs. Case-by-case analysis is therefore recommended to assess the relevance of direct solid digestate recirculation for a given biogas plant. Under certain conditions, this approach can be considered good management practice that might allow agricultural CSTR biogas plant energy efficiency to increase without any additional CAPEX and with a low OPEX (short daily labour time, associated tractor operations and additional low mixing costs). Such a potential increase is significant for biogas plant operators and represents a relevant way to reduce biogas production costs.

In order to fully complete this study in the future, better knowledge of the existing relationships between digestate TS, digestate viscosity and energy mixing requirements for a diversity of mixing systems would need to be developed, and the cost-relevance of SD could thus also be determined with higher accuracy.

Following this work, several recommendations/perspectives emerge, to gain further knowledge on SD direct recirculation practices. This includes: (i) additional potential co-benefits of direct SD recirculation such as an improved hydrolysis step or a higher buffering capacity may be further assessed at lab or pilot scales; (ii) a study covering the impact of this practice on plant sustainability (via a life cycle analysis) and digestate agronomic properties. It is likely that this second strategy would be equally beneficial regarding greenhouse gas emissions of biogas plants. Indeed, during recirculation, additional carbon is converted to methane, while this would otherwise have been released in the form of carbon dioxide during SD composting. (iii) performing full-scale trials to fully confirm that direct SD recirculation following an addition strategy may effectively enhance plant methane production; (iv) better understanding the synergies that might exist with other type of digestate flows that can be recirculated within a biogas plant (raw and liquid digestates).

Acknowledgment

National Research and Technology Association (ANRT) is gratefully acknowledged for the PhD grant allocated to Ulysse Brémond (reference CIFRE N° 2016/0617).

Declaration of interest

Declarations of interest: none

Appendix A

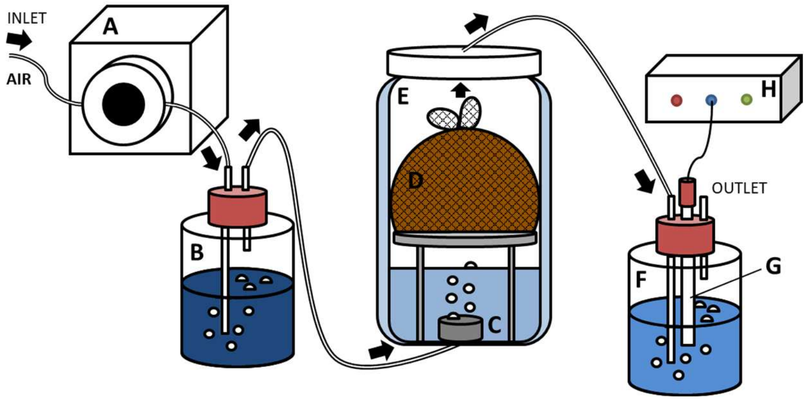


Figure A: Experimental set-up for short-term aerobic post-treatment of SD. (A) Calibrated peristaltic pump; (B) Air CO2 trap using 2M NaOH solution; (C) Humidifier system; (D) SD contained in a net; (E) 2.5 litres double jacket aerobic reactor at 30°C; (F) NaOH 0.5M trap for CO2 emitted from SD respiration; (G) Conductivity probe; (H) Acquisition system.

Appendix B

In the case of a CSTR biogas plant equipped with a phase separator and producing SD and LD, it is possible to describe the system according to **figure B**:

