Trends and emissions of ammonia and its influence on air quality in the UK

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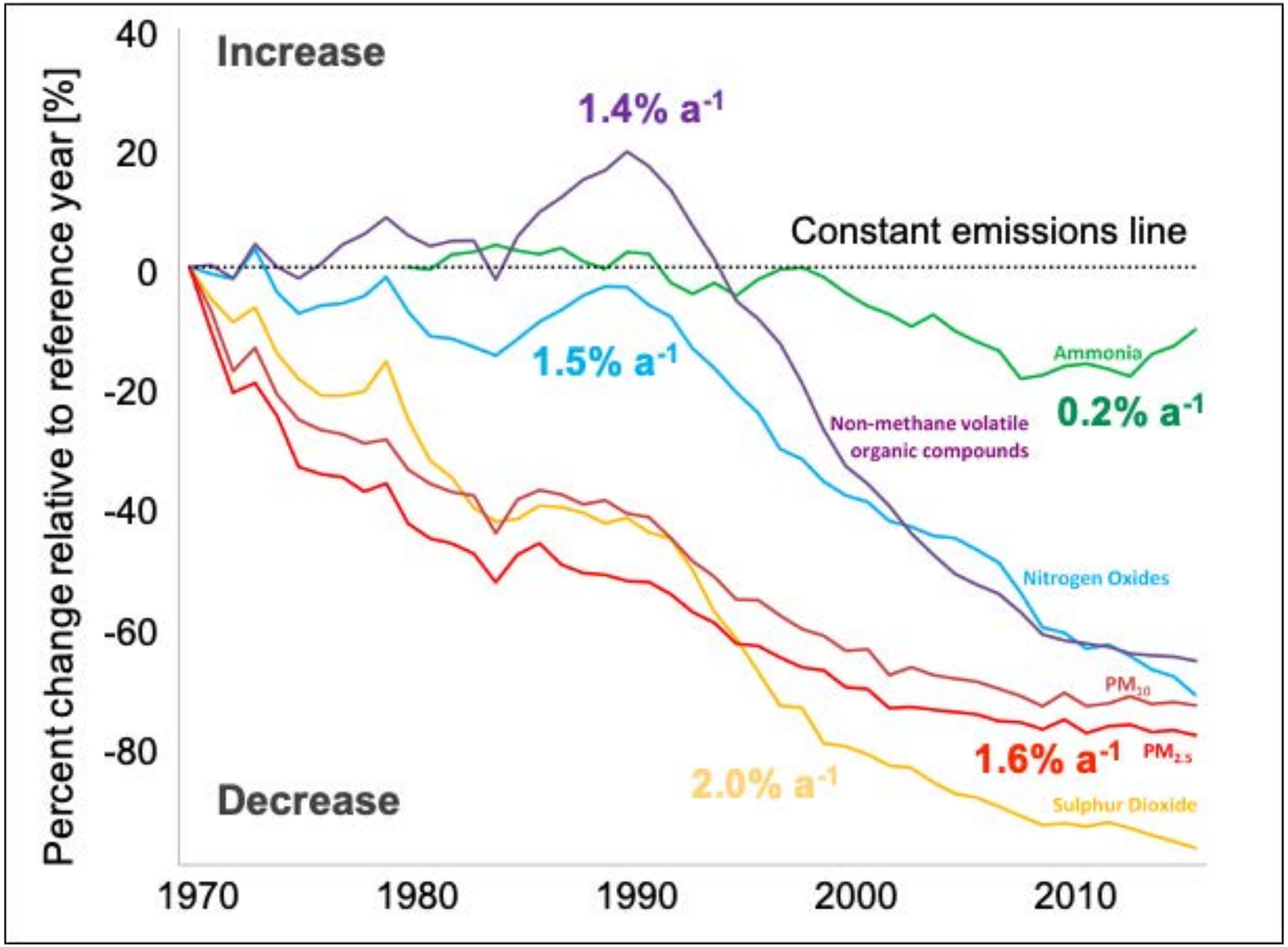
Ammonia emissions in the UK: the bottom-up perspective

Temporal (Time) Variability in Emissions



Green: ammonia

Purple: non-methane volatile organic compounds

Blue: nitrogen oxides

Orange: primary PM_{10}

Red: primary $PM_{2.5}$

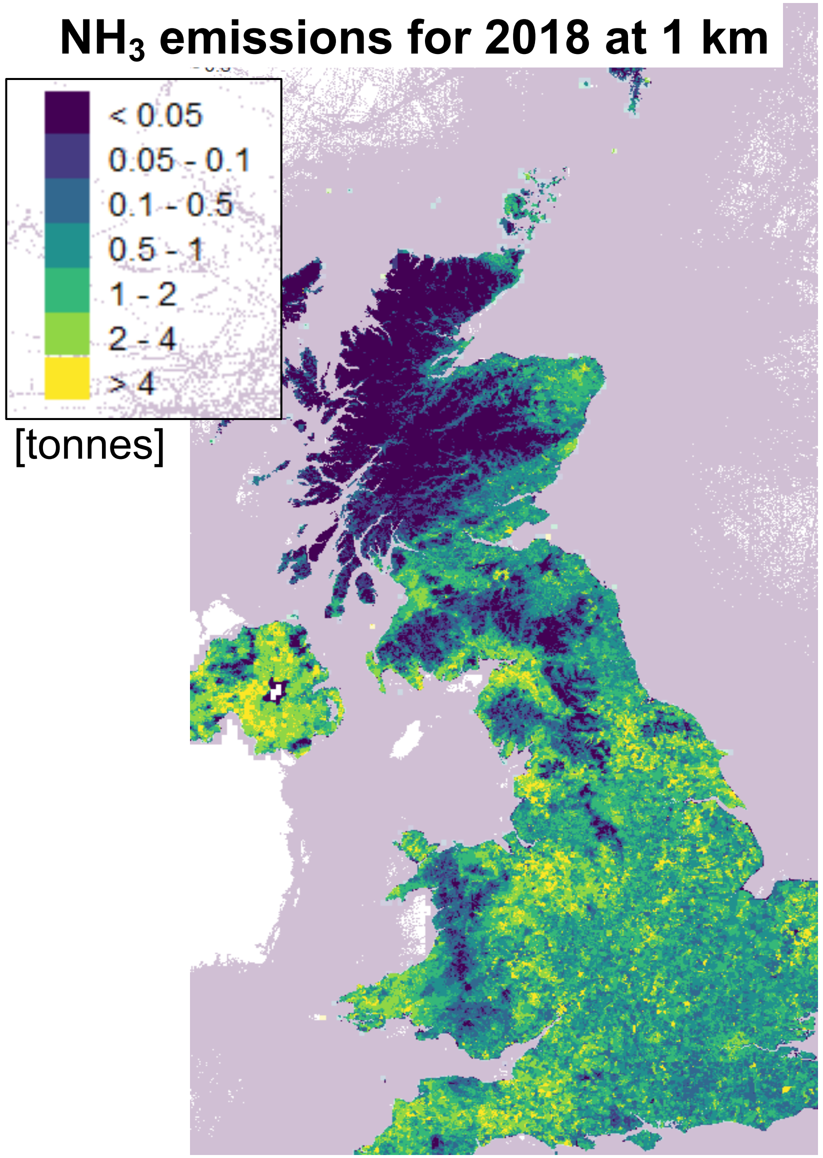
Yellow: sulfur dioxide

[*Adapted from* Defra, 2018]

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

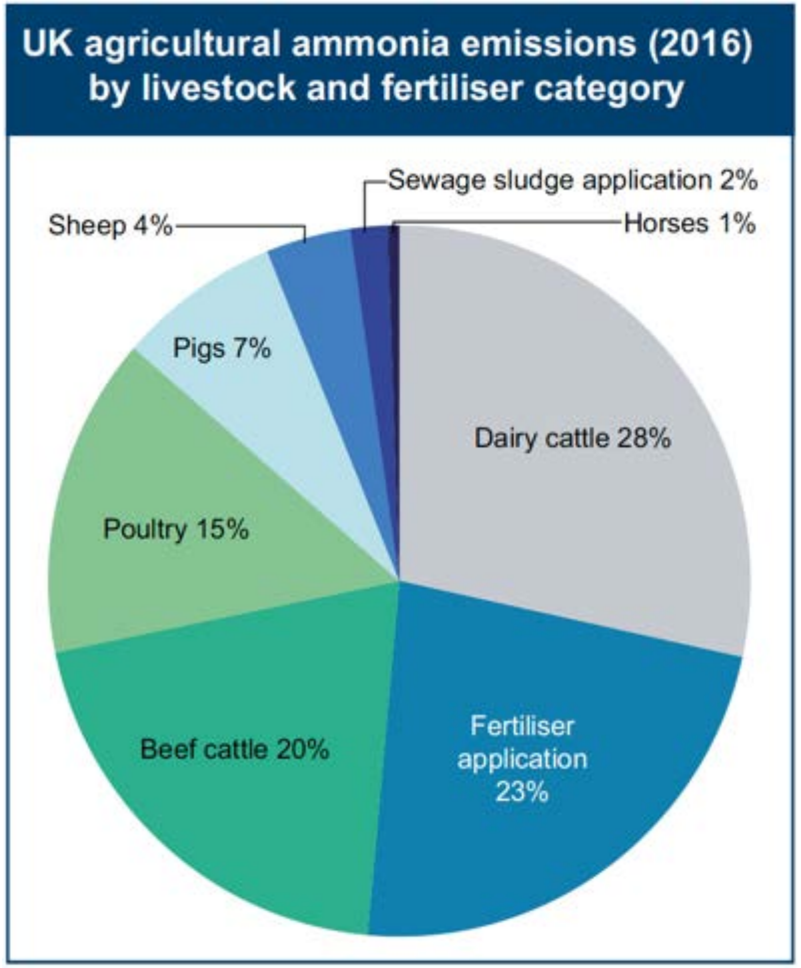
Ammonia emissions in the UK: the bottom-up perspective

