Overview of Lightning NOx Production

Deep Convective Clouds and Chemistry (DC3) Experiment

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Lightning NO_x

- **Lightning is responsible for approximately 10-15% of NO_x emissions globally.   This is roughly 2 – 8 Tg N yr-1 [Schumann and Huntrieser, 2007]. Much of uncertainty stems from little knowledge of NO_x production per flash or per unit flash length.**

- **Most of lightning-produced NO_x (LNO_x) is injected into middle and upper troposphere. Lifetime is relatively long (a few days) relative to lower troposphere (hours). NO_x in this region plays a key role in the chemistry of ozone, the importance of which as a greenhouse gas maximizes in the UT.**

- **Methods used to estimate LNO_x/flash include theoretical estimates, laboratory experiments, analysis of aircraft NO_x observations and flash rates, cloud-resolved chemistry modeling constrained by aircraft obs., and analysis of satellite NO_2 data.**

Previous investigations of lightning NO_x production for individual storms

Method	Moles NO/flash (Notes)	Reference
Theoretical	1100 (CG), 110 (IC)	Price et al., 1997
Laboratory	~103	Wang et al., 1998
Aircraft data, cloud model	345-460 (STERAO-A)	DeCaria, et al., 2005
Aircraft data, cloud model	360 (STERAO-A, EULINOX)	Ott et al., 2007; 2010
Aircraft data, cloud model	590-700 (CRYSTAL-FACE)	Ott et al., 2010
	500 (Mean midlat. from model)	Ott et al., 2010
Satellite (OMI)	440 (Central US, Gulf)	Pickering et al. (in prep)
LMA/Theoretical	484 (CG), 34 (IC)	Koshak et al., 2012 (in press)
Aircraft data	70-210 (TROCCINOX)	Huntrieser et al., 2008
Aircraft data	121-385 (SCOUT-O3 Darwin)	Huntrieser et al., 2009
Aircraft data	70-179 (AMMA)	Huntrieser et al., 2011
Aircraft data, cloud model	500 (Hector)	Cummings et al., 2012 (in press)
Satellite (GOME)	32-240 (Sub-Tropical)	Beirle et al., 2006
Satellite (OMI)	87-246 (TC4 – tropical marine)	Bucsela et al., 2010
	174 (TC4 mean from OMI)	Bucsela et al., 2010
Satellite (SCIAMACHY)	33-50 max. (global analysis)	Beirle et al., 2010

Recent aircraft/cloud model studies suggest intracloud (IC) flashes at least as productive as cloud-to-ground (CG) flashes

Mid-latitude storms possibly more productive per flash than tropical storms (Huntrieser et al., 2008)

Topics

- Brief review of facilities/data used in DC3 LNOx analyses
- Flash rates and other flash characteristics from LMA data
- LNOx per flash from aircraft data analysis
- LNOx per flash from cloud-resolved modeling of DC3 storms
- Improvement of representation of LNOx in models using DC3 LMA/radar data
- LNOx analysis from regional and global models
- LNOx per flash from satellite NO2 observations

Facilities Required for Acquiring Data Necessary for LNOx Analyses

Facilities

- **<u>Dual Doppler polarimetric radar</u> - defines storm volume; provides velocity, reflectivity, and microphysical fields to evaluate models; enables relating flash locations to storm dynamical and microphysical fields**

  CO: CHILL, Pawnee, NWS NEXRADs

  AL: ARMOR, MAX, KHTX

  OK/TX: KOUN, SR-1, SR-2

- **<u>Lightning Detection</u> – provides lat/lon, time, peak current, multiplicity for each flash; gridded flash rates as f(time) can be constructed.**

  National Lightning Detection Network (NLDN) – detects CG flashes with 90-95% efficiency; detects some not-well defined fraction of IC flashes; some rough estimate of IC flashes can be obtained from climatology of IC/CG ratio applied to CG flashes.

Facilities

- <u>Lightning Detection</u> –

  Lightning Mapping Arrays - detect VHF radiation sources from flash components

  Northern Alabama

  Oklahoma and Oklahoma western extension

  West Texas

  Northeast Colorado

  Flashes constructed from source points using time and space criteria; gridded flash count files being developed

- <u>Environmental Soundings</u> – provide profiles of temperature, dew point, winds in pre-storm and inflow environments; can be used to evaluate regional and cloud-resolved models

Facilities

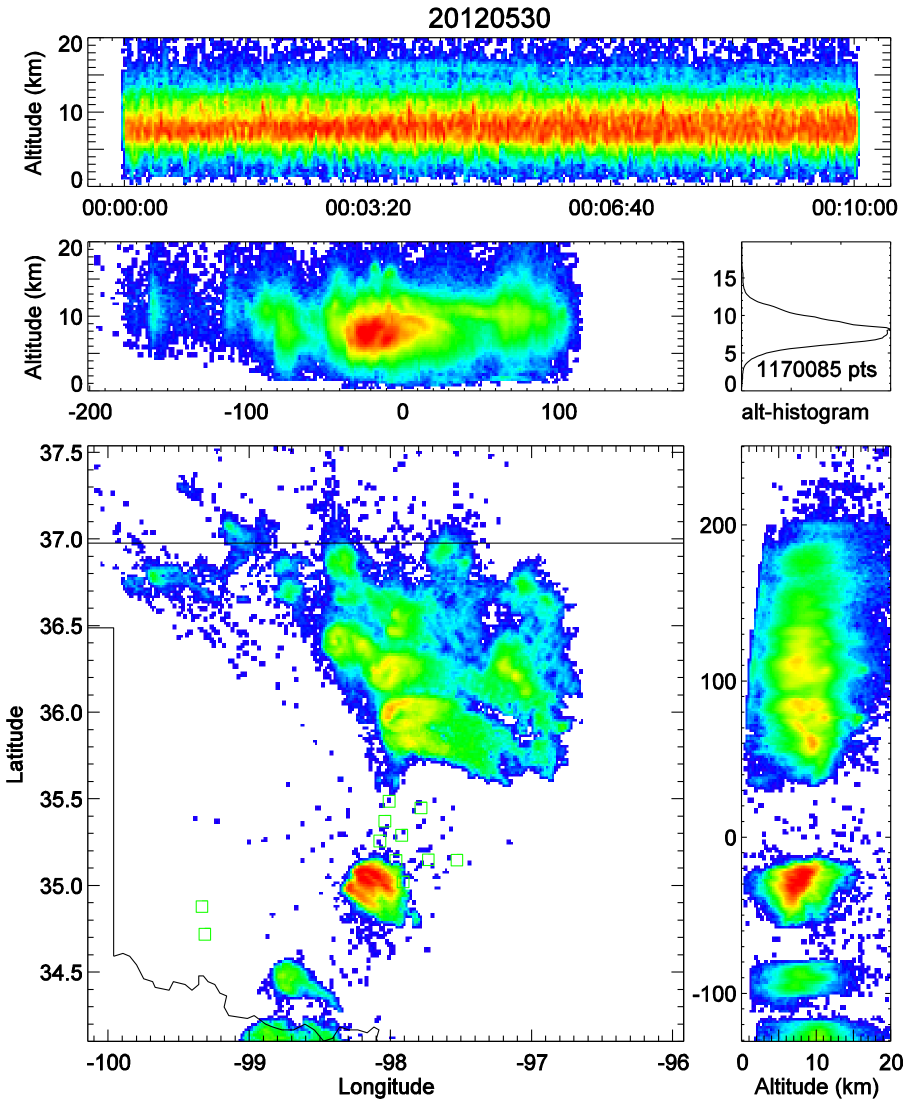
- **<u>Aircraft Observations of Trace Gases</u> – provide inflow and outflow observations of CO, O_3, VOCs, halocarbons, NO, NO_2; tracers used to help define inflow and outflow regions and to evaluate convective transport in cloud-resolved models.**

  Need NO_x observations at multiple levels, multiple distances from convective core, and as a function of time within anvil to best constrain NO production per flash.

Flash Rates and Flash Characteristics

Eric Bruning, Don MacGorman, Paul Krehbiel, Ron Thomas, Larry Carey, Bill Koshak

Oklahoma LMA Plot for a 10-min Segment of 29-30 May Storm



Time vs. altitude

X vs. Z plot

Altitude histogram of source points

Lat./Lon. Map of source points

Y vs. Z plot

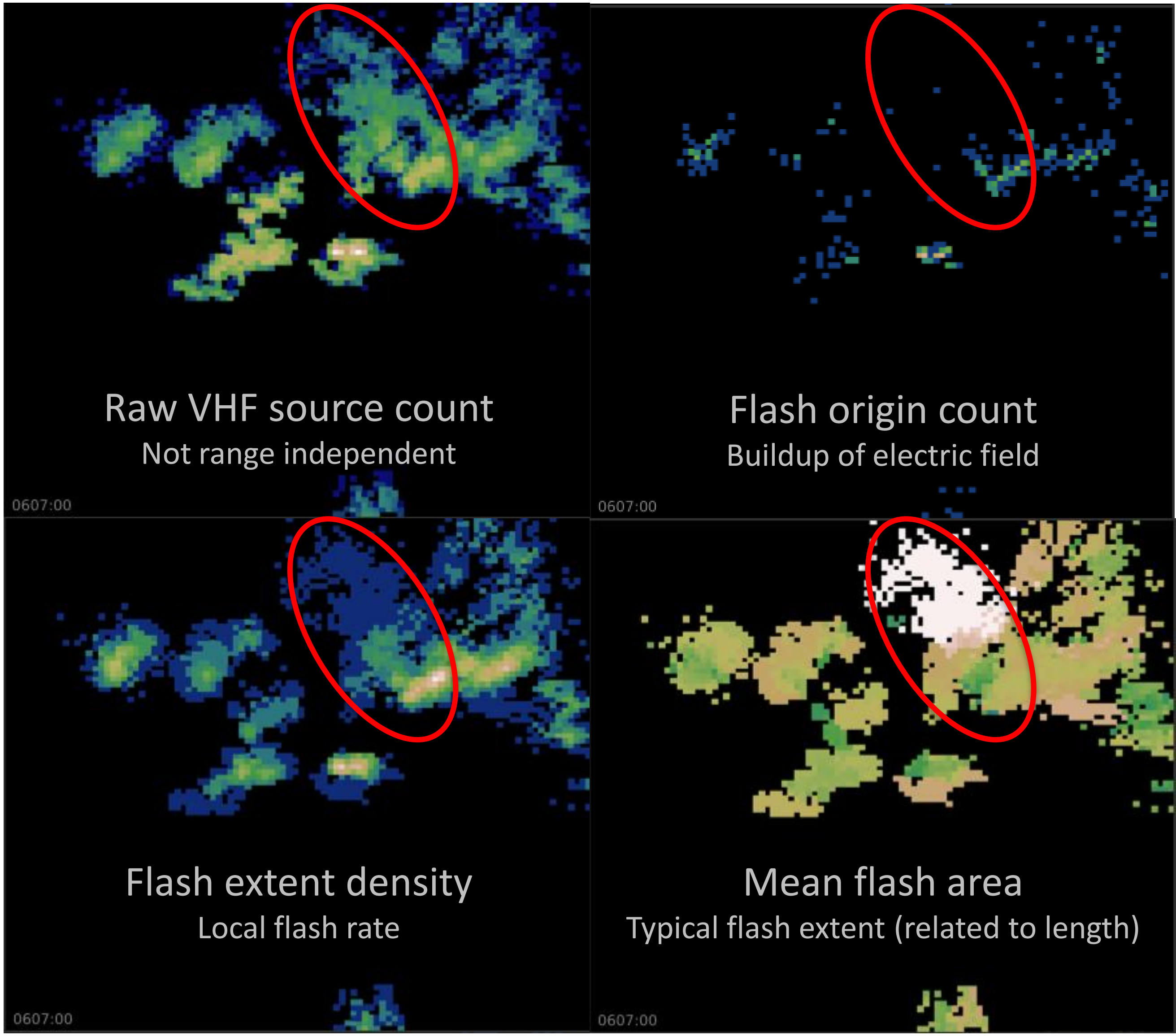
LMA Products

- **Gridded products -- 3-km horizontal resolution; 5-min time res.**
  - **Source density – number of VHF sources per grid cell**
  - **Flash initiation density – number of flashes initiating in a grid cell (flash counts)**
  - **Flash extent density – number of flashes passing through a grid cell (local flash count)**
  - **Mean flash area – Sum of areas of each flash passing through a grid cell divided by flash extent density (related to flash length)**

