

# Observations of enhanced reactive Hg in the free troposphere and Hg emissions from urban areas

L. Gratz<sup>1</sup>, J. Ambrose<sup>1</sup>, D. Jaffe<sup>1,2</sup>, L. Jaeglé<sup>2</sup>, V. Shah<sup>2</sup>, N. Selin<sup>3</sup>, S. Song<sup>3</sup>

<sup>1</sup>University of Washington-Bothell, Bothell, WA

<sup>2</sup>University of Washington, Seattle, WA

<sup>3</sup>MIT, Cambridge, MA

Southeast Atmosphere Study Data Workshop

Boulder, CO

March 31, 2014



# Acknowledgments

- **Funding**



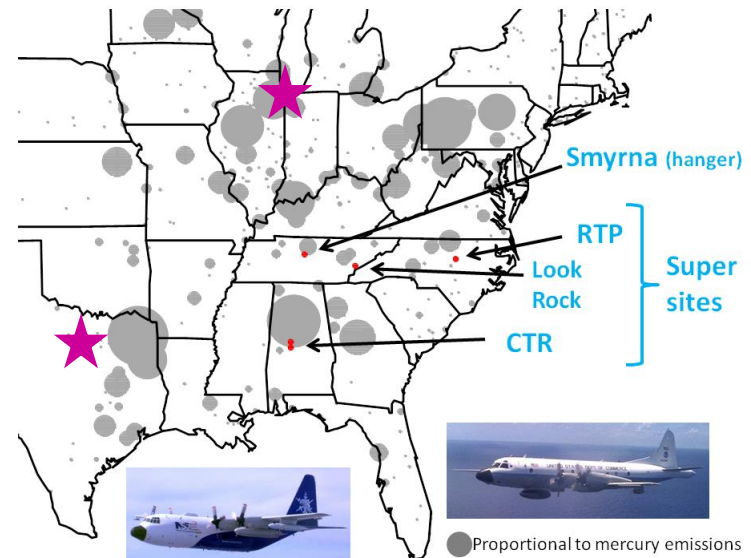
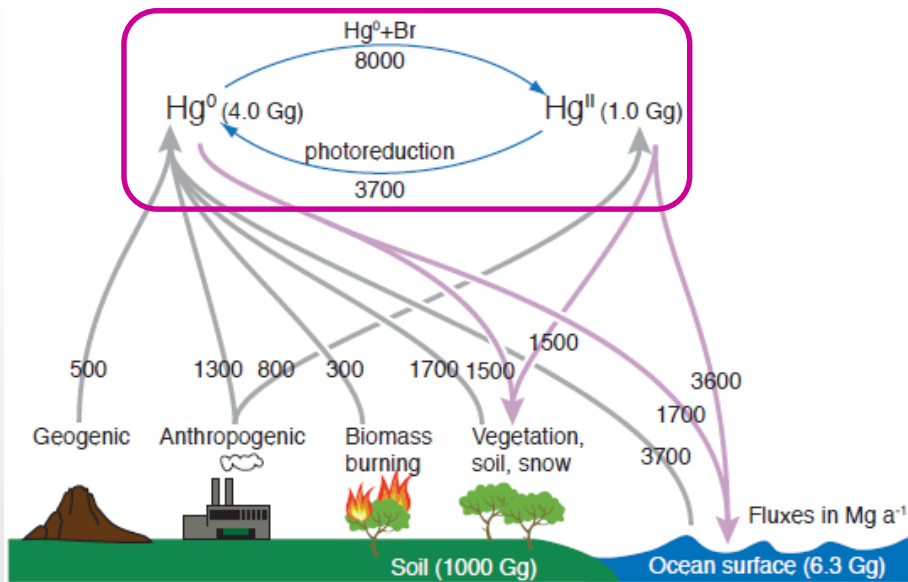
- **NOMADSS Collaborators**

- NOMADSS Science Team
- NCAR Research Aviation Facility

*NOTE: Some data used in this presentation are preliminary*

# NOMADSS Hg Objectives

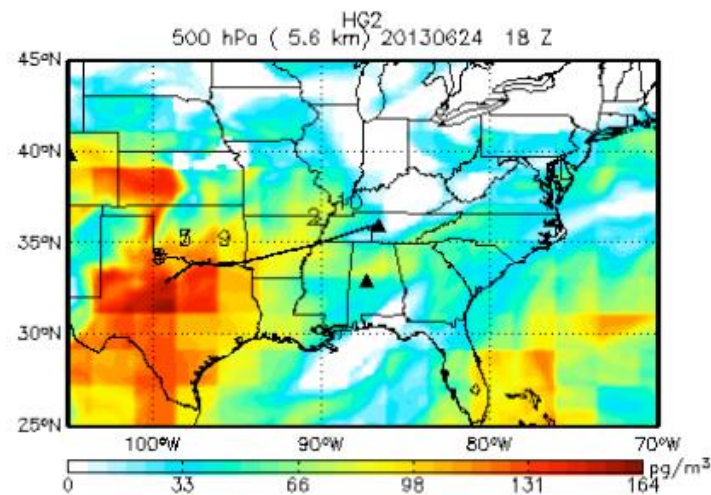
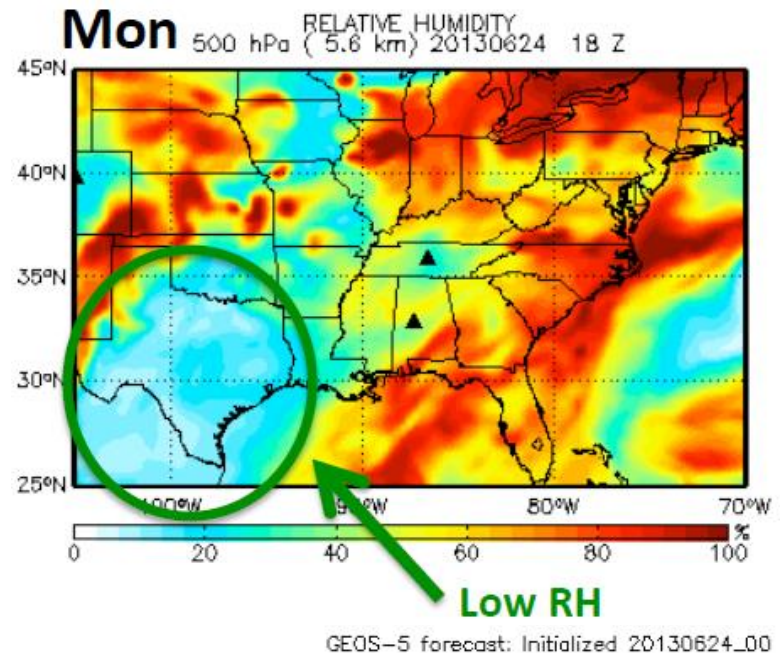
1. Quantify the distribution and chemical transformation of speciated Hg in the free troposphere (FT).
  - **What are the concentrations and sources of reactive Hg (RM)?**
2. Constrain emissions of Hg from major source regions in the Eastern U.S.
  - **What are the relative Hg emissions from major urban areas?**



Holmes *et al.* (2010)

