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

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Abstract

This study assesses the technological, environmental and economic feasibility of biodrying to valorise cellulosic sludge as renewable energy source. Specifically, three different aeration strategies were compared in terms of biodrying performance, energetic consumption, gaseous emissions, quality of end-products and techno-economic analysis. The overall drying efficiency of the process was evaluated through 3 different aeration strategies. Those 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) are proposed to better assess the initial and operational conditions that are favouring the maximum energy process efficiency and the highest quality of the end-products. The end-products obtained always presented moisture contents below 40% and lower heating values above 9.4 MJ·kg^{-1}, being the best values achieved, 32.6% and 10.4 MJ·kg^{-1} for moisture content and lower heating value respectively. Low N_2O and CH_4 emissions confirmed the effective aeration during all the three strategies carried out, while the rest of the gases monitored 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, techno-economic analysis.

Abbreviation list:

AT4	4 days cumulative oxygen consumption
B.A.	Bulking Agent
B.I.	Biodrying Index
B.P.I.	Biodrying Performance Index
CAPEX	Capital expenditures
COD	Chemical Oxygen Demand
C.S.	Cellulosic Sludge
D.R.I.	Dynamic Respirometric Index
E.B.I.	Energetic Biodrying Index
E.C.	Energy Consumption
E.P.	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
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

1. Introduction

The recovery of resources (materials and energy) from wastewater is a promising solution that contributes to achieve the relevant sustainability challenges of water utilities and modern societies. There is a wide range of innovative technologies that are being applied that additionally make the wastewater treatment plants more efficient; reducing the amount of sludge produced, reducing the energy consumed and clearly providing environmental and economic benefits (Conca et al., 2020; Da Ros et al., 2020). These new technologies are applied at different stages of the water treatment facilities, 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). Nevertheless, the impact on the overall improvement of the Wastewater Treatment Plant (WWTP) efficiency and energy savings, although it is positive, still has some limitations and margin for improvement.

To increase the WWTP efficiency while increasing the resource recovery capacity, new technologies are developed and applied at the first stages of the main stream (Reijken et al., 2018; Larriba et al., 2020). A part of the direct impacts on resource recovery, these technologies have also an indirect impact on improving the efficiency of the next stages since they significantly reduce the chemical oxygen demand (COD) and total solids (TS) content and consequently, reducing the aeration needs and the amounts of sewage sludge produced that can be translated in important energy savings.

Among these innovative promising technologies, Cellvation® is aiming to maximise the recovery and recycling of cellulose replacing, partially or totally, the primary settler.

Cellvation®, consists of an initial grit and hair removal in a rotating drum filter followed by a 350 µm fine sieve (Salsnes Filter, Norway), followed by a cellpress, a hygienisation step and a pelletiser. Finally, cellulose is recovered 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 directly considered as suitable material for being valorised via energy recovery technologies.

Among the different technologies that could be applied, biodrying is presented as an innovative, energy-saving and environmentally friendly alternative process for CS and sewage sludge energetic valorisation. Biodrying, that can be considered as a composting-like process, 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 is aiming to preserve, in the final biomass fuel produced, most of the organic matter content present in the raw material (Huiliñir and Villegas, 2014).

Biodrying performance is normally assessed using two main indices: the daily drying rate and Biodrying Index (BI). However, theses indices present some limitations, since daily drying rates does not consider the organic carbon biodegraded and BI does not consider the external energy consumed.

CS presents some difficulties over other organic wastes for its valorisation through biodrying, being the most significant its low porosity and high moisture content, which can hamper the proper air diffusion throughout the raw material. This technology has not been optimized yet for low porosity organic wastes and there is still room for the improvement of its performance and efficiency that would importantly increase its possible applications to valorise different types of sewage sludge.

Assuming suitable initial conditions (e.g. organic content of raw materials and 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. In literature, there are some studies assessing the 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 $min^{-1}kg^{-1}$VS-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 optimized 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 maximising the net energy recovery. The key quality indicators of the biodried products obtained are a 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 weaknesses were indeed related to the high MC of final products (Navaee-Ardeh et al., 2006). In addition, 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, there also exists a lack of information in the literature about gaseous emissions during biodrying processes, 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-deep performance assessment of biodrying 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 are proposed to overcome the limitations of the currently used indices and have 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 $m^3 \cdot h^{-1}$ of wastewater. This system reduces the total suspended solids up to 40%, that can be translated into energy savings up to 15% and reduction of sewage sludge production up to 20% (Cellvation, B.V., 2018). The raw material used in this study is a mix of intermediate cellulose-rich flows, the so-called, CS. The main physic-chemical characteristics of CS are presented in Table 1, including a comparison with other conventional sludge produced.

