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Mark S. Coyne
University of Kentucky, mark.coyne@uky.edu

R. A. Gilfillen
University of Kentucky

Robert L. Blevins
University of Kentucky

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Atmospheric Pollutants and Trace Gases

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M. S. Coyne,* R. A. Gilfillen, and R. L. Blevins

ABSTRACT

Adding carbon-rich materials to fields, like manure, may enhance denitrification. Grass filters, which are used to trap surface runoff from these fields, may also provide a carbon-rich environment that favors water infiltration and denitrification. Nitrous oxide (N_2O) may be evolved in these settings. It is a radiatively important trace gas and intermediate in the denitrification pathway and several other microbial processes. We measured N_2O flux, after simulated rain, using a soil cover technique in poultry-manured plots and grass filters receiving their runoff. Intact soil cores were used to relate the N_2O flux to the denitrification potential of the plots. Nitrous oxide fluxes were smaller in grass filters than in manured plots, even though more denitrifying bacteria were present. The average N_2O flux in the three most dynamic erosion plots was 755 µg N_2O-N $m^{-2}h^{-1}$, which was 39% of the maximal denitrification rate measured in acetylene-blocked, NO_3^--amended soil cores. Nitrous oxide flux immediately after rainfall was greater than N_2O flux measurements reported for similar agricultural settings.

AGRICULTURAL SOILS in Kentucky are increasingly used to dispose of animal wastes from livestock production and processing facilities—particularly an expanding poultry (*Gallus gallus domesticus*) industry (Kentucky Agric. Stat., 1992, p. 59). One by-product of waste application to agricultural land is nitrate-N (NO_3^-), due to mineralization and nitrification. If NO_3^- is formed in excess of crop uptake, it can contribute to groundwater contamination after transport in surface water runoff.

Grass filters are used to control runoff from agricultural land (Gross et al., 1991). They remove some of the soluble nutrients, like NO_3^-, which are intercepted during surface flow. Groffman et al. (1991) found that denitrification in grass filters removed 14 to 25% of the NO_3^- applied depending on the grass variety. If additional C was added, the potential NO_3^- removal by denitrification increased. They suggested that runoff containing available C, in addition to NO_3^-, such as runoff from manure-amended fields, might promote greater denitrification in grass filters than runoff poor in C.

If denitrification were promoted in grass filters, nitrous oxide (N_2O) evolution could also increase, because it is one of the principle intermediates in the denitrification pathway (Firestone, 1982). Because N_2O may contribute to global warming and atmospheric ozone destruction, information about its sources in agricultural settings is important for modeling global N_2O flux. It is particularly

important to obtain flux measurements when N_2O evolution is most dynamic—such as immediately after rainfall.

We measured N_2O flux after simulated rain in poultry manured soil and grass filters receiving their runoff. Our goal was to assess the N_2O flux in poultry-manured soil immediately after rain in comparison to reported N_2O flux measurements from similar agricultural settings. We also wanted to assess N_2O flux from grass filters receiving the runoff from these poultry-manured fields.

MATERIALS AND METHODS

Site. This study was done at the University of Kentucky Agricultural Experiment Station in Lexington during June and July 1992. Experimental plots were on a Maury silt loam soil (fine, mixed, mesic Typic Paleudalf) with an average natural slope of 9% and soil permeability ranging from 5 to 15 cm h^{-1} (Blevins et al., 1990). Six individual erosion plots 4.6 m wide by 22.1 m long were used (Fig. 1). A grass filter either 4.5 m or 9.0 m in length abutted each erosion plot. The grass filters were a mixed sod composed of tall fescue (*Festuca arundinacea* L.) and 'Kentucky' bluegrass (*Poa pratensis* L.). For 8 continuous years before this study, the tillage management used on these plots was conventional tillage in Plots 1 and 3, no-tillage in Plots 2 and 4, and chisel plow tillage in Plots 3 and 6. In 1992, the tillage management in all plots was chisel plow tillage.

Site Treatment. Poultry litter mixed with sawdust and wood shavings bedding from a breeder house was briefly stockpiled and then uniformly spread over each erosion plot on 29 June 1992 at 16.5 Mg ha^{-1} (wet wt.). The poultry litter contained 2.8% total N, 2.93% total P, and 1.78% total K (wet basis) at a moisture content of 34.2%. Poultry litter was partially incorporated into each plot with a chisel plow as the only tillage practice. Erosion plots were covered with black plastic tarps to protect them from natural rain but which allowed air circulation. Grass filters were not covered.