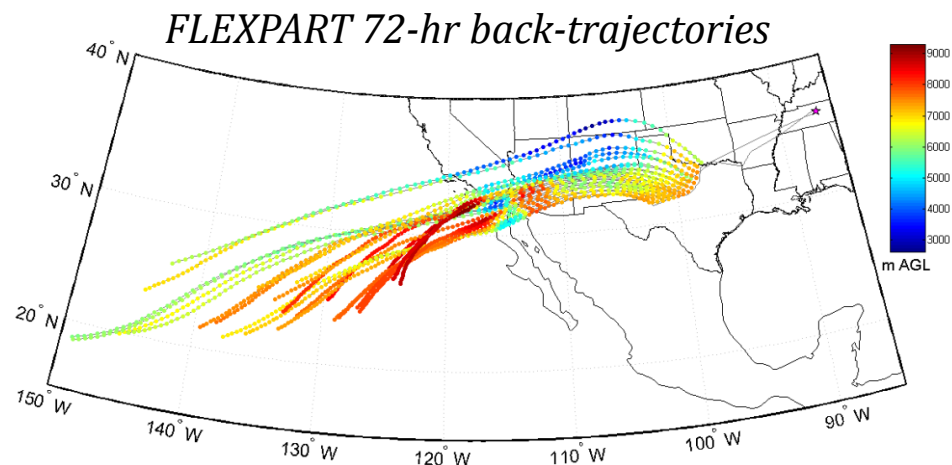
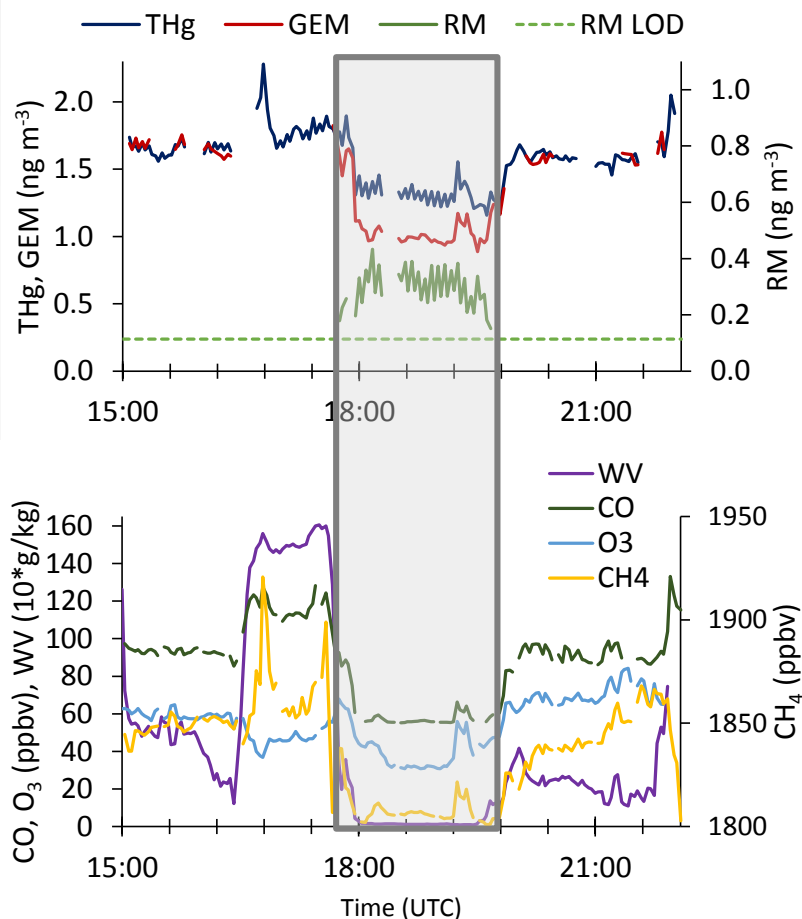
# 1. RM in the Free Troposphere

- Enhanced RM was often forecast in the dry free troposphere over Texas.
- We sampled this free tropospheric RM pool on two NOMADSS flights:
  - ✓ RF-06 on 6/19/2013
  - ✓ RF-09 on 6/24/2013



# 1. RM in the Free Troposphere

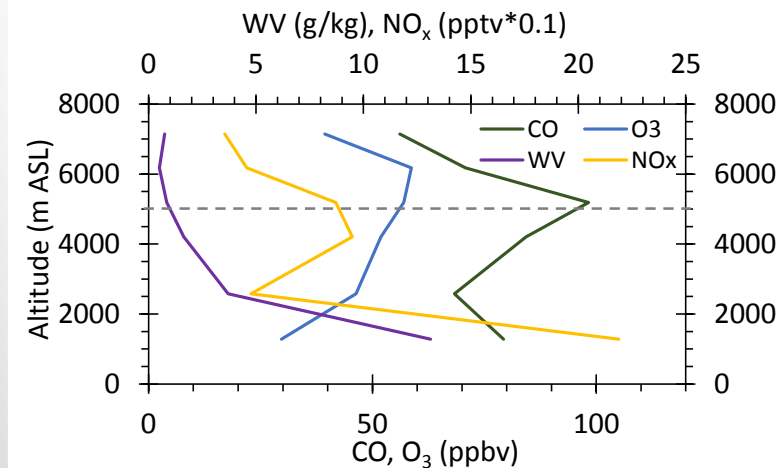
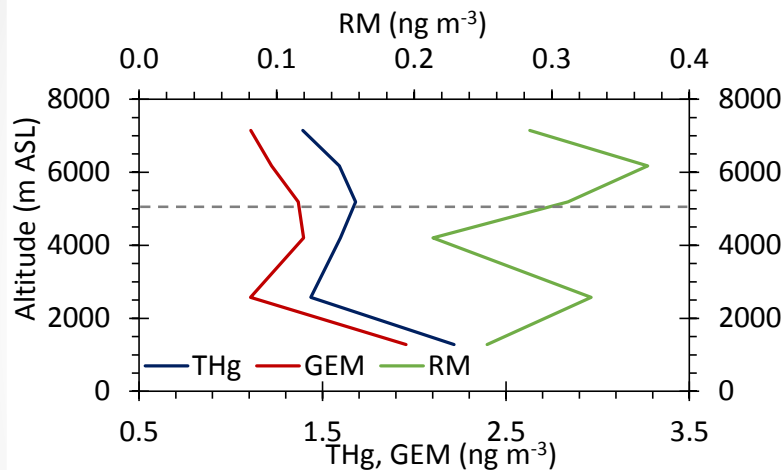
## RF-06: Enhanced RM over TX



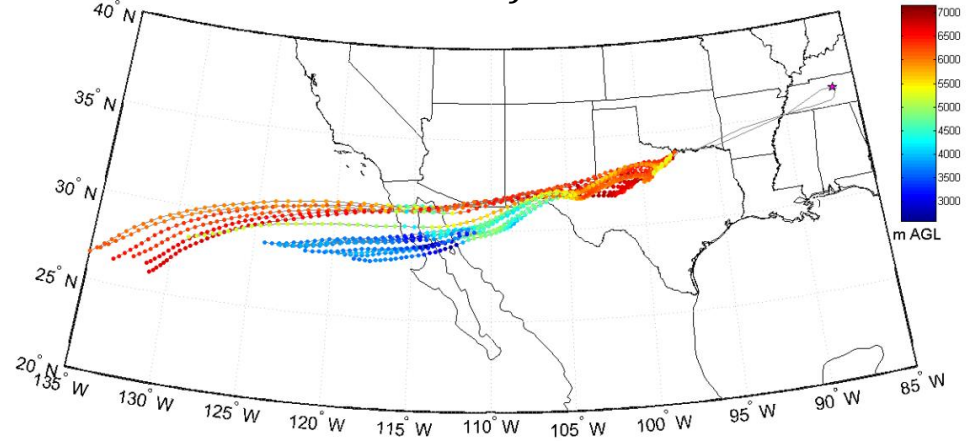
- Observations and trajectories suggest RM source is the FT pool over eastern sub-tropical Pacific.
- Air mass may have circulated around Pacific High for many days.
- No indication for influences of anthropogenic or marine emissions, or stratospheric subsidence.

