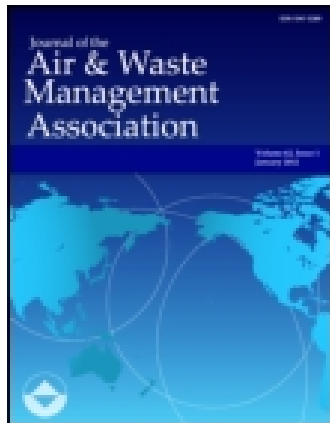


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Journal of the Air & Waste Management Association

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/uawm20>

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Published online: 01 Mar 2012.

To cite this article: Viney P. Aneja, Benny D. Holbrook & Wayne P. Robarge (1997) Nitrogen Oxide Flux from an Agricultural Soil During Winter Fallow in the Upper Coastal Plain of North Carolina, U.S.A., Journal of the Air & Waste Management Association, 47:7, 800-805, DOI: [10.1080/10473289.1997.10463933](https://doi.org/10.1080/10473289.1997.10463933)

To link to this article: <http://dx.doi.org/10.1080/10473289.1997.10463933>

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Nitrogen Oxide Flux from an Agricultural Soil During Winter Fallow in the Upper Coastal Plain of North Carolina, U.S.A.

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ABSTRACT

Incorporation of the remaining crop residue, including the root system, of grain (soybean and corn) and fiber (cotton) crops into the soil following harvest is a common agricultural practice. The crop residue represents a substantial portion of nitrogen initially applied as fertilizer, and thus is a potential source of nitrogen for NO emissions during the winter fallow period. Fluxes of NO and NO₂ were measured from fallow fields from February 7 to March 23, 1994, using a dynamic chamber technique (ambient air as the carrier gas). Average NO flux rates, as a function of previous crop residue, were 9.2 (range -4.2 to 76) ng-N m⁻² s⁻¹ for soybean, 6.1 (range -11.7 to 110) ng-N m⁻² s⁻¹ for cotton, and 4.7 (range -0.2 to 40) ng-N m⁻² s⁻¹ for corn. Maximum NO fluxes were observed in mid-morning when soil temperatures were lowest. Minimum NO flux occurred after mid-afternoon when soil temperature reached a maximum. The decrease in NO flux with increase in soil temperature (5 cm depth) reflected the existence of a NO compensation concentration (i.e., the rate for the NO consumption reactions continued to increase with increase in temperature). NO₂ deposition was

calculated for 92% of the data points, with no trend in deposition between the three fields and their corresponding crop residue. These results indicate that significant fluxes of NO are generated from fallow agricultural fields following incorporation of the residue from the previous crop.

INTRODUCTION

In 1991, the National Academy of Sciences report "Rethinking the Ozone Problem" concluded that efforts to attain the National Ambient Air Quality Standards (NAAQS) for ozone within many areas of the United States have failed, primarily because of the lack of adequate precursor emission inventories.¹ Emphasis has now shifted toward evaluating rural sources of these precursors, especially the emission of nitrogen oxides from intensively managed (i.e., high rates of nitrogen fertilizer application) agricultural soils. Nitrogenous fertilizer usage in the United States has been estimated by Matthews² to be in excess of 10,000 × 10⁹ grams nitrogen o_r year. This quantity of fertilizer nitrogen usage represents a potential source of nitric oxide (NO) and nitrogen dioxide (NO₂) emissions that could have a significant impact on tropospheric ozone formation. This is especially true in the southeastern United States, which has approximately 40% of the ozone non-attainment areas in the country,³ and rural summertime ozone concentrations are among the highest recorded in the United States.⁴

A number of studies and reviews have been published concerning NO emissions from agricultural soils,⁵⁻¹² but of these, relatively few have reported measurements of NO emissions in the southeastern United States. Valente and Thornton⁸ suggested that agricultural soils, together with pasture and forest soils, may equal approximately 60% of the current NO and NO₂ emissions from all power

IMPLICATIONS

The results presented here demonstrate that significant fluxes of NO can be generated during winter fallow periods when crop residue is mixed back into the soil, a common agricultural practice. The data also demonstrates that successful modeling of NO flux from agricultural soils must take into account the four soil parameters that influence soil NO emissions: soil temperature, water content (percent water-filled pore space), available nitrogen, and microbial activity. An over-reliance on predictive models based on a single soil parameter (e.g., soil temperature) may fail to provide an accurate assessment of NO flux from soils.

