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AN EDDY-CORRELATION MEASUREMENT OF NO_2 FLUX TO VEGETATION AND COMPARISON TO O_3 FLUX*

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Abstract—Eddy-correlation measurements with a newly developed fast-response NO_x sensor indicate that the deposition velocity at a height of about 6 m above a soybean field has a maximum value near 0.6 cm s^{-1} for NO_x and is usually about 2/3 of that found for ozone. In these studies, over 90% of the NO_x is NO_2. The corresponding minimum surface resistance for NO_x calculated as the quantity remaining after atmospheric resistances are subtracted is about 1.3 s cm^{-1}, which is larger than expected on the basis of leaf stomatal resistance alone. Emission of NO from sites in the plant canopy and soil where NO_2 is deposited and reduced to NO or release of NO_x as a result of biological activity may have lessened the downward fluxes of NO_x as measured. During windy conditions at night, surface resistances are found to have values of about 15 s cm^{-1} for NO_x (again, greater than 90% NO_2) and 1.8 s cm^{-1} for O_3, corresponding to deposition velocities of 0.05 cm s^{-1} and 0.3 cm s^{-1}, respectively.

INTRODUCTION

Contaminants in the lower atmosphere are significantly affected by processes of removal and emission at the surface of the earth. Nitrogen oxides ($NO_x \equiv NO + NO_2$) are of interest because they can cause injurious biological effects and they strongly influence the photochemical production of other potentially-harmful substances, especially oxidants such as ozone. To estimate oxidant budgets for the lower atmosphere, the amount of nitrogen oxides present should be known (e.g. National Research Council, 1977). Also, knowledge of factors that control the amount of NO_x in the atmosphere is important in studies of "acid rain", a subject receiving increasing attention at present.

Processes that control the exchange of trace gases between the atmosphere and the surface of the earth are highly varied, but a limiting factor can often be identified as the ability of the surface to "capture" the trace materials. This ability can thus determine the rate of cleansing of the lower atmosphere, and is highly dependent on the chemical and physical properties of the gases and surfaces considered. Important chemical properties of gases include solubility in water and the oxidizing or reducing potential, while the corresponding surface properties such as moisture content and the presence of easily reduced or oxidized compounds should be considered also.

Gases very soluble in water are usually taken up by vegetation at rates dependent on the sizes of leaf stomatal apertures on a plant canopy with a given number and distribution of stomata, while less-soluble gases are usually taken up more slowly (e.g. Hill, 1971). Highly-reactive gases, such as HF are usually removed quite efficiently by the outer leaf surfaces as well as through stomata (Benedict *et al.*, 1965; Jacobson *et al.*, 1966). However, there are many exceptions to such generalizations. The removal of ozone by vegetation, which is very weakly soluble in pure water and highly reactive with many surfaces, is rapid and controlled mainly by stomatal variations (Rich *et al.*, 1970; Bennett *et al.*, 1973; Wesely *et al.*, 1978; Leuning *et al.*, 1979). Apparently, ozone reacts strongly with compounds in the leaf (Thomson *et al.*, 1966; Heath, 1975). For carbon dioxide, it is well known that removal by vegetation is somewhat controlled by the size of stomatal apertures, but is also lessened considerably by mesophyllic resistances dependent on processes of metabolism and photosynthesis.

The nature of NO and NO_2 deposition to vegetation in the field has not been determined yet. The major global source of NO_x is natural biological action, but locally high levels are usually associated with man-made sources. Due to photochemistry, NO_2 usually predominates during the daytime. Both NO and NO_2 can affect plants, but NO_2 has more lasting, less reversible effects (Bennett and Hill, 1975). Chamber work by Bennett and Hill has shown that NO is taken up very slowly by alfalfa, while NO_2 is taken up rapidly. The uptake of NO_2 by wet surfaces should be

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approximately as large as that for highly-soluble gases such as SO_2 because NO_2 dissolves and reacts quickly in water to form nitrous and nitric acid. Another interesting feature found in the chamber work is that NO seems to be emitted from alfalfa exposed to NO_2, possibly because of conversion of NO_2 to NO in the chamber, perhaps by the vegetation (Hill, 1971).

