Nitrogen Oxide Emissions Constrained by Space-based Observations of NO_2 Columns

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Introduction

- NO_x Sources
  - Fuel combustion (mobile, power plants and etc.)
  - Biomass burning
  - Lightening
  - Microbial processes in soil amplified by fertilizers, rain and burning
- NO_x Roles
  - Ozone production
  - Effect on the global climate indirectly by perturbing greenhouse gases
  - Adverse health effects
  - A precursor for ammonium nitrate, an important PM
  - Acidification and eutrophication of soils and surface waters

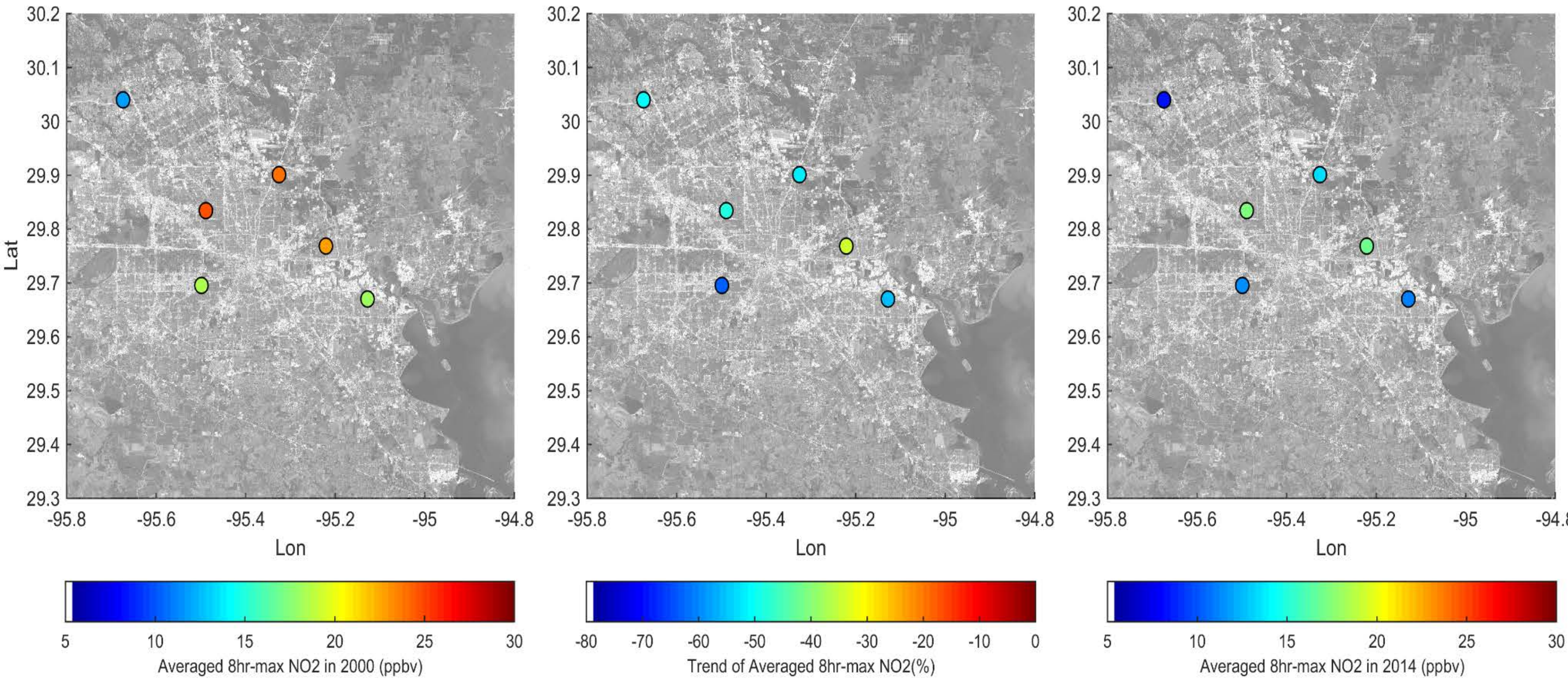
Introduction

- ‘Bottom-up’’ inventories
    - Fuel
    - Land use statistics
    - In-tunnel measurements of NO_x emission
    - Agricultural data
    - Estimates of burned areas

- Labor-intensive and expensive
- Done every 3 years in U.S.
- Have high uncertainty (e.g., ~50% for NEI-2005)
- Very soon become obsolete

