

Health effects of agricultural emissions of ammonia:

A focus on the UK with global implications



Ammonia pathway leading to adverse health outcomes

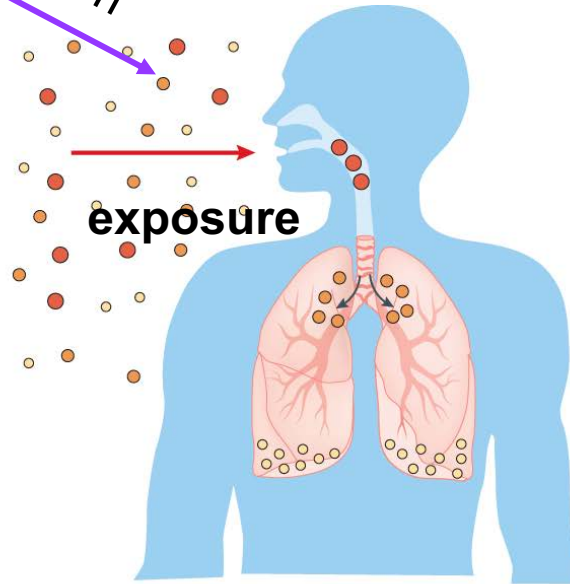
Ammonia (NH_3)
emissions

transport

deposition

aerosol formation

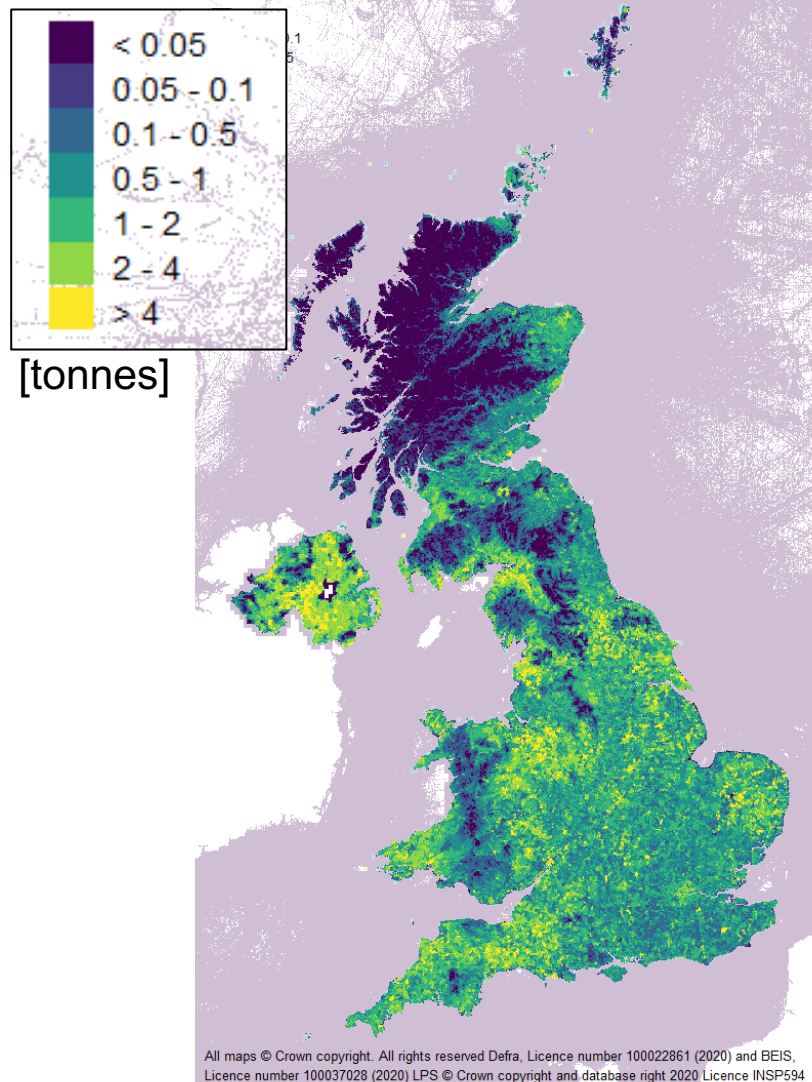
exposure



Ammonia emissions in the UK: the bottom-up perspective

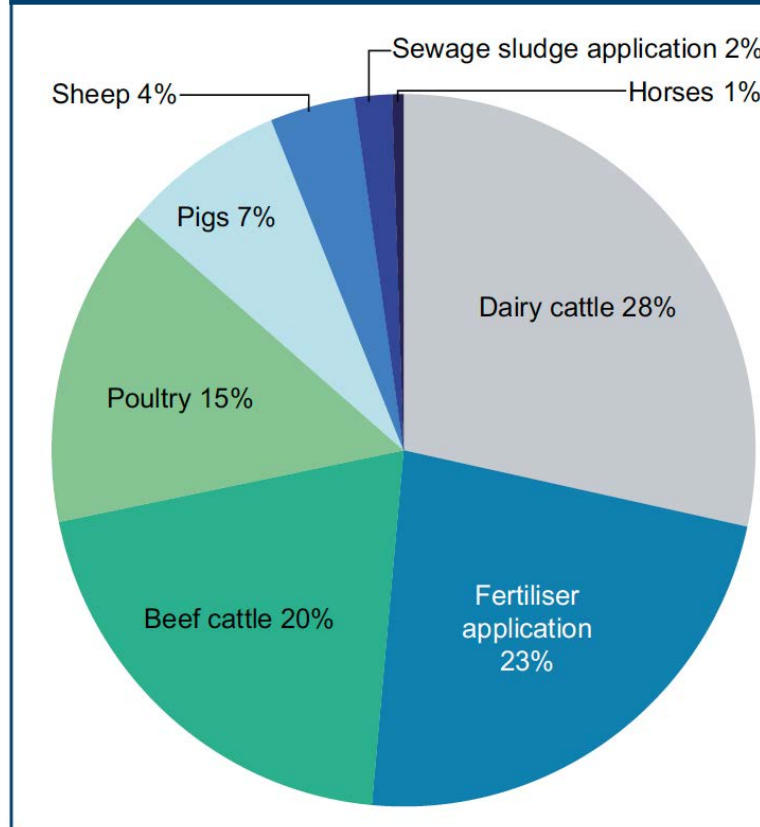
Emissions Spatial Variability

Annual NH_3 emissions at 1 km

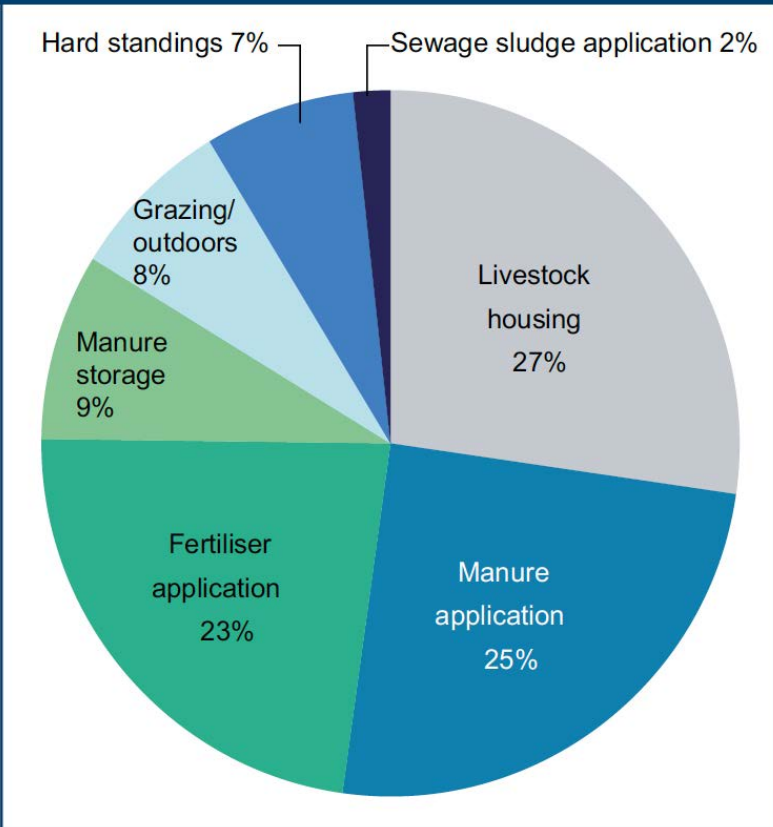


Contributions of activities to ammonia emissions

UK agricultural ammonia emissions (2016)
by livestock and fertiliser category



UK agricultural ammonia emissions (2016)
by management category



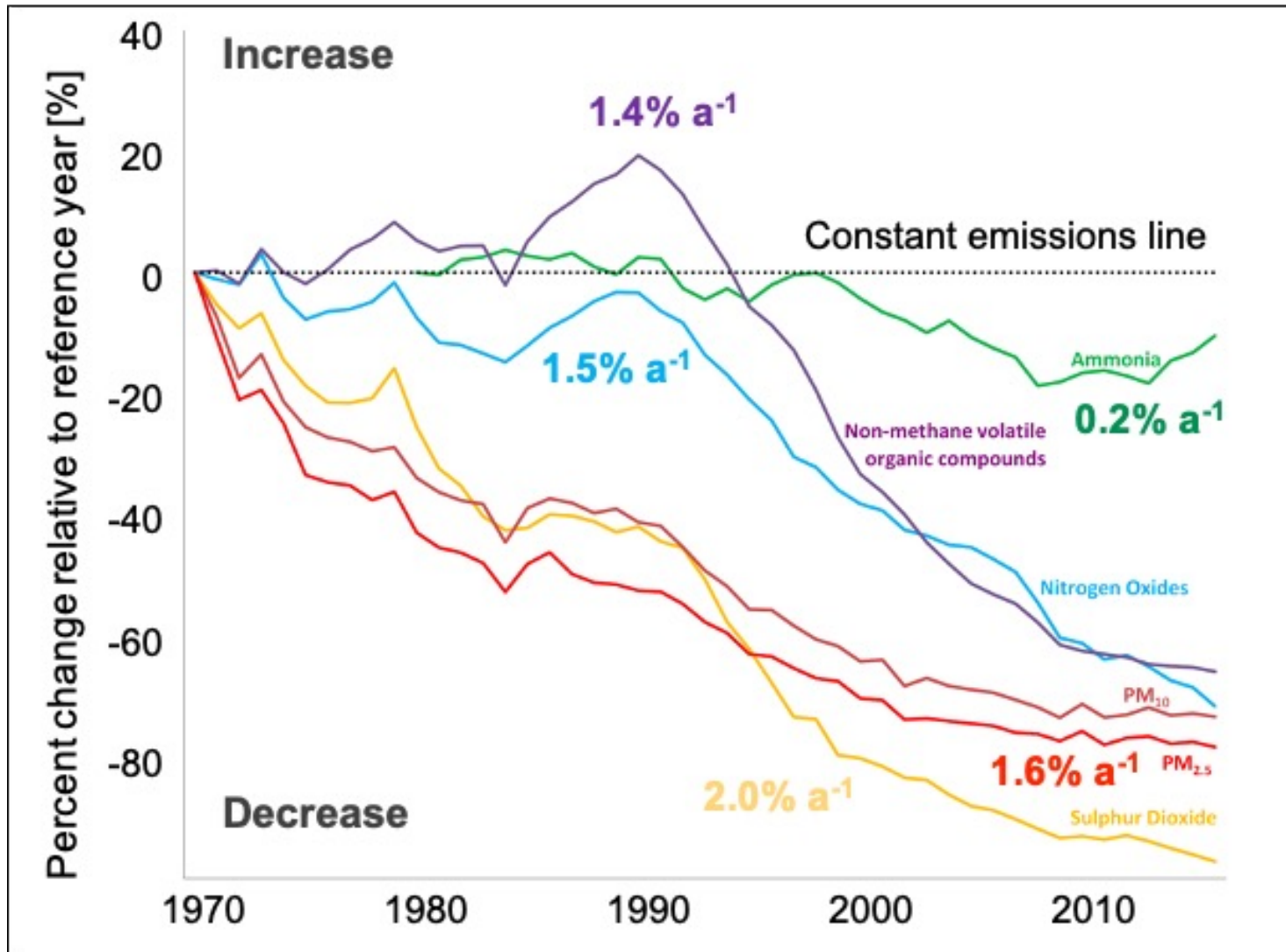
[UK Clean Air Strategy, 2019]

