Evaluating NO_x Emissions Using Satellite Observations

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Overview

1. Approach

Top-down assessment of NO_x emissions at a sector level using

- Atmospheric NO_2 columns retrieved from satellites
- In-situ aircraft sampling
- Calculations by regional chemical-transport model

2. Applications

NO_x controls on Eastern US power plants

S.-W. Kim et al. (2006), Satellite-observed US power plant NO_x emission reductions and their impact on air quality, *Geophys. Res. Lett., 33,* L22812, doi:10.1029/2006GL02774

NO_x emitted from Western US power plants and urban areas

S.-W. Kim et al. (2009), NO2 columns in the western United States observed from space and simulated by a regional chemistry model and their implications for NOx emissions, *J. Geophys. Res., 114,* D11301, doi:10.1029/2008JD011343

NO_x emitted from Texas power plants, cities, industry, & ports

S.-W. Kim et al. (2011), Evaluations of NOx and highly reactive VOC emission inventories in Texas and their implications for ozone plume simulations during the Texas Air Quality Study 2006, *Atmos. Chem. Phys.*, *11*, 11361–11386

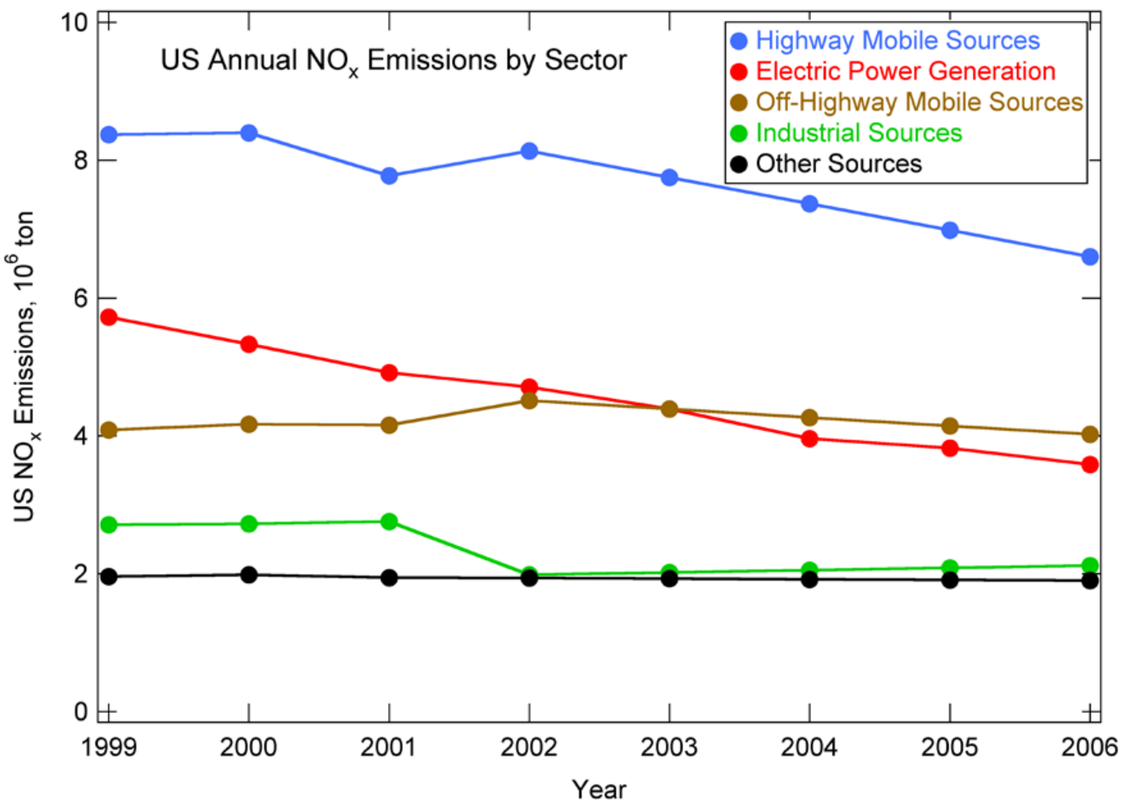
J. Brioude et al. (2011), Top-down estimate of anthropogenic emission inventories and their interannual variability in Houston using a mesoscale inverse modeling technique, *J. Geophys. Res., 116*, D20305, doi:10.1029/2011JD016215

3. Conclusions

Motivation

Bottom-up emissions inventories are generally based on process models that rely on emission factors and activity data derived from a few representative measurements and a variety of estimation techniques. Ideally, inventories would be evaluated with independent information based on observations.

Example: US EPA National Emissions Inventory



EPA National Emissions Inventory (NEI) Air Pollutant Emissions Trends Data
http://www.epa.gov/ttn/chief/trends/index.html

- 1999-2006 decreases > 20%
- 2 biggest sectors show largest declines
  - Highway and most other sources:
    - *emission process models**
    - *(emission factors + activity data)*
  - Electric power generation:
    - *in-stack measurements*

**Examples of emission process model quantities for mobile sources*

- Emission factors
  - Gasoline vs. diesel vs. ethanol
  - Vehicle operation: speed, cold start, load
  - Condition of vehicle
- Activity data
  - Fuel use
  - Fleet composition
  - Road network
  - Traffic volume

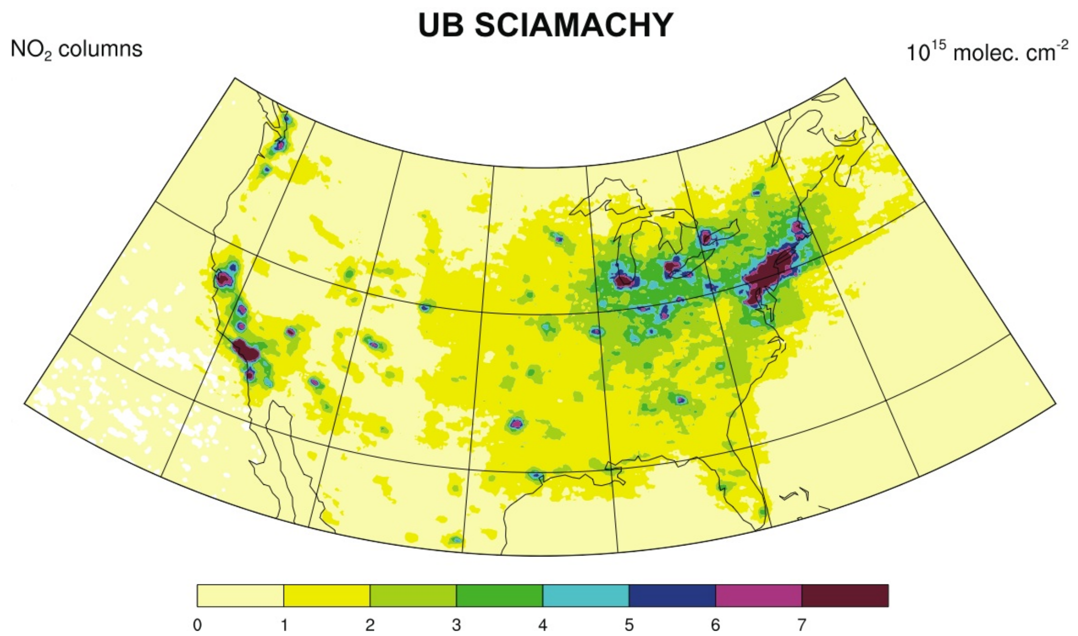
Top-Down Emissions Assessment by Satellites

NO_x emissions $\propto$ NO_2 columns
summer day $\Rightarrow$ short NO_x lifetime

NO_2 columns retrieved from satellites and calculated by chemical-transport models

- ➢ Understand, improve NO_x emissions
- ➢ Understand uncertainties of methods

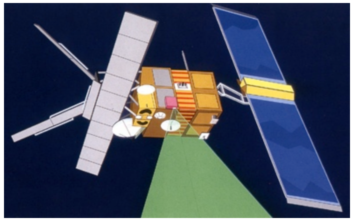
Martin et al., 2003; Beirle et al., 2003; Boersma et al., 2004; Richter et al., 2005; van der A, 2006; Kim et al., 2006 & 2009; etc

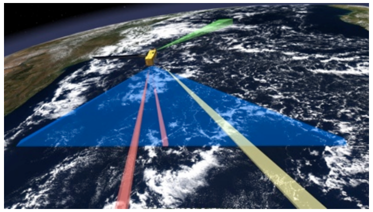


Our research using satellite NO_2 columns to understand NO_x emissions

- Information on absolute NO_x emission strengths, temporal trends, sectoral partitioning
- US power plants measure in-stack NO_x emissions (CEMS)
  - ➢ More accurate modeled NO_2 columns
  - ➢ Evaluate & calibrate satellite columns over power plants with chemical-transport model
- Satellite NO_2 columns used to evaluate inventories for other NO_x sources

Satellite NO_2 Column Retrievals





Satellite Instrument	Period	Overpass time	Global coverage	Pixel size
GOME (ERS-2)	1995/4-2003/6	10:30 LT	3 days	340 x 40 km^2
SCIAMACHY (ENVISAT)	2002/3-present	10:00 LT	6 days	60 x 30 km^2
OMI (AURA)	2004/7-present	13:30 LT	1 day	27 x 13 km^2 (nominal)
GOME-2 (METOP)	2007/3-present	09:30 LT	1.5 days	80 x 40 km^2

NO_2 Column Retrieval Process

1. Spectral fittings to get NO_2 slant column (S)
2. Subtraction of stratospheric NO_2 column
3. Calculation and application of air mass factor (AMF) to get NO_2 vertical column (V)

➔ $V = S / AMF$

AMF calculation

- *A priori* NO_2 profile
- *A priori* aerosol profile
- Aerosol optical depth
- Terrain height
- Surface albedo
- Temperature & pressure
- Radiative transfer equation

➢ *These quantities can come from models, climatologies, satellite data, etc.*

Chemical-Transport Modeling of NO_2 Vertical Columns

WRF-Chem

(***W***eather ***R***esearch and ***F***orecasting - ***Chem***istry model)

- www.wrf-model.org/WG11
- Simulates atmospheric chemistry online within WRF meteorological model
- Various modules for gas and aerosol chemistry, planetary boundary layer dynamics, aerosol and cloud microphysics, radiation, and convection

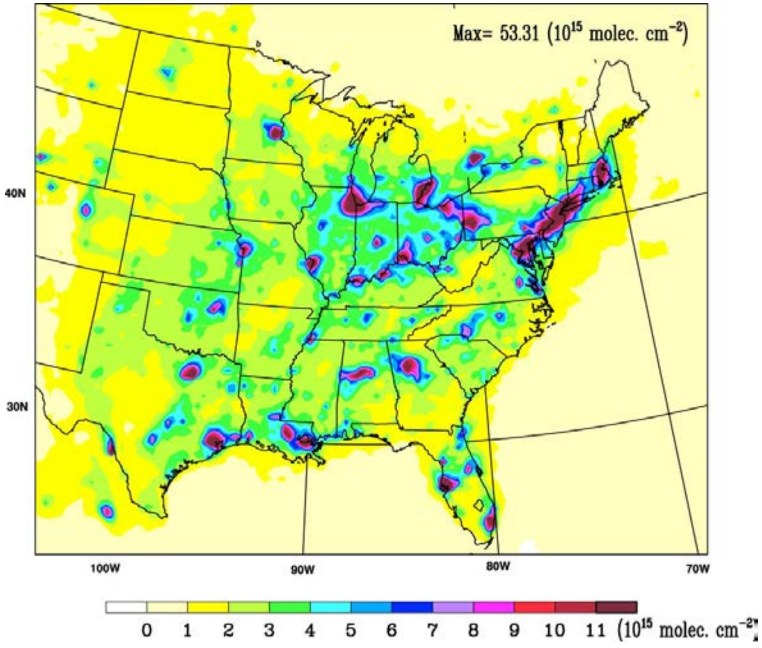
NOAA ESRL High Performance Computing System

