2006 Fall IMAQS Group Seminar

September 29th, 2006

HARC Project H60 Final Report

Regional Air Pollution Transport Modeling:

Task: CMAQ HDDM Simulations

Daewon Byun, Soontae Kim, Fang-Yi Cheng, Beata Czader,
In-Bo Oh, Meongdo Jang, Chang-Keun Song, Hyun-Cheol Kim,
Peter Percell, Fong Ngan, Violeta Coarfa,
Daniel Cohan*

Institute for Multi-dimensional Air Quality Studies
University of Houston

***Department of Civil & Environmental Engineering**
Rice University

Outline

- Emissions Preparation for 2005 episodes (5 slides, 7 minutes)
- Base Simulations with CAMx and CMAQ with CB4 & SAPRC99 (6 slides, 8 minutes)
- CMAQ HDDM Description
- HDDM Simulations
  - Contribution of NOx and VOC emissions from four neighboring states to DFW ozone concentrations (First main topic in the presentation)
  - Source apportionment and sensitivity of NOx and VOC emissions over HGB 8-county area (Second main topic in the presentation)
  - Contribution of VOC emissions to HCHO concentrations (Examples)
  - Contribution of biogenic emissions to Houston ozone concentrations (Examples)

Outline

- **Emissions Preparation for 2005 episodes**
- Base Simulations with CAMx and CMAQ
- CMAQ HDDM Description
- HDDM Simulations
  - Contribution of NOx and VOC emissions from four neighboring states to DFW ozone concentrations
  - Source apportionment and sensitivity of NOx and VOC emissions over HGB 8-county area
  - Contribution of VOC emissions to HCHO concentrations
  - Contribution of biogenic emissions to Houston ozone concentrations

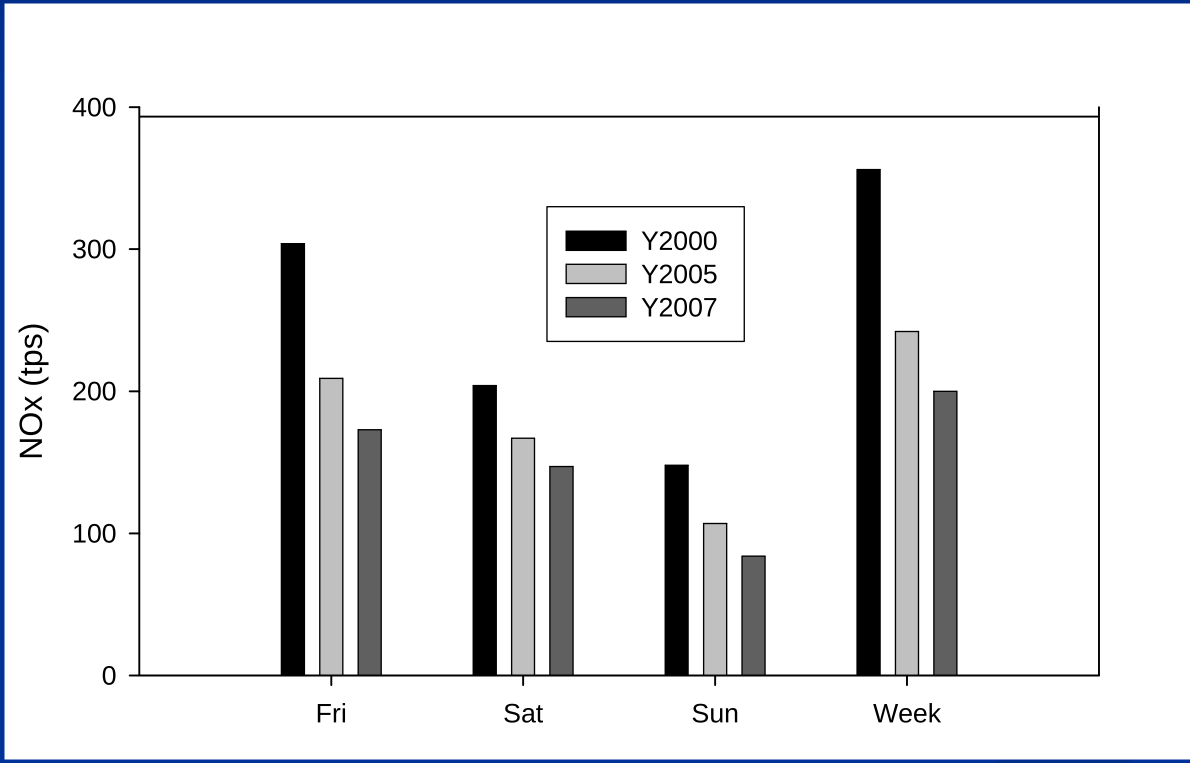
Mobile Source

	Category	Description	Data	EI year
1	Texas HPMS MOBILE6 emissions	County-based emissions for Texas counties excluding HGB 8 counties and DFW 9 counties	TCEQ	2000, 2007
2	HGB Link MOBILE6 emissions	Link-based emissions for HGB 8 counties; Brazoria, Chambers, Fort Bend, Galveston, Harris, Liberty, Montgomery, Waller	TCEQ	2000, 2007
3	DFW HPMS MOBILE6 emissions	County-based emissions for DFW 4 counties: Ellis, Johnson, Kaufman, Parker	NCTCOG	2005
4	DFW Link MOBILE6 emissions	Link-based emissions for DFW 5 counties; Collin, Dallas, Denton, Rockwall, Tarrant	NCTCOG	2005
5	NEI99	County-based emissions inventory for outside Texas and PM species	U.S. EPA	1999

The various component mobile source emissions inventories

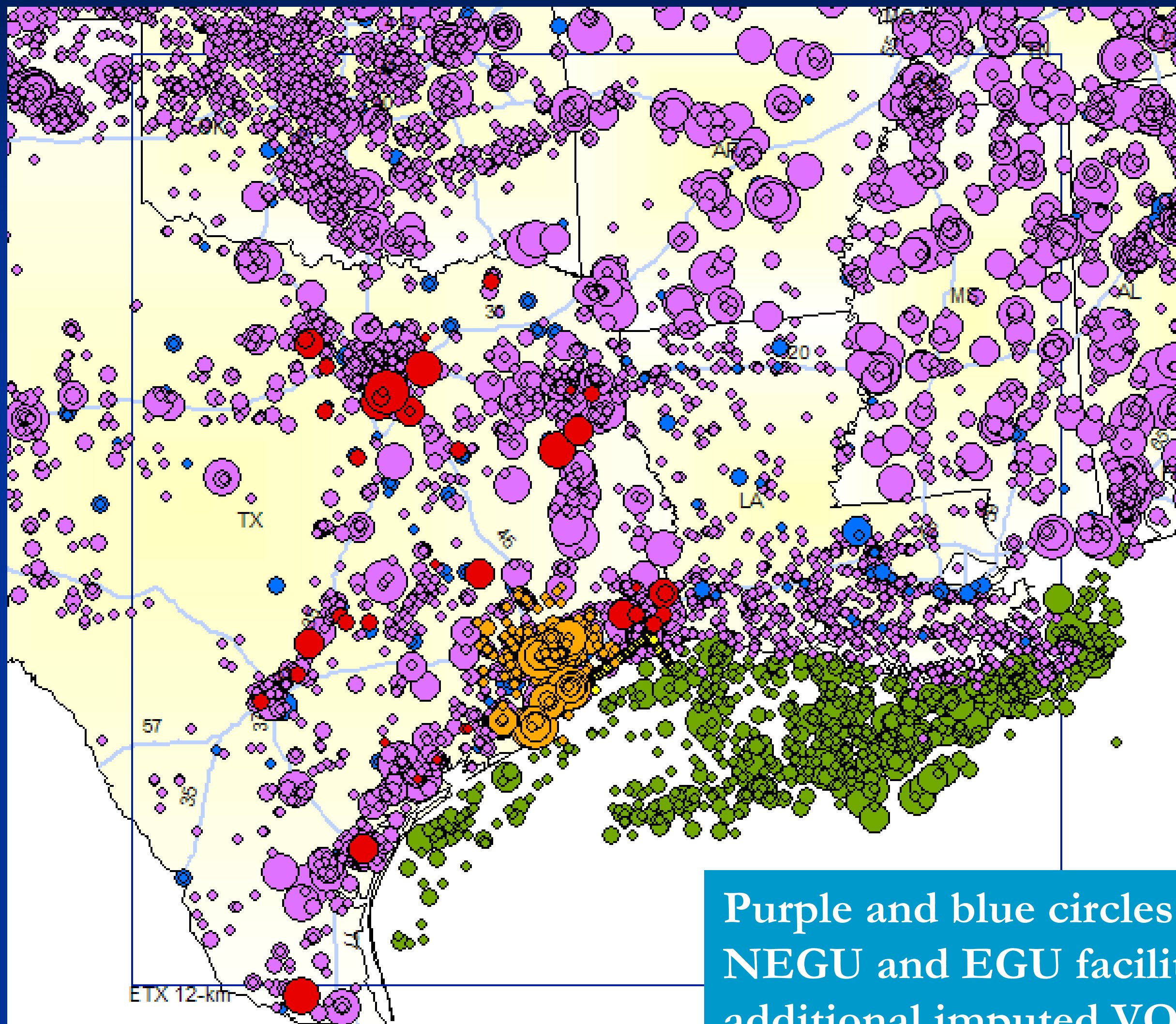
Mobile Source

NOx emissions rates from HGB 8 counties for different days of the week



Compared to 2000, NOx emissions in 2005 decreased by ~ 30% but were still higher than those in 2007.

Point Source

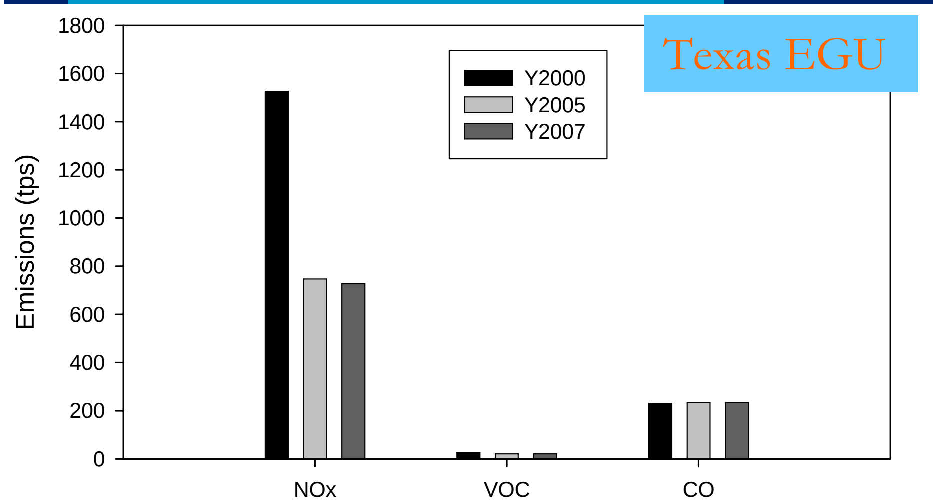


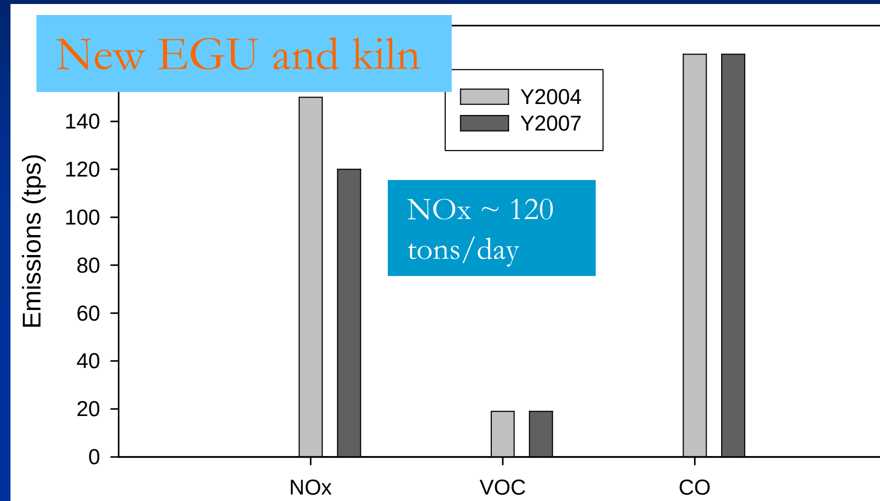
Purple and blue circles represent NEGU and EGU facilities; Orange for additional imputed VOC emitter, green for offshore sources, and red for new EGU and kiln (2004) point sources

Point Source

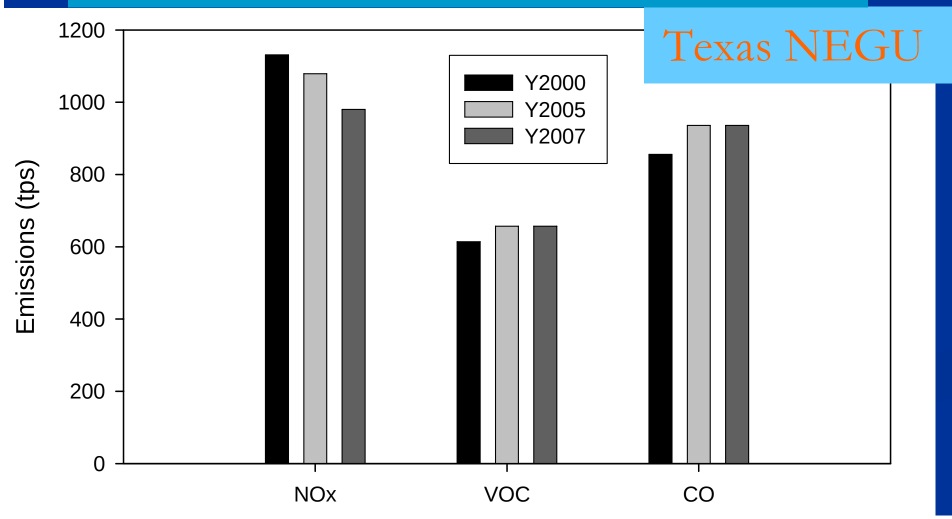
Growth and control factors from TCEQ:
http://www.tceq.state.tx.us/assets/public/implementation/air/sip/sipdocs/2005-09-BPA/ado_BPA_D.pdf

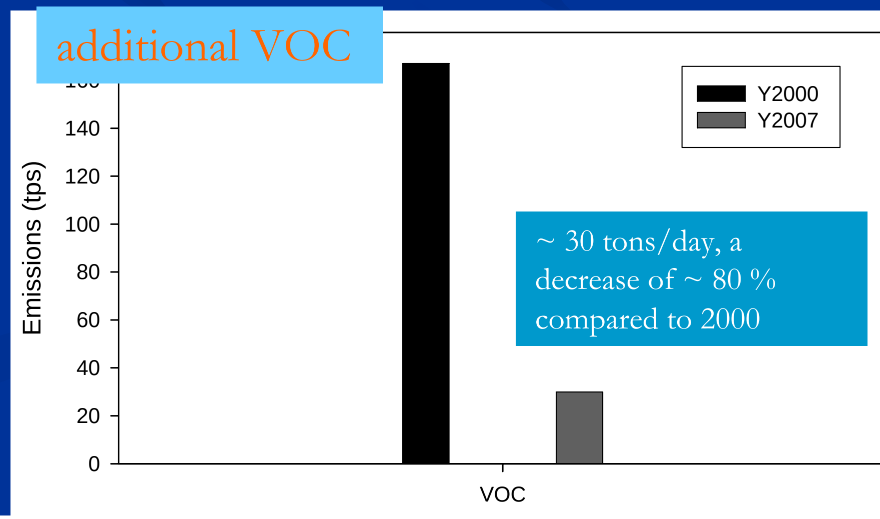
NOx emissions from Texas EGU facilities were reduced by ~ 700 tons/day compared to 2000



