

Bridging knowledge gaps in atmospheric science:

From tropical cities to the remote troposphere and the mesosphere 50-80 km aloft



Fast-growing tropical megacities

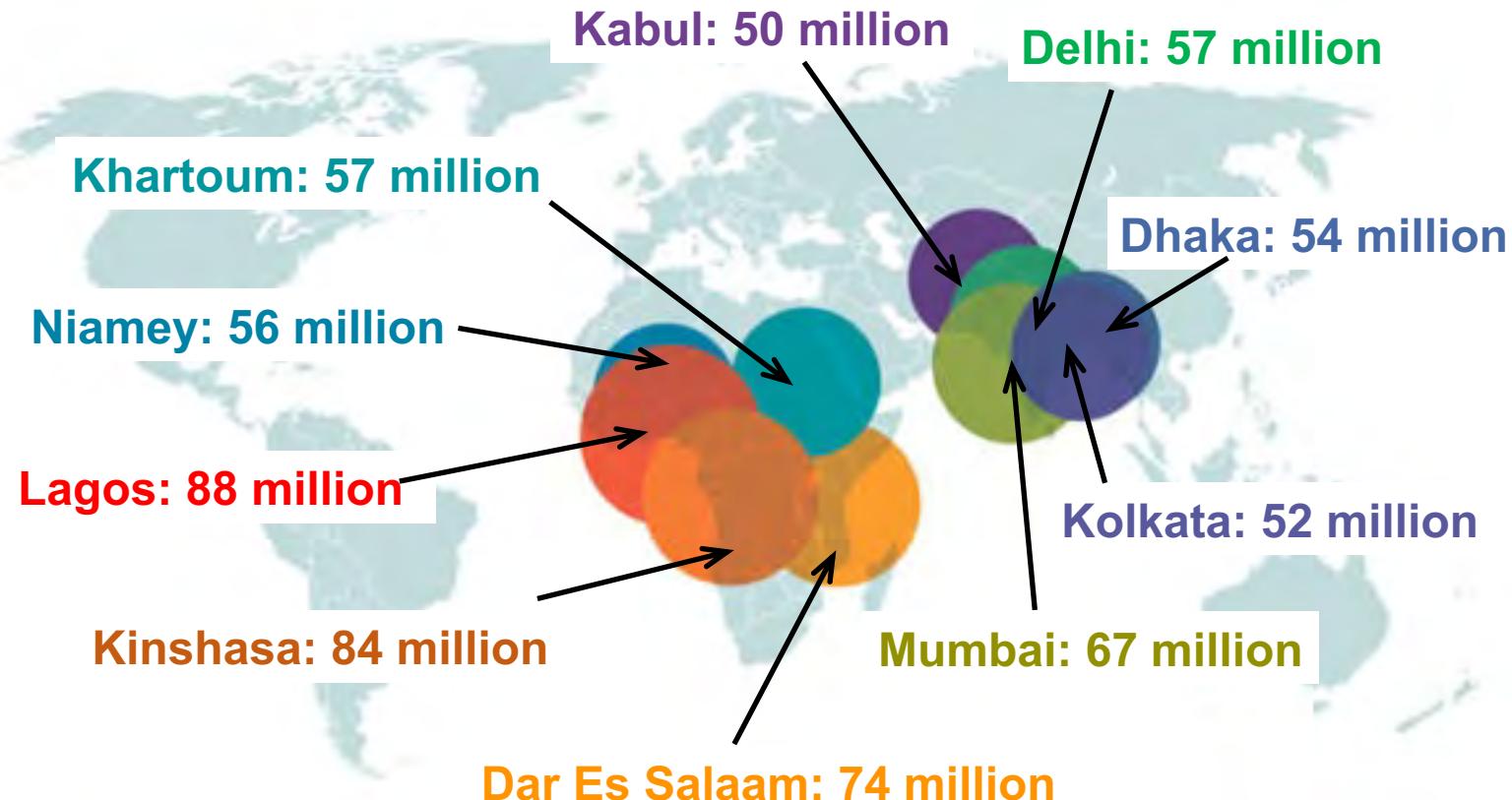


Karn Vohra
postdoc

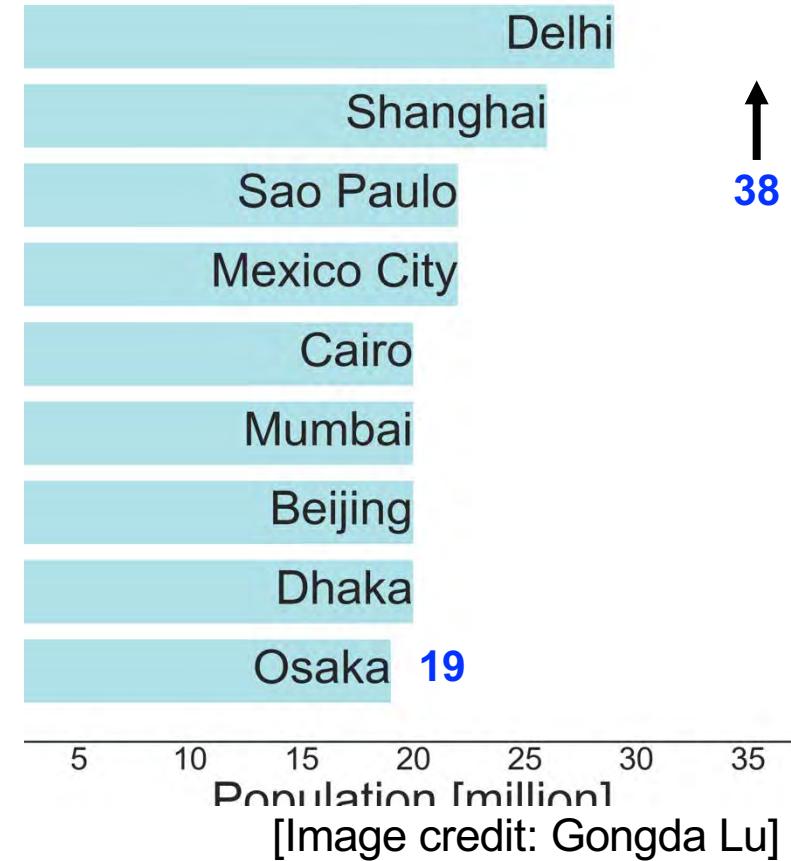
The largest future megacities are all in the tropics

Mostly in tropical Africa and Asia, where air quality knowledge gaps are largest

WORLD'S LARGEST CITIES IN 2100



Largest cities in 2020

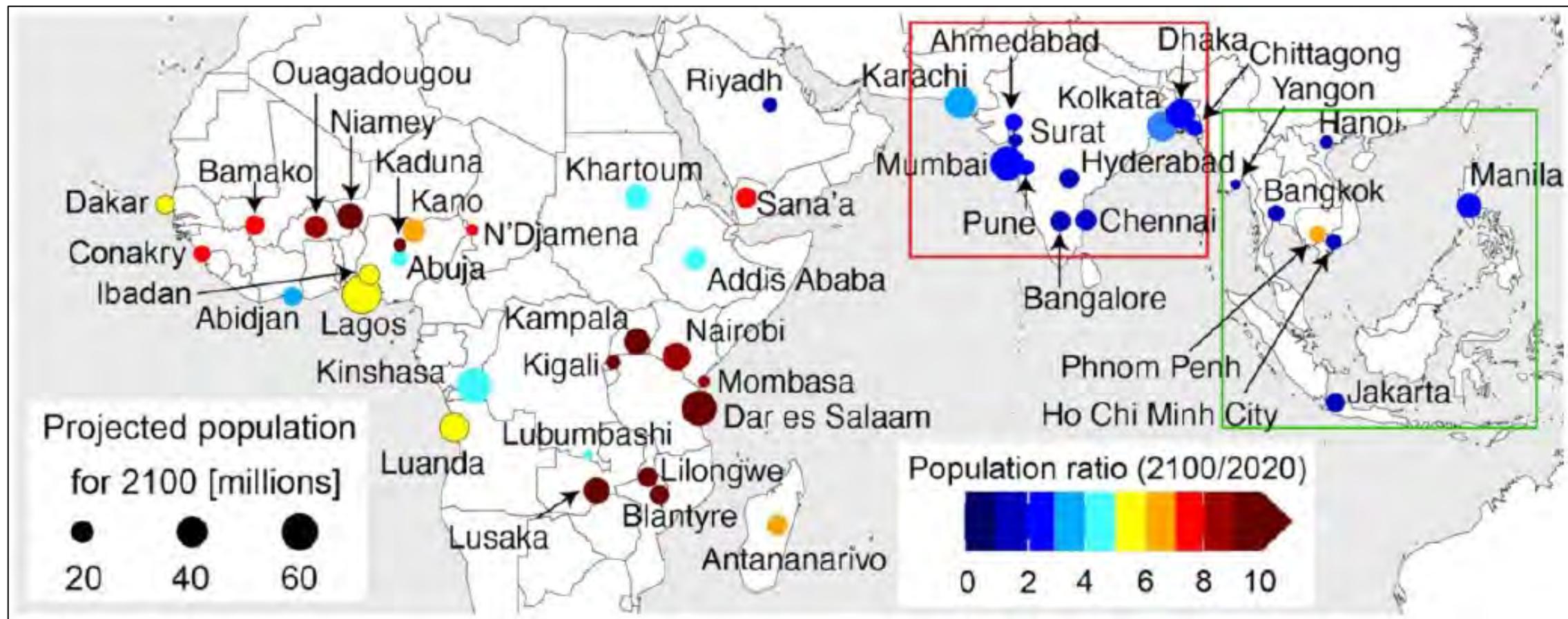


Adapted image: <https://medium.com/ensia/here-come-the-megacities-1b0f8a2287f2>

Projections: <https://journals.sagepub.com/doi/full/10.1177/0956247816663557>

Fastest-growing cities are in the tropics

Population growth in the 46 fastest-growing cities in tropical Africa, Asia and the Middle East



Regional annual projected population growth rates for 2020-2100 [Hoornweg & Pope, 2017]:
3-31% for Africa, 0.8-3% for **South Asia**, 0.5-7% for **Southeast Asia**

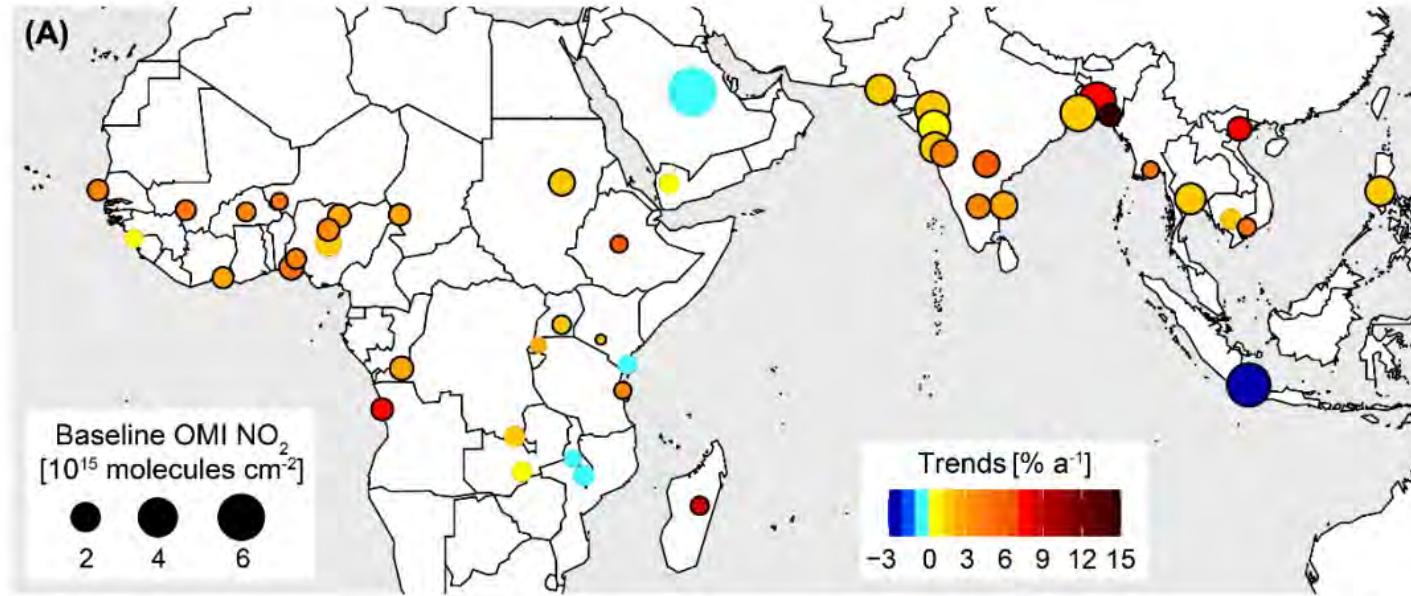
Steep annual increases in NO_x and NH_3

NO_2 trends
(proxy for NO_x)
[2005-2018]

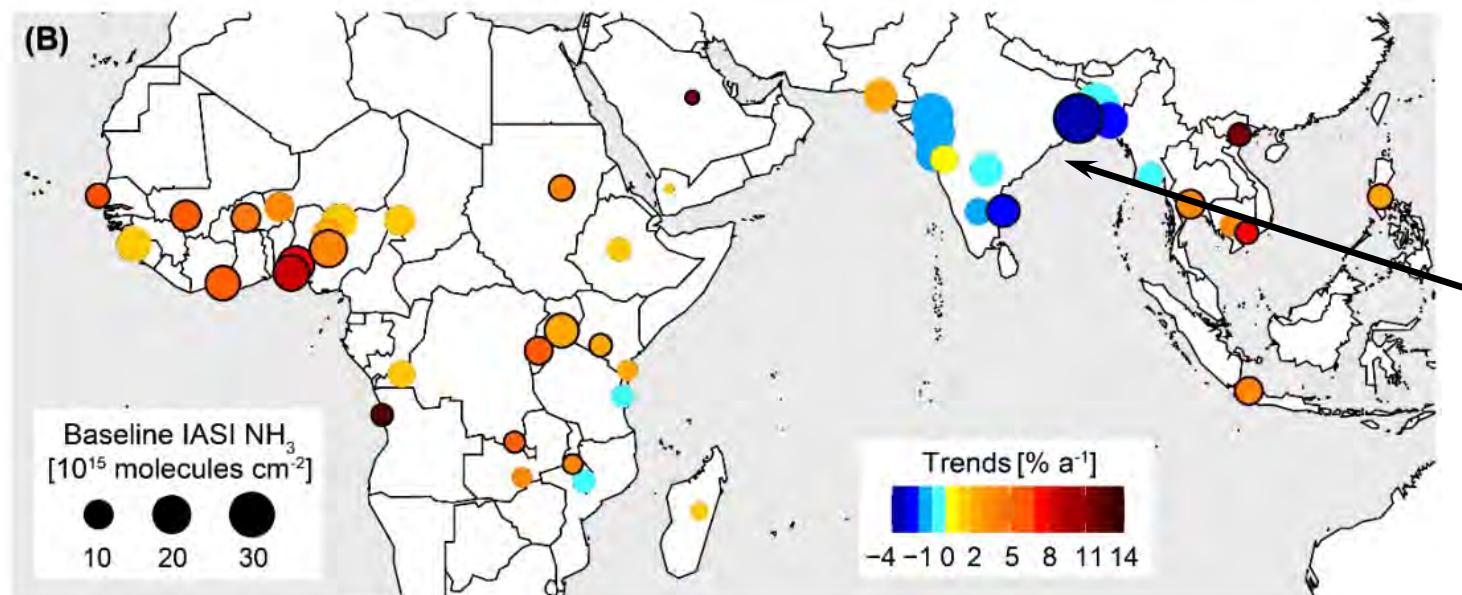
OMI: Ozone Monitoring Instrument

NH_3 trends
(depends on acidic aerosol abundance)
[2008-2018]

