The Effects of Deep Convection on Atmospheric Chemistry

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Effects of Deep Convection

Convection over "Polluted Regions"

- Venting of boundary layer pollution
- Transport of NO_x, NMHCs, CO, and HO_x precursors to the upper troposphere (UT) and sometimes to the lower stratosphere (LS), where chemical lifetimes are longer and wind speeds greater
- Downward transport of cleaner air to PBL
- Transported pollutants allow efficient ozone production in UT, resulting in enhanced UT ozone over broad regions

NO + HO₂
$$\rightarrow$$
 NO₂ + OH
NO₂ + hv \rightarrow NO + O*
O₂ + O* + M \rightarrow O₃ + M

- Increased potential for intercontinental transport
- Enhanced radiative forcing by ozone

Effects of Deep Convection

Convection over "Clean" Regions

- In remote regions low values of PBL O_3 and NO_x are transported to the upper troposphere
- Potential for decreased ozone production in UT
- Larger values of these species tranported downward to PBL where they can more readily be destroyed

Convection over all Regions

- Lightning production of NO (much more over land)
- Perturbation of photolysis rates
- Effective wet scavenging of soluble species
- Nucleation of particles in convective outflow

Observations and Models

- Combination of observations and model simulations is a powerful tool to better understand physical and chemical processes in thunderstorms
- Convection/chemistry field experiments (the last 25 years):

PRESTORM – OK, KS 1985

ABLE-2A – Brazil 1985

ABLE-2B – Brazil 1987

STEP – Australia 1987

NDTE – North Dakota 1989

TRACE-A – Brazil 1992

STERAO – Colorado 1996

EULINOX – Germany 1998

CRYSTAL-FACE – Florida 2002

TROCCINOX – Brazil 2005

SCOUT-O3/ACTIVE – Australia 2005

AMMA – West Africa 2006

TC4 – Costa Rica 2007

Observations and Models

Cloud-resolved chemistry models:

- Storm simulation with Goddard Cumulus Ensemble (GCE) Model or cloud-resolved MM5 drives offline transport and chemistry model with lightning NO production.
- Cloud-resolved WRF-Chem (online transport/chemistry) now being used.

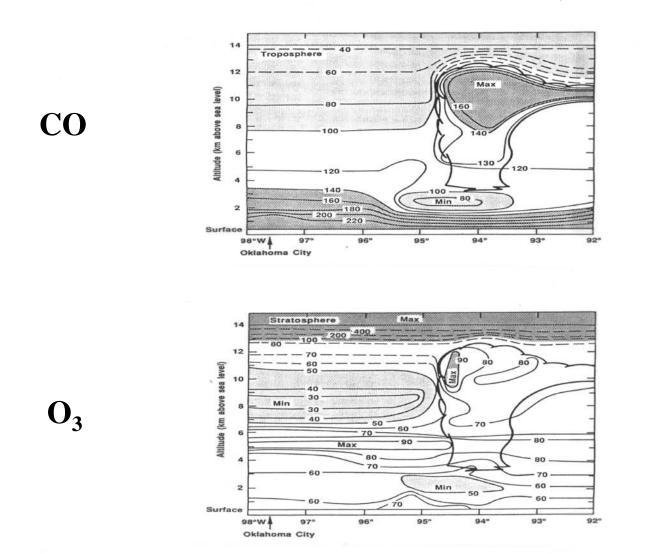
• Regional chemistry models:

- Driven by WRF with parameterized convection (examples: offline CMAQ; on-line WRF-Chem). Lightning schemes being developed.
- Global chemical transport models:
 - Offline global chemistry and transport in UMD-CTM and NASA/GMI CTM driven by GEOS-DAS from Goddard GMAO. Lightning parameterized with model convective mass fluxes and constrained with satellite observations.
 - Online chemistry and transport in GEOS-5 Chemistry and Climate Model, allowing chemistry to feedback to meteorology through perturbations to radiative fluxes. Physically-based lightning scheme under development.

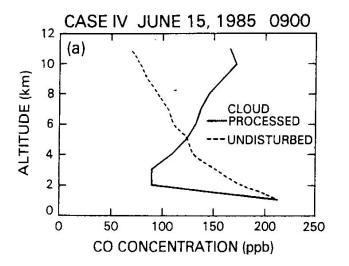
Midlatitude Convection

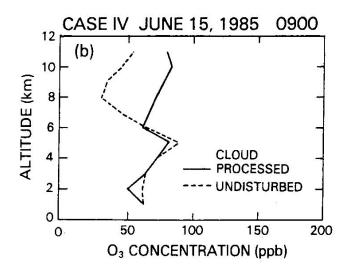
Examples from field experiments and models

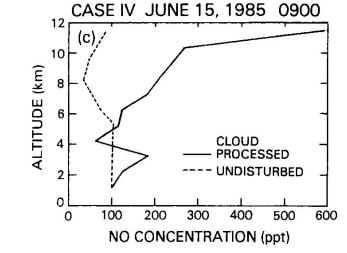
Aircraft Measurements of Trace Gas Redistribution in Oklahoma PRESTORM June 15, 1985 MCC



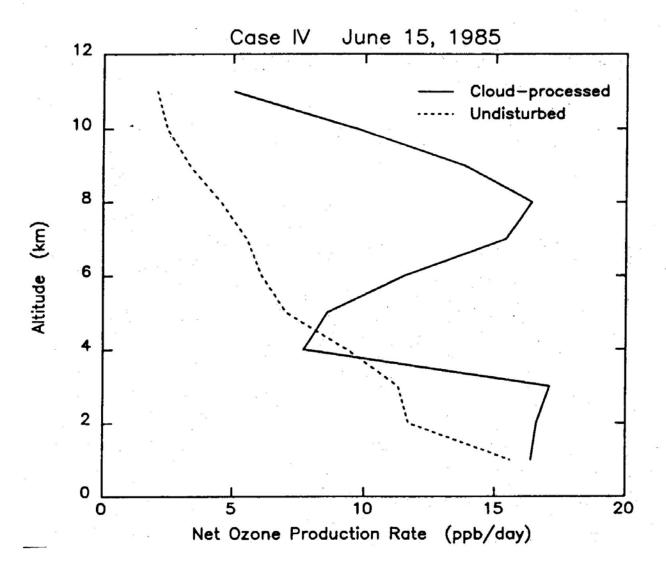
Dickerson et al., 1987, Science







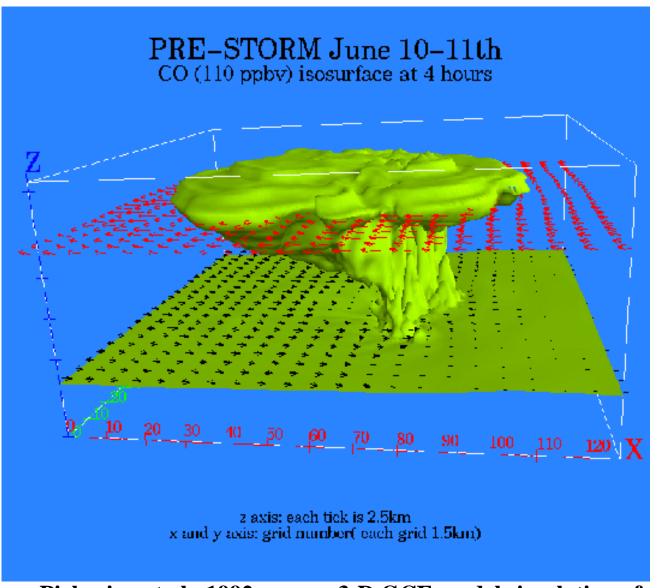
Pickering et al., 1990



Mid – upper trop. ozone production enhanced by factor of 4

Pickering et al., 1990

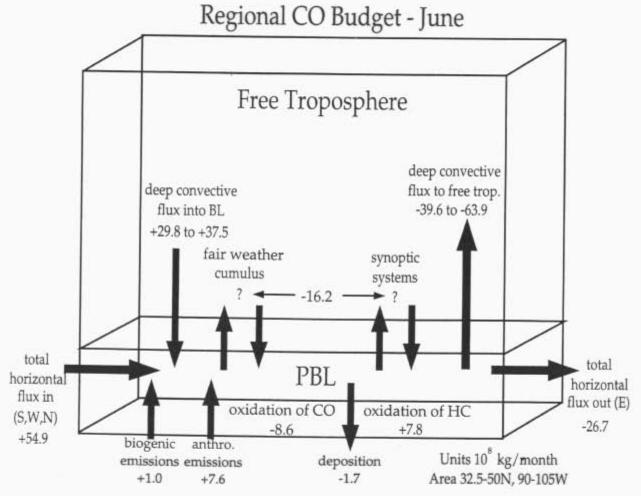
Kansas-Oklahoma Squall Line Cell



Pickering et al., 1992

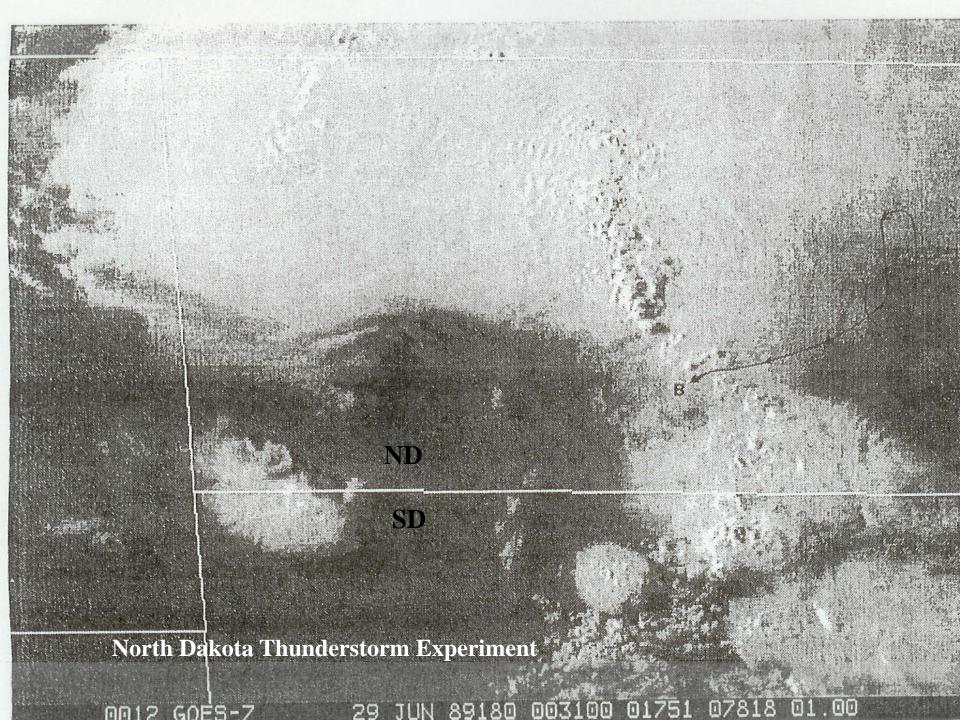
3-D GCE model simulation of one squall line cell

