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NO_x Production From Lightning

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A Proposal to the LLNL LDRD Labwide Program

NO_x Production From Lightning

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ABSTRACT

This proposal requests funds to construct a theoretical model for the production of NO_x from lightning. NO_x production can cause changes in the atmospheric ozone distribution which are biologically harmful. Lightning will also result in the production and/or destruction of other gases which can be used as observational tracers of the lightning process. These tracers can be used to provide an observational calibration of the production mechanism. A new and very interesting aspect of this work is to provide modeling support for the conjecture that the lightning process will destroy CFC compounds in the disturbed air for long periods of time and this would provide a very good observational tracer. The ultimate product of this effort would be an accurate prediction of the amount of NO_x produced per unit energy of the lightning. Use will be made of LLNL developed 1-D radiation-hydrodynamic models to predict the temporal and spatial behavior of the temperature and density in the air in the vicinity of the lightning channel as a function of altitude. A chemical kinetics model will be used to predict the time variation of the trace gas species in the disturbed air parcels as a function of the temperature and density profiles calculated by the hydrodynamics model.

INTRODUCTION

A current and major concern in the field of atmospheric science is the behavior of the ozone distribution in the earth's atmosphere as well as the chemical processes which control the production and depletion of ozone. One of the major factors in the determination of the ozone distribution is the abundance of atmospheric NO_x (NO , NO_2) (Kroening and Ney, 1962; Crutzen, 1970; Johnston, 1971). In the troposphere, increased NO_x emissions can cause increased ozone abundances (Chameides and Walker, 1973; Crutzen, 1970) while in the stratosphere increased NO_x emissions can cause decreased ozone abundances (Crutzen, 1970; Johnston, 1971). Thus, increased NO_x emissions, in general, result in harmful effects to the ozone distribution. NO_x emissions have both natural and anthropogenic sources. The major anthropogenic NO_x sources are fossil fuel emissions, biomass burning, and jet aircraft emissions (Penner et al., 1994). The primary natural NO_x sources (Penner et al., 1994) are soil emissions (5 Tg/yr, range 1 - 20) and lightning (10 Tg/yr, range 2 - 100). Lawrence et al. (1995, "LR") calculates NO_x production from lightning at 2 Tg/yr with a range 1 - 8 Tg/yr. This uncertainty range is enough to have significant effects on the atmospheric NO_x budget as well as the atmospheric ozone distribution (LR). NO_x production by lightning can be calculated as the product of three terms, the NO_x molecules produced per Joule of lightning energy, the energy per lightning flash, and the number of lightning flashes per unit time on a global basis (LR). LR indicates that the NO_x production per Joule of lightning energy lies in the range $\sim 1.2 - 26 \times 10^{16}$ molecules/J, taking into account both laboratory and theoretical estimates. The most likely estimates however, are in the range of $\sim 5 - 17 \times 10^{16}$ molecules/J (Chameides, 1979, "CM").

The modeling of the production of NO_x from lightning is a difficult problem. First, the physics of lightning has to be characterized in order to determine the amount of energy available in the lightning channels. Second, the spatial and temporal heating of the channel as well as the air surrounding the channel has to be determined. Third, the abundance of NO_x and other significant trace species formed as a result of the lightning must be calculated. Fourth, long lived trace species, or the absence of measurable trace species, need to be determined for observational calibration of the models. Recent work by Price et al. (1995) at LLNL has provided models for the energy available in the lightning channels. Prior work by Lin (1954), CM, Goldenbaum and Dickerson (1993) as well as Chameides et al. (1977) have used hydrodynamic models combined with limited chemistry models to characterize NO_x production by lightning discharges. Chang and Grossman (1996) have obtained an analytic solution to the approximate "freezing theory" model for the NO_x production by lightning in the square wave, strong shock approximation given by Zel'dovich and Razier (1967) and CM. Bhetanabhotla et al. (1985) have used a detailed chemistry model to calculate the temporal behavior of 10 important trace species formed in the heated core channel. An important result of this work, in addition to the NO_x production, was that the N_2O concentration in the heated channel was significantly reduced over that in ambient air for periods on the order of 100 seconds after the lightning strike. They indicate however that N_2O measurements in the air around thunderstorms actually shows an increase in N_2O concentration that is probably due to processes taking place in the air immediately outside of the heated channel.

The major goals of this proposal are to, 1) use detailed LLNL developed radiation-hydrodynamic computer codes to model the spatial and temporal temperature and density distribution of the heated air around a lightning channel, 2) combine this calculation with a LLNL developed detailed high temperature chemistry model which predicts the time variation of observationally significant trace species and NO_x . A particular aspect of this work which would be tested is the conjecture that CFC concentrations (chloro-fluoro-carbons, CCl_3F for example) would be significantly decreased in air in the vicinity of the lightning activity and this might be a very long lived tracer of NO_x production by lightning. If this is the case then CFC sampling of the air in the vicinity of thunderstorms correlated with NO_x sampling in the same air mass would provide the calibration of the theoretical models which fix and quantify NO_x production per unit energy.

