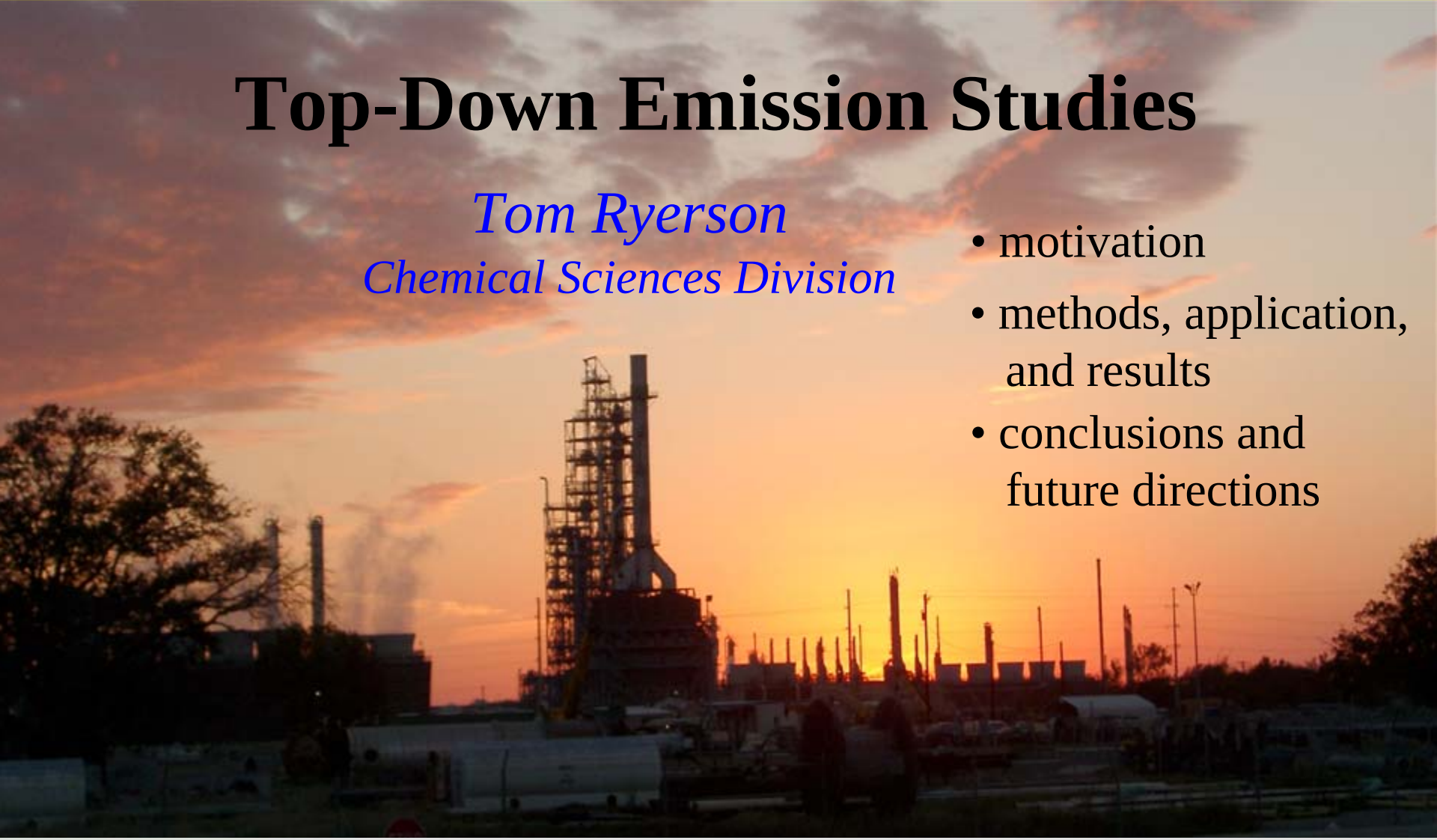
Top-Down Emission Studies

Tom Ryerson
Chemical Sciences Division

- motivation
- methods, application, and results
- conclusions and future directions



NOAA Earth System Research Laboratory

ESRL Atmospheric Chemistry Review

January 29 - 31, Boulder, Colorado

Motivation for top-down inventory assessment

- Most inventories compiled from bottom-up estimates, where reported data from many individual sources are summed

  an enormous undertaking
  regulatory / legal "baggage" } *difficult, and slow, to update*
  independent assessments are needed to evaluate accuracy

- Emissions routinely change over all time scales

  hours - power plant loads - 30%
  days - urban mobile fleet mix - 50%
  years - response to control strategies - up to 80%

- Accurate inventory input is critical for models *(**AQ**, C Cycle, climate).*

Evaluation methods and 4 selected applications:

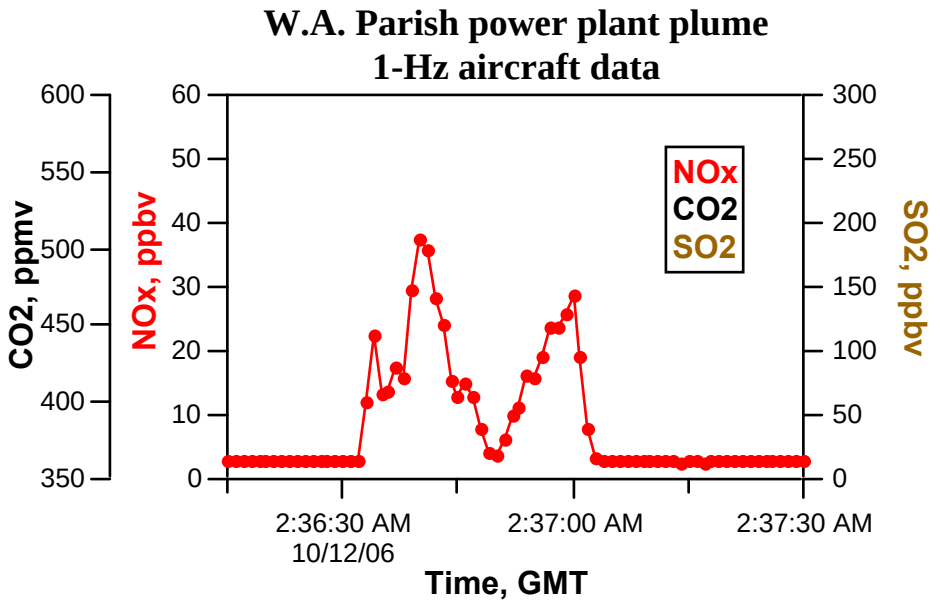
Field data, satellite retrievals, and models