Gridded products completed for W. TX and AL; soon to be run for OK

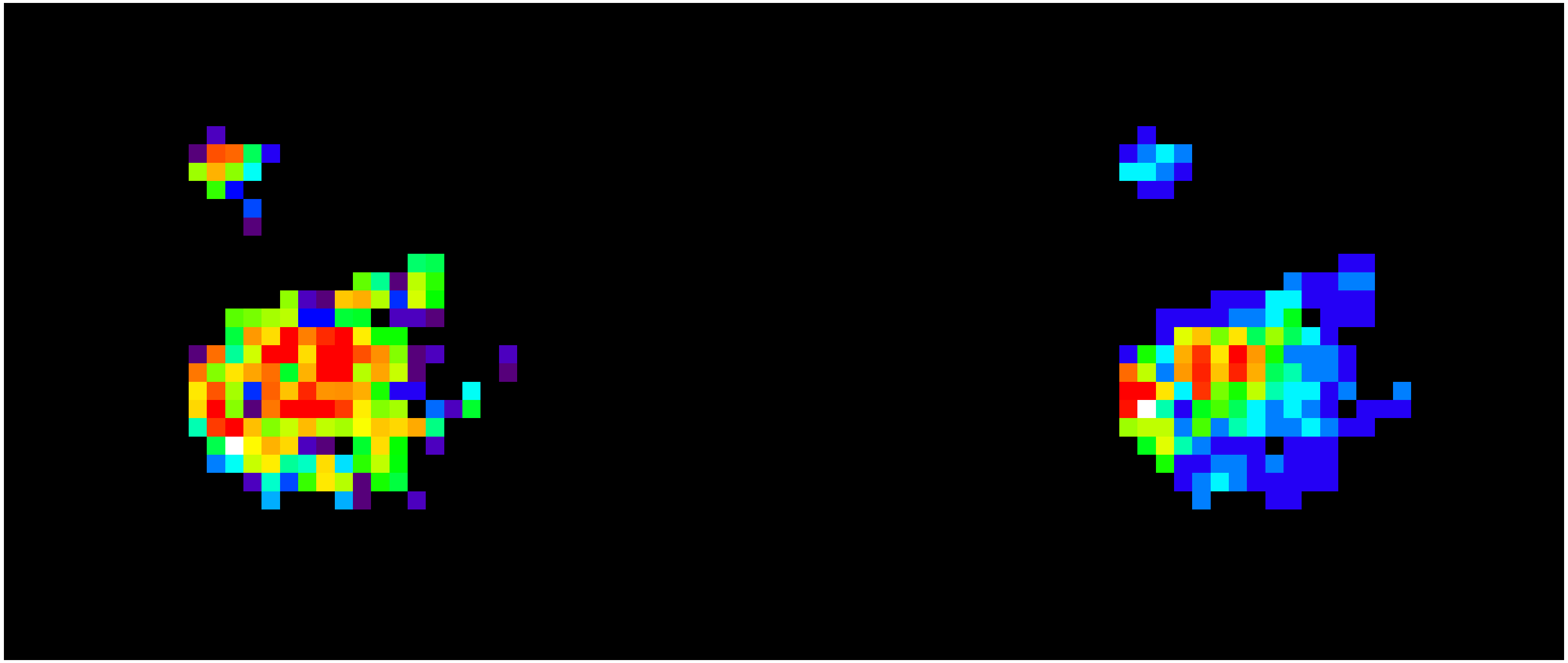
Sample preliminary LMA flash products West Texas, 12 June 2012, 0607 UTC

- 400 x 400 km domain
- 4 km x 1 min grids
  - Final products will be 3 km to match model grids
  - CF-compliant NetCDF
- Flash sorting: McCaul et al., 2009
  - Results in HDF5 format w/ VHF source and flash metadata
  - Include CG yes/no
- Gridding: open source
  - https://bitbucket.org/deeplycloudy/lmatools
- Products emphasize differences in typical local flash characteristics
  - Consider region highlighted by oval
  - Low-rate, extensive in stratiform regions.
  - High-rate, compact in convective regions, especially new updrafts



Example Gridded LMA Products from CO-LMA

June 22, 2012 22:46:29 – 22:47:00



3-km gridded VHF sources **3-km gridded flash counts**

R. Thomas, NMT

Flash length algorithms: planned comparison for DC3

- 3 estimation methodologies
  - LNOM (Koshak/Peterson, MSFC), Thomas (NMT), Bruning (TTU)
  - Generally similar methods (connect-the-dots, box coverage, fractal ideas), but tuning can result in factor-of-two differences
- Compare length estimates with each method on a set of common datasets
  - Total length, altitude distributions of channel segments
  - Across networks, expect some differences in detection of + leaders, average number of sources per unit of length
  - Utilize local strengths
    - Relatively mature LNOM estimates from Alabama
    - Very well-resolved channels in Colorado
    - Large domain in OK/TX for studies of large, long-lived systems

Fractal channel length estimates

Bruning, with Thomas, Koshak, Peterson

- Global properties: Fractal dimension, channel stepping length, convex hull area or volume filled by flash
  - Independent of LMA detection efficiency
  - Highly nonlinear sensitivity to choices of D, step.
  - Good for whole-storm estimates
  - AMS 2013: consensus on D=1.5, step ~= 150 m?
    - Matches LNOM results for one hour of data from NALMA

$$L = N_s b_s = b_s \left(\frac{\sqrt{A_h}}{b_s} \right)^D = \frac{(\sqrt{A_h})^D}{(b_s)^{D-1}}$$

- Local flash length (vertical distribution):
  - Weight global total by local natural neighbor volume given by Delaunay triangulation of VHF sources
  - Nearly identical to vertical distribution of VHF sources

$$L_i = k \frac{S_i}{P_i} L = \sqrt[3]{\frac{V_h^D}{V_i^{D-1}}}$$

- Future work: variation of step length with height; polarity dependence?
- See Bruning et al. poster, this meeting

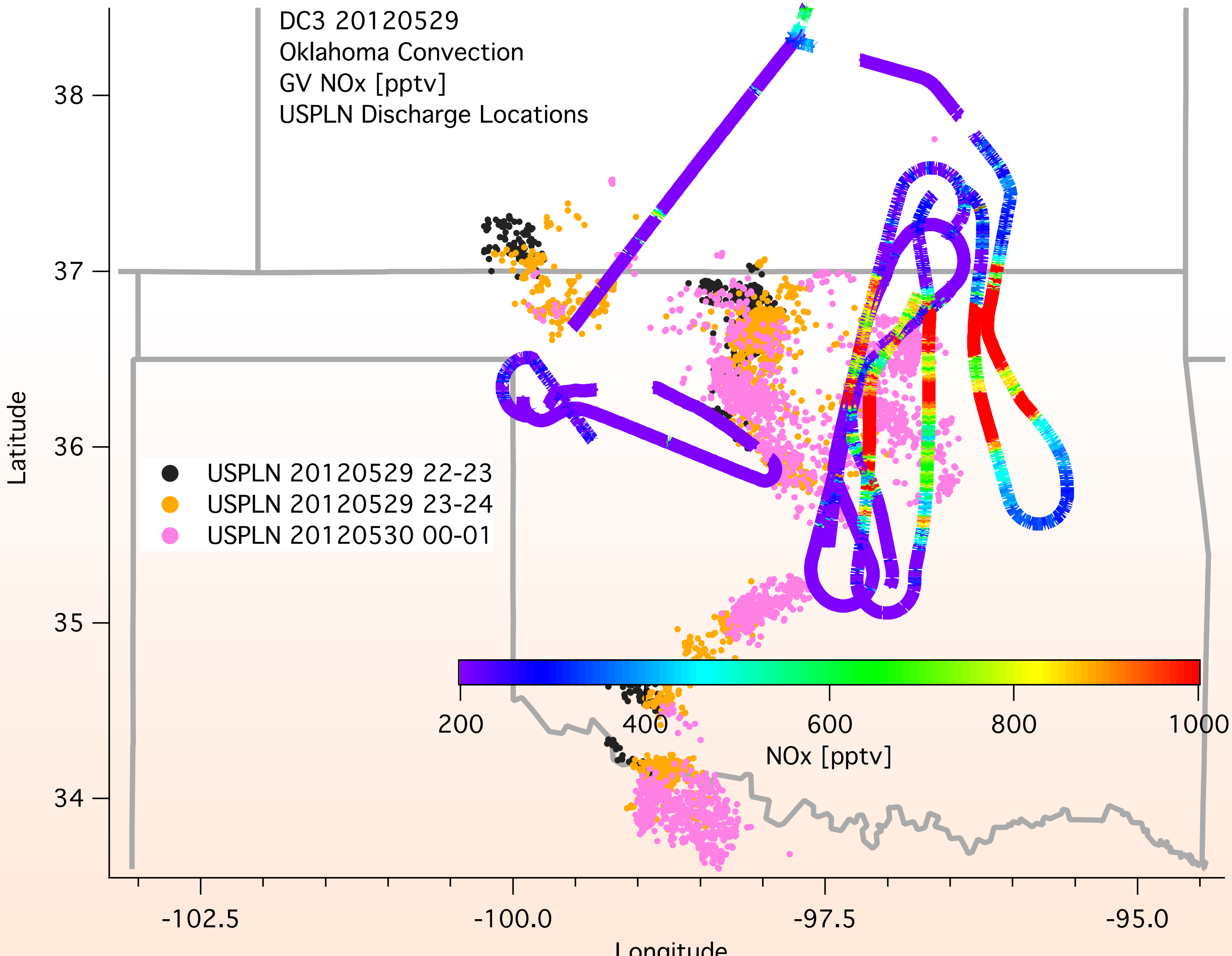
LNOx Estimates from Aircraft Data Analysis

Ilana Pollack, Andy Weinheimer, Heidi Huntrieser

Oklahoma Storms

- **Andy Weinheimer - interest in May 29 and June 16 cases**
- **Ilana Pollack – interest in May 19, 25, 29 and June 16 cases**

Weinheimer


DC3 20120529
Oklahoma Convection
GV NOx [pptv]
USPLN Discharge Locations
USPLN 20120529 22-23
USPLN 20120529 23-24
USPLN 20120530 00-01
Latitude
38
37
36
35
34
200
400
600
800
1000
NOx [pptv]
-102.5
-100.0
-97.5
-95.0
Longitude

Pollack et al.

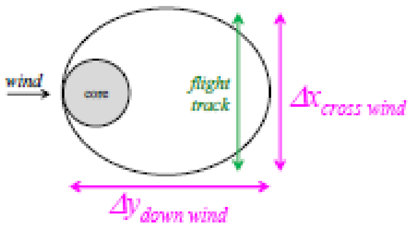
See poster

May 19
Oklahoma

3. NOx production using Volume method

Molecules NO_x estimated from **volume**
Ridley et al. (1996, 2004), Koike et al. (2007)

Top View



$$P(NO_x) \propto LNO_x^{enh} * Volume$$

$P(NO_x)$ in units of molecules

Divide by #flashes to get molecules $flash^{-1}$

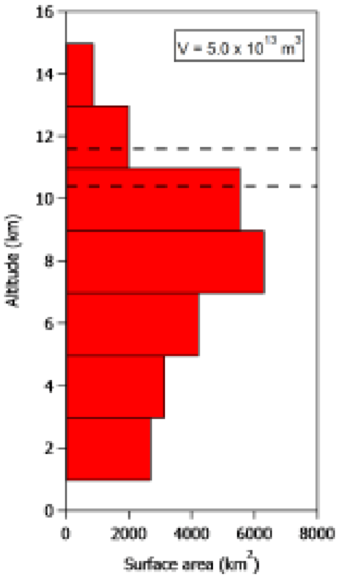
	z (km)	Mean $NO_x^{outflow}$ (ppbv)	Mean NO_x^{trans} (ppbv)	Mean $LNO_x^{enh}(z_i)$ (ppbv)	NO_x (molecules m^{-3})
Pass 1	10.4	1.24	0.22	1.02	8.5×10^{15}
Pass 2	11.6	1.64	0.21	1.43	10.3×10^{15}

Average $NO_x = 9.4 \times 10^{15}$ molecules m^{-3}

1097 cloud-to-ground (CG) lightning flashes estimated from NLDN → *Flashes counted from storm start to sampling end (Δt = 4 hrs) in LAT/LON region of storm*

$$P(NO_x) = \frac{(9.4 \times 10^{15} \text{ molecules m}^{-3}) * (5.0 \times 10^{13} \text{ m}^3)}{(1097 \text{ CG flashes})} = 42 \times 10^{25} \text{ molecules CG flash}^{-1}$$

Estimated storm volume from composite radar images



- Use CAPPI (constant altitude plan position indicator) images - 2 km vertical resolution
- Images selected during outflow sampling and when storm is most developed
- Image surface area calculated from pixels > 20 dZB; Total volume = 2 km*∑(surface areas)

Pollack et al.