Central United States

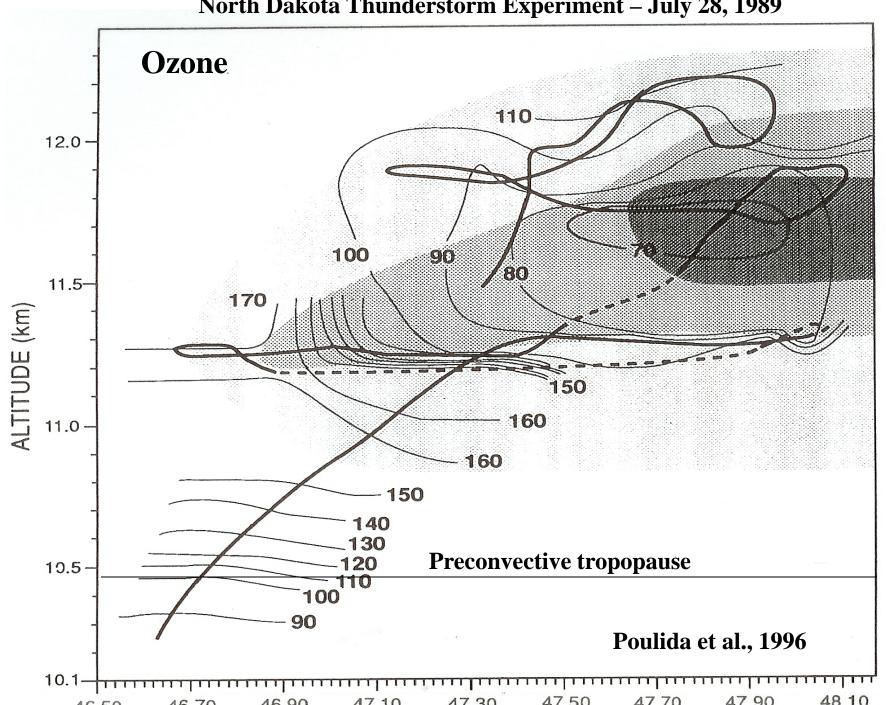


Thompson et al., 1994

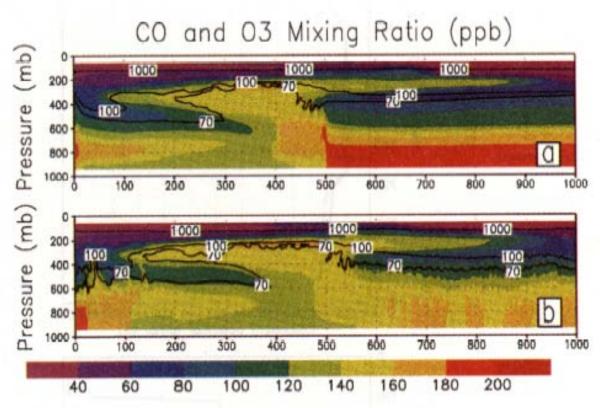
Uses cloud-resolved model transport statistics and ISCCP convective cloud climatology



North Dakota Thunderstorm Experiment – July 28, 1989



CO and O₃ Tracer Simulation for June 28, 1989 NDTP storm



 $CO - color scale; O_3 - isolines$

(a) base simulation; (b) moist boundary condition simulation

Note downward ozone transport near rear anvil

Stenchikov et al. (1996)

CO and O₃ Tracers Along Anvil Passes for July 10, 1996 STERAO storm

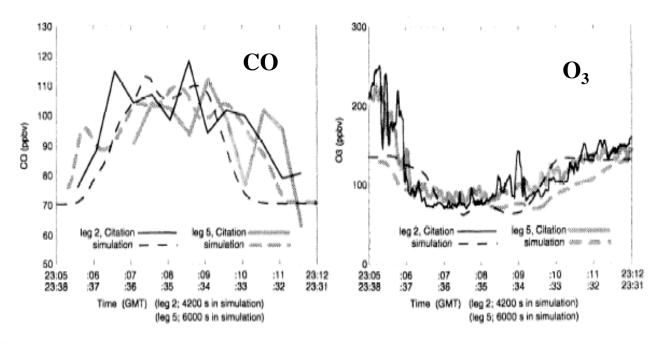


Figure 11. Citation CO and O₃ measurements for anvil passes close to the southeasternmost convective cell (leg 2, 10 km downwind, 11.6 km msl) and downwind (leg 5, 50 km downwind, 11.2 km msl), along with analogous tracks taken through the simulation. The plot tracks are from the southwest (left) to the northeast (right).

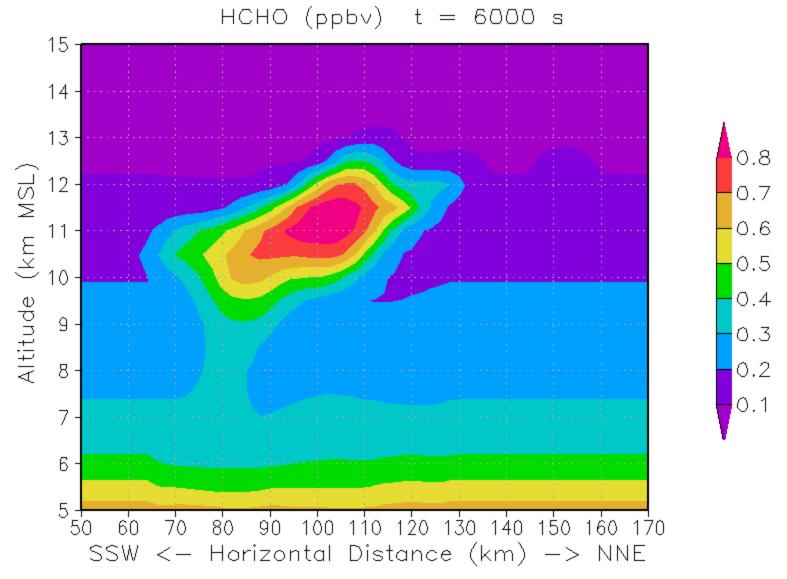
Note enhanced ozone at southwest (upwind) edge of anvil

Skamarock et al. (2000)

Enhanced UT HO_x Production

- Jaeglé et al. (1997) and Prather and Jacob (1997) noted that deep convection is effective in transporting HO_x precursors to the upper troposphere.
- Water vapor, acetone, methylhydroperoxide, and formaldehyde shown to be important as HO_x precursors.
- Enhanced HO_x leads to enhanced O_3 production

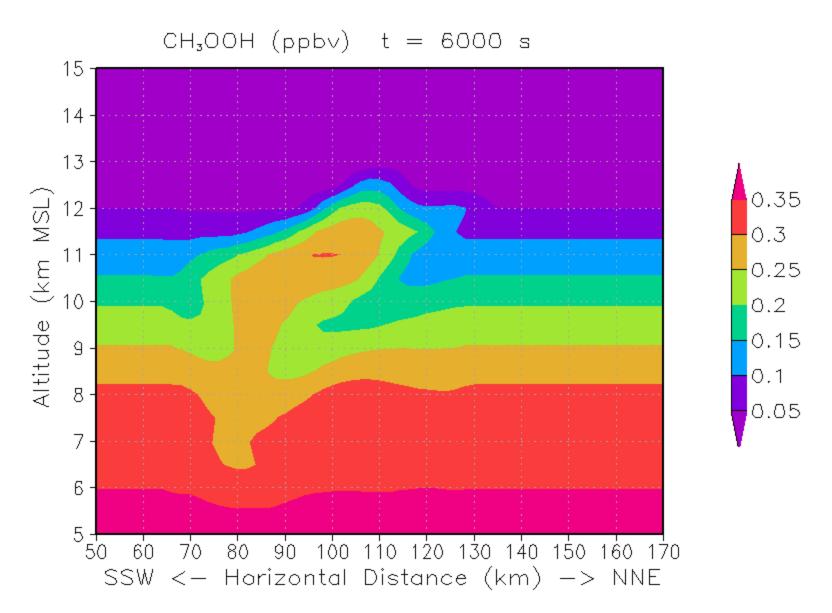
STERAO-A July 10, 1996



Fried et al.(2008) INTEX-A obs showed 46% of samples had HCHO enhanced above background, and 30% of these samples resulted from direct convective injection to the UT

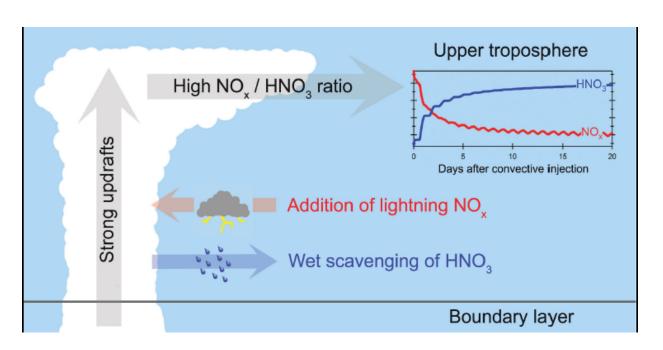
Ott et al. (2006)

STERAO-A July 10, 1996



Ott et al. (2006)

NASA INTEX-A Observations over US – Summer 2004



NO_x/HNO₃ ratio used as a chemical clock to determine time since air was influenced by convection

