Space-based constraints on reactive nitrogen:

From emissions of NH_3 at the surface to NO_x in the upper troposphere

Eloïse Marais

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e.marais@ucl.ac.uk

First Estimate of Upper Tropospheric NO_2 from TROPOMI

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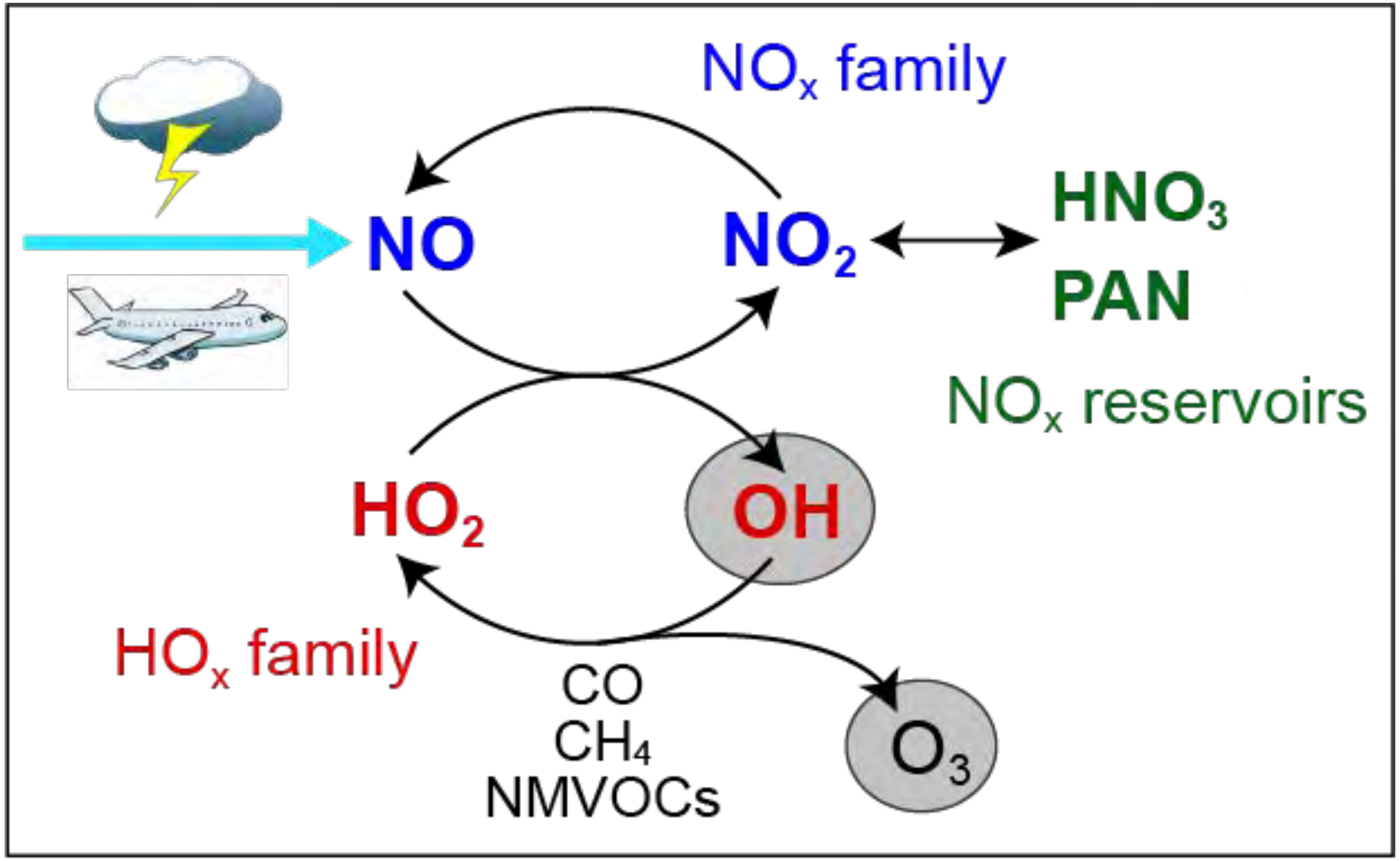
Contributors: J. F. Roberts, R. G. Ryan, H. Eskes, K. F. Boersma, S. Choi, J. Joiner, N. Abuhassan, A. Redondas, M. Grutter, A. Cede, L. Gomez, M. Navarro-Comas

Nitrogen oxides (NO_x) in the Upper Troposphere (8-12 km)



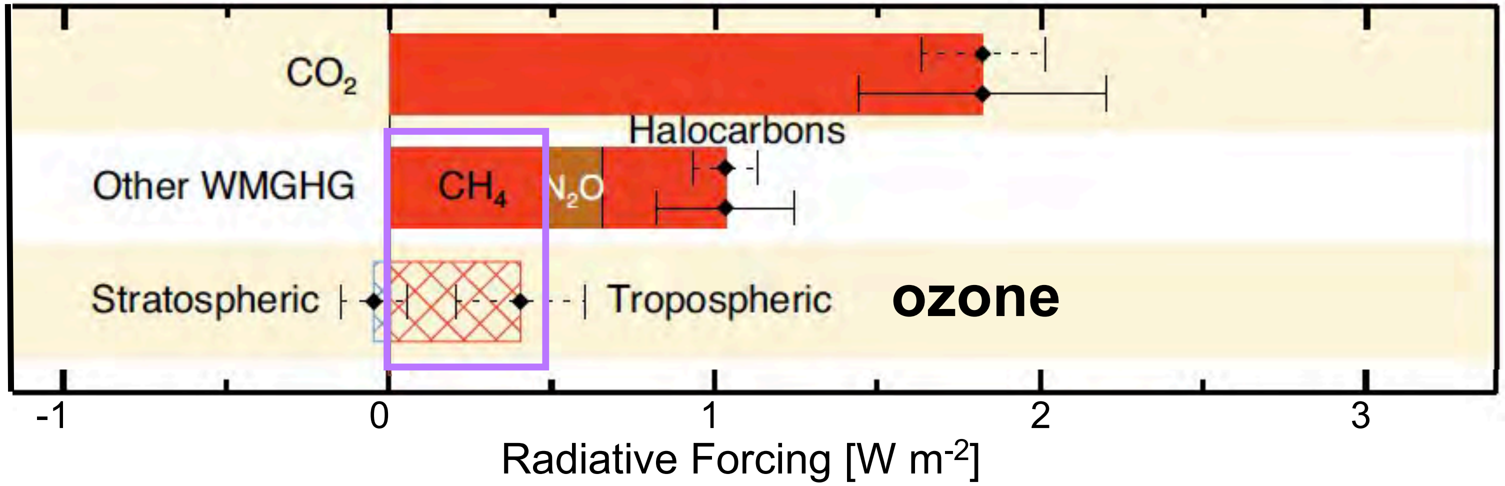
Influence atmospheric oxidants (OH, O_3) and climate (O_3 formation, methane persistence)

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

Why uncertainties in the upper troposphere matter

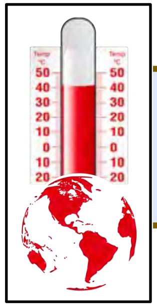
Warming due to tropospheric ozone is similar to that of methane (CH_4)

Tropospheric ozone and methane have near-equal climate impacts

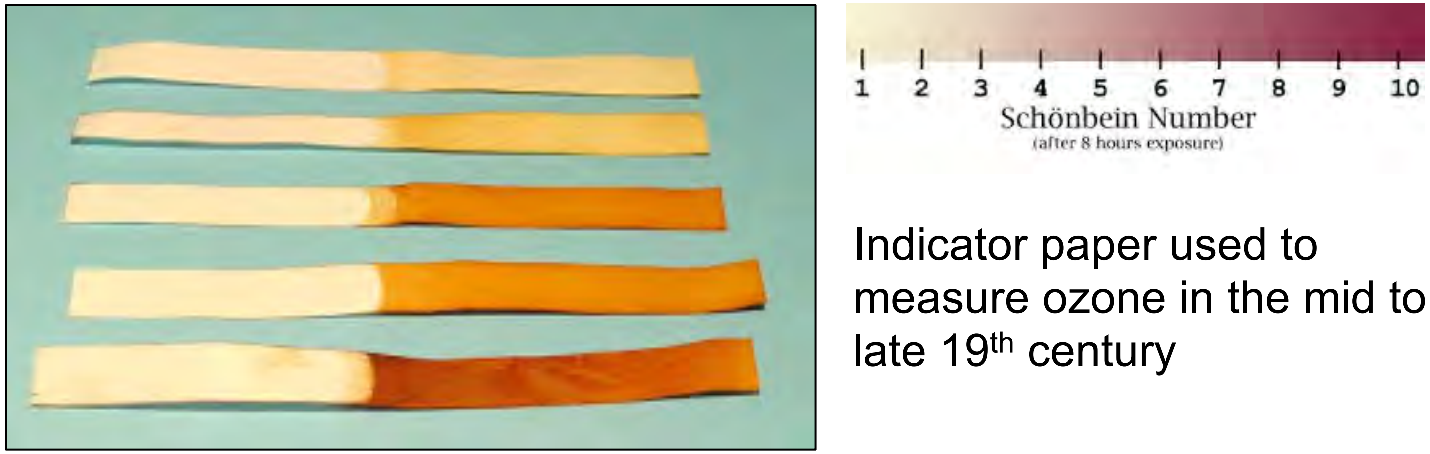


[IPCC AR5, 2013]

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



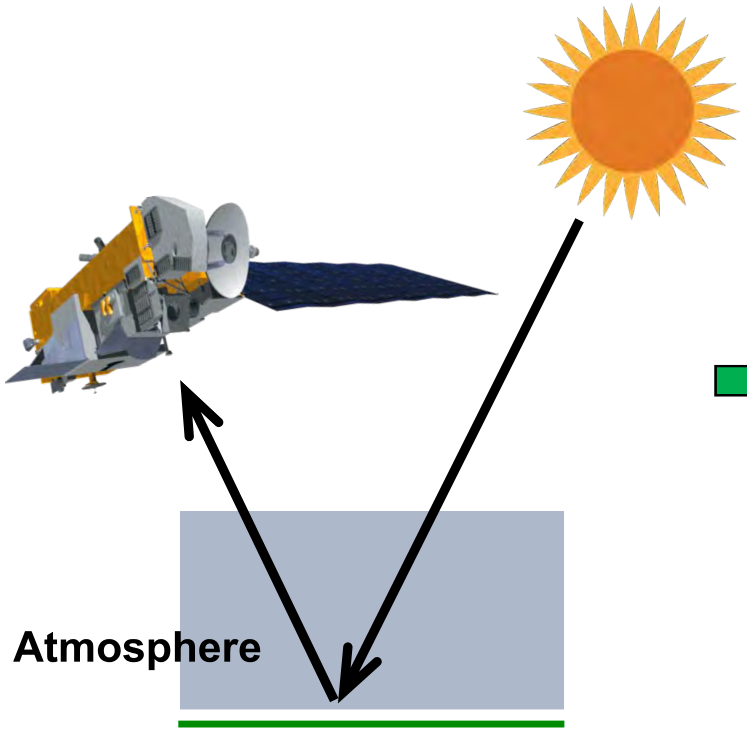
Errors in tropospheric ozone radiative forcing



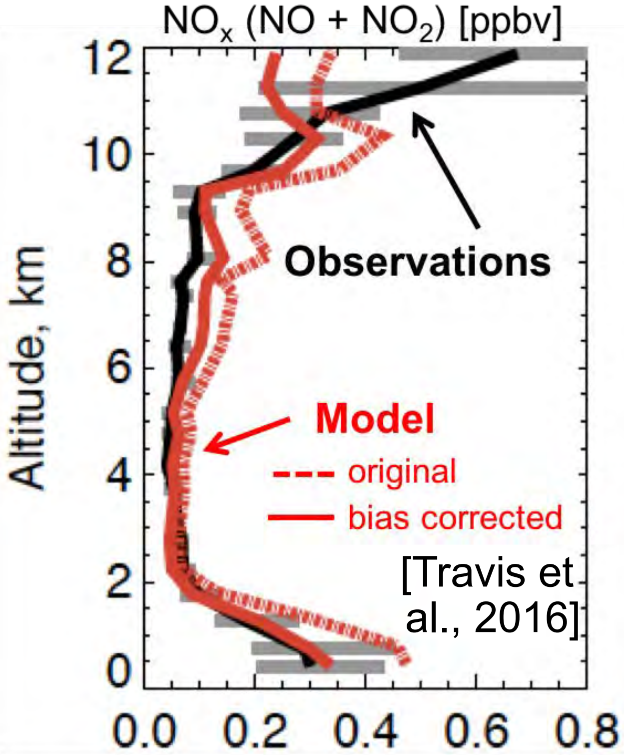
Indicator paper used to measure ozone in the mid to late 19th century

Why uncertainties in the upper troposphere matter

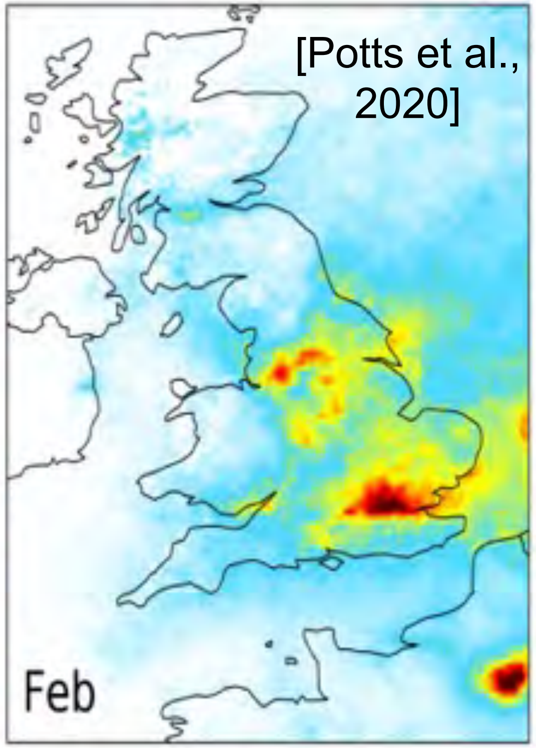
We rely on models to retrieve atmospheric composition from satellite instruments