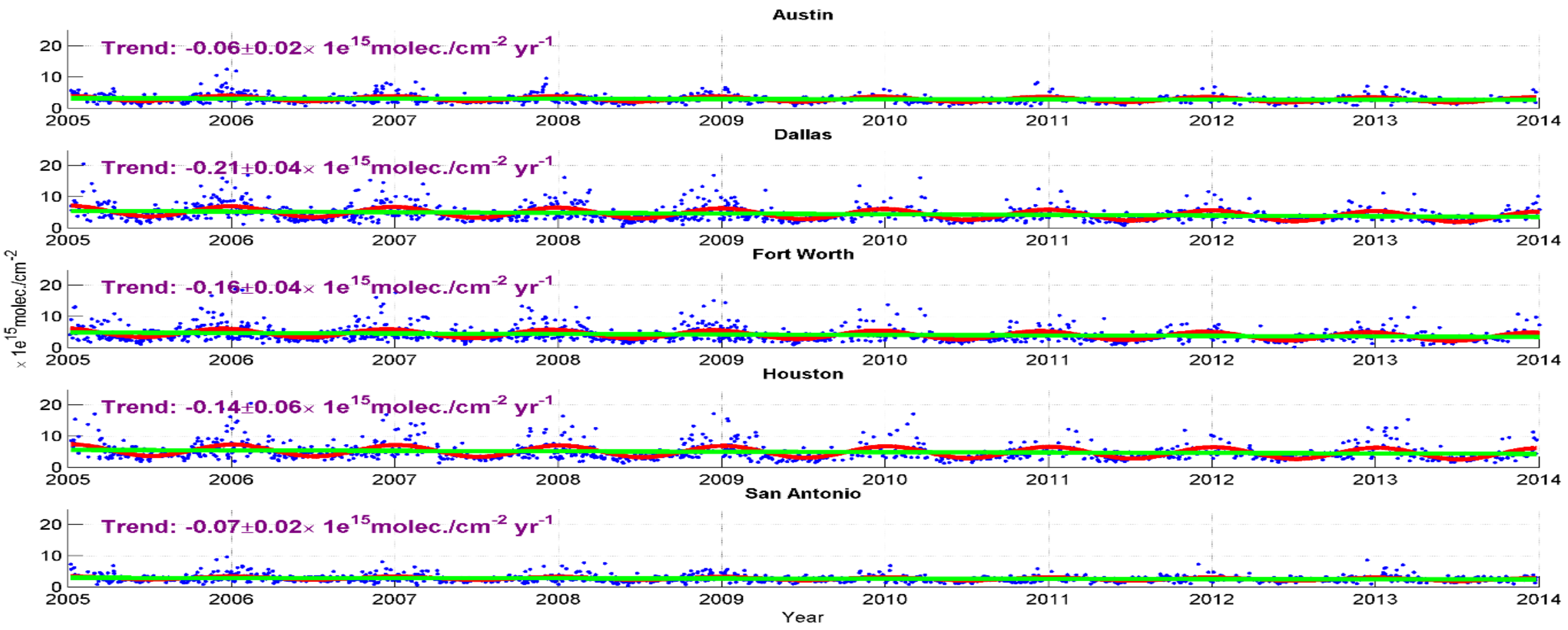
Long-term NO_2 trends

- A large and continuous decline in CAMS NO_2 levels during 2000-2014.



Long-term NO_2 trends

- A continuous decline in OMI NO_2 levels during 2005-2014.



Introduction

- "Top-down" approach
  - Satellite observations (y)
  - Emissions (x)
  - A Jacobian matrix (K) from a forward model

$$\mathbf{y} = \mathbf{K}\mathbf{x}$$

- When a physical quantity is not directly accessible for measurement, it is common to proceed by observing other quantities that are connected with it by physical laws.
- The notion of an inverse problem corresponds to the idea of inverting these physical laws to gain indirect access to the quantity.

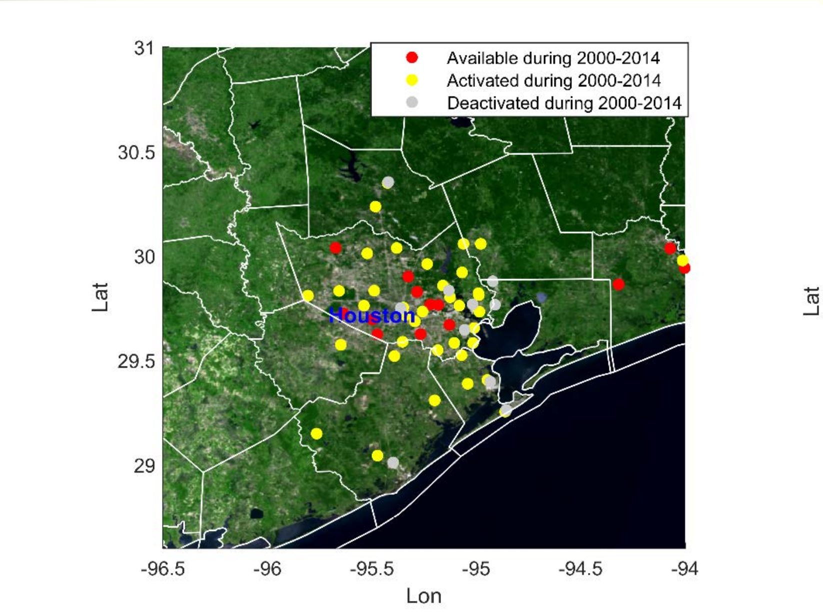
Research objective

- Quantify the posteriori NO_x emissions from a priori emissions (e.g., point, area, mobile, and soil sources) using an inverse method with tropospheric OMI NO_2 columns.

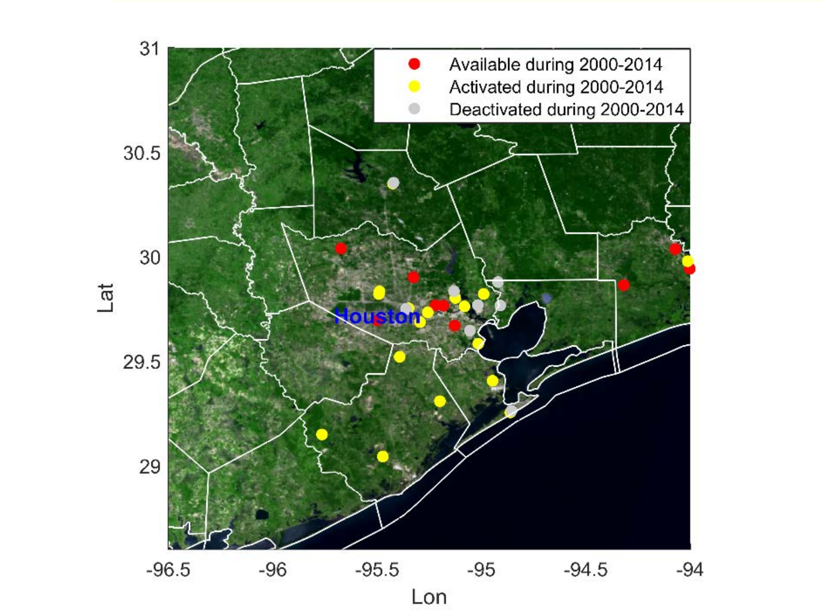
    - Use high spatial resolution of OMI NO_2
    - Improve WRF-CMAQ simulation using objective analysis
    - Utilize the Bayesian framework for inverse modeling
    - Evaluate the adjusted emissions with aircraft and ground-based observations.

