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

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23 production could rise by 0.6 to 6.3% or that a potential feedstock shortage of 64 to 1431
24 tons/year could be compensated. Secondly, the relevance of additional post-treatments for
25 improving these initial results was evaluated. Thermo-chemical post-treatments successfully
26 increased SD biodegradability by 30 to 46% although their costs were not compensated by
27 additional methane production. Short-term aerobic post-treatments failed in increasing SD
28 biodegradability (up to 21% loss in biomethane potential). Hence, at full scale, a quick and
29 direct recirculation of SD excluding any post-treatment appears to be the optimal condition
30 to apply. Finally, conditions for the full-scale implementation of direct SD recirculation were
31 theoretically studied. This practice has proved to increase the solid retention time by 11 to
32 38% and the plant total solid content by 6 to 20%. Thus, the critical point for its
33 implementation should be the capacity of the plant mixing system to handle such an
34 increase in solids. The relevance of SD recirculation needs to be determined on a case-by-
35 case basis. Consequently, for the first time, this article provides a framework where the
36 conditions can be identified for direct SD recirculation to be a relevant digestate
37 management practice. Overall, this article demonstrates how direct SD recirculation can be a
38 simple and low-cost mean for improving agricultural CSTR biogas plant efficiency. It also
39 highlights the importance of achieving efficient digestate management in the biogas sector
40 in order to reduce the costs of biogas production.

41 1. Introduction

42 One of the main challenges of the 21st century is to decorrelate growing worldwide energy
43 consumption from the use of fossil fuels. Their substitution with low-carbon/renewable
44 options is crucial for reducing greenhouse gas (GHG) emissions and mitigating global
45 warming. Among these renewable options, anaerobic digestion (AD) displays several
46 advantages since it processes low-cost organic waste via a robust and naturally occurring

47 biological process to produce biogas. It is a versatile energy carrier that can be used for
48 generating power and heat, injected into the gas grid or used as a fuel for vehicles [1]. In
49 Europe, incentive policies have led to the emergence of a biogas industry, mainly based on
50 agricultural feedstocks (agricultural residues, energy crops, catch crops) which represent the
51 largest available deposit of organic matter. Biomethane potential from agricultural
52 feedstocks was estimated at 58.9 billion Nm³, three times higher than potential from other
53 organic wastes, such as food waste or sewage sludge [2].

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

69 For SD, the most widespread storage method implies composting before use as a soil
70 amendment. However, according to certain studies: (i) SD can have a biomethane potential
71 as high as $240 \text{ Nm}^3 \text{ CH}_4 \cdot \text{ton}^{-1}$ volatile solids (VS) [8]; (ii) strong ammonia emissions occur
72 during the composting treatment of agricultural SD since significant amounts of $\text{NH}_3/\text{NH}_4^+$
73 contained in SD are volatilized, while carbon is emitted as CO_2 [9]. To avoid this loss of
74 organic matter during composting, three main strategies have been identified to recover
75 more energy from SD [10]: (i) fermentation of SD to produce bioethanol [11]; (ii) thermal
76 conversion processes of SD such as pyrolysis that produce syngas, biochar and bio-oil [12],
77 hydrothermal and vapothermal carbonisation that produce mainly hydrochar [13] or
78 combustion that provides heat [14]; (iii) recirculation of SD within the biogas plant to recover
79 the residual methane potential [8].

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

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

92 176% increase in SD BMP has been observed for thermal treatment [15], milling [16] and
93 enzymatic treatment [17]. However, in all of these studies, the economic impact of
94 recirculation for agricultural biogas plants and possible post-treatment costs were rarely
95 discussed. Sambusiti *et al.* estimated that direct recirculation of SD could lead to a 14%
96 increase in economic profits, although this was based on very optimistic assumptions: a high
97 solid separation efficiency of the screw press (73%) and a recirculation of the total amount
98 of SD produced [17]. Solid separation efficiency indexes of a screw press vary around 30% for
99 total solids (TS) [6]. Finally, only a partial amount of SD can be recirculated as some solids
100 should exit the system, notably to eliminate the inorganic matter that would otherwise
101 accumulate and cause inhibitions (e.g. heavy metals). Realistic hypotheses need to be
102 applied in order to fully evaluate the economic relevance of such post-treatment practices in
103 a recirculation scheme.

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

- 110 (i) Which strategies and conditions are relevant for the application of SD recirculation?
111 (ii) Are additional post-treatments prior to SD recirculation economically viable?
112 (iii) What are the impacts of SD recirculation on plant methane production, solid retention
113 time and digester TS content?

114

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

122

123 2. Materials and methods

124 2.1. The different strategies for SD recirculation

125 It is important to define the three possible implementation strategies for SD recirculation.

126 These are represented in **Figure 1**:

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

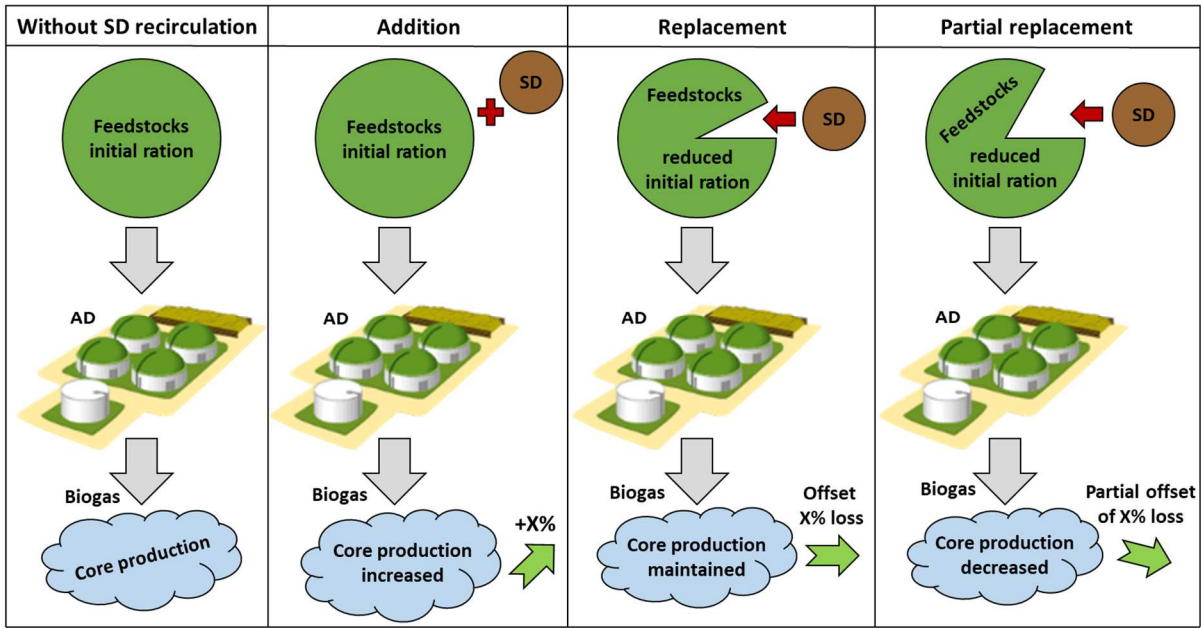
129 (ii) SD can also be integrated to the ration and replace certain feedstocks that are
130 lacking (shortage/voluntarily non-produced/further stored, in the case of a future
131 anticipated shortage). In this second case, SD recirculation would allow to totally
132 offset ration reduction and maintain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

136 (iii) Finally, SD can also be integrated to the ration and replace certain feedstocks that
137 are lacking although the amount of recirculated SD would not allow for a total

138 offset of ration reduction or maintain a steady biogas production over time. In
 139 this case, it would simply limit loss in biogas production. This could be either due
 140 to a too high quantity of lacking feedstock that cannot be offset even when all
 141 yearly produced SD is recirculated, either due to a quantity of recirculated SD that
 142 is lower than the quantity of SD recirculated to reach the replacement strategy
 143 ($\dot{m}_{SD_replacement}$ in tons per year). In this latter case the quantity of recirculated SD
 144 ($\dot{m}_{SD_partial_replacement}$ in tons per year) would vary within the following range:

$$0 < \dot{m}_{SD_partial_replacement} < \dot{m}_{SD_replacement}$$

146 This strategy will be further called the “partial replacement strategy”



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

149 In the present paper focus is mainly put on the addition and replacement strategies for cases
 150 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 ($\dot{m}_{\text{feedstock}}$ in kg or t/year), the HRT before phase separation, the pH, the temperature, the average percentage of methane in biogas (x_{CH_4} 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 ($Q_{\text{bioCH}_4\text{,hour}}$ in Nm^3 biomethane/hour) or combined heat and power valorisation (CHP) engine nominal power ($\text{CHP}_{\text{power}}$ in kW) were provided by the plant operators. In all calculations, the number of days of operation during the year ($D_{\text{operation}}$ 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.

