Improving our understanding of upper tropospheric NO_x

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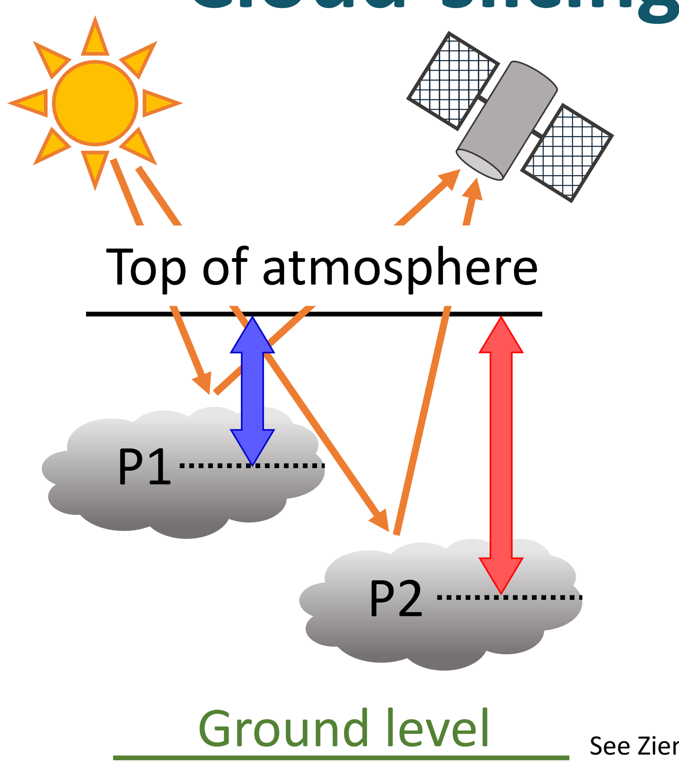
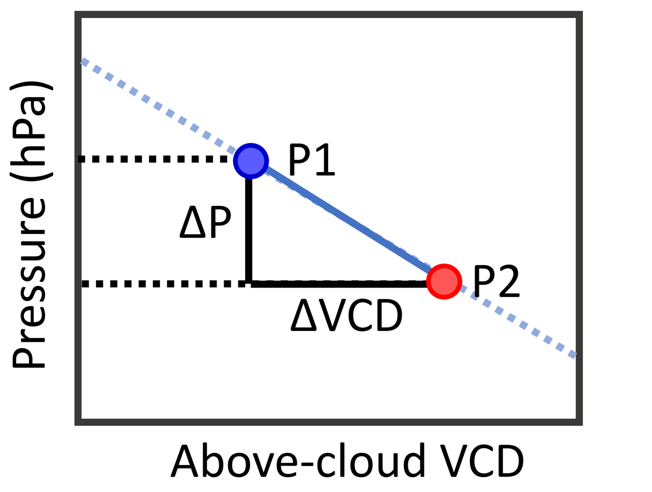
Cloud-slicing of satellite data



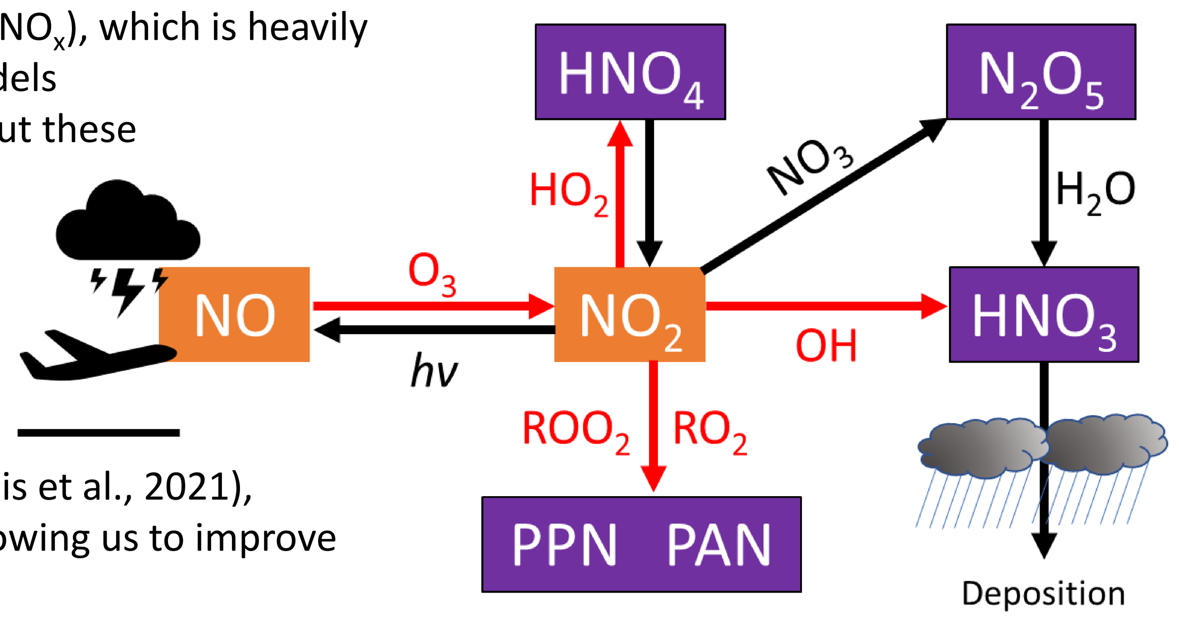
$$NO_2\ vmr = \frac{\Delta VCD}{\Delta P} \times \frac{k_b g}{R_{air}}$$



