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Variations in the fate and risk analysis of amoxicillin and its degradation products during pig manure aerobic composting

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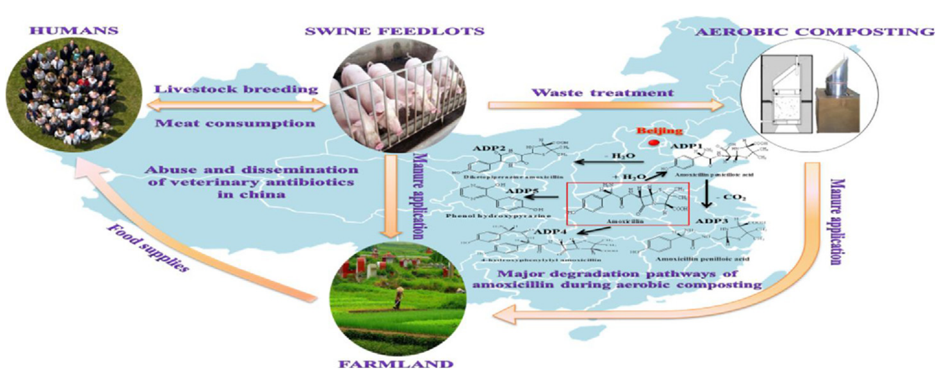
HIGHLIGHTS

- AMX is more unstable and degradable in alkaline conditions than acidic and neutral.
- The effect of pH is more significant than divalent metal ions on AMX degradation.
- Aerobic composting of pig manure can completely remove AMX and most of its DPs.
- AMX stable end product appears to be nonallergenic and nontoxic after composting.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, 100 mg/kg of amoxicillin (AMX) was added to pig manure during aerobic composting using a laboratory-scale reactor system, for better understanding the degradation of AMX and its potential risks. Qualitative and quantitative analysis of AMX and its main degradation products (DPs) were then conducted using solid phase extraction combined with high performance liquid chromatography-tandem mass spectrometry. Additionally, hydrolysis testing was performed to monitor DPs produced from AMX degradation under various controlled conditions. The results showed that AMX was unstable in compost and rapidly degraded into AMX penicilloic acid, AMX penilloic acid and AMX diketopiperazine, which all eventually degraded, leaving a structurally simple and stable end product, 3-(4-hydroxyphenyl) pyrazin-2-ol, which appeared to be nonallergenic and less toxic than DPs generated earlier in the composting process. Besides, AMX was more unstable and more apt to generate multiple DPs in alkaline conditions than in acidic or neutral conditions or in ultrapure water, while pH exerted a larger effect on AMX degradation than did divalent metal ions (Cu^{2+} and Zn^{2+}). This study provides technical and methodological support to better achieve AMX residual treatment and to evaluate the safety of AMX DPs produced in huge quantities in compost in China.

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1. Introduction

As important pharmaceuticals used for prophylaxis, therapy and to boost animal growth, antibiotics have been widely used for medical treatment and in large-scale livestock and poultry industries

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[1–7]. In recent years, China has become both the largest producer and consumer of antibiotics in the world, with a total production of 248,000 tons in 2013 and an annual usage of 84,240 tons in animals (pig: 52.2%) [6,8,9]. Consequently, 30–90% of these antibiotics and their metabolites are excreted in feces and urine after use, with eventual dissemination into the ecotope [1,2,10–13]. Of great concern, such large quantities of antibiotics and degradation products (DPs) have been demonstrated to promote horizontal gene transfer of antibiotic resistance genes (ARGs) among indigenous microorganisms and pathogens. ARGs ultimately threaten the efficacy of contemporary medicine and pose a serious threat to human health [3–7,9,10,12,13]. Amoxicillin (AMX), one of the most frequently used antibiotics, belongs to the semi-synthetic penicillin group of β-lactam antibiotics and accounts for nearly 50–70% of total antibiotic usage [14–17]. The β-lactam ring, the basic AMX structure responsible for its antibacterial activity, interferes with bacterial cell wall biosynthesis to eventually cause cell lysis [16,18,19]. Studies reported that AMX is one of the major antibiotics used in livestock [20], which is also usually used in swine and cattle breeding process [21]. The concentration of the most commonly used antibiotics have been reported to be as high as 216 mg/L of swine, beef, and poultry/turkey manures [21], while AMX can be mixed as therapeutic in pig feed at levels between 250 and 500 mg/kg [22]. As reviewed in Zhang et al. [8], AMX had the largest usage for both human and animal use among the 36 antibiotics in China; and β-lactams (including penicillin), tetracyclines and macrolides are the majority antibiotic active ingredients sold in veterinary medicinal products in the USA and UK, of which the three drugs plus fluoroquinolones consuming more in China. With a large portion of the administered doses is excreted via urine and feces into the environment, substantial β-lactam antibiotics are expected to be discharged [16,23]. Research showed that typical concentrations of antibiotics in livestock manure are between 1 and 10 mg/kg but can be as high as 200 mg/kg [21,24]. Previous studies have shown that AMX is extremely unstable and rapidly breaks down to form various DPs, which can participate in consecutive reactions under neutral or alkaline conditions [1,17,25,26]. The β-lactam ring is susceptible to multiple factors, such as abiotic and biotic processes [15,16,26,27]. However, AMX DPs are suspected of being potentially more unstable and toxic than the parent compound AMX [17,26,28]. Although the main DPs of AMX, such as AMX penicilloic acid (ADP1) and AMX diketopiperazine (ADP2), have lost the antibacterial function of their parent compound, they possess potential allergenic properties which can result from reactions between the β-lactam carbonyl group and the amino groups of proteins [29–31]. These toxic and potentially allergenic DPs likely pose an increasing risk to both human and ecosystem health as a consequence of the increase in long-term application of manure fertilizers to farmland [3–5,32,33]. Studies showed that manure is a major reservoir of Antibiotics and ARGs with a high risk of dissemination [7,33]. Zhou et al. [7] investigated ARGs in feces from 17 dairy farms in China and found that β-lactam was the top three predominant ARGs in fecal samples.

