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

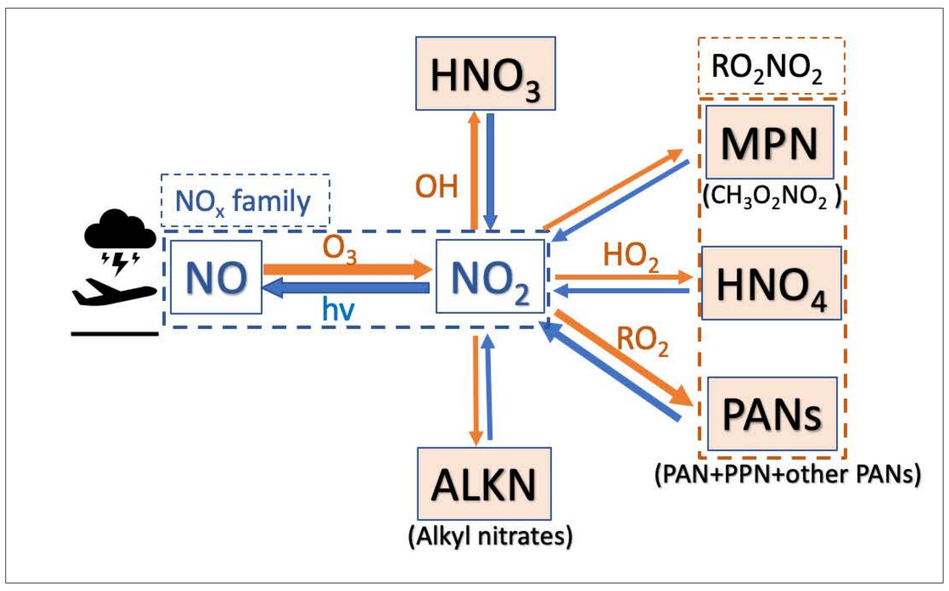
Nana Wei (nana.wei.21@ucl.ac.uk), Eloise. A. Marais, J. F. Roberts, R. G. Ryan, G. Lu, and NASA DC8, MOZAIC and IAGOS Teams

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.

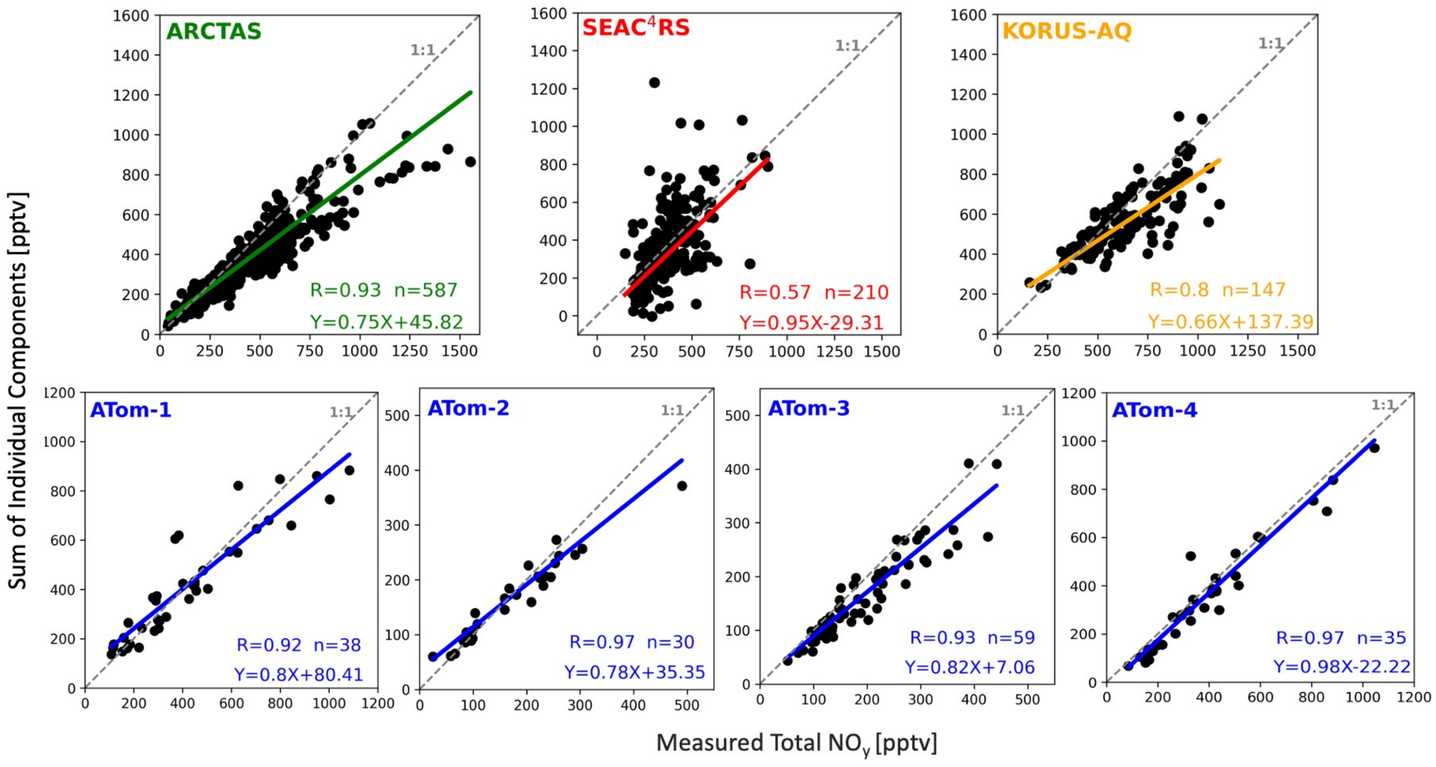
Recycling pathways of UT NO_y components



- 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.

