

Observations and Model Analysis of Enhanced Reactive Mercury in the Free Troposphere

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NOMADSS Data Discussion

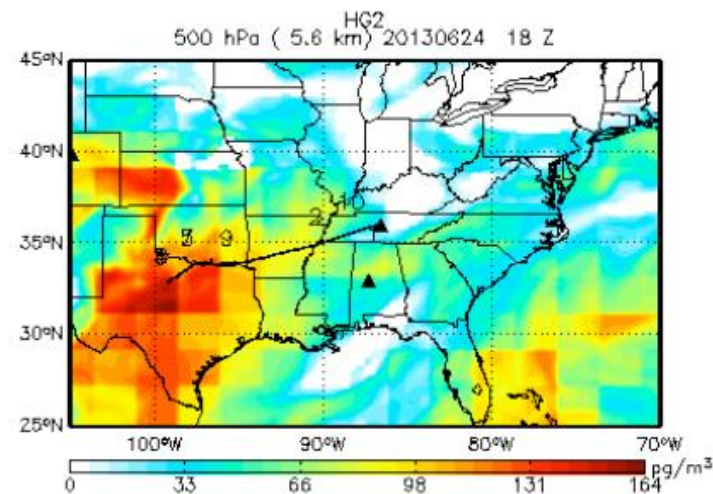
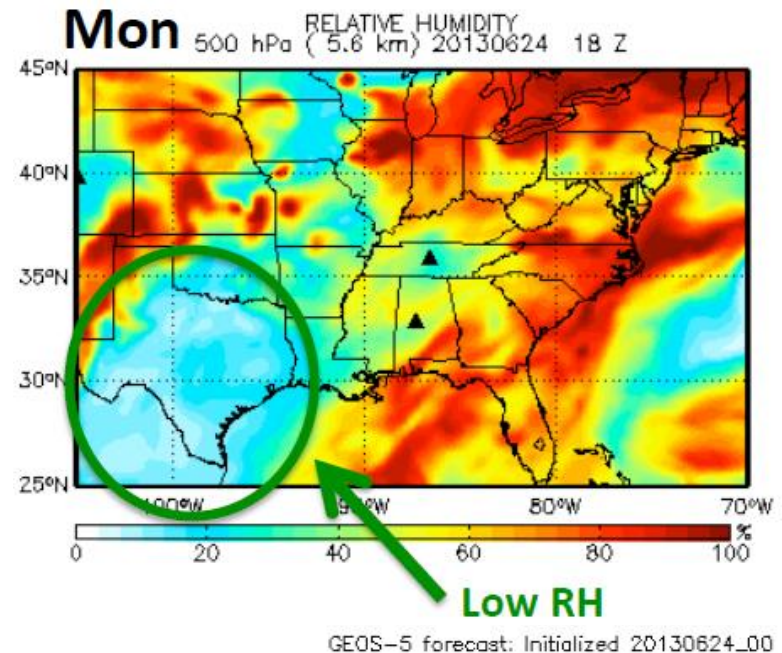
May 12, 2014

Reactive Mercury (RM) in the Free Troposphere (FT) during NOMADSS

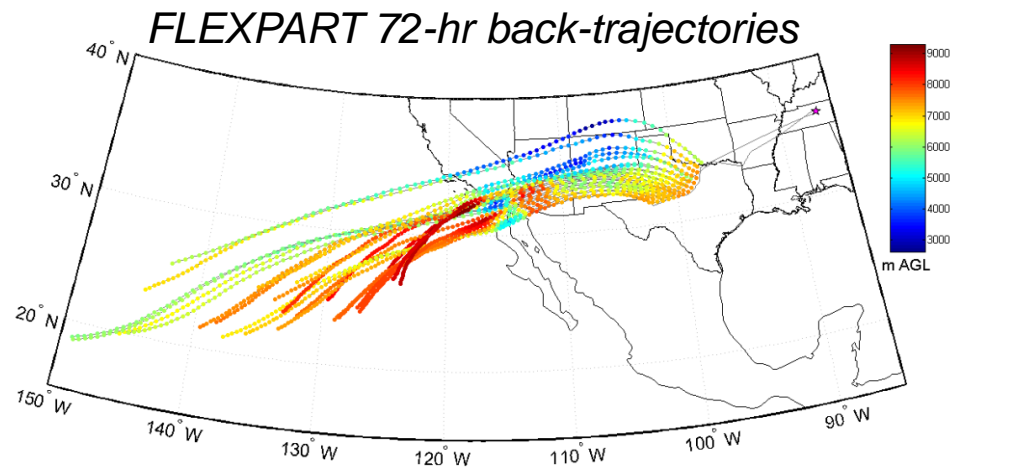
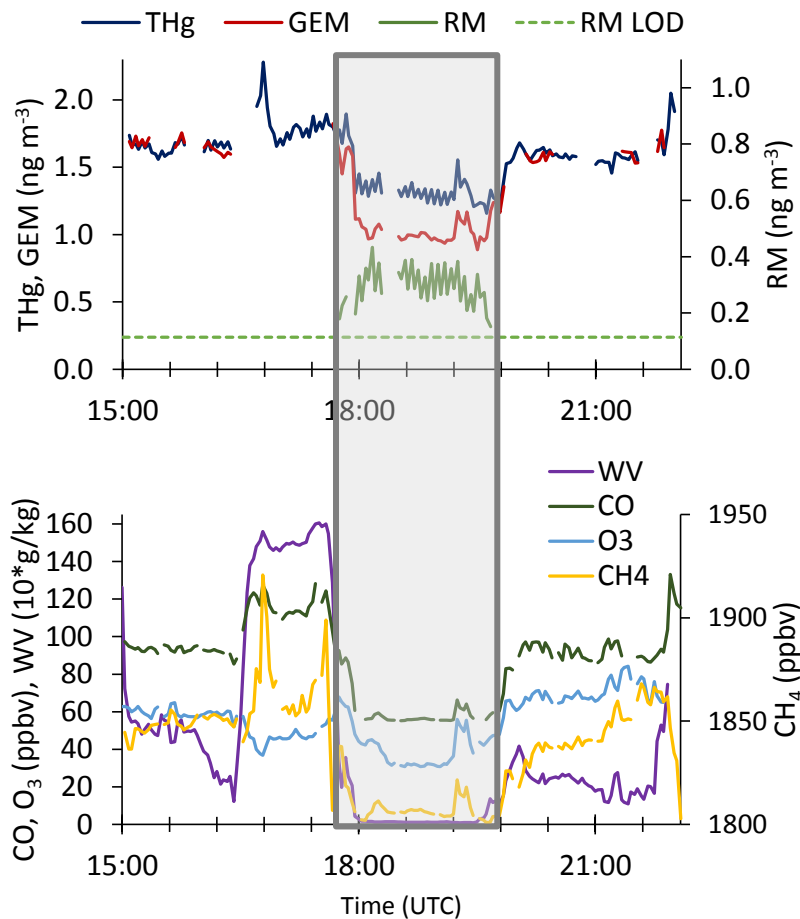
- A limited number of ground-based and airborne studies have identified the presence of enhanced RM in the FT.
- Chemical Transport Models (CTMs; e.g. GEOS-Chem) suggest oxidation of GEM by Br is the major source of RM in the FT.
- We extensively measured enhanced RM in the FT in two different locations during NOMADSS:
 - North Texas
 - Atlantic Coast
- Standard GEOS-Chem underestimates RM in the FT by factor of 3-5.
- GEOS-Chem 3xBr model reproduces RM over Texas, but not over the Atlantic.

