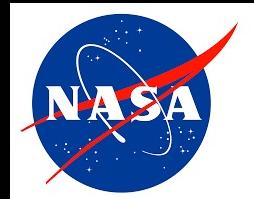


# Space-based constraints on reactive nitrogen:

From emissions of  $\text{NH}_3$  at the surface to  $\text{NO}_x$  in the upper troposphere



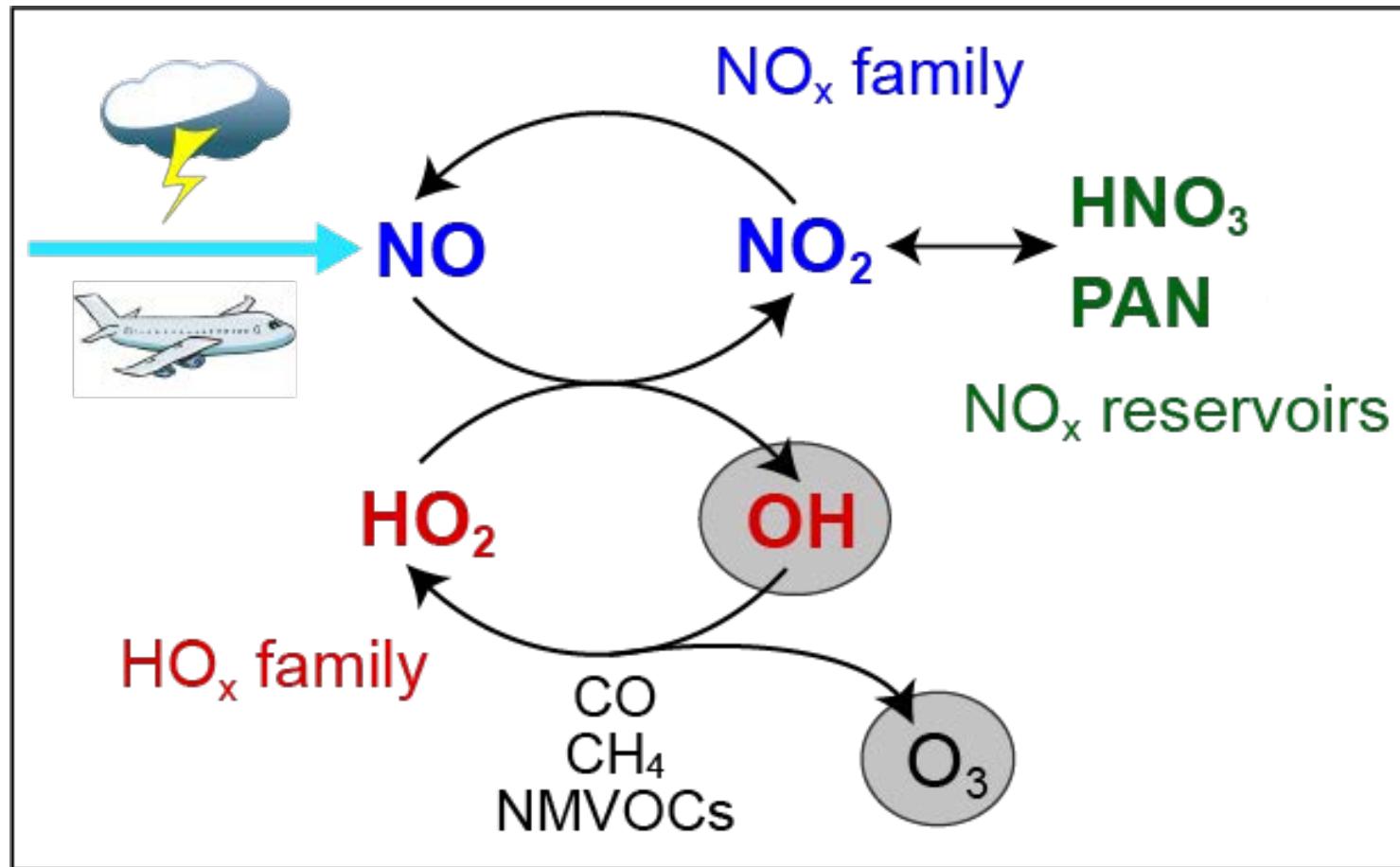
# First Estimate of Upper Tropospheric NO<sub>2</sub> from TROPOMI



Just published: <https://doi.org/10.5194/amt-14-2389-2021>

**Contributors:** J. F. Roberts, R. G. Ryan, H. Eskes, K. F. Boersma, S. Choi, J. Joiner, N. Abuhassan, A. Redondas, M. Grutter, A. Cede, L. Gomez, M. Navarro-Comas

# Nitrogen oxides ( $\text{NO}_x$ ) in the Upper Troposphere (8-12 km)



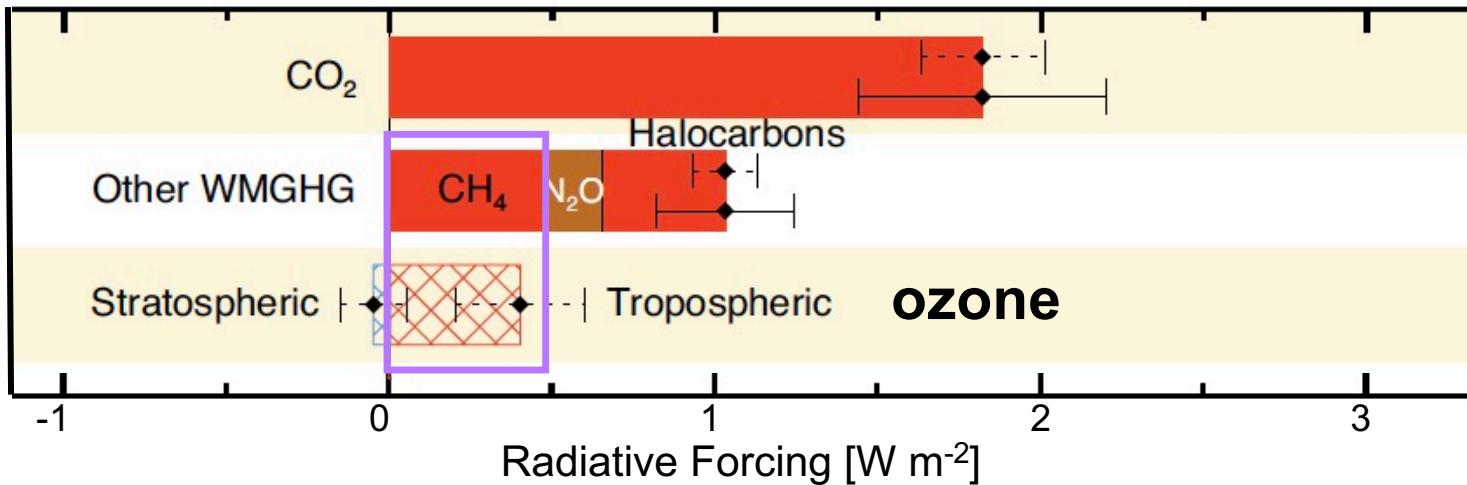
Influence atmospheric oxidants ( $\text{OH}$ ,  $\text{O}_3$ ) and climate ( $\text{O}_3$  formation, methane persistence)

**Other sources:** injection of surface pollution, rockets (?), long-range transport

# Why uncertainties in the upper troposphere matter

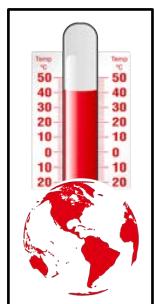
Warming due to tropospheric ozone is similar to that of methane ( $\text{CH}_4$ )

Tropospheric ozone and methane have near-equal climate impacts

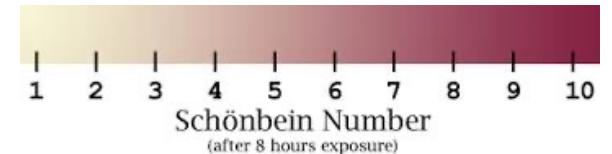


[IPCC AR5, 2013]

We are 100% reliant on models to estimate pre-industrial ozone



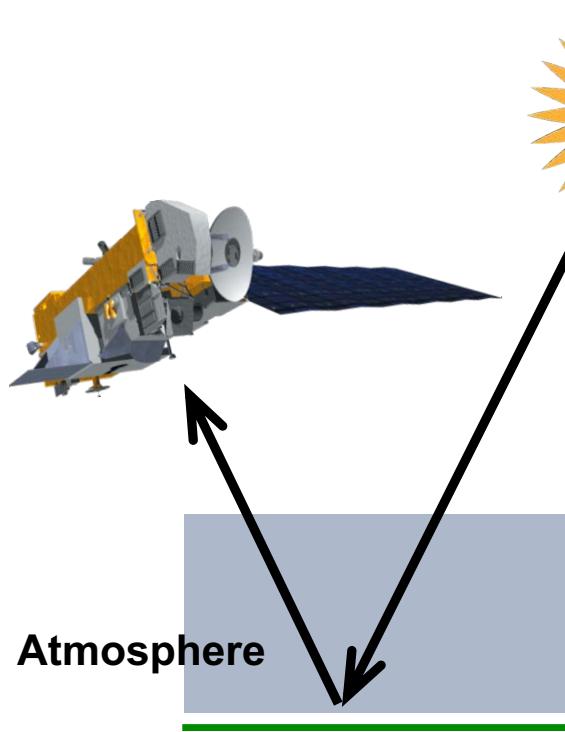
Errors in tropospheric ozone radiative forcing



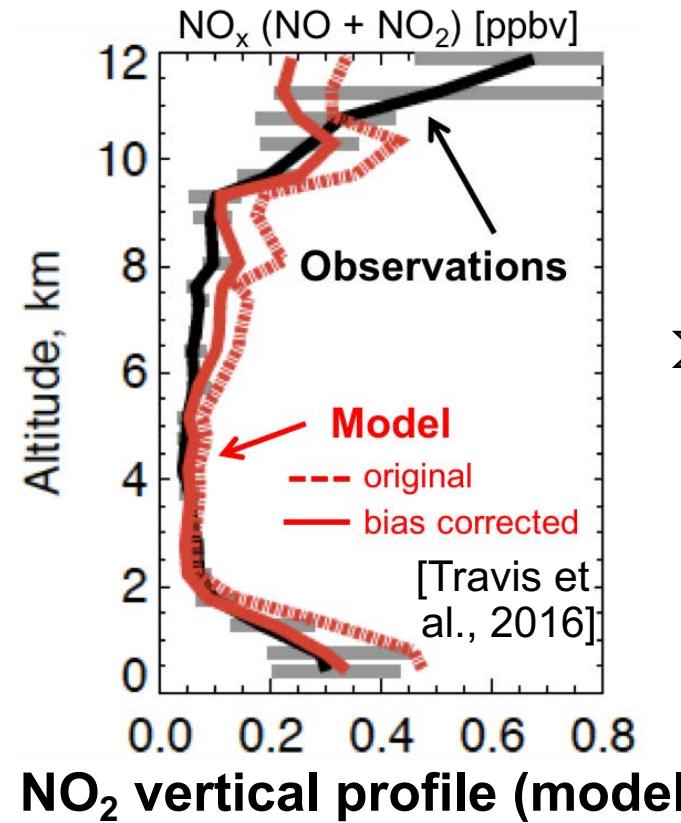
Indicator paper used to measure ozone in the mid to late 19<sup>th</sup> century

# Why uncertainties in the upper troposphere matter

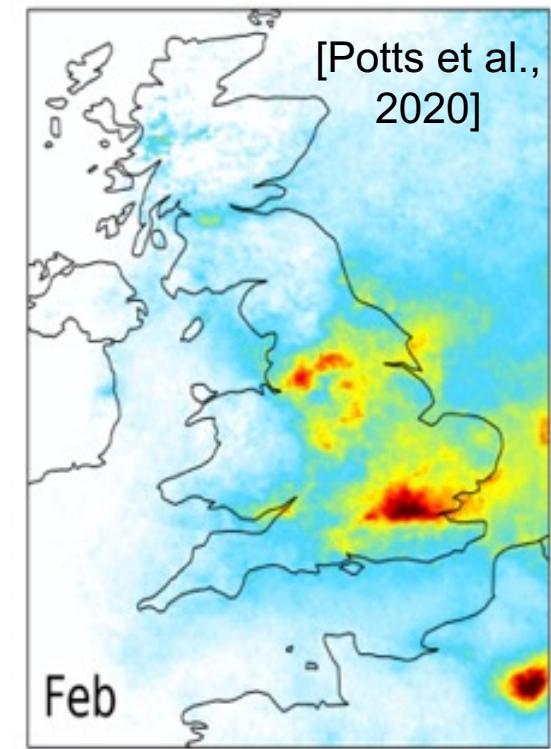
We rely on models to retrieve atmospheric composition from satellite instruments



