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Characterization of biogenic nitric oxide source strength in the southeast United States

Viney P. Aneja*, Paul A. Roelle and Wayne P. Robarge

North Carolina State University, Raleigh, NC 27695-8208, USA

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Abstract

Emissions of nitric oxide (NO) were measured during the summer of 1995 from 4 crops, located at three different sites throughout North Carolina. These sites were chosen to represent major physiographic regions of the Southeast US, in an effort to compare fluxes from different agriculturally managed soils. Emission rates were determined using a dynamic flow-through chamber system. In order to understand the NO flux from the different soil and crop types, measurements were made on corn and soybean crops in the coastal region, tobacco in the piedmont region, and corn in the upper piedmont region of North Carolina. Average NO fluxes were 5.5 ± 2.2 ng N m^{-2} s^{-1}, 20.7 ± 19.2 ng N m^{-2} s^{-1}, 4.1 ± 1.4 ng N m^{-2} s^{-1}, and 8.5 ± 4.9 ng N m^{-2} s^{-1} respectively for corn and soybean in the coastal region, tobacco in the piedmont region, and corn in the upper piedmont region. We were only able to detect an exponential dependence of NO flux on soil temperature at two of the locations. The composite data of all the research sites revealed a general trend of increasing NO flux with soil water content or increasing extractable nitrogen in the soil, however, the day to day variations within each site did not reveal the same trends. We feel that acquisition of a soil NO flux data set in this fashion, which consists of observations collected over different points in both space and time, makes attempts to model soil NO flux in terms of different soil parameters difficult.

Keywords: Natural emissions, nitric oxide, corn, soybean, tobacco, agricultural soils, soil moisture content, soil extractable nitrogen

Introduction

Ozone photochemistry in the troposphere is regulated by oxides of nitrogen (NO_X = NO + NO_2). Currently, the only known pathway for the production of ozone is the photolysis of NO_2 (NO_2 + hv → NO + $O(^3P)$), which further reacts with O_2 to produce ozone (O_3) by the reaction $O(^3P)$ + O_2 → O_3. In a pseudo-photostationary environment, the O_3 produced would react with the NO that was generated via the photolysis of NO_2 in the following reaction: NO + O_3 → NO_2 + O_2. Hence, there is no net production of O_3. However, in the real atmosphere, hydroxyl radicals combine with volatile organic compounds (VOC's) to produce new peroxy radicals which preferentially react with NO to produce NO_2, allowing a net O_3 accumulation. Regions such as the Southeast US are classified as NO_x limited and increased emissions of NO into the troposphere are likely to produce increased O_3 concentrations (SOS, 1993; Aneja et al., 1996b). In a region which currently maintains 40% of the ozone non-attainment areas of the US, the southeast must develop a better understanding of these ozone precursors in order to more successfully develop control strategies for the emissions of ozone precursors.

The strongest sources of NO_x are known to be anthropogenic and located in confined geographical areas, such as the combustion of fossil fuels from power plants and automotive exhaust (Logan, 1983; Hameed and Dignon, 1988; Levy and Moxim, 1989). Being confined to known areas, these anthropogenic sources can be well quantified and modeled. In contrast, biogenic emissions are not as clearly understood. Recent studies have estimated that soil emissions can be 50% of the NO_x budget in remote agricultural areas of the U.S., and even exceed 75% of the NO_x budget during certain months of the year (Yienger and Levy, 1995; Aneja and Robarge, 1996).

* Corresponding author. Tel.: +1-919-515-7808; fax: +1-919-515-7802; e-mail: viney_aneja@ncsu.edu

Researchers have found that obtaining accurate estimates by computer modeling is difficult to achieve due to the extremely high variability of biogenic soil NO emissions. Seemingly, homogeneous soils have been shown to differ by more than a factor of 10 between adjacent sites (Williams and Fehsenfeld, 1991). Although the apparent spatial variability of soil NO emissions would appear to limit attempts to model soil NO flux, there are some environmental parameters which have been shown to have a reasonably consistent relationship with NO flux. These include soil temperature, soil water content and the nitrogen content of the soil (Anderson and Levine, 1987; Davidson, 1992; Slemr and Seiler, 1991; Hutchison and Brams, 1992; Sullivan et al., 1996; Aneja et al., 1996a).

