

# Bridging knowledge gaps in atmospheric science:

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



UCL Atmospheric Composition and Air Quality Research Group

<https://maraisresearchgroup.co.uk/>



# Fast-growing tropical megacities



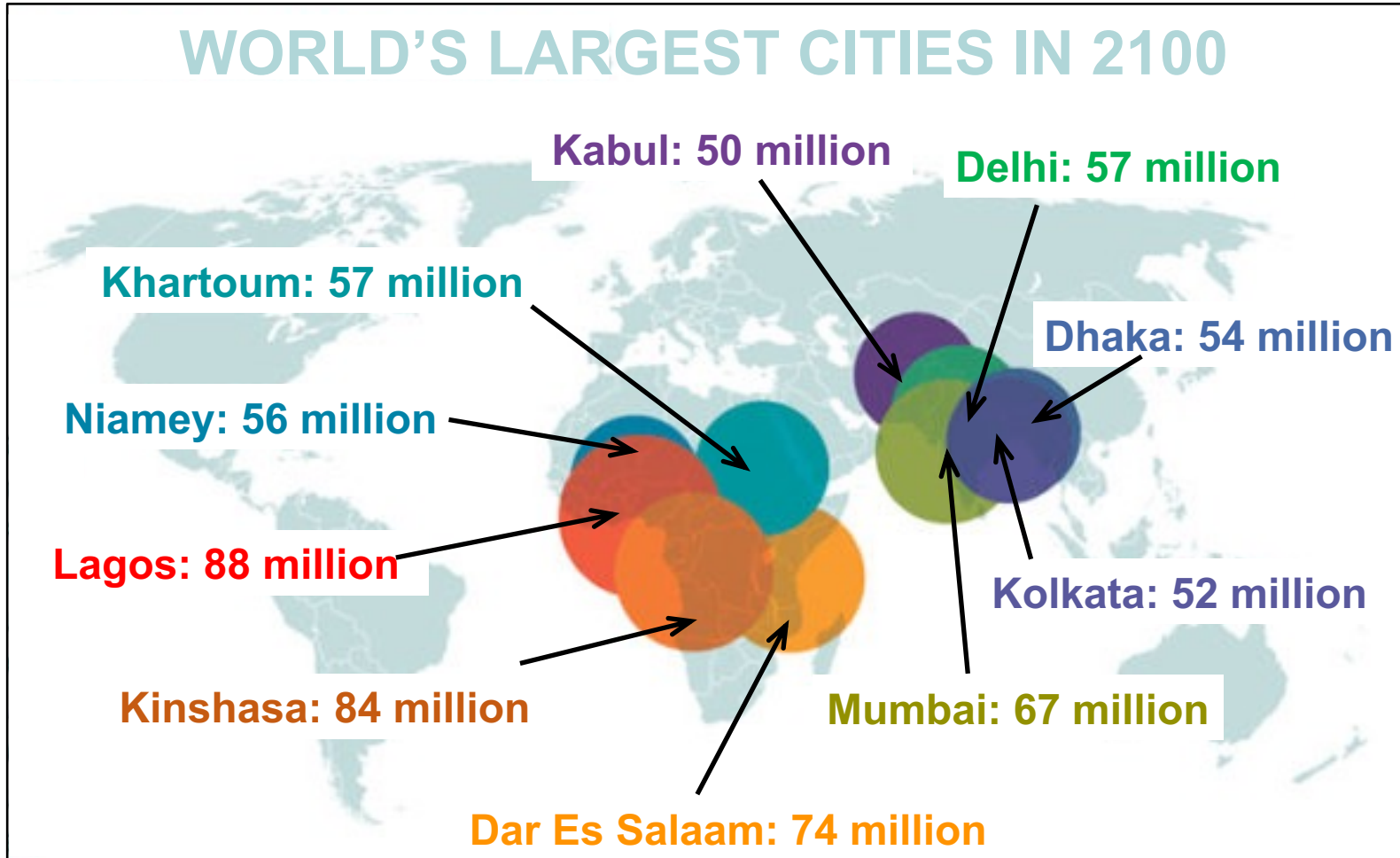
**Karn Vohra**  
postdoc



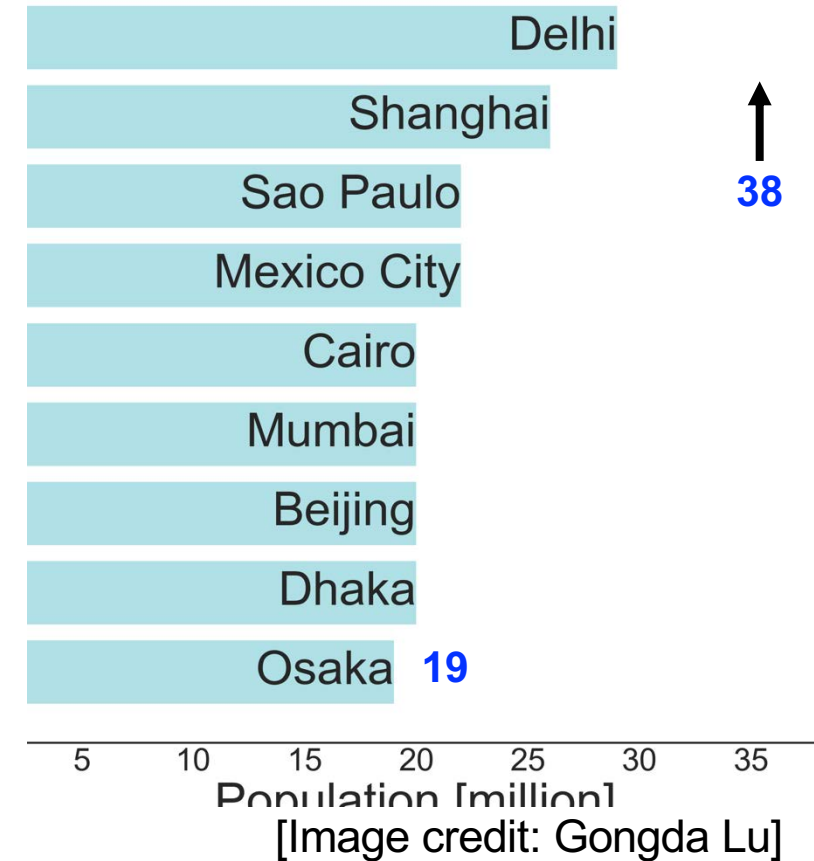
# The largest future megacities are all in the tropics

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

## WORLD'S LARGEST CITIES IN 2100



## Largest cities in 2020

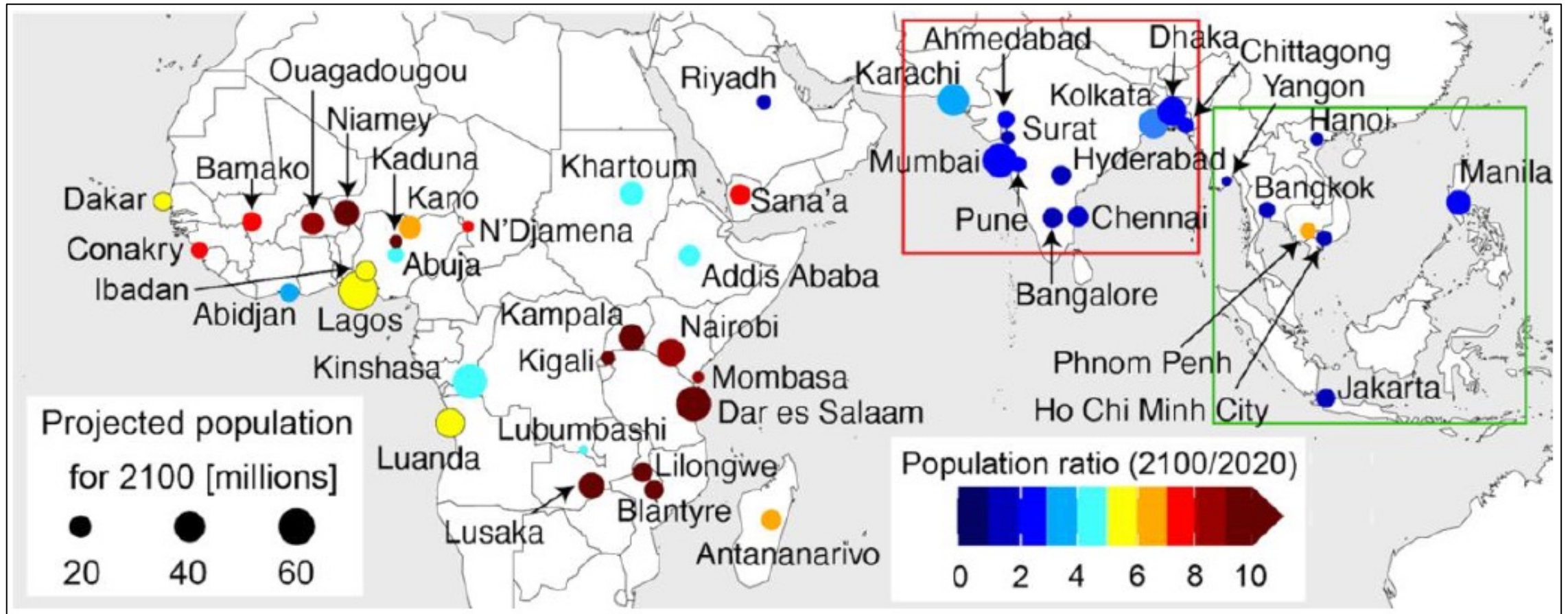


Adapted image: <https://medium.com/ensia/here-come-the-megacities-1b0f8a2287f2>

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]:

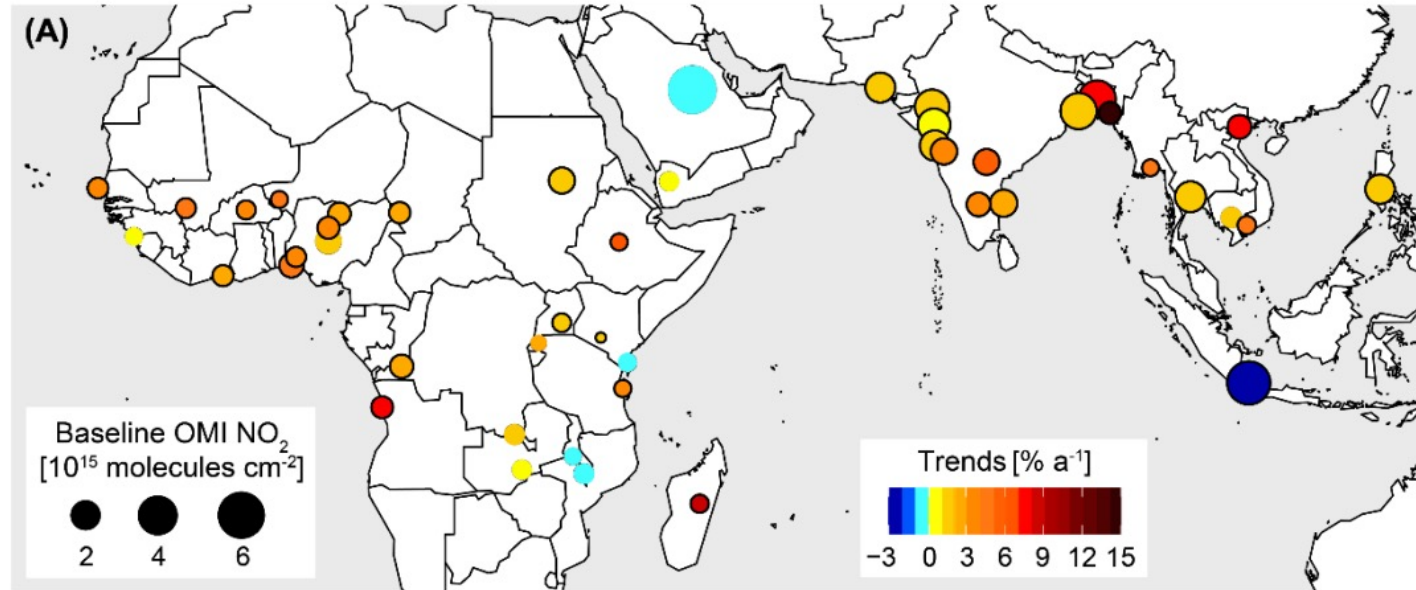
3-31% for Africa, 0.8-3% for **South Asia**, 0.5-7% for **Southeast Asia**



# Steep annual increases in $\text{NO}_x$ and $\text{NH}_3$

$\text{NO}_2$  trends  
(proxy for  $\text{NO}_x$ )  
[2005-2018]

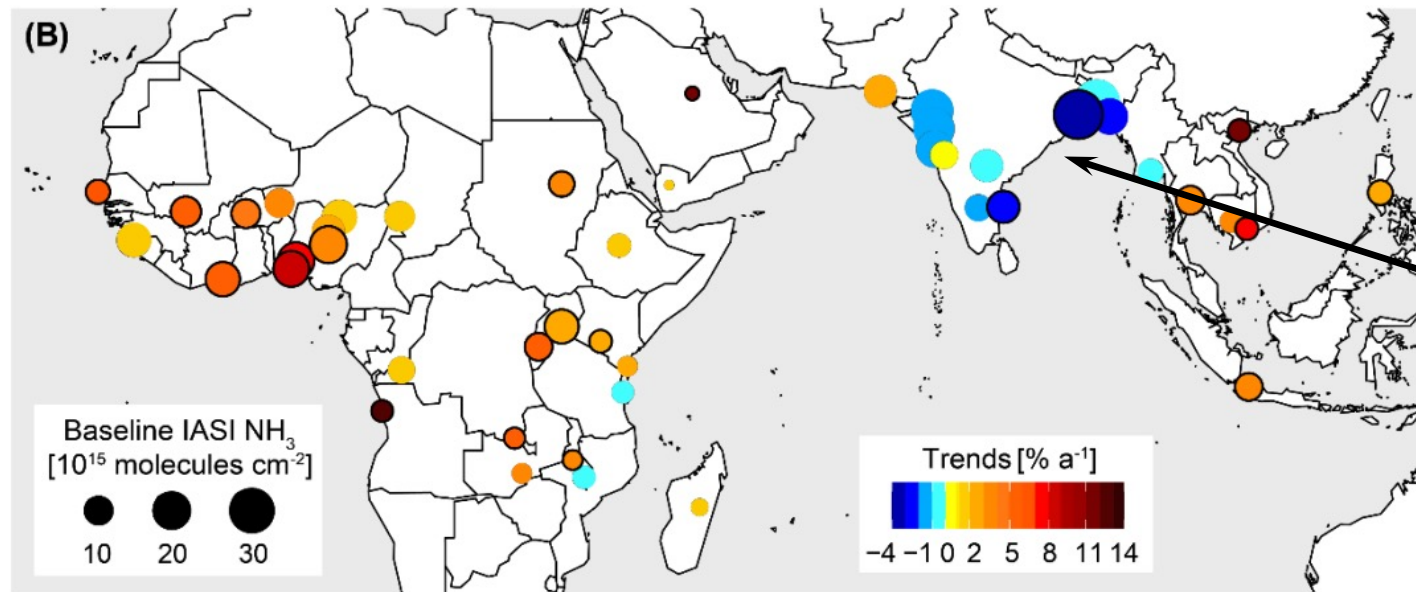
OMI: Ozone  
Monitoring  
Instrument



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

$\text{NH}_3$  trends  
(depends on acidic  
aerosol abundance)  
[2008-2018]

