Scientific Program Overview (SPO)

for a

Deep Convective Clouds and Chemistry (DC3)

Field Campaign

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A. Cover Page

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B. Project Summary

The Deep Convective Clouds and Chemistry Project (DC3) field campaign investigates the impact of deep, midlatitude continental convective clouds, including their dynamical, physical, and lightning processes, on upper tropospheric (UT) composition and chemistry. The DC3 field campaign makes use of extensively instrumented aircraft platforms and ground-based observations. The NSF/NCAR Gulfstream-V (GV) aircraft is the primary platform to study the high altitude outflow of the storms, and is instrumented to measure a variety of gas-phase species, radiation, and cloud particle characteristics. The GV is also documenting the downwind chemical evolution of the convective plume The NASA DC-8 aircraft complements the GV via in situ observations to characterize the convective storm inflow and provides remote sensing to aid in flight planning and column characterization. Ground-based radar networks are used to depict the physical and kinematic characteristics of the storm and provide input to the aircraft operations. The impact of lightning on outflow composition is constrained through detailed measurements from lightning mapping arrays. The forecasting and analysis is improved through other observations such as radiosondes and precipitation collection and its chemical analysis. Satellite data is used to place the airborne and ground-based measurements in the context of the wider geographical region and help guide sampling strategies. At the same time, DC3 measurements help satellite retrievals of atmospheric constituents such as NO_2 near storms.

The observations are conducted in three locations: 1) northeastern Colorado, 2) central Oklahoma, and 3) northern Alabama in order to gather data on different types of storms and with different boundary layer compositions as well as to ensure sampling of convection during the time period of the field campaign. The types of storms being sampled are air mass, multicell, and supercell convection.

The DC3 project addresses the following goals, 1) Quantify and characterize the convective transport of fresh emissions and water to the upper troposphere within the first few hours of active convection, investigating storm dynamics and physics, lightning and its production of nitrogen oxides, cloud hydrometeor effects on wet deposition of species, surface emission variability, and chemistry in the anvil. 2) Quantify the changes in chemistry and composition in the upper troposphere after active convection, focusing on 12-48 hours after convection and the seasonal transition of the chemical composition of the UT. The DC3 field experiment will improve current knowledge of convection and chemistry by providing a comprehensive suite of chemical measurements within the context of excellent kinematic, microphysical and electrical ground-based measurements. These measurements will provide the necessary information to estimate ozone sources and sinks in the upper troposphere where ozone is radiatively active as a greenhouse gas.

The DC3 project provides broader impacts to society via extensive education and outreach activities, and via improved understanding of sources of UT ozone, an important constituent to climate and air quality, for assessment reports and resulting policy implications. Further, DC3 measurements are instrumental in improving model parameterizations of convective transport, production of NO by lightning, and wet deposition of chemical species. Students from both undergraduate and graduate school are participating in DC3 in a variety of ways including airborne and ground-based observations, design and construction of instruments, operation and improvement of numerical models, precipitation collection and analysis, and reporting of the results to the scientific community through presentations and publications. Outreach days for the public and media provide a valuable means to engage the public in atmospheric science.

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D. Project Description

D.1 Overall scientific rationale & goals of the project

The upper troposphere and lower stratosphere (UTLS) is an important region for Earth's climate because water vapor, ozone, cirrus clouds and aerosols in this region strongly contribute to radiative forcing of the climate system. The UT and LS have very different chemical compositions resulting in strong gradients across the tropopause. Further, the UTLS is a highly dynamic region influenced by a broad range of scales, from deep convection and gravity waves, to tropospheric weather systems and the stratospheric large-scale circulation. Convective transport is a major pathway for rapidly moving chemical constituents and water from the boundary layer to the upper troposphere and in some cases to the lower stratosphere. Yet the impact of convective transport on the UTLS composition and chemistry has not been fully characterized on either the global or continental scale. The Deep Convective Clouds and Chemistry (DC3) field experiment is proposed to study the impact of continental, midlatitude deep convection on the UTLS composition and chemistry above the continental U.S. during the lifetime of the storm itself and during the period 12-48 hours after active convection. The primary goals of DC3 are to:

1) *quantify and characterize the convective transport of fresh emissions and water to the upper troposphere within the first few hours of active convection, investigating storm dynamics and physics, lightning and its production of nitrogen oxides, cloud hydrometeors effects on scavenging of species, surface emission variability, and chemistry in the anvil.*
2) *quantify the changes in chemistry and composition in the upper troposphere after active convection, focusing on 12-48 hours after convection and the seasonal transition of the chemical composition of the UT.*

These goals will be addressed by sampling convection during May and June in three U.S. locations, northeast Colorado, central Oklahoma, and northern Alabama. These locations give us the opportunity to contrast the effects, and the storm processes influencing those effects, on UT composition for regions of remote continental air versus those with air masses more influenced by anthropogenic emissions and for the much different storm kinematics and microphysics of regions with abundant boundary layer moisture versus those with drier environments. While previous field experiments have either focused on storm kinematics, microphysics, lightning, and their influence on convective transport and nitric oxide production or on chemistry downwind of convection, no previous field experiment has gathered the detailed storm and suite of chemistry data needed to fully characterize the influence of thunderstorms on UTLS composition and chemistry.

D.2 Current State of Knowledge

Convection is common and frequent throughout much of the world, over land and over oceans, in the tropics and midlatitudes. Some characteristics of midlatitude, continental convection are similar to tropical convection but differences occur in surface forcing and vertical extent. Because the tropical tropopause is ~3 km higher than the midlatitude tropopause, tropical storms tend to have greater vertical extent. Larger surface forcing and greater low level buoyancy over land result in continental convection having significantly stronger updrafts than oceanic storms. Consequently mixed phase processes in continental convection are robust and lead to heavy convective rainfall associated with the fallout and melting of graupel and hail. Continental convection produces more lightning than oceanic convection (Christian et al. 2003), and as a result makes a major contribution to the quantity of nitrogen oxides in the UT. Lastly the boundary layer composition differs significantly for continental and oceanic storms. The oceanic boundary layers have less of an influence from anthropogenic and terrestrial biogenic sources, but contain more halogens.