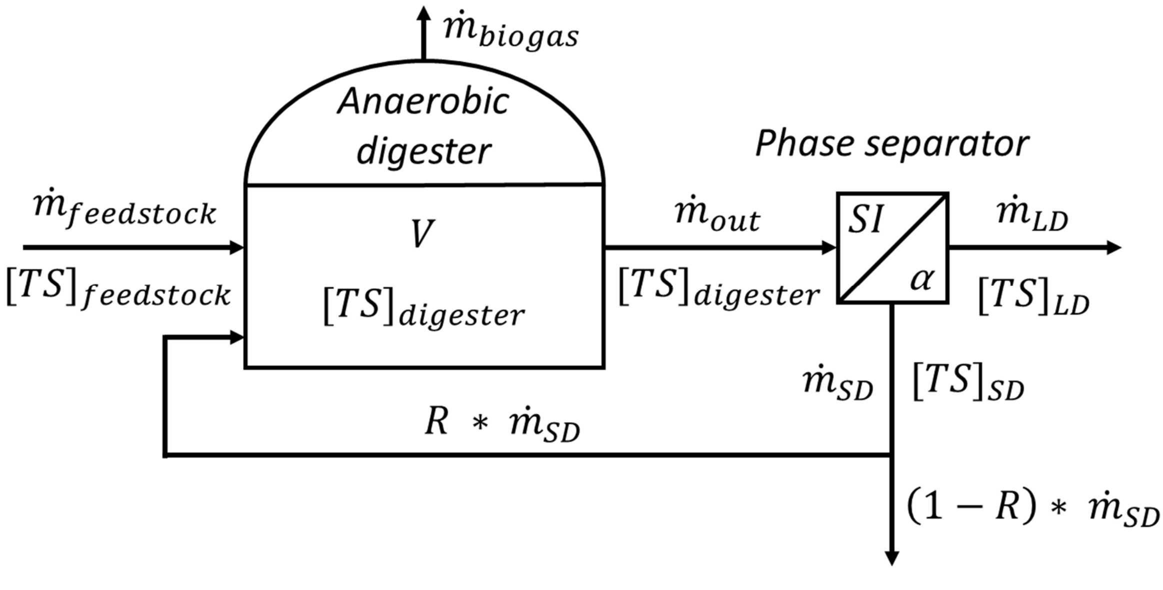


Figure B: CSTR biogas plant equipped with a phase separator and performing solid digestate recirculation

With the following known variables:

- $\dot{m}_{feedstock}$: mass of feedstocks incorporated per day (tons/day)

- $\dot{m}_{biogas}$: mass of biogas produced per day (tons/day)
- V : volume of the anaerobic digester (m^3)
- $[TS]_{feedstock}$: concentration in total solids of feedstock (ton TS/ton feedstock)
- SI : efficiency of the separation unit - Defined
- α : the repartition factor of mass flow between solid digestate and liquid digestate -
- R : the percentage of recirculated solid digestate - Defined

As well as the following unknown variables:

- $\dot{m}_{out}$: mass of raw digestate leaving the digester per day (tons/day)
- $\dot{m}_{LD}$: mass of liquid digestate produced per day (tons/day)
- $\dot{m}_{SD}$: mass of solid digestate produced per day (tons/day)
- $[TS]_{digester}$: concentration in total solids of raw digestate (ton TS/ton raw digestate)
- $[TS]_{LD}$: concentration in total solids of liquid digestate (ton TS/ton liquid digestate)
- $[TS]_{SD}$: concentration in total solids of solid digestate (ton TS/ton solid digestate)

It is important to specify that for the rest of the calculations the following strong hypotheses were made:

- Density of all feedstocks and digestate were equal to one. This allows switching freely from mass to volume.
- $[TS]_{digester}$ was considered to be the total solids content of the raw digestate entering in the phase separator ($\dot{m}_{out}$). Ideally, $[TS]_{post\text{-}digester}$ should be considered.

The seven following equations can be defined, based on a steady state system and existing definitions:

$$\dot{m}_{feedstock} - \dot{m}_{biogas} = \dot{m}_{LD} + (1 - R) * \dot{m}_{SD}$$

(B.1)

$$\dot{m}_{feedstock} * [TS]_{feedstock} - \dot{m}_{biogas} = \dot{m}_{LD} * [TS]_{LD} + (1 - R) * \dot{m}_{SD} * [TS]_{SD}$$

(B.2)

$$\dot{m}_{out} = \dot{m}_{LD} + \dot{m}_{SD} \quad \text{(B.3)}$$

$$\dot{m}_{out} * [TS]_{digester} = \dot{m}_{LD} * [TS]_{LD} + \dot{m}_{SD} * [TS]_{SD}$$