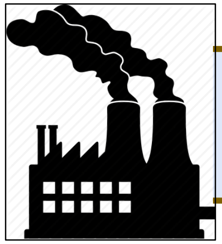
NO_2 along the viewing path



NO_2 vertical profile (model)



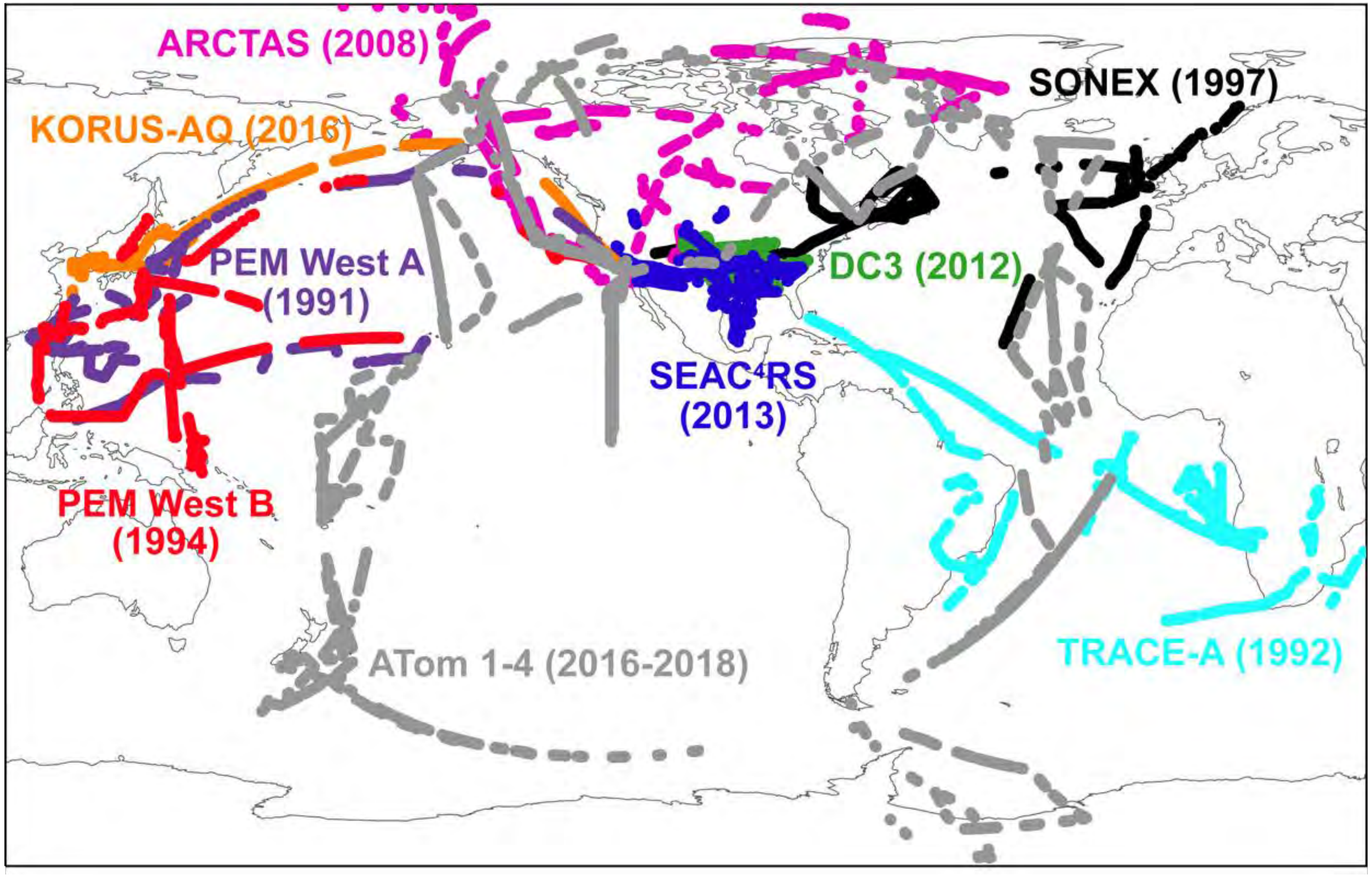
Vertical column densities



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

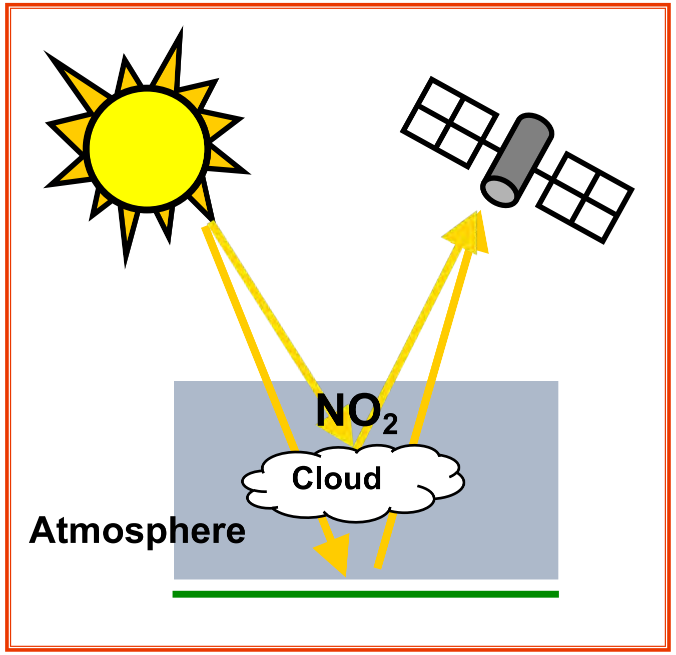
All use instruments that are susceptible to large biases in the upper troposphere

Satellite products obtained using the cloud-slicing technique

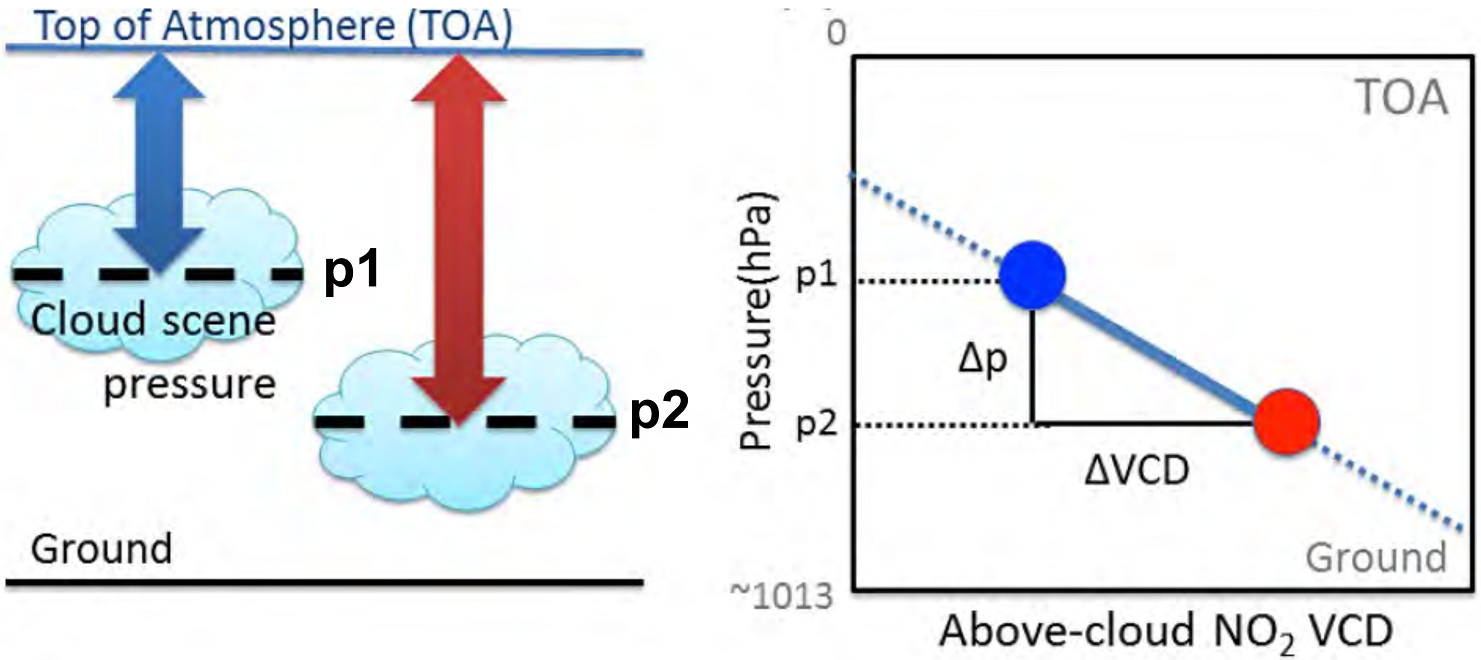
First applied by Ziemke et al. [2001] to TOMS ozone

Retrieve partial NO_2 columns over cloudy scenes at different heights

APPROACH



Use cloud height variability to derive partial columns



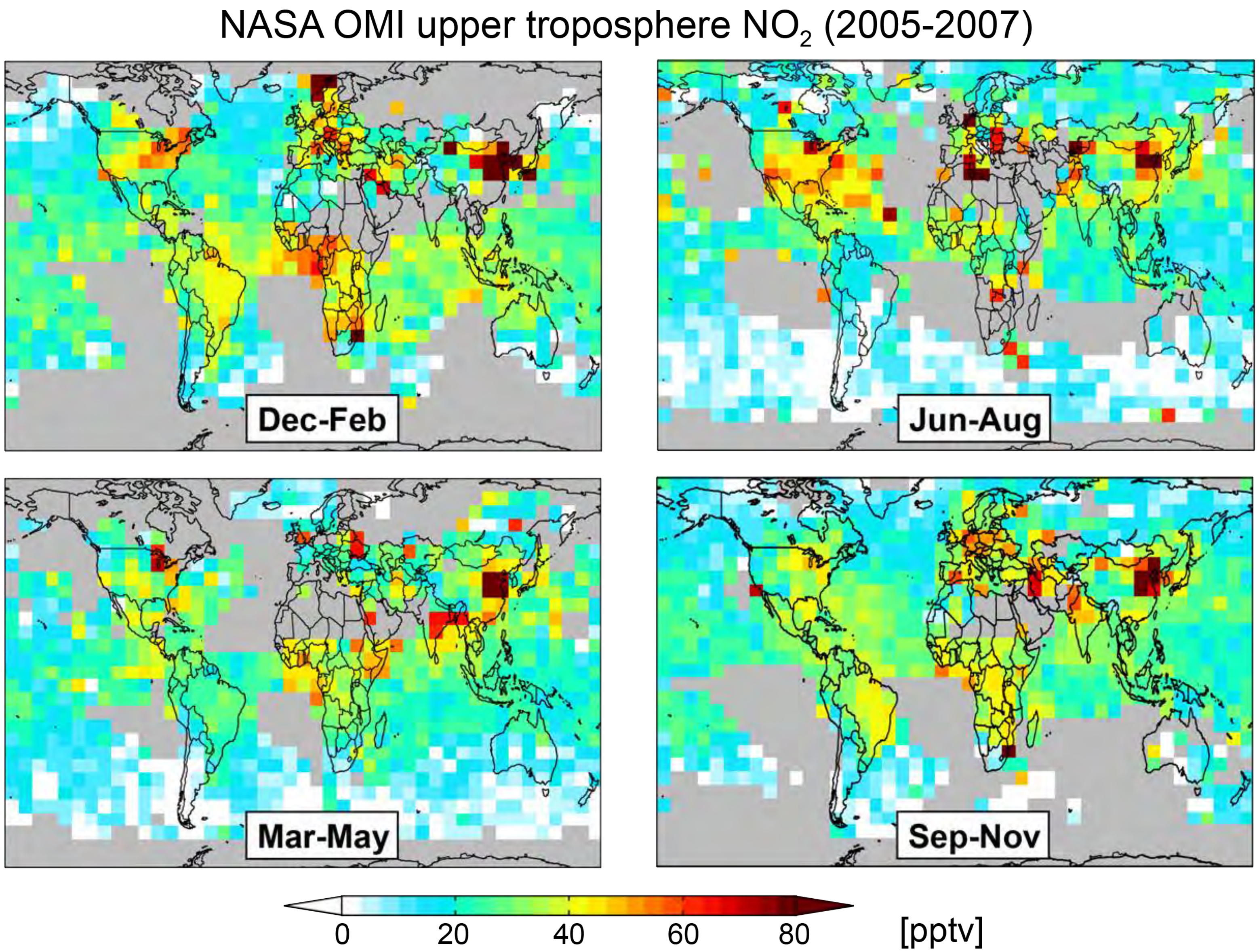
[adapted from Choi et al., 2014]

NO_2 volume mixing ratio (VMR) between clouds at p1 and p2

$$\mathrm{NO_2\ VMR} = \frac{\Delta \mathrm{VCD}}{\Delta \mathrm{p}} \times \frac{k_B g}{R_{\mathrm{air}}}$$

Products of upper tropospheric NO_2 from satellite observations

Near global spatial coverage of seasonal mean UT NO_2 at $5° \times 8°$ (50 km × 80 km)