IASI: Infrared  
atmospheric  
sounding  
interferometer



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

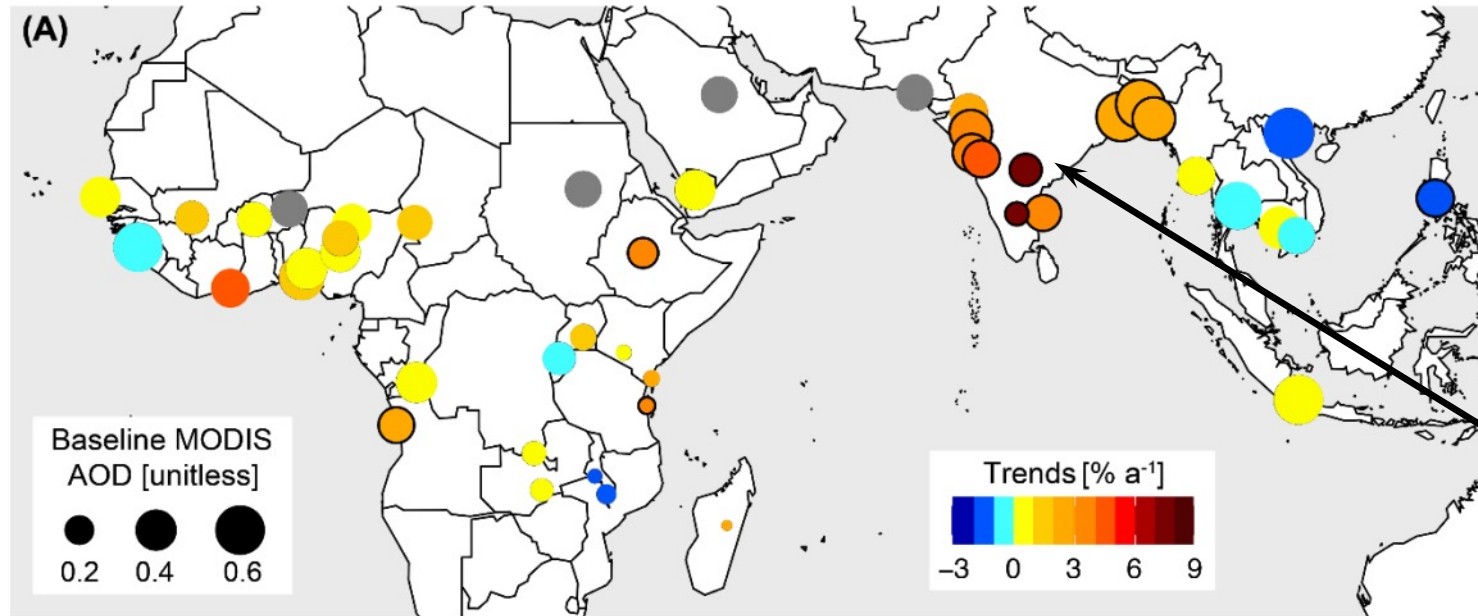
$\text{NH}_3$  data from M. Van Damme, L. Clarisse, P.-F. Coheur at ULB



# Annual changes in PM<sub>2.5</sub> and ozone production regimes

**AOD trends**  
(proxy for PM<sub>2.5</sub>)  
[2005-2018]

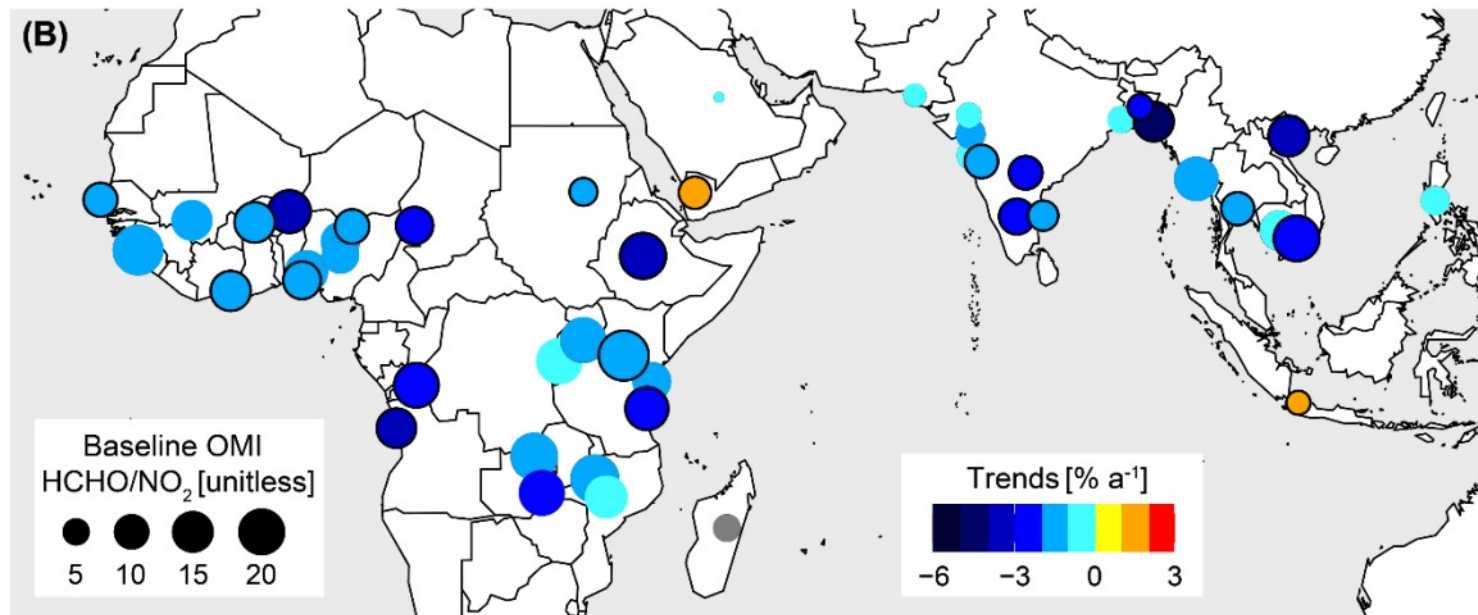
**MODIS:** Moderate  
resolution imaging  
spectroradiometer



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

Increases in PM<sub>2.5</sub>  
precursors SO<sub>2</sub>,  
NH<sub>3</sub>, NO<sub>x</sub>

**HCHO/NO<sub>2</sub> trends**  
(proxy for ozone  
production regime)  
[2005-2018]



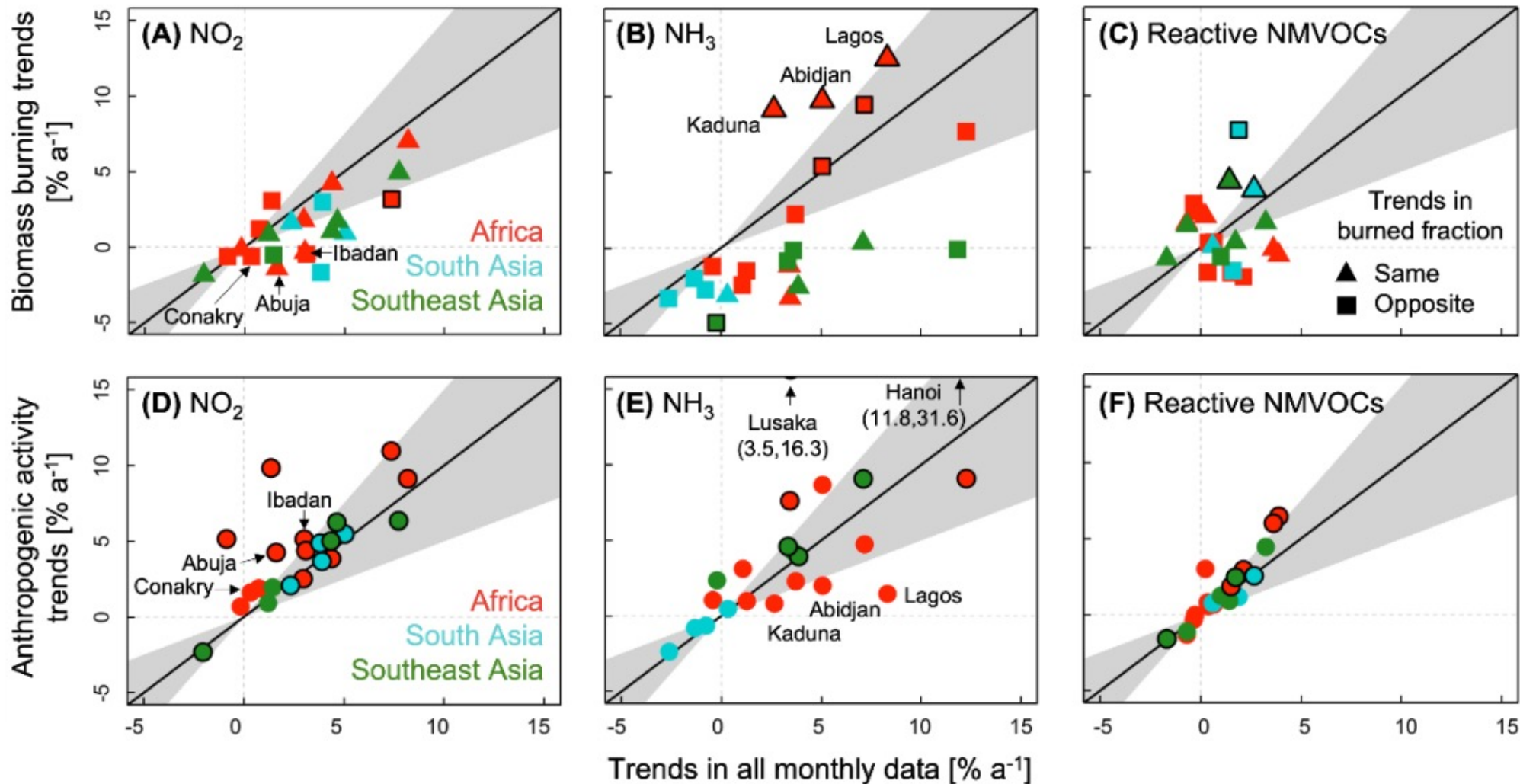
**Ratio > 5:**  
O<sub>3</sub> production  
sensitive to NO<sub>x</sub>

Transitioning to NO<sub>x</sub>  
saturated or VOC  
sensitive



# What's driving the observed trends?

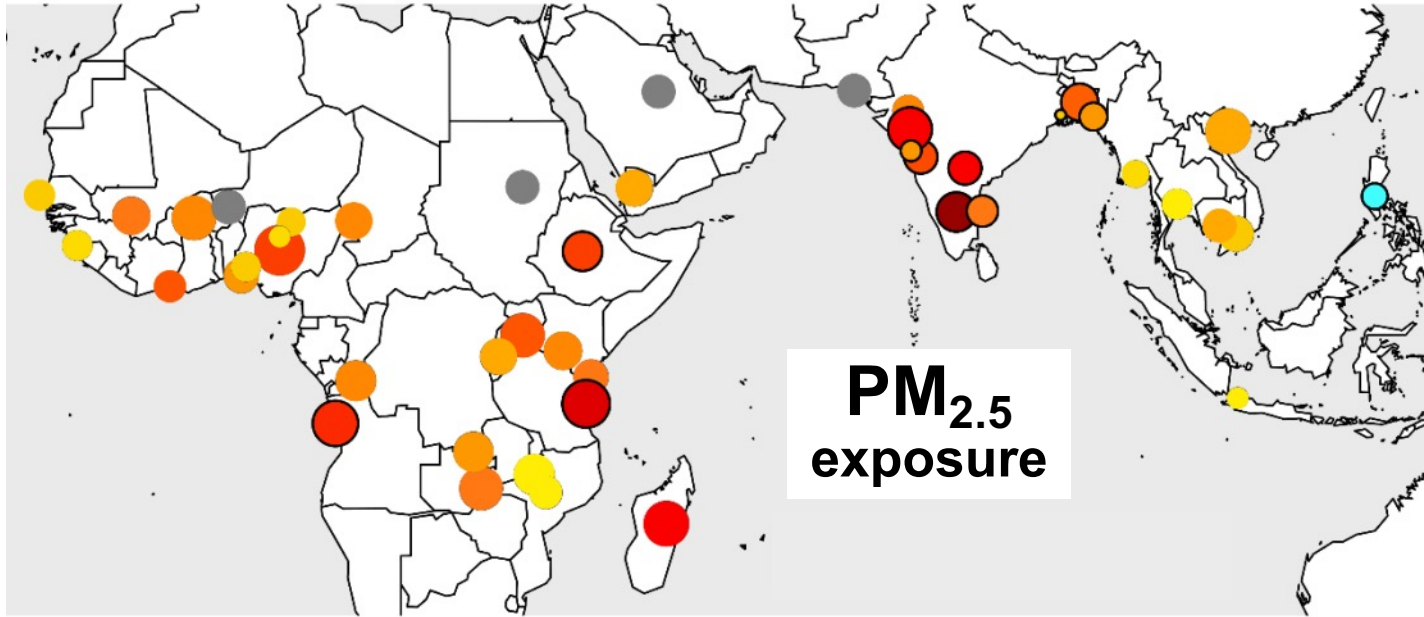
We use a statistical approach and knowledge of seasonality of emissions to assess the relative role of anthropogenic and biomass burning emission



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



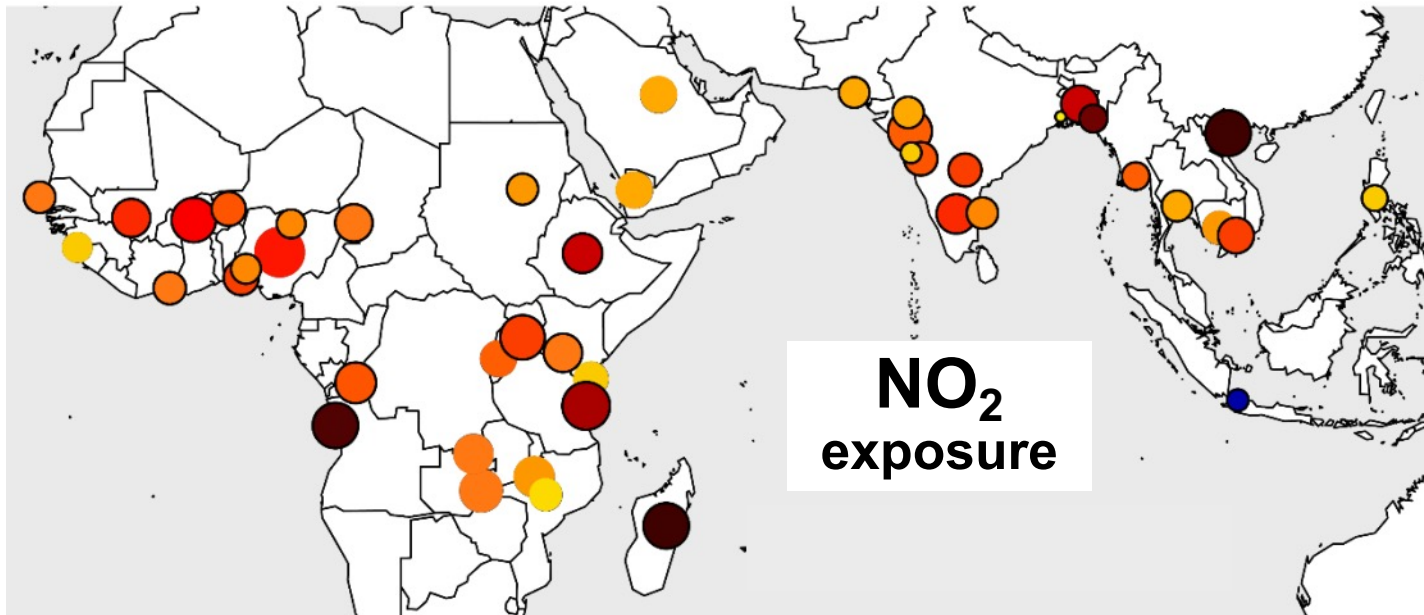
# Increase in urban population exposure to air pollution



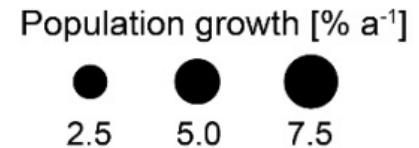
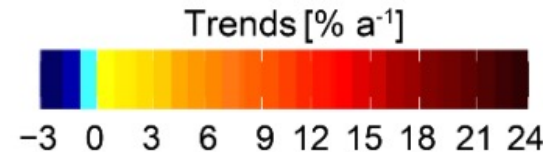
Combined effect of rapid air quality degradation, increase in population and urbanization

