Tellus (1997), 49B, 357–363
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TELLUS
ISSN 0280–6509

Nitrous oxide emissions from landfill cover soils in Sweden

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(Manuscript received 27 November 1996; in final form 7 March 1997)

ABSTRACT

Emissions of nitrous oxide measured at 4 landfill sites were found to be higher where sewage sludge was used as a landfill cover, ranging from −0.011 to 35.7 mg N_2O-N m^{-2} h^{-1}. From landfill sites covered with mineral soils, N_2O-emissions ranged from −0.0017 to 1.07 mg N_2O-N m^{-2} h^{-1}. However, extrapolation to the national level showed that sewage sludge could only be a minor source of nitrous oxide, since the amounts of sewage sludge are too small to give rise to any considerable fluxes compared with other sources. Nitrous oxide emissions from sewage sludge are comparable to emissions from most fertilizers, i.e., the N_2O-emissions are around 1% of the N-input per year.

1. Introduction

Nitrous oxide (N_2O) is an important greenhouse gas whose atmospheric concentration of N_2O has increased since pre-industrial times from about 270 to 315 ppbv (Leuenberger and Siegenthaler, 1992), corresponding to an annual rate of increase of 3.1–4.7 Tg N (Prather et al., 1995). Anthropogenic emissions are estimated at 3.7–7.7 Tg N/year, and the natural sources account for about twice this amount, while the only estimated sink, photolysis in the stratosphere, removes 9–16 Tg N per year (Prather et al., 1995). In a national inventory of N_2O-sources in Sweden (Robertson, 1991), natural sources accounted for 25.8 Gg and anthropogenic sources for 20.5 Gg N_2O-N per year.

The largest sources for N_2O discovered so far are the terrestrial, microbiologically mediated processes in soil, i.e., nitrification and denitrification (Bremner and Blackmer, 1978). For example, the turn-over of nitrogen in soils is estimated to result in the emission of 2 ± 1 kg of N_2O-N to the atmosphere per 100 kg of N applied, with low variation between different types of soils, fertilizers and crops (Mosier et al., 1996).

Very few estimates have been made on N_2O-emissions from wastewater-treatment plants and sewage sludge. Sümer et al. (1995) estimated that the release of N_2O from a German wastewater-treatment plant was 0.001% of the total amount of N received. They also calculated that the contribution of German waste purification plants was 4.5 Mg N_2O annually, which is equivalent to 56 mg N_2O per person and year. In a U. S. investigation, the estimate was higher, i.e., 3.5 g N_2O per person and year in New Hampshire. In that study 96% of the emissions derived from aeration treatments of sewage sludge were considered (Czepiel et al., 1995). These authors also estimated that the mean for the US was 3.3 g N_2O per person and year (Czepiel et al., 1996). Mosier et al. (1982) found that 1.4% of the N in sewage sludge (applied at a rate of 83.5 kg N ha^{-1}) used as fertilizer in spring barley was lost as N_2O.

The annual production of sewage sludge in Sweden is ca. 230 000 tons dry weight (Swedish Environmental Protection Agency, 1995), with a nitrogen content of 3.5% (range 2–4.9; 3.4–3.7 for the Stockholm region). Approximately 1/3 of the sludge is used as fertilizers, half the amount is

landfilled, and the other 15–20% is used for landfill covers (Statistics Sweden, 1996 and O. Palm, personal communication). The length of time that the sludge is stored, and thus exposed to the atmosphere, varies greatly depending on local regulations and market prices.

Substantial rates of N_2O-efflux were observed from sewage sludge used as landfill cover, and are reported here in comparison with N_2O-effluxes from mineral soil covers. The suitability of sewage sludge as landfill cover in this respect is discussed, and the potential N_2O-emissions from deposited sewage sludge in Sweden were calculated.

2. Methods

2.1. Site descriptions

Three municipal landfills were used in this study, all of them within 60 km from Uppsala, in south-east Sweden. Some properties of the landfills are given in Table 1.

