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REVIEW

Effect of exogenous additives on heavy metal passivation and nitrogen retention in pig manure composting

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KEYWORDS

Additives, composting, heavy metals passivation, nitrogen retention, pig manure

HIGHLIGHTS

- Research on heavy metal passivation and nitrogen emissions is necessary for the pig industry.
- Mechanism of heavy metal passivation and nitrogen retention by different additives was introduced.
- Development and prospect of metal passivation, nitrogen preservation technology were discussed.

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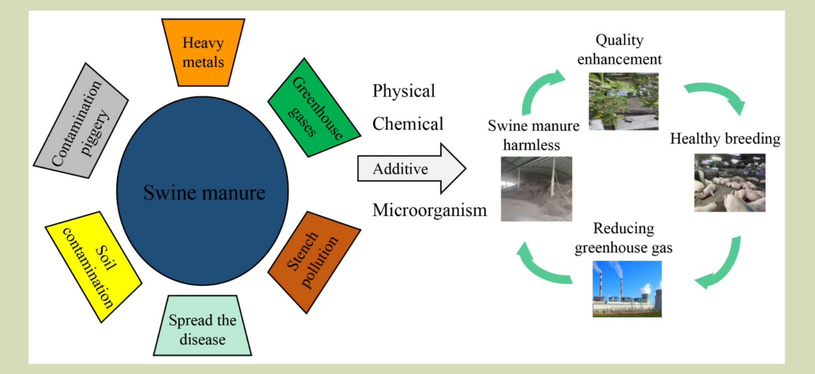
GRAPHICAL ABSTRACT



ABSTRACT

The widespread use of feed additives in intensive and large-scale pig farming has resulted in high levels of heavy metals in pig manure. The long-term application of organic fertilizers containing high levels of heavy metals leads to the accumulation of heavy metals in the soil, which not only causes heavy metal pollution in the soil, and also affect food safety and endanger human health. Composting is an economical and effective technical measures to achieve environmentally-sustainable treatment of pig manure and is a practical method to reduce the problem of heavy metals and to improve the resource value of pig manure. The composting process is accompanied by high temperatures and the production and emission of gases, and also lead to changes in the nitrogen content of the compost and provide opportunity for heavy metal passivation additives. This paper summarizes the forms and types of heavy metals present in pig manure and reviews the progress of research as well as the techniques and problems of in the composting process, and provides recommendations for research on heavy metal passivation and nitrogen retention in pig manure composting.

1 Introduction

In recent years, with the expansion of the livestock industry in China and the increasing intensive production, the number of pigs slaughtered in China reached 0.4 billion at the end of 2020[1]. The rapid development of scale and intensification of the pig industry has produced an increasing amount of pollutants[2]. The annual production of livestock manure in China in 2021 is about 3.8 Gt[3]. A survey showed that as a proportion of livestock and poultry manure in China was about 29% of gas emissions come from pig manure, and for heavy metal pollution pig manure contributed about 71%[4].

Heavy metal contamination from pig manure is widespread in agricultural soils in China, with average concentrations ranging from 0 to 10 $mg\cdot kg^{-1}$[5]. A survey found that in some areas pig manure contained 767 $mg\cdot kg^{-1}$ of copper and 3130 $mg\cdot kg^{-1}$ of zinc[6], and heavy metals are present in agricultural soils for a long time and most of them are toxic, causing impacts on the growth and development of soil organisms and crops, and even affecting animal and human health through the food chain.

Nitrogen, one of the important nutrients for crop growth, but during the fermentation of compost, there is a significant release of greenhouse and harmful gases, including CO_2, NH_3, CH_4 and N_2O. Most nitrogen loss is through NH_3, with a smaller proportion through N_2O. China aims to be carbon neutral by 2035, so greenhouse gas emissions from animal industries cannot be ignored. Understanding and control of greenhouse gas emissions from manure is necessary. Nitrogen retention in pig manure compost is as important as the passivation of heavy metals. Also addition of heavy metal passivators can be used to change the form of heavy metals present and also changes the composting environment, thus affecting the release and retention of nitrogen.

Therefore, it is crucial to investigate the origin, form and type of heavy metals in pig manure, passivation techniques, and the impact of passivators on nitrogen retention in compost in order to make this treatment of pig manure suitable for applying the material to agricultural fields.

2 Sources, forms and bioavailability evaluation of heavy metals in pig manure

2.1 Heavy metal sources

The use of high doses of Cu and Zn has been increasing since it was discovered that there metals promote piglet growth, reduce diarrhea and increase daily feed intake and performance[7]. This has resulted in a widespread problem of excess Cu and Zn in pig manure[8]. Although China has clearly defined the concentration thresholds of heavy metals in agricultural soils in the Environmental Quality Control Standards for Contaminated Soil on Agricultural Land[9]. However, some companies still add large amounts of these feed additives containing high doses of metals for financial gain. In general, the concentration of feed Cu can reach 200–300 $mg\cdot kg^{-1}$ and selenium 0.3–0.5 $mg\cdot kg^{-1}$. However, 95% of Cu^{2+} and Zn^{2+} in $CuSO_4$ and ZnO is not absorbed and utilized livestock. Pigs have a low utilization rate of metal additives in feed, most of which are eliminated in feces and urine[10]. Feed processing also results in residues of trace metal elements, which combined with the misuse of feed additives, leading to serious pollution of agricultural soils by heavy metals such as zinc, copper, lead, cadmium, chromium, mercury, arsenic and nickel[11,12]. Soil heavy metal contamination affects the survival of soil microorganisms, leading to a decrease in soil microbial load and changes in microbial activity and structure[5]. In addition, heavy metal migration in soil following rainwater can lead to water pollution, deteriorating water quality and toxicity to aquatic organisms.

The most concentrated heavy metals in pig manure are mostly Cu, Zn and Cr[13], and this mostly comes from feed additives. It has been found that the excess Cu in manure of fattening pigs and piglets was 82% and 97%, respectively, and the content of Zn all exceeded the standard[14]. Heavy metal overload has become the biggest source of pollution in pig industry in China[15].