The objective of this study was to measure NO emissions from several different physiographic regions in the southeast US, in an attempt to relate NO flux to different physical and chemical properties of soil. Further, these relationships could then be extrapolated to similar physiographic regions of the Southeast US to better model emissions of NO from biogenic processes within soils.

Methods and materials

Sampling sites and crop characterizations

NO flux measurements were made on three different crop types (corn, soybean, tobacco) at three different research sites during Summer of 1995. The research sites were located in Lenoir County, NC, approximately 5 km northeast of Kinston, NC; Granville County, NC, approximately 5 km west of Oxford, NC; and Rockingham County, NC, approximately 10 km southwest of Reidsville, NC. All three of these sites were operated by the North Carolina Department of Agriculture using management practices typical for their respective crops and physiographic locations. The agricultural fields at the Kinston, NC research facility are classified as Rains fine sandy loam. The dominant soil types of the Oxford, NC and Reidsville, NC research sites were classified as Vance sandy loam and Pacolet sandy loam respectively. The location of these research sites can be seen in Fig. 1.

NO flux measurements at the Kinston, NC research site were conducted on both corn and soybean fields. The corn crop was planted in early April, 1995 and the seed was drilled directly into soybean stubble (no-till planting) from the preceding year. The corn crop received a total of 190 kg N per hectare. The corn crop, which was in a mature growth stage, had fully developed ears and had reached an approximate height of 183 cm. The soybean crop at Kinston was planted during the first week of June, 1995. The soybean seed was planted directly into the residue of wheat (no-till planting), which was harvested a few days prior to the soybeans being planted. Although the wheat crop received approximately 157 kg N per hectare during the month of February, 1995, the soybean crop did not receive any N fertilizer. The soybean crop, which was in a vegetative growth stage, was approximately 10 cm tall at the beginning of our measurement period and grew to a height of approximately 25 cm at the conclusion of our measurement period.

The research conducted in Oxford, NC was on a tobacco crop. The tobacco plants were initially grown in tobacco plant beds and transplanted to the growing field in early May, 1995, when the individual plants were approximately 13 cm tall. The tobacco plants were planted in raised beds, which were spaced approximately 1 meter apart. After transplanting, the tobacco crop received a total of 70 kg N per hectare. During our measurement period, the tobacco crop was in a mature growth stage, reaching an approximate height of 145 cm. Leaves from these tobacco plants were being harvested throughout our measurement period.

The research conducted in Reidsville, NC was on a corn crop that was planted in early April, 1995. The corn seed was drilled directly into stubble (no-till planting),

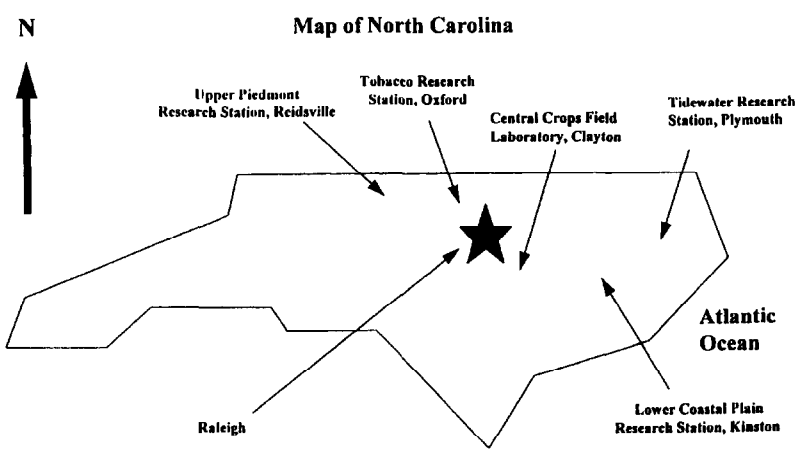

Fig. 1. Location of NO flux measurement sites in various physiographic regions of North Carolina.

from corn planted the preceding year, and received a total of 197 kg N per hectare. This crop was in a mature growth stage, and had reached an approximate height of 289 cm, which was attributed to the large amounts of rain during the early part of the growing season. Portions of the crop were being harvested during our measurement period.

Sampling scheme

The sampling scheme of the multi-site experiment was to measure concentrations of NO at ground level at three distinctively different physiographic locations in North Carolina. Measurements were conducted for a minimum of four days at each site with at least one continuous 24-hour experiment conducted at each location.