Setup for these case studies

- Eastern US
    - Summer 2004 simulation period
    - 27 x 27 km^2 horizontal resolution
- Western US
    - Summer 2005 simulation period
    - 15 x 15 km^2 horizontal resolution
- Texas
    - Summer 2006 simulation period
    - Model resolution: 20 x 20 km^2
- *Emissions*
    - EPA NEI1999 and NEI2005
    - CEMS power plant emissions

WRF-Chem Summer 2004 Average

NO_2 Vertical Columns



1. Eastern US Power Plants

Effects of NO_x controls on large point sources in the Eastern US beginning in the late 1990s

- Acid Rain Program, NO_x SIP Call, NO_x Budget Trading Program
- Focus on coal-burning power plants
- Improved burner technology, post-burner ammonia scrubbers

S.-W. Kim et al., *Geophys. Res. Lett.*, 2006

NO_x Controls at Eastern US Power Plants

Effects of NO_x controls on large point sources in the Eastern US beginning in the late 1990s

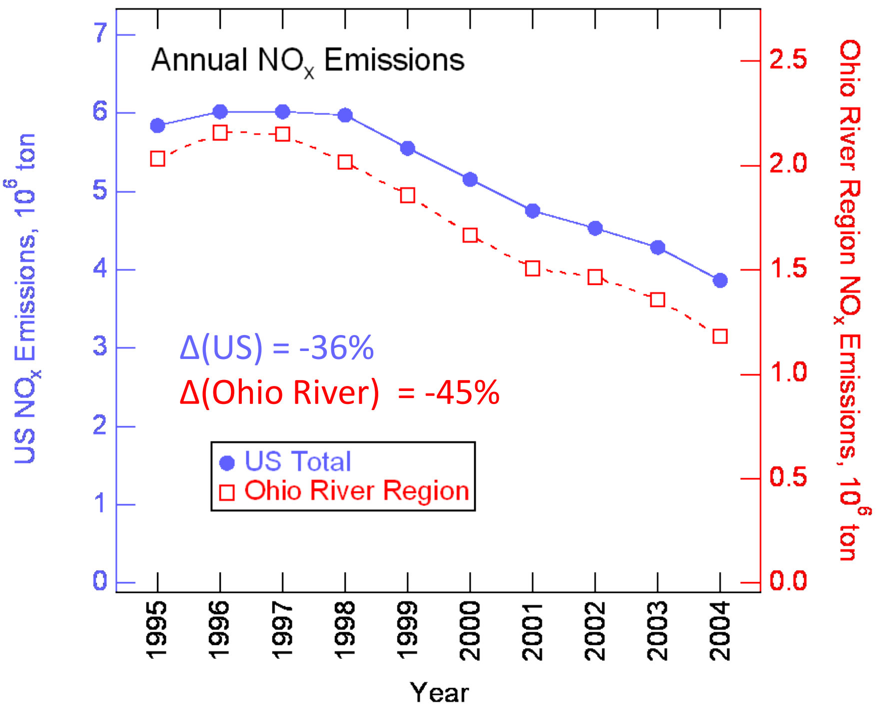
- National and regional pollution control programs
- Focus on coal-burning power plants
- Improved burner technology, post-burner scrubbers

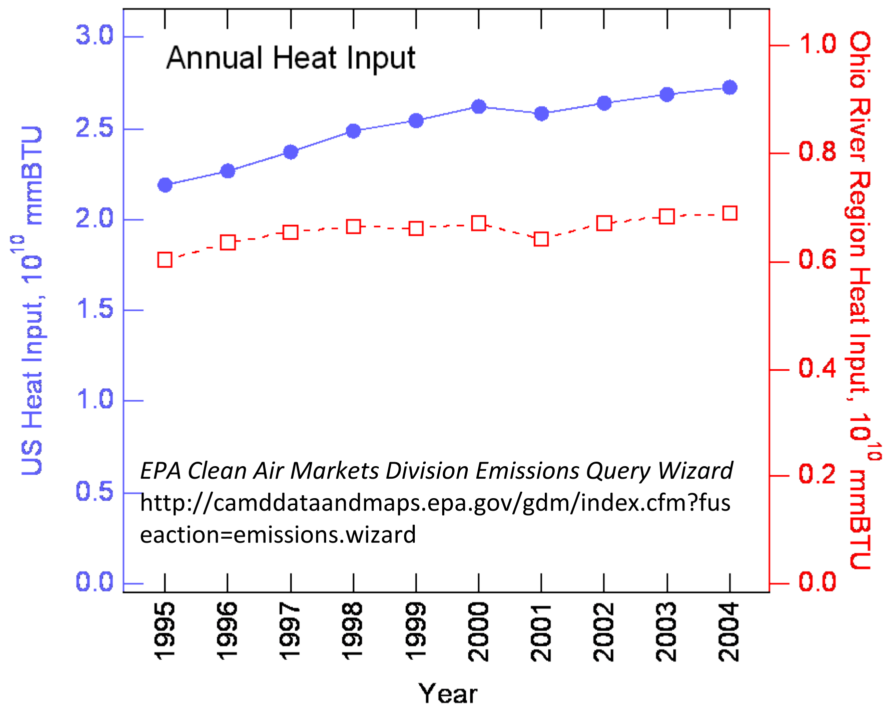
Continuous Emission Monitoring Systems (CEMS)

- *Stack measurements of hourly NO_x, SO_2, and CO_2 emissions made by utility companies*
- *Data for 966 facilities in 1999 and 1427 facilities in 2004*

CEMS data show:

- Substantial NO_x emission reductions since late 1990's
- Amount of electric power generated has increased



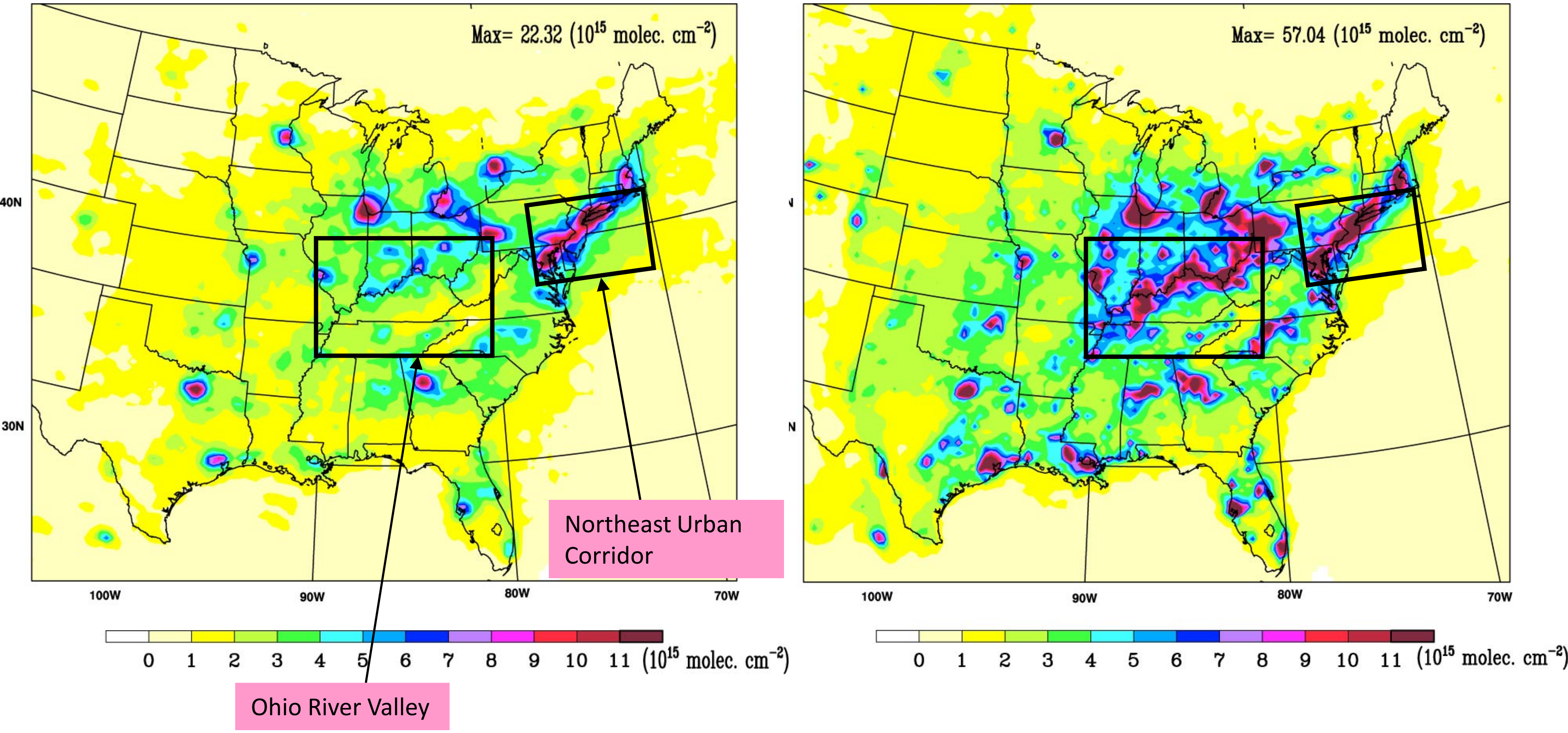


EPA Clean Air Markets Division Emissions Query Wizard
http://camddataandmaps.epa.gov/gdm/index.cfm?fuseaction=emissions.wizard

Eastern US Power Plant NO_x Controls Detected by Satellite

Summer 2004 Average NO_2 Vertical Columns

SCIAMACHY **WRF-Chem, Reference Emissions (NEI 99)**



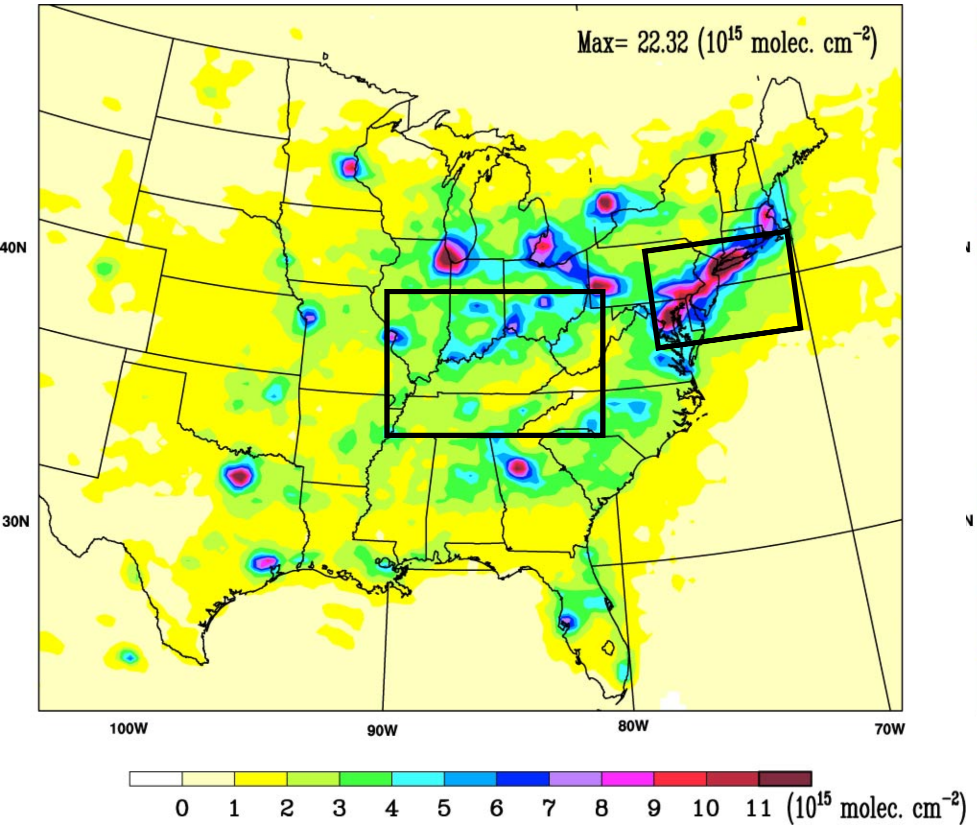
- Model reproduces satellite NO_2 vertical columns over urban areas
- Model NO_2 columns too large over power plants using 1999 emissions

