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**Short Term Responses of  
 Nitrogen Trace Gas Emissions  
 to Nitrogen Fertilization in Tropical Sugar Cane:  
 Variations due to Soils and Management Practices**

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**ABSTRACT.** Nitrogen (N) fertilization of agricultural systems is thought to be a major source of the increase in atmospheric N<sub>2</sub>O; NO emissions from soils have also been shown to increase due to N fertilization. While N fertilizer use is increasing rapidly in the developing world and in the tropics, nearly all of our information on gas emissions is derived from studies of temperate zone agriculture. Using chambers, we measured fluxes of N<sub>2</sub>O and NO following urea fertilization in tropical sugar cane systems growing on a variety of soil types in the Hawaiian Islands, USA. On the island of Maui, where urea is applied in irrigation lines and soils are mollisols and inceptisols, N<sub>2</sub>O fluxes were elevated for a week or less following fertilization; maximum average fluxes were typically less than 30 ng cm<sup>-2</sup> h<sup>-1</sup>. NO fluxes were often an order of magnitude less than N<sub>2</sub>O. Together, N<sub>2</sub>O and NO represented from 0.01 - 0.5% of the applied N. In fields on the island of Hawaii, where urea is broadcast on the surface and soils are andisols, N<sub>2</sub>O fluxes were similar in magnitude to Maui but remained elevated for much longer periods after fertilization. NO emissions were 2-5 times higher than N<sub>2</sub>O through most of the sampling periods. Together the gases losses represented approximately 1.1 - 3% of the applied N. Laboratory studies indicate that denitrification is a critical source of N<sub>2</sub>O in Maui, but that nitrification is more important in Hawaii. Experimental studies suggest that differences in the pattern of N<sub>2</sub>O/NO and the processes producing them are a result of both carbon availability and placement of fertilizer, and that the more information-intensive fertilizer management practice results in lower emissions.

## Introduction

Application of fertilizer nitrogen (N) has increased rapidly in the last several decades. In 1970, approximately 32 Tg N were produced globally, with most of it applied in the developed countries of the temperate zone. In 1990, around 80 Tg were produced, and the application of that nitrogen was approximately evenly split between the more and less developed countries of the world (*FAO, 1990; Vitousek and Matson, 1993*); of this, approximately 40% was applied in the tropics or subtropics (*Matthews, 1994*). While fertilizer use in the developed world is leveling off and even declining in some regions, quantities and applications rates are expected to continue to increase in the developing world as food production attempts to keep pace with human population growth. Current projections suggest that 100 Tg/yr of N fertilizer will be applied in the developing world in 2025 in comparison with less than 50 in the now-developed countries of the world (*EPA, 1990*).

This intensification and expansion in fertilizer use may have consequences for a number of biogenic trace gases, including nitrous oxide (N<sub>2</sub>O), methane (CH<sub>4</sub>), and nitric oxide (NO). The first two are greenhouse gases, contributing about 25% of the change in radiative forcing over the last decade. Tropospheric concentrations of these gases are increasing, but there is uncertainty concerning their global and regional sources and sinks (*Cicerone and Oremland, 1988; Matson and Vitousek, 1990; Watson et al., 1992*). Methane and NO are also important in tropospheric and stratospheric chemistry

(*Williams et al., 1992a; Cicerone and Oremland, 1988; Watson et al., 1992*), and through their effects on tropospheric ozone concentrations, have the potential to affect biological processes (*Field et al., 1992; Mooney et al., 1987*).

The consequences of fertilizer use on emissions of N<sub>2</sub>O have been evaluated in agricultural systems of the temperate zone (see *Eichner et al., 1990; Mosier, 1993; Bouwman, 1994*). In current budgets, global emissions of N<sub>2</sub>O are estimated by assuming that all agricultural systems respond in the same way that temperate, developed-world agricultural systems respond to fertilizer N additions. Cultivation is believed to be the largest single source of increase (*Watson et al., 1992*). The possibility that fertilization has different consequences in tropical agricultural systems has not been evaluated, yet there is reason to believe that they might differ. Tropical forest soils in general have higher fluxes of N<sub>2</sub>O and lose more N<sub>2</sub>O per unit of nitrogen cycled than do temperate systems (*Matson and Vitousek, 1990; Vitousek and Matson, 1993*).

Emission of NO as a consequence of fertilization has received much less attention in the temperate zone, and virtually none in developing world agriculture outside of pasture systems (*Davidson et al., 1991; Keller and Matson, 1994*). We could locate only 14 published estimates of NO flux in fertilized agricultural systems (all in temperate areas), but these studies indicate significant effects of fertilization on NO fluxes (*Johannson and Granat, 1984; Williams et al., 1988; Hutchinson and Brams 1992; Shepard et al., 1991; Williams et al. 1992a, 1992b; Hall et al., unpublished report*).

Because of the dearth of field data, no statistical analysis of the role of fertilizer type or management has been possible.

This research addresses the effects of the increasing use of fertilizers in agriculture in tropical environments on fluxes of N gases. In this paper, we first present patterns of N trace gas emissions as they vary among sugar cane fields on the islands of Maui and Hawaii in the Hawaiian Islands, USA. These patterns are then related to controlling processes and to variation in tropical soil types and management practices.

## **Methods**

### *Site Description*

We measured trace gas fluxes following operational and experimental fertilization events in 8 sugar cane fields on Maui and 4 on the Island of Hawaii. The Maui fields are owned and managed by Hawaiian Commercial & Sugar Company (HC&S), Puunene (20°52'N, 156°27'W). In these fields, fertilizer is typically applied as urea in irrigation water fed through drip lines buried between cane rows. Irrigation line areas cover approximately 18% of the field surface; cane rows cover 38% and interrow areas cover 43% (Figure 1). Multiple split applications of urea are timed to crop requirements (based on analyses of foliar N concentration); N application rates range from 20-80 kg/ha, and over the entire 22-24 month rotation, application of N ranges from 300-500 kg/ha.

Two Hawaii fields are located on the south slope of Mauna Loa (19°14'N, 155°30'W) and managed by Ka'u Agribusiness; the other

two are located on the west slope of Mauna Kea (19°45'N, 154°10'W) and are owned and managed by Mauna Kea Agribusiness. These fields received urea fertilizer in pellet form, broadcast either by tractor when the cane is small or by airplane. When applied by tractor, urea is applied in a strip over the cane rows; fertilized strips account for approximately 45% of the field area and interrow areas account for 55%. When applied from planes, fertilizer is broadcast across the entire field. Fertilizer is applied in fewer splits and larger amounts than on Maui. Application rates for the fields we studied were all around 100 kg/ha; over the 24-60 month rotation, N applications range from 500-800 kg/ha. Site information is provided in Table 1.

#### *Gas Collection and Analyses*

We sampled nitrous oxide (N<sub>2</sub>O) and nitric oxide (NO) prior to and following fertilization of Maui and Hawaii fields. Gas samples were collected using two-piece chambers (*Matson et al., 1990; 1991; 1992*); a 25 cm diameter polyvinylchloride (PVC) base was inserted before measurement and then capped with an 10 cm high acrylonitrile-butadiene-styrene (ABS) plastic cover at the time of sampling. Gas samples routinely were collected from 8-16 randomly placed chambers; on Maui, 4-6 chambers were placed in each of the irrigation line, cane line and interrow positions (Figure 1); on Hawaii, where irrigation lines are not used, 4-6 chambers were placed in cane line and interrow positions. Gas samples were collected 4 times over a 30 minute period using nylon stop-cocked

syringes, and were immediately returned to the laboratory for analysis of N<sub>2</sub>O.

Nitrous oxide was analyzed using Shimadzu Mini gas chromatograph Model 2 configured with an electron capture detector (ECD) containing <sup>63</sup>Ni as the isotope source and using a argon/methane carrier gas (*Garcia-Mendez et al., 1989*). Standards (0.1, 0.5 and 1.0 ppm; Scott Research Laboratory, Inc., Plumsteadville, PN) bracketed every 12 samples, and concentrations of the chamber gases were calculated using least squares linear regression. Fluxes were calculated by regressing N<sub>2</sub>O concentration within the chamber against sampling time, and correcting for temperature and the ratio of chamber volume to soil surface area covered. Minimum detectable flux was approximately 0.1 ng cm<sup>-2</sup> h<sup>-1</sup>.

Within 1 h (usually within 30 minutes) preceding or following sampling for N<sub>2</sub>O, NO was measured in the same rings using a Scintrex LMA-3 chemoluminescence detector modified for field measurements (*Davidson et al., 1991; 1993*). In brief, a vented cover was placed on the PVC base and a continuous flow of air at approximately 0.2 L/min was drawn through the chamber. Ambient air at a flow of approximately 0.8 L/min was passed through Drierite and Ascarite to remove water vapor and nitrogen oxides, and then mixed with chamber air before entering a CrO<sub>3</sub> converter where NO was oxidized to NO<sub>2</sub>. The NO<sub>2</sub> was detected using the LMA-3. We carried out infrequent tests for soil NO<sub>2</sub> emissions, in which sample air by-passed the CrO<sub>3</sub> converter. Because emissions of NO<sub>2</sub> were very low or zero in those tests, we here refer to all soil

emissions as NO. Chamber air NO values were recorded every 15 seconds, converted to nitric oxide concentrations using standard curves (using dilution of a 0.1 ppb standard) run in the field before and after soil measurements, and flux was calculated using linear regression of concentration over time. Minimum detectable flux was approximately  $0.05 \text{ ng cm}^{-2} \text{ h}^{-1}$ .

### *Soil Sampling and Analyses*

During gas sampling, soil temperatures were taken from the outside, shaded side of the chambers at 1-2 cm depth. Following gas sampling, soil samples were collected to 10 cm depth from positions adjacent (within 1 m) to each ring. Within 3 h after collection, soils were mixed by hand and plant material and inclusions > 3 mm were removed. Moisture content was determined on subsamples of the soil by weighing field moist samples before and after oven drying at 100 °C for 48 hr. Samples from each position were composited and pH was determined using a method described by McLean (1982) using a Corning Model 10 pH meter with a standard electrode; we used 20 g of air dried soil and 20 ml of deionized water.

For estimates of nitrogen concentrations in soil, two 10 g subsamples were removed from each sample. One was placed in 100 ml 2 N KCL, shaken, and allowed to equilibrate for 18 to 24 hr; the second subsample was incubated in a closed cup for 7 days, after which it was extracted in KCl as above. Nitrate-N ( $\text{NO}_3$ ) and ammonium-N ( $\text{NH}_4$ ) concentrations were analyzed using a continuous flow autoanalyzer (*Perstorp Analytical, 1990*).



One or two times during the period of study, soil samples from most fields were collected from each position, composited by field, oven-dried and shipped to the University of California, Berkeley for analysis of total carbon (C) and N using a Carlo Erba CN analyzer.

In 1994, three 237 cm<sup>3</sup> soil samples were collected to 10 cm depth in each position at most of the Maui and Hawaii fields and bulk density was determined (*Blake and Hartge, 1986*). Care was taken not to compress the sample. After removing and estimating the volume of inclusions > 3 mm in diameter, soils were dried for 48 h at 100°C. Inclusions typically represented less than 5% of the core volume. Water-filled pore space (WFPS) was then calculated using inclusion free bulk density, gravimetric moisture data and particle density; particle density for each field was assigned according to soil type (*Soil Survey Staff, 1972; U.S Soil Survey, 1976*)

### *Operational Fertilization Events*

Gas flux and soil measurements were taken preceding, during, and after 8 fertilizer events in 7 fields on Maui and 3 events in 3 different fields on Hawaii. Field numbers, sampling dates, cane age at sampling, and amount of fertilizer N applied are listed in Table 2. Measurements typically were initiated within 24 h before fertilizer was applied, and were continued at daily to less frequent intervals until N<sub>2</sub>O and NO fluxes fell to near pre-fertilization levels.

