Vertical profiles of global tropospheric nitrogen dioxide (NO_2) and ozone (O_3) obtained via cloud-slicing TROPOMI partial columns

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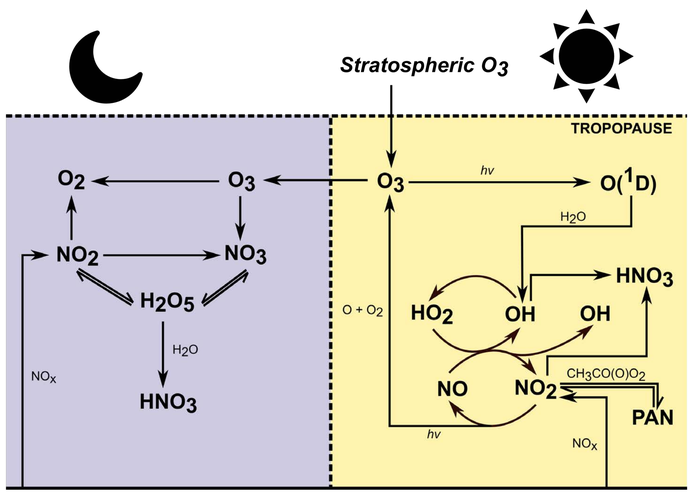
Major Finding: Cloud-slicing of TROPOMI NO_2 performs well between 800-320 hPa when compared to NASA DC-8 observations. GEOS-Chem underestimates NO_2 by up to 50% in the remote oceans. Cloud-slicing of TROPOMI O_3 performs well across the whole troposphere when compared to in-situ observations.

1. Motivation

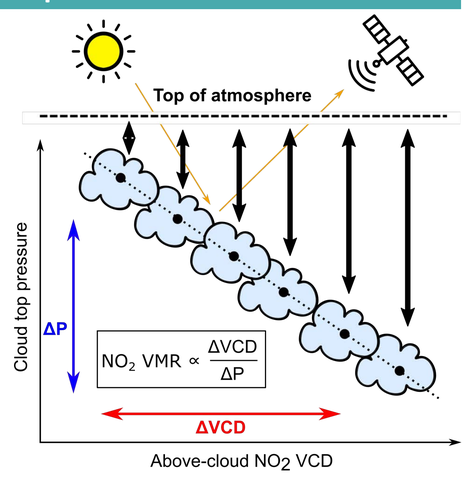
Cycle of NO_x in the troposphere during the day and at night



- Nitrogen oxides ($NO_x \equiv NO + NO_2$) are strongly linked to the formation of ozone (O_3) in the NO_x-limited regions of the troposphere.
- O_3 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 O_3 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 NO_2 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