# 1. RM in the Free Troposphere

## RF-09: Vertical Profile over TX



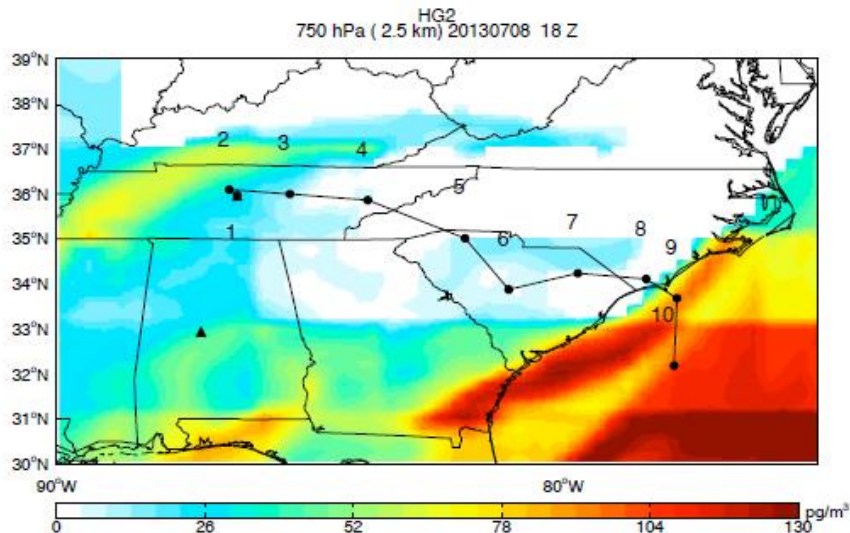
*FLEXPART 72-hr back-trajectories > 5000 m AGL*



- Like RF-06, RM source is the FT pool over eastern sub-tropical Pacific.
- Relatively clean and dry air mass.
- Observed RM concentrations in the FT were 3-5x larger than forecast or near-real time model analysis.

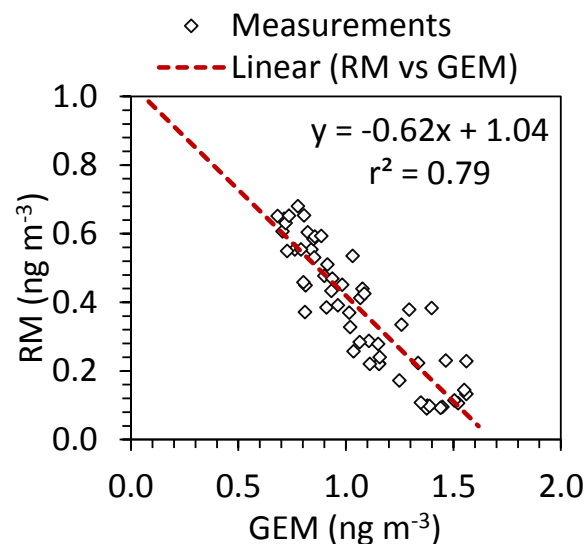
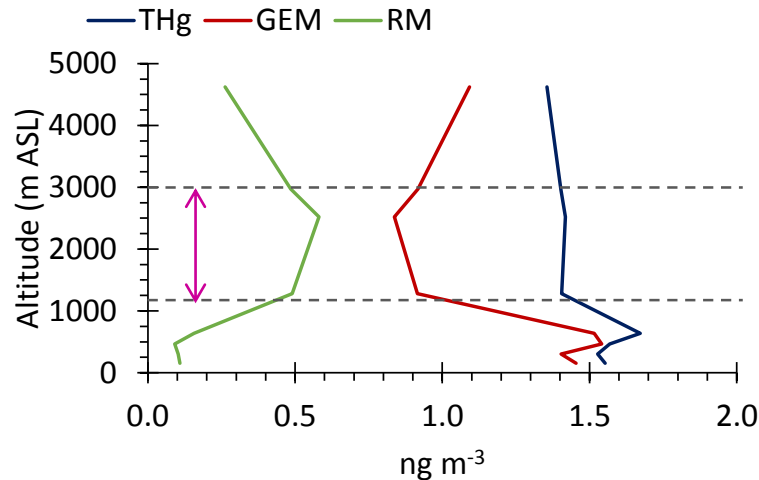
# 1. RM in the Free Troposphere

## RF-16: Vertical Profile over Atlantic



*C-130 flight track & GEOS-5 Forecast for RM at 750 hPa on 7/8/2013.*

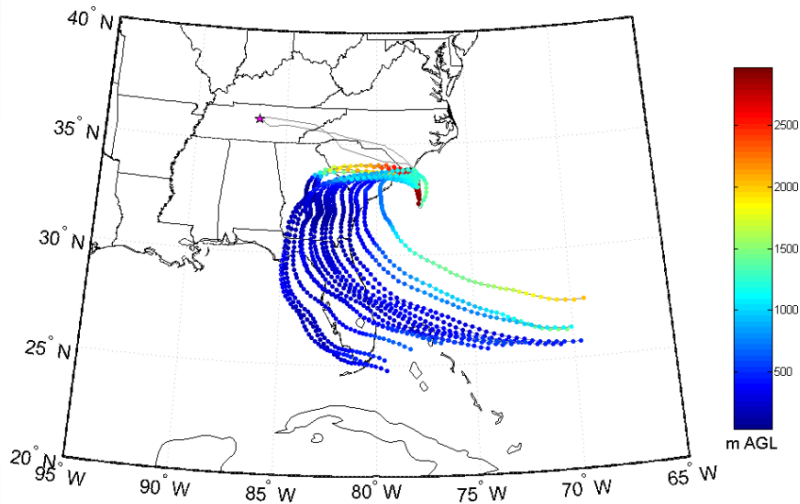
- Enhanced RM was observed in a layer between 1250 – 3000 m ASL.
- RM vs. GEM across the profile shows strong evidence for in situ oxidation (different from RF-09).



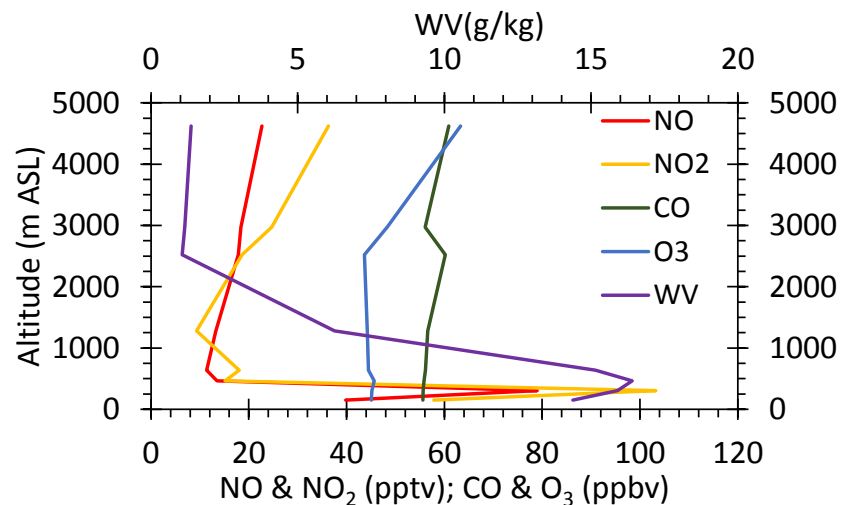
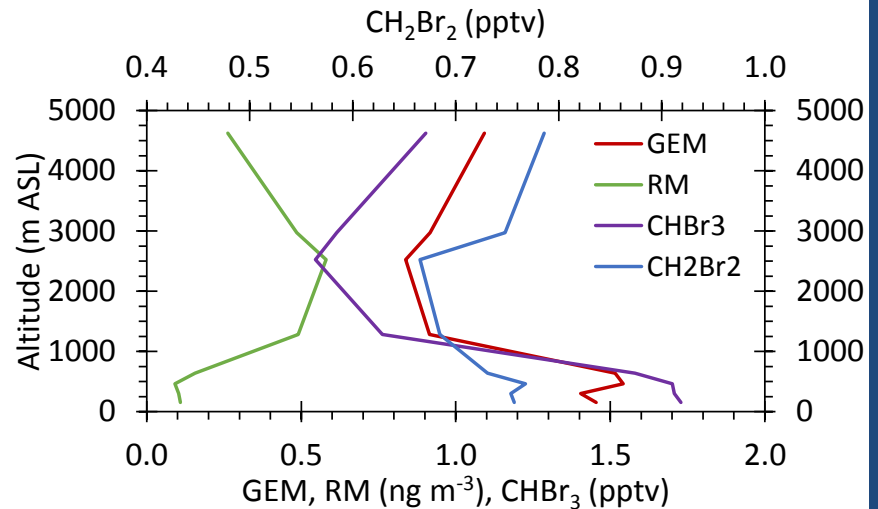
# 1. RM in the Free Troposphere

