Agricultural Ammonia (NH_3) Emissions in the UK

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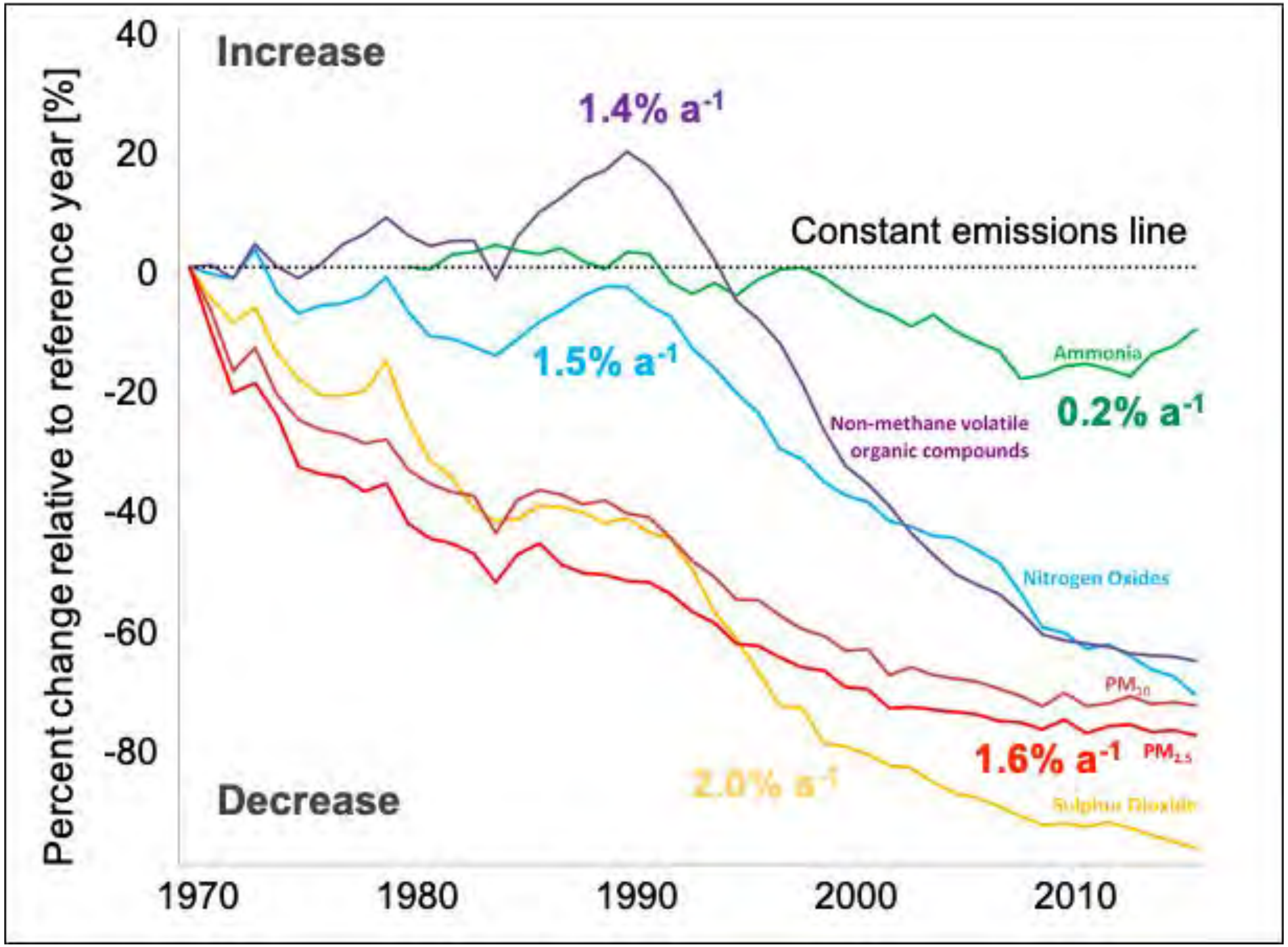
Ammonia emissions in the UK: the bottom-up perspective

Temporal (Time) Variability in Emissions



Green: ammonia

Purple: non-methane volatile organic compounds

Blue: nitrogen oxides

Orange: primary PM_{10}

Red: primary $PM_{2.5}$

Yellow: sulfur dioxide

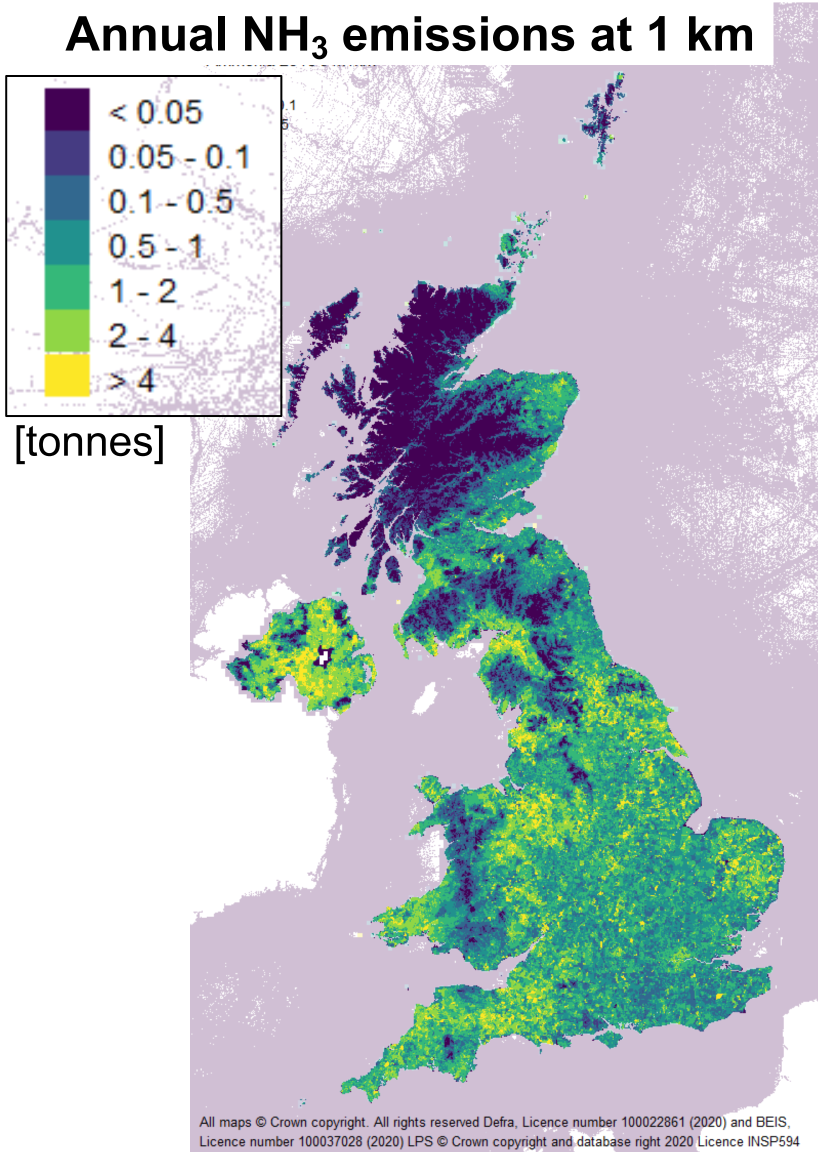
[*Adapted from* Defra, 2018]

Successful decline in all primary $PM_{2.5}$ sources and precursor emissions, except ammonia (NH_3)

Ammonia emissions in the UK: the bottom-up perspective

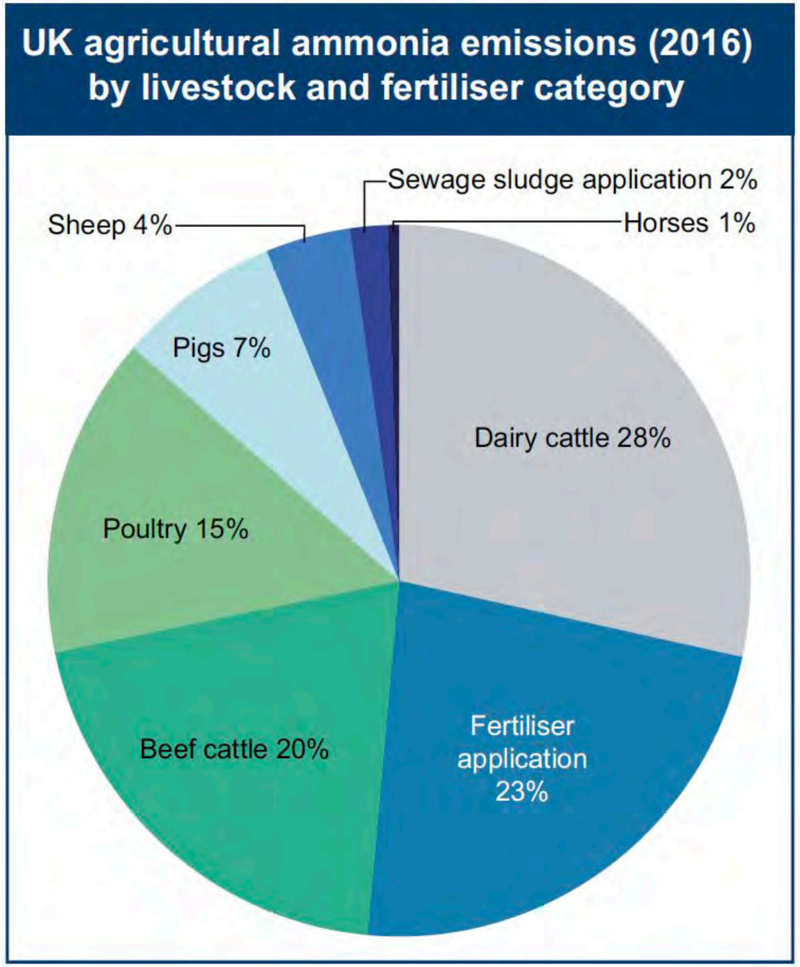
Emissions Spatial Variability

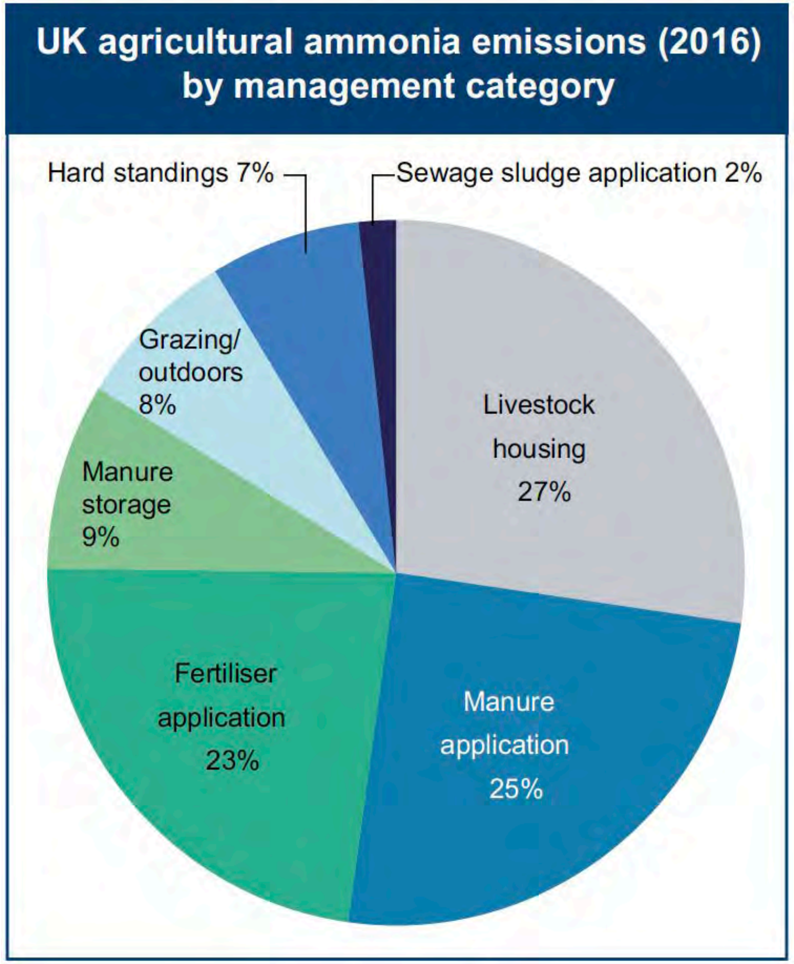
Annual NH_3 emissions at 1 km



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

Contributions of activities to ammonia emissions



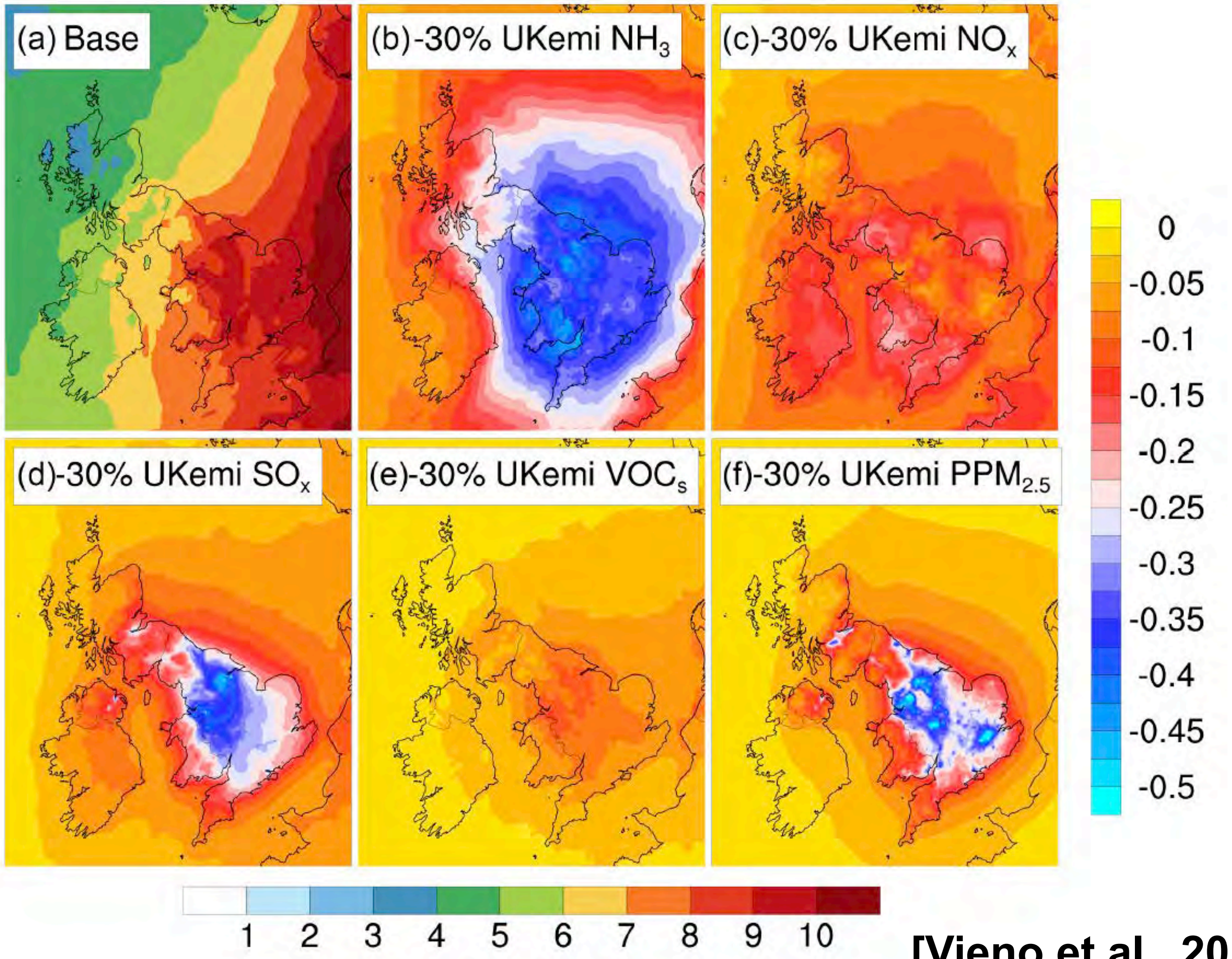


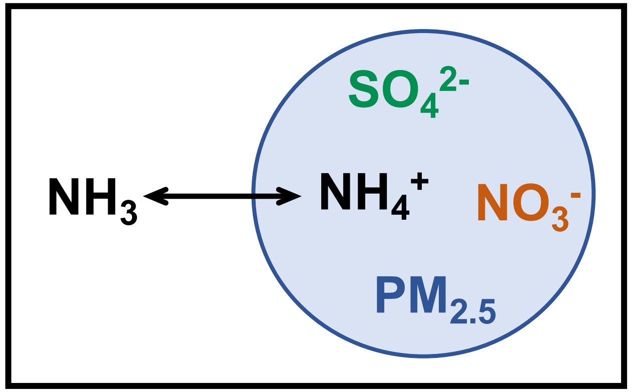
[UK Clean Air Strategy, 2019]

