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INTRODUCTION TO A SPECIAL SECTION

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Special Section:
Deep Convective Clouds and Chemistry 2012 Studies (DC3)

Key Point:

- Key findings from the DC3 project on convective transport, chemistry, lightning, and cloud physics are summarized

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Introduction to the Deep Convective Clouds and Chemistry (DC3) 2012 Studies

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Abstract The Deep Convective Clouds and Chemistry (DC3) project aimed to determine the impact of deep, midlatitude continental convective clouds on tropospheric composition and chemistry. The DC3 field campaign was conducted over a broad area of the central United States during May–June 2012. Data collected by DC3 have been extensively analyzed, with many results published in Deep Convective Clouds and Chemistry 2012 Studies (DC3), a joint special section of *JGR Atmospheres* and *Geophysical Research Letters*. This paper highlights key results from the DC3 project as an introduction to the special issue.

1. Introduction

The Deep Convective Clouds and Chemistry (DC3) project aimed to determine the impact of deep, midlatitude continental convective clouds on tropospheric composition and chemistry. The DC3 field campaign was conducted over a broad area of the central United States during May–June 2012. Three extensively instrumented research aircraft were based in Salina, Kansas, and were flown to thunderstorms that occurred in northern Alabama, northeastern Colorado, and west Texas to central Oklahoma, where ground-based research radars, lightning mapping arrays (LMAs), and radiosondes were located. Over 300 participants, including more than 100 students and 30 postdoctoral scientists, were involved in the sampling of 20 storms over these three regions. Data collected by DC3 have been extensively analyzed, with many results already published in Deep Convective Clouds and Chemistry 2012 Studies (DC3), a joint special section of *JGR Atmospheres* and *Geophysical Research Letters*.

The DC3 project had two major goals. The first was to quantify and characterize the convective transport of fresh emissions and total water (vapor and condensate) 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, and chemistry in thunderstorm anvils. The second goal was to quantify the changes in chemistry and composition in the upper troposphere following active convection, focusing on 12–48 hr after convection and the seasonal transition of the chemical composition of the upper troposphere. To accomplish the first goal, the DC3 science team performed research-grade numerical weather predictions for thunderstorm forecasts and employed a team of nowcasters to provide weather updates while the research aircraft collected measurements on the composition in the inflow and outflow regions of thunderstorms, ground-based radar documented cloud structure and kinematics, and ground-based LMAs recorded lightning characteristics, most notably flash rates and the 3-D location of lightning channels in thunderclouds. DC3 was the first campaign to gather a comprehensive suite of airborne composition measurements while simultaneously collecting detailed storm characteristics. To address the second goal, locations of aged convective outflow regions of previously sampled storms were forecasted with numerical models and trajectory models. These regions were then targeted for aircraft sampling, using ratios of various trace gases to identify air recently transported from the boundary layer. The second goal included sampling the convective outflow of a dissipating mesoscale convective system (MCS). DC3 was unique compared to previous field experiments in that it intentionally investigated the photochemical aging of the convective outflow by tracking the outflow plume to regions downwind. More details of the experimental design and highlighted storm cases can be found in Barth et al. (2015).

As is common with field experiments, science discoveries on topics other than the main goals were made. Here we introduce the findings from the DC3 project aligned with the following science topics: (1) convective transport and scavenging, (2) production of nitrogen oxides (NO_x) by lightning, (3) lightning flash characteristics, especially related to storm dynamics and microphysics, (4) chemistry in convective outflows, (5) stratosphere-troposphere exchange, (6) tropospheric composition (trace gases and aerosols), and (7) cloud structure.