Beef, dairy, and fertilizer use dominate

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

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: Coarse particles

Red: Fine particles

Yellow: sulfur dioxide

[Adapted from Defra, 2018]

Successful decline in all primary PM_{2.5} sources and precursor emissions, except ammonia (NH₃)

Assess national inventory estimates of ammonia emissions with satellite observations

Published in JGR: Atmospheres:

<https://agupubs.onlinelibrary.wiley.com/doi/epdf/10.1029/2021JD035237>



Engineering and
Physical Sciences
Research Council



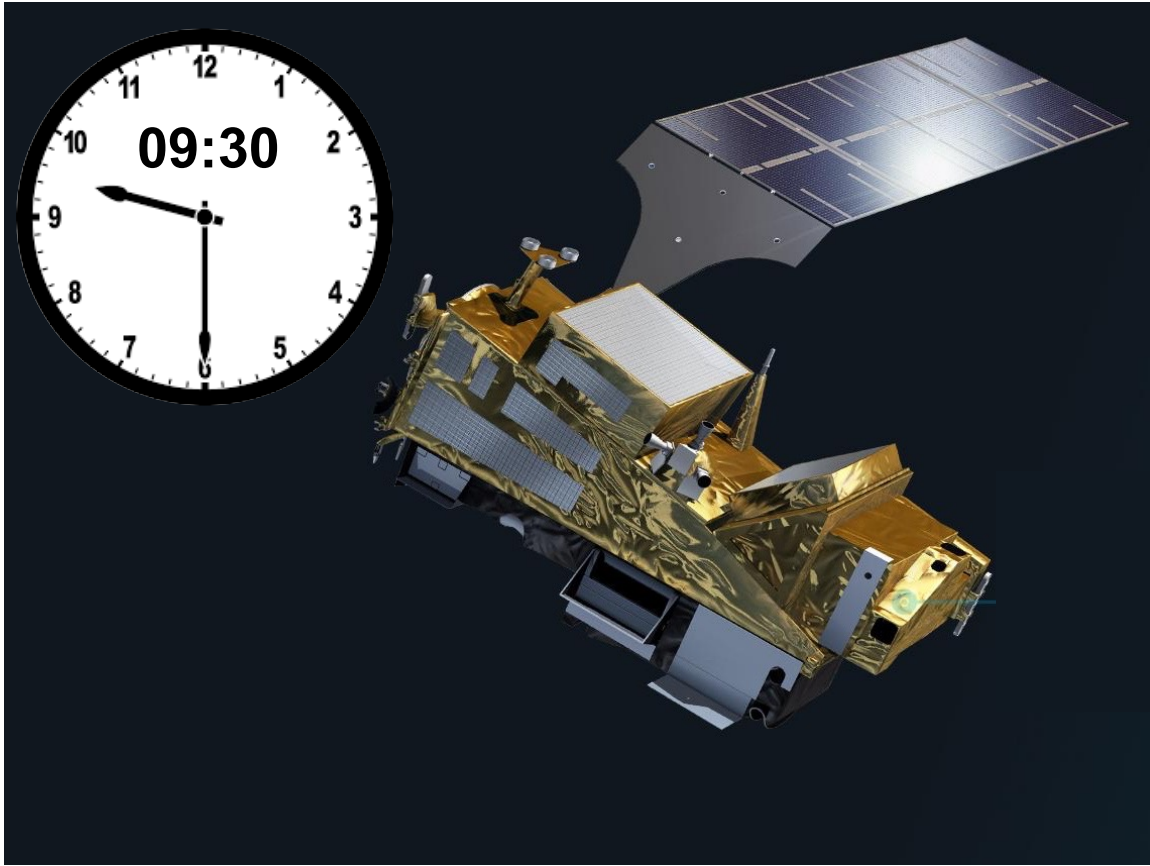
Natural
Environment
Research Council



Department
for Environment
Food & Rural Affairs

Instruments in space measuring NH_3 column densities

IASI: Infrared Atmospheric Sounding Interferometer



Resolution: 12 km at nadir

Swath width: 2200 km

Launch date: 2006 (2012, 2018, 2024, 2031, 2038)

Years used: 2008-2018

CrIS: Cross-track Infrared Sounder



Resolution: 14 km at nadir

Swath width: 2200 km

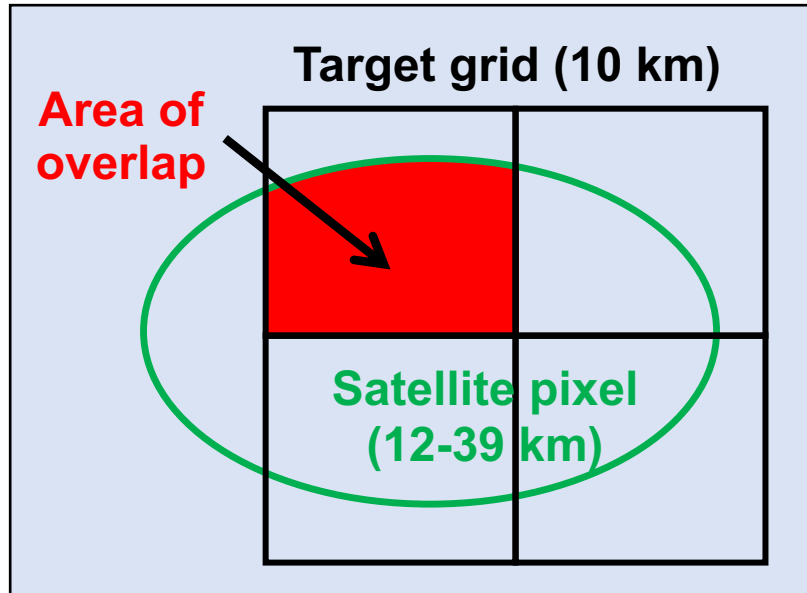
Launch date: 2011 (2017, 2022, 2027, 2032)

Years used: 2013-2018

Fine-scale regridding of satellite observations by oversampling

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

Oversampling Technique

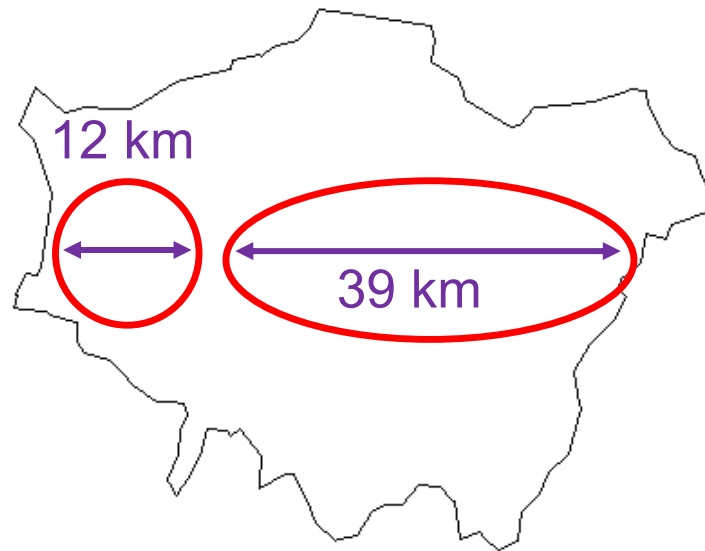


Weights each IASI NH₃ pixel by area of overlap and the reported uncertainty

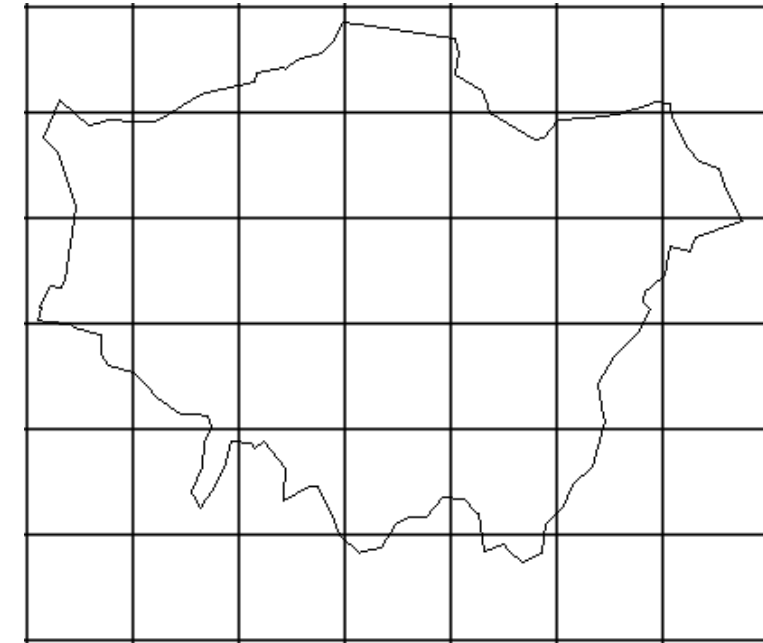
Oversampling code: L. Zhu,
SUSTech (Zhu et al., 2017)

Oversampling technique over London

IASI ground pixel



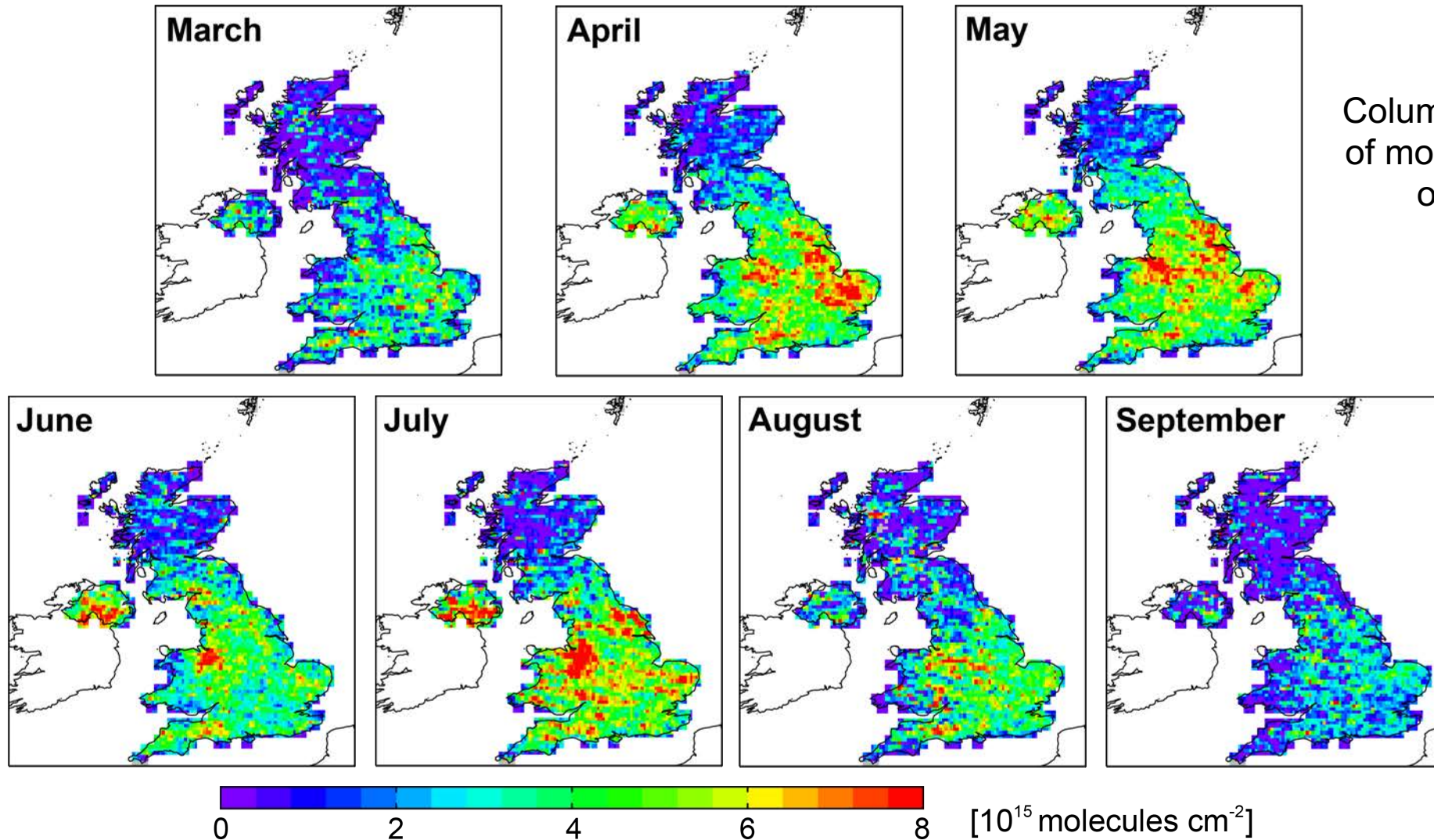
0.1° x 0.1° (~10 km) grid



Lose time (temporal) resolution; gain spatial resolution

Multiyear means from the IASI (morning overpass) instrument

Multiyear (2008-2018) monthly means for warmer months of the year



Column densities: number of molecules from surface of Earth to top of atmosphere

Climatological mean to be consistent with bottom-up ammonia emissions

Top-down estimate of ammonia emissions

Employ simple mass balance approach:

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

ABUNDANCES

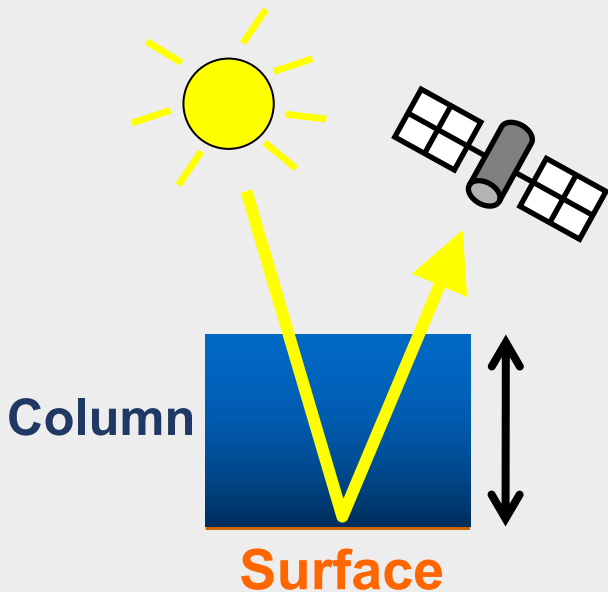


Conversion Factor

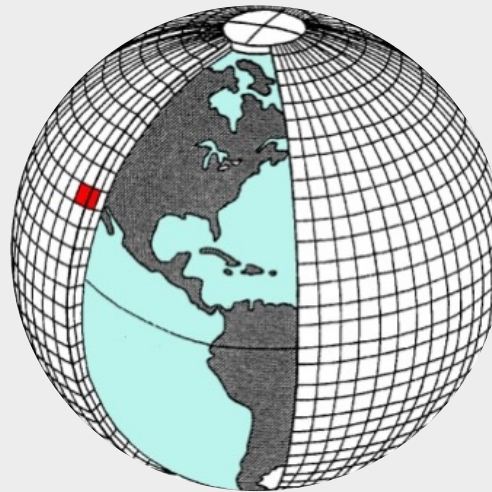


EMISSIONS

Satellite column densities



Model Concentration-to-Emission Ratio



Satellite-derived Surface Emissions

Emission

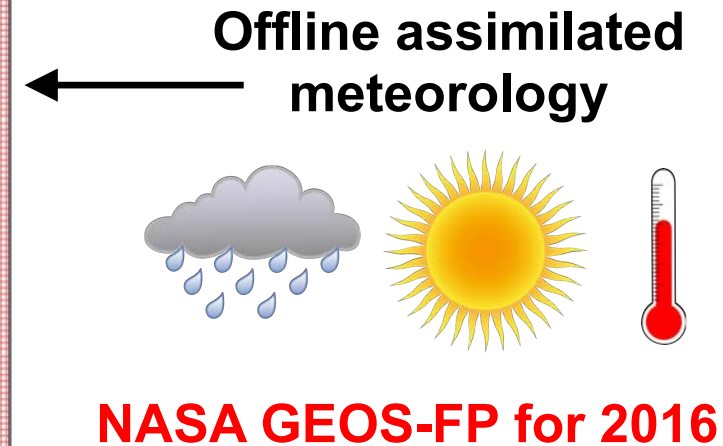
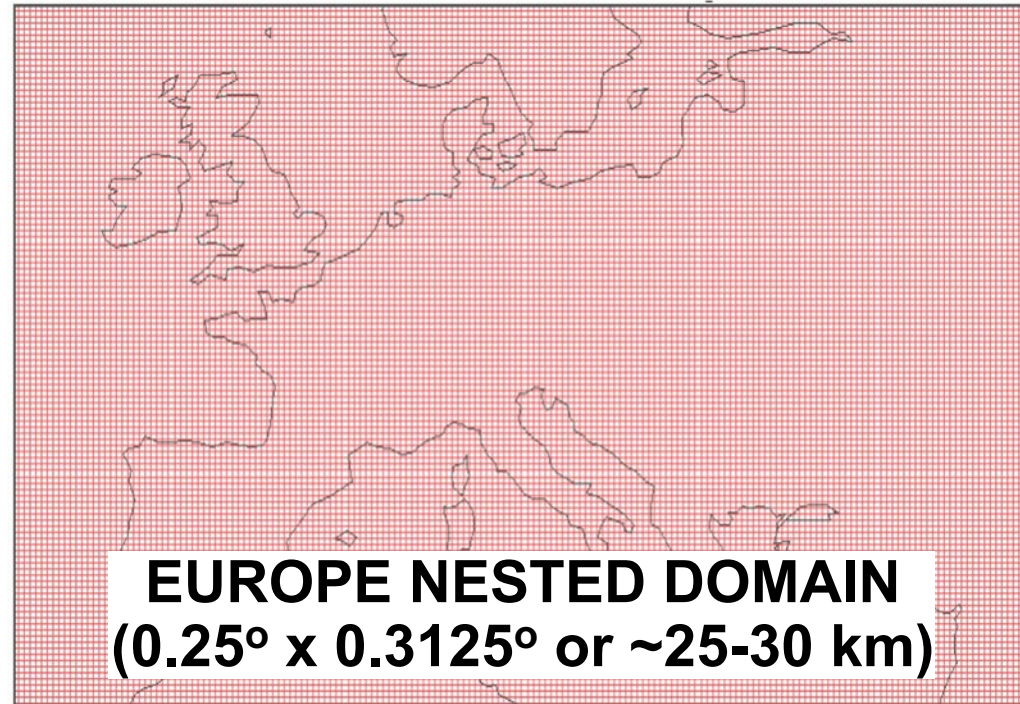
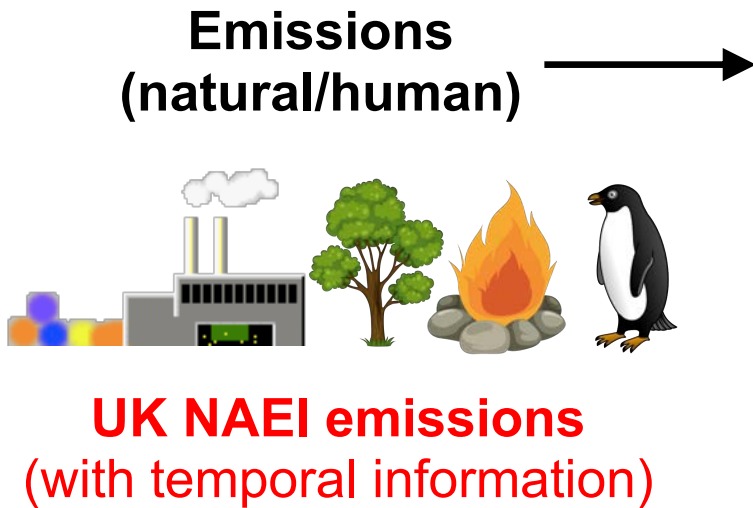


This approach possible as NH_3 has a relatively short lifetime (2-15 hours) at or near sources

Modelled concentration-to-emissions-ratio from GEOS-Chem

GEOS-Chem

3D Atmospheric Chemistry Transport Model



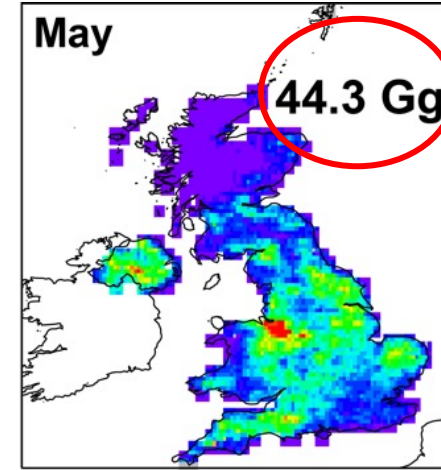
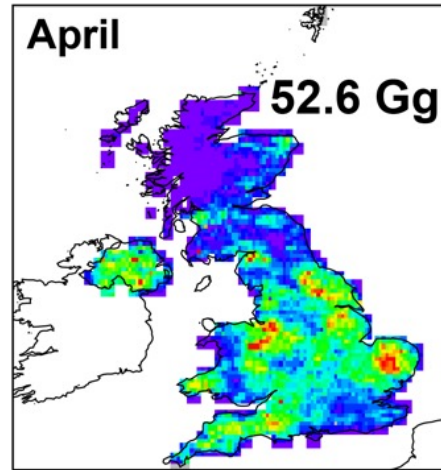
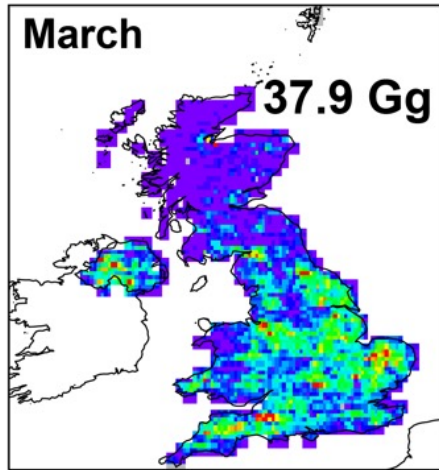
Gas phase and heterogeneous chemistry
Transport
Dry/wet deposition