S.-W. Kim et al., *Geophys. Res. Lett.*, 2006

Eastern US Power Plant NO_x Controls Detected by Satellite

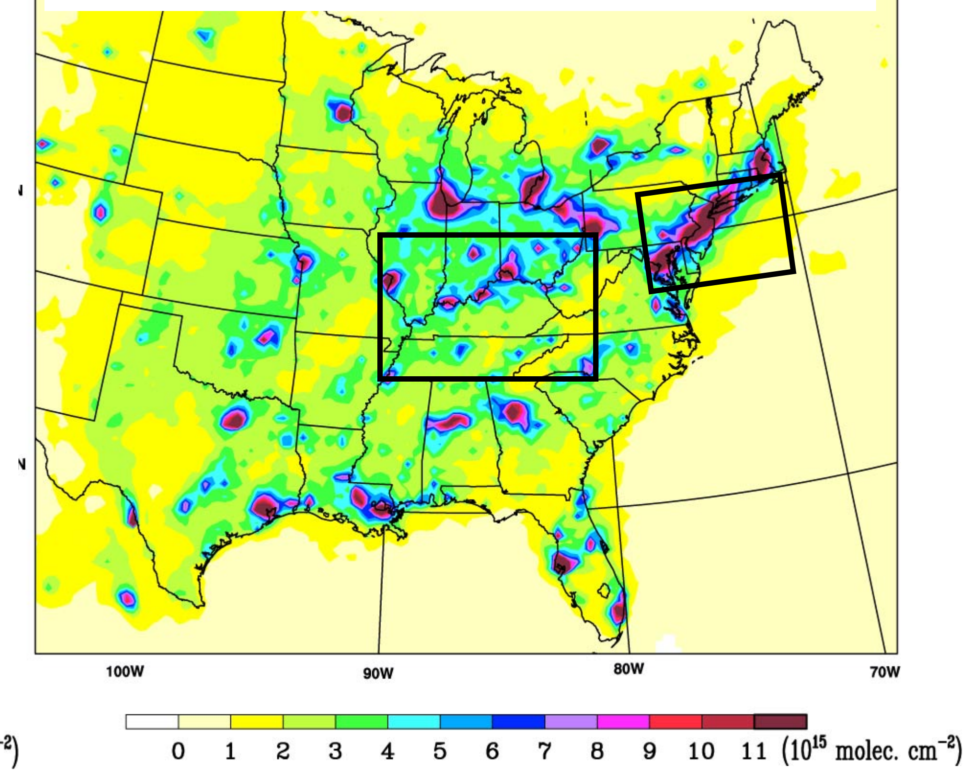
Summer 2004 Average NO_2 Vertical Columns

SCIAMACHY



WRF-Chem, Updated Power Plant Emissions

- ➢Power plants = CEMS 2004 monthly averages
- ➢All other sources = NEI 1999

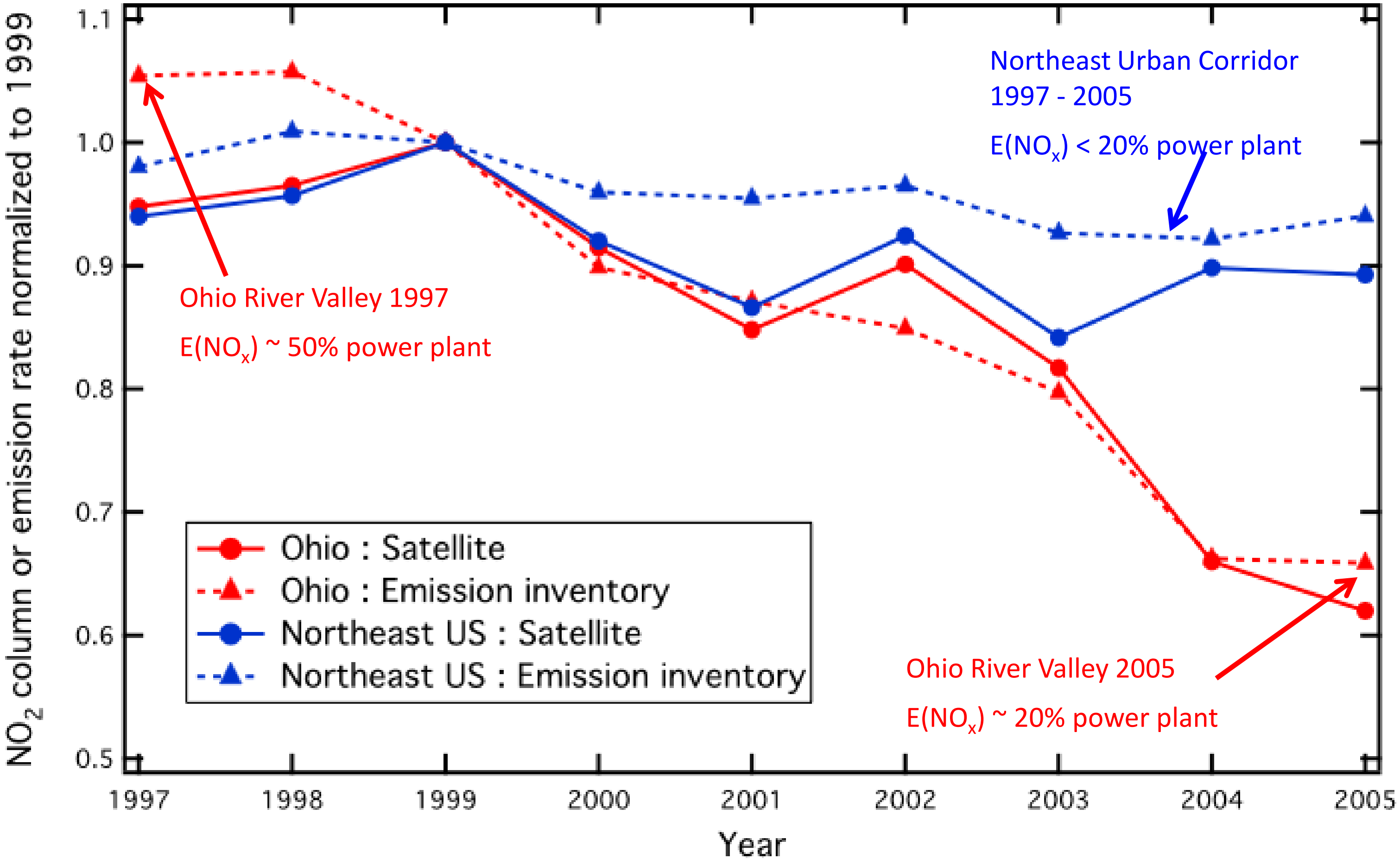


➢Model with summer 2004 power plant emissions agrees much better with satellite NO_2 columns over power plants

➢Satellite detects changes in Ohio River Valley from recent power plant NO_x emission controls

S.-W. Kim et al., *Geophys. Res. Lett.*, 2006

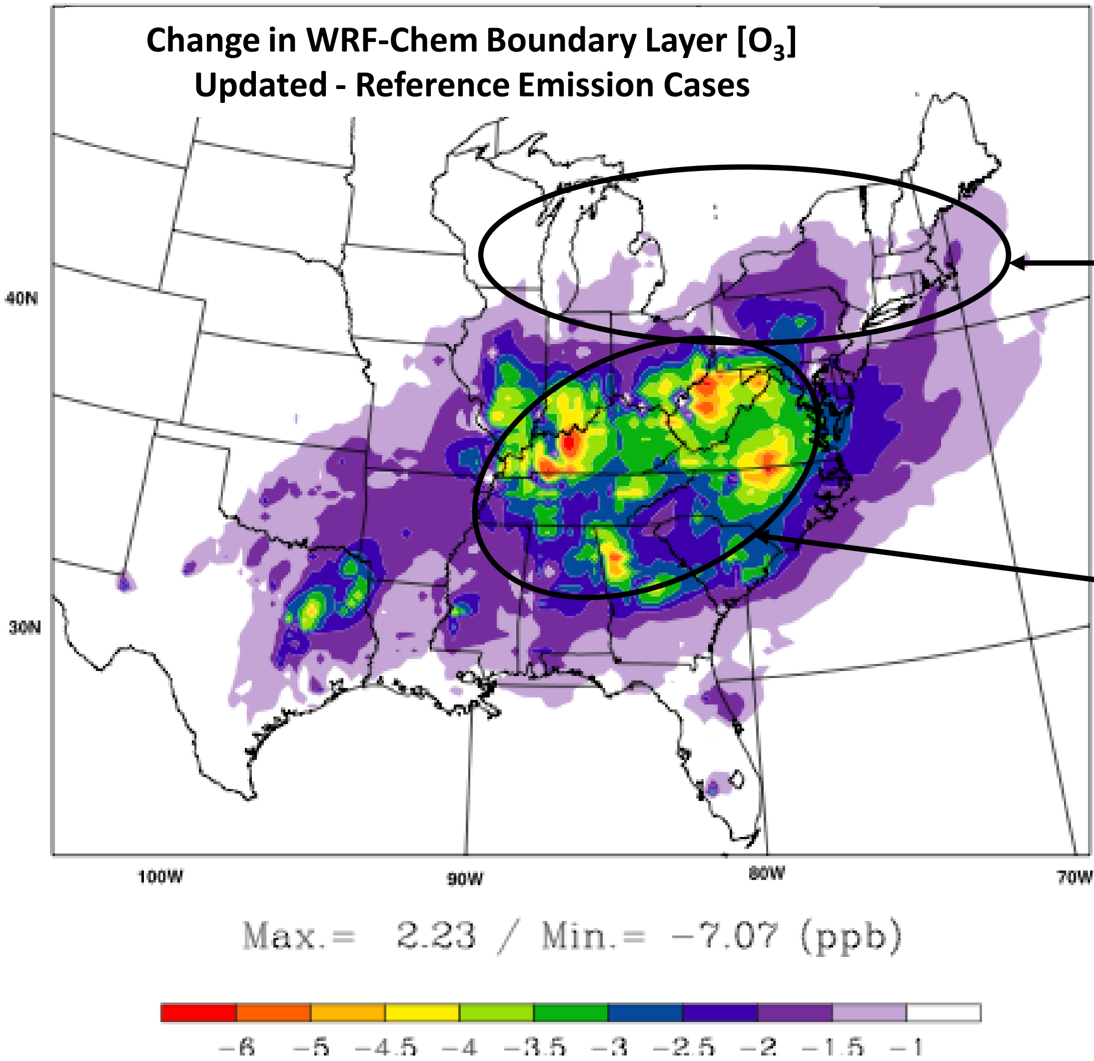
Trends in Eastern US Satellite NO_2 and NO_x Emissions



- Similar trends in satellite NO_2 columns and NO_x emissions
  - *Power plant NO_x controls have decreased NO_2 columns*
  - *Mobile NO_x emission changes smaller than those from power plants*

S.-W. Kim et al., *Geophys. Res. Lett.*, 2006

Boundary Layer O_3 Response to NO_x Emission Reductions



Average of all model output between 0 & 1 km at 20 UTC (1500 EST) for all days June-August 2004

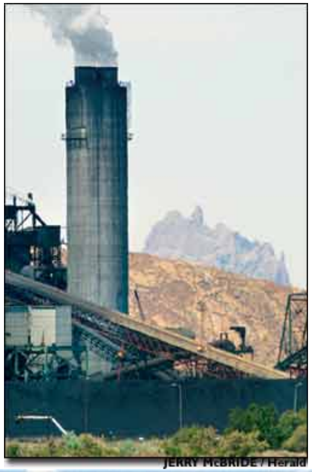
- *O_3 generally decreases in response to power plant NO_x emission reductions*
- *Small $\Delta[O_3]$ in northern US*

$\Rightarrow$ persistent cold fronts and unusually cold conditions in summer 2004

- *Up to 10% [O_3] decreases in Ohio River Valley, VA, NC, and GA*

S.-W. Kim et al., *Geophys. Res. Lett.*, 2006

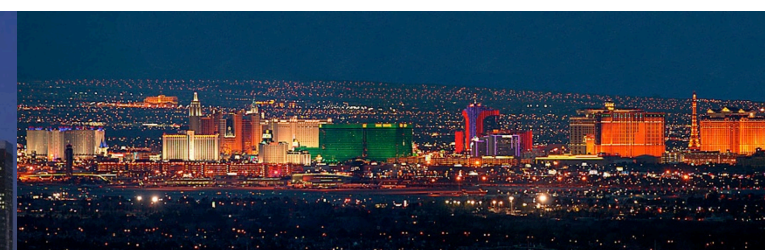
2. Western US Power Plants and Cities

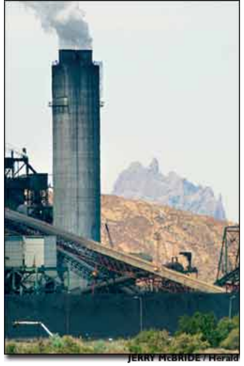


Use discrete satellite signals in Western US to evaluate NO_x emissions from individual power plants and urban areas

- Steady, well-known power plant emissions
  - "Calibrate" satellite and model algorithms
- Rapidly growing urban areas with lots of motor vehicles
  - How well are mobile source NO_x emissions understood?
  - Are overall NO_x emissions declining?

