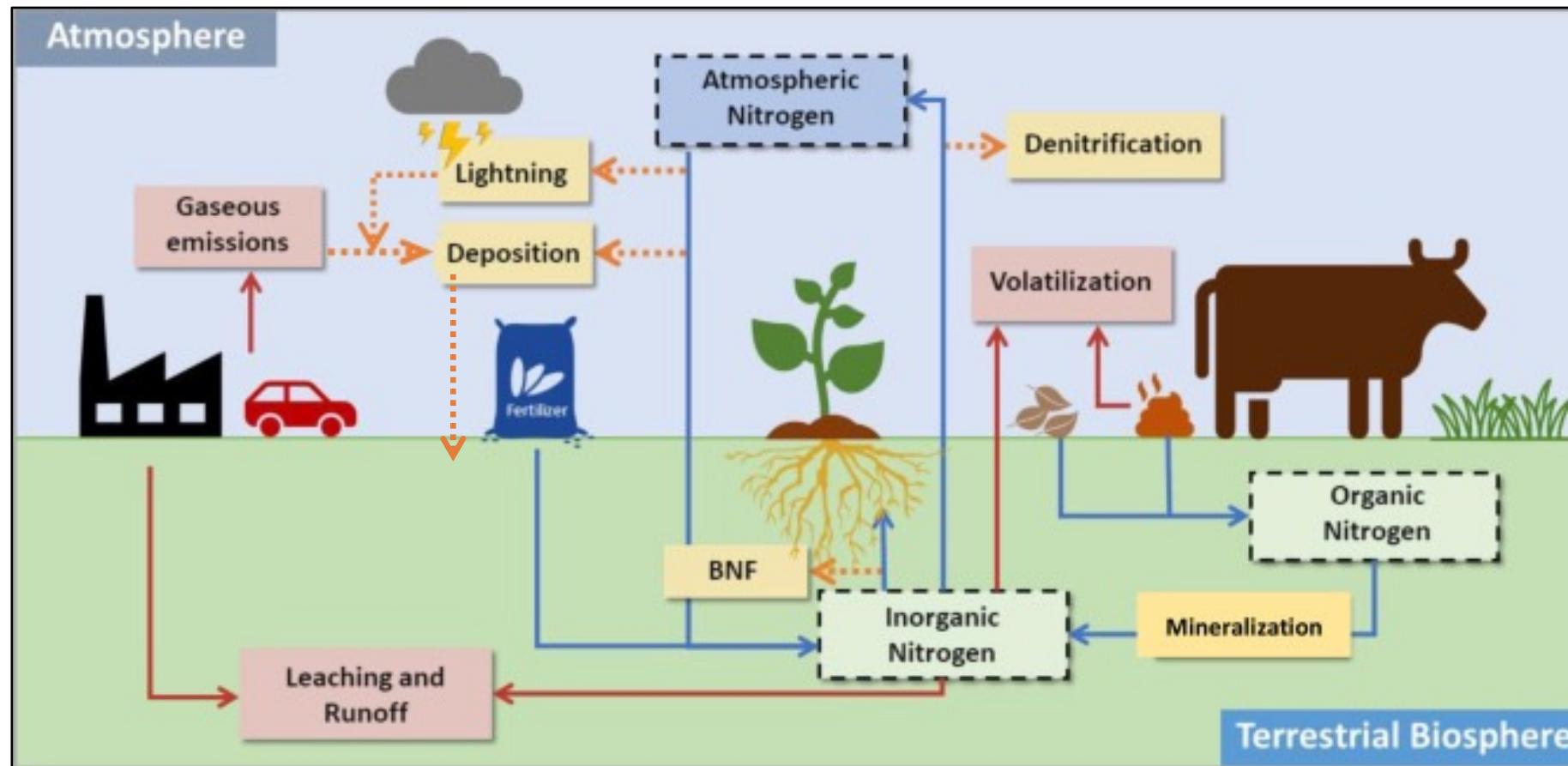


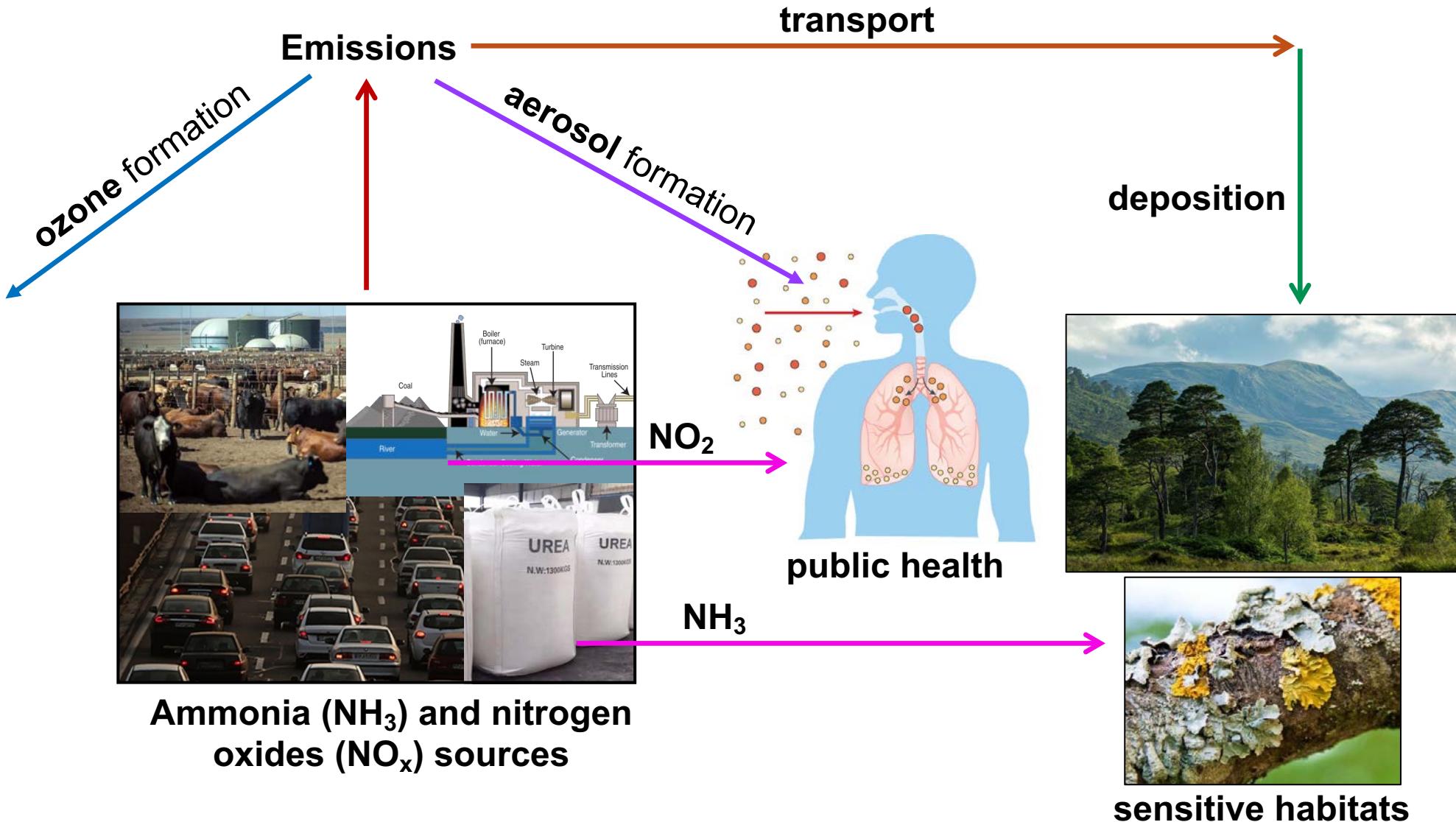
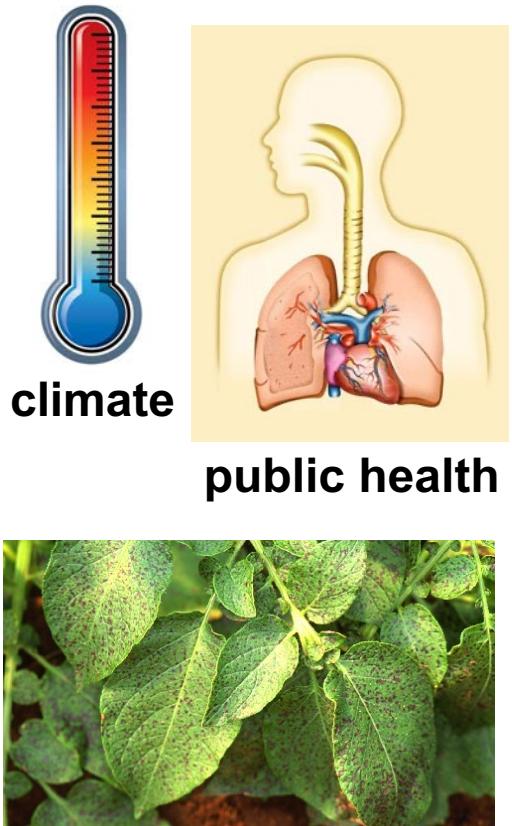
Deriving new and exploiting existing remote sensing observations to better understand sources and abundances of natural and anthropogenic reactive nitrogen