This work is a continuation of efforts to measure and parameterize the dry-deposition rates of various atmospheric contaminants in natural conditions. The nature of the vertical flux of NO_x, mostly NO_2 during this experiment, above a common agricultural surface, soybeans (*Glycine max.* L. Merr.), is investigated. Comparisons with simultaneous measurements of ozone flux are made. The eddy-correlation techniques used and the theoretical approach applied are largely explained elsewhere (Wesely and Hicks, 1977; Wesely *et al.*, 1978), and are only briefly outlined here. The main limitation to the study of NO_x flux by eddy correlation in the field is the lack of a suitably sensitive and fast-responding sensor for the gas. The sensor used in this study is the first designed specially for eddy-correlation measurements.

EXPERIMENTAL PROCEDURES

Eddy-correlation measurements of the vertical fluxes of momentum, heat, water vapor, ozone, and NO_x ($NO + NO_2$) were taken during August, 1979, near Manheim, in Lancaster County, Pennsylvania, U.S.A. Sampling points were about 5.2 m above the aerodynamic displacement height of a soybean canopy, which was about 0.9 m tall. The soybeans were near a maximum in vigor and amount of ground cover.

The site had been carefully chosen to provide a large uniform surface upwind of the sampling point, in order that measurements be taken in an atmospheric surface layer in "equilibrium" with the surface. Fetch-to-height ratios were greater than 100 during collection of the data reported here. Thus, the vertical fluxes at the sampling height of 5.2 m should have been practically the same as those that would have been found much closer to the surface in representative portions of the field.

Data were collected with a minicomputer and digital-tape system at a rate of $20\,s^{-1}$, and were analyzed in real time as well as later in a more thorough analysis of the tapes. Some of the sensors had notably large response and delay times, near 1 s, so that some corrections were applied to account for the poorly-detected high frequencies that are associated with the vertical fluxes. See Wesely *et al.* (1978) for a brief explanation of the corrections, along with descriptions of the wind, temperature, and ozone sensors. One of the two ozone sensors employed chemiluminescent reaction with NO (Eastman and Stedman, 1977) and one employed chemiluminescent reaction with C_2H_4 (Wesely *et al.*, 1981). A commercially-available Lyman-alpha hygrometer was used to measure humidity fluctuations.

The instrument used to measure the concentration of NO_x employed the chemiluminescent reaction of NO and O_3 (Stedman *et al.*, 1972, and references therein). Upstream of the instrument and near the sampling point, a converter (containing molybdenum metal turnings at 400°C and manufactured by Columbia Scientific*) was used to reduce NO_2 in the sampled air to NO. The NO detection system was specially designed to provide sensitivity and detection limits much lower than commercial NO_x detectors. The system delay time in the field was 0.6 s and the response time 0.15 s. Efficient mixing of sample air and ozone was accomplished in a reaction chamber designed by M. McFarland of the NOAA Aeronomy Laboratory, Boulder, CO (McFarland *et al.*, 1979). The intensity of light from chemiluminescence was measured by a red-sensitive photomultiplier tube (EMI 9658A*) maintained at −50°C, and operated in the pulse counting mode. The detector background signal, or "zero" level, was obtained by altering the flow route so that the O_3 and sample air were mixed in a pre-reactor vessel, so that chemiluminescence was completed before the sample air entered the reaction chamber and thus was out of view of the photomultiplier tube.

The sample air flow was limited by a glass capillary intake. At the typical sample flow rate of $4\ell\ \mathrm{min}^{-1}$, the pressure in the inlet plumbing was close to that in the reaction chamber itself, less than 1333 Pa (10 torr). The low pressure in the inlet minimized losses of NO_x to the tubing walls. The conversion efficiency was found to be stable and greater than 97%, at concentration of 1–30 ppb. This measurement of efficiency used a NO/O_3 titration technique which is independent of flow rates and of absolute concentrations of source gases (Stedman, 1976). Knowledge of the conversion efficiency allowed "absolute" standards of both NO and NO_2 to be used in calibration.

An important aspect of this detector is that, while the chemiluminescence intrinsically measures NO, use of the convertor in the intake system causes the sum (NO_x) of NO and other materials which can be converted to NO to be detected. If vegetation or other surface materials were oxidized by NO_2 and some of the NO_2 thus reduced to NO re-emitted, then a lessened net NO_x flux would be measured. In fact, the re-emitted NO would react rapidly with normal ambient ozone to produce NO_2; the lifetime of NO emitted into air with 50 ppb of ozone is about 50 s. Thus, some of the conversion would take place below the eddy-correlation sampling point, causing the apparent ozone flux at that height to increase slightly and the NO_2 (as well as NO_x) flux to decrease. Of course, other sources of NO at the surface, such as bacterial production in the soil, would also lessen the measured downward flux of NO_x.

