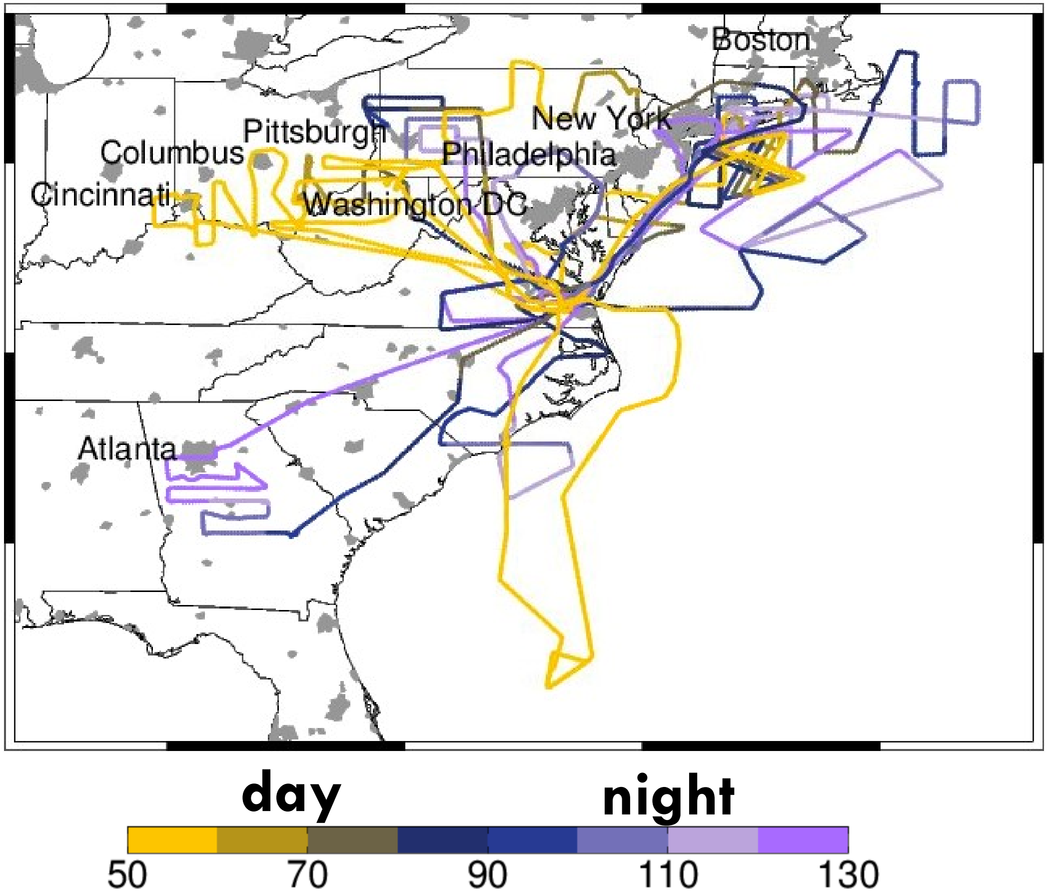
Co-PIs: L. Jaeglé (UW)
S. Brown (NOAA)
R. Cohen (UCB)
J. Jimenez (CU)
R. Weber (GT)
J. Dibb (UNH)

$$ Funding: NSF-AGS $$
NOAA ESRL
NOAA ARL
NASA GSFC and LRC

Special Issue in *JGR-Atmospheres*
Also relevant publications in PNAS and GRL

Joel Thornton thornton@atmos.uw.edu
Department of Atmospheric Sciences; University of Washington

WINTER campaign Feb 1 – March 13, 2015



➢ **50% of flights hours at night**
➢ **71% within 1 km of the surface**

Measurements

- Gas-phase: NO_2, NO, HNO_3, N_2O_5, ∑PANs, ∑ANs, HONO, NOy, $ClNO_2$, HCl, Cl_2, CO, O_3, SO_2, NH_3, HCHO HNCO, HCOOH, alkanes, alkenes, alkynes, CFCs, halons
- Aerosol composition (ToF-AMS, PILS, Filter): organic aerosol, SO_4^{2-}, NH_3^+, NO_3^-, Cl^-, Na^+, Mg^{2+}, Ca^{2+}
- Aerosol size distribution, actinic flux, meteorological parameters

Modeling

→**GEOS-Chem (UW) nested grid (25km)**
→NOAA ARL 12km PM model
→FLEXPART (UW) w/NO_x tracer

WINTER vs. AQUARIUS

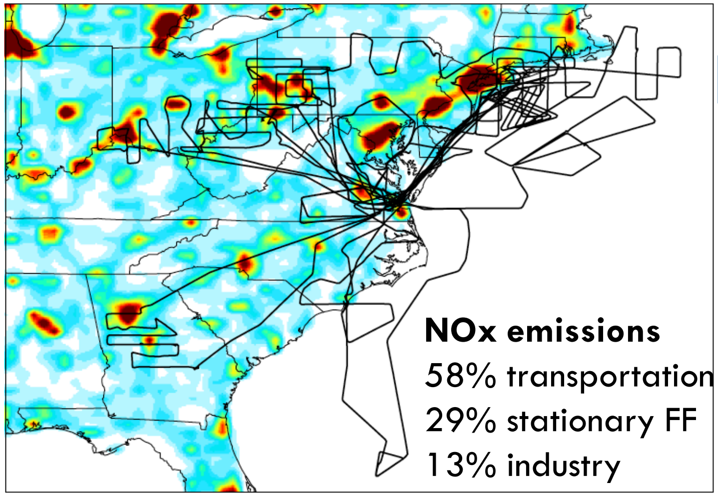
- Eastern U.S. meteorology can be rather different from west / mountain west
  - Stagnation episodes with strong inversions
  - Frontal systems and cold-air outbreaks (off-shore transport)
  - Solar radiation

