



Effect of woody and non-woody bulking agents on biodrying of long-term storage sludge

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ABSTRACT

Storage sludge is associated with a range of environmental problems and biodrying such sludge via exothermic microbial decomposition can improve its stability and quality and decrease its mass. Bulking agents can be mixed with storage sludge to increase biodrying performance, but no studies have yet investigated differences between woody and non-woody bulking agents in the biodrying of storage sludge. This study investigated storage sludge mixed with four types of bulking agent, both woody (sawdust, wood shavings and pin chips) and non-woody (straw). Variations in temperature, moisture content, organic components, element contents, microbial populations and lignocellulase activity were measured under the laboratory scale for a period of 18 d. Straw performed better than woody bulking agents. It had the highest water removal capacity, followed by sawdust, wood shavings and pin chips. In such straw-sludge samples, the highest temperature reached 57.1°C, the moisture content decreased from 71.83% to 56.89%, and the volatile solids (VSs) decreased from 60.95% to 51.01% while the moisture content only decreased from 69.34% to 64.91% in pin chips-sludge samples. Water soluble components, hemicellulose and cellulose decreased while humus increased in all treatments during biodrying process, indicating that the biodrying process can improve the stability of storage sludge. Bacteria and actinomycetes had similar trends during the biodrying process, while fungi decreased. Carboxymethyl cellulase, lignin peroxidase and xylanase played key roles in lignocellulose degradation.

Keywords: Storage sludge; Biodrying; Woody and non-woody bulking agents; Organic matter; Microbial populations; Lignocellulase activity

1. Introduction

The long-term storage sludge (SS) is a type of sludge produced by urban sewage treatment plants and deposited in sludge landfill for many years while undergoing only very simple toxic treatment. Due to long-term accumulation, the characteristics of the organic matter and microbial communities existing in SS differ from those of raw sludge. First, the organic matter content of SS is lower than that of raw sludge [1–4]. Likewise, the microbial population of SS is less than that of raw sludge, while more anaerobic microorganisms are found in SS [3].

In recent years, with the increase of sludge yields, more and more SS are being produced, and it has caused serious environmental problems such as land occupation, soil and groundwater pollution, and even threats to the health of nearby residents. Therefore, dealing with SS is an urgent issue. Despite depositing in the sludge landfill for long time, the moisture content is still as high as 75%–85% owing to the precipitation [3]. However, the moisture content of sludge should be less than 60% if it is to be used for sanitary landfills, and less than 45% if used as a barrier material [2,5–8]. Hence, an additional dewatering step is often needed to reduce the moisture content to a level that enables SS to be safely disposed of [8].

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Biodrying is a novel alternative method of composting to reduce the moisture content of waste material [9–13]. The main mechanism of biodrying is convective evaporation, which uses the bio-heat generated by microbial aerobic degradation of organic compounds to evaporate water. This is more economical and energy-saving than traditional thermal drying [13–17]. Moisture reduction during the biodrying process occurs as follows: water molecules evaporate from the material surface into the air, and is transported and removed by airflow [11,17]. Although biodrying technology has been widely used for raw sludge and anaerobically digested sludge pre-treatment in recent years [18,19], few studies have been reported for SS treatment [20,21]. It was found in our previous study that the stability of the SS can be improved after biodrying [20]. Biodrying can fix toxic pollutants in SS, and its volume can be considerably reduced by the evaporation of water. Additionally, the process can enhance organic matter degradation and promote deep stabilization of SS [14,20], which makes SS more suitable for subsequent resource utilization.

It is well known that bulking agents play a significant role during the biodrying or composting processes [22–24]. Bulking agents are usually mixed with waste materials to adjust its moisture content and C/N ratio, and provide structural support and high free air space, which allows easier air movement [24–26]. Due to the relatively low organic matter content, high moisture content, and the lack of porosity of SS, the contribution of bulking agent is much more important in the SS biodrying process [14,23]. Wheat straw, sawdust, wood chips, and shredded rubber are frequently recommended as bulking agents for biodrying or composting of dewatered sludge [22–24]. Many reports have focused on the biodrying of sewage sludge, anaerobically digested sludge, and municipal solid waste [14,23,27]. However, few studies have considered the effect of bulking agents in the SS biodrying process [20,21], let alone distinguished between woody and non-woody bulking agents.