RM in the FT: Texas

- Enhanced RM was often forecast in the dry FT 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**



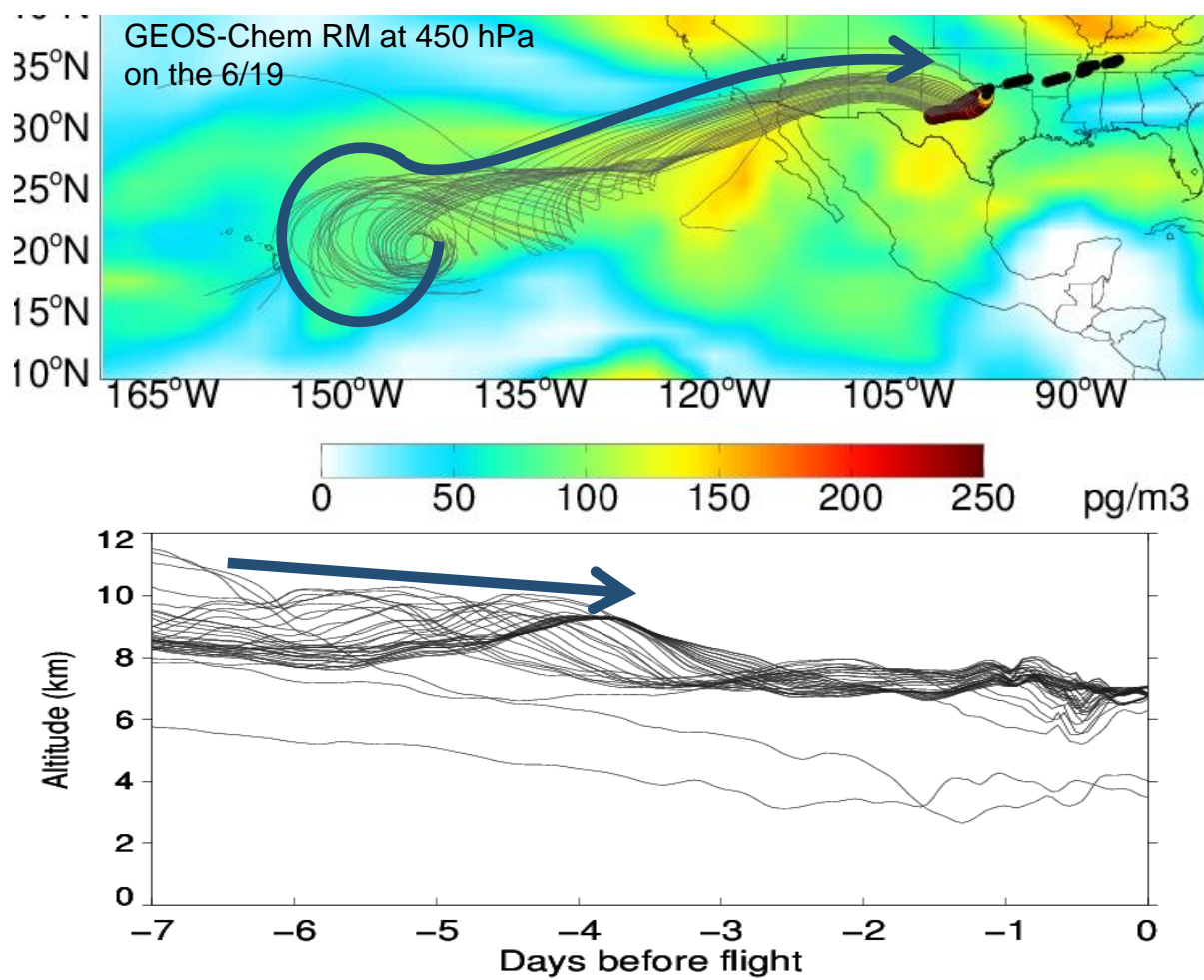
RM in the FT: Texas



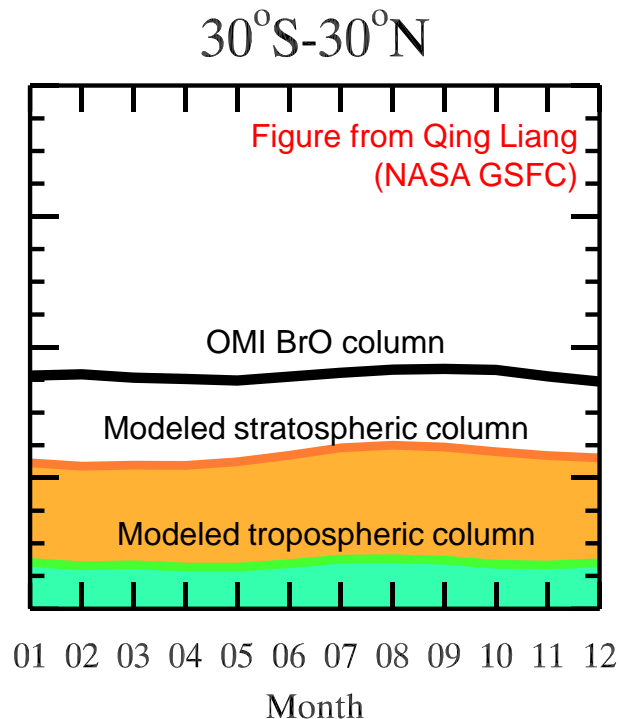
- Observations and back-trajectories suggest RM source is the eastern sub-tropical Pacific FT.
- Very clean air mass - may have circulated around Pacific High for days.
- No indication for influences of anthropogenic or marine emissions, or stratospheric subsidence.

RM in the FT: Texas

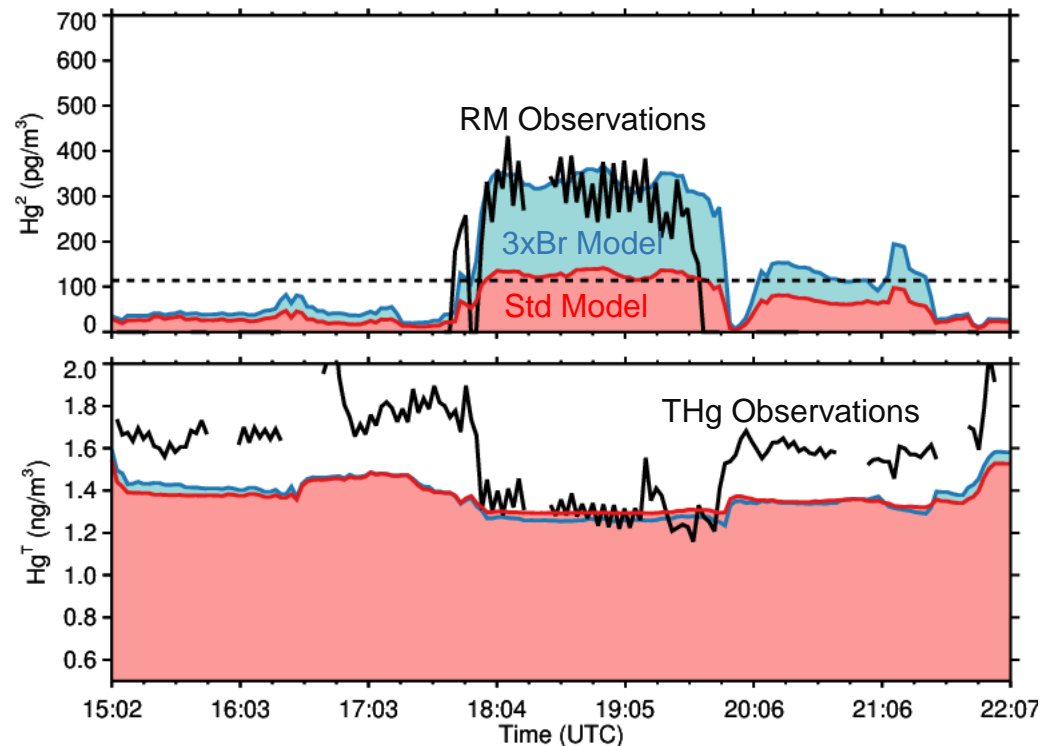
HYSPLIT 7-day back-trajectories overlaid on Standard GEOS-Chem RM at 450 hPa



