P2.13 NOx Production in Laboratory Simulated Blue Jet and Sprite Discharges

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1. INTRODUCTION

There are high concentrations of NO_x present in the middle atmosphere (Callis 2002). For example, Gordley et al (1996) found NO_2 mixing ratios of 1 ppb at 30 mb, increasing to 8 ppb at 10 mb. NO_2 vertical column densities in the stratosphere are greatest in the polar daytime. In contrast, they are lowest in the polar winter and in the tropics (Wenig 2004). The source of this NO_x is currently unknown. N_2O reacting with oxygen atoms has been looked at as a potential source of stratospheric NO_x (Brasseur 1986). Transport of NO_x across the tropopause has also been examined, though Kasibhatla et al (1991) found that tropospheric NO_x levels were not greatly influenced by stratospheric NO_x production. A general consensus is that stratospheric NO_x is dominated by processes other than transport from the troposphere. Some of the more stable NO_y compounds (NO_y includes NO, NO_2, NO_3, HNO_3, $ClNO_3$, N_2O_5, and HNO_4) may decompose to NO_x following transport across the tropopause (Strand 1994). Energetic electron precipitation, as caused by geomagnetic storms in the magnetosphere and upper ionosphere, is

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thought to contribute to NO in the mesosphere and lower thermosphere (Callis 2002). In this paper, we provide experimental evidence pointing to blue jets, red sprites, and other transient luminous
events (TLEs) as a source of middle atmosphere NO_x

NO_y found above 55 km is primarily NO (Callis 2002). NO_x present in the mesosphere and lower thermosphere is then transported to the stratosphere during polar winter, as a result of the global meridional circulation. At the same time, NO_x in the stratosphere plays an important role in catalytic ozone loss (Funke 2005). Thus, any NO_x produced in the mesosphere by TLEs would be partially responsible for ozone destruction in the stratosphere.

TLEs include a wide variety of electrical discharges occurring at heights between the tops of thunderstorms, up to and including the ionosphere (see figure 1). Blue jets begin around 10 km, and extend from the tops of thunderstorms to as high as 40 km or more. A related phenomenon, gigantic jets, extends upward from the cloud top to the ionosphere, as described by Pasko et al (2002 A) and Su et al (2003). Red sprites occur higher up, extending to the ionosphere. They begin as high at 90 m, and extend as far downward as 40 km (Lyons 2003).