Microorganisms play an important role in biodrying system, as the water is removed by the bio-heat which is generated by the aerobic microbial fermentation [1]. Biodrying is a dynamic process and the presence of specific microorganisms can indicate its progress [14,22,28]. Although microorganisms in the composting system have been studied by many researchers [29–32], variations in microorganisms involved in biodrying have not been fully investigated. Microorganisms and enzymes play key roles in the degradation of organic

compounds. The substrates in the biodrying system are degraded by microorganisms that secrete enzymes to hydrolyze macromolecular substrates and degrade monomer hydrolytes. Bacteria are mainly responsible for the degradation of easily degradable substrates. Most of fungi and actinomycetes are involved in the degradation of lignocelluloses by producing lignocellulase [10,19,33,34]. Variation in enzyme activities is consistent with the level of organic matter degradation, that is, the higher the enzyme activity, the greater the organic losses. Amylase, protease and lipase are responsible for the degradation of easily degradable substrates while carboxymethyl cellulase (CMCase) is involved in the degradation of cellulose. Xylanase and lignin peroxidase mainly degrade hemicellulose and lignin, respectively [19]. Therefore, studying the evolution of enzyme activities could help to understand the biodegradation of substrate after biodrying. However, while many reports have focused on protease, amylase and lipase, etc. [10,12,27,35], few have investigated lignocellulase [19]. Accordingly, this study will investigate the role of lignocellulase in biodrying.

The objectives of this study are to: (1) evaluate the effect of woody (sawdust, wood shavings and pin chips) and non-woody (straw) bulking agents on the biodrying process; (2) investigate the variations in organic matter composition and organic elements; and (3) analyze the evolution of microbial populations and lignocellulase activity during the biodrying process.

2. Materials and methods

2.1. Materials

The SS used in this study was obtained from a sludge landfill in Shenyang, China. Sawdust, wood shavings and pin chips were obtained from a local timber mill in Harbin, China. Straw was collected from the nearby farmland in Harbin, China. The bulking agents and SS were mixed to obtain a moisture content of approximately 70%. Characteristics of the raw materials are outlined in Table 1.

2.2. Experimental design

The biodrying experiments were performed in four cylinder reactors made of PVC plastic with an inner height of 600 mm and a diameter of 400 mm (Fig. 1). The effective volume of each reactor was approximately 56L. The outer

Table 1
Characteristics of the raw materials

Parameters	Raw sludge	Storage sludge (SS)	Sawdust	Wood shavings	Pin chips	Straw
pH	7.52	6.93	6.31	6.53	6.42	6.04
Moisture content (wet basis, %)	80.14	75.92	7.94	6.11	5.80	1.25
VS content (dry basis, %)	69.08	40.56	99.00	99.12	98.13	88.19
C content (dry basis, %)	32.34	17.34	45.68	47.76	47.42	40.39
N content (dry basis, %)	3.50	2.82	0.39	0.28	0.20	0.93
H content (dry basis, %)	3.80	3.45	4.86	5.17	4.88	4.54
S content (dry basis, %)	1.67	1.45	0.12	0.19	0.25	0.75
C/N ratio	9.24	6.14	117.13	170.57	237.10	43.34
C/H ratio	7.19	5.03	9.40	9.24	9.72	8.90

wall was covered with a 15 cm layer of polyurethane foam to reduce heat loss. A layer of straw was used to cover the matrix at the top of the column to prevent heat loss and vapor condensation. A perforated baffle with a 2-mm hole was fixed above the bottom of each reactor to support the matrix and promote ventilation. Three sampling pots were located on one side of each reactor [36].

Four trials with different bulking agents were run with a ratio of 5:1 (w/w, wet basis) for storage sludge and bulking agents: storage sludge/sawdust (T-1), storage sludge/wood shavings (T-2), storage sludge/pin chips (T-3) and storage sludge/straw (T-4). The aeration rate was 1.4 L/(min kg wet basis) with 10 min run/20 min stop during the biodrying process. The matrix was homogenized on the ninth day by turning. All experiments were run for a period of 18 d.

2.3. Sampling and analytical methods

To ensure representative sampling, the samples were collected from three different depths of the matrix every 3 d and mixed for analysis. Analyses were performed in triplicate for all samples. Moreover, the calibration curves were established each time when the enzyme activities were determined.

During the biodrying process, the matrix temperature was monitored by a thermometer (GS200-ET, Zhituo Co., Zhejiang, China) with sensors located at the top, middle and bottom points and the average was reported. The moisture content of samples was determined by drying the samples at 105°C for 24 h [37]. The content of VS was analyzed by heating the sample at 550°C for 5 h in a muffle furnace [37]. The C, N, H and S contents in the materials were analyzed using an element analyzer (Vario EL III, Elementar, Germany).

The organic matter components, including the water soluble components (WSCs), lipids, hemicellulose, cellulose, lignin and humus contents were analyzed by the fractional extraction and gravimetric method as previous description [38]. The WSCs were extracted with a neutral detergent at 100°C for 60 min and the hemicellulose fraction was extracted with an acid detergent. Cellulose was obtained using strong acid extraction and lignin was obtained by ashing. Humus was extracted by Na₄P₂O₇ and NaOH solution and measured by K₂Cr₂O₇ volumetric method [38]. Lipid (LIP) was measured by acid-hydrolysis method [38].