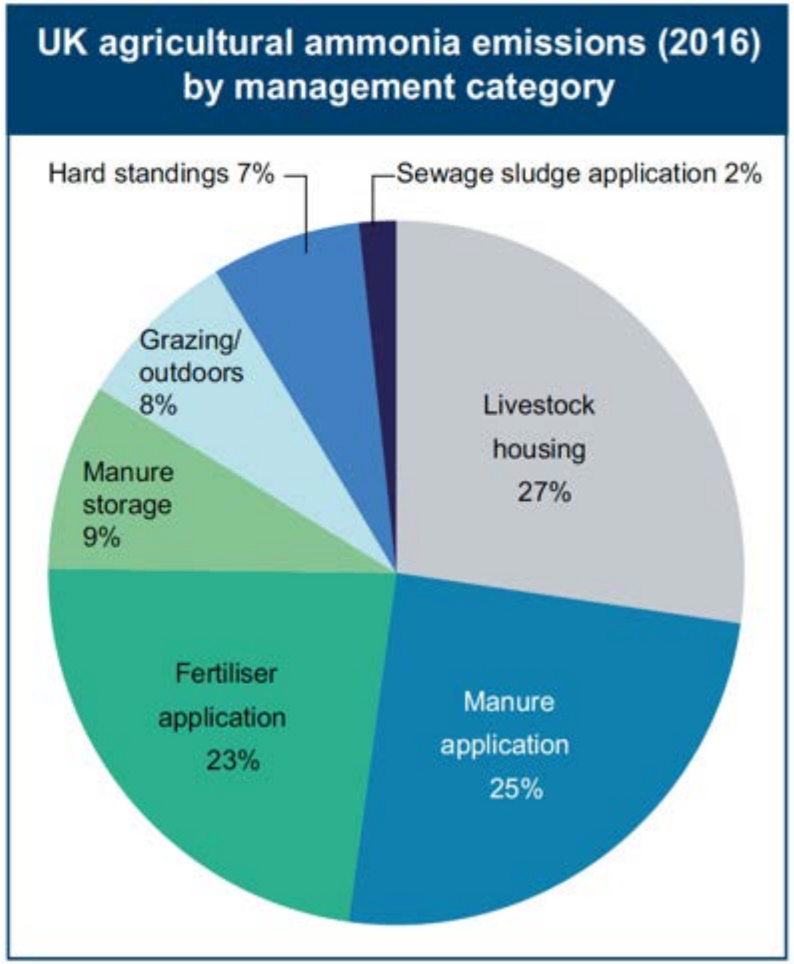
Emissions Spatial Variability



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

Contributions of activities to ammonia emissions



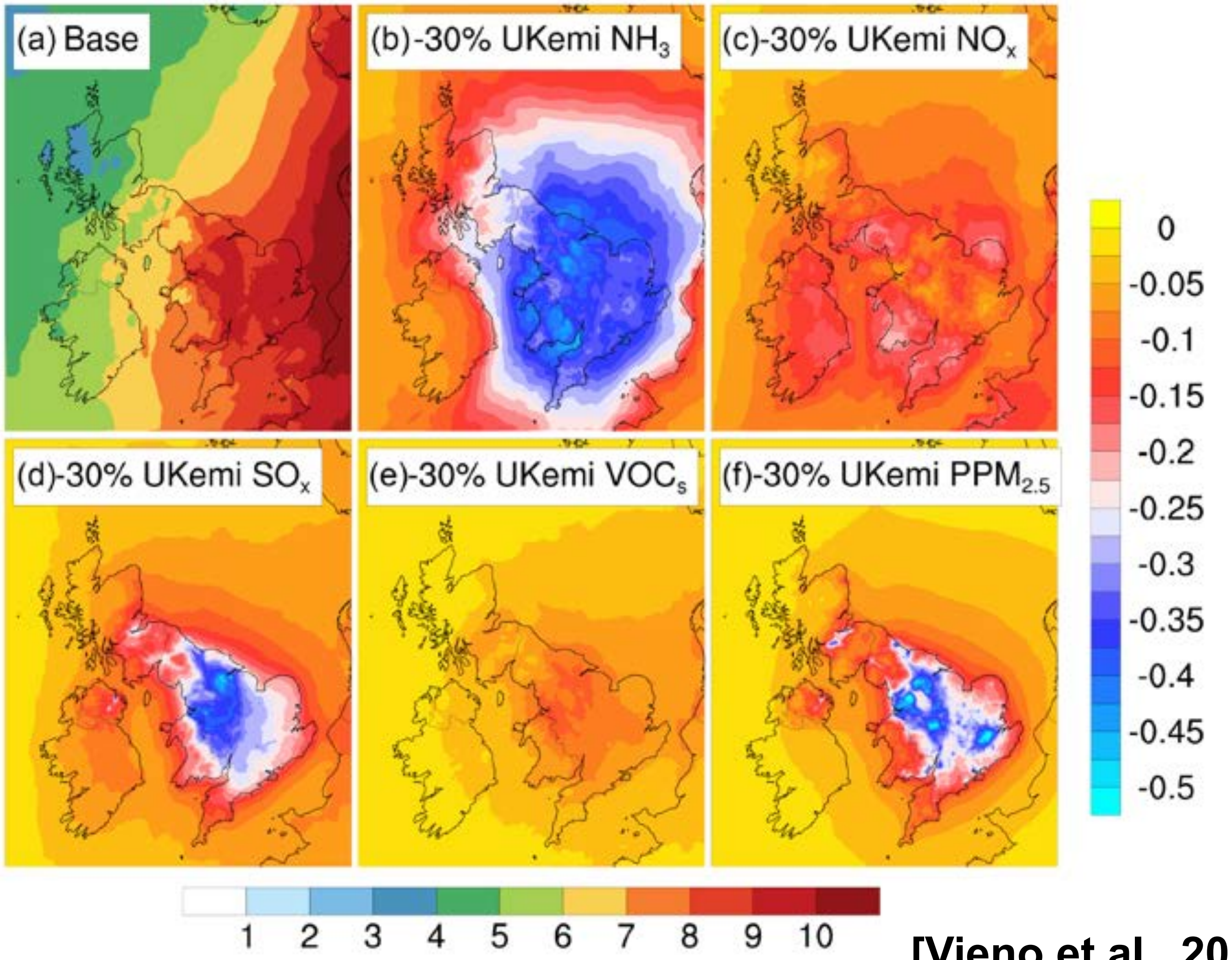


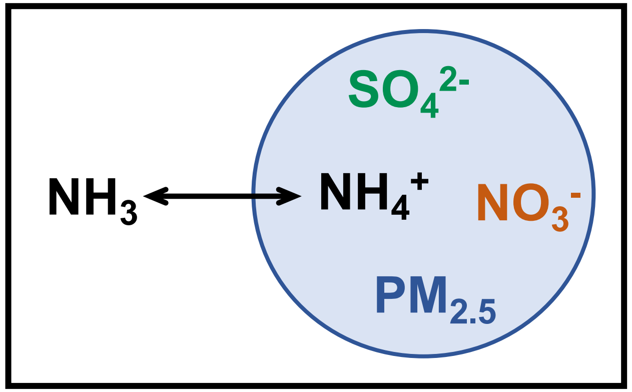
[UK Clean Air Strategy, 2019]

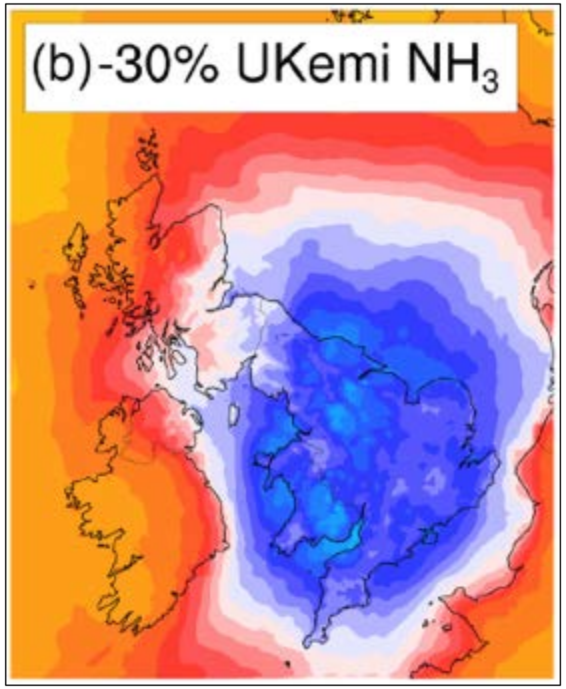
Beef, dairy, and fertilizer use dominate

Ammonia impact on air pollutants hazardous to health

Effect of emission controls on $PM_{2.5}$





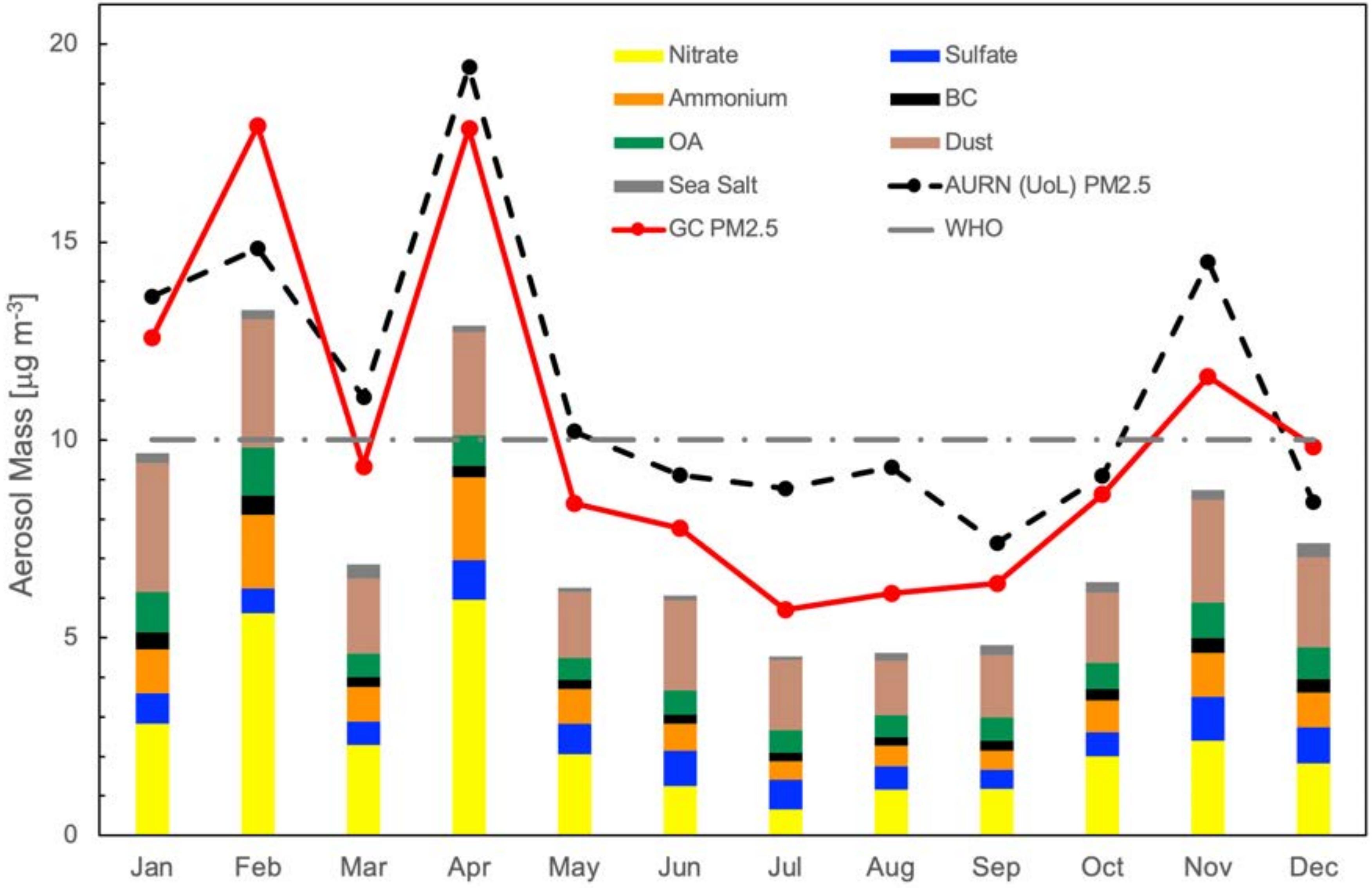


[Vieno et al., 2016]

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

Ammonia is a large contributor to $PM_{2.5}$ in an East Midlands City

Modelled and observed $PM_{2.5}$ mass in Leicester in 2019



Defra-funded project with Leicester City Council

Model similar to AURN $PM_{2.5}$, except in summer. NH_3 underestimate?

Ammonium (orange) large component of $PM_{2.5}$ in most months

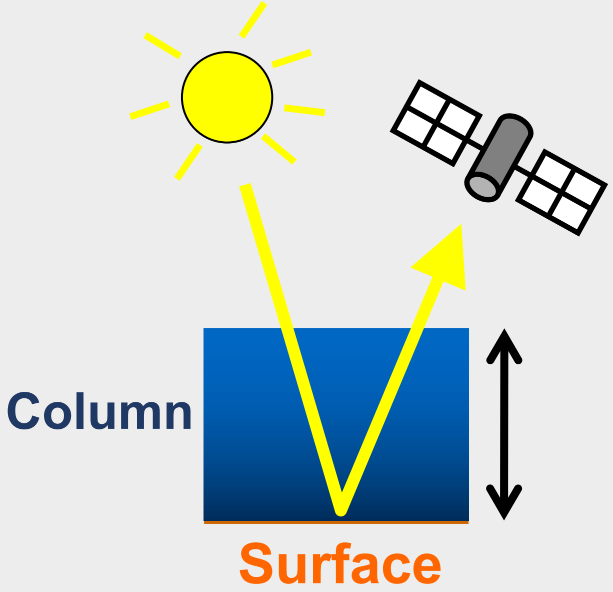
Winter: excess NH_3, cold temperatures favor promote nitrate and ammonium formation

Top-down emissions estimated with satellite observations

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

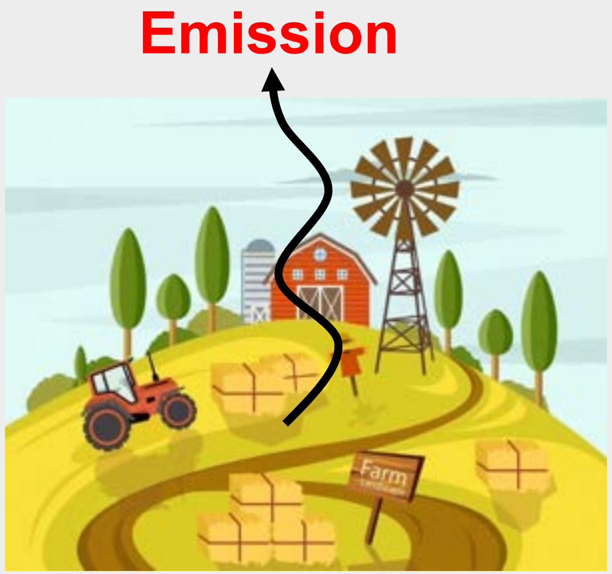
ABUNDANCES → **Conversion Factor** → **EMISSIONS**

