Cloud-slicing to obtain vertical profiles of tropospheric NO_2 and ozone: Challenges and opportunities with geostationary instruments

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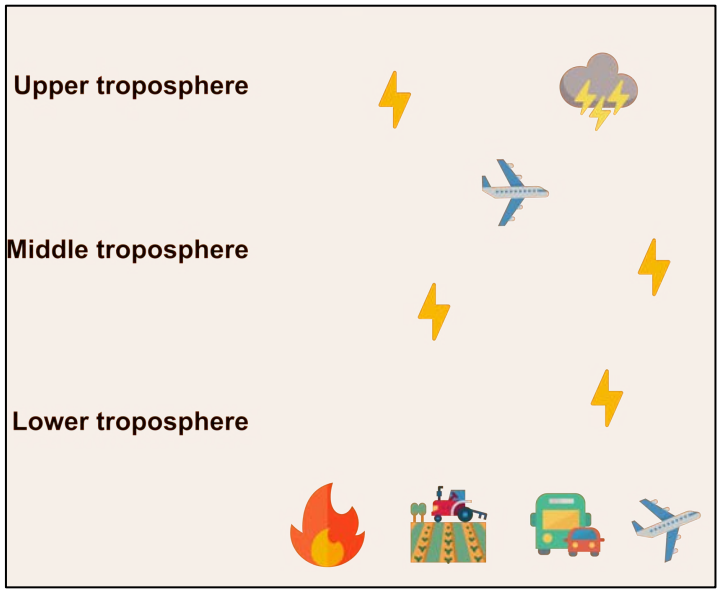
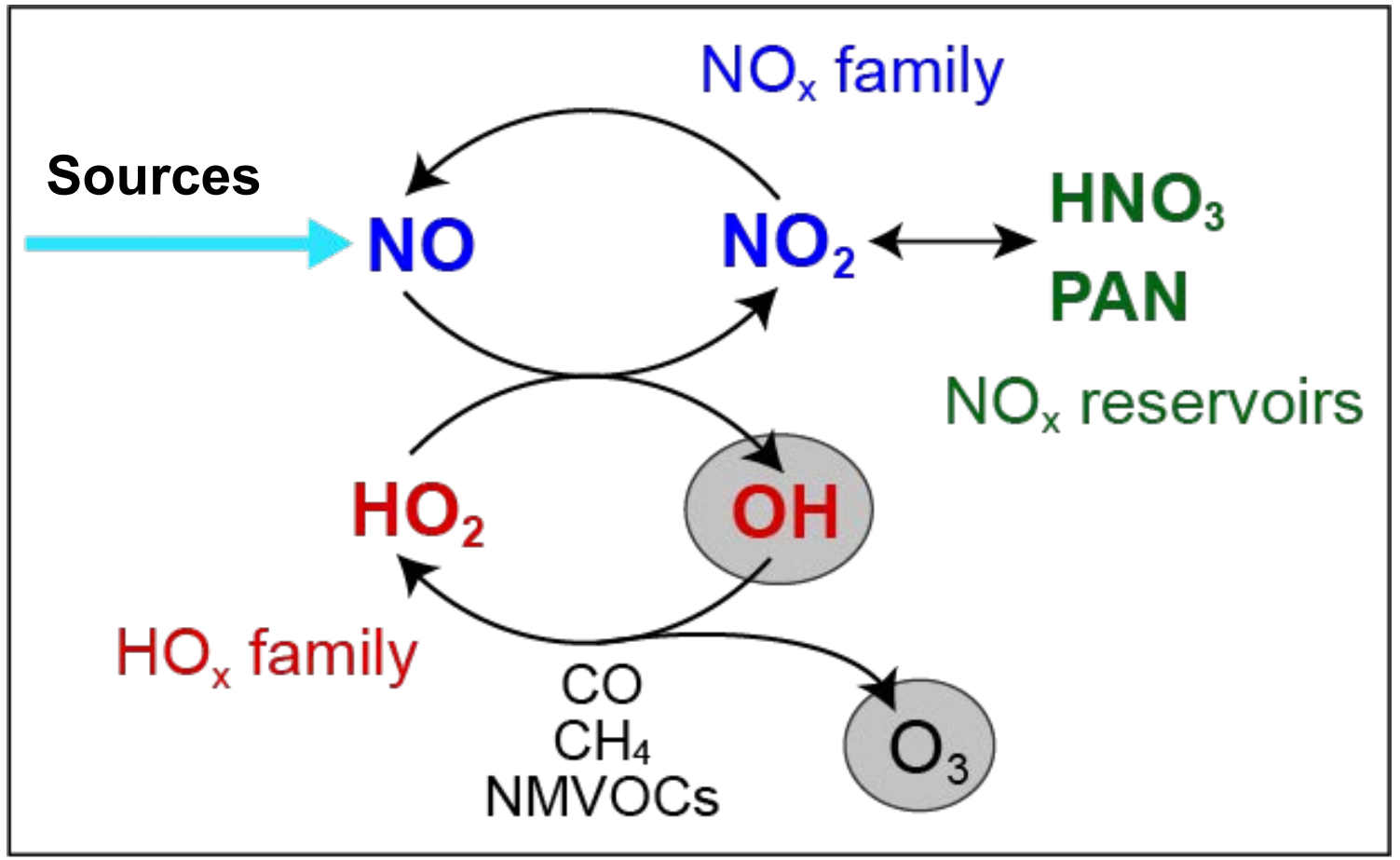
Left to Right: Nana Wei (PhD), Eloise Marais (lead), Connor Barker (postdoc), Karn Vohra (postdoc), Eleanor Gershenson-Smith (PhD), **Bex Horner (PhD)**

Tropospheric nitrogen oxides ($NO_x \equiv NO + NO_2$)





Influences climate, air quality, food security, oxidative capacity of the troposphere

Limitations of current observing systems

TROPOMI tropospheric NO_2 [μmol m^{-2}]