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



[Image source: Khattar et al., 2023]

Environmental Impacts of Atmospheric Reactive Nitrogen



Remote Sensing Observations Used by the Group

IASI: NH₃



LIS: lightning properties



TROPOMI: NO₂



MAX-DOAS: HONO



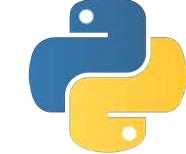
CrIS: NH₃

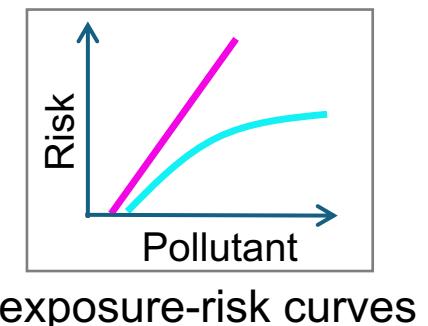
We also use:



in-situ measurements

GEOS
Chem
models


python

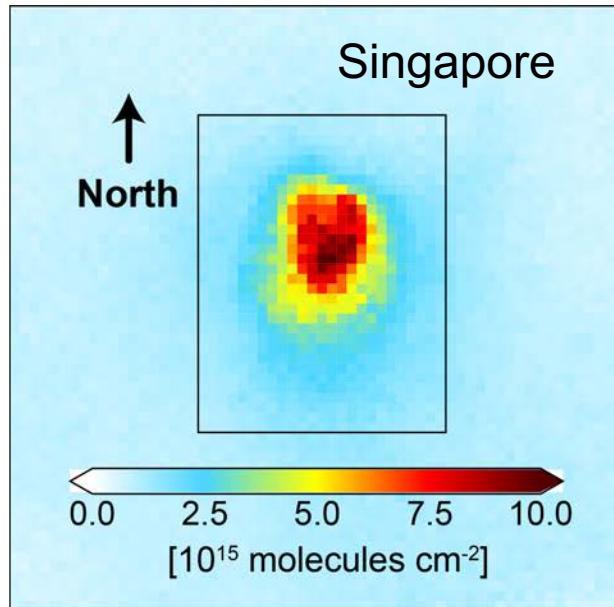


exposure-risk curves

Top-down Estimate of NO_x Emissions

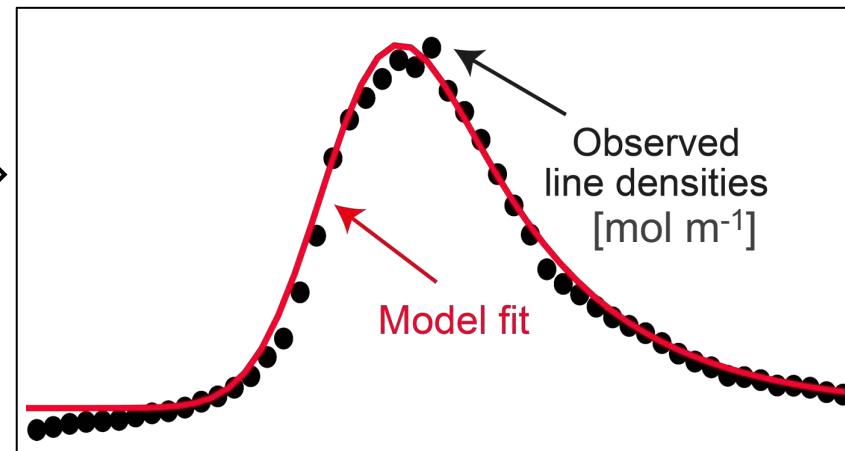
Derive NO_x emissions of isolated hotspots viewed by UV-visible space-based sensors

Wind rotated TROPOMI NO₂



Model fit to yield best-fit parameters

Across-wind sum of vertical columns



City NO_x Plume Emissions

112 mol NO_x s⁻¹

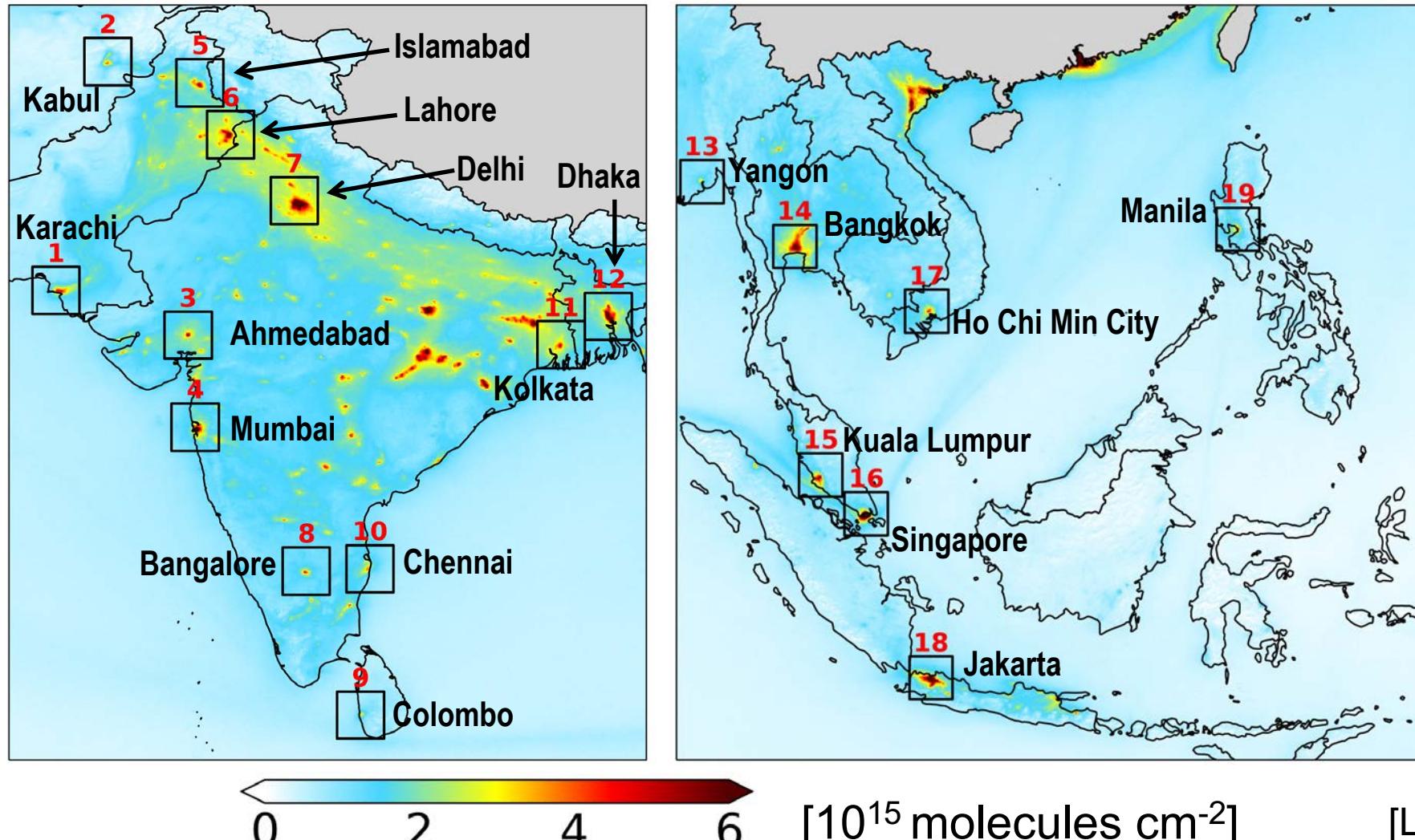


Enhance success of well-established method by defining many (54) rather than one sampling area

