


A New Resource Recovery Process for Refining Sludge Generated by a Paper Manufacturing Company

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ABSTRACT

A new sludge processing method was developed by the Beta Company, a paper manufacturing company. The new method will shorten the time for transforming sludge into organic fertilizers, which will improve the process of handling the waste generated by a paper manufacturing company. The paper industry is facing the most rigorous regulations for processing wastewater and sludge to date in the history of many industrialized and developing countries. The Beta Company attempted to improve the conventional method implemented in-company for handling sludge generated during the paper manufacturing process, instead of shipping sludge out-of- company for processing by a third party. The current experimental results show positive improvement, indicating the Beta Company is moving one step closer toward environmental sustainability commitments.

KEYWORDS

Environmental Sustainability, Infrared Spectrum, Organic Fertilizer, Paper Industry, Paper Sludge, Resource Recovery

INTRODUCTION

The paper industry is one of the important fundamental raw material industries in China. The major production areas are in Shandong, Guangdong, Zhejiang, and Jiangsu provinces. In 2016, the production amount of paper pulp was 18 million and 500 thousand tons, 18 million and 400 thousand tons, 16 million and 900 thousand tons, and 12 million and 850 thousand tons for Shandong, Guangdong, Zhejiang, and Jiangsu provinces, respectively (United Credit Ratings Co. Ltd. [UCRCL], 2018). In China, there is a shortage of wood resources. The average forest area per person is only one quarter of the world's average area per person. At the same time, the demand for wood resources is high (Hu et al., 2001; UCRCL, 2018; G. Wu, 2007). Thus, the issue of environmental sustainability is more important than ever. The Beta Company, where the proposed new process for refining paper sludge will take place, is located in the Guangdong province. The paper pulp that will be transformed into organic fertilizers is from the plantation forest, which is owned by the Beta Company. It is less

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likely to contain heavy metal contamination such as that from the paper pulp generated from a recycled paper company. If the Beta Company successfully develops a new process for transforming sludge into organic fertilizers, it will enhance its capability of handling production waste.

Water pollution is the major type of production pollution from paper manufacturers, and the sludge produced during the process of making paper pulp needs special attention (Chinese Paper Company [CPC], 2020; Education for good, 2019). In recent years, China has created, and is enforcing, several environmental laws and regulations, such as “The Law on Environmental Protection of The People’s Republic of China (PRC),” “The Law of Cleaning Production Reinforcement of The PRC,” “The Law of Circular Economy Reinforcement of The PRC,” “The Law on the Prevention and Treatment of Water Pollution of The PRC,” “Opinions Regarding Accelerate Developing of Circular Economy by State Council (China),” and “The Index System of Clean Production Evaluation of Paper Industry (China)” (UCRCL, 2018). One of the major purposes of implementing these laws and regulations is to enhance the operational capability of the paper industry and make it fulfill the standards of the laws and regulations enacted by the government. Thus, the industry may need to eliminate outdated production processes and consolidate.

LITERATURE REVIEW

The paper industry needs to recycle the organic waste it produces in an economically viable and environmentally acceptable way. Sludge from paper mills has been successfully used as a soil conditioner for agricultural and forest crops (Cline & Chong, 1991; Henry, 1991), used as reclaimed land (Bellamy et al., 1990; Pervaiz & Sain, 2015), and also used as a medium production vessel for producing crops in cultivation greenhouses and nurseries (Chong & Cline, 1993). Another popular strategy is incineration, which is used to reduce capacity and generate electricity (Faubert et al., 2016). Other less common methods include anaerobic digestion (Meyer & Edwards, 2014), pyrolysis (Reckamp et al., 2014), bioethanol and hydrogen production (Moreau et al., 2015), and composites are used for various applications (Faubert et al., 2016). However, most of these methods fail because when dealing with a large amount of primary paper mill sludge (PPMS), it is often uneconomical and even harmful to the environment. Under these situations, biological treatment of sludge is more beneficial because, in addition to the volume reduction, a product with resale value can be produced, thereby increasing the efficiency of waste management.

Sludge in paper mills is usually stacked in piles by mixing in organic waste, such as animal manure or other agricultural waste, and letting it decompose for three to five weeks, with frequent heaping into a precocious compost (Campbell et al., 1991; Chong et al., 1991). After another four to six weeks of static stages of stacking and occasionally turning the pile into fully cured compost, the end product becomes an acceptable medium for producing container plants (Chong & Cline, 1991).

The use of compost produced by aerobic stacking with other excipients and pulp sludge is a feasible and effective method. For example, Jokela et al. (1997) developed compost made from primary and secondary sludge from wastepaper mills and the compost was forced to be ventilated, and the amount of ventilation was also adjusted. Meanwhile, the compost temperature was controlled at 55°C to 60°C. The high-temperature fermentation period can be maintained for 24 hours, the total solids content can be increased from 31.3% to 63.8%. The paper sludge has been proven to be feasible for aerobic composting treatment. Rantala et al. (1999) utilized compost, which was also made by mixing five different types of papermaking sludge with bark. The mixing ratios are from 3:4 to 1:2. During the composting period, the compost stack is completely turned over three to four times. The duration of composting time is four months. All sludge can be easily fermented and can be used for soil improvement in crop cultivation. Rantala et al. (2000) further mixed two different types of papermaking sludge with bark in 3:4 and 4:7 ratios, and then added chicken manure for aerobic composting fermentation compost. During this period, the compost stack was turned over for a total of seven times, and the composting time was six months. It was found that both kinds of

sludge could be fermented smoothly. The original sludge was toxic, but the toxicity was reduced during the fermentation process, and the fermentation-completed sludge was proven to be non-toxic. Fermented papermaking sludge has been proven suitable for soil improvement for crop cultivation.

Hazarika and Khwairakpam (2018) used a mixture of PPMS, cow dung, and sawdust as a fermentation material. It was found that at a ratio of 6:3:1 fermentation using the aerobic composting method of turning could produce the most effective fermentation effect with a high-temperature state of up to about 10 days and could improve the utilization rate of phosphorus by about 67%. PPMS can thus be transformed into potential value-added products that can be safely applied to soil.

In recent years, Simão et al. (2018) provided an overview of the characteristics of pulp and paper mill manufacturing processes, the waste generated, the main destinations, and alternatives to recycling. Studies have shown that lime sludge can be used in agriculture as a soil conditioner, but also in agriculture and environmental technologies, mainly for wastewater treatment. Grit is commonly used in construction, while pulp mill sludge is shown in applications in agriculture, construction, and energy processes. Several success cases of recycling waste from pulp and paper mills were also included. Rasa et al. (2020) used three different types of paper-based organic sludge and studied their effects on grain yield, soil carbon content, and fungal and bacterial composition. It was found that there was a direct interaction between the added granular organic matter and the microorganisms that stabilized the soil aggregates.

In soils with low organic matter content, the application of paper industry by-products can be a viable measure to mitigate soil erosion. Goel et al. (2021) intended to make bricks with recycled paper-mill-sludge compost (PPMC). It was found that fired bricks incorporated with PPMC could be safe for use in conventional applications as non-load-bearing and in-filled walls. Because the European Green Deal focuses on the circular economy, this study is timely and fulfills the goals of the United Nations Sustainable Development Goals (SDGs). Zawadzińska et al. (2021) conducted an experiment using plant biomass in the form of waste and by-products from various industries and transformed the waste and by-products into a valuable material for the production of compost and growing media for urban horticulture. In this study, pulp and paper mill sludge, fruit and vegetable waste, mushroom-used matrices, and rye straw were used to produce compost, further used as a medium component for tomato container cultivation. This study shows that compost made from natural materials from a variety of sources is a valuable potted medium supplement that has a positive effect on tomato yield and nutritional value.

