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

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Comprehensive observations from NASA DC8 + Multiple years of observations from MOZAIC + State-of-science atmospheric chemistry from GEOS-Chem = Improved understanding of reactive nitrogen

1. Introduction and Methodology

- UT NO_y impact global climate, air quality, and atmospheric oxidants
- Large uncertainties of UT NO_y exist in models (Stevenson et al., 2013)
- NASA DC8 aircraft sampling in the UT (450-180 hPa) provides global coverage and has a long consistent record of NO_y and NO_y components. These include SONEX, ARCTAS, DC3, SEAC4RS, KORUS-AQ and ATom from 1997 to 2018.
- MOZAIC include multiyear UT NOy measurements to assess its climatology
- The GEOS-Chem model includes detailed treatment of NO_y chemistry.
- We use DC8, MOZAIC, and GEOS-Chem to better understand **global UT NO_y**
- We filter out stratospheric influence using the O_3-to-CO > 1.25 mol mol^{-1}, as is standard (Hudman et al., 2007; Marais et al., 2018)

2. Results: Does NASA DC8 Capture Most NO_y Components?

- Regression slopes of 0.7-1.1 suggest that most (near 100%) of the UT NO_y budget can be explained by a handful of measurements (NO_x, PANs, HNO_3, HNO_4 and C1-C5 organic nitrates) during DC8 campaigns.
- Slopes < 1.0 are due to missing observations (such as HNO_4 and NO_2 for SONEX)