**NO<sub>2</sub> along the viewing path**



**NO<sub>2</sub> vertical profile (model)**



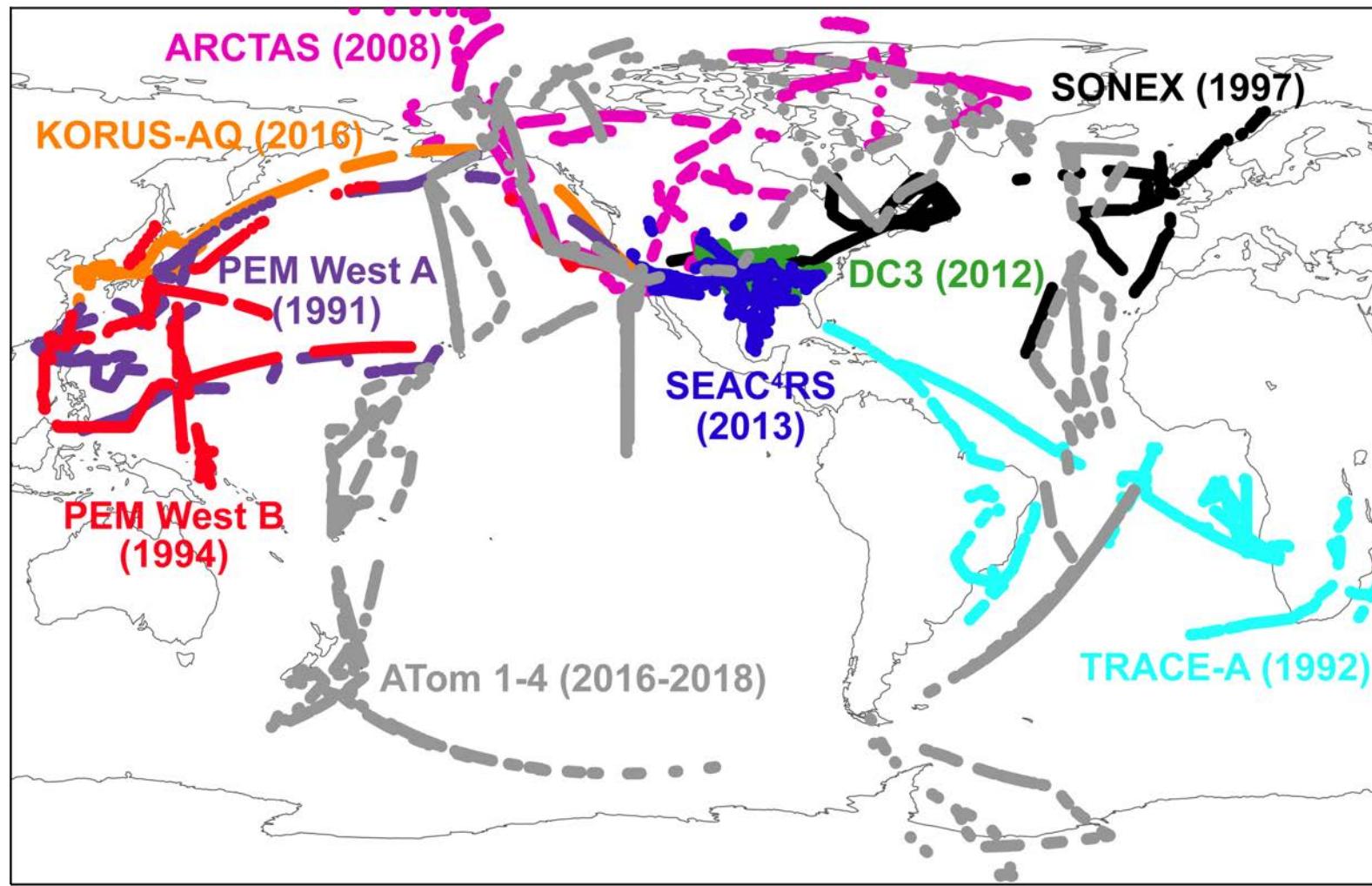
**Vertical column densities**



**Uncertain global air quality constraints from satellites**

# Sampling of the upper troposphere is limited

NASA DC8 research aircraft flight tracks in the upper troposphere



There are also measurements from commercial aircraft

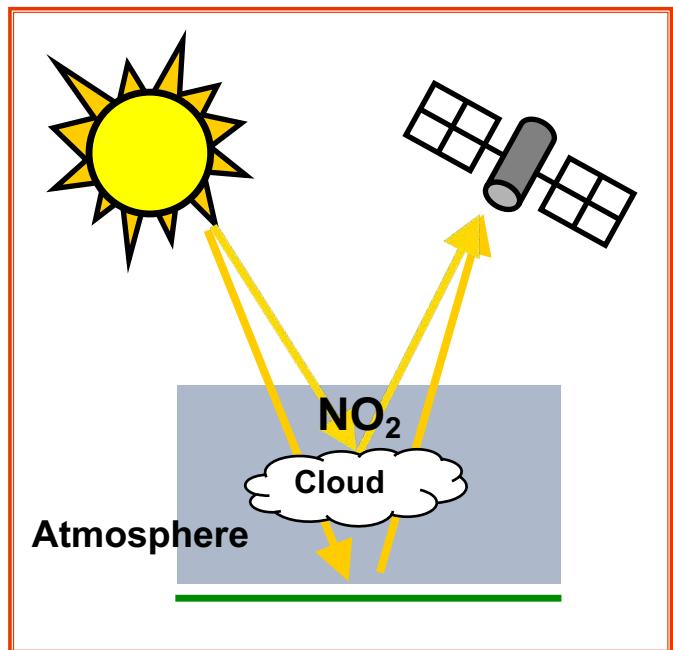
All use instruments that are susceptible to large biases in the upper troposphere

# Satellite products obtained using the cloud-slicing technique

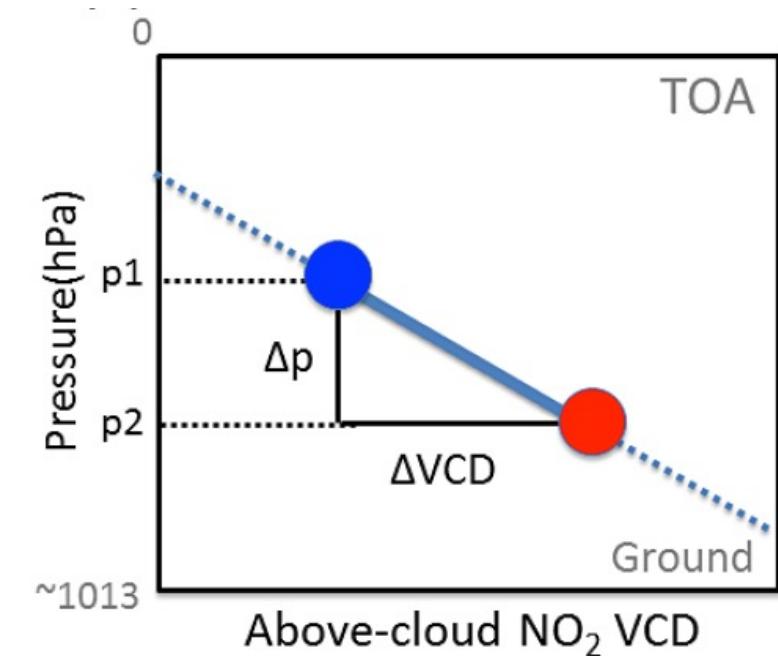
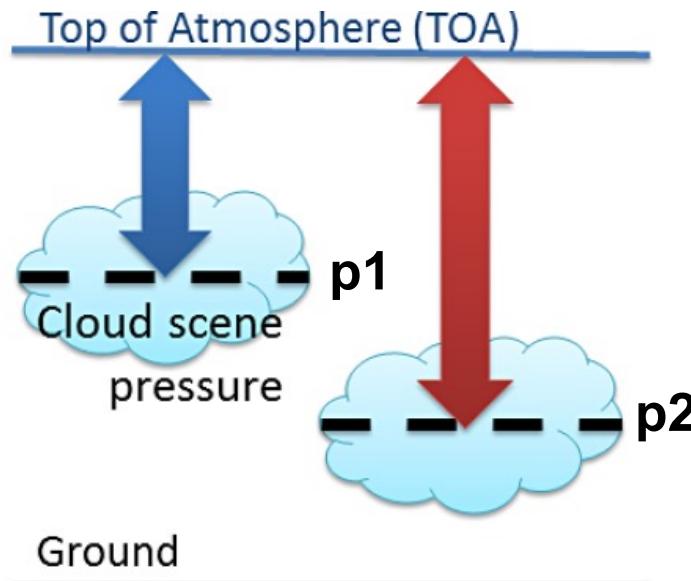
First applied by Ziemke et al. [2001] to TOMS ozone

Retrieve partial NO<sub>2</sub> columns over cloudy scenes at different heights

## APPROACH



## Use cloud height variability to derive partial columns



[adapted from Choi et al., 2014]

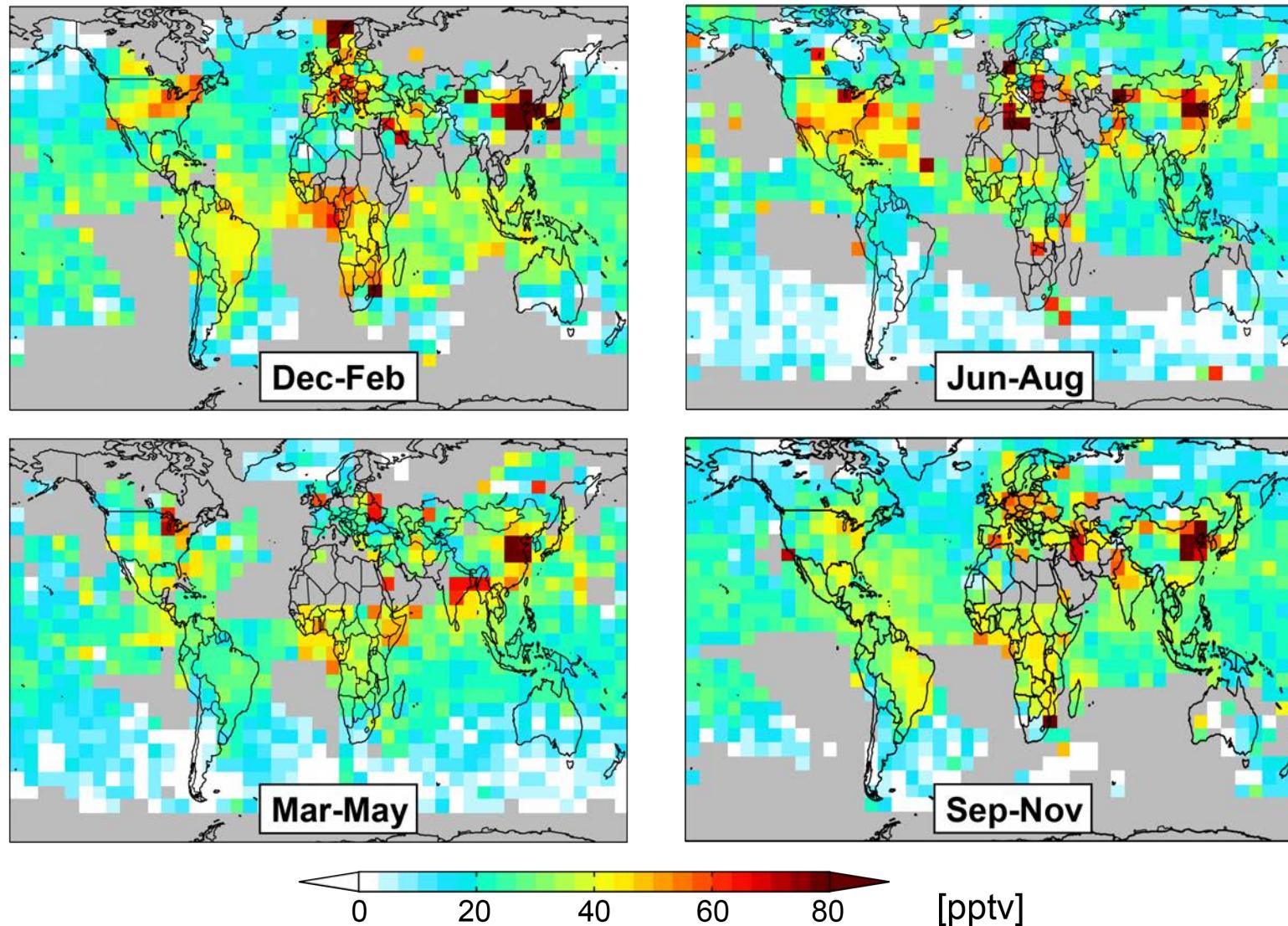
NO<sub>2</sub> volume mixing ratio (VMR) between clouds at p1 and p2

$$\text{NO}_2 \text{ VMR} = \frac{\Delta \text{VCD}}{\Delta p} \times \frac{k_B g}{R_{\text{air}}}$$

# Products of upper tropospheric NO<sub>2</sub> from satellite observations

Near global spatial coverage of seasonal mean UT NO<sub>2</sub> at  $5^\circ \times 8^\circ$  (50 km  $\times$  80 km)

NASA OMI upper troposphere NO<sub>2</sub> (2005-2007)

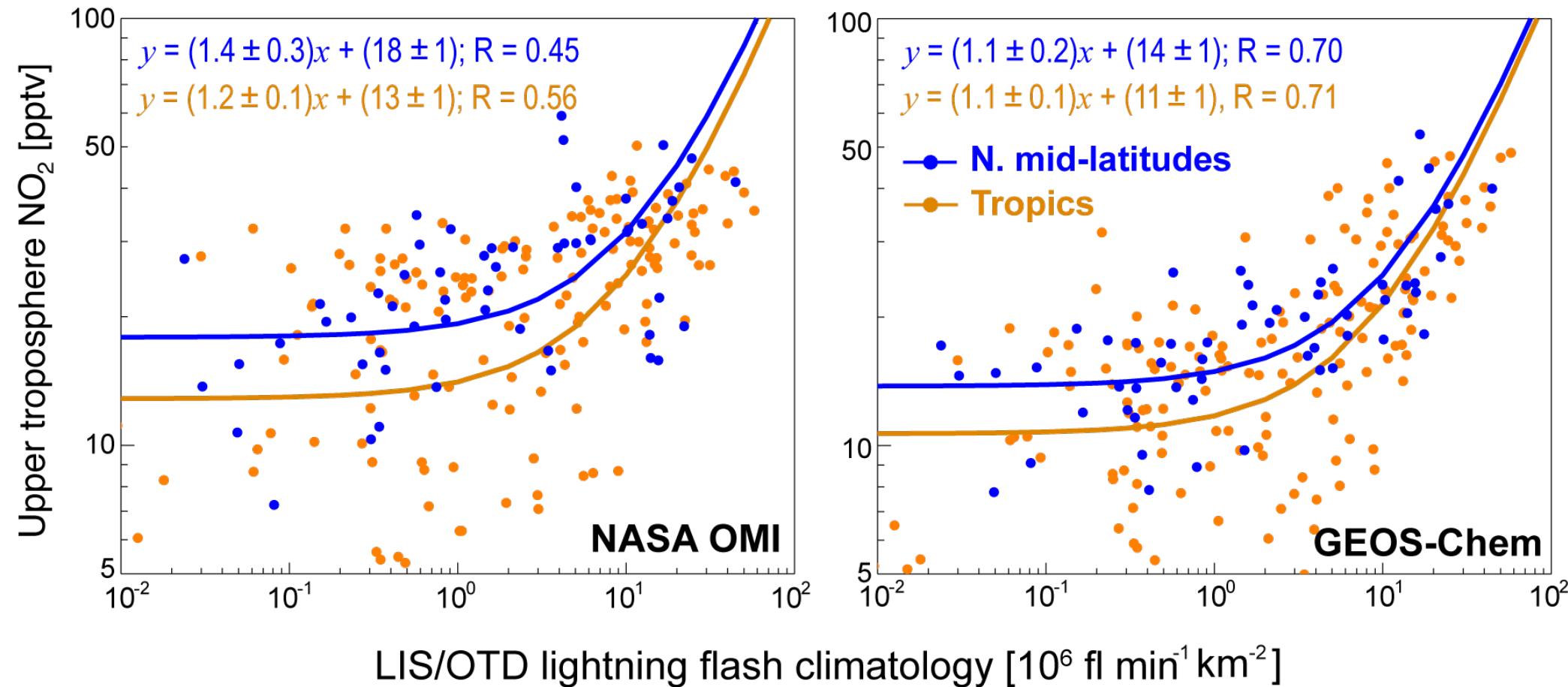


**OMI:**  
Ozone Monitoring  
Instrument

[Marais et al., 2018;  
Choi et al., 2014]