Turner et al.'s (2022) review article pointed out that historical and recent literature of land disseminated on pulp and paper mill sludge (PAPMS), combined with knowledge of European and British regulations and through the pretreatment of the sludge by composting, can easily mitigate the risks associated with potential nitrogen fixation in the soil after application. The benefits to crops have been well demonstrated, and to date no negative ecological impacts have been observed at typical field application rates. Therefore, it is necessary for the compost made from pulp and paper mill sludge to continue to be applied on land as an environmentally responsible and sustainable option. However, there are current gaps in the literature regarding the long-term effects of PAPMS applications in agriculture and the possible presence of new contaminants in certain PAPMS materials.

PAPER MANUFACTURING PROCESS AND PRODUCTION WASTE

Paper Manufacturing Process

The paper manufacturing process consists of two types of processes, pulping and papermaking. Pulping uses mechanical or chemical methods to separate vegetative fibers, artificial fibers, lignin, and dissolvable ingredients from other ingredients (China Technology Service Publisher [CTSP], 2013). The fiber then goes through the purification, bleaching, and drying process to make pulp or cardboard. At present, the most widely used pulping methods are mechanical, chemical, and semi-chemical methods. The chemical pulping process is widely used in paper mills. The papermaking

process mixes crude paper pulp and thickens it after bleaching the paper pulp. After the plasma refining, seminal plasma refining, papermaking, and drying processes, the paper pulp will turn into a finished product. During these processes, the amount of white water generated is quite large; white water is the main component of wastewater.

Waste of Papermaking Process

The pollutants of wastewater discharged from the pulp/paper industry can be divided into three categories: suspended solids, soluble organic matter, and soluble inorganic matter (CTSP, 2013):

1. The solids contained in suspended solids are wood chips and bark produced by the preparation chamber, the fibers produced during pulping and papermaking, and the fillings in the papermaking process, such as calcium carbonate, titanium dioxide, and clay.
2. Soluble organic matter refers to wood in addition to fibers and lignin. There are about 30% of non-fibers, such as sugar, which if discharged in the river will cause rapid growth of microorganisms in the water. This results in a lack of oxygen in the river, which affects the growth of aquatic organisms. If the lack of oxygen is severe, anaerobic organisms can reproduce, causing a foul odor.
3. There are two main sources of soluble inorganic matter. One is cooking liquids used in the cooking process of the chemical and semi-chemical pulping methods, and the other is a variety of chemical agents added in the papermaking process.

SLUDGE TREATMENT METHODS AND AN EXPERIMENT

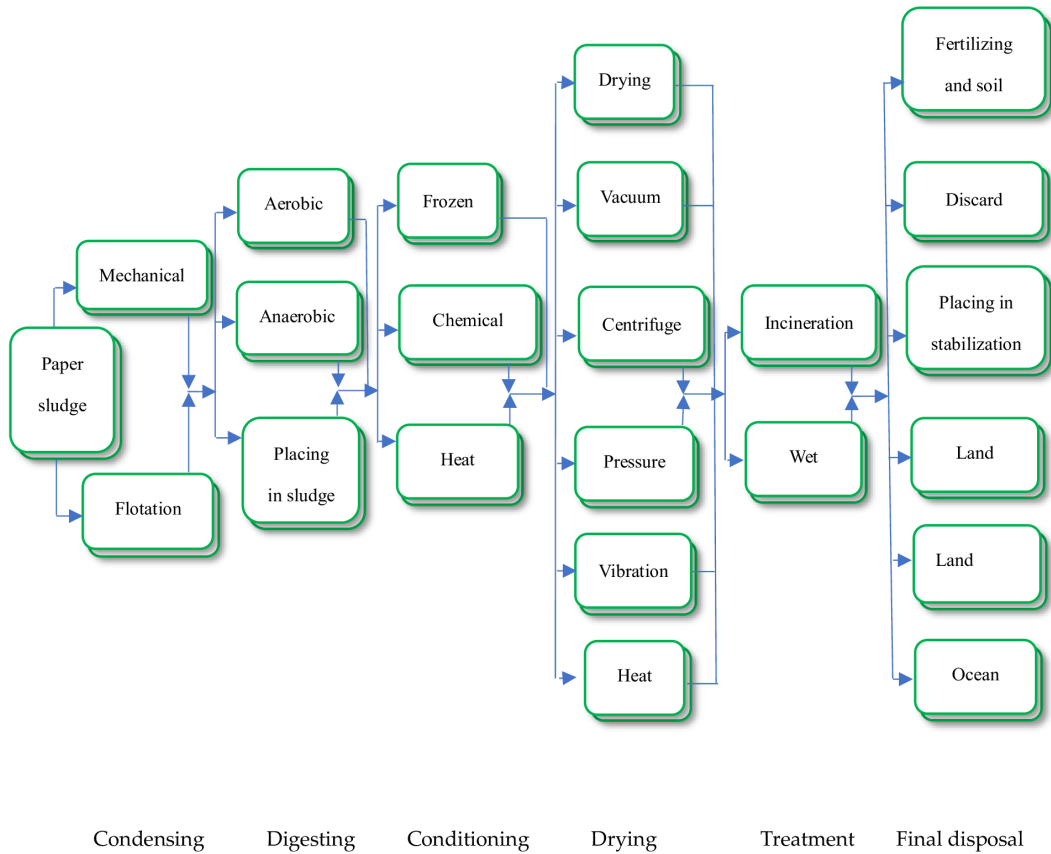
Traditional Sludge Treatment Methods

There are several traditional final treatment alternatives for paper sludge (see Figure 1), such as ocean disposal, land filling, land scattering, incineration, making fertilizer and soil conditioning (CTSP, 2013). Sludge and ash disposal at the final stage of the process faces a variety of challenges, as does ocean disposal, which has been prohibited by international marine law. Land filling or scattering on land have the advantages of potential restoration of landforms, restoration of vegetation, and restoration of land. The disadvantages of land filling or scattering are that it requires large amounts of land and there is the potential of causing secondary pollution of the land, groundwater, and atmosphere. If treating paper sludge through incineration, there is the high cost of incineration and the potential for causing secondary pollution. Therefore, sludge reduction, stabilization, and harmless treatment (such as converting it into a resource) have become the main development direction (CPC, 2020; CTSP, 2013).

Several characteristics and disadvantages associated with sludge processing stages are presented as follows (CTSP, 2013):

1. **Sludge Condensing:** (1) When the gravity thickeners method is used in this stage, raw sludge in a raw primary precipitation tank or abandoned activated sludge is continuously brought into a thickening tank. Using a truss rod or vertical fences while stirring the sludge slowly, clear water passes through an upper-opened ditch and goes back to the primary precipitation tank. The condensed sludge at the bottom of a precipitation tank is sucked into a digestion tank or a dehydration facility. (2) When the flotation thickeners method is used in this stage, the flotation thickeners tank is always used for handling abandon activated sludge. If not using chemical ingredients, under normal circumstances, 4% of solid type sludge could be obtained, while 85% of solid substance could be obtained from the input.
2. **Sludge Digestion:** The major ingredients of sludge in a paper-manufacturing factory are inorganic substances, lime, coagulant, and organic substances. The fibers within the organic substances cannot be digested and do not decompose easily.