Bacteria, fungi and actinomycetes were counted by the plate dilution method [39]. CMCase activity was determined

according to Nakamura and Kitamura [40]. Xylanase activity was measured using the dinitrosalicylic acid method [30]. Lignin peroxidase activity was measured using the visible spectrophotography [30]. One international unit (IU) of lignin peroxidase, CMCase and xylanase activity was defined as the amount of enzyme required to release 1 μmol of veratryl aldehyde, glucose and xylose, respectively, per minute under the given assay conditions.

2.4. Statistical analysis

All statistical analyses were performed using SPSS 20.0 (SPSS, Inc., Chicago, USA). The statistically significant difference between the two groups was compared by *t*-tests and were considered statistically significant at *p* < 0.05.

3. Results and discussion

3.1. Temperature evolution during the biodrying process

Temperature is a crucial parameter affecting water evaporation and organic substance degradation during the biodrying process [12,13,35]. The variation in average temperature is presented in Fig. 2. The temperature profiles of the four trials presented the typical temperature changes:

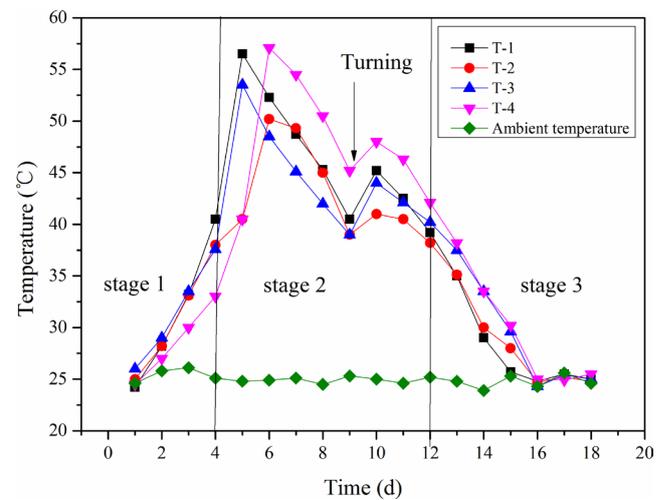


Fig. 2. Variation of temperature during the biodrying process.

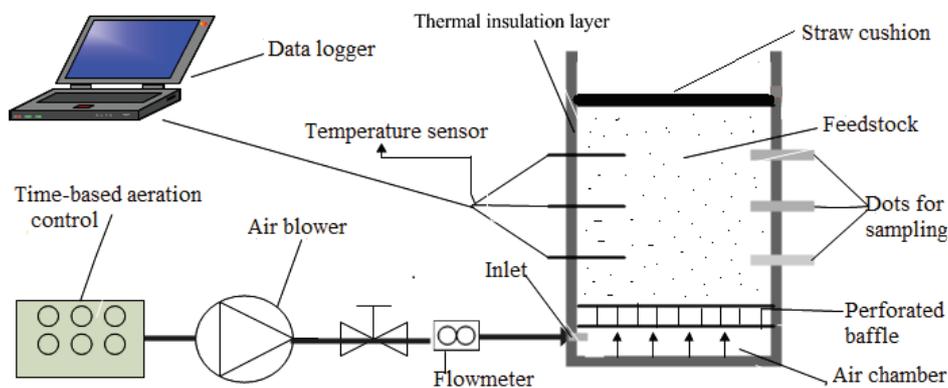


Fig. 1. Schematic diagram of the biodrying system.

stage 1 (heating stage), stage 2 (thermophilic stage) and stage 3 (cooling stage).

The T-1, T-2, T-3 and T-4 trials reached their highest temperatures on day 5 (56.5 °C), day 6 (50.2°C), day 5 (53.5°C) and day 6 (57.1°C), respectively. The matrix temperature was relatively lower than that in composting process due to a larger aeration rate [23]. Notably, the T-4 trial reached the highest temperature (57.1°C) and achieved the longest thermophilic stage (>6 d), followed by trials T-1, T-3 and T-2.

For all trials, 5 or 6 d were needed to reach the peak value, which is longer than reported in previous studies (2–3 d) [12,13,23,41], which can be attributed to the characteristic of SS. Because the physicochemical properties and microbial activity of SS had changed because of long-term stacking in sludge landfill [3].

The highest temperature and the longest thermophilic stage of T-4 trial can be attributed to the large biodegradation potential of straw. It has been reported that straw has substantial biodegradation potential in the aerobic process, while woody materials have poor capacity to be degraded [23,42]. It was found that the biodegradation of the bulking agent contributed a much higher proportion of bio-generated heat during the biodrying process than that of sludge [14,20,21], which was consistent with our previous study [21]. We found that the contribution of the bulking agent could account for 82.35%–86.67% of the bio-generated heat in biodrying system [21]. Li et al. [14] also found that wheat residues contributed approximately 86.01% of bio-generated heat during the biodrying process. Additionally, Zhao et al. reported that slowly biodegradable fraction came from some bulking agents rather than sludge itself, which contributed substantial amounts of bio-generated heat to enhance the matrix temperature in sludge biodrying or composting processes [23]. Thus, the role of bulking agents was very important during the biodrying process and the performance of T-4 was better than that of the other trials, which all used woody materials as bulking agents. Therefore, compared with woody materials, straw was more suitable as bulking agent for the biodrying of storage sludge used in this study.