172 The loading rate (LR in kg feedstock/m³/day) was obtained using the quantity of feedstocks
 173 used per year ($\dot{m}_{feedstock}$ in kg/year) and the digester reactor volume ($RV_{Digester}$ in m³)
 174 according to the following equation:

$$175 \quad LR = \frac{\dot{m}_{feedstock}}{RV_{Digester} * D_{operation}} \quad (1)$$

176 In absence of biogas flowmeters on plants, calculation of the plant biogas production
 177 (V_{biogas_year} in Nm³/year) was different as a function of the valorisation type. In the case
 178 where upgrading and biomethane injection occurred, all produced methane was assumed to
 179 be injected and the following equation was used.

$$180 \quad V_{biogas_year} = \frac{Q_{bioCH_4_hour} * D_{operation} * 24}{x_{CH_4}} \quad (2)$$

181 In the case of CHP, several hypotheses were made: (i) The CHP engines were sized to have an
 182 optimal efficiency, therefore only 85% of the CHP engine nominal power was used; (ii) The
 183 electric efficiency of the CHP system was 40% as usually reported in the literature [17]; (iii)
 184 Five percent of additional energetic loss to avoid under-supply of the CHP system [19]; (iv) A
 185 lower heating value (LHV) of 9.94 kWh/Nm³ was used for methane; (v) all biogas produced
 186 was sent to the CHP system. Total electrical energy produced per year (E_{elec} in kWh_{el}/year)
 187 and plant biogas production (V_{biogas_year} in Nm³/year) were calculated using these two
 188 equations:

$$189 \quad E_{elec} = CHP_{power} * 0.85 * D_{operation} * 24 * 0.4 \quad (3)$$

$$190 \quad V_{biogas_year} = \frac{CHP_{power} * 0.85 * D_{operation} * 24}{0.95 * LHV * x_{CH_4}} \quad (4)$$

191 It is noteworthy that calculated values are close to the approximate figures provided by
 192 plant operators. Finally, the average methane yield from feedstock (Y_{CH_4} in Nm³ CH₄/t

193 feedstock) was calculated from the volume of methane produced by the plant each year
194 ($V_{CH_4_year}$ in Nm³ biomethane/year by multiplying V_{biogas_year} with x_{CH_4}):

$$195 \quad Y_{CH_4} = \frac{V_{CH_4_year}}{\dot{m}_{feedstock}} \quad (5)$$

196 For each biogas plant, given and calculated features in this section, are displayed in **Table 1**.

197 For reasons of confidentiality, a letter was attributed to each plant.

198

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 (m³)	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/m³/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 (Nm³/hour)	145	/	140	160	/
CHP engine nominal power (kW)	/	1890	/	/	1090
Plant biogas production (Nm³/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 (Nm³/year)	1,189,000	1,395,000	1,148,000	1,312,000	805,000
Methane yield (Nm³ CH₄/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

199 2.3. Solid digestate characterisation

200 Several methods were used to characterise the different SD. The TS and VS contents were
201 obtained following the standard methods of the American Public Health Association [20].
202 Total carbon (TC) content was measured using a Shimadzu TOC-VCSN Analyser coupled to a
203 Shimadzu solid sample module SSM-5000A. An AutoKjeldahl Unit K-370, BUCHI was used to
204 determine the total Kjeldahl nitrogen. Aqualytic 420721 COD Vario Tube Test MR (0-1.5
205 g·L⁻¹) were used to get the chemical oxygen demand (COD). For liquids, 2 ml of sample
206 adequately diluted were pipetted into each tube and oxidation reactions in tubes were
207 performed in a HACH COD reactor at 150 C for 2 hours. Finally, COD concentrations were
208 measured using an Aqualytic MultiDirect spectrophotometer. For solids, in duplicate, 0.25 g
209 of samples were poured into 10 ml of 98% H₂SO₄ and set under strong agitation overnight to
210 solubilize solid particles. Dilution with Milli-Q water up to 250 ml allowed pipetting. The
211 obtained liquid was used as previously in COD tubes.

212 SD matter was characterised via sequential chemical extractions according to the protocol
213 developed by Jimenez et al. [21]. Samples were freeze-dried, then grounded at 1 mm using a
214 Fritsch Pulverisette 19 and associated stainless steel sieve. Subsequent sequential
215 extractions were performed using 0.5 g of samples. Three fractions of decreasing
216 accessibility were then obtained by applying increasingly strong chemical solutions: (i)
217 soluble OM (SPOM) using CaCl₂ (10 mM) that corresponds to soluble sugars and proteins; (ii)
218 slowly extractable OM (SEOM) using NaOH (0.1 M), which corresponds to remaining proteins
219 and sugars, some humic substances and lipids; (iii) poorly extractable OM (PEOM) using
220 H₂SO₄ 72% w/w that corresponds to hemicellulose and cellulose. At each step, the
221 solubilized VS was recovered in the supernatant by centrifugation (18,750 g for 20 min at 4
222 °C) and filtered at 0.45 µm. VS of each fraction were then characterized via COD

223 measurement as described before. Finally, the non-extractable OM (NEOM) was calculated
224 by subtracting the four fractions of VS extracted from the sample from the total VS. Total VS
225 were obtained by measuring COD on SD solid sample powder as previously described. As VS
226 correspond to organic matter, the former term will only be used thereafter.

227 BMP tests were performed on each SD. SD were anaerobically digested in batch mode at
228 35°C under agitation. Bottles with a working volume of 400 mL, were used. Bottles were
229 filled with: 4mL of trace elements solution (FeCl₂, 2 g·L⁻¹; CoCl₂, 0.5 g·L⁻¹; MnCl₂, 0.1 g·L⁻¹;
230 NiCl₂, 0.1 g·L⁻¹; ZnCl₂, 0.05 g·L⁻¹; H₃BO₃, 0.05 g·L⁻¹; Na₂SeO₃, 0.05 g·L⁻¹; CuCl₂, 0.04 g·L⁻¹;
231 Na₂MoO₄, 0.01 g·L⁻¹), 8.6 ml of macroelement solution (NH₄Cl, 26.6 g·L⁻¹; KH₂PO₄, 10 g·L⁻¹;
232 MgCl₂, 6 g·L⁻¹; CaCl₂, 3 g·L⁻¹), 20.8 mL of bicarbonate buffer (NaHCO₃, 50 g·L⁻¹), an inoculum at
233 5 g of VS·L⁻¹, the SD at 5 g of VS·L⁻¹ and potentially with distilled water to complete to 400
234 mL. Anaerobic conditions were then obtained by flushing the headspace with nitrogen gas
235 and closing bottles with red butyl rubber septum-type stoppers. A control was made by
236 preparing the same mixture with the inoculum but without the substrate to evaluate the
237 endogenous methane production. The inoculum was the same for all tests; a granular sludge
238 coming from a mesophilic anaerobic digester of a sugar refinery was mixed with water and
239 crushed using a kitchen immersion hand blender. Then, a 25-mesh sieve was used to remove
240 most of the remaining solid/minerals and to obtain a homogenous liquid inoculum.
241 According to standardized practices, triplicates were launched and incubated under agitation
242 at 35°C. Biogas production was frequently monitored by measurement of the headspace
243 pressure and its composition using a PerkinElmer® Clarus 580 gas chromatograph as
244 described in a previous study [22]. Endogenous methane production was evaluated by
245 subtracting the gas produced in the controls and BMP tests were stopped when the daily

246 methane production during three consecutive days was <1% of the accumulated volume of
247 methane [23]. All volumes are expressed in normal conditions.