Figure 1. The flow chart of tradition typical sludge treatment



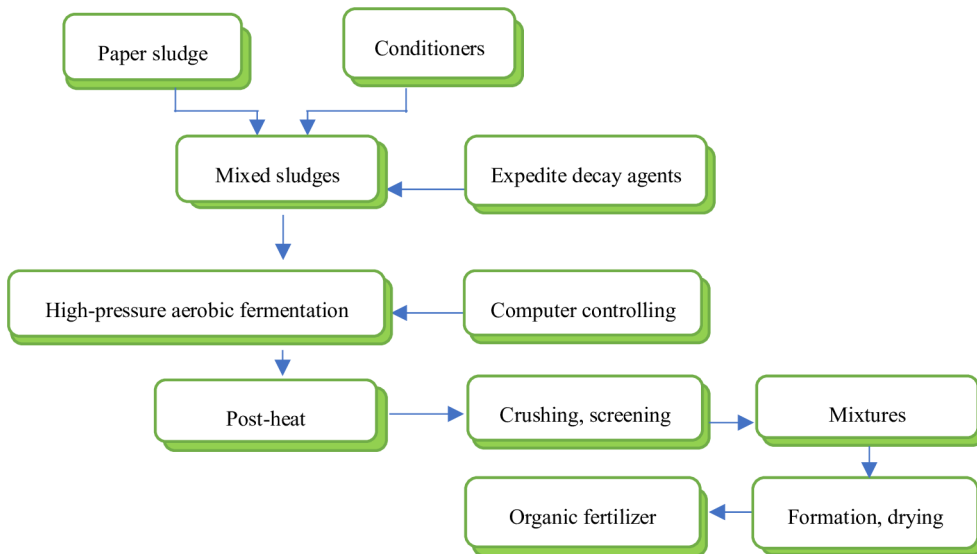
3. **Sludge Conditioning:** The methods for sludge conditioning are the freezing method, chemical conditioning method, and heat treatment method. The chemical conditioning method is the most often used and the most feasible solution.
4. **Sludge Final Disposal:** (1) *Sludge incineration*: sludge incineration includes two stages, the front stage drying, and post-combustion. (2) *Sludge pond*: this method is no longer adopted by most paper manufacturers because of the generation of anaerobic foul odors and the increase of BOD (biochemical oxygen demand) of the effluent. (3) *Land filling*: the treatment of dehydrated sludge with a substance high in lime, which is suitable for this method.

New Method for Sludge Conversion to Organic Fertilizer

S. Zhao et al. (2011) proposed a method (see Figure 2) for making organic fertilizers. This method mixed paper sludge and conditioning agents; added the accelerant decay agents; caused high-pressure aerobic fermentation; added heat; crushed and screened the materials; and added nitrogen, phosphorus, potassium, and other elements to produce the organic fertilizers.

The procedure proposed in this paper is different from that of S. Zhao et al. (2011), which is able to produce organic fertilizer, and is more than 20% faster than the various other procedures that currently exist (Oladejo et al., 2018). Because this procedure was successful in the laboratory, it will be implemented in factories on a large scale. The waste sludge does not have to be transported outside the plant, so as not to cause any possible secondary pollution. The process is shown in Figure 3.

Figure 2. A method of using paper sludge to produce organic fertilizers



First, the pulp sludge is mixed with chicken manure, phosphorus powder, and fermentation bacteria, and then stirred. Then the sludge is forcibly ventilated and placed in an aerobic fermentation tank, followed by the procedure for ripening, and then becoming composted.

The main functions of aerobic bacteria used in this experiment are to eliminate foul odor and shorten the fermentation cycle time (He et al., 2019; Jian, 2010; Liang et al., 2021; Naicker et al., 2020; Y. Xu et al., 2020). The factory produces pulp sludge in the daily amount of 10 tons to 15 tons. If entrusted to a third party to deal with, the cost per ton is more than US\$30 (US dollars). The current processing costs continue to rise. Another main consideration is the uncertainty of the third-party treatment method, which is not easy to confirm. At present, the experimental method has achieved initial success, so that the factory does not have to ship sludge out of the factory. It can make its own organic fertilizers. In addition to being used in their own afforestation forest, surplus fertilizers can be sold. The factory has successfully achieved its processing waste reduction target and enhanced the added value of production waste.

The estimated sludge treatment cost and revenue of organic fertilizers are presented in Table 1. The selling price of organic fertilizers is US\$0.50/kg and the treatment cost by a third party is US\$30 per ton. The estimated sludge daily processing cost is US\$450, resulting in an annual processing cost of US\$118,800. The daily revenue from selling organic fertilizers is US\$7,500, while the annual revenue

Table 1. The estimation of the cost of paper sludge treatment and revenue of organic fertilizers

	Processing Cost of Paper Sludge	Revenue of Selling Organic Fertilizers
Daily Production of Paper Sludge	15 tons * \$30/ton = \$450/day	15 tons * \$0.5/kg ^b = \$7,500/day
Monthly Production of Paper Sludge	\$450 * 22 days = \$9,900/month	\$7,500 * 22 days = \$165,000/month
Yearly Production of Paper Sludge	\$9,900 * 12 months = \$118,800/year	\$165,000 * 12 months = \$1,980,000/year

^aNote: Currency unit used is US\$(US Dollars);^bcurrent market price of organic fertilizers for 20kg package

is US\$1,980,000. After deducting the sludge treatment cost and the increase of revenue from selling organic fertilizers, the potential annual revenue could reach US\$2,098,800. The sludge processing method proposed by the Beta Company is not only to acquire a positive return in the economy, but to also demonstrate its commitment to environmental sustainability.

An Experiment

Introduction

Primary paper mill sludge (PPMS) is the main waste released by the paper pulp industry, causing soil and water pollution through non-degradable organic and inorganic components. However, these pollutants can be converted into high-value soil improvement materials with a small investment of money and time. Sludge treatment and disposal of paper pulp are common and difficult problems faced by papermaking enterprises. At present, the disposal alternatives of paper pulp sludge are as landfill and through incineration, and the recycling alternatives are to use sludge to make bricks, ceramic grains, filler utilization, cement (Chen, 2019), sheet materials or, fertilizer or soil conditioners (D. Wang et al., 2003). Many studies have shown that the organic content of pulp and paper sludge is high, mainly including high-molecular organic matter such as cellulose, hemicellulose and lignin (M. Wang, 2011), and contains the elements needed for crop growth such as nitrogen, phosphorus and potassium (Huang et al., 2011). During the composting process, there is an opportunity for the organic matter to be broken down by microorganisms and further synthesized into stronger, harmless, and usable organic matter.

Experiments showed that the composting treatment and agriculturalization of pulp and paper sludge are alternative methods of resource recovery, beyond landfilling and incineration (Bellamy et al., 1995). Composted products can be used in agriculture or forestry productions while the wood can be used in pulp and papermaking, which would achieve the intention of integrating production and recycling. MSG (monosodium glutamate) waste liquid is rich in proteins, amino acids, bacteria, and other nutrients (Jia et al., 2007; Körner et al., 2001), and is added to paper sludge because of its nitrogen-rich properties for promoting joint fermentation.

Using MSG waste liquid as a nitrogen source can speed up the process of compost fermentation, and the effect of MSG waste liquid as a nitrogen source is higher than urea (J. Zhao et al., 2008). The use of aluminum-rich paper sludge compost can significantly slow down soil acidification, improve soil quality, and will not cause the accumulation of activated aluminum in the soil (Lin et al., 2010; J. Zhao et al., 2008). Paper sludge is relatively free of chemical pollutants and is mainly used as an improved organic agent rather than fertilizer, because its mineral nutrient content is very low. Greenhouses, seedling container substrates, and field tests have shown that the growth and desired growth of various horticultural and agricultural crops have improved. It has proven the beneficial reaction of using pulp sludge as a potted organic conditioner and in field soils (Bellamy et al., 1995). The paper sludge is dried with fertilizer and ground into particles and applied to soil. Its performance is like that of clay, which has a strong moisture retention capacity and can be used as mulching soil (Schut, 2004). Paper mill sludge can become a soil conditioner by adding different proportions of cow manure and wood chips. The best ratio is 6:3:1 (sludge to cow manure to wood chips), and it was found that it could be transformed into a potential value-added product (Hazarika & Khwairakpam, 2018).

