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Biodegradation of nonylphenol during aerobic composting of sewage sludge under two intermittent aeration treatments in a full-scale plant

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ABSTRACT

The urbanization and industrialization of cities around the coastal region of the Bohai Sea have produced large amounts of sewage sludge from sewage treatment plants. Research on the biodegradation of nonylphenol (NP) and the influencing factors of such biodegradation during sewage sludge composting is important to control pollution caused by land application of sewage sludge. The present study investigated the effect of aeration on NP biodegradation and the microbe community during aerobic composting under two intermittent aeration treatments in a full-scale plant of sewage sludge, sawdust, and returned compost at a ratio of 6:3:1. The results showed that 65% of NP was biodegraded and that *Bacillus* was the dominant bacterial species in the mesophilic phase. The amount of NP biodegraded in the mesophilic phase was 68.3%, which accounted for 64.6% of the total amount of biodegraded NP. The amount of NP biodegraded under high-volume aeration was 19.6% higher than that under low-volume aeration. *Bacillus* was dominant for 60.9% of the composting period under high-volume aeration, compared to 22.7% dominance under low-volume aeration. In the thermophilic phase, high-volume aeration promoted the biodegradation of NP and *Bacillus* remained the dominant bacterial species. In the cooling and stable phases, the contents of NP underwent insignificant change while different dominant bacteria were observed in the two treatments. NP was mostly biodegraded by *Bacillus*, and the rate of biodegradation was significantly correlated with the abundance of *Bacillus* ($r = 0.63$, $p < 0.05$). Under aeration, *Bacillus* remained the dominant bacteria, especially in the thermal phase; this phenomenon possibly increased the biodegradation efficiency of NP. High-volume aeration accelerated the activity and prolonged the survival of *Bacillus*. The risk of organic pollution could be decreased prior to sewage sludge reuse in soil by adjusting the ventilation strategies of aerobic compost measurements.

1. Introduction

The Bohai Sea, also known as the Bohai Gulf, is the innermost gulf of the Yellow Sea and Korea Bay on the coast of Northeastern and North China. It is approximately 78,000 km^2 in area, and its proximity to Beijing, the capital of China, makes it one of the busiest seaways in the world. Some of the rivers entering the gulf include the Yellow, Hai, Liao, and Luan Rivers. There are also important oil reserves in the vicinity of the gulf, including the Shengli Field and Liaohe Oilfield. The coastal region of the Bohai Sea is the largest pool of modern industry and is the heavy and chemical industrial base of China. The coastal region of the Bohai Sea shows large developmental potential as the center of the international economy shifts to the Asia-Pacific region; however, a large amount of wastewater, exhaust gas, and waste residue was released without treatment and reached the national standard beginning in the 1980s. Based on reporting from Economic Information Daily on August 10, 2015, annually, approximately 28×10^4 t waste water and 70×10^4 t solid waste flowed into the Bohai Sea through the 57

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rivers, which accounts for 50% of China's total marine pollutants (Bai, 2015). Pollutants have made environmental pollution particularly serious in the Bohai Sea, which has consequently harmed the development of the Bohai economic circle (Chen et al., 2017; Zhang et al., 2017; Jiang et al., 2018).

In recent years, the wastewater treatment industry has developed rapidly in the coastal region of the Bohai Sea. By 2016, the rate of municipal wastewater treatment (MWT) was 90.58%, 92.08%, 93.61%, 95.37% and 96.21% in Beijing, Tianjin, Liaoning, Hebei and Shandong, respectively. Sewage sludge is the end product of sewage treatment. The improvement in sewage treatment capacity has gradually increased the production of sludge. Therefore, problems regarding the disposal of sewage sludge must be urgently addressed. Despite the risks of heavy metals, pathogens, organic pollutants, and other pollutants that sewage sludge brings, this method can be applied similarly to soil fertilizer after proper treatment because it can provide plant nutrients and humus. In the EU-27 countries, 53% of urban sludge is treated through composting (Kelessidis and Stasinakis, 2012). Thus, the principle of harmless and resource-oriented treatment and disposal of sewage sludge can and should be followed. Composting is a suitable method for sewage sludge treatment. The heat produced during composting can kill most of the pathogenic bacteria, and sewage sludge can be effectively stabilized after composting. Compost can also be used to produce organic fertilizer.

Sewage sludge composting is not designed to treat organic pollutants, as evidenced by the lack of early warning and monitoring mechanisms for organic pollutants in sewage sludge treatment plants; thus, the presence of organic pollutants in sludge may pose a risk to the environment (Luo et al., 2014). Nonylphenol (NP) is an estrogen and mainly originates from the biodegradation of NP ethoxylates (NPEs), which are widely used in industrial products (Bai et al., 2017; Jambor et al., 2017). During sewage treatment, NPEs easily biodegrade into NP, which then contaminates sewage sludge (Samaras et al., 2013; Esteban et al., 2016; Murdoch and Sanin, 2016). NP can be detected in sewage sludge at a concentration ranging from $1 \times 10^3\ \mu g\ kg^{-1}$ to $1 \times 10^6\ \mu g\ kg^{-1}$ (Stasinakis, 2012). Meng et al. (2016) conducted a systematic review and a meta-analysis of organic contaminants in Chinese sewage sludge based on the literature published over the past 30 years and found the amount of 4-nonylphenol to be $3.14 \times 10^3\ \mu g\ kg^{-1}$. NP is also an endocrine-disrupting chemical that may enter the human body through the food chain and affect the liver and reproductive organs once released into the environment (Durán and Beiras, 2016; Esteban et al., 2016). Therefore, the NP content of sewage sludge must be controlled.