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

$$251 \quad BD_{ana} = \frac{BMP * 100}{0.35 * COD_{tot}} \quad (6)$$

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

254 2.4. Thermo-alkaline post-treatment

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

261 2.5. Short-term aerobic post-treatment

262 Short-term aerobic post-treatment was applied to SD from plants A and B. 300 grams of SD
263 were placed inside 2.5 L bioreactors and aerated according to the set-up displayed in
264 **Appendix A**. SD was contained in a fine polyester net to avoid loss of material. A calibrated
265 peristaltic pump sent air into the system. Airflow first entered a concentrated NaOH (2M)
266 solution to trap atmospheric CO₂. The air was then moisturized by bubbling into water at the
267 bottom of the double jacket 2.5L bioreactor. For all experiments, temperature in the aerobic

268 reactors was set at 30°C. Finally, carbon loss due to respiration was evaluated by trapping
269 the emitted CO₂ in a 0.5 M soda solution set at 30°C, of which conductivity was measured
270 and recorded every five minutes using an internally developed acquisition system. The
271 relationship between conductivity (mS·cm⁻¹) and trapped carbon (mg C-CO₂·L⁻¹) was
272 determined according to a calibration line obtained by measuring conductivity of the soda
273 solution with different quantities of Na₂CO₃ dissolved inside and 0.273 as a correction factor
274 to translate the mg CO₂·L⁻¹ trapped into mg C-CO₂·L⁻¹. The total amount of carbon in the SD
275 placed in the bioreactor was calculated from the mass of SD, its TS and the average TC
276 content. From this set-up, several short durations and low to strong airflow were tested at
277 30°C. For plant A, 1.75, 3.6 and 6 days with an airflow of 1.5, 10 and 20 L·h⁻¹·kg⁻¹ of TS of SD
278 were tested. For plant B, 2.5 and 5 days with an airflow of 1.65, 16.5 and 33 L·h⁻¹·kg⁻¹ of TS of
279 SD were applied.

280 Samples from aerated SD were also used as substrates for additional BMP tests. These tests
281 involved aerated SD, the corresponding untreated SD (blank) and the control without SD for
282 endogenous production. For aerated SD, BMP of final post-treated matter (BMP_{FPTM} in
283 mL CH₄·g⁻¹ VS final) was defined as the methane directly measured and produced by the
284 remaining matter without any respiration correction. It can be assimilated to
285 biodegradability of post-treated SD. To determine their BMP, carbon loss was taken into
286 account. It was obtained from raw BMP values corrected according to the hypothesis that all
287 carbon losses as CO₂ during the post-treatment would otherwise have produced biogas
288 (ratio 60% CH₄/40% CO₂).

289 2.6. Impact of SD recirculation on plant methane production

290 The impact of SD recirculation on plant methane production (gain or offset) is evaluated as a
291 percentage of the total plant methane production. This percentage ($\%_{\text{total_plant_CH}_4}$) is
292 obtained by dividing the volume of methane produced from recirculation ($V_{\text{CH}_4_SD}$) by the
293 plant methane production ($V_{\text{CH}_4_year}$). $V_{\text{CH}_4_SD}$ is calculated using the measured BMP from SD,
294 the quantity of SD recirculated each year ($\dot{m}_{SD_recirculated}(R)$) and the %VS of SD ($[VS]_{SD}$). For
295 BMP values, in the case of SD from plants A, B and C, the considered value was the average
296 of values obtained during the three different BMP tests (control values in the case of each
297 post-treatment). As an example, in the case of plant A, BMP for each control were 130, 155
298 and 153 mL $\text{CH}_4 \cdot \text{g}^{-1}$ VS, respectively. The considered value was therefore their average: 146
299 mL $\text{CH}_4 \cdot \text{g}^{-1}$ VS. For SD from plants C, D and E, the BMP values were only obtained from one
300 set of trials (control for simple BMP; no post-treatment tested on these SD). Finally, a 0.8
301 correction factor was applied to all BMP values from plants A, B, C, D, E and F, in order to
302 simulate a recirculation process at full scale rather than lab scale and to assess realistic
303 values [23]. These new values are called corrected SD BMP ($\text{BMP}_{\text{corrected_SD}}$). Besides, it is
304 important to underline that all produced SD cannot be recirculated. Indeed, certain types of
305 recalcitrant organic matter as well as minerals must exit the system to avoid AD inhibitions
306 and be spread on land to recycle carbon and nutrients. Calculations were performed on
307 three amounts of SD that can be recirculated within the biogas plant. They correspond to
308 30%, 50% and 80% of the total amount of SD produced each year and appear to be realistic
309 in the case of a full-scale application. Therefore, $\dot{m}_{SD_recirculated}$ varies according to these
310 percentages. All these values were used to calculate the impact of SD recirculation on plant
311 methane production:

$$312 \quad \%_{total_plant_CH4} = \frac{V_{CH4_SD}}{V_{CH4_year}} = \frac{BMP_{corrected_SD} * \dot{m}_{SD_recirculated} * [VS]_{SD}}{V_{CH4_year}} \quad (7)$$

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

317 As all plants are in France, FIT for injected biomethane and electricity, set by the French
 318 government, were obtained by using the calculator provided by the Association Technique
 319 Energie Environnement (ATEE) Club biogaz [25]. Tariffs are given in **Table 1**. For all plants,
 320 the purchase agreement of biomethane or electricity was assumed to be signed on the 1st
 321 January 2019. For biomethane injection, a higher heating value (HHV) of biomethane was set
 322 at 10.8 kWh/Nm³ and bonuses for the presence of catch crops, sludge or municipal waste
 323 inside the feedstock mix were taken into account to obtain the feed-in tariff. In the case of
 324 electricity, similarly bonuses due to the use of livestock manure were considered.

325 In French biogas legislation, as a standard rule, methane energy equivalence is calculated
 326 with the HHV coefficient in the case of biomethane injection, while the lower heating value
 327 coefficient is used for CHP valorisation. According to this rule, additional economic profits
 328 were obtained using two formulas according to the type of valorisation. For upgrading
 329 purposes, the HHV value (kWh/Nm³), the additional amount of produced methane
 330 ($V_{additional_CH4}$ in Nm³ CH₄/year) and the corresponding feed-in tariff for the biogas plant
 331 ($FIT_{biomethane}$ in €/MWh) were used as displayed in **Eq. 8**. For CHP, the LHV (9.94 kWh/Nm³),
 332 the electrical conversion efficiency set to 40% and 5% of additional energetic loss to avoid
 333 the under-supply of the CHP system and the corresponding feed-in tariff for the biogas plant
 334 ($FIT_{electrical}$ in €/MWh) were used as displayed in **Eq. 9**.

$$335 \quad AEG_{biomethane} = \frac{V_{CH_4,SD} * HHV * FIT_{biomethane}}{1000} \quad (8)$$

$$336 \quad AEG_{electrical} = \frac{V_{CH_4,SD} * LHV * 0.95 * 0.4 * FIT_{electrical}}{1000} \quad (9)$$

