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Characteristics and kinetics of ammonia and N_2O emissions of aged refuse irrigated from landfill leachate

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ABSTRACT

This is the first attempt to report the gaseous nitrogen emissions from landfill leachate filtration methods by irrigating the aged refuse. A first-order reaction model was a good fit for the increase in ammonia emissions from aged refuse, clay and sandy soil incubated for 120 h after adding the leachate-N solution. The emissions of ammonia and N_2O by the three experimental materials fit well to first-order and zero-order models, respectively. The maximum ammonia emission from aged refuse was approximately 1.17 mg NH_4^+—N kg^{-1} d.w. and the calculated emission factor was 1.95‰, which was 3.76 and 2.67 times lower than that of sandy and clay soils, respectively. The tendencies of NH_4^+—N nitrification and NO_3^-—N generations fit well to the zero-order reaction model and the net nitrification rate by the aged refuse was 1.30 ($p < 0.05$) and 1.71 ($p < 0.05$) times that of clay soil and sandy soil, respectively. At the same time, the net NO_4^-—N generation rate by the aged refuse was 1.56 ($p < 0.05$) and 2.33 ($p < 0.05$) times that of clay soil and sandy soil, respectively. The quantity of nitrogen emitted by aged refuse as N_2O was 2.46 times greater than that emitted as ammonia. The emission factor for N_2O from aged refuse was 8.28 ($p < 0.05$) and 16.11 ($p < 0.05$) times greater than that of clay and sandy soils, respectively. For the leachate irrigation, N_2O emissions should be of greater concern than ammonia emissions.

1. Introduction

Nitrous oxide (N_2O) is a greenhouse gas accounting for 7.9% of observed global warming (Zhang et al., 2009). It is estimated to have 298 times the radiative force per molecule of CO_2, partly due to its prolonged residence time (IPCC, 2007). N_2O is produced predominantly in soils by microbial processes as a byproduct of nitrification and an intermediate product of denitrification (Inamori et al., 2008; Ruser et al., 2006). Enhanced N_2O emissions are largely stimulated by fertilizers (i.e., the application of mineral fertilizers, poultry manure, sewage sludge, and the drying–wetting cycles caused by irrigation or precipitation) (Inubushi et al., 2000; Kravchenko et al., 2002; Murry et al., 2004; Priemé and Christensen, 2001; Towprayoon et al., 2005).

Landfill leachate is typically characterized by a high loading of ammonium to thousands of milligrams per liter. It is a cost-effective operation for leachate treatment by using aged refuse and/or soil as filter materials (Li et al., 2010; Xie et al., 2012; Zhang et al., 2010a, 2009). Aged refuse – also termed as mineralized refuse or waste soil – contains a wide spectrum and large quantity of microorganisms that have a strong nitrification capability for the ammonia nitrogen (NH_3–N) present in some wastewaters, such as leachate, coke-plant wastewater and livestock and domestic wastewater (Xie et al., 2012; Zhang et al., 2012; Zhao et al., 2002; Zhu et al., 2012). These documented data were very focused on how to elevate the removal efficiency of the ammonia nitrogen from wastewater through the microbial nitrification and denitrification processes. Aged refuse from irrigated landfill leachate should be favorable for the microbial production of N_2O because aged refuse has been reported to be rich in organic matter and microbial activity, and landfill leachate can also provide soluble substrates of both carbon and nitrogen (Zhang et al., 2010a; Zhang et al., 2009). However, there has been far less information yielded from the study on the characteristics and kinetics of the emissions of N_2O by aged refuse with strong nitrification capacities.

Moreover, with a high loading of ammonium to thousands of milligrams per liter in landfill leachate, a high potential risk of ammonia (NH_3) emissions should exist within the irrigated aged refuse or soil. Ammonia emissions contribute to the acidification and eutrophication of soils and waters in nitrogen-limited ecosystems (Klaasen, 1994; Sutton et al., 1998) and are also a secondary source of N_2O emissions after deposition in the soil. In addition, NH_3 is a key precursor to neutralize H_2SO_4 and HNO_3 in the air and from $(NH_4)_2SO_4$, NH_4HSO_4 and NH_4NO_3, which contribute to reduced visibility, regional haze and health impacts associated

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with fine particulate matter (Zhang et al., 2010b). A host of evidence indicates that agricultural sources (i.e., volatilization from livestock manure and mineral fertilizer application) contribute to the majority of NH_3 emissions and comprise approximately 80–90% of the total anthropogenic NH_3 emissions (Liu et al., 2011; Sommer and Hutchings, 2001). The cumulative amount of ammonia emissions from agriculture fields after organic manure and chemical fertilizer application within a short period accounted for more than 30% of the total nitrogen applied (Huijsmans et al., 2003; Lu et al., 2010; Sommer and Hutchings, 2001). When wastewater treatment uses filtration methods, the wastewater should be considered similar to liquid fertilizer due to the high NH_4^+—N loadings. To best of our knowledge, there is scarce information on ammonia emissions from wastewater treatment in high NH_4^+—N loadings when using the irrigation method.

For the emissions of ammonia as well as N_2O and CO_2, the soil physicochemical properties are the intrinsic factors (Inubushi et al., 2000; Murray et al., 2004; Priemé and Christensen, 2001; Van der Weerden and Jarvis, 1997; Zhang et al., 2010a). Kravchenko et al. (2002) reported more than 6.0 times difference between the N_2O fluxes of loam and loamy sand soils amended with the same amount of $(NH_4)_2SO_4$ solution. Besides N_2O emissions, CO_2 is mainly produced by aerobic decomposition of the organic matter, i.e., microbial respiration, which is also influenced by the physicochemical properties of the soil. Murray et al. (2004) also documented that a factor of more than one order of magnitude difference was generated between the CO_2 emissions from two grassland soils amended with the same organic carbon solution. Thus, a potential solution existed by selecting the specified physicochemical properties of aged refuse or of soil for the mitigation of greenhouse gas emissions.

