- 1 Biomass fuel production from cellulosic sludge through biodrying: aeration
- 2 strategies, quality of end-products, gaseous emissions and techno-economic
- 3 assessment
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Abstract

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This study assesses the technological, environmental and economic feasibility of biodrying to valorise cellulosic sludge as a renewable energy source. Specifically, three different aeration strategies were compared in terms of biodrying performance, energetic consumption, gaseous emissions, quality of end-products and technoeconomic analysis. These strategies were based on different combinations of convective drying with biogenic heat produced. Two innovative biodrying performance indicators (Energetic Biodrying Index and Biodrying Performance Index) were proposed to better assess the initial and operational conditions that favour the maximum energy process efficiency and the highest end-product quality. The end-products obtained consistently presented moisture contents below 40% and lower heating values above 9.4 MJ·kg-1. However, the best values achieved were 32.6% and 10.4 MJ·kg-1 for moisture content and lower heating value, respectively. Low N₂O and CH₄ emissions confirmed the effective aeration of all three strategies carried out, while NH₄ and tVOCs were related either to temperature or biological phenomena. A techno-economic analysis proved the economic viability and attractiveness of the biodrying technology for cellulosic sludge in all the strategies applied. Keywords: cellulosic sludge, biodrying, aeration strategies, gaseous emissions, technoeconomic analysis.

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45 **Abbreviation list:**

AT4 4 days cumulative oxygen consumption

BA Bulking Agent BI. Biodrying Index

BPI Biodrying Performance Index

CAPEX Capital expenditures

COD Chemical Oxygen Demand

CS. Cellulosic Sludge

DRI Dynamic Respirometric Index
EBI Energetic Biodrying Index
EC Energy Consumption
EP Energy Production
GHG Greenhouse Gases
FAS Free Air Space

HHV Higher Heating Value IRR Internal Rate of Return LHV Lower Heating Value MC Moisture Content MSW Municipal Solid Waste NPV Net Present value

OPEX Operational expenditures
PE Population Equivalents
SRF Solid Recovered Fuels

TIP Temperature Increasing Phase

TS Total Solids

tVOC Total Volatile Organic Compounds

VS Volatile Solids

VS-CS Volatile Solids from Cellulosic Sludge

WWTP Wastewater Treatment Plant

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1. Introduction

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The recovery of resources (materials and energy) from wastewater is a promising solution to the relevant sustainability challenges of water utilities in modern societies There is a wide range of innovative technologies that are currently being applied, which not only increase the efficiency of wastewater treatment plants (WWTP), but also reduce the amount of sludge produced, reduce the energy consumed and provide clear environmental and economic benefits (Conca et al., 2020; Da Ros et al., 2020). These new technologies are applied at different stages during water treatment, but mostly in side streams and down streams. On the one hand, these flows present high concentrations of COD, TS and nutrients, and are normally considered suitable candidates to implement resource recovery strategies (Raheem et al., 2018). On the other hand, the impact on the overall increase in WWTP efficiency and energy savings, although positive, still has some limitations and margin for improvement. To increase WWTP efficiency while increasing resource recovery capacity, new technologies were developed and applied during the first stages of the main stream (Reijken et al., 2018; Larriba et al., 2020). A part from the direct impacts on resource recovery, these technologies also have indirect impacts on the efficiency of the subsequent stages; they significantly reduce the chemical oxygen demand (COD) and total solids (TS) content and consequently, reduce the aeration needs and the amount of sewage sludge produced, translating into important energy savings. Among these innovative promising technologies, Cellvation® aims to maximise the recovery and recycling of cellulose, replacing, partially or totally, the primary settler. Cellvation® consists of several steps, an initial grit and hair removal step in a rotating

drum filter, followed by a 350 µm fine sieve (Salsnes Filter, Norway), and subsequently, a cellpress, a hygienisation step and finally, cellulose recovery in the form of Recell® cellulose pellets. However, a cellulosic sludge (CS) is also produced. To avoid the loss of resources and minimise disposal costs, the CS could be further valorised considering the high potential energetic content of this material due to the high content in cellulose and hemicellulose. Thus, all the cellulose-rich sludge obtained after the cellpress could be considered as suitable material for valorisation via energy recovery technologies. Among the different technologies that could be applied, biodrying is presented as an innovative, energy-saving and environmentally friendly alternative for CS and sewage sludge energetic valorisation. Biodrying, considered similar to composting, is an aerobic biological process that uses the biogenic heat produced during the decomposition of biodegradable organic matter to remove as much moisture as possible in the shortest operation time (Cai et al., 2012). Additionally, biodrying aims to preserve most of the organic matter present in the raw material, in the final biomass fuel produced (Huiliñir and Villegas, 2014). Biodrying performance is normally assessed using two main indices: the daily drying rate and Biodrying Index (BI). However, these indices present some limitations, since daily drying rates do not consider the organic carbon biodegraded and BI does not consider the external energy consumed presents some difficulties over other organic wastes for its valorisation through biodrying, where the most significant issue relates to its low porosity and high moisture content, which can hamper proper air diffusion through the raw material. This technology has not yet been optimized for low-porosity

organic wastes, and improvement is still needed in terms of its performance and

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efficiency. This could amplify possible valorisation opportunities and applications for different types of sewage sludge.

Assuming suitable initial conditions (e.g., organic content of raw materials and

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matrix structure and porosity), water evaporation in the biodrying process depends mainly on two operational parameters: (1) airflow temperature (inlet and outlet) and (2) airflow rate. Previous studies have assessed the technical performance of biodrying processes using continuous and discontinuous aeration strategies and a wide range of specific airflow rates from 0.5 to 6.2 L min⁻¹kg⁻¹VS-CS (Zhao et al., 2010; Huiliñir and Villegas, 2014). However, an effective biodrying process should not only be considered from a technical perspective but also by its environmental and economic sustainability. Therefore, an optimised biodrying process should guarantee: (1) low energy consumption and low harmful gaseous emissions, allowing, in turn, (2) the production of a high-quality biomass fuel, hence maximising the net energy recovery. The key quality indicators of the biodried products obtained are low moisture content (MC) and high calorific potential. There are few previous references about economic viability of biodrying technologies applied to low-porosity materials. In these studies, the main economic weaknesses were indeed related to the high MC of final products (Navaee-Ardeh et al., 2006). Nonetheless, electricity demand, particularly for aeration, is recognised to be the main operational cost during biodrying processes (Psaltis and Komilis, 2019). Consequently, choosing the most appropriate aeration strategy will lead to important energy savings as well as the improved environmental performance of the process.

