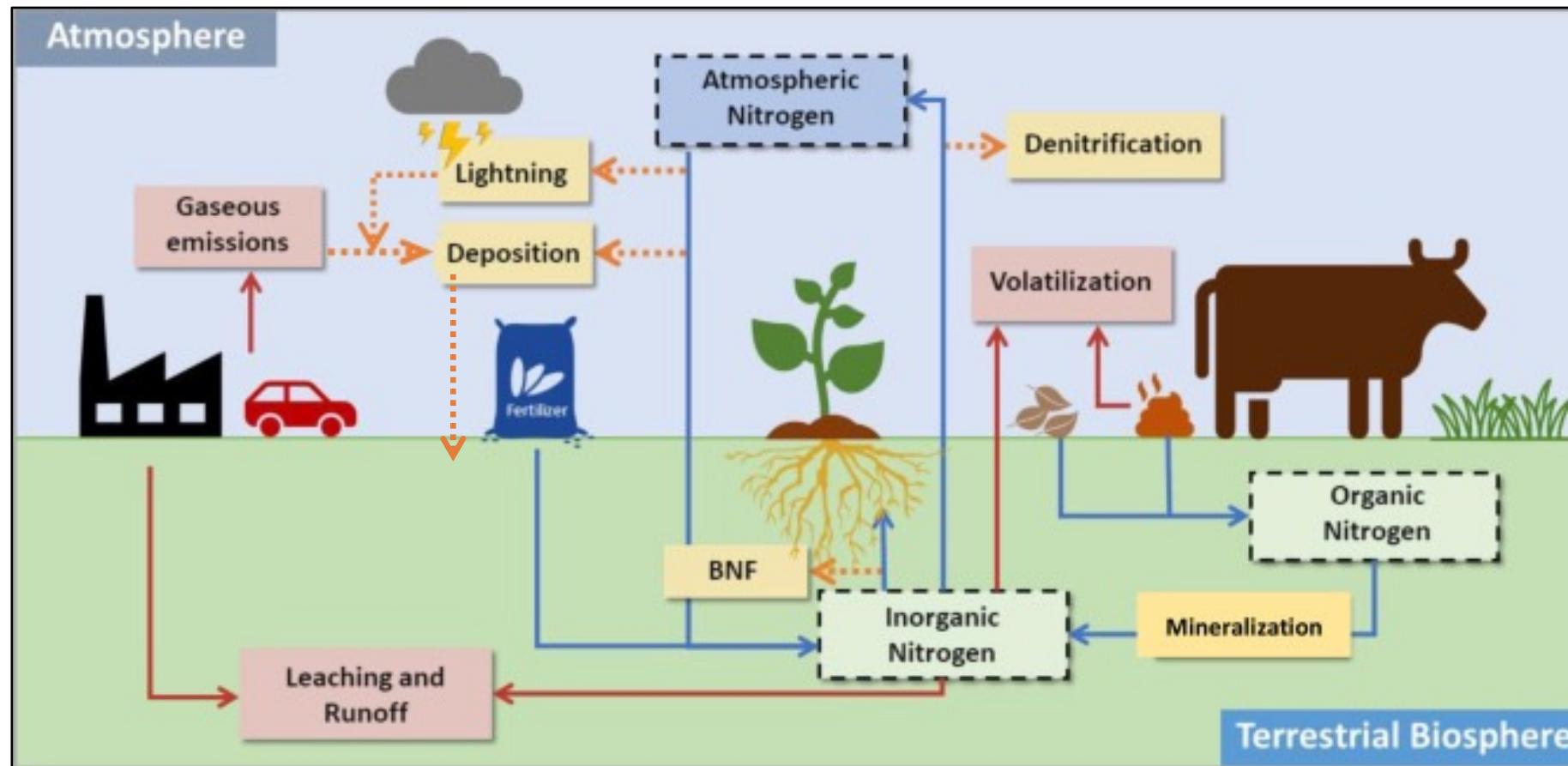


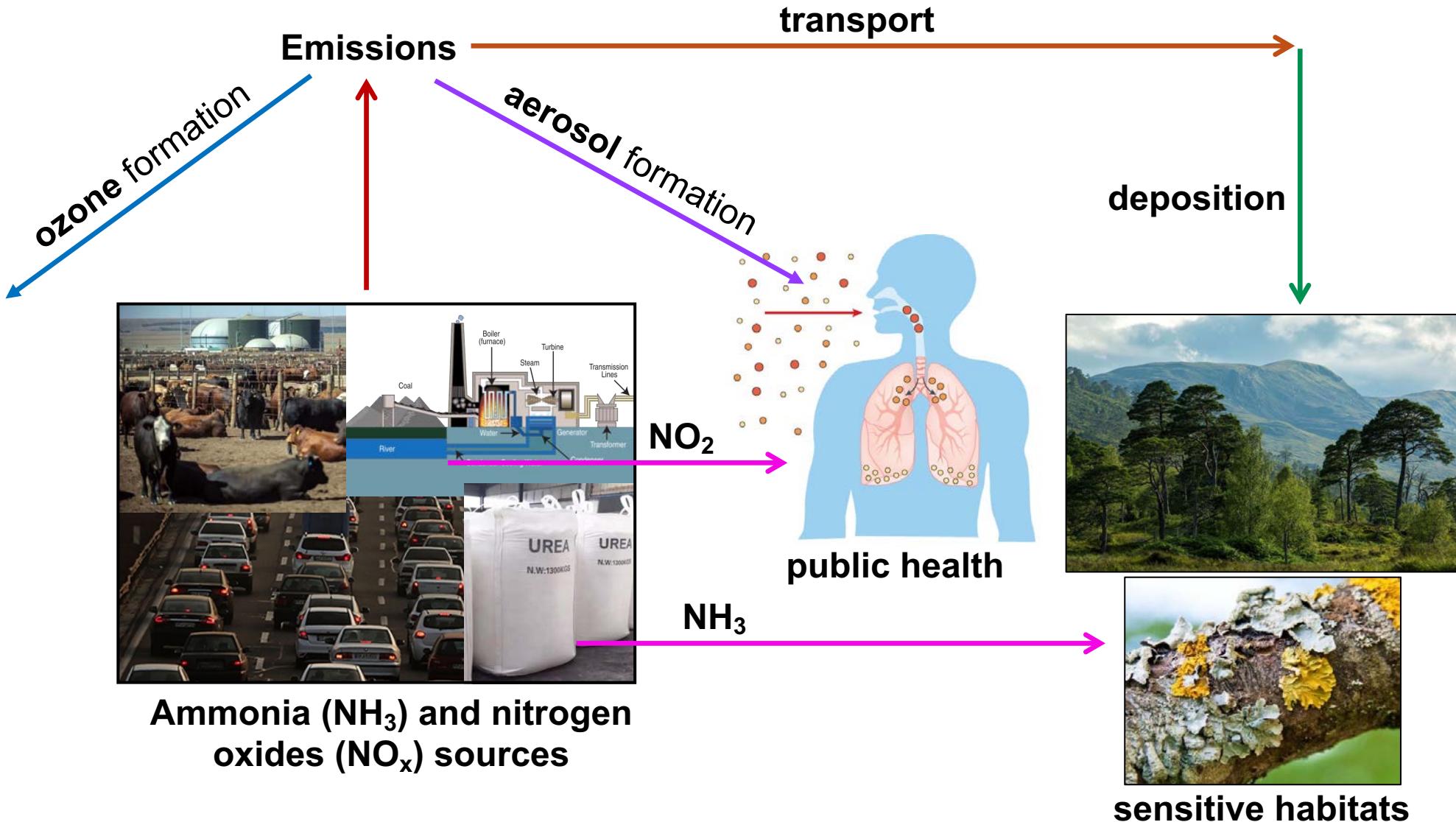
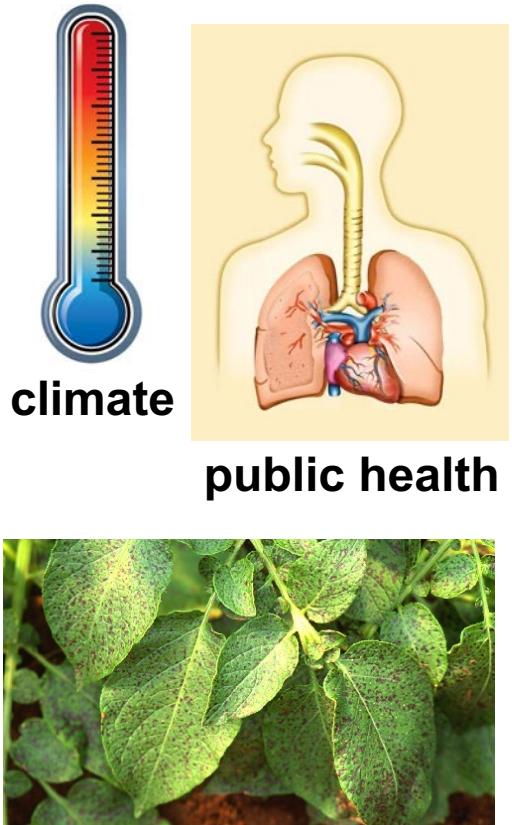
# Deriving new and exploiting existing remote sensing observations to better understand sources and abundances of natural and anthropogenic reactive nitrogen