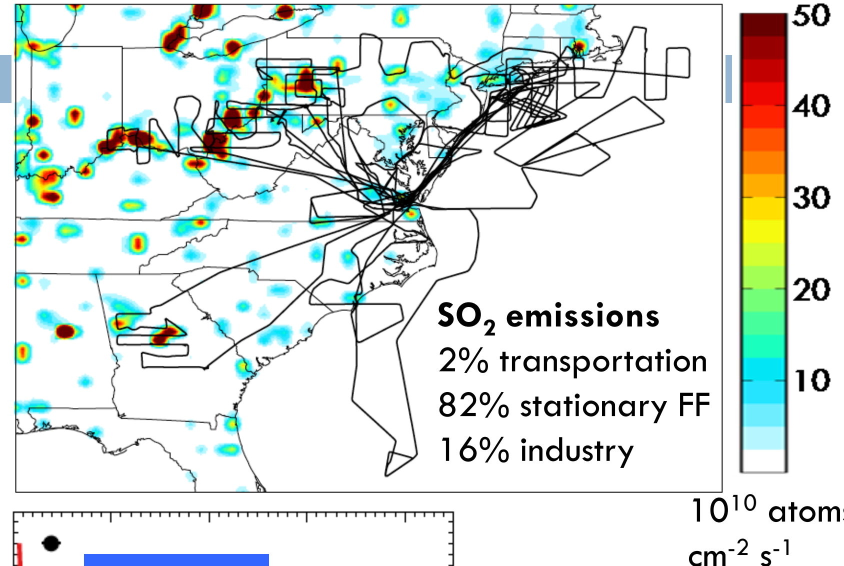
- WINTER campaign objectives were more regional in scope, not with a specific urban focus

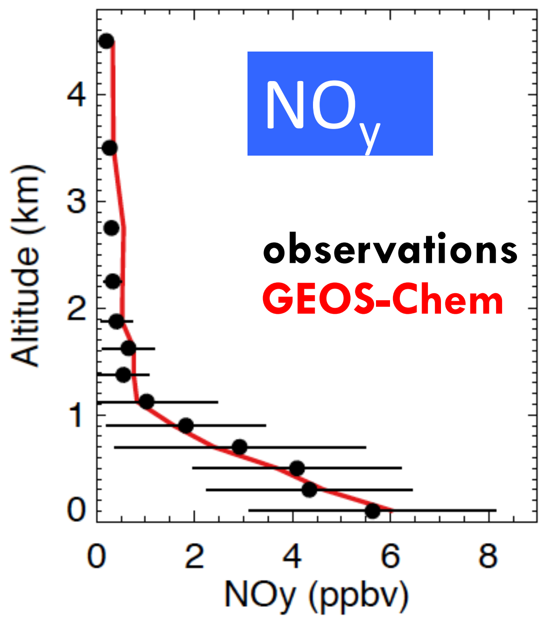
- Mix of emissions, especially role of agriculture, likely quite different for some AQUARIOUS regions