Using a high temperature and aerobic composting process to make pulp sludge harmless is a low-cost method, while adding other harmless food sludge and converting paper sludge into organic fertilizer is the best way for resource recovery. In the sludge composting process, due to the varieties and complexities of reactions, the resulting intermediate products are also mostly complex substances. Compost maturity is generally analyzed using physical, chemical and biological parameters. According to current research, physical indicators such as temperature, color, odor and E_4/E_6 give a more direct representation of the degree of maturity during composting. Chemical indicators such as pH and organic carbon can also be observed as indicators, while seed germination rate and its index are also commonly used as biological indicators in evaluating the maturity of the compost.

The Fourier transform infrared spectroscopy analysis technique is also frequently used to evaluate the structure-changing characteristics of organic matter while composting, and its results serve as the theoretical basis for composting maturity (Yu et al., 2016). This experiment uses the aerobic treatment by using the paper mill primary pulp sludge, adding the food sludge, and composting the mixture. An evaluation of the possibility of recycling pulp sludge for agricultural land use in a safer way was also conducted.

Experiment Materials

The raw materials for compost in this experiment are pulp sludge from a paper factory in Guangdong (in-plant sewage treatment terminal sludge dehydrator), white mud from a paper factory in Guangdong (a product of causticizing reaction from an alkali recovery workshop in the pulping process), bark, sludge, and waste liquids from a food plant in Guangdong. Molasses and pyroligneous acid were purchased as accessory materials (see Tables 2 and 3).

Experiment Equipment

The equipment used were composting bins, air pumps (aerations), precision electronic scales, pH meters, probe-type thermometers, multi-use speed oscillators, ultra-violet spectrophotometers, and IRTracer-100 Fourier transform infrared spectrometers.

Experiment Parameters and Processes

Treatment one (T1): 1:1 (3 kg:3 kg) sludge-to-bark wet weight ratio. Add 5% food plant sludge and 1.5 L(liters) nutrient solution (10 mL [milliliter] pyroligneous acid, 10 mL food plant waste liquid, and 50 mL molasses per liter of nutrient).

Treatment two (T2): 2:1 (4 kg:2 kg) sludge-to-bark wet weight ratio. Add 5% food plant sludge and 1.5L nutrient solution (10 mL pyroligneous acid, 10 mL food plant waste liquid, and 50 mL molasses per liter of nutrient).

Treatment three (T3): To 7 kg of sludge, add 10% food plant sludge, and add 2L nutrient solution (10 mL pyroligneous acid, 10 mL food factory waste, and 100 mL molasses per liter nutrient).

Table 2. Physico-chemical characteristics of paper mill sludge and material for compost

Parameters	Moisture %	pH	Organic Matter %	Carbon%	Nitrogen%	C/N Ratio	Fiber Content %	EC mS/cm	Iron %	Calcium %
Paper mill sludge	79.03	7.73	51.12	29.72	0.9	33.0	19.2	2.4	0.32	2.33
Bark	61.61	6.06	—	52.4	0.2	262	—		—	—
Food factory sludge	81.78	5.41	76.65	23.35	5.6	4.16	12.17		0.26	0.96
Food factory liquid waste	78.96	6.4	—	—			—		—	—

Table 3. Physico-chemical characteristics of white mud

Component Names	Moisture (%)	pH	Residual Alkali (%)	Calcium Carbonate (%)	Calcium Oxide (%)	Ferric Oxide (%)
White mud	22.3	12.26	0.28	75.1	0.18	0.1

Treatment four (T4): 8:1 (8 kg:1 kg) sludge-to-white-mud wet weight ratio. Add 10% of the food plant sludge and 2L nutrient solution (10 mL pyroligneous acid, 10 mL food factory waste, and 100 mL molasses per liter of nutrient).

Treatment five (T5): Pure sludge 8 kg. Add molasses 100 mL (100 mL with 1500 mL).

Detailed experiment parameters of compost samples are shown in Table 4.

The compost equipment is a purchased composting tank with a volume of about 55 L, and the air pump inflates from the ventilation hole of the compost box with approximately 0.5 L/min flow to the reactor body, breathing about eight hours per day. The compost in tank is flipped two times a day.

Test Methods

The probe thermometer is inserted into the stack of compost at a depth of about 20 cm at 8:30 am and 4:30 pm per day, respectively, by measuring three points, taking down the readings, and calculating the average temperature as the fermentation temperature of the compost. In the second decay phase, the same method was used and was recorded every six days. A 5-point sampling method was used for 3, 6, 9, 12, 19, 26, 33, and 40 days after the composting process was activated. After mixing the compost evenly, a test was conducted to measure the pH, moisture content, organic carbon, and E_4/E_6 . The pH value was measured by a Rex pH meter, after oscillating horizontally at 200 r/min, at room temperature, using the fresh compost samples with deionized water in a ratio of 1:10(W(g): V(mL)). The moisture content was measured using a fresh compost sample put in a 105 °C oven for baking. The organic carbon measurement used a wet oxidation method, where the compost was dried at 80 °C and ground into powder. A 0.1 g sample was placed in a 500 mL cone bottle, and 20 mL 1 N potassium dichromate ($K_2Cr_2O_7$) and 40 mL concentrated sulfuric acid (H_2SO_4) were added. The mixture was heated to 45 °C and kept at the same temperature for 30 minutes. After the mixture cooled, 200 mL of pure water was added to the conical bottle. Several drops of phenanthroline ($C_{12}H_8N_2$), and then 0.5N ferrous sulfate were dripped into the mixture until it became fresh green. Then the content of organic carbon (%) = $(1\text{ N} \times 20\text{ mL} - 0.5\text{ N} \times \text{titration mL}) \times 3 \times f$ (f : correction factor, about 1.3). The measurement of E_4/E_6 was taken using a compost sample of about 2-4 mg and dissolving it in 10 mL of 0.05N of sodium carbonate (Na_2CO_3) and adjusting to the optimal pH value. Its absorbance at wavelengths 465 nm and 665 nm were measured. The seed germination rate was measured by taking 5 g compost, adding 100 mL of 60°C warm water and placing in a 200 mL beaker, filtering after three hours in a water bath at 60°C. Two sheets of paper were put in a petri dish, with a 10 mL liquid filter. Twenty-five cabbage seeds were placed in a petri dish, which was kept in temperature-controlled room. After one or two days, seed germination was observed. The compost sample and comparison sludge were pressed by KBr (potassium bromide) and analyzed by the IRTracer-100

Table 4. Experiment parameters of compost samples

Treatment	Sludge	Wet Bark	White Mud	Food Factory Sludge	Nutrients			
					Pyroligneous Acid	Waste Liquid From Food Plant	Molasses	Water
T1	3 kg	3 kg	—	5%	15 mL	15 mL	75 mL	1395 mL
T2	4 kg	2 kg	—	5%	15 mL	15 mL	75 mL	1395 mL
T3	7 kg	—	—	10%	20 mL	20 mL	200 mL	1760 mL
T4	8 kg	—	1 kg	10%	20 mL	20 mL	200 mL	1760 mL
T5	8 kg	—	—	—	—	—	100 mL	1400 mL

Fourier Transform Infrared Spectrometer. The transmission mode was scanned 32 times. The pilot method flow is shown in Figure 3.

