Identification of Source Regions of High NOx During ACCLIP

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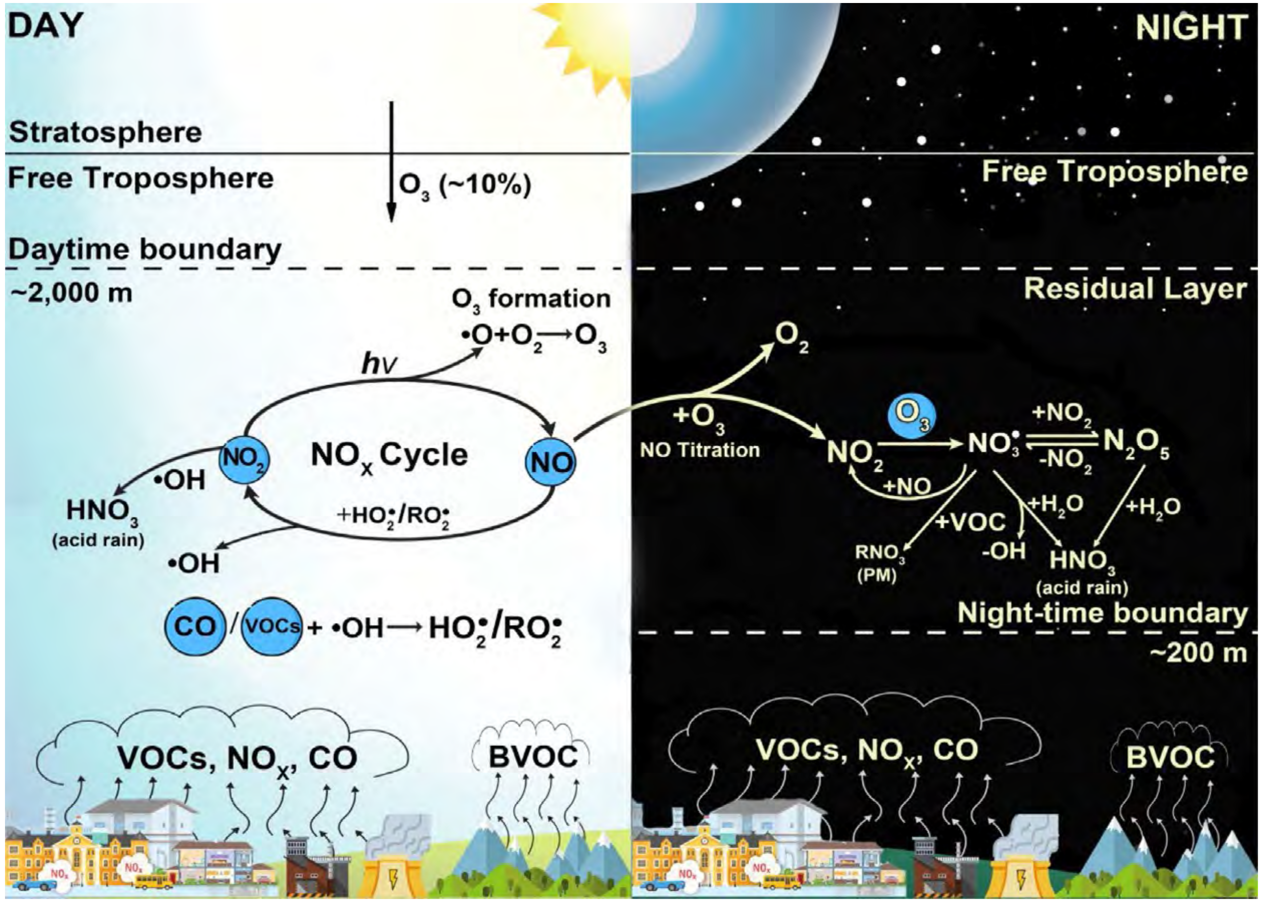
Overview – Atmospheric Nitrogen

1. Diurnal cycle

2. Daytime: NO and NO2 cycle back and fourth, producing O3 in the process.

3. Temperature dependent
   a) Slower at lower temperatures
      i. Shorter lifetime near surface (hours).
      ii. Longer lifetime in the free troposphere (days).

Makes Linking Observations to Sources a Challenge



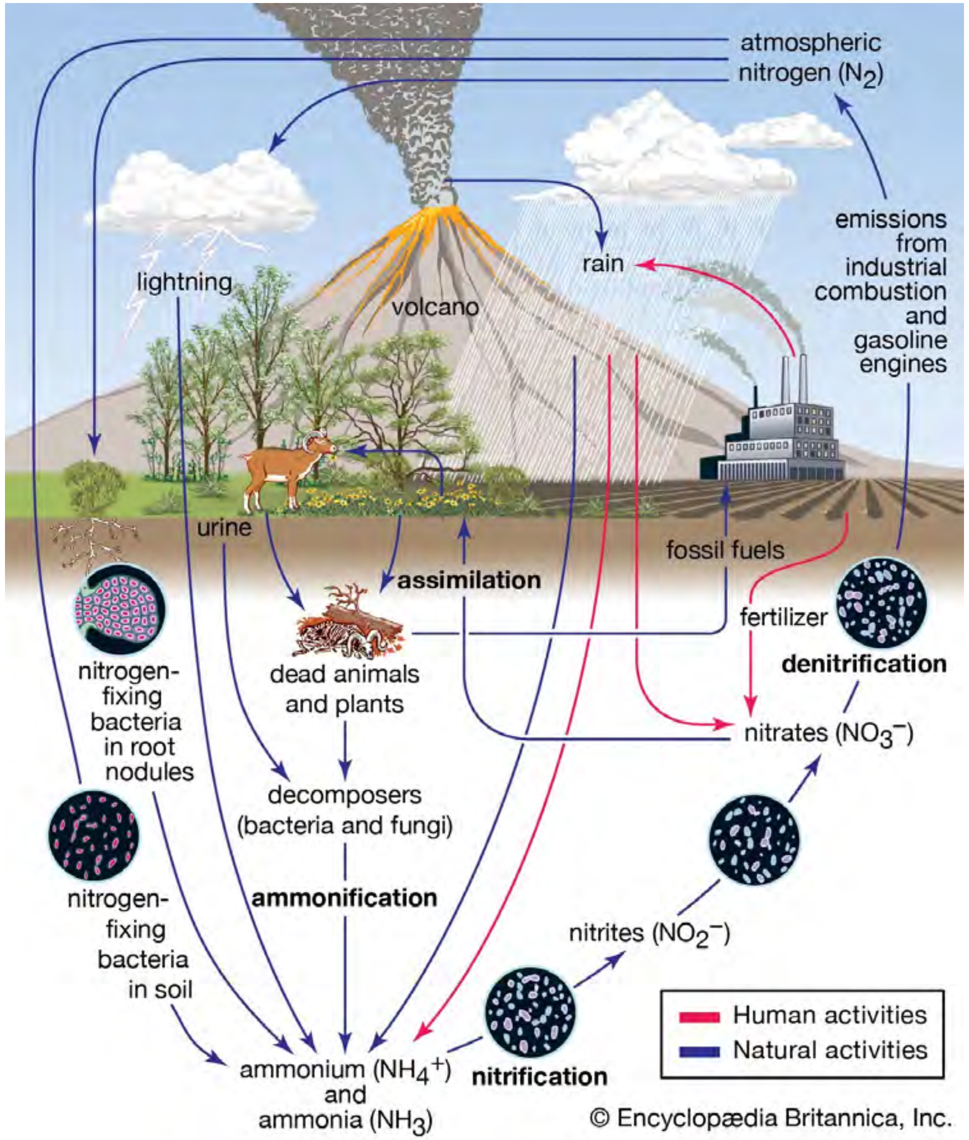
Overview – NOx Sources

Potential Sources of Upper Tropospheric NOx in ACCLIP Measurements:

- Stratospheric Intrusions
  - Not a Common Occurrence in the ACCLIP domain.
- Volcanoes
  - No major eruptions that affected observations.
- Wildfires/Biomass Burning
  - Estimates of 15%-20% of global NOx budget[1].
- Lightning
  - Estimated 70% NOx contribution in subtropical free trop[2].
  - Large uncertainty in modeled lightning NOx production.
- Anthropogenic
  - Industrial sources in large cities
- Aircraft Exhaust
  - ACCLIP flew in very busy aircraft corridors and often on busy airways.

[1]Denman et al., 2007
[2] Pickering et al., 2016

Nitrogen Cycle – UT NOx Lifetime ~ Several Days



From https://www.britannica.com/science/nitrogen-cycle

Method – General Flow

<u>GOAL</u>: Investigate the source regions and source types of high NOx events observed during ACCLIP.

Segregate High NOx
- GV Data Only
- WB57 data will eventually be ingested

Identify Source Regions
- Lagrangian Back Trajectories
- Convective Cloud Tops

Collect information at Source regions
- Satellite Data
- Lightning Data
- Reverse Domain Filling

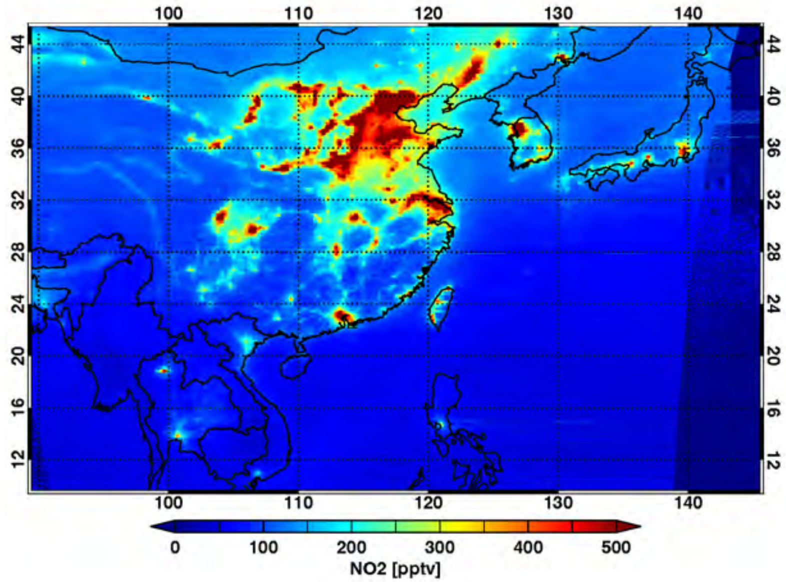
Analyze Collected Information
- Time Series
- Convective Influence
- Scatter Plots
- Etc.

Exploratory Stages

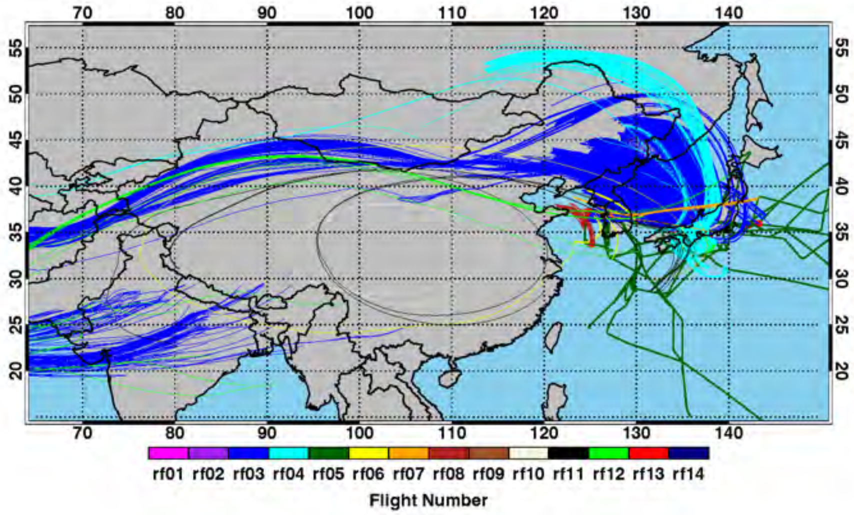
Narrow the gap between model and observations

Data & Tools

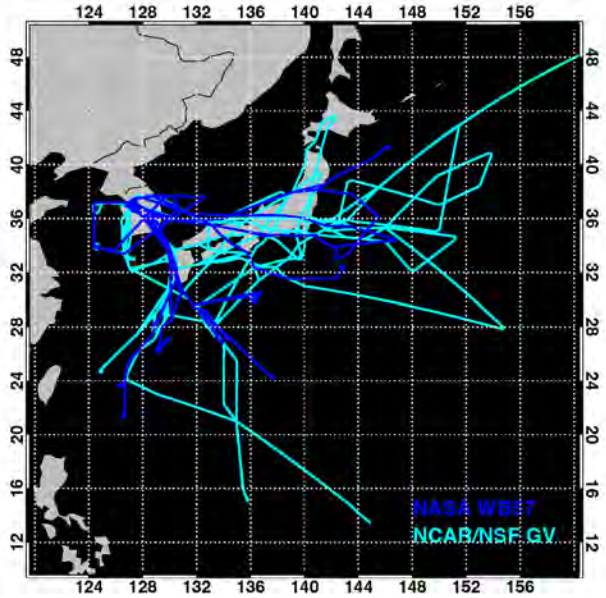
August 2022 mean GEMS Tropospheric Column NO_2



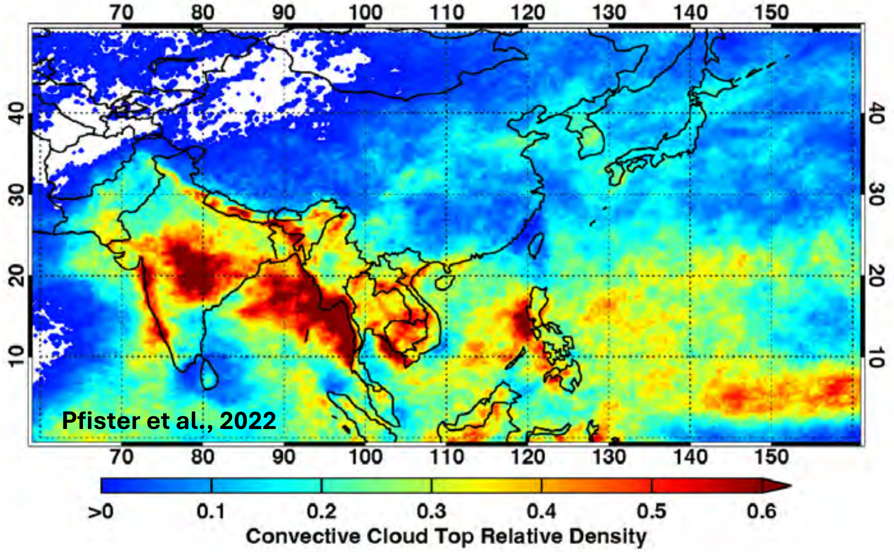
Lagrangian Kinematic Back Trajectories (TRAJ3D)



