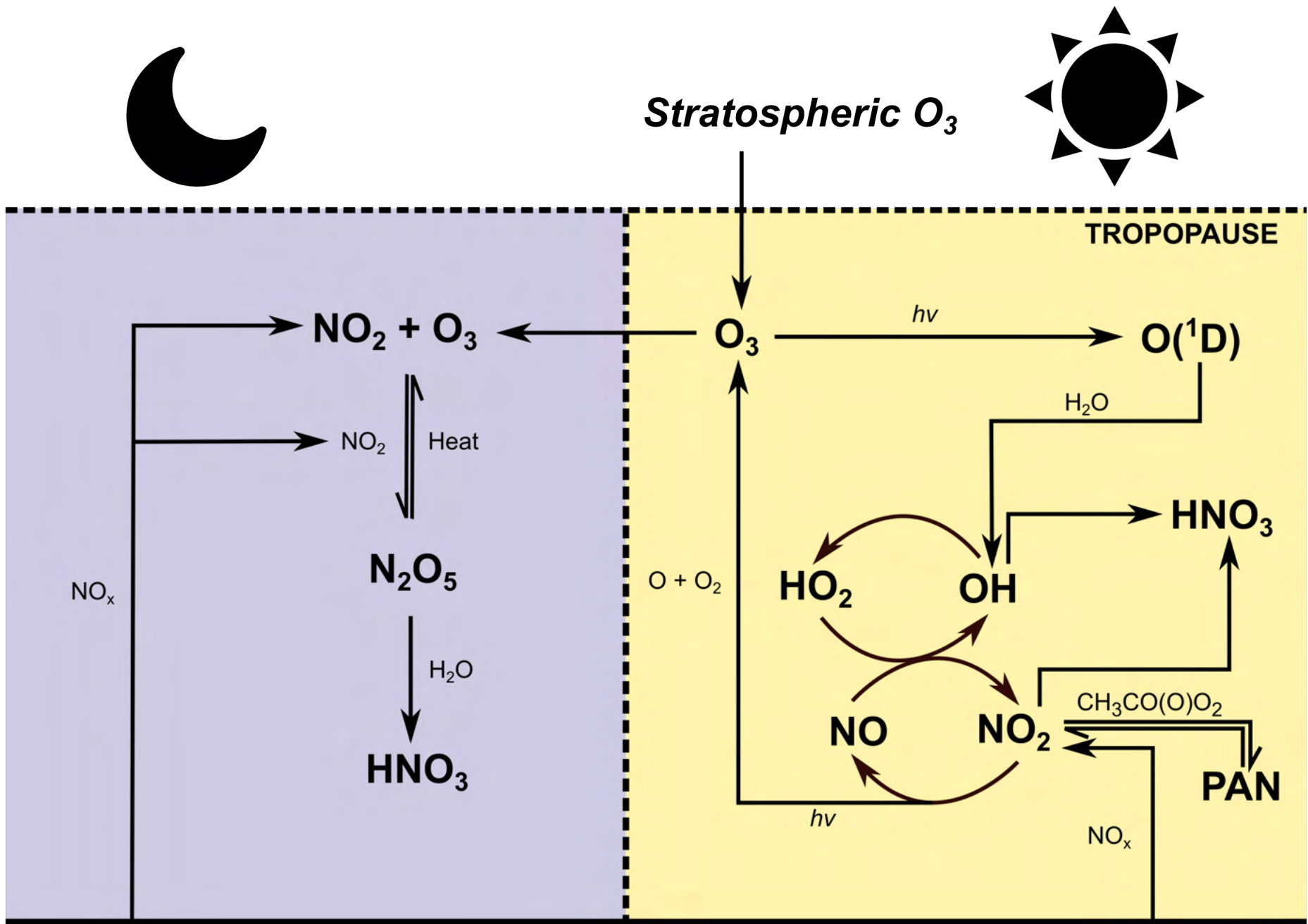


Major Finding: Cloud-slicing of TROPOMI NO₂ performs well globally between 320-800 hPa when compared to NASA DC-8 aircraft observations and GEOS-Chem underestimates by ~60% in this region of the troposphere.

