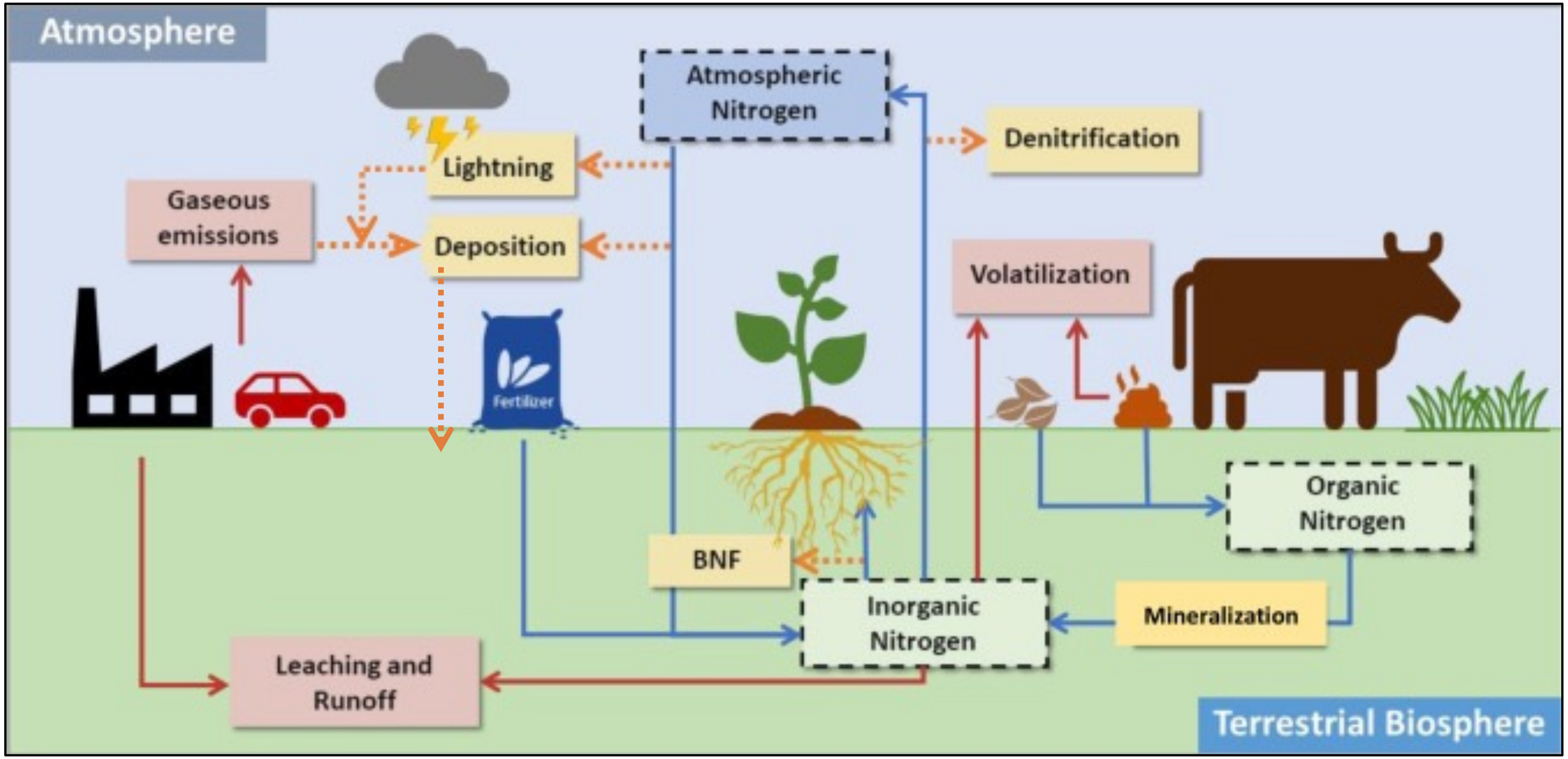
Deriving new and exploiting existing remote sensing observations to better understand sources and abundances of natural and anthropogenic reactive nitrogen

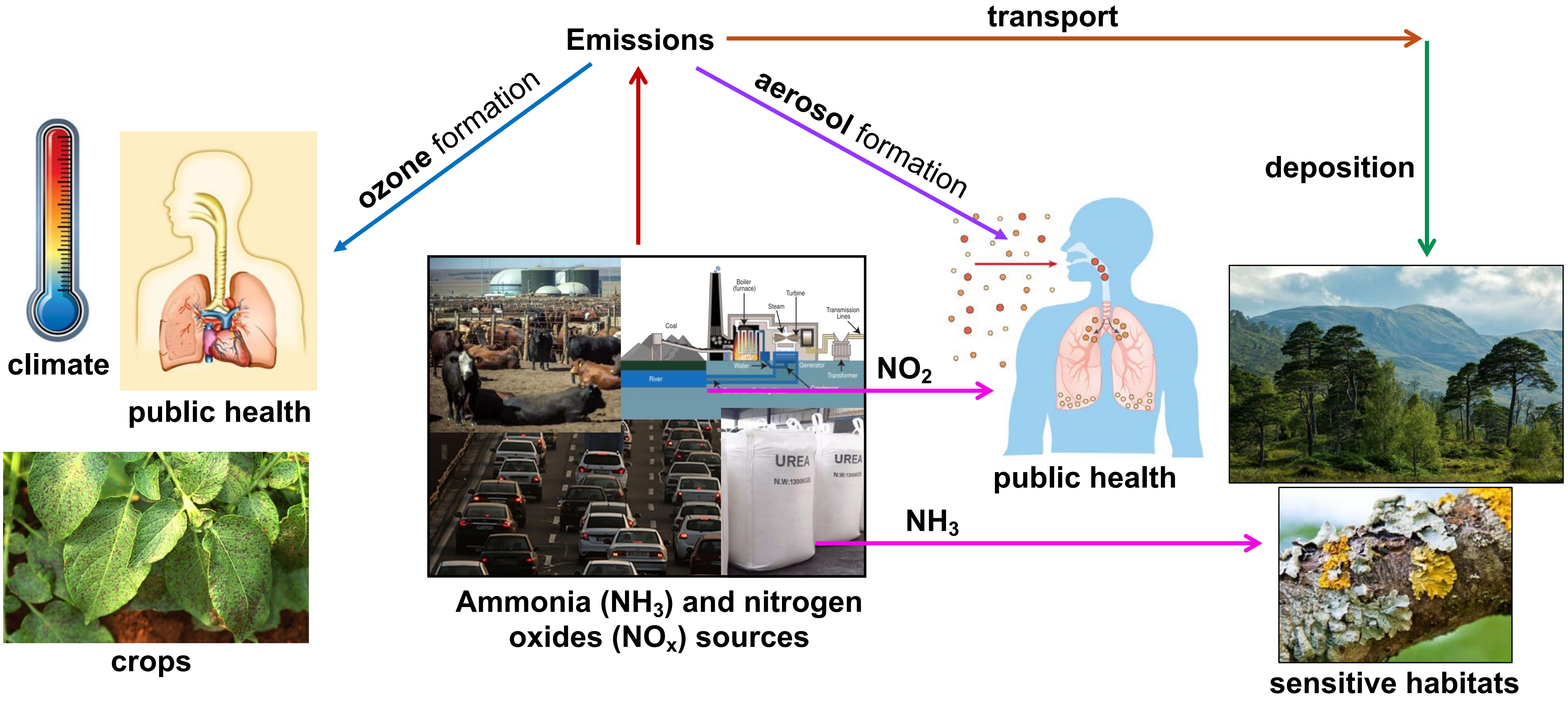
Eloise Marais, Lab website: https://maraisresearchgroup.co.uk/



[Image source: Khattar et al., 2023]

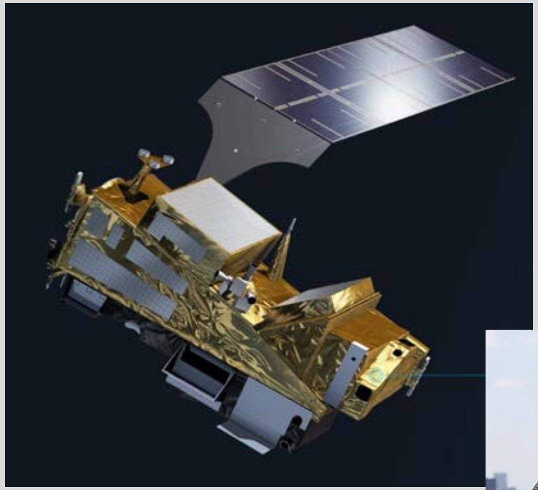
Royal Society UK-China Bilateral Meeting, 22-23 May 2024

