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Evaluation of total greenhouse gas emissions during sewage sludge composting by the different dicyandiamide added forms: Mixing, surface broadcasting, and their combination

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ABSTRACT

The aim of this work was to compare the impact of different adding forms of dicyandiamide (DCD) on NH_3 and greenhouse gas (GHG) emissions during sewage sludge (SS) composting. Four treatments were set up using SS mixed with sawdust, to which DCD was then added by mixing (M), surface broadcasting (B), and a combination of the two (M+B). The treatment without DCD applied was used as the control. The results indicate that the addition of DCD slightly inhibited the organic matter (OM) degradation, but that it had no significant effect on CO_2 emission. The surface mulching of DCD has no significant effect on NH_3, N_2O, and CH_4 emissions. The mixing addition of DCD significantly increased the NH_3 emission by 32.5% compared to that of the control. The N_2O emission for the M and M+B treatments significantly decreased by 35.1% and 51.8%, respectively. The CH_4 emission for the M and M+B treatments decreased by 33.9% and 31.8%, respectively. In addition, the total GHG emissions for the M and M+B treatments were significantly reduced by 16.7–25.7% ($P < 0.05$) compared to those of the control. Therefore, to reduce the total GHG emissions of the SS composting process, the addition of DCD by a combination of mixing and surface mulching is strongly recommended as a highly efficient solution.

1. Introduction

Forty million tons of sewage sludge (SS) (with 80% moisture content) was produced in China in 2016 (NBSC, 2016); this amount is increasing due to expedited urbanization and the increasing capacity of municipal wastewater treatment facilities (Jain et al., 2018; Wang et al., 2018a). The improper management of SS will cause secondary pollution such as pathogenic microbes, organic micro-pollutants, and toxic heavy metals; therefore, sustainable and ecofriendly SS management is urgently required (Sadef et al., 2016).

Composting is a simple and low-cost method for converting SS into a safe and stable product that could be used as organic fertilizer or as a soil conditioner for farming (Meng et al., 2018; Zhang et al., 2018). However, harmful gases, such as ammonia (NH_3), nitrous oxide (N_2O), and methane (CH_4), which are emitted due to the mismanagement of the composting process, not only reduces the agronomic value of compost as a soil fertilizer or amendment but also decreases the environmental benefits of composting (Santos et al., 2018; Wang et al., 2018c). As global warming worsens and the greenhouse effect intensifies, emissions of N_2O and CH_4 during the composting process have attracted the interest of researchers (Awasthi et al., 2018; Jain et al., 2018). Additionally, CH_4 and N_2O have 30 and 210 times higher impact on global warming than that of carbon dioxide (CO_2) emissions (IPCC, 2007), respectively. It was reported that approximately 0.1–9.9% of nitrogen (N) in compost material was lost as N_2O via nitrification or denitrification during composting (Sanchez et al., 2015; Tsutsui et al., 2015), and CH_4 emissions amounted to 0.8–14% of the initial carbon (C) (Ermolaev et al., 2014; Luo et al., 2013). With respect to the aforementioned issues, in the last few decades, substantial research has been conducted on SS composting, focusing in particular on employing different additives to reduce the emissions of greenhouse gases (Awasthi et al., 2017b; Sanchez et al., 2015).

Dicyandiamide (DCD, $C_2H_4N_4$) is a well-known nitrification inhibitor (NI) that was proven to be effective in reducing N_2O emission from cropland (Cahalan et al., 2015; Kelliher et al.,

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2008). In one hand, DCD could reduce the *amoA* gene in ammonia oxidizing bacteria and thus slow nitrification, resulting in the decrease of N_2O production and emission rates. In the other hand, it also reduced nitrate concentrations in soil, which decreases the potential for N_2O production from denitrification (Akiyama et al., 2010; Dai et al., 2013). It has been documented that N_2O emissions were reduced by 17–90% when using DCD in comparison to that using conventional urea (Dobbie and Smith, 2003; Jiang et al., 2017b; Kelliher et al., 2008). Considering the obvious inhibition on N_2O emissions, several researchers have conducted studies on the influence of DCD on N_2O emissions and N immobilization during composting. Jiang et al. (2016) reported that DCD significantly inhibits nitrification, which could reduce the N_2O emission by 76.1–77.6% for composting pig faeces and corn stalks. In addition, Luo et al. (2013) found that when DCD was added to pig manure composting, the total N_2O losses were reduced by 33–35% compared to those of the control.

During composting, N_2O can be produced through both nitrification and de-nitrification processes (Jiang et al., 2016). Nitrification involves the initial transformation of ammonia to nitrite by different genera of ammonia-oxidizing bacteria, whereas during denitrification, nitrite is reduced to NO by nitrite reductase, which is further reduced by nitric oxide reductase to N_2O (Moënne-Loccoz and Fee, 2010; Sanchez et al., 2015). The rates of nitrification and denitrification are affected by the raw materials' C/N ratio, water content, NH_4^+-N, pH and turning frequency (Kebreab et al., 2006; Sanchez et al., 2015). In addition, the oxygen concentration is one of the main factors that control N_2O emissions because nitrification occurs in aerobic conditions, while denitrification occurs in anaerobic conditions. Due to the heterogeneity of composting materials, aerobic and anaerobic conditions can exist simultaneously in the composting pile, since different oxygen concentration gradients are created along the pile (Beckfriis et al., 2000; Hao et al., 2004). Relatively speaking, denitrification mainly occurred in the inner part of the pile, where the O_2 was quickly consumed by microorganisms, whereas nitrification required oxygen concentrations in the range of 1–10%, so the aerobic pile surface could satisfy this condition (Béline et al., 1999).