Pruning waste was used as bulking agent (B.A). It was obtained from the MSW composting plant of the Parc Ambiental de Bufalvent located in Manresa, Spain.

Sludge and bulking agent were mixed manually. The mixture ratio used was 1:2.5 of CS to pruning waste, allowing to have a MC and Free Air Space (FAS) within the optimal range, close to 50-60% (Villegas and Huiliñir, 2015) and close to 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 2 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 $L\cdot min^{-1}$) was continuously pumped and analysed using O_2 and CO_2 sensors (O_2A_2 and IRC A_1, 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

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

2.3. Analytical methods

All analysis were made following the Standard Methods for the Examination of Water and Wastewater (APHA, 1995) except for 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 & Huiliñir, 2014). Biological stability, by means of Dynamic Respiration Index (DRI) and 4 days cumulative oxygen consumption (AT4), were determined in a dynamic respirometer developed by Ponsa et al. (2010).

Higher calorific 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 sample between 0,6-1g was electrically ignited in pure oxygen environment (30atm) and afterwards heat of combustion produced, 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 mineralization during biodrying was calculated according to the ash conservation principle (Cai et al., 2012). Accordingly, final Volatile Solids (VS) weight was calculated from VS content of representative products after homogenization and grinding. The VS loss ratio was estimated for every stage (lag, thermophilic and late mesophilic-cooling stages) from the percentage of cumulative O_2 consumption monitored in each stage. Then, those values were used to calculate moisture content removal correcting it from monitored weight 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, the 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 in the BI calculation obtaining two new indices. Hence, those parameters could reflect the energetic, economic and environmental viability of a certain biodrying process performance. Consequently, the new Energetic Biodrying Index (EBI) and Biodrying Performance Index (BPI) are presented (Equation 1 and 2, respectively).

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

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

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 CS treated) and EP is the energy potential production in terms of HHV of sieved product considering its specific production ratio (corrected per dry mass of CS treated).

Additionally, an indicator referred to the mass conservation efficiency, that indirectly measures the VS conservation capacity, is suggested by means of the specific production ratio, as defined in Equation 3.

$$Specific\ production\ ratio = \frac{m_{TS}\ product}{m_{TS-C}\ \ fed} \qquad (Equation\ 3)$$

Referring m_{TS} *product* to the absolute mass of TS contained in the product and $m_{TS\text{-}CS}$ *fed* to the absolute TS mass from CS fed in 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. CH_4 and N_2O analysis were carried out using an Agilent 6890 N Gas Chromatograph (Agilent Technologies, Inc.) equipped with a flame ionization detector and an electron capture detector for CH_4 and N_2O detection, respectively. Total Volatile Organic Compounds (tVOC), NH_3 and H_2S 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 in the CS valorisation step by using biodrying process that produces a biomass fuel with economic value.

Real scale WWTP data were provided by Cirtec B.V. The specific CS production of the WWTP studied was of 1.1E-02 $t \cdot PE^{-1} \cdot y^{-1}$, given total annual CS production of 2,920 tons while serving to 262,000 Population Equivalents (P.E.). 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 biodrying trenches made of concrete including a cover for roofing and aeration system based on blowers. For yearly costs calculation, energy consumption (electricity and diesel), personnel costs, B.A. cost, pelleting, maintenance and insurance costs were estimated. The energetic consumption of equipment was upscaled based on experimental data obtained and adapted to the information provided by the industrial composting plant consulted (Aigües de Manresa S.A., Spain). The market price value of end-products 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 Expendidures (OPEX).

The economical parameters calculated were: CAPEX, OPEX, Revenues, Net Present value (NPV), Internal Rate of Return (IRR) and Payback Period Calculation detail of NPV and IRR are facilitated in Equation 4 and 5.

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

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

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

Payback period was calculated through the sum of annual cash flows over time until a positive value is 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 the three trials are shown in Fig. 1. In general, temperature profiles are comparable to those found in literature regarding sewage sludge biodrying (Zhao et al., 2010). Maximum temperatures achieved were equivalent for S1 and S3 (72ºC and 73.4ºC, respectively) while temperature profiles were roughly similar until the process entered in a late mesophilic stage, when the aeration rate clearly differed. As expected, the temperature profile with S2 was different, reaching 55ºC after 24h and maintained for 2 more days.

After almost 4 days of operation, the maximum temperature peak of 63.5ºC was achieved. Thermophilic temperatures were maintained for 5.3, 4.1 and 4.9 days with S1, S2 and S3, respectively. Compared to what was obtained by other authors working on a similar scale, both maximum temperatures achieved and the length of the thermophilic stage with S1 and S3 were improved in the current study (Zhao et al., 2010; Vilegas and Huiliñir, 2014).