IASI: Infrared atmospheric sounding interferometer



Circle Features:
Size: start of record
Color: trend
Outline: significant



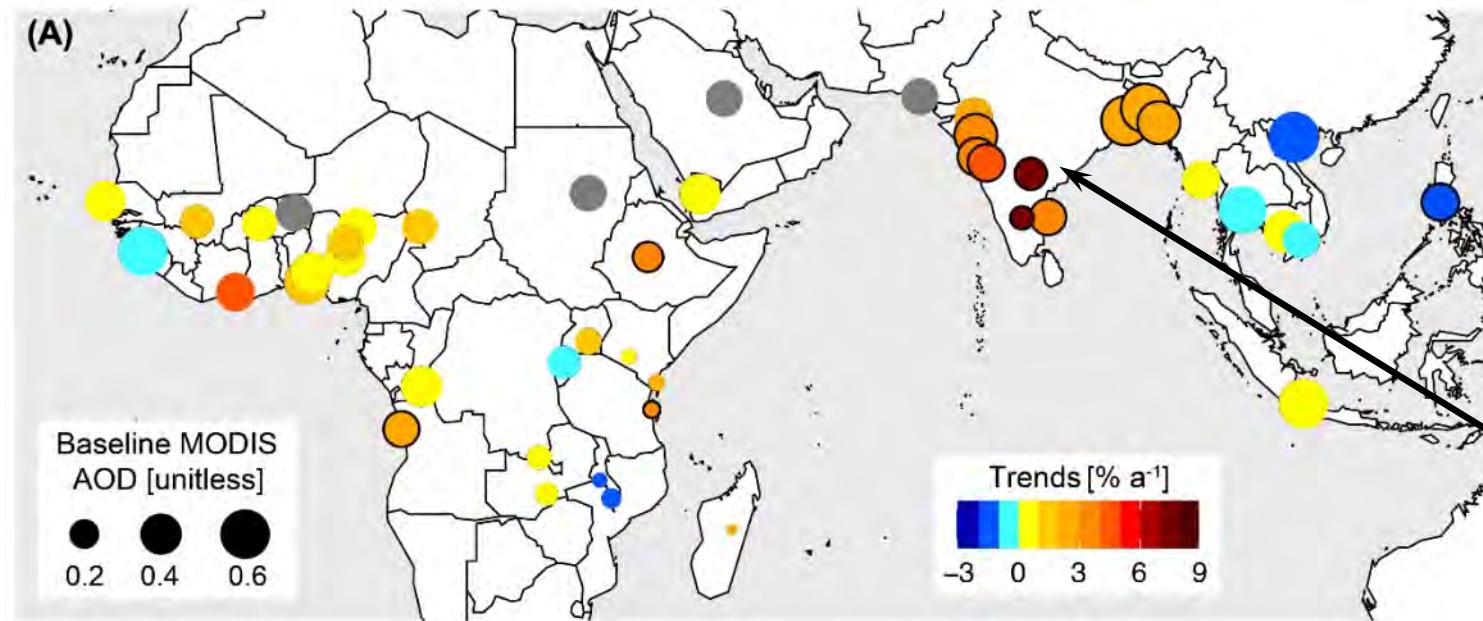
Decline over Indian subcontinent due to increase in uptake to acidic aerosols

Annual changes in PM_{2.5} and ozone production regimes

AOD trends
(proxy for PM_{2.5})
[2005-2018]

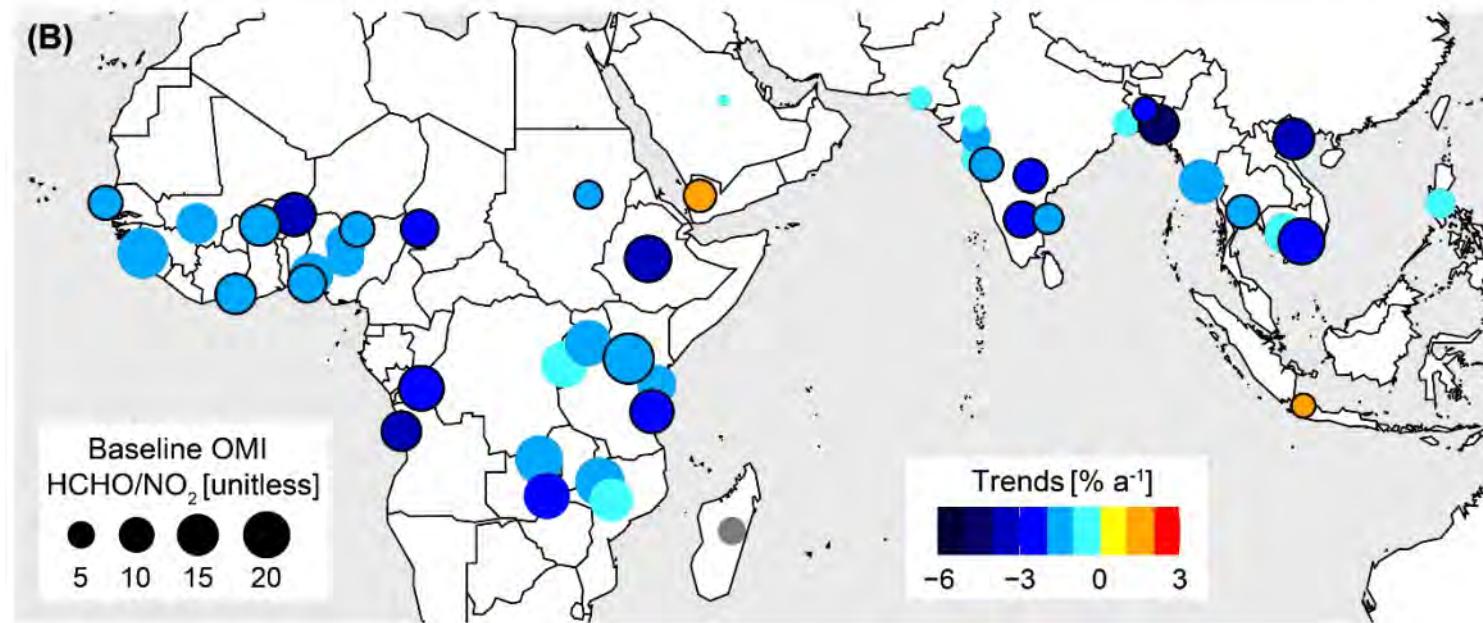
MODIS: Moderate resolution imaging spectroradiometer

HCHO/NO₂ trends
(proxy for ozone production regime)
[2005-2018]



Circle Features:
Size: start of record
Color: trend
Outline: significant

Increases in PM_{2.5} precursors SO₂, NH₃, NO_x

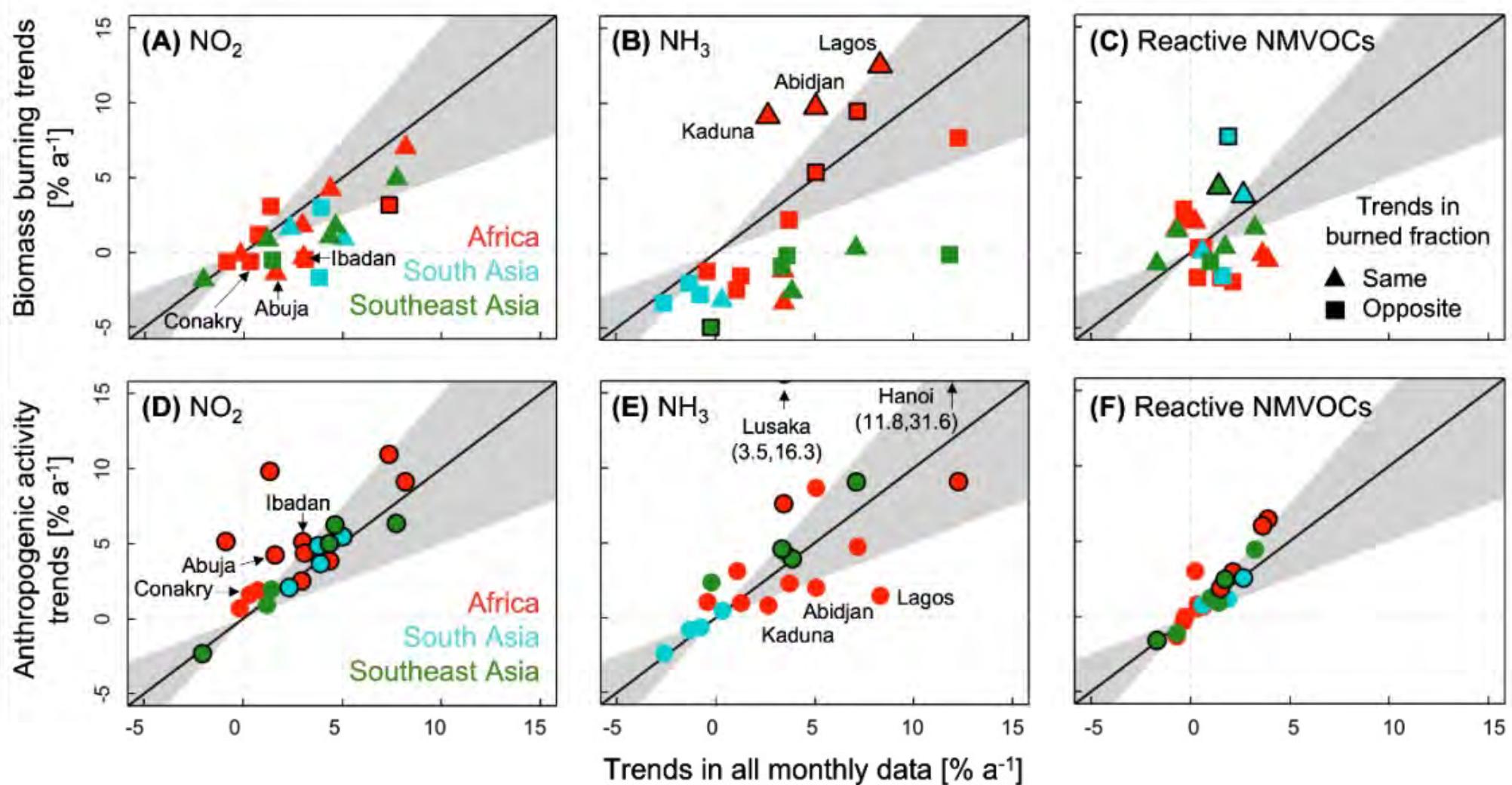


Ratio > 5:
O₃ production sensitive to NO_x

Transitioning to NO_x saturated or VOC sensitive

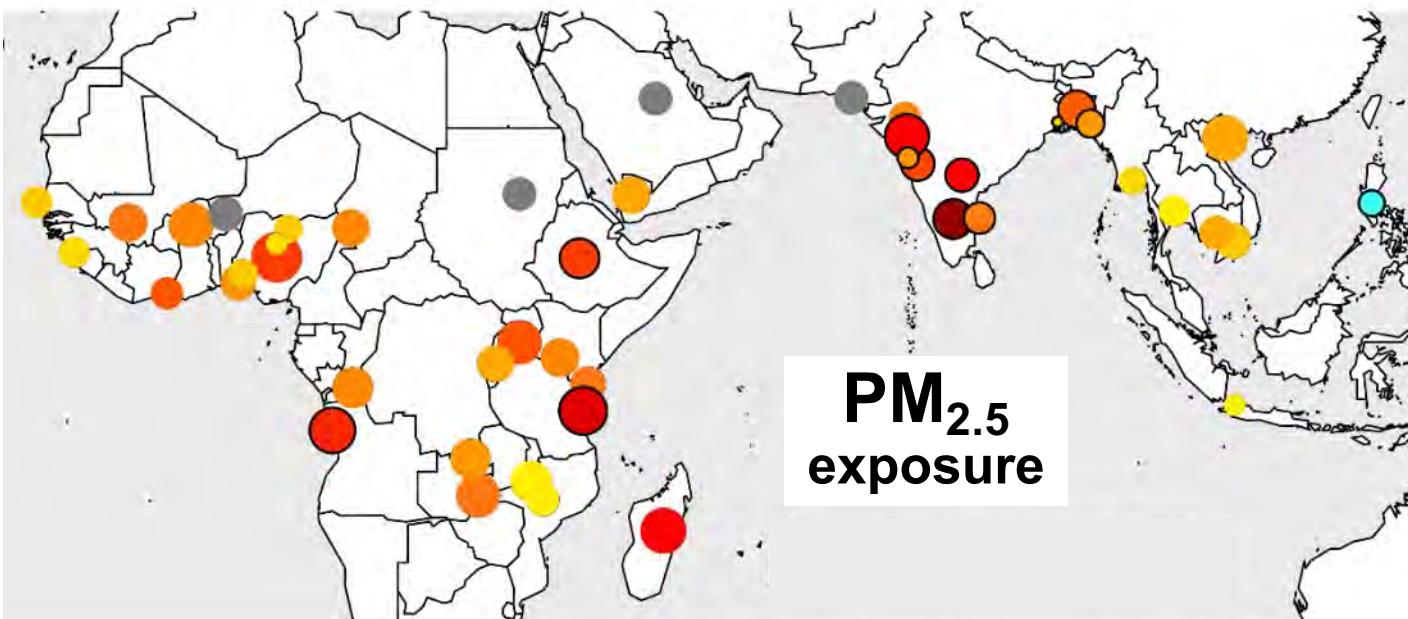
What's driving the observed trends?

We use a statistical approach and knowledge of seasonality of emissions to assess the relative role of anthropogenic and biomass burning emission



Consistency in trends for anthropogenic influenced months and all data months supports anthropogenic emissions as air pollution trend drivers with some offsetting from decline in agricultural activity

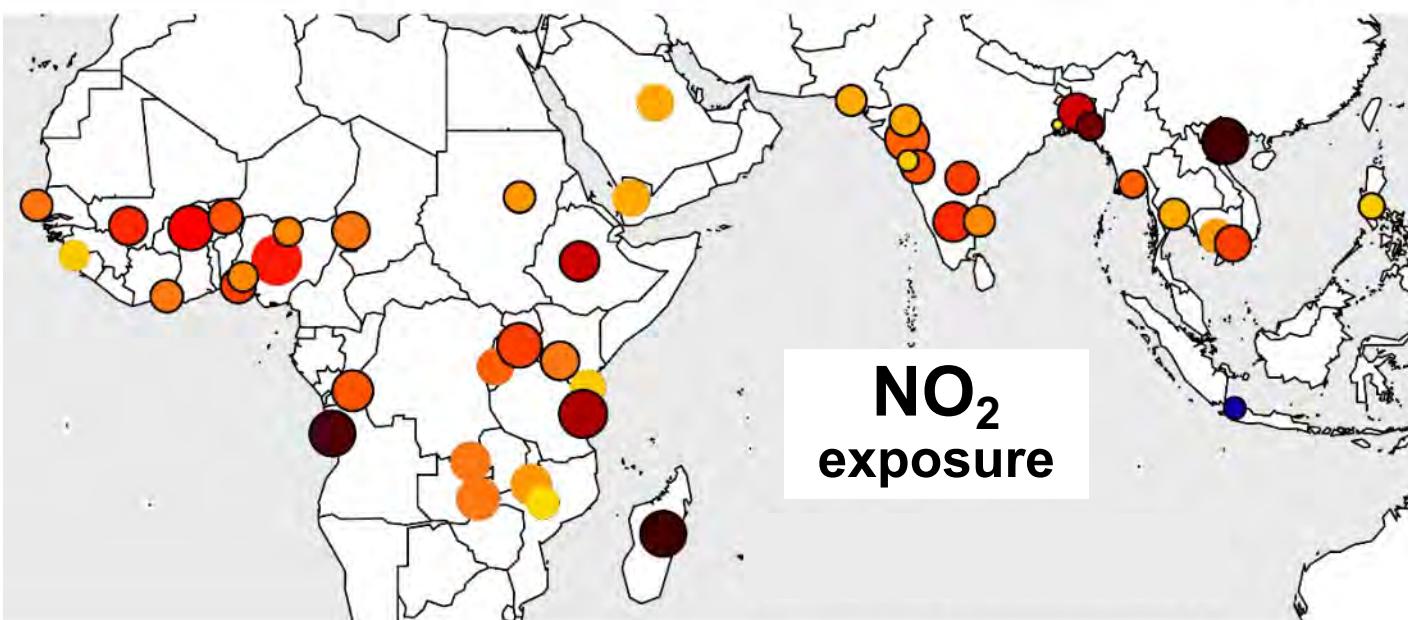
Increase in urban population exposure to air pollution



Combined effect of rapid air quality degradation,
increase in population and urbanization

Up to **18 % a⁻¹** increase
in PM_{2.5} in India

Increased incidence in many health adverse
health outcomes leading to premature death