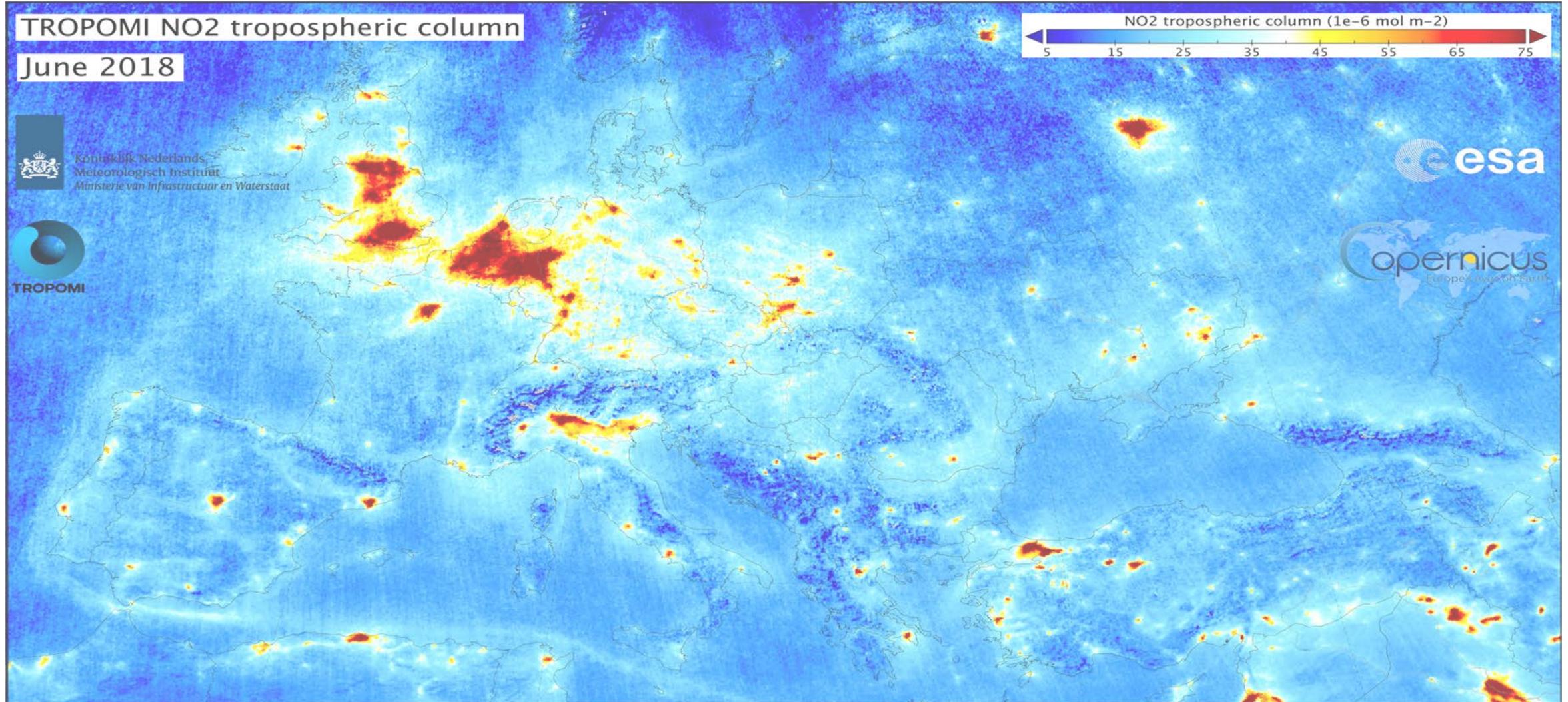
# Provide improved constraints on lightning NO<sub>x</sub> emissions

Log-log relationship between UT NO<sub>2</sub> from OMI and GEOS-Chem and satellite observations of lightning flashes in the **northern midlatitudes** and **tropics**



Similar slope in northern midlatitudes and tropics supports similar lightning NO<sub>x</sub> production rates

# Can we do better with high-resolution TROPOMI measurements?

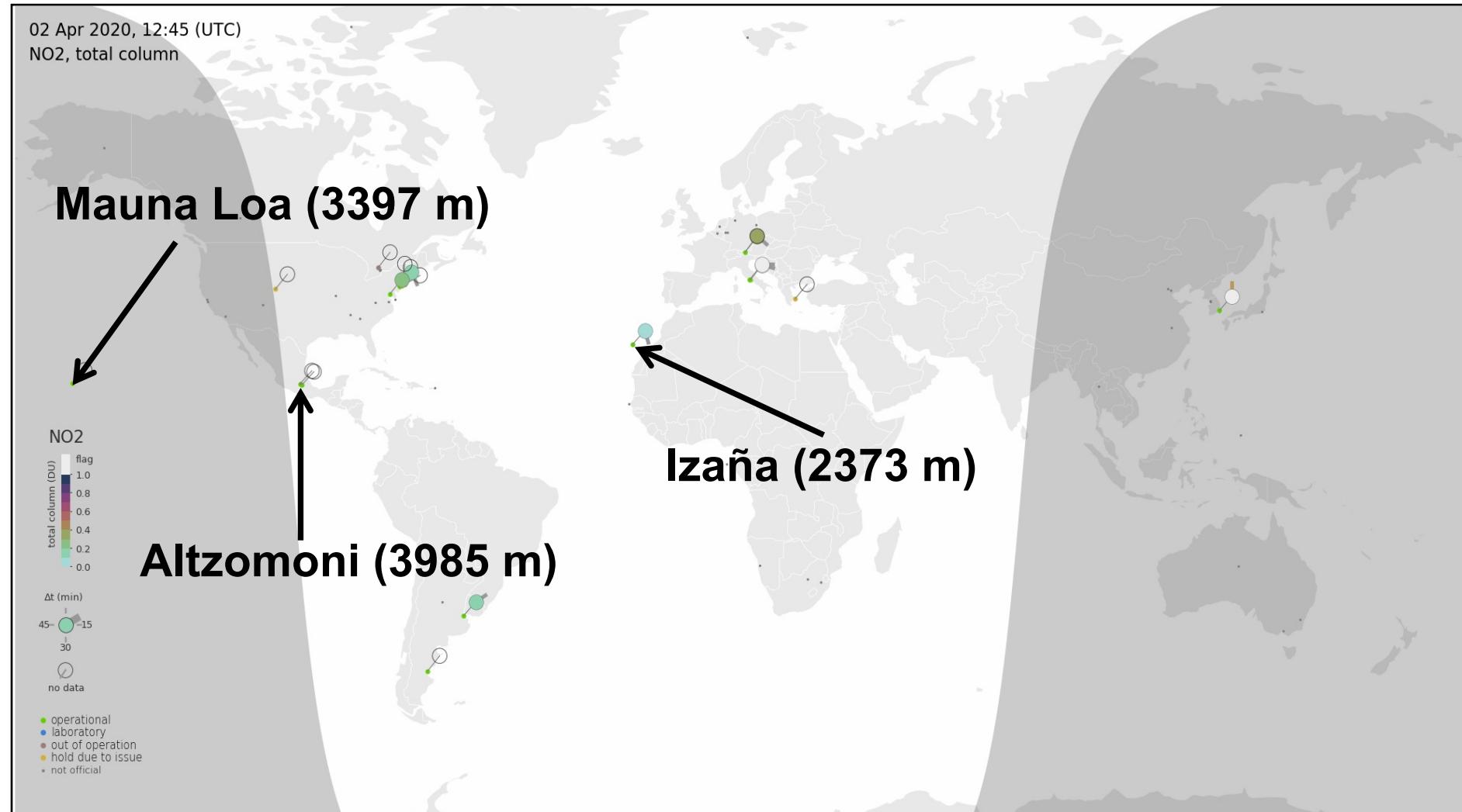


[Source: <http://www.tropomi.eu/data-products/nitrogen-dioxide>]

Nadir spatial resolutions in km (along × across): **13 × 24 (OMI)**; **5.6 × 3.5 (TROPOMI)**

# Evaluate TROPOMI with ground-based measurements

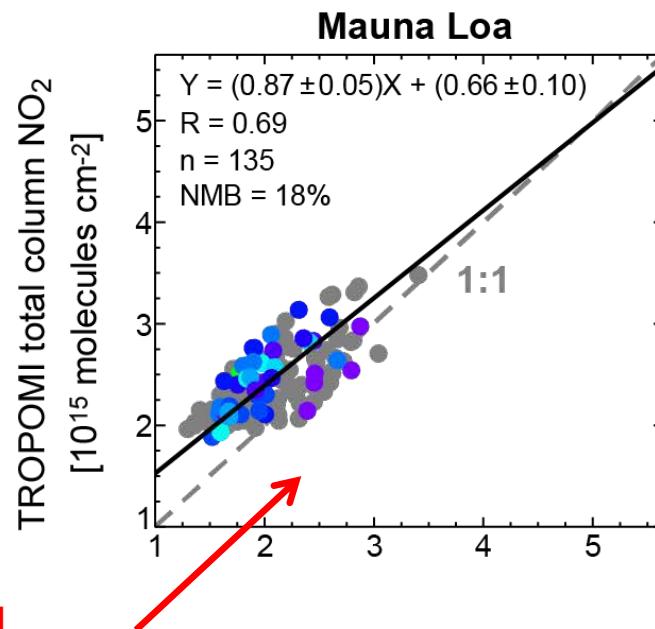
Global Pandora network, indicating locations of high-altitude sites (large relative contribution from the UT) used to evaluate TROPOMI



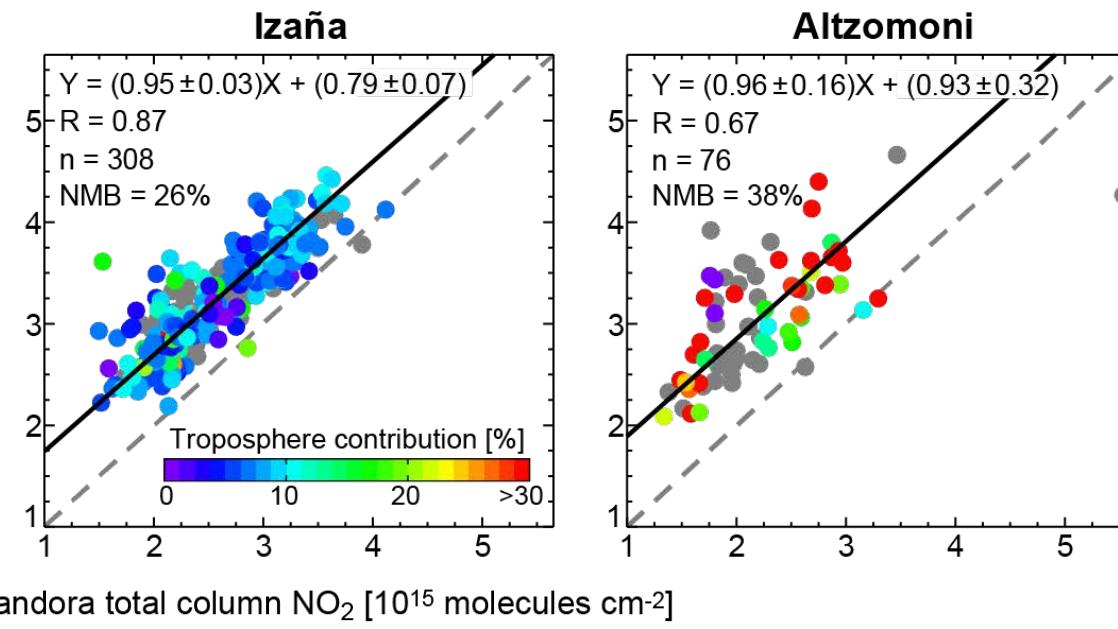
[Source: <https://www.pandoria-global-network.org/>]

# Evaluate TROPOMI with ground-based measurements

## Stratosphere:



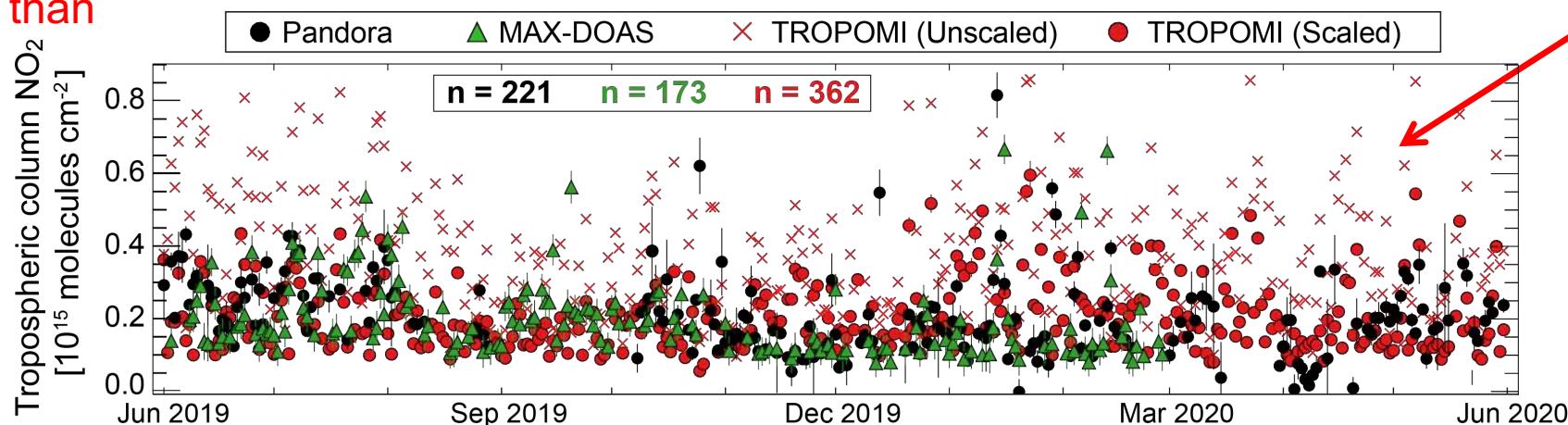
## Stratosphere + free troposphere:



TROPOMI stratospheric column variance is less than Pandora

TROPOMI free tropospheric column is more than Pandora and MAX-DOAS

## Free troposphere:



# TROPOMI cloud-sliced upper tropospheric NO<sub>2</sub>

Seasonal mean UT  
NO<sub>2</sub> mixing ratios  
from 2 distinct cloud  
products at 1° × 1°

Range: 30-80 pptv

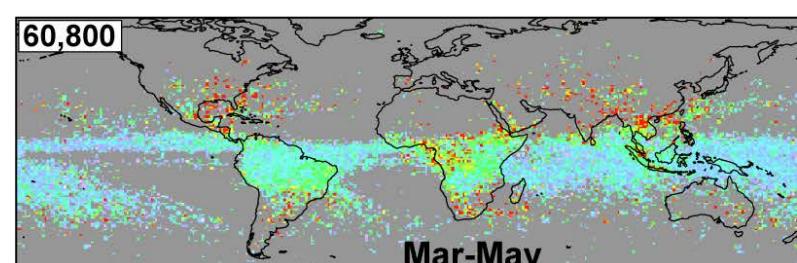
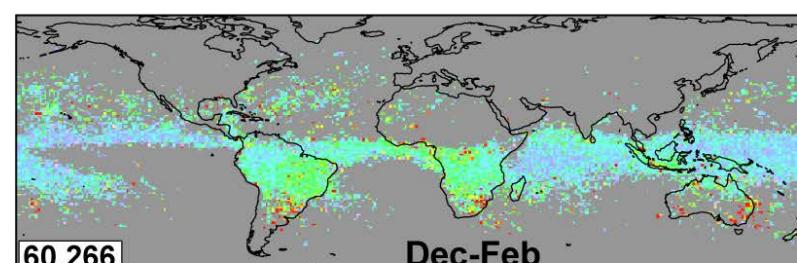
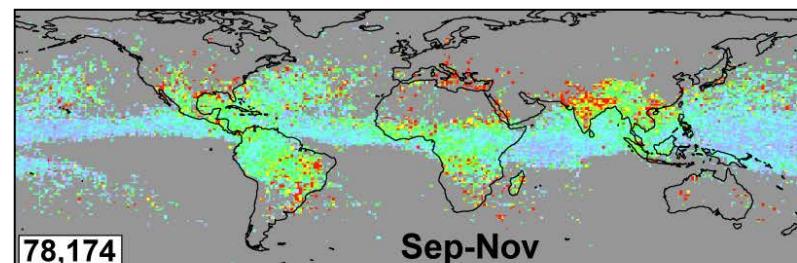
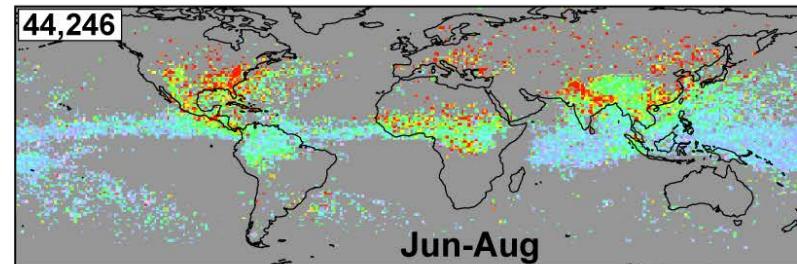
Background: ~30 pptv

Cloud products give  
similar UT NO<sub>2</sub> in the  
tropics

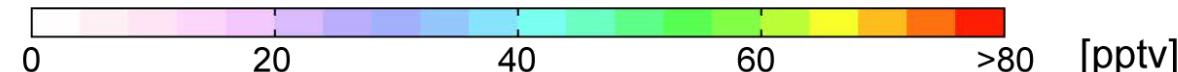
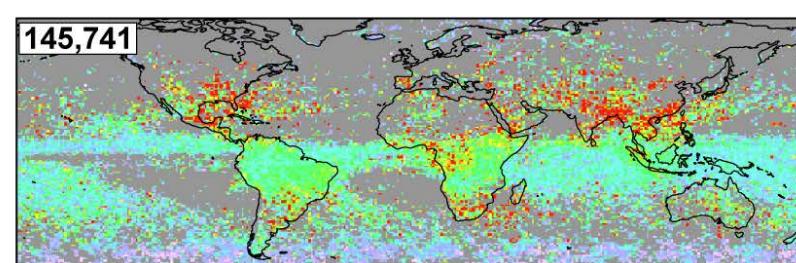
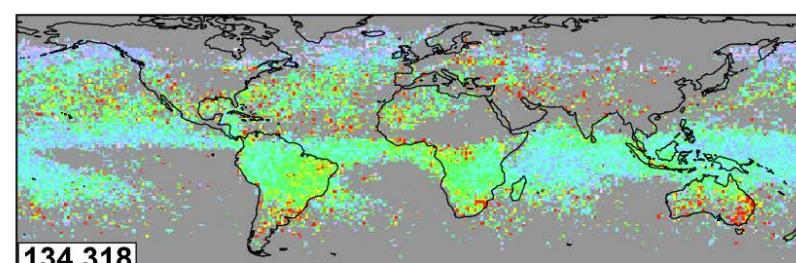
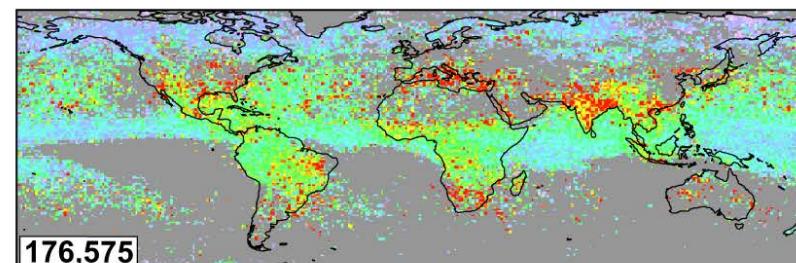
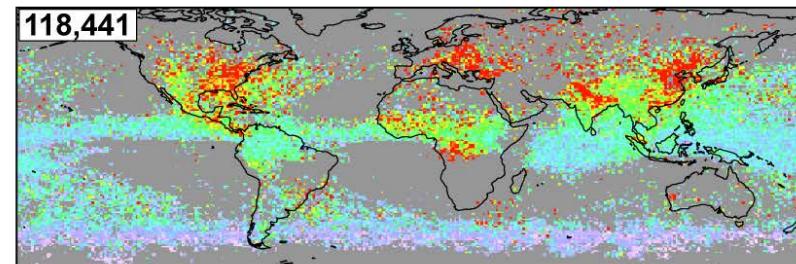
Greater coverage with  
ROCINN-CAL

Some contamination:  
Australia (fires), North  
China (pollution)

Cloud Product One (FRESCO-S)

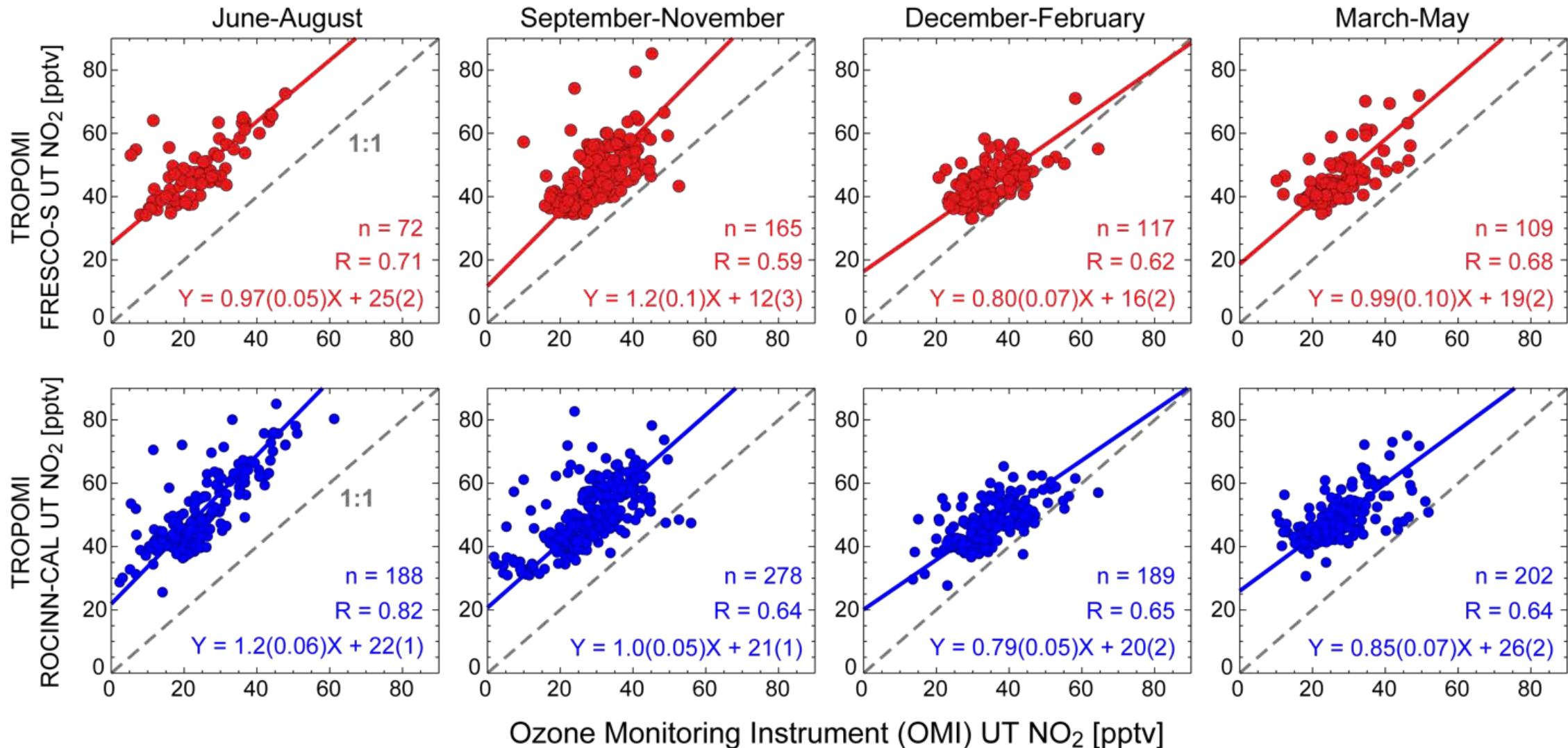


Cloud Product Two (ROCINN-CAL)



# Comparison to the NASA OMI product

TROPOMI UT NO<sub>2</sub> obtained at 1° × 1° and gridded to the NASA product resolution (8° × 5°)

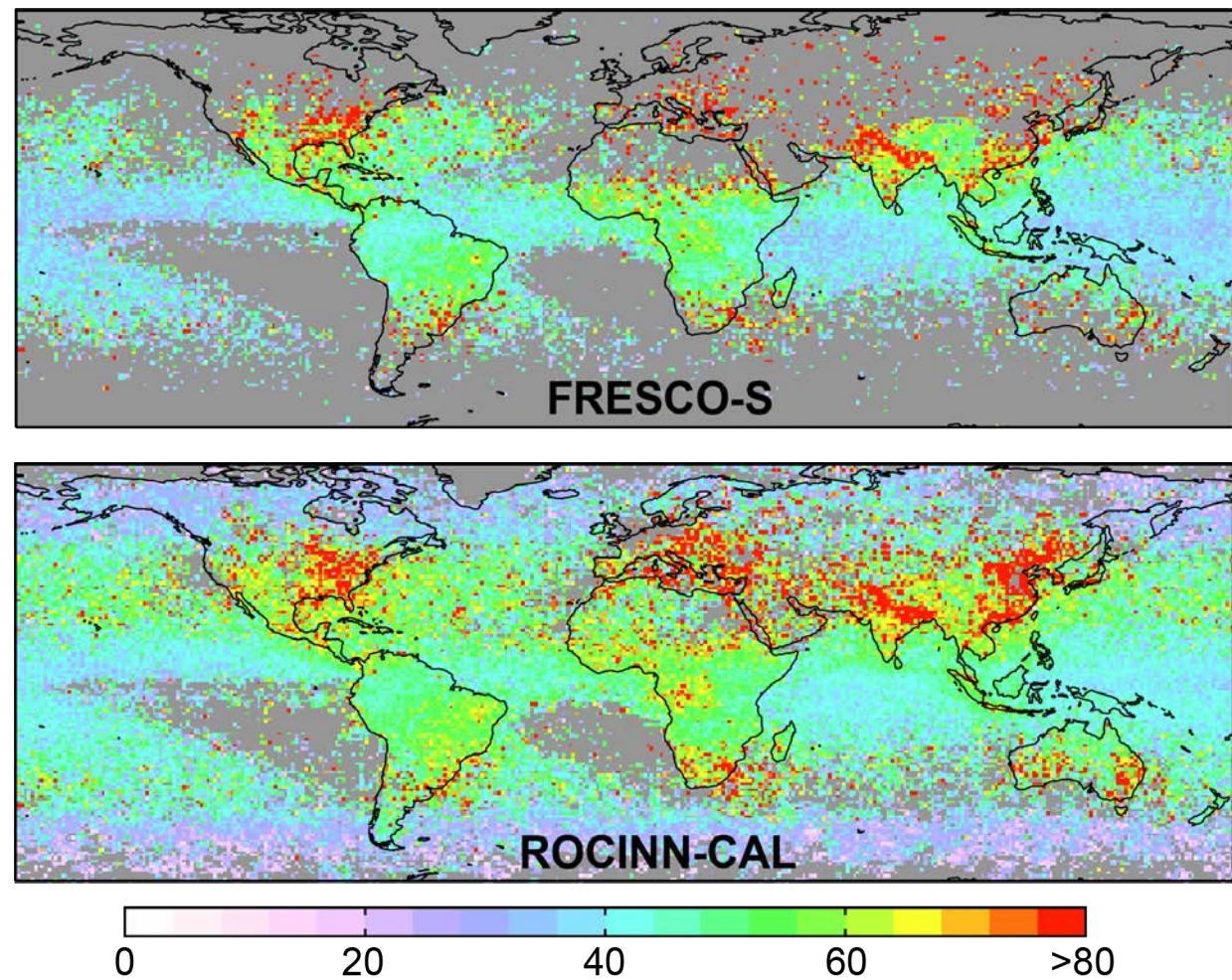


Spatial consistency. TROPOMI is 12-26 pptv more than OMI (retrieval, clouds, vertical NO<sub>2</sub> profile)