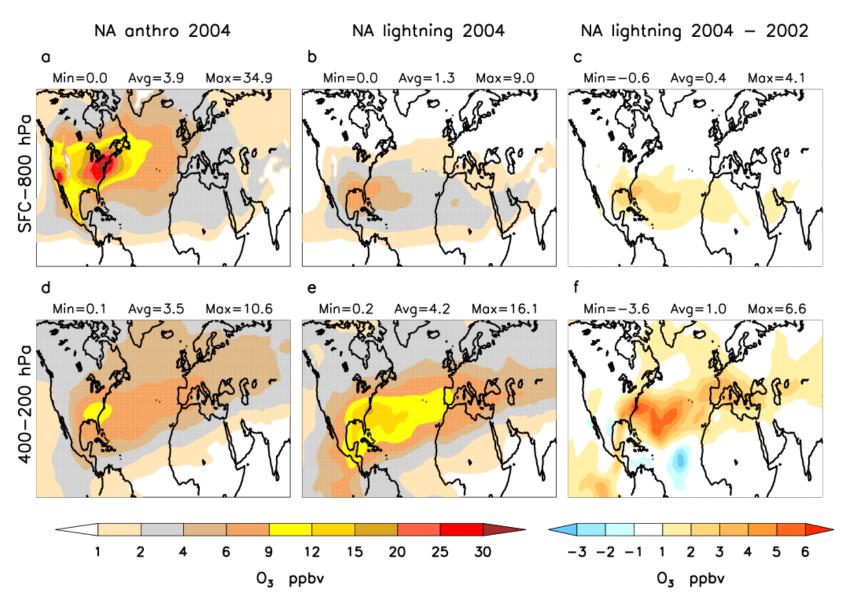
Altitude	Frequency
7.5-8.5 km,	$f_{< 2 \text{ Days}} = 0.43$
8.5-9.5 km,	$f_{< 2 \text{ Days}} = 0.56$
9.5-10.5 km,	$f_{< 2 \text{ Days}} = 0.69$
10.5-11.5 km,	$f_{< 2 \text{ Days}} = 0.43$

0.54

Mean

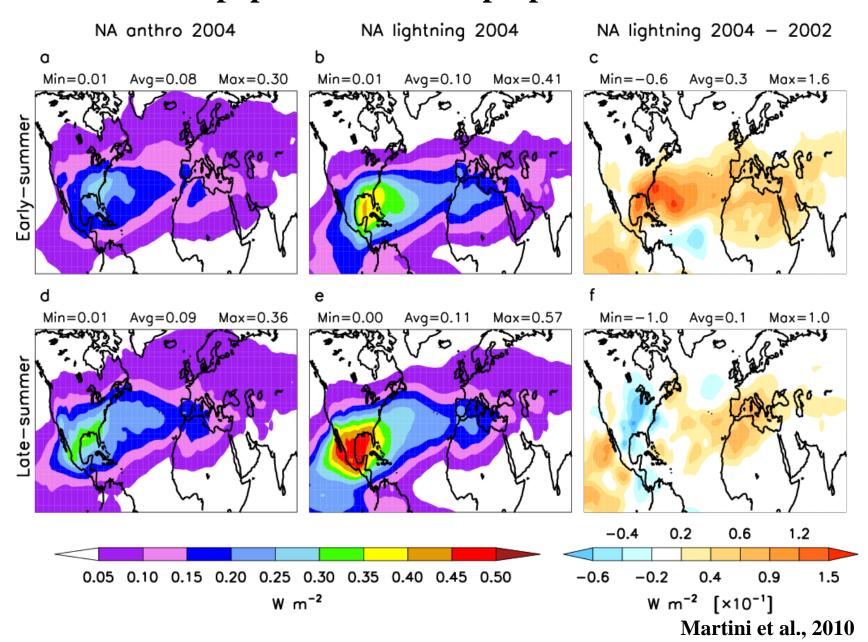
Bertram et al., 2007

Ozone Export from North America – Early Summer

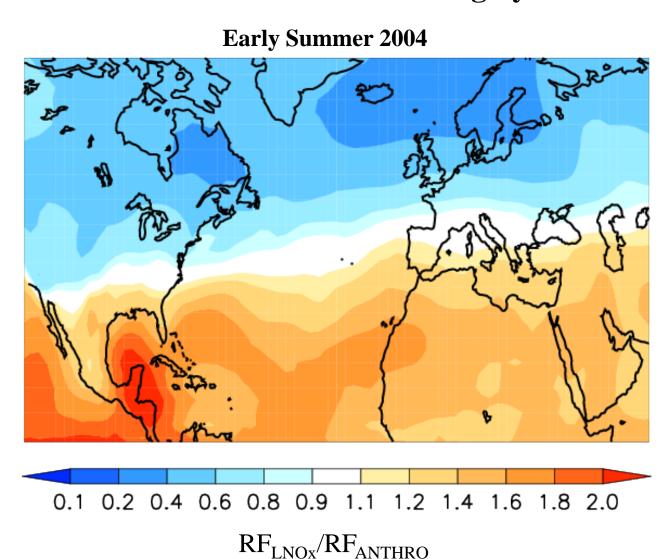


Martini et al., 2010

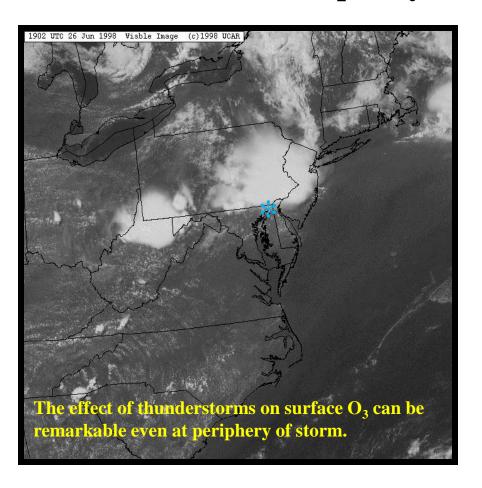
Net Downward LW Radiative Flux Perturbations at the Tropopause Due to Tropospheric Ozone



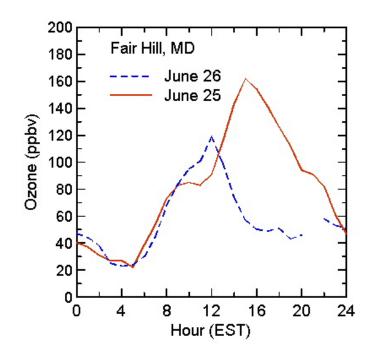
Relative Importance of Anthropogenic and Lightning Emissions on Radiative Forcing by Ozone



Surface air quality effects of deep convection



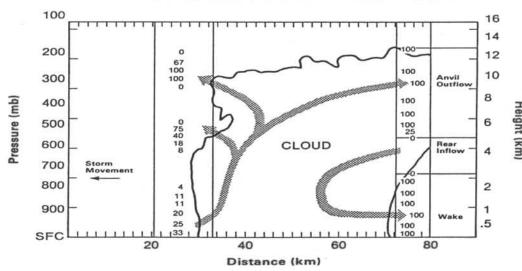
On the third day of a high O_3 episode (June 24-26 1998), a line of thunderstorms passed just north of the Fair Hill, MD monitoring station.



Tropical Convection

AUGUST 3, 1985 ABLE 2A CLOUD MODEL SIMULATION TIME = 240 MIN

Tropical squall line over Amazon Basin

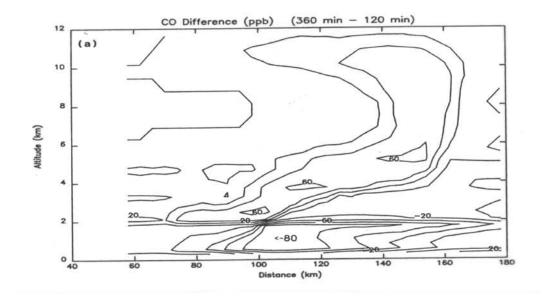


Arrows indicate major transport paths

Columns of numbers indicate percentage of air at these locations that is cloud outflow based on trajectory analysis

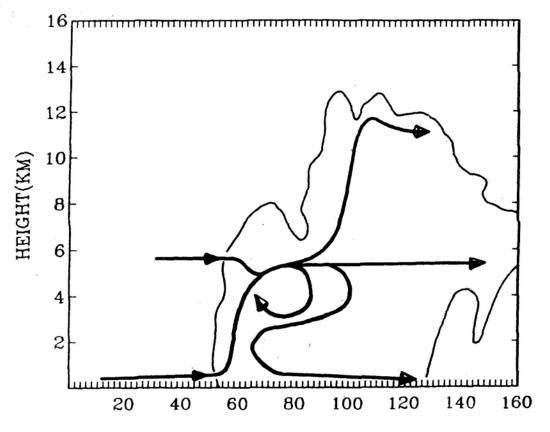
Dry Season

Pickering et al, 1991



CO redistribution from biomass burning plume

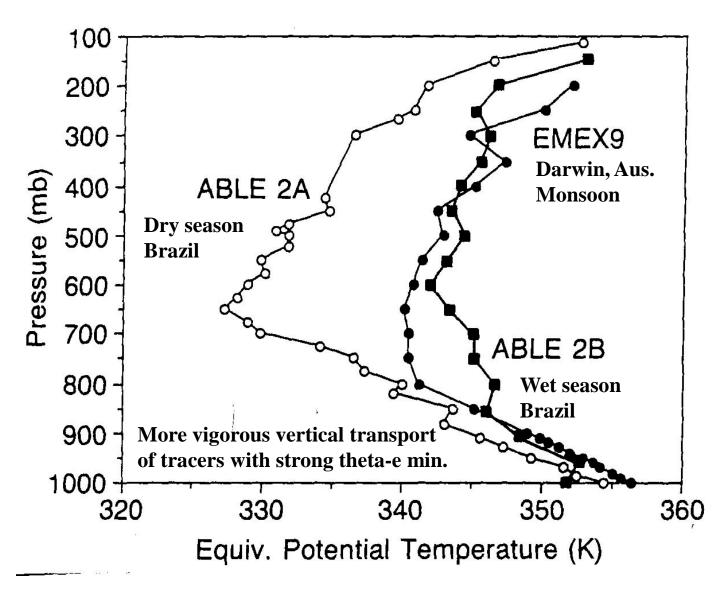
ABLE-2B April 26, 1987 Brazil Squall Line



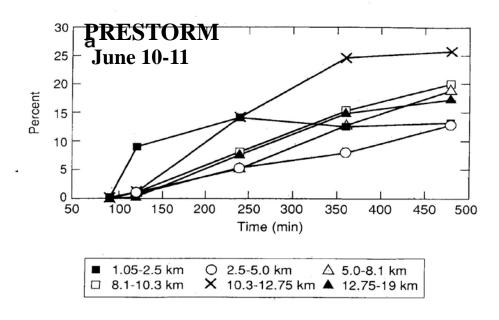
Arrows indicate major transport pathways