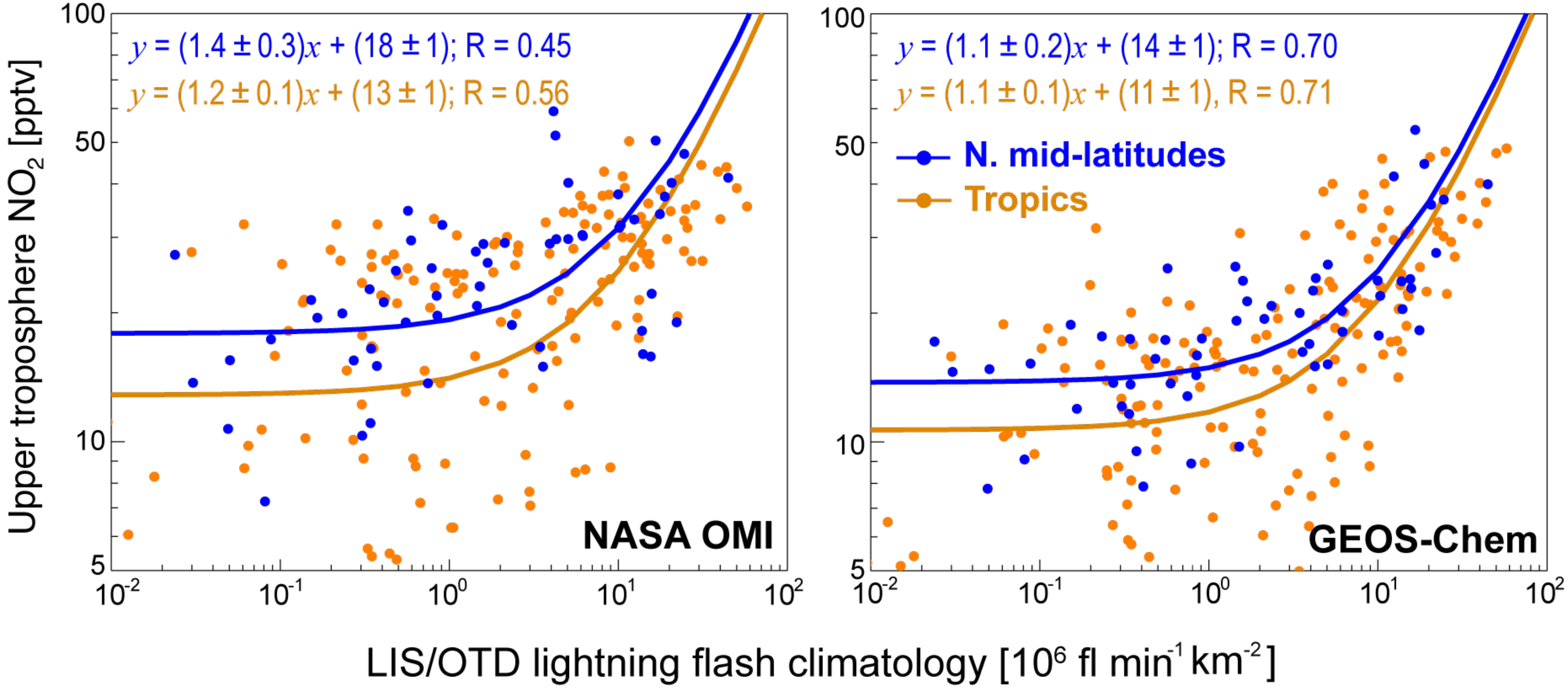


OMI:
Ozone Monitoring Instrument

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

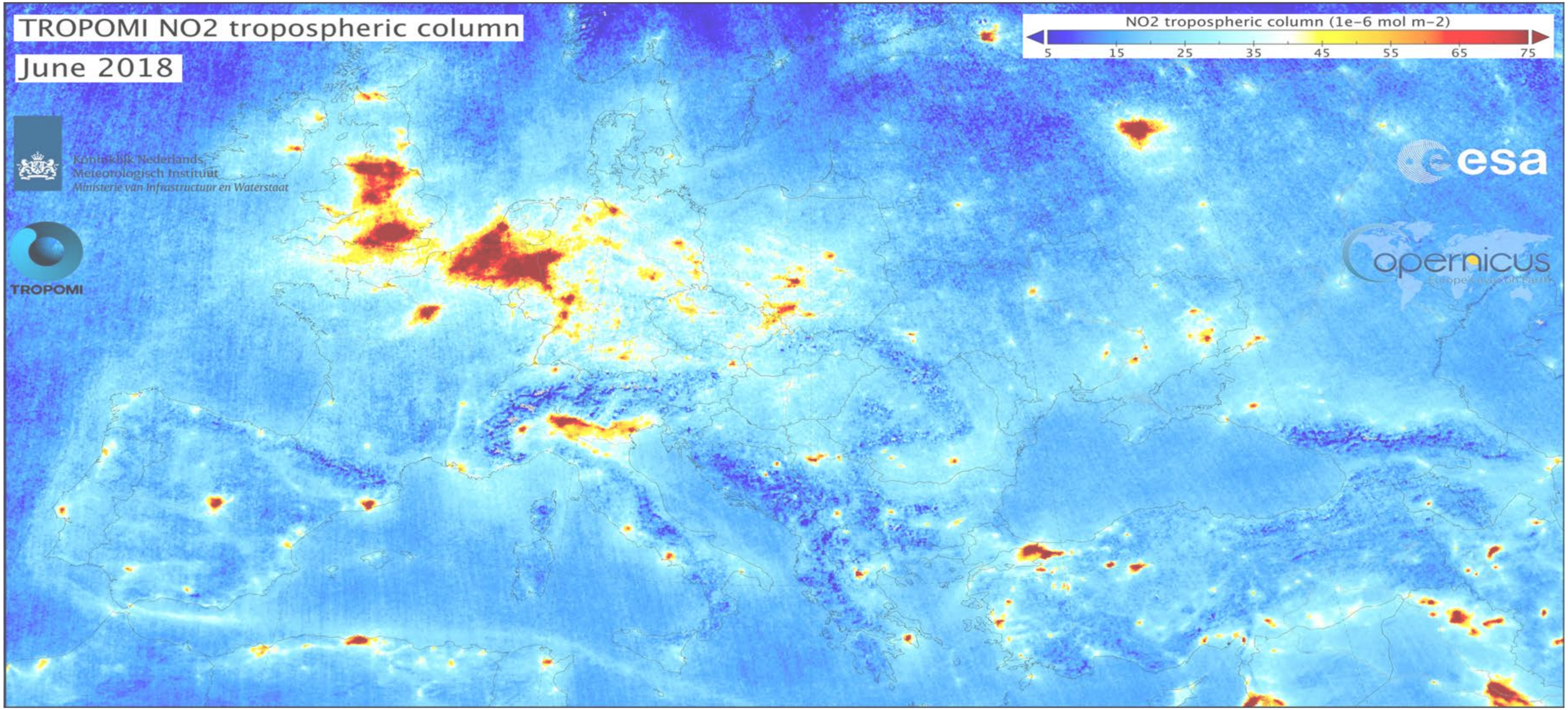
Provide improved constraints on lightning NO_x emissions

Log-log relationship between UT NO_2 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_x production rates

Can we do better with high-resolution TROPOMI measurements?

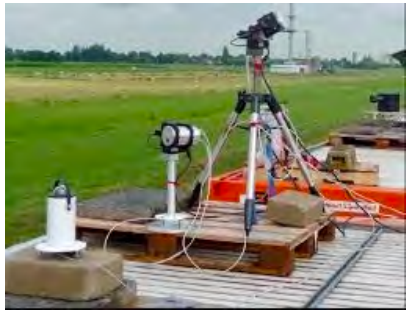


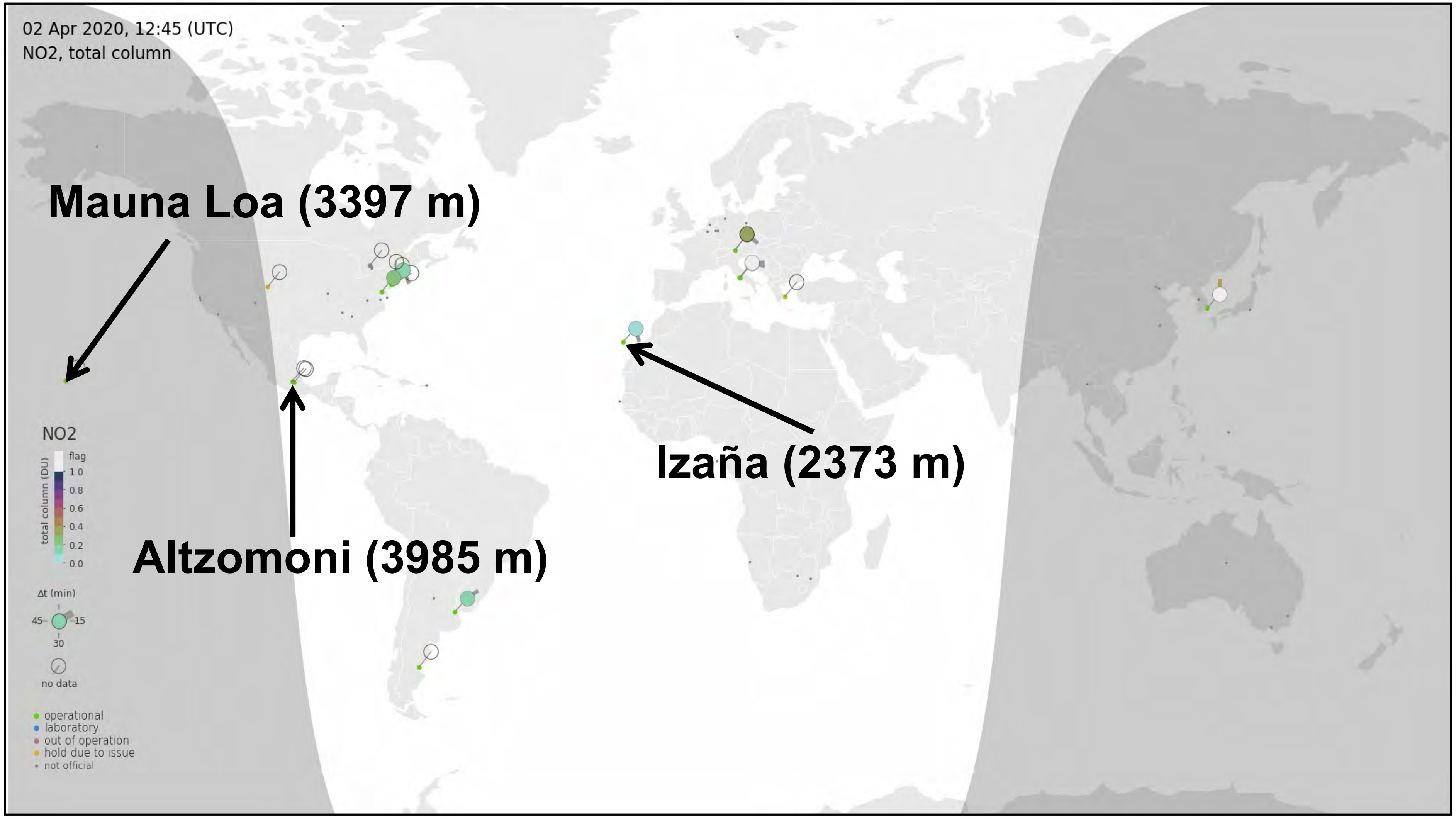
[Source: http://www.tropomi.eu/data-products/nitrogen-dioxide]

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



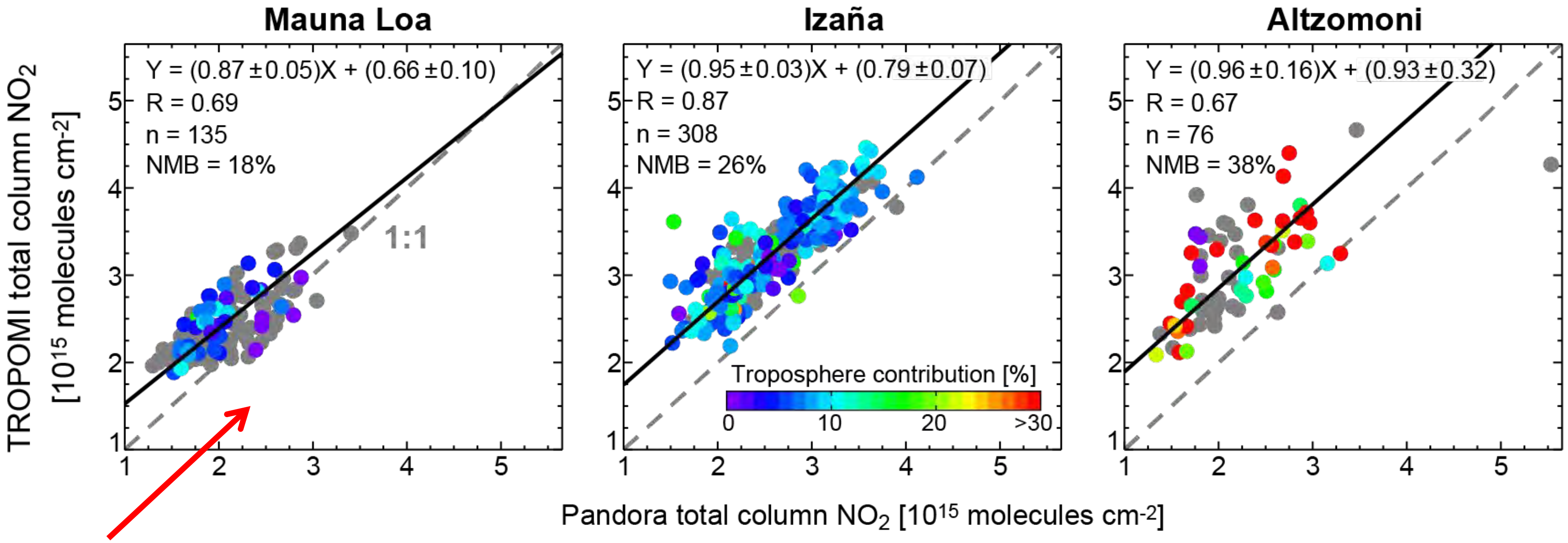


[Source: https://www.pandonia-global-network.org/]

