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/

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Harvard Seminar

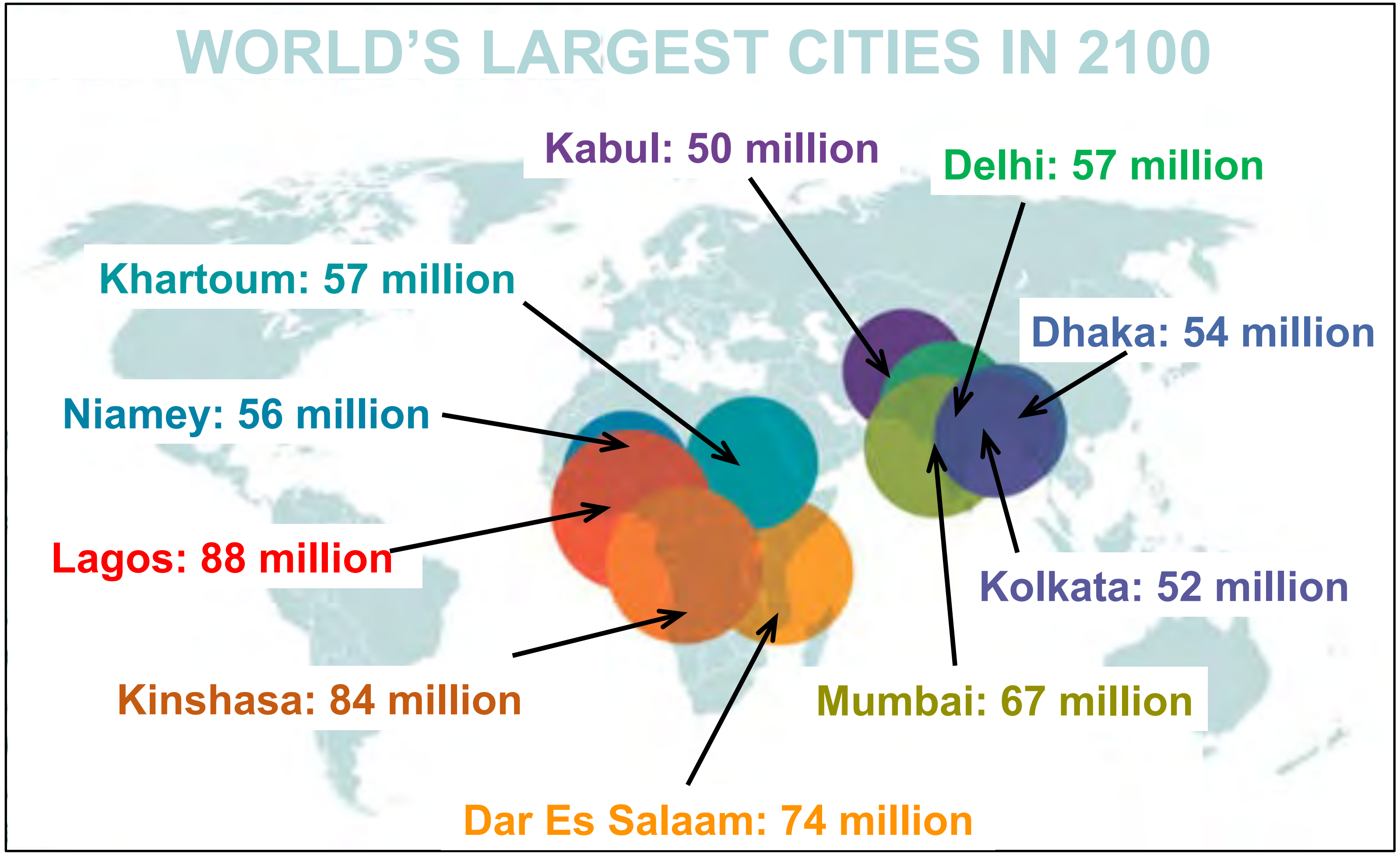
9 November 2022

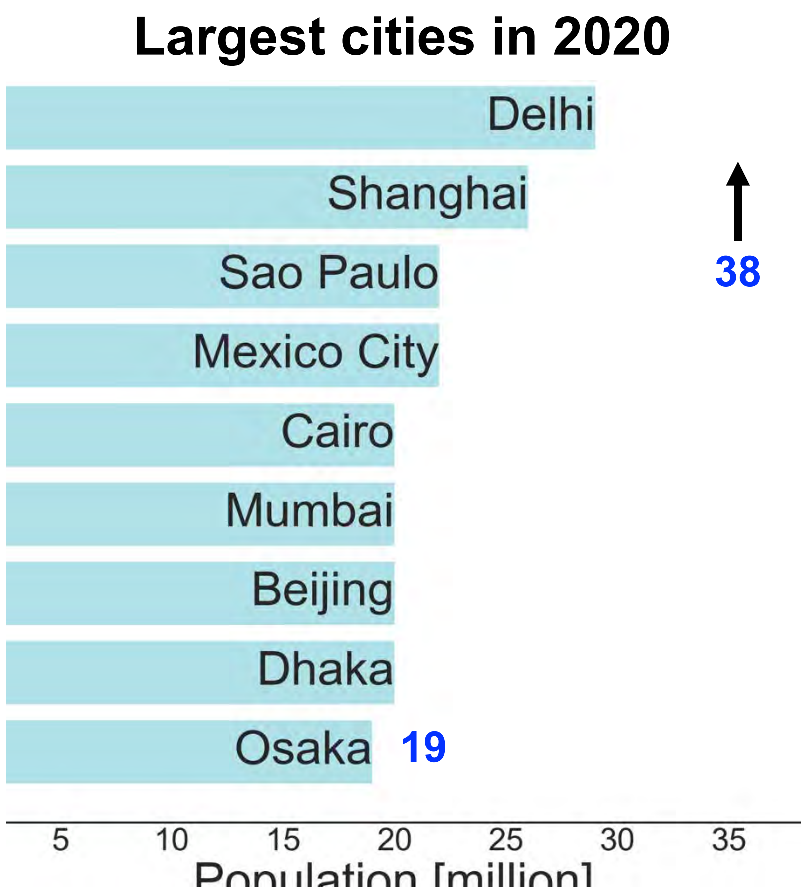
Fast-growing tropical megacities

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





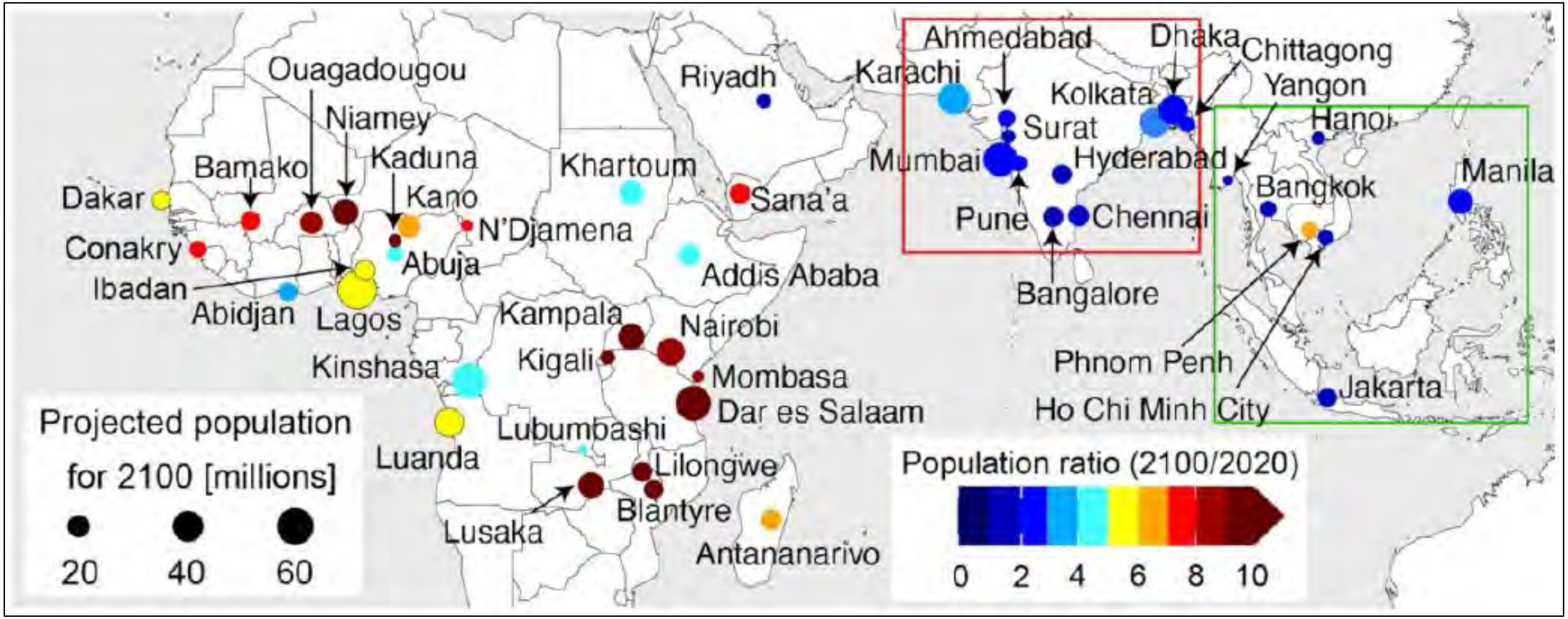
[Image credit: Gongda Lu]

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**

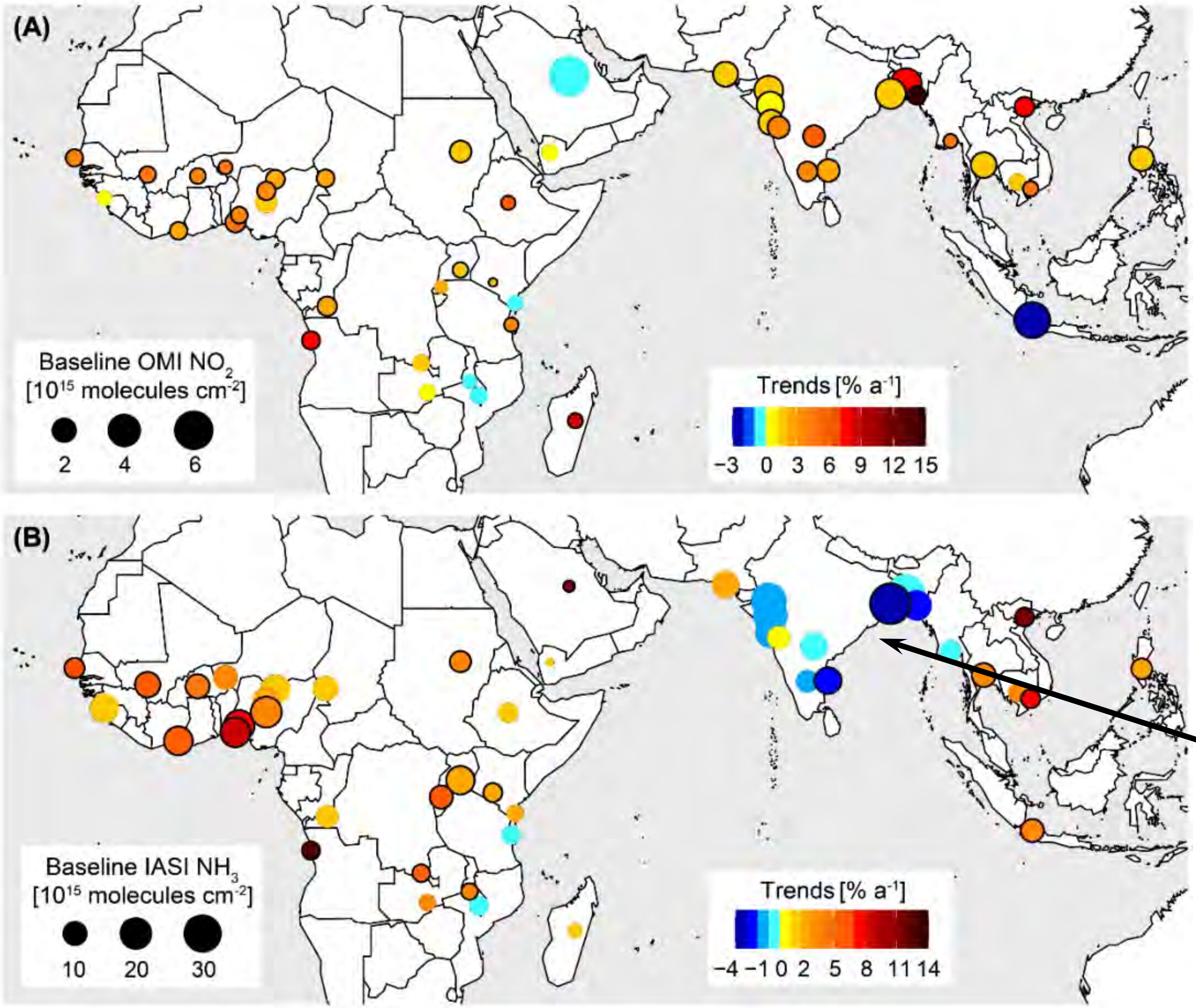
Steep annual increases in NO_x and NH_3

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

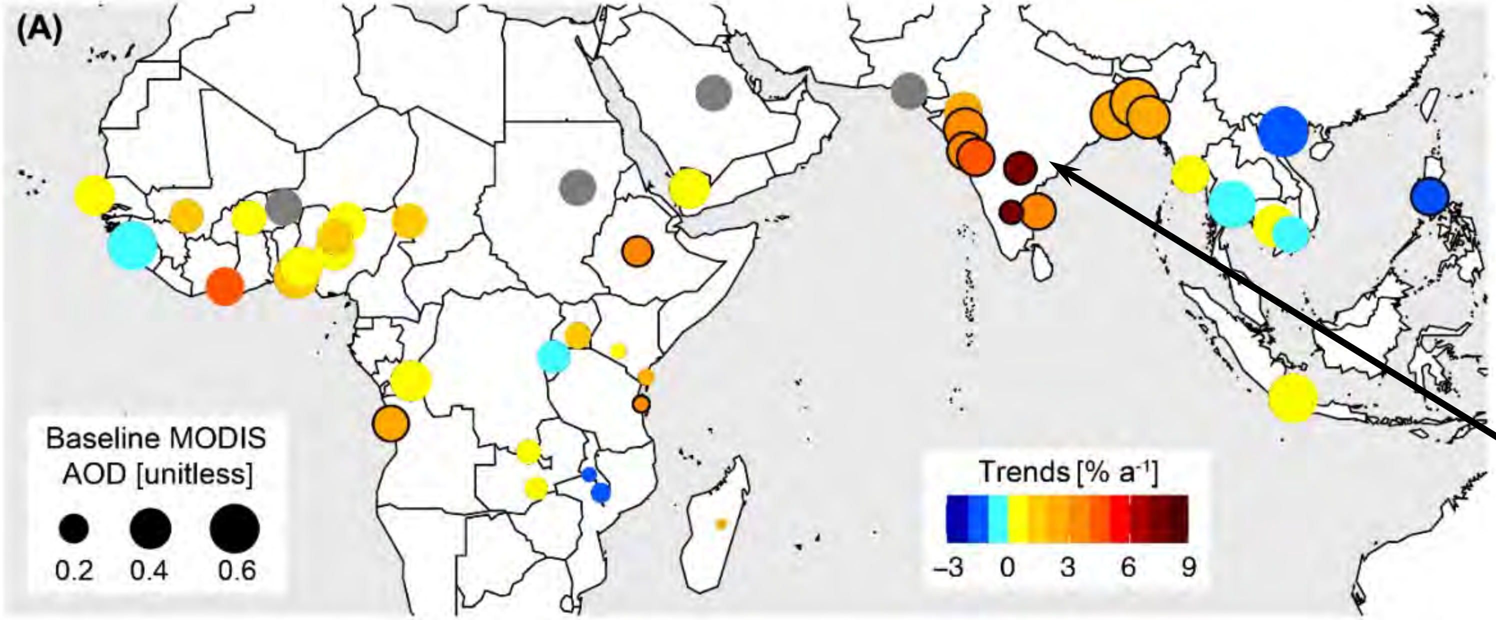
Decline over Indian subcontinent due to increase in uptake to acidic aerosols