3.2. Moisture content evolution with different bulking agents in SS biodrying

The main purpose of biodrying is to utilize thermal energy, generated by aerobic degradation of organic matter in waste, to evaporate water, thus achieving self-drying [4,15]. The variation of moisture content is shown in Fig. 3.

Moisture contents decreased from $71.51\% \pm 0.12\%$, $69.60\% \pm 1.01\%$, $69.34\% \pm 0.78\%$ and $71.83\% \pm 0.45\%$ to $61.41\% \pm 0.76\%$, $57.89 \pm 0.56\%$, $64.91\% \pm 0.47\%$ and $56.89 \pm 0.78\%$ for trials T-1, T-2, T-3 and T-4, respectively, representing reduction rates of 14.12%, 16.82%, 6.4% and 19.68%, respectively. This was because air carried moisture from bottom to top of the matrix. Thus, the dried layer extended to the top little by little, which drove moisture loss [13,43]. T-4 trial had the highest moisture reduction rate mainly due to the generation of greater bio-heat, as the factors affecting moisture removal were mainly bio-heat and ventilation mode, and the ventilation modes used in all trials were the same in this study.

Additionally, moisture content change was corresponding to the temperature variation (Fig. 2) and the water reduction was greatest in stage 2. Thus, stage 2 was the main functional period. Statistical analysis showed the type of bulking agent had a significant influence on the water removal ($p = 0.001$) through modifying moisture content, free air pore volume and soluble organic matter content.

3.3. Organic matter variation during the biodrying process

3.3.1. Organic element variation during the biodrying process

The variation in organic elements (C, N, H and S) during the biodrying process is outlined in Table 2. Organic elements contents decreased during the biodrying process, indicating that oxidation, dehydrogenation and desulfurization

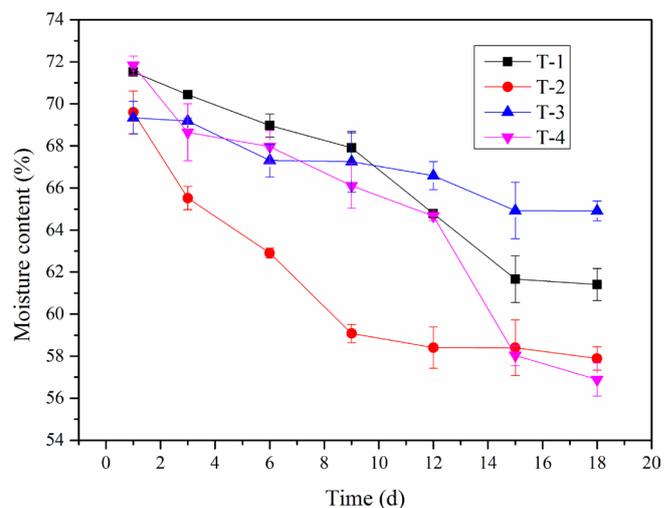


Fig. 3. Variation of moisture content during biodrying process.

Table 2
Elements content variation during biodrying process

Parameters	T-1		T-2		T-3		T-4	
	Initial	Final	Initial	Final	Initial	Final	Initial	Final
C (%)	25.08 ± 0.66	20.94 ± 0.46	25.29 ± 0.68	22.44 ± 0.57	24.85 ± 0.89	22.79 ± 0.59	23.65 ± 0.97	18.06 ± 0.55
N (%)	3.60 ± 0.21	3.48 ± 0.33	3.61 ± 0.35	3.52 ± 0.11	3.55 ± 0.28	3.51 ± 0.17	3.70 ± 0.11	3.50 ± 0.06
H (%)	3.81 ± 0.11	3.43 ± 0.29	3.84 ± 0.11	3.64 ± 0.19	3.69 ± 0.36	3.53 ± 0.08	3.71 ± 0.09	3.19 ± 0.10
S (%)	2.38 ± 0.09	1.80 ± 0.26	1.94 ± 0.08	1.73 ± 0.05	1.95 ± 0.07	1.80 ± 0.05	1.96 ± 0.09	1.77 ± 0.07
C/N	6.96	6.01	7.00	6.37	7.00	6.49	6.39	5.16
C/H	6.58	6.10	6.58	6.16	6.73	6.45	6.37	5.66

occurred [14,44]. The C content of T-4 decreased from 23.65% to 18.06%, and the N content decreased from 3.70% to 3.50%, which was mainly due to the degradation of organic compounds. Compared with other trials, the final samples in T-4 contained lower contents of C, N, H and S, indicating higher degree of maturity. However, some studies have reported that the N content increased which was inconsistent with our result [45,46], which indicated that in the present study the rate of degradation of N-containing compounds was greater than the rate of their formation.

