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Biodrying of storage sludge and analysis of the stability and agricultural properties of biodried products

Tiantian Liu, Chongwei Cui, Junguo He*, Jian Tang

School of Municipal and Environmental Engineering, Harbin Institute of Technology, Harbin 150090, China, emails: junguohe@263.net (J. He), tian080816@163.com (T. Liu), cuichongwei1991@126.com (C. Cui), tangtang100@126.com (J. Tang)

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ABSTRACT

Biodrying was first used for treatment storage sludge by adding bulking agents to reduce its moisture content and improve its quality and stability. The influence on the stability and agricultural application of biodried products after adding bulking agents was investigated by analyzing the variation of the AT_4 index, organic matter, and heavy metals of biodried products. Two treatments were considered: the addition of straw (trial A) and sawdust (trial B) to storage sludge as bulking agents before biodrying. After 18 d of biodrying, trial A with straw as bulking agent achieved the higher water-removal rate by 19.24% and also had the higher matrix temperature (57°C). Additionally, the content of protein decreased by 38.45%, while the polysaccharide content only decreased by 5.96%. Additionally, the content of humic substances increased by 48.58%. Further investigation showed that most of the heavy metals transformed from acid soluble fraction to oxidizable and residual fractions, the contents of heavy metals increased insignificantly (6.29%–10.81%), and Zn content exceeded the maximum permissible value stipulated for soil in China (GB 4284-1984). Thus, the risk for farm-oriented storage sludge still exists.

Keywords: Storage sludge; Biodrying; Bulking agents; Bioproducts; Stability; Agricultural risk

1. Introduction

Long-term storage sludge is the sludge which is arbitrarily stacked in sludge landfill sites, undergoing only very simple toxic treatment for many years. In recent years, with the increase of sludge yield, more and more storage sludge has been produced and caused serious environmental problems such as land occupation, soil, and groundwater pollution and even threats to the health of the residents in surrounding areas. Therefore, dealing with storage sludge is an urgent issue. As the storage sludge is placed in an open field, its moisture content is still high (about 80%) due to the impact of rainfall [1]. However, the moisture content of sludge should be less than 60% if it is to be used for sanitary landfills and land use, and less than 45% if used as barrier material [2–4]. The sludge can be coincinerated when the moisture content is about 60%–80%, and sludge (around 14 MJ/kg dry mass) with 50% moisture content can be incinerated without adding any fuel [4,5]. Therefore, it is important to choose a suitable method to reduce the volume and quantity of the storage sludge and improve its stability, and biodrying can be considered [6].

Biodrying, which is derived from but differs from composting, is a novel sludge reduction method and was first proposed as a pretreatment method of solid waste in the 1990s [7,8]. The aim of biodrying is to achieve the highest water removal in a short duration [9–12]. Traditional drying technology uses an external heat source to cause the water in the material to evaporate [3] while biodrying reduces the water of material using its own bioheat without an additional heat source [13]. Therefore, it is cost-effective and environmentally friendly [6]. Although many studies have reported that biodrying has been widely used as a reduction method for raw sludge and anaerobically digested sludge [9,21] before the biodried products can be used for landfilling, incineration, and land utilization [14], few researches have focused on the biodrying of storage sludge.

* Corresponding author.

Because of the long-term stacking that occurs in the environment, characteristics of the organic matter and microbial composition of storage sludge differ from raw sludge. First, due to the degradation action of microbes during the long process of stacking, the content of organic matter in storage sludge is lower than that in raw sludge [1,4,10,14,15], especially the easily degradable organic matter. Second, the microbial population of storage sludge is also less than that in raw sludge, which is mainly due to the addition of lime and other fungicides in the process of stacking [1]. Additionally, the lack of oxygen in the middle of the sludge landfill sites results in the decrease of aerobic or facultative aerobic microorganisms and a greater number of anaerobic microorganisms are found in the storage sludge [1].

As a result of the differences found in raw sludge compared with storage sludge, the performance of storage sludge biodrying will be different from raw sludge. Thus, investigating the biodrying of storage sludge could be extremely useful and important. If the biodrying of storage sludge is feasible, the volume and quantity can be reduced and the harmfulness of the toxic pollutants can be minimized; additionally, the degradation of organic matter and its deep stabilization would be promoted, which would be conducive to the subsequent disposal [4].