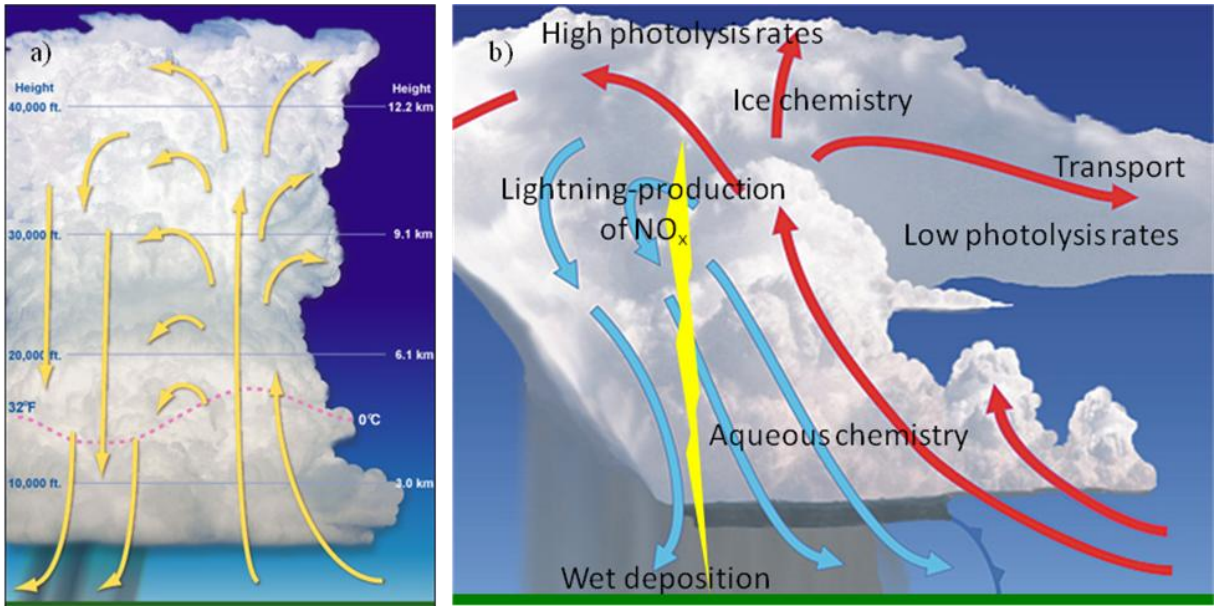

Figure 1. Schematic of a) a mature airmass thunderstorm and b) a squall line vertical cross section. Taken from the NOAA National Weather Service http://www.srh.weather.gov/srh/jetstream/index.htm. Superimposed on panel b are processes affecting chemical species that are ingested into storms.

Convective storms over the central U.S. are often an everyday occurrence during the late spring and summer (Carbone et al., 2002). These storms range from airmass thunderstorms, to multicellular thunderstorms and supercells, to mesoscale convective systems depending on the instability, wind shear and mesoscale forcing of the atmosphere (see Figure 1 for schematics of airflow in airmass and squall line storms). These storms can have a significant impact on UT composition as seen from observations taken during the July and August 2004 NASA INTEX-A project. Results from this project showed that much of the UT region over the U.S. is influenced by convection (Cooper et al., 2006; Bertram et al., 2007; Hudman et al., 2007; Snow et al., 2007; Fried et al., 2008).

There have been many experimental and theoretical studies exploring the influence of deep convection on the chemical composition of the upper troposphere (e.g. Chatfield and Crutzen, 1984; Dickerson et al., 1987; Pickering et al., 1990; Jaeglé et al., 1997; Ridley et al., 2004a,b; Huntrieser et al., 2002; Ancellet et al., 2008). The PRESTORM campaign in 1985 provided some of the first observations clearly showing convective redistribution of ozone (O_3), nitric oxide (NO), carbon monoxide (CO), and non-methane hydrocarbons (NMHC) in a mid-latitude storm (Dickerson et al., 1987). Subsequent modeling and analysis (Pickering et al., 1990) found that convective redistribution could dramatically increase ozone production in the upper troposphere by as much as a factor of four with peak net ozone production rates of 15 ppbv per day. This increase was shown to rely on the transport of both NO and NMHC from the boundary layer as well as NO from lightning. Thus, transport, production of NO from lightning, and scavenging of soluble species (Figure 1b) were found to be important processes occurring in thunderstorms affecting the chemical composition of the troposphere.

Deep convection is also important for hydrogen oxides HO_x (sum of hydroxyl and hydroperoxy radicals: $HO_x = OH + HO_2$) and HO_x precursors, such as hydrogen peroxide (H_2O_2), methyl hydroperoxide (CH_3OOH) and formaldehyde (CH_2O). The influence of convective transport on HO_x precursors was first hypothesized by Chatfield and Crutzen (1984), but renewed attention came when the first upper tropospheric HO_x observations over the tropical Pacific Ocean during the STRAT (1996) campaign sometimes exceeded theoretical expectations by a factor of two or more (Jaeglé et al., 1997). It was proposed that convectively transported peroxides and formaldehyde could provide the additional source of HO_x needed to explain observations. Unfortunately, no measurements of peroxides or formaldehyde were available to

confirm the hypothesis. Crawford et al. (1999) subsequently examined upper tropospheric chemistry measurements (which lacked HO_x but included peroxides) by the NASA DC-8 over the tropical Pacific during the PEM West A, PEM West B and PEM Tropics A campaigns. While model results did support the possibility of a source of HO_x from convectively transported peroxides, the PEM peroxide observations fell far short of the levels needed to explain the discrepancy in the STRAT observations.

Diagnosing convective transport of formaldehyde and peroxides is complicated by their solubility and assumptions regarding the role of ice in scavenging processes (Barth et al., 2001; 2007a). Furthermore, observations are needed as close as possible to the time and place of convection since these species are rather short-lived, photochemically-active trace gases that can immediately undergo rapid change in the UT convective outflow where high photolysis rates can accelerate photochemistry in the bright regions of the storm and anvil (Figure 1b). Conversely, low photolysis rates at the bottom of the anvil can slow the photochemistry. Recently, a cloud resolving model analysis of observations from STERAO (1996) have included the effects of convected peroxides and formaldehyde on ozone production (DeCaria et al., 2005). Large enhancements in H_2O_2 and CH_3OOH were predicted in cloud outflow at 10 km, however in-cloud observations of these species were only available at 6.7 km where no significant enhancement was predicted. This emphasizes the strong need for peroxide and formaldehyde observations at the appropriate altitude and timing needed to evaluate the full chemical impact of outflow from convective systems.

The main strength of these previous studies is the clear demonstration that ozone photochemistry is perturbed by convective outflow. The most important advances needed to improve current knowledge call for field studies that extend the altitude range of observations, provide a comprehensive suite of chemical measurements, and sample in close proximity to convection where storm kinematic, microphysical, and electrical characteristics can be documented. These three capabilities have existed separately but have never been exploited simultaneously in a focused experiment. In addition, measurements are needed one to two days transport time downwind to aid in understanding the photochemical evolution of the outflow.

D.3 Hypotheses to be tested

DC3 seeks to quantify and characterize convective transport, composition and chemical transformation within the first few hours of active convection, and secondly to quantify the changes in chemistry and composition in the upper troposphere after active convection, focusing on 12-48 hours after convection and the seasonal transition of the chemical composition of the UT. There are several hypotheses that can be tested during the DC3 campaign. We have prioritized these into eight hypotheses that we think are critical to address.

D.3.a) Goal 1: Quantify and characterize the convective transport of fresh emissions and water to the upper troposphere within the first few hours of active convection

- **Convective transport of air, water and chemical constituents**

Because transporting air, water, and chemical constituents from the boundary layer to the upper troposphere is the primary process associated with deep convection, quantifying the amount of transport and the altitudes to and from which the constituents are being transported are required to assess the importance of other processes such as scavenging and chemistry. Further, understanding the convective transport is important both for improvement of convective parameterizations of air, water, heat, and chemical constituents, as well as proper measurement and simulation of latent heating depths, which are crucial for momentum and radiative budgets (e.g. Schumacher et al., 2004; Alexander et al., 2004).

DC3 will use two aircraft platforms that can concomitantly sample the inflow and outflow regions of isolated convection while the storm is remotely measured from the ground by polarimetric Doppler radars and a lightning mapping network. This experimental setup will allow for analysis of the storm kinematics by both in situ and dual-Doppler radar measurements and for trace gas flux estimates using in situ aircraft

measurements. Using this same experimental setup in three locations gives us the opportunity to contrast mass fluxes of air, water, and chemical species and their level of outflow for storms ranging from airmass thunderstorms to isolated severe convection and squall lines.