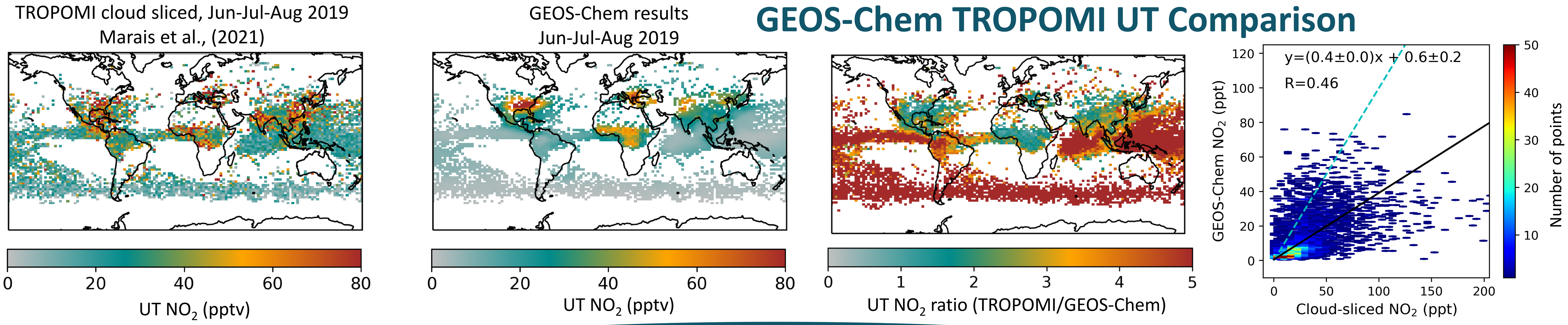
See Ziemke et al., (2012), figure adapted from Choi et al. (2014)

Importance of the upper troposphere (UT)

- Upper tropospheric (~450-180 hPa) nitrogen oxides ($NO + NO_2 \equiv NO_x$) are long-lived, enhancing the production efficiency of the potent greenhouse gas ozone.
- Dominant direct UT NO_x source is lightning (LNO_x), which is heavily parameterized in atmospheric chemistry models
- UT NO_x can be measured in-situ by aircraft, but these measurements are subject to interferences
- Updating chemistry by including missing processes (sinks of peroxypropionyl nitrate, PPN) and latest laboratory results has little effect on UT NO_x.
- Cloud-slicing total columns of NO_2 from TROPOMI, above optically thick clouds (Marais et al., 2021), provides near-global coverage of UT NO_2, allowing us to improve constraints on global lightning NO_x emissions