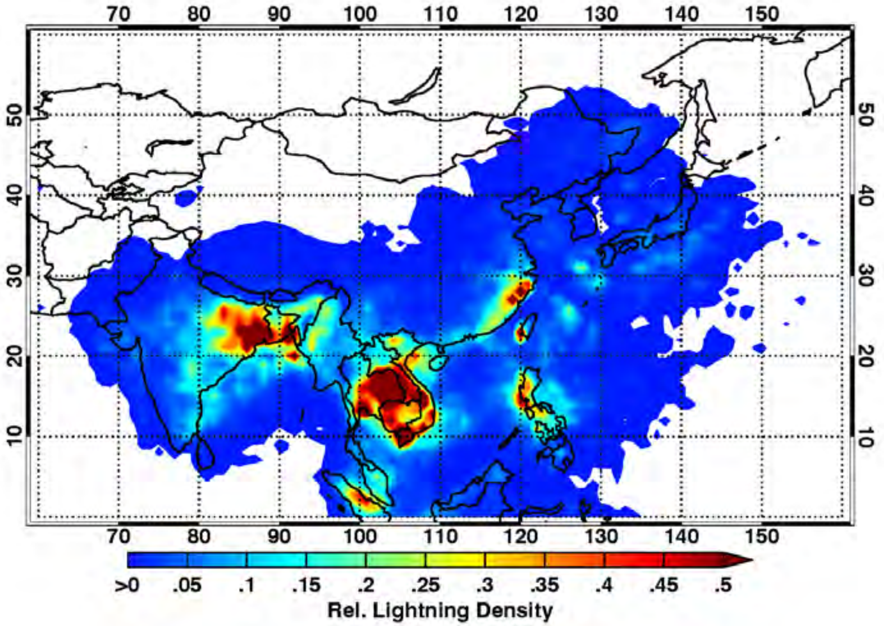
ACCLIP Aircraft Data



Convective Cloud Tops > 10 km



Earth Networks Lighting > 10 km (Intracloud)

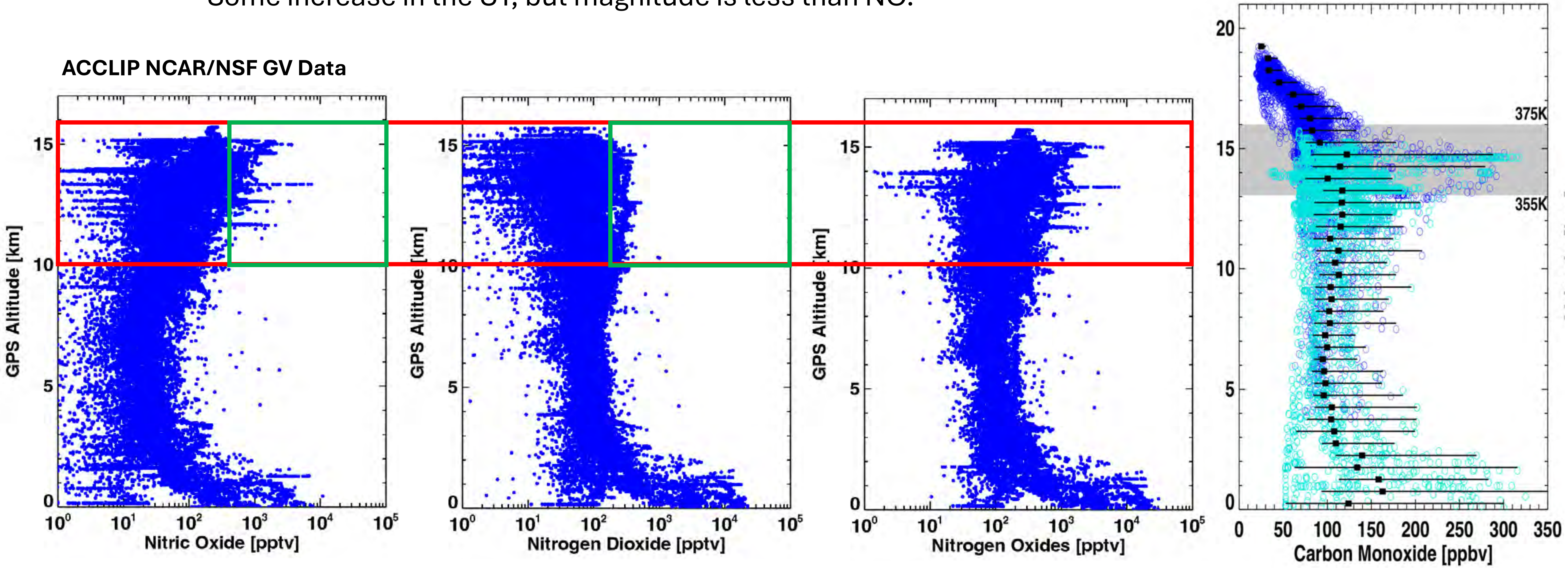


NOx - What Did We Observe

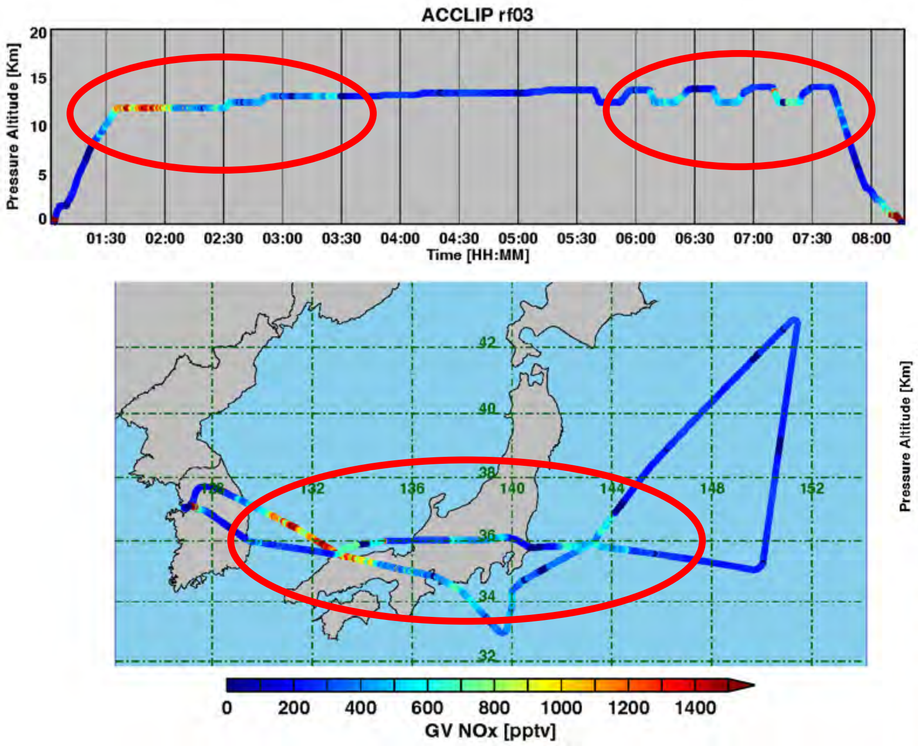
- Nitric Oxide (NO) exhibits the same C-shape profile seen in other tracer profiles (i.e. CO).
- Nitrogen Dioxide (NO2) has more of an L-shape.
  - Some increase in the UT, but magnitude is less than NO.

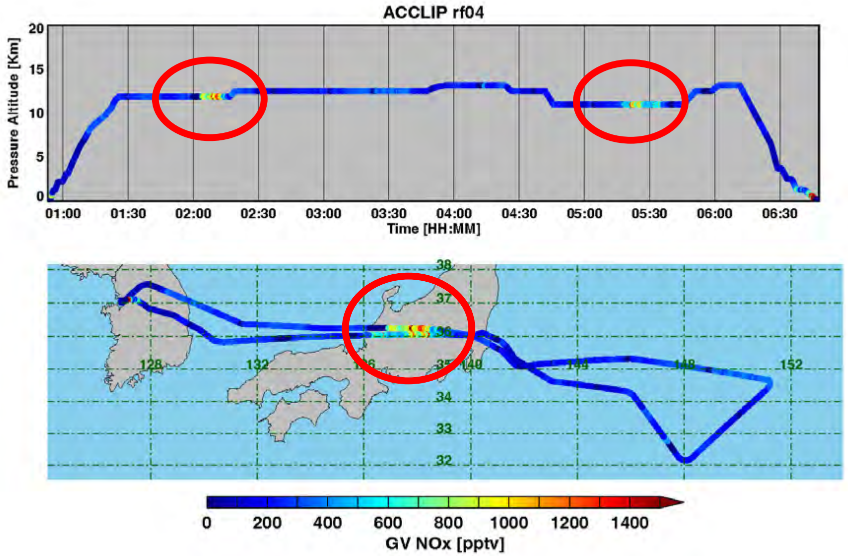
Pan et al., 2024

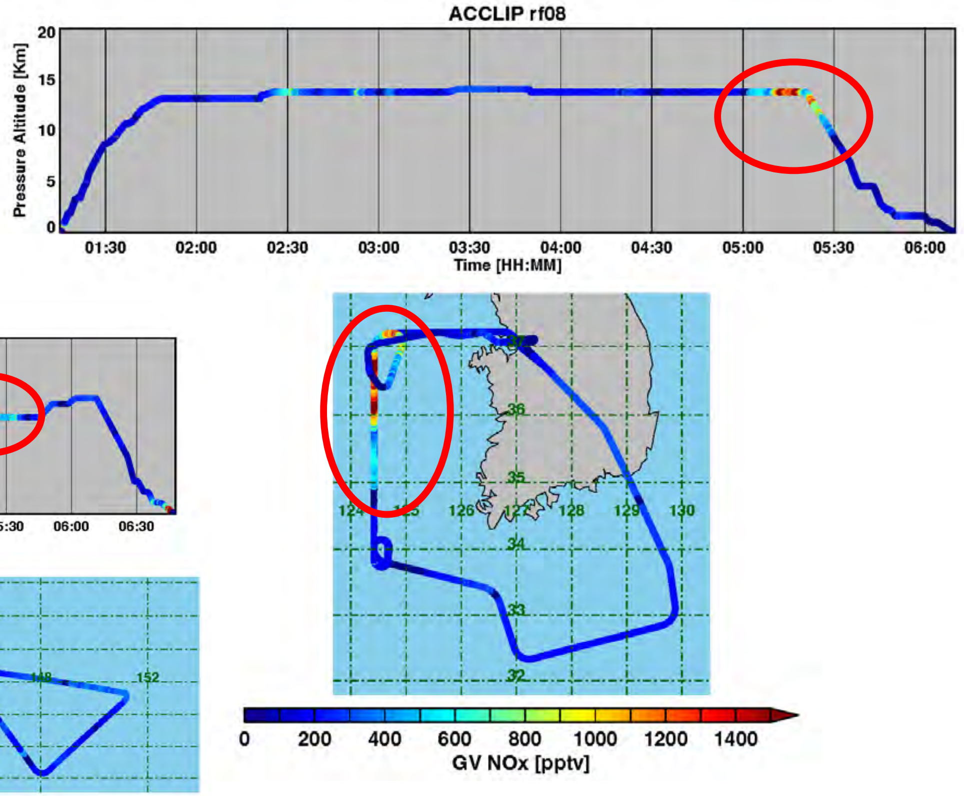
ACCLIP NCAR/NSF GV Data



NOx - What Did We Observe







High NOx: Mainly from three flights

1. RF03 – Aug 6th
2. RF04 – Aug 7th
3. RF08 – Aug 19th

Method - Segregate High NOx

Criteria:

1. NO: > 400 ppt
2. NO2: > 200 ppt
3. Alt: > 10 km

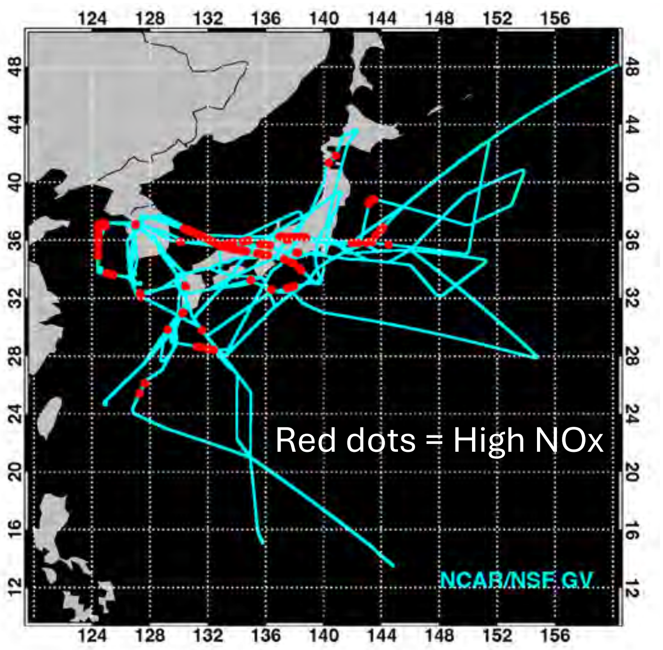
About 4000 GV 1Hz sampling rate data points to work with

Most from 3 GV flights:

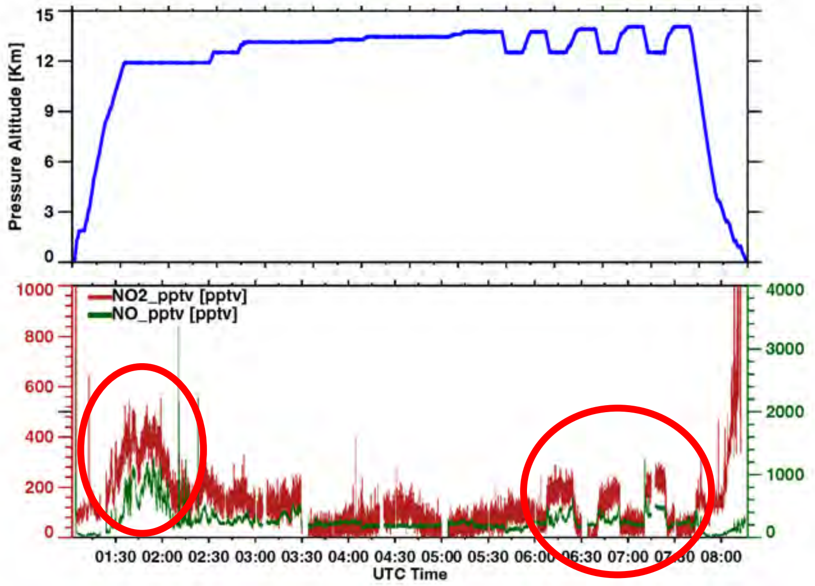
1. RF03
2. RF04
3. RF08

Areas Covered:

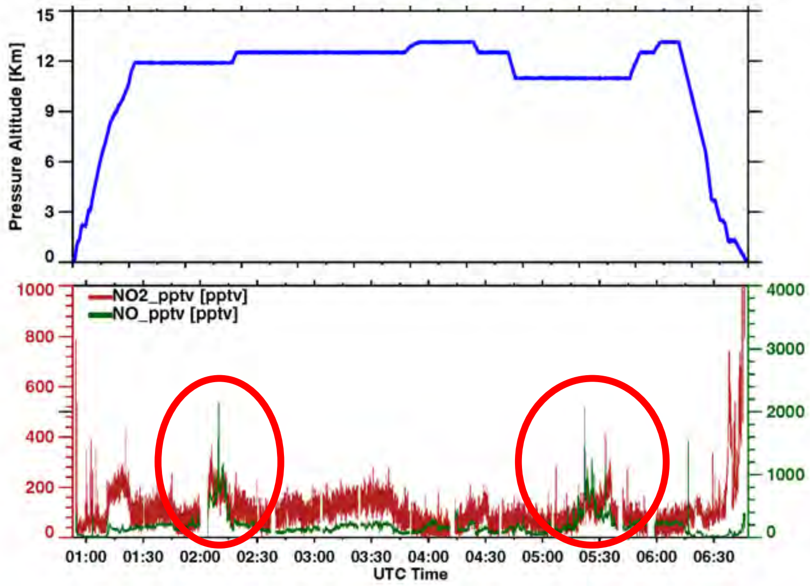
1. Japan
2. Yellow Sea
3. Sea of Japan
4. Pacific Ocean



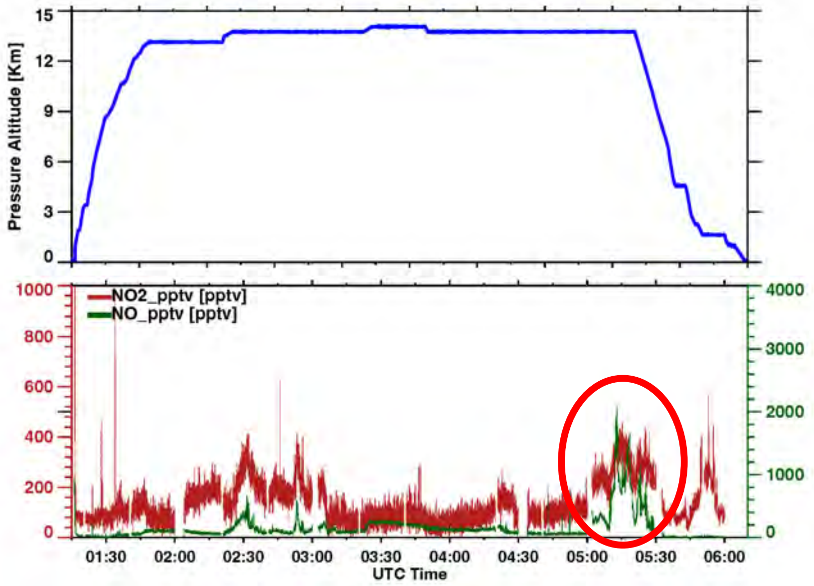
RF03 2022/08/06



RF04 2022/08/07



RF08 2022/08/19

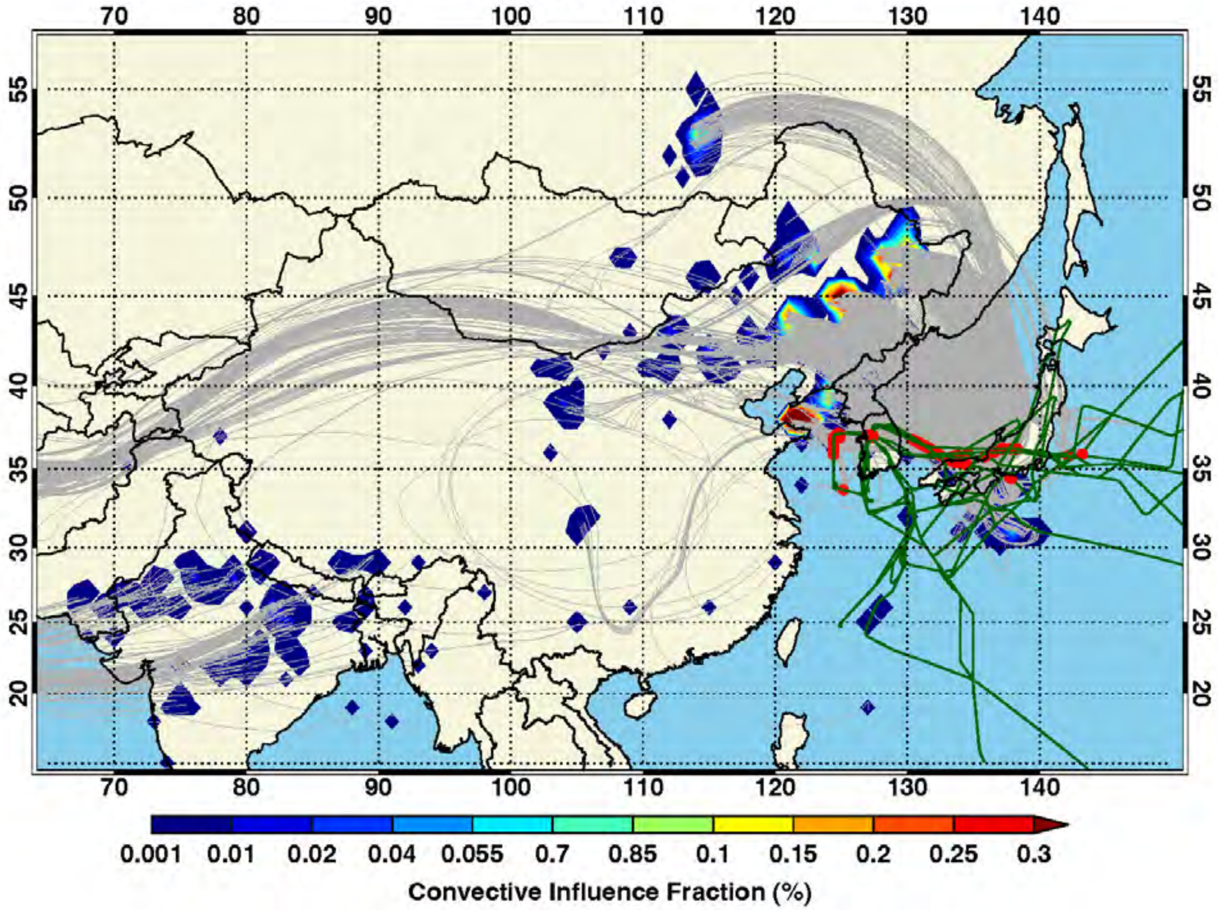


Method - Trace Sources via Lagrangian Trajectories

- Lagrangian kinematic back trajectories (TRAJ3D).
- Driven by hourly ERA5 wind fields.
- Initiated at each of the ~4000 tagged high NOx data points.
- Trajectories run backward for a maximum of 10 days.
- Terminated at convective cloud tops > 10 km.
- Trajectories that go the entire 10 days without a convective encounter are disregarded.

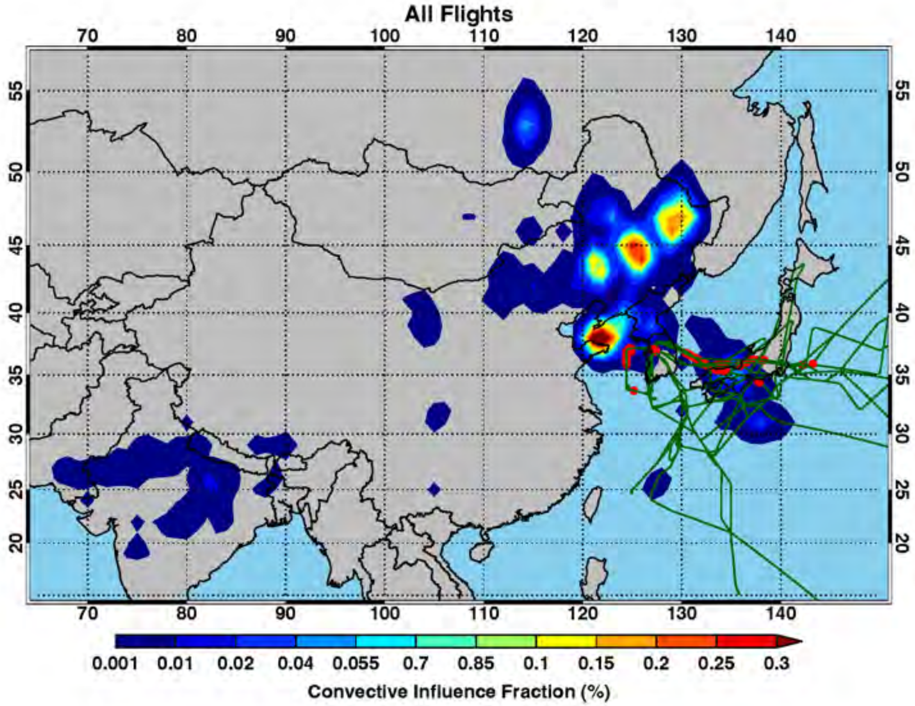
~95% of the trajectories terminated at a convective cloud within 10 days.

Convective influence fraction and trajectories from all flights



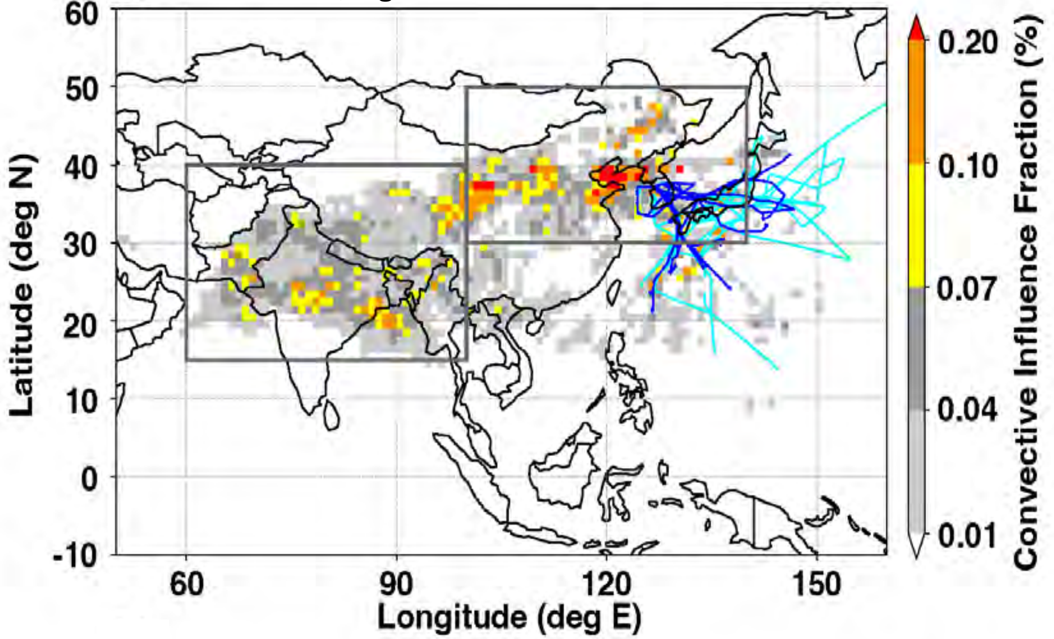
Results - Source Regions

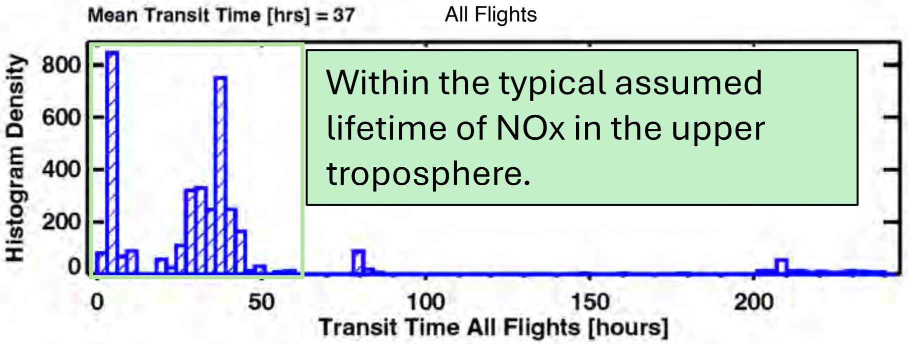
➢ Most of the high NOx originates from eastern China within a couple of days – good agreement with Pan et al. 2024 (with exception of RF04).