Up to **18 % a<sup>-1</sup>** increase in PM<sub>2.5</sub> in India

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

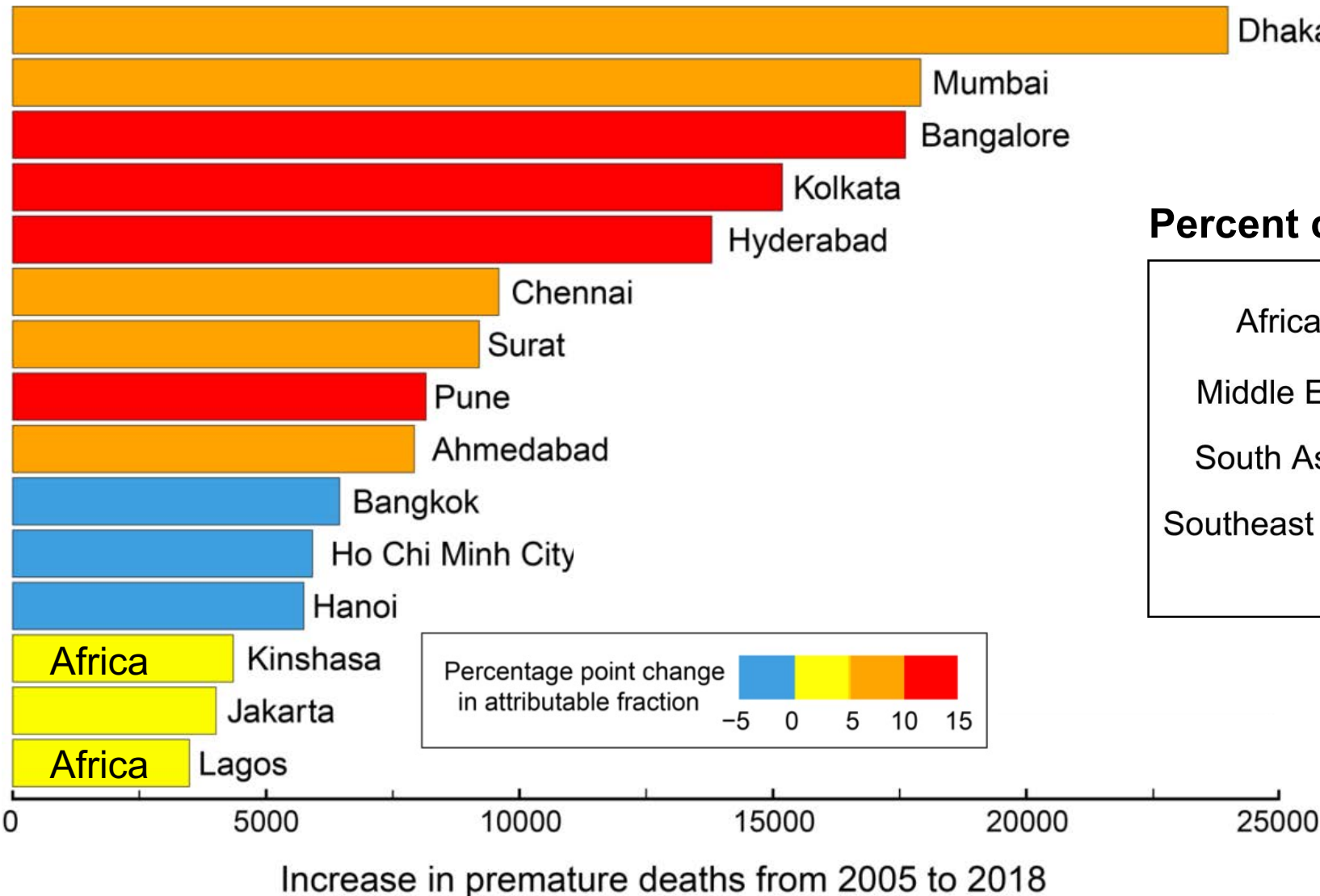


Up to **23% a<sup>-1</sup>** increase in NO<sub>2</sub> in many cities

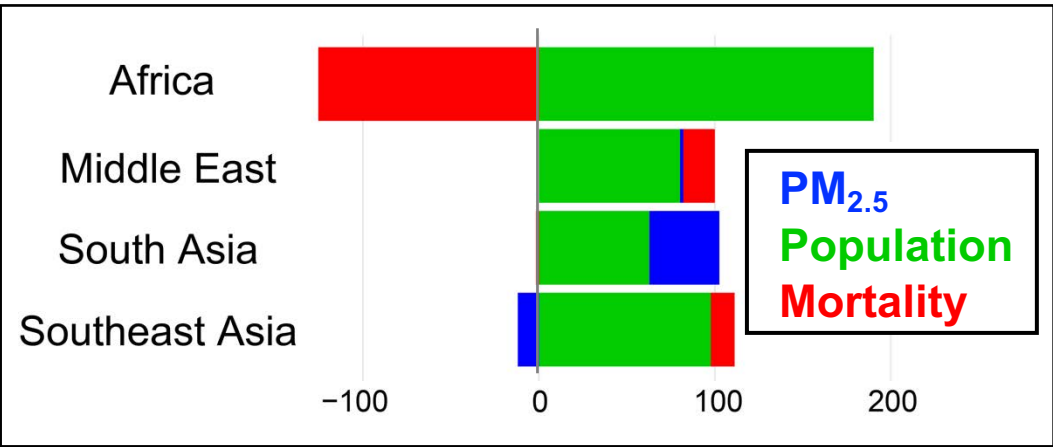


# Premature mortality attributable to rise in PM<sub>2.5</sub> exposure

## 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<sub>x</sub> 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



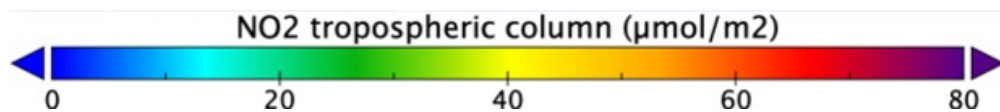
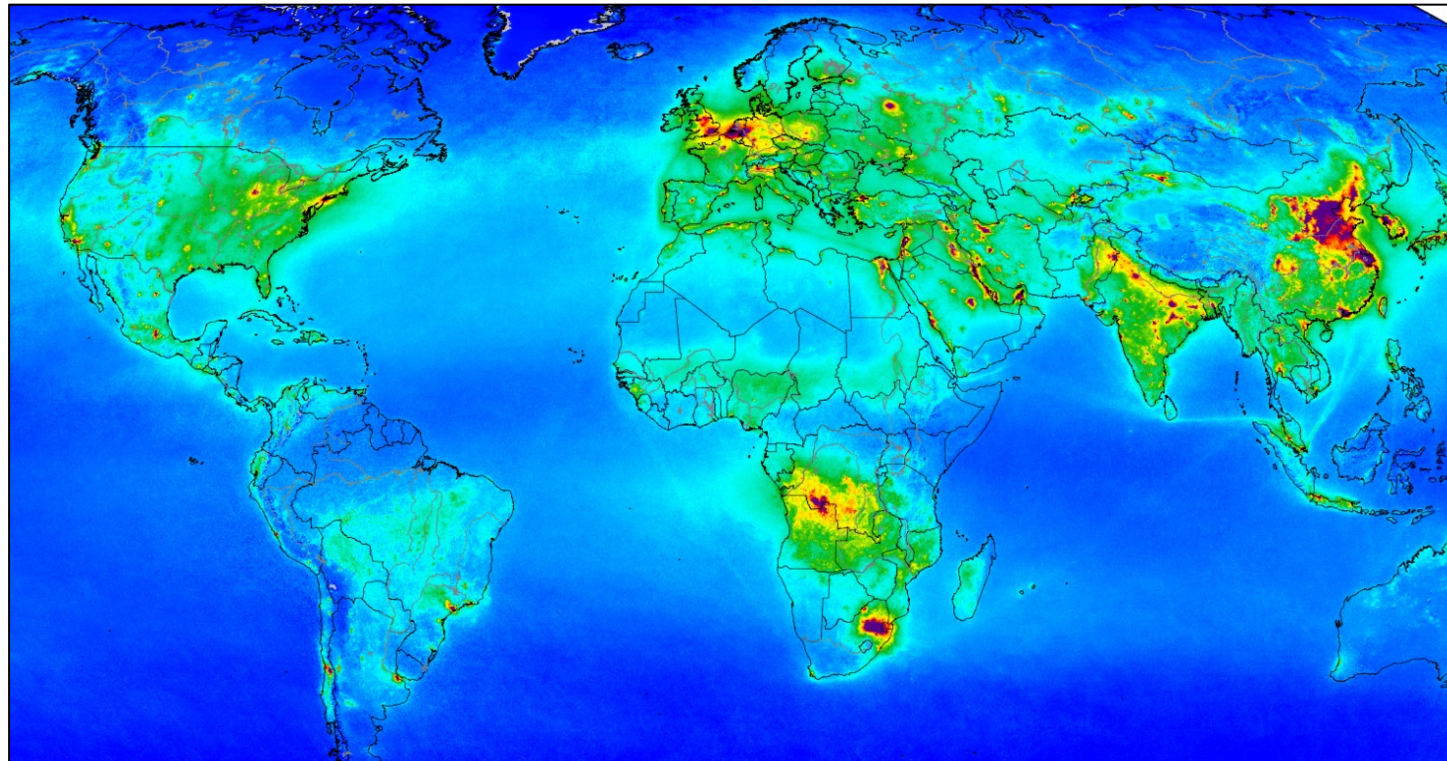
**Eleanor Smith**  
PhD



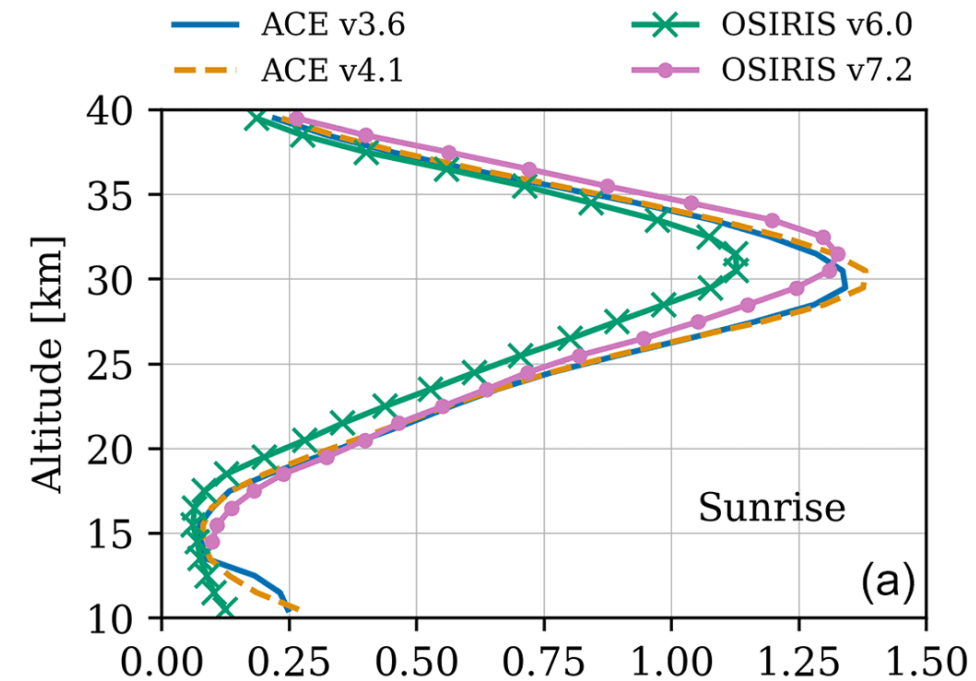
# Reactive nitrogen in the remote troposphere

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

Nadir-viewing instruments observe the whole column



Limb-viewing instruments not sensitive to troposphere

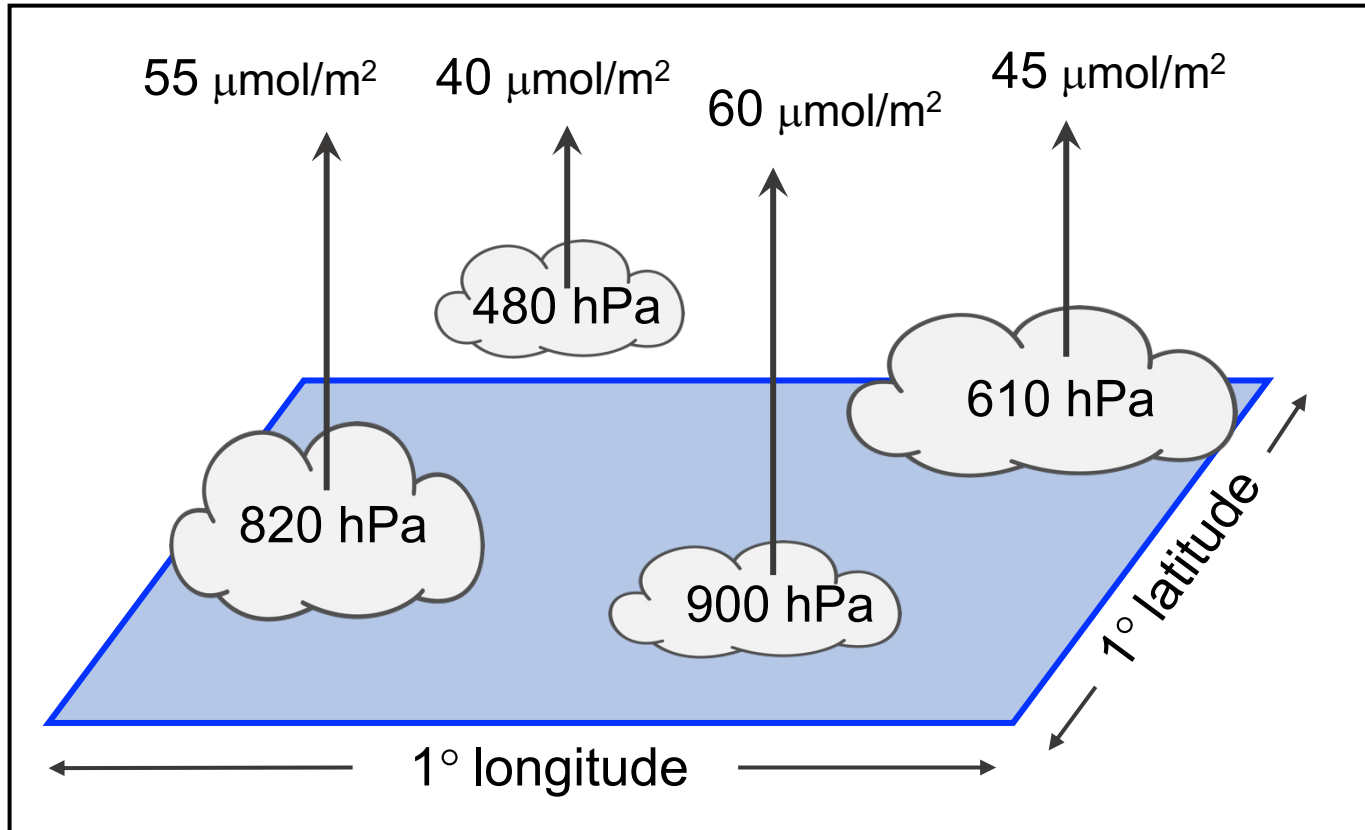


[Dubé et al., 2022]

Aircraft observations limited in space and time

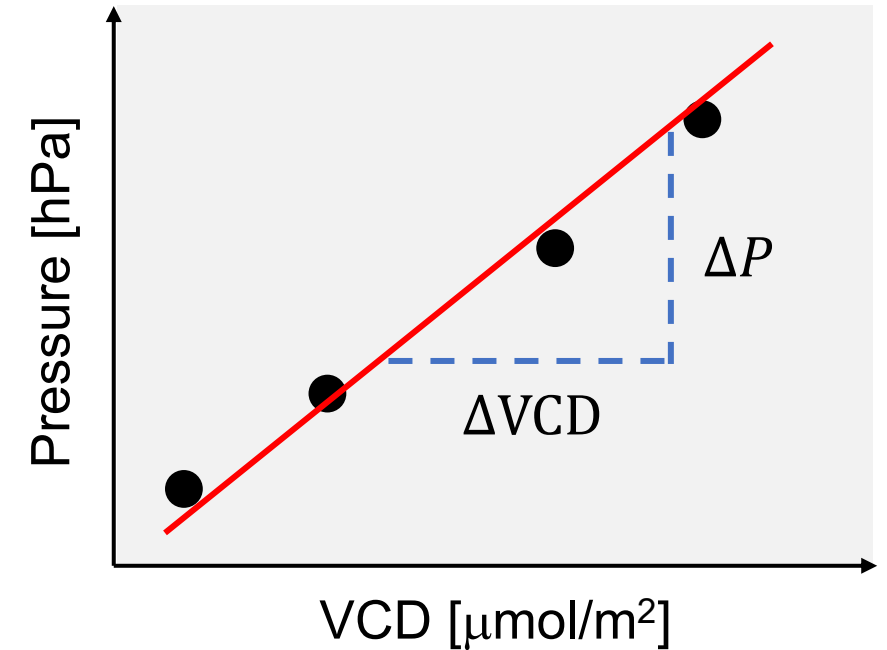
# Cloud-slicing satellite observations to address data scarcity

Clusters of partial columns above optically thick clouds:



Calculate average mixing ratio between target pressure ranges:

Regress cloud top pressures against partial vertical column densities (VCDs):