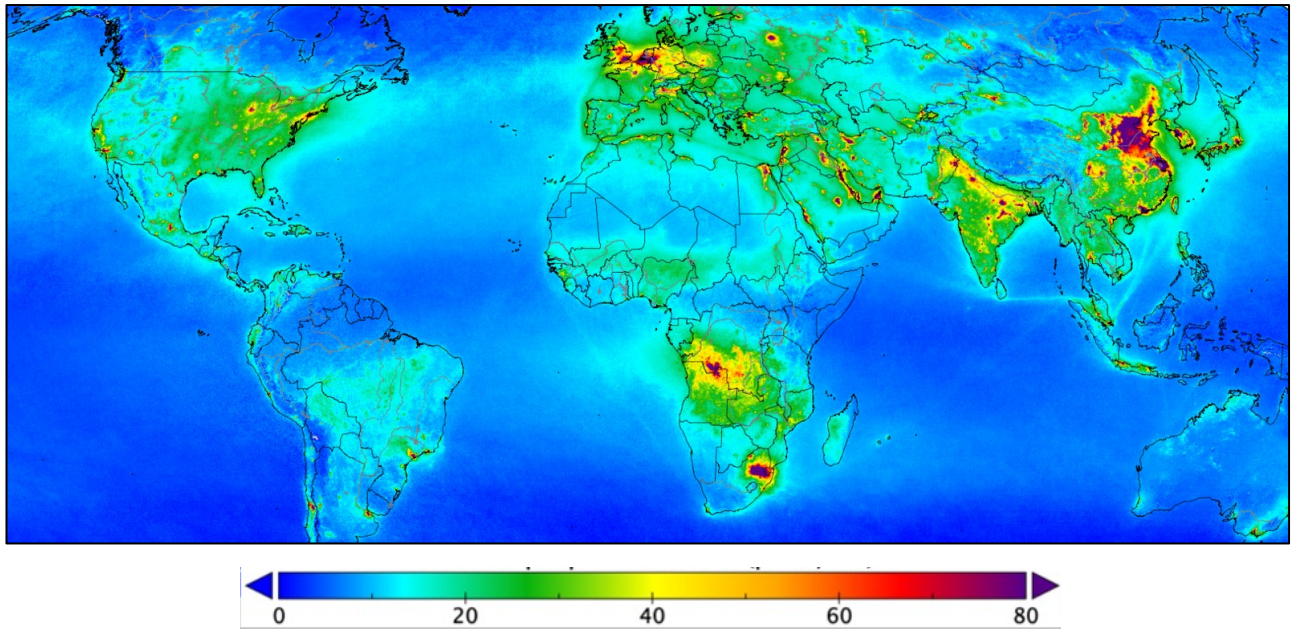
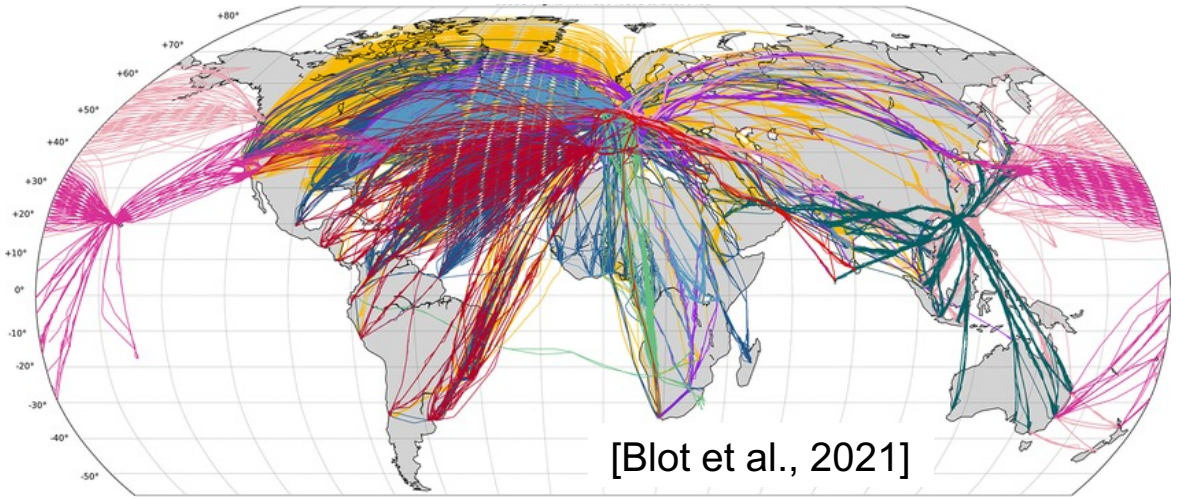
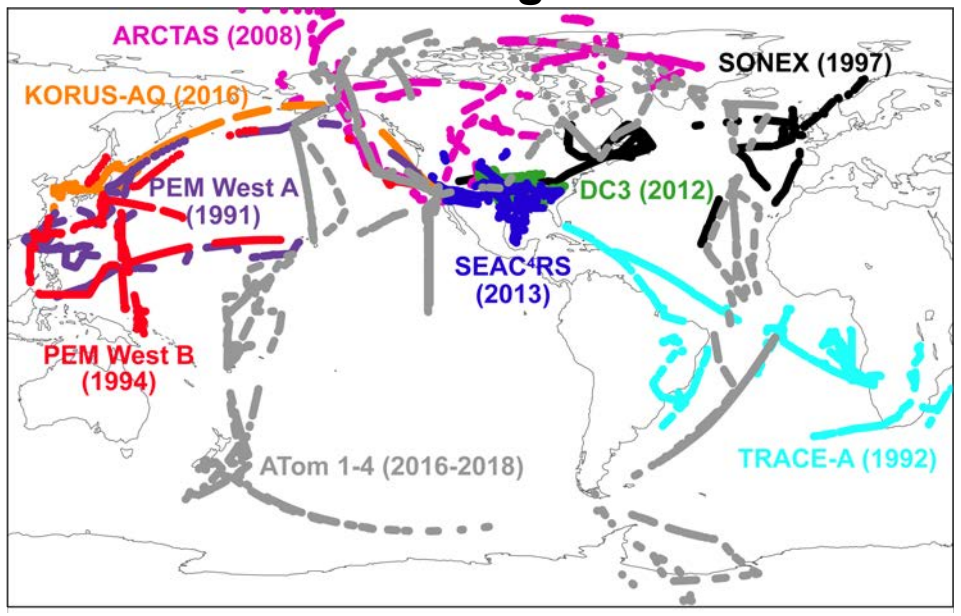


Image source: https://www.esa.int/Applications/Observing_the_Earth/Copernicus/Sentinel-5P/Nitrogen_dioxide_pollution_mapped

IAGOS campaign flight tracks



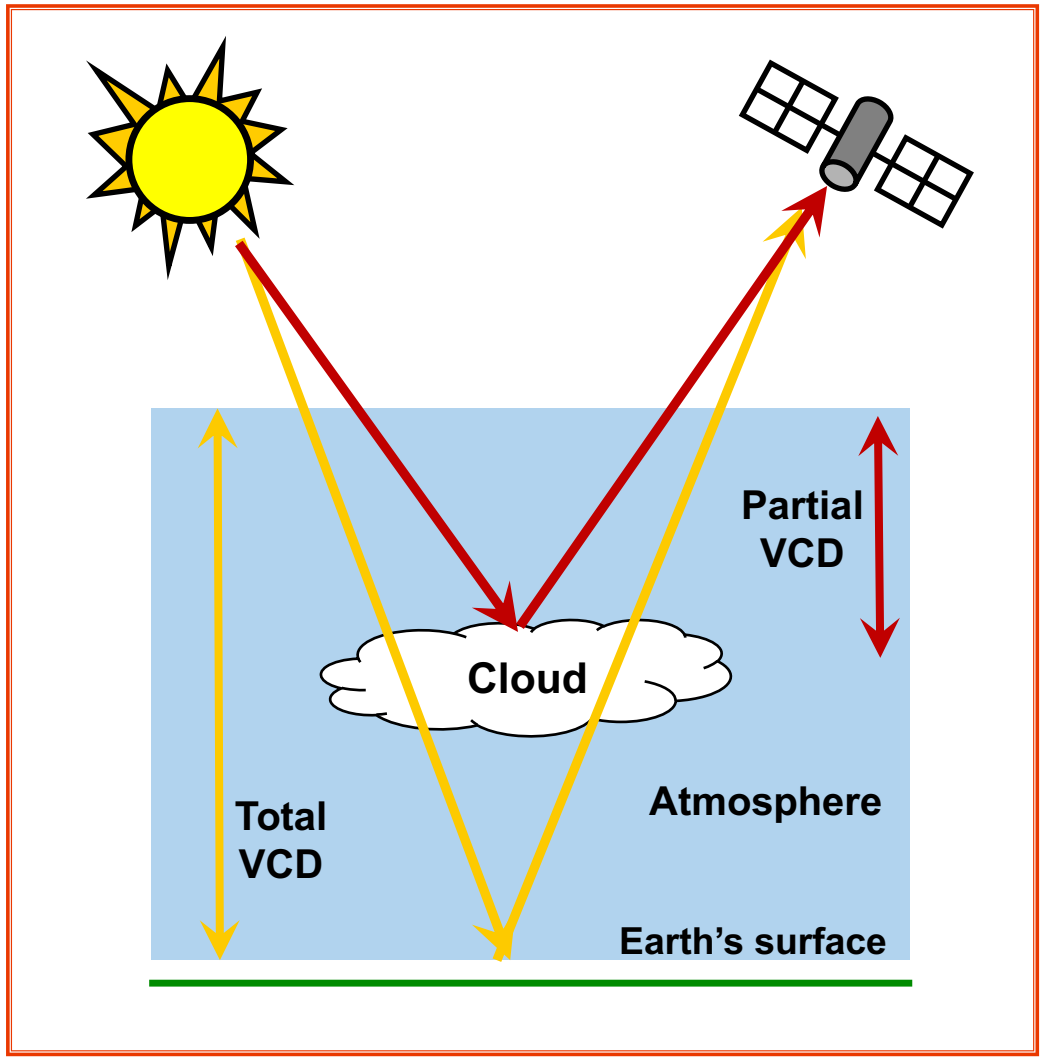
NASA DC8 flight tracks



Operational satellite data products offer one piece of vertical information

Aircraft observations intermittent and in situ instruments prone to interference from reservoir compounds