As shown in Fig. 1, airflow rates supplied were significantly different for the three strategies. Use of S2 high airflow rates (up to 3.5 $L \cdot min^{-}1 \cdot kg^{-1}$ VS-CS) probably led to a delayed temperature peak as well as a comparatively higher heat loss after 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, Temperature Increasing Phase (TIP) and Thermophilic stages in S1 followed the same trend than in S3. However, the higher aeration rates used in S3 from day 6 on, significantly affected its temperature profile. Probably, after day 8, the biogenic temperature generation was not able to counterbalance the heat loss due to high aeration rate and consequently temperature decreased to 25ºC and remained constant until the end of the experiment. Accordingly, with S3, it could be assumed that only convective drying occurred after day 8.

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 but at a fixed VS mass consumption, corresponding to the minimum value obtained among the 3 strategies that was found for S3 (1.19 kg VS), the moisture removed applying S2 would still be 38%

and 11% higher than that removed with S1 and S3, respectively, demonstrating the high efficiency of S2.

In terms of water removal efficiency, biodrying performance of CS was generally high in comparison to the values 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 of sludges with other biodegradable wastes is 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, led to the most significant MC removal occurring during this stage, compared to S2. 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 B.A biodegradation negligible, maximum VS consumption from cellulosic sludge (36.7% of initial VS-CS content) occurred when applying aeration S1. In contrast, when applying S3, the lowest VS consumption was determined (12.6% of the initial VS-CS content). Again, when comparing the results at 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, finding maximum absolute values in S1, more than double than in 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 authors 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 the tree strategies implemented in terms of VS conservation, as even the highest VS consumption found for S1 (14.9% of bulk mixture VS) resulted to be lower than most of the other authors obtained (normally between 15 and 40% of VS consumption) (Zhao et al., 2010).

3.2 Process performance assessment

The most relevant performance assessment parameters to assess biodrying processes are: (i) moisture removal; (ii) VS consumption and; (iii) the energy consumption along the process. Moisture removal should be intended to be maximised, while VS consumption must be limited, and energy consumption minimised. The Biodrying Index (BI) is usually reported in the literature as 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 the tree of them, adding an energy consumption parameter into performance efficiency assessment. When the above-mentioned indices are determined daily, they would allow the semi-continuous process performance monitoring and the optimisation of the biodrying process efficiency.

Process monitoring, by means of BI and EBI, for the three compared aeration strategies are shown in Fig. 2a and b, respectively. When comparing the three strategies assessed, the best final BI was obtained when applying S2 (9.8 $kgH_2O \cdot kg^{-1}VS$), followed closely by S3 (9.2 $kgH_2O \cdot kg^{-1}VS$), and finally by S1 (4.3 $kgH_2O \cdot kg^{-1}VS$). Comparatively, the lowest BI obtained for S1 was indeed expected due to its higher VS consumption, which doubled those found for S2 and S3. The limitation of organic carbon mineralization is a key point for the improvement of biodrying performance since it would affect the end-product quality as 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 in 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 authors, all the strategies tested, but 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; Villegas & Huiliñir, 2014). However, some of the authors reported higher BI values (up to 20 $kgH_2O \cdot kg^{-1}VS$) compared to those presented in this study. 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 general, in higher MC removal ratios, but also significantly higher VS consumption values, lowering in these cases the overall BI values (up to 6 $kgH_2O \cdot kg^{-1}VS$) (Hao et al., 2018; Zhang et al., 2018; González et al., 2019a).

Some authors expose that overall water carrying capacity when using high airflow rates should be substantially higher than that achieved due to high temperatures (Sharara et al., 2012). The better drying performance of S2 compared to S3 during late mesophilic stage is probably due to the difference in bulk temperature during that stage. Although airflow rates are equivalent, the under mesophilic bulk 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 ratios, a minimum bulk temperature around 35-40ºC 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.

In overall terms, the most efficient strategy was found to be again S2 (0.99 $kgH_2O·kg^{-1}VSkwh^{-1}$), followed closely by S3 (0.85 $kgH_2O·kg^{-1}VS·kwh^{-1}$).

Conversely and although it was the one with the lowest overall energy consumption, S1 obtained the lowest EBI value (0.62 $kgH_2O·kg^{-1}VS·kwh^{-1}$), particularly due to the high VS consumption, which did not actually improve moisture removal efficiency.

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

Nevertheless, energy consumption data per water removed are more favourable, 0.4-0.9 $kwh·kg^{-1}H_2O$ in the present study vs. 2.2-2.5 $kwh·kg^{-1}H_2O$ obtained in Sharara et al., (2012), 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 shows promising results even considering that it can be further optimised during the mesophilic-cooling stage. The information provided by the indices proposed together with different aeration strategies used would certainly facilitate the upscaling of the biodrying process.