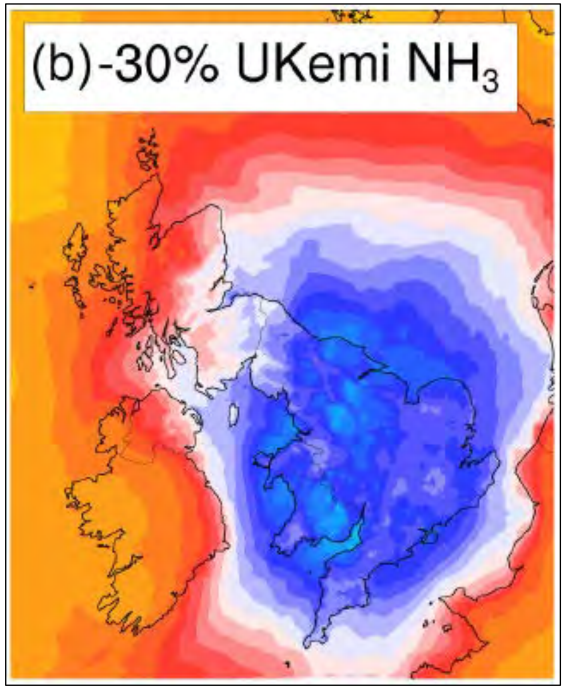
Beef, dairy, and fertilizer use dominate

Ammonia impact on air pollutants hazardous to health

Effect of emission controls on $PM_{2.5}$





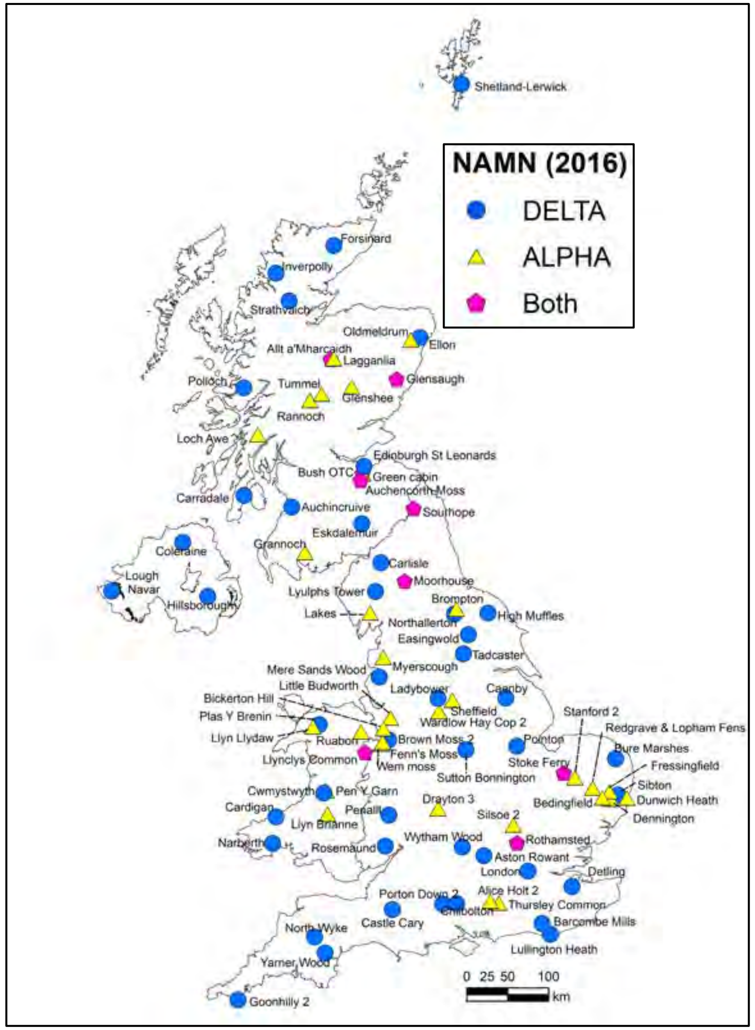


[Vieno et al., 2016]

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

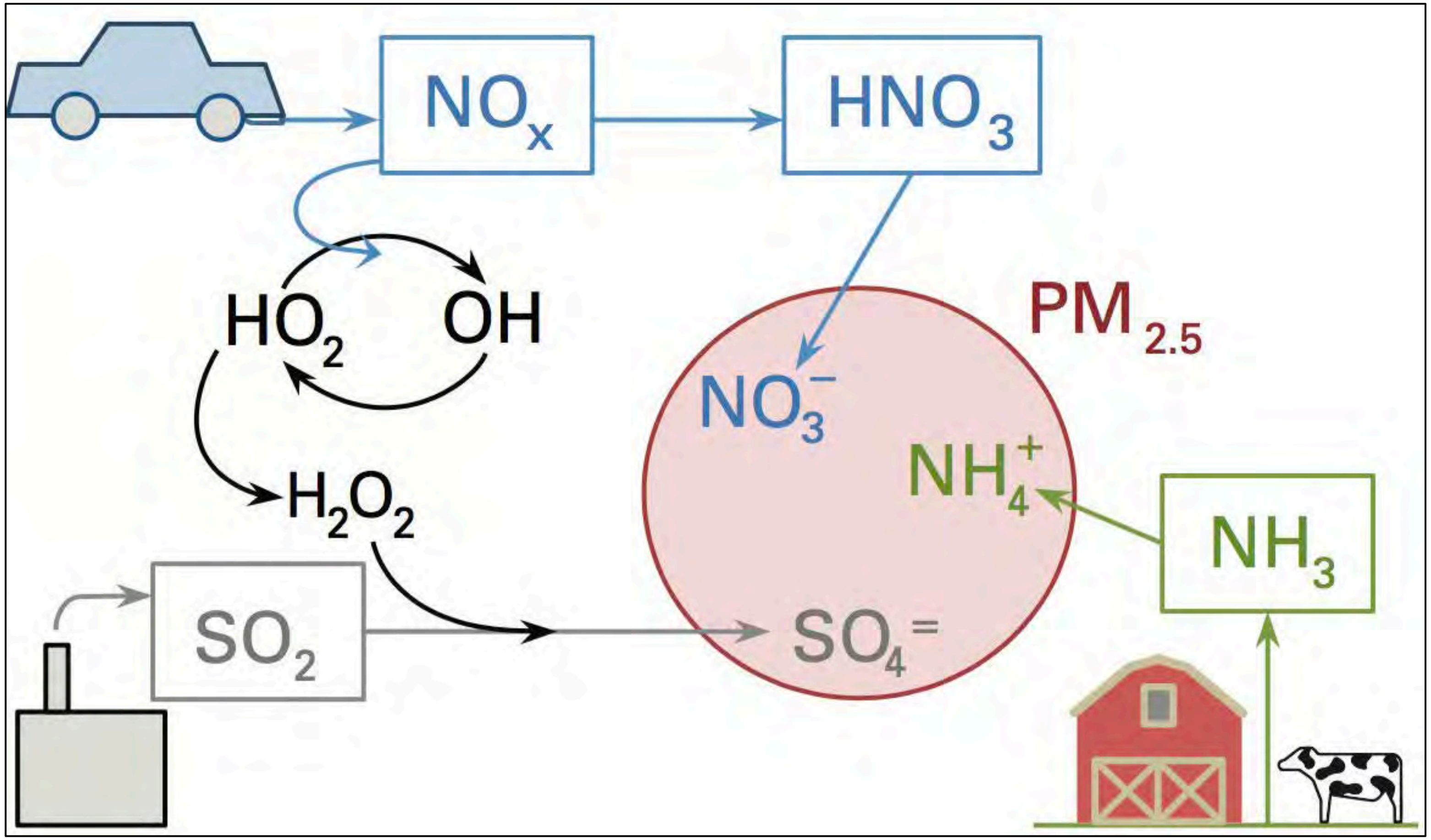
Ammonia emissions are challenging to calculate and validate

Site coverage is limited



Source:
http://www.pollutantdeposition.ceh.ac.uk/content/ammonia-network

Ammonia (NH_3) abundance is complicated

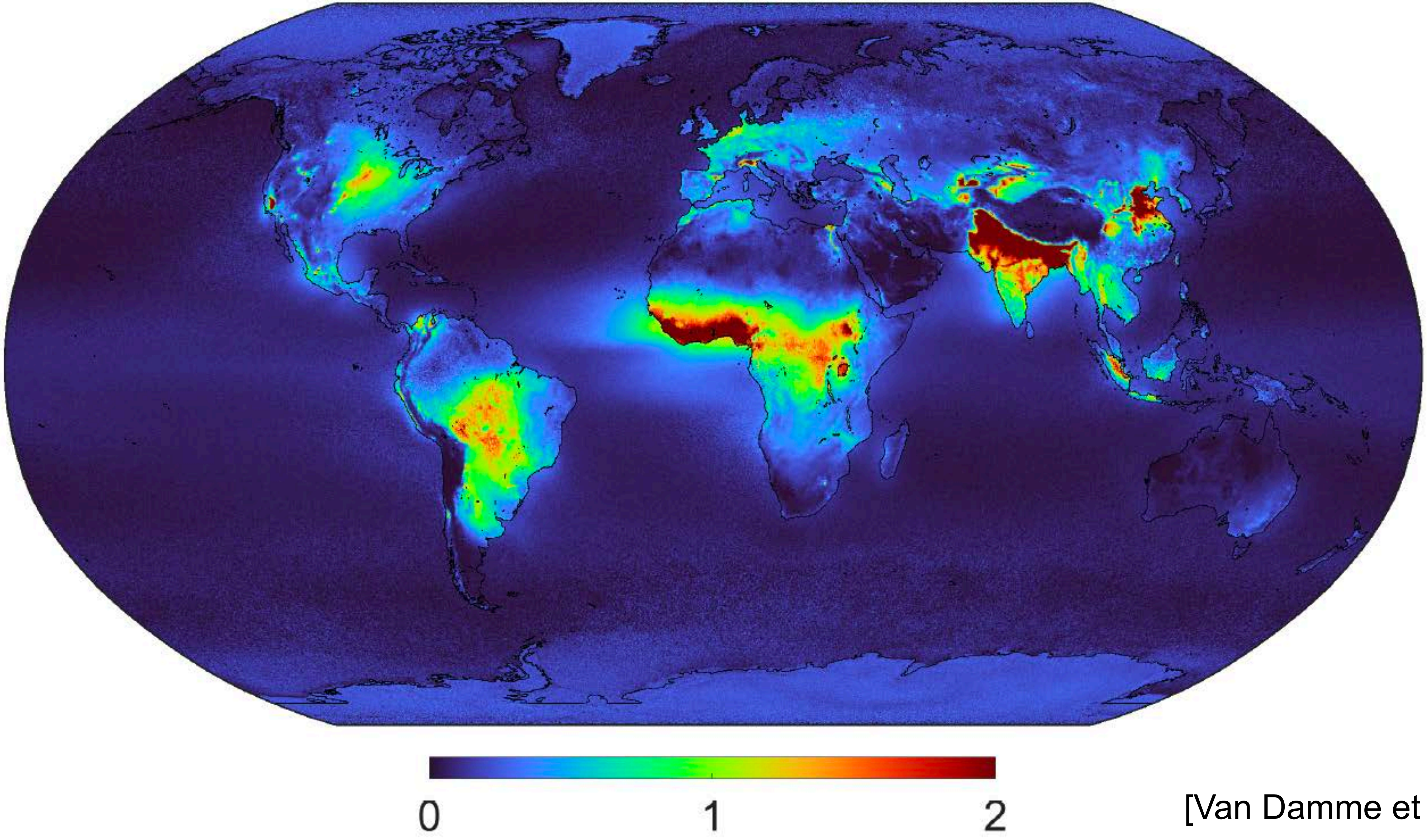


[http://climate-science.mit.edu/]

NH_3 emissions depend on **environmental factors** (T, RH)

Satellite observations offer global coverage of NH_3 columns

NH_3 from Infrared Atmospheric Sounding Interferometer (IASI) for 2008-2018
[10^{16} molecules cm^{-2}]

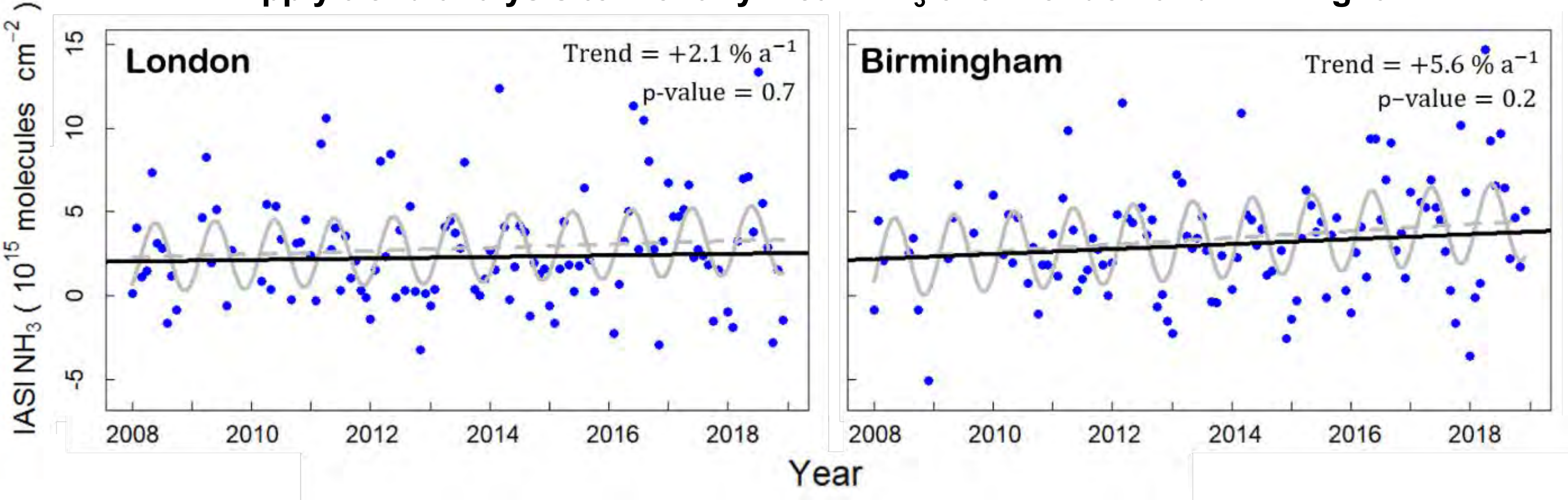


[Van Damme et al., 2020]

Infrared Atmospheric Sounding Interferometer (IASI) Instrument

Exploit the long record (2008-2018) from IASI to assess trends of NH_3 in cities in the UK

Apply trend analysis to monthly mean NH_3 over London and Birmingham



[Vohra et al., accepted, ACP, 2021]

NH_3 concentrations increasing in both cities, but the trend is not significant

Top-down emissions estimated with satellite observations

Convert atmospheric **column concentrations** to surface **emissions** by relating the two with a **model**
Possible as NH_3 has a relatively short lifetime (2-15 hours at or near sources)