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 NO₂ at ~5 km resolution



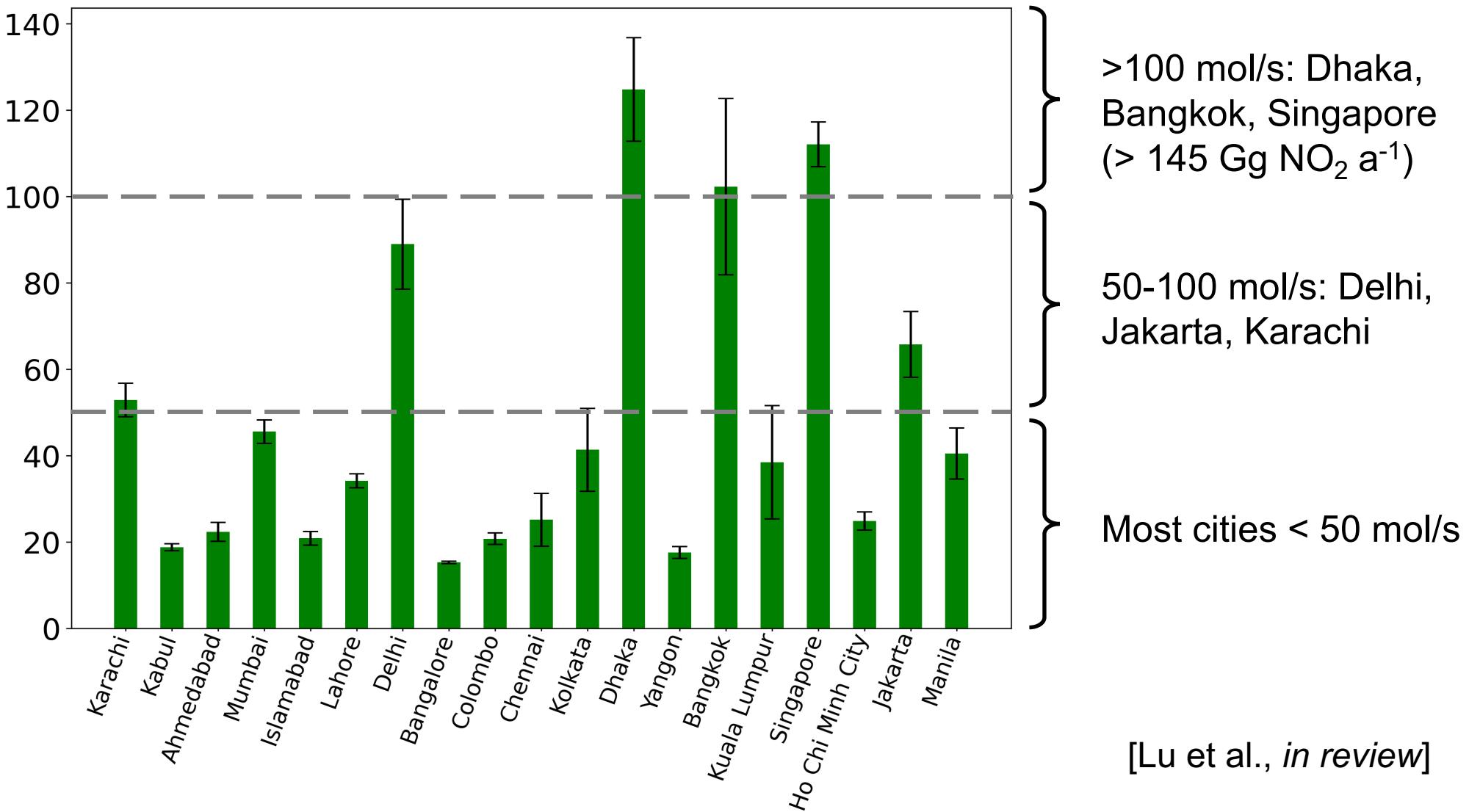
19 isolated city hotspots selected (other hotspots: industries, power plants or not isolated)

Derive City-Specific NO_x Emissions and Fit Uncertainties

City NO_x emissions for 2019 [mol/s]

Relative error range:
4-34%

Range of past studies:
10-40%

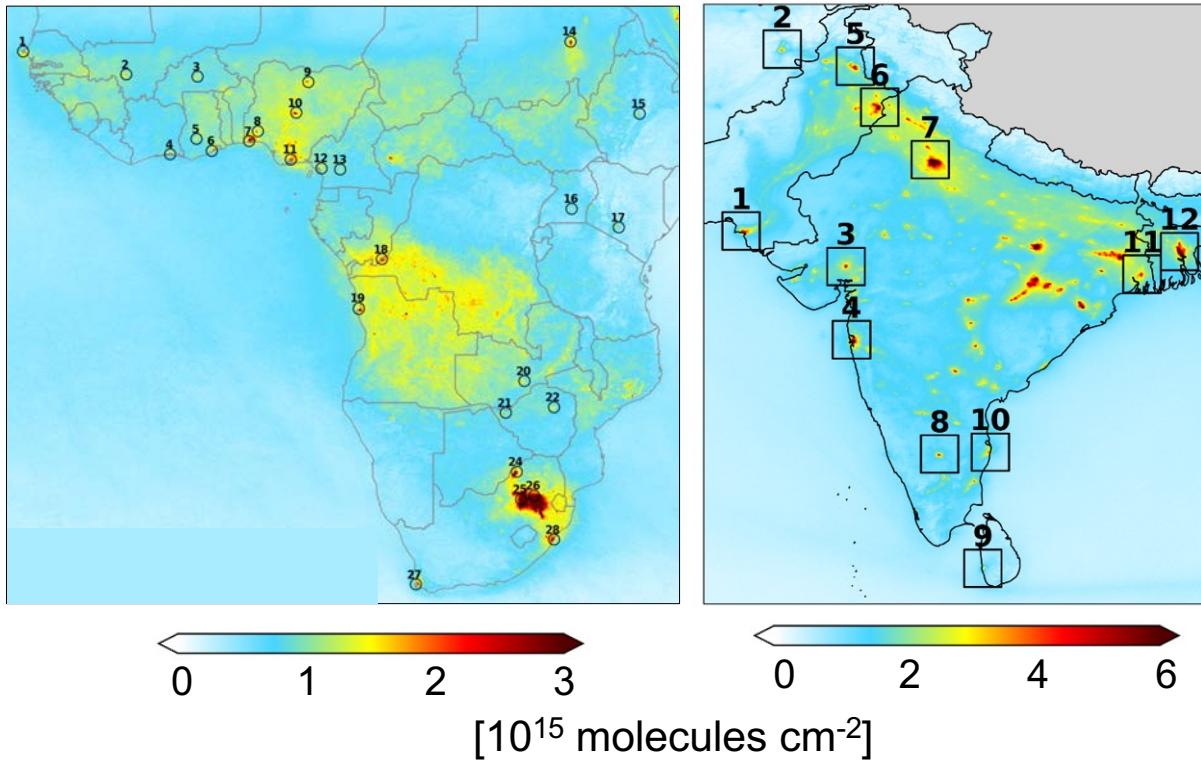


[Lu et al., *in review*]

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

NO_x Emissions of Hotspots in Sub-Saharan Africa

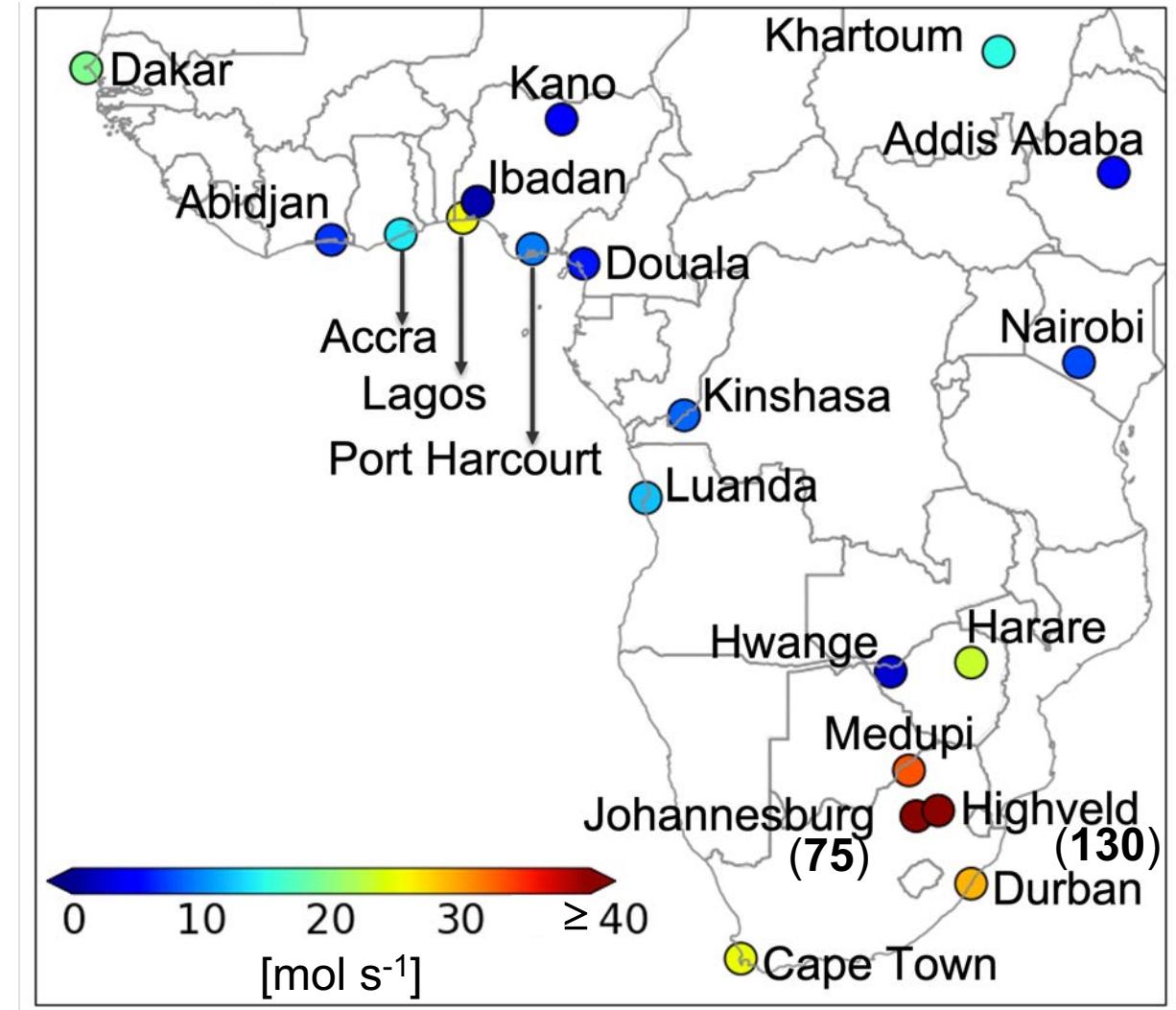
Annual mean TROPOMI NO_2 for 2019



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)