Environmental Impacts of Atmospheric Reactive Nitrogen



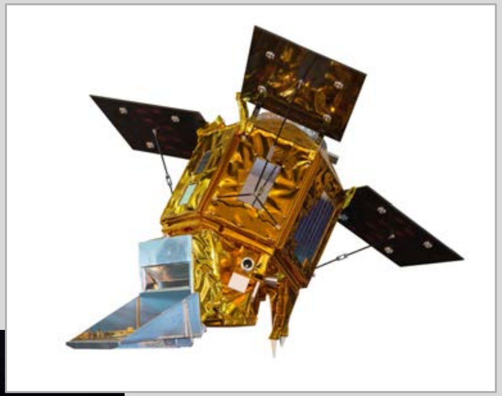
Remote Sensing Observations Used by the Group

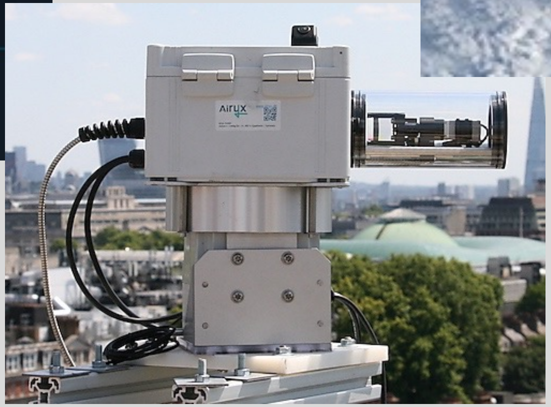
IASI: NH_3



LIS: lightning properties

TROPOMI: NO_2





MAX-DOAS: HONO

CrIS: NH_3

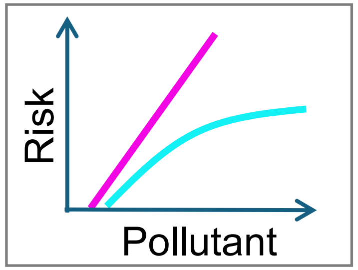
We also use:

in-situ measurements

GEOS Chem

models

python

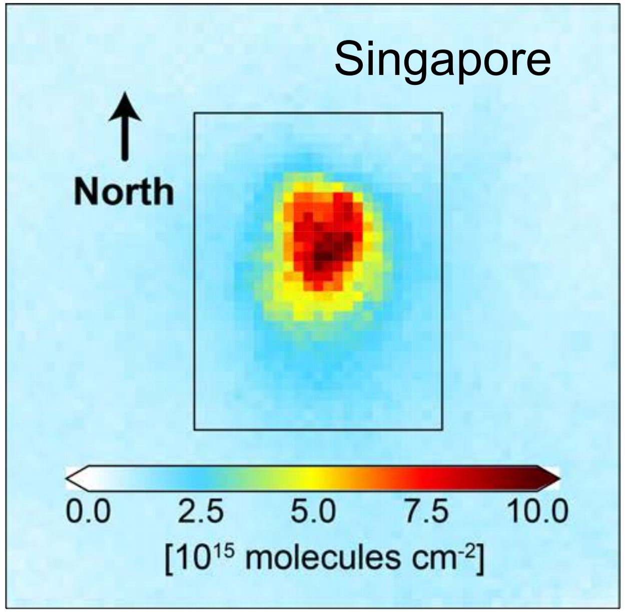


exposure-risk curves

Top-down Estimate of NO_x Emissions

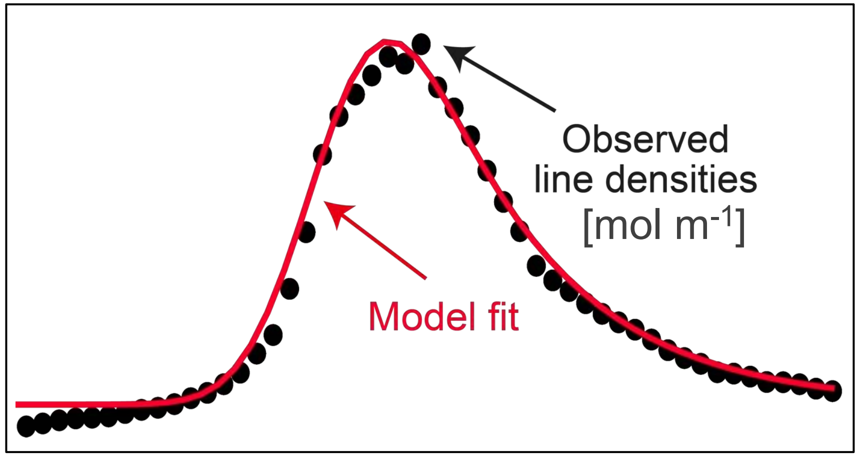
Derive NO_x emissions of isolated hotspots viewed by UV-visible space-based sensors

Wind rotated TROPOMI NO_2



Model fit to yield best-fit parameters

Across-wind sum of vertical columns



City NO_x Plume Emissions

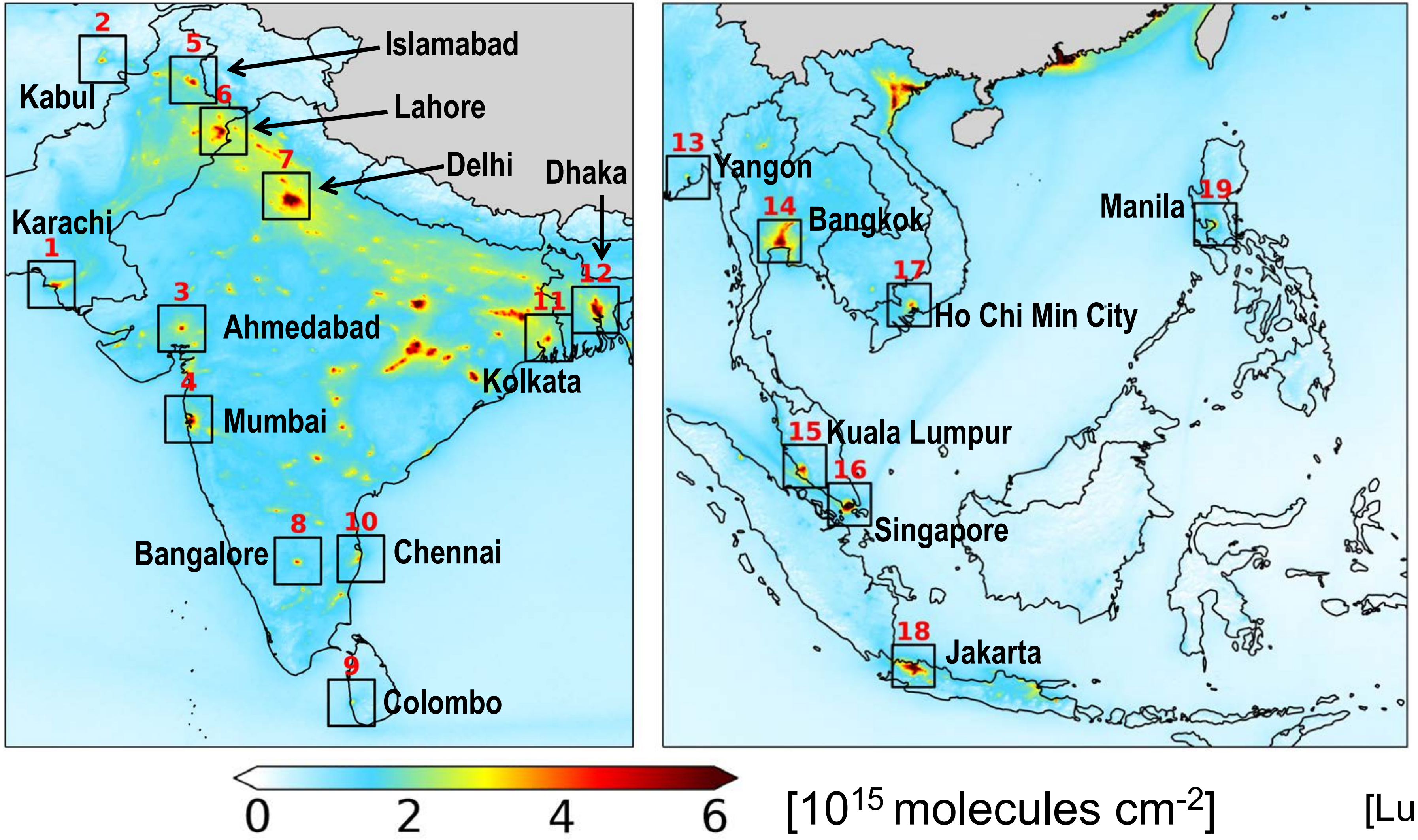
112 mol NO_x s^{-1}

Enhance success of well-established method by defining many (54) rather than one sampling area

