Addressing model uncertainty in upper tropospheric NO_x

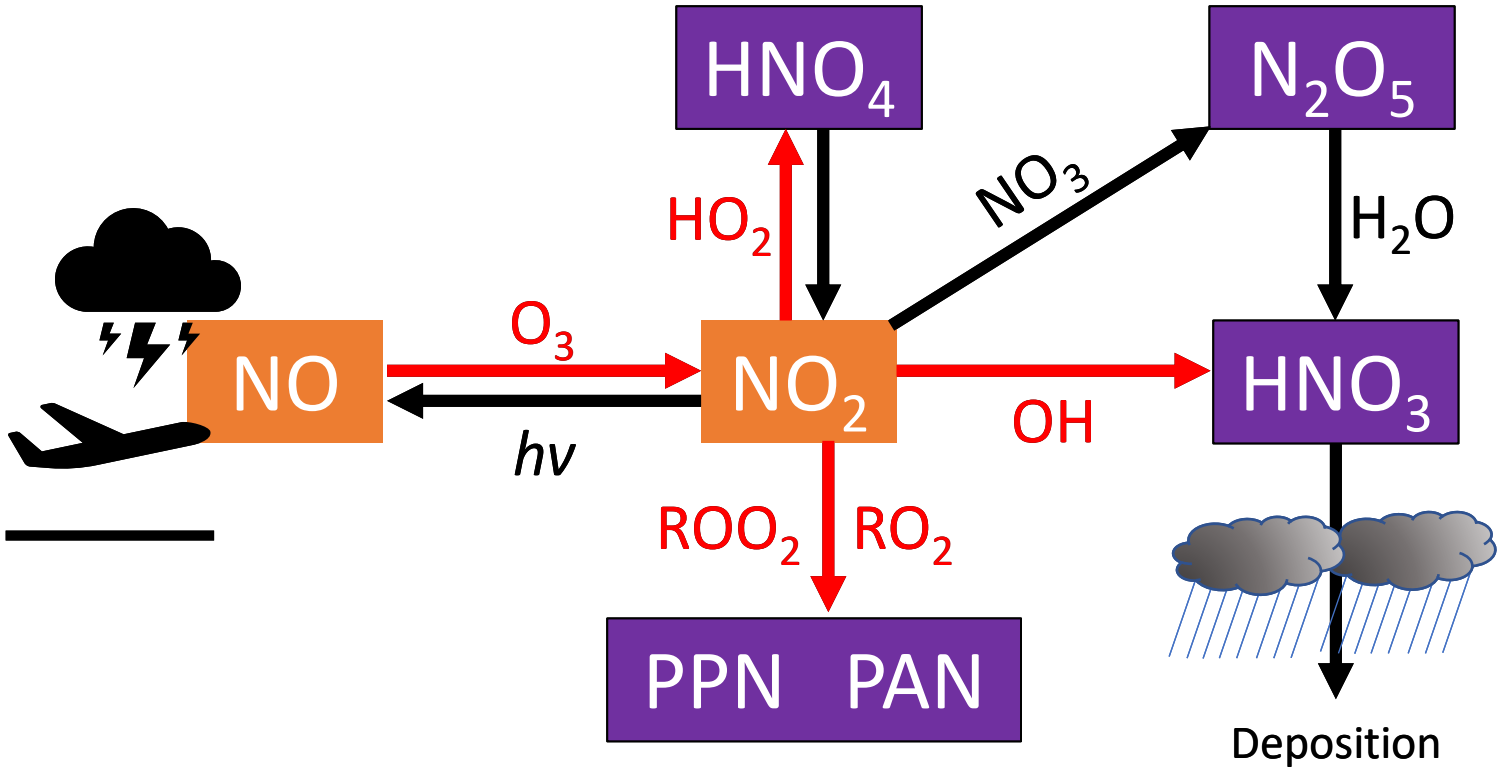
Robert Ryan, Eloise Marais, Nana Wei
University College London

