Heterogeneity of biochar amendment to improve the carbon and nitrogen sequestration through reduce the greenhouse gases emissions during sewage sludge composting

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HIGHLIGHTS

- Biochar additions increased the dehydrogenase activity during composting.
- Biochar have a potential impact on reduction of N losses and gaseous emissions.
- Biochar amendments shorten the thermophilic phase period.
- Biochar additions increased CO_2 emission and reduce the ammonia losses.
- Biochar additions have significant correlation with carbon and nitrogen losses.

GRAPHICAL ABSTRACT
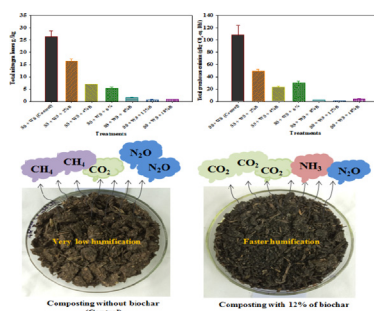
ABSTRACT

This study was performed to investigate the effects of biochar as an amendment to a gaseous emissions and sewage sludge (SS) composting dynamics. Six dosage of biochar [low dosage of biochar (LDB) – 2%, 4% and 6%; and higher dosage of biochar (HDB) – 8%, 12% and 18%] were amended to a mixture of SS and wheat straw (4:1 ratio on dry weight basis) and compared to control or without additive. The HDB significantly reduced CH_4, N_2O and NH_3 emission by 92.85–95.34%, 95.14–97.30% and 58.03–65.17%, but not the CO_2 emission. Meanwhile, humification results indicated that humic and fulvic acid 35–42% and 24–28% higher in the HDB amended treatments than those in the LDB and control treatments. The HDB significantly decreased total nitrogen losses and greenhouse gas emission, while LDB had significantly ($p < 0.001$) higher CH_4 and N_2O emissions. Due to effective performance of HDB, the 12% biochar was recommended to be used in SS composting practice.

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1. Introduction

Nowadays, greenhouse gas (GHG) emissions from various anthropogenic organic waste management processes are attracted the attention of environmental scientists because of the growing concerns on global warming (Chen et al., 2010). Recently, Bong

et al. (2016) has reviewed the influence of different waste management strategies on GHG emissions and emphasis the side-effect of the emissions during organic waste composting and mitigation strategies. However, one of the major part of GHG emission is carbon dioxide (CO_2) which represent 63%, whereas methane and nitrous oxide produced 24% and only 3%, respectively (Luo et al., 2013; Jiang et al., 2016). But particular CH_4 and N_2O emission have a high warming potential, 30 and 210 times higher than CO_2 (IPCC, 2007). The huge quantity of CO_2 is generated during organic matter degradation, but its global warming influence is negligible as compare to CH_4 and N_2O for the evaluation of the impact of waste management practices because CO_2 evolutions are of biogenic origin (IPCC, 2007; Awasthi et al., 2016b). In consequence, some developing countries e.g. China is one of the highest populated country and produced huge quantity (30 million tons/year) of putrescible characteristics of sewage sludge (SS) from different wastewater treatment plants, and its ecofriendly management by traditional composting process generated extensive quantity of GHG (Zhang et al., 2014; Awasthi et al., 2016c). Therefore, increasing the demand for improving and expansion of composting strategies with the formulation of novel feedstock to reduce not only CH_4 and N_2O emission, but also mitigate significant amount of CO_2 production and total nitrogen as well as carbon loss during various kinds of organic waste composting.

In order to control global warming, recently several earlier researchers have applied to reduce the CH_4 and N_2O emission during various organic waste composting employing different bulking agents like saw dust, agricultural wastes (Szanto et al., 2007; Zhang et al., 2014) and amended with different kinds of mineral additives such as lime, zeolite, bentonite and medical stone (Li et al., 2012; Awasthi et al., 2016b,c; Wang et al., 2016). But no any previous study reported to reduce CO_2 emission combined with other gases like NH_3, CH_4 and N_2O, because huge quantity of CO_2 emission and less amount of other gases such as NH_3, CH_4 and N_2O can cause secondary pollution and reduces the environmental benefits of compost. Consequently, these gases emission combined together contribute to maximum organic nutrient loss from the end product and also lower the quality of the compost. However, CO_2 emission is inevitable during composting when the organic matter mineralization and the temperature and pH are high (Zhang et al., 2014; Jiang et al., 2016; Awasthi et al., 2016b), while CH_4 and N_2O emission are significant GHG. But previous studies have proved that more than 30–40% of total organic carbon, 70–74% of initial total nitrogen (TN) is lost during organic waste composting (Beck-Friis et al., 2000; Fukumoto et al., 2003). Among these, most TOC is lost in form of CO_2 emission (Awasthi et al., 2016c,d), while 10–46% of TN is lost in the form of NH_3 (Sanchez-Monedero et al., 2010; Luo et al., 2013; Wang et al., 2016) and 0.1–10% is lost in the form of N_2O (Shen et al., 2011; Awasthi et al., 2016b). The CH_4 emission is a byproduct of anaerobic degradation during the composting but can be substantially reduced the emission with proper aeration and addition of bulking agent. Beside this, Sommer and Moller (2000) has reported that between 0.01% and 0.03% of the initial total organic carbon (TOC) may be lost in the form of CH_4, when settlement of composting mass and then some anaerobic pockets formed inside.

Recently, wheat straw biochar production and its amendment for SS composting is considered to be as cost effective and ecofriendly technology with successful composting (Malinska et al., 2014; Jiang et al., 2016; Awasthi et al., 2016b), because biochar has excellent ability to adsorbs GHG, and ammonia as well as increases the humification of organic waste (Dias et al., 2010; Jindo et al., 2012; Chowdhury et al., 2014). Consequently, last few years several earlier researchers observed that biochar amendment led to faster mineralization and reduced total GHG emission by 42.8% during the poultry manure composting (Dias et al., 2010;

Chowdhury et al., 2014), while Chen et al. (2010) and Jiang et al. (2016) reported that ~45 to 55% GHG emission reduction pig feces and pig manure mixed with struvite salt and coffee husk biochar. But most of previous studies have only focused on CH_4, N_2O and NH_4 reduction employing biochar as amendment for various types of organic waste materials composting (Steiner et al., 2010; Wang et al., 2013; Bong et al., 2016), while CO_2 and ammonia losses are another problematic issue. Therefore, we hypothesized that biochar amendment into mixture of feed stock will be provide various benefits during the SS composting. Because porous micro-structure of biochar leads to provide optimal aeration as per indigenous microbial demand and not only reduce the CH_4 and N_2O emission, but act as biofilter to decreased the CO_2 and ammonia losses (Chen et al., 2010; Sonoki et al., 2013), consequently biochar amendment alleviate the initial low pH during the thermophilic stage of composting (Wei et al., 2014; Czekala et al., 2016). Remarkably, from an extensive literature search and conclude that no previous studies reported reduction of CO_2 combined with CH_4 and N_2O emission as well as its relationship with humification. Therefore, the purpose of the present study was (i) to evaluate heterogeneity of biochar for total organic carbon loss mitigation through CO_2 and CH_4 emission and nitrogen conservation by N_2O and NH_4 reduction, and as consequence to determine the most effective dosage biochar for SS composting; (ii) to study the relationship between the mechanisms involved in the total gaseous emission, carbon, nitrogen losses and humification of the composting mixtures.