Target cities in understudied regions of the world (South and Southeast Asia, Sub-Saharan Africa)

Cities Targeted in South and Southeast Asia

Annual (2019) mean TROPOMI NO_2 at ~5 km resolution



[10^{15} molecules cm^{-2}]

[Lu et al., *in review*]

19 isolated city hotspots selected (other hotspots: industries, power plants or not isolated)

Derive City-Specific NO_x Emissions and Fit Uncertainties

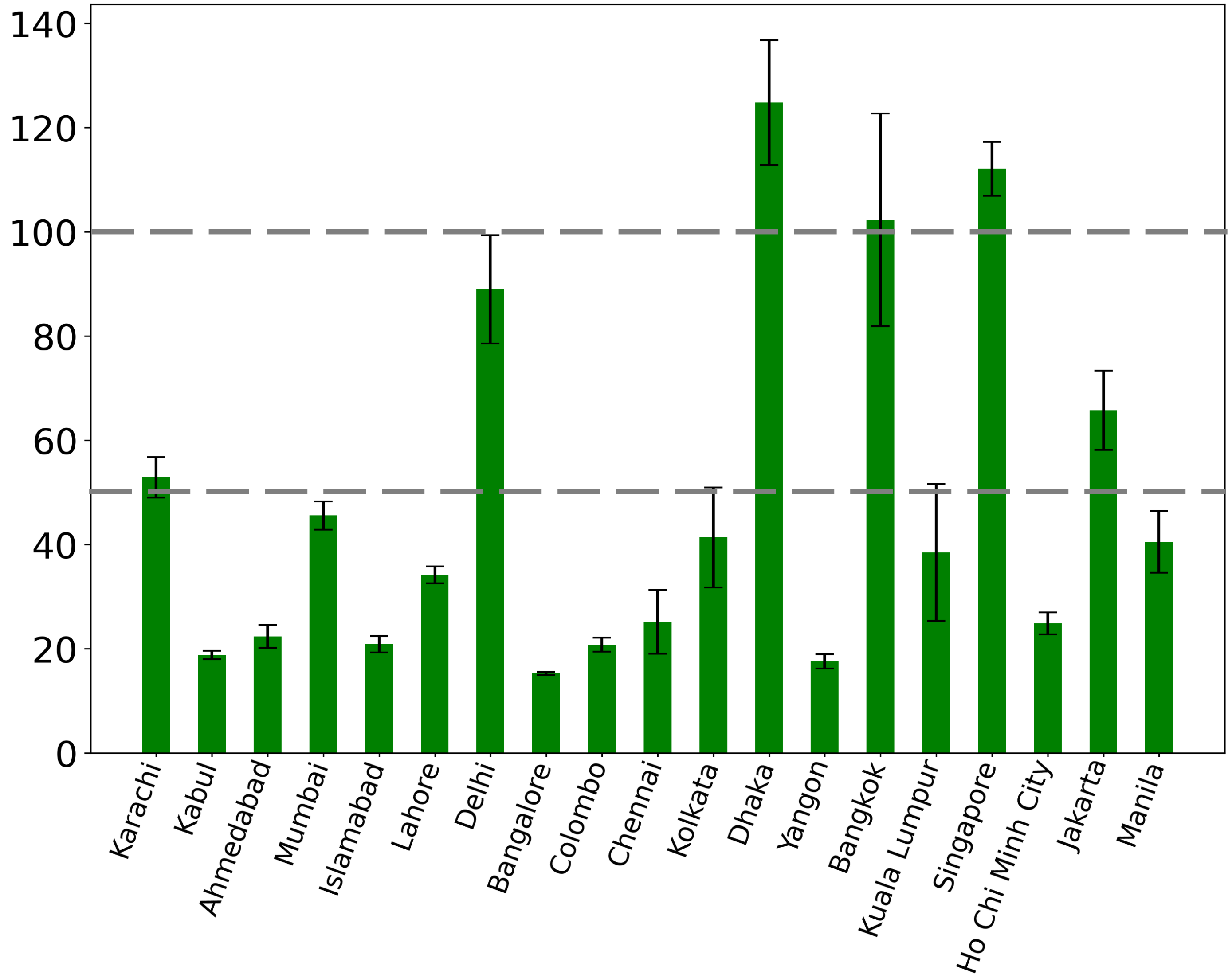
Relative error range:

4-34%

Range of past studies:

10-40%

City NO_x emissions for 2019 [mol/s]



>100 mol/s: Dhaka, Bangkok, Singapore (> 145 Gg NO_2 a^{-1})