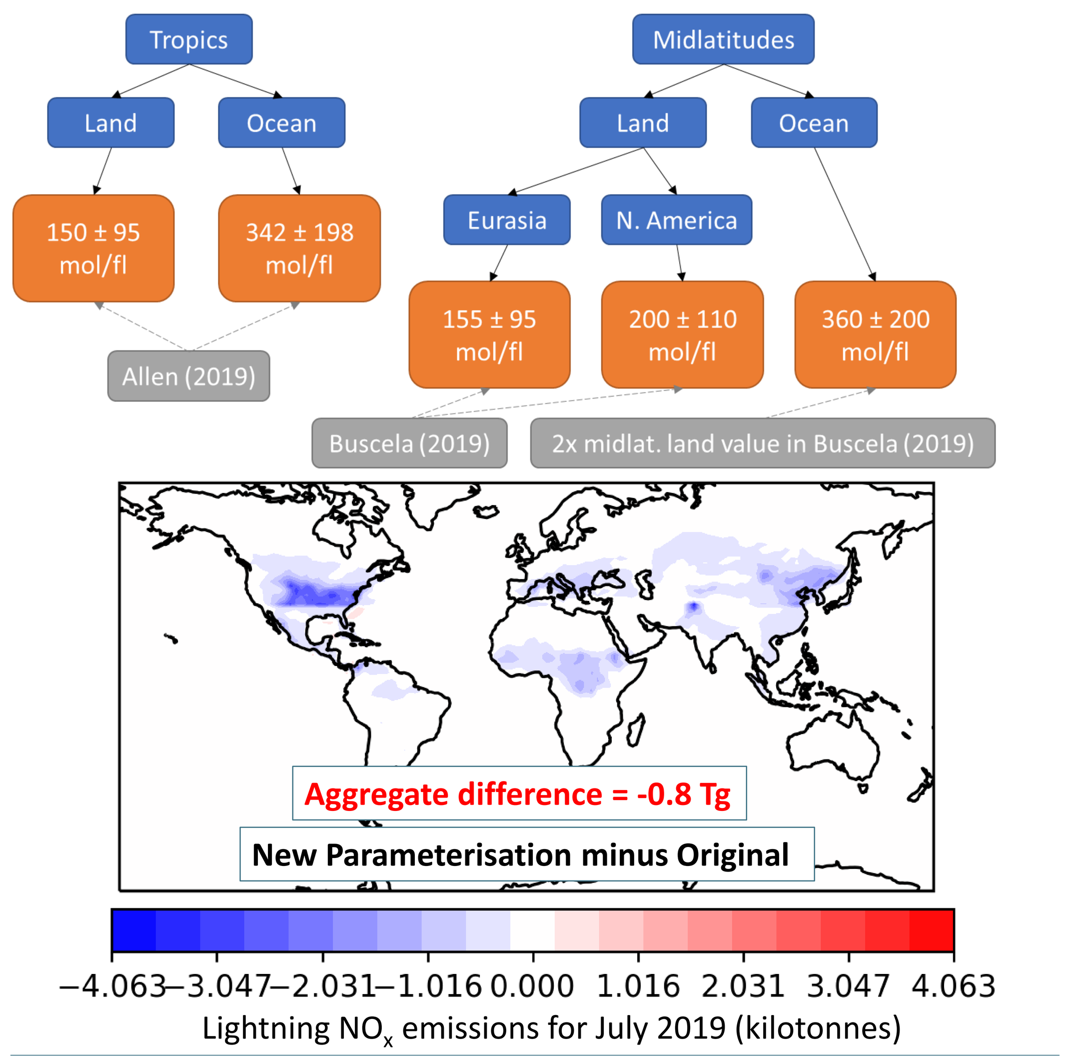
GEOS-Chem TROPOMI UT Comparison



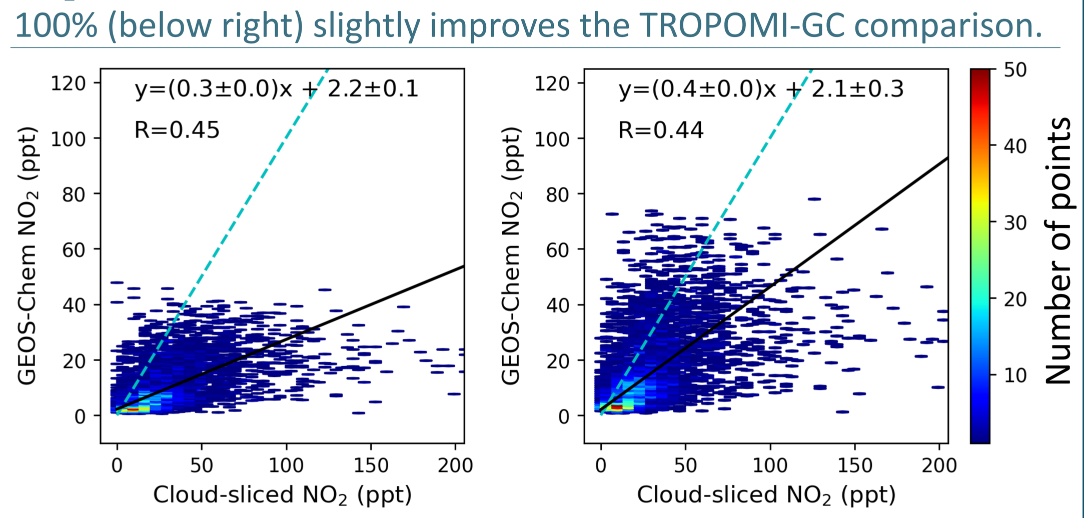
EVEN WITH LATEST CHEMISTRY, MODELS UNDERESTIMATE UPPER TROPOSPHERIC NO_2

Three times more lightning NO_x is required!

