Sleuthing Errors in Reactive Nitrogen in the Global Upper Troposphere

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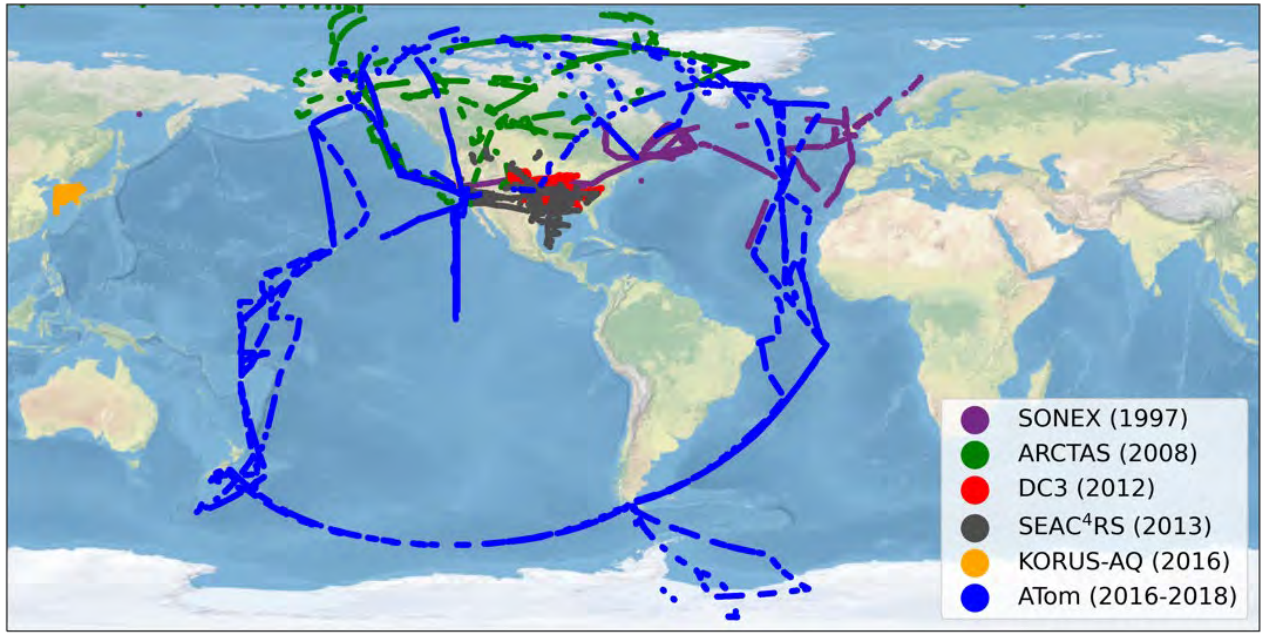
Historic NASA DC8 campaigns combined to achieve near-global coverage of the upper troposphere