Regarding environmental performance, a lack of information in the literature exists in regards to gaseous emissions during the biodrying process, for both sewage sludge and Municipal Solid Waste (MSW) valorisation (Ragazzi et al., 2011; González et al., 2019a).

Therefore, the main objective of this study was to develop an in-depth performance assessment of biodrying processes from a technical, environmental and economic point of view, in the particular case of CS used as raw material. Specifically, process performances and quality of end-products under three different aeration strategies were compared in terms of process efficiency, gaseous emissions and economic feasibility. In addition, new process performance indices were proposed to overcome the limitations of the currently used indices, in order to give a more detailed and comprehensive assessment of biodrying processes.

2. Materials and methods

2.1. Raw materials and initial mixture

Cellulosic sludge was collected from the WWTP of Geestmerambacht, the Netherlands. In this case, Cellvation® cellulose recovery technology treats 30-80 m³·h¹¹ of wastewater. This system reduces the total suspended solids up to 40%, which can be translated into energy savings of up to 15% and a reduction of sewage sludge production of up to 20% (Cellvation, B.V., 2018). The raw material used in this study was a mix of intermediate cellulose-rich flows, the so-called, CS. The main physicochemical characteristics of CS are presented in Table 1, including a comparison with other conventional sludges.

Pruning waste was used as bulking agent (BA), obtained from the Parc Ambiental de

Bufalvent MSW composting plant located in Manresa, Spain.

The sludge and the bulking agent were mixed manually. The mixture ratio used was

1:2.5 of CS to pruning waste, allowing an optimal range of MC and Free Air Space

(FAS) close to 50-60% (Villegas and Huiliñir, 2015) and 70%, respectively, of all the

initial mixtures used in this study.

2.2. Experimental equipment and operation

2.2.1 Biodrying reactor operation

A near-to-adiabatic reactor with a working volume of 100L was used for all biodrying trials. The reactor was aerated through a diffusion grid in the bottom using an air compressor (Dixair DNX 2050, Worthington Creyssensac) and a flowmeter/controller (D-6311-DR, Bronkhorst High-Tech B.V.). Humidity of inlet air was controlled by installing a set of two filters for moisture and particle removal before the flow meter/controller. During the biodrying trials, inlet air and matrix temperatures were monitored using proves (Pt-100). A representative sample of exhaust gases (0.14 L·min⁻¹) was continuously pumped and analysed using O₂ and CO₂ sensors (O₂A₂ and IRC A₁, respectively, Alphasense). Weight loss was monitored with a scale (Gram Precision / k3-k3i, Gram group). Arduino UNO was used for data acquisition and LabView2017 (National Instruments) software was used for data analysis, process monitoring and airflow control. Material homogenization, was carried out using a maze spiral compost aerator. The turning frequency criteria adopted was once per day during the thermophilic stage of the process while it was once per two days during late mesophilic and cooling stages.

2.2.2. Control system

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Three different aeration strategies were adopted for cellulosic sludge biodrying: (1) set an airflow that can maintain the highest bulk material temperature and the longest thermophilic phase duration (S1); (2) set high airflows, triple the values of S1 airflows (S2); and (3) a combined strategy where S1 airflows were maintained until the thermophilic phase was over (below 45°C), plus S2 airflows thereafter (S3). An algorithm to adapt aeration rates to 5 temperature ranges (<35°C, 35-45°C, 45-55°C, 55-70°C and > 70°C) was developed, in which aeration rates per range were adapted to the particular strategy assessed. For the first strategy, optimal aeration levels typically used during composting processes were chosen, with the aim to use bulk temperature as the main water removal driver. For the second strategy, aeration levels were set significantly higher, particularly in the thermophilic stage, in order to facilitate the extraction of the evaporated water and ultimately improve water removal (Navaee-Ardeh et al., 2006). In the S3, a combination of the previous strategies was tested, aiming to maximise moisture removal within the two stages, by firstly maximising temperature (equivalent to S1) during the thermophilic stage and secondly maximising aeration rates (equivalent to S2) during the mesophilic-cooling stage.

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2.3. Analytical methods

All analysis were made following the Standard Methods for the Examination of Water and Wastewater (APHA, 1995), with the exception of pH and conductivity measurements, which were carried out following the Test Methods for the Examination of Composting and Compost (US Department of Agriculture and US Composting

Council, 2001). C/N and FAS values were estimated from chemical characterisation as suggested and used elsewhere (Richard et al., 2002; Villegas and Huiliñir, 2014). Biological stability, by means of Dynamic Respiration Index (DRI) and 4 days cumulative oxygen consumption (AT4), were determined using a dynamic respirometer developed by Ponsa et al. (2010).

Higher heating value (HHV) of wastes was determined using a bomb calorimeter (1341 Plain Jacket Calorimeter with the 1108 Oxygen Combustion Vessel, Parr) according to manufacturer instructions. Briefly, the pelletised biodried samples between 0.6-1g was electrically ignited in pure oxygen environment (30 atm) and the heat of combustion was monitored for subsequent calculations. Lower heating value (LHV) was calculated from HHV by correcting it following the equation given by Koppejan and Van Loo, (2012) and applied elsewhere (Gonzalez et al., 2019a).

2.4. Calculation of mass balances and performance indicators

Organic matter mineralisation during biodrying was calculated according to the ash conservation principle (Cai et al., 2012). Accordingly, final Volatile Solids (VS) mass was calculated from VS content of representative products after homogenisation and grinding. The VS loss ratio was estimated for every stage (lag, thermophilic and late mesophilic-cooling stages) from the percentage of cumulative O₂ consumption monitored in each stage. Then, those values were used to calculate moisture content removal, correcting it from monitored mass loss. Biodegradation of the bulking agent was assumed to be negligible (Ponsá et al., 2011) as it was confirmed through dynamic respirometry tests.

From the process efficiency point of view, daily drying rates are typically used to assess experimental results. This parameter is clearly scale-dependent, and it does not consider the organic carbon consumed. As the aim of the biodrying process is to obtain a high-quality biomass fuel with high calorific potential, degradation of VS during the process should be considered. Regarding this, the ratio of moisture removed per mass unit of organic matter lost is presented as the appropriate indicator reflecting the efficiency of the process, the so-called biodrying index (BI) (Hao et al., 2018). In the current study, apart from the overall BI, daily indices were also calculated to identify and consider, stage per stage, the most important parameters affecting the process.