utilities in the state of Tennessee during the summer months. Anderson and Levine¹¹ reported NO flux values ranging from 0.2 to 67 ng-N m⁻² s⁻¹ for an agricultural site in Virginia, which is similar to the range in NO flux observed by Aneja et al.⁷ for agricultural soils in North Carolina. Kim et al.¹³ reported that NO emissions from an abandoned pasture site in North Carolina (0.1 to 6.7 ng-N m⁻² s⁻¹) were substantially less than from agricultural soils. Using an empirical model of soil-biogenic NO_x emissions, Yienger and Levy¹⁴ and Aneja and Robarge¹⁵ have estimated that agricultural systems account for about 41% of the current annual budget for soil-derived NO. With continued usage of N fertilizer, Yienger and Levy¹⁴ have calculated that global soil-biogenic NO_x emissions will approach 6.9 T_g N with agricultural soils accounting for more than 50% of the global source by the year 2025. These estimates are consistent with Galloway's et al.¹⁶ projections that the anthropogenic N-fixation rate will increase by 60% by the year 2020, primarily due to a combination of fossil-fuel combustion and fertilizer use.

Typically, studies dealing with NO flux from agricultural soils focus on emissions during the growing season, especially immediately following the application of nitrogen fertilizers when emissions are thought to be at their greatest.⁹ Less emphasis has been given to the winter fallow period, when the stubble from the prior crop has been incorporated back into the soil. Since the remaining crop residue after harvest (including the roots) represents a substantial portion of the nitrogen initially applied as fertilizer, the common agricultural practice of incorporation of this material into the soil represents a potential significant source of nitrogen for soil NO emissions. Anderson and Levine¹¹ estimated that 24% of the annual flux of NO from an agricultural site in Virginia occurred between November and April. In the southeastern United States, the majority of the fallow agricultural soils are not frozen or covered by snow during the winter months, thus favoring the emissions of trace gases. Moreover, it may be hypothesized that the net NO emissions from fallow fields may be higher than from fields with fully grown canopies. Due to the longer lifetime in winter, the NO emissions may be as important for the ozone formation as the NO emissions in summer.

In this paper we present NO and NO₂ flux measurements made in late winter/early spring 1994, herein referred to as "winter," from a soil type (Norfolk silt loam) with characteristics of agricultural soils in the Upper Coastal Plain region of North Carolina. The measurements were made on fields containing different crop residues that had been incorporated the previous fall. A winter cover crop was also present, which is in keeping with common agricultural practice in the southeast United States.

METHODS AND MATERIALS

Sampling Site

Flux measurements were made from February 7 to March 23, 1994, in three general crop (non-irrigated) fields at the Central Crops Field Laboratory (105 m MSL) in Clayton, NC. The field laboratory is owned by the North Carolina Agricultural Research Service and is operated by North Carolina State University. It is located approximately 10 km east-southeast of Raleigh, in the Upper Coastal Plain of North Carolina. The dominant soil type in each of the fields sampled is Norfolk sandy loam (fine-loamy, siliceous, thermic typic paleudult).¹⁷

Following harvesting in the fall (September-October) of 1993, the remaining crop stubble (soybean, *Glycine max*; cotton, *Gossypium sp.*; or corn, *Zea mays*) was leveled with a flail mower and the residue disked into the top 15 cm of soil. Approximately one month later, winter wheat (*Triticum sp.*) was planted as a cover crop. The wheat was approximately 5 cm in height during the actual sampling period. For the purposes of brevity, each field sampled will be referred to according to the previous crop (i.e., soybean field, cotton field, or corn field).

Flux Measurements

A dynamic chamber technique was used to measure NO_x flux from the soil.⁷ The dynamic chamber is an open bottom Teflon-lined cylinder (diameter ~27 cm, height ~42 cm, and volume ~25 l) held in place by a stainless steel frame driven into the ground to a depth of ~10 cm. Ambient air is pumped through the chamber at a constant flow rate ($Q = 9$ l p.m.), and the air in the chamber is well mixed by a motor-driven Teflon stirrer (~20 cm diameter, 120 r.p.m.). Air samples were collected after reaching steady-state conditions (~30 min of operation) at both ports of the chamber using Teflon bags (~10 l). The collection period was typically ~5 min. The air samples in these bags were then immediately analyzed (<3 min) for their NO and NO₂ concentrations.