Data: in-situ surface

- CAMS for surface ozone and NO_x data



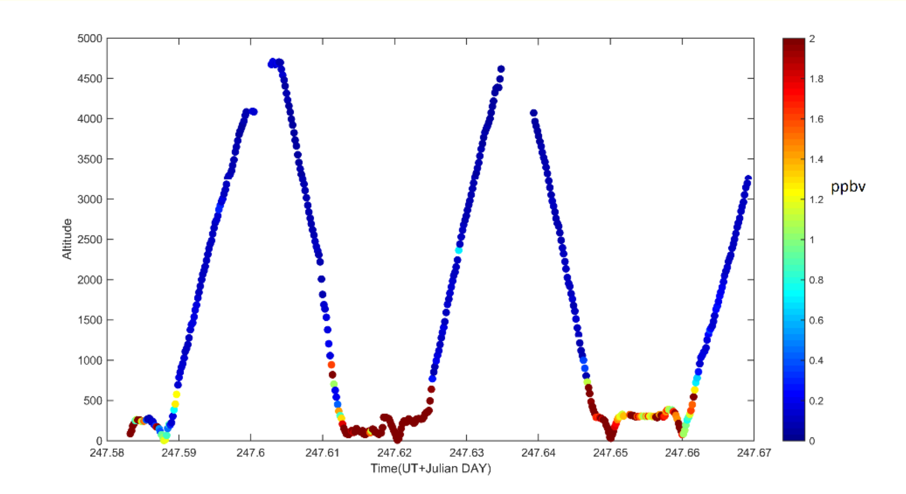
Surface ozone



NO_x

Data: Aircraft and emission

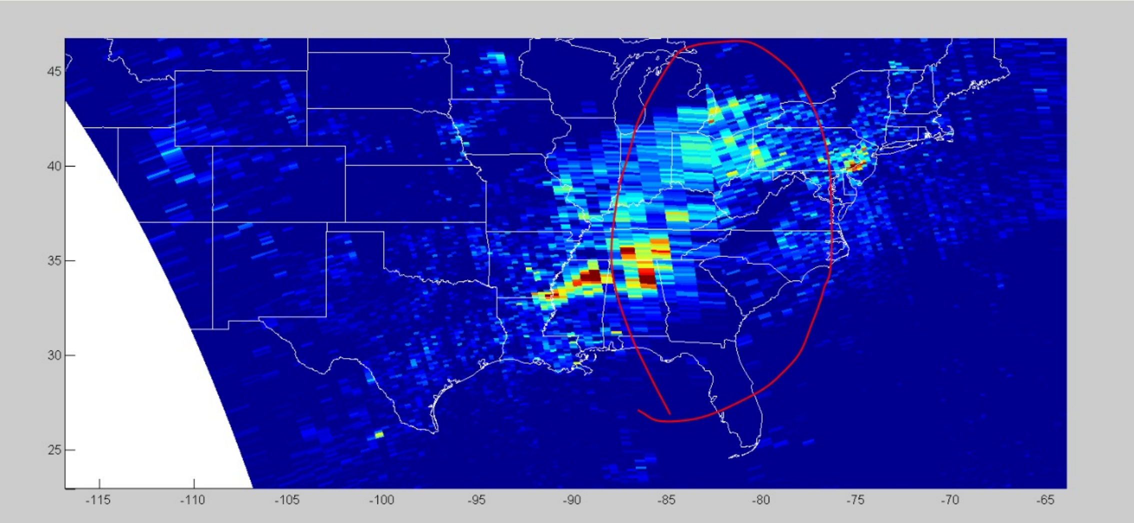
- Aircraft measurements (various gases including ozone and NO_x) (10 flights in September 2013)



- NO_x emission inventories from four different sources (area, mobile, biogenic and point) based on NEI-2011