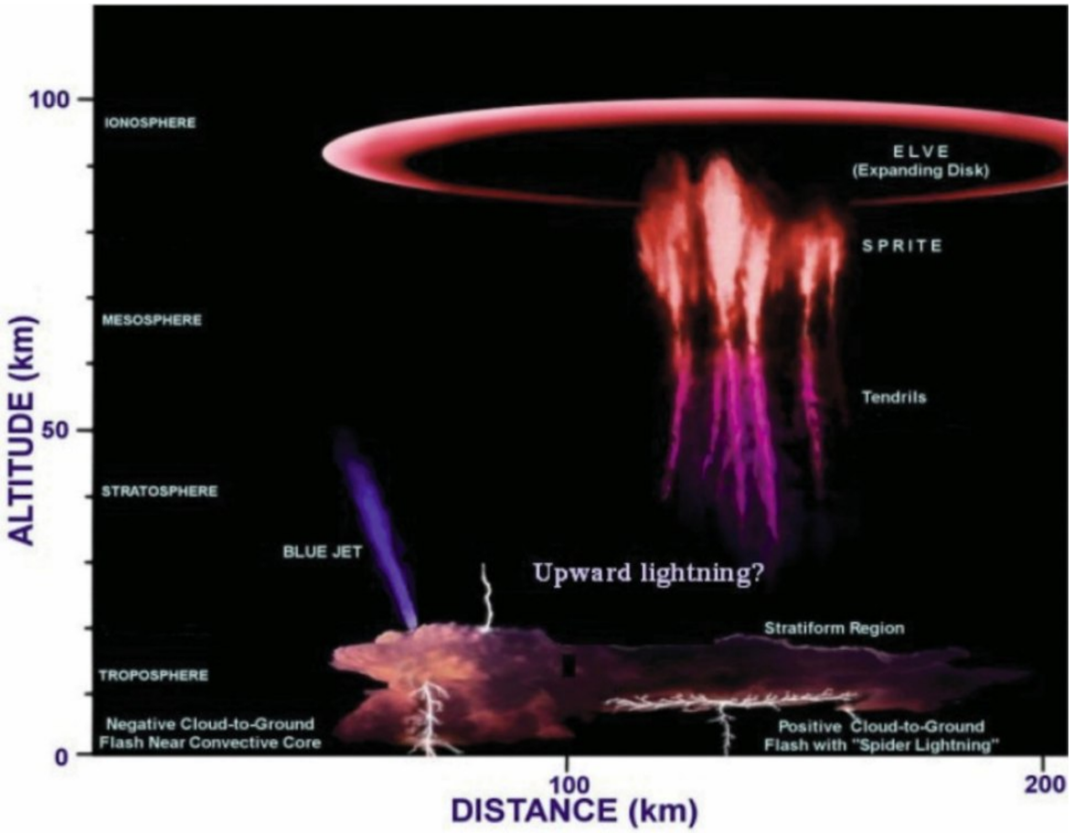


Figure 1. Summary of transient luminous events (TLE's) from Physics Today article.

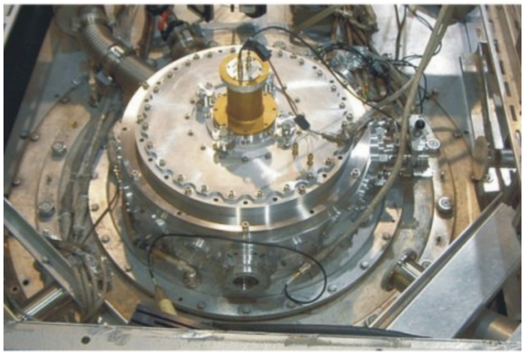
Simulations of upper tropospheric and stratospheric electrical discharges have been conducted in the laboratory at pressures from 1-500 mb in an attempt to determine the contribution of TLE's to middle atmospheric NO_x. Chemiluminescence was used to detect NO_x produced in these discharges as a function of discharge current and energy, as well as measuring the ratio of NO to NO_2. It will be shown that while total NO_x production per TLE is lower than for typical tropospheric lightning events, the production per unit of energy is greater for discharges at pressures down to 10 mb. TLE discharges are much more spatially distributed, involving much lower current densities than typical intracloud and cloud-to-ground discharges. Hence the increased production of NO_x per unit of energy with decreasing pressure may indicate that TLE's are a significant source of stratospheric NO_x.

2. EXPERIMENTAL METHOD

The experimental setup consisted of an evacuated discharge chamber, a 50 kV DC power source, discharge circuitry, and a chemiluminescence NO_x detector. The chamber (figure 2) consists of two large plates and an acrylic chamber with a volume of 10.4 L. A conical brass electrode on the bottom plate (cathode) and a thick piece of copper wire attached to the grounded top plate formed a 4 centimeter discharge gap in the center of the chamber. Discharge currents ranged from a 350 amps at 500 mb to a few amps at 1 mb. Currents were limited to avoid tortuosity in the discharge channels so that measured NO_x production was for an approximately fixed discharge channel length. The chamber is designed to fit on top of the "Zebra" machine (figure 3) at the Nevada Terawatt Facility at the University of Nevada, Reno, a high energy discharge device which can provide discharges of 100's of kiloamps at up to one million volts. Future high energy discharges with Zebra are currently in the planning stage. Experiments investigating discharges in the presence of water droplets, ice crystals (the chamber can be

Figure 2. Discharge chamber and NO_x analyzer used in the study.

cooled to -30 ^{o}C or so) and aerosols are also planned.



The DC power source was used to create electric fields with voltages between 0 and 35 kV. The electric field could be built up slowly over any specified period of time until breakdown occurred, or could be pulsed with a high voltage relay connected to a capacitor that could send the voltage to the bottom plate of the chamber all at once, resulting in current rise times of approximately 5-50 microseconds. The pulsed approach was used for most of the measurements presented here because larger currents could be obtained by this method.