1. Motivation

Cycle of NO_x in the troposphere during the day and at night

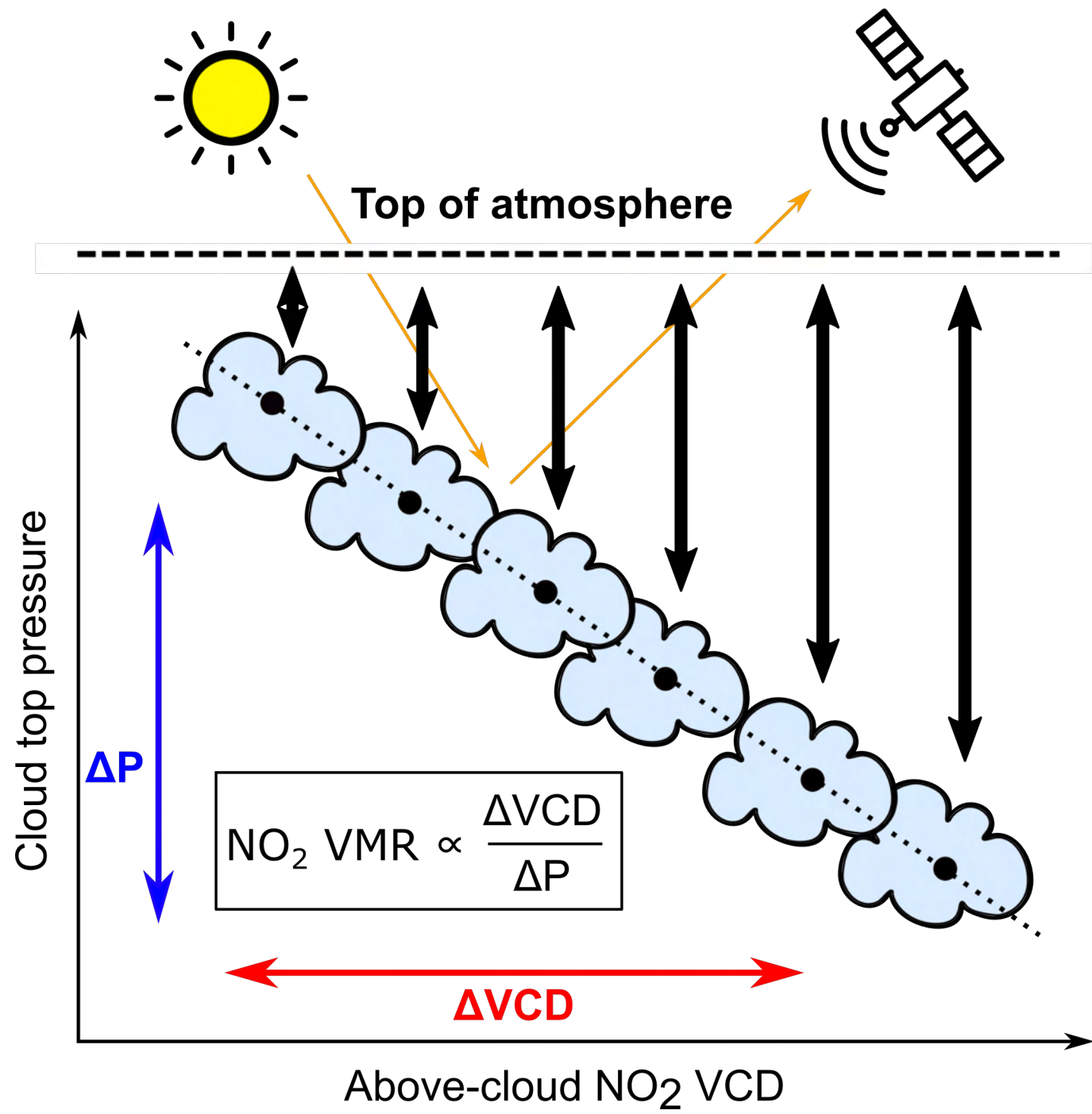


- Nitrogen oxides (NO_x ≡ NO + NO₂) are strongly linked to the formation of ozone (O₃) in the NO_x-limited regions of the troposphere.
- O₃ is a key contributor to the oxidation state of the atmosphere and maintaining the oxidation capacity of the troposphere.
- Well-mixed greenhouse gases have been responsible for a **radiative forcing of 2.45 W/m²** and **O₃ is responsible for 26%** of this.