It is well-known that the addition of bulking agent to the waste materials can modify properties, improve C/N ratio, provide structural support, provide high free air space, and adjust moisture content [15–20]. Although many reports have focused on the biodrying of sewage sludge, anaerobically digested sludge, and municipal solid waste [20–22], few studies have studied the effect of bulking agents in the storage sludge biodrying process.

Just like composting, biodrying also involves the biostabilization of organic waste materials, and biodrying stability is related to the bioavailability of organic matter [23]. Degradable organic matter is mineralized to carbon dioxide, ammonia, and water, and other organic matter is transformed into refractory humic substances (HS) [24]. Therefore, HS can reveal the stabilization and maturity of organic matter. HS include humic acid (HA), fulvic acid (FA), and other components, such as humin [25]. The HA content usually increases while the FA content decreases during composting [26]. The reports on humus variation in biodrying are very few [4,25], while many studies have been conducted on composting [4,27,28]. Therefore, the investigation into the changes of HS during the biodrying process has important guiding significance for the stability and maturity of dried products.

This paper investigates storage sludge biodrying, and the aims of the study are to compare the biodrying effect of different bulking agents (sawdust and straw); to study the stability of biodried products; and finally, to evaluate the feasibility of farm-oriented biodried products.

2. Materials and methods

2.1. Materials

The storage sludge was obtained from a sludge landfill site in Shenyang, China. Sawdust was obtained from a local timber mill in Harbin, China, and straw was collected from nearby farmland in Harbin. The bulking agents and storage sludge were mixed to adjust the moisture content to about 70%, constituting the biodrying material. The characteristics of the raw materials are outlined in Table 1.

2.2. Experimental equipment and process operation

The biodrying experiments were performed in two cylinder reactors (Fig. 1) made of PVC plastic, with an inner height

Table 1
Characteristics of storage sludge and bulking agent

Parameters	Storage sludge	Sawdust	Straw
pH	6.93	6.31	6.04
Moisture content (wet basis, %)	75.92	7.94	1.25
VS content (dry basis, %)	40.56	87.13	89.12
C content (dry basis, %)	17.34	45.68	40.39
N content (dry basis, %)	2.12	0.39	0.93

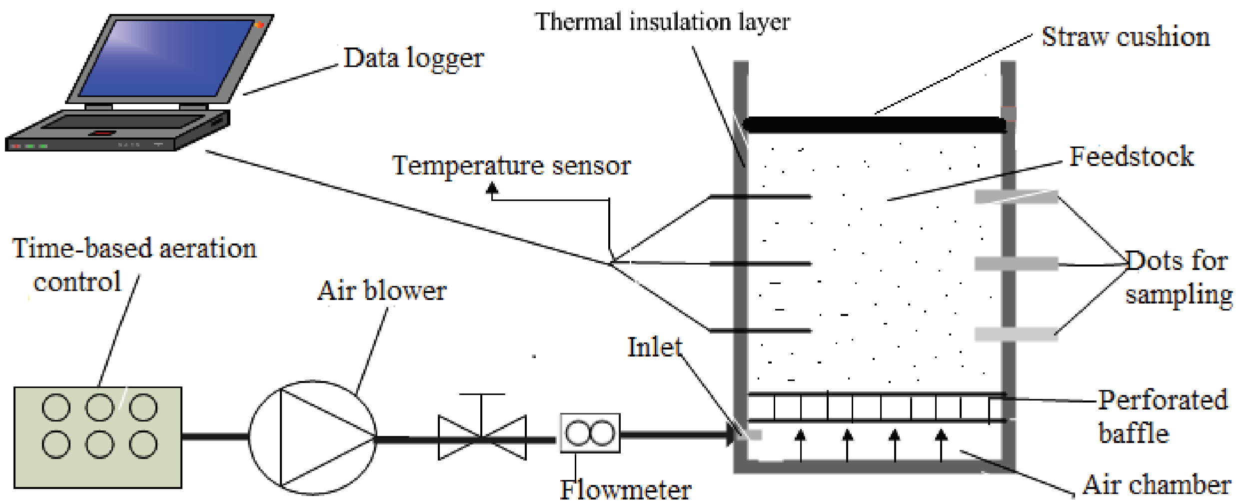

Fig. 1. The schematic diagram of the biodrying system.

of 600 cm and a diameter of 400 cm. The effective volume of the reactors was approximately 56 L. The outer wall was covered with a 15-cm layer of polyurethane foam to reduce heat loss. A layer of straw was covered on 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 the reactor to support the matrix and promote ventilation. Three sampling pots were located on one side of the reactor [29].