Power plant NO_x, petrochemical VOCs, urban CO and VOCs

(but not biogenics, in this talk)

Top-down methods for inventory assessment

Emission ratios derived from field data “snapshots” agree with continuous emission monitoring systems (CEMS) at power plants



NOAA WP-3D aircraft transect, 650m AGL
6 km from W.A. Parish power plant
Sept. 12, 2006; Houston, TX

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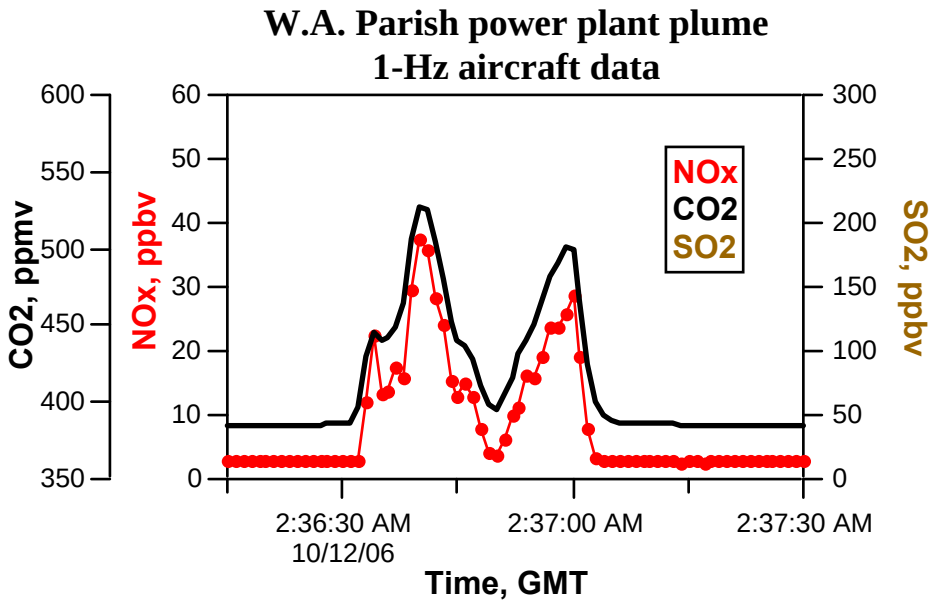

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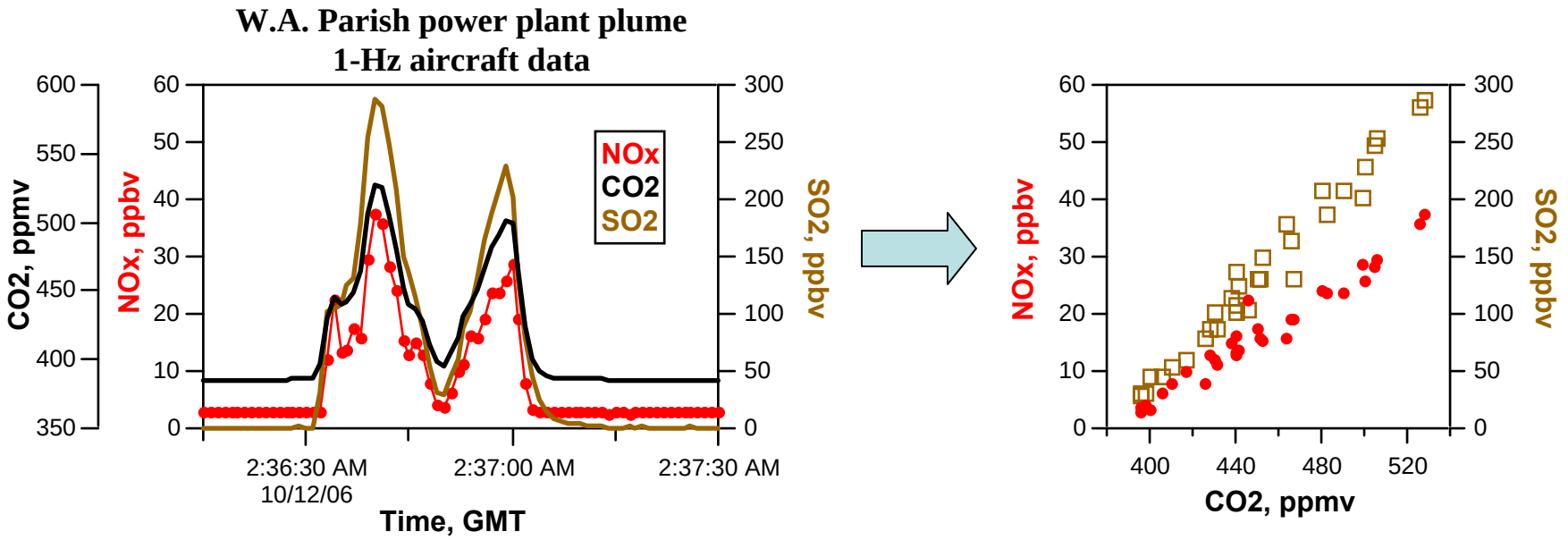