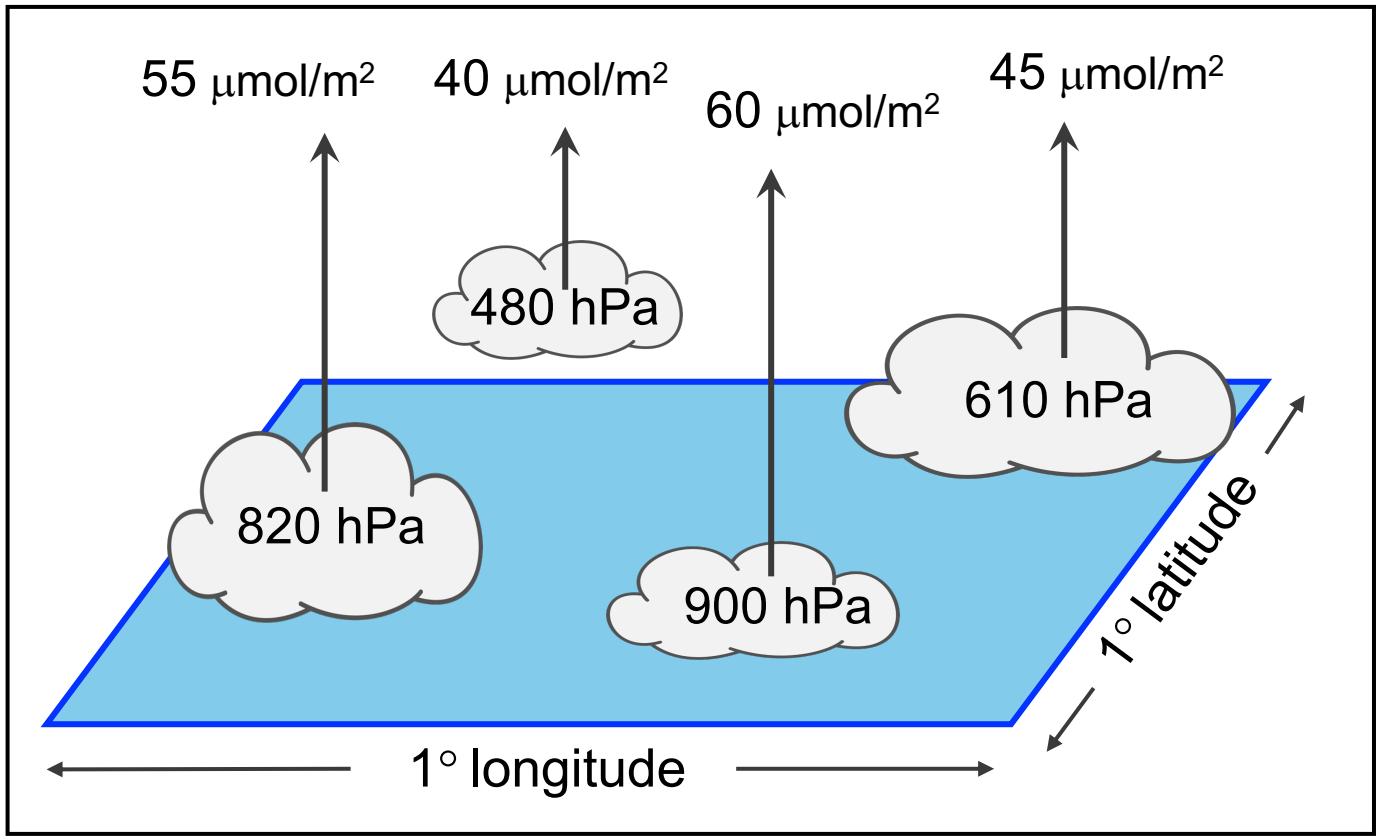
Optically thick clouds split up the troposphere



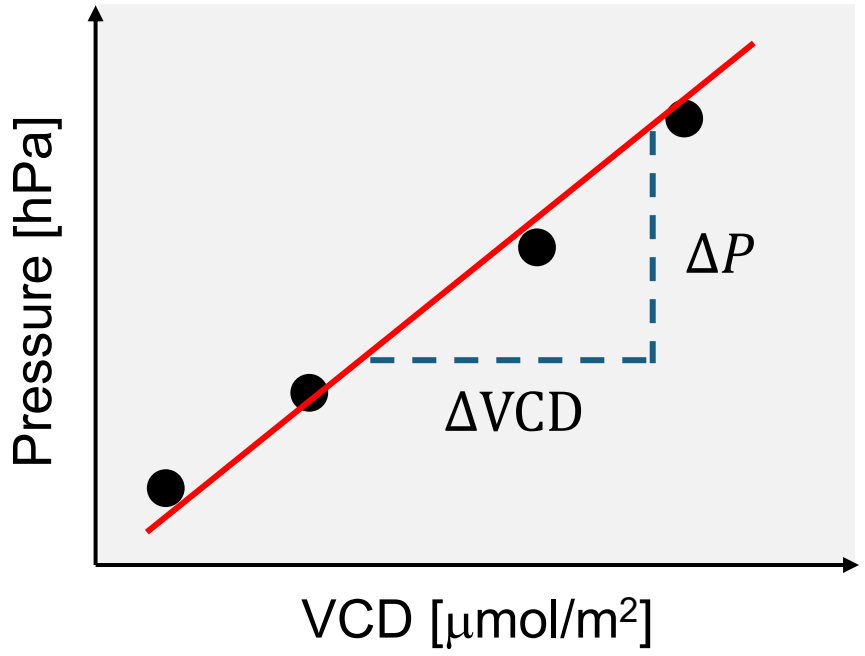
Retrieve partial columns above optically thick clouds (data typically discarded)

Convert partial columns to NO_2 mixing ratios:
Cloud-slicing

Clusters of partial columns above optically thick clouds:



Regress cloud top pressures against partial vertical column densities (VCDs):

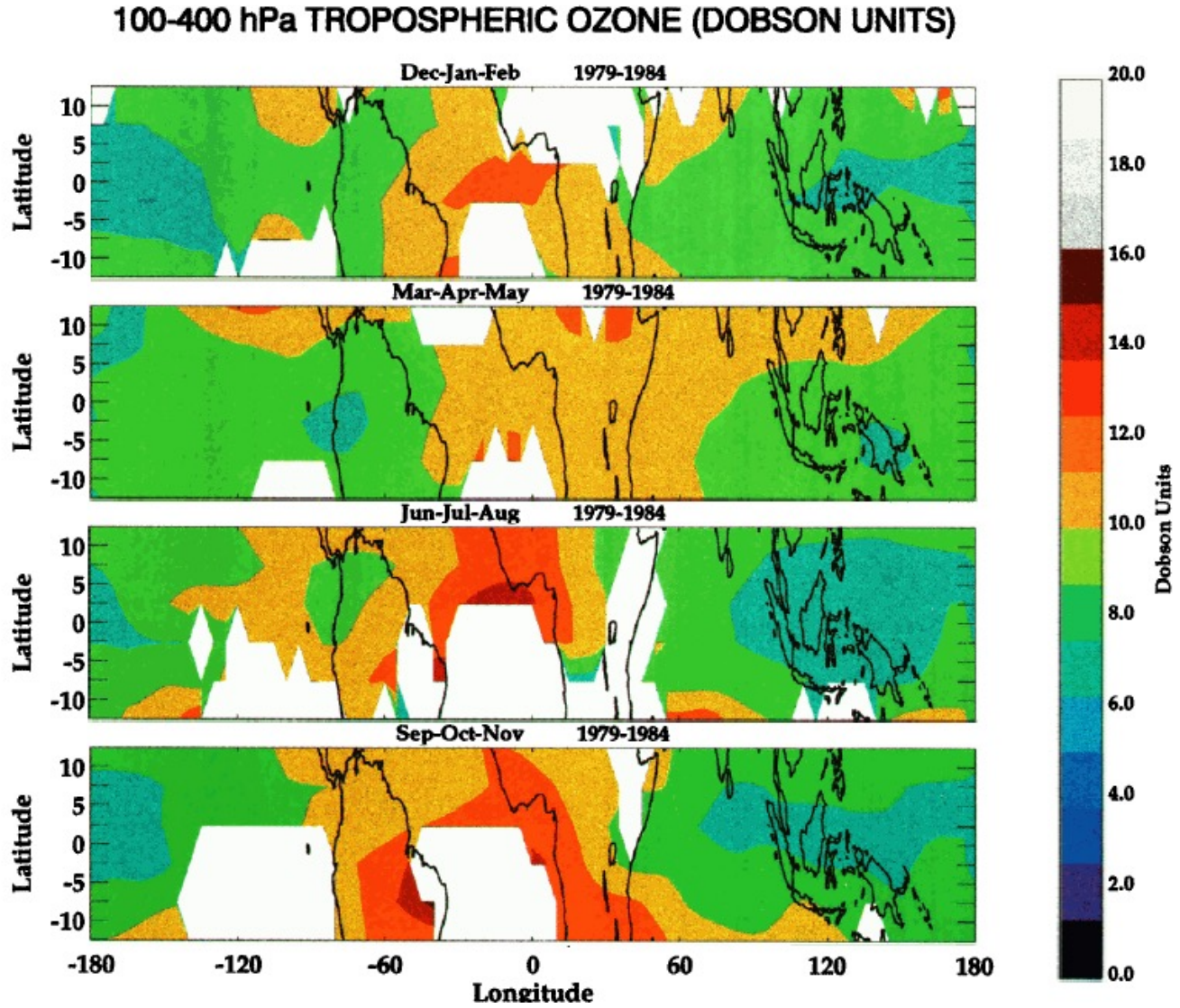


Calculate average mixing ratio between target pressure ranges:

$$\mathrm{NO_2\ VMR} = \frac{\Delta \mathrm{VCD}}{\Delta P} \times \mathrm{const}$$