2. Materials and methods

2.1. Raw materials collection and processing

The SS and wheat straw were used as raw materials in this investigation. SS was obtained from a local municipal wastewater treatment plant (Yangling, Shaanxi Province, China) and wheat straw (WS) was taken from the local agricultural farm research station of Yangling Northwest A&F University. Wheat straw biochar (WSB) was purchased from Yangling Pvt. Ltd., Shaanxi Province, China. To achieve the appropriate moisture content (~55%) and C/N ratio ~25, SS and WS were mixed at a ratio of 4:1 (dry weight basis). In addition, 1 kg of plastic spheres was mixed with initial feed stock to adjust the initial bulk density to ~0.5 kg/L according to our previous work experience (Awasthi et al., 2016a). Air dried WSB was crushed into fine particles and sieved to 2–5 mm; and then used as an amendment for composting, while the characteristics of raw materials are shown in Table 1.

2.2. Experiment design and compost sample collection

The composting process was performed in polyvinyl chloride (PVC) reactors, each with a total working volume of 130-L under controlled ambient temperature; and systematic layout of the

Table 1
Selected physicochemical properties of raw materials used in the present experiments (dry weight basis).

Parameters	SS	WS	WSB
Moisture content (%)	82.10 ± 2.17	12.86 ± 0.26	3.30 ± 0.08
pH (solid:water = 1:5)	7.18 ± 0.06	4.93 ± 0.10	8.83 ± 0.12
EC (mS cm^{-1}) (solid:water = 1:5)	5.36 ± 0.12	0.89 ± 0.02	1.05 ± 0.04
Total organic matter (%)	79.30 ± 2.18	98.38 ± 2.31	95.60 ± 2.17
Total organic carbon (%)	42.72 ± 2.13	58.14 ± 1.04	65.08 ± 1.10
Total Kjeldahl nitrogen (%)	2.70 ± 0.06	0.80 ± 0.03	0.58 ± 0.02
C:N ratio	15.82 ± 0.14	72.70 ± 1.20	112.2 ± 4.31

ND – Not detected, SS – Sewage sludge, WS – Wheat straw, WSB – Wheat straw biochar and EC – Electrical conductivity. Results are the mean of three replicates ± standard deviation.

reactor and operational process were used for the present investigation is already described in our previous study (Li et al., 2012). Seven treatments were conducted in triplicate to evaluate the effect of biochar at 2, 4, 6, 8, 12 and 18% (on SS dry weight basis) to determine the best optimum dosage of biochar for GHG emission reduction with special emphasis to decreased the CO_2 emission and TOC as well as nitrogen losses. The SS + WS without any additives amendment was used as control for comparison purpose. About 100-L of each mixture was composted for 56 days in a reactor. Moisture content of the composting mixture was readjusted to 55% periodically on turning days 0, 3, 7, 10, 14, 21, 28, 35, 42, 49 and 56; meanwhile about 250 g compost samples were collected from each treatment for further analysis. Fresh samples were separated into three parts; one part was air dried, properly ground, sieved through 0.1 mm sieve and then used for total nutrients content analysis, while two other parts were stored as fresh sample on 4 °C and −20 °C for chemical and microbiological analysis. The composting biomass temperature was monitored every day four times (6 h) by the using of thermocouple probe inserted into the center of the composting materials and averaged temperature was reported, respectively. Despite this the room temperature was also recorded. The thermocouple, air inlet and outlet pipes were disconnected from the reactor when the composting mass was manually turned and mixed.

2.3. Greenhouse gas sampling and measurement

To evaluate the gaseous emission during the composting, the ammonia was absorbed in boric acid (2%) for 1 h and concentration was determined according to standard method of Meeker and Wagner (1933), while GHG (CO_2, CH_4 and N_2O) were daily collected during the first 2 weeks and then 3 times per week by the using of 1-LTedlar® PLV gas sampling bag w/Thermogreen®LB-2 Septa (Sigma–Aldrich) according our previous study (Awasthi et al., 2016b). The concentration of GHG were analyzed within 12 h using gas chromatography (Agilent Technologies 6890 N Network GC system, China) as explained by Colón et al. (2012), while data for non-measured days were calculated by averaging the closest measured days.

2.4. Compost analysis

Moisture content was determined by oven drying at 105 °C till constant weight basis. Fresh compost samples 1:5 aqueous extract (dry weight basis) was used for the analysis of pH, electrical conductivity (EC), extractable ammonium (NH_4^+-N) and germination index (GI). The pH and EC were measured using pH meter with a glass electrode (INESA PHSJ-3F, China) and conductivity electrode (INESA DDS-307, China). Total organic matter (TOM), NH_4^+-N, total Kjeldahl nitrogen (TKN) and TOC were determined as per the standard Test Methods for the Examination of Composts and Composting (TMECC, 2002). The dehydrogenase enzyme activity (DEA) was analyzed using aqueous extracts of fresh compost samples as per the standard method of Barrena et al. (2008) and activity is expressed in mg TPF g^{-1} dw h^{-1} on a dry weight basis. Humic substances (humic acid and fulvic acid) from the air-dried compost samples were extracted as per the standard procedure and analyzed using the KBr pellet method (Wei et al., 2007) with the Nicolet 550 Magna-IR spectrometer (Nicolet Instrument Corporation, Madison, WI) using a detection range between 350 and 4000 cm^{-1}.

2.5. Statistical analysis

All the physic-chemical and enzymatic analyses were performed in triplicate. Meanwhile data were subjected on the basis of two-way analysis of variance (ANOVA) and multiple comparison tests were also performed to compare the least significance difference (LSD) at $p = 0.05$ values using SPSS v.21 software package for windows. The principal component analysis (PCA) was performed using the *R* language v3.0 software with prcomp function and the package ggbiplot, while Redundancy analyses of physiochemical properties, greenhouse gas, ammonia emission and studied enzymes activity during the composting were conducted by Canoco 5.

3. Results and discussion

3.1. Effect of biochar amendment on temperature, pH and dehydrogenase activity

For higher dosage of biochar (HDB) amended (8%, 12% and 18%) treatments, the temperatures sharply increased at the beginning of composting and exceeded more than 60 °C within 3 days (Fig. 1a), which reflect rapid degradation of organic waste. The thermophilic phase lasted around 2 weeks and then gradually decreased with the deficiency of easily available degradable organic matter; this bio-oxidation phase is long enough to destroy the pathogenic microbes and weeds seeds (Jindo et al., 2012; Zhang et al., 2014; Awasthi et al., 2016b). When compare to the low dosage of biochar (LDB) added treatments, the thermophilic phase in this trial is significantly late and shorter, possibly due to LDB application cannot accelerate the degradation of organic matter from the beginning of composting; and its thermophilic phase lasted only four to five days as well as temperature never go more than 55 °C. In contrast, without biochar added or control treatment showed very low temperature through the composting process and never reached more than 50 °C, which could be attributed due to low pH and higher moisture content (data not shown). The acidic pH and higher moisture content not only inhibit the composting, but also increase the anaerobic clumps, volatile organic acids (VOAs) and NH_4^+-N formation in the composting feedstock that further decreased the degradation of organic matter and accelerate the GHG emission as well as cause odor problems (Steiner et al., 2010; Sanchez-Garcia et al., 2015; Awasthi et al., 2016c).