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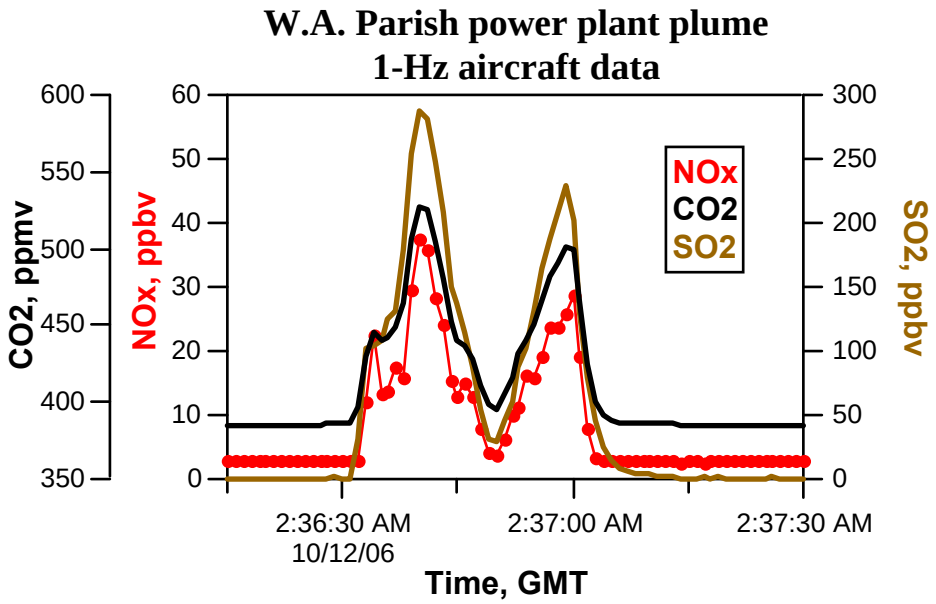
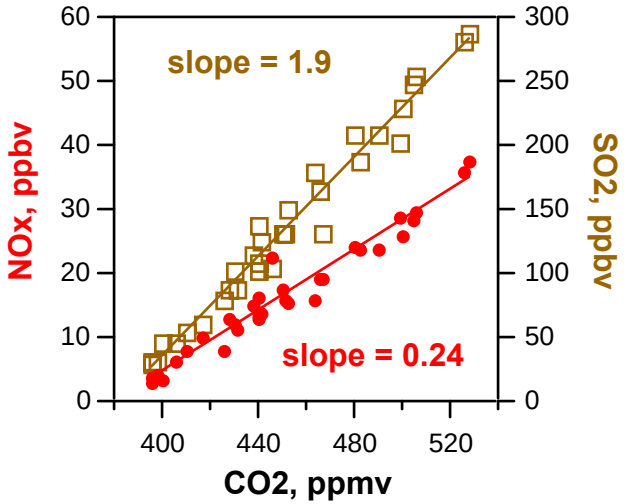




- **Method is accurate**: within ±25% of inventory values from CEMS
  - *apply top-down approach to wider variety of sources with reasonable confidence*

- **Top-down data necessary to quantify atmospheric concentration changes resulting from emissions control strategies**

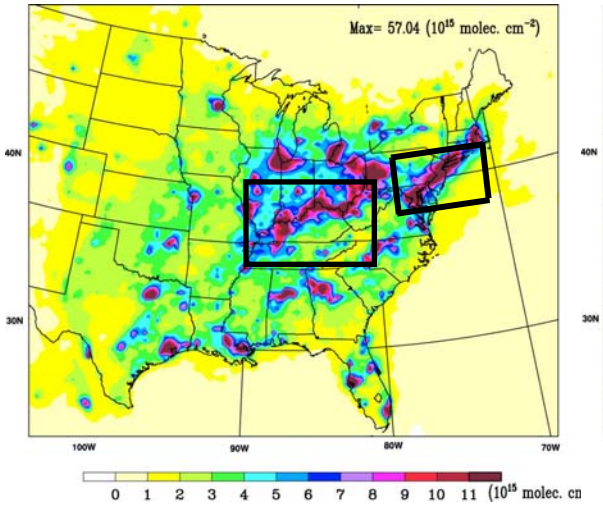
W.A. Parish emission estimates compared

	NO_x/CO_2	SO_2/CO_2
observed slope	**0.24** ±0.03	**1.9** ±0.3
CEMS inventory	**0.20**	**1.9**

Top-down methods for inventory assessment

Documenting atmospheric response to reductions on regional scales

modeled NO_2 columns
Summer 2004
WRF-Chem with EPA NEI 99



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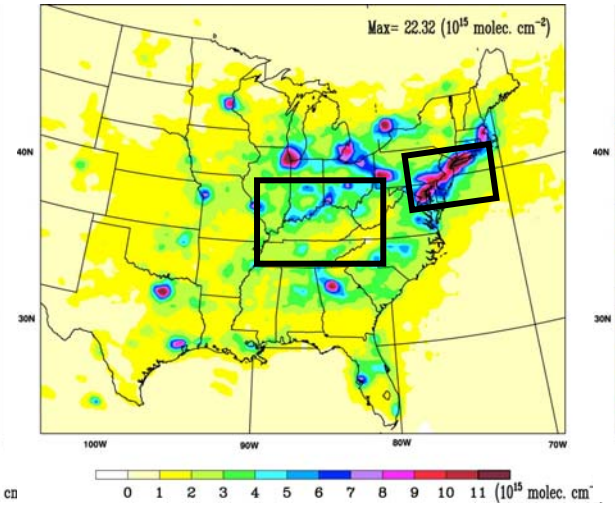