# Concluding Remarks for TROPOMI UT NO<sub>2</sub>

- Developed seasonal mean UT NO<sub>2</sub> from TROPOMI using cloud-slicing at finer resolution than ever before
- Two datasets from distinct cloud products
- Differences in cloud products, particularly in higher latitudes, lead to differences in coverage
- Consistency with existing OMI product
- Potential to address data sparsity in the UT
- We're using TROPOMI UT NO<sub>2</sub> to detect the influence of aircraft on UT NO<sub>2</sub> and quantify uncertainties in reactive nitrogen in the UT with GEOS-Chem

TROPOMI annual mean upper tropospheric NO<sub>2</sub> [pptv]



Interested in using the TROPOMI UT NO<sub>2</sub> product? Contact me: [e.marais@ucl.ac.uk](mailto:e.marais@ucl.ac.uk)

**Back to Earth's surface...**

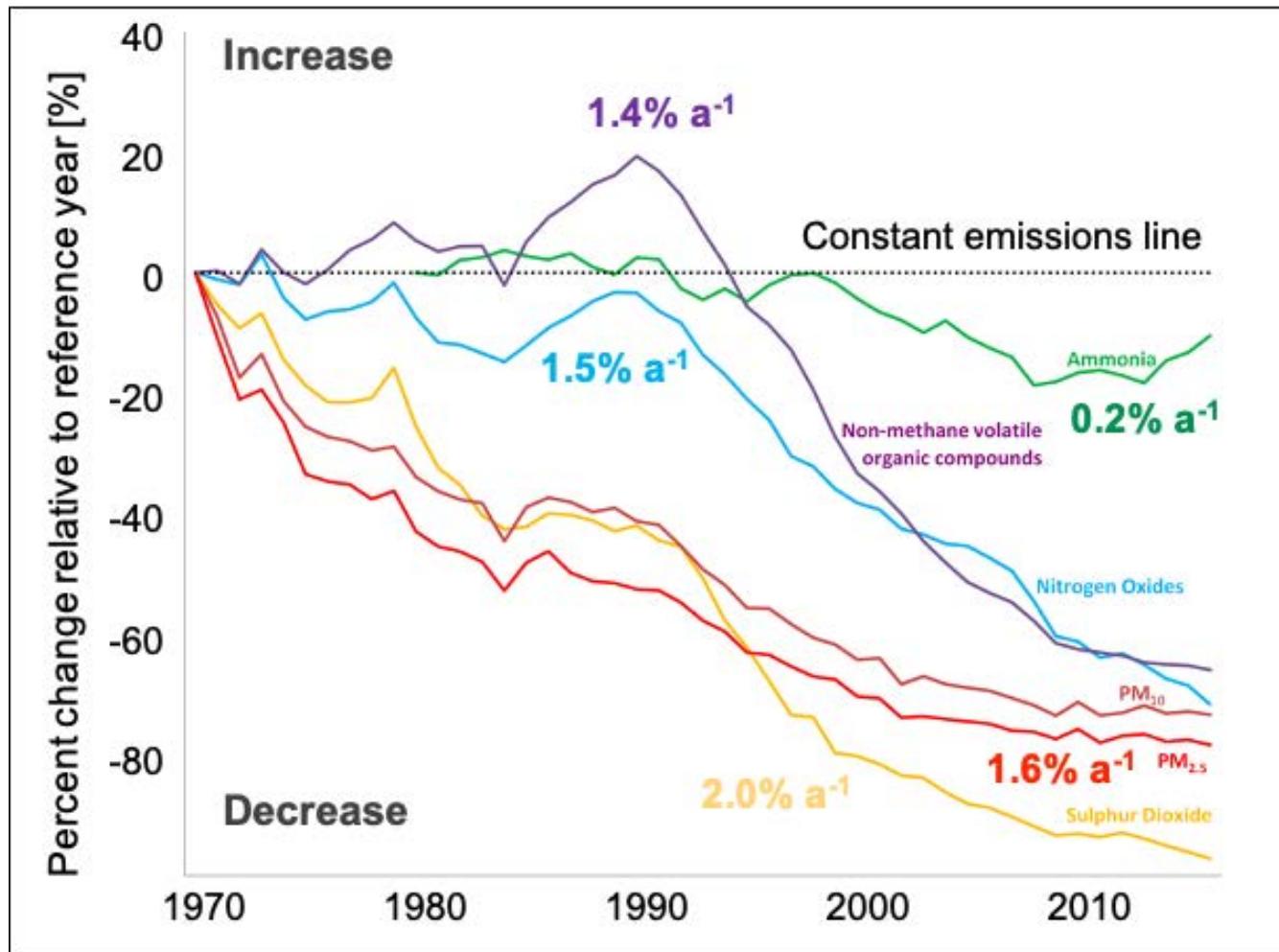
# Top-down estimate of UK ammonia emissions



**Contributors:** A. Pandey, M. Van Damme, L. Clarisse, P. F. Coheur, M. Shephard, K. Cady-Perreira, T. Misselbrook, L. Zhu, F. Yu, G. Luo

# Ammonia emissions in the UK: the bottom-up perspective

## Temporal (Time) Variability in Emissions

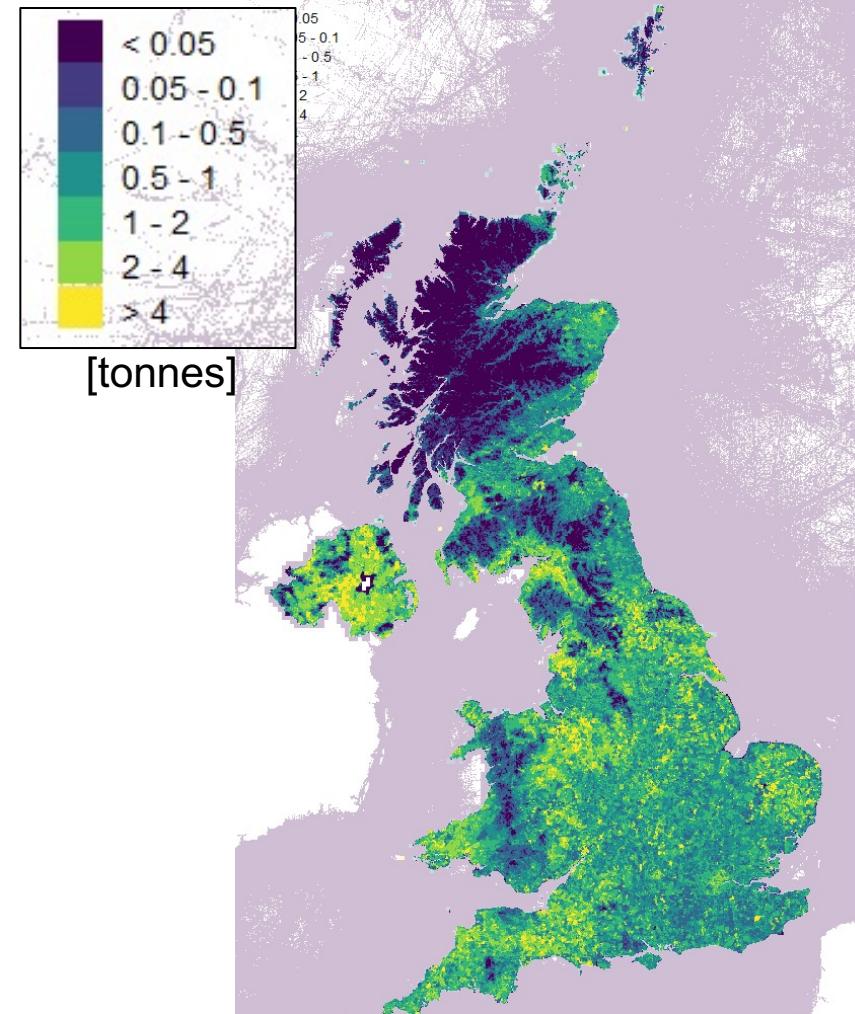


[Adapted from Defra, 2018]

Successful decline in all emissions, except ammonia (NH<sub>3</sub>)

## Spatial Variability in Emissions

### NH<sub>3</sub> emissions for 2018 at 1 km

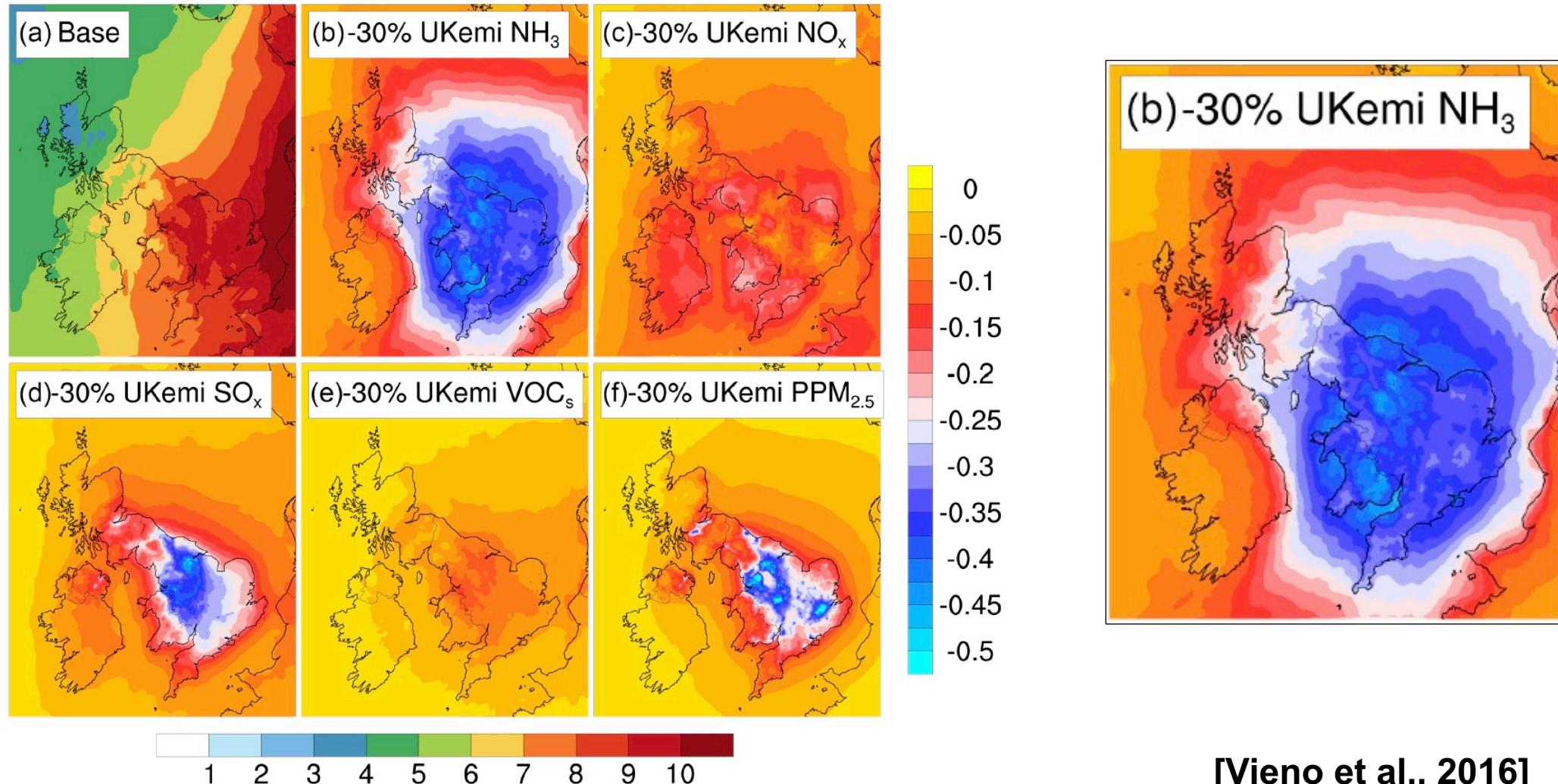


All maps © Crown copyright. All rights reserved Defra, Licence number 100022861 (2020) and BEIS, Licence number 100037028 (2020) LPS © Crown copyright and database right 2020 Licence INSP594

[Adapted from <https://naei.beis.gov.uk/data/>]

# Ammonia impact on air pollutants hazardous to health

## Effect of precursor emission reductions on 2010 PM<sub>2.5</sub>



Largest and most extensive decline in PM<sub>2.5</sub> achieved by targeting ammonia sources