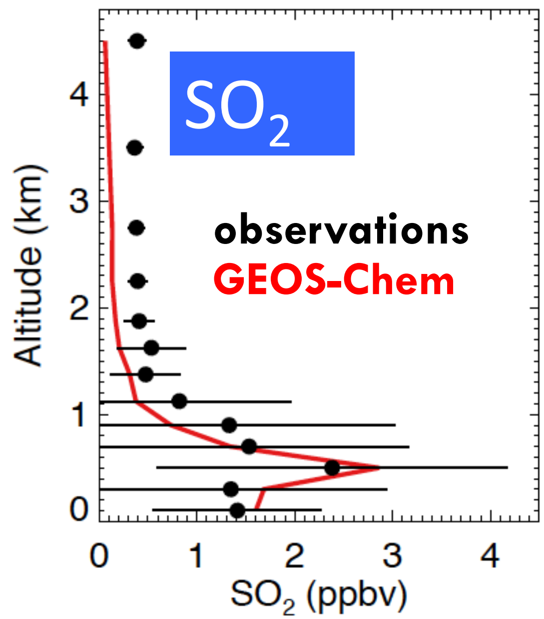
Emissions

Anthropogenic NO_x and SO_2 Emissions







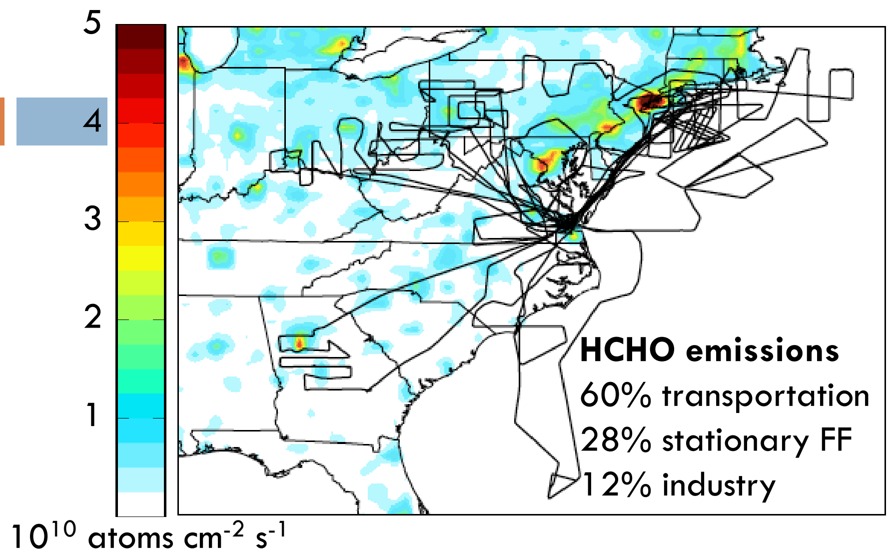


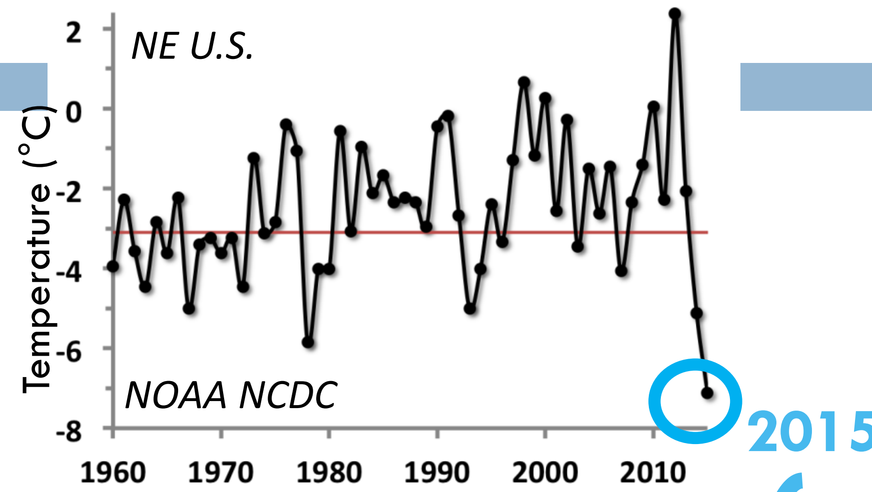
Vertical profiles over land

Jaeglé, et al JGR 2018

→ Reasonable emissions: model within 10% of observations

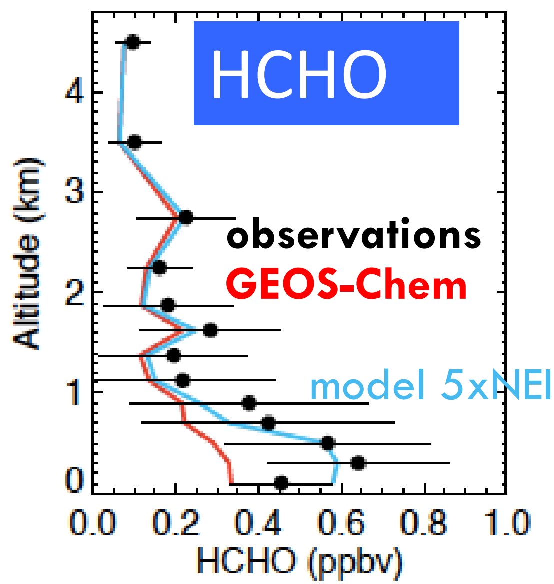
Formaldehyde (HCHO) and NH_3 emissions

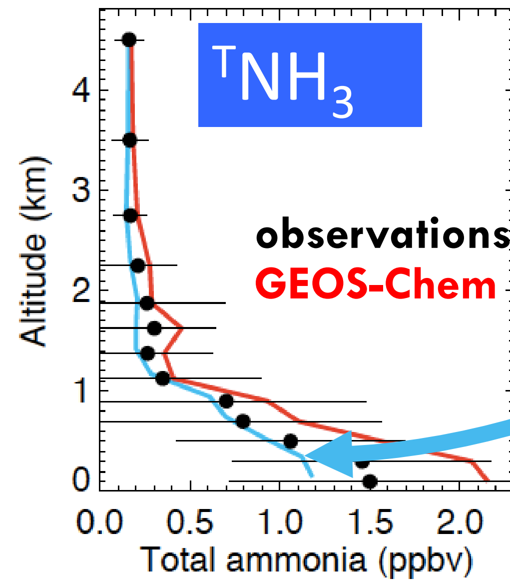




Underestimate of <u>cold-start</u> OVOC emissions?

→ Increase NEI HCHO by x5





Rescale livestock $E_{NH3} \sim 10^{0.48T}$
[Aneja et al., 2000]