The measurement campaign began in Kinston, NC from June 30 until July 13, 1995. Typically only one crop was measured at each of the sites, however this site had two different crops planted side-by-side facilitating NO flux measurements on both corn and soybean, with one week being spent at each crop. Oxford, NC was the second site of the measurement campaign, with NO flux measurements made on a tobacco crop from July 20 until July 27, 1995. The measurement campaign concluded in Reidsville, NC with NO flux measurements made on a corn crop from August 1 until August 10, 1995.

Soil NO flux was measured on a daily basis using a dynamic flow-through chamber technique positioned over bare soil. Ambient air was used as the carrier gas in the chamber. Figure 2 is a schematic diagram of the dynamic flow-through chamber system utilizing ambient air as the carrier gas.

NO concentrations within the chamber were measured every 15 min, usually from 9:00 a.m. until 4:00 p.m. The chamber was flushed with zero grade air for approximately 45 min prior to the first measurement to allow the system to reach a steady state. The stainless-steel collar was relocated in the evening after each experiment, which attempted to remove a potential bias from soil NO flux due to soil disturbance generated by insertion of the stainless-steel collar into the soil.

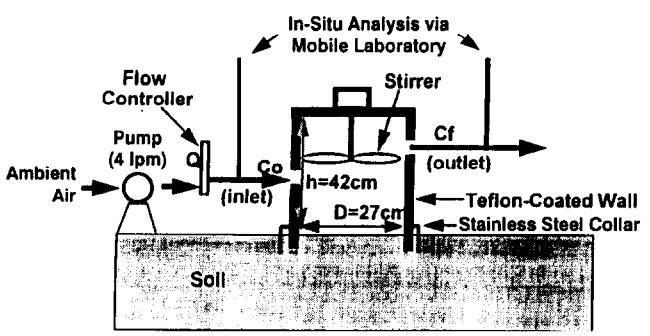

Fig. 2. A schematic of the dynamic flow-through chamber used for measuring the flux of NO from soils. The walls and all internal surfaces are fluorinated ethylene propylene (FEP) Teflon.

Flux calculation

The NO flux was calculated from a mass balance equation (Kaplan et al., 1988; Kim et al., 1994). The mass balance equation is:

$$\frac{dC}{dt}=\left(\frac{q[C_{air}]}{V}+\frac{J}{h}\right)-\left(\frac{L}{h}+\frac{q}{V}\right)[C] \quad (1)$$

where h is the internal height of the chamber, J the emission flux per unit area, L the loss by chamber wall per unit area assumed first order in [NO], q the flow rate through the chamber, V the volume of the chamber, C the NO concentration in the chamber, and C_{air} the NO concentration in the stream entering the chamber

Assuming the chamber is well mixed, the concentration [C] measured can be assumed to be the same everywhere within the chamber. Additionally, at steady-state conditions, the change of concentration, with respect to time, will be zero. Equation (1) reduces to:

$$\frac{J}{h}=\left(\frac{L}{h}+\frac{q}{V}\right)C_{eq}-\frac{qC_{air}}{V} \quad (2)$$

(C_{eq}) is the concentration measured at the outlet of the chamber. During most of the measurements, the NO concentrations in the ambient air (C_{air}) adjacent to the chamber were less than 1 ppbV.

In eq. (2), the total loss term (L) is the sum of the loss of NO through reactions with the chamber walls and chemical reactions of NO with existing oxidants in the carrier gas, such as ozone and peroxy radicals (Kim et al., 1994; Aneja et al., 1995). The total loss term was determined empirically utilizing a method developed by Kaplan et al. (1988). This method plots the value of $-\ln\frac{C_{eq}-C}{C_{eq}-C_0}$ against time (t). C_0 is the NO concentration in te chamber when NO reaches the first equilibrium state at an initial flow rate. C_{eq} is the NO concentration in the chamber after the flow rate is reduced and allowed to reach a second equilibrium. From the linear relationship between the value of $-\ln\frac{C_{eq}-C}{C_{eq}-C_0}$ and time during the experiment, the slope is found to represent $\left(\frac{L}{h}+\frac{q}{V}\right)$. The values of L/h agree with those found by Kim et al. (1994), and are used in eq. (2) to calculate the NO flux during the experimental period.

