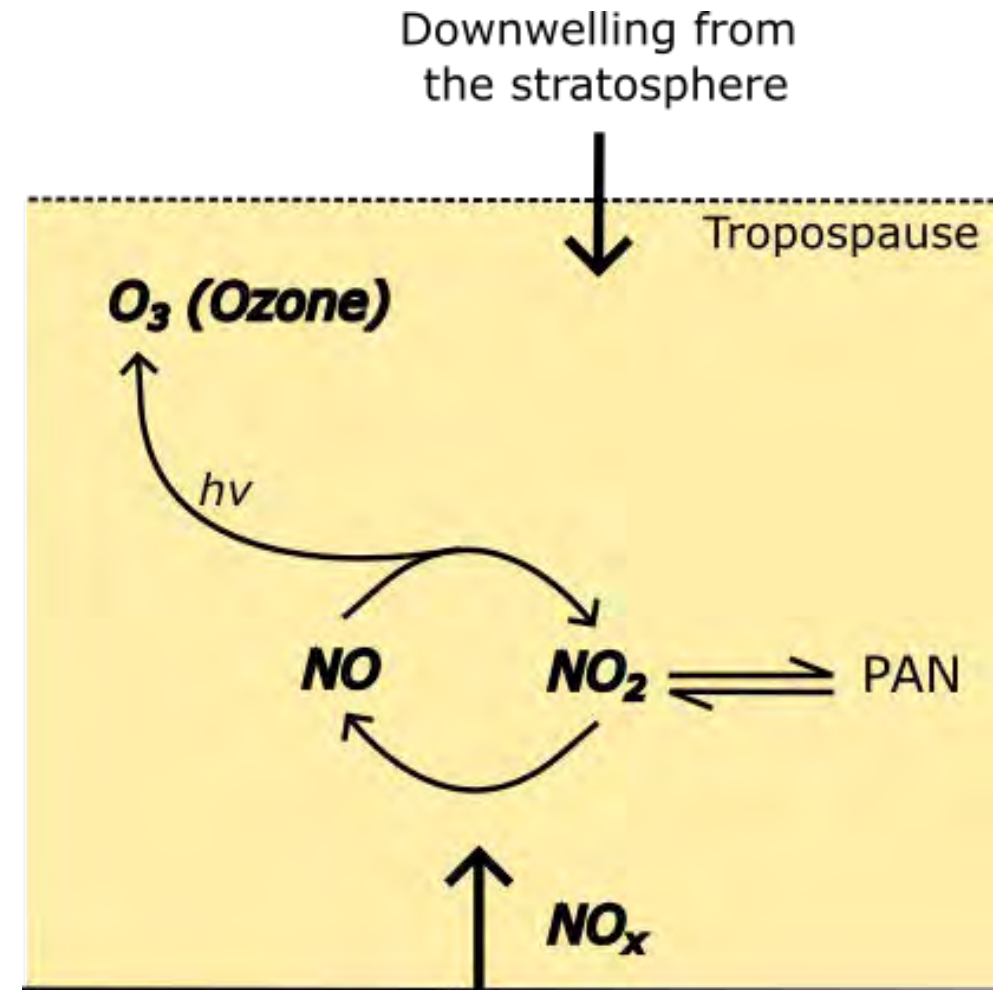
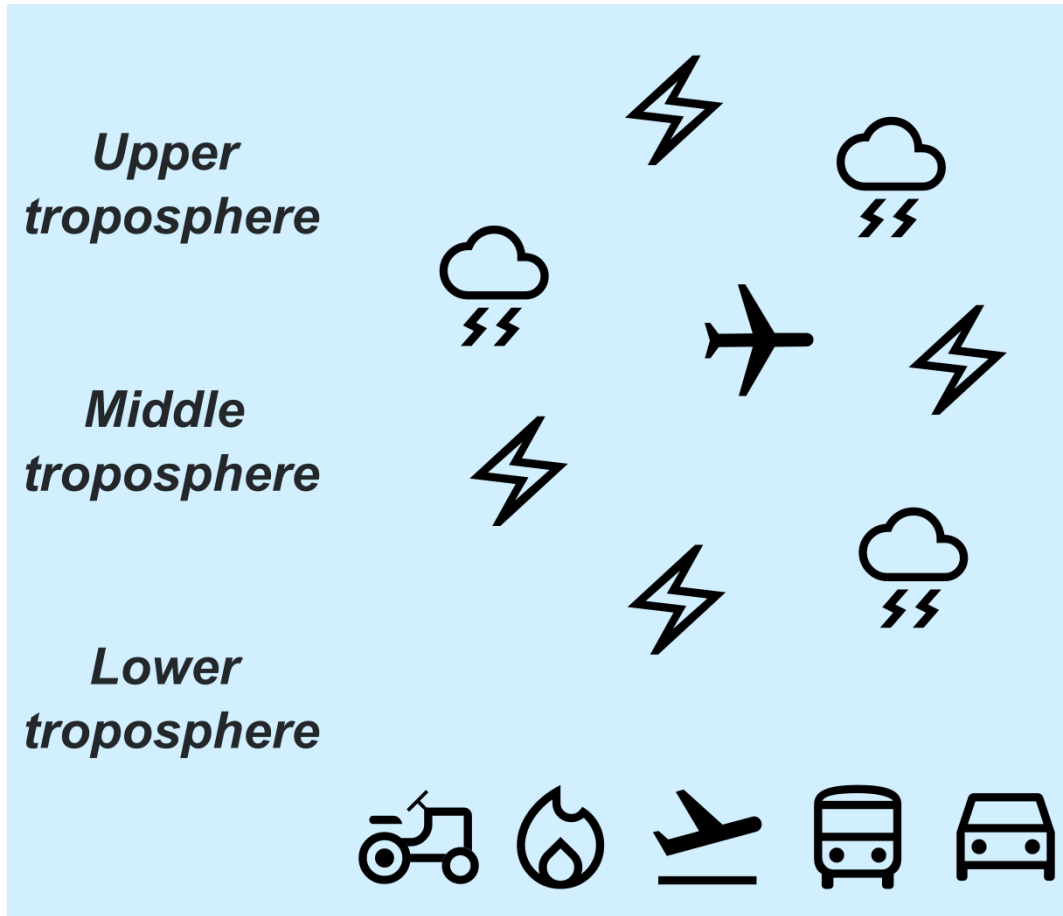


Evaluation of GEOS-Chem vertical profiles of nitrogen dioxide and ozone using cloud-sliced TROPOMI columns

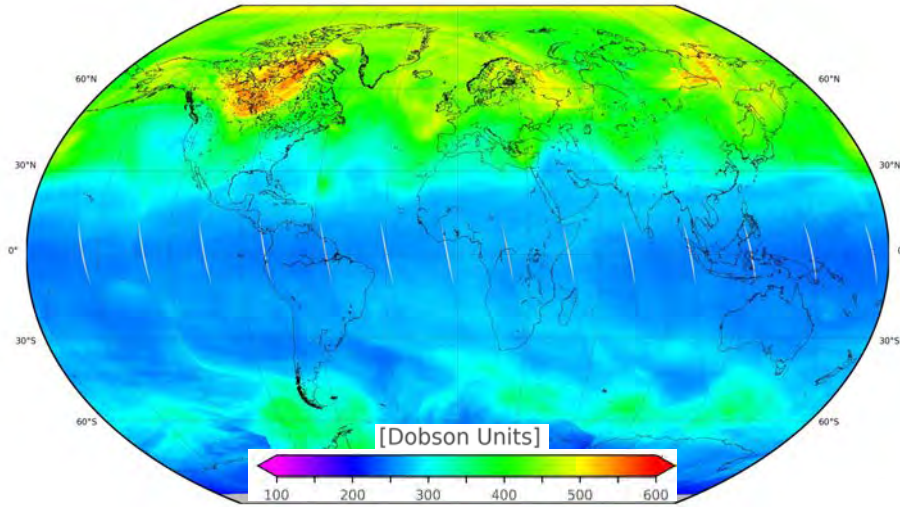
NO_x plays an important role in the formation of O_3 in the troposphere



