Deep Convective Clouds and Chemistry (DC3) Experiment

When: Summer 2009 Where: Central U.S.
Major Facilities: Aircraft (high altitude and low altitude planes),
Radar (Doppler and polarimetric), Lightning Mapping Array

Background:
Convective transport is a major pathway of rapidly moving chemical constituents (including water) from the boundary layer to the upper troposphere (UT) and in some cases to the lower stratosphere (LS). Yet the global-scale impact of convective transport on the UTLS composition and chemistry has not been characterized. Dissolution and aqueous reactions of chemical species are an effective way of cleansing the atmosphere through wet deposition. Often associated with deep convection, lightning produces NOx, an essential precursor of ozone. The higher NOx concentrations accelerate hydrocarbon oxidation, HOx production, and O_3 production. Because of these multiple processes, quantifying the convective processing of NOx, HOx, its precursors, and volatile organic compounds (VOCs) is important to determining the impact of convection on the UTLS composition and photochemistry.

Understanding the photochemistry in the UTLS and the influence of convection on the UTLS composition and photochemistry are also critical to predicting ozone trends, which ultimately impact the earth's radiation budget, the UV flux to the surface, and the production of radical species that are responsible for removal of primary pollutants. In the upper troposphere, chemistry involving HOx and NOx radicals in the presence of VOCs generally results in net ozone production. In the lower stratosphere, catalytic chemical cycles involving radical species (HOx and/or various halogen species, such as chlorine and bromine oxides) result in net ozone destruction.

Interactions between cloud microphysics, chemistry and aerosols are important factors in understanding the impact of convection on UTLS composition and chemistry. The cloud microphysics, in particular the interactions between water and ice particles, affects the fate of chemical species dissolved in the drops. Thus, measurements of soluble species in convective inflow and outflow will elucidate the role of microphysics on chemical species distributions. Additionally, adsorption of species, such as nitric acid, onto ice is another important mechanism for redistributing species in the troposphere.

Entrainment of air into the cloud core and anvil dilutes the water content of the cloud and reduces the buoyancy, thereby affecting the dynamics and kinematics of the cloud. It also brings into the cloud aerosols and gases that are of different composition, concentration, and origin than the air that ascends from the boundary layer. This entrained air can affect local thermodynamics as well as chemical and microphysical processes.

There is ample evidence that convection perturbs the chemical composition in the upper troposphere. However, only a few studies have attempted to examine the detailed dynamics of deep convection and the concomitant redistribution, production, or removal of reactive constituents. Experiments, such as STERAO and EULINOX, included measurements of cloud structure, lightning flashes, and a suite of chemical species. They provided some of the first detailed in situ observations of NOx production and related it to measurements of lightning activity and numerical cloud models that simulated NOx production and convective transport. However, neither of these experiments included measurements of HOx precursors (e.g. peroxides, carbonyls, and hydrocarbons) in both the inflow and outflow of the storms. Experiments, such as INTEX and TRACE-P, obtained measurements of HOx and its precursors in convective outflow regions, but did not have measurements of these species in the inflow regions of the storms, nor did they include measurements of cloud structure and kinematics, or lightning, which connect the chemical measurements in the convective outflow to specific cloud processes. Hence, the incomplete data from these field programs do not allow us to establish a complete description of convective cloud processing on HOx and its precursors and on NOx.

The experiment proposed here will obtain measurements of enough chemical species to characterize the effects of convection on the transport and transformation of ozone and its precursors. For example, HOx species, its precursors, and NOx species in both inflow and outflow regions of deep convection will be measured along with microphysical properties, storm kinematics, and lightning discharges. These measurements are planned for over the Great Plains of the United States, where we hope to contrast regions of remote continental air to those more influenced by anthropogenic emissions.

Primary Goals:

1. *To quantify the impact of continental, midlatitude convective storm dynamics, multiphase chemistry, lightning, and physics on the transport of chemical constituents to the upper troposphere,*

Candidate Hypotheses:

a) The transport efficiency of inert tracers to the upper troposphere varies depending on the type of convection (e.g., multicell, supercell) which is affected by characteristics of the environment, such as convective available potential energy (CAPE) and wind shear. Soluble species are transported less efficiently compared to inert species because of scavenging and multiphase chemical reactions which depend on the relative amounts of ice and liquid water in the storm.

b) The amount of NOx produced from lightning correlates with the length and intensity of the lightning flash with most of the NOx produced at heights extending from just above the core liquid water region to near the top of the anvil. On average an intracloud flash produces an approximately equivalent amount of NOx as that produced on average by a cloud to ground flash. NOx production in cloud to ground flashes correlates with peak current.