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

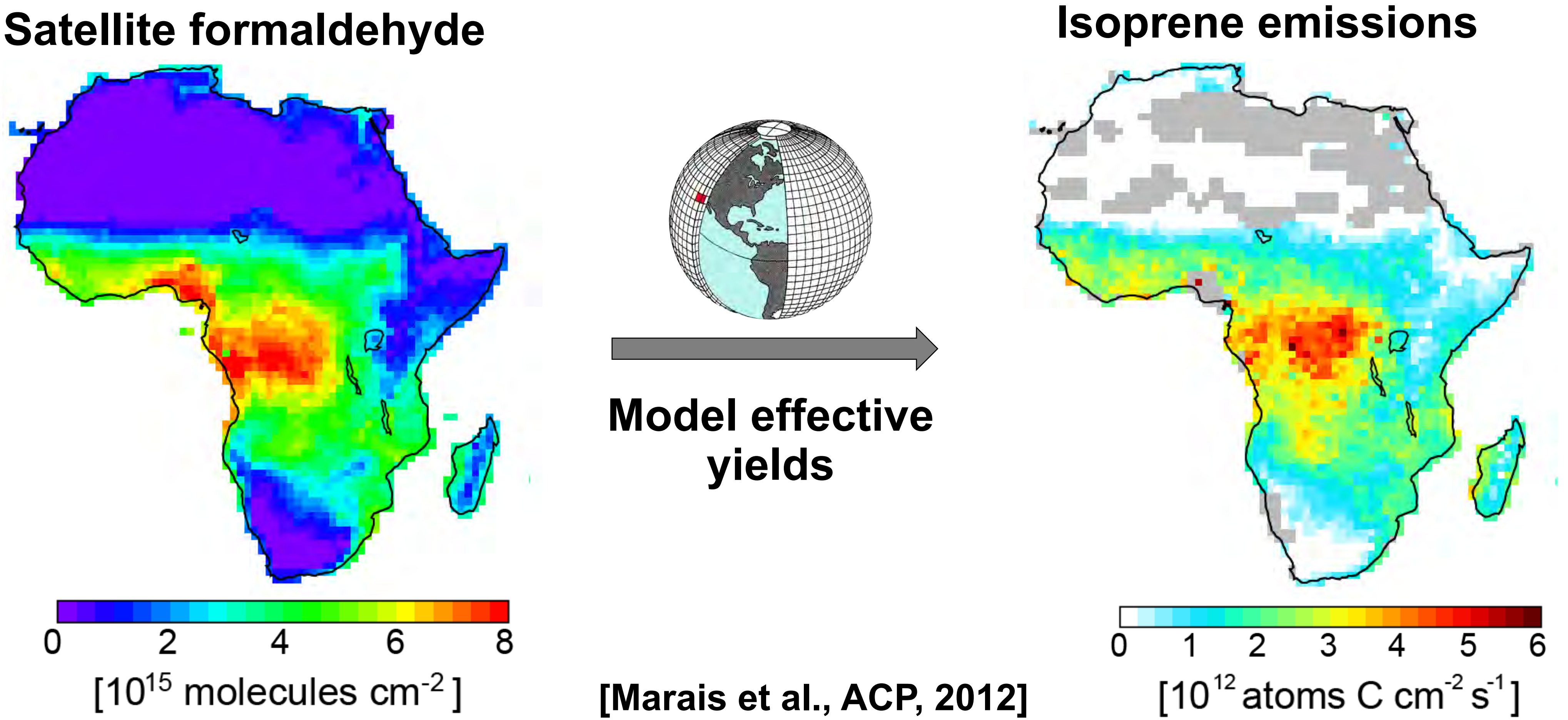
Satellite column densities | **Model Concentration-to-Emission Ratio** | **Satellite-derived Surface Emissions**

Widely used to estimate emissions and surface concentrations

Works for atmospheric components that are short-lived and form promptly and in high yield

Concentrations → emissions: formaldehyde → isoprene, NO_2 → NO_x

Column → surface: formaldehyde → formaldehyde, NO_2 → NO_2, AOD → $PM_{2.5}$



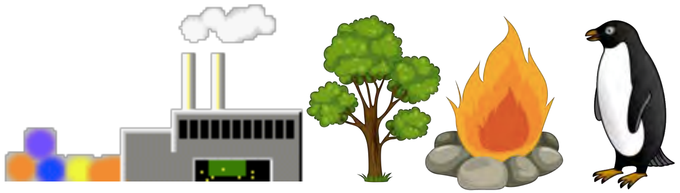
[Marais et al., ACP, 2012]

Surface SO_2 concentrations calculated with GEOS-Chem

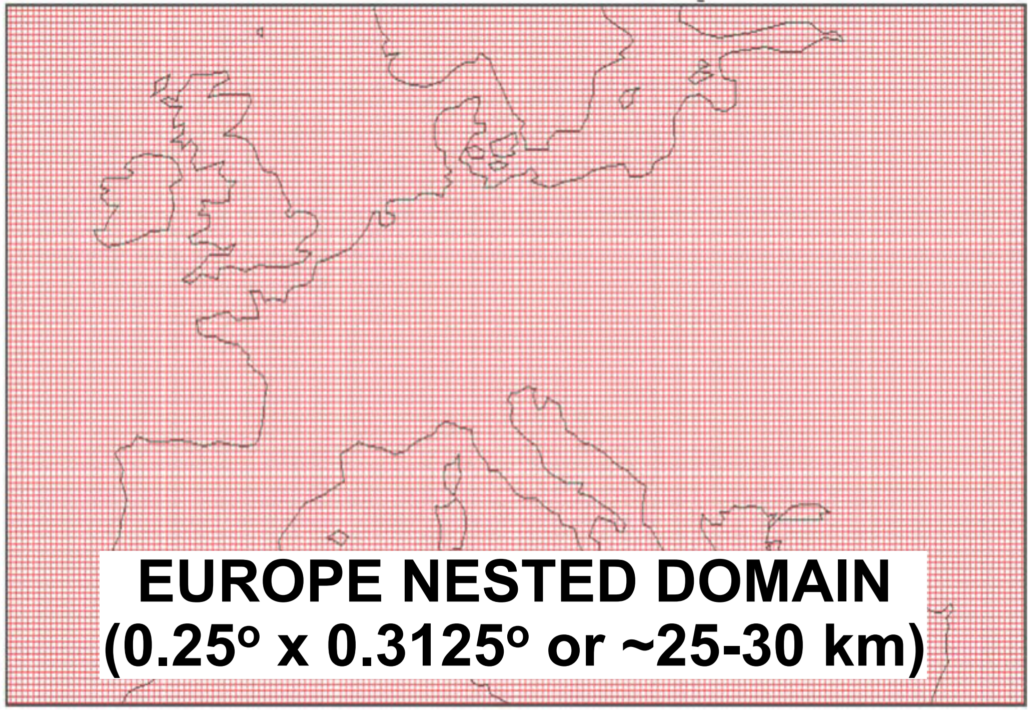
GEOS-Chem

3D Atmospheric Chemistry Transport Model

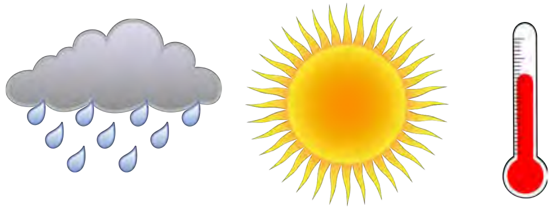
Emissions (natural/human)



UK NAEI emissions
(with temporal information)



Offline assimilated meteorology



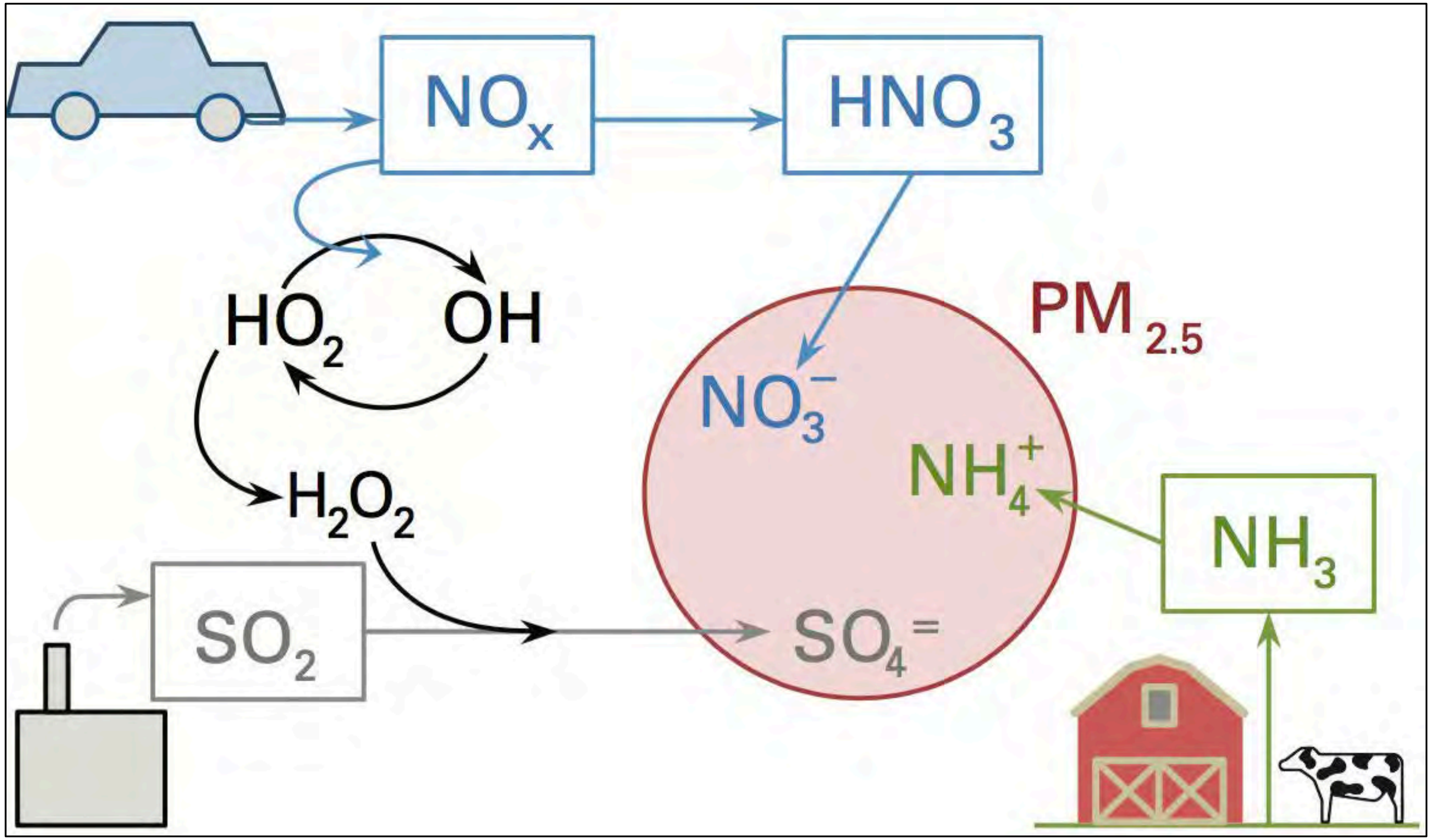
NASA GEOS-FP for 2016

Gas phase and heterogeneous chemistry
Transport
Dry/wet deposition

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

Ammonia abundance depends on numerous factors

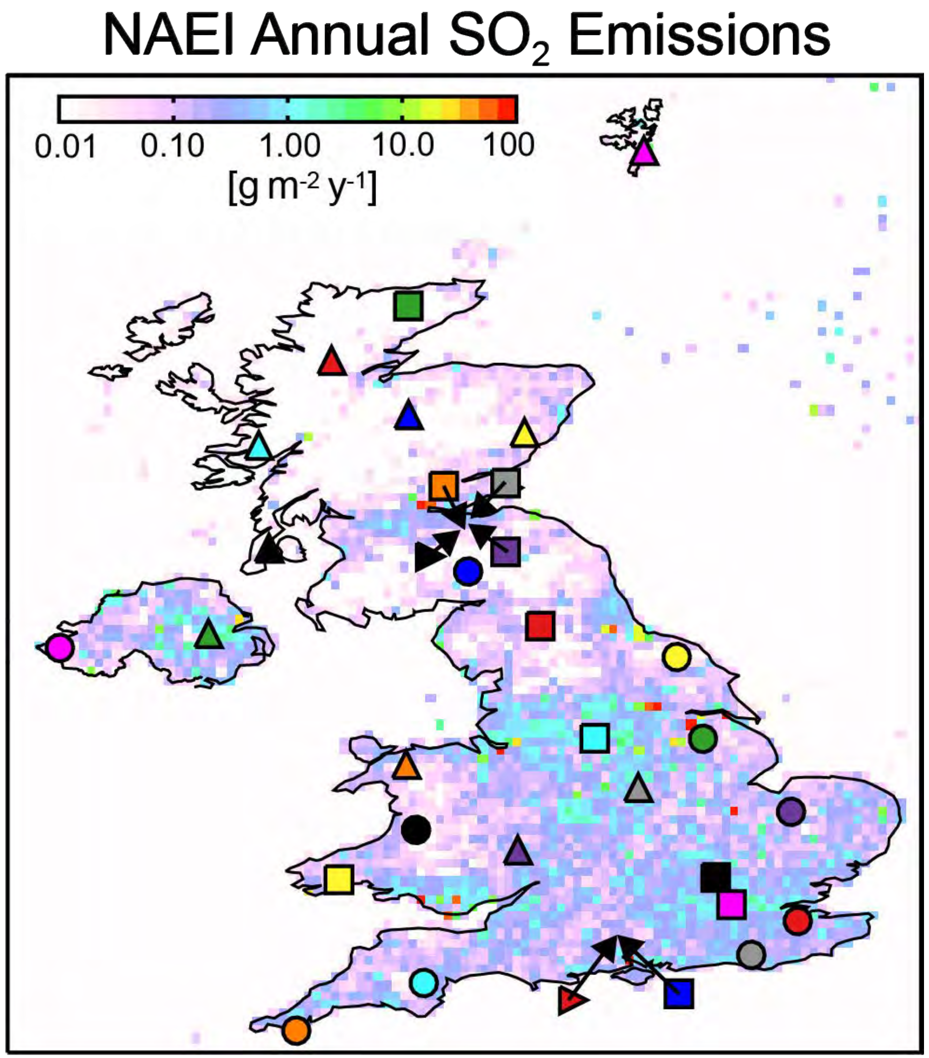
Ammonia buffers acidic aerosols formed when SO_2 oxidizes to form sulfate (SO_4^{2-})



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

Abundance of gas-phase of ammonia (NH_3) depends on emissions of SO_2

Ammonia abundance depends on numerous factors



Symbols: SO_2 concentration monitors

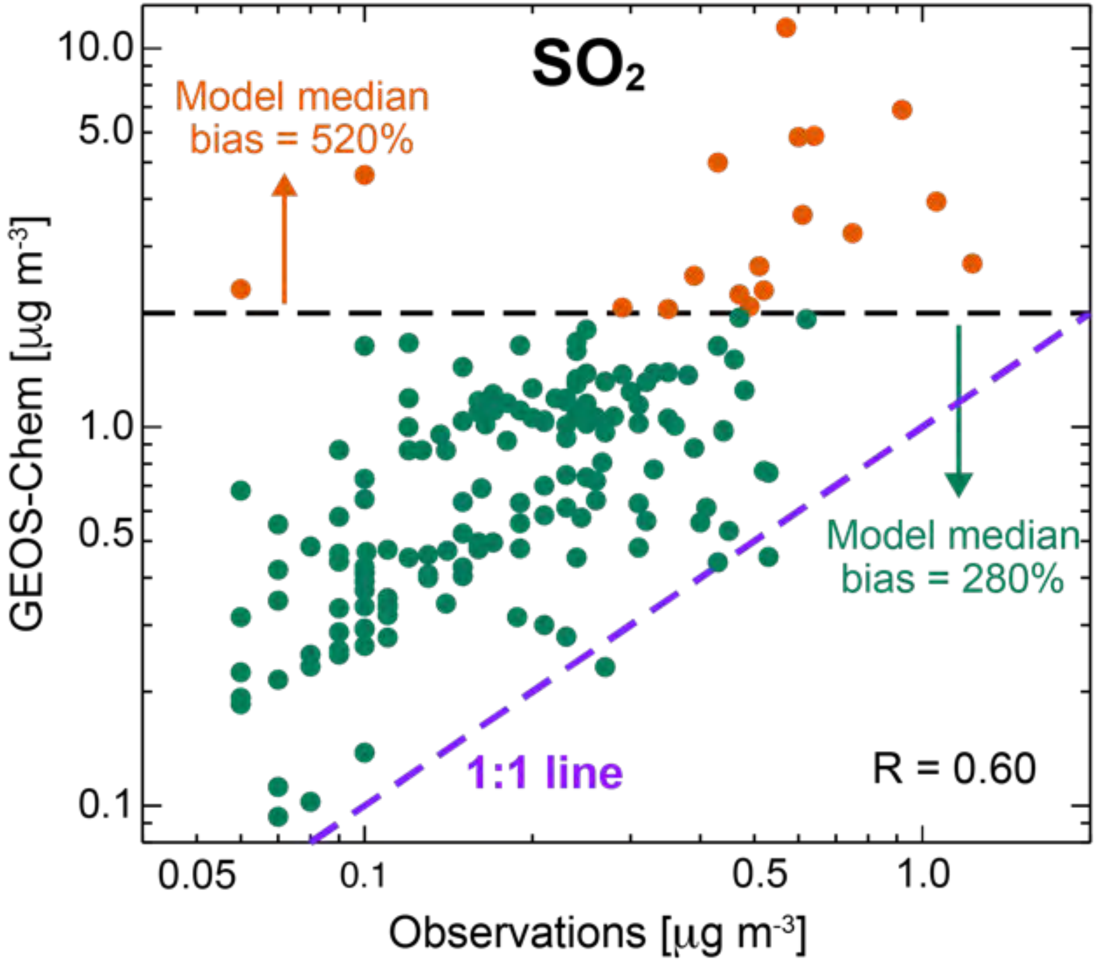
UKEAP:
~30 sites
offline denuder measurements
0.05 μg m^{-3} detection limit