Hypothesis 1) Inert tracers are transported primarily to the upper troposphere within 3-5 km of the tropopause in shear-driven storms, such as those found in Colorado and Oklahoma, and can be used to determine the maximum outflow altitude, which will be different than cloud top height, the level of neutral buoyancy, and the maximum ice content altitude. These same inert tracers are transported throughout the free troposphere in airmass thunderstorms, more common in the southeastern U.S. This implies that shear-driven thunderstorms contribute more to UTLS chemistry, ozone production, and cross tropopause transport than airmass thunderstorms.

Results from the PRESTORM campaign showed high CO mixing ratios within ~4 km of the tropopause indicating transport of boundary layer air, and low CO mixing ratios transported from the unperturbed upper troposphere to near cloud base (Dickerson et al., 1987; Luke et al, 1992). Other subsequent studies (e.g., Pickering et al., 1992; Scala et al., 1990; Thompson et al., 1994; Hauf et al., 1995; Skamarock et al., 2000) have corroborated this type of transport pattern for mid-latitude deep convection. However, these studies have also indicated that transport of boundary layer air to the anvil depends strongly on storm dynamics and, in particular, the degree of entrainment that occurs in any one storm.

Because airmass storms grow, mature, and dissipate during a short time period (1 hour or less), the altitudes of convective outflow in airmass storms should vary with time. During the growth stage, the convective outflow is occurring at cloud top in the mid- to upper troposphere (Kingsmill and Wakimoto, 1992), while at later stages the convective outflow is in the UT. During the dissipation stage of the storm, a downdraft is formed bringing air from the upper regions of the storm to mid and low levels. With an ensemble of airmass thunderstorms occurring in a region, the net effect of transport of inert species is to move boundary layer air to altitudes throughout the free troposphere.

The level of neutral buoyancy (LNB) and cloud-top heights are generally used to estimate the convective updraft depths and air mass convective outflow profiles from operational datasets. However, because entrainment is highly variable and depends on the local environmental profile, storm classification and storm size (Cohen, 2000; Mullendore et al., 2005), the LNB does not satisfactorily predict the level of maximum outflow (Sherwood et al., 2004). In prior field campaigns, convective outflow profiles have been constructed from aircraft in-situ data or from dual-Doppler derived winds, but never have both observations been available concurrently. The DC3 proposed measurement platforms will provide detailed convective outflow profiles that are constrained by both observed storm motions (dual-Doppler derived winds and in-situ velocity measurements) and also in-situ chemical tracer measurements.

- **Cloud hydrometeor effects on wet scavenging of species,**

Less is known about the transport efficiency of HO_x precursors and other species that are soluble and reactive within convection (such as, CH_2O, H_2O_2, and HNO_3) because of the lack of concomitant measurements of these species in deep convective inflow and outflow regions. Recent cloud-resolving model simulations (Pickering et al., 2001; DeCaria et al., 2005; Barth et al., 2007a) have given significantly different results. These are exemplified by a model intercomparison study (Barth et al., 2007b) that showed a large variation in predictions of CH_2O, H_2O_2 and HNO_3 (Figure 2). Much of the variability seen in Figure 2 results from the assumptions made regarding ice, the riming of cloud water, and the retention of a soluble species during cloud drop freezing. Further, re-emission of soluble species during ice sublimation back into the gas phase is a major uncertainty in our understanding of cloud scavenging. Measurements in convective outflow regions (Pickering et al., 1996; Ravetta et al., 2001; Fried et al., 2008) report enhancements of H_2O_2 and CH_2O compared to the background UT mixing ratios. The discrepancies between the different model simulations and between model simulations and observations emphasize the

need to make measurements of HO_x precursors in convective outflow for a variety of conditions (e.g. outflow altitude, boundary layer composition, and storm type) so that their impact on upper tropospheric ozone can be ascertained.

Hypothesis 2) In the anvil and near the convective cores, soluble species, e.g. HNO_3, H_2O_2 and CH_2O, will be depleted compared to their background UT mixing ratios because ice scavenges the dissolved species in cloud water within the convective core. Furthermore, because of the short time an air parcel is in contact with liquid water and the high updraft speeds, transport of soluble species to the UT will be more efficient in the high plains (Colorado) storms compared to the storms in northern Alabama. The warmer cloud bases and greater moisture contents in Oklahoma and Alabama have larger liquid water regions resulting in more efficient scavenging of soluble species.

Barth et al., (2001) estimated the time an air parcel was in contact with the liquid water region for the simulated 10 July 1996 STERAO storm to be less than 10 minutes. This short time period is related to the high cloud base of the storm (3.2 km m.s.l.) and the high updraft speeds (> 25 m s^{-1}). We expect a different result in both Oklahoma and Alabama storms because of the lower cloud base heights. By sampling soluble species (HNO_3, H_2O_2 and CH_2O) in both the inflow and outflow regions of storms in all three regions, we will get the needed information to determine the importance of scavenging including the role of the ice phase.