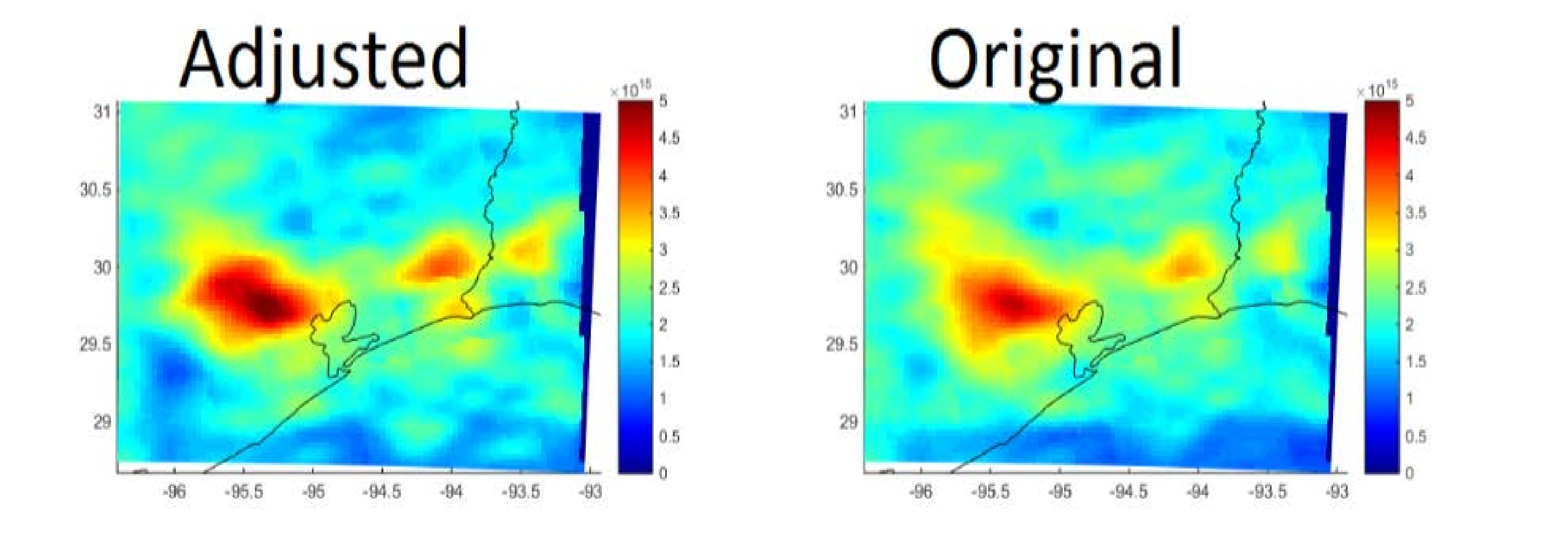
Data: remote sensing

- Satellite tropospheric OMI NO_2 column.
  - Noisy pixels filtered out based on cloud fraction, RMSE in the retrieval, VCD quality and etc.
  - OMI footprint is larger in pixels far from nadir. A remedy is to use splines to correct geometric distortions based on Kuhlmann et al. (2014)



Data: remote sensing

- The influences of priori NO_2 profiles removed by using Air Mass Factor in each granule and model simulation (e.g., Choi et al., 2008; Duncan et al., 2014).
- Without this adjustment, OMI shows underprediction and overprediction in urban and rural areas respectively.



Method: Inverse modeling

- Inverse modeling:

$$\mathbf{y} = \mathbf{K}\mathbf{x} + \epsilon_{\Sigma}$$

- For well-conditioned linear problems, under the assumption of independent and normally distributed data errors, least-squares (maximum likelihood principle) can be used.

- The Bayesian approach
  - Random variables
  - The approach can naturally use a priori

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Bayesian method

$$\mathbf{y} = \mathbf{K}\mathbf{x} + \boldsymbol{\varepsilon}_{\Sigma}$$

$$J(\mathbf{x}) = (\mathbf{y} - \mathbf{K}\mathbf{x})^{\mathrm{T}}\mathbf{S}_{\Sigma}^{-1}(\mathbf{y} - \mathbf{K}\mathbf{x}) + (\mathbf{x} - \mathbf{x}_a)^{\mathrm{T}}\mathbf{S}_a^{-1}(\mathbf{x} - \mathbf{x}_a)$$

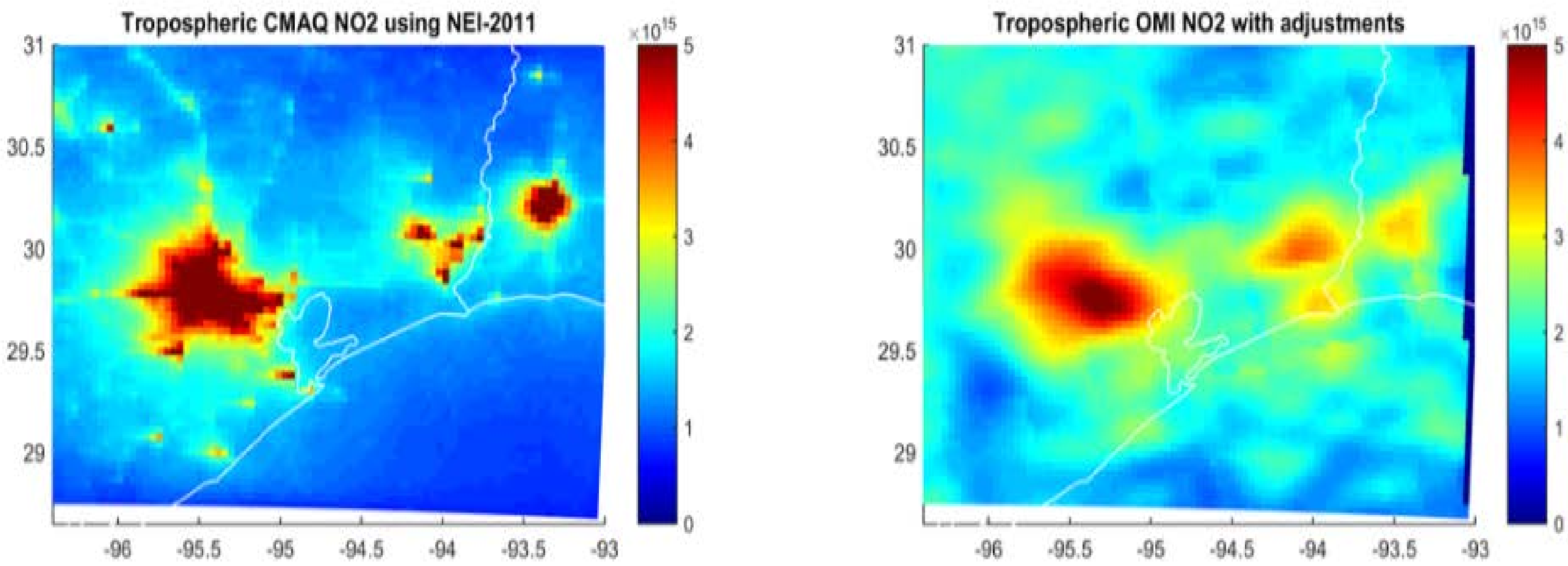
$$\hat{\mathbf{x}} = \mathbf{x}_a + \mathbf{S}_a\mathbf{K}^{\mathrm{T}}\left(\mathbf{K}\mathbf{S}_a\mathbf{K}^{\mathrm{T}} + \mathbf{S}_{\Sigma}\right)^{-1}(\mathbf{y} - \mathbf{K}\mathbf{x}_a)$$

$$\hat{\mathbf{S}}^{-1} = \mathbf{K}^{\mathrm{T}}\mathbf{S}_{\Sigma}^{-1}\mathbf{K} + \mathbf{S}_a^{-1}$$