MARGA:
2 sites
semi-continuous denuder measurements
0.04 μg m^{-3} detection limit

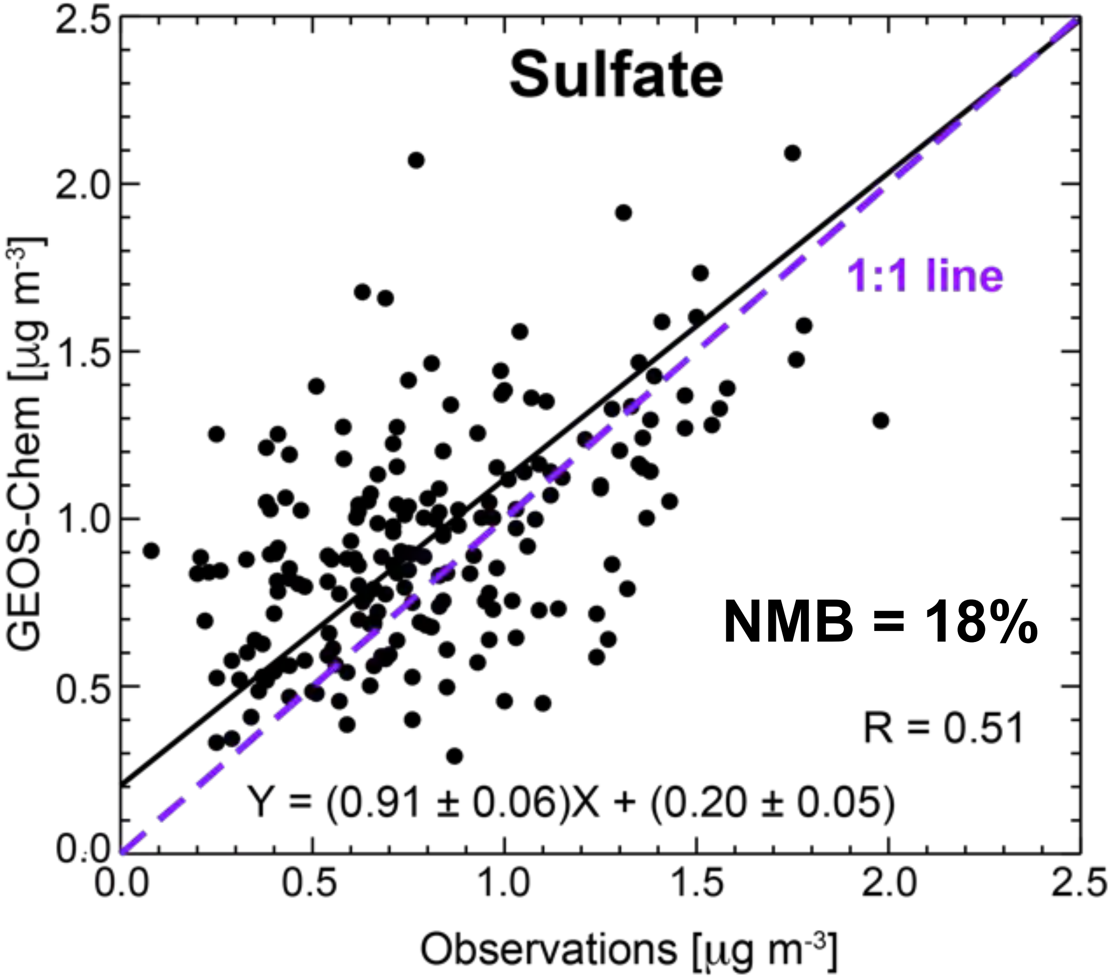
UV fluorescence instruments:
Not used. Poor detection.
4 μg m^{-3} detection limit

Assessment of NAEI SO_2

Modelled vs observed surface SO_2 concentrations



Modelled vs observed surface sulfate concentrations



Model overestimate in SO_2 and sulfate supports overestimate in SO_2 emissions. Possibly due to overestimate in SO_2 biomass emissions?

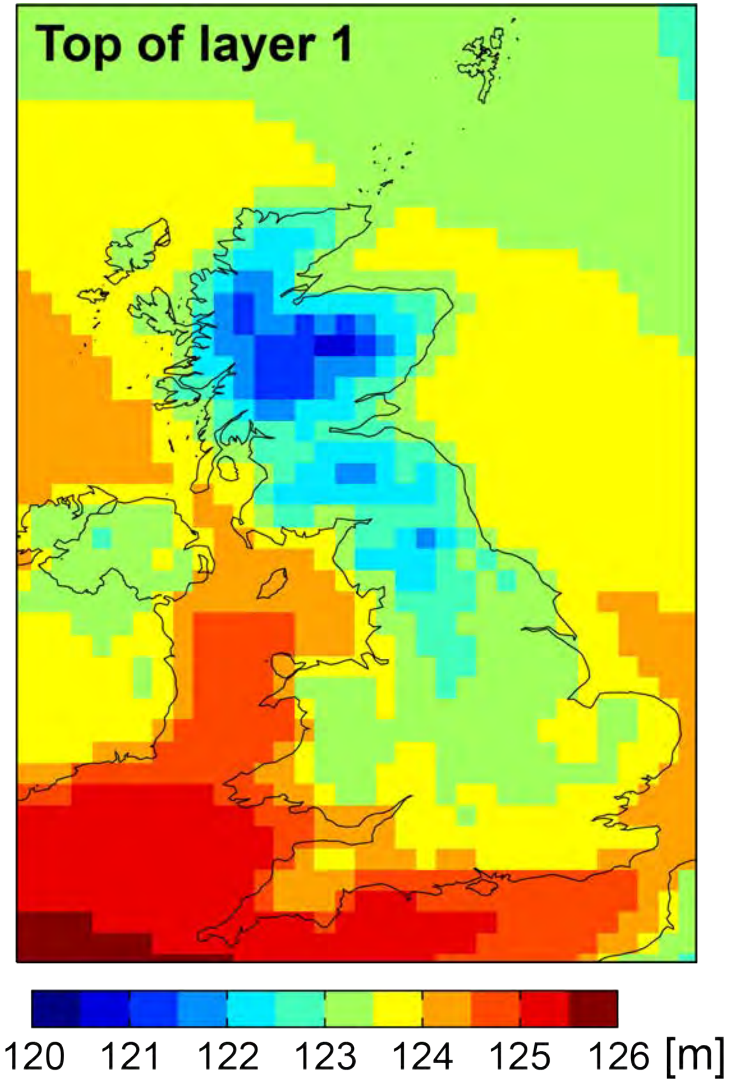
Vertical distribution of SO_2 point sources

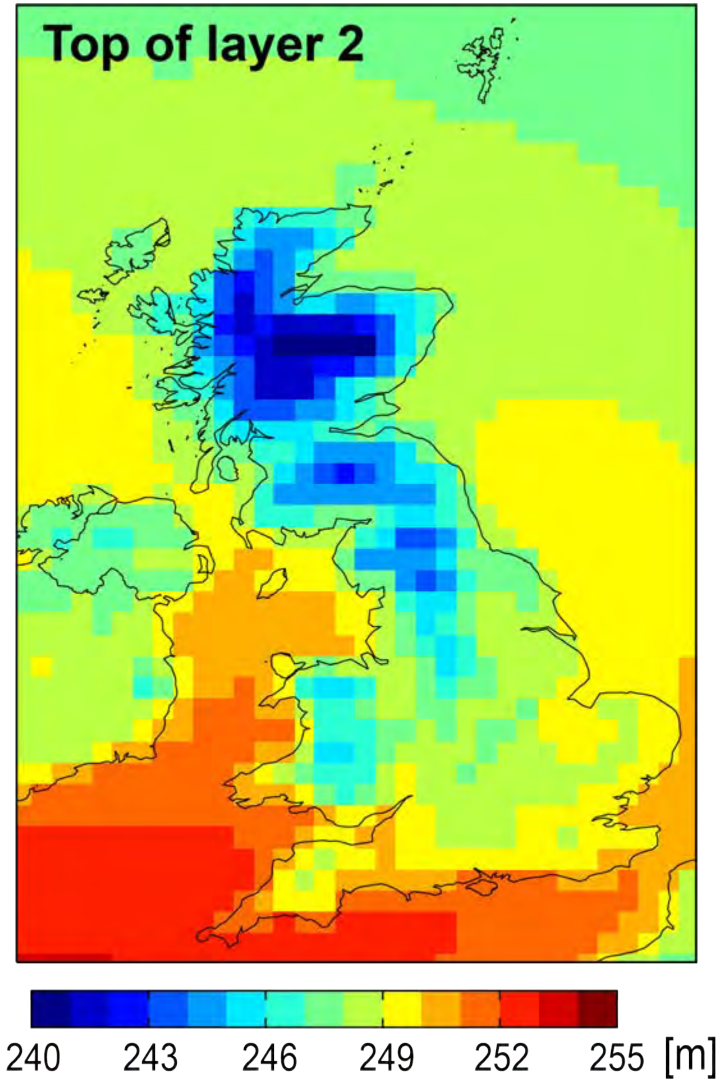
No vertical distribution data provided with the NAEI emissions

Drax cooling towers: 114 m
Drax chimney: 259 m
Other tall stacks: 160-240 m

GEOS-Chem:
Layer 1: 120-126 m
Layer 2: 240-255 m

Map of annual average model layer heights





Vertical distribution of SO_2 point sources

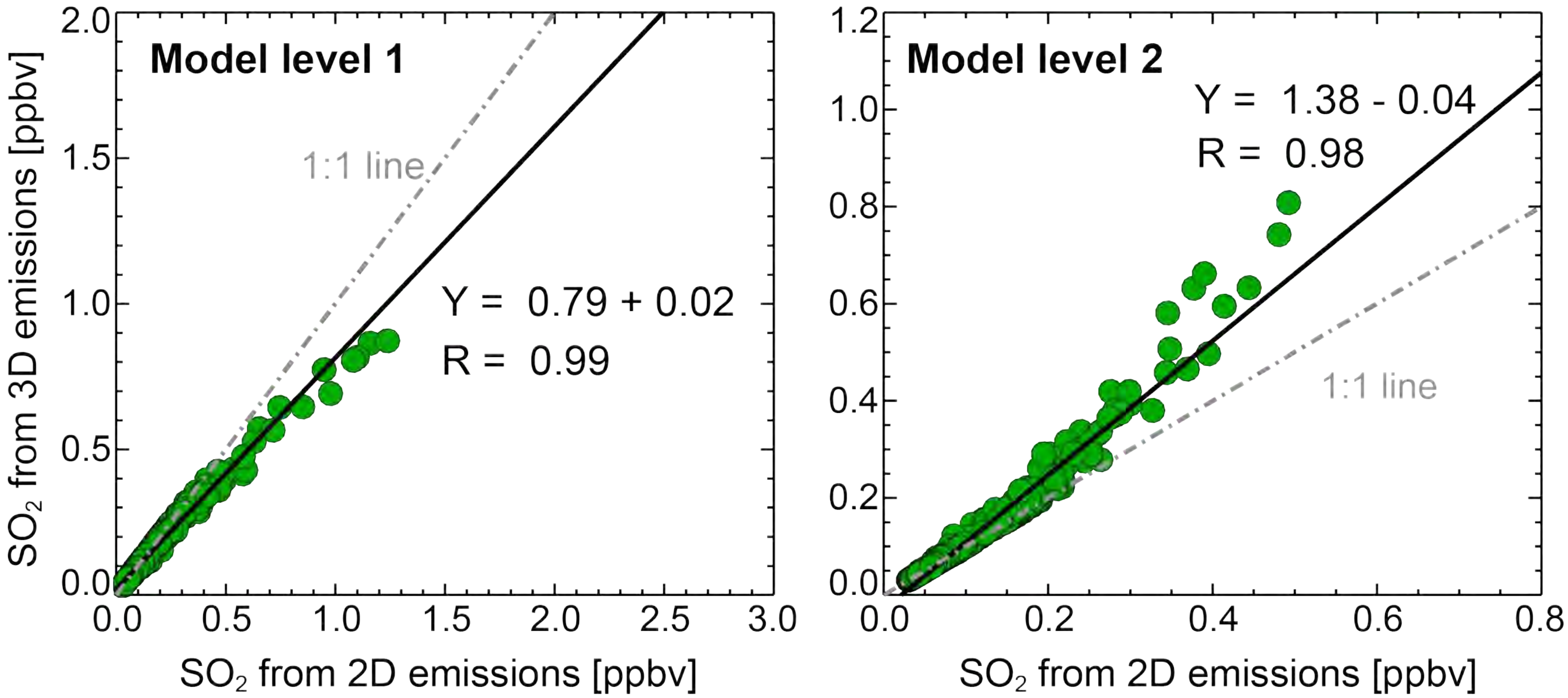
Test with GEOS-Chem the effect of placing ALL point source emissions of SO_2 in model layer 2

2016 SO_2 emissions:

All land-based: 164 Gg

Point sources: 96 Gg

Effect of 3D SO_2 emissions on SO_2 concentrations in **July**



Extreme test leads to 20% decrease in surface SO_2, 38% increase in layer 2. Similar results for July.