On 1, 7, 9, 14, 16, or 21 July 1992, rain was simulated on one of the erosion plots beginning with Plot 1. The plastic tarp was removed and simulated rain was delivered to each erosion plot at about 6.4 cm h^{-1}. This intensity approximates a 1-in-10-yr storm event in central Kentucky. A storm of such intensity occurred in Lexington on 16 July 1992 (Fig. 2). Moore et al. (1983) have previously described the rain simulator used in our study.

The duration of simulated rainfall varied from plot to plot, because each erosion plot had different runoff characteristics, and because simulated rain continued until runoff was measured for at least 1 h at the bottom of grass filters with two different lengths (Fig. 1). Consequently, simulated rain lasted for 115 min in Plot 1, 135 min in Plot 2, 140 min in Plot 3, 136 min in Plot 4, 100 min in Plot 5, and 110 min in Plot 6. Surface runoff from

Department of Agronomy, N-122 Agricultural Science Building North, Univ. of Kentucky, Lexington, KY 40546-0091. Received 13 May 1993. *Corresponding author (mscoyn00@ukcc.uky.edu).

Abbreviations: ECD, electron capture detector; TCD, thermal conductivity detector; MPN, most probable number.
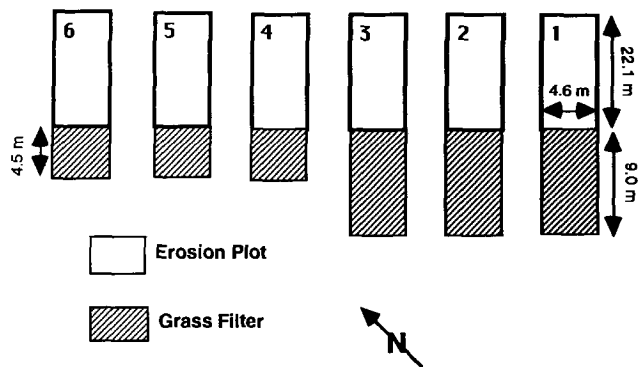
Fig. 1. Diagram of the study plots.

the erosion plots was usually observed 20 to 30 min after simulated rain began.

Soil Cover Measurements. After simulated rain ceased, soil covers were immediately inserted to a depth of 2.5 cm in the middle of the erosion plot. The soil covers placed in the grass filters were within 1 m of the erosion plots; this location became saturated by surface runoff during simulated rain. The soil covers were coffee cans 17.1 cm high by 15.6 cm diam. with the bottoms removed and a rubber septum penetrating the original plastic lid. Preliminary experiments with N_2O indicated that the cans remained gas tight for the duration of field measurements (data not shown). Five replicates were used at each location. At 0-, 15-, 30-, and 60-min intervals, gas samples were removed from the soil cover head space and stored in preevacuated Vacutainers (Beckton Dickinson, Rutherford, NJ) for N_2O analysis.

Intact Core Denitrification Measurements. Five, randomly distributed, intact soil cores 15 cm in depth were collected from each erosion plot and grass filter strip in 21 cm high by 2.5 cm diam. plastic sleeves about 1 mo after simulated rainfall studies. The cores were saturated with 10 mM KNO_3 and allowed to stand for 30 min before excess solution was drained and the plastic sleeve sealed at both ends with sleeve-type rubber stoppers. The head space was evacuated and flushed three times with N_2. Ten milliliters of head space gas were removed and replaced by an equal volume of reagent-grade acetylene. One-milliliter gas samples were removed from the head space at 0-, 2-, and 6-h intervals.
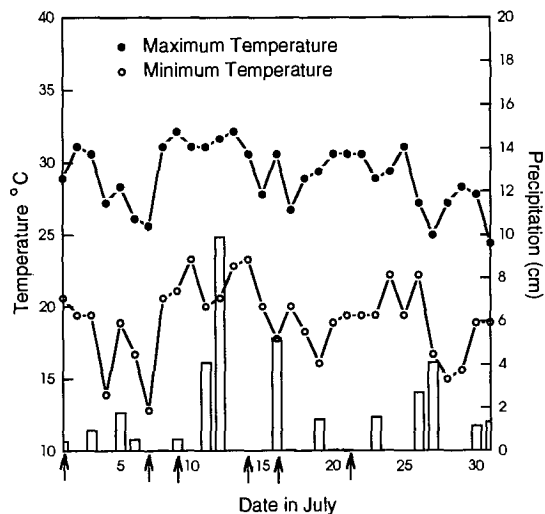

Fig. 2. Precipitation and average daily temperatures during the study (arrows indicate dates of rain simulations).