NH_3 data from M. Van Damme, L. Clarisse, P.-F. Coheur at ULB

Annual changes in $PM_{2.5}$ and ozone production regimes

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

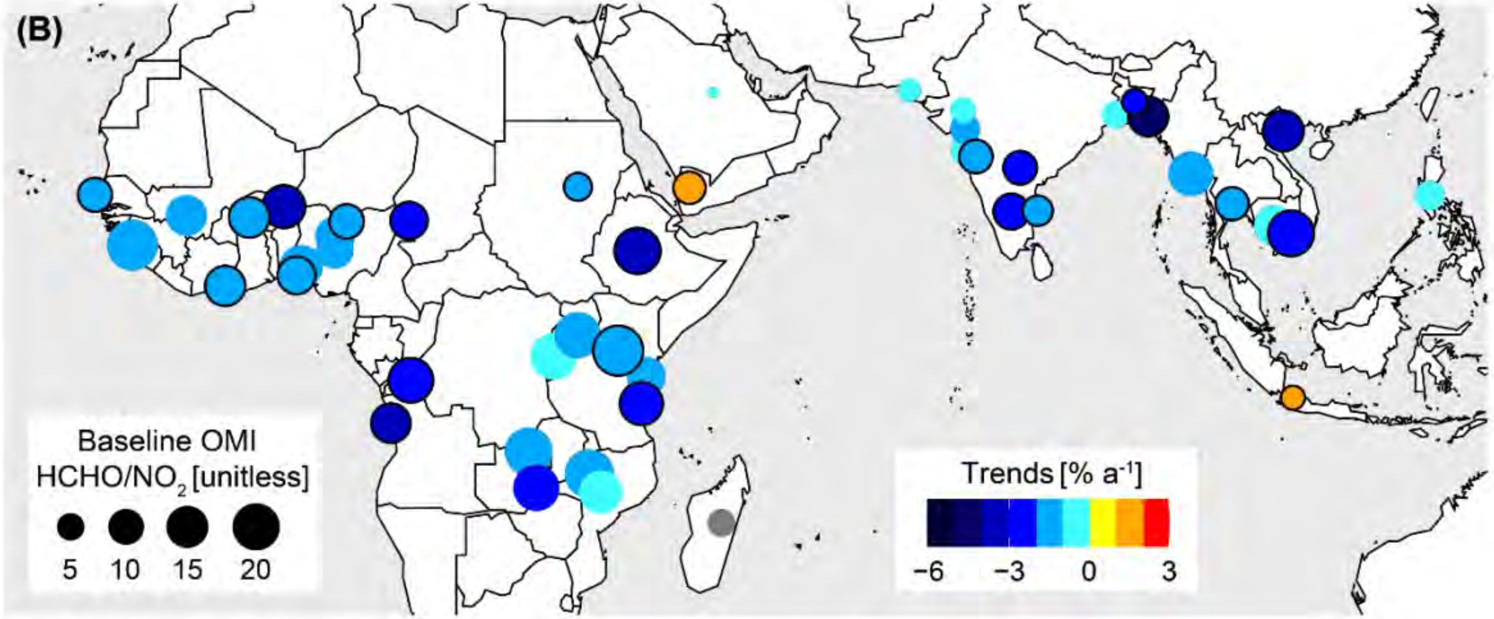
MODIS: Moderate resolution imaging spectroradiometer



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

Increases in $PM_{2.5}$ precursors SO_2, NH_3, NO_x

$HCHO/NO_2$ trends
(proxy for ozone production regime)
[2005-2018]

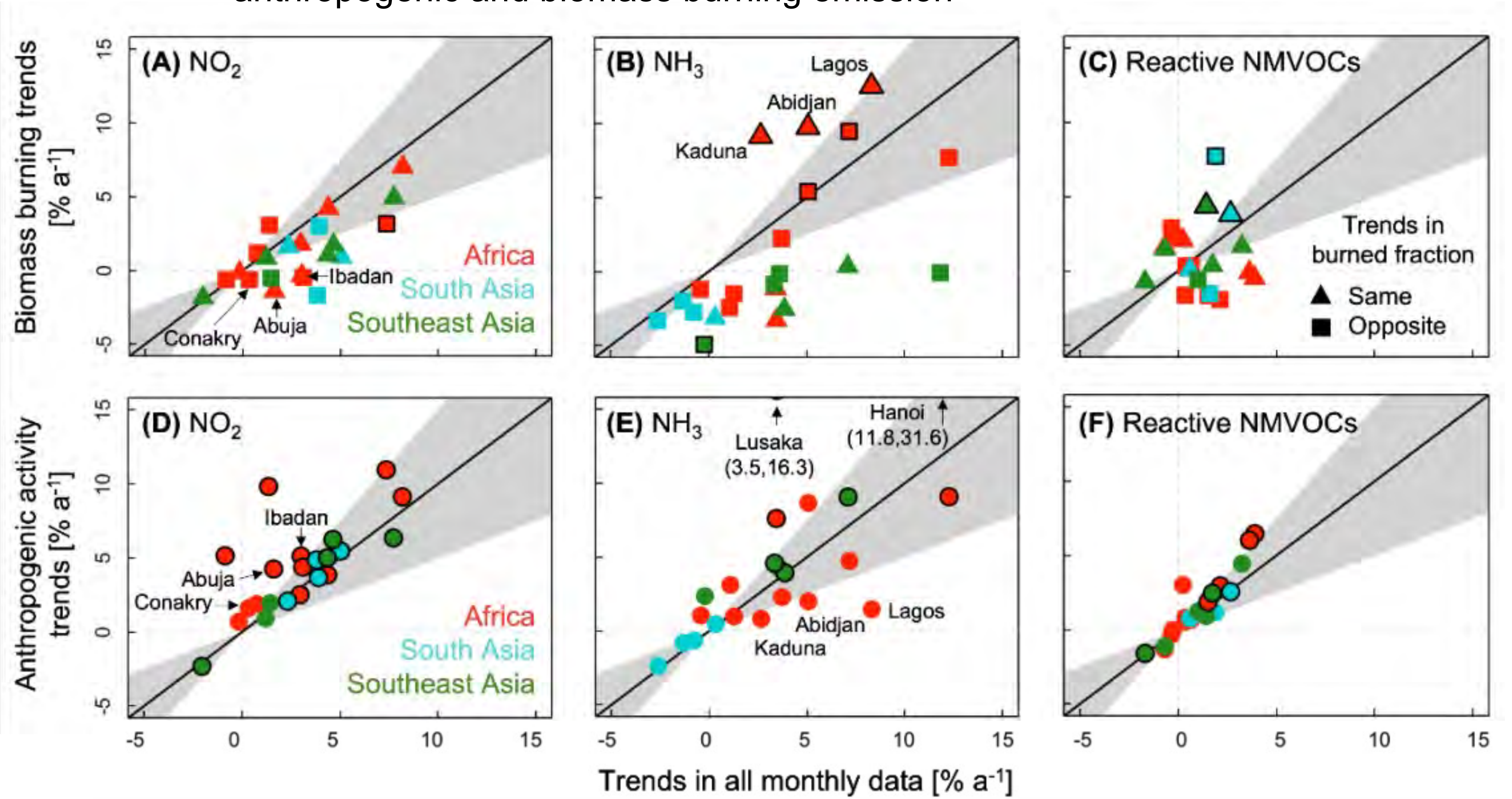


Ratio > 5:
O_3 production sensitive to NO_x

Transitioning to NO_x saturated or VOC sensitive

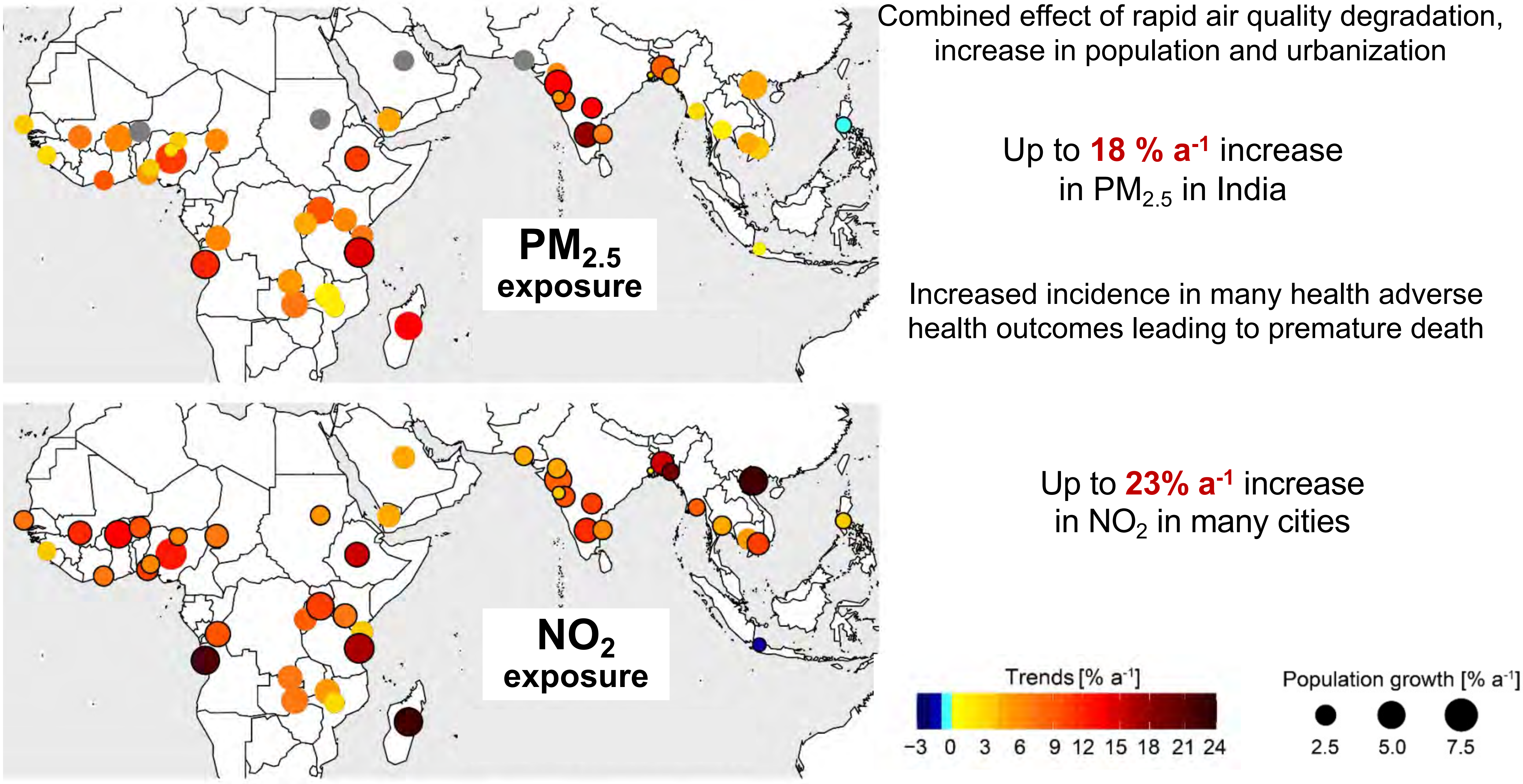
What's driving the observed trends?

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



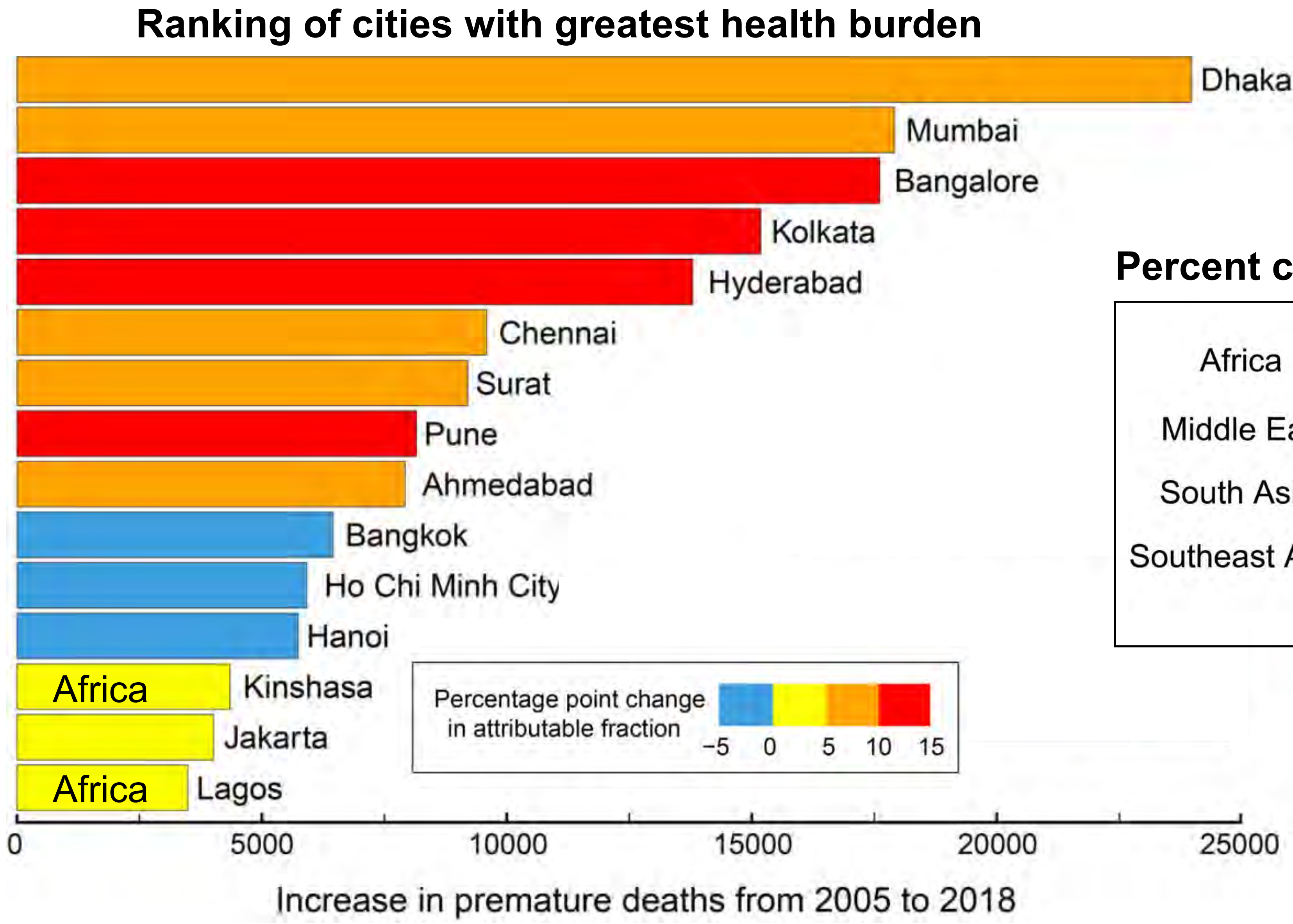
Combined effect of rapid air quality degradation, increase in population and urbanization