To understand the consequences of landfill leachate irrigated aged refuse and of soil on the emissions of ammonia and N_2O as well as of CO_2, batch incubation was conducted to study the characteristics and kinetics of aged refuse and two different soils to compare them after the addition of leachate. The conversion between the NH_4^+—N and NO_3^-—N contents in the aged refuse and the soil samples were also evaluated.

2. Materials and methods

2.1. Aged refuse and soil samples

The landfill site selected for sampling is situated at the foot of Jiaozishan Mountain, which is located in a northern suburb of Nanjing City (31°59′N, 118°54′E) in eastern China. The mean annual temperature was 16.7 °C, and the annual precipitation amounted to 1200 mm in 2010. The aged refuse used in this study was excavated in 2010 from a single enclosed chamber that had been covered for 14 years. The domestic refuse has been dumped approximately 1000 t/d currently. The landfilled domestic refuse was sampled according to the Sampling and Analysis Methods for Domestic Waste (GB CJ/T 313-2009) by the first-degree sample ranged from 3 to 200 mm. The composition of the current landfilled refuse was analyzed as wet weights: food waste (60%); plastics (20%); organic matter, such as bamboo, wood, paper and textiles (15%); and inorganic matter, such as stone, sand, metals and other materials (5%). Compared with the landfilled refuse 10 years ago, the food waste was increased by 10% and stone and other materials were decreased by 10% accordingly.

After removing the cover soil, the excavated aged refuse has been collected from the inside of the landfilled pile. The excavated refuse was air-dried for 48 h in the laboratory, and the larger-sized non-degradable matter (e.g., stones, glass bottles, plastic film, plastic bags, and rubber) were removed by manual sorting (Zhang et al., 2012; Zhu et al., 2012). Larger particles were removed and then the finer portion of the waste was crushed and passed through a 2 mm sieve. The two different types of soil sample – alkaline clay soil and acidic sandy soil – for the comparative study were collected from a rice field and a landscape field in the Taihu Lake basin, Jiangsu Province, China, respectively (31°29′N, 119°59′E). All experimental materials were sieved through 2 mm screens to remove the large particulate matter, and the physicochemical properties of aged refuse and the soils are given in Table 1. The much higher NO_3 content in age refuse might result from the oxidation of the accumulated NH_4^+ after the excavation.

2.2. Incubation assays

Nitrification experiments, ammonia emissions, N_2O and CO_2 emissions experiments were carried out in the laboratory as described by Cheng et al. (2004), with some modifications (Fig 1). Samples of 100 g (oven-dried weight) from each soil and aged refuse sample were put into three 250-mL wide-mouth glass bottles, and sealed tightly with a rubber stopper and screw top according to Zhang et al. (2010a). Moreover, each 100 g of soil sample was weighed and distributed among the 250-mL glass bottles in triplicate. The soil and the aged refuse samples were incubated in the laboratory at a constant air temperature of 25 ± 0.5 °C, which is the optimal temperature for most soil microbial processes (Sánchez-Martín et al., 2008). The landfill leachate (approximately 8.0 mL) and/or distilled water were then added to each bottle with a mini-pipette to adjust the soil sample to the moisture content of 18% or 25% (oven-dry weight), which were linked to 46% and 70% water-filled pore space (wfps) for N_2O and CO_2 emissions experiments, respectively. During the incubation, lost water was replaced every 2 days by distilled water through mini-pipette.

The loading of 200 mg N kg^{-1} d.w. was achieved by adding $(NH_4)_2SO_4$ into the landfill leachate, which corresponds to the recommended fertilizer level (Inubushi et al., 2000). The landfill

Table 1
Physicochemical properties of the aged refuse and the soils (sample no = 3).

Physicochemical properties[a]	Aged refuse	Clay soil	Sandy soil
pH ($CaCl_2$)	7.47 ± 0.25	8.05 ± 0.16	6.75 ± 0.19
Cation exchange capacity (cmol kg^{-1})	70.89 ± 1.90	53.21 ± 1.81	41.71 ± 1.34
Specific surface area ($m^2\ g^{-1}$)[b]	0.74 ± 0.10	0.80 ± 0.17	0.77 ± 0.12
Organic matter (%)	11.23 ± 1.94	2.51 ± 0.52	1.81 ± 0.38
NH_4^+—N content (mg kg^{-1})	49.48 ± 0.24	13.65 ± 1.21	10.78 ± 0.70
NO_3^-—N content (mg kg^{-1})	32.31 ± 0.70	17.42 ± 2.31	15.25 ± 0.03
Total nitrogen (%)	0.136 ± 0.017	0.066 ± 0.009	0.043 ± 0.003
Clay (in mass%)	35.3	43.5	23.4
Silt (in mass%)	33.7	32.1	30.9
Sandy (in mass%)	31.0	24.4	45.7

[a] The physicochemical properties of the soil and aged refuse were determined according to the Chinese Soil Society Guidelines (Lu, 2000), In Section of 2.3.
[b] Through the 2 mm sieve. The specific surface area of the soil and aged refuse has been determined by BET methods (Lu, 2000).