The aeration rate was 640 L/h with 10 min run/20 min stop during the biodrying process. The matrix was homogenized by turning on the ninth day. All experiments were conducted over a period of 18 d. Two trials with different bulking agents were run: storage sludge/straw (A) and storage sludge/sawdust (B).

2.3. Sampling and analysis

To ensure representative sampling, samples were collected from three different depths of the matrix for every 3 d and divided into two parts: one part was preserved at 4°C and the other part was dried and sieved for analysis.

During the biodrying process, the matrix temperature was monitored using 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 the samples was determined by drying the samples at 105°C for 24 h using the Standard Method [30]. The content of volatile solid (VS) was analyzed by heating the sample at 550°C for 5 h in a muffle furnace using the Standard Methods [30].

AT_4 is the cumulative consumed oxygen (O_2) per unit of dry matter (DM) in 4 d (mg O_2/g DM). AT_4 was used to determine the biological stability of the dried products. First, 10 g samples were evenly spread on the bottom of the 1 L conical flask and incubation was performed without inoculum at 35°C for 4 d in sealed bottles. The air in the bottles was deflated every day, and the O_2 concentration was determined with a CYS-1 O_2 analyzer (Xuelian Co., China). Then, the inner air was replaced by ambient air and the bottles were sealed again for successive incubation [31].

The protein was determined using the bicinchoninic acid (BCA) protein concentration assay kit (Sigma, BCA1) from the Sigma-Aldrich Company (Shanghai, China), using the improved Lowry method [32]; and the polysaccharide was determined using the anthrone sulfuric acid method [33]. Humus and its composition were extracted by 0.1 mol/L $Na_4P_2O_7$ and 0.1 mol/L NaOH solution and measured by $K_2Cr_2O_7$ volumetric method [24]. The content of organic elements (C, H, S, and N) was analyzed using an element analyzer (Vario EL III, Elementar, Germany).

The content of heavy metals was measured by atomic fluorescence spectrometry using wet digestion and the heavy metal fraction was extracted by the modified Community Bureau of Reference (BCR) sequential extraction scheme [34].

3. Results and discussion

3.1. Performance of the storage sludge biodrying process of different bulking agents

The mean temperature of the bottom, middle, and top of the matrix during the storage sludge biodrying process with different bulking agents is shown in Fig. 2(a). The mean temperature increased and reached the first peak value of 57.1°C and 52.5°C on the fourth and fifth days respectively, for A and B after which a tendency to decrease was observed and a slight upward tendency was presented on the ninth day as a result of turning the material. Due to greater aeration