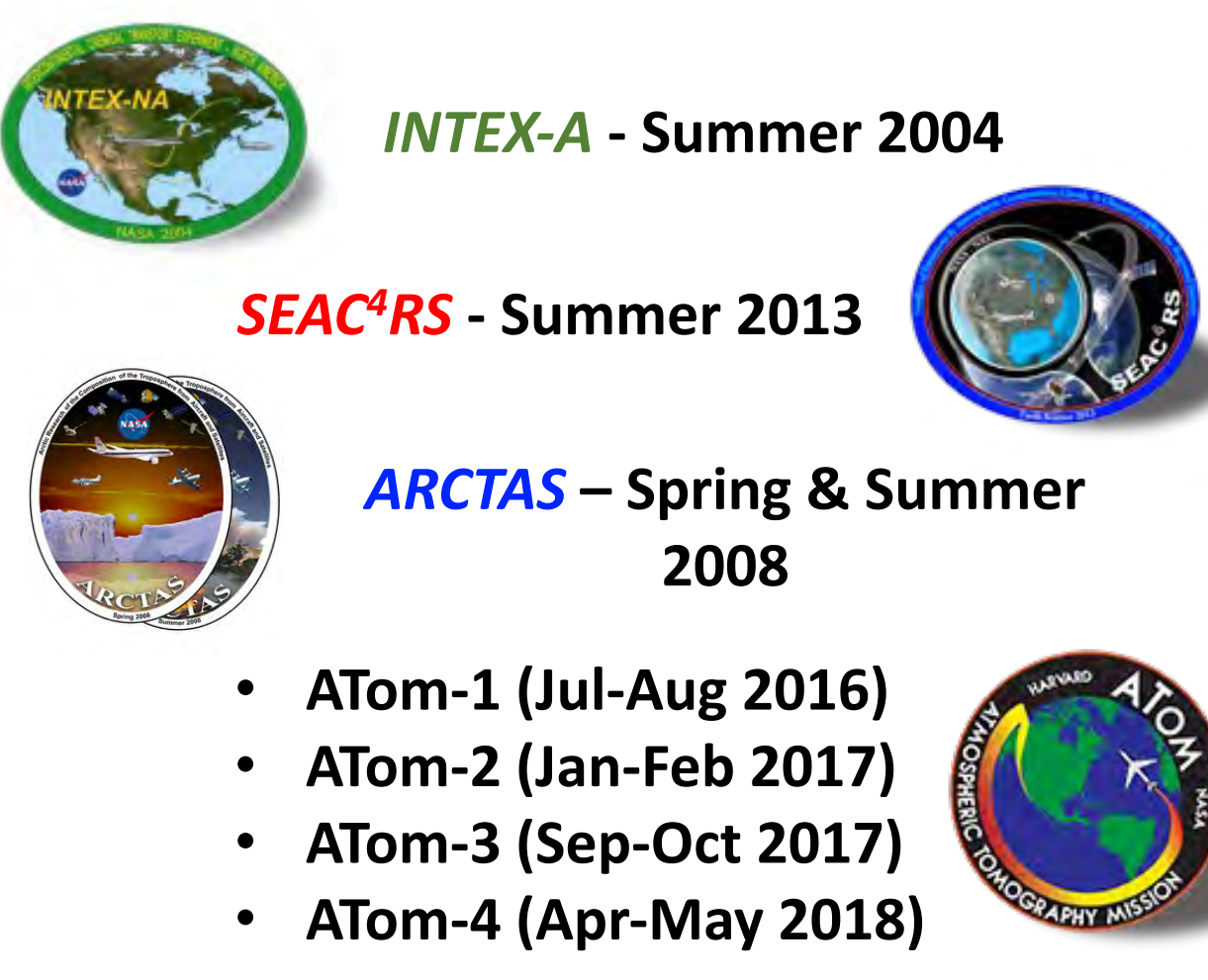
