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# Measurements of N<sub>2</sub>O from Composted Organic Wastes

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The current atmospheric mixing ratio of nitrous oxide (N<sub>2</sub>O), a chemically significant trace gas, is increasing at a rate of 0.25–0.31% yr<sup>-1</sup>, apparently well correlated with human activity. Better quantification of all N<sub>2</sub>O sources is required in efforts to stabilize this rate of increase. N<sub>2</sub>O emissions from the composting of wastewater sludge and livestock wastes were measured during 1993 and 1994 using enclosure methods. Static chambers were placed on the surface of the compost piles, and N<sub>2</sub>O mixing ratios within the chamber headspace were measured over time from which fluxes were calculated. The flux measurements resulted in mass-based emission factors of 0.7 g of N<sub>2</sub>O (dry kg)<sup>-1</sup> and 0.5 g of N<sub>2</sub>O (dry kg)<sup>-1</sup> for the sludge compost and the livestock waste compost, respectively. The derived emission factors were used in conjunction with recent waste generation and disposal statistics to estimate potential global N<sub>2</sub>O emissions from the treatment of organic wastes. Livestock waste treatment appears to hold the greatest N<sub>2</sub>O potential with estimated global emissions of  $1.2 \times 10^{12}$  g of N<sub>2</sub>O yr<sup>-1</sup>.

## Introduction

Nitrous oxide (N<sub>2</sub>O) is an important trace gas that has been identified as a significant constituent in several aspects of atmospheric science. Its efficiency in absorbing infrared radiation (about 200 times that of carbon dioxide) makes it important as a greenhouse gas (1, 2). N<sub>2</sub>O is also the dominant source of nitrogen oxides to the stratosphere which play a catalytic role in ozone destruction (3). The current atmospheric mixing ratio of N<sub>2</sub>O is about 310 ppbv and is increasing at a rate of 0.25–0.31% yr<sup>-1</sup> (4). This rate of increase implies a 40% excess of sources over sinks (5).

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To understand and possibly reduce the rate of increase of N<sub>2</sub>O in the atmosphere, all sources, both natural and anthropogenic, must be identified and quantified. Total global N<sub>2</sub>O emissions are estimated to be  $21 \pm 2$  Tg of N<sub>2</sub>O yr<sup>-1</sup> (5). Significant natural sources, which are estimated to account for about 60% of total N<sub>2</sub>O emissions, include undisturbed soils, oceanic waters, and possibly atmospheric formation (4). Numerous anthropogenic sources have also been identified but remain poorly quantified. These sources include cultivated soils, biomass burning, combustion, industrial processes, and the treatment of high organic content wastes (5, 6). Anthropogenic sources present an opportunity to reduce the current rate of N<sub>2</sub>O increase, but better quantification is necessary to establish a source hierarchy to guide the application of mitigation resources.

The major biogenic processes responsible for N<sub>2</sub>O production are nitrification and denitrification (7, 8). Nitrification is a two-step process that, under aerobic conditions, oxidizes NH<sub>4</sub><sup>+</sup> to NO<sub>2</sub><sup>-</sup> followed by the oxidation of NO<sub>2</sub><sup>-</sup> to NO<sub>3</sub><sup>-</sup>. N<sub>2</sub>O is produced as a byproduct of these reactions. Denitrification, ordinarily associated with anaerobic conditions, is a process in which NO<sub>3</sub><sup>-</sup>, NO<sub>2</sub><sup>-</sup>, N<sub>2</sub>O, or NO can be used by bacteria as terminal electron acceptors (9). NO, N<sub>2</sub>O, or N<sub>2</sub> are produced as end products or byproducts of these reactions.

The availability of oxygen (O<sub>2</sub>) is a determining factor in N<sub>2</sub>O production. During nitrification, the oxidation of NH<sub>4</sub><sup>+</sup> at low O<sub>2</sub> partial pressures results in significant N<sub>2</sub>O production, possibly as an intermediate between NH<sub>2</sub>OH and NO<sub>2</sub><sup>-</sup> (10). During denitrification, NO<sub>3</sub><sup>-</sup> is reduced to N<sub>2</sub> without appreciable N<sub>2</sub>O production in the absence of O<sub>2</sub>. But, as O<sub>2</sub> concentration increases, N<sub>2</sub>O production increases to a maximum and decreases with additional O<sub>2</sub> (11). Therefore, N<sub>2</sub>O production by both nitrifiers and denitrifiers is optimized under microaerobic conditions (12, 13).

A microaerobic environment, high mineral N availability, and a high concentration of organic C substrate have been identified as favorable conditions for N<sub>2</sub>O production (9, 14). These conditions are prevalent in several organic waste treatment systems, particularly composting systems. The theory and operation of organic waste composting systems are discussed in detail in several comprehensive reviews (15–17).

The purpose of this study was to quantify and characterize the emissions of N<sub>2</sub>O to the atmosphere from the composting of common organic wastes and to derive waste mass-based N<sub>2</sub>O emission factors from these results. A comprehensive study of N<sub>2</sub>O emissions from municipal wastewater sludge composted in aerated static piles was performed from April 1993 through January 1994. In addition, preliminary measurements of N<sub>2</sub>O emissions from livestock wastes composted in windrows was performed from September 1992 through December 1993. The resulting emission rate data were then utilized to estimate N<sub>2</sub>O emissions from composting and other organic waste treatment systems in the United States and globally.