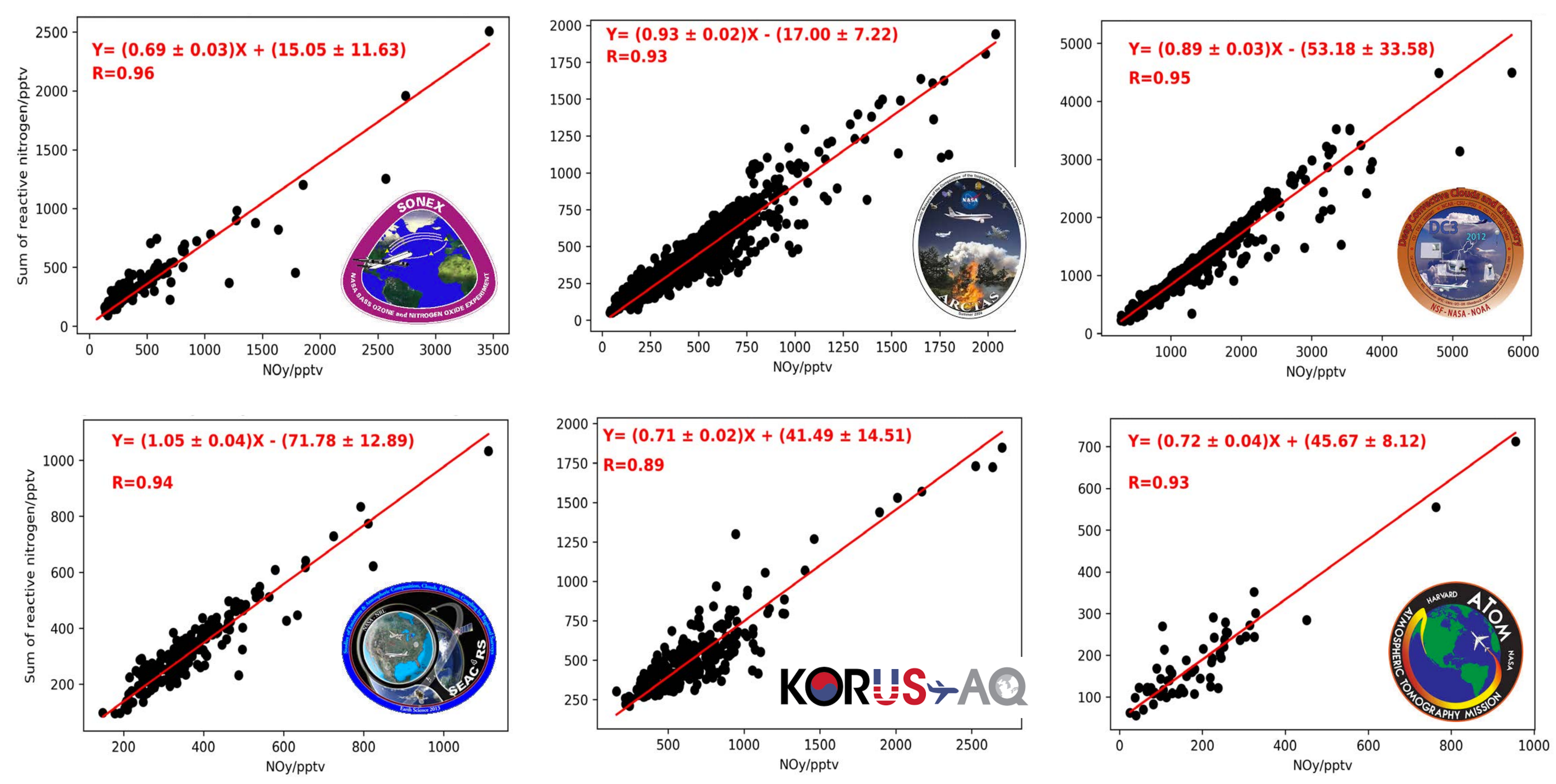

Figure 1: Budget of UT components measured during each campaign

3. Results: Are DC8 and MOZAIC UT NO_y consistent?

- Long-term abundance of UT NO_y is relatively consistent between MOZAIC and DC8 in all seasons, except winter, as indicated by regression slopes of 0.7-1.3.
- Winter comparison is poor due to a limited number of overlapping grids.

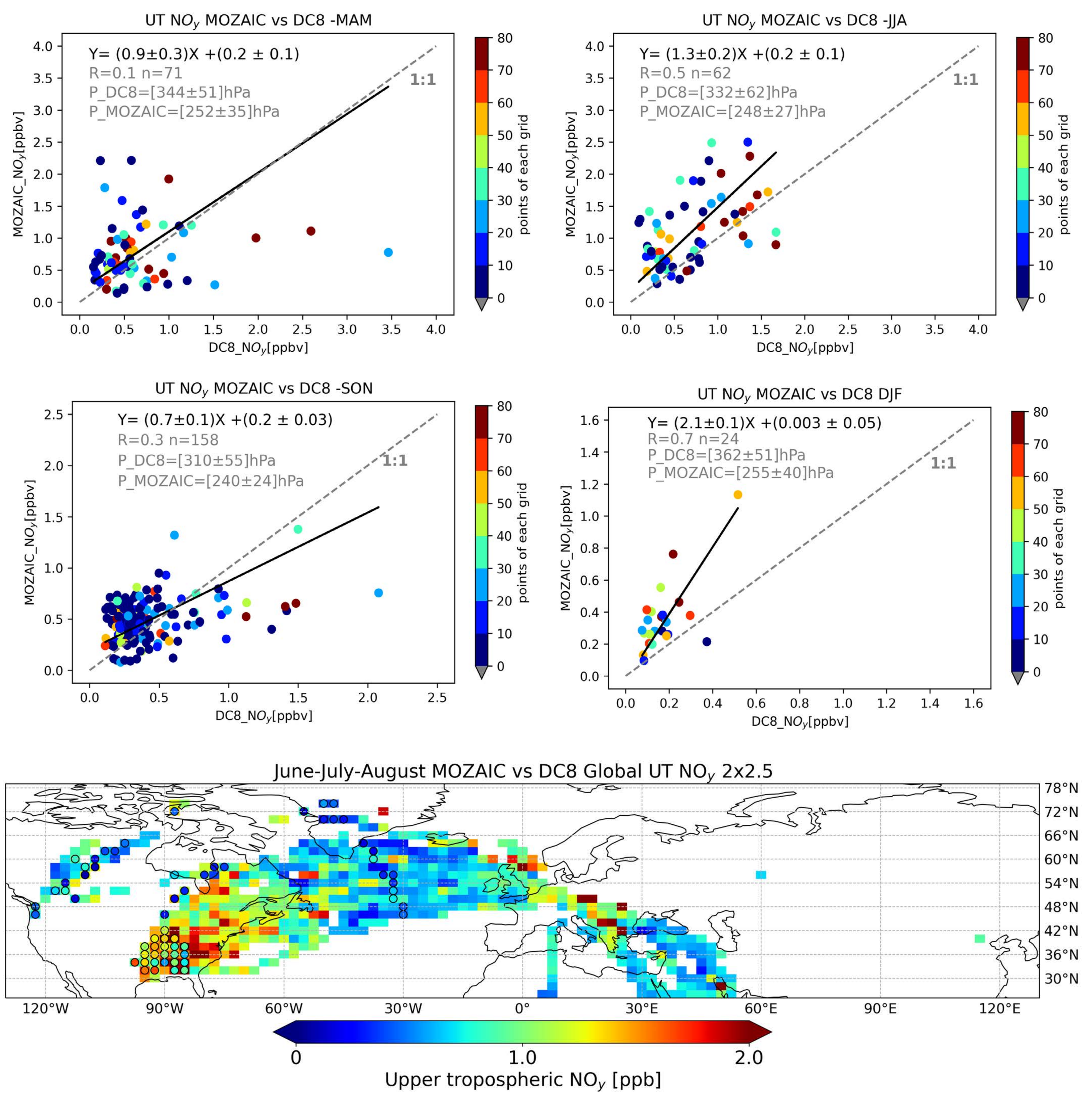

Figure 2: comparison of total measured reactive nitrogen in the upper troposphere between DC8 and MOZAIC during each season and spatial distribution of mean NO_y abundance, squares represent MOZAIC NO_y and points are DC8 NO_y

4. Results: Does GEOS-Chem reproduce observed UT NO_y?

- Model overestimates total NO_y for ATom1 but observed NO_y has large variability.
- Model overestimates HNO_3 for ATom1 measured in 2016.