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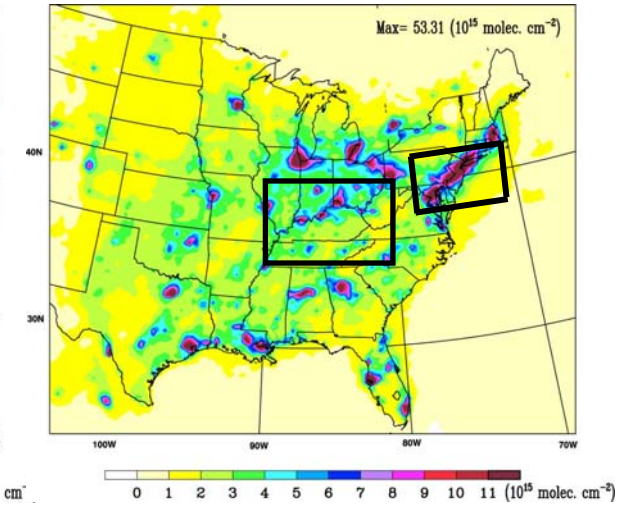

- EPA National Emissions Inventory **1999** v3 was released in **2004**
  - *NEI 1999 was the most up-to-date gridded national inventory available in late 2005*

- **major changes** in electric utility point source NO_x emissions took place in 2003
  - *documented by observations and CEMS; update to gridded inventory <u>still</u> pending*

Kim et al., (2006) GRL 33, doi:10.1029/2006GL027749

Top-down methods for inventory assessment

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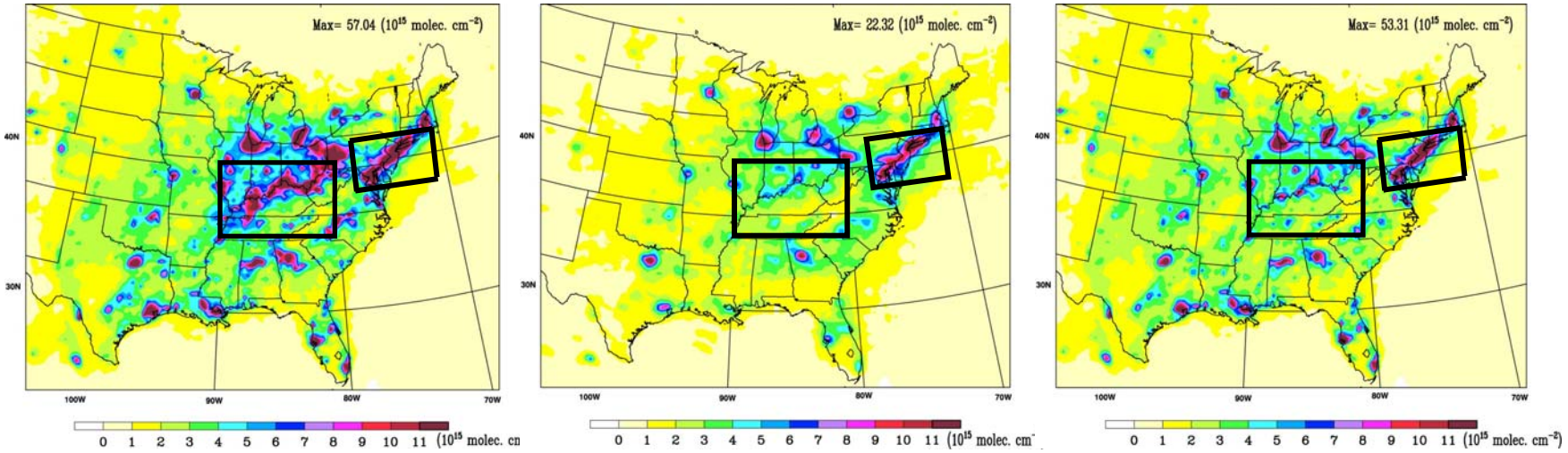

- Model cannot simulate post-2003 NO_2 in the Ohio River Valley without 'unofficial' (i.e., top-down) and time-consuming adjustments to the gridded national inventory
  - *this effect is also seen using inverse methods in global models*

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Top-down methods for inventory assessment

Documenting atmospheric response to reductions on regional scales

modeled NO_2 columns
Summer 2004
WRF-Chem with EPA NEI 99

observed NO_2 columns
Summer 2004
SCIAMACHY on Envisat

modeled O_3 difference
Summer 2004
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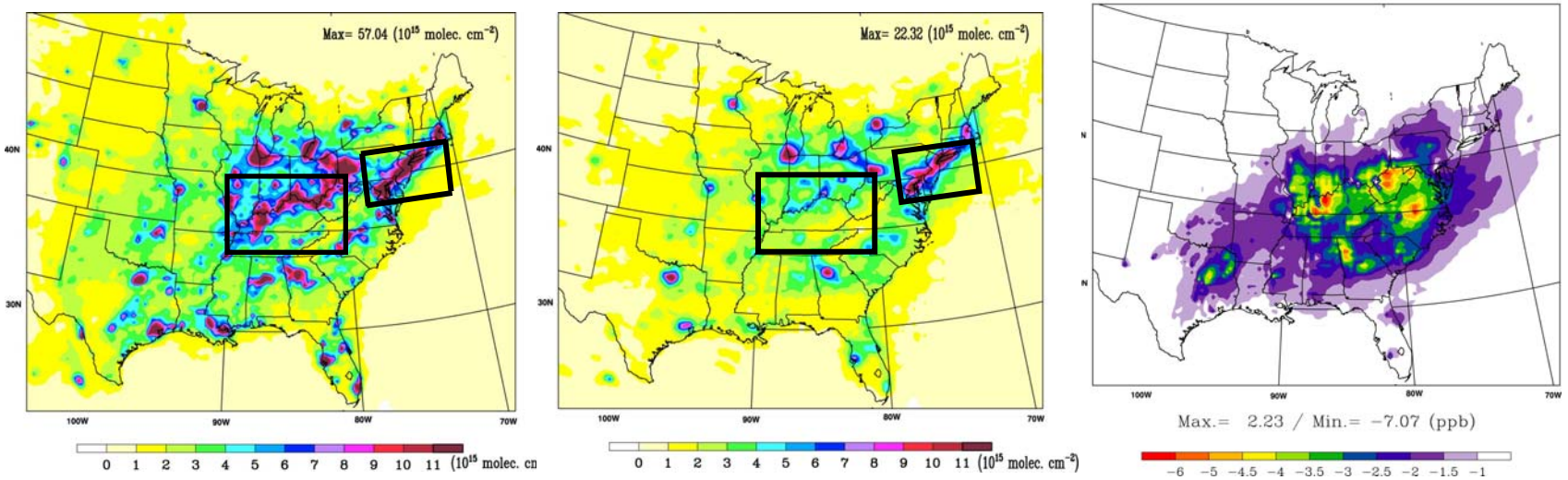

