Reactive Nitrogen in the Global Upper Troposphere from Aircraft Campaigns and GEOS-Chem

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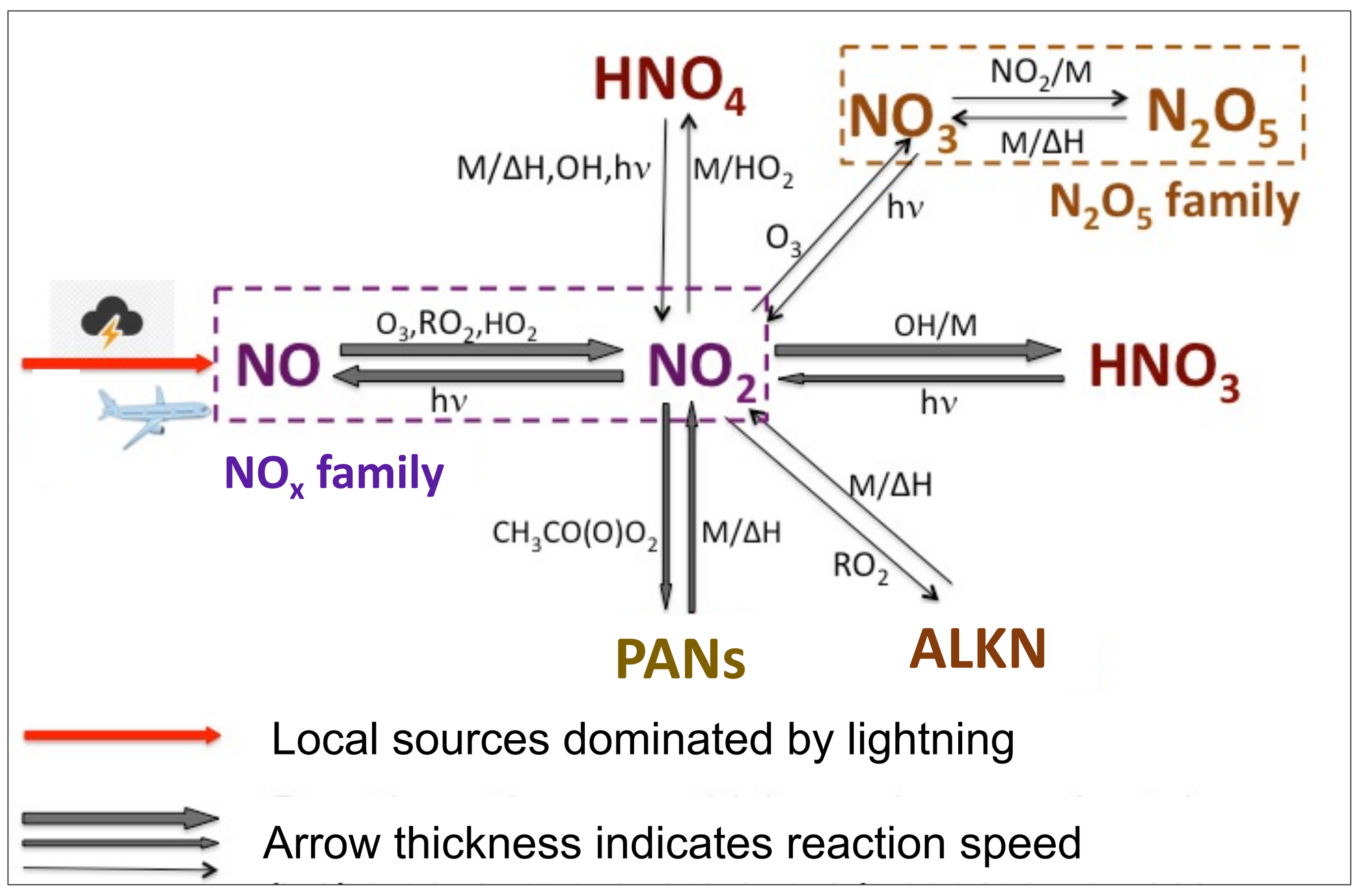
Major Finding: We identified that missing loss processes in GEOS-Chem for the PAN-like compound PPN accounts for almost 50% of the model underestimate in NO_2 in the upper troposphere compared to NASA DC8 aircraft observations.