2. Findings From DC3

2.1. Convective Transport

Although it is understood that severe storms transport more air and boundary layer constituents to the upper troposphere than air-mass type storms, the underlying mechanisms causing the differences in convective transport were not well understood. To examine the convective transport of trace gases in three different types of storms sampled during DC3, measurement analysis and cloud-resolving modeling were applied to an air-mass storm in Alabama, a severe convective storm in Oklahoma, and a MCS in Missouri/Arkansas. This analysis found that convective transport of trace gases, which was the strongest (per unit area of storm) in the severe thunderstorm compared to the other two storms, was controlled more by the vertical gradient of the mass flux rather than the vertical gradient of the trace gas mixing ratio (Li et al., 2017). Within MCSs, it was found that rear inflow jets may weaken the convective transport of boundary layer constituents by transporting clean air into the storm that is entrained into the inflow air. These same storms were simulated at convective parameterization scales ($\Delta x = 12$ km and $\Delta x = 36$ km) using the Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) to evaluate the capabilities of five convective parameterizations with observations. Li et al. (2018) showed that the convective transport was weaker than the cloud-resolving simulations and was controlled more by the convective parameterization during the early phase of the storm rather than during the later stage of the storm. Importantly, Li et al. (2018) pointed out the need to have a convective transport parameterization consistent with the subgrid convective cloud scheme. In addition, there is a need for having a physically realistic representation of ice microphysics in the convective parameterization.

As air near the Earth's surface is ingested into thunderstorms, we expect most aerosols to be removed by precipitation via nucleation scavenging and impaction scavenging. DC3 observations indicate that over 80% of the sulfate, nitrate, and organic aerosol mass was removed during transport to the upper regions of the storm (Yang et al., 2015). Corr et al. (2016) found that more than half of the coarse-mode dust mass concentrations are transported to convective outflow regions, suggesting deep convection may be an important mechanism for transporting coarse-mode dust to the upper troposphere, where these dust particles serve as ice nuclei for cirrus cloud formation and are therefore important for cloud radiation and climate effects.

With the aim of understanding and quantifying ozone production in the upper troposphere, it is important to first determine how much of the precursors that chemically react to form ozone are being transported to the upper troposphere via deep convection, namely, hydrogen oxide radicals (HO_x), volatile organic compounds, and nitrogen oxides (NO_x). Barth et al. (2016), Fried et al. (2016), and Bela et al. (2016) quantified scavenging efficiencies of key ozone precursors, formaldehyde and two peroxides, by comparing these trace gas concentrations in the storm inflow and outflow regions. They found that 80–90% of hydrogen peroxide, 40–60% of formaldehyde, and 12–84% of methyl hydrogen peroxide were scavenged by a variety of storms sampled during DC3. The removal of hydrogen peroxide and formaldehyde did not strongly depend on the fraction of trace gas retained in ice during the cloud drop freezing process, likely because these more soluble trace gases were mostly removed in the liquid-dominated region (at $T > 0$ °C) of storms, while methyl hydroperoxide scavenging had some dependence on its retention in freezing drops (Bela et al., 2018). Using the WRF-Chem model at convective parameterization scales, Li et al. (2019) found that wet scavenging overestimated removal of these soluble trace gases, but the predicted scavenging efficiency improved compared to observations when ice retention fractions for each trace gas were included in the parameterization. In general, observations in the upper troposphere outflow regions found that formaldehyde had the highest mixing ratios of these three HO_x precursors and was calculated to be the dominant HO_x precursor for ozone formation downwind of the storms (Brune et al., 2018).

2.2. Lightning Flash Rates and Lightning-NO_x Production

Lightning plays an important role in atmospheric chemistry through its production of NO_x (= NO + NO_2). Model parameterizations of lightning-generated NO_x depend on lightning flash rates predicted for the storm and an estimate of how much NO is produced per lightning flash, which may depend on the type of flash (i.e., cloud-to-ground or intracloud). Because explicit prediction of charge in a model is computationally expensive, predicting the lightning flash rate often depends on empirical relationships of storm parameters and lightning flash rate (e.g., Deierling & Petersen, 2008). Evaluation of these storm parameter-flash rate relationships in previous studies gave variable results (some relationships predicted flash rates well for one type of storm but not another; Barthe et al., 2010), suggesting more studies need to be done. Analysis of flash rates and storm characteristics for severe convective storms in Colorado suggested that existing parameterization schemes of flash rate based on updraft intensity replicated observed flash rates less successfully than a new parameterization based on the >35-dBZ radar volume of the storm (Basarab et al., 2015). In addition to flash rate, the three-dimensional flash size (Bruning & Thomas, 2015) has been recognized to be relevant to the production of NO_x, indicating lightning-NO_x parameterizations could include flash size as well as flash rate.