### *Surface vs. Subsurface Fertilization Experiments*

To determine the relative importance of application practice on nitrogen trace gas emissions, we carried out several comparisons

of surface vs. subsurface applications of urea within single fields, using fields 100 and 908 on Maui (where fertilizer is typically applied subsurface), and field Ka'u 26 in Hawaii (where fertilizer is applied on the surface). Field 100 was 5 mo old at the time of the experiment, field 908 was 9 mo old, and Ka'u 26 was 8 mo old.

In field 100, urea was applied at the rate of 33 kg/ha, but because it was focused on irrigation lines only (as is the operational application), the effective rate of fertilization was 187 kg/ha. Because soils were moist, no water was added. Gases were sampled from 8 chambers (and soils sampled adjacent) beginning one day prior to treatment and continuing for 5 days, when operational subsurface irrigation and fertilization (also at the rate of 33 kg/ha) occurred. Gas fluxes and soil characteristics of the subsurface treatment were measured daily for 5 days in an area of the field adjacent to the earlier experimental surface application. We compare the two fertilizer application treatments directly despite the fact that measurements were of necessity acquired approximately one week apart.

In addition to measurements of N<sub>2</sub>O and NO fluxes in field 100, we also used acid traps to collect ammonia (NH<sub>3</sub>-N) emitted from the soil (*Schlesinger and Peterjohn, 1991*). Immediately following measurement of N<sub>2</sub>O and NO, 10 ml of 2% sulfuric acid solution were poured from acid washed screw-top test tubes into acid washed beakers and placed in the center of each ring. Chamber top vents and sampling ports were sealed and tops were placed over the rings for two hours, after which the acid solutions were resealed in the test tubes. Acid solutions that were transferred from test tubes to

beakers and back to sealed test tubes were treated as blanks. The acid solutions were analyzed for  $\text{NH}_4\text{-N}$  as described above.

In field 908, treatments and sampling were the same as above, except that application to the surface and subsurface were both at the rate of 20 kg/ha, again focused on irrigation lines. Here, pre-fertilization measurements were done within several hours of application, and continued daily for 7 days after fertilization.

In field 26 in the Ka'u plantation on the Island of Hawaii, where urea is typically applied to the surface, we used a completely randomized experimental design with 4 replicates of 3 treatments: water application, subsurface injection of urea dissolved in water, and surface application of urea in water. Here, operational fertilization rates are typically 100 kg/ha of urea-N applied to the entire surface of the field; we applied urea to 30 cm x 30 cm plots (with chamber rings centered within) in the cane lines at the rate of 100 kg/ha N; urea was dissolved in 1 L water. Subsurface applications were carried out by injecting solution to approximately 8 cm depth using syringes and tubing. Pre-fertilization measurements were taken 48 h prior to treatment; post-fertilization measurements were taken every other day for 11 days post fertilization. To compare the surface and subsurface fertilizer treatments separately from the effects of watering, we report data for the fertilizer treatment fluxes minus the water treatment flux.

### *Succinate and Urea Experiments*

In order to evaluate the importance of variation in available carbon as a factor in determining differences in gas fluxes between

Maui and Ka'u sites, we carried out succinate and urea additions in fields 100 (6 mo old at the time of sampling) and 105 (1 mo old) on Maui and field 26 (8 mo old) at Ka'u, Hawaii. Our factorial treatments included 3-4 replicates of a water control, surface urea application, surface succinate application and surface succinate plus urea application. Succinate and urea were dissolved in 1 L water. On Maui, treatments were applied to the area inside chamber rings plus a 5 cm band around the rings; treatments were applied on irrigation lines only. Urea N was applied at similar rates to the operational fertilization (33 kg/ha for whole field; focused in the irrigation line, approximately 187 kg/ha), and succinate-C was applied proportionally to N at a ratio of 6:1 (a typical C:N ratio of microbial biomass). In Ka'u 26, treatments were applied to the cane rows and were applied to 30 x 30 cm plots with chamber rings centered within. Here, urea was applied at the rate of 100 kg/ha to simulate typical broadcast applications, and succinate-C was again applied at a ratio of 6:1 C:N.

#### *Acetylene Inhibition Experiments*

To distinguish between autotrophic nitrification and denitrification as sources of  $N_2O$ , an acetylene ( $C_2H_2$ ) inhibition method described by Davidson (1992) was used. This approach relies on the fact that low levels of  $C_2H_2$  inhibit nitrification but not denitrification. Two 25 g subsamples of hand mixed, field moist soil were removed from each of 4-8 soil samples taken from irrigation lines in fields 100, 907, and 817 on Maui at two different times in November and Dec 1993, from two different surface applications in

field 100, and from cane rows in surface fertilized field MK67 in Hawaii in January 1994.

Each subsample was placed in a 0.475 L (1 pint) mason jar and sealed with lids fitted with rubber septa. The headspace gas of 1 jar was brought to 10 Pa by injection of  $C_2H_2$  (generated from calcium carbide); the second served as the control. One hour following injection of  $C_2H_2$ , 5 mL of headspace gas was removed from each jar and analyzed for  $N_2O$  (see above). A second gas sample was removed and analyzed at 7 hours following  $C_2H_2$  injection. The results from the 1 hour sample was subtracted from the 7 hour time point to estimate emission. Percent of  $N_2O$  derived from denitrification was estimated by dividing the 10 Pa  $C_2H_2$  treatment values by control values.

### *Statistical Analyses*

Trace N gas and soil N variables were log-transformed prior to statistical analysis using Statview II (*Abacus Concepts, 1992*). Surface vs. subsurface comparisons were made using a t-test to distinguish differences between means at each time point following fertilization. Treatment and interaction effects in the succinate/urea experiment were evaluated using a factorial design ANOVA.

Stepwise regression was used to determine the relative importance of soil N, WFPS and temperature variables in relation to N trace gas flux at each sampling point within individual fields. Stepwise regression also was used to evaluate relationships between total trace gas fluxes over entire sampling periods vs. mean

values of soil N, soil temperature, WFPS, total carbon and nitrogen, and the amount of N applied per operational fertilization event among fields.

Total fluxes of N<sub>2</sub>O and NO were calculated for each operational fertilization event. Because diel measurements taken at 3 different times in the crop cycle did not show significant difference in fluxes over 24 h periods, we assumed that all measured fluxes were constant over 24 h. In Maui fields, collection points were usually 24 hours apart and continued for approximately one week. In Hawaii fields, fertilization events were followed for 2-4 weeks and collection points were often more widely spaced, especially later in the sampling period. When measurements were collected every 48 h or less, fluxes at one time point were assumed to be constant until the next measurement time point. When fluxes were separated by more than 48 hours, estimates for unsampled time periods were interpolated at 24 hour intervals.

Mean fluxes for each position over each time interval were then weighted on an area basis and summed to give a weighted flux per interval. In Maui, the weightings for each position were 0.18 for irrigation lines; 0.38 for cane line; and 0.43 for interrows. On Hawaii, the spatial weightings were 0.45 for interrow areas and 0.55 for cane lines. The weighted fluxes for each interval were then summed to provide a total flux (kg/ha) for the fertilization event. Total fluxes were divided by the amount of N applied in each fertilizer event to calculate percent loss of fertilizer N as either N<sub>2</sub>O-N or NO-N.

## Results

### *Chemical and Physical Characteristics of Soils*

Soil characteristics differed markedly between Maui and Hawaii (Table 3). Maui soils were mostly clay and clay loam, with bulk densities ranging from 1.06-1.26; in contrast, Hawaii's andepts soils had silt loam textures and bulk densities well below 1. Percent carbon and nitrogen contents were very low in the Maui soils compared to Hawaii, and Maui pH's were generally higher than Hawaii's (Table 3).

### *Trace Gas Fluxes in Response to Fertilization*

The temporal pattern of nitrous oxide response to fertilizer application was similar among the Maui and Hawaii sites, with  $\text{N}_2\text{O}$ -N fluxes very low before fertilization (but after the initiation of irrigation on Maui), increasing rapidly within 24 hours after fertilization, peaking within 2-3 days, and gradually returning to background levels (Figure 2,3). In the Maui sites, fluxes returned to background levels within 7 days (Figure 2), whereas they remained elevated for 2 weeks and more in the Hawaii sites (Figure 3).

Within-field spatial patterns of  $\text{N}_2\text{O}$ -N flux depended on the fertilizer application practice. In the Maui sites, fluxes above the buried irrigation line, where fertilizer was applied, were nearly always significantly higher than on the cane line; interrow areas had very low fluxes that showed little response to fertilization or irrigation (Figure 2). In the Hawaii fields, there were typically no significant differences between cane rows and inter-row positions

when fertilizer was applied by plane (ie, MK32 and MK67), but there were differences ( $p < .05$ ) when fertilizer was broadcast on the cane rows by machine (as in Ka'u 31) (Fig 3).

Maximum mean  $\text{N}_2\text{O-N}$  fluxes on both islands were usually less than  $30 \text{ ng cm}^{-2} \text{ h}^{-1}$ , but very high fluxes of over  $100 \text{ ng cm}^{-2} \text{ h}^{-1}$  were measured in Maui field 817 (at the 7/93 fertilization) and in MK32 on Hawaii.

When weighted by position (irrigation line, cane row and inter-rows) and summed over the entire response period, fluxes of  $\text{N}_2\text{O-N}$  from the Maui fields ranged from  $0.006 \text{ kg/ha}$  to  $0.171 \text{ kg/ha}$ , representing from  $0.013$  to  $0.493\%$  of the N applied in fertilization (Table 4). Fluxes summed and interpolated over the longer response periods in the Hawaii fields ranged from  $0.325$ - $1.245 \text{ kg/ha}$ , and represented from  $0.346$ - $1.00\%$  of the added fertilizer.

Temporal patterns of  $\text{NO-N}$  flux were less distinct than for  $\text{N}_2\text{O-N}$ , but peak flux times generally lagged behind peak  $\text{N}_2\text{O}$  fluxes. There was also a striking contrast in magnitude of emissions between Maui and Hawaii (Figures 4,5). On Maui, mean  $\text{NO-N}$  fluxes were often well below  $3 \text{ ng cm}^{-2} \text{ h}^{-1}$  (Figure 4), and were often an order of magnitude less than  $\text{N}_2\text{O-N}$ . On Hawaii, mean  $\text{NO-N}$  emissions reached above  $90 \text{ ng cm}^{-2} \text{ h}^{-1}$  (with the highest flux of  $153.6 \text{ ng cm}^{-2} \text{ h}^{-1}$ ) (Figure 5), and were much higher than  $\text{N}_2\text{O}$  (often by factors of 2-5) throughout much of the response periods.

Over the entire fertilizer response period,  $\text{NO-N}$  losses were very small (ranging from  $0.001$ - $0.013 \text{ kg/ha}$ ) in the Maui fields, representing from  $0.004$ - $0.038\%$  of the applied fertilizer (Table 4). In contrast, summed  $\text{NO}$  losses from the Hawaii sites were



relatively large (0.713 and 1.953 kg/ha for fields Ka'u31 and MK67 respectively), and represented a more substantial proportion of the fertilizer N (0.758 and 2.06% respectively; Table 4).