- *Green lines denote GV flight tracks.*
- *Red dots on aircraft tracks denote high NOx locations.*
- *Convective Influence Fraction is for trajectories initiated in red areas on the GV flight track.*

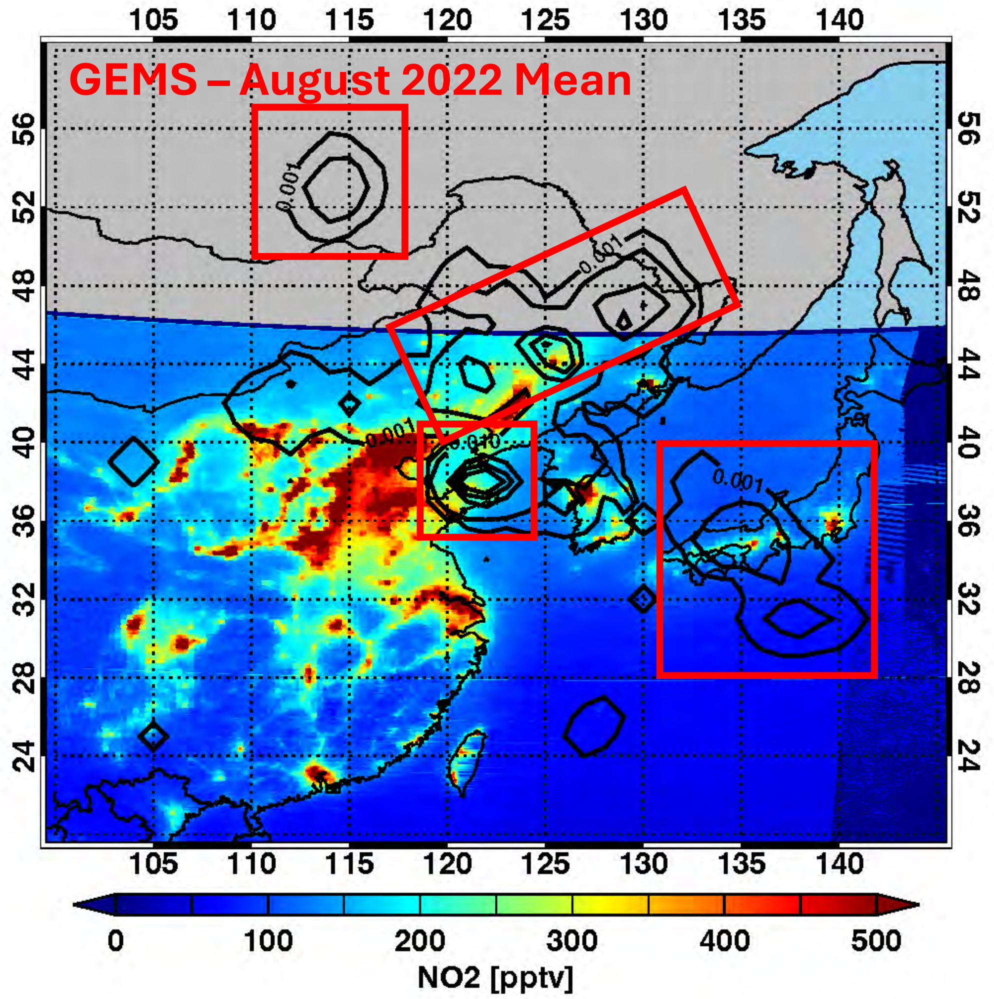
Pan et al. 2024, Figure 3

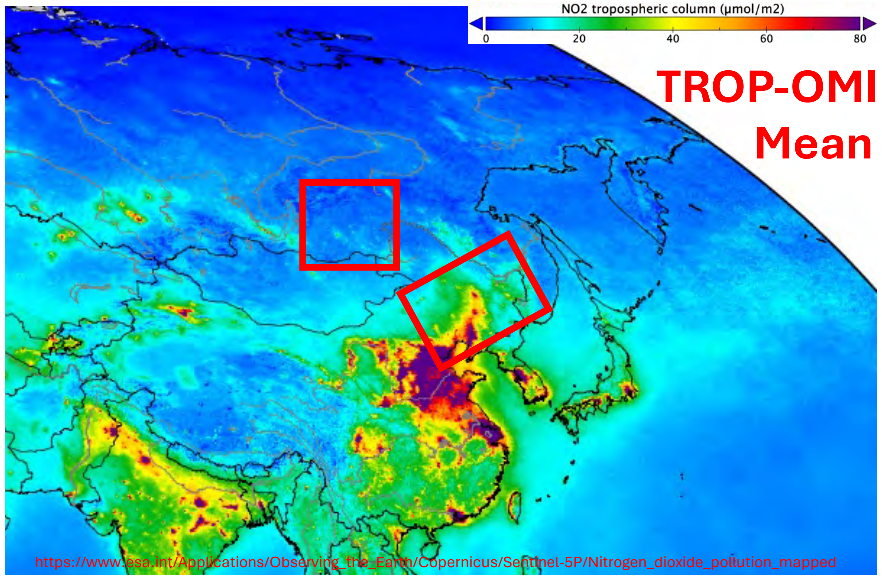


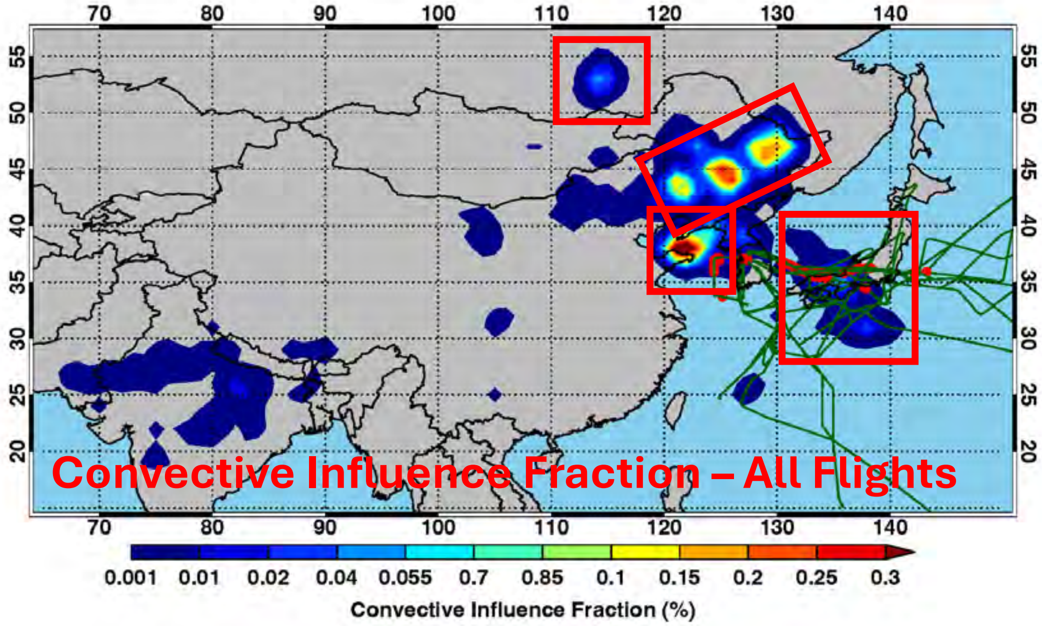


Results - Source Regions

Anthropogenic sources surely have at least some influence on observations though convective influence areas don't heavily intersect.

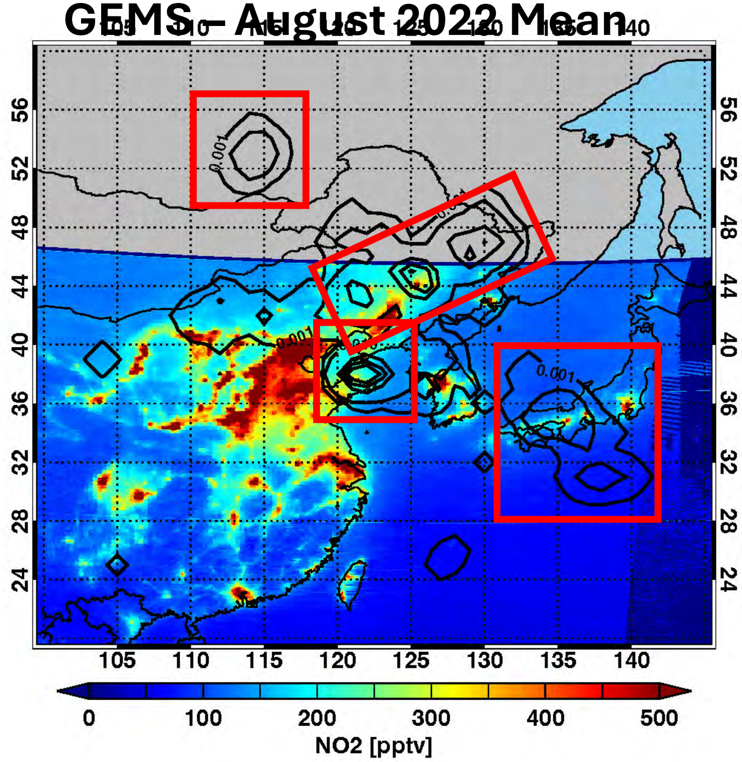






Results - Source Regions

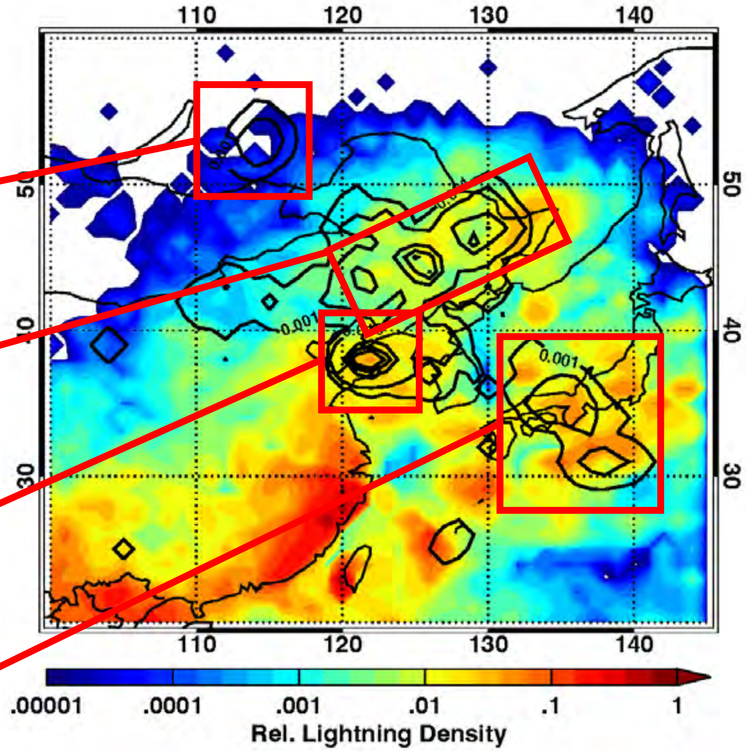
GEMS – August 2022 Mean



Lighting data adds more information

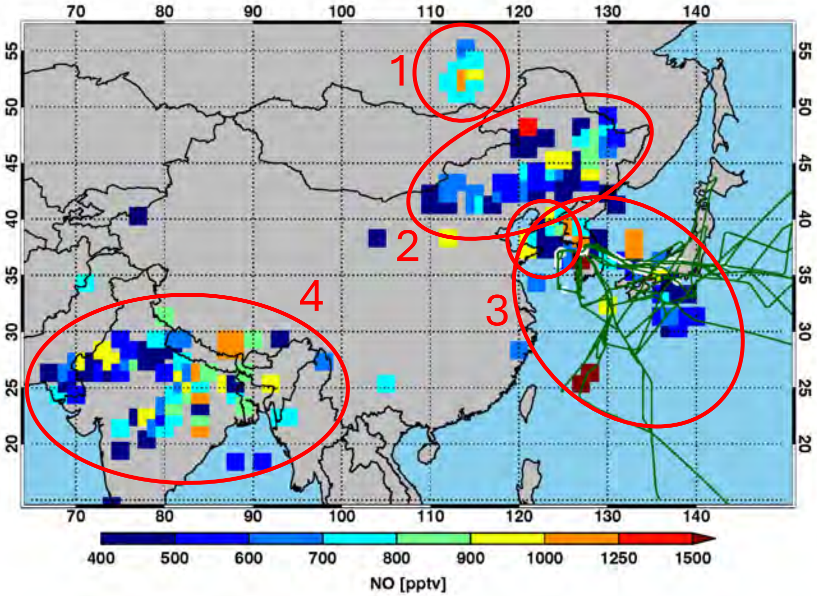
- Upper left box not associated with much lightning.
- Increased lightning activity on right area of upper middle box.
- More lighting in lower middle box.
- Bottom right box associated with most lighting compared to other contoured areas.

