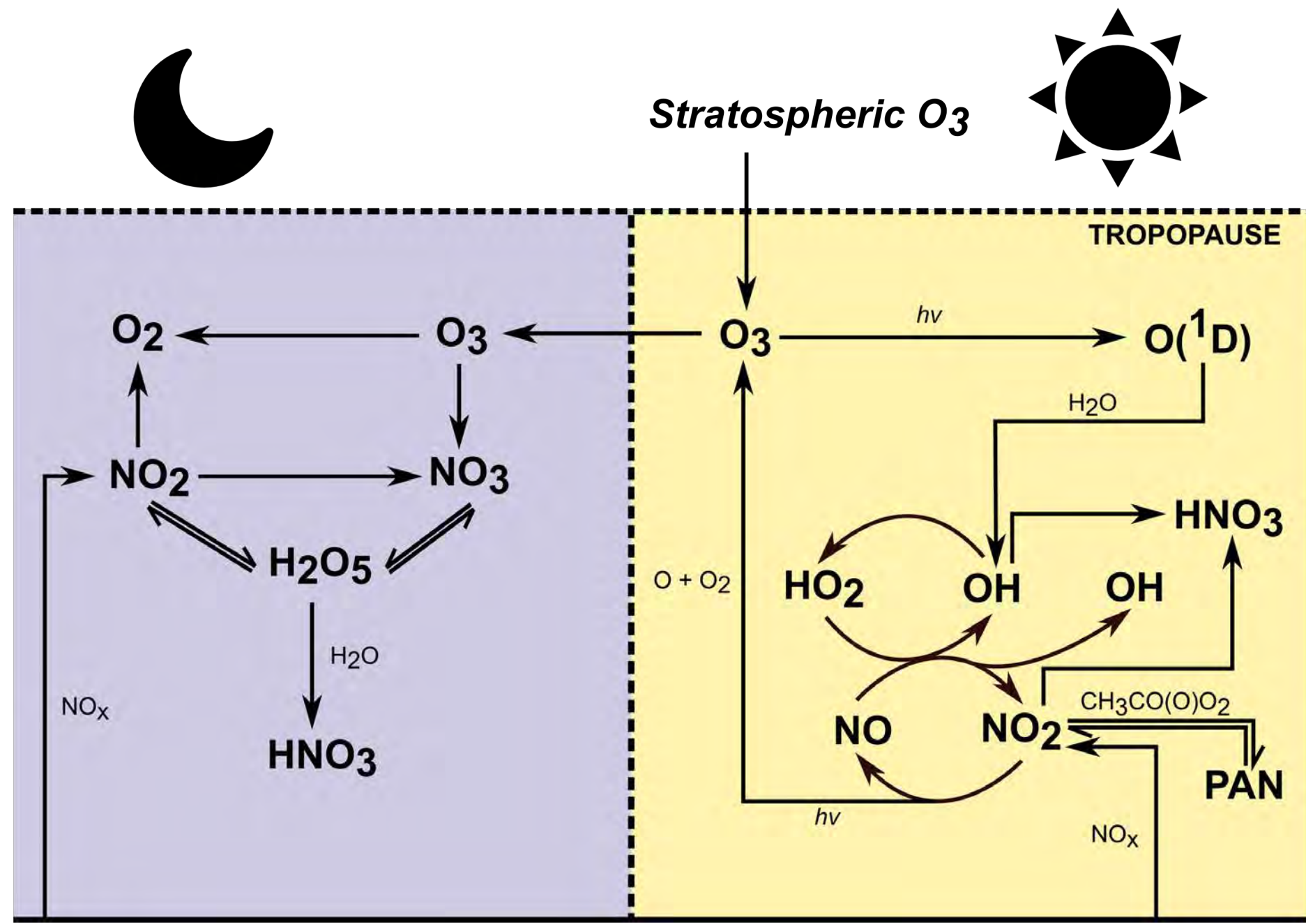


Major Finding: Cloud-slicing of TROPOMI NO₂ performs well between 800-320 hPa when compared to NASA DC-8 observations. GEOS-Chem underestimates NO₂ by up to 50% in the remote oceans. Cloud-slicing of TROPOMI O₃ performs well across the whole troposphere when compared to in-situ observations.