Data Processing

The experimental data was analyzed and processed using Origin8.0 and Microsoft Excel 2007 software, and OMNIC8.0 software for infrared spectral semi-quantitative analysis.

Results and Discussions

Temperature

Temperature can reflect the changes of microbial activity and indicate the degree of maturity of the compost. The results of this experiment on the compost temperature are shown in Figure 4. During the composting process, the organic matter in the sludge decomposes under the action of aerobic microorganisms, while releasing a large amount of heat, causing the composting temperature of

Figure 3. The proposed new method for making organic fertilizers from paper sludge

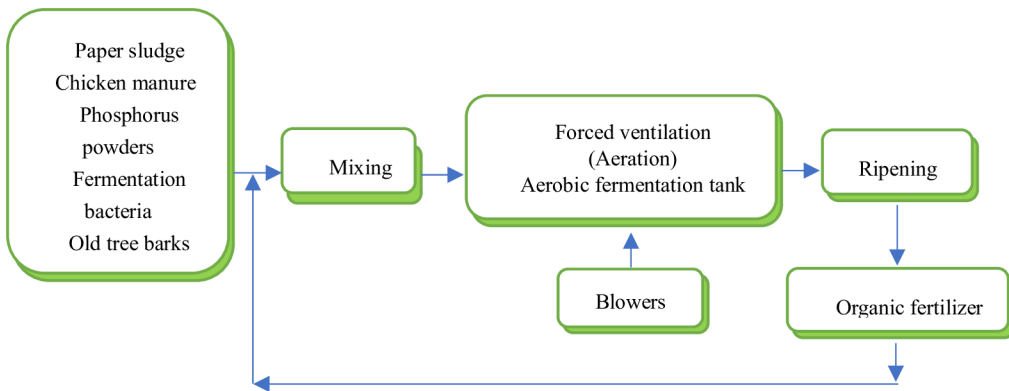
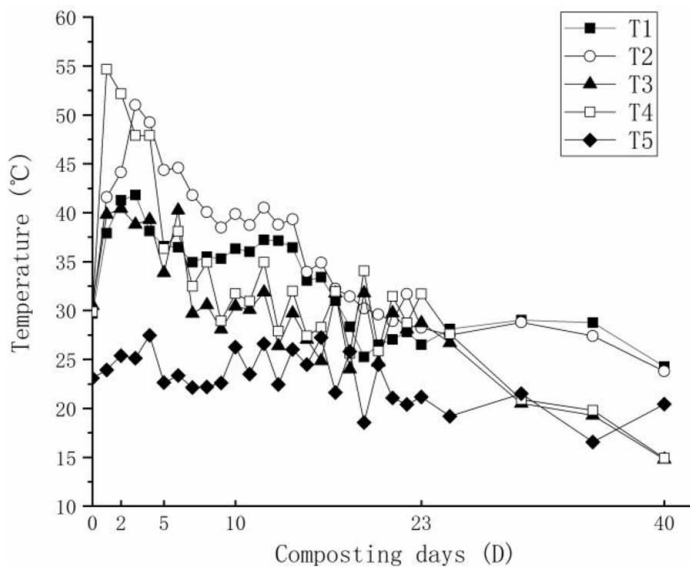


Figure 4. The changes of temperature in sludge composting process



each treatment to rise and reach peak, with the peak temperature of the five groups of treatments (T1, T2, T3, T4, T5) being 41.8°C, 51.0°C, 40.4°C, 54.7°C, and 27.5°C, respectively. The addition of food factory sludge and waste liquid, pyroligneous acid, and other auxiliary materials caused a rise in the initial temperature of the compost, perhaps because the food factory sludge and waste liquid provided a source of bacteria; molasses provided a carbon source; and the appropriate concentration of pyroligneous acid promoted the activity of microorganisms, promoted the growth of microorganisms, and reduced the production of organic matter heat. Fundamentally, there are two types of composting systems: temperature limits and non-temperature limits. The self-limiting system achieves a suppressed temperature ($> 60^{\circ}\text{C}$), weakening the microbiome and inhibiting decomposition, heat output, and moisture removal. In contrast, a non-self-limiting temperature system ($< 60^{\circ}\text{C}$) provides a powerful bacterial community that promotes decomposition, heat output, and dehydration (MacGregor et al., 1981). The peak temperature of each processing group of this test lasts only one to two days, resulting from three possible reasons: (1) the stack size may be too small and not conducive to insulation; (2) the ventilation time may be long, resulting in heat dissipation that is faster than heat production, or; (3) the stacks may be turned too frequently.

T2 more easily achieved a 50°C composting temperature than T1, which may be due to the additional amount of sludge. More sludge means more organic substances, resulting in more microbial reproduction and more heat energy being released. The composting temperature of T4 is higher than T3, mainly due to the addition of white mud to increase pH, which regulates the acidity and alkalinity of the stack to increase microbial activity and release more heat. This phenomenon is similar to the process of composting food waste; an extended initial acidic phase may occur, resulting in a similar low degradation rate. In a successful composting process, in the initial stage is a process of high-speed composting at a pH higher than neutral. A combination of temperatures above 40°C and pH below 6 severely inhibits the composting process (Sundberg, 2005). The result of adding white mud helps composting fermentation.

Moisture

Moisture is the basis for microbial life activity in the process of composting. Studies have shown that the moisture rate of composting experiments should be maintained at 50% to 60%, for the results of composting to be best (I. Zhao et al., 2020). When the moisture rate exceeds 70%, it is easy to cause an anaerobic state, inhibiting the growth and reproduction of aerobic microorganisms and reducing the decomposition rate of organic matter. Figure 5 shows that the moisture rate of each treatment is more than 20% during the composting process, which will fulfill the minimum requirement of humidity for microbial activity. As the composting experiment progressed, the moisture content changed, which was mainly due to the fact that microorganisms also consumed water when converting organic matter to produce other substances. Moisture was constantly being evaporated in the states of ventilation and stacking. By the end of composting, the moisture rate of T1 was 54.7%; T2, 46.02%; T3, 26.38%; and T4, 30.3%, which when compared to the start of composting, was down by 11.31%, 15.99%, 21.24%, and 16.22%, respectively. The moisture rates of T1 and T2 are higher than those of T3 and T4, possibly because the bark absorbed moisture and accumulated in the stacks, however, the moisture rates of the five groups of treatment showed a clear downward trend.

pH

The changes of pH indicated the activity and overall condition of microorganisms during composting. Studies have shown that composting pH can effectively promote good microbial growth at 6.7 to 9.0. The pH change trend in the stack first goes up then goes down. During the early stage of the composting process, microbial metabolism is active; nitrogen-containing organic matter decomposes under ammonia, producing a large amount of ammonium nitrogen, resulting in pH rising. And then, due to air circulation and ammonia volatilization, ammonia is weakened, and in the degradation process, organic acid and humic acid are generated, resulting in pH decreasing. As shown in Figure 6, the pH remained at 6.64