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



# Application to high-resolution TROPOMI instrument



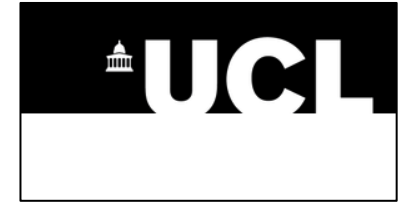
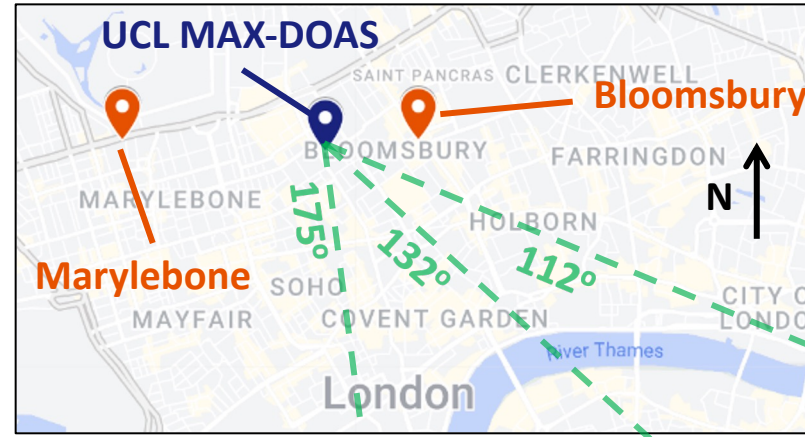
UV/vis spectrometer

13h30 overpass time

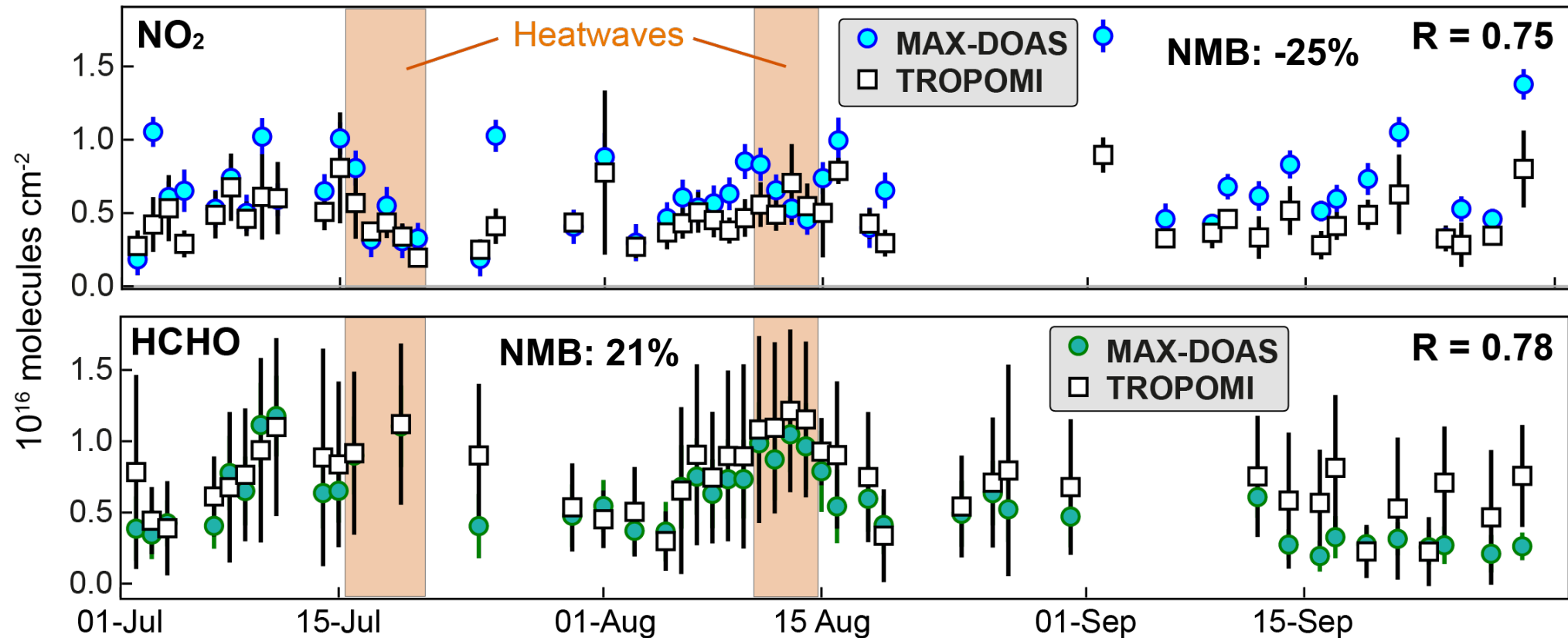
Daily global coverage

5.6 km × 3.5 km resolution

# TROPOMI validation with MAX-DOAS during 2022 heatwaves



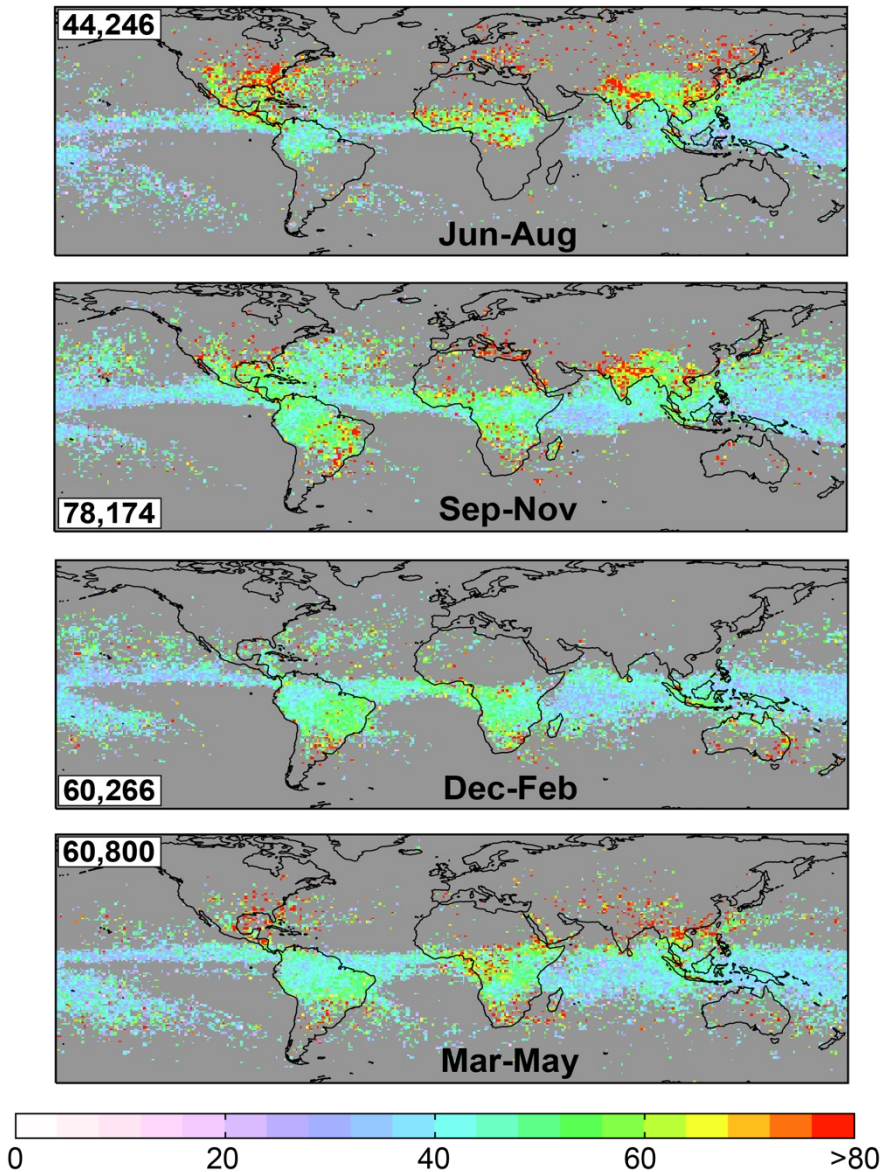
Capital Equipment  
Fund



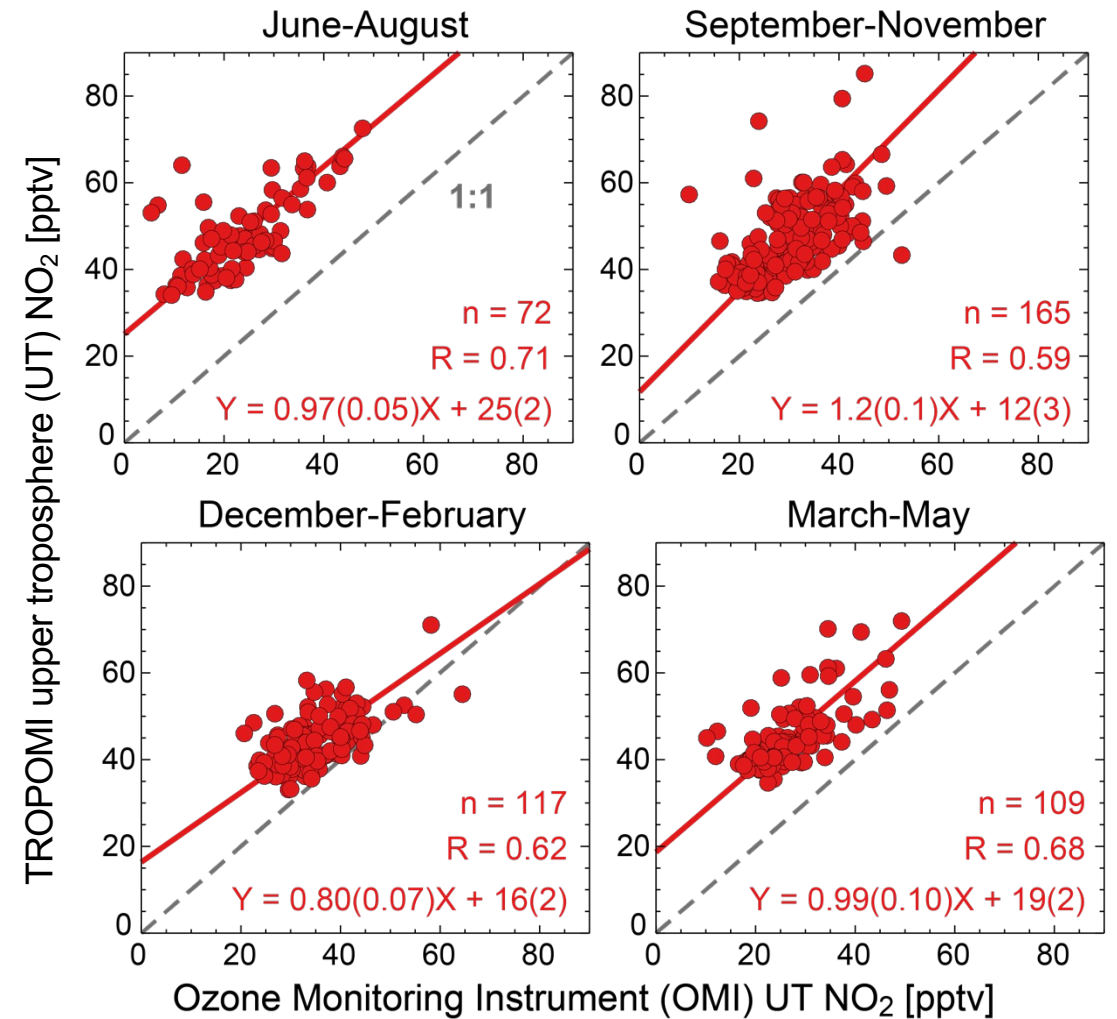


# Seasonal means of NO<sub>2</sub> in the upper troposphere

Seasonal means at 8-12 km



Evaluation against product from OMI



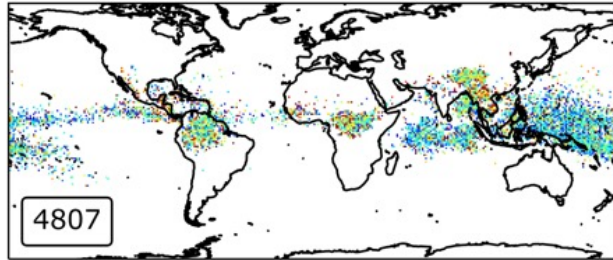
[Marais et al.,  
AMT, 2021]

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

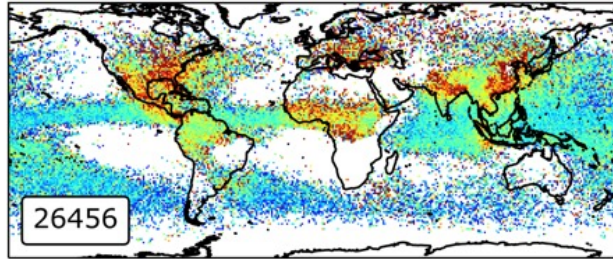
# Vertical profiles of NO<sub>2</sub> derived with the TROPOMI instrument

## Vertical profiles in Jun-Aug

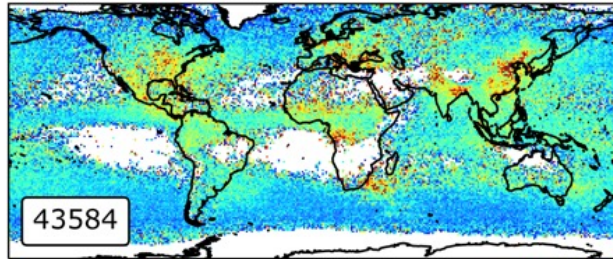
180-320 hPa  
9-12 km



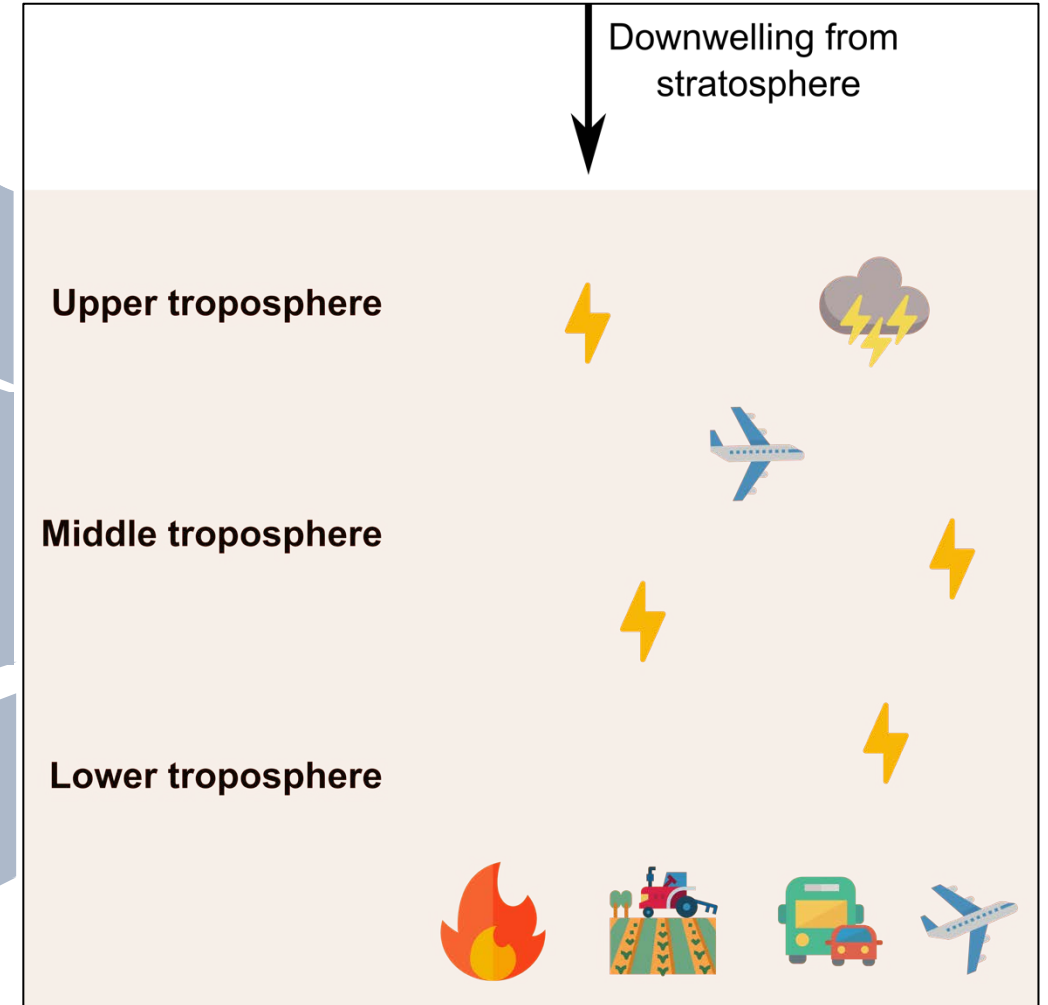
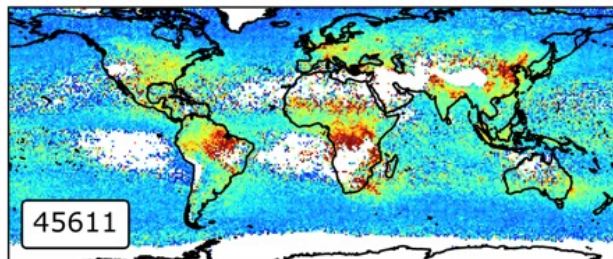
320-450 hPa  
6-9 km