## Field Sites

The wastewater sludge composting facility is located on the site of the Dover, NH, municipal wastewater treatment

TABLE 1

## Typical Physical and Chemical Characteristics of Sludge Prior to Composting, Finished Sludge Compost, and Finished Livestock Waste Compost

	total solids (%)	total volatile solids (%)	pH	total nitrogen, TKN (dry wt %)	total phosphorous (dry wt %)	potassium (dry wt %)
sludge	25	85	6.5	3.4	0.6	0.7
sludge compost	45	65	6.7	2.9	0.4	0.6
livestock waste compost	53	NA <sup>a</sup>	6.6	1.5	NA	NA

<sup>a</sup> NA, not available.

(WWT) plant. The WWT plant serves a population of 25 000 and generates approximately 1800 dry kg day<sup>-1</sup> of sludge from primary and secondary WWT processes. Selected sludge and sludge compost physical and chemical characteristics are presented in Table 1. The sludge is mixed with wood ash as a bulking agent in a 1:1 mixture by dry weight. Typical values for bulk density and particle density, as measured by the facility staff, were 0.47 and 0.95 g cm<sup>-3</sup>. The mixture is composted in aerated static piles (ASP) on a concrete slab under a roofed enclosure. Aeration is maintained by introducing air into the base of the piles through perforated polyethylene pipes placed on the concrete slab prior to pile construction. The aeration cycle, typically 2–5 min long, is computer controlled and determined by the feedback of temperature from probes inserted into the center of each pile. The aeration period, generally 1–3 min long, is controlled to maintain optimal temperatures for thermophilic microbial activity in a range from 50 to 60 °C. Ten compost piles are maintained on a continuously rotating basis with a new pile started approximately every 4 days. Each pile is 18 m long, 3 m wide at the base, and 2 m high and is actively aerated for approximately 40 days. The composted material is then placed outside in finishing piles for an additional 30 days prior to distribution for use in local ornamental agriculture.

The livestock waste compost piles are located at the Compost Research Center of the Kingman Research Farm at the University of New Hampshire in the town of Madbury, NH. The site consists of a 1.6 ha formerly wooded area cleared of vegetation and topsoil. Livestock waste is placed uncovered in separate static piles called windrows, approximately 11 m long, 3.7 m wide at the base, and 1.5 m high. The livestock waste consists of 25% manure and 75% seasoned bedding hay by dry weight. The manure consists of approximately equal parts cattle and horse manure. The physical and chemical characteristics of the finished livestock waste compost is presented in Table 1. Measured values of bulk density in the livestock waste compost piles ranged from a mean of 0.11 g cm<sup>-3</sup> at compost pile initiation to a mean of 0.19 g cm<sup>-3</sup> at the end of the measurement period. The windrows are turned regularly with a mechanical windrow turner to ensure nutrient homogeneity and proper aeration. These conditions help to maintain optimal temperatures for thermophilic microbial activity in a range from 50 to 60 °C. The composting process continues for approximately 1 year and is considered complete when the internal pile temperature after turning falls to ambient. The finished compost is then bagged and sold as a soil amendment.

## Methods

**Gas Sampling.** A static enclosure technique was used to measure fluxes from the surface of the compost piles (18,

19). An open aluminum collar was inserted approximately 10 cm into the compost surface and allowed to stabilize for 5–10 min prior to each flux measurement to minimize disturbance of the surface at the time of the measurement. An aluminum enclosure (51 × 51 × 26 cm) was then lowered into a water-filled channel at the top edge of the collar during each measurement to provide an air-tight seal. The enclosure surface area and volume were 0.26 m<sup>2</sup> and 93.6 L, respectively.

A battery-operated fan circulated the air within the enclosure, and a thermistor mounted inside measured enclosed air temperature. Enclosure headspace samples were collected at 0.5-min intervals in 20-mL nylon syringes (VWR Scientific) and analyzed within 4 h of collection. The gas flux,  $E_{N_2O}$  (mass N<sub>2</sub>O area<sup>-1</sup> time<sup>-1</sup>) from the surface was calculated by

$$E_{N_2O} = \frac{V}{A} p \left( \frac{\Delta C}{\Delta t} \right) \quad (1)$$

where  $V$  is the volume of the enclosure,  $A$  is the enclosed surface area,  $p$  is the gas density at the enclosure headspace temperature, and  $\Delta C/\Delta t$  is the change in N<sub>2</sub>O concentration in the enclosure headspace.  $\Delta C/\Delta t$  is calculated by linear regression of the N<sub>2</sub>O concentration in four or five headspace samples with a required correlation coefficient of 0.90 or higher, significant to the 95% confidence level for  $n = 4$  or 5 (20). The minimum measurable N<sub>2</sub>O flux was 1 mg m<sup>-2</sup> day<sup>-1</sup>.

The sludge compost was sampled approximately bimonthly from April 1993 to November 1993 using a stratified random sampling approach. Two flux measurements were performed in random locations on each of three compost piles during a sampling session. Piles were chosen randomly in each of three age categories: 0–15 days, 16–30 days, and greater than 30 days.