Temperature and soil analysis

Soil and air temperature were recorded every 15 min in conjunction with NO flux measurements. Soil and air temperatures were recorded and stored in a laptop computer using temperature probes. The soil temperature probe was inserted 5 cm into the soil, adjacent to the

chamber, while the air temperature probe was positioned at a height of 1 m, shielded from direct sunlight.

A soil sample was taken from the center of the dynamic flow-through chamber footprint at the end of each experimental period (approx. 1 sample per day). Samples were taken with a bucket auger which removed the top 20 cm of soil. Soil parameters, such as total extractable nitrogen, moisture content, and pH for the various research sites were determined for each soil sample collected. Nitrate (NO_3^-) and ammonium (NH_4^+) in a 2 M KCL soil extract (Keeney and Nelson, 1982) were determined using standard autoanalyzer techniques (Lachat Instruments, 1990).

Results and discussion

NO Flux

The intersite comparison revealed that measured soil NO flux was heavily dependent on location and crop type (Table 1). The highest average NO flux, 20.7 ± 19.2 ng N m^{-2} s^{-1}, occurred from a soybean field in Kinston, NC, although it should be noted that the average NO flux from this soybean crop would have been 12.8 ± 5.2 ng N m^{-2} s^{-1} if we neglected one day of measurements after a rain event. We observed the largest amount of NO flux from the soybean crop, unlike what has typically been observed by other researchers (Aneja et al., 1995; Sullivan et al., 1996). However, the different sampling periods of the various crops should be taken into consideration when trying to account for this apparent anomaly. We were sampling the soybean crop within 4 weeks of the seed being drilled directly into the wheat stubble. This method of planting left the root system from the previous wheat crop undisturbed. The subsequent decomposition of the wheat roots may have provided an abundant nitrogen source explaining the higher than expected NO flux values.

The diurnal variation in which NO flux increases in the afternoon, coinciding with the rise in soil temperature throughout the afternoon, can be seen in Fig. 3, which shows the average NO flux (6:00 a.m. to 6:00 p.m.) for each of the crops sampled. Figure 3 also reveals a morning peak of NO emissions, between 6:00 a.m. and 10:00 a.m. for soybean. Similar result were observed by Holbrook (1994), and the hypothesis which was proposed was that the roots of the plants exude organic compounds during the morning hours. These organic compounds are then utilized by denitrifying bacteria which reduce NO_3^- generating emissions of NO greater than would be predicted by soil temperature.

The plot for soybean in Fig. 3 contains three of the four sample days, because the NO flux which we recorded on 11 July 1995 was an order of magnitude larger than the rest of the sampling days, and we felt this was unrepresentative of average NO emissions at this site. The increased NO flux for this particular day, evident by the change in scale of the *y*-axis in Fig. 4 can be

Table 1

Data Summary for Kinston, Oxford, and Reidsville, NC (Summer 1995). All NO flux data and soil data were calculated from the 15 minute binned averages. The total extractable N and % soil moisture content were calculated from the soil sample collected from the center of the chamber footprint at the end of each measurement period.

		Soil temp. (°C)	Total Extractable N (mg N kg dry $soil^{-1}$)	% Soil moisture (dry wt.)	NO Flux (ng N m^{-2} s^{-1})
Site: Kinston, NC	Average	24.8	10	12.6	5.5
Crop: Corn	Standard Deviation	2.4	2	1.5	2.2
Growth Stage: Maturity	Minimum	19.3	8	10.7	3.0
Date: June 30–July 5, 1995	Maximum	32.5	12	14.5	14.4
Site: Kinston, NC	Average	25.7	14	12.8	20.7
Crop: Soybean	Standard Deviation	3.0	3	0.8	19.2
Growth Stage: Vegetative	Minimum	21.5	11	11.6	6.1
Date: July 10–July 14, 1995	Maximum	31.9	19	13.4	80.0
Site: Oxford, NC	Average	27.3	8	5.6	4.1
Crop: Tobacco	Standard Deviation	2.1	2	2.3	1.4
Growth Stage: Maturity	Minimum	23.5	6	2.7	1.7
Date: July 20–July 27, 1995	Maximum	32.5	13	8.1	8.0
Site: Reidsville, NC	Average	22.6	13	11.3	8.5
Crop: Corn	Standard Deviation	2.3	12	2.4	4.9
Growth Stage: Maturity	Minimum	19.7	4	10.0	1.9
Date: August 2–August 11, 1995	Maximum	29.0	32	15.6	20.5