1. Motivation and Approach

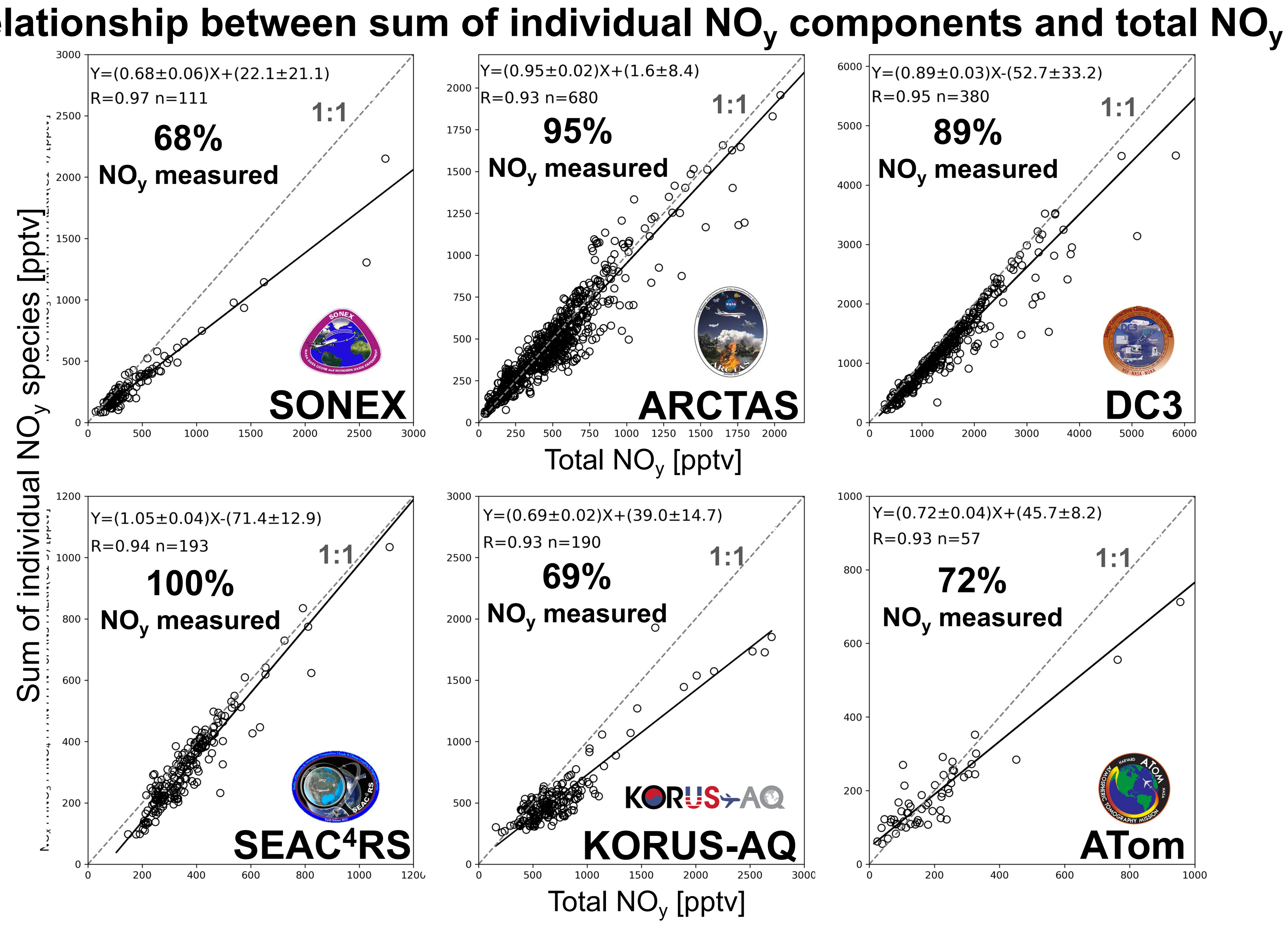
- Reactive nitrogen (NO_y) in the upper troposphere (UT; 8-12 km) impacts global climate, air quality, and atmospheric oxidants.



- There are large uncertainties in NO_y in the UT, evidenced by discrepancies between state-of-science models and observations.
- We use NASA DC8, MOZAIC, and IAGOS aircraft observations and GEOS-Chem v13.0.2 to identify and quantify these errors.
- Aircraft observations are screened for stratospheric influence and aircraft plumes