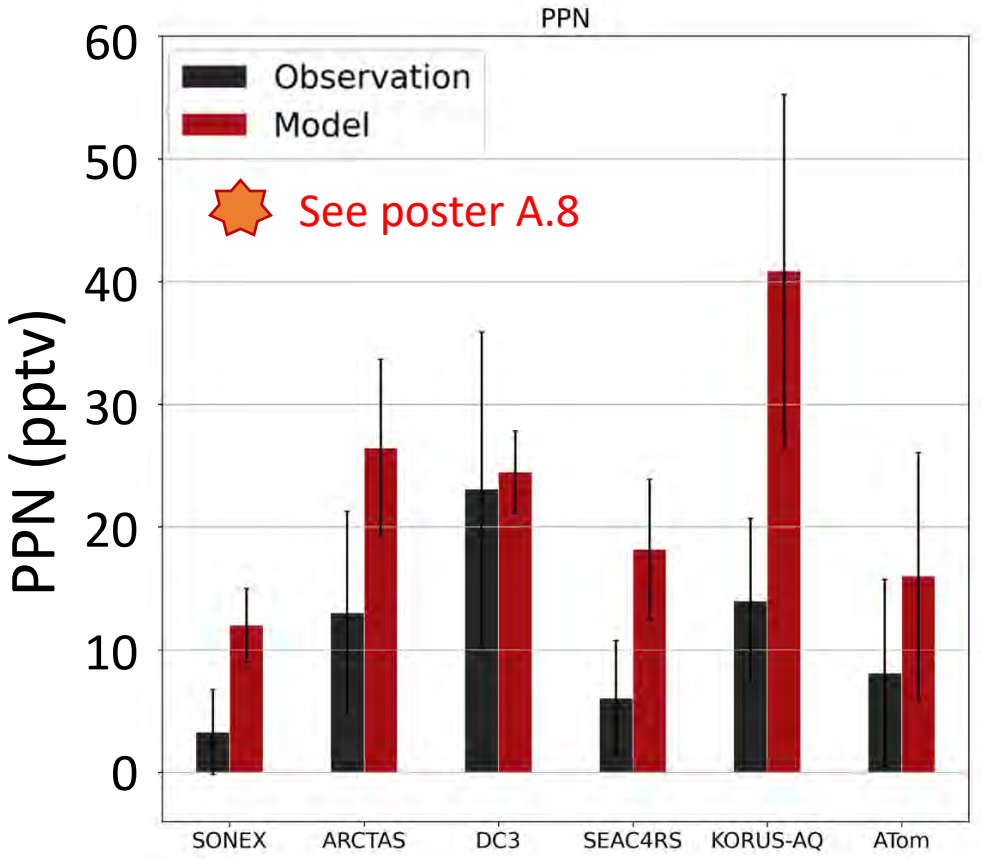
UCL

IGC10
St Louis
2022

GEOS-Chem

Controls on upper tropospheric NO_x





Aircraft campaigns

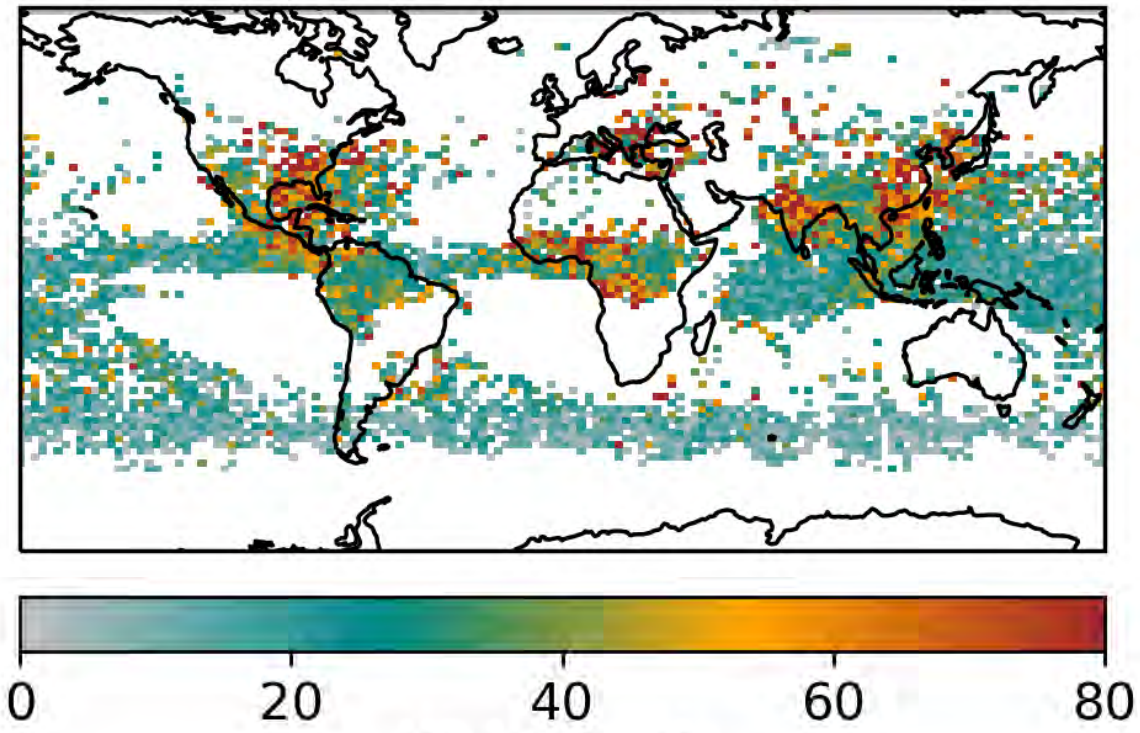
$NO + O_3 \rightarrow NO_2 + O_2$ (Rate too slow: Silvern et al., 2018)