Wet Season

Scala et al., 1990



Pickering et al., 1993



26% of BL tracer reaches 10-12 km

Weak vertical transport to upper troposphere due to midlevel overturning

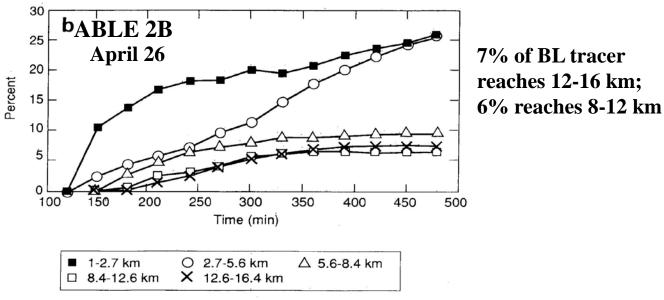


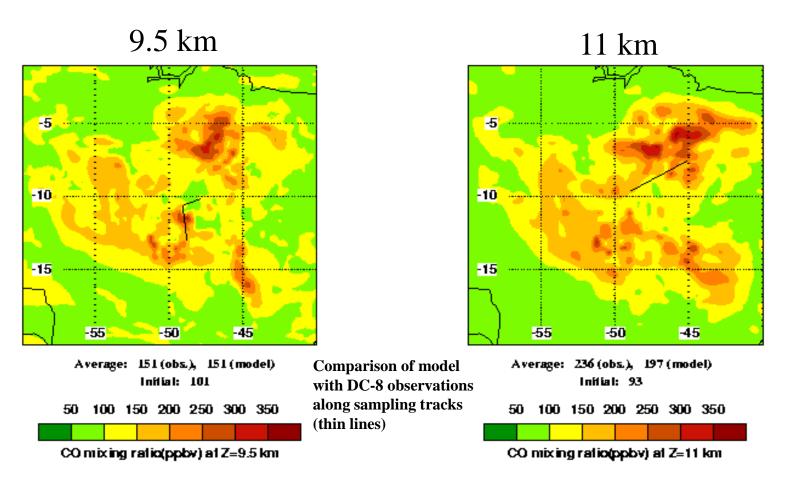
Fig. 6. Fractional redistribution of tracer mass initially in lowest kilometer to other indicated layers for (a) Kansas-Oklahoma PRE-STORM event (June 10–11, 1985) and (b) April 26, 1987, Amazonian ABLE 2B event.

Pickering et al., 1992

Convective Transport of Biomass Burning Emissions over Brazil

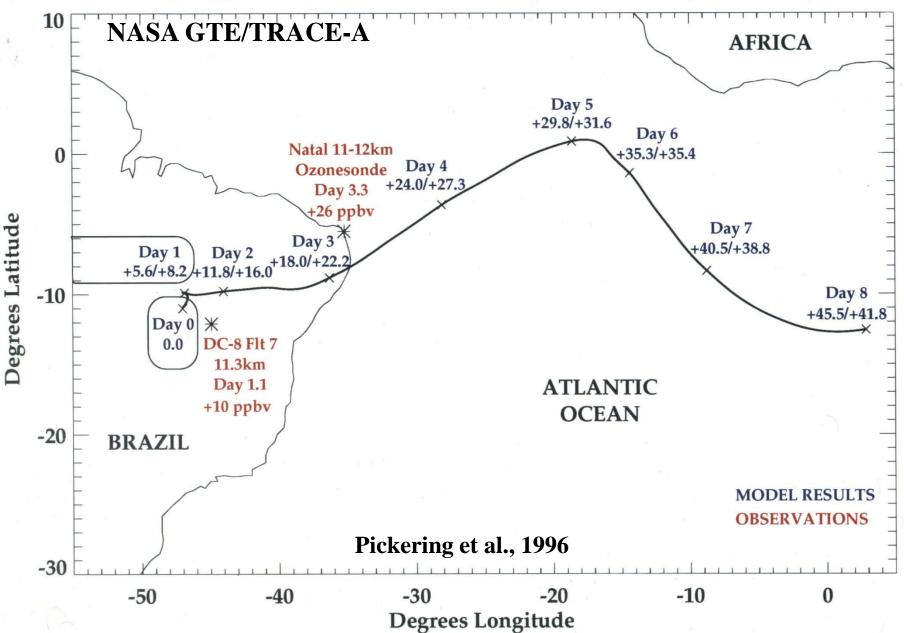
Kain-Fritsch Convective Parameterization MM5 Simulation of System Sampled on GTE/TRACE-A

Positive definite scheme, grid+subgrid transport



Pickering et al., 1996

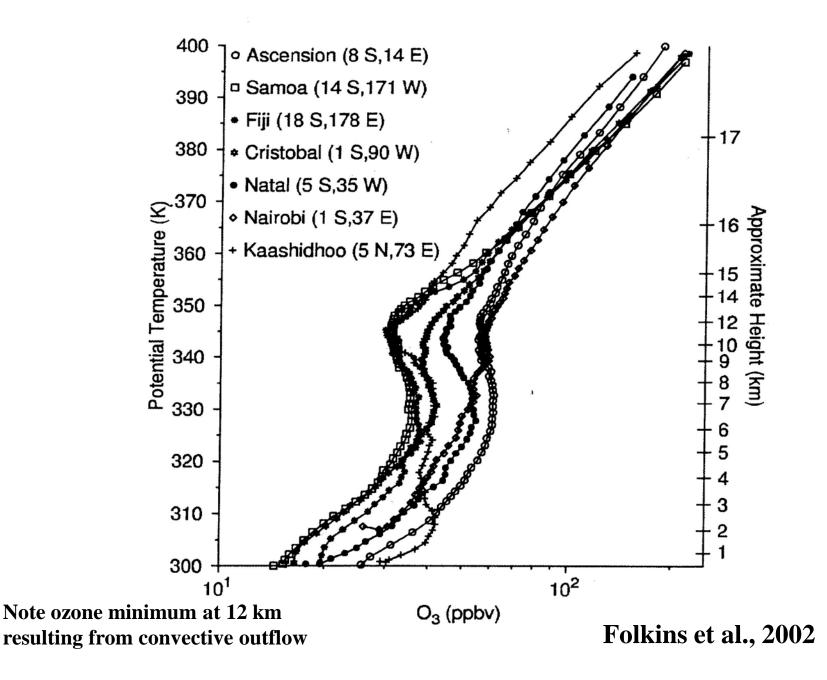
Ozone Production Downstream of 26-27 Sept 92 Convection Box Model Calculations vs. Observations (9.5-12km)



Convection over Remote Oceans

• Pickering et al. (1993) noted O₃ and NO_y minima in UT convective outflow in STEP (near Darwin, Australia) and computed a decrease in P(O₃) due to convection.

• Kley et al. (1996) reported very low ozone measurements near tropopause over the Central Pacific. Resulted from convective transport of very clean marine boundary layer air.



Low Ozone Events in UT Indicative of Convective Frequency

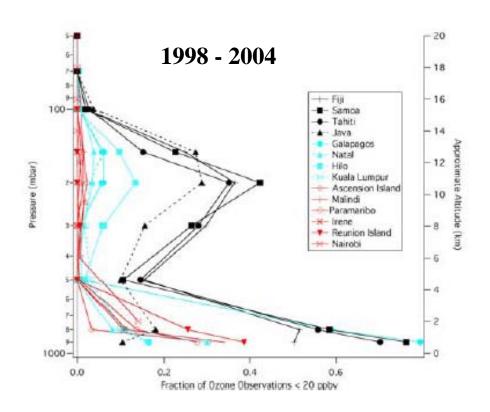
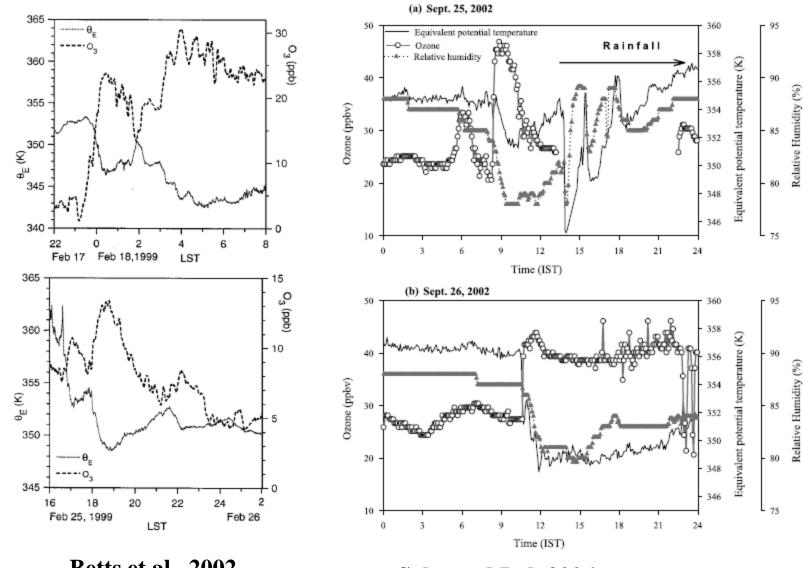


Table 1. Changes in Reduced Ozone in the Upper Troposphere From the Late 1970s to Present

Period	Number of Sondings	Percent of Reduced Ozone Events at 200 mbar
	Samoa	
1986-1990	115	10 (<20 ppbv)
1995-1996	96	18
1997-1999	82	47
2000-2002	104	42
2003-mid 2005	73	32
	Hawaii	
1982-1985	84	7 (<20 ppbv)
1986-1990	221	4
1991-1995	184	15
1996-1999	159	16
2000-2004	189	14
	Natal	
1979-1982	32	3 (<30 ppbv)
1990-1992	65	2
1997-2000	123	18
2001-2002	117	14
2003-mid 2005	103	5

Increases in frequency of low ozone events in the UT in the mid to late 1990s suggest increased convection

Convective Downdrafts Transport Ozone Downward into PBL



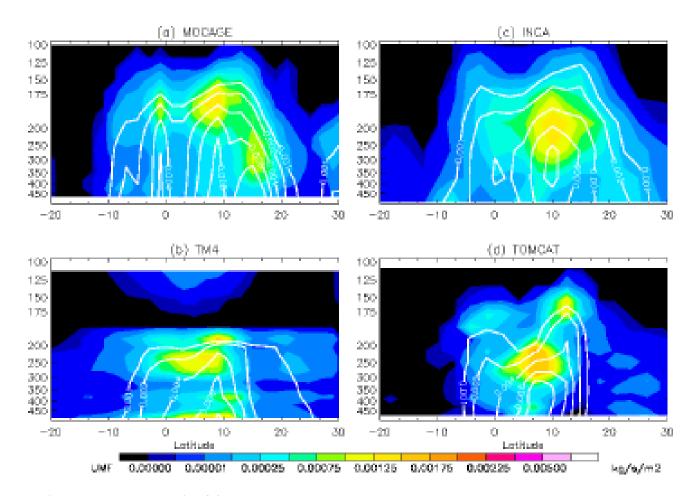
Betts et al., 2002 Rondonia, Brazil

Sahu and Lal, 2006 Bay of Bengal

NET EFFECT OF CONVECTION ON TROPOSPHERIC OZONE

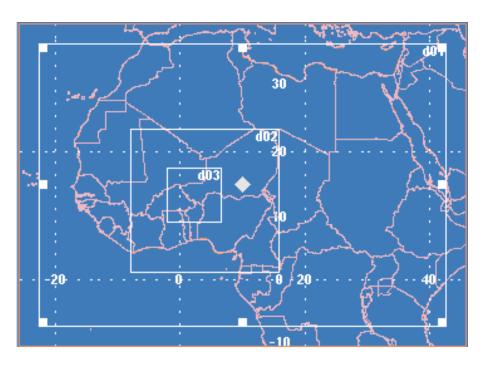
- Lelieveld and Crutzen (1994) model calculations indicate that dominant effect of convection is to enhance ozone destruction
- Lawrence et al. (2003) with better treatment of convection and hydrocarbons found that convection caused an overall net increase of tropospheric ozone
- Doherty et al. (2005) found that convective overturning of ozone dominates over changes in ozone chemistry. Obtained a decrease in global tropospheric ozone burden with convection. Differences in convective and chemical schemes yield results in contrast to Lawrence et al. (2003)