It is possible that the O_3 and NO_x sensors could interfere with each other because the NO_x sensor produces large excesses of O_3 and the O_3 sensor produces excess NO_x. However, the outlets for these unwanted gas products are easily placed downwind of the sampling points, thus avoiding the problem.

RESULTS AND DISCUSSION

Fluxes and concentrations

Figure 1 summarizes the measurements, which encompass nearly 24 h of data collection. A full diurnal cycle of measurements is desirable in order to study the entire range of resistances associated with soybean leaf stomata which, as is well known, close at night and open in the daytime. For the night considered, the winds were sufficiently strong to allow successful use of the eddy-correlation apparatus; very light winds at night often result in greatly damped turbulence confined to relatively high frequencies, which prevents effective use of the rather slow-response instruments for eddy-correlation. Also, with light winds and strong surface cooling at night, the formulations currently available to describe flux-gradient relationships in the

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* This does not connote approval or recommendation of the product by sponsors of this work, Argonne National Laboratory, or the University of Michigan, to the exclusion of other products which may be suitable.

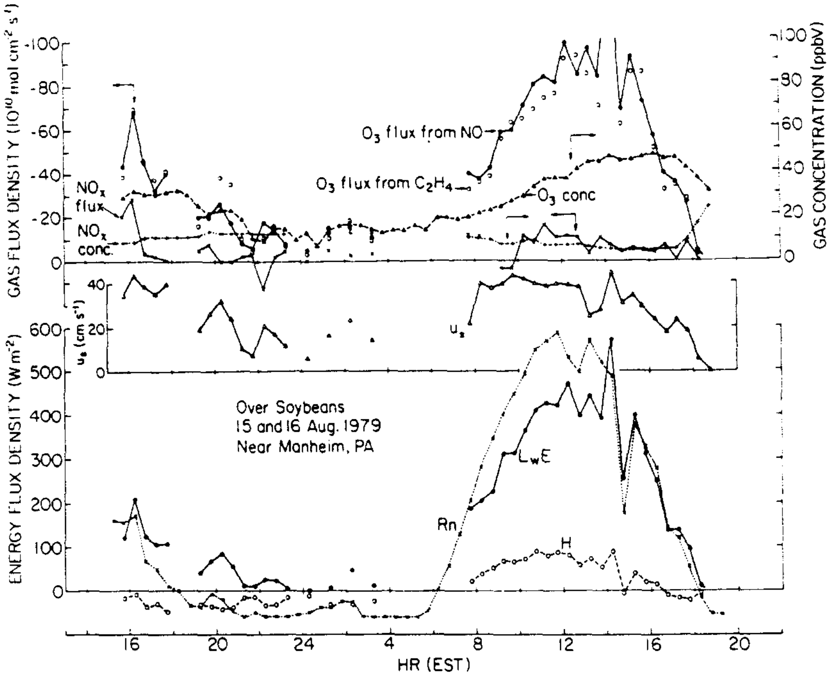

Fig. 1. Summary of half-hour averaged measurements. Straight lines connect sequential data points except for the O_3 flux from C_2H_4 where the lines are omitted for clarity. In addition to some of the terms described in the text, u_* is the friction velocity, H the sensible heat flux, L_wE is the latent heat flux, and Rn the net radiation.

atmospheric surface layer are usually inadequate.

The energy balance terms shown in the lower portion of Fig. 1 depict a rather common situation for well-watered soybeans under mostly cloudless skies. The large variations evident between 1200 and 1600 h Eastern Standard Time (EST) are due to scattered clouds. The latent heat flux L_wE is large and the sensible heat flux seems suppressed during the daytime, which is suggestive of heat advection. In the authors' experience, such a case of mild heat advection is not unusual for soybeans with a good supply of soil moisture; it is not known whether this was large-scale advection or local advection from the nearby fields.