The amount of NO produced per flash has been estimated in a number of previous field campaigns, model studies, and theoretical analyses resulting in an order of magnitude range (32- to 664-mol NO_x per flash) for lightning-NO_x production per flash (Schumann & Huntrieser, 2007). A compilation of these previous studies shows higher lightning-NO_x production per flash in midlatitude storms compared to tropical storms (Pickering et al., 2016), possibly due to vertical wind shear affecting flash length (Huntrieser et al., 2008). To try to reduce uncertainty in the estimate of lightning-NO_x production, an analysis of Colorado and Oklahoma severe convection found lightning-NO_x production to range from 142- to 291-mol NO_x per flash (Pollack et al., 2016) and showed no preference to intracloud or cloud-to-ground lightning flashes but did show a small positive correlation with flash size and with 0- to 6-km altitude wind shear. However, analysis of an air-mass (prefrontal) convective storm in Alabama found the majority of lightning-NO_x production came from cloud-to-ground flashes and that flash rate correlated better with lightning-NO_x production than flash extent (Carey et al., 2016). The observation-modeling analysis of the Alabama storm suggested a much higher lightning-NO_x production (1,000-mol NO per flash) in cloud-to-ground flashes than in intracloud flashes (100-mol NO per flash).

These two DC3 studies based their results on data collected near the storm cores (<1-hr travel time from core to aircraft sample). However, estimating lightning-NO_x production based on aircraft sampling further downwind can be problematic. Nault et al. (2017) conducted a chemical analysis of organic nitrate compounds in convective outflow regions finding, when chemical data on nitrate formation rates are updated, a much shorter NO_x chemical lifetime (~3 hr) near the active convection than further downwind (0.5- to 1.5-day lifetime). They concluded that lightning-NO_x production may be greater than estimates determined from convective outflow plumes >6 hr away from active convection, suggesting 500-mol NO per flash is an optimal NO_x production emission estimate in the Goddard Earth Observing System with chemistry (GEOS-chem) model.

The vertical distribution of lightning-NO_x generation is important for the subsequent chemistry producing ozone. DeCaria et al. (2005) proposed a midlevel (at $T = -15$ °C) peak distribution for cloud-to-ground flashes and a bimodal distribution for intracloud flashes. Analyses of storms in northern Alabama revealed that vertical distributions of intracloud flashes did not have the expected bimodal initiation distribution and that hybrid flashes (i.e., flashes with both intracloud and cloud-to-ground components) were consistently larger in size than intracloud or cloud-to-ground flashes (Mecikalski & Carey, 2017, 2018a). Mecikalski et al. (2017) and Mecikalski and Carey (2018b) found that these three types of flashes do not have the same parent distribution and therefore should be treated separately in lightning-NO_x parameterizations.

Ott et al. (2010) noted that the vertical distribution of lightning-NO_x differed in midlatitude and subtropical storms, but little research has been done to examine the effect of the storm environment on lightning-NO_x production. Inverted (anomalous) and normal polarity thunderstorms occur in different storm environments, offering a means to explore the impact of storm environment on lightning-NO_x production. Using kinematic data from radar observations and lightning flash channel source observations, Davis et al.

(2019) examined the lightning-NO_x production and advection in normal polarity and inverted polarity storms. The combination of stronger vertical motions and higher flash rates in inverted storms compared to normal storms led to markedly higher lightning-NO_x concentrations in the upper troposphere, despite the fact that lightning channels in inverted storms are located considerably lower in the troposphere relative to normal polarity storms. Even though inverted storms occur less frequently compared to normal storms, they may have a disproportionate impact on upper tropospheric NO_x. An analysis by Fuchs and Rutledge (2018) of the location of lightning channels in normal and inverted storms (examining hundreds of individual storms) showed in general that flash channels are indeed at lower altitudes in inverted storms relative to normal storms by ~2 km.