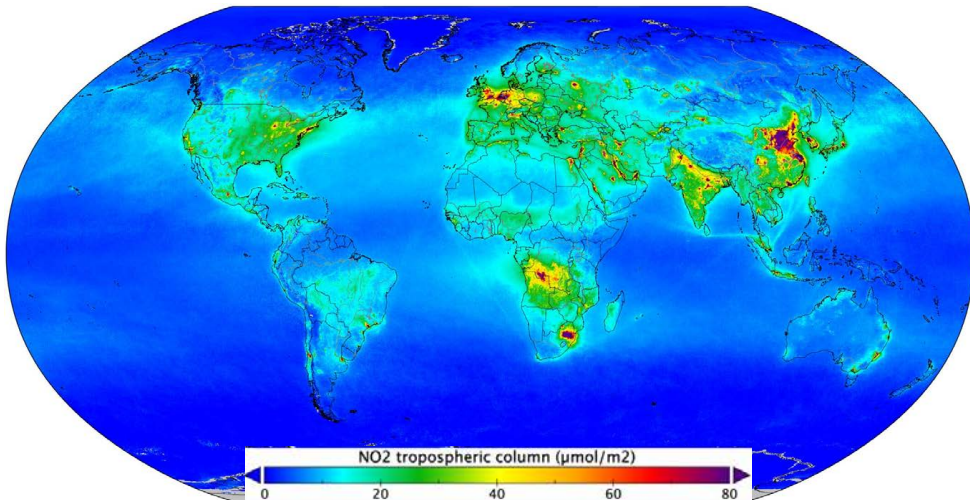
Lifetime of NO_x increases with altitude \rightarrow NO_x has a large influence on tropospheric ozone