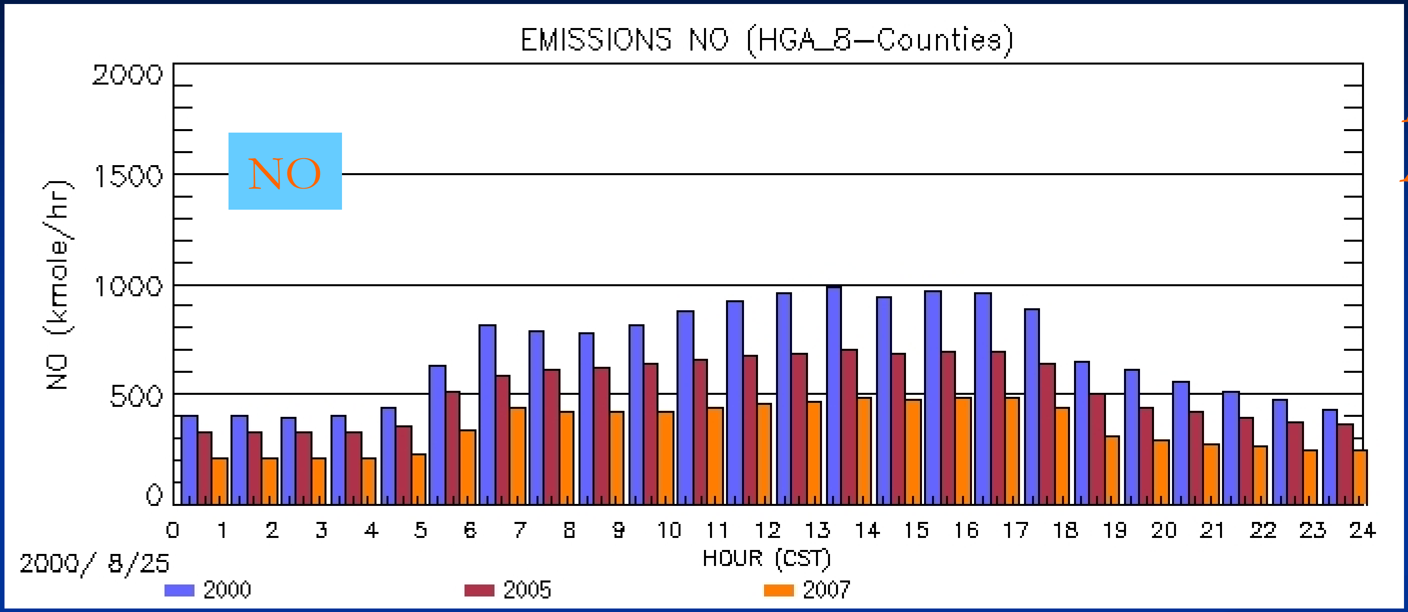


NOx emissions in 2005 reduced by ~ 50 tons/day, and VOC and CO emissions increased by ~ 10 %, compared to 2000

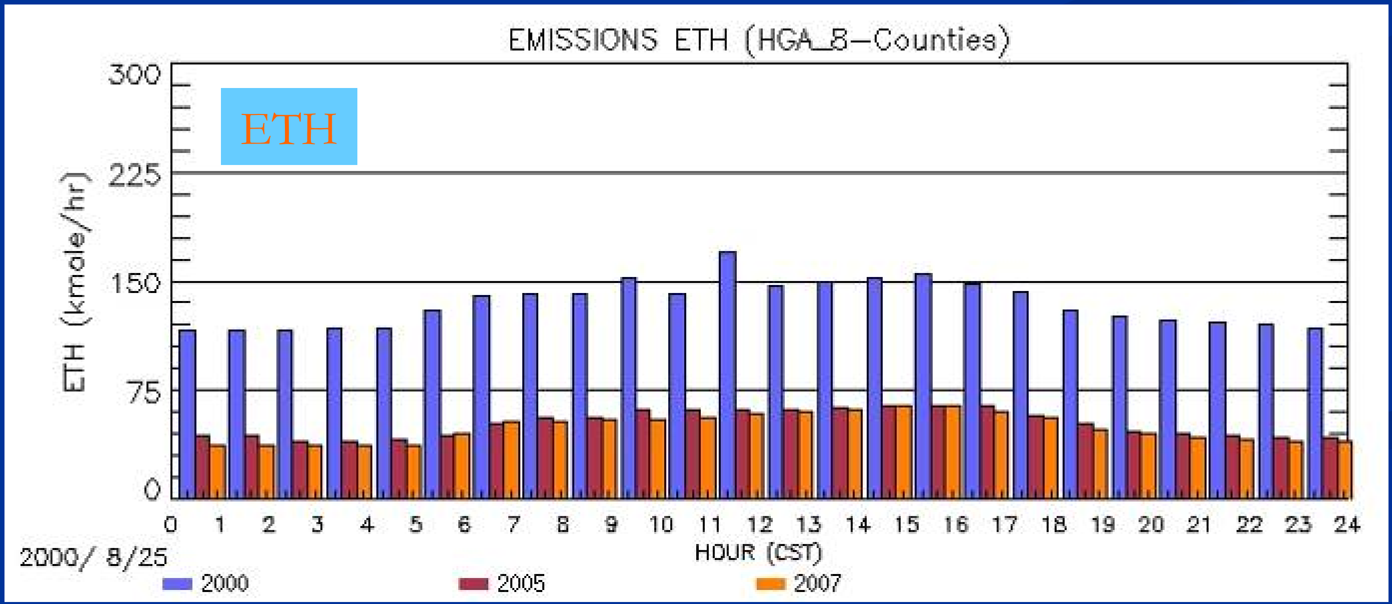




Base & Future Year Emissions



HGB 8 counties, August 25th



Outline

- Emissions Preparation for 2005 episodes
- **Base Simulations with CAMx and CMAQ**
- CMAQ HDDM Description
- HDDM Simulations
  - Contribution of NOx and VOC emissions from four neighboring states to DFW ozone concentrations
  - Source apportionment and sensitivity of NOx and VOC emissions over HGB 8-county area
  - Contribution of VOC emissions to HCHO concentrations
  - Contribution of biogenic emissions to Houston ozone concentrations

2005 Base Simulations

ETX 12-km domain

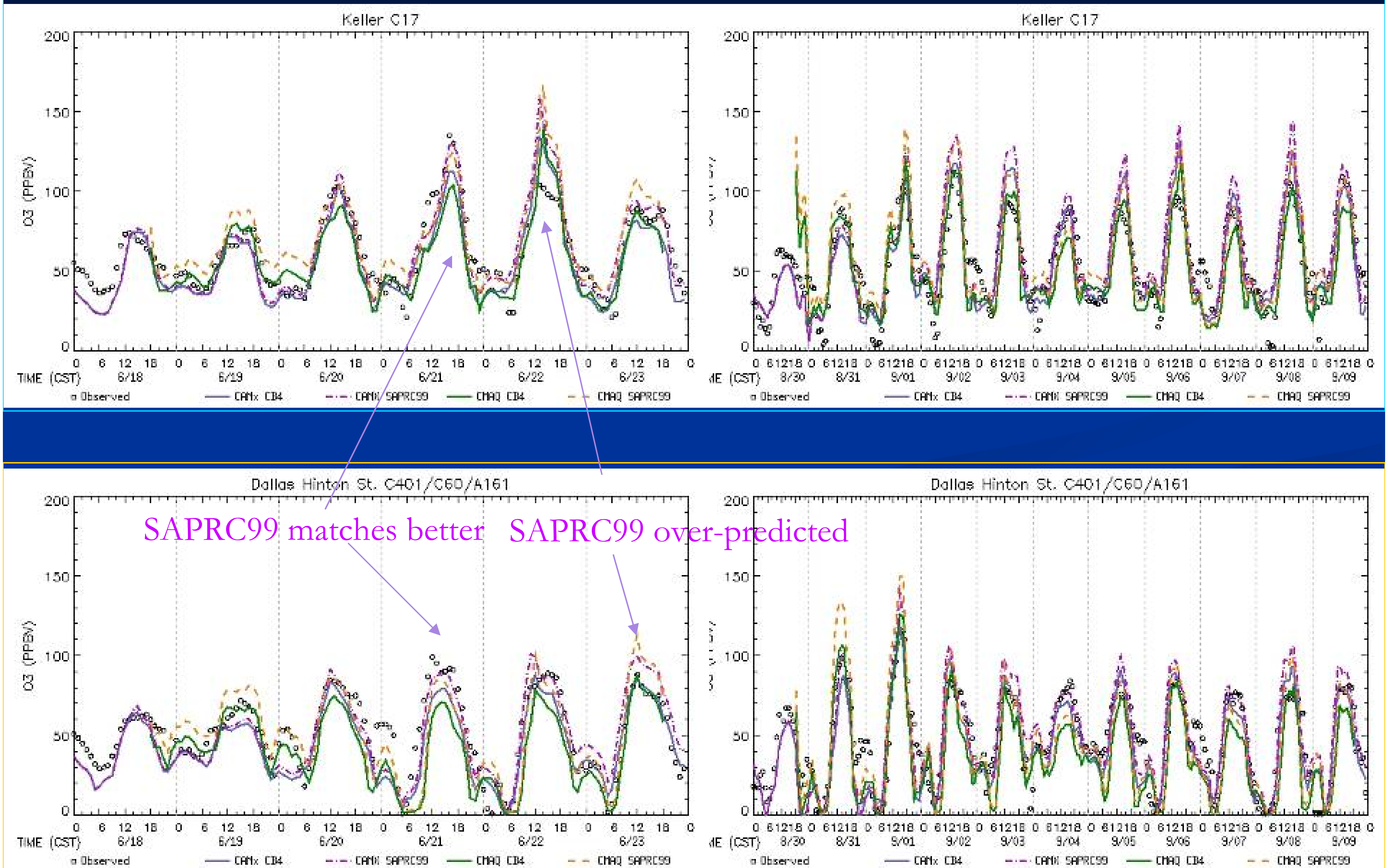
Run	Description
X41	CAMx 4.31, CB4, TAMU MM5, June 18 ~ June 23, 2005
X42	CAMx 4.31, SAPRC99, TAMU MM5, June 18 ~ June 23, 2005
X43	CAMx 4.31, CB4, TAMU MM5, August 30 ~ September 9 2005
X44	CAMx 4.31, SAPRC99, TAMU MM5, August 30 ~ September 9 2005
Q84	CMAQ 4.4, CB4, TAMU MM5, June 19 ~ June 23, 2005
Q85	CMAQ 4.4, SAPRC99, TAMU MM5, June 19 ~ June 23, 2005
Q86	CMAQ 4.4, CB4, TAMU MM5, August 31 ~ September 9 2005
Q87	CMAQ 4.4, SAPRC99, TAMU MM5, August 31 ~ September 9 2005

- John Nielsen-Gammon, of Texas A&M University (TAMU) prepared the MM5 simulation results for interesting 2005 modeling episodes for Dallas. These episodes were selected based on the model meteorology's favorability for high ozone, as well as (in most cases) actual observations of high ozone.

- UH selected two of these episodes, June 19-23 and August 30-September 9, for Houston and Dallas air quality simulations for the comparison study between CAMx and CMAQ.

