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Challenging our understanding of upper tropospheric NO_x

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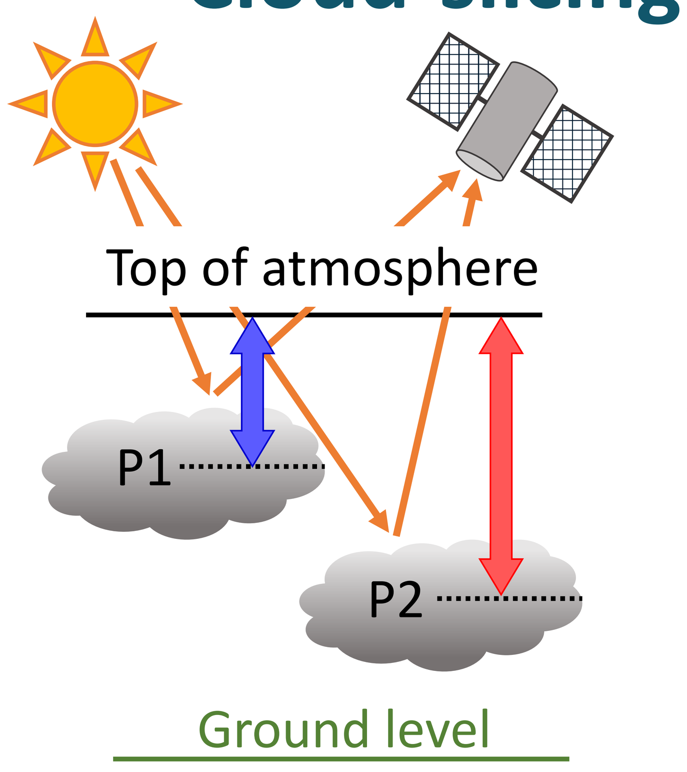
Cloud-slicing of satellite data



$$NO_2\ vmr = \frac{\Delta VCD}{\Delta P} \times \frac{k_b g}{R_{air}}$$

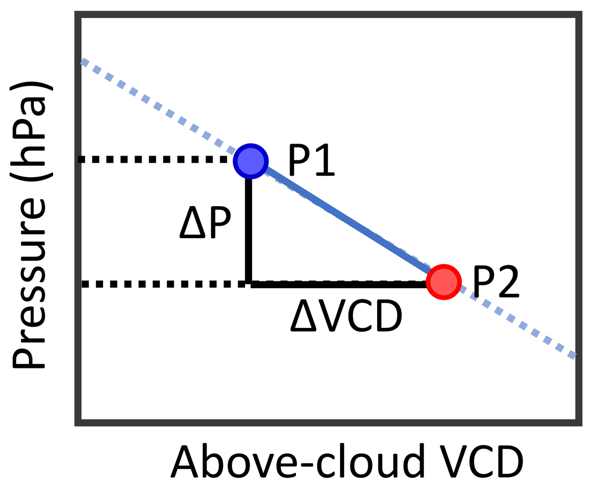
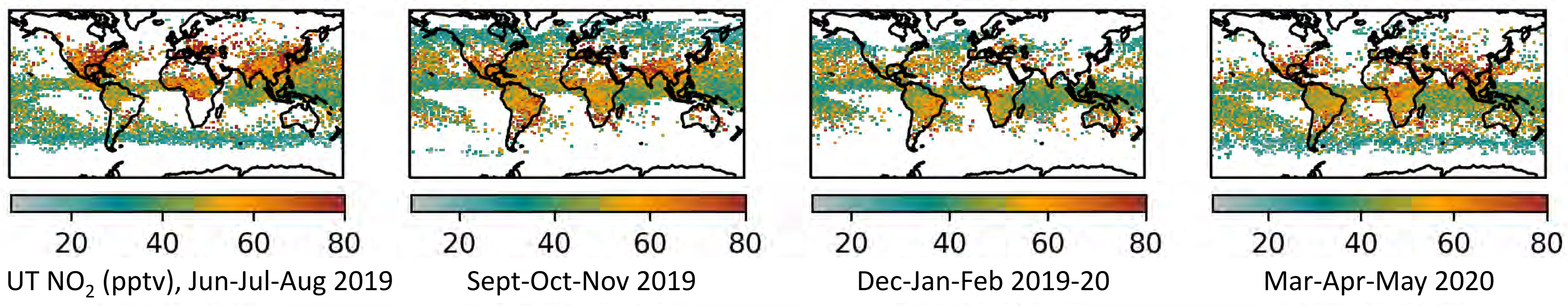
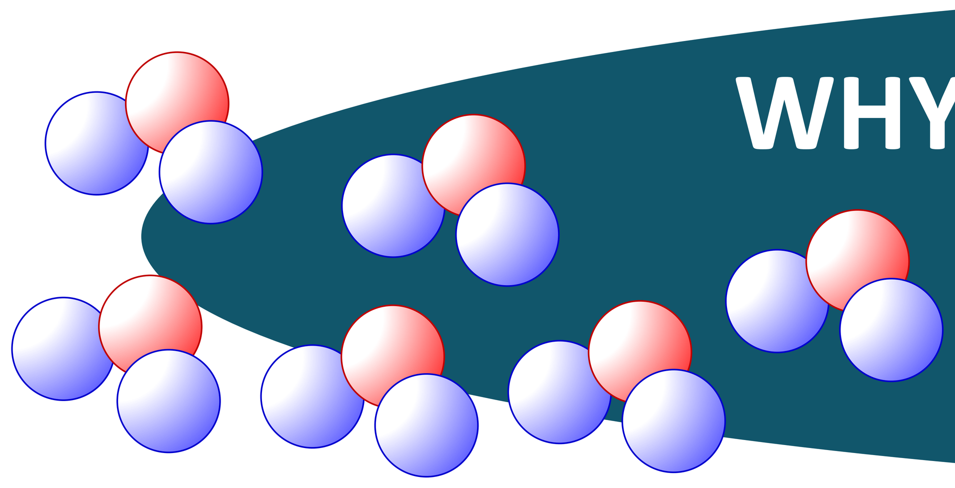