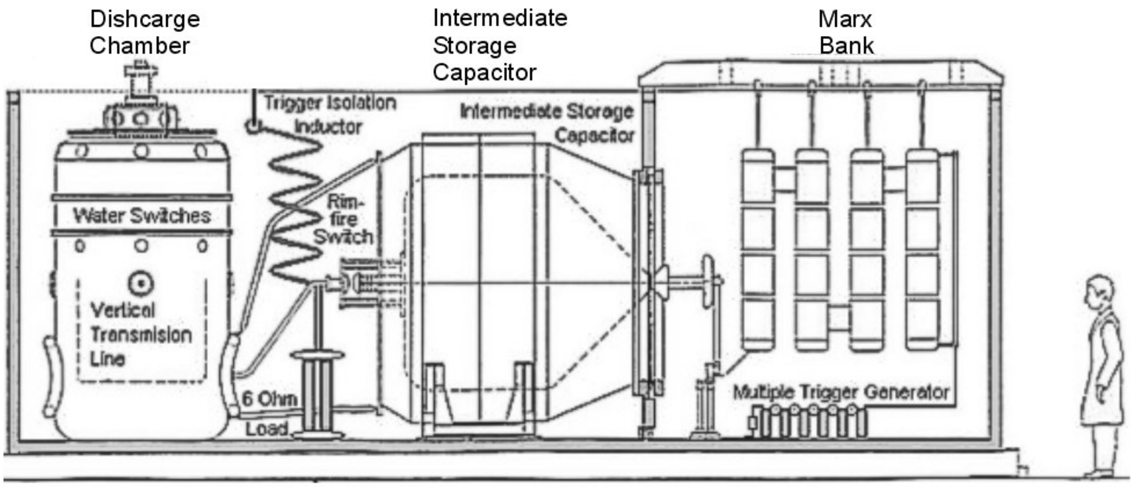


Figure 3a and 3b. Figure 3a (above) is the discharge chamber on top of the "Zebra" machine at the Nevada Terawatt Facility; figure 3b is a schematic of Zebra.

Using this setup, TLEs were simulated by producing sparks and glow discharges in the chamber for pressures of 1-500 mb. A transition region where chamber pressures were between 100 and 5 mb produced sparks that were a mix of blue and red rather than white as seen in figure 4, but which otherwise had similar visual characteristics to discharges at higher pressures. In sprites, the red light is emitted by excited nitrogen molecules (Inan 2002). Specifically, the N2 1 PG (3-1) transition emits photons of wavelength 762.7 nm (Yair 2004). In blue starters, a type of TLE related to blue jets, the blue color is from the 427.8 nm first negative transition in N_2^+ molecules (Pasko 2002 B). Occasionally, a faint orange glow (assumed to be excited NO) was observed near the copper wire. These discharges were bluish in color around 3 mb, and tended toward a mix of blue and red down to 0.5 mb. Observations show a transition occurs in sprites from a lower streamer region, to a transition region with a mean altitude of 78.2 km and one to two km in depth (Gerken 2005), to an upper diffuse region (Pasko 2002 C???). While the chamber pressure in these discharges is higher than in the transition region of sprites, the current densities are similar.