2.2 Heavy metal forms

Currently single and step-by-step chemical extraction methods are often used to study the morphological distribution of heavy metals in livestock manure and amended soil[16]. In 1979, Tessis et al.[17] proposed that heavy metals in soil or sediment could be classified after a five-step sequential extraction method as exchangeable, carbonate-bound, Fe manganese oxide bound, organic bound and residue, in order of biological availability[18]. A standardized extraction protocol was also proposed by the European Bureau of Reference Materials and modified in 1999, referred to as the BCR stepwise extraction method. This method classifies the heavy metals into a weak acid extracted state, a reduced state, an oxidized state and a residue state[19]. For heavy metals in pig manure, the biological effectiveness of different forms varies. The main way to control heavy metal pollution in livestock and poultry manure is to fix heavy metals in livestock and poultry manure by changing their forms and reducing their mobility and availability, and then to remove them from the manure. The main method to control heavy metal pollution in livestock manure is to use livestock manure composting[20,21]. It has been shown that composted sludge has about 7.3% to 16% less heavy metals due to the dilution effect of conditioning agents and significant changes the morphology of heavy metals[22]. More recently, it has been found that an effective way is to add heavy metal passivator to the composting process to transform the form of heavy metals from high to low activity, reducing bioavailability.

2.3 Heavy metals bioavailability evaluation

The bioeffective properties of heavy metals reflect the degree of their uptake and utilization by organisms, which is a key parameter in heavy metal pollution risk assessment and is receiving more attention in practical research and application[23,24]. However, biological effectiveness is based on both chemical and biological concepts. Nelson[25] defines the bioavailability of heavy metals as a property of heavy metals that produces toxic effects on organisms or leads to uptake by organisms, including toxicity and bioavailability, as evaluated by indirect toxicity data or organism concentration data. Biological effectiveness has also been viewed as a dynamic process that must be considered in relation to the environment around the organism[26] but there is no unified international standard for the evaluation of biological effectiveness of heavy metals[27,28].

The study of morphological distribution of heavy metals in pig manure is important for predicting the biological effectiveness and environmental risk of heavy metals in pig manure, which can provide more useful information on the migration transformation, biological effectiveness and leaching of heavy metals after applying livestock manure to agricultural fields[29].

3 Nitrogen and heavy metals

Nitrogen is an essential element for plant growth and one of the main elements of the compost microenvironment[30]. Nitrogen conversion in composting systems is a fundamental biochemical process mediated by a variety of microorganisms[31], mainly including ammonification, ammonia assimilation, nitrification and denitrification (Fig. 1). These processes occur simultaneously with heavy metal passivation. Compost heavy metal passivators are mainly divided into three categories: physical, chemical and microbial. Physical technology mainly uses activated carbon, zeolite, bentonite and other substances with high adsorption capacity, which can effectively adsorb heavy metals and reduce their biological effectiveness. These additives also affect the emission of nitrogen from the composting process. Likewise, chemical and microbial additives for heavy metal passivation can affect nitrogen emissions. Therefore, it is necessary to study heavy metal passivation and nitrogen retention in parallel.

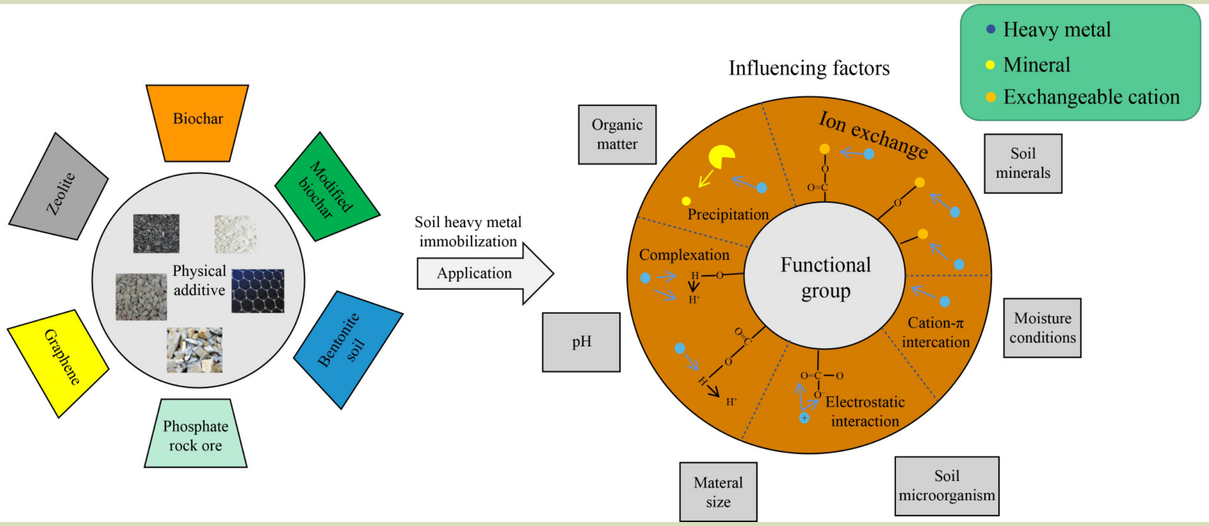


Fig. 1 Mechanisms of physical additives passivation of heavy metals.

4 Methodological initiatives to improve the efficiency of heavy metal passivation

Heavy metal contamination is a major limiting factor in the application of composting products. Therefore, determining how to effectively reduce the toxicity of heavy metals in the composting process has become a primary concern in composting[32]. There are many studies on heavy metal passivation methods and the effectiveness of passivation.

4.1 Physical additives

Biochar, zeolite, bentonite and other additives has been extensively researched and used for the safe, minimization of toxins as well as resource recovery of agricultural waste[33,34]. In agricultural production, the application of biochar can reduce soil nutrient runoff[35], improve land fertility and raise the pH of acidic soils duet to the high specific surface area, pore size characteristics and strong adsorption capacity[36]. Improving the final quality of the compost can effectively reduce the biological impact of manure in terms of organic or inorganic pollutants[37].

Figure 1 shows the interaction mechanism between physical additives and fecal heavy metals. In general, the interaction between physical additives and fecal heavy metals involves cation exchange, complexation, coprecipitation, electrostatic adsorption, cation-π interactions and physical adsorption[38].

Kong et al.[39]. found that the addition of biochar, calcium magnesium phosphate fertilizer and spent mushroom substrate to compost with 42% to 53% exchangeable zinc and 2.5% to 39%, reducible zinc, the exchange zinc content was reduced by 5.9% to 6.8%. Zinc passivation achieved by the treatments increased in the order: CK < biochar < mushroom substrate < calcium magnesium phosphate fertilizer. Ingelmo et al.[40] found that Zn availability was positively associated with the fulvic acid (FA) fraction and negatively associated with the humic acid fraction. Therefore, the availability of zinc was strongly affected by the humification process.