Figure adapted from Choi et al., ACP, 2014

Importance of the upper troposphere (UT)

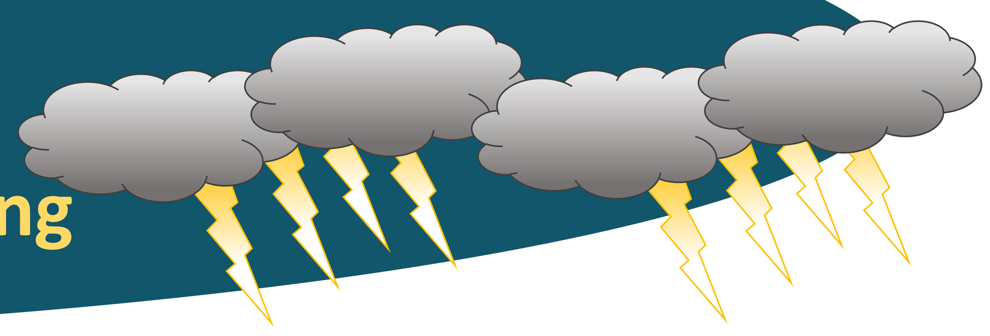
- Upper tropospheric (~450-180 hPa) nitrogen oxides ($NO + NO_2 \equiv NO_x$) are especially long-lived. This allows UT NO_x to play a key role in the chemistry of tropospheric O_3, an important greenhouse gas.
- UT NO_x has natural (lightning) and anthropogenic (aircraft) sources
- UT reactive nitrogen chemistry is controlled by cycling between NO_x and its reservoir compounds (HNO_4, HNO_3, PAN, & PPN). Cycling rates in this cold, low-pressure part of the atmosphere are uncertain.
- UT NO_x can be measured in-situ by aircraft, but these measurements are subject to interferences
- Cloud-slicing total columns of NO_2 from TROPOMI provides near-global coverage of NO_2 in the UT in each season





WHY DO MODELS UNDERESTIMATE UPPER TROPOSPHERIC NO_2?