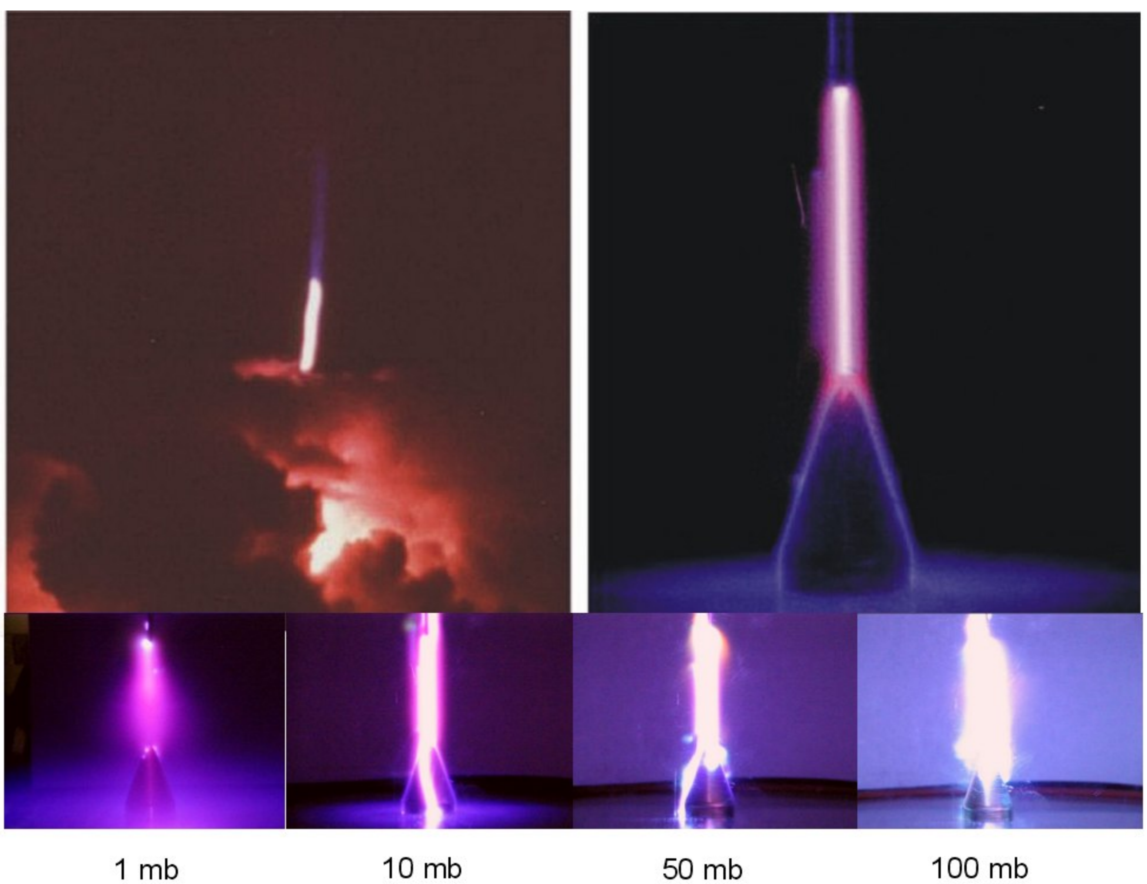


Figure 4. (upper) Image of a blue jet compared with a CCD camera image of a discharge at 10 mb; (lower) CCD images of discharges at various pressures and currents.