(B.4)

$$\alpha * \dot{m}_{SD} = \dot{m}_{LD} \quad \text{(B.5)}$$

$$SI = \frac{\dot{m}_{SD} * [TS]_{SD}}{\dot{m}_{out} * [TS]_{digester}}$$

(B.6)

$$1 - SI = \frac{\dot{m}_{LD} * [TS]_{LD}}{\dot{m}_{out} * [TS]_{digester}} \quad \text{(B.7)}$$

It is known from previous works on sludge recirculation [43] that solid retention time corresponds to the following equation:

$$SRT = \frac{V * [TS]_{digester}}{\dot{m}_{LD} * [TS]_{LD} + (1 - R) * \dot{m}_{SD} * [TS]_{SD}}$$

(B.8)

Based on **Eq. (B.6)** and **Eq. (B.7)** we can write that:

$$\dot{m}_{SD} * [TS]_{SD} = SI * \dot{m}_{out} * [TS]_{digester} \quad \&$$

$$\dot{m}_{LD} * [TS]_{LD} = (1 - SI) * \dot{m}_{out} * [TS]_{digester}$$

It is therefore possible to express the denominator of the SRT equation as a function of $\dot{m}_{out}$ and to get rid of $[TS]_{digester}$.

$$SRT = \frac{V}{(1-R*SI)*\dot{m}_{out}}$$

(B.9)

Hence, combining **Eq. (B.1)** and **Eq. (B.5)** leads to the following equation:

$$\dot{m}_{feedstock} - \dot{m}_{biogas} = (1+\alpha - R)*\dot{m}_{SD}$$

And a combination of **Eq. (B.3)** and **Eq. (B.5)** can be expressed as:

$$\dot{m}_{out} = (1+\alpha)*\dot{m}_{SD}$$

Thus, $\dot{m}_{out}$ can be expressed as a function of $\dot{m}_{feedstock}$, $\dot{m}_{biogas}$, α and R:

$$\dot{m}_{out} = \frac{(1+\alpha)*(\dot{m}_{feedstock} - \dot{m}_{biogas})}{(1+\alpha-R)}$$

(B.10)

This new expression of $\dot{m}_{out}$ can be reinjected in **Eq. (A.9)**, leading to the following final equation:

$$SRT = \frac{(1+\alpha-R)}{(1+\alpha)*(1-R*SI)}\frac{V}{(\dot{m}_{feedstock} - \dot{m}_{biogas})}$$

(B.11)

α is a positive real number. The higher the value, the lower the quantity of SD produced. The SI index is comprised between 1/(1+ α) and 1. When SI is equal to 1/(1+ α), SD is not enriched in TS in comparison to LD and raw digestate is only separated in two flows (according to α). When SI is equal to 1 it means that the efficiency of the phase separation is maximal and that all total solids end up in the SD. α and SI values can be selected according to the average mass and total solids distribution profiles provided from a recent study on digestate mechanical separation [6]. In the case of a low efficiency separator (e.g. screw press), typical values for α and SI are 9 and 0.38 respectively. In the case of a high efficiency separator (e.g. centrifuges), typical values for α and SI are 2.45 and 0.81 respectively. R is comprised between 0 and 1. When R is equal to 0, no SD is recirculated and the SRT is equal to the HRT. When R is equal to 1, all produced SD is recirculated.