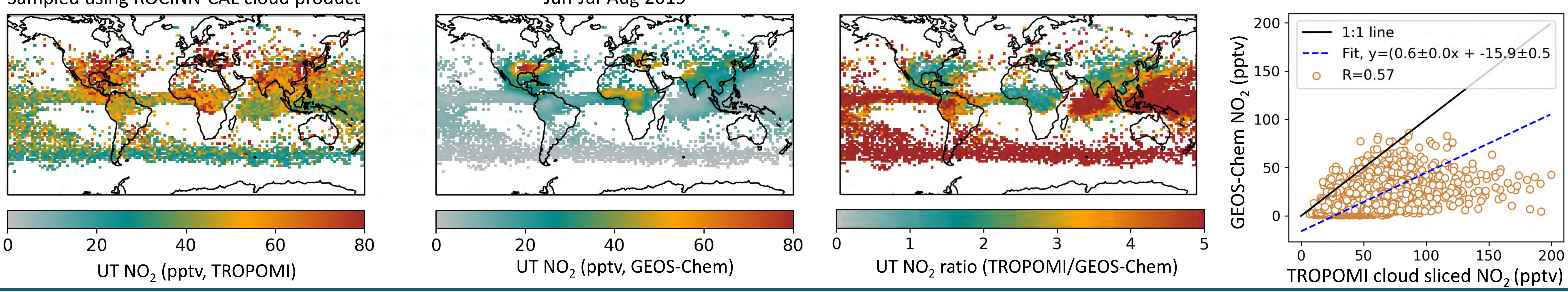
We look at the role of chemistry and lightning



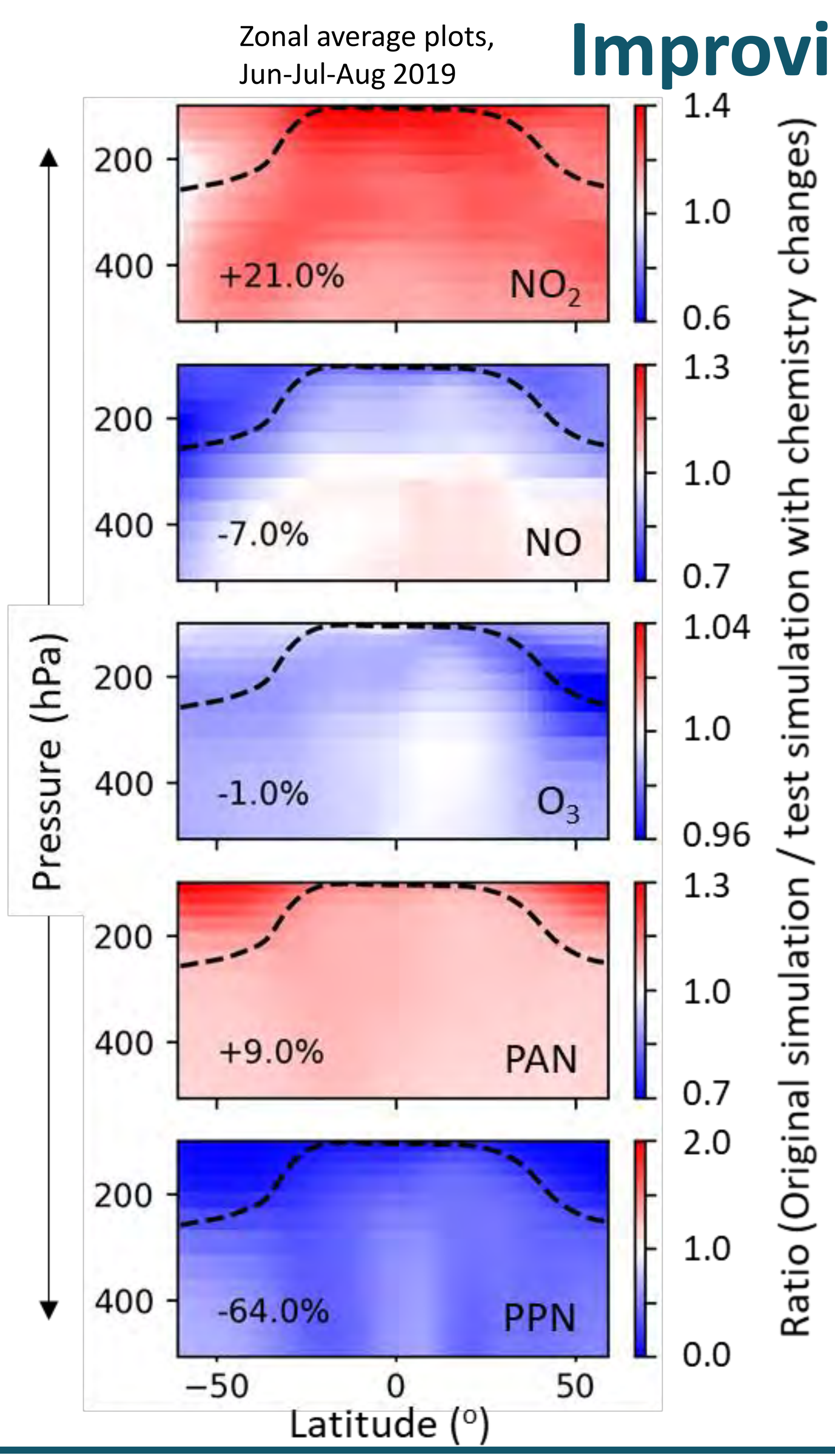
Original GEOS-Chem vs TROPOMI UT Comparison