In addition, interstitial gas samples were collected at various depths in the profile of several sludge compost piles to determine in situ N<sub>2</sub>O concentrations. Gas samples were collected in 20-mL nylon syringes by inserting an end-perforated 3.175 mm o.d. stainless steel tube into the compost in 5–10-cm increments. Integrated compost samples, in 10-cm depth increments, were collected at the locations of interstitial gas sampling to determine moisture contents. Compost samples were collected from the surface to a depth of 40 cm. Moisture contents were determined in all samples gravimetrically by oven drying at 105 °C for 24 h (21).

Two locations were sampled repeatedly on each of two livestock waste windrows. The livestock waste windrows were sampled approximately bimonthly, one from its inception in September 1992 through December 1992 and the second from its inception in May 1993 through August 1993.

**Compost Incubations.** Sludge compost samples were incubated in the laboratory to determine the  $\text{N}_2\text{O}$  generation rate per unit mass of compost as well as the sensitivity of generation to temperature. A composited compost sample was collected from all active piles to guarantee an age-averaged generation rate estimate. A trowel was used to gather approximately 50 compost samples from a regular grid over the surface of the active piles. The samples were placed in a 20-L bucket and mixed to ensure homogeneity. Samples ranging from 50 to 75 g were then withdrawn from the composite for incubation. Moisture contents were determined in all samples as previously described.

To determine  $\text{N}_2\text{O}$  generation potential, each compost sample was subjected to a series of incubation experiments performed over a range of temperatures using methods similar to those described by ref 22. Compost samples were placed in 1.0-L air-tight glass jars fitted with 3.175-mm Swaglock unions and silicone septa. Jar headspaces were sampled by removing 3 mL of gas in 10-mL glass syringes five times during each 25-min incubation. The  $\text{N}_2\text{O}$  emission factor,  $E_{\text{N}_2\text{O}}$  (mass  $\text{N}_2\text{O}$  time $^{-1}$  dry mass compost $^{-1}$ ) was calculated by

$$E_{\text{N}_2\text{O}} = \frac{V}{m} p \left( \frac{\Delta C}{\Delta t} \right) \quad (2)$$

where  $V$  is the jar volume,  $p$  is the density of the gas at sample temperature,  $m$  is the sample dry mass, and  $\Delta C/\Delta t$  is the linear increase in  $\text{N}_2\text{O}$  concentration in the jar headspace.  $\Delta C/\Delta t$  is calculated by linear regression of the  $\text{N}_2\text{O}$  concentration in the headspace samples with statistical requirements as described for eq 1.

**Gas Analysis.** All samples were analyzed as described by Martikainen (23). Samples were immediately returned to the lab, allowed to equilibrate to laboratory temperature for 1–2 h, and analyzed for  $\text{N}_2\text{O}$  within 3–4 h after collection. Samples and standards were dried across  $\text{CaSO}_4$  as they were injected into the gas chromatograph injection loop. Analyses for  $\text{N}_2\text{O}$  were accomplished using an electron capture detector (ECD) after constituent separation using a 2 m  $\times$  3.2 mm o.d. column packed with Porapak Q. Carrier gas was a 95% Ar–5%  $\text{CH}_4$  mixture. Peaks were quantified with Hewlett Packard Chemstation software on a personal computer. The ECD was calibrated for  $\text{N}_2\text{O}$  over the range 310 ppbv to 100 ppmv during each analysis period using  $\text{N}_2\text{O}$  in  $\text{N}_2$  standard gases obtained from Scott Specialty Gases with  $\pm 2\%$  certified accuracy. Precision of analysis for calibration gases was  $\pm 0.3\%$  ( $n = 15$ ).

$\text{O}_2$  in interstitial gas samples was quantified using a Shimadzu GC-8A gas chromatograph with a thermal conductivity detector (TCD) after constituent separation using a 2 m  $\times$  3.2 mm o.d. column packed with molecular sieve 5A (80/100 mesh). Carrier gas was helium; column temperature was 70  $^\circ\text{C}$ . The TCD was calibrated for  $\text{O}_2$  over the range 1–21% by volume during each analysis period using standard gases obtained by Scott Specialty Gases with  $\pm 5\%$  certified accuracy and a NOAA/CMDL certified air standard. Precision of analysis for calibration gases was  $\pm 0.8\%$  ( $n = 12$ ).

## Results

**Sludge Compost.** A total of 74  $\text{N}_2\text{O}$  flux measurements were performed on the sludge compost piles. Thirty-three fluxes were measured with the blowers active, and 41 were measured with the blowers inactive. Figure 1 presents the