1. Motivation

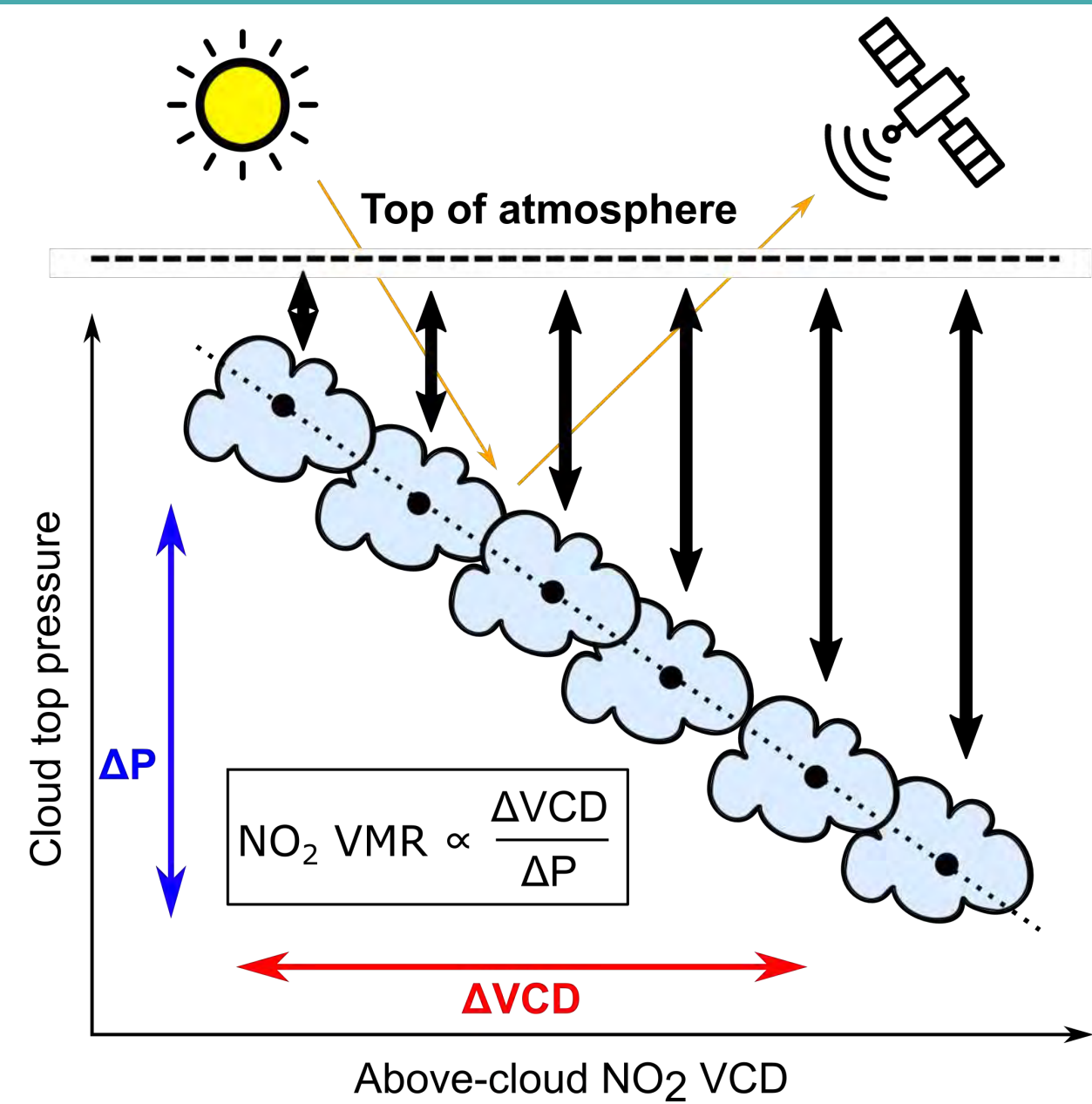
Cycle of NO_x in the troposphere during the day and at night