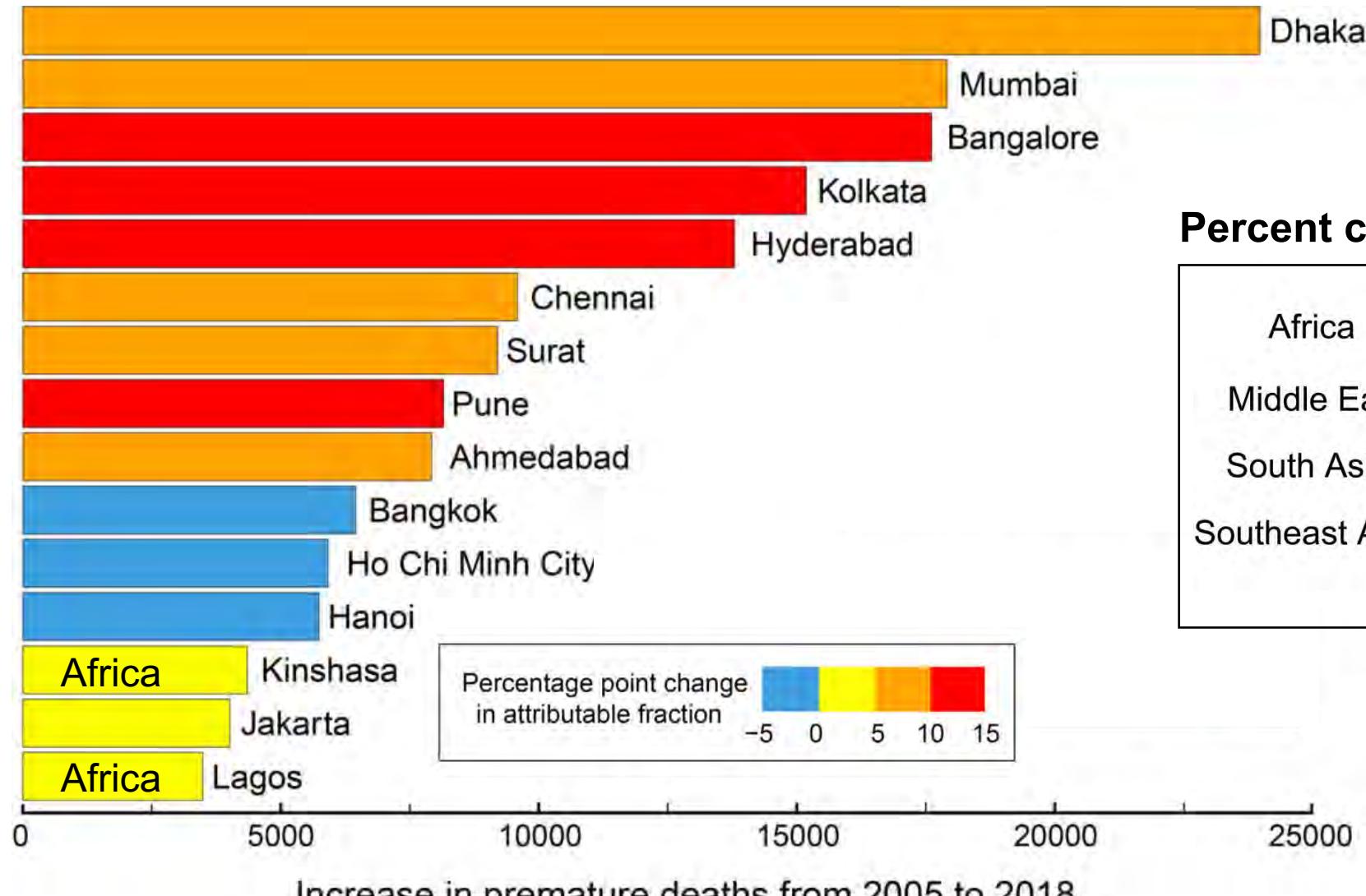


Up to **23% a⁻¹** increase
in NO₂ in many cities

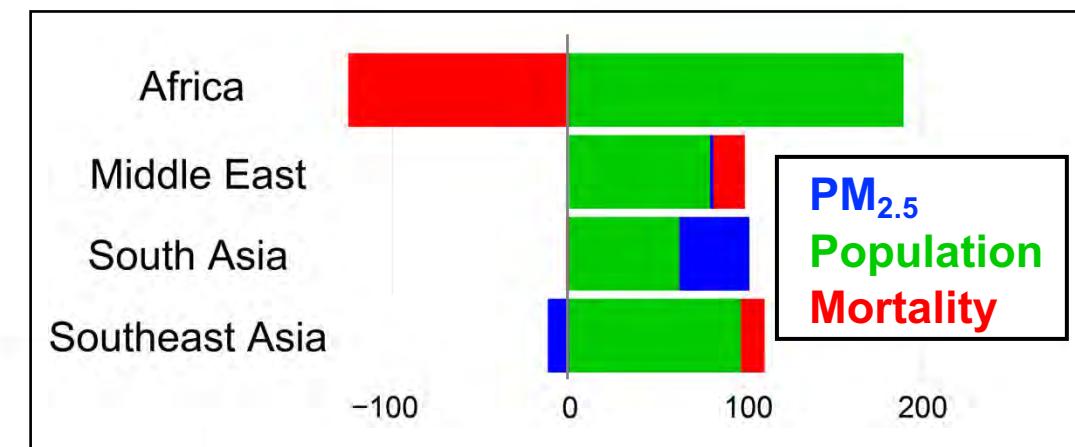


Premature mortality attributable to rise in PM_{2.5} exposure

Ranking of cities with greatest health burden



Percent contribution of individual factors



Total: 179,550

[95% CI: -227,131 to 586,231]

Highest ranked are almost all in Asia. Worst effects in Africa buffered by improvements in healthcare.

Take-homes and additional findings from this work

Shift in dominance from traditional (biomass burning) to a mix of anthropogenic sources

Trends in cities opposite to national and regional trends in Africa

Inventories underestimate growth in precursor emissions suggested by trends from satellite observations

Ozone production transitioning to dependence on volatile organic compounds that are more challenging than NO_x to regulate

Health impacts in cities in Asia likely to occur in cities in Africa in the next 2-3 decades

Link to paper: <https://www.science.org/doi/reader/10.1126/sciadv.abm4435>

Link to New York Times article:

<https://www.nytimes.com/2022/04/08/climate/air-pollution-cities-tropics.html>

Reactive nitrogen in the remote troposphere



Rob Ryan
postdoc



€ 1.5 million
Starting Grant



Nana Wei
PhD



Bex Horner
PhD

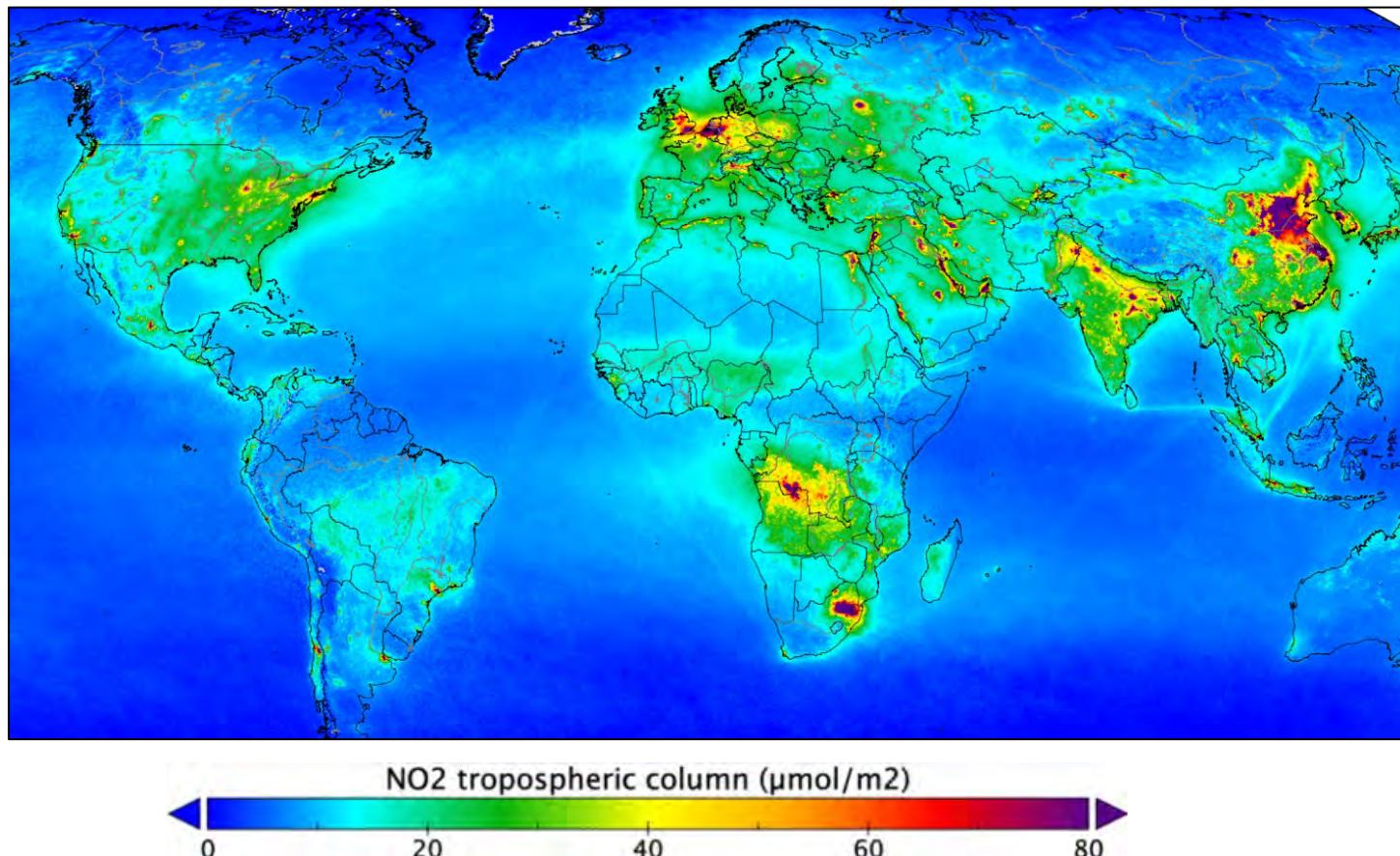


Eleanor Smith
PhD

Reactive nitrogen in the remote troposphere

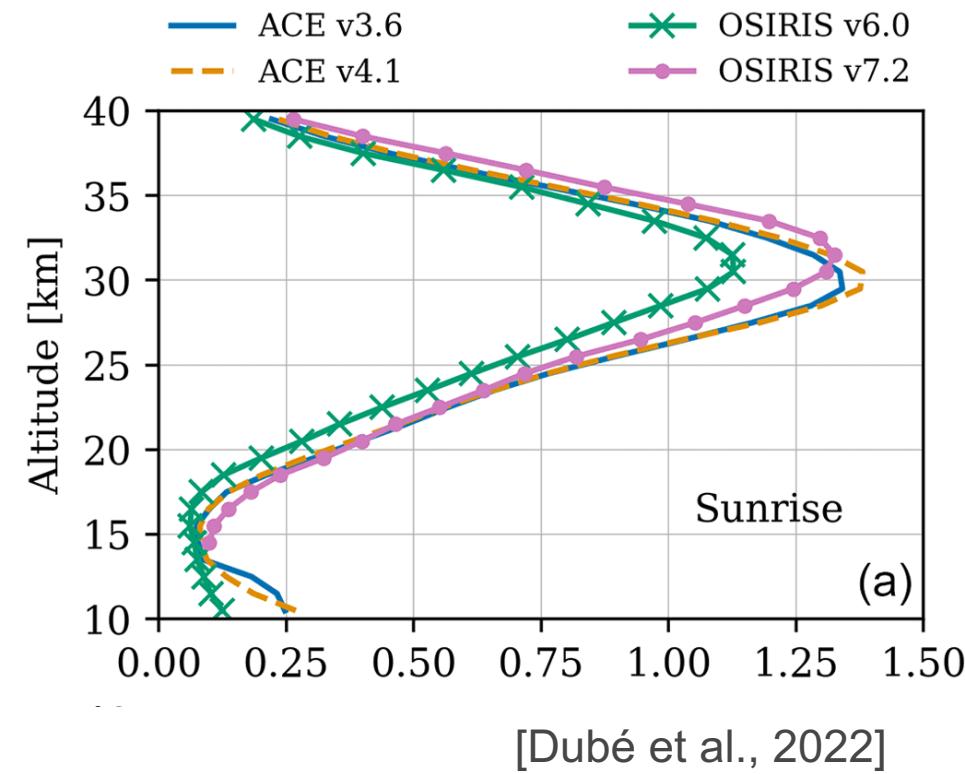
Key to formation of the greenhouse gas tropospheric ozone, but observations are limited

Nadir-viewing instruments observe the whole column



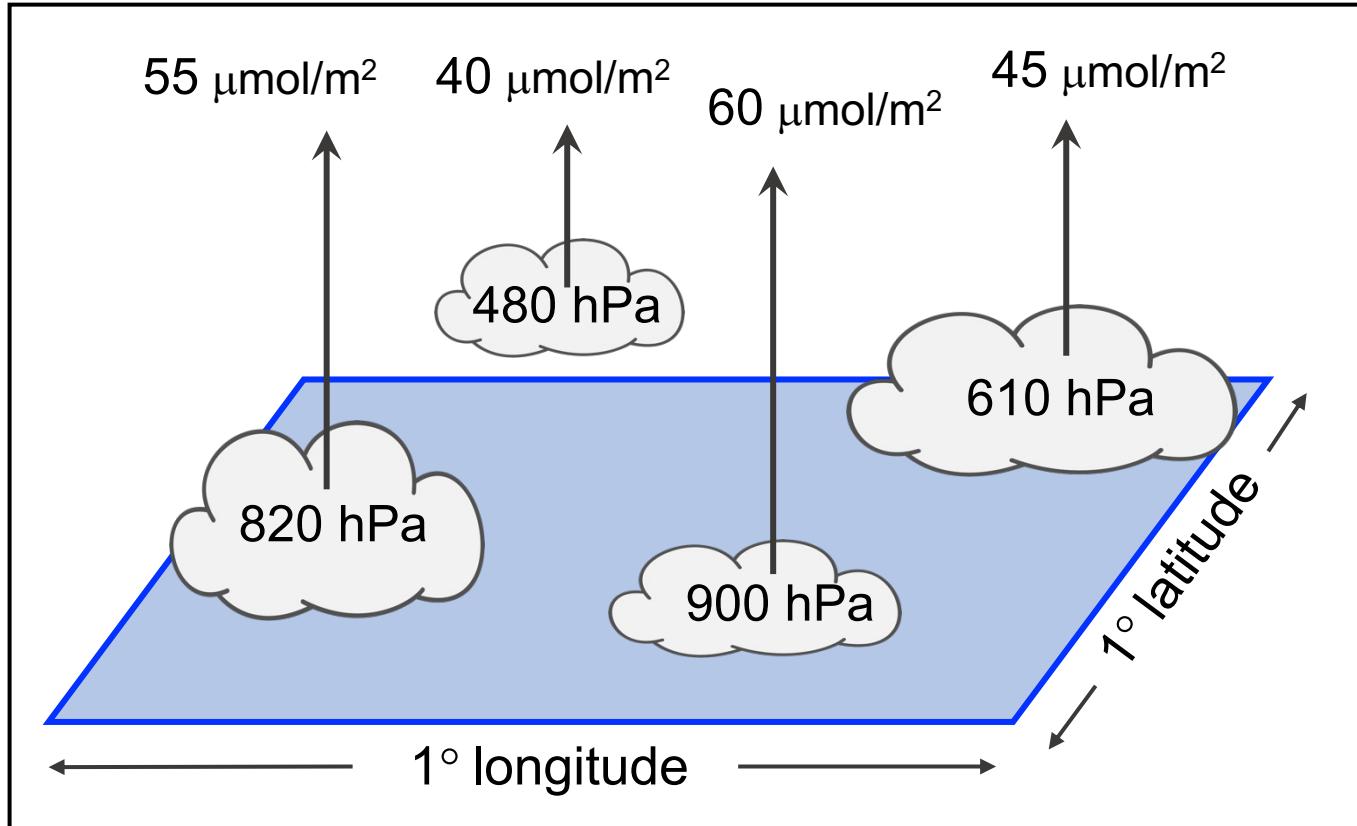
Aircraft observations limited in space and time

Limb-viewing instruments not sensitive to troposphere

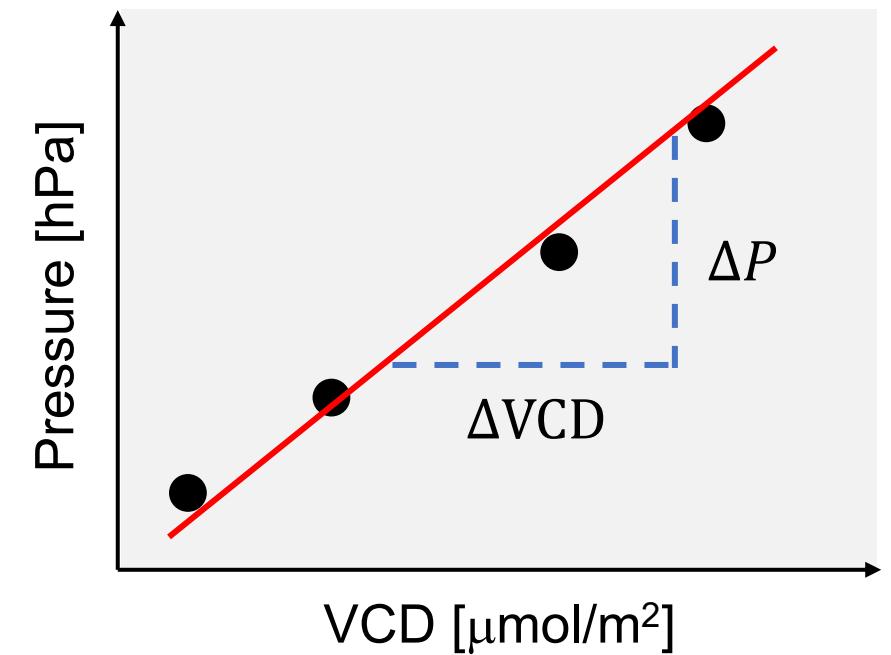


Cloud-slicing satellite observations to address data scarcity

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:

$$\text{NO}_2 \text{ VMR} = \frac{\Delta \text{VCD}}{\Delta P} \times \text{const}$$