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- **Top-down methods are vital for timely evaluation of emissions control strategies**

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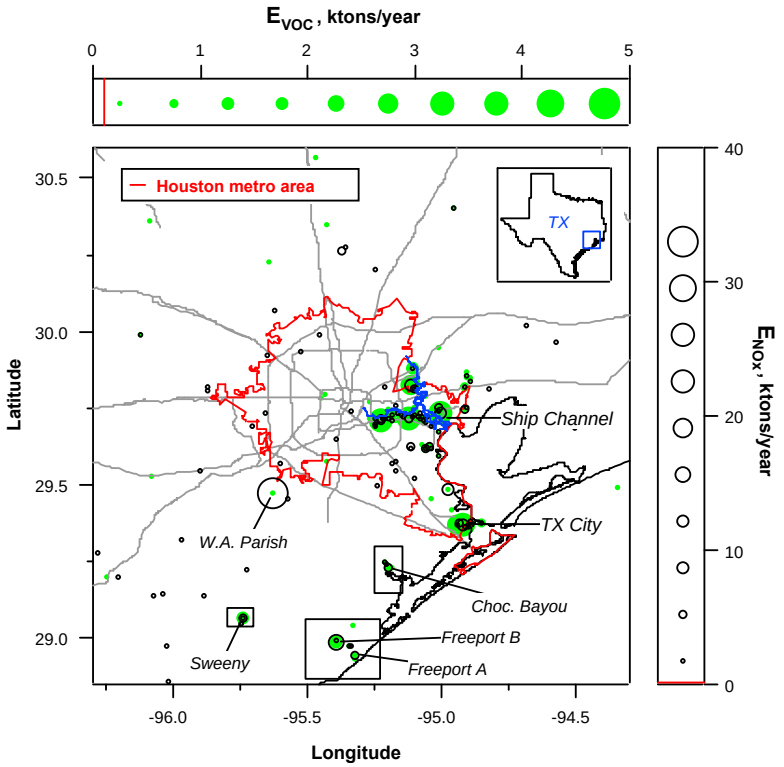
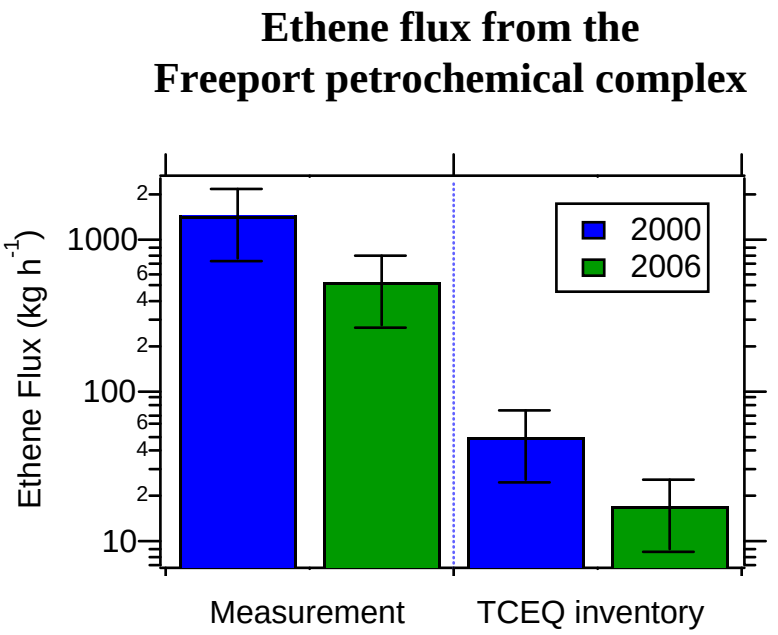
Top-down methods for inventory assessment

Errors in **petrochemical VOC** inventories persist in Houston





- Top-down methods point to **large (factor of 30) underestimates** of alkene emissions from petrochemical industries in Houston, TX in 2000 and 2006

de Gouw et al., (2008), in preparation.

Top-down methods for inventory assessment

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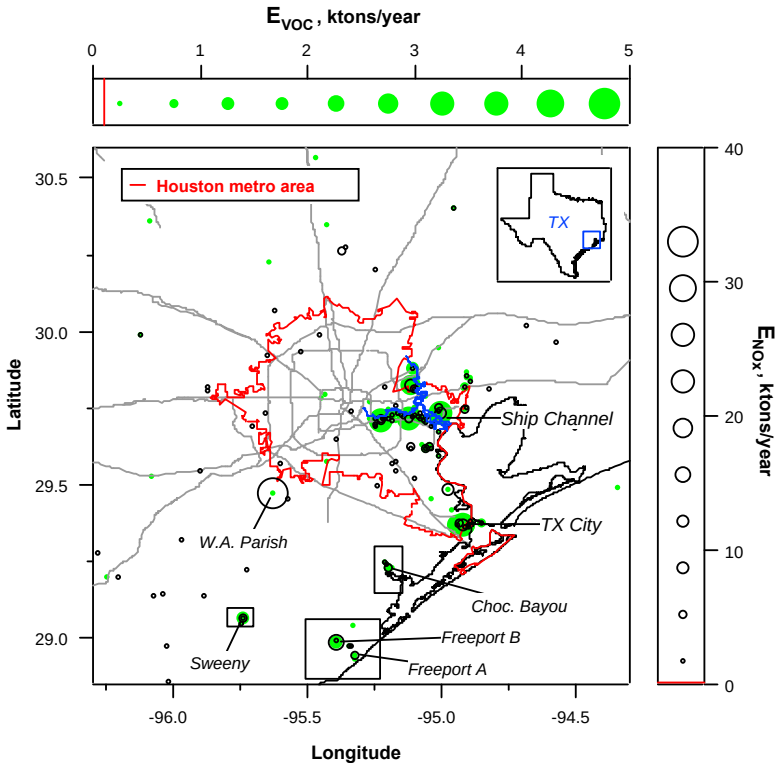