The C/N and C/H ratios are also described in Table 2. The C/N ratio decreased in all trials during the biodrying process, which was similar to previous studies [45–48], possibly due to strong degradation of organic carbon compounds. C/N ratio corresponds to the maturity degree of the studied organic material (compost, soil, sewage sludge) [48], and, thus, a decrease in the C/N ratio indicated that the mineralization and stabilization of organic waste were enhanced through biodrying. The final samples in T-4 achieved the lowest C/N ratio of 5.16, implying that T-4 had a higher maturity degree than trials T-1, T-2 and T-3.

The C/H ratio is a useful qualitative parameter for comparing the aromaticity of the matrix [46]. Biodrying also caused a decrease in C/H ratio, which was similar to the previous reports on biodrying [14] that also suggested strong degradation of organic carbon compounds. This contrasted with results for compost [44,45], indicating that composting was more conducive to the formation of aromatic compounds than

biodrying, because of its longer composting cycle. Overall, the stability and maturity of the matrix were improved by biodrying, and trial T-4 achieved the best stability and maturity. Additionally, we can infer that the degree of aromaticity of the biodrying products was lower than that of the composting products because of the shorter duration of biodrying.

3.3.2. Organic component evolution during the biodrying process

The variation in organic components is outlined in Fig. 4. At the beginning of all trials, water soluble fraction comprised the highest proportion of organic components, followed by cellulose, hemicellulose, lignin, humus and lipid. After biodrying, the proportions of WSC, hemicellulose, cellulose, lignin and lipid decreased, while humus increased, indicating that the materials gradually became stable and mature, as the increased humus is associated with the stability and maturity of organic matter [44]. The humus content in T-4 increased from 9.87% to 15.89%, and it increased from 9.96% to 15.27% in T-1. The reduction of WSCs of T-4 decreased from 25.93% to 16.26% and the reduction proportion was 9.67%, followed by T-1 (6.48%), T-2 (4.77%) and T-3 (3.71%), indicating that T-4 achieved the highest proportion of reduction. The remarkable reduction in WSCs indicated that they were easily biodegradable organic compounds during the biodrying process [1]. Hemicellulose, cellulose and other refractory materials showed slight declines, such