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: NH<sub>3</sub>



LIS: lightning properties



TROPOMI: NO<sub>2</sub>



MAX-DOAS: HONO



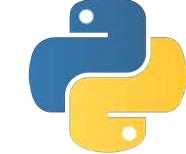
CrIS: NH<sub>3</sub>

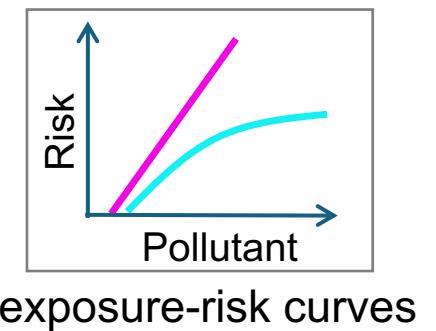
We also use:



in-situ measurements

**GEOS**  
**Chem**  
models

  
python

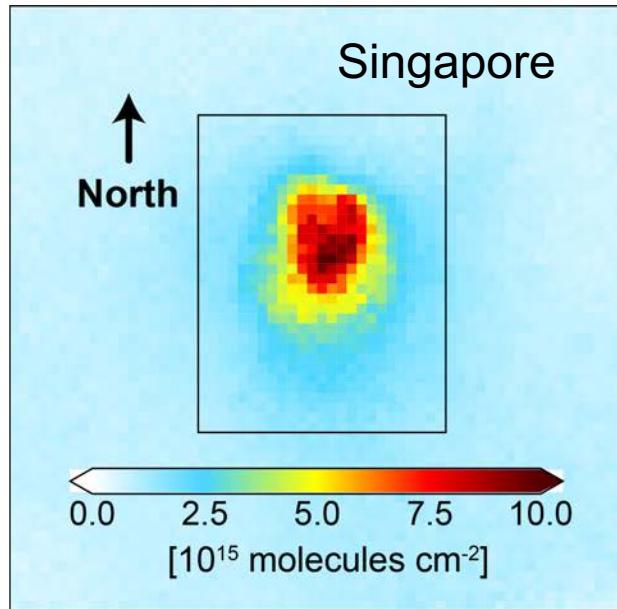


exposure-risk curves

# Top-down Estimate of NO<sub>x</sub> Emissions

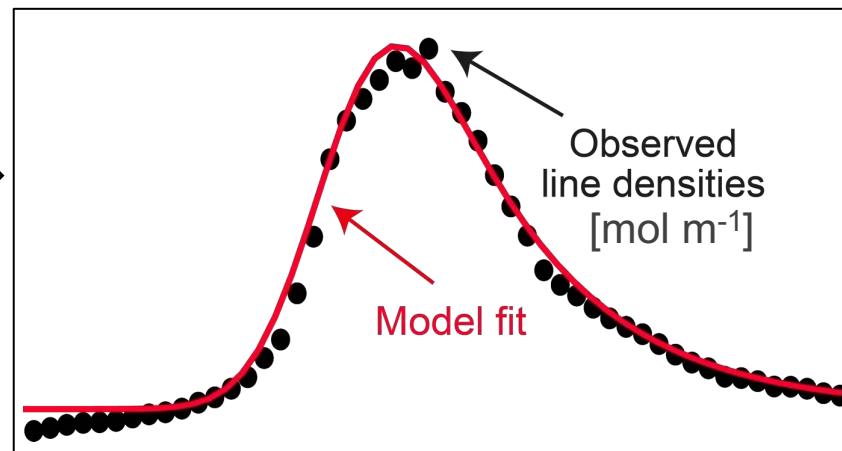
Derive NO<sub>x</sub> emissions of isolated hotspots viewed by UV-visible space-based sensors

## Wind rotated TROPOMI NO<sub>2</sub>



## Model fit to yield best-fit parameters

Across-wind sum of vertical columns



## City NO<sub>x</sub> Plume Emissions

**112 mol NO<sub>x</sub> s<sup>-1</sup>**

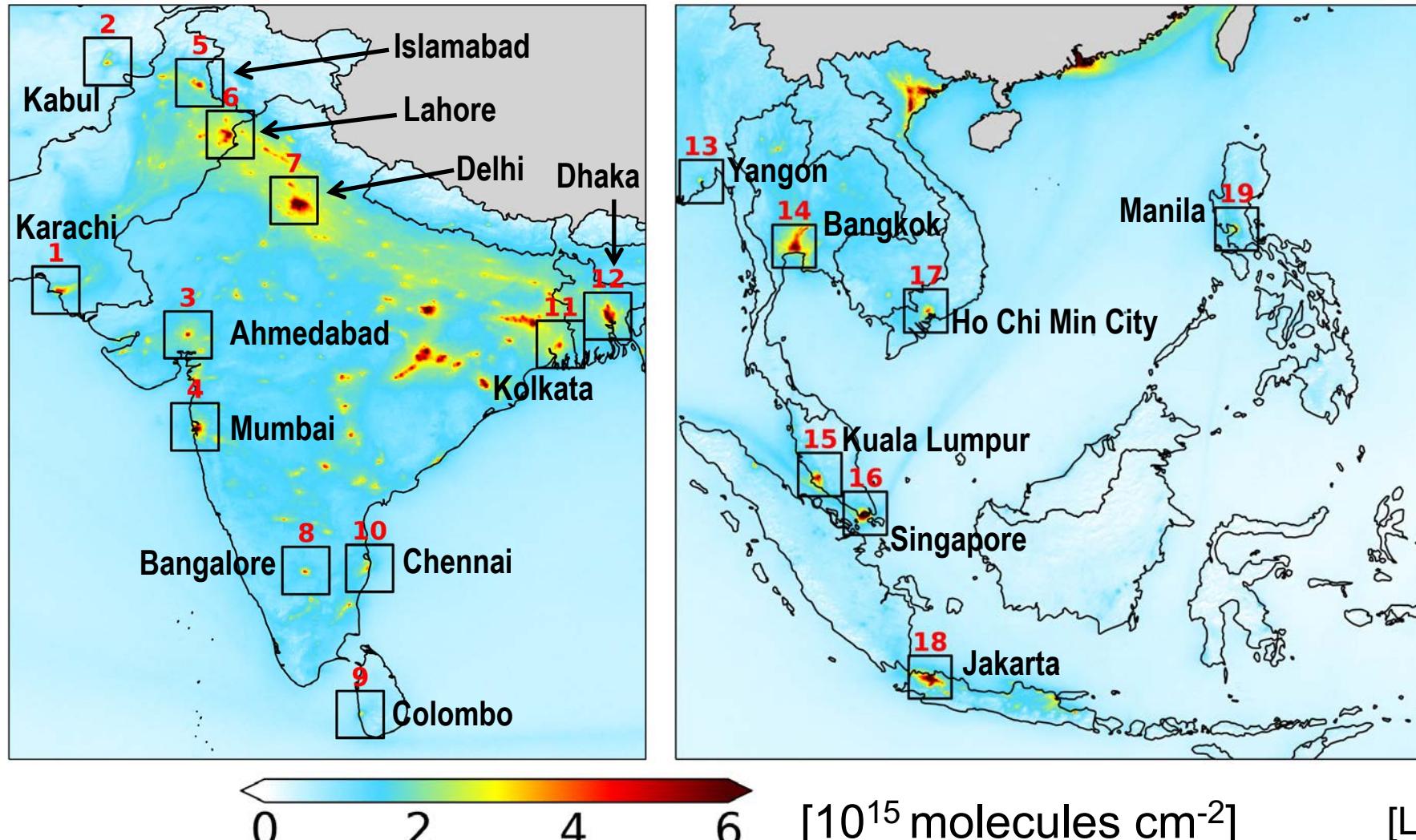


Enhance success of well-established method by defining many (54) rather than one sampling area

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<sub>2</sub> at ~5 km resolution