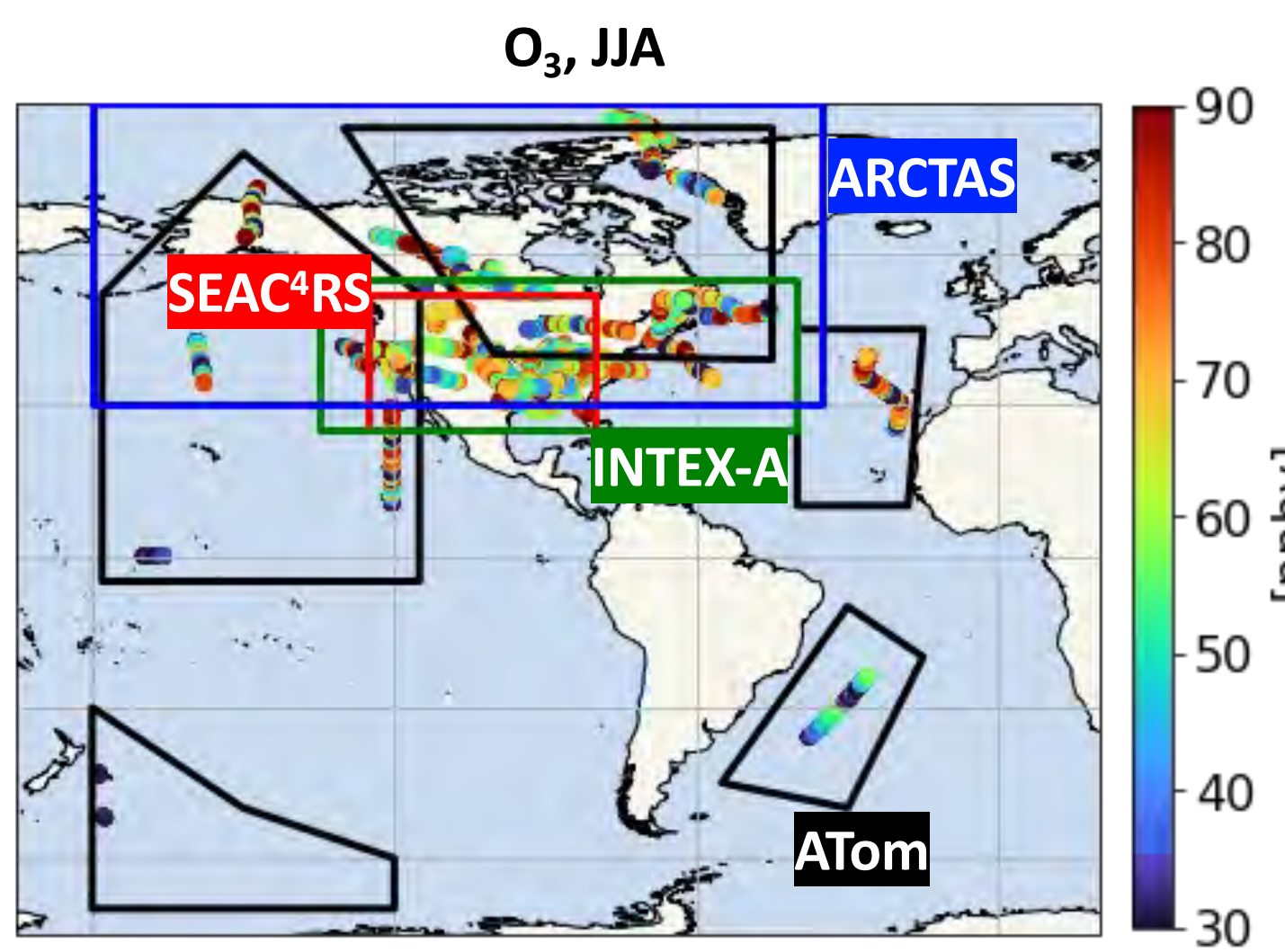
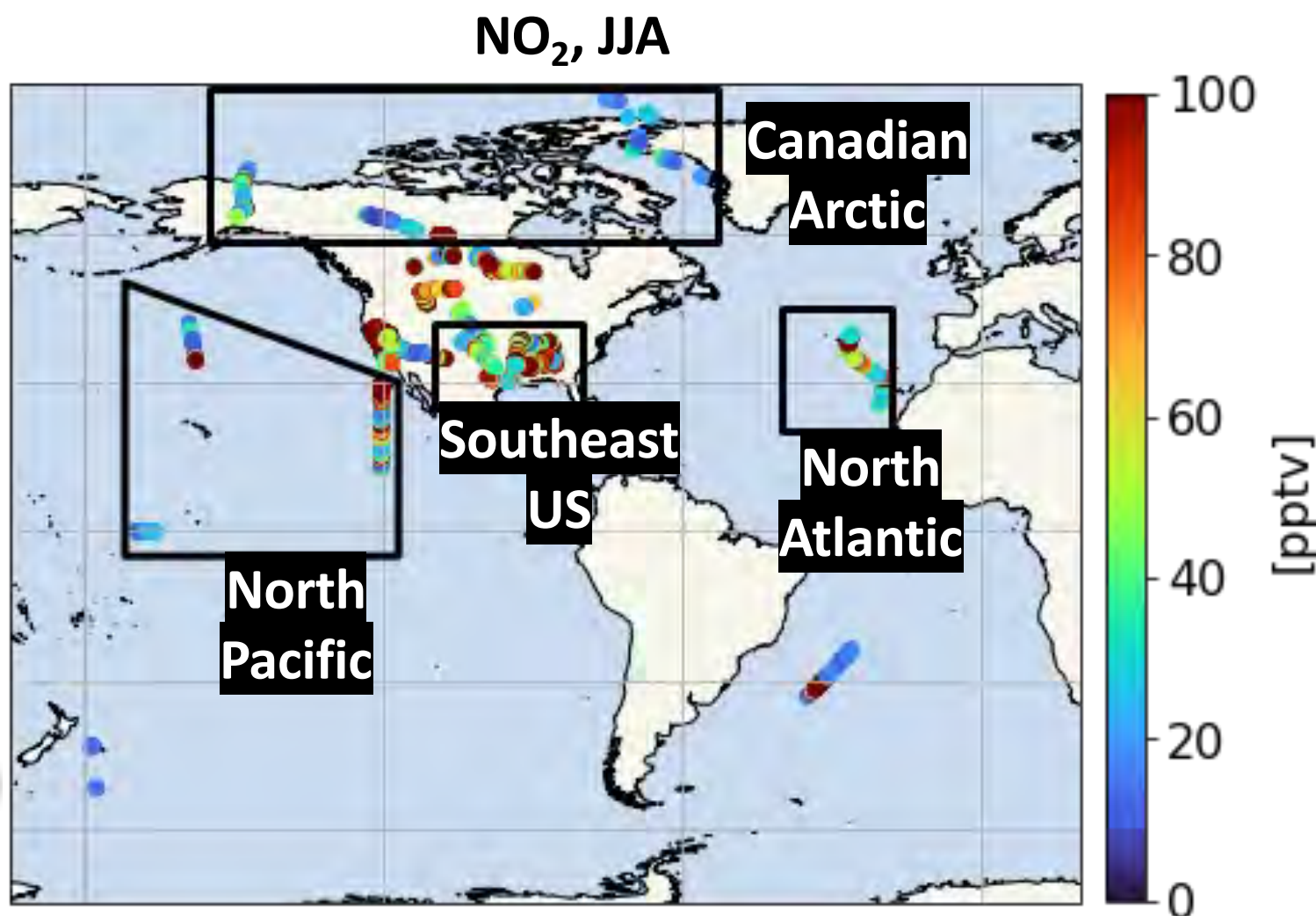
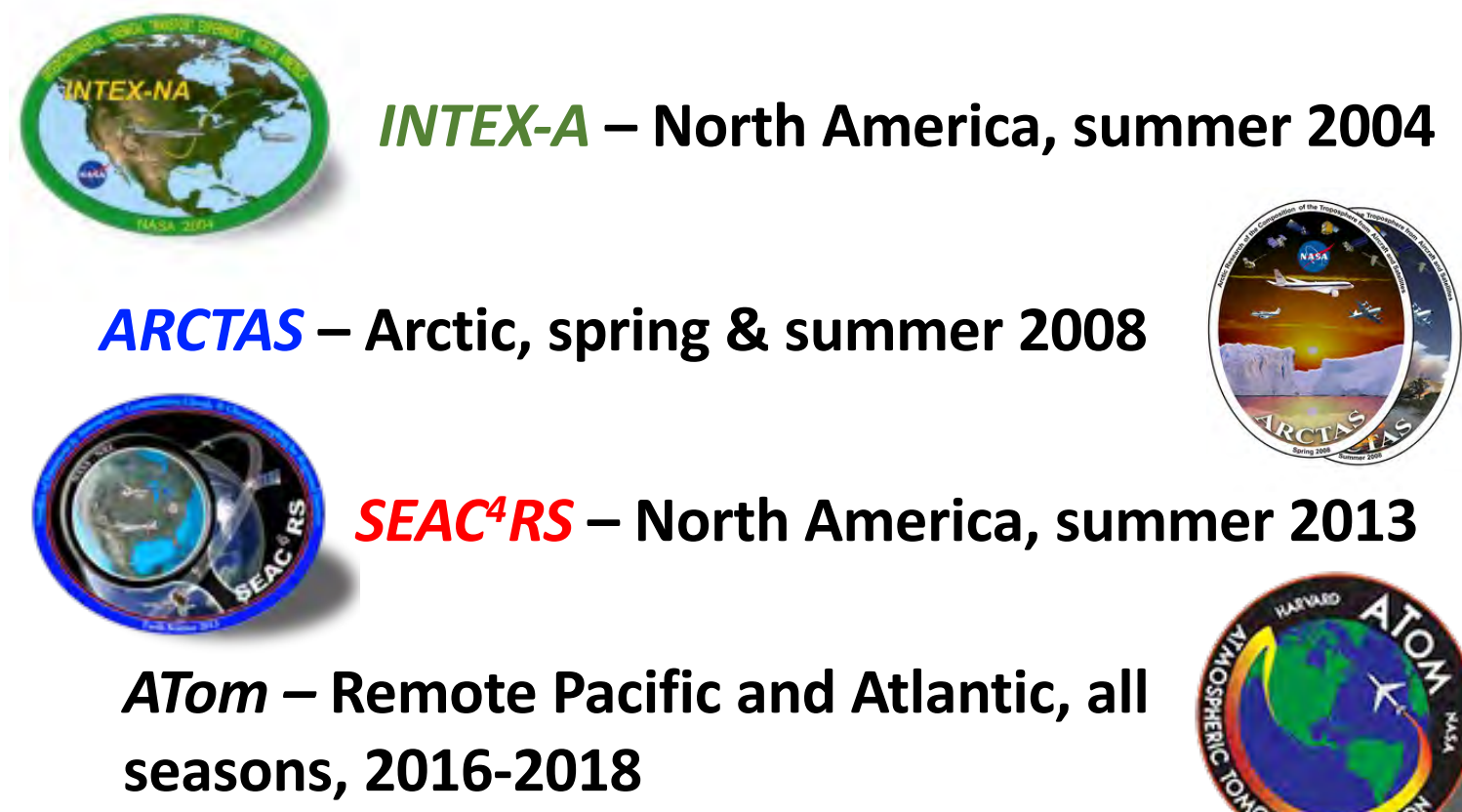
- Nitrogen oxides (NO_x ≡ NO + NO₂) are strongly linked to the formation of ozone (O₃) in the NO_x-limited regions of the troposphere.
- O₃ is a key contributor to the oxidation state of the atmosphere and maintaining the oxidation capacity of the troposphere.
- Well-mixed greenhouse gases have been 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).
- This technique allows us to eliminate the contribution of the stratosphere.