Past Application of Cloud-slicing to TOMS ozone

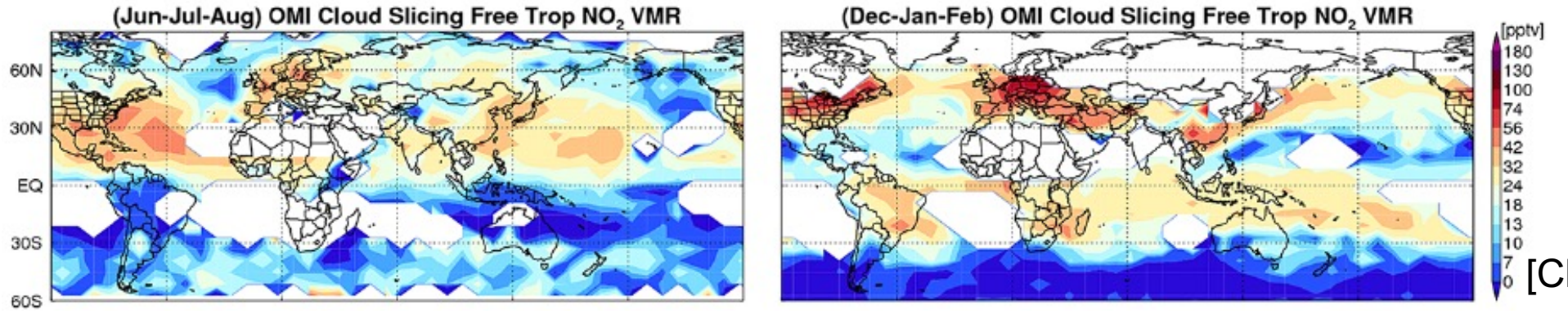
Cloud-slicing applied to Total Ozone Mapping Spectrometer (TOMS) to derive upper tropospheric ozone



[Ziemke et al., 2001]

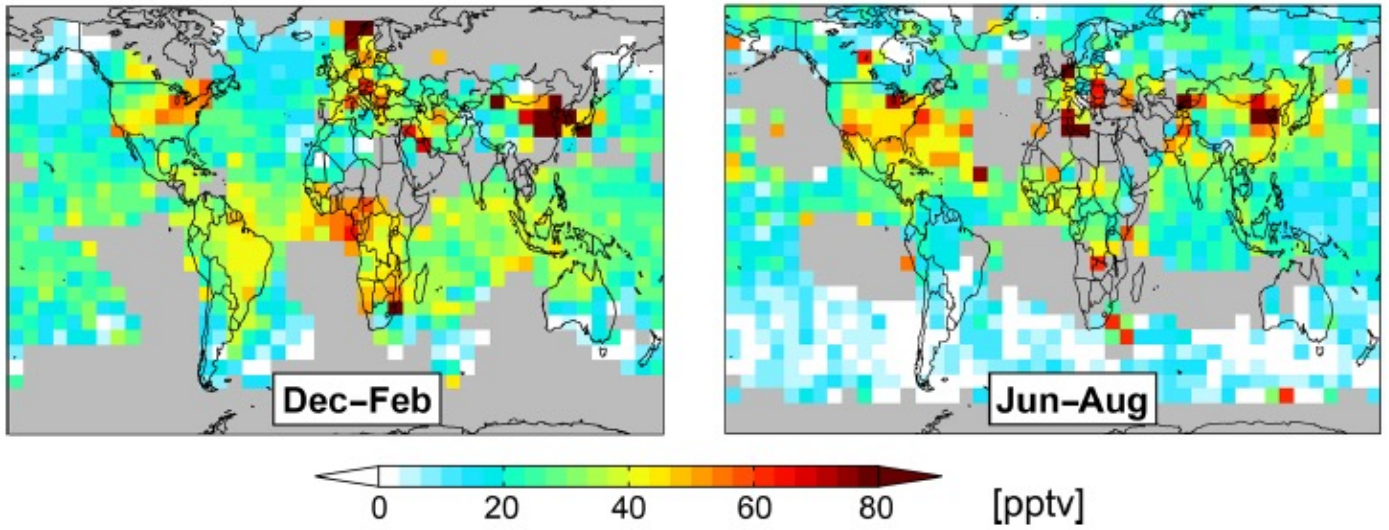
Past Application of Cloud-slicing to OMI NO_2

Cloud-slicing applied to OMI to yield NO_2 in the middle troposphere (Choi et al., 2014), multiple layers in the troposphere (Belmonte Rivas et al., 2015) and upper troposphere (Marais et al., 2018)



Resolution:
5° lat x 8° lon

[Choi et al., 2014]



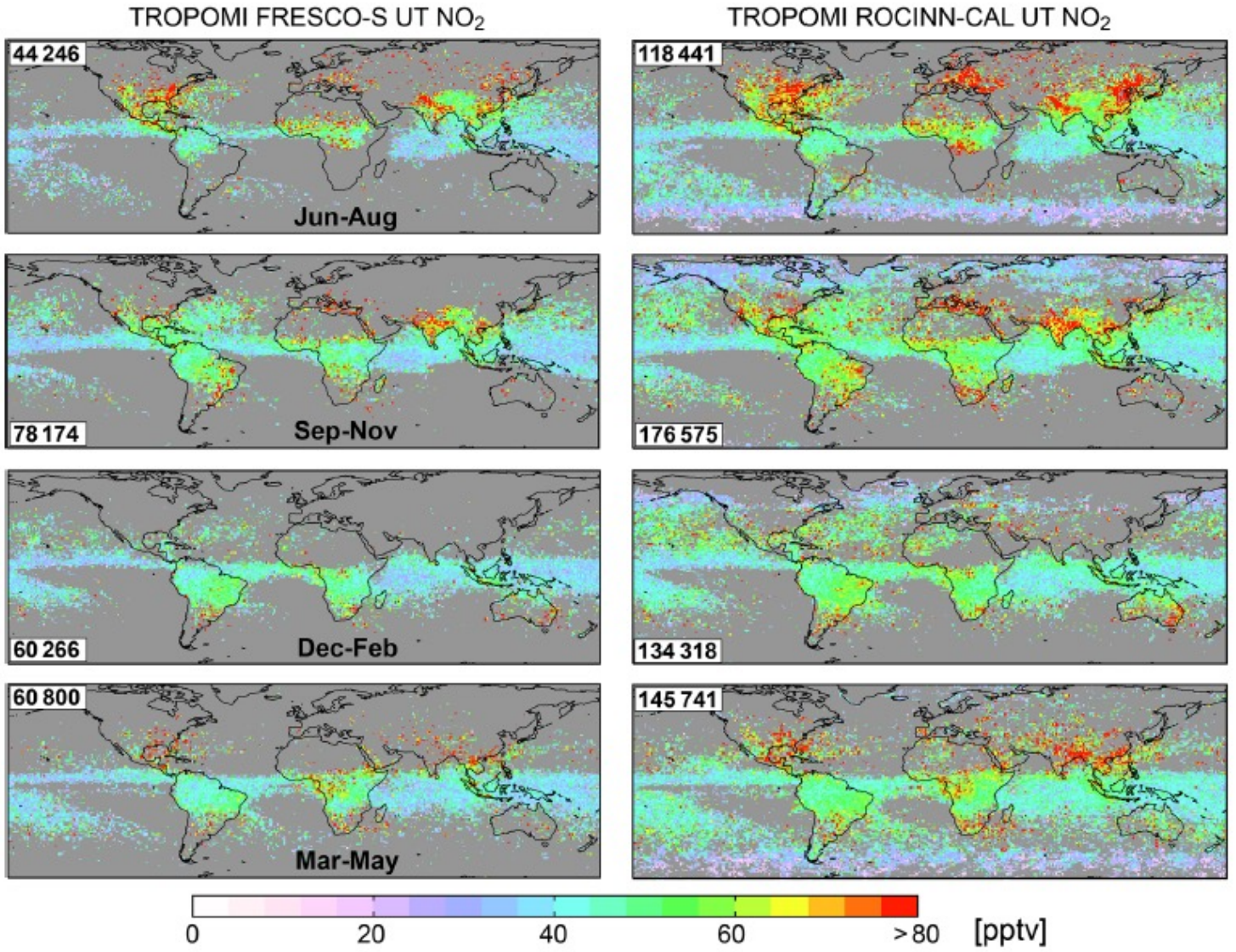
Resolution:
5° lat x 8° lon

[Marais et al., 2018]