Aircraft emission

Biogenic-anthropogenic interactions



Pollution to Arctic

Thunderstorms

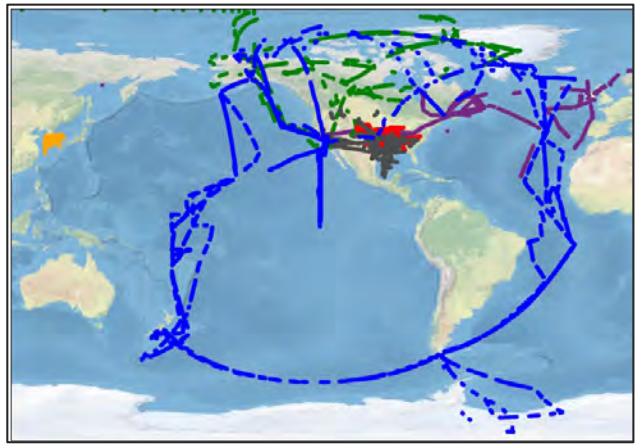
KORUS-AQ

Air quality in South Korea

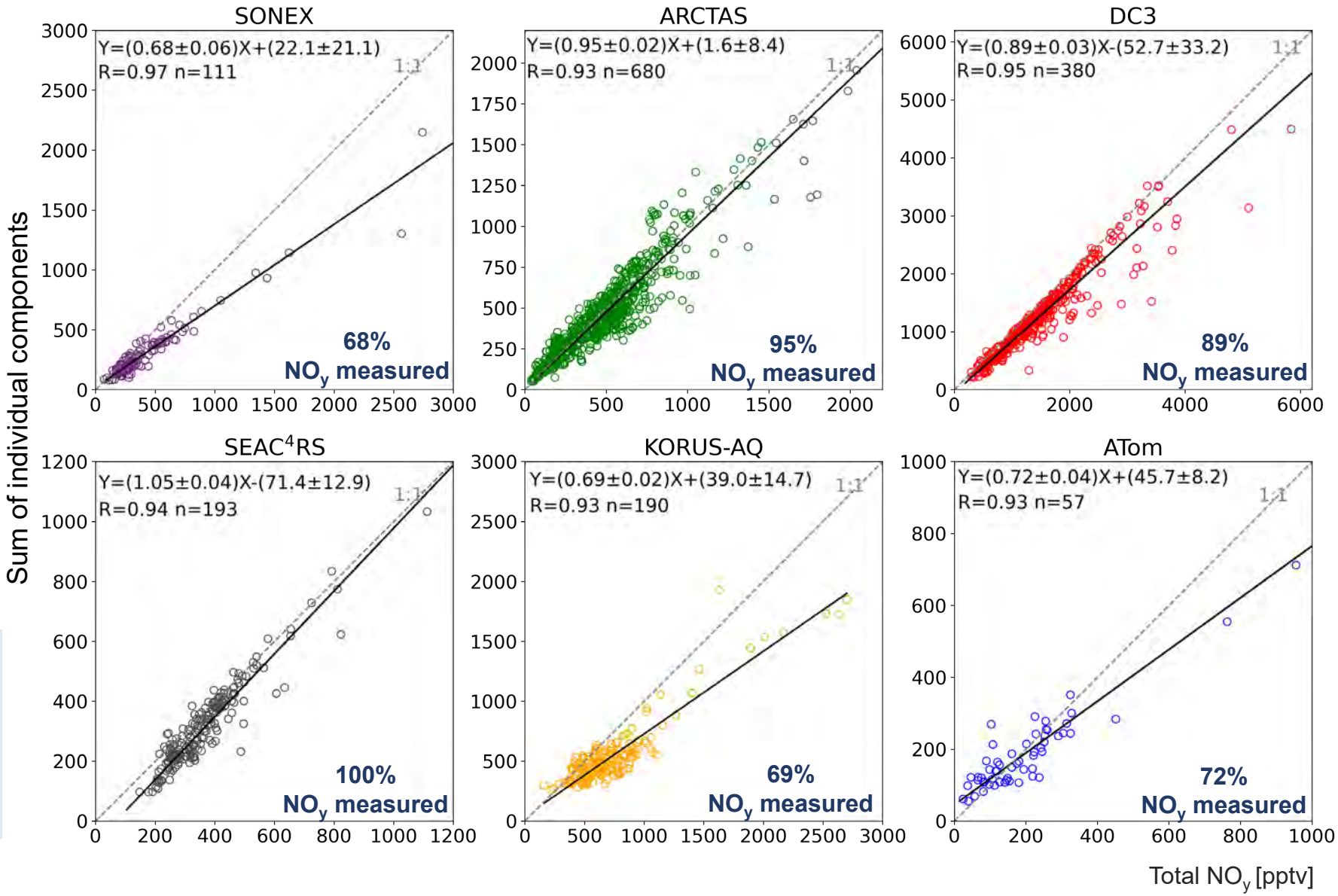
Remote troposphere

The DC8 campaigns extend from SONEX in the North Atlantic in 1997 to ATom covering the northern and southern hemisphere in all seasons from 2016 to 2018.