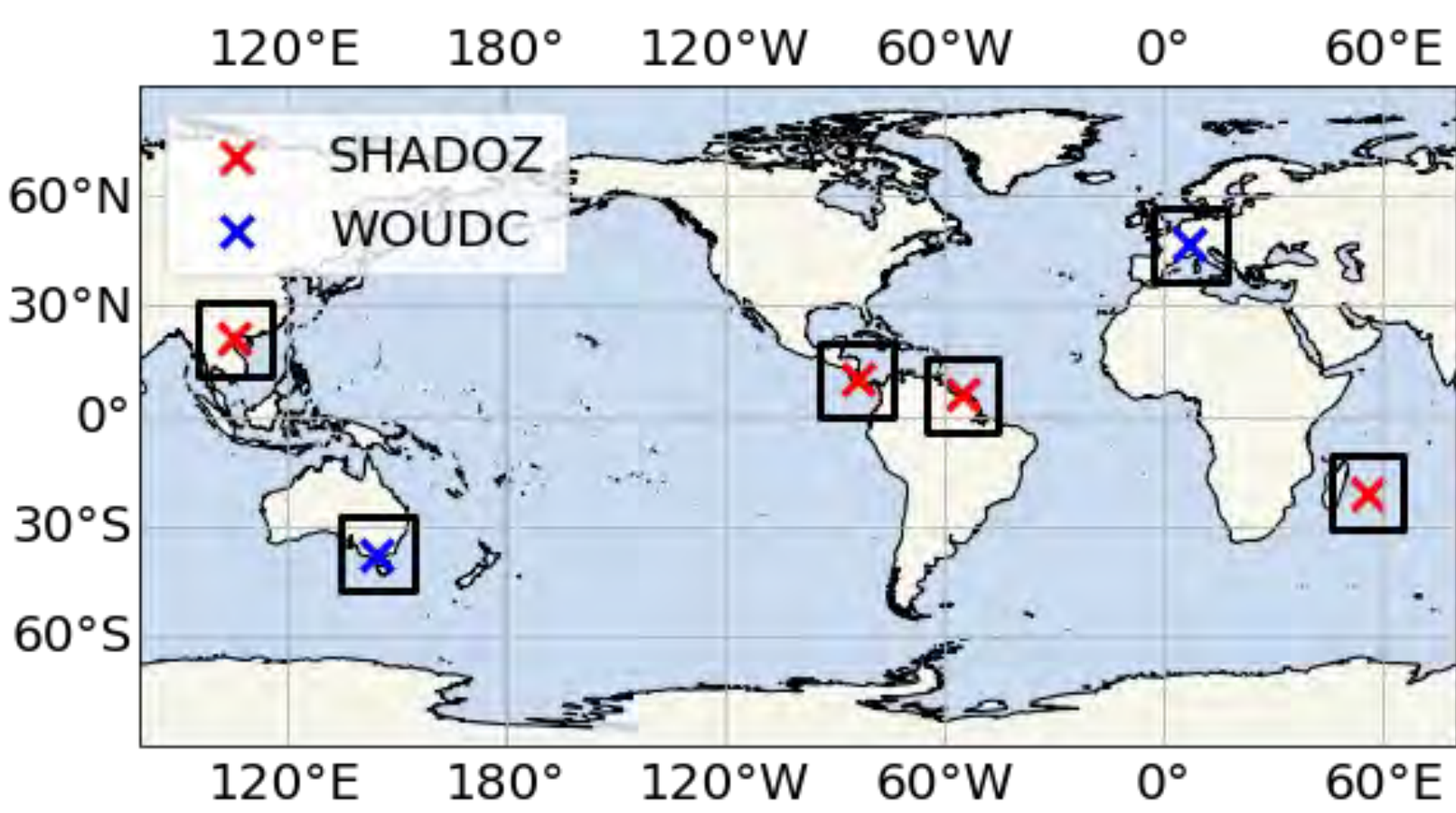


3. NASA DC-8 aircraft campaigns



- Aircraft observations (shown here for JJA) are compared to cloud-slicing in each box.
- For NO₂, we compare in the Canadian Arctic, Southeast US, North Pacific and North Atlantic calculated under photochemical steady state (PSS).
- For O₃, we compare in regions that cover each NASA DC-8 aircraft campaign.