$NO_2 + OH \rightarrow HNO_3$ (Rate too fast: Henderson et al., 2012, Nault et al., 2016)

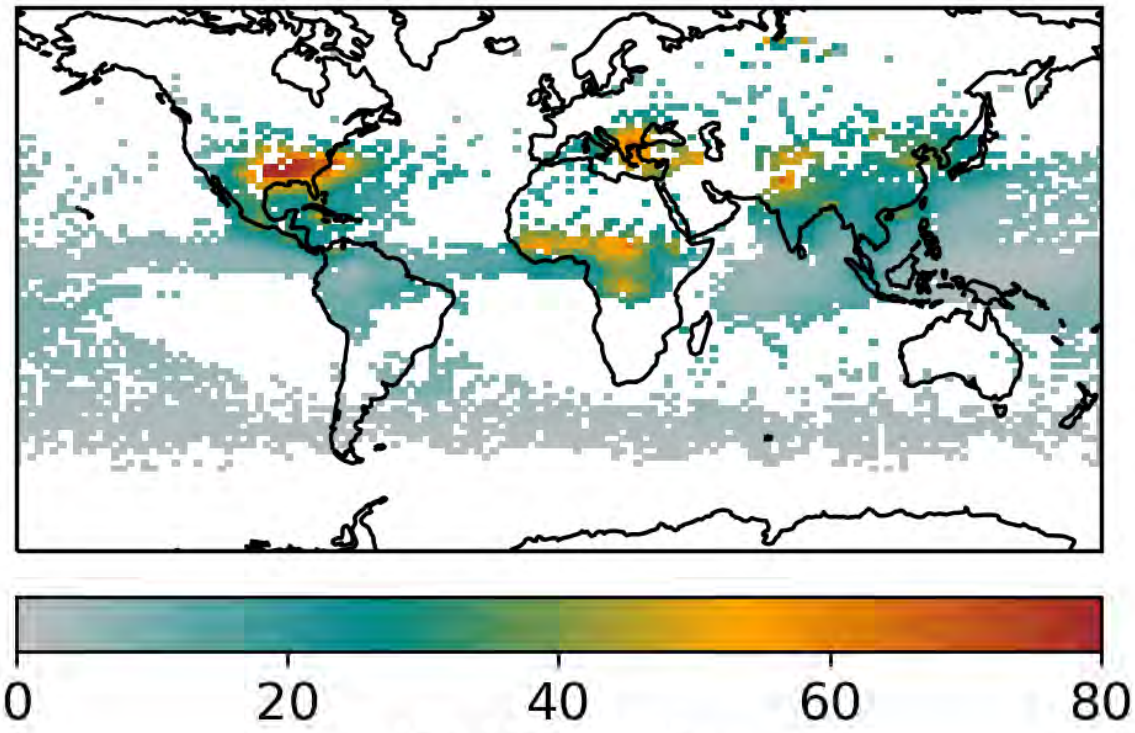
$NO_2 + HO_2 \rightarrow HNO_4$ (Rate too fast: Nault et al., 2016)