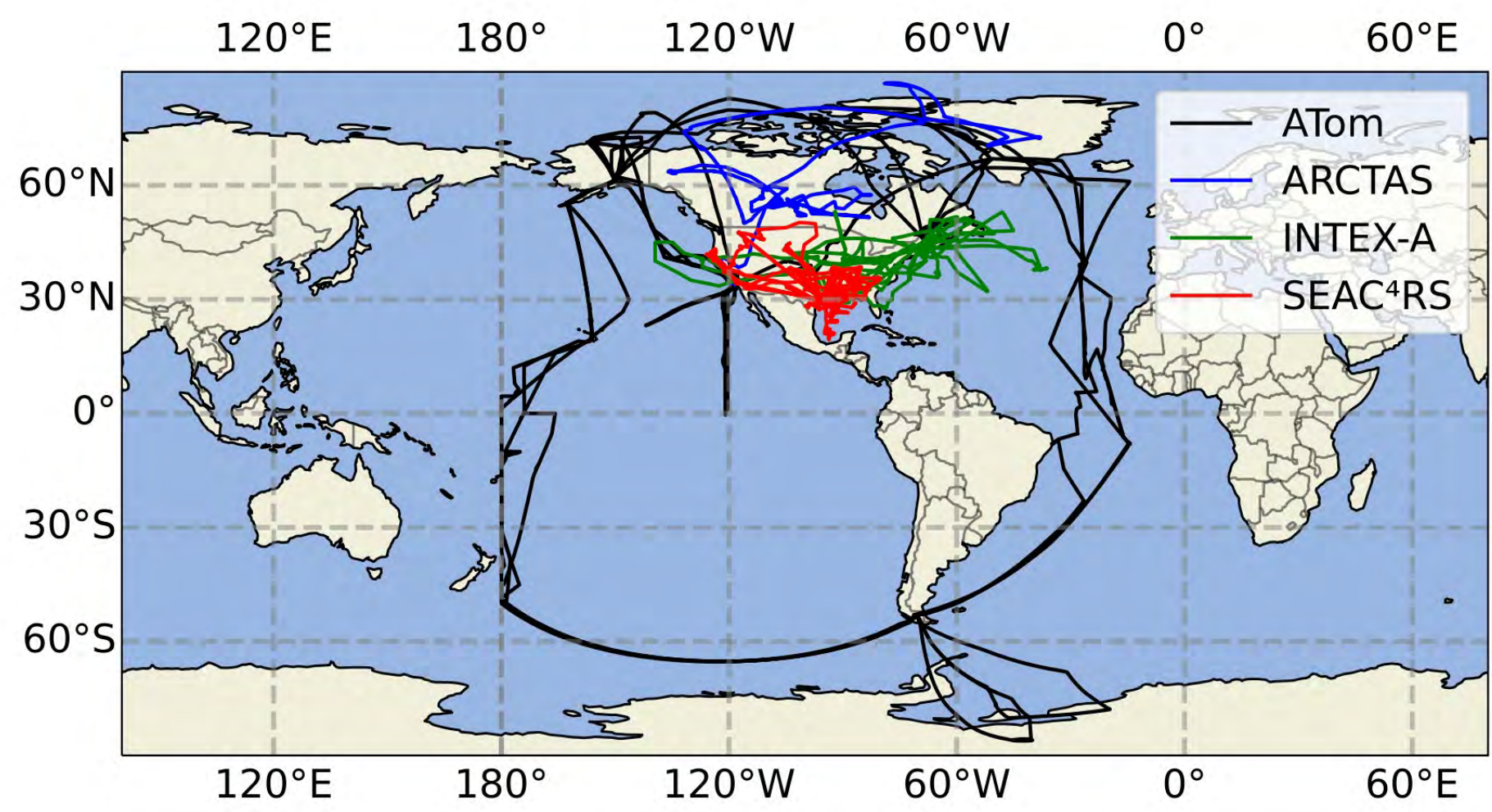
2. The cloud-slicing technique