4. Ozone sonde measurements



- We use WUOZ & SHADOZ ozone sonde measurements at Hanoi, Costa Rica, Paramaribo and Réunion Island, Melbourne and Switzerland
- We compare these measurements to cloud-slicing results in the **10° x 10° area** surrounding each site

Next steps

Use these cloud-slicing results to assess the effectiveness of GEOS-Chem of representing lightning NO_x in the troposphere

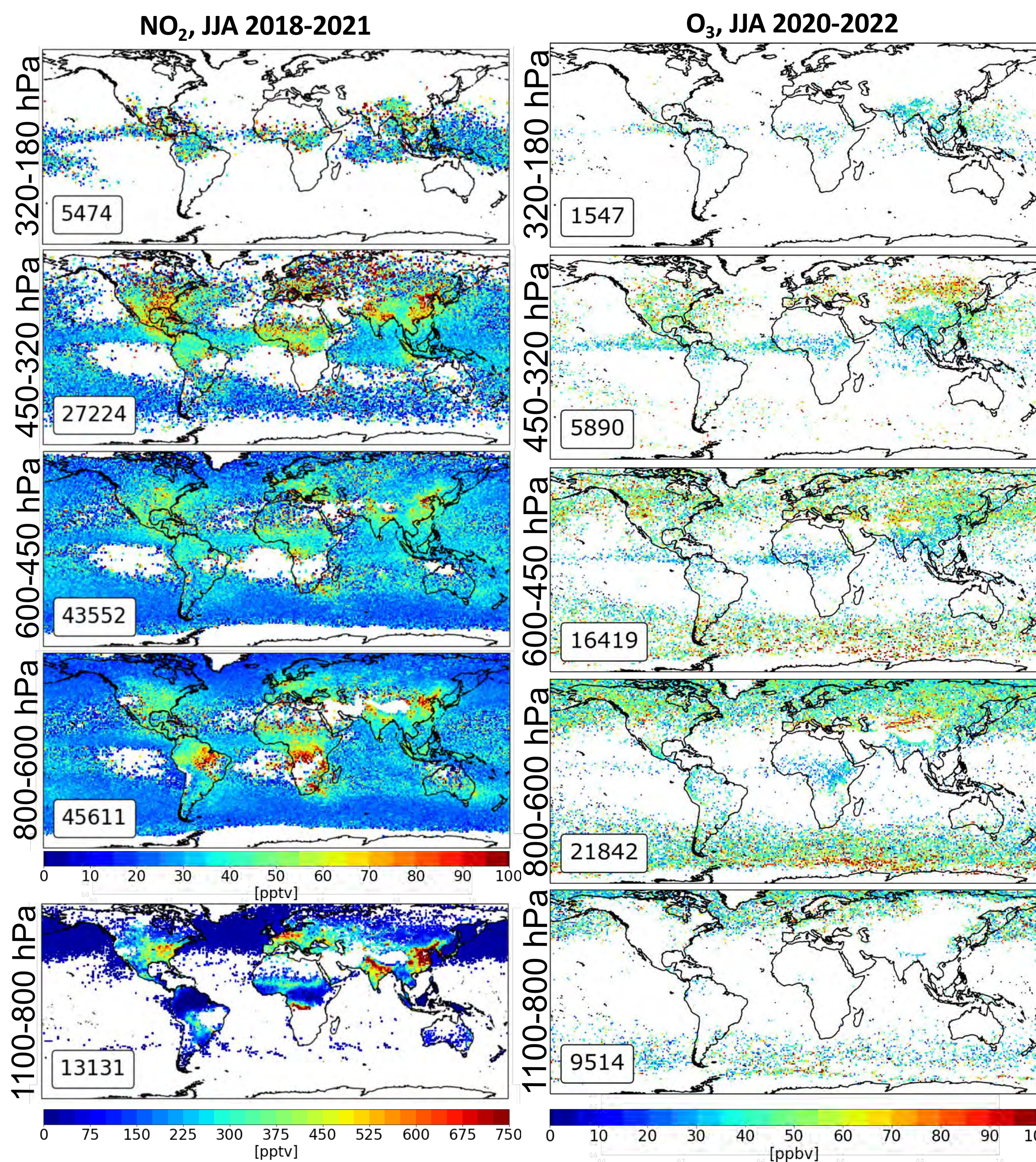
References

- [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

Acknowledgements

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- We are grateful to the NASA DC-8 Science Teams for for access to aircraft observations.