## RF-16: Vertical Profile over Atlantic

*FLEXPART 72-hr trajectories, 1250-3000 m AGL*



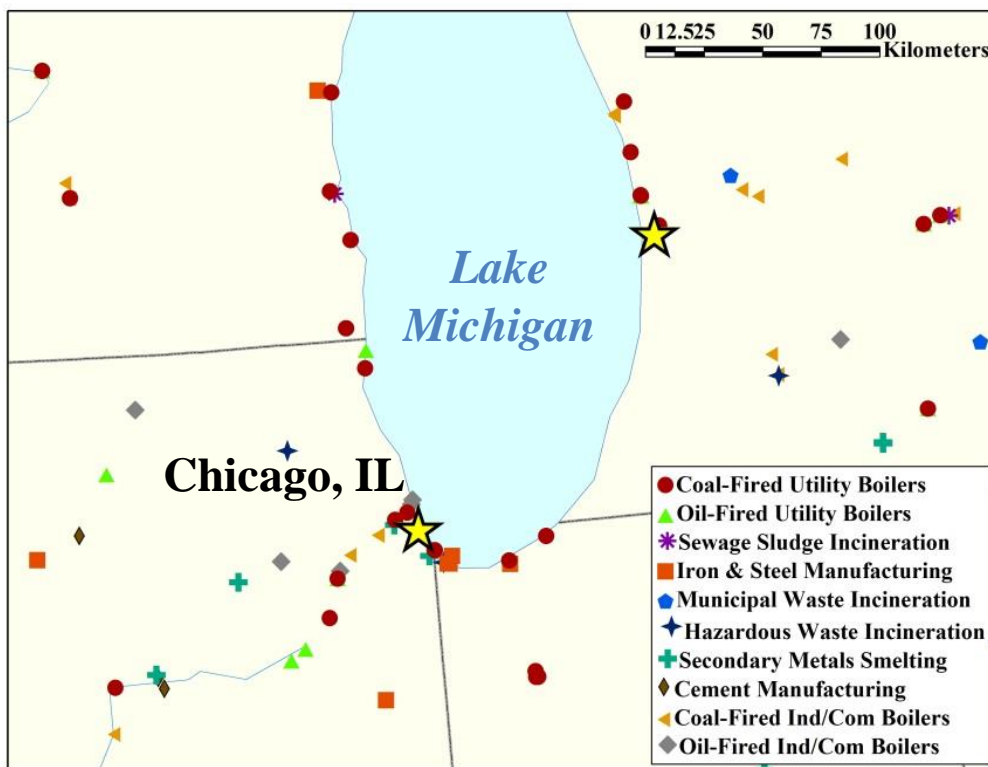
- Trajectories suggest MBL influence.
- No indication that air mass picked up anthro. emissions over land.
- Anti-correlation between RM and CHBr<sub>3</sub> may suggest Br oxidation.





## 2. Hg in Urban Emissions

### RF-15: Chicago/Gary Urban/Industrial Plume



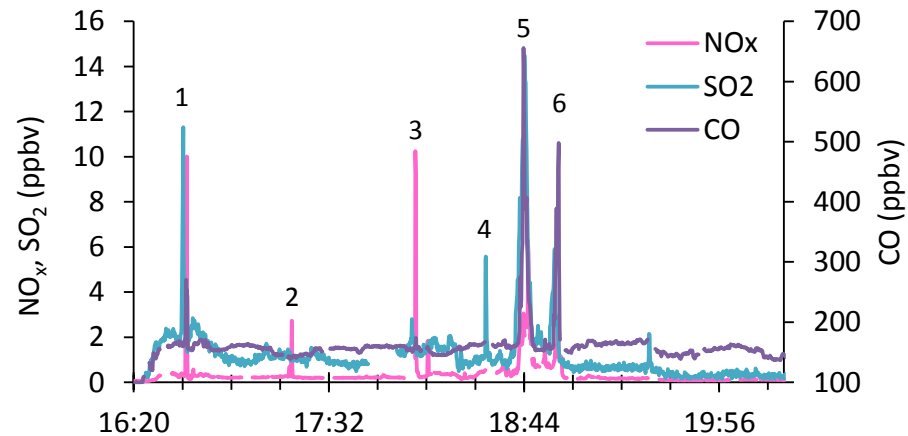
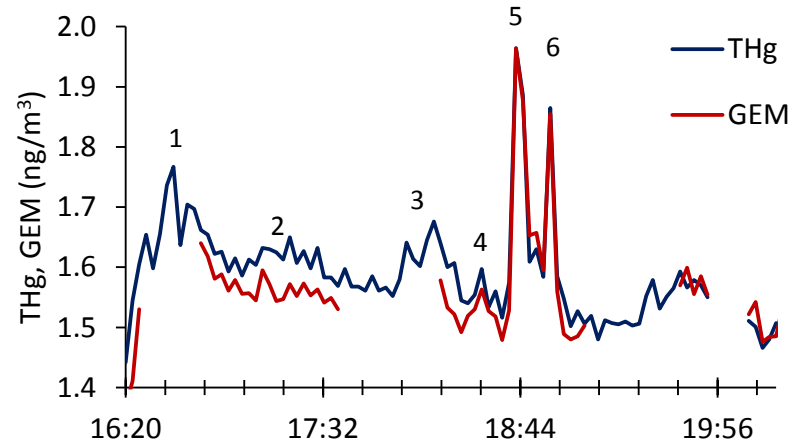
Hg point sources that emit  $\geq 0.1 \text{ kg Hg yr}^{-1}$  based on the 2005 EPA NEI (Gratz et al., 2012).

C-130 flight track during RF-15 on 7/7/2013.

# 2. Hg in Urban Emissions

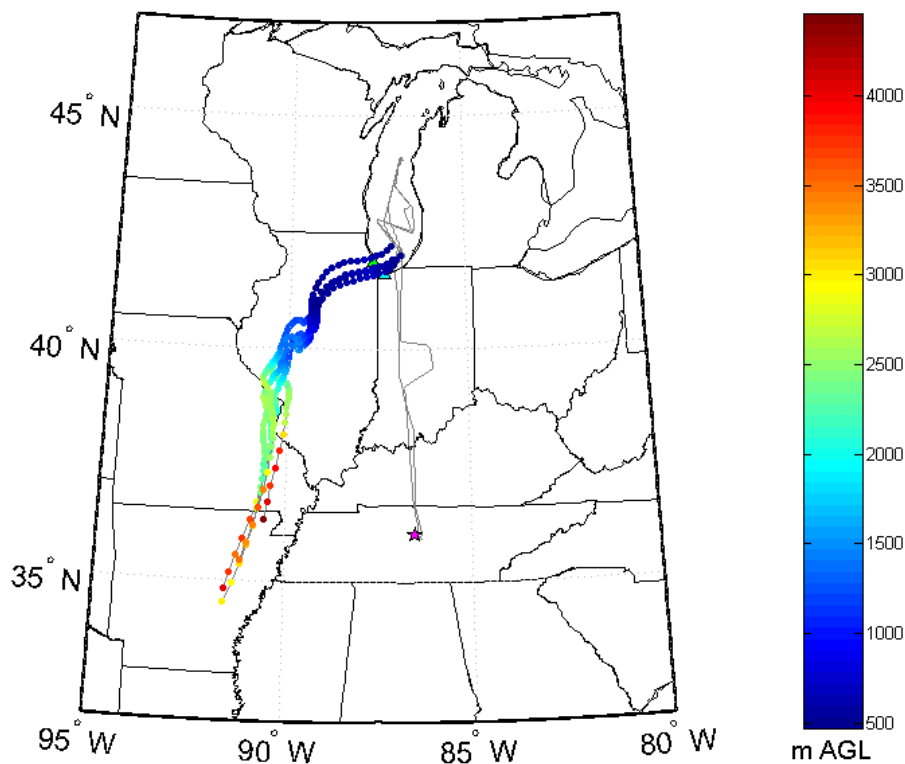
## RF-15: Chicago/Gary Urban/Industrial Plume