337

338 In the case of a recirculation replacement strategy, this percentage of the total plant
 339 methane production should allow to calculate the tons of feedstocks ($\Delta_{feedstock}$) that could be
 340 replaced by the amount of recirculated SD ($\dot{m}_{SD_recirculated}$) per year. This amount of feedstock
 341 can be calculated using the methane yield (Y_{CH_4} – obtained via **Eq. 5**) and according to the
 342 following equation:

$$343 \quad \Delta_{feedstock} = \frac{V_{CH_4,SD}}{Y_{CH_4}} \Leftrightarrow \frac{[VS]_{SD} * BMP_{SD} * \dot{m}_{SD} * R}{Y_{CH_4}} \quad (10)$$

344 2.7. Impact of SD recirculation on CSTR solid retention time and TS content

345 For agricultural CSTR biogas plants, hydraulic retention time (HRT in days) represents the
 346 retention time of liquids in the system and can be calculated in two different manners. The
 347 most common and simplest way uses digester and post-digester volumes ($RV_{digester}$ and
 348 $RV_{post-digester}$ in m^3) as well as the feedstock intake ($\dot{m}_{feedstock}$ in t/year divided by the days of
 349 operation during the year $D_{operation}$ in days/year) to calculate HRT ($HRT_{Classical}$ in days)
 350 according to the **Eq. 11**. It is generally assumed that feedstock densities are equal to one,
 351 therefore tons can be converted to cubic meters.

$$352 \quad HRT_{Classical} = \frac{RV_{digester} + RV_{post-digester}}{\frac{\dot{m}_{feedstock}}{D_{operation}}} \quad (11)$$

353 HRT displayed in **Table 1** are calculated in this way and are similar to values provided by
 354 operators. However, with this approach, calculations do not take into account the gas
 355 production from the feedstock, thus leading to inaccurate results. However, in these biogas

356 plants, the amount of biogas produced (V_{biogas_year} in $Nm^3/year$) as well as its average
 357 composition is known (x_{CH_4} defined as the methane percentage in the biogas). The mass of
 358 biogas produced (\dot{m}_{biogas} in t/year) can therefore be approximated using average methane
 359 and carbon dioxide densities (d_{CH_4} - 0.67 and d_{CO_2} - 1.87 kg/ Nm^3 at 15°C and atmospheric
 360 pressure), according to the following equation:

$$361 \quad \dot{m}_{biogas} = \frac{V_{biogas_year} * x_{CH_4} * d_{CH_4} + V_{biogas} * (1 - x_{CH_4}) * d_{CO_2}}{1000} \quad (12)$$

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

$$366 \quad HRT_{accurate} = \frac{RV_{digester} + RV_{post-digester}}{\frac{\dot{m}_{feedstock} - \dot{m}_{biogas}}{D_{operation}}} \quad (13)$$

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

370 2.7.1. Direct SD recirculation - Addition strategy

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

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

379 (14)

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

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

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

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

393 In the rest of this article, SI and α will be constant but further details concerning their range
 394 of validity for the general formula can be found in **Appendix B**. Since the study concerns
 395 agricultural biogas plants using screw presses that are low-efficiency phase separators, the
 396 average mass distribution profiles provided from a recent study on digestate mechanical

397 separation [6] can be used. Hence, SI can be set to 38% and α to 9 (90% of flow weight goes
 398 to the LD and 10% to the SD). Previous equations can thus be simplified as follows:

$$399 \quad SRT = \frac{(1 - \frac{R}{10})}{(1 - 0.38 * R)} * HRT_{accurate}(R) \quad (18)$$

$$400 \quad [TS]_{digester} = \frac{(1 - \frac{R}{10})}{(1 - 0.38 * R)} * \frac{(\dot{m}_{feedstock} * [TS]_{feedstock} - \dot{m}_{biogas}(R))}{(\dot{m}_{feedstock} - \dot{m}_{biogas}(R))} \quad (19)$$

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

405 2.7.2. Direct SD recirculation - Replacement strategy

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

$$415 \quad \dot{m}1_{feedstock} = \dot{m}_{feedstock} - \Delta_{feedstock}(R) \quad (20)$$

416

417 3. Results and discussion

418 3.1. Characterization and classification of solid digestates

419 **Table 2** summarizes the physico-chemical and biological properties of the five SD. TS were
 420 comprised between 22.5% and 25.7% and their VS between 82.5% and 87.3% of TS. It is in
 421 the range of values found in literature for SD coming from a screw press comprised between
 422 20 and 30% for TS and 80-90% of TS for VS [24]. Average C/N ratio for the five SD is 21,
 423 slightly lower than the optimal C/N ratio for AD often found between 25 and 30 [26].

424 **Table 2: Solid digestate physico-chemical and biological properties. Main molecules**
 425 **present in the different OM fractions: SPOM – Soluble sugars & proteins; SEOM – Proteins;**
 426 **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

427
 428 BMP values are in accordance with values found in the literature for SD (60 to 240 Nm³ CH₄/t
 429 VS) [10]. High BMP (> 145 Nm³ CH₄/t VS) and anaerobic biodegradability (32-45% COD_{tot}),
 430 that were obtained for plants B, D and E, can be explained by the low to intermediate HRT of
 431 these plants (84, 88 and 40 days respectively). Intermediate to low BMP values were
 432 obtained for plants A and C (99-130 Nm³ CH₄/t VS) leading to a lower anaerobic
 433 biodegradability (< 30% COD_{tot}). For plant A and C, whey, catch crop and beet pulp are the
 434 main feedstocks. They are easy to degrade, and applied HRT of 100 and 130 days,

435 respectively, allow for most of the biogas from the feedstocks to be recovered, thus lowering
436 the BMP value.

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

447 Decision was made to: (i) further work on SD from plants A and C as they are representative
448 of the agricultural CSTR biogas plant sector with long HRT, mainly easy to degrade feedstocks
449 (> 40% total ration) and SD with high lignin-like content; (ii) eliminate among the last
450 subgroup SD from plants D and E since they have too short HRT compared to the amount of
451 feedstock they treat each year and to their mix (manure rich); (iii) also select SD from plant B
452 as it displays an interesting profile with intermediate BMP and high hollocellulose content
453 that is due, despite a long HRT and thermophilic conditions, to the high amount of manure
454 used as feedstock (60%). The application of post-treatments on SD from plants A, B and C
455 aims to increase their BMP. This can be obtained notably by a specific degradation of the
456 lignin-like fraction that enhances the bioaccessibility of the organic matter.

457 3.2. Effect of thermo-alkaline and short-term aerobic post-treatments on solid
 458 digestate biomethane yield

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

465 **Table 3: Effect of thermo-alkaline and short-term aerobic post-treatment on SD on the**
 466 **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%

467
 468 In a study on the effect of composting storage on BMP SD, Menardo *et al.* showed that
 469 composting between two and four weeks led to an approximate 30 % decrease in BMP [15].
 470 During the composting process, mesophilic and thermophilic phases favour the removal of
 471 easily degradable matter, cellulose and hemicellulose. Lignin components are mainly
 472 degraded during the maturation phase [28]. SD VS evolution was not studied by Menardo *et*

473 *al.*, but their results seem to indicate that over a medium-term, easily biodegradable
474 fractions are better degraded than complex ones, thus explaining the lower BMP.

475 In this study, the effect of shorter and controlled aeration on SD was evaluated for its
476 potential as a low-cost biological post-treatment practice at full scale [29]. It was
477 hypothesized that short-term forced aeration can favour lignin degradation over easily
478 degradable fractions and therefore increase biodegradability and BMP. SD from plants A and
479 B were selected from the two previously described subgroups, as the lignin-like fraction
480 (NEOM) content was high (44% COD_{tot}) for the former and low (31% COD_{tot}) for the latter.