Application to high-resolution TROPOMI instrument



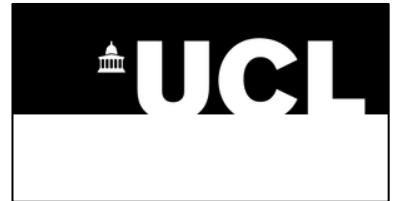
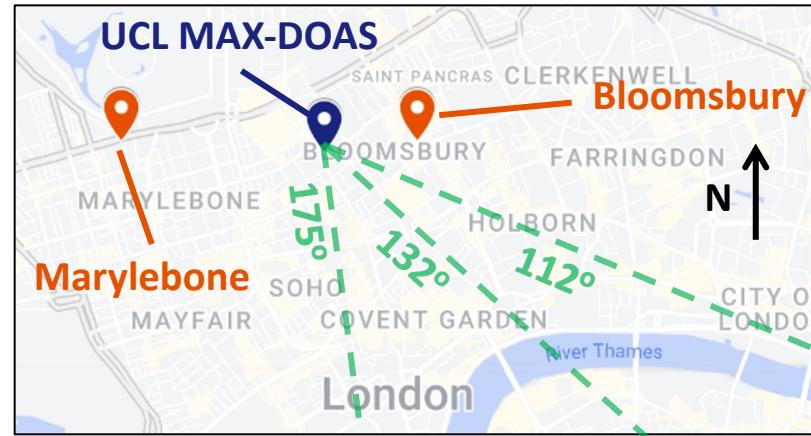
UV/vis spectrometer

13h30 overpass time

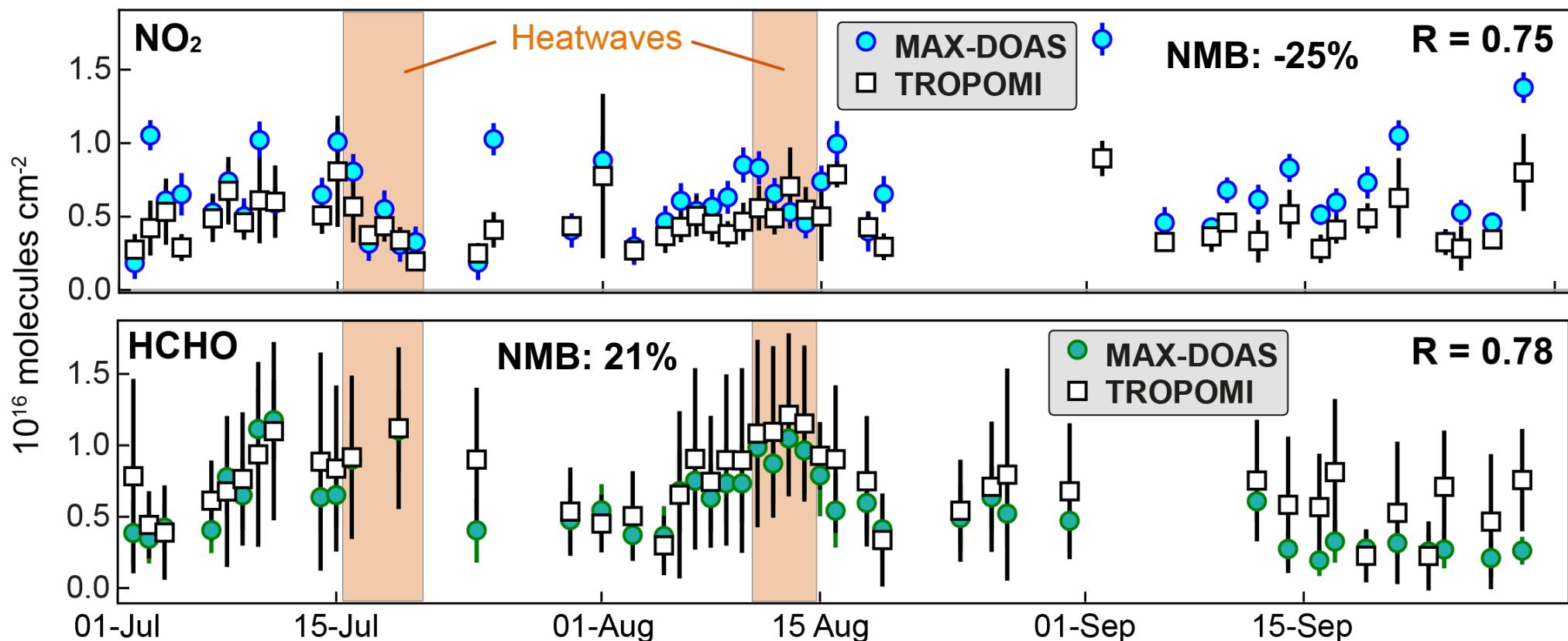
Daily global coverage

5.6 km × 3.5 km resolution

TROPOMI validation with MAX-DOAS during 2022 heatwaves

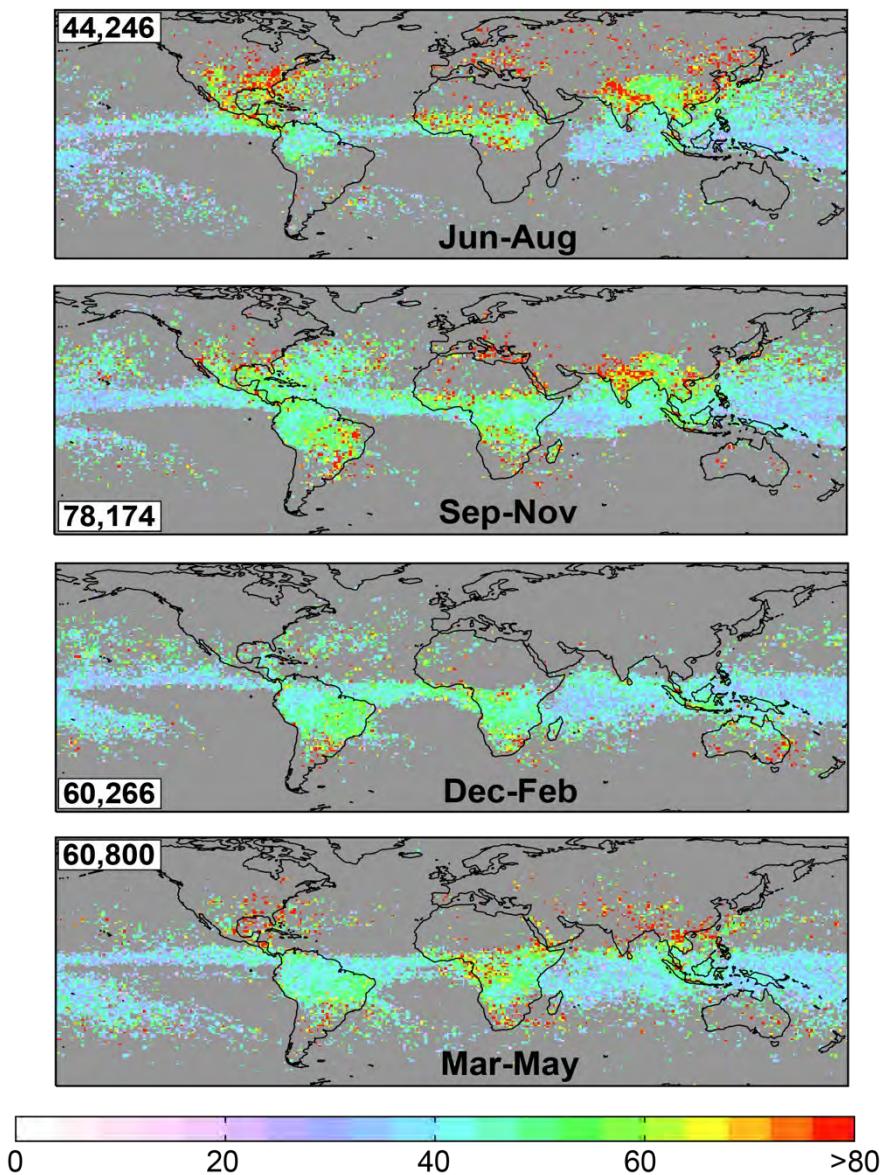


Capital Equipment
Fund

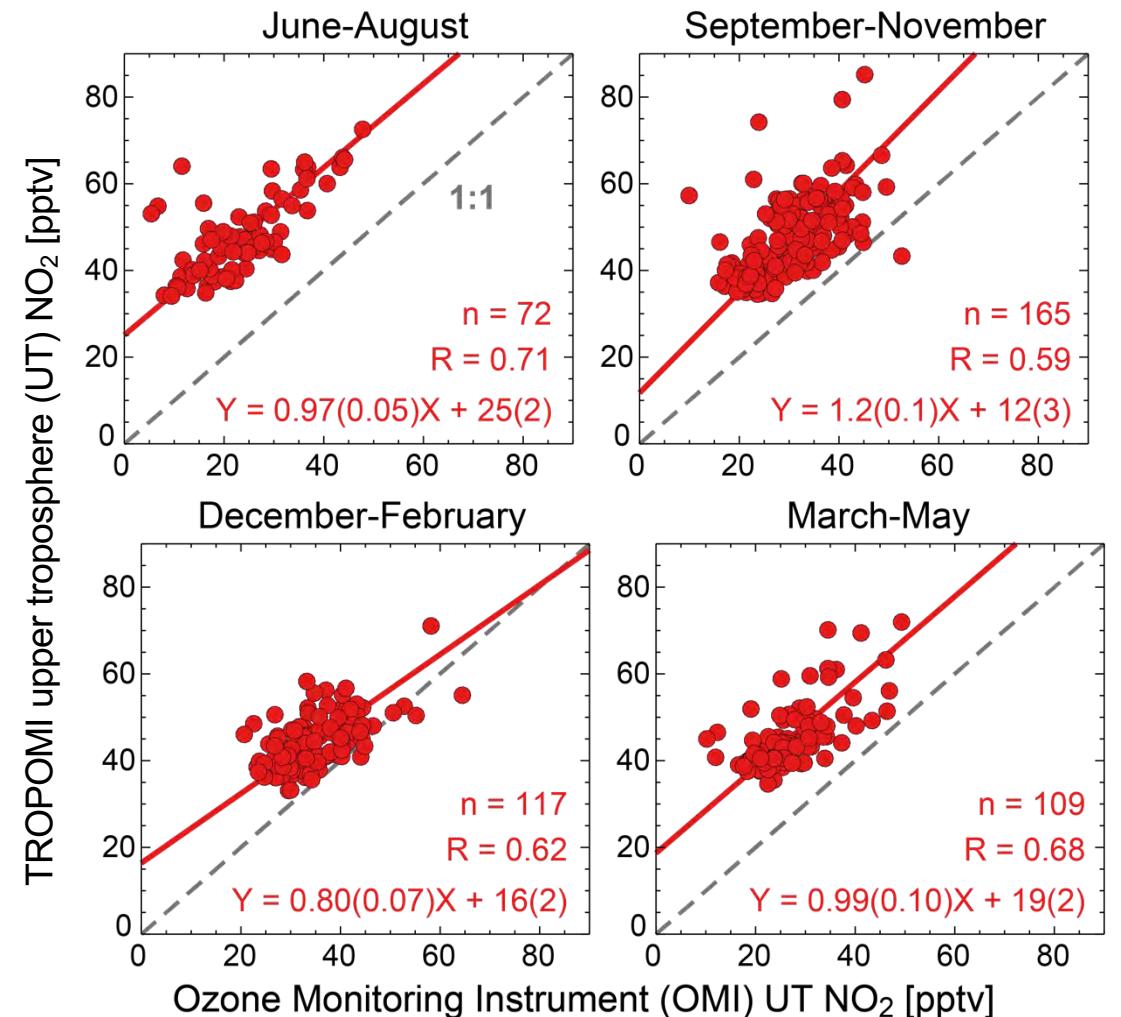


Seasonal means of NO₂ in the upper troposphere

Seasonal means at 8-12 km



Evaluation against product from OMI



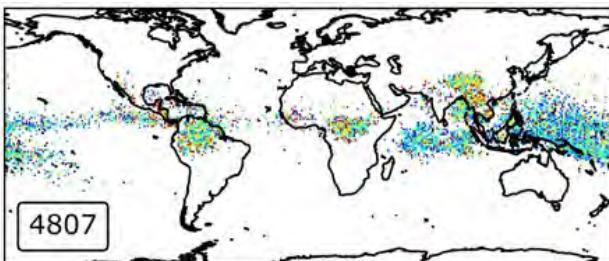
[Marais et al.,
AMT, 2021]

OMI data from S. Choi and J. Joiner at NASA

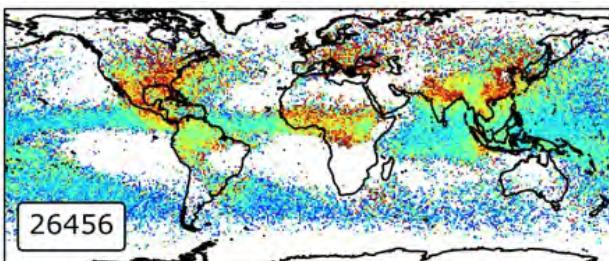
Vertical profiles of NO₂ derived with the TROPOMI instrument