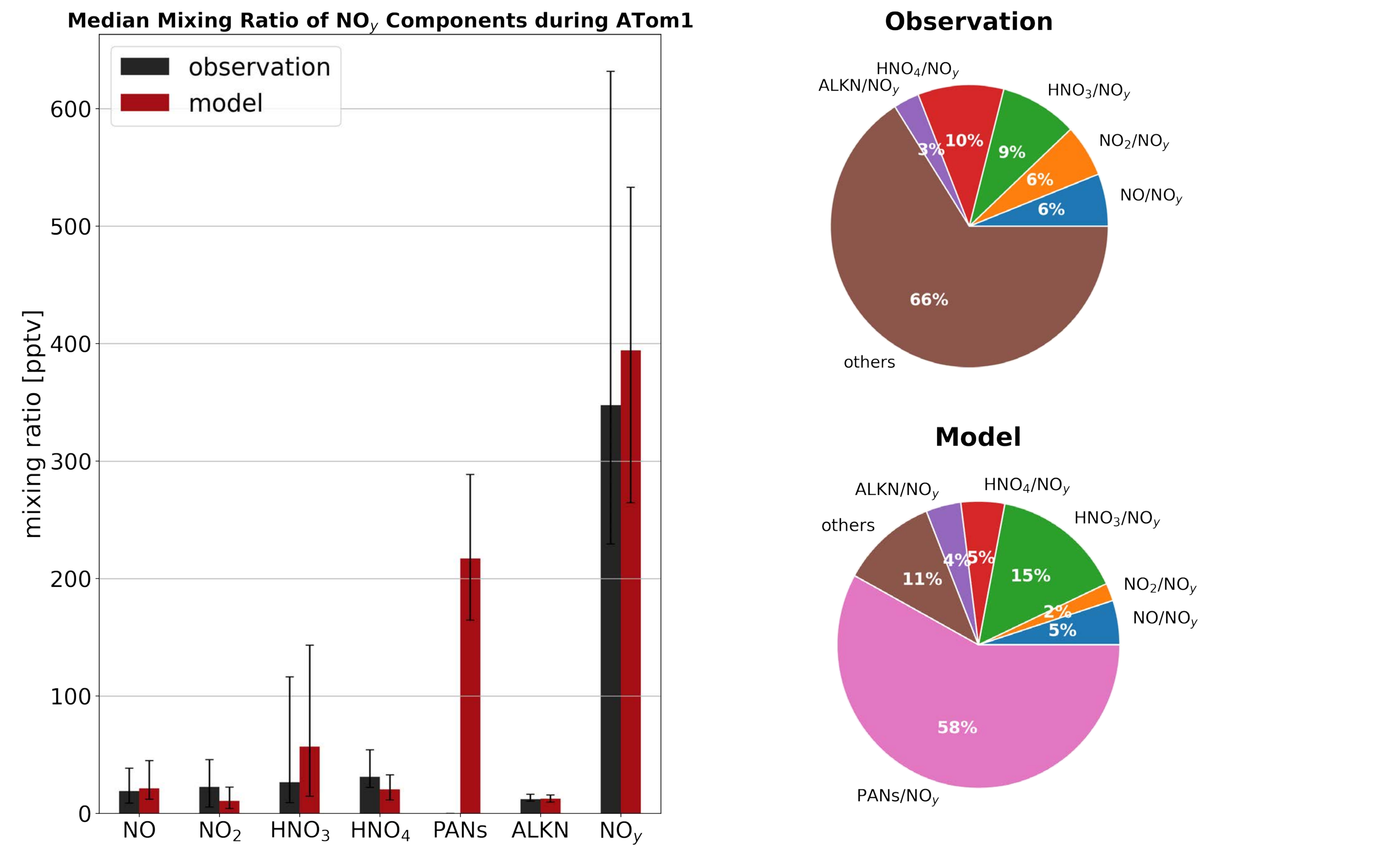

Figure 3: Compare mixing ratio of NO_y and NO_y components and the percentage of each component to NO_y between observation and model during ATom1

- Model underestimated total NO_y for SONEX but observed NO_y has large variability.
- Model underestimated HNO_3 for SONEX measured in 1997.
- coarse instrument at very earlier campaign is one reason but cannot fully explain the large discrepancy.