Current observations of NO₂ and O₃ vertical profiles are limited

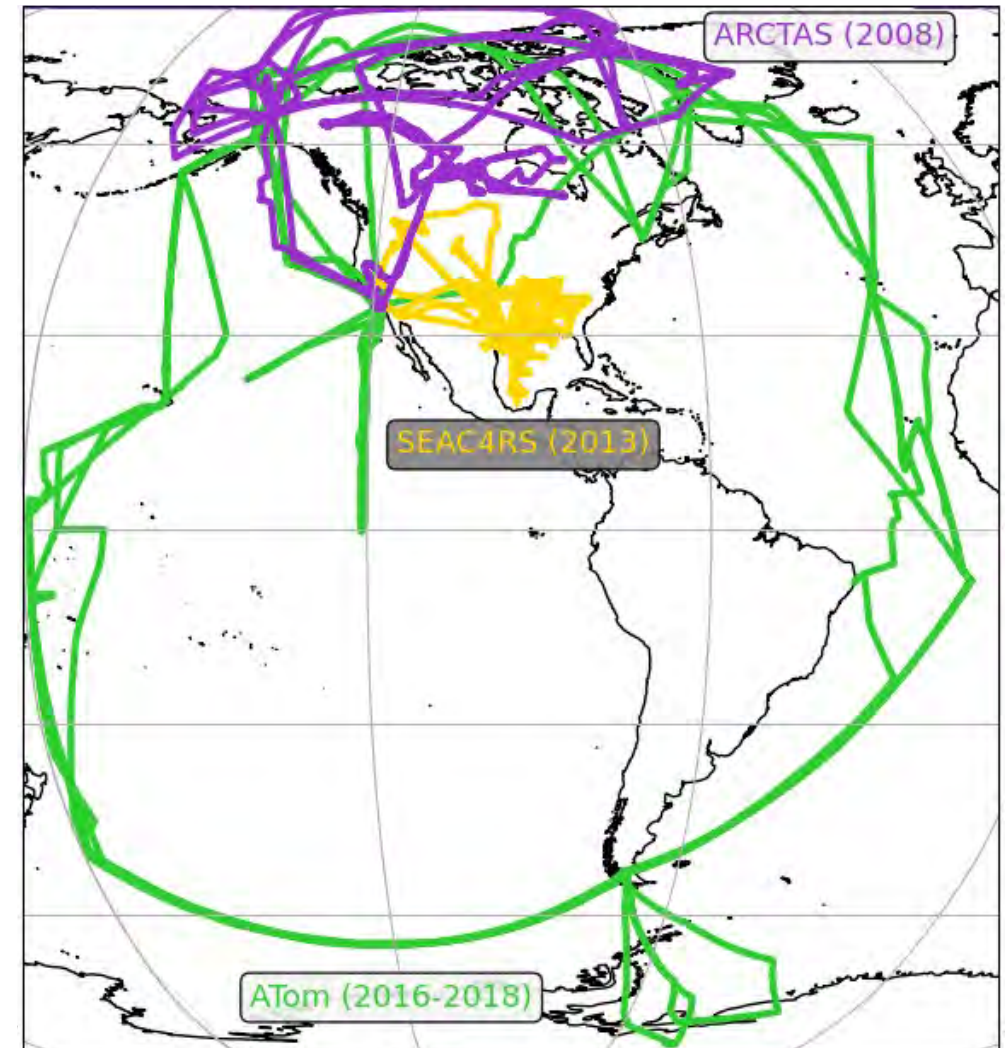
Total ozone column, 29th March 2018



Total nitrogen dioxide column, April-September 2018

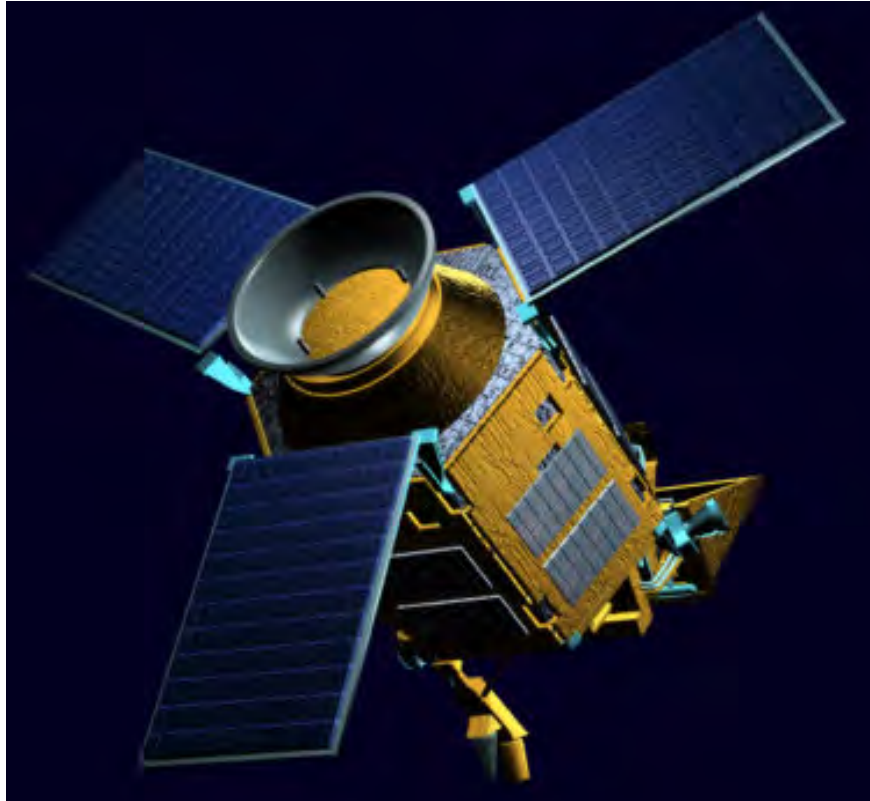


Sample of NASA DC8 aircraft data

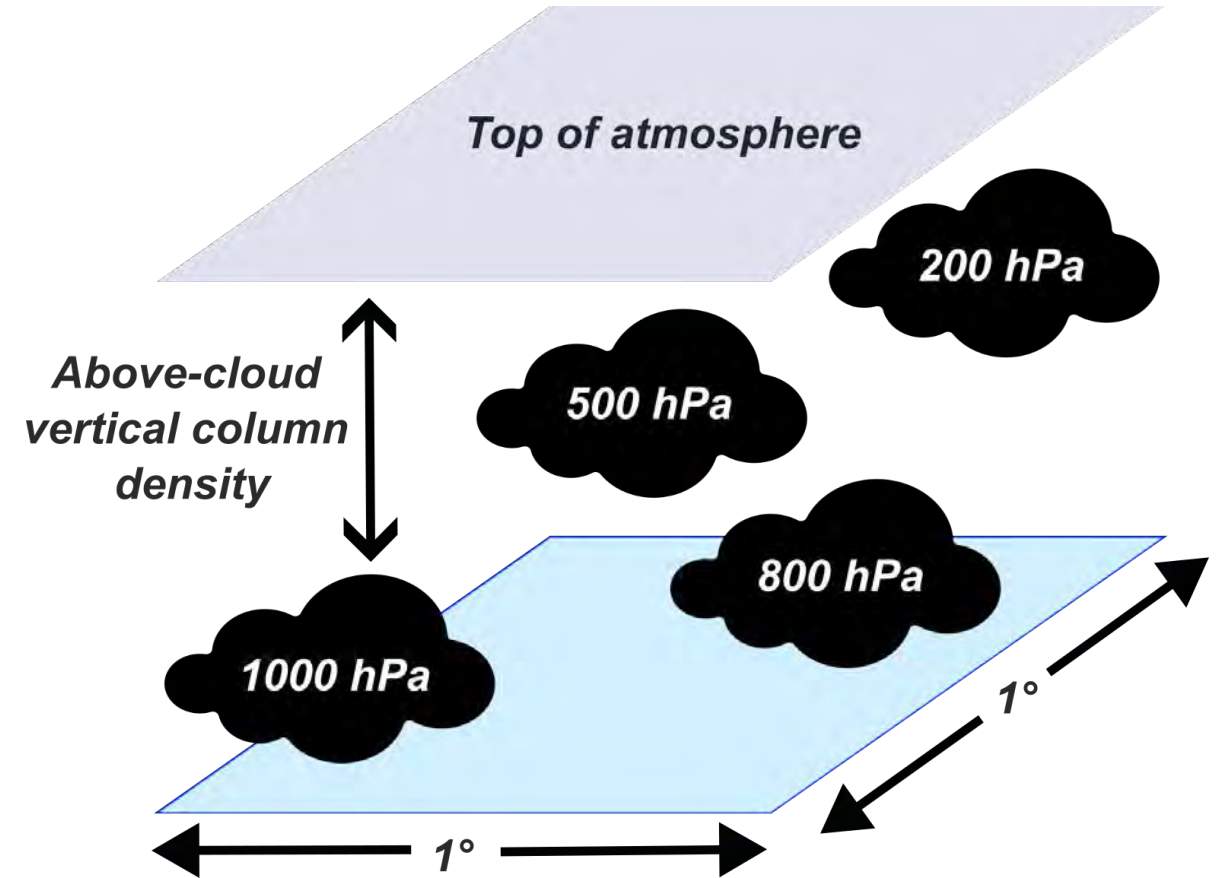


We use cloud-slicing to look at the vertical distribution from satellite measurements

TROPOMI



Launched 13th October 2017
Spatial resolution of 5.5 km x 3.5 km



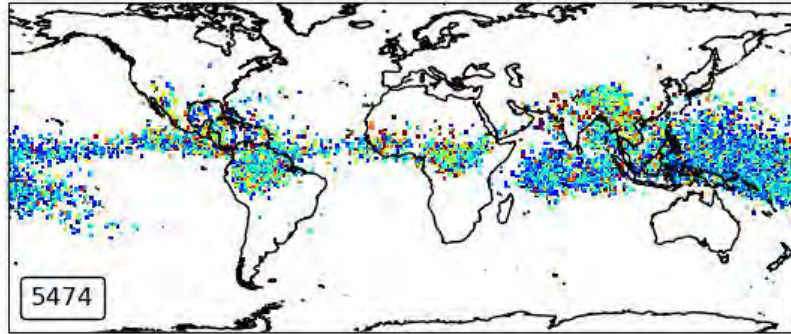
$$\text{VMR} \propto \frac{\Delta \text{ vertical column density}}{\Delta \text{ cloud-top pressure}}$$

NO₂ cloud-slicing results

NO₂ vertical profiles from cloud-slicing of TROPOMI data

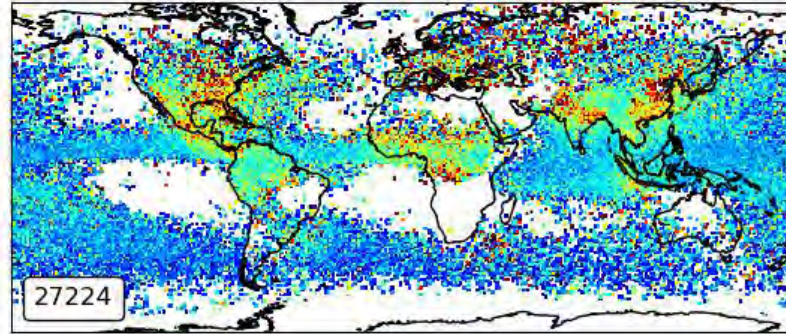
Multiyear seasonal mean for JJA 2018-2021 at a resolution of 1° x 1°

9-12 km (320-180 hPa)



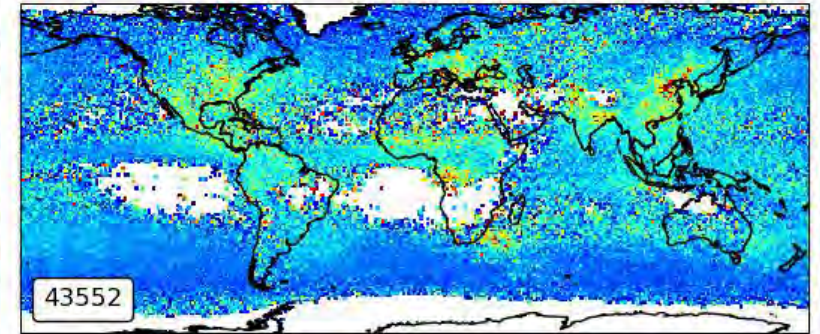
8% coverage

6-9 km (450-320 hPa)



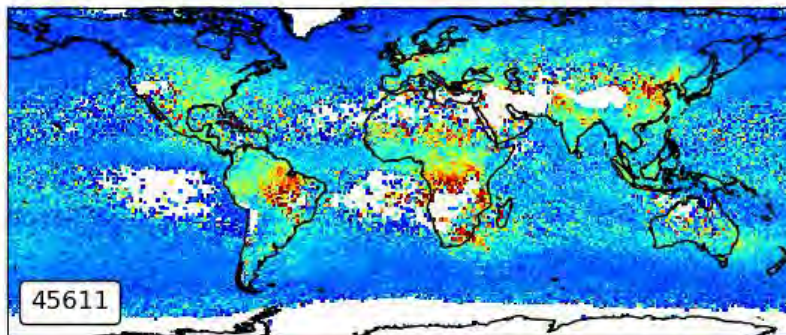
42% coverage

4-6 km (600-450 hPa)



