Executive Summary

A secondary goal of the HEAT project is to examine the effects of Houston thunderstorms on atmospheric chemistry through lightning-produced NO_x ($NO_x = NO + NO_2$) and transport and processing of pollutant species. Convective clouds transport, scavenge, and mix species between the atmospheric boundary layer and upper troposphere. They are also an important and poorly understood source of NO_x through lightning. Due to this processing they affect the distribution and deposition of air pollutants. Through precursor transport and NO_x production, they impact upper tropospheric ozone and HO_x concentrations, influencing global radiative forcing and the cleansing capacity of the troposphere. The HEAT project provides an ideal venue to investigate the mechanisms and effects of thunderstorms on urban air pollutant fate and upper tropospheric chemistry. Investigations will include determination of possible urban-enhanced lightning production of NO_x, study of thunderstorm dynamical and microphysical processes affecting NO_x and urban pollutant fate, and elucidation of the impacts of convective venting of pollutant species and NO_x on upper tropospheric photochemistry.

Part of Section 1.1 GOALS section of SOD

Effects of Urban Influenced Thunderstorms on Atmospheric Chemistry

Thunderstorms have important impacts on air pollutant transport and dispersal through complex air motions (updrafts, downdrafts, entrainment, detrainment), scavenging of pollutants by hydrometeors, and gas and aqueous phase chemistry (Hales and Dana, 1979; Dickerson et al, 1987; Jaegle et al, 1997; Barth et al., 2001; Pickering et al., 2001). In addition, they are important sources of NO_x through lightning (e.g., Liaw et al, 1990). In polluted areas, increased lightning may further enhance this NO_x production. By venting surface-emitted pollutants and lightning-produced NO_x to the upper troposphere, thunderstorms impact upper tropospheric abundances of ozone (Pickering et al., 1990) and HO_x (Jaegle et al., 1997). Hence, they affect global radiative forcing (Park et al., 2001) and the cleansing capacity of the troposphere (Thompson, 1992). In polluted areas, these effects may be enhanced. In regions of increased aerosol concentrations, Andreae et al (2004) found that smoke aerosols suppressed drop coalescence and wet scavenging, allowing more smoke particles to detrain from clouds in the upper troposphere and even the lower stratosphere. Finally, at local to regional scales, thunderstorm dispersal and scavenging of air pollutants can impact deposition of acids. The HEAT project provides a unique opportunity to study the impacts of pollution-influenced thunderstorms on transport and fate of volatile and particulate contaminants (including NO_x from lightning), as the Houston area experiences extreme air pollution and lightning enhancement. Many previous studies of thunderstorm effects on tropospheric composition have occurred in minimally polluted or pristine regions (e.g., PRESTORM, STERAO, ABLE-2B, PEM Tropics-B). The HEAT project provides the opportunity to measure freshly-emitted volatile organic compounds, species that play an important role in ozone formation within the Houston environment and in thunderstorm outflow regions. High concentrations of NO_x, volatile organic compounds (VOCs) and aerosols in this polluted region were documented through airborne and surface measurements in the Texas Air Quality Study – 2000 (e.g., Ryerson et al., 2003; Karl et al., 2003; Brock et al., 2003).

Through the HEAT project, we will improve our knowledge of NO_x production by lightning, the mechanisms through which storms of various types impact air pollutant and NO_x redistribution, and the impact of urban influenced thunderstorms on upper tropospheric photochemistry. This will be achieved through aircraft observations of chemical concentration of species of interest (CO, O_3 , NO, NO_2 SO_2,, hydrocarbons, peroxides, formaldehyde, and aerosols) in distinct storm regions (inflow, downdraft, and outflow) for a variety of storm types. Concentrations of nitrates and sulfates in precipitation at surface sites will also be measured.

Comparison of chemical concentration with lightning data from the lightning mapping system (LDAR II) and NLDN will explain lightning production of NO_x. Mass flux balance calculations and chemical transport and reaction modeling will also be employed in the analyses.