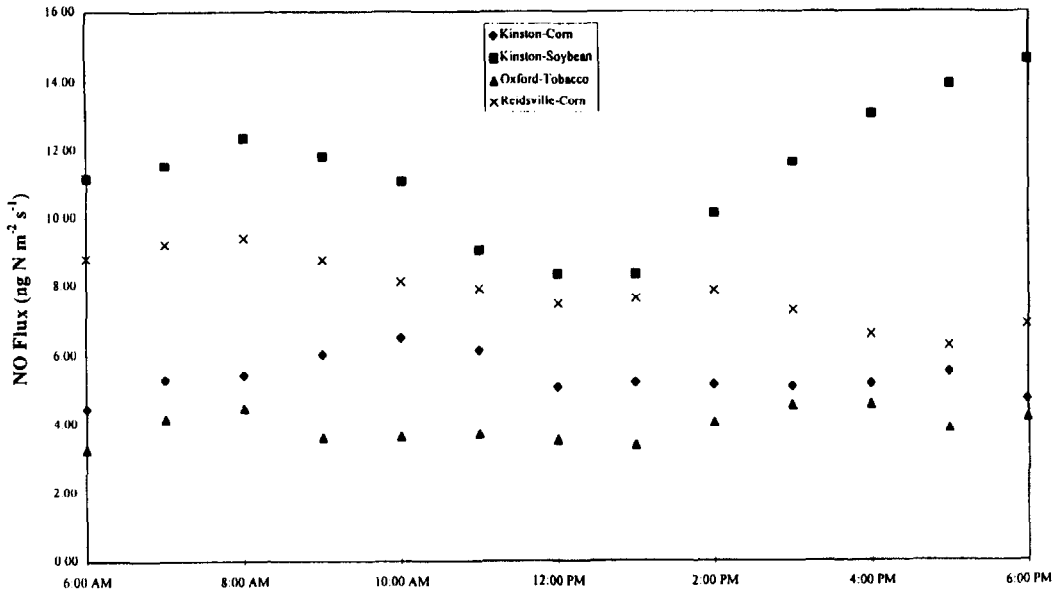

Fig. 3. The composite hourly averaged NO flux (6:00 a.m. to 6:00 p.m.) for the three research sites and the four crops. Note: The plot of soybean at Kinston does not contain the data from measurements conducted on the day following a thunderstorm.