Satellite column densities



Model Concentration-to-Emission Ratio

Satellite-derived Surface Emissions

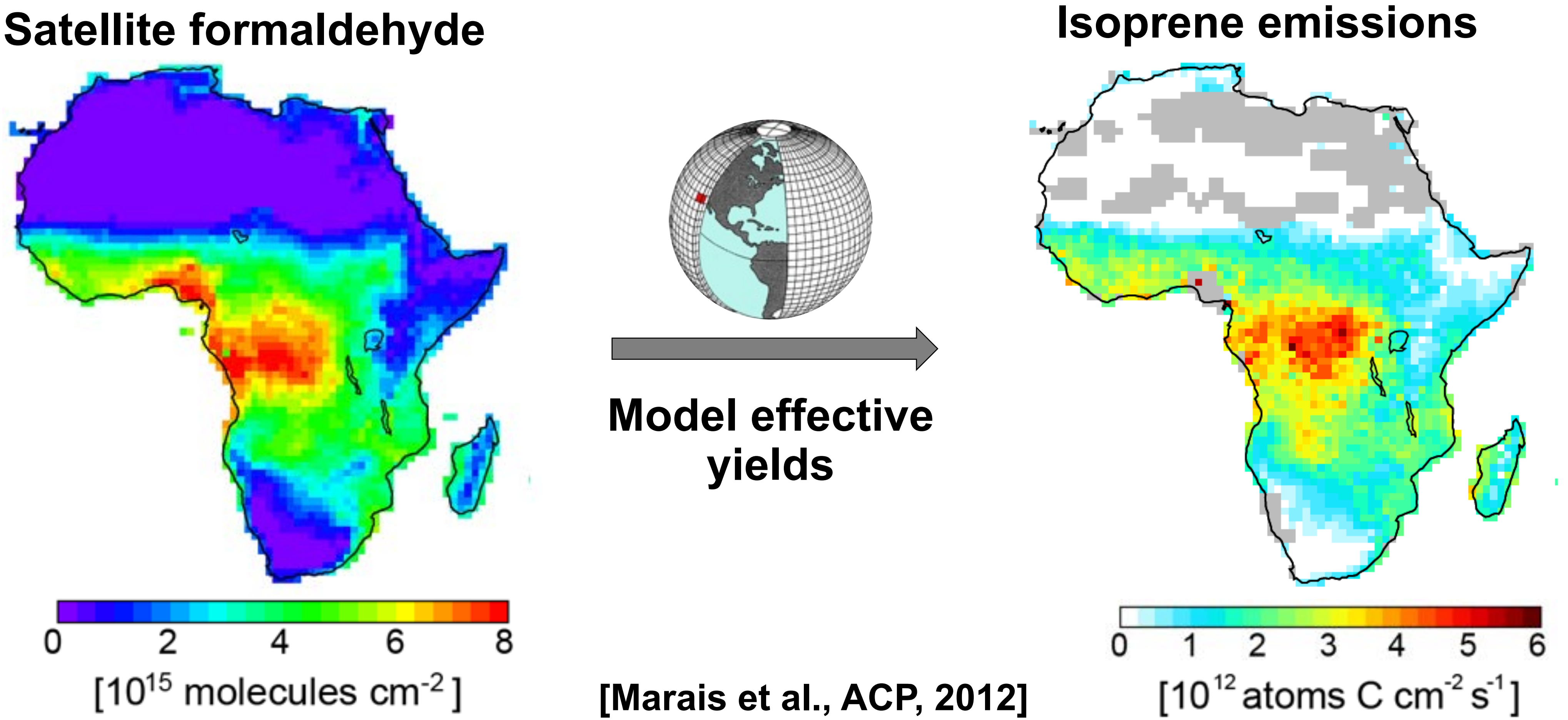


Widely used to estimate emissions and surface concentrations

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

Concentrations → emissions: formaldehyde → isoprene, NO_2 → NO_x

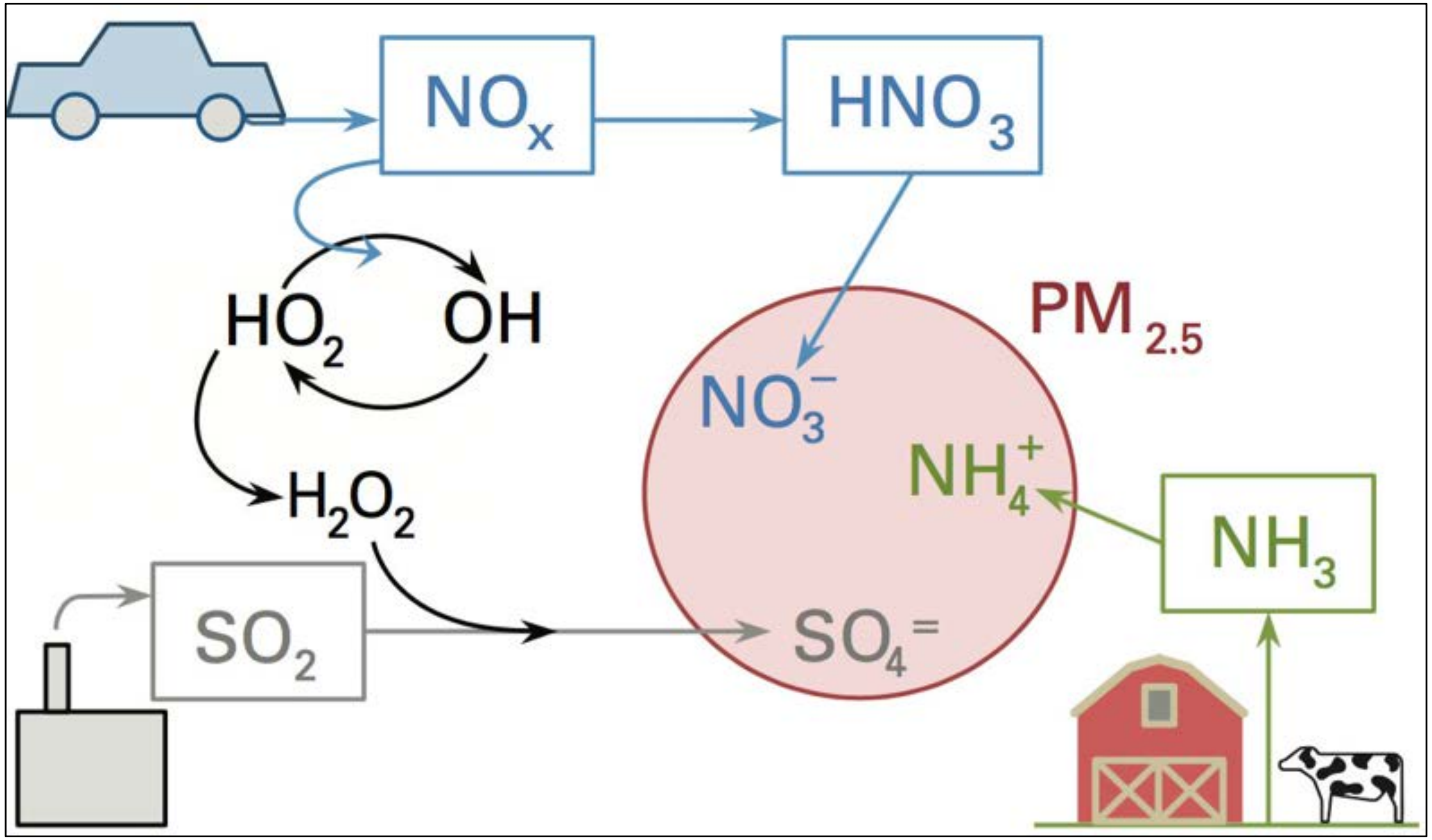
Column → surface: formaldehyde → formaldehyde, NO_2 → NO_2, AOD → $PM_{2.5}$



[Marais et al., ACP, 2012]

Ammonia abundance depends on numerous factors

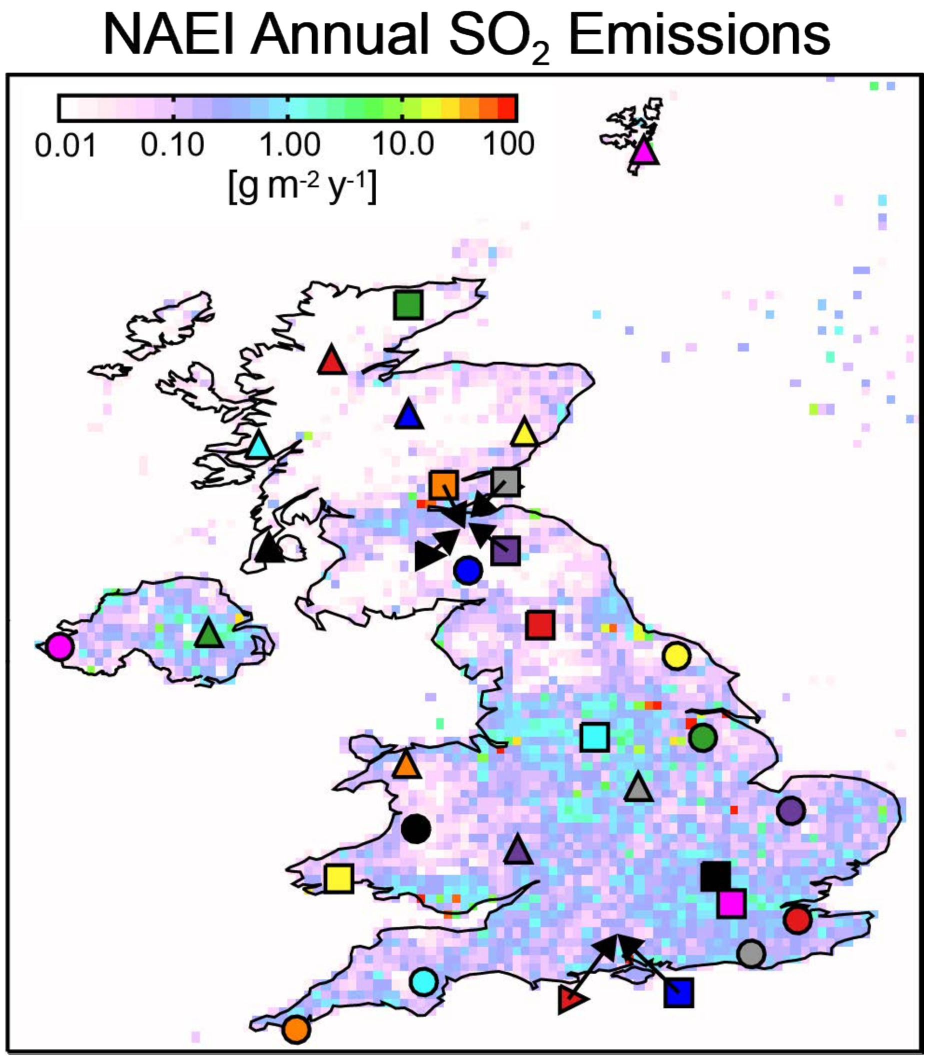
Ammonia buffers acidic aerosols formed when SO_2 oxidizes to form sulfate (SO_4^{2-})



[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

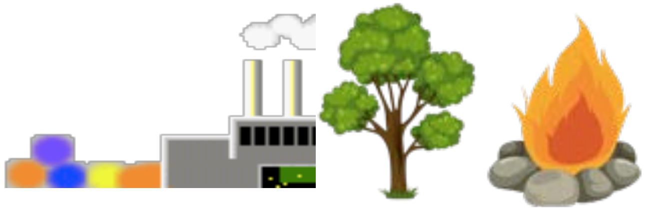
UKEAP:
~30 sites
offline denuder measurements
0.05 $\mu g\ m^{-3}$ detection limit

MARGA:
2 sites
semi-continuous denuder measurements
0.04 $\mu g\ m^{-3}$ detection limit

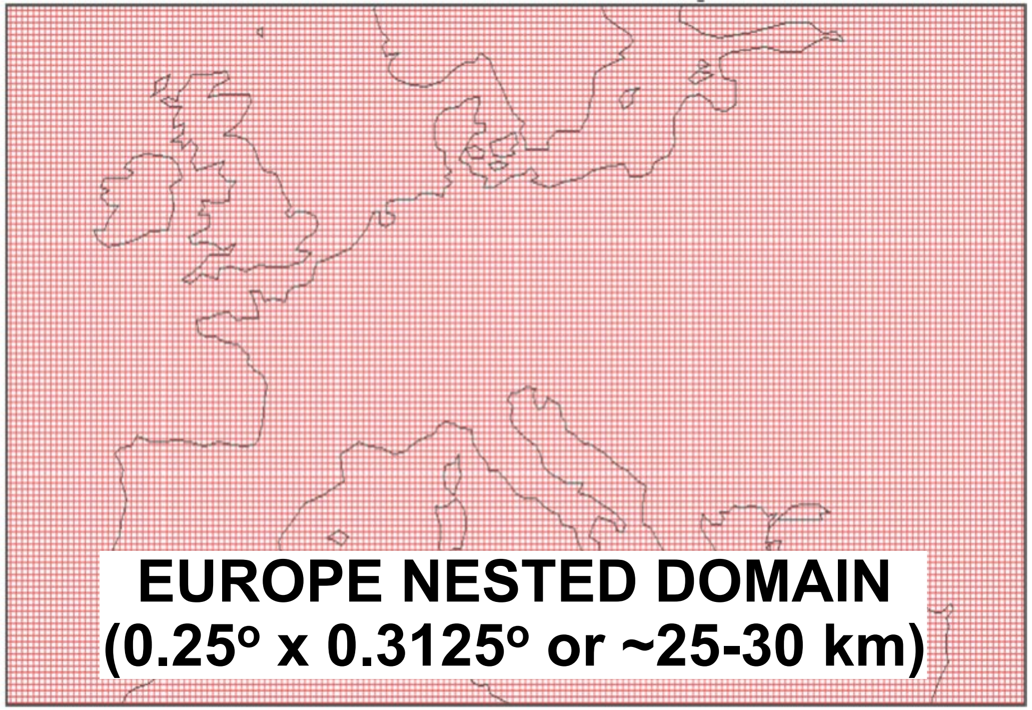
Surface SO_2 concentrations calculated with GEOS-Chem

3D Atmospheric Chemistry Transport Model

Emissions (natural/human)



UK NAEI emissions
(with temporal information)