Vertical profiles in Jun-Aug

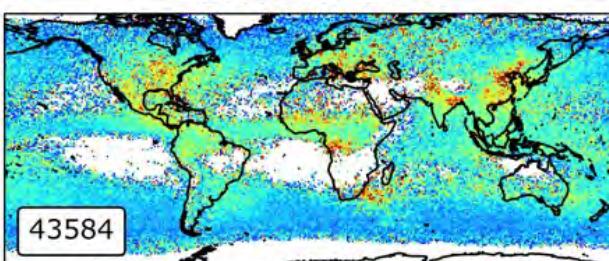
180-320 hPa
9-12 km



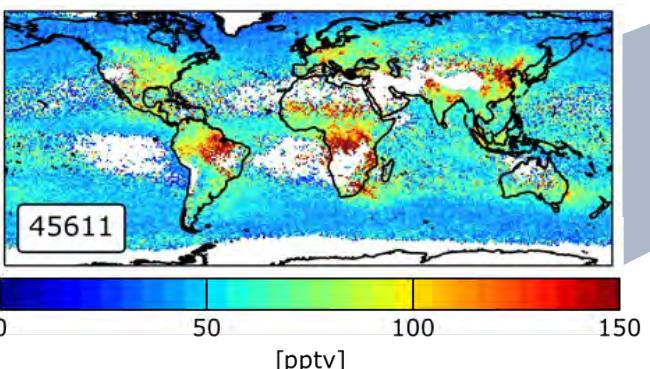
320-450 hPa
6-9 km



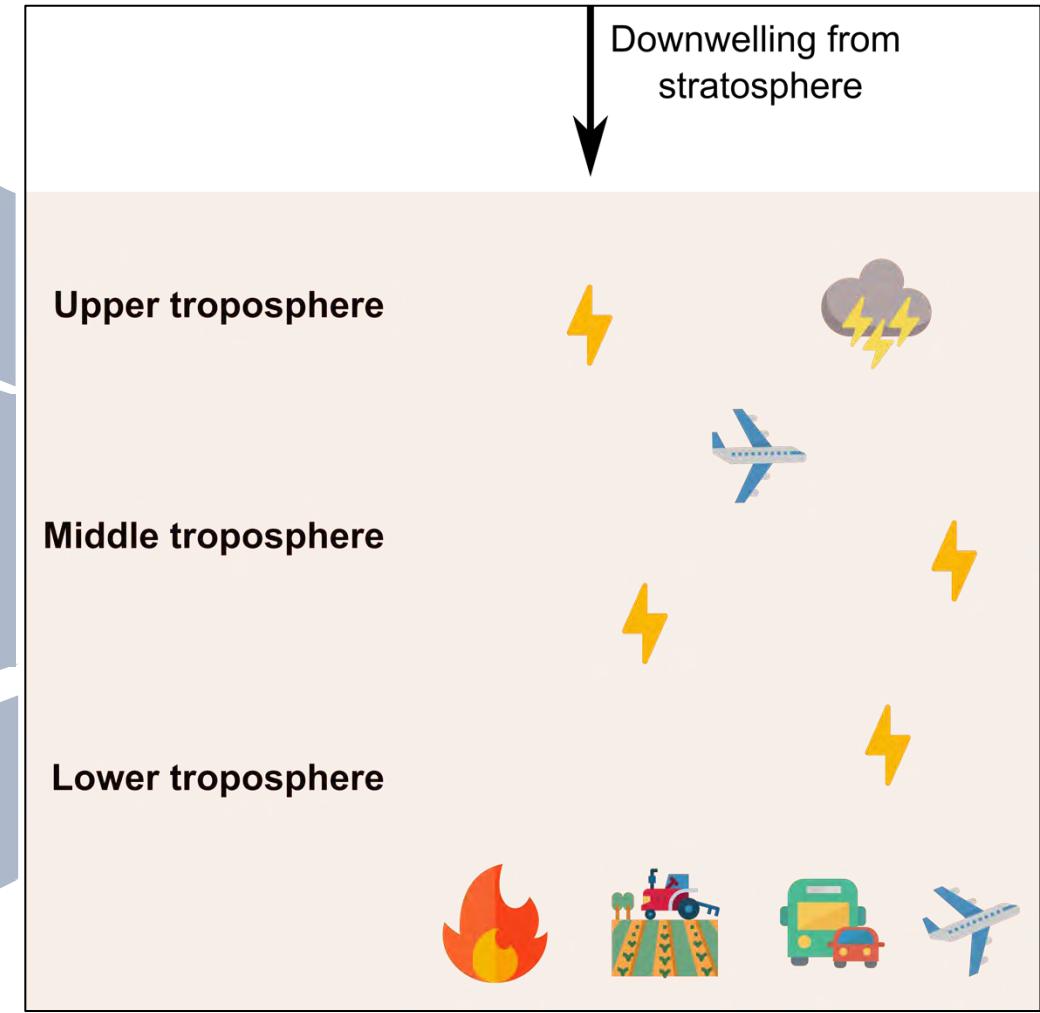
450-600 hPa
4-6 km



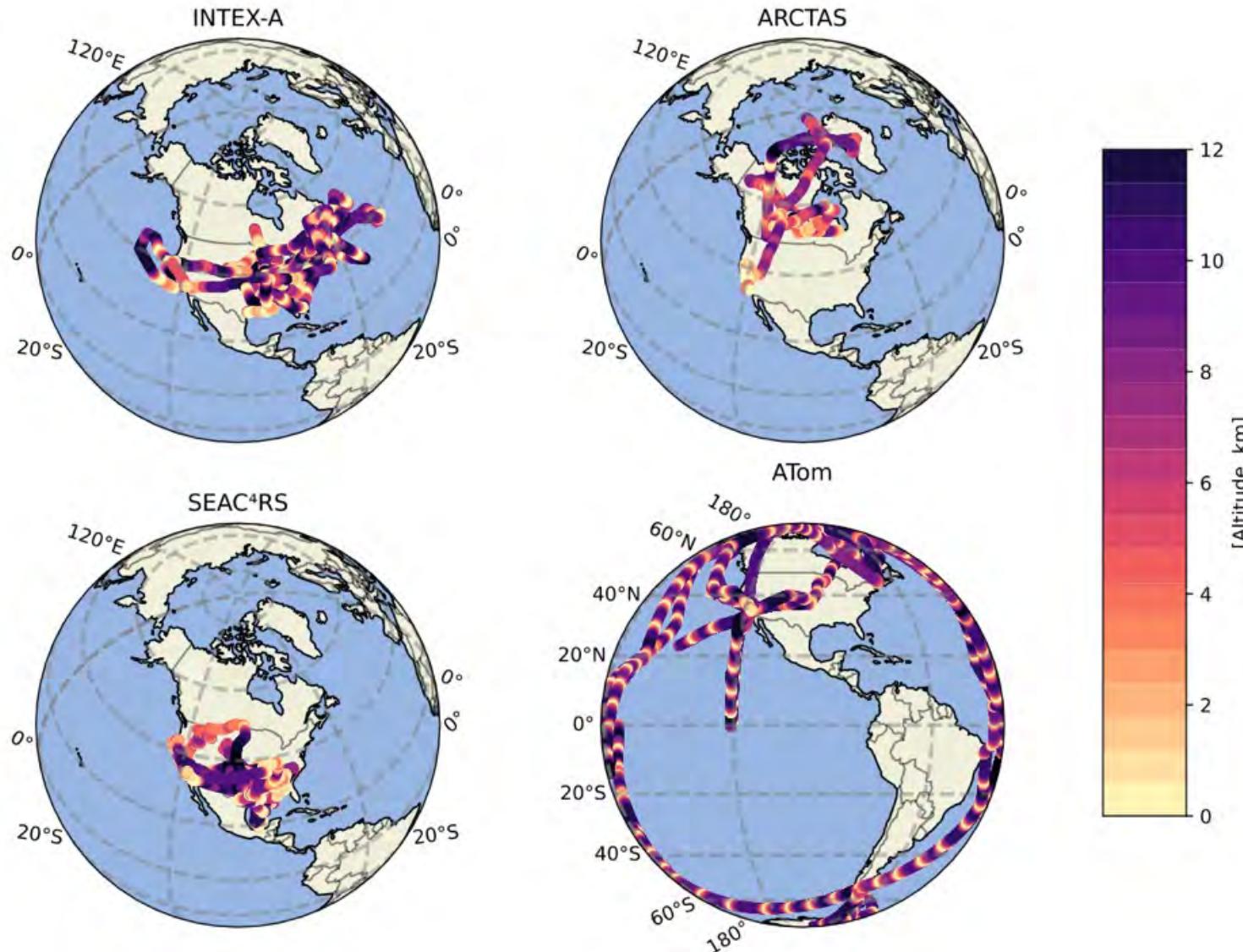
600-800 hPa
2-4 km



[Horner et al.,
in prep]



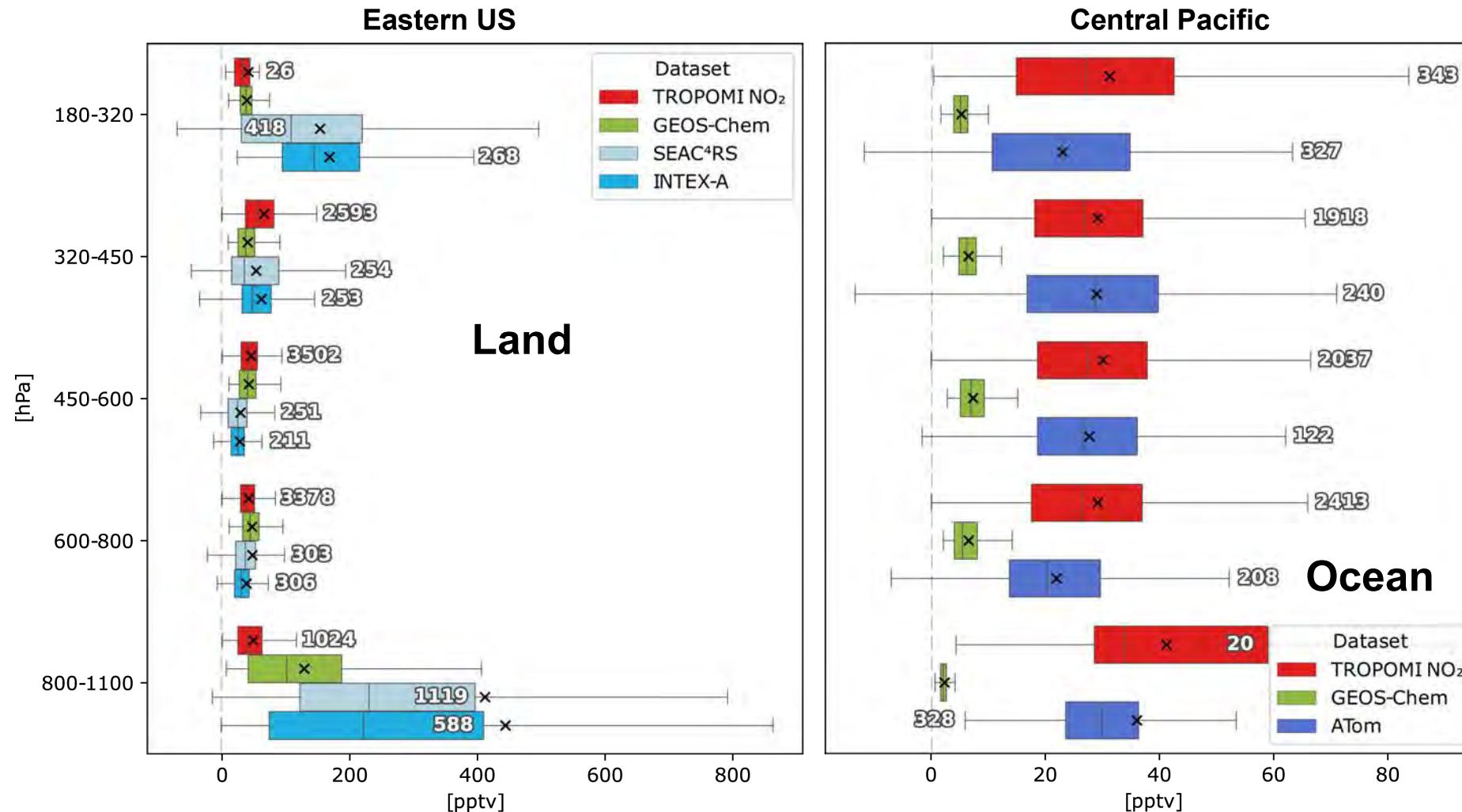
Cloud-sliced product validation with aircraft observations



Data provided by NASA DC8 Science Teams

Cloud-sliced product validation with aircraft observations

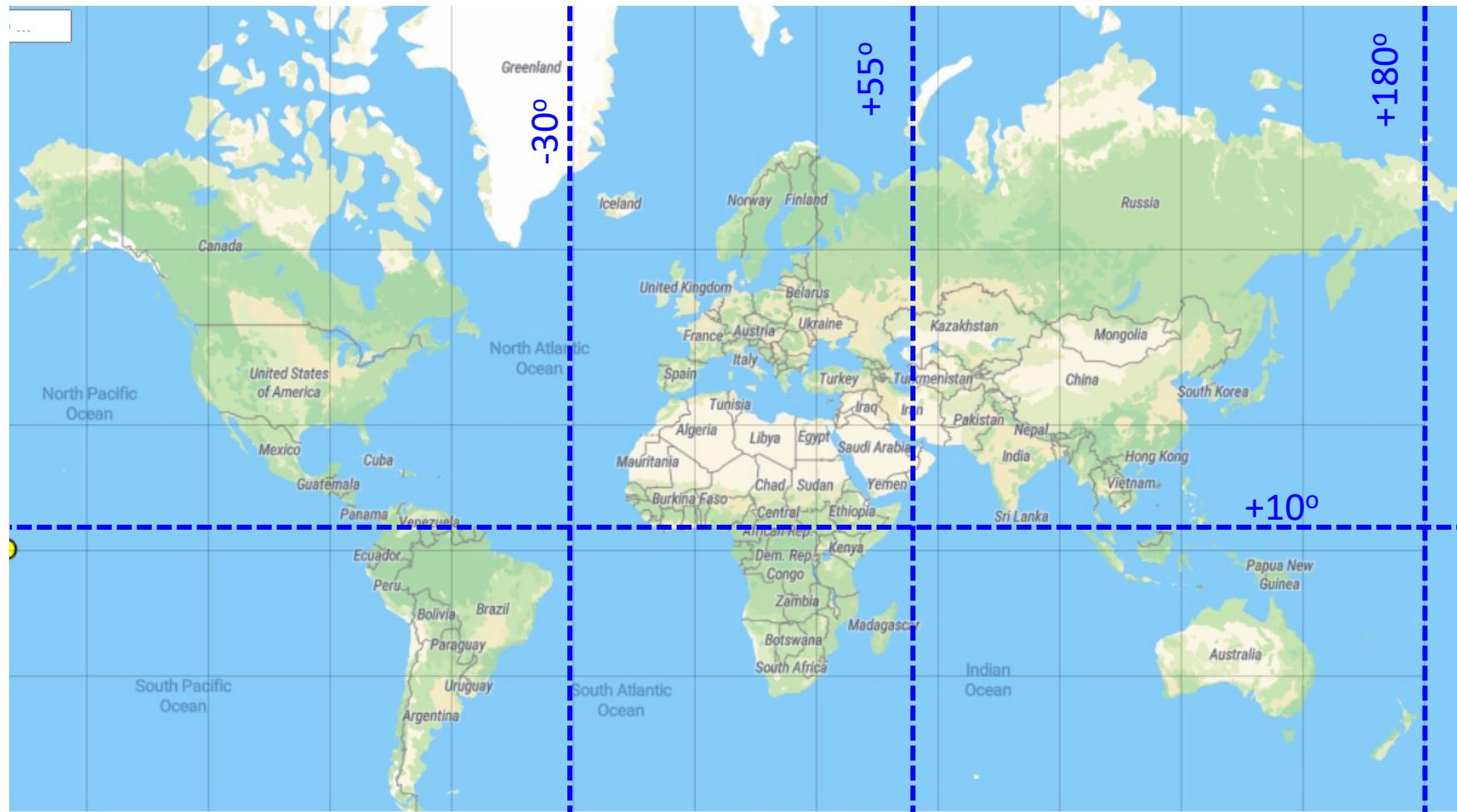
Comparison of collocated NO₂ mixing ratios in June-August



Aircraft and TROPOMI consistent (20-30 pptv) in the mid troposphere

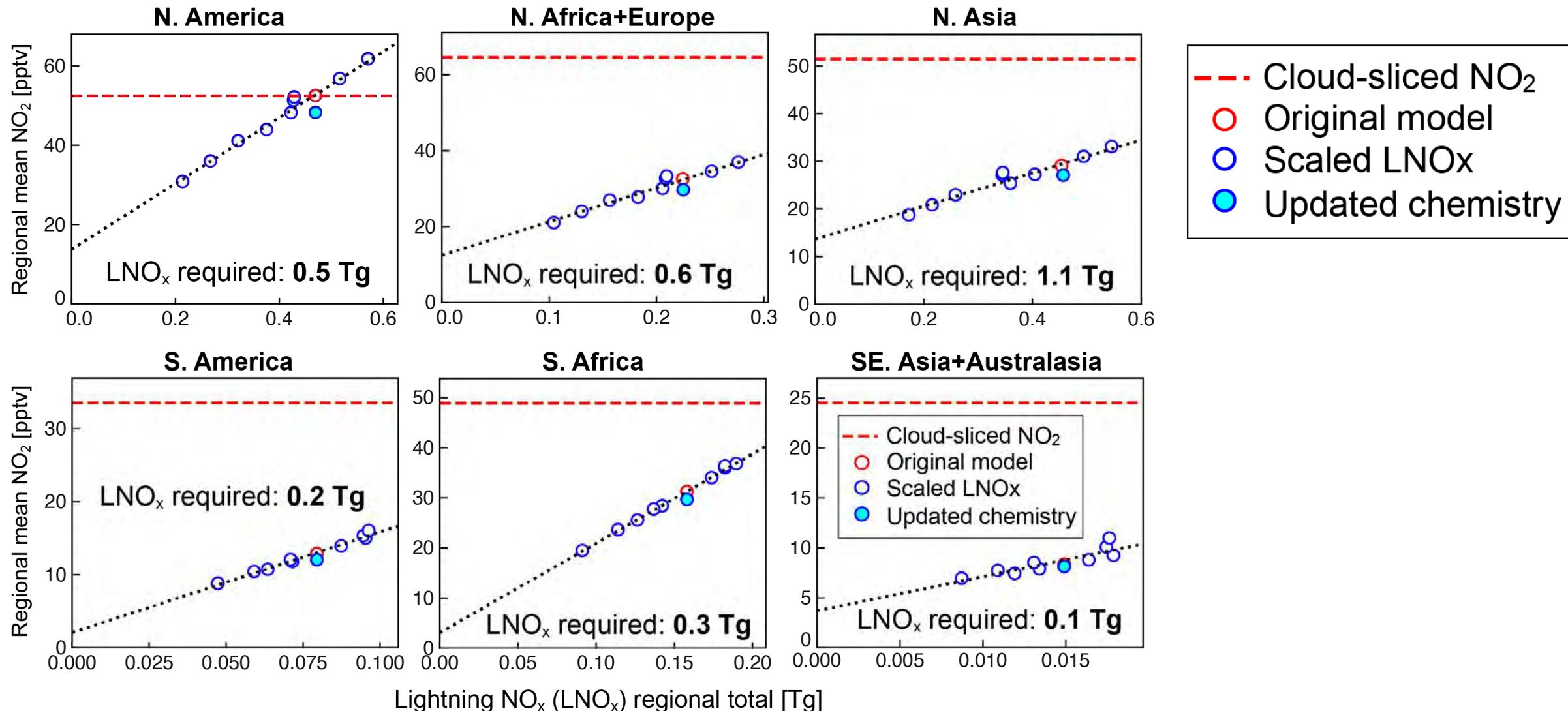
Model (GEOS-Chem) biased low throughout troposphere over remote ocean