OMI coarse resolution and not fit for use after row anomaly (low data density)

Recent Application of Cloud-slicing to TROPOMI NO_2

Limited to upper troposphere for a single year. Achieve much finer resolution (1º) than OMI

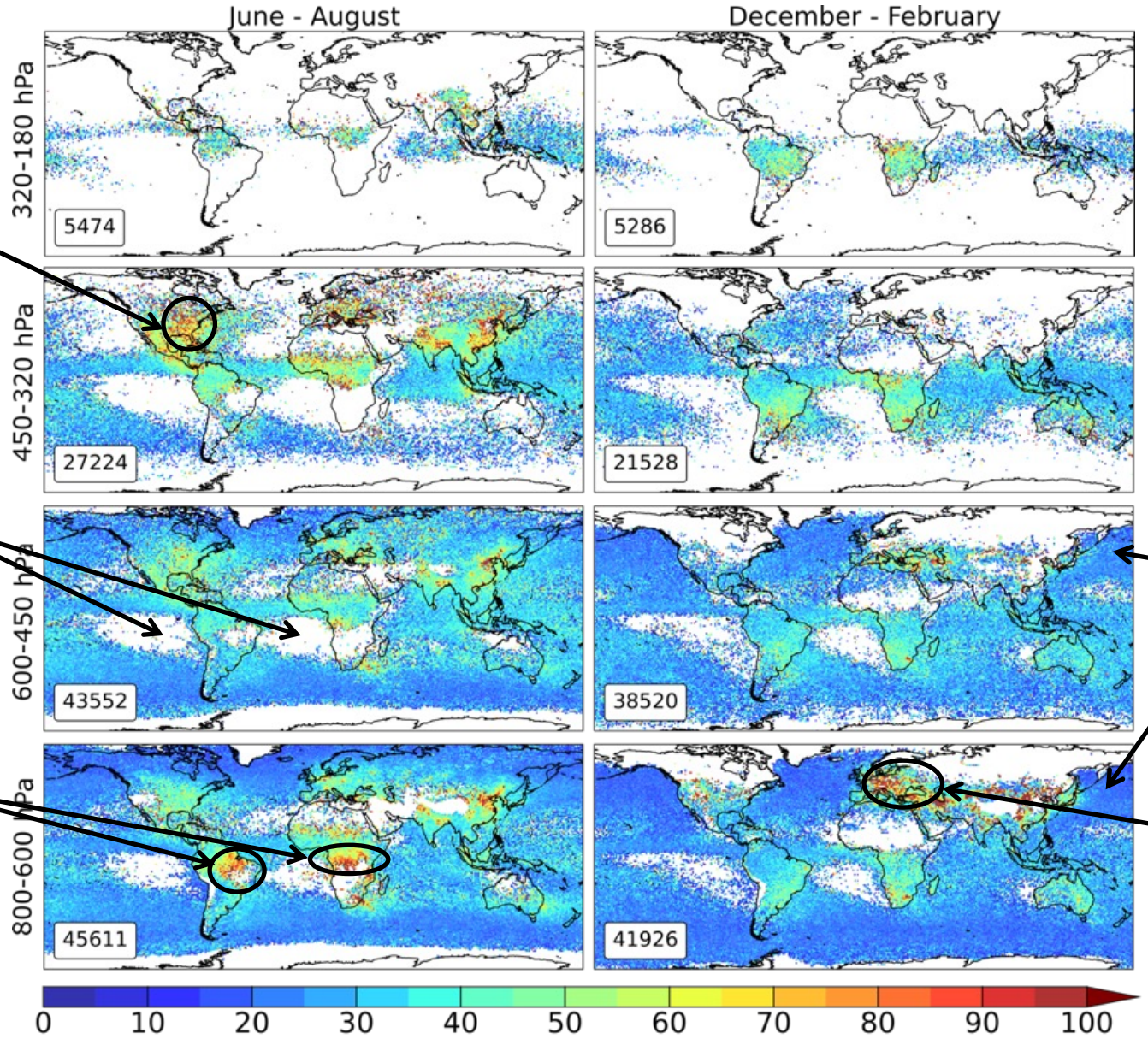


Seasonal means obtained with different cloud products

[Marais et al., 2021]

Current Application to TROPOMI: free troposphere

Cloud-sliced multiyear mean free tropospheric NO_2 [pptv]



Lightning

Little coverage → persistent subsidence

Open fires

Little coverage in top layer (few clouds)

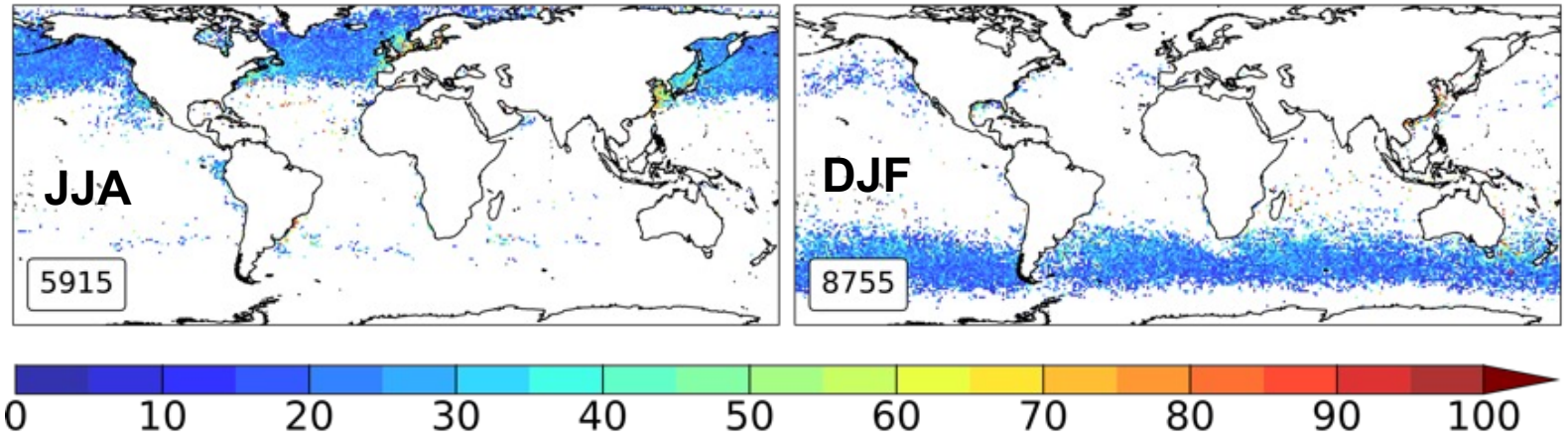
Remote background NO_2 of ~20-30 pptv

Long winter lifetimes

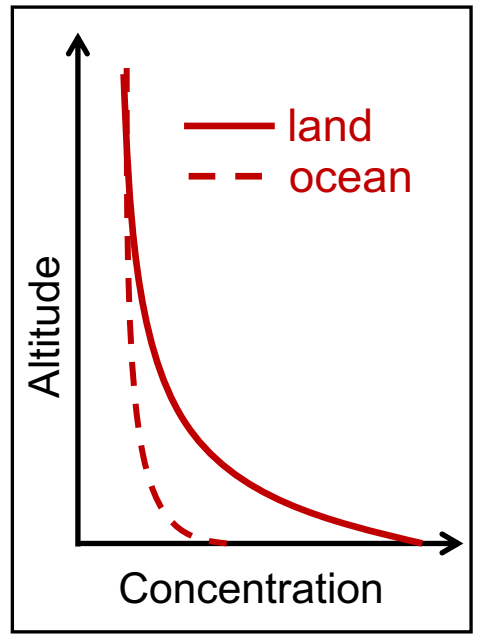
[Horner et al., *submitted* 2024]