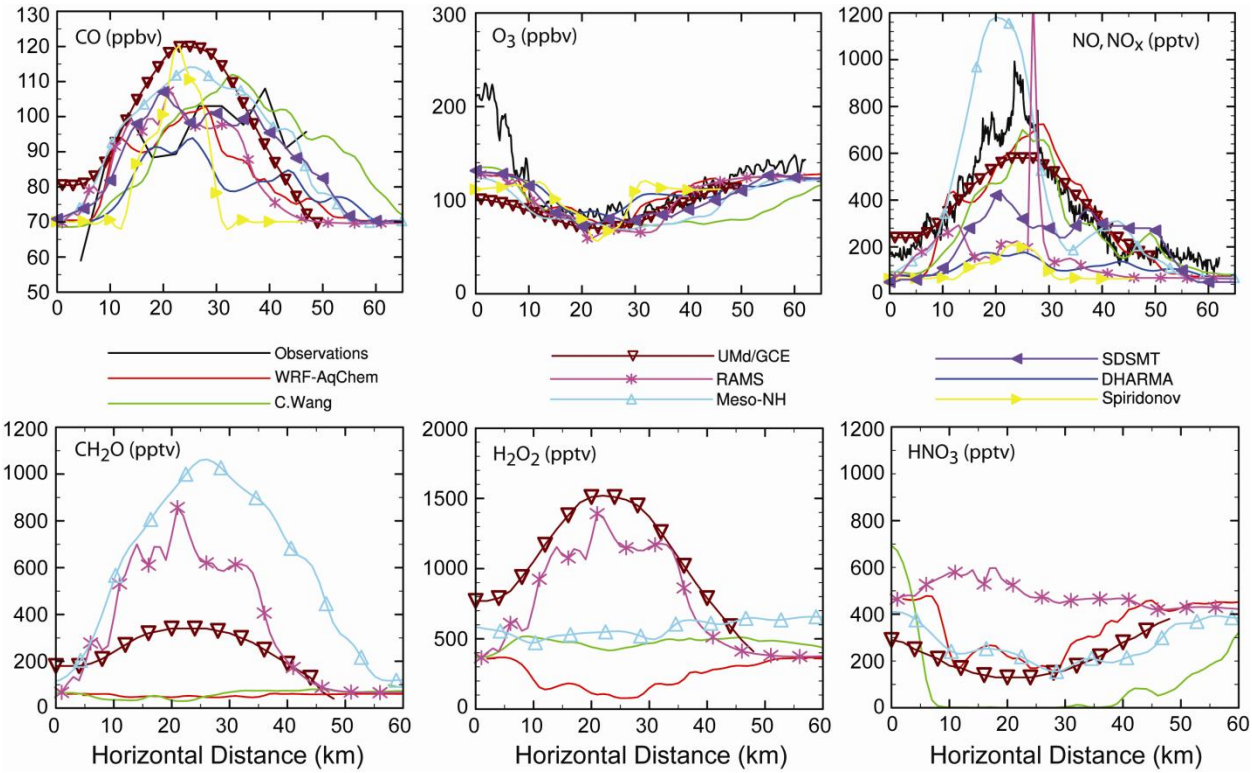

Figure 2. Results from a model intercomparison (Barth et al., 2007b) of passive tracers (CO, O_3), lightning enhanced NO_x, and soluble tracers (CH_2O, H_2O_2, HNO_3) plotted as an aircraft transect across the modeled anvil 50 km downwind of the storm. For CO, O_3, and NO_x, UND-Citation observations from the 10 July 1996 STERAO storm are included. Passive tracers compare well, while soluble species (including key HO_x precursors) show substantial variability among models.

- **Lightning and its production of nitrogen oxides**

Lightning is considered to be the largest natural tropospheric source of nitrogen oxides (NO_x = NO + NO_2; e.g., MacGorman and Rust, 1998). The strength of the global NO_x source from lightning has been estimated to range over an order of magnitude (2-20 Tg N yr^{-1}) (e.g., Bradshaw et al., 2000), although more recent estimates are in the 2-8 Tg N yr^{-1} range (Schumann and Huntrieser, 2007). Many factors affecting the contribution by lightning are still understood poorly. Such factors include the relative numbers of cloud-to-ground and intracloud flashes, the height distribution of flashes, the location of lightning relative to updrafts and downdrafts, and the amount of NO_x produced per flash or per unit length of channel or as a function of power dissipated by the channel. While field programs such as STERAO and EULINOX have examined some of these factors and have been important in constraining global estimates of lightning-produced NO_x, information on the height and structure of lightning obtained concurrently with chemistry observations in storm anvils remains elusive (e.g., Dye et al. 2000).

The current generation of lightning mapping systems (Rison et al. 1999) provides unprecedented information on the structure of lightning channels and higher order information such as flash duration and flash length (Thomas et al. 2004). Several recent studies (e.g., Wiens et al. 2005, Bruning et al. 2007, Deierling et al. 2008) have demonstrated the ability of VHF systems to map lightning relative to the kinematic and microphysical structure of storms to examine factors influencing lightning production and flash characteristics. DC3 will take advantage of VHF lightning mapping systems to determine factors influencing lightning-produced NO_x.

Hypothesis 3: The contribution of lightning to NO_x concentrations in the anvil, and subsequently in the upper troposphere, depends on overall flash rates and aggregate channel lengths at heights extending from just above the melting level to the uppermost region of the convective core. The amount of NO_x produced by a cloud-to-ground flash is on average roughly equivalent to that produced by an intracloud flash.

This hypothesis is based on the intuitive notion that the lightning that contributes to upper tropospheric NO_x is the lightning along trajectories that flow into the upper troposphere. Results from storm modeling studies (e.g., Pickering et al. 1998; DeCaria et al. 2000, 2005; Ott et al. 2005, 2007; Zhang et al. 2003b; Barthe and Pinty, 2007) suggest that the final vertical distribution of lightning-produced NO_x depends critically on the vertical distribution of the lightning itself and may vary from storm to storm.

Before 1996, the relative production of NO_x by individual intracloud flashes (IC) and cloud-to-ground flashes (CG) had been based on early studies that suggested IC flashes were less energetic than CG flashes (e.g. Holmes et al., 1971), and many estimates of lightning NO_x production have assumed that the amount of NO_x produced per IC flash (P_{IC}) is less than the production by a CG flash (P_{CG}). Price et al. (1997), for example, assumed that P_{IC} was one tenth of P_{CG} in calculating global lightning NO_x production. On the other hand, Gallardo and Cooray (1996) suggested that IC flashes may dissipate nearly as much energy as CG flashes and therefore P_{IC} may be on the order of P_{CG}. Estimates from field data (Figure 3) indicate that in some cases P_{IC} may be greater than P_{CG}, and is quite likely that it is at least comparable to P_{CG} on average. While DC3 measurements will not measure P_{IC} or P_{CG} directly, it should be possible to evaluate their relative magnitudes from measurements of NO_x in small storms having relatively low flash rates monitored by lightning mapping arrays that can locate flashes in 3 dimensions.

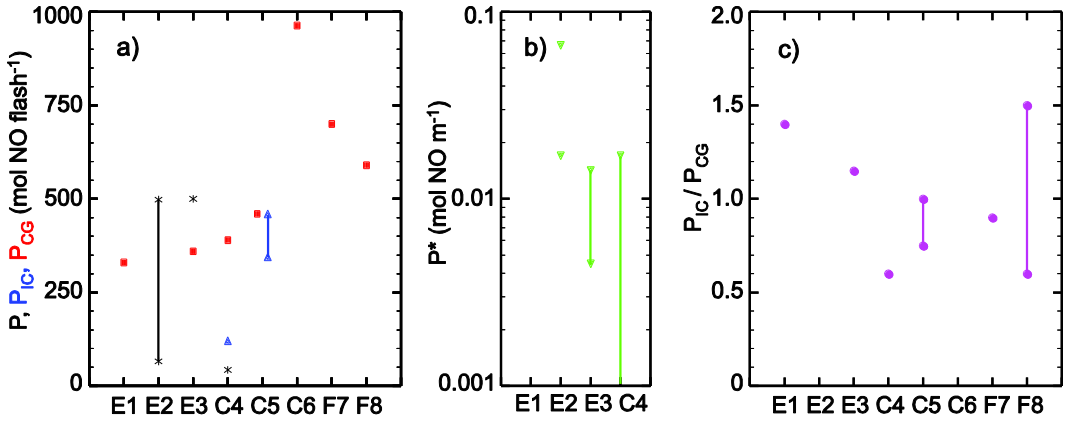

Figure 3. Production of NO from lightning, literature review. a) Estimate of total (black), intracloud (blue), and cloud-to-ground NO production per lightning flash as a function of storm studied; b) estimate of NO production per lightning flash length as a function of storm studied; and c) estimated ratio of NO production from IC flashes to CG flashes as a function of storm studied. E1 is the 21 July 1998 EULINOX storm (Fehr et al., 2004); E2 a LINOX storm in S. Germany and Switzerland (Huntrieser et al., 1998; Höller et al., 1999); E3 the 21 July 1998 EULINOX storm (Huntrieser et al., 2002; Thery et al., 2000; Ott et al., 2007). C4 is the 10 July 1996 STERAO storm (Stith et al., 1999; Skamarock et al., 2003; Ott, 2006; Barthe and Barth, 2008); C5 the 12 July 1996 STERAO storm (DeCaria et al., 2000, 2005); C6 the 12 September 2002 Colorado storm (Langford et al., 2004). F7 is the 16 July 2002 CRYSTAL-FACE storm (Ott et al., 2005); F8 the 29 July 2002 CRYSTAL-FACE storm (Ott et al., 2005).