Proportion of UT NO_y measured during DC8 campaigns

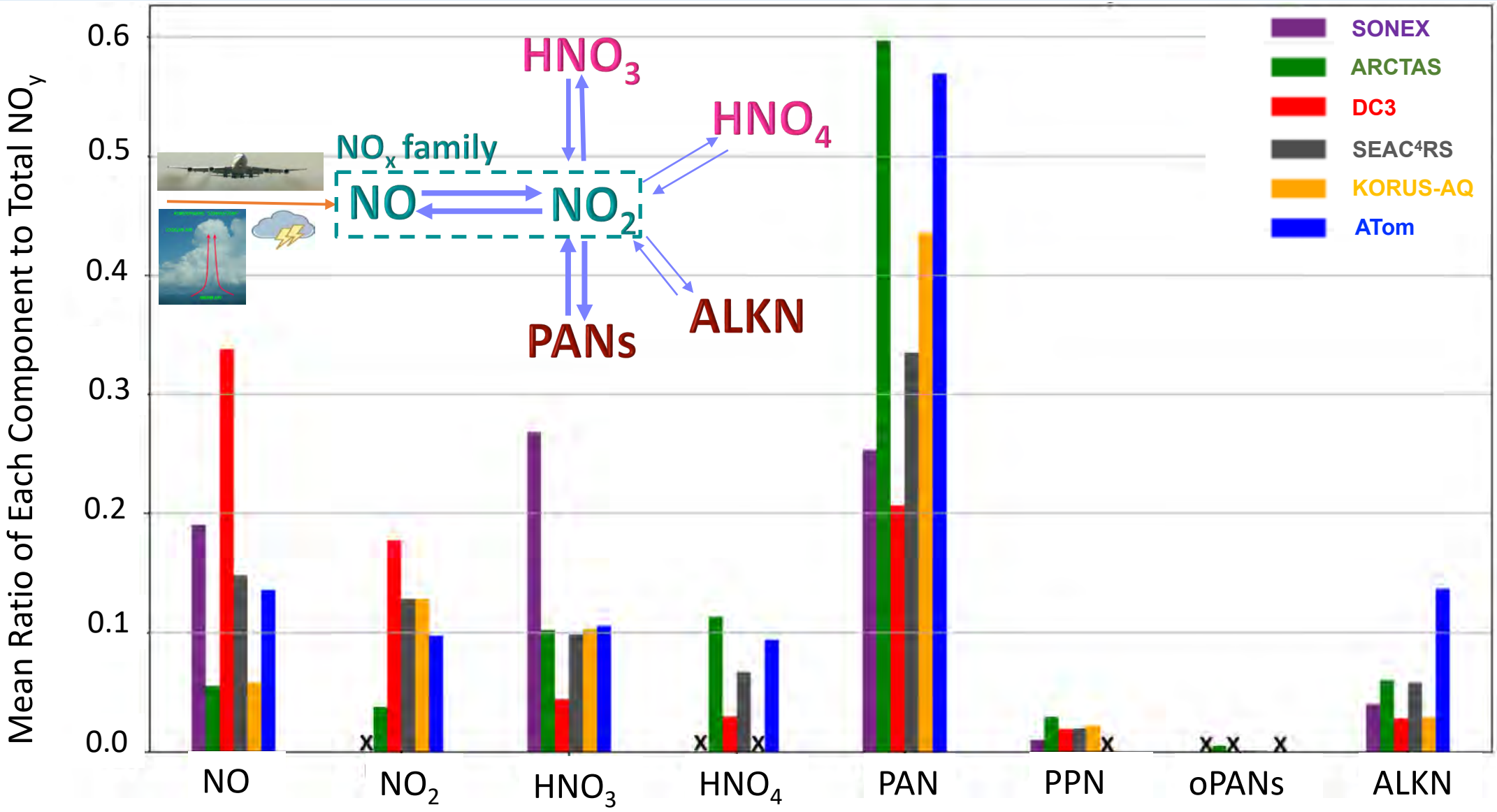


Relationship between sum of individual NO_y components and total NO_y



Most of UT NO_y contributed by a handful of species including NO_x, PANs, HNO_3, HNO_4 and organic nitrates.

Contribution of dominant individual components to total NO_y

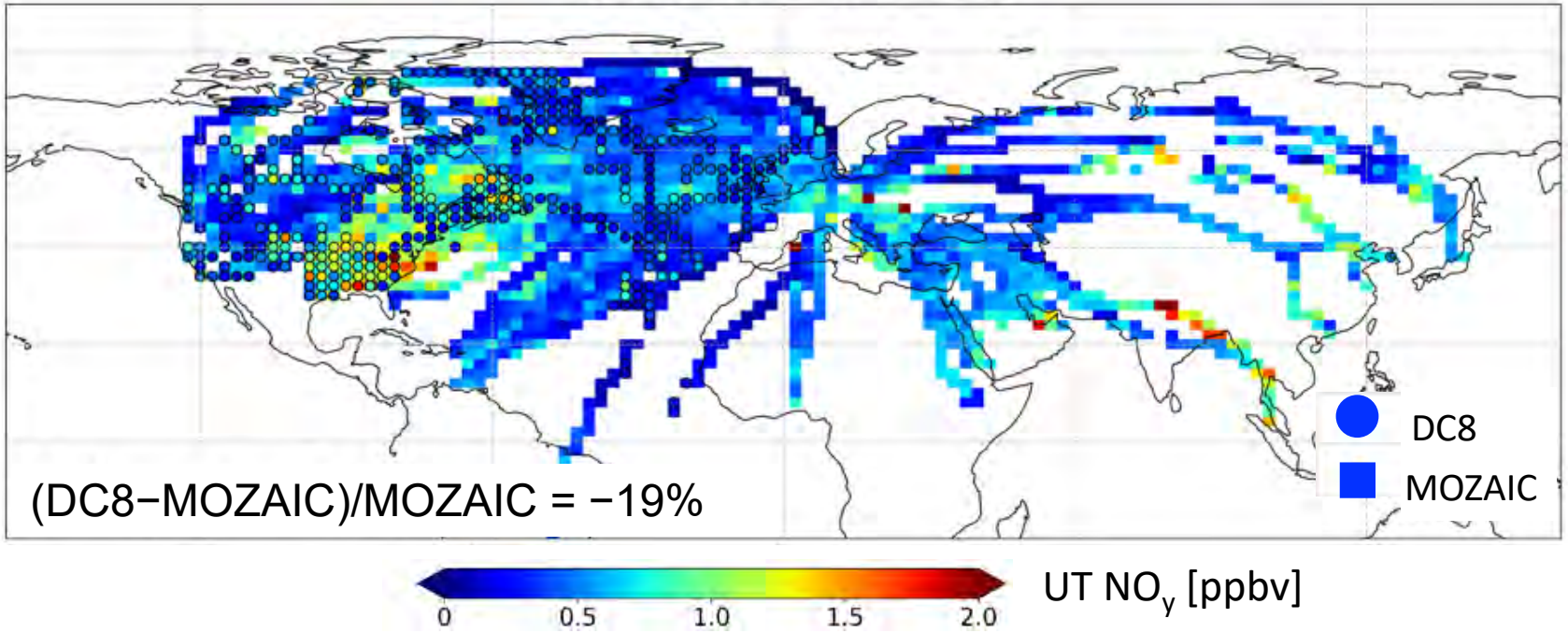


PAN is the dominant NO_y component, ranging from 21% to 60%.