Site A. The Hagby landfill is situated in the northern Stockholm region, and is run by SÖRAB, a private company. Most of the household waste is incinerated, but every summer ca. 10 000 cubic metres are landfilled in separate cells. A 1-m thick layer of mineral soil (mostly sandy loam) is used to cover all cells. The cell we measured was covered 10 months earlier.

Site B. The Hökhuvud landfill is situated 100 km north of Stockholm and was actively used between 1968 and 1980. It was filled with municipal solid waste from an area serving around 23 000 inhabitants (1 January 1995). The cover consists of thin layers of ashes, bark and glass wool, coated with 0.10–0.80 m of sand. Further details are given by Börjesson and Svensson (1997).

Site C and D. The Högbytorp landfill is located in Bro, NW of Stockholm, and is managed by Ragn-Sells, a private company. Site C was landfilled between 1983 and 1986, after which it was covered with a 25-cm thick layer mineral soil (intended as an intermediate cover). In 1988, a gas extraction system was applied, and around 0.5 m sewage sludge was added to the cover. In the investigated part the cover was 0.3–0.5 m thick. Soil samples taken in December 1992 were determined to contain 1.0% total Kjeldahl-N.

Site D, also at Högbytorp, is a cell measuring 25×25 m and filled with waste from households and building sites (0.73 tons/m^3). Deposition started in September 1991 and continued until the end of June 1992. Initially a 0.5-m thick layer of sewage sludge cover was applied in November 1992. In February 1993, half of the area was overlaid with another 0.5 m of sewage sludge, and the other half was covered with 0.5 m of clay.

2.2. Experimental set-up

All equipment and its handling is described by Börjesson and Svensson (1997).

(1) On site A, emission measurements were made on one occasion with 16 instantly applied 6.9-L chambers. A clay/water mixture was placed around the bottom of each chamber to serve as a seal. On sites B and C frames (basic area 0.205 m^2) were installed two weeks before the first sampling occasion. They were put in a square with 4-m sides on top of the landfill. On site D frames were installed for emission measurements with four chambers in a line on each part (clay and sewage sludge, respectively). Upon these frames, 51.3-l chambers were applied immediately before measurement and were then removed. 5 samples were withdrawn at ca. 10-min. intervals during 1 h.

(2) Soil gas concentrations were measured with permanent probes installed at the 0.50-, 0.70- and 0.90-m depths on site D, with duplicates on each part (=12 altogether), on the other 3 sites with one probe at each depth, or as deep as possible. The gas probes were sampled immediately after emission measurements had been made. On two occasions N_2O concentrations were also determined in samples taken from the gas extraction system of site D, 2.5, 6 and 10 m below ground.

(3) Soil temperatures were measured with termistors permanently installed on PVC-bars at 0.025, 0.075, 0.125, 0.175, 0.225, 0.40, 0.60 and 0.85 m depth. One such probe was put on site A and one on site B, while four probes were used on site D, with two probes on each part. On site C a soil thermometer (Kebo) was used down to 10-cm depth.

Soil moisture, expressed as % wetness (weight water of dry weight soil), was measured gravimetrically by drying at 105°C, from soil sampled down to 0.90-m depth.

All gas chromatographic analyses were made on a Chrompack 9000 equipped with an EC-detector, according to Hallin and Pell (1994).

Table 1. *Investigated landfills and some of their properties*

Landfill site	Hagby (59°28′N, 17°58′E) A	Hökhuvud (60°13′N, 18°12′E) B	Högbytrop (59°32′N, 17°38′E) C	Högbytrop (59°32′N, 17°38′E) D[1]
final covering	autumn 1992	1980	1986–88	autumn 1992
sampling period	6 August 1993	13 May –23 July 1992	15 Nov. 1991 –22 July 1992	16 Feb. 1993 –1 Nov. 1994
no. samples (occasions*chambers)	16 (1*16)	12 (3*4)	32 (7*4)	40 (5*8)
gas extraction system in use	yes	no	yes	yes
landfilled amount (tons)	12 250	ca. 60 000	100 000	12 100
area (m^2)	2000	16 000	20 000	625
height (m)	6	6–10	8–10 m	14
Soil cover				
cover thickness (m)	0.5–1	0.1–0.8	0.3–0.5	1.0
textural soil classes	–[b]	sand	loam	–[b]
coarse sand (%)		65	15	
fine sand (%)		26	12	
silt (%)		5.2	37	
clay (%)		2.4	10	
organic matter content (% weight loss on ignition 550°C)	3.2	1.3	25	38/4.9[a]
porosity (%)	35	45	69	79/48[a]