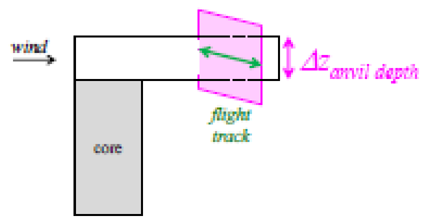
See poster

May 19
Oklahoma

4. NOx production using Flux method

NO_x **flux** out of anvil
Chameides et al. (1987), Huntrieser et al. (1998, 2002)

Vertical Cross Section



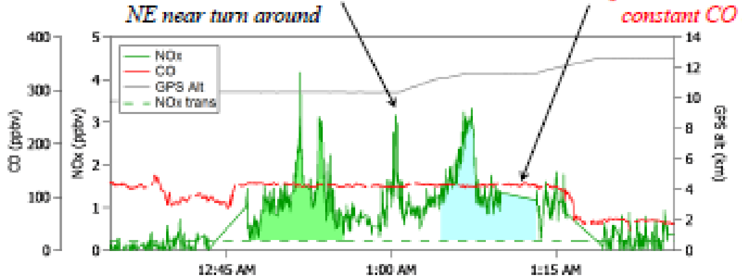
$$P(NO_x) \propto v_{wind} * \int^{z} n_{air} * \int^{x} LNO_x$$

$P(NO_x)$ in units of molecules s^{-1}

Divide by #flashes s^{-1} to get molecules $flash^{-1}$

sampled different storm to NE near turn around

NO_x enhancements detected above constant CO background



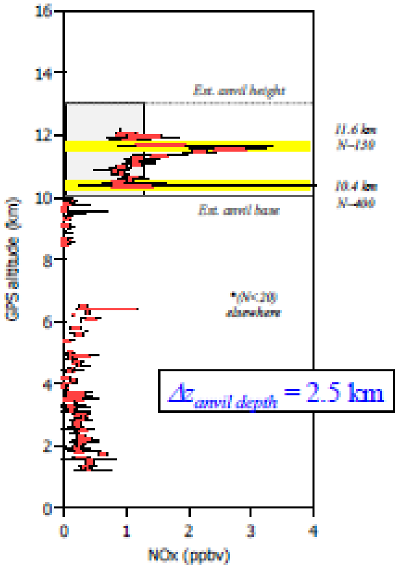
DC-8 data

pass 1	*pass 2*	
2.3 x 10^{25} molecules s^{-1}	3.5 x 10^{25} molecules s^{-1}	Average = 2.9 x 10^{25} molecules s^{-1}

Average CG flash rate (0.08 flashes s^{-1})
→ assumes constant average distribution of 1097 flashes over Δt = 4 hrs

$$P(NO_x) = \frac{(2.9 \times 10^{25} \text{ molecules s}^{-1})}{(0.08 \text{ CG flashes s}^{-1})} = 38 \times 10^{25} \text{ molecules CG flash}^{-1}$$

Estimated anvil depth from NO_x vertical distribution

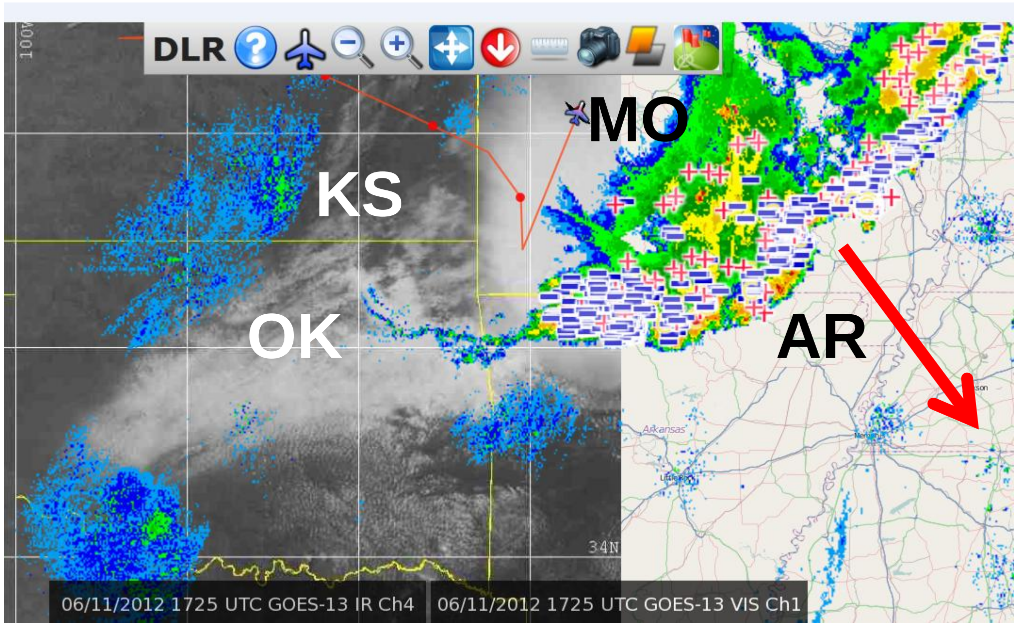


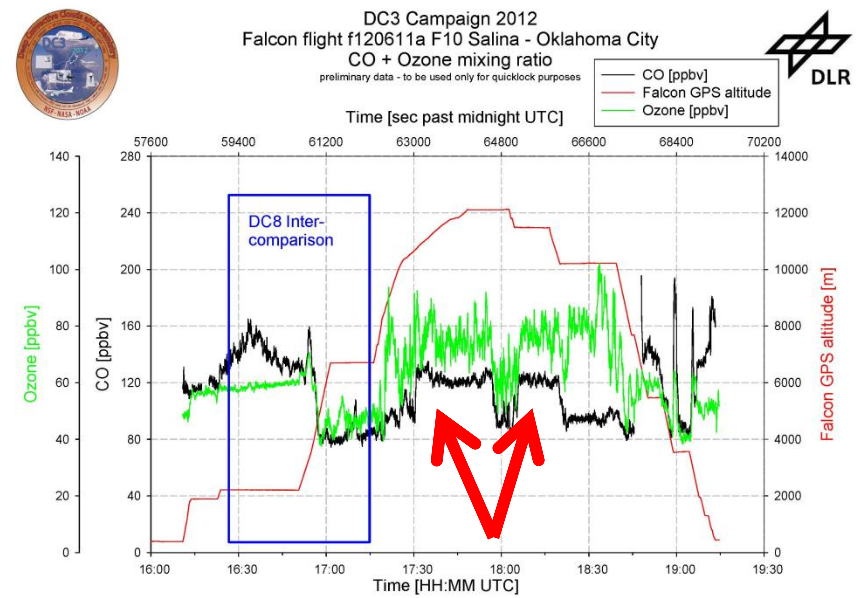
- Minimal samplings, but similar medians
- Assumes constant average NO_x distribution

DLR Falcon Flights

- Several flights in fresh anvil outflow on **29, 30 May and 5, 11, and 12 June 2012**
- → however unfortunately most anvil penetrations **outside of the DC3-LMA** domain
- → **NLDN and LIS data will be used to estimate the flash rate in the storms**

Falcon A-flight on 11 June 2012: Widespread MCS over Missouri and Arkansas





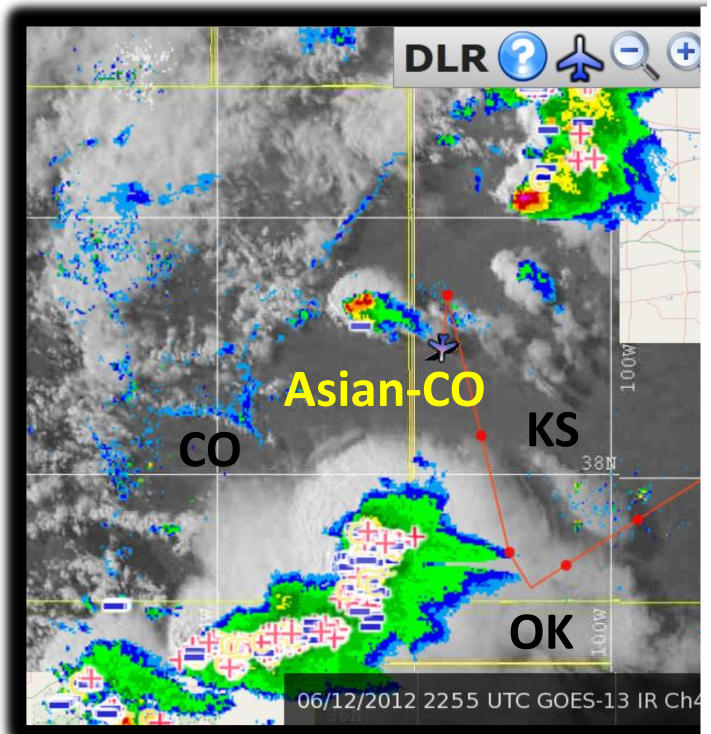
Flight on 11 June 2012 to Oklahoma City:

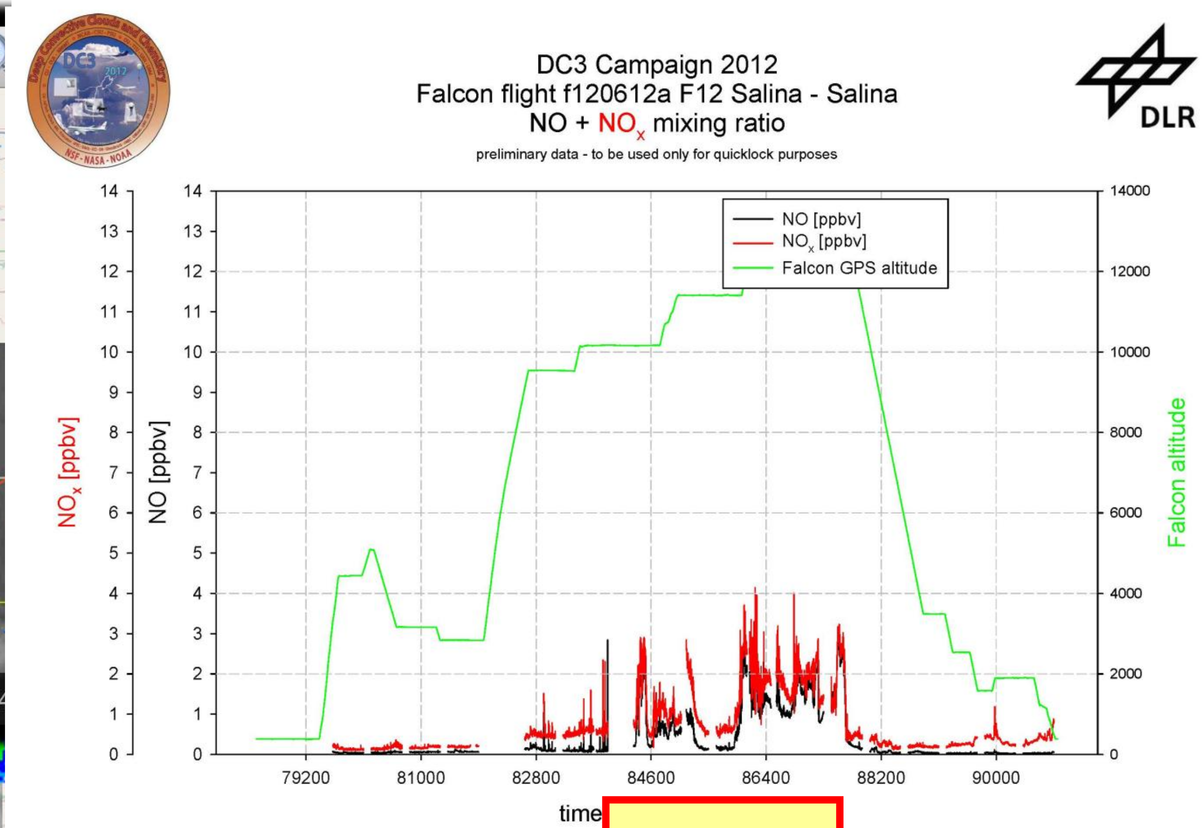
Flight A: 5 transects in anvil outflow ~10-12 km during ~1 h

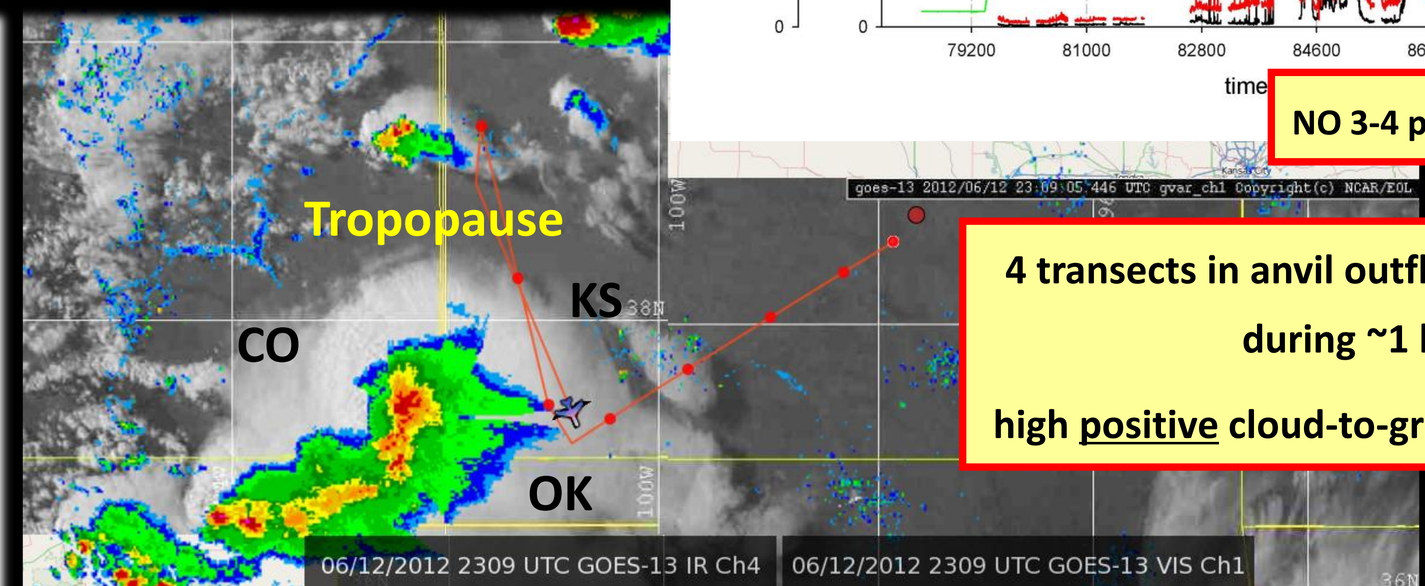
Flight B: 2 transects in anvil outflow ~11-12 km during ~0.5 h