Moreover, for a more appropriate assessment of the process, energy consumption and energy production potential parameters were introduced into the BI calculation obtaining two new indices. Hence, those parameters could reflect the energetic, economic and environmental viability of a certain biodrying process. Consequently, the new Energetic Biodrying Index (EBI) and Biodrying Performance Index (BPI) are presented (Equation 1 and 2, respectively).

$$EBI = \frac{1}{m_{VS}} \cdot \frac{1}{EC/m_{H2O}}$$
 (Equation 1)

 $BPI = \frac{1}{m_{VS}} \cdot \frac{1}{EC/m_{H2O}} xEP$ (Equation 2)

Where m_{H2O} is the water content lost (in mass) in the period of interest, m_{VS} is the VS content consumed in the same period, EC is the overall specific energy consumption during the period (per dry mass of treated CS) and EP is the energy potential production

of sieved product in terms of HHV considering its specific production ratio (corrected per dry mass of treated CS).

Additionally, an indicator referred to as the mass conservation efficiency was used,
which indirectly measured the VS conservation capacity, and is suggested by means of
the specific production ratio (Equation 3).

235 Specific production ratio =
$$\frac{m_{TS} \, product}{m_{TS-CS} \, fed}$$
 (Equation 3)

Referring m_{TS} product to the absolute mass of TS contained in the product and m_{TS-CS} fed to the absolute TS mass from CS fed into the process at the beginning of the batch.

2.5. Gas and odour emissions: sampling and analysis

Samples were daily collected in Nalophan® bags by using a semi-spherical stainless-steel flux chamber (Scentroid, IDES Canada Inc.) and a vacuum pump. CH₄ and N₂O analysis were carried out using an Agilent 6890 N Gas Chromatograph (Agilent Technologies, Inc.) equipped with a flame ionisation detector and an electron capture detector for CH₄ and N₂O detection, respectively. Total Volatile Organic Compounds (tVOC), NH₃ and H₂S concentration in exhaust gas were measured *in situ* using a MultiRAE Lite analyser (RAE Systems). The extended sampling method and gas analysis can be found in González et al. (2019a).

2.6. Techno-economical assessment

An economic assessment for the implementation of the biodrying technology in WWTPs was performed, focusing solely on the CS valorisation step by using a biodrying process that produces a biomass fuel with economic value.

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Real scale WWTP data were provided by Cirtec B.V. The specific CS production of the WWTP studied was 1.1E-02 t·PE⁻¹·y⁻¹, given total annual CS production of 2,920 tons while serving to 262,000 Population Equivalents (PE). Raw material characteristics were defined as an average of the values obtained experimentally. From this starting point, performance efficiencies experimentally obtained, were assumed for the mass balance calculation. Real budget data were used for the calculation of investment costs, assuming the construction of concrete biodrying trenches, a cover for roofing and an aeration system based on blowers. For yearly costs calculations, energy consumption (electricity and diesel), personnel costs, BA costs, pelleting, maintenance and insurance costs were estimated. The energy consumption of equipment was upscaled based on experimental data and adapted to the information provided by the industrial composting plant consulted (Aigües de Manresa S.A., Spain). The market price value of endproducts was determined according to the specific energy content of biodried products and biomass energy selling price reported by Avebiom (2019). Annual revenues were corrected from product selling earnings considering yearly Operational Expenditures (OPEX).

The economic parameters calculated were: Capital Expenditure (CAPEX), OPEX,
Revenues, Net Present value (NPV), Internal Rate of Return (IRR) and Payback Period
using Equations 4 and 5,

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$$NPV(\in) = \sum_{t=1}^{T} \frac{B_t - C_t}{(1+r)^t} - K$$
 (Equation 4)

 $\frac{(B_t - C_t)}{(1 + IRR)^t} = 0$ (Equation 5)

where Bt are the annual benefits coming from the full-scale implementation of the CS biodrying system, in this specific case product selling and sludge management fees; Ct are the annual costs of the project implementation (OPEX); r is the discount rate, in which a value of 7% was used and derived from Imeni et al., (2019) for these types of projects, and K are the investment costs expressed in € (CAPEX). t is the lifespan of the project which was fixed to 25 years.

Payback period was calculated through the sum of annual cash flows over time until a positive value was achieved, once this time is reached, net profit period would start.

From this initial framework, a breakeven point analysis was performed to find the zero-profit scenario and determine the minimum feasible capacity of a treatment plant (Imeni et al., 2019).

3. Results and discussion

3.1. Process evolution: temperature, moisture content and airflow rates

Temperature profiles, moisture and airflow evolution obtained during all three trials are shown in Fig. 1. In general, temperature profiles are comparable with those found in literature regarding sewage sludge biodrying (Zhao et al., 2010). Maximum temperatures achieved were equivalent for S1 and S3 (72°C and 73.4°C, respectively), and temperature profiles remained roughly similar until the process entered a late mesophilic stage, when the aeration rate clearly differed. As expected, the temperature profile with S2 was different, reaching 55°C after 24h and maintained for 2 more days.

After nearly 4 days of operation, a maximum temperature peak of 63.5°C was achieved. Thermophilic temperatures were maintained for 5.3, 4.1 and 4.9 days with S1, S2 and S3, respectively. Compared to previous studies using a similar scale (Zhao et al., 2010; Vilegas and Huiliñir, 2014), both the maximum temperatures achieved and the length of

the thermophilic stage with S1 and S3 were improved in the current study.

As shown in Fig. 1, airflow rates supplied were considerably different for the three strategies. Use of S2 high airflow rates (up to 3.5 L·min-1·kg-1 VS-CS) probably led to a delayed temperature peak as well as a comparatively higher heat loss after the temperature peak (shorter thermophilic stage). However, thermophilic temperatures and satisfactory biodrying performance were achieved, demonstrating that selected airflow rates for S2 were not high enough to impair biodrying process. As expected, the Temperature Increasing Phase (TIP) and Thermophilic stages in S1 followed the same trend as in S3. However, the higher aeration rates used in S3 from day six onwards, significantly affected its temperature profile. It is likely that, after day eight, the biogenic temperature generation was not able to counterbalance the heat loss due to high aeration rate and consequently, the temperature decreased to 25°C and remained constant until the end of the experiment. Accordingly, with S3, it could be assumed that only convective drying occurred after day eight.