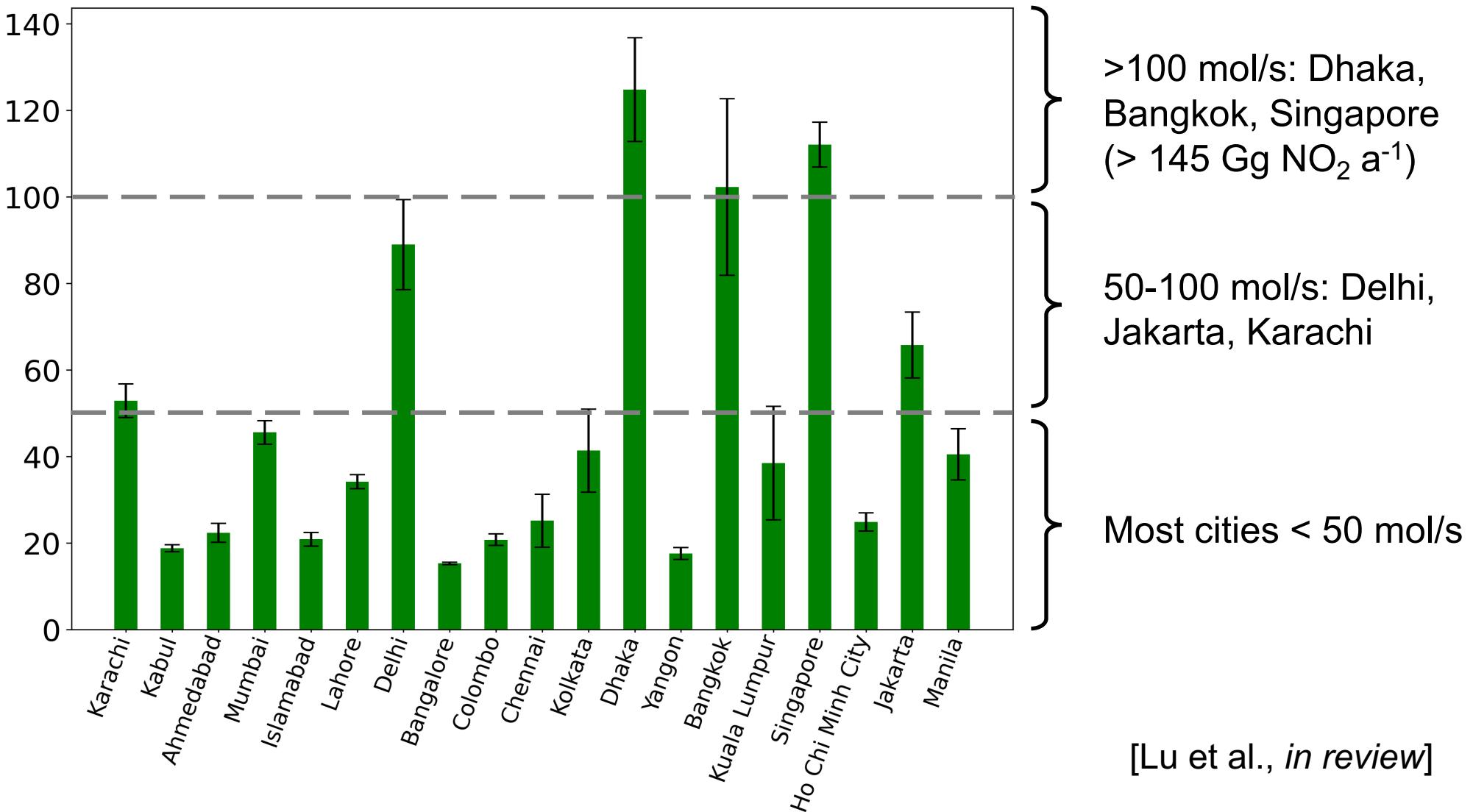
19 isolated city hotspots selected (other hotspots: industries, power plants or not isolated)

# Derive City-Specific NO<sub>x</sub> Emissions and Fit Uncertainties

City NO<sub>x</sub> emissions for 2019 [mol/s]

Relative error range:  
**4-34%**

Range of past studies:  
**10-40%**

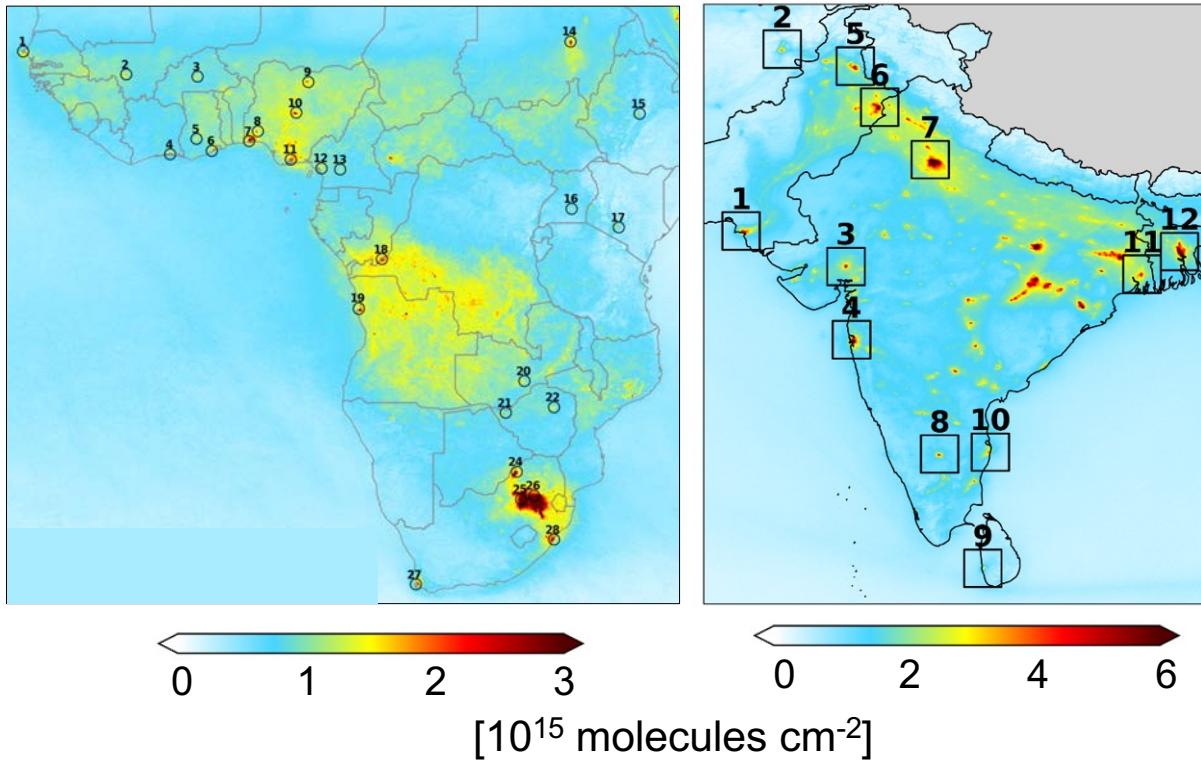


[Lu et al., *in review*]

NO<sub>x</sub> emissions from mean of individual successful fits. Standard deviation provides fit error.

# $\text{NO}_x$ Emissions of Hotspots in Sub-Saharan Africa

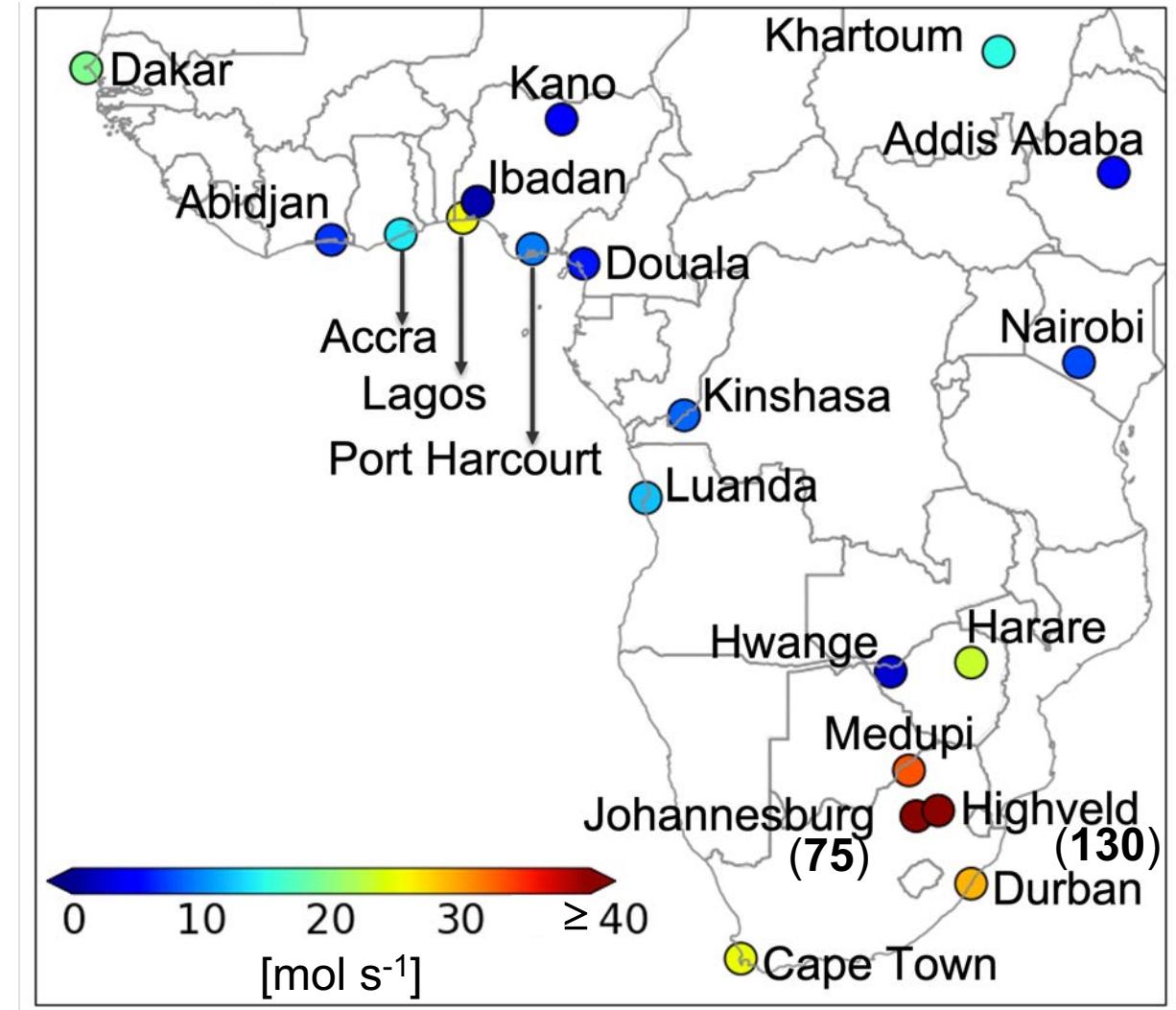
Annual mean TROPOMI  $\text{NO}_2$  for 2019