GEOS-Chem vs TROPOMI

TROPOMI Cloud-sliced 450-180 hPa
ROCINN-CAL cloud product (clouds as layers)
June-July-August 2019

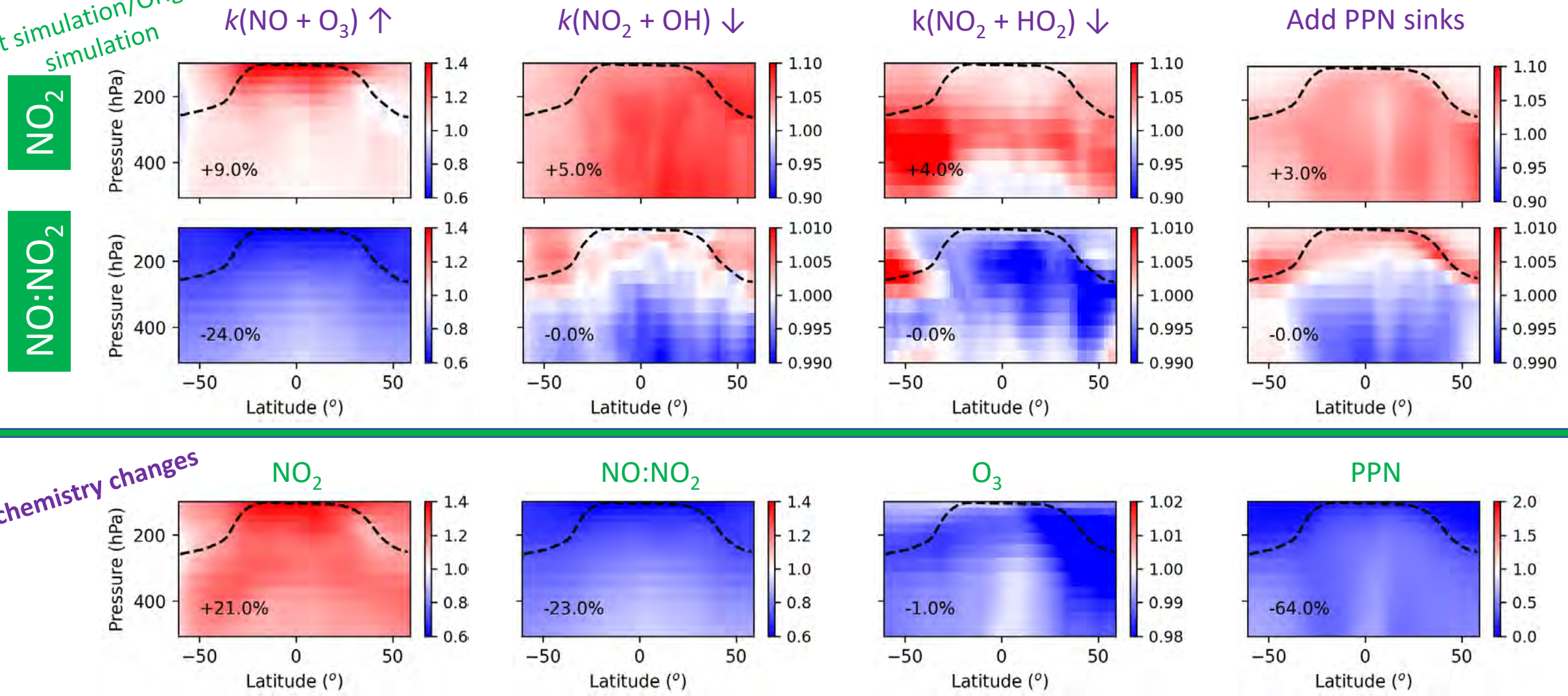


GEOS-Chem v13.3.4, 2° x 2.5°
June-July-August 2019



Improved kinetics for UT NO_x cycling

Test simulation/Original simulation



All chemistry changes