- **high negative cloud-to-ground flash rate**

Falcon flight on 12 June 2012: Squall line SW Kansas







NO 3-4 ppbv

4 transects in anvil outflow ~10-12 km during ~1 h

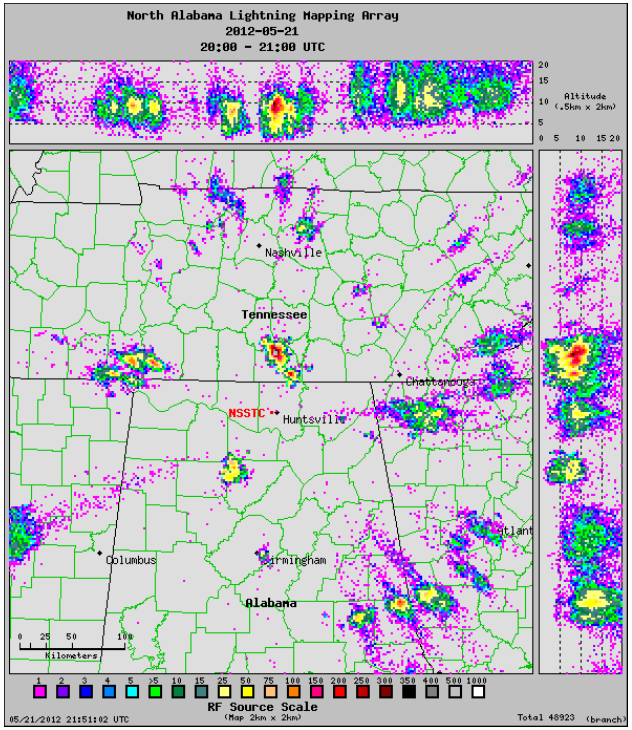
high <u>positive</u> cloud-to-ground flash rate

LNO_x Production Estimates from Cloud-Resolved Modeling Constrained by Lightning and Aircraft NO_x Observations

Ken Pickering, NASA/GSFC
Kristin Cummings, UMD
Yunyao Li, UMD
Megan Bela, NCAR
Mary Barth, NCAR

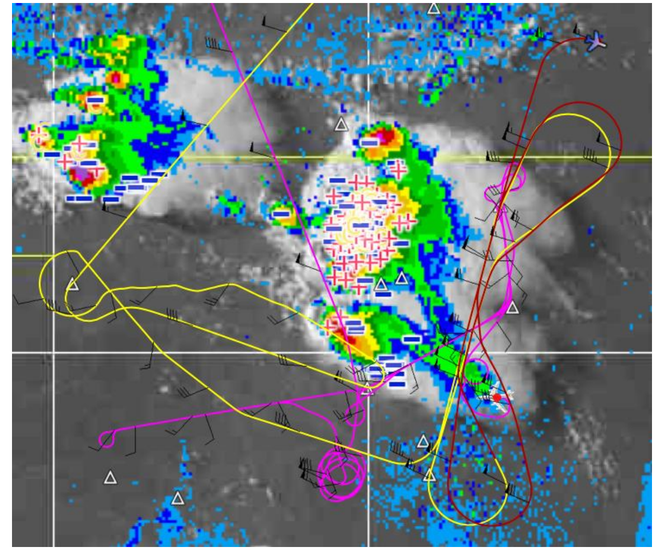
Potential DC3 Simulation Cases

Alabama
21 May 2012



- Sampling by GV and DC-8 aircraft
  - Isolated convection
- Coverage within dual-Doppler (ARMOR, MAX, and Hytop) and LMA regions
- Mobile soundings before and after cell formation

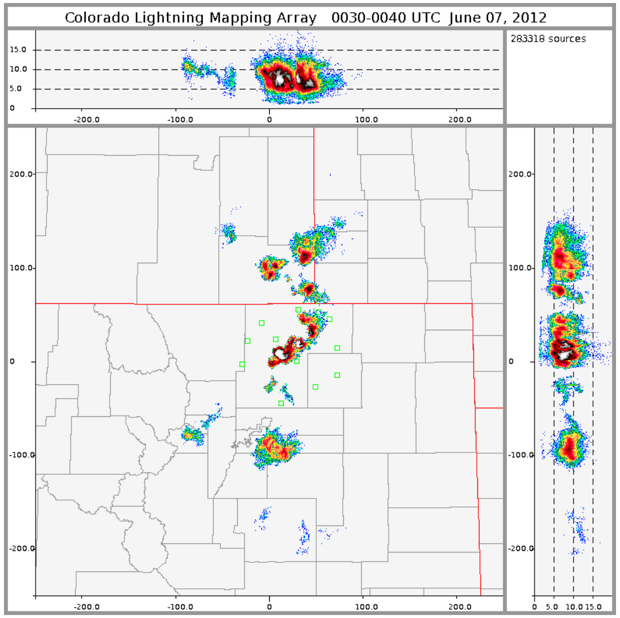
Oklahoma
29 May 2012



- Sampling of inflow and outflow by the GV, DC-8, and Falcon aircraft (two convective cells)
- High quality dual-Doppler radar data
- LMA coverage
- Environmental and EFM soundings

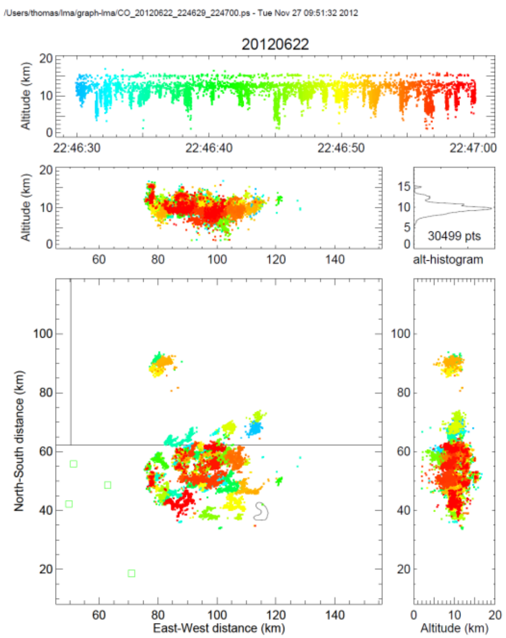
Potential DC3 Simulation Cases

Colorado
6 June 2012



- Sampling of inflow and outflow by the GV and DC-8 aircraft was comprehensive
  - Isolated cell
  - Squall line
- Coverage within dual-Doppler and LMA regions
- Environmental and EFM soundings

Colorado
22 June 2012



- Excellent sampling by both DC-8 and G-V
- Best radar coverage for early part of storm
- Smoke plume from High Park fire
- 7 environmental soundings

Flash Rate Parameterization Schemes used in 3-km Resolution WRF Forecasts during DC3

Type of FRPS	Equation (*flashes min^{-1}*)
• Updraft volume*	$f = 6.75 \times 10^{-11} w_5 - 13.9$
• Maximum vertical velocity	$f = 5.7 \times 10^{-6} \times w_{max}^{4.5}$
• Cloud top height	$f = 3.44 \times 10^{-5} H^{4.9}$

f = total flash rate*;* IC/CG ratios based on *Boccippio et al. (2001)*

* Used in LNOx calculations

These and other schemes will be run in cloud-resolved 3-km resolution WRF simulations of selected DC3 storms and tested against LMA flash counts

Treatment of LNO_x in Cloud-Resolved WRF-Chem

- LNO_x parameterization scheme *(DeCaria et al., 2005)*
  - Gaussian vertical distributions of IC (bimodal) and CG (single mode) NO production based on typical lightning flash channel distributions
  - Lightning channels set to maximize at -15°C (CG and IC) and -45°C (IC)
  - 500 moles NO per IC and CG flash *(Ott et al., 2010)*
  - Horizontal placement of NO based on reflectivity ≥ 20 dBZ

- LNO mixing ratios simulated in post-mission WRF-Chem
  - Sum of IC and CG LNO produced at each model time step injected into grid cells as designated above.
  - After evaluating model storm evolution and characteristics against radar data and chemical initial conditions against aircraft observations:

  Evaluate convective transport in the model simulation using tracers

  Compare model and aircraft-observed NO_x mixing ratios at various altitudes within the cloud to determine if the assumed mean LNO_x moles NO $flash^{-1}$ for IC and CG flashes in the model is over or underestimated. Test additional values as necessary.

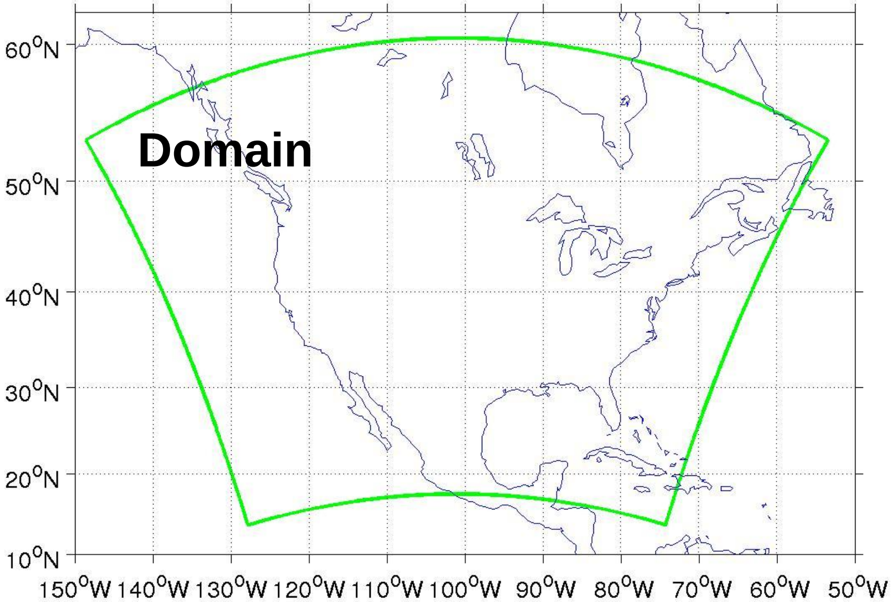
Potential Improvements to the LNO_x Scheme in WRF-Chem

- Evaluate model flash rates based on LMA flash counts; compare LMA rates with adjusted NLDN data
- Identify flash rate parameterization schemes that perform best for specific DC3 regions and storm types
- Use LMA data to:
  - Improve vertical distribution of LNO_x production based on LMA data (vertical distribution of flash segments)
  - Modify region within storm where LNO_x is placed (flash extent information)
  - Use IC/CG ratio from LMA data
  - Modify model to use flash length data rather than flash counts

LNOx Estimates from Regional and Global Models

Mary Barth, Megan Bela, Louisa Emmons, Frank Flocke

WRF-Chem Setup



Grid spacing: dx = 15 km, 40 vertical levels to 50 hPa (~650 m in UT)

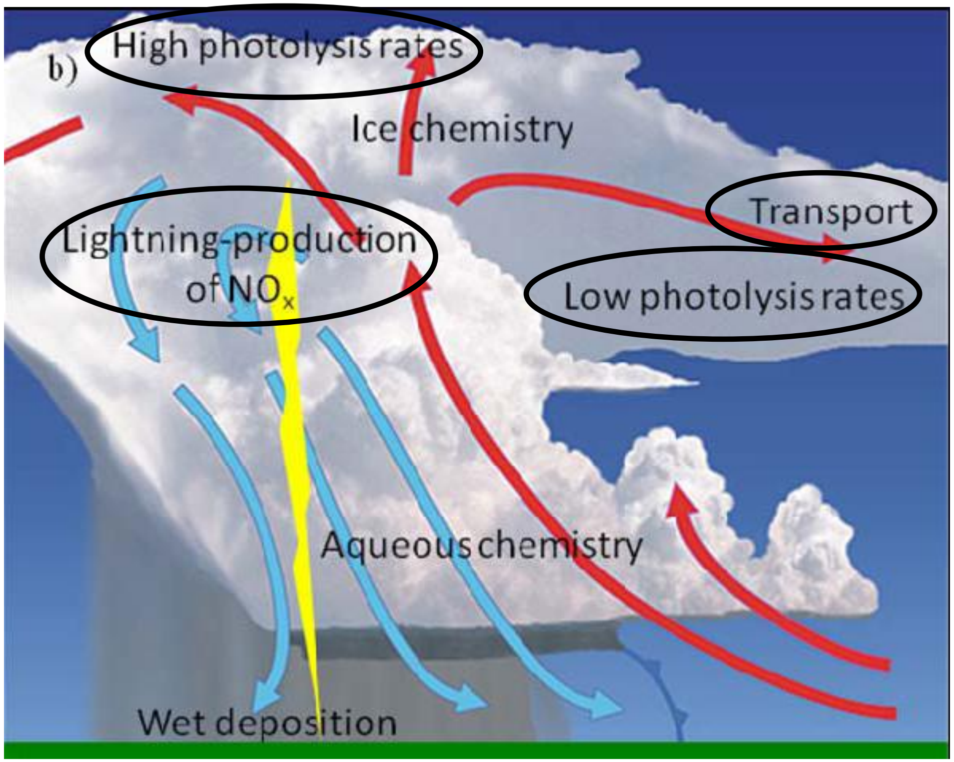
Initial/Boundary Conditions: DART (met), MOZART (chem)

Physics: Grell 3D convection, Morrison cloud microphysics, MYJ PBL

Chemistry: MOZART gas chemistry mechanism; GOCART aerosol scheme

Emissions: EPA NEI 2005 anthropogenic (2012 NO/NO2 based on OMI NO2), aircraft from Baughcum (1999), MEGAN v2.0.4 biogenic, FINN fire

Included processes:

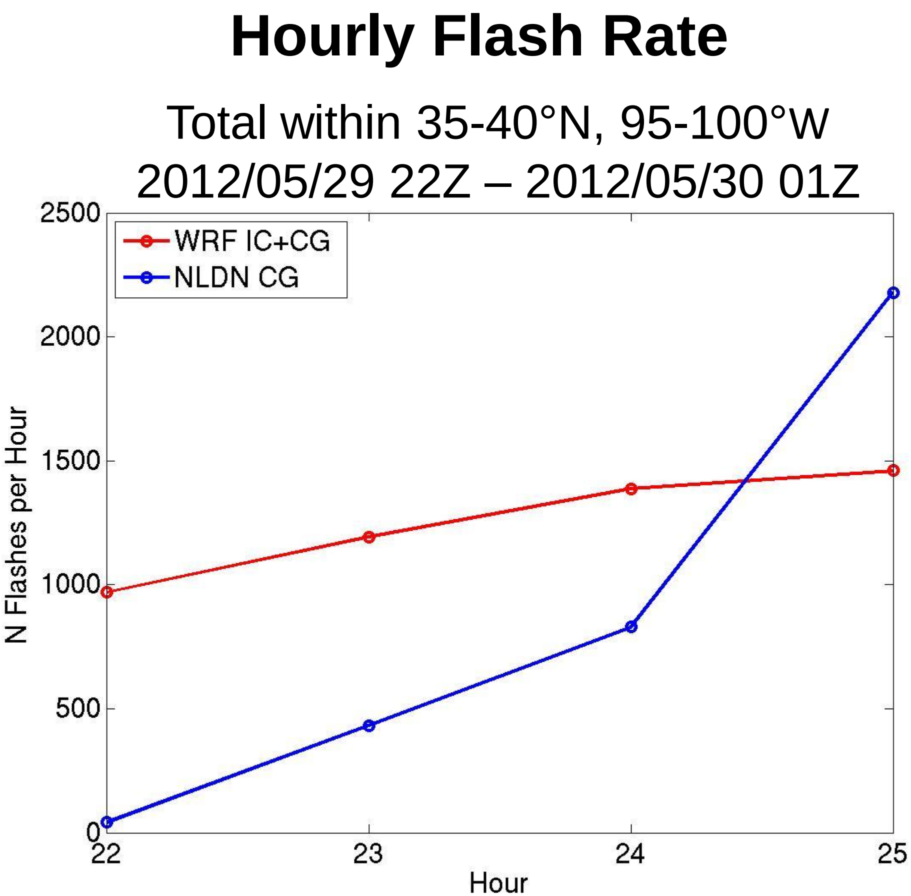


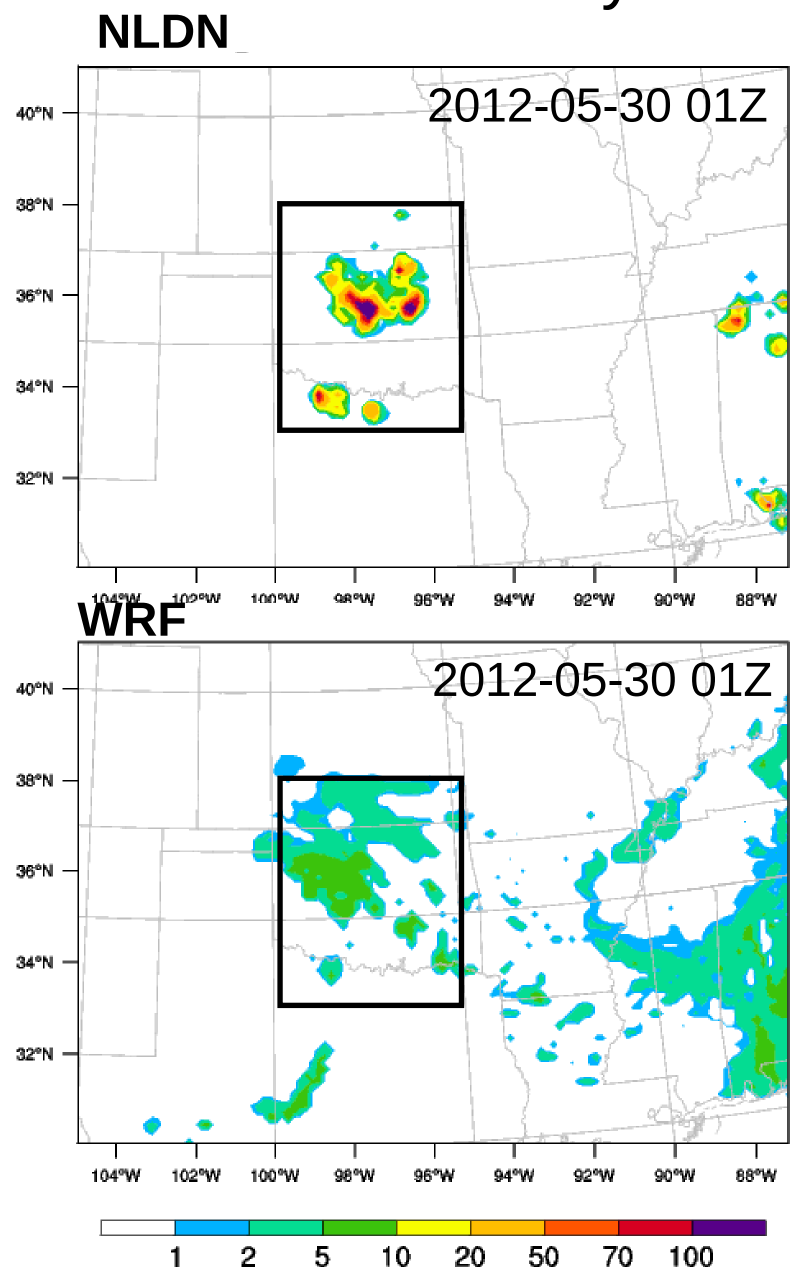
Lightning-NOx : $FR = 3.44 \times 10^{-5}\, z_{top}^{4.9}$

z_{top} = cloud top height = level neutral buoyancy – 2 km (Wong et al., 2012)

500 moles NO/flash placed vertically following Ott et al. (2010) curves

WRF-Chem overpredicts total hourly flash rate in storm region compared to NLDN, but underestimates intensity



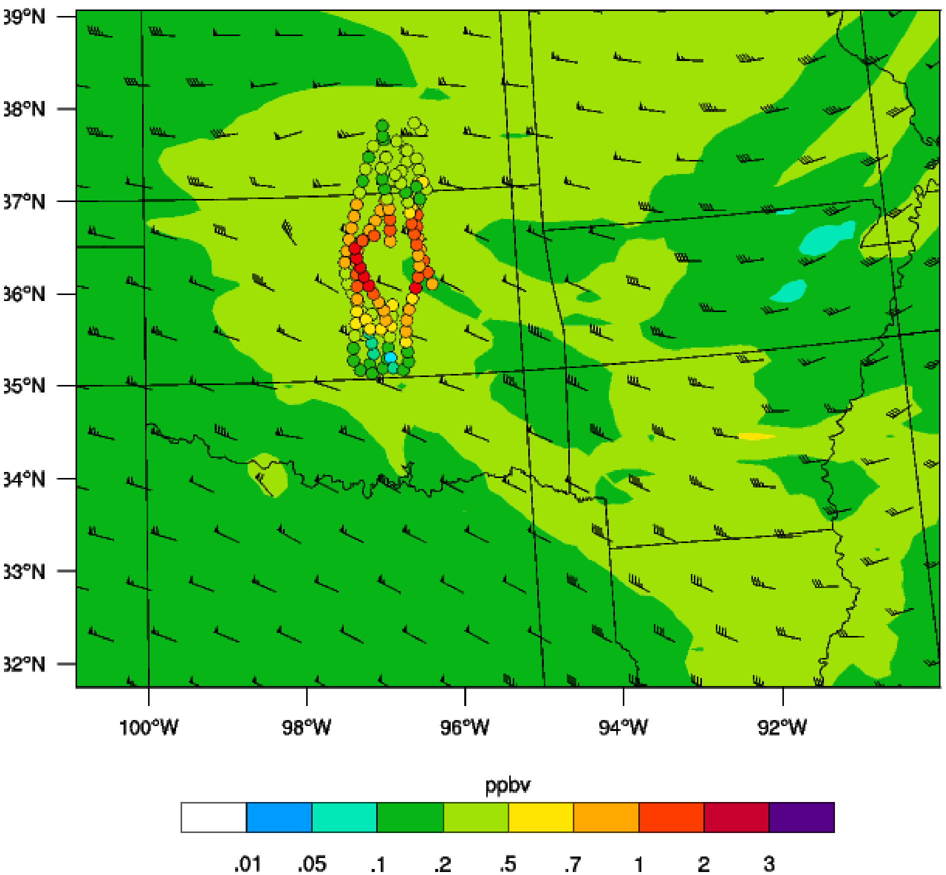


Note: NLDN data includes Cloud-to-Ground (CG) flashes only, while WRF data is CG plus Intracloud (IC)

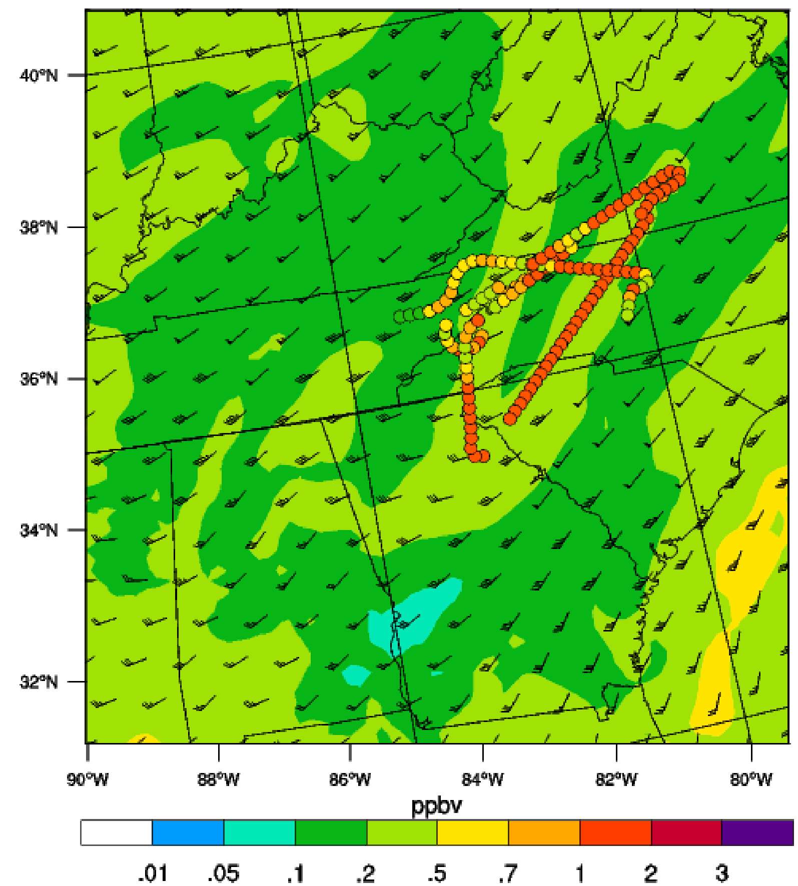
WRF-Chem NO_x values at 11km are too low by a factor of ten compared with DC8 and GV observations for May 29 storm and May 30 downwind flights

NOx (ppbv) simulated by WRF-Chem at 11km and observed by DC8 and GV for 10<z<12km

2012-05-30 0Z



2012-05-30 22Z



Large-scale impact of lightning NOx on the UT over the US

Frank Flocke, Louisa Emmons – NCAR/ACD

Hypothesis:

T-storms firing off the Rockies and moving E, (some becoming MCSs), and T-storms firing over the plains and moving E, should increase upper trop. NOx over the Eastern US compared to UT NOx over Western US.

Analysis:

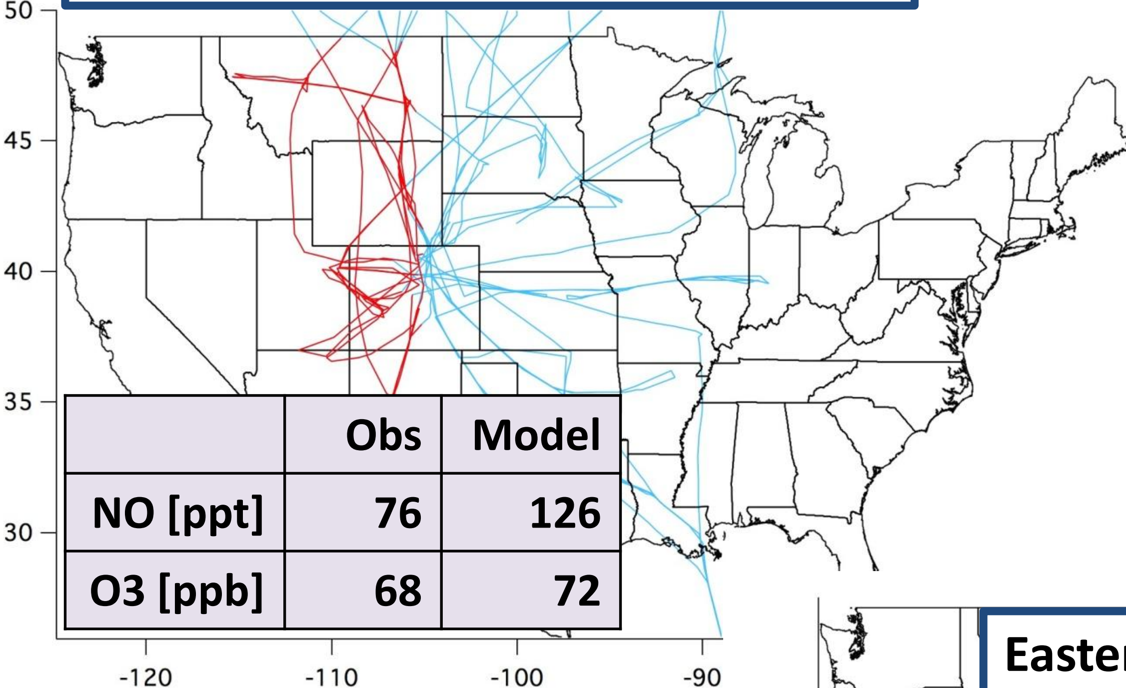
Compare regional averages from observations over Western and Eastern US with regional model averages.