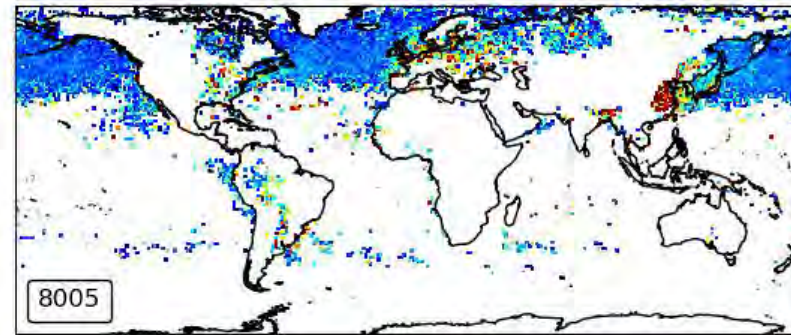
67% coverage

2-4 km (800-600 hPa)



70% coverage

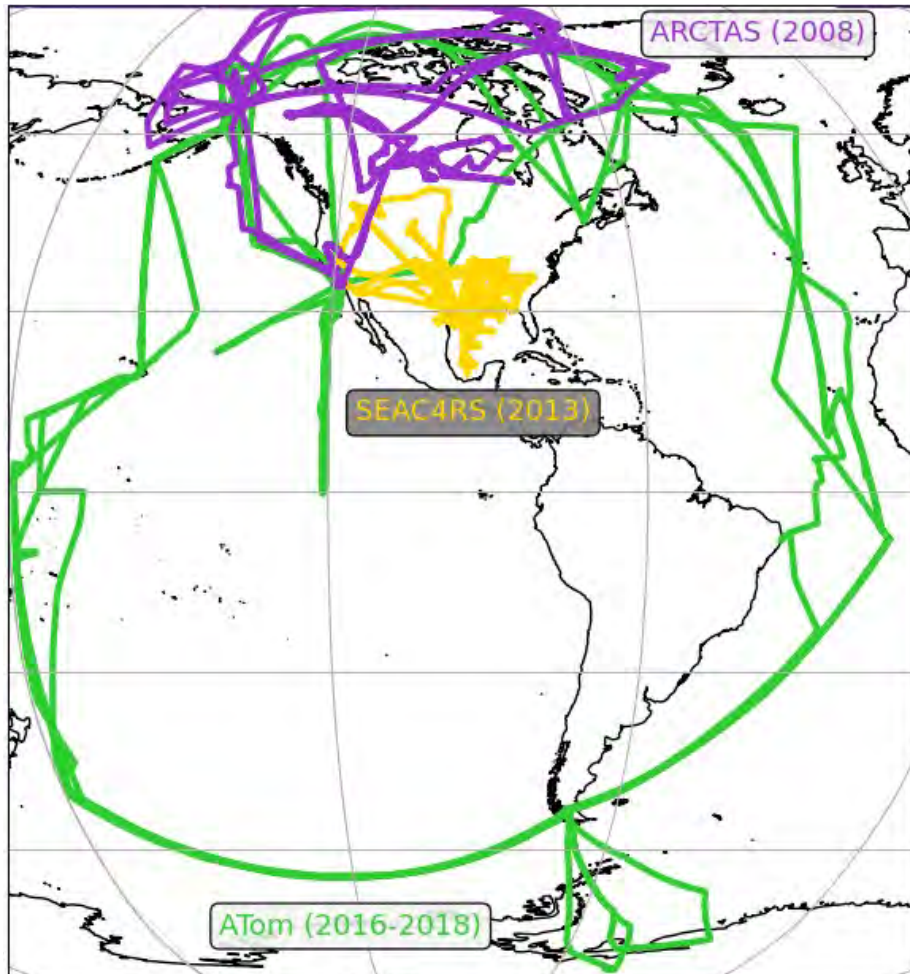
< 2 km (1100-800 hPa)



12% coverage



We use aircraft observations to compare to cloud-slicing results



SEAC⁴RS – Central US, summer 2013



ATom – Remote Pacific & Atlantic, once in all 4 seasons from 2016 to 2018



ARCTAS – Canada & Arctic Circle, spring and summer 2008

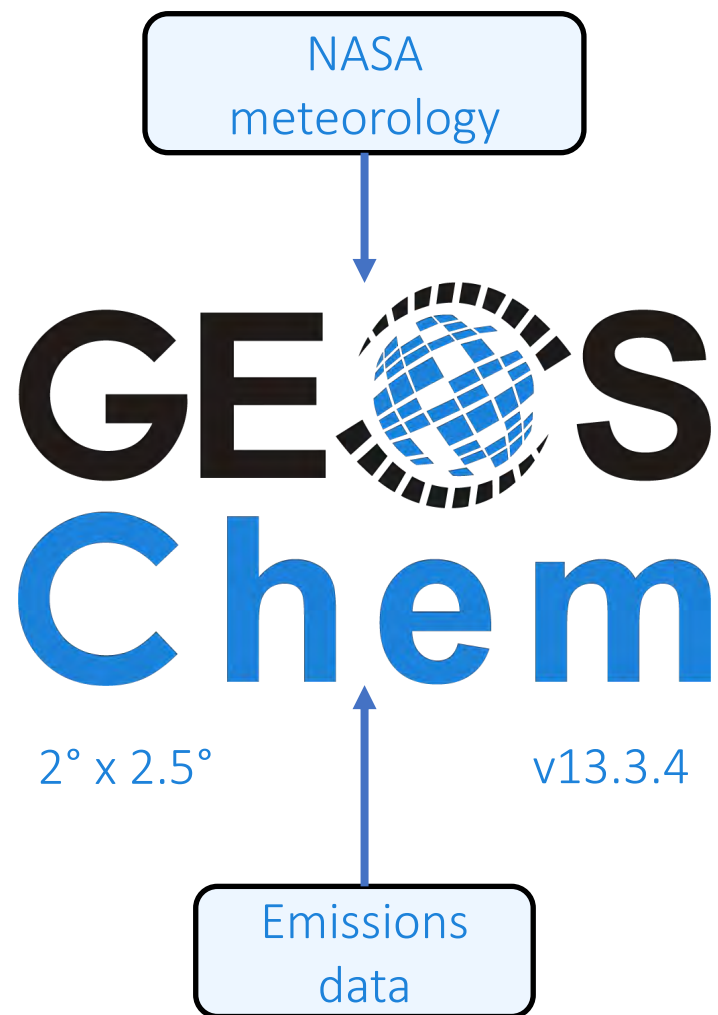
Aircraft measurements are filtered where:

1. The local solar time is similar to the TROPOMI overpass time
2. NO measurements are 2x the instrument detection limit

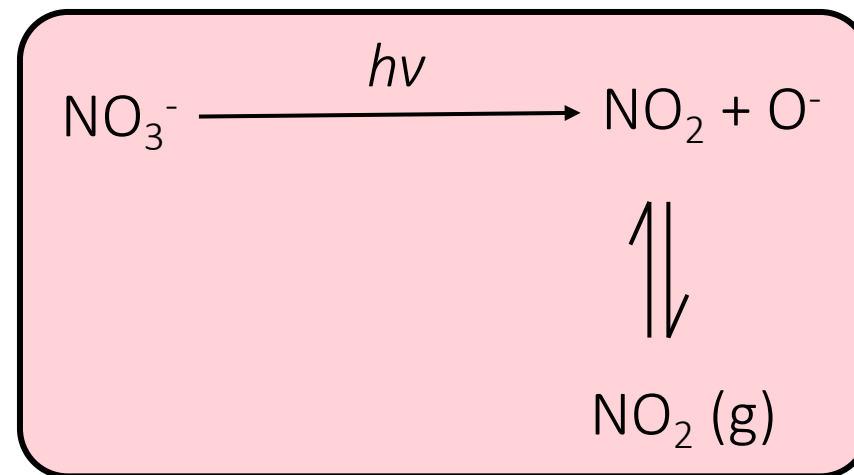
$$PSS = \frac{[NO]}{[NO_2]} \approx \frac{j_{NO_2}}{k_1[O_3] + k_2[HO_2]} \approx \frac{j_{NO_2}}{k_1[O_3]}$$