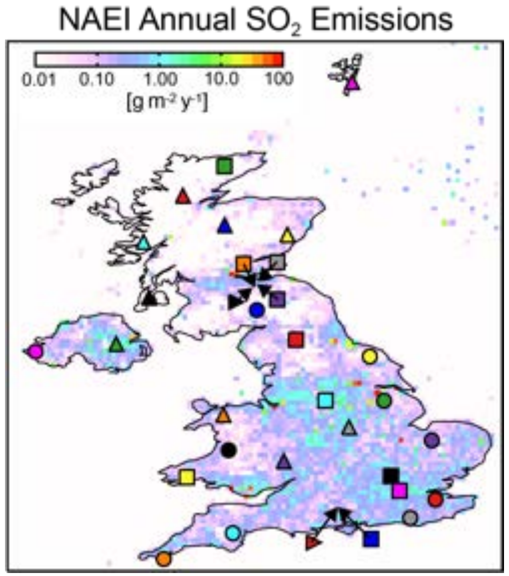
Offline assimilated meteorology

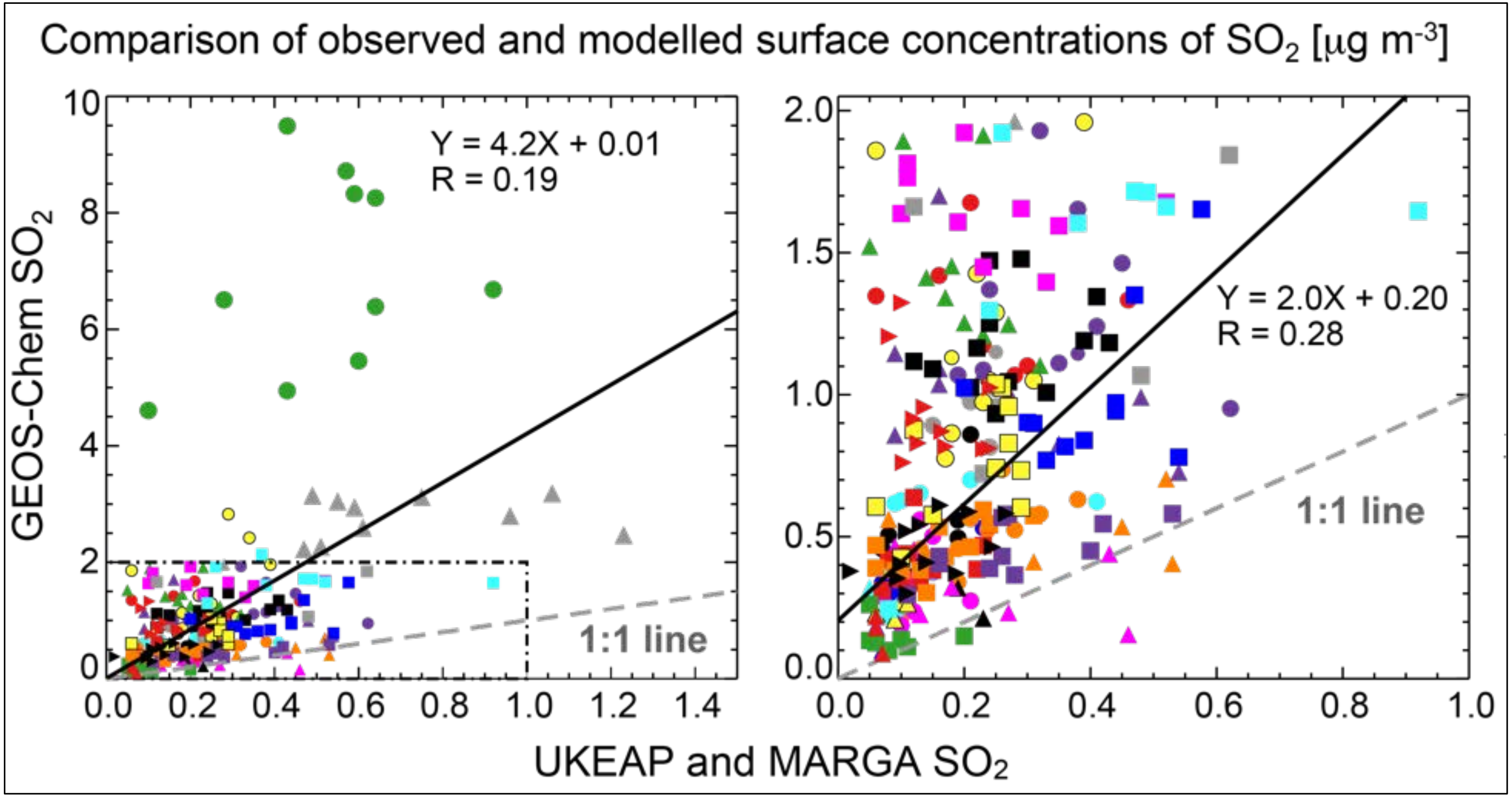
NASA GEOS-FP for 2016

Gas phase and heterogeneous chemistry
Transport
Dry/wet deposition

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

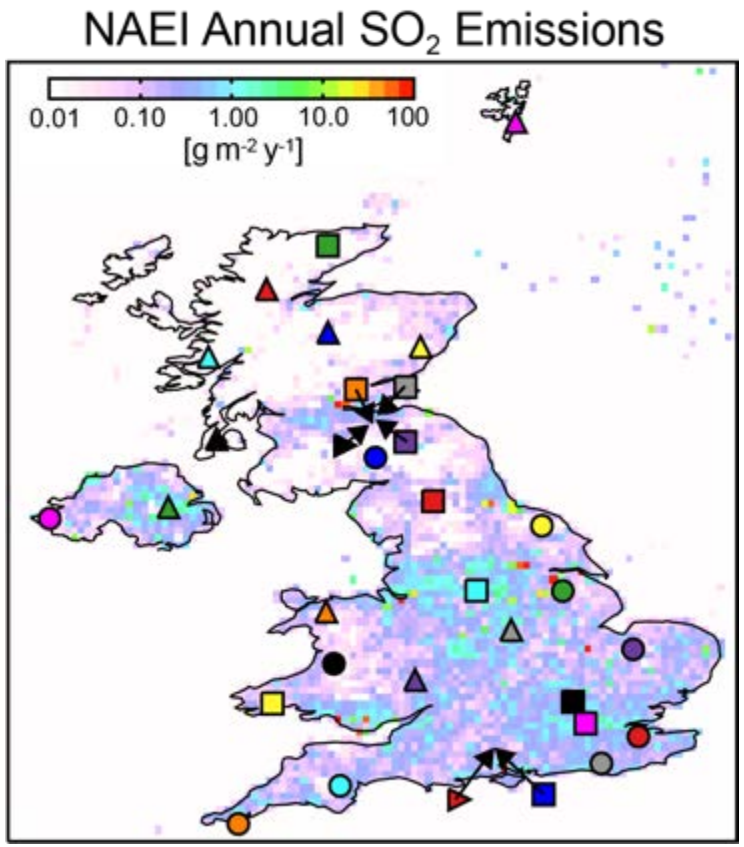
Modelled (GEOS-Chem-NAEI) versus observed SO_2

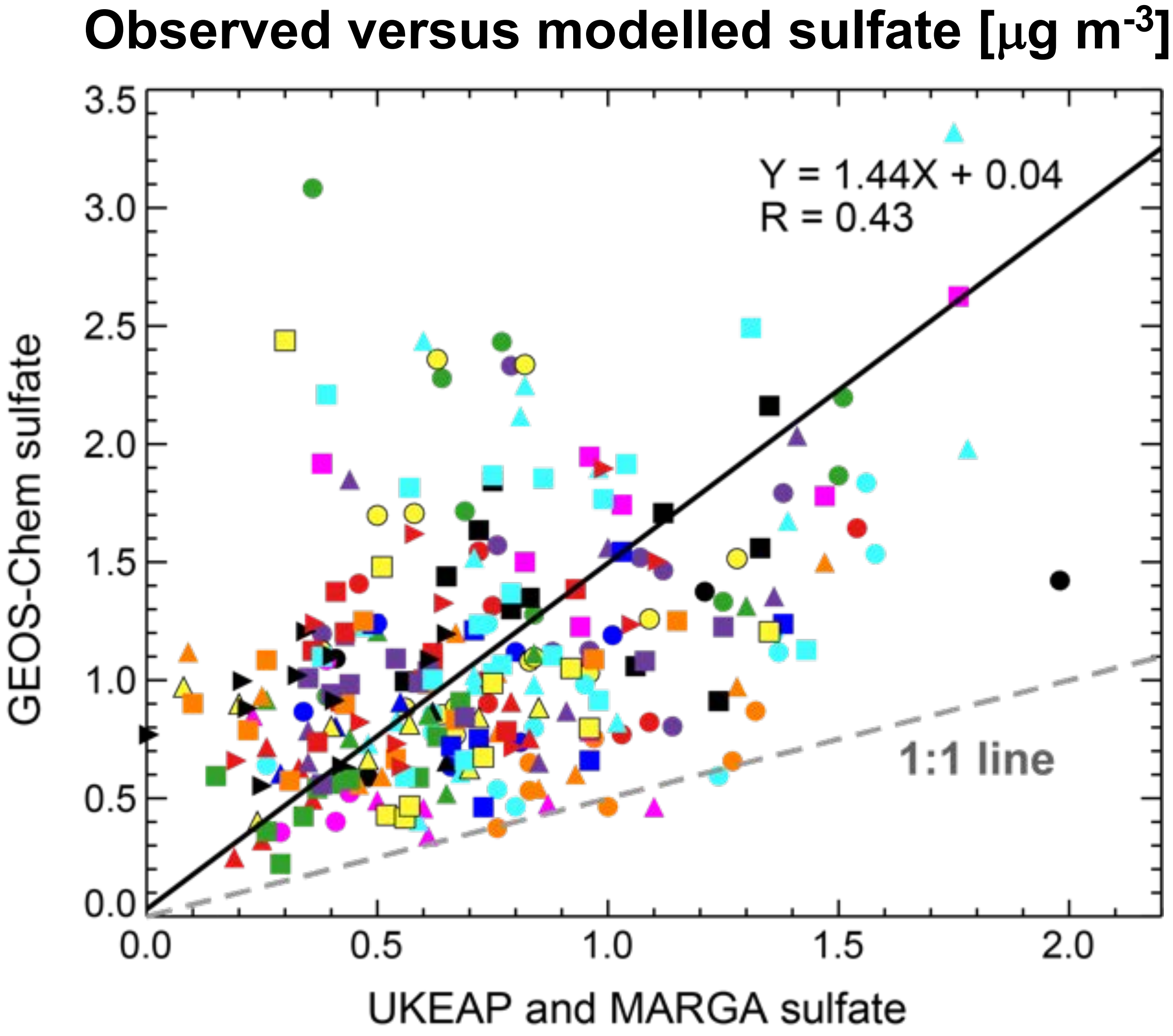




Comparison supports large overestimate in NAEI SO_2 emissions (in particular point sources)

Modelled (GEOS-Chem-NAEI) versus observed sulfate





Better spatial consistency for sulfate (R = 0.43). Model also overestimates sulfate (by 52%)

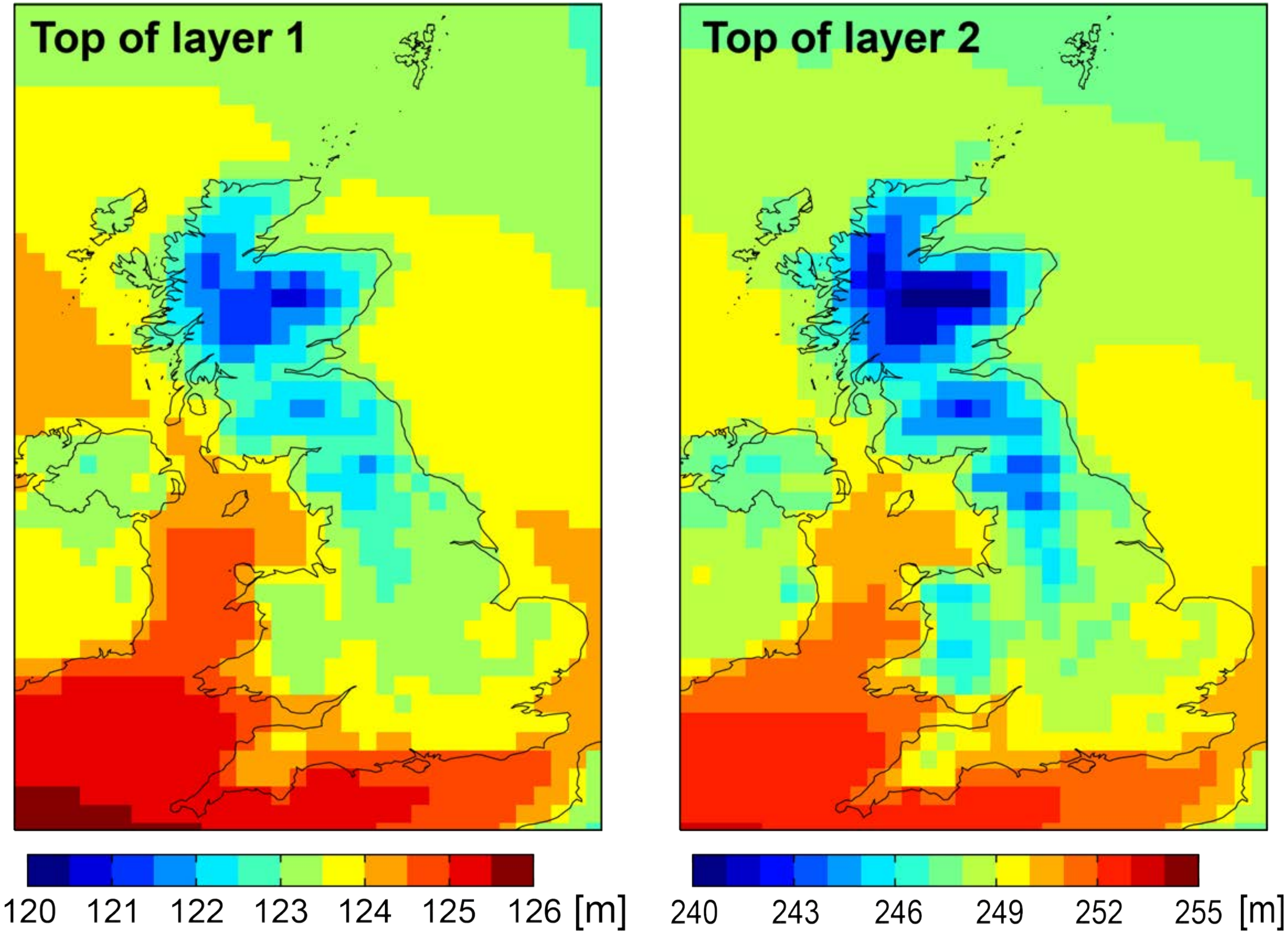
Vertical distribution of SO_2 point sources

