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ORIGINAL PAPER

Gaseous Emissions During Composting of Biowaste Digestate with Different Biochar Ratios at Laboratory Scale

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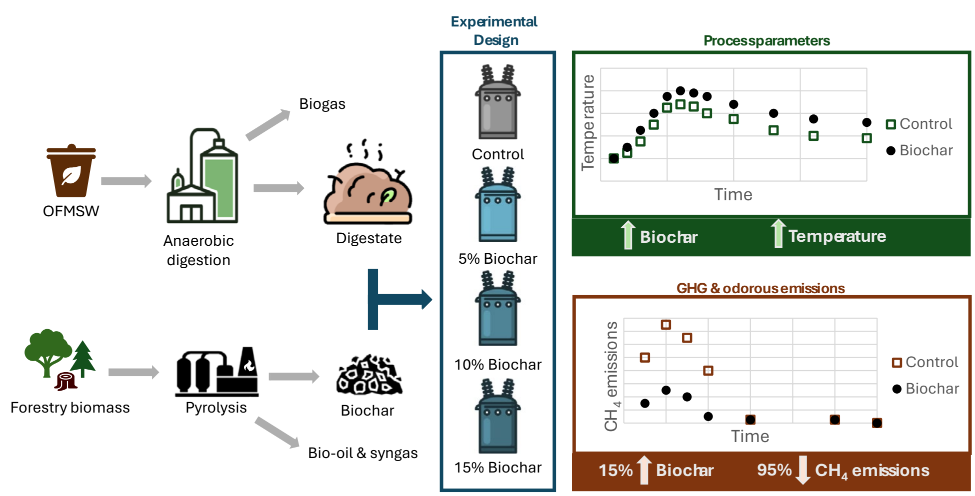
Abstract

Digestate from organic source-selected municipal solid waste is a material that has experienced an exponential growth in its production, due to the current boost of anaerobic digestion. Composting is an environmentally-friendly technology required by EU legislation for the proper management and valorisation of this waste prior to its application to soil as organic amendment. Although some studies have examined the impact of biochar on the composting of raw organic waste and its resulting emissions, the effects of biochar on the composting of low-biodegradability organic waste, such as digestate, have rarely been investigated. In this work, biochar from forestry biomass was tested at different application ratios as a material to minimize the gaseous emissions from biowaste digestate composting, including: ammonia, methane, volatile organic compounds (VOC) and odours. Biochar composting ratios were 5%, 10% and 15% (based on digestate dry matter) together with a control treatment without biochar at bench scale (10 L) during the typical times used in industrial scale digestate composting (12 days). The addition of biochar resulted in a reduction of up to 95% of methane, 60% of total VOC and 70% of odour units emissions, being these numbers obtained with the highest application ratio of biochar (15%). On the contrary, ammonia emissions increased significantly threefold with 5% biochar, according with the high process temperature reached. In conclusion, biochar has the potential to reduce the emission of certain pollutant gases during digestate composting. However, further large-scale experiments are needed to investigate the effect of composting mass on temperature and, consequently, the quantity and composition of gaseous emissions.

Graphical Abstract



Keywords Biochar · Composting · Digestate · Greenhouse emissions · Odours · Volatile organic compounds

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Introduction

In the current scenario of municipal solid waste treatment and valorisation, biological technologies have adopted a worldwide predominant role when dealing with the organic fraction that this waste stream contains [1]. From the environmental perspective, several studies have highlighted the benefits of anaerobic digestion (and subsequent composting of the digestate) when compared to other strategies implemented for waste management, using complete life cycle assessments tools [2–4]. Additionally, configurations where anaerobic digestion and composting (in that order) coexist have been identified as the best options to generate profit from the organic fraction of municipal organic solid waste through the production of locally sourced renewable energy and a well-stabilised mature compost [5, 6].

If this strategy for the source-selected organic fraction of municipal solid waste (OFMSW or biowaste) is chosen, the composting process is, inherently, different from that for raw OFMSW [7]. As the composting substrate has been anaerobically digested (therefore called digestate), its biodegradability is significantly lower, whereas the content of ammonia forms of nitrogen is higher [8, 9]. This results in a significantly shorter composting process (around 12 days at commercial scale) and a different profile regarding the gaseous emissions, which is of especial relevance for methane and ammonia emissions, among others [10]. Although gaseous emissions from composting of other typologies of organic waste have been widely studied [11], there are few studies on the biowaste digestate composting emissions. This knowledge gap is particularly relevant, since anaerobic digestion is currently experiencing a worldwide boost due to ongoing geopolitical developments that have an impact on easily available sources of energy [12].

Simultaneously to the implementation of biological treatments for biowaste, the use of biochar in a wide range of environmental technologies is experiencing an extraordinary expansion, including wastewater treatment, anaerobic digestion or soil bioremediation, among others [13]. Biochar is a carbon-rich solid generated from the pyrolysis of waste biomass (using typically agricultural and forestry residues) [14]. Biochar has been proposed as a way to mitigate climate change through long-term carbon preservation [15, 16]. Besides, the role of biochar in the mitigation of gaseous emissions from the composting pile has also been recently reviewed [17, 18].