Decrease annual NAEI SO_2 emissions 161 Gg to 84 Gg and rerun GEOS-Chem

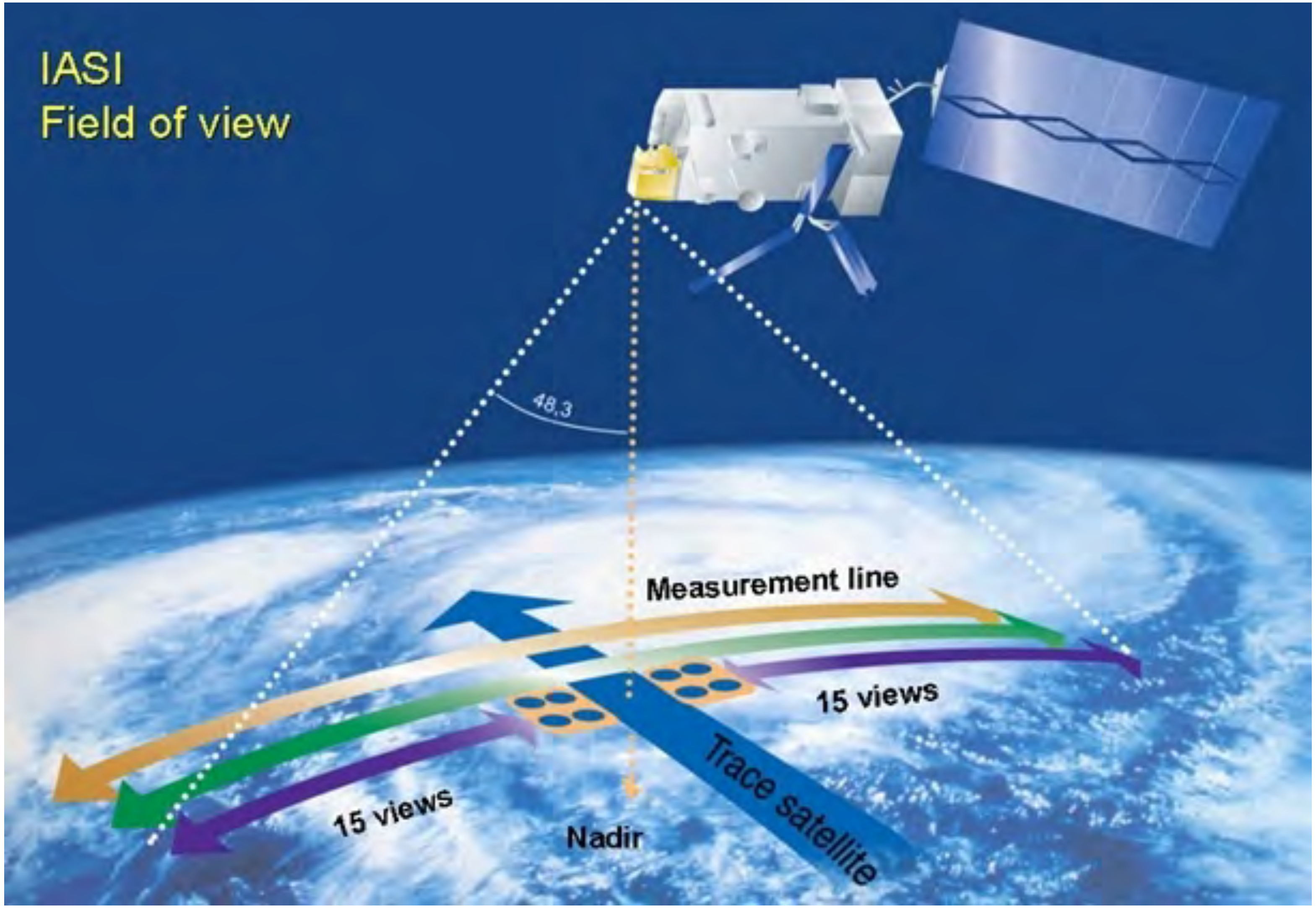
Infrared Atmospheric Sounding Interferometer (IASI) Instrument

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

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

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

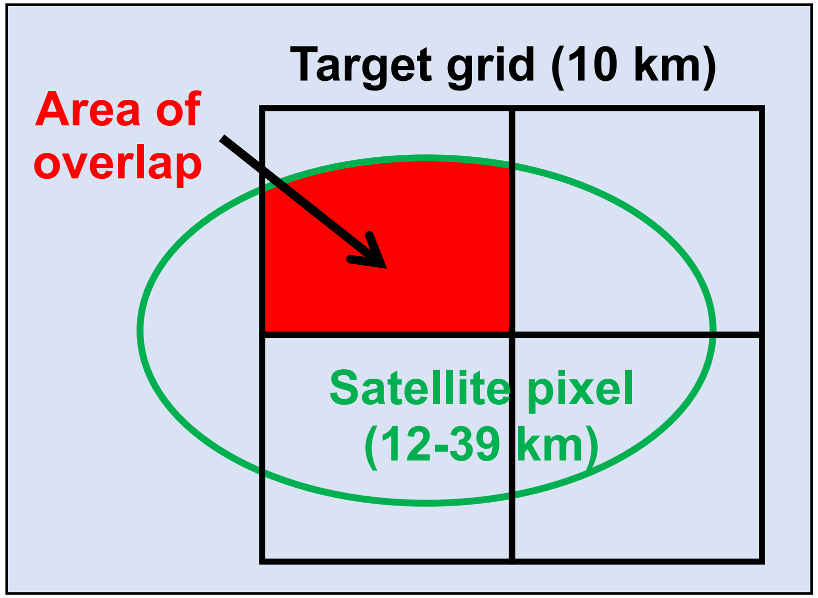
<u>Launch date:</u>
October 2006



Fine-scale sampling of IASI using Oversampling

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

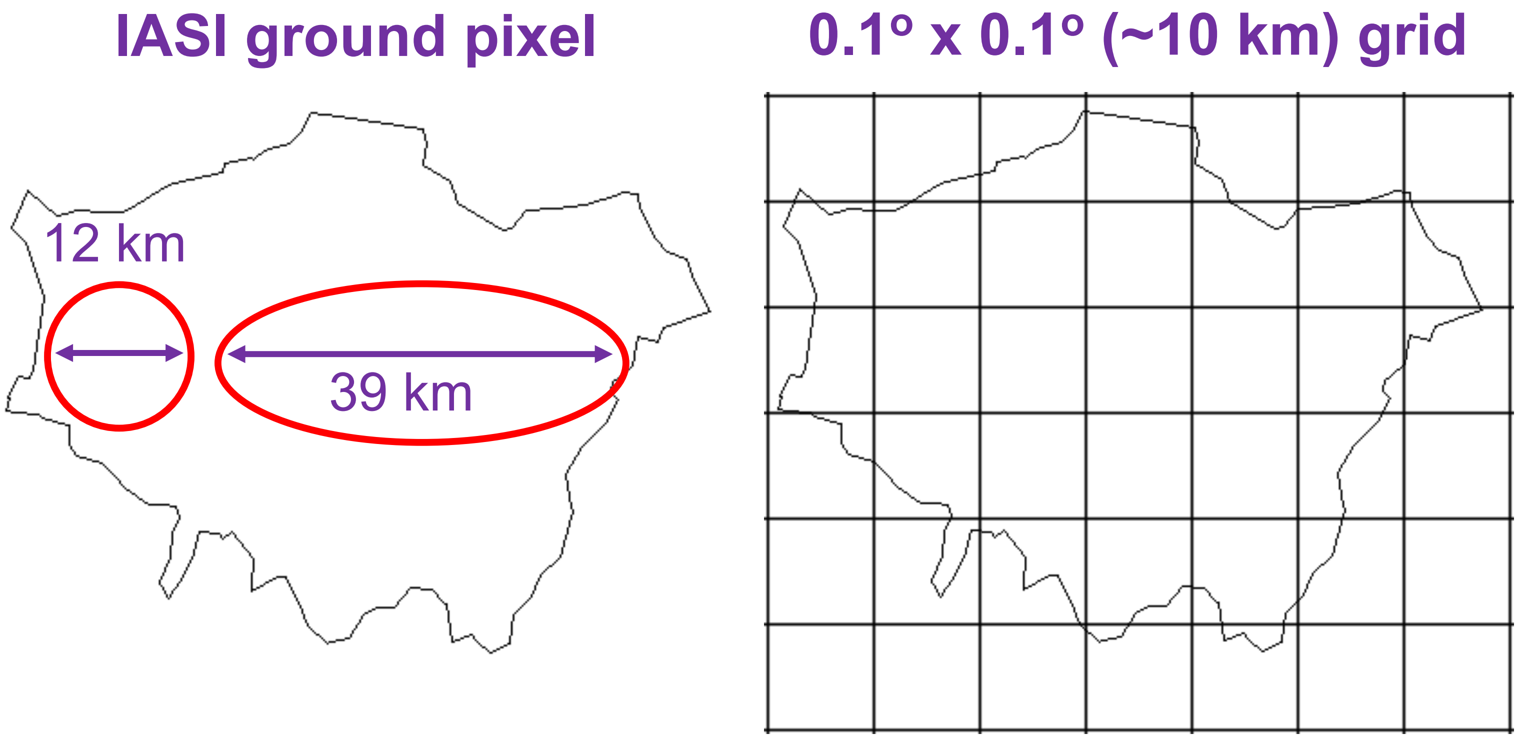
Oversampling Technique



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

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

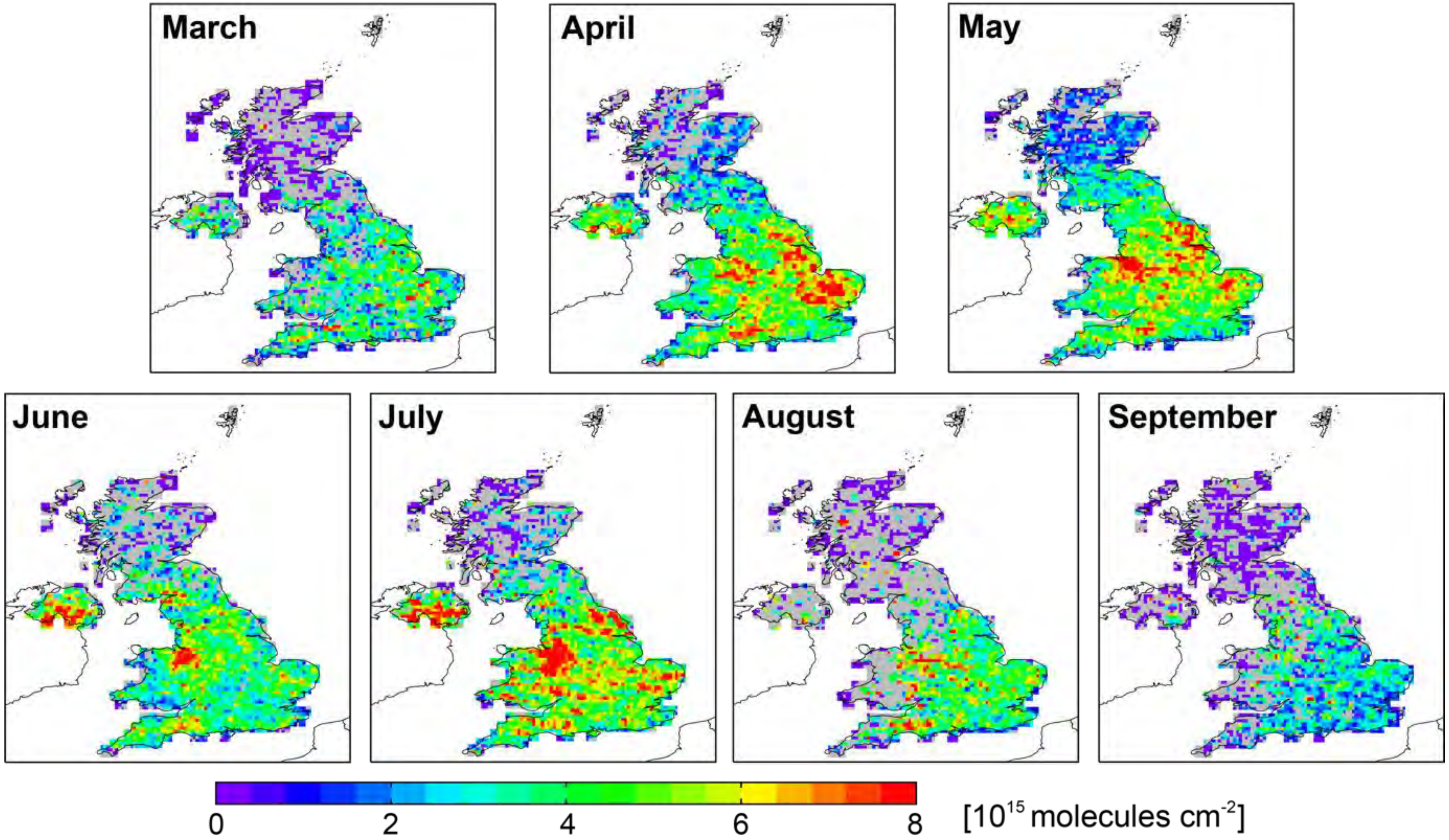
Oversampling technique over London



Trade off temporal resolution for finer spatial resolution

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

IASI NH_3 retrieved using machine learning (neural network)



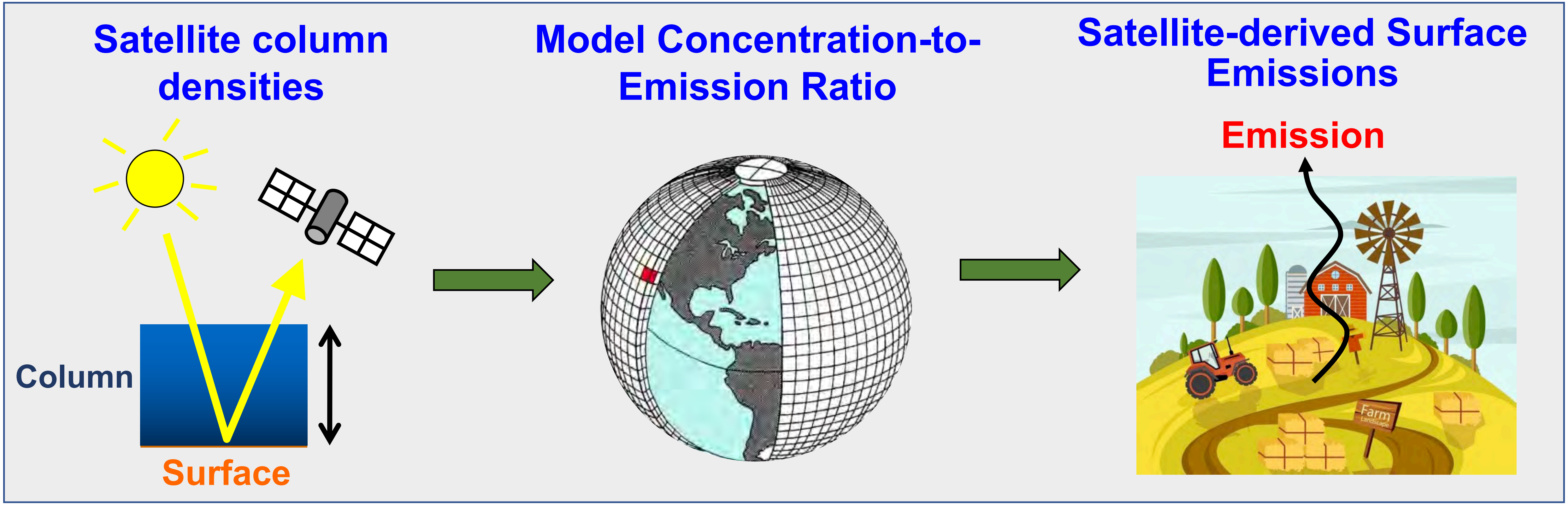
Data for Oct-Feb and over Scotland have low signal and large relative retrieval error (> 100%)

<u>IASI data providers</u>: M. Van Damme, L. Clarisse, P.-F. Coheur, ULB, Belgium

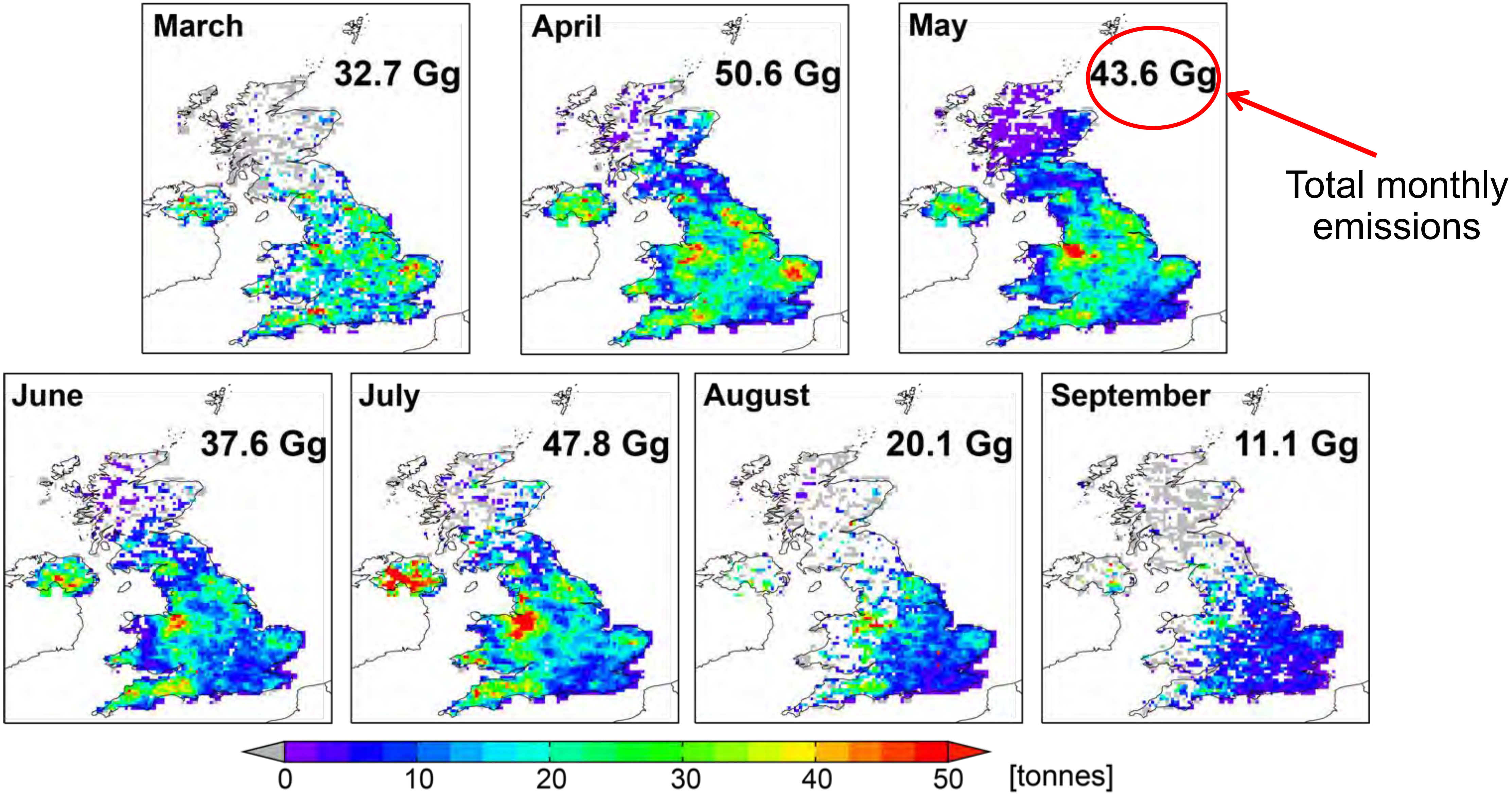
Top-down emissions estimates with satellite observations