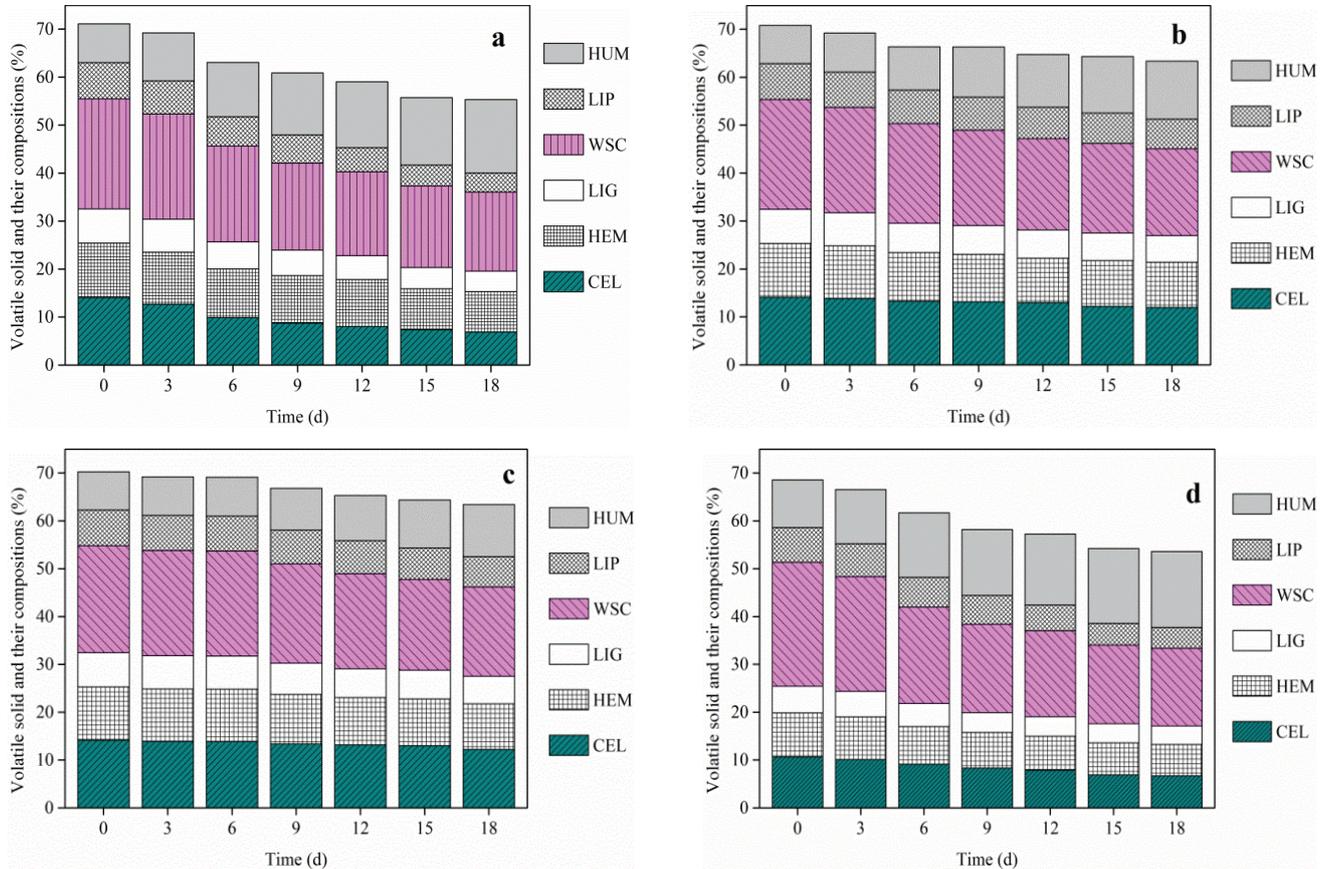


Fig. 4. Evolution of organic component during biodrying process: (a) T-1; (b) T-2; (c) T-3; (d) T-4.

as the highest reduction proportion of hemicellulose was only 2.63% among all treatments, indicating that they can be partially degraded and transformed [22].

Organic matter contents decreased in all trials, indicating that biodrying leads to the degradation of organic matter due to the degradation of aerobic microorganisms. Organic matter contents in all trials decreased by 11.28%, 7.46%, 6.52% and 14.96% for T-1, T-2, T-3 and T-4, respectively. Organic matter in T-4 got the best degradation effect while the effect of T-3 was the worst.

Overall, the performance of the T-4 trial was better than that of the other trials, which was mainly because the straw used contained more easily degradable organic matter than the woody bulking agents used in the other trials [23]. Therefore, straw was most suitable for biodrying. Statistical analysis showed that there were significant differences between the organic matter contents of the four trials ($p < 0.001$).

3.4. Evolution of microbial populations and lignocellulase activity during the biodrying process

3.4.1. Changes in the microbial populations during the biodrying process

The temporal evolution of microbial populations is shown in Fig. 5. In all trials, bacterial populations increased

first and then decreased gradually until the end of the process (Fig. 5(a)). T-4 obtained the highest value, which could indicate that straw was favorable to the establishment of dominant aerobic bacteria. However, compared with T-4, the woody bulking agents did not cause greater bacteria proliferation. This was mainly because straw contained large amounts of easily degradable organic matter while the woody bulking agents contained lots of lignin, cellulose and other refractory organic compounds which were not easily used by bacteria [23].

The variation of actinomycetes in the biodrying system is presented in Fig. 5(b). Although the actinomycetes populations decreased at the late stage, it still remained at a relative high level. This was because actinomycetes can decompose cellulose, lignin and other refractory organic compounds [19], which were the main available nutrients during the late stage. T-4 reached the maximum value of 12 log₁₀ CFU/g on the 6th day, which was significantly higher than other three trials.

Fungi not only secrete enzyme and hydrolyze organic substances but also cause physical damage to materials through their mycelium machinery alternation, which makes it more useful to the decomposition of organic matter [49]. In contrast to bacteria and actinomycetes, fungi presented a downward-upward-downward trend (Fig. 5(c)). Due to the poor heat tolerance of most fungi, the number of thermophilic fungi was very little in the biodrying system. Tiquia