Convective Mass Flux and Detrainment During AMMA as Computed by Four Models



Averaged over 0 -30 deg. E Colors – detrainment White contours – upward convective mass flux

AMMA WRF Simulations



Resolutions: 18, 6 and 2 km

Grid size: 391x271, 424x412, 466x466, and

61 vertical layers

 $\Delta t = 18$ seconds

Starting time: 00Z 08/06/2006

Initial and Boundary Conditions:

NCEP/GFS, no data assimilation

Physics:

• Cu parameterization:

Kain-Fritsch scheme (for the outer grid only)

• Cloud microphysics:

Goddard microphysics 3ice-Graupel

• Radiation:

New Goddard radiation scheme for both longwave and shortwave

• PBL parameterization:

Mellor-Yamada-Janjic TKE scheme

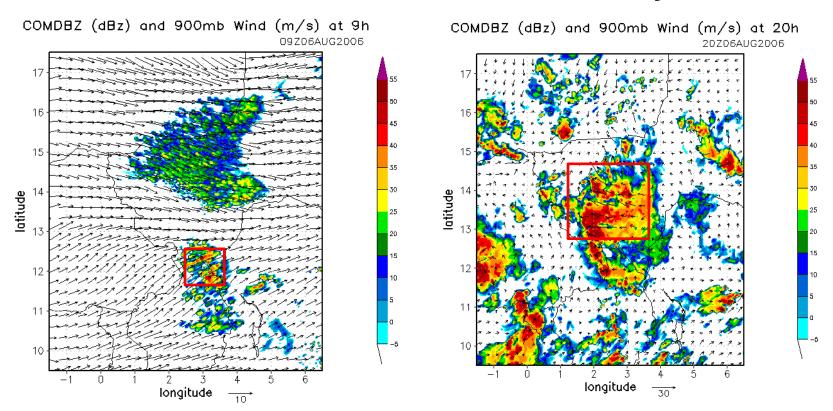
Surface Layer:

Monin-Obukhov (Janjic)

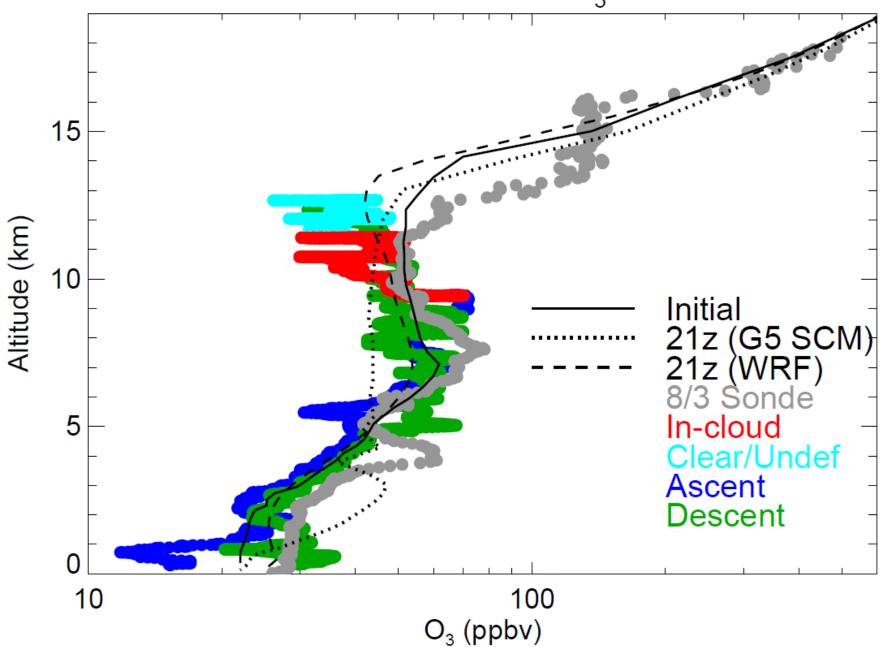
Land Surface Model:

Noah land-surface

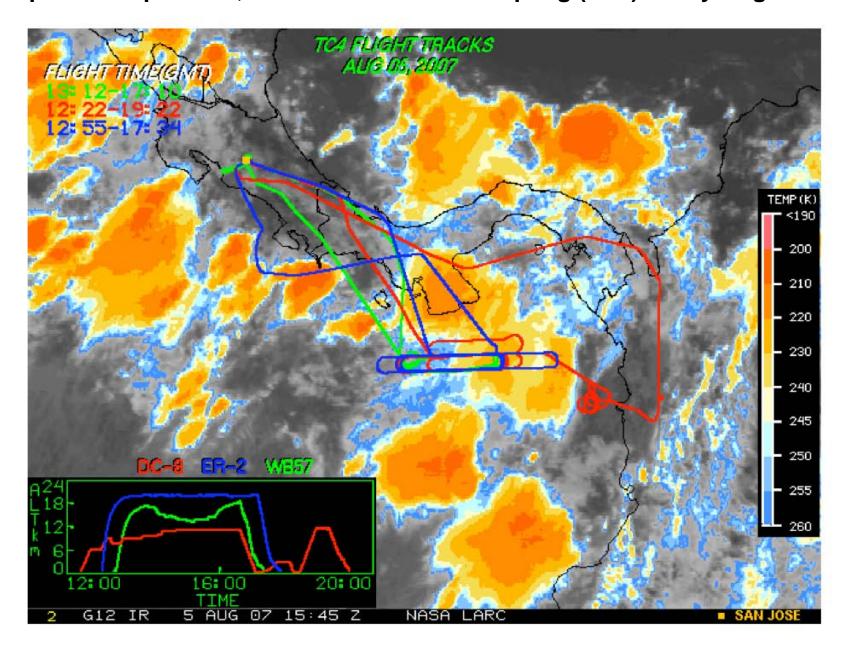
WRF-Calculated Radar Reflectivity



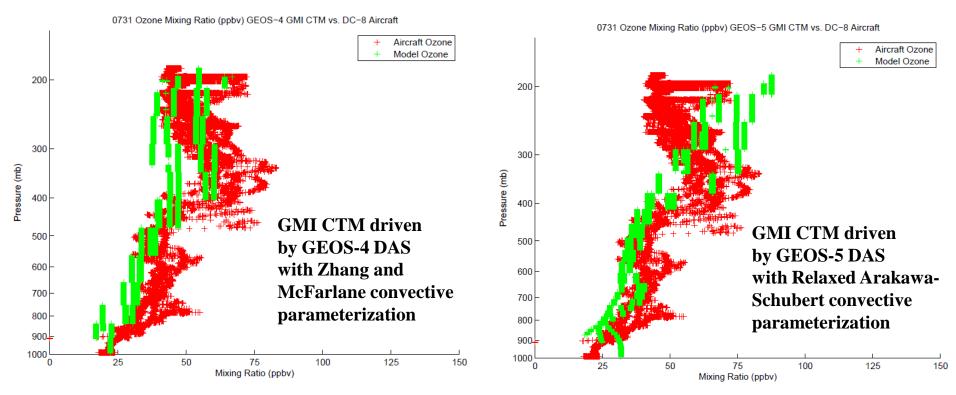
Observed and Simulated O₃ - 20060806



Tropical Composition, Cloud and Climate Coupling (TC4) – July/August 2007



Evaluation of Parameterized Convective Transport in the Offline NASA Global Modeling Initiative (GMI) Chemistry and Transport Model



NASA DC-8 data from TC4 flight matched in time and with nearest grid cell in GMI model with deep convection

Wet Scavenging

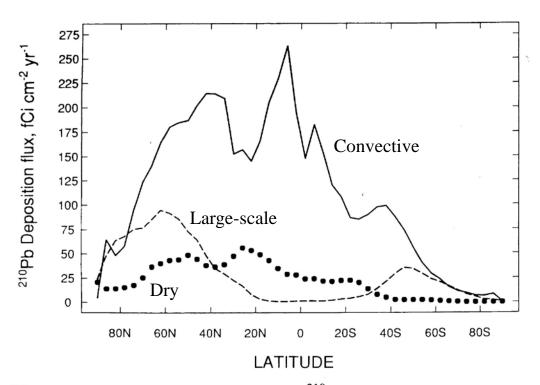
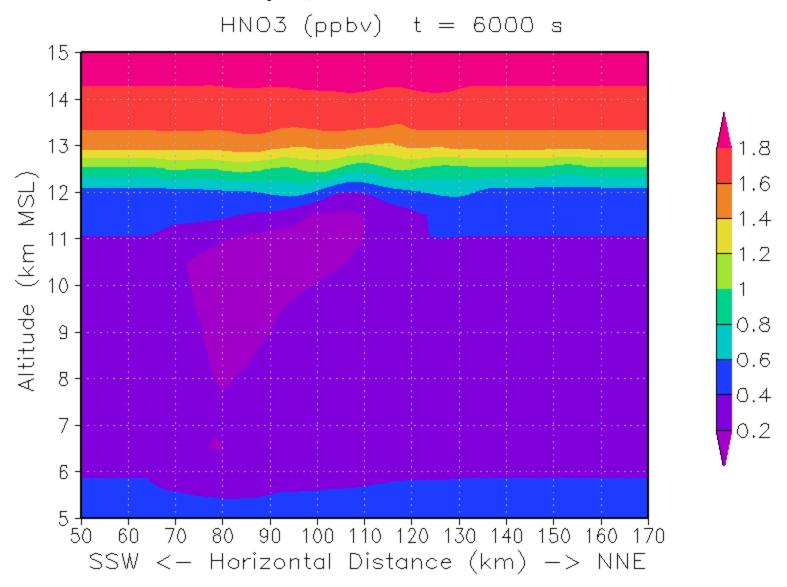