2005 Base Simulations

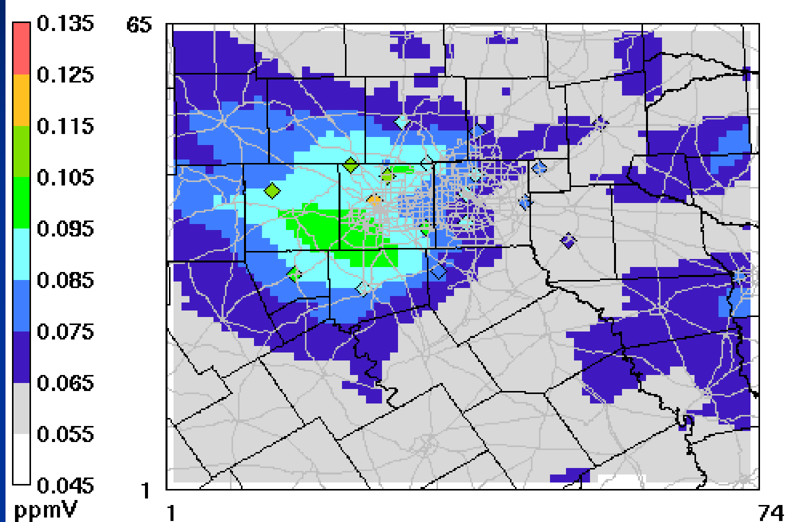
ETX 12-km domain
1-hr Ozone



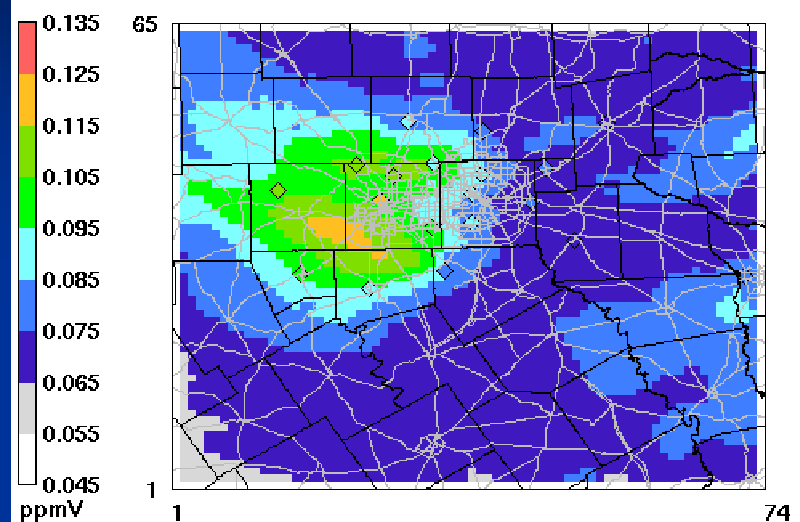
2005 Base Simulations

ETX 12-km domain
DFW 8-hr daily max O3
June 21st, 2005

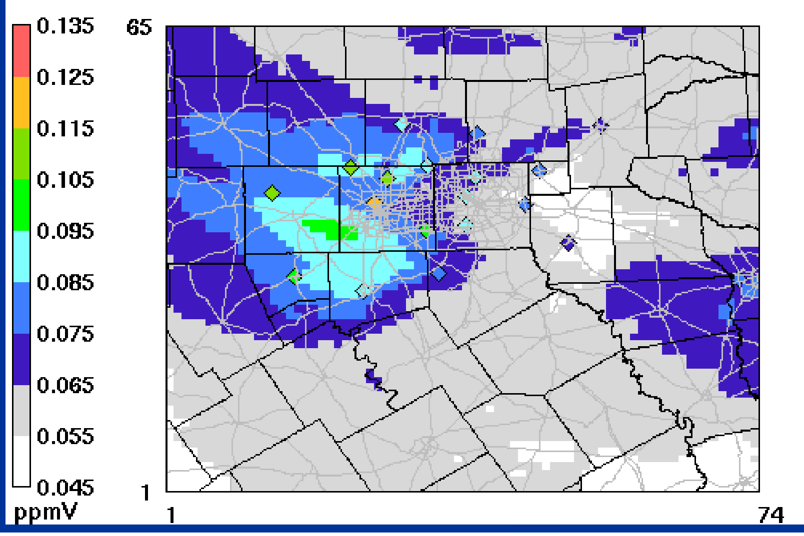
CAMx CB4



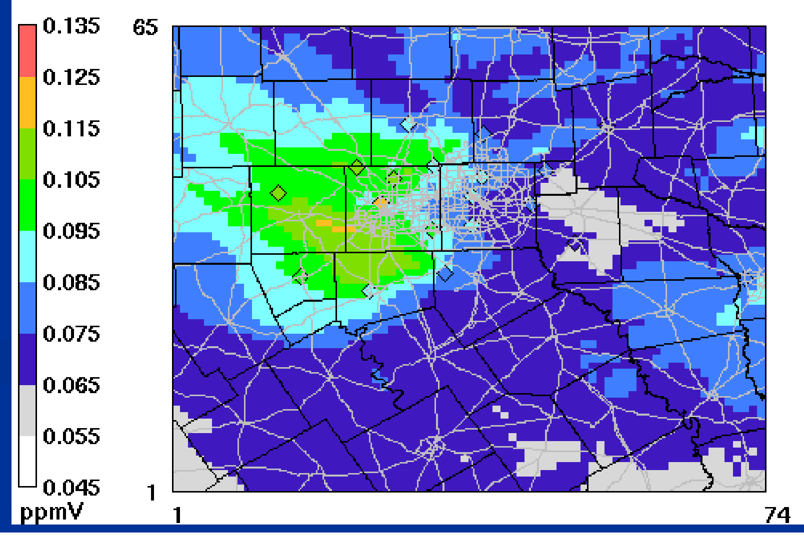
CAMx SPARC99



CMAQ CB4

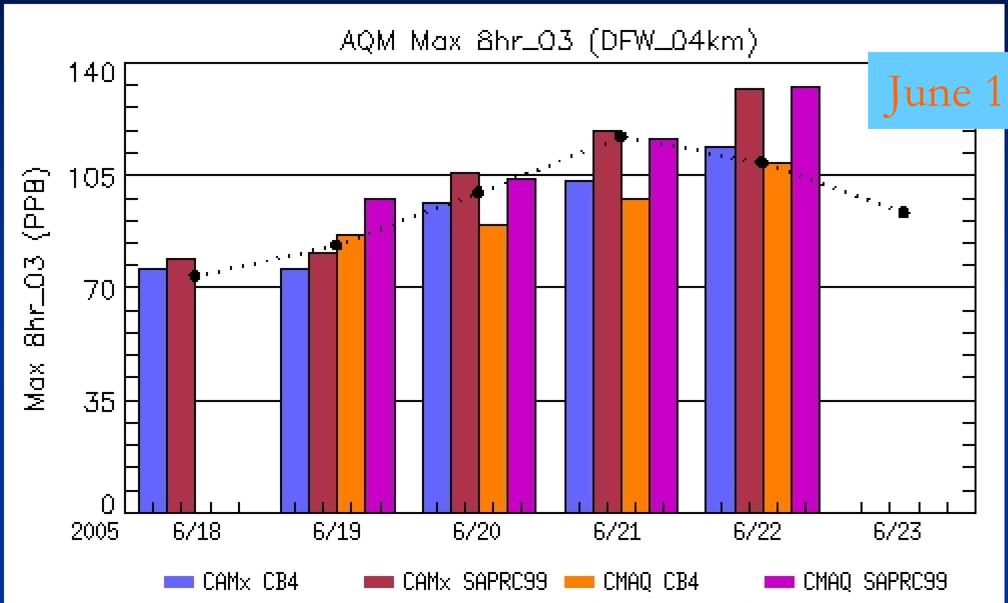


CMAQ SAPRC99

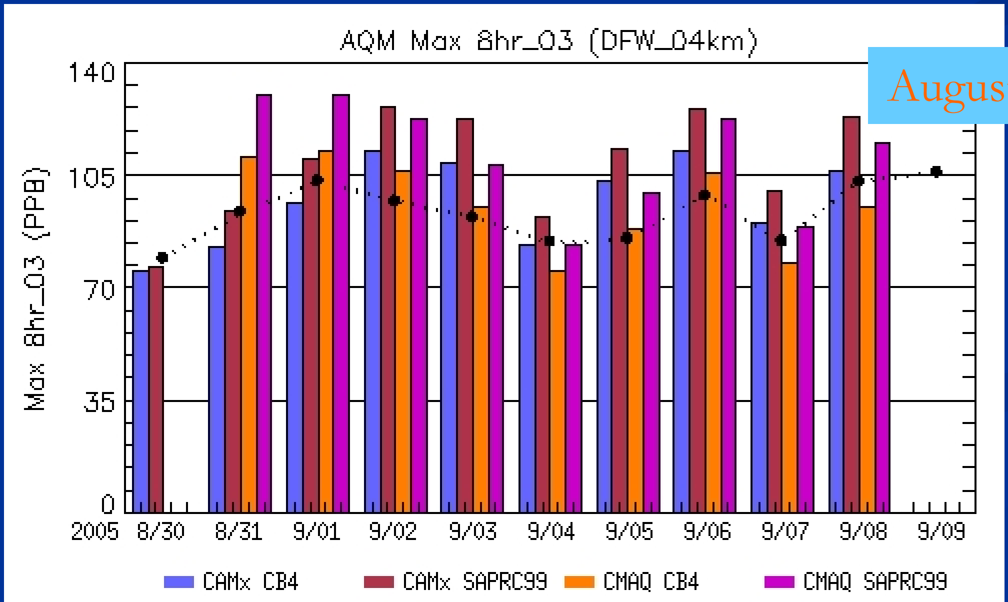


2005 Base Simulations

ETX 12-km domain
DFW 8-hr daily max O3



June 19th ~ 22nd, 2005

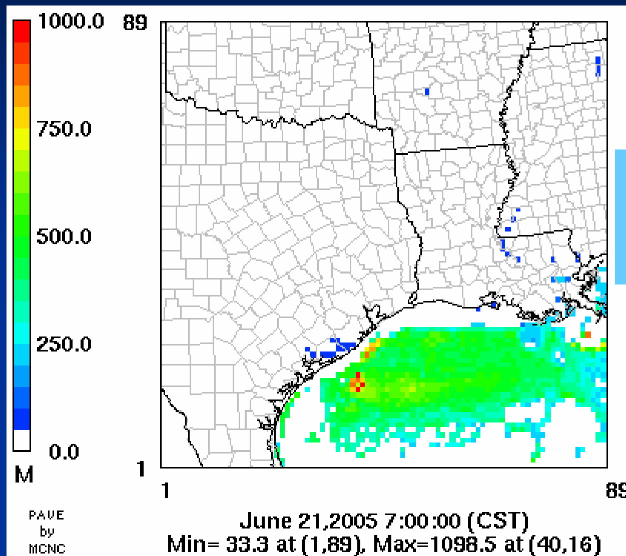


August 31st ~ September 8th, 2005

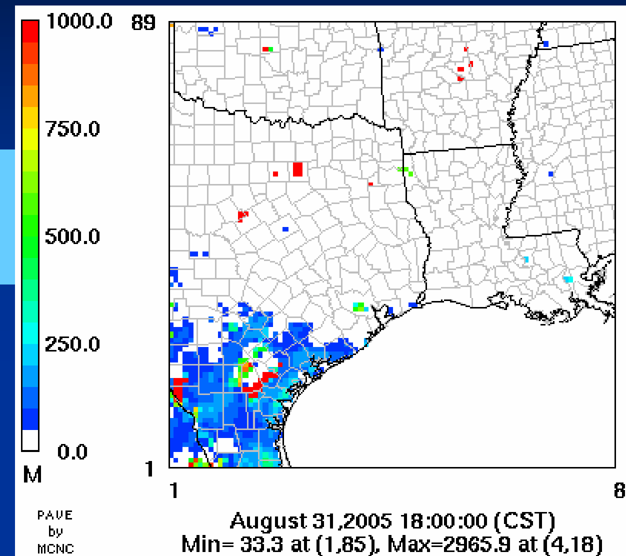
Black dots refer to observations.

ETX 12-km domain

Serious Uncertainties in PBL heights

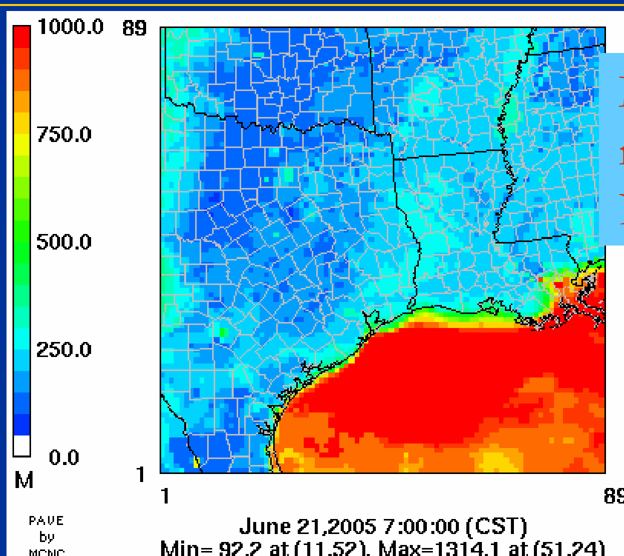


MM5-estimated PBL heights

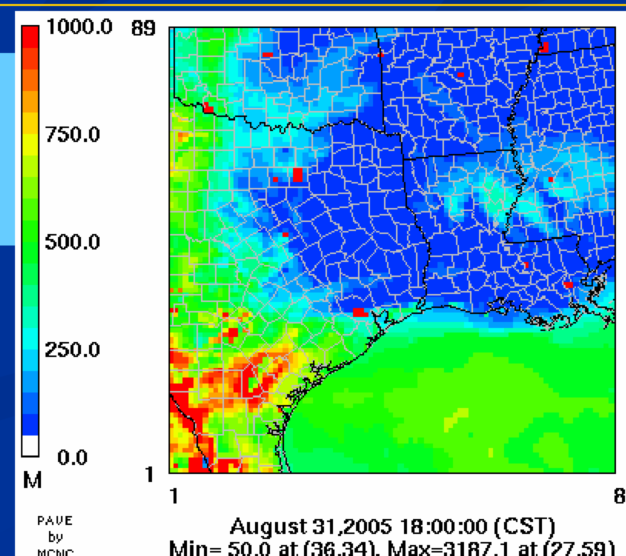


Early morning

Late afternoon



MCIP-recalculated PBL heights



Summary (1)

- For the DFW area, both CAMx and CMAQ behave similarly and compare well with observations from the Keller and Dallas Hinton St. sites, except on some days (e.g., June 22nd) when the models over-predicted ozone concentrations.

- CAMx and CMAQ simulations with SARPC99 chemical mechanism generally showed higher O3 concentrations than those with CB4.

- With the SAPRC99 chemical mechanism, CAMx and CMAQ showed a better performance for medium O3 concentration range.

Outline

- Emissions Preparation for 2005 episodes
- Base Simulations with CAMx and CMAQ
- **CMAQ HDDM Description**
- HDDM Simulations
  - Contribution of NOx and VOC emissions from four neighboring states to DFW ozone concentrations
  - Source apportionment and sensitivity of NOx and VOC emissions over HGB 8-county area
  - Contribution of VOC emissions to HCHO concentrations
  - Contribution of biogenic emissions to Houston ozone concentrations

CMAQ HDDM (1)

Cohan et al (2005)

- Perturbation

$$p_j = \varepsilon_j B_j = (1 + \Delta\varepsilon_j) B_j \qquad (1)$$

B_j : An unperturbed (base case) values of the sensitivity parameter

ε_j : A perturbation scaling variable with a nominal value of 1

p_j : Perturbations in a sensitivity parameter such as emissions rate, initial condition, or boundary condition

CMAQ HDDM (2)

Cohan et al (2005)

- First-order sensitivity coefficients

$$S_j^{(1)} = B_j \frac{\partial C}{\partial p_j} = B_j \frac{\partial C}{\partial(\varepsilon_j B_j)} = \frac{\partial C}{\partial \varepsilon_j} \quad (2)$$

$S_j^{(1)}$: The semi-normalized first-order sensitivity coefficients of concentration response to a model parameter (p_j) can be defined by scaling the local sensitivities

CMAQ HDDM (3)

Cohan et al (2005)

- Second-order sensitivity coefficients

$$S_{j,k}^{(2)} = B_j \frac{\partial}{\partial p_j}\left(B_k \frac{\partial C}{\partial (p_k)} \right) = \frac{\partial^2 C}{\partial \varepsilon_j \partial \varepsilon_k} \qquad (3)$$

: When $j = k$, the second-order sensitivity becomes the local curvature of the concentration-parameter relationship, while when $j \neq k$, it represents a cross-sensitivity interaction between the sensitivities of two different parameters.

CMAQ HDDM (4)

Cohan et al (2005)

- Sensitivity parameter for a perturbation p_j

$$C_{j,p_j} \approx C_0 + \Delta\varepsilon_j S_j^{(1)} + \frac{1}{2}\Delta\varepsilon_j{}^2 S_{j,j}^{(2)} + \cdots \quad (4)$$

: The second-order term scales with $\Delta\varepsilon_j^2$ and the importance increases with the size of the perturbation.

CMAQ HDDM (5)

Cohan et al (2005)

- Zero-out source contribution (ZOC) of an emission source

: is defined as a concentration achieved when the source does not exist. Due to the nonlinearities in ozone chemistry, ozone concentrations do not strictly corresponds to the sum of zero-out contributions. The second-order approximation of ZOC can be computed with $\Delta\varepsilon_j = -1$ from equation (4)

$$ZOC(B_j) \cong C_0 - C_{j,p_j=0} = S_j^{(1)} - \frac{1}{2} S_{j,j}^{(2)} \quad (5)$$

CMAQ HDDM (6)

Cohan et al (2005)

- Sensitivity parameter & ZOC for multiple perturbations p_j & p_k

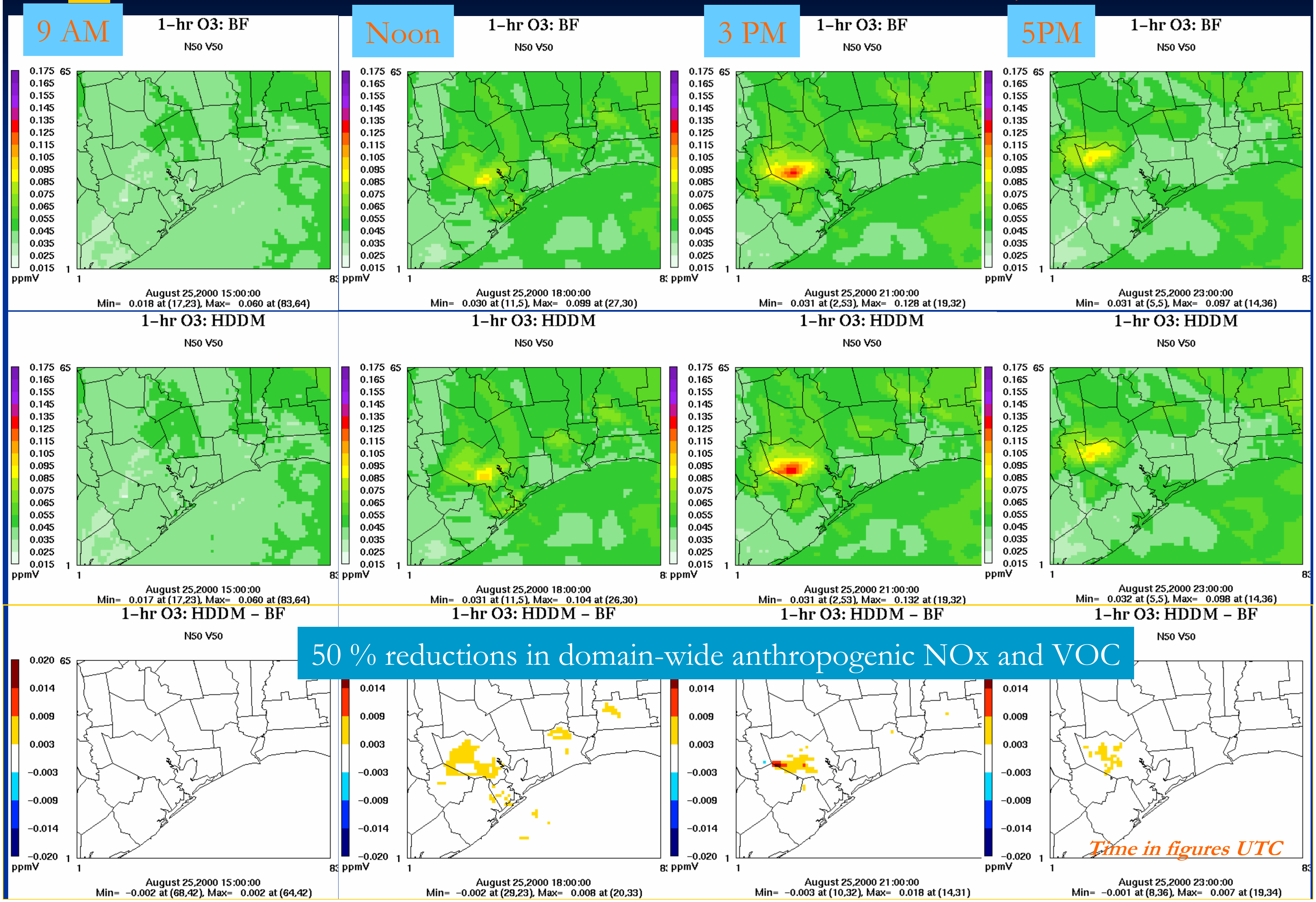
$$C_{j+k} \cong C_0 + \Delta\varepsilon_j S_j^{(1)} + \Delta\varepsilon_k S_k^{(1)} + \frac{1}{2}\Delta\varepsilon_j{}^2 S_{j,j}^{(2)} + \frac{1}{2}\Delta\varepsilon_k{}^2 S_{k,k}^{(2)} + \frac{1}{2}\Delta\varepsilon_j \Delta\varepsilon_k S_{j,k}^{(2)} + \ldots \quad (6)$$

$$ZOC(B_j + B_k) \approx \left(S_j^{(1)} - \frac{1}{2}S_{j,j}^{(2)}\right) + \left(S_k^{(1)} - \frac{1}{2}S_{k,k}^{(2)}\right) - S_{j,k}^{(2)} \quad (7)$$