The ozone concentrations are moderately small, peaking at nearly 50 ppb by volume. The measured amount of NO_x (NO plus NO_2, expressed in units of ppb of NO_2) is rather small, usually less than 13 ppb. Measurements of NO taken directly by bypassing the converter in the NO_x sensor at intervals of a few hours showed that concentrations of NO are considerably less than 10% of the NO_x for the present data. Nitric oxide is a significant fraction of the total only after 1800 h on 16 August when winds became very light and the concentration of NO_x increased rapidly. We assume for the present that all the NO_x was NO_2 for this analysis, and do not use the data for after 1830 h on the 16th.

The two ozone sensors provide nearly identical values of the vertical flux, which quite closely parallel the trends in L_wE. The flux of NO_2 at times has similar trends, but is occasionally directed upward, a confusing factor. While the possibility that the surface was a source of NO_x at times cannot be totally discarded, it is likely that the "noise" inherent in this preliminary version of the NO_x sensor caused some spurious readings that were particularly strong at these times. Indeed, most of the variability in the NO_2 fluxes from 1/2 h to 1/2 h does not seem to be correlated with any of the other factors considered, and thus could be due to the noisiness of the sensor. For frequencies greater than 1 Hz, a large amount of noise was noted in the output signal of the NO_x sensor. This noise was random in the sense that it was not correlated with vertical wind speed, and thus should not have systematically altered the NO_2 fluxes.

Surface resistances

To eliminate some of the seemingly random variations of NO_2 flux and thus obtain a clearer picture of the processes that control the flux, data are averaged over at least two 1/2 h intervals. Then a residual surface resistance r_c is calculated following the procedures of Wesely and Hicks (1977):

$$r_c = v_d^{-1} - r_a - r_s, \qquad (1)$$

where the deposition velocity v_d is the negative of the ratio of the measured flux (positive when directed upward) to the concentration at the height of measurement, the aerodynamic resistance r_a from the sample point to near the surface is estimated on the basis of some of the micrometeorological flux measurements and inferred surface physical properties, and the boundary-layer resistance r_s of the quasi-laminar air sublayer enveloping surface elements is calculated based on momentum flux and surface roughness. According to Equation (1), r_c is the resistance that remains after all aerodynamic resistances above the surface elements are removed and thus represents the bulk resistance of the plant canopy and soil to uptake, as seen from above. Notably, r_c results from a highly stylized formulation and is difficult to relate precisely to the properties of any one surface element such as leaf, stem, or particular area of soil surface. Similar resistances are calculated for water vapor and ozone, which should mainly represent the bulk canopy stomatal resistance, with perhaps some lessening due to evaporation from the soil or ozone destruction at the soil and outer plant surfaces.

For the night-time data between 1915 and 0315 h, the average surface resistances for H_2O, O_3 and NO_2 are $3.0 \pm 0.74\,s\,cm^{-1}$ for 13 1/2 h samples, $1.8 \pm 0.4\,s\,cm^{-1}$ for 13 samples, and $15 \pm 2\,s\,cm^{-1}$ for 11 samples, respectively. These are logarithmic averages, which are appropriate because of nearly log-normal distributions, as are typically found in resistance data. Also, the two cases of NO_2 flux being directed upward are not used; the value $15\,s\,cm^{-1}$ thus represents a lower limit. These averages indicate that, while evaporation from the soil was significant and ozone destruction at the soil and possibly the outer plant surfaces was rather large, the net flux of NO_2 was severely limited. Hence, it appears that penetration of well-mixed air into the plant canopy and down to the soil surface was substantial, and that ozone is much more easily removed than NO_2 from the air by soil and plants with stomata closed. Of course, there remains the possibility that NO emitted from the soil as a result of microbial activity or of reduction of NO_2 to NO at the surface could have substantially decreased the net flux of NO_x.