Each day ($n = 5$ days per field), the chamber was positioned in the center of a different 15-m × 15-m plot that was chosen at random from a grid of 15 such plots constructed in each field. Hourly samples (normally 0600 to 2000 Eastern Standard Time) were taken using FEP Teflon bags at the inlet and outlet of the chamber. Analysis of the NO and NO₂ concentrations in the samples was carried out using a TECO 42S chemiluminescent high sensitivity NO analyzer (Thermo Environmental Instruments, Inc.),¹⁸ and a LMA-3 Luminol-based NO₂ analyzer (Scintrex, Ltd.)¹⁹ as discussed in Kim et al.¹³

The flux of NO or NO₂ was calculated using a mass balance equation with appropriate loss terms to account for reactions within the chamber and sample bags.^{7,13} The mass balance for NO in the chamber is given by

$$\frac{dC}{dt} = \frac{Q [C]_o}{V} + \frac{JA}{V} - \frac{LA [C]_f}{V} + \frac{Q [C]_f}{V} + F \quad (1)$$

where

- A = soil surface area covered by the chamber
- V = volume of the chamber
- Q = flow rate through the chamber
- J = emission flux per unit area
- C = NO concentration in the chamber
- [C]_o = NO concentration at the inlet of chamber
- [C]_f = NO concentration at the outlet of chamber
- L = loss term by chamber wall per unit area assumed first order in [C]
- R = chemical production/destruction rate in the chamber

For a well-mixed chamber, [C]_f may be assumed to be equal to the NO concentration in the chamber.

Soil Analysis

Soil temperature (5 cm depth) was measured using a thermocouple probe inserted into the soil adjacent to the dynamic chamber. Air temperature within the chamber was measured by inserting a thermocouple probe into the chamber outlet immediately after removing the Teflon sample bag. Soil samples were obtained from the center of the chamber placement footprint using a bucket auger (0-20 cm depth) at the end of each measurement period. Total soil water content was calculated as (initial weight-oven dry [105 °C weight])/oven dry weight, and used to calculate percent water-filled pore space.²⁰ Extractable ammonium and nitrate in a 2M KCl extract²¹ was determined using standard autoanalyzer techniques²² (Lachat Instruments, 1990). Additional soil physical and chemical properties are described in Aneja et al.⁷

RESULTS AND DISCUSSION

Soil temperature ranged from 0.9 to 21.0 °C (Table 1) during the measurement period (February 7 to March 23, 1994). Highest soil temperatures were recorded in mid-afternoon (1500 EST), and the lowest recorded soil temperatures were usually just before sunrise (600 EST). Change in soil temperature during the

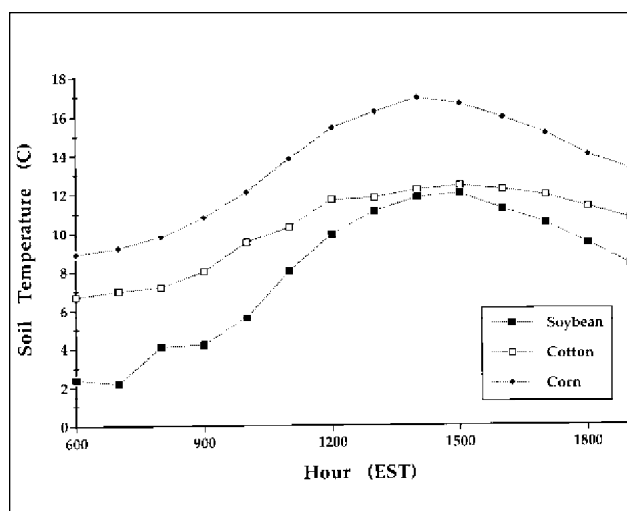


Figure 1. Daily variation in mean (n = 5 days) soil temperature (5 cm depth). Legend indicates type of crop residue incorporated into the soil the previous fall.

day was greatest for measurements made in the soybean field, and the least for the cotton field (Figure 1). Temporal trends in the air temperature within the dynamic chamber were similar to soil temperature, with the mean air temperature approximately 4 °C warmer than the soil temperature (Table 1). Largest differences between air temperature within the dynamic chamber and soil temperature adjacent to the chamber were observed in mid-afternoon.