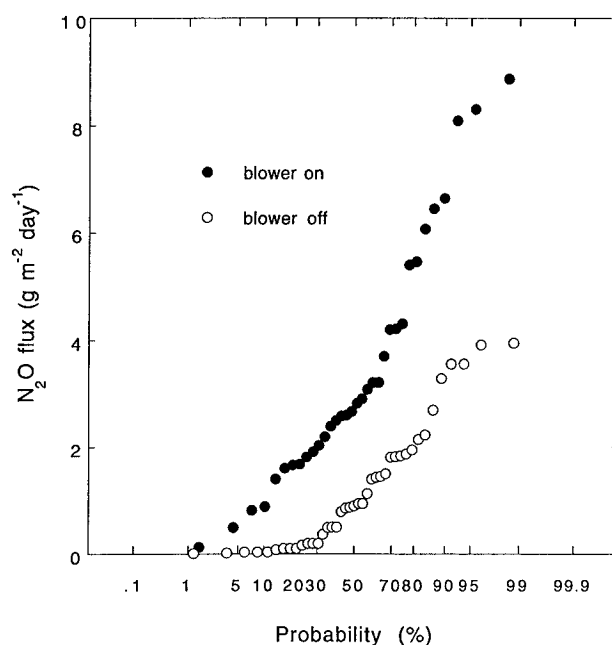


FIGURE 1. Cumulative probability plot of the measured  $\text{N}_2\text{O}$  flux data. Solid circles represent fluxes measured during active aeration; open circles represent fluxes measured with aeration inactive.

cumulative probability plot of the measured flux data with aeration active and inactive. A clear difference in the distribution of the fluxes is apparent under different aeration conditions. The mean of the fluxes during active aeration was 3.4 g of  $\text{N}_2\text{O}$   $\text{m}^{-2}$   $\text{day}^{-1}$  while the mean of the fluxes measured with the blowers off was 1.2 g of  $\text{N}_2\text{O}$   $\text{m}^{-2}$   $\text{day}^{-1}$ . The mean value of the entire flux data set, weighted by the fractional distribution of fluxes determined by aeration activity (on or off), was 2.2 g of  $\text{N}_2\text{O}$   $\text{m}^{-2}$   $\text{day}^{-1}$ .

A limited number of flux measurements were also performed on the finishing piles. Fifteen fluxes were measured in different locations between late winter and early summer, which ranged from 0.6 to 1.3 g of  $\text{N}_2\text{O}$   $\text{m}^{-2}$   $\text{day}^{-1}$  with a mean value of 0.9 g of  $\text{N}_2\text{O}$   $\text{m}^{-2}$   $\text{day}^{-1}$ .

A mass-based  $\text{N}_2\text{O}$  emission factor can be calculated for the sludge from the measured emissions. Approximately 1800 dry kg of sludge is generated daily on site and is accumulated for 4 days. A total of 7200 dry kg of sludge per pile is then composted with the added bulking agent under aeration for 40 days with a mean emission rate of 2.2 g of  $\text{N}_2\text{O}$   $\text{m}^{-2}$   $\text{day}^{-1}$ . The pile emitting surface area is 45  $\text{m}^2$ . The resulting mass-based emission rate is 0.5 g of  $\text{N}_2\text{O}$  (dry kg sludge) $^{-1}$ . A similar methodology can be used to estimate emissions from the nonaerated finishing piles. Assuming an emission rate of 0.9 g of  $\text{N}_2\text{O}$   $\text{m}^{-2}$   $\text{day}^{-1}$  and a finishing period of 30 days yields an emission rate of 0.2 g of  $\text{N}_2\text{O}$  (dry kg sludge) $^{-1}$ . The resulting total mass based  $\text{N}_2\text{O}$  emission rate is 0.7 g of  $\text{N}_2\text{O}$  (dry kg sludge) $^{-1}$ .

The disparity between fluxes under different aeration conditions is the result of the dominance of different gas transport mechanisms under varying blower conditions. Both thermal convection and gaseous diffusion transport microbially generated trace gases from the compost to the atmosphere when the blowers are inactive. When the blowers are active, advective air movement dominates transport and effectively flushes the trace gases out of the piles and into the atmosphere.

The age of the compost was also recorded at the time of each flux measurement. Figure 2 presents the measured

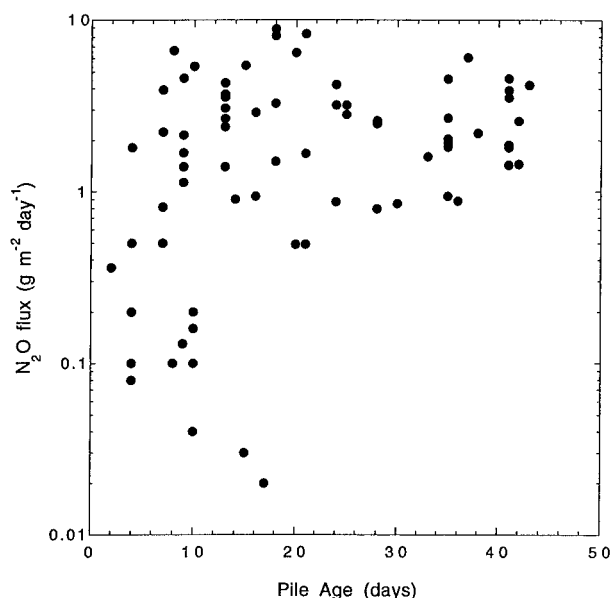


FIGURE 2. Measured  $\text{N}_2\text{O}$  fluxes as a function of compost age.

fluxes as a function of compost age. No significant statistical correlation was observed between  $\text{N}_2\text{O}$  flux and compost age. The only significant feature of this relationship is the decreasing variability in measured emissions with compost age. The highest flux variability was observed in compost less than approximately 20 days old. The interstitial gas depth profiles may help explain this variability trend.