Yang et al.[41] investigated the immobilization properties and mechanisms of heavy metals using biochar modified by KH_2PO_4 during the anaerobic digestion of pig manure. This was found to it make these heavy metals more stable and reduce the biological risk. The modified biochar was quite efficient in passivating copper and lead. Therefore, KH_2PO_4 modified biochar can be used as a new type of heavy metal passivation remediation agent. The effect of additives biochar and humic acid, on the passivation of heavy metals in pig manure compost was investigated by Zhou et al.[42] Their results showed that the addition of fertilizer passivators to compost products with sawdust charcoal improved the passivation of heavy metals during composting. The deactivation rates of copper, lead and zinc were greatly reduced. The application of compost also increases the yield of oilseed rape. Chen et al.[43] added biochar and H_3PO_4 modified biochar to explore the effects on heavy metal stability, antibiotic resistance genes. Biochar and H_3PO_4 modified biochar significantly reduced the concentration of DTPA extractable copper and zinc compared to the control. There was also a significant reduction in the total abundance of nine antibiotic resistance genes and five metal resistance genes in the compost products treated with biochar and H_3PO_4 modified biochar compared to the control. Wang et al.[44] studied the effect of adding different proportions of biochar to pig manure to increase biogas production and reduce the biological effectiveness of heavy metals, while the potential biological risk of heavy metals in the digestate was also assessed. The results showed that methane production significantly increased with the addition of different proportions of biochar. Also, there were significant changes in heavy metal morphology in each group. Cui et al.[45] investigated the correlation between bacterial diversity and heavy metal fractions during the composting of with a range of doses of biochar. Their results showed that the bioavailability coefficients of zinc, copper, cadmium and lead were significantly reduced in all groups after the addition of biochar. This indicates that biochar had an effect on bacterial population structure and heavy metal passivation in the pig manure compost. The bioavailability coefficients of heavy metals were significantly reduced by biochar-amended compost compared to the control. Addition of 12% date palm biochar to pig manure compost resulted in significantly higher passivations of Cu and Zn. Also, the maximum composting temperature was advanced regardless of the percentage of date palm biochar added[46]. Composting was significantly improved and the composted product was less toxic[47]. The effect of heavy metal passivation through adsorption and humification during aerobic composting of zeolites of various particle sizes has been investigated[48]. The morphological changes of heavy metals during composting were also observed. In comparison to the control, zeolites significantly reduced the bioavailability coefficients of heavy metals, especially copper, cadmium and lead. Redundancy analysis and structural equation modeling indicated that the passivation promotion of heavy metals was comparable for coarse and fine zeolites and was accelerated by surface adsorption for cadmium and lead and by humification for copper. Wang et al.[49] study inorganic additives phosphate rock and boron waste were added to the compost and monitored for temperature, pH, EC, OM, TN, TP and other indicators during composting. It was found that phosphate rock and boron waste provided certain nutrients and promoted the degradation of organic matter in the humus chelating Cu and Zn, and the passivation of Cu was promoted. Xu et al.[50] investigated the effect of sodium hydroxide addition on the properties and environmental risks of heavy metals in pig manure biochar. It was found that the addition of various doses of sodium hydroxide at different temperatures increased pH, EC, ash content, yield, aromaticity and hydrophilicity, while promoting the conversion of mobile fractions of Cu, Zn and Cd to oxidizable fractions. This study also found that NaOH-assisted pig manure pyrolysis can be an effective method for immobilizing heavy metals. The addition of NaOH promoted the formation of more stable metal compounds and increased the alkalinity of the biochar. Li et al.[51] natural bentonite and microwave-treated bentonite were added to composted pig manure. The analysis of the samples and the extraction by BCR method showed that the percentage of heavy metal Cr in the oxidizable and residual state increased, indicating that anaerobic fermentation reduced the biological activity of heavy metal Cr in pig manure. Microwave treated modified bentonite effect is better than the addition of natural bentonite treatment. At the same time, surface source pollution is reduced and energy material and high quality organic manure is obtained. It was found that the addition of maize straw significantly increased the degradation of organic matter in pig manure during composting[52]. The migration rate of Cu and Zn decreased at an initial C:N of 25:1 and eventually 90% Cu was the residual fraction. In addition, the compost mixture with an initial C:N of 25:1 accelerated the decrease in urease activity compared to the compost mixtures with an initial C:N of 15:1 and 20:1. Hui et al.[53] use of agricultural lime and a newly designed alabaster activated carbon composite (AACC) in a pig manure composting system enhanced the stability of heavy metals and improved the degradation of antibiotics. The addition of AACC reduced the enrichment of chromium, cadmium, lead and arsenic during the composting process. Concurrently, the high heavy metal hazard in the manure inhibited the dissipation of antibiotics. The addition of lime and AACC promoted the fixation of heavy metals in the manure and accelerated the breakdown of antibiotics. Therefore, mixing pig manure and AACC for composting not only significantly reduced compost contaminants, but also improved nitrogen conversion and reduces the phytotoxicity of compost products. Liu et al.[54] found that the addition of magnetic Fe_3O_4/FA composite additive to pig manure anaerobic compost could promote methane production in the anaerobic digestion system by adding different concentrations of Fe_3O_4/FA. Also, the introduction of Fe_3O_4/FA composites could effectively control the migration of heavy metals and Fe_3O_4/FA enhanced the passivation of Cu and Zn in the solid digestion residue. The mechanism of heavy metal passivation by Fe_3O_4/FA mainly involves physical adsorption during anaerobic digestion. This process converts heavy metals into stable mineral precipitates and reduces the solubility and mobility of heavy metals.

4.2 Chemical additives

Chemical techniques are mainly used to remove heavy metals from surface acidification, ion exchange, solubilization, complexing agents and surfactants. The diagram below shows the mechanism by which chemicals passivate heavy metals (Fig. 2). The chemical additives for treatment fecal heavy metals mainly act as acidification treatments, surfactants and complexing agents. Acidifiers remove large amounts of heavy metals by dissolving them. Surfactants and complexing agents convert insoluble metals into soluble metals or complexes which can be removed.

Liphadzi et al.[55] investigated the conversion of heavy metals in compost from an insoluble compound to a soluble ionic state as the principle of the chemical approach to remove heavy metals from compost. The most studied chemical method is acidification, and the other method is the use of complexing agents.