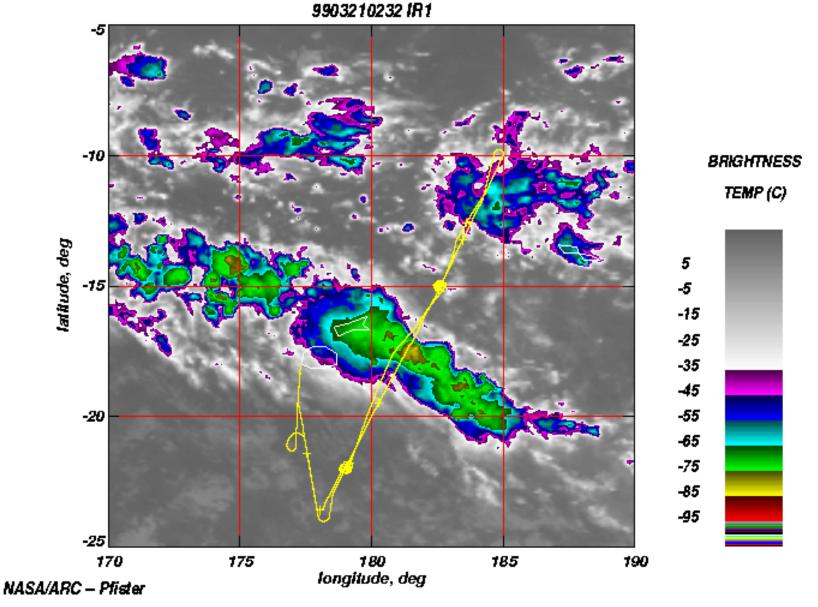
Fig. 2. Zonal mean deposition fluxes of ²¹⁰Pb in the model contributed by convective precipitation (solid line), large-scale precipitation (dashed line), and dry processes (dotted line). Values are yearly averages.

Balkanski et al. (1993)

July 10, 1996 STERAO storm



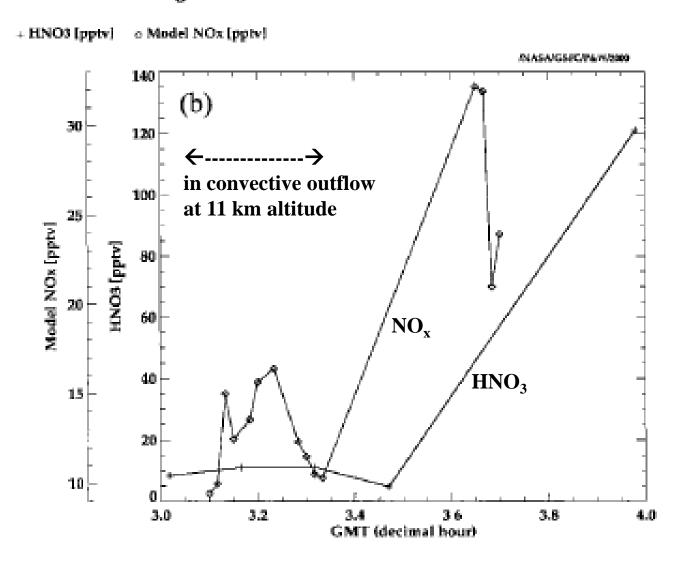
South Pacific Convergence Zone Convection Near Fiji



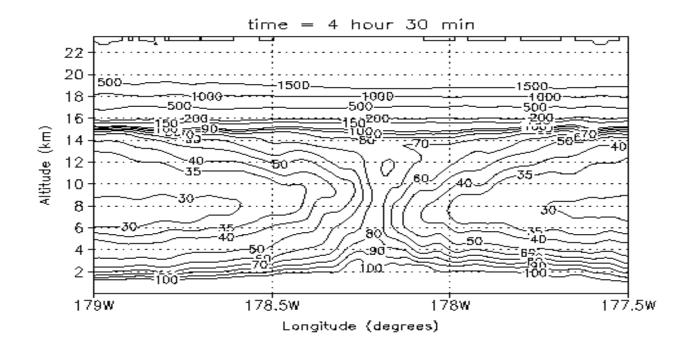
Pickering et al. (2001)

DC-8 Aircraft Measurements in SPCZ System

PEM TROPICS-B Flight 10 [3.00000 - 3.60000] GMT



Pickering et al., 2001



HNO3 simulation for SPCZ convection without wet scavenging (~80 pptv at 11 km)

Comparison with observations (~5-10 pptv) at 11 km within storm suggest ~90-95% removal

Lightning NO Production

• How much NO is produced per cloud-toground (CG) flash and per intracloud (IC) flash? Or per meter of flash length?

Varies over two orders of magnitude

• How are lightning channels distributed throughout a storm?

Some indication of bimodal distribution in the vertical

• How is the NO distributed in the vertical at the end of the storm?

Mostly in middle and upper troposphere

How many flashes occur globally?

Satellite observations indicate ~44
flashes/s

How are the flashes distributed geographically?

At least 75% occur over continents

What is the IC/CG flash number ratio, and how does it vary from storm to storm?

Over continental U.S. annual mean varies from ~1.5 to ~10, with mean ~3

What is the global annual production?

Literature estimates range from 2-20 Tg/yr in the most recent decade, but 2-8 Tg/yr appears most likely

Motivation for Lightning NO Studies

- Production of NO by lightning is an important part of the tropospheric NO_x budget (tropical UT: >50-60%), but it is also the most uncertain component.
- In most of the free troposphere O₃ production rates are highly sensitive to NO_x mixing ratios.
- The maximum effectiveness of ozone as a greenhouse gas is in the UT/LS. Ozone is the third most important greenhouse gas.
- Global annual lightning NO production has been estimated to be 2-20 Tg N/yr, but recent observations and modeling have reduced the range to 2-8 Tg N/yr (Schumann and Huntrieser, 2007)
- Lightning observations from surface networks and satellites are being used in conjunction with cloud-resolving and global models in attempts to further reduce this uncertainty.

Requirements for Specifying Lightning NO Production in Global/Regional Chemical Transport and Climate Models

- 1) NO production per flash (DeCaria et al., 2000; 2005; Ott et al. (2006; 2007; 2010)
- 2) A method of specifying the effective vertical distribution of lightning NO_x at the end of a storm (e.g., Pickering et al., 1998; Ott et al., 2010).
- 3) Flash rates need to be estimated for the times and locations for which parameterized convection is active in the model (e.g., Allen and Pickering, 2002).

Methods of Estimating NO Production Per Flash

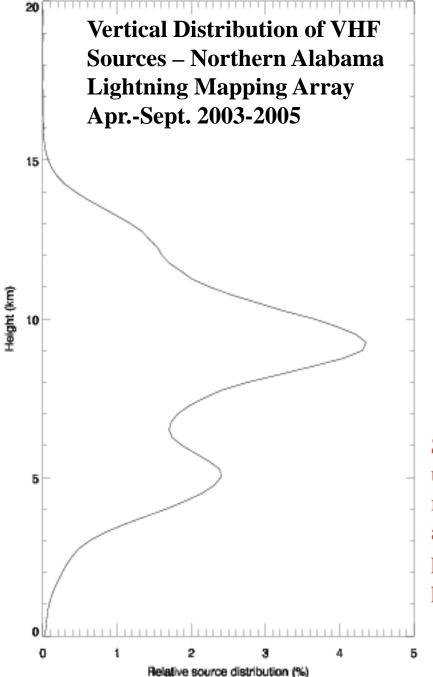
- Theoretical estimates (e.g., Price et al., 1997) 6.7×10^{26} molecules/CG flash ~ 1100 moles/flash 6.7×10^{25} molecules/IC flash ~ 110 moles/flash
- Laboratory experiments (e.g., Wang et al., 1998) 6.2×10^{25} molecules/flash ~ 103 moles/flash
- Field experiments anvil measurements by aircraft of NO from individual flashes and integrated effects (e.g., STERAO-A, EULINOX, CRYSTAL-FACE)
- Cloud-resolved models, lightning parameterizations, anvil measurements (e.g., Pickering et al., 1998; DeCaria et al., 2000; 2005; Ott et al., 2006; 2007; 2010)
- Cloud-resolved models with explicit electrophysics
- Satellite-based NO_2 observations and flash counts (Bucsela et al., 2010)

Cloud-Resolved Model Lightning Placement Parameterization

NO production by lightning injected into the model based on either observed <u>flash rates</u> or <u>flash lengths</u>.

Flash Rate Scheme

- Lightning NO production is calculated using observed CG and IC flash rates over 3-minute periods and specified production of NO per CG and per IC flash.
- NO production is assumed to be proportional to pressure and to the vertical distribution of lightning channel segments which is assumed to be bimodal.
- In each model layer, lightning NO production is distributed uniformly within the 20 dBZ contour.



Similar shape factors used in cloud/chemistry model along with assumption of NO production being proportional to pressure

D. Buechler, NASA/MSFC

NO Production Parameterization

Make initial estimate of P_{CG} using Price et al. (1997) equation: $P_{CG} = E_{CG} P$ where $E_{CG} = 1.823 \times 10^5 I_o$ Joules $P = 1 \times 10^{17}$ molecules NO/Joule $I_o = peak$ current from observations

- Let $P_{IC} = \alpha P_{CG}$ and test production scenarios with various values of α . Compare simulation results with anvil NO_x observations in terms of :
 - 1) mean anvil profile (peak value, shape)
 - 2) probability distributions at specific altitudes
 - 3) anvil column mass

Simulated Storms

<u>Storm</u> <u>Location</u> <u>References</u>	<u>Storm</u>	Location	References
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STERAO - 7/12/96 NE Colorado DeCaria et al.(2000, JGR;

2005, JGR)

STERAO – 7/10/96 NE Colorado Ott et al. (2010, JGR)

EULINOX – 7/21/98 Bavaria Ott et al. (2007, JGR)

Fehr et al. (2004, JGR)

CRYSTAL-FACE

7/29/02* S. Florida Ott et al. (2006;