- Model (CMAQ or CAMx) determines, $K = dNO_2/dE\_NO_x$
- A posteriori x is determined at last

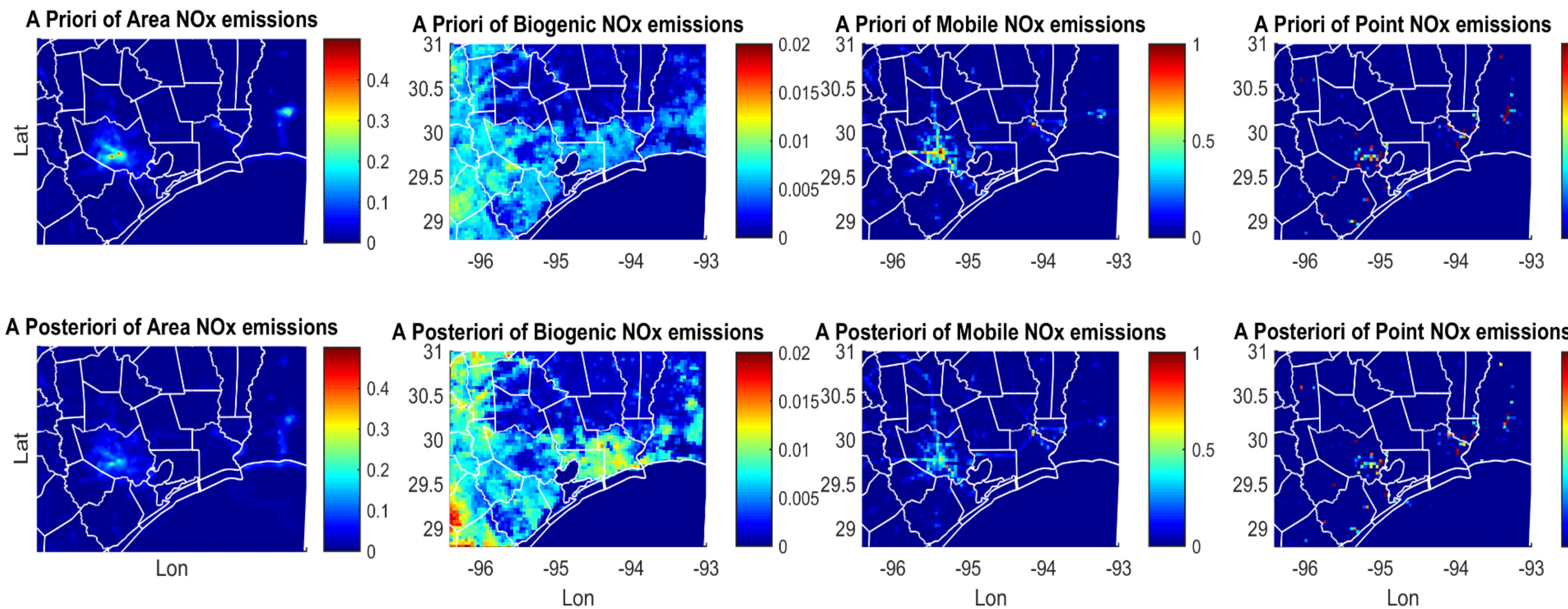
Results

CMAQ overpredicted NO_2 in urban regions and underpredicted in rural ones, which is similar to those by Choi et al. (2012) and Choi (2014)



Results

Overall, all the anthropogenic NO_x emissions reduced, while biogenic emissions increased. Both reduction and enhancement not occurred evenly over the domain.