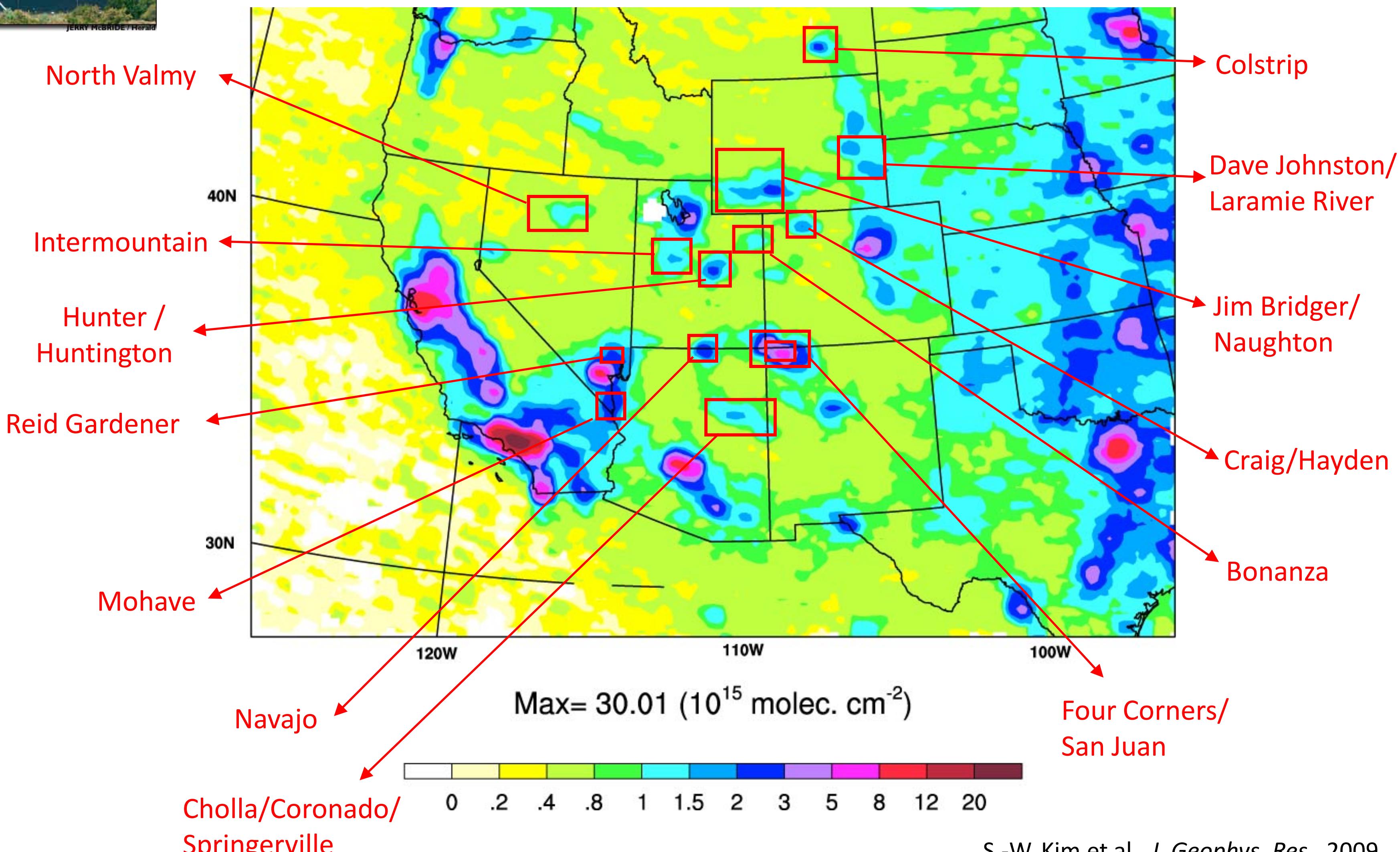
S.-W. Kim et al., *J. Geophys. Res.*, 2009





NO_x Emissions from Western US Power Plants

- *Isolated plants have discrete signatures in satellite retrievals*
- *Power plant emissions are measured continuously at each stack*
- *Currently no NO_x pollution controls on large coal-burning plants in the West*
- ➢ *"Calibration" for satellite-model comparison*

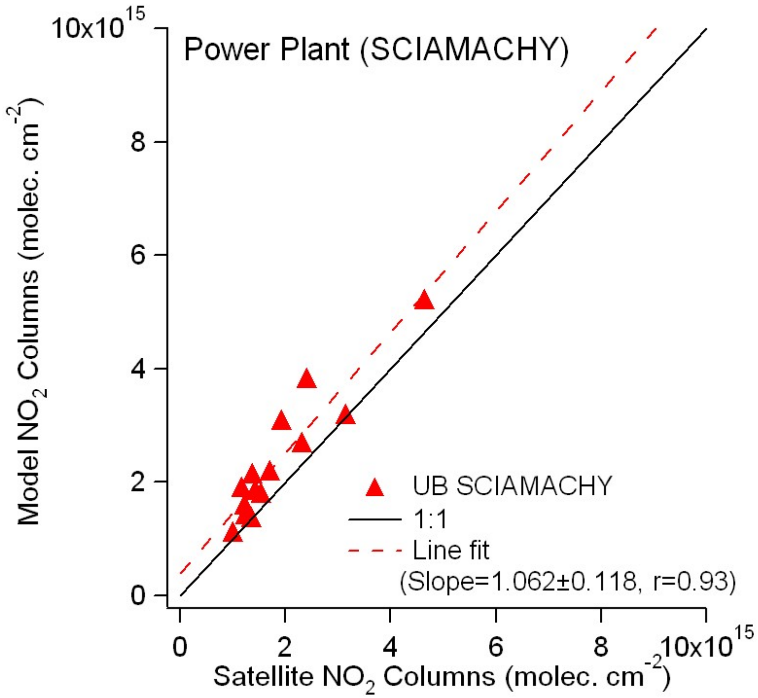


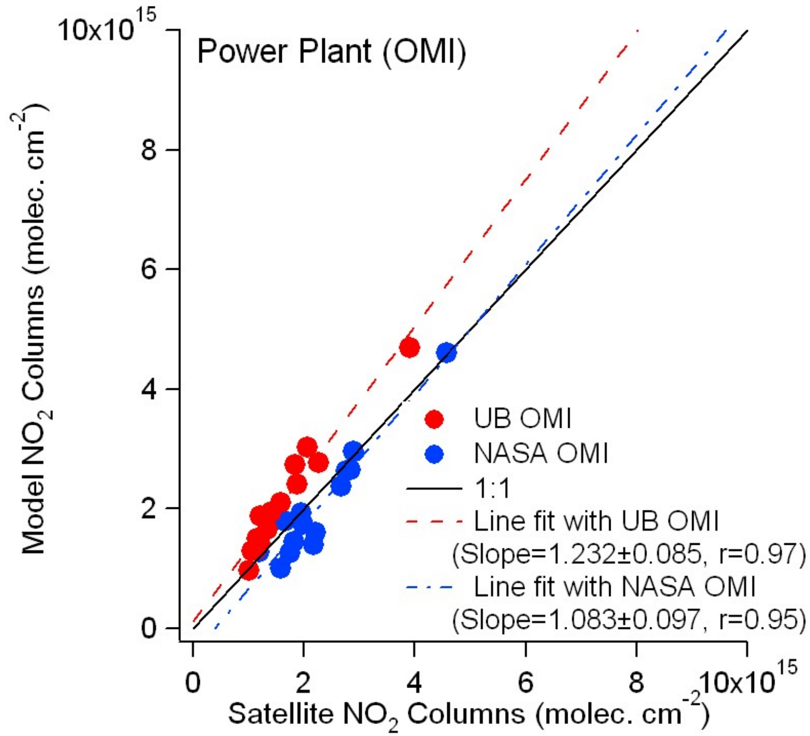
S.-W. Kim et al., *J. Geophys. Res.*, 2009

SCIAMACHY, Summer 2005

Satellite - Model NO_2 Column Comparison: Power Plants

Model uses observed emissions (CEMS) for power plants





Satellite and model NO_2 columns nearly equal over Western US power plants

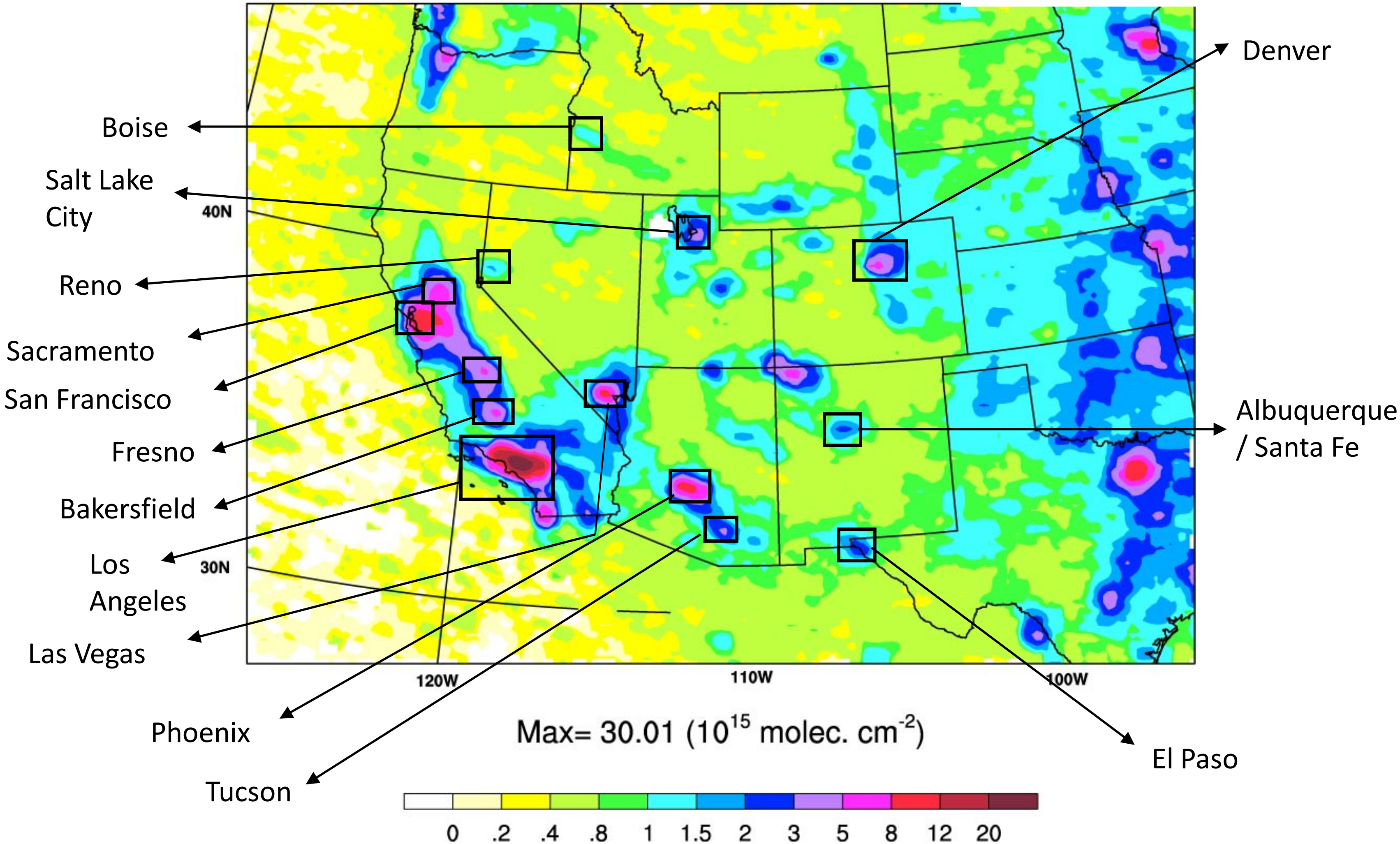
- Optimize satellite column retrievals and model parameterizations
- Model enables comparison of different satellite retrieval approaches
- Consistency for different retrievals: confidence in conclusions about emissions