RM in the FT: Texas

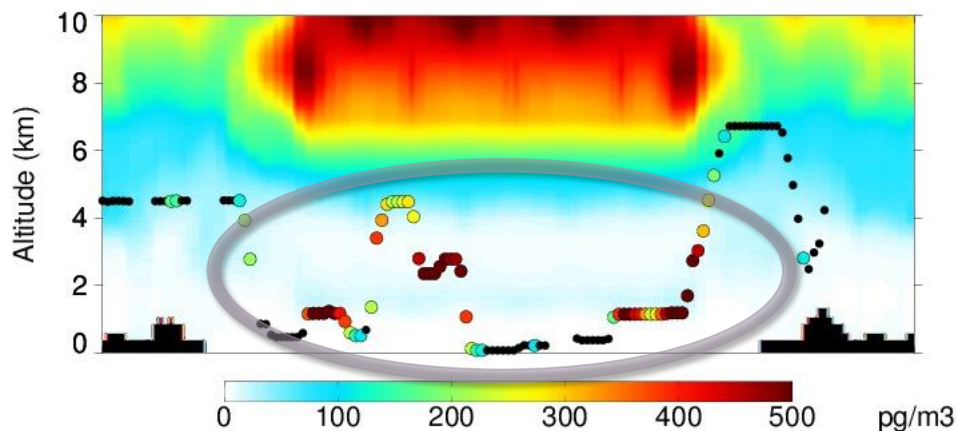


Modeled tropospheric BrO
concentrations are biased low

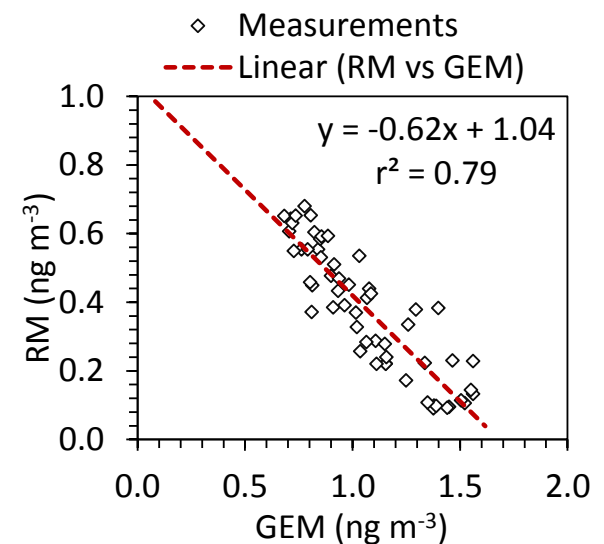
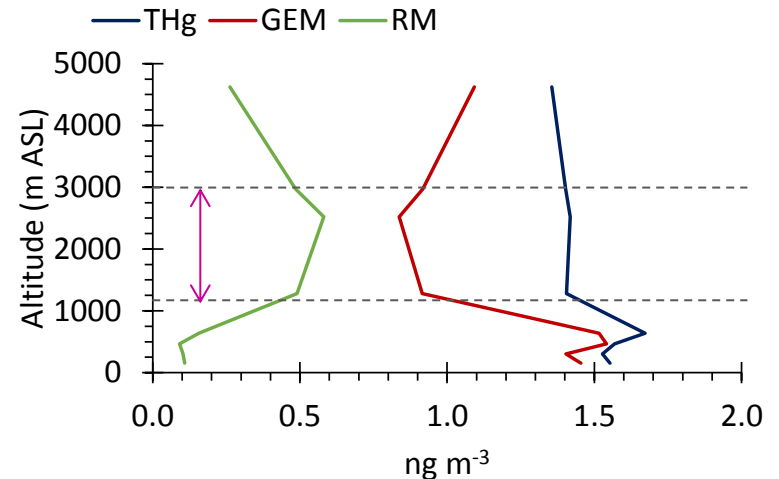


Tripling concentrations of Br at 500 hPa