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

IASI-derived multiyear (2008-2018) monthly mean NH₃ emissions

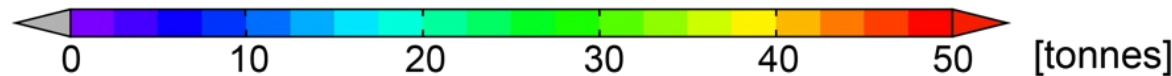
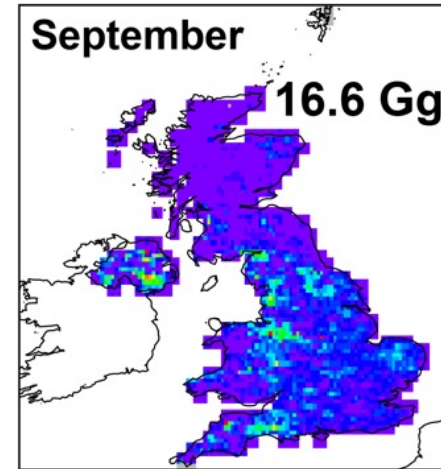
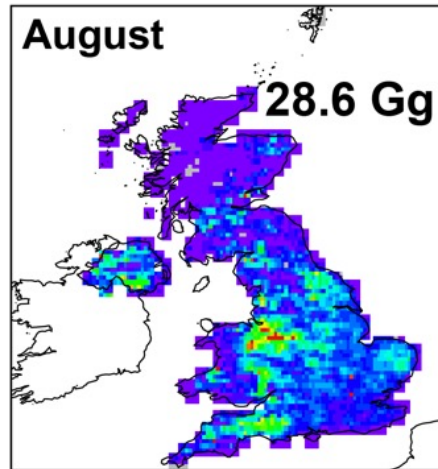
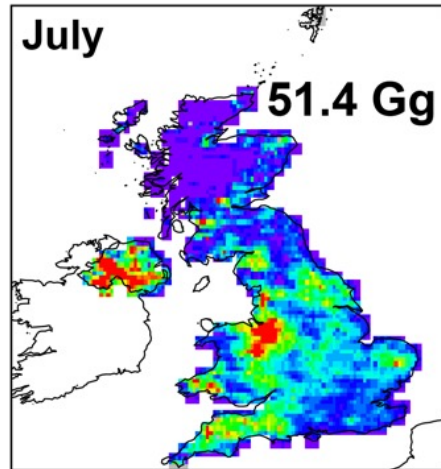
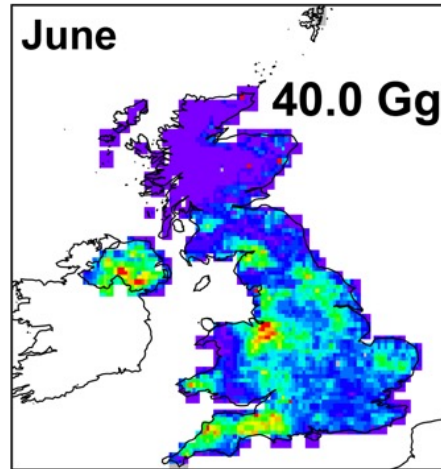
Focus on **Mar-Sep** when warm temperatures and clearer conditions increase sensitivity to surface NH₃

IASI: morning
overpass



Total monthly
emissions

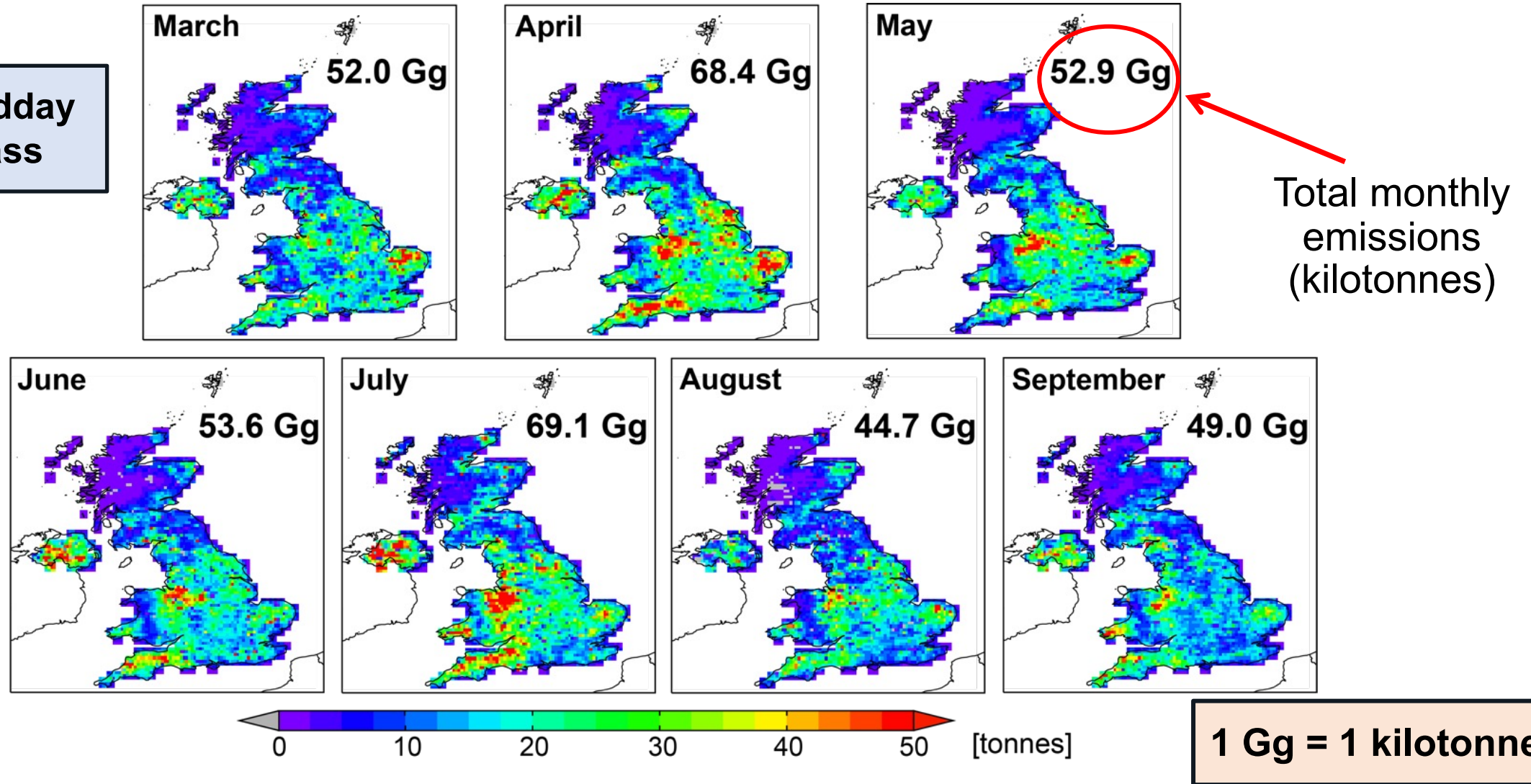
1 Gg = 1 kilotonne



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

CrIS-derived multiyear (2008-2018) monthly mean NH₃ emissions

CrIS: midday
overpass

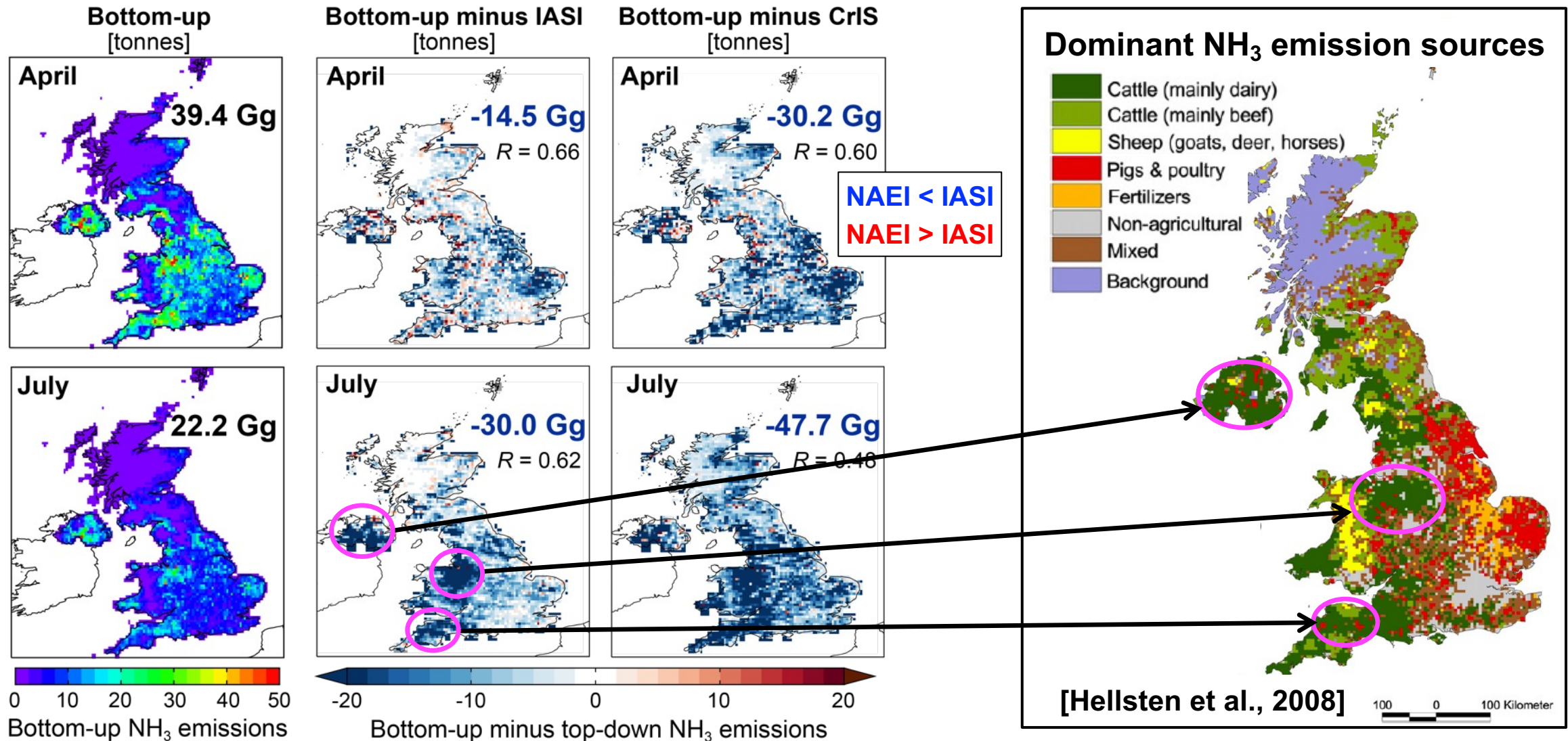


Monthly emissions for March-September from **CrIS**-derived estimates sum to **389.6 Gg**

CrIS is 43% more than IASI. Largest difference of >a factor of 2 in September.