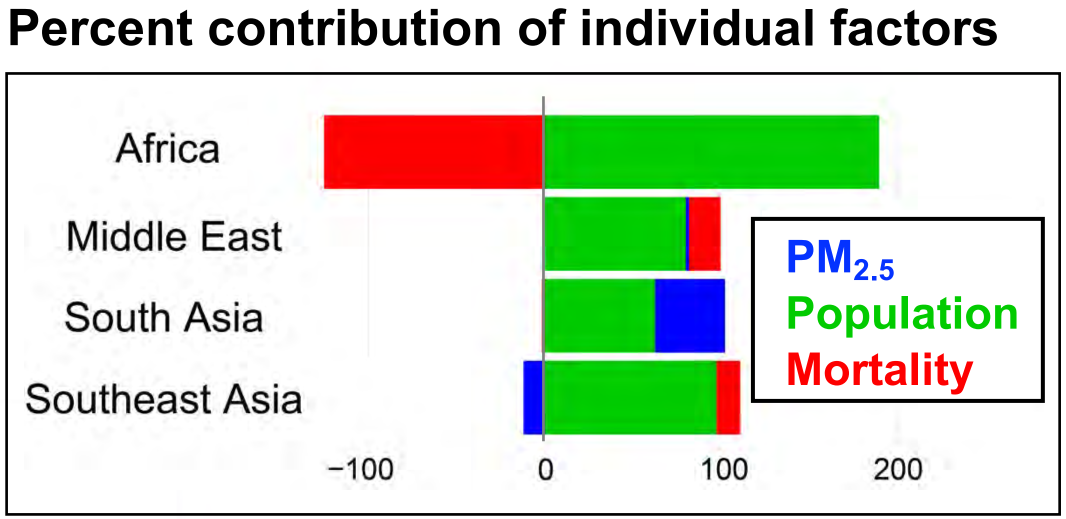
Up to **18 % a^{-1}** increase in $PM_{2.5}$ in India

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

Up to **23% a^{-1}** increase in NO_2 in many cities

Premature mortality attributable to rise in $PM_{2.5}$ exposure





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

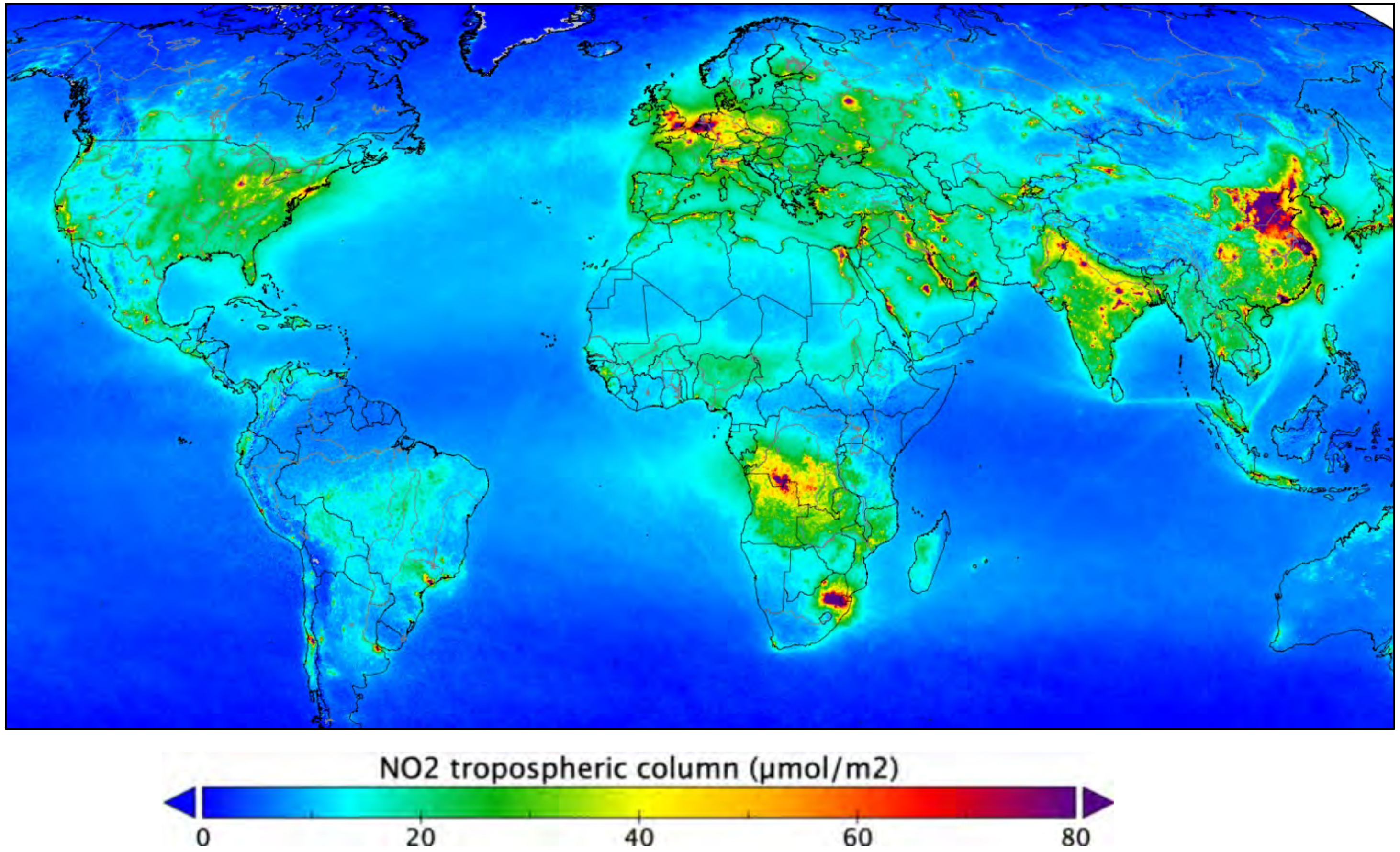
Bex Horner
PhD

Eleanor Smith
PhD

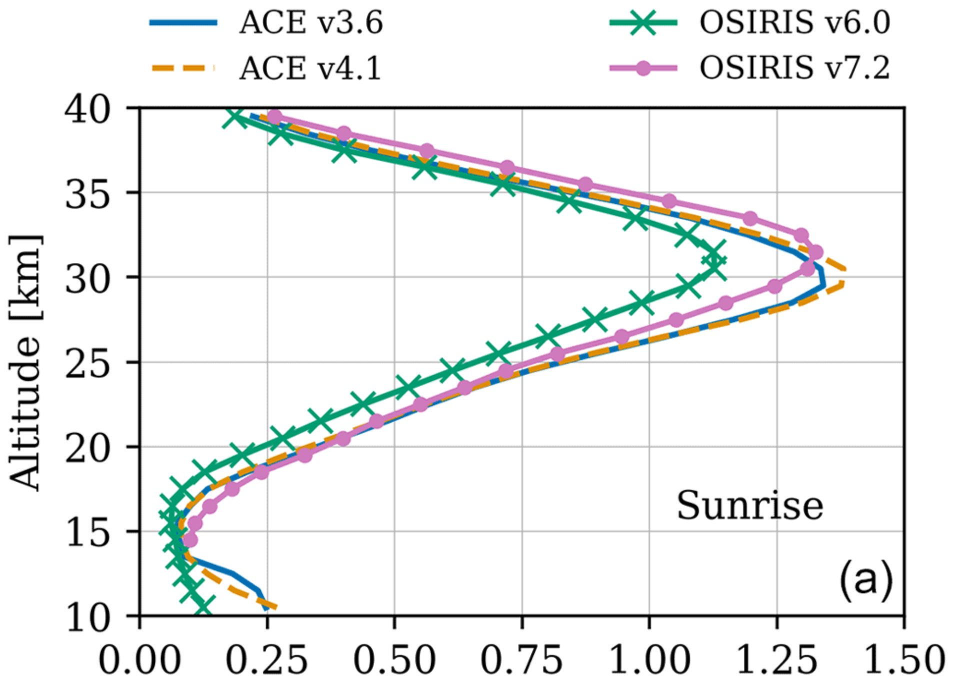
Reactive nitrogen in the remote troposphere

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

Nadir-viewing instruments observe the whole column



Limb-viewing instruments not sensitive to troposphere

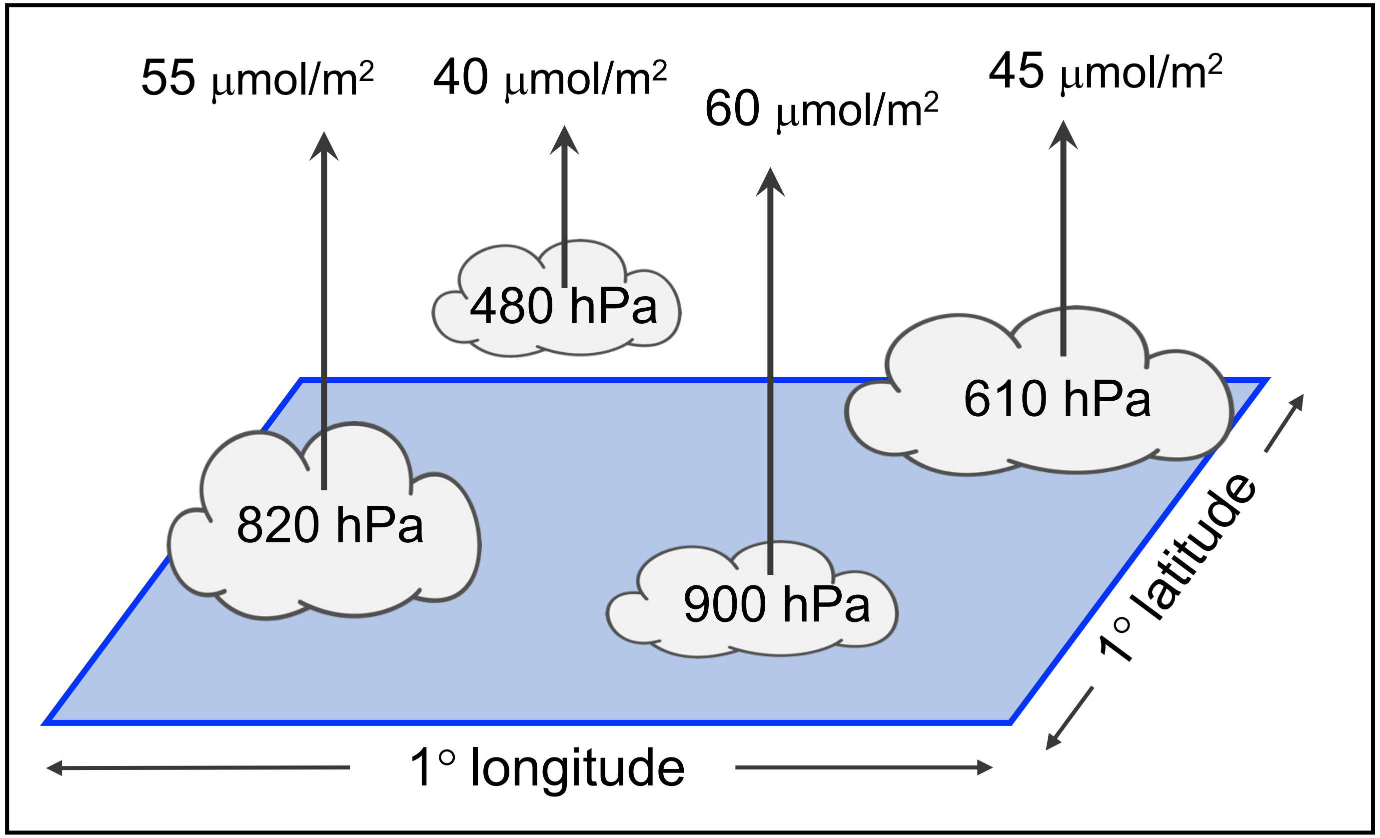


[Dubé et al., 2022]

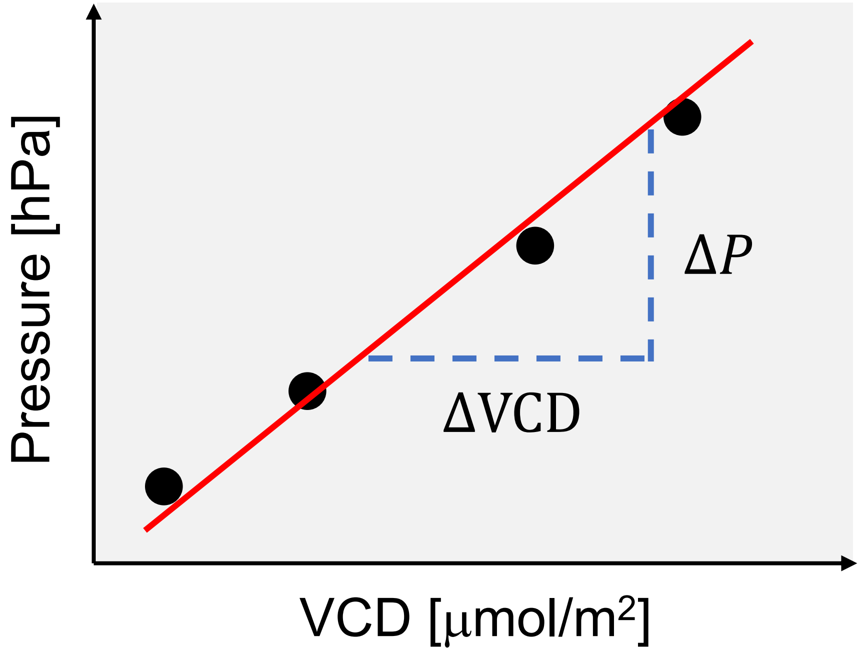
Aircraft observations limited in space and time

Cloud-slicing satellite observations to address data scarcity

Clusters of partial columns above optically thick clouds:



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

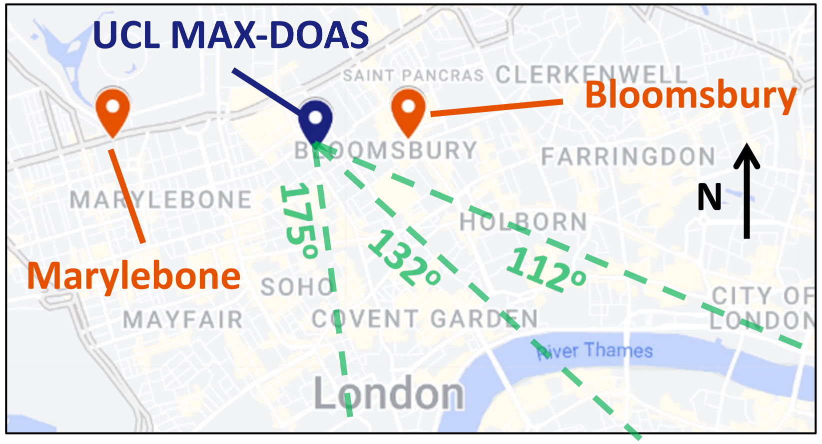


Calculate average mixing ratio between target pressure ranges:

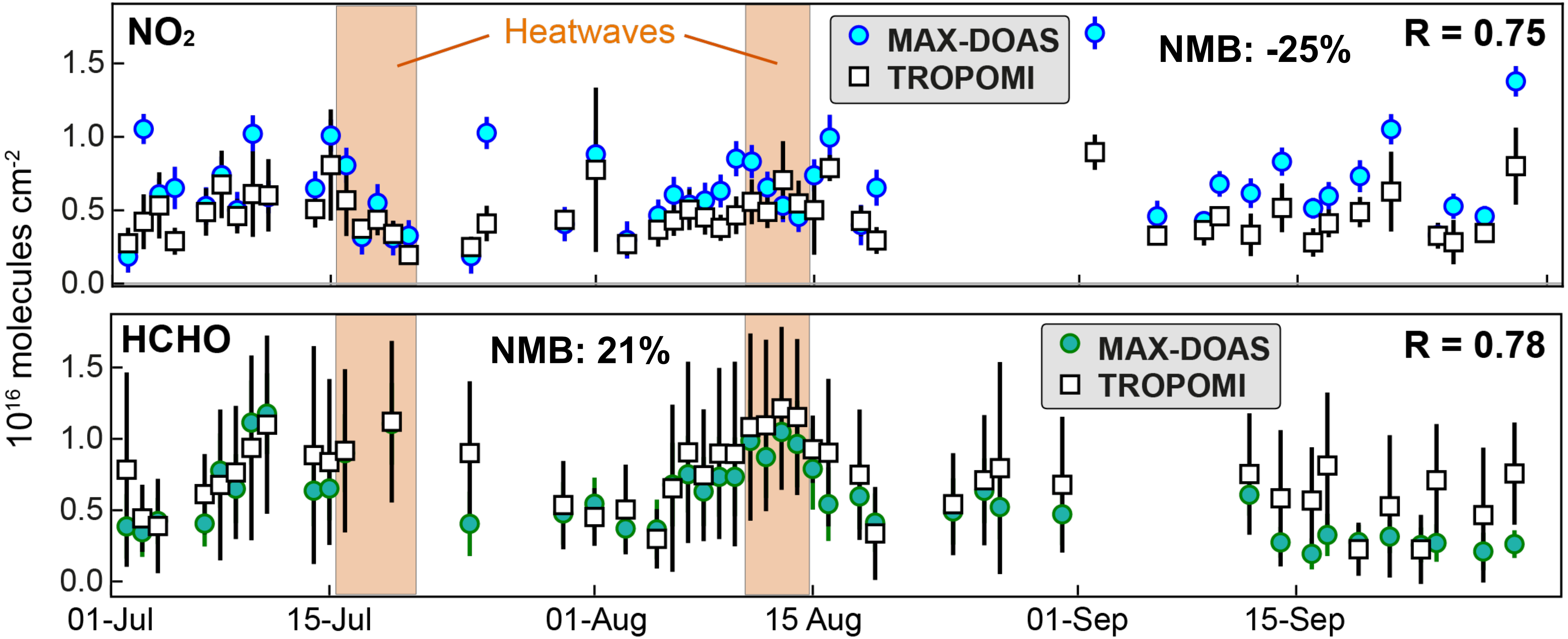
$$\mathrm{NO_2\ VMR} = \frac{\Delta \mathrm{VCD}}{\Delta P} \times \mathrm{const}$$

Application to high-resolution TROPOMI instrument

TROPOMI validation with MAX-DOAS during 2022 heatwaves

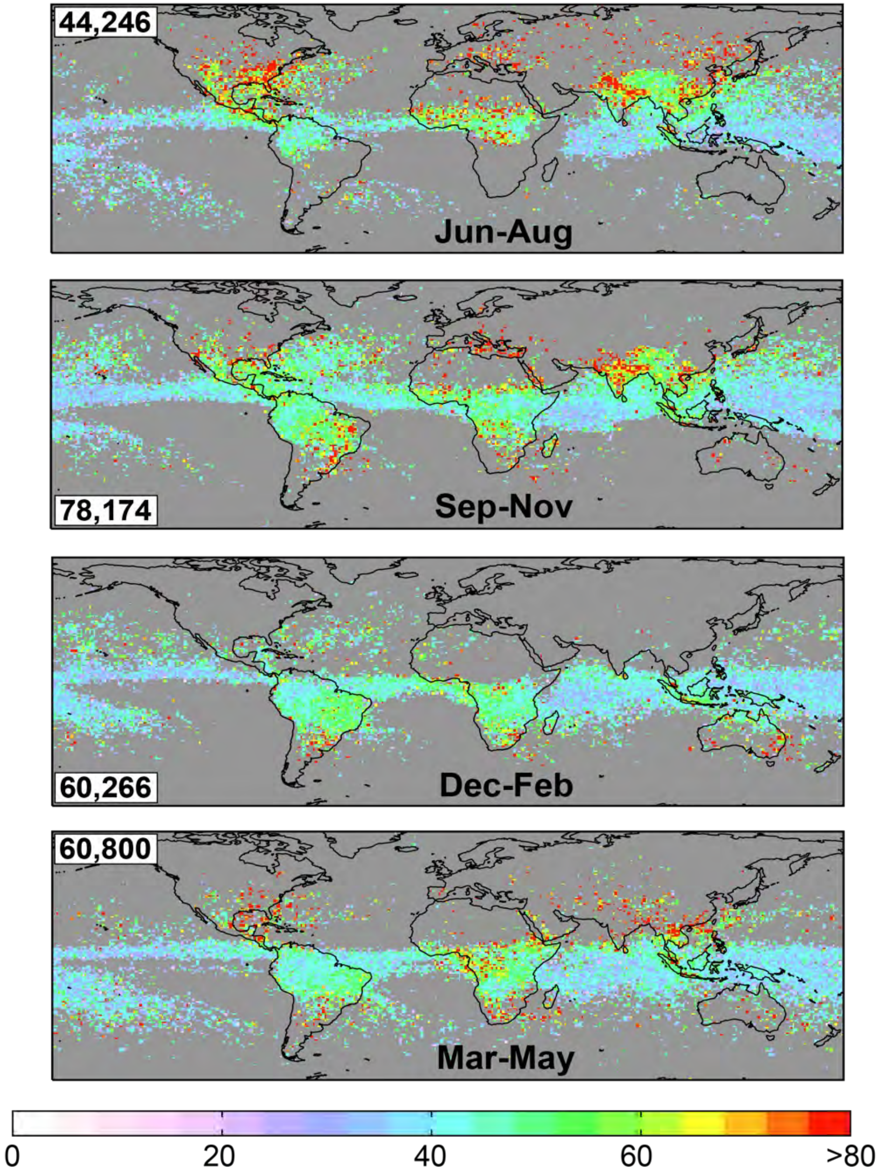


Capital Equipment Fund



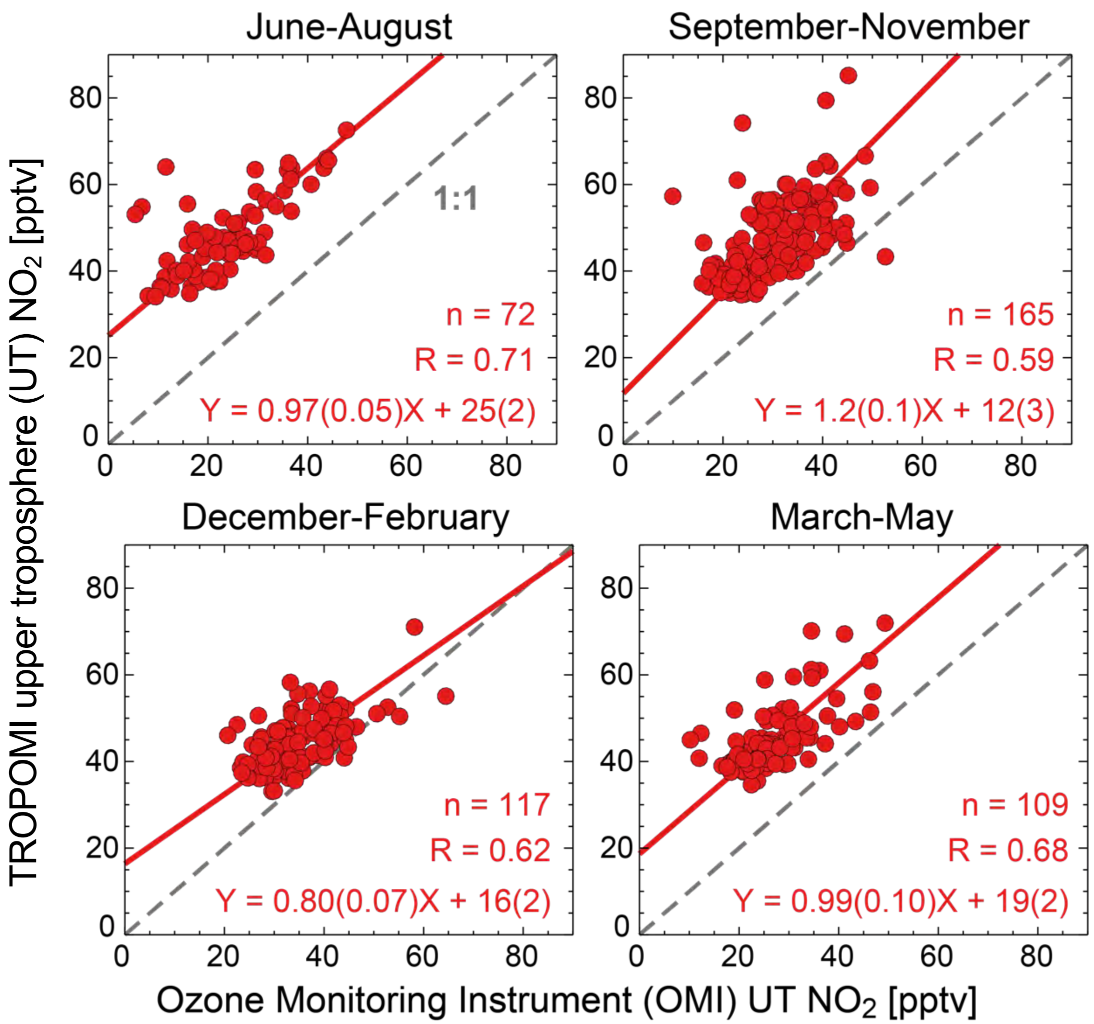
Seasonal means of NO_2 in the upper troposphere

Seasonal means at 8-12 km



[Marais et al., AMT, 2021]