50-100 mol/s: Delhi, Jakarta, Karachi

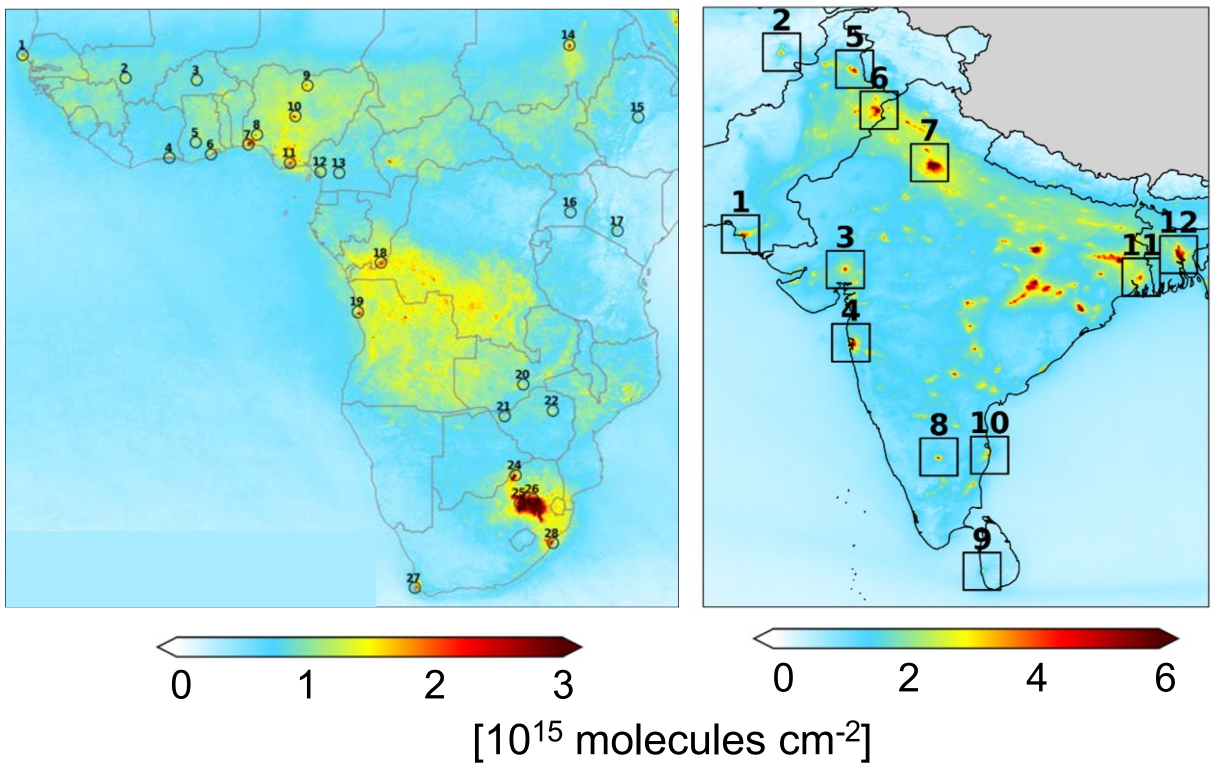
Most cities < 50 mol/s

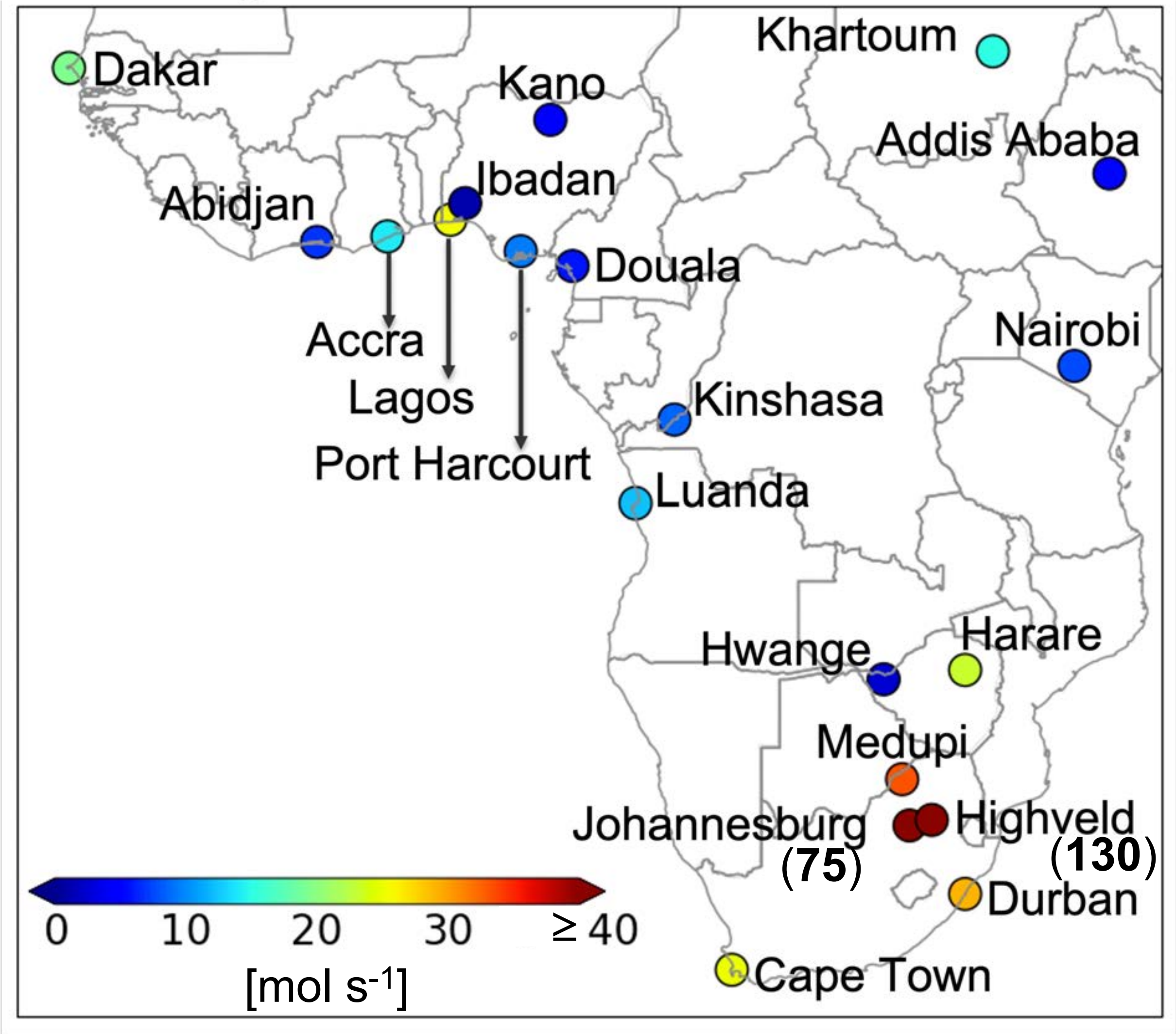
[Lu et al., *in review*]

NO_x emissions from mean of individual successful fits. Standard deviation provides fit error.

NO_x Emissions of Hotspots in Sub-Saharan Africa

Annual mean TROPOMI NO_2 for 2019





Emissions obtained for 20 hotspots:
18 cities, 1 power station, 1 industrial area

South Africa hotspot emissions far greater (28-130 mol/s) than others (<28 mol/s)

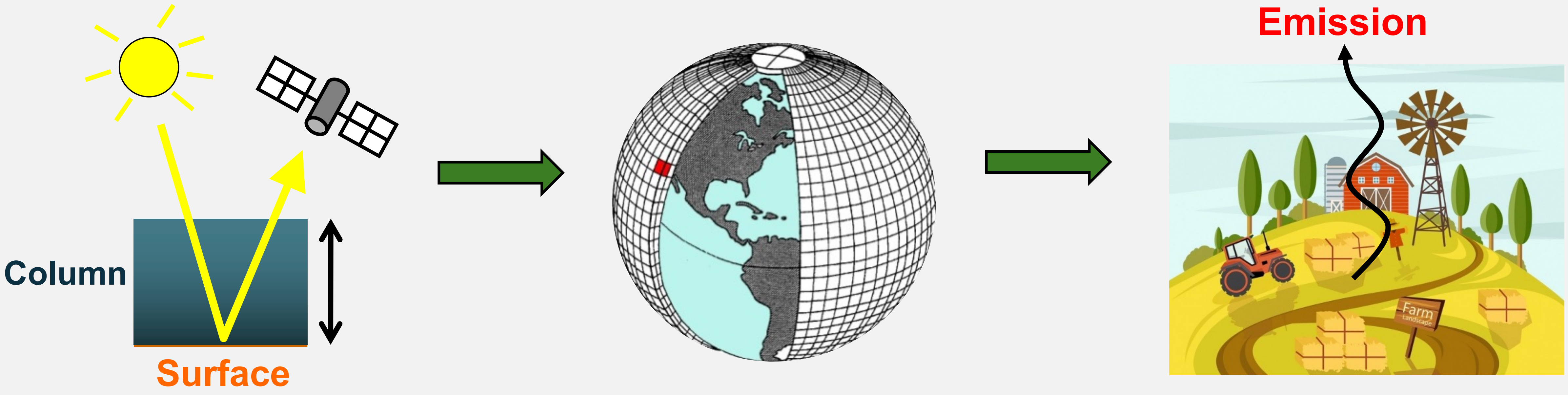
[Wei et al., *in progress*]

Top-down Estimate of Ammonia (NH_3) Emissions

Simple mass balance approach:
Convert atmospheric **column concentrations** to surface **emissions** by relating the two with a **model**

ABUNDANCES → **Conversion Factor** → **EMISSIONS**

Satellite column densities | **Model Concentration-to-Emission Ratio** | **Satellite-derived Surface Emissions**