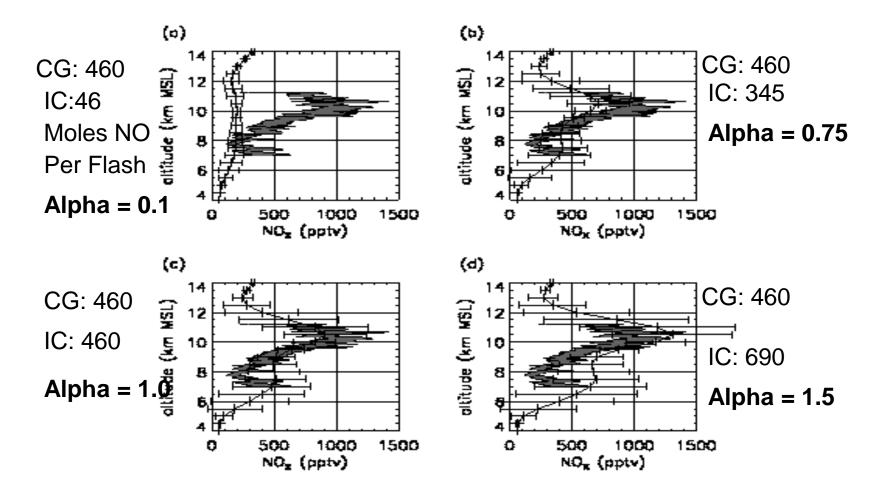
7/16/02** S. Florida 2010, JGR)

^{*} Run using MM5

^{**} Run using ARPS

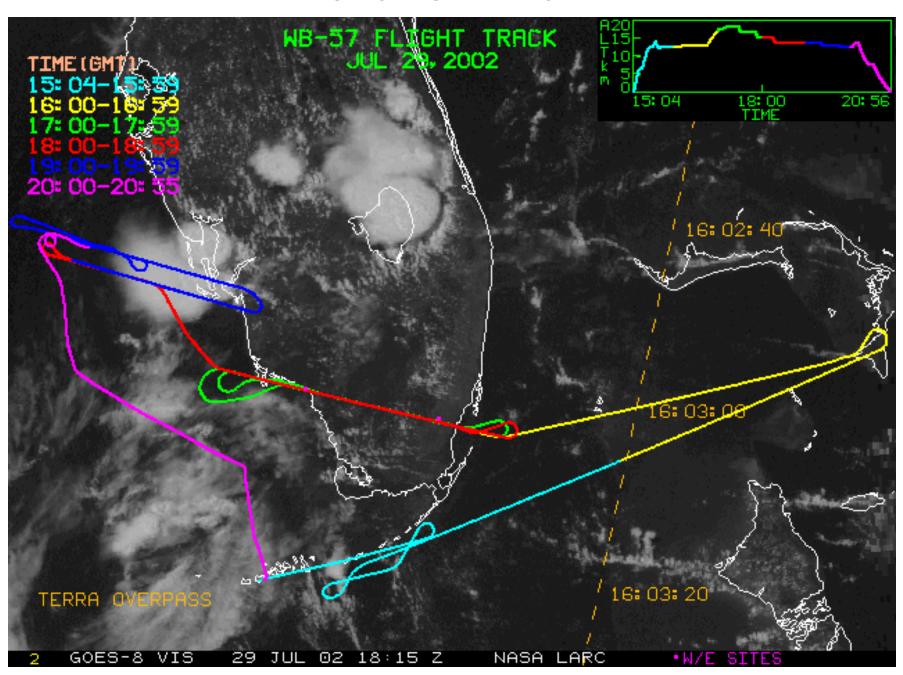


July 12, 1996 STERAO-A Storm – NE Colorado

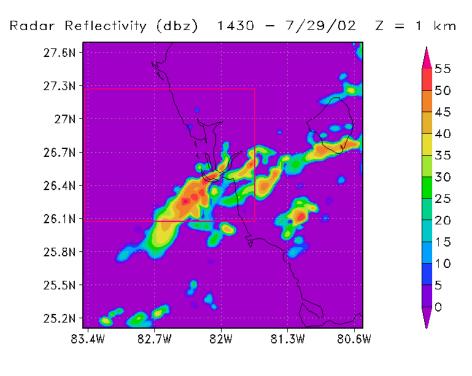


Model-simulated vs. Measured NOx Profiles For Four Lightning NO Production Scenarios DeCaria et al. (2005)

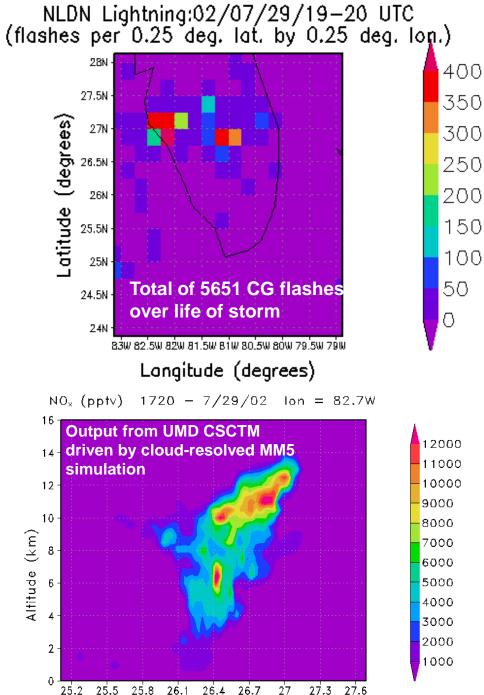
NASA CRYSTAL-FACE

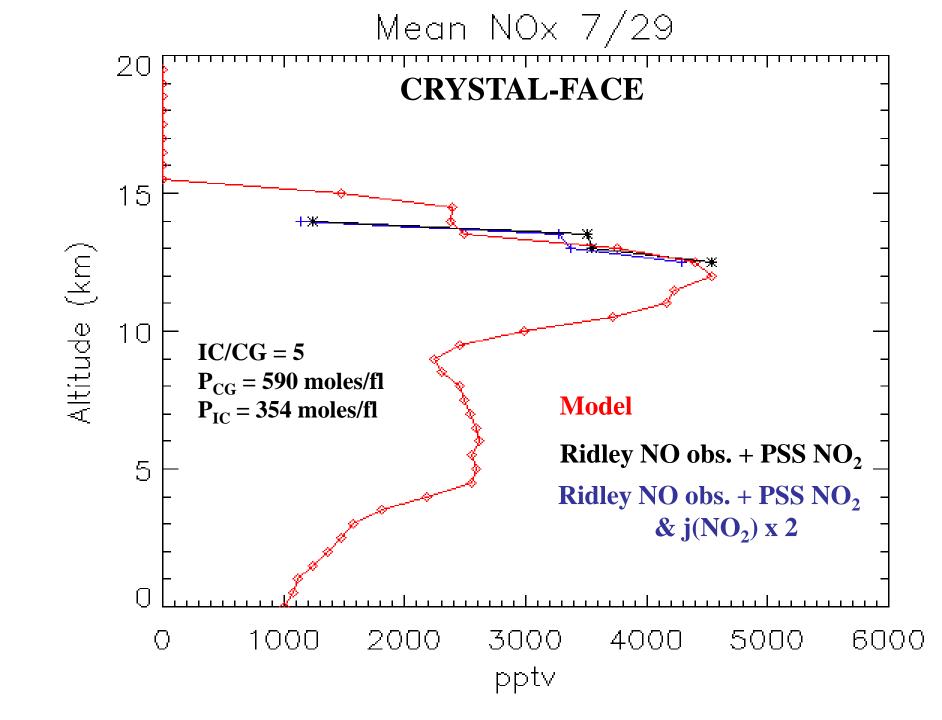


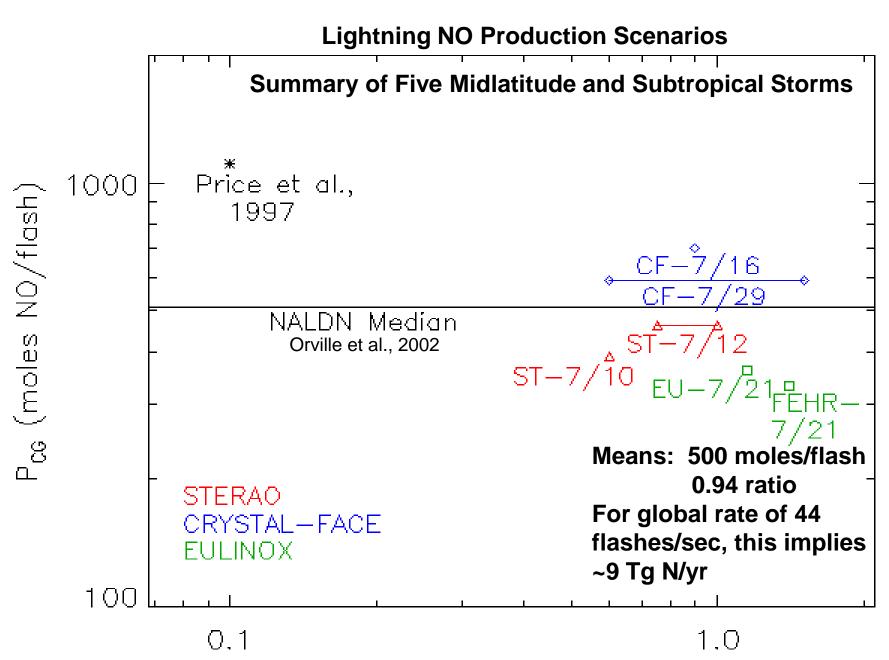
From MM5 simulation run at 2-km horiz. res.



CRYSTAL-FACE South Florida July 29, 2002







Ott et al., 2010, JGR

 P_{IC}/P_{CG}

Vertical Distribution of Lightning NO_x

- Analysis performed by Pickering et al. (1998, JGR) using 2-D cloud/transport model with simple lightning parameterization. These profiles have been used in many regional/global CTMs.
- New calculations of vertical profiles using output from 3-D CSCTM containing a more realistic lightning parameterization have been performed for five midlatitude and subtropical events (Ott et al., 2010). Now used in NASA GMI CTM.