*Soil nutrients and water, and relationships with fluxes*

Temporal patterns of soil  $\text{NH}_4\text{-N}$  concentrations indicated that conversion of urea to  $\text{NH}_4$  occurred rapidly; ammonium concentrations peaked and began to decline within 2-3 days after fertilization in the Maui sites, and within a week in the Hawaii sites (see Figure 6a and b for examples). Peak concentrations of  $\text{NO}_3\text{-N}$  often lagged behind  $\text{NH}_4$ . Spatial patterns reflected the site of fertilizer application, with highest  $\text{NH}_4$  and  $\text{NO}_3$  concentrations occurring on the irrigation line (in the Maui sites) and cane rows (in the Ka'u31 site in Hawaii). Ammonium and  $\text{NO}_3$  concentrations did not change significantly in the interrow areas where fertilizer was not applied, although pools did increase somewhat in the cane lines adjacent to irrigation lines in the Maui sites.

While patterns of change in  $\text{NH}_4$  and  $\text{NO}_3$  were similar between islands, concentrations were dramatically different (Figure 6). On Maui, pre-fertilization levels of  $\text{NH}_4$  were always less than 5  $\mu\text{g/g}$  and peak mean concentrations following fertilization rarely exceeded 30  $\mu\text{g/g}$ . Pre-fertilization concentrations of  $\text{NO}_3$  were less than 30  $\mu\text{g/g}$  and post-fertilization levels exceeded 60  $\mu\text{g/g}$  only one day in one site. Within each Maui field, pre-fertilization  $\text{NO}_3$  was often higher than  $\text{NH}_4$  (possibly due to  $\text{NO}_3$  in irrigation water, which was not measured in this study), but post-fertilization

mean and maximum  $\text{NH}_4$  concentrations were usually higher than  $\text{NO}_3$  concentrations.

On Hawaii, both  $\text{NH}_4\text{-N}$  and  $\text{NO}_3\text{-N}$  concentrations were much higher. In all Hawaii sites, pre-fertilization  $\text{NO}_3$  was between 35-100  $\mu\text{g/g}$  and ranged as high as 700  $\mu\text{g/g}$  after fertilization. Pre-fertilization  $\text{NH}_4\text{-N}$  concentrations ranged from 30-100  $\mu\text{g/g}$  in the Mauna Kea sites, and peaked at near 200 $\mu\text{g/g}$ . In the Ka'u 31 site, average  $\text{NH}_4$  concentrations were less than 1  $\mu\text{g/g}$  several hours before fertilization, jumped to around 70  $\mu\text{g/g}$  within several hours after fertilization, and peaked at 625  $\mu\text{g/g}$  within 6 days.

Soil moisture in the Maui sites was controlled by the irrigation that always occurred prior to and during fertilization as well as by precipitation events. Percent water filled pore space (WFPS), which is calculated using soil texture, particle size and density, and soil moisture, was always above 40% in the irrigation lines before fertilizer application and remained consistently high throughout our sampling periods (Table 5). Values in irrigation lines were always higher than cane line and interrow initially, but in several of the sites, cane line values were no longer significantly lower than irrigation lines 24 h post fertilization. Soil moisture and WFPS did not change significantly in the interrow areas, and were typically 15-20 percentage points lower than in irrigation lines. The exception to this was field 105, where rains brought cane line and interrow WFPS to between 35-40%, while irrigation line values ranged from 40-45%.

In the Hawaii sites, soil moisture and WFPS did not vary by position, and reflected rain events only. In all three sites, there

were no significant differences by time or position. The average WFPS were 29.7%, 28.7% and 36.1% for MK32, MK67 and Kau31 respectively (Table 5).

Analysis of residuals of linear regressions of N<sub>2</sub>O and NO vs. soil variables within each site suggested that the variables we measured do not account for variation in trace gases among sampling points. Therefore, our regression analysis of point data for individual sites does not fit the assumptions required for the regression model. When we carried out stepwise regressions between total fluxes for each site (in kg/ha/sampling period) and average soil nitrate, ammonium, WFPS, pH, total C, total N and amount of fertilizer applied for each site, we found significant relationships between N<sub>2</sub>O-N and WFPS ( $p < .05$ ,  $R^2 = .579$ ), and WFPS plus pH ( $p < .02$ ,  $R^2 = .867$ ). Nitric oxide was significantly related to total C ( $p < .01$ ,  $R^2 = .739$ ); this relationship is not likely to be mechanistic, but probably results because of the very large differences in concentrations of total C in andept soils on Hawaii vs. the Maui soils.

#### *Surface vs. Subsurface Fertilization*

Under operational fertilization practices, Maui fields always had much larger fluxes of N<sub>2</sub>O-N than NO-N; on Hawaii, NO was as high or higher than N<sub>2</sub>O. To evaluate the degree to which these differences resulted from fertilizer application practice (subsurface on Maui, surface on Hawaii), we experimentally compared the effects of surface vs. subsurface urea application on N gases emissions in two Maui fields and one Hawaii field. On Maui,

experimental surface applications of urea in Field 100 resulted in  $\text{N}_2\text{O}$ -N fluxes that were a factor of 6 lower than NO fluxes after 4 days (Figure 7a). In contrast, operational subsurface fertilization with the same amount of urea resulted in  $\text{N}_2\text{O}$  fluxes that were 2-fold higher than NO, at least for the first two days following fertilization (Figure 7a). Nitrous oxide fluxes were significantly higher in the subsurface treatment than in the surface treatment on every day except the first and sixth day after fertilization ( $p < .005$  or lower). Interestingly, NO-N fluxes in the subsurface fertilization event were not consistently significantly different from NO in the surface event and were much higher than typically seen with subsurface fertilization (Figure 4).

Experimental surface fertilization in field 908 also led to NO-N fluxes that were high in comparison with  $\text{N}_2\text{O}$ -N fluxes. NO emissions in the subsurface treatment were significantly lower than in the surface treatments on all days except 3 and 4 days post fertilization ( $p < .0001$ ), while  $\text{N}_2\text{O}$  fluxes did not differ between treatments (Figure 7b).

Experimental surface fertilization in Hawaii's Ka'u 26 field also resulted in very high NO fluxes in comparison with  $\text{N}_2\text{O}$ . Despite high variability, NO emissions from subsurface fertilized plots were significantly lower than in surface fertilized plots 4 and 7 days after fertilization ( $p < .02$ ; Figure 7c).  $\text{N}_2\text{O}$  fluxes were higher in the subsurface treatment than in the surface treatment two days after fertilization ( $p < .05$ ), but were slightly lower in the subsurface treatment after one week ( $p < .05$ ; Figure 7c).

Results of acid trap collections of  $\text{NH}_4\text{-N}$  in field 100 on Maui also indicated strong qualitative differences in  $\text{NH}_3$  emissions between surface and subsurface applications. Ammonia (measured as ammonium in the acid vials) was significantly higher with surface than with subsurface fertilization. No ammonia was detected following subsurface fertilization, while concentrations up to 33  $\mu\text{g/g}$  were collected during 2 h measurement periods in the surface fertilized plots (Figure 8).

#### *Succinate and Urea Applications*

Nitrogen gas response to surface application of urea and/or succinate varied between Maui and Hawaii fields. In Maui fields 105 and 100,  $\text{N}_2\text{O}$  fluxes responded significantly within 24 h to succinate application ( $p < .0005$  and  $p < .0001$  for fields 105 and 100 respectively); response to urea application was significant in field 100 ( $p < .001$ ) but not in field 105 (Figure 9a,b). Nitric oxide fluxes showed no response in field 105, and a small but significant response ( $p < .001$ ) to urea application in field 100 (Figures 9a,b). Interactions between succinate and urea treatments were not significant in either field for either  $\text{N}_2\text{O}$  or  $\text{NO}$ .

In Hawaii,  $\text{N}_2\text{O-N}$  fluxes responded significantly to urea treatment ( $p < .05$  in the 23 h measurement;  $p < .013$  in the 95 h measurement), but not to succinate addition (Figure 9c).  $\text{NO-N}$  fluxes were significantly reduced by succinate application ( $p < .0001$ ).

#### *Acetylene Inhibition Experiments*

The  $\text{N}_2\text{O-N}$  produced in soils treated with 10 Pa  $\text{C}_2\text{H}_2$  (which blocks nitrification) was assumed to be produced during denitrification. As a proportion of untreated soils, denitrification-derived  $\text{N}_2\text{O-N}$  ranged from 22-107% in the Maui fields receiving operational subsurface fertilization. In contrast, the mean proportions from denitrification measured in soil from the two experimental surface applications on Maui were only 11.1% and 13.3%; the proportion in the soils collected from surface fertilized Ka'u31 in Hawaii was 3.4% (Table 6).

## Discussion

Patterns of N trace gas fluxes in response to urea fertilization varied consistently between Maui and Hawaii, with the Hawaii sites having elevated gas emissions for longer periods and substantially more  $\text{NO-N}$  than  $\text{N}_2\text{O-N}$  emitted. These differences may be related to the apparently greater importance of denitrification as a dominant gas source in the Maui sites in contrast to Hawaii. Experimental results suggest that the patterns in gas fluxes and controlling microbial processes reflect differences in soil characteristics as well as differences in fertilizer management.

### *Nitric Oxide vs. Nitrous Oxide Fluxes*

Perhaps the most remarkable difference between Maui and Hawaii sites was in  $\text{NO}$  flux. On Maui,  $\text{NO}$  emissions were always very low and represented a small fraction of the total gaseous N lost. On Hawaii,  $\text{NO}$  fluxes were high, and they represented the

majority of the measured gaseous N loss. Results from our experimental comparison of surface vs. subsurface fertilization suggest that these differences are in part due to the different fertilizer application methods employed on the two islands. Experimental surface fertilization in sites on both islands resulted in higher NO fluxes in comparison with subsurface applications, even when WFPS did not differ.

The much greater NO flux following surface fertilization rather than subsurface fertilization is probably not simply a result of differences in production. Nitrification occurred in both surface and subsurface fertilized situations. Within a given site, nitrate concentrations in the 0-10 cm soil profile were not different depending on application method. However, our sampling protocol did not differentiate vertical variations within the 0-10 cm increment. It is likely that in the subsurface treatments, nitrification was focused in a diffuse band several centimeters below the surface. Given NO's low rate of diffusion through most soils, especially under moist to wet conditions (*Galbally and Johnson, 1989*), it is possible that much of the NO produced during nitrification was consumed or further reduced before it could diffuse to the soil surface. With surface application, there is no delay to emissions and thus little potential for uptake within the soil.

Subsurface fertilization typically resulted in higher N<sub>2</sub>O than NO fluxes. The explanation for this may lie in the predominance of denitrification as a N<sub>2</sub>O source (Table 4). Higher rates of denitrification with subsurface fertilization in the field may be in

part due to high moisture levels and low redox potentials focused at several centimeters depth, although these were not measured in this study. Results of most studies agree that  $N_2O$  emission predominates over NO during denitrification in field environments, probably because the high water content that causes the low oxygen environment necessary for denitrification also restricts the diffusion of NO (*Tortoso and Hutchinson, 1990; Firestone and Davidson, 1989; Hutchinson and Davidson, 1993*).

#### *Total Losses in Response to Fertilization*

Nitrous oxide fluxes summed over the entire response periods were generally lower in the Maui fields than in Hawaii fields, in large part because the elevated fluxes seen in the Hawaii sites were sustained over longer periods of time. While the Hawaii fields had higher application rates than did the Maui sites, this is unlikely to explain the difference. Within the Maui sites, where application rates ranged from 20-84 kg/ha N, no significant relationship between amount of fertilizer applied and total or peak flux was evident. Moreover, the Maui sites in fact had higher effective application rates than did the Hawaii fields, because the urea was applied in narrow bands rather than in broad strips or over the entire field; nevertheless,  $N_2O$  fluxes were always short-lived in the Maui sites.