- The cloud-slicing technique was first used to derive upper tropospheric ozone measurements from the TOMS satellite instrument^[1].
- This takes advantage of the **optically thick clouds** present in the troposphere.
- The NO₂ volume mixing ratio (VMR) is calculated using the relationship between the cloud top pressure and the vertical column density (VCD).

$$\text{NO}_2 \text{ VMR} \propto \frac{\Delta \text{VCD}}{\Delta P}$$
- This technique allows us to eliminate the contribution of the stratosphere.



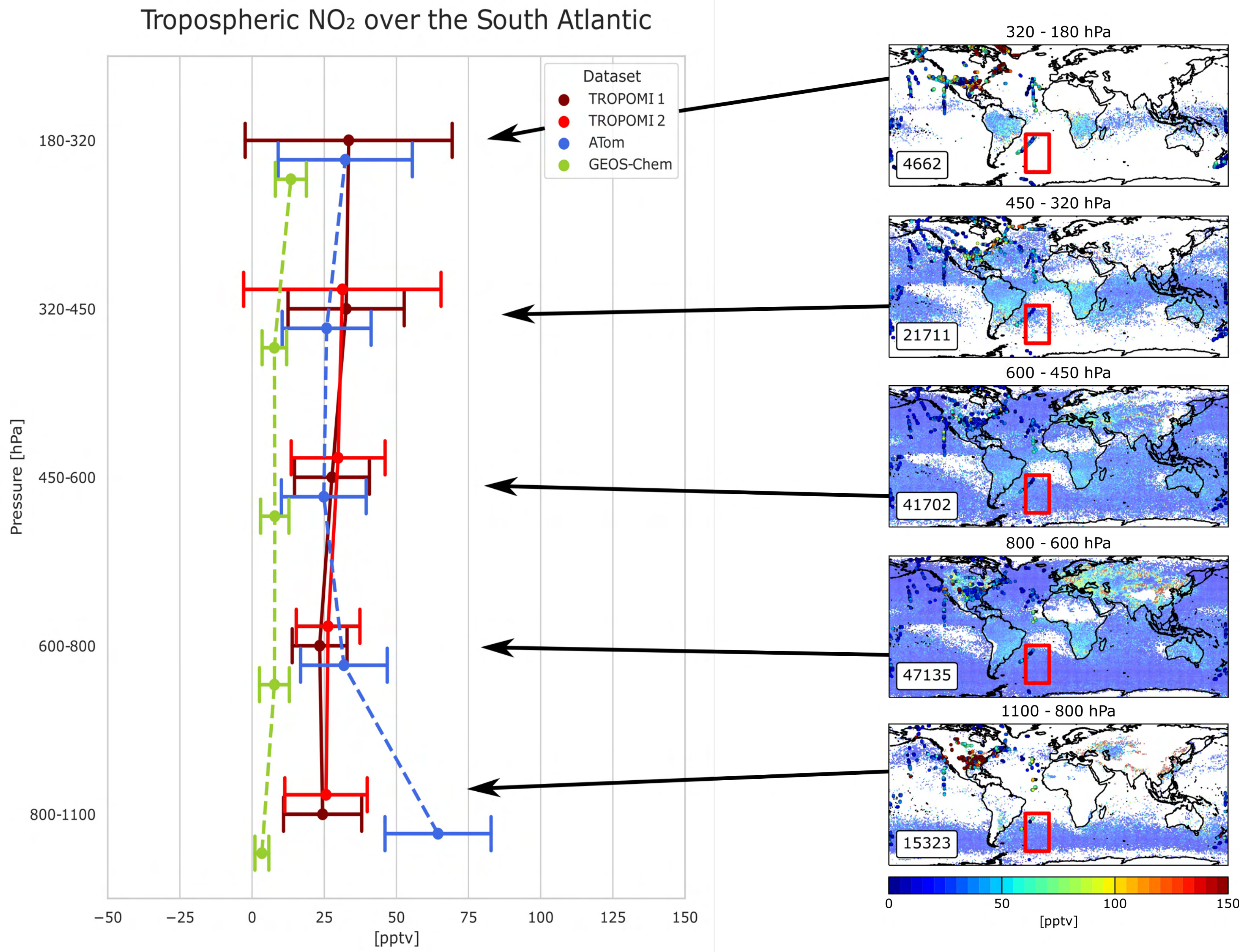
3. NASA DC-8 aircraft campaigns



- ATom-1 (Jul-Aug 2016)
- ATom-2 (Jan-Feb 2017)
- ATom-3 (Sep-Oct 2017)
- ATom-4 (Apr-May 2018)

5. Creating vertical profiles of cloud-sliced NO₂ TROPOMI data

The mean of all the cloud-sliced data points within a given area is calculated and plotted to create a profile and the error bars represent the standard deviation. **GEOS-Chem v13.3.4** is used at a **resolution of 4° x 5°**. GEOS-Chem simulations are shown here for the period **DJF 2015/2016** and cloud-slicing observations averaged over the DJF season between 2018 and 2021.



- TROPOMI 1:** Uses cloud top height and cloud fraction information from the **FRESCO** product that minimises the difference between measured and simulated spectra in the O2 A-band between 752-766 nm.
- TROPOMI 2:** Uses cloud top height and cloud fraction information from the **O22CLD** product that measures oxygen absorption in the O2-O2 band between 460-490 nm.

Next Steps

Explore the cause of inconsistencies in the boundary layer using AMF data and evaluating the correlation between ΔVCD and ΔP. Apply cloud-slicing to the OMPS instrument, as OMPS missions are sustained to 2040.