GEOS-Chem is updated to include nitrate photolysis

Seasonal multiyear means 2015-2019



NITRATE PHOTOLYSIS



NITRATE PHOTOLYSIS RATE

$$\frac{\text{JHNO}_3}{\text{JNIT}} \times 100$$

FINE MODE NITRATES

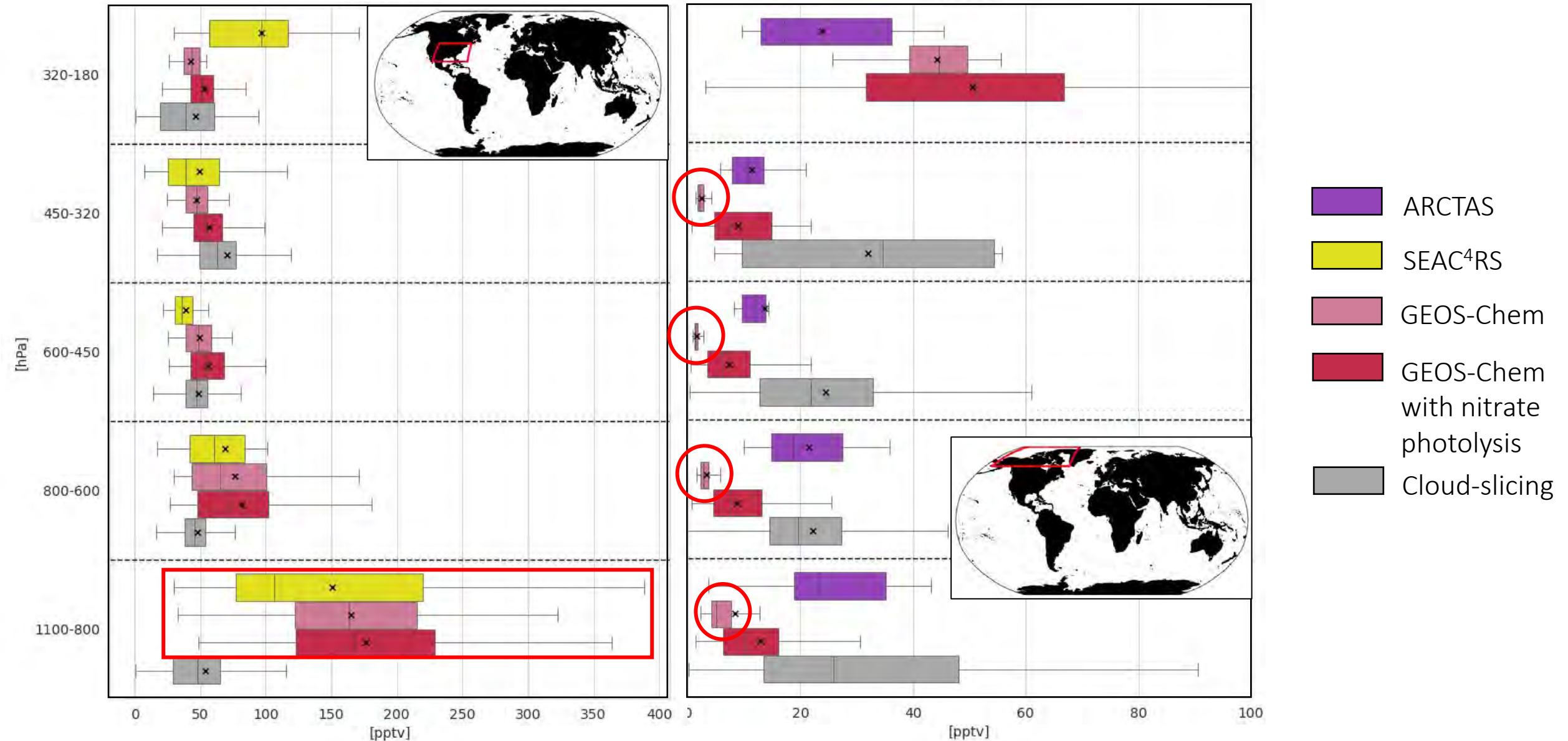
$$\frac{[\text{SSA}]}{[\text{SSA}] + [\text{pNO}_3^-]}$$

[Dang et al., 2022, Shah et al., 2022]