S.-W. Kim et al., *J. Geophys. Res.*, 2009

NO_x Emissions from Western US Urban Areas

Build on satellite-model comparisons for power plants

➢*Evaluate urban area emission inventories and monitor changes*

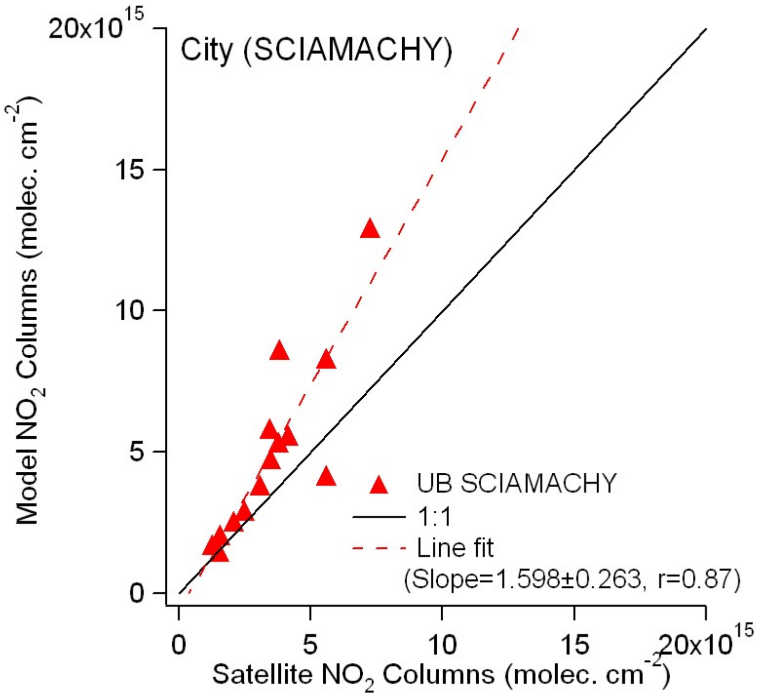


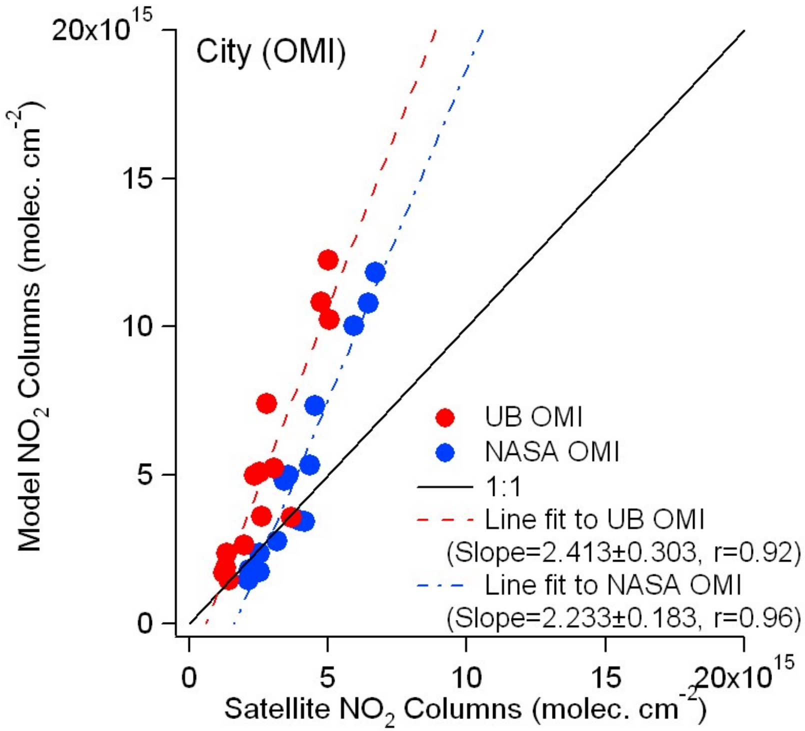
S.-W. Kim et al., *J. Geophys. Res.*, 2009

SCIAMACHY, Summer 2005

Satellite - Model NO_2 Column Comparison: Urban Areas

Note: weekend days are omitted in this analysis



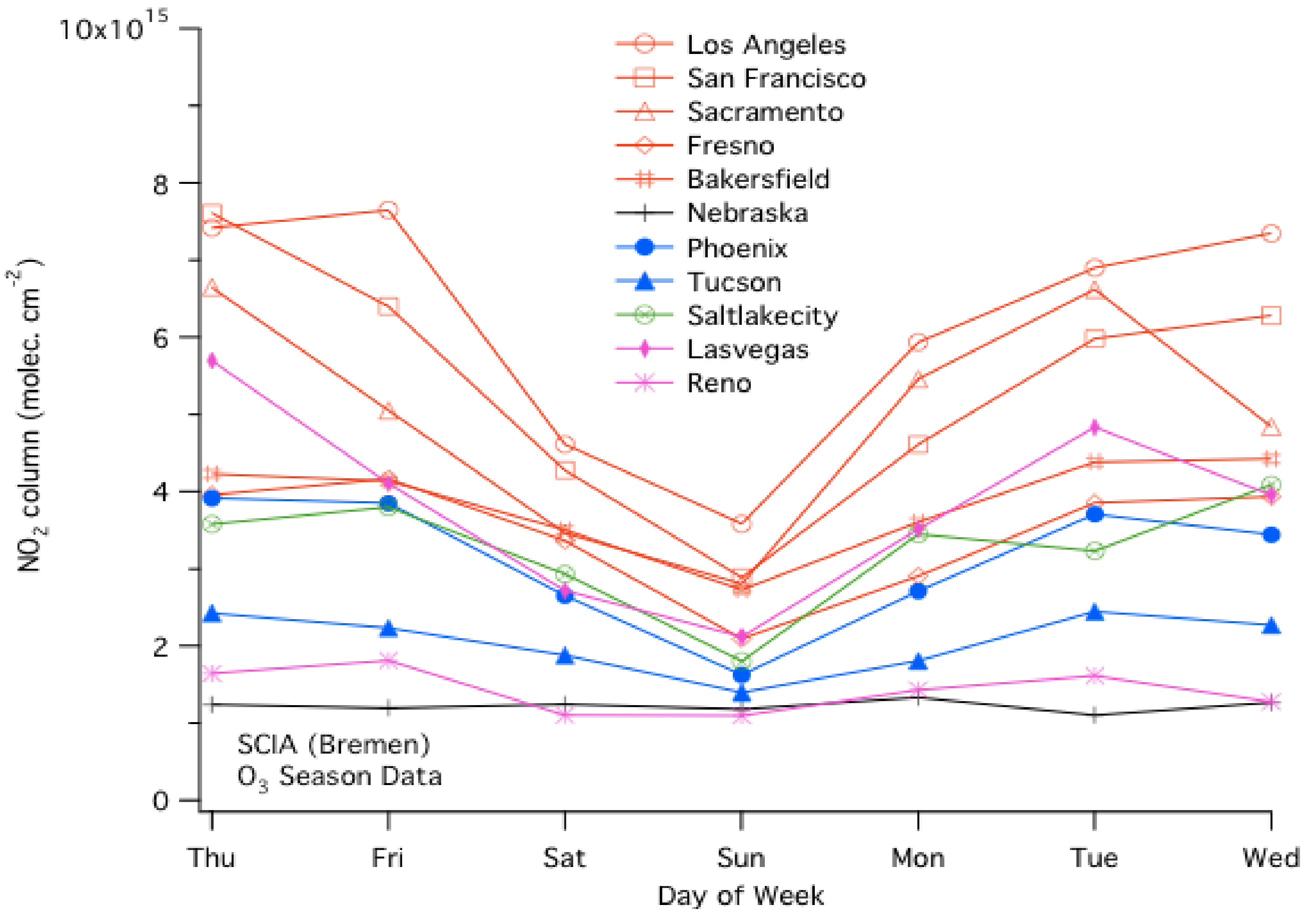


Large satellite - model NO_2 column differences over many Western US cities

- Urban emissions not well represented by 1999 inventory
- Urban model NO_2 columns higher than satellite retrievals
- Trends in NO_x emissions since 1999?

S.-W. Kim et al., *J. Geophys. Res.*, 2009

Day-of-Week Trends in Urban Satellite NO_2 Columns



Satellites show weekend decline in urban NO_2 columns

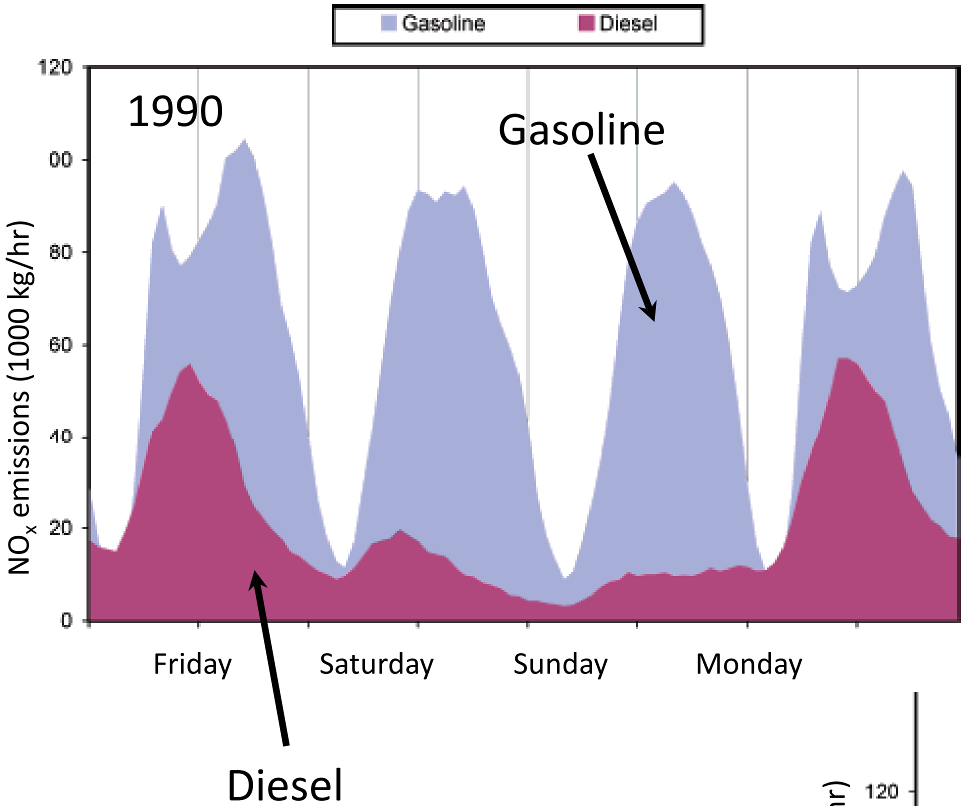
- Reduced traffic, particularly heavy-duty diesel vehicles
- Lower mobile source NO_x emissions on weekends
- Consistent with roadside monitoring