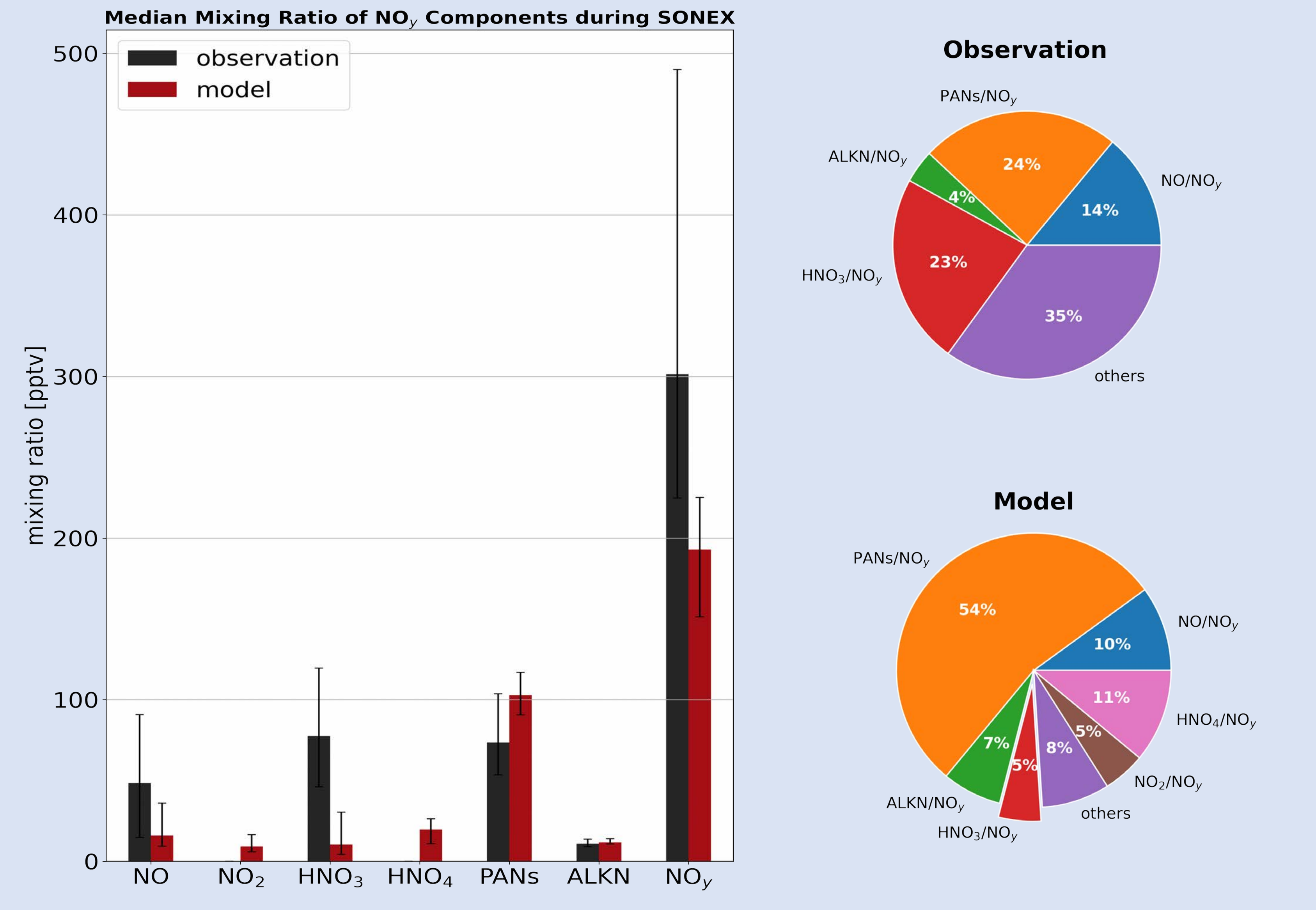

Figure 4: Compare mixing ratio of NO_y and NO_y components and the percentage of each component to NO_y between observation and model during SONEX

5. Concluding Remarks

1. Most total measured reactive nitrogen in the upper troposphere is from a few individual components and DC8 is roughly consistent with MOZAIC climatology during all seasons except winter.
2. Initial model comparison to DC8 suggests the model routinely overestimates NO_y due to a positive bias in HNO_3, as has been reported before (Travis et al., 2020).

6. Next Steps

1. Compare model to other DC8 campaigns.
2. Build the climatology of global UT NO_y.
3. Identify consistent model biases in all campaigns and diagnose error sources.

References

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