Comparison of NO₂ cloud-slicing to GEOS-Chem and aircraft observations

Eastern US, JJA

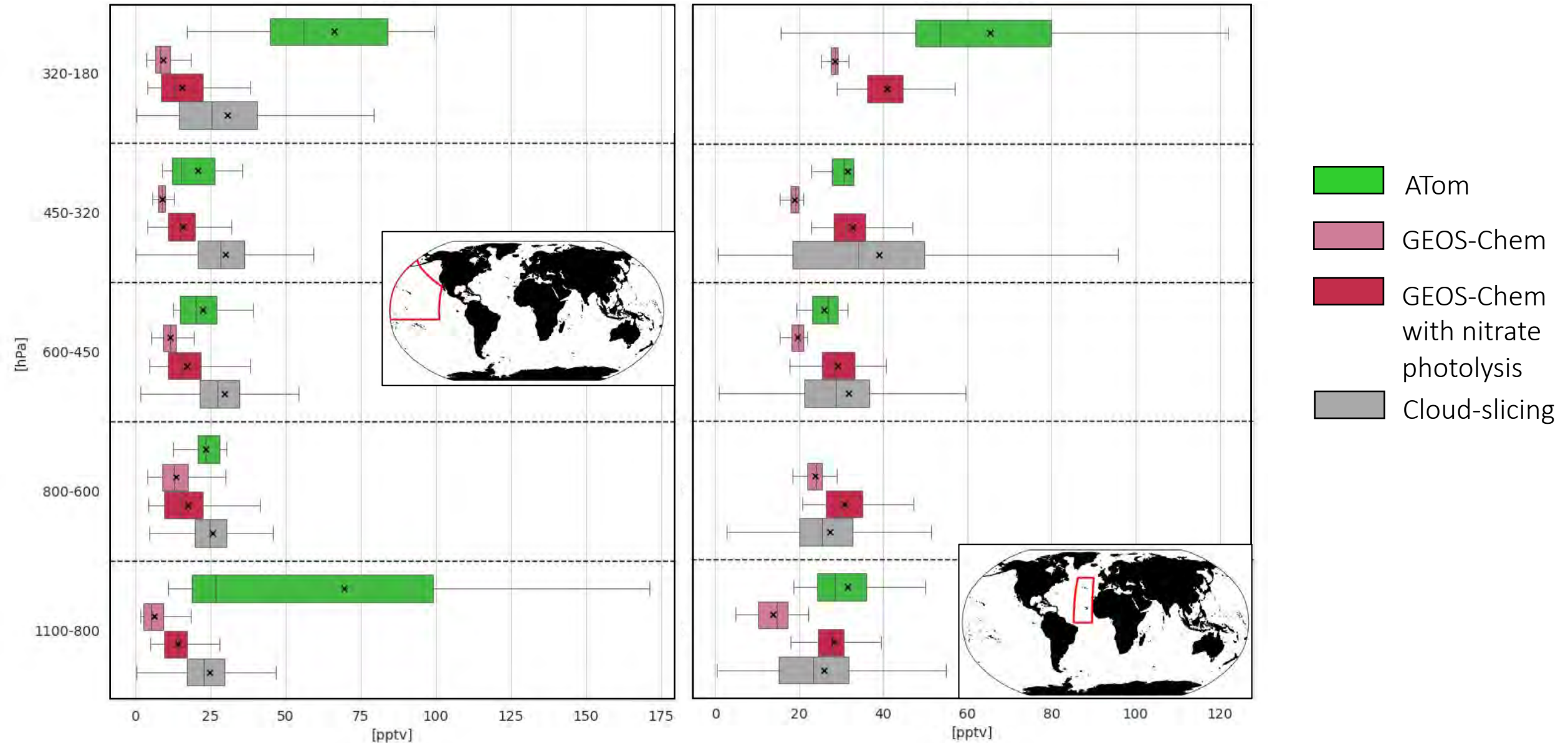
Arctic, MAM



Comparison of NO₂ cloud-slicing to GEOS-Chem and aircraft observations

Pacific, JJA

North Atlantic, JJA

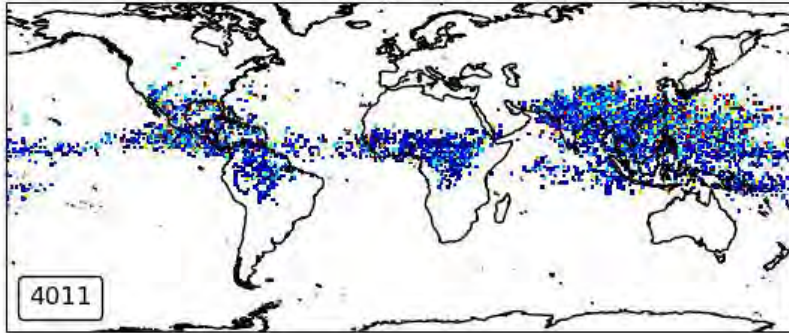


O₃ cloud-slicing results

O₃ vertical profiles from cloud-slicing TROPOMI data

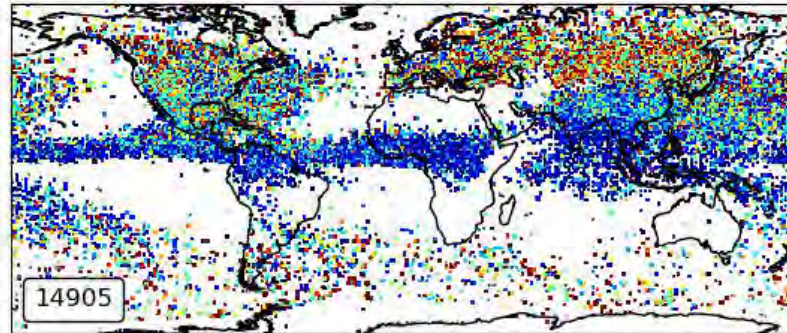
Multiyear seasonal mean for JJA 2020-2022 at a resolution of 1° x 1°

9-12 km (320-180 hPa)



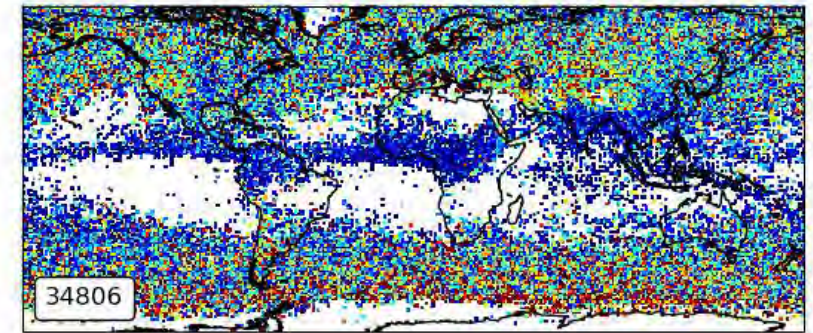
6% coverage

6-9 km (450-320 hPa)



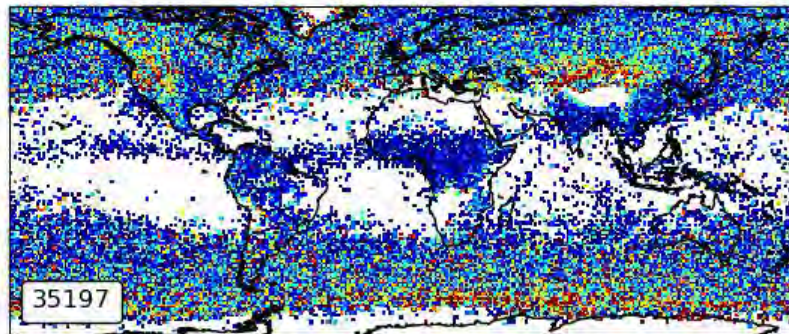
23% coverage

4-6 km (600-450 hPa)



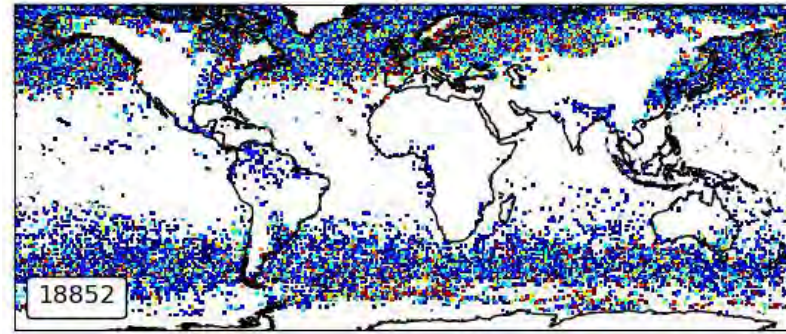
53% coverage

2-4 km (800-600 hPa)



54% coverage

< 2 km (1100-800 hPa)



29% coverage

O₃ [ppbv]



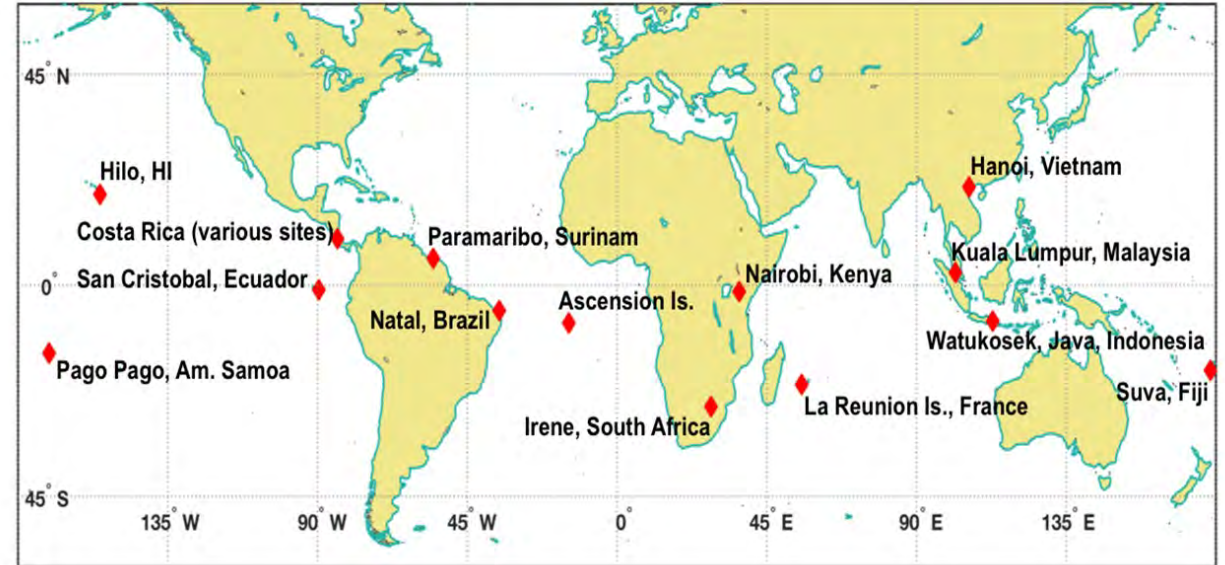
We use ozonesonde measurements to compare to cloud-slicing