Model did not include day-of-week variations in NO_x emissions

Day of week changes in satellite NO_2 columns first reported by:
S. Beirle et al. (2003), Weekly cycle of NO_2 by GOME measurements: a signature of anthropogenic sources, Atmos. Chem. Phys., 3, 2225-2232

S.-W. Kim et al., *J. Geophys. Res.*, 2009

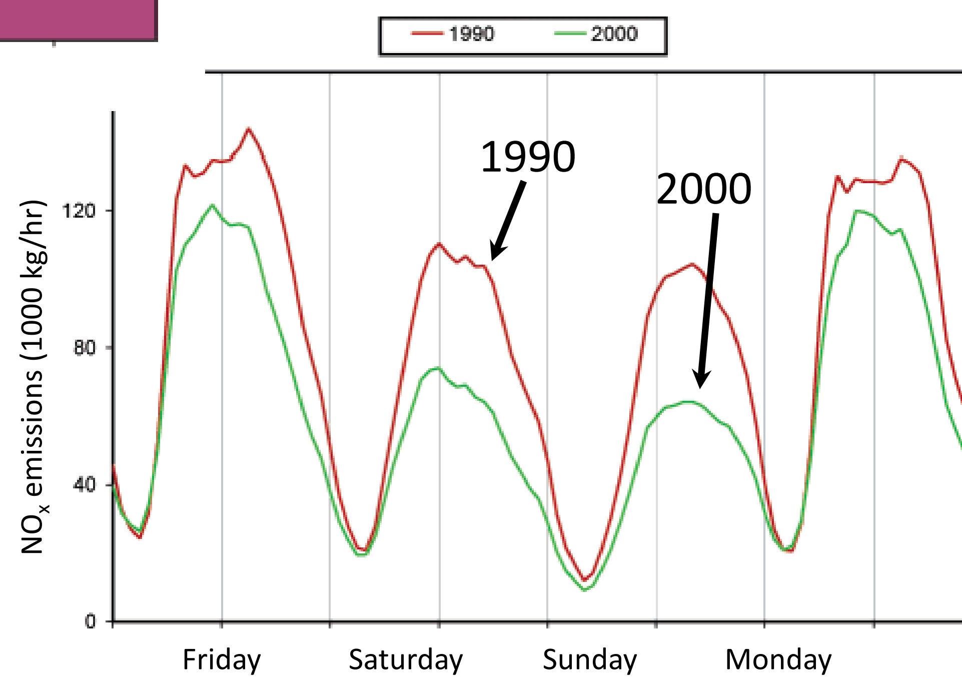
Day-of-Week Variations in Roadside NO_x Measurements



California statewide NO_x emissions from motor vehicles based on roadside monitoring, traffic counts, and fuel use
(Harley et al., UC Berkeley)

Weekend-weekday differences resulting from traffic patterns

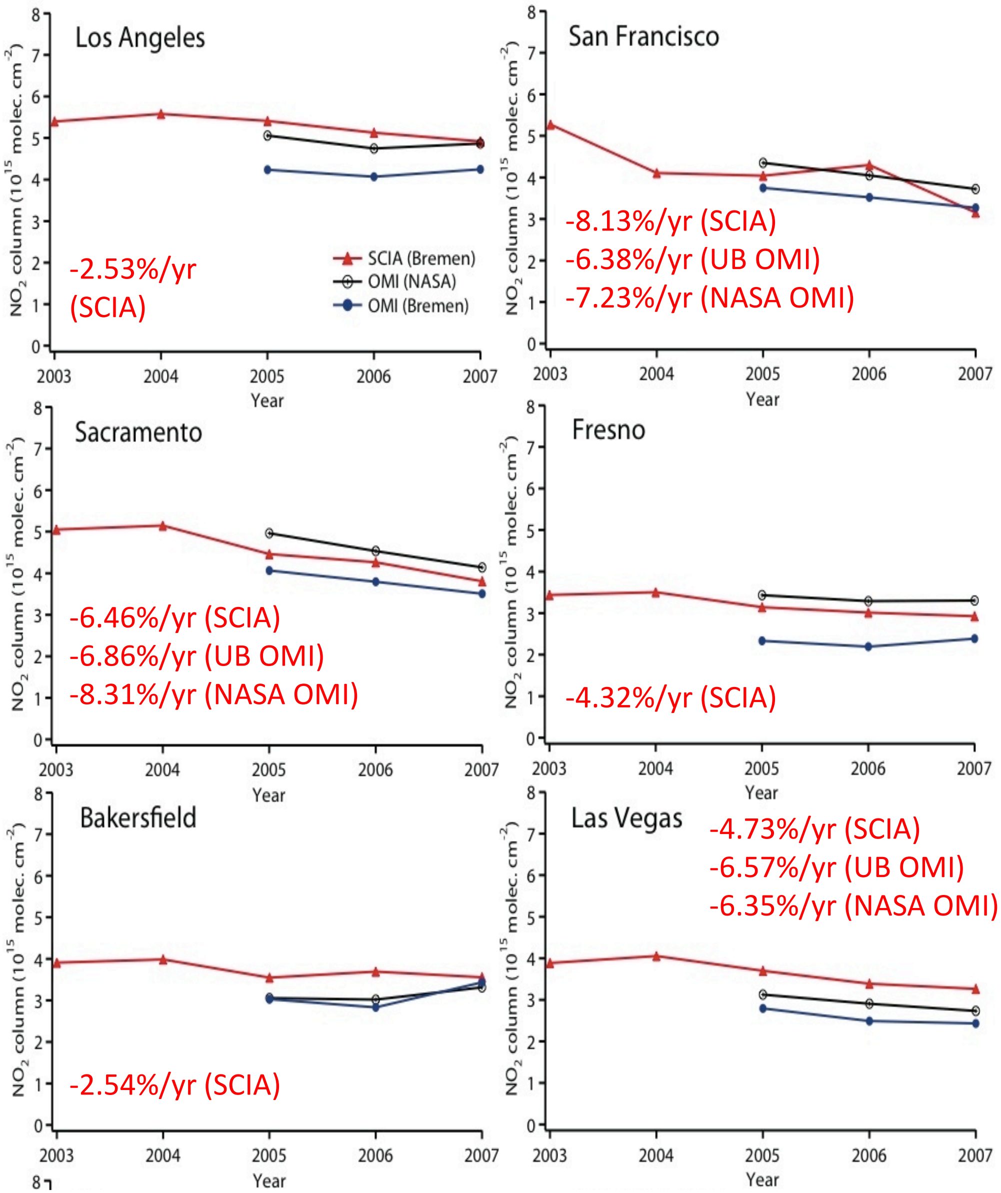
1990 - 2000 change in weekend-weekday difference:
cleaner gasoline vehicles



R. A. Harley et al., (2005), Changes in motor vehicle emissions on diurnal to decadal time scales and effects on atmospheric composition, *Environ. Sci. Technol.*, *39*, 5356-5362

Year-to-Year Trends in Urban Satellite NO_2 Columns

Left axis: NO_2 columns averaged over box



- Year-to-year declines in satellite NO_2 columns in many Western US cities
  - ➢ Coincide with NO_x declines seen by roadside monitoring (Ban-Weiss et al., 2008; Dallmann and Harley, 2010; Bishop and Stedman, 2008).
  - ➢ Effect of cleaner engines, especially light-duty gasoline vehicles

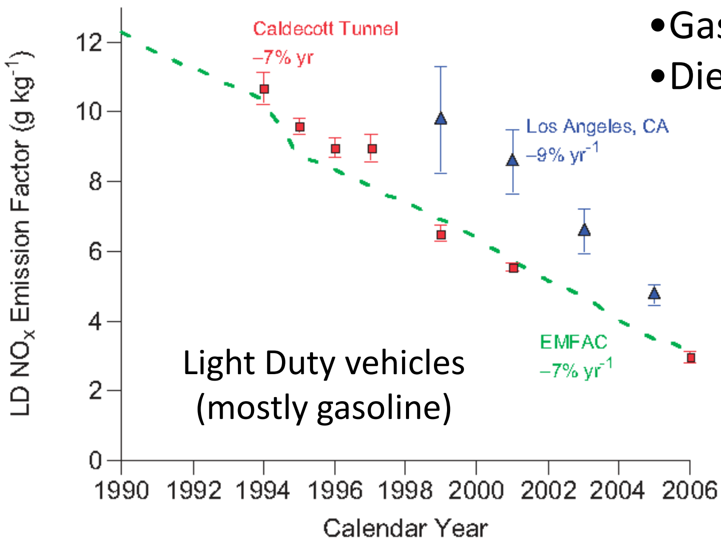
- Declining trends in aerosols are not included in retrievals
  - ➢ Declines in satellite NO_2 column might be underestimated

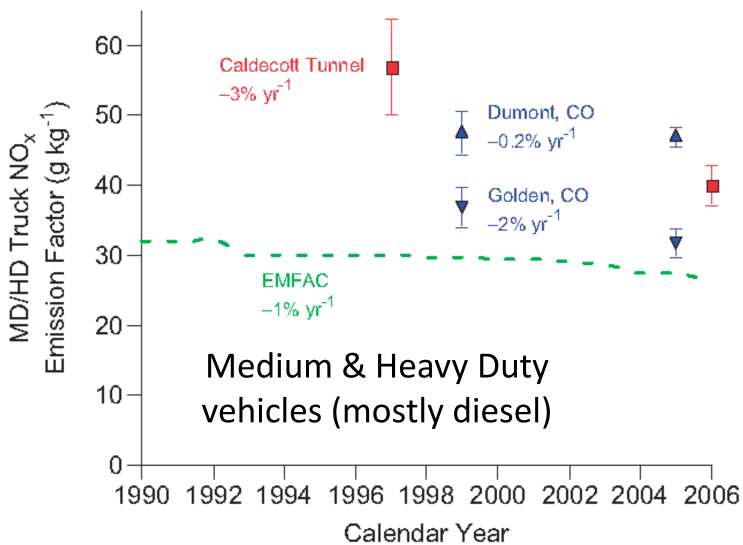
S.-W. Kim et al., *J. Geophys. Res.*, 2009