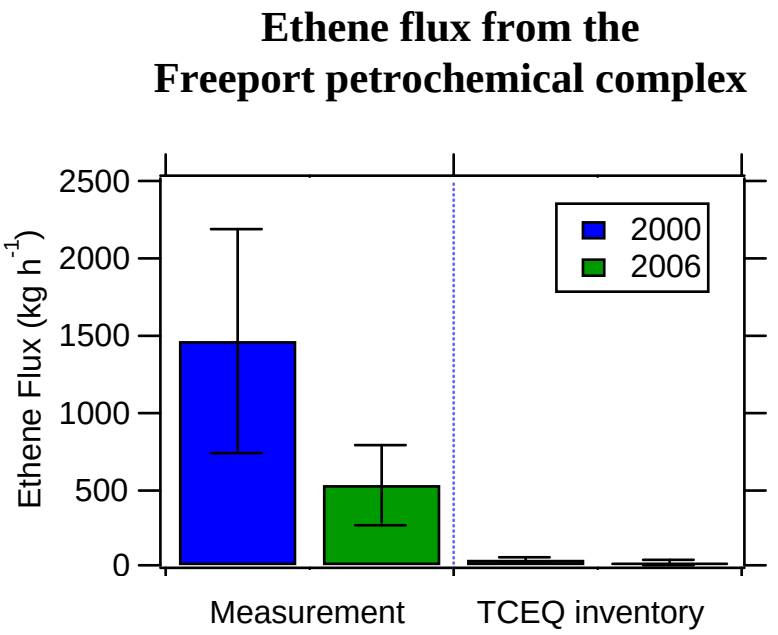

- Top-down methods point to **large (factor of 30) underestimates** of alkene emissions from petrochemical industries in Houston, TX in 2000 and 2006
- These alkenes dominate the most extreme (>150 ppbv) ozone exceedances

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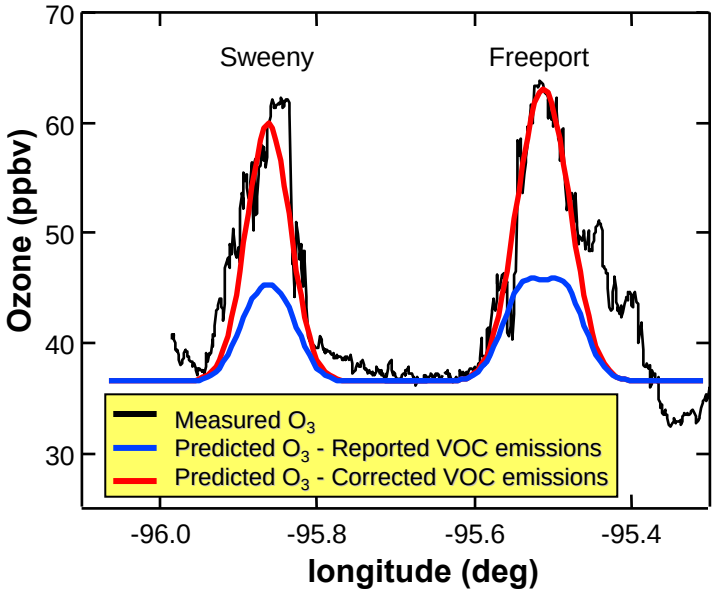
Top-down methods for inventory assessment

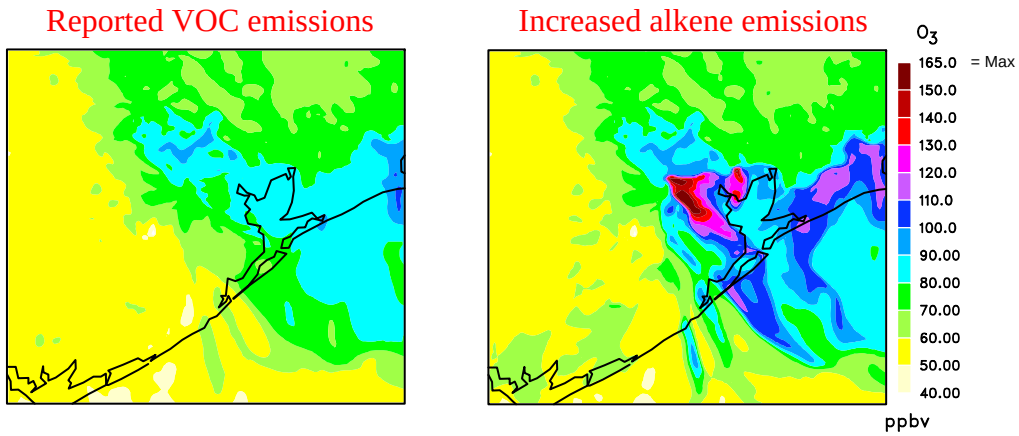
Errors in **petrochemical VOC** inventories persist in Houston