Ozonesonde measurements



Measures vertical distribution of atmospheric ozone up to 30-35 km

SHADOZ ozonesonde network

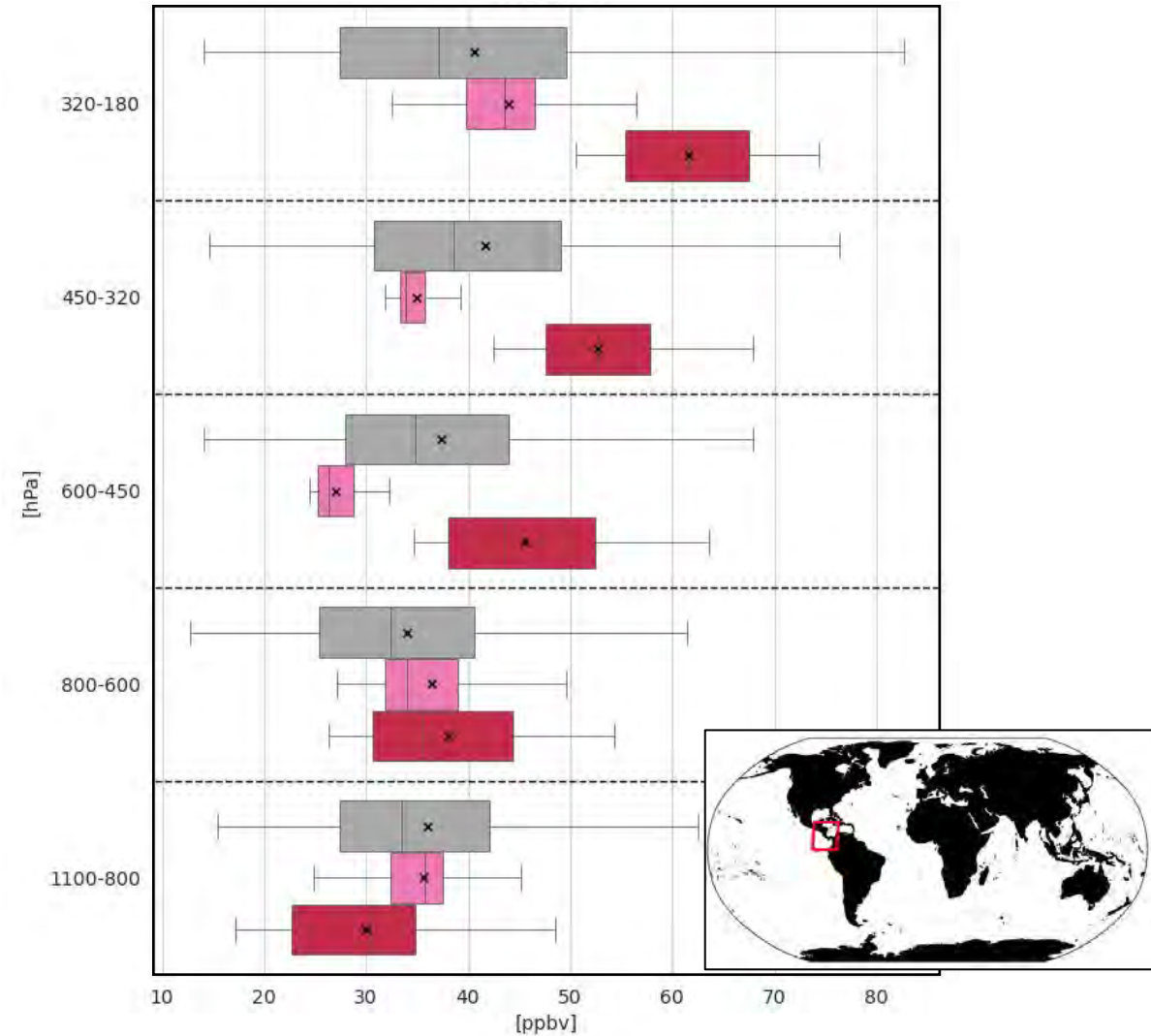


SHADOZ is primarily focused around the tropics and subtropics

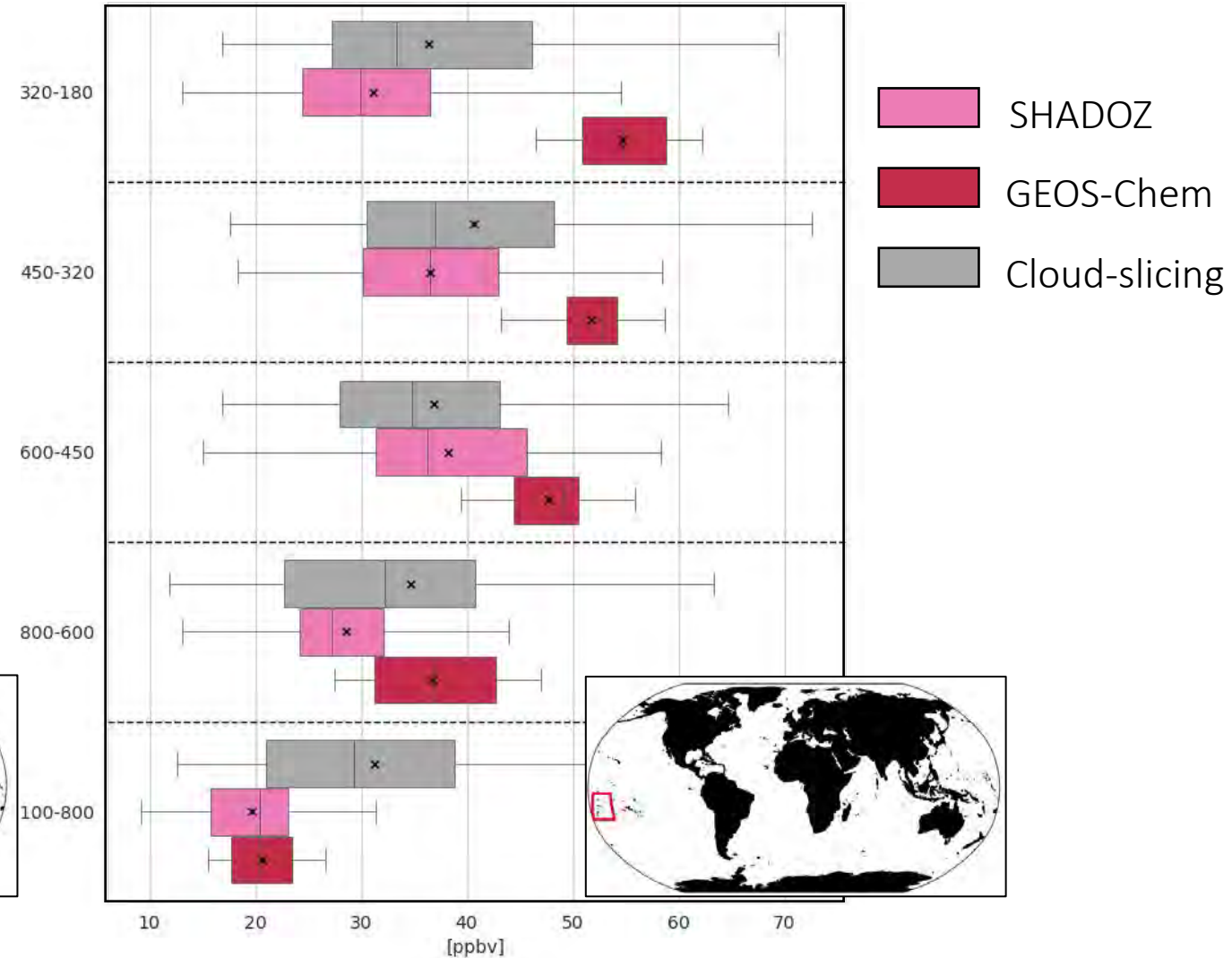
We filter ozonesonde data where local solar time is similar to the TROPOMI overpass time