Current Application to TROPOMI: boundary layer

Cloud-sliced multiyear mean boundary layer NO_2 [pptv]

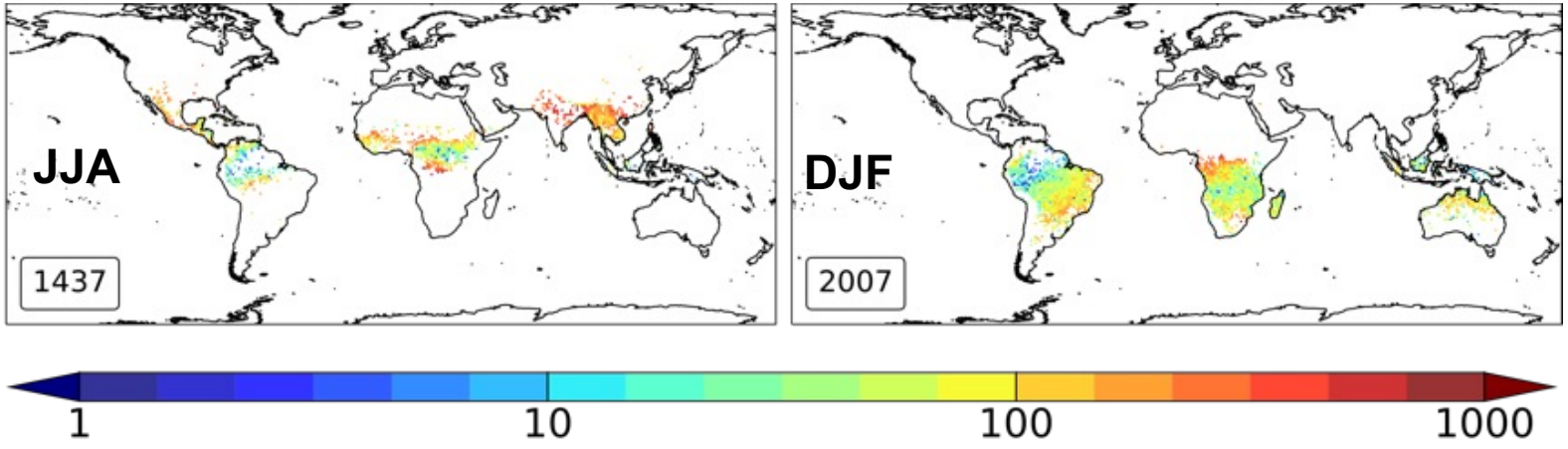


NO_2 vertical profile



Cloud-slicing assumes NO_2 uniform within each layer. Assumption doesn't hold over surface source regions, so take difference of cloud-sliced and tropospheric columns:

Differenced multiyear mean boundary layer NO_2 [pptv]



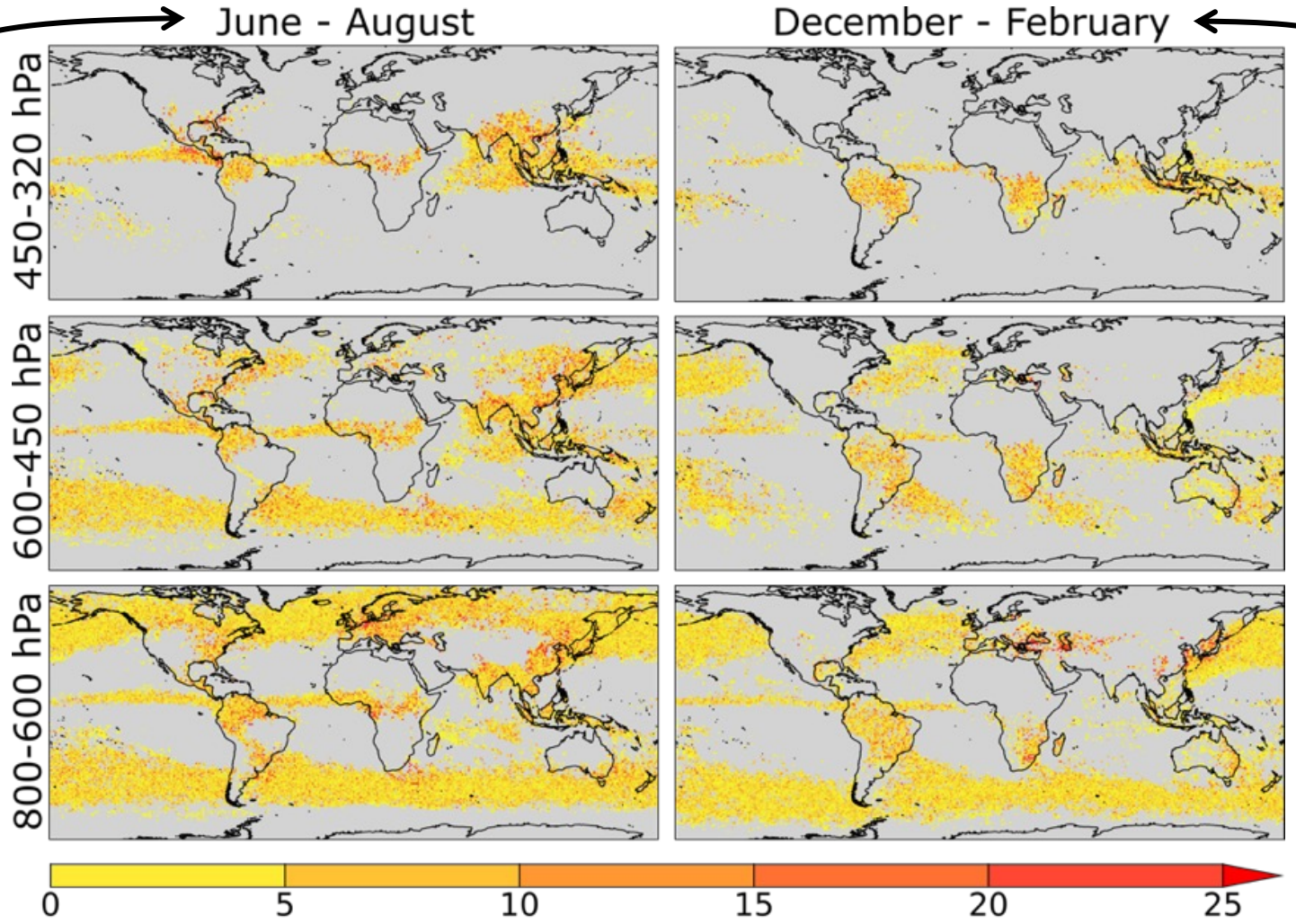
[Horner et al., *submitted* 2024]

NO_2 Interannual Variability from Cloud-sliced Data

Interannual variability of cloud-sliced NO_2 [pptv]

JJA 2021 minus 4-year mean

Dec 2020 to Feb 2021 minus 4-year mean



5-10 pptv (25-30%) over remote regions

<25 pptv (~40%) over source regions

First ever observationally-derived interannual variability in the vertical distribution of tropospheric NO_x suggests NO_x relatively stable from year to year

[Horner et al., *submitted* 2024]