Interstitial gas samples were collected from six compost piles of various ages from depths of 5–60 cm. Figure 3 presents the concentration profiles of  $\text{N}_2\text{O}$  and  $\text{O}_2$  in the oldest and youngest compost piles examined. The concentration of  $\text{N}_2\text{O}$  in the 9-day-old compost is highest in the top 10 cm and is negligible below 20 cm. This decrease in  $\text{N}_2\text{O}$  is accompanied by a decrease in  $\text{O}_2$  concentration from near atmospheric at 5 cm to less than 2% at 60 cm. The decline of interstitial  $\text{O}_2$  is associated with increasing compost water content with depth, which limits  $\text{O}_2$  diffusion. The measured gravimetric water contents in the 0–10-, 10–20-, 20–30-, and 30–40-cm depth intervals were 62%, 84%, 103%, and 106%, respectively. The volumetric water content, as percent of water-filled pore space (WFPS), was determined from the gravimetric water content and reported bulk density and particle density values. Calculated WFPS in the 0–10-, 10–20-, 20–30-, and 30–40-cm depth intervals were 58%, 78%, 96%, and 98%, respectively.  $\text{N}_2\text{O}$  production in the 9-day-old compost appears to be limited to the shallow depths. Water content in those areas appears to be high enough to provide an optimal moisture environment but low enough to provide enough air-filled pore space to supply sufficient  $\text{O}_2$ , ensuring an optimal microaerobic environment.

In the 38-day-old compost, the concentration of  $\text{N}_2\text{O}$  is minimal in the top 10 cm, gradually increases to a maximum at 20 cm, slowly decreases below 20 cm, and is negligible below 60 cm. The corresponding  $\text{O}_2$  concentrations are near atmospheric above 20 cm and gradually decrease to 4.5% at 60 cm.  $\text{N}_2\text{O}$  production in this pile has been significantly affected by moisture loss due to both diffusion and blower-induced advection. The measured gravimetric water contents in the 0–10-, 10–20-, 20–30-, and 30–40-cm depth intervals were 29%, 44%, 69%, and 89%, respectively. Calculated WFPS in the 0–10-, 10–20-, 20–30-, and

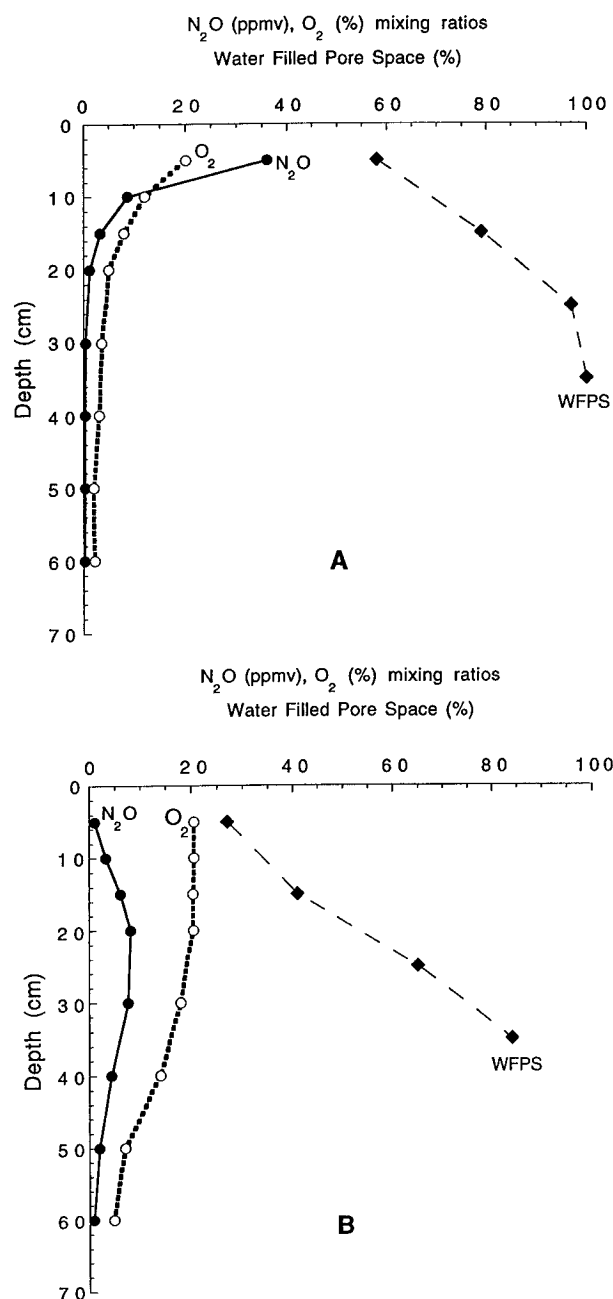


FIGURE 3. Profiles of  $\text{N}_2\text{O}$  (solid circles),  $\text{O}_2$  (open circles), and moisture content as percent water-filled pore space (solid diamonds) in the oldest and youngest compost piles examined. (A) 9-day-old compost; (B) 38-day-old compost.