2. *To determine the role of anvil dynamics, multiphase chemistry, microphysics, radiation, and electrification on the chemical composition of convective outflow,*

Candidate Hypothesis:

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, a lower layer with low radiation fluxes and nighttime-like radical chemistry, and ice crystals containing significant amounts of soluble species that have been entrained or produced from photochemistry in the anvil. This will result in nitric acid and other adsorbed species being brought to the mid-troposphere by the fallout of ice and sublimation. NOx concentrations will be elevated by production from lightning and the degree and vertical profile of enhancement will depend on the frequency and location of the lightning in the storm and the storm dynamics and structure.

3. *To determine the effects of convectively-perturbed air masses on ozone and its related chemistry in the midlatitude upper troposphere and lower stratosphere 12-48 hours after the near convection region is sampled,*

Candidate Hypothesis:

In sampling the convective plume 12-48 hours after convection that occurred over a relatively unpolluted region, we expect to find that 10-15 ppbv ozone will be produced due to high NOx and enhanced concentrations of radical reservoir species. Less ozone will be produced in convective plumes from storms occurring over more heavily polluted regions because of a lower ozone production efficiency associated with larger anvil mixing ratios of NOx. That is, determine the amount of O_3 produced and contrast O_3 production from storms in clean regions versus storms from more polluted regions.

4. *To contrast the influence of different surface emission rates on the composition of convective outflow.*

Candidate Hypothesis:
Convection that ingests relatively unpolluted air will have convective outflow regions with less HOx precursors but approximately the same NOx as convection that ingests more polluted air. Consequently ozone production will be more efficient in the relatively unpolluted convective outflow.

Other objectives benefiting from the DC3 field experiment are:

1. *To determine partitioning of reactive halogen and reservoir species in the UTLS*
2. *To determine the mass fluxes of air and trace gases into and out of the storm, including entrainment (determine fraction of boundary layer air that reaches LS, UT; determine fraction entrained; determine what part of the boundary layer is ingested by the storm; determine quantity of stratospheric ozone entrained into anvil)*
3. *To improve our understanding of cloud electrification and lightning discharge processes*
4. *To investigate the role of deep convection in contributing to the UT water vapor and in the transport of water vapor into the lowermost stratosphere*
5. *To connect aerosol and cloud droplet and ice particle number concentrations with convection characteristics and trace gas convective processing*

Experimental Strategy:

- Use a low to mid-troposphere aircraft to measure in-situ HOx and HOx reservoir species and NOy species in the inflow regions (both below cloud base and in regions entraining air into the storm) of midlatitude deep convection and to support an upward-looking LIDAR.
- Use high altitude aircraft (including HIAPER) to measure radical species, their precursors, nitrogen oxides, passive tracers, and cloud particles in the outflow region of deep convection.
- Conduct measurements in the convective outflow plume as it advects downwind. Measurements of one plume may occur at plume ages of several hours to a day.
- Perform extensive airborne comparison (both during and before the proposed mission) of common measurements on board the aircraft.
- Use ground-based radars and lightning interferometers, an NO_2 spectrometer, rain collectors, and meteorological soundings to obtain information on cloud structure, lightning flash rate, length, and location, precipitation amount and composition, and the environmental state of the atmosphere. Quantifying the storm dynamics, physics, and electrical activity is crucial to interpreting the chemistry measurements.
- Make use of available satellite data, e.g. from the AURA satellite, for field experiment planning.
- Use regional and global-scale models to forecast convective events and to guide flights.
- Simulate the chemistry in and around the deep convection via cloud resolving models, regional-scale models, and global models.
- Conduct studies over the relatively clean lower troposphere, e.g. the upper Great Plains, and in more polluted regions near urban centers. Sample a variety of types of storms.

Critical Measurements:

Airborne:

HIAPER – remote and in-situ measurements of radicals, their precursors, NOx, and tracers, and other related measurements (e.g. state parameters, actinic flux, and cloud particle concentrations), in-situ turbulence, cloud ice and liquid particles

Low-altitude aircraft – state parameters, remote and in-situ measurements of HOx, its precursors, NOx, and tracers, wind field via cloud radar (ELDORA or Doppler radar on P3), aerosol size, concentration, and chemical composition, CCN, liquid and solid sampling of precipitation.

Ground-based:

Radar – reflectivity and polarized to determine hydrometeor type

Lidar –aerosol backscatter, O_3 DIAL

Lightning interferometers –flash rate and length; 3D location of lightning discharges (combine with cooperation of NLDN network)

Rain collectors – amount of precipitation, soluble species and ions, e.g. CH_2O, H_2O_2, CH_3OOH, HNO_3, SO_4, organic acids (HCOOH, dicarboxylic acids)

Soundings

Meteorological soundings

Ozone soundings

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