Satellite vs inventory NH₃ emissions: spatial distribution

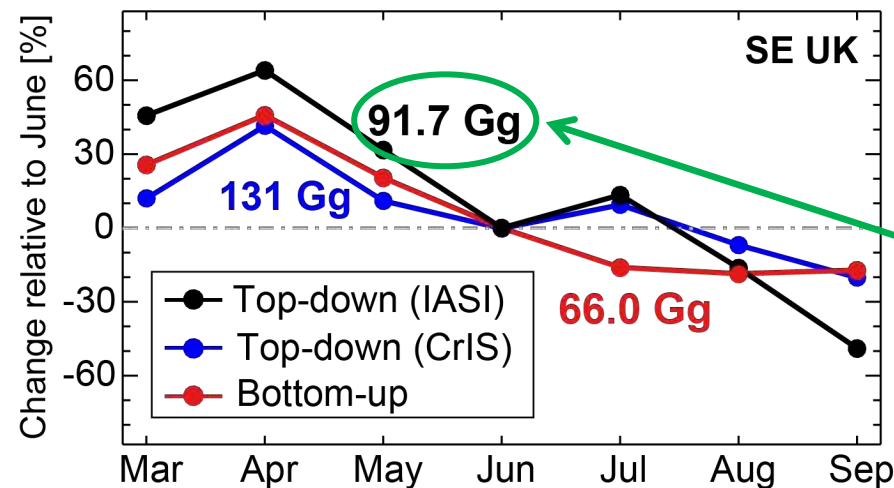
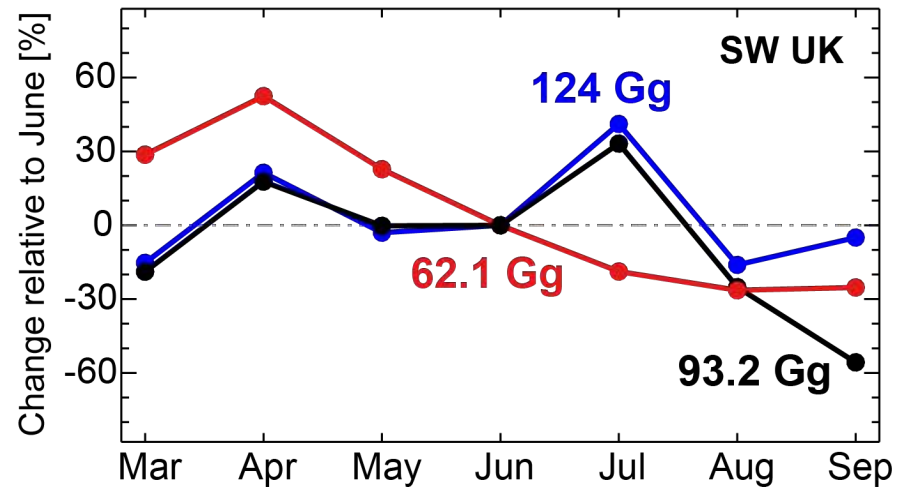
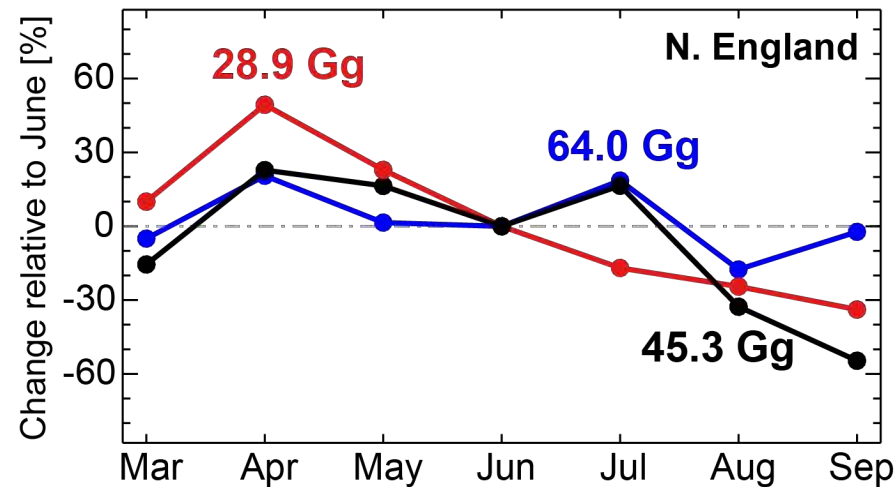
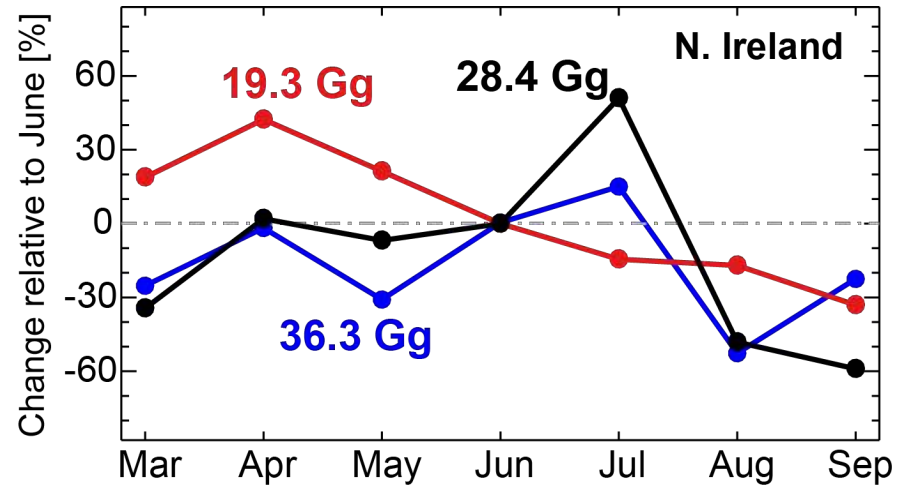
Comparison of months with peak emissions according to IASI and CrIS (April and July)



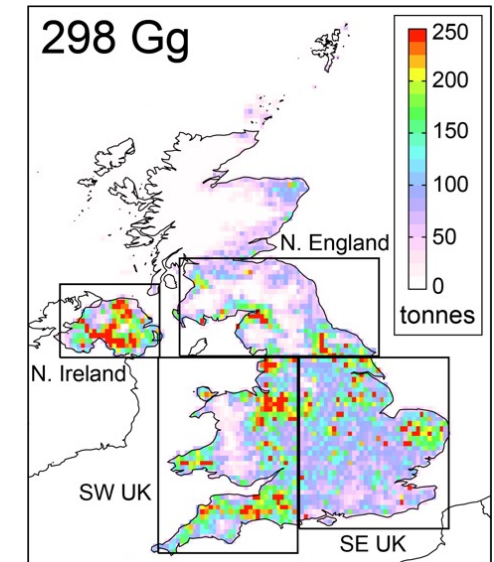
Large July difference over locations dominated by dairy cattle. Inventory is 27-49% less than the satellite values.

Satellite vs inventory NH₃ emissions: seasonality

Seasonality shown as emissions in each month relative to June



Regions and annual inventory emissions



Mar-Sep emission totals in each region

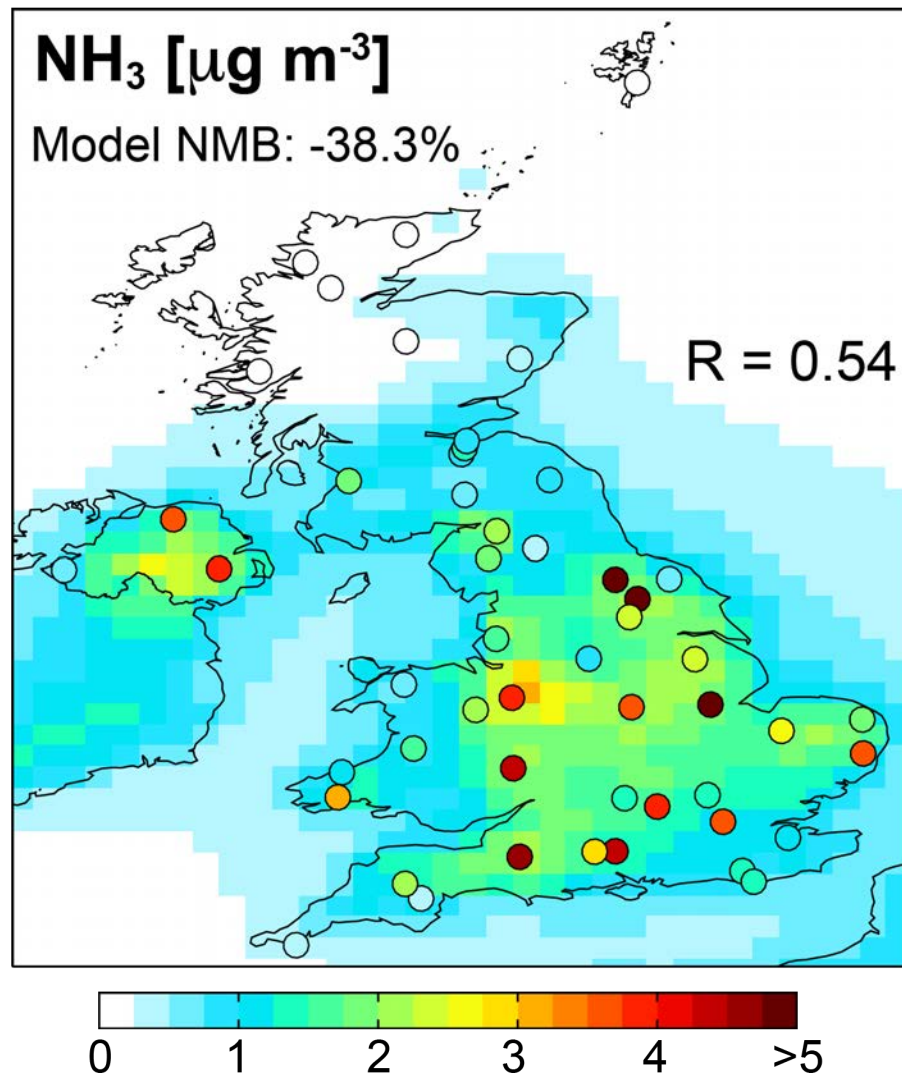
1 Gg = 1 kilotonne

All reproduce spring April peak (fertilizer & manure use). Only the satellite show summer July peak (dairy cattle?).

The increase in emissions in September in CrIS is spurious.

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%**)

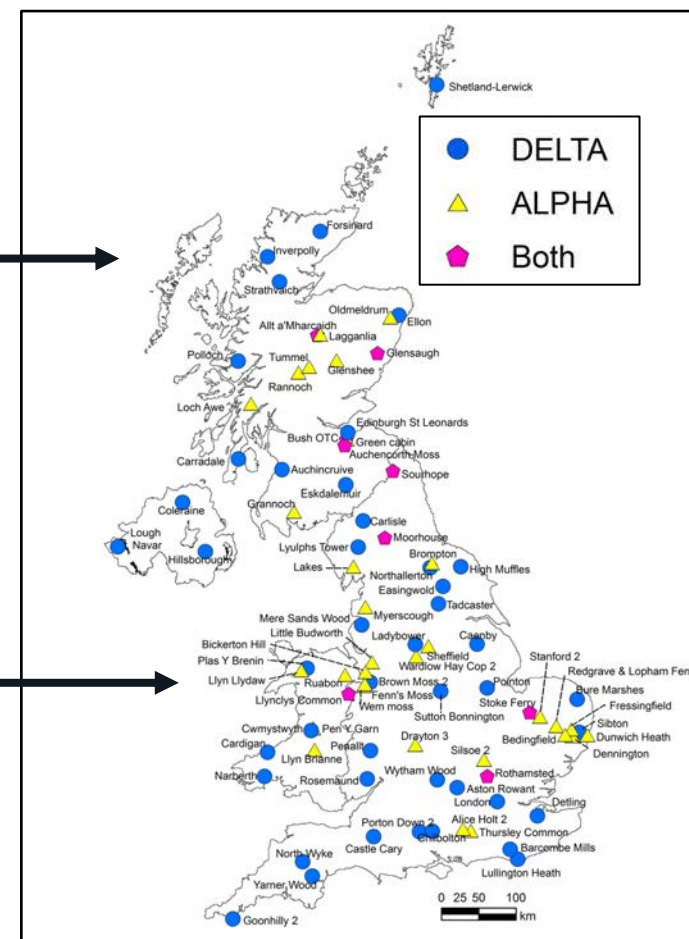


Image source:

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

GEOS-Chem underestimate in surface NH_3 driven with the NAEI corroborates results from IASI

Rural ammonia emissions influence on fine particles (PM_{2.5}) in UK cities

J. Kelly et al., 2022, *under review*



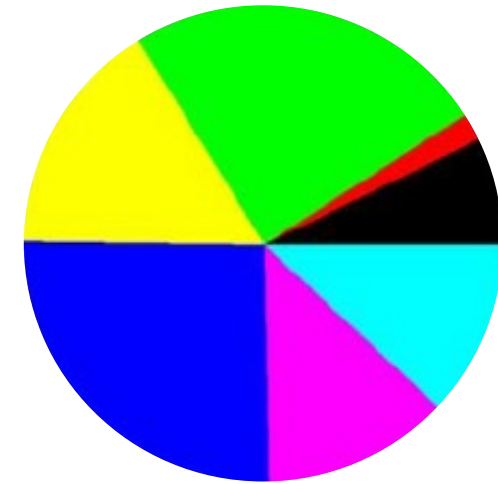
Department
for Environment
Food & Rural Affairs

Particles are a mix of components that persist for days

Direct emission
of PM_{2.5}
(primary)

Emission of gas-phase
precursors
(secondary)