DC3 will evaluate these hypotheses and thereby quantify lightning's effectiveness as a source of NO_x in storm anvils and the upper troposphere for three geographic regions with very different storm characteristics. To extend these results more widely and to use them in global chemistry models, it will be necessary also to determine how meteorological factors affect the height, storm-relative location, flash rates, and channel lengths of lightning – which are topics of great interest to the field of storm electrification, as well as to the tropospheric chemistry community.

Climatologies of U.S. storm and lightning characteristics (e.g. Boccippio et al., 2001; Williams et al., 2005), reveal that storms in the southeastern U.S. (Alabama) often are associated with very moist subcloud layers, low cloud base heights (CBH), and weak shear, with warm rain processes making a significant contribution to precipitation formation. Storms in the high plains regions (e.g., Colorado) are associated with drier subcloud layers, increased cloud base heights, and form in environments with strong low level shear. Little, if any, precipitation forms by warm rain processes. Supercell storms, one of the principal targets of DC3, can occur in all three environments, but are most prevalent in the central plains region (Oklahoma), which tends to have intermediate values of subcloud moisture and CBH and large values of low-level shear. Besides affecting the transport and processing of chemical species by storms, these variations in storm properties (e.g., CBH, convective available potential energy, and the amount of sub-cloud moisture) may also modulate flash rates, IC to CG ratios, and the propensity for high peak current positive CG lightning (Knapp, 1994; Lang and Rutledge, 2002; Williams et al., 2005), which are in turn controlled by storm lifecycle, precipitation physics and storm dynamics. Boccippio et al. (2001), for example, found that the proportion of cloud-to-ground flashes tends to be larger in the southeastern U.S. compared to the central and western high plains. Similarly, MacGorman et al. (2006) showed that CG lightning production can be delayed and reduced in storms on the high plains and hypothesized that the delay is related to the low precipitation efficiency of the high plains storms. DC3 investigators expect to sample a wide spectrum of storms, including weak isolated storms, low and high precipitation supercell storms, to evaluate how various storm characteristics affect lightning production by storms and NO_x production by lightning.

Hypothesis 4: The flash rates of a storm are proportional to the volume of updrafts greater than 10 m s^{-1} in the -10˚C to -40˚C layer and to storm graupel echo volume. Cloud-to-ground lightning occurrence usually follows the occurrence of precipitation in the 0˚C to -10˚C layer after graupel has appeared in this region or higher regions. Conversely, cloud-to-ground lightning occurrence is inhibited in storms that produce little precipitation.

Several modeling studies (Kuhlman et al., 2006, Cohen, 2008) and observational studies (Wiens et al., 2005, Lhermitte and Krehbiel, 1979) have found that lightning rates increase as the updraft and graupel echo volume increase (Figure 4). However, only a few cases have been documented with co-located Doppler, polarimetric radar, and VHF lightning network observations. Observations of a wider spectrum of storms are needed to quantify and more fully test this relationship. DC3 will provide such measurements.

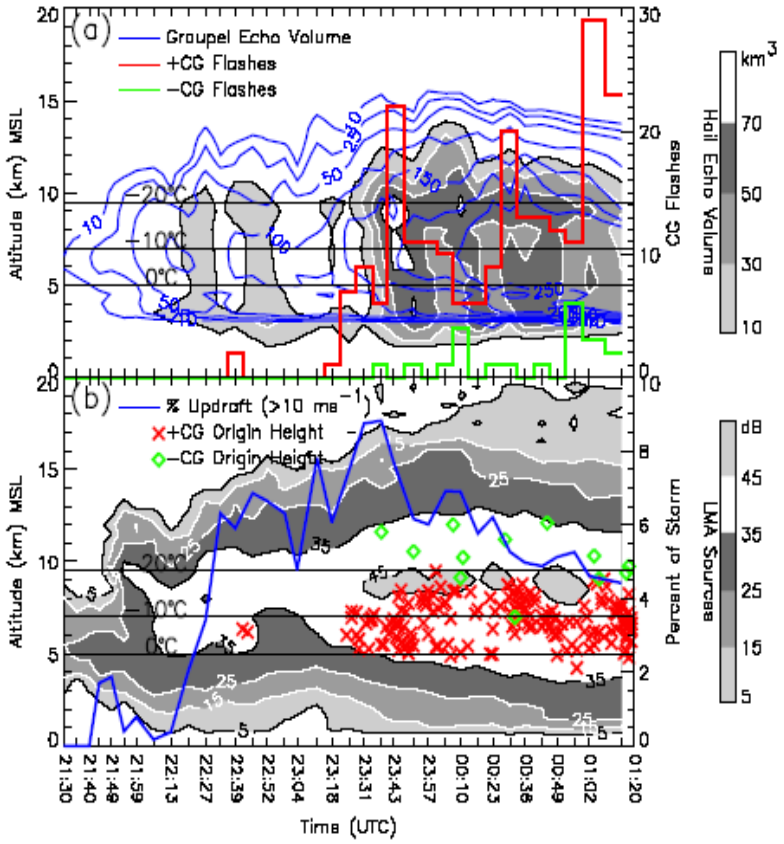

Figure 4. (a) Contours of the number of radar grid points at each time and height classified as hail (in gray scale) and graupel (blue). The number of grid points has been multiplied by the dimensions of a grid cell $(0.5 \text{ km})^3$, resulting in units of echo volume. The total number of +CG and –CG flashes during each volume scan are plotted in red and green, respectively. Here, the CG flashes are summed (not averaged) over the duration of each volume scan. (b) Time height contours of total LMA sources (in gray scale) normalized to a five-minute time interval. Contour values are in decibel units (i.e., $10log_{10}$(# sources)) due to the large range of values. The percent volume of updraft exceeding 10 m s^{-1} is overlaid onto (b) as a thick blue line. Origin altitudes of +CG and –CG flashes are overlaid onto (b) as red X's and green diamonds, respectively. Adapted from Wiens et al. (2005).

Several case studies (e.g., MacGorman et al., 1989; Carey and Rutledge, 1996; Bruning et al. 2007) have found that production of cloud-to-ground lightning typically follows the appearance of precipitation, particularly graupel, near and slightly colder than the freezing level, but investigation of this behavior is needed to better quantify the behavior and to examine whether it is true in a broader spectrum of storm

types. The availability of DC3's lightning data for regions with widely divergent climatologies, including the low-precipitation storms of the high plains, will allow this hypothesis to be thoroughly tested.

Hypothesis 5: Storms that produce inverted-polarity IC flashes in the upper part of storms and inverted-polarity CG flashes are those in which a large fraction of the adiabatic liquid water profile is realized as cloud liquid in the mixed phase region.