- Identified 6 plumes in data while over Lake Michigan.
- Plumes 5 and 6 represent best capture of the Chicago/Gary urban/industrial plume.
- No detectable RM in plume.
- Plume 5 enhancements:
  - THg: 1.96 ng m<sup>-3</sup>
  - NO<sub>x</sub>: 3.0 ppbv
  - SO<sub>2</sub>: 14.5 ppbv
  - CO: 656 ppbv

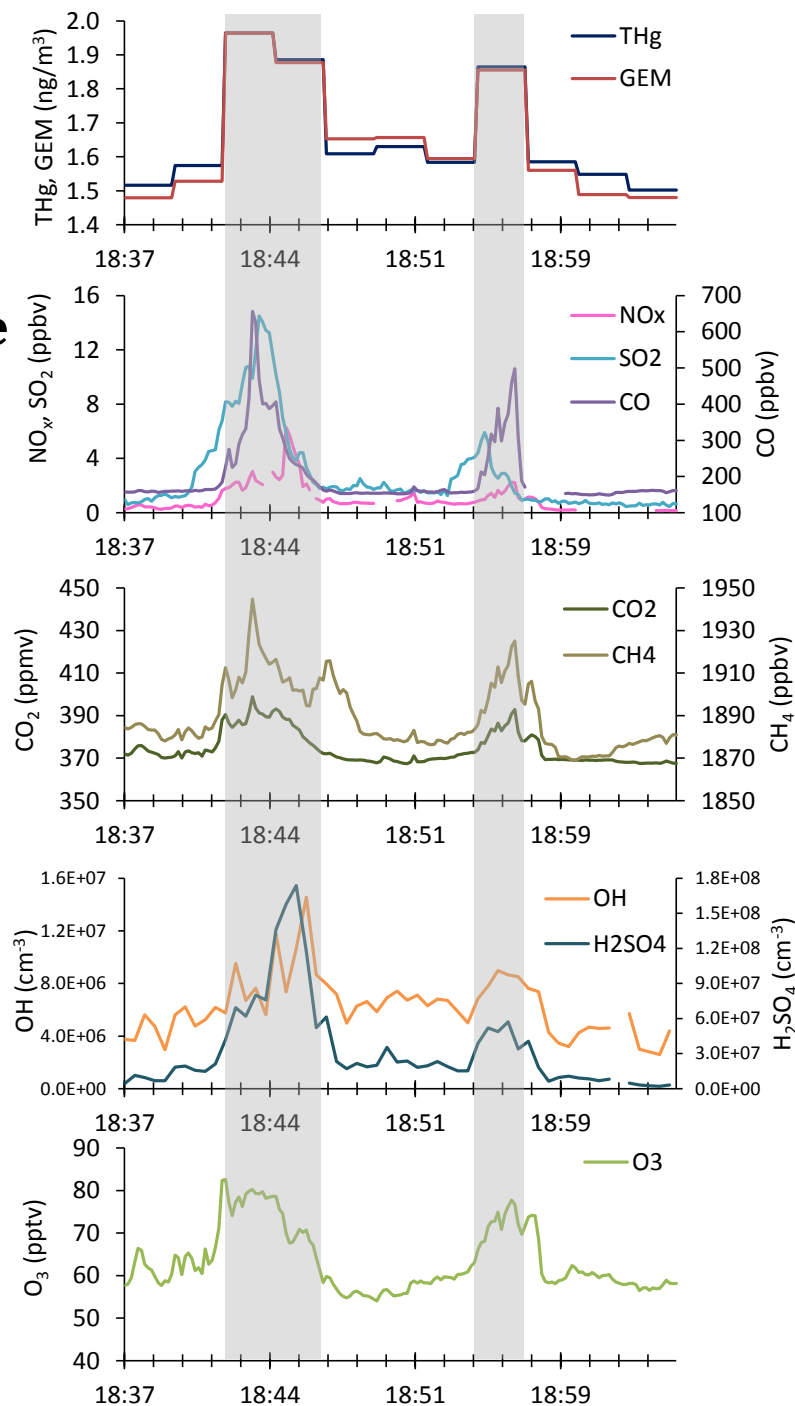


# 2. Hg in Urban Emissions

## RF-15: Chicago/Gary Urban/Industrial Plume

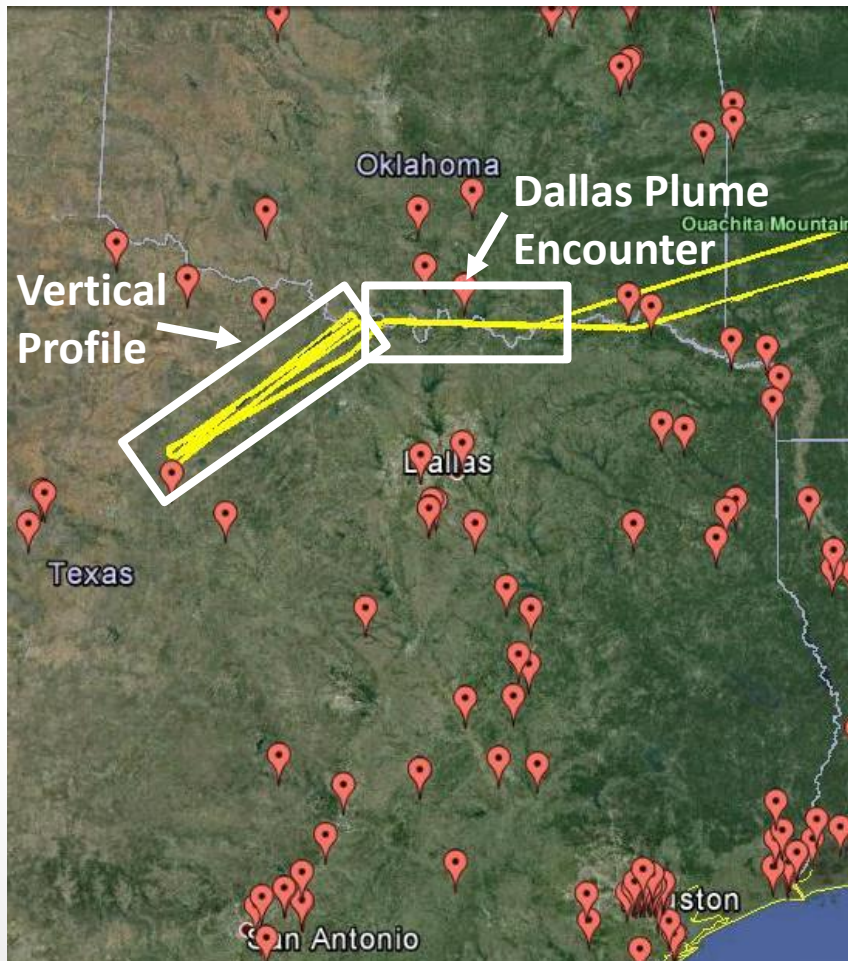


*FLEXPART 72-hour back-trajectories from 18:40 UTC – 18:55 UTC*



## 2. Hg in Urban Emissions

### RF-09: Dallas Urban Plume

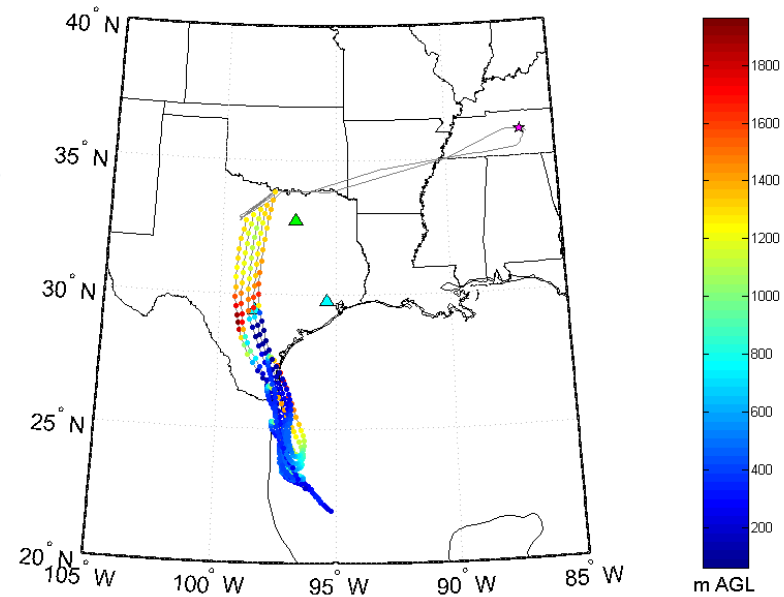
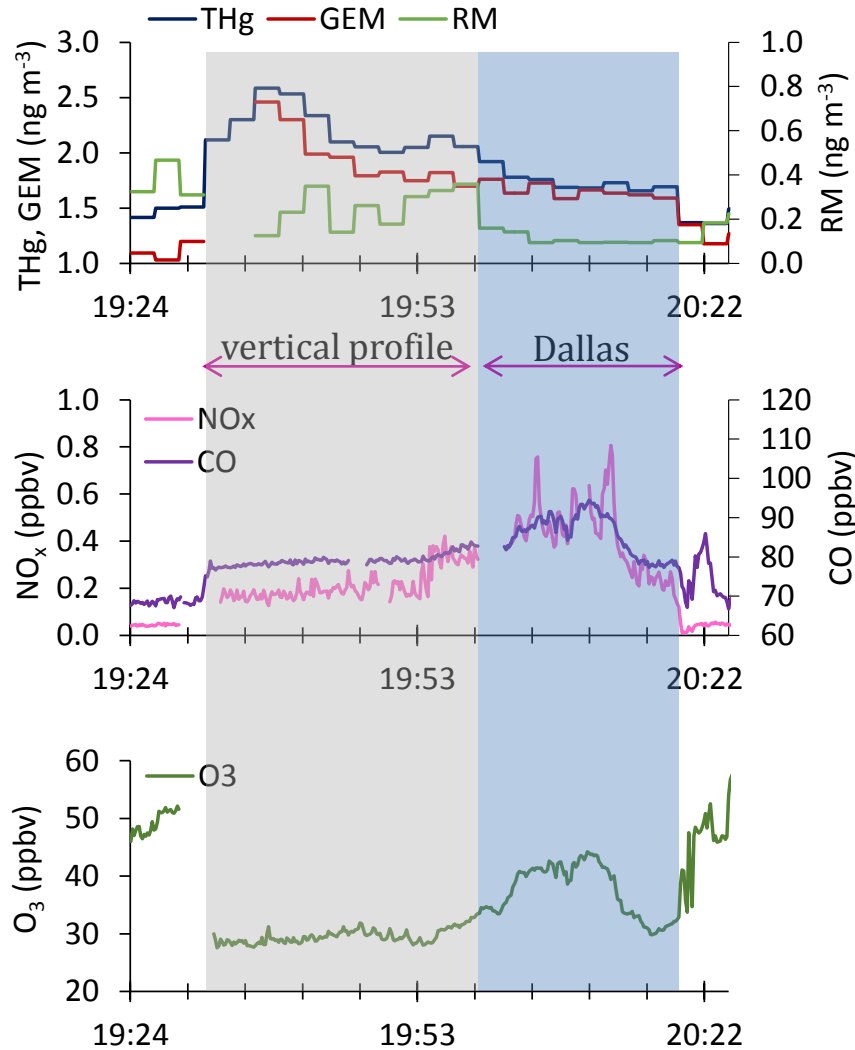


- After BL segment of vertical profile, C-130 traveled east to capture Dallas plume.