NO Production in Midlatitude, Subtropical, and Tropical Flashes

- Cloud-resolved modeling of observed midlatitude and subtropical storms yields an average of ~500 moles NO per flash (both CG and IC).
- This result is supported for North America by GEOS-Chem model simulations by Hudman et al. (2007, JGR) for the ICARTT period of 2004 evaluated with NASA DC-8 data and by Jourdain et al. (2008, ACPD) evaluated with TES O_3 data.
- Huntrieser et al. (2008, ACP) has hypothesized that on average a tropical flash may produce less NO than a flash in a midlatitude or subtropical storm. This may be due to lesser vertical wind shear in the tropics, leading to shorter flash channel lengths.
- Recent tropical experiments will aid in obtaining improved estimates of LNO_x production per flash

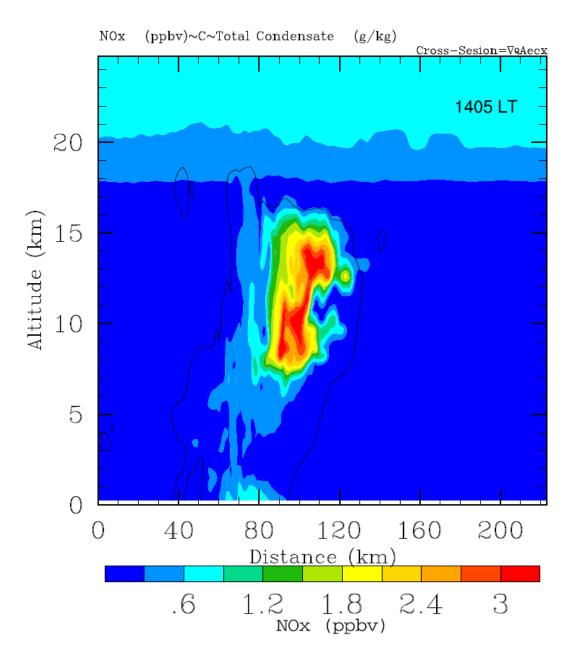
TROCCINOX	São Paulo State, Brazil	Feb. 2005
SCOUT-03/ACTIVE	Darwin, Australia	Nov 2005 – Jan 2006
AMMA	West Africa	Aug. 2006
TC ⁴	Costa Rica, Panama	July – Aug. 2007

 TC^4 = Tropical Composition, Cloud and Climate Coupling

Hector Simulations

- Cloud-resolved chemistry simulation of "Hector" storm observed over Tiwi Islands near Darwin, Australia during the SCOUT-O3 and ACTIVE field experiments. Goal: Estimate LNOx production per flash.
- "Hector" thunderstorm is simulated with the WRF-AqChem cloudresolving model (Barth et al., 2007) at 1-km horizontal resolution, and cloud simulation is evaluated with radar, satellite, and aircraft data
- Trace gases are transported and chemical reactions are computed on-line in the cloud simulation. Same lightning scheme as in GCE/CSCTM.
- How does NO prod. per flash in Hector storm compare with that in higher latitude storms? Run simulation with assumption of 500 moles/flash.
- Work in progress!

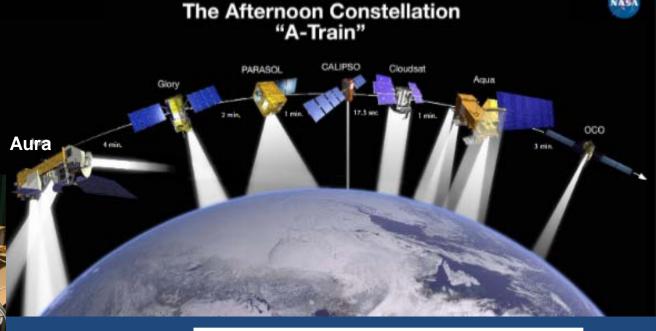
Hector WRF-AqChem simulation with Lightning NOx



K. Cummings

Aura/OMI

Ozone Monitoring Instrument



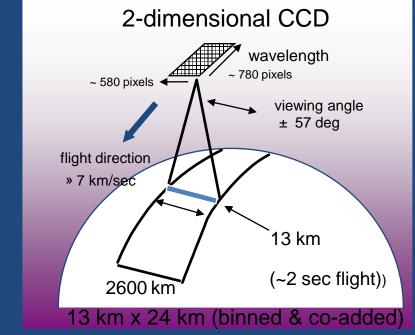
Wavelength range: 270 – 500 nm

Sun-synchronous polar orbit; Equator crossing at 1:30 PM LT

2600-km wide swath; horiz. res. 13 x 24 km at nadir

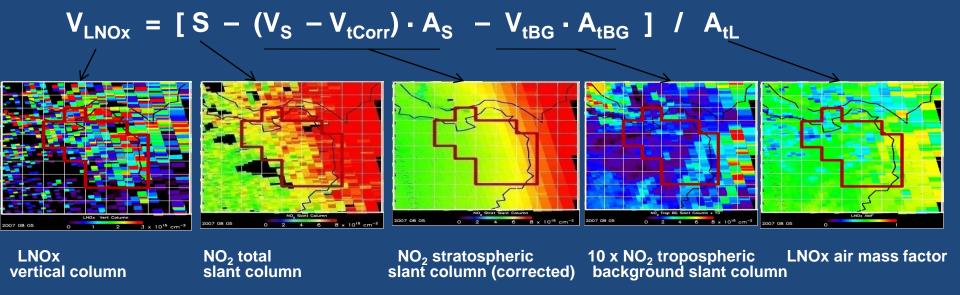
Global coverage every day

O₃, NO₂, SO₂, HCHO, aerosol, BrO, OCIO



Analysis of LNO_x from OMI

The vertical column of NO_x due to lightning is:



V_s = OMI-derived stratospheric NO2 vertical column ("clean" region data with wave-2 pattern imposed)

 V_{tCorr} = GMI model correction (about ~10%) of V_s for tropospheric contamination

V_{tBG} = GMI model tropospheric background in region of lightning measurement

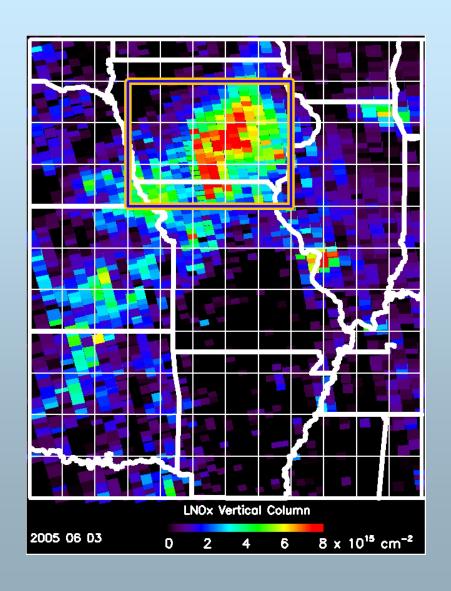
Analysis of Results from TC4 cases

<u>Day</u>	LNO _x (moles/flash)	300 hPa NCEP wind (m/s)
7/31	246 ± 287	8
8/5	227 ± 223	14
7/17	87 ± 252	4
7/21	135 ± 114	2

Smaller LNO_x production per flash is associated with weaker upper tropospheric wind speeds in these example cases.

Perhaps lesser flash length associated with weaker anvil-level winds?

Case 1: Production Per Flash



Summing LNOx over the box and adjusting for 2-4 day chemical lifetime (7.2% decay in this case) we obtain:

 8452 ± 4858 kmoles

Dividing by vertically massweighted flash count of 15,829 flashes yields:

534 ± 351 moles LNOx per flash

Vertical Distribution of Lightning NO_x

- Analysis performed by Pickering et al. (1998, JGR) using 2-D cloud/transport model with simple lightning parameterization. These profiles have been used in many regional/global CTMs.
- New calculations of vertical profiles using output from 3-D CSCTM containing a more realistic lightning parameterization have been performed for five midlatitude and subtropical events (Ott et al., 2010). Now used in NASA GMI CTM.
- Direct use of vertical profiles derived from 3-D Lightning Mapping Array data being used in CMAQ and WRF-Chem regional models.

Summary

- Convection is an effective mechanism for transporting pollution from the boundary layer to the upper troposphere where it can more readily contribute to intercontinental transport.
- As a result, ozone production is enhanced, contributing to enhanced radiative forcing.
- Ozone destroyed as a result of remote marine convection.
- Net effect of convection on tropospheric O_3 remains uncertain.
- Lightning is a major contributor to the upper tropospheric NO_x budget and to ozone production.
- On a per flash basis, IC flashes are nearly as productive of NO as CG flashes. For five simulated storms, estimates of mean NO production per flash vary by a factor of two.
- Approximately 500 moles NO produced per flash on average over the five midlatitude and subtropical storms → ~9 Tg N/yr. Simulations of tropical events in progress.

Future Research

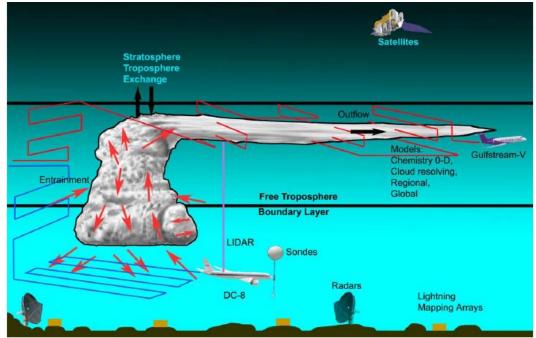
- Lightning NO need more field projects with comprehensive data collection (radar, 3-D lightning flash mapping, chemistry)
- Ozone measurements downstream of convection to evaluate model estimates of ozone production
- Wet scavenging better 3-D precipitation fields needed for use in CTMs; measurements of soluble species in cloud-scale field experiments
- Aerosol effects on convective strength and lightning
- Studies of new particle production in convective outflows
- Evaluation and improvement of convective parameterizations in regional and global models

Deep Convective Clouds and Chemistry – DC3

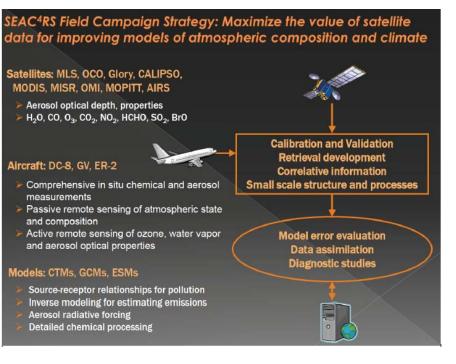


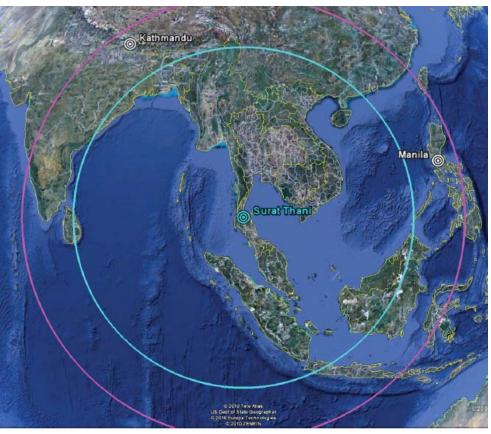
May/June 2012





Southeast Asia Composition, Cloud, Climate Coupling Regional Study (SEAC4RS)





Measurement Objectives:

- 1) Characterize the chemical gradients associated with the dynamical background of the Asian Monsoon Anticyclone.
- 2) Characterize the chemical composition of convective outflow and microphysical properties of anvil cirrus.
- 3) Characterize the chemical and meteorological impact of biomass burning plumes.

August/September 2012

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