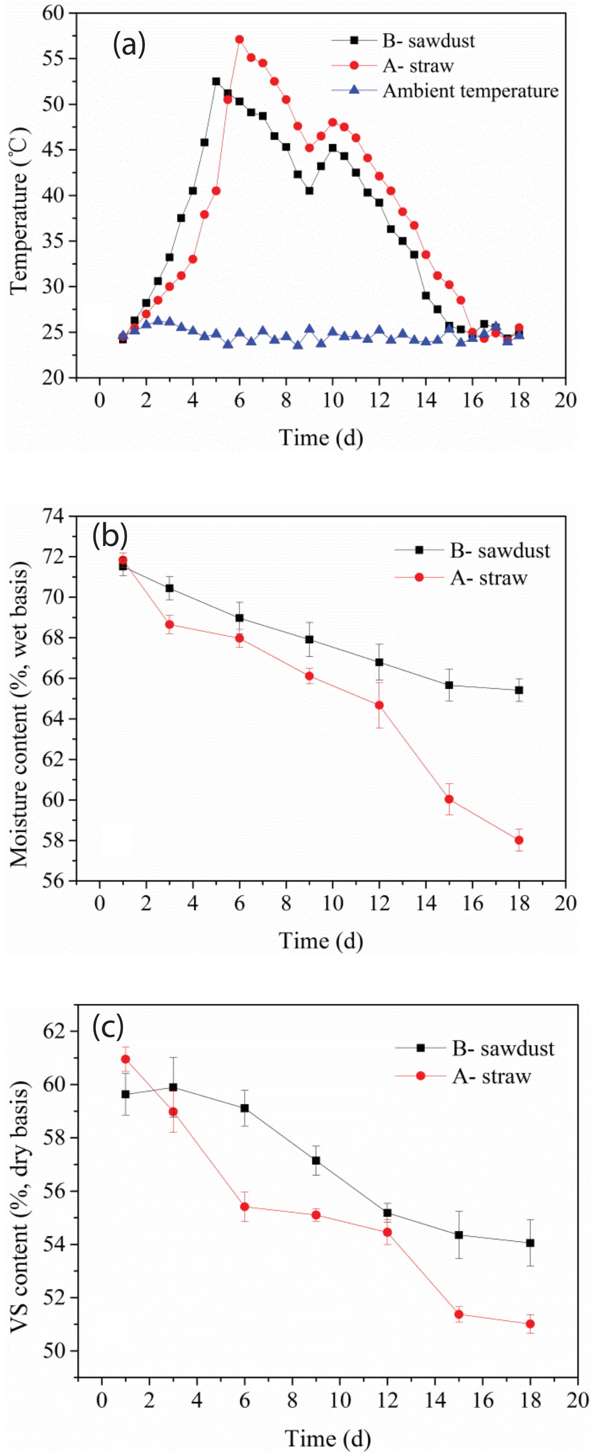

Fig. 2. Performance of the storage sludge biodrying process: (a) mean temperature; (b) moisture content; and (c) VS content.

that would be in a typical composting process, the temperature of the storage sludge biodrying process was relatively low [15,21]. The temperature reached as high as 57.1°C when straw was used as the bulking agent while only 52.5°C was observed for sawdust. The temperature of A was higher than B, indicating that the straw was more conducive to maintaining a higher temperature. Straw contains more easily degradable organic matter than sawdust and the contribution of the bulking agent to the biogenerated heat (about 80%) in the biodrying system is more than sludge (about 10%–20%) [15,21]. Therefore, the heat generated by the degradation of organic matter in straw could maintain a higher temperature, which was similar to the previous report [15].

In turn, the high temperature of the reactor was conducive to the rapid evaporation of water, and the moisture content during the storage sludge biodrying process is shown in Fig. 2(b). The moisture content of A decreased from 71.83% ± 0.35% to 58.01% ± 0.54% and that of B decreased from 71.51% ± 0.45% to 65.41% ± 0.56% during the biodrying process. The variation of VS content during the biodrying process is presented in Fig. 2(c). The VS content of A decreased from 60.95% ± 0.46% to 51.01% ± 0.35% and that of B decreased from 59.63% ± 0.78% to 54.05% ± 0.87%. Therefore, straw was more suitable for maintaining a higher temperature, higher water removal ability and organic degradability during the storage sludge biodrying process, which was because the biodegradation potential of straw was stronger than sawdust and the aerobic incubation tests under conditions similar to sludge biodrying in the previous literature confirmed this [15].

3.2. Stability analysis of biodried products

The evolution of the AT_4 index under different bulking agents is presented in Fig. 3. To some extent, the AT_4 index can represent the biological stability of biodried products. The AT_4 index increased gradually at the beginning and decreased after the sixth day. It was observed that the initial AT_4 index for A and B was 54 and 43 mg O_2/g DM, respectively, and the maximum value was 60 and 48 mg O_2/g DM, respectively. At the end of biodrying process, the AT_4 index decreased to 15 and 36 mg O_2/g DM, respectively. During the biodrying process, the AT_4 index of B varied more slowly than A. The AT_4 index increased slightly at the beginning, which was likely due to the consumption of dissolved organic matter and the depolymerization of polyacrylamide (PAM) organic macromolecular flocs that release soluble substrates for aerobic strains [35]. The AT_4 index decreased slowly with the stabilization of the matrix [31], indicating that the biological stability of biodried products increased gradually with the biodrying process and also that A was more stable than B.