One possible explanation for the low total  $N_2O$  fluxes may be the low availability of carbon as a reductant for the denitrification process, which appears to be an important source of  $N_2O$  in the Maui subsurface-fertilized sites. On Maui, the addition of succinate



resulted in very large increases in nitrous oxide emissions, while urea alone did not. This suggests that in these sites, denitrification fluxes of N<sub>2</sub>O are limited by carbon availability, not nitrogen. Thus, even high additions of N would not be expected to lead to sustained high levels of gas emissions. In contrast, Hawaii fields already have very high organic carbon contents; additions of available carbon in these soils do not lead to increased gas emissions, while additions of urea do. In these fields, increasing additions of N would be expected to lead to increased denitrification losses of N<sub>2</sub>O, providing that moisture conditions and WFPS are high enough to restrict O<sub>2</sub> availability and allow denitrification to occur.

#### *Implications for Regional and Global Estimates*

Our results support the suggestion that soil characteristics, such as soil texture and organic matter content, and agricultural management practices are critical controls in the emission of N trace gases (*Bouwman, 1990;1994; Eichner,1990; Hutchinson and Davidson,1993; Mosier, 1993;1994; .Williams et al., 1992b*) Therefore, in attempting to estimate N gas fluxes at regional to global scales and predict how they will change with changing fertilizer use, a knowledge of soil characteristics and management as well as simply the amount and type of fertilizer may be required (*Mosier, 1994*). A number of local and global scale models and budgets of N gas fluxes have begun to incorporate these controlling variables (*Parton et al.,1988; Li et al., 1992a,b; Bouwman,et al., in press; Potter et al., in press*).

Our results also suggest that the proportion of urea fertilizer lost as  $N_2O$ -N may be larger than assumed in some global emissions estimates. Eichner (1990) reported  $N_2O$ -N loss as a proportion of urea fertilizer at .07-.18%; these estimates are currently the basis for global estimates (Watson, 1992). Our values of .03-1.0% may be more in line with those reported in a review by Bouwman (1994). Using his compilation of published data, we calculate an average  $N_2O$ -N loss of applied urea N of 0.5% (S.E. 0.16), with a range of .1-1.8%. However, all the values reported by Bouwman were based on measurement periods greater than 3 months following fertilization; his analysis suggests that as measurement periods are extended, the proportional  $N_2O$  flux values increase. Given the relatively short measurement periods in our study, our values are likely to be underestimates.

If the high percentage losses that we measured in Hawaii are common for tropical soils that are relatively high in organic matter (either as a result of soil type, fertilizer type -- i.e. organic fertilizers, or time since conversion from forest), current global calculations of  $N_2O$  flux from agricultural sources may be underestimated.

Fertilizer management practices also appear to be a very important control on  $NO$  emissions. Subsurface fertilization resulted in almost no N loss in the form of  $NO$  -- the highest proportional loss of urea N was .04%. Likewise, subsurface fertilization resulted in no ammonia loss, in accordance with earlier studies in sugar cane (unpublished Hawaii Sugar Planters Association data). On the other hand, broadcast surface fertilization

in Hawaii fields lead to very high NO fluxes; our mean flux values are among the highest reported in the literature (see reviews by *Williams et al., 1992b, Hall et al., unpublished report*). Moreover, these high fluxes were sustained over relatively long periods, resulting in proportional losses of up to 2% of the applied urea-N. In the only other study of NO flux from urea fertilized agricultural soils, Slemr and Seiler (1984) estimated a 3.25% loss of fertilizer applied; their fluxes were measured on bare soils previously planted in beans.

While such proportions may not be of great interest to farmers concerned with fertilizer loss, they do represent a potentially important flux in terms of atmospheric chemistry (*Jacob and Wofsy, 1990; Penner et al., 1991; Williams et al., 1992b*) and in downwind nitrogen deposition (*Melillo et al., 1989; Vitousek and Matson, 1993*).

In the comparisons reported here, the more complex fertilizer management practice resulted in lower N trace gas fluxes to the atmosphere. This method, in which fertilizer is applied below the soil surface and is carefully timed to crop requirements, must be considered information-intensive, requiring knowledge about crop physiology and phenology as well as up-front capital inputs in establishment of the application technology. It is clear, however, that such approaches can increase fertilizer-use efficiency, reduce the loss of fertilizer, and consequently limit the effects of agriculture on the atmosphere.

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## Figure Legends

Figure 1. Illustration of Maui sugar cane field spatial structure; irrigation lines run through mounds between double cane rows.

Figure 2.  $\text{N}_2\text{O-N}$  fluxes ( $\text{ng cm}^{-2} \text{h}^{-1}$ ) for seven fertilizer events in Maui sugar cane fields, separated by irrigation line, cane line, and interrow positions. Note the change in scale for each position.

Figure 3.  $\text{N}_2\text{O-N}$  fluxes ( $\text{ng cm}^{-2} \text{h}^{-1}$ ) fluxes following fertilization in 3 Hawaii sugar cane fields.

Figure 4.  $\text{NO-N}$  fluxes ( $\text{ng cm}^{-2} \text{h}^{-1}$ ) for seven fertilizer events in Maui sugar cane fields.

Figure 5.  $\text{NO-N}$  fluxes ( $\text{ng cm}^{-2} \text{h}^{-1}$ ) fluxes following fertilization in 2 Hawaii sugar cane sites.

Figure 6. Change in  $\text{NH}_4\text{-N}$  and  $\text{NO}_3\text{-N}$  concentrations ( $\mu\text{g/g}$  dry weight of soil) for Maui field 105 (a) and Hawaii field MK32 (b).

Figure 7. Comparisons of  $\text{N}_2\text{O-N}$  and  $\text{NO-N}$  fluxes ( $\text{ng cm}^{-2} \text{h}^{-1}$ ) in surface and subsurface fertilized plots in Maui field 100 (a), Maui field 908 (b) and Hawaii field Ka'u26 (c). For Ka'u26,  $\text{N}_2\text{O}$  and  $\text{NO}$  values are surface or subsurface treatment values minus values measured in the water (control) plots.

Figure 8.  $\text{NH}_4\text{-N}$  concentrations ( $\mu\text{g cm}^{-2} \text{h}^{-1}$ ) in acid traps in Maui field 100, reflecting the amount of ammonia emitted from soil surface within the chamber, trapped and converted to ammonium.

Figure 9.  $\text{N}_2\text{O-N}$  and  $\text{NO-N}$  fluxes ( $\text{ng cm}^{-2} \text{h}^{-1}$ ) measured 23 h after surface application of succinate and urea in Maui field 105 (a), Maui field 100 (b) and Hawaii field Ka'u26 (c). For Ka'u26, a 72 h post-treatment time point is also shown.

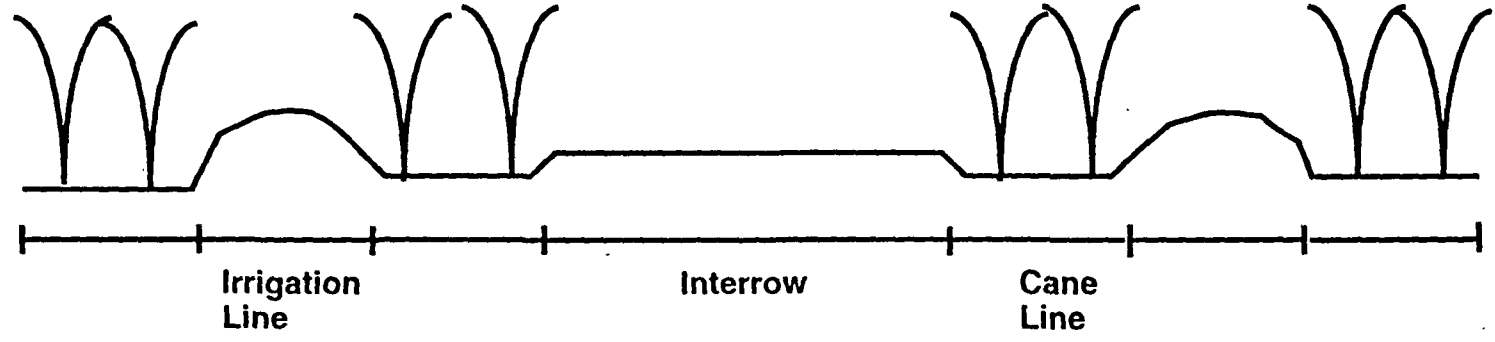


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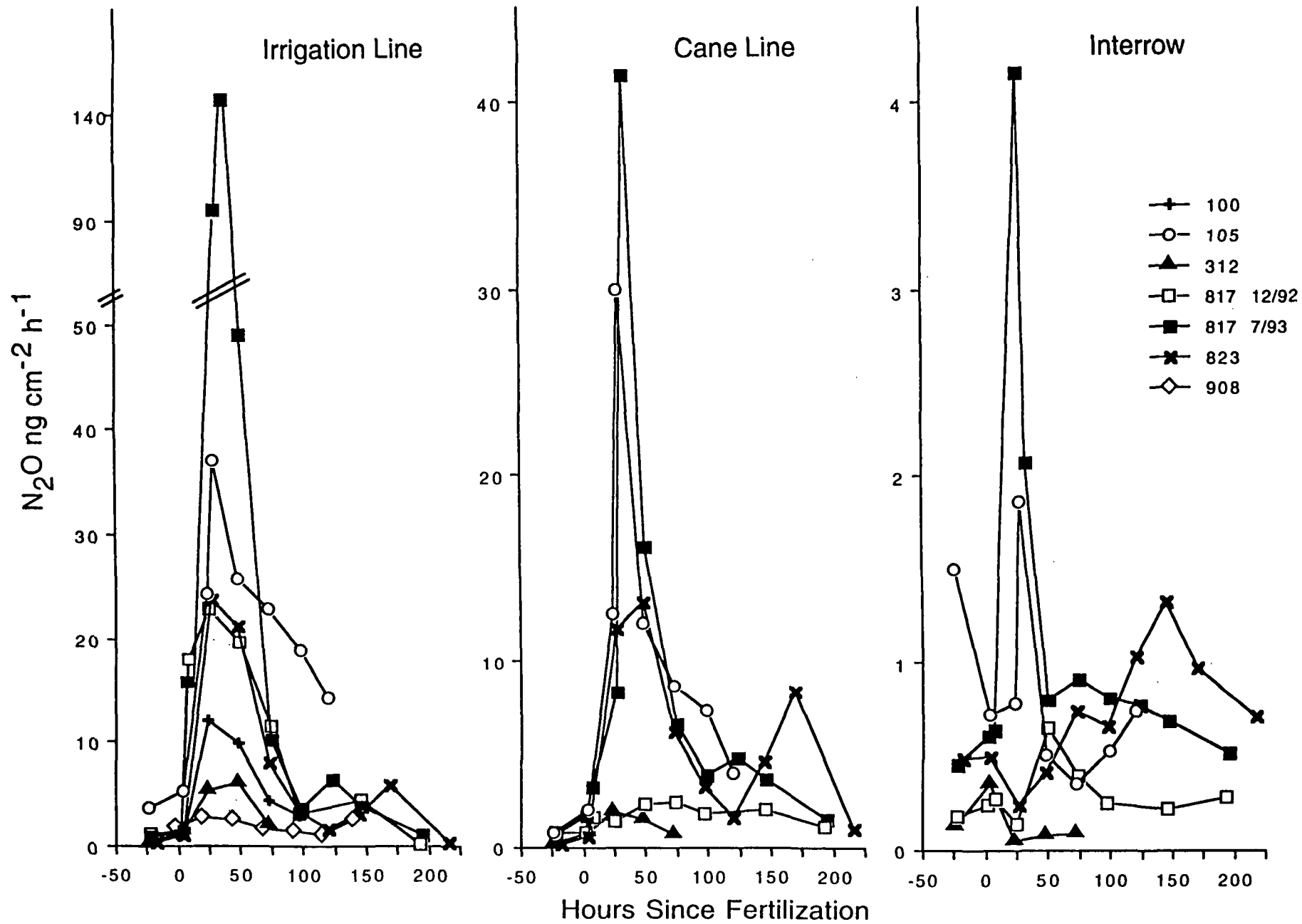