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 ( $\text{NH}_3$ ) Emissions

Simple mass balance approach:

Convert atmospheric **column concentrations** to surface **emissions** by relating the two with a **model**

**ABUNDANCES**

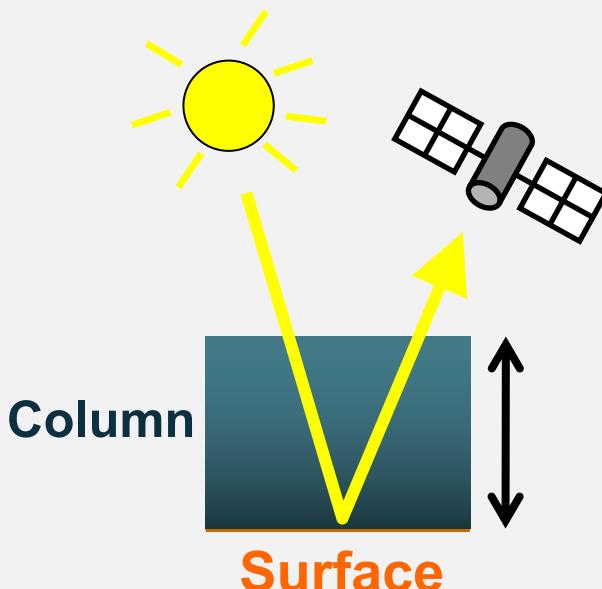


**Conversion Factor**

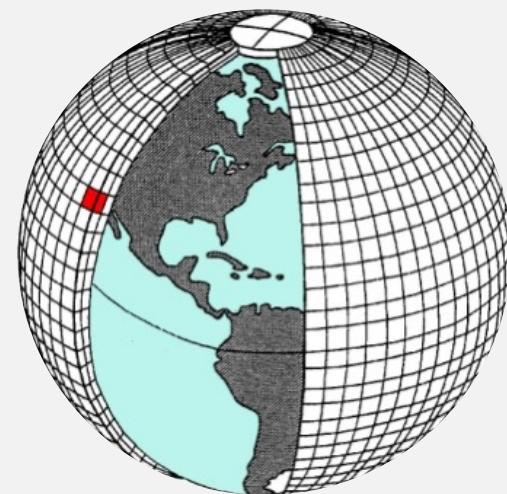


**EMISSIONS**

**Satellite column densities**



**Model Concentration-to-Emission Ratio**



**Satellite-derived Surface Emissions**

**Emission**

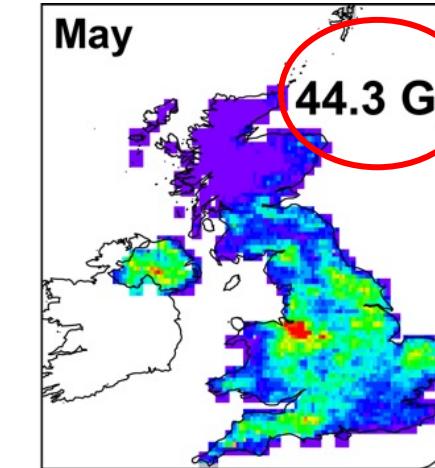
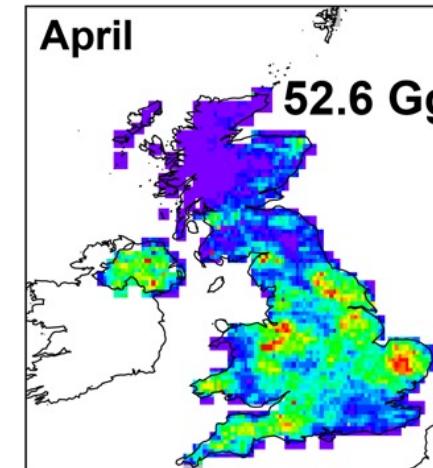
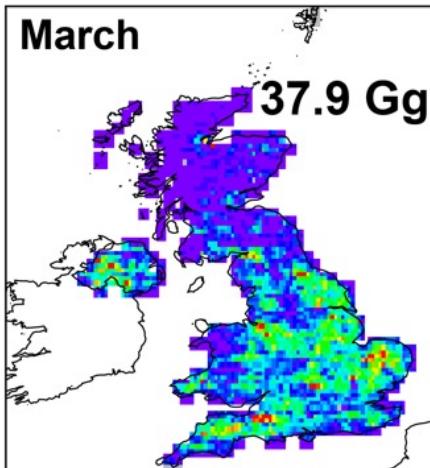


This approach possible as  $\text{NH}_3$  has a relatively short lifetime (2-15 hours) at or near sources

# High-Resolution Agricultural NH<sub>3</sub> Emissions for the UK

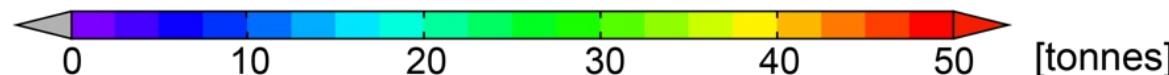
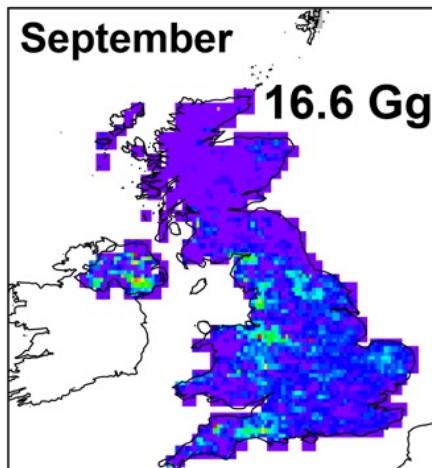
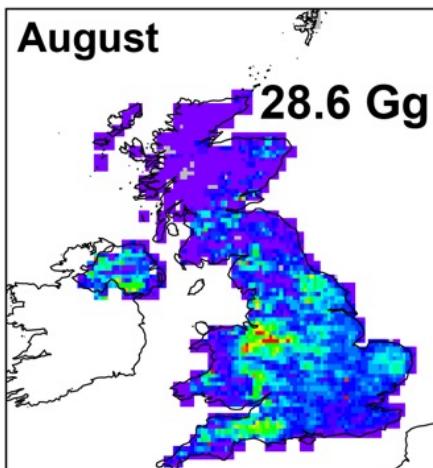
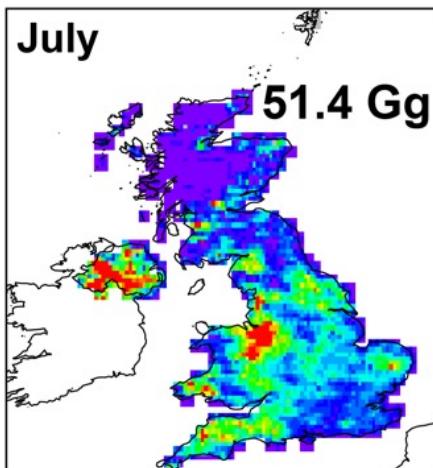
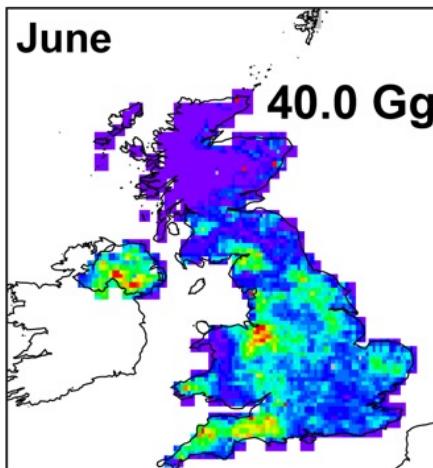
Focus on Mar-Sep when warm temperatures and clearer conditions increase sensitivity to surface NH<sub>3</sub>