The pH of the initial feedstock was slightly alkaline in all biochar amended treatments than their initial values or control treatment (Fig. 1b). LDB (2%, 4%, and 6%) added treatments were showed slightly acidic pattern of pH during the early phase of composting and HDB (85, 12% and 18%) amended treatments showed neutral range of pH profile with a rapid activation of composting process, while control treatment showed acidic pH through the composting (Fig. 1b). Consequently, the results indicated that HDB addition (8%, 12% and 18%) adequately buffer the VOAs production during the thermophilic phase. Beside this, LDB resulted low buffering ability during the early phase of composting and as consequence low microbial activity, humification and low CO_2 emission were observed at the beginning of composting (Fig. 2). This fact is in line with the previous studies using biochar as amendment or co-substrate for various kind organic waste composting (Wang et al., 2013; Wei et al., 2014; Czekala et al., 2016).

During composting, the starting raw stocks were degraded by variety of biochemical process in which different kind of substrate specific group of enzymes (dehydrogenase, cellulase, protease, amylase, xylase and chitinase) involved, but the DEA showed the overall microbial dynamics of the composting process as like temperature profile. Because DEA was directly involved with the biogenic activity, which could collectively provide the comprehensive information about total gaseous emission and end product quality (Fukumoto et al., 2003; Luo et al., 2013; Awasthi et al., 2016d). DEA in all HDB amended treatments rapidly increased during the thermophilic phase and then gradually decreased with the
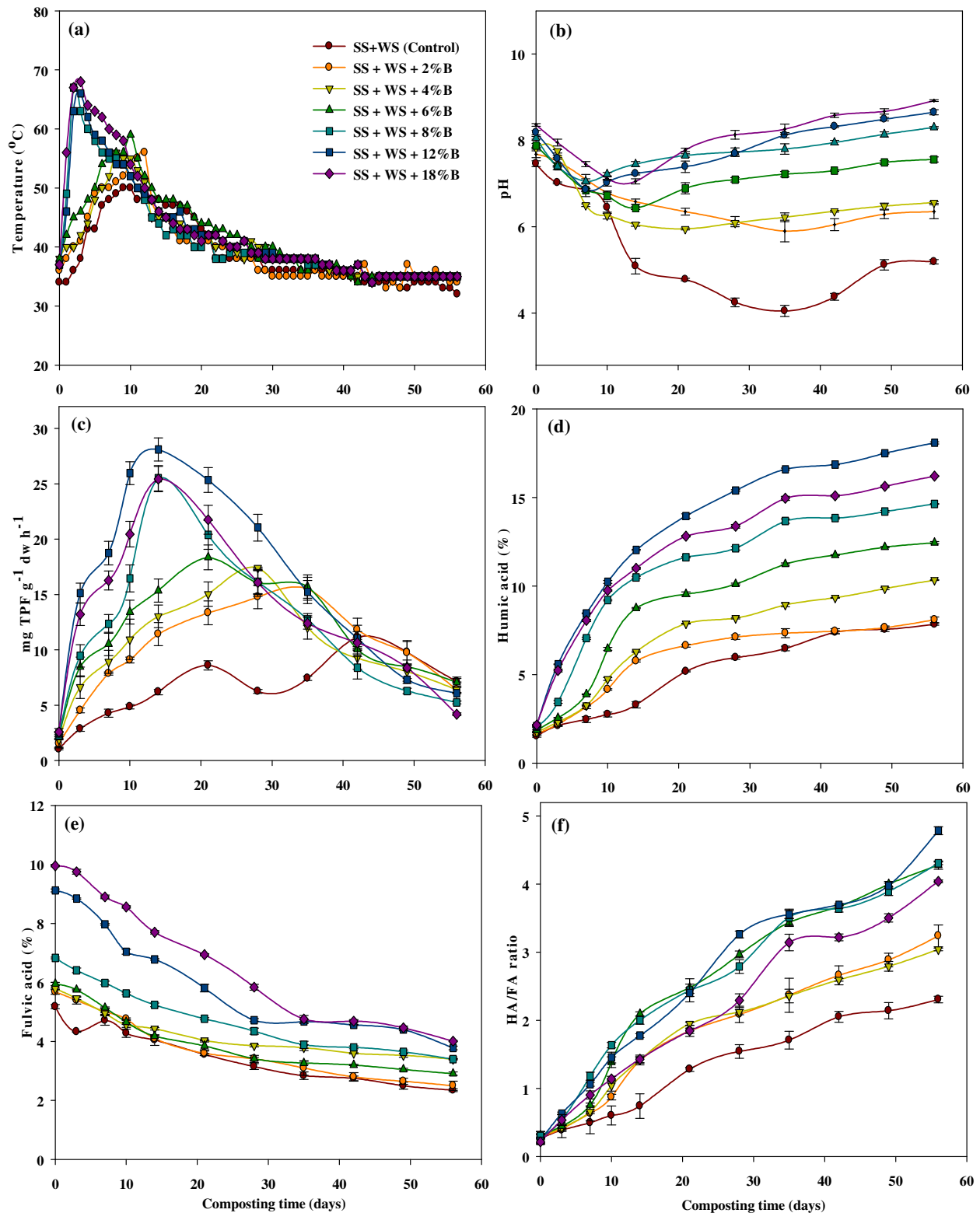
Fig. 1. Evaluation of temperature (a), pH (b), dehydrogenase (c), humic acid (d), fulvic acid (e) and humic/fulvic acids ratio (f) profile of different treatments during composting of sewage sludge. SS + WS: sewage sludge + wheat straw (Control); SS + WS 2% B: sewage sludge + wheat straw + 2% Biochar; SS + WS + 4% B: sewage sludge + wheat straw + 4% Biochar; SS + WS + 6% B: sewage sludge + wheat straw + 6% Biochar; SS + WS + 8% B: sewage sludge + wheat straw + 8% Biochar; SS + WS + 12% B: sewage sludge + wheat straw + 12% Biochar and SS + WS + 18% B; sewage sludge + wheat straw + 18% Biochar. Results are the mean of three replicates and error bars indicates standard deviation.

maturation phase of composting, which is indicated that there was active degradation of TOM (data not shown) and CO_2 emission (Fig. 2). The maximum DEA was obtained in HDB (8%, 12% and 18%) amended treatments (28.10 mg TPF g^{-1} dw h^{-1}) in 12%