Evaluate TROPOMI with ground-based measurements

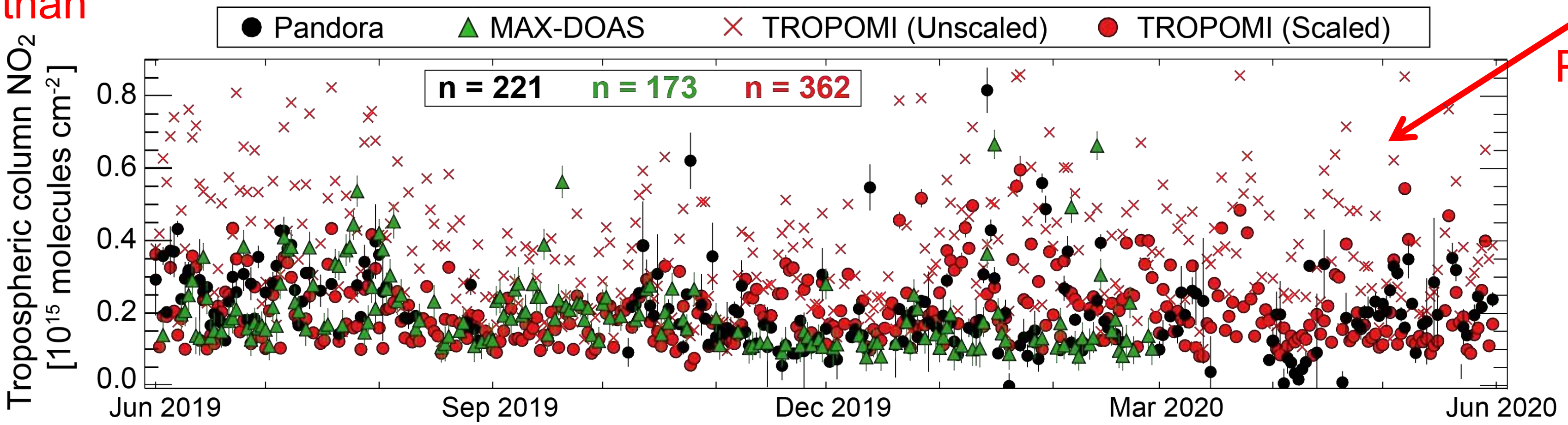
Stratosphere:

Stratosphere + free troposphere:



TROPOMI stratospheric column variance is less than Pandora

Free troposphere:



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

TROPOMI cloud-sliced upper tropospheric NO_2

Seasonal mean UT NO_2 mixing ratios from 2 distinct cloud products at 1° × 1°

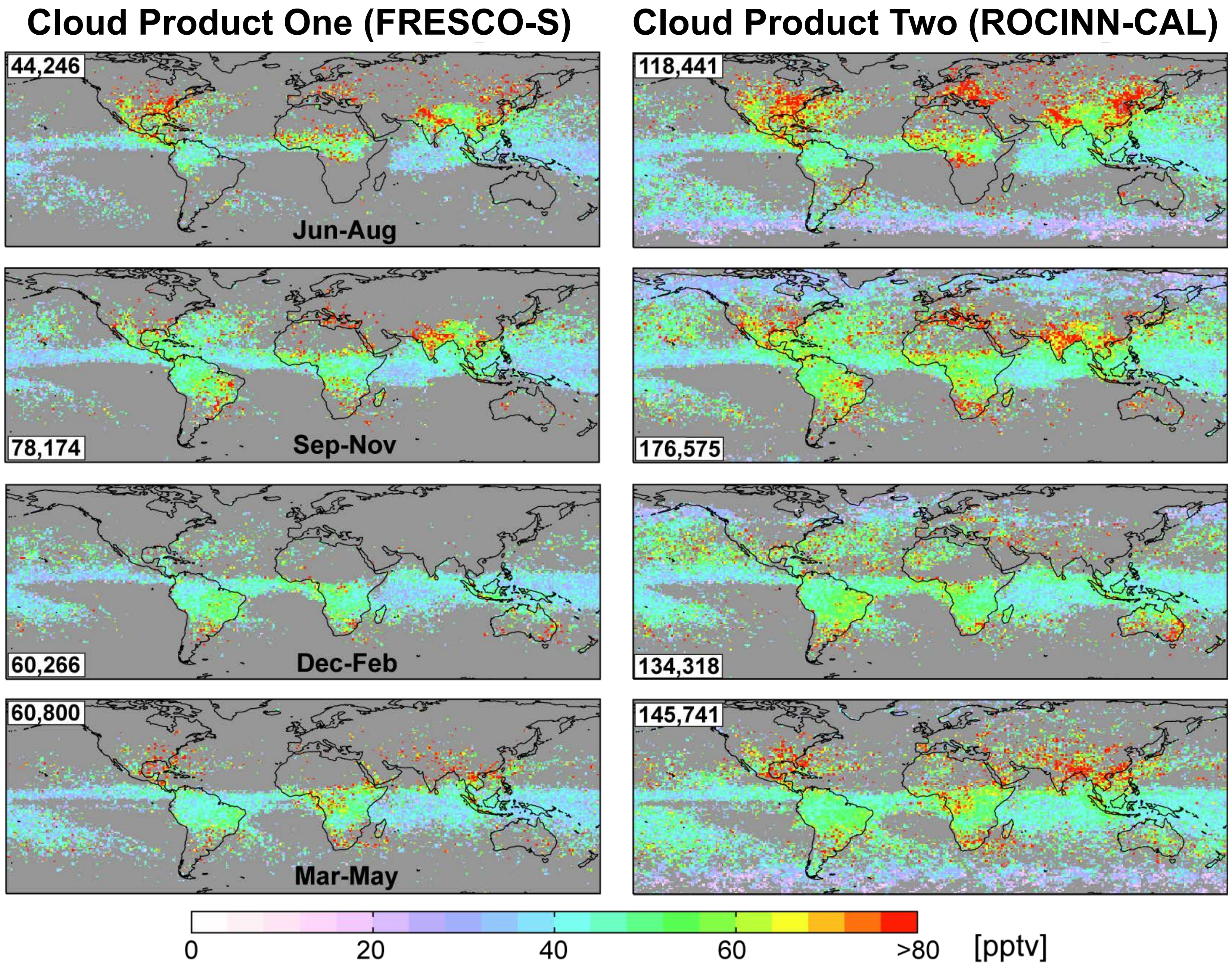
<u>Range</u>: 30-80 pptv

<u>Background</u>: ~30 pptv

Cloud products give similar UT NO_2 in the tropics

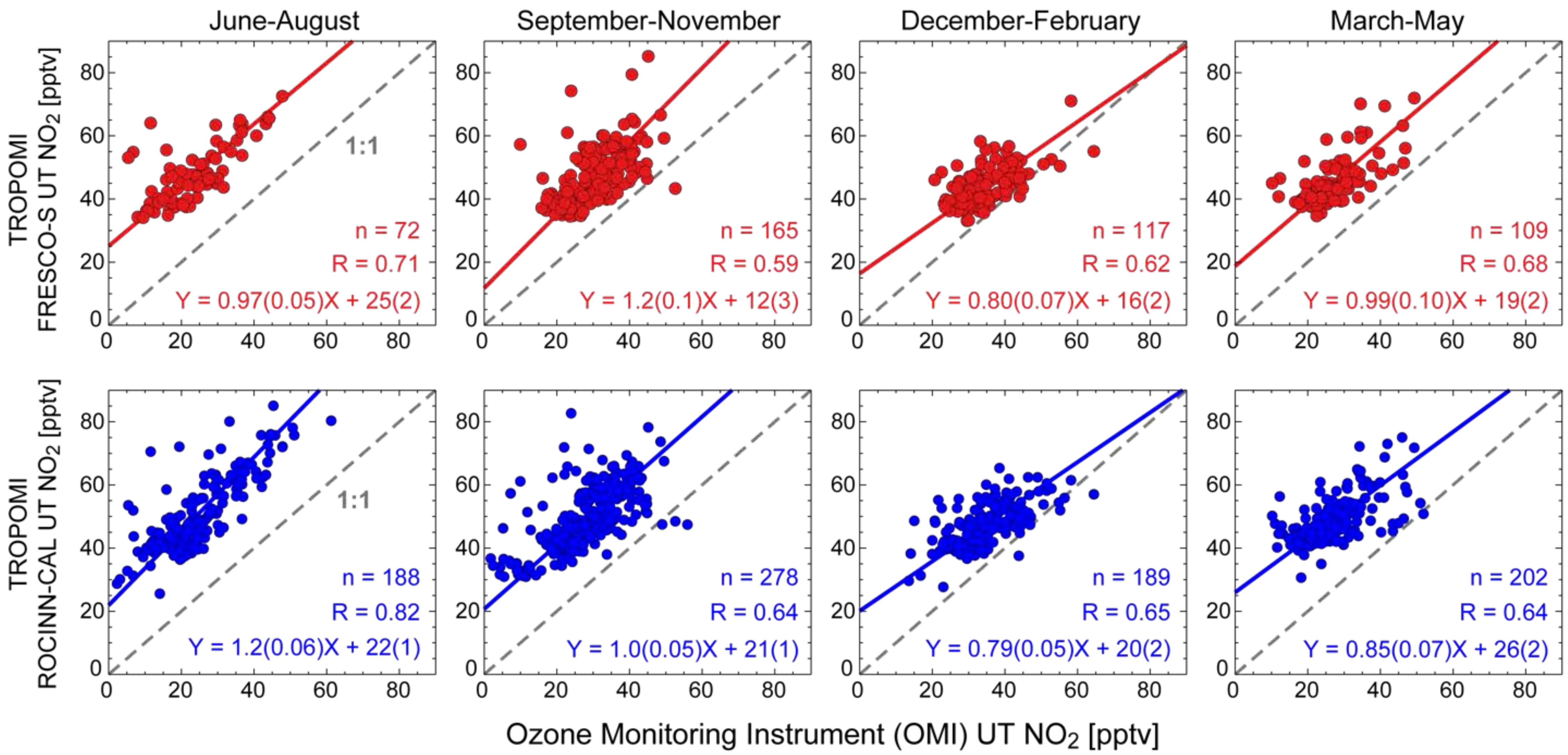
Greater coverage with ROCINN-CAL

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



Comparison to the NASA OMI product

TROPOMI UT NO_2 obtained at 1° × 1° and gridded to the NASA product resolution (8° × 5°)

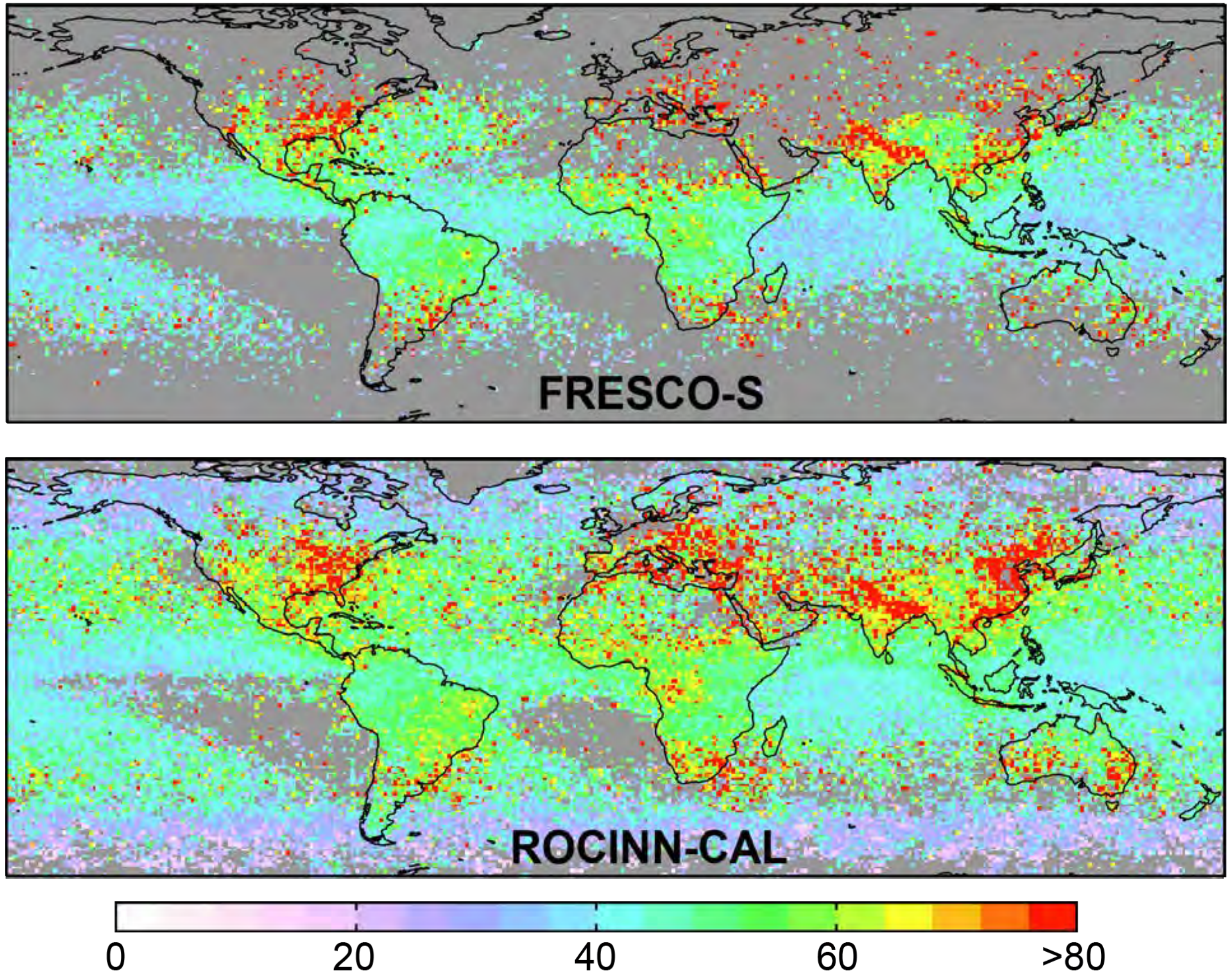


Spatial consistency. TROPOMI is 12-26 pptv more than OMI (retrieval, clouds, vertical NO_2 profile)

Concluding Remarks for TROPOMI UT NO_2

- Developed seasonal mean UT NO_2 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_2 to detect the influence of aircraft on UT NO_2 and quantify uncertainties in reactive nitrogen in the UT with GEOS-Chem

TROPOMI annual mean upper tropospheric NO_2 [pptv]