These studies reveal that there are still major uncertainties in estimating the production of lightning-NO_x and in representing this process in numerical models. The new data on lightning size and flash currents coupled with novel analysis techniques may further reduce these uncertainties.

2.3. Lightning Characteristics

LMAs (Rison et al., 1999) have significantly advanced the field of atmospheric electricity with respect to quantifying types of lightning flashes, flash initiation regions, and sizes of lightning flashes, especially flash area and channel length. LMAs were already established in northern Alabama and central Oklahoma before the DC3 field campaign. New arrays were installed in northeastern Colorado, west Texas, and southwest Oklahoma preceding the DC3 campaign. The performance of the LMA location and detection efficiency of lightning was found to depend on station configuration but generally showed >95% detection efficiency within 100 km of the LMA network (Chmielewski & Bruning, 2016). A new analysis technique based on the moments of the flash area distribution has allowed a reliable way to estimate lightning channel length, flash energy, and flash rate (Bruning & Thomas, 2015). Using these parameters, it has been shown that flash size and flash rate are inversely correlated (Barth et al., 2015; Mecikalski et al., 2015).

Lightning and storm kinematic and microphysical properties are closely linked. Analysis of the 29 May 2012 Oklahoma severe convection found that the kinematic and microphysical features of the storm were consistent with the noninductive charge mechanism for triggering lightning (DiGangi et al., 2016), which is thought to be the main mechanism for cloud electrification.

Storms with inverted or anomalous polarity charge structure tend to have positive cloud-to-ground flashes (e.g., Lang et al., 2004), which have stronger electric fields, higher peak charge, longer flash duration, and potentially cause greater damage than negative cloud-to-ground flashes. The DC3 project provided data to investigate the processes that create anomalous polarity thunderstorms by examining the links between environment, microphysics, kinematics, and charge structure. Using LMA and radar observations, comparisons of normal and inverted polarity storms were obtained. Fuchs et al. (2018) found that anomalous polarity storms in Colorado have stronger and wider updrafts and increased supercooled liquid water content in the mixed phase region of the storm, which promotes positive charge on graupel and thus the inverted charge structure characteristic of this storm type. Normal polarity storms tended to have weaker and narrower updrafts, reducing supercooled water contents and thereby promoting negatively charged graupel. A key finding in Fuchs et al. (2018) is that anomalous versus normal storms are often differentiated by the depth of the warm cloud layer. Anomalous storms formed in environments that promote shallow warm cloud depths, which reduce the time available to form raindrops via coalescence. Instead, condensate is transported to the mixed phase region leading to high supercooled liquid water content. The actual mechanism that leads to positive charge on riming graupel particles is far from being clearly understood however.

2.4. Chemistry in Convective Outflows

In the early 1990s, it was hypothesized that convective outflow regions would be conducive to ozone production. Modeling studies estimated ozone formation of 4–15 ppbv/day for midlatitude convection (Pickering et al., 1990). Measurements of high ozone in the summertime, upper troposphere were attributed to convective outflow regions over North America (Bertram et al., 2007; Cooper et al., 2006, 2007, 2009). However, these previous studies did not gather direct evidence of ozone production in convective outflow regions. During DC3, scientists observed high ozone concentrations in convective outflow regions in the upper troposphere. Apel et al. (2015) estimated ozone production in the 22 June 2012 Colorado convective outflow regions of two, side-by-side severe storms, one with and one without wildfire smoke (the smoke contains much higher concentrations of volatile organic compounds than the air from the planetary boundary layer). Their calculations suggested that 10–15 ppbv of ozone would be produced over 2 days as calculated by a photochemical model and underscored the observation that more lightning-generated NO_x creates more ozone. The 21 June 2012 DC3 case of a decaying MCS provided measurements of the chemical composition of the dissipating anvil region and convective outflow of the MCS over an 11-hr period during daytime. During the morning period (the first 5 hr of measurements), ozone increased by ~12 ppbv (Brune et al., 2018), consistent with observed HO_2 and box model calculations, and another 8 ppbv (20 ppbv in total) by midafternoon (Barth et al., 2015). Results from these two case studies are in the upper end of the range reported by previous studies. The combination of MCSs being strongest at night and dissipating during the day with the sunlight to fuel the photochemistry is hypothesized to be the most direct way to quantify the chemistry associated with ozone formation in convective outflow regions.