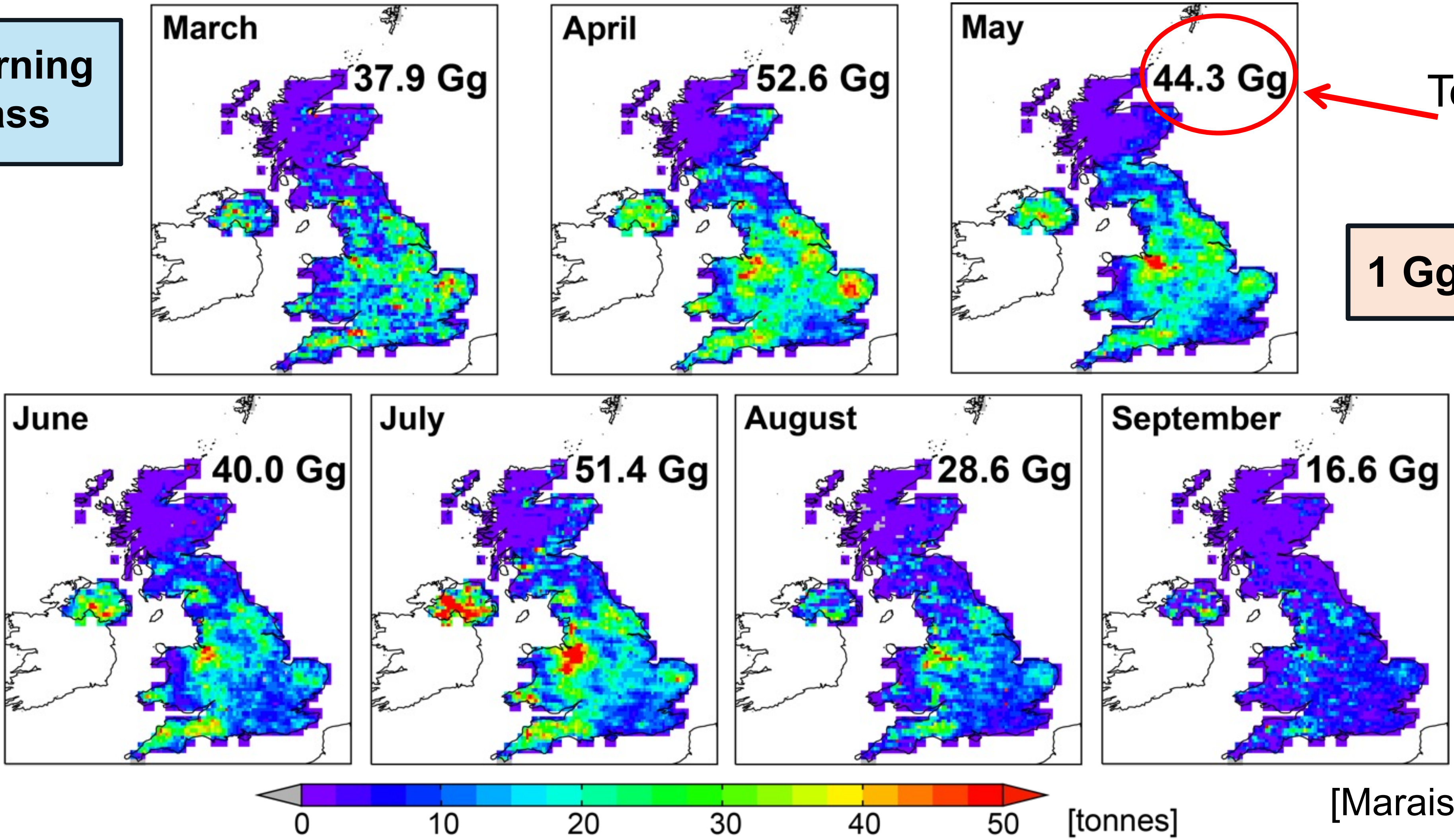


This approach possible as NH_3 has a relatively short lifetime (2-15 hours) at or near sources

High-Resolution Agricultural NH_3 Emissions for the UK

Focus on **Mar-Sep** when warm temperatures and clearer conditions increase sensitivity to surface NH_3

IASI: morning overpass



Total monthly emissions

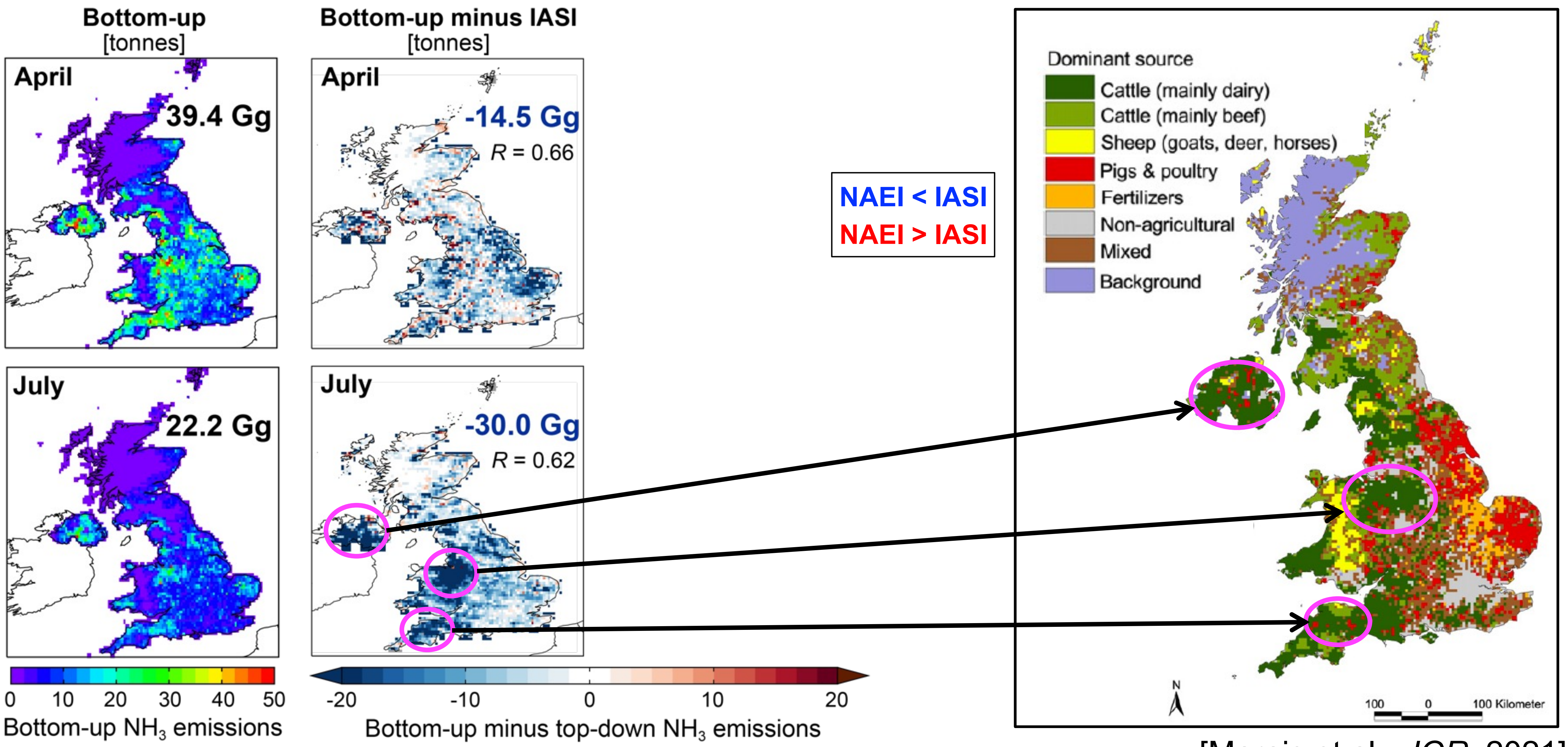
1 Gg = 1 kilotonne

[Marais et al., *JGR*, 2021]

Monthly emissions for March-September from **IASI**-derived estimates sum to **271.5 Gg**

Satellite vs inventory NH_3 emissions: spatial distribution

Comparison of months with peak emissions according to IASI and CrIS (April and July)

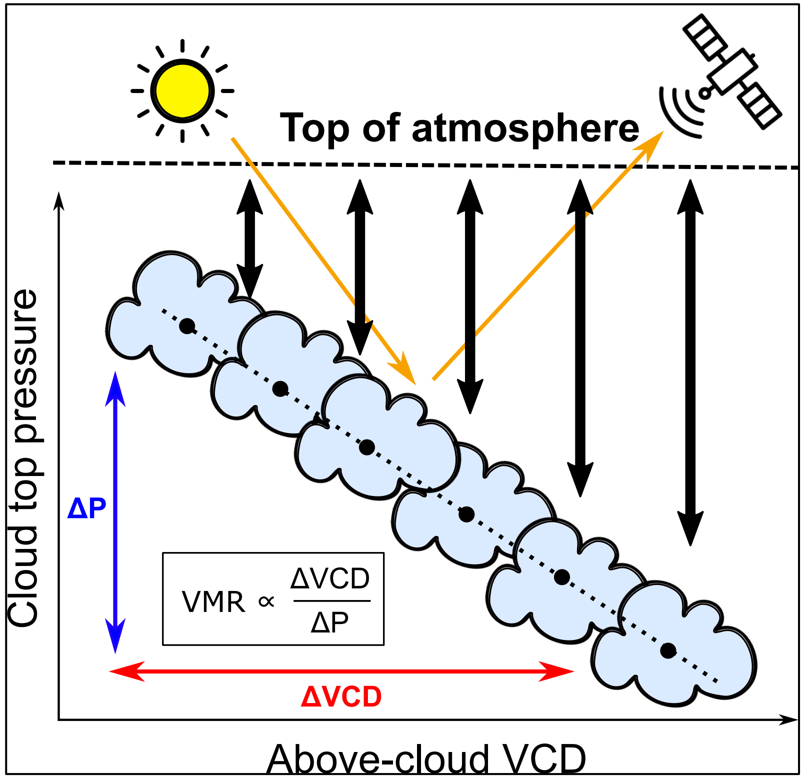


[Marais et al., *JGR*, 2021]

Large July difference over locations dominated by dairy cattle. Inventory is 27-49% less than the satellite values.