- Expect Dallas emissions to have more urban signature compared to mixed urban/industrial signature of Chicago/Gary.

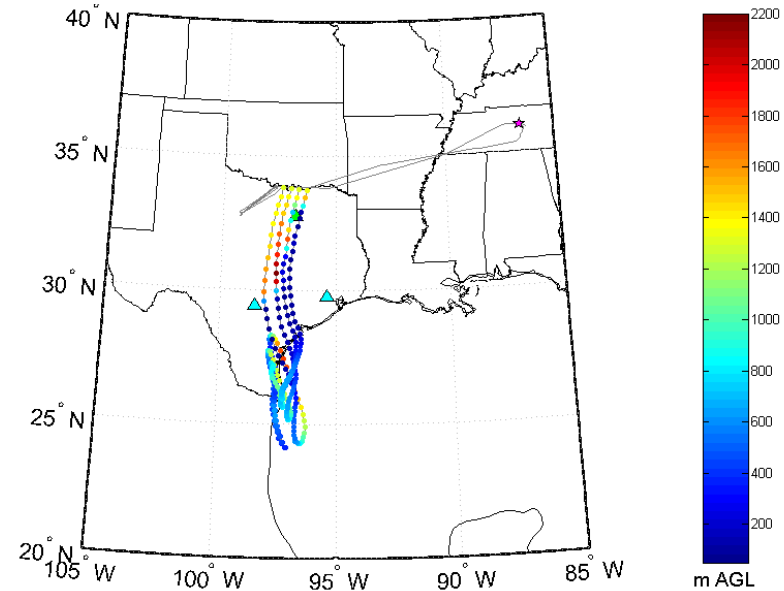
**LEFT:** C-130 flight track over North Texas during RF-09, and Hg point sources in the 2011 TRI.

# 2. Hg in Urban Emissions

## RF-09: Dallas Urban Plume

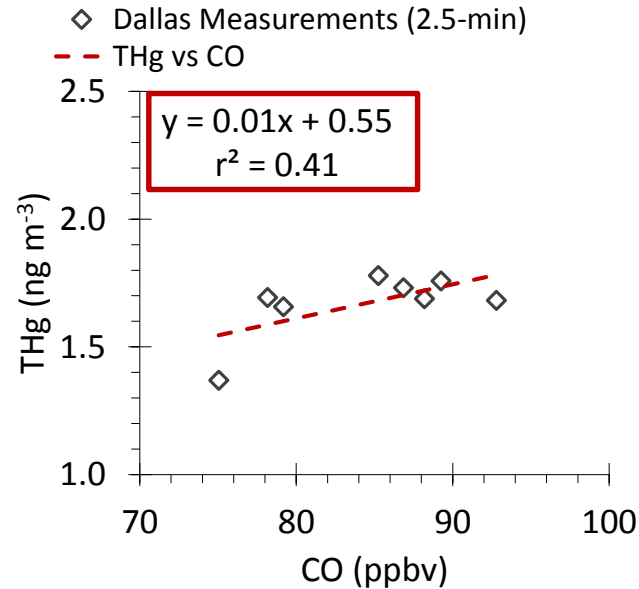
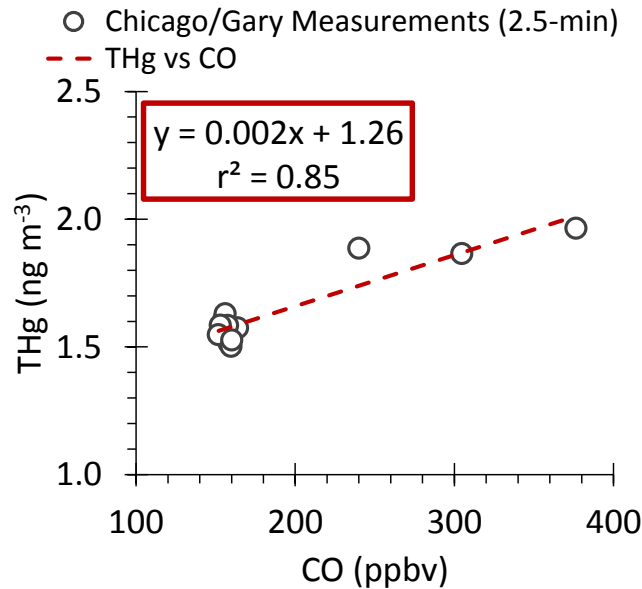


*FLEXPART 72-hour trajectories during BL segment of vertical profile.*



*FLEXPART 72-hour trajectories during the Dallas plume encounter*

## 2. Hg in Urban Emissions



- Ratios of Hg to anthropogenic tracers (e.g. CO) can be used to characterize urban emissions.
- Comparing measured ratios to GEOS-Chem modeled ratios will allow us to evaluate how well emission inventories characterize emissions from all Hg sources in selected urban areas.