GEOS-Chem overestimates concentrations at individual ozonesonde sites

Costa Rica, JJA



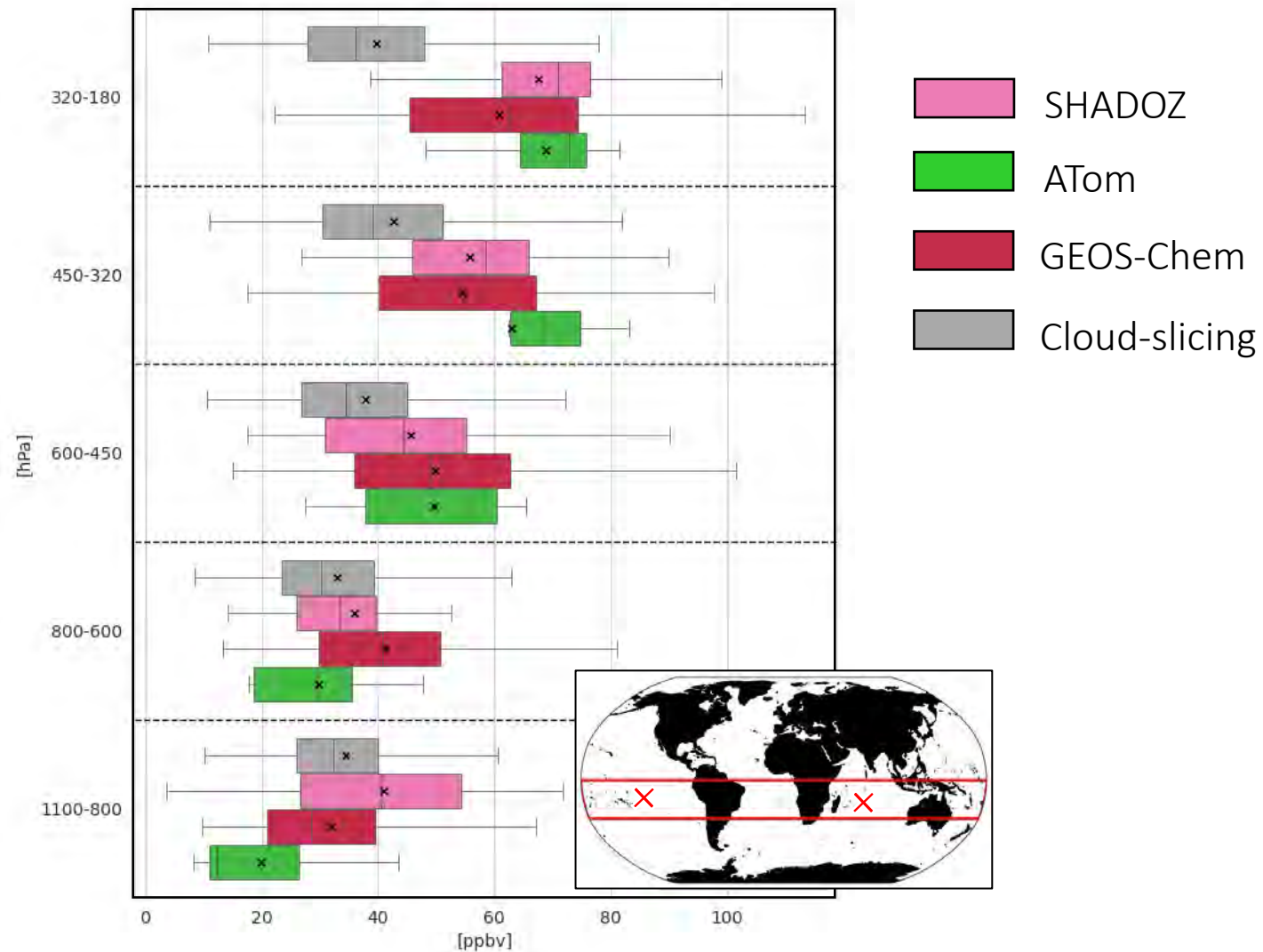
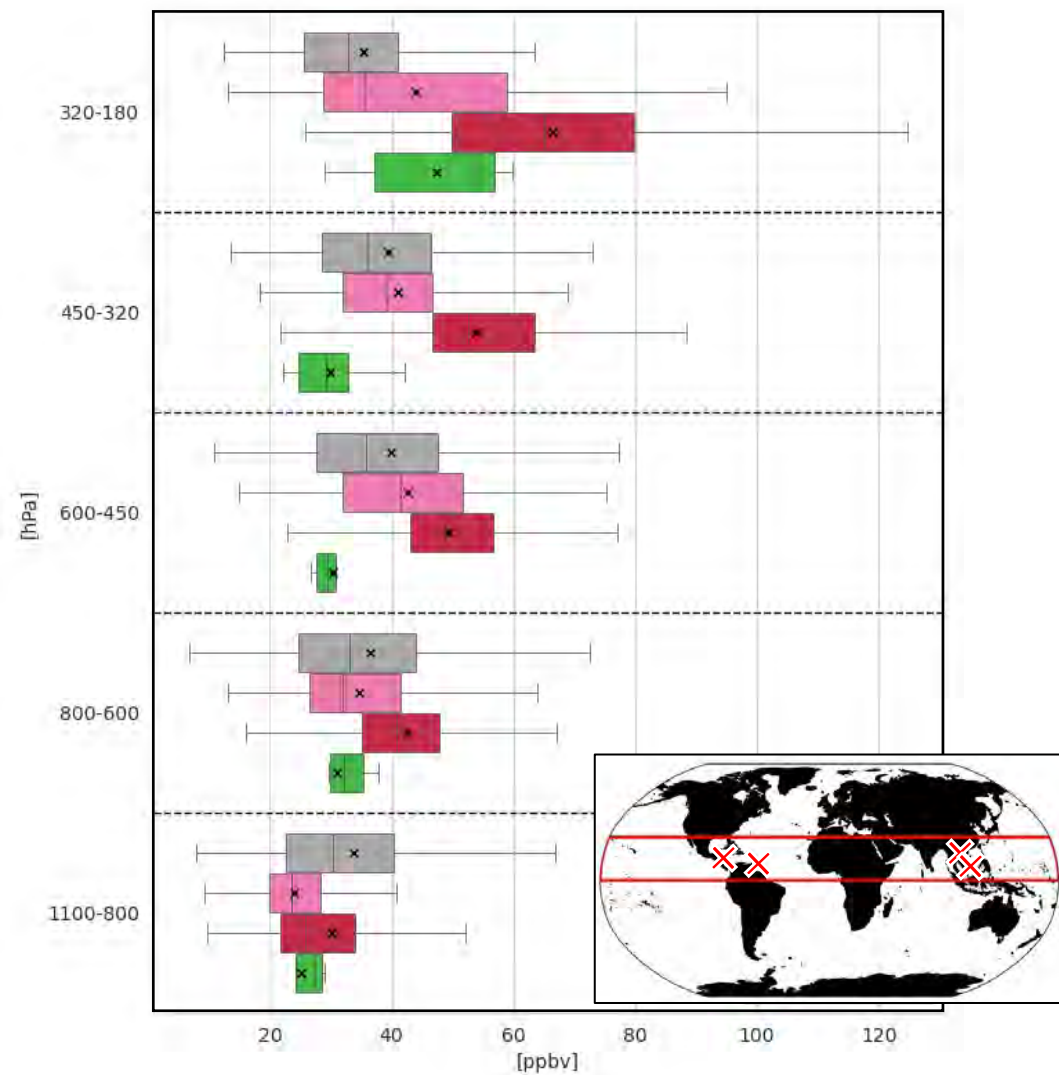
Samoa, JJA



There is good agreement between datasets cross latitude bands

0°N – 30°N, JJA

0°S – 30°S, JJA



Concluding remarks

- The cloud-slicing technique **improves global coverage** of NO_2 and O_3 vertical profiles
- **Cloud-slicing underestimates NO_2 concentrations in the urban terrestrial boundary layer** due to large land-based anthropogenic pollution sources
- **GEOS-Chem underestimates NO_2 concentrations** in the remote troposphere by as much as 20 pptv → this is improved by including nitrate photolysis in simulations.
- **Cloud-sliced O_3 and SHADOZ measurements are in good agreement** in the Northern Hemisphere however in some regions there is an overestimate from GEOS-Chem