$\frac{\partial C}{\partial \varepsilon_j}$

Brute Force vs. HDDM sensitivity coefficients

Snapshots for August. 25th, 2000

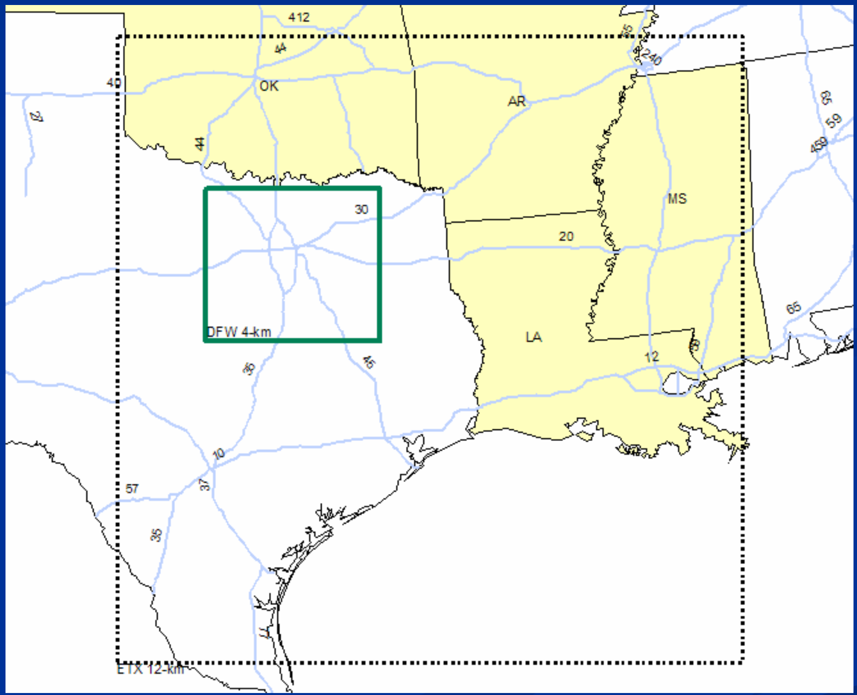


Outline

- Emissions Preparation for 2005 episodes
- Base Simulations with CAMx and CMAQ
- CMAQ HDDM Description
- **HDDM Simulations**
  - Contribution of NOx and VOC emissions from four neighboring states to DFW ozone concentrations
  - Source apportionment and sensitivity of NOx and VOC emissions over HGB 8-county area
  - Contribution of VOC emissions to HCHO concentrations
  - Contribution of biogenic emissions to Houston ozone concentrations

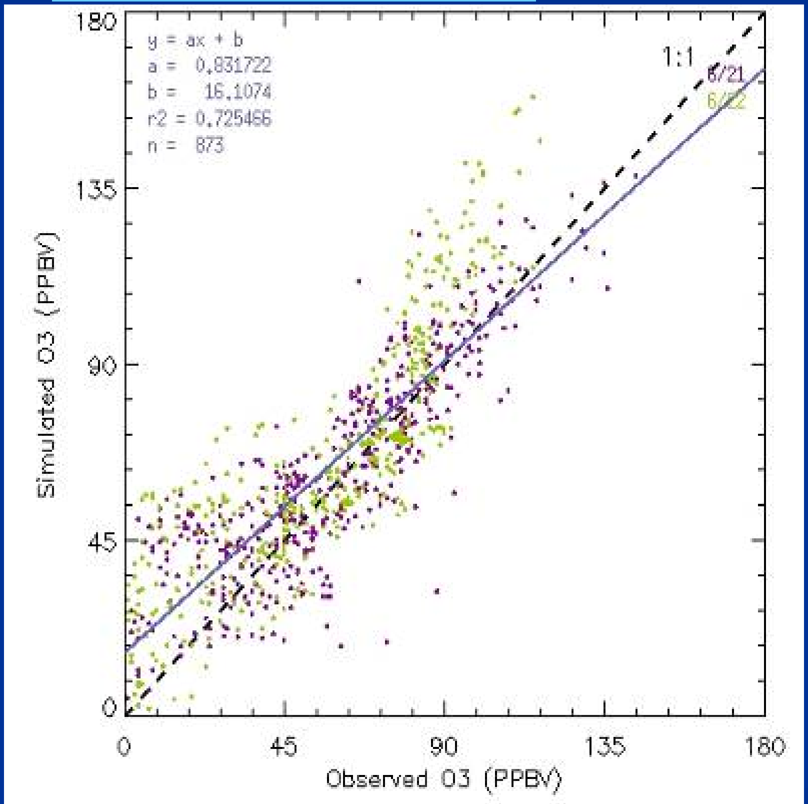
Case Study 1 - DFW

- To quantify the contributions of NOx and VOC emissions from neighboring states (Oklahoma, Louisiana, Arkansas, and Mississippi) to Dallas and Texas ozone concentrations, source regions were predefined with grid cells over the domain
  - A set of simulations was designed over a 12-km resolution domain for the periods of June 19 ~ 23 (5 days) and August 31 ~ September 5 (6 days) in 2005, when base CMAQ and CAMx runs showed better matches to the observations, and northeasterly winds were predominant.

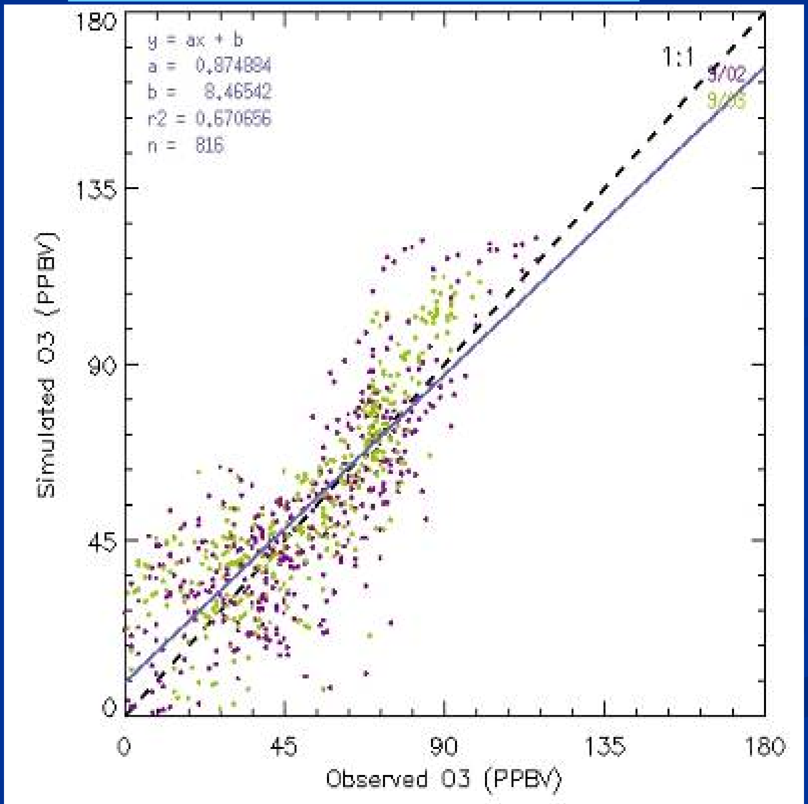


1-hr Ozone for the episodes

June 21st and 22nd, 2005



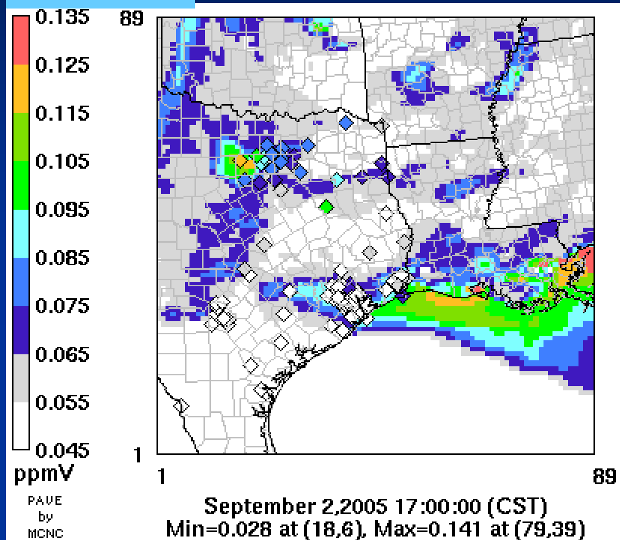
September 2nd and 3rd, 2005



Ozone & ZOC of Four State Emissions

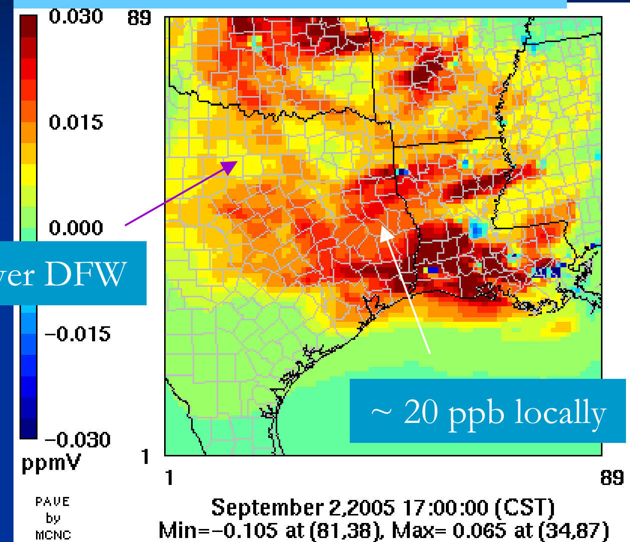
Case 1 - DFW
Snapshots for Sept. 2nd, 2005 at 17 CST