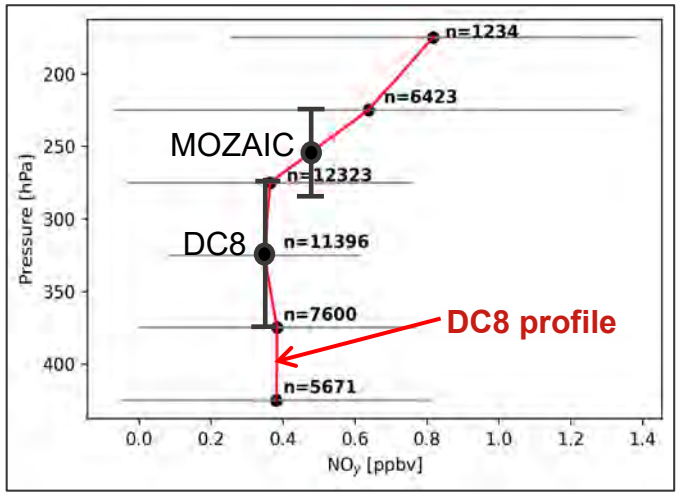
Comparison of short-term DC8 and multiyear commercial aircraft NO_y

We assessed how representative of DC8 UT NO_y to normal condition

Spatial distribution of UT NO_y during DC8 and MOZAIC



Vertical sampling extent of aircraft campaigns



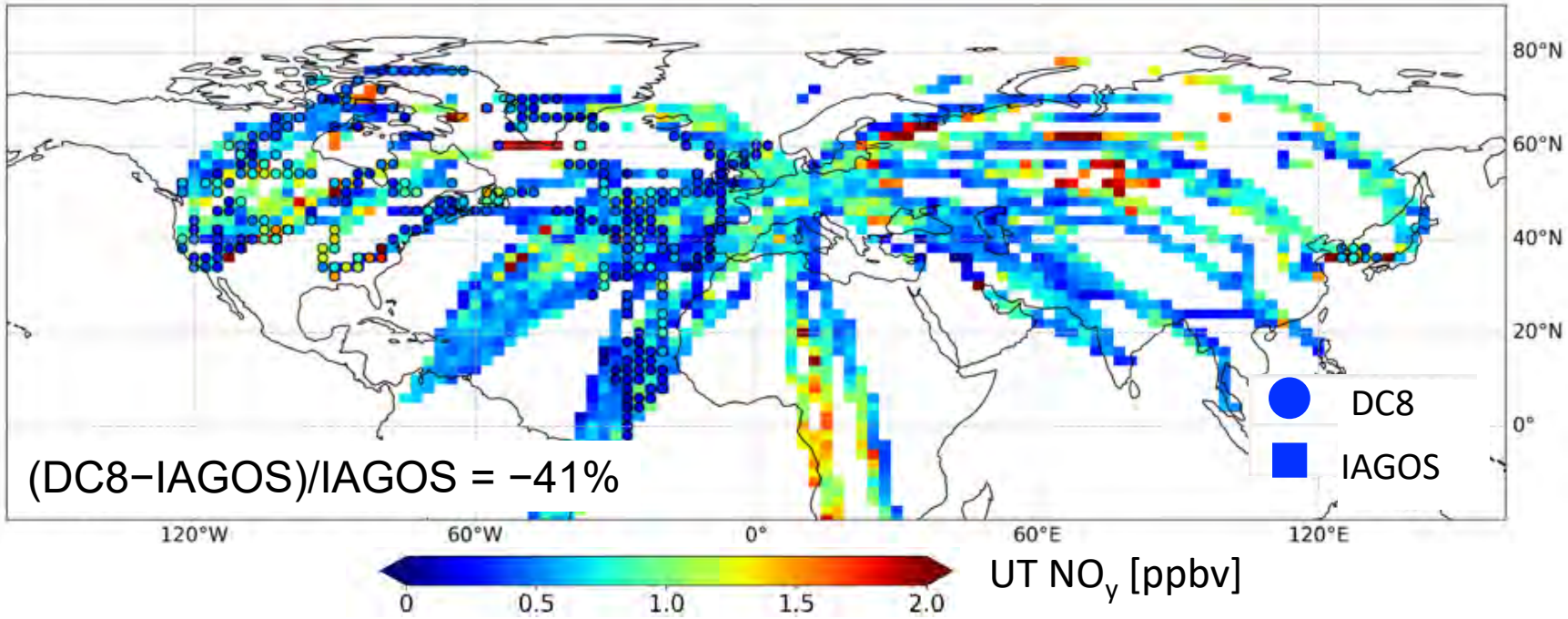
NO_y 22% less at DC8 altitude

DC8 NO_y is about 19% less than MOZAIC and the difference is attributed to the different altitude sampled.