5. Cloud-slicing results between 1100-180 hPa

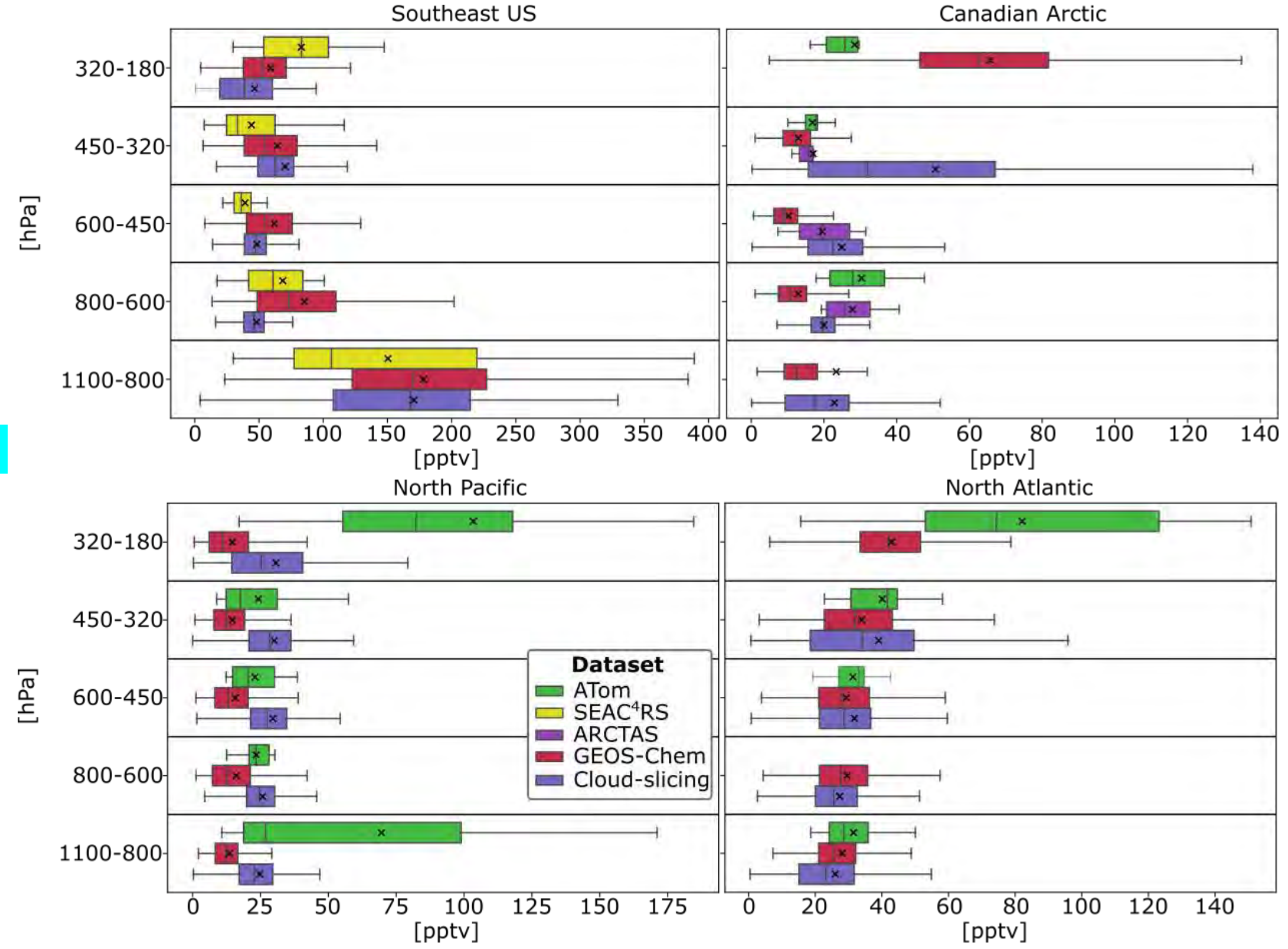


- Previous studies have carried out cloud-slicing on TOMS measurements of tropospheric O₃ in two pressure bands between 1000-100 hPa^[1]. Cloud-slicing has also been carried out on OMI measurements of tropospheric NO₂ between 900-650 hPa^[2] and between 450-280 hPa^[3] as well as on TROPOMI measurements between 450-180 hPa^[4].
- This has been expanded here using **TROPOMI** data at five pressure intervals between 1100-180 hPa for both NO₂ and O₃.
- Between 1100-800 hPa, NO₂ concentrations are from the difference between the tropospheric column and cloud-slicing concentrations between 800-180 hPa.