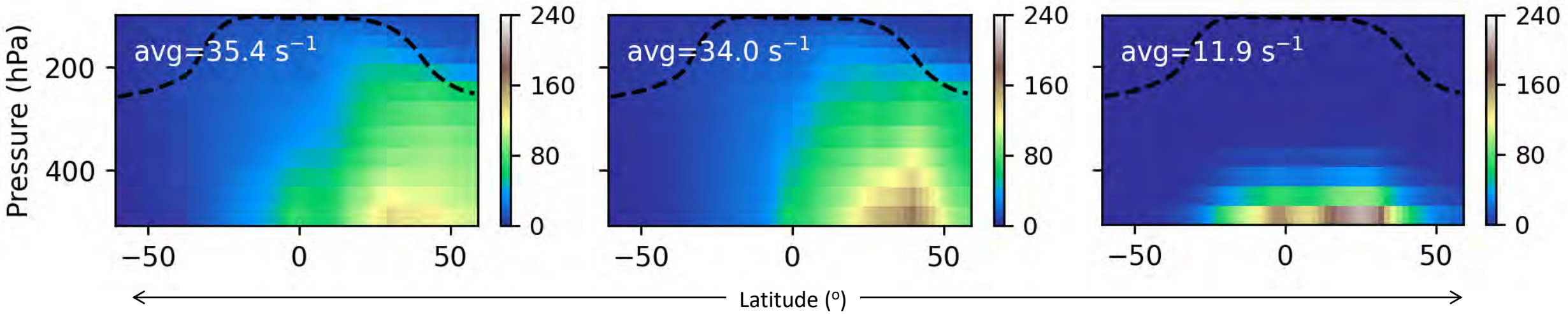
Focusing on PPN

Above 500 hPa

PPN photolysis

PPN+OH

Thermal decomposition



Whole troposphere

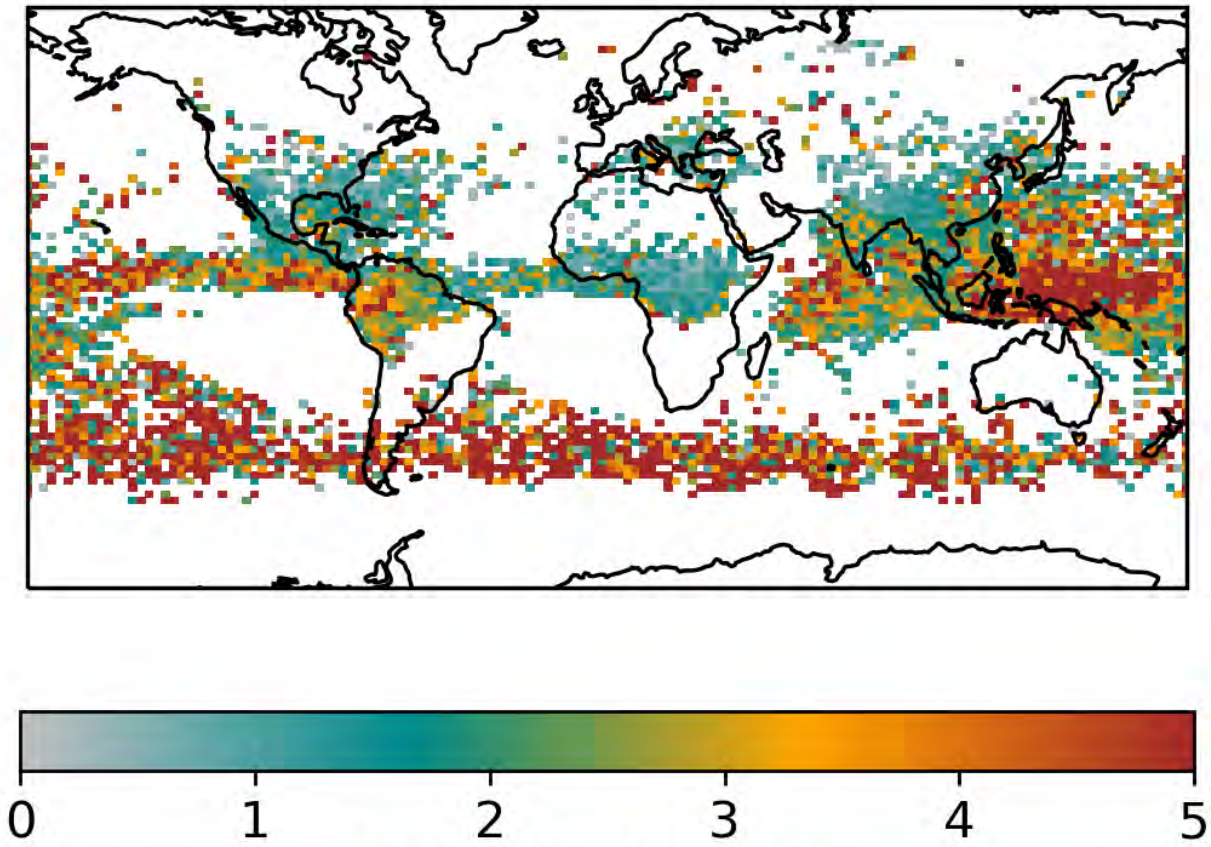
60 s^{-1}

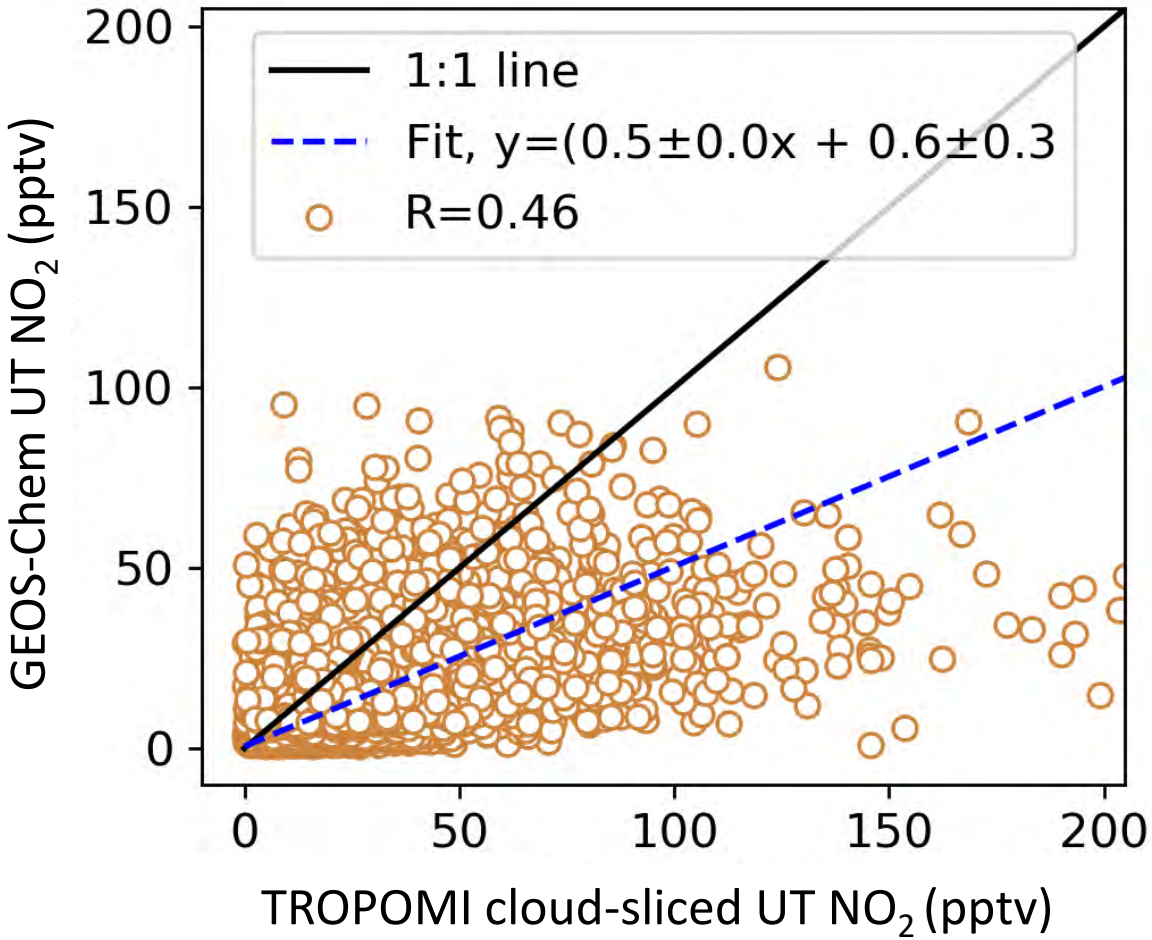
75 s^{-1}

8x10^3 s^{-1}

Recommendation: PPN+OH and PPN photolysis should be included in GEOS-Chem