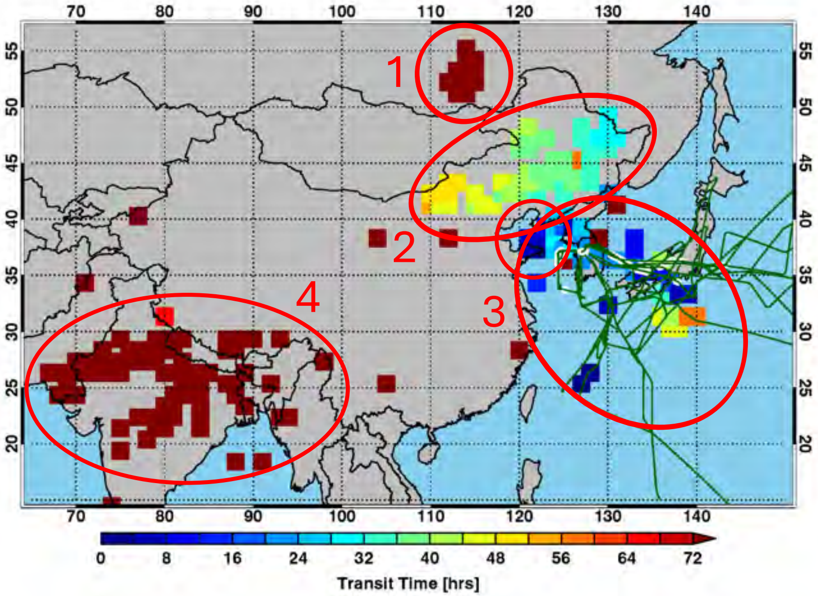
Earth Networks Lightning JAS Mean

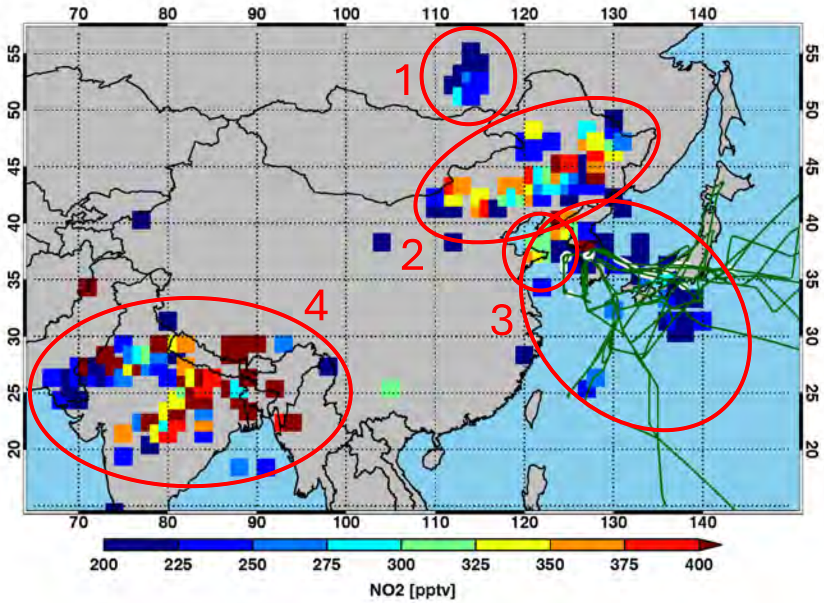


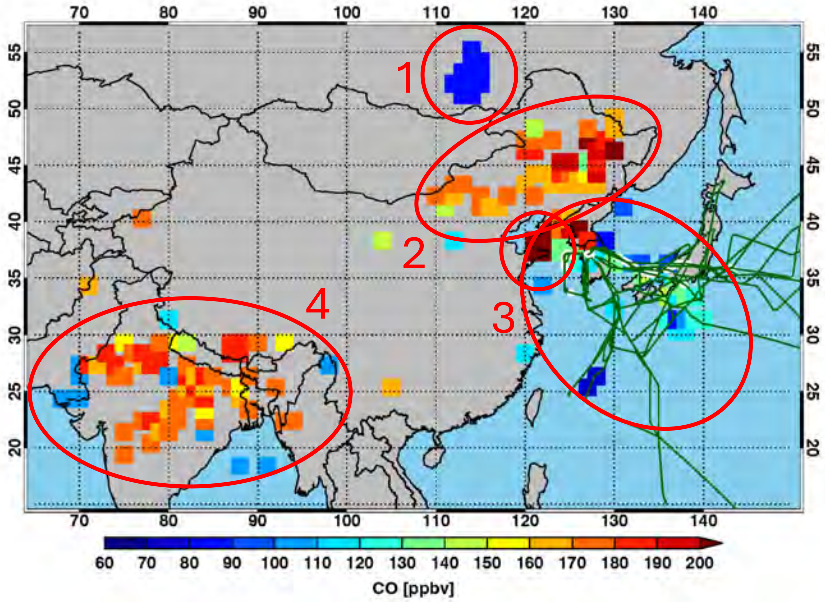
Results - Source Regions - RDF

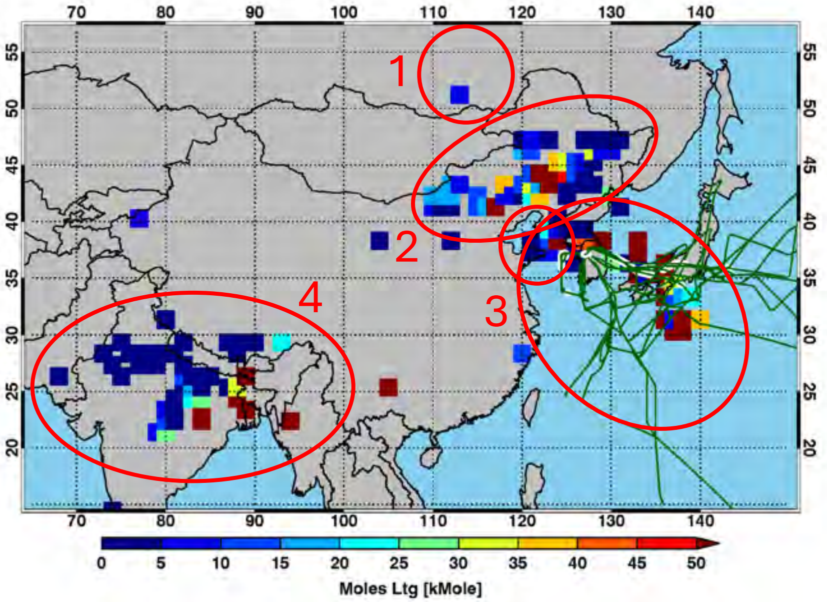
1. High NO but outside lightning window.
   a) Trajectories mapped wrong location?
2. High CO, high NOx, lots of lightning.
   a) Some industrial influence?
3. Lower CO/NO2. Shortest transit time.
   a) Mostly lightning influence?
4. Long transit time & small # of trajectories.
   a) Disregard?





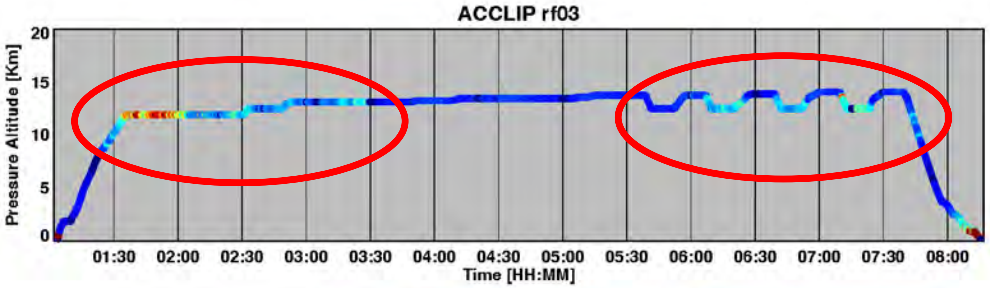


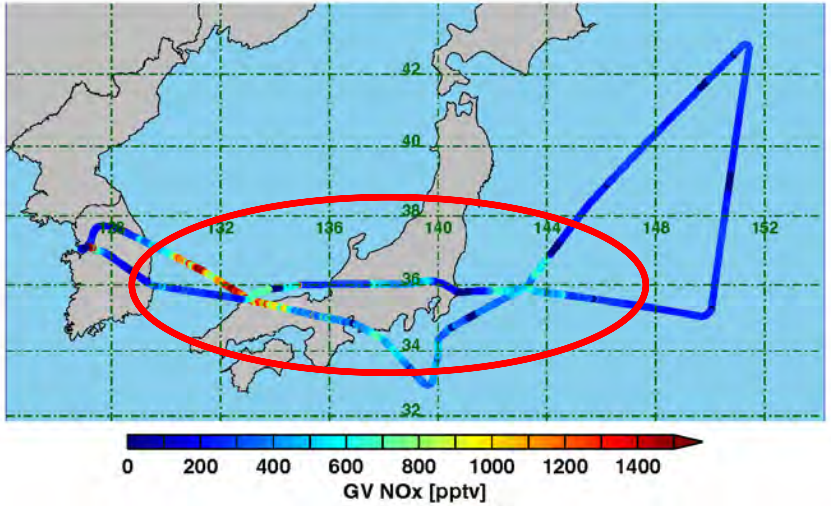




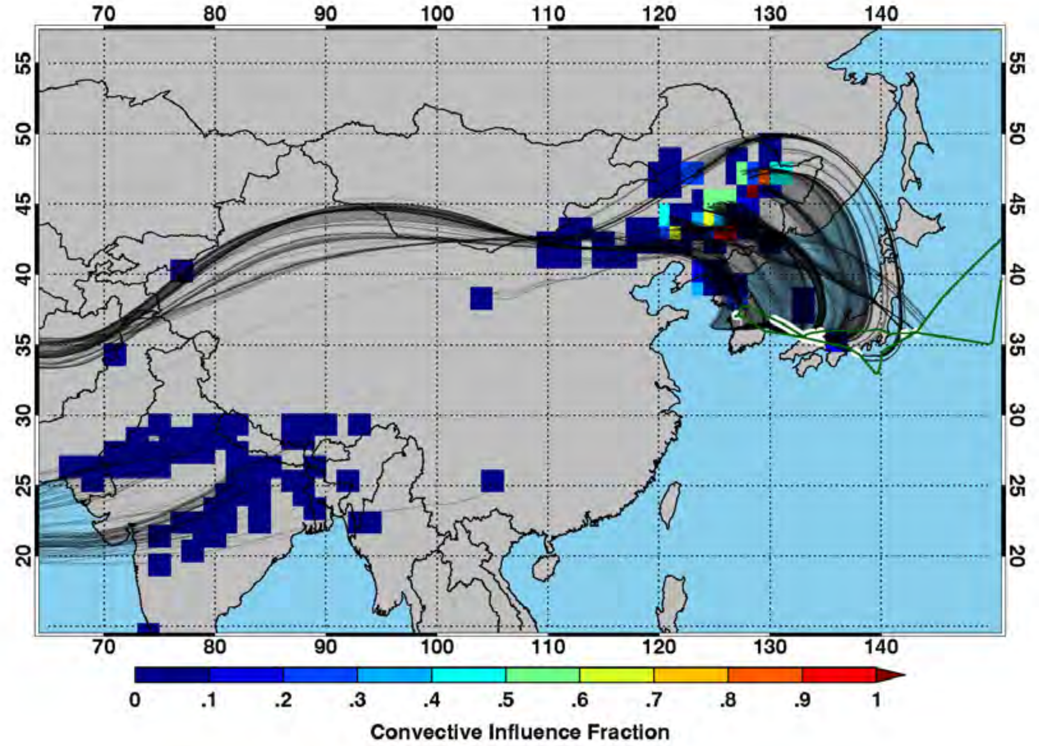
Assumes 80 moles Ltg. NOx / flash

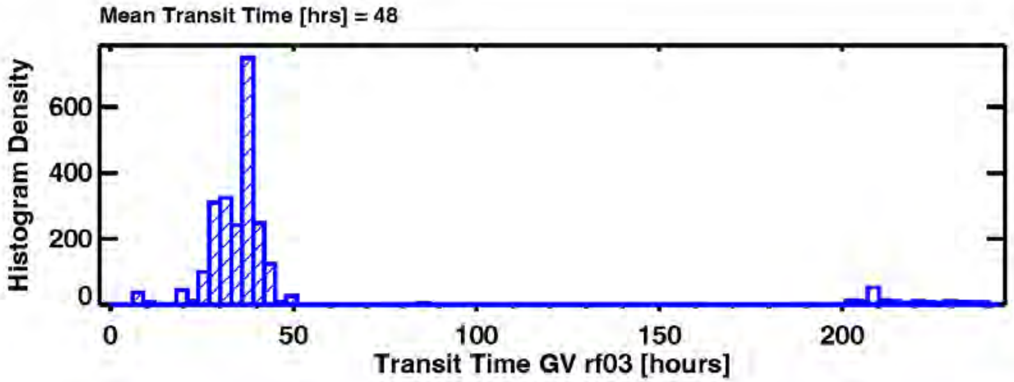
Results – RF03

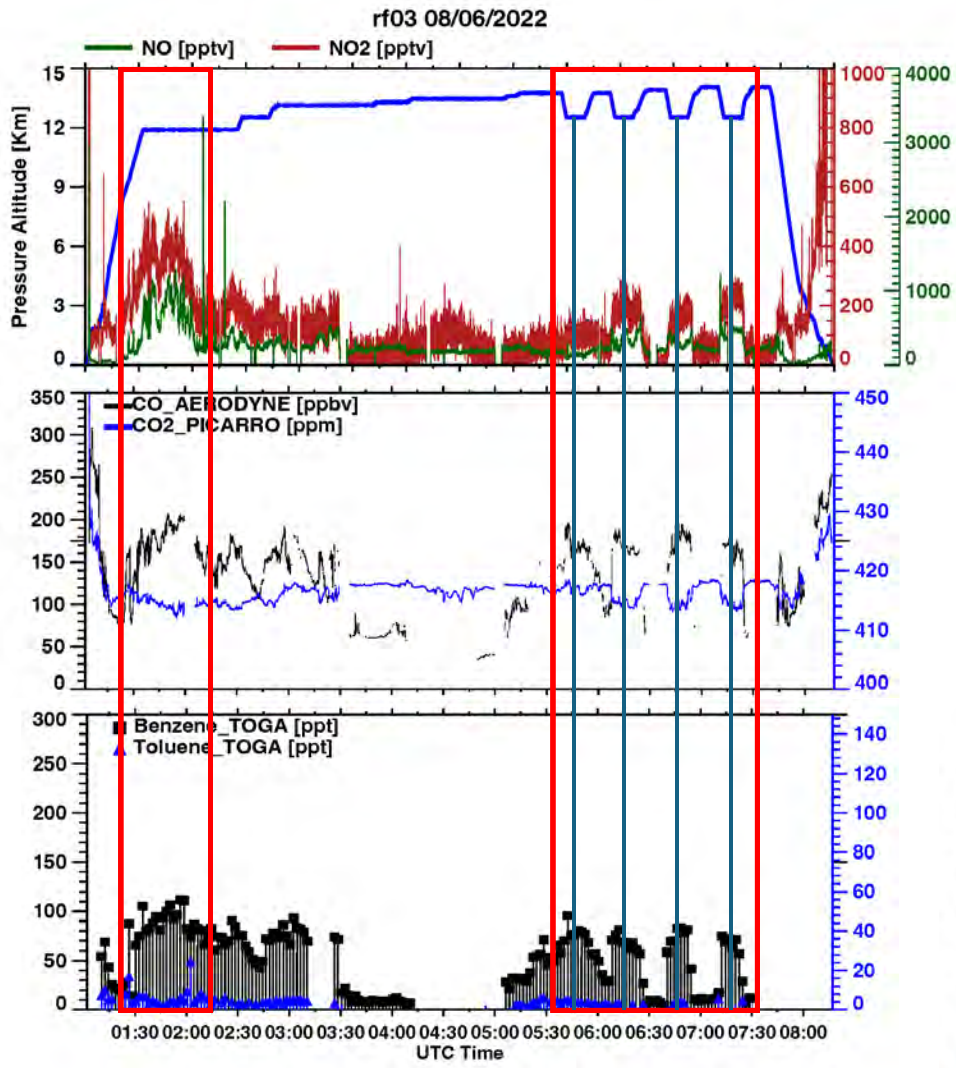




- Plume over Yellow sea - > NE China
- Bottom of Saw tooth – India
- Transit time – about 2 days (excluding India)