Regional sensitivity to lightning NO_x emissions



Regional sensitivity to lightning NO_x emissions

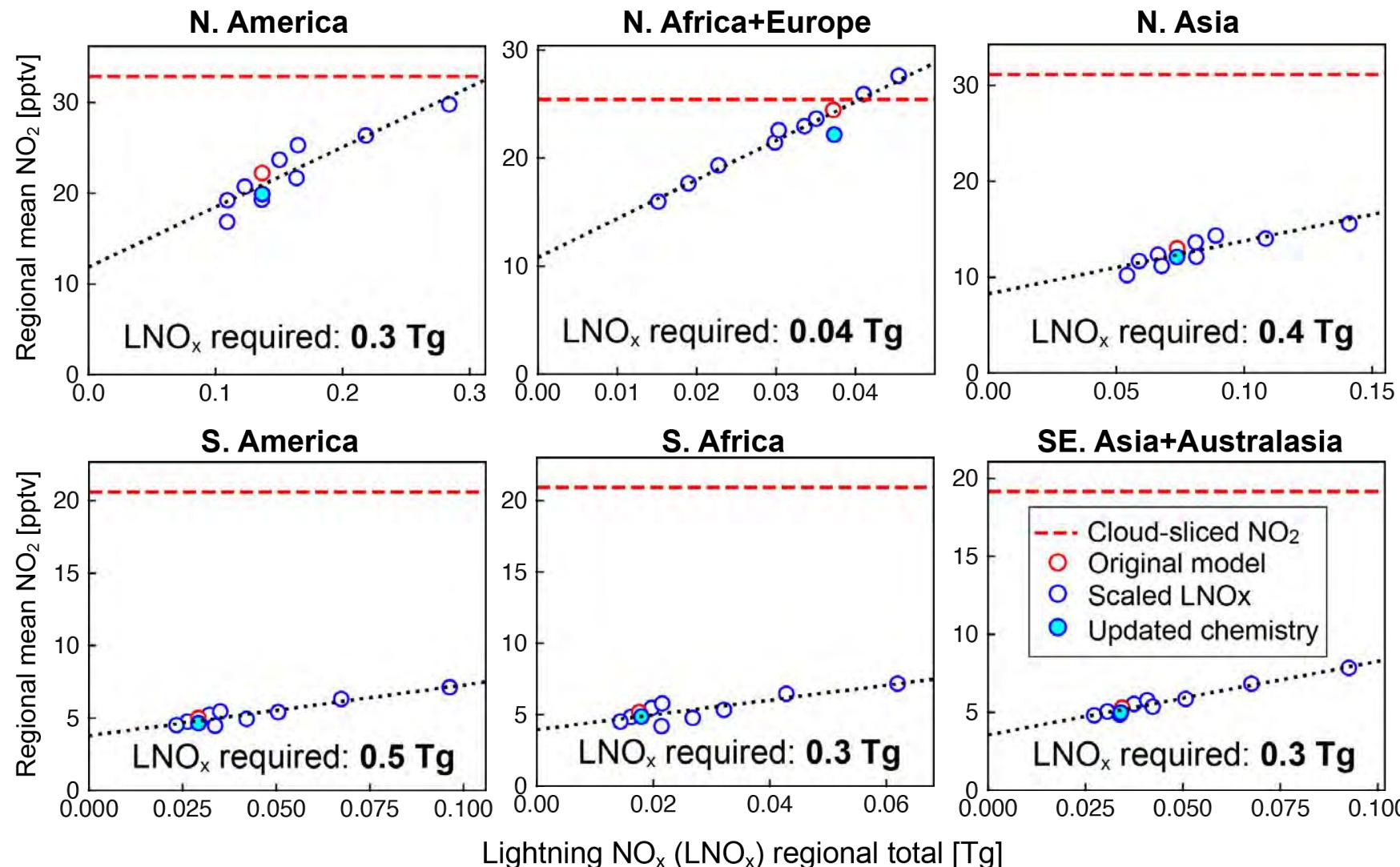
Observed versus modelled June-August upper tropospheric regional mean NO₂ over land



Original model emissions of 1.4 Tg NO, whereas 2.7 Tg NO required to match cloud-sliced NO₂

Regional sensitivity to lightning NO_x emissions

Observed versus modelled June-August upper tropospheric regional mean NO₂ over the ocean

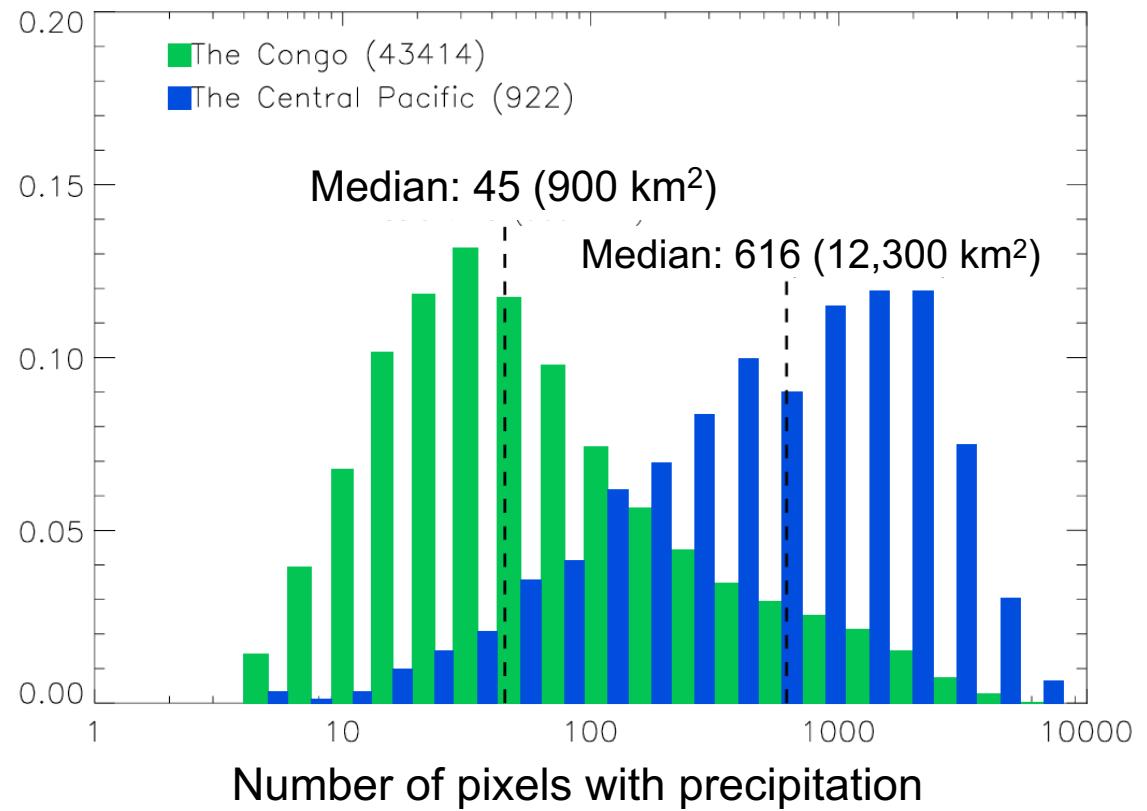


Original model emissions of 0.3 Tg NO, whereas 1.9 Tg NO required to match cloud-sliced NO₂

Lightning characteristics over the ocean and over land

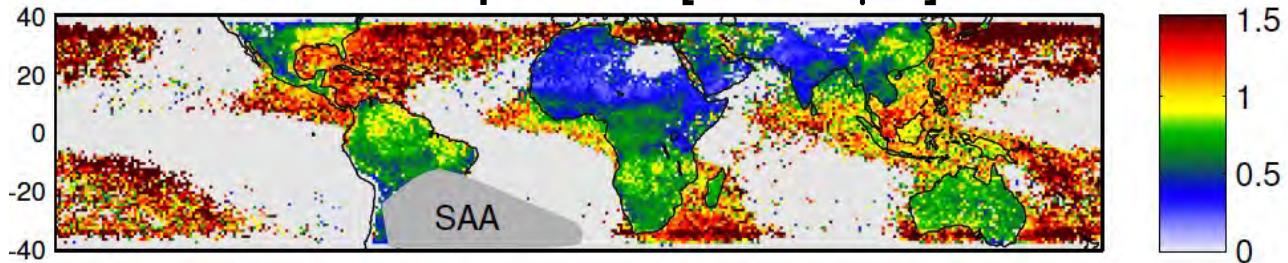
Support for larger, more persistent and higher energy lightning flashes over the ocean than over land

Frequency distribution for all radar precipitation features with lightning

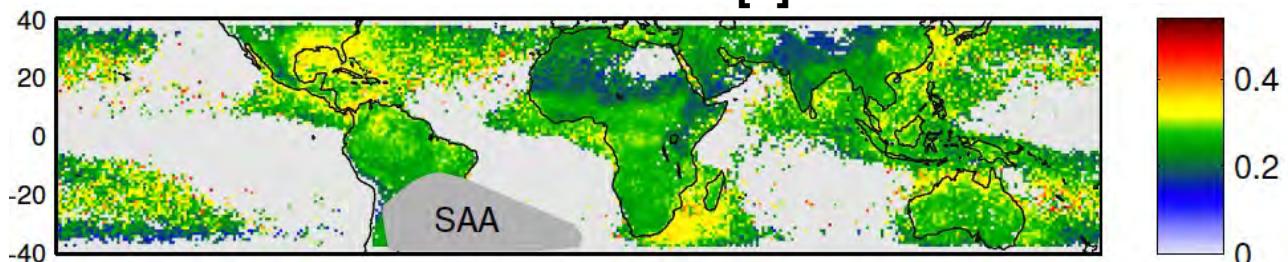


[Bang & Zipser, 2015]

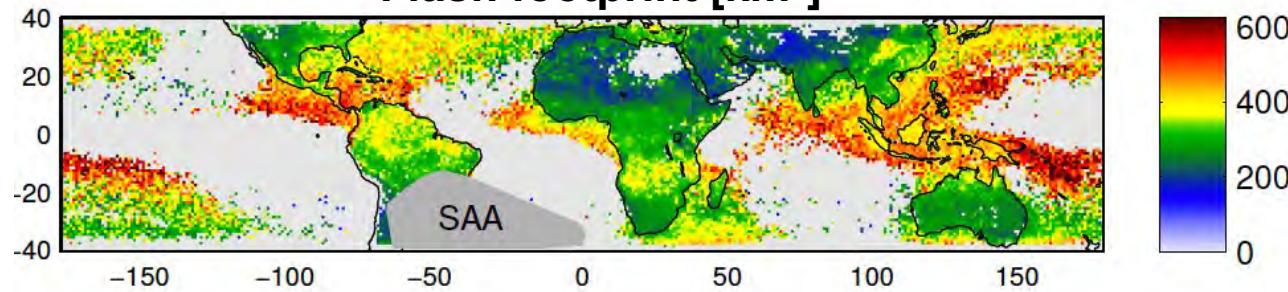
Radiance per flash [J/m²/sr/μm]



Flash duration [s]



Flash footprint [km²]



[Beirle et al., 2014]

Concluding Remarks

Cloud-sliced profiles of NO₂ in the mid-troposphere consistent with aircraft observations

GEOS-Chem reproduces observations over land, but has a large low bias over the remote ocean

Modelled regional mean NO₂ sensitive to lightning NO_x emissions

Addressing the model bias requires almost 3-fold increase in global lightning NO_x emissions with implications for tropospheric ozone production

Environmental impact of the modern space sector



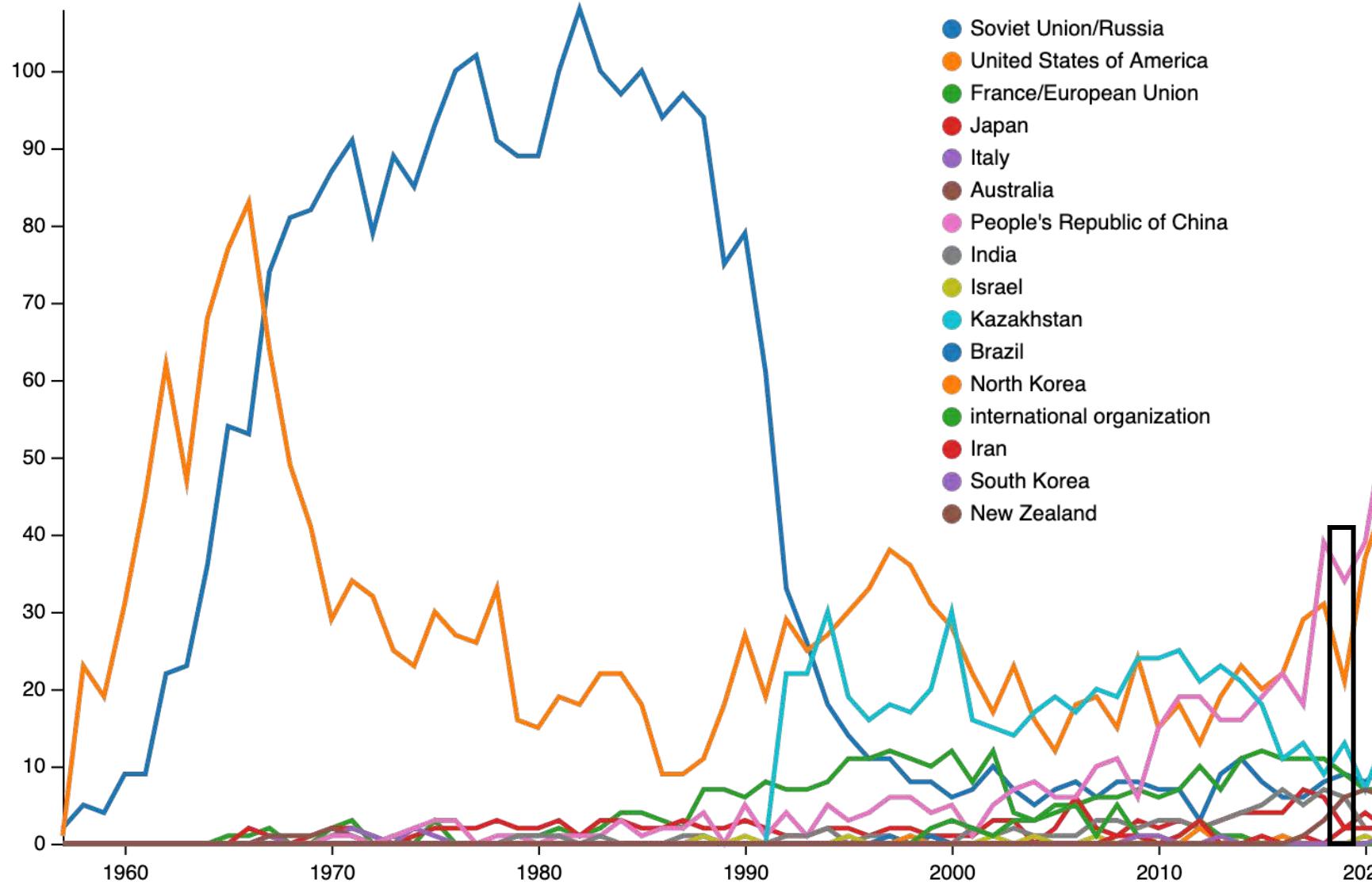
Chloe Balhatchet
summer student



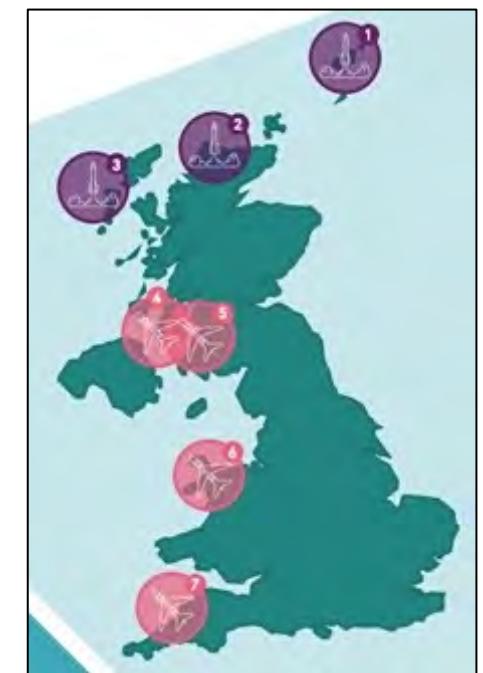
Rob Ryan
postdoc

More diverse space sector than the original space race

Number of rocket launches per country in each year



Even the UK is joining the race:



Dramatic increase in objects in space

Number of objects launched each year

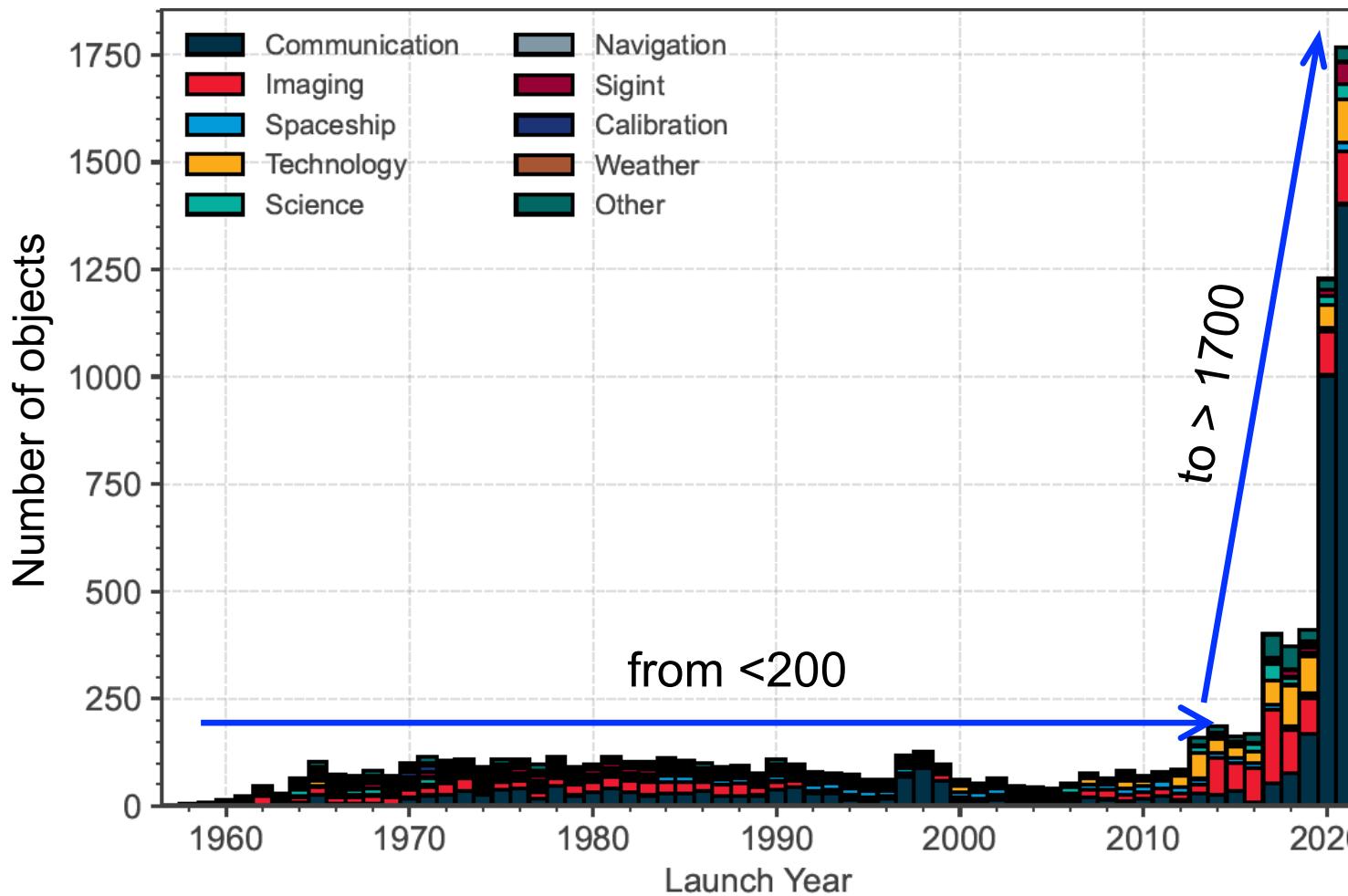


Image from ESA's Annual Space Environment Report, 2022

Only viable disposal method is complete burn up by re-entering Earth's atmosphere

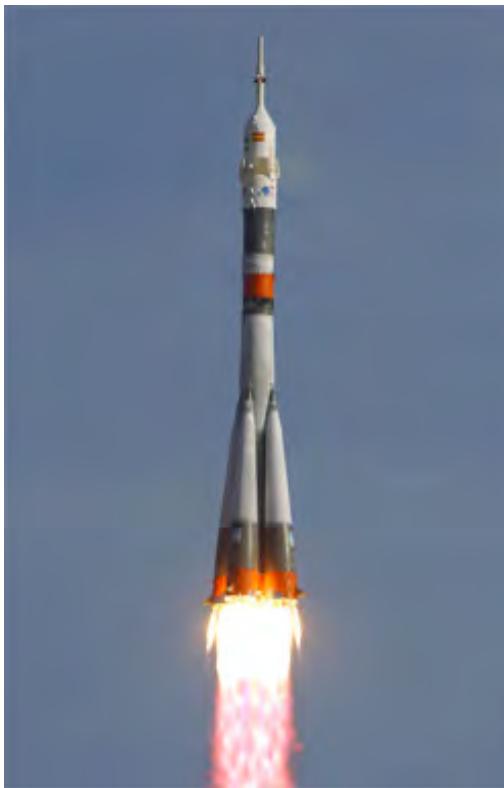
Air pollutant emissions from rocket launches

Solid



NO_x
 HCl+Cl
 Al_2O_3
 H_2O
BC

Hypergolic



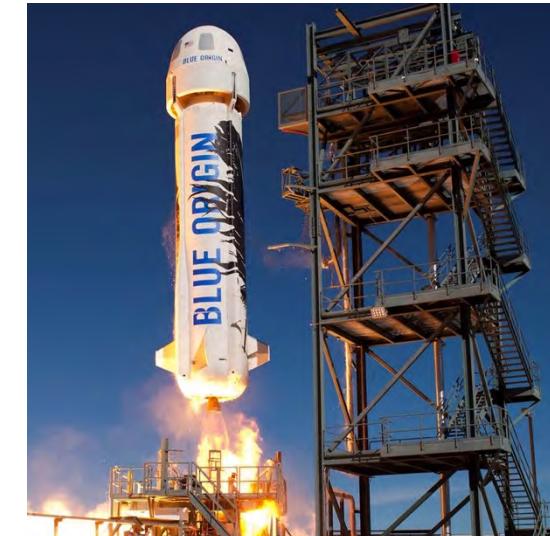
NO_x
 H_2O
BC

Kerosene



NO_x
 H_2O
BC

Cryogenic



NO_x
 H_2O

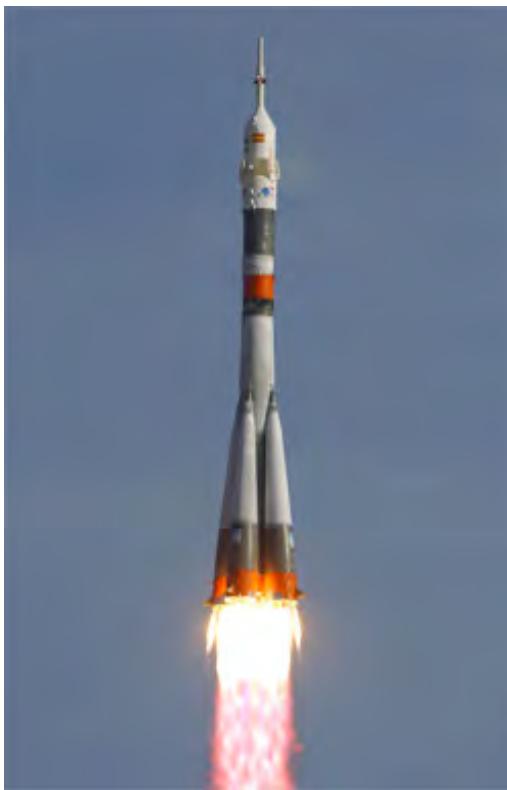
Air pollutant emissions from rocket launches

Solid



NO_x
 HCl+Cl
 Al_2O_3
 H_2O
 BC

Hypergolic



NO_x
 H_2O
 BC

Kerosene



NO_x
 H_2O
 BC

Cryogenic



NO_x
 H_2O

Climate
concern

Air pollutant emissions from rocket launches

Solid



NO_x
 HCl+Cl
 Al_2O_3
 H_2O
BC

Hypergolic



NO_x
 H_2O
BC

Kerosene



NO_x
 H_2O
BC

Cryogenic



NO_x
 H_2O

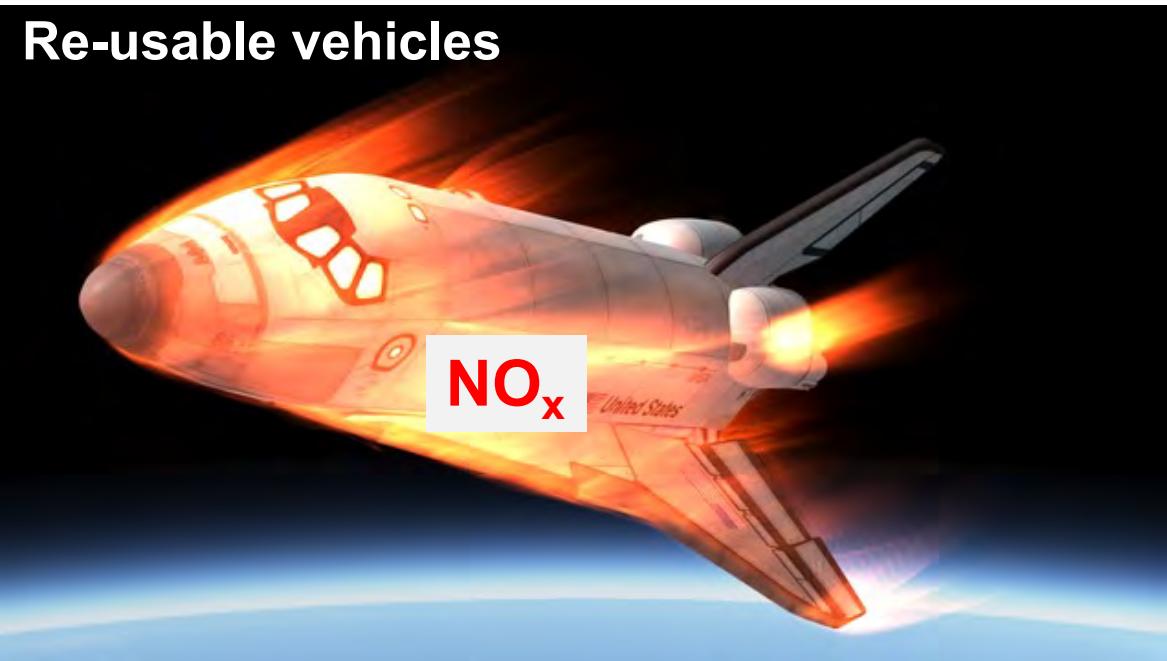
Ozone
depletion

Air pollutant emissions from re-entry

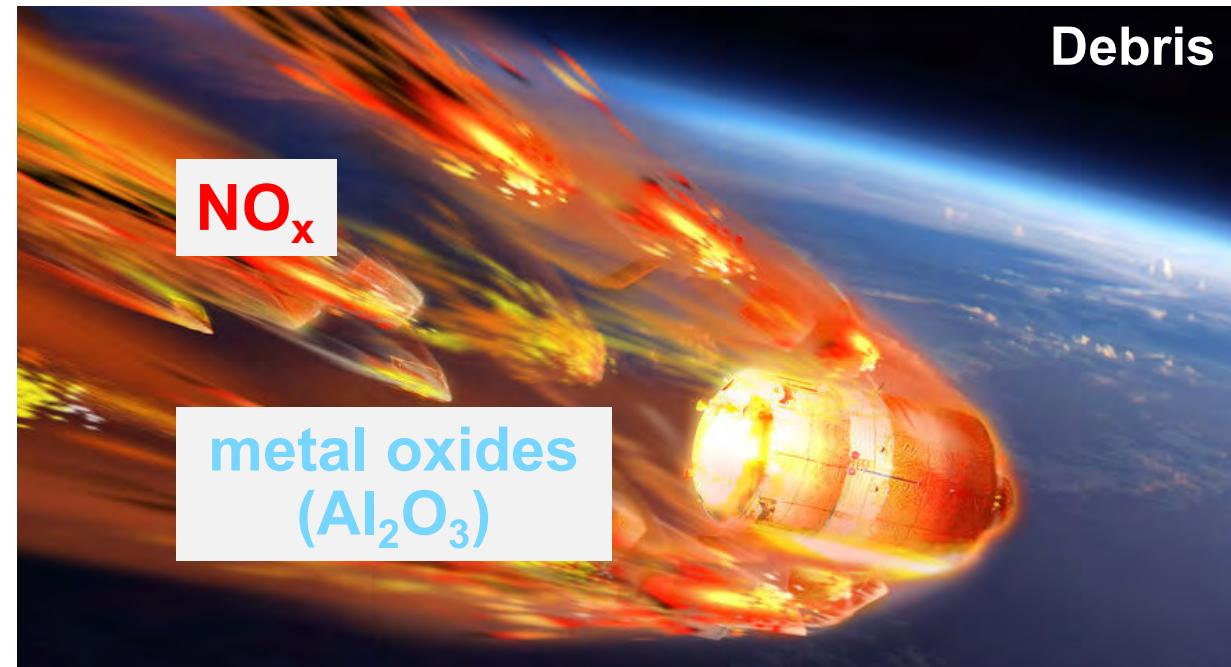


2-40 Gg NO_x per year

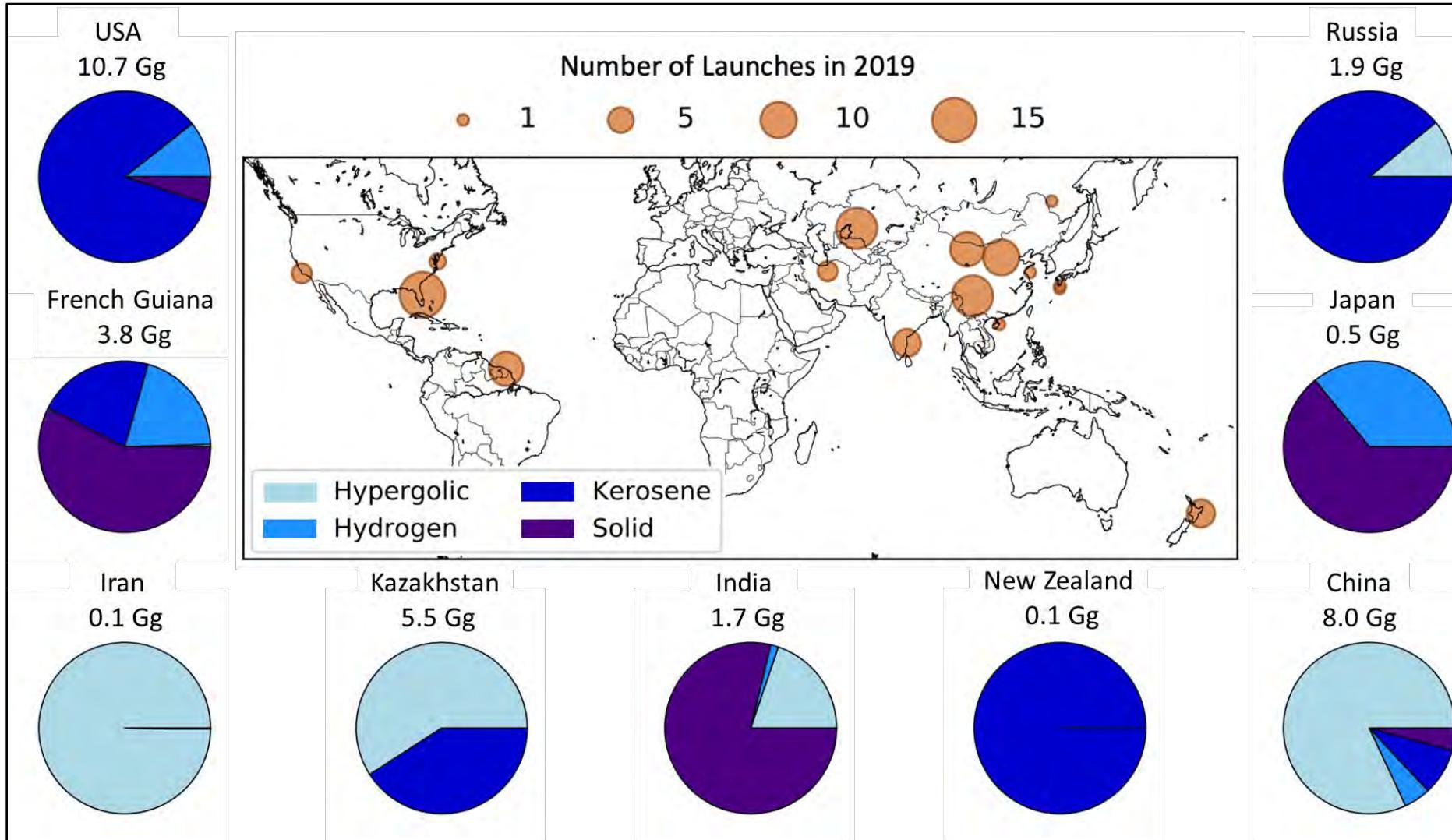
Re-usable vehicles



Debris



Calculate and map a single year of emissions



Annual Emissions:

H₂O: 11 Gg
BC: 0.5 Gg
Al₂O₃: 2 Gg
HCl: 1 Gg
Launch NO_x: 0.2 Gg
Re-entry NO_x: 2 Gg

Artificial NO_x similar
to lower end estimate
of natural NO_x

~100 successful launches in 2019

Reaches 135 in 2021. Already 148 in 2022.