The successful completion of the work described in this proposal should position LLNL for future funding of the climatic consequences of these studies from DOE and/or NASA.

SCIENTIFIC AND TECHNICAL APPROACH

I. ENERGETICS OF LIGHTNING CHANNEL

The energy in a lightning flash (stroke) can be determined in two ways:

$$E_1 = \int \frac{I(t)^2}{\sigma(t)\pi r(t)^2} dt \quad [\text{Joules/meter}]$$

$$E_2 = V \int I(t) dt = VQ \quad [\text{Joules}]$$

where $I(t)$ is the current flowing through the channel, $\sigma(t)$ is the electrical conductivity of the lightning channel, $r(t)$ is the radius of the channel and V is the breakdown potential. The conductivity is strongly dependent on the lightning channel temperature. Peak temperatures between 30000-40 000 K are reached within a few microseconds (Orville, 1968). The electrical conductivity varies by 11 orders of magnitude as the temperature rises from 2000 K to 30000 K (Yos, 1963; Uman and Voshall, 1968). Furthermore, the channel radius varies by two orders of magnitude, from 1mm to 10cm, during the lifetime of the flash. Both these parameters are highly uncertain, resulting in large uncertainties in E_1 . In addition, E_1 has units of Joules/meter. This implies an additional assumption has to be made regarding the channel length before calculating the total energy.

Using the second formulation for energy (E_2), one only has to assume a value for the breakdown potential (V) which does not vary over the lifetime of a lightning flash, and is better known than $\sigma(t)$ and $r(t)$. The breakdown potential can vary from storm to storm, but is fairly constant during a particular storm. As clouds become increasingly electrified during a storm's development, breakdown simply occurs more often, resulting in larger lightning frequencies. However, the value of the breakdown potential remains

fairly constant throughout the storm. In addition, using E_2 also provides one with the total energy (Joules), without making assumptions for channel length. For the above reasons we have decided to use E_2 for the calculation of the energies in lightning flashes.

Price et al. (1995) have derived a formulation for the charge deposited by a lightning flash, which is dependent on the peak current in the discharge (I_0):

$$Q = 5.9 \times 10^{-4} I_0$$

For the typical breakdown potential ($2.5 - 3.5 \times 10^8$ V), a mean value of 3×10^8 V is used, giving an energy of:

$$E = QV = 1.77 \times 10^5 I_0$$

For mean negative peak currents ranging from 10 - 60 kA (90% of observed values), the above implies a flash energy of $1.8 \times 10^9 - 1.1 \times 10^{10}$ J, in good agreement with previously published values (Uman, 1987). For lightning channel lengths on the order of 5 km, this energy range gives energy depositions on the order of $3.6 \times 10^5 - 2.2 \times 10^6$ J/m.

II. HYDRODYNAMIC EXPANSION AND HEATING OF THE AIR

Based on an initial condition for the energy input to the lightning channel at a given altitude, a radiation - hydrodynamics model will be used to predict the temperature-density profile of the air as a function of time and distance from the center of the channel. Various hydrodynamic codes are available at LLNL that can model the behavior of the shock heated air in and around the channel. In particular, a treatment of the radiation field will be included in order to determine the temperature variation of the air mixture. Attempts will be made to calculate the proper partitioning of the energy input to the channel into radiation, thermal, kinetic, and chemical bins. In this manner a temperature profile of the disturbed air down stream of the shock-wave formed in the lightning channel can be determined. The chemical dissociation taking place in these air parcels can play an important role in the final abundances of trace gas species.

III. CHEMICAL KINETICS MODEL

A chemical kinetics framework which predicts the time variation of the concentration of gas species as a function of temperature has been developed at LLNL. To this framework must be added data giving the appropriate rate constants of the relevant chemical reactions, tabulated in the JANAF (1970) database, for the disturbed air in and around the lightning channel as a function of temperature. The temperature profiles are determined by the hydrodynamic solutions. In particular, a model for CFC chemistry will be added to the kinetics framework in order to determine the effectiveness of the absence CFC's as a long lived tracer of lightning activity. This coupled with the prediction of NO_x generation will provide the basis for atmospheric measurements which, in turn, will calibrate the theoretical model.

OUTLINE OF NO_x PRODUCTION FROM LIGHTNING PROJECT

YEAR 1.

- Assemble data giving range of energy/length factors for different types of lightning.
- Adapt LLNL high temperature chemistry code to calculate appropriate trace gas species,
  1. Bench mark calculations and comparison of results with prior published work.
  2. Add CFC species to chemical model.

- Adapt LLNL radiation - hydrodynamics model to calculate air temperatures and densities of air in and around heated lightning channel.

1. Comparison of results with prior published results.

YEAR 2.

- Lightning channel hydrodynamics and temperature calculations at different altitudes in the ambient atmosphere.
- Chemistry calculations for NO_x , CFC, N_2O abundances at different altitudes using spatial and temporal density and temperature profiles obtained from hydrodynamic models.
- Write paper on results of theoretical models.

YEAR 3.

- Collaboration with field programs to obtain measurement data that can calibrate theoretical models.
- Combine observational data with model runs to fix NO_x production as a function of energy input.

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