between 40°S and 40°N allows GOES-Chem to reproduce the observed RM concentrations over Texas.

RM in the FT: Atlantic Ocean

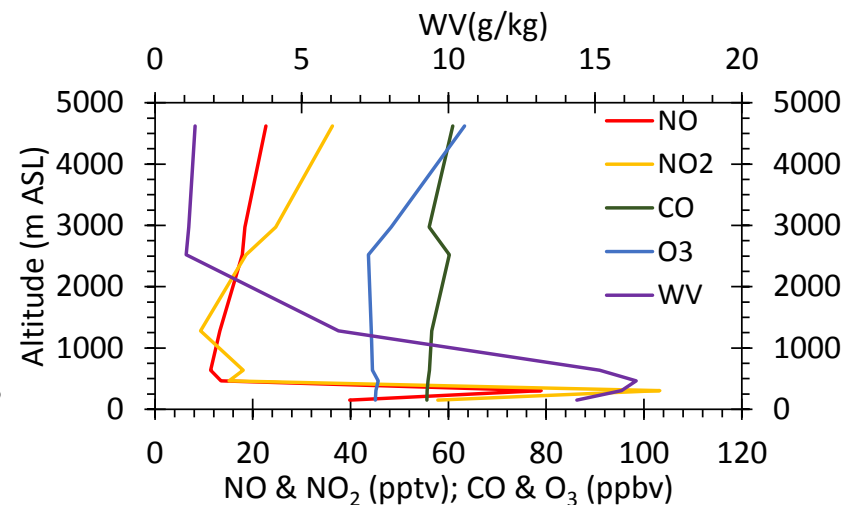
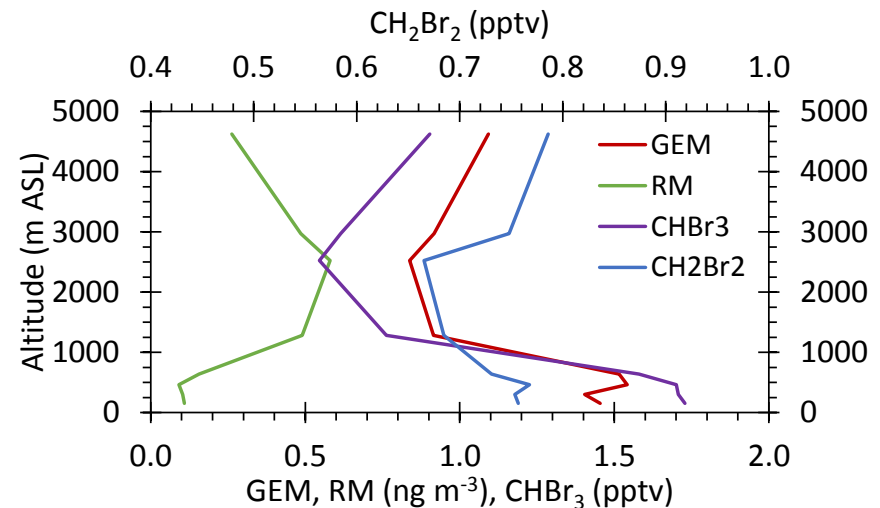
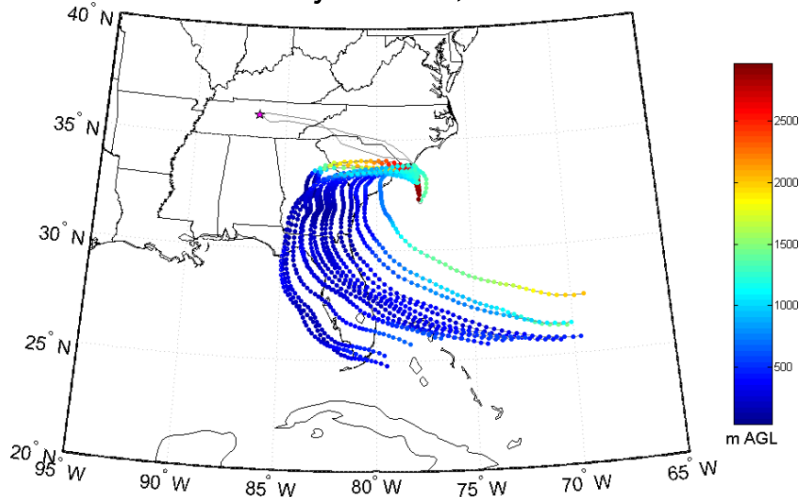