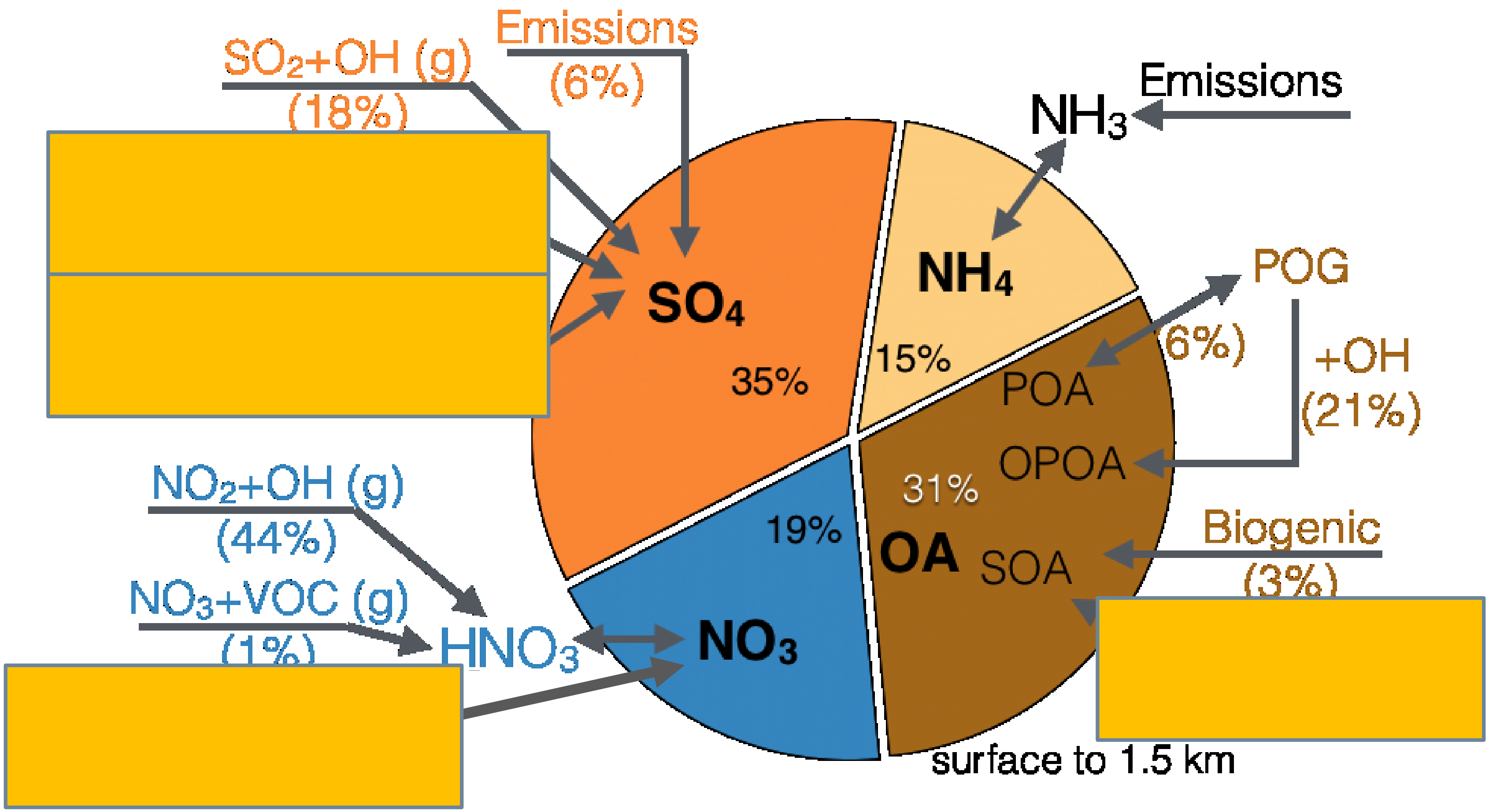
model with 2015 T

→ Default HCHO too low by 50%, $^{T}NH_3$ too high by 50%

Secondary Aerosol Formation

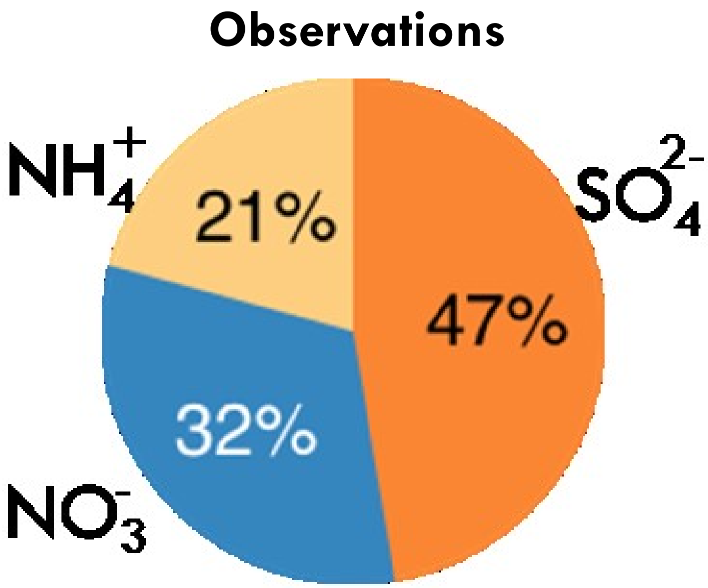
>95% of PM1 mass formed in the atmosphere

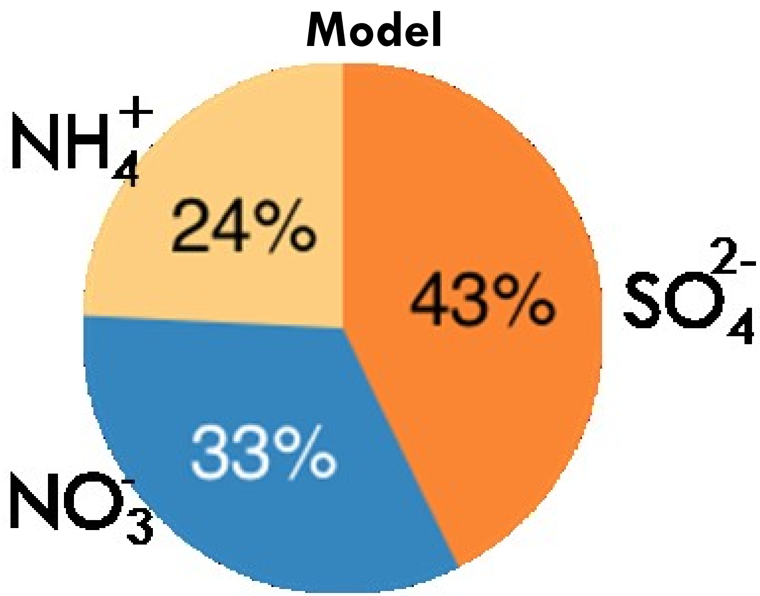
35% from multiphase aerosol/cloud chemistry

