



# Improved lightning NO<sub>x</sub> emission inventory evaluated with vertical profiles of NO<sub>2</sub> and ozone obtained by cloud-slicing TROPOMI

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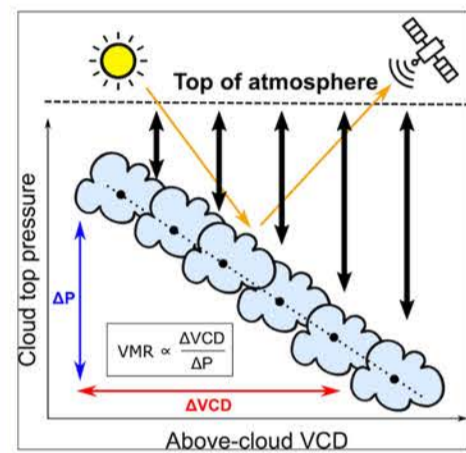
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We derive global vertical profiles of tropospheric NO<sub>2</sub> and O<sub>3</sub> from TROPOMI that we use to identify the need to improve representation of lightning NO<sub>x</sub>

## 1 Cloud-slicing to yield vertical profiles of pollutants

### Schematic of cloud-slicing

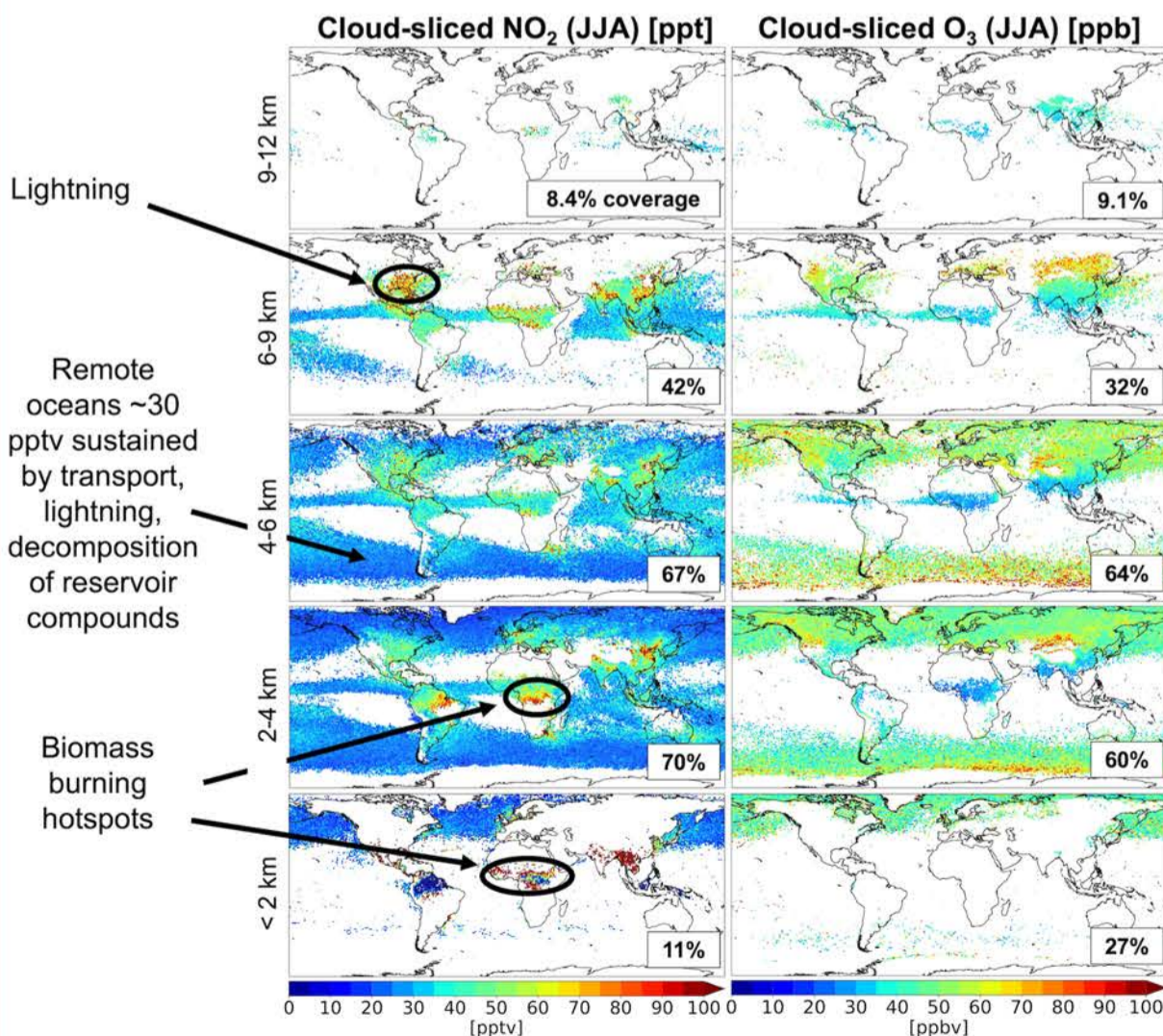


- Nitrogen oxides (NO<sub>x</sub>) are key contributors to tropospheric ozone (O<sub>3</sub>).
- Observations of the vertical profiles of tropospheric NO<sub>x</sub> and O<sub>3</sub> are severely limited.
- Cloud-slicing (left) addresses this by taking advantage of satellite partial columns separated by optically thick clouds<sup>[1]</sup>.