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

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

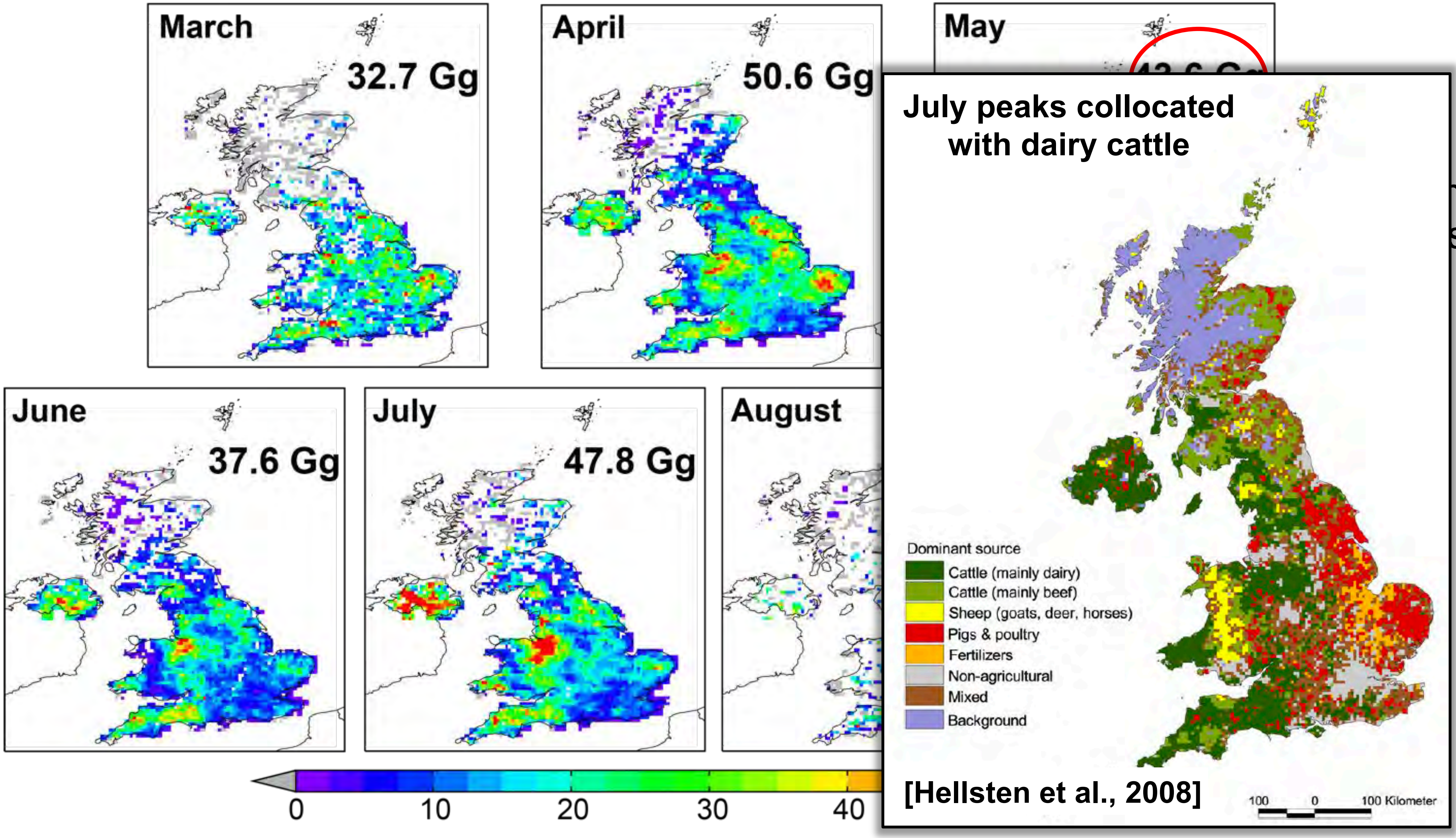


IASI-derived NH_3 emissions at 0.1° x 0.1°



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

IASI-derived NH_3 emissions at 0.1° x 0.1°



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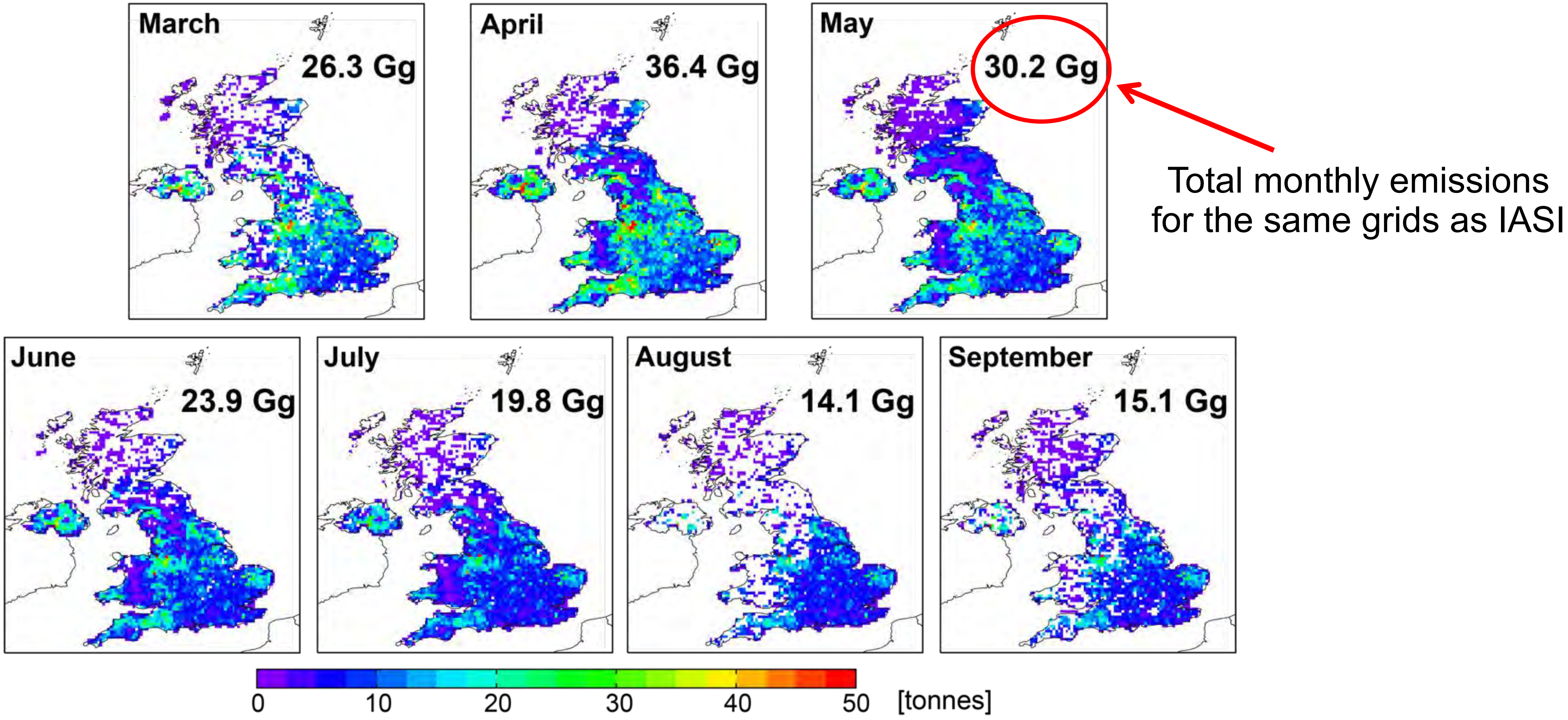
Sum of IASI-derived emissions for retained grids: **243.5 Gg**

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** (> Gothenburg protocol 297 Gg ceiling)

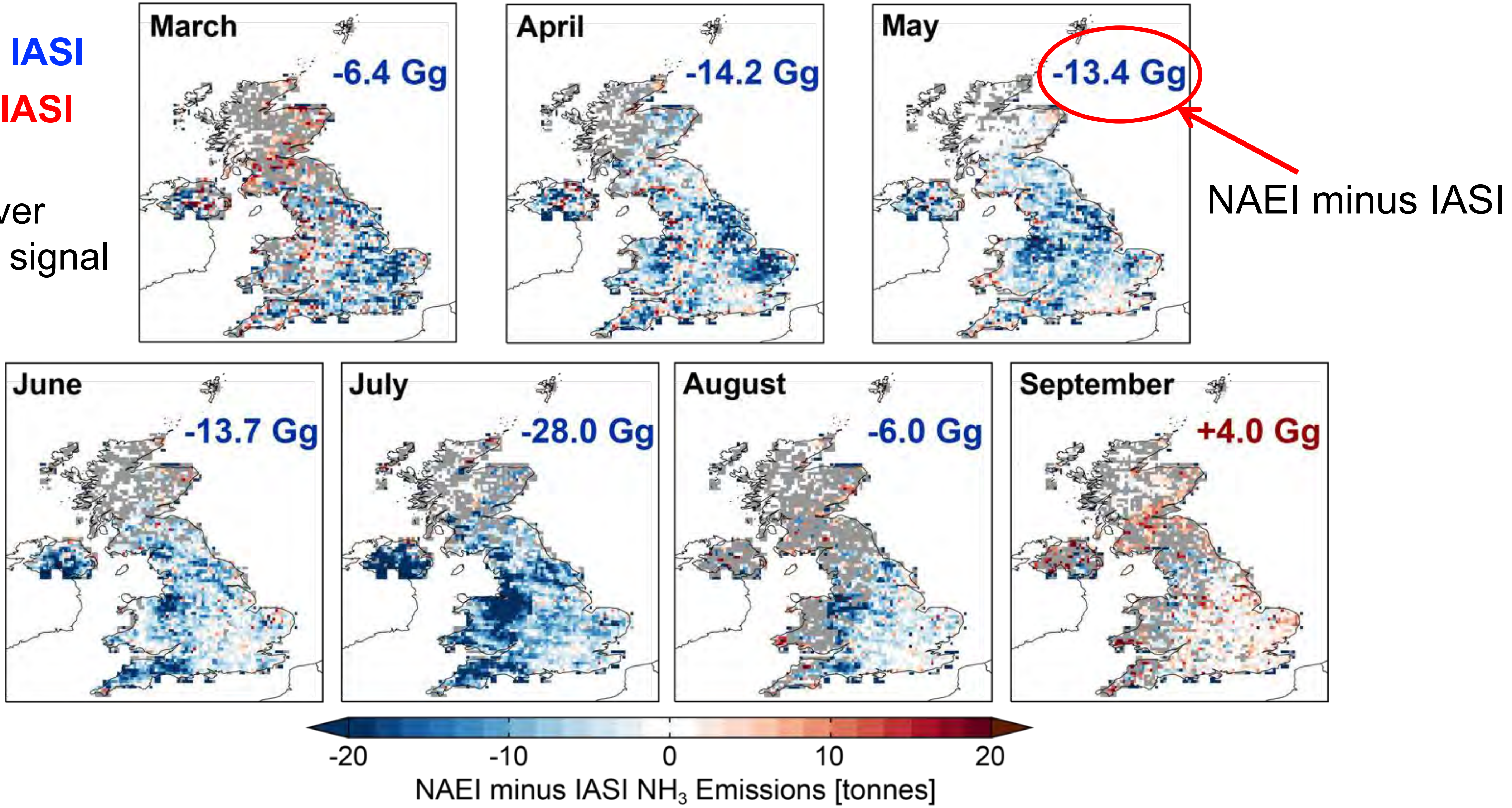
Differences in spatial distribution of IASI and NAEI