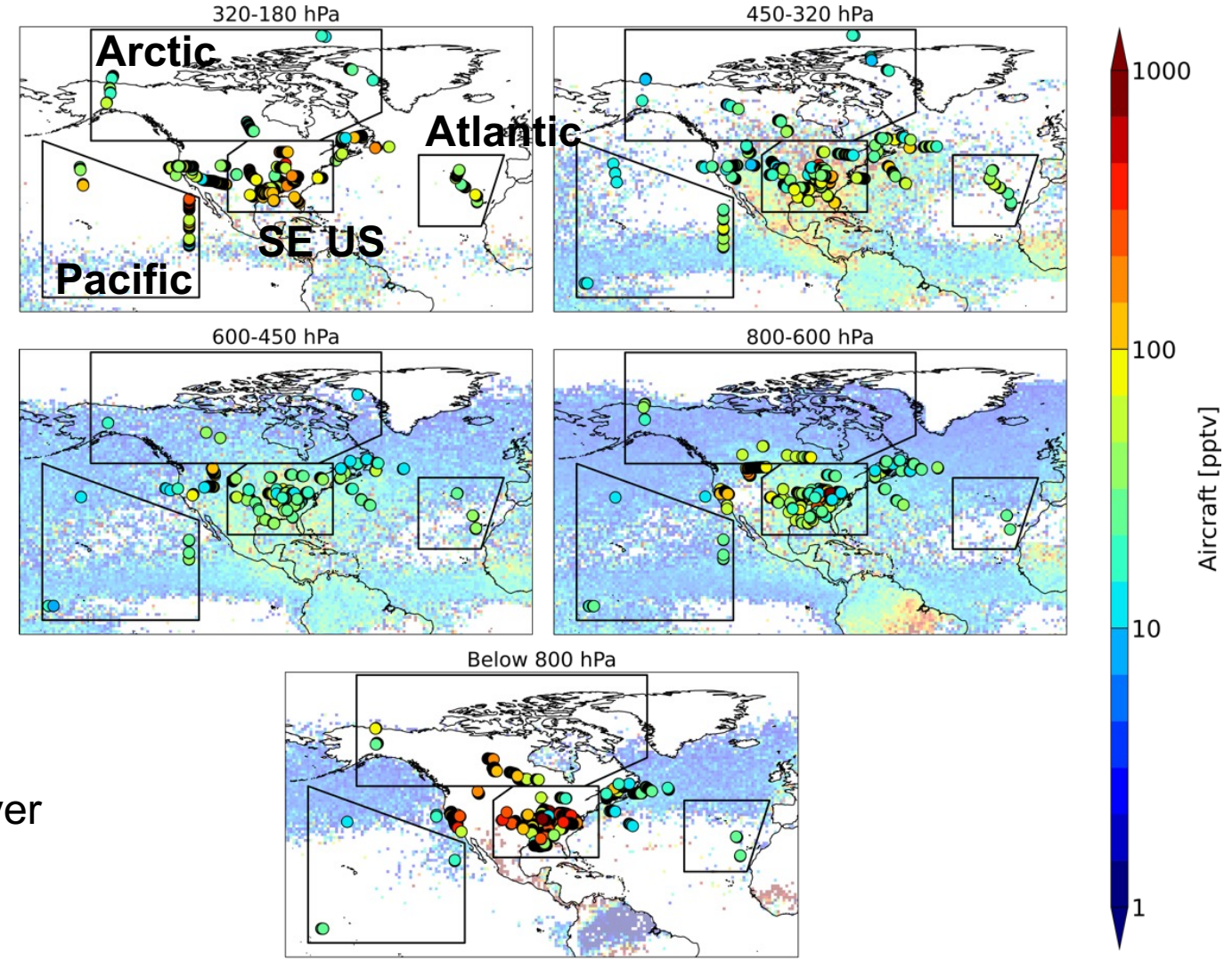
Assessment of Cloud-sliced NO_2

Use NASA DC8 aircraft observations for campaigns in 2008-2018

Upper troposphere

Middle troposphere

Boundary layer



Symbols: aircraft NO_2

Background: cloud-sliced NO_2

In situ data for different years

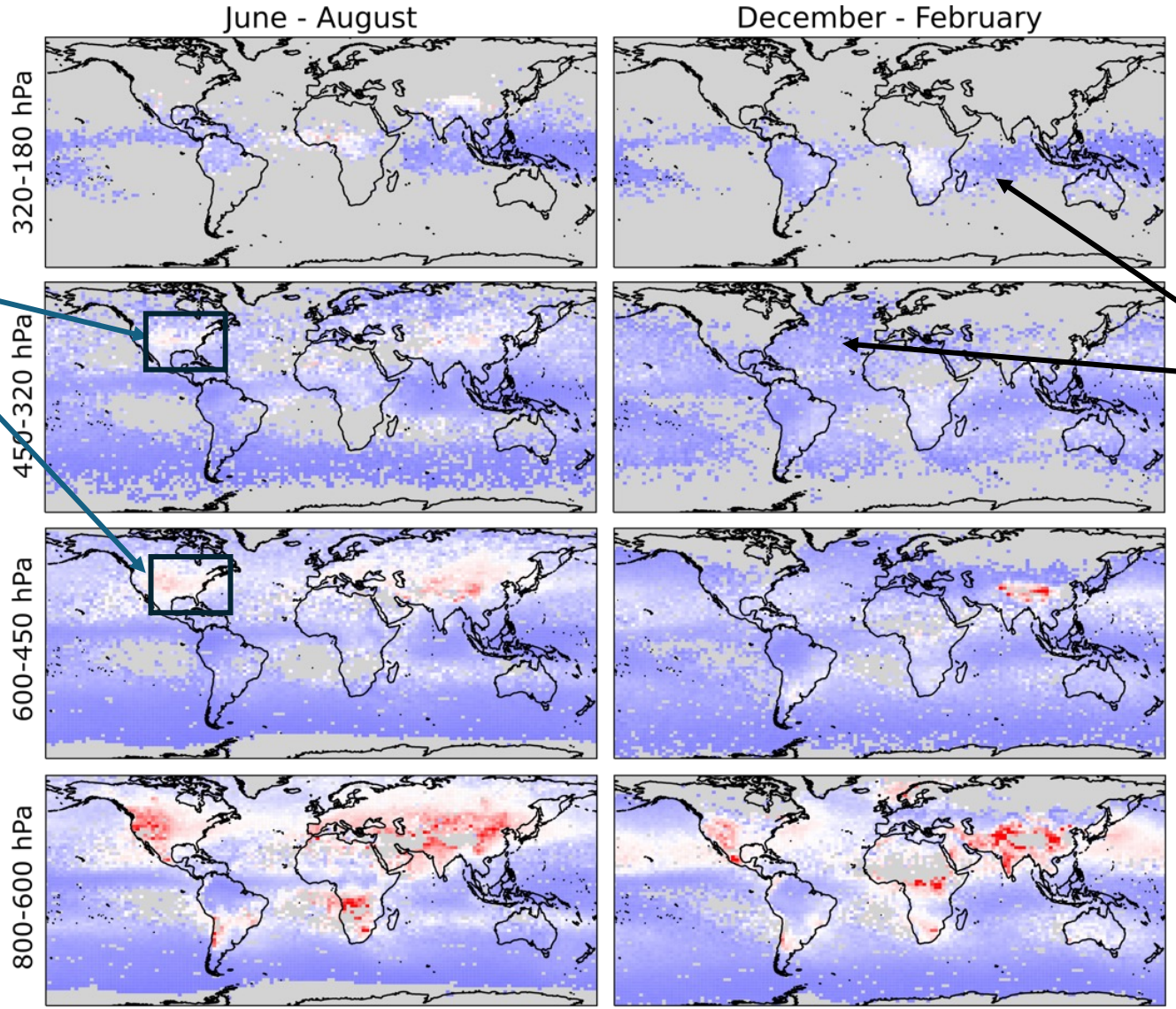
When spatial coverage similar, aircraft and cloud-sliced data difference < 10 pptv

But, few instances of coincidence

[Horner et al., *submitted* 2024]

Application of Cloud-sliced NO_2

Assess best understanding of tropospheric NO_2 as simulated by the GEOS-Chem model



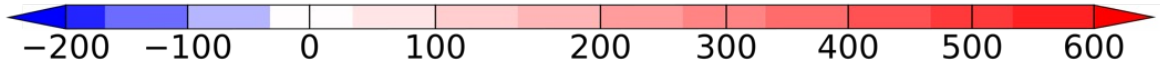
Model < Observed

Model > Observed

Model high bias over Southeast US (lightning NO_x)