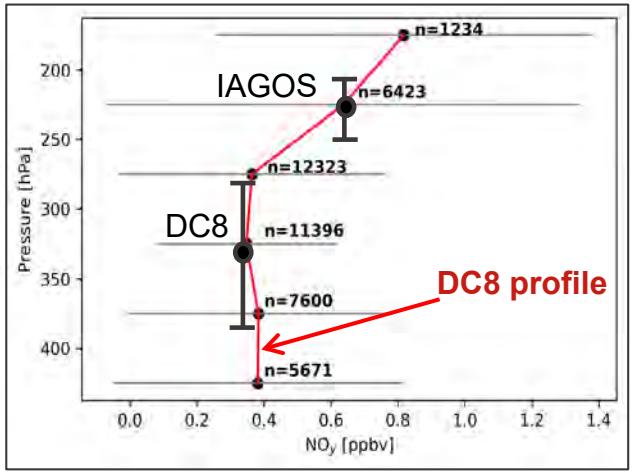
Comparison of short-term DC8 and multiyear commercial aircraft NO_y

We assessed how representative of DC8 UT NO_y to normal condition

Spatial distribution of UT NO_y during DC8 and IAGOS



Vertical sampling extent of aircraft campaigns

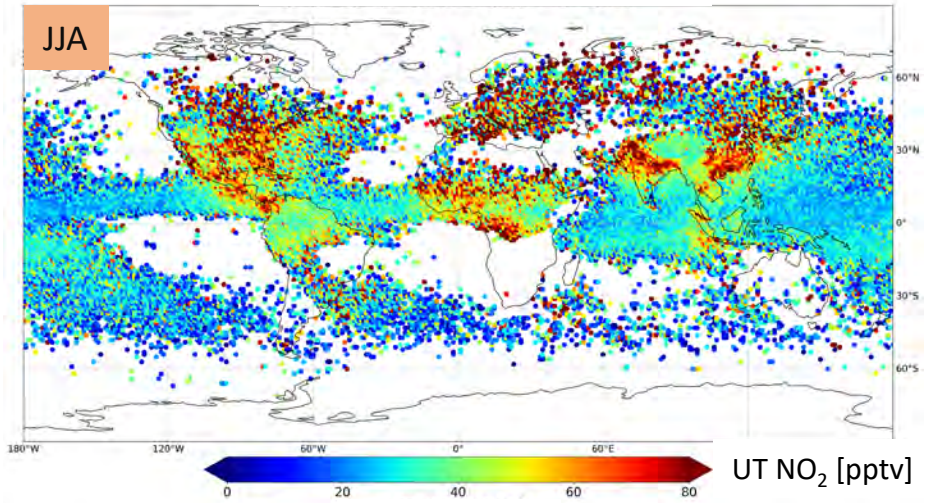


NO_y 41% less at DC8 altitude

DC8 NO_y is 41% less than IAGOS and the difference is attributed to the different altitude sampled.

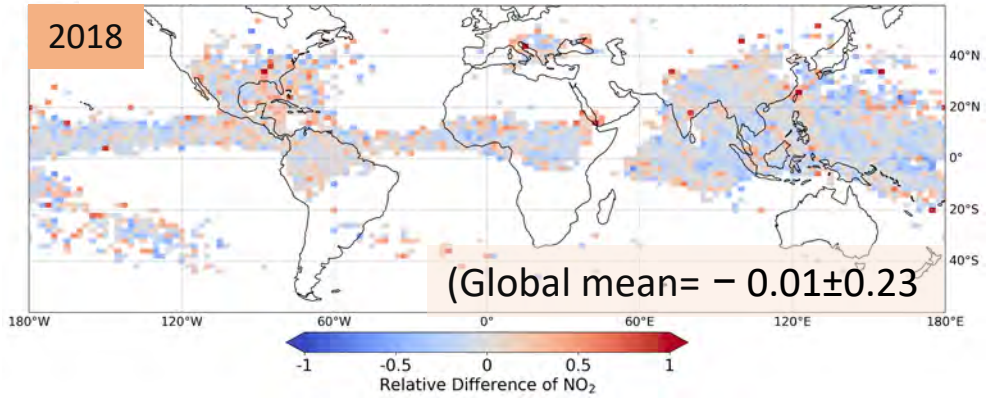
Further assessment of representativeness of single year measurements