WSB, (25.51 mg TPF g^{-1} dw h^{-1}) in 8% WSB and (25.43 mg TPF g^{-1} dw h^{-1}) in 18% WSB on day 14, respectively, while treatment amended with LDB (25%, 4% and 6%) showed significantly lower DEA (15.52, 17.41 and 18.35 mg TPF g^{-1} dw h^{-1})
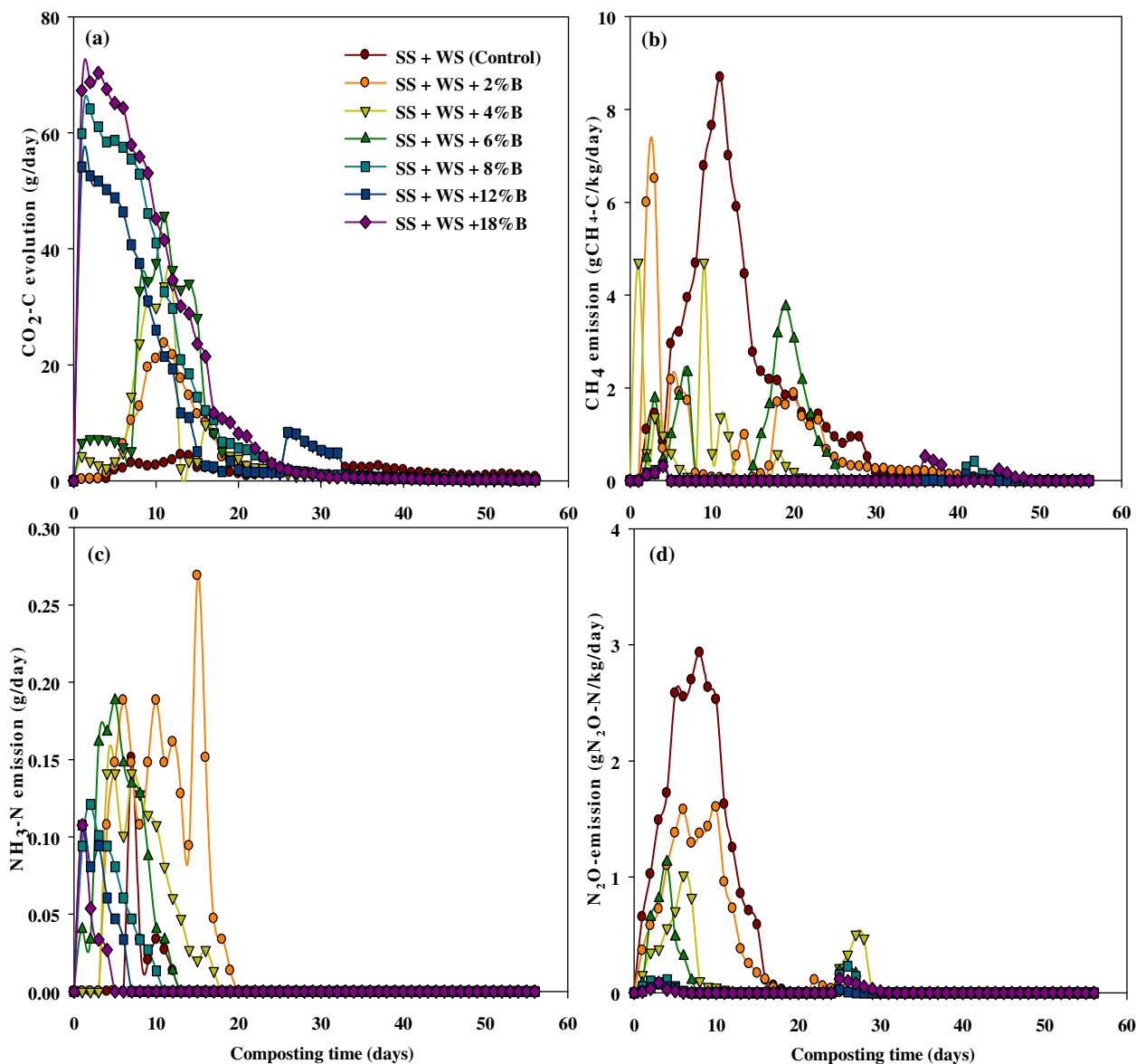
Fig. 2. Evaluation of CO_2 (a), CH_4 (b), NH_3 (c) and N_2O emission (d) profile of different treatments during composting of sewage sludge. SS + WS: sewage sludge + wheat straw (Control); SS + WS + 2% B: sewage sludge + wheat straw + 2% Biochar; SS + WS + 4% B: sewage sludge + wheat straw + 4% Biochar; SS + WS + 6% B: sewage sludge + wheat straw + 6% Biochar; SS + WS + 8% B: sewage sludge + wheat straw + 8% Biochar; SS + WS + 12% B: sewage sludge + wheat straw + 12% Biochar and SS + WS + 18% B; sewage sludge + wheat straw + 18% Biochar. Results are the mean of three replicates and error bars indicates standard deviation.

on day 35, 28 and 21, respectively (Fig. 1c). Because HDB addition provide a suitable habitat for indigenous microorganism proliferation and as results exerts a positive effect on feedstock properties such as porosity, surface area and optimum moisture content. These changes in substrates promoted a microbial enzymatic activity, which is also reflected these treatments temperature, pH and gaseous emission profile (Figs. 1 and 2). Whereas among the all biochar amended treatment, 12% biochar addition showed significantly maximum DEA (28.10 mg TPF g^{-1} dw h^{-1}). Consequently, when compare with the control treatment the lowest DEA was observed through the composting, this might be due to the low pH (Fig. 1b) and high moisture content (data not shown) that cause unfavorable condition for microbial growth; hence it can be also considered for low CO_2 and ammonia emission profile, while maximum CH_4 and N_2O emission (Sanchez-Garcia et al., 2015; Bong et al., 2016). In addition, the change in DEA could be used as the indicator of the state of composting and provide the information about heterogeneity of biochar dosage to reduce the GHG emission

and nitrogen loss as well as compost maturity period (Barrena et al., 2008; Ermolaev et al., 2014). In contrast, the results indicated that LDB addition is not effective for SS composting, because it cannot buffer the pH from around 6.0–7.5 during the thermophilic phase of composting and as results cannot provide proper aerated conditions that would not help to increased the DEA. The result of our study was in line with the previous studies of Zhang et al. (2014) and Awasthi et al. (2016d), who reported that the addition of HDB had positive impact to increase the humification and DEA.

3.2. Effect of biochar amendment on humification

Most of biochemical transformation of complex organic compounds took place during the thermophilic phase of composting. Therefore, it's important to investigate the characterization of humic compounds during the composting, because this was critically important for understanding the mechanism of organic waste mineralization and gaseous emission during the composting. As

shown in Fig. 1, the FA content gradually decreased in all biochar amended treatments, but the reduction rate significantly very low from 5.18 to 2.35%, 5.68% to 2.50%, 5.79% to 3.40% and 5.95% to 2.91 in the control and LDB (2%, 4% and 6%) amended treatments, respectively, while in HDB amended treatments decreasing trend of FA content is much faster than LDB added treatments. In contrast, HA content underwent a gradually increased with the mineralization of TOM and as a consequence HA/FA ratio was also increased until the end of composting (Fig. 1f). The maximum HA content was observed 18.09% in 12% biochar amended treatment at the end of experiment, which is also represented the rapid break down of organic materials and as indicating the gaseous emission (Fig. 2).

Furthermore, the HA/FA ratio also increase in all treatments with highest value (4.78%) was found in 12% biochar applied treatment, followed by 4.30%, 4.27%, 4.05%, 3.24% and 3.04% in 8%, 6%, 18%, 2% and 4% biochar added treatments, while significantly very low HA/FA ratio observed in control (2.30%). The higher HA/FA content in HDB amended treatments, might be due to the production of excess amount of complex molecules (HA) and polymerization of simple molecules (FA), meanwhile biodegradation of non-humic or FA fraction followed by the formation of more polycondensed aromatic structures. Similar fluctuations were also reported by Zhang et al. (2014) and Awasthi et al. (2016b) in a previous study for SS composting, attributed to be initial instability of HA formation and rapid mineralization with the influence of different dosage of additives or biochar amendment.