It is also possible to determine the impact of SD recirculation on digester TS content according to **Eq. (B.2)**. By using **Eq. (B.6)** and **Eq. (B.7)**, **Eq. (B.2)** can be transformed into:

$$\dot{m}_{feedstock} * [TS]_{feedstock} - \dot{m}_{biogas} = \dot{m}_{out} * [TS]_{digester} * (1 - R * SI)$$

Since the previous expression of $\dot{m}_{out}$ is expressed as a function of $\dot{m}_{feedstock}$, $\dot{m}_{biogas}$, α and R, the $[TS]_{digester}$ can be expressed according to the following final equation:

$$[TS]_{digester} = \frac{(1 + \alpha - R)}{(1 + \alpha) * (1 - R * SI)} * \frac{(\dot{m}_{feedstock} * [TS]_{feedstock} - \dot{m}_{biogas})}{(\dot{m}_{feedstock} - \dot{m}_{biogas})}$$

(B.12)

Generally, $[TS]_{digester}$ without recirculation (R=0) is a known value, therefore the equation obtained from above can be used to determine the $[TS]_{feedstock}$:

$$[TS]_{feedstock} = \frac{[TS]_{digester} * (\dot{m}_{feedstock} - \dot{m}_{biogas}) + \dot{m}_{biogas}}{\dot{m}_{feedstock}}$$

(B.13)

Finally, for more precision in SRT and $[TS]_{digester}$ calculations, $\dot{m}_{biogas}$ can be modified according to R. Indeed, when SD is recirculated, the additional produced biogas should be added to the initial biogas from feedstocks ($\dot{m}_{biogas}$). In this equation $\dot{m}_{biogas}(R)$ is expressed as a function of $\dot{m}_{SD\_recirculated}(R)$. This corresponds to a fraction (R) of the estimated amount of

SD produced per year (from biogas plant operator knowledge). $\dot{m}_{biogas}(R)$ is also expressed as a function of SD BMP (in Nm^3 CH_4/t VS), volatile solid contents, methane density as well as carbon dioxide density. Thus the following equation:

$$\dot{m}_{biogas}(R) = \dot{m}_{biogas}(0) + \frac{\dot{m}_{SD\_recirculated}(R) * [VS]_{SD} * BMP_{SD} * (d_{CH4} + \frac{(1-x_{CH4})}{x_{CH4}} * d_{CO2})}{1000}$$

B.14)

This can be reinjected in equations **Eq. (B.11)** and **Eq. (B.12)**, resulting in the two final equations presented in section 4.2.

$$\boldsymbol{SRT} = \frac{(1+\alpha-R)}{(1+\alpha)*(1-R*SI)} \frac{V}{(\dot{m}_{feedstock} - \dot{m}_{biogas}(R))}$$

(B.15)

$$[\boldsymbol{TS}]_{digester} = \frac{(1+\alpha-R)}{(1+\alpha)*(1-R*SI)} * \frac{(\dot{m}_{feedstock} * [TS]_{feedstock} - \dot{m}_{biogas}(R))}{(\dot{m}_{feedstock} - \dot{m}_{biogas}(R))}$$

(B.16)

Appendix C

After short-term aerobic post-treatments, the fractionation method, developed by Jimenez et al. [21], was performed on aerated SD from biogas plant B. However, instead of using COD tests to evaluate the distribution of VS in the different fractions, the TC content was measured to understand carbon distribution after the post-treatment. TC was measured on the four fractions (SPOM, SEOM, PEOM and NEOM) and on the raw sample. Besides, the fluorescence spectra of liquid extracts were recorded on a Perkin Elmer LS55 and a complexity ratio was calculated accordingly to Jimenez et al. [33]. This index is defined as the ratio of the sum of the fluorescence volumes of the most complex molecules (lignin, humic acid…) over the sum of the fluorescence volumes of the protein-like molecules.