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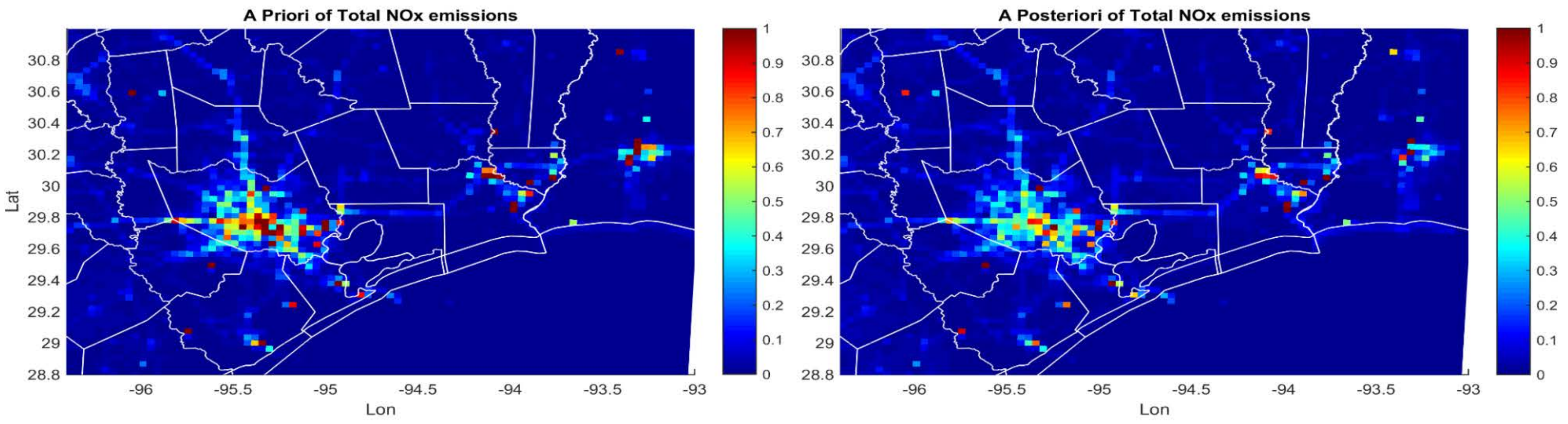
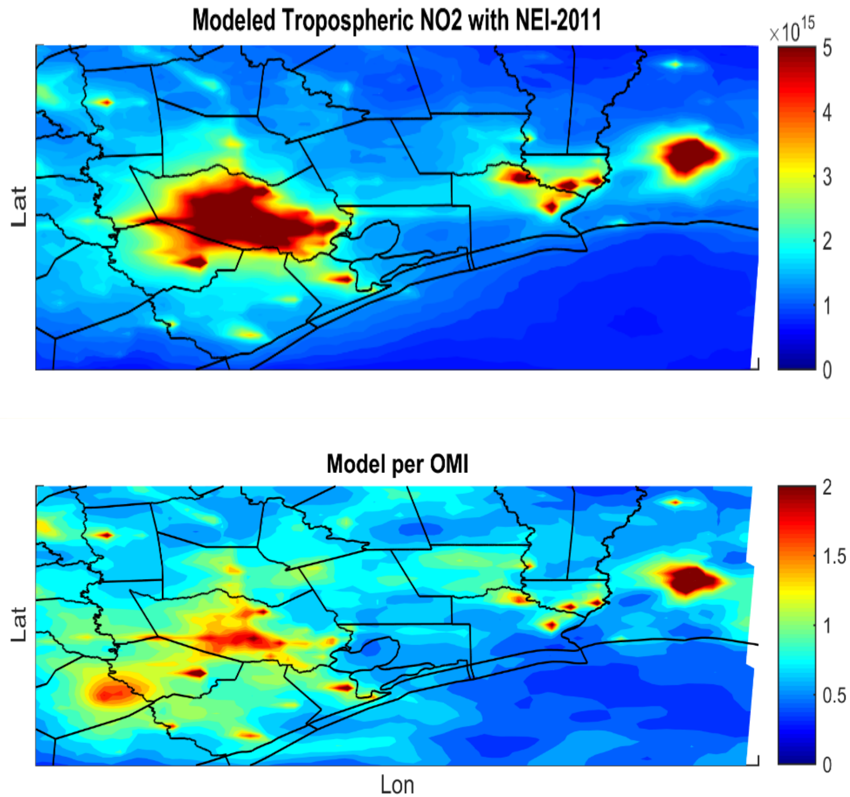
Results

- Total NO_x emission overview:



Results

Comparison to OMI NO_2 columns



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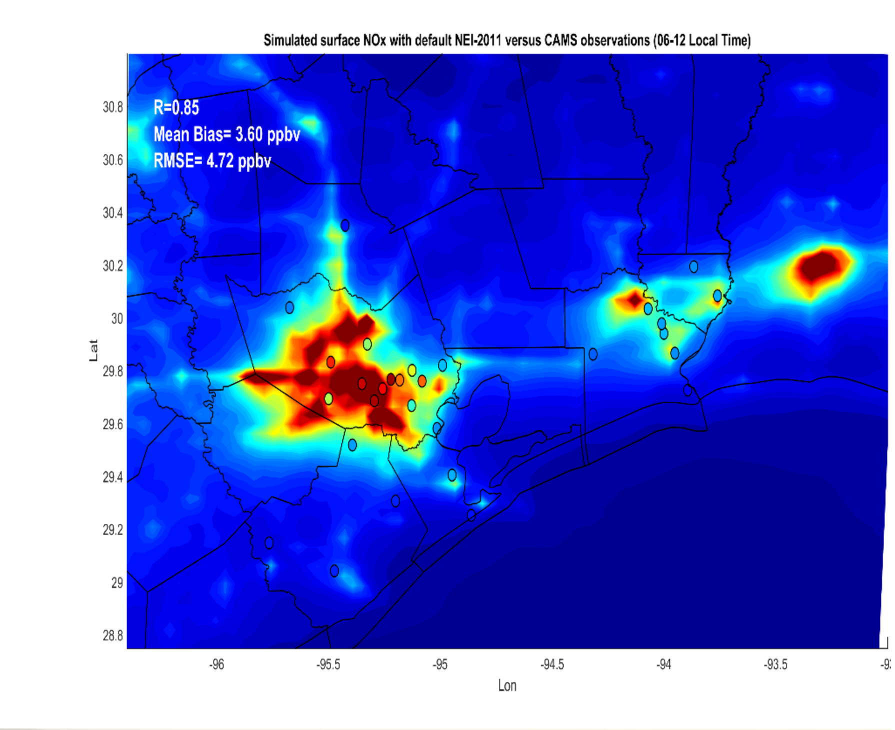
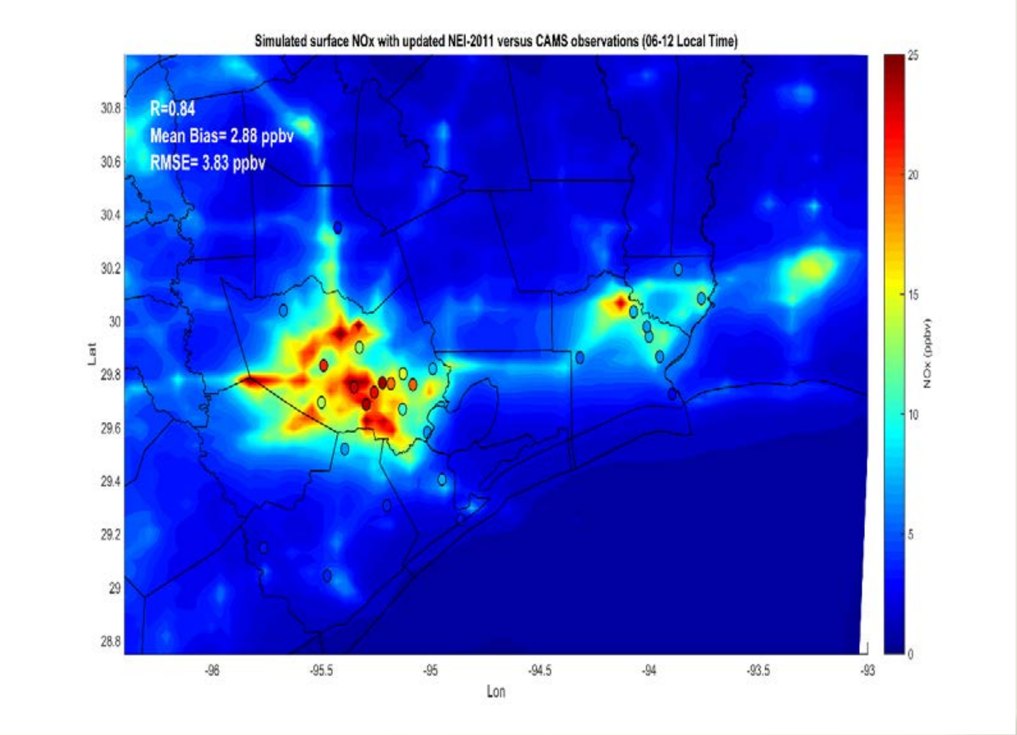
Results