Table 1. Summary of NO and NO₂ flux measurements and site variables.

| Crop Field | | Soil Temp* (C) | Air Temp† (C) | Total Extractable Nitrogen‡ | % WFPS @ | Air Carrier | |
|------------|-----------|----------------|---------------|-----------------------------|----------|-------------|------------------------|
| | | | | | | NO Flux** | NO ₂ Flux** |
| Soybean | Average | 8.3 | 12.2 | 4.1 | 47.2 | 9.2 | -14.1 |
| | Std. Dev. | 3.9 | 5.6 | 0.9 | 3.2 | 14.6 | 26.3 |
| | Min. | 0.9 | 0.0 | 3.1 | 43.1 | -4.2 | -141.0 |
| | Max. | 14.2 | 23.9 | 5.3 | 51.5 | 75.9 | 38.5 |
| Cotton | Average | 10.3 | 13.2 | 6.5 | 55.0 | 6.1 | -6.0 |
| | Std. Dev. | 5.6 | 9.0 | 3.4 | 6.1 | 13.8 | 11.4 |
| | Min. | 3.3 | 1.7 | 3.2 | 48.0 | -11.7 | -63.5 |
| | Max. | 21.0 | 31.5 | 10.3 | 62.2 | 109.6 | 15.1 |
| Corn | Average | 13.4 | 17.0 | 4.5 | 34.1 | 4.7 | -8.1 |
| | Std. Dev. | 4.3 | 9.2 | 0.4 | 7.2 | 5.5 | 11.2 |
| | Min. | 3.3 | -1.5 | 4.2 | 26.0 | -0.2 | -64.6 |
| | Max. | 20.7 | 32.2 | 5.2 | 43.4 | 40.4 | 25.3 |

* = Soil temperature measured at 5 cm depth adjacent to chamber

† = Air temperature measured inside the chamber

‡ = Units are mg-N/kg

@ = Percent Water-Filled Pore Space (WFPS)

** = Units are ng-N m⁻² s⁻¹

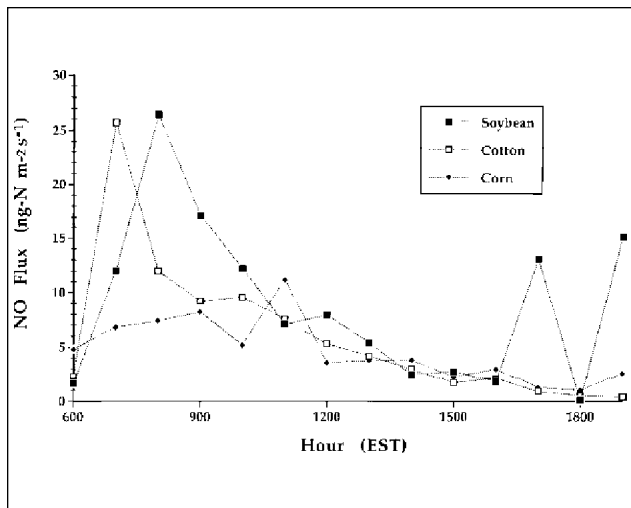


Figure 2. Daily variation in mean ($n = 5$ days) NO flux. Legend indicates type of crop residue incorporated into the soil the previous fall.

Percent water-filled pore space varied from $47.2 \pm 3.2\%$ in the soybean field to $54.9 \pm 6.1\%$ in the cotton field and $34.1 \pm 7.2\%$ in the corn field. This range in percent water-filled pore space is considered optimum for NO production from soils,²⁰ although the corn field was actually somewhat drier than the other fields during the measurement period. This may account in part for the higher soil temperatures observed for the corn field (Figure 1). Total extractable nitrogen (ammonium plus nitrate) was relatively constant within and between the three fields (Table 1), which is consistent with the lack of a fall application of nitrogen fertilizer and the presence of the winter wheat cover crop.