IASI-derived emissions minus NAEI-GEOS-Chem emissions

Blue: NAEI < IASI

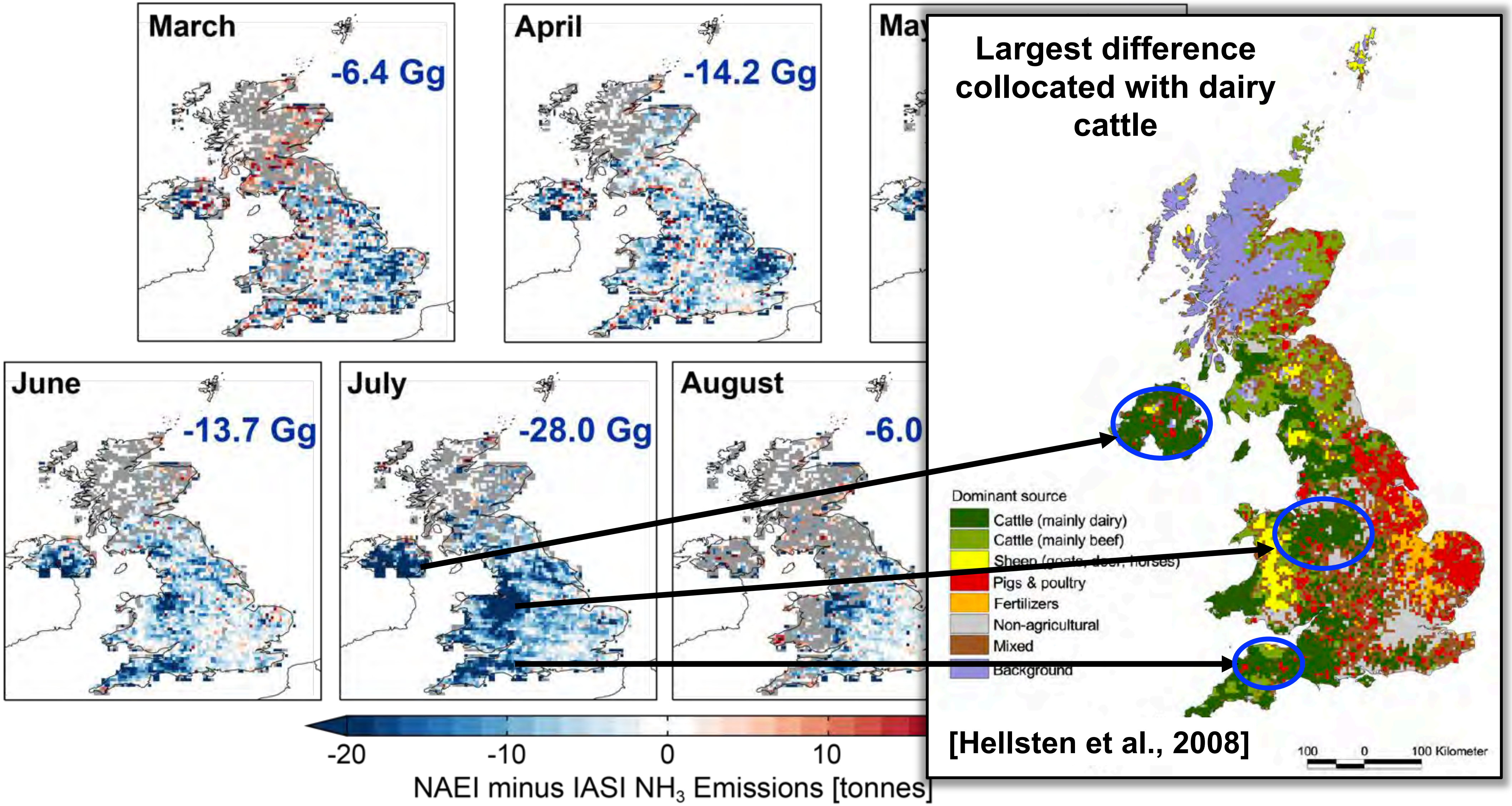
Red: NAEI > IASI

Red mostly over grids with low signal



Differences in spatial distribution of IASI and NAEI

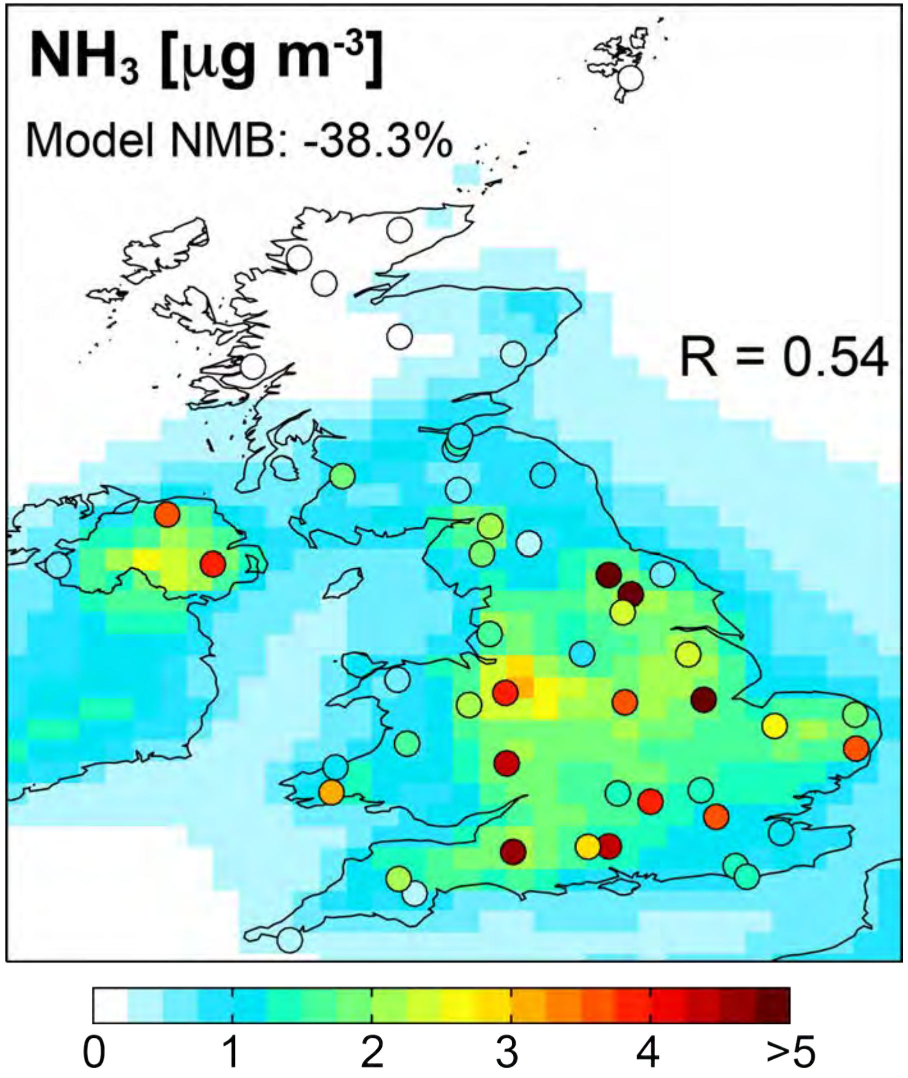
IASI-derived emissions minus NAEI-GEOS-Chem emissions



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

Assessment of NAEI NH_3 compared to surface observations

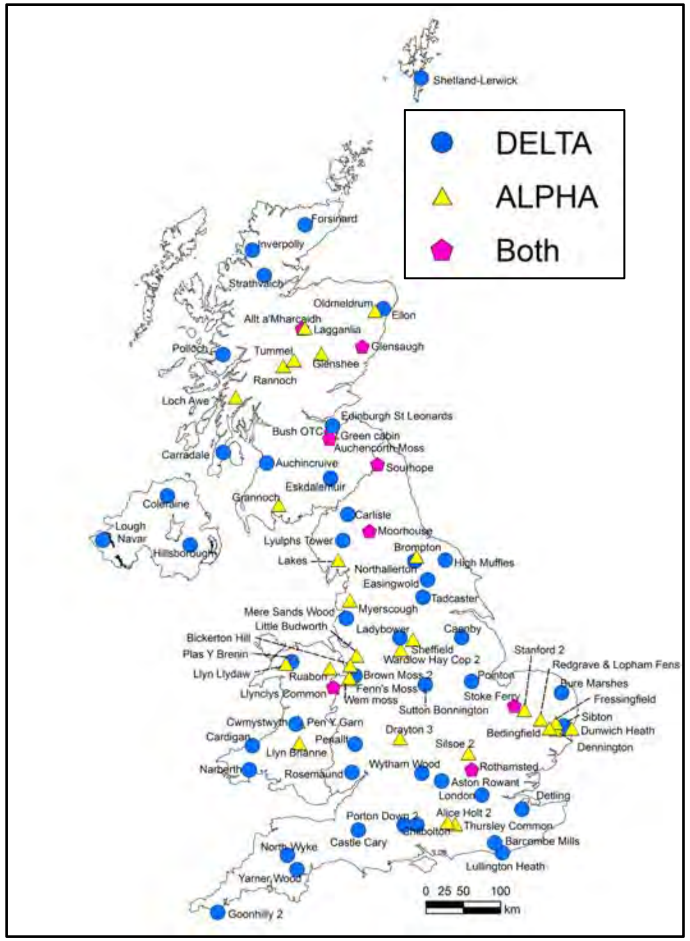
Network (points) and model (background) surface NH_3 in Mar-Sep



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

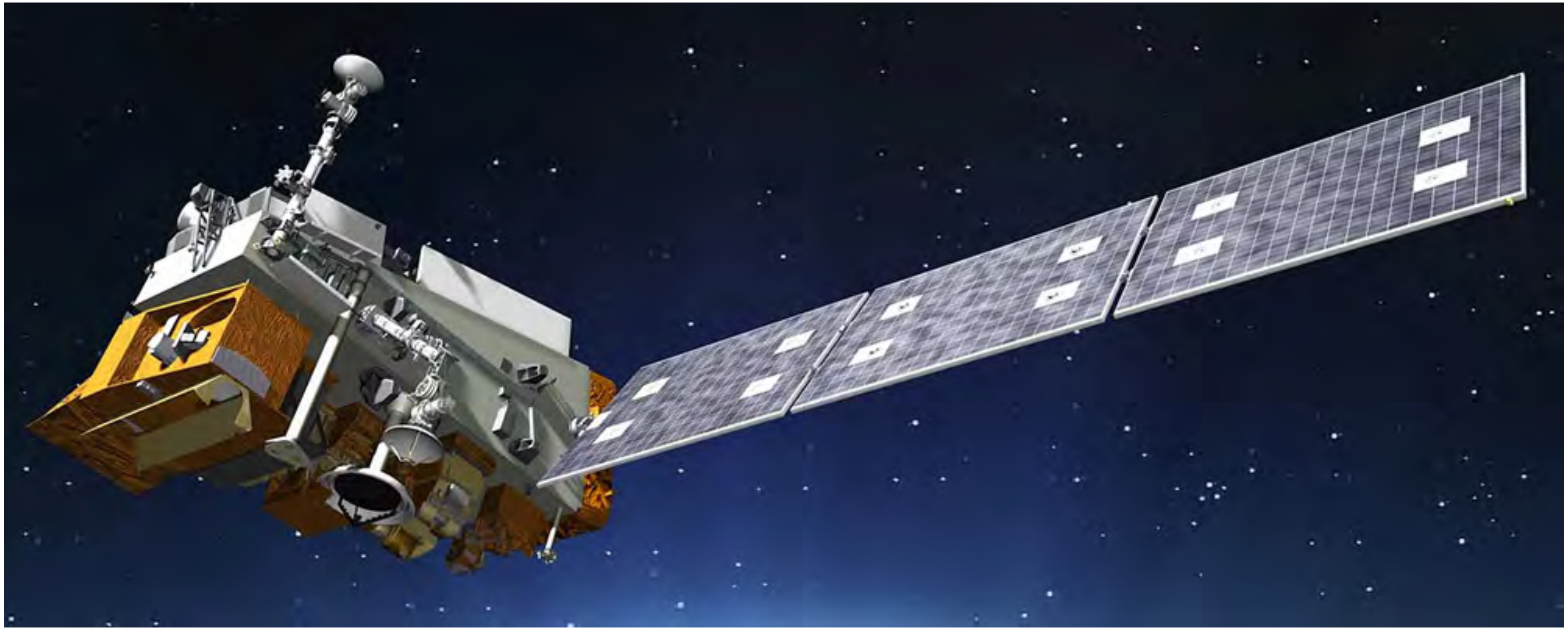
Cross-Track Infrared Sounder (CrIS) Instrument

<u>Overpass:</u>
13:30 local solar time

<u>Spatial resolution:</u>
14 km at nadir

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

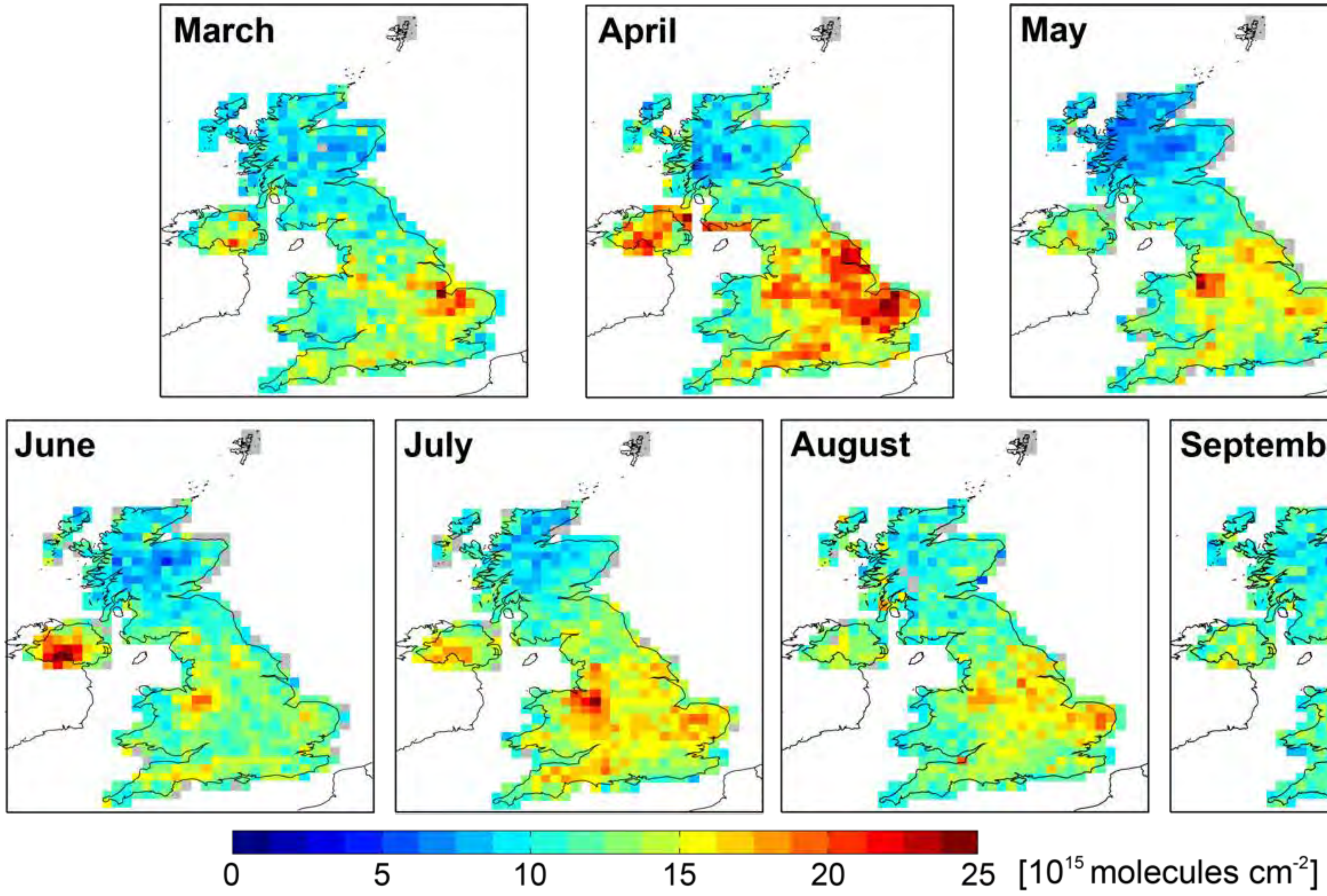
<u>Launch date:</u>
October 2011

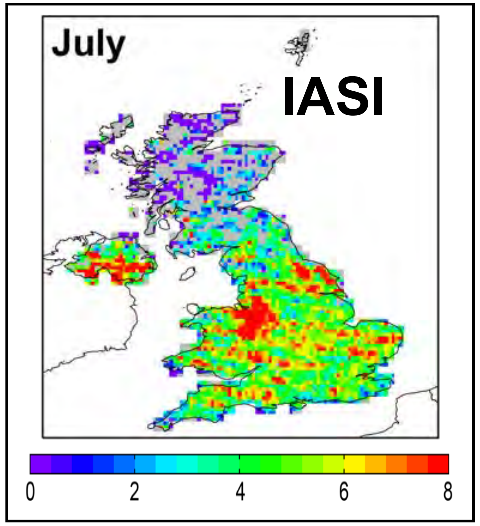


CrIS instrument is 4-times more sensitive than IASI instrument

Multiyear (2013-2018) monthly mean CrIS NH_3

CrIS NH_3 retrieved using optimal estimation (constrained with a prior)





IASI retrieval is 10-times more sensitive to NH_3 than CrIS

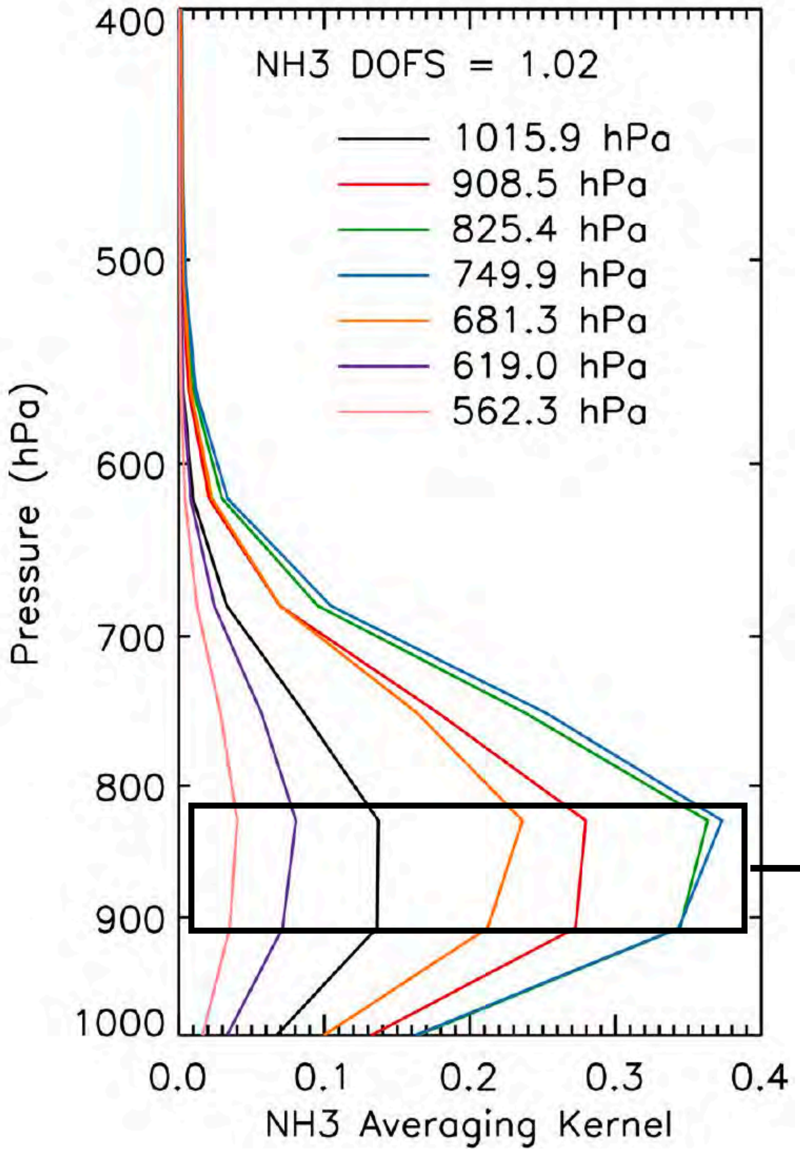
Data are at the same spatial resolution as GEOS-Chem (0.25° x 0.3125°)