The carbon distribution across the different fractions of plant B SD (aerated or not) is given in **Table C.** Distribution of carbon in the different fractions after aeration was significantly different between control SD and SD placed under strong aeration (SD 3). Carbon content in SPOM, PEOM and NEOM was reduced by 18%, 8% and 20%, respectively in comparison to the control. These lower carbon contents can be explained by the carbon respired during the post-treatment (10.5% of total initial carbon). Carbon loss was distributed among several fractions. It can be assumed that microbial endogenous activities under these aerobic conditions were not only ligninolytic but also proteolytic, cellulolytic and hemicellulolytic. SPOM fraction complexity index also increased, thus implying that soluble lignin-like molecules had been released. Finally, biodegradability (assessed by BMP_{FPTM} values and BMP of SD) decreased due to a lower quantity and higher complexity of the most accessible fraction (SPOM). This fraction and its complexity were found to strongly correlate with biodegradability and BMP in a previous study [44]. For SD placed under intermediate aeration (SD 2) carbon respired was also close to 10% of the total initial carbon. The carbon content in SPOM, SEOM and PEOM fell by 25%, 8% and 13.5% respectively, in comparison to the control. However, NEOM did not decrease significantly. For SD placed under low aeration (SD 1), only PEOM fell by 8%. It thus appears that for these airflows, significant degradation of carbon from the lignin-like fraction did not occur, while it was observed for the more accessible fractions. This explains the lower BMP. In the case of SD and taking these results into account, it is likely that under aerobic conditions, degradation of complex fractions may only take place once the easily degradable matter has begun to be degraded.

Table C: Short-term aerobic post-treatment on plant B SD: conditions and carbon distribution

Plant B SD sample	**Control SD**	**SD 1**	**SD 2**	**SD 3**
Aeration (L air/h/kg TS)	/	1.65	16.5	33
Duration (days)	/	5	5	5
% of total carbon loss/respired	0	3.1	8.6	10.5
% of total carbon in SPOM	2.8±0.2	2.6±0.1	2.1±0.1	2.3±0.1
% of total carbon in SEOM	10.2±0.1	10.2±0.1	9.4±0.3	10.2±0.1
% of total carbon in PEOM	53.2±1.4	46±1.8	46.1±0.1	49.1±0.6
% of total carbon in NEOM	32.6±3	33.6±5.8	28.4±1.6	26±1.4
% of total carbon to close balance	1.2±2	4.5±4.1	5.4±0.4	1.9±0.3
Complexity index of SPOM (in %)	130.5	156.8	161.1	153.6
Complexity index of SEOM (in %)	338.8	296.7	268.9	304

Appendix D

Table D: Features of principal mixer types for CSTR adapted from [39,45]

Type of mixer	**Upper TS content range**	**Approx. operation speed (rpm)**	**Installed max. power (kW)**
Submersible motor mixer with propeller	8%	500 – 1,500	35
Submersible motor mixer with large-blade rotor	12%	50 – 120	20
Central mixer	12%	12 – 18	25
Paddle mixer	14%	10	30
Shaft mixer	18%	40 – 50	11

References

[1] Weiland P. Biogas production: Current state and perspectives. Appl Microbiol Biotechnol 2010;85:849–60. https://doi.org/10.1007/s00253-009-2246-7.

[2] Scarlat N, Fahl F, Dallemand JF, Monforti F, Motola V. A spatial analysis of biogas potential from manure in Europe. Renew Sustain Energy Rev 2018;94:915–30. https://doi.org/10.1016/j.rser.2018.06.035.

[3] Wellinger A, Murphy JD, Baxter D. The biogas handbook Science, production and applications. 2013. https://doi.org/10.1533/9780857097415.

[4] Tambone F, Orzi V, D'Imporzano G, Adani F. Solid and liquid fractionation of digestate: Mass balance,

chemical characterization, and agronomic and environmental value. Bioresour Technol 2017;243:1251–6. https://doi.org/10.1016/j.biortech.2017.07.130.

[5] Paavola T, Rintala J. Effects of storage on characteristics and hygienic quality of digestates from four co-digestion concepts of manure and biowaste. Bioresour Technol 2008;99:7041–50. https://doi.org/10.1016/j.biortech.2008.01.005.

[6] Guilayn F, Jimenez J, Rouez M, Crest M, Patureau D. Digestate mechanical separation: Efficiency profiles based on anaerobic digestion feedstock and equipment choice. Bioresour Technol 2019;274:180–9. https://doi.org/10.1016/j.biortech.2018.11.090.

[7] Guilayn F, Jimenez J, Martel JL, Rouez M, Crest M, Patureau D. First fertilizing-value typology of digestates: A decision-making tool for regulation. Waste Manag 2019;86:67–79. https://doi.org/10.1016/j.wasman.2019.01.032.