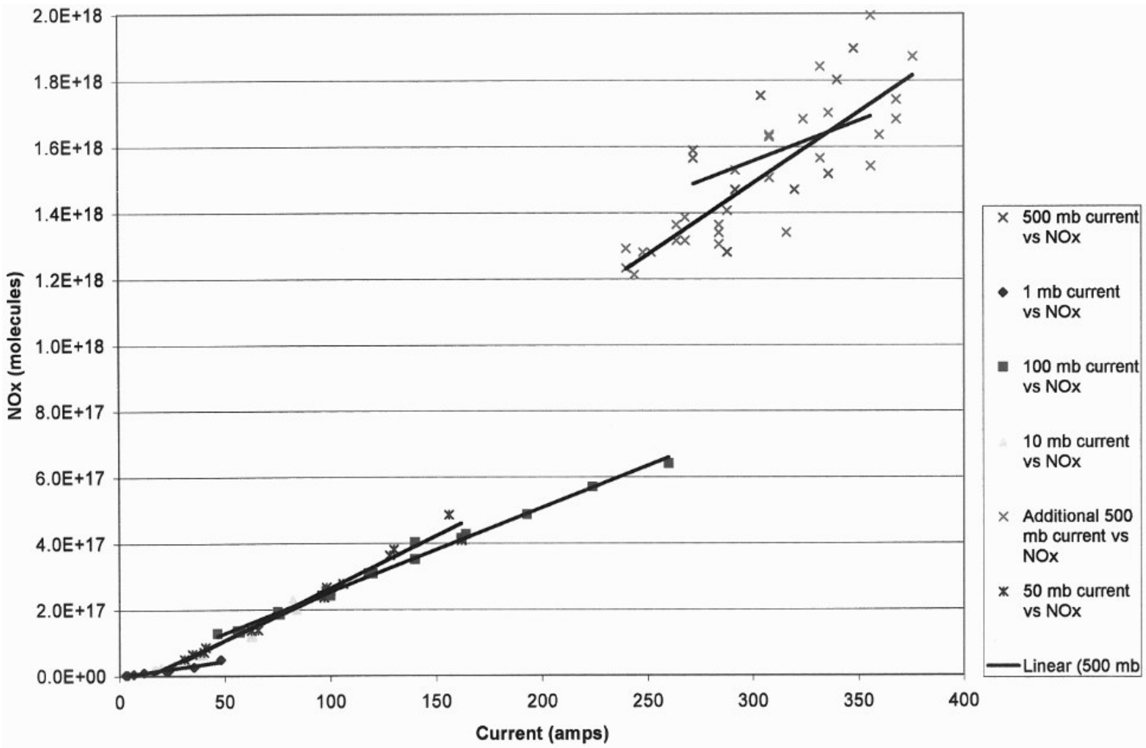


Figure 5. Comparison of NOx production versus current for pressures of 1-500 mb..

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NO_x was detected using a Monitor Labs 8440 chemilumi-nescence NO_x detector with a maximum detection level of 5ppm (with this type of analyzer, NO and total NO_x is measured, with NO_2 determined by differencing these two measurements). Following a discharge, the chamber was quickly brought up to room pressure, and then the diluted contents were sampled by the NO_x detector through a short length of Teflon tubing. Sampling typically occurred within 30-60 seconds following discharge and dilution. Detected NO_x levels remained relatively constant for the 30 seconds or so that were required for the detector to provide a stable measurement (about 5 detector time constants). The response of the NO_x detector, and uncertainties resulting from dilution, were checked by introducing a calibration gas to the chamber of 5.0 ppm of NO_x in nitrogen, and then simulating a dilution and NO_x measurement cycle. Uncertainties in NO_x measurements were typically a few percent, approximately within the precision and stability of the NO_x detector, except for the largest current discharges at 500 mb. At 500 mb and a few hundred amps of currents, NO_x levels exceeded 5 ppm, so the dilution had to be performed twice in order to bring the NO_x levels within the detection range of the analyzer. This process was also checked with the calibration gas and found to involve only a slightly larger uncertainty.