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



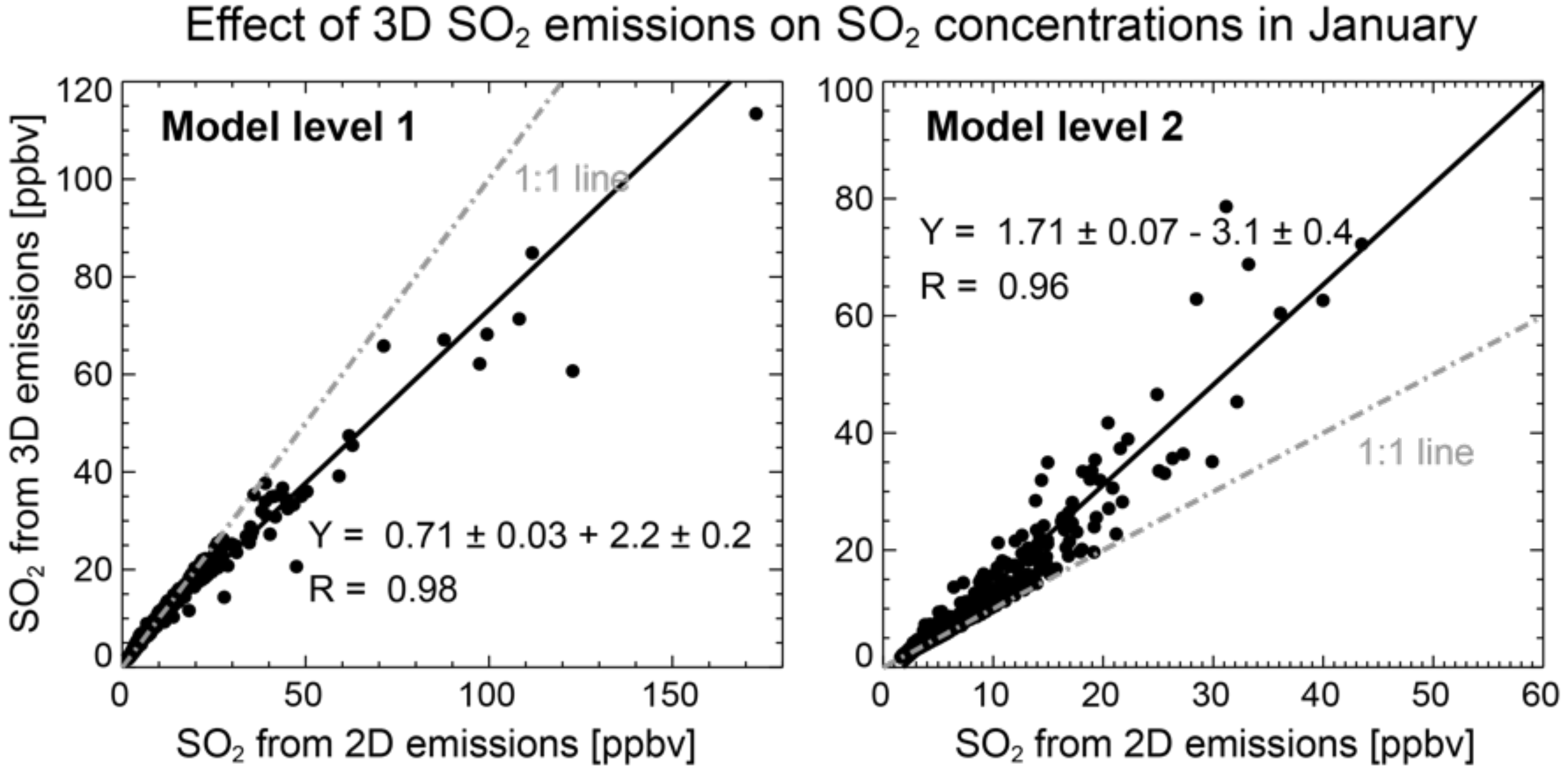
Vertical distribution of SO_2 point sources

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

2016 SO2 emissions:

All land-based: 164 Gg

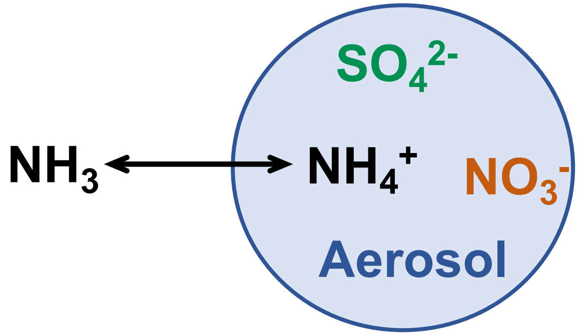
Point sources: 96 Gg



Extreme test leads to 30% decrease in surface SO_2, 70% increase in layer 2. Similar results for July.

Decrease annual NAEI SO_2 emissions 161 Gg to 87 Gg and rerun GEOS-Chem

Evaluate model representation of surface NH_x ($NH_3 + NH_4^+$)



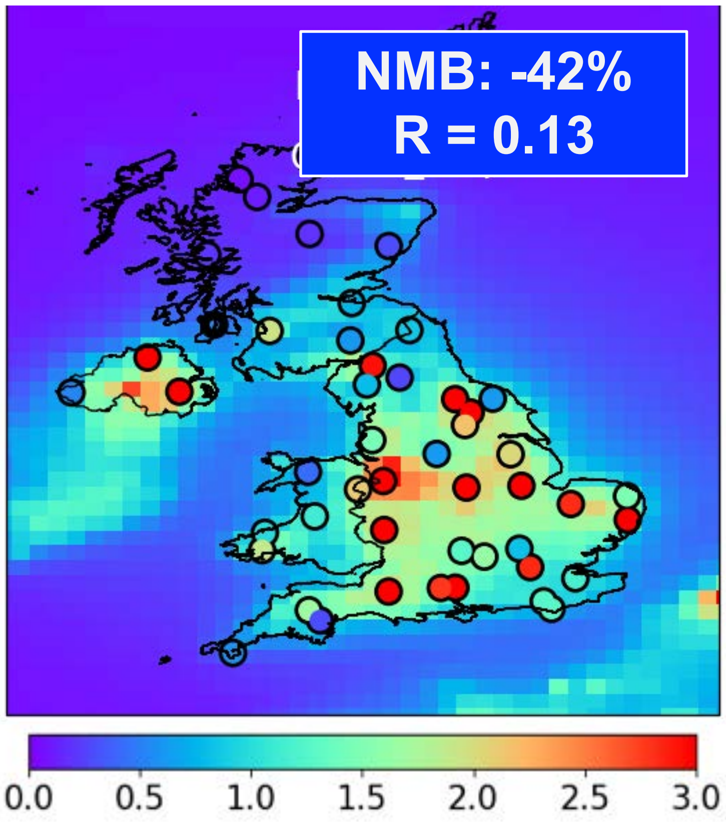
Gas-phase abundance of NH_3 depends on sulfate and nitrate

Does the model get this balance right?

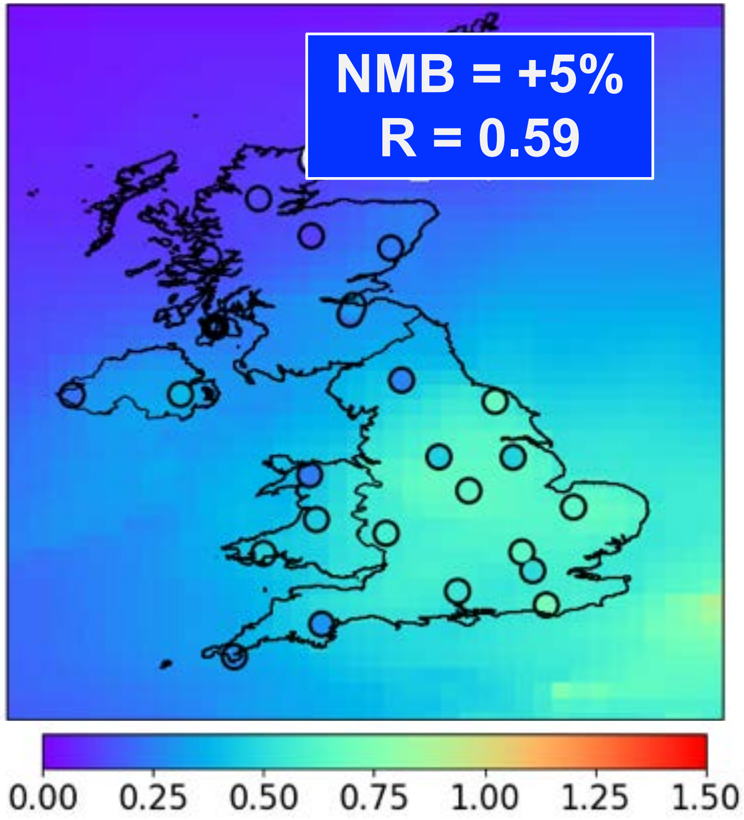
Focus on March-September (agricultural activity)

NMB: Model normalized mean bias

Mar-Sep mean NH_3 [$\mu g\ m^{-3}$]



Mar-Sep mean NH_4^+ [$\mu g\ m^{-3}$]



Model underestimates NH_3, slightly overestimates ammonium (NH_4^+)

Model underestimates surface NH_x by 40%

Remaining positive bias in SO_2

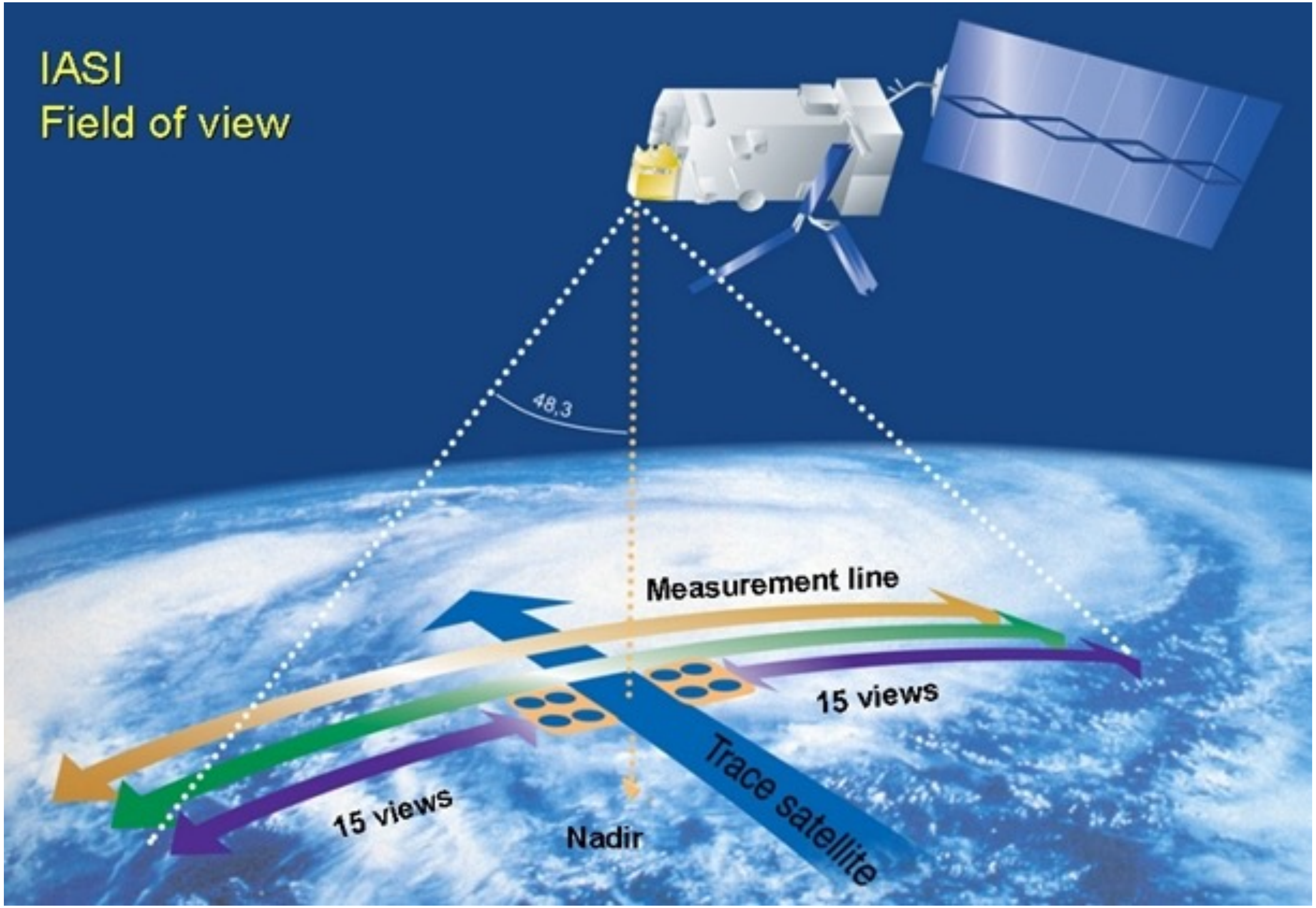
Infrared Atmospheric Sounding Interferometer (IASI) Instrument