TROPOMI cloud sliced, Jun-Jul-Aug 2019
Sampled using ROCINN-CAL cloud product

GEOS-Chem results
Jun-Jul-Aug 2019



Improving UT chemistry scheme

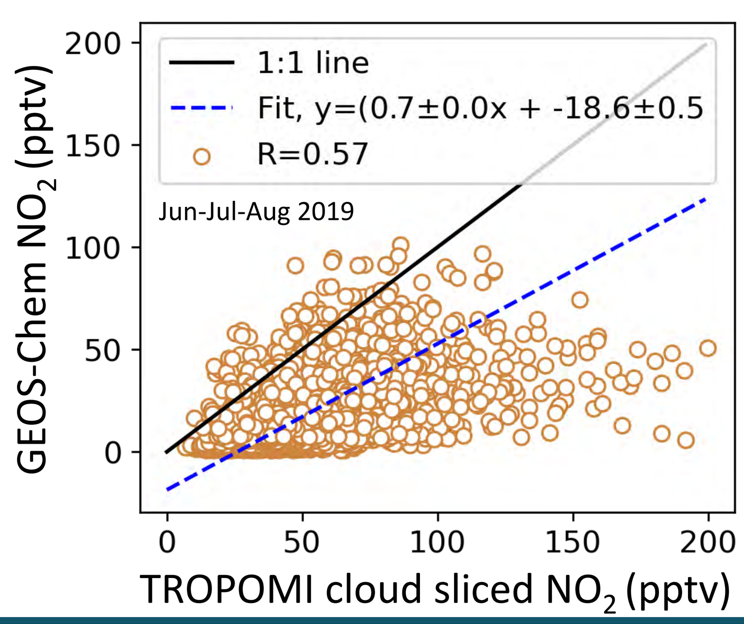


Reaction rates under UT conditions have been updated based on aircraft observations of reactive nitrogen:

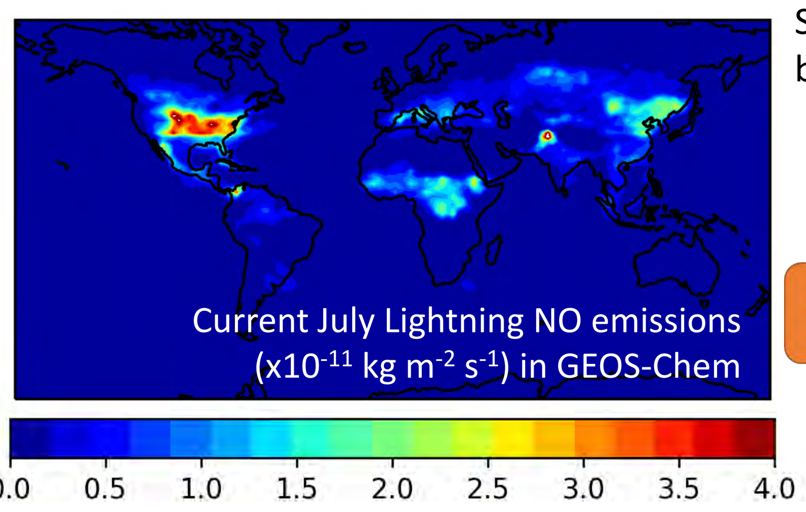
- Speed up $NO + O_3 \rightarrow NO_2 + O_2$
- Slow down $NO_2 + OH \rightarrow HNO_3$
- Slow down $NO_2 + HO_2 \rightarrow HNO_4$
- Add sinks (OH oxidation and photolysis) for peroxypropionyl nitrate (PPN)