3.3. Effect of biochar amendment on gaseous emission

3.3.1. Carbon dioxide and methane emission

The CO_2 and CH_4 emissions (Fig. 2a and b) were directly correlated with the overall microbial activity like DEA, humification and temperature profiles, because this is inevitable byproducts of overall biogenic activity and its production showed the variation between biochar dosages. After composting began, CO_2 emissions from all HDB (8%, 12% and 18%) amended treatments increased rapidly and the peaked value 70.32, 64.10 and 54.11 g CO_2-C observed on day 4, 3 and 2, respectively (Fig. 2a) and then gradually decreased until the maturation phase of composting, this could be due to HDB addition provide favorable condition for biological activities and thus rapid mineralization of organic waste occur. In contrast, significantly very low CO_2 emissions occur from LDB amended treatment (2%, 4% and 6%) during the first week of composting, which attributed to be low pH during the thermophilic stage and as consequence slow down the microbial activities. As we know, more than 90% of CO_2 was emitted during thermophilic phase of composting and the major differences in CO_2 emissions were observed between all HDB amended treatments, showing an average emission over the SS composting process of 873.8, 749.2 and 577.1 gCO_2-C emission per day with 18%, 8% and 12% biochar amended. In all biochar added treatments, the highest CO_2 emission occurred mainly during the initial 20 days of composting, with a gradually decrease until day 35, which coincides with the pH and temperature profile (Fig. 1a and b). Subsequently, CO_2 emission gradually decreased during the end of the thermophilic phase (20 days) to levels lower than 10 gCO_2-C production per day, and finally CO_2 production during maturation phase dropped to levels below 10 gCO_2-C emissions per day. Because CO_2 emission is characteristics of a gradual stabilization of the feedstock since this is frequently used as a maturity index, which reflects the overall microbial activities (Barrena et al., 2008; Ermolaev et al., 2014; Czekala et al., 2016). When compared with the control, addition of HDB increased cumulative CO_2 emission by approximate 60%, which might be obvious from that biochar addition caused a relatively long duration of thermophilic phase

and promoted the decomposition of SS, while LDB amendments have significantly lower cumulative CO_2 emission in this study (data not shown), which opposite with other studies reported that the addition of 4 and 6% biochar increased the CO_2 emission during SS and poultry manure composting (Jindo et al., 2012; Malinska et al., 2014; Sanchez-Garcia et al., 2015).

Overall, the contradictory findings might be result from the different physicochemical properties of raw materials (e.g., the status of available carbon content) in which indigenous microbes survived and utilized. From our results, we can assume that maximum CO_2 emissions from HDB amended was considerable due to biochar addition increased the porosity between the composting matrix and accelerate the decomposition of organic carbon, while LDB addition cannot provide optimum pH to buffer the composting mass and aeration as per the microbial demand. Finally, average CO_2 emissions statistically differ between HDB and LDB added treatments (873.8, 577.1, 749.2, 416.9, 257.5 and 213.8 g CO_2-C emission from 18%, 125%, 8%, 6%, 4%, and 2%, respectively). In addition, our results clearly indicated that at the end of composting in LDB applied and control treatments, CO_2 emissions were significantly lower than HDB amended treatments, which could be due to higher moisture content (data not shown) and low pH cannot favor the microbial activities, but it could not be considered as lower carbon loss.

In consequence, several earlier researcher reported that maximum CO_2 evolution occurred at the beginning of the composting, as found in our present investigation and other composting materials like green waste, municipal solid waste and manure (Sommer and Moller, 2000; Sanchez-Monedero et al., 2010; Wang et al., 2016). To date, the number of studies analyzing CO_2 emissions during various types of organic waste composting, but very limited investigations are available to work for total organic carbon loss reduction through CO_2 emissions or any of the waste properties whatsoever, because the mechanism and prediction on long-term CO_2 emission from different kind of organic wastes are difficult (Chen et al., 2010; Wei et al., 2014; Czekala et al., 2016). Beside this, Luo et al. (2013) and Sanchez-Garcia et al. (2015) proved that the CO_2 emission is depend on several abiotic factors like carbon/nitrogen ratio, moisture and not by indigenous microbes even if they commonly exist during the initial thermophilic phase of organic waste mineralization when microbial activities are high. Therefore, the HDB addition was not only beneficial to buffer the low pH during the early phase of composting, but also maintained the C/N ratio and proper aeration as per indigenous microbes' demand and as results improved the humification or composting. Recent studies have also proved that biochar was able to increase the rate of decomposition and CO_2 emission during municipal solid waste and poultry manure composting (Steiner et al., 2010; Sanchez-Garcia et al., 2015; Czekala et al., 2016).

Normally, CH_4 is released by the deoxidization of produced carbon dioxide/carbon monoxide and VOAs by methanogens under oxygen limited circumstances (Shen et al., 2011). In the present investigation, the CH_4 emissions mainly observed from all LDB (2%, 4% and 6%) amended and control treatments during the early phase of composting and significantly very low CH_4 emitted from HDB (8%, 12% and 18%) added treatments, because HDB provide favorable condition (extra porosity and water holding capacity) for rapid mineralization of organic matter, which is consistent with previous findings and underline regions are well explained (Fukumoto et al., 2003; Sanchez-Garcia et al., 2015). The statistical analysis report showed that there is significant difference ($P < 0.05$) on CH_4 emission profile among the all biochar amended treatments; while significantly major difference for CH_4 emissions was observed between HDB (8%, 12% and 18%) and control treatments, while no significance difference was found among the LDB (2%, 4% and 6%) amended treatments. In control or without

any amended treatment, maximum CH_4 emission was observed from day 8–14, due to the slow degradation of TOM, which may relate to the low microbial activities (data not shown), as evidenced short thermophilic phase and low pH through the composting (Fig. 1) as previous also observed by Ermolaev et al. (2014) and Awasthi et al. (2016b, c) for home and SS composting. Although, CH_4 release from aerobic composting can be temporally observed at the early or maturation stages because consistent air supply and turning apply during the composting to prevent the develop of anaerobic pockets, but due to large amounts of available nutrients and organic compounds utilized by microbes for metabolic activities and thus oxygen deficiency in the reactors during the maturation phase of composting occurred (Jindo et al., 2012; Sanchez-Garcia et al., 2015). Consequently, some previous researchers also investigated that SS with high moisture and LDB could lead to the formation of large clumps during the early phase of composting that can originate anaerobic zones and reduce the composting efficiency, while later on during the maturation phase due to degraded feedstock compaction and settlement decreased the porosity as well as microbial activities (Chen et al., 2010; Sonoki et al., 2013; Wei et al., 2014; Bong et al., 2016). However, material compaction could be found at the end of the process as the composters were neither opened or composting material was not turned during the composting. And it might not be easily observed whether the compaction convicted at the thermophilic or maturation phase of the composting, because there is a significance difference between initial and final porosity of composting mass (Dias et al., 2010; Awasthi et al., 2016c). In addition, a progressive large size clumps formation was responsible for maximum CH_4 emission from the control and LDB amended treatments at the beginning of composting regardless of turnings. In consequence, our present study results indicated that HDB addition (8%, 12% and 18%) avoided the creation of anaerobic pockets and facilitated the oxygen diffusion through the composting matrix; meanwhile Zhang et al. (2014) and Sanchez-Garcia et al. (2015) also reported that the formation of large clumps was very less noticeable with HDB for SS and poultry manure composting. Furthermore, some previous scientists have proved that biochar was able to reduce the CH_4 emissions during manure and municipal solid waste composting (Sonoki et al., 2013; Chowdhury et al., 2014). However, HDB application rates (8%, 12% and 18%) were used in this study, because comparatively LDB (2%, 4% and 6%) addition used in our present investigation was not enough to improve the aerobic conditions at the required level for efficient composting. The CH_4 emission was completely consistent with the trend of temperature and humification profile among the all biochar added treatments. The LDB added treatments have the highest CH_4 emission, this was probably because low amount of biochar amendment cannot provide sufficient oxygen for aerobic bacteria proliferation, and hence the anaerobic conditions prevail in the composting feedstock. Therefore, HDB addition decreased the CH_4 emission and increased the humifcation or organic matter degradation (data not shown), hence our study provided clear landmark on correlation between composting progress and humification, while most of previous available literature only focused on importance of biochar for nitrogen conservation or HMs immobilization and nutrient transformation, respectively.