Acidification treatment usually uses sulfuric acid, hydrochloric acid, nitric acid, phosphoric acid, organic acid and other chemical reagents to dissolve a wide range of metals. However, it has been shown that when sulfuric acid is used alone, the removal effect of copper and lead is not ideal[56]. While using 1:1 hydrochloric acid/sulfuric acid to treat sludge the removal rate of heavy metal copper, lead, zinc and tin can achieve 60% or more. The use of nitric acid leaching of sludge can achieve copper and arsenic removal rates of 87% and nickel removal of up to 100%[57].

The use of organic complexing agents for the removal of heavy metals is based on the principle that the addition of complexing agents to insoluble metal compounds converts them into soluble metal complexes for removal. Studies have shown that organic complexing agents (EDTA and DTPA) are very effective in removing heavy metals. For example, EDTA can form stable compounds with many heavy metals and $0.1\ \mathrm{mol \cdot L^{-1}}$ EDTA was able to remove Pb. It was found that the extraction of Pb by EDTA could reach over 60%[58]. A study on the removal of heavy metals by surfactants found that surfactants mixed with EDTA were effective in removing heavy metals in the order: Cd > Pb > Zn[59]. Surfactants can remove

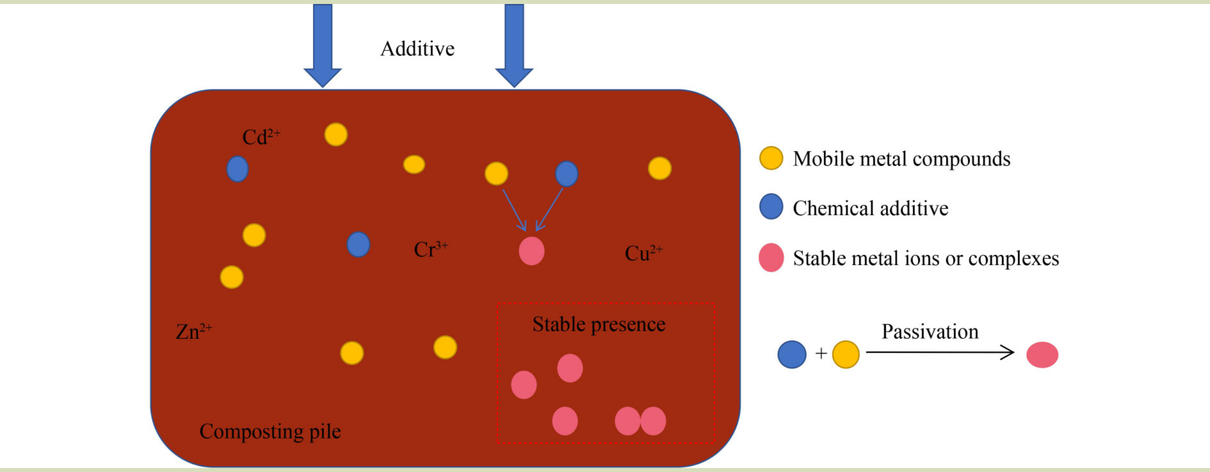


Fig. 2 Chemical passivation agent passivation mechanism.

heavy metals from soil by desorption, but may have serious environmental impacts. So, it is important to use rapidly degradable and non-toxic surfactants to remove heavy metals from manure.

4.3 Microbial additives

The addition of microbial agents to the composting process has become one of the most effective ways to reduce the biological activity of heavy metals[60]. The dosage of microbial additives in compost generally ranges from 0.05% to 5.0%, and the amount of bacterial agent varies for different times of composting[61]. Phosphate mineralizing bacteria can also mineralize heavy metals. Many bacteria and fungi can mineralize heavy metals in soil and water bodies by hydrolyzing inorganic phosphate sources (apatite minerals) or organic phosphate sources (phenolphthalein diphosphate, glycerophosphate, acetylmethamidophosphate, glycerol 2-phosphate and phytate) in the presence of phosphatase or phytase to release phosphate, thereby reducing the mobility of toxic ions[62]. The diagram below shows the mechanism of mineralization by phosphate-mineralization bacteria (Fig. 3).

Li et al.[63] studied composting supplemented with different types and doses of biochar and found these treatments increased fixation of heavy metals in pig manure composts. Seventy percent of copper was fixed by addition of peanut shell biochar and microbial preparations. The fulvic and humic acid-like substances in the humus increased to nearly twice the pre-compost level. The conversion of copper and zinc to a more stable state was closely related to the formation of fulvic and humic acids, and the immobilization and humification of

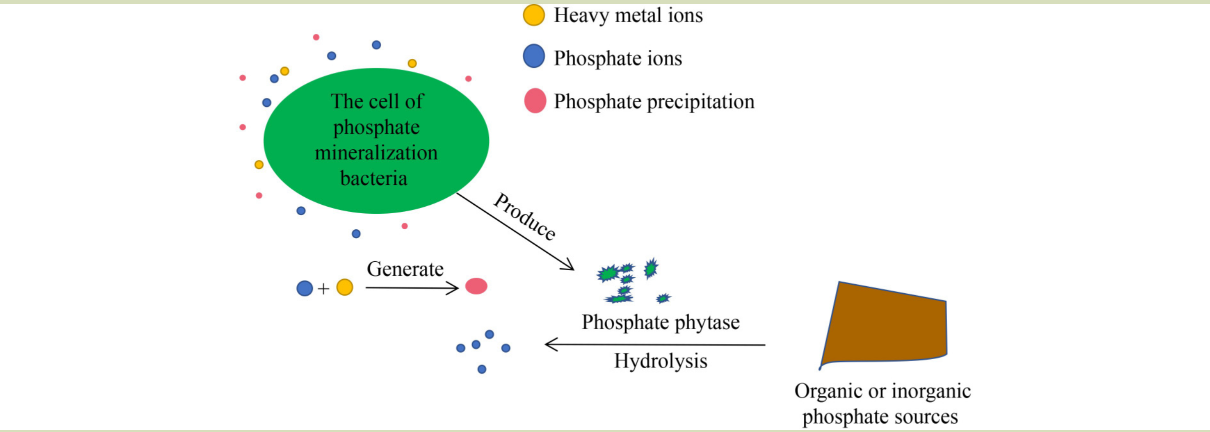


Fig. 3 Mechanism of mineralization by phosphate-mineralization bacteria.

copper and zinc were positively correlated with pH. Cui and Wang[64] used *Thiobacillus* ferrous oxide amended sludge to examine if biological leaching could effectively dissolve the heavy metals in the sludge. After 4–10 d of biological leaching, the removal rates of Cr, Cu and Zn reached 80%, 80% and 100%, respectively. Cao et al.[65] found that anaerobic fermentation could mitigated heavy metal contamination with migration rates higher under high temperature ana fermentation. The metabolic activity of microorganisms such as fungi, bacteria and algae can be used to effectively mitigate heavy metal toxicity and contamination. Luo and Wu[66] report that the addition of earthworms to the compost was able to reduce the heavy metal content, but the reduction was dependent on the in earthworm species. The production of biogas and passivation of heavy metals during anaerobic digestion were analyzed using treated rice straw as feedstock. It was found that more than 80% of the copper was present in the oxidizable and residual fractions, also, that zinc was effectively improved by microwave and ultrasonic treatment for adsorption[67]. The addition of rice straw also effectively improved the passivation efficiency of copper and zinc. Therefore, the addition of rice straw facilitated the passivation of heavy metals, but the cost for removing heavy metals in the substrate and pretreatment needs to be further investigated.