30–40-cm depth intervals were 27%, 41%, 65%, and 84%, respectively.  $\text{N}_2\text{O}$  production is low above 20 cm, apparently inhibited by low moisture contents. Between 20 and 30 cm production is maximized under optimal moisture and  $\text{O}_2$  conditions and decreases with gradually decreasing  $\text{O}_2$  and increasing water.

The distribution change of  $\text{N}_2\text{O}$  production in the compost profile that accompanies drying may account for the decreasing variability of flux with age. Initially, production occurs near the surface where spatial variability is maximized due to the narrow production zone. As the production zone widens with increasing pile age, lateral spatial variability is effectively damped by the increasing depth of the  $\text{N}_2\text{O}$  emission zone. This effect may also contribute to the lack of correlation between emissions and pile age. High production within a low volume in young

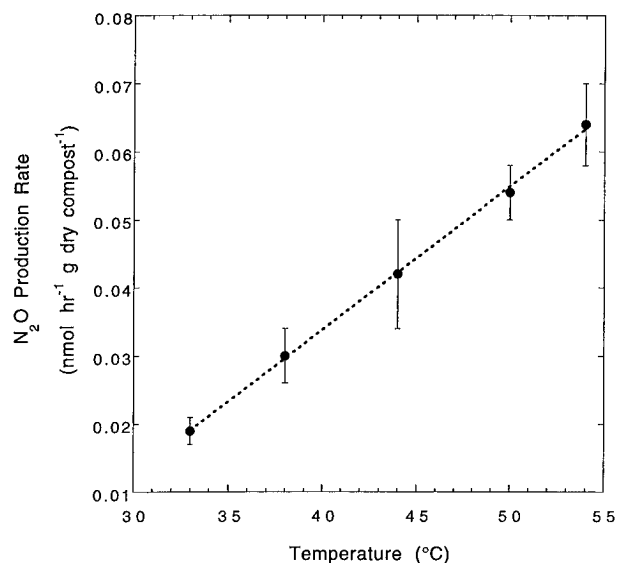


FIGURE 4. N<sub>2</sub>O production rates as a function of temperature from jar incubations of 10 composite compost samples. Solid circles represent the mean production rate; error bars represent the standard deviation.

compost appears to result in emissions similar in magnitude to those from older compost where production is low and volume is high.

Incubations of composite sludge compost samples were performed to determine the temperature sensitivity of N<sub>2</sub>O production. Near-surface compost temperature surveys were performed prior to incubations to establish the proper experimental temperature range. Temperature was measured on two occasions at a depth of 15 cm over a regular grid including compost of all ages. The temperature on April 16 ranged from 33 to 56 °C with a mean of 43 °C, while the temperature on August 1 ranged from 32 to 61 °C with a mean of 42 °C.

Incubation temperatures were chosen to simulate the observed range of field temperatures. The N<sub>2</sub>O production rate, shown in Figure 4, is presented as the mean and standard deviation of 10 samples from the original composite at each incubation temperature. N<sub>2</sub>O production was observed to increase linearly from 0.019 nmol of N<sub>2</sub>O h<sup>-1</sup> (g of dry compost)<sup>-1</sup> at 33 °C to 0.064 nmol of N<sub>2</sub>O h<sup>-1</sup> (g of dry compost)<sup>-1</sup> at 54 °C. The resulting slope of the observed N<sub>2</sub>O production–temperature curve is 0.002 nmol of N<sub>2</sub>O h<sup>-1</sup> (g of dry compost)<sup>-1</sup> deg<sup>-1</sup>.

**Livestock Waste Compost.** N<sub>2</sub>O emissions from two livestock waste windrows are shown in Figure 5. Emissions are presented as the average of two fluxes measured in two locations on each pile with error bars representing the range of measured fluxes. The presented values are those measured under normal circumstances consisting of calm to low surface winds, no recent or current precipitation, and a static pile condition. N<sub>2</sub>O emissions increased during the first 35–55 days after pile inception and decreased thereafter. The peak N<sub>2</sub>O emission rates measured on the livestock waste piles were 1.3 and 0.8 g of N<sub>2</sub>O m<sup>-2</sup> day<sup>-1</sup> from piles 1 and 2, respectively. The average of emissions measured on both piles during the observation period was 0.5 g of N<sub>2</sub>O m<sup>-2</sup> day<sup>-1</sup>.

A mass-based N<sub>2</sub>O emission factor can be estimated for the livestock waste from the measured emissions. A pile volume of 41.6 m<sup>3</sup> and an initial average dry bulk density of 133 kg m<sup>-3</sup> yields a total pile dry mass of 5533 kg.