Key results:

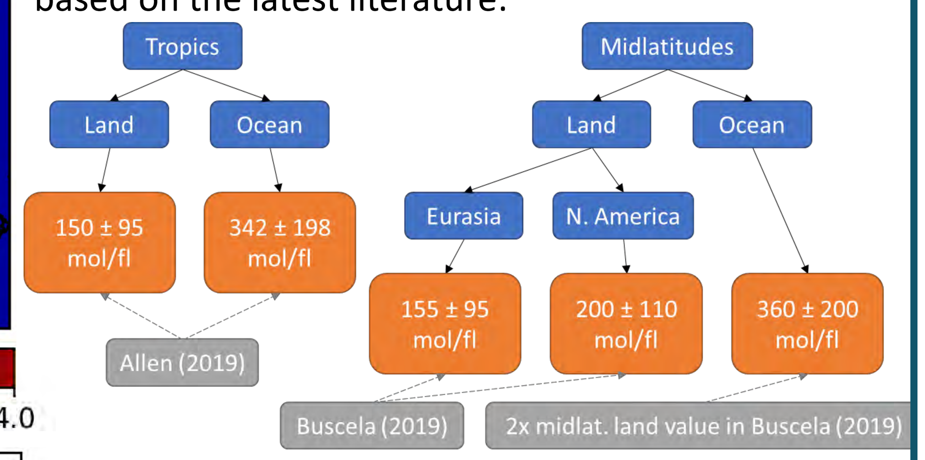
- UT PPN decreased by 64 %
- UT O_3 unchanged
- UT NO_2 increased by 21 %, slight improvement in GEOS-Chem vs TROPOMI comparison slope (0.6 → 0.7).