1-hr O3

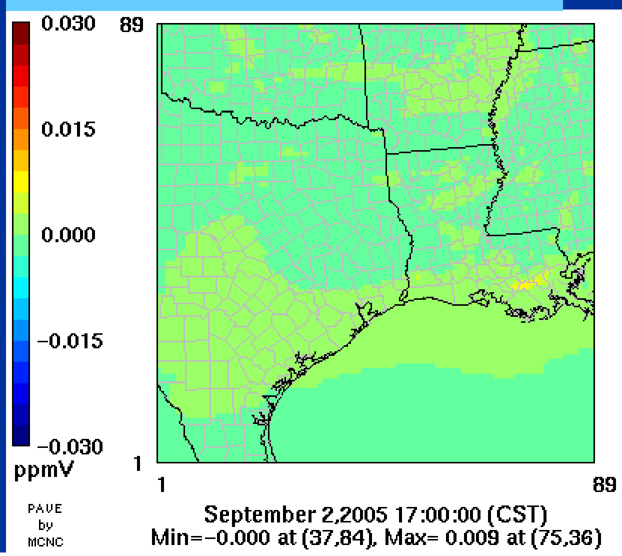


ZOC of NOx from 4 states

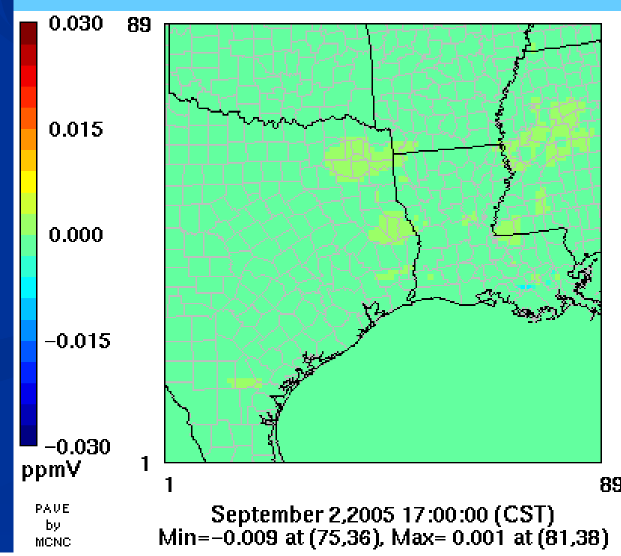
~ 10 ppb over DFW



ZOC of VOC from 4 states



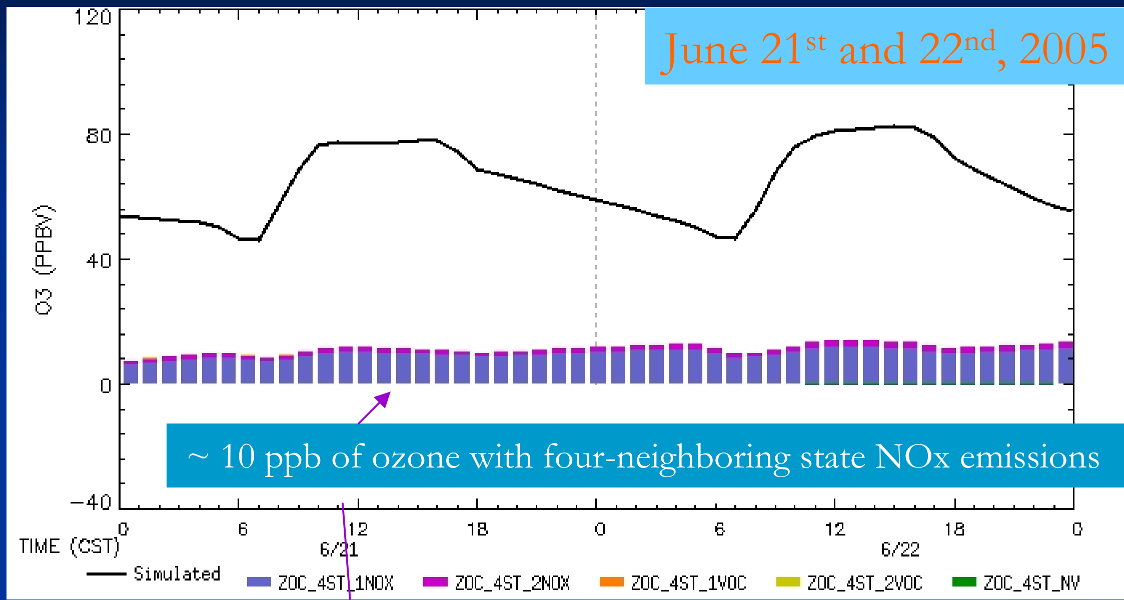
ZOC of NOx x VOC from 4 states

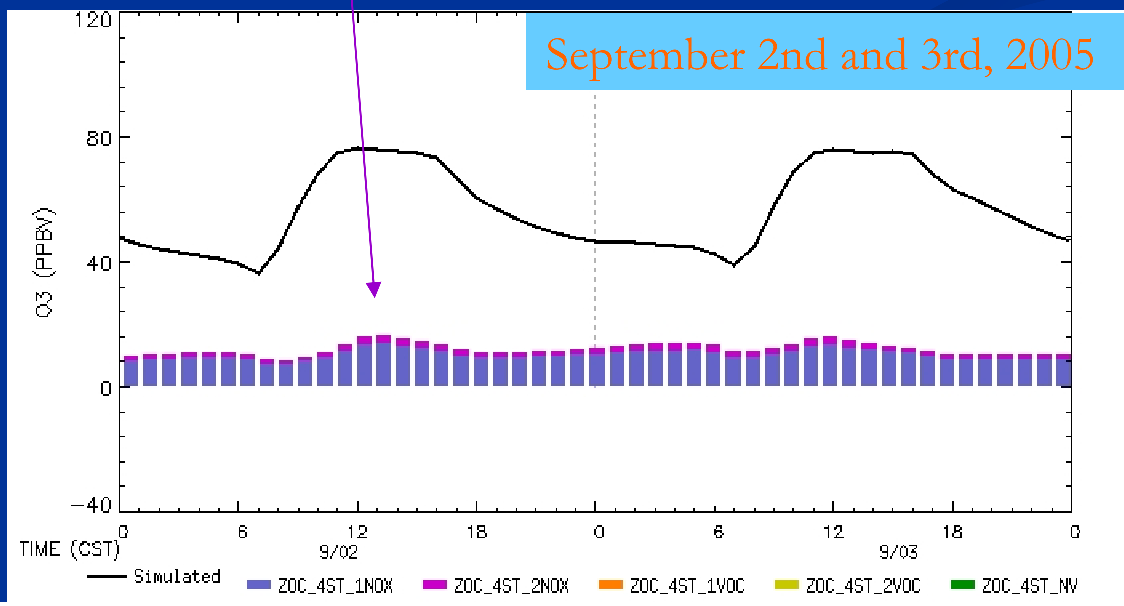


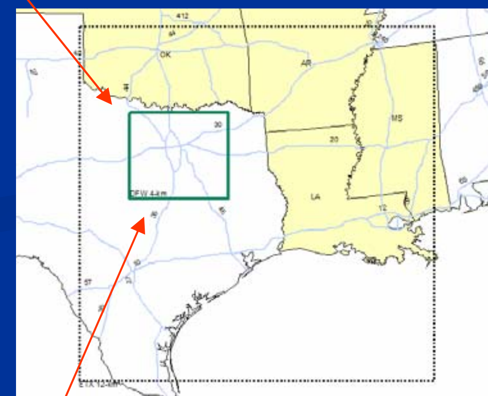
Cross term

Ozone & ZOC of Four State Emissions

Case 1 - DFW
Over DFW 4-km domain



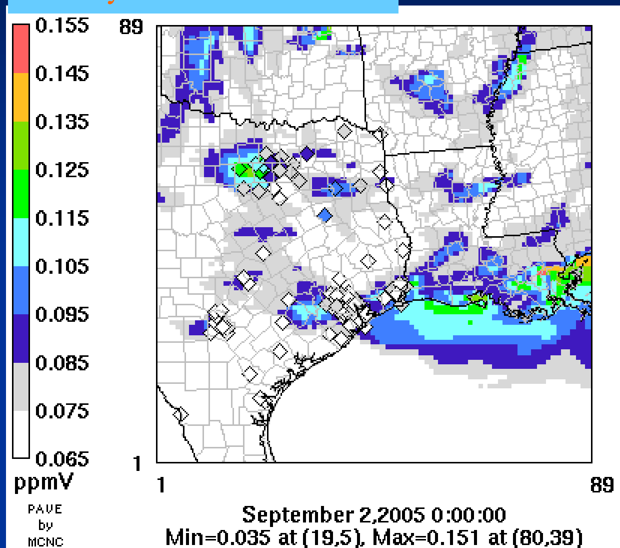




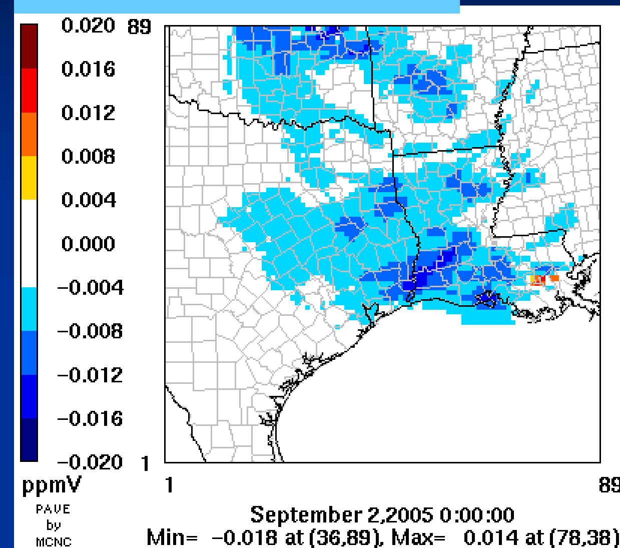
Daily maximum 1-hr ozone & the changes for NOx emissions from 4 states

Case 1 - DFW
Sept. 2nd, 2005

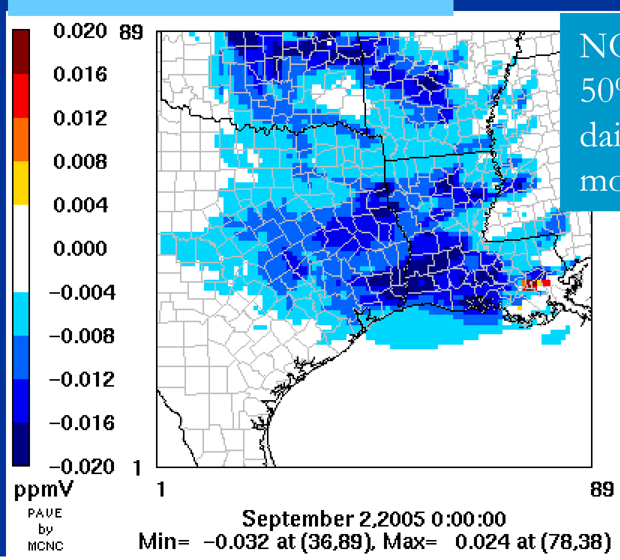
Daily max 1-hr O3



30 % NOx reduction

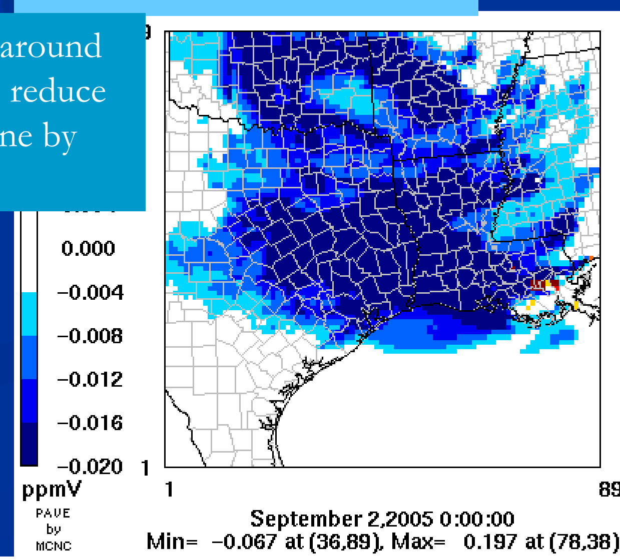


50 % NOx reduction



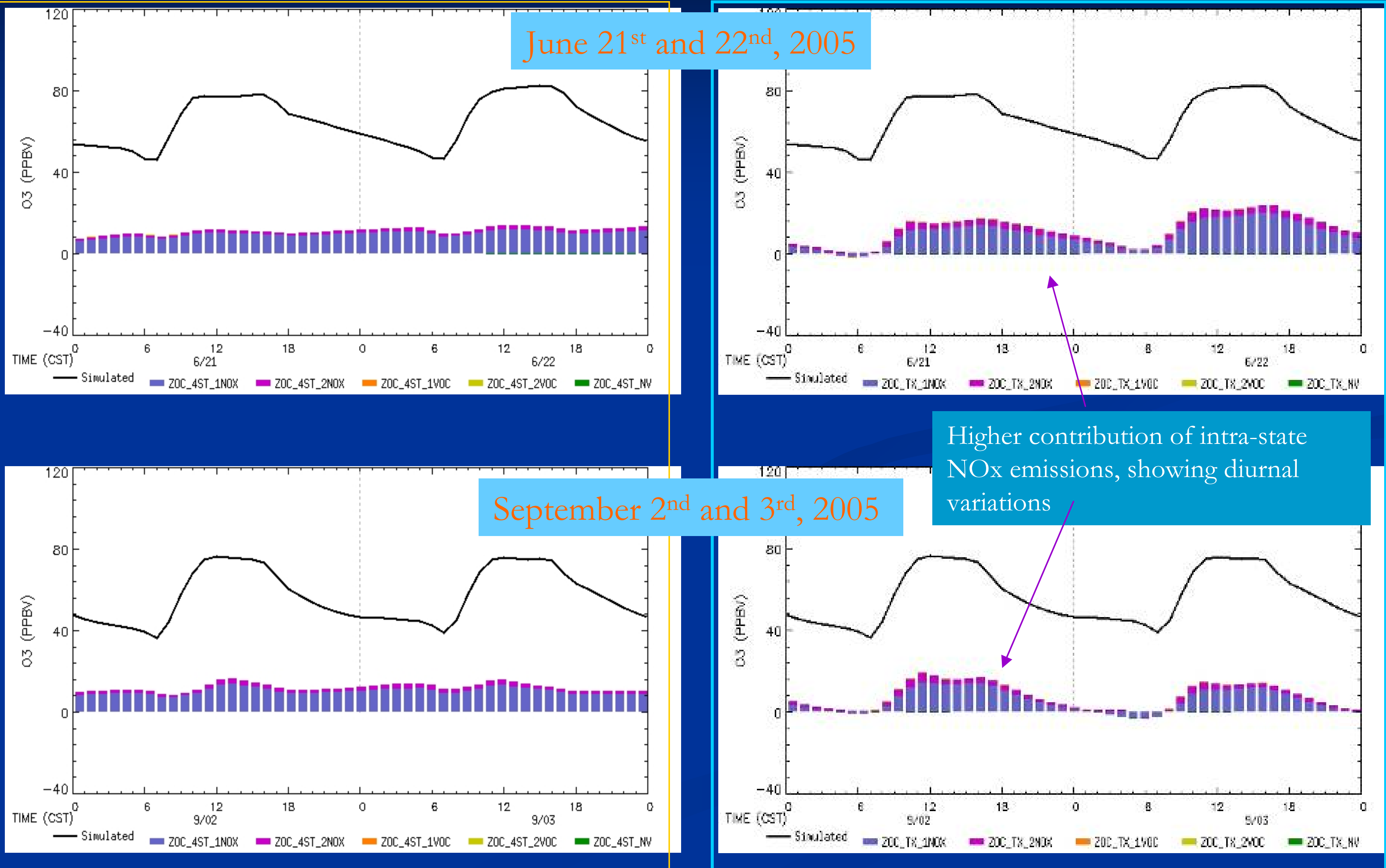
NOx reductions of around 50% are required to reduce daily maximum ozone by more than 4 ppb

100 % NOx reduction



Case 1 - DFW
Over DFW 4-km domain

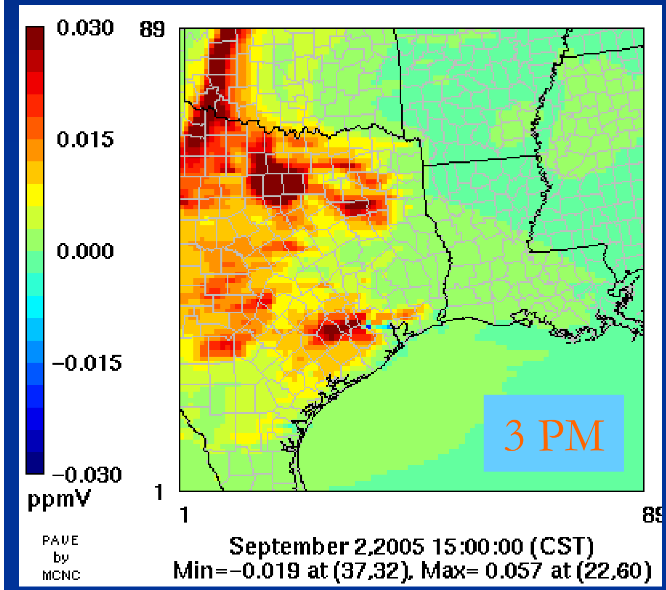
Ozone & ZOC of inter-/intra-state emissions



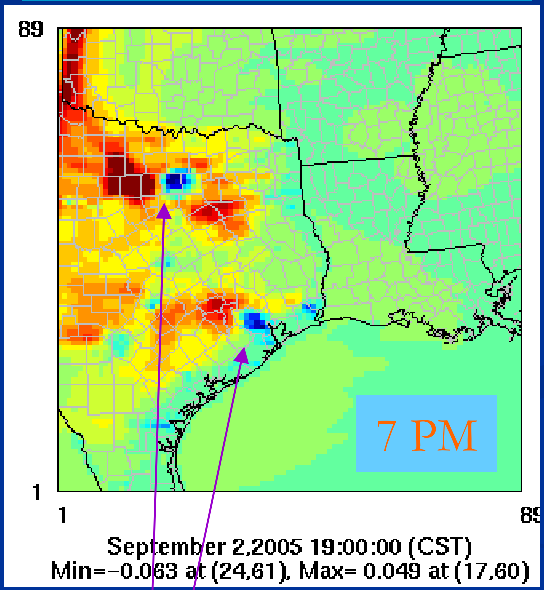
Ozone & ZOC of TX NOx emissions (intra state)

Case 1 – DFW
September 2nd, 2005

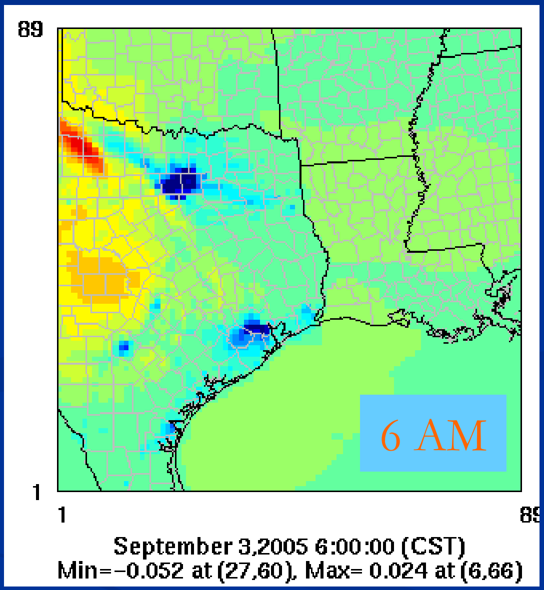
NOx is used to form ozone



O3 titration begins to appear



Mostly O3 titration



Depends on the source location and time.