# Conclusions and Future Work

## 1. RM in the free troposphere

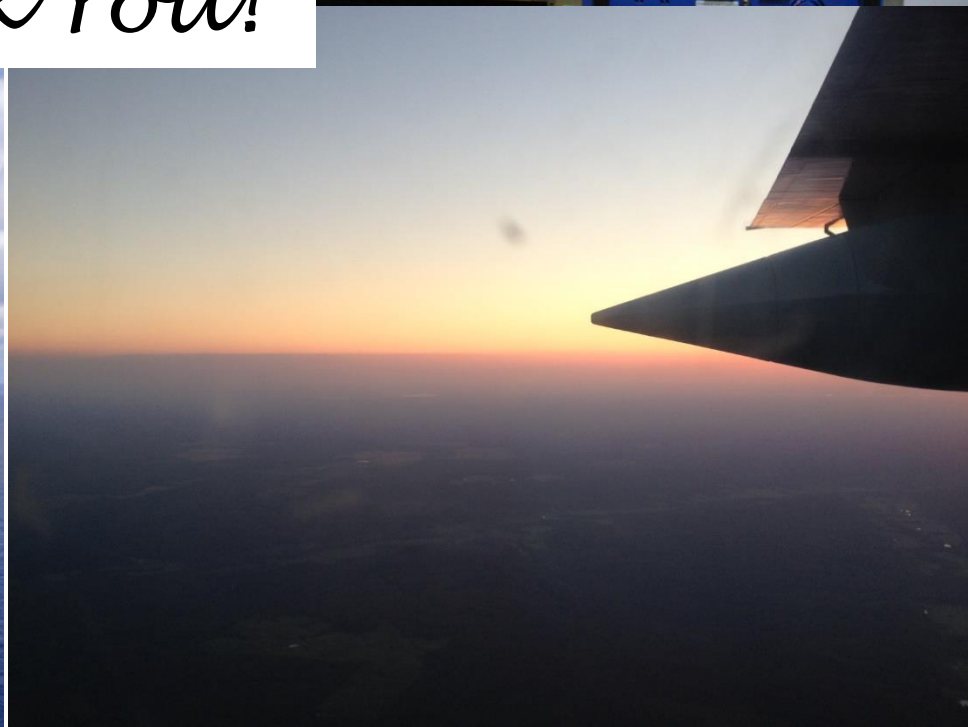
- We identified different sources of RM in the FT over Texas and over the Atlantic.
- We will continue exploring C-130 measurements and model output to evaluate sources of RM in the FT (e.g. marine Br source).

## 2. Hg emissions from major urban areas

- There are significant differences in Hg emissions from urban/industrial areas vs. urban areas.
- Other C-130 measurements (e.g. VOCs) may be useful in characterizing urban emissions.
- We will examine other urban areas (e.g. Houston, Birmingham) to compare/contrast with Chicago and Dallas.
- We will compare measured vs. GEOS-Chem modeled Hg concentrations and ratios to evaluate inventories and model representation of urban Hg emissions.



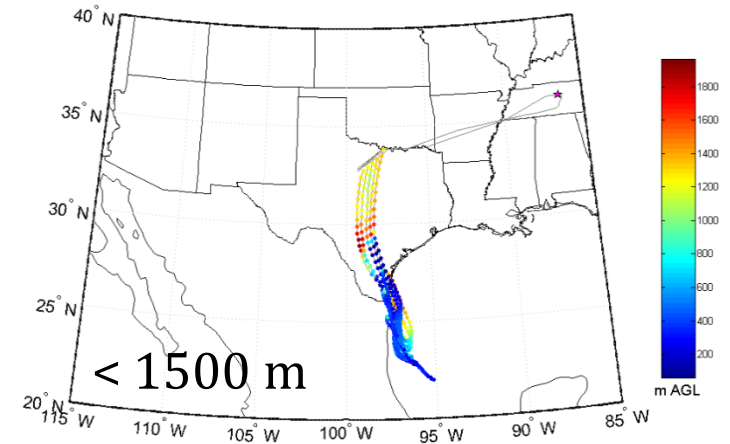
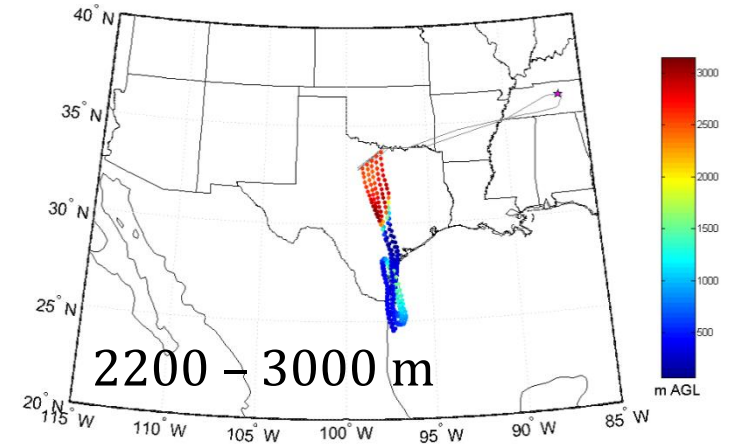
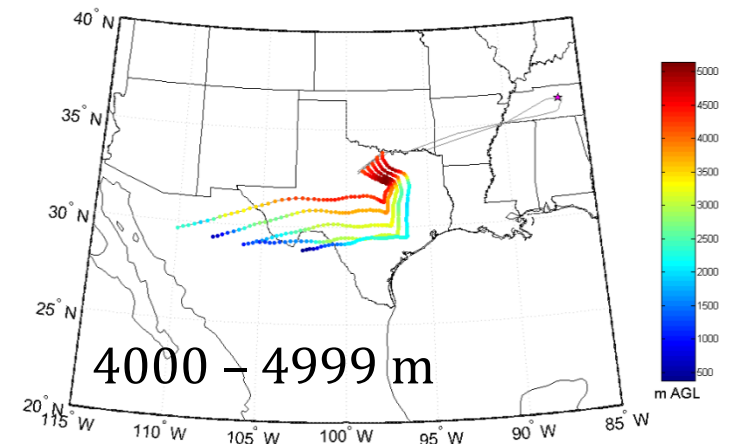
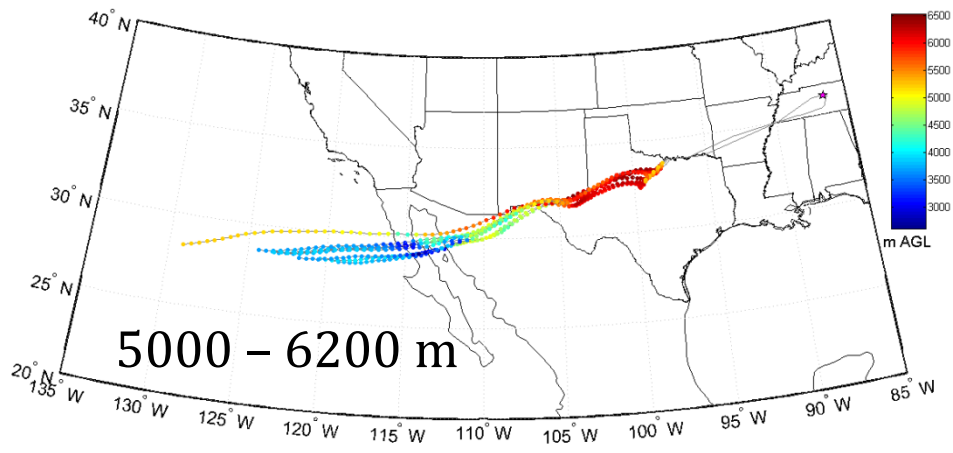
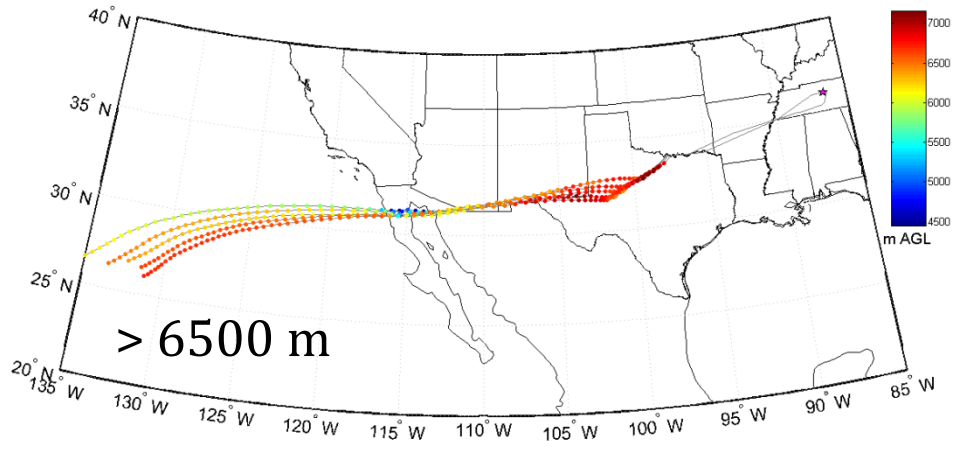
*Thank You!*