Since there is previous literature pertaining to the study of DCD during composting, in those studies, DCD was all mixed with raw materials (Luo et al., 2013; Jiang et al., 2016). However, it is likely that the mixing addition of DCD is only significant in inhibiting denitrification in the inner pile but that does not inhibit the nitrification on the aerobic pile surface. In addition, few studies have been performed to evaluate surface mulching or the combination of mixing and surface mulching of DCD on the composting process. Thus, the purpose of the present study is to compare the impact of DCD by mixing, surface mulching, and their combination on NH_3 and emissions of greenhouse gases (CO_2, N_2O, and CH_4) during SS composting and to determine the most effective adding form for nitrogen conservation and total greenhouse gas reduction.

2. Materials and methods

2.1. Raw materials and additive forms

Fresh dewatered SS and sawdust (SD) were used as raw materials in this research. SS was collected from a local municipal wastewater treatment plant (Xinxiang, Henan, China), and was the mixture of the primary and secondary sludge. SD, the residual material remaining after working timber, was used as the bulking agent and was obtained from a local furniture factory. The SD was chopped into 1.0 cm pieces, and applied to adjust the moisture content (~55–65%) and C/N ratio (~25) of the initial compost. The physicochemical characteristics of raw materials are presented in Table 1. The DCD used in this study was a chemically pure reagent grade purchased from Wuxi Yatai United Chemical Industry Co. Ltd., China.

2.2. Experimental design and composting process

To simulate the forced aeration system, the composting experiments were conducted in a series of 14 L cylindrical polyethylene reactors, with an inner diameter and height of 200 mm and 450 mm, respectively. The aeration rate was fixed at 0.40 L/min and programmed with intermittent aeration (start 30 min, stop 30 min) by air pumps. To achieve the appropriate moisture content and C/N ratio, initially, 3.6 kg of fresh SS was mixed with 0.9 kg SD; then, the DCD was added to each treatment in three adding patterns. In Treatment 1 (M), 6 g (0.4% dry basis) of DCD was mixed with the raw materials at the beginning of composting. In Treatment 2 (B), 6 g of DCD was dispersed uniformly on the surface of the raw materials at the beginning of composting. In Treatment 3 (M+B), 3 g of DCD was mixed with the raw materials, and half of the DCD by mass was dispersed uniformly on the surface of the raw materials at the beginning of composting. When mixed directly with the compost pile, e.g., 6 g in the M treatment and 3 g in the M+B treatment, the weight of DCD was very low compared to that of the pile (4.5 kg). Therefore, the DCD was first dissolved in 100 mL of deionized water and then mixed with the compost pile to ensure that the mixture was homogenous. In comparison, for surface dispersion, DCD was directly sprayed on the surface without the pre-dissolving DCD. This is because, the diameter of the reaction column is 200 mm, so spray in powder form can achieve even dispersion of the DCD on the surface. Additionally, when dissolved in water, the solution might sink to the bottom of the reaction column and affect the mixing pattern. The treatment without additives was taken as the control (CK) and mixed with 100 mL of deionized water for the purpose of comparison.

Four piles of mixtures were filled in four identical cylindrical reactors, and the composting experiments were carried out over 22 days. To maintain the heat of the composting pile during composting, the four reactors were placed in a water-bath with temperatures set to 1.0–2.0 °C below that of the lowest temperature of the four treatments (Mason and Milke, 2005). The temperature of the compost and the water bath were monitored twice daily and recorded.

2.3. Sampling and physicochemical analysis

Homogeneous compost samples were collected after 0, 3, 6, 9, 12, 16 and 22 days. The representative sample was divided into two parts. One part was immediately stored at 4 °C until analysis, and the other part was air-dried, passed through a 0.25 mm sieve and stored in a desiccator for further analysis. The fresh samples were used to determine pH, electrical conductivity (EC), seed germination index (GI), NH_4^+-N, and NO_3^--N, according to laboratory procedures (Jiang et al., 2018; Jiang et al., 2017a). The GI was determined using Chinese pakchoi (*Brassica campestris* L. ssp. *chinensis Makino*) seeds, and the measurement was repeated three times. For each determination, ten seeds were evenly scattered on filter paper and moistened with 5 mL of the compost extract and then incubated at 25 °C for 48 h. In this set-up, distilled water was used as the control. To determine the dissolved organic carbon (DOC), fresh samples were extracted at 25 ○C with ultrapure water with a ratio of 1:10 (w/w) using a shaker running at 150 rpm for 1 h. This step was followed by centrifugation at 12000 rpm for 10 min and filtration through a 0.45 μm membrane (HainingYatai Pharmaceutical Machinery Co., Ltd.). Finally, the sample was measured by an automated TOC analyser (Shimadzu TOC-V). The