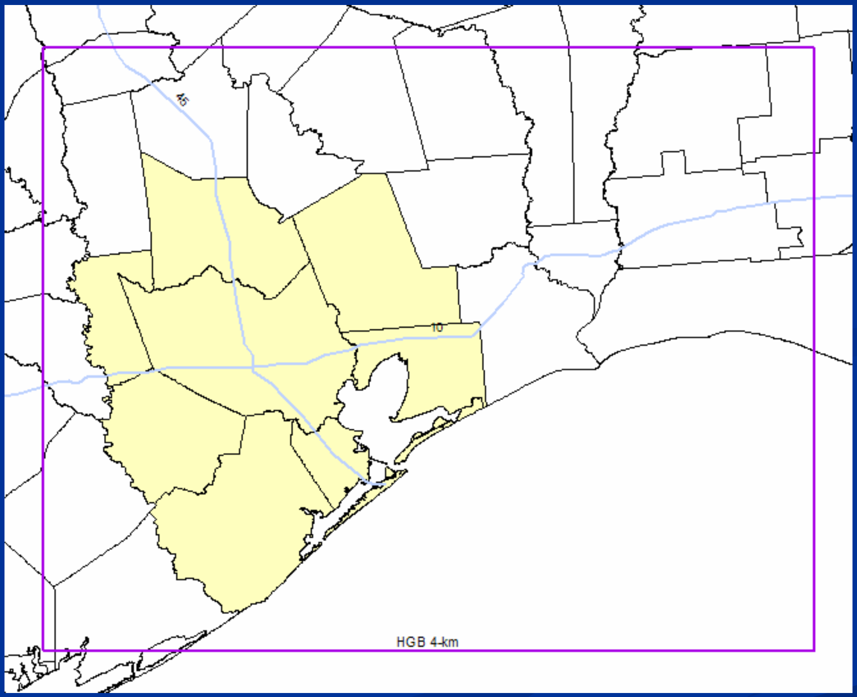
Summary (2)

- HDDM simulations assessing the impact of regional transport on DFW ozone concentrations reveal that the Texas intra-state contribution is dominated by NOx and can exceed 20 ppb, whereas the combination of sources in the neighboring states of Oklahoma, Arkansas, and Louisiana contribute up to around 10 ppb.

- NOx reductions of around 50% in the four neighboring states of Oklahoma, Arkansas, and Louisiana, and Mississippi are required to reduce daily maximum ozone by more than 4 ppb (roughly comparable to the current SIP ozone attainment deficit based on projected 2009 8-hour ozone design values).

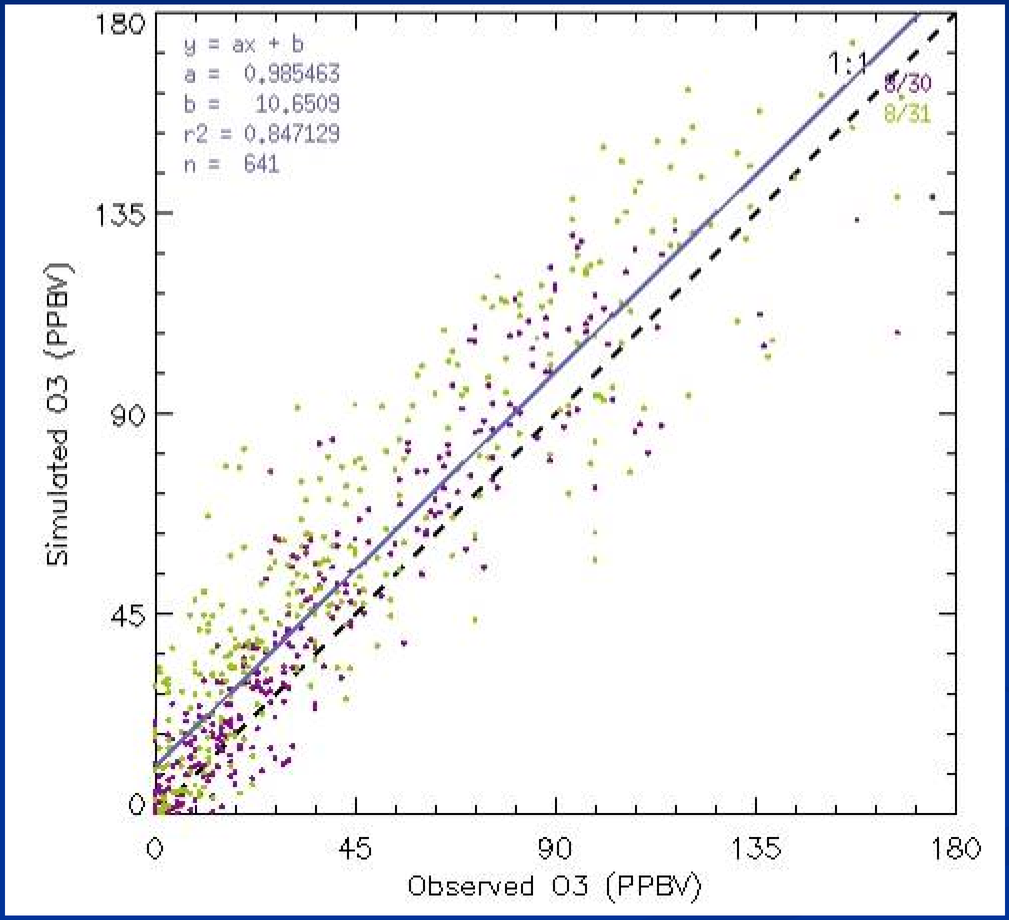
Case Study 2 - HGB

- To assess the contributions of emission sources to high ozone concentrations observed during TexAQS 2000 (August 28 ~ September 1, 2000).
  - Sources of interest include mobile NOx emissions from HGB counties and HRVOC emissions such as ethane and olefins from the Ship Channel area.
  - Anthropogenic and biogenic emissions are considered separately.



Case 2 – HGB
August 30th ~ 31st, 2000

Predicted Surface Ozone compared to CAMS

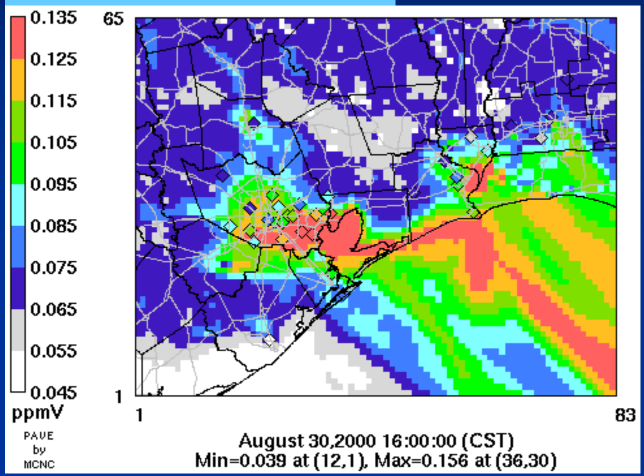


Ozone & ZOC of NOx and VOC

Case 2 - HGB
August. 30th, 2000 at 16 CST

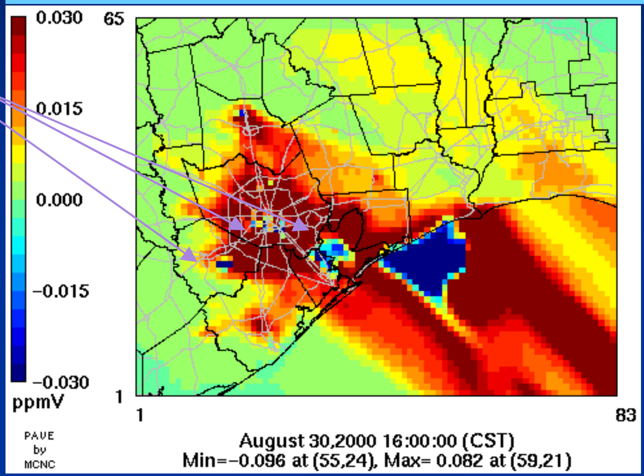
daily maximum sensitivity coefficients

Daily max 1-hr O3

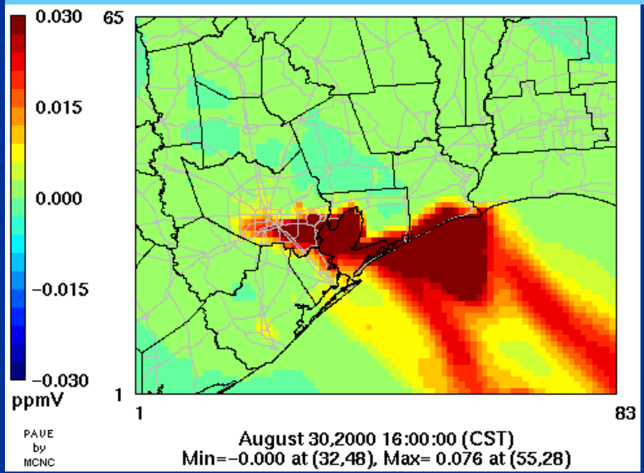


High
NOx
sources

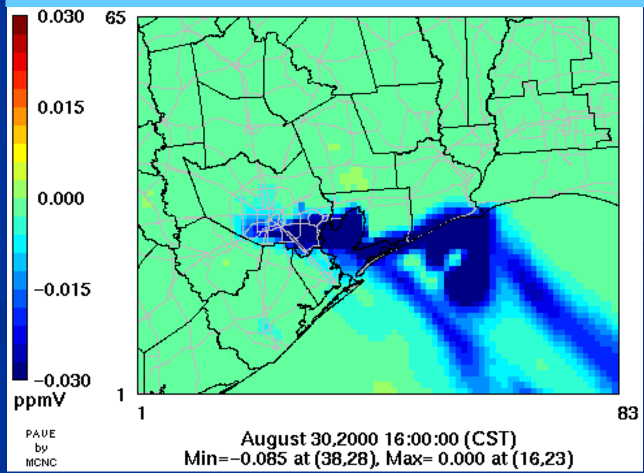
HGB NOx from all anthropogenic sources



HGB VOC from all anthropogenic sources



Cross-term between NOx and VOC from all HGB anthropogenic sources

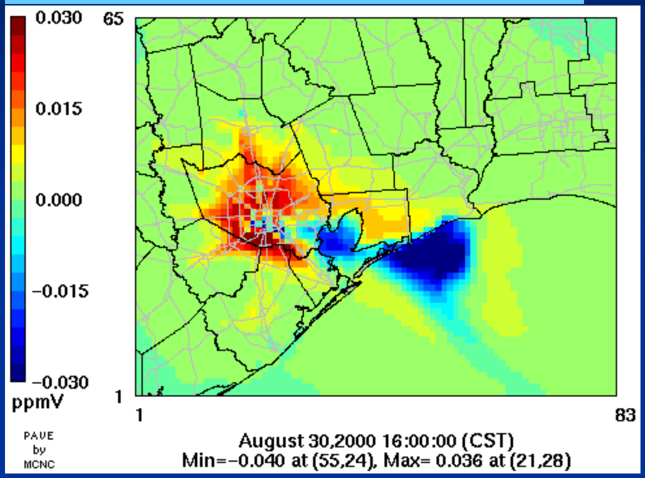


Ozone & ZOC of NOx and VOC *(continued)*

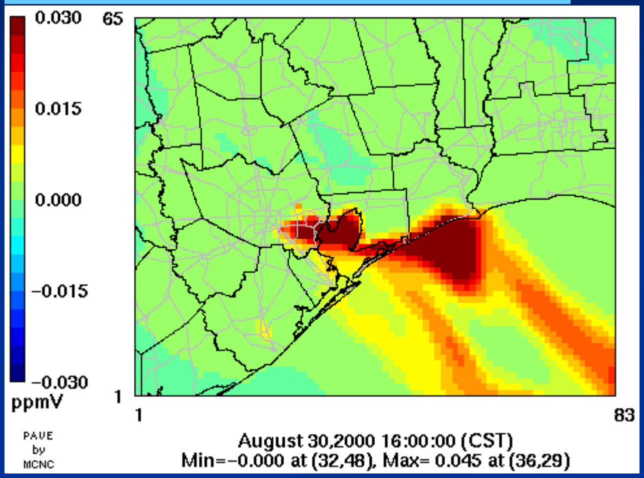
Case 2 - HGB

August. 30th, 2000 at 16 CST

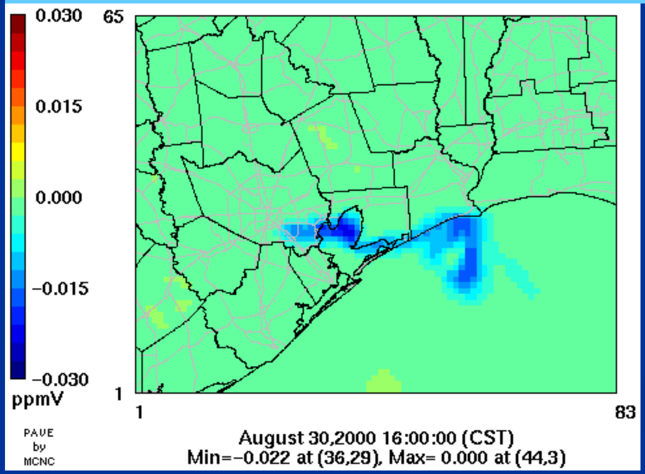
HGB NOx from mobile sources



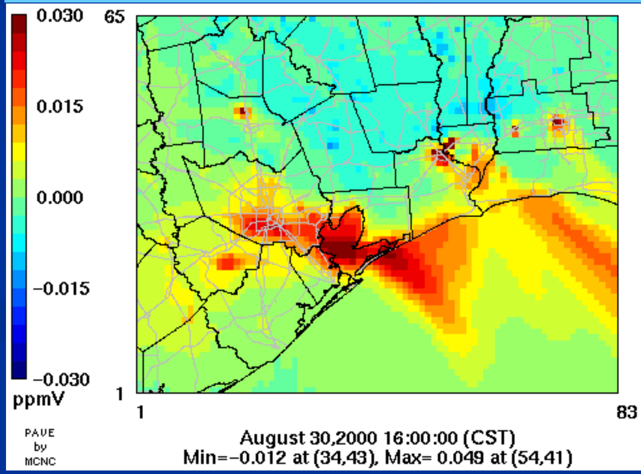
HGB VOC from point sources



Cross-term between mNOx and pVOC



Domain-wide biogenic emissions



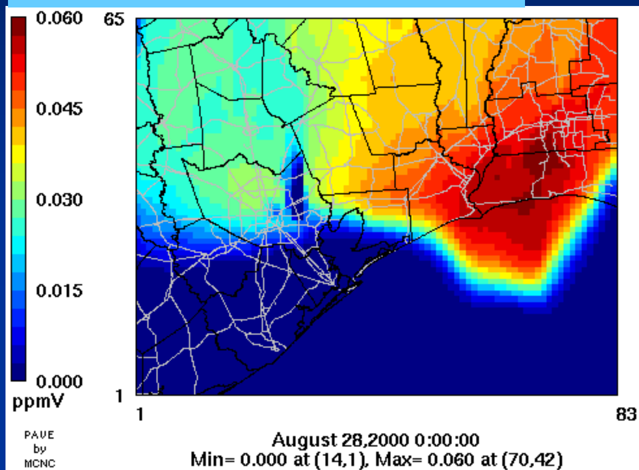
Ozone & ZOC of NOx and VOC *(continued)*