Biochar's impact on the mitigation of several gases, such as ammonia and methane, during composting has been well-documented [19, 20], through a mechanism similar to that of active carbon [21]. Reductions up to 50% in ammonia or methane emissions have been reported during composting a variety of organic wastes [22]. Recently, the role of direct interspecies electron transfer (DIET) has been also highlighted as a potential reason why biochar decreases the emission of other gases such as nitrous oxide [23], although some other phenomena as changes in redox potential may occur [24]. Studies on the impact of biochar on the release of volatile organic compounds (which include a large number of compounds and families) during composting are very limited [25, 26]. Finally, the effect of biochar on odour emissions during composting has been scarcely treated in scientific literature, although some works can be found with mixed municipal solid waste and biowaste [27, 28], but not with biowaste digestate, which is expected to have a different behaviour in terms of odours, because of the chemical composition and the low biodegradability level of organic matter [9].

Therefore, the objective of this research is to evaluate the gaseous emissions generated in the composting of biowaste digestate at a laboratory scale when applying different ratios of biochar, particularly ammonia, methane, nitrous oxide, volatile organic compounds and odours. The effect of biochar on the stability of digestate has been also studied through respiration techniques.

Materials and Methods

Biochar

Biochar was obtained from the high-temperature pyrolysis (700 °C) of forestry biomass. Table 1 shows the main characteristics of biochar.

The specific surface area, the average pore size (Table 1) and the scanning electron microscope images of biochar (Fig. S1, Supplementary Information) reveal that it is a very porous material regarding both macro (< 2 nm) and microporosity (> 50 nm) [29]. Biochar was

Table 1 Characterization of biochar used in this study

Parameter (units)	Value
Bulk density (kg/m^3)	348
Moisture (%, weight basis)	23.2
pH	8.7
Electrical conductivity (µS/cm)	230
Specific surface area (m^2/g)	7.78
Average pore size (nm)	6.40
C (%, w/w)	77.42
H (%, w/w)	2.41
N (%, w/w)	0.70
S (%, w/w)	< 0.1
H/C$_{org}$ (%, w/w)	0.37

also analysed with pyrolysis GC–MS and random reflectance showing an inert fraction inertinite-like of 100%. Therefore, it is safe to assume that the difference between the starting material in each reactor is non-existent, as it is a homogeneous material.

Supplementary Information (Table S1 and S2) includes biochar production information and different SEM (Scanning Electron Microscope) images of biochar (Fig. S1). The characterization analyses for biochar were carried out in an external laboratory: SAQ (*Servei d'Anàlisi Química de la UAB*, https://www.uab.cat/ca/serveianalisi-quimica). Microscopic images of biochar were taken using a Scanning Electron Microscope (Merlin FE-SEM). Surface area and pore size were also measured after degassing the samples at 80 °C for 20 h and analyzed with nitrogen adsorption/desorption at 196 °C using a BET analyzer (ASAP 2020 Micrometrics Inc.). Quantitative determination of C, H, N and S in solid samples was determined by combustion of biochar samples at 1200 °C in an oxygen atmosphere and further quantification by Gas Chromatography (CHNS Thermo Scientific Flash 2000 Elemental Analyzer). Semi-quantitative estimation of inorganic elements was carried out by Inductively Coupled Plasma Mass Spectroscopy (ICP-MS) using an Agilent (model 7900) equipment. Surface functional groups were analyzed using a Fourier transform Infrared (FTIR) spectrometer (Alpha II Bruker, United States) with a Diamond Crystal ATR (Attenuated Total internal Reflectance) accessory. The biochar samples were placed directly on the diamond crystal plate, and the transmittance data were analyzed with Opus 7.8 software (Bruker, United States).

Biowaste Digestate

Biowaste digestate was collected from the municipal solid waste treatment plant located in Granollers (Barcelona, Spain). This plant annually treats 40,000 tons of source-selected organic fraction of municipal waste. The material is pre-treated to remove impurities and fed to two one-stage, mesophilic anaerobic digesters of 3000 m^3 of capacity. Digestate is filtered by pressure until a cake of 25% of dry matter is obtained. A complete characterization of the biowaste digestate can be found at Table 2, showing a low standard deviation in the main parameters that affect composting (moisture, organic matter, pH, etc.), which is characteristic of a highly homogeneous material. In fact, digestate can be considered homogenous, contrarily to the raw organic fraction of municipal solid waste coming from source-selection street collection schemes, since anaerobic digestion causes the loss of the labile organic matter, as can be confirmed by a low respiration index.

Table 2 Characterization of biowaste digestate used in this study. Deviation values correspond to the variability of two digestate independent samples collected at different times. DM: dry matter

Parameter (units)	Value
Moisture (%)	71.9 ± 0.7
Organic Matter (%, DM basis)	66.3 ± 0.7
Bulk density (kg/m^3)	0.58 ± 0.2
pH	8.75 ± 0.02
Electrical conductivity (μS/cm)	2.69 ± 0.2
Dynamic respiration index (g O_2 g^{-1} DM h^{-1})	1.9 ± 0.5
C/N ratio	13.44
Total N (%, DM basis)	3.69
Organic N (%, DM basis)	2.48
N-NH_4 (%, DM basis)	1.21
P (%, DM basis)	0.939
K (%, DM basis)	0.686

Composting Mixtures and Experimental Setup

Biochar was sieved to a particle size in the range of 0.7 and 1.6 mm for all treatments. This size range was selected to achieve a balance between fine and big biochar particles, as fine particles are difficult to manage and they create airborne dust, and coarse particles can reduce the mixture homogeneity. No additional pretreatment was applied to biochar, which was then mixed with biowaste digestate at different ratios of 5%, 10% and 15% on dry matter basis (for this, only the digestate is considered, not the bulking agent). Thereafter, wood chips were used as bulking agent to the mixture to provide porosity to the composting starting material. Wood chips were previously sieved to a size larger than 7 mm to obtain a proper porosity. A 1:1 ratio of bulking agent was selected (on a volumetric basis) according to Gea et al. [30].