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

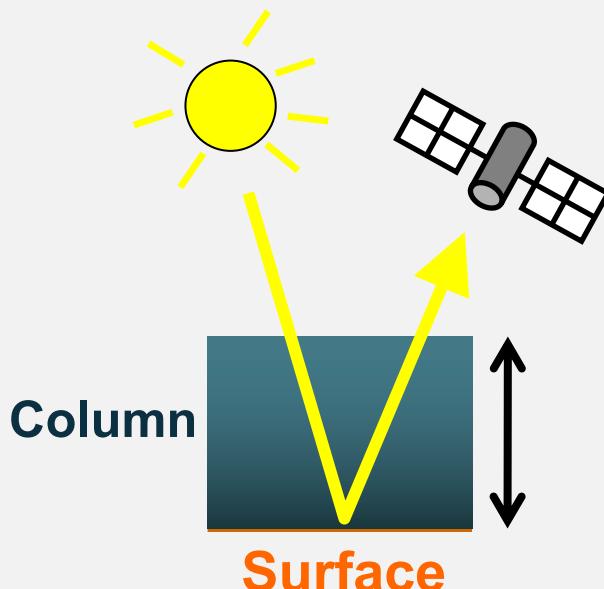


Conversion Factor

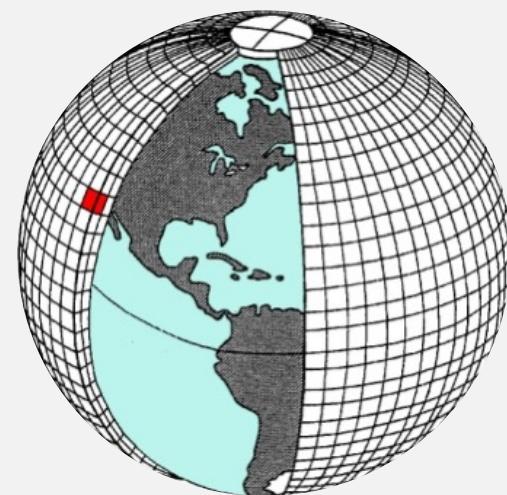


EMISSIONS

Satellite column densities



Model Concentration-to-Emission Ratio



Satellite-derived Surface Emissions

Emission

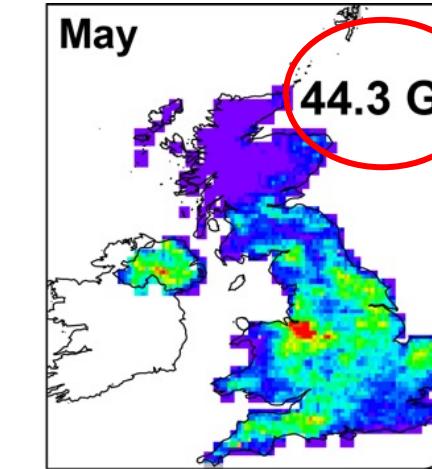
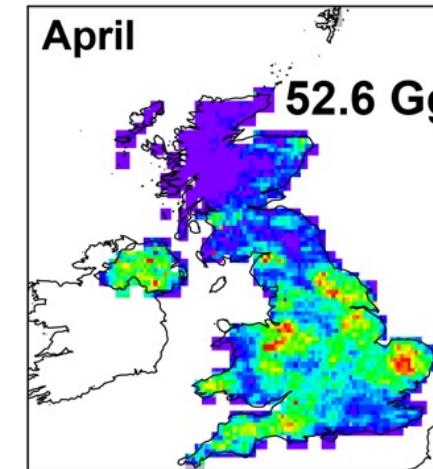
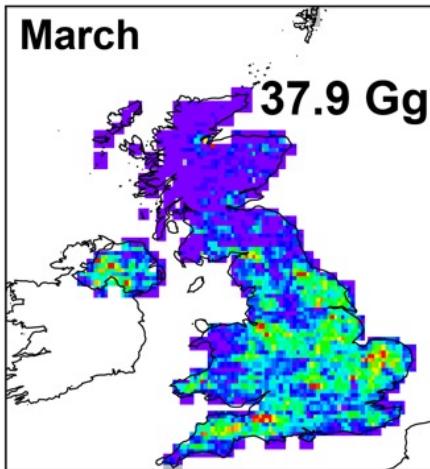


This approach possible as NH_3 has a relatively short lifetime (2-15 hours) at or near sources

High-Resolution Agricultural NH₃ Emissions for the UK

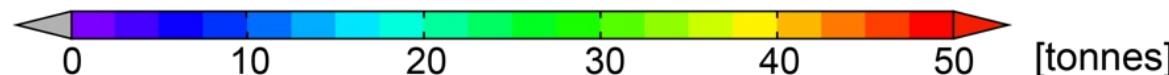
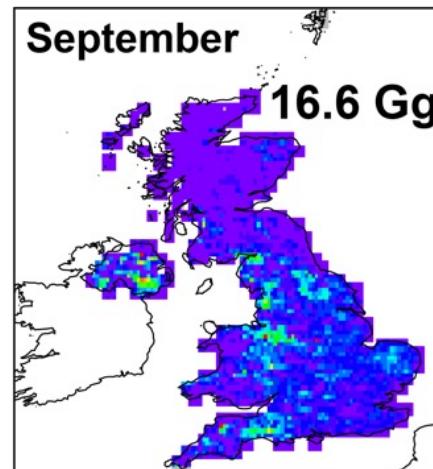
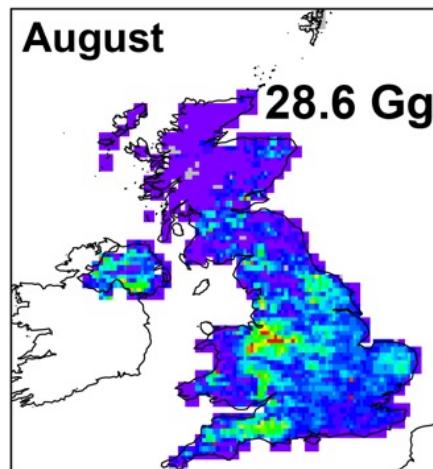
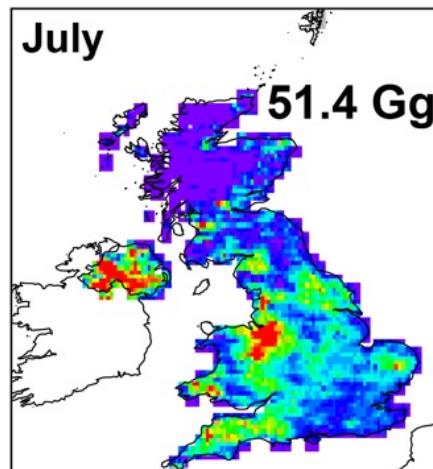
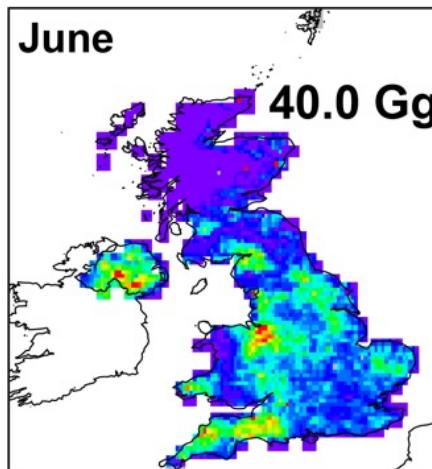
Focus on Mar-Sep when warm temperatures and clearer conditions increase sensitivity to surface NH₃