2. Proportion of UT NO_y measured during DC8 campaigns

Relationship between sum of individual NO_y components and total NO_y

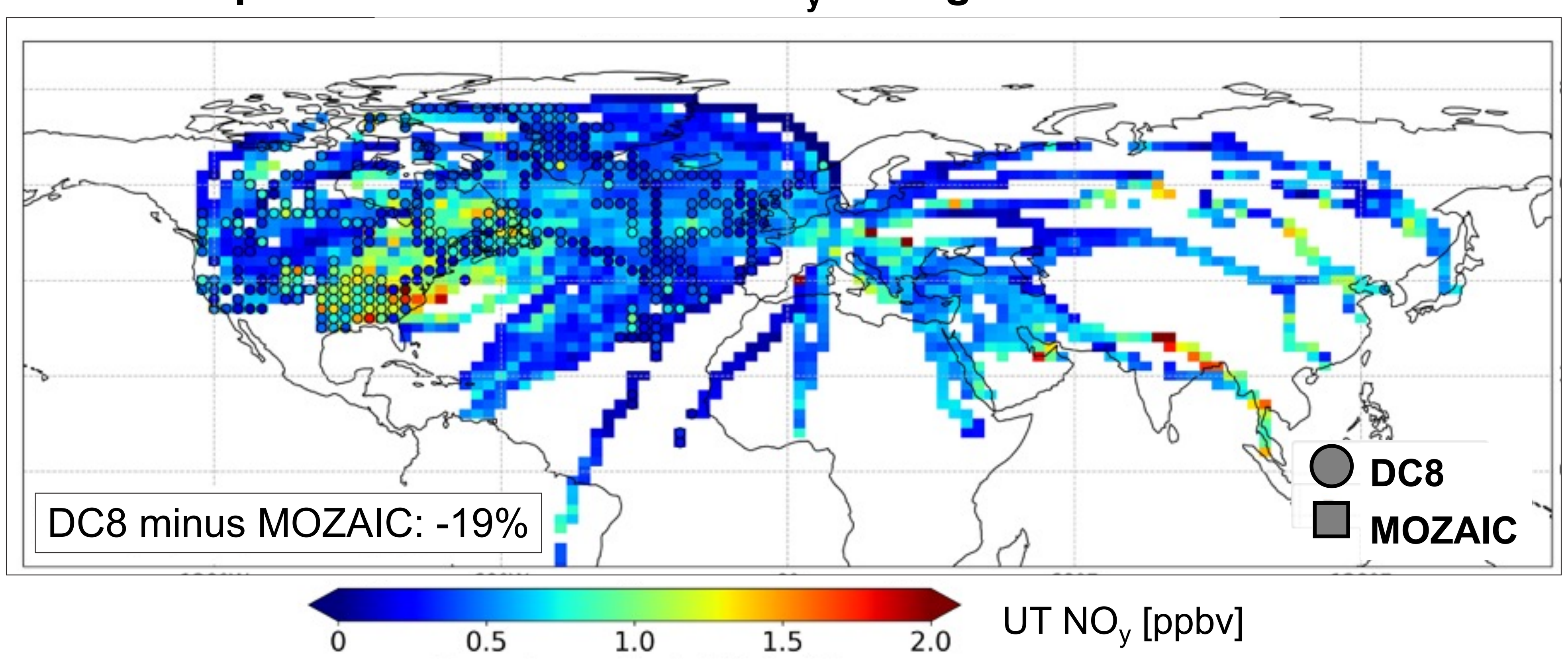


- Most (68-100%) UT NO_y contributed by a handful of species (NO_x, PANs, HNO_3, HNO_4 and organic nitrates). No NO_2 during SONEX. KORUA-AQ influenced by biomass burning.