A quasi-adiabatic 10-L composting reactor Fig. S2 was filled with the mixture biochar/biowaste digestate/bulking agent at different ratios of biochar, as explained above. A treatment without biochar was used as control. The total composting mass of the reactor was ca. 1.7–1.8 kg of mixture. Briefly, the reactor is a 10-L Dewar® vessel adapted to composting experiments. It is force-aerated from the bottom with humidified fresh air with a constant flow of 300 mL/min. Temperature sensors (standard Thermochron iButton device, Maxim Integrated, U.S.) were placed at the bottom, middle and top of the reactor to monitor the evolution of the process temperature. Oxygen profile was obtained treatment using an electrochemical oxygen sensor located at the reactor exhaust gas outlet (Alphasense O2-A2, UK). With this level of aeration, all treatments were over 12% of oxygen concentration, which ensure the prevalence of aerobic conditions. Each treatment lasted 12 days, as for the full-scale plant.

Gaseous Collection and Sampling

Detailed information about sampling and gas analysis from composting and other environmental facilities can be found in previous papers, for ammonia and VOC [11] and for methane, nitrous oxide and odours [31]. Specifically, for these bench-scale composting treatments, the sampling methodology was adapted to monitor gaseous emissions at low scale composting, similarly to that of home and community composting [32]. Briefly, Nalophan® sampling bags connected to the exhaust gas of the composter were used to obtain the gaseous samples, which were later analysed for ammonia, methane, total Volatile Organic Compounds (VOCs) and odour concentration (in European odour units). For the characterization of specific VOCs, stainless steel sorbent tubes packed with two different hydrophobic sorbents (Tenax® TA and Carbograph™ 1TD, Markes International, Inc., Gold River, CA, USA) were used as detailed by the USEPA TO-17 method [33] and reported elsewhere [11].

Nitrous oxide was measured given its expected presence during the composting process [5, 34], however, during the 12 days of experiment the concentrations were extremely low, close to the detection limit of the analytical method used (1 ppm). In consequence, these results are not shown, and its effect was not considered.

Gaseous emissions are presented as daily specific emission rates (mass of pollutant per dry matter of biowaste digestate treated per time) together with the corresponding emission factors (mass of pollutant per dry matter of biowaste digestate treated). These units, as opposed to concentrations [27], allow the comparison with other systems, as gaseous emissions from composting largely depend on the composting matrix and system, the aeration regime and, more specifically, on temperature.

Analytical Methods for Gaseous Compounds

A detailed description of the analytical methods used to determine the concentrations of ammonia, methane, total and individual VOCs and odour units can be found at González et al. [32]. Briefly, total VOC and ammonia concentrations were measured with a MiniRAE 3000 portable analyser (RAE Systems, San José, CA, USA), equipped with a 10.6 eV PID lamp for VOC measurement (detection range 0–15,000 ppmv with 0.1 ppmv increments) and an iBrid™ MX6 multi-gas monitor (Industrial Scientific Co., Pittsburgh, PA, USA) equipped with an electrochemical sensor for ammonia (detection range 0–500 ppmv with 1 ppmv increments), respectively.

The analysis of methane and nitrous oxide was performed using gas chromatography (GC): an Agilent 6890N GC equipped with a flame ionization detector (FID) for methane, and an Agilent 8860 GC equipped with an electron capture detector (ECD) for nitrous oxide. A HP-PLOT Q semi-capillary column was used for both gases' quantification using the appropriate standards.

VOC individual characterization was based on González et al. [11], using a UNITY-2 thermal desorber (Markes International, Inc.) to desorb the VOCs retained in the sorbent tubes (Sect. "Gaseous Collection and Sampling") and an Agilent 7820 GC coupled to an Agilent 5975 Mass Spectrometer (MS) for the separation and analysis of the different VOC species. An Agilent DB-624 capillary column was used. The Agilent 5975 MS acquired data in scan mode with m/z interval ranging from 35 to 355 amu. Wiley275 mass spectra library® was used to identify the different VOCs based on their mass spectra, with a minimum match quality of a 90%.

Odour analysis was performed using a Scentroid SM-100 portable field dynamic olfactometer (IDES Canada, Inc., Whitchurch-Stouffville, ON, Canada), as previously described [11]. The olfactometer is factory-calibrated, with a calibration range from 1.9 to 4288.9 ou m^{-3}. Samples were analysed in triplicate by the same panellist in a clean, closed, and well-ventilated lab within the next 24 h after sampling. This measure is expensive and time consuming and, in this work, only some punctual determinations could be conducted (Fig. 4).

Analytical Methods for Composting Solid Samples

Dynamic Respiration Index

Dynamic respiration index (DRI) was used to determine the biological stability of compost. Details of DRI determination can be found elsewhere [35]. DRI was calculated as the average oxygen uptake rate measured during the 24 h of maximum activity. A dynamic respirometer at constant temperature (37 °C) with constant airflow was used [36]. DRI was expressed as g of oxygen consumed per kg of dry matter (DM) and per hour (g O_2 kg^{-1} DM h^{-1}). All measurements were conducted in triplicates.