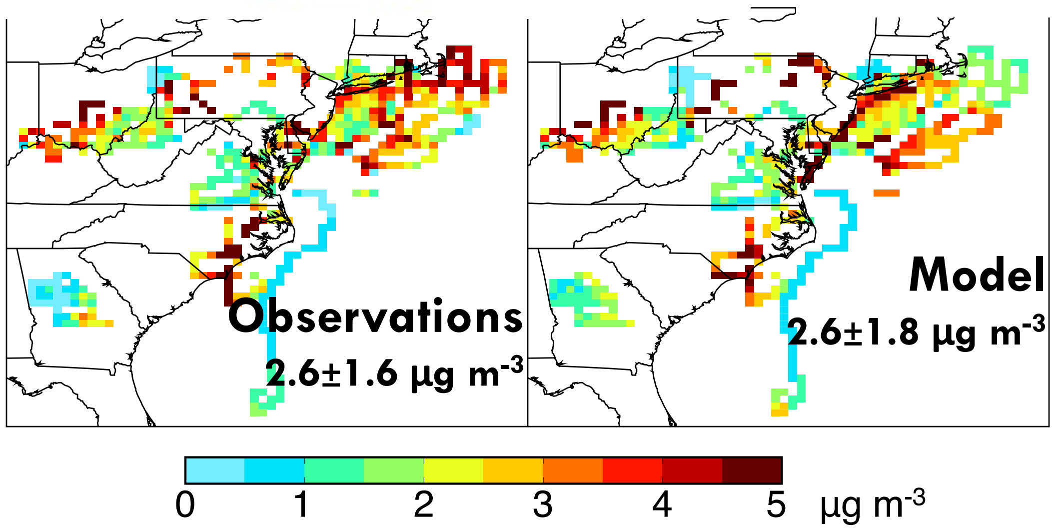


Shah et al PNAS 2018

Inorganic aerosol components



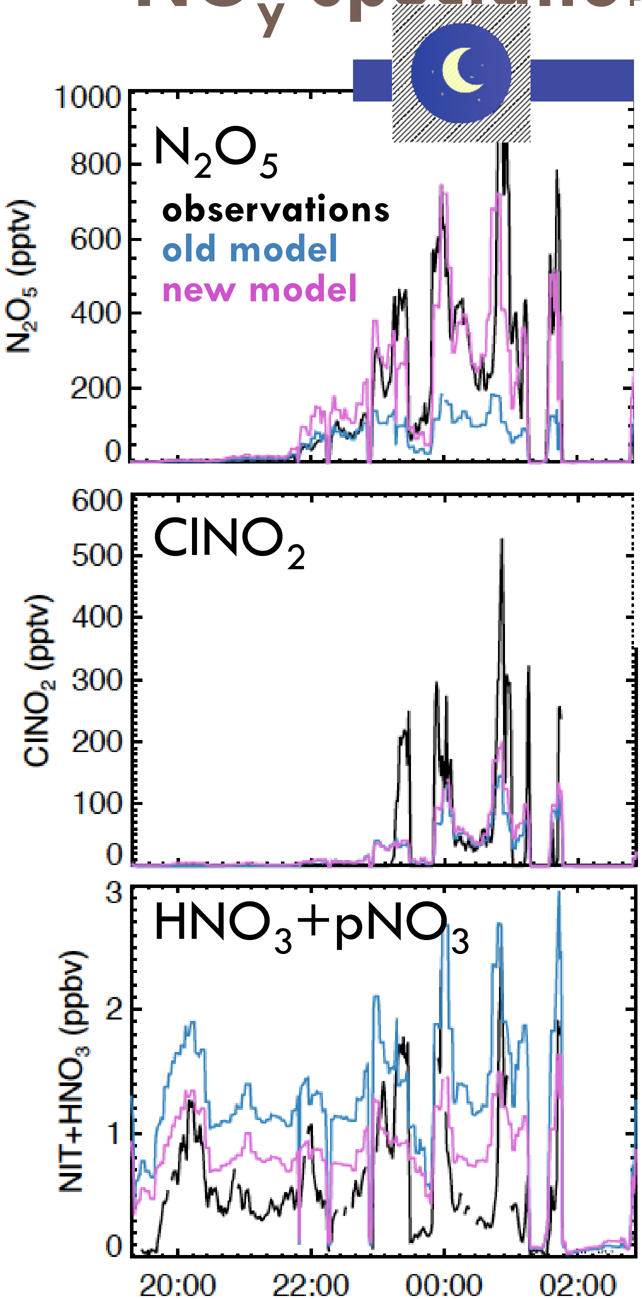


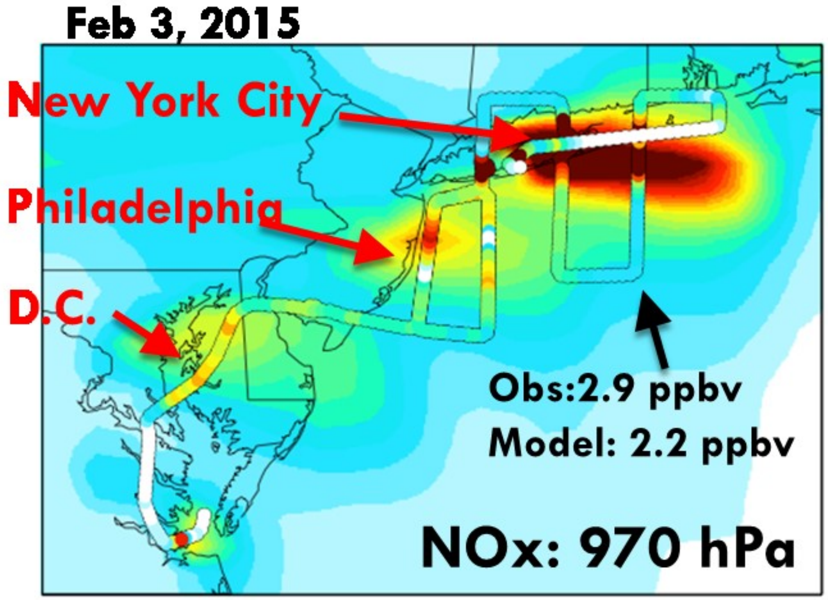


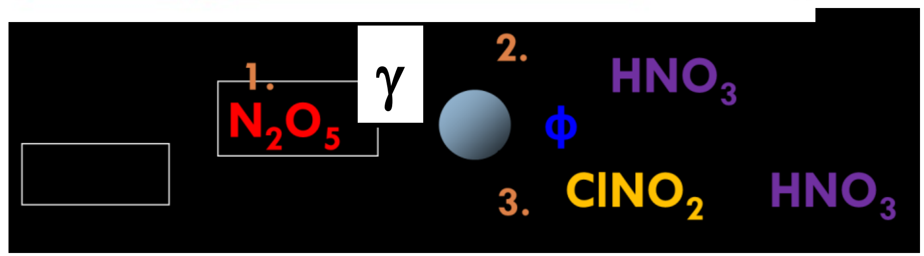
Normalized Mean Bias: 5%
FAC2: 78%
(0.5 < model/obs < 2)