[8] Thygesen O, Sommer SG, Shin SG, Triolo JM. Residual biochemical methane potential (BMP) of concentrated digestate from full-scale biogas plants. Fuel 2014;132:44–6. https://doi.org/10.1016/j.fuel.2014.04.062.

[9] Rincón CA, De Guardia A, Couvert A, Le Roux S, Soutrel I, Daumoin M, et al. Chemical and odor characterization of gas emissions released during composting of solid wastes and digestates. J Environ Manage 2019;233:39–53. https://doi.org/10.1016/j.jenvman.2018.12.009.

[10] Monlau F, Sambusiti C, Ficara E, Aboulkas A, Barakat A, Carrère H. New opportunities for agricultural digestate valorization: Current situation and perspectives. Energy Environ Sci 2015;8:2600–21. https://doi.org/10.1039/c5ee01633a.

[11] Sambusiti C, Monlau F, Barakat A. Bioethanol fermentation as alternative valorization route of agricultural digestate according to a biorefinery approach. Bioresour Technol 2016;212:289–95. https://doi.org/10.1016/j.biortech.2016.04.056.

[12] Monlau F, Sambusiti C, Antoniou N, Barakat A, Zabaniotou A. A new concept for enhancing energy recovery from agricultural residues by coupling anaerobic digestion and pyrolysis process. Appl Energy 2015;148:32–8. https://doi.org/10.1016/j.apenergy.2015.03.024.

[13] Funke A, Reebs F, Kruse A. Experimental comparison of hydrothermal and vapothermal carbonization. Fuel Process Technol 2013;115:261–9. https://doi.org/10.1016/j.fuproc.2013.04.020.

[14] Kratzeisen M, Starcevic N, Martinov M, Maurer C, Müller J. Applicability of biogas digestate as solid fuel. Fuel 2010;89:2544–8. https://doi.org/10.1016/j.fuel.2010.02.008.

[15] Menardo S, Balsari P, Dinuccio E, Gioelli F. Thermal pre-treatment of solid fraction from mechanically-separated raw and digested slurry to increase methane yield. Bioresour Technol 2011;102:2026–32. https://doi.org/10.1016/j.biortech.2010.09.067.

[16] Lindner J, Zielonka S, Oechsner H, Lemmer A. Effects of mechanical treatment of digestate after anaerobic digestion on the degree of degradation. Bioresour Technol 2015;178:194–200. https://doi.org/10.1016/j.biortech.2014.09.117.

[17] Sambusiti C, Monlau F, Ficara E, Musatti A, Rollini M, Barakat A, et al. Comparison of various post-treatments for recovering methane from agricultural digestate. Fuel Process Technol 2015;137:359–65. https://doi.org/10.1016/j.fuproc.2015.04.028.

[18] Brémond U, Bertrandias A, Steyer J, Bernet N, Carrere H. A vision of European biogas sector development towards 2030 : Trends and challenges. J Clean Prod 2020. https://doi.org/10.1016/j.jclepro.2020.125065.

[19] Couturier C. Techniques de production d'électricité à partir de biogaz et de gaz de synthèse. 2009.

[20] APHA. Standard methods for the examination of water and wastewater. American Public Health

Association, Washington, DC. Am Public Heal Assoc Washington, DC 2005.

[21] Jimenez J, Aemig Q, Doussiet N, Steyer JP, Houot S, Patureau D. A new organic matter fractionation methodology for organic wastes: Bioaccessibility and complexity characterization for treatment optimization. Bioresour Technol 2015;194:344–53. https://doi.org/10.1016/j.biortech.2015.07.037.

[22] Brémond U, Bertrandias A, Loisel D, Jimenez J, Steyer J-P, Bernet N, et al. Assessment of fungal and thermo-alkaline post-treatments of solid digestate in a recirculation scheme to increase flexibility in feedstocks supply management of biogas plants. Renew Energy 2020;149. https://doi.org/10.1016/j.renene.2019.12.062.