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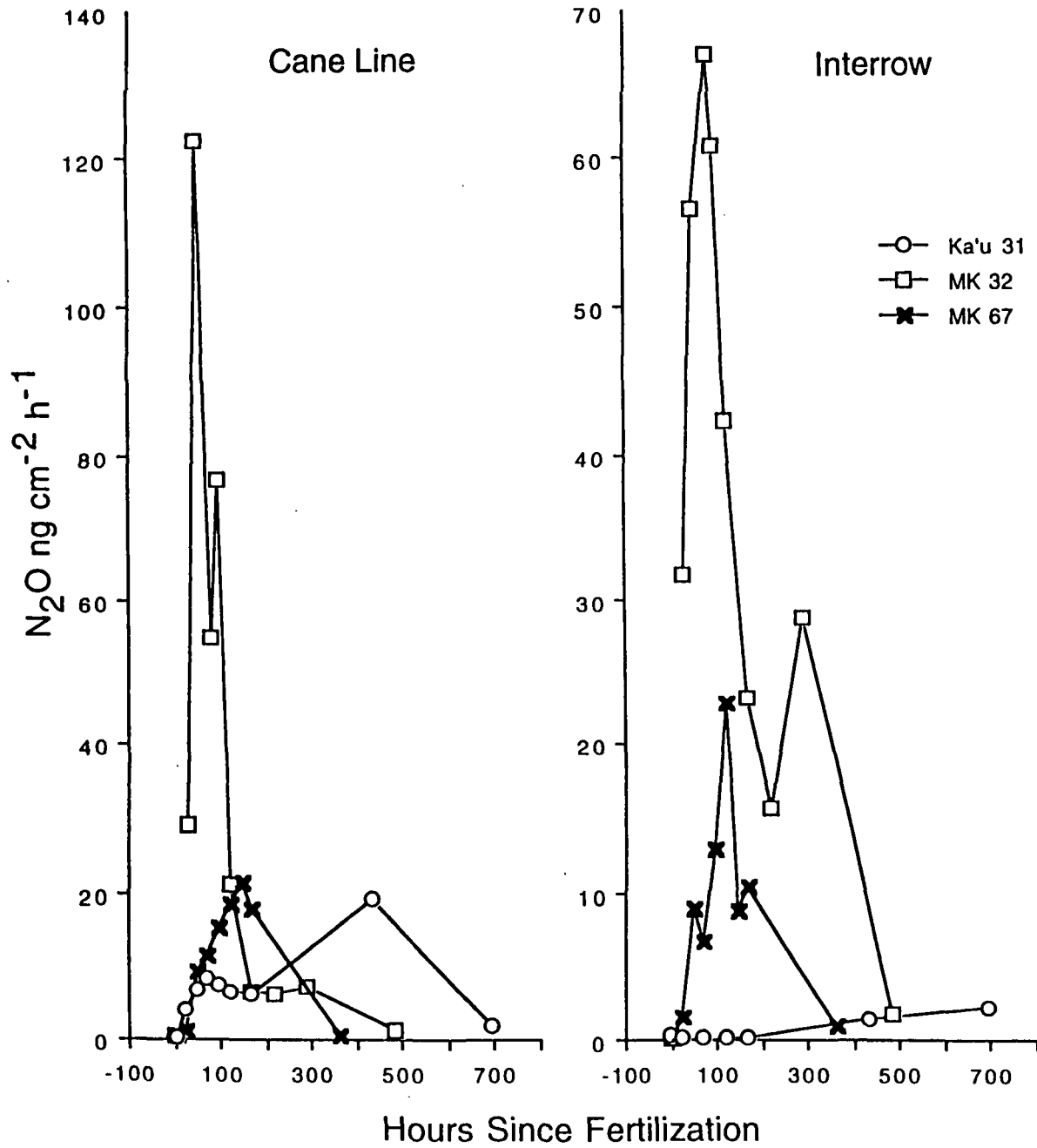


Figure 2



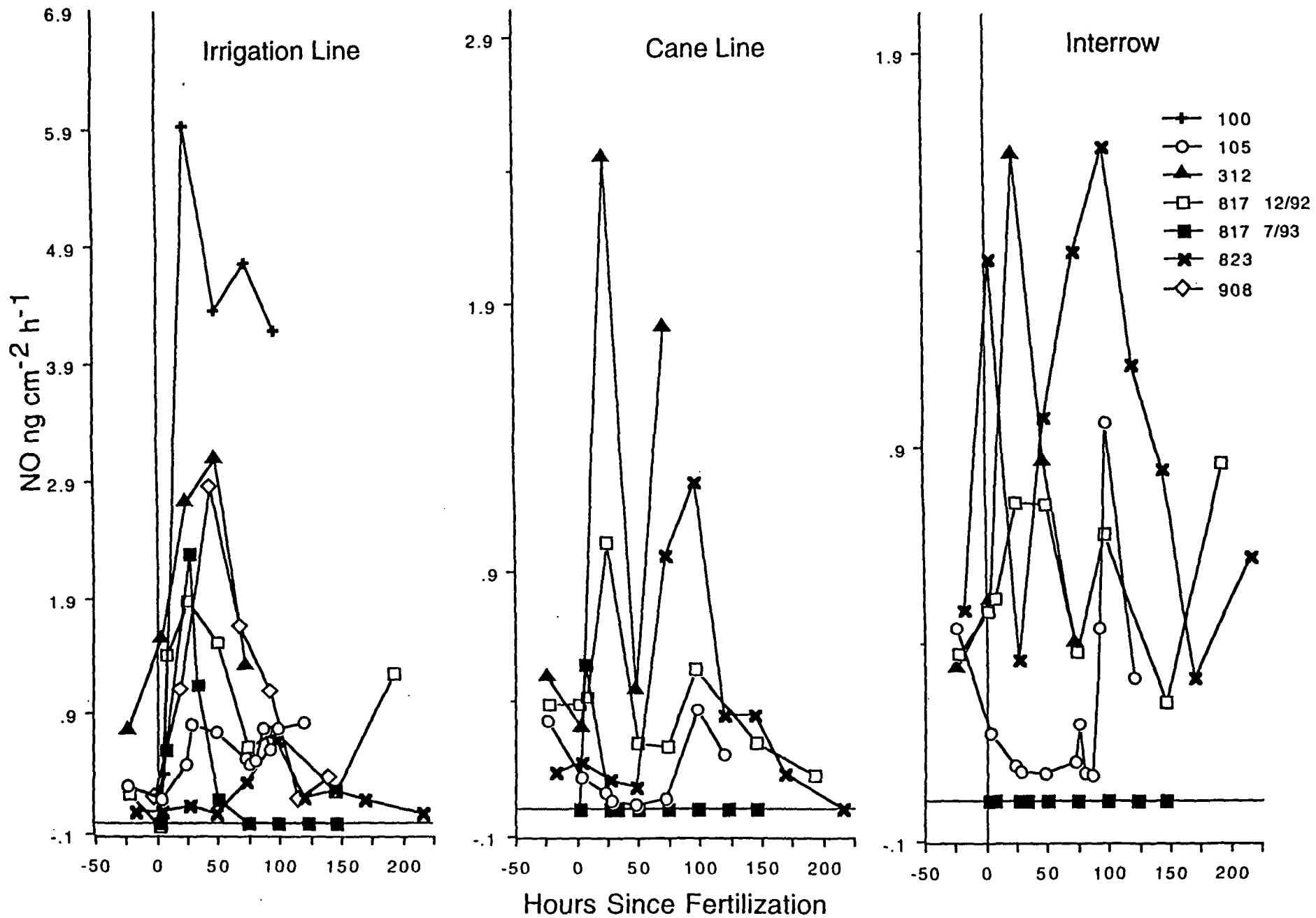


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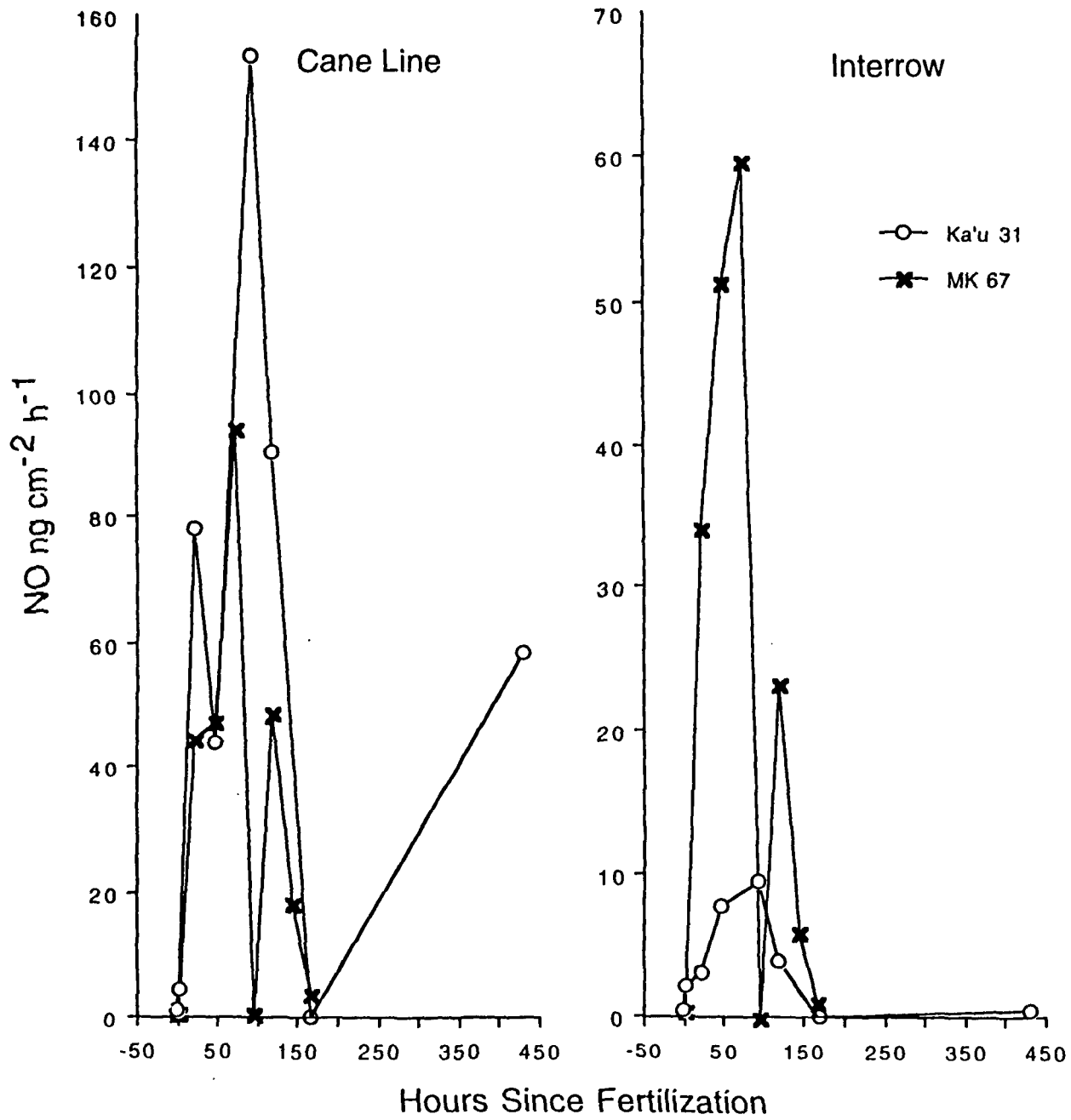