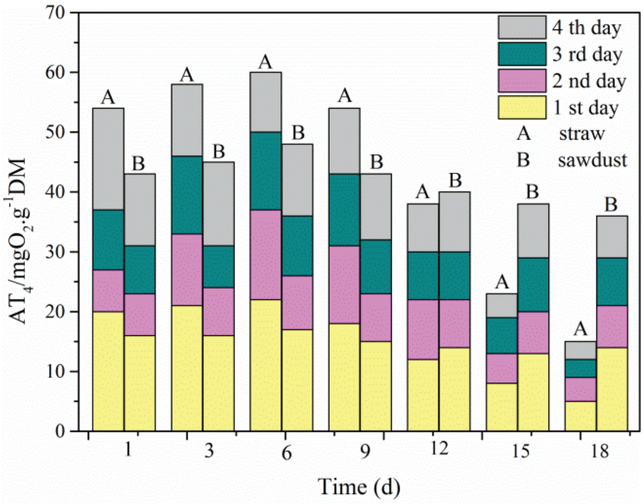

Fig. 3. Variation of biodegradability of biodried products.

3.3. Variation of major organic matter during the biodrying process

3.3.1. Change of protein and polysaccharide content during the biodrying process

Water in material was evaporated and removed by air ventilation [22,36,37] and bioheat generated by the aerobic degradation of organic matter (protein, polysaccharide, etc.) [15,21,25], which in turn can reveal the extent of the biodrying process. Thus, it is necessary to study the variation of organic matter in the process of biodrying. The variation of protein and polysaccharide content is shown in Fig. 4. For A (straw as bulking agent), the protein content decreased from 23.12% ± 0.29% to 14.23% ± 0.61% and for B (sawdust as the bulking agent), it decreased from 18.56% ± 0.76% to 12.91% ± 0.34%. The degradation rate was 38.45% and 30.44%, respectively, which implied that more protein was degraded when straw was used as the bulking agent. A large amount of heat was generated by degradation of protein, which was consistent with the higher temperature, lower moisture content, and more VS degradation when straw was used as the bulking agent. The polysaccharide content of A decreased from 15.11% ± 0.19% to 14.21% ± 0.33% while in B it decreased from 11.12% ± 0.13% to 10.34% ± 0.16%, which was not an obvious change. For both A and B, the protein content obviously decreased while the polysaccharide content remained relatively, indicating that protein and not polysaccharide was the main degradation substance during the biodrying process which may be related to the higher protease content in the storage sludge [35,38,39].

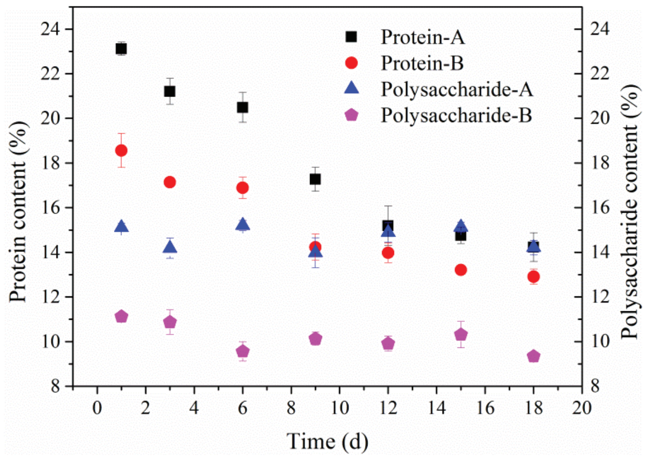

Fig. 4. The variation of protein and polysaccharide during biodrying process.