Routine Analytical Methods

Typical composting parameters such as dry matter and organic matter content, pH and electrical conductivity were determined according to standard methods [37].

Statistics

Statistical analysis was based on one-way ANOVA ($p < 0.05$ confidence) with the Tukey test. Significant differences were analysed using the software package Minitab 16 (Minitab Inc.).

Results

Composting Performance

The process of biowaste digestate composting presents several important differences when compared to that of raw biowaste composting in terms of general performance. Complete temperature profiles are presented in Fig. 1 (average values for each biochar percentage). The dosage of biochar 15% shows a significant temperature increase in comparison to the other treatments, particularly in the temperature peak. The standard deviation was averaged to be below 2,6 °C for all treatments, thus showing a homogeneous behaviour across the reactor.

Another critical point related to any composting process is the level of stability of the end product, which is measured through DRI, a consistent measure of biological stability [38]. Biowaste digestate presented a DRI of 1.9 ± 0.6 g O_2 kg^{-1} DM h^{-1}, with a relatively high dispersion of values that is typically found in the anaerobic digestion of biowaste.

Gaseous Emissions

Ammonia

Ammonia is the main compound in the exhaust gas from composting, excluding carbon dioxide and water vapour [39]. The control experiment shows clearly a lower emission rate than the biochar amended treatments. The highest emission rates are observed during the first 5 days with the 5% biochar addition. A peak for the 10% addition of biochar is observed after 7 days of the experiment, with similar values as the previously mentioned (Fig. 2).

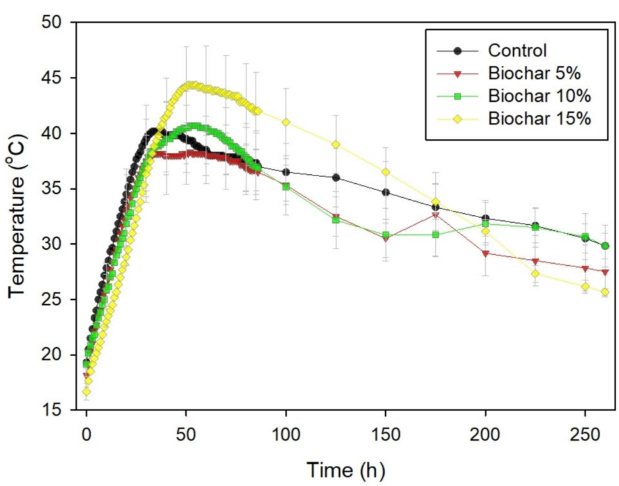

Fig. 1 Temperature profiles for all the experiments using different percentages of biochar and the control experiment. Detailed temperature profiles for different heights of the composting reactors can be found in the Supplementary Information (Tables S3 and Fig. S3)

Methane

Methane is the main contributor of greenhouse gas (GHG) emissions from composting, although when composting is performed under strict aerobic conditions, its content is quite low [40]. Figure 3 presents the main trends related to methane emissions using different ratios of biochar and the control treatment. Methane emissions show a clear peak during the first 4 days. The control emissions are the highest, in comparison to the biochar amended. A 15% addition of biochar shows a peak, while 5 and 10% have marginal emissions throughout the process (Fig. 3).

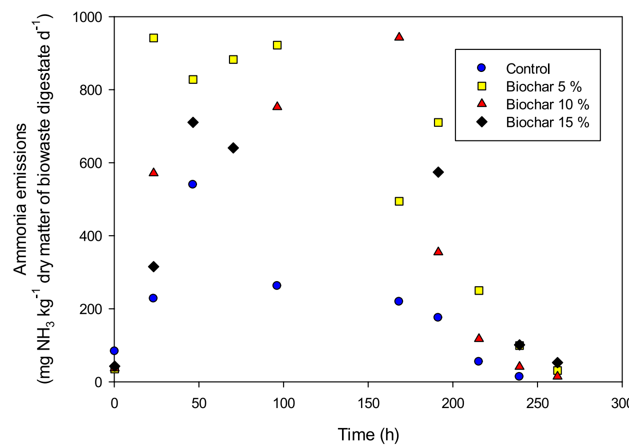

Fig. 2 Profile of ammonia specific emission rate during the composting process of biowaste digestate at different ratios of biochar

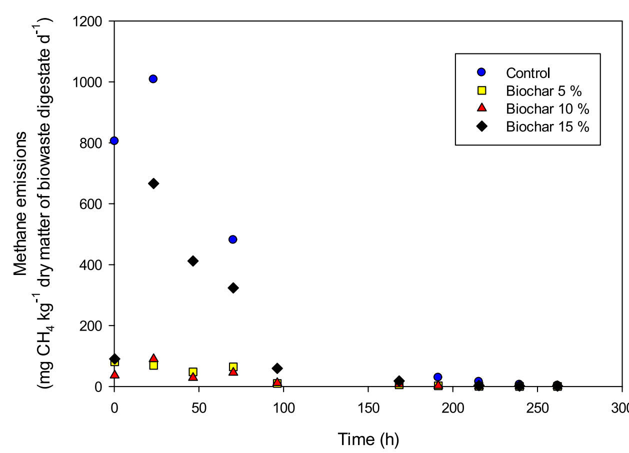

Fig. 3 Profile of methane specific emission rate during the composting process of biowaste digestate at different ratios of biochar

Odours

As expected, odour emissions decrease linearly throughout the process. No significant difference is observed between the different treatments. In general, the control odours are higher than for the biochar treatments (Fig. 4).