IASI: morning overpass



Total monthly emissions

1 Gg = 1 kilotonne

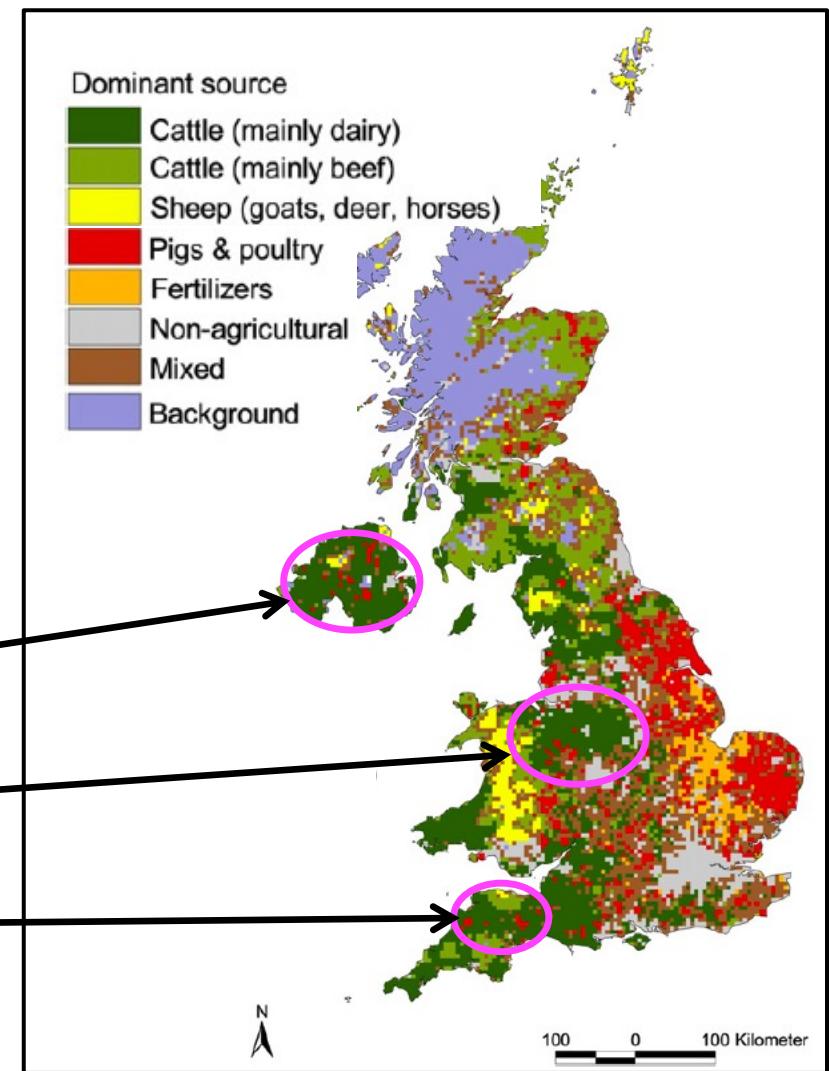
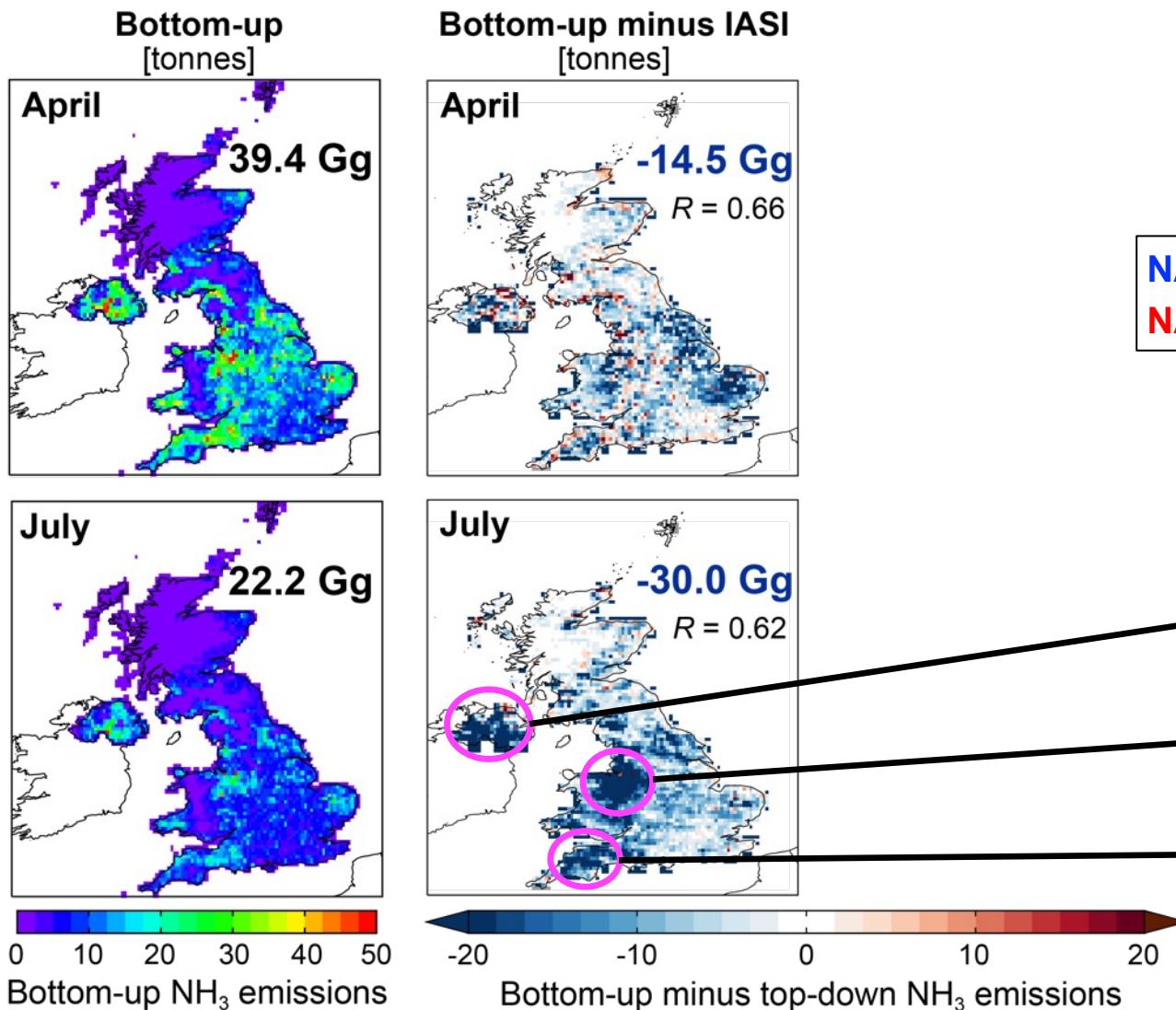


[Marais et al., JGR, 2021]

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

Satellite vs inventory NH₃ emissions: spatial distribution

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

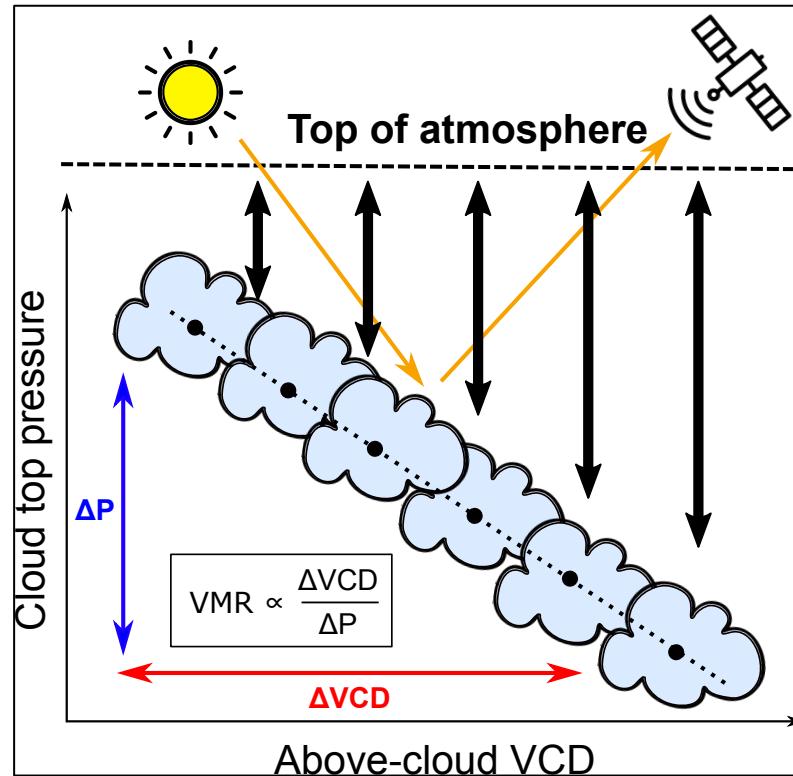


[Marais et al., JGR, 2021]