# Top-down emissions estimated with satellite observations

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

**ABUNDANCES**

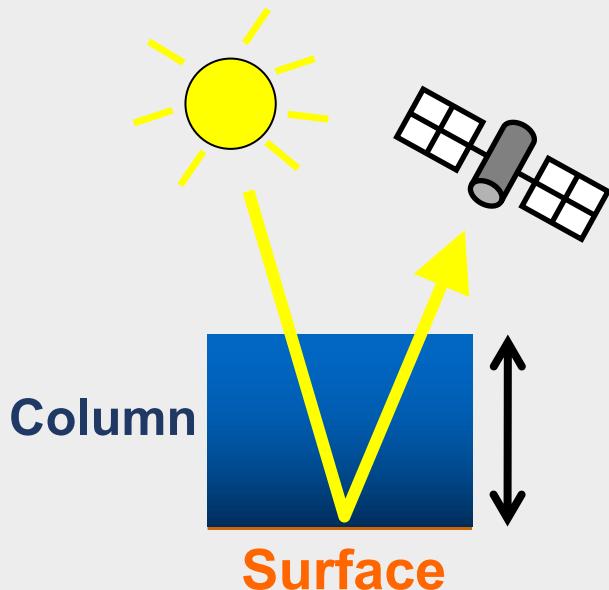


**Conversion Factor**

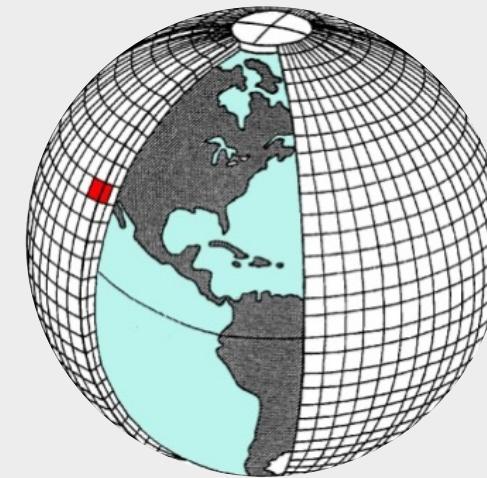


**EMISSIONS**

**Satellite columns  
(IASI)**



**Column-to-Emission ratio  
(GEOS-Chem)**



**Satellite-derived  
Surface Emissions**

**Emission**



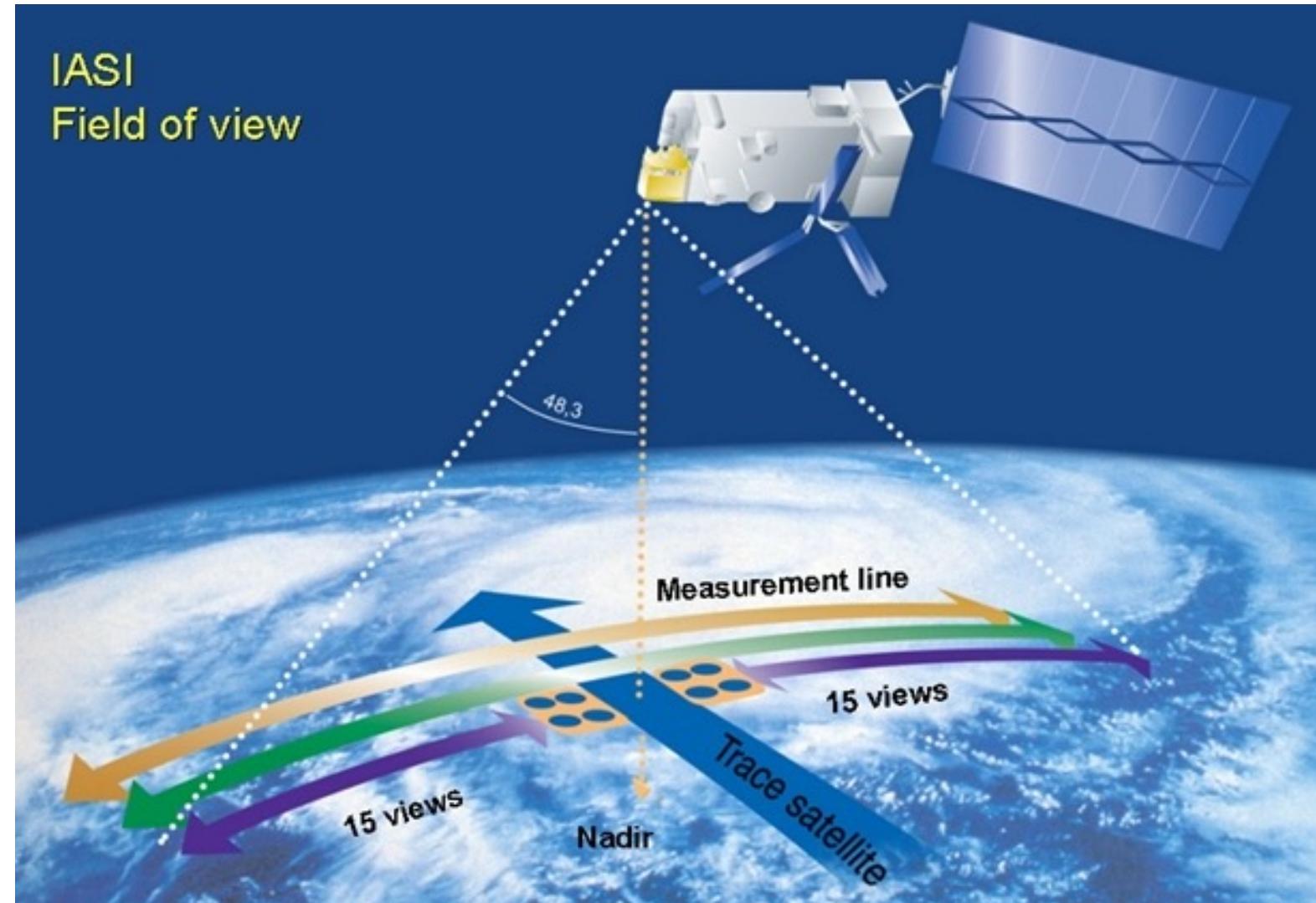
# Infrared Atmospheric Sounding Interferometer (IASI) Instrument

Overpass:  
9:30 local solar time

Spatial resolution:  
12 km to 39 km

Swath width:  
2200 km

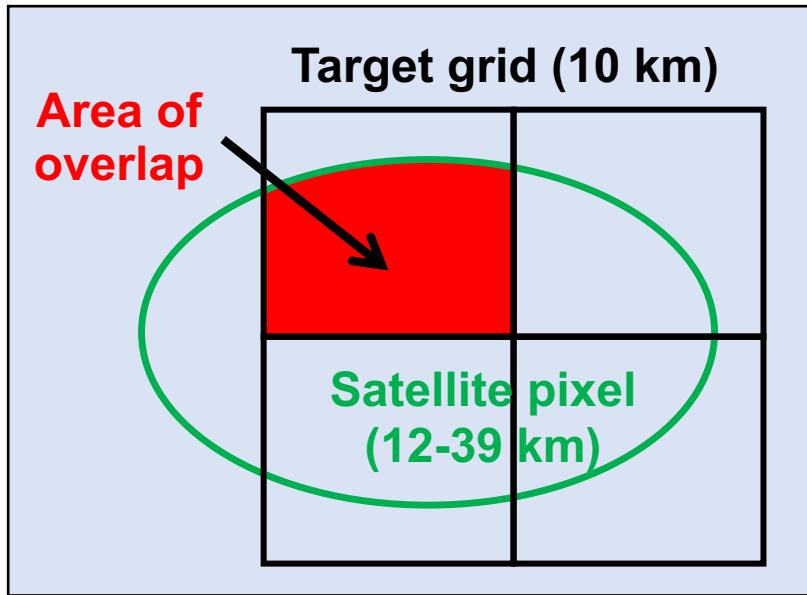
Launch date:  
October 2006



# Ammonia emissions in the UK: the top-down perspective

Enhance the spatial resolution relative to the native resolution of the instrument by oversampling

## Oversampling Technique

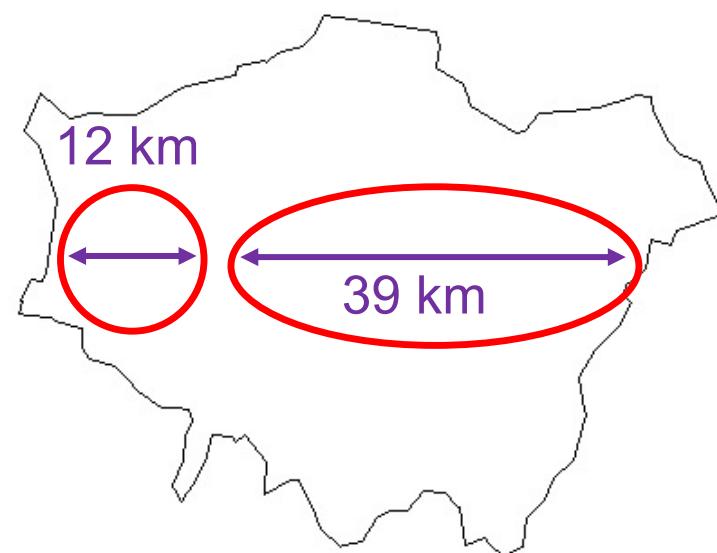


Weights each IASI NH<sub>3</sub> pixel by area of overlap and the reported uncertainty

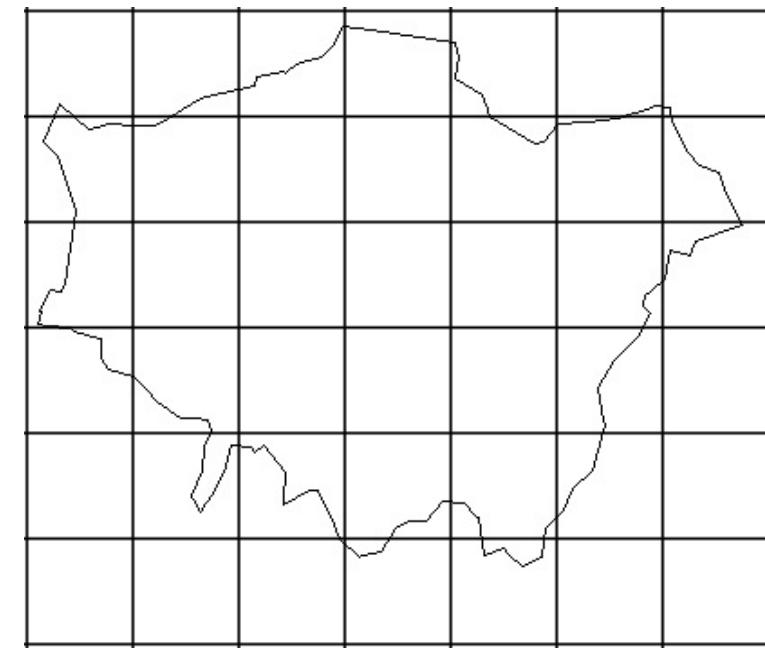
Oversampling code: L. Zhu,  
SUSTech (Zhu et al., 2017)

## Oversampling technique over London

IASI ground pixel



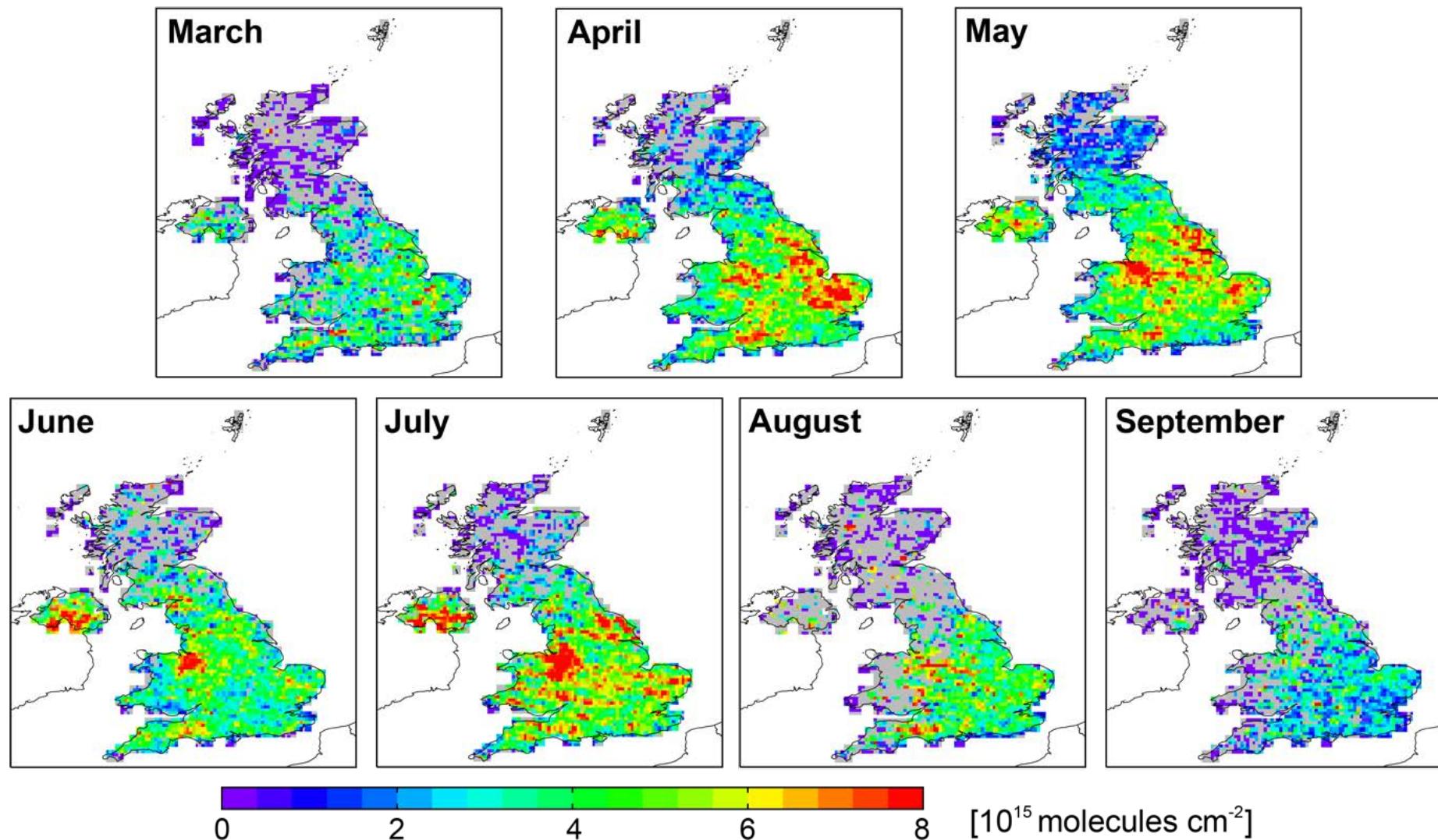
0.1° x 0.1° (~10 km) grid



Lose time (temporal) resolution; gain spatial resolution

# Multiyear (2008-2018) monthly mean IASI NH<sub>3</sub> at 0.1° x 0.1°

IASI NH<sub>3</sub> retrieved using spectral enhancement due to NH<sub>3</sub> and machine learning (neural network)