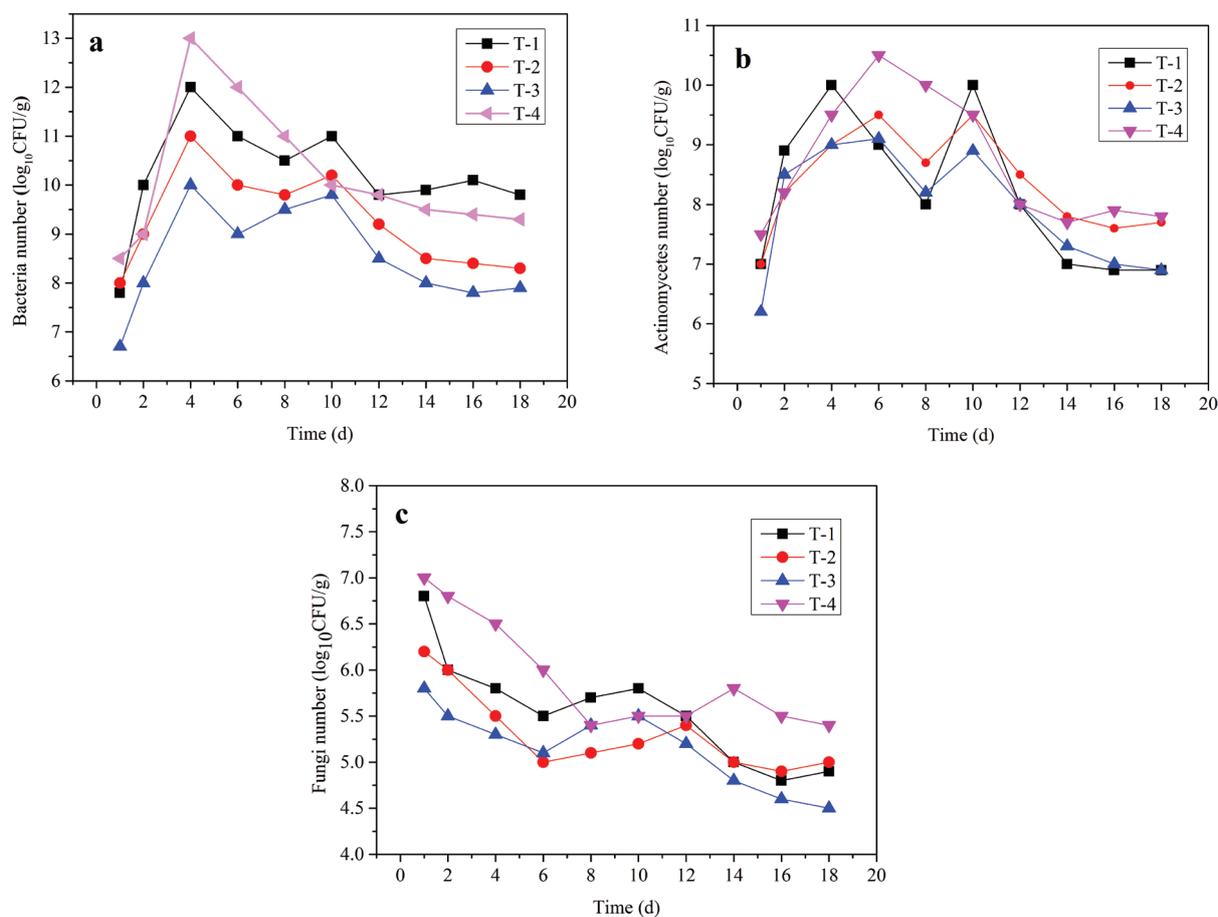


Fig. 5. Variation of microbial populations during biodrying process: (a) bacteria; (b) actinomycetes; (c) fungi.

[50] reported that most fungi were eliminated when temperature exceeded 50°C but the populations recovered when temperature decreased to below 45°C. Therefore, mesophilic fungi reappeared in the material following the end of the thermophilic period.

In brief, in early stage of biodrying, bacteria were the major microorganisms, which were responsible for the initial decomposition of organic matter and the generation of bio-heat [14,22]. Statistical analysis showed that the type of bulking agents had a significant influence on the microbial populations ($p < 0.004$).

3.4.2. Lignocellulase activity analysis

Temporal evolutions of CMCase, xylanase and lignin peroxidase activities are outlined in Fig. 6. The enzyme activity was proportional to the amount of the degrading substrates [27]. The variation in lignocellulase activity, which is responsible for the degradation of recalcitrant substrates, was different from that of the other enzymes (i.e., amylase, protease and lipase) which was responsible for the degradation of easily degradable organics [27]. CMCase activity decreased while xylanase and lignin peroxidase activities increased during the biodrying process in all treatments. The enzyme activities in trial T-4 were higher than those of the other three

trials, which was corresponding to microbial populations (Fig. 5), as the substrates were degraded by microorganisms to secrete enzymes. In trial T-4, CMCase activity decreased from 0.85 to 0.52 IU g⁻¹. In contrast, lignin peroxidase activity increased from 0.48 to 0.83 IU g⁻¹ and xylanase activity increased from 0.63 to 1.78 IU g⁻¹. Statistical analysis showed that bulking agents had a significant influence on enzyme activity ($p < 0.001$).

CMCase activity had higher value in the first two stages (heating and thermophilic stages), while lignin peroxidase and xylanase activities were lowest in stage 2 (Fig. 2). Enzyme activities indicated that higher temperature may favor CMCase activity, while lower temperature may facilitate lignin peroxidase and xylanase activities, which suggested that CMCase played a key role in the first two stages, whereas lignin peroxidase and xylanase played key roles during the second thermophilic phase and the cooling phase. This could explain why cellulose showed its highest degradation rate between days 3 and 12 (Fig. 4). Lignin peroxidase and xylanase were responsible for the degradation of lignin and hemicellulose, respectively. They experienced lower degradation during the initial stage of biodrying and then showed significant degradation (Fig. 4), which agreed well with the changes in enzyme activities.