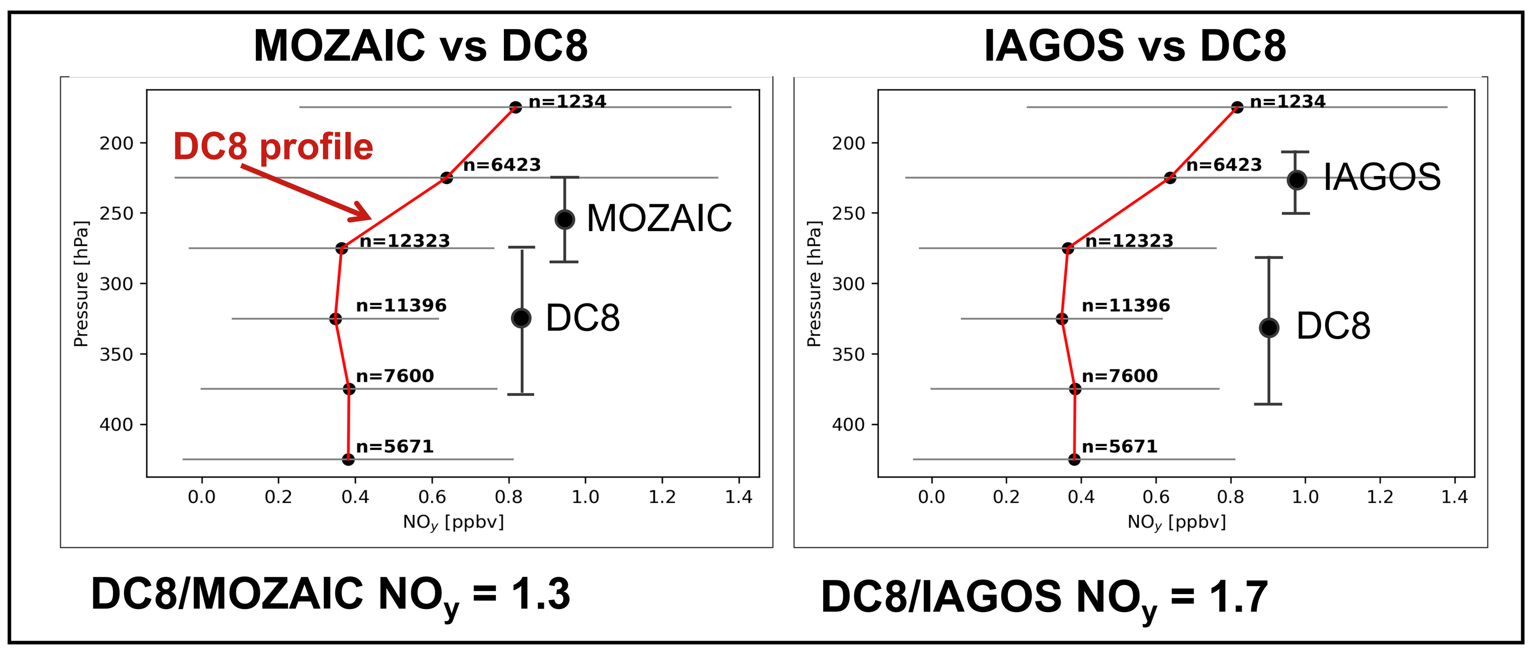
3. Consistent between aircraft observations of total UT NO_y from DC8, MOZAIC and IAGOS

- MOZAIC and IAGOS commercial aircraft campaigns have multiple years of observations of total NO_y at cruising altitude, so can be used to assess whether DC8 measurements offer a climatology of reactive nitrogen in the UT. Comparison requires accounting for different vertical sampling of UT.