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 NO₂

Cloud-slice TROPOMI NO₂
above optically thick clouds



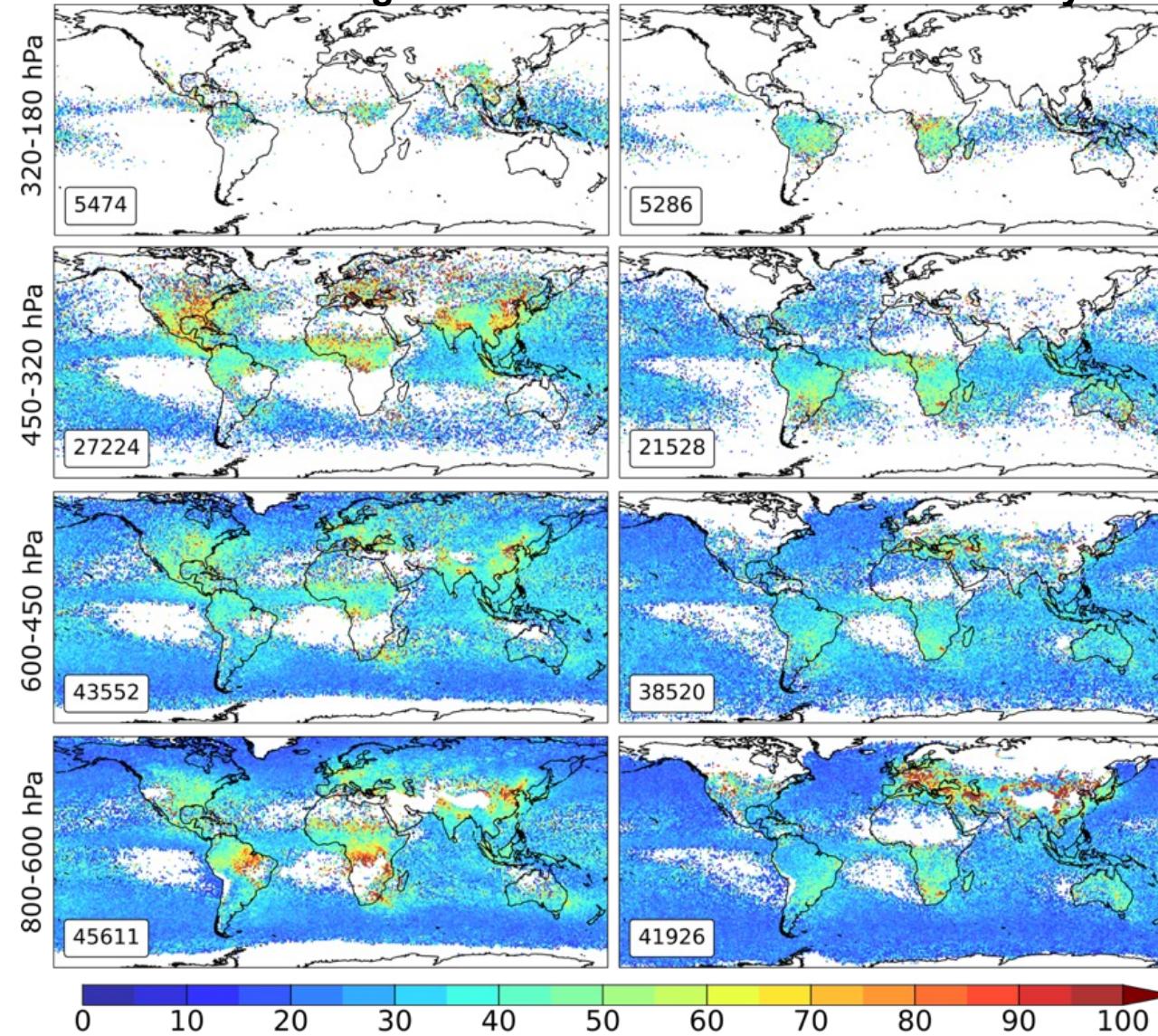
Addresses absence of routine
observations

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

Seasonal multiyear mean NO₂ mixing ratios [pptv]

June-August

December-February

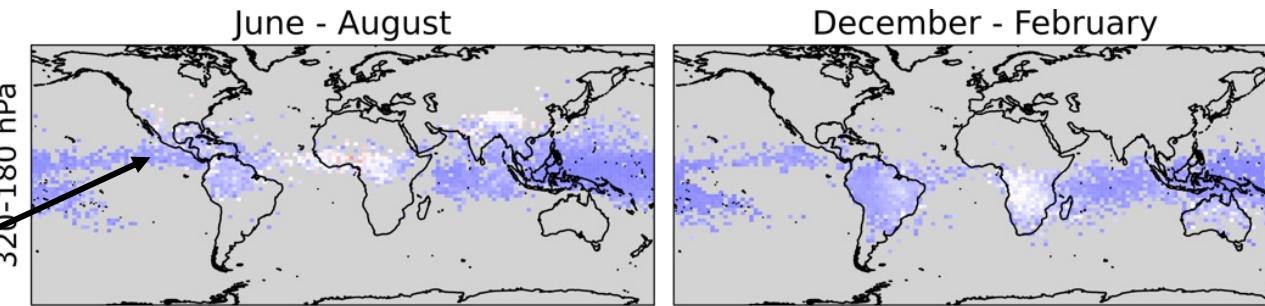


[Horner et al., submitted, ACP]

Use New Datasets to Assess State of Knowledge

Difference between GEOS-Chem and cloud-sliced NO₂ [%]

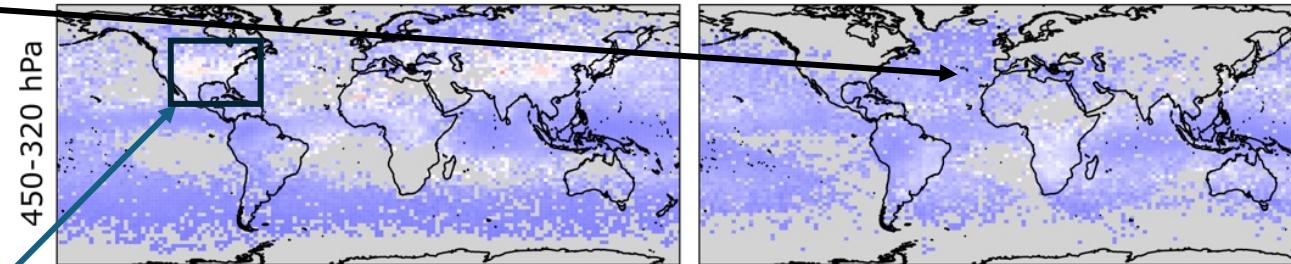
Model low bias over remote regions



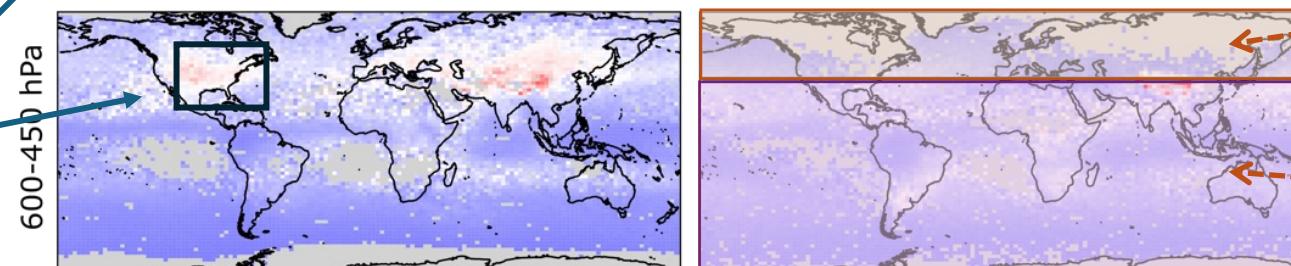
Model < Observed

Model > Observed

Model high bias
over Southeast US
(lightning NO_x)