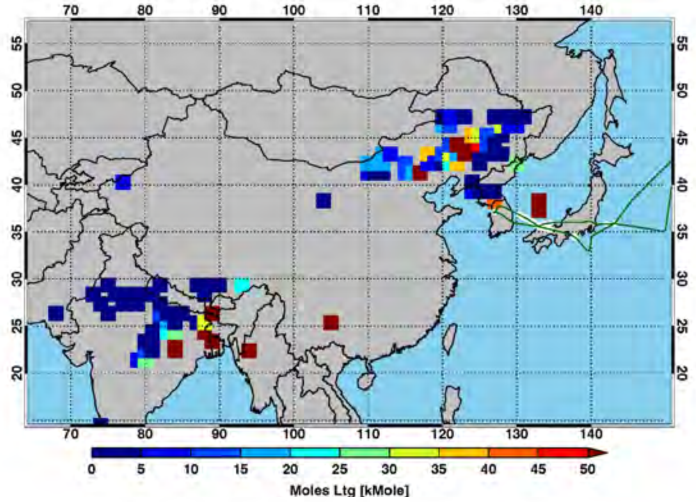


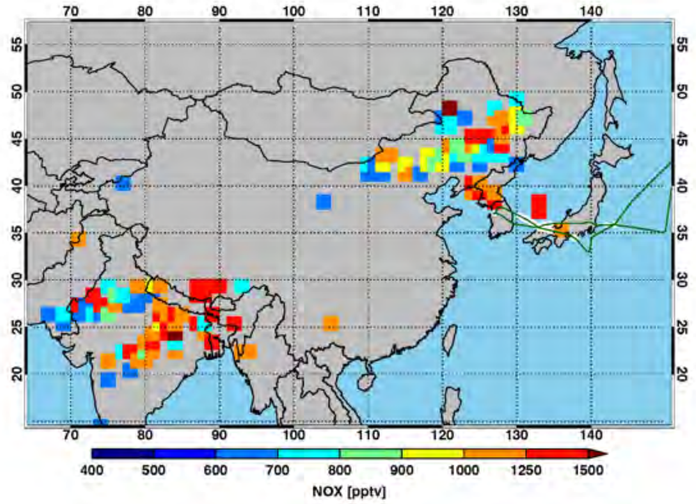


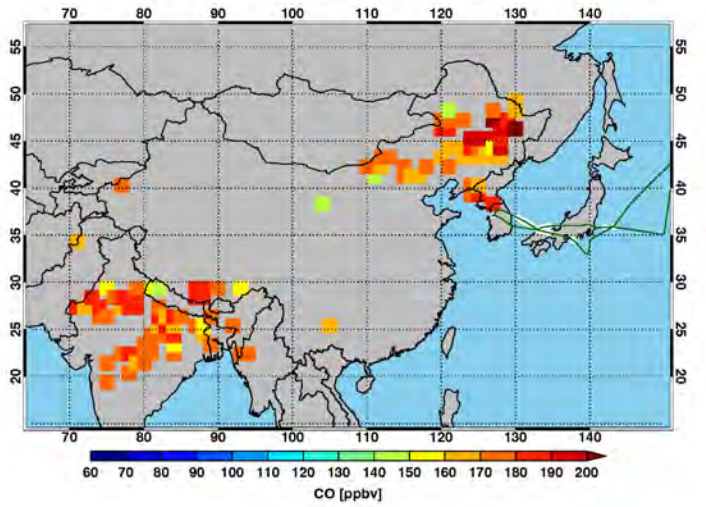


Results – RF03

- Elevated CO
- Background CO_2
- Elevated Benzene
- Low toluene – consistent with being a couple of days old.
- Pockets of high lightning density.



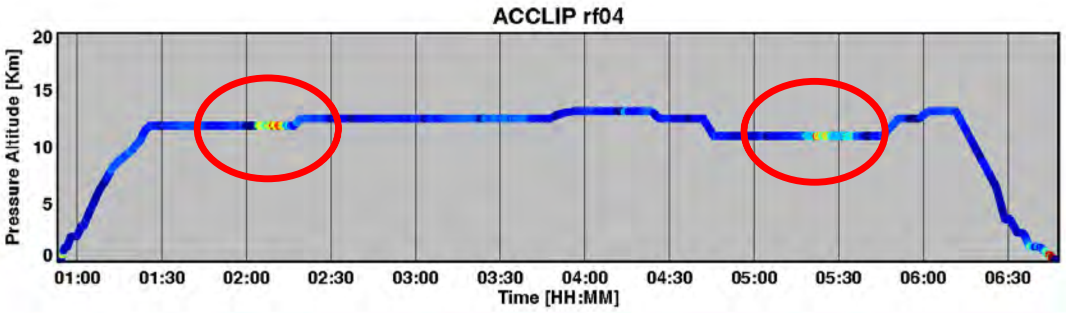


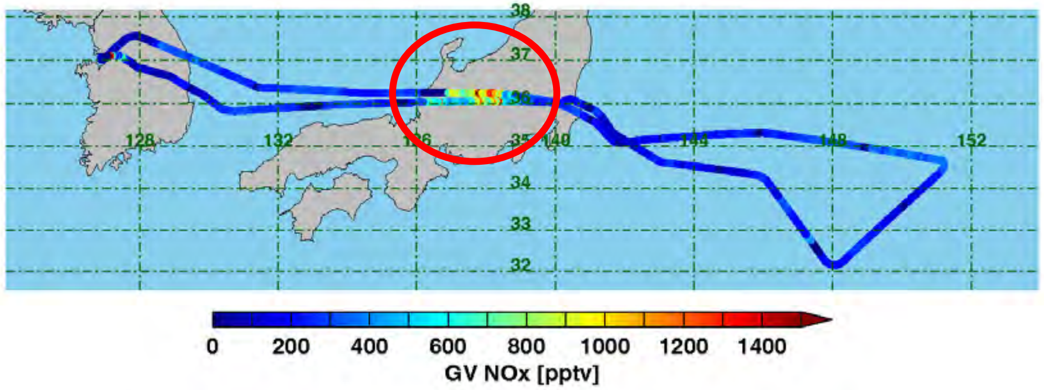


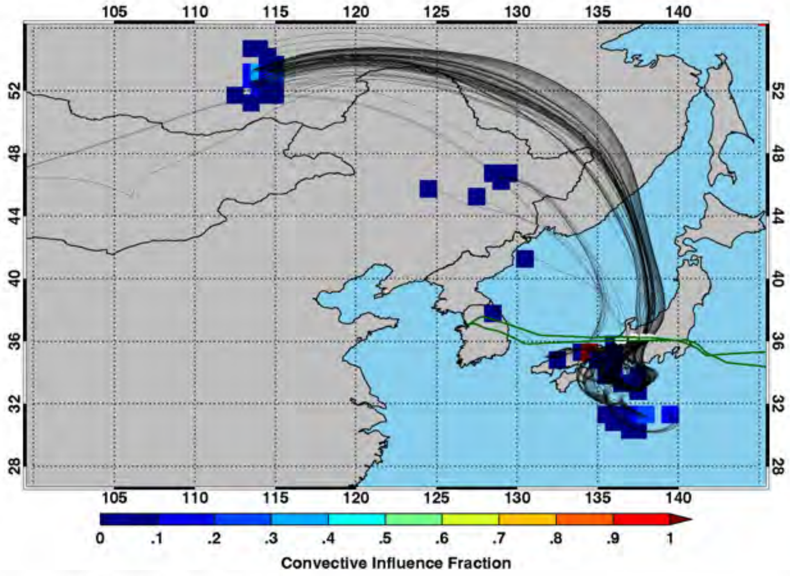
Consensus:
Mix of Pollution and Lightning

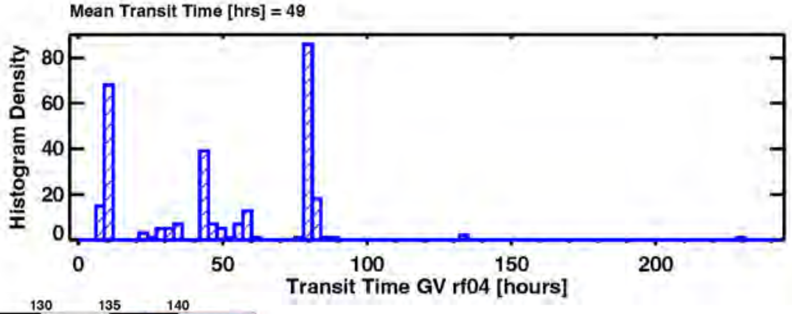
<u>Typical Atmospheric Lifetimes:</u>
Benzene : 12 days – pollution signature
Toluene: 2 days – pollution signature
NOx: ~ several days to a week in UT
CO/CO_2: Months - CO: pollution signature, CO_2: aircraft exhaust

Results – RF04

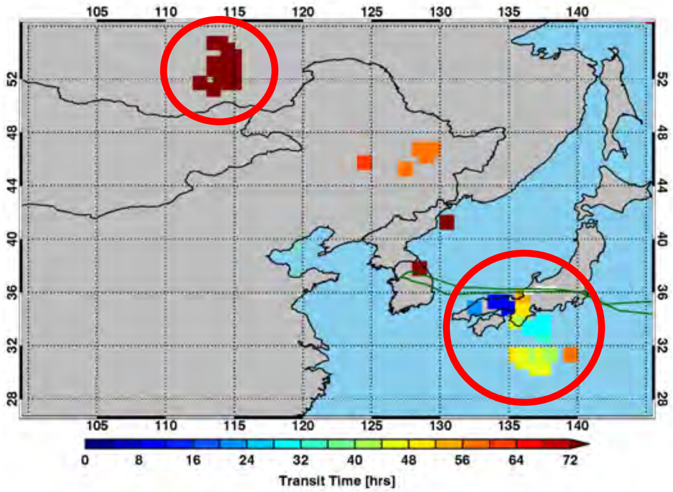




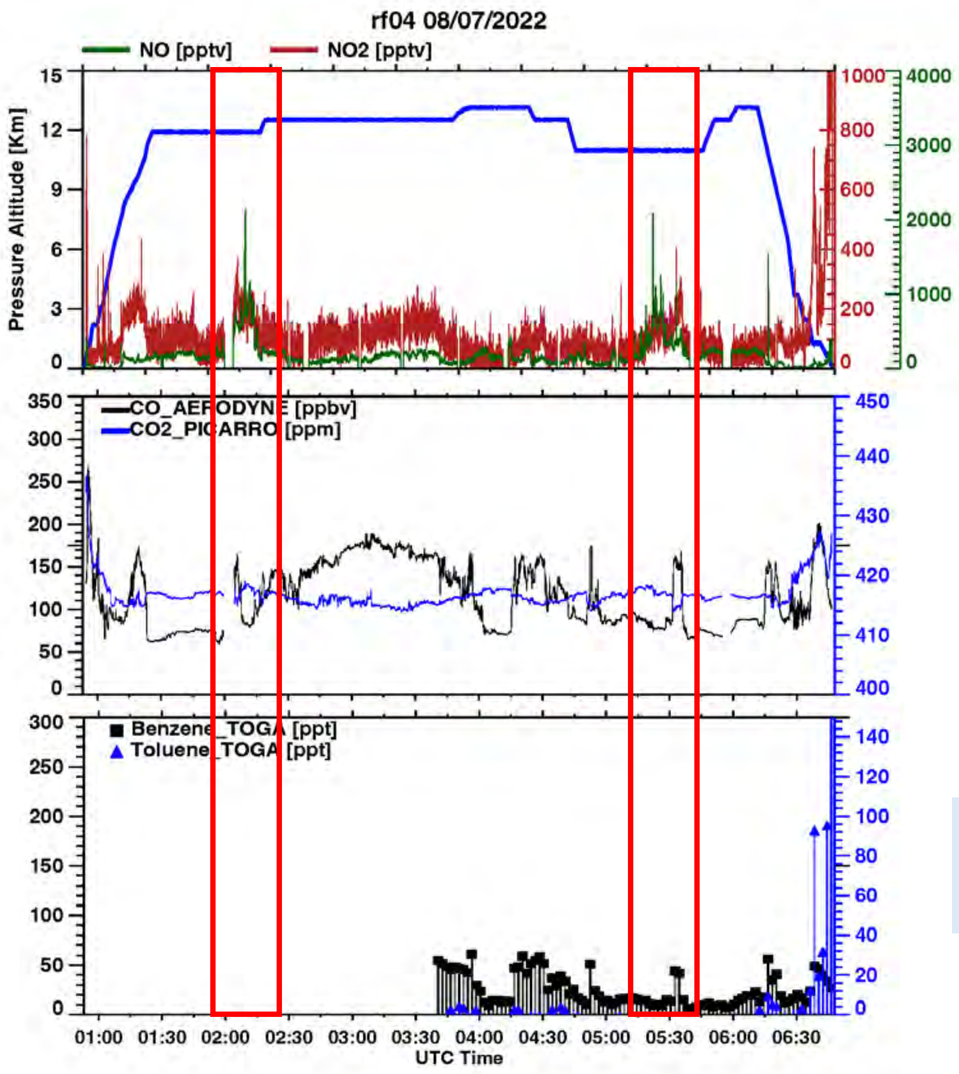




- Trajectories that go to Russia are on the outbound leg.
- Appears to have seen same plume 3 hours later but in a different wind regime based on trajectories.