Figure 5. The variations of moisture rate

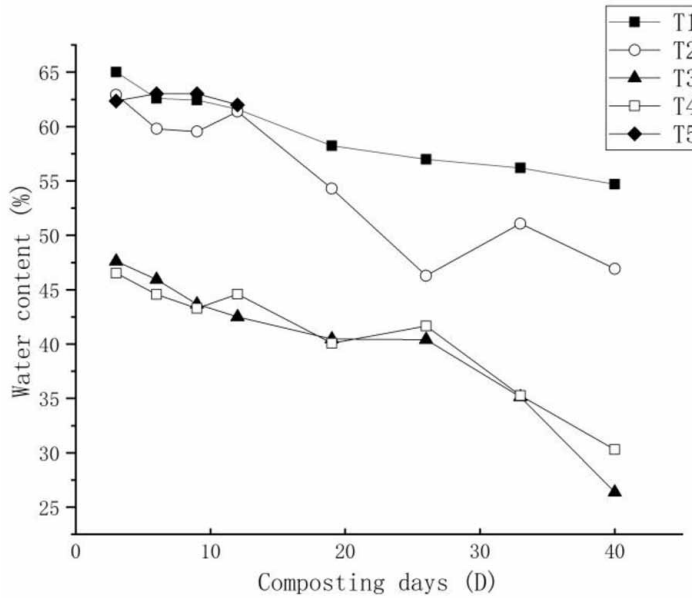
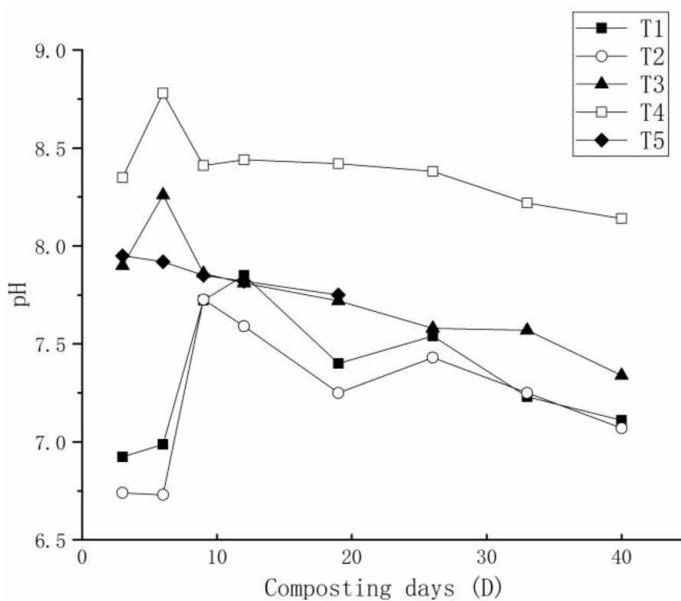


Figure 6. The changes of pH



to 8.14 in this composting experiment. The pH values were in the trend of rising first, possibly with the release of NH_3 in the composting process. The five groups of treatments are basically in the pH range appropriate for composting. The initial pH of T4 is higher than in other treatments, possibly due to the addition of white mud. The main component is calcium carbonate, which is strongly alkaline. The acidic substances produced in the process of composting reduced the pH of the stack. Zhang et al.

(2018) proposed that adding lime powder can adjust the pH of the stack to be neutral or weakly alkaline to prevent the stack from being acidic, which is not conducive to fermentation. Adding an appropriate amount of white mud makes T4 ultimately weakly alkaline.

Variations of E_4/E_6 Ratio

E_4/E_6 ratio can be used for reflecting the polymerized aromatic degree and molecular weight of the humus during a composting process. The smaller the E_4/E_6 ratio, the greater the molecular weight of the compost humus, and the higher the level of formation of humus and polymerization. As the experiment progressed, the E_4/E_6 value decreased, indicating a significant degree of polymerization during composting (C. Li et al., 2015). As can be seen from the change in the ratio of E_4/E_6 in Figure 7, the ratio of E_4/E_6 increases first and then decreases in the recurring trend. The results show that there may be some large molecular substances in the initial composting period, which may be degraded by microorganisms into small molecular substances, resulting in an increase in their proportion, and thus generating new and more complex substances, and then reducing the proportion of E_4/E_6 . T1 is more similar to the T2 trend, mainly because the bark was already undergoing a decay phase (the stage of becoming old bark) before it was added to the compost test, where there was humic acid in the bark. E_4/E_6 ratio decreased with an increase in the molecular weight of humic acid or the increase in the degree of contraction. The ratio of E_4/E_6 changed significantly, and the biochemical process of organic and humic acid was strong, causing the compost to decay.

Variations of Organic Carbon

In the composting process, microorganisms decompose organic substances into water, carbon dioxide, and stable humic acid, providing the necessary substances for their own growth and reproduction (Liu et al., 2012). As shown in Figure 8, the quality score of organic carbon of each treatment is in a downward trend, while the score of T2 drops most significantly. The decrease of organic carbon in the early stage of each compost sample was greater, because in the initial stage of composting, each compost sample reached the peak of composting temperature. The microorganisms mainly use the

Figure 7. The variations of E_4/E_6 ratio

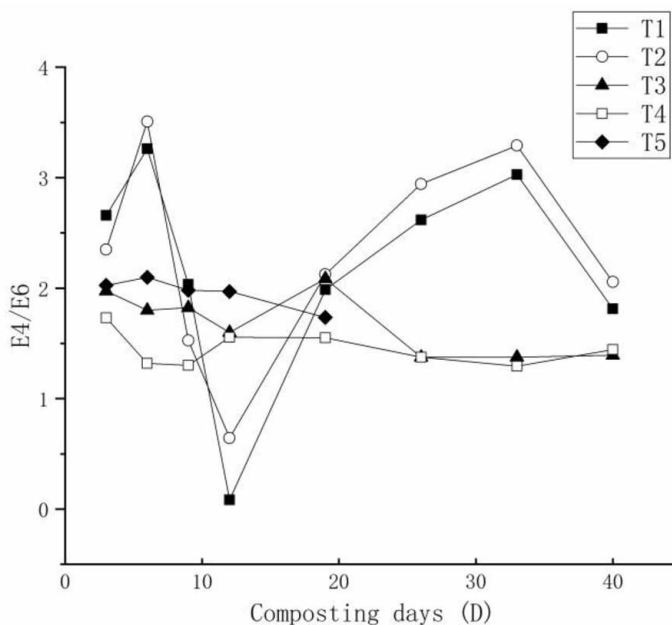
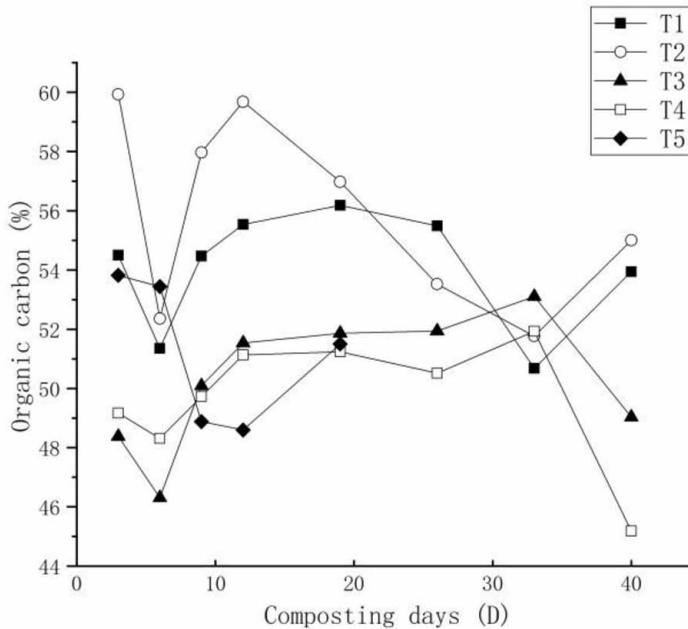


Figure 8. The variations of organic carbon



easily degradable organic substances in nutrients, such as sugar and starch to progress metabolism with a high decomposition speed, while organic carbon declines quickly. In an organic carbon recovery process, microorganisms use nutrients for reproduction, and thus cause a short-term rise of organic carbon. As the composting experiment progresses, the organic carbon content decreases. The microorganisms can only use hemicellulose, cellulose, and lignin as carbon sources. Microbial activity weakens and some organisms even die.