Figure 2 shows that values of surface resistances during the daytime of 16 August. There was an inadvertent release of nitrogen oxides in the vicinity of the NO_x sensor at about 0845 h, possibly causing the appearance of an upward-directed flux of NO_2 near 0900 h (see Fig. 1). These cases of upward fluxes are not used in the analysis producing Fig. 2. Further, the raw NO_x data are averaged over more than 1 1/2 h in order to smooth the variations of values of r_c calculated. The overall trends of the three resistances plotted in Fig. 2 are similar and conform to the type of variation expected for bulk canopy stomatal resistance. That is, leaf stomata open fairly quickly in the morning and gradually close during late afternoon. Because of the rather strong winds and probably continued evaporation throughout the previous night, dewfall was probably not substantial on the leaves in the morning. Thus, the resistance for H_2O during the morning and the other times shown in Fig. 2 should be representative of bulk canopy stomatal resistance, but probably lessened a little due to the effects of evaporation from the rather-wet soil surface. During late afternoon, the resistance to ozone removal seems to increase too rapidly, as the average value of $r_c = 1.8\,s\,cm^{-1}$ found for the previous night is exceeded. An adequate explanation has not been found. Similar trends are seen on other days during transition to the very stable conditions that are rather common at night. During such stable conditions, aerodynamic resistances are very large, so that deposition velocities are extremely small regardless of the value of r_c. Thus, it is of little consequence that r_c during night-time strongly-stable conditions is somewhat larger than suggested during night-time well-mixed conditions.

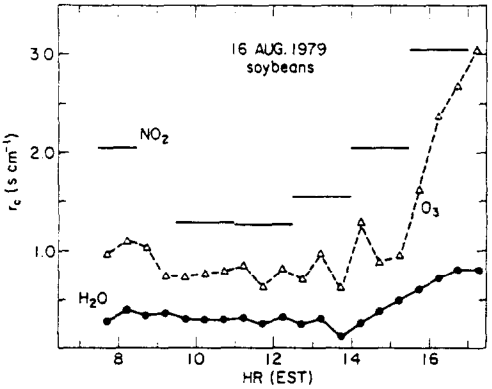

Fig. 2. Summary of bulk canopy resistances to exchanges of NO_2, O_3 and H_2O measured during the daytime.

If the only mechanism controlling the surface resistance to H_2O and O_3 flux were stomatal resistance, the differences in the rates of molecular diffusion in air through leaf stomatal openings should account for the differences in H_2O and O_3 resistances. That is, the ratio of diffusivities (1.64) multiplied by r_c for H_2O should equal r_c for O_3. However, r_c for O_3 is substantially greater, which indicates that evaporation from the soil might be quite large, removal of O_3 at the soil surface might be weak, or that O_3 might be destroyed with considerably less than 100% efficiency at surfaces exposed in substomatal cavities. This situation is in contrast to that found recently for maize in drier soil, where ozone removal was considerably greater than expected on the basis of H_2O resistance (Wesely *et al.*, 1978).

Surface resistance to NO_2 removal is clearly much larger than would be expected if uptake through leaf stomata were limited only by molecular diffusion (NO_2 has about the same diffusivity in air as O_3). This could imply that mesophyllic resistance to NO_2 uptake is substantial, at least $0.5\,s\,cm^{-1}$, if the difference in Fig. 2 between the r_c's for O_3 and NO_2 is taken as a guide.

Chamber work by Hill (1971), however, suggests that NO_2 uptake by vegetation is rapid, is limited mainly by stomatal resistance rather than mesophyllic resistance, and might be accompanied by substantial NO emission. It is possible that the situation in the present study is similar. Research on different types of surfaces suggest that re-emission of a large portion of deposited NO_2 as NO might be fairly common (Judeikis and Wren, 1978). Also, soil bacteria release NO, perhaps at a greater rate during the daytime than at night because of warmer temperatures more favorable to microbial activity during the daytime. Let us assume, for example, that NO is released in substantial quantities from the vegetation and soil and is all rapidly converted to NO_2 in the atmosphere very close to the surface. Further, let us assume that the actual value of r_c for NO_2 is $0.7\,s\,cm^{-1}$ near noon rather than the value of about $1.3\,s\,cm^{-1}$ measured. This lower value should be a typical minimum value of bulk canopy stomatal resistance for gases with a diffusivity equal to that of NO_2 (Wesely and Hicks, 1977). It can then be calculated that roughly 50% of the NO_2 initially removed from the surface would seem to reappear in the atmosphere due to surface emissions. Even if very little of the NO emitted were converted to NO_2 by the time a height of several meters is reached, this mechanism of emission is still quite feasible since the sensor measured total NO_x, accomplished by conversion of any NO_2 to NO within the system. Conversion of NO to NO_2 at greater heights would still keep the concentration of NO as small as was observed.