Climatology of UT NO_2

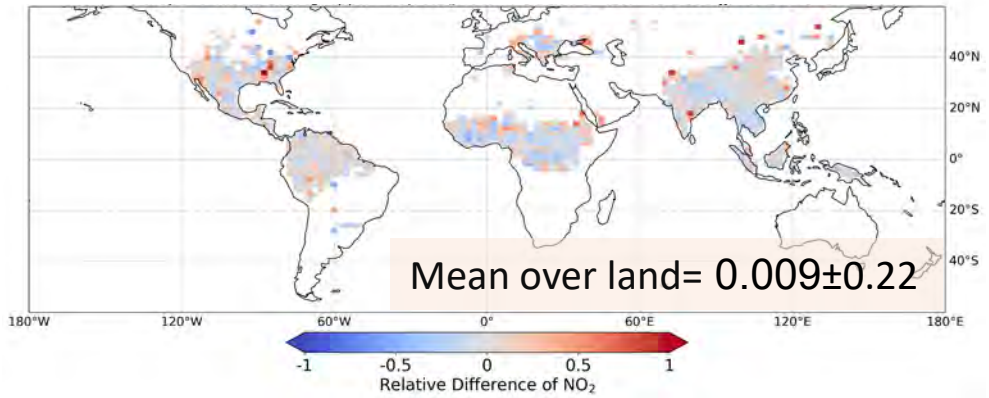


Determined by quantifying interannual variability in UT NO_2 from multiple years (2018-2021) of cloud-sliced TROPOMI from Marais et al. (2021).