3.3 Gaseous emissions

3.3.1 GHG emissions

Regarding Greenhouse Gases (GHG), maximum CH_4 and N_2O emission rates were found during the first hours (0h for N_2O and 24h for CH_4) of the process. These maximum emissions were probably related to anaerobic conditions during the dewatering and shipping of raw materials (Han et al., 2018a). In fact, after adjusting the initial structure, porosity and moisture content of the material, N_2O stored in sludge was probably stripped-out by forced aeration (Han et al., 2018a; González et al., 2019a) and it was not detected anymore. CH_4 emissions have been related to an inadequate mixture structure 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 the worst-case scenario, maximum daily emission rates found for N_2O and CH_4 were 91.8 $mg \cdot d^{-1}$ and 16.3 $mg \cdot d^{-1}$, respectively. Additionally, the overall emission factors calculated for N_2O and CH_4 were 6.8E-03 $gN_2O \cdot kg^{-1}TS$ and 2.6E-03 $gCH_4 \cdot kg^{-1}TS$, which were lower than values reported in biodrying and composting literature (Han et al., 2018a; González et al., 2019a). Regarding the global warming effect, the maximum cumulated value was 2.13 g $CO_2eq \cdot kg^{-1}TS$, corresponding to S1. This value is almost 3 times lower than the values reported in conventional sewage sludge biodrying (González et al., 2019a) and even significantly lower than those of sewage sludge composting (Yuan et al., 2016; Han et al., 2018a).

3.3.2 H_2S, NH_3 and total VOC emissions

In aerobic degradation processes such as composting or biodrying, H_2S, NH_3, and tVOCs are the main compounds related to unpleasant odour emissions, which are recognised to be a significant weakness of those processes (Han et al., 2018a). Emission profiles of NH_3 and tVOCs are shown in Fig. 3a and b, respectively. H_2S was never detected, reinforcing the effective aerobic conditions of biodrying mixtures with all the aeration strategies implemented (Han et al., 2018b). NH_3 and tVOC emissions followed a typical profile where maximum NH_3 emission peaks were related to thermophilic temperatures whereas tVOCs were emitted mainly in the first days of operation (Maulini-Duran et al., 2013; González et al., 2019a). Maximum emission rates for NH_3 were detected with S1 (570 mg $NH_3 \cdot d^{-1}$), whereas peak emissions were 90% lower with S2 (58.3 mg $NH_3 \cdot d^{-1}$), and 95.7% lower with S3 (25.8 mg $NH_3 \cdot d^{-1}$). Furthermore, the highest overall NH_3 emission factor was found during S1 (11.5E-01 g $NH_3 \cdot kg^{-1}$TS), which emitted 80.9% and 96.1% more NH_3 than strategies S2 (2.2E-02 g $NH_3 \cdot kg^{-1}$TS) and S3 (4.5E-03 g $NH_3 \cdot kg^{-1}$TS) respectively. Comparatively, those values were always lower than those of the sewage sludge biodrying (2.7E-01 g $NH_3 \cdot kg^{-1}$TS) (González et al., 2019a) and composting processes (values found between 0.4 and 10.95 g $NH_3 \cdot kg^{-1}$TS) (Yuan et al., 2016; Han et al., 2018a).

Maximum tVOC emission rates found were 107.2; 41 and 80.8 mg C-VOC$\cdot d^{-1}$ for strategies S1, S2 and S3, respectively. In all cases, those maximum values were detected in the first hours (48h approximately), later decreasing to barely detectable values. These results are in line with what other authors found during 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$\cdot kg^{-1}$TS), being 65.7%

and 30.7% higher than S2 (4.8E-03 g C-VOC·kg^{-1}TS) and S3 (9.7E-03 g C-VOC·kg^{-1}TS), respectively. Probably, the more adjusted aeration rates used in S1 and in the thermophilic stage of S3, could lead to increase the anaerobic spots in the bulk mixture, leading to significant tVOC emissions (Maulini-Duran et al., 2013). Compared to values found in literature, there is limited information about tVOC emissions in biodrying process and in this regard, only one work was found. All trials in the present study emitted 55-85% less tVOCs than sewage sludge biodrying (3.1E-02 g C-VOC·kg^{-1}TS) (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, upgrading such efficiency up to 74-80% when reducing the MC below 40% (Gebreegziabher et al., 2013). Besides, 20% of MC was claimed to be the most appropriate value for pelleting process of Solid Recovered Fuels (SRF) (Rezaei et al., 2020). All mixed products achieved MCs significantly lower than other authors working with 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 from LHV above 4MJ·kg^{-1} (Hao et al., 2018). All the biodried mixed products obtained in this work presented LHV above 9MJ·kg^{-1}, being equivalent to other conventional biomass fuels used in boilers while the mixed product obtained when applying S3 can be classified into group 4 according to the SRF quality standard (EN 15359). LHV determined for all the three mixed end-products are, in general, higher than those found in 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., 2018).