Considering the results shown in Table 2, maximum moisture removal ratio was obtained when applying S2. MC removal ratios obtained were 55.0%, 62.4% and 57.5% for S1, S2 and S3 respectively. When comparing these results using a fixed VS mass consumption of 1.19 kg VS from S3, which was the minimum value obtained among the three strategies, the moisture removed by applying S2 would still be 38% and 11% higher than S1 and S3, respectively, demonstrating the high efficiency of S2. Moisture

removal ratios obtained in the current work were generally in the high range of what was previously reported in literature for similar low-porosity wastes, where most of the MC removal ratio values found were between 45 and 60% (Zhao et al., 2010; Huiliñir and Villegas, 2014). Only co-biodrying processes of sludges with other biodegradable wastes were reported to improve water removal efficiency (60-90%) (Zhang et al., 2018; Hao et al., 2018).

Due to the high temperatures achieved when applying S1 and S3 during the thermophilic stage together with its longer duration, this stage presented the highest MC removals. Conversely, for S2, the MC removal ratios were balanced among the thermophilic and mesophilic-cooling stages.

Cumulative oxygen consumption profiles were used to estimate VS consumption ratios in each stage of the processes (Table 2). Considering BA biodegradation negligible (Ponsá et al., 2011), maximum VS consumption from cellulosic sludge (36.7% of initial VS-CS content) occurred when applying S1 aeration. In contrast, when applying S3, the lowest VS consumption was determined (12.6% of the initial VS-CS content). Again, when comparing the results with a fixed value of moisture removed (11 kg of water corresponding to S3), there would be a 57% and 52% lower VS consumption for S2 and S3, respectively, than in the case of S1. As expected, maximum VS biodegradation occurred during the thermophilic stage, in which maximum absolute values in S1 were more than double that of the other two strategies. The low VS consumption obtained when applying high airflow rates (along all stages of S2 and mesophilic-cooling stage of S3) reinforces what other studies previously found about high airflow rates limiting biological activity and degradation of organic matter (Huiliñir and Villegas, 2014).

It is worthwhile to highlight the potential effectiveness of all three strategies implemented in terms of VS conservation, as even the highest VS consumption found for S1 (14.9% of bulk mixture VS) was found to be lower than previous studies (normally between 15 and 40% of VS consumption) (Zhao et al., 2010).

3.2 Process performance assessment

The most relevant performance assessment parameters for biodrying processes are:

(i) moisture removal; (ii) VS consumption and; (iii) the energy consumption during the process. Therefore, the aim would be to maximise moisture removal, while limiting VS consumption, and minimising energy consumption. The Biodrying Index (BI) is usually reported in the literature as a performance efficiency index that interrelates the first two of the mentioned key parameters. Additionally, Energetic Biodrying Index (EBI) is presented in this study as a new index integrating all three parameters, adding an energy consumption parameter into the performance efficiency assessment. When the abovementioned indices are determined daily, they would allow a semi-continuous process performance monitoring and an optimisation of biodrying efficiency.

Process monitoring, by means of BI and EBI, for the three strategies assessed, the

shown in Fig. 2a and b, respectively. When comparing the three strategies assessed, the best BI was obtained when applying S2 (9.8 kgH₂O·kg⁻¹VS), followed closely by S3 (9.2 kgH₂O·kg⁻¹VS), and finally by S1 (4.3 kgH₂O·kg⁻¹VS). The lowest BI obtained for S1 was expected due to its higher VS consumption, which were double those found for S2 and S3. The limitation of organic carbon mineralization is key to improving biodrying performance since it would affect the end-product's quality as an energy source. On the contrary, the best BI obtained corresponds to S2 mainly due to the high

MC removal ratio and the moderate VS consumption. Accordingly, some authors also reported that the airflow rate had more effect on moisture removal than on VS consumption (Vilegas and Huiliñir, 2014). The comparison of CS biodrying performance results with values reported in literature is presented in Table 3. Compared to other studies, all the strategies tested, especially S2 and S3, obtained satisfactory results in terms of process efficiency, due to high MC removal ratios, but more particularly due to the reduced VS consumption reported (Zhao et al., 2010; Huiliñir and Villegas, 2014). However, some of the authors reported higher BI values (up to 20 kg H₂O·kg⁻¹VS) compared to those presented in the current study (Villegas and Huiliñir, 2014). This difference could be due to the particularly low VS consumption associated to their low temperature profiles. In addition, when comparing values reported in sludge co-biodrying studies, the use of co-substrates resulted in higher MC removal ratios in general, but also significantly higher VS consumption values, lowering in these cases the overall BI values (up to 6 kgH₂O·kg⁻¹VS) (Hao et al., 2018; Zhang et al., 2018; González et al., 2019a). Some authors expose that overall water carrying capacity should be substantially higher when using high airflow rates, than that achieved due to high temperatures (Sharara et al., 2012). The better drying performance of S2 compared to S3 during the late mesophilic stage is probably due to the difference in bulk temperature during that stage. Although airflow rates were equivalent, the below-mesophilic temperatures found in S3 clearly hampered the drying efficiency compared to S2. The depletion of most biodegradable VS during the first half of the S3 trial seemed to have reduced the biogenic heat production in later stages, leading to low bulk temperatures. Thus, during the late mesophilic stage, although high airflow rates can result in good MC removal

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ratios, a minimum bulk temperature around 35-40°C seems to be necessary for an improved drying efficiency.

Fig. 2b shows the EBI profile along the three biodrying strategies assessed,

presenting clear differences among them, mainly related to their different aeration strategies.

Overall, considering the energy consumption during the process and the EBI, the most efficient strategy was found to be S2 (0.99 kg $H_2O\cdot kg^{-1}VS\cdot kWh^{-1}$), followed closely by S3 (0.85 kg $H_2O\cdot kg^{-1}VS\cdot kWh^{-1}$).

Conversely and although it had the lowest overall energy consumption, S1 obtained the lowest EBI value (0.62 kgH₂O·kg⁻¹VS·kWh⁻¹), particularly due to the high VS consumption, which did not particularly improve moisture removal efficiency.

Energy consumption data in biodrying studies are scarce and only a few studies present some data. Sharara et al., (2012), determined energy consumption values around $1 \text{ kWh} \cdot \text{kg}^{-1}_{\text{mix}}$, when treating livestock waste and using equivalent airflows as in S1.