481 **Table 3** presents anaerobic biodegradation results. They suggest that the BMP of final post-
482 treated matter (BMP_{FPTM}) was not improved in comparison to the control for both plant SDs.
483 For plant A SD, the BMP_{FPTM} ranged from 126 to 154 Nm³ CH₄/t VS_{final} with values either
484 significantly lower or similar to the control. For plant B, raw BMP_{FPTM} values were also always
485 significantly lower than the control. These lower methane potentials are due to carbon
486 losses during the short-term aerobic post-treatment.

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

490

491 3.3. Impact of various strategies of solid digestate recirculation on plant methane
492 production and evaluation of potential economic gains or capacity offset.

493 **Table 4** displays calculation results that evaluate the effect of direct recirculation of 30%,
494 50% and 80% of SD produced each year on plant methane production. Moreover, the

495 potential additional economic gains (AEG) in an addition strategy case and the amount of
496 feedstock that can be offset ($\Delta_{\text{feedstock}}$) in a replacement strategy case are also provided.

497 Firstly, the higher the amount of SD directly recirculated, the higher the impact on plant
498 methane production. Direct recirculation of 30% of SD represents between 0.6% and 2.3% of
499 total plant methane production and when recirculating 80% of SD reaches between 1.6%
500 and 6.3%. Plants D and E that have a high amount of feedstock, a short to medium HRT and
501 mesophilic conditions display the highest impact of recirculation on plant methane
502 production that can be explained by the relatively high BMP as well as SD amount produced
503 yearly. Interestingly, the lowest impact of recirculation on plant methane production (only
504 1.6% at 80% SD recirculation) is obtained for plant C. It can mainly be explained by the long
505 HRT and the use of easy to degrade feedstocks that leads to low SD BMP.

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

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

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

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

535 **Table 5** presents for plant A, B and C the effect of recirculation of post-treated SD on plant
536 methane production. Additional thermo-alkaline post-treatment increases SD BMP between
537 30 and 46%. In comparison to direct recirculation, this allows an additional increase of total
538 plant methane production between 0.2% (plant B – 30% SD recirculated) and 1.2% (plant A –
539 80% SD recirculated). Consequences vary as a function of the type of recirculation but
540 conclusions remain similar:

541 (i) In an addition strategy, recirculation of post-treated SD leads to AEG between
542 10.1k€ and 59k€ per year. In comparison to direct recirculation, AEG after a
543 thermo-alkaline post-treatment are increased (+2.1k€ to +17.7k€). However,
544 these economic gains will hardly compensate for additional post-treatment
545 expenditures. Purchase of soda (540€/t at 2% w/w), on average already accounts
546 for 88% of the additional gains of plant A, 300% of the additional gains of plant B
547 and 67% of the additional gains of plant C. Besides, other expenditures can be
548 added to the cost of soda. OPEX has to be increased due to additional labour and
549 heating (thermophilic conditions) and the potential capital expenditure (CAPEX),
550 such as the cost for a post-treatment tank (at least 30k€). Therefore, such a
551 strategy does not prove to be economically viable.

552 (ii) Similar conclusions can be drawn in the case of a replacement strategy. Soda
553 post-treated SD allow to offset an additional amount of feedstock comprised
554 between 49 t (plant B – 30% SD recirculated) and 163 t (plant A – 80% SD
555 recirculated) per year (in this specific case, regarding the loading rate, it
556 represents 8 days of feedstocks). However, investment costs are again too high
557 and buying feedstocks should represent a less expensive strategy. As an example,
558 in the case of plant A with 80% SD recirculation, cost for soda (15.5k€ per year)
559 could be allocated instead to buy at least 346 t of catch crops at a high price of
560 45€ per ton (cost ranges normally between 15 and 45€/t) [30]. This amount is
561 more than two times higher than the additional 163 t of feedstocks offset by
562 recirculated post-treated SD. Consequently, in this case, buying feedstock largely
563 outcompetes the thermo-alkaline post-treatment of SD.

564 Short-term aeration post-treatments lead to lower SD BMP for plant A and B SD. Calculations
565 based on the recirculation of 50% of the total produced SD show that the recirculation
566 impact on total plant methane production is lower than for direct recirculation. Thus, in the
567 case of an addition strategy, slightly lower AEG are obtained in comparison to direct
568 recirculation (-800€ to -1,800€). Similarly, for a replacement strategy case, the amount of
569 feedstock that can be offset is slightly lower (-16 to -20 t/year). These results suggest that
570 short-term aeration is not a significant strategy either, despite its low cost. Besides, they also
571 indicate that SD should not be aerated nor stored in open air for too long before being
572 recirculated, to avoid methane losses in the form of CO₂ respiration.

573 Finally due to the low impact of SD direct recirculation on the total plant methane
574 production (mainly originating from low SD BMP), a post-treatment, however effective it
575 may be in tested scenarios, is unlikely to be profitable. Indeed, additional CAPEX and OPEX
576 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
30%	BMP at lab scale (Nm³ CH₄/t VS)	146	155	114	191	155
	BMP_{corrected_SD} (Nm³ CH₄/t VS)	117	124	91	153	124
	$\dot{m}_{SD_recirculated}$ (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} (Nm³ CH₄/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} (Nm³ CH₄/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
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} (Nm³ CH₄/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
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	30%	BMP_{corrected_SD} (Nm³ CH₄/t VS)	167	156	150
		V_{CH4_SD} (Nm³ CH₄/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
		Δ_{feedstock} – replacement strategy case (t/year)	203	238	106
		Δ_{feedstock} variation compared to direct recirculation (t/year)	+62	+49	+42
	50%	V_{CH4_SD} (Nm³ CH₄/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
		Δ_{feedstock} – replacement strategy case (t/year)	336	397	177
			Δ_{feedstock} variation compared to direct recirculation (t/year)	+101	+82
	80%	V_{CH4_SD} (Nm³ CH₄/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
		Δ_{feedstock} – replacement strategy case (t/year)	539	635	283
		Δ_{feedstock} variation compared to direct recirculation (t/year)	+163	+131	+112
Short-term aeration		BMP_{corrected_SD} (Nm³ CH₄/t VS)	109	116	/
	50%	V_{CH4_SD} (Nm³ CH₄/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	/
		Δ_{feedstock} – replacement strategy case (t/year)	219	295	/
			Δ_{feedstock} variation compared to direct recirculation (t/year)	-16	-20

530 3.4. Assessment of the impact of SD recirculation on HRT, SRT, digester TS and of
531 mixing costs on CSTR biogas plant

532 The evolution of CSTR plant HRT_{accurate} and SRT as a function of the applied recirculation
533 strategy and for three different percentages of recirculated SD (30-50 and 80% of m_{SD}) are
534 presented in **Table 6**. For the five CSTR biogas plants studied and in the case of an addition
535 strategy, HRT_{accurate} only slightly increases (less than one percent on average) since the
536 additional methane production from SD is relatively low in comparison to total methane
537 production. However, for SRT, the increase is more significant. SRT are on average 10%, 18%
538 and 33% higher, comparatively to SRT without recirculation, when 30%, 50% and 80% SD are
539 recirculated, respectively. For a replacement strategy, HRT_{accurate} and SRT increases are
540 slightly better, compared to an addition strategy because the initial feedstock supply was
541 reduced. Thus, comparatively to HRT_{accurate} and SRT without recirculation, HRT_{accurate} are on
542 average 2%, 3% and 4% higher and SRT are on average 11%, 21% and 38% higher, when 30%,
543 50% and 80% SD are recirculated, respectively.

544 For both strategies, recirculation of a fraction of total produced SD has a negligible impact
545 on the HRT_{accurate} but allows for the SRT to increase because the SD remains longer within
546 the biogas plant. These higher SRT values lead to a higher methane recovery (as SD BMP
547 between 67 and 191 $\text{Nm}^3 \text{CH}_4/\text{t VS}$). This can account for the previously determined increase
548 in plant methane production as well as the higher biogas plant efficiency. Finally, 200 days of
549 AD are seemingly required to reach a 95% methane recovery from the most complex
550 feedstocks to degrade, such as manure [31]. For plant SRT to approach such a value via SD
551 recirculation would be noteworthy for biogas plants that treat complex feedstocks (e.g. plant
552 A, B or D) in order to allow a high biogas plant efficiency. However, impact of recirculation
553 on the digester TS content should also be taken into account.