When comparing to MSW biodried products the results are more variable. Some authors obtained LHVs as high as 21MJ·kg^{-1} (Tambone et al., 2011), although such high values can be related to their content of plastics and papers (Shao et al.,2010).

The mixed product quality assessment is the most usual study that is found in the literature. Nevertheless, bulking materials (normally pruning waste or wood chips) are hiding or diluting the real values corresponding to the waste streams that are being valorised as biomass fuels, as it is the case in the present study (Table 4). Therefore, in this study, the results corresponding to sieved materials will be prioritised.

Sieved materials presented always higher MC and consequently, lower LHV than mixed materials. The lowest LHV of 5.4 MJ·kg^{-1} was found in the product obtained when applying S1 and the highest value (7.9 MJ·kg^{-1}) when applying S3. This last value was comparable, to those obtained in other sewage sludge and paper mill sludge biodrying studies but considering the mixed products obtained (Huiliñir and Villegas, 2014; Hao et al., 2018; González et al., 2019a).

Additionally, the energy production per energy consumed (EP/EC) and the biodrying performance index (BPI) are presented in the current work as suitable indicators for the evaluation of the process by means of end-product quality. Almost 2 and 3 kwh can be recovered from sieved product per each kwh consumed in the process, demonstrating the energetic efficiency of the process in all the three cases. Moreover, the new BPI proposed in this work could be used as an overall biodrying process efficiency indicator, facilitating the decision making and comparison of process efficiencies, as it considers all the main factors involved in the biodrying performance, including the quality, in terms of 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 when applying 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-economical assessment

An economic model was developed and upscaled based on experimental results obtained from S3, that it is considered the best performing strategy. In this study, only CS valorisation step though biodrying was considered in the model as alternative strategy to sludge disposal. The overall economic study of the WWTP after the implementation of Cellvation® followed by the biomass fuel production through biodrying would provide a more detailed analysis of the economic viability of this technological innovation in a WWTP, however, this integrated assessment is out of the scope of this current work. A breakeven point analysis of a hypothetic biodrying plant was performed to find the minimum plant capacity size, in terms of population served, which would lead to an economically sustainable scenario. To do so, economic parameters of a biodrying plant were calculated according to variable mass flow of CS treated, that 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 supplementary material, Table 1S). For the smallest scale plants main OPEX and CAPEX costs were associated to personnel costs and construction of windrows respectively. For largest 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.

* K refers to thousand

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), the 56% of Spanish WWTP, providing services to approximately 95% of total Spanish population, would have enough treatment capacity to guarantee the economic viability of a complementary biodrying plant producing a new source of renewable energy and significantly reducing the waste generated.

The IRR values obtained for WWTP providing 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 2 and 5 years, achieving worthy benefits (over yearly 100K €) in the case of the largest plants (Table 3).

4. Conclusions

Two new process performance efficiency indexes, EBI and BPI, were proposed and their relevance and appropriateness to monitor, assess and compare biodrying process was confirmed. These two new indicators will contribute to better design, monitor and assess current and future biodrying systems.

All three aeration strategies assessed (S1, S2 and S3) showed good performance results and acceptable quality of the end-products obtained, 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 IRR higher than 40% and payback time of 2 years, for the best-case scenario (medium-large WWTP).

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

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Type of sludge	Total Solids (%, wb)	Volatile Solids (%, db)	N-TKN (%, db)	N-NH4 (%, db)	HHV (MJ kg^{-1}TS)	LHV (MJ kg^{-1})	pH	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	5-28	60-80	1.5-4				5.6-6.9		
Secondary 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		

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

Data gathered from: Navaee-Ardeh et al., 2006; Pagans et al., 2006; Rihani et al., 2010; Bayr & Rintala, 2012; Maulini-Duran et al., 2013; Zhang et al., 2014; Crutchik et al., 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 in 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 m^3 per kg of VS fed from CS.

		Duration	**Air supply**		**Weight loss**	**Water removal**	**VS consumption**
		Days	**Total m^3**	**Av. $m^3kg^{-1}VS\text{-}CS\cdot d^{-1}$**	**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%)
S3	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 is referred to Temperature Increasing Phase

Table 3 Comparative biodrying overall efficiencies in literature for similar high moisture organic wastes.