4.4 Comparison of heavy metal passivation methods for pig manure

By comparing the operability, efficiency, cost effectiveness and environmental safety of several heavy metal removal methods, it was found that physical methods are less efficient than chemical and microbiological methods for removal of heavy metals from sludge, but such methods were less expensive and simpler to operate[68]. The use of chemical and microbiological methods to reduce the content of heavy metals can achieve in higher heavy metal removal rate, but the high cost, complex operation and environmental friendless limit their practical application[69]. The following table lists the advantages and disadvantages of the different additives available today (Table 1).

5 Effect of exogenous additives on nitrogen retention in compost

Nitrogen losses during composting are mainly as NH_3 and N_2O emissions and account for 79% to 94% and 0.2% to 9.9% losses, respectively[70]. The main pathways of nitrogen conversion in aerobic composting include ammonification, nitrification, denitrification and biofixation (Fig. 4)[71].

NH_3 emissions are mainly produced by the biodegradation of nitrogenous substances. Ammoniation, also known as deamination, is a process by which microorganisms decompose organic nitrogen compounds to produce ammonia[72] by the following two processes: (1) protein is hydrolyzed into peptide by the catalysis of microbial protease; then (2) deamination, which primarily involves the separation of amino acids to produce NH_4^+/NH_3. The most common type of amino acid degradation is oxidative deamination. NH_4^+ and NH_3 maintain chemical equilibrium through $NH_3{\cdot}H_2O$ in the aqueous phase. Under high pH and high temperature conditions, the equilibrium moves in favor of producing NH_3, which results in the release of NH_3 into the atmosphere.

Nitrification is a critical step to accelerate conversion of ammonium nitrogen to oxidized N and reduce ammonia

Table 1 Advantages and disadvantages of different additives

Type of additives	Advantages	Disadvantages	References
Physical additives	Easy operation Low cost Wide range of sources	Less effective Low repeatability	[40–51]
Chemical additives	Better effects Fast results	Difficult operation Higher costs Waste liquid pollution Application difficulties	[56–60]
Microbial additives	Reusable Better effects Great prospects	High technical demands Higher costs Further research needed	[63–67]

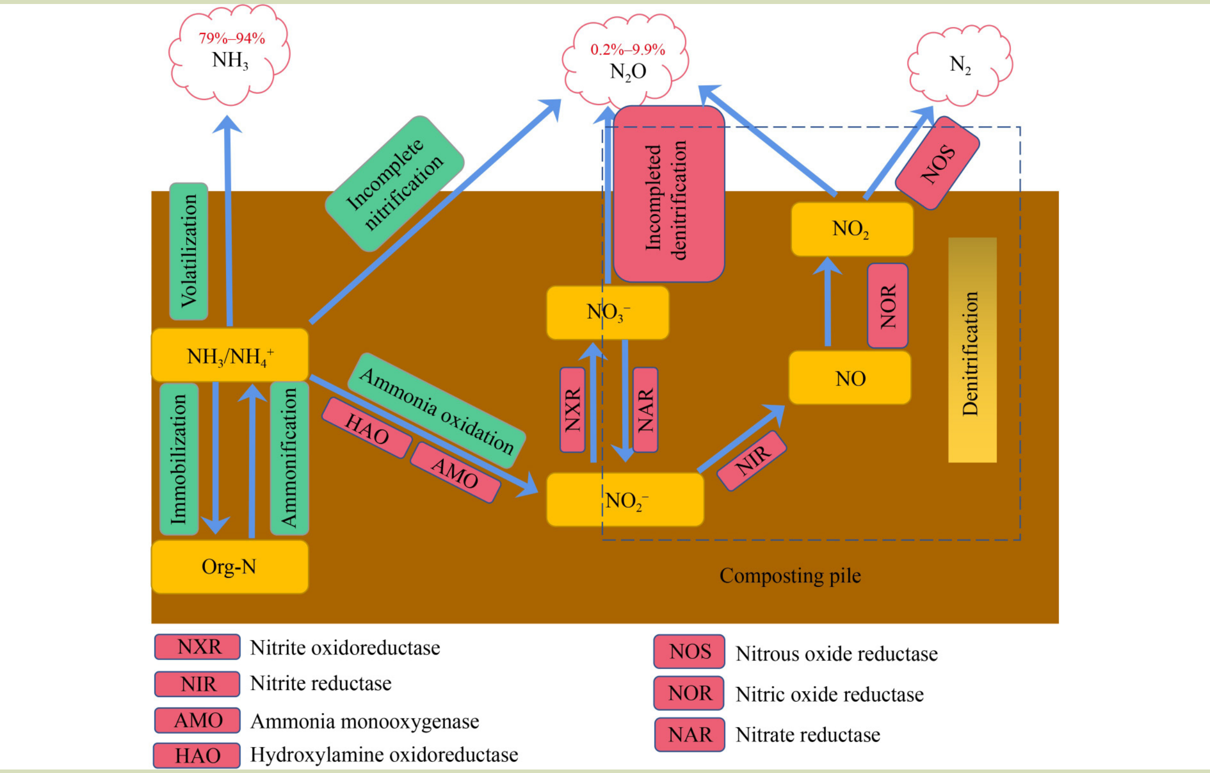


Fig. 4 Nitrogen conversion processes in composting.

volatilization. Ammoxidation is the first and rate-limiting step of nitrification, including two reactions, wherein NH_3/NH_4^+ forms hydroxylamine by ammonia monooxygenase and then NO_2^- is formed by hydroxylamine oxidoreductase[73]. Given that nitrifying microorganisms are sensitive to temperature, their growth is inhibited by high temperature in composting, so that nitrification is weak in the thermophilic phase of composting[74].