Evaluation against product from OMI



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

Vertical profiles of NO_2 derived with the TROPOMI instrument

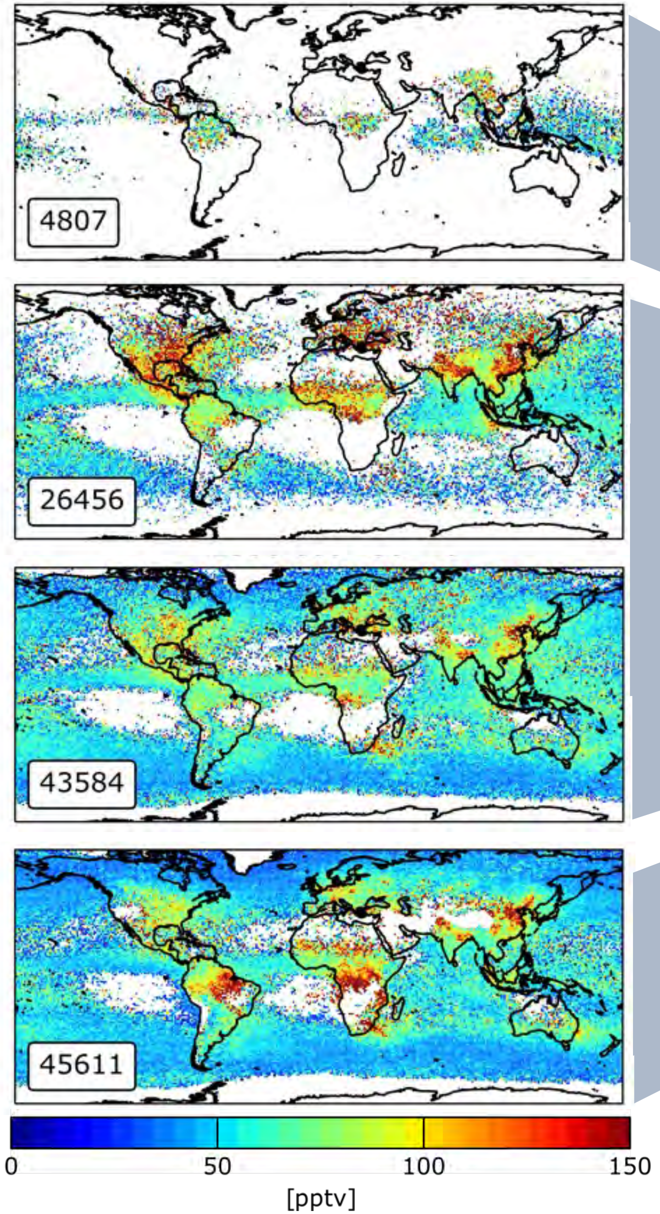
Vertical profiles in Jun-Aug

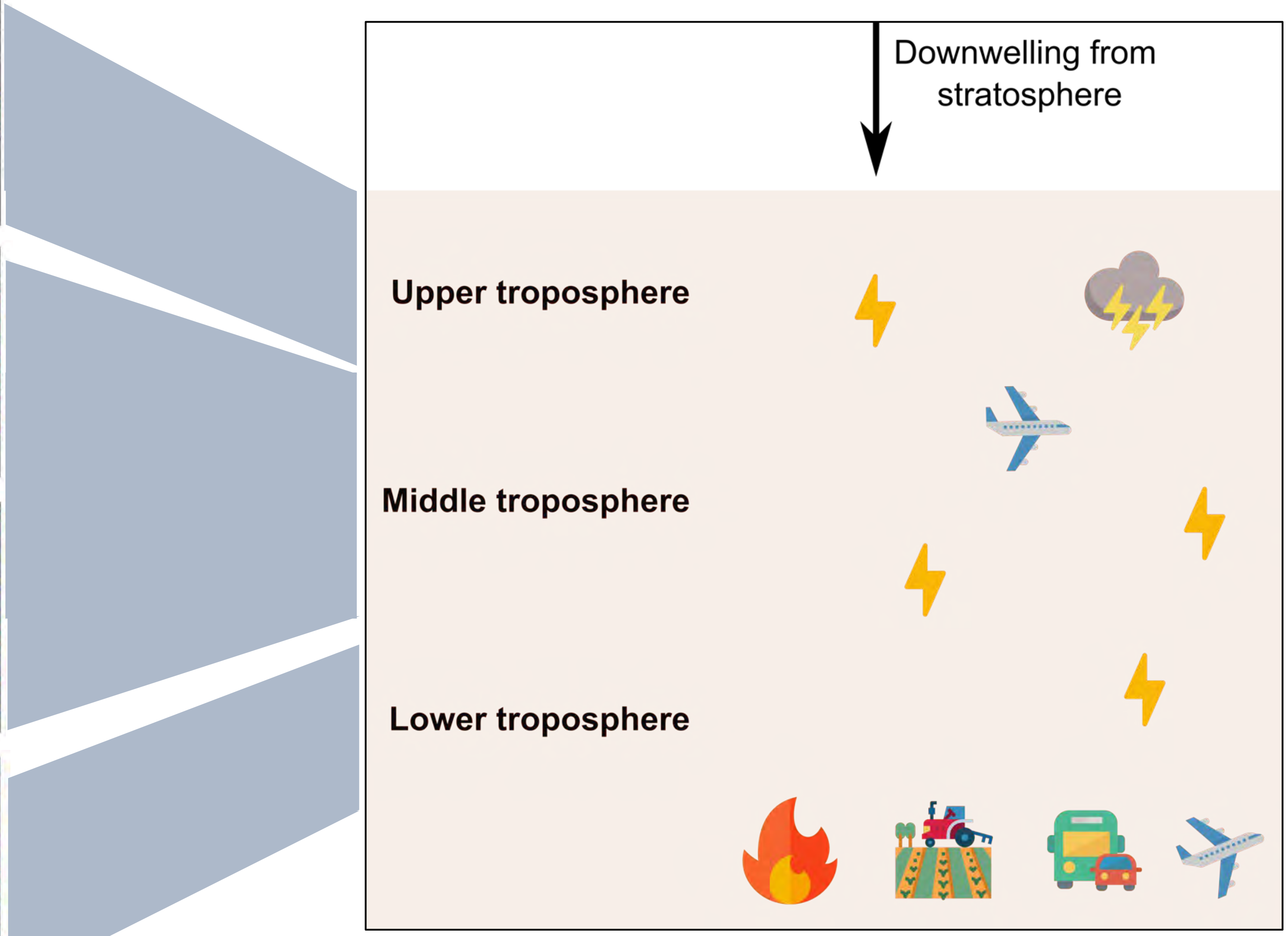
180-320 hPa
9-12 km

320-450 hPa
6-9 km

450-600 hPa
4-6 km

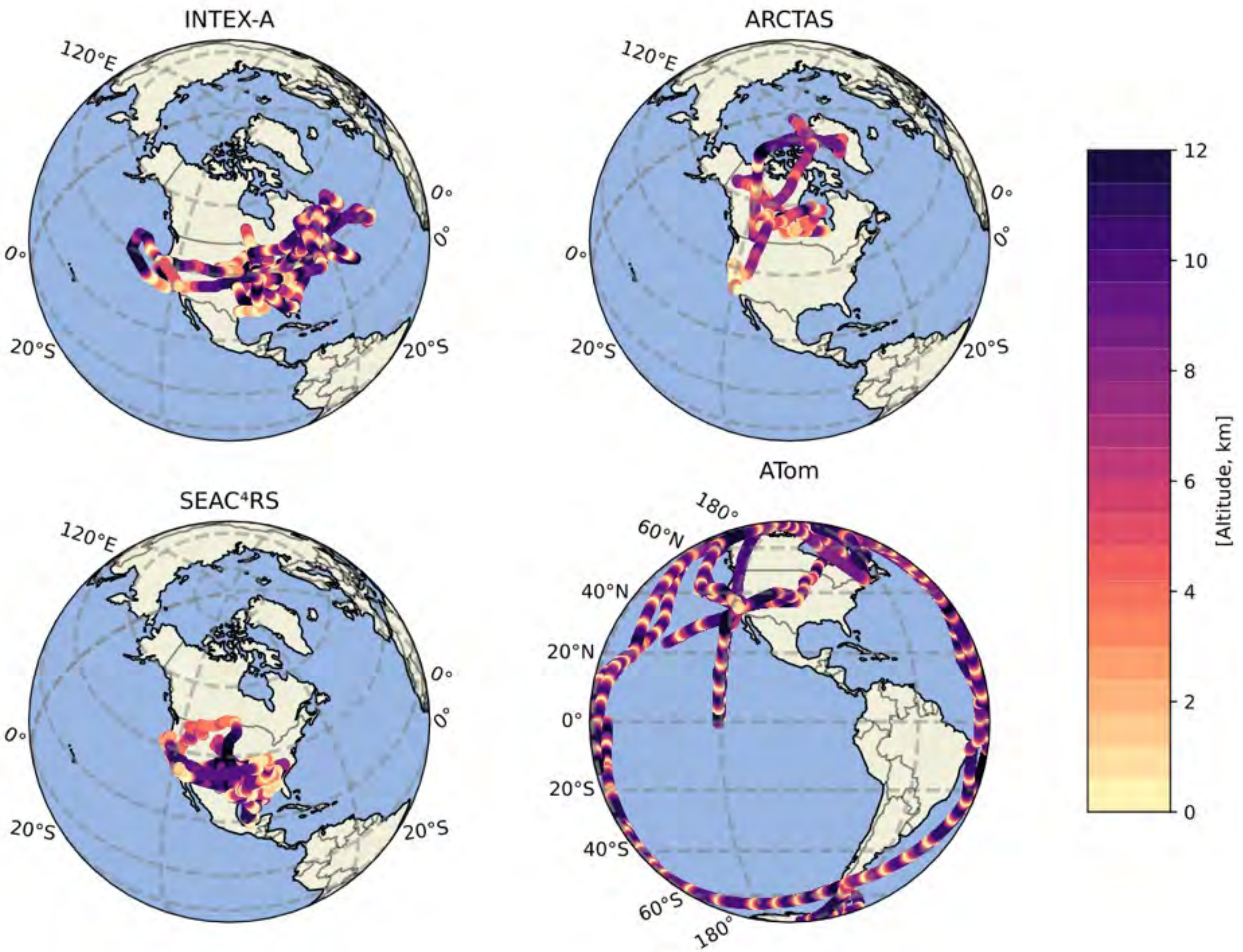
600-800 hPa
2-4 km





[Horner et al., in prep]

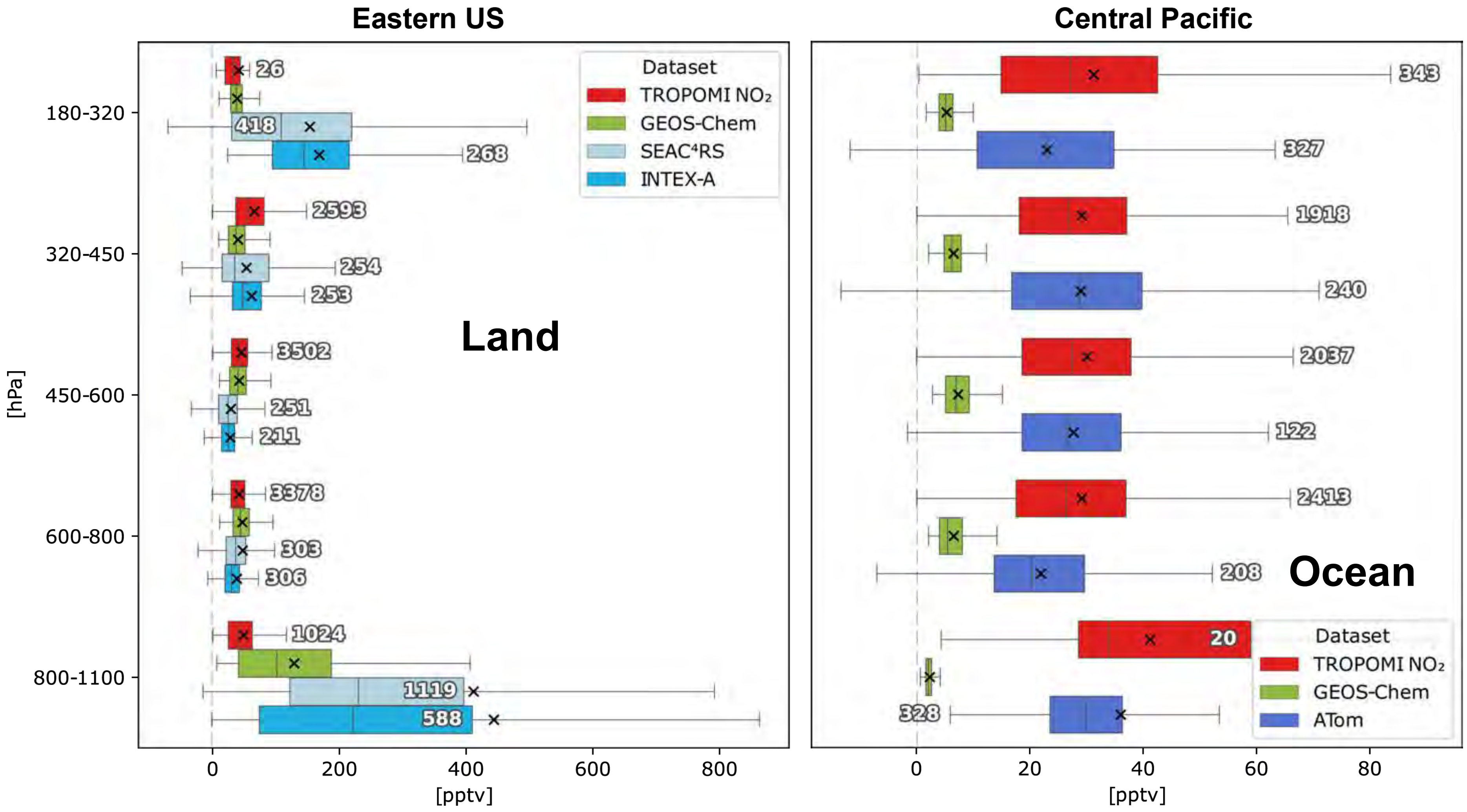
Cloud-sliced product validation with aircraft observations



Data provided by NASA DC8 Science Teams

Cloud-sliced product validation with aircraft observations

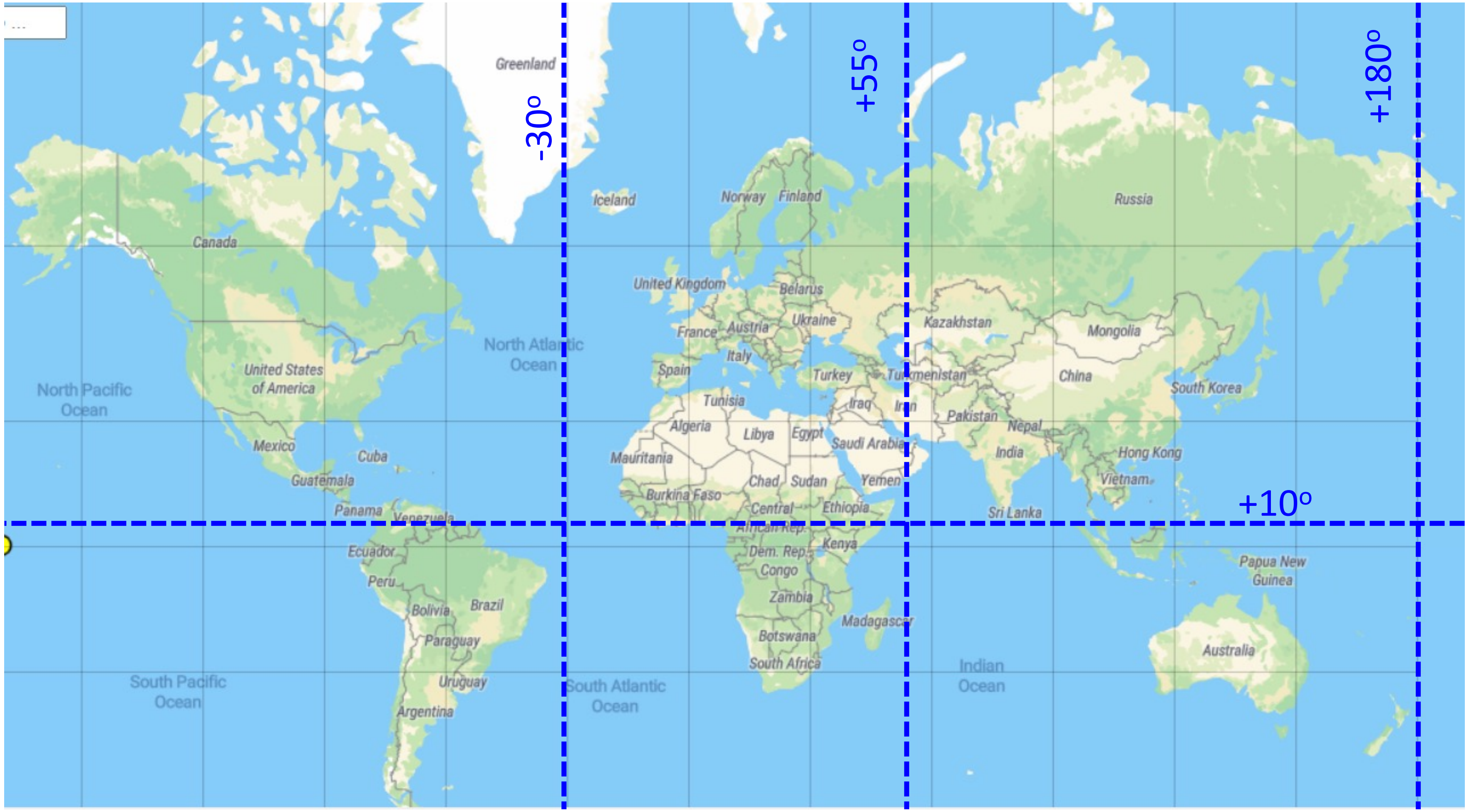
Comparison of collocated NO_2 mixing ratios in June-August



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

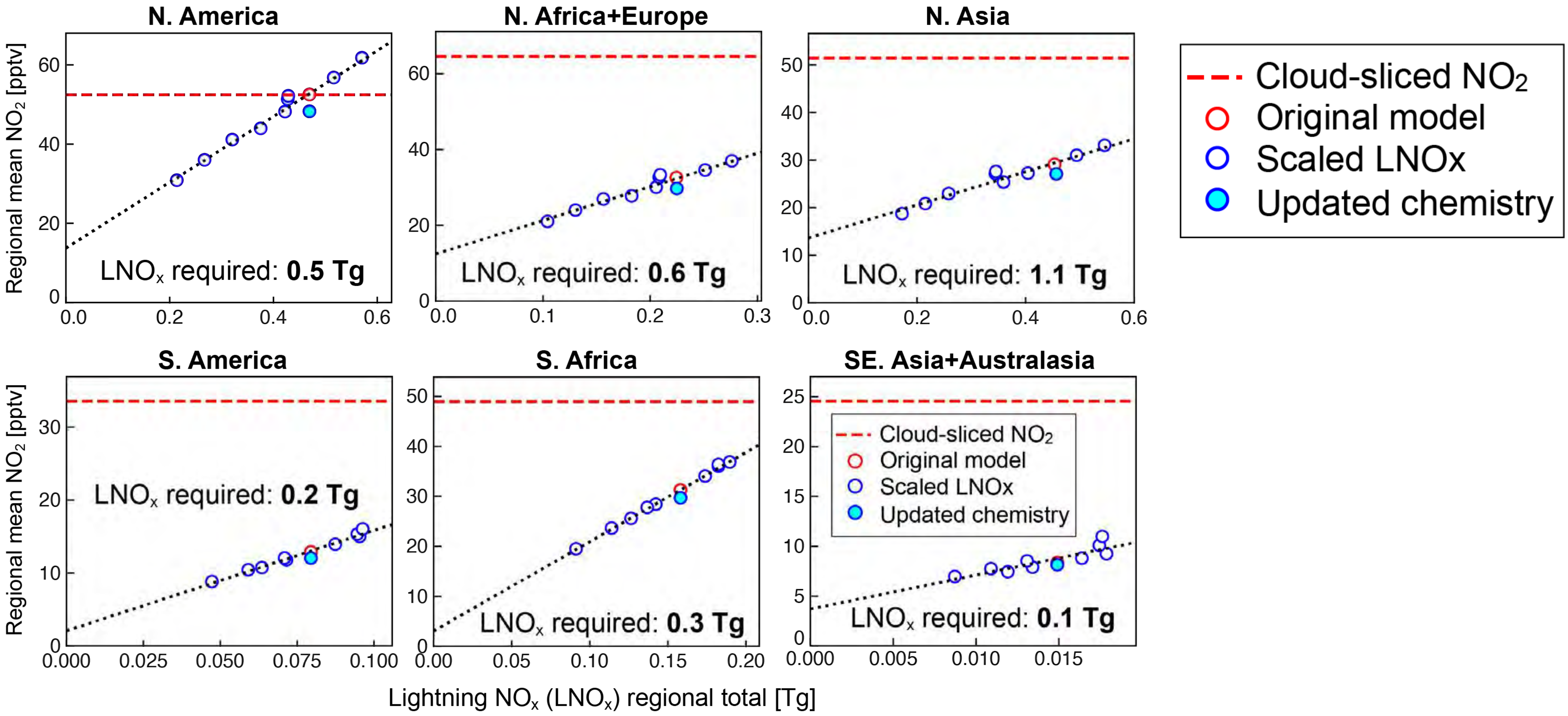
Model (GEOS-Chem) biased low throughout troposphere over remote ocean