Reference	Raw material	Co-substrate (Y/N; which)	Scale	Specific aeration (L ·min^{-1}·kgVS^{-1})	Initial MC (%)	Final MC (%)	MC removal ratio (%)	VS consumption ratio (%)	BI kg $_{H2Or}$· kg$_{VSc}^{-1}$	EBI kg $_{H2Or}$· kg$_{VSc}^{-1}$·Kwh^{-1}
This study	Cellulosic sludge	N	Bench (100L)	S1	51.9	35.1	55.0	14.9	4.3	0.62
				S2	57.8	32.5	62.4	10.0	9.8	0.99
				S3	51.8	31.5	57.5	6.9	9.2	0.85
González et al., 2019a	Secondary sludge	Y (diatomaceous earth)	Bench (100L)	Variable	54.6	35.9	58.8	14.5	5.7*	
Hao et al., 2018	Dewatered 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	Dewatered 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	Dewatered secondary 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 & Ahn, 2012	Beef manure	N	Pilot (0.9 m^3)	0.65	59	30	59	8.1	15.5*	0.126*
	Swine manure				60	28	58	5.8	19.8*	0.08*
	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

Parameter	**S1**		**S2**		**S3**	
	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 (*MJ·kg^{-1} TS)*	17.1 ± 0.05	17.1 ± 0.1	17.2 ± 0.1	16.9 ± 0.3	16.9 ± 0.1	17.71 ± 0.00
HHV % lost from initial	*9.9*	*4.2*	*4.9*	3.4	10	*6.1*
LHV (*MJ·kg^{-1})*	5.4 ± 0.03	9.5 ± 0.2	6.57 ± 0.06	9.4 ± 0.2	7.88 ± 0.07	10.6 ± 0.00
LHV % gained from initial	*46.1*	*27.0*	*206.9*	53.5	60.8	*30.5*
Specific production ratio (kgTS product·kg^{-1} TS-CS fed	*0.65*	-	*0.81*	-	0.87	-
EP/EC (kwh·kwh^{-1})	1.8	-	2.1	-	3.1	-
BPI	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 €	140.368 €	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 €	179.038 €	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 €**	**143.320 €**	**323.698 €**	**645.377 €**	**1.071.696 €**	**1.388.017 €**	**1.674.811 €**	**2.489.490 €**	**3.194.170 €**	**4.567.239 €**	**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