3.3.2. Change of humic substances during the biodrying process

The variation of HS and its composition of HA and FA is presented in Fig. 5. The HS content and HA content increased, while the FA content decreased for both trials A and B during the biodrying process. The HS content of trial A increased from 29.78 ± 1.87 to 57.91 ± 0.96 g/kg (an increase rate of 48.58%), while for trial B it increased from 25.61 ± 0.87 to 41.77 ± 1.34 g/kg (an increase rate of 38.69%). Thus, the increased degree of trial A was more obvious. The HA content of trial A increased from 11.11 ± 0.66 to 40.83 ± 1.39 g/kg and for B it increased from 7.32 ± 1.56 to 30.15 ± 1.35 g/kg. The FA content of trial A decreased from 25.11 ± 0.56 to 17.07 ± 1.55 g/kg and for B it decreased from 18.12 ± 1.53 to 11.98 ± 1.56 g/kg. It was found that the biodrying process was conducive to the formation of HS and HA but not to the formation of FA, possibly because the molecular weight of HA is larger than FA. Furthermore, similar to composting, the biodrying process was conducive to the formation of macromolecular humus [40,41]. The increase of molecular weight implied that the HS transformed from water-soluble state to solid state. Additionally, the HA/FA ratio increased gradually during the biodrying process for trial A and B from 0.44 to 2.39 (trial A) and 0.40 to 2.52 (trial B), respectively, which also indicated that the biodrying process was beneficial to the formation of HA. These results showed that just like composting, the biodrying process could cause the organic matter of storage sludge to gradually become stable and move in the direction of the humus, which would prove useful in increasing the value of the land use and reducing environmental risk.

3.3.3. Variation of organic elements during biodrying process

The variation of organic elements (C, N, H, and S) of the storage sludge during the biodrying process is outlined in Fig. 6. The organic elements of A and B had similar trends. The content of C, N, H, and S decreased during the biodrying process. For trial A, the content of C decreased from 23.65% ± 0.55% to 18.06% ± 0.55% (a reduction rate of 36.32%), while the reduction rate of B was 31.90%. Similarly, the loss of N and H in B was also lower than that of A, implying that the degradation degree of organic matter in A was higher than in B. The reduction rate of A was 9.69% while that of B was 18.53%, indicating that more S was lost in B. Additionally, the C/N and C/H ratio in A and B decreased during the biodrying process, possibly due to the strong degradation of organic carbon compounds, which indicated an increase in the maturity degree of the storage sludge by biodrying [21,42].

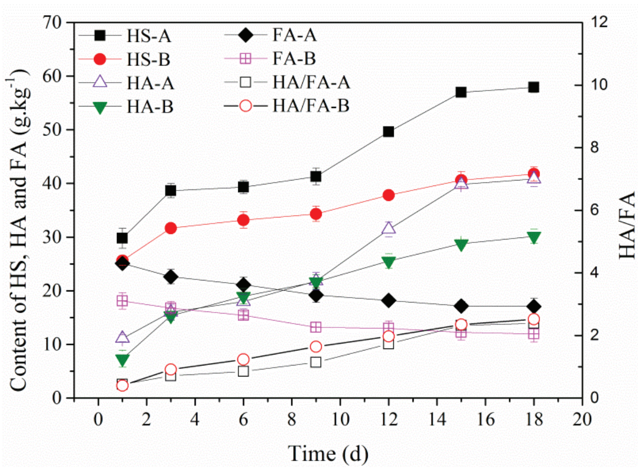

Fig. 5. Variation of humic matter and its composition during biodrying process.

3.4. Variation of heavy metal content and speciation during the biodrying process

3.4.1. Analysis of heavy metal content during the biodrying process

Heavy metal content is an important index for the agricultural application of storage sludge. The content change of different heavy metals (Cu, Zn, Cr, Ni, Pb, Cd, As, and Hg) during the biodrying process and the standards of different countries are shown in Table 2. The content of all heavy metals increased slightly. An increase of 6.29%–10.81% was