- We observed enhanced RM between 1250 – 3000 m ASL.
- RM vs. GEM across the profile shows strong evidence for GEM oxidation.



RM in the FT: Atlantic Ocean

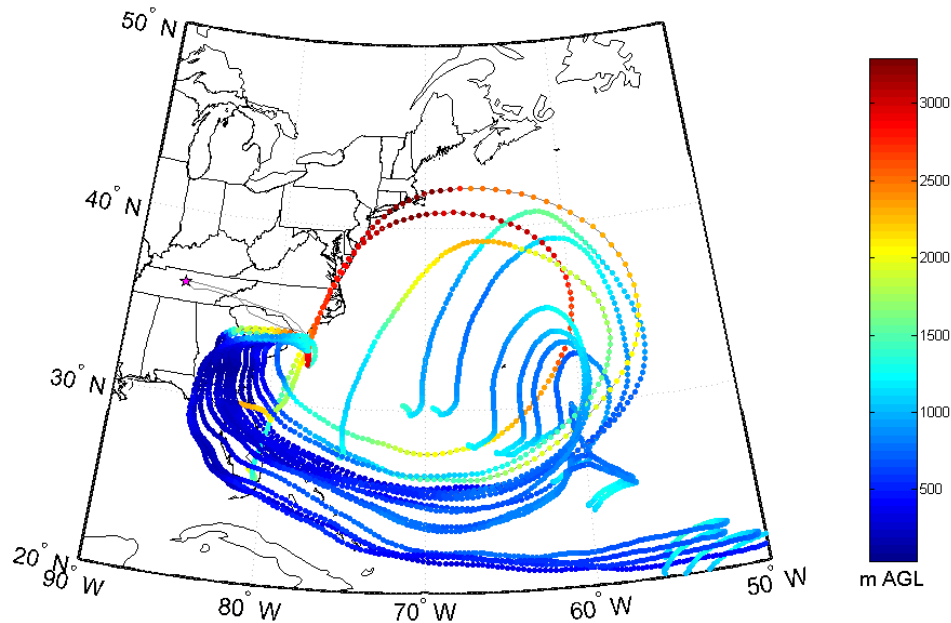
FLEXPART 72-hr trajectories, 1250-3000 m AGL



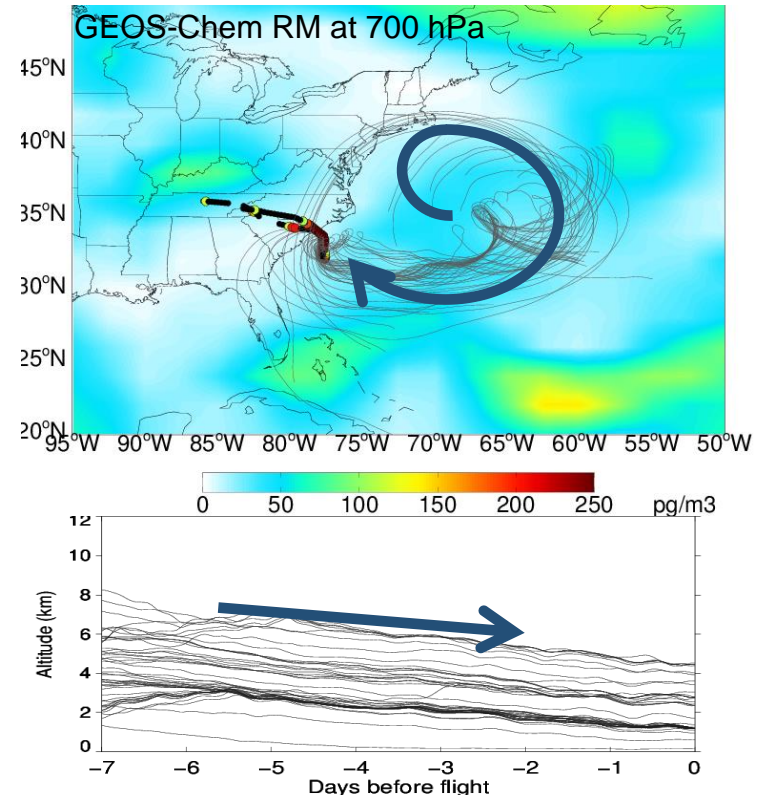
- 72-hr FLEXPART trajectories suggest MBL air transported over SE U.S.
- BUT... No indication that air mass picked up fresh marine or anthropogenic emissions.
- Anti-correlation between RM and CHBr₃ may suggest sampling of different air masses (tropical marine air vs. FT) at different altitudes.