Shah et al PNAS 2018

NO_y speciation constrains key parameters

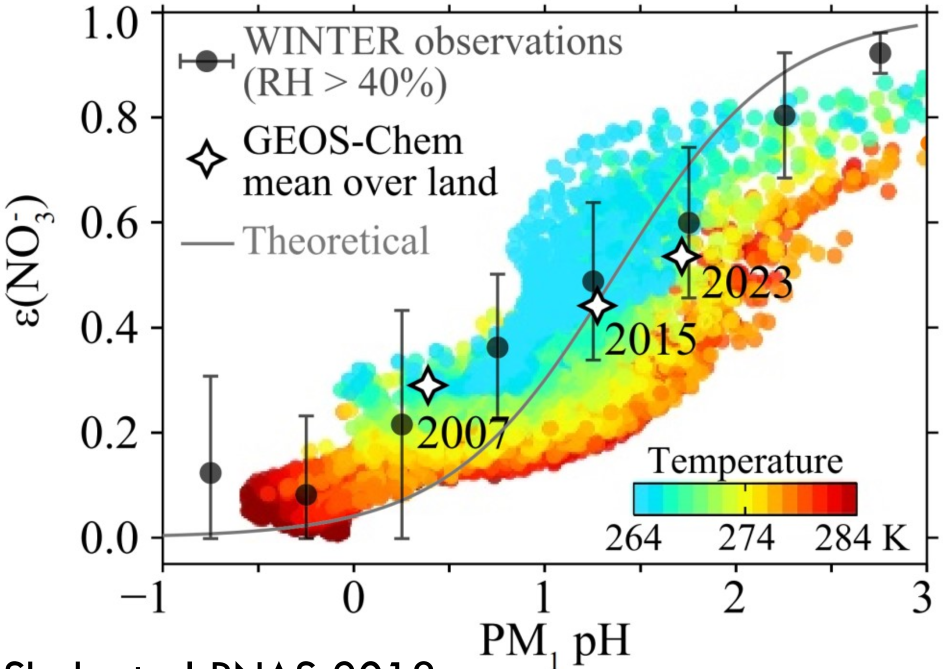




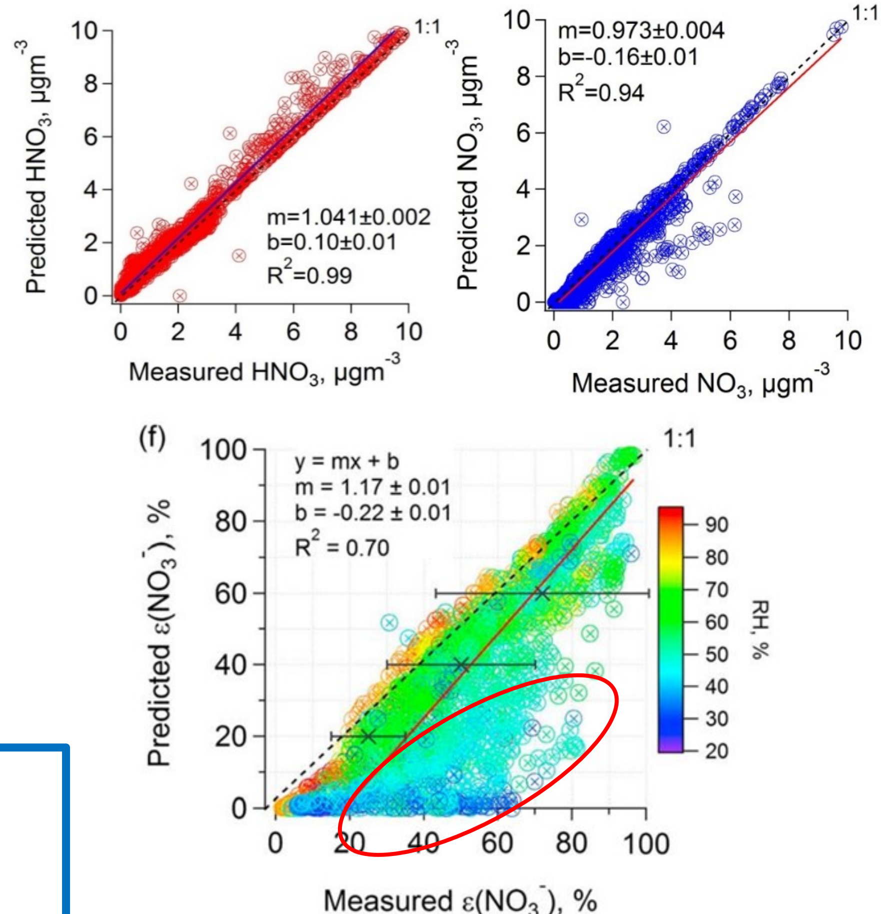


- Updates to $\gamma(N_2O_5)$ improve modeled N_2O_5
- $^TNO_3$ also improved, remaining difference related to dry deposition of HNO_3
- Jaeglé et al, JGR 2018; McDuffy et al JGR, 2018

Nitrate partitioning



Shah et al PNAS 2018

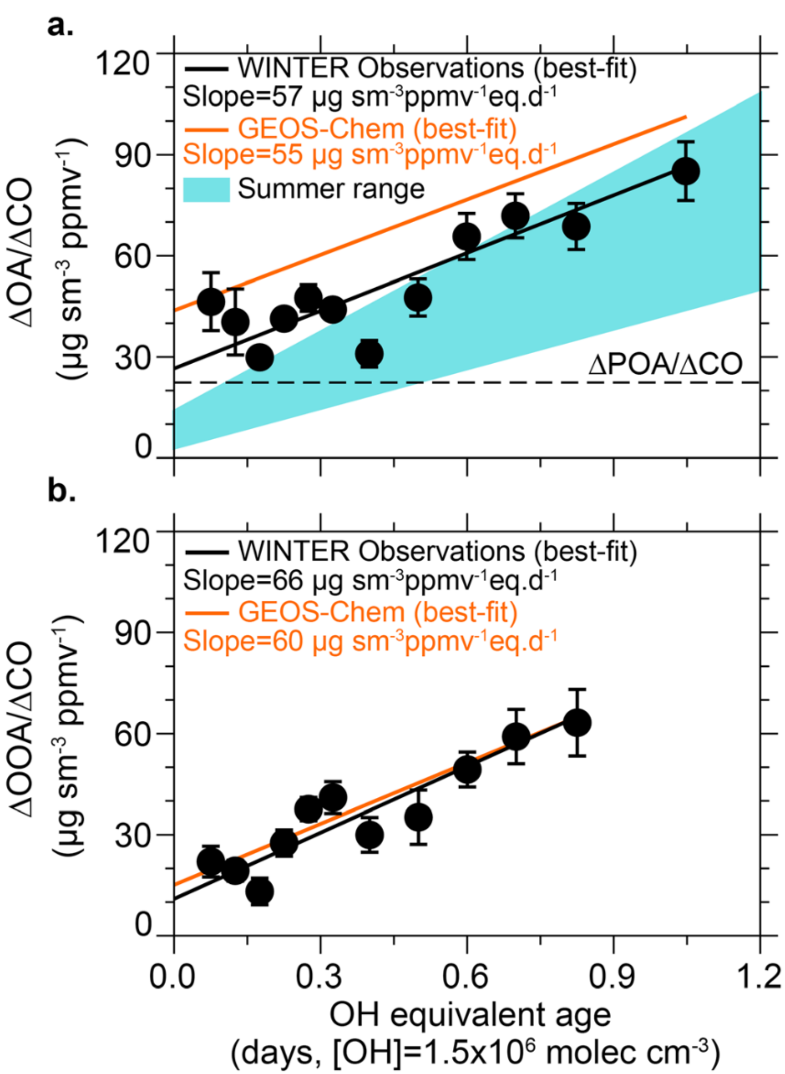


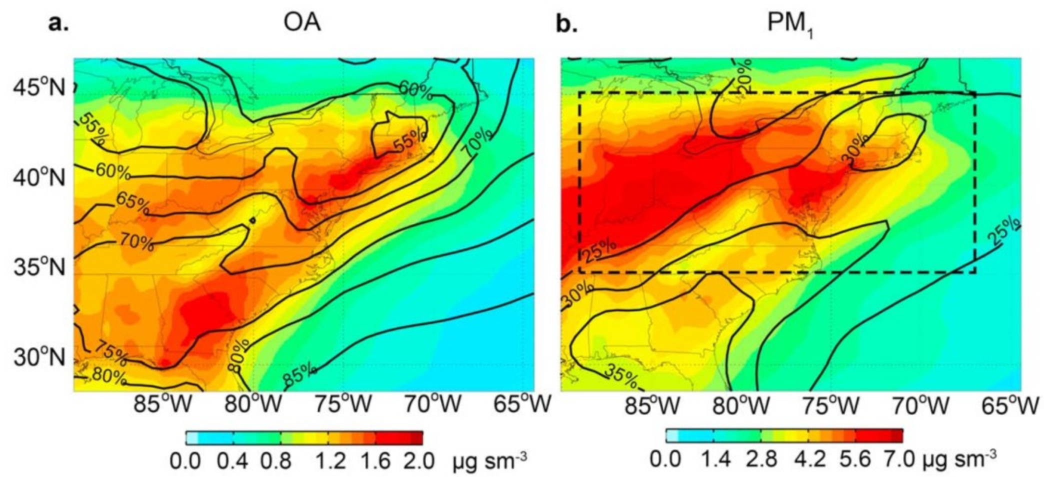
Guo et al JGR 2016

Reasonable agreement between observed and predicted nitrate partitioning during WINTER

How does this agreement change at the extremes (very low temperature, low RH, very high NH_3)?