Multiple studies on the transport, sorption, and biodegradation of NP in soil and river sediments have been conducted (Chen et al., 2011; Mao et al., 2012; Liu et al., 2014; Lu and Gan, 2014; Lyons et al., 2014; Wang et al., 2014, 2015; Zhang et al., 2016), whereas studies on the biodegradation of NP during sewage sludge composting are limited. A few studies have found that NP in soil and river sediments can be biodegraded by various microorganisms, such as *Pseudomonas* (Watanabe et al., 2012; Xie et al., 2015), *Sphingomonas* (Weert et al., 2010; Collado et al., 2013), *Acidovorax* (Watanabe et al., 2012), and *Bacillus* (Hsu et al., 2013). The oxygen content, temperature, and organic matter content of windrow during sewage sludge composting differ, however, from those of soil and river sediments. The effect of these microorganisms on the sewage sludge composting process remains unclear. Hao et al. (2008) studied the bio-degradation pathways of NP in wastewater treatment, and the results showed that the long branches in nonyl of NP are first degraded into short methyl branches, which creates a series of short chain alkyl phenols with different branch structures. Given that the biodegradation of NP always starts from the oxygen-adding open loop of benzene, the amount of oxygen exerts a limited effect on such biodegradation. Studies have shown that NP is completely biodegraded under the effect of aeration; in contrast, the rate of biodegradation in the absence of aeration is only 60% under laboratory scale (Abargues et al., 2013; Lu et al., 2015). The biodegradation efficiencies of laboratory scale are insufficient to satisfy the demand of environmental governance (Bai et al., 2017). During sewage sludge composting, forced aeration can increase the oxygen content of a material and the duration of aerobic conditions. In the absence of forced aeration or under a low amount of aeration, the oxygen content of a material is depleted completely, thereby resulting in an anaerobic environment (Shen et al., 2011). Therefore, aeration may be an important affecting factor of the biodegradation of NP during sludge composting (Zheng et al., 2015).

Research on the succession of the microbial community structure and the biodegradation kinetics of NP in sludge compost can reveal which microorganisms are involved in such biodegradation. Measures can then be taken to promote the biodegradation of NP during sewage sludge composting and consequently reduce the risks posed by the land application of sludge compost products. Full-scale composting for aeration optimization and development, particularly an aeration method for sewage sludge composting with improved efficiency for the biodegradation of NP, should be developed. Thus, the objective of this work is twofold. First, the dynamic changes in the biodegradation of NP during aerobic composting of sewage sludge under two intermittent aeration treatments in a full-scale plant are assessed. The abundance of microorganisms is studied by high-throughput sequencing in addition to the composting process. Second, the relationship between the microbe community and NP biodegradation is investigated by comparing the effect on the biodegradation of NP and the abundance of microorganisms by using different aeration treatments.

2. Materials and methods

2.1. Instruments and reagents

Dichloromethane, methanol, and acetone (chromatographically pure) were purchased from Sigma–Aldrich (St. Louis, USA). Ethanol and acetone (analytically pure) were purchased from the China National Pharmaceutical Group (Beijing, China). Standard NP solution (2500 mg/L, dissolved in methanol:acetone = 1:1 mixture) was purchased from AccuStandard (New Haven, USA). DNA extraction kits (PowerSoil@ DNA Isolation Kit) were purchased from MO BIO Laboratories (Carlsbad, Canada). Ultrapure water, 10 × reaction buffer, PCR primers (5′ and 3′ primers), dNTPs, and pfuTaq enzyme were used in DNA PCR amplification. A solid-phase extraction column (Waters Sep-Pak tC18, 6 mL, 500 mg, 37–55 μm) and high-performance liquid chromatography (HPLC) column (C18 symmetry, 5 μm, 4.6 mm × 250 mm) were purchased from Waters (Milford, USA).

2.2. Sludge, conditioner, and returning compost

Sludge was collected from the Qinhuangdao Sludge Treatment Plant in Hebei Province, China. The water content of the sludge was approximately 80% after mechanical dewatering. The sawdust was obtained from a local timber processing plant; the equivalent diameter of the sawdust was approximately 1.0 cm, and the moisture content was 4.0–5.0%. Returning compost was obtained from matured compost and then directly transported from the composting workshop to the mixing workshop. The initial moisture content of the windrow was approximately 60%, and the C/N ratio was approximately 25:1. On the basis of the moisture content and