[a] Values on cover divided into pure sludge and mineral soil, respectively.
[b] Not determined.

Before August 1993, analyses were made with a split-flow for concurrent N_2O- and CO_2-analysis according to Klemedtsson et al. (1986).

2.3. *Statistical treatment*

Statistical analyses were made with JMP version 3 (SAS Institute Inc.). For emission rates linear regressions of accumulated gas concentrations were treated as zero in cases where coefficients had probability values exceeding 0.05. Of these zero-rates no chamber had more than 0.45 ppmv N_2O accumulated. The Shapiro-Wilk's W-test was used to test for normality, with a prob $< W$-value less than 0.05 indicating that data are not normally distributed. Where possible, an ANOVA (Analysis of Variance) was made with the Student t-test ($\alpha = 0.1$). For non-normally distributed data non-parametric tests (Wilcoxon/Kruskal-Wallis) were used.

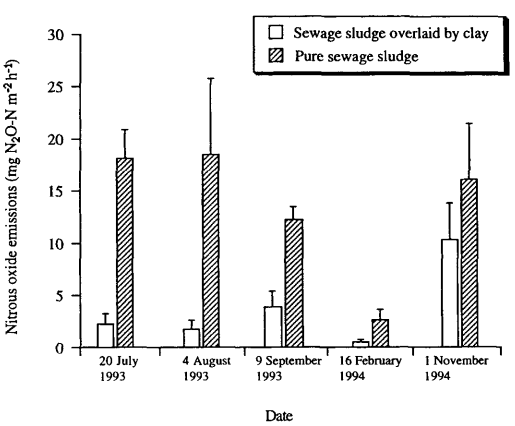

Fig. 1. Nitrous oxide emissions, given as arithmetic mean values ($n = 4$), as measured on five occasions at the Högbytorp landfill 59°32′N, 17°38′E (site D), comprising two different cover types. Bars indicate standard errors.

3. Results

3.1. Nitrous oxide emissions

Sites A, B and C emitted relatively moderate amounts of nitrous oxide (Table 2). These emissions were neither normally nor lognormally distributed. The fluctuations were quite large, especially on site A, where effluxes were recorded from only three of 16 chambers.

The site-D emissions ranged from -0.011 to 35.7 mg N_2O-N m^{-2} h^{-1} (Fig. 1). Emission-values from the mixed cover were closer to a normal distribution when converted to their natural logarithms (prob $< W = 0.209$) than in their original form (prob $< W = 0.0004$). The opposite was the case for the original values obtained for the area with only sludge as the cover (prob $< W = 0.246$). However, in this case a lognormal distribution can also be argued for (prob $< W = 0.053$). A 90% confidence interval about the mean (Student t) for emissions from the pure sludge was within 13.5 ± 11.5 mg N_2O-N m^{-2} h^{-1}. Correspondingly, emissions from the mixed cover were within 1.84 ± 3.00 mg N_2O-N m^{-2} h^{-1}, calculated from logged values ($\ln + 1$). Since the same permanent frames were used on every sampling occasion, there were three (4-1) degrees of freedom for each site.

3.2. Soil gas concentrations of N_2O

The mineral soil covers on site A and B did not show any raised levels (over atmospheric concentration) except for one sample (35 ppmv) taken in February on site A.

At site C (Högbytorp, old part) a maximum of 40 ppmv N_2O was observed at 0.175-m depth in November 1991. A few measurements of soil gas concentrations during 1992 also showed values slightly higher than atmospheric levels, up to 33 ppmv at 0.50 m depth in July.