Highest calculated fluxes for NO were during the morning hours, with pronounced peaks in NO emissions occurring in both the soybean and cotton fields (Figure 2). The temporal trend depicted in Figure 2 was consistent for each field for each measurement period ($n = 5$ days per field); only the magnitude in peak NO emissions varied from day to day. Averaged over time, the NO flux for the soybean field was $9.2 \pm 14.6 \text{ ng-N m}^{-2} \text{ s}^{-1}$, compared to $6.1 \pm 13.8 \text{ ng-N m}^{-2} \text{ s}^{-1}$ for the cotton field and $4.7 \pm 5.5 \text{ ng-N m}^{-2} \text{ s}^{-1}$ for the corn field (Table 1). Seven of the 199 data points (3.5%) calculated suggested NO deposition. Some peaks in NO flux were also observed during the evening hours for the soybean residue. We currently have no explanation for these peaks.

The tendency for NO flux to decrease during the day (Figure 2) resulted in a negative trend for NO flux with increase in soil temperature (Figure 3). The strength of this relationship varied between the fields (soybean, $R^2 = 0.37$; cotton, $R^2 = 0.13$; and corn, $R^2 = 0.16$), but the consistency in the data illustrates that an NO emission precursor became limiting during the day with increase in soil temperature.

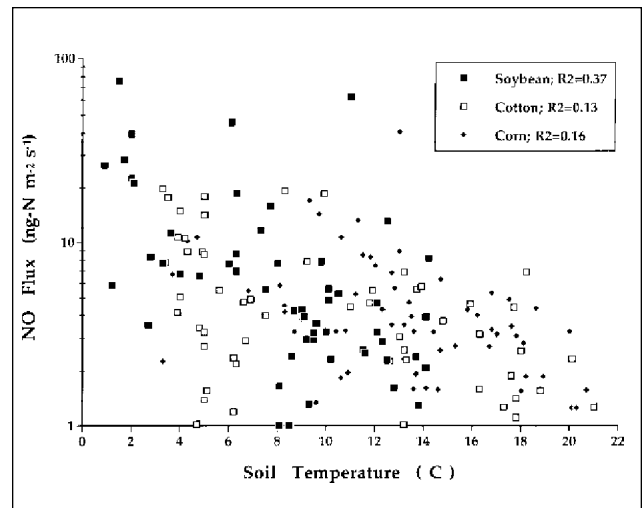


Figure 3. NO flux versus soil temperature (5 cm depth). Legend indicates type of crop residue incorporated into the soil the previous fall. The R^2 term is the coefficient of determination for the regression of NO flux versus soil temperature.

The majority of the data points calculated for NO₂ flux (92%) were negative in value, indicating deposition of NO₂ (Figure 4). Temporal trends in the composite data suggest highest deposition occurred for soybean and cotton fields during the morning hours (Figure 4), but the trend is less evident than for NO emissions (Figure 2). Averaged over time, the NO₂ flux for the soybean field was $-14.1 \pm 26.3 \text{ ng-N m}^{-2} \text{ s}^{-1}$, compared to $-6.0 \pm 11.4 \text{ ng-N m}^{-2} \text{ s}^{-1}$ for the cotton field and $-8.1 \pm 11.2 \text{ ng-N m}^{-2} \text{ s}^{-1}$ for the corn field (Table 1).

February and March are not considered high ozone months in North Carolina such that the calculated NO fluxes should be representative of NO emissions from intensively managed agricultural fields in the southeast United States during the winter fallow period. The early

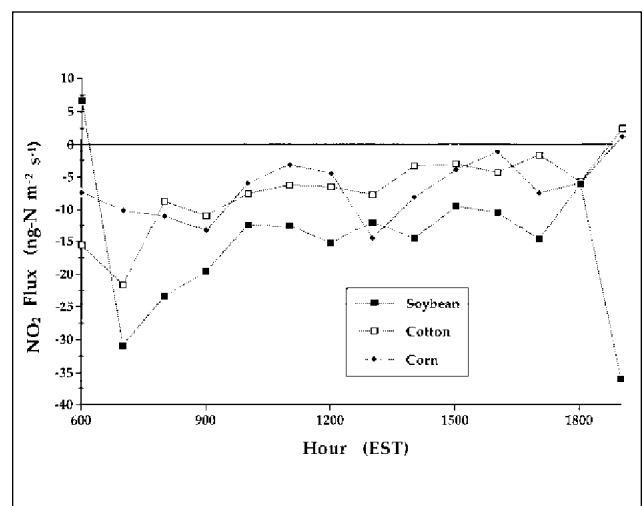


Figure 4. Daily variation in mean ($n = 5$ days) NO₂ flux. Legend indicates type of crop residue incorporated into the soil the previous fall.