Derive New Datasets: Vertical Profiles of Tropospheric NO_2

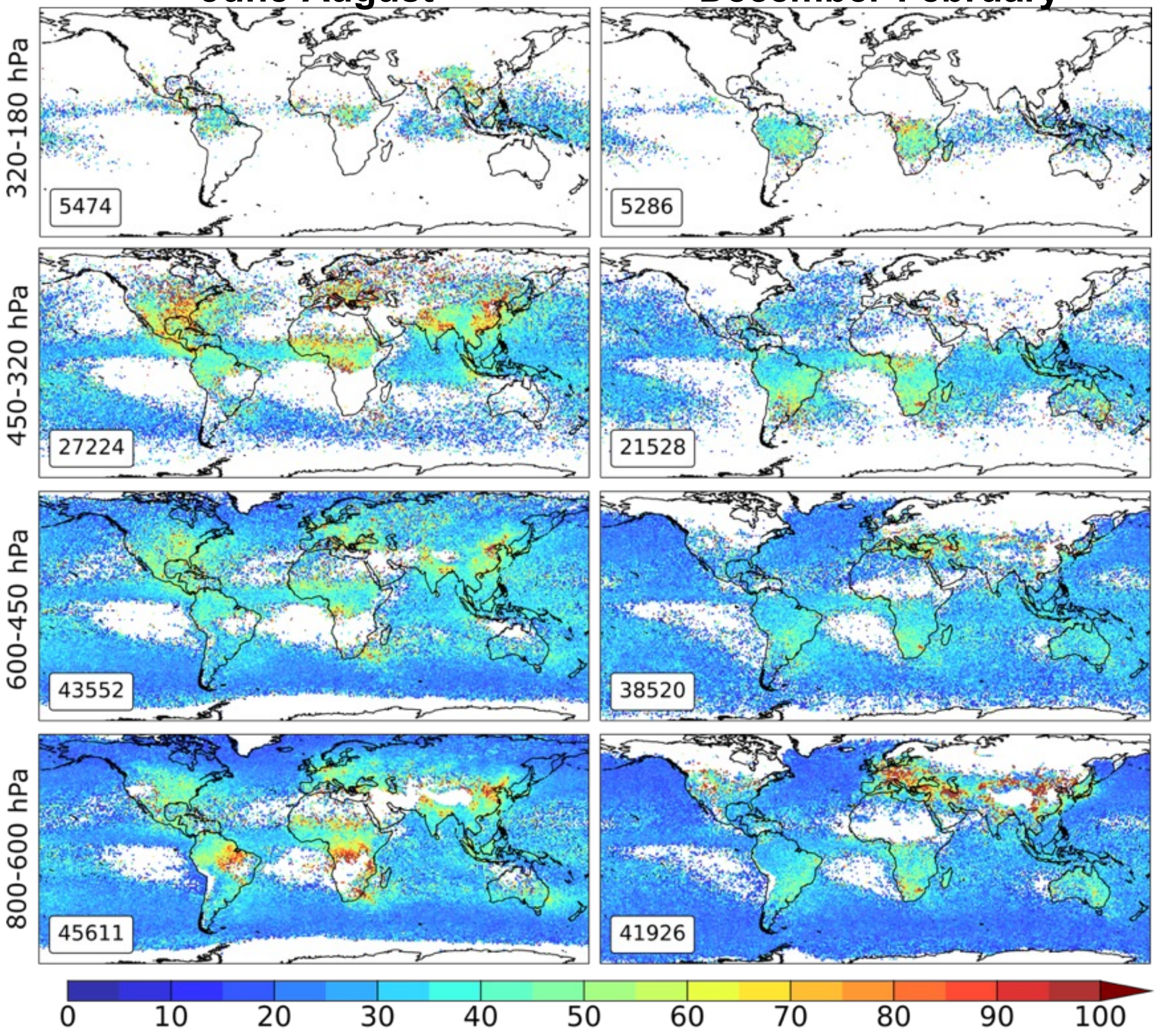
Cloud-slice TROPOMI NO_2 above optically thick clouds



Addresses absence of routine observations

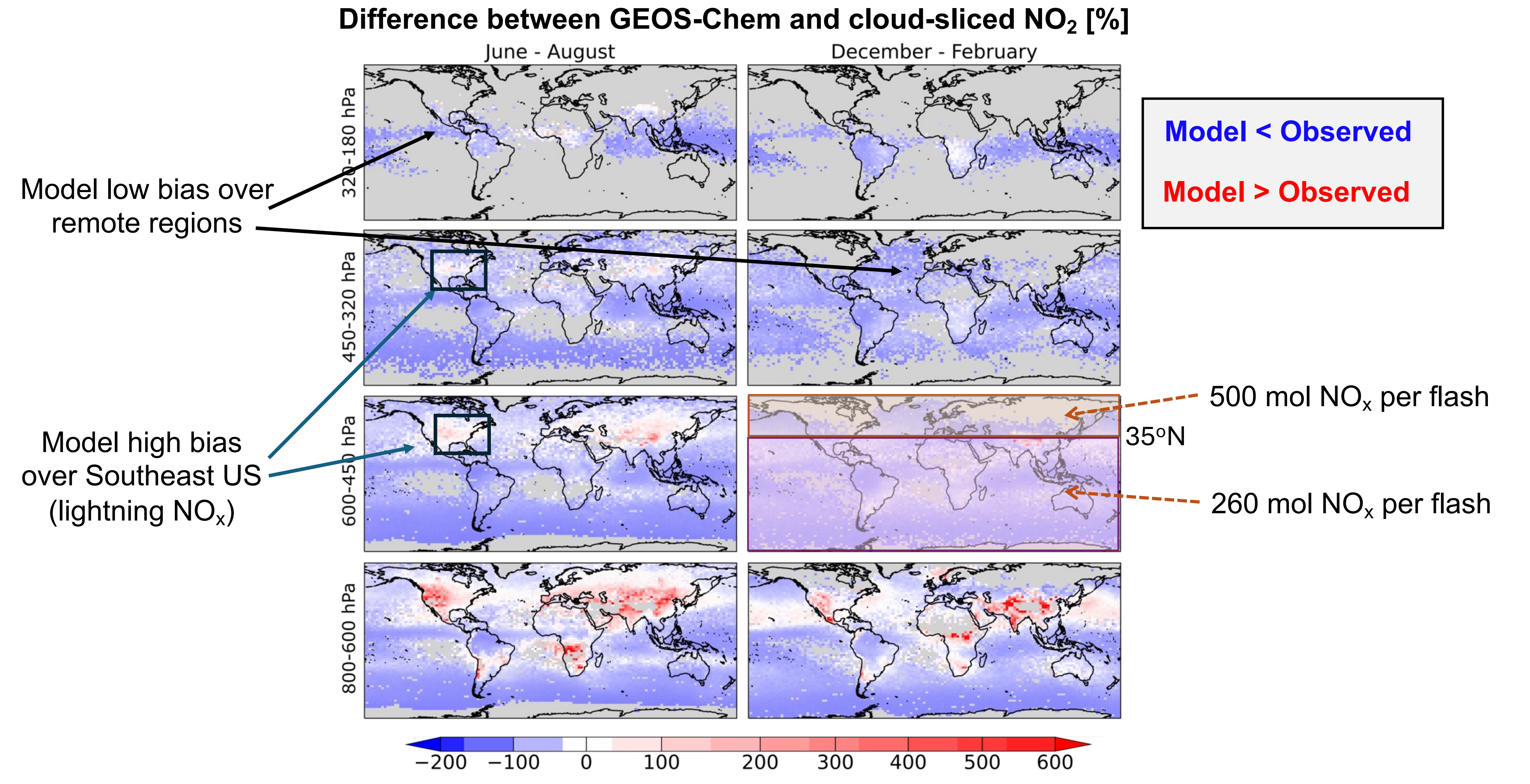
Consistent with in situ observations from NASA DC-8 aircraft campaigns

Seasonal multiyear mean NO_2 mixing ratios [pptv]



[Horner et al., *submitted*, ACP]