554
555

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
$RV_{\text{post-digester}}$ (m ³)	2,000	2,200	2,000	2,700	1,200
$\dot{m}_{\text{feedstock}}$ (t/year)	13,600	18,000	10,500	25,000	20,000
HRT _{classical} (days)	100	84	130	88	40
$\dot{m}_{\text{biogas}(0)}$ (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

556
557

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

566 5.30 m and switch off at 5.20 m). Thanks to this system, digestate can flow from the digester
567 to the post-digester. The position of the overflow pipeline coincides with the maximum
568 height that the digestate can reach and thus sets the volume of the CSTR biogas plant.
569 Hence, if the SRT increases while the volume of the biogas plant is fixed, the TS content of
570 digester should increase.

571

572 **Table 6** displays the evolution of digester TS according to the applied recirculation strategy
573 and to the different percentages of recirculated SD. For the five studied CSTR biogas plants,
574 results point out that an increase in the amount of recirculated SD also leads to an increase
575 in digester TS for both recirculation strategies. Thus, for an addition strategy, the initial
576 digester TS content rises, on average, by 7%, 14% and 25% (representing an average +0.8,
577 +1.5, +2.8% increase of the initial digester TS value), when 30%, 50% and 80%, respectively,
578 of the total produced SD is recirculated,. In the case of a replacement strategy digester, the
579 initial digester TS content rises on average by 6%, 11% and 20% (representing an average
580 +0.6, +1.2, +2.2% increase of the initial digester TS value), when 30%, 50% and 80%,
581 respectively, of the total produced SD is recirculated. Slightly lower increases in TS values for
582 the replacement strategy than for the addition strategy can be explained by the removal of
583 certain feedstocks from the initial feedstock supply. Such rises in digester TS content are not
584 negligible; furthermore they should modify digestate viscosity and potentially affect digester
585 and post-digester mixing. Digestate rheology and mixing costs need to be further discussed,
586 to better define conditions of application for SD recirculation.

587 Rheological studies in anaerobic digestion have been historically focused on activated sludge
588 and raw manure. Only recently have several studies have been performed on digestate from
589 agricultural biogas plants [32]. In most of these studies, agricultural digestate originates from

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

609

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

614 (0.61 to 6.25% of total methane produced). If this value is used as an example and is specific
615 to one digester, it implies that the mixing energy consumption is significant and should be
616 taken into account when evaluating the SD recirculation strategy. If the TS content and
617 digestate viscosity increase after SD recirculation, it is likely that the mixing energy
618 consumption would also increase. However, it remains difficult to determine precisely, and
619 even more to generalise, the extent to which the energy consumption can rise. For instance,
620 for 13 full-scale biogas plants, no clear relationship has been observed between the average
621 TS content during operation and mixing energy consumption, since the highest TS content is
622 not associated to the highest mixing energy consumption [39]. This can be explained by the
623 fact that mixing energy consumption is not only dependant on the TS content as well as
624 digestate viscosity but also on the type of mixers, their numbers, the type of impellers, the
625 agitation frequency and the agitation speed [39]. All these parameters can greatly vary from
626 one biogas plant to another; therefore the sensitivity of mixing energy consumption towards
627 an increase in TS can also be likely to vary according to the biogas plant.

628

629 Indeed, because of i) the specificity of each biogas plant mixing system and of ii) the current
630 lack of precise correlation between a digestate TS and its viscosity, a turnkey equation
631 allowing for the variation in mixing costs as a function of digester TS cannot be provided. For
632 a given biogas plant, the relationship between mixing costs and digestate TS should rely on
633 the operator's empirical knowledge. Overall, the impact of SD recirculation on digestate
634 viscosity and mixing costs can vary strongly from one biogas plant to another. The relevance
635 of SD recirculation therefore has to be evaluated on a case-by-case basis; this will be further
636 described in section 3.5. Nevertheless, regarding the potential increase in digester TS
637 content displayed in **Table 6**, recirculation above 50% of total SD produced is likely to be

638 unreasonable and not economically profitable. A noteworthy potential trade-off between
639 additional methane or offset capacity and higher mixing cost may even consist in low to
640 medium recirculation percentage values ($R \leq 0.5$).

641 3.5. A potential approach to determine the significance of SD recirculation for a given CSTR 642 biogas plant

643 In order to evaluate the significance of SD recirculation for a given CSTR biogas plant, the
644 following approach can be applied. Firstly, the operator must determine the VS content and
645 BMP of its SD as well as the annual quantity of SD produced from the phase separation
646 system ($\dot{m}_{SD\text{year}}$). This information would allow for the potential gains/offset capacity to be
647 calculated using **Eq. 7, 8, 9** and **10**. The potential increase in TS could then be calculated
648 according to the % of SD recirculated (R) using **Eq. 14** as well as **Eq. 17** based on average
649 $[TS]_{\text{digester}}$, annual quantity of feedstock incorporated ($\dot{m}_{\text{feedstock}}$), annual quantity of biogas
650 produced (\dot{m}_{biogas}) and phase separator properties (α and SI). The new calculated digester TS
651 values should then be compared with the working range of installed mixing equipment.
652 **Appendix D** provides an overview of the estimated upper TS content range that a certain
653 type of mixer can handle. It shows that except for submersible motor mixers equipped with
654 a propeller, most existing mixing technologies can handle TS contents above 10%. If the new
655 calculated digester TS lies within the working range, the operator should be able to
656 determine (from a historical monitoring of the plant) the potential impact of a given increase
657 in TS on the mixing costs. When applicable, observations dating from periods with a similar
658 TS content might be useful for assessing the extent of the increase in the electrical
659 consumption or maintenance rate. Besides, the presence of an on-site solid size-reduction
660 device would have to be taken into account since it would reduce the impact of SD
661 recirculation on digestate viscosity. If the plant operator should consider that the potential

662 increase on mixing costs would be sufficiently low in comparison to the potential
663 gains/offset capacity, then the implementation of SD recirculation can be initiated.