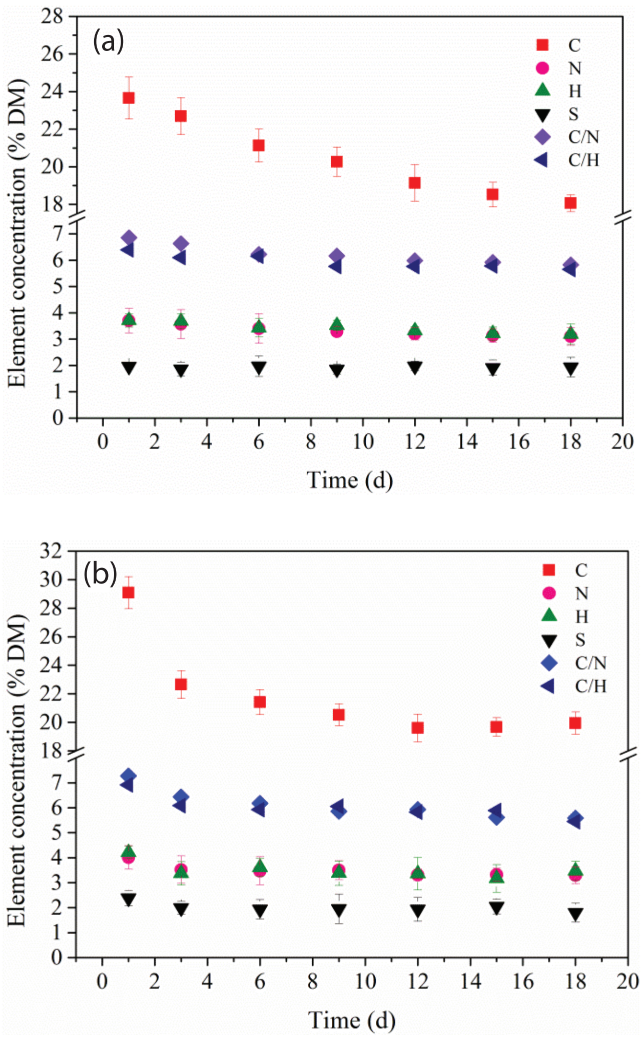

Fig. 6. Element variation during biodrying process: (a) trial A (straw as bulking agent) and (b) trial B (sawdust as bulking agent).

Table 2
Content of heavy metals during biodrying process of storage sludge (mg/kg)

Time (d)	Heavy metal species							
	Cu	Zn	Cr	Ni	Pb	Cd	As	Hg
1	430.81	1,006.11	150.72	62.01	52.32	4.32	9.81	4.18
3	443.76	1,045.22	156.34	65.33	52.24	4.33	9.57	4.22
6	438.21	998.35	153.29	66.24	55.54	4.56	9.78	4.27
9	450.35	1,000.32	157.23	64.39	54.30	4.51	9.98	4.31
12	443.24	980.34	153.21	65.45	55.69	4.48	10.01	4.29
15	442.14	1,060.34	158.23	66.34	59.78	4.73	10.76	4.53
18	457.89	1,080.23	164.34	67.25	56.32	4.77	10.87	4.62
Chinese standard	500	1,000	1,000	200	1,000	20	75	15
EU standard	1,750	4,000	1,000	400	1,200	40	–	25
France standard	1,000	3,000	1,000	200	800	15	–	10
Germany standard	800	2,500	900	200	900	10	–	8
Sweden standard	600	800	100	50	100	2	–	2.5

observed, indicating that the heavy metals were concentrated during the biodrying process. On one hand, the degradation of organic matter and the reduction of pile volume resulted in the concentration of heavy metals in storage sludge which was shown as "relative concentration effect" [43]. On the other hand, some dissolved heavy metals were lost due to leaching, which caused a decrease in heavy metals [43]. For the comprehensive effect of concentration and leaching, the content of heavy metals had little change, which showed that the impact of concentration was greater than the impact of leaching.

Cu and Zn were found in storage sludge to be predominant followed by Cr, Ni, Pb, As, Cd, and Hg. The content of Cu was about 400 mg/kg and Zn was found to be 1,000 mg/kg. The content of Cd, As, and Hg was very low (less than 10 mg/kg). It was discovered that all heavy metals, except Zn, met the requirements outlined in the Chinese standards for soil, and furthermore they did not exceed the maximum permissible content standards of other countries. More than half of the arable land in China is lacking in zinc [44], and it is necessary to use zinc fertilizer extensively in agricultural production. However, the standard limit of Zn content outlined in the Chinese agricultural standard is too strict, which limits the use of sludge in agriculture.