Acknowledgements

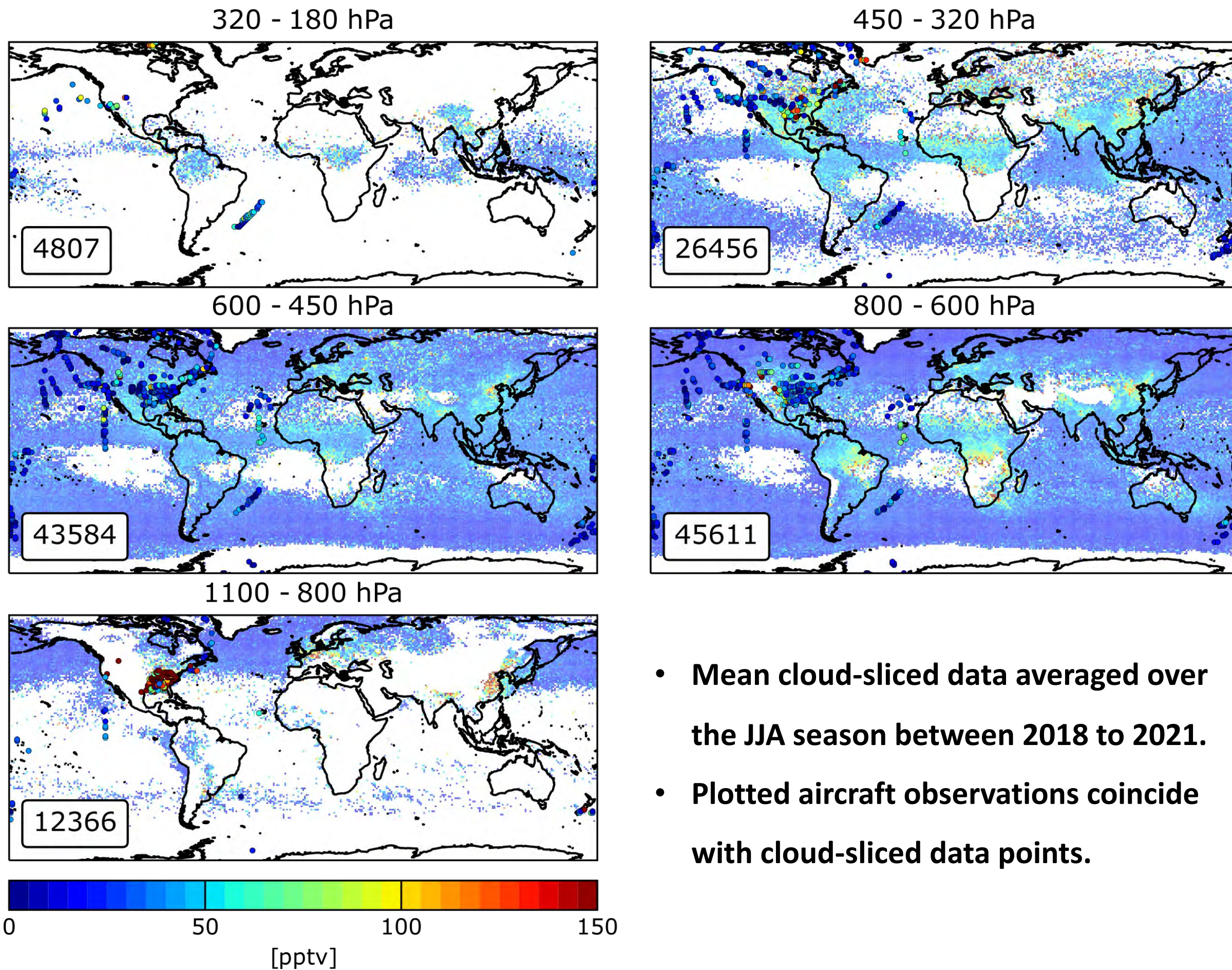
- This work is funded by the European Research Council.
- We are grateful to the NASA DC-8 Science Teams for for access to aircraft observations.

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- [1] Ziemke et al., JGR, <https://doi.org/10.1029/2000jd900768>, 2001
- [2] Choi et al., ACP, <https://doi.org/10.5194/acp-14-10565-2014>, 2014
- [3] Marais et al., ACP, <https://doi.org/10.5194/acp-18-17017-2018>, 2018
- [4] Marais et al., AMT, <https://doi.org/10.5194/amt-14-2389-2021>, 2021