2. Proportion of UT NO_y measured during DC8 campaigns

Relationship between sum of individual NO_y components and total NO_y

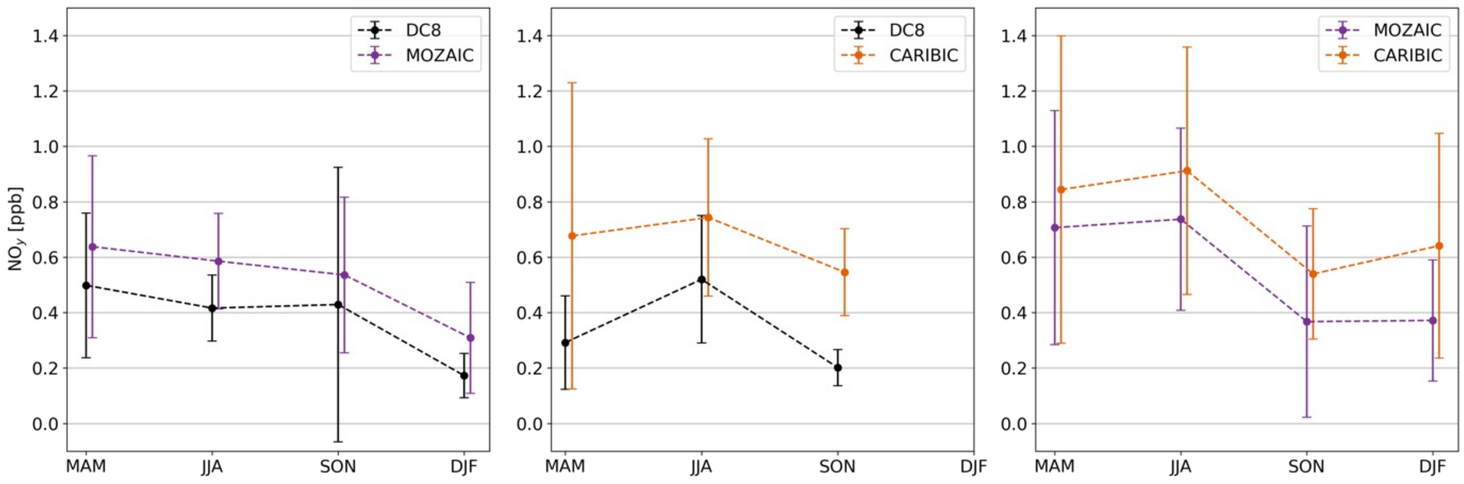


- Most (66-98%) UT NO_y contributed by a handful of species (NO_x, HNO_3, ALKN and peroxy nitrate (RO_2NO_2) including PANs, HNO_4 and MPN). ATom 1-2 measured only PAN, with no measurements of PPN or other PANs species.

3. Consistent between aircraft observations of seasonal UT NO_y from DC8, MOZAIC and CARIBIC

- MOZAIC and CARIBIC 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 sampling years.

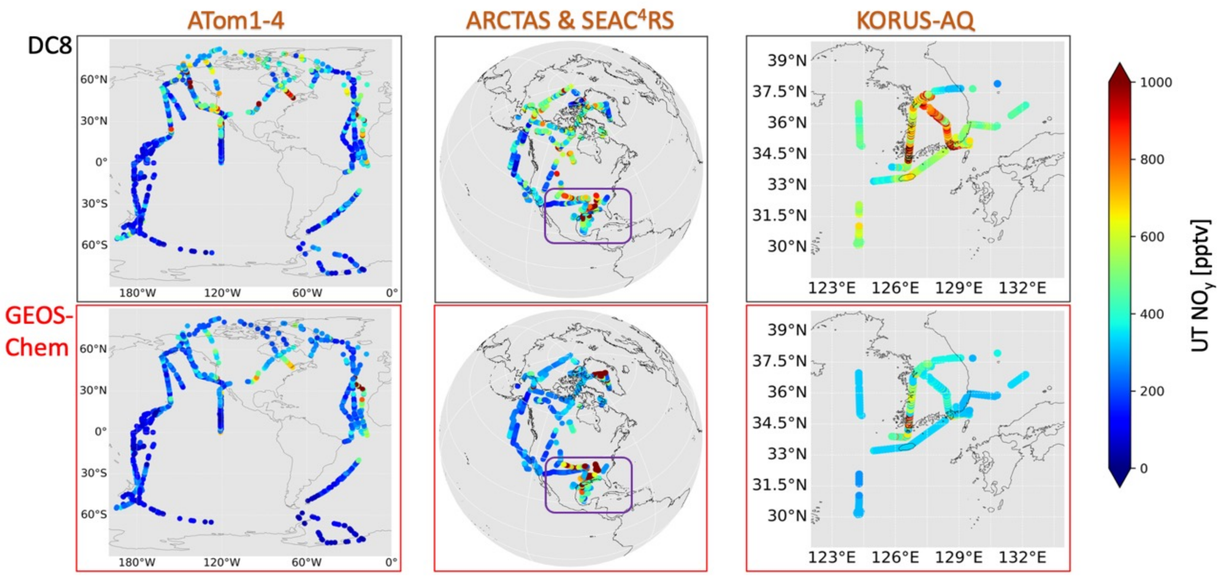
Seasonal mean NO_y mixing ratio in overlapping regions