IASI: morning overpass



Total monthly emissions

1 Gg = 1 kilotonne

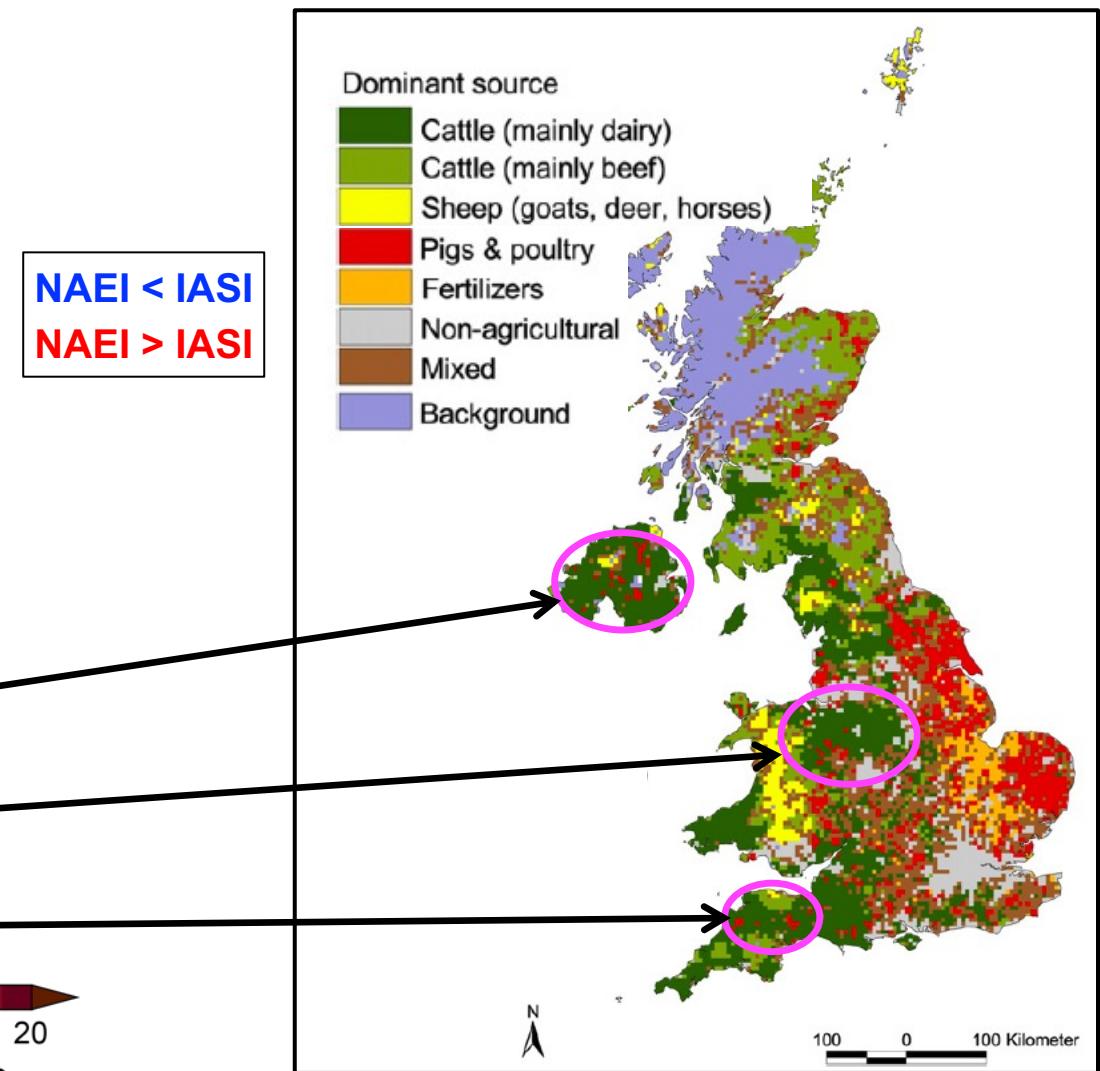
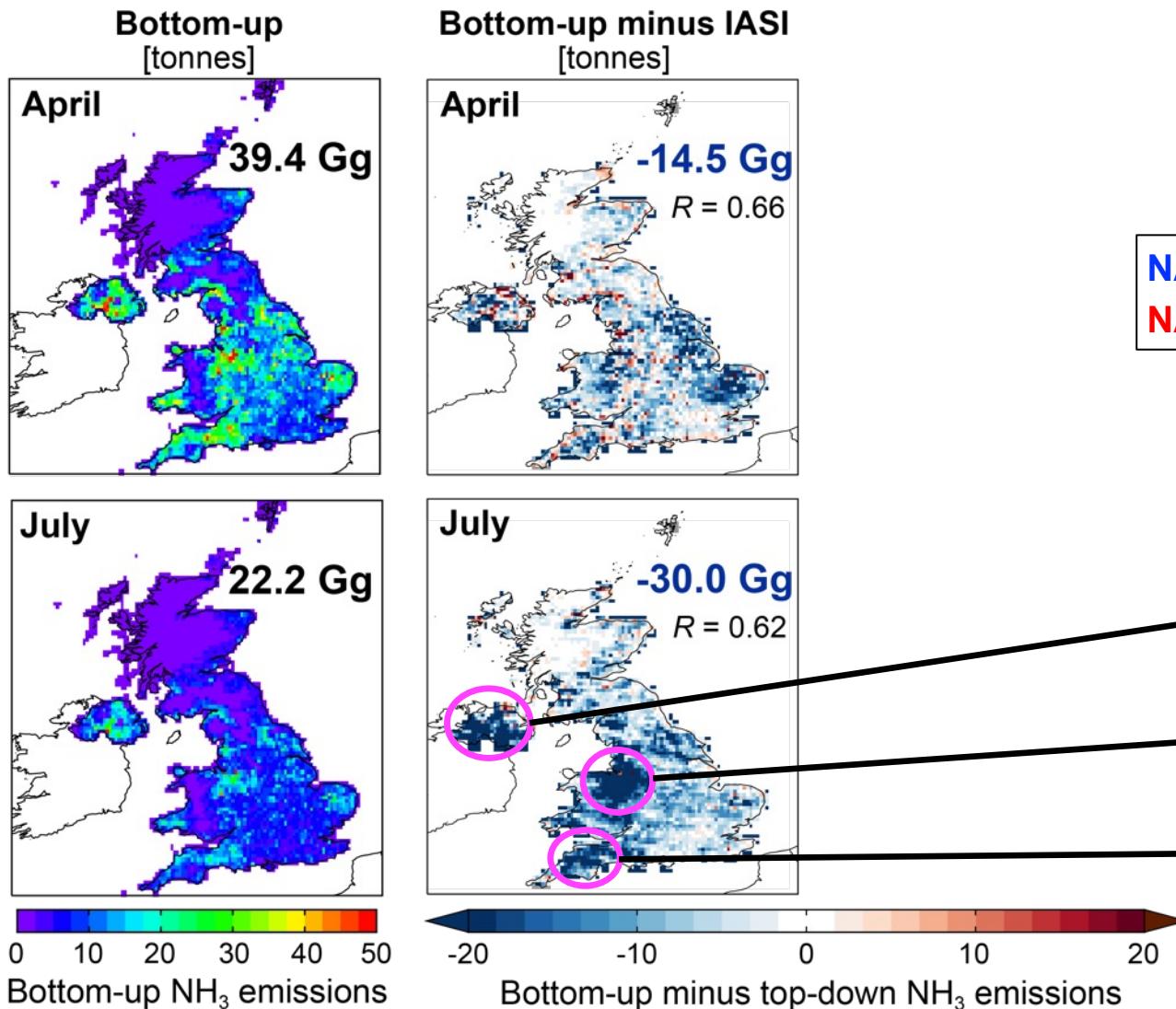


[Marais et al., JGR, 2021]

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

# Satellite vs inventory NH<sub>3</sub> emissions: spatial distribution

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

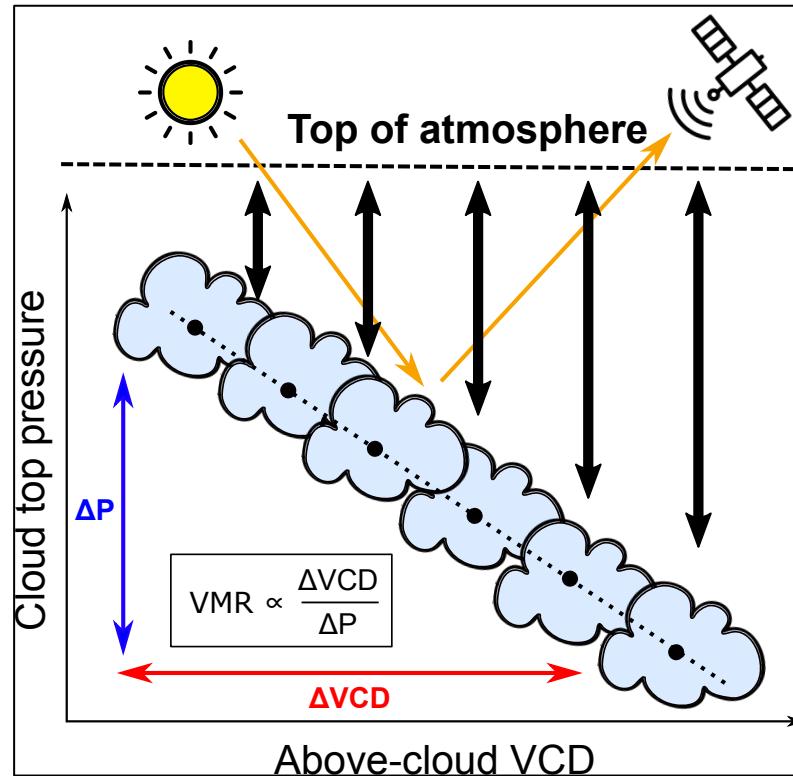


[Marais et al., JGR, 2021]