Denitrification is a process in which microorganisms successively reduce NO^{3-} to NO_2^-, NO, N_2O and N_2[75]. In general, microbial denitrification occurs in the anoxic microenvironment of composting, including four enzymatic reduction reactions corresponding to the following four reductases: nitrate reductase, nitrite reductase, nitric oxide reductase, and nitrous oxide reductase. Nitrite and nitrous oxide reductases contribute to denitrification.

5.1 Physical additives

Physical additives are either natural or synthetic, with large specific surface area, strong adsorption, strong ion exchange ability, through its adsorption capacity to reduce the conversion of NH_4^+ to NH_3[48]. Similar to heavy metal physical passivation, physical additives (Table 2) can significantly reduce NH_3 and N_2O emissions during composting, with average values reaching 35% and 60%, respectively.

The effects of adding bamboo charcoal and bamboo vinegar liquid to compost piles on nitrogen retention and copper and zinc fixation during the composting of pig manure were investigated[78]. Total Kjeldahl nitrogen loss, as well as copper and zinc migration, decreased with increasing bamboo charcoal addition. Total Kjeldahl nitrogen loss and mobility of copper and zinc were significantly reduced compared to the control. The addition of BV further reduced the total Kjeldahl nitrogen losses. It also influenced the changes in compost temperature, pH and germination index (GI), shortening the time to the high temperature phase of composting, reducing the pH at the high temperature stage and increasing the GI of the compost product. Therefore, the addition of bamboo charcoal or bamboo charcoal + vinegar to pig manure compost is an effective way to reduce total Kjeldahl nitrogen losses and control Cu and Zn migration. The effect of additives such as apple pomace, bentonite and calcium superphosphate on nitrogen, carbon and phosphorus conversion and compost

Table 2 Effects of physical additives on nitrogen retention in pig manure compost

Feedstock	Additive	Impact on N conservation (%)				Reference
		TN	CH_4	NH_3	N_2O	
Pig manure and maize straw	$Ca(H_2PO_4)_2$ + $MgSO_4$	65↓	59↓			[72]
Pig manure and maize straw	Guano crystals + nitrification inhibitors			50↓		[76]
Pig manure and wheat straw	Biochar (10%)^a	54.5↓	51↓			[77]
	Biochar (10%)^a + zeolite (10%)^a			63.4↓	78.13↓	
	Biochar (10%)^a + zeolite (10%)^a + wood vinegar solution (2%)^a			74.32↓		
Pig manure and sawdust	Raffinate + zeolite + biochar + wood	55.5↓	80↓			[75]
	vinegar solution		74↓	69↓		
Pig manure and sawdust	Clay (10%)^a		45↓	86↓		[78]
Pig manure and wheat straw	Calcium superphosphate	22↑				[79]
	Apple dregs	87↓				
	Bentonite		55↓			
Pig manure and sawdust	Bamboo charcoal + bamboo vinegar liquid	74↑				[78]
Pig manure and maize straw	Phosphogypsum (10%)	22↓				[80]

Note: ^{a} Dry weight.

maturity during the composting of pig manure has been investigated[79]. It was found that the additives all prolonged the high temperature phase of composting compared to the control. Calcium superphosphate helped facilitate composting and significantly reduced ammonia volatilization during the high temperature phase and increased the total nitrogen and phosphorus content of the compost, while bentonite increased ammonia volatilization and reduced the total nitrogen concentration. These results indicate that calcium superphosphate is an effective additive for maintaining nitrogen during composting of pig manure. The effect of clay on greenhouse gas emissions and humification during the composting of pig manure has also been investigated[81]. The results show that the addition of clay resulted in a significant reduction in CH_4 and N_2O emissions, promoted the degradation of organic matter and facilitated the synthesis of humic acids. Dissolved organic matter spectra showed that the addition of clay promoted the formation of aromatic carbon compounds and the degradation of aliphatic carbon. Also, the decomposition of tyrosine and tryptophan and the formation of humic acid-like substances were promoted, increasing humification[82].

5.2 Chemical additives

Chemical additives reduce nitrogen losses during composting, for example, by adding acidic chemicals or nitrification inhibitors, or by inducing guano stone crystallization[49]. The mechanism for this is shown in Fig. 5. Chemical additives reduce nitrogen loss in the composting process by adding acidic chemicals and nitrification inhibitors (dicyandiamide) and inducing struvite crystallization. Chemical additives (acidic chemicals and struvite crystallization) can significantly reduce NH_3 emissions in the composting process, but N_2O emission reduction was low.

Table 3 lists the effects of different chemical additives in pig manure and straw compost on nitrogen content of the compost. Of the effects, the retention and emission of total nitrogen, ammonia and nitrous oxide were improved by the additives.

The effects of four different struvite crystallization process (SCP) on pig manure compost were compared[76]. Four combinations of magnesium and phosphate were evaluated and compared with a control group without the addition of additives. The addition of magnesium salts and phosphates with an initial nitrogen content of 15%. SCP significantly reduced the NH_3 emissions. With the addition of sulfate, CH_4 emissions were significantly reduced and a fully mature compost was produced. In practice, $Ca(H_2PO_4)_2$ + $MgSO_4$ can be used for nitrogen retention in compost. The results showed

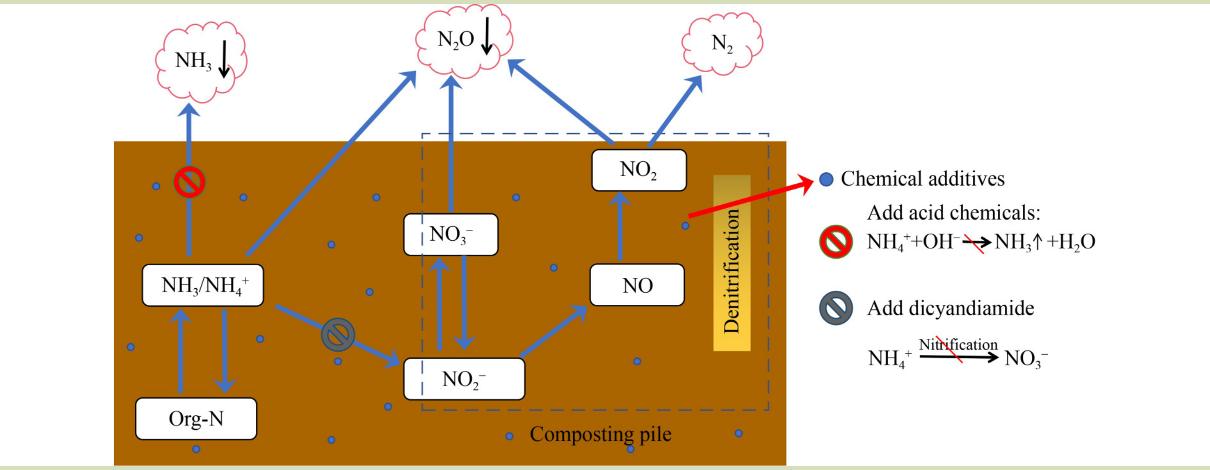


Fig. 5 Mechanism of action of chemical additives on nitrogen loss during composting.