- UT NO_y levels are higher in spring and summer than autumn and winter due to increased lightning activity and photochemistry during MAM and JJA.
- MOZAIC and CARIBIC consistently record higher seasonal UT NO_y levels than DC8, due to larger instrument interference from HCN conversion efficiency.
- IAGOS measures higher UT NO_y levels than MOZAIC in northern hemisphere regions, due to differing sampling years (IAGOS: 2008-2018, MOZAIC: 2002-2005).

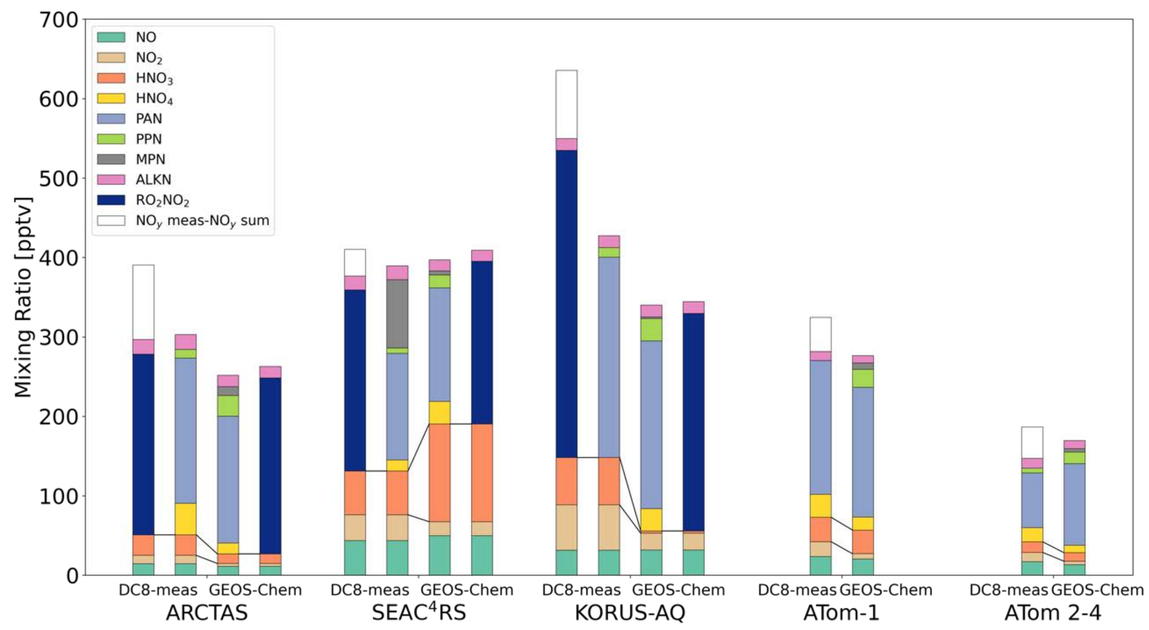
4. GEOS-Chem skill at simulating UT reactive nitrogen

Spatial distribution of UT total NO_y from DC8 and GEOS-Chem



- GEOS-Chem underestimates total NO_y in the UT during ARCTAS and KORUS-AQ due to a lower bias in NO_2, HNO_3 and peroxy nitrates.
- GEOS-Chem routinely overestimates the PAN-like PPN compound due to missing PPN photolysis processes, accounting for almost 50% of NO_2 underestimation.

Comparison of UT NO_y components from DC8 and GEOS-Chem



- The underestimation of NO_2 by GEOS-Chem during KORUS-AQ is also attributed to its underestimation of O_3 since NO_2 is mainly formed from the reaction of NO with O_3 in the UT.

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 years sampled.
3. GEOS-Chem v13.0.2 underestimates UT NO_y mainly due to an underestimation in NO_2, HNO_3 and peroxy nitrates.
4. GEOS-Chem overestimates the PAN-like PPN compound due to missing loss processes in the model.
5. GEOS-Chem underestimates UT NO_2 due to its locking NO_2 as PPN and underestimation of O_3, the precursor of UT NO_2.

Acknowledgements
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