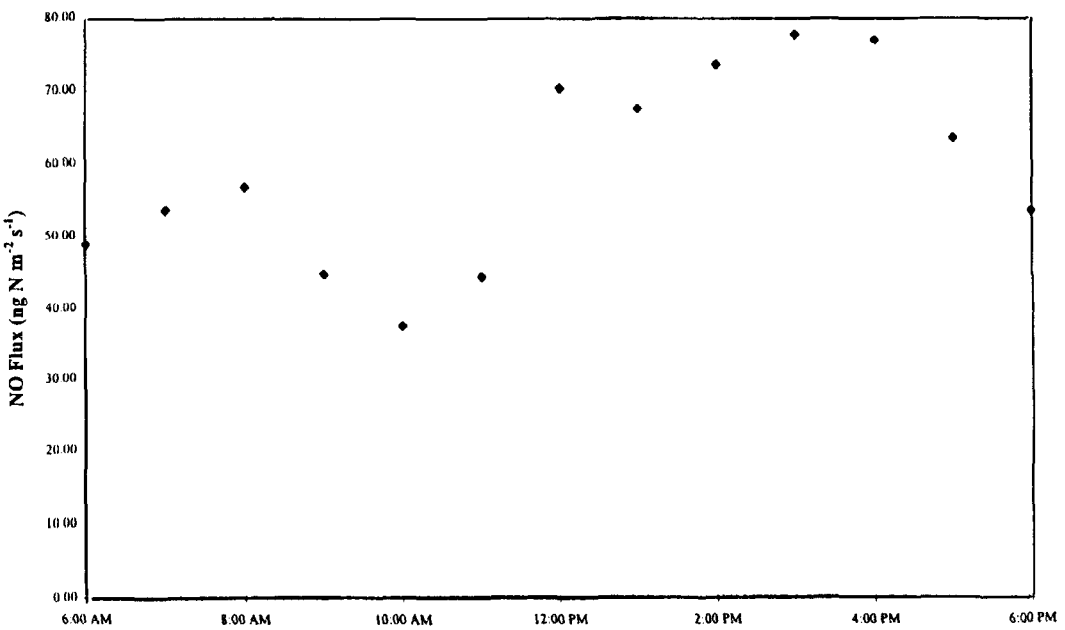

Fig. 4. Hourly averaged NO flux (6:00 a.m. to 6:00 p.m.) for the soybean crop in Kinston, NC, after a thunderstorm the previous evening.

attributed to rainshowers that occurred the previous evening, which is consistent with a process referred to as "pulsing" by Yienger and Levy (1995). These pulse fluxes, which occur as the result of a rain event, can be 10–100 times background values and have been observed to last for a few days to a few weeks (Stocker et al., 1993; Valente and Thornton, 1993; Williams et al., 1987; Williams and Fehsenfeld, 1991). The duration of the increased flux will vary depending on the moisture condition of the field prior to the rain event and the amount of rain. We observed one day of increased NO flux immediately after the rainshower with the NO flux returning to pre-rain event values on July 12, 1995.

Each crop received different amounts of N fertilizer (Kinston: corn, 190 kg N per hectare; Kinston: soybean, 0 kg N per hectare; Oxford: tobacco, 70 kg N per hectare; Reidsville: corn, 197 kg N per hectare). By taking a ratio of the average summertime flux of NO to the amount of total N fertilizer applied to each crop, we can estimate the amount of N fertilizer returned to the atmosphere, during the summer months, via NO flux. For each of the crops which were fertilized, the percentage of nitrogen applied as fertilizer which was returned to the atmosphere via NO flux was less than 1%.

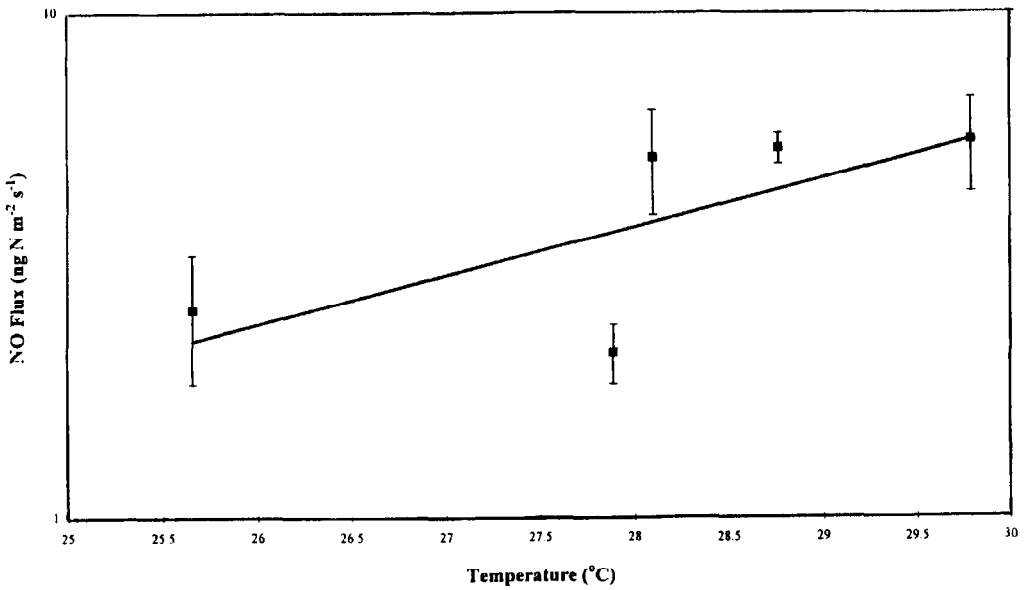

Fig. 5. Daily averaged NO flux versus daily averaged soil temperature (9:00 a.m. to 5:00 p.m.) plotted on a logarithmic scale. Vertical bars indicate one standard deviation of the average NO flux. This plot represents data collected from a tobacco crop in Oxford, NC.