450-600 hPa  
4-6 km



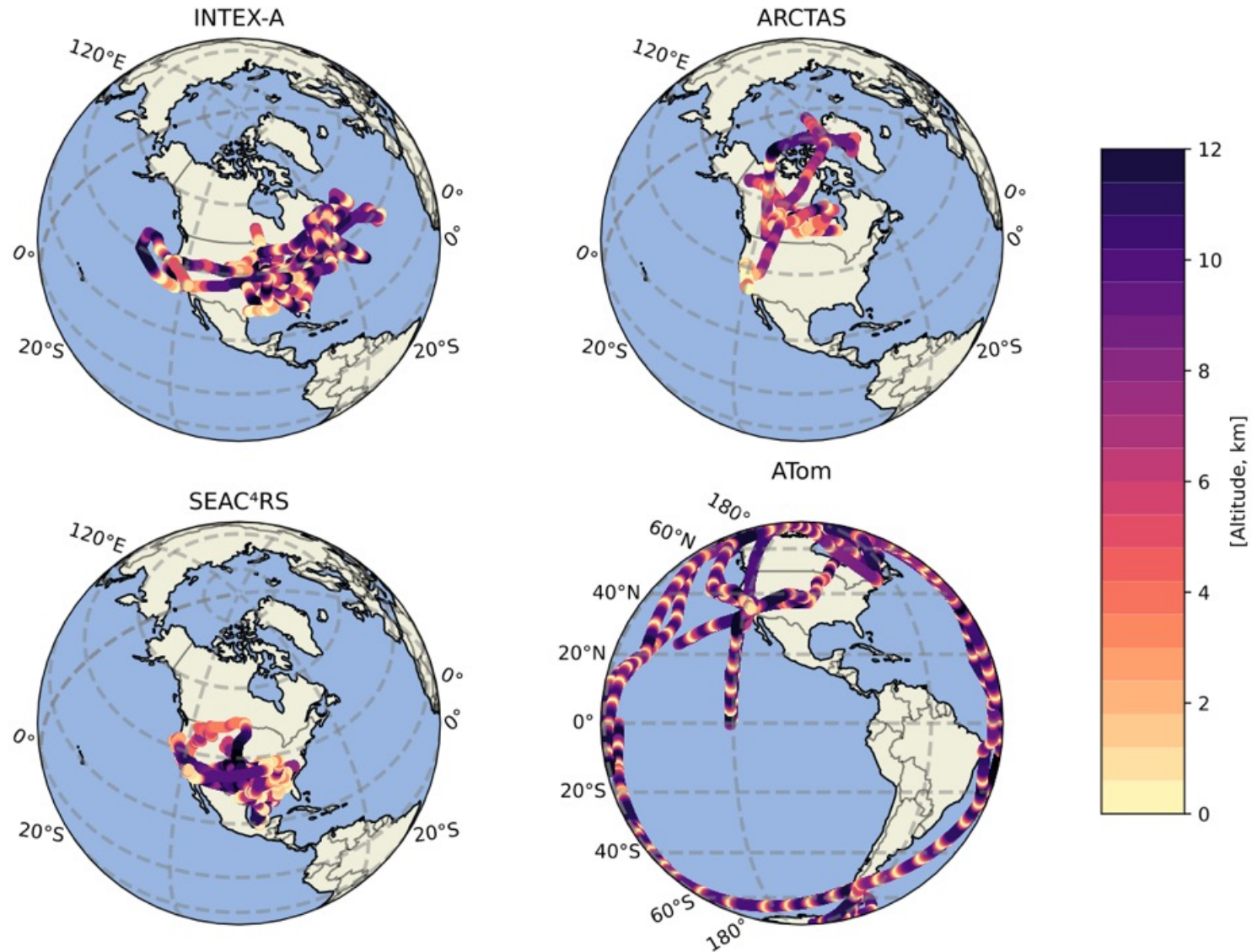
600-800 hPa  
2-4 km



[Horner et al.,  
in prep]



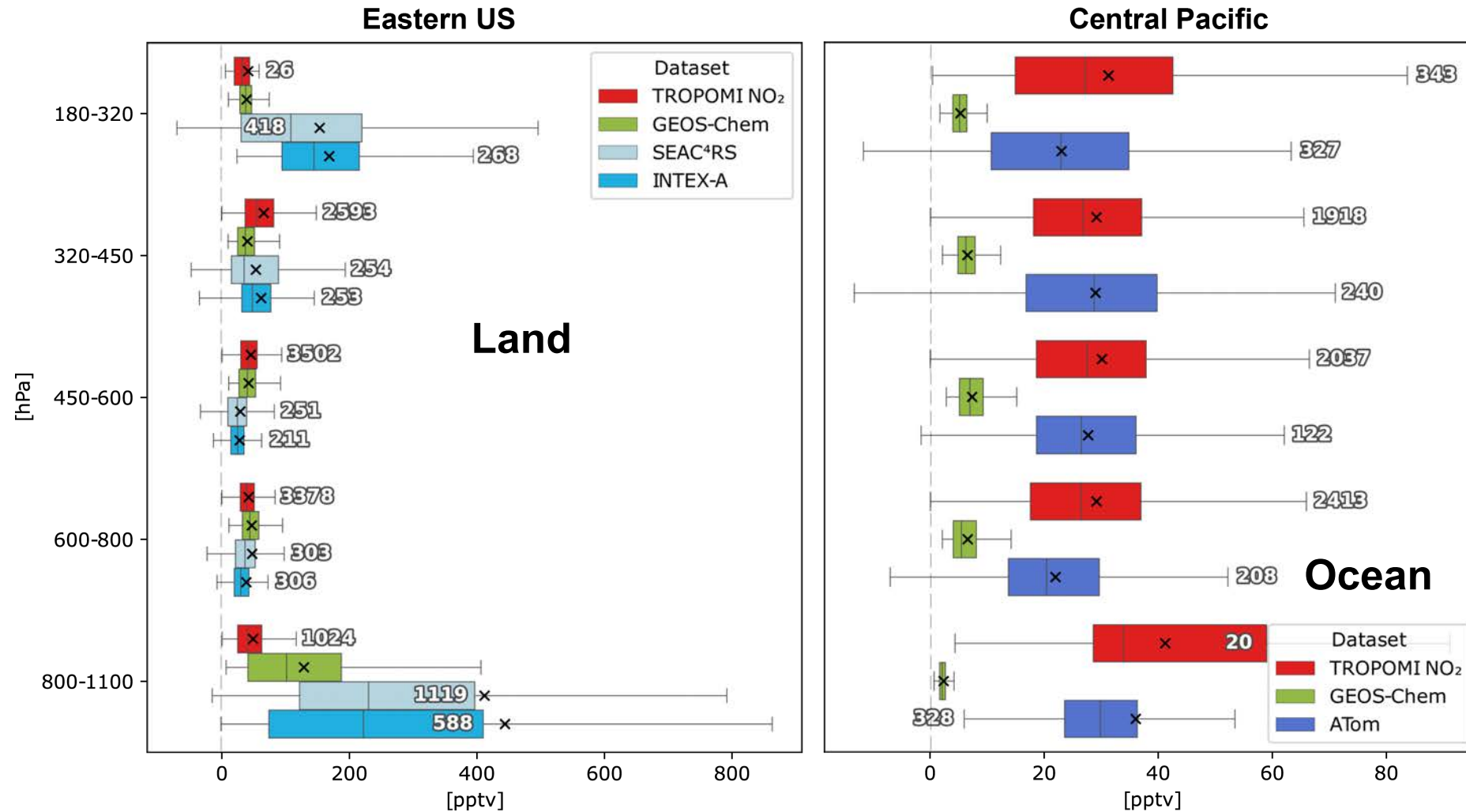
# Cloud-sliced product validation with aircraft observations



Data provided by NASA DC8 Science Teams

# Cloud-sliced product validation with aircraft observations

Comparison of collocated NO<sub>2</sub> mixing ratios in June-August

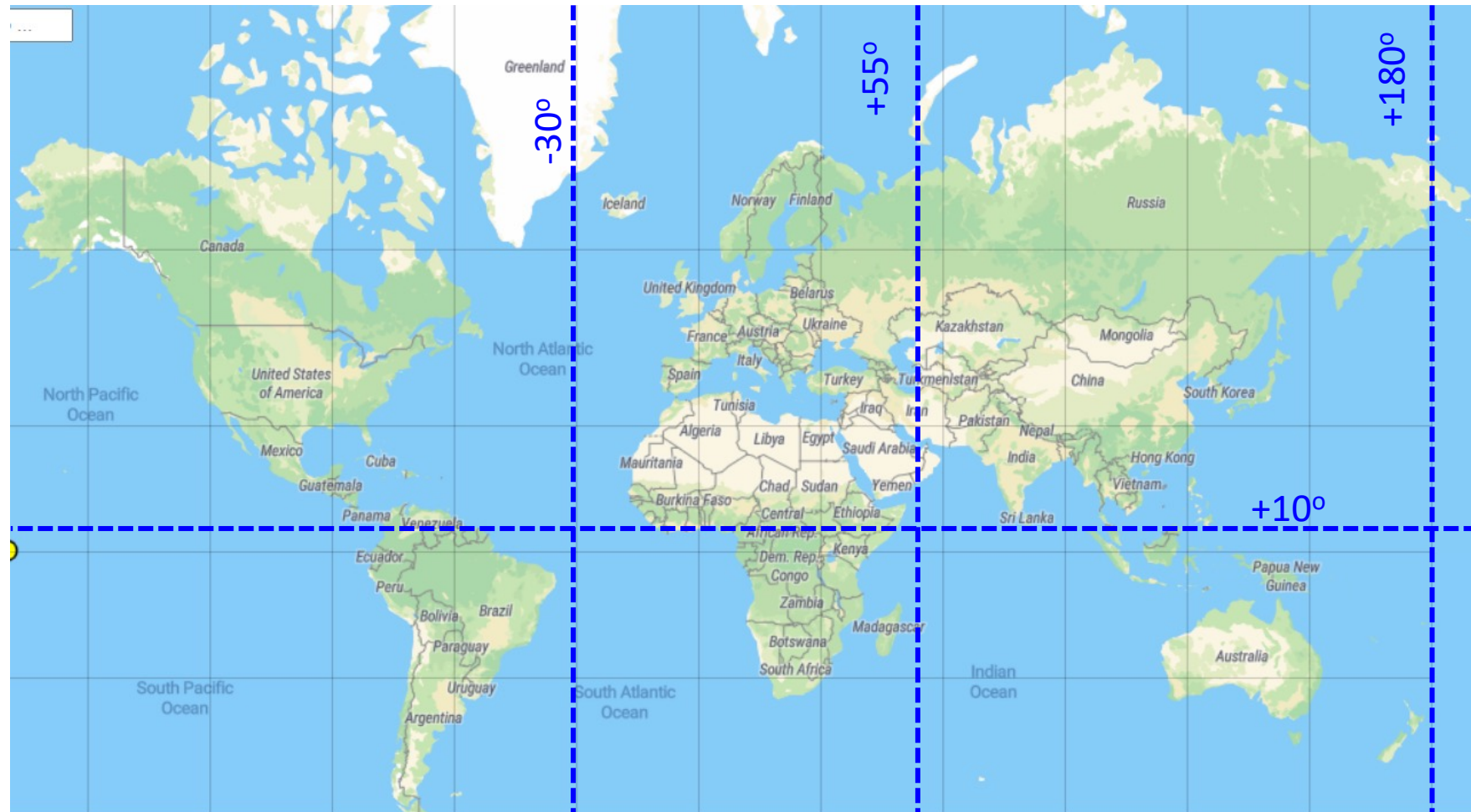


Aircraft and TROPOMI consistent (20-30 pptv) in the mid troposphere

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

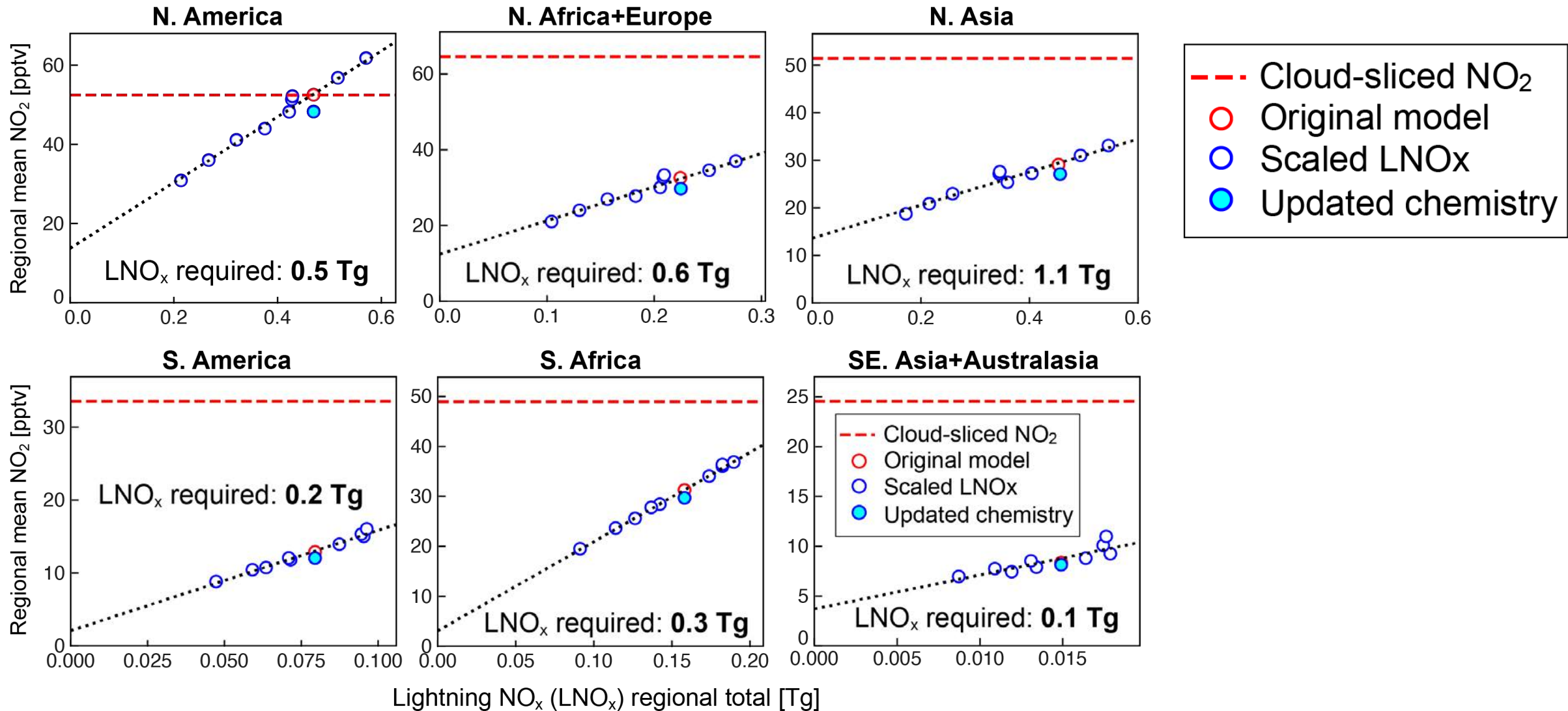


# Regional sensitivity to lightning NO<sub>x</sub> emissions



# Regional sensitivity to lightning $\text{NO}_x$ emissions

Observed versus modelled June-August upper tropospheric regional mean  $\text{NO}_2$  over land