Nevertheless, energy consumption data per water removed are more favourable, in the present study (0.4- 0.9 kwh·kg⁻¹H₂O) vs. those obtained in Sharara et al., (2012) (2.2-2.5 kwh·kg⁻¹H₂O), demonstrating the effective use of the biogenic heat produced combined with appropriate aeration strategies to improve moisture removal.

In summary, when analysing the efficiency parameters proposed, S2 seems to be the most efficient when considering moisture removal, BI and EBI. However, S3 also showed promising results, even though the mesophilic-cooling stage could be further optimised. The information provided by the indices proposed together with different aeration strategies would certainly facilitate the upscaling of the biodrying process.

3.3 Gaseous emissions

3.3.1 GHG emissions

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In regards to Greenhouse Gases (GHG), maximum N₂O emission rates were found 416 within the first hour of the process and at the 24th hour for CH₄. These maximum 417 emissions were likely related to anaerobic conditions during the dewatering and 418 419 shipping of raw materials (Han et al., 2018a). In fact, after adjusting the initial structure, 420 porosity and moisture content of the material, N₂O stored in sludge was probably stripped-out by forced aeration (Han et al., 2018a; González et al., 2019a) and was no 421 422 longer detectable. CH₄ emissions have been related to an inadequate mixture structure 423 and insufficient oxygen supply, leading to anaerobic conditions (Maulini-Duran et al., 2013; Yuan et al., 2016). For instance, when applying S1, which can be considered as 424 the worst-case scenario, maximum daily emission rates found for N₂O and CH₄ were 425 91.8 mg·d⁻¹ and 16.3 mg·d⁻¹, respectively. Additionally, the overall emission factors 426 calculated for N₂O and CH₄ were 6.8E-03 gN₂O·kg⁻¹TS and 2.6E-03 gCH₄·kg⁻¹TS, 427 428 which were lower than the values reported in biodrying and composting literature (Han 429 et al., 2018a; González et al., 2019a). Regarding the global warming effect, the maximum cumulated value was 2.13 g CO₂eq·kg⁻¹TS, corresponding to S1. This value 430 is almost three times lower than the values reported in conventional sewage sludge 431 biodrying (González et al., 2019a) and even lower than those of sewage sludge 432 433 composting (Yuan et al., 2016; Han et al., 2018a). 3.3.2 H₂S, NH₃ and total VOC emissions 434 In aerobic degradation processes such as composting or biodrying, H₂S, NH₃, and 435

tVOCs are the main compounds related to unpleasant odour emissions, and are

recognised as a significant weakness of those processes (Han et al., 2018a). Emission

profiles for NH₃ and tVOCs are shown in Fig. 3a and b, respectively. H₂S was never detected, reinforcing the effective aerobic conditions of biodrying mixtures with all the aeration strategies implemented (Han et al., 2018b). NH₃ and tVOC emissions followed a typical profile where maximum NH₃ emission peaks coincided with thermophilic temperatures, whereas tVOCs were emitted mainly in the first few days of operation (Maulini-Duran et al., 2013; González et al., 2019a). Maximum emission rates for NH₃ were detected with S1 (570 mg NH₃· d^{-1}), whereas peak emissions were 90% lower with S2 (58.3 mg NH₃·d⁻¹), and 95.7% lower with S3 (25.8 mg NH₃·d⁻¹). Furthermore, the highest overall NH₃ emission factor was found during S1 (11.5E-01 g NH₃·kg⁻¹TS), which emitted 80.9% and 96.1% more NH₃ than strategies S2 (2.2E-02 g NH₃·kg⁻¹TS) and S3 (4.5E-03 g NH₃·kg⁻¹TS) respectively. Comparatively, those values were always lower than those of the sewage sludge biodrying (2.7E-01 g NH₃·kg⁻¹TS) (González et al., 2019a) and composting processes (values found between 0.4 and 10.95 g NH₃·kg⁻ ¹TS) (Yuan et al., 2016; Han et al., 2018a). Maximum tVOC emission rates found were 107.2, 41 and 80.8 mg C-VOC·d⁻¹ for strategies S1, S2 and S3, respectively. In all cases, those maximum values were detected within the first 48h approximately, later decreasing to barely detectable values. These results coincide with what other studies found during the composting of sewage sludge (Maulini-Duran et al., 2013, González et al., 2019b). The highest tVOC emission factor was found when applying aeration S1 (1.4E-02 g C-VOC·kg⁻¹TS), being 65.7% higher than S2 (4.8E-03 g C-VOC·kg⁻¹TS) and 30.7% higher than S3 (9.7E-03 g C-VOC·kg⁻¹ ¹TS). It is likely that the more adjusted aeration rates used in S1 and in the thermophilic stage of S3, led to an increase in anaerobic spots in the bulk mixture, leading to significant tVOC emissions (Maulini-Duran et al., 2013). Compared to values found in

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literature, there is limited information about tVOC emissions in the biodrying process, and in this regard, only one study was found. All trials in the present study emitted 55-85% less tVOCs than sewage sludge biodrying (3.1E-02 g C-VOC·kg⁻¹TS) (González et al., 2019a).

3.4 Quality assessment of final biodried products obtained

For a complete end-product quality assessment, both mixed and sieved end-products were assessed in the present study and results are presented in Table 4. Although a sustained combustion in a conventional biomass boiler can occur with a MC up to 55% (Navaee-Ardeh et al., 2010), the maximum boiler efficiency is directly dependent on the MC of the product, where efficiency can be upgraded up to 74-80% when reducing the MC below 40% (Gebreegziabher et al., 2013). Nevertheless, 20% of MC was claimed to be the most appropriate value for the pelleting process of Solid Recovered Fuels (SRF) (Rezaei et al., 2020). All mixed products achieved MCs significantly lower than other studies on sludges or SRF (Shao et al., 2010; Cai et al., 2012; Villegas and Huiliñir, 2014; Yasar et al., 2018).

Apart from the MC, LHV is the other key parameter that determines the quality of the biomass fuel produced. It seems that sustained combustion can occur with LHV above 4MJ·kg⁻¹ (Hao et al., 2018). All the biodried mixed products obtained in this study presented LHVs above 9MJ·kg⁻¹, which were equivalent to other conventional biomass fuels used in boilers. The mixed product obtained in S3 reached 10MJ·kg⁻¹

which can be classified into group 4 according to the SRF quality standard (EN 15359).