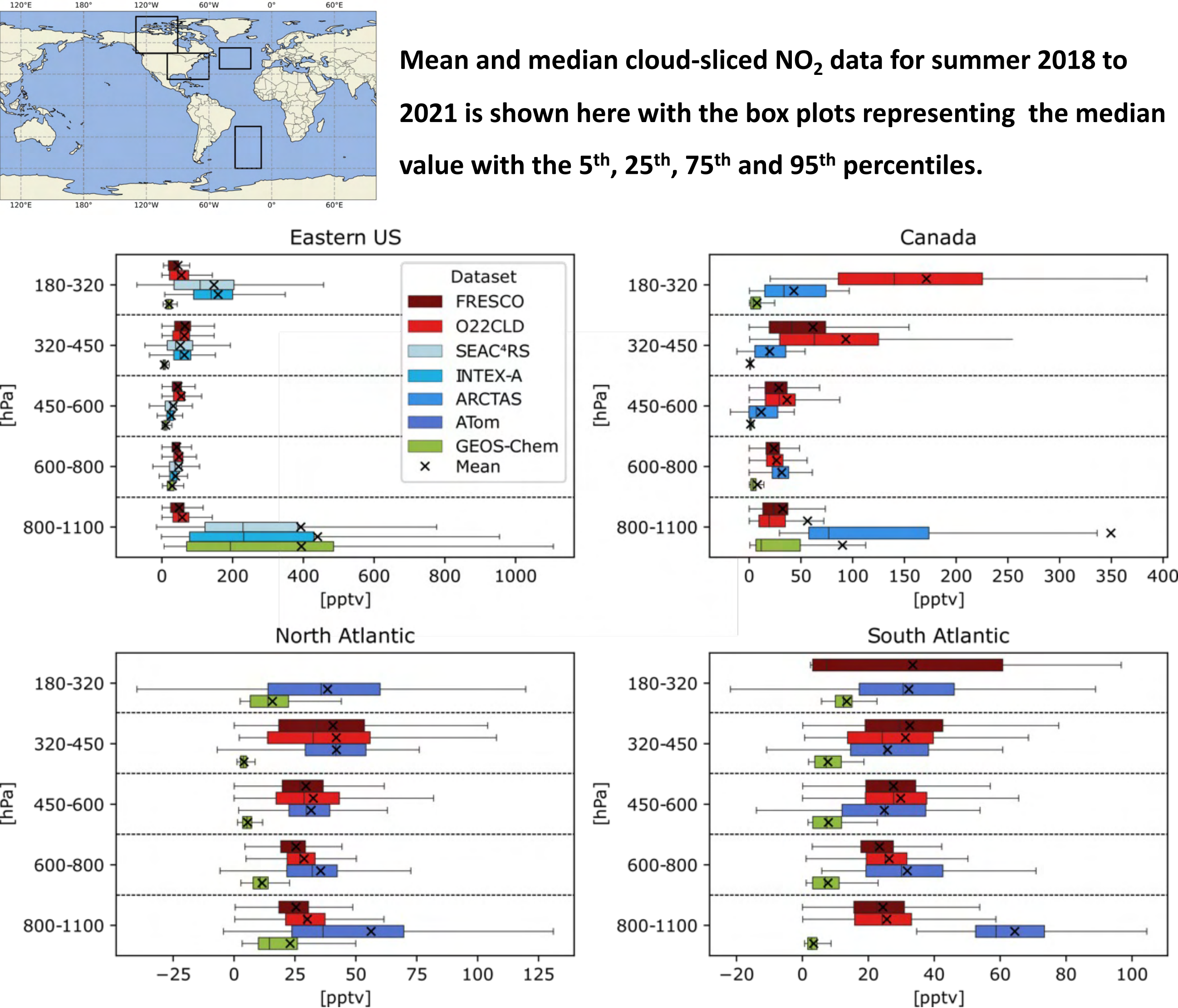
4. Implementing the cloud-slicing technique between 180-1100 hPa

- Previous studies have used cloud-sliced OMI total columns of NO₂ to obtain NO₂ mixing ratios between 650-900 hPa^[2] and between 280-450 hPa^[3] as well as on TROPOMI measurements between 180-450 hPa^[4].
- This has been expanded here using **TROPOMI** data at five pressure intervals between 180-1100 hPa and compared to measurements from the **NASA DC-8** aircraft campaigns.



- Mean cloud-sliced data averaged over the JJA season between 2018 to 2021.
- Plotted aircraft observations coincide with cloud-sliced data points.

6. Comparing vertical profiles between different global regions



- Concentrations of NO₂ deviate by **less than 15%** between cloud-slicing and aircraft observations in the mid-troposphere where data density is increased.
- Cloud-slicing results underestimate NO₂ concentrations compared to aircraft observations in the boundary layer where cloud-slicing is up to **300 ppbv** below aircraft observations.
- Differences of up to 150 ppbv in the upper troposphere (320–180 hPa) may be due to the low sampling frequency here (see Box 3).