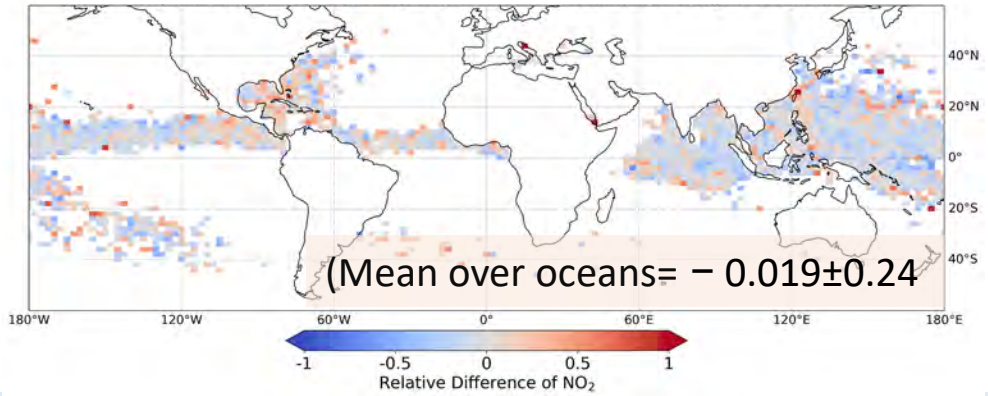
Interannual variability of UT NO_2



Interannual variability of UT NO_2 over land

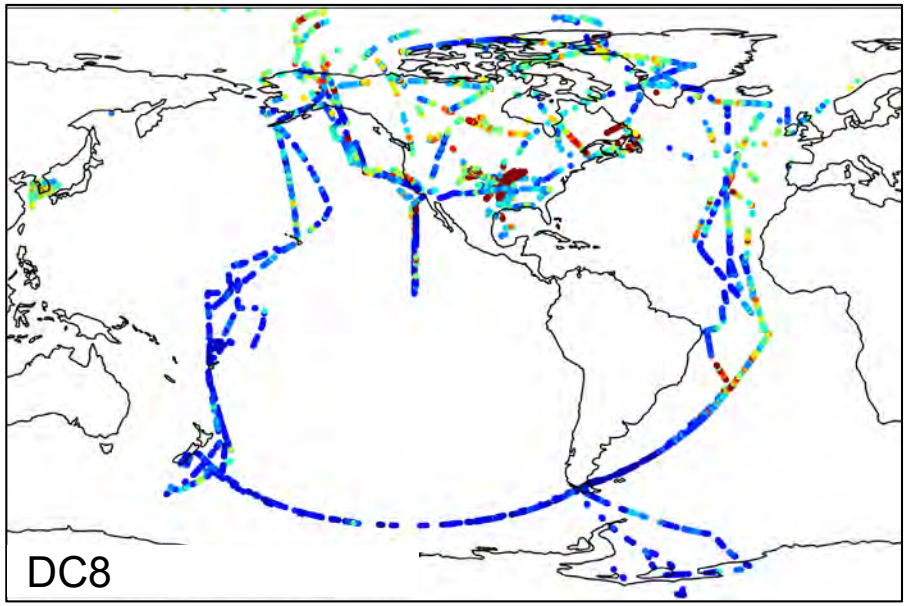


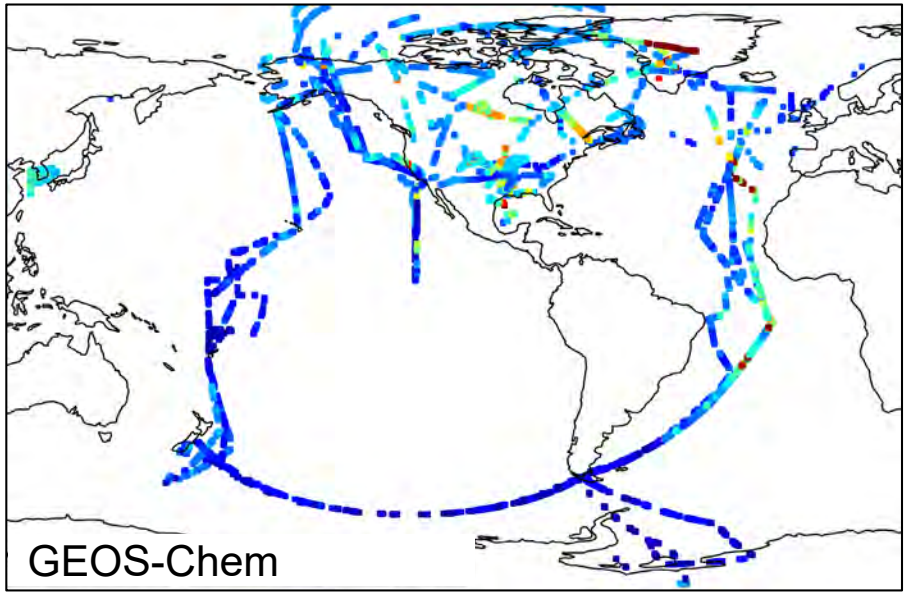
Interannual variability of UT NO_2 over oceans

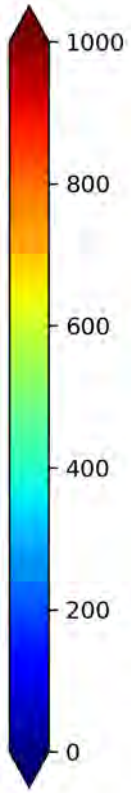


Negligible interannual variability of UT NO_2 gives us confidence to use single year observations of DC8

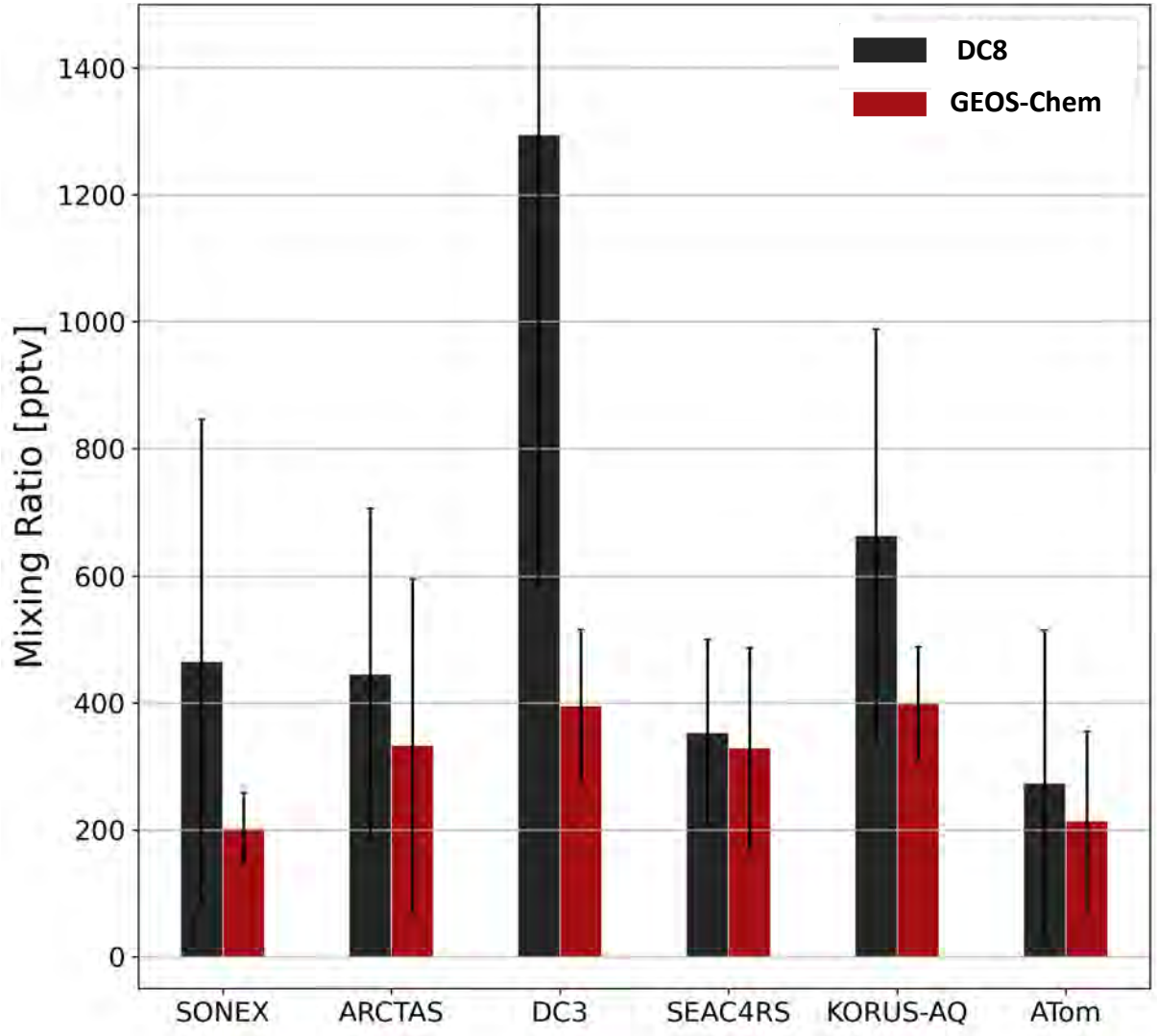
GEOS-Chem skill at simulating UT total NO_y