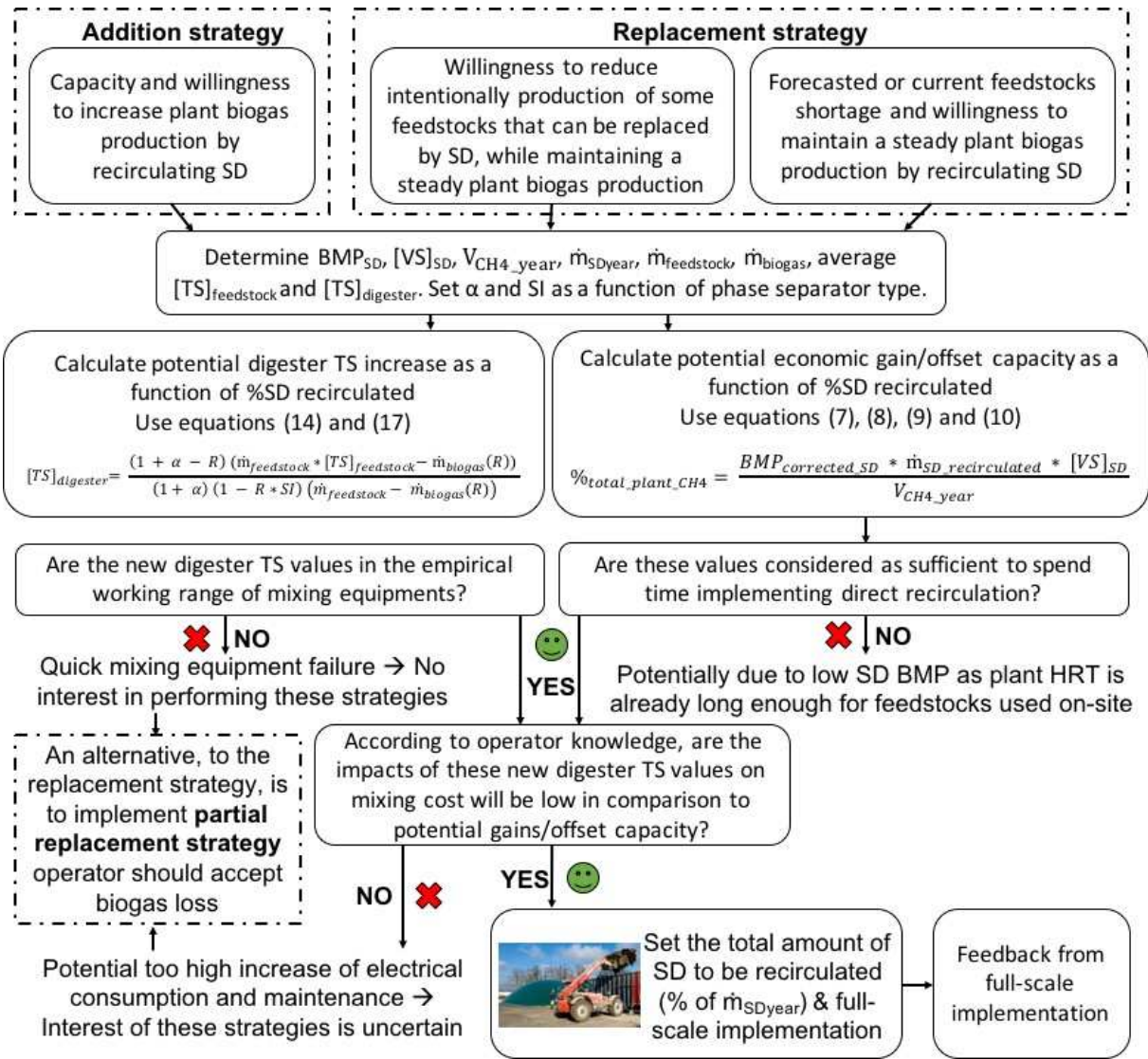
664 During the implementation of SD recirculation, it is likely that, while the digester TS rises, SD
665 production from the screw press should also increase. Thus, if SD recirculation should be
666 performed over a long-term basis, it may be expected that the total produced SD during the
667 initial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

673

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

681

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



687

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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.

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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

699 strategy unless it should remain at low levels ($R \leq 0.1$). A partial replacement strategy could
700 also be envisioned if ever feedstock shortage occurs. For plant B, the digester is equipped
701 with one Flyght® 4430 submersible motor mixer with large-blade rotors (Xylem, New York,
702 United-States) and two Biogator® HPR 1 paddle mixers (REMA GmbH, Hausen, Germany).
703 For this plant, the average TS content in the digester without recirculation (10.5%) is in the
704 low range of mixing equipment working capacity (13 to 14% TS). Besides, the presence of a
705 PreMix incorporator, equipped with a RCX® Rotacut shredder (Vogelsang, Essen, Germany)
706 may potentially reduce the impact of SD recirculation on digestate viscosity. Here,
707 recirculation can be envisioned as a potential strategy; indeed, even at a high percentage of
708 recirculation ($R=0.8$), the TS in the digester would remain within the working range. The
709 relevance of this strategy remains to be assessed by the plant operator according to the
710 potential gain/offset capacity and to the estimated increase in mixing costs.

711

712 Finally, different full-scale implementation cases for solid and liquid digestate recirculation
713 can be identified. LD recirculation can be used to dilute high TS feedstocks. In cases, where
714 storage tank is not covered and heated, additional methane might be obtained from LD.
715 However, particular attention should be paid to ammonia inhibition, especially for plants
716 treating nitrogen rich feedstocks, as it may accumulate in digesters with LD recirculation.
717 Intermediate air stripping devices for nitrogen removal [40] or adaptation of the
718 recirculation rates [41] might represent potential solutions to avoid inhibitions. In
719 comparison, SD recirculation may serve to increase the TS content for plants treating large
720 amounts of liquid feedstocks. Besides, as indicated for composted solid digestate, it may
721 have an additional buffering impact in the digester and allow for stabilisation. In addition,
722 according to the recirculation rate, both LD and SD recirculation are likely to allow for an

723 increase or at least sustain anaerobic microbial load and community in the digester. The
724 application of these recirculation strategies should depend on the features of the biogas
725 plant as well as the different stages of its operation (onset of acidogenesis, seasonal high
726 loading of TS feedstocks...). Further full-scale studies to increase our understanding of such
727 practices (for instance on potential heavy metals accumulation in the digester) are required
728 as they are likely to be part of the occasional/permanent tools to reach excellence in biogas
729 plant operation.

730

731 4. Conclusions

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

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

746 low mixing costs). Such a potential increase is significant for biogas plant operators and
747 represents a relevant way to reduce biogas production costs.

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

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

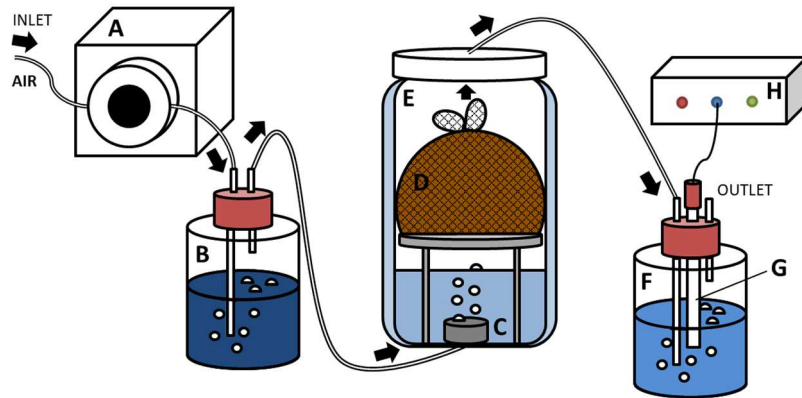
764 [Acknowledgment](#)

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767 [Declaration of interest](#)