- We apply cloud-slicing to TROPOMI NO<sub>2</sub> and O<sub>3</sub> total columns in 2018 to 2022.

Cloud-slicing yields global vertical profiles of seasonal mean NO<sub>2</sub> and O<sub>3</sub> volumetric mixing ratios (VMRs) in 5 tropospheric layers

## 2 Vertical profiles of NO<sub>2</sub> and O<sub>3</sub> from TROPOMI

Global cloud-sliced multiyear mean NO<sub>2</sub> and O<sub>3</sub> obtained at high resolution (1° x 1°) in 5 discrete layers: 2 upper troposphere (6-9 km & 9-12 km), 2 mid-troposphere (2-4 km & 4-6 km), and 1 boundary layer (BL) (< 2 km).



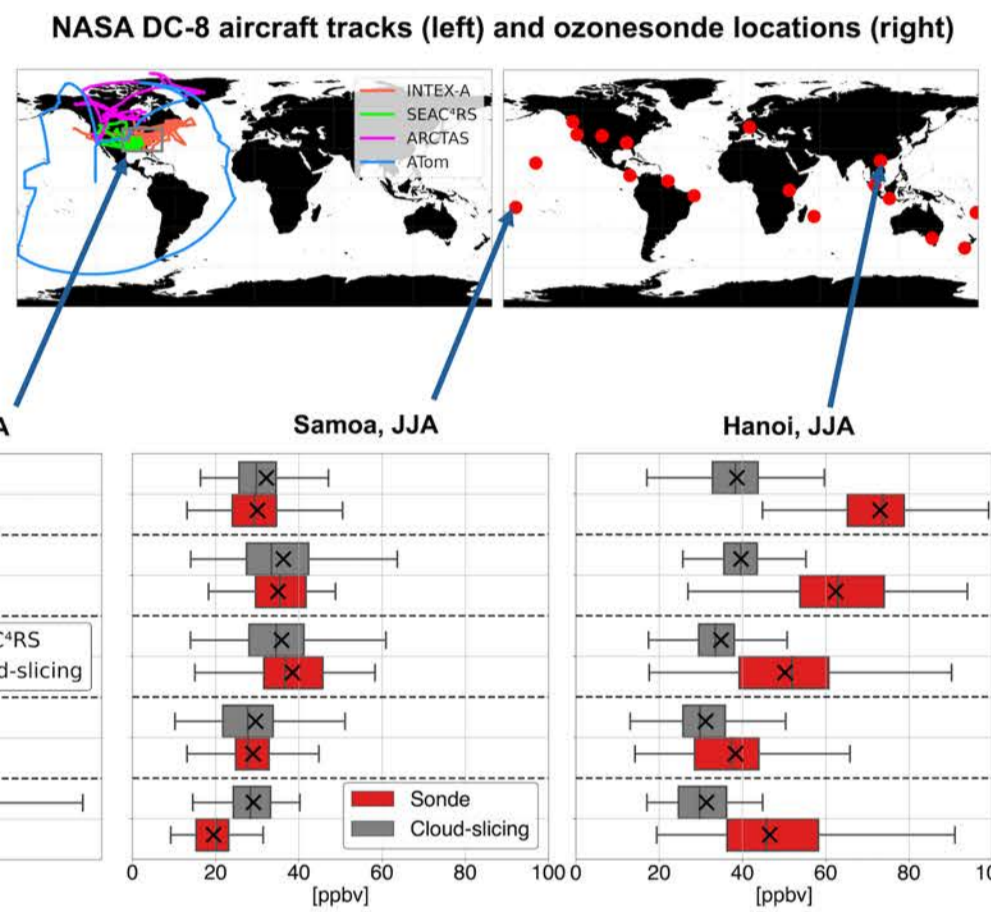
Lightning  
 Remote oceans ~30 pptv sustained by transport, lightning, decomposition of reservoir compounds

Biomass burning hotspots

Greatest **coverage** in the mid troposphere where clouds are abundant. Data **range** from ~ 30 ppt (remote oceans) to > 100 ppt (BL and mid troposphere over biomass burning locations) for NO<sub>2</sub> and ~35 ppb (tropics) to >200 ppb (northern midlatitudes influenced by pollution) for O<sub>3</sub>.