Ubiquitous anthropogenic SOA formation



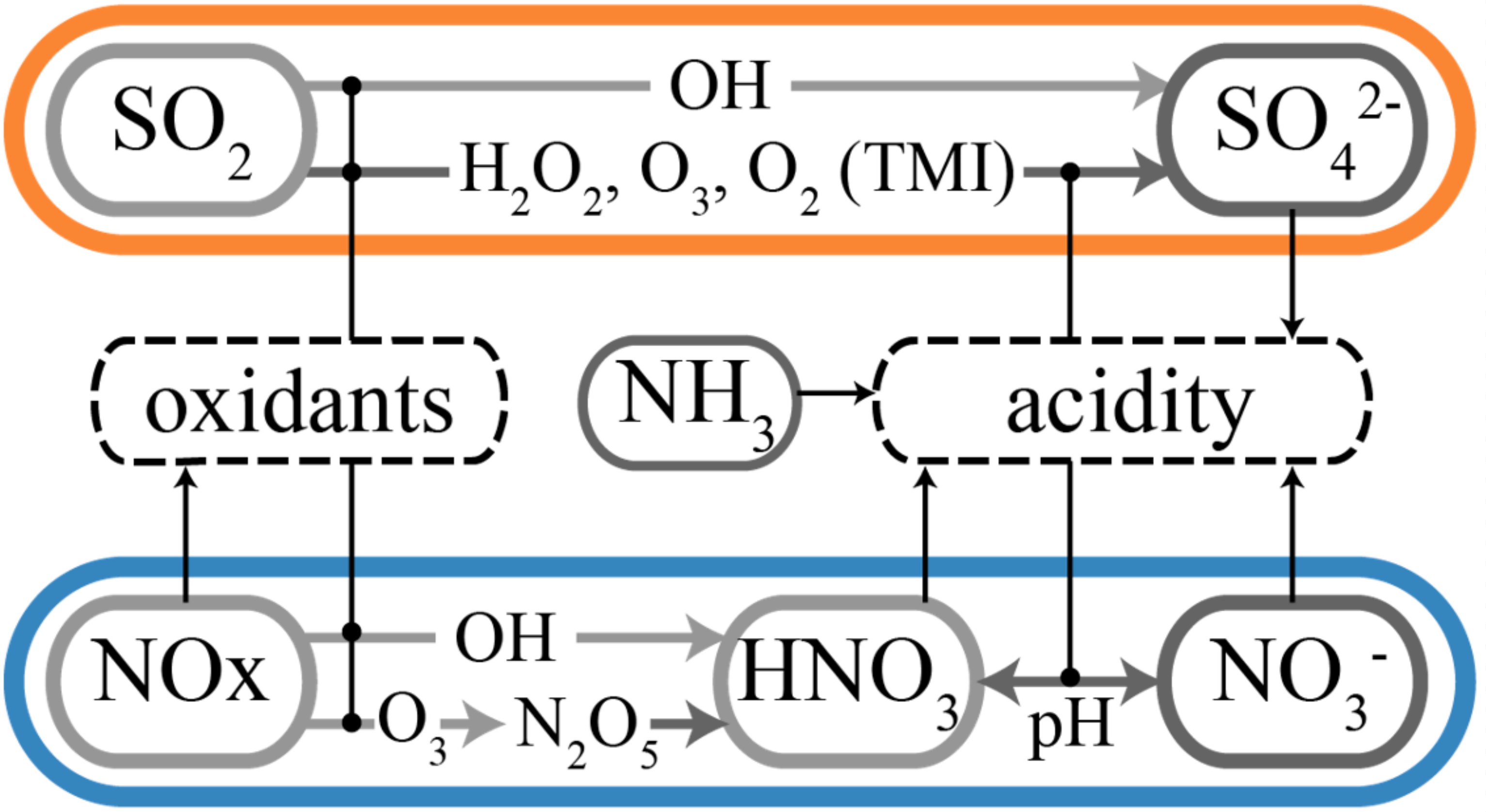


What are the important VOC precursors (and the sources thereof)?