Work underway to improve lightning NO_x emissions

Model low bias over remote regions

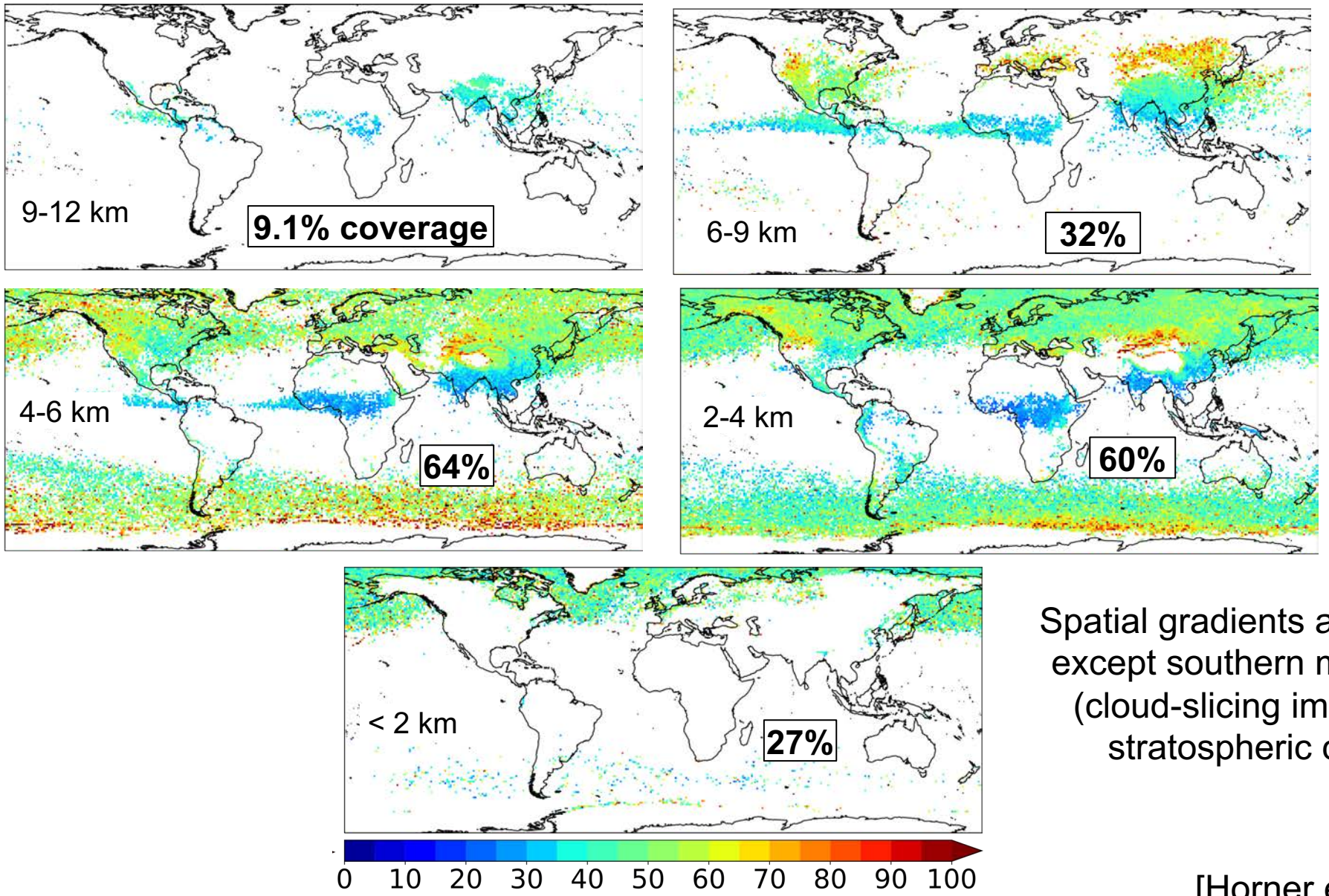


Model minus satellite [%]

[Horner et al., *submitted* 2024]

Extending Cloud-slicing to TROPOMI ozone

June-August multiyear mean ozone [ppbv]



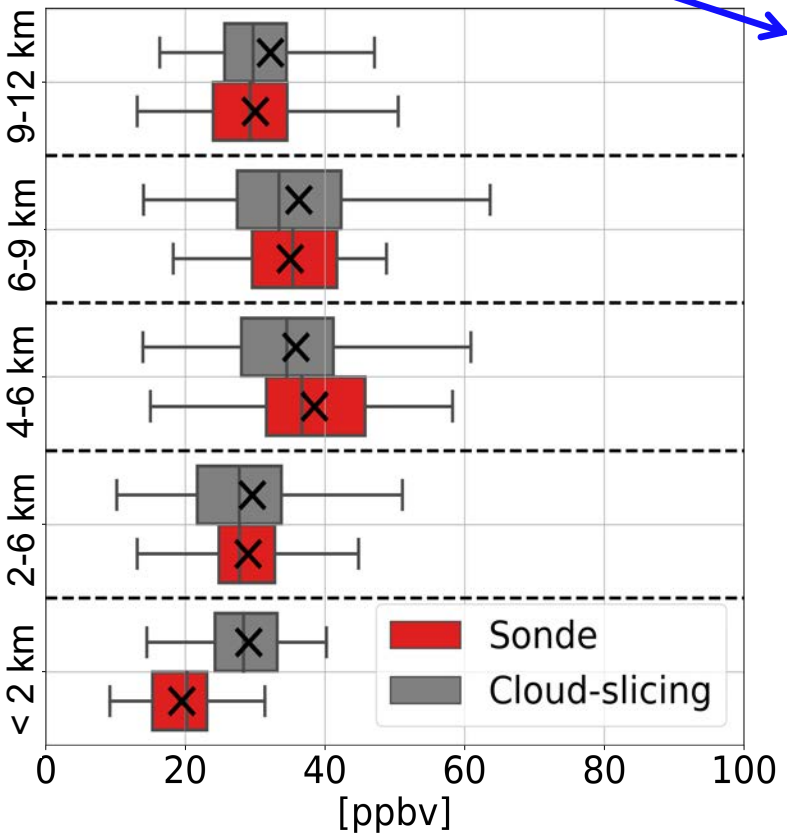
Spatial gradients as expected, except southern midlatitudes (cloud-slicing impacted by stratospheric column)

[Horner et al., *in progress*]

Assess Cloud-slicing Ozone with Ozonesondes

Ozonesonde network sites used

Samoa, JJA

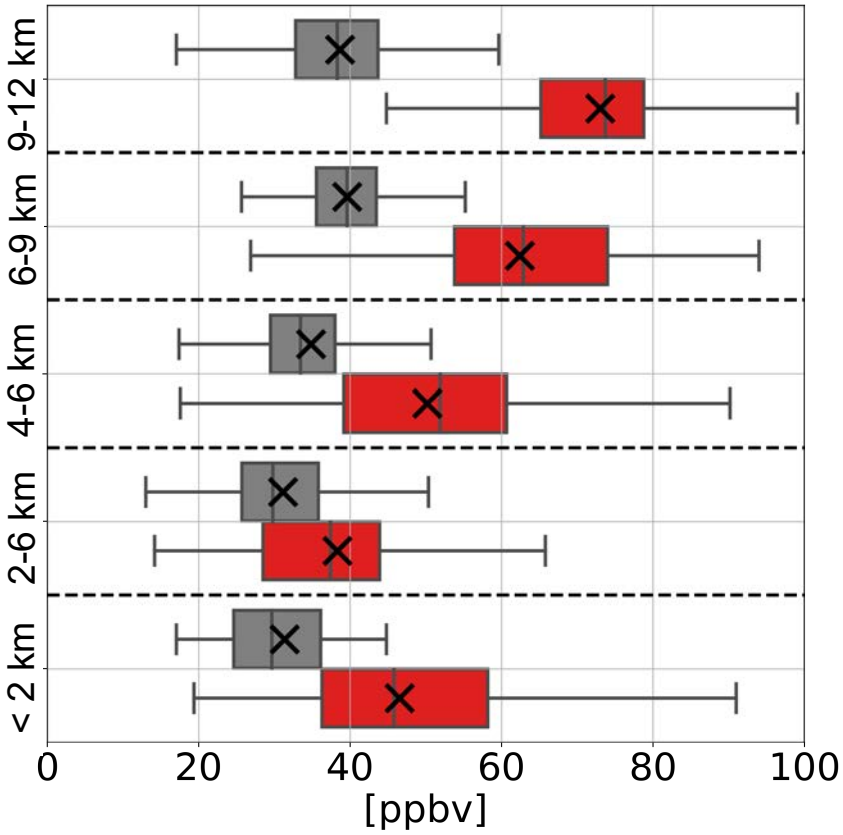


Ozonesondes with measurements during TROPOMI overpass

Most with good agreement like Samoa (left)

Only 3 with poor agreement like Hanoi (right)

Hanoi, JJA



Validation against ozonesondes encouraging

[Horner et al., *in progress*]

Opportunities & Challenges with GEO Instruments

- All cloud-sliced products are from LEO instruments, so only provide one snapshot (midday) of atmospheric composition
- <10% of original pixel data retained for cloud-slicing
- GEO offers greater number of observations and could be used to interrogate diurnal variability in NO_2 in distinct layers in the troposphere
- Need reliable, coincident, independent observations to validate cloud-sliced data

Resources:

Cloud-sliced vertical profiles of NO_2 data: https://doi.org/10.5522/04/25782336

Accompanying ACP preprint should appear on EGUsphere soon!

UCL-CSC Visiting research (PhD) student scheme:
https://www.ucl.ac.uk/scholarships/ucl-csc-visiting-research-student-award