morning peaks in NO emissions observed for the cotton and soybean fields could not be ascribed to soil disturbance with insertion of the chamber into the soil. The chamber was installed the evening prior to the next day's sampling, allowing a minimum of 10 hours between insertion of the chamber into the soil and the start of measurements. Furthermore, the pronounced peak in NO emissions (Figure 2) was not observed for the field with the incorporated corn stubble. We prefer to accept the temporal trends in NO flux shown in Figure 2 as an illustration of the competition between the various soil parameters that influence NO emissions from soils.

The soil parameters that influence NO_x emissions are known and include temperature,^{11,13,23,24} water content,^{18,25-27} available nitrogen,^{9,28,29} and microbial activity.^{20,30} The interaction of these soil parameters, which resulted in the trends in soil NO emissions shown in Figure 2, can probably best be described using the concept of the NO compensation concentration. Conrad³¹ has defined the compensation concentration as the concentration at which the rate of production of NO equals the rate of consumption (net soil NO flux equal to zero). The decrease in NO flux with increase in soil temperature (Figure 3) indicates that the rate of NO consumption processes approaches NO production. Both NO production and consumption have been observed in a range of soil types and different soil horizons (Baumgärtner and Conrad),²⁵ with observed NO compensation mixing ratios ranging from 3 to >1,400 ppbv NO. However, Baumgärtner and Conrad²⁵ noted that the majority of compensation points they observed were less than 50 ppbv NO. Field observations by Slemr and Seiler²⁹ suggest NO compensation mixing ratios may be as low as 0 to 5.5 ppbv NO.

Skiba et al.⁹ noted a reversal of the normal diurnal variation in NO emissions for soil NO flux values <0.15 ng-N m⁻² s⁻¹ on rye grass plots (clay loam soil) and <2 ng-N m⁻² s⁻¹ on winter wheat plots (sandy loam soil). NO deposition was recorded on the unfertilized rye grass plots during the day as soil temperature increased from 4 to 16 °C. Maximum NO deposition appeared to coincide with maximum soil temperature. Baumgärtner and Conrad²⁸ also noted a positive correlation between NO uptake by soil cores and temperature, and concluded that NO deposition to soils is controlled solely by the activity of soil microorganisms. Denitrification reactions are known to consume NO₂³² and may have been responsible for NO consumption at our site even though percent water-filled pore space was <55%. The presence of the crop residue may have enhanced the formation of microsites within the surface soil that were anaerobic, and thus potential consumers of NO, especially with an increase in soil temperature.²⁵ Our data suggests that the rate for the NO consumption reactions increased steadily

during the day beginning after sunrise, and became approximately equal to the NO production rate by sunset (Figure 2).

The results presented here demonstrate that significant fluxes of NO can be generated during winter fallow periods when crop residue is mixed back into the soil, a common agricultural practice. Similar results involving incorporating plant residues back into the soil have been reported by Slemr and Seiler.¹² Our average NO flux values are also similar in magnitude to those reported by Valente and Thornton (Figure 7)⁸ for a fallow corn field in central Tennessee, and by Anderson and Levine (Table 3)¹¹ from an agricultural site in Virginia. The data also demonstrates that successful modeling of NO flux from agricultural soils must take into account the four soil parameters that influence soil NO emissions: soil temperature, water content (percent water-filled pore space), available nitrogen, and microbial activity. An over-reliance on predictive models based on a single soil parameter (e.g., soil temperature, Williams et al.)¹⁰ may fail to provide an accurate assessment of NO flux from soils.

ACKNOWLEDGMENTS

This research has been funded through a cooperative agreement with the U.S. Environmental Protection Agency (CR 822058-01) as part of the Characterization of Emissions of Nitrogen Oxides from the Soils of Southeast U.S. Project. We sincerely acknowledge George Clark and Jimmy Prince of the Central Crops Field Laboratory for providing the field logistical support; Dr. B. Gay, T. Pierce, and C. Geron of the EPA for the technical discussions; L. Sullivan and T.C. Moore for assistance during the experiment; and M. DeFeo in the preparation of the manuscript.

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