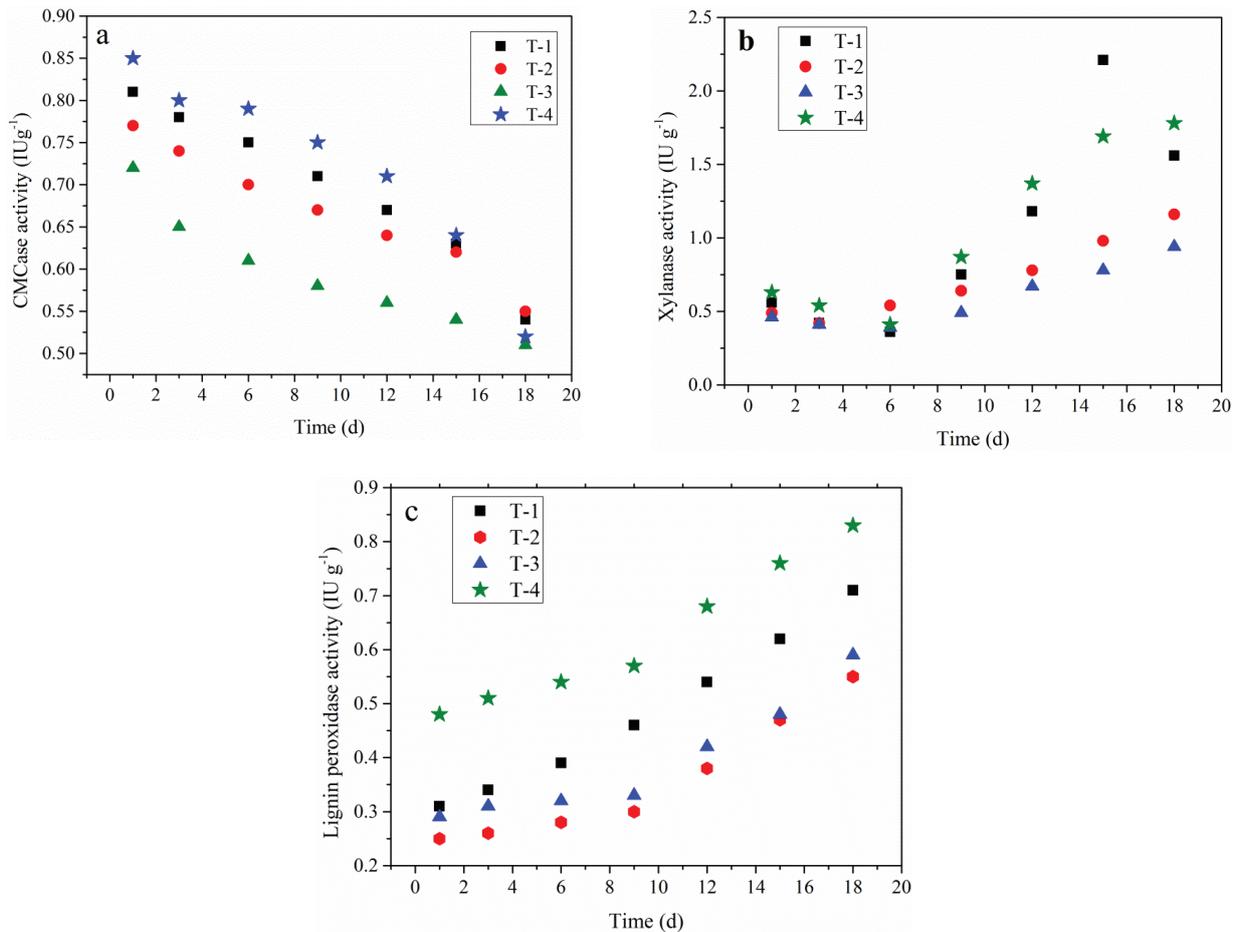


Fig. 6. Temporal evolution of lignocellulase activity during biodrying process: (a) CMCase activity; (b) xylanase activity; (c) lignin peroxidase activity.

4. Conclusion

The type of bulking agents had a strong effect on the biodrying of storage sludge. The non-woody bulking agent (straw) achieved the best performance by the highest temperature (57.1°C), longest thermophilic stage (>6 d) and highest water removal capacity (19.68%), indicating that it was more suitable for biodrying than woody bulking agents. All organic component contents decreased except for humus, indicating that biodrying could cause an increase of stability and maturity of SS, which was also indicated by the decrease in the C/N ratio. It was inferred that the degree of aromaticity of the biodrying products was lower than that of the composting products. Bacteria and actinomycetes had similar dynamics (upward-downward), which were different from that of fungi (continuous declination). Lignocellulase was mainly responsible for the degradation of recalcitrant substrates, such as lignocelluloses.

Acknowledgments

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