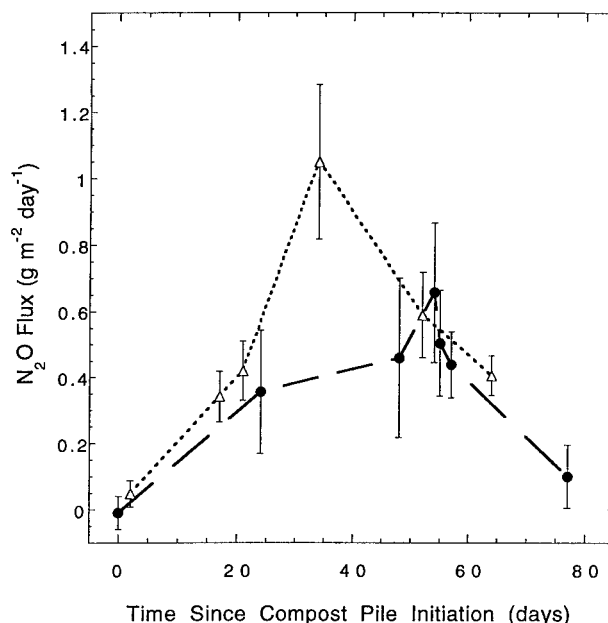


FIGURE 5. N<sub>2</sub>O emissions from livestock waste piles 1 (open triangles) and 2 (solid circles). Symbols represent flux averages; error bars represent the range of the measured fluxes.

Approximately 25% by weight or 1383 kg of this mass is manure. Total N<sub>2</sub>O emissions,  $E_{N_2O}$ , from the compost can be estimated by

$$E_{N_2O} = \frac{fAt}{m} \quad (3)$$

where  $f$  is the average emission rate (0.5 g of N<sub>2</sub>O m<sup>-2</sup> day<sup>-1</sup>),  $A$  is the effective emitting surface area (16 m<sup>2</sup>), and  $t$  is the emission period, 90 days (an extrapolation conservatively based on the observed temporal pattern of emissions), and  $m$  is the livestock manure mass (1383 kg). The resulting mass-based N<sub>2</sub>O emission factor is 0.5 g of N<sub>2</sub>O (kg of dry waste)<sup>-1</sup>.

## Discussion

Our results demonstrate the potential for significant emissions of N<sub>2</sub>O from the treatment of high organic content wastes. N<sub>2</sub>O is produced under microaerobic conditions during the processing of organic wastes with high moisture contents in the presence of adequate oxygen. These conditions are prevalent in the composting systems we observed as well as in other organic waste management systems. This similarity can be utilized to make preliminary estimates of N<sub>2</sub>O emissions from various waste management systems.

An estimate of potential global N<sub>2</sub>O emissions from wastewater sludge treatment and disposal can be generated using recent sludge production and disposal statistics from several urbanized regions of the industrialized world. Among the countries that generate large quantities of sludge, only the United States and Japan compost a significant fraction (24). The amount of municipal sludge treatment by composting in the United States in 1992 was  $1.2 \times 10^9$  dry kg yr<sup>-1</sup> (25). Approximately 50% was treated by ASP, 25% by windrowing, and 25% by in-vessel treatment. If we assume that the ultimate N<sub>2</sub>O emission potential calculated from our observations, 0.7 g of N<sub>2</sub>O (kg of dry waste)<sup>-1</sup>, is applicable to all composting systems with potentially different generation periods, then total N<sub>2</sub>O emissions from

TABLE 2

Estimated Global Livestock and Poultry Manure Generation ( $10^{11}$  wet kg  $\text{yr}^{-1}$ ) and Waste Management Systems Utilized in 1992 (29)

species	system types					
	anaerobic lagoons	liquid systems	daily spread	solid storage and drylot	pasture, range, paddock	fuel generation <sup>a</sup>
nondairy cattle	minimal	3.9	3.9	4.5	40.0	9.0
dairy cattle	5.8	5.5	5.5	8.4	6.6	1.7
swine	0.7	5.7	minimal	6.2	none	0.4
poultry	minimal	minimal	none	minimal	1.5	minimal
other <sup>b</sup>	none	none	none	minimal	20.0	none
total	6.5	15.1	9.4	19.1	68.1	11.1

<sup>a</sup> Anaerobic digestion with gas burned for fuel. <sup>b</sup> Includes goats, horses, mules, donkeys, and camels.

sludge composting in the United States in 1993 were  $8.4 \times 10^8$  g of  $\text{N}_2\text{O}$   $\text{yr}^{-1}$ .

In Japan, approximately  $1.1 \times 10^8$  dry kg  $\text{yr}^{-1}$  of municipal wastewater sludge was composted in 1988 (24). No data were available regarding specific composting methods. Using the assumptions previously presented yields an annual emission rate of  $8.0 \times 10^7$  g of  $\text{N}_2\text{O}$   $\text{yr}^{-1}$ .

Other methods of treatment and disposal of sludge and manure may also present the microaerobic conditions necessary for  $\text{N}_2\text{O}$  emissions. Land disposal of organic wastes appears to hold the most significant potential for  $\text{N}_2\text{O}$  emissions globally due to its widespread application and the large volume processed. Amending soils with wastewater sludge and animal manure has been observed to substantially increase  $\text{N}_2\text{O}$  emissions (9, 26, 27). However, the observed  $\text{N}_2\text{O}$  emission rates in these amended soils are lower and the generation periods are longer than those observed in more concentrated waste environments such as our compost piles. This appears to be the result of the influence of lower temperatures and variable moisture conditions on microbially mediated  $\text{N}_2\text{O}$  production. A preliminary estimate of potential  $\text{N}_2\text{O}$  emissions from land application can be calculated if we assume that the ultimate yield of  $\text{N}_2\text{O}$  from land-applied wastes is similar to that observed in our compost measurements, albeit at different rates and under different physical conditions.