Gaussian plume model



Gaussian model and analysis:
M. Trainer - NOAA CSD

WRF-Chem 3D model

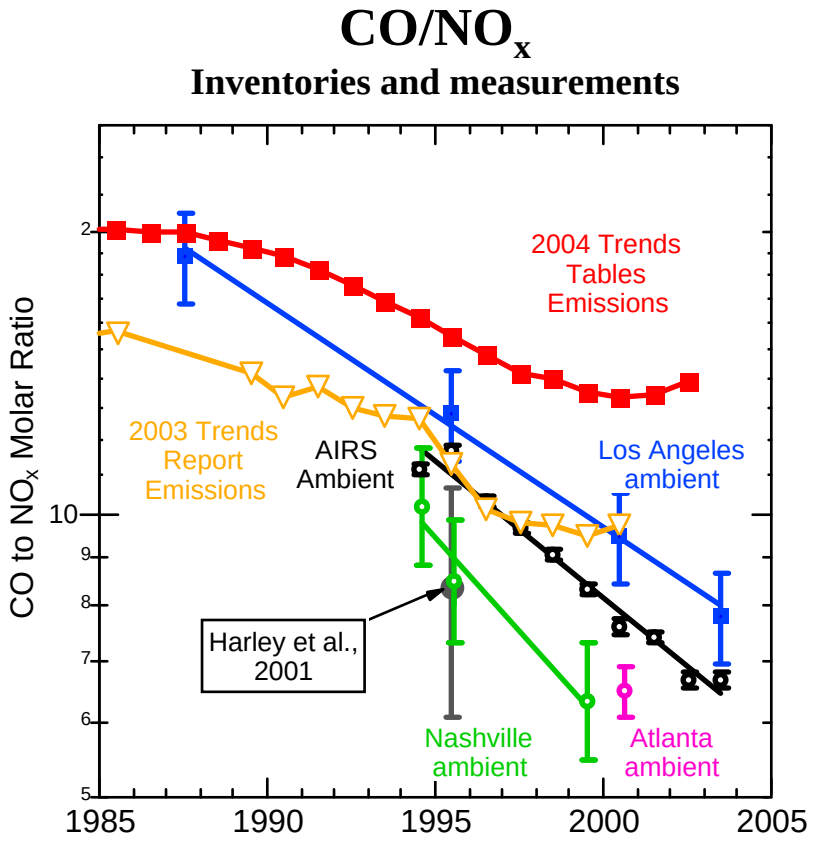


3D model and analysis:
Grell, Bao, McKeen - NOAA GSD and CSD

- Models realistically simulate observations of ozone and other photoproducts **<u>only after</u> reactive VOC emissions are made consistent with top-down assessments**
- State of Texas changed its emissions control strategy as a result of these findings

Top-down methods for inventory assessment

Urban CO emissions overestimated in inventory



- Mobile source bottom-up inventories have varied considerably over the past 15 years; no clear improvement over time
- Analyses of ambient data show the inventories do capture the dramatic decrease in mobile CO/NO_x ratios
- Top-down analyses using measured CO/NO_x and CO/CO_2 data suggest that **urban CO emissions are overestimated** (factors of 2-3) in bottom-up inventories

inventory errors are suggested by these observations … what are the effects?

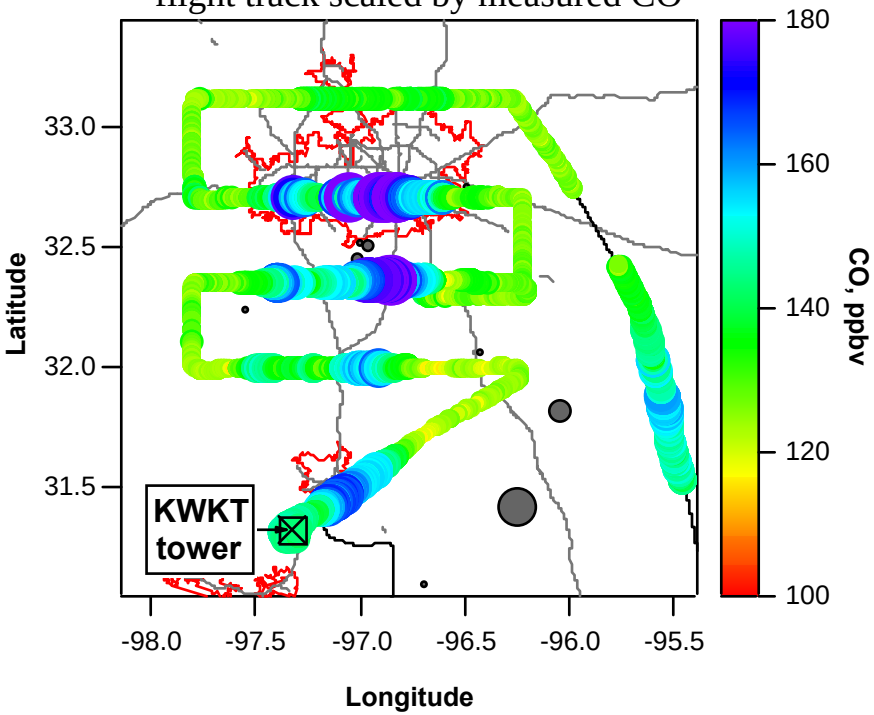
Parrish, D.D., (2006), Atmos. Environ. 40, 2288-2300.