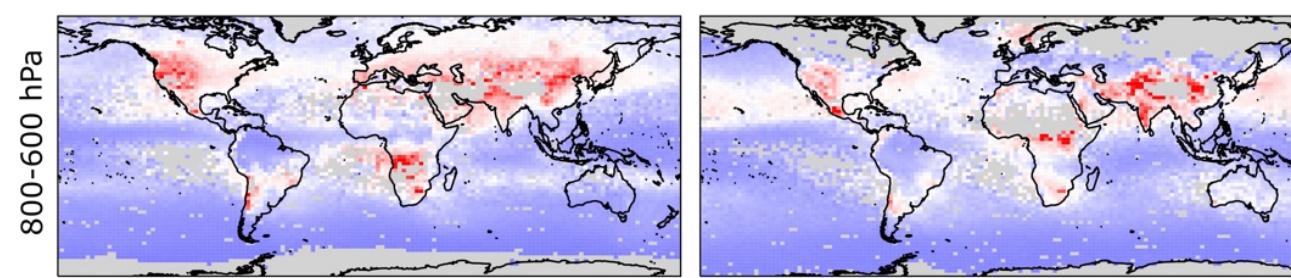


500 mol NO_x per flash



35°N

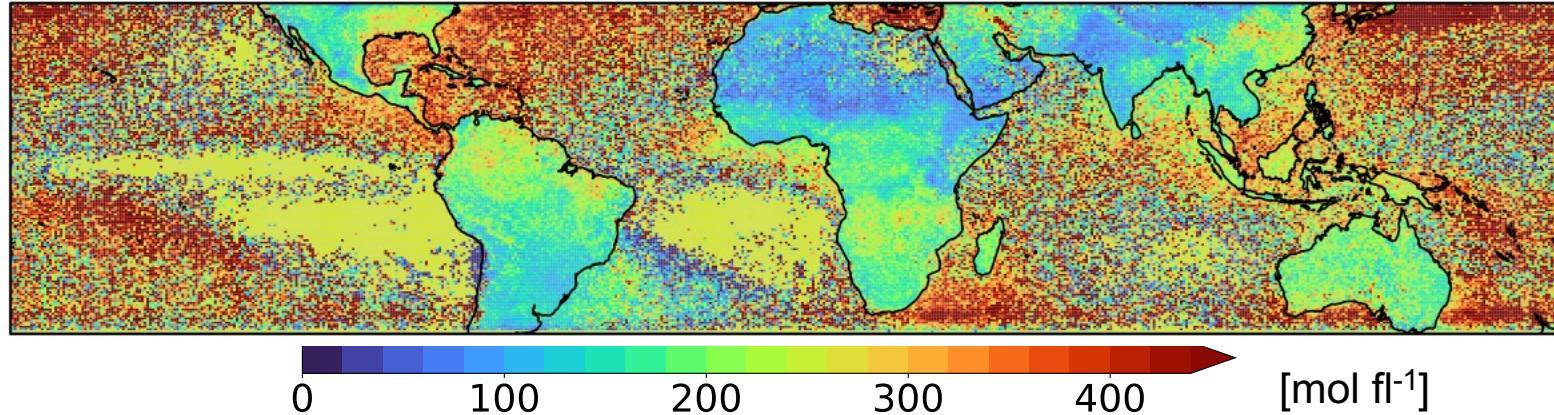
260 mol NO_x per flash



Time- and Space-Resolved Lightning NO_x Production

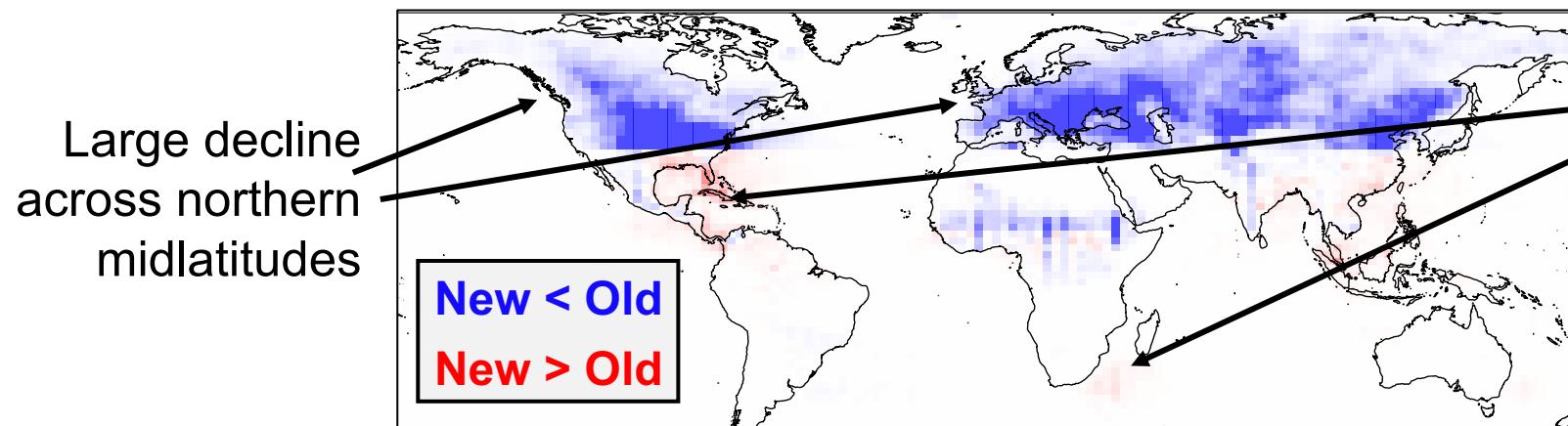
Use satellite observations of lightning flash energy to calculate hourly gridded production rates to GEOS-Chem lightning NO_x emission inventory

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)



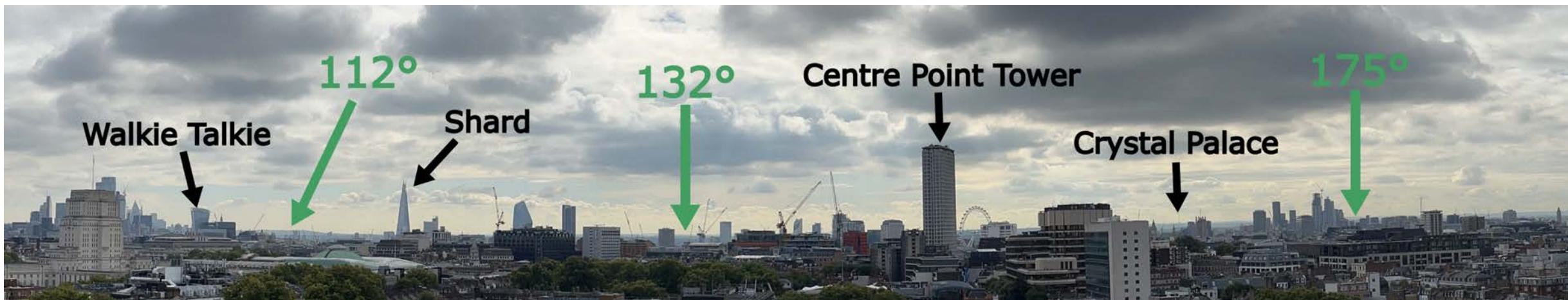
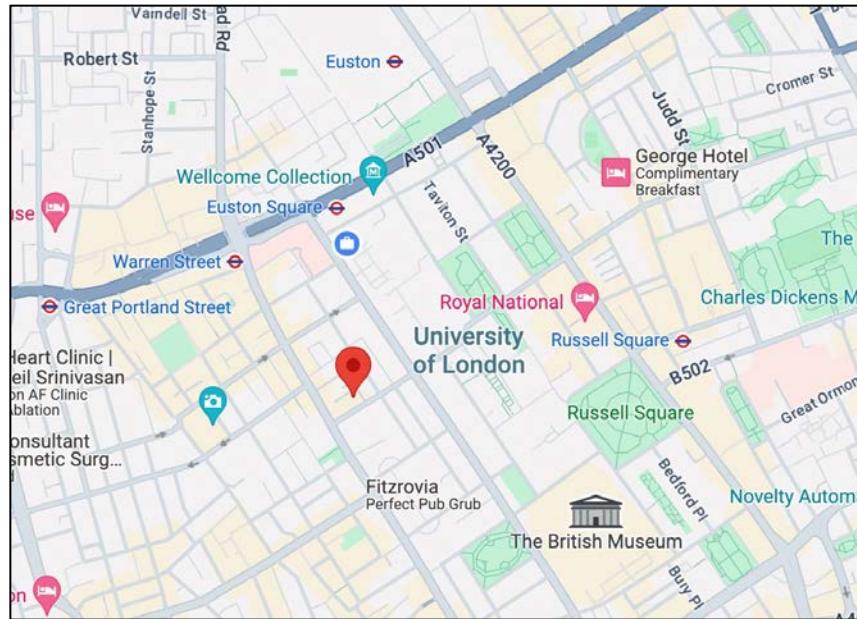
Small increases over oceans

Large decline across northern midlatitudes

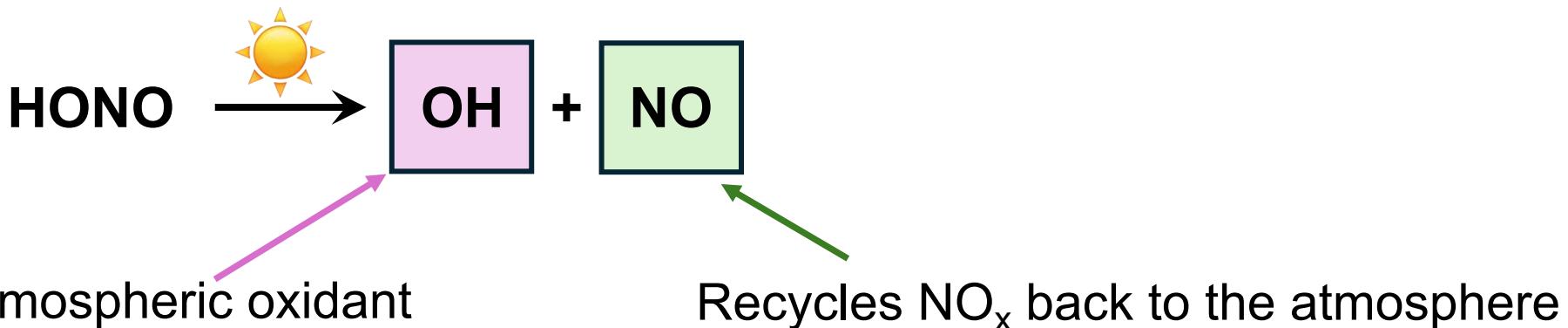
[Horner et al., *in progress*]