3.3.2. Ammonia and nitrous oxide emission

The NH_3 emission was positively correlated with temperature ($p < 0.01$ in all treatments) and CO_2 production profile during the SS composting as shown in Fig. 2c. The NH_3 emission is inevitable product during the composting, which is rapidly increased from the beginning of treatments amended with biochar when optimum pH favored the microbial activities and higher temperature also accelerated the NH_3 volatilization (Fukumoto et al., 2003; Shen

et al., 2011; Jiang et al., 2016), but NH_3 emission significantly very low in HDB (8%, 12% and 18%) amended treatments as compare to LDB (2%, 4% and 6%) applied treatments (Fig. 2). However, negligible amount of NH_3 emission was observed from the control treatment or without any amendment at the first week of composting, might be due to low pH and high moisture did not favored the indigenous microbial activities and thus slowed down the mineralization of organic matter. But after first turning significant amount of NH_3 emission was observed from the control treatment (Fig. 2c), while it could be considered as significant loss of NH_3. Among the all biochar added treatments, 90–95% of NH_3 was emitted during the first 20 days composting process and then emission rates sharply decreased thereafter (Fig. 2c). Compared to the previous studies, in our present investigation LDB addition was not effective to decrease the NH_3 emission and promoted the rate composting but HDB application (8%, 12% and 18%) significantly reduced cumulative NH_3 emission by 43.78%, 59.10% and 65.09%, respectively (Fig. 2c). Because HDB amendment increased the amount of degradable carbon and porosity as well as increased the C/N ratio which provide the favorable condition for microbial growth and hence accelerate the immobilization of ammonium into microbial biomass (Sommer and Moller, 2000; Wang et al., 2013).

In addition, throughout the whole SS composting process maximum cumulative NH_3 emissions was observed from the LDB amended treatments (2%, 4% and 6%) 2.07, 1.16 and 1.18 NH_3–N g/day and the HDB added treatments (8%, 12% and 18%) have significantly lower cumulative NH_3 emission such as 0.68, 0.42 and 0.23 NH_3–N g/day, respectively. Because the NH_3 emission were mainly depend on the ammonification rate of organic nitrogen and the aeration; thus statistical analysis results showed that the HDB amendment had a significant influence on NH_3 emissions ($p < 0.001$) reduction, while no significant differences were found between the 4% and 6% biochar added treatments. However, our result is dissimilar with the Malinska et al. (2014), who reported that 4% biochar addition much more effective to reduce the maximum nitrogen loss during the SS composting because large surface area and porous structure of biochar can adsorbed the NH_3 and thus reduced the emission. Therefore, HDB addition may increased the potential to reduced the NH_3 emission during the SS composting in comparison to the LDB added treatments; and our NH_3 emission results was consistent with the previous finding (Steiner et al., 2010; Ermolaev et al., 2014), who also reported that HDB addition much more effective for maximum nitrogen conservation during the poultry manure composting. Meanwhile, Chowdhury et al. (2014) and Awasthi et al. (2016d) also investigated that HDB addition can facilitate the maximum nitrogen conservation during cattle slurry and SS composting, and Luo et al. (2013) found that 20% of biochar on wet weight basis combined with poultry litter can be decrease up to 64% ammonia emission. Beside this, some researchers described that high moisture content and C/N ratio reduced the porosity and as a consequence prohibited organic matter mineralization and then reduced the NH_3 emission, while N_2O emission was started due to the presence of anaerobic pockets in composting mass (Chen et al., 2010; Luo et al., 2013; Jiang et al., 2016).

During composting, the mechanism of N_2O evolution is very complex because N_2O can be emitted by two different ways such as denitrification or incomplete nitrification (Shen et al., 2011; Wei et al., 2014; Sanchez-Garcia et al., 2015; Awasthi et al., 2016b,c). The N_2O emissions of the LDB amended treatments were high during the first 15 days after the start of the composting, and then sharply decreased thereafter; while HDB added treatments have comparatively very low N_2O emissions through the experiment (Fig. 2b). In contrast, maximum N_2O was produced from the control or without any amended treatment between 2 and 14 days and gradually decreased thereafter; this N_2O emission profile clearly indicated that the presence of larger clumps or

anaerobic sites during the start of composting, and may be the result of feed stock settlement also caused the O_2 deficiency. These conditions formed despite the optimum forced aeration and turning events provided to assure aerobic conditions in all treatments or eliminate the anaerobic pockets formation in the composting mass. It seems like that most of N_2O was formed by denitrification, when raw materials stored and processed for short period before began the experiment. This assumption was closely related with El Kader et al. (2007) and Steiner et al. (2010) observation, who reported that high concentration of N_2O produced at the starting of composting that may have been generated before the experiment began. Because maximum amount of N_2O emission was observed in control and LDB added treatments during the early phase of composting, while significantly ($p = 0.001$) very low amount of N_2O was produced from the HDB amended treatments which attributed due to the rapid degradation of total organic matter and low NO_3^- concentration during the thermophilic phase or higher soluble ammonia concentration (Fig. 2d). Consequently, it seems like cumulative N_2O emissions to be responsible for a significant fraction of nitrogen losses in the control and LDB added treatments, as shown in Fig. 2d. However, most of N_2O emissions were detected during the bioactive phase (from day 1 to 14) from the control and LDB amended (2%, 4% and 6%) treatments, but significant amount of N_2O was emitted from the HDB (8%, 12% and 18%) added treatments during the maturation phase of composting, which could be due to the nitrification of NH_4^+-N and that accumulated during the thermophilic phase, and as results nitrifying microbial activity increased the significant amount of N_2O emission (Szanto et al., 2007; Wei et al., 2014; Awasthi et al., 2016c). During the whole composting process, maximum cumulative N_2O emissions observed from the control and LDB added treatments were 26.0, 14.23, 5.72 and 4.17 gN_2O-N, respectively. When compared with the control and LDB added treatments, the HDB amended treatments have the lowest N_2O emission, this could be explained that HDB application provide favorable condition for rapid mineralization of organic matter and temperature as well as CO_2 profile also support this evidence; so the most N_2O was emitted from the LDB and control treatments. In addition, the statistical analysis results also showed that there was a significance ($P < 0.001$) difference in N_2O emissions profile between LDB and HDB added treatments, while no difference was observed among the all HDB amended treatments. The N_2O emission profile was in line with the previous reported studies (El Kader et al., 2007; Malinska et al., 2014; Sanchez-Garcia et al., 2015; Wang et al., 2016), where the proportion of bulking agent, mixing of biochar or other additives and turning frequency were important factors in modulating the total nitrogen losses during the composting.