Deposition velocities

Deposition velocity, found as the ratio of the downward component of flux to the concentration at a specified height, incorporates the effects of both aerodynamic and surface properties, and thus is often more difficult to describe in general terms than a variable such as r_c that is meant to reflect only surface properties. Nevertheless, numerical models often use estimates of deposition velocity was an expedient to determine total removal at the surface from the atmosphere. For this reason, a brief summary of deposition velocites found in this experiment is appropriate. The nighttime average values found at a height of 5.2 m above the soybean field during 15 and 16 August are $0.29\,cm\,s^{-1}$ for ozone and $0.05\,cm\,s^{-1}$ for NO_2 (which again neglects the two cases of NO_x flux directed upward). During the daytime, the deposition velocity has a maximum value of about $0.56\,cm\,s^{-1}$ for NO_2 and about $0.84\,cm\,s^{-1}$ for O_3. These and other estimates for the deposition velocity during daytime are easily calculated from the values of r_c shown in Fig. 2, with the additional information that the bulk gas-phase aerodynamic resistance above the leaves totals about $0.5\,s\,cm^{-1}$ between 0800 and 1600 h on 16 August. That is, deposition velocity is given approximately as $(r_c + 0.5)^{-1}$.

A crude means of parameterizating deposition over large areas by use of r_c has been suggested recently by Sheih *et al.* (1979), for application in numerical models. It is assumed that aerodynamic resistances will be calculated independently in order to derive deposition velocities. Table 1 summarizes the present results in a form similar to that suggested by Sheih *et al.* Since these results are from only one day of measurements, further confirmation of the values in Table 1 is highly desirable. Of course, many types of surfaces need to be considered in order to consider large areas.

CONCLUSIONS

The removal of NO_2 from the atmosphere by a full-canopied soybean field is limited by a bulk surface resistance that is quite large at night, near $15\,s\,cm^{-1}$, and during daytime is 1.5–2.0 times as large as that for ozone. Ozone destruction at the field surface is rather large, corresponding to a calculated residual resistance of about $1.8\,s\,cm^{-1}$ at night, which probably indicates a substantial uptake by the soil surface, and during daytime is near that expected if ozone were nearly perfectly removed at inner leaf surfaces. The corresponding deposition velocities are about $0.05\,cm\,s^{-1}$ for NO_2 and $0.3\,cm\,s^{-1}$ for O_3 at a height of 5–6 m during windy conditions at night, and vary to maximum values of slightly less than $0.6\,cm\,s^{-1}$ for NO_2 and slightly more than $0.8\,cm\,s^{-1}$ for O_3 during daytime. Actually, NO_x was measured rather than NO_2, but the NO portion was small enough, less than 10%, to be ignored in the present experiment.

The rather small deposition velocity for NO_2 during the daytime suggests that there might be a substantial mesophyllic resistance to NO_2 uptake amounting to at least $0.5\,s\,cm^{-1}$. This is somewhat unexpected since NO_2 dissociates rapidly in water and presumably also in the watery solution surrounding leaf cells. Apparently, the ability to be taken up by water can be secondary in importance to reactivity with inner leaf surfaces. Chamber work by Hill (1971) suggests that NO_2 is taken up quite effectively by alfalfa and is

Table 1. Summary of data in the generalized fashion suggested by Sheih *et al.* (1979), for the soybean field studied. (The wind speed $\bar{u}$ is for a height of about 5 m above the canopy, L is the Obukhov scale length and r_c is the surface resistance)

Measurement period (h)	Stability category	Stability	L (m)	$\bar{u}$ ($m\,s^{-1}$)	r_c for O_3 ($s\,cm^{-1}$)	r_c for NO_x ($s\,cm^{-1}$)
0730–1600	A, B, C	unstable	-61 ± 9	3.7 ± 0.1	0.84 ± 0.04	1.6 ± 0.2
1600–1800	D	near-neutral	large	2.2 ± 0.1	2.4 ± 0.5	2.8 ± 1.6
1900–0330	E	slightly stable	15 ± 5	2.3 ± 0.1	1.8 ± 0.4	15 ± 2

largely controlled by stomatal resistance, but that the uptake of NO_2 seems to be accompanied by a release of NO. It is possible that this mechanism lowered the apparent net uptake of NO_2 (measured as NO_x in the present experiment). If so, then as much as 50% of the NO_2 initially removed from the atmosphere during the daytime could have been released as NO by the soybeans. More generally, any source of NO_x, whether from biological activity or from reduction of NO_2 to NO and subsequent entry into the atmosphere, could have caused the rather low values measured.

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