Comparison to CAMS NO_x values in morning time (06-12 LT) of Sep 2013 (before and after inverse modeling)





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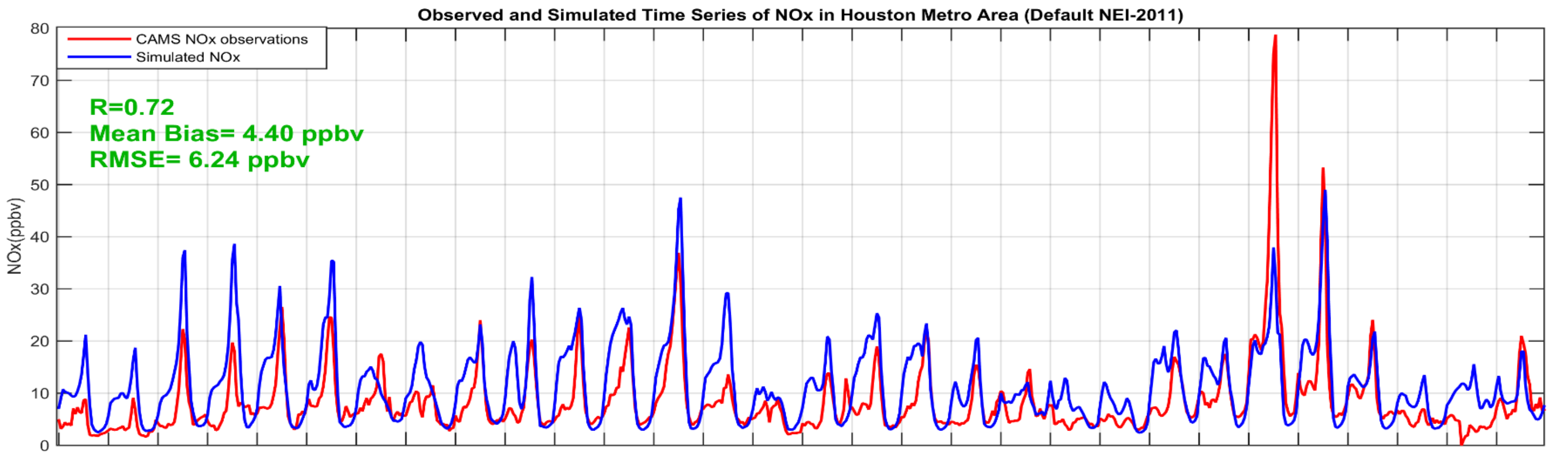
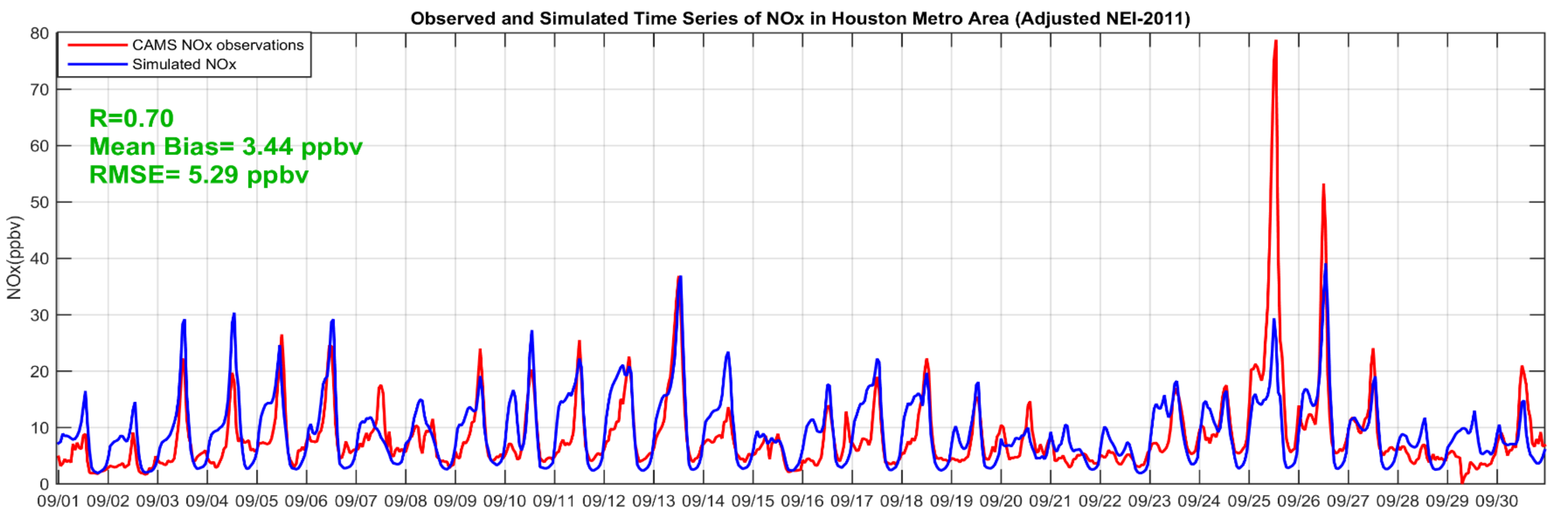
Results