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<sub>2</sub>

Cloud-slice TROPOMI NO<sub>2</sub>  
above optically thick clouds



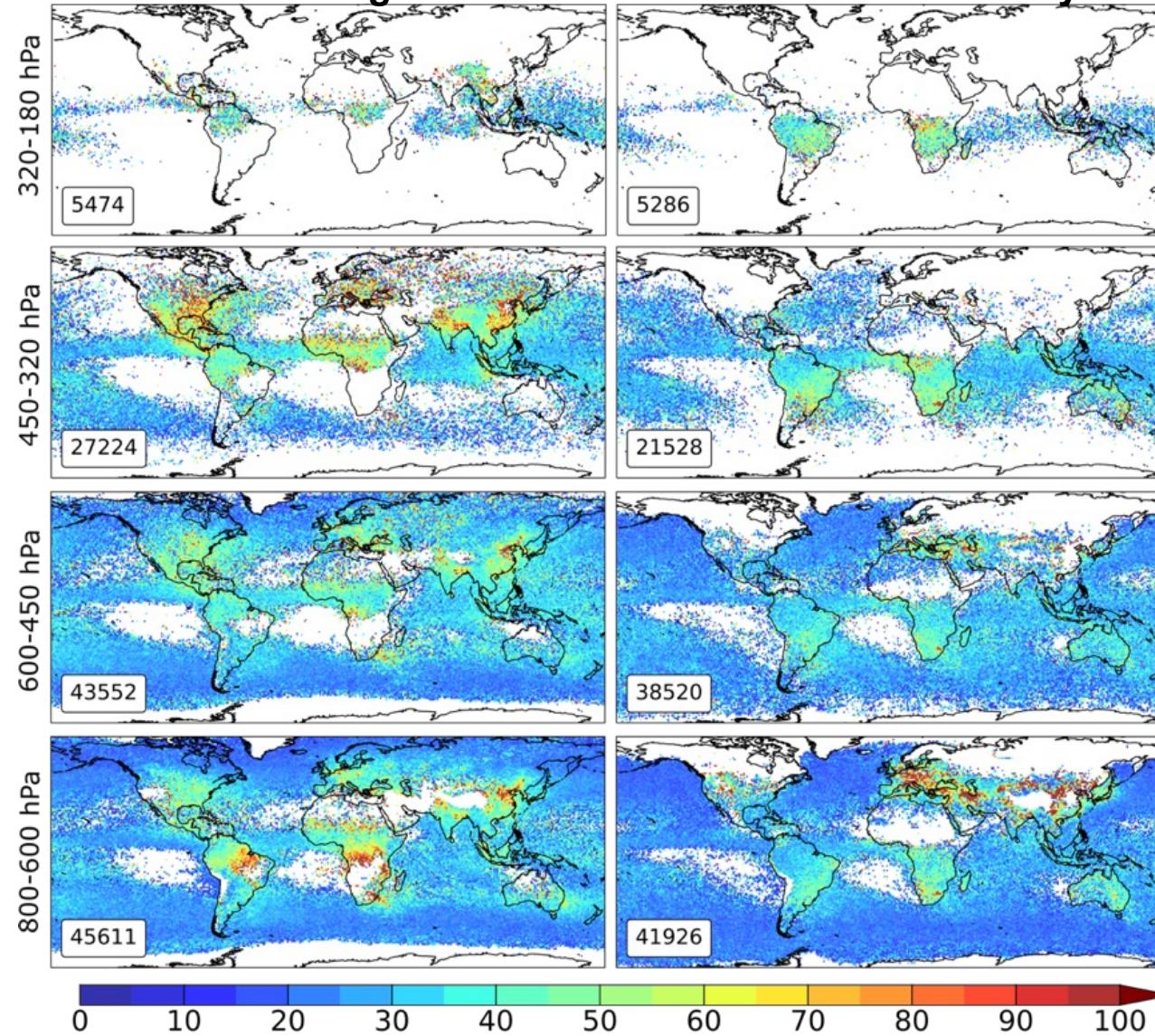
Addresses absence of routine  
observations

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

Seasonal multiyear mean NO<sub>2</sub> mixing ratios [pptv]

June-August

December-February

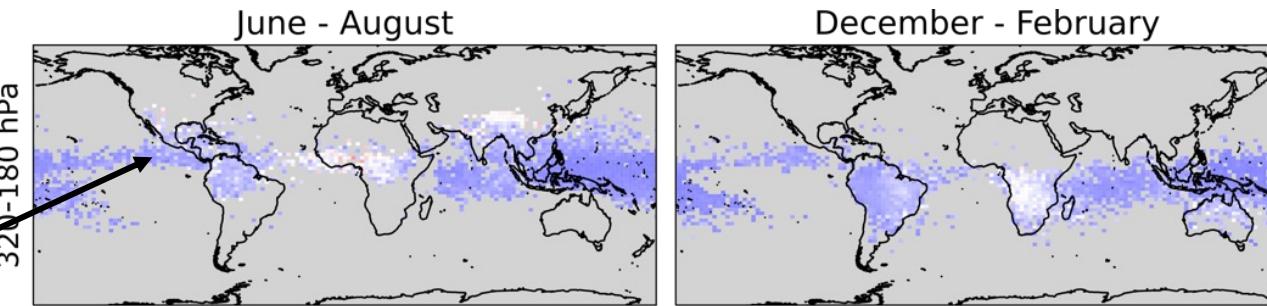


[Horner et al., submitted, ACP]

# Use New Datasets to Assess State of Knowledge

Difference between GEOS-Chem and cloud-sliced NO<sub>2</sub> [%]

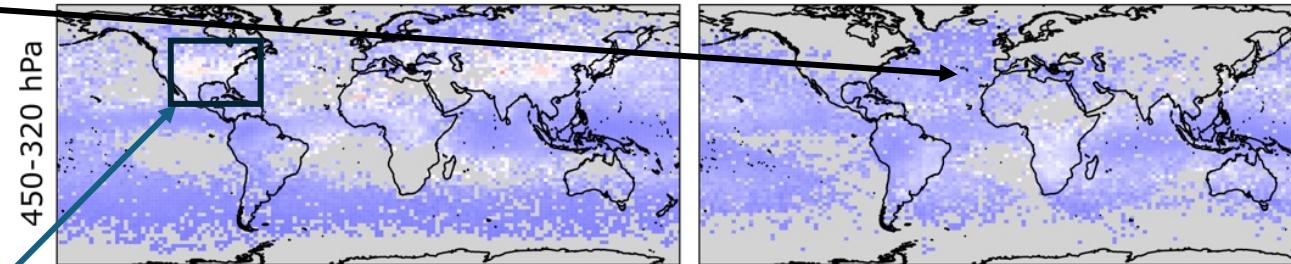
Model low bias over remote regions



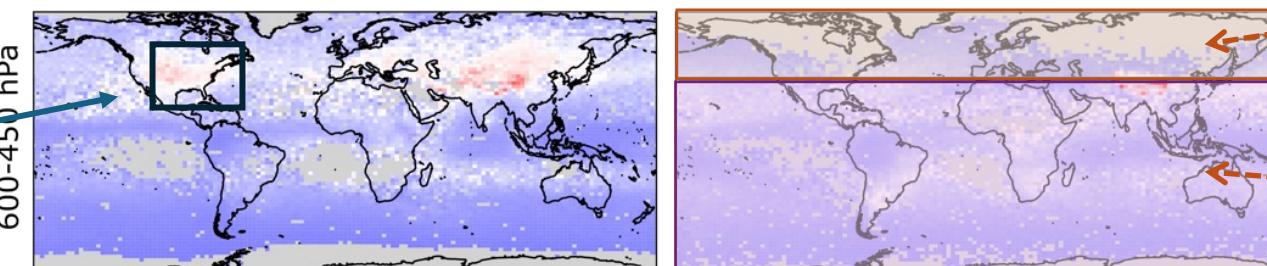
Model < Observed

Model > Observed

Model high bias  
over Southeast US  
(lightning NO<sub>x</sub>)

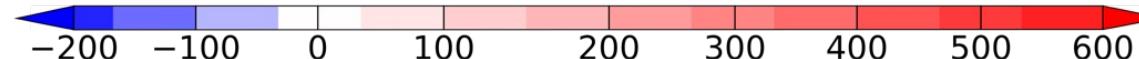
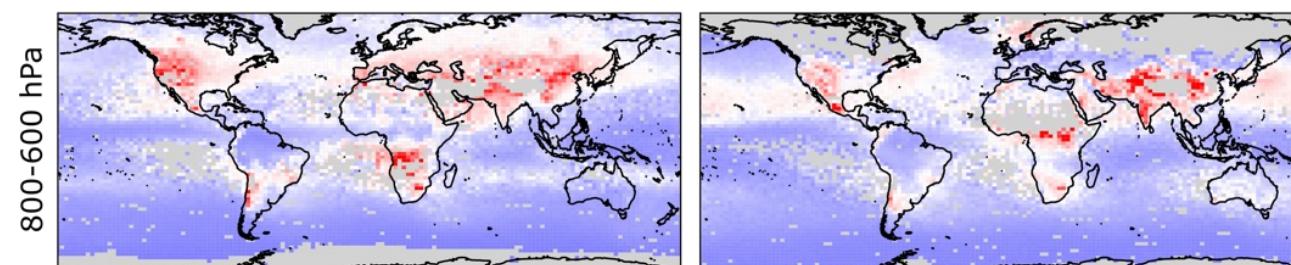