The measurements that are planned for DC3 and the types of storms that will be observed are expected to allow this important topic to be investigated. A major finding of the STEPS field program (Lang et al. 2004) was that a large fraction of storms in the high plains have inverted-polarity lightning, and in fact, somewhat unexpectedly, appear to have inverted-polarity electrical structures. This has also been observed in some severe storms on the central plains (MacGorman and Burgess 1994), but rarely if ever occurs in the southeastern U.S. (Knapp 1994; Carey et al. 2003). Williams et al. (2005) and Carey and Buffalo (2007) attributed the inverted-polarity lightning to the strong updrafts, high cloud bases, and negligible warm rain processes in storm updrafts on the high plains. MacGorman et al. (2005, 2008) attributed the behavior to the low subcloud moisture and low precipitation efficiencies of storms in the region, because of the reduced recirculation of precipitation into the updraft. Both explanations suggest that the result is to reduce scavenging of cloud liquid particles below the mixed phase region, so that graupel riming is large enough to cause graupel to gain positive charge over most of the mixed phase region, thereby inverting the usual charge distribution.

- **Chemistry in the anvil.**

Processing of trace gases in the anvil as the air flows away from the storm core(s) is crucial to determining the concentrations of gases once the cloud particles have subsided or evaporated. The anvil region is the ice cloud outflow resulting from the divergence of the convective core updrafts occurring just below the tropopause. The anvil is characterized as a region of dissipating turbulence from the convective cores, but as time proceeds radiative cooling at the anvil top can create a weak convective eddy circulation. The chemical composition of the anvil region will depend on both the composition of the updraft reaching the anvil including the enhancement of NO_x from lightning production, and the solar radiation field in the anvil, which is especially dependent on the presence of small ice crystals.

Hypothesis 6) The chemical composition of the convective outflow within and near the visible anvil will be stratified into a top layer with high radiation fluxes accelerating radical chemistry, and a lower layer with low radiation fluxes and near nighttime-like radical chemistry.

The composition of the outflow of the convective cores is the initial condition for the subsequent photochemistry of the convective outflow air mass. This air will include low solubility chemical constituents, such as CO, NO, NO_2, O_3, and hydrocarbons, transported from the boundary layer (below cloud). Soluble species, such as sulfur dioxide (SO_2), CH_2O, H_2O_2, CH_3OOH, and carboxylic acids, will be partially in the gas phase (interstitial air) and partially in cloud particles. Nitric acid and hygroscopic aerosol species will enter the anvil in the cloud particles. The NO_x vertical distribution in the anvil will depend on the frequency and location of the lightning and the storm dynamics and structure. Ozone in the anvil may be either greater than or less than that in the storm surroundings depending on the relative amounts in the PBL and UT prior to the storm and on whether photochemical production or loss occurs during the convection.

The main factor affecting the anvil composition is the role of shortwave radiation driving the photochemistry. Madronich (1987) showed for stratiform type clouds that the flux impinging on a sphere (i.e. the actinic flux) is enhanced above cloud, can be even more enhanced just below cloud top, and reduced near and below cloud base compared to clear sky actinic fluxes. Measurements of the NO_2 photolysis rate coefficient vertical profile (Früh et al., 2000) confirm the enhanced rate coefficients near cloud top and

reduced rate coefficients near cloud base. With enhanced photolysis rates near cloud top, the ozone radical photochemistry will be accelerated. For example, in a modeling study of tropical deep convection and chemistry, Wang (2005) found increased ratios of NO_2 to NO below the cloud anvil and within the storm and decreased ratios at and near the top of the cloud anvil due to cloud scattering of solar radiation. The study also reported that OH concentrations were increased by 20% due to enhanced photolysis rates in the uppermost cloud layer.

D.3.b) Goal 2: Quantify the changes in chemistry and composition after active convection, focusing on 12-48 hours after convection and the seasonal transition of the chemical composition of the UT

- **12-48 hours after convection**

Downstream of the active convection, the chemistry in the convective outflow, which has elevated NO_x and volatile organic compounds (VOC) mixing ratios, can dramatically increase ozone production in the upper troposphere. Model estimates of ozone production rates for midlatitude convection range from 4-15 ppbv/day (Pickering et al., 1990). The mechanistic details of ozone photochemistry in the upper troposphere are still not fully understood because there continues to be a discrepancy between HO_x observations and theoretical predictions for the UT. Photochemical models predict for upper tropospheric NO_x values greater than a few hundred pptv, that production of nitric acid becomes an important HO_x loss. Thus, further NO_x increases reduce HO_x levels and lower the efficiency with which NO_x produces ozone. Recent INTEX HO_x observations (Ren et al., 2008), however, do not respond to increases in NO_x as expected and continue to remain elevated even for NO_x levels exceeding 1 ppbv. Furthermore, the HO_x precursor, CH_2O, also has measurement-model discrepancies for high NO conditions in the UT (Fried et al., 2008). The elevated HO_x suggests that there are stronger radical sources than models predict which would enable much greater ozone production rates than expected. There are numerous measurements of high NO_x levels in the upper troposphere, but very few with associated HO_x and CH_2O measurements. The observations proposed for DC3 are needed to see if there is some higher threshold of NO_x beyond which observed HO_x begins to decrease and to help elucidate the possible chemical mechanisms occurring.

Ozone production rates calculated by numerical models have shown a dependence on boundary layer composition and lightning flash rate production of NO_x. For the low pollution, moderate flash rate of STERAO (~1 ppbv NO_x in anvil), downstream O_3 production rates were ~8 ppbv/day (Ott, 2006) and for the moderate pollution, high flash rate of EULINOX (~3.5 ppbv NO_x in anvil) were ~10 ppbv/day (Ott et al., 2007). These results show that downstream ozone production may be less efficient with storms with high flash rates or with those transporting significant pollution. By sampling convection and its convective outflow in 3 locations with different boundary layer composition, the DC3 campaign can contrast O_3 production downwind of active convection.

Hypothesis 7) In sampling the convective plume 12-48 hours after convection, we expect to find that 8-12 ppbv ozone will be produced per day due to high NO_x and enhanced concentrations of HO_x precursor species. The ozone production will vary in a complex nonlinear fashion depending on the NO_x and VOC abundance transported to the anvil from the boundary layer and the amount of NO_x produced by lightning.

We expect to find large variations in surface emissions (Table 1) in the DC3 regions allowing us to study a wide range of chemical environments. The emission rates listed in Table 1 are based on the low level air flow coming from the SSE which is common in all 3 locations. Any variations in the airflow may give very different emission scenarios (e.g. the cleaner air to the NNW of the CSU-CHILL radar). Further, actual emissions on any given day are likely to be quite different from these typical summer day emissions used here. Emissions in Colorado exemplify a lower NO_x, higher CO and VOC scenario. In Oklahoma anthropogenic NO_x, CO, and VOC emissions are lower relative to the other DC3 sites, while in

Alabama these emissions are higher than the other 2 DC3 sites. Biogenic VOC emissions (Figure 5 and Table 1) are greatest in Alabama and weakest in northern Colorado.

Table 1. Anthropogenic emission rates (kg per day per km^2) for a typical summer day in the three DC3 regions. Sources are July weekday average EPA NEI-2005 on-road and non-road transportation sector, August 2006 weekday average point emissions from CEMS, and EPA NEI-2002 area source emissions inventory. Biogenic isoprene emissions are calculated by the Model of Emissions of Gases and Aerosols from Nature (Guenther et al., 2006). Courtesy of Stu McKeen (NOAA/ESRL) and Christine Wiedinmyer (NCAR).