2.5. Stratosphere-Troposphere Exchange

Sampling cloud-scale stratosphere-troposphere exchange caused by thunderstorm dynamics was somewhat unexpected. Measurements found stratospheric ozone mixing into storm anvils (Huntrieser et al., 2016b; Schroeder et al., 2014) and even wrapping around the anvil in one case (Pan et al., 2014), bringing high ozone concentrations into the upper troposphere. Analysis of aged convective outflow air revealed enhanced ozone mixing ratios by 20–50 ppbv mainly from dynamical processes (Huntrieser et al., 2016a). Although we expect ozone to be formed in the upper troposphere convective outflow plumes, transport of ozone from the stratosphere also contributes to increased upper troposphere ozone mixing ratios, and this dynamical process needs to be better represented in chemistry transport models.

In addition to stratospheric air being mixed into the upper troposphere, some of the DC3 storms were shown to transport water vapor well into the stratosphere (Homeyer et al., 2014), where water has important impacts on radiation and climate. Analysis of these storms has led to the hypothesis that weakened stability associated with double-tropopause environments facilitates convective injection deep into the stratosphere.

2.6. Tropospheric Composition

Besides direct sampling of storm inflow and outflow regions and aged convective outflow, the DC3 aircraft frequently sampled background free troposphere air. Airborne measurements of aerosol concentrations, optical properties, and hygroscopicity were taken during DC3, providing further insights into their distributions and impact on radiation and clouds. Measurements of brown carbon showed that it is prevalent throughout the troposphere with increasing prevalence relative to black carbon with increasing altitude (Liu et al., 2014). Because of its light-absorbing properties, brown carbon is important to the atmosphere's radiation budget. Analysis of the brown carbon optical properties suggest that brown carbon contributes to 20% of top-of-atmosphere aerosol forcing (Liu et al., 2014). Black carbon, also important to atmosphere radiation as an absorbing aerosol, was also measured providing observations to constrain global model data. Schwarz et al. (2017) found that global models participating in the AEROCOM model intercomparison simulated black carbon mixing ratios that compared well to observations in the lowest 5 km of the troposphere but overpredicted black carbon in the upper troposphere. K. Heimerl, H. Huntrieser, J. P. Schwarz, A. Minikin, R. Baumann, M. A. Fenn, D. Fütterer, J. W. Hair, H. Schlager, and B. Weinzierl suggest that this discrepancy in upper troposphere black carbon mixing ratios may be due to partial wet removal in convection (Transport and Washout of Refractory Black Carbon in North American Biomass Burning Layers in the Free Troposphere, submitted to *J. Geophys. Res.*).

Other aerosol optical properties (e.g., extinction coefficient, single scattering albedo, and asymmetry factor) depend upon the refractive index of aerosols. The real part of the refractive index was characterized over North America to have an average value of 1.52, typical for organic aerosols (Aldhaif et al., 2018). These measurements were found to correlate positively with the O:C ratio in air that was unaffected by biomass burning. Generally, the refractive index was found to be lower at altitudes >8 km compared to altitudes <4 km in nonstorm conditions (Sorooshian et al., 2017), although the chemical composition of the aerosols did not consistently change between these altitude regions, suggesting other factors (like water) affect the refractive index. These authors also found that hygroscopicity of the aerosols was enhanced in convective outflow regions.

2.7. Cloud Structure

Analyses of cloud physical properties were conducted using aircraft measurements of cloud particle size distributions (Stith et al., 2016) and water vapor (Diao et al., 2017), as well as balloon-borne video disdrometer (Waugh et al., 2018). The recently developed balloon-borne microphysics probe can provide observations at fine scales and give calculated cloud particle mixing ratios and reflectivity in agreement with radar and cloud-resolving, kinematic model results.