Table 3 Effect of chemical additives on nitrogen retention in pig manure compost

Feedstock	Additive	Impact on N conservation (%)				Reference
		TN	CH$_4$	NH$_3$	N$_2$O	
Pig manure and maize straw	H_3PO_4 (15%)[a], MgO (15%)[a]	49.8↑		55.4↓		[76]
	H_3PO_4 (15%)[a], $Mg(OH)_2$ (15%)[a]	44.5↑		50.8↓		
	$Ca(H_2PO_4)_2$ (15%)[a], $MgSO_4$ (15%)[a]	53.1↑		59.1↓		
	H_3PO_4 (15%)[a], $MgSO_4$ (15%)[a]	73.9↑		81.9↓		
Pig manure and maize straw	H_3PO_4 (15%)[a], MgO (15%)[a]				78.6↓	[80]
	Dicyandiamide (10%), H_3PO_4 (15%)[a] and MgO (15%)[a]				53.7↓	
Pig manure and maize straw	Calcium magnesium phosphate (10%)[b]			42.9↓		[83]
Pig manure and maize straw	Phosphogypsum (10%)[b]			21.6↓		[84]
	Phosphogypsum and dicyandiamide (0.2%)[b]			29.3↓		
Pig manure and maize straw	Calcium magnesium phosphate (20%)			29.3↓		[85]

Note: [a] Initial molar ratio of nitrogen and [b] dry weight.

that the combination of SCP and dicyandiamide was phytotoxic, SCP significantly reduced NH_3 losses, dicyandiamide significantly inhibited nitrification at higher levels and it significantly reduced N_2O emissions[80]. Li et al.[85] in a study of the nitrogen retention properties of phosphogypsum and calcium magnesium phosphate on pig manure composts it was found that mixing phosphogypsum with calcium magnesium phosphate was effective in retaining nitrogen. The mixture of these two additives can synergistically regulate NH_3 and N_2O emissions, thereby increasing NH_4^+-N and the total N content of the compost. The addition of phosphogypsum alone can also reduce NH_3 emissions. The addition of calcium magnesium phosphate alone can slow the release of N_2O. Regardless of the addition, both additives can improve the maturity and quality of the compost by introducing additional nutrients. Liu et al.[83] found that calcium magnesium phosphate fertilizer, biochar and spent mushroom substrate significantly reduced total emissions of nitrogen- and sulfur-containing gases. It also effectively reduced emissions of NH_3, H_2S, dimethyl sulfide and dimethyl disulfide, while improving GI values.

5.3 Microbial additives

Microbial additives can contain a single strain of bacteria, but also a combinations of strains. Currently, these are commonly species of *Bacillus*, *Lactobacillus*, *Pseudomonas* and/or *Streptomyces,* or wood mold or white rot bacteria[86]. Different bacterial products have their own characteristics and the appropriate microbial preparation should be selected according to the types and characteristics of the organic matter being composted. Figure 6 shows the mechanism of reducing nitrogen loss during composting with microbial inoculants. Adding exogenous microorganisms to the compost can reduce ammonia emissions and retaining more nitrogen by changing the metabolism of carbon and nitrogen. Microbial additives can significantly reduce NH_3 and N_2O emissions during composting. During the thermophilic phase of composting, microorganisms rapidly decompose and utilize organic nitrogen substances, resulting in the formation of a large amounts of NH_4^+, and the increase in pH accelerates the transformation from NH_4^+ to NH_3. The presence of nitrate in compost was previously considered as a clear indicator of maturity[71].

As shown in Table 4, it was found that the addition of *Bacillus, Lactobacillus paracasei* or a mixture of bacteria to pig manure composting materials reduced total N losses by 22%, 17% and 35%, respectively, while the addition of *Bacillus megaterium* and bentonite reduced NH_3 and N_2O emissions.

Li et al.[90] found that inoculation with 1% *Bacillus immobilis*, prolonged the high temperature phase of composting, increased the number of heat-tolerant bacteria and the concentration of NH_4^+-N and NO_3-N, and increased cellulase activity during the cooling phase of composting. The final results indicate that inoculation of microorganisms accelerated the composting process and significantly modulated microbial function. Analysis of the effect of biochar on changes in compost bacterial communities showed that bacterial activity

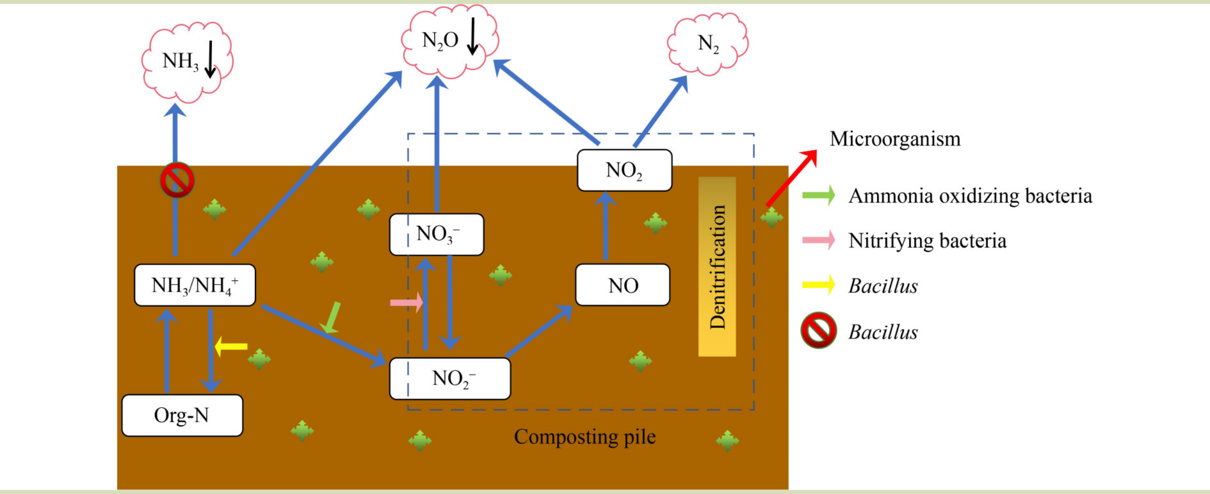


Fig. 6 Mechanism of reducing nitrogen loss during composting by inoculating microorganisms.