Soil temperature

Strong relationships between NO flux and soil temperature, in which NO flux doubles for each 10°C increase in soil temperature, has been observed by several researchers (Williams et al., 1988; Williams and Fehsenfeld, 1991; Kim et al., 1994; Sullivan et al., 1996). Unlike most of these researchers who were sampling from fairly uniform and stable forest and grassland systems, our research was conducted from intensively managed agricultural soils. Although we occasionally detected this same relationship between soil temperature and NO flux, we were not surprised that our results were not consistent across the three different soil and crop types. Figure 5 is a graph of daily average NO flux, plotted on a log scale, versus daily average soil temperature (9:00 a.m. to 5:00 p.m.) at the tobacco crop located in Oxford, NC. This graph displays a relationship between soil temperature and NO flux that we detected in our multi-site experiment ($R^2 = 0.54$). A possible relationship between soil temperature and NO flux ($R^2 = 0.35$) was suggested for the soybean crop grown in Kinston, NC (excluding the day after the rain event). However, no NO flux dependence on soil temperature was observed for the two corn crops sampled at Kinston and Reidsville. These results from Kinston and Reidsville, which were sampled approximately two months after the last N fertilizer application, are consistent with some of the results of the data that we collected from a corn crop located in Plymouth, NC. Research at Plymouth, NC was segregated into two time periods, which were before and after the final application of N fertilizer. The Plymouth, NC site revealed no NO flux dependence on soil temperature until immediately after the field was fertilized. The fact that no relationship between soil temperature and NO flux was detectable until excess amounts of N were present in the top few centimeters of the soil surface suggests that NO flux is also being controlled by the application of N fertilizer.

Total extractable nitrogen and soil moisture

NH_4^+ and NO_3^- are two forms of nitrogen which are utilized by soil microbes and can lead to the release of NO gas in soils. The total extractable nitrogen ($NH_4^+ + NO_3^-$) present in the soil, therefore should give an indication of NO flux. Figure 6 is a graph of NO flux versus total extractable N for the different measurement sites. The general trend among all of the data points is an increase in NO flux as total extractable N increases, however within each crop type, the relationship is not as evident. This figure reveals, as other researchers have also reported, that a change in extractable nitrogen, by itself, does not lead to increased NO flux (Williams and Fehsenfeld, 1991; Cardenas et al., 1993; Sullivan et al., 1996).

Although we have seen NO emissions follow changes in soil temperature, and to some degree follow changes in extractable nitrogen content, the correlation's are further complicated by the interactions of soil moisture. Researchers have shown that biogenic NO emissions can take place over a wide range of soil moisture conditions as long as the soils are not stressed by a lack of water or are not water saturated (Slemr and Seiler, 1984; Anderson and Levine, 1987; Johansson et al., 1988). A model has been proposed by Davidson (1993) which shows no effect of soil moisture on NO flux, as long as the field is in an optimum range, meaning it is neither water stressed nor water logged. The optimum range of soil