500 mol NO<sub>x</sub> per flash



35°N

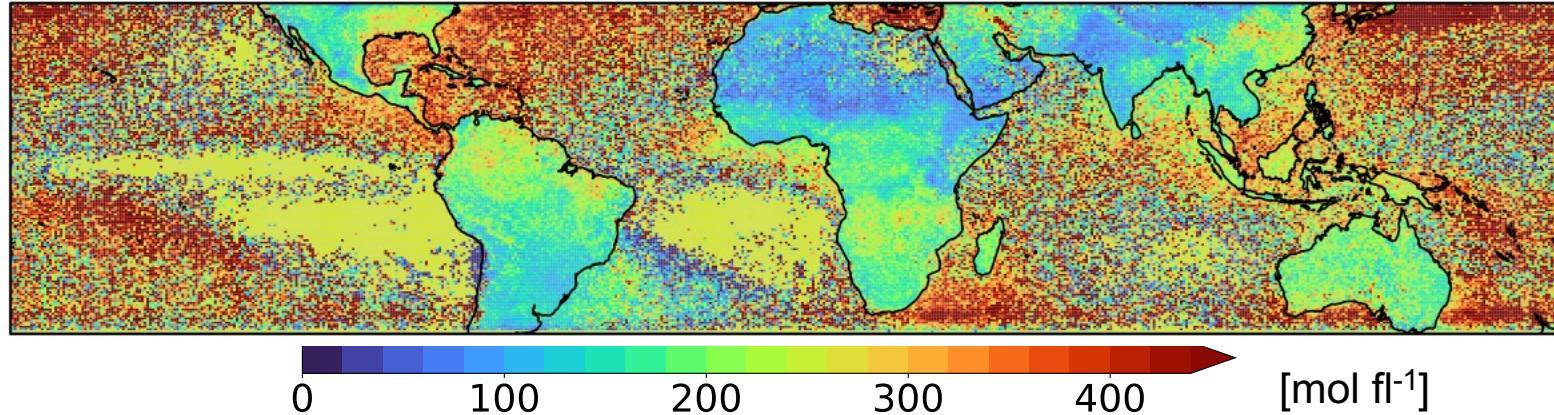
260 mol NO<sub>x</sub> per flash



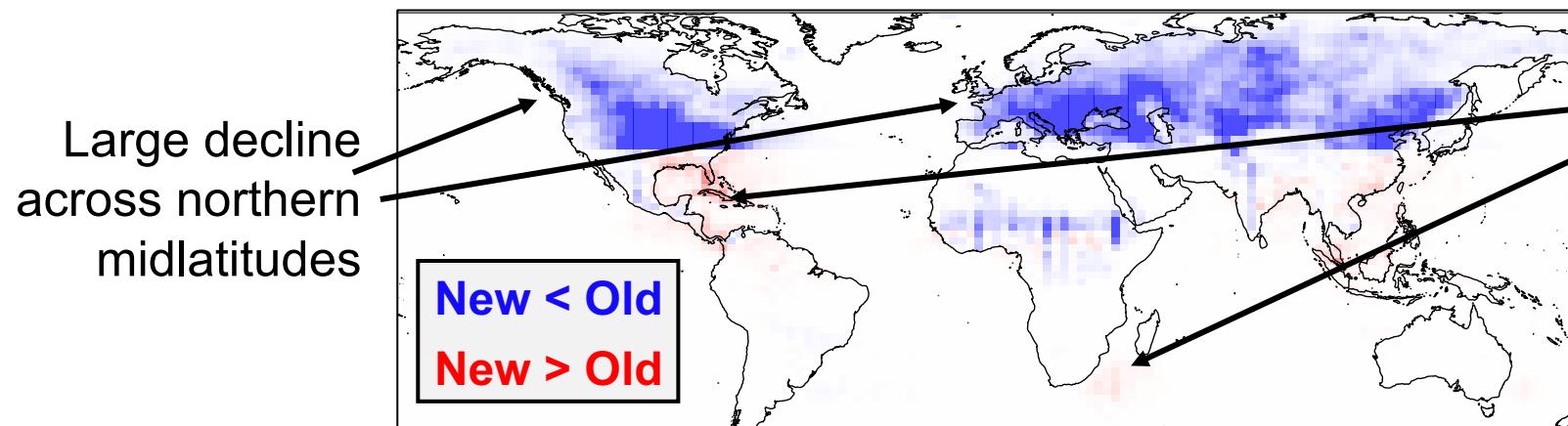
# Time- and Space-Resolved Lightning NO<sub>x</sub> Production

Use satellite observations of lightning flash energy to calculate hourly gridded production rates to GEOS-Chem lightning NO<sub>x</sub> emission inventory

Gridded lightning NO<sub>x</sub> production rates



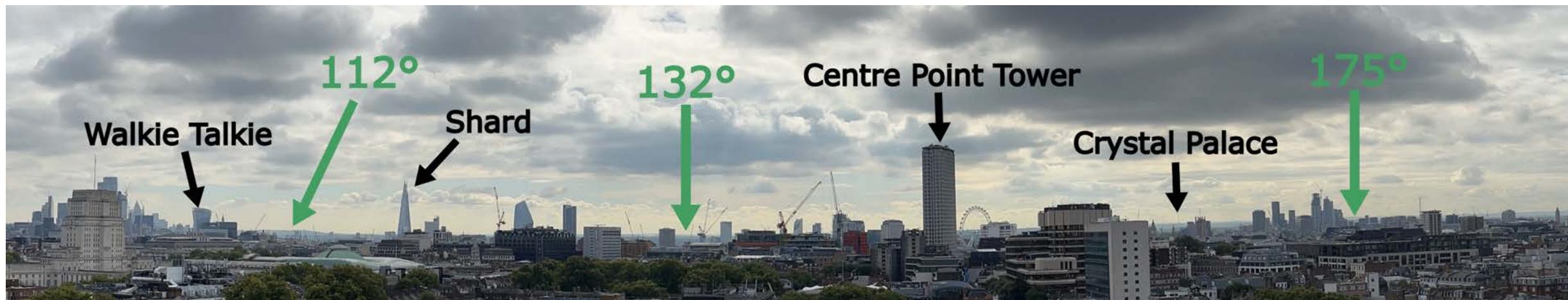
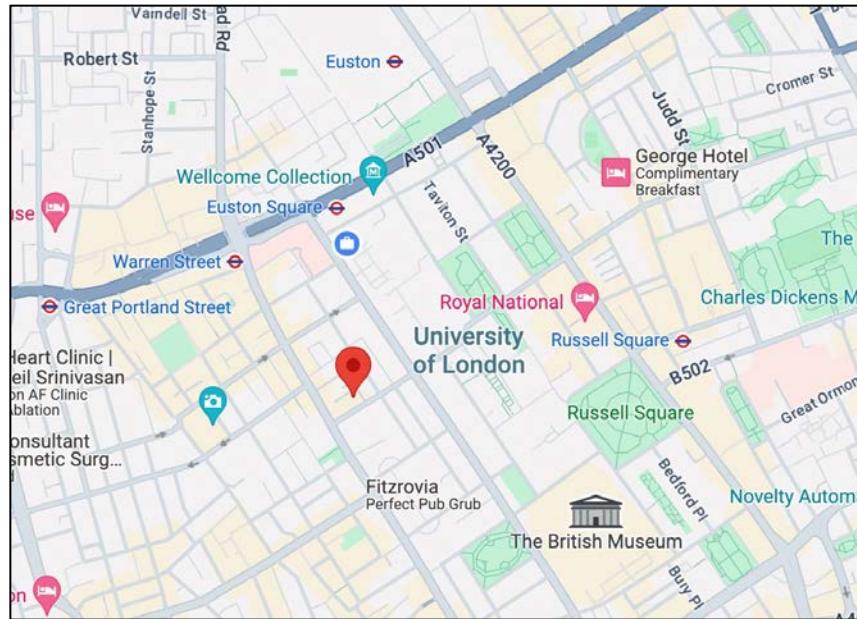
Effect of gridded lightning NO<sub>x</sub> production on emissions for single month (June)



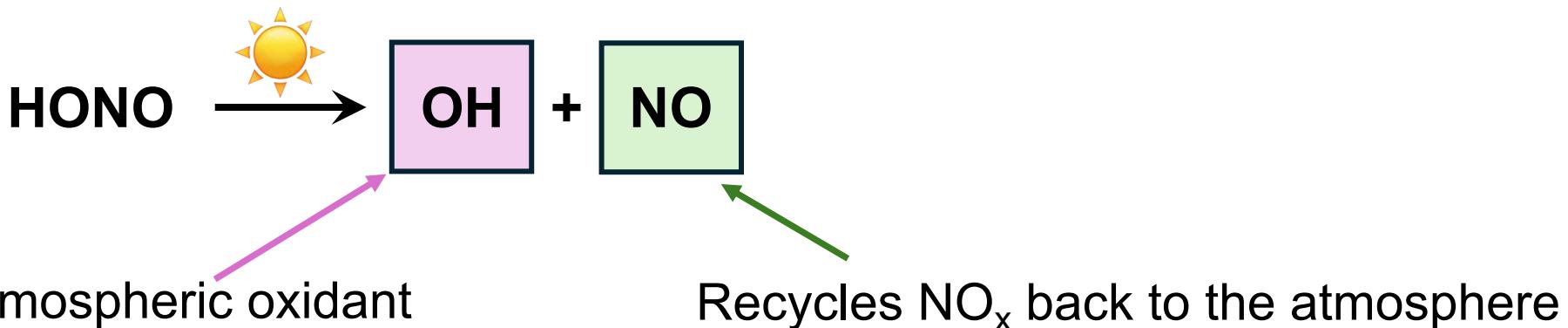
[Horner et al., *in progress*]