Top-down methods for inventory assessment

Urban CO emissions overestimated in inventory

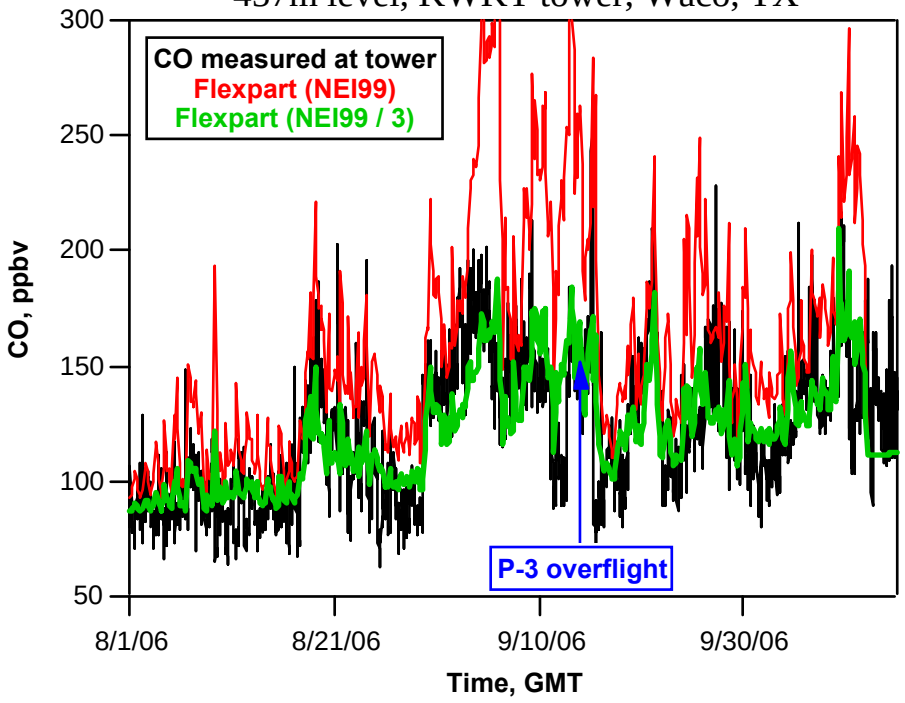
NOAA WP-3D aircraft data

flight track scaled by measured CO



NOAA Tall Tower Network data

457m level, KWKT tower, Waco, TX



- ties field project inventory "snapshots" to longer-term monitoring time scales
- top-down assessments are critical for both **carbon cycle…** and **air quality** issues
  - *determining biospheric CO_2 uptake using tall-tower data requires accurate knowledge of urban CO/CO_2 signatures*
  - *urban VOC emissions are coupled to urban CO, which together determine OH reactivity --> <u>ozone & PM implications</u>*

Additional data, analysis: A. Andrews, NOAA GMD

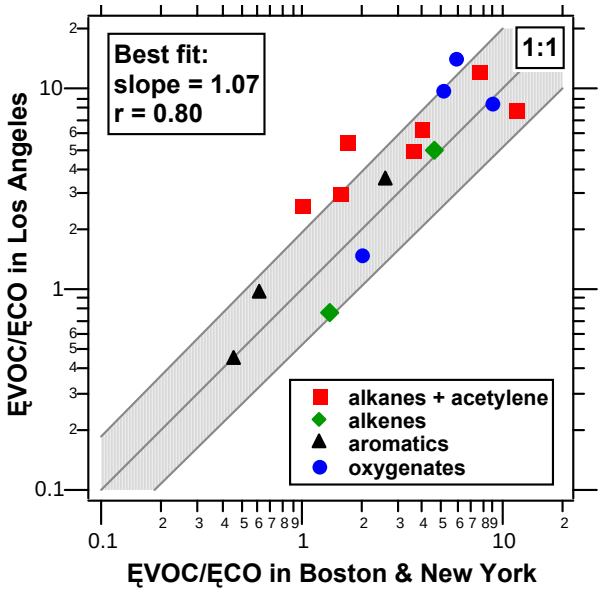
Flexpart model: A. Stohl, NILU

Top-down methods for inventory assessment

Urban VOC emissions not well represented in inventory

urban VOC/CO

Observations in different cities

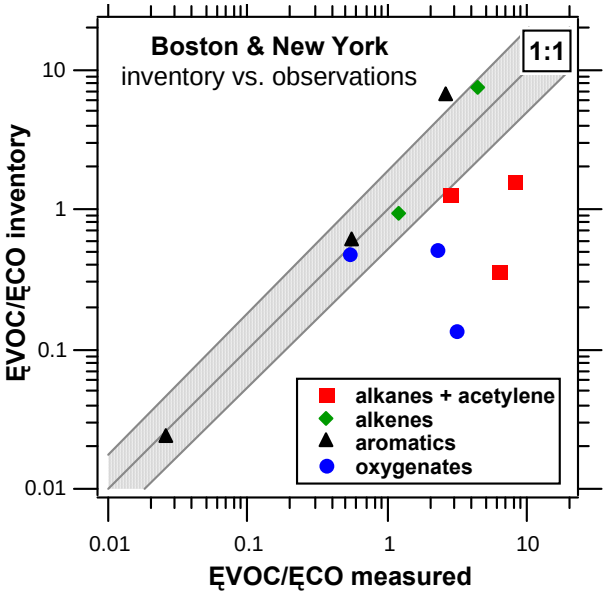


Compare NEI 99

to observations in Boston/NYC

urban VOC/CO

NEI 99v3 compared to observations



- Similar VOC/CO ratios observed
  (LA, NYC, Boston, Dallas, Houston…)
- Inventory captures VOC reactivity <u>per CO emitted</u>
  - *outliers represent relatively unreactive classes of compounds*

- since (reactive VOC/CO) ratios are reasonably represented in bottom-up inventory, **urban CO inventory errors imply large urban VOC reactivity errors**
  - *trend in VOC/NO_x is large and poorly represented in official inventory*

Warneke et al., (2007), JGR 112, doi:10.1029/2006JD007930

Top-down methods for inventory assessment

Conclusions

- **Tabulated inventories can be substantially in error**, by factors of 2 to 10 or more, for many important chemical species from many of the largest source types.
  - *continuing need for top-down, independent assessment of inventories*

- **Errors are sufficiently large to confound development of scientifically sound control strategies** based on anthropogenic emissions reductions.
  - *relevant to air quality, carbon cycle, and climate issues*

- Bottom-up inventories appear to be accurate to ±25% or better **only when directly measured monitoring data are routinely available** (e.g., point source CEMS)

Future directions for top-down assessments and AQ research

- **Continue to translate findings from observations to models**
  AQ payoff: quantify processes for improved understanding and prediction

- **Development of high-resolution mobile source CO_2 inventory**
  AQ payoff: criteria pollutants on a per-carbon basis

- **Add measurements for non-CO_2 GHG tropospheric source surveys**
  AQ payoff: CH_4 couples AQ and climate; source terms highly uncertain

- **Expanded surveys of NH_3 point and area emissions sources**
  AQ payoff: quantify effects of anticipated emissions changes on aerosol composition