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

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

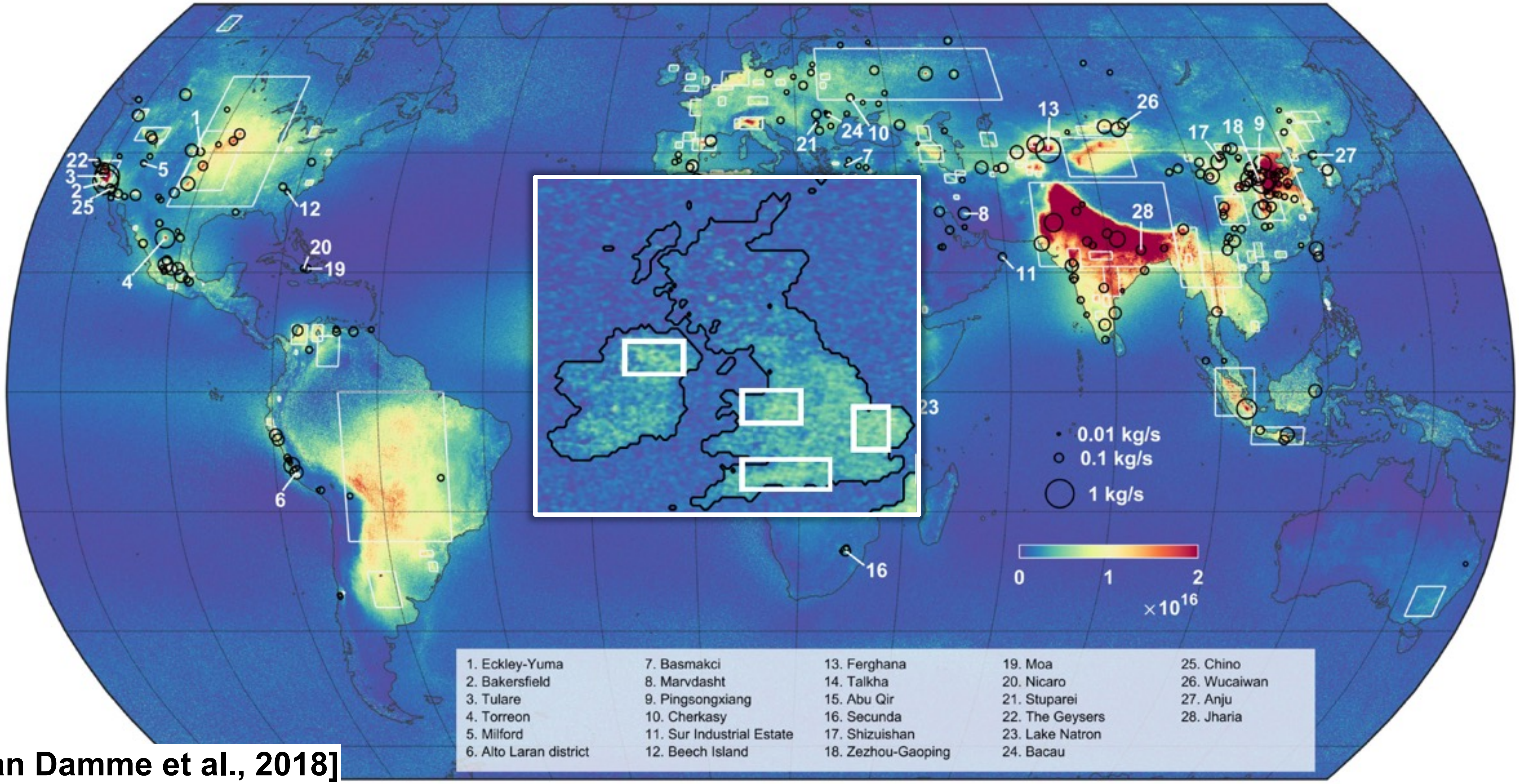
<u>Swath width:</u>
2200 km

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



Infrared Atmospheric Sounding Interferometer (IASI) Instrument

IASI extensively used to identify and assess inventories of large point sources of NH_3

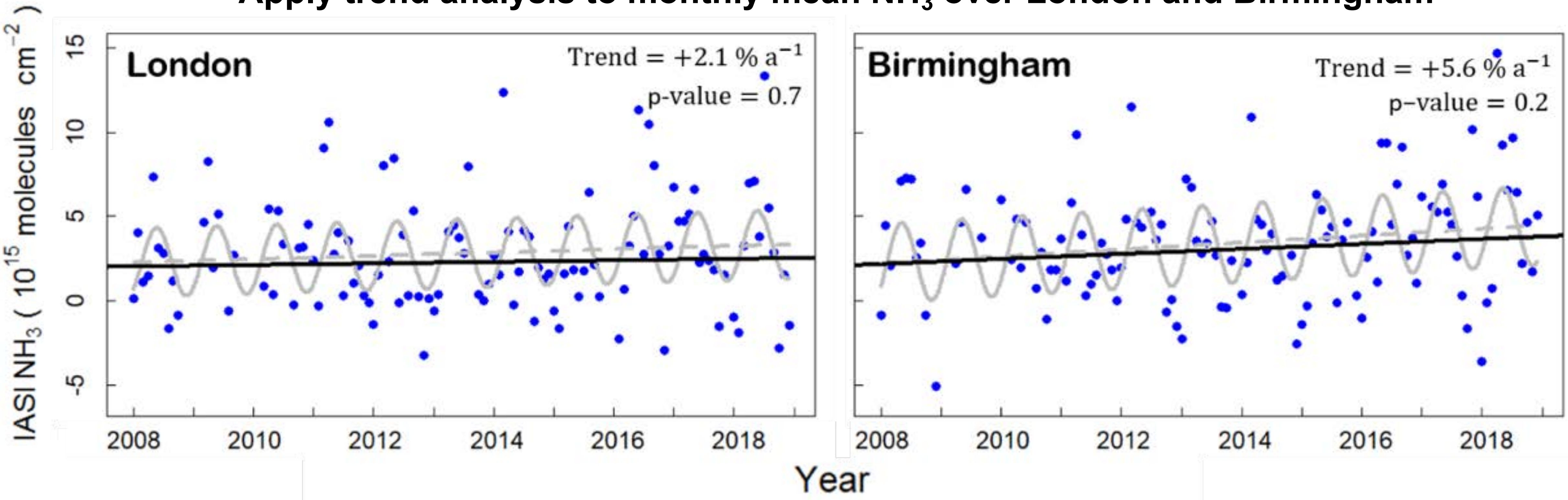


[Van Damme et al., 2018]

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



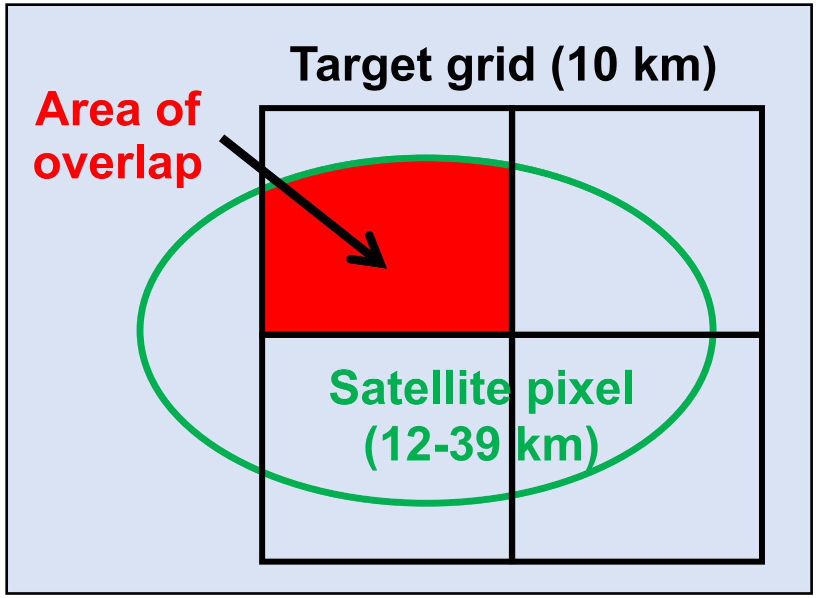
[Vohra et al., ACPD, 2020]

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

Fine-scale sampling of IASI using Oversampling

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

Oversampling Technique

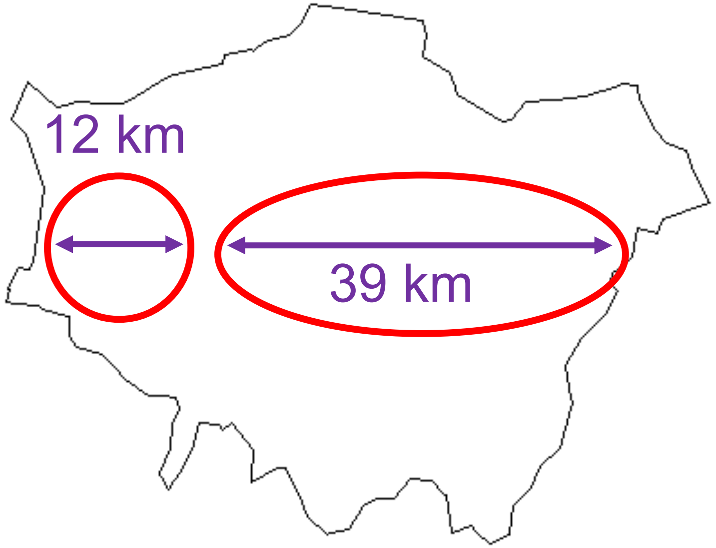


Weights each IASI NH_3 pixel by area of overlap and the reported uncertainty

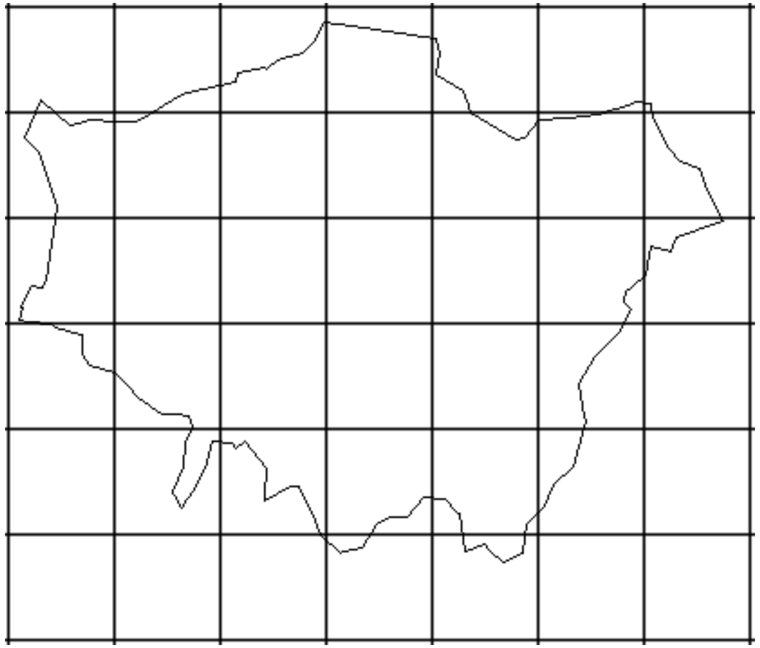
Oversampling code: L. Zhu, SUSTech (Zhu et al., 2017)

Oversampling technique over London

IASI ground pixel



0.1° x 0.1° (~10 km) grid

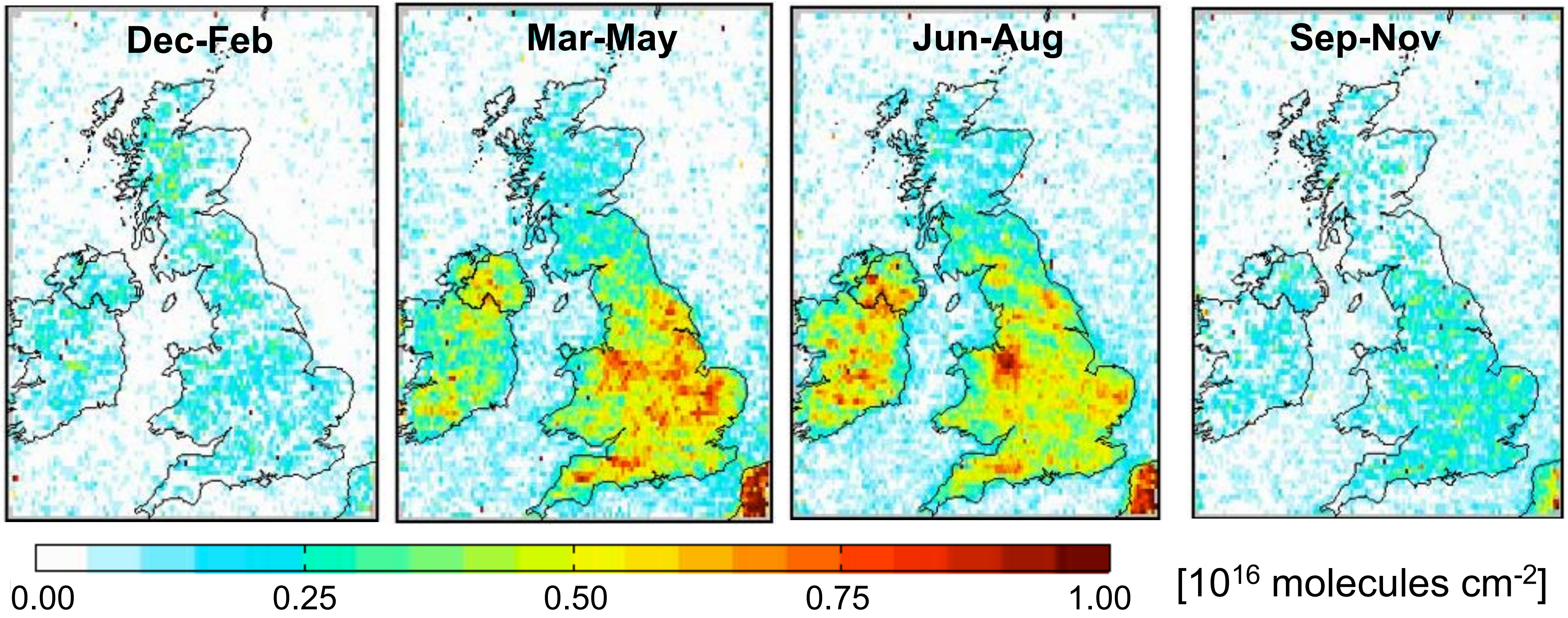


Lose time (temporal) resolution; gain spatial resolution

Multiyear seasonal mean oversampled IASI NH_3

Observations of column densities are available since 2007 from the IASI instrument

