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Minimizing N_2O fluxes from full-scale municipal solid waste landfill with properly selected cover soil

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Abstract

Municipal solid waste landfills emit nitrous oxide (N_2O) gas. Assuming that the soil cover is the primary N_2O source from landfills, this study tested, during a four-year project, the hypothesis that the proper use of chosen soils with fine texture minimizes N_2O emissions. A full-scale sanitary landfill, a full-scale bioreactor landfill and a cell planted with *Nerium indicum* or *Festuca arundinacea* Schreb, at the Hangzhou Tianziling landfill in Hangzhou City were the test sites. The N_2O emission rates from all test sites were considerably lower than those reported in the published reports. Specifically, the N_2O emission rate was dependent on soil water content and nitrate concentrations in the cover soil. The effects of leachate recirculation and irrigation were minimal. Properly chosen cover soils applied to the landfills reduced N_2O flux.

Key words: municipal solid waste landfill; N_2O flux; cover soil; leachate; nitrification/denitrification; environmental factors

Introduction

Nitrous oxide (N_2O) is a greenhouse gas with high ozone depletion potential (Chen *et al*., 2000; Baggs *et al*., 2002). Atmospheric N_2O has 310 times the radiative force per molecule relative to CO_2, partly because of its long atmospheric lifetime of 127 years (Kiese and Butterbach-Bahl, 2002). Soil produce N_2O via microbial nitrification and denitrification processes (Holtan-Hartwig *et al*., 2002). Fluxes of N_2O from different ecosystems, such as tilled fields, pastures, wetlands and forests were documented (Ghosh *et al*., 2003; Cheng *et al*., 2004; Maljanen *et al*., 2004; Wang *et al*., 2005).

Municipal solid waste (MSW) landfills have massive amounts of substrates containing nitrogen, which are potential sources of N_2O (He *et al*., 2000; Hui *et al*., 2003). Rinne *et al*. (2005) indicated that N_2O fluxes from the Ammalssuo landfill were higher than those from northern European agricultural soils and boreal forests by 1–2 orders of magnitude. A field survey of the Guangzhou Likang landfill implied that N_2O from leachate-contaminated soil was at minimum 2–5 times higher than that from the Illinois landfill (Lee *et al*., 2002). The highest N_2O fluxes recorded were in the Sweden Hőgbytorp landfill, 2–3 orders of magnitude higher than those at all other landfills measured (Bőrjesson and Svensson, 1997). The high N_2O fluxes from these landfills resulted from high organic matter concentrations in fertile cover soil or introduced by leachate or sewage sludge (Bőrjesson and Svensson, 1997; Inubsushi *et al*., 2000; Hui *et al*., 2003).

The bioreactor landfill approach is characterized by leachate recirculation or liquid injection in MSW landfills (Berge *et al*., 2006; Khire and Mukherjee, 2007). Vegetation added to soil increases N_2O emissions (Arnold *et al*., 2005; Dick *et al*., 2006); whereas leachate is commonly applied to vegetated or landscaped cover at landfills (Tyrrel *et al*., 2002; Duggan, 2005). Emissions from bioreactor landfill or from vegetated cover soil with leachate irrigation have not been reported.

If cover soil is responsible for most N_2O emissions from MSW landfills, as in other ecosystems, selecting a proper soil should minimize the amount of this greenhouse gas emitted regardless of landfill age or leachate recirculation. To test this hypothesis, this study conducted a four-year project at the Hangzhou Tianziling landfill in Hangzhou City of China, utilizing selected sandy soils with low nitrogen content and fine texture as the final cover. The N_2O emission rates from the cover soils at the sanitary landfill site were measured. This study was also applied to a full-scale bioreactor landfill site that received the same MSW as the sanitary landfill. This study proved that properly selected cover soils exhibited low potential for N_2O emissions even when leachate was recycled to landfilled waste body by the bioreactor landfill or applied to irrigate the vegetated cover soil for volume reduction.