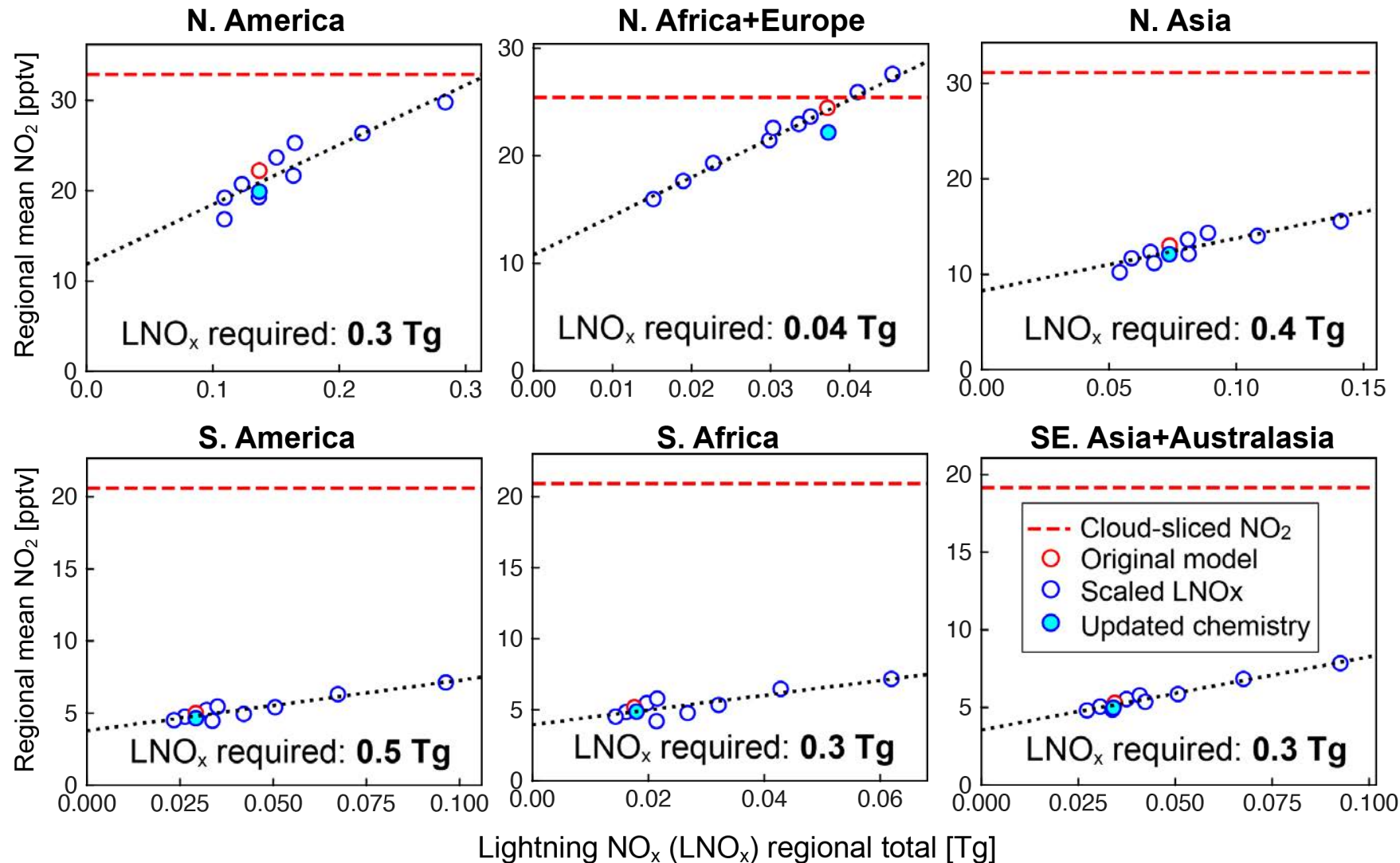


Original model emissions of **1.4 Tg NO**, whereas **2.7 Tg NO** required to match cloud-sliced  $\text{NO}_2$



# Regional sensitivity to lightning $\text{NO}_x$ emissions

Observed versus modelled June-August upper tropospheric regional mean  $\text{NO}_2$  over the ocean

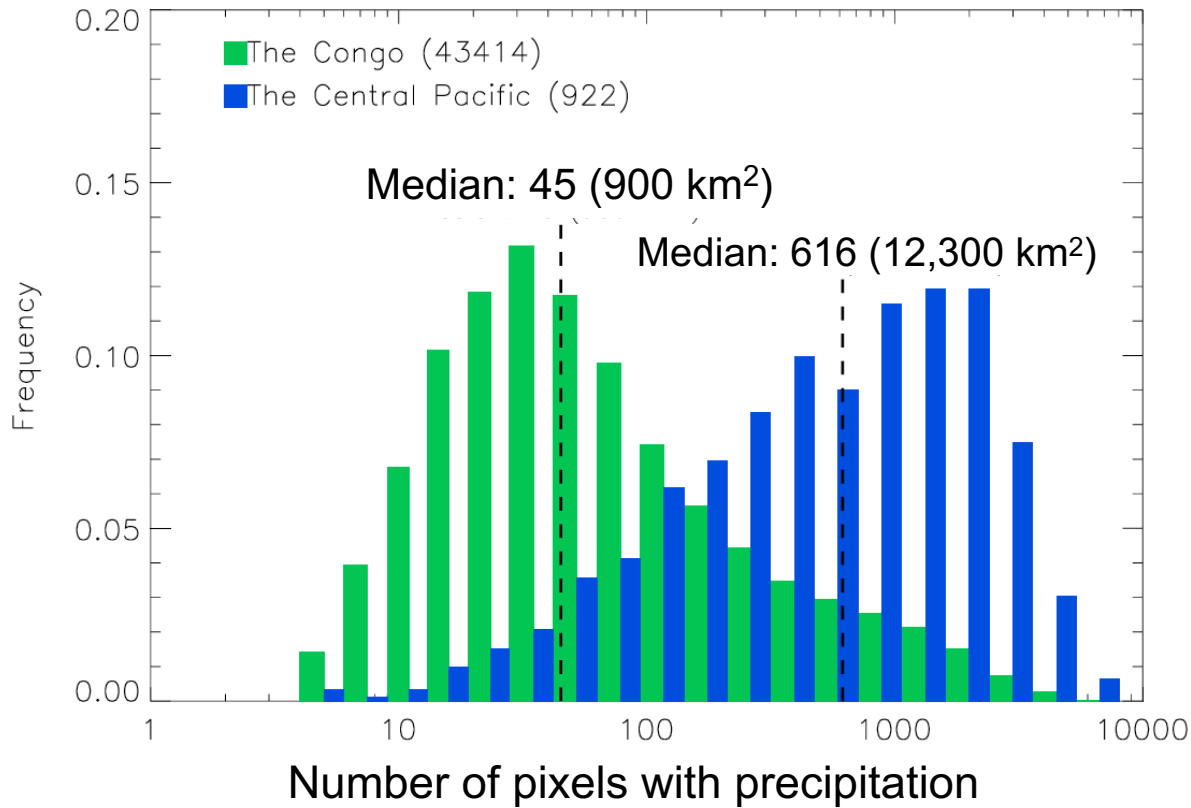


Original model emissions of **0.3 Tg NO**, whereas **1.9 Tg NO** required to match cloud-sliced  $\text{NO}_2$

# Lightning characteristics over the ocean and over land

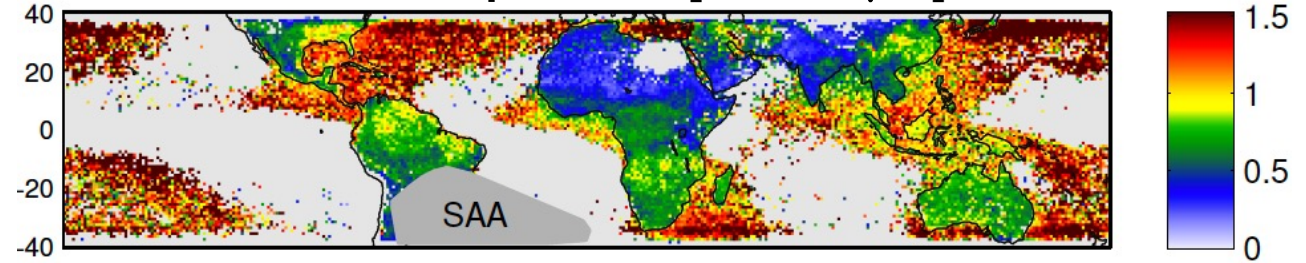
Support for larger, more persistent and higher energy lightning flashes over the ocean than over land

Frequency distribution for all radar precipitation features with lightning

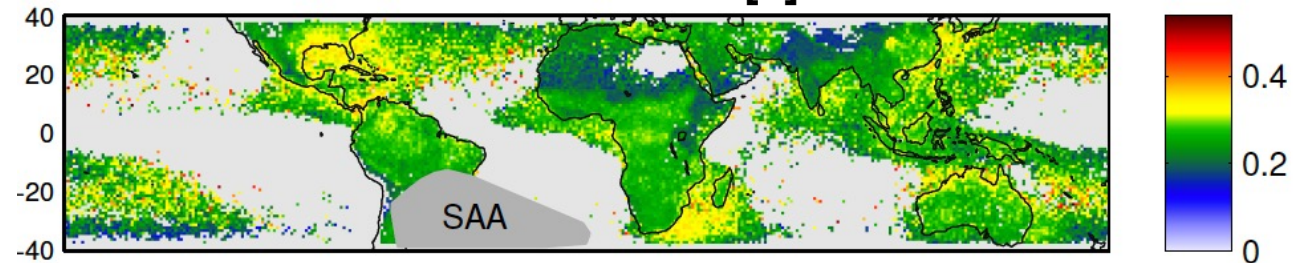


[Bang & Zipser, 2015]

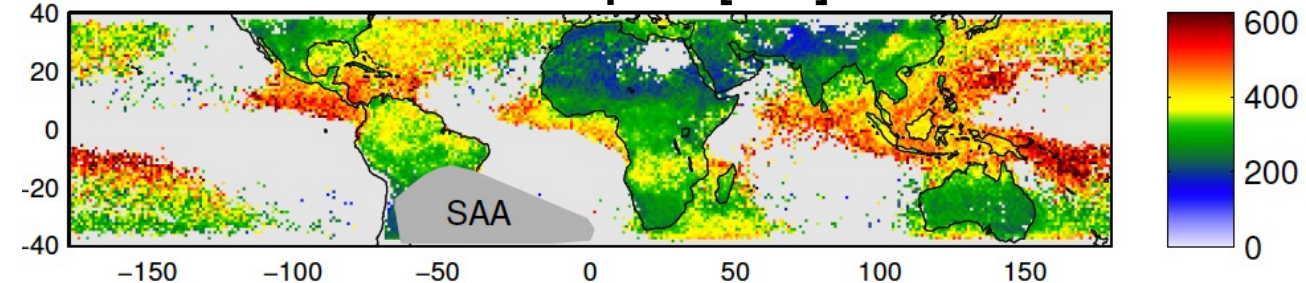
Radiance per flash [ $\text{J/m}^2/\text{sr}/\mu\text{m}$ ]



Flash duration [s]



Flash footprint [ $\text{km}^2$ ]



[Beirle et al., 2014]



# Concluding Remarks

Cloud-sliced profiles of  $\text{NO}_2$  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  $\text{NO}_2$  sensitive to lightning  $\text{NO}_x$  emissions

Addressing the model bias requires almost 3-fold increase in global lightning  $\text{NO}_x$  emissions with implications for tropospheric ozone production

# Environmental impact of the modern space sector



**Chloe Balhatchet**  
summer student

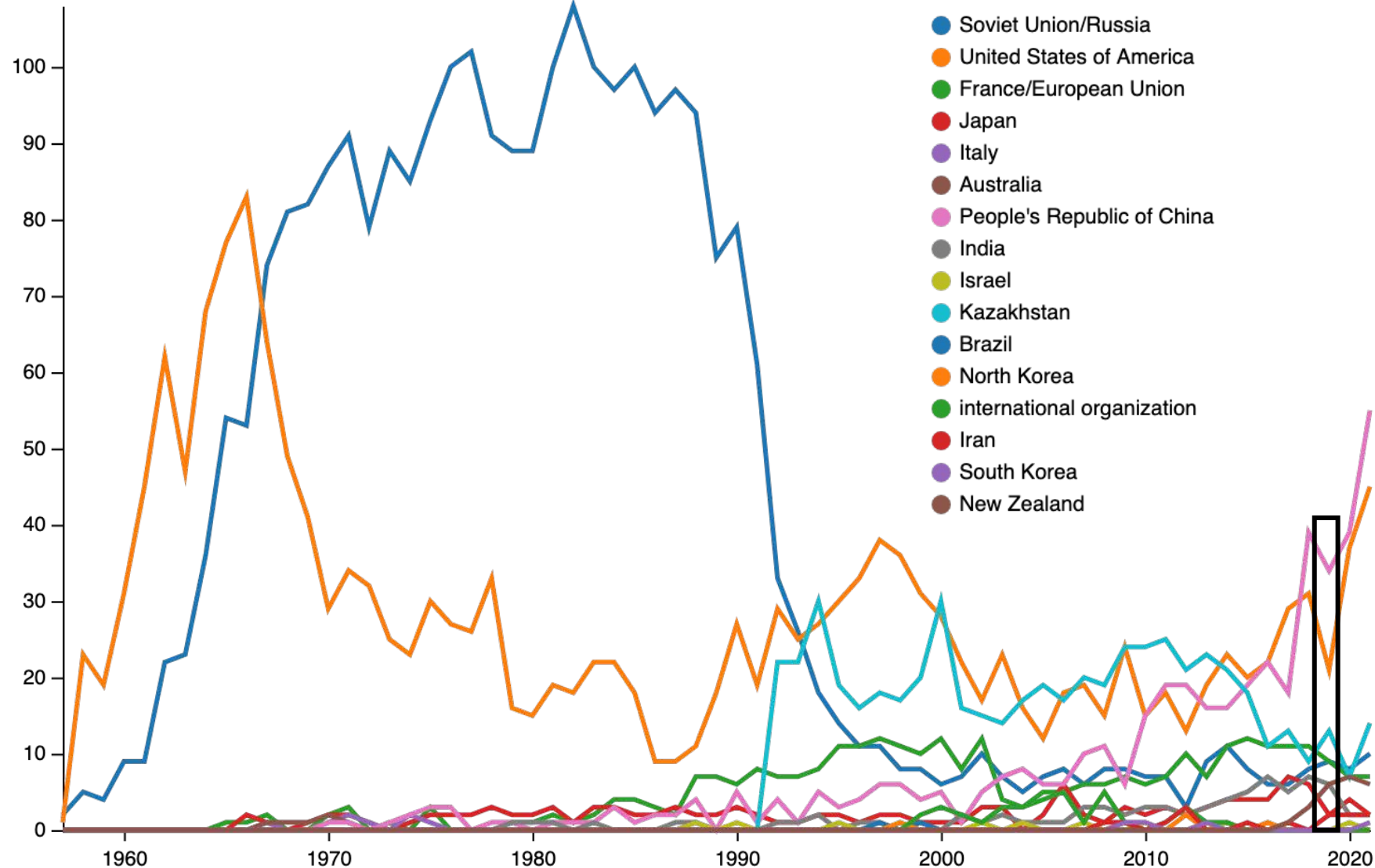


**Rob Ryan**  
postdoc

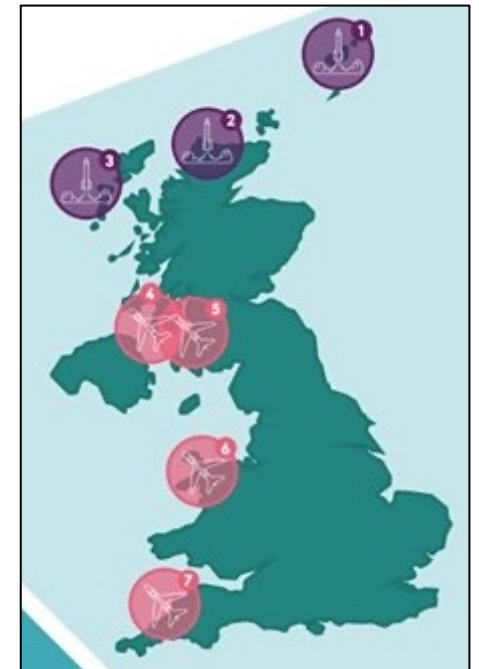


# More diverse space sector than the original space race

Number of rocket launches per country in each year



Even the UK is joining the race:



# Dramatic increase in objects in space

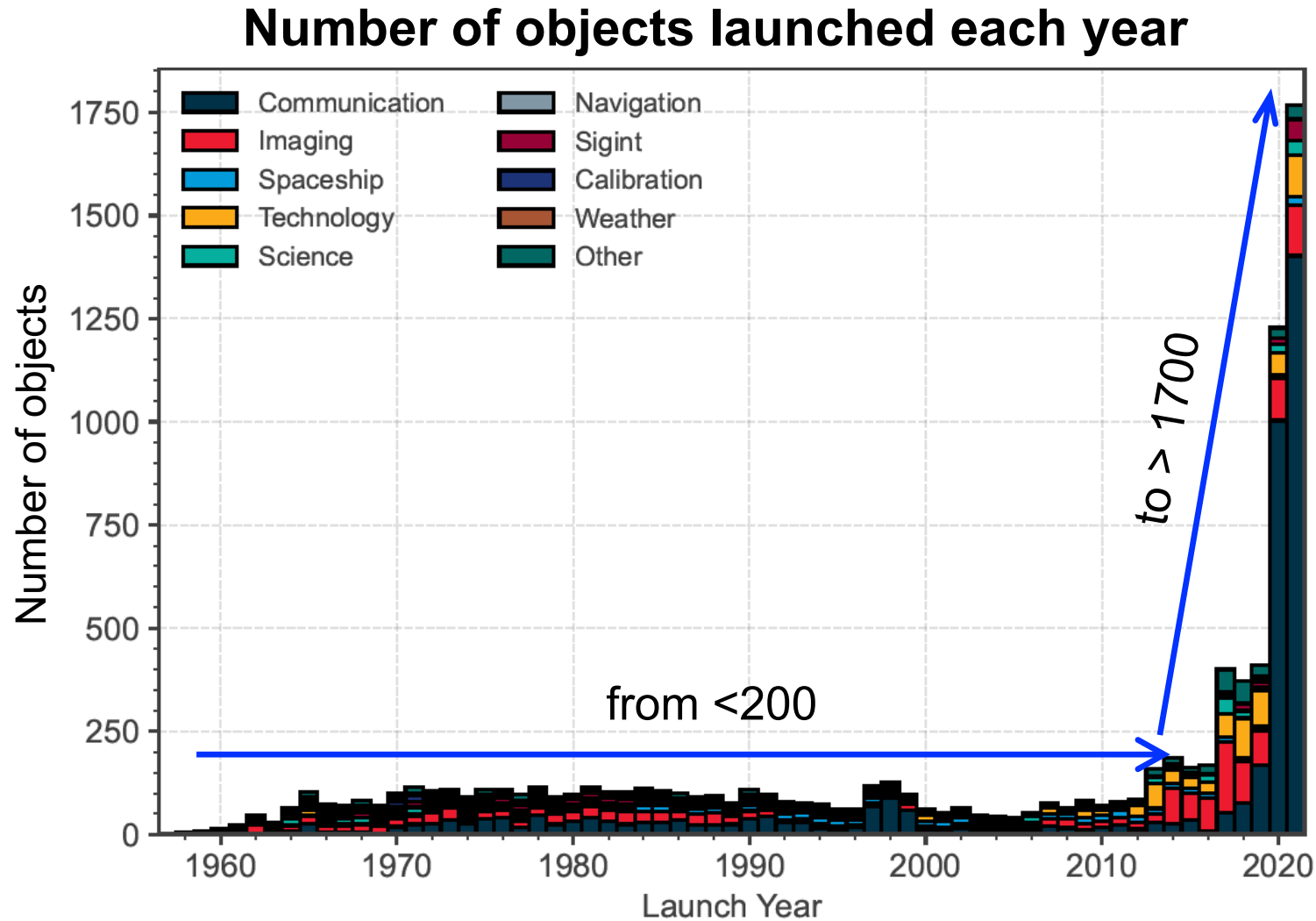


Image from ESA's Annual Space Environment Report, 2022

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



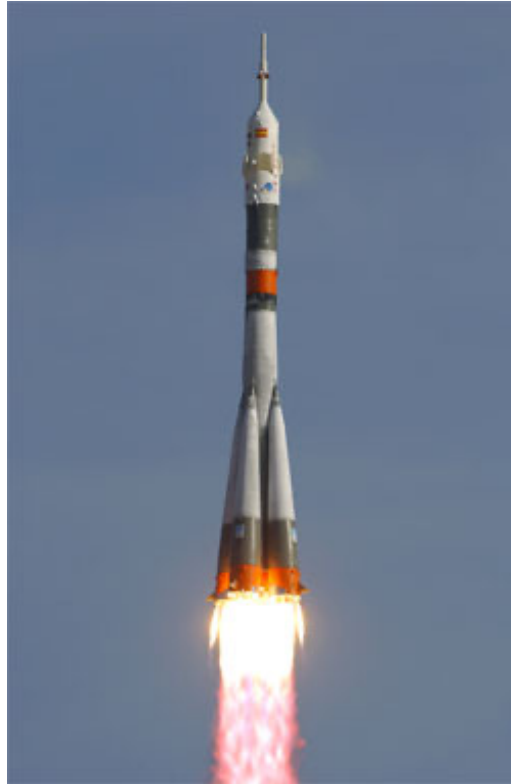
# Air pollutant emissions from rocket launches

## Solid



$\text{NO}_x$   
 $\text{HCl} + \text{Cl}$   
 $\text{Al}_2\text{O}_3$   
 $\text{H}_2\text{O}$   
 $\text{BC}$

## Hypergolic



$\text{NO}_x$   
 $\text{H}_2\text{O}$   
 $\text{BC}$

## Kerosene



$\text{NO}_x$   
 $\text{H}_2\text{O}$   
 $\text{BC}$

## Cryogenic



$\text{NO}_x$   
 $\text{H}_2\text{O}$

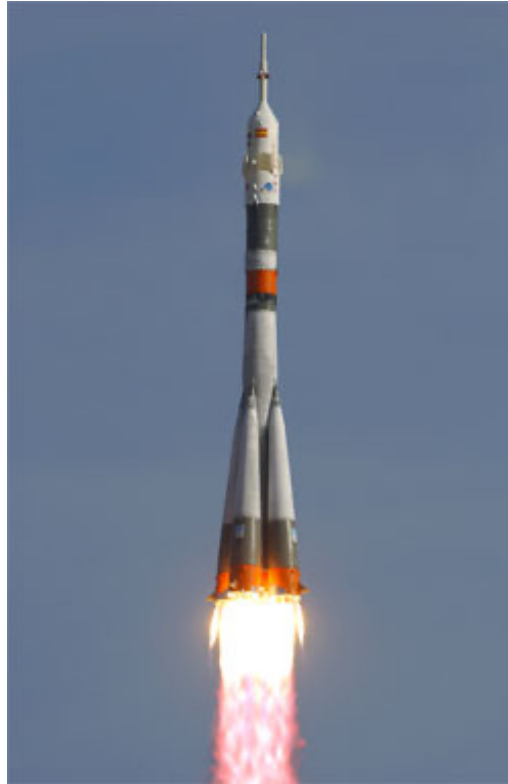
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$\text{NO}_x$   
 $\text{H}_2\text{O}$   
 $\text{BC}$

## Cryogenic



$\text{NO}_x$   
 $\text{H}_2\text{O}$

Climate  
concern



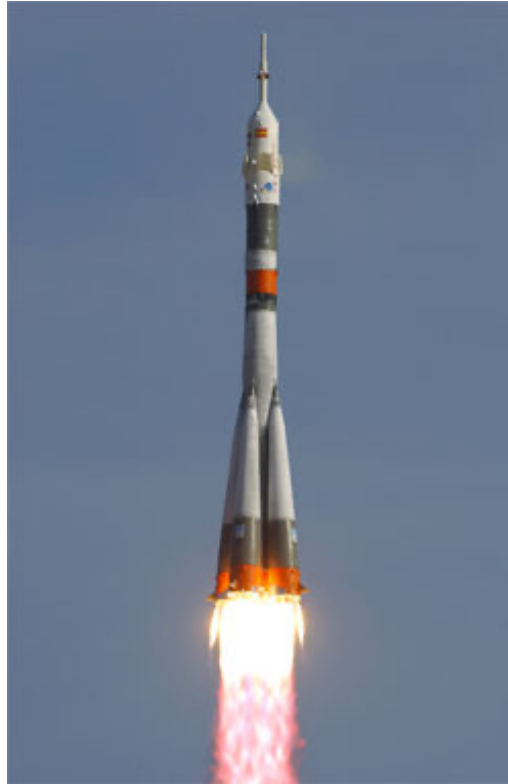
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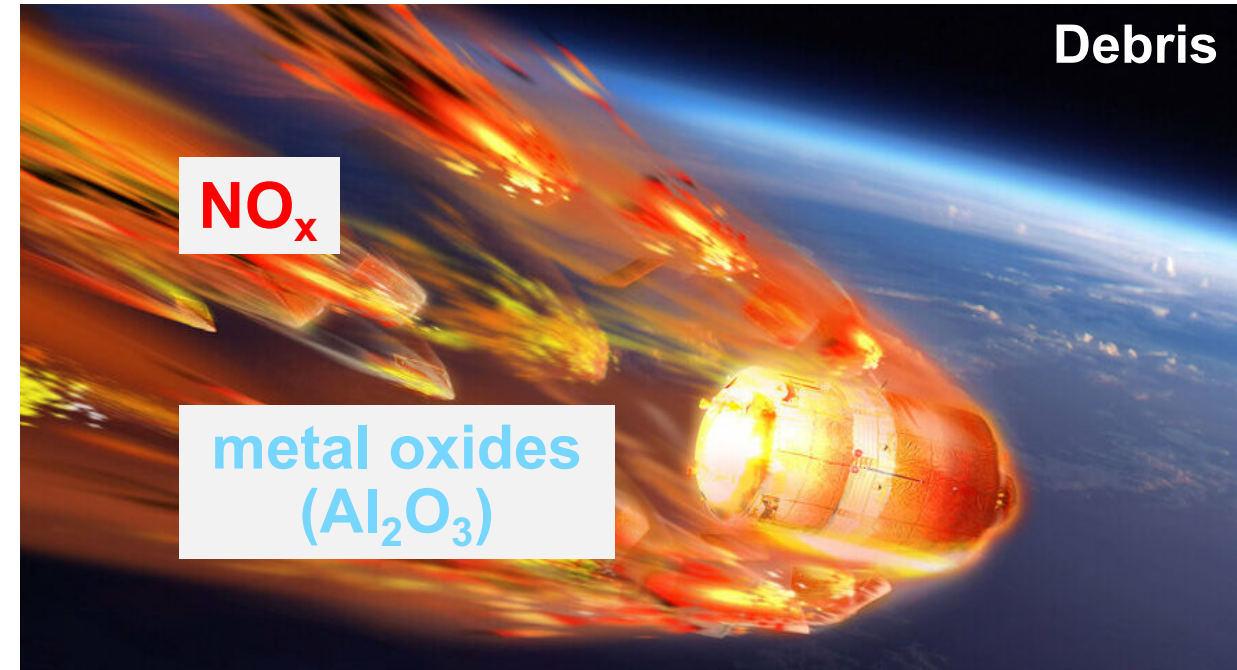
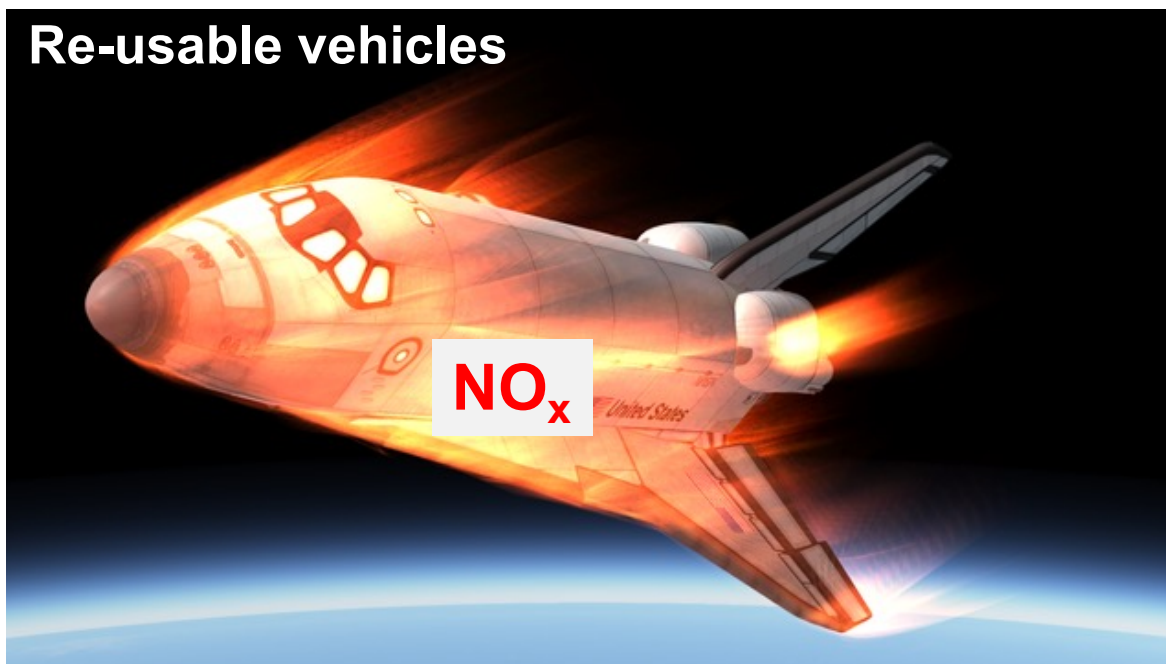
$\text{NO}_x$   
 $\text{H}_2\text{O}$

Ozone  
depletion

# Air pollutant emissions from re-entry

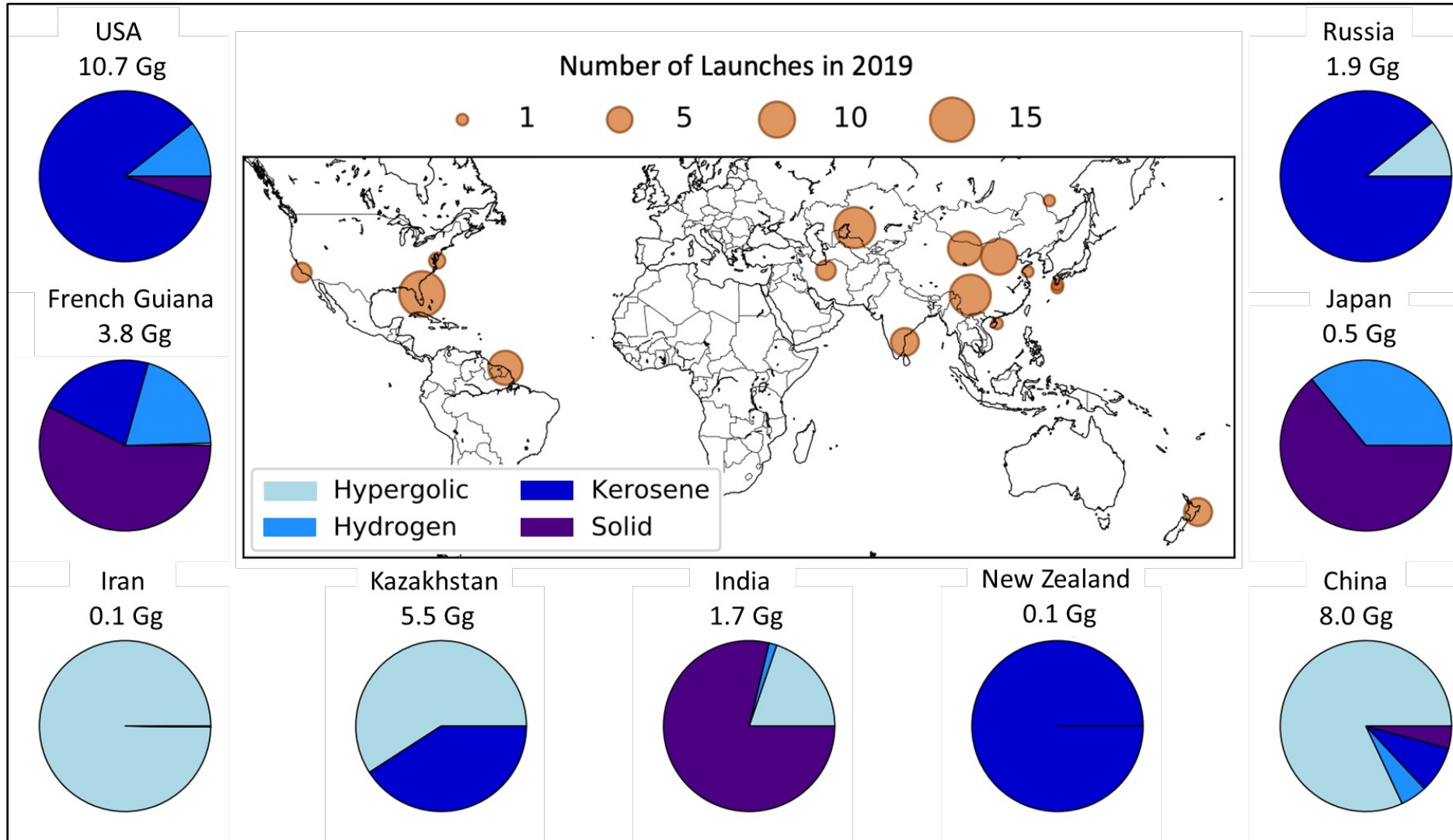


2-40 Gg  $\text{NO}_x$  per year





# Calculate and map a single year of emissions



## Annual Emissions:

H<sub>2</sub>O: 11 Gg  
BC: 0.5 Gg  
Al<sub>2</sub>O<sub>3</sub>: 2 Gg  
HCl: 1 Gg  
Launch NO<sub>x</sub>: 0.2 Gg  
Re-entry NO<sub>x</sub>: 2 Gg

Artificial NO<sub>x</sub> similar  
to lower end estimate  
of natural NO<sub>x</sub>

~100 successful launches in 2019

Reaches 135 in 2021. Already 148 in 2022.