485 LHVs for all the three mixed end-products were, in general, higher than those found in 486 literature for conventional sewage sludge and pulp and paper mill sludge biodried products (5.5-7.5MJ·kg⁻¹ in the best cases) (Huiliñir and Villegas, 2014; Zhang et al., 487 488 2018). When comparing to MSW biodried products, the results are more variable. Some 489 authors obtained LHVs as high as 21MJ·kg⁻¹ (Tambone et al., 2011), although such 490 high values can be related to their plastic and paper content (Shao et al., 2010). 491 492 The mixed product quality assessment is the most common study that is found in the literature. Nevertheless, bulking materials (normally pruning waste or wood chips) may 493 494 hide or dilute the real values corresponding to the waste streams that are being valorised 495 as biomass fuels, as it is the case in the present study (Table 4). Therefore, in this study, 496 the results corresponding to sieved materials were prioritised. 497 Sieved materials consistently presented higher MC and consequently, lower LHVs than mixed materials. The lowest LHV (5.4 MJ·kg⁻¹) was found in the product obtained 498 when applying S1 and the highest value (7.9 MJ·kg⁻¹) when applying S3. This last value 499 500 was comparable to those obtained in other sewage sludge and paper mill sludge biodrying studies, for the mixed products obtained (Huiliñir and Villegas, 2014; Hao et 501 502 al., 2018; González et al., 2019a). 503 Additionally, the energy production per energy consumed (EP/EC) and the biodrying performance index (BPI) were presented in the current work as suitable indicators for 504 the evaluation of the process by means of end-product quality. Nearly 2 to 3 kWh can 505 506 be recovered from sieved products per each kWh consumed in the process, 507 demonstrating the energetic efficiency of the process in all three cases. Moreover, the 508 new BPI proposed in this work could be used as an overall biodrying efficiency

indicator, facilitating decision making and efficiency comparisons. It considers all the main factors involved in biodrying performance, as well as product quality parameters, thus energy recovery potential of the end-products obtained. The best BPI was achieved when applying S3 (35.1) mainly due to its high specific production ratio.

Comparatively, BPI values for S2 and S1 were 27% and 55% lower than S3 values, respectively.

In general terms and considering all the efficiency indicators described in this work, S3 was considered the best performing strategy.

Additionally, the end-product stability analysis was carried out, indicating that these materials were not totally stable (DRI above 3 $g \cdot kgVS^{-1} \cdot min^{-1}$ and AT4 above 200 $g \cdot kg^{-1}VS$).

Since S3 was considered the best control strategy, the techno-economic analysis presented in Section 3.5 was based on the results and data determined from this trial.

3.5 Techno-economic assessment

An economic model was developed and upscaled based on experimental results obtained from S3, which was the best performing strategy. In this study, only the CS valorisation step though biodrying was considered in the model as an alternative strategy to sludge disposal. An overall economic study of the WWTP after Cellvation® and biomass fuel production through biodrying, would provide a more detailed analysis of the economic viability of this WWTP technological innovation, however, this integrated assessment is out of the scope of the current study. A breakeven point analysis of a hypothetical biodrying plant was performed to find the minimum plant capacity size, in terms of population served, and would indicate the most economically

sustainable scenario. To do so, economic parameters of a biodrying plant were calculated according to the variable mass flow of CS treated, which directly depends on the treatment capacity of the WWTP related to PE served. Table 5 specifies the main economic parameters and financial indicators of the scenarios studied (more detailed information can be found in the supplementary material, Table S1). For the small-scale plants, main OPEX and CAPEX costs were associated to personnel costs and construction of windrows, respectively. For large-scale plants, main CAPEX costs were also related to construction of windrows, while OPEX costs were distributed among electricity, personnel and pelleting costs. In general terms, 53% of the yearly revenues are related to product selling while the rest are due to avoided costs from external sludge management or disposal.

The zero-profit analysis determined that the minimum economically feasible WWTP capacity is >60.000 PE. As an example, according to the Waterbase-UWWTD dataset provided by EEA (EEA, 2020), 56% of Spanish WWTPs, providing services to approximately 95% of the Spanish population, would have enough treatment capacity to guarantee the economic viability of a complementary biodrying plant. This could produce a new source of renewable energy, whilst significantly reducing the waste generated.

The IRR values obtained for WWTP's that provide services to more than 100,000 PE are always above 40%, indicating the economical attractiveness of biodrying processes. Complementarily, payback periods obtained for medium to large WWTP capacity, were between two and five years, achieving valuable benefits (over $100K \in$, yearly) in the case of the largest plants (Table 3).

4. Conclusions

Two new process performance efficiency indices, EBI and BPI, were proposed and their relevance and appropriateness to monitor, assess and compare biodrying processes were confirmed. These two new indicators will contribute to improving the design, monitoring and assessing of current and future biodrying systems.

All three aeration strategies assessed (S1, S2 and S3) showed good performance results and acceptable end-product quality, compared to literature results. Among them, S3 was selected as the best aeration strategy due to the highest BPI values obtained and therefore the highest net energy recovery potential. Moreover, the three aeration strategies used showed low gaseous emissions and, therefore, low environmental impacts are expected. Additionally, promising techno-economic indicators were determined for the best aeration strategy (S3), obtaining an IRR greater than 40% and a payback time of 2 years, for the best-case scenario (medium-large WWTP).

In general terms, biodrying was proven to be an adequate technology to valorise CS in terms of economic and environmental indicators.

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Table 1. Main physico-chemical characteristics of CS and other conventional sludges.

Type of sludge	Total Solids (%, wb)	Volatile Solids (%, db)	N-TKN (%, db)	N-NH4 (%, db)	HHV (MJ kg ⁻	LHV (MJ kg ⁻¹)	pН	CE (mS cm ⁻¹)	DRI (gO ₂ kgVS ⁻ ¹ h ⁻¹)
Cellulosic sludge	25-37	85-93	3-12	1.9-2.5	18-19	2.1-4.9	4.7-6.9	0.5-1.6	2.3-3
Primary sludge Secondary	5-28	60-80	1.5-4				5.6-6.9		
sludge	15-25	52-76	3-6		11-17	0.5-0.9	6.4-7.9		3-7
Mixed sludge	26-38	60-70	2.5-4	0.5-1			5.9-7.1	1.2-1.8	6-7
Anaerobically digested sludge	17-38	53-70	2.6-7	0.7			7.6-7.9	1.2-2.1	1.2-3.7
Pulp & Paper mill sludge	19-26	80-85	0.5-5		18-21		6.2-7.8		

Data gathered from: Navaee-Ardeh et al., 2006; Pagans et al., 2006; Rihani et al., 2010;

⁷³⁹ Bayr and Rintala, 2012; Maulini-Duran et al., 2013; Zhang et al., 2014; Crutchik et al.,

^{740 2017;} Hao et al., 2018; Zhang et al., 2018; Toledo et al., 2019; Da Ros et al., 2020.