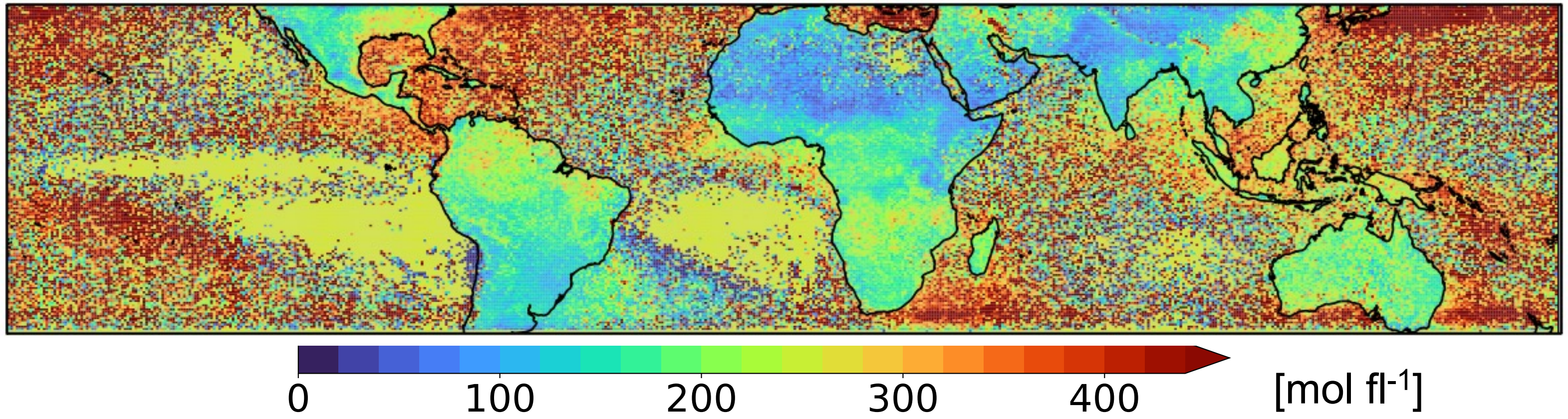
Use New Datasets to Assess State of Knowledge



Time- and Space-Resolved Lightning NO_x Production

Use satellite observations of lightning flash energy to calculate hourly gridded production rates to GEOS-Chem lightning NO_x emission inventory

Gridded lightning NO_x production rates

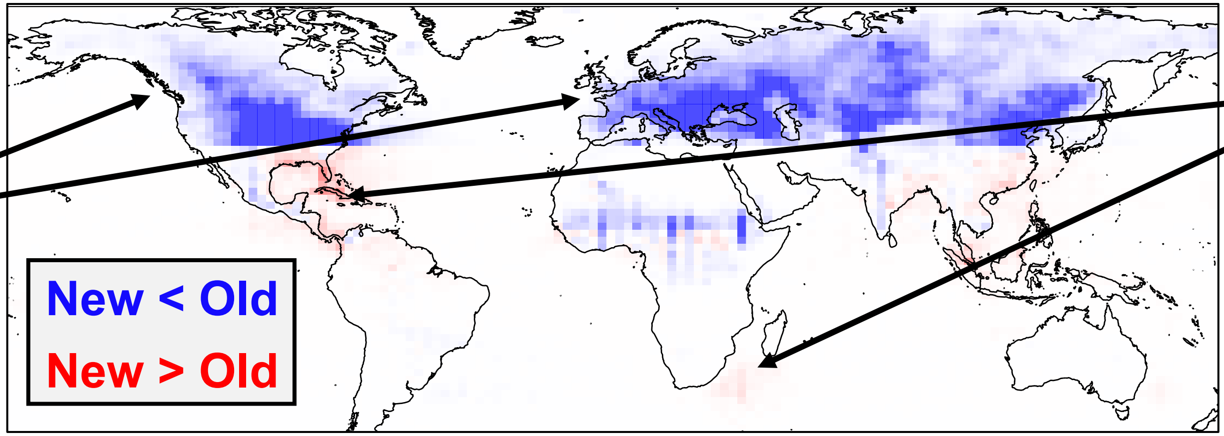


Ocean > land well known

Reason seems to be in dispute

Effect of gridded lightning NO_x production on emissions for single month (June)

Large decline across northern midlatitudes

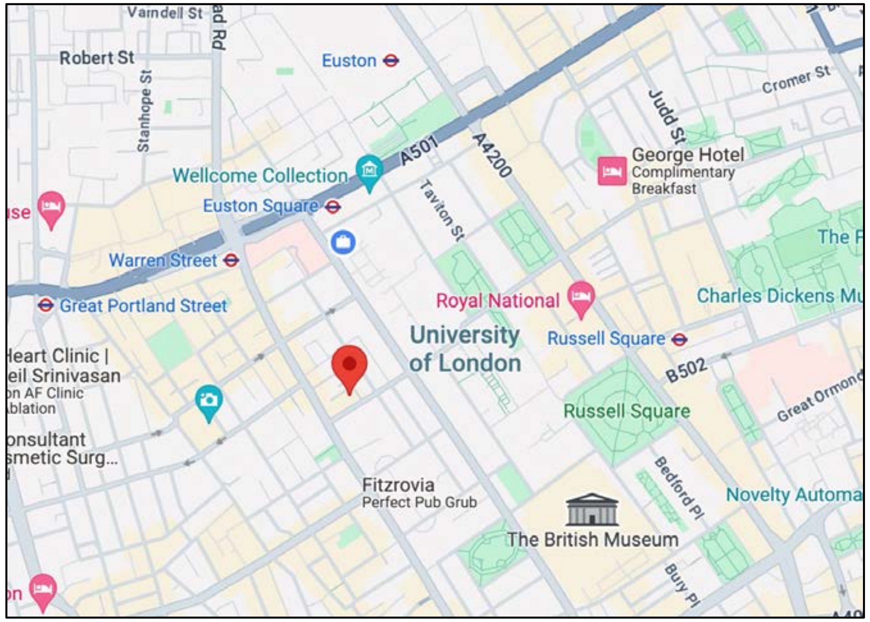


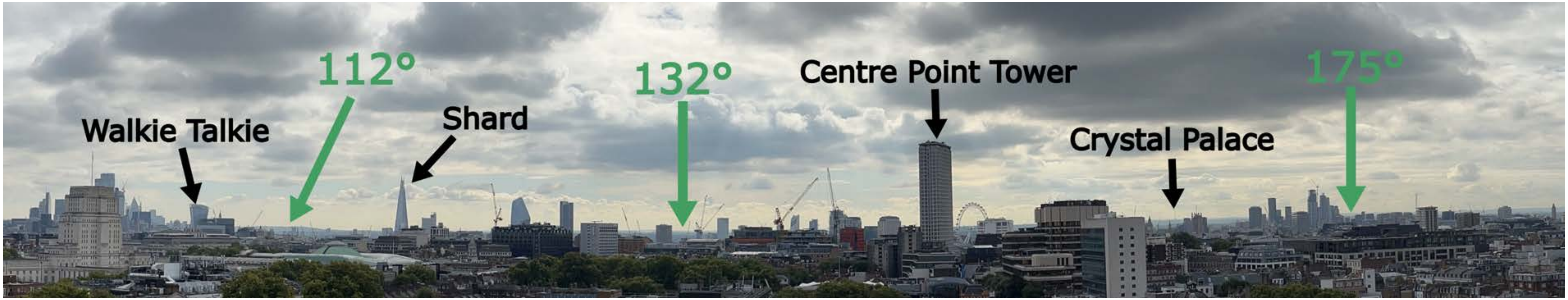
Small increases over oceans

[Horner et al., *in progress*]

Next step is to implement emissions in GEOS-Chem to test effect on tropospheric NO_x and ozone

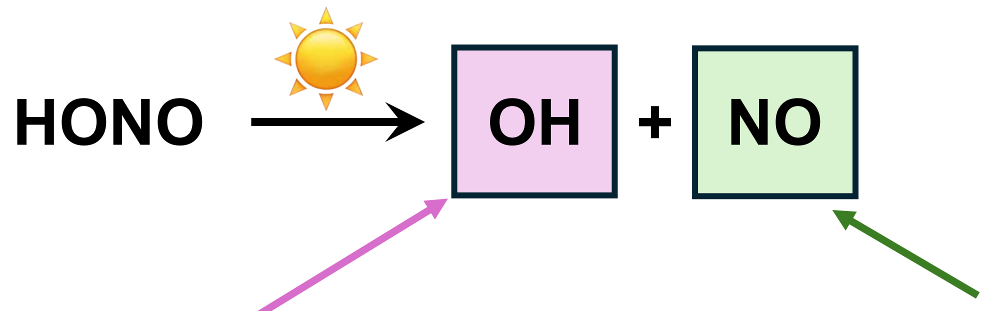
Observations of Nitrous Acid (HONO) in Central London





[Ryan et al., ACP, 2023]