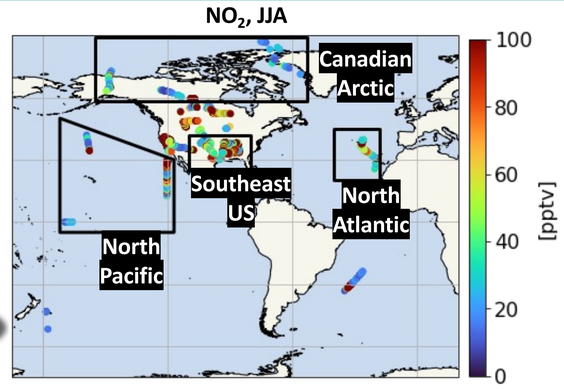
INTEX-A – North America, summer 2004

ARCTAS – Arctic, spring & summer 2008

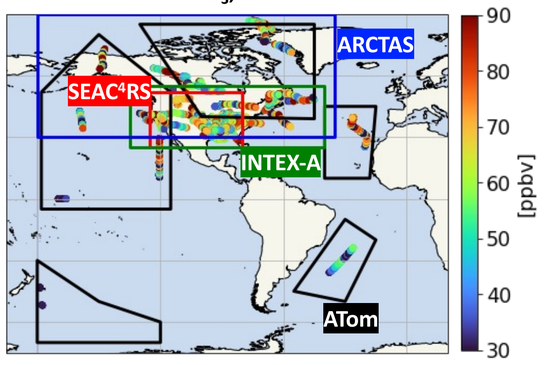
SEAC⁴RS – North America, summer 2013

ATom – Remote Pacific and Atlantic, all seasons, 2016-2018

NO_2, JJA

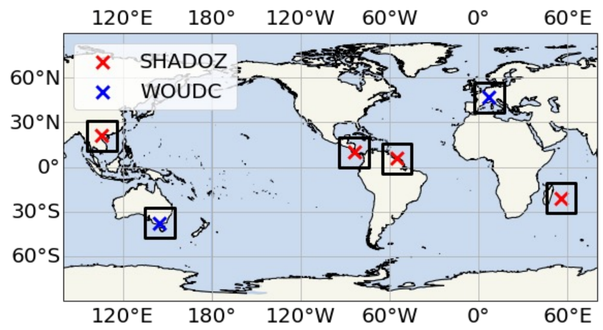


O_3, JJA



- Aircraft observations (shown here for JJA) are compared to cloud-slicing in each box.
- For NO_2, we compare in the Canadian Arctic, Southeast US, North Pacific and North Atlantic calculated under photochemical steady state (PSS).
- For O_3, we compare in regions that cover each NASA DC-8 aircraft campaign.

4. Ozonesonde measurements



- We use WOUDC & SHADOZ ozonesonde 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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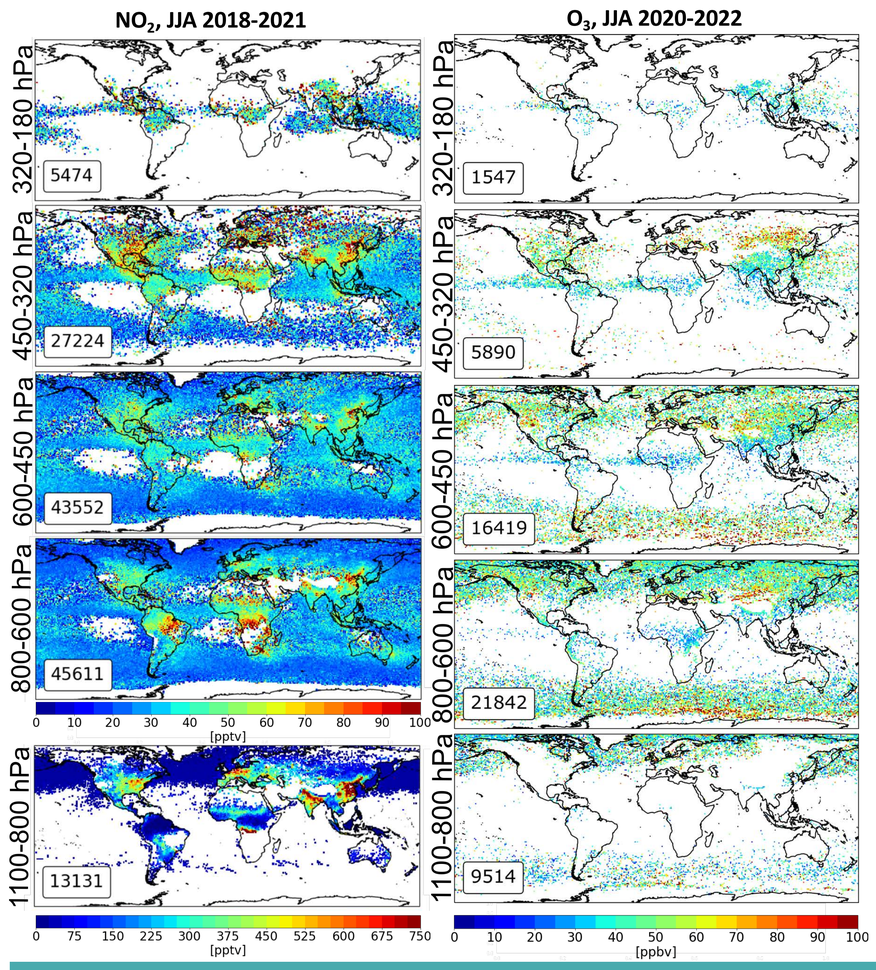
- We are grateful to the NASA DC-8 Science Teams for for access to aircraft observations.