PM_{2.5} includes a mix of components



Black carbon **primary**

Sulfate

Nitrate

Ammonium

secondary

Other inorganics

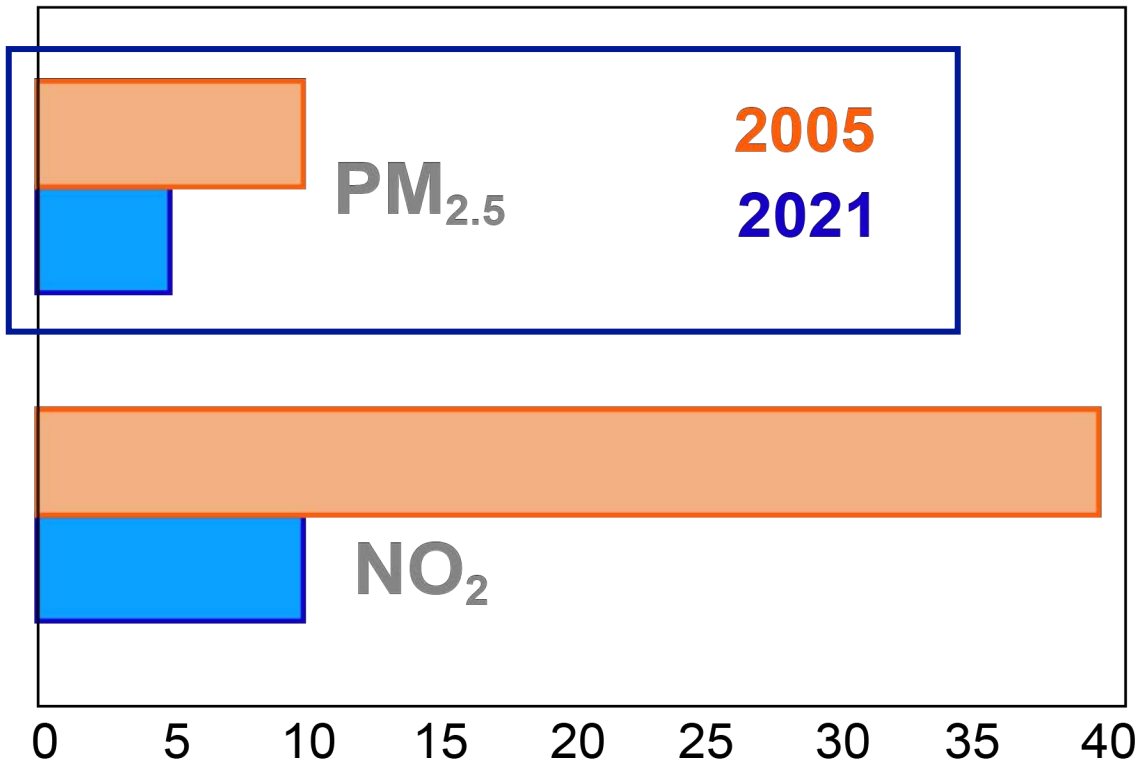
Organic aerosols **primary+secondary**

PM_{2.5} challenging to regulate, as includes many sources and has a long atmospheric lifetime (2-3 weeks)

Stricter World Health Organization (WHO) Guideline

(<https://apps.who.int/iris/handle/10665/345329>)

WHO Annual Air Quality Guidelines [$\mu\text{g m}^{-3}$]

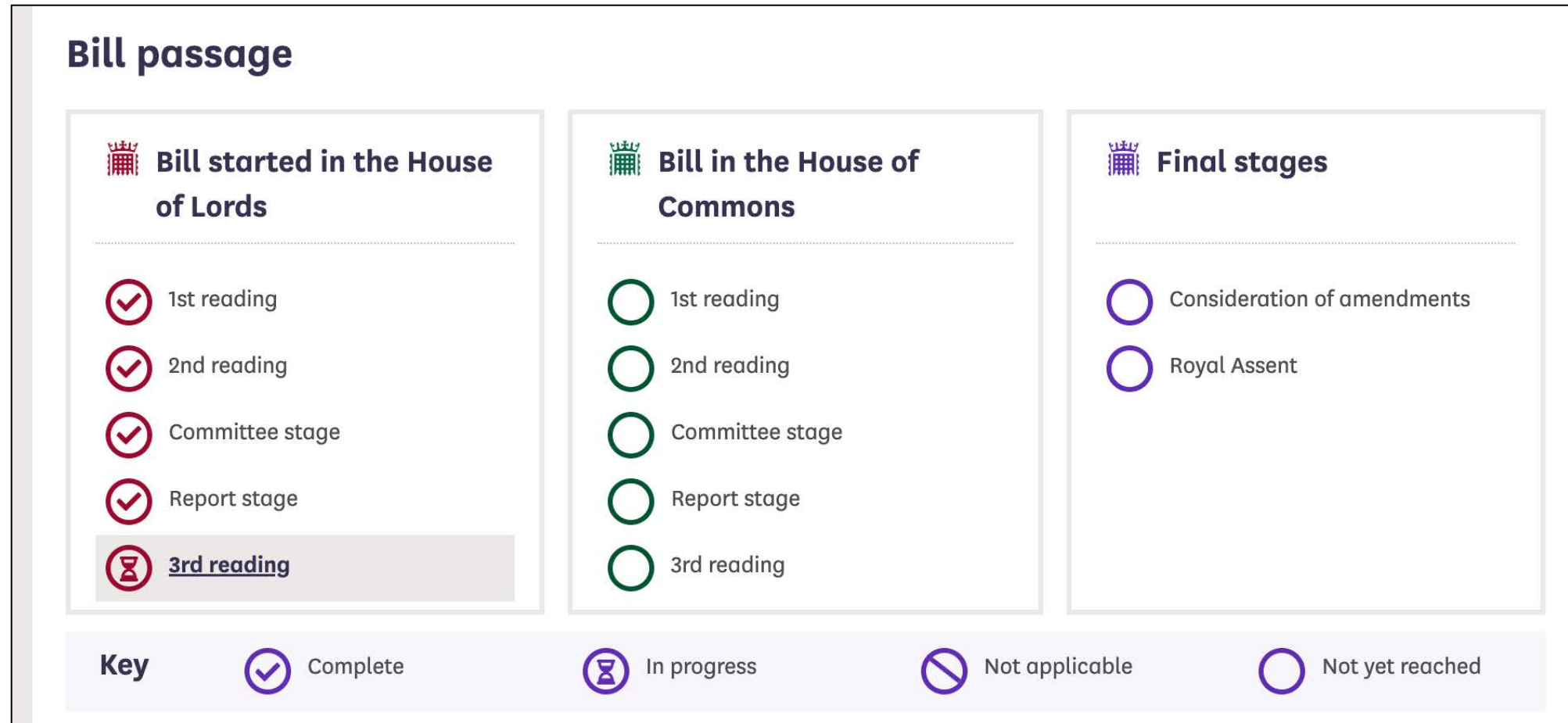


Source: WHO Facebook page

UK air pollution targets making its way through parliament

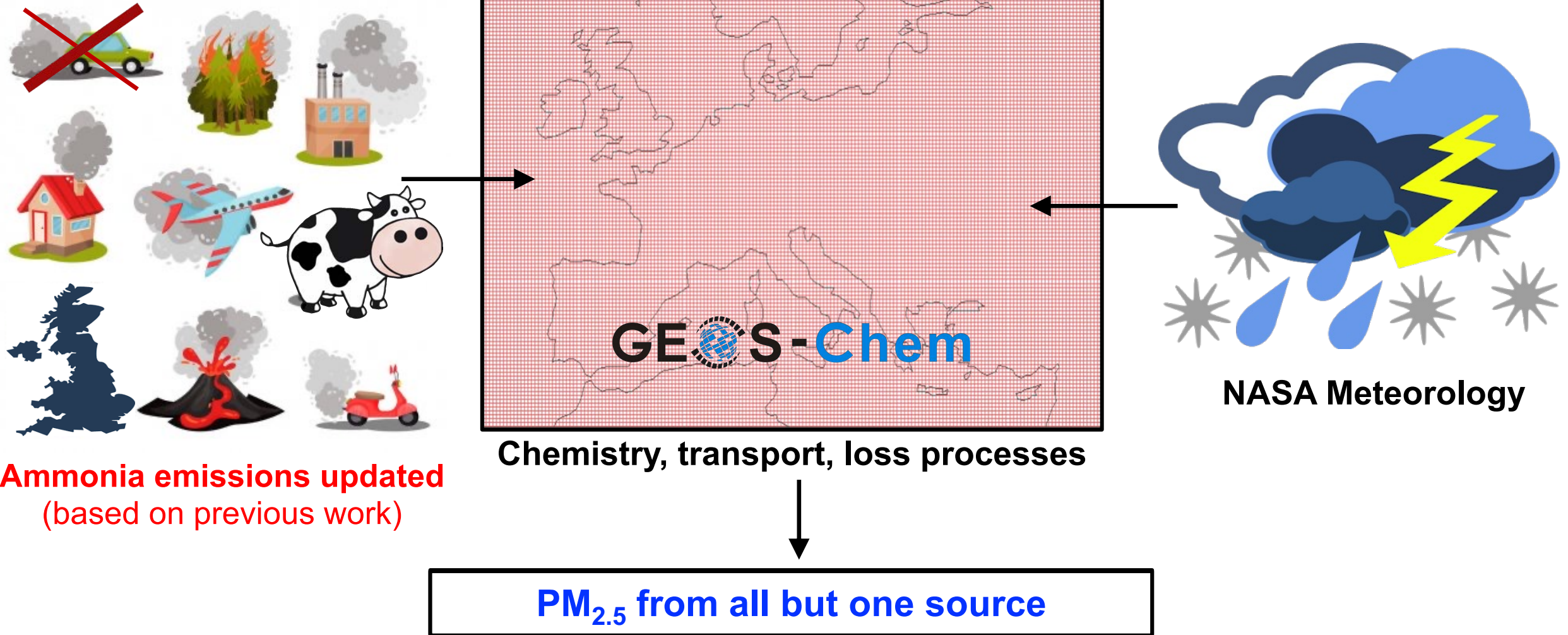
Pretty meek target: “PM_{2.5} to be less than or equal to 10 µg m⁻³ by 1 January 2030.”

Status at 9am yesterday (<https://bills.parliament.uk/bills/3161>)



Simulate PM_{2.5} with GEOS-Chem

3D Atmospheric Chemistry Transport Model



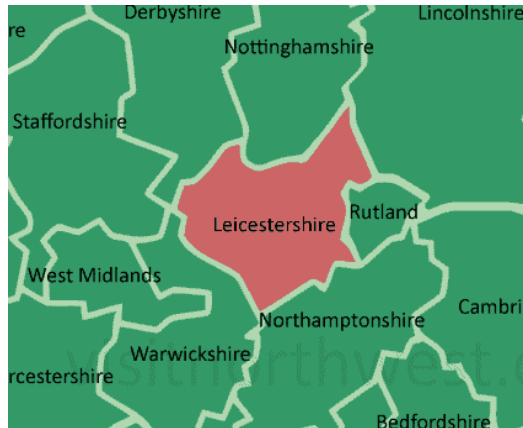
GEOS-Chem manual: <http://acmg.seas.harvard.edu/geos/>