Regional sensitivity to lightning NO_x emissions



Regional sensitivity to lightning NO_x emissions

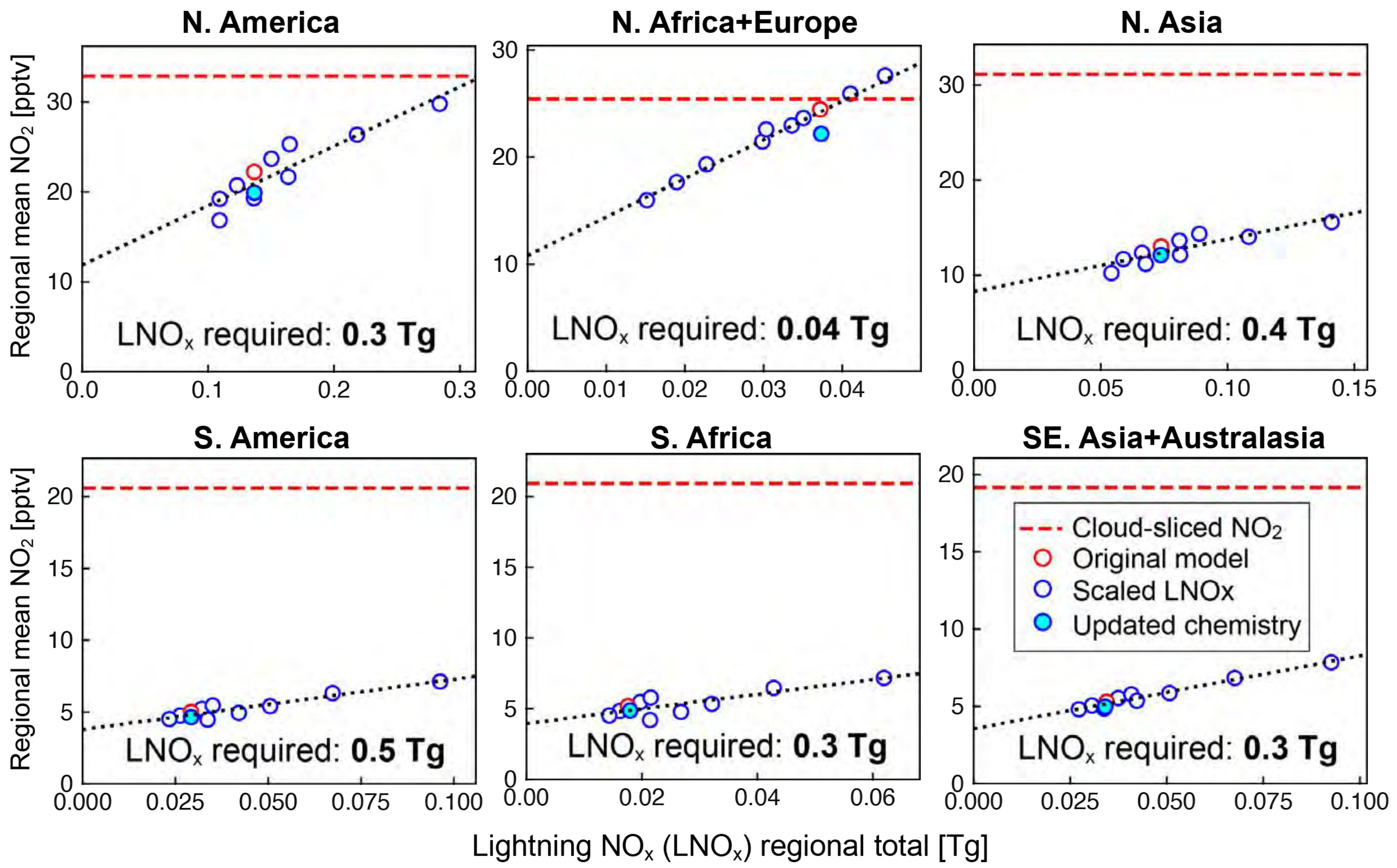
Observed versus modelled June-August upper tropospheric regional mean NO_2 **over land**



Original model emissions of **1.4 Tg NO**, whereas **2.7 Tg NO** required to match cloud-sliced NO_2

Regional sensitivity to lightning NO_x emissions

Observed versus modelled June-August upper tropospheric regional mean NO_2 **over the ocean**

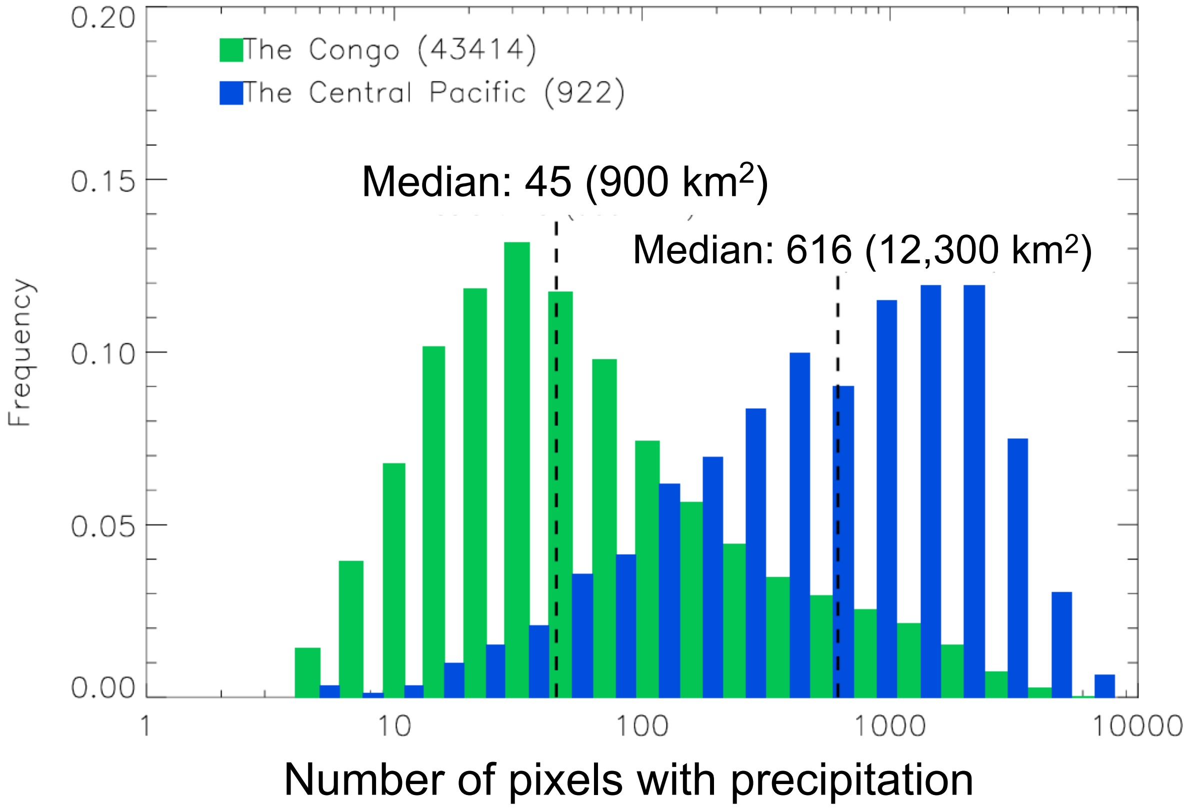


Original model emissions of **0.3 Tg NO**, whereas **1.9 Tg NO** required to match cloud-sliced NO_2

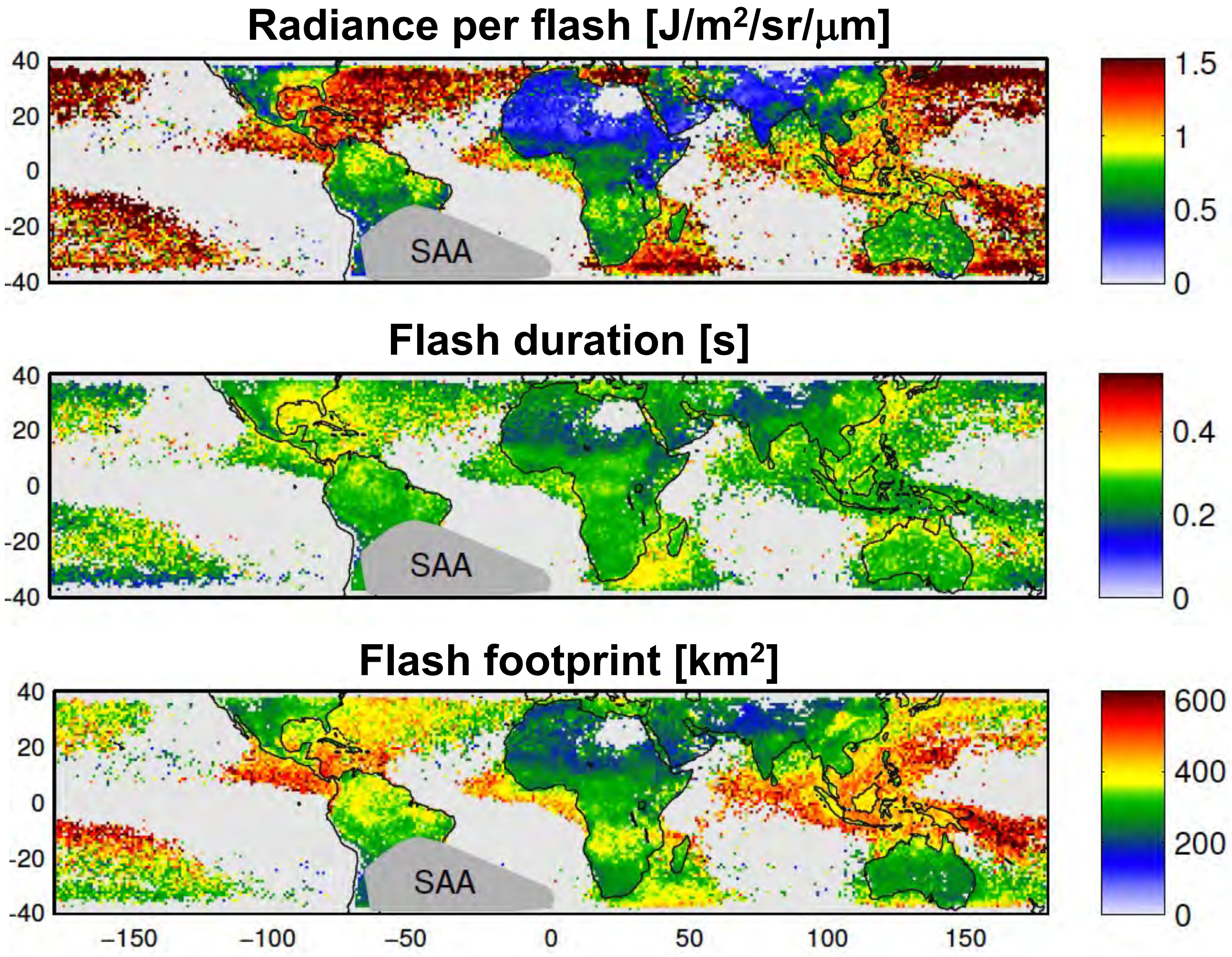
Lightning characteristics over the ocean and over land

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]



[Beirle et al., 2014]

Concluding Remarks

Cloud-sliced profiles of 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 NO_2 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

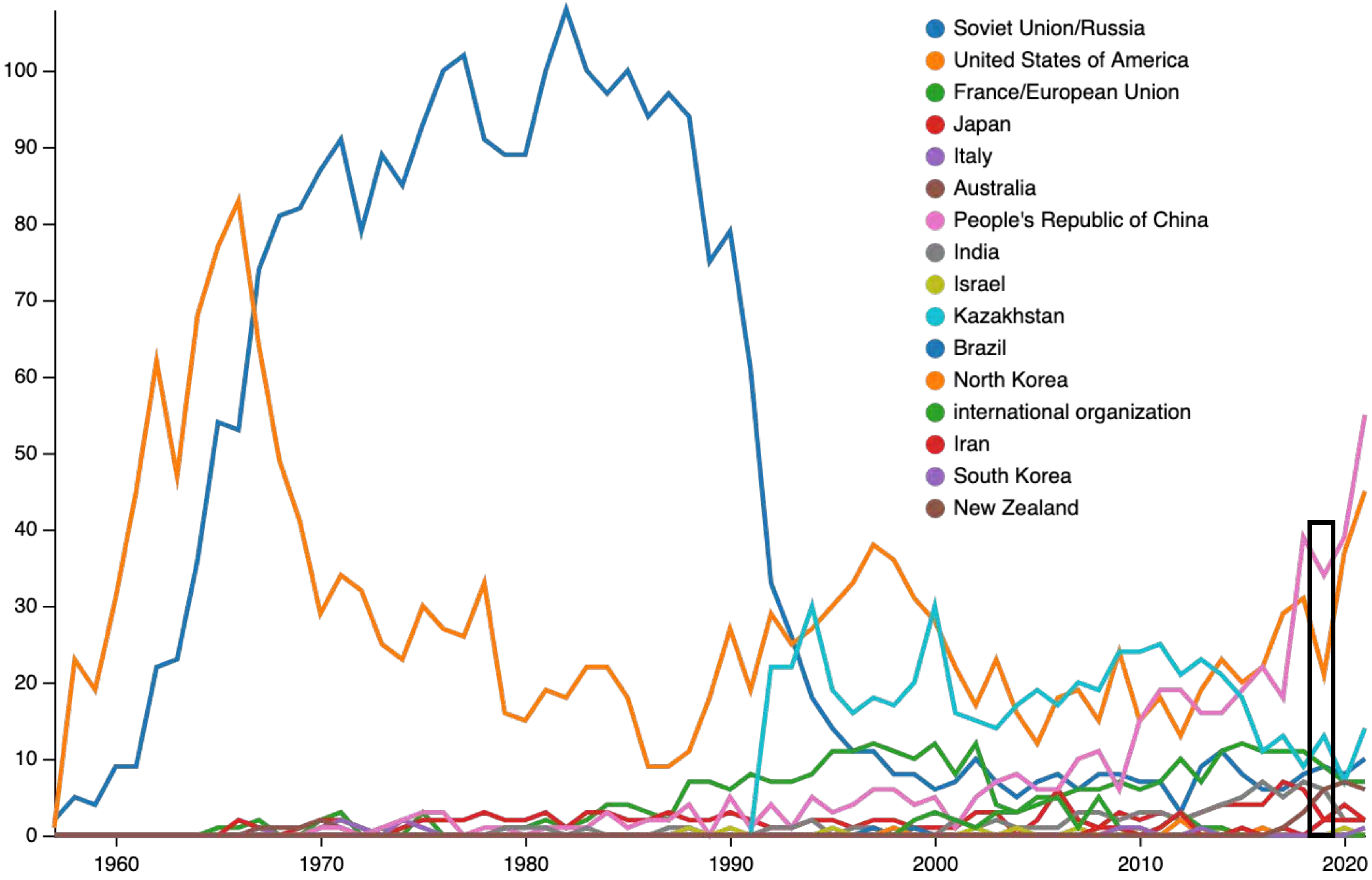
Environmental impact of the modern space sector