Analyses of cloud particles revealed the presence of frozen drop aggregates accompanied by high NO_x concentrations in the cores of convectively generated anvils. On the edges of anvils, more individual frozen droplets are found along with small ice crystals and lower NO_x concentrations (Stith et al., 2014). The frozen drop aggregates are formed by electrical forces associated with high electric fields accompanying lightning. Since lightning is also the source of lightning-NO_x, these studies suggested that electrically active storms may have characteristically different ice particle types in the anvil compared to less electrically active storms, which may have an impact on the radiative impacts of the anvil clouds (Stith et al., 2016).

Another critical process with impacts on atmospheric radiation is ice nucleation. Improving model parameterizations of ice nucleation is a high priority for climate studies. High-resolution modeling at $\Delta x = 0.25$ km simulated well the ice supersaturated regions and ice nucleation when the threshold for RHice was set to 130% (Diao et al., 2017), providing guidance for better parameterizations in weather and climate models. Simulations of the 22 June 2012 Colorado severe storms using WRF-Chem at $\Delta x = 1$ km found that storms with cold cloud bases and therefore small contributions of the warm-rain processes to precipitation caused enhanced heterogeneous ice nucleation compared to the homogeneous ice nucleation process. When heterogeneous ice nucleation was enhanced, the storm anvils were optically thinner (Takeishi & Storelvmo, 2018).

3. Summary

The DC3 project brought together scientists from different subcommunities of atmospheric science to understand more holistically the cloud physics, electricity, and dynamics of thunderstorms and their impact on tropospheric composition. The success of the field campaign was rooted in the centralized operations base utilizing internet-based communications among the aircraft and ground-based crews, which allowed for real-time adjustments in deployment activities. A crucial aspect of DC3 operations was the daily weather forecasts by a team of forecasters composed of a lead forecaster at the operations base and regional forecasters at each DC3 focused region. Being tested during DC3 field operations were probabilistic forecasts and automated decision algorithms to aid in field campaign decisions. Hanlon, Small, et al. (2014) found that automated decision algorithms would have increased the yield of data from the field campaign, while Hanlon, Young, et al. (2014) showed that their probabilistic forecasts had predictive skill over climatology for each of the DC3 regions. Another pivotal reason for the success of DC3 was the strong cooperation and interaction between the lead PIs for the project.

Often, field experiments provide a venue for new measurements as well as educational experiences. DC3 was no exception. There are several papers published describing new instrumentation deployed during DC3 as well as undergraduate education activities.

With the new knowledge gained from the DC3 science analysis, new questions naturally arise and new opportunities are envisioned. The scavenging of trace gases has been found to be a complex topic complicated by the role of the cloud physics. Determining whether the scavenging efficiencies found in the severe convection sampled during DC3 are applicable to different types of storms, from weak convection to tropical convection to synoptic storms, is an important next step. Uncertainties in the scavenging calculations can be reduced by gathering more data at the top of the updraft of storms as well as conducting chemical analysis of rain and cloud drop and ice samples.

The production of NO_x from lightning is also a complex topic because of the need to combine lightning characteristics and cloud physics and dynamics to determine NO_x production. While DC3 provided more information on the process, especially in the context of total flash rates, further analysis in terms of other flash characteristics still need to be pursued. In addition, gathering NO_x measurements in thunderstorm cores near the lightning flashes would give more direct information on the lightning-NO_x production.

DC3 provided sampling in both near-storm and downwind convective outflow regions to determine ozone formation downwind of convection. It was learned that MCSs, which typically are strongest at night over the central United States and dissipate during the day, provide an excellent region to probe ozone formation in convective outflow. Sampling of several MCS convective outflow regions would provide a more robust data set for documenting ozone production as well as new particle formation.

Finally, contrasting the influence of thunderstorms in midlatitude regions like the United States with other continents (e.g., Asian and West African monsoons and South America) and oceanic regions should provide a wealth of knowledge on the processes that control atmospheric trace gas and aerosol composition near and downwind of storms.

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