Interested in using the TROPOMI UT NO_2 product? Contact me: e.marais@ucl.ac.uk

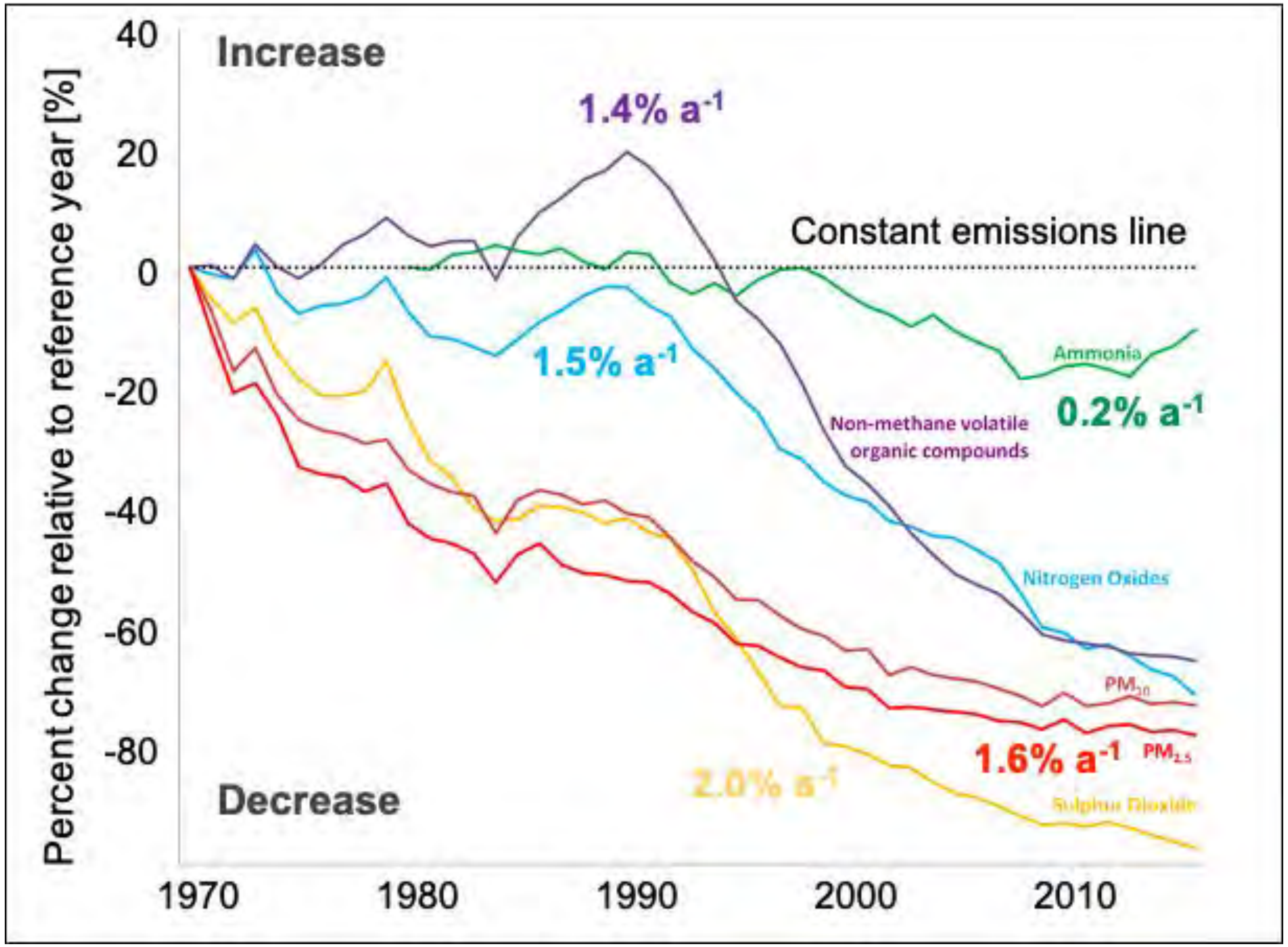
Back to Earth’s surface…

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

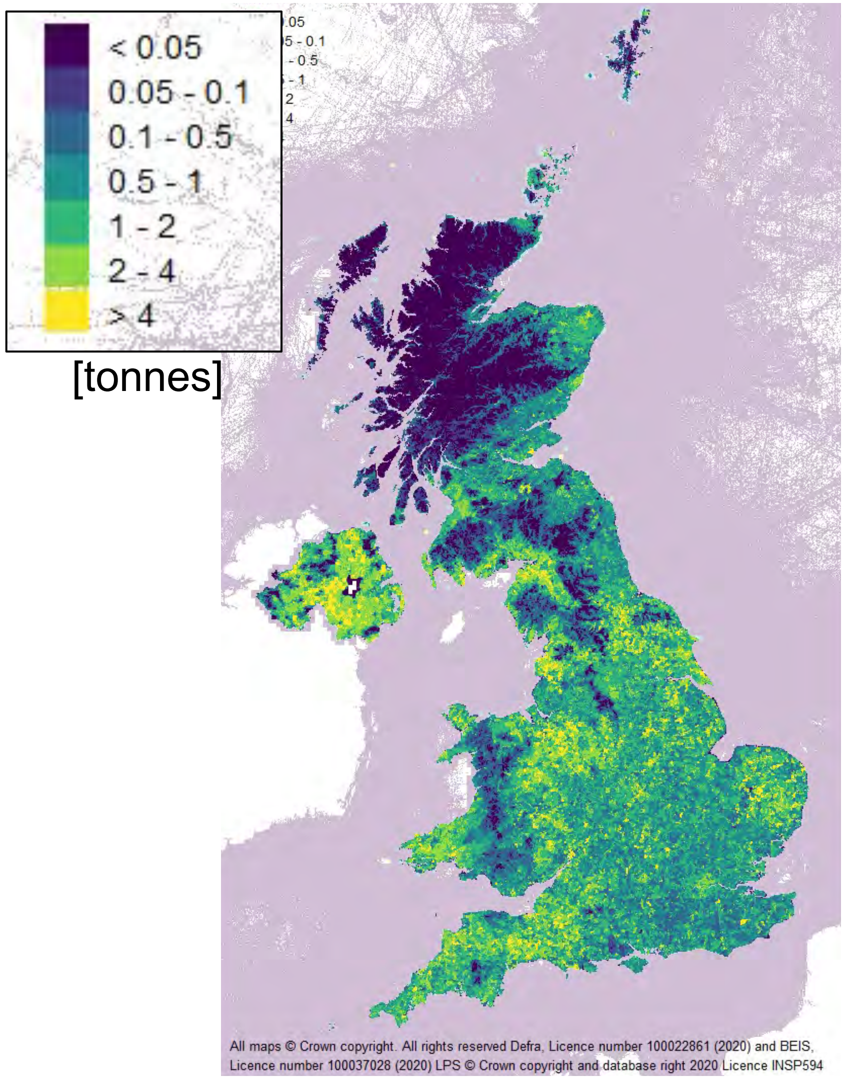


[*Adapted from* Defra, 2018]

Successful decline in all emissions, except ammonia (NH_3)

Spatial Variability in Emissions

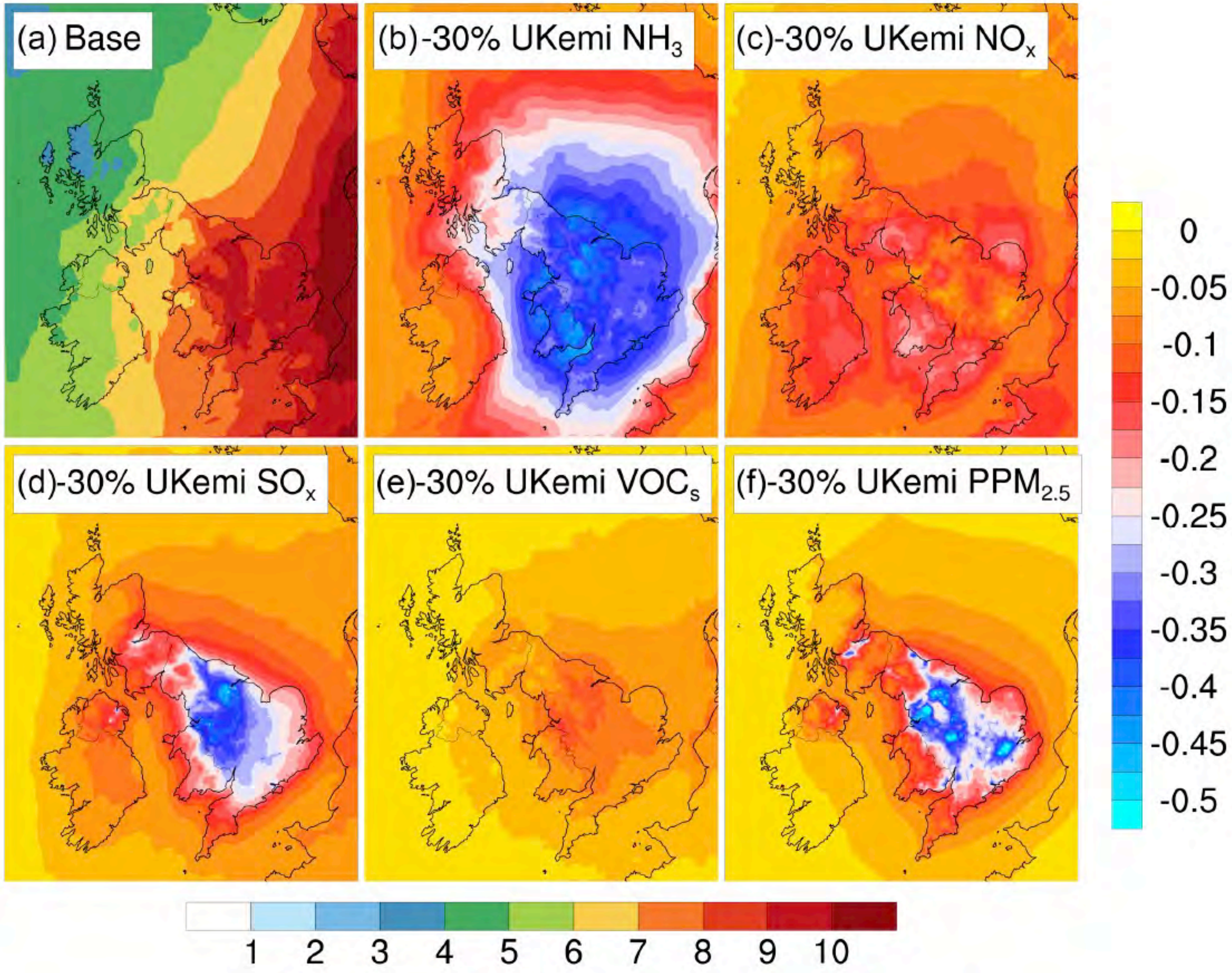
NH_3 emissions for 2018 at 1 km

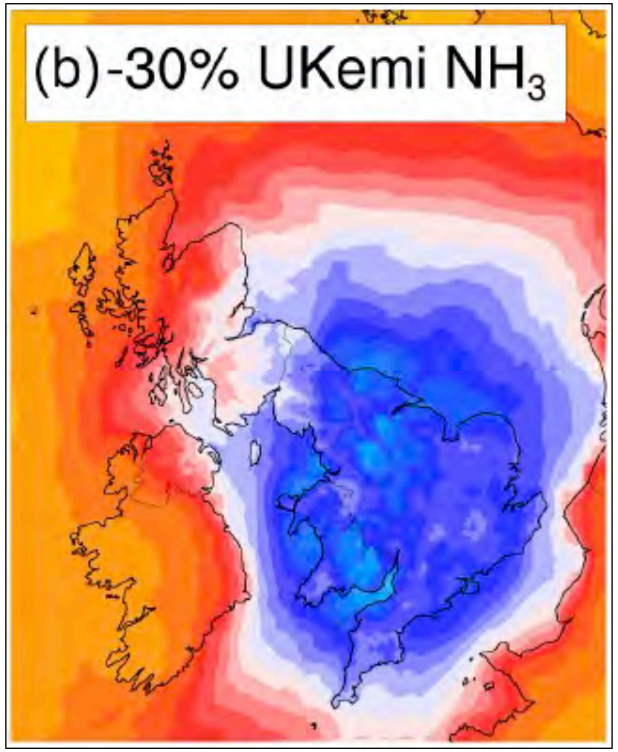


[*Adapted from* https://naei.beis.gov.uk/data/]

Ammonia impact on air pollutants hazardous to health

Effect of precursor emission reductions on 2010 $PM_{2.5}$





[Vieno et al., 2016]

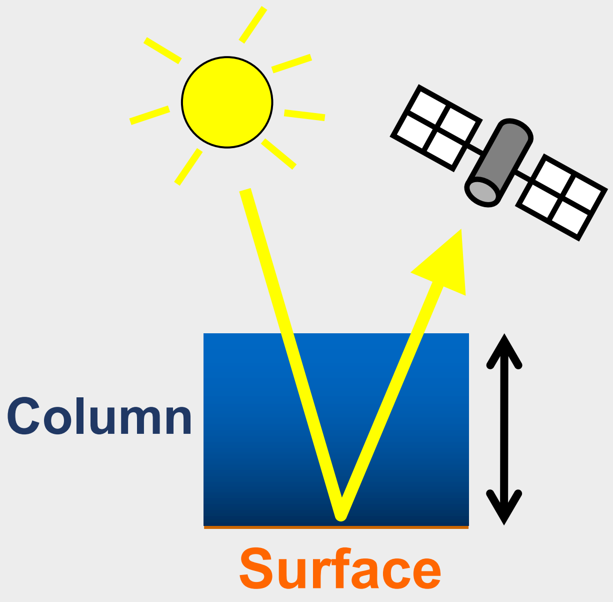
Largest and most extensive decline in $PM_{2.5}$ achieved by targeting ammonia sources

Top-down emissions estimated with satellite observations

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

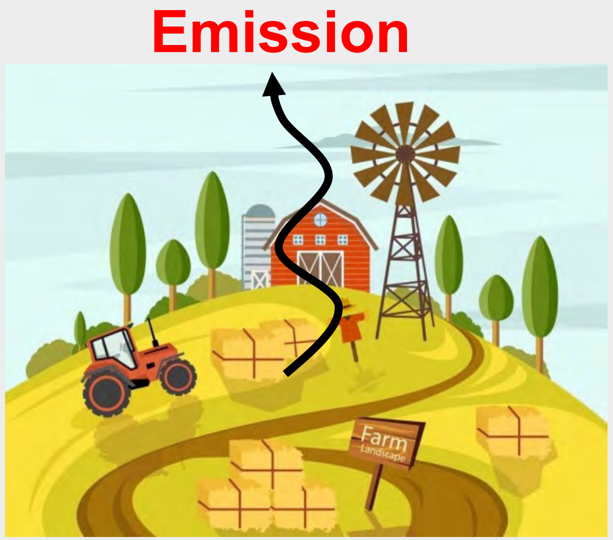
ABUNDANCES → **Conversion Factor** → **EMISSIONS**