## 3 Cloud-slicing assessed with independent measurements

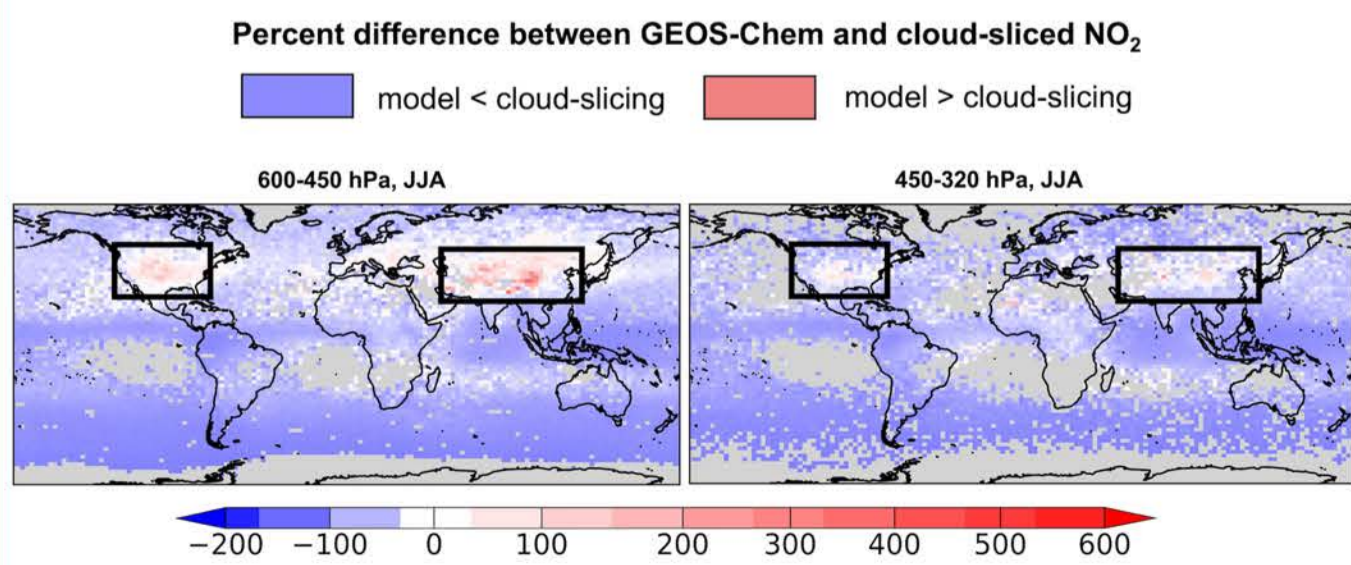
We compare cloud-sliced data to NASA DC-8 aircraft data for NO<sub>2</sub> and SHADOZ, WOUDC and NOAA ozonesondes for O<sub>3</sub> coincident with TROPOMI overpass.



NO<sub>2</sub> data differences are small (10-15 ppt) when data density and sampling are consistent, but this is rare. O<sub>3</sub> data is consistent at most sites (see Samoa above), but a few sites exhibit large differences in the upper troposphere (see Hanoi above).

## 4 Assessing understanding of tropospheric NO<sub>x</sub> and O<sub>3</sub>

We use GEOS-Chem as state-of-knowledge of tropospheric NO<sub>x</sub> and O<sub>3</sub> and compare it to the cloud-sliced data on the **global model grid** (2° x 2.5°). The model is updated to include liberation of NO<sub>x</sub> via peroxypropionyl nitrate (PPN) photolysis and particle nitrate photolysis.<sup>[2]</sup>