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

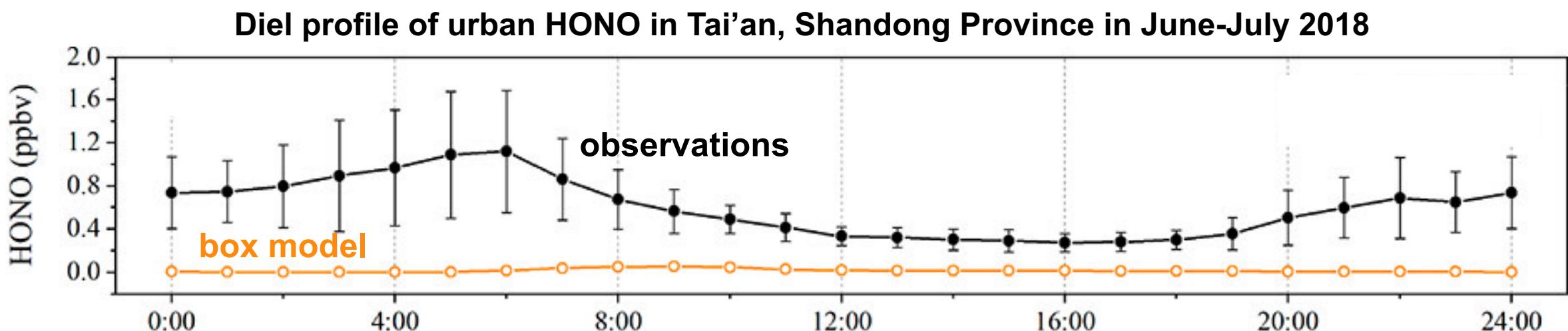
Observations of Nitrous Acid (HONO) in Central London



Observations of Nitrous Acid (HONO) in Central London

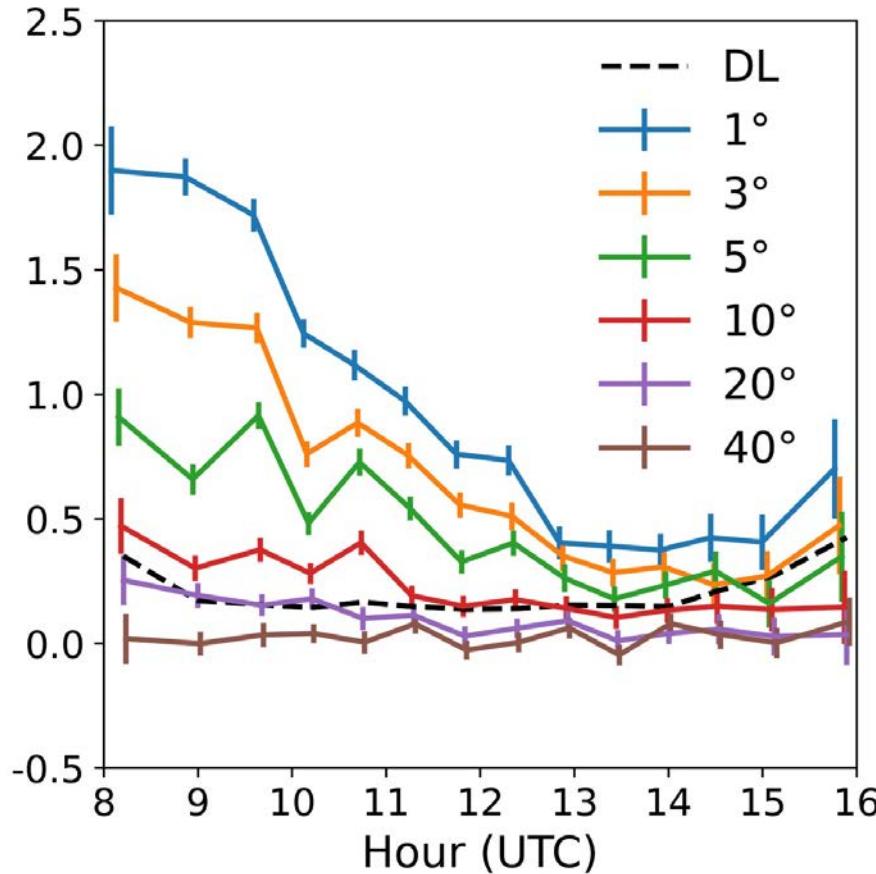


Best understanding of HONO (models) routinely fails to reproduce reality:



Observations of nitrous acid (HONO) in Central London

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

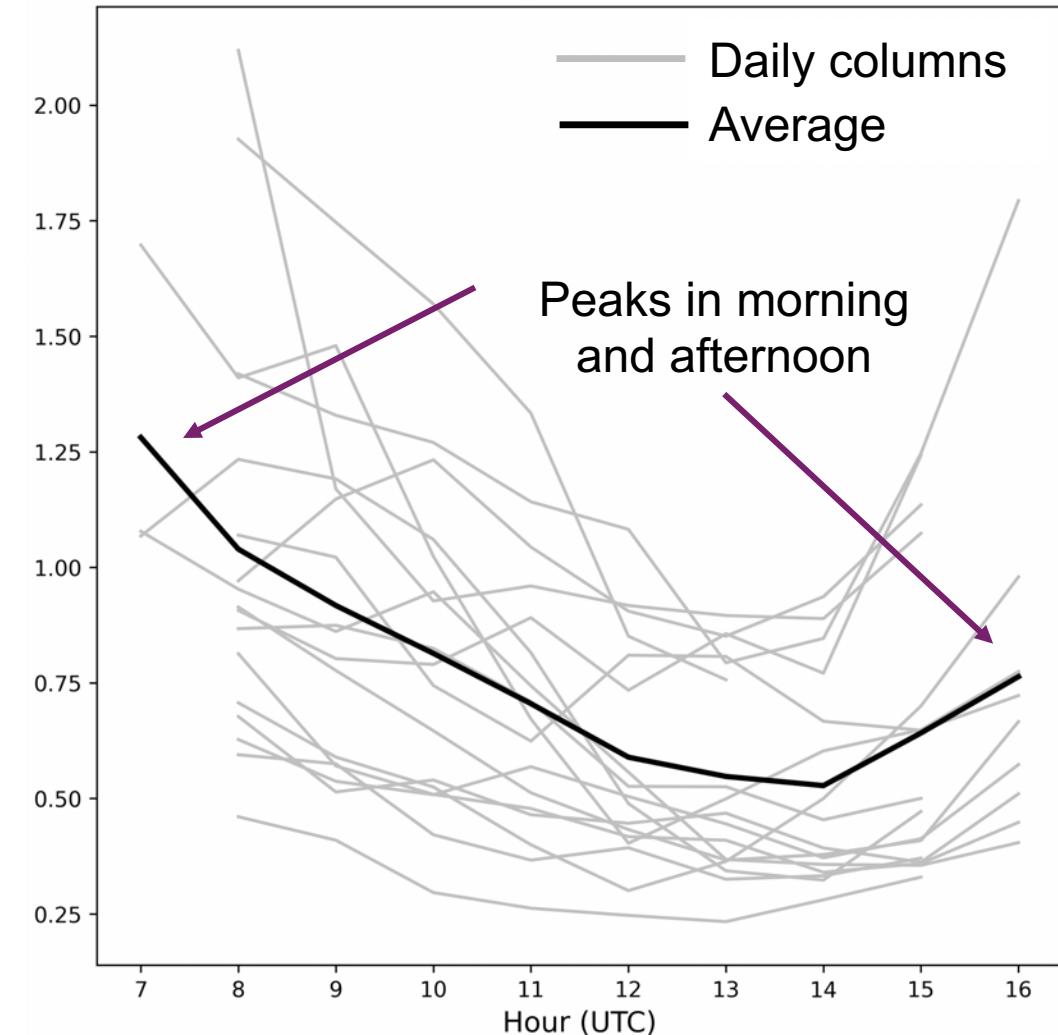


dSCDs: differential slant column densities

DL: detection limit

Good dSCDs separation → HONO present

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



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

Summary and Outlook

- Use existing satellite observations of NH₃ to estimate UK agricultural emissions and assess the national inventory
- NH₃ publication most downloaded JGR-Atmospheres paper in 2022
- Led to investigation of contribution of NH₃ to UK urban PM_{2.5} pollution and risk of NH₃ 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 NO₂ 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>