RM in the FT: Atlantic Ocean

FLEXPART 10-day back-trajectories, 1250-3000 m

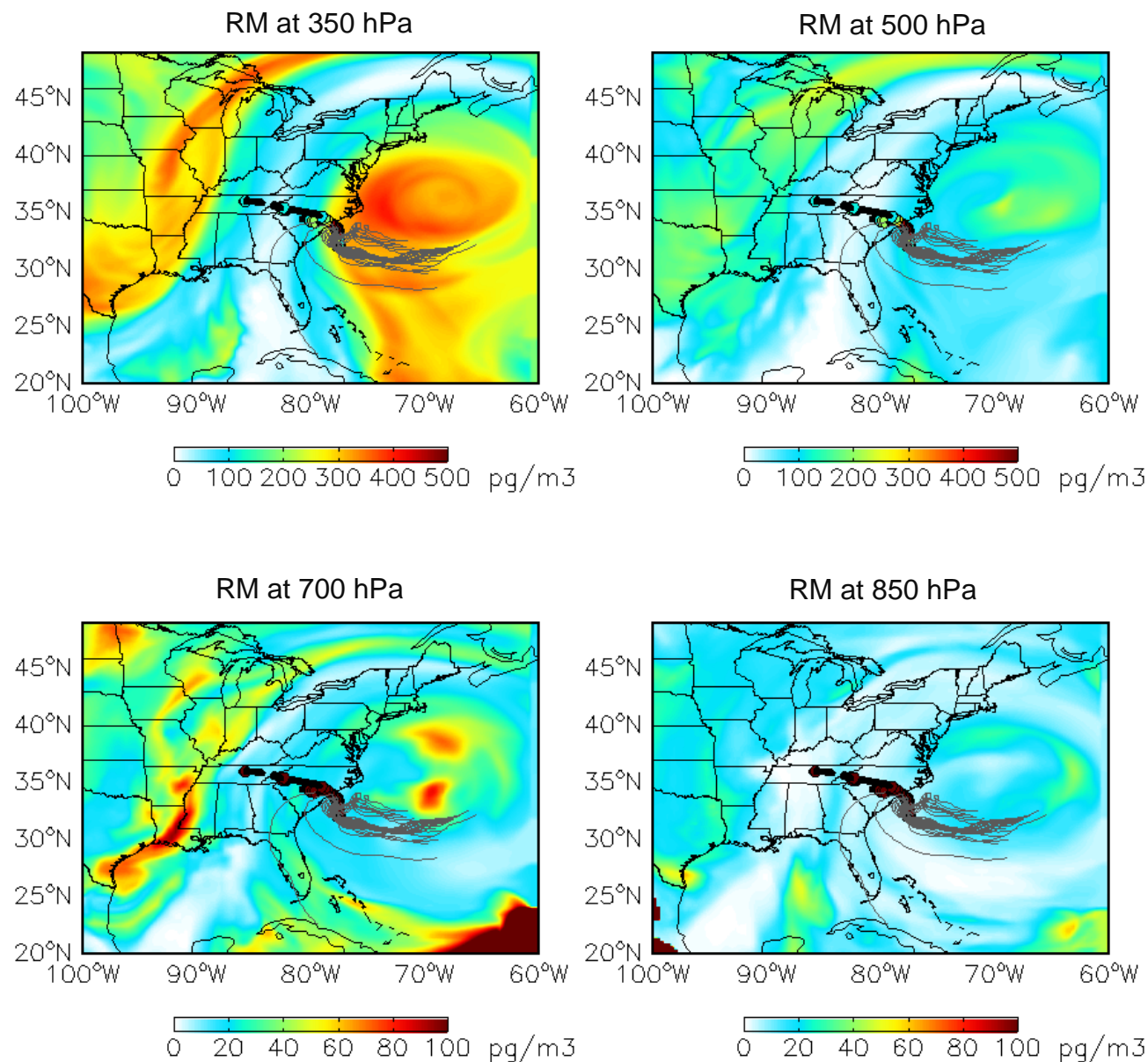


HYSPLIT 7-day back-trajectories, all altitudes

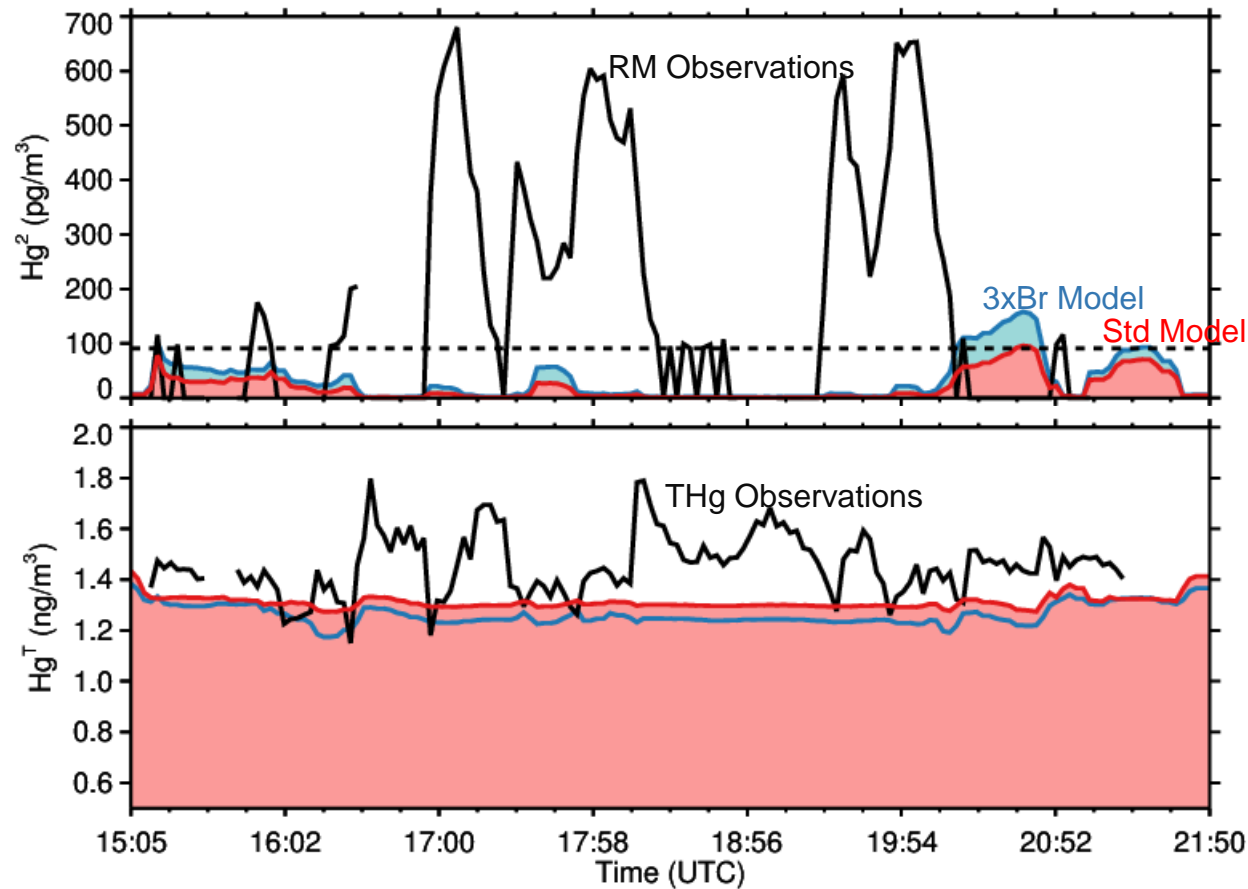


Longer back-trajectories (> 7-days) suggest high-RM air mass circulated around Atlantic high pressure system. But, there are differences between the modeled transport paths in HYSPLIT and FLEXPART.

72-hour HYSPLIT back-trajectories starting between 1000-3000m overlaid on GEOS-Chem modeled RM on 07/05/13 (3 days before flight).



RM in the FT: Atlantic Ocean



Even after tripling Br concentrations above 500 hPa between 40°S and 40°N GOES-Chem cannot reproduce the observed RM over the Atlantic.