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1 Materials and methods

1.1 Study site

The test landfill is an MSW landfill at a valley in northern Hangzhou City. The MSW lists compositions were loaded at 2000 t/d to the sanitary landfill site at the test landfill (Table 1). The sanitary landfill was equipped with bottom liner using compacted clay, also with leachate collection pipes and active landfill gas (LFG) collection system (the flow rate was about 20,000 m^3/d). The designed elevation of the landfill was from 48 to 180 m (based on Yellow Sea level, hereafter likewise). The landfilling operation was carried out by multi-lift method; from 1999 to 2006 the operation lifts were between 115–177.5 m elevation, with 12.5 m for each lift (Fig.1a). The slope face of each lift is 500–600 m width with typical slope 3:1. The lifts in the range 115–165 m elevations have been covered finally with 0.5–1.0 m the selected soil and sparse vegetation on the cover surfaces. At present, the waste is being filled at the lift 165–177.5 m.

Table 1 Compositions of MSW produced in Hangzhou City

Compositions	Year		
	1999–2001	2002–2004	2005–2006
Food waste (%, w/w)	52.0±0.99	60.1±8.11	59.3±15.5
Papers (%, w/w)	12.6±0.24	9.97±1.49	7.50±3.86
Texture (%, w/w)	1.21±0.37	1.84±0.80	2.19±1.85
Bamboo/woods (%, w/w)	1.53±1.09	2.55±1.71	4.16±4.07
Plastics (%, w/w)	12.6±2.29	14.3±1.44	21.9±11.6
Metals (%, w/w)	0.84±0.06	1.12±0.34	0.77±0.21
Glass (%, w/w)	0.87±0.31	1.96±0.38	1.05±0.52
Stone and others (%, w/w)	18.4±1.98	8.35±5.04	3.09±0.80

Sample number n = 3.

The amount of landfilled food waste increased and accounted for approximately 60% of the total landfilled MSW since 1999, resulting in a high potential for conversion of nitrogen-containing organic matters to N_2O. The tested cover surface where N_2O sampling points were set is located at the slope face of lifts 115–177.5 m at the landfill (Fig.1a). This study selected a cover soil as the final cover (Table 2). The organic content in these cover soils was typically low.

The bioreactor landfill was located adjacent to the sanitary landfill site in a canyon at the 58–85 m elevation. The bioreactor landfill was constructed with bottom liners using high density polyethylene (HDPE) membrane and compacted clay, leachate collection pipes, LFG collection wells and leachate recirculation system.

In 2004–2005, approximately 121,000 t of MSW was landfilled in the bioreactor landfill with leachate recirculated back to the MSW layer at 10–35 m^3/d. The final cover of the bioreactor landfill was completed in January 2006, with top soils (0.6–0.8 m) same as those in the sanitary landfill. N_2O sampling points were set on Platform 58 m and Platform 85 m, as well as the slope face between them (Fig.1b). The area of the bioreactor landfill occupied approximately 20000 m^2. During the sampling period, the landfill gas (50%–60% v/v of CH_4) was extracted by pump at a rate of approximately 500 m^3/d.

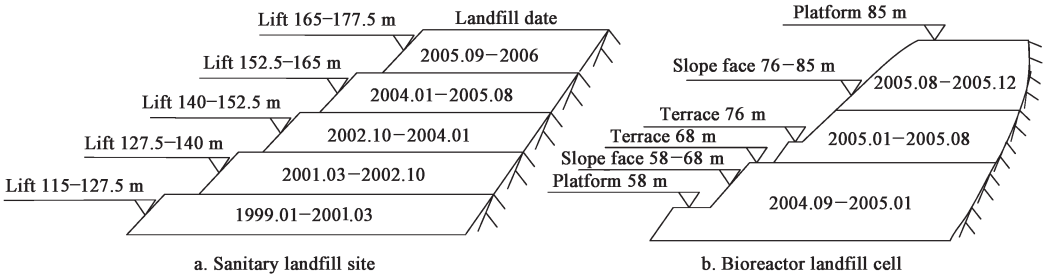

Fig. 1 Diagrammatic sketch of the sampling sites.