Total VOC and VOC Characterisation

Although some studies have presented the VOC emission profile during composting, the references with digestate are very scarce, and often related to a specific typology of VOC or total VOC [41, 42]. In this work, a complete characterization of VOC, showing total VOC emission profile (Fig. 5) and families' speciation with its relative percentage are presented (Fig. 6).

In Fig. 4, the highest VOC emission rates observed are for the control treatment. The peak is observed on the first day of experiment, while for 15% biochar, the peak is observed on day 4. All treatments decrease their emissions during the second week, without any significant difference between them. In Fig. 5, terpenes show to be the highest emitted gases with relative abundances above 80% for all treatments. Alkanes, aromatic hydrocarbons, alkenes and sulphur compounds increase their relative abundance for the biochar treatments.

Cumulative Gas Emissions

Cumulative gas emissions have been determined for ammonia, methane and VOC in Table 3. Ammonia total gas emissions increase with an augment on the biochar ratio, as observed in Fig. 2. Methane emissions decrease an order of magnitude with the biochar ratios of 5 and 10%, however the 15% ratio only halves the cumulative emissions of the control treatment. Total VOC remain within the same range for all treatments.

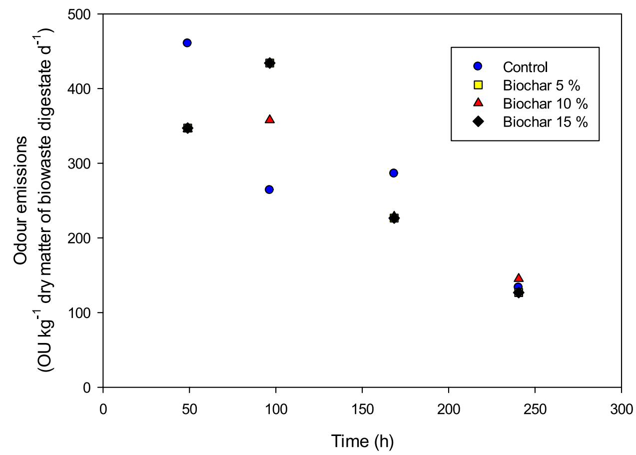

Fig. 4 Odour specific emission rate during the composting process (selected points) of biowaste digestate at different ratios of biochar (ratio of biochar 15% overlaps with 5% in many points)

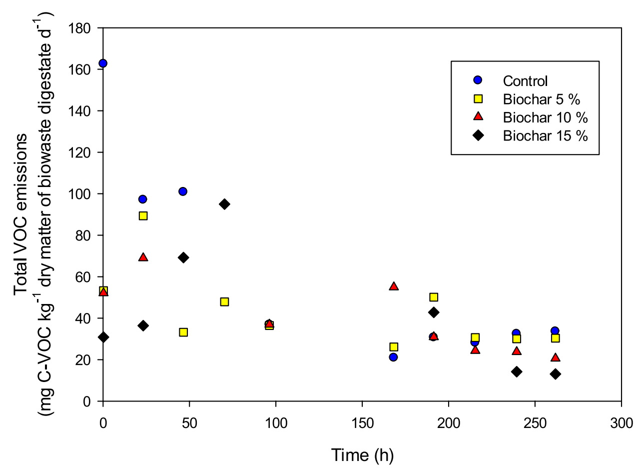

Fig. 5 Total VOC specific emission rate during the composting process of biowaste digestate at different ratios of biochar

Discussion

Composting Process

According to the results presented in this study, biochar properties can have a critical effect on digestate composting performance, as porosity is a crucial parameter for a properly process development [43]. The observations shown in Table 1 and Fig. S1 (Supplementary Information) regarding the specific surface area and the average pore size, jointly with the scanning electron microscope images of biochar, explains its usefulness as an adsorbent and as a support for microbial growth, as previously reported [18, 44]. Moreover, several authors have reported that the feedstock used and the pyrolysis conditions significantly affect the characteristics of produced biochar [45], being our results similar to those reported for non-functionalized biochar [46].

This is of special importance with low-porosity organic waste, such as digestate, as its high moisture contents results in a low level of porosity, which hampers the development of full aerobic composting conditions, promotes methane formation and emission and results in the need of high ratios of bulking agent [36, 43]. Digestate also presents a high content of nitrogen in form of ammonia with a relatively alkaline pH (Table 2), which it is expected to result in high ammonia emissions, as discussed later. This phenomenon has been reported with other high-nitrogen content organic waste, such as wastewater sludge or animal by-products [39].

Another specific property of digestate as organic waste to be composted is its relative low biodegradability. As shown