768 Declarations of interest: none

769 Appendix A

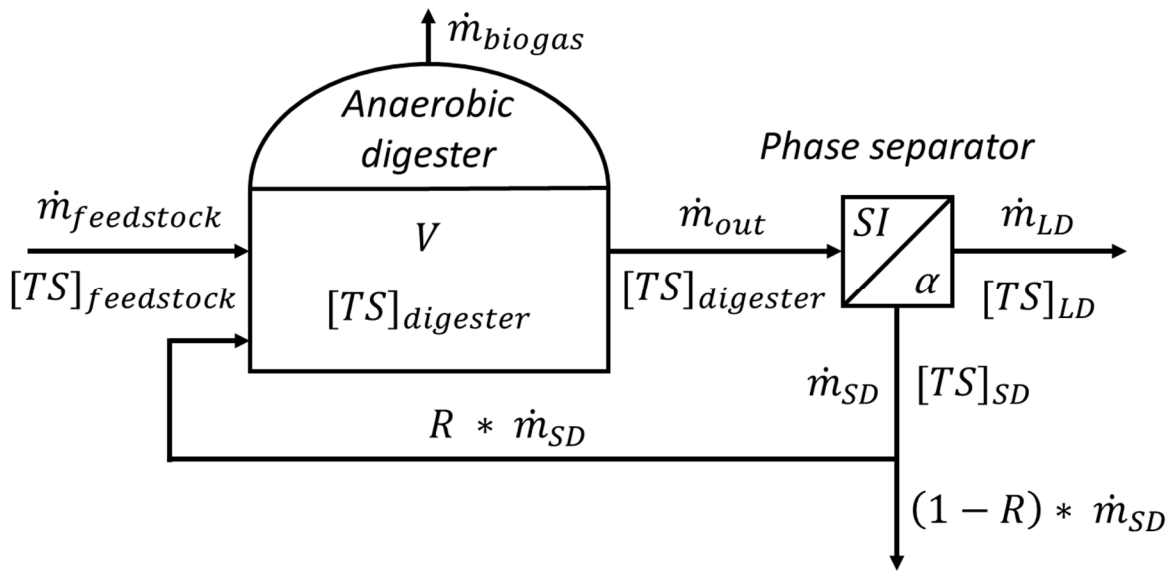


770
 771 *Figure A: Experimental set-up for short-term aerobic post-treatment of SD. (A) Calibrated*
 772 *peristaltic pump; (B) Air CO2 trap using 2M NaOH solution; (C) Humidifier system; (D) SD*
 773 *contained in a net; (E) 2.5 litres double jacket aerobic reactor at 30°C; (F) NaOH 0.5M trap for*
 774 *CO2 emitted from SD respiration; (G) Conductivity probe; (H) Acquisition system.*

775

776 Appendix B

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



779
 780 *Figure B: CSTR biogas plant equipped with a phase separator and performing solid digestate*
 781 *recirculation*

782

783 With the following known variables:

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

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

791 As well as the following unknown variables:

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

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

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

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

$$806 \quad \dot{m}_{feedstock} - \dot{m}_{biogas} = \dot{m}_{LD} + (1 - R) * \dot{m}_{SD}$$

807 (B.1)

$$808 \quad \dot{m}_{feedstock} * [TS]_{feedstock} - \dot{m}_{biogas} = \dot{m}_{LD} * [TS]_{LD} + (1 - R) * \dot{m}_{SD} * [TS]_{SD}$$

809 (B.2)

$$810 \quad \dot{m}_{out} = \dot{m}_{LD} + \dot{m}_{SD} \tag{B.3}$$

$$811 \quad \dot{m}_{out} * [TS]_{digester} = \dot{m}_{LD} * [TS]_{LD} + \dot{m}_{SD} * [TS]_{SD}$$

812 (B.4)

$$813 \quad \alpha * \dot{m}_{SD} = \dot{m}_{LD} \tag{B.5}$$

$$814 \quad SI = \frac{\dot{m}_{SD} * [TS]_{SD}}{\dot{m}_{out} * [TS]_{digester}}$$

815 (B.6)

$$816 \quad 1 - SI = \frac{\dot{m}_{LD} * [TS]_{LD}}{\dot{m}_{out} * [TS]_{digester}} \tag{B.7}$$

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

$$819 \quad SRT = \frac{V * [TS]_{digester}}{\dot{m}_{LD} * [TS]_{LD} + (1 - R) * \dot{m}_{SD} * [TS]_{SD}}$$

820 (B.8)

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

$$822 \quad \dot{m}_{SD} * [TS]_{SD} = SI * \dot{m}_{out} * [TS]_{digester} \quad \&$$

$$823 \quad \dot{m}_{LD} * [TS]_{LD} = (1 - SI) * \dot{m}_{out} * [TS]_{digester}$$

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

$$826 \quad SRT = \frac{V}{(1-R*SI)*\dot{m}_{out}}$$

827 (B.9)

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

$$829 \quad \dot{m}_{feedstock} - \dot{m}_{biogas} = (1 + \alpha - R) * \dot{m}_{SD}$$

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

$$831 \quad \dot{m}_{out} = (1 + \alpha) * \dot{m}_{SD}$$

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

$$833 \quad \dot{m}_{out} = \frac{(1 + \alpha) * (\dot{m}_{feedstock} - \dot{m}_{biogas})}{(1 + \alpha - R)}$$

834 (B.10)

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

$$837 \quad SRT = \frac{(1 + \alpha - R)}{(1 + \alpha) * (1 - R * SI)} \frac{V}{(\dot{m}_{feedstock} - \dot{m}_{biogas})}$$

838 (B.11)

839 α is a positive real number. The higher the value, the lower the quantity of SD produced. The
840 SI index is comprised between $1/(1 + \alpha)$ and 1. When SI is equal to $1/(1 + \alpha)$, SD is not
841 enriched in TS in comparison to LD and raw digestate is only separated in two flows
842 (according to α). When SI is equal to 1 it means that the efficiency of the phase separation is
843 maximal and that all total solids end up in the SD. α and SI values can be selected according

844 to the average mass and total solids distribution profiles provided from a recent study on
 845 digestate mechanical separation [6]. In the case of a low efficiency separator (e.g. screw
 846 press), typical values for α and SI are 9 and 0.38 respectively. In the case of a high efficiency
 847 separator (e.g. centrifuges), typical values for α and SI are 2.45 and 0.81 respectively. R is
 848 comprised between 0 and 1. When R is equal to 0, no SD is recirculated and the SRT is equal
 849 to the HRT. When R is equal to 1, all produced SD is recirculated.

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

$$852 \quad \dot{m}_{feedstock} * [TS]_{feedstock} - \dot{m}_{biogas} = \dot{m}_{out} * [TS]_{digester} * (1 - R * SI)$$

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

$$855 \quad [TS]_{digester} = \frac{(1 + \alpha - R)}{(1 + \alpha) * (1 - R * SI)} * \frac{(\dot{m}_{feedstock} * [TS]_{feedstock} - \dot{m}_{biogas})}{(\dot{m}_{feedstock} - \dot{m}_{biogas})}$$

856 (B.12)

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

$$859 \quad [TS]_{feedstock} = \frac{[TS]_{digester} * (\dot{m}_{feedstock} - \dot{m}_{biogas}) + \dot{m}_{biogas}}{\dot{m}_{feedstock}}$$

860 (B.13)

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

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

$$868 \quad \dot{m}_{biogas}(R) = \dot{m}_{biogas}(0) + \frac{\dot{m}_{SD_recirculated}(R) * [VS]_{SD} * BMP_{SD} * (d_{CH_4} + \frac{(1-x_{CH_4})}{x_{CH_4}} * d_{CO_2})}{1000}$$

869 B.14)

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

872

873

$$874 \quad SRT = \frac{(1 + \alpha - R)}{(1 + \alpha) * (1 - R * SI)} \frac{V}{(\dot{m}_{feedstock} - \dot{m}_{biogas}(R))}$$

875 (B.15)

$$876 \quad [TS]_{digester} = \frac{(1 + \alpha - R)}{(1 + \alpha) * (1 - R * SI)} * \frac{(\dot{m}_{feedstock} * [TS]_{feedstock} - \dot{m}_{biogas}(R))}{(\dot{m}_{feedstock} - \dot{m}_{biogas}(R))}$$

877 (B.16)

878

879 [Appendix C](#)

880 After short-term aerobic post-treatments, the fractionation method, developed by Jimenez
 881 et al. [21], was performed on aerated SD from biogas plant B. However, instead of using COD
 882 tests to evaluate the distribution of VS in the different fractions, the TC content was
 883 measured to understand carbon distribution after the post-treatment. TC was measured on
 884 the four fractions (SPOM, SEOM, PEOM and NEOM) and on the raw sample. Besides, the

885 fluorescence spectra of liquid extracts were recorded on a Perkin Elmer LS55 and a
886 complexity ratio was calculated accordingly to Jimenez et al. [33]. This index is defined as the
887 ratio of the sum of the fluorescence volumes of the most complex molecules (lignin, humic
888 acid...) over the sum of the fluorescence volumes of the protein-like molecules.

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

908 these results into account, it is likely that under aerobic conditions, degradation of complex
 909 fractions may only take place once the easily degradable matter has begun to be degraded.

910 *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

911

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913

914 Appendix D

915 *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

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