Updated LNO_x parameterisation based on latest literature

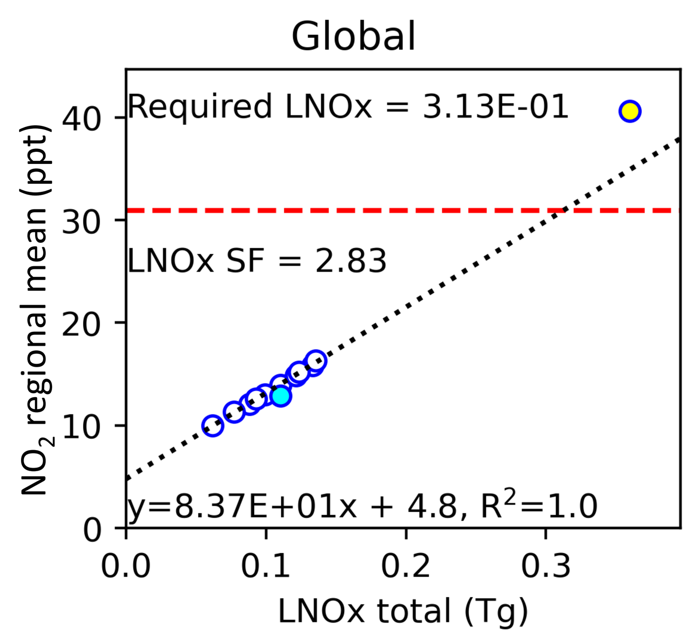


Parameterising LNO_x as suggested in Allen et al., (2019) and Buscela et al., (2019) reduces the amount of LNO_x and the amount of UT NO_2 (above and below left). Increasing this parameterisation by 100% (below right) slightly improves the TROPOMI-GC comparison.



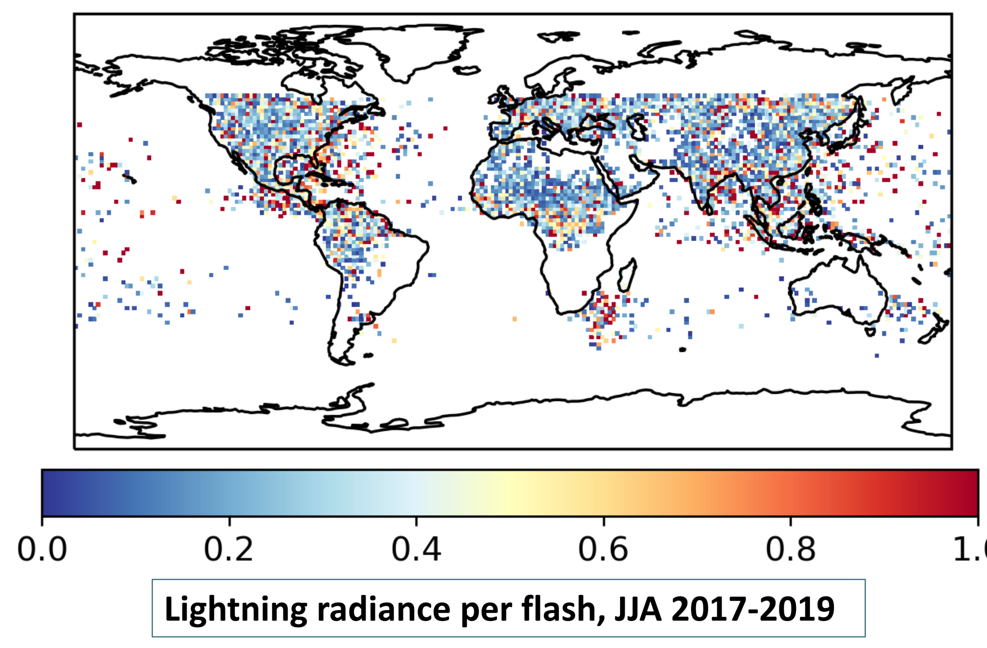
Adjusting the 'per-flash' LNO_x parameterisation