Table 2 Physico-chemical properties of cover soils in Hangzhou Tianziling landfill

Test site		Soil texture	Soil temp. (°C)	pH	Water cont. (%)	Organic carbon (%)	Total nitrogen (%)	Vegetation
Sanitary landfill	Lift 165–177.5 m	Sandy clay	33.2	6.7±1.3	22.9±11.7	2.50±1.97	0.231±0.206	Nil
	Lift 152.5–165 m	Sandy clay	27.9	7.5±0.56	13.4±1.62	1.55±0.15	0.116±0.020	Nil
	Lift 140–152.5 m	Sandy clay	32.2	4.5±0.36	15.7±1.27	0.37±0.08	0.075±0.081	Sparsity
	Lift 127.5–140 m	Sandy clay	37.2	7.5±0.31	13.9±5.88	1.31±0.48	0.111±0.054	Sparsity
Planted soil				4.6±0.06	13.0±0.43	0.61±0.16	0.059±0.014	Nil
	Lift 115–127.5 m	Sandy clay	29.0	7.5±0.01	22.6±4.72	0.52±0.03	0.039±0.010	*F. arundinacea*
				6.2±2.2	18.9±2.61	0.60±0.14	0.054±0.025	*N. indicum*
Bioreactor landfill	Platform 85 m	Fine sandy soil	30.2	4.9±0.01	11.5±0.68	0.33±0.00	0.037±0.001	Sparsity
	Slope face 76–85 m	Fine sandy soil	29.3	4.8±0.26	12.1±1.34	0.33±0.12	0.044±0.013	Sparsity
	Terrace 76 m	Fine sandy soil	29.1	4.7±0.16	14.0±1.95	0.25±0.04	0.033±0.013	Sparsity
	Terrace 68 m	Fine sandy soil	28.8	6.1±1.7	9.9±1.6	0.34±0.12	0.038±0.008	Sparsity
	Slope face 58–68 m	Fine sandy soil	29.5	5.1±0.22	12.2±1.36	0.28±0.01	0.043±0.005	Sparsity
	Platform 58 m	Fine sandy soil	30.1	5.7±1.7	15.8±4.49	0.34±0.11	0.030±0.025	Sparsity

Sample number n=3; Nil: without vegetation.

To assess the emission potential of N_2O from the well-vegetated cover soil with leachate irrigation, the cover soil on the slope face of Lift 115–127.5 m was vegetated in three equally sized sites (20 m × 50 m) using *Nerium indicum*, *Festuca arundinacea* Schreb and non-vegetable soil remained as control, respectively. An HDPE membrane underneath the cover soil prevents contact with leachate from landfilled MSW. Leachate collected from the sanitary landfill site was irrigated on these sites through subsurface pipe net for two years for comparison of N_2O emission potentials (Table 2) with those without irrigation.

1.2 Analytical methods

Sampling of N_2O gas from the cover soil was performed during August 2006 under humidity of 70%–73% and temperature of 32.2–34.0°C. Homemade acrylic chambers (40 cm × 40 cm × 40 cm) were equipped with fans directly mounted on the cover soil sites at a chamber-to-chamber distance of 10 m. The N_2O gas emitted from the cover soil was accumulated in the headspace of collection chambers. Four gas samples were collected from each collection chamber in 60-ml plastic syringes at 10–15 min intervals on the sampling day (Towprayoon *et al*., 2005). The N_2O content was immediately analyzed using a gas chromatography (GC-122, Jingke Instrument Co., Shanghai, China) with a glass column packed with Porapak Q (80/100 mesh) and an electron capture detector. Oven and detector temperatures were 55 and 330°C, respectively. The N_2O concentration in the sample gas was calculated by comparing the peaks for the sample gas and standard gas that appeared after 3.5 min. Before sample analysis, standard N_2O gas was diluted of 5 ppmv span to calibrate GC by the 99.99% purity gas procured from Shanghai Jiliang Standard Reference Gas Co. Ltd., China.

Soil samples at a depth of 0–10 cm were collected in triplicate at the sites next to the N_2O sampling sites. The pH of the soil suspension (soil:water = 1:2.5, w/v) was determined using a glass electrode. Soil water content (SWC) was measured by drying at 105°C for 24 h. Organic matter (OM) content in each soil sample was determined as a loss of ignition (550°C, 2 h). The NH_3^+-N, NO_3^--N, and NO_2^--N contents were extracted by shaking 10 g soil samples with 50 ml of 2 mol/L KCl for 1 h before filtering and analyzing using standard methods (State Environmental Protection Administration of China, 1997). Total nitrogen (TN) content was determined using the Kjedahl method (Lu, 2000). The soil organic carbon (SOC) was measured using the potassium dichromate-volumetric method (Lu, 2000).