Satellite columns (IASI)



Column-to-Emission ratio (GEOS-Chem)

Satellite-derived Surface Emissions



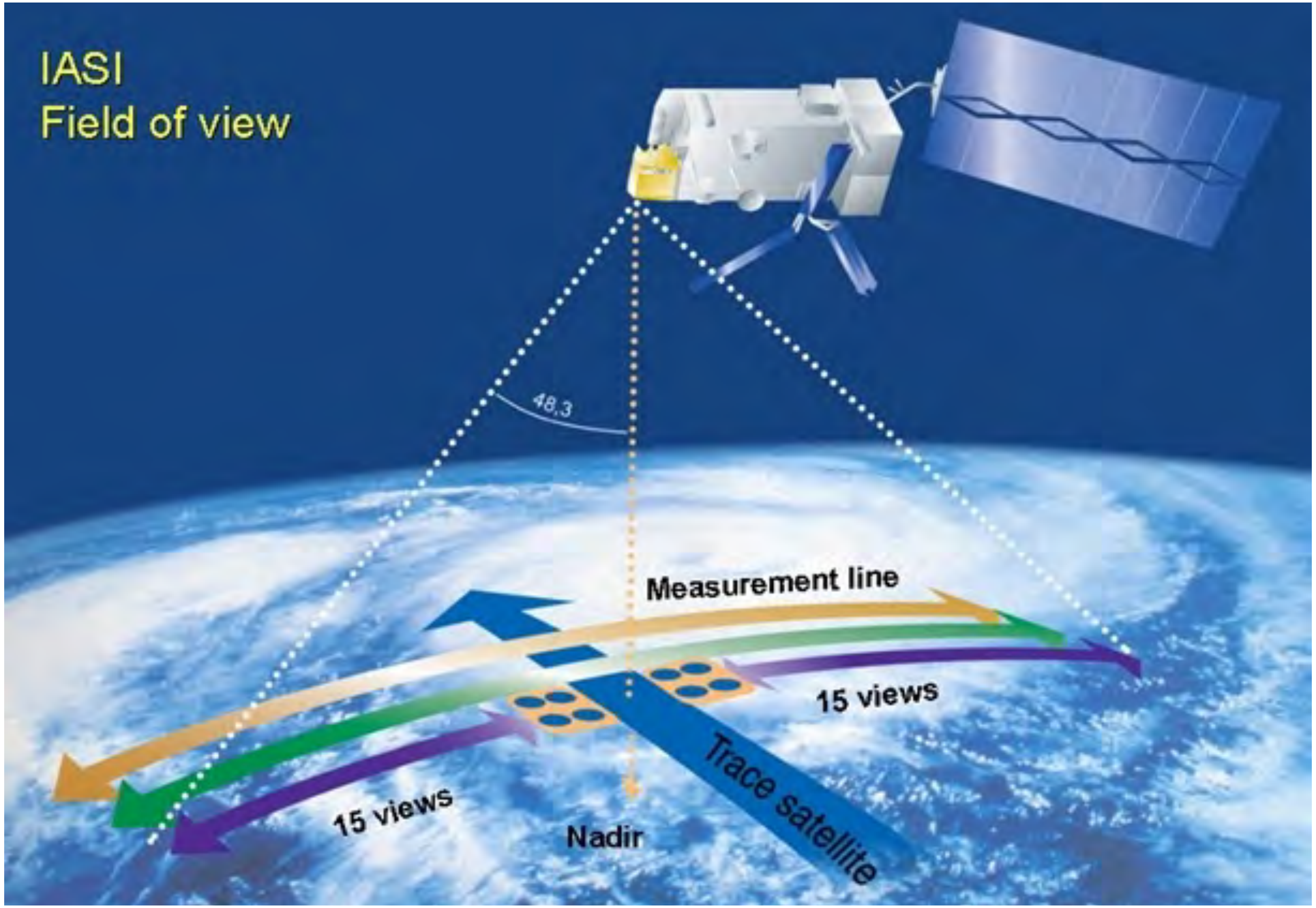
Infrared Atmospheric Sounding Interferometer (IASI) Instrument

<u>Overpass:</u>
9:30 local solar time

<u>Spatial resolution:</u>
12 km to 39 km

<u>Swath width:</u>
2200 km

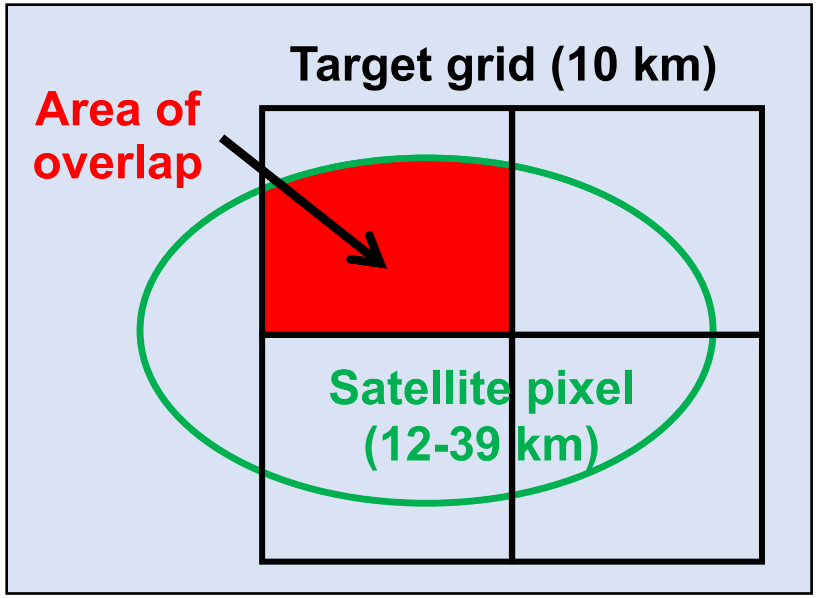
<u>Launch date:</u>
October 2006



Ammonia emissions in the UK: the top-down perspective

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

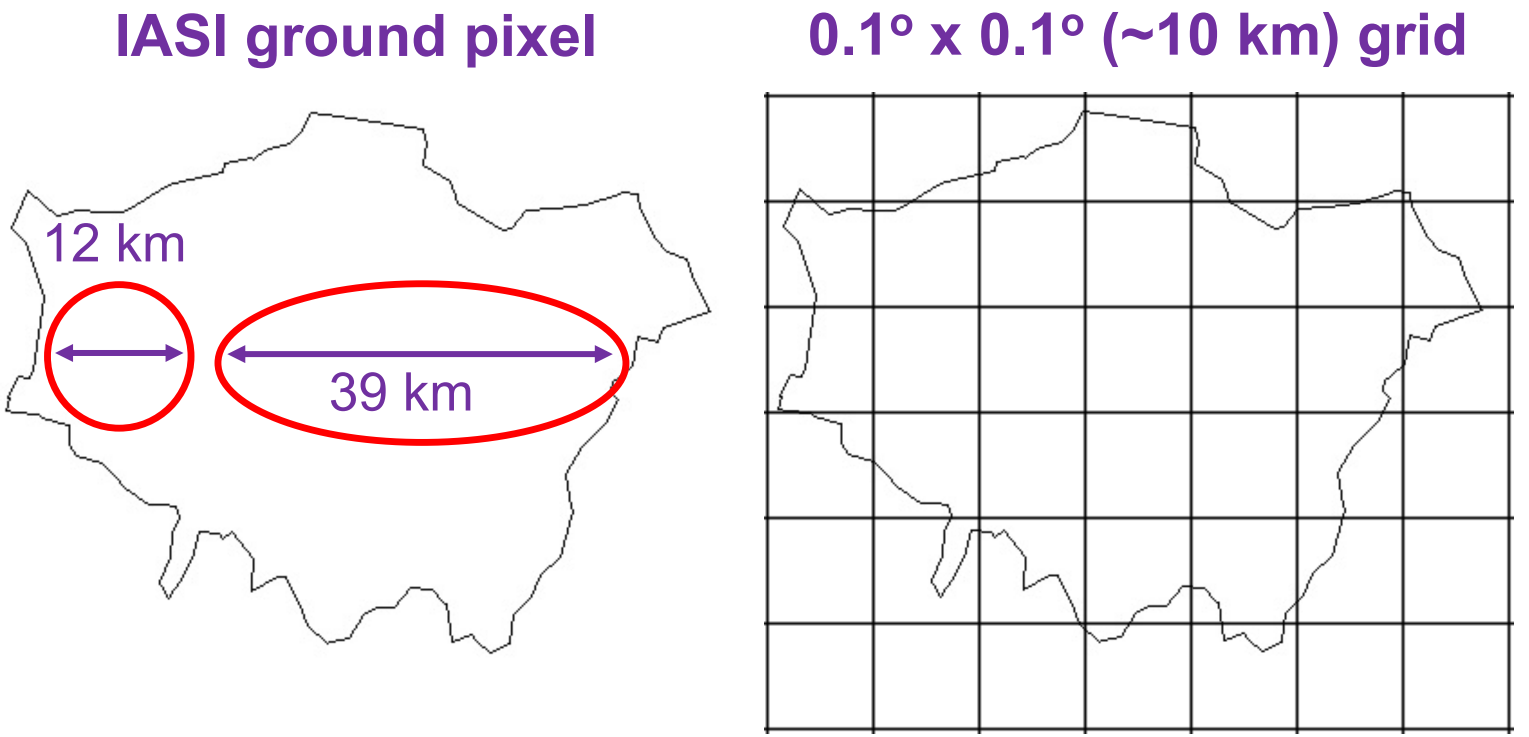
Oversampling Technique



Weights each IASI NH_3 pixel by area of overlap and the reported uncertainty

<u>Oversampling code</u>: L. Zhu, SUSTech (Zhu et al., 2017)

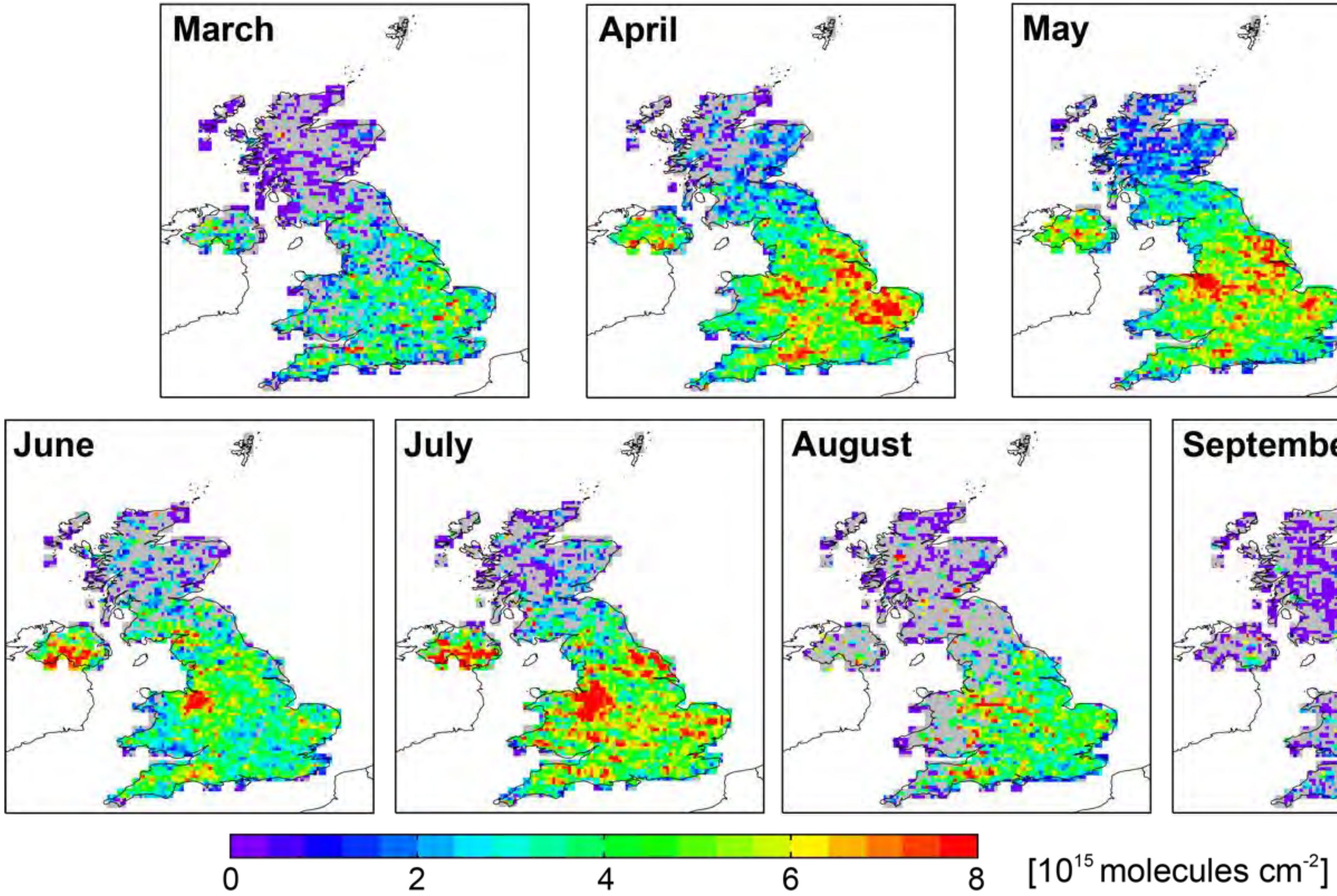
Oversampling technique over London



Lose time (temporal) resolution; gain spatial resolution

Multiyear (2008-2018) monthly mean IASI NH_3 at 0.1° x 0.1°

IASI NH_3 retrieved using spectral enhancement due to NH_3 and machine learning (neural network)