Summary

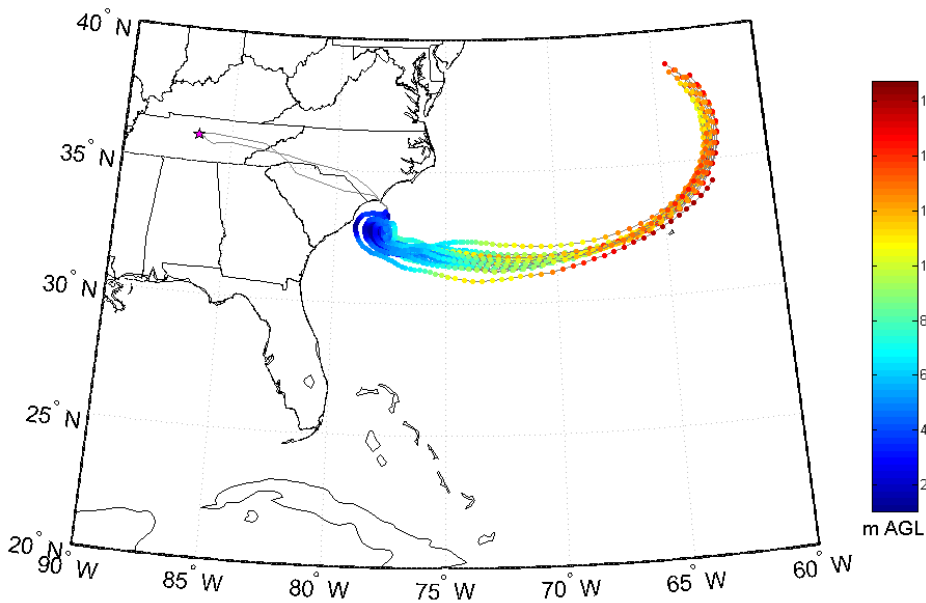
- RM observations in the FT over Texas and the Atlantic are much higher than models have predicted.
- Enhanced RM over Texas appears associated with transport from the sub-tropical Pacific where the air mass likely circulated around / subsided within the Pacific High prior to transport over N. America.
- Enhanced RM over the Atlantic may be associated with subsidence and GEM oxidation within the Atlantic high pressure system.
- Standard GEOS-Chem model does not reproduce RM observations in either case.
- 3xBr GEOS-Chem reproduces enhanced RM over Texas but not over the Atlantic. **WHY?**