Case 2 - HGB

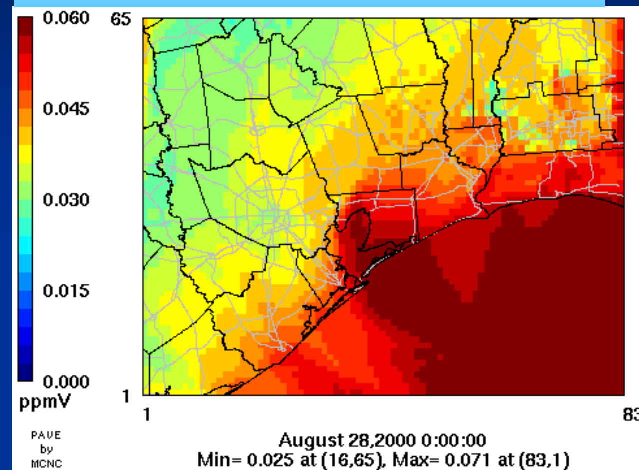
Daily max on August. 28th & 30th, 2000

1st day of the simulation

Initial O3 concentrations

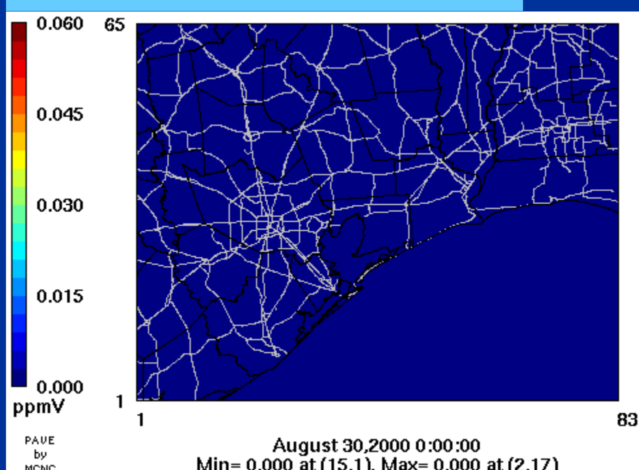


Boundary O3 concentrations

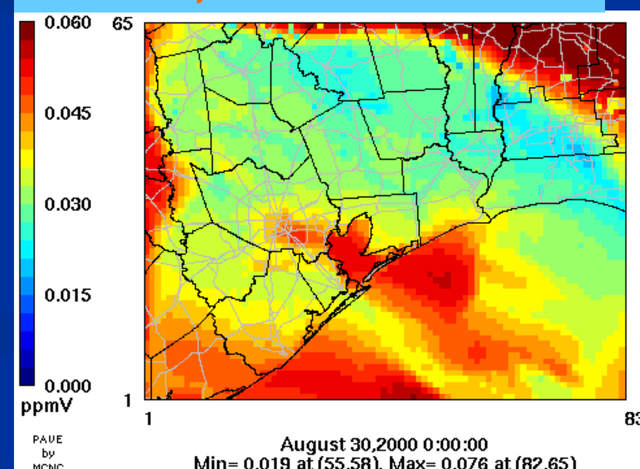


3rd day of the simulation

Initial O3 concentrations

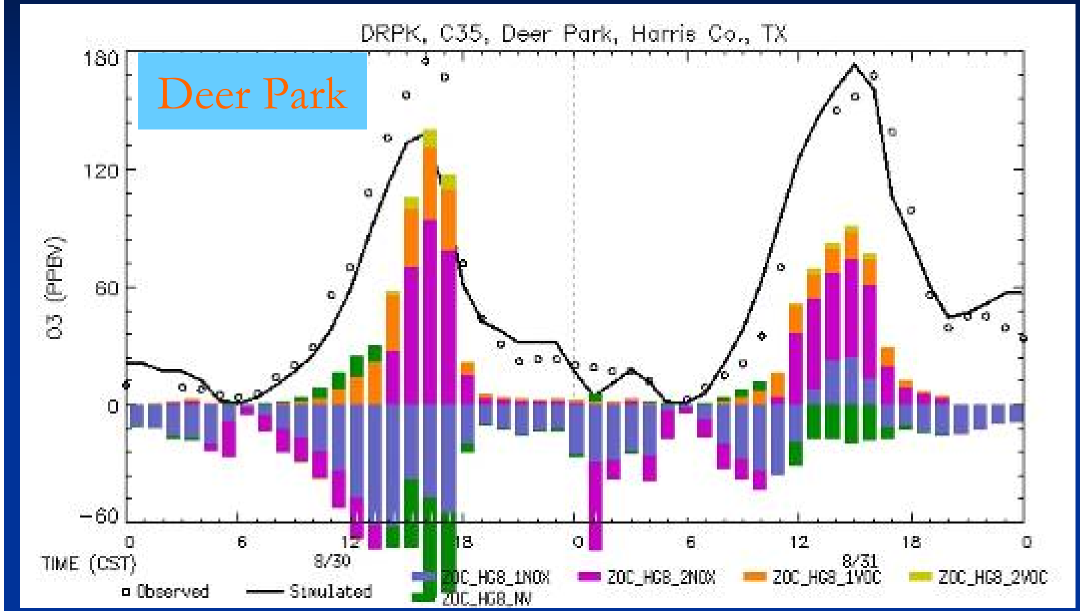


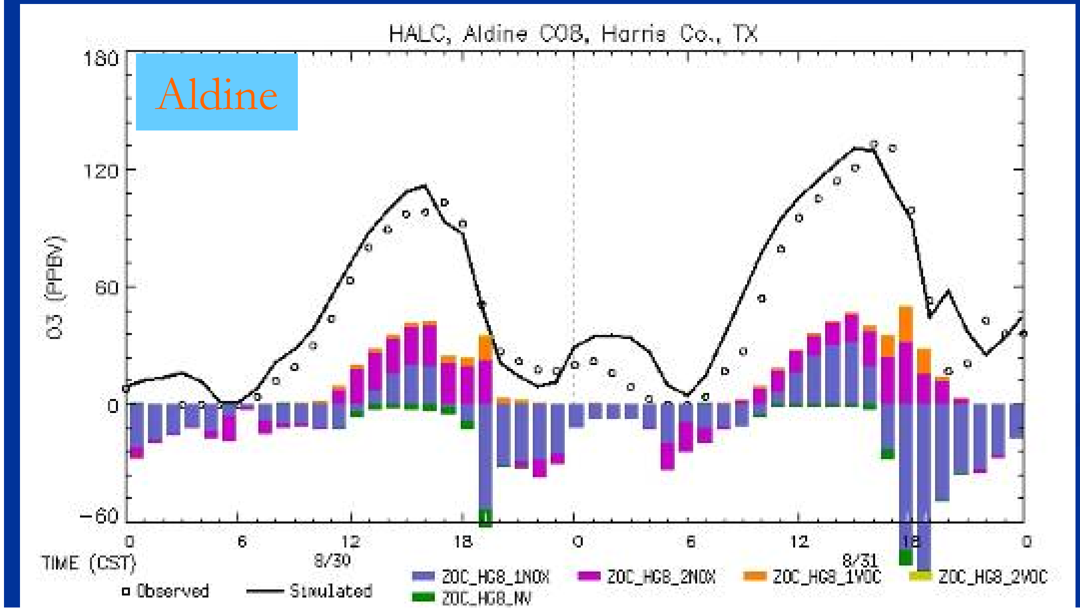
Boundary O3 concentrations



ZOC of Anthropogenic NOx and VOC emissions from HGB 8 counties

Case 2 – HGB
August 30th ~ 31st, 2000



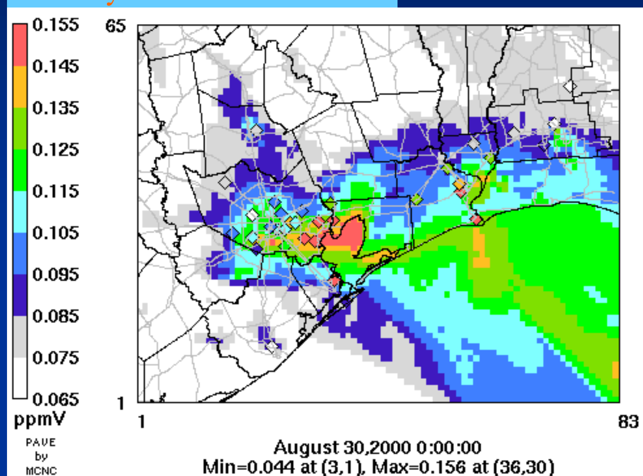


- 1-hour ozone at the Deer Park (Ship Channel) and Aldine (North Houston) monitoring sites
- At both stations, the first order ZOC for NOx is responsible mainly for NOx titration of ozone (especially on Aug. 30th), whereas the second order ZOC of NOx contributes primarily to ozone production.
- The VOC contributions are stronger at Deer Park than at Aldine, due to the large primary emissions of VOCs in the Ship Channel and the aging of pollution plumes that eventually reach North Houston.
- The NOx-VOC cross term is mostly negative at both sites, but relatively small compared to other terms at Aldine

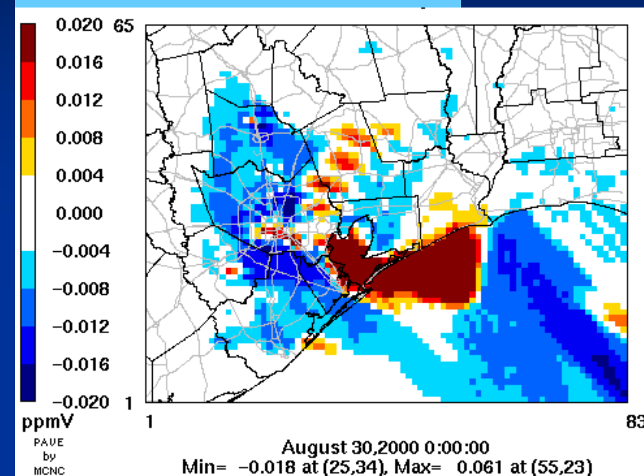
Daily maximum 1-hr ozone & the changes for anthropogenic NOx & VOC reductions

Case 2 - HGB
August. 30th, 2000

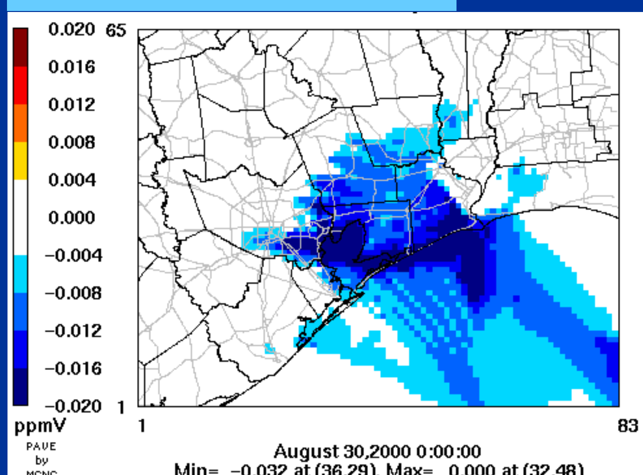
Daily max 1-hr O3



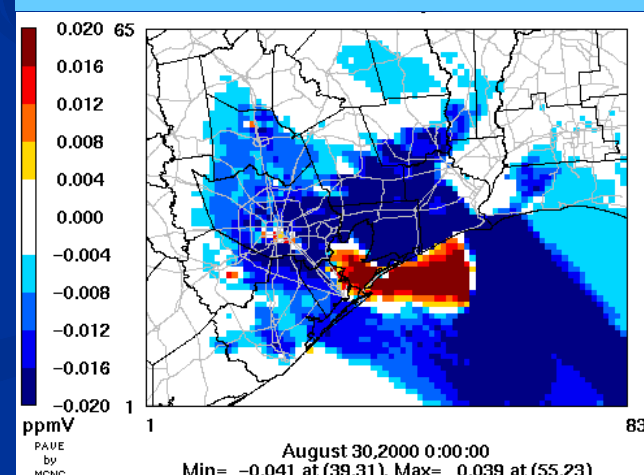
50 % NOx reduction



50 % VOC reduction

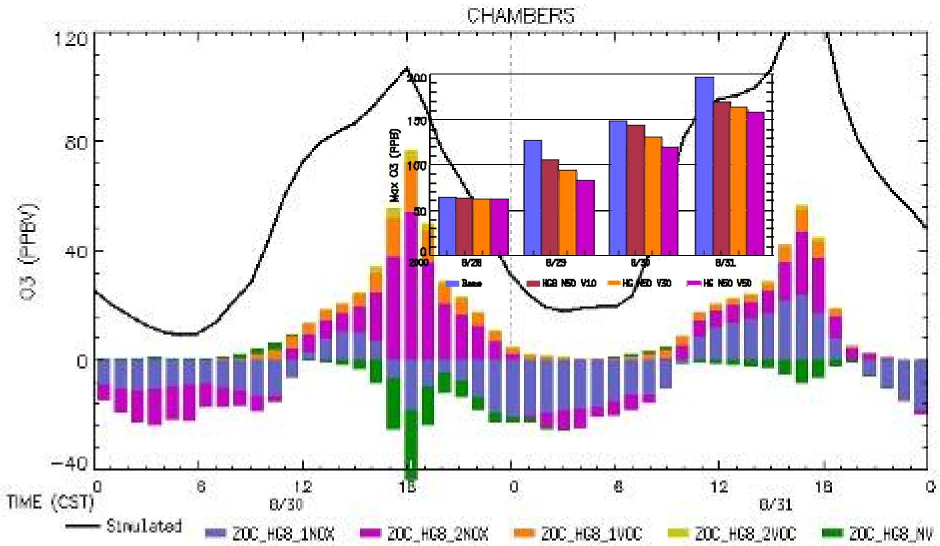


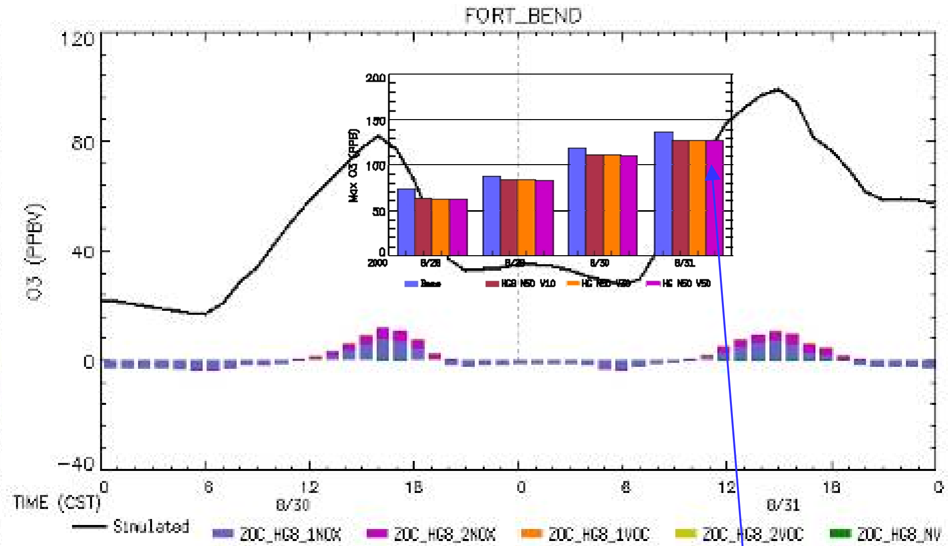
50 % NOx & VOC reduction



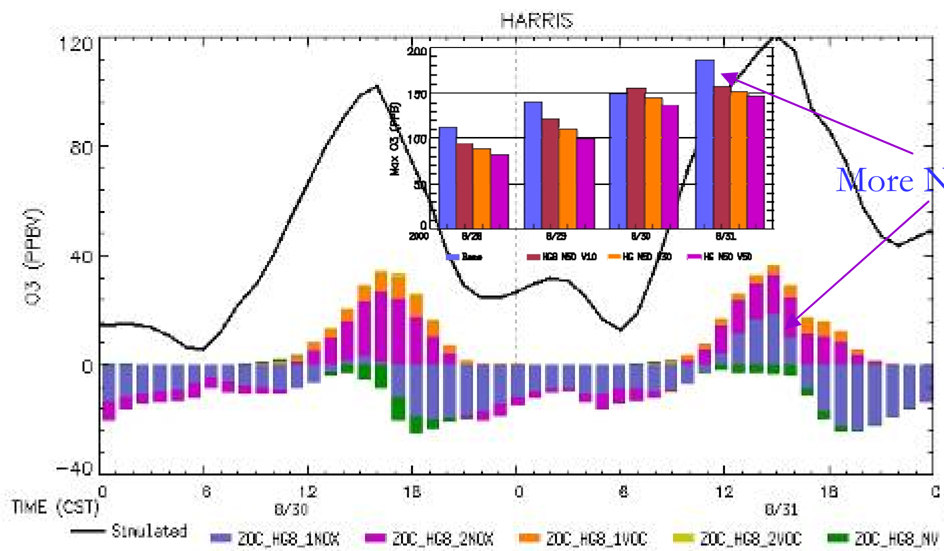
ZOC of Anthropogenic NOx and VOC emissions from HGB 8 counties