# Incorporate these in GEOS-Chem

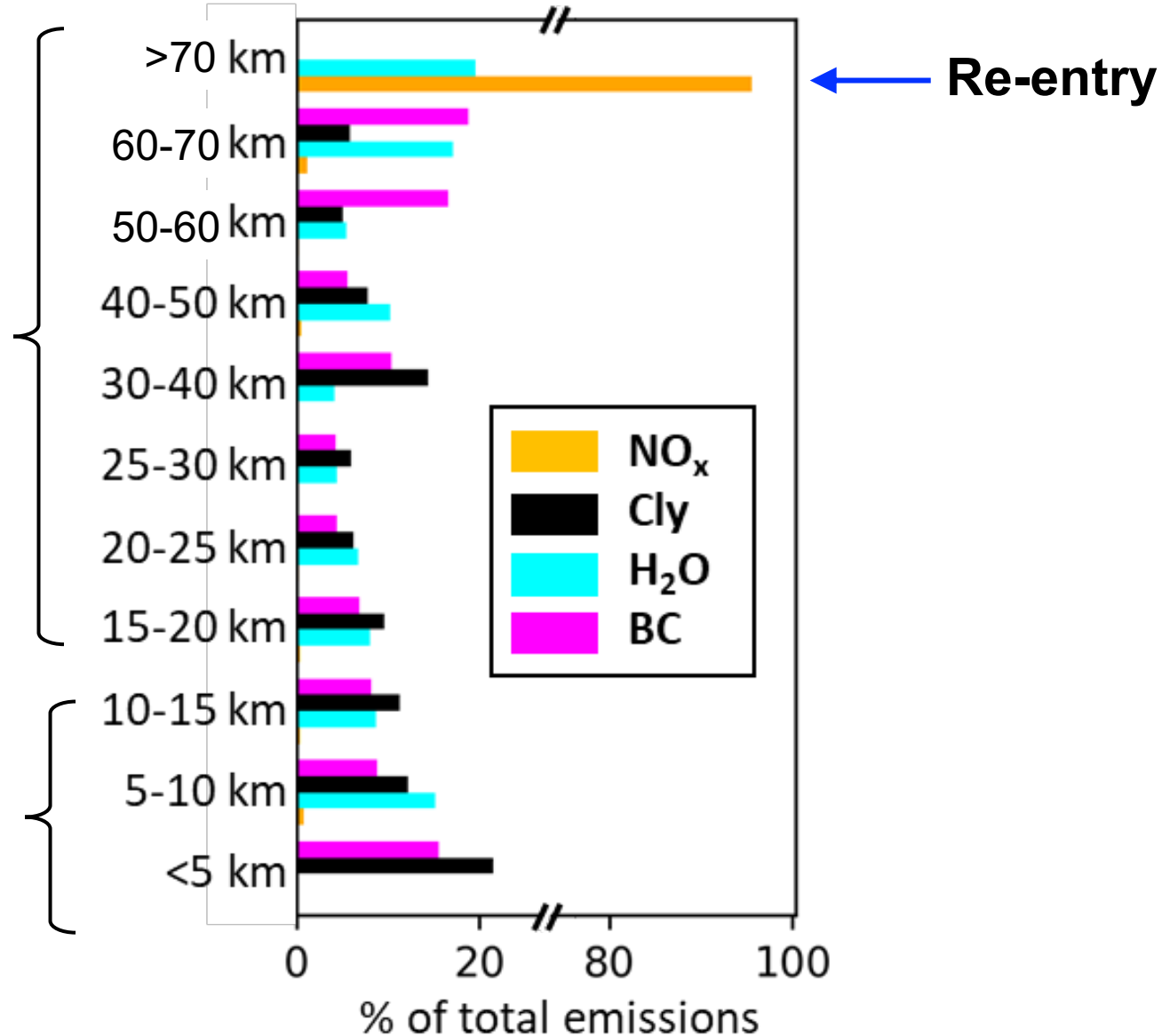
GEOS-Chem extends  
to **80 km**

## Stratosphere & mesosphere:

lifetime >2 years  
(*gravitational settling*)

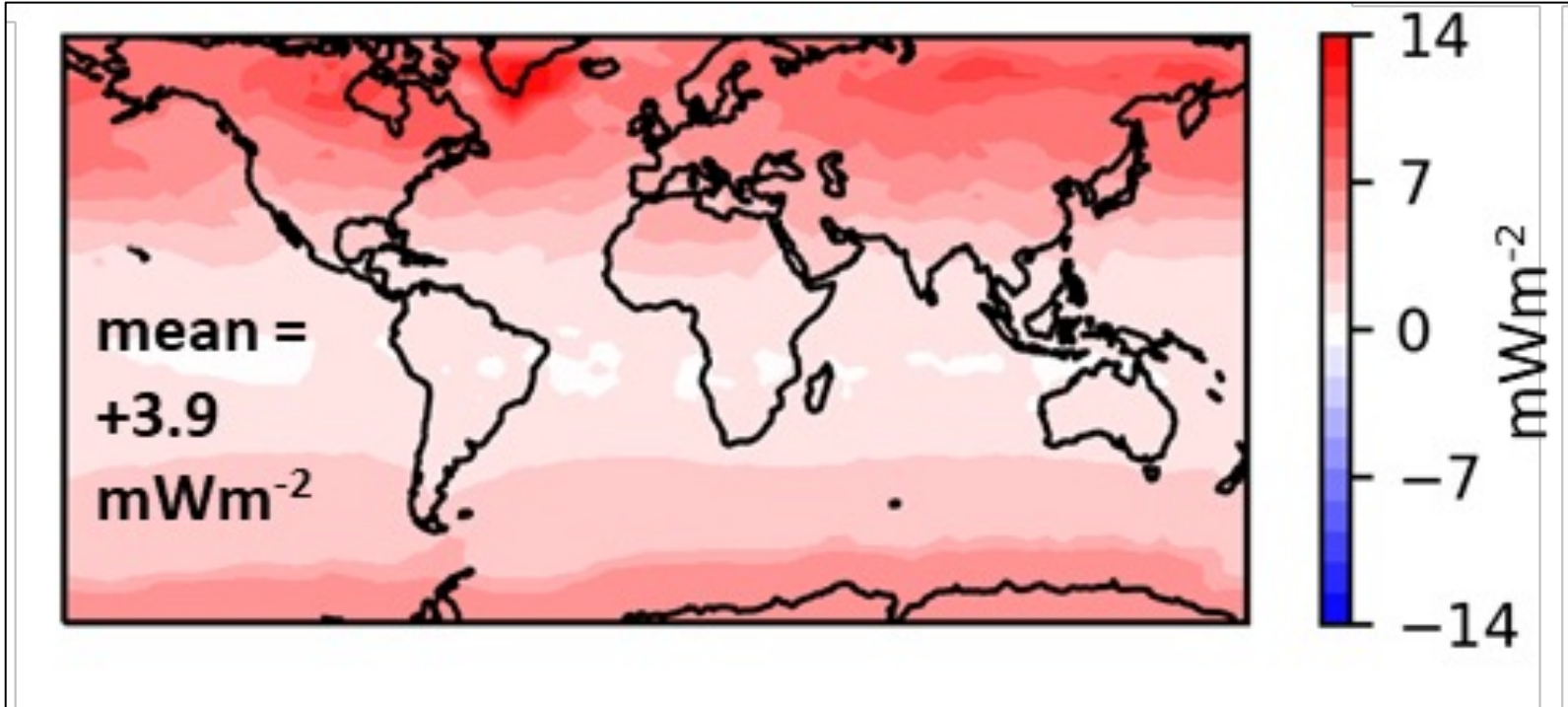
## Troposphere:

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

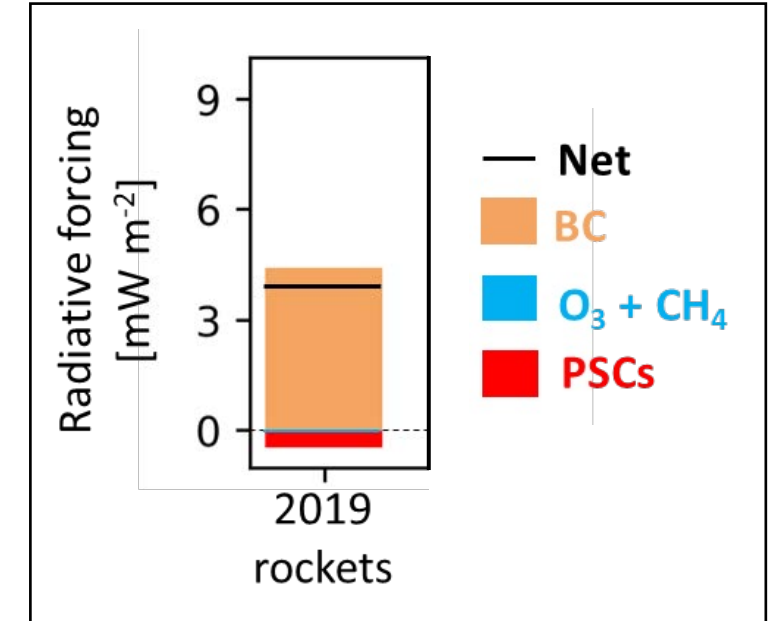


# Radiative forcing due to black carbon emissions

After 10 years of emissions assuming modest growth



Mostly due to BC

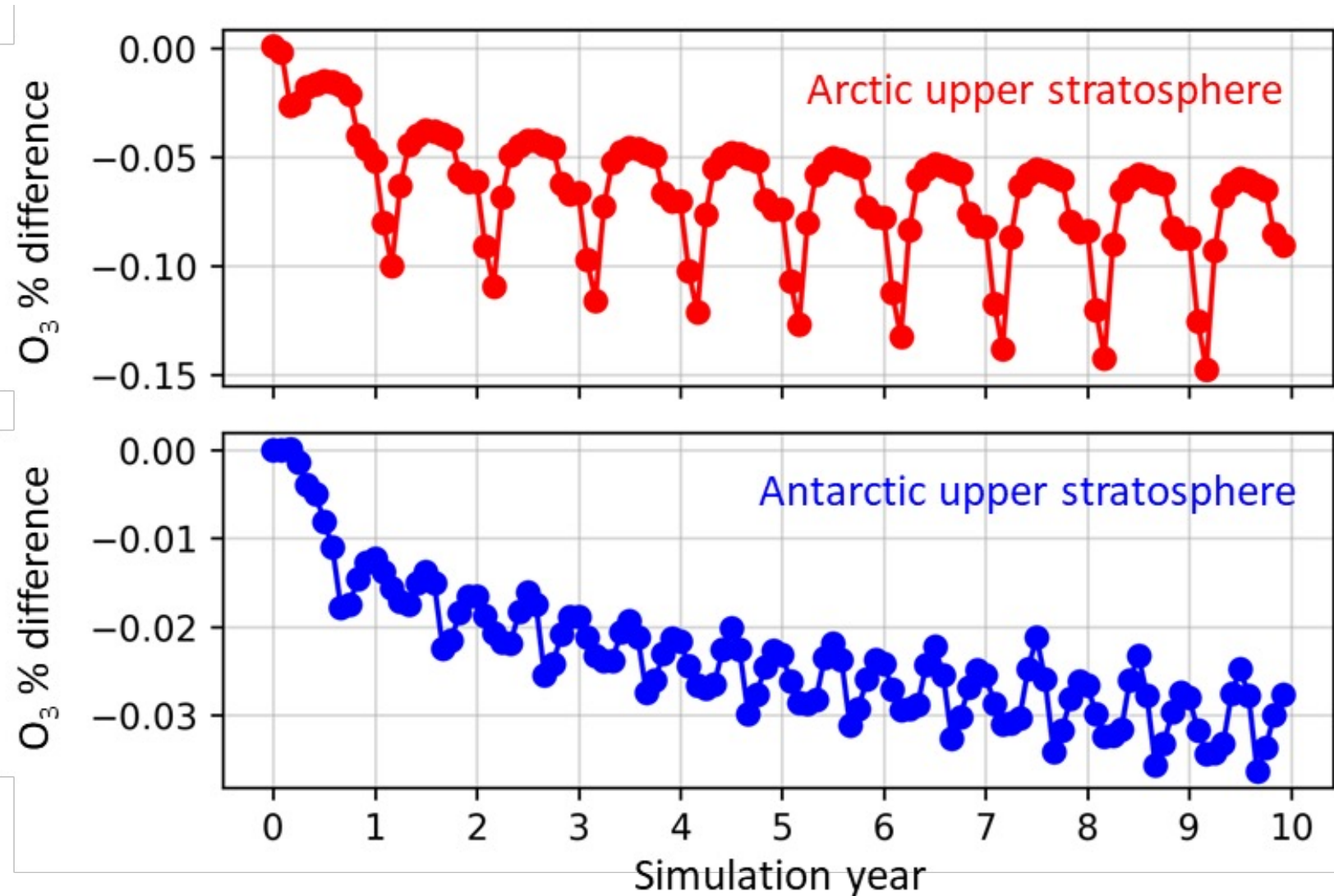


Rockets ~3% of BC radiative forcing from all anthropogenic sources, but only 0.01% of emissions.

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<sub>x</sub> 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



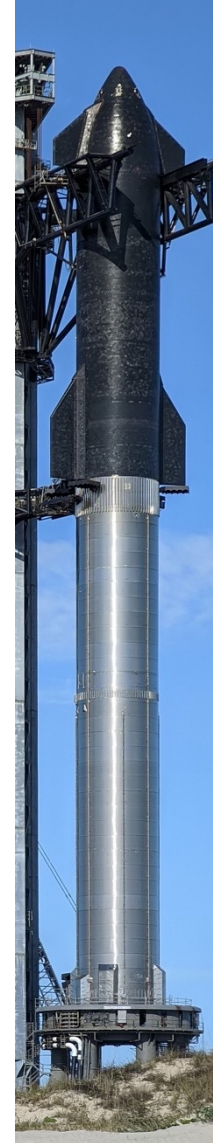
26 tonnes



60 satellites

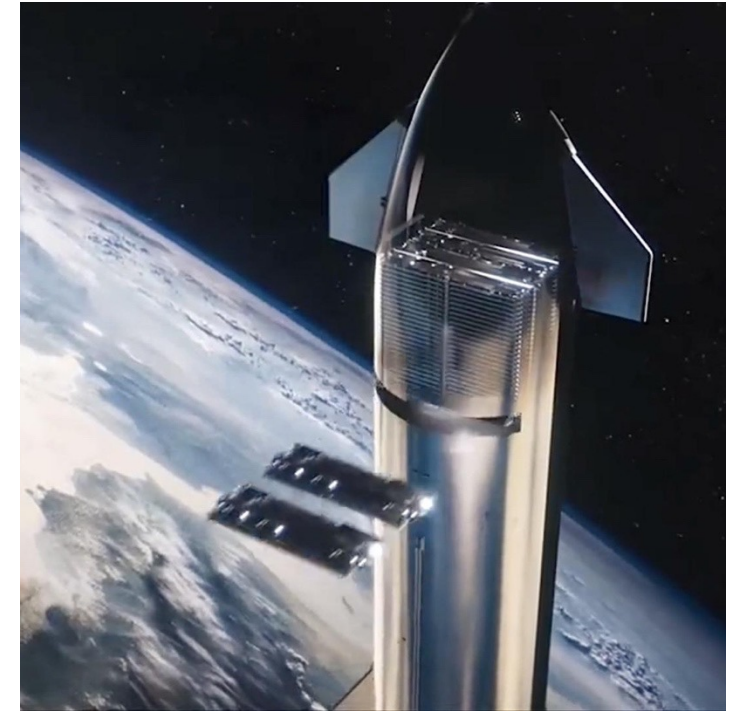
3,558 launched to date  
**318 deorbited**

Raptor



## SpaceX StarShip

~200 tonnes



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

# Take-homes and future work

Re-entry  $\text{NO}_x$  comparable with lower end estimate of natural  $\text{NO}_x$  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.