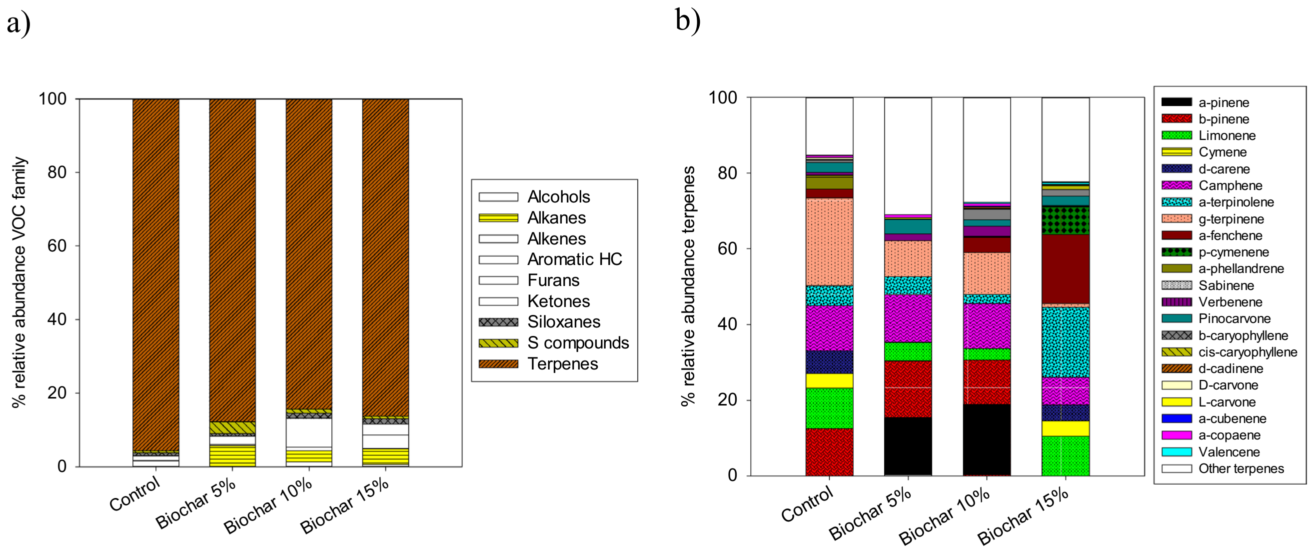

Fig. 6 Volatile Organic Compounds (VOC) characterisation at the time with maximum temperature at different ratios of biochar (control, 15, 10 and 5% of biochar): **a** VOC families (in percentage) for the different ratios of biochar, **b** Main compounds detected in the family of terpenes

Table 3 Ammonia, total VOC (volatile organic compounds) and methane emission factors determined for the composting process of biowaste digestate at different ratios of biochar in lab-scale reactors

Composting treatment	Ammonia (g NH_3 kg^{-1} DM biowaste digestate)	Methane (g CH_4 kg^{-1} DM biowaste digestate)	Total VOC (g C-VOC kg^{-1} DM biowaste digestate)
Control	2.41	4.09	0.57
Biochar 5%	6.61	0.36	0.46
Biochar 10%	5.83	0.28	0.47
Biochar 15%	5.15	1.98	0.58

in Table 2, the digestate properties are relatively similar to other anaerobically digested materials, such as wastewater sludge or several types of manure [47]. The respiration index of the digestate (1.9 ± 0.5 g O_2 g^{-1} DM h^{-1}) meets the criteria for a medium-to-low biodegradable material [48]. This low biodegradability of digestate has important implications on the composting process, specifically in the parameters' evolution. First, it is expected a lower oxygen consumption in the process, which conditions the aeration requirements and the process duration [49] and it was reflected in values of oxygen over 12% in all the treatments during the entire composting process (data not shown). Second, it is probable that thermophilic temperatures are difficult to reach as they are the result of metabolic heat retention, especially at full-scale [50]. In this later case, sanitation, which is related to the fulfilment of specific time–temperature conditions for a safety (free of pathogens) compost manipulation, can be affected [51, 52].

In our study, the highest ratio of biochar (15%) results in temperatures higher than those obtained with lower ratios (including control treatment) in all the locations of the reactor (Fig. 1, Fig. S3a, b, c, Supplementary Information). This is especially relevant as composting is used for organic waste sanitation prior to the use of the resulting compost as organic amendment in soil [51, 53]. In the case of digestate, thermophilic sanitation temperatures are difficult to achieve because of the low digestate biodegradability [54], which are reflected in a low respiration index, as shown in Table 2 [48]. Therefore, the presence of biochar is clearly positive according to this objective. The reason for this temperature increase in the presence of biochar can be due to two main effects: an increase in the biological activity or because of its thermal insulating properties in organic matrices. Both phenomena have been observed in composting environments, but not with digested materials [55, 56]. In consequence, it can be hypothesised that the presence of high ratios of biochar in low biodegradable organic waste results in an increase of the thermal insulating properties of organic matter [56]. Anyway, this fact deserves further studies at larger scales, as sanitation is a critical issue regarding the use of composted digestate in organic agriculture [57].

Another directly related effect of an initial low biodegradability in an organic waste is the resulting stability of compost. In this study, digestate biodegradability values presented some level of dispersion (around 25%, as shown in Table 2), which is a well-known property of highly heterogeneous organic waste [6, 58]. This notwithstanding, the level of stability of the material after a short composting process results in DRI values well below 0.5 g O_2 kg^{-1} DM h^{-1} for all the tested ratios of biochar (including the control treatment), which is an evident validation of the efficiency of the experimental system [35]. From a practical point of view, this level of stabilization provides a proper base for the study of gaseous emissions profiles [5], as explained in the following point.

Gaseous Emissions

Since the gases emitted from a composting process are produced during different stages of the process and are generated as a consequence of different composting conditions [59], it is important to discuss them separately for each specific waste, being biowaste digestate an organic waste with very limited information in composting scientific literature. Table 3 presents the emission factors of the gases studied in this research, which will be discussed for each compound or family of compounds. To our knowledge, this is the first work where these values are presented regarding this type of organic waste.