Next step is to implement emissions in GEOS-Chem to test effect on tropospheric NO<sub>x</sub> and ozone

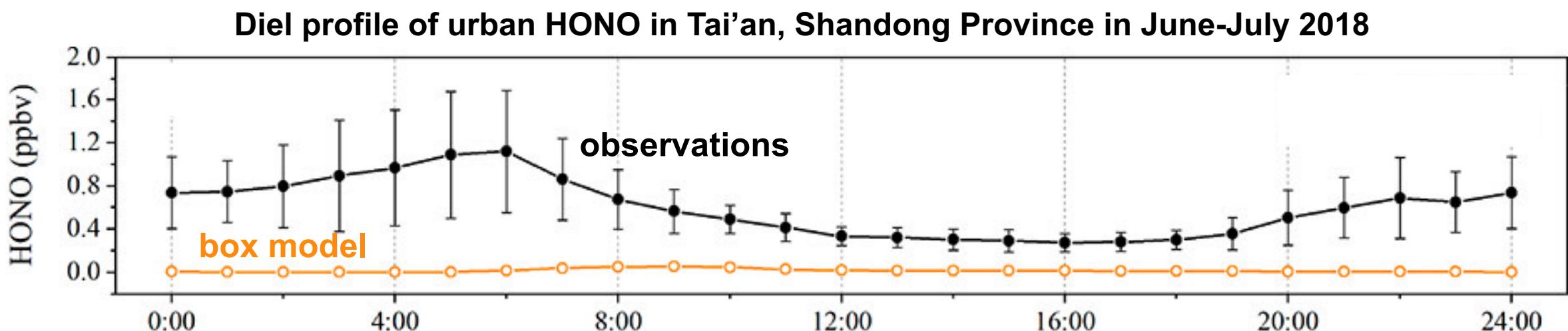
# Observations of Nitrous Acid (HONO) in Central London



# Observations of Nitrous Acid (HONO) in Central London

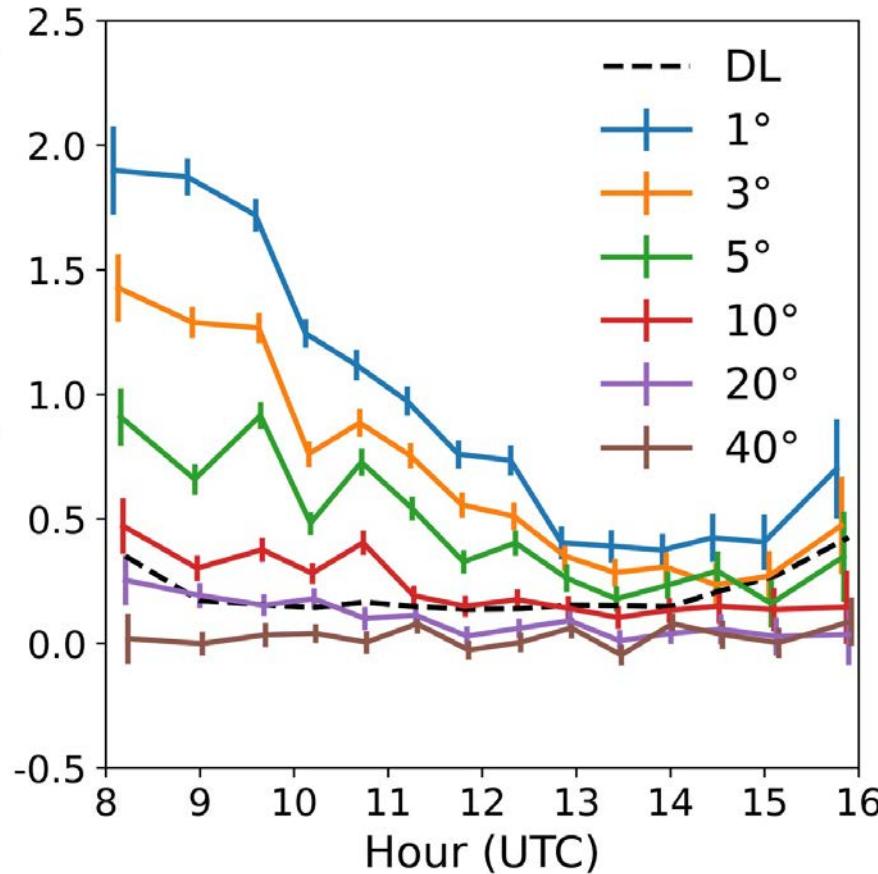


Best understanding of HONO (models) routinely fails to reproduce reality:



# Observations of nitrous acid (HONO) in Central London

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

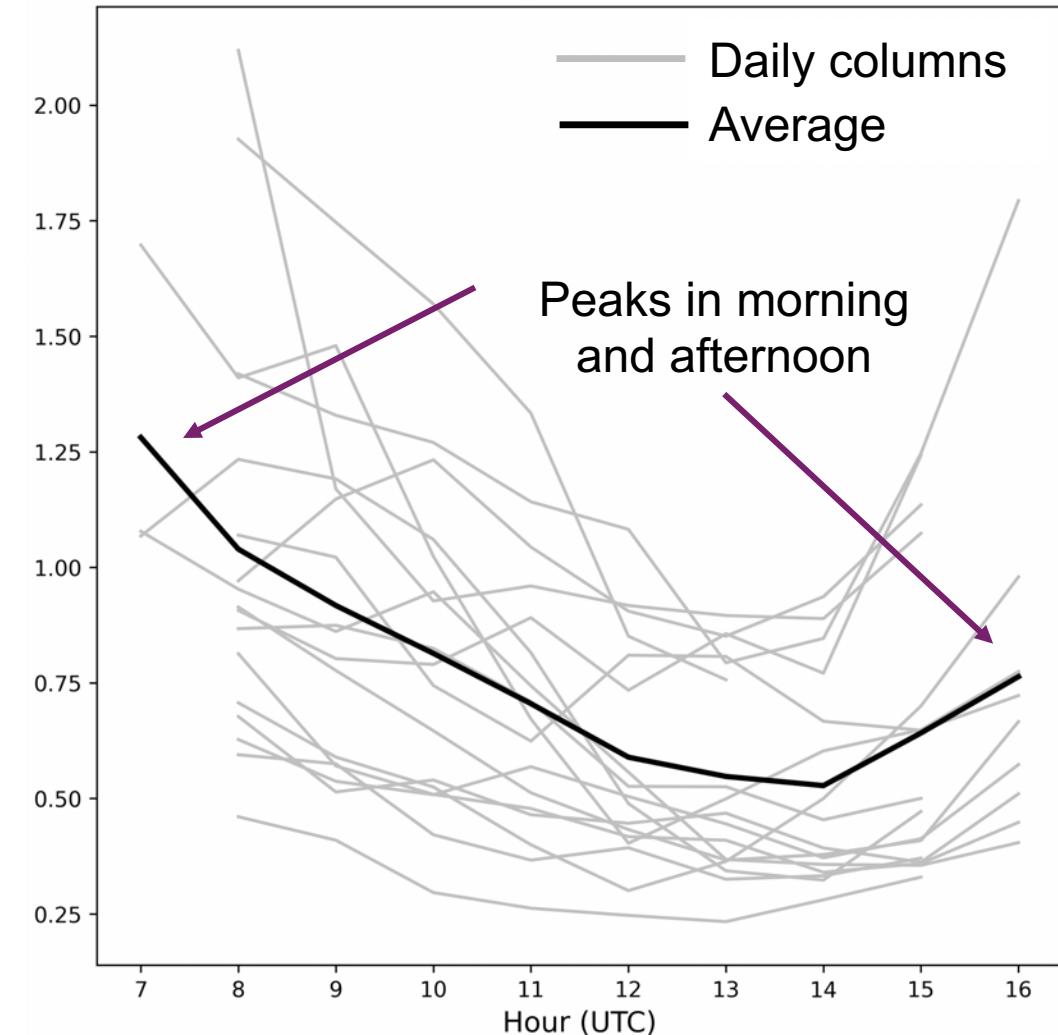


dSCDs: differential slant column densities

DL: detection limit

Good dSCDs separation → HONO present

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



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

# Summary and Outlook

- Use existing satellite observations of NH<sub>3</sub> to estimate UK agricultural emissions and assess the national inventory
- NH<sub>3</sub> publication most downloaded JGR-Atmospheres paper in 2022
- Led to investigation of contribution of NH<sub>3</sub> to UK urban PM<sub>2.5</sub> pollution and risk of NH<sub>3</sub> 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<sub>2</sub> and identify and address model shortcomings in representing lighting NO<sub>x</sub> 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>