FTIR (Fourier Transform Infrared) Spectroscopy Analysis of Compost Treatments

The FTIR spectroscopy analysis method can identify the characteristics of the functional group of the compound. According to the results of previous studies, the various bands in the infrared spectrum are represented as follows: 3500 to 3200 cm^{-1} indicates O-H stretching vibration of carbohydrate and water molecules, the absorbance of N-H stretching vibration of protein and acylamide compound; 2935 to 2900 cm^{-1} indicates C-H stretching vibration of aliphatic; 1650 to 1640 cm^{-1} indicates C = O stretching vibration of amide acid, asymmetric COO stretching vibration of organic acid; 1430 to 1419 cm^{-1} indicates COO stretching vibration of organic acid, or double bond or carbonyl group connection of -CH transforming vibration in lignin and aliphatic compound and inorganic NH_4^+ or NO_3^- ; 1170 to 950 cm^{-1} indicates C-O stretching vibration of polysaccharide or polysaccharide analogues, possible -C-O peak of cellulose or hemicellulose; 890 to 870 cm^{-1} indicates CO_3^{2-} out-of-plane bending, or C-O out-of-plane bending vibration of carbonate; and 470 to 460 cm^{-1} indicates Si-O stretching vibration of silicate minerals or silicon dioxides (Kang et al., 2010; G. Li et al., 2009; MacGregor et al., 1981; G. Wang et al., 2019; Z. Wu et al., 2004; Y. Xu et al., 2004, 2014).

T0 is a control group of non-composting sludge. Combining Figure 9 with Table 5, the research found that 3370, 2923, 1644, 1424, 1021, and 873 cm^{-1} have obvious and similar absorbance peaks in each situation. The strength of the characteristic peak can reflect the relative content of the functional group. The greater the strength of the absorbance peak, the higher the content of the functional group (MacGregor et al., 1981). Based on the semi-quantitative analysis of Table 5,

Figure 9. FTIR spectra of pre-composting and post-composting of treatments

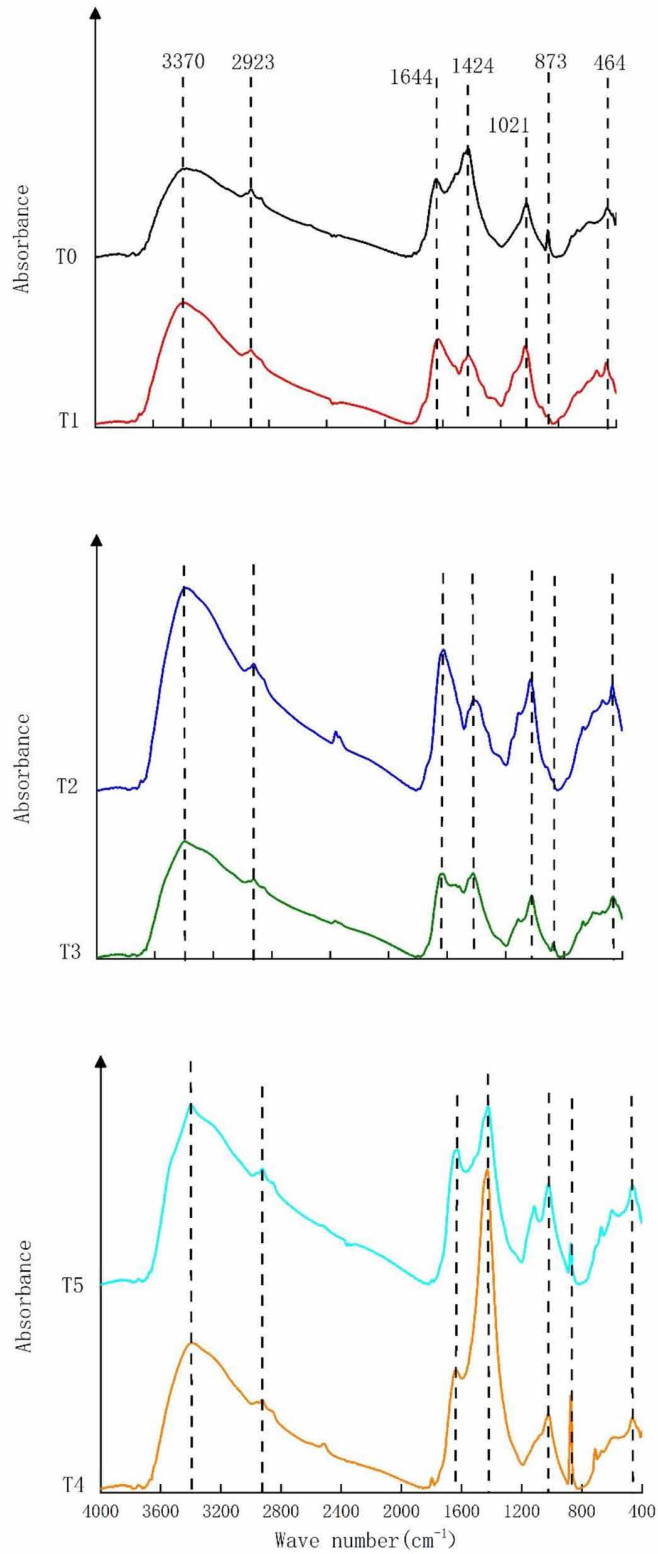


Table 5. The relative intensity of each absorbance peak

Treatment	Absorbance Spectrum()					
	3370	2923	1644	1424	1021	873
T0	65.63%	1.14%	6.21%	9.96%	15.88%	1.17%
T1	67.72%	1.42%	10.55%	5.30%	14.91%	0.10%
T2	68.59%	1.38%	11.37%	5.77%	12.82%	0.07%
T3	78.36%	0.65%	6.95%	6.91%	6.64%	0.49%
T4	54.33%	0.16%	2.46%	30.21%	10.64%	2.19%
T5	75.38%	0.26%	6.99%	10.06%	6.41%	0.90%

the absorbance peak of T4 decreased when T4 was at 3370 cm^{-1} , indicating a decrease in the total amount of carbohydrate and aromatic compounds. The absorbance peak of this same position in the remaining treatment groups increased, with T3 composting having increased the most. Thus, the changes in carbohydrates and aromatic compounds were more significant. The absorbance intensity decreased when T3, T4, and T5 were in 2923 cm^{-1} , indicating that the amount of aliphatic decreased in the composting process, while the absorbance intensity in T1 and T2 increased. The absorbance intensity of T1 and T2 increased at 1644 cm^{-1} , indicating an increase of aromatic analogues and acylamide compounds with a nitrogen group. The absorbance intensity of T5 was slightly decreased in this situation, while T4 increased, indicating the amount of aromatic compound decreased. The absorbance intensity of T4 and T5 increased at 1424 cm^{-1} . The increase of T4 was most obvious, increasing from the original 9.96% to 30.21%, which indicated the accumulation of humic acid and organic acid substances continuously, thus reducing the pH value of the stack (compost). All absorbance intensities of treatments were below T0's at 1021 cm^{-1} , possibly because the polysaccharide analogues continuously decomposed during the composting process. The appearance of an absorbance peak at 873 cm^{-1} indicated the five treatments have carbonate substances in the composting process. The five treatments have similar major functional groups in the composting process. Since the sources of composting materials are complicated, the absorbance peak of different functional groups may overlap and interfere with each other.