Recently, aerobic composting has been demonstrated to be a practical and effective approach to remove antibiotics from animal manure, ultimately reducing ARGs levels [24,32–34]. For example, as reviewed in Youngquist et al. [24], composting has been demonstrated to significantly reduce levels of 16 extractable antibiotics in livestock manure across 11 different studies of both bench-scale [32] and farm-scale composting systems. Research on AMX in the soil, municipal wastewater, compost and sludge fields has attracted much attention [17,25,26,35–39] in recent years. Moreover, some studies showed that although photocatalytic AMX degradation efficiency can be high [28,35], the toxicity of resulting products may not be completely eliminated, as measured by toxic DPs effects measured using the water flea *Daphnia magna* and the green alga *Pseudokirchneriella subcapitata* [28]. Meanwhile, other toxicity studies have revealed no toxicity of AMX toward green algae relative to other antibiotics and only low toxicity toward cyanobacteria (also known as cyanophytes) [17,35,38]. Lately, several groups have worked to characterize AMX DPs and evaluate their environmental prevalence [17,26], which were mainly focusing on laboratory hydrolysis experiments under controlled conditions followed by monitoring of aquatic environments. However, AMX and its DPs are rarely reported in the environment [8,40,41] especially in manure and compost, due to their instability in feces [36,37] and their quantitative difficulties owing to matrix effects [8,23]. Simultaneously, comprehensive qualitative and quantitative studies of AMX degradation characteristics and risk analysis of AMX and DPs during aerobic composting have not yet been reported.

In this study, pig manure aerobic composting experiment was carried out using a laboratory-scale reactor system to study AMX and DPs produced during aerobic composting. Concurrently, the degradation characteristics of AMX and its four important DPs, as well as their potential risks, were analyzed during aerobic composting for a variety of compost matrix characteristics and related physicochemical parameters. The results collectively provide theoretical, technical and methodological support for evaluation of AMX degradation and safety evaluation of AMX DPs in compost that is currently being produced in large quantities in China.

2. Materials and methods

2.1. Materials

Fresh pig manure (PM) was collected from a concentrated swine feedlot located in the Shunyi district of Beijing. Sampled pigs did not receive any antibiotics throughout the feeding regimen. Wheat straw (WS) was sourced from the Shangzhuang Experimental Station of China Agricultural University and was cut into lengths of 3–5 cm [42,43]. The basic physicochemical properties of composting materials are listed in Table S1.

Only chromatographic grade chemicals and reagents were used in this study. AMX (98%) was purchased from Beijing Huamaike Biological Technology Company, Ltd. (China). ADP1 (95%), ADP2 (95%), AMX penilloic acid (ADP3, 96%), 4-hydroxyphenylglyl AMX (ADP4, 96%) and 3-(4-hydroxyphenyl) pyrazine-2-ol (ADP5, 95%) were purchased from Shenzhen Standardmed, Ltd. (China). Internal standard propranolol (IS, 100%) was provided by the National Institutes for Food and Drug Control (China). Acetonitrile, methanol, formic acid and ammonium acetate were purchased from Honeywell (NJ, USA). Six standard materials were prepared in acetonitrile/water (50:50, v/v) into 1000 μg/mL standard stock. IS was formulated in acetonitrile as a 10 μg/mL internal standard stock, which was fully mixed and temporarily stored at −20 °C for up to 48 h before use.

2.2. Experimental design

2.2.1. Aerobic composting experiment

In this experiment, 5.2 kg of fresh pig manure and 100 mg/kg (dry weight) of AMX ultrapure aqueous solution (UAS) were thoroughly mixed and equilibrated for 2 h [33] followed by mixing with 0.7 kg of wheat straw in a C/N ratio of 20:1 and obtained an initial moisture content (MC) to about 65%. Next, all materials were thoroughly mixed and filled into a 15-L cylindrical composting reactor (0.40 m height × 0.25 m internal diameter) as Fig. S1, which was described in our previous study [42]. High concentration (100 mg/kg) of AMX was selected according to previous studies [25,44] and with respect to probable fast degradation of AMX and some DPs covering a worst-case scenario. The intermittent ventilation mode [43] was used during the process, with a time interval of

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