1.3 N_2O emission rate

The rates of N_2O emissions from cover soil samples were estimated by the following equation (Towprayoon *et al*., 2005):

$$f = (V/A)(\Delta C/\Delta t) \quad (1)$$

where, f is the N_2O flux (μgN/(m^2·h)); V is the chamber headspace (m^3); A is the cross-sectional area of the chamber (m^2); ΔC is the concentration difference between time zero and time t (μg/m^3); Δt is the time interval between samplings (h).

1.4 Statistical analysis

Statistical analyses utilized SPSS 12.0 (SPSS, Inc. 2003. Chicago, Illinois, USA). The dependence of N_2O fluxes on a single factor was analyzed by bivariate correlation analysis.

2 Results

Table 3 lists the N_2O fluxes of soil samples collected from different lifts at the sanitary landfill site. The N_2O fluxes decreased substantially with increasing landfill age. The fluxes from young MSW layers (landfill age of 1 week or 1 year) were roughly 6 and 2 times greater than those from uncovered layers and from old layers (age of 2.5 years or 4 years), respectively. The observed N_2O fluxes from just-landfilled MSW (1 week) were 9.1–172 (μgN/(m^2·h) with a peak coefficient of variation (179%). The negative flux suggested that the soil adsorbed some N_2O from the atmosphere (Wang *et al*., 2005). Nevertheless, this value is quite low.

Table 4 lists the N_2O fluxes from Platforms or Slope faces in the bioreactor landfill site. The average N_2O fluxes from the bioreactor landfill site (5.57 μgN/(m^2·h)) were only 31% of that from the sanitary landfill site (18.1 μgN/(m^2·h)). A large data variation existed amongst the collected samples. For example, the N_2O fluxes from Terrace 76 and 68 m were only 5%–7% of that from Lift 152.5–165 m (18.6 μgN/(m^2·h)) of the same landfill age (one year).

In bioreactor landfill site, the leachate was recycled back inside the MSW layers. However, the N_2O emission rates detected from the bioreactor landfill site were lower than those from the sanitary landfill site (Table 3).

The N_2O emission fluxes from leachate-irrigated soils (Table 5) were higher than those from sanitary landfill site (Table 3). The N_2O emission rates from vegetated

Table 3 N_2O fluxes from sanitary landfill site

Site	Landfill age	N_2O flux (μgN/(m^2·h))		CV (%)
		Mean ± SD	Range	
Lift 165–177.5 m (center)	Fresh MSW (uncovered)	8.0±3.4	6.2–13.1	42.3
Lift 165–177.5 m (rim)	1 week	47.1±84.3	–9.1–172	179
Lift 152.5–165 m	1 year	18.6±8.8	7.8–29.3	47.2
Lift 140–152.5 m	2.5 years	8.1±5.2	0–14.9	64.0
Lift 127.5–140 m	4 years	8.6±8.5	0–22.5	99.1

Sample number n=4. CV: coefficient of variation.

Table 4 N_2O fluxes from bioreactor landfill site with leachate recirculation

Site	Landfill age (year)	N_2O flux (μgN/(m^2·h))		CV (%)
		Mean ± SD	Range	
Platform 85 m	0.8	1.7±2.0	0.47–4.0	114
Slope face 76–85 m	0.8	7.7±9.4	1.81–18.6	122
Terrace 76 m	1	0.89±0.24	0.57–1.65	27.0
Terrace 68 m	1	1.4±0.96	0.49–2.4	68.1
Slope face 58–68 m	1.5	1.2±0.89	0.57–2.2	77.4
Platform 58 m	1.5	20.6±14.0	9.7–36.4	60.0

Sample number n=3.

Table 5 N_2O fluxes from planted soils (Lift 115–127.5 m) with leachate irrigation

Site	N_2O flux (μgN/(m^2·h))		CV (%)
	Mean ± SD	Range	
Unplanted soil	39.7±40.5	11.0–68.3	103
F. arundinacea planted soil	117±129	27.3–297	110
N. indicum planted soil	64.0±21.0	49.2–78.8	32.8

Sample number n=3.

soil were higher than those without vegetation (Table 5), an experimental finding coincident with observations of other vegetation systems (Kiese and Butterbach-Bahl, 2002; Arnold *et al*., 2005). Vegetation on soils further enhanced N_2O flux, with that from *F. arundinacea* planted soils being higher than that from *N. indicum* planted soils (Table 5).