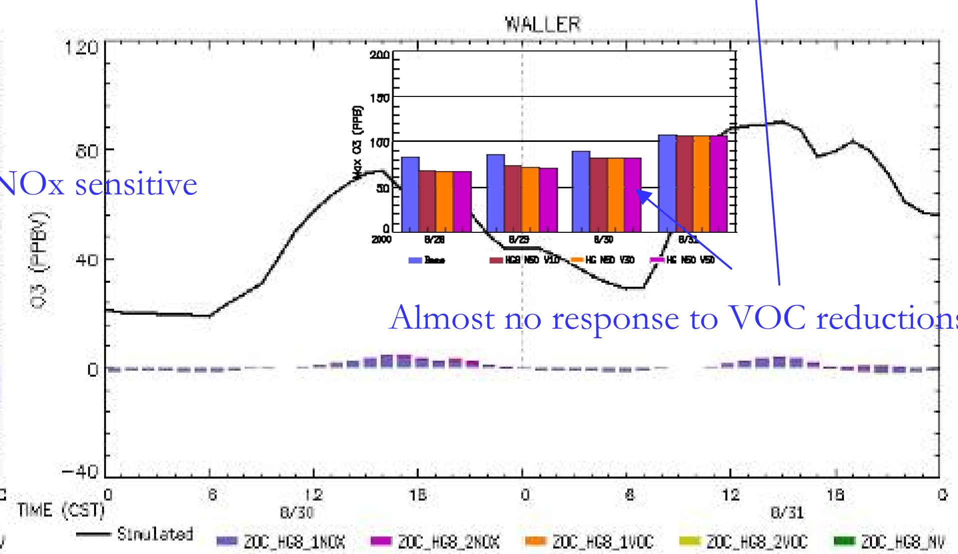
Case 2 – HGB
August 30th ~ 31st, 2000





Small bar plots - Blue: Base, Brown: -50% NOx & -10% VOC
Orange: -50% NOx & -30% VOC Purple: -50% NOx & -50% VOC





More NOx sensitive

Almost no response to VOC reductions

Summary

- Local NOx and VOC contributions to daily maximum ozone are comparable during the HGB 2000 episode. The VOC point source contribution maximizes on the east side of Houston in the area of the Ship Channel, whereas the mobile NOx contribution maximizes on the west side of Houston (on Augst 30th and 31st, 2000).

- When HDDM analysis was applied to the Deer Park and Aldine sites, the first order contribution by NOx was found to be responsible mainly for NOx titration of ozone, whereas the second order term contributed primarily to ozone production. The VOC contributions were stronger at Deer Park than at Aldine, due to the large primary emissions of VOCs in the Ship Channel and the aging of pollution plumes that eventually reach North Houston. The NOx-VOC cross term is mostly negative at both sites, but relatively small compared to other terms at Aldine.

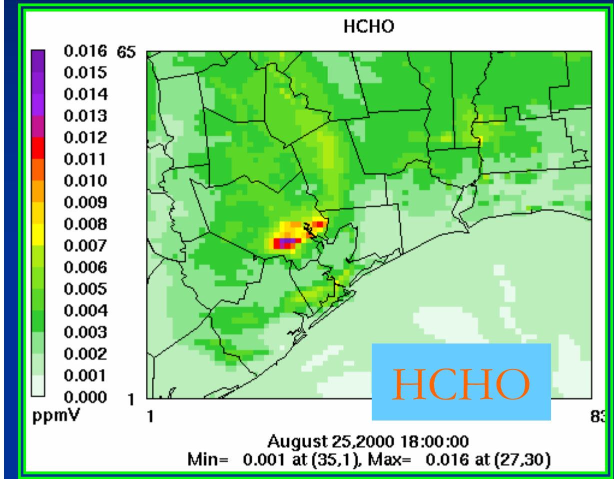
- Based on analysis with the 2000 HGB episode, deep NOx reductions of the order of 50% are required to reduce ozone by up to 18 ppb, in the Houston region. A 50% reduction in VOCs can result in significant ozone reductions of 30 to 40 ppb, especially when combined with a 50% NOx reduction. The results of the HDDM analysis show that the Houston airshed in 2000 was of mixed sensitivity to VOC and NOx, consistent with the results of the previous DDM-SAPRC sensitivity analysis conducted by Pun et al. (2005).

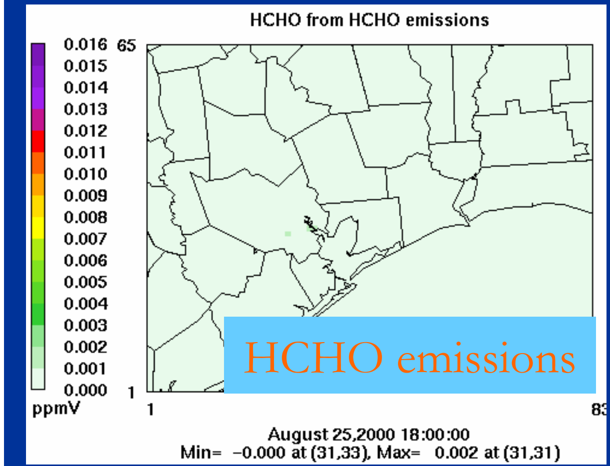
Other preliminary HDDM Simulations

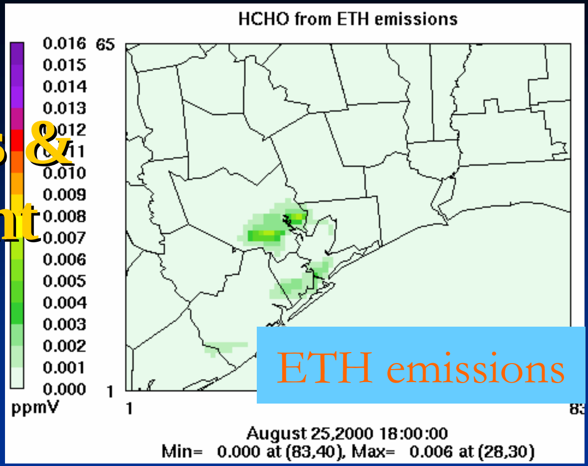
- Case 4: Sensitivity of VOC species to HCHO
  - Emissions of HCHO, ISOP, ETH, OLE, ALK, and ARO → 70 %
- Case 5: Contribution of biogenic NO and VOC emissions
  - GloBEIS vs. BEIS, spatial and temporal variation → 20 %

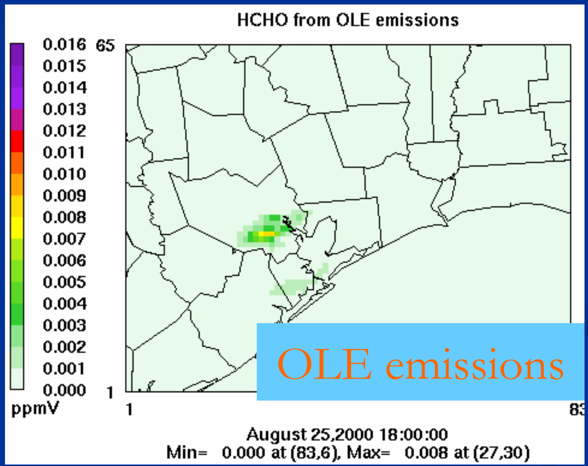
Case 4:
HCHO Concentrations & Source Apportionment

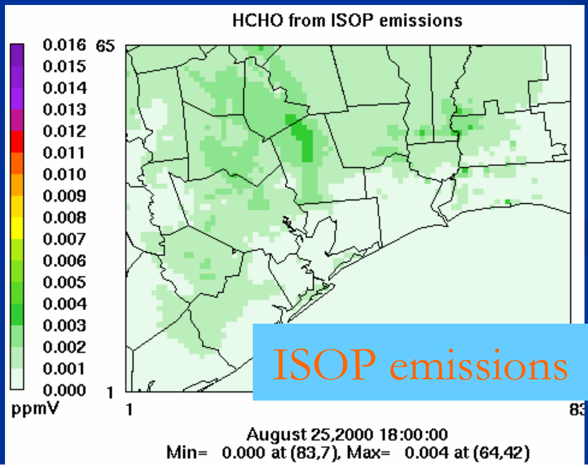
Snapshots for August. 25th, 2000 at 12 CST

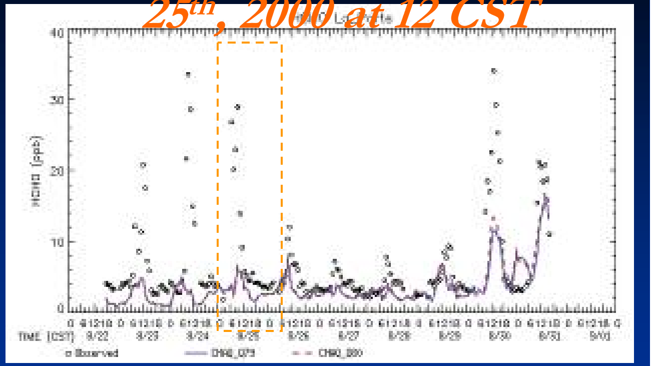


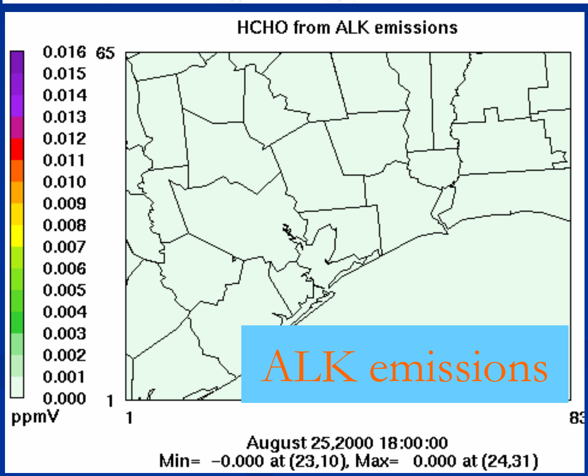


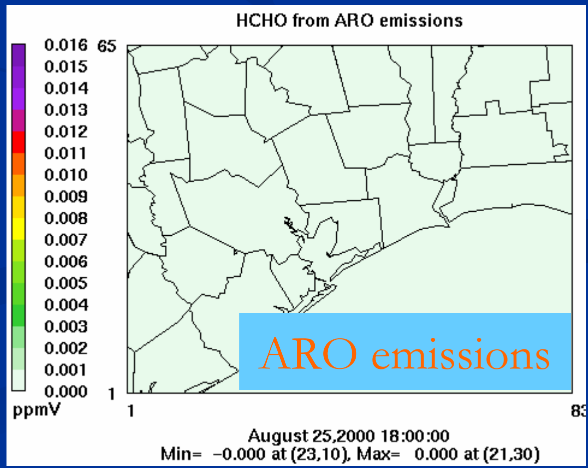












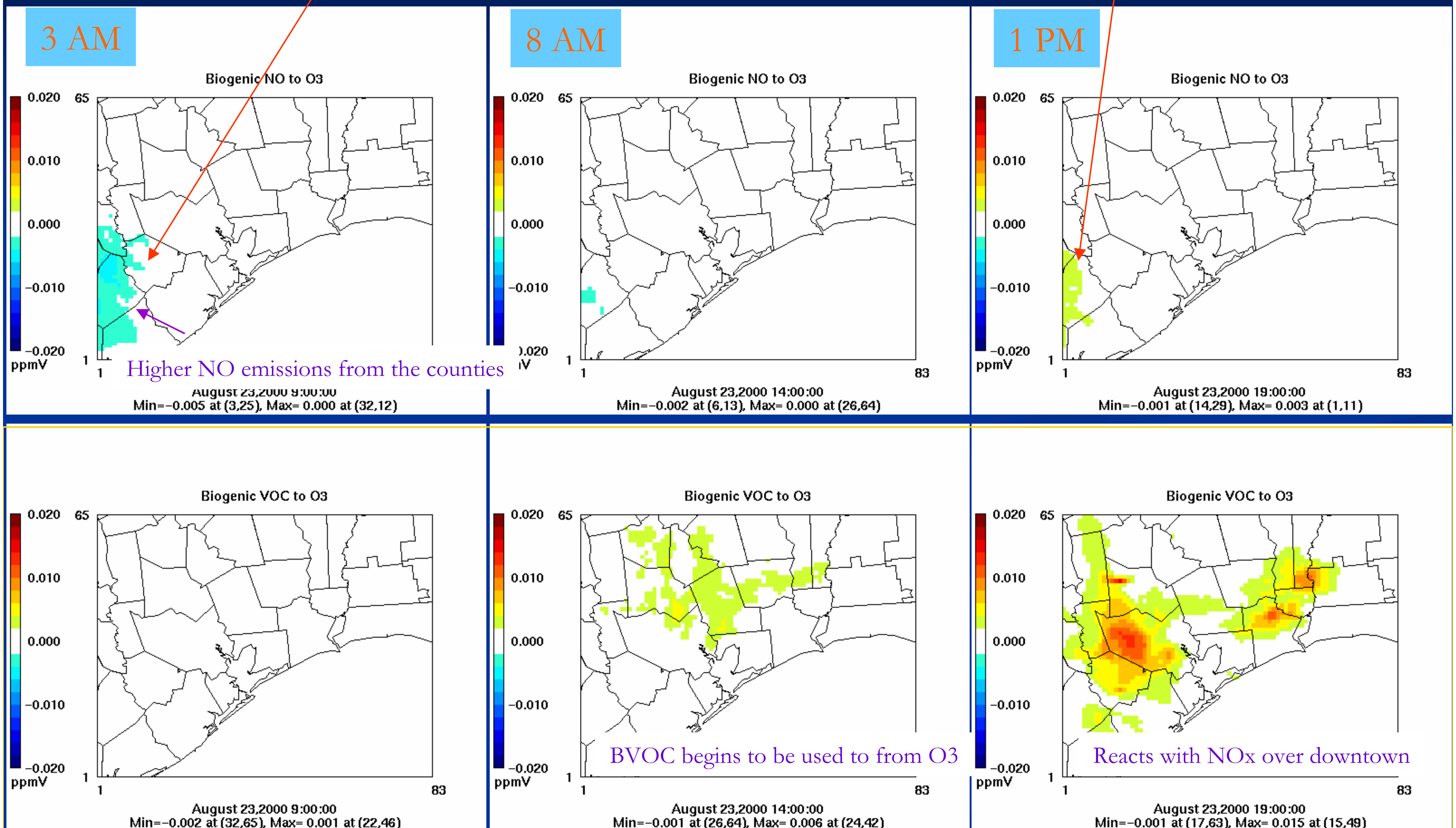
Time in figures UTC

Case 5: Biogenic NO and VOC to O3

Snapshots for August. 23rd, 2000

O3 titration by night

O3 formation by daytime



Time in figures UTC