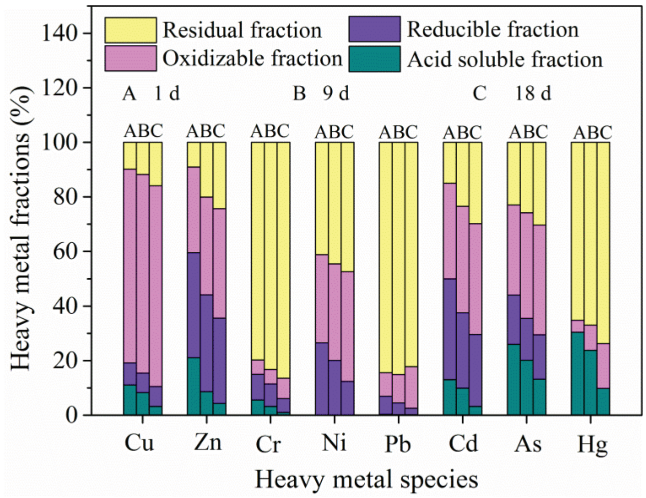

Fig. 7. The variation of heavy metal fractions during biodrying process.

3.4.2. Analysis of heavy metal fraction during the biodrying process

The variation of heavy metal fraction in storage sludge during biodrying process is shown in Fig. 7. For all heavy metals, the proportion of the oxidizable fraction and residual fraction were higher than the acid soluble fraction and reducible fraction. Cu, Zn, As, and Cd dominated the oxidizable fraction, while Cr, Pb, and Hg existed predominantly in the residual fraction. Furthermore, Ni was evenly distributed in both oxidizable and residual fractions during the biodrying process. The acid soluble fraction was not detected for Ni and Pb, indicating that the acid soluble fraction of Ni and Pb was not stable.

Most heavy metals were transformed from the acid soluble fraction to the oxidizable and residual fractions during the biodrying process, and the variation of the reducible fraction was not obvious. For Cu, the proportion of the oxidizable fraction increased from 70.99% to 73.64%, and the residual fraction increased from 9.86% to 20.88%, while the acid soluble fraction decreased from 11.12% to 3.23% and the reducible fraction from 8.03% to 7.25%. It was found that the change of the reducible fraction was insignificant while the variation of the residual fraction was the most obvious. The proportion of the oxidizable fraction and the residual fraction was 94.56% when the biodrying process was completed. Thus, Cu was mainly immobilized as complexes of the oxidizable fraction and the residual fraction because Cu had a high affinity with organic matter and was directly bound to two or more organic functional groups (carboxylic, carbonyl, and phenolic groups) [43,45,46]. Just like composting, the heavy metals changed more stable during the biodrying process. The acid soluble fraction is considered to be unstable, and the migration activity and bioavailability activity are larger than other factions. The reduction of the acid soluble fraction content revealed that the effective state depressed and the toxicity of heavy metals was weakened. Therefore, biodrying could effectively fix the heavy metal of storage sludge. When compared with sawdust, the straw was more easily degraded [15]. Thus, the degradation of organic matter and humus formation could restore, adsorb, and fix heavy metal ions, which was beneficial to the transformation of heavy metals to other forms and in turn reduced the bioavailability of heavy metal.

3.4.3. The interaction between heavy metals and humic substances

The content of macromolecular HS increased, as shown in Fig. 5 and the form of the heavy metals was transformed from effective state to steady state (Fig. 7) with a decreasing bioavailability. HS played an important role in the transformation of heavy metals, as water-soluble HS could bind with heavy metals through a complexation reaction while solid HS do so by an adsorption reaction [47]. The water solubility of heavy metal complexes determined their migration, which in turn affected the bioavailability of heavy metals. Furthermore, solid-phase macromolecular humus was able to adsorb, fix heavy metals, and reduce their mobility and effectiveness [48]. Therefore, the increase in the content of macromolecular HS was conducive to the transformation and stabilization of heavy metals due to the increased heavy metal binding sites [49].

4. Conclusion

Adding straw produced higher matrix temperature and water-removal capacity compared with sawdust, and the treatment in trial A achieved a better performance. During the biodrying process, more protein (38.45%) was degraded compared with polysaccharide (5.96%). The addition of different bulking agents could result in different variation of the AT_4 index, organic elements, and HS, which implied that biodrying caused an increase in the biostabilization of biodried products. All the heavy metals, except Zn, did not exceed the maximum permissible content used for soil in China (GB4284-1984). The heavy metal form transformed from acid soluble fraction to oxidizable and residual fractions, which could reduce the hazards associated with heavy metals. Additionally, further investigation demonstrated that HS played an important role in the transformation of heavy metals. However, dried products of biodrying to some extent still carry agricultural risks.

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