3 Discussion

3.1 Role of cover soils

Although there were large fluctuations in data, certain correlations were still evident in collected emission data (Tables 3, 4 and 5). The N_2O emission rate declined as landfill age increased, suggesting that nutrient limitation occurred in the cover soil for N_2O production. The N_2O emission rates from cover soil at the bioreactor landfill site were lower than those measured at the sanitary landfill site. This observation correlates with lower soil organic carbon content in the cover soil applied at the bioreactor landfill (0.25%–0.34%) than that at the sanitary landfill site (0.37%–2.50%). Vegetation on soil further enhanced the N_2O flux, with *F. arundinacea* generating more N_2O than *N. indicum*. The interactions between plants and soil determined N_2O flux.

This study clearly demonstrated that with the selected infertile soils–which have low nitrogen content and/or soil organic matter–as final cover, the N_2O emission rates from the full-scale sanitary landfill site, from the full-scale bioreactor landfill site and from the tested vegetated sites were all substantially lower than those reported at the Guangzhou Likang landfill, the Finnish Ammalssuo landfill, and Sweden's Hagby, Hőkhuvud and Hőgbytorp landfills. The MSW compositions at the Hangzhou Tianziling and the Guangzhou Likang landfills are typically considerably higher in organic nitrogen content than that in Finland or Sweden (Bőrjesson and Svensson, 1997; Lee *et al*., 2002). However, the soil organic carbon and organic matter contents at the Guangzhou Likang landfill and the Finnish Ammalssuo landfill were 3.2%–14.4% and 20%–40%, respectively, resulting in high N_2O emissions (Table 6). The organic matter in the cover soil at the Sweden Hőgbytrop landfill was at a level similar to that in this study; however, wastewater sludge at the Sweden Hőgbytrop landfill was applied as the cover soil to yield high emission potential of N_2O. At the Guangzhou Likang landfill, the untreated leachate was flooded over the cover soil, thereby producing a high N_2O flux. All these observations demonstrated that cover soil controls N_2O emissions from landfill sites.

Hence, the very low N_2O emission rates at the Hangzhou Tianziling landfill should not be attributed to a certain level of denitrification reactions occurring inside MSW layers; that is, the characteristics of cover soil controlled the emission flux.

3.2 Factors controlling N_2O flux

All experimental observations identified the minor influence of landfilled MSW and leachate recirculation, suggesting that cover soil has a significant influence on N_2O emissions from landfills. The soil parameters analyzed were water content (%), organic matter (%), pH, NH_4^+-N (mg/kg), NO_3^--N (mg/kg), NO_2^--N (mg/kg), total nitrogen (%), SOC (%) and C/N ratio.

Table 7 summarizes the r-values for bivariate correlation analyses. For all data from sanitary landfill site

Table 6 N_2O fluxes from Hangzhou Tianziling landfill and comparison with published report data

Site	Sampling period	N_2O flux (μgN/(m^2·h))		Reference
		Range	Mean	
Guangzhou Likang landfill	Jun.–Aug., 2000	618–797	–	Lee *et al*., 2002
Finland Amma1ssuo landfill	Aug., 2003	1350–1930	–	Rinne *et al*., 2005
Swenden Hagby landfill	Aug., 1993	0–575	44	Bőrjesson and Svensson, 1997
Swenden Hőkhuvud landfill	May–Jul., 1992	–1.7–163	40	Bőrjesson and Svensson, 1997
Swenden Hőgbytorp landfill (mixed cover)	Nov., 1991–Jul., 1992	0–1070	139	Bőrjesson and Svensson, 1997
Swenden Hőgbytorp landfill (pure sludge)	Feb., 1993–Nov., 1994	–10.9–35700	1800	Bőrjesson and Svensson, 1997
Hangzhou Tianziling landfill	Jul.–Aug., 2006	–9.1–172	11.8	This study