Table 2. Air supplied and overall mass balances within the different stages of cellulosic sludge biodrying trials operated with different control strategies. Time is given in days (d) and air supply as an average of the period in m³ per kg of VS fed from CS.*

		Duration		Air supply	Weight loss	Water removal	VS consumption	
		Days	Total m ³	Av. m³kg-¹VS-CS·d-¹	kg (%)	kg (%)	kg (%)	
S1	TOTAL	12.2	128.5	1.4	14.6	11.8 (55.0%)	2.8 (14.9%)	
	TIP	0.6	1.4	0.3	1.2 (8.2%)	1.1 (5.1%)	0.1 (0.6%)	
	THERMOPHILIC ST.	5.4	76.6	1.9	10.4 (71.2%)	8.4 (39.0%)	2.0 (10.8%)	
	MESOPHILIC ST.	6.3	50.7	1.1	3.0 (20.6%)	2.3 (10.9%)	0.7 (3.5)	
S2	TOTAL	13.0	207.4	2.7	13.6	12.3 (62.4%)	1.26 (10.0%)	
	TIP	1.0	2.2	0.4	0.4 (2.9%)	0.4 (1.8%)	0.04 (0.3%)	
	THERMOPHILIC ST.	4.1	101.8	4.2	7.3 (53.7%)	6.8 (34.2%)	0.5 (4.2%)	
	MESOPHILIC ST.	8.0	103.4	2.2	5.9 (43.4%)	5.2 (26.3%)	0.7 (5.4%)	
S 3	TOTAL	13.2	211.6	1.7	12.2	11.0 (57.5%)	1.19 (6.9%)	
	TIP	1.1	2.5	0.2	0.2 (1.6%)	0.2 (0.8%)	0.05 (0.3%)	
	THERMOPHILIC ST.	4.9	64.6	1.4	8.8 (72.1%)	8.0 (41.6%)	0.8 (4.8%)	
	MESOPHILIC ST.	7.2	144.4	2.1	3.2 (26.2%)	2.9 (15.1%)	0.3 (1.8%)	

The mass balances were done according to bulk mixtures, to be consistent with other authors. TIP refers to Temperature Increasing Phase.

⁷⁵¹ The numbers in bold reflect the overall moisture removal ratio and VS consumption ratio of each strategy assessed.

Table 3. Comparison of overall biodrying efficiencies between the current study and other studies for similar high moisture organic wastes

Reference Raw material		Co- substrate	Scale	Specific aeration	Initial MC	Final MC	MC removal ratio	VS consumption ratio	BI	EBI
	(Y/N; which)		$(L \cdot min^{-1} \cdot kgVS^{-1})$	(%)	(%)	(%)	(%)	kg _{H2O} · kg ⁻¹ vs	kg _{H2O} · kg ⁻ 1 _{VS} ·Kwh ⁻¹	
	Cellulosi		Bench	S 1	51.9	35.1	55.0	14.9	4.3	0.62
This study	c sludge	N	(100L)	S2	57.8	32.5	62.4	10.0	9.8	0.99
	c sludge		(100L)	S 3	51.8	31.5	57.5	6.9	9.2	0.85
		Y								
González et al., 2019a	Secondar y sludge	(diatomac eous earth)	Bench (100L)	Variable	54.6	35.9	58.8	14.5	5.7*	
Hao et al., 2018	Dewatere d sewage sludge	Y (Spent Coffee Ground)	Lab (28.3L)	1.37	68.3- 71.6	46.2	79.7	43.5	4.37	
Zhang et al., 2018	Dewatere d sewage sludge	Y (MSW)	Lab (19.44L)	0.49-0.56	70	45.1- 68.3	45.1-78.6	35.1-46.7	3.3-4.6	
Villegas and Huiliñir, 2014	Dewatere d secondar y sludge	N	bench (64L)	1.05-3.14	58	51- 52.5	16.9-24	5-14.3	16-20	

Table 3 cont.

Huiliñir and Villegas, 2014	Pulp and Paper secondary sludge	N	Lab (9L)	0.51-5.26	64.4-65.2	62-45	20-58	0-18	2.5-12.7*	
Winkler et al., 2013	Dewatered sewage sludge	N	Industrial (1900 m ³)	Variable	75	27.4	90.5	26	11.1*	183.6*
Cai et al., 2012	Sewage sludge	N	Pilot (1.6m ³)	Variable	66.1	54.7	46.1			
Sharara et al., 2012	Dairy manure	N	Bench (147 L)	0.05-1.5	55.9	28-35	70.7-79.1	26.3-41.9	2.6-3.2*	24.7-346.7*
Sadaka and Ahn,	Beef manure				59	30	59	8.1	15.5*	0.126*
2012	Swine manure	N	Pilot (0.9 m^3)	0.65	60	28	58	5.8	19.8*	0.08*
2012	Poultry manure				61	40	53	5.9	19.0*	0.11*
Tambone et al., 2011	residual MSW	N	Industrial	Variable	32.7	17.8	65.5	29	2.26*	
Shao et al., 2010	MSW	N	Bench (150L)	1.4	73	48.3	79.9	37.3	7.02*	
Zhao et al., 2010	Dewatered sewage sludge	N	Bench (81L)	3.1-6.1	67.8	30.5-41.9	57.5-68.2	31.0-36.7	5.9-6.1*	
Frei et al., 2004	Pulp and Paper mixed sludge	N	Pilot (1m ³)		52.5-75.5	34.3-59.5	47-53.5	5.5-18	5.9-21.7*	

^{*}Estimated from the values provided in the work

Table 4. Quality assessment parameters of cellulosic sludge biodrying end-products from three different aerations strategies for each sieved and mixed products.