3. RESULTS AND DISCUSSION

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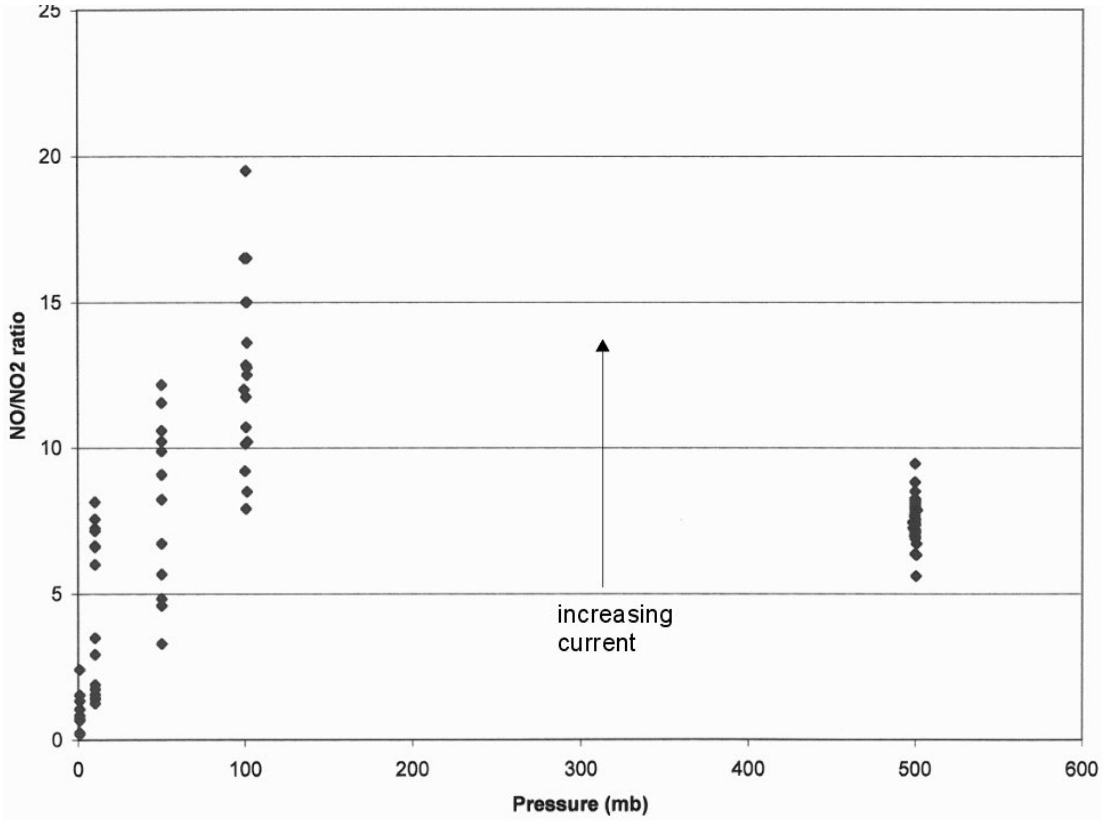

Figure 6. Mixing ratios of NO to NO_2 as a function of pressure and increasing current..

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A summary of NO_x production as a function of pressure and current is shown in figure 5. Discharges taking place at pressures below 100 mb follow a linear relationship between current and pressure, while two sets of discharges at 500 mb followed a different linear relationship. The linear fits for discharges at 100 mb and below had R-squared values from 0.95 all the way up to 0.99, while the 500 mb data had an R-squared value of 0.76. The larger variation at 500 mb is most likely a reflection of the increased tortuosity observed in the discharges at high current levels, the actual length of the discharge somewhat exceeding the 4 cm electrode gap length. Since there were no discharges conducted at pressures between 100 and 500 mb, the nature of NO_x production as a function of current in this transition region has yet to be determined. The results of this initial study suggests a nonlinear relationship.

It was also noted that discharges produced a greater fraction of NO_x as NO as pressures decreased from 500mb to 100 mb, but, this ratio decreased for pressures lower than 100 mb (figure 6). At 10 mb the ratio of NO to NO_2 was about 5:1 which contrasts with measurements showing mesospheric NO_x to occur primarily as NO (Callis 2002). The presence of corona in the lower pressure discharges, as visible in Figure 4, could have generated ozone and subsequently converted NO to NO_2.

The NO production rate as a function of discharge energy is shown in figure 7. The figure includes production as a function of stored energy in the discharge capacitor and production as a function of energy dissipated in the actual spark current. Wang et al. (1998), in their analysis of laboratory NO_x production in simulated large cloud-to-ground discharges, noted a serious problem with results reported in the literature from various past experiments which involved scaling NO_x production to stored energy, e.g in a capacitor bank, rather than to the actual energy deposited in the spark or discharge channel. At large voltages and high current levels, there are significant inductive and resistive losses in the discharge circuit external to the electrode gap. In such cases, the majority of stored energy can be dissipated in the external circuit, and not in the actual spark. Dividing production by the stored energy as