Model:

CAM-chem (NCAR Community Atmosphere Model with MOZART-4 chemistry) driven by GEOS-5.

Lightning NO emissions: 3 Tg-N/yr

Western US: START-08 (June 2008)



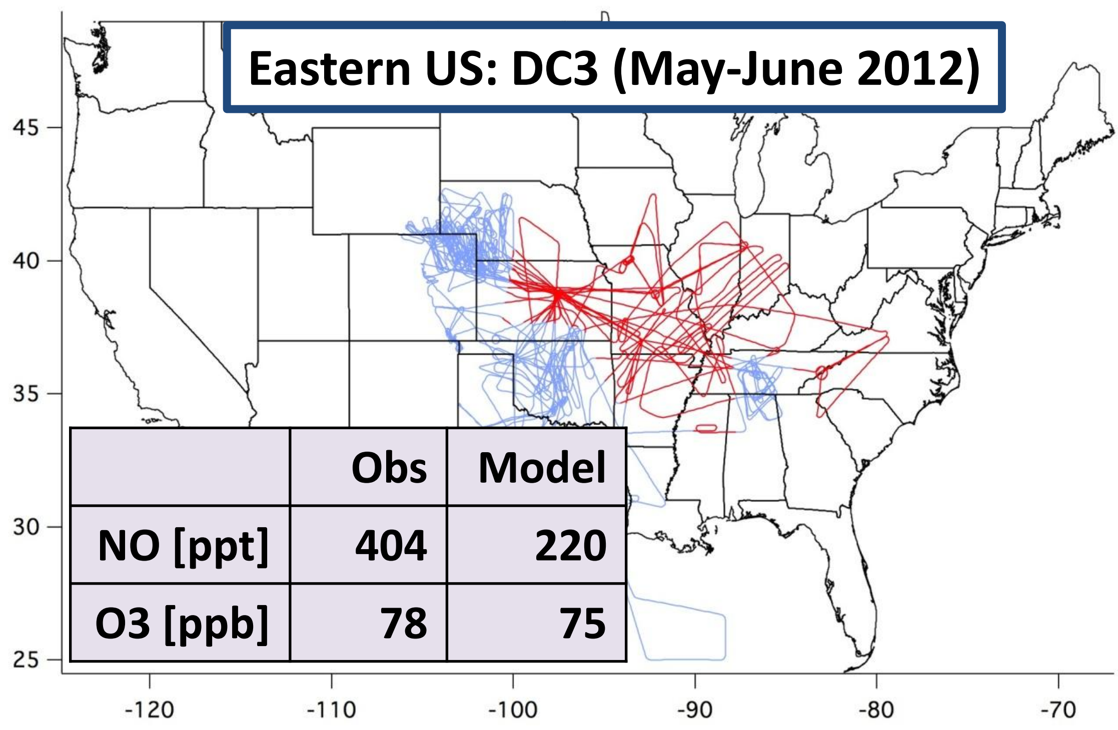
	Obs	Model
NO [ppt]	76	126
O3 [ppb]	68	72

Red tracks mark flight data used for each set of averages

Further restrictions: 30-40 kft, O_3 < 100 ppb

CAM-chem model results averaged over same regions, daytime only

Eastern US: DC3 (May-June 2012)



	Obs	Model
NO [ppt]	404	220
O3 [ppb]	78	75

Conclusions:

- Both model and obs show higher NO and O_3 in UT over eastern than western U.S.
- Model under-predicts E-W enhancement, perhaps due to low lightning NO
- Approach may be useful for model evaluation

Estimates of Lightning NO_x Production from OMI NO_2 Observations During DC3

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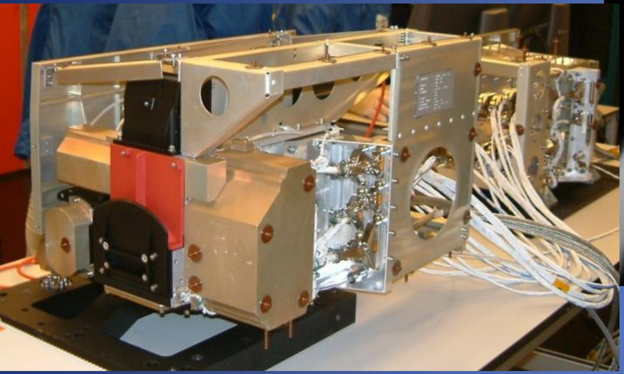
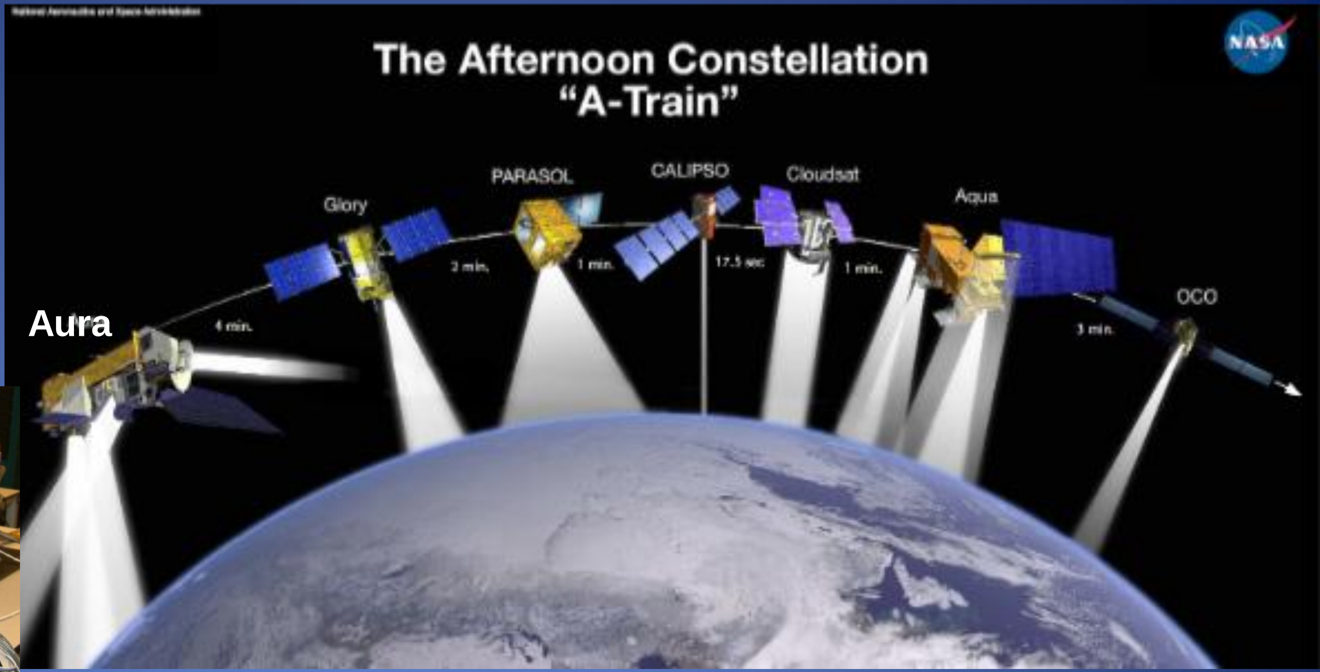
Yunyao Li, UMD/AOSC
Lok Lamsal, GESTAR/GSFC
Ed Celarier, GESTAR/GSFC
Bill Swartz, JHU/APL
Nick Krotkov, NASA/GSFC

Aura/OMI

Ozone Monitoring Instrument





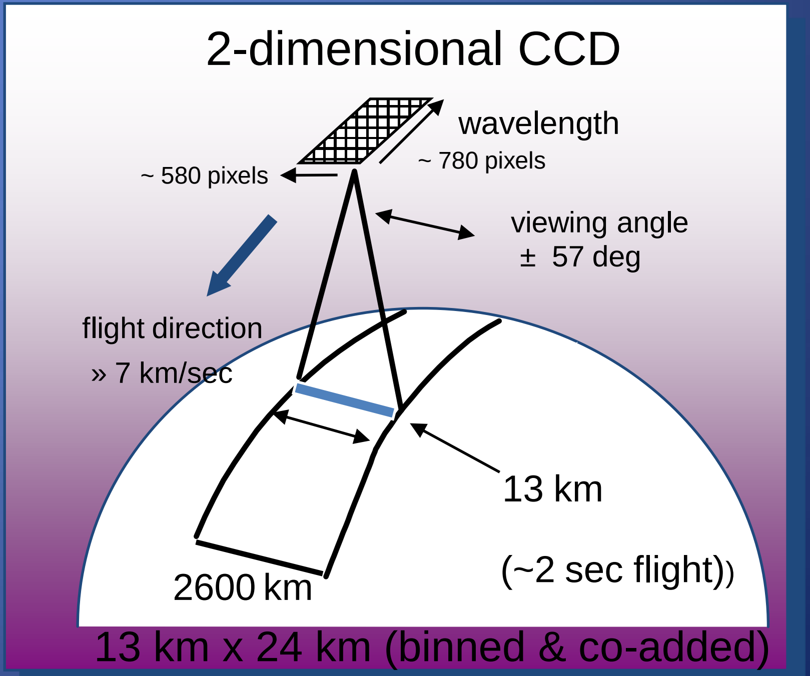
Wavelength range: 270 – 500 nm

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

O_3, NO_2, SO_2, HCHO, aerosol,
BrO, OClO



Lightning NO_2 retrieval algorithm developed for DC3

$$\beta = \frac{\Omega_{NoL}^{GMI}}{\Omega_{L}^{GMI}}$$

GMI CTM simulations

$$\Omega_{NoL}^{OMI} = \beta \times \Omega_{clim}^{OMI}$$

Based on OMI tropospheric NO2 from case day ±3 days

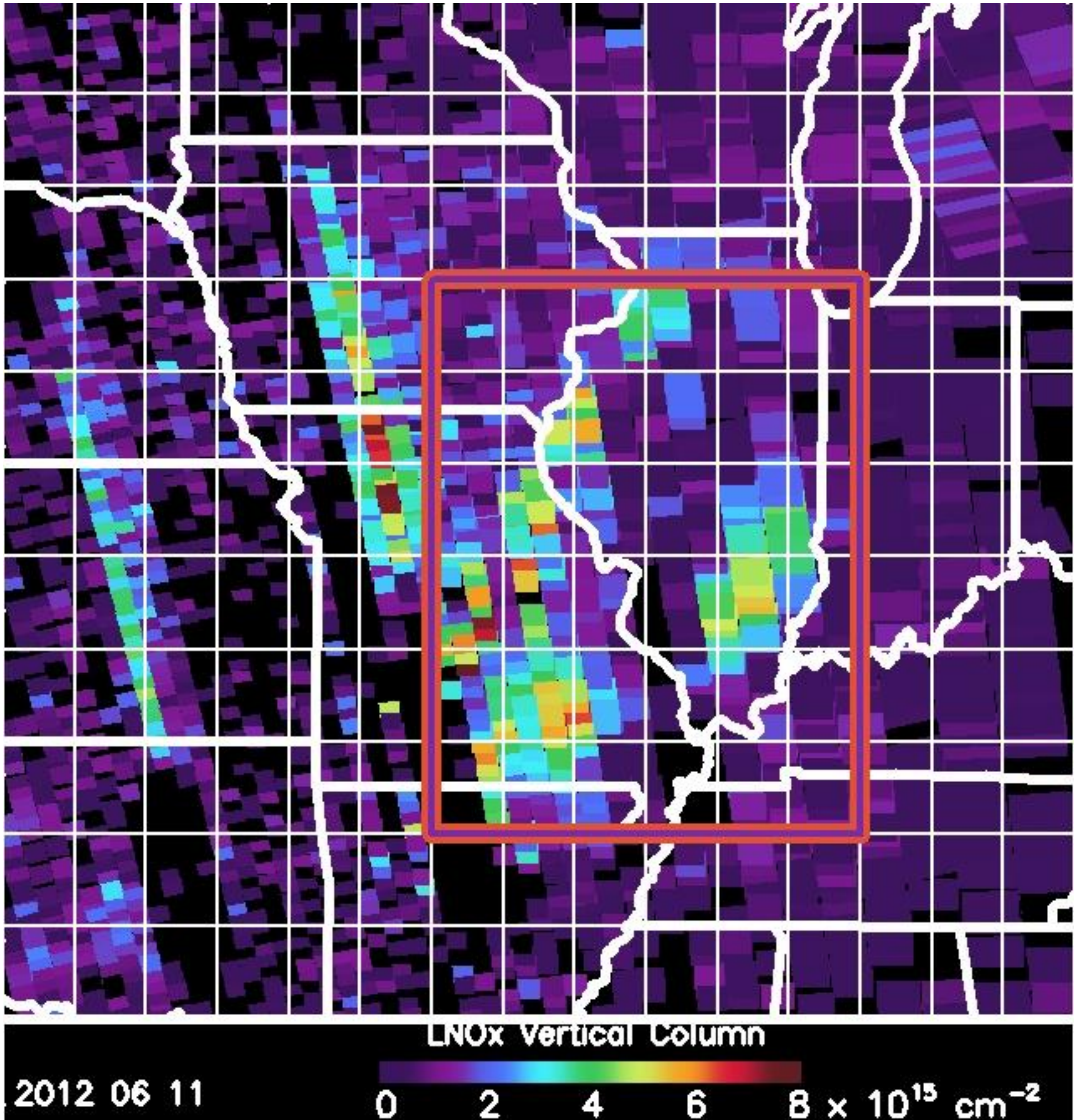
$$\Omega_{LNO2} = \frac{\Omega_{total}^{slant} - \Omega_{strat} \times AMF_{strat} - \Omega_{NoL}^{OMI} \times AMF_{trop}}{AMF_{LNO2}}$$