3.4. Total carbon and nitrogen losses through gaseous emissions

At the end of the composting, the cumulative CO_2, CH_4, NH_3, and N_2O emissions were expressed to get total carbon and nitrogen loss, while CH_4 and N_2O were converted into CO_2-equivalent, and then added to obtain total GHG emissions ranged from 1.12 to 108.61 g/kg CO_2-eq. dry matter (Fig. 3). The results indicated that biochar amended treatments were significantly reduced the total GHG emissions and nitrogen loss as compare to control (Fig. 3), which was in line to the previous findings that biochar can mitigate the GHG emissions during various kind of organic waste composting (Dias et al., 2010; Wang et al., 2013; Sanchez-Garcia et al., 2015). In present study, the negligible effects of biochar amendment on total carbon loss mainly resulted from their effects on total CH_4 emission, which accounted less than 0.3–0.5% of the initial TOC loss. In addition, the HDB had the maximum TOC loss, and TOC loss for LDB was the lowest. This finding is more obvious like several previous studies (Szanto et al., 2007; Luo et al., 2013;
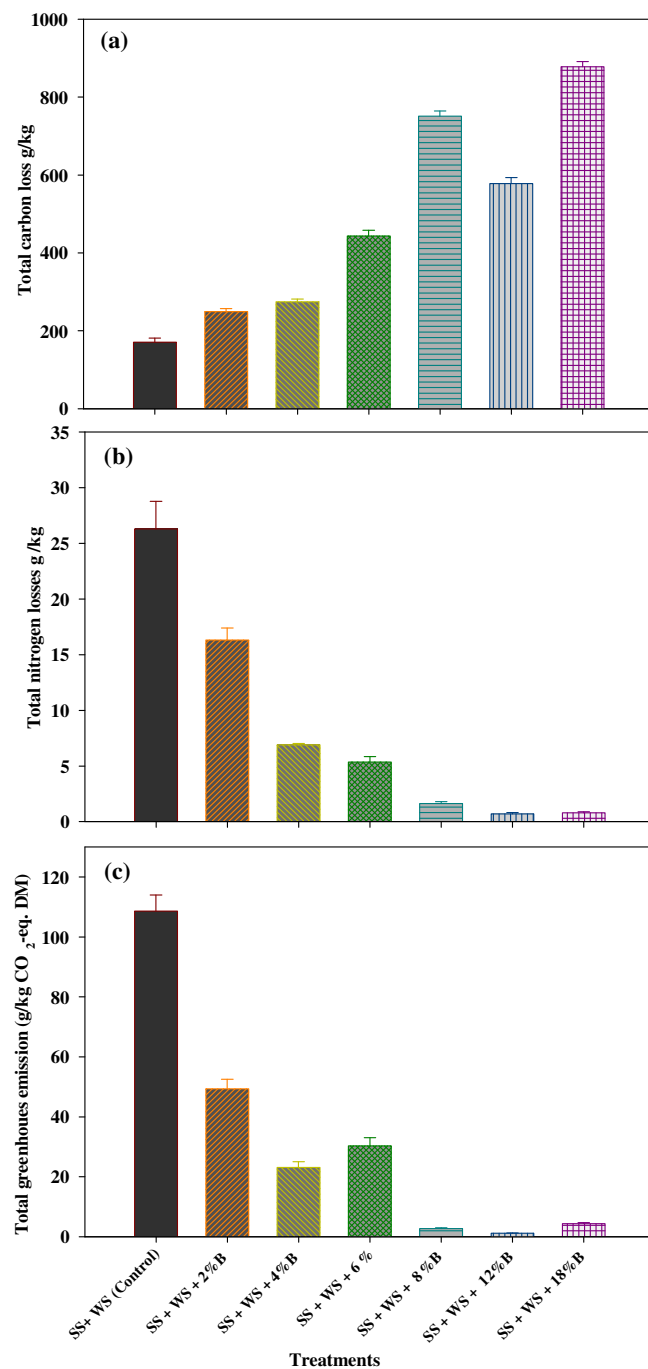

Fig. 3. Evaluation of total carbon loss (a), total nitrogen loss (b), and total greenhouse gases emission (c) profile in different treatments at the end of composting. SS + WS: sewage sludge + wheat straw (Control); SS + WS + 2% B: sewage sludge + wheat straw + 2% Biochar; SS + WS + 4% B: sewage sludge + wheat straw + 4% Biochar; SS + WS + 6% B: sewage sludge + wheat straw + 6% Biochar; SS + WS + 8% B: sewage sludge + wheat straw + 8% Biochar; SS + WS + 12% B: sewage sludge + wheat straw + 12% Biochar and SS + WS + 18% B; sewage sludge + wheat straw + 18% Biochar. Results are the mean of three replicates and error bars indicates standard deviation.