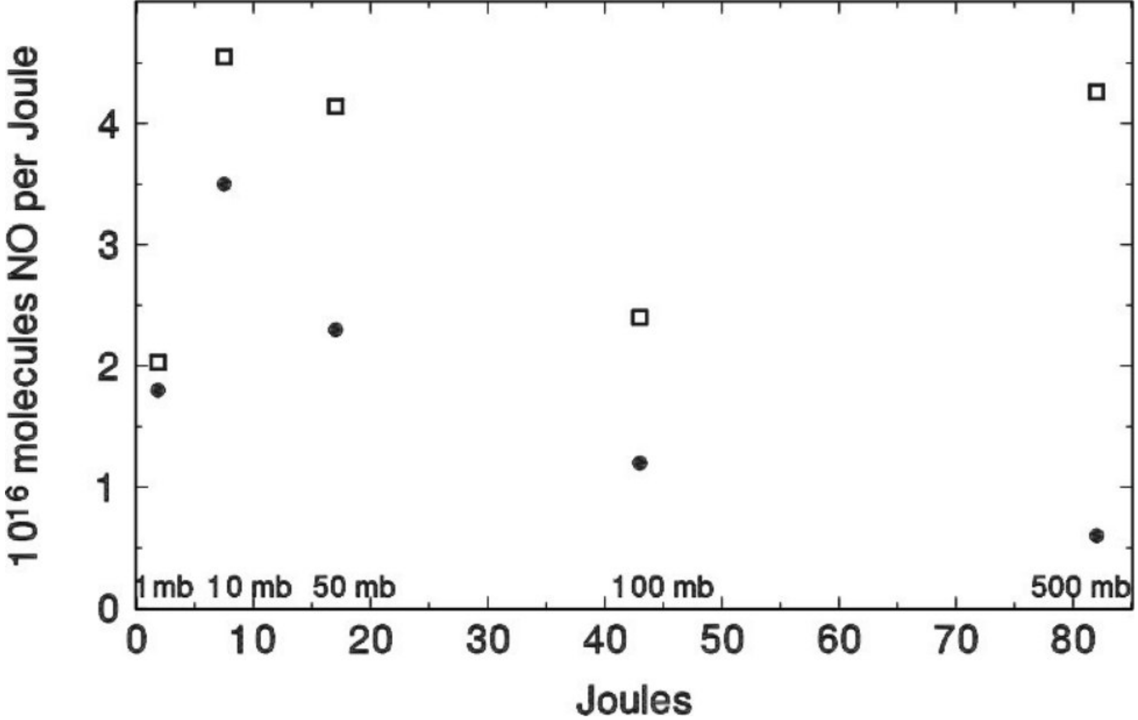

Figure 7. Production rate of NO as a function of energy. Production rate as a function of stored energy prior to discharge is represented by open squares, while the rate corrected for circuit losses during discharge are shown as solid circles, representing the production as a function of energy actually deposited in the spark channel.

opposed to actual energy deposited in the spark leads to an underestimation of the production rate.

This effect is clearly seen in figure 6 where current wave forms measured with an oscilloscope were used to perform standard RCL circuit analysis in order to calculate circuit losses. The correction is particularly significant at 500 mb where that largest currents and discharge voltages were investigated. When this correction is taken into account, more NO_x per Joule of discharge energy is produced by discharges at stratospheric pressures than those at tropospheric pressures.

4. CONCLUSION

NO_x produced by TLEs potentially plays an important role in stratospheric ozone depletion. Therefore it is desirable to estimate this production through laboratory simulations. A relationship for NO_x production as a function of current was found for discharges at stratospheric pressures which differed from the relationship found for discharges at tropospheric pressures. While discharges at stratospheric pressures produce less NO_x than those at tropospheric pressures, they actually produce more NO_x per Joule of discharge energy. Discharges at 10 mb were found to produce more than three times as much NO_x per Joule as compared with discharges at 500 mb. In the future, it will be desirable to use these results to estimate global NO_x production by TLEs and to estimate stratospheric ozone depletion by these discharges. A similar study by Rozanov et al (2005) found that NO_y produced by energetic electron precipitation reduced ozone levels by 5% at mid-latitudes, increasing to a 30% reduction near the south pole. This had the additional effect of reducing stratospheric and tropospheric temperatures, pointing to wide ranging effects if TLEs do prove to be a significance source for middle atmosphere NO_x.

In the future, it is planned to expand these preliminary results to include discharges at lower pressures, pressures between 100-500 mb, and discharges at higher overall currents, but with lower current densities, which more would more accurately simulate TLE discharge conditions.

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