Seasonal multiyear (2008-2018) mean IASI NH_3 on a 0.1° x 0.1° (~10 km) grid



<u>Units</u> are number of ammonia molecules in a column of air from the surface to the satellite

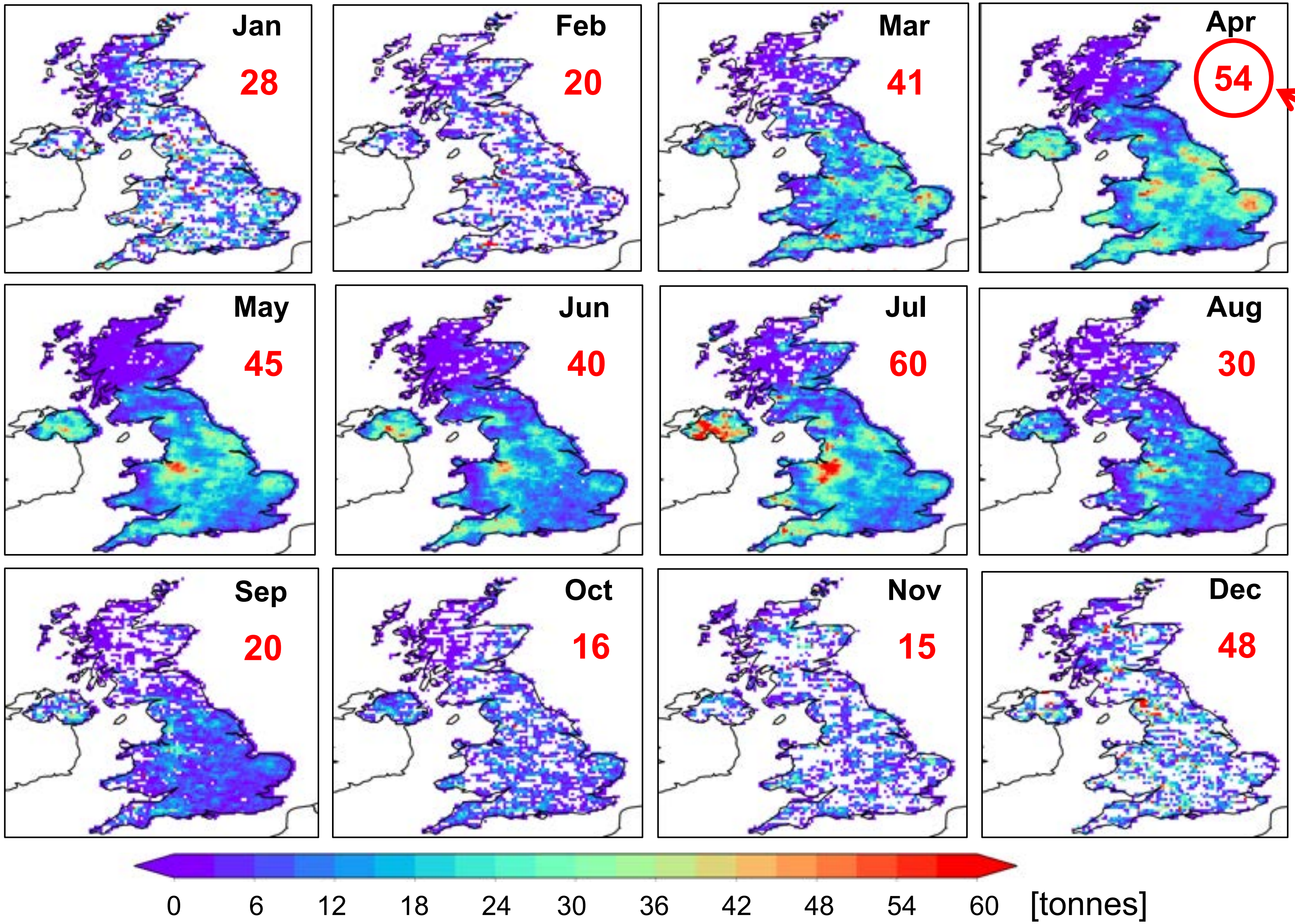
<u>IASI data providers</u>: M. Van Damme, L. Clarisse, P.-F. Coheur, ULB, Belgium

UK IASI-derived ammonia emissions

Convert IASI NH_3 column concentrations to surface emissions of NH_3

Data noisy in winter, start of spring, and end of autumn

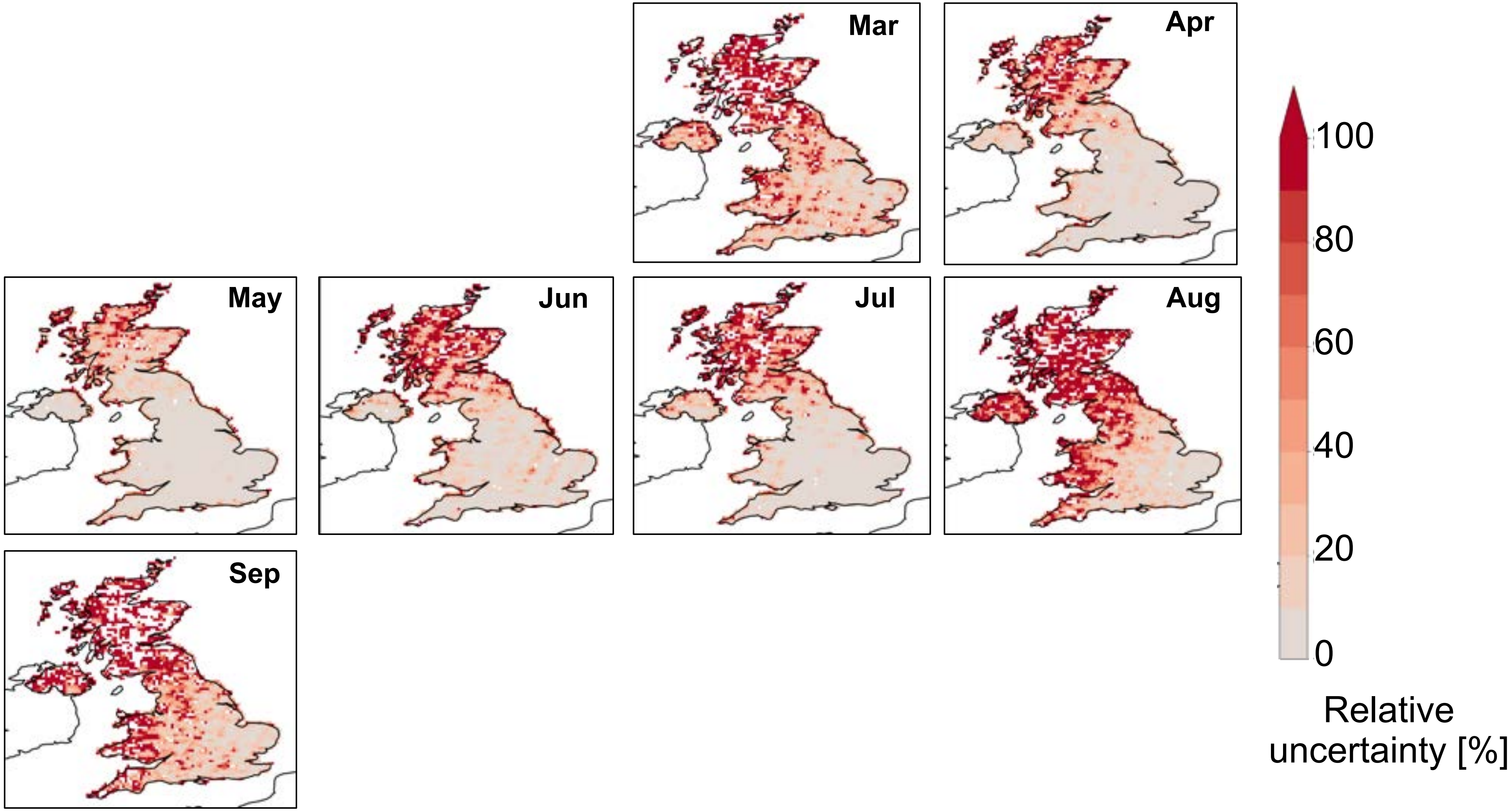
Challenging to retrieve NH_3 in these months



Total monthly emissions (Gg)

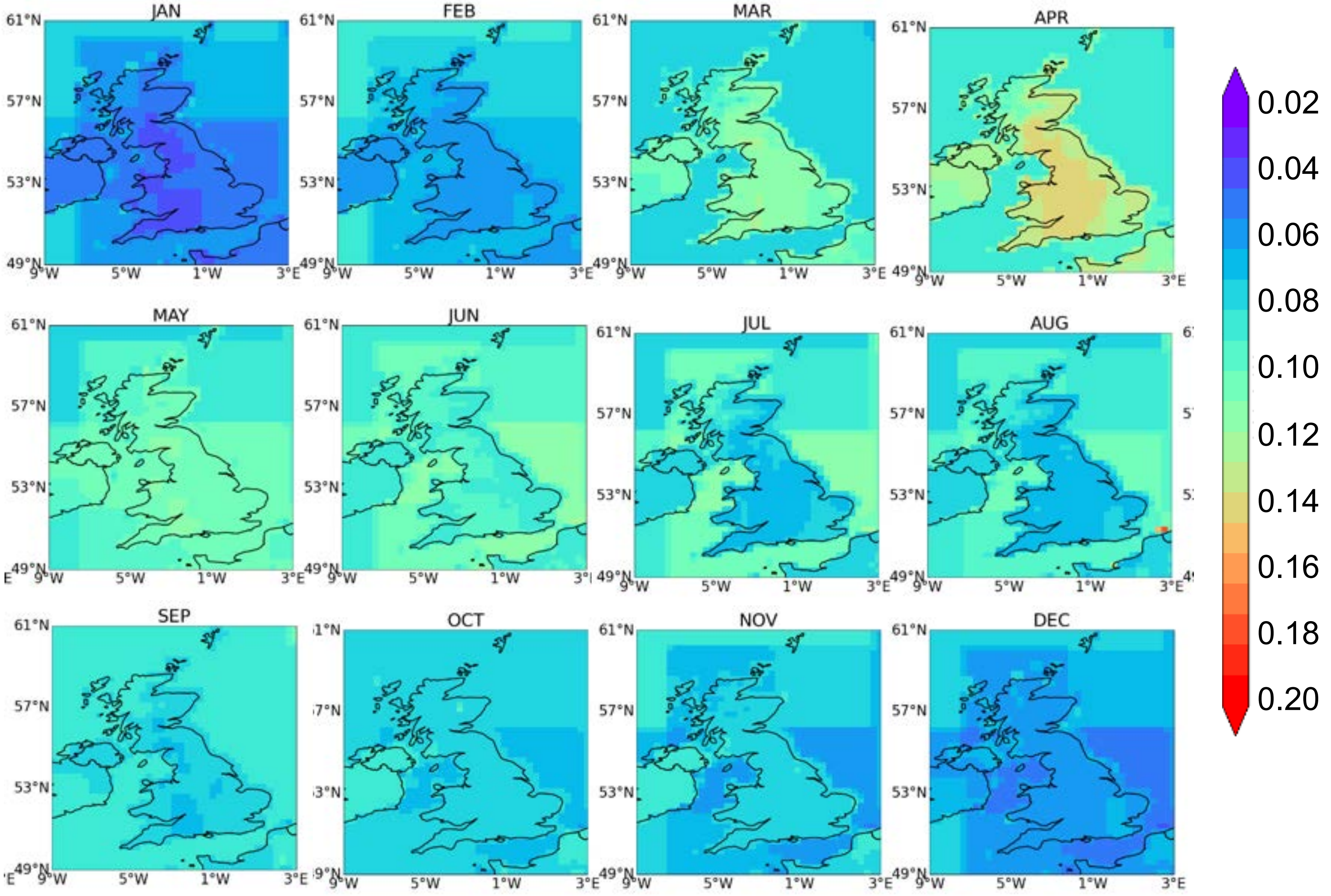
Account for Observation Uncertainties

IASI NH_3 column concentrations susceptible to large uncertainties in cold months