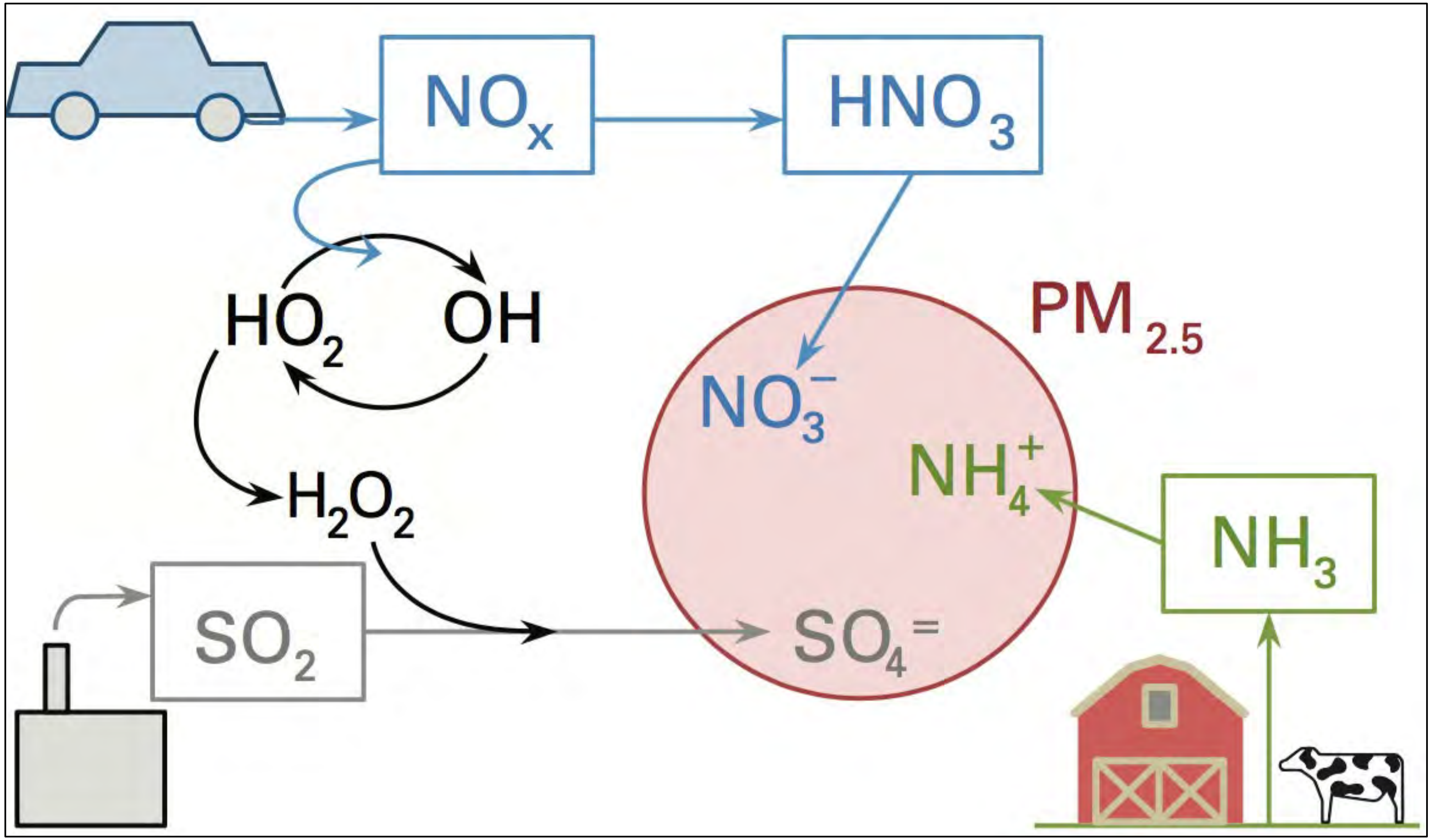


Data for Oct-Feb and over Scotland have low signal and large relative retrieval error (> 100%)

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 $PM_{2.5}$



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

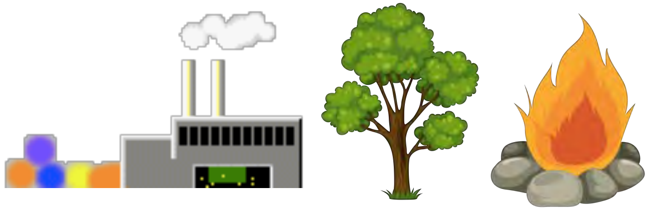
Partitioning of ammonia (NH_3) to pre-existing aerosols depends on abundance of NO_x and SO_2

Surface SO_2 concentrations calculated with GEOS-Chem

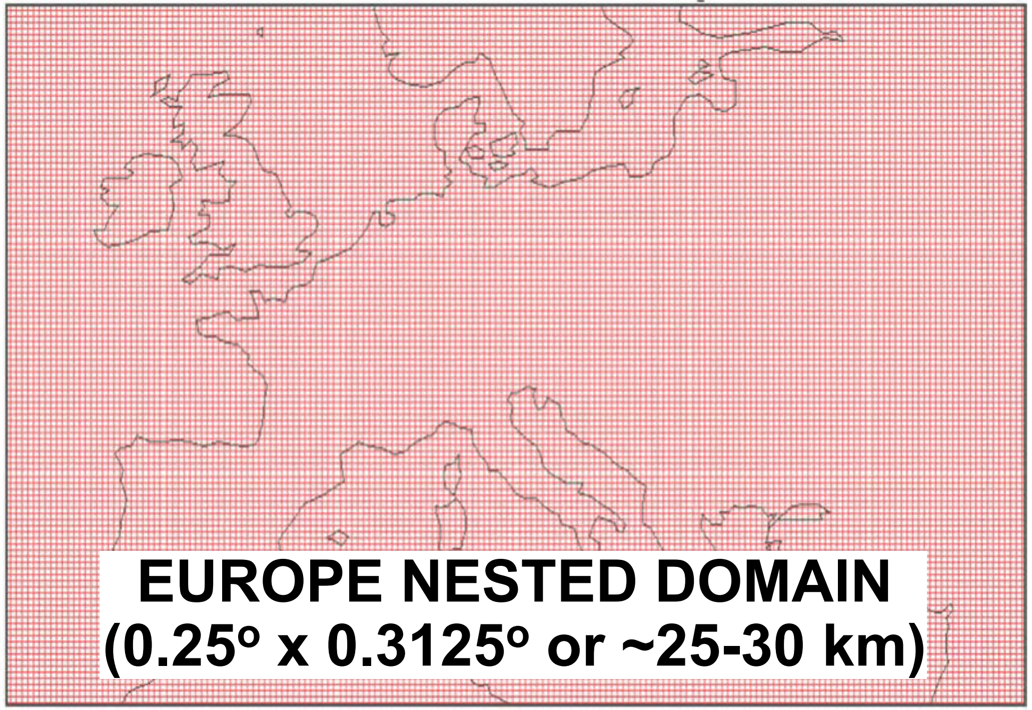
GEOS-Chem

3D Atmospheric Chemistry Transport Model

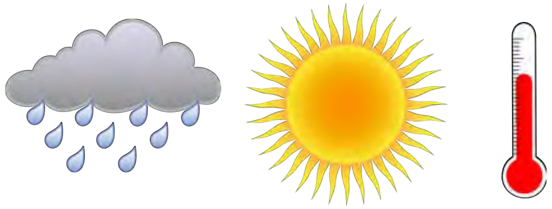
Emissions (natural/human)



UK NAEI emissions
(with temporal information)



Offline assimilated meteorology

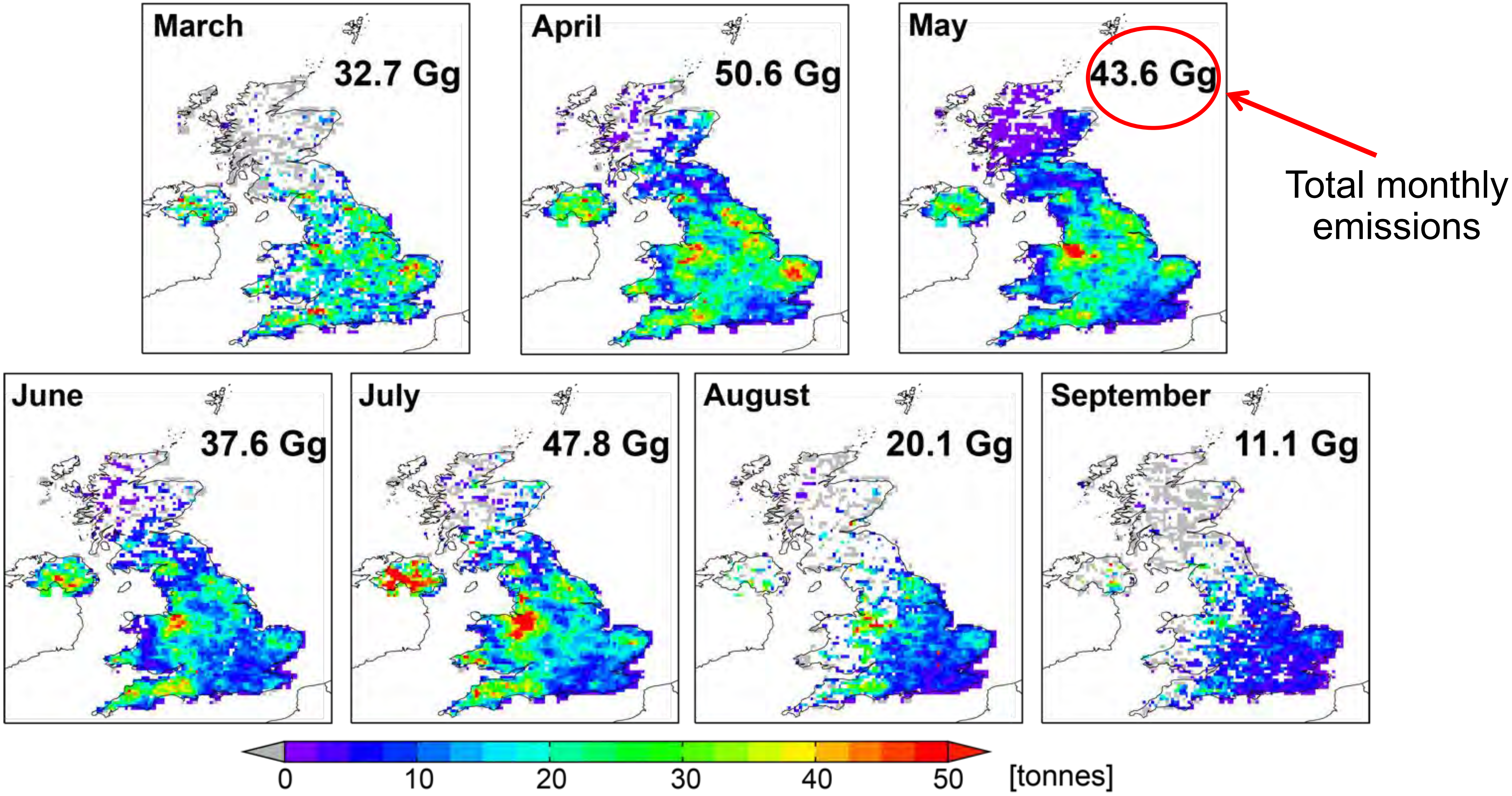


NASA GEOS-FP for 2016

Gas phase and heterogeneous chemistry
Transport
Dry/wet deposition

GEOS-Chem **version 12.1.0** (doi:10.5281/zenodo.1553349)

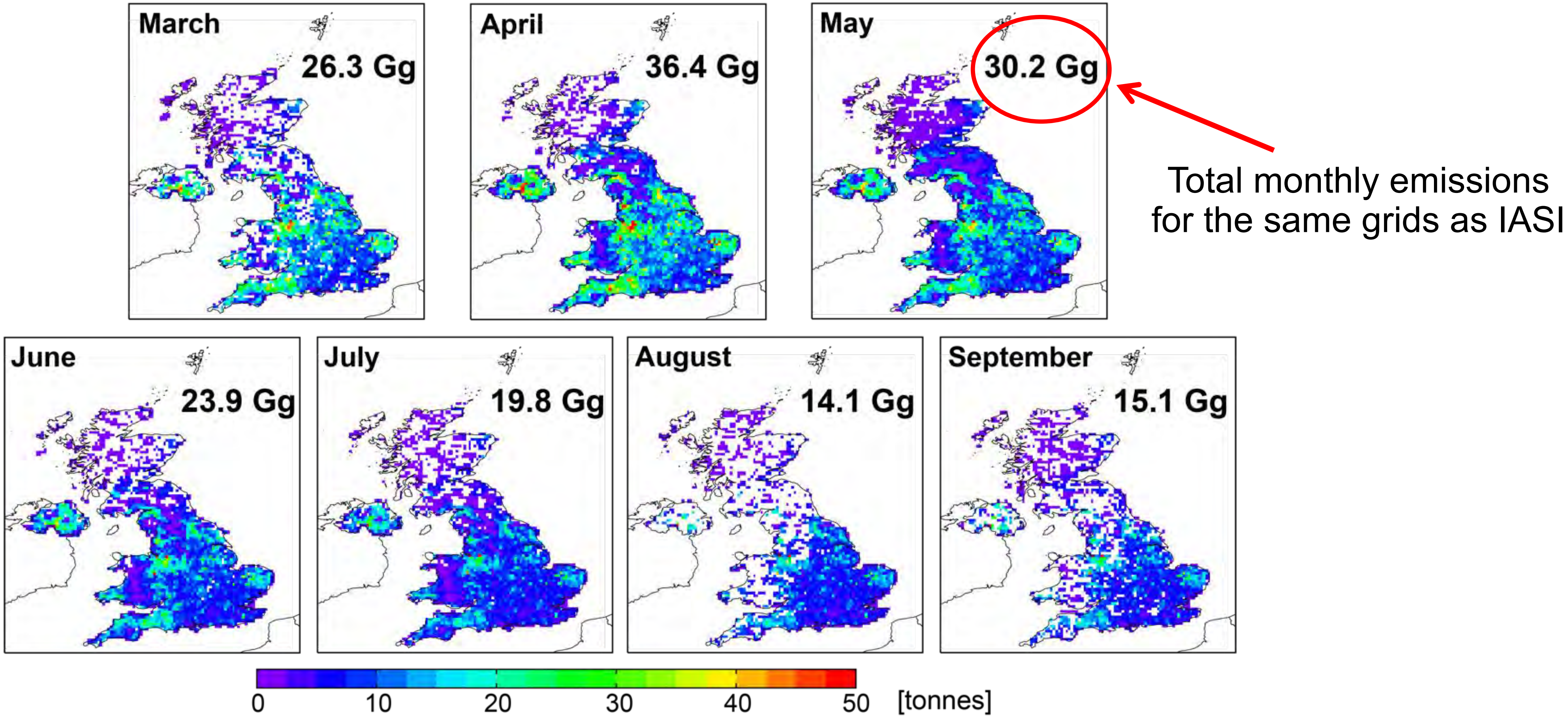
IASI-derived NH_3 emissions at 0.1° x 0.1°