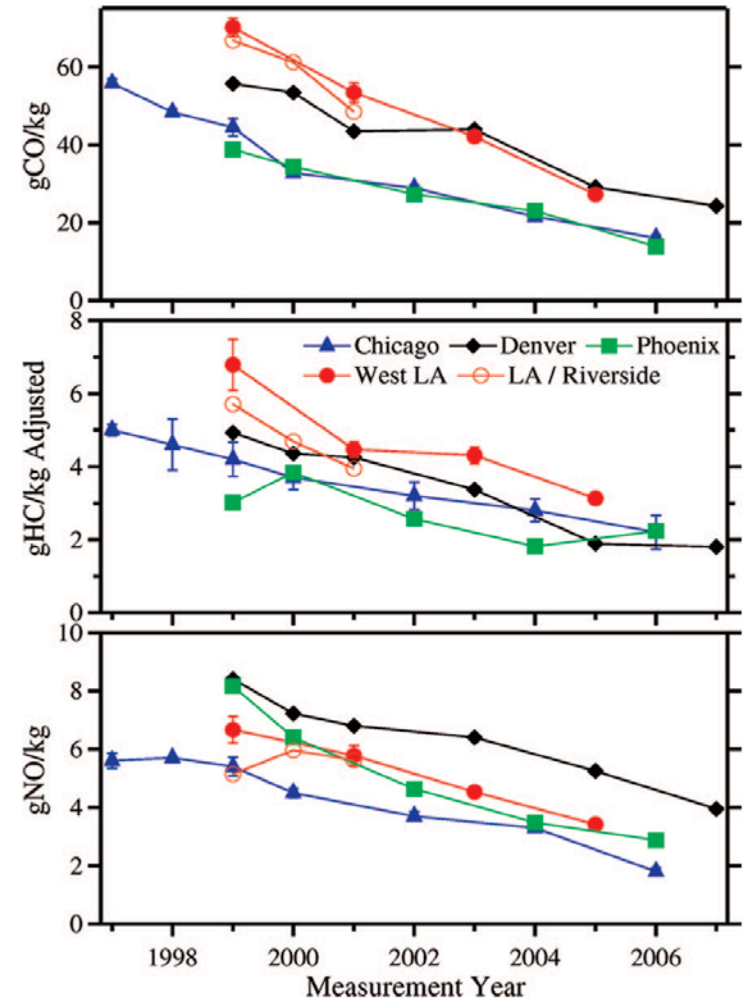
Year-to-Year Trends in Roadside NO_x Measurements

Roadside monitoring of fuel-specific NO_x emission factors
(g NO_x emitted per kg fuel burned)

- Gasoline: declined 6-9% per year since 1990's
- Diesel: small change, discrepancy with emission models







G. A. Ban-Weiss et al. (2008), Long-term changes in emissions of nitrogen oxides and particulate matter from on-road gasoline and diesel vehicles, Atmos. Environ., 42, 220-232

G. A. Bishop and D. H. Stedman (2008), A decade of on-road emissions measurements, Environ. Sci. Technol., 42, 1651-1656

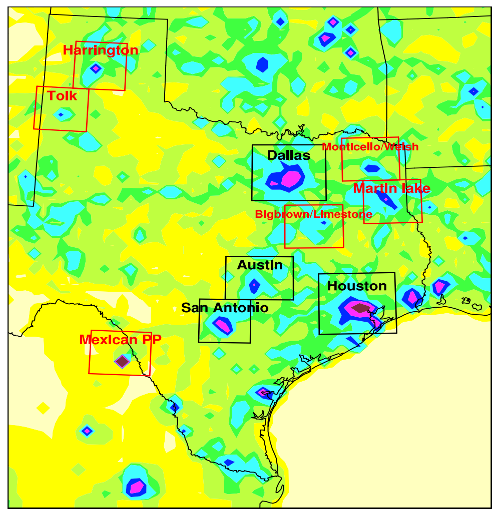
3. Texas Urban, Industrial, and Shipping Sources

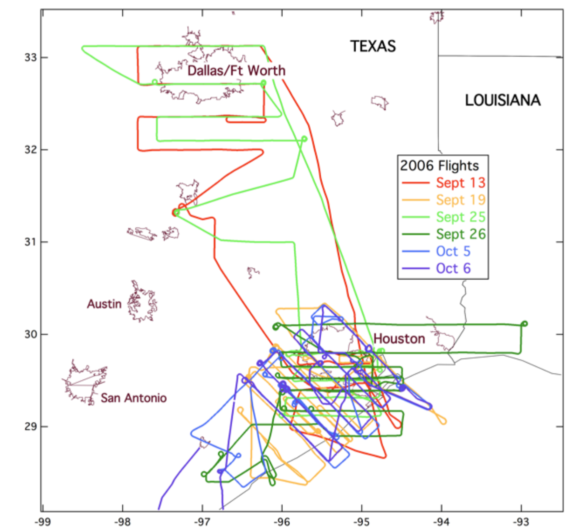
Eastern Texas has many large emissions sources, including power plants, urban areas, industry, and shipping

Two large field campaigns conducted in region in 2000 and 2006

Can we use satellites to establish trends in emissions sources?

Can we use aircraft measurements to inform model calculations, constrain satellite retrievals, and derive information on emission trends?

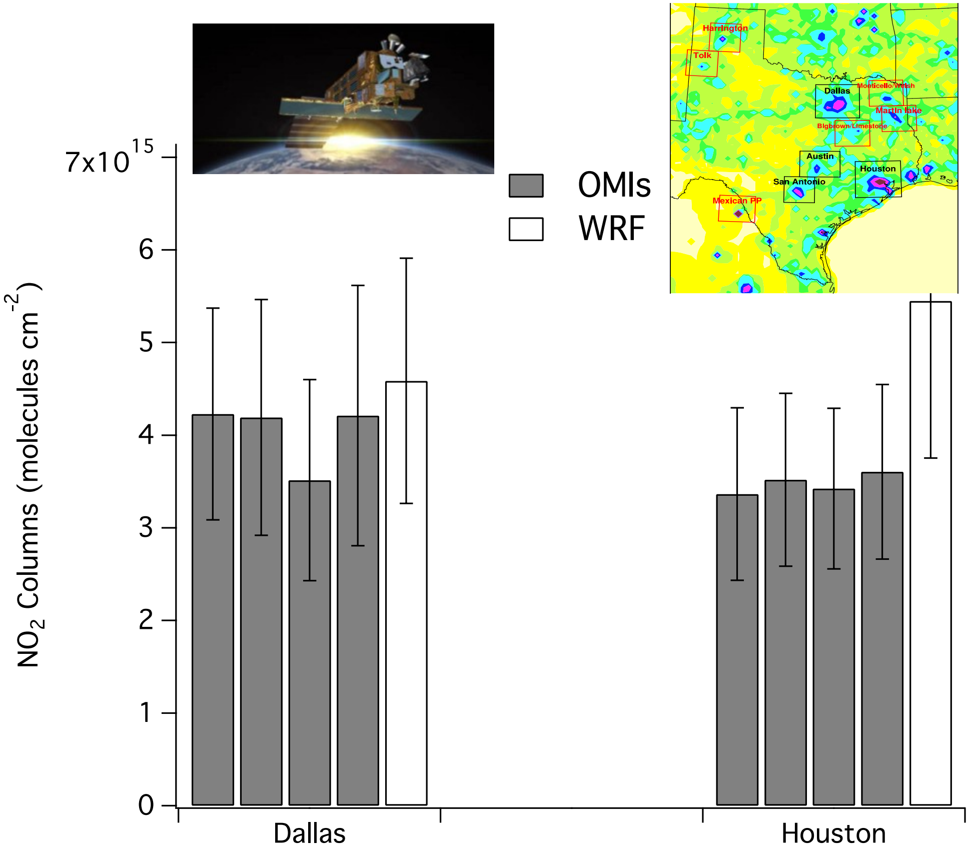




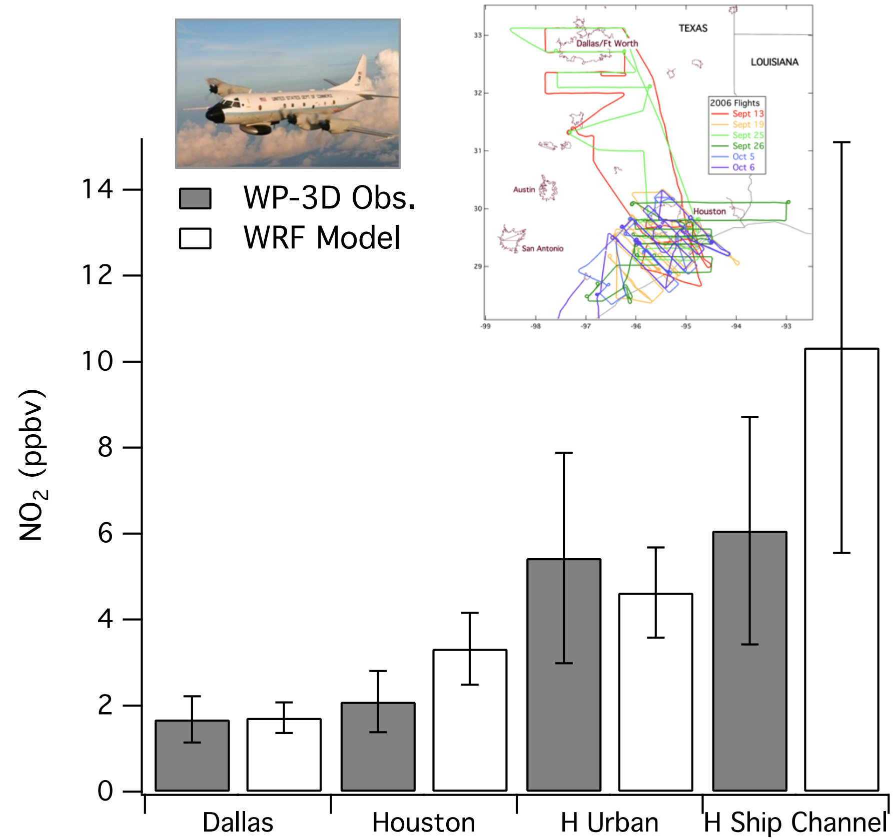
S.-W. Kim et al., *Atmos. Chem. Phys.*, 2011
J. Brioude et al., *J. Geophys. Res.*, 2011

Observed and Modeled NOx in Houston

Satellite (OMIs) vs. Model (WRF) NO_2



Aircraft (WP-3D) vs. Model (WRF) NO_2



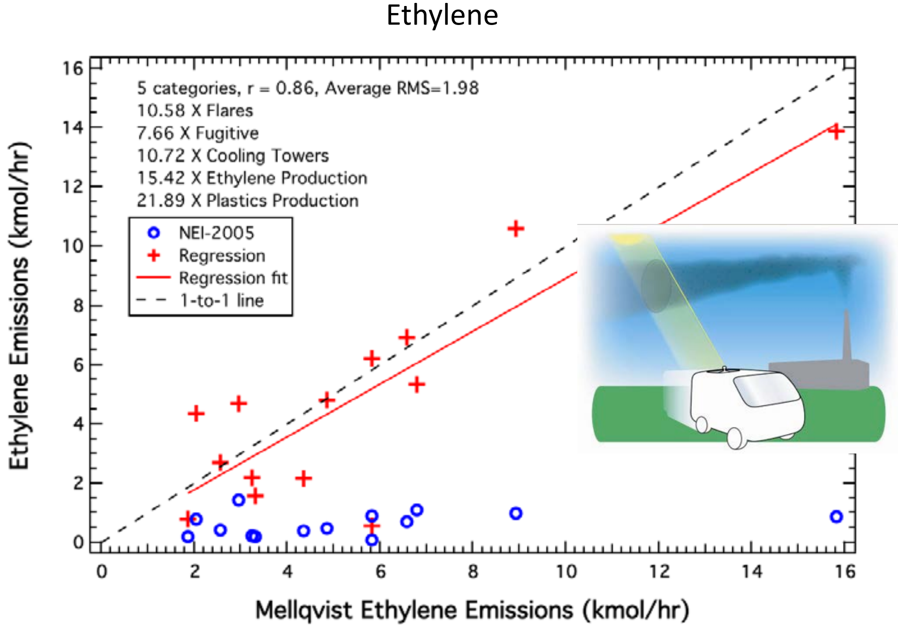
- Consistency: Satellite or aircraft observations vs. model with NEI2005
- Dallas: NO_x observations and model agree
- Houston: Model overestimates NO_x observations