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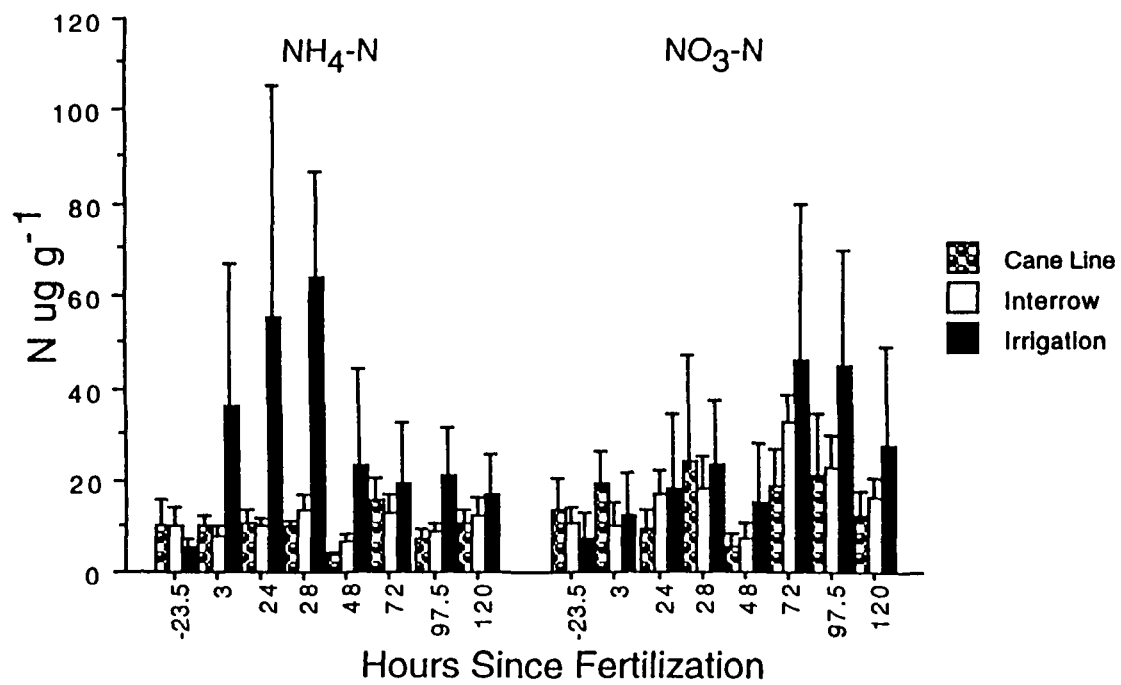


Figure 6a.

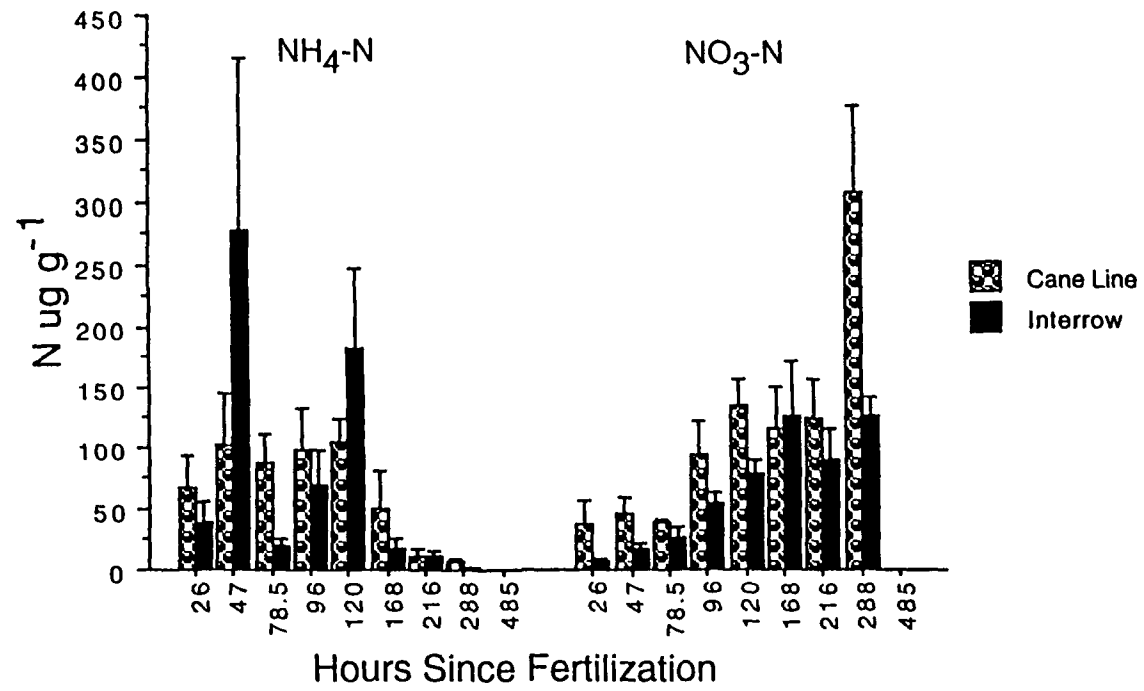


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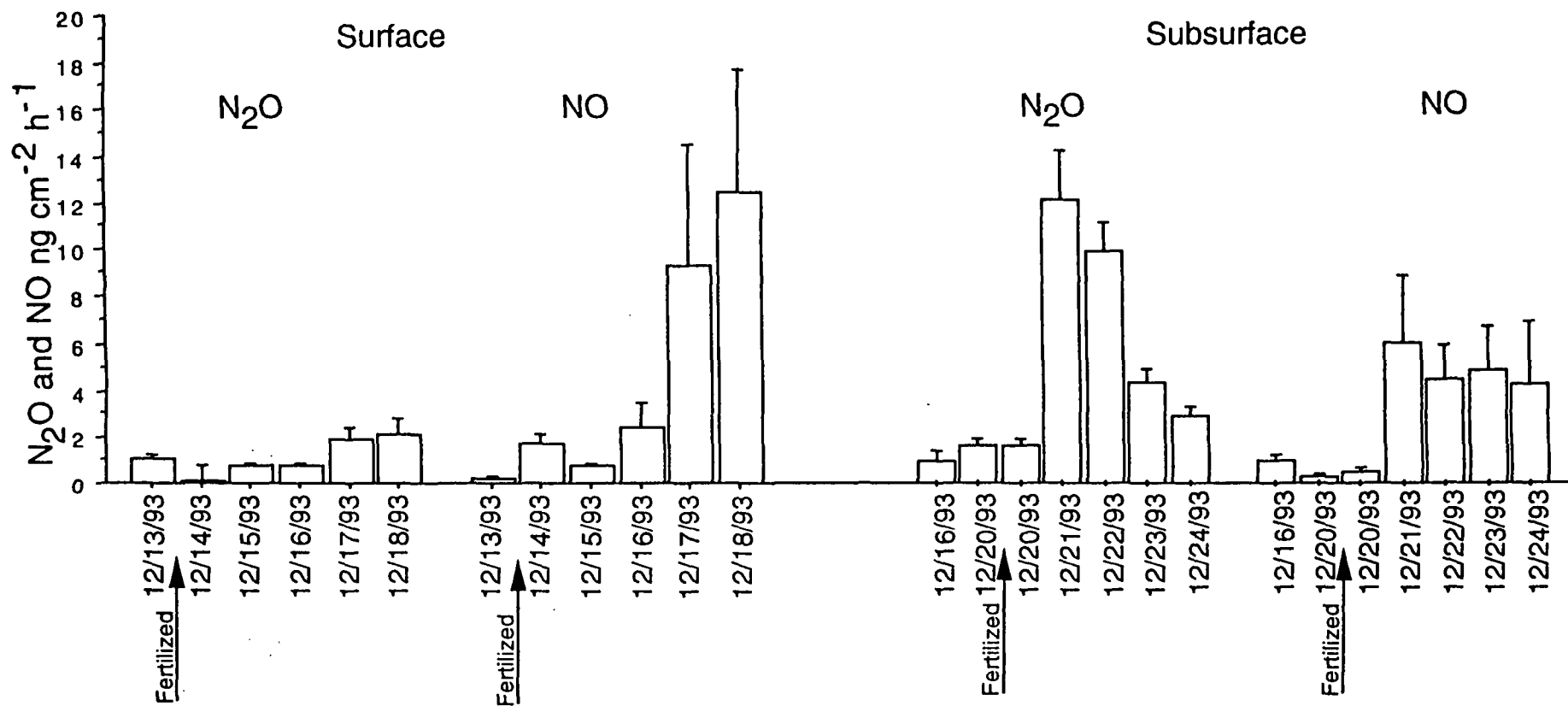


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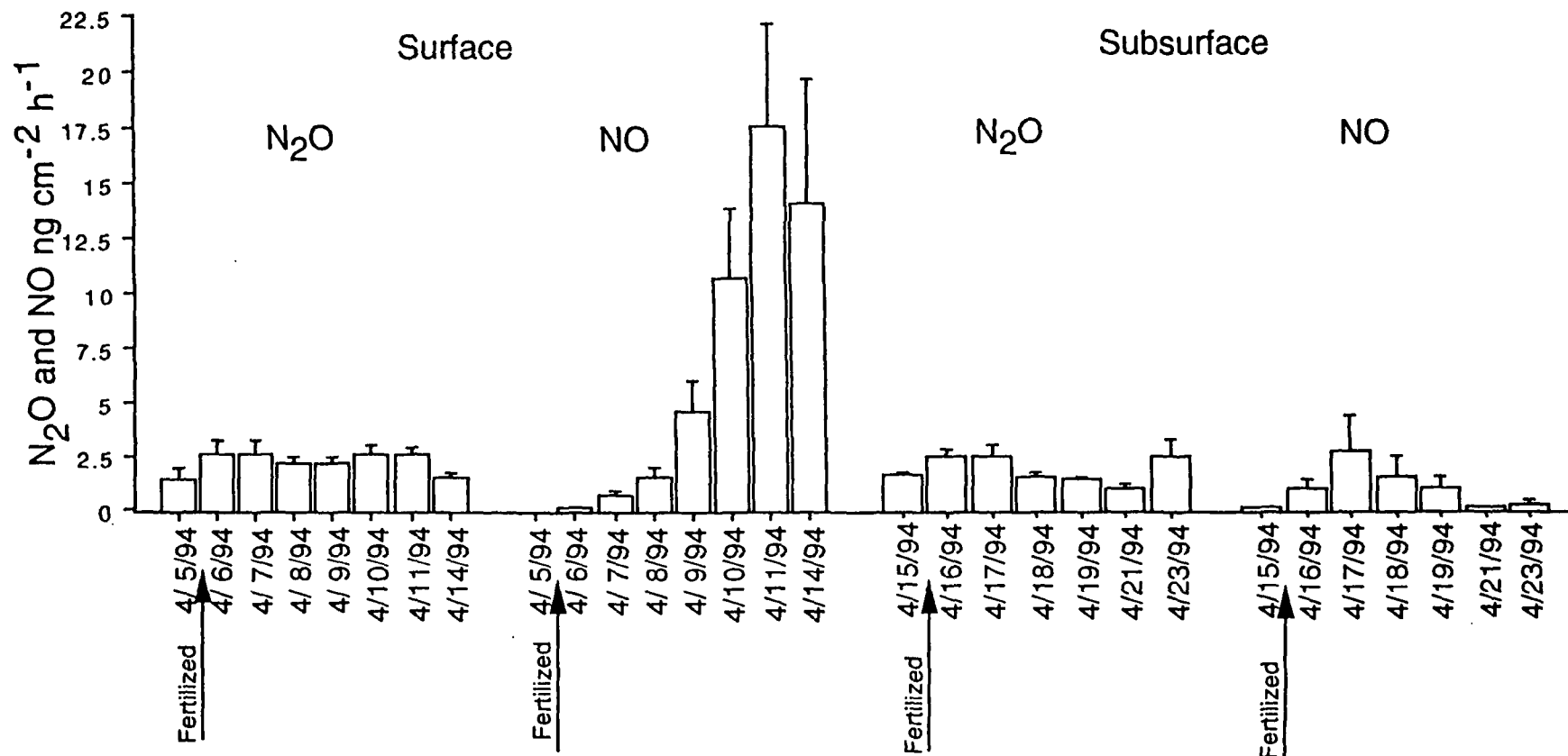