Species	Colorado	Oklahoma	Alabama
NO_x	7.73	6.93	12.5
CO	42.3	29.6	60.8
VOC	14.4	8.08	13.2
Biogenic C_5H_8	1.90	10.5	21.8

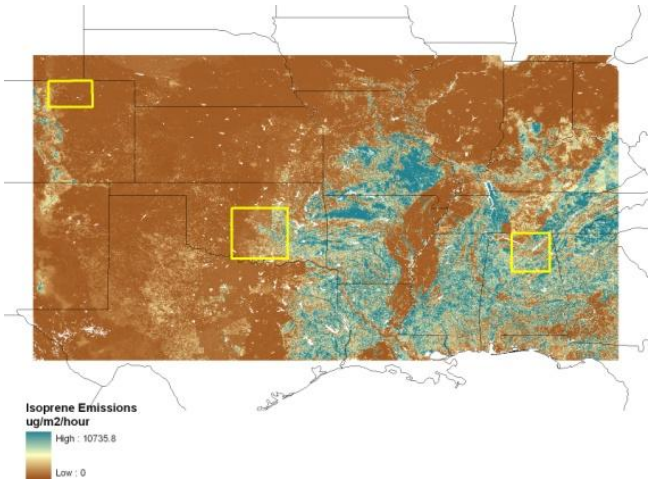

Figure 5. Isoprene emissions calculated by the MEGAN model (Guenther et al., 2006) for the southern central U.S. on a typical June day. Courtesy of Christine Wiedinmyer.

- **Seasonal transition of O_3 in the UT**

The extension of the impact of deep convection on upper tropospheric composition from the 1-2 day time period to the month-long and seasonal impact is also important. Modeling studies by Zhang et al. (2003a) and Li et al. (2005) suggest that a widespread UT ozone enhancement should form above the southern U.S. during summer due to convective transport of O_3 and its precursors (especially formaldehyde) followed by in situ production with NO_x (both convectively transported from the surface and produced from lightning). The presence of the UT ozone enhancement was confirmed with a broad network of measurements that utilized IONS ozonesondes and MOZAIC aircraft ozone profiles during July-August 2004 and August 2006 (Cooper et al., 2006, 2007). The UT ozone enhancement is associated with the North American monsoon and its upper tropospheric anticyclone that traps ozone precursors over the southern U.S. and northern Mexico. The long residence times in the anticyclone allow for strong ozone production that is mainly driven by lightning NO_x, producing average monthly ozone enhancements above the southeastern U.S. of 30-50 ppbv in comparison to upwind sites along the North American west coast (Cooper et al. 2007). A modeling study exploring the fate of a NO_x tracer released from the locations of all detected CG lightning flashes across North America during summer 2004 and 2006 (Cooper et al., 2009) verifies the strong connection between the observed increase of upper tropospheric NO_x above the southeastern U.S. and the formation and strengthening of the UT anticyclone.

Hypothesis 8) Survey flights at the end of June from the central U.S. to the northern Caribbean will find the greatest UT ozone and NO_x mixing ratios above the Gulf of Mexico and Florida. Daily ozonesonde/lidar profiles from Huntsville, Alabama will document the seasonal buildup and decay of the UT ozone enhancement from May to September.

The upper tropospheric ozone enhancement is the largest average ozone feature in North America with upper tropospheric ozone values exceeding lower tropospheric values by 20 ppbv (Cooper et al., 2007). Ozone measurements have captured the strength of the ozone enhancement along its northern and western boundaries, and have so far shown that the greatest enhancements occur above Texas or Alabama, but measurements are lacking across the Gulf of Mexico and the southern and eastern boundaries. Model studies indicate that NO_x and ozone may reach maximum values anywhere from northern Mexico, across the Gulf of Mexico to Florida (Cooper et al, 2007, 2008; Hudman et al., 2007). While these same studies have shown skill at predicting the general position and strength of the ozone maximum, they still under predict ozone values near the center of the ozone maximum, and also under predict median NO_x mixing ratios. DC3 survey flights across the upper tropospheric ozone maximum and daily sonde/lidar ozone profiles from Huntsville, Alabama will provide important information on the build-up of ozone and ozone precursors within the upper tropospheric anticyclone. These data will not only place DC3 within the broader context of the seasonal and regional impact of thunderstorms on the UT composition, but will place better constraints on model simulations of lightning NO_x emissions and ozone production.

D.3.c) Ancillary goals complementary to DC3

The DC3 experiment has two ancillary studies. These studies are complementary to the core objectives of DC3, and are expected to produce significant results for studies related to aerosols and thunderstorms and to UTLS halogen chemistry.

- **Thunderstorms and aerosols**

Studying the processing of aerosols by thunderstorms and conversely the effect of aerosols on thunderstorm structure, dynamics, and lightning is of paramount importance because of the implications for climate and water resources. The DC3 focus, however, is on the impact of thunderstorms on UT composition and chemistry, therefore placing aerosol – thunderstorm interactions as a secondary objective. As part of DC3, we will characterize with payload-accommodating instruments the aerosol number and mass concentrations in the inflow and outflow regions of the storms. Where permitted we will also obtain ice concentration measurements in the anvil of the storms. These measurements will give valuable information on physical characteristics of the aerosols that can be used to set up a more detailed and focused aerosol – thunderstorm interaction experiment.

- **Transport of halogens**

Quantifying the amount of inorganic bromine (Br_y) in the UTLS region is important for understanding its impact on stratospheric ozone trends and the associated chemistry/climate interactions. Br_y can result from the rapid breakdown of short-lived organic source gases, such as $CHBr_3$ and other brominated methanes. Convective transport of source gases and chemical processing in the upper troposphere are expected to significantly influence the amount of Br_y near the tropopause. As part of DC3, we will include measurements of long and short-lived organic halogen source gases and the amount of at least one inorganic bromine and chlorine containing compound (e.g. BrO and HCl) in order to calculate inorganic halogen amount and the partitioning among various reactive and reservoir species.

D.4 Conduct of the research, experiment techniques, observing systems

D.4.a) Brief description of experimental techniques

Two aircraft, the NSF/NCAR GV (HIAPER) and the NASA DC-8, are requested to be flown during DC3 and will be based initially in Colorado and then in Kansas or Oklahoma. These aircraft will sample storm inflow and outflow air in three central U.S. locations: northeastern Colorado, central Oklahoma, and northern Alabama. Because convection at one location often occurs for several consecutive days followed by a quiescent period, the sampling strategy will be to give priority to storms occurring in one of the 3 regions for a 4-9 day period and then to shift the focus to another region.

DC3 will attempt to characterize fully the flow of air into, through and out of deep convection. The deployment of the aircraft platforms will be driven by forecasts for the timing and location of the development of convective storms in the areas under study. Once the aircraft have reached one of the three study regions, close interaction with scientists at that radar location will be established to fine tune flights plans. The high-altitude aircraft (HIAPER) will sample the convective outflow in the anvil and downstream, while the low-altitude aircraft (DC-8) will sample primarily the inflow, and the air at mid-troposphere altitudes. Both aircraft will be utilized to locate and sample convective outflow air 12-48 hours downstream of the active convection. The GV (HIAPER) aircraft will also fly one or more survey flights at the end of June in the upper troposphere from the central U.S. to the northern Caribbean. Ground-based Doppler and polarimetric radar will sample the storms to give kinematic and microphysical information, and lightning mapping arrays will be used to provide 3-dimensional lightning locations. The ground-based networks will document all storms within range during DC3, regardless of whether, or not, the DC3 aircraft are operating in that study region. Other ground-based observations include balloon sondes for thermodynamic and limited chemical information as well as precipitation collection and its chemical analysis. Satellite data will be used to place the airborne and ground-based measurements in the context of the wider geographical regions and help guide sampling strategies.