➢ Errors in emissions estimates for industry, shipping

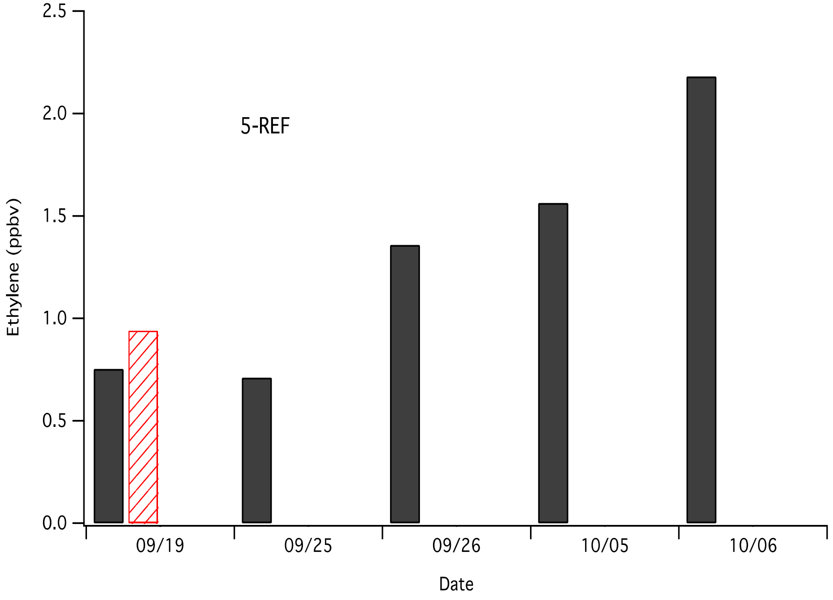
S.-W. Kim et al., *Atmos. Chem. Phys.*, 2011

Observed and Modeled VOCs in Houston

Inventory vs Van Observations
(Mellqvist, Solar Occultation Flux)



Aircraft (WP-3D) vs.
Model (WRF) VOCs

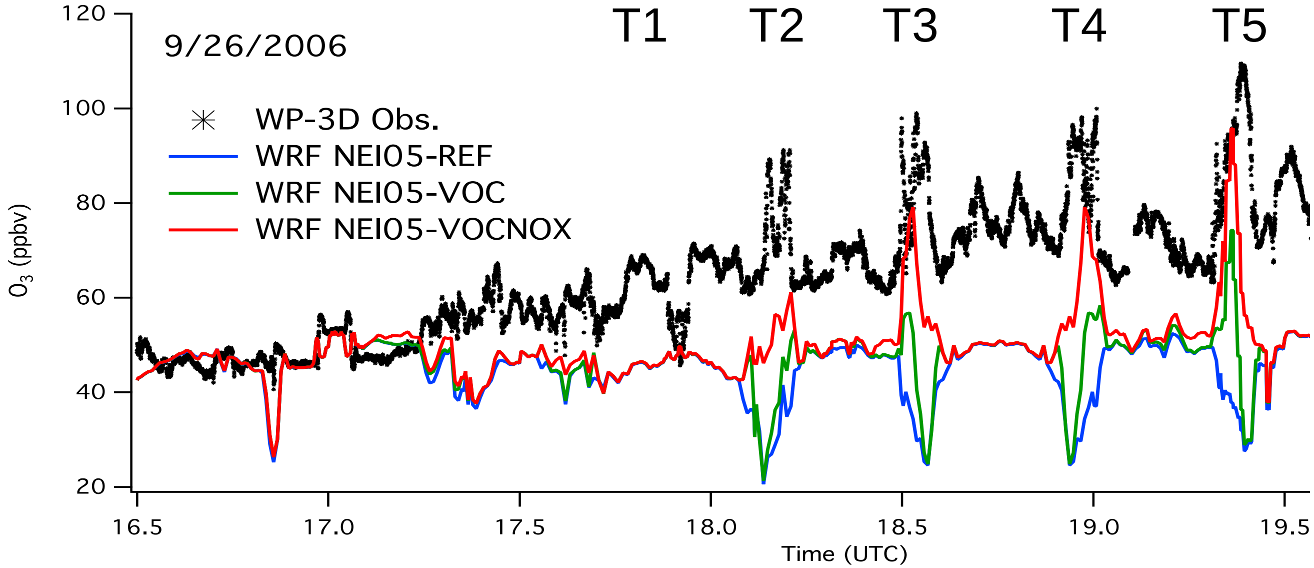


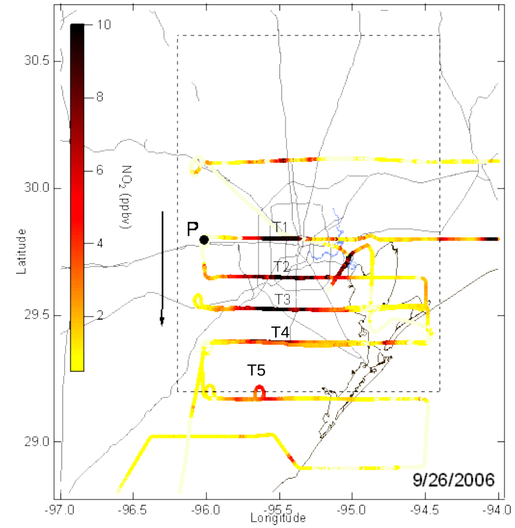
- Observed reactive VOC concentrations (ethylene, propylene) are 2-3 times higher than model or inventory predictions
- Consistent in aircraft and mobile surface observations in 2000 and 2006
- ➢ Large underestimates of business-as-usual VOC emissions from petrochemical industry

S.-W. Kim et al., *Atmos. Chem. Phys.*, 2011

Sensitivity of Houston Ozone to Emissions

Boundary layer O_3 observed by aircraft (WP-3D) and modeled (WRF)





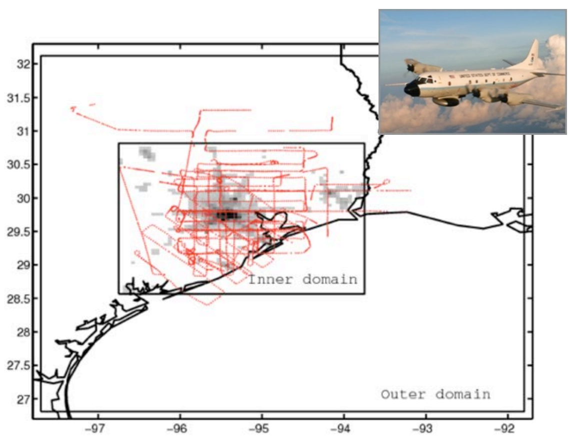
- Investigate sensitivity of modeled ozone to different emissions inventories
- Use measurements to adjust NOx and VOC emissions in sensitivity tests

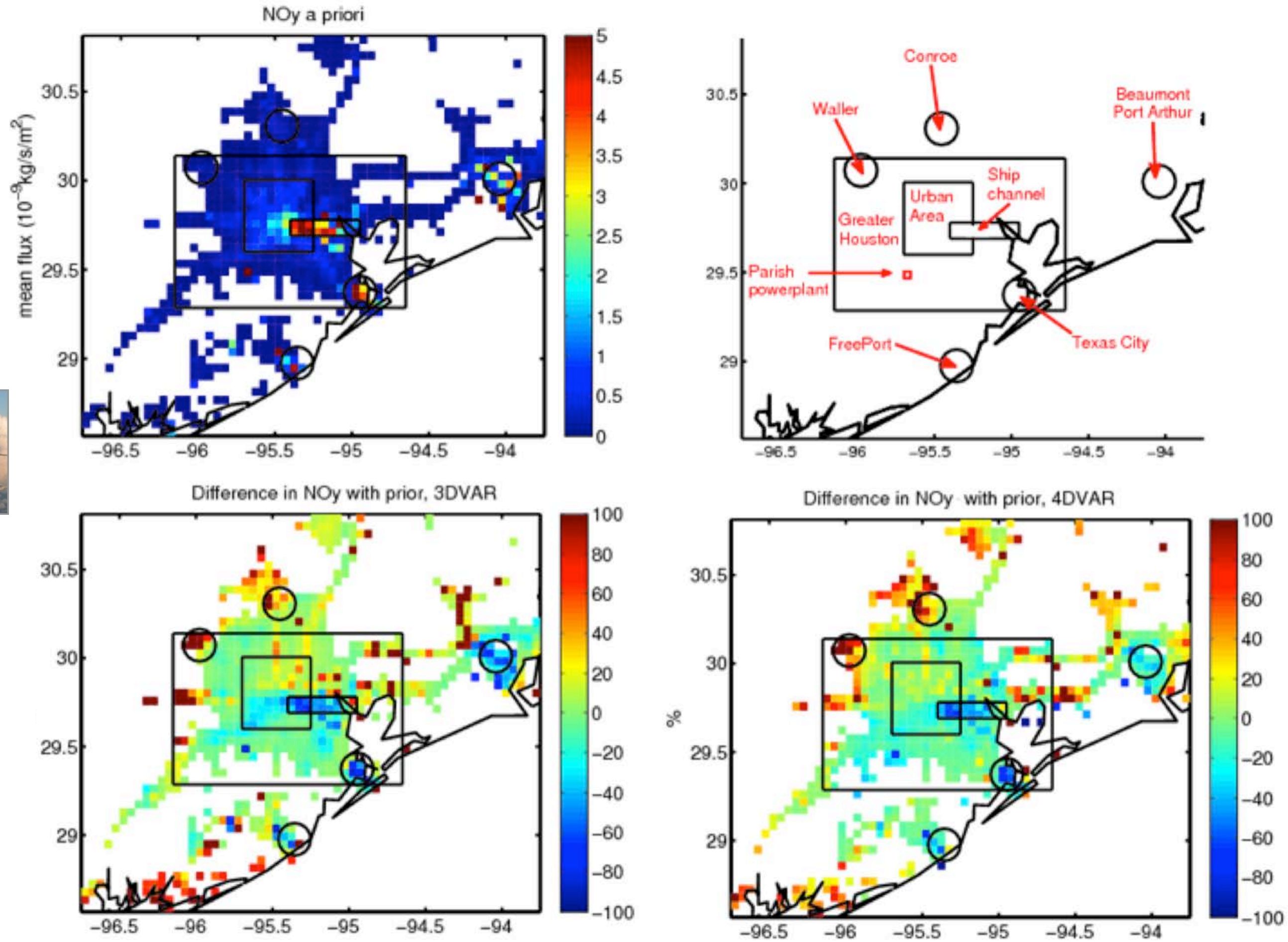
➢ Plume ozone very sensitive to both NOx and VOC emissions

S.-W. Kim et al., *Atmos. Chem. Phys.*, 2011

Inverse Modeling of Houston NOx Emissions

- Aircraft data drives NOx inverse modeling
- Optimize prior NOx emissions (NEI2005) with 3DVAR, 4DVAR





NEI2005 overestimates 2006 NO_x in Houston

- Greater Houston: NEI2005 prior – posterior = 28±4%
- Urban Houston: NEI2005 prior – posterior = 6±3%
- Ship Channel: NEI2005 prior – posterior = 50±5%

➢ Consistent with Kim et al. (2011) direct model-observational results

J. Brioude et al., *J. Geophys. Res.*, 2011

Conclusions

- Combination of satellite retrievals and chemical-transport modeling provides useful evaluation of NO_x emission inventories and trends
- Pollution control strategies have resulted in widespread changes to atmospheric NO_x levels that satellites can measure
- Large Western US power plants help calibrate NO_2 columns
- Biases in bottom-up urban, industrial, & port NO_x emissions identified with satellite data
- Aircraft observations provide confirmation for satellite and model

Issues for Further Research

- Capabilities for NO_x emissions inferred from satellite + model NO_2 columns
    - Absolute emission rates ≈ ±25% (best case)
    - Trends ≈ a few % per year (best case)
    - Identify/quantify emerging or sporadic sources (e.g. oil & gas production, fires)
- Satellite retrievals
    - Impact of aerosols, temperature, cloudiness, land use/albedo
    - External validation by aircraft & surface in-situ/remote-sensing observations
- Modeling
    - Impact of NO_x changes on O_3 and PM
- Interactions with inventory developers
    - Understand differences in top-down vs bottom-up approaches, national vs state data