Gas mixing ratios of N_2O in the soil profiles of site D were recorded on 3 occasions (Table 3). There were higher concentrations of nitrous oxide in the pure sewage sludge cover than in the mixed part (Wilcoxon/Kruskal-Wallis; prob $> \chi^2 = 0.0054$; medians 70 ppmv and 1.72 ppmv N_2O, respectively). The latest observation in 1994 suggested that the main part of the production had moved downwards in the profile, where a concentration of 1.8% N_2O was found at 0.90-m depth in the pure sludge part (Table 3).

Concentrations of nitrous oxide inside the D-site landfill were close to atmospheric levels: 0.202 ppmv (2.5 m) and 0.303 ppmv (10 m depth) 9 September 1993; 0.296 ppmv (2.5 m), 0.586 ppmv (6 m) and 0.403 ppmv N_2O (10-m depth) 16 February 1994.

3.3. Climatic factors

Dry conditions prevailed at sites A and B during sampling. Site A had an air temperature of 21°C and an average soil moisture down to 0.6 m depth of 16% wetness (water of dry weight soil). Air temperatures at site B were 1.5, 5.5 and 33°C with soil moistures of 22, 12 and 15% wetness for the three sampling occasions in May, June and July, respectively. This was far from the water-saturated porosity of 31% wetness in the B-site soil.

The soil of site C was almost water-saturated (around 100% wetness) at all sampling occasions except on 26 June and 21 July 1992, when the soil moisture was only around 50% wetness. The air temperatures were between $-1.5°$ and 29.5°C.

Table 2. *Emissions of N_2O from four investigated Swedish landfill sites (mg N_2O-N m^{-2} h^{-1})*

Landfill site	No. samples[a]	Range		Median	Arithmetic mean values
		min.	max.		
A	16 (3)	0	0.575	0	0.044
B	12 (7)	−0.0017	0.163	0.004	0.040
C	28 (25)	0	1.07	0.066	0.139
D mixed cover	20 (17)	−0.0109	16.7	0.931	3.46
D pure sludge	20 (20)	1.23	35.7	11.6	13.5

[a] Numbers in parenthesis are rates different from zero.

Table 3. *Mixing ratios of N_2O (ppmv) in soil gas samples taken from the landfill cover soils at Högbytorp, site D (mean of duplicate probes in each type of cover material where ranges ± are shown)*

Date	Depth (m)	Mixed cover	Pure sewage sludge cover
6 July 1993	0	1.05	1.05
6 July 1993	0.07	7.1 ± 4.8	162 ± 102
6 July 1993	0.5	3.4 ± 0.2	388 ± 384
6 July 1993	0.7	1.1 ± 0.3	3.3 ± 2.1
6 July 1993	0.9	1.6 ± 0.1	2.2 ± 0.9
9 Sept. 1993	0.5	1.2 ± 0.4	322 ± 241
9 Sept. 1993	0.7	1.1 ± 0.3	51.0 ± 43.9
9 Sept. 1993	0.9	0.78 ± 0.05	2.0
1 Nov. 1994	0.25	325 ± 24	225 ± 85
1 Nov. 1994	0.5	–	1830 ± 1100
1 Nov. 1994	0.7	3.4 ± 1.1	11.1 ± 9.6
1 Nov. 1994	0.9	320 ± 297	11 400 ± 6800

–: not determined.

Nitrous oxide emissions were not correlated either with moisture or temperature.

In the pure sludge part of site D the highest emissions occurred on the day with the lowest moisture (65% wetness; 4 August 1993). A positive correlation was found between N_2O-emissions and soil temperature: 46% of the variation in the pure sludge part could be explained by soil temperature at 17.5-cm depth ($p = 0.0038$). The regression line showed an increase by 0.74 mg N_2O-N per degree Celsius from an intercept of 2.6 mg N_2O-N m^{-2} h^{-1} at 0°C. No such correlation was evident for the mixed cover part ($p = 0.75$).