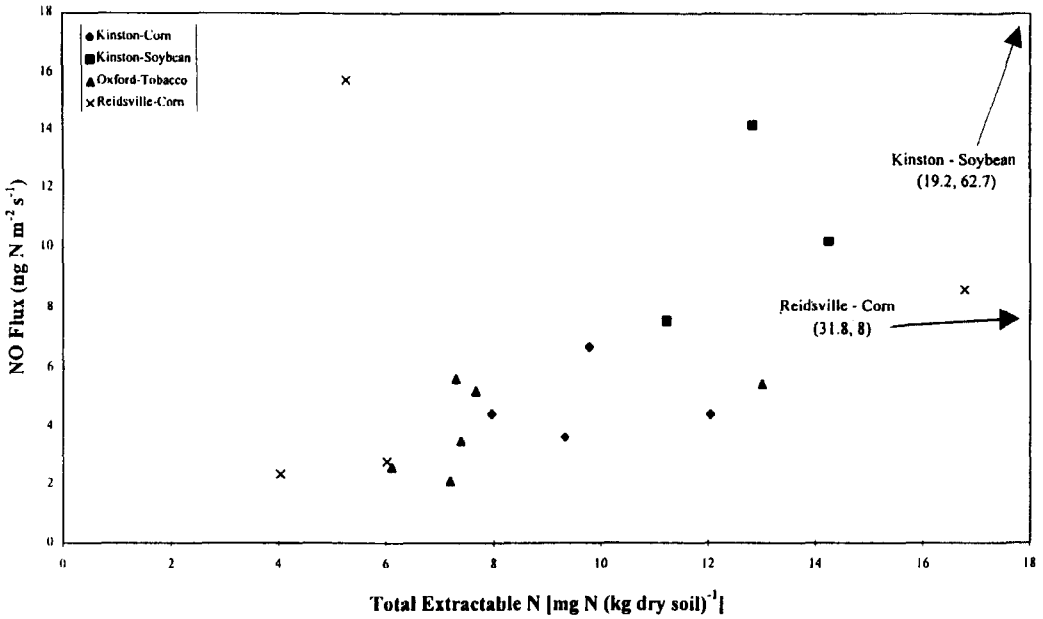

Fig. 6. Daily averaged NO flux (9:00 a.m. to 5:00 p.m.) versus total extractable N. The arrows indicate two data points located off the scale.

moisture for any given field will differ due to soil and crop type. The range of soil moisture (dry weight) that we observed from our multi-site experiment ranged from 2.9–15.7%, however our data does not specify the optimum range of soil moisture for the individual fields.

Data set bias

Sanchez et al. (1985) recognized two types of data sets for studying soil dynamics. A Type I data set is one in which changes in soil properties are monitored with time at the same site. An example of a Type I data set, although confined to a relatively short period of time, is the four-day measurement period at each of the individual research sites (Kinston, Oxford, Reidsville). A Type II data set is one in which several soils of nearby sites are sampled at the same time. Type I data, when conducted over extensive periods of time, best characterizes a soil system. However, obtaining this type of data is both costly and time consuming, and therefore not readily available. Type II data sets produce results quickly and are more readily available, however they suffer from the inherent problem that the initial conditions of the research site and the initial soil properties are unknown. Therefore, differences among the research sites may be the result of different initial soil properties, or crop management techniques, and not necessarily the current physical or chemical properties of the soil system. The data we collected, which would be classified as Type II data, came from physiographic regions in North Carolina which represent different crop and soil types. Although we know the dates of planting, and the amount of N fertilizer applied, we can not assume that the soils were all identical in their potential to produce NO during the measurement period used in this study.

The variability inherent in our measurement sites makes it difficult to see simple trends in the data. We were unable to detect some of the same relationships that other researchers have identified, at relatively stable ecosystems such as pastures and fallow fields, between NO flux and total extractable nitrogen, % soil moisture or soil temperature (Slemr and Seiler, 1984; Johansson and Granat, 1984; Williams et al., 1988; Kaplan et al., 1988; Williams and Fehsenfeld, 1991; Hutchison and Brams, 1992; Kim et al., 1994; Aneja et al., 1995). In order to see a simple two dimensional relationship between NO flux and total extractable nitrogen, % soil moisture or temperature, requires that the other variables be at optimum conditions for the production of NO, or not important in terms of NO production. The fact that we were unable to detect consistent trends between NO flux and the environmental variables suggests that agricultural soil systems are too dynamic to apply simple environmental dependence functions.

Conclusions and recommendations

NO flux from intensively managed agricultural soils displayed variations from one physiographic location to another. Relationships between environmental variables and NO flux were found to exist at some locations but no one relationship was consistent throughout all of the sites. The soybean crop yielded the highest NO flux, while corn yielded the next highest NO flux and tobacco produced the least NO flux. We feel this observation was a result of the sampling dates of our research. Unlike the corn and tobacco crops which were close to being harvested, the soybean was just recently planted in a

nitrogen-rich source of decaying wheat roots. Temperature seemed to be a fairly good indicator of NO flux at two of the sites (Kinston-soybean, Oxford-tobacco). We noticed an unusual peak in NO emissions in soybean which occurred during the morning hours.

This observational based study consisted of sampling during summer during the changing conditions of the crops growth cycle. Additionally, the sampling occurred from different crop types in physiographically different regions of the state of N.C. Sanchez et al. (1985) elucidated this problem when he described two different types of data sets used in studying soil dynamics. Future work should consist of controlled field experiments where some of these soil conditions can be controlled and remove some of the uncertainty inherent in studies conducted at different crops in space and time.

Acknowledgments

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