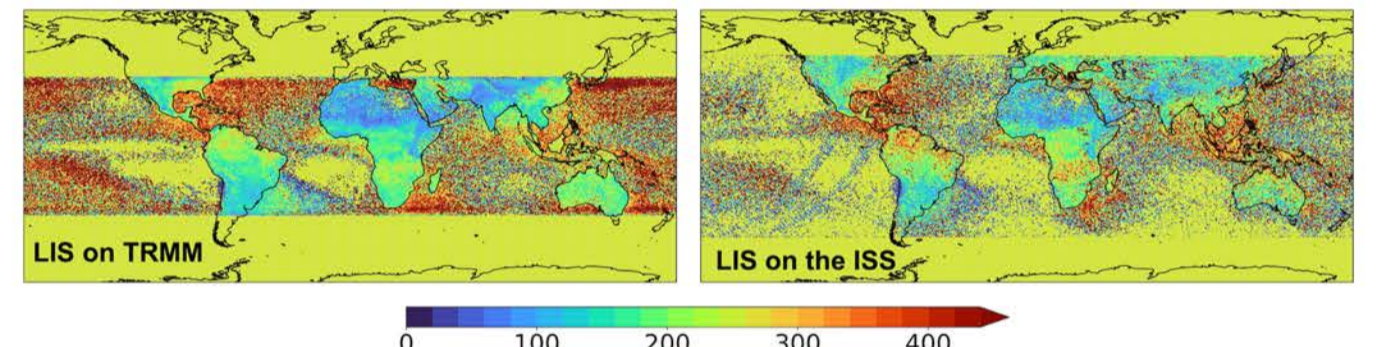


GEOS-Chem overestimates NO<sub>2</sub> in the mid and upper troposphere over northern mid-latitudes, as lightning NO<sub>x</sub> production is double (500 mol/fl) everywhere else (260 mol/fl).

## 5 Lightning NO<sub>x</sub> production rates from flash energy proxies

Lightning imaging sensor (LIS) flash energies (radiances) are used with observationally-constrained global mean lightning NO<sub>x</sub> production (280 mol/fl) to calculate spatially resolved values for input to models<sup>[3]</sup>.

Gridded (0.5° x 0.625°) lightning NO<sub>x</sub> production rates in moles per flash

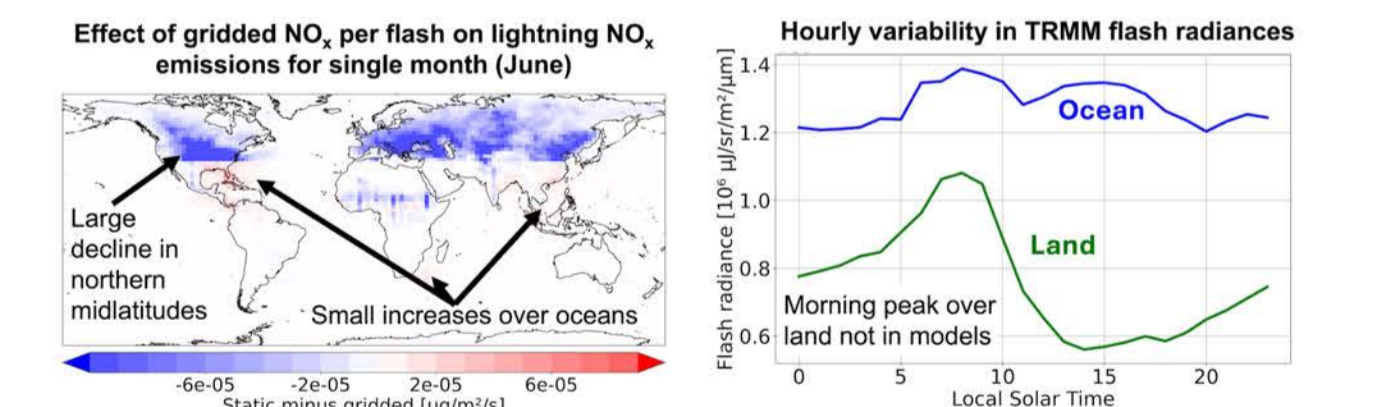


NO<sub>x</sub> per flash is far greater over oceans (250-500 mol/fl) than land (35-200 mol/fl), but differences between ISS and TRMM over oceans needs to be resolved to exploit greater latitudinal extent of ISS.

NO<sub>x</sub> per flash for use in CTMs is far more variable than fixed values currently invoked. Greater NO<sub>x</sub> per flash over oceans may resolve remote ocean low bias shown in Box 4.

## 6 Implications of gridded NO<sub>x</sub> per flash on lightning NO<sub>x</sub> emissions

We incorporate gridded NO<sub>x</sub> per flash in the emissions component (HEMCO) of GEOS-Chem (left plot), yielding annual emissions (6.5 Tg N) greater than the original model version (5.8 Tg N).



Mass of nitrogen emitted by lightning is relatively unaffected, but spatial distribution (shift from land to ocean) is. Hourly variability effects on atmospheric composition needs to be tested.

## 7 Ongoing work

- Resolve TRMM and LIS differences
- Account for hourly variability in lightning NO<sub>x</sub> production rates, as LIS data indicate greatest NO<sub>x</sub> production in the morning.
- Assess the impact of more mechanistic lightning NO<sub>x</sub> parameterisation on tropospheric NO<sub>x</sub>, O<sub>3</sub>, oxidants, and compounds affected by oxidant abundance.

## Acknowledgements

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## Key references

- [1] Ziemke J.R. et al. (2001) JGR: Atmospheres 106(D9)
- [2] Shah V. et al. (2023) ACP 23(2)
- [3] Chronis T. & Koshak W.J. (2017) BAMS 98(7)