Only consider months with relatively low IASI uncertainty: March-September

Temporal scaling factors



NAEI emissions are annual

IASI-derived emissions monthly

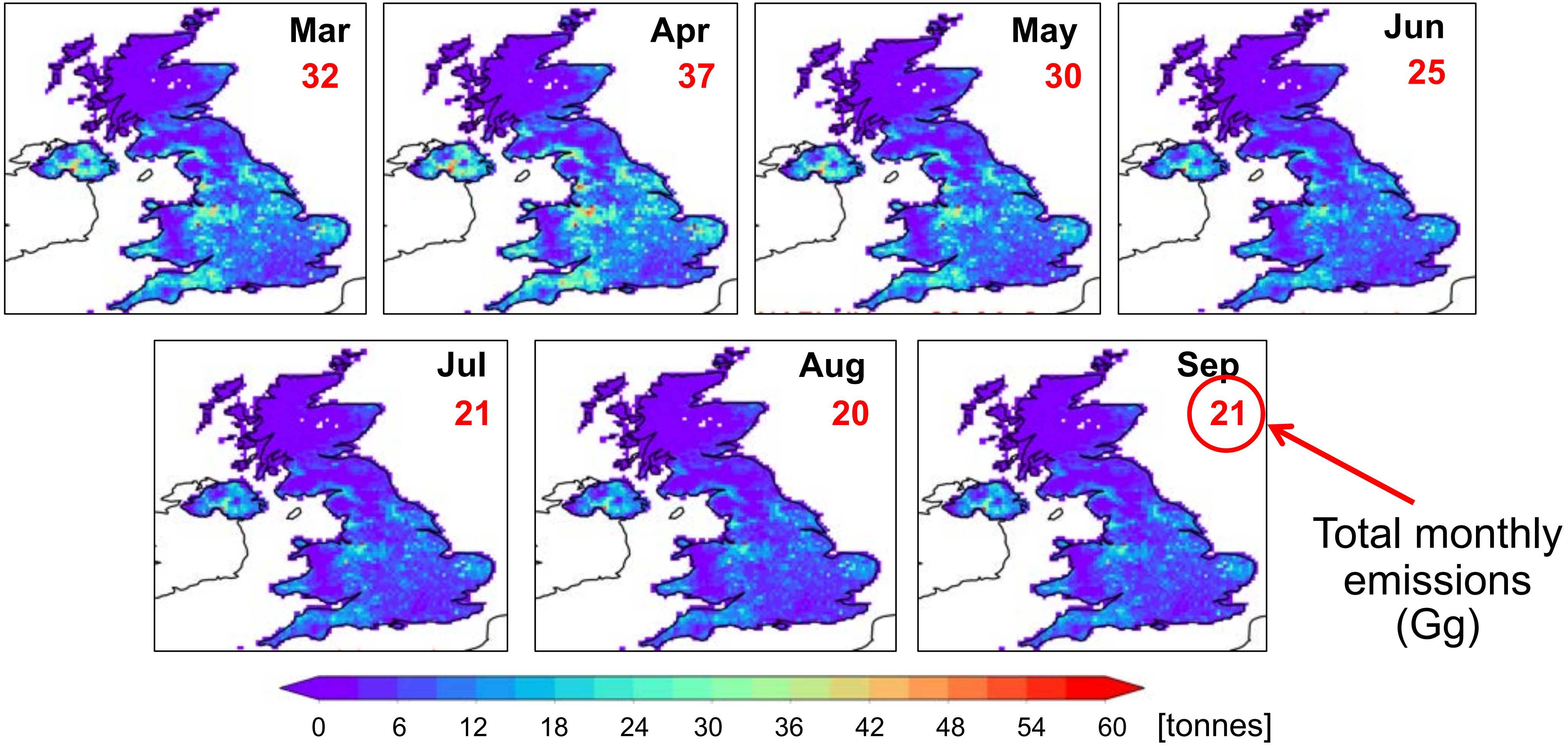
Monthly scale factors used in GEOS-Chem applied to NAEI

Generally within narrow range (4-14%)

Assumed to peak in April

NAEI ammonia emissions with assumed seasonality

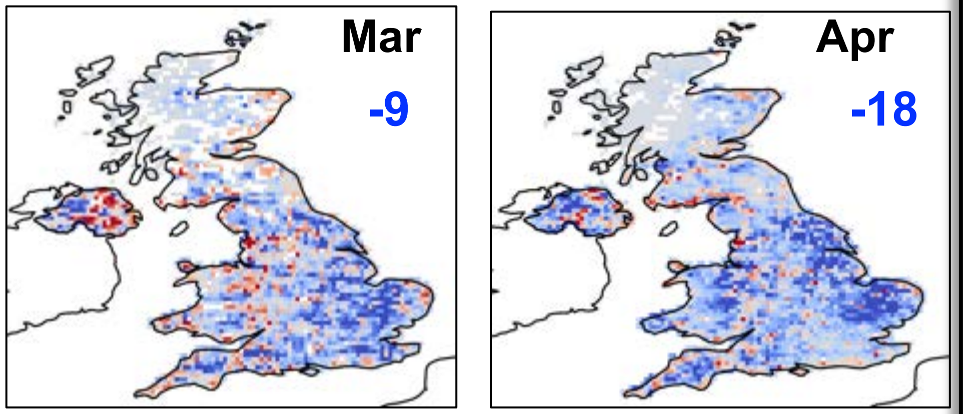
NAEI NH_3 emissions with monthly scaling factors used in GEOS-Chem applied

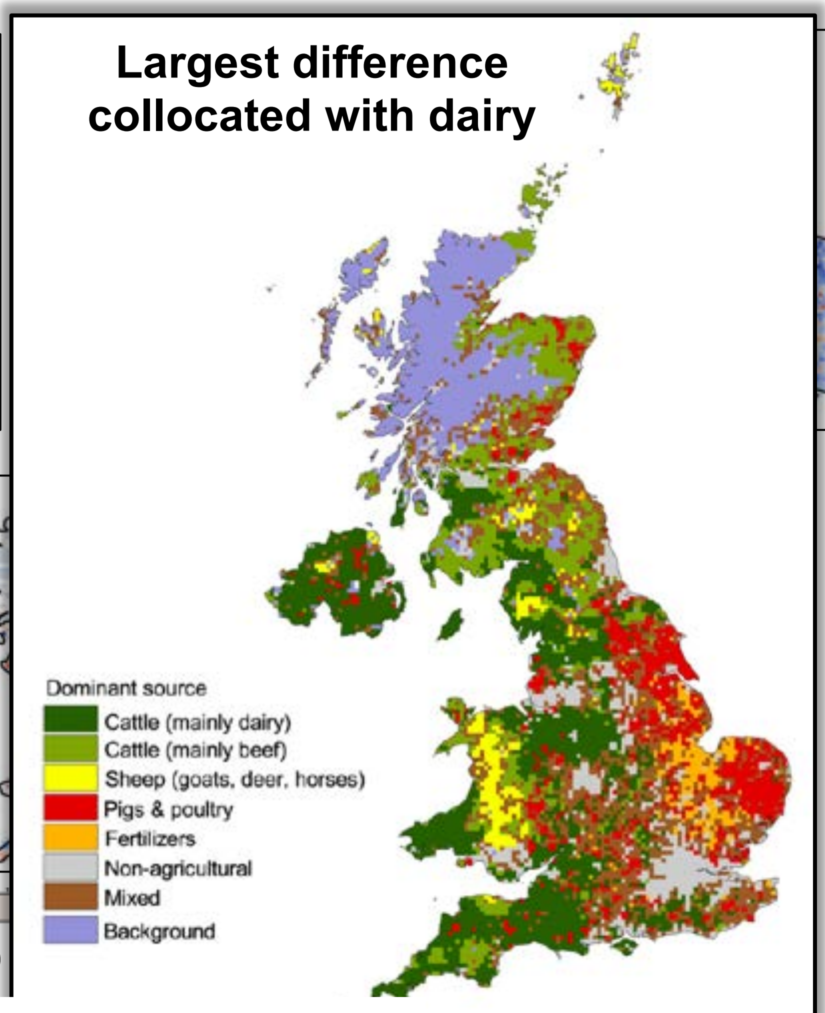


NAEI NH_3 emissions in March-September are 67% of annual NAEI NH_3 emissions

Assessment of the UK National Emission Inventory

Compare IASI-derived and NAEI NH_3 emissions with representative scaling factors applied to the NAEI





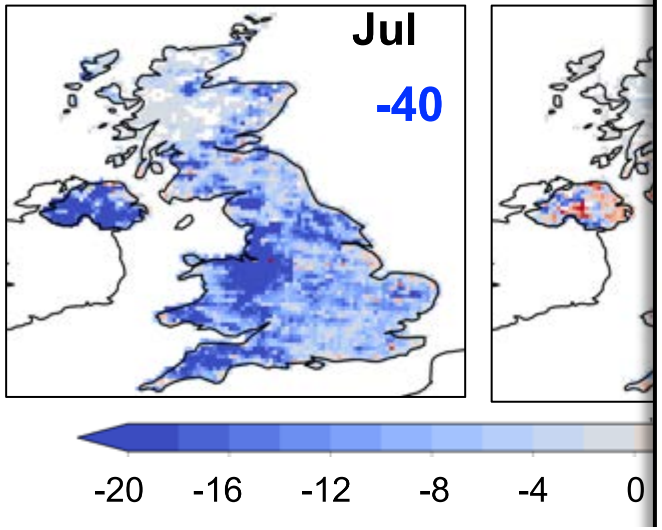
[Hellsten et al., 2008]

Difference in monthly emissions (Gg)

Blue: NAEI < IASI

Red: NAEI > IASI

Red mostly large uncertainties



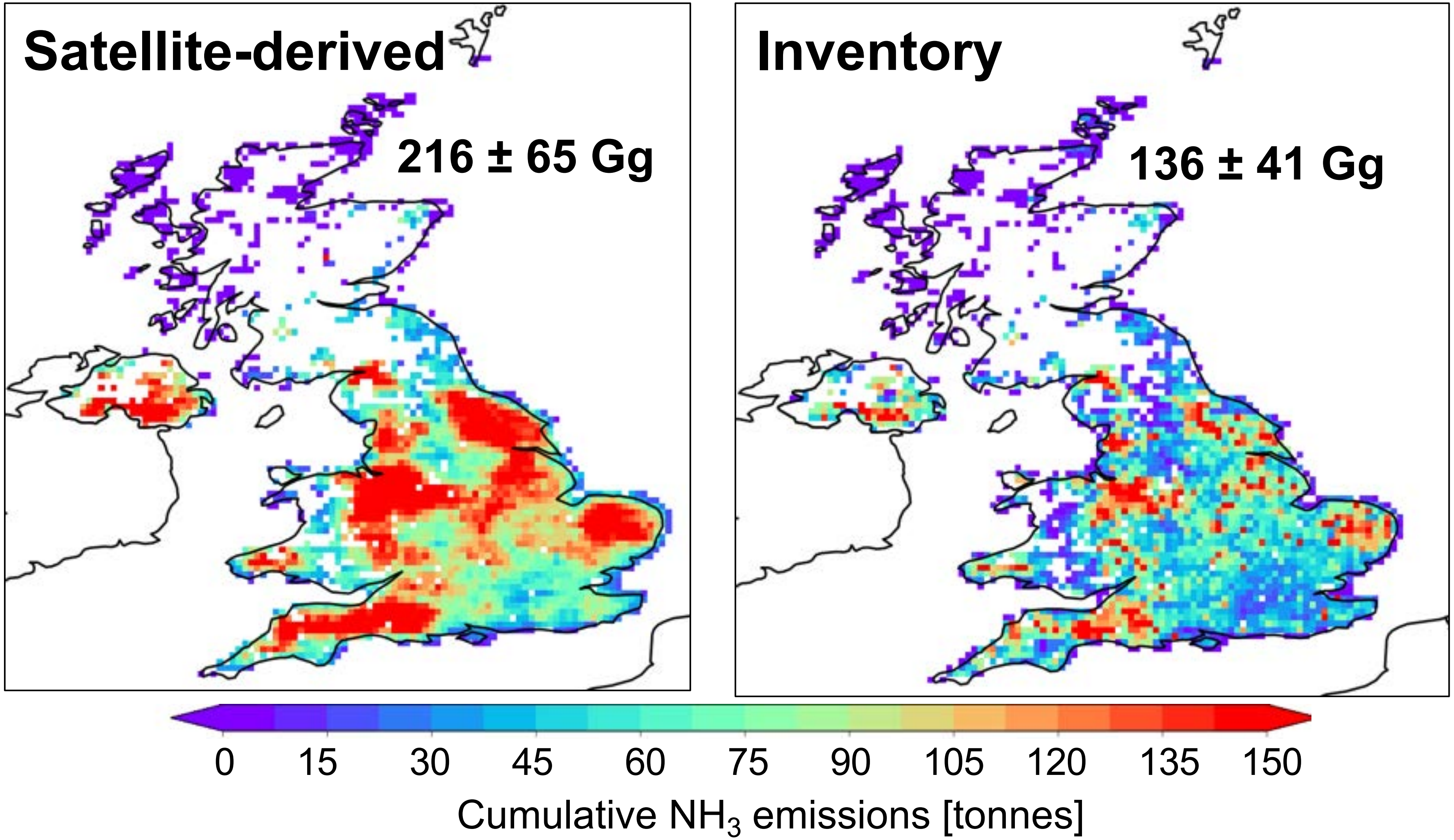
Largest discrepancy over locations dominated by **dairy farms**

Comparison of total March-September emissions

Comparison suggests satellite-derived estimate suggests NAEI underestimates emissions.

Similar relative errors:
IASI: 29%
NAEI: 31%

Satellite-derived emissions 60% more than NAEI emissions



Implication: underestimate $PM_{2.5}$, in particular ammonium and nitrate components

Concluding Remarks

- UK NAEI overestimates SO_2 emissions by more than a factor of 2 for point sources
- Satellite-derived NH_3 emissions from IASI and GEOS-Chem are 60% more than the UK NAEI estimate, but likely 40-60% if factor in SO_2 emission uncertainties.
- Largest underestimate in NAEI is over dairy farms that have peak emissions in July, according to the IASI-derived emissions
- Underestimate in NAEI NH_3 emissions obtained with IASI corroborated by surface observations and another instrument (CrIS) with a midday overpass (not shown here)
- Implication for air quality models that use the NAEI is an underestimate in nitrate and ammonium components of $PM_{2.5}$ due to underestimate in NH_3 emissions

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

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

I have 2 PhD studentships and 1 postdoc position in my group.

For queries about today's presentation or for details about these opportunities, contact me at e.marais@ucl.ac.uk