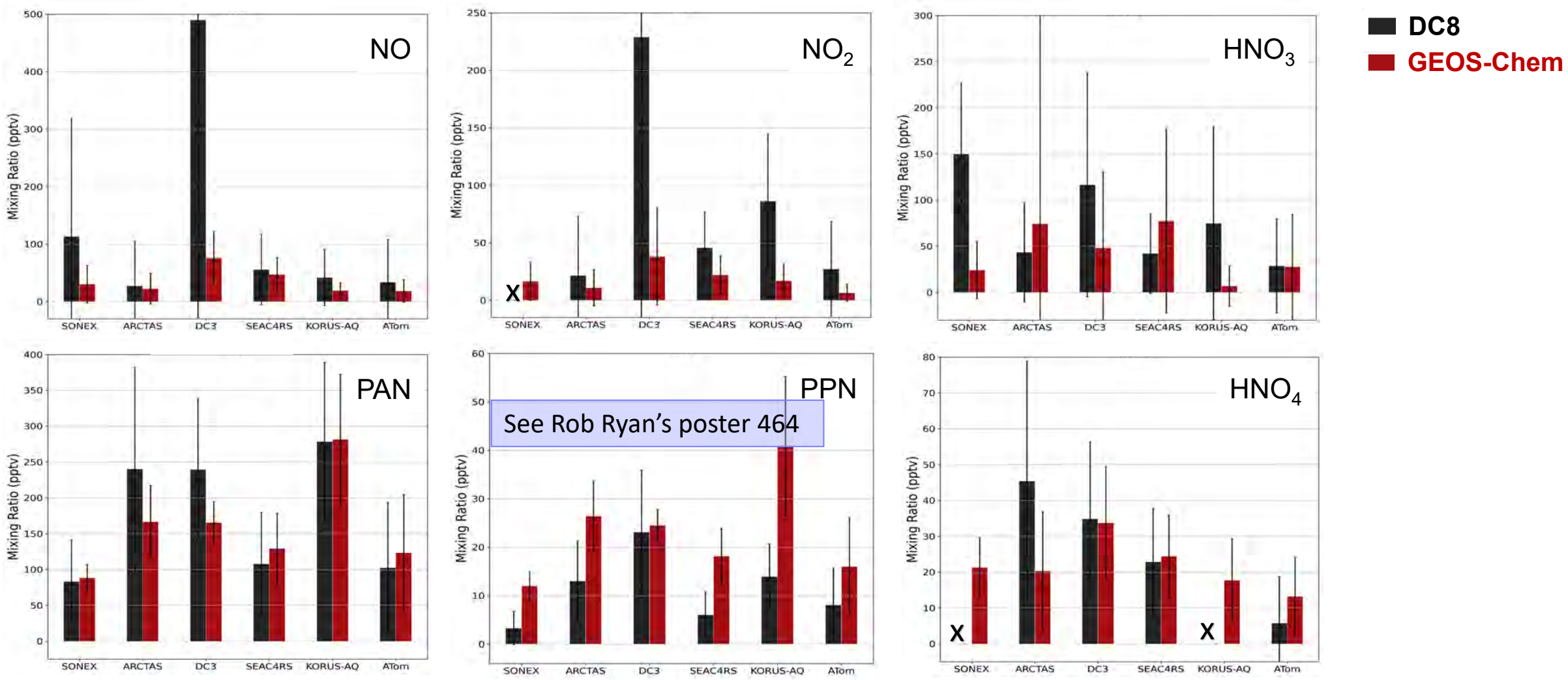


NO_y [pptv]



GEOS-Chem underestimates UT NO_y

GEOS-Chem skill at simulating each UT NO_y component



GEOS-Chem underestimates NO by 5-80 ppt and NO_2 by 11-24 ppt, as it locks up ~10 ppt NO_2 as PPN that is overestimated in the model due to missing loss processes.

Conclusion

- Most total measured reactive nitrogen in the upper troposphere is from a few individual components.
- PAN is the dominant component of upper tropospheric reactive nitrogen globally.
- DC8 UT NOy consistency with MOZAIC and IAGOS and near-negligible interannual variability in UT NO_2 support the use of single-year observations from DC8 for assessing GEOS-Chem.
- GEOS-Chem underestimates all NO_y components, except PPN.
- The model underestimating NO_y implies underestimating ozone.

Any Questions? Please contact Nana Wei (nana.wei.21@ucl.ac.uk) @NanaWei_Nevaeh