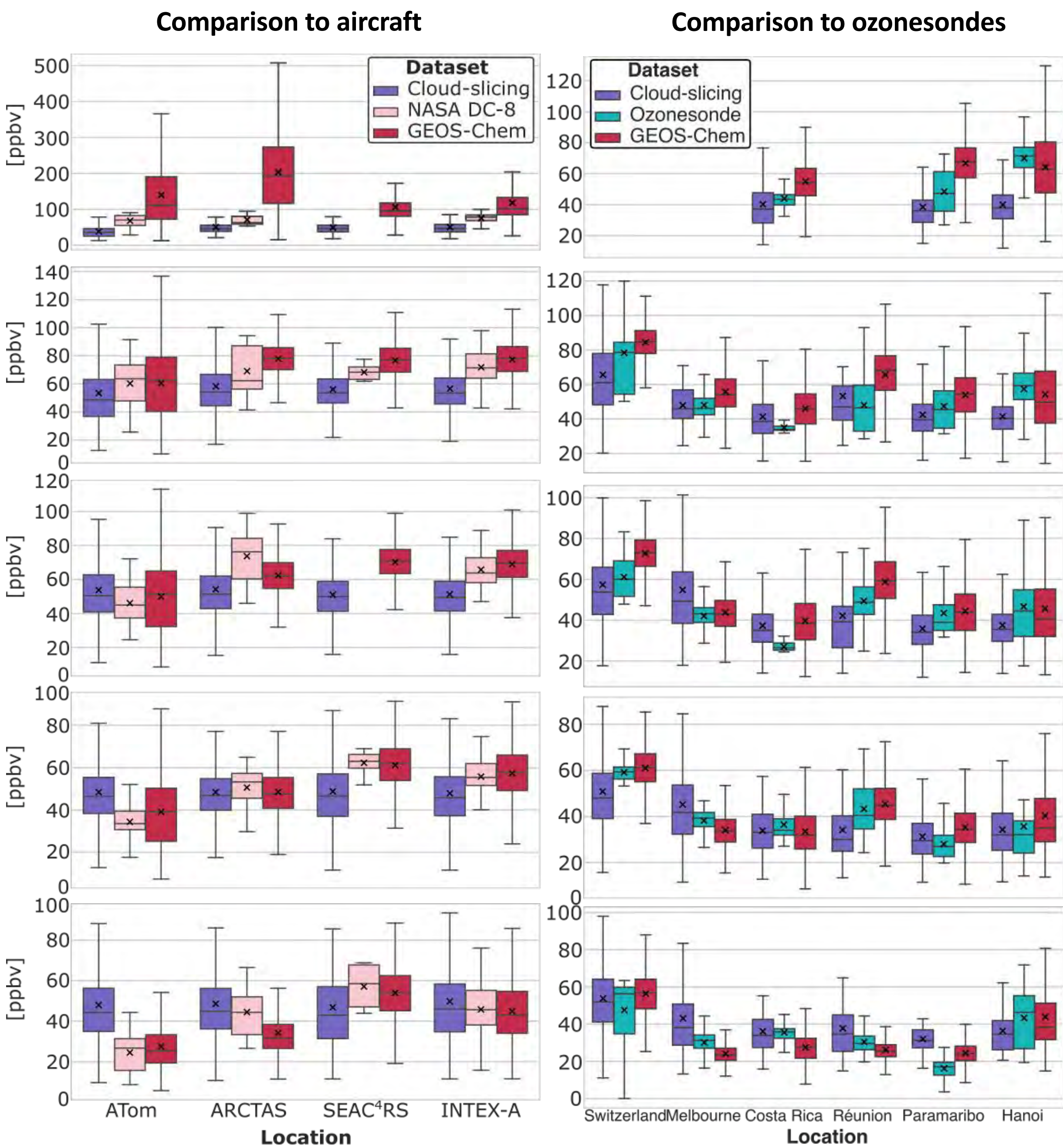
6. Comparing NO2 vertical profiles to in-situ measurements and GEOS-Chem

Mean and median cloud-sliced NO₂ data is shown here with the box plots representing the median value with the 5th, 25th, 75th and 95th percentiles.

- The median of all the cloud-sliced data points within a given area is calculated and plotted to create a profile. **GEOS Chem v13.3.4** is used at a **resolution of 2° x 2.5°** between 2016-2019 with PPN and nitrate photolysis added to simulations.
- Concentrations of NO₂ deviate by **less than 15%** between cloud-slicing and aircraft observations in the mid-troposphere where data density is increased.
- 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).



7. Comparing O3 vertical profiles to in-situ measurements and GEOS-Chem



- Concentrations of O₃ from cloud-slicing deviate from ozonesonde observations by **< 40 ppbv across the whole troposphere.**
- When compared to ozonesonde measurements, GEOS-Chem overestimates concentrations at 320-180 hPa by as much as **150 ppbv.**
- Across the whole troposphere, concentrations of O₃ from cloud-slicing deviate from aircraft observations by **< 20 ppbv** except for in the upper troposphere where differences can reach **40 ppbv** due to lower cloud-slicing data density (Box 3).