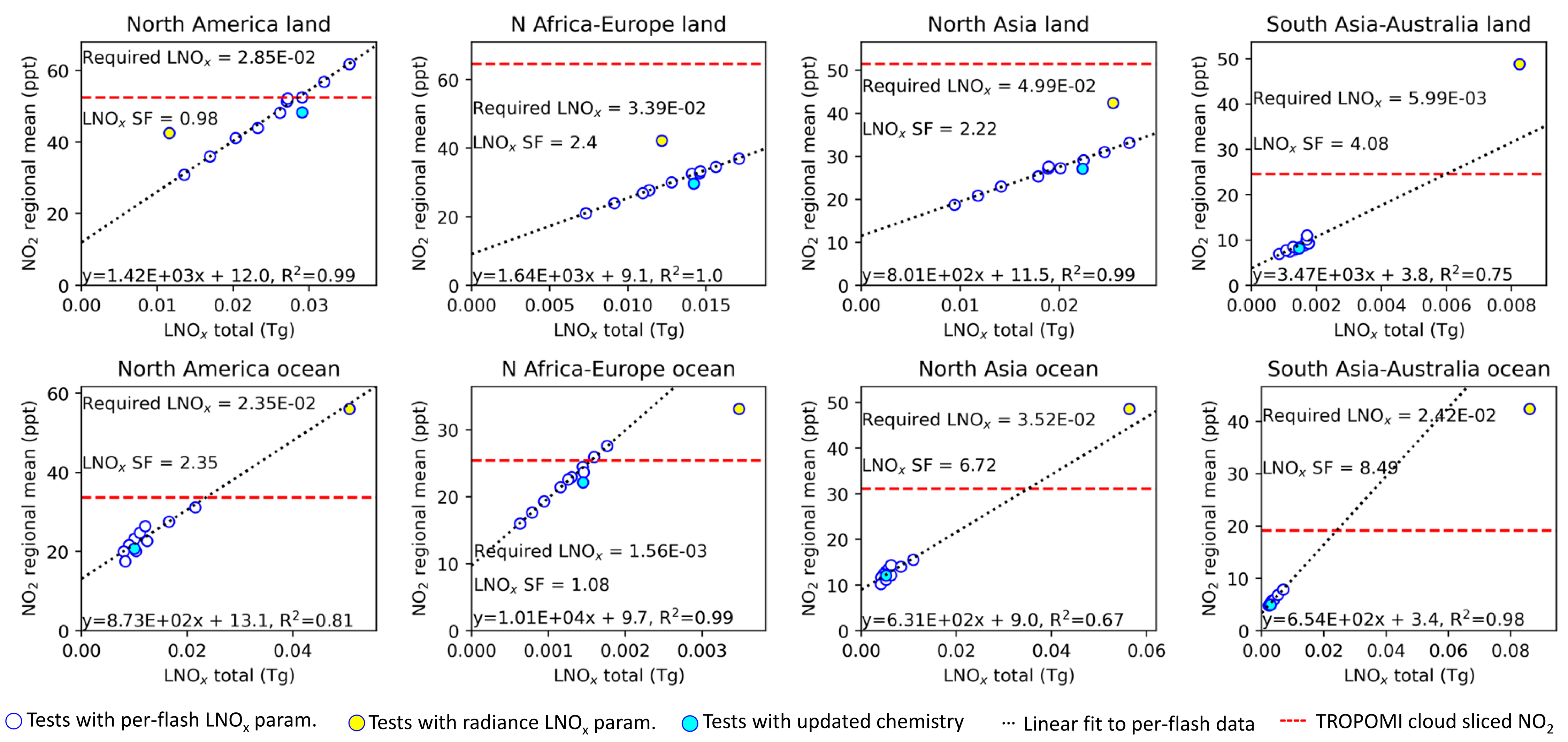
The relationship between total monthly modeled LNO_x and the resulting UT NO_2 mixing ratio, for a range of tests where the 'per-flash' parameterisation has been altered, is linear. This allows a 'Required LNOx' to be calculated which is the LNOx amount corresponding to the cloud-sliced UT NO2 value. Regional that in scaling factors (LNO_x SF) can be calculated, which is the amount the original GEOS-Chem LNO_x needs to be multiplied by to produce the Required LNOx. Using a global linear relationship and also by summing the regional Required LNOx values shows July global LNOx should be ~3 x higher than currently in GEOS-Chem.



Could we parameterise LNO_x by energy instead?

Other measured lightning metrics such as flash radiance and footprint have different spatial characteristics to flash density. We develop a new approach that uses radiances measured by the Lightning Image Sensor onboard the International Space Station to estimate NO_x emissions from lightning.





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