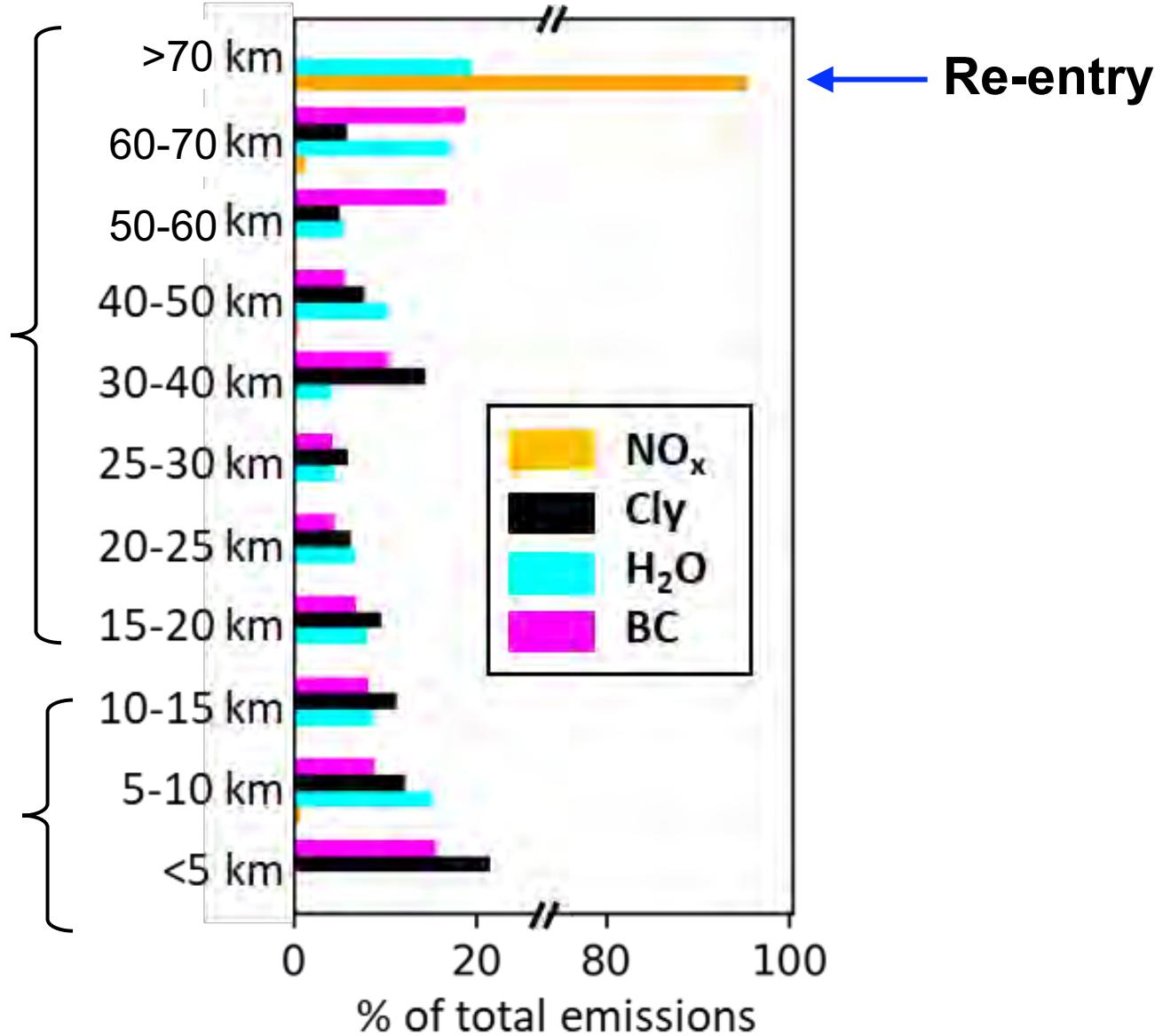
Incorporate these in GEOS-Chem

GEOS-Chem extends
to 80 km

Stratosphere & mesosphere:

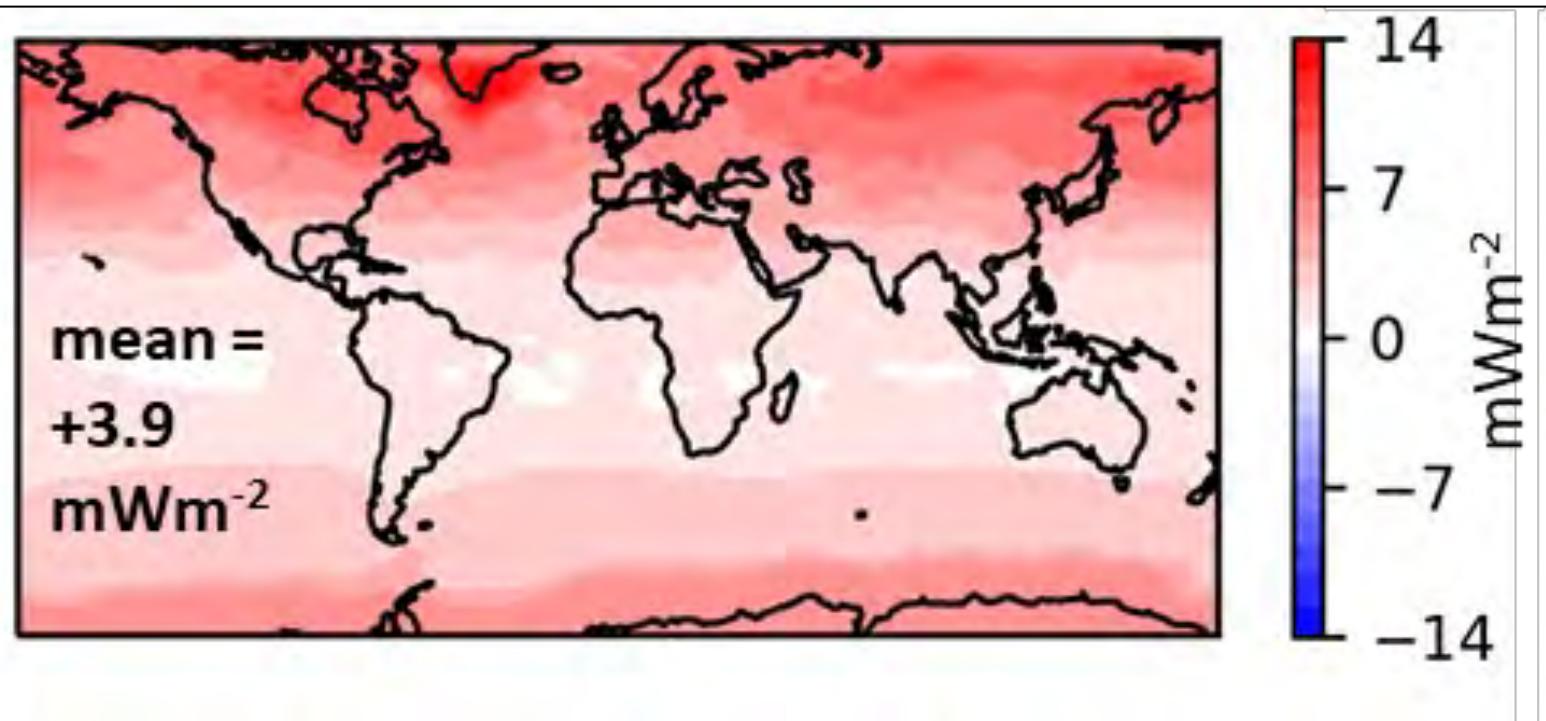
lifetime >2 years
(*gravitational settling*)

Troposphere:
lifetime weeks to months
(*wet and dry deposition,
subsidence, chemical losses*)

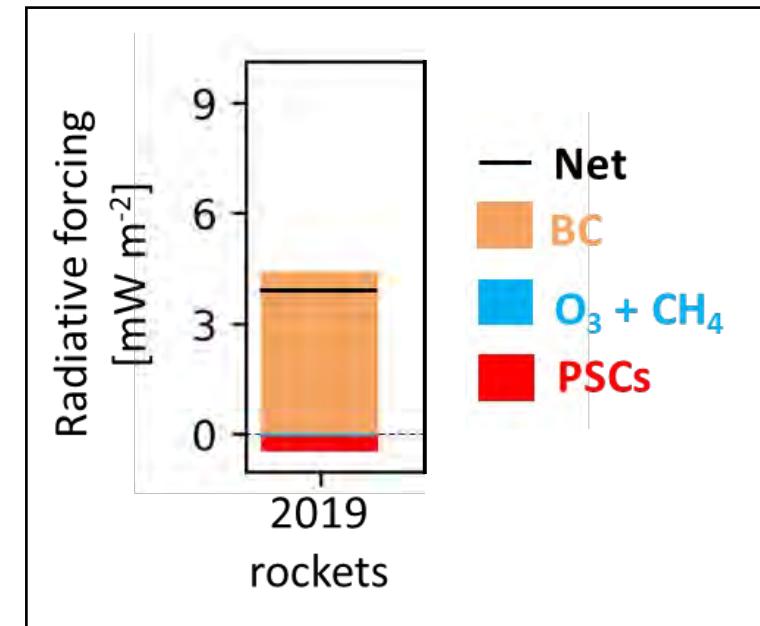


Radiative forcing due to black carbon emissions

After 10 years of emissions assuming modest growth



Mostly due to BC

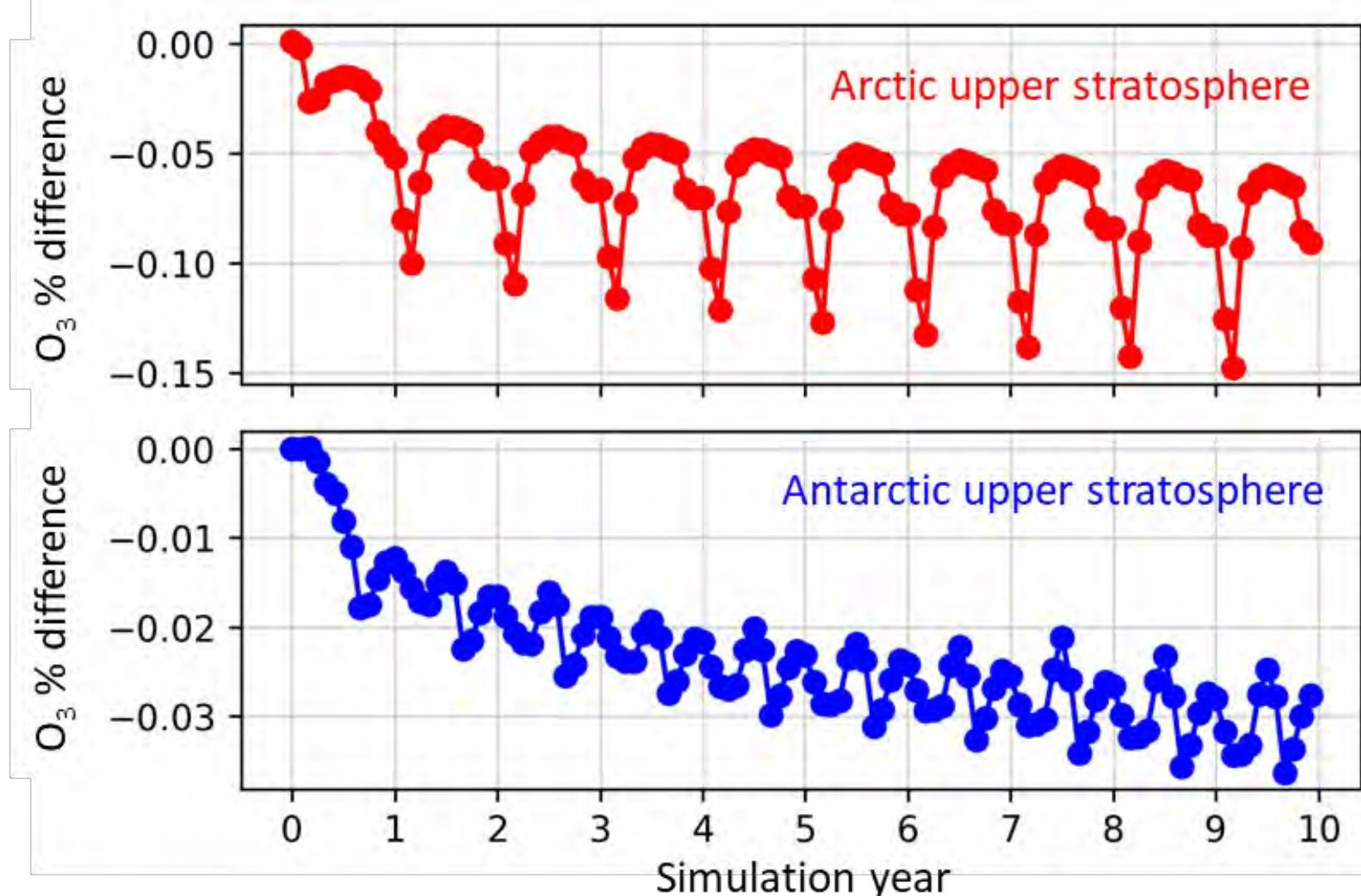


Rockets ~3% of BC radiative forcing from all anthropogenic sources, but only 0.01% of emissions.

BC from rockets **400-500 times greater radiative effect** than BC from Earth-bound sources

SpaceX Starship mission plan is 3 launches per day, so 10-fold increase in annual launches

Stratospheric ozone depletion due to 2019 rockets and re-entry



Oscillatory pattern takes
2-3 years to establish

Seasonality tracks
sunlight chemistry

50:50 contribution from
re-entry NO_x and rocket
launch chlorine

Peak decline in spring is
0.15% in the NH and
0.04% in the SH

Springtime Arctic upper stratospheric ozone depletion reaches **~0.15%** after a decade of launches
This is **~10%** of upper stratospheric ozone recovery attributed to Montreal Protocol ban on ODS

Recent and anticipated megaconstellations

SpaceX StarLink

Falcon 9



26 tonnes



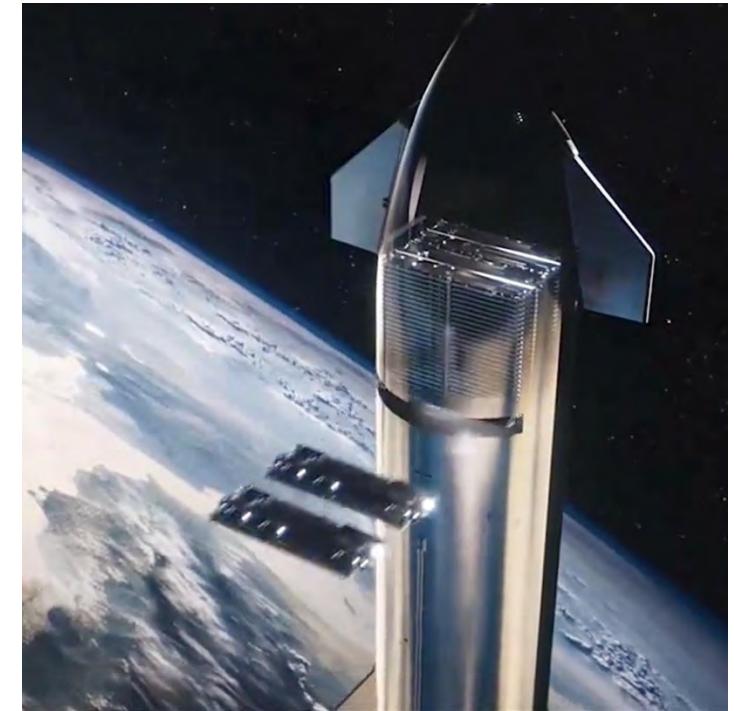
60 satellites

3,558 launched to date
318 deorbited

Raptor



~200 tonnes



Ambition is 3 launches per day
and total launch of 30,000
satellites

SpaceX StarShip

Take-homes and future work

Re-entry NO_x comparable with lower end estimate of natural NO_x from meteorites

Ozone depleting chemicals have a very local effect on upper stratospheric springtime Arctic

Positive radiative forcing due to BC of most concern. Exacerbated by anticipated growth in space sector.

Lots to do on this topic! Account for re-entry emissions of metal oxides, use the current observing system to detect signals associated with launch and re-entry emissions.

Link to paper: <https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2021EF002612>

Media coverage by BBC, Times, Forbes, MSN, Sky and many more.