Results – RF04



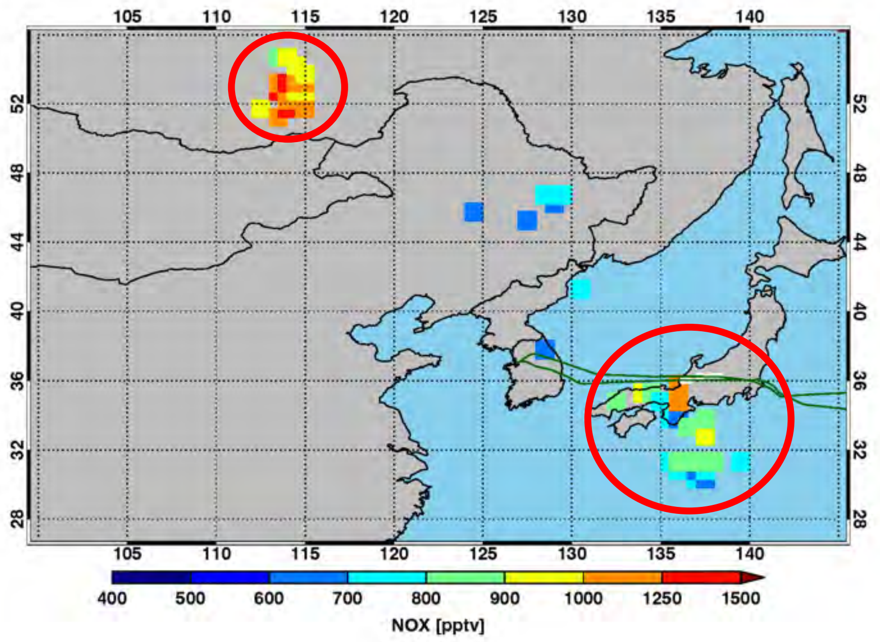
Best guess:

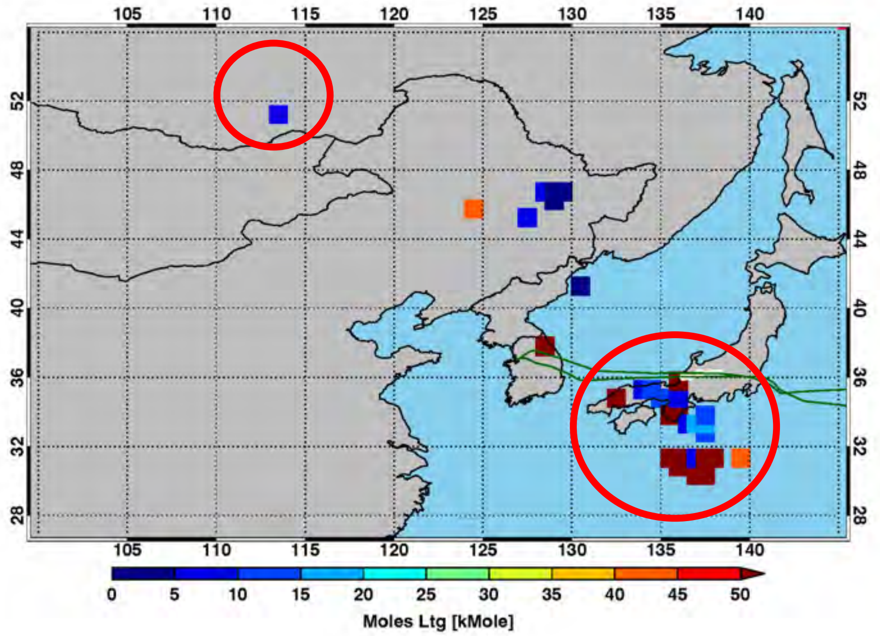
Trajectories over Russia should have terminated over/south of Japan.

Not consistently polluted (CO/CO_2/Benzene/Toluene).

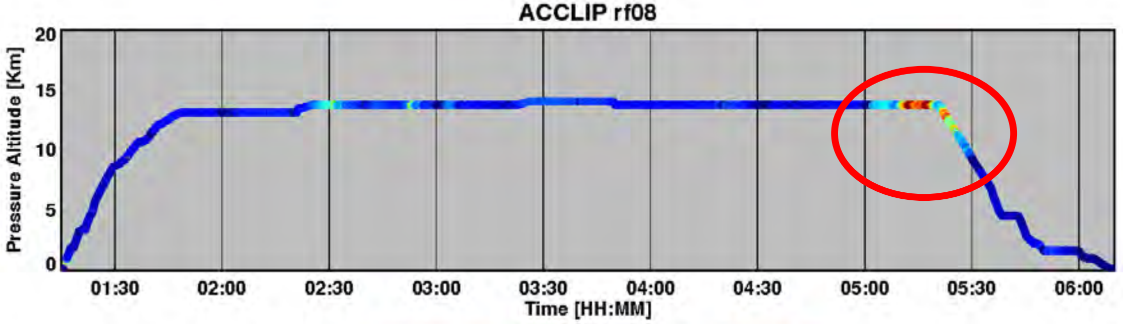
Strong lightning signature.

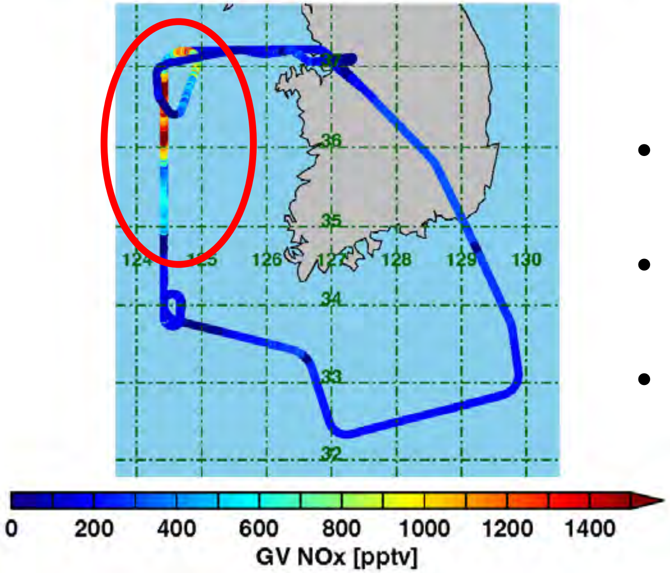
Consensus:
Primarily from Lightning

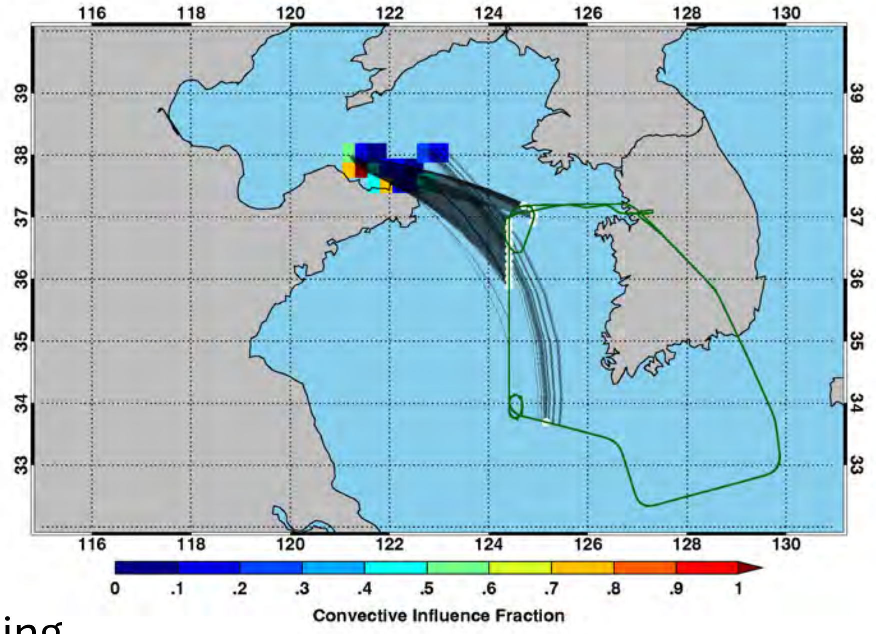




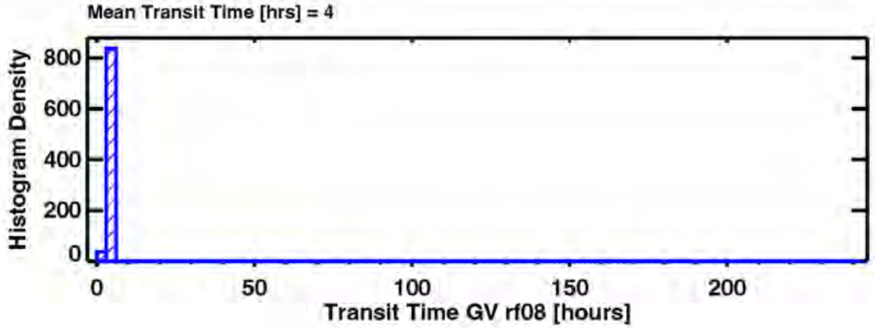
Results – RF08



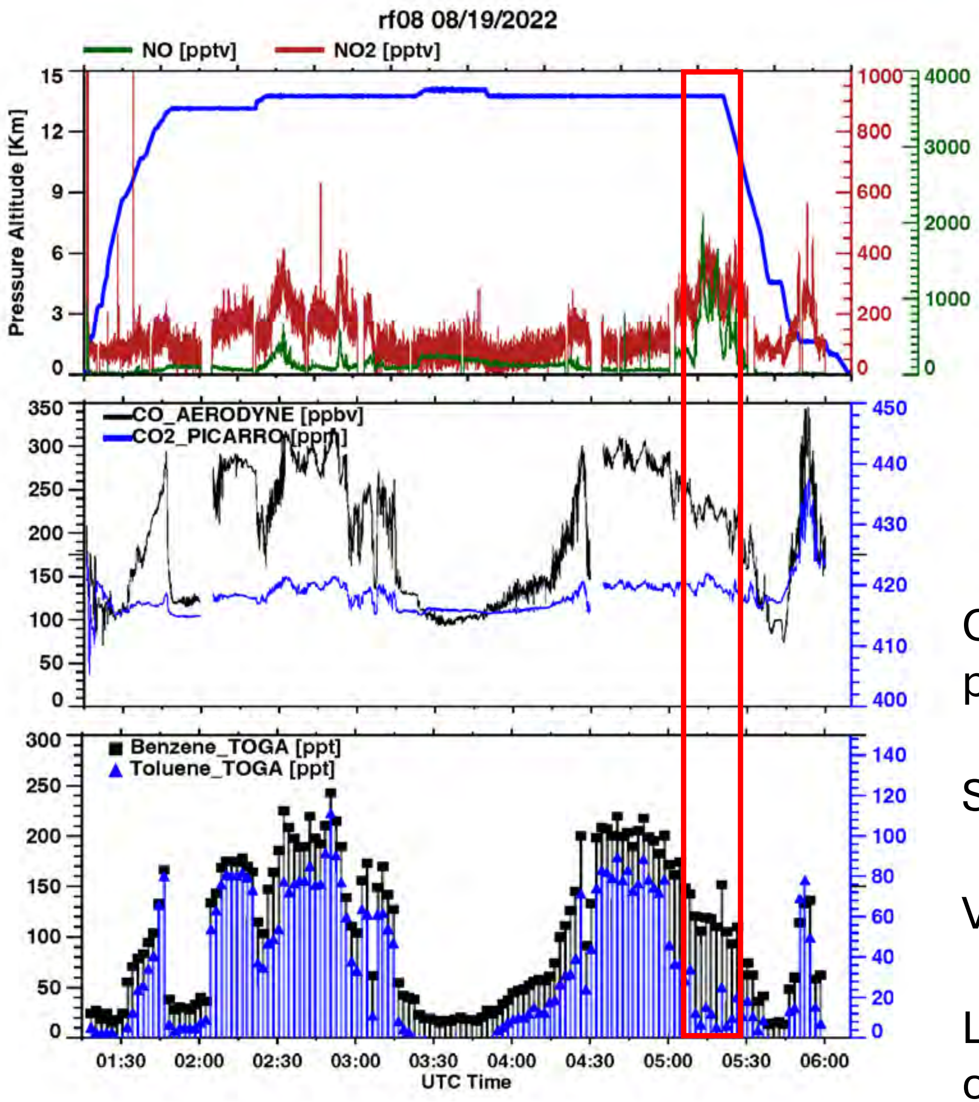


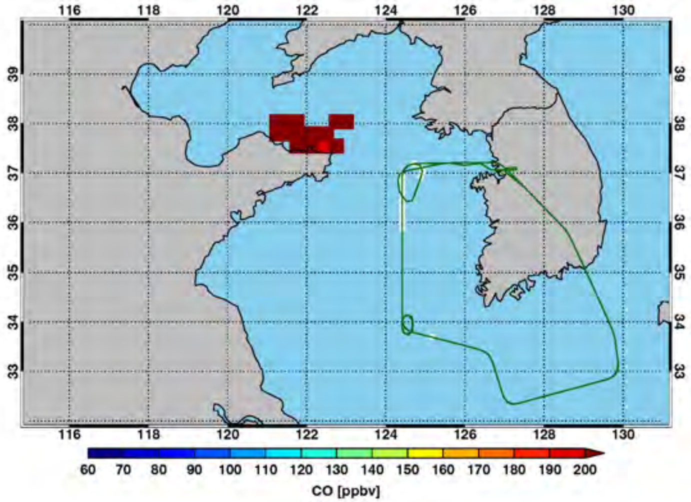


- Last leg of the flight before landing.
- Concentrated to the Yellow Sea.
- Very short transit time.