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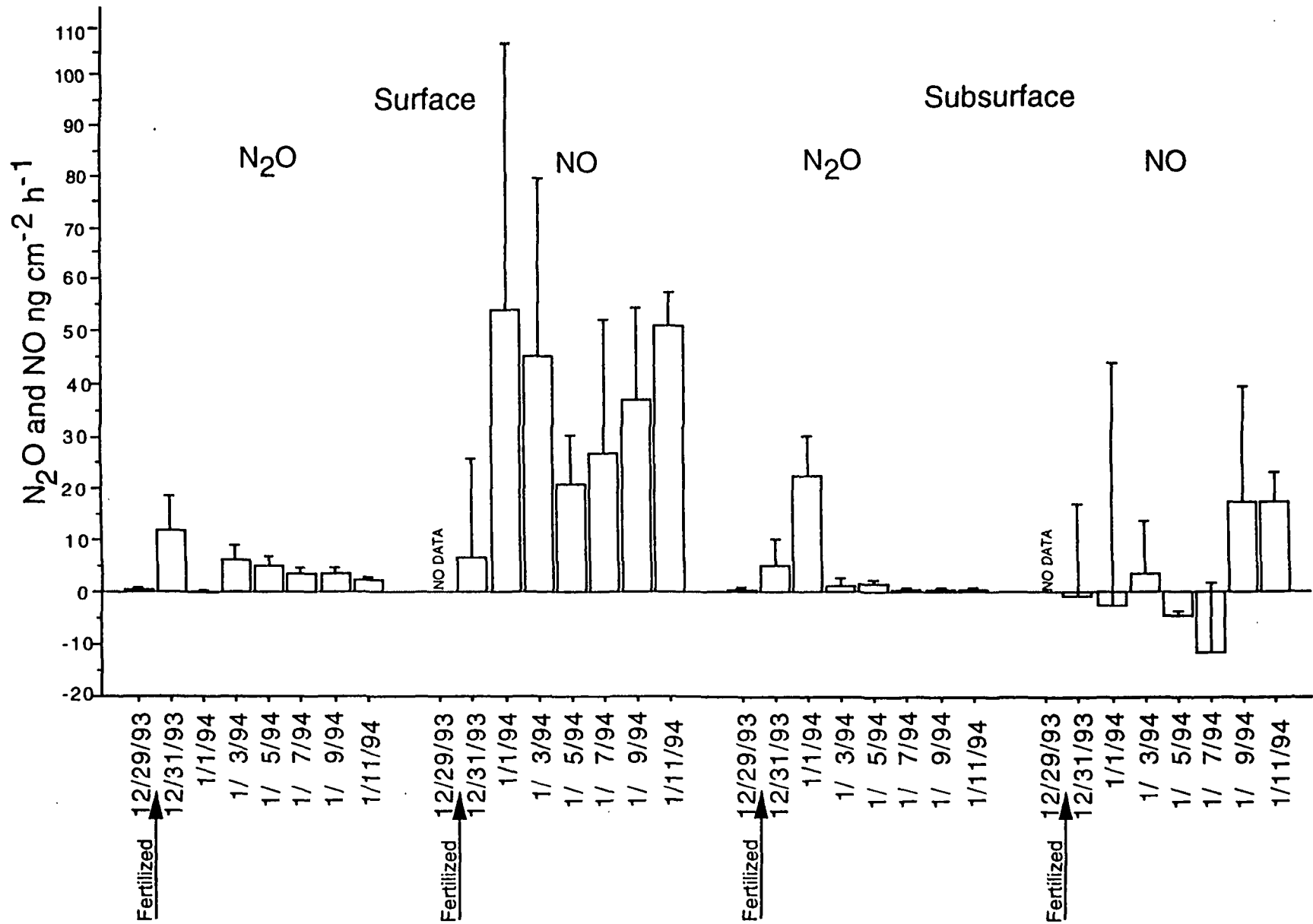


Figure 7c

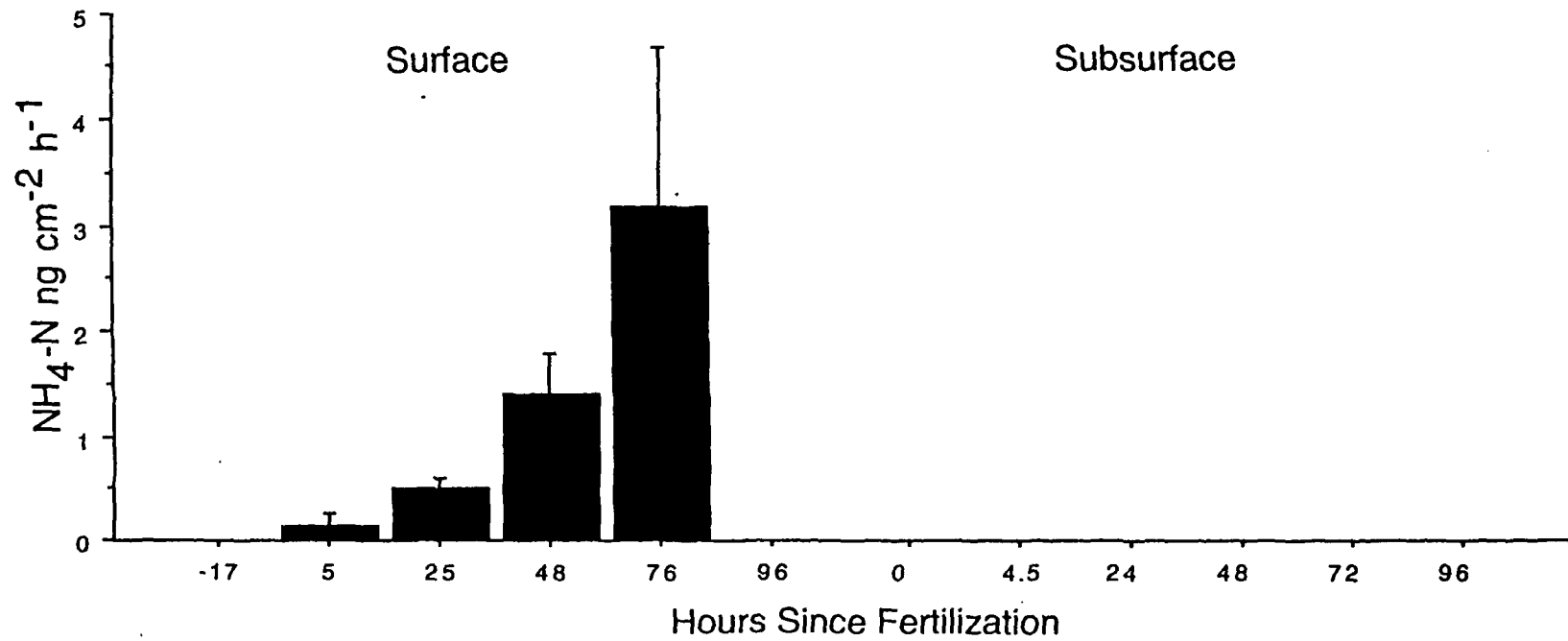


Figure 8.



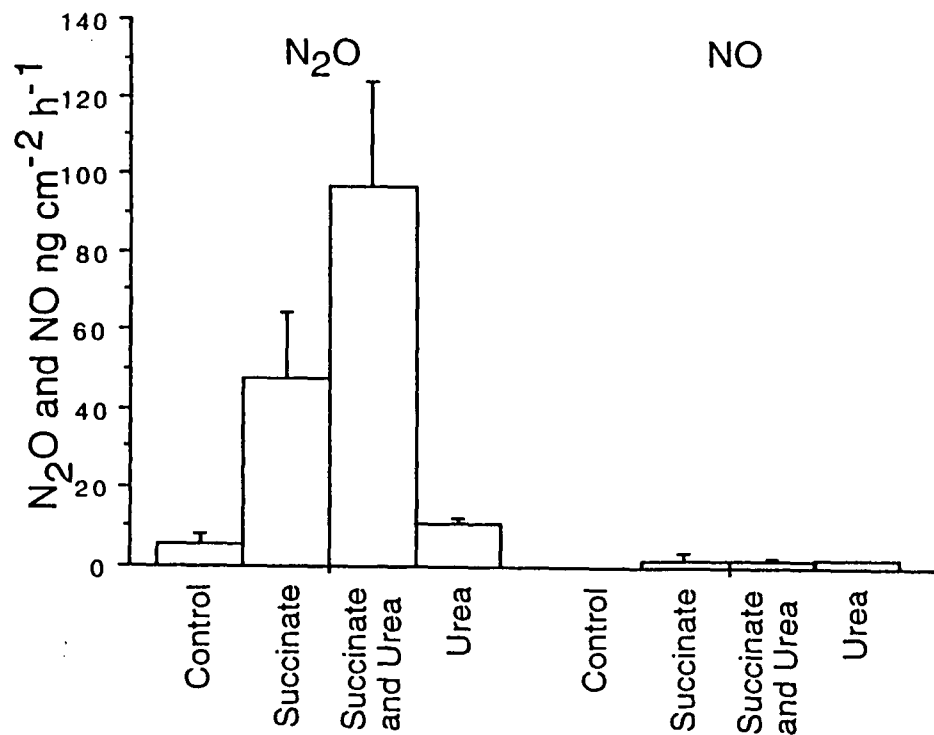


Figure 9a.