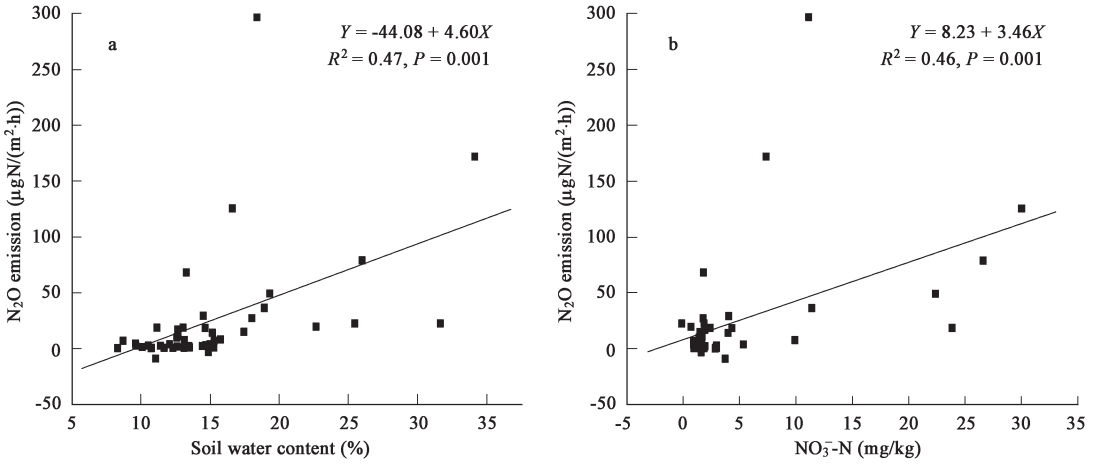

Fig. 2 Linear correlation between total N_2O fluxes and soil water content (a) and NO_3^--N content (b) for all test sites.

Table 7 Bivariate correlation analysis between N_2O flux and soil physico-chemical parameters

Soil parameter	N_2O emission *r*-value		
	All data	Sanitary landfill	Bioreactor landfill
Water content (%)	0.474**	0.705**	0.705**
Organic matter (%)	0.233	0.459*	0.324
pH	–0.090	0.146	–0.174
NH_4^+-N (mg/kg)	0.261	0.764**	0.423
NO_3^--N (mg/kg)	0.461**	0.450*	0.693**
NO_2^--N (mg/kg)	–0.023	0.008	nd
TN (%)	0.229	0.683**	–0.404
SOC (%)	0.267	0.737**	–0.104
C/N ratio	0.095	–0.042	0.706**

* $p < 0.05$ (2-tailed); ** $p < 0.01$ (2-tailed). nd: not detected.

and bioreactor landfill site, the soil water and NO_3^--N contents correlated significantly with N_2O emission rates ($p < 0.01$). However, water content correlated with the N_2O flux better than NO_3^--N content. Fig.2 presents regression plots for soil water content and NO_3^--N content. Furthermore, N_2O fluxes were significantly correlated with soil organic matter, soil organic carbon, NH_4^+-N and total nitrogen in sanitary landfill site as well, while N_2O fluxes from bioreactor landfill site were only significantly correlated with soil C/N ratio (Table 7). Obviously, soil water content, soil organic carbon, nitrogen content and distribution of mineralized-N compounds were important controlling factors for N_2O emission flux variations (Baggs *et al*., 2002; Maljanen *et al*., 2004). Nevertheless, there was no significant correlation between N_2O fluxes and above-mentioned factors in bioreactor landfill site may attribute to the quite narrow ranges of both soil C and N concentrations, SOC: 0.25%–0.34%; TN: 0.030%–0.044% (Table 2).

The effects of moisture content in soil from grasslands or agricultural lands on N_2O emission potential have been documented (Rudaz *et al*., 1999; Weitz *et al*., 2001; Wang *et al*., 2005). The observation that N_2O emission rates increased as NO_3^--N content increased demonstrated that the denitrification reaction in soil controlled the emission process; however, the conclusion will seek more evidence in the further studies. Thus, adequate moisture and nitrogen sources generated the noted N_2O emissions when an appropriate anaerobic environment was established with cover soil.

4 Conclusions

The N_2O fluxes from three test sites of Hangzhou Tianziling landfill can be minimized by utilizing selected sandy soils with low nitrogen content and fine texture as the final cover even when leachate was recycled to landfilled waste body by the bioreactor landfill or applied to irrigate the vegetated cover soil. Specifically, the N_2O emission rate was dependent on moisture content and nitrate concentrations in the cover soils. Adequate moisture and nitrogen sources generated the noted N_2O emissions when an appropriate anaerobic environment was established with cover soil.

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