Test Contribution of Potentially Influential Sources

Local



City



County

National



Nearby large cities

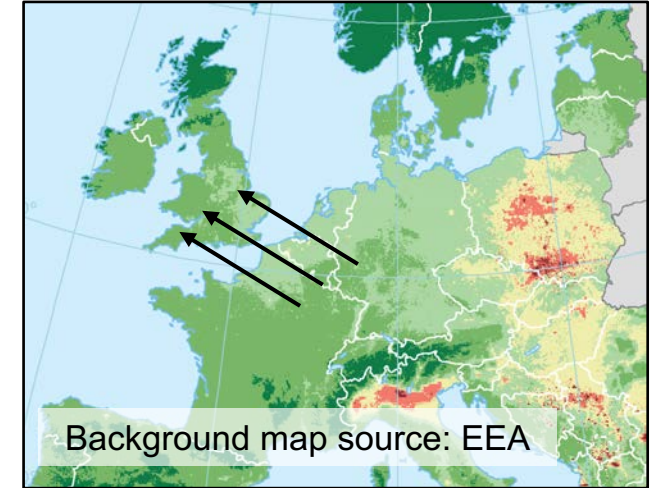


Transport



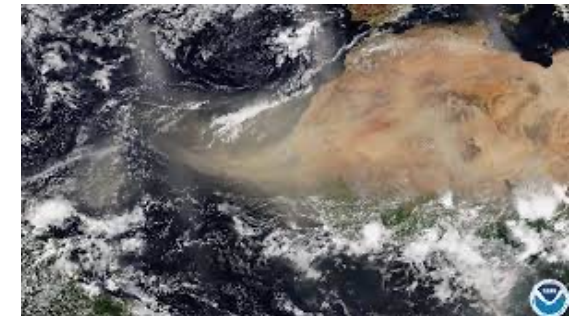
Agriculture

Regional



Mainland Europe

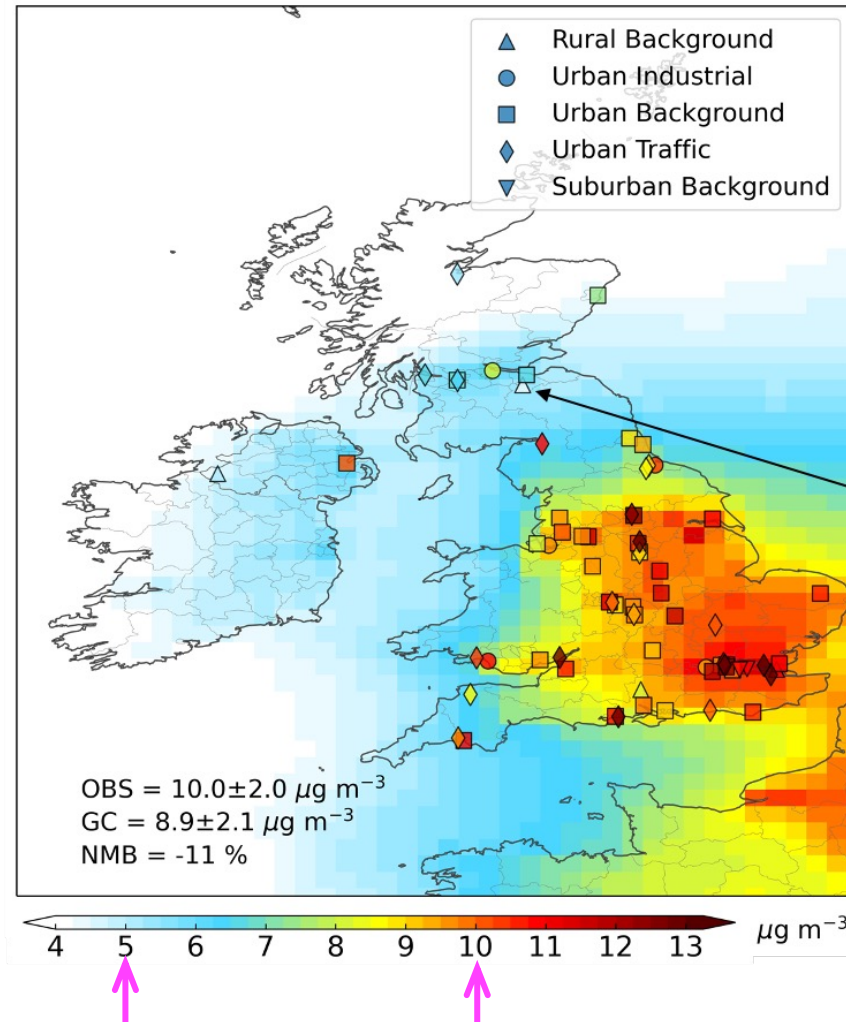
Global



Desert Dust

Assess Validity of Model using Permanent Networks

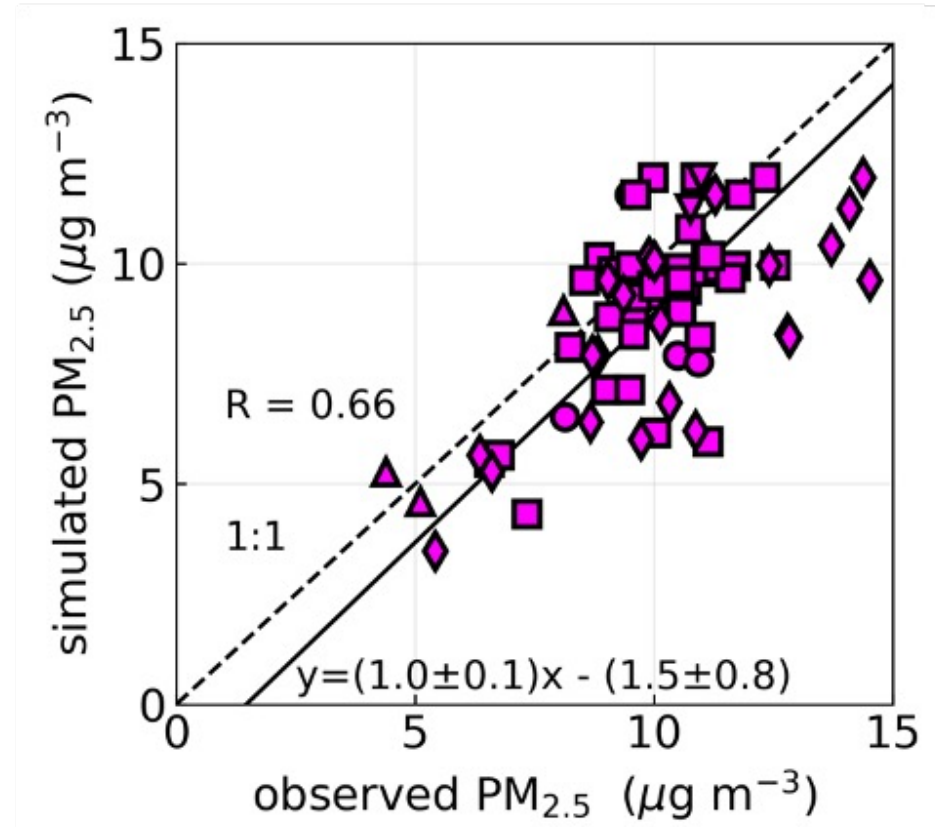
Use total PM_{2.5} observations from the Automatic Urban and Rural Network (AURN) to assess model



WHO (2021) WHO (2005), new UK target

74% of UK exceeds updated WHO guideline

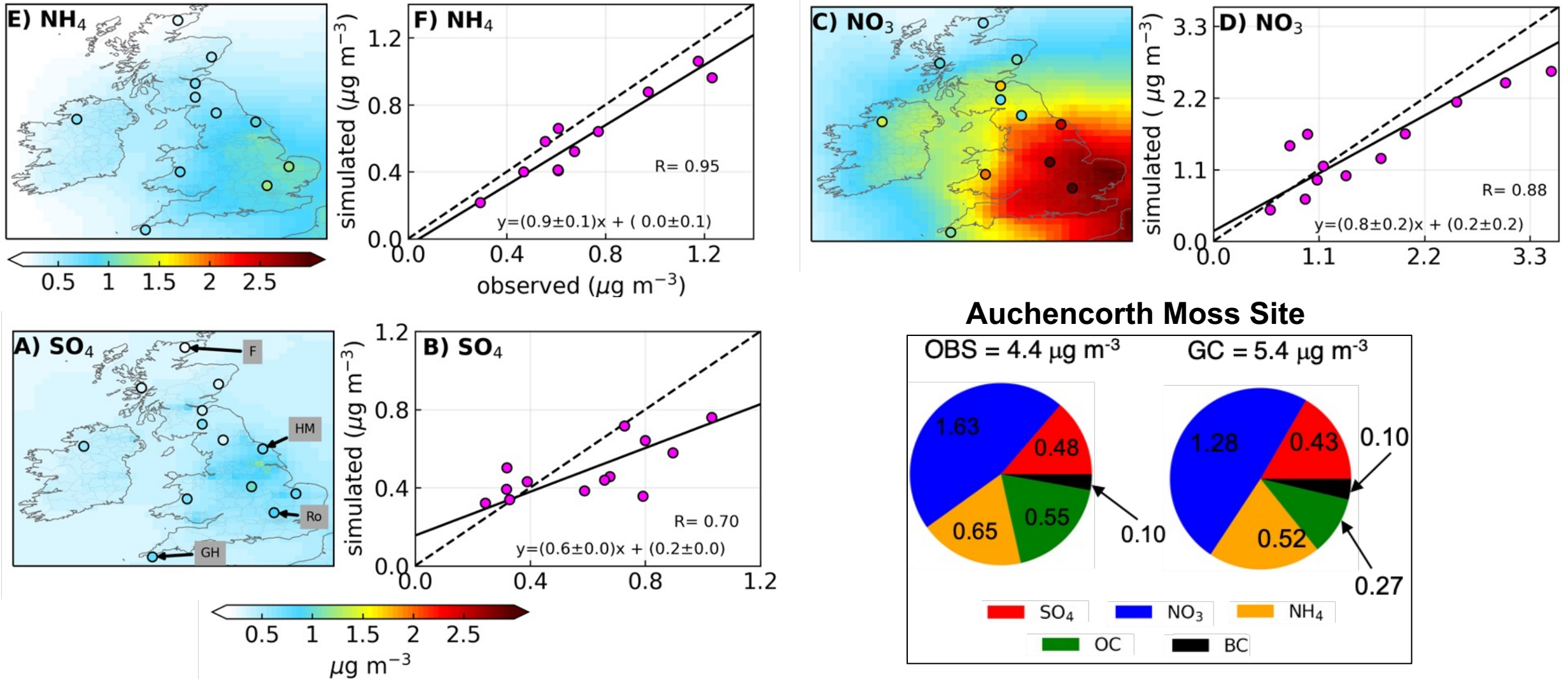
Compare annual mean surface concentrations of PM_{2.5} for 2019



Consistent spatial pattern ($R = 0.66$) and variance (slope = 1.0). Model 11% less than observations

Assess Validity of Model using Permanent Networks

Use PM_{2.5} composition measurements from UKEAP and EMEP sites to assess model

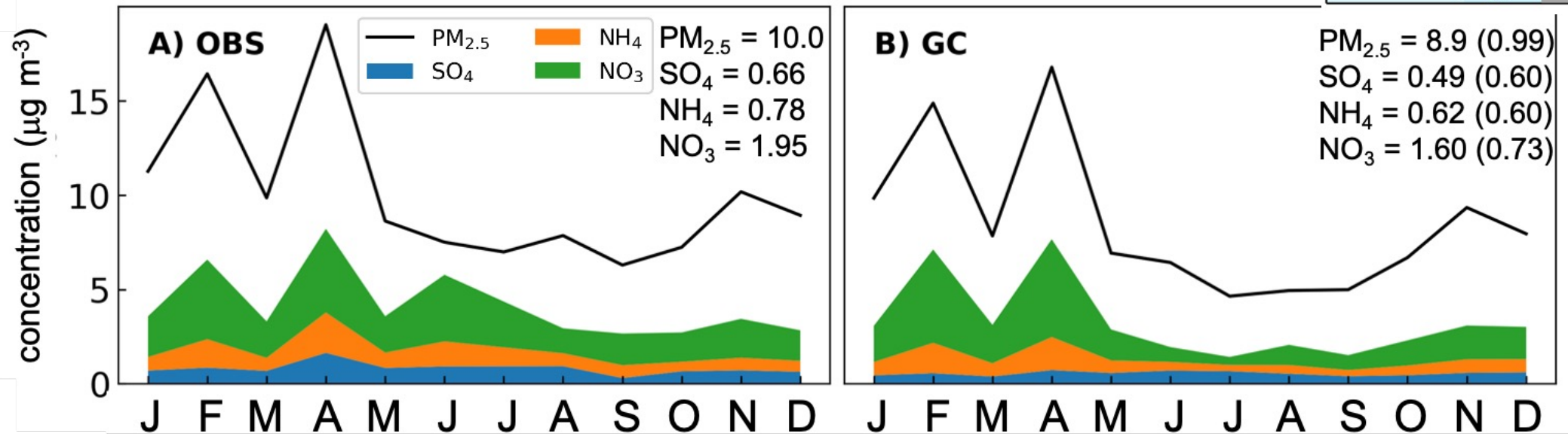
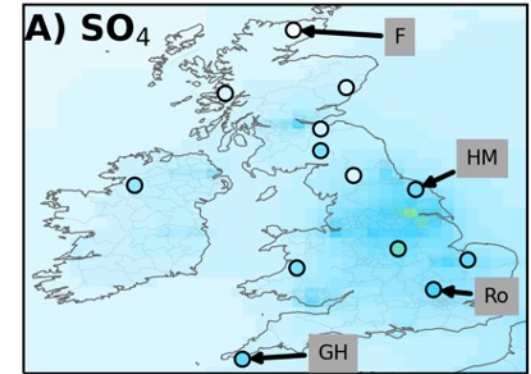