In the United States,  $1.6 \times 10^9$  dry kg  $\text{yr}^{-1}$  of municipal wastewater sludge was land disposed in 1988 while in western Europe, where sludge composting is minimal,  $2.4 \times 10^9$  dry kg  $\text{yr}^{-1}$  of municipal wastewater sludge was land disposed in 1992 (24, 28). Assuming the ultimate  $\text{N}_2\text{O}$  yield from our compost observations (0.7 g of  $\text{N}_2\text{O}$  (kg of dry waste) $^{-1}$ ) and a total of  $4.0 \times 10^9$  dry kg  $\text{yr}^{-1}$  of municipal wastewater sludge land disposed (yields annual  $\text{N}_2\text{O}$  emissions of  $2.8 \times 10^9$  g of  $\text{N}_2\text{O}$   $\text{yr}^{-1}$ ). Land disposal of sludge in Japan is not significant in the sludge disposal budget.

The total  $\text{N}_2\text{O}$  emissions potential from the wastewater sludge treatment and disposal systems considered is estimated to be  $3.7 \times 10^9$  g of  $\text{N}_2\text{O}$   $\text{yr}^{-1}$ . Unfortunately, few reliable data exist regarding wastewater treatment and sludge generation in the increasingly populated urban regions of Asia, Africa, and Central and South America. The addition of sludge statistics from these areas could be expected to significantly increase estimated  $\text{N}_2\text{O}$  emissions from the treatment of wastewater sludge.

The treatment and disposal of livestock wastes appear to hold the greatest potential for  $\text{N}_2\text{O}$  emissions globally. Approximately  $13.2 \times 10^{12}$  wet kg  $\text{yr}^{-1}$  of livestock and

poultry manure was generated globally in 1992 (29). The distribution of generated wastes by species and the waste management systems utilized are presented in Table 2. The microaerobic conditions necessary for  $\text{N}_2\text{O}$  production are common in several of these systems including the following: daily spread and paddock, where manure is collected daily and spread on fields; pasture and range, where manure is left "as is" on fields; drylot, where manure is dried on feedlots and later spread on fields; solid storage, where manure is collected and stored for longer than daily periods and ultimately field spread, which may include composting. Also, the final disposal of liquid storage is generally field application of the accumulated wastes. Lagoon and digestion systems, where wastes are treated under anaerobic conditions, would not be expected to produce significant quantities of  $\text{N}_2\text{O}$ , although disposal of the resulting sludge, which is generally field spread, may hold some potential for  $\text{N}_2\text{O}$  emissions.

We can estimate potential  $\text{N}_2\text{O}$  emissions using the global manure production numbers presented in Table 2 if we make the assumption that the ultimate yield of  $\text{N}_2\text{O}$  from land-applied manure is similar to that observed in our compost measurements.  $11.2 \times 10^{12}$  wet kg  $\text{yr}^{-1}$  of livestock and poultry manure was treated and disposed by the systems previously described that provide the microaerobic conditions necessary for  $\text{N}_2\text{O}$  production. Assuming a composite total solids content as excreted of 20% (30) yields a manure dry mass of  $2.3 \times 10^{12}$  kg  $\text{yr}^{-1}$ . Using the mass-based ultimate  $\text{N}_2\text{O}$  yield derived from our livestock waste compost observations, 0.5 g of  $\text{N}_2\text{O}$  (kg of dry waste) $^{-1}$ , yields annual  $\text{N}_2\text{O}$  emissions from livestock waste of  $1.2 \times 10^{12}$  g of  $\text{N}_2\text{O}$   $\text{yr}^{-1}$ .

Elevated  $\text{N}_2\text{O}$  mixing ratios have recently been observed around manure piles and feed lots (4). A global emission rate of  $0.3 \times 10^{12}$ – $0.8 \times 10^{12}$  g of  $\text{N}_2\text{O}$   $\text{yr}^{-1}$  was extrapolated from observations of the observed ratios of methane to  $\text{N}_2\text{O}$  and estimated global methane emissions from cattle. Our estimate compares favorably with this range if the contribution of other livestock wastes are also considered. Cattle wastes account for approximately 75% of total global livestock wastes (Table 1). Including only cattle wastes in our estimate would reduce estimated emissions to  $0.9 \times 10^{12}$  g of  $\text{N}_2\text{O}$   $\text{yr}^{-1}$ , close to the upper end of the previously estimated range.

Our extrapolation of global  $\text{N}_2\text{O}$  emissions should be considered a probable upper bound due to the significant physical differences between the compost and land-applied environments as well as the limited range of corresponding methods and source materials investigated. These esti-

mates are a first step toward a quantitative evaluation of global N<sub>2</sub>O emissions from the treatment and disposal of organic wastes. Several further steps must be taken to better characterize this apparently significant N<sub>2</sub>O source. More comprehensive measurements must be performed that encompass the full range of composting technology in current use. Reliable N<sub>2</sub>O emission factors for land-applied wastes based on quantitative field measurements need to be established to determine if the predicted potential is actually realized. A more detailed compilation of global waste generation, treatment and disposal statistics is necessary to determine the quantity of wastes that are exposed to conditions favorable for N<sub>2</sub>O production.

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