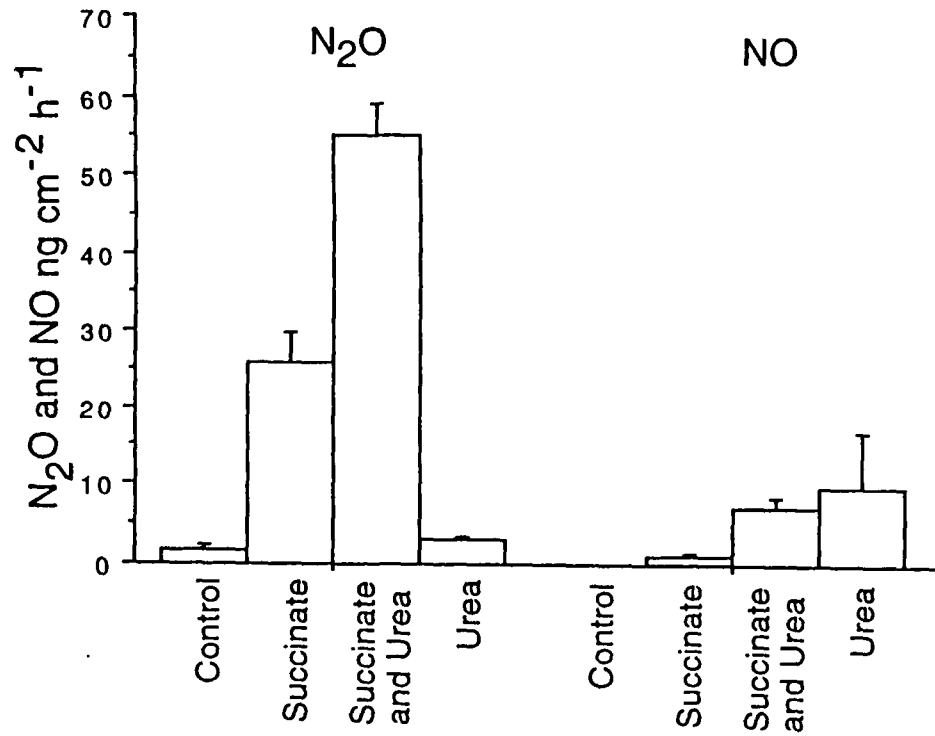


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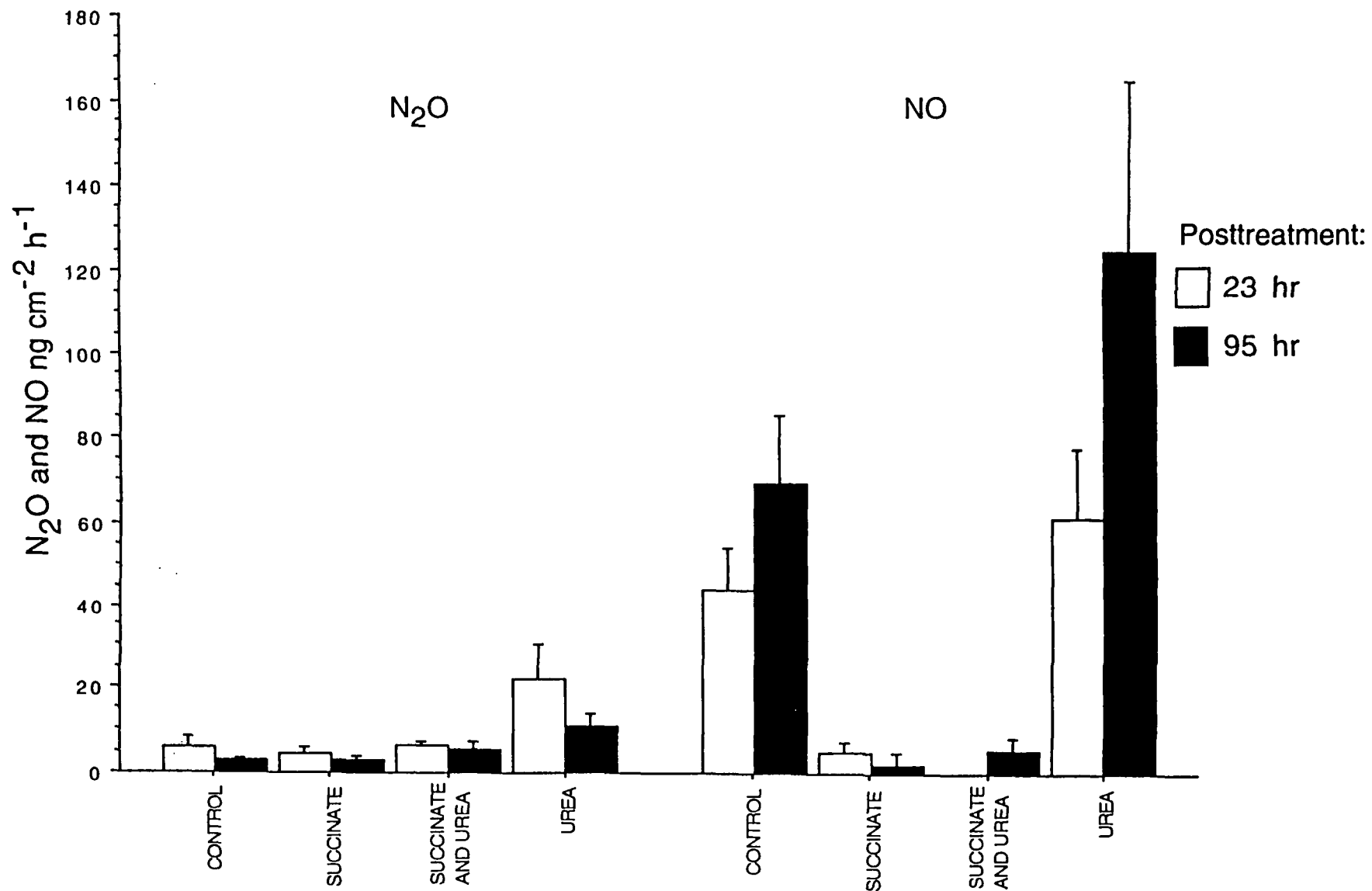


Figure 9c.

Table 1. General description of sugar cane fields located in Maui and Hawaii.

<u>Field*</u>	<u>Location</u>	<u>Elevation (m)</u>	<u>Precipitation (mm)</u>	<u>Soil Order</u>	<u>Soil Classification**</u>	<u>Preparation</u>	<u>Planted</u>
<u>Maui</u>							
312	Windward	256-329.2	1000-1500	Inceptisol	Ustoxic Humitropept	Ripped, disked	6/27-7/1/91
105	Windward	6.1-44.2	1000-1250	Mollisol	Typic Haplustoll	Ripped, disked	10/9-10/13/91 11/12/93
101	Windward	53.3-129.5	1000-1250	Mollisol	Typic Haplustoll	Ripped, disked	9/27/92
100	Windward	97.5-134.1	1000-1250	Mollisol	Typic Haplustoll	Ripped, disked	6/25/93
817	Leeward	24.6-81.5	250-300	Mollisol	Aridic Haplustoll	Ripped	7/30-8/5/92
907	Leeward	7.3-43.3	250-300	Mollisol	Cumulic Haplustoll	Ripped, disked	6/26-6/30/91
823	Leeward	130.5-83.8	250-300	Mollisol	Cumulic Haplustoll	Ripped	1/30/94
908	Leeward	2.7-19.2	250-300	Mollisol	Cumulic Haplustoll	Ripped, disked	8/13/93
<u>Hawaii</u>							
Ka'u 31	S. Slope, Mauna Loa	427	1100	Andisol	Lithic Haplustand	Ripped	11/92
Ka'u 26	S. Slope, Mauna Loa	476	1270	Andisol	Typic Hydrudand	Ripped	5/93
MK 32	Windward, Mauna Kea	152	3810	Andisol	Typic Hydrudand	Ripped	10/92
MK 67	Windward, Mauna Kea	228	3800	Andisol	Typic Hydrudand	Ripped	11/92

\*Maui fields owned and managed by Hawaiian Commercial and Sugar Company (HC&S), Puunene, Maui.

\*Hawaii fields owned and managed by Ka'u Agribusiness and Mauna Kea Agribusiness

\*\*Maui data: Soil Survey for Islands of Kauai, Oahu, Maui, Molokai, and Lanai, State of HI.

\*\*Hawaii data: personal communication, Bob Gavenda, Natural Resource Conservation Service, Kealahou, HI.

Table 2. Operational fertilizer events during which gas flux and soil measurements were carried out.

<u>Field</u>	<u>Date Planted</u>	<u>Date Sampled</u>	<u>Amount Fertilizer Applied (kg/ha)</u>	<u>Age of Sugar Cane (Months)</u>
<u>Maui</u>				
312	7/91	6/92	84	11
817	8/92	12/92	45	4
817	8/92	7/93	35	11
100	6/93	12/93	22	5
908	8/93	4/94	20	9
823	2/94	7/94	39	5
105	11/93	7/94	34	8
<u>Hawaii</u>				
Ka'u 31	11/92	6/93	95	8
MK 32	10/92	7/93	124	10
MK 67	11/92	7/93	94	9

Table 3. Chemical and physical characteristics of soil in fields on Maui and Hawaii.

<u>Field</u>	<u>Soil Texture</u>	<u>Bulk Density</u> (g/cm <sup>3</sup> )	<u>% Nitrogen</u>	<u>% Carbon</u>	<u>pH</u>
<u>Maui</u>					
312	Clay	1.06 (.01)	.14	1.64	5.5
105	Clay*	1.12 (.07)	.13	1.81	6.0
101	Clay*	1.12 (.07)*	N/A	N/A	5.8
100	Silty clay*	1.12 (.07)*	N/A	N/A	5.5
817	Clay*	1.26 (.05)	.12	1.49	7.4
907	Clay loam*	1.15 (.04)	.12	1.39	7.0
823	Silt loam*	1.15 (.04)*	N/A	N/A	7.0
908	Silt loam*	1.15 (.04)*	N/A	N/A	6.9
<u>Hawaii</u>					
Ka'u 31	Silt loam	.70 (.03)	0.71 (0.04)	8.12 (0.28)	4.29 (0.05)
Ka'u 26	Silty clay loam	.65 (0.04)	N/A	N/A	N/A
MK 32	Silt loam	.45 (0.01)	0.28 (0.02)	5.64 (0.14)	5.78
MK 67	Silt loam	.42 (0.02)	0.28 (0.02)	5.48 (0.09)	5.37 (0.03)

% Nitrogen, % Carbon, pH and Bulk density data are means (SE).

\* Soil Survey.

Table 4. Total N<sub>2</sub>O-N and NO-N flux (kg/ha/event) and percent of the applied N lost as N<sub>2</sub>O-N or NO-N during operational fertilization events in Maui and Hawaii.

<u>Field</u>	<u>Sample Date</u>	<u>Sampling Duration (hr)</u>	<u>N<sub>2</sub>O kg/ha/event</u>	<u>N<sub>2</sub>O % loss</u>	<u>NO kg/ha/event</u>	<u>NO % loss</u>
<u>Maui</u>						
312	6/92	96	.011	.013	.012	.014
817	12/92	217	.052	.117	.012	.027
817	7/93	218	.171	.493	.001	.004
100	12/93	240	.013	.059	.008	.038
908	4/94	103	.006	.029	.003	.017
823	7/94	240	.092	.235	.013	.034
105	7/94	144	.113	.336	.004	.012
<u>Hawaii</u>						
Ka'u 31	7/93	695	.381	.401	1.953	2.065
MK 32	6/93	509	1.245	1.001	N/A	N/A
MK 67	7/93	382	.325	.346	.713	.758

Table 5. Prefertilization and postfertilization WFPS (%) by position in fields on Maui and Hawaii: irrigation (ir), cane line (cl), interrow (in).

Field	Position	WFPS %			
		Prefertilization Mean	Postfertilization		Sampling Period Mean
			Min Daily Mean	Max Daily Mean	
<u>Maui</u>					
312	ir	43.5	37.6	43.5	41.0
	cl	34.0	31.1	41.7	38.1
	in	34.9	31.0	34.8	32.6
817(12/92)	ir	53.1	51.0	61.9	55.8
	cl	46.3	47.9	54.6	51.9
	in	39.1	34.6	40.3	37.4
817 (7/93)	ir	57.9	51.0	62.7	56.1
	cl	41.2	31.3	50.6	41.9
	in	28.4	24.2	28.5	26.7
100	ir	47.4	41.8	47.4	42.9
	cl	N/A	N/A	N/A	N/A
	in	N/A	N/A	N/A	N/A
908	ir	32.6	30.9	49.8	37.9
	cl	N/A	N/A	N/A	N/A
	in	N/A	N/A	N/A	N/A
823	ir	48.0	40.6	51.9	44.6
	cl	37.8	32.1	48.6	39.7
	in	22.2	22.9	27.9	25.2
105	ir	40.6	37.3	43.6	40.1
	cl	39.2	29.0	35.0	32.8
	in	36.3	31.2	35.3	32.9
<u>Hawaii</u>					
Ka'u 31	cl	36.6	32.0	38.1	35.8
	in	36.6	33.2	40.1	36.3
MK 32	cl	N/A	24.8	31.2	29.5
	in	N/A	28.3	31.3	29.9
MK 67	cl	29.4	25.7	30.0	28.9
	in	27.4	27.2	29.8	28.6