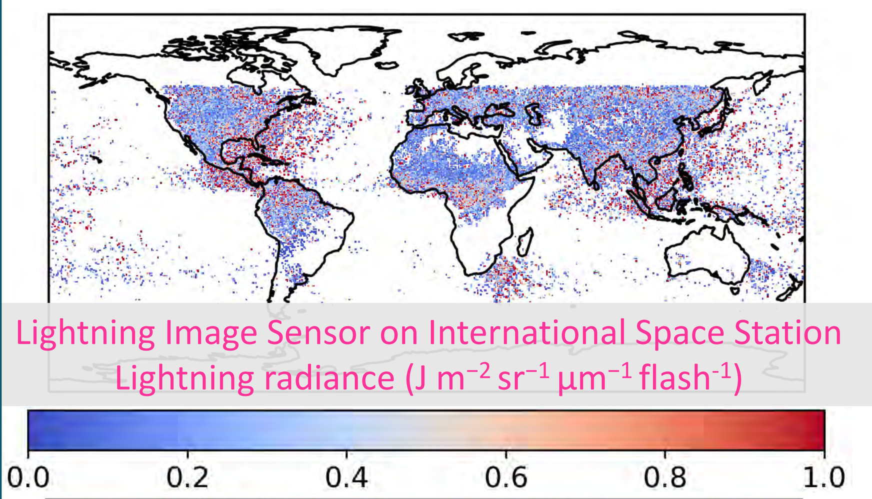


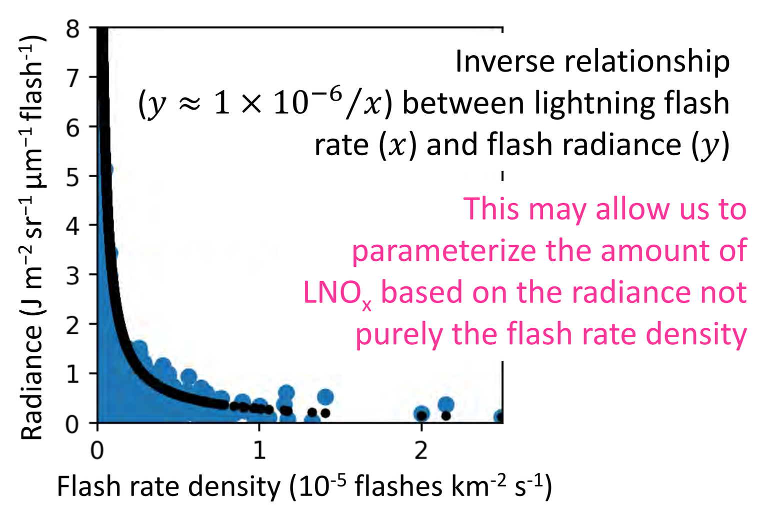
Improving lightning NO_x (LNO_x) scheme



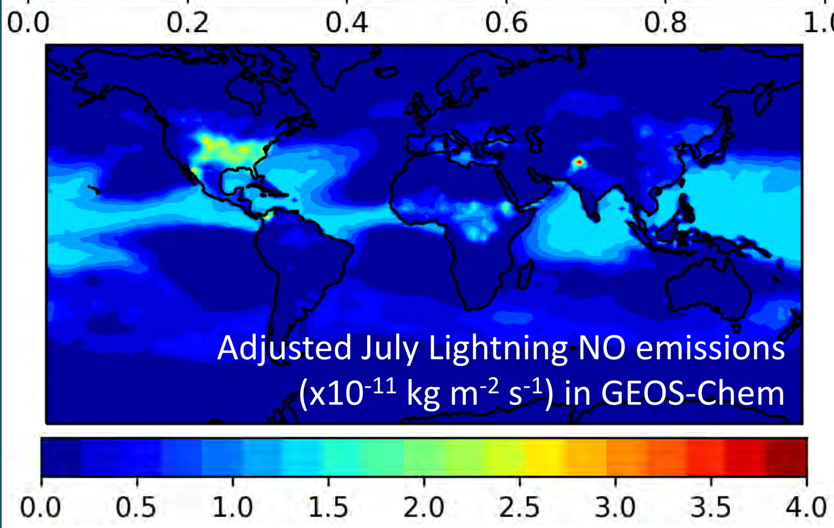
Suggested update to per-flash LNO_x parameterization based on the latest literature:



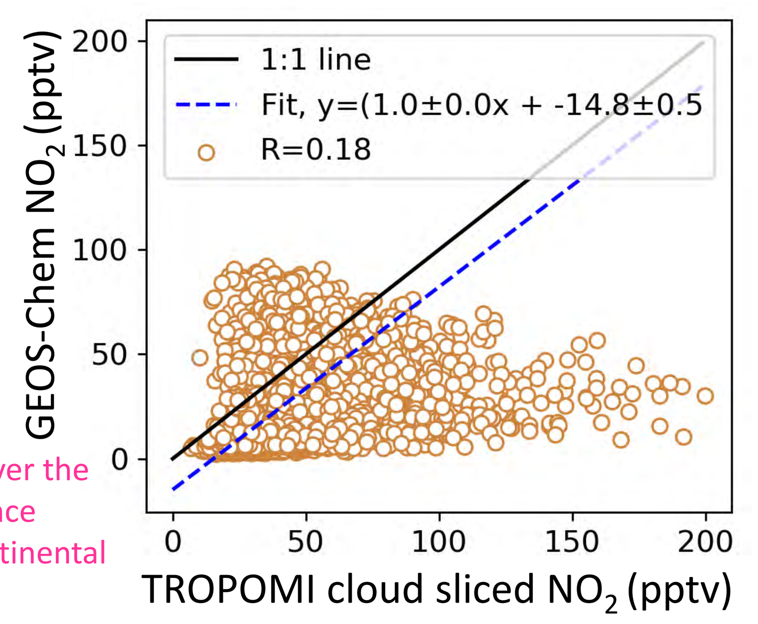




This may allow us to parameterize the amount of LNO_x based on the radiance not purely the flash rate density



In this experiment we target the underestimation of LNO_x over the oceans by parameterizing LNO_x over the oceans using radiance rather than flash rate. This still leaves significant remote continental areas of UT NO_2 underestimated in GEOS-Chem.



Summary

GEOS-Chem underestimates TROPOMI cloud sliced UT NO_2 by ~40 %.

We account for 21% of this by updating the chemistry scheme, especially for the reservoir compound PPN. Large discrepancies between TROPOMI and GEOS-Chem coincide with high lightning radiance, which is inversely related to flash rate. We are currently experimenting with LNO_x parameterization by radiance rather than flash rate.

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