5. Cloud-slicing results between 1100-180 hPa

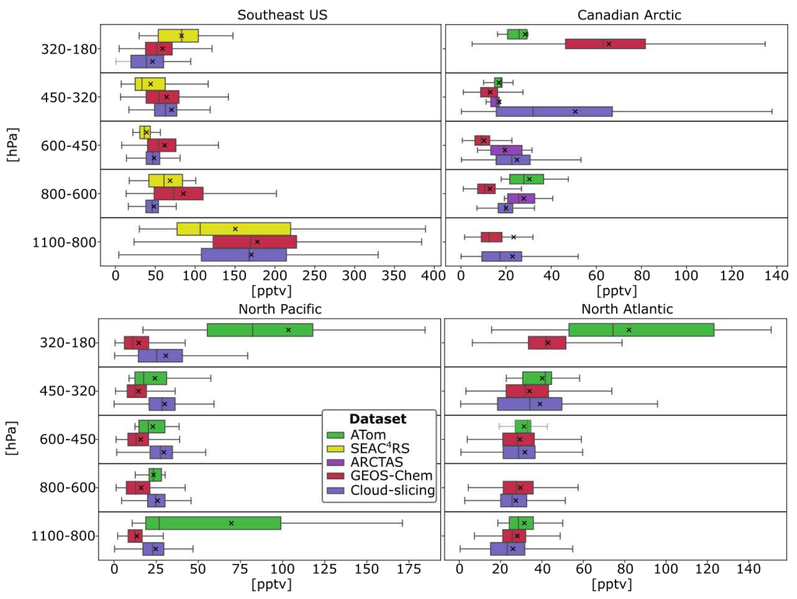


- Previous studies have carried out cloud-slicing on TOMS measurements of tropospheric O_3 in two pressure bands between 1000-100 hPa[1]. Cloud-slicing has also been carried out on OMI measurements of tropospheric NO_2 between 900-650 hPa[2] and between 450-280 hPa[3] as well as on TROPOMI measurements between 450-180 hPa[4].
- This has been expanded here using TROPOMI data at five pressure intervals between 1100-180 hPa for both NO_2 and O_3.
- Between 1100-800 hPa, NO_2 concentrations are from the difference between the tropospheric column and cloud-slicing concentrations between 800-180 hPa.

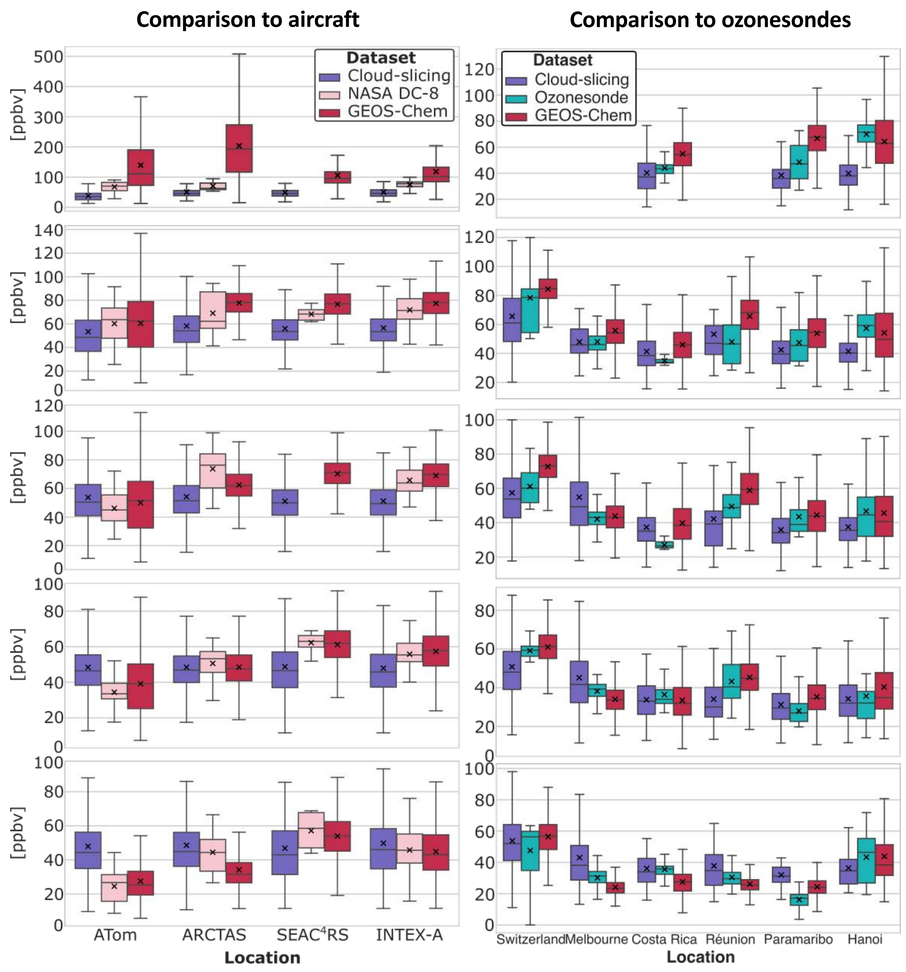
6. Comparing NO2 vertical profiles to in-situ measurements and GEOS-Chem

Mean and median cloud-sliced NO_2 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° × 2.5° between 2016-2019 with PPN and nitrate photolysis added to simulations.
- Concentrations of NO_2 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 pptv 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 O_3 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 O_3 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).