Next Steps

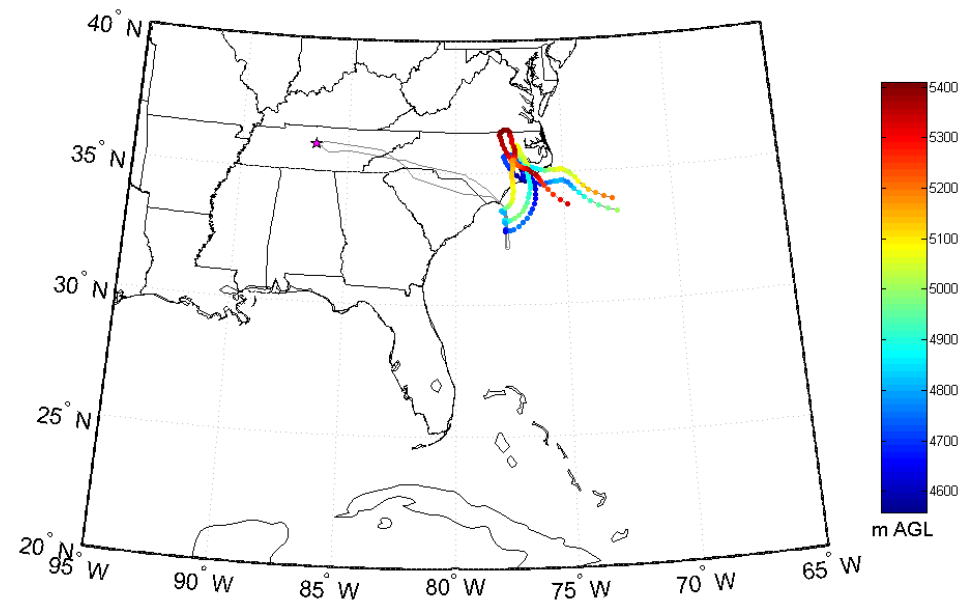
- Determine why GEOS-Chem 3xBr model does not reproduce RM observations over the Atlantic.
 - Problems with model meteorology?
 - Different oxidant? Multiple oxidants?
- Continue exploring measurement dataset for indications of air mass origin during RM enhancement over Atlantic.
- N. Selin will run GEOS-Chem simulation that will track time from scavenging to show how long an air parcel in the model has been dry. Maybe model has too much scavenging in the FT → not enough time for conversion reactions.
- Other suggestions?

Extra Slides

RM in the FT: Atlantic Ocean

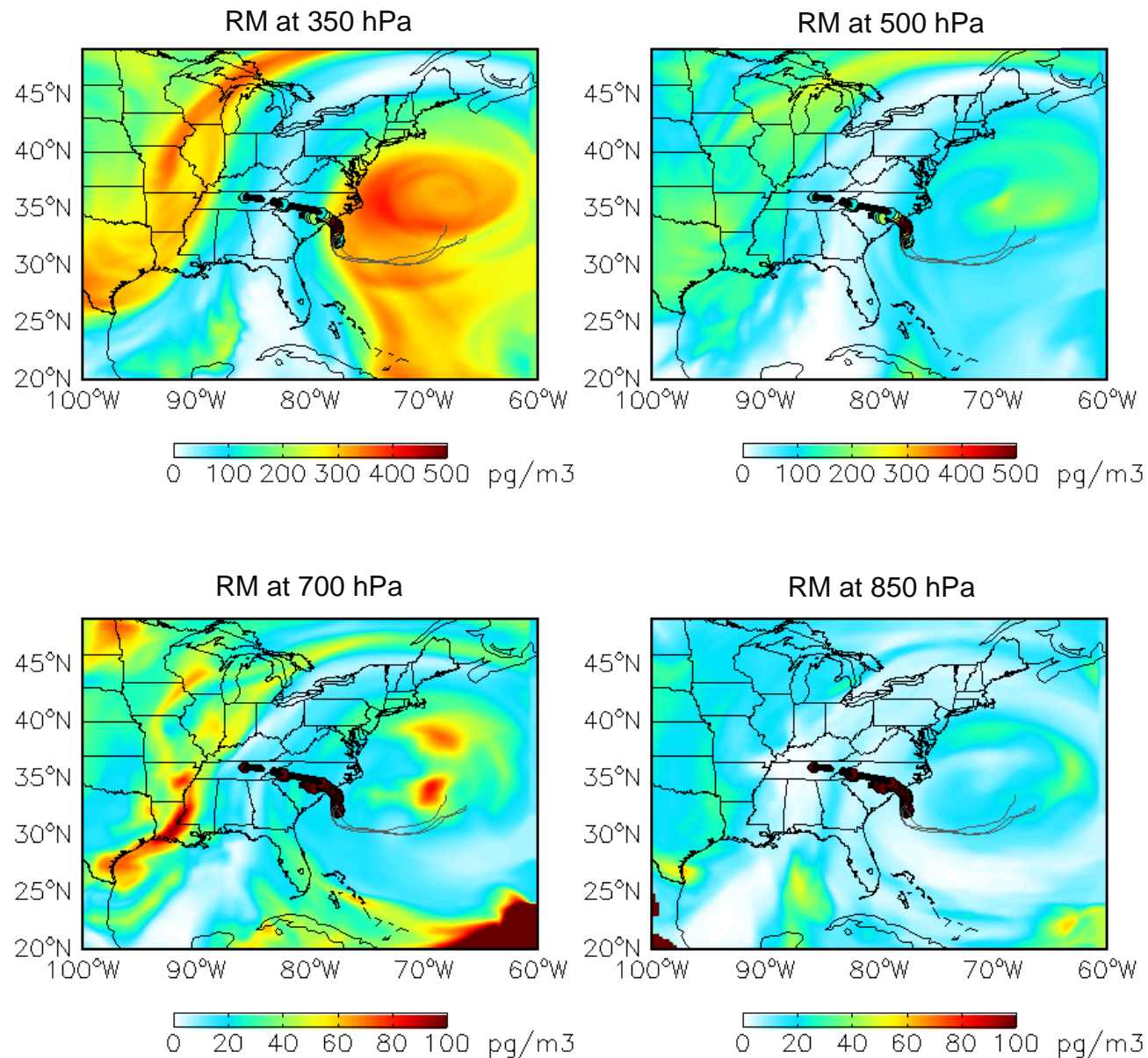


FLEXPART 72-hr trajectories, < 1000 m AGL



FLEXPART 72-hr trajectories, > 4000 m AGL

72-hour HYSPLIT back-trajectories starting between 0-1000m overlaid on GEOS-Chem modeled RM on 07/05/13 (3 days before flight).



72-hour HYSPLIT back-trajectories starting between 3000-7000m overlaid on GEOS-Chem modeled RM on 07/05/13 (3 days before flight).