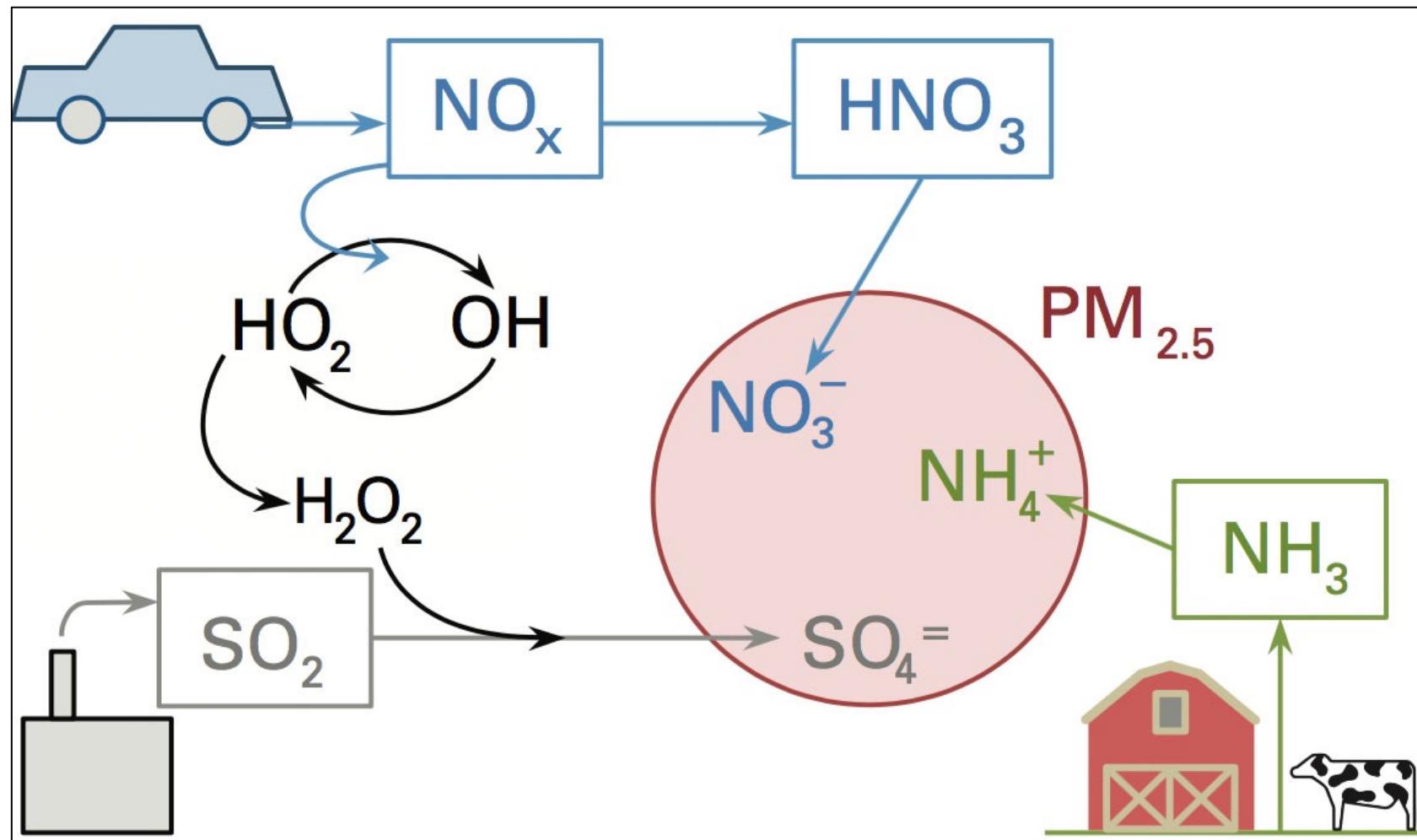


Data for Oct-Feb and over Scotland have low signal and large relative retrieval error (> 100%)

**IASI data providers:** M. Van Damme, L. Clarisse, P.-F. Coheur, ULB, Belgium

# Ammonia abundance depends on numerous factors

Ammonia partitions to aerosols to form PM<sub>2.5</sub>



[<http://climate-science.mit.edu/>]

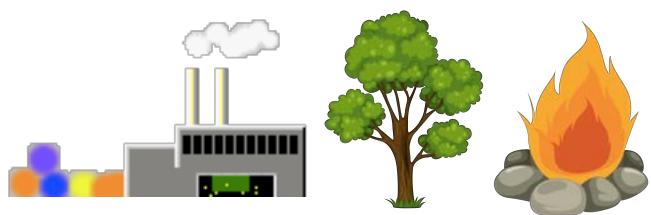
Partitioning of ammonia (NH<sub>3</sub>) to pre-existing aerosols depends on abundance of NO<sub>x</sub> and SO<sub>2</sub>

# Surface SO<sub>2</sub> concentrations calculated with GEOS-Chem



3D Atmospheric Chemistry Transport Model

Emissions  
(natural/human)

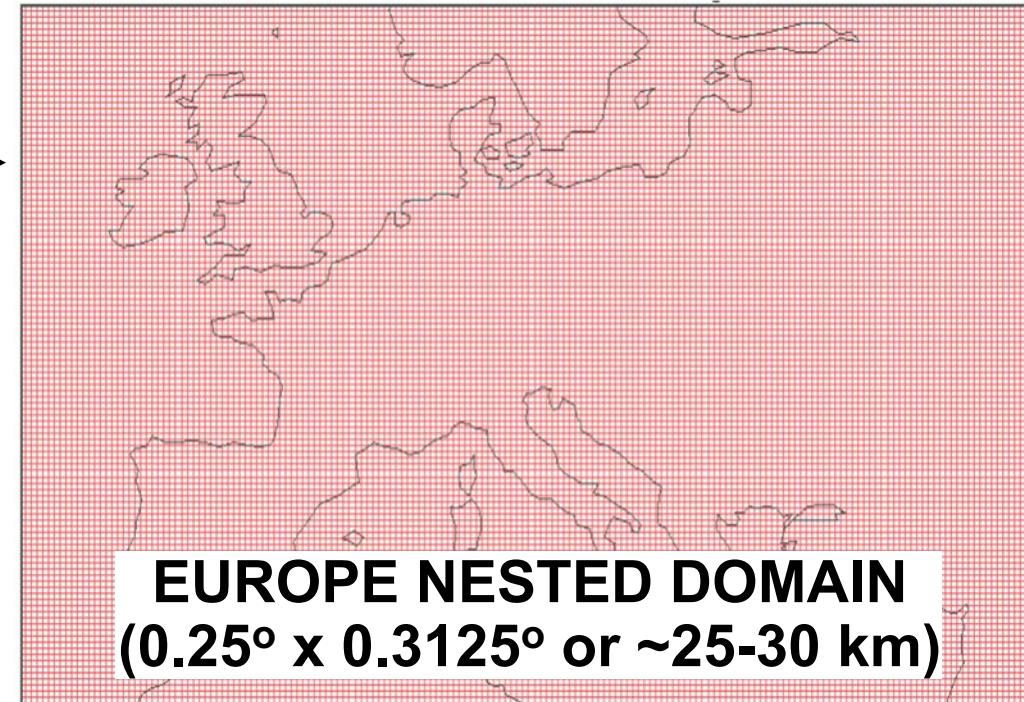


UK NAEI emissions  
(with temporal information)

Offline assimilated  
meteorology

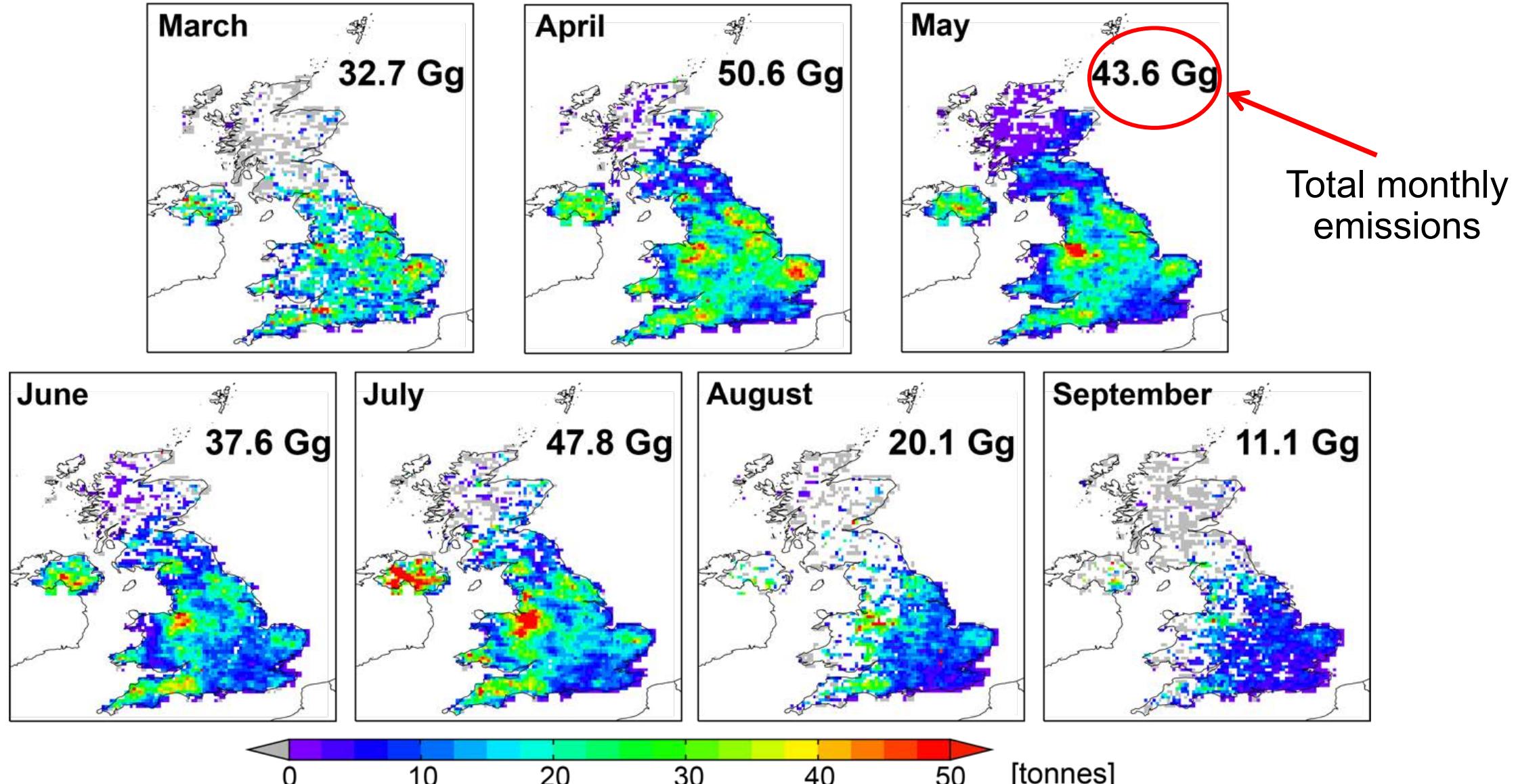


NASA GEOS-FP for 2016



Gas phase and heterogeneous chemistry  
Transport  
Dry/wet deposition

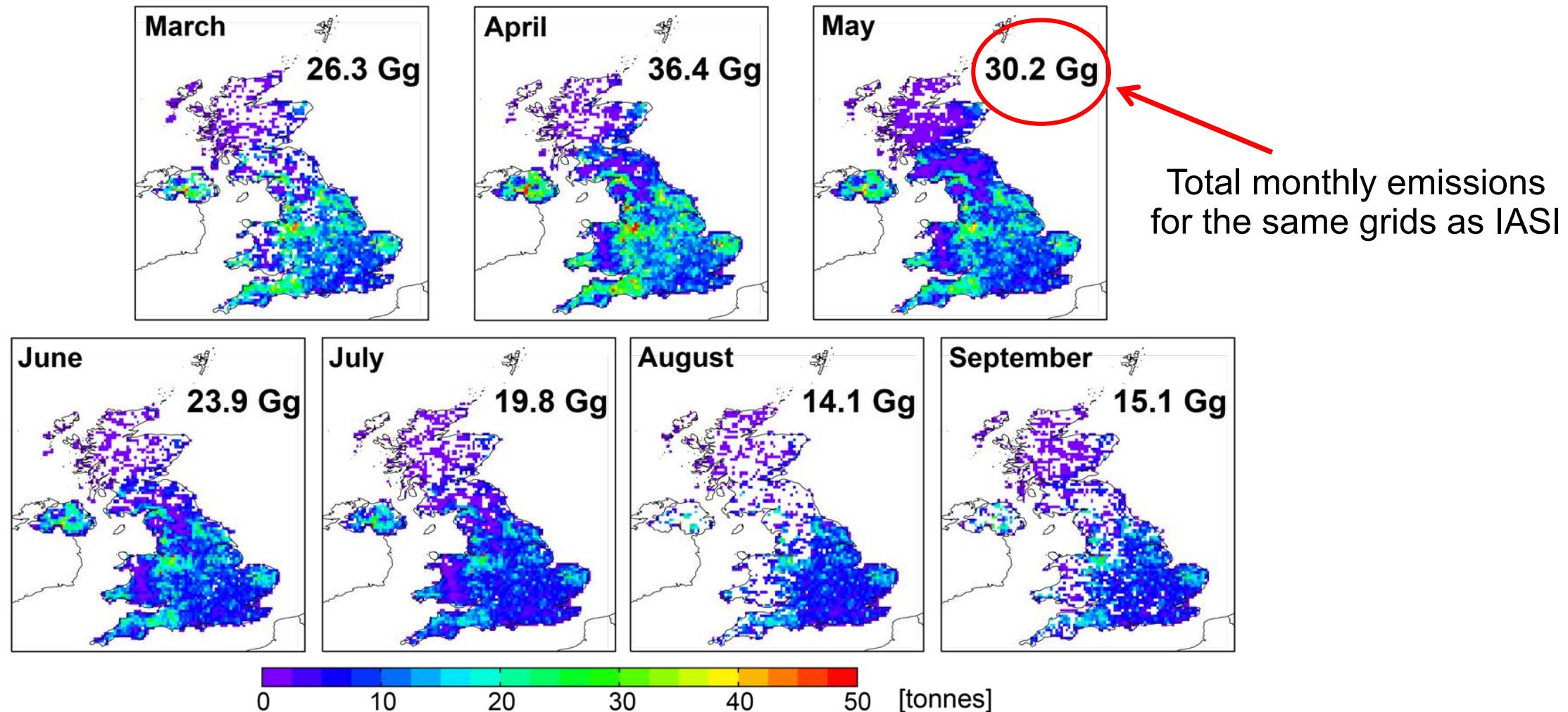
# IASI-derived $\text{NH}_3$ emissions at $0.1^\circ \times 0.1^\circ$



Sum of IASI-derived emissions for retained grids: **243.5 Gg**

# Monthly emissions from NAEI and GEOS-Chem at $0.1^\circ \times 0.1^\circ$

Obtained by multiplying NAEI annual emissions by GEOS-Chem emissions seasonality



Sum of NAEI emissions: **165.8 Gg** (56% of annual total, 32% less than IASI)