Gas Analysis. Gas samples were analyzed for N_2O on a Varian 3700 gas chromatograph with 2 m Porapak Q columns using an electron capture detector (ECD) for soil cover samples and a thermal conductivity detector (TCD) for intact cores. Analysis conditions for the ECD were: detector temperature, 360°C; column temperature, 60°C; carrier gas, 95% argon, 5% methane; carrier gas flow 30 mL min^{-1}; and sample volume, 1.0 mL. Analysis conditions for the TCD were: detector temperature, 120°C; filament temperature, 140°C; column temperature, 60°C; carrier gas, helium; carrier gas flow rate, 35 mL min^{-1}; and sample volume 1.0 mL. Machine response to N_2O was measured and compared to standard curves for N_2O generated from gas standards of known concentration.

Chemical Analyses. Chemical analysis of poultry litter was done in the University of Kentucky Regulatory Services soil testing lab. Soil samples for chemical characterization were taken from the 0- to 15-cm depth interval in both the erosion plots and the grass filters before addition of poultry litter. The pH was measured in a 1:1 soil/water slurry. Percent soil C was measured on a CR 12 Leco Carbon Determinator (Leco Corp., St. Joseph, MI).

Microbial Analysis. Soil samples used to determine denitrifier most probable number (MPN) were removed from the 0- to 15-cm depth interval in both the erosion plots and the grass filters about 24 h after rain simulation in each plot. The denitrifier MPN in erosion plots and grass filters was determined as outlined by Tiedje (1982). A 10-fold serial dilution of soil in physiological saline (8 g L^{-1} NaCl in distilled H_2O) was used to inoculate five replicate tubes per dilution. Growth media was Tryptic Soy Broth with 1 g L^{-1} KNO_3. The tubes were incubated 28 d at 26°C and residual NO_3^- was detected with diphenylamine in concentrated sulfuric acid. The MPN of denitrifiers were determined using published tables (Alexander, 1982).

Statistical Analysis. ANOVA and t-tests were made using the CoStat (CoHort Software, Berkeley, CA) statistical software package for personal computers.

RESULTS AND DISCUSSION

Nitrous oxide flux rates immediately after simulated rain were greater in the erosion plots than they were in the grass filters ($\alpha = 0.05$; Table 1). The average coefficient of variation in flux rates between plots was 56% in erosion plots (range 25–100%) and 91% in grass filters (range 36–131%). This spatial variability is not unique for N_2O field measurements (Goodroad et al., 1984; Mosier et al., 1986). The average N_2O flux rate in Plots 1, 2, and 3 (775 µg N_2O-N m^{-2} h^{-1}), the first plots treated, was greater than the average N_2O flux rate in Plots 4, 5, and 6 (134 µg N_2O-N m^{-2} h^{-1}; $\alpha = 0.05$). We did not find this difference in the grass filters.

Nitrous oxide fluxes have been measured in numerous environments. Based on an evaluation of various field experiments, Eichner (1990) estimated that 2% of N fertilizer is lost as N_2O over a 1-yr period in fertilized and manured soils. Average daily flux was 24 µg N_2O-N m^{-2} h^{-1} from ammonium-fertilized grass and 7 µg N_2O-N m^{-2} h^{-1} from soil (Eichner, 1990). The N_2O flux immediately after rain in poultry-manured soils suggests that this flux could be much greater, albeit, for a short period. The flux rates we measured may underestimate the true N_2O flux rate due to the solubility of N_2O in water. Nitrous oxide has an adsorption coefficient between 0.544 and 0.472 mL N_2O per mL of water from 25 to 30°C (Tiedje, 1982);

Table 1. Mean N_2O evolution immediately after simulated rainfall in poultry-manured erosion plots and grass filters receiving their runoff.