GEOS-Chem vs TROPOMI

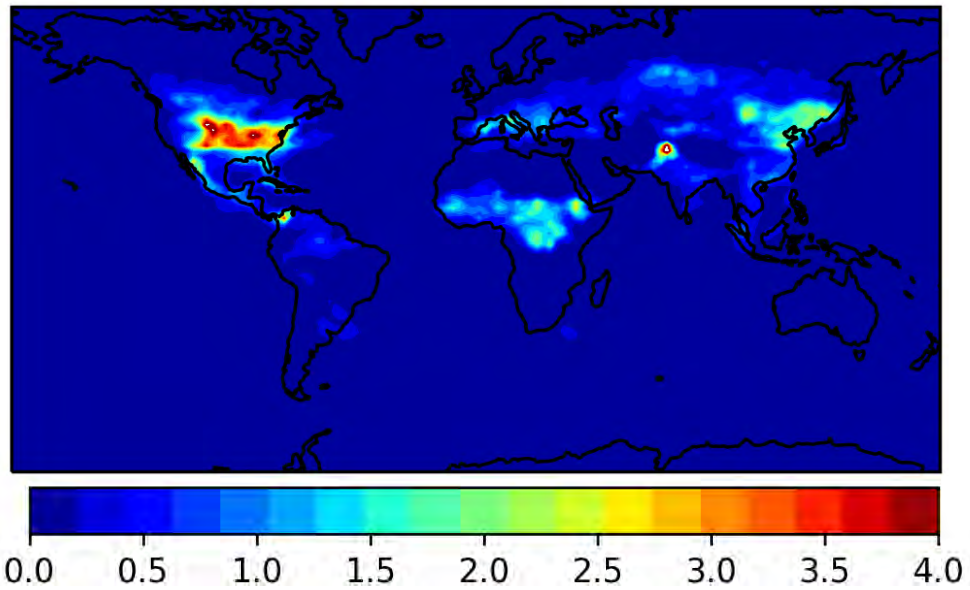




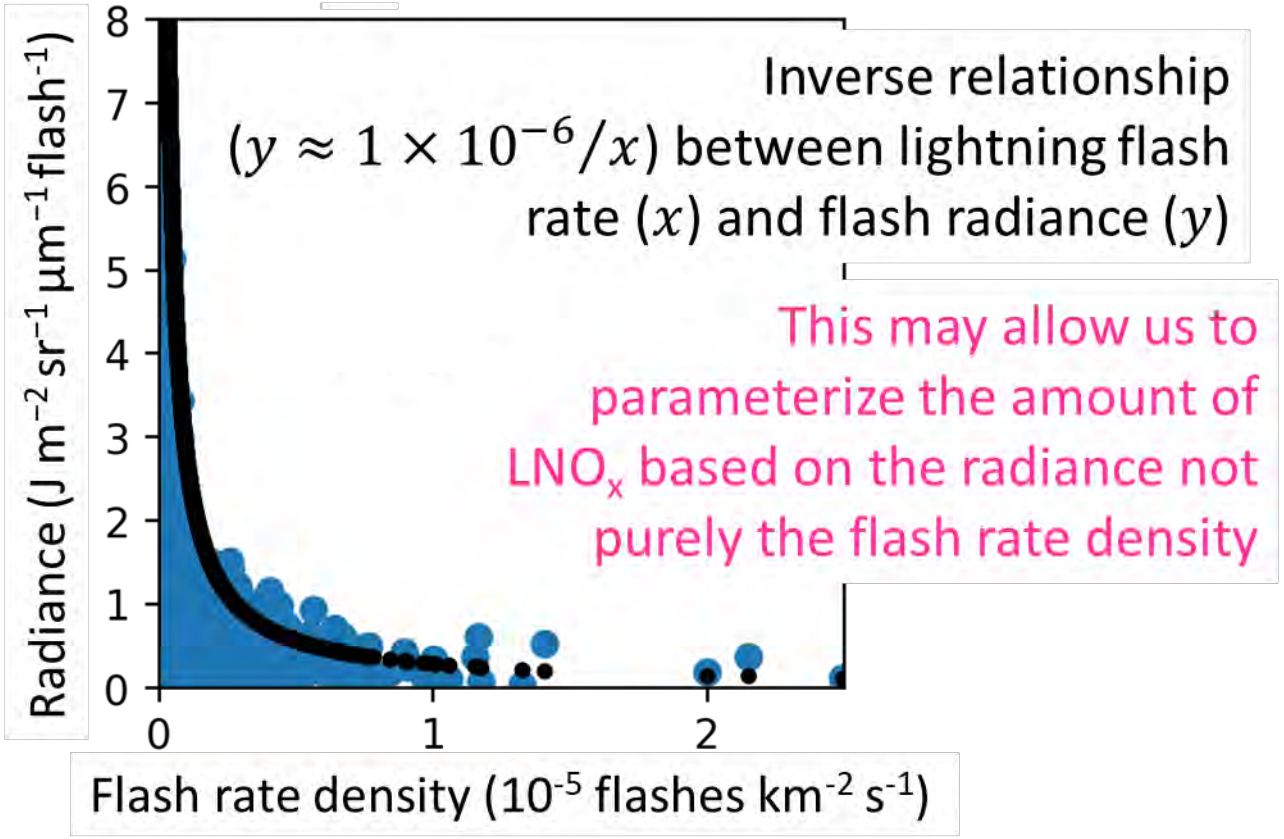
Routes to improving lightning NO_x scheme

Current lightning NOx parameterisation (Price & Rind, 1992)

- Lightning located parameterised by cloud top heights
- Then constrained by LIS/OTD observations
- 500 mol LNOx/flash in NH extratropics
- 260 mol LNOx/flash elsewhere



Current July Lightning NO emissions
(x10^{-11} kg m^{-2} s^{-1}) in GEOS-Chem



Summary

More UT NO_x information on Nana's poster: A.8

- We compare cloud-sliced TROPOMI UT NO_2 observations with GEOS-Chem
- GEOS-Chem ~ 60 % lower than TROPOMI

- Updates to kinetics increase GEOS-Chem UT NO_2 by ~ 20 %
- Key improvements: NO + O_3, NO_2 oxidation, PPN sinks

- PPN sinks added to GEOS-Chem (+OH, photolysis) outcompete thermal decomposition in the UT

- We are working on re-parameterising lightning NO_x based on radiance – any suggestions welcome!