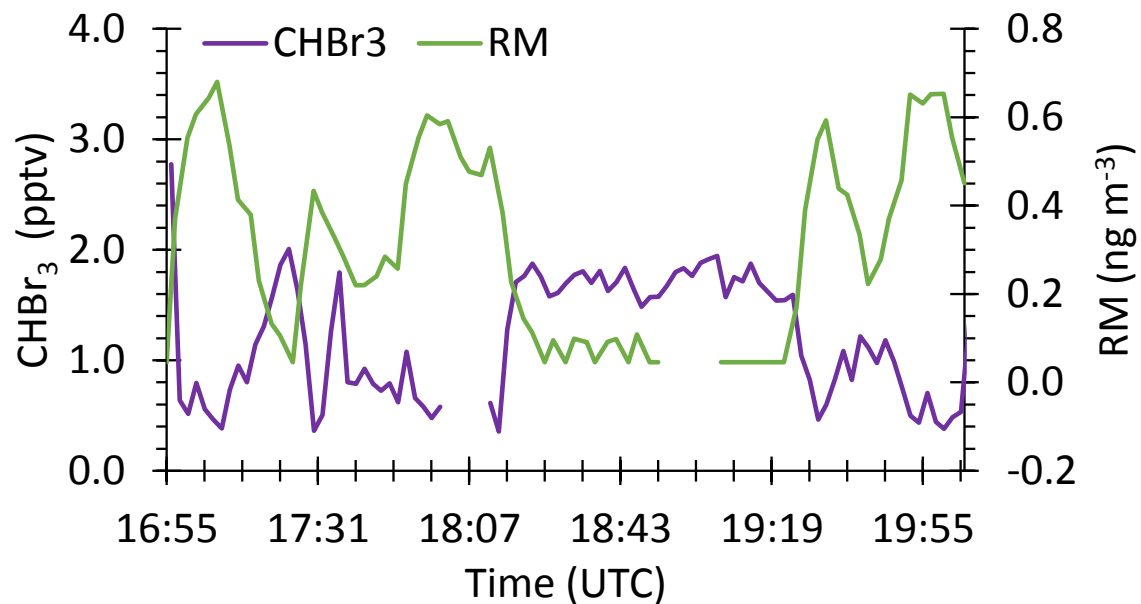


# Extra Slides

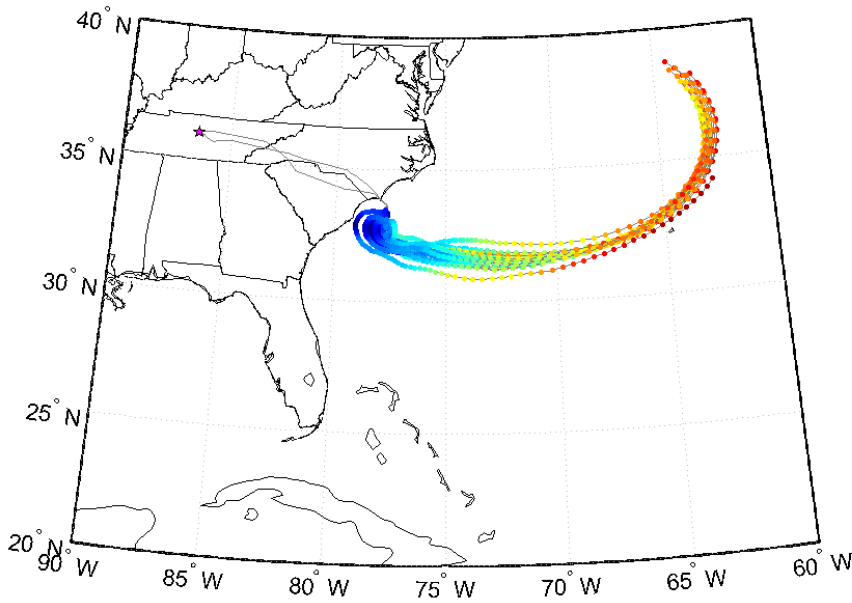
# RF-09 Trajectories



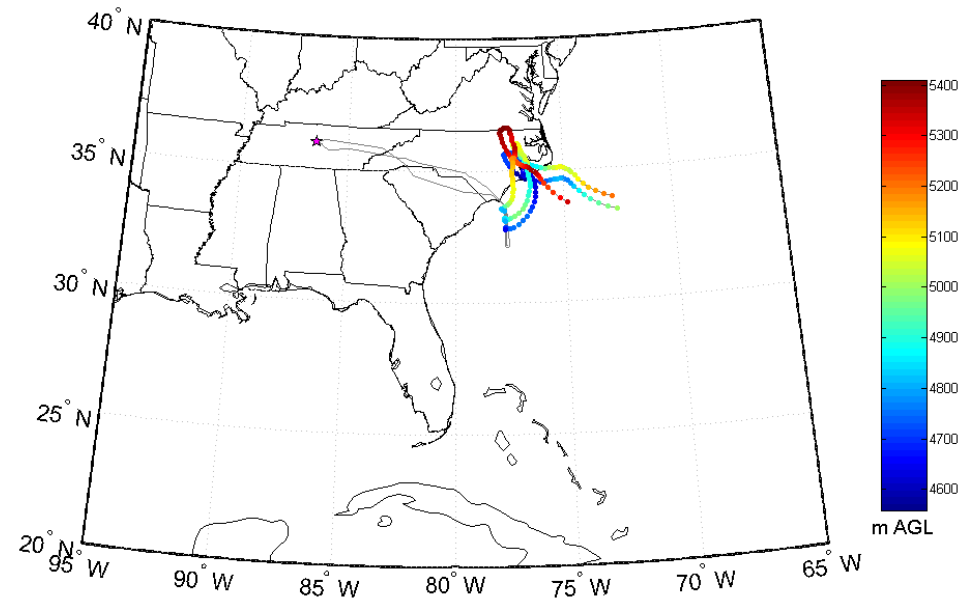
# RF-16: RM and CHBr<sub>3</sub> Time Series



# RF-16 Trajectories



*FLEXPART 72-hr trajectories, < 1000 m AGL*



*FLEXPART 72-hr trajectories, > 4000 m AGL*

# RF-15 Trajectories