Plot	Days after litter applied	Erosion plot†	Grass filter
		——— μg N_2O-N m^{-2} h^{-1} ———	
1	2	1000 ± 527	148 ± 177
2	8	559 ± 191	58 ± 76
3	10	763 ± 581	85 ± 44
4	15	149 ± 110	79 ± 57
5	17	51 ± 51	76 ± 83
6	22	201 ± 51	76 ± 83

† Mean of five soil covers ± 1 SD.

Table 2. Chemical and biological differences between erosion plots and grass filters.

	Average of Plots 1, 2, and 3		Average of Plots 4, 5, and 6	
Soil property	Erosion plot	Grass filter	Erosion plot	Grass filter
pH	5.5	6.7*	5.7	6.6*
% C	2.08	1.93	1.98	1.97
MPN† Denitrifiers	2.8×10^4	4.0×10^6	1.2×10^3	2.6×10^5

* Indicates a difference between the erosion plot and grass filter in each group of plots (t-test, α = .05).
† MPN (most probable number) of denitrifiers g^{-1} oven dry soil. The 95% confidence interval is ± 3.3 × MPN.

this range spans the soil temperatures observed during flux measurements.

Goodroad et al. (1984) found a mean N_2O flux from a manure-amended, no-till corn (*Zea mays* L.) experiment in Wisconsin of about 100 μg N_2O-N m^{-2} h^{-1}. However, during spring thaw, when soils were presumably saturated, N_2O flux reached 634 μg N_2O-N m^{-2} h^{-1}. A period in their study, comparable to conditions we created with our rainfall simulation, occurred during the first rainfall after manure addition to soil. Goodroad et al. (1984) measured a N_2O flux of about 400 μg N_2O-N m^{-2} h^{-1}. This is comparable to the average N_2O flux rates we observed in our first three plots after simulated rain. Mosier et al. (1986) observed that, in irrigated corn fertilized with 200 kg N ha^{-1} as NH_4SO_4, N_2O emissions peaked at 565 and 504 μg N_2O-N m^{-2} h^{-1} after a 7.8-cm and 7.0-cm rain, respectively.

Nitrous oxide may be evolved during autotrophic and heterotrophic nitrification (Robertson and Tiedje, 1987) and NO_3^- respiration (Smith and Zimmerman, 1981) as well as during denitrification. We cannot rule out the first three processes as the source of N_2O because we did not selectively inhibit them (Robertson and Tiedje, 1987). However, the intensity and duration of rainfall would have created soil conditions favorable for denitrification.

The NO/N_2O ratio may also be a useful indicator of whether N-oxide flux comes from denitrification or some other process (Davidson, 1991). Anderson and Levine (1986) found that the NO/N_2O ratio was 0.01 to 0.3 for denitrifiers and 0.9 to 5.6 for nitrifiers and NO_3^- respirers. The Vacutainers used for gas sampling were contaminated with NO. However, even with this background NO, the NO/N_2O ratio was <0.01 (data not shown), which suggests that, initially, denitrification was the main source of N_2O from these plots.

What could account for the different flux rates between the erosion plots and grass filters, and the dramatic difference in N_2O flux rates between the first three erosion plots and the last three? We suspected that if denitrification were a major N_2O source, different denitrifier population size might be responsible. When examined, Plots 1, 2, and 3 had 10-fold greater denitrifier MPN than Plots 4, 5, and 6 (Table 2). We believe this difference may be because erosion Plots 4, 5, and 6 were covered for an extended period by a plastic tarp when maximum daily temperatures exceeded 27°C (Fig. 2). However, we did not

test this hypothesis by comparing pre- and postexperiment denitrifier MPN.

The grass filters were not covered during the experiment, yet we also found a difference in the average denitrifier MPN of Plots 1, 2, and 3 and Plots 4, 5, and 6. If intrinsic soil properties varied across the experiment site from Plot 1 to Plot 6, they were not among the parameters we measured. Neither pH nor %C differed significantly across erosion plots or across grass filters (Table 2).