Model underpredicts observed (sulfate, nitrate, ammonium) and possibly overpredicts unobserved (dust) components. Model captures variance of components from NO_x (nitrate) and ammonia (ammonium)

Assess Validity of Model using Reference Monitors

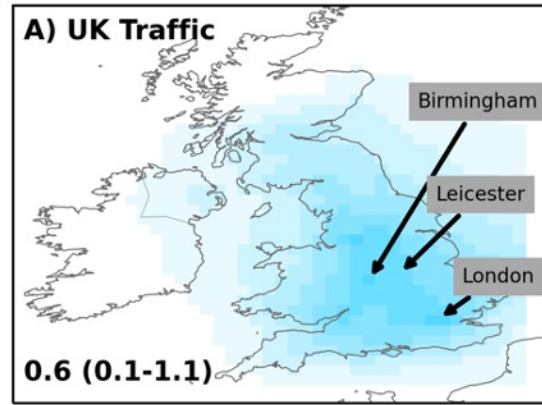
Also evaluate model skill at reproducing observed seasonality in $\text{PM}_{2.5}$

SO_4 : sulfate; NO_3 : nitrate; NH_4 : ammonium

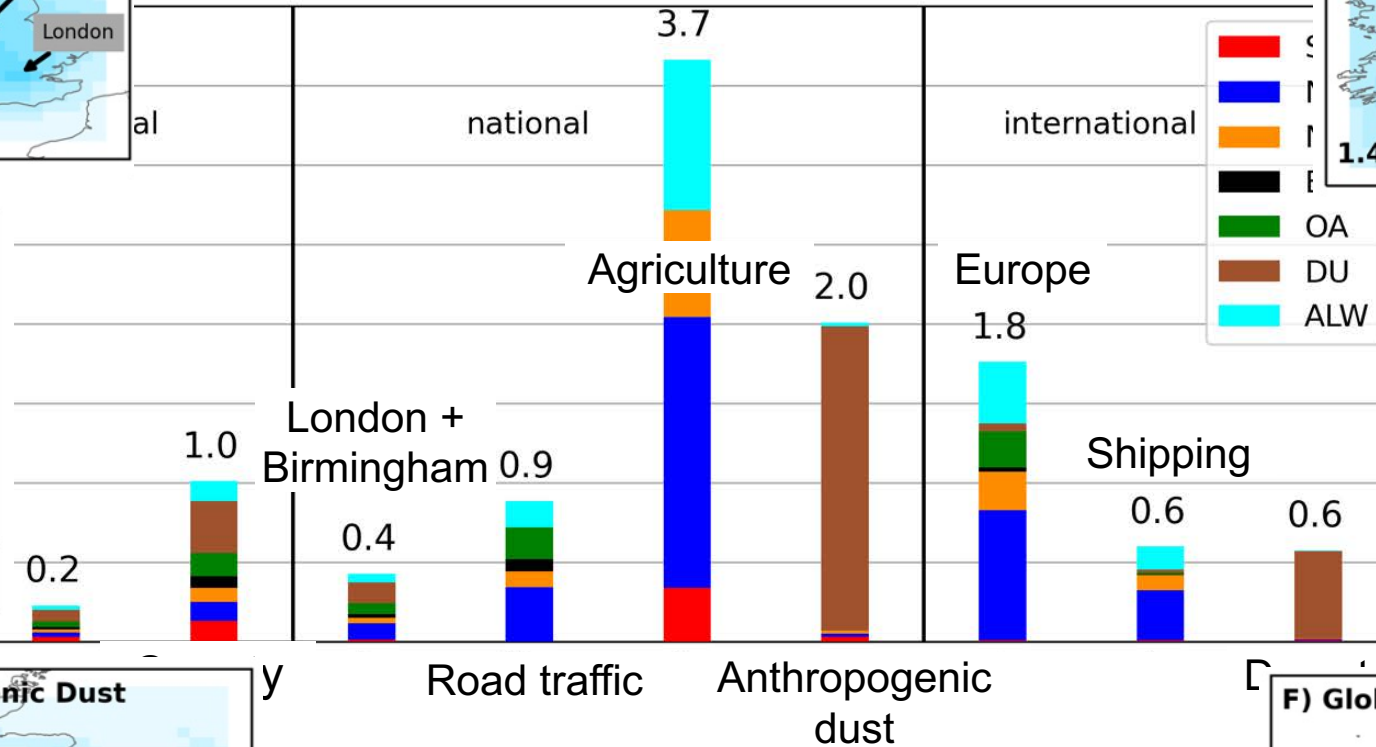
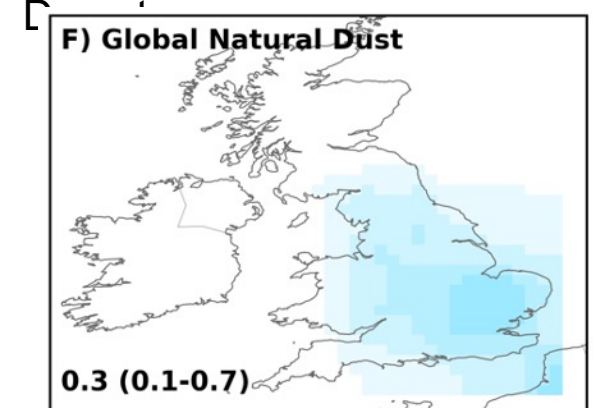
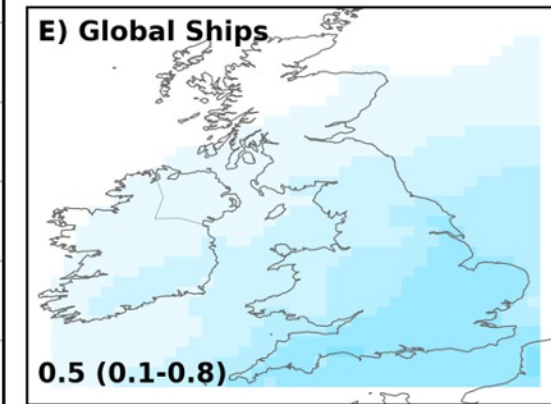
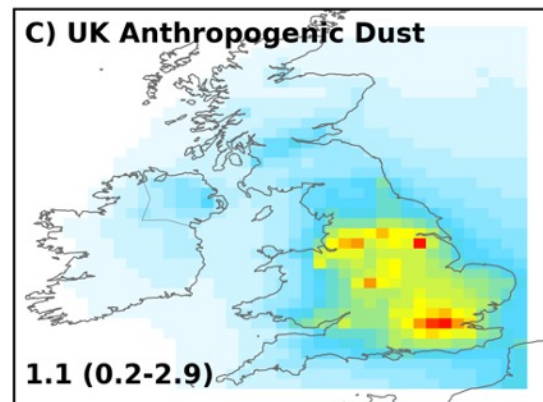
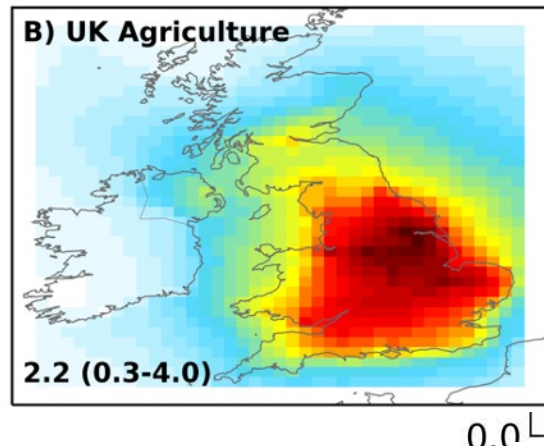
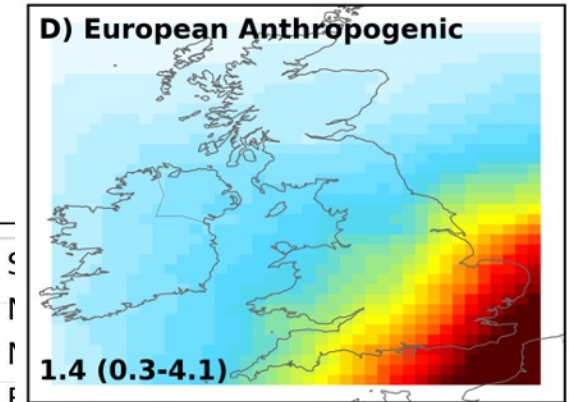


Enhancements in cold months and when ammonia emissions from agriculture peak due to application of synthetic fertilizer in March-April

Contribution of Sources to annual PM_{2.5} in Leicester



SO₄: sulfate; **NO₃**: nitrate; **NH₄**: ammonium
BC: black carbon; **OC**: organic carbon; **DU**: dust



Colour scale for maps of PM_{2.5} in µg m⁻³



Support for Small Local Contribution to PM_{2.5}

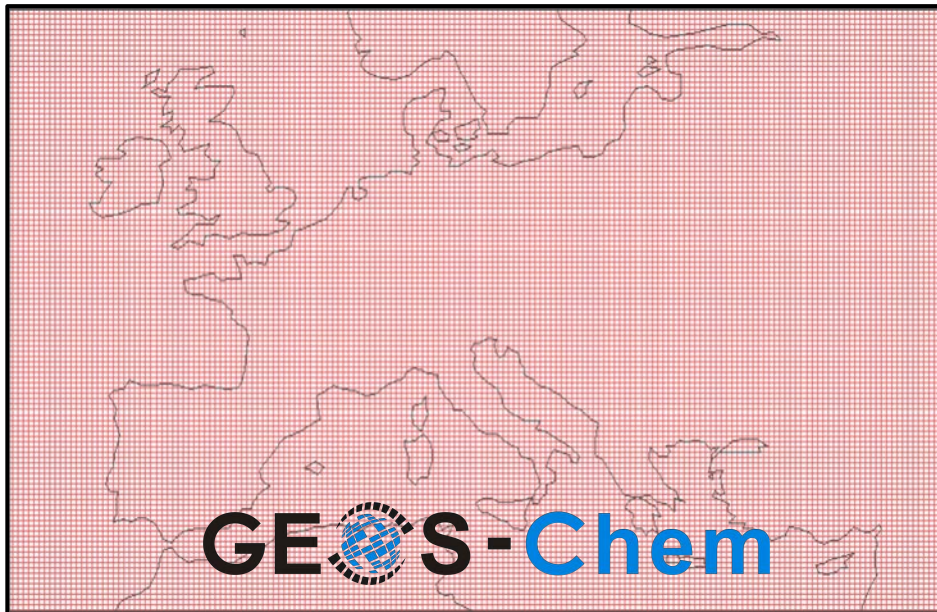
Low-cost network of Zephyr® sensors distributed throughout Leicester since November 2020

AURN network

GEOS-Chem model

EarthSense low-cost sensors

Jan 2019 Jul 2019 Jan 2020 Jul 2020 Jan 2021 Present