Parameter	S1		S	52	S3		
rarameter	Sieved	Mixture	Sieved	Mixture	Sieved	Mixture	
MC (%, w.b.)	57.3	35.1	51.4	35.5	43.3	31.5	
VS (%, d.b.)	88.7	88.7	85.5	84.7	91.9	94.2	
HHV (<i>MJ</i> · <i>kg</i> ⁻¹ <i>TS</i>)	17.1 ± 0.05	17.1 ±	17.2 ± 16.9 ±		16.9 ±	17.71 ±	
IIII (MJ Ng 15)	17.1 ± 0.03	0.1 0.1		0.3	0.1	0.00	
HHV % lost from initial	9.9	4.2	4.9	3.4	10	6.1	
T TTX (MT 11)	5.4 + 0.02	9.5 ±	6.57 ± 9.4 ±		7.88 ±	10.6 ±	
LHV $(MJ \cdot kg^{-1})$	5.4 ± 0.03	0.2	0.06	0.2	0.07	0.00	
LHV % gained from initial	46.1	27.0	206.9	53.5	60.8	30.5	
Specific production							
ratio (kg TS product·kg	0.65	-	0.81	-	0.87	-	
¹ TS-CS fed							
EP/EC (kWh·kWh ⁻¹)	1.8	-	2.1	-	3.1	-	
BPI (kg _{H2O} · kg ⁻¹ vs)	15.7	-	25.6	-	35.1	-	

Table 5. Economic parameters and financial indicators of variable CS input scenarios. NPV values are given considering a lifetime of 25
 years and considering a discount rate of 7%.

	20K	40K	60K	80K	100K	150K	200K	250K	300K	400K	500K	750K	1000K
CAPEX (€)	20,012	23,878	27,743	33,309	37,664	66,762	99,082	136,888	140,36 8	176,319	246,721	351,540	467,793
OPEX (€ • y -¹)	28,393	32,105	35,663	39,740	43,673	58,595	67,509	82,288	99,608	122,881	148,187	234,227	339,788
REVENUE (€·y ⁻¹)	11,936	23,872	35,808	47,743	59,679	89,519	119,359	149,198	179,03 8	238,717	298,397	447,595	596,794
BENEFITS (€·y ⁻¹)	-16,457	-8,233	145	8,004	16,006	30,924	51,849	66,910	79,430	115,837	150,210	213,368	257,006 €
NPV (€)	- 410,134	- 221,846	- 29,968	143,32 0	323,69 8	645,37 7	1,071,69 6	1,388,017	1,674,8 11	2,489,4 90	3,194,1 70	4,567,2 39	5,440,284
IRR (%)	-	-	-	23	42	46	52	49	56	66	61	61	55
PAYBACK (y)	INF	INF	INF	5	3	3	3	3	2	2	2	2	2

K refers to thousand and INF refers to no payback possibility

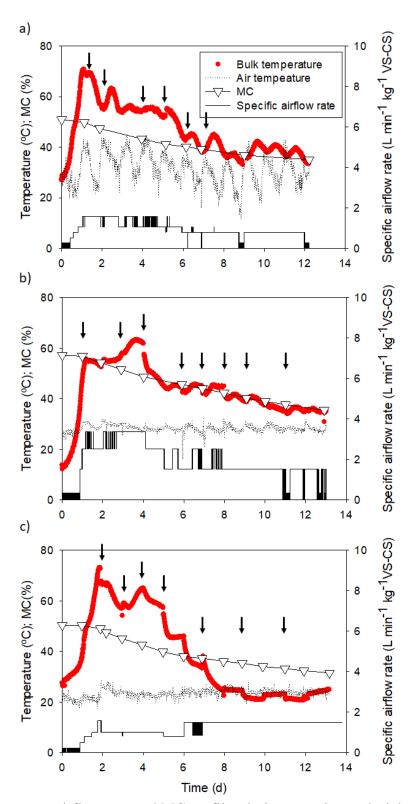


Fig. 1. Temperature, airflow rate and MC profiles during experimental trials for
 strategies S1, S2 and S3, shown in figures a, b and c, respectively. Arrows indicate
 when the mixture was turned.

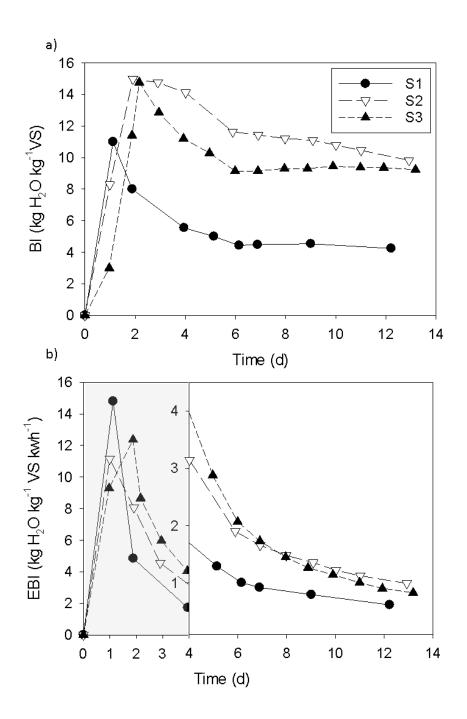


Fig. 2 Daily comparative biodrying performance efficiency indexes: (a) Biodrying Index (kgH₂O kgVS⁻¹) and (b) Energetic Biodrying Index (kgH₂O kgVS⁻¹ kwh⁻¹), the grey area roughly indicates the thermophilic stages during the trials. Axes for the EBI profile differed between the thermophilic stage and the rest of the process, as they were adjusted to the values obtained in each phase.

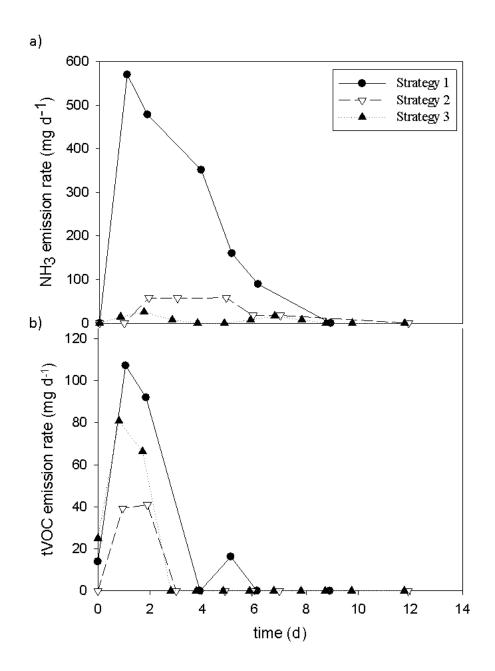


Fig. 3. NH₃ and tVOC emission patterns during the three biodrying aeration strategies
 implemented.