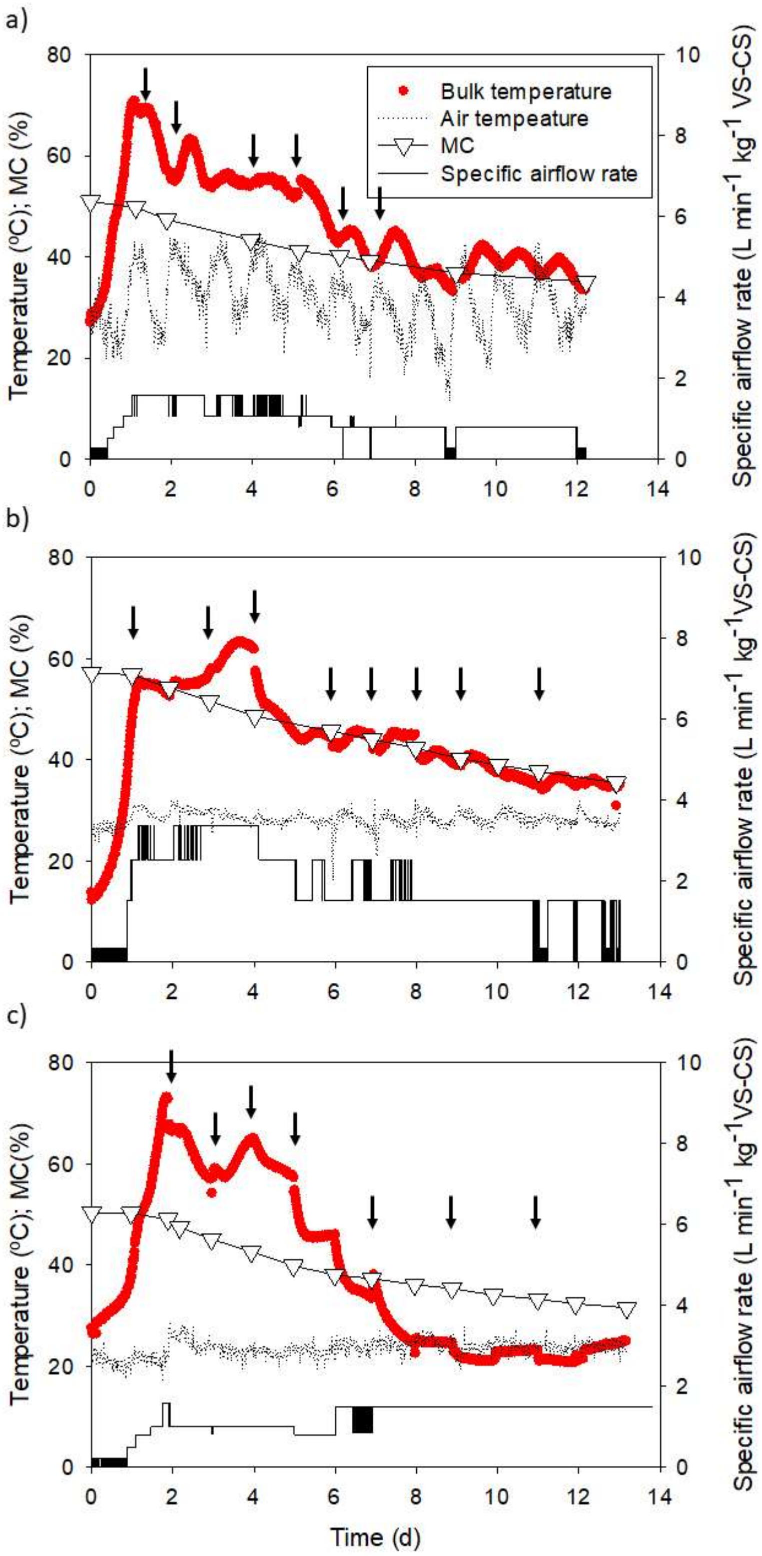

Fig. 1 Temperature, airflow rate and MC profiles during experimental trials implementing strategies S1 , S2 and S3 are shown in figures a, b and c, respectively. Arrows indicate whenever mixture was turned.