AMF_{trop} - using NO2 profile shapes based on GMI no-lightning simulation
AMF_{LNO2} - using NO2 profile shape representative of convective outflow

Convert LNO2 to LNOx using upper tropospheric NO_x/NO_2 ratios from the DC-8 and G-V observations

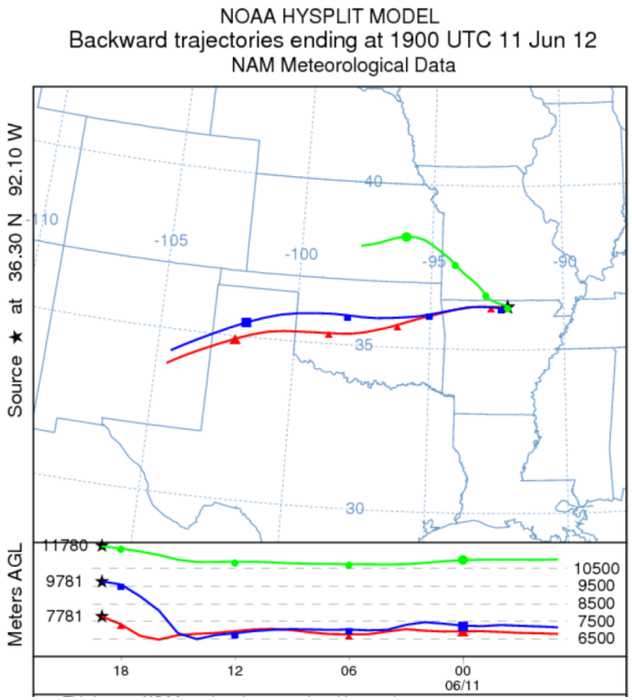
Account for LNO2 loss during transport from storms to location at overpass time using exponential decay with assumed UT lifetime of 2 days. Transport time estimated from back trajectories.

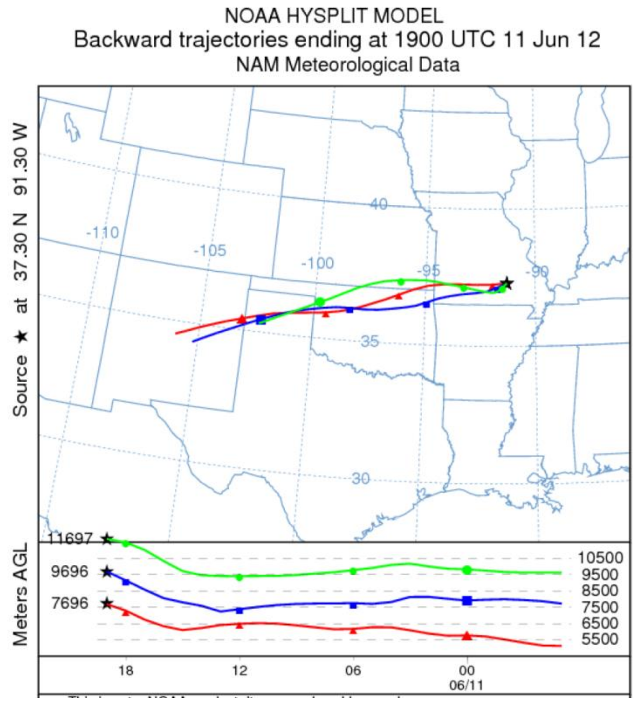
Case 1: June 11, 2012 – Active Mesoscale Convective System

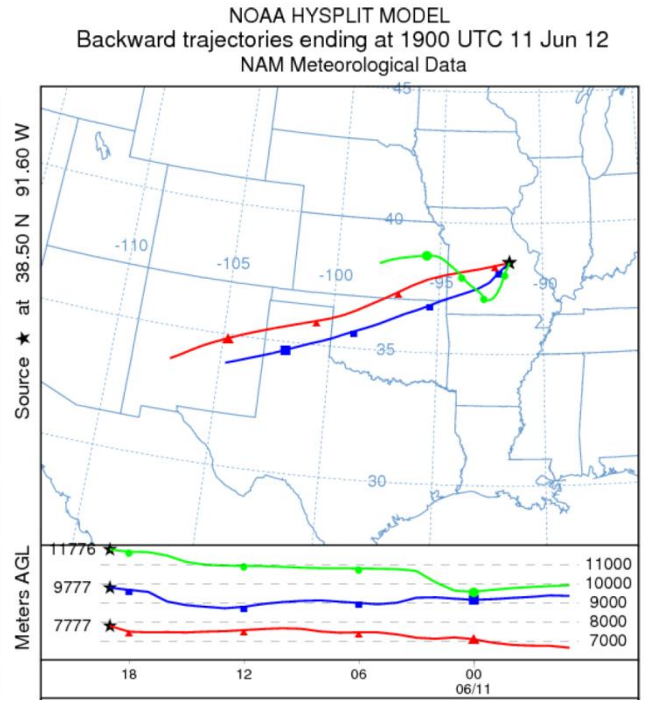


6.989 Mmoles LNOx within box

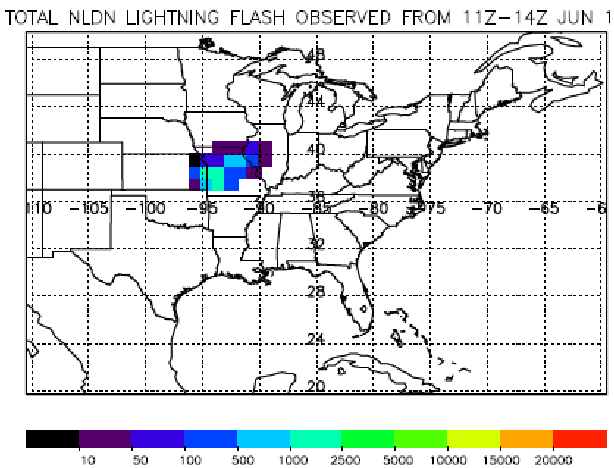
8.963 Mmoles LNOx after considering loss during transport