[23] Holliger C, de Laclos HF, Hack G. Methane production of full-scale anaerobic digestion plants calculated from substrate’s biomethane potentials compares well with the one measured on-site. Front Energy Res 2017;5:1–9. https://doi.org/10.3389/fenrg.2017.00012.

[24] Maynaud G, Druilhe C, Daumoin M, Jimenez J, Patureau D, Torrijos M, et al. Characterisation of the biodegradability of post-treated digestates via the chemical accessibility and complexity of organic matter. Bioresour Technol 2017;231:65–74. https://doi.org/10.1016/j.biortech.2017.01.057.

[25] Atee Club Biogaz. Calculateur - Injection du Biogaz dans le réseau 2019.

[26] Wang X, Lu X, Li F, Yang G. Effects of temperature and Carbon-Nitrogen (C/N) ratio on the performance of anaerobic co-digestion of dairy manure, chicken manure and rice straw: Focusing on ammonia inhibition. PLoS One 2014;9:1–7. https://doi.org/10.1371/journal.pone.0097265.

[27] Jurado E, Skiadas I V., Gavala HN. Enhanced methane productivity from manure fibers by aqueous ammonia soaking pretreatment. Appl Energy 2013;109:104–11. https://doi.org/10.1016/j.apenergy.2013.03.075.

[28] Lin L, Xu F, Ge X, Li Y. Improving the sustainability of organic waste management practices in the food-energy-water nexus: A comparative review of anaerobic digestion and composting. Renew Sustain Energy Rev 2018;89:151–67. https://doi.org/10.1016/j.rser.2018.03.025.

[29] Brémond U, de Buyer R, Steyer JP, Bernet N, Carrere H. Biological pretreatments of biomass for improving biogas production: an overview from lab scale to full-scale. Renew Sustain Energy Rev 2018;90:583–604. https://doi.org/10.1016/j.rser.2018.03.103.

[30] Chambre d’agriculture des Landes. Essais couverts végétaux 2018:47–65.

[31] Muha I, Linke B, Wittum G. A dynamic model for calculating methane emissions from digestate based on co-digestion of animal manure and biogas crops in full scale German biogas plants. Bioresour Technol 2015;178:350–8. https://doi.org/10.1016/j.biortech.2014.08.060.

[32] Schneider N. Density and Viscosity of Biomass from Agricultural Biogas Plants 2018:1–229.

[33] Hreiz R, Adouani N, Fünfschilling D, Marchal P, Pons MN. Rheological characterization of raw and anaerobically digested cow slurry. Chem Eng Res Des 2017;119:47–57. https://doi.org/10.1016/j.cherd.2017.01.005.

[34] Mönch-Tegeder M, Lemmer A, Hinrichs J, Oechsner H. Development of an in-line process viscometer for the full-scale biogas process. Bioresour Technol 2015;178:278–84. https://doi.org/10.1016/j.biortech.2014.08.041.

[35] Mbaye S, Dieudé-Fauvel E, Baudez JC. Comparative analysis of anaerobically digested wastes flow properties. Waste Manag 2014;34:2057–62. https://doi.org/10.1016/j.wasman.2014.06.021.

[36] Björn A, Šafarič L, Karlsson A, Danielsson A, Ejlertsson J, Svensson BH, et al. Substrate and operational conditions as regulators of fluid properties in full-scale continuous stirred-tank biogas reactors - implications for rheology-driven power requirements. Water Sci Technol 2018;78:814–26. https://doi.org/10.2166/wst.2018.352.

[37] Liu Y, Chen J, Song J, Hai Z, Lu X, Ji X, et al. Adjusting the rheological properties of corn-straw slurry to reduce the agitation power consumption in anaerobic digestion. Bioresour Technol 2019;272:360–9. https://doi.org/10.1016/j.biortech.2018.10.050.