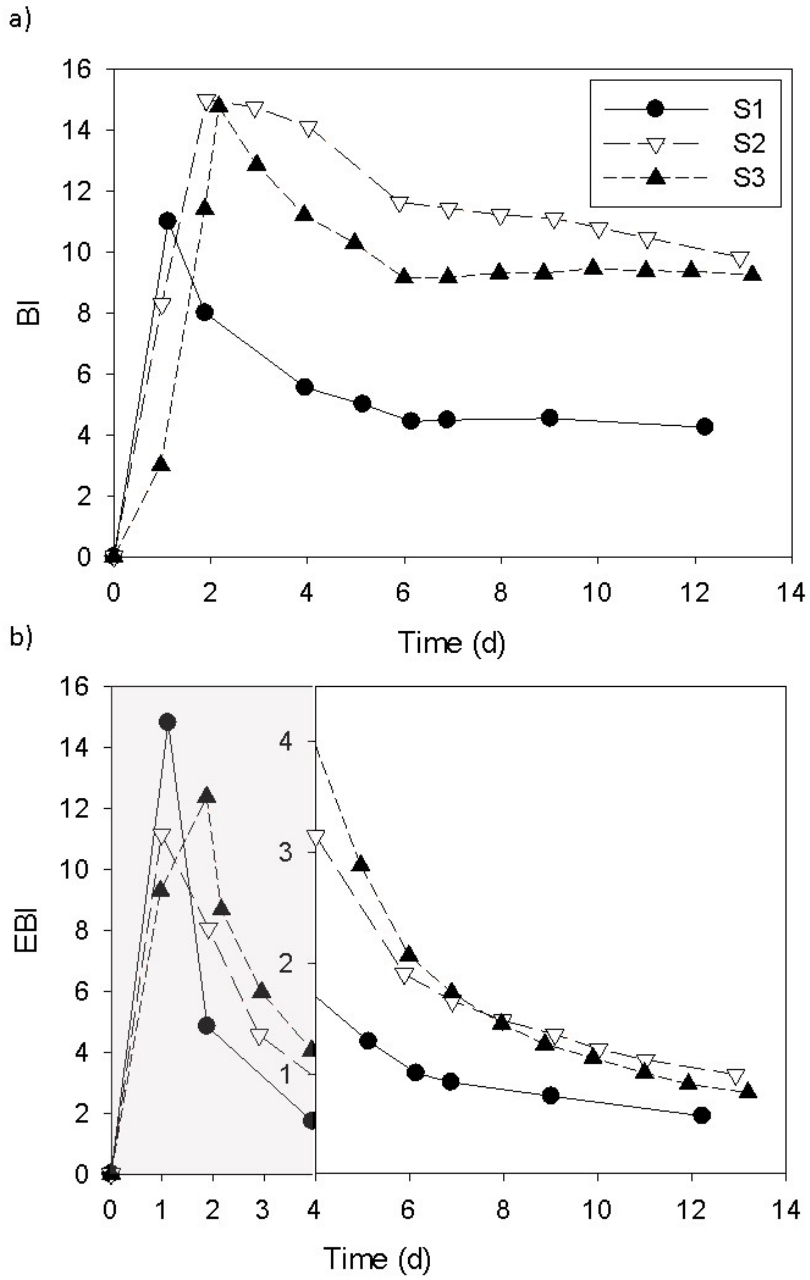

Fig. 2 Daily comparative biodrying performance efficiency indexes: (a) Biodrying Index ($kgH_2O\ kgVS^{-1}$) and (b) Energetic Biodrying Index ($kgH_2O\ kgVS^{-1}\ kwh^{-1}$), the grey area indicates roughly thermophilic stages during trials. Axes for thermophilic and the rest of the process of EBI profiles differ as they were adjusted to the values obtained in each phase.

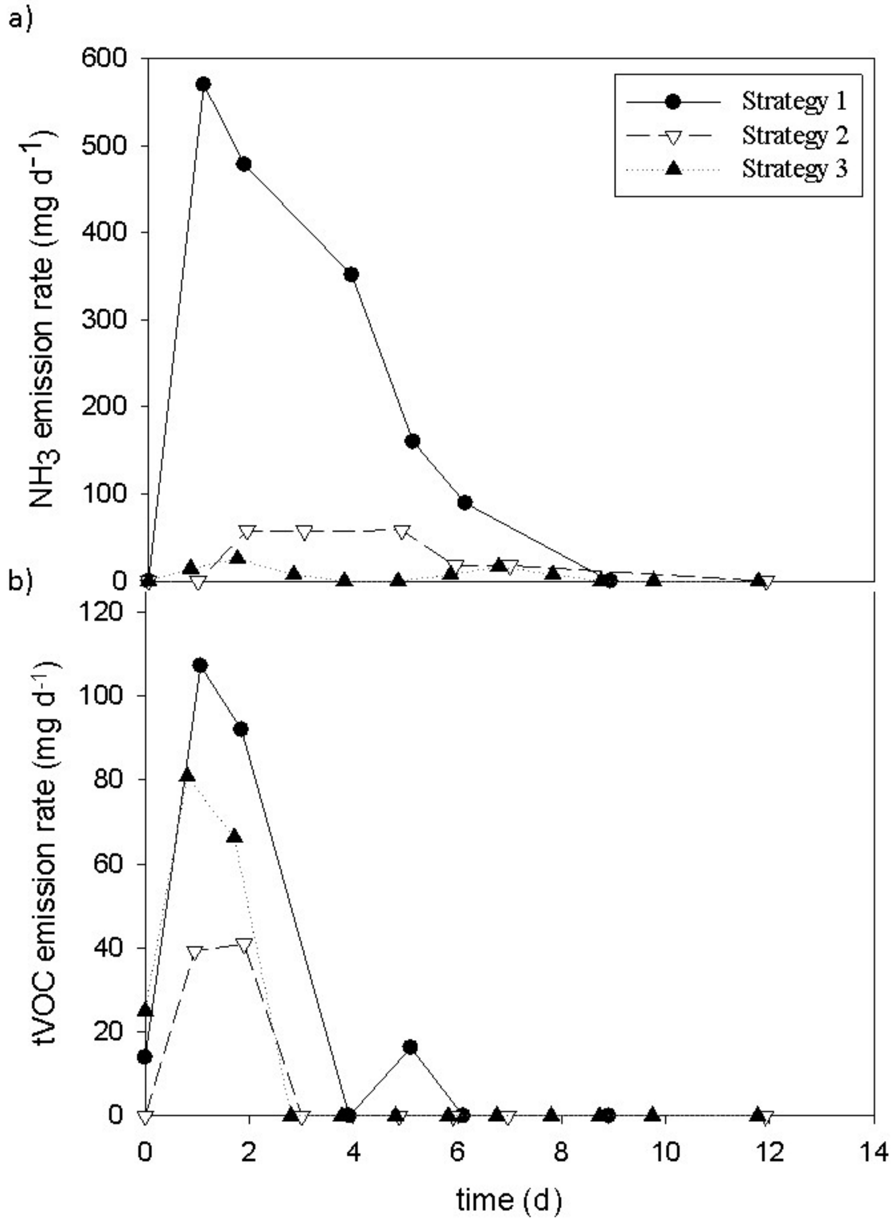

Fig. 3. NH_3 and tVOC emission patterns during the three biodrying aeration strategies implemented.

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