Soil temperature was consistently higher in the sludge part, indicating either that this material had a larger heat capacity or that the sludge was decomposing at a high rate.

4. Discussion

To our knowledge, data on N_2O-emissions from landfills have not been published before. Our results show that the mineral cover soils of sites A and B had low emission rates.

When applied as a landfill cover, fresh sewage sludge showed high rates of nitrous oxide emissions. Sludge cover in direct contact with air demonstrated higher emissions than sludge overlaid with 0.5 m soil at site D in our investigation. By contrast, there was no statistical difference between emissions of old sewage sludge at site C and the fluxes observed from the mineral cover soils of sites A and B. Thus, as sludge material ages and decomposes the production of N_2O decreases. The lower variability and the normal distribution of the D-site emission values were probably due to the more homogenous cover on the D-site compared with the covers on sites A, B and C. There are at least two explanations for the high emissions when the pure sludge part had a low moisture content: (1) N_2O formed by nitrification as a result of increased supply of oxygen to a larger depth of the sludge cover; (2) increased nitrification and thus supply of NO_3^- which would be denitrified after leaching into anaerobic parts of the sludge. Also the release of N_2O to the atmosphere will be easier at a lower moisture content, a factor which is likely a part of the observed increase in the N_2O-emissions.

4.1. Soil gas

The results from soil gas analyses reflect the pattern from emission measurements, with relatively low N_2O concentrations at sites A, B and C, while site D reached percentage levels. The N_2O concentrations around atmospheric level observed in the gas extraction system of site D indicate that N_2O was being produced somewhere in the system. Most likely, the reducing environment normally existing under methanogenic conditions in the waste would not allow for nitrous oxide production. However, the gas extraction system may bring in oxygen, which could then oxidize ammonium-nitrogen to nitrate which

could be denitrified when reaching the anaerobic refuse and thus give rise to N_2O.

4.2. *Extrapolated nitrous oxide emissions*

To evaluate the importance of sewage sludge landfill covers as a source of atmospheric N_2O we extrapolated the emission rates. For this purpose we assumed that measurements from July to November were also valid for April to June, while the February fluxes were adequate to winter (December until March). This yielded a weighted annual average of 11.7 mg N_2O-N m^{-2} h^{-1}, or 1.03 tons N_2O-N ha^{-1} $year^{-1}$. This is much higher than the highest N_2O-emission rate reported from soils, i.e., 165 kg N_2O-N ha^{-1} $year^{-1}$ (Duxbury et al., 1982), according to Eichner (1990). By using the density value for the sludge (0.37 tons m^{-3}), the emission rates were converted to 277 g N_2O-N ton $sludge^{-1}$ $year^{-1}$. If we assume a 3.5% N-content (the mean value given by the Swedish E. P. A. 1995 for Swedish sludge), 1.6% of the deposited N was emitted as N_2O-N during the first two years of exposure to the air.

As the N_2O-production seemed to reach the bottom of the cover after two seasons (Table 3), we assume that the emissions at high rates will level off after a time-course of two years. Furthermore, if we assume that the 20% of the Swedish sludge used as landfill cover will emit N_2O to the atmosphere at substantial rates during two years, an extrapolation of the rates calculated above will give 0.026 Gg N_2O-N per year from this source in Sweden. This would represent around 0.1% of the Swedish anthropogenic sources of N_2O, according to Robertson (1991). Estimated per capita, this is equivalent to 3.0 g N_2O-N (or 4.7 g N_2O) per person and year in Sweden, which is similar to the value of 3.5 g N_2O per person and year estimated for aerobic sewage sludge treatment processes in the US (Czepiel et al., 1996). In conclusion: emissions of nitrous oxide from sewage sludge used as landfill cover material may be high, but extrapolations of these rates demonstrate that this source of N_2O is neglectable compared with other sources.

5. Acknowledgements

This work was part of project funded by NUTEK, under contract no. 706 005–1. We would also like to thank SÖRAB, Ragnsells and the community of Östhammar for allowing us to use the sites.

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