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

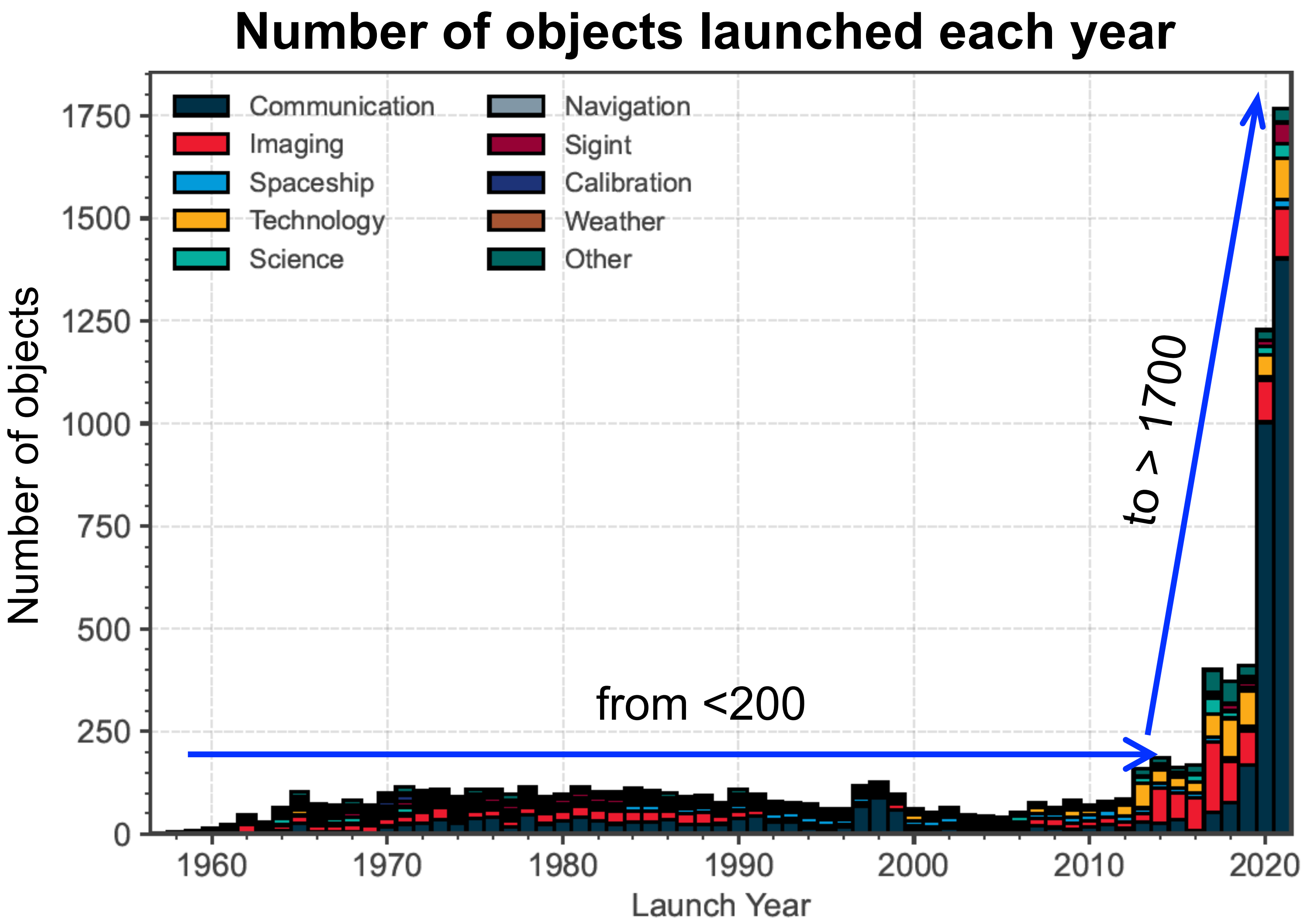


Image from ESA's Annual Space Environment Report, 2022

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

Air pollutant emissions from rocket launches

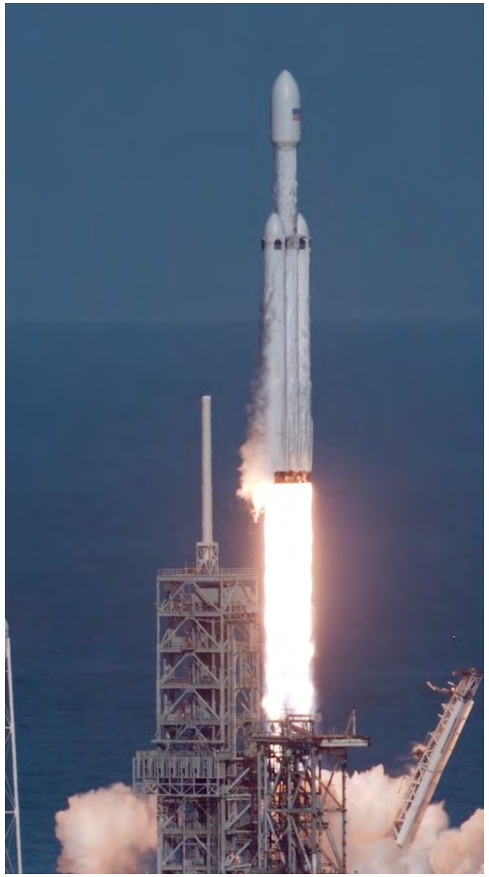
Solid

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

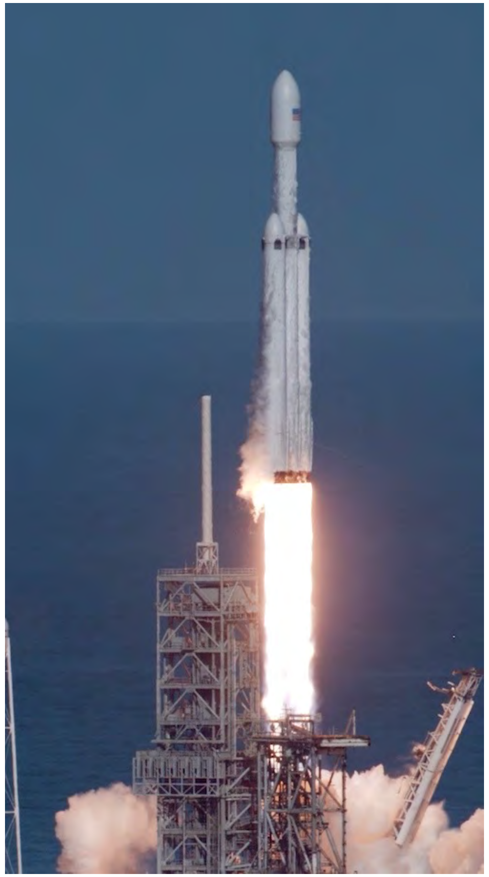
Solid

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

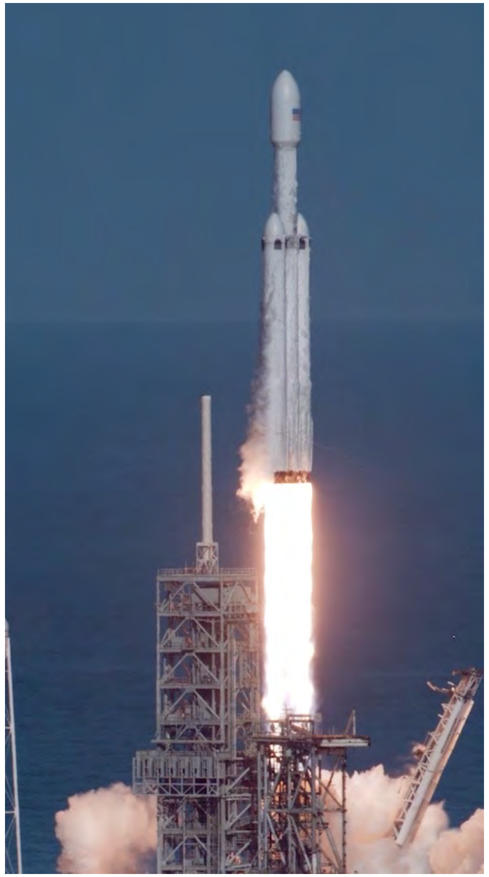
Solid

NO_x
$HCl+Cl$
Al_2O_3
H_2O
BC

Hypergolic

NO_x
H_2O
BC

Kerosene



NO_x
H_2O
BC

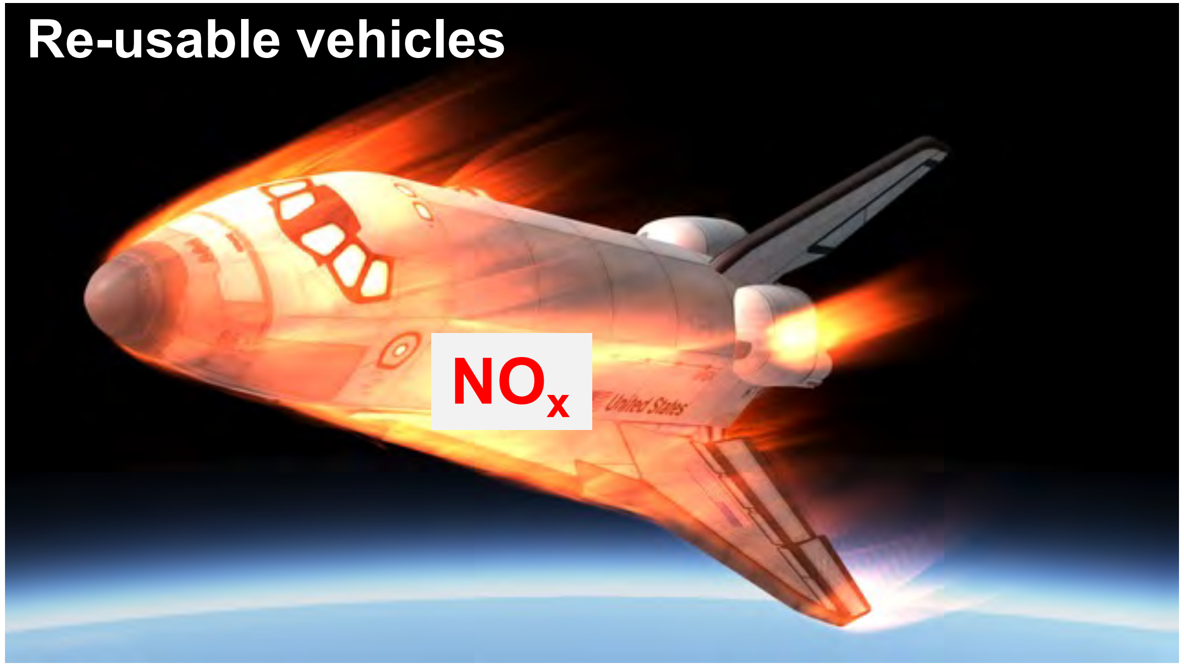
Cryogenic

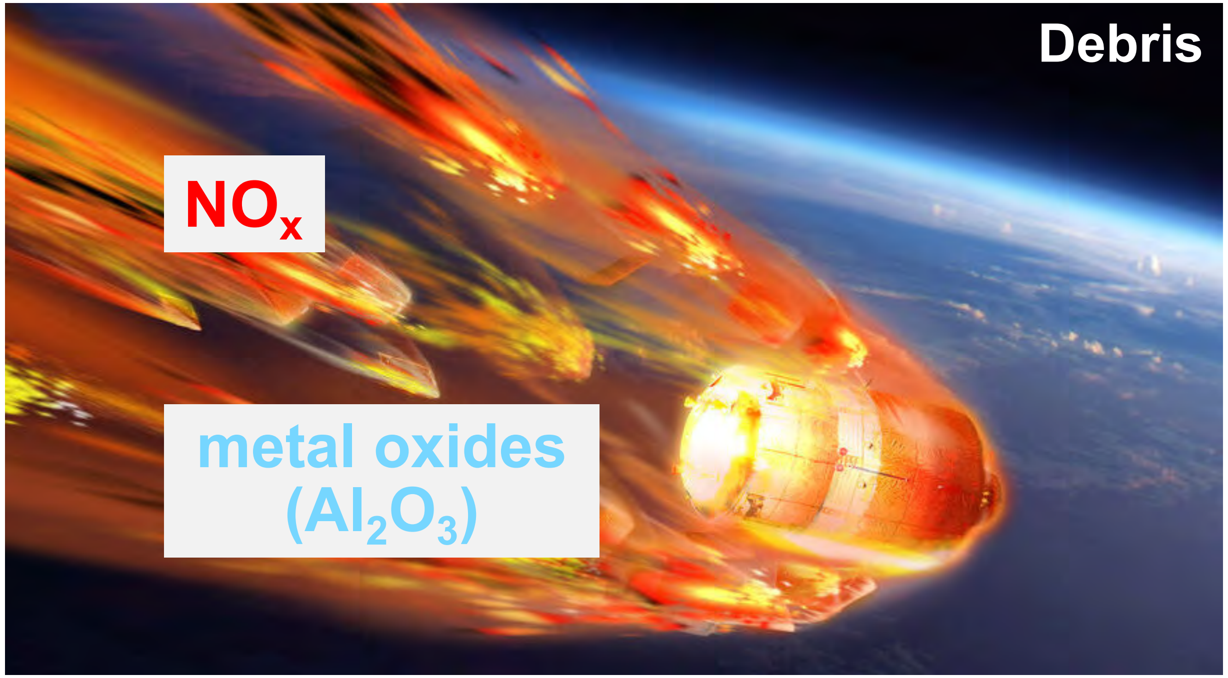
NO_x
H_2O

Ozone depletion

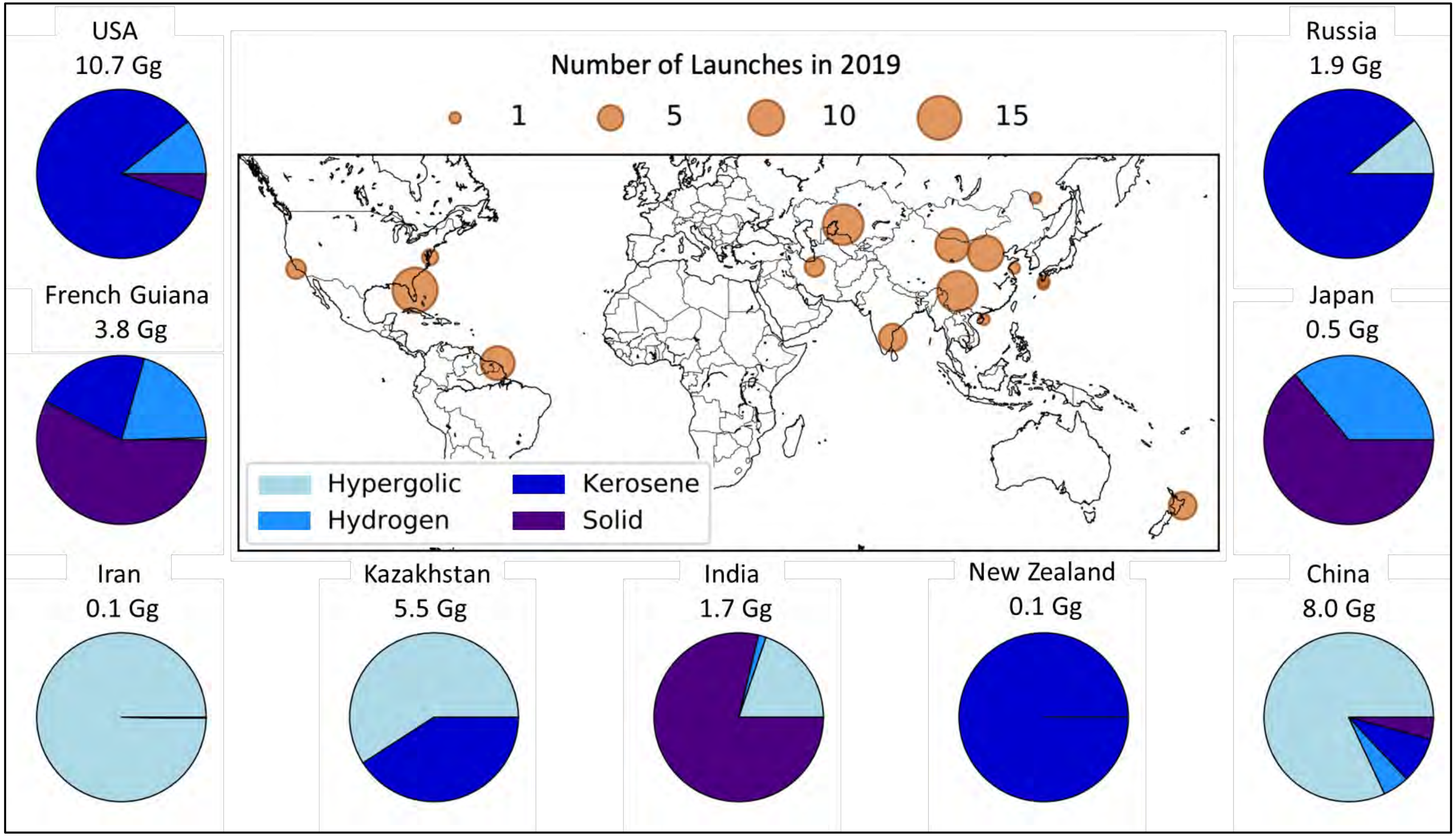
Air pollutant emissions from re-entry

2-40 Gg NO_x per year





Calculate and map a single year of emissions



Annual Emissions:

H_2O: 11 Gg
BC: 0.5 Gg
Al_2O_3: 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

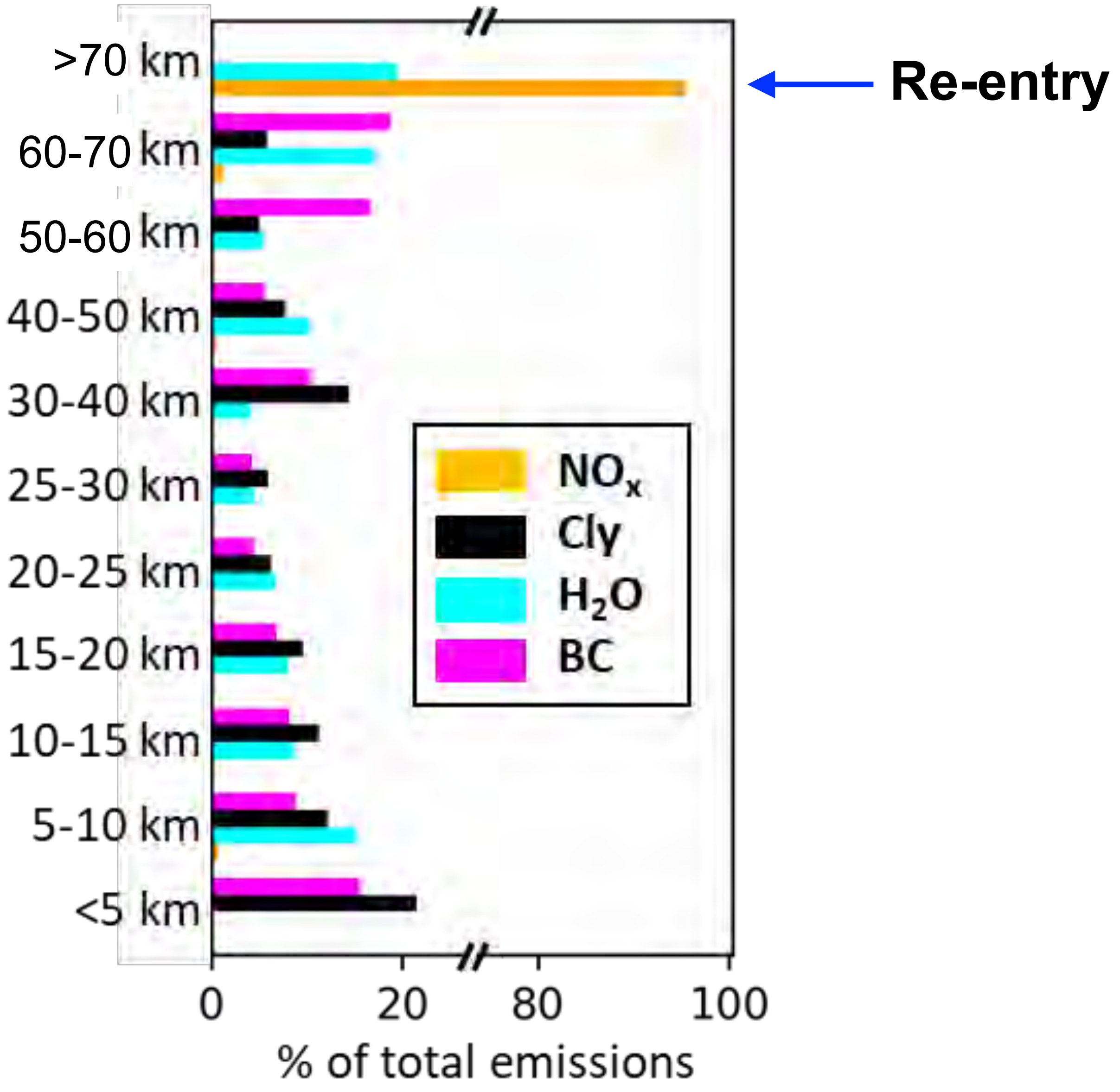
Reaches 135 in 2021. Already 148 in 2022.

Incorporate these in GEOS-Chem

GEOS-Chem extends to **80 km**

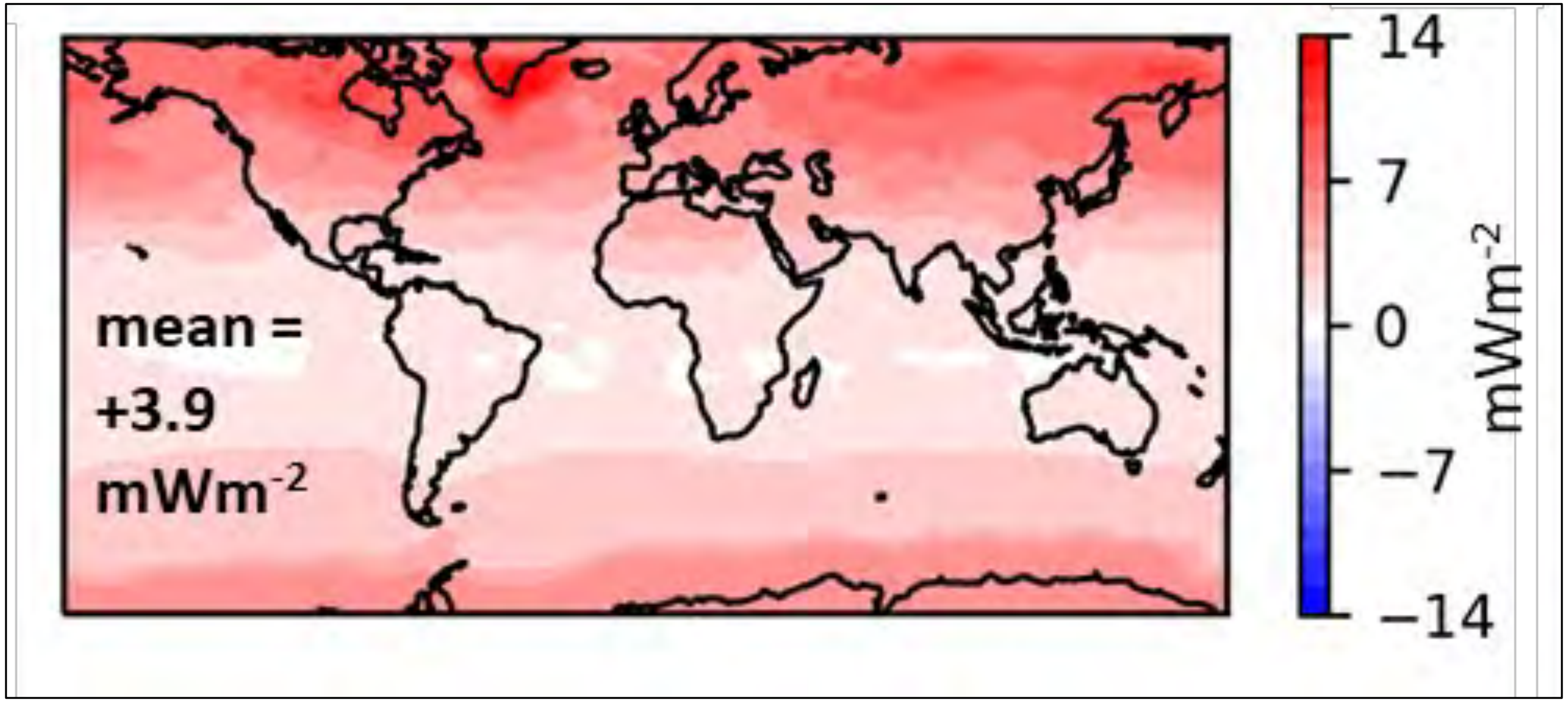
<u>Stratosphere & mesosphere:</u>
lifetime >2 years
(*gravitational settling*)

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

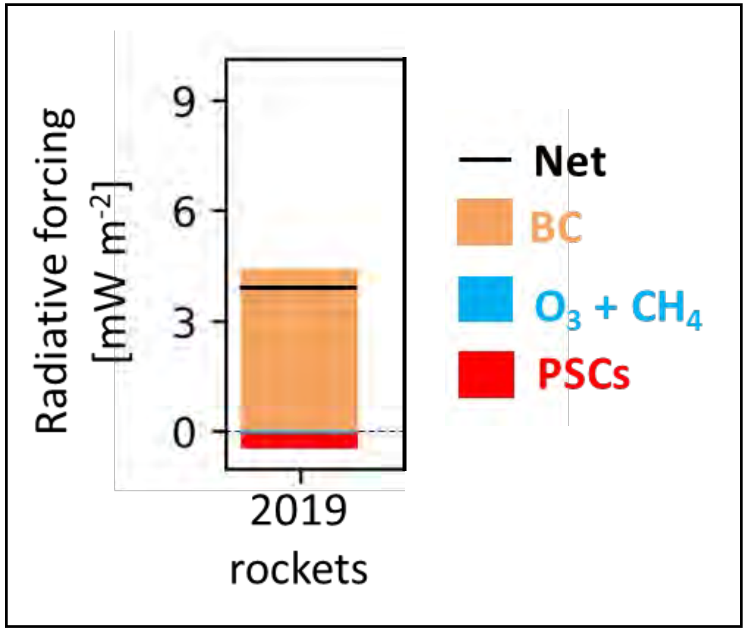


Radiative forcing due to black carbon emissions

After 10 years of emissions assuming modest growth



Mostly due to BC

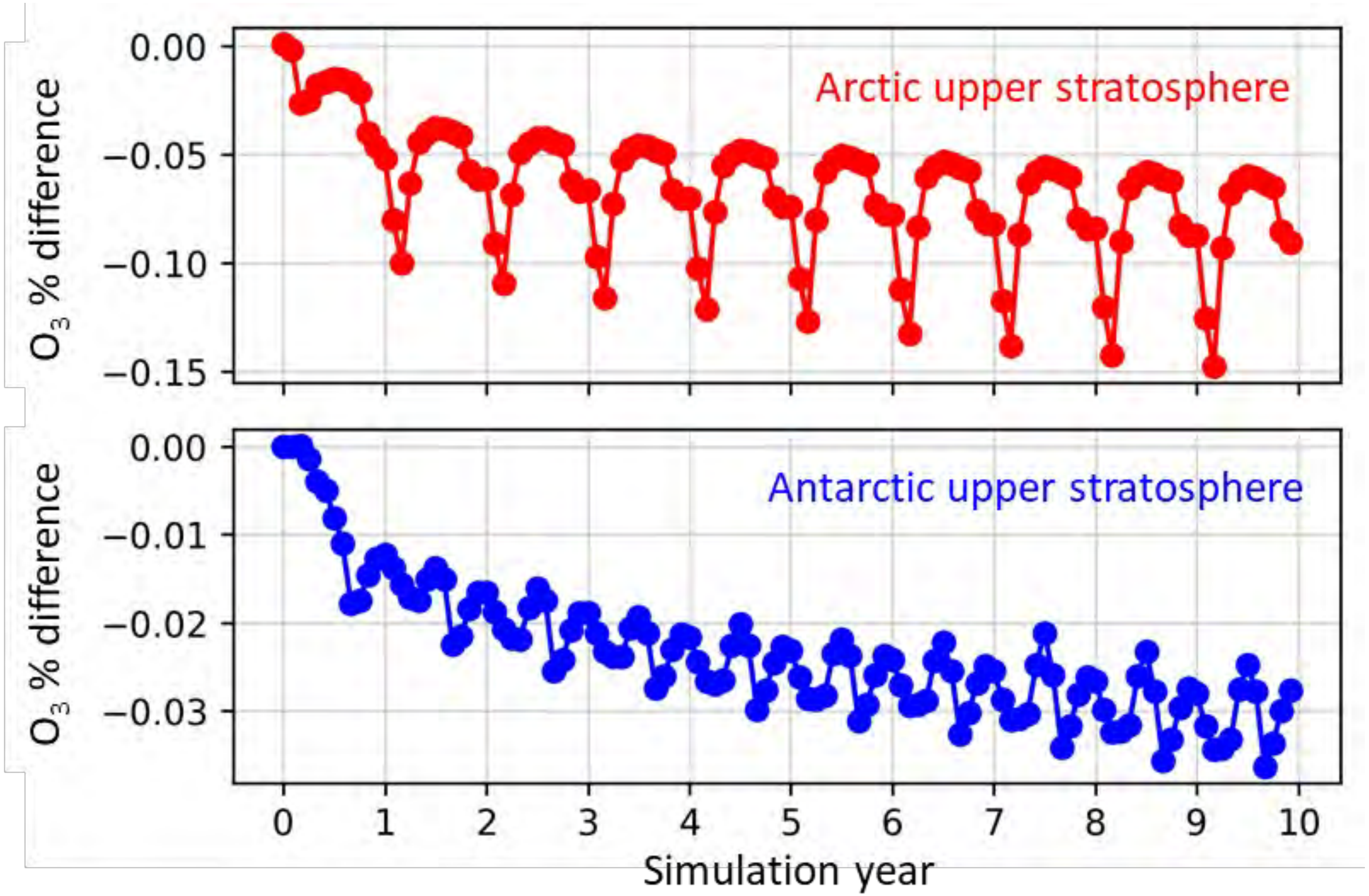


Rockets ~3% of BC radiative forcing from all anthropogenic sources, but only 0.01% of emissions.

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

SpaceX StarShip

Raptor

~200 tonnes

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

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.