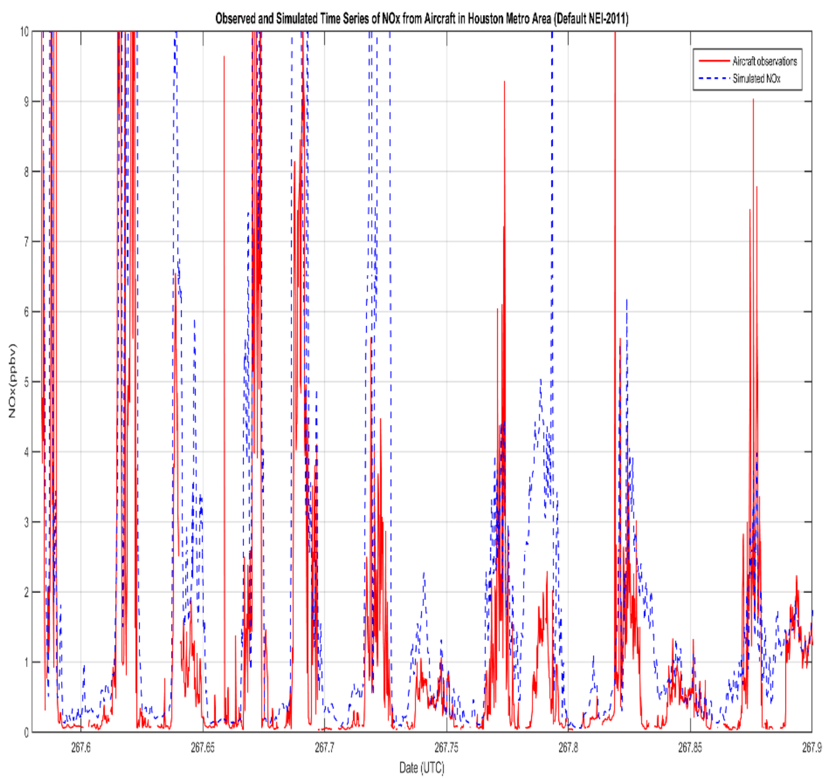
Time series of CAMS NO_x before (upper) and after (lower) adjusting emissions:

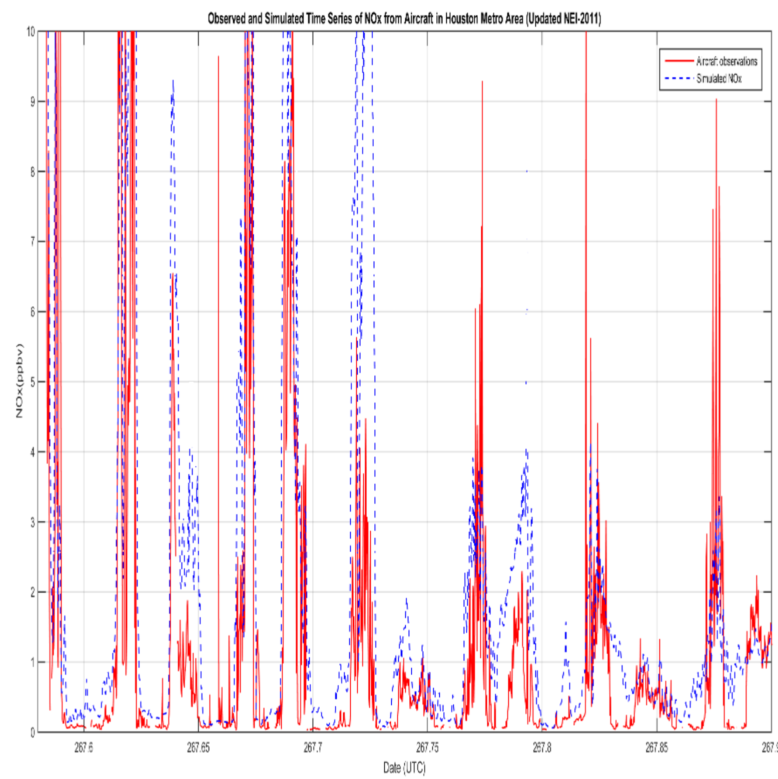




Results

- RMSE and bias between aircraft NO_x and simulated ones are 2.4 and 6.0ppbv for NEI2011 (left) and 1.9 and 4.1ppbv for adjusted NEI-2011 (right).
- A snapshot for Sep. 24th





Conclusion and following works

- CMAQ using NEI-2011 showed NO_2 overprediciton in urban areas and undeprediction in rural areas.

- Evidence to show that tropospheric OMI NO_2 can be used to constrain the emission.

- Anthropogenic emissions reduced after the update, but biogenic emission increased.

- The bias between observations and simulated NO_x decreased after the emission is updated.

- Following works:
    - Inverse modeling for biomass burning (e.g., FINN, GFED or QFED) and HCHO (a proxy for VOC)

Thanks for your attention