Ammonia emissions from the composting of different organic waste have been largely studied in literature because of three main reasons: (i) it needs a specific treatment because it is a pollutant, (ii) it is a well-known malodours and stringent odorant and (iii) its loss as gaseous emission results in a compost with less nitrogen content, being this an important property of compost when used as fertilizer [60]. It is also widely studied that biochar prevents ammonia emission in the composting of several organic waste, with a wide range among 5–40% of reduction related to the initial nitrogen content [56, 61]. However, Fig. 2 shows a different trend. Thus, the presence of biochar increases the ammonia emissions, which results in a significant increase even at low biochar concentration (5%), and even doubling them depending on the biochar ratio used.

To explain this fact, the main hypothesis is the different temperature patterns observed when biochar is present in the composting mass. The bioreactor used in these treatments is based on a Dewar® vessel, which is known for its extraordinary heat insolation. Moreover, biochar also enhances the thermal insulating properties of organic matter, as it has been recently demonstrated with heat transfer models [56]. These combined factors result in a higher temperature in the presence of biochar, as it can be confirmed with the temperature profiles in the three sections of the composter (Fig. S3, Supplementary Information). The observed differences in temperature can reach 8–10 °C during several days in the biochar-amended composting treatment. Since it has been reported that the dependence of ammonia emissions with temperature is exponential [39], the ammonia emissions shown in Fig. 2 follow this trend. Additionally, the nature of biowaste digestate, which contains high amounts of nitrogen in form of ammonium and highly-labile organic nitrogen (Table 2) can contribute to these high emissions in the presence of biochar [9]. In composting, temperature is also dependant on the reactor scale because of the "thermal inertia" effect [62], therefore, it is probable that this phenomenon is even more relevant at full-scale, although only one reference has been found in literature to the best of our knowledge [63]. In this study, however, this increase in temperature in the presence of biochar has another positive effect, which is material sanitation, as commented above. This is especially interesting with low biodegradable substrates as digestate inherently is (Table 2) and it implies that digestate composting should be carefully analysed when designing exhaust gas cleaning operations, which will be different than those used for standard composting using raw non-digested organic waste.

As a conclusion, and only considering ammonia emissions, it is evident that with this kind of composting substrates, which are expected to be predominant in the next future because of anaerobic digestion expansion, biochar addition must be carefully analysed in economic and environmental terms and other novel strategies can be evaluated [64]. This analysis should extend beyond the scope of biological treatment of municipal biowaste, including anaerobic digestion and composting, particularly with regard to the application of compost amended with biochar. It is well known that the presence of biochar reduces ammonia emissions in soil when applied to crop production [15, 26]. Unfortunately, to the authors' knowledge, there are no studies on the use of compost containing biochar from the outset of the composting process. Such studies would enable an overall nitrogen mass balance to be performed, which could serve as a basis for a sustainability assessment related to this critical fertilisation element.

In the case of methane, a completely different emission pattern can be observed (Fig. 3). In the control treatment, a pronounced peak of methane is detected at the very beginning of the composting process (around 1000 mg CH_4 kg^{-1} DM of biowaste digestate d^{-1}), which is higher than those published in composting literature with municipal waste or even wastewater sludge [5, 64, 65]. A possible explanation for this high peak is that this part of methane is released from biowaste digestate once the material is conditioned for composting (bulking agent addition) and the application of forced aeration causes the stripping of the entrapped methane [66]. Although this phenomenon could not be experimentally confirmed, the fact that a first high peak of ammonia and methane appears at the beginning of the composting process has been confirmed by the plant managers (personal communication) and partially observed with other similar substrates as digested wastewater sludge [10]. In this previous study, the initial emissions of methane were reported to be 60 times higher when using digested sludge instead of secondary sludge. In the case of biowaste digestate, which is also supposed to have a large amount of methane coming from anaerobic digestion, the behaviour seems to be similar. Nevertheless, it is evident that this fact deserves further research given the importance of methane as GHG.

In the presence of biochar, this initial methane peak is significantly lower (Fig. 3). Thus, in biochar ratios of 5–10%, no methane peaks are observed and the specific emission rates during the entire composting process do not reach 100 mg kg^{-1} DM of biowaste digestate d^{-1}. Moreover, methane emission factors determined for each treatment (Table 3) support these observations, presenting a reduction of up to a 93% of the total methane released throughout the whole composting process when using a biochar ratio of 10%. This fact is very relevant as it confirms the adequacy of biochar to mitigate methane emissions in the composting of biowaste digestate, as previously reported with other organic waste [22, 55]. With these ratios of biochar, a reduction of methane emission during the composting could be estimated to be more than 90%. In this sense, biochar has been reported to act as a strong adsorbed for hydrophobic hydrocarbons, such as methane, close to the activated carbon capacity [67]. However, with a high ratio of biochar (15%, dry basis), the observed reduction in methane emission is significantly lower (around 50%). Although the explanation of this fact is not clear, it can be hypothesised that, in this situation, although a certain adsorption capacity of methane remains, the effect of high temperature prevents an almost complete adsorption as observed with ratios of 5–10%. The temperature profile observed with different ratios (Fig. 1, Fig. S3, Supplementary Information) confirms that temperature for the treatment with 15% of biochar is higher than those observed for lower ratios of biochar in 5–7 °C during the thermophilic composting phase. Additionally to adsorption, methane emissions could be mitigated by the biochar's microporosity. Anaerobic areas, where methane is produced, are not only prevented by the macroporosity added by the bulking agent, but also the macro and microporosity provided by the biochar. In consequence, the phenomena observed with methane could be similar to that of the ammonia emissions, as previously commented. In summary, it can be concluded that the selection of biochar ratio plays a key role in the resulting composting gaseous emissions, being this a general conclusion. This has been confirmed by previous works were the ratio of biochar in composting has been optimized in terms of gaseous emissions [68]. However, to our knowledge, this is the first work with biowaste digestate that presents an overall picture of all the possible composting emissions.