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

Monthly emissions from NAEI and GEOS-Chem at 0.1° x 0.1°

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)

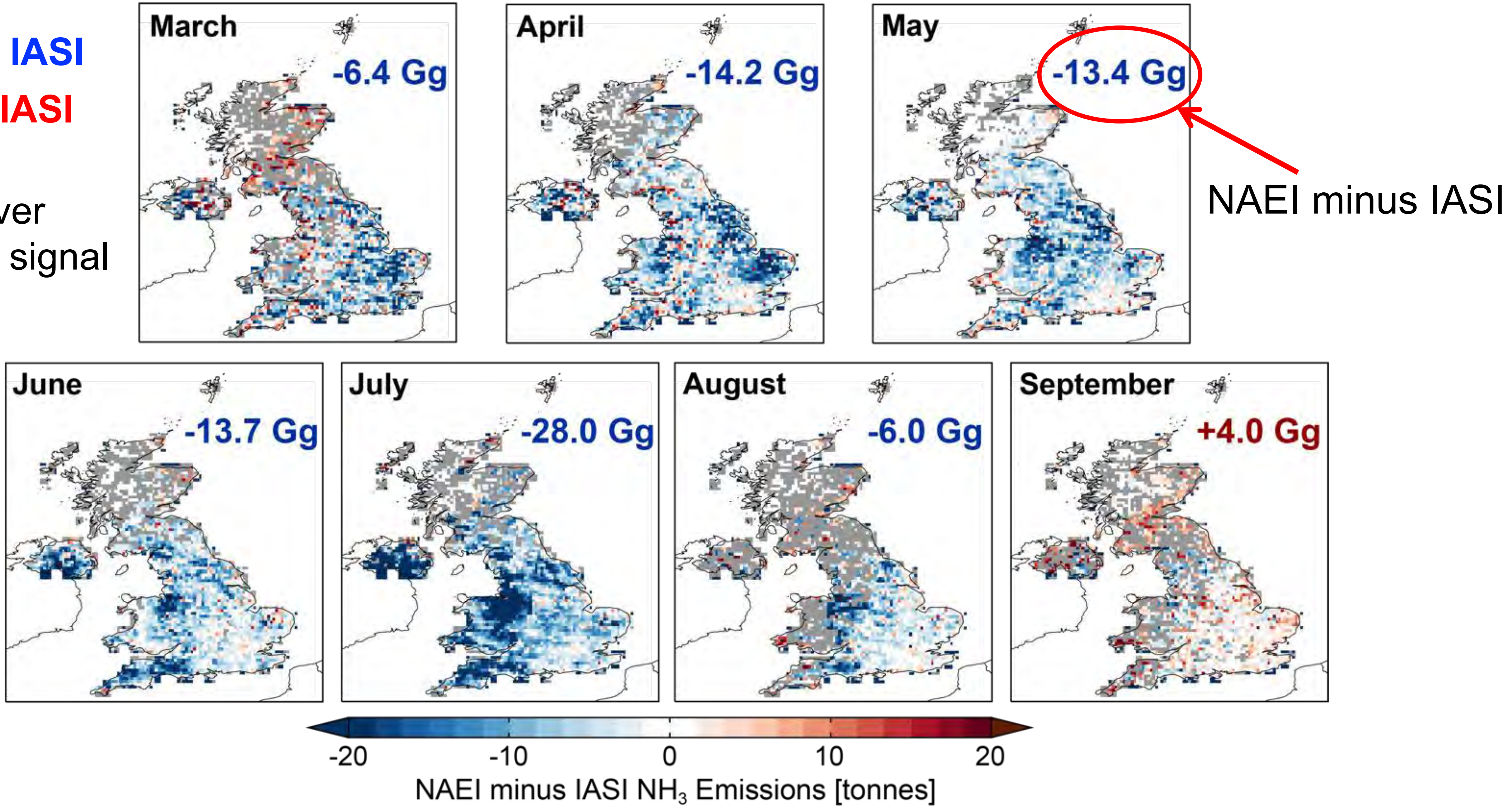
Differences in spatial distribution of IASI and NAEI

IASI-derived emissions minus NAEI-GEOS-Chem emissions

Blue: NAEI < IASI

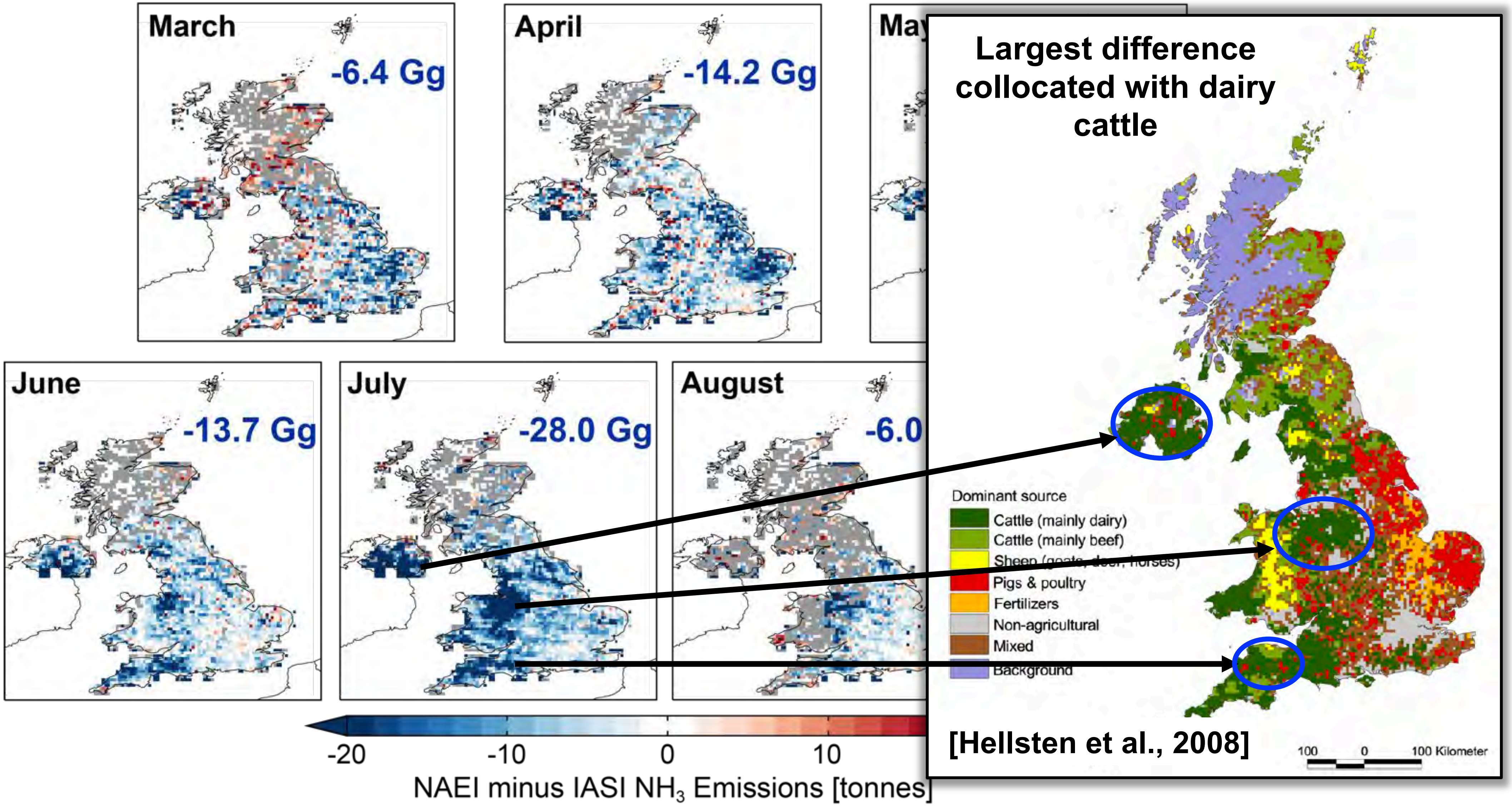
Red: NAEI > IASI

Red mostly over grids with low signal



Differences in spatial distribution of IASI and NAEI

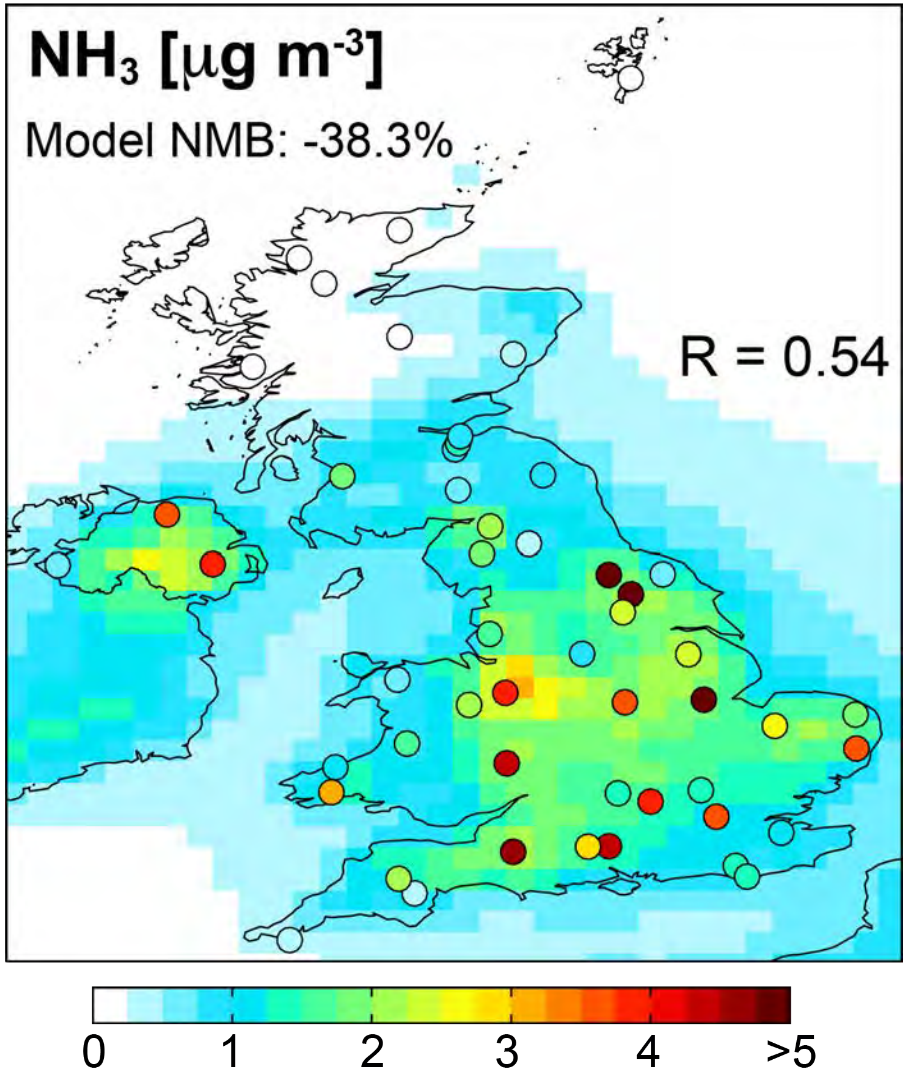
IASI-derived emissions minus NAEI-GEOS-Chem emissions



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

Surface network observations corroborate top-down results

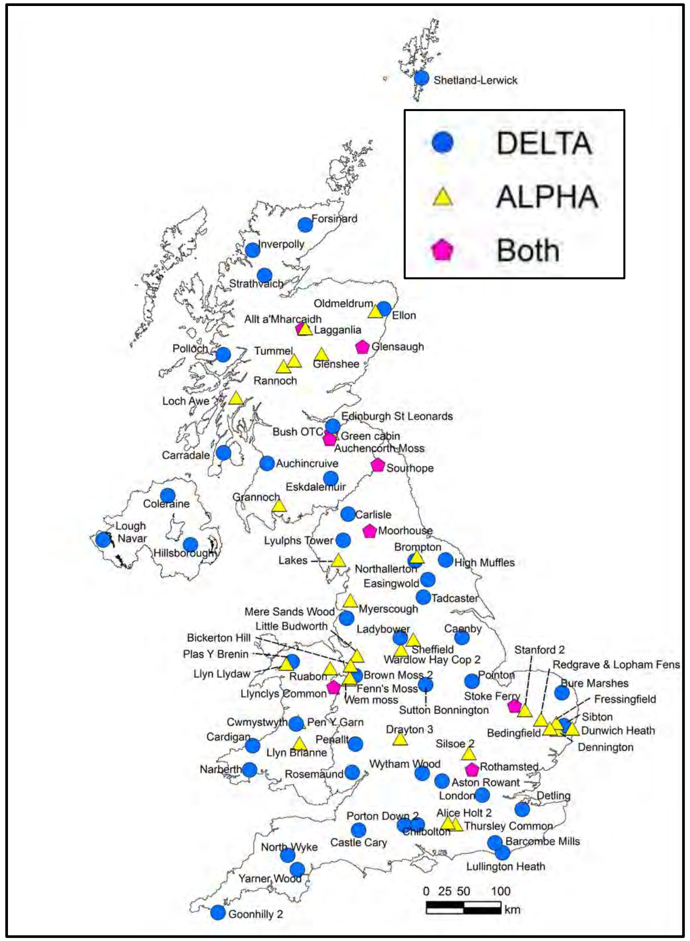
Network (points) and model (background) surface NH_3 in Mar-Sep



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_3 driven with the NAEI corroborates results from IASI

Concluding Remarks for top-down NH_3 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.