D.4.b) Distribution and use of facilities and platforms

DC3 will make use of ground-based facilities, balloon sondes, precipitation collection, and extensively instrumented aircraft platforms. Each instrument and facility is selected to specifically address one or more DC3 goal. We will utilize a variety of ground-based facilities tailored to each measurement site:

1. Northeastern Colorado centered on the CSU-CHILL and Pawnee radar facilities operated by Colorado State University;
2. Central Oklahoma using facilities maintained by the National Severe Storms Laboratory and University of Oklahoma including SMART-Rs, the KOUN polarimetric radar, and Oklahoma lightning mapping array; and
3. Northern Alabama using facilities operated by the University of Alabama-Huntsville and NASA Marshall Space Flight Center including the ARMOR polarimetric radar, the MAX X-band polarimetric radar, northern Alabama lightning mapping array, and MIPs mobile sounding facility.

In all three regions, multiple Doppler and polarimetric radars will provide airflow and hydrometeor information for the observed storms. For these same regions, three-dimensional VHF lightning mapping data (LMA networks) will be supplemented with cloud-to-ground lightning data from the National Lightning Detection Network operated by Vaisala to provide ground strike locations, peak current, current polarity, and number of strokes per flash (multiplicity). The availability of 3-D lightning mapping arrays is pivotal for DC3 proposed science. These networks are in place in Oklahoma and Alabama and a portable system will be deployed in Colorado for the field campaign. Balloon sondes for thermodynamic and limited chemical information will be deployed, as well as precipitation collection and its chemical analysis. Besides the two primary aircraft platforms, the NSF/NCAR GV (HIAPER) and the NASA DC-8, DC3 would provide an excellent opportunity to operate a storm penetrating aircraft (e.g. the A-10) if it

is available by the time of DC3. Other aircraft platforms may also contribute, potentially including the German DLR GV or Falcon, the UK BAe-146, the UND Citation, the NASA WB-57, and the DOE G-1. These platforms are viewed as complementary to DC3 as the primary aircraft being requested are the NSF/NCAR GV and the NASA DC-8. Satellite data will be used for storm tracking and placing the ground-based and in situ measurements in the regional scale context. Satellite data that will be utilized include geostationary satellite images from the GOES-EAST and GOES-WEST platforms, TRMM/LIS data measuring lightning flashes, Aura/TES CO and O_3 profiles, Aura OMI O_3, NO_2, CH_2O, SO_2 and aerosol columns, Terra and Aqua/MODIS data of aerosol optical depth, Terra/MOPITT CO data, Aqua/AIRS CO data, CloudSat cloud radar reflectivity, and Calipso aerosol profiles and cloud top data.

D.4.c) Optimal time frame for field operations

The DC3 experiment is planned for May – June 2012. The first week of the field deployment will be used to begin the intensive operations and to mitigate any instrument, aircraft and forecast model problems that inevitably arise at the beginning of large-scale field missions. The following 6-7 week period has several advantages, which are described here. 1) The month of June is a good "intersection" month that yields relatively high precipitation often driven by daytime convection at all three sites. This month also provides a high incidence of air mass thunderstorms, with the additional occurrence of shear driven convection during the first half of the month. 2) The last half of May provides a high incidence of shear driven convection at all three sites. 3) While mesoscale convective systems and complexes occur in this time frame they do not dominate convective activity, providing ample, isolated deep convective storms. 4) This is the best time period for deep, isolated convection within the range of the CSU-CHILL radar in northeast Colorado. 5) The month of June provides the longest possible daylight hours allowing aircraft missions to extend into the early evening, plus ample daylight for the investigation of photochemistry.

D.4.d) Education and Outreach

A variety of education and outreach activities are planned for DC3, with opportunities especially for students at undergraduate and graduate levels. The field operations of DC3 will include the participation of undergraduate and graduate students who will contribute to instrument development and/or operation, analysis and integration of data, and numerical modeling of convection and related chemistry. We expect to support a number of graduate students who will analyze DC3 data as part of their theses and dissertations. We will work with CoCoRaHS (Colorado Cooperative Rain and Hail Studies) volunteers to collect and report precipitation and with undergraduate and high school students (local to each DC3 field site) to collect and analyze precipitation for its chemical composition. Through the UCAR SOARS program, students from underrepresented groups will contribute to data collection and analysis. Other education activities will include a seminar series and/or student forecast and flight planning class. There will also be several different Internet web sites with descriptive and instructional material about the project. Outreach days will be planned for the public and local media at some of the project's locations. These days will allow the public to tour the aircraft and ground facilities and engage the local media to educate the public on the goals and status of DC3.

D.4.e) Broader Impacts of DC3 Measurements

Post-experiment analysis will include modeling at both cloud-resolving scales and cloud-parameterized scales (e.g. regional and global models). The cloud resolving models will be evaluated with the field observations and used to develop parameterizations for large-scale models. Parameterization development will be for convective transport of air, water, and trace gases, production of NO by lightning (e.g. Ott et al., 2007; Barthe and Barth, 2008), and wet deposition of soluble species. The large-scale models will be used to analyze the regional impact of convection on the chemistry. Process-scale chemistry models will be used to analyze mechanistic aspects of the photochemistry in the UT (e.g. Crawford et al., 2000; Fried et al., 2008). The experimental data and model analyses will be provided for mod-

el intercomparison exercises and assessment reports. The data collected during DC3 will undergo quality assurance during the first year after the experiment and will be released to the community two years after the experiment.

D.4.f) Related prior field campaigns

Several previous experimental studies in the continental midlatitudes focused on the perturbations to NO_x (e.g. STERAO, CRYSTAL-FACE, EULINOX) by individual storms utilizing detailed storm kinematic, microphysical, and electrical measurements, but did not address convective processing of HO_x or its precursors (e.g. peroxides, carbonyls, and hydrocarbons). Other field studies inferred the influence by deep convective storms upwind of the measurement locations (e.g. INTEX-A, TRACE-P), but the storm structure and dynamics were not measured. The main strength of these studies was the clear demonstration that ozone photochemistry is perturbed by convective outflow. The recent START08 field campaign flew the NSF/NCAR GV with a limited chemistry payload near convection that did not have supporting ground facilities to fully analyze the effect of deep convection on UT chemistry. The incomplete data and sampling strategies from these prior field programs do not allow us to establish a complete description of convective cloud processes on HO_x, its precursors and NO_x. Knowledge of HO_x, its precursors and NO_x mixing ratios together is needed to ascertain ozone production in the upper troposphere. Finally, DC3 will build on results from STEPS 2000 that demonstrated relationships between storm kinematics, microphysics, electrical structures and lightning.

The DC3 field experiment will improve our current knowledge of convection and chemistry by providing a comprehensive suite of chemical measurements near convection and downstream of convection at altitudes that extend to at or near the tropopause. These chemical measurements will be obtained within the context of excellent kinematic, microphysical and electrical ground-based measurements. The suite of chemical, kinematic, microphysical, and electrical measurements will provide the necessary information to estimate ozone sources and sinks in the upper troposphere where ozone is radiatively active as a greenhouse gas.

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