3.5 Effects of Urban Influenced Thunderstorms on Atmospheric Chemistry

<u>OBJECTIVE A1</u>: NO_x production by lightning

PROCESS TO BE STUDIED: The availability of nitrogen controls many aspects of biogeochemistry, as it is a key nutrient for plant growth [Schlesinger 1991]. In addition, nitrogen oxides are integral in tropospheric photochemistry [Crutzen 1973]. Lightning is an important source of bioavailable nitrogen as NO_x [Liaw et al. 1990]. However, production of NO_x by lightning remains a significant uncertainty in the global nitrogen budget [Lee et al. 1997; Price et al. 1997]. Contributions to this global budget uncertainty include uncertainties in the global flash rate, the representation of average thunderstorm lightning production, the average production of NO_x per lightning flash, and the relative contributions of cloud-to-ground versus intra-cloud lightning to NO_x production (DeCaria et al., 2000).

GOALS OF STUDY: The main goal of this objective is to quantify lightning production of NO_x. Subgoals include estimation of the amount of NO_x produced per meter of flash, per flash, and per thunderstorm (to estimate global lightning-produced NO_x), intra-cloud (IC) versus cloud-to-ground (CG) production of NO_x, NO_x production rates for the different components of a flash, and the NO_x production for different storm types (isolated, squall line, supercell).

MEASUREMENT REQUIREMENTS: To achieve the goals of this objective, measurements of lightning flashes, NO and NO_2 mixing ratios in and around storms, and nitrate concentrations in precipitation are needed. Thunderstorm penetrations (particularly at anvil levels) to measure chemical concentrations will be conducted by the University of North Dakota Cessna Citation or the Weather Modification, Inc. (WMI) Lear jet. A lower-level aircraft (either the University of Wyoming King Air or a Baylor University aircraft) will sample near the base of the storm. They will observe clouds with and without lightning, and at different stages of development. The radars (WSR-88D, NCAR S-Pol, and CSU CHILL) will be used to determine storm structure and to direct aircraft to certain storm regions (inflow, downdraft, and anvil). Nitrate concentration in precipitation will be measured from collection receptacles at mesonet sites. The lightning mapping system (LDAR II) will reveal regions of lightning activity and will be used to determine flash lengths. Flash type (IG versus CG) will be determined using data from the lightning mapping system and National Lightning Detection Network (NLDN). Determination of NO production per meter flash, per flash, and per storm will be determined by comparison of lightning data with aircraft data, chemical transport modeling, and mass flux balance analysis.

<u>OBJECTIVE A2</u>: Transport and fate of pollutants in thunderstorms

PROCESS TO BE STUDIED: Thunderstorms vent gaseous and particulate air pollutants to the upper troposphere leading to enhanced pollutant atmospheric lifetimes and impacts on upper tropospheric chemistry and global radiative forcing. Through entrainment and detrainment of air at mid-levels, thunderstorms also impact regional dispersal of air pollutants. Scavenging of soluble and low volatility species leads to ground deposition of pollutants (acid rain) with resulting negative impacts on ground ecosystems. Finally, gaseous- and aqueous-phase chemical reactions in thunderstorms can transform trace species to forms with different properties (e.g.

volatility, solubility) and hence impact pollutant fate. Transport of trace pollutants, including VOCs, oxygenated VOCs, NO_x, and black carbon (soot), to the upper troposphere will impact photochemical production of ozone (e.g. VOCs and NO_x) or directly affect absorption of radiation (e.g. soot and ozone). Climatically-important radiative effects on the atmosphere may result (Jacobson, 2001; Park et al., 2001). The HEAT project provides an ideal environment to improve our understanding of how thunderstorms affect urban pollutant transport and fate.

GOALS OF STUDY: The goals of this objective are to understand the mechanisms by which trace species (e.g. volatile and particulate air pollutants and NO_x) are redistributed by thunderstorms and to quantify their fate during and after different storm types. This will include determining the relative importance of thunderstorm processes (convective motions, entrainment, scavenging by liquid and ice phase hydrometeors, and gas and aqueous phase chemistry) to the fate of specific air pollutant species of interest (e.g. NO_x, hydrocarbons, and aerosols) and categories of species with similar characteristics (e.g. solubility).