Chowdhury et al., 2014), who reported that HDB leads to maintain optimum air convection as per microbial demand and retain moisture losses in composting matrix, which is conducive to the rapid degradation of total organic carbon. On the other hand, total CO_2 emission during the whole experiment were not influenced by biochar amendments, a possible reason was that increased total CO_2 emission caused by rapid mineralization of TOC in all biochar
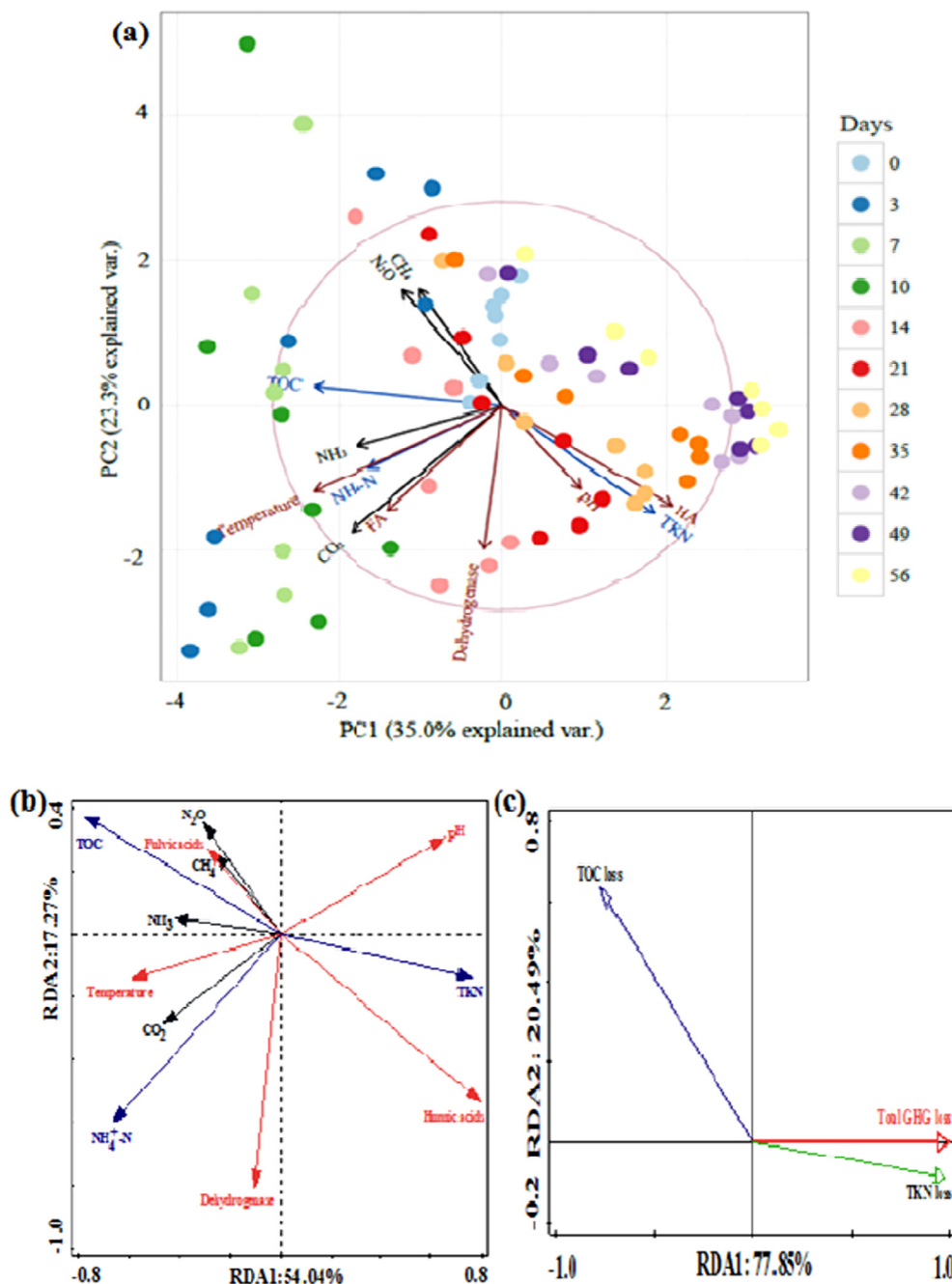
Fig. 4. (a) Principal component analysis where vectors correspond to the variables that define the treatments and points correspond to sampling during the composting of sewage sludge, (b) Redundancy analysis (RDA) of gaseous emission, humification and physicochemical properties and (c) RDA analysis of total greenhouse emission and its correlation with carbon and nitrogen losses during the sewage sludge composting. The correlation between gaseous emission and humification activity as well as physiochemical properties variables can be approximated by a perpendicular projection of the three different color variable arrow-tips onto the line overlaying the ordination axes. The length of the gaseous emission arrows is the multiple correlations of that humification and physicochemical properties with the ordination axes.

amendments were offset by the reduction of total N_2O and CH_4 emissions. Because CO_2 emission is inevitable as an indicator of rapid biogenic activity and which was excluded from the total GHG accounting. This result is similar to that of Beck-Friis et al. (2000) in organic household waste and Czekala et al. (2016) for poultry manure composting. Beside this, about 30.1% and 55.4% of initial total nitrogen was lost during SS composting; among all treatments most of initial total nitrogen was lost in the form of NH_3 (25.80–32.20%), 0.69–26.30 g/kg of initial total nitrogen was lost cumulatively in the form of N_2O and NH_3 which is 74.23–82.47% main source of nitrogen losses. This result and underline

region well explained in previous studies (Luo et al., 2013; Malinska et al., 2014; Jiang et al., 2016). Total combined N_2O–CH_4 emissions for LDB (2%, 4%, and 6%) added treatments were 49, 23, and 30 g/kg CO_2-eq. DM, respectively, which is higher compared to HDB applied treatments, while with the control or without biochar added treatment having the maximum total GHG emissions. However, total combined N_2O–CH_4 emission was reduced by biochar amendment, but total CO_2 emission reduction is another problematic issue of current study because more than 95% TOC loss by early CO_2 emission and recently major contributor of total GHG emission. Therefore, in terms of total N_2O–CH_4

emissions reduction, HDB addition would be strongly recommended as effective ways to SS composting, but further study is urgently required to reduce the CO_2 emission and TOC loss.

3.5. Composting parameters and its correlation with humification

The analysis of principal components (PC) and redundancy analysis (RDA) were considered with the gaseous emission, humification and physico-chemical variable shown in Fig. 4. The principal component 1 (PC1) accounted for 35.0% of the variance and significantly separated to three groups ($p < 0.0001$). The composting parameters important to determine the differences between the groups through PC1 were, fundamentally, CO_2 ($r = 0.98$, $p < 0.0001$), CH_4 ($r = 0.95$, $p < 0.0001$), NH_3 ($r = 0.97$, $p < 0.0001$), N_2O ($r = 0.94$, $p < 0.0001$) and TOC ($r = 0.96$, $p < 0.0001$). Those parameters that subscribe to intersect the second groups during the principal component 2 (PC2), which estimated for 23.3% of the variance, were pH ($r = 0.98$, $p < 0.0001$), TKN ($r = 0.98$, $p < 0.0001$), humic acid ($r = 0.95$, $p < 0.0001$) and fulvic acid ($r = 0.94$, $p < 0.0001$); while the third group were significantly different from each other ($F_{2,45} = 23.62$, $p < 0.0001$). The biochar added treatments initial feed stocks have slightly alkaline pH and contained high amounts of TOC loss through CO_2 emission and low amounts TKN losses with rapid humification. HDB amended treatments were characterized by low level of NH_3 and methane emission, while control treatment was identified by higher levels of total GHG and NH_3 emission ($p < 0.0001$), and TKN losses ($p < 0.0001$) during the composting process than those obtained at the end of experiment; control treatment showed lower levels of humification or HA and FA ($p < 0.0001$) than what was achieved via especially HDB amended treatments (Fig. 4b). Total GHG emission at the end of composting in control or without amended treatment was higher (108.61 g/kg) than the HDB added treatments between 2 and 4 g/kg; likewise, the percentage of nitrogen losses was also greater in the control (26.30 g/kg) than in the HDB applied treatments between 0.69 and 1.62 g/kg, respectively. All treatments presented significantly different parameters during the composting process (56th day); these differences, might be due to the significant variation of the C/N ratio and clumps formation which was also observed for gaseous emission between control, LDB and HDB applied treatments.

4. Conclusions

HDB treatments significantly reduced the NH_3, CH_4 and N_2O emission by 58.03–65.17%, 92.85–95.34% and 95.14–97.28% as compare to control treatments but not the CO_2, respectively. Although, the LDB addition had significantly the low NH_3 emission compare to control, but it cannot adequately buffered the compost, and then inhibited the humification rate. Furthermore, it was estimated that HDB could reduce the length of the active phase and enhance the humifcation with significant reduction of total N loss and GHG emissions. Overall, the addition of 12% HDB for composting demonstrated to be a beneficial practice for the management of SS.

Acknowledgements

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