Spatial distribution of UT NO_y during DC8 and MOZAIC

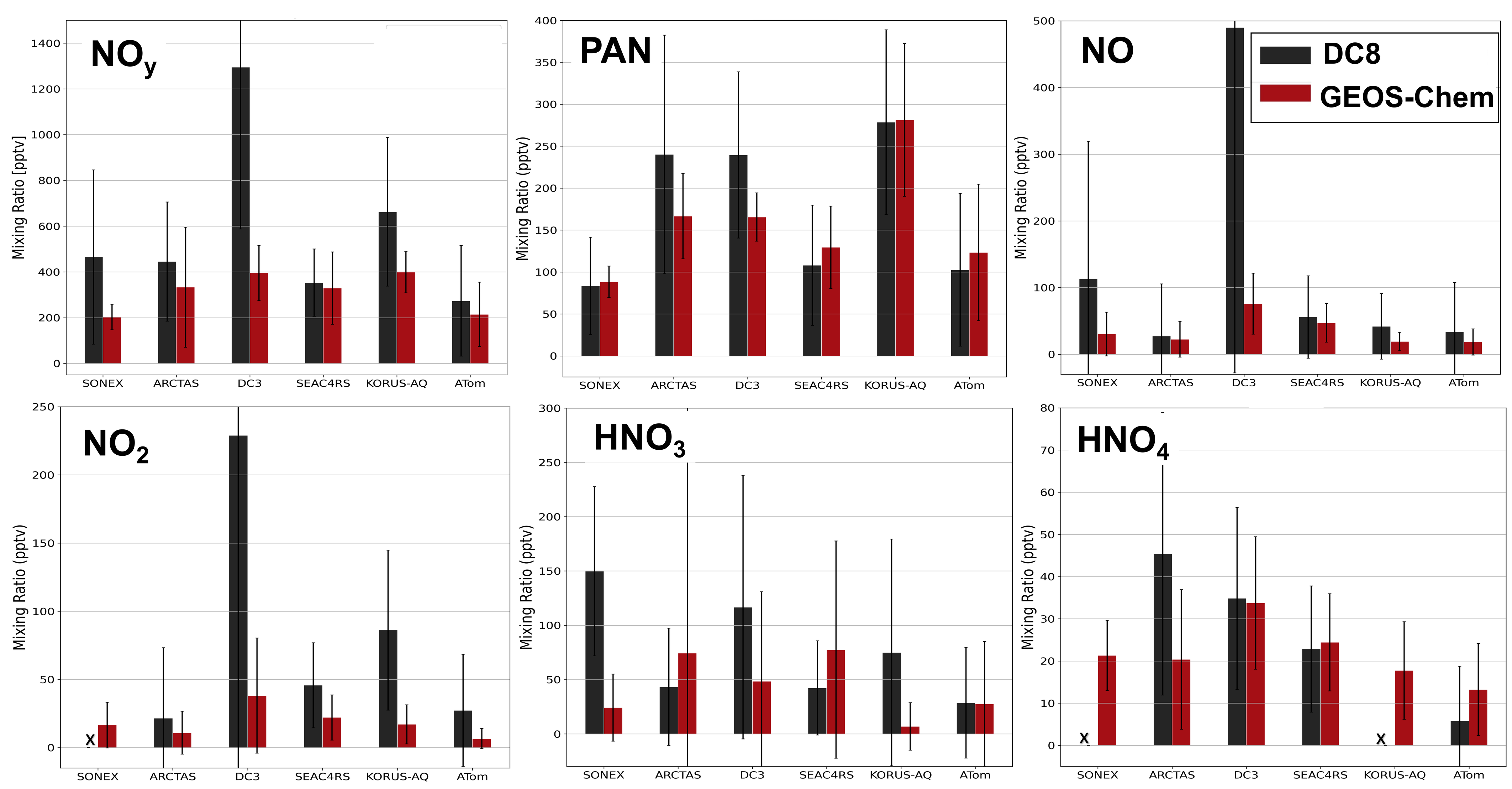


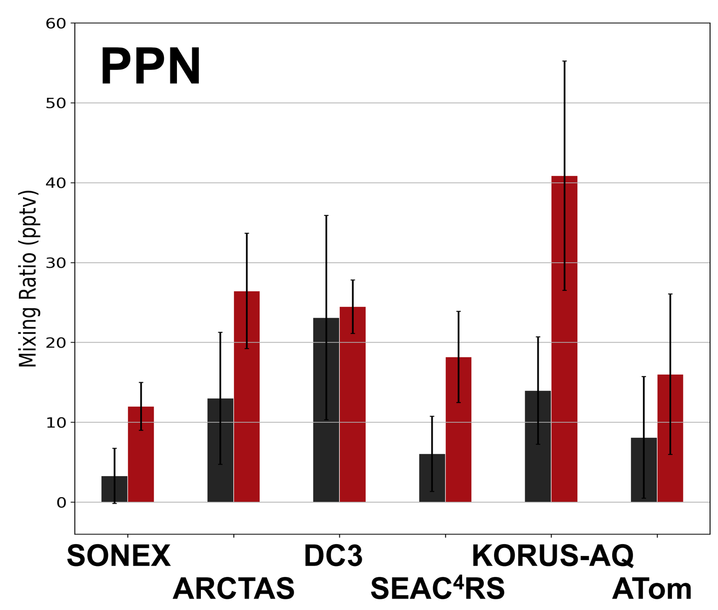
Vertical sampling extent of aircraft campaigns



DC8 less than MOZAIC (by 19%) and than IAGOS (by 41%; not shown). Difference is consistent with altitude sampled (commercial aircraft at higher altitude where NO_y more abundant).

4. GEOS-Chem skill at simulating UT reactive nitrogen





GEOS-Chem underestimates total NO_y from the underestimating NO by 5.0-80 ppt and NO_2 by 11-24 ppt (excluding DC3 and KORUS-AQ).

DC3 anomalous, as sampling thunderstorms with fresh NO_x from lightning.

GEOS-Chem locks up about 10 ppt NO_2 as the PAN-like compound PPN that is overestimated in the model due to missing PPN loss processes.

5. Concluding Remarks

1. Most total measured reactive nitrogen in the upper troposphere is from a few individual components.
2. DC8 is roughly consistent with MOZAIC and IAGOS climatology after accounting for different altitudes sampled.
3. GEOS-Chem v13.0.2 underestimates UT NO_y due to an underestimate in NO_x.
4. The model overestimates the PAN-like PPN compound due to missing loss processes in the model.
5. Next: Evaluate DC8 against NO_2 obtained from cloud-slicing TROPOMI partial columns.

References

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Stevenson et al., 2013, doi:10.5194/acp- 13-3063-2013

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