MEASUREMENT REQUIREMENTS: To achieve these goals, measurements of chemical vertical concentration profiles before, during, and after several storms of different types are needed. In addition, chemical concentrations in storm inflow regions, in outflow regions, and in regions containing liquid and solid hydrometeors are needed. Species of interest include:
Priority 1 - CO, O_3, NO, NO_2 ; Priority 2 – (in ranked order) peroxides and formaldehyde, continuous hydrocarbon measurements; grab sampling for laboratory hydrocarbon, nitrate, and CFC analysis; soot; and SO_2 . Species of interest in precipitation include nitrate and sulfate. The University of Wyoming's King Air or the Baylor aircraft will conduct survey flight patterns at 3 or 4 altitudes in the expected inflow region before the thunderstorm activity to characterize the variability of trace species of interest. Also as the convection is developing the jet aircraft should perform spiral profiles from the boundary layer to the upper troposphere. During thunderstorm activity, the King Air or Baylor aircraft will intensively profile the inflow region, and the UND Citation or the Weather Modification, Inc. (WMI) Lear jet will conduct flight legs in the anvil. The anvil work can be comprised of transects across the anvil at various downwind distances, flight tracks along the anvil axis, or upward spirals through the anvil. If possible, chemical concentration in the low-level storm outflow will also be measured by instrumentation on the King Air or Baylor aircraft. Concentrations in precipitation will be determined at mesonet sites. The radars (WSR-88D, NCAR S-Pol, and CSU CHILL) and the LDAR will be used to direct the aircraft to specific regions of the storm. CO will be used as the primary tracer of air motion through clouds. In conjunction with chemical transport and reaction modeling and mass flux balance analysis, these measurements will elucidate the mechanisms of redistribution of air pollutants and NO_x as well as their post storm fate for a variety of storm types.

<u>OBJECTIVE A3:</u> Effect of urban thunderstorms on upper tropospheric chemistry

PROCESS TO BE STUDIED: Studies have found that convection may have significant impacts on ozone and odd hydrogen chemistry of the free troposphere through venting of boundary layer air (Pickering et al, 1990, 1992; Jaegle et al 1997). NO_x produced by lightning may have additional impacts, due to the role of NO_x in ozone chemistry. Because of the close proximity of thunderstorms observed during the HEAT project to recent emissions, the contribution of freshly-emitted hydrocarbons and their oxidative products on upper troposphere ozone photochemistry can be assessed.

GOALS OF STUDY: The goals of this objective are to explain the effects of urban air pollution on upper tropospheric ozone chemistry through storm venting of surface emitted pollutants and enhanced production of NO_x (due to urban enhanced lightning). A sub goal is to quantify the relative contributions of lightning produced NO_x and surface emitted NO_x to upper tropospheric NO_x concentrations for a variety of storm types.

MEASUREMENT REQUIREMENTS: To achieve these goals, quantification of air pollutant venting and lightning-produced NO_x to the upper troposphere and quantification of the contributions of all NO_x sources are needed. Measurements of ozone in storm outflow air are needed in the anvil during the active thunderstorm and during transport of the outflow downstream. The Citation or LearJet will likely need to make two flights on some days to achieve this objective. The first flight would be to probe the active storm anvil and the second flight would locate and sample the upper tropospheric outflow plume at a distance of several hours downwind of the convective cores. Species to measure are the same as those listed in Objective A2. Chemical transport and reaction modeling on the regional scale will be instrumental to this objective.

MEASUREMENT ISSUES

Chemical measurements have been prioritized under Objective A2 above. However, all of the aircraft that are proposed are relatively small and will allow only limited chemistry payloads in addition to the cloud microphysics instrumentation. Decisions will need to be made concerning the extent and complexity of the chemistry payload relative to the other measurements. In order to effectively use the chemical data in analysis and modeling, instrument intercomparison flights must be conducted by the two aircraft during the field project.

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