[38] Naegele HJ, Lemmer A, Oechsner H, Jungbluth T. Electric energy consumption of the full scale research biogas plant “unterer lindenhof”: Results of longterm and full detail measurements. Energies 2012;5:5198–214. https://doi.org/10.3390/en5125198.

[39] Singh B, Szamosi Z, Siménfalvi Z. State of the art on mixing in an anaerobic digester: A review. Renew Energy 2019;141:922–36. https://doi.org/10.1016/j.renene.2019.04.072.

[40] Wu S, Ni P, Li J, Sun H, Wang Y, Luo H, et al. Integrated approach to sustain biogas production in anaerobic digestion of chicken manure under recycled utilization of liquid digestate: Dynamics of ammonium accumulation and mitigation control. Bioresour Technol 2016;205:75–81. https://doi.org/10.1016/j.biortech.2016.01.021.

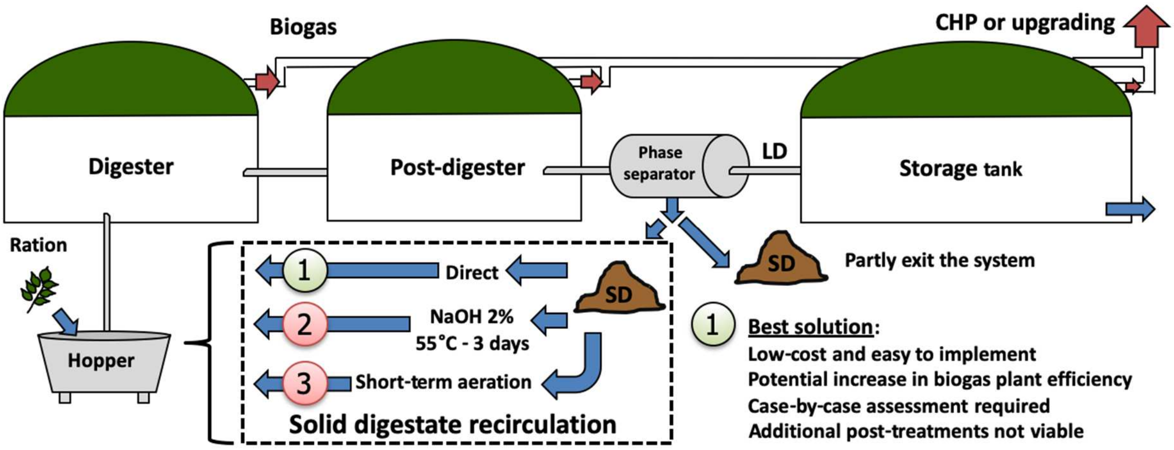
[41] Ni Z, Liu J, Zhang M. Short-term pre-aeration applied to the dry anaerobic digestion of MSW, with a focus on the spectroscopic characteristics of dissolved organic matter. Chem Eng J 2017;313:1222–32. https://doi.org/10.1016/j.cej.2016.11.020.

[42] Wagner AO, Janetschek J, Illmer P. Using Digestate Compost as a Substrate for Anaerobic Digestion. Chem Eng Technol 2018;41:747–54. https://doi.org/10.1002/ceat.201700386.

[43] Degremont. Water Treatment Handbook. 7th ed. 2007.

[44] Jimenez J, Lei H, Steyer JP, Houot S, Patureau D. Methane production and fertilizing value of organic waste: Organic matter characterization for a better prediction of valorization pathways. Bioresour Technol 2017;241:1012–21. https://doi.org/10.1016/j.biortech.2017.05.176.

[45] FNR. Guide to Biogas: From production to use. Fachagentur Nachwachsende Rohstoffe e V FNR, 2012:232.



Biogas
CHP or upgrading
Digester
Post-digester
Phase separator
LD
Storage tank
Ration
Hopper
1
Direct
SD
2
NaOH 2%
55°C - 3 days
3
Short-term aeration
Solid digestate recirculation
SD
Partly exit the system
1
Best solution:
Low-cost and easy to implement
Potential increase in biogas plant efficiency
Case-by-case assessment required
Additional post-treatments not viable