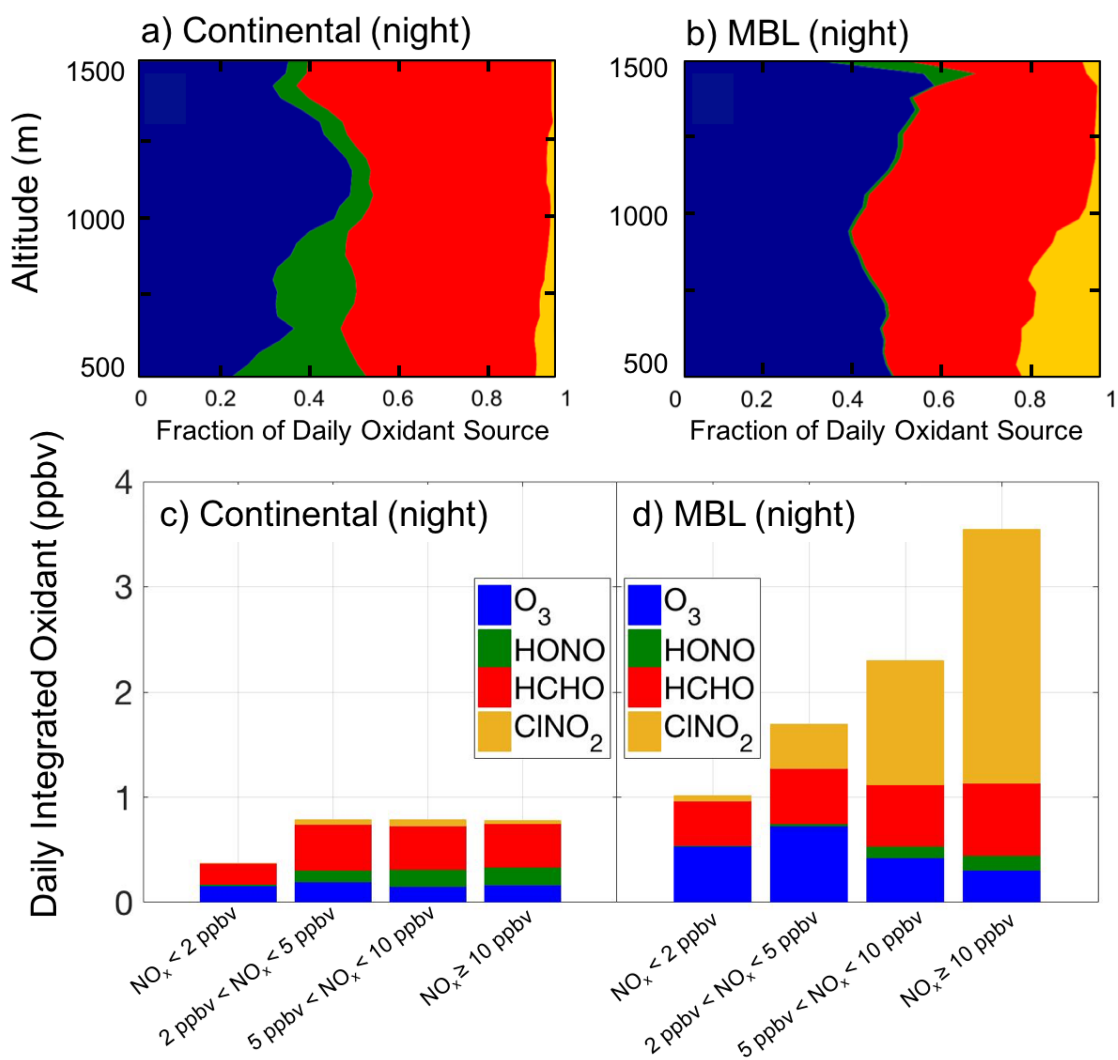
Can laboratory parameterizations of SOA formation from these precursors explain observed SOA?

Shah et al, GRL 2019
Schroeder et al, JGR 2018

Chemical processing



WINTER primary radical sources



>70% of primary radical source from HCHO, $ClNO_2$, and HONO

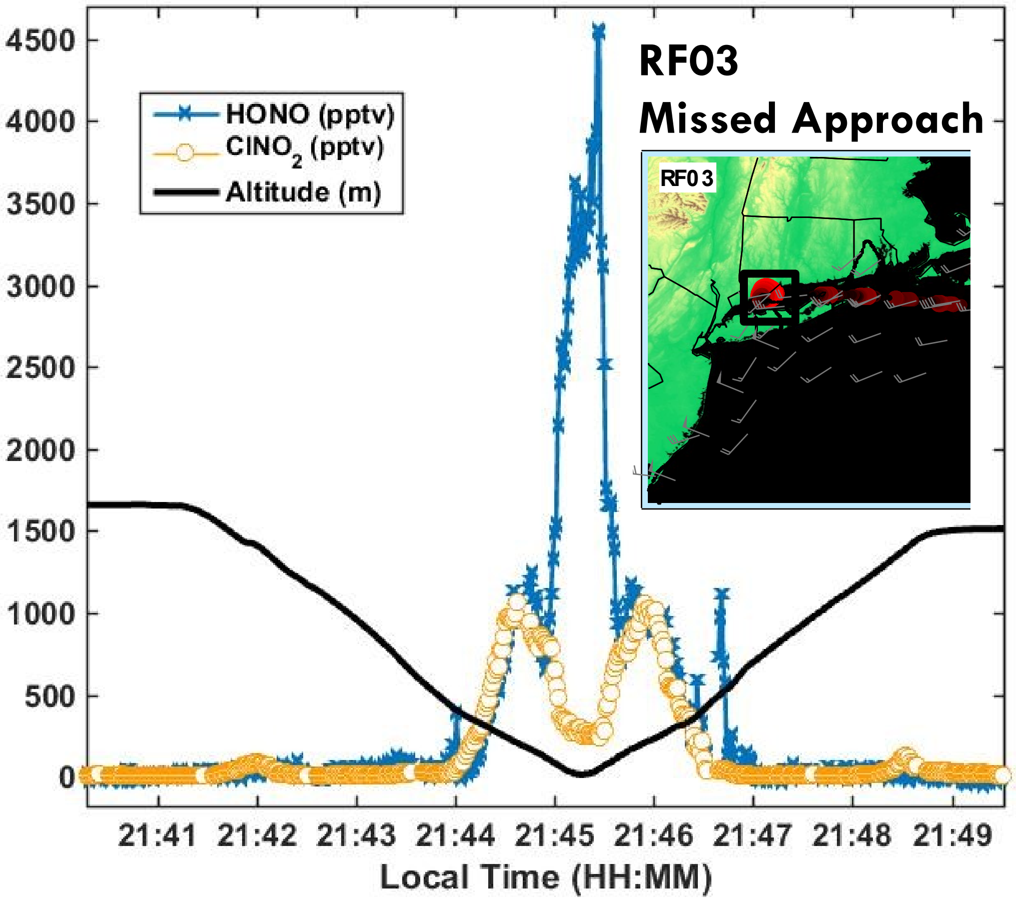
What drives HCHO formation? Are other RCHO important?

Will there be significant Chloride chemistry in AQUARIUS regions?

What are HONO vertical profiles during strong inversions and can these help elucidate sources?

Haskins et al, submitted to GRL 2019

WINTER primary radical sources



>70% of primary radical source from HCHO, $ClNO_2$, and HONO

What drives HCHO formation? Are other RCHO important?

Will there be significant Chloride chemistry in AQUARIUS regions?

What are HONO vertical profiles during strong inversions and can these help elucidate sources?

Haskins et al, submitted to GRL 2019

Some concluding thoughts
RE: AQUARIUS vs WINTER

- Couple potentially stronger photochemistry with stagnation/inversions, then based on WINTER expect,
  - → more localized but mostly secondary pollutants
  - → increased role for multi-phase processes

- Do we understand chemistry of "extremes" (high NH_3, high aerosol / fog surface area, high NO_x)

- How best can observations and models be obtained / utilized in such events to test hypotheses about emissions and chemistry?