Suggests annual total IASI emissions of **435 Gg** (> NAEI annual total of 298 Gg ceiling)

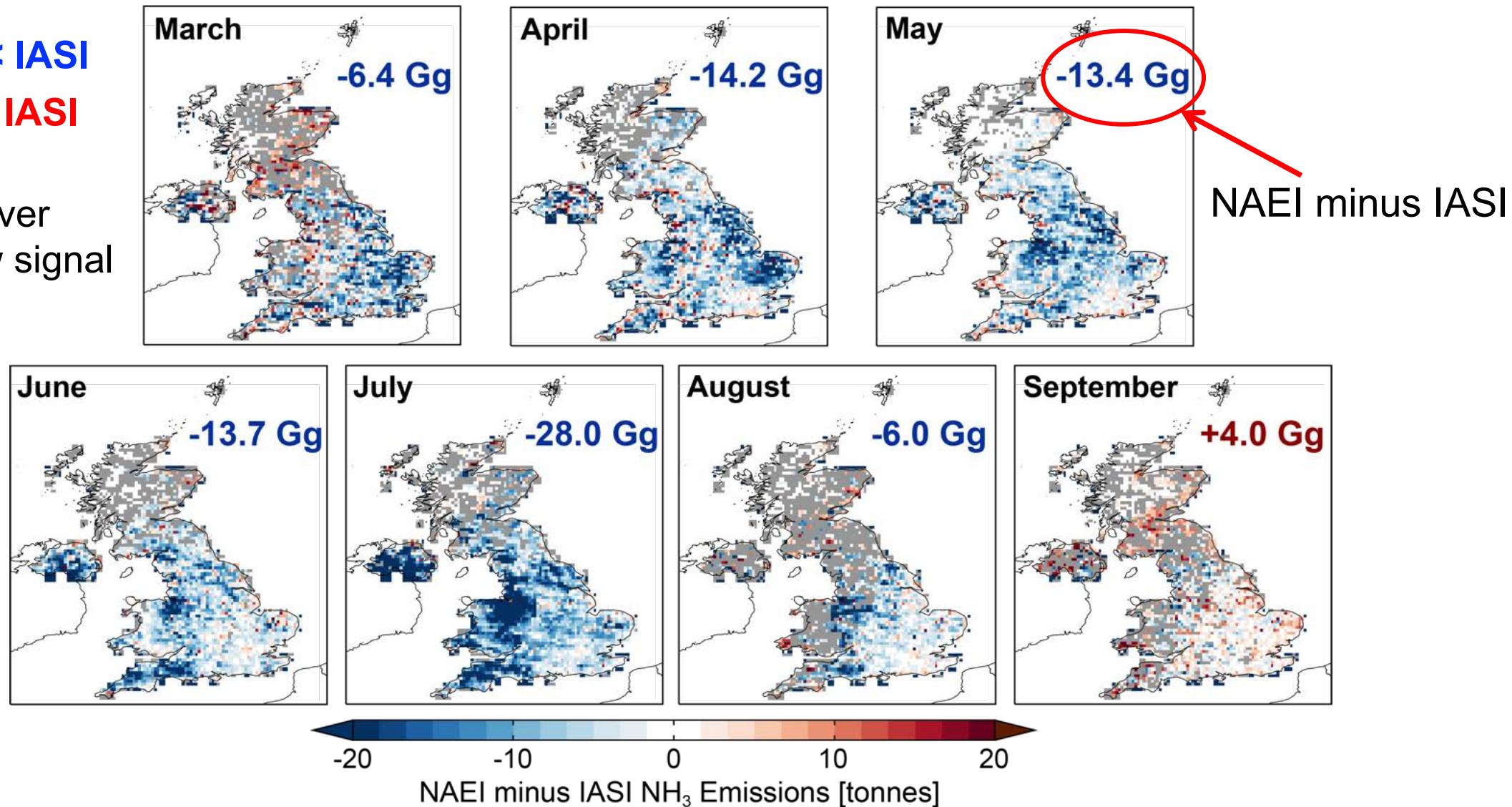
# Differences in spatial distribution of IASI and NAEI

IASI-derived emissions minus NAEI-GEOS-Chem emissions

Blue: NAEI < IASI

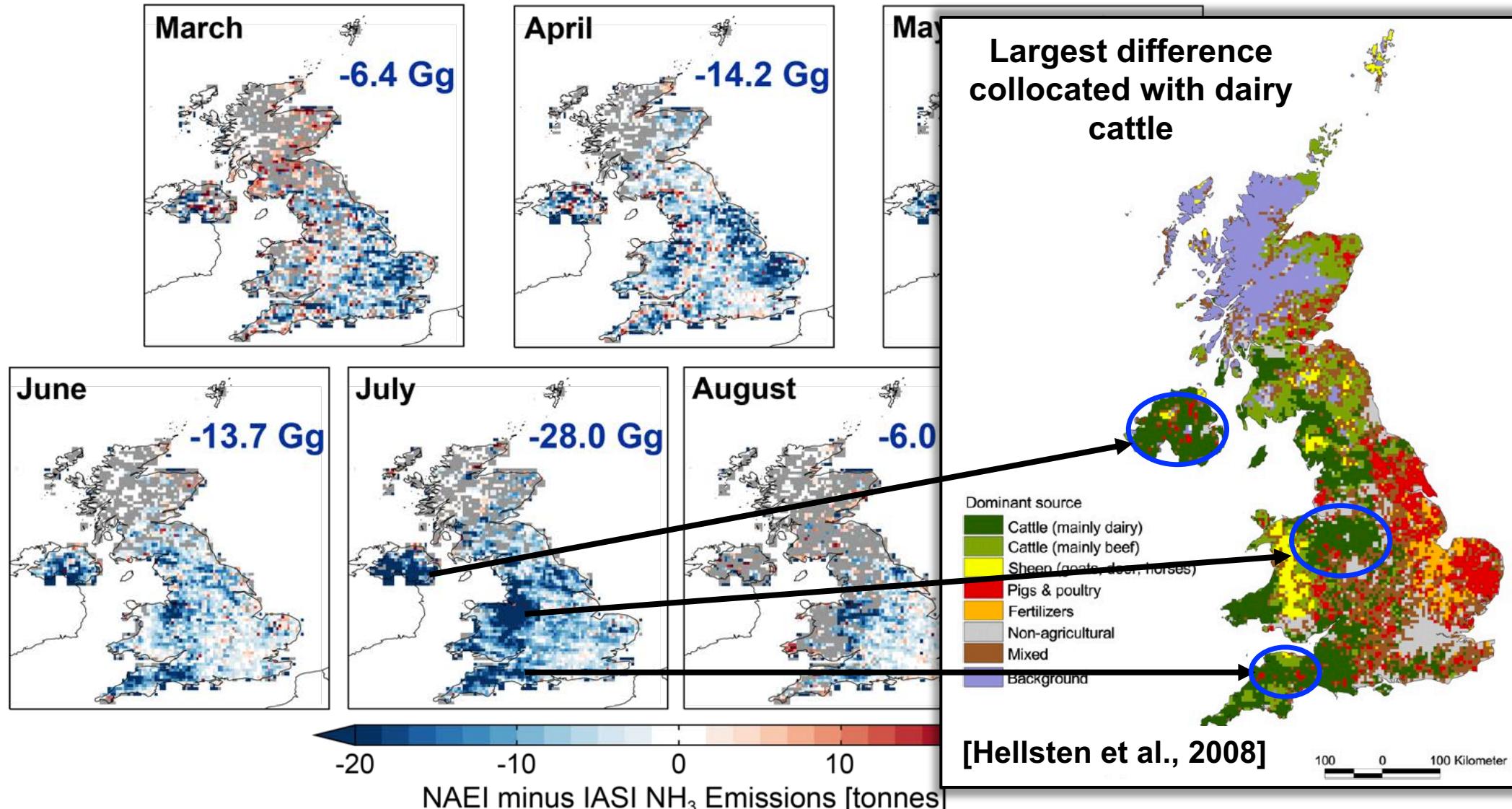
Red: NAEI > IASI

Red mostly over  
grids with low signal



# Differences in spatial distribution of IASI and NAEI

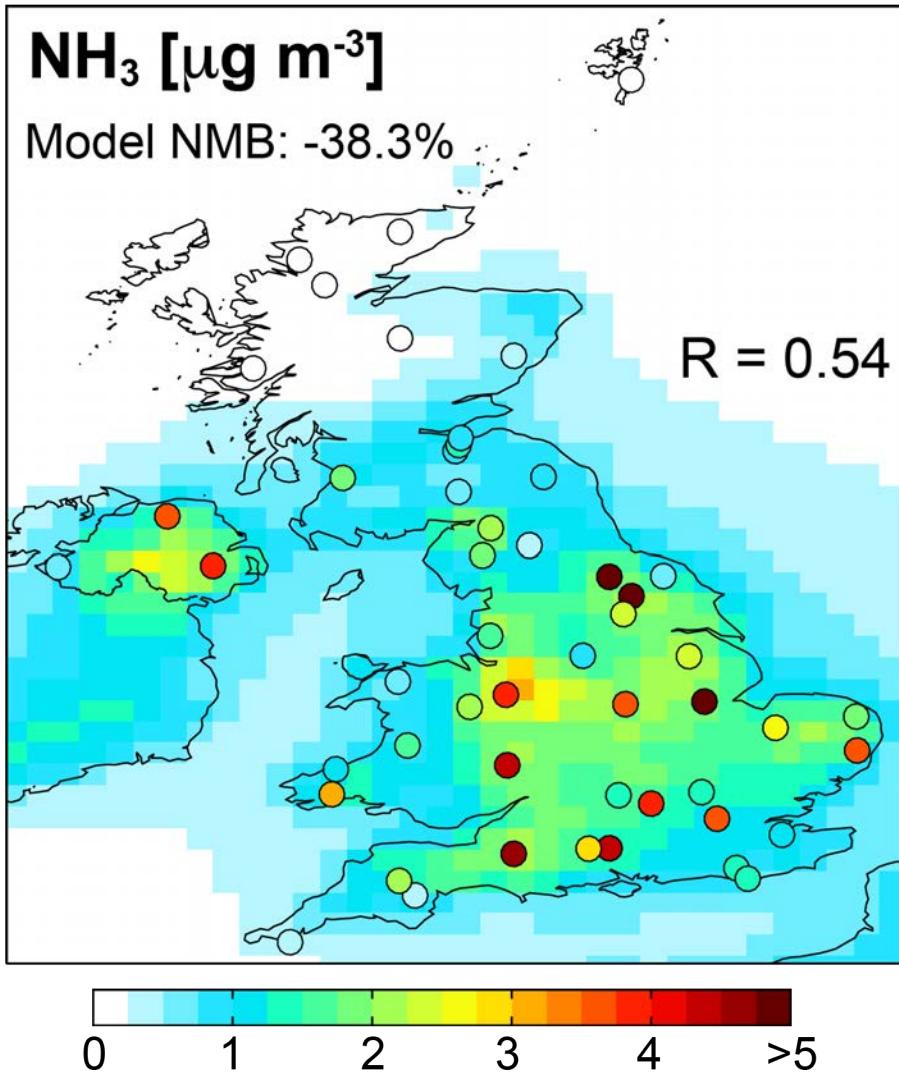
IASI-derived emissions minus NAEI-GEOS-Chem emissions



Largest discrepancy over locations dominated by **dairy farms**

# Surface network observations corroborate top-down results

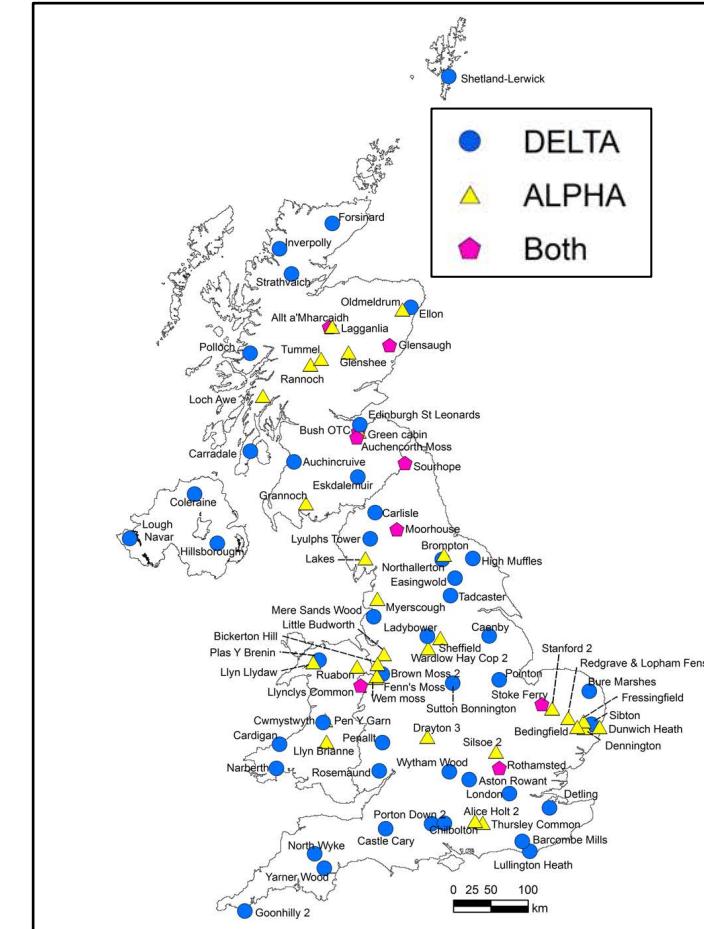
Network (points) and model (background)  
surface NH<sub>3</sub> in Mar-Sep



Points are for DELTA  
instruments (blue circles)

DELTA instruments support  
model underestimate  
(NMB = -38%)

So do passive low-cost  
ALPHA instruments (yellow  
triangles)  
(NMB = -41.5%)



GEOS-Chem underestimate in surface NH<sub>3</sub> driven with the NAEI corroborates results from IASI

# Concluding Remarks for top-down NH<sub>3</sub> emissions

- IASI-derived emissions are 34-46% more than those from the UK National Atmospheric Emission Inventory (NAEI).
- IASI-derived emissions random error is 4-35%.
- Largest discrepancy between top-down and bottom-up inventory is in July in locations dominated by dairy farms, but IASI data may be susceptible to a clear-sky bias in July.
- NAEI-IASI difference is similar to reported errors for the NAEI (31%), but problematic for policy development and estimating environmental impact.