Results – RF08



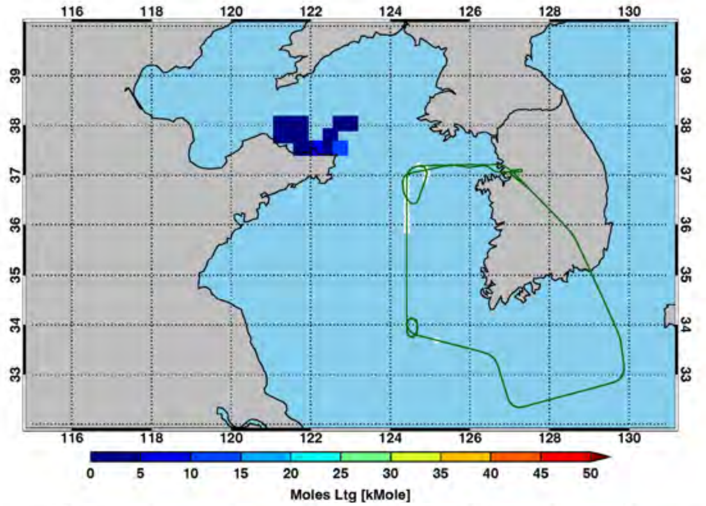


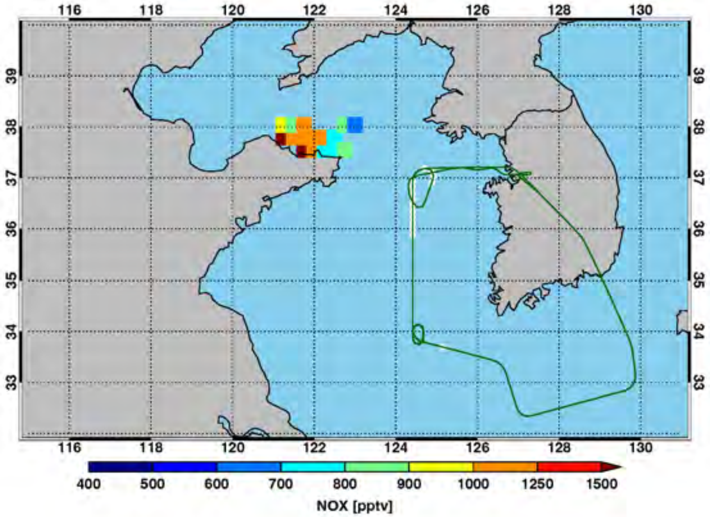
Overall flight is heavily influenced by pollution, *however*…

Shrinking pollution signature.

Very short transit time.

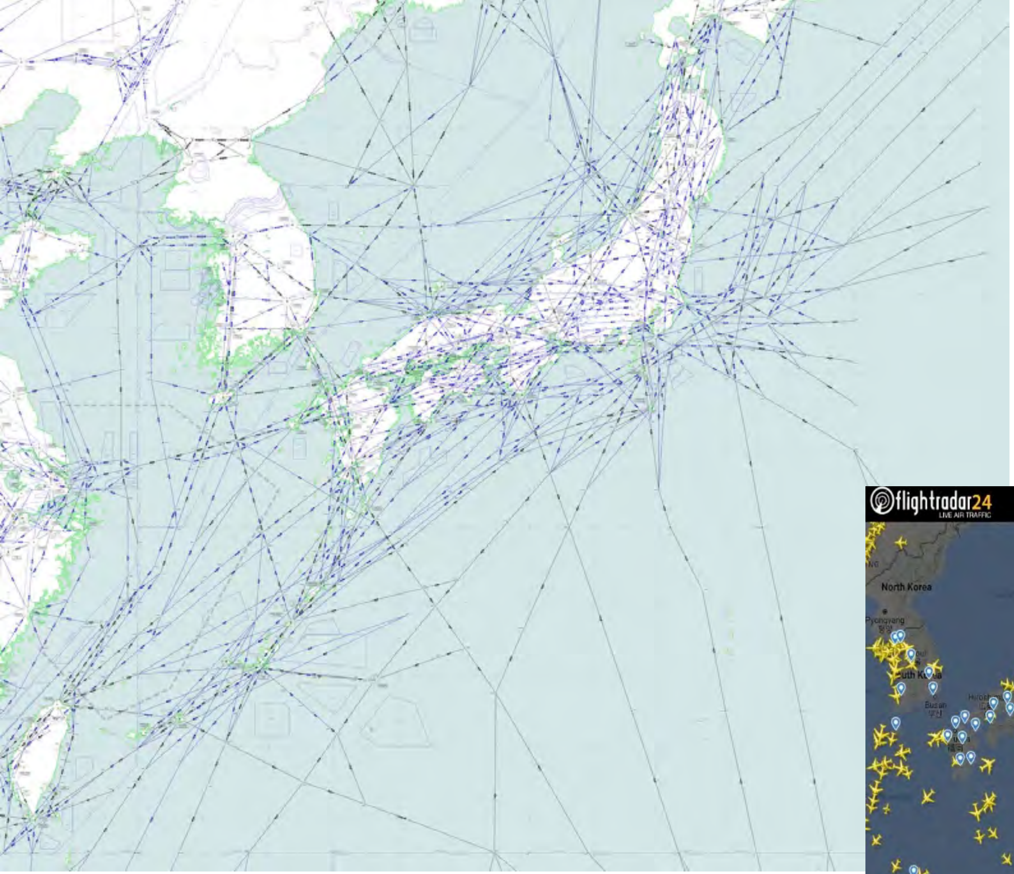
Low lightning density but very fresh outflow.





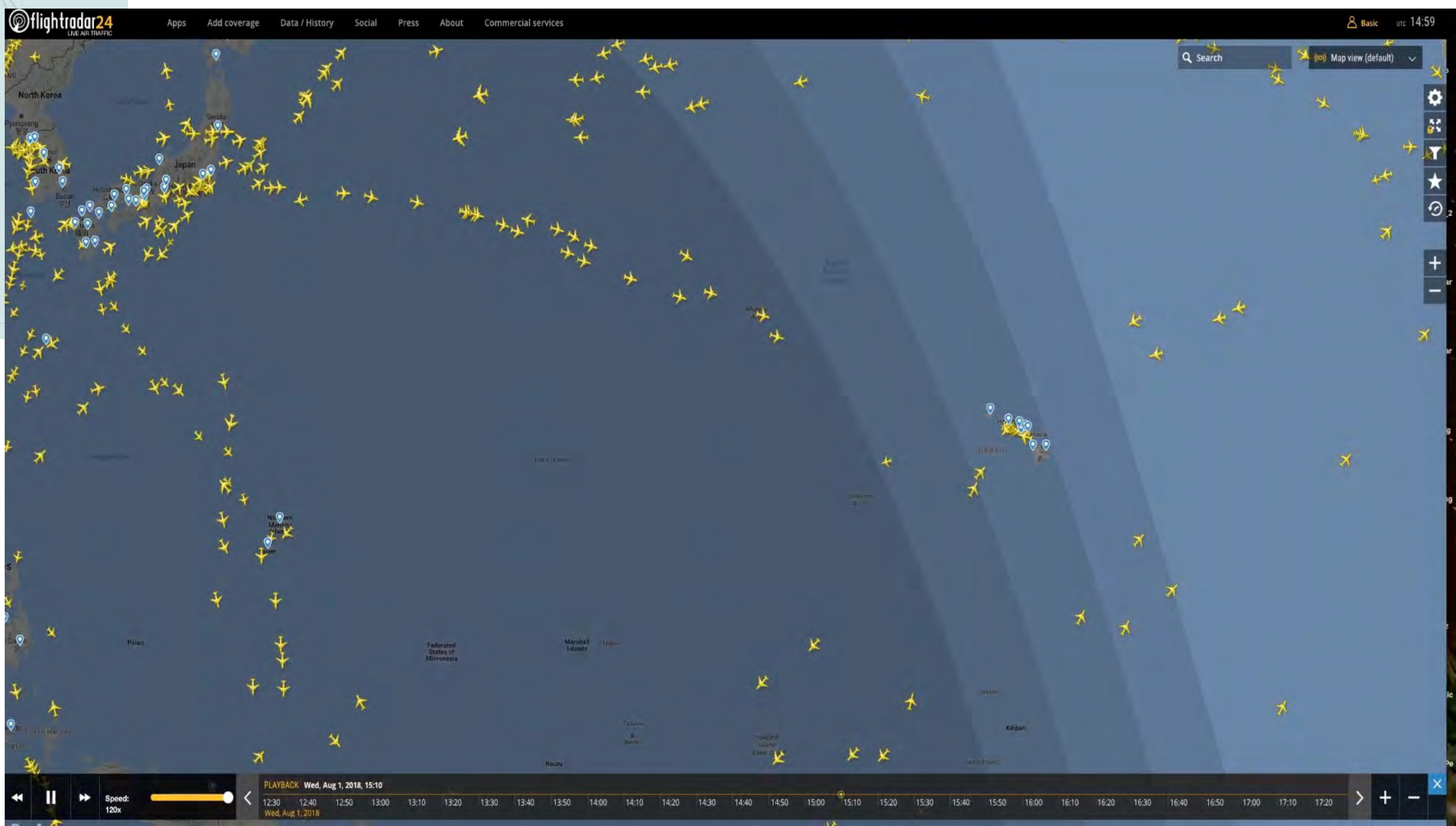
Consensus:
More lightning than Pollution.

Aircraft Exhaust



A lot of air traffic .

Spent most of our time on airways.



Aircraft exhaust:

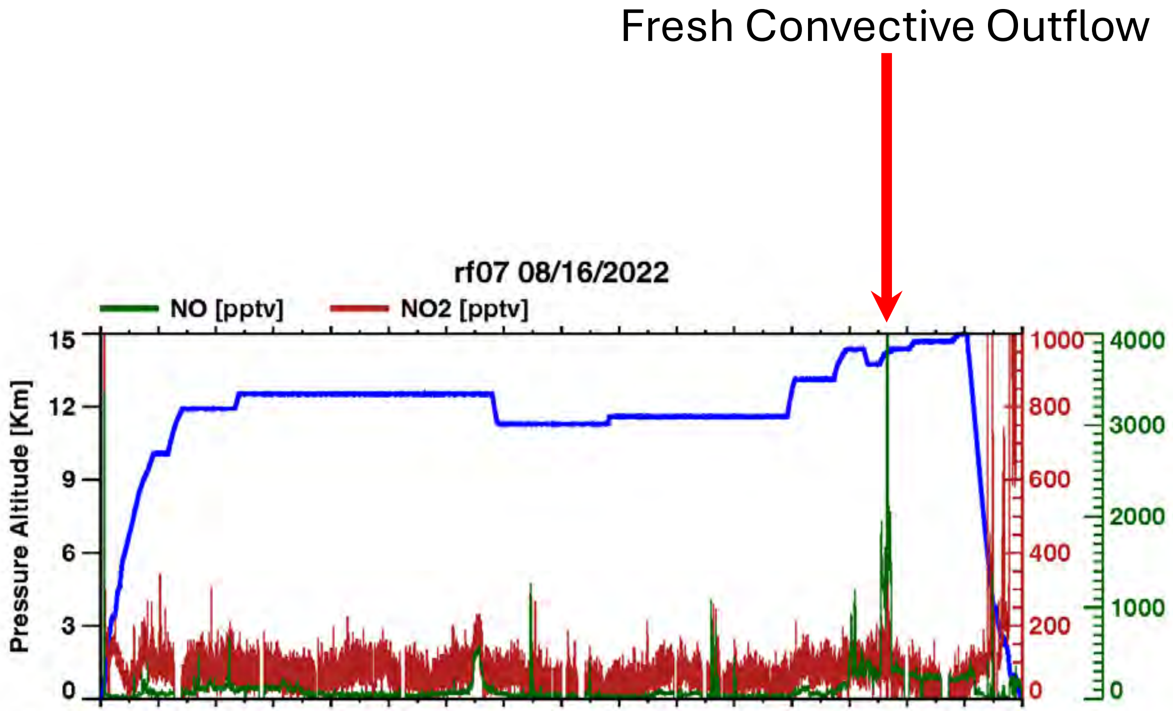
- CO
- CO_2
- H_2O
- BC
- NOx
- Hydrocarbons
- Sulfur

Summary & Conclusions

- High NOx events observed by the NCAR/NSF GV during ACCLIP were explored using a suite of tools.
- Source regions were located by Lagrangian kinematic back trajectories terminated at convective cloud tops.
  - The primary source regions were found to be Northeast China (RF03), the Yellow Sea region (RF08), and areas in and around Japan (RF04).
- **Initial conclusions about the origins of observed high NOx**:
  - RF03: Even mix of anthropogenic and lightning.
  - RF04: Primarily from lightning.
  - RF08: Mix of anthropogenic and lighting, but more heavily weighted towards a lightning influence.
  - Can’t yet confirm if there is an aircraft exhaust signature present.
  - None of the other sources appear to be present (Intrusions, Volcanoes, wildfires).
  - Analysis suggests that high GV NOx observations have a stronger lightning influence than anthropogenic.

Continuing Work

- Add more satellite (GEMS) analysis.
  - Take advantage of the hourly data output.
- Try to quantify sources.
- Use lightning data to tighten up the convective termination points of back trajectories.
- Compare a broader spectrum of GV species.
- Add in WB57 data to the analysis.
- Work RF07 into the analysis and compare signatures with the other flights…
  - Narrow swath of data.
  - Skirted an active convective storm and measured the highest NOx of the campaign.



Thank You!

Questions/Comments?