Observations of Nitrous Acid (HONO) in Central London

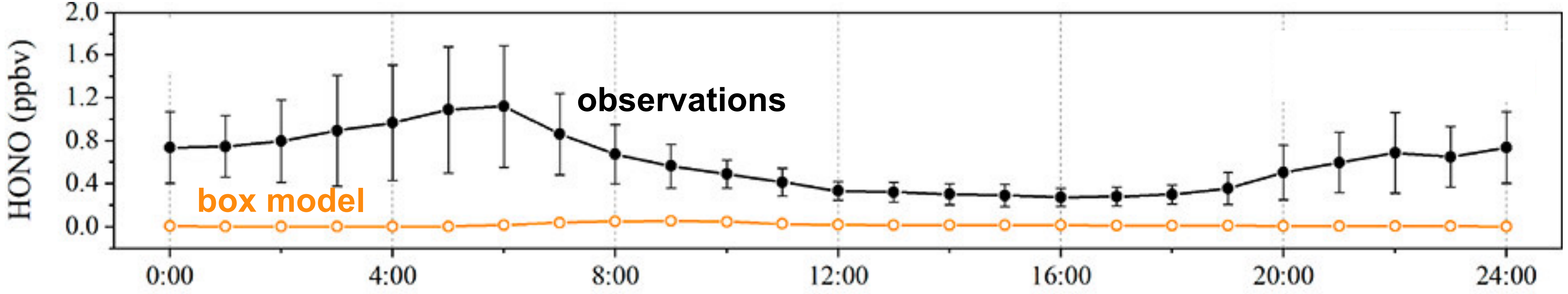


Source of the main atmospheric oxidant

Recycles NO_x back to the atmosphere

Best understanding of HONO (models) routinely fails to reproduce reality:

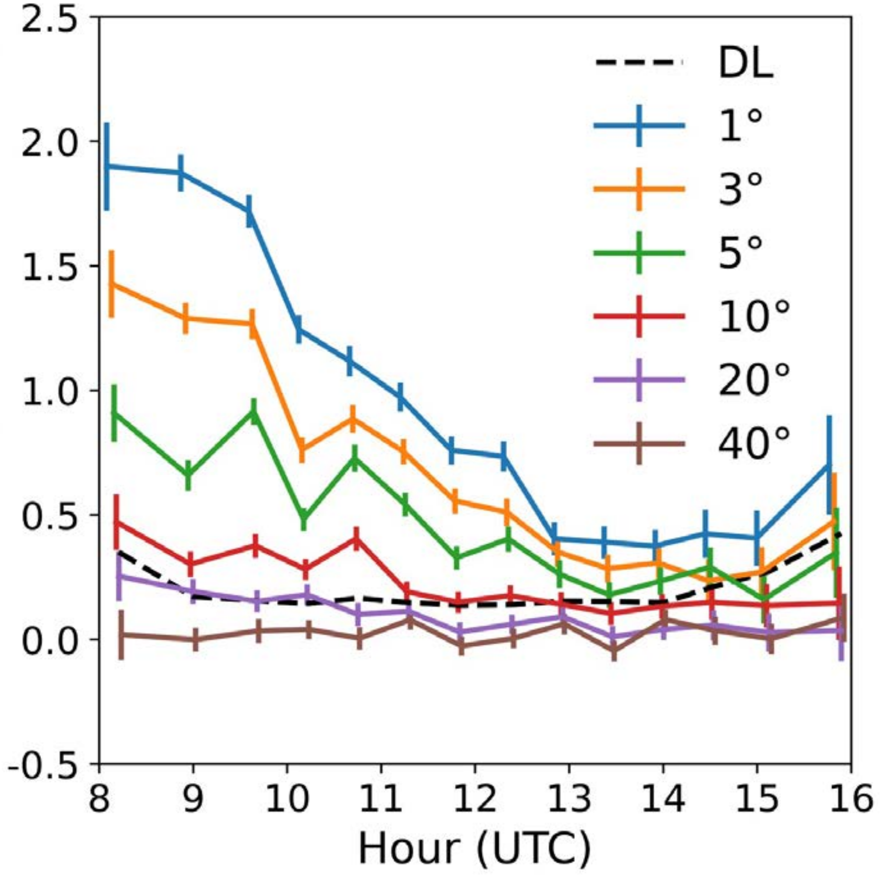
Diel profile of urban HONO in Tai'an, Shandong Province in June-July 2018



[Liu et al., 2022]

Observations of nitrous acid (HONO) in Central London

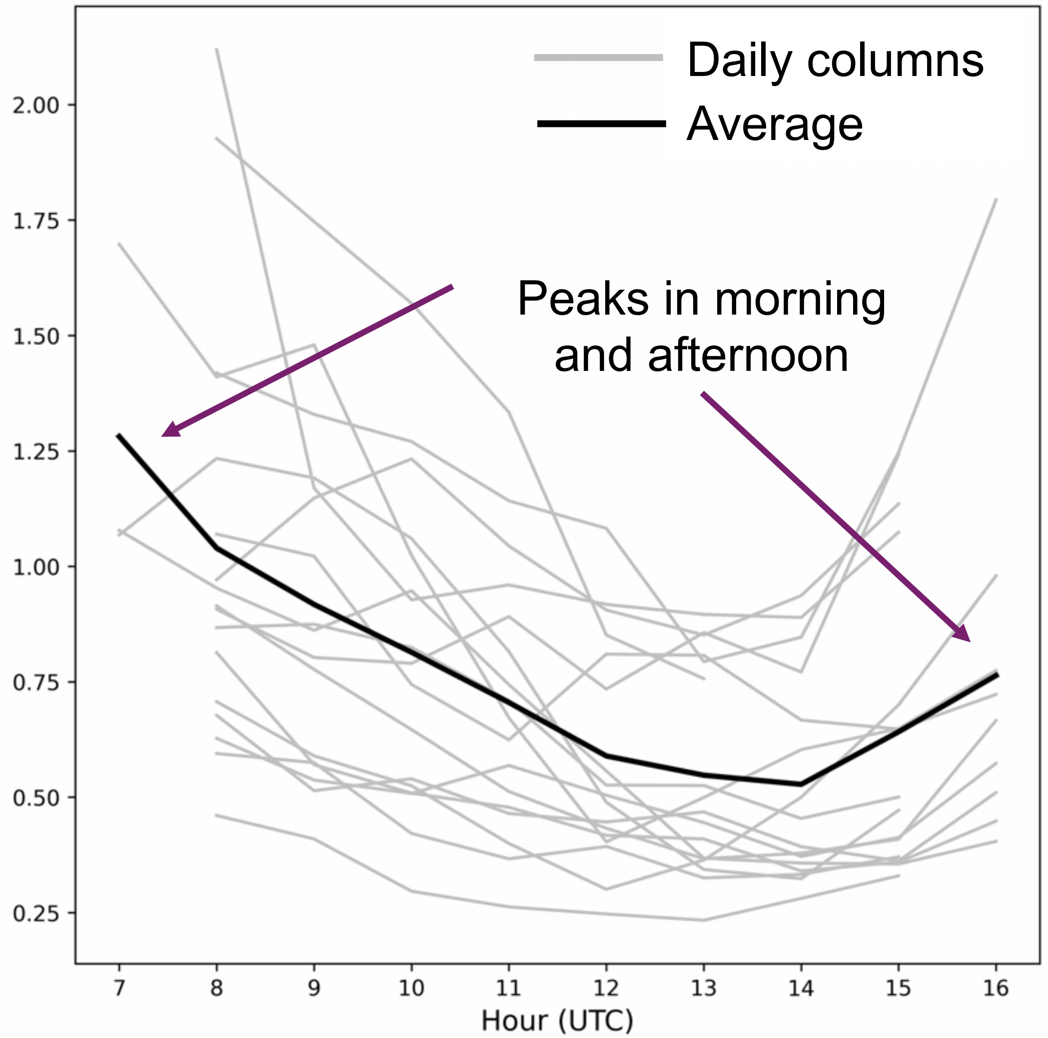
HONO along the viewing path of each elevation angle [10^{16} molecules cm^{-2}]



Retrieval

(optimal estimation)

HONO vertical column densities
[10^{15} molecules cm^{-2}]



dSCDs: differential slant column densities
DL: detection limit

Good dSCDs separation → HONO present

[Gershenson-Smith et al., *in progress*]

Summary and Outlook

- Use existing satellite observations of NH_3 to estimate UK agricultural emissions and assess the national inventory
- NH_3 publication most downloaded JGR-Atmospheres paper in 2022
- Led to investigation of contribution of NH_3 to UK urban $PM_{2.5}$ pollution and risk of NH_3 to UK habitats and public health and the efficacy of regulation and technology at mitigating this burden
- Increase the success of existing method to derive hotspot emissions by application to undersampled parts of the world
- Derive new observations of vertical profiles of NO_2 and identify and address model shortcomings in representing lighting NO_x emissions
- Investigate processes governing urban HONO with remote sensing observations in Central London
- To access publications: https://maraisresearchgroup.co.uk/publications.html
- To access data accompanying publications: https://maraisresearchgroup.co.uk/datasets.html