Table 4 Effect of microbial additives on nitrogen in pig manure compost

Feedstock	Additive	Impact on N conservation (%)				Reference
		TN	CH_4	NH_3	N_2O	
Pig manure and sawdust	*Bacillus*	22↑				[87]
Pig manure and wheat bran	*Lactobacillus paracasei*	17↑				[88]
Pig manure and wheat straw	*Bacillus megaterium* (5%)[a]			31.3↓	53.1↓	[89]
	Bentonite (5%)[a]			18.8↓	72.6↓	
	Bacillus megaterium and bentonite (mix 5%)[a]			23.7↓	63.4↓	
Pig manure and wheat straw	Ammoniation bacteria, nitrifying bacteria and nitrogen fixing bacteria	35↑				[32]

Note: [a] Dry weight.

in the thermophilic stage was controlled by the dissolved organic carbon content and the temperature of the compost mixture, while conductivity and total Kjeldahl nitrogen also influenced the maturity stage of the compost[91]. In experiments investigating the preservation of nitrogen and sulfur and the passivation of heavy metals during the composting of sewage sludge with KH_2PO_4 and $FeSO_4$, it was found that the addition of KH_2PO_4 and $FeSO_4$ resulted in a significant reduction in the rate of N loss during the composting process[92]. The addition of KH_2PO_4 resulted in lower migration rates of Cu, Zn and Pb after composting compared to before composting. Also, the addition of $FeSO_4$ retained more nitrogen. $FeSO_4$ enhanced the passivating effect of KH_2PO_4 on Pb. He et al.[93] report the effect of different types and particle sizes of biochar on greenhouse gas and ammonia emissions during composting. Compared to powdered biochar, granular biochar improved pore connectivity, favored methanogenic activity and reduced CH_4 emissions. At the same particle size, bamboo biochar had higher pore volume and better aerobic microenvironment than straw biochar. Bamboo biochar had a higher concentration of aromatic compounds and NO_3^-, which inhibits denitrifying bacteria and reduces N_2O emissions. The effects of biochar and soybean residue on nitrifying and denitrifying bacteria and on NH_3 and N_2O emissions were in vestigated[94]. The soybean residue + biochar treatment produced significantly lower peak NH_3 and N_2O emissions compared to the soybean residue treatment. This was mainly due to the suppression of NH_3 and N_2O emissions by limiting nitrification and denitrification during the composting process, reduced nitrogen losses from the composting process.

6 Outlook

As discussed above, heavy metal passivation and nitrogen retention in pig manure composting are issues that need to be urgently addressed. Especially, the mechanism of heavy metal passivation needs to be further investigated and new additives discovered. It is not difficult to see the limitations in the above studies and experiments summarized above. Here, several aspects that need to be addressed and emphasized in future research are:

(1) Most of the studies were small-scale, lacking large-scale trials for validation, and it is uncertain if the benefits of the additives tested, such as biochar for pig manure composting, will occur in large-scale practice. Theories and results from the laboratory need to be scaled up in practice to verify their effectiveness.

(2) In recent years, many studies on composting pig manure or other livestock manure have focused on compost gas emissions and nitrogen retention. There is a lack of follow-up research on the effects of compost products on improving the soil environment and enhancing soil microorganisms after intensive use. More field trials are needed to assess the potential of additive composts, such as biochar, in the agro-environment and resource use of processed manure.

(3) In many trials on compost additives the additives were pretreated under certain conditions. In real life production, it is necessary to consider the cost of additives and the complexity of the composting process, both for companies and individuals. Therefore, research needs to control the actual cost of the method or the additives while considering factors such as improving the composting effect.

(4) More sustainable materials or additives need to be developed and researched, for example, developing combinations of additives that reduce the use of physical and chemical additives, starting with microorganisms as renewable additives.

7 Conclusions

This article has reviewed research progress on heavy metal passivation and nitrogen retention in pig manure composting in recent years. There has been considerable progress in the passivation of heavy metals. For the treatment of heavy metals in compost for production applications, physical additives are still the main choice, due to their wide availability and simplicity of operation, but their heavy metal passivation is insufficient. Chemical and microbiological additives have yet to be further promoted for practical production applications. Chemical passivators are effective, technically mature and time-consuming, but as the volume of manure treated increases, so does the consumption of passivators, which not only increases costs but also makes subsequent treatment more likely to cause secondary pollution. Microbial additives for passivation of heavy metals have advantages over physical and chemical methods, and are more economic and effective, but the difficulty of extracting microorganisms from microbial passivators is a challenge that must be faced. For current production applications dealing with heavy metals in compost, the main choice is still physical additives, mainly because of the wide range of sources and simplicity of operation. Therefore, in promoting the application of new heavy metal passivators, the ease and time of use is also a points that needs more attention.

For the retention of nitrogen during composting, the main focus is reducing the emission of nitrogenous gases and the solid nitrogen content. Physical additives are generally natural or synthetic substances with a large surface area and strong adsorption properties to ammonia and ammonium ions, reducing their conversion to ammonia. Chemical additives reduce the loss of nitrogen from the compost by adding chemicals, nitrification inhibitors and inducers of guano stone crystallization. Microbial additives alter the metabolism of carbon and nitrogen by adding exogenous microorganisms, such as ammonia oxidizing archaea, thermophilic ammonia tolerant bacteria, nitrogen fixing bacteria and nitrifying *bacilli*, thus retaining more nitrogen in the compost. Currently physical, chemical and microbial additives each have their own advantages in terms of nitrogen retention in compost. Combining all three additives is a key area for future research. Compound additives can reduce the overall amount of additives used, and show great promise for both nitrogen retention and heavy metal passivation in compost.

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Compliance with ethics guidelines

Ziqi Wang, Guotao Sun, Jiamin Wang, and Gongshe Yang declare that they have no conflicts of interest or financial conflicts to disclose. This article does not contain any studies with human or animal subjects performed by any of the authors.

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