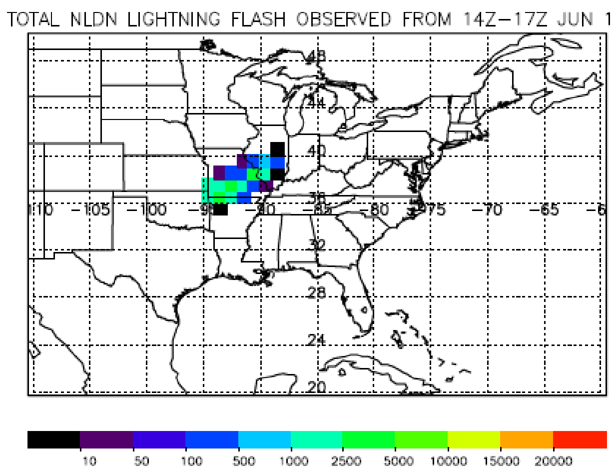


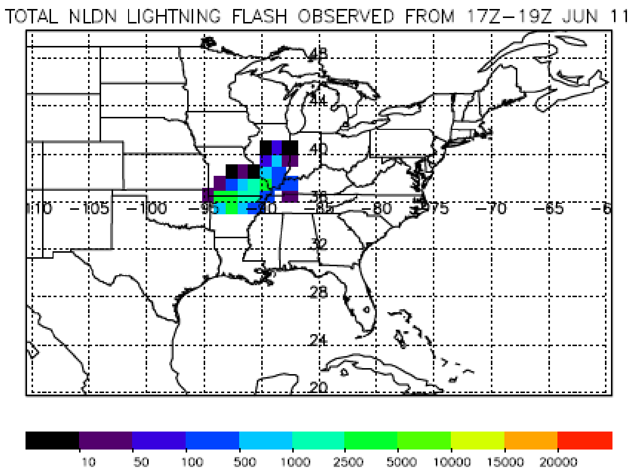




Back trajectories suggest transport times of 0 – 8 hours; mean = 4.7 hours



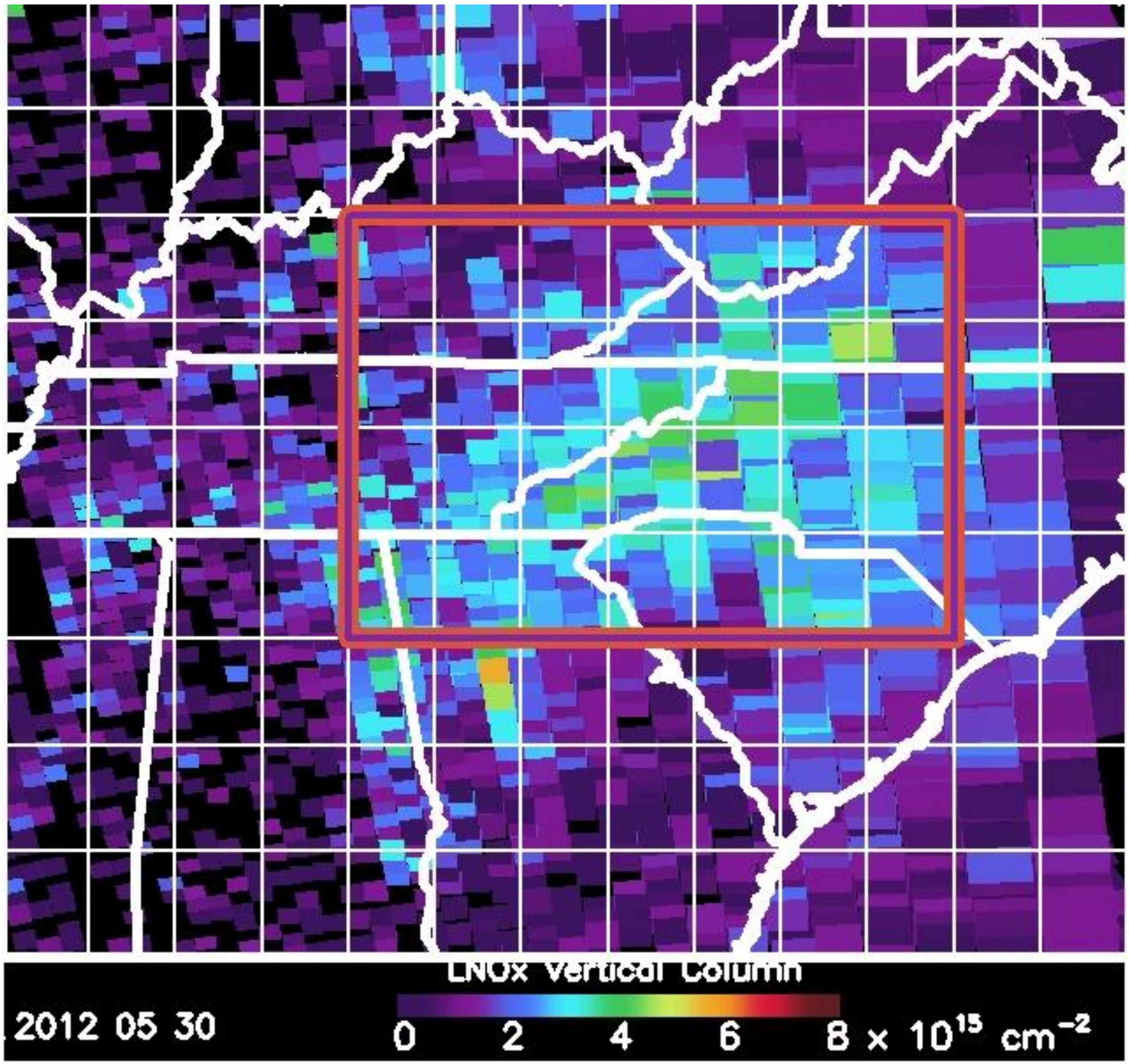




Total contributing flashes = 27384 +70483 + 48436 = 146,303

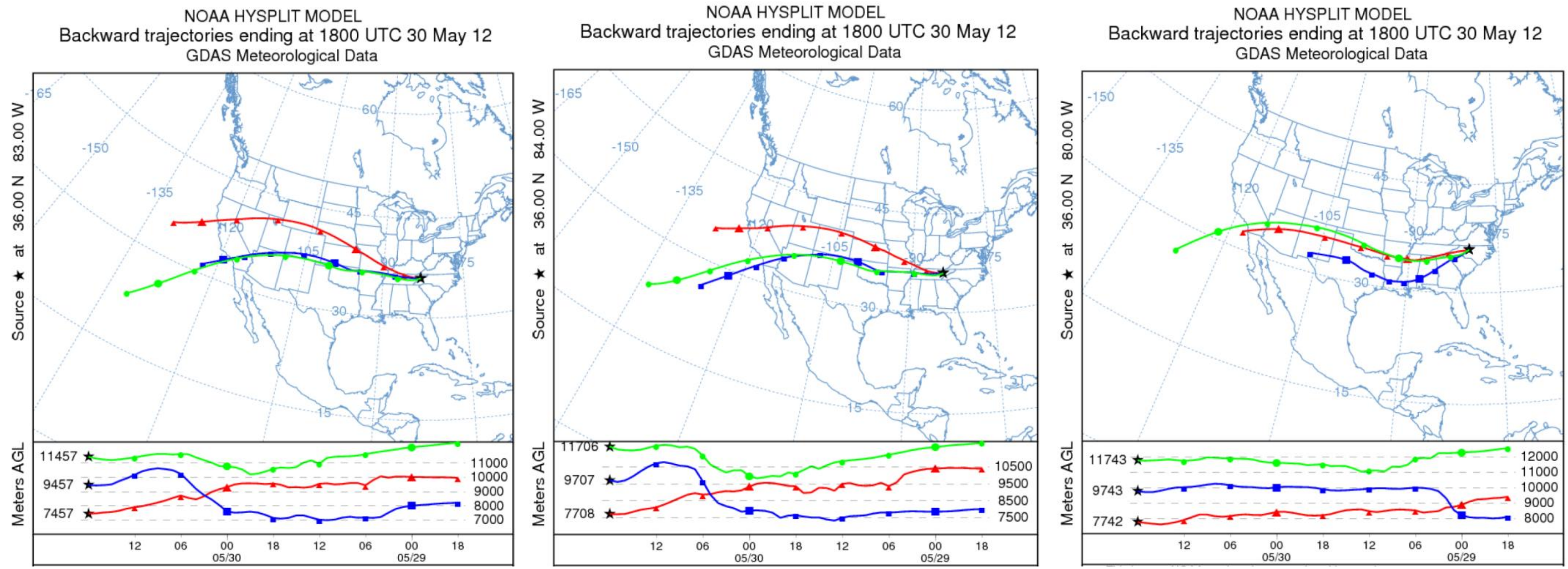
8.96×10^6 moles/146303 flashes = 61.3 moles LNOx/flash

Case 2: May 30, 2012 – Downwind of previous day's OK convection

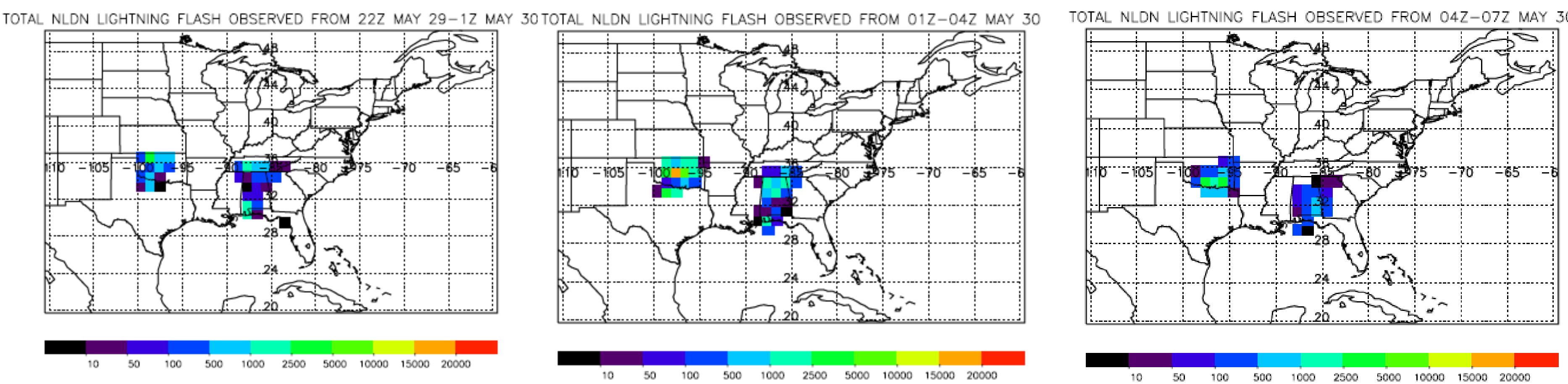


7.449 Mmoles LNOx within box

10.823 Mmoles LNOx after considering loss during transport



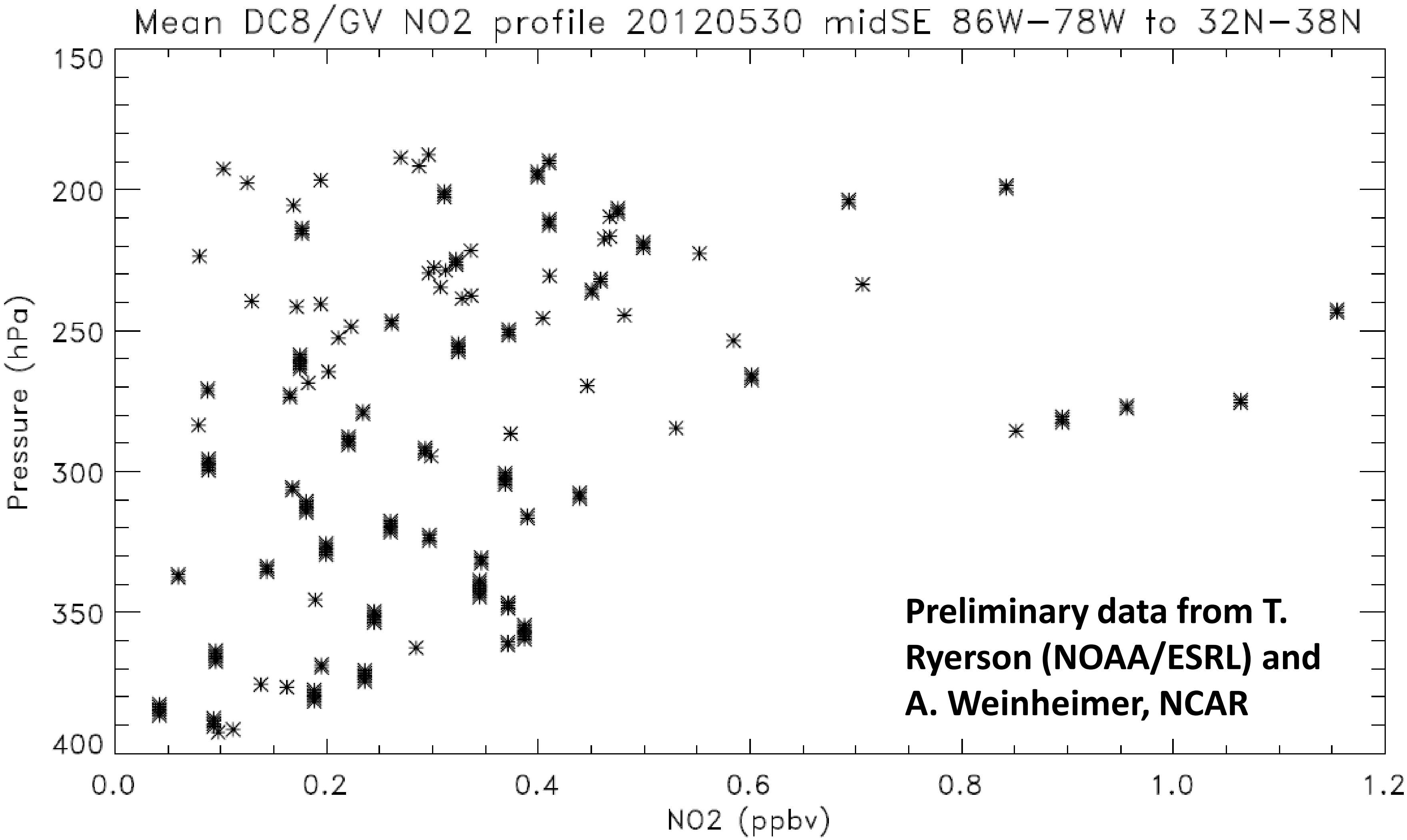
Back trajectories suggest transport times of 9 – 18 hours; mean = 16.1 hours



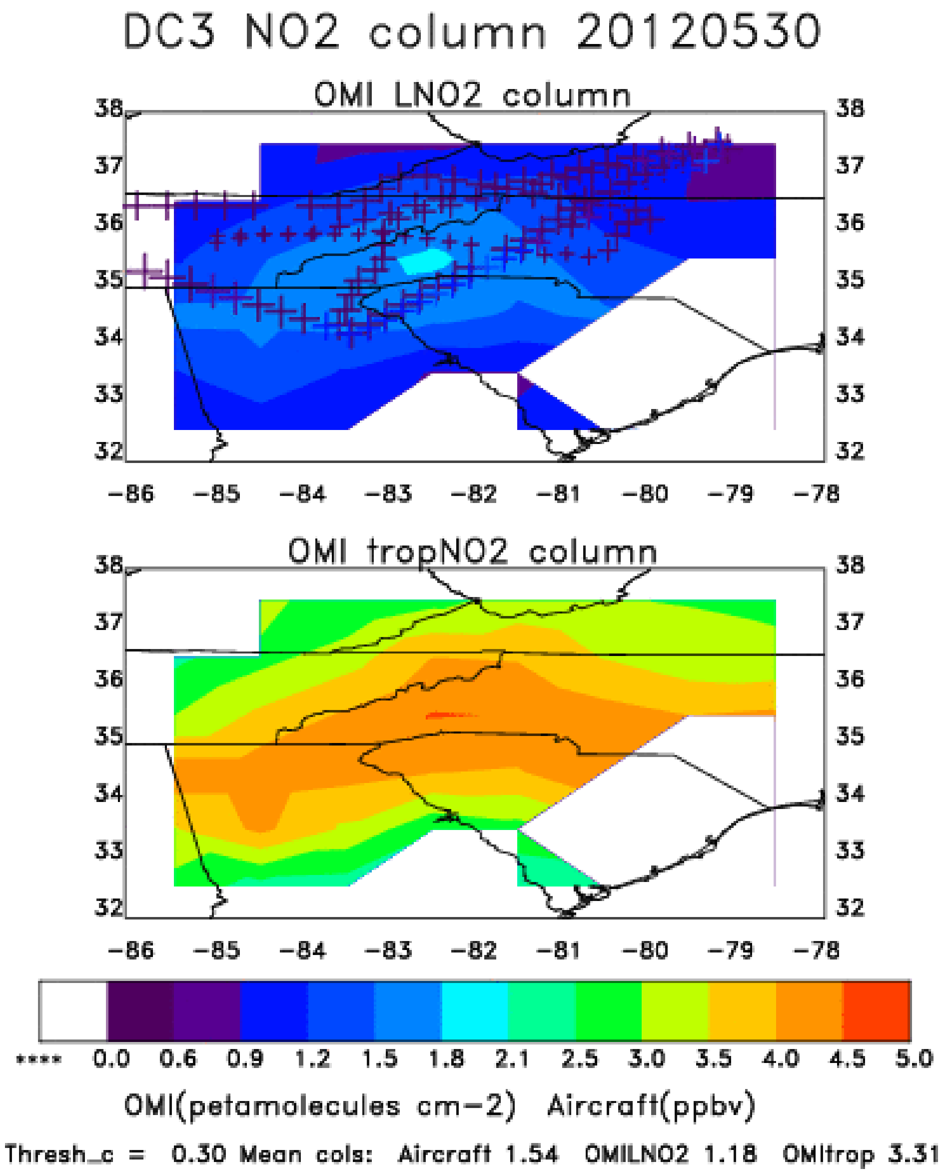
Total contributing flashes = 47626 + 84255 + 50772 = 182,653

1,08 x 10^7 moles/182,653flashes = 59.3 moles LNOx/flash

In-situ NO_2 Observations by NASA DC8 and NCAR G-V



DC-8 data binned into 1 hPa intervals and vertically integrated yielding 1.40×10^{15} molec cm^{-2}



Mean OMI LNO2 column over enhanced region = 1.18×10^{15} molec cm^{-2}

Possible 16% low bias compared with DC-8 observations

Adjusting LNOx production estimates:

Case 1: 61 moles/flash → 70

Case 2: 59 moles/flash → 68

36 – 47% of trop NO2 column in case study region due to upwind lightning

Summary

- DC3 experiment provided excellent facilities in three regions of the US to obtain the necessary data for estimating LNO_x production:
  - Lightning Mapping Arrays
  - Dual-doppler and polarimetric radars
  - Aircraft chemical obs. in storm inflow and anvil outflow
- LMA data products being archived: initially flash count & flash extent; ultimately flash channel lengths and vertical distribution of channel segments
- Analysis of aircraft NO_x data in relation to flash rates and WRF-Chem modeling of flash rates & LNO_x production are underway
- Regional and global models underestimating NOx when “standard “ LNOx formulations are used; cloud-resolved simulations just getting underway.
- Preliminary OMI-based estimates for two DC3 cases are lower (~70 moles/flash) than previous estimates (~500 moles/flash) over the continental US and Gulf of Mexico, but within the range of literature values.

Initial List of Issues for Breakout Discussion

- Refinements of techniques for LNOx estimates from aircraft observations
- Refinements of LNOx schemes in models
- Why are OMI based estimates coming out relatively low?
- How does mean LNOx production per flash vary from storm to storm, one region to another?
- Is LNOx production different in anomalous polarity storms vs. normal polarity storms