Spatial correlation with IASI is R = 0.45 (Sep) and R = 0.54-0.75 (Mar-Aug)

CrIS data providers:
K. Cady-Pereira, M. Shephard

Comparison of CrIS and GEOS-Chem column densities

CrIS Averaging Kernels



[Shephard & Cady-Pereira, 2015]

Peak sensitivity

Apply instrument vertical sensitivity to GEOS-Chem for consistent comparison:

$$\boxed{x_{\text{smoothed}} = \exp\big((\mathbf{I} - \mathbf{A})\ln x_a + \mathbf{A}\ln x_{\text{resolved}}\big)}$$

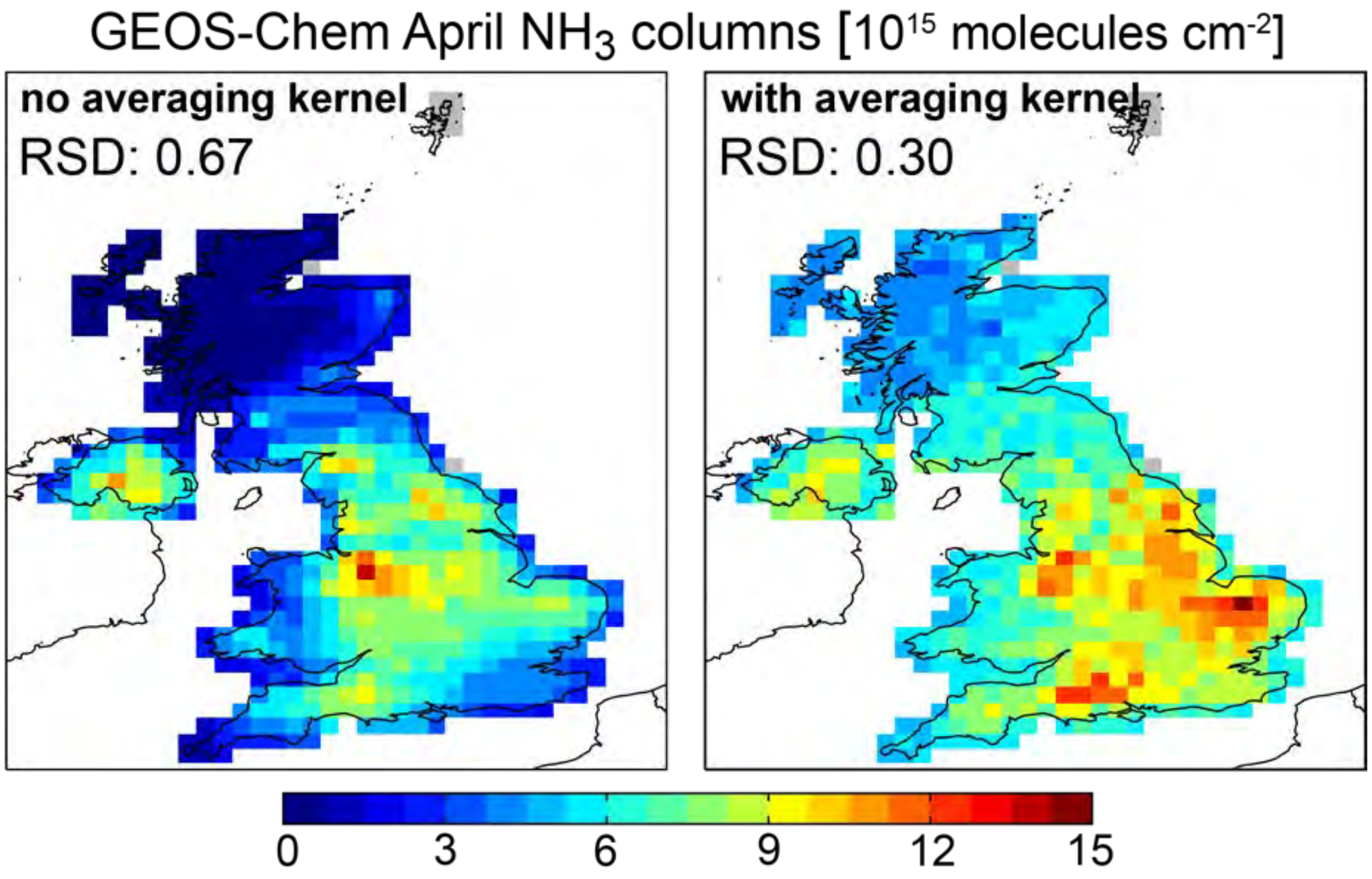
$\mathbf{A}$: averaging kernel matrix

x_{a}: CrIS a priori NH_3 profile

x_{resolved}: GEOS-Chem vertical profile

Averaging maximum boundary layer altitude in London is 1.5 km

Application of CrIS averaging kernels to GEOS-Chem



Substantial smoothing of spatial variability

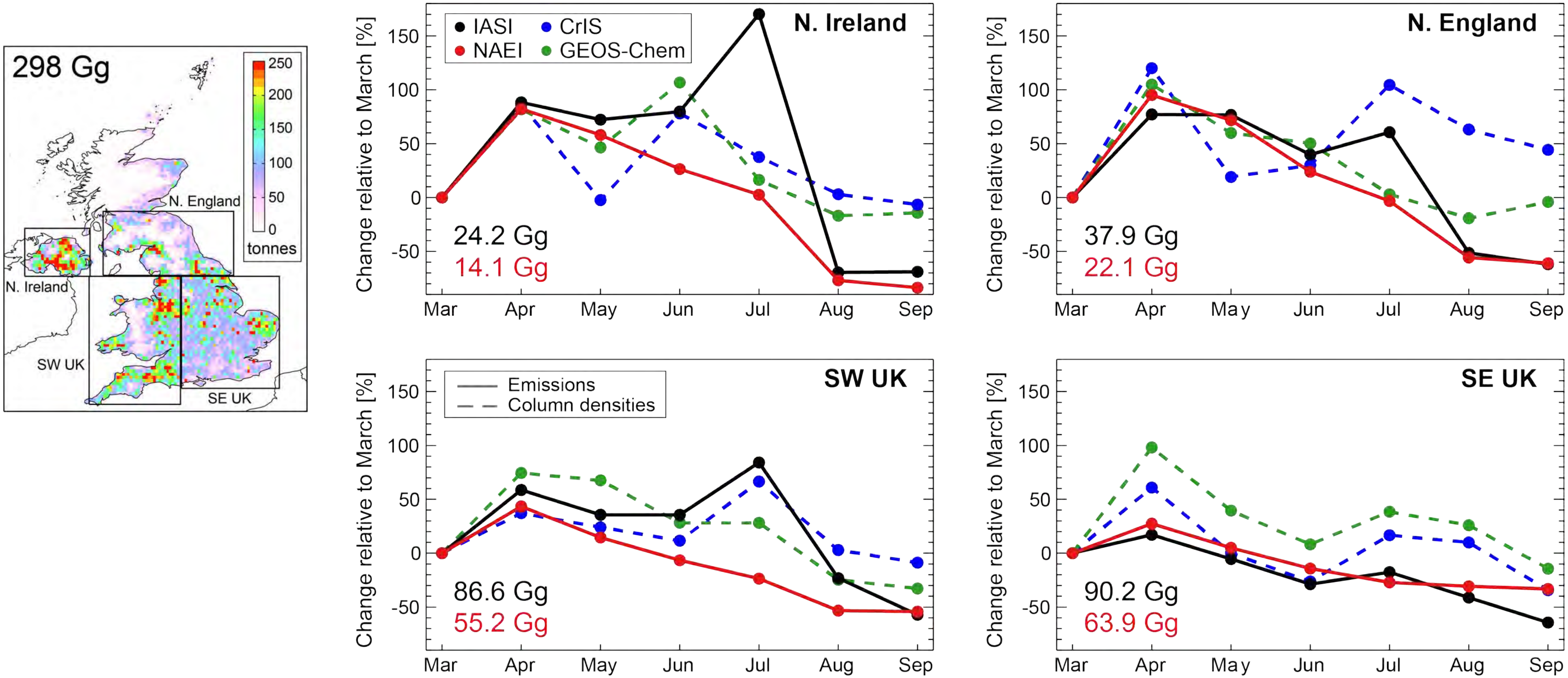
Spatial correlation between GEOS-Chem and CrIS is reasonable in Sep (R = 0.5) and strong in Mar-Aug (R = 0.7-0.9).

Substantial smoothing of spatial variability (halves relative standard deviation or RSD)

Model is biased low by ~50%, but does not change much (~45%) with a doubling in NH_3 emissions

Seasonality in NH_3 in the UK

Month-month variability in NH_3 plotted below relative to March

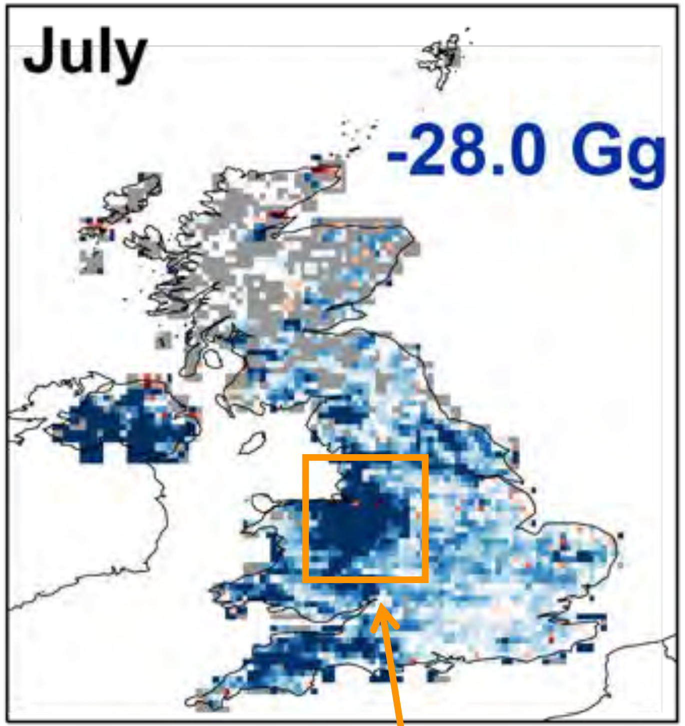


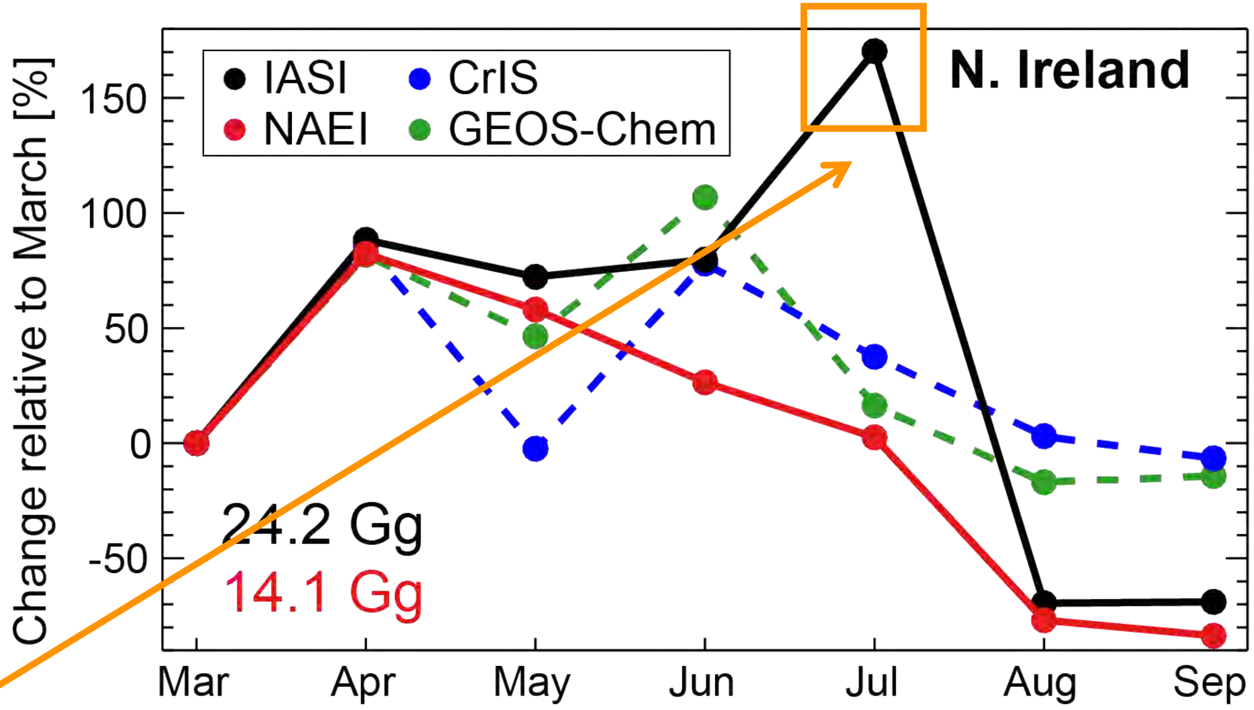
Consistent spring April peak (fertilizer use), inconsistent second summer peak

Large IASI peak in July in N Ireland may be erroneous.

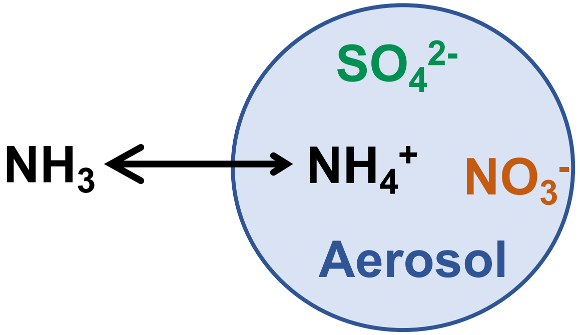
IASI clear-sky bias in July

Month-month variability in NH_3 plotted below relative to March





IASI samples cloud-free scenes → warmer atmosphere → semi-volatile NH_3 favours the gas phase



Clear-sky boundary-layer temperature 5.6ºC more than it is for all-sky in July. It's 0.8-4ºC more in the other months.

If exclude July, the NAEI emissions are 34% less than the IASI-derived emissions

Concluding Remarks

- NAEI overestimates SO_2 emissions by more than a factor of 2 likely due to errors in biomass burning emissions.
- NAEI NH_3 emissions are underestimated by 25-32% compared to IASI-derived emissions and the relative error is 8-18% compared to 31% for NAEI.
- UK may be out of compliance with the Gothenburg protocol emissions ceiling.
- Low bias in NAEI emissions is corroborated by surface observations.
- CrIS has limited sensitivity to NH_3 emissions, but both IASI and CrIS provide constraints on seasonality.
- Consistent peak in NH_3 emissions in April in bottom-up and top-down estimates due to fertilizer application, but summer peak in satellite-derived emissions may by biased by sampling cloud-free conditions.
- Low bias in NAEI NH_3 emissions affects ability to model $PM_{2.5}$.

Acknowledgements

Defra for funding

Data analysis by **Alok Pandey** and **Karn Vohra**

Martin Van Damme, **Lieven Clarisse**, and **Pierre-F. Coheur** for IASI NH_3

Karen Cady-Perreira and **Mark Shephard** for CrIS NH_3

Lei Zhu for oversampling code

UKEAP and MARGA teams for maintaining very precious surface monitoring networks

CEH, **Tom Misselbrook** for helpful discussions on UK NH_3 sources and temporal variability