Seed Germination Tests

Since the final product of the compost will be used in agricultural production, seed germination tests are the most direct and reliable method of measuring the toxicity of planting using the compost's products (Su et al., 2016). T0 is a pure water control group. By using seed germination rate, it can be easily determined whether the stack contains toxicity. According to Table 6, the seed germination rate can be close to 50% for T0 after 24 hours, while after 48 hours, the seed germination rate can

Table 6. Seed germination rate of various treatments (%)

Time (hrs) ^a	Treatment							
	T0	T1	T2	T3	T4	T5	Sludge Without Fermentation	White Mud Without Fermentation
24 h	48±2	39±7	34±6	17±5	31±3	10±6	6±6	7±4
48 h	98±2	94±6	95±5	89±4	93±4	88±4	85±5	76±4

^aNote: hrs = hours

be close to 100%. The germination cycle in this environment for the cabbage seed is two days. The germination rate of each treatment ranked from low to high after 24 hours was: T1>T2>T4>T3>T5. The difference in seed germination rate after 24 hours and 48 hours was greater in each treatment (T1 to T5) than the difference in seed germination rate after 24 hours and 48 hours for T0, the control group. T5 had the lowest seed germination rate among the treatments.

The seed germination rate of each treatment increased after 48 hours. However, the final results showed that the ranking of the seed germination rate was similar to that of 24 hours. The final germination rates of T1, T2, and T4 were all above 90%; thus, one may conclude the toxicity of these treatments is low. The germination rates of T3 and T5 were low, although their final germination rates were close to 90%. There were different degrees of inhibition in time, while the inhibition in the 24-hour stage was the most obvious. The germination rate of each treatment was greater than the un-composted sludge, indicating the composted sludge was less toxic than the un-composted sludge, resulting in low inhibition of seed germination. T4 had a higher germination rate than that of un-composted sludge, mainly because the latter's strongly alkaline and high calcium carbonate content inhibits seed germination to some extent. Table 5 shows that, although the germination rate of each treatment against the pure water control group is generally slightly lower, the toxicity of each treatment has a small negative effect on seed germination. T1, T2, and T4 germination rates were higher than T3's, which shows that adding bark and white mud to the compost is more ideal than composting only with sludge.

Each treatment has different degrees of inhibition in time. Placing just composted sludge for a few days and waiting until it is more stable before application can reduce its adverse effects, to a certain extent. Paper pulp sludge as the main compost material either contains less toxic and harmful substances from the start or produces less harmful substances during the composting process, which allows the final product to be more advantageous for plant growth and agricultural use.

Experiment Conclusions

This experiment used paper pulp sludge to study resource recovery, and added bark, white mud, pyroligneous acid, molasses, and sludge from food plants as composting accessories. Researchers studied these accessories' effect on composting and analyzed the temperature, moisture content, pH, E_4/E_6 , and organic carbon content during the composting process.

The study found that the sludge microorganisms have higher chemical activity in the initial stage of the composting process, and the addition of auxiliary materials consisting of pyroligneous acid, sludge and waste liquid from food plants, and molasses, can make the temperature of the compost rise significantly, and can speed up the maturity of the compost. The temperature of the fermentation of pulp sludge compost is low and lasts only one to two days. Analyzing the stack after it was finally in a decaying state, the inhibition to seed germination was found to be small. It was noted that by using the aerobic ventilation composting method, composting fermentation time could be shortened to achieve compost maturity.

The addition of white mud could adjust the pH of the stack, so that microorganisms could reproduce at a more appropriate level of acidity and alkalinity. Organic carbon had a short-term rise in the composting process, indicating the microorganisms reproduced and were highly active in the process. In analyzing the variations of carbohydrate by FTIR, the aromatic structure components and degree of aroma increased, while the degradation of polysaccharides was used for the growth and reproduction of microorganisms.

The seed germination rate of each treatment was higher than that of the un-composted sludge and white mud. The toxicity of the sludge and white mud after composting was low, which had less of an adverse effect on seed germination. Placing just-composted sludge for several days before application could reduce the adverse effects on seed germination, to a certain extent.

The effect of composted sludge on crops was smaller than the direct application of pure sludge. Adding bark is beneficial to the process of sludge composting, especially when the sludge-to-bark

ratio is 2:1. This ratio can make the composting temperature rise above 50°C which is conducive to the decay of the stack, and minimally inhibits seed germination.

Adding a moderate amount of white mud can regulate the pH of the stack, so that microorganisms can reproduce at more appropriate acidity levels to promote decay. The addition of sludge and waste liquid from the food factory was to provide the source of bacteria for the stack. Adding molasses was to provide a carbon source, make up for the shortage of carbon sources in simple sludge, so that the microorganisms could reproduce in large numbers and degrade organic matter to promote the fermentation and decay of the stack.

CONCLUSION AND SUGGESTIONS

This study used the aerobic stacking method. Although this method has been practiced for a long time, it uses waste containing higher nitrogen as excipients, which will significantly shorten the stacking time required by the general traditional aerobic composting method and obtain a reasonable C/N ratio of compost. It has an economic savings benefit for the subsequent adjustment of compost composition. In addition, this method uses intermittent ventilation to maintain a good aerobic state of the fermentation process, which is also helpful for saving fermentation costs.

From a technology development aspect, experiment results show that the aerobic process and the appropriate carbon source can promote the rapid decay and fermentation of pulp sludge. After adding different proportions of bark, the relative intensity of the infrared spectral absorbance peak of the aliphatic compound and aromatic compound in compost doubled, showing an increase in the degree of aroma and the overall decay of the compost. Proper addition of white mud can adjust the acidity of the compost and help mature the compost.

The seed germination rate experiment showed that the germination rate of the sludge compost treatment with bark is slightly higher than the treatment without adding bark, and the seed germination rate of the treatment with white mud is also higher than the treatment without adding white mud. This experiment shows that composting the pulp sludge through an aerobic process can shorten the time it takes the compost to mature, can reduce the growth inhibition of plants, and can be applied to the production of fertilizer and soil conditioner. All of these can accomplish material circulation.

From the factory management aspect, this paper proposes a method of turning paper pulp sludge into fertilizer that has achieved initial success, and that has effectively shortened the time needed to convert pulp into organic fertilizer. It greatly improved the plant's efforts to reduce production waste and strengthened its goal of promoting sustainable forestry. In order to fulfill the various environmental protection regulations enacted by the Chinese government in recent years, the factory pulp source is taken from the forest farm planted by the company. Only trees that are at least six years old can be used for making paper pulp, to align with size standards. Processing wastewater and sludge produced by the pulping process is a great challenge. Wastewater and sludge treatment have met the requirements of government regulations, and because the sludge is high quality and is from the original pulp, it has the potential to become a value-added product. One of the tasks of the research group was to find other valuable elements in sludge, beyond the production of organic fertilizer.

The issue of environmental sustainability deserves the paper industry's continuous attention and efforts. The paper industry is a high pollution industry in the public's mind, but it can also be a very environmentally friendly industry (CPC, 2020; Education for good, 2018) The top managers of the paper industry need to put more effort toward environment friendly objectives.

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Chen; data curation, C. Chen and J. Chen; writing/original draft preparation, C. Chen and J. Chen; writing/review and editing, C. Chen and J. Chen; visualization, C. Chen and J. Chen; supervision, C. Chen and J. Chen; project administration, C. Chen and J. Chen. All authors have read and agreed to the published version of the manuscript. The authors contributed equally and are co-first authors.

CONFLICTS OF INTEREST

The authors of this publication declare there is no conflict of interest.

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