Grass filters had 100-fold greater denitrifier MPN and greater pH than erosion plots (α = 0.05; Table 2). So, denitrifier population size is an unsatisfactory explanation for the differences in N_2O flux rates between erosion plots and grass filters. A better explanation is that more NO_3^- or NH_4^+ was available in the erosion plots than the grass filters. In 1991, all erosion plots were fertilized with 170 kg N ha^{-1} as NH_4NO_3 and subjected to at least two separate rate simulations. The poultry manure we added to each erosion plot in 1992 contained about 304 kg N ha^{-1}. Fresh poultry manure typically consists of 25 to 30% urea and ammonium forms (Rasnake et al., 1991), some of which would have been lost by volatilization under the plastic tarps. The remaining ammonium and mineralizable N forms would nitrify over time.

We did not measure soil NO_3^- and NH_4^+ concentrations either before or after rain simulation. However, we measured NO_3^- concentrations in surface runoff. The maximal NO_3^- concentration in surface runoff from erosion plots increased from Plot 1 to 6. This corresponds with the order in which simulations were done (Table 3). The only source of N added to the grass filters was runoff from the erosion plots. The final NO_3^- concentration in surface runoff from the erosion plots immediately before N_2O flux measurements began was greater than in grass filters

Table 3. Concentration of NO_3^--N in surface runoff.

		Maximal		Final	
Plot	Days after litter applied	Erosion plot	Grass filter	Erosion plot	Grass filter
			mg NO_3^--N L^{-1}		
1	2	0.53	0.56	0.52	0.42
2	8	0.73	0.57	0.57	0.50
3	10	1.02	0.45	0.39	0.38
4	15	1.12	1.15	0.43	0.43
5	17	0.69	0.64	0.49	0.43
6	22	1.50	0.55	0.59	0.36

Table 4. Mean N₂O evolution in acetylene-blocked, NO₃⁻-amended, intact soil cores from erosion plots and grass filters.

Plot	Erosion plot	Grass filter
	μg N₂O-N m⁻² h⁻¹†	
1	393 ± 596	297 ± 483
2	2590 ± 3580	4810 ± 4310
3	2750 ± 2810	2710 ± 1260
4	3840 ± 7850	557 ± 1250
5	190 ± 279	629 ± 818
6	112 ± 155	ND‡

† Mean of five cores ± 1 SD.
‡ ND = no data.

(α = 0.05; Table 3). This evidence, along with the fertilization history of the erosion plots, suggests that the erosion plots contained more available NO_3^- than the grass filters at the start of N_2O flux measurements.

Was the potential N_2O flux from an erosion plot simply greater than in a grass filter? If part of the N_2O flux were associated with denitrification activity, denitrification potential in erosion plots and grass filters should reflect the different N_2O flux rates observed. Intact soil cores were removed from erosion plots and grass filters to test this hypothesis.

The coefficients of variation were considerably greater in our intact core studies than they were in our field measurements. The average coefficient of variation was 147% for erosion plot cores (range 102–205%) and 131% for grass filter cores (range 47–224%). There was no significant difference between the denitrification potential of the two sets of cores (α = 0.05; Table 4).

The average N_2O flux from erosion Plots 1, 2, and 3, 755 μg N_2O-N m⁻² h⁻¹, was equivalent to 39% of the average total N-flux we observed in NO_3^--amended, acetylene blocked, intact cores from Plots 1, 2, and 3 (Tables 1 and 4). In contrast, the average N_2O flux from the grass filters in Plots 1, 2, and 3, 97 μg N_2O-N m⁻² h⁻¹, was only 4% of the average total N gas flux under acetylene blocked conditions. Groffman et al. (1991) suggested that surface runoff from manured soils might carry sufficient C to enhance denitrification in adjacent grass filters, thus removing NO_3^- before it reaches groundwater. If denitrifying conditions were created in the grass filters, the smaller N_2O flux rates may simply be due to more N_2 production than in the corresponding erosion plots.

CONCLUSION

If an accurate estimate of agricultural N_2O input to global N_2O flux is to be made, models of global atmospheric N_2O flux from agricultural soils must account for N_2O flux in soils undergoing wetting and drying cycles. Nitrous oxide flux immediately after rainfall can exceed 1 mg m⁻² h⁻¹ for an indeterminate period in poultry manure-amended fields. More refined field studies are needed to demonstrate the source of N_2O evolved in these settings. If the major N_2O source is denitrification, further research must demonstrate whether denitrification in grass filter strips is enhanced by the C contained in surface runoff from adjacent manured fields.

ACKNOWLEDGMENTS

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