In the case of nitrous oxide, which is a powerful GHG emitted from composting [5], all the values measured were close to the detection limit of the analytical method (1 ppm) and consequently discarded. Probably, the short composting period and the high level of oxygen prevents the nitrification-(partial) denitrification process, which is the responsible for nitrous oxide composting emissions [69].

Odours, measured through dynamic olfactometry, are scarcely reported in composting literature. In this study, and although the number of points where odour concentration was determined is low, it can be concluded that a similar trend than that of methane is observed (Fig. 4). Thus, biochar seems to partially abate some odour compounds when compared to control. However, high ratios of biochar do not present positive results. In this case, it is evident that more research is needed to confirm these preliminary results, especially with digestate derived from different organic waste, as odour emission has been pointed as a factor to be included in the environmental performance and social acceptance of waste treatment plants [70].

Finally, the emission of total VOC showed similar trends among the four different treatments, with the only particularity of the suppression effect shown by the biochar treatments for the first VOC emission peak, which is reduced up to an 80% when biochar is present in the initial mixture (Fig. 5). When comparing the total VOC emission factors determined for each treatment (Table 3), the differences between the control and the biochar trials are minimal, showing a reduction of up to a 19% on the total VOC emission when using a biochar ratio of 10% with respect to the control treatment. However, total VOC emission is often not informative enough, as VOC are composed of a large number of families of different properties and behaviour during composting [11].

The case of VOC families' characterization is similar to that of odour emissions in terms of cost and complexity of the analytic method. Figure 6a shows the relative percentage of VOC with different ratios of biochar in the thermophilic stage of the composting process. This figure can be confusing, as the percentage of terpenes was so high that hampers the visualization of the evolution of the rest of VOC families. This phenomenon has been observed in the composting of other organic waste [71], especially when high amounts of bulking agent are used, as is the case of digestate due to its low porosity [43]. Figure 5b presents the most predominant VOC found in biowaste digestate composting. To our knowledge, no similar results have been presented in scientific literature, and this information can be the base for further studies. In summary, it is evident that biochar and its ratio have a strong influence in the particular emission of each VOC. Again, considerable differences can be observed in terpenes content. As VOC are involved in several atmospheric processes such as the formation tropospheric ozone

[72], apart from the inherent toxicity of some of them, it is evident that the effect of biochar must be more specifically addressed, as biochar seems very effective in the adsorption of some VOC. In terms of total VOC, it is also evident that biochar is an effective adsorbent as it hampers the initial VOC release. However, with high ratios of biochar, a similar behaviour than that observed for methane and odours can be also observed. In the case of VOC, it is again evident that the ratio of biochar plays a significant role in gaseous emissions, both in the case of total VOC and the families they comprise. Regarding this, it is important to highlight that, to our knowledge, no studies showing this phenomenon could be found, as no articles dealing with digestate or other low-biodegradability similar substrates with different biochar ratios presented detailed composting emission results [73]. However, this differs from what happens in other biological treatments of biowaste, such as anaerobic digestion [74]. In this case, it was observed that increasing the dose of biochar ratio (typically over 10%), this material had no further effect on process performance. While it is evident that composting and anaerobic digestion are different processes, the improvement in anaerobic digestion performance due to biochar is typically attributed to the adsorption of inhibitors, such as free ammonia or volatile fatty acids. This resembles the effect that biochar can have on composting emissions, although further studies should demonstrate this experimentally.

Conclusions

This study shows the performance of biowaste digestate composting at bench scale regarding its associated gaseous emissions and the compost stabilization when using different ratios of biochar. The results obtained show that the overall emissions picture using biowaste digestate is considerably different than that obtained when composting other more biodegradable organic waste. The reason for this phenomenon seems to be due to the inherent characteristics found in digested materials: low biodegradability and high content of ammonia forms. The presence of biochar changes the emissions pattern of gaseous greenhouse gases, odours and some groups of volatile organic compounds, with a decreasing trend. On the contrary, ammonia emissions are higher due to the increase of process temperature. This increase seems to be related to the thermal insulating properties of biochar, as respiration rates are not significantly altered in the presence of biochar and results in well-stabilised compost. This increase in temperature is of interest as sanitation is an important issue when dealing with waste that has low biodegradability and short composting processes. However, an increase in temperature is also expected to affect the profile of gaseous emissions in terms of quantity and composition. Therefore, the effect of biochar on digestate composting at an industrial scale should be studied. Other areas for future study could include using chemically modified biochar to reduce gaseous emissions, or analysing the potential for using pyrolysis to obtain biochar from green waste while kitchen waste undergoes anaerobic digestion in a biorefinery context. From a sustainability perspective, the overall benefits of biochar on biowaste digestate composting should be carefully considered in further studies regarding its economic viability and environmental performance.

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Data Availability The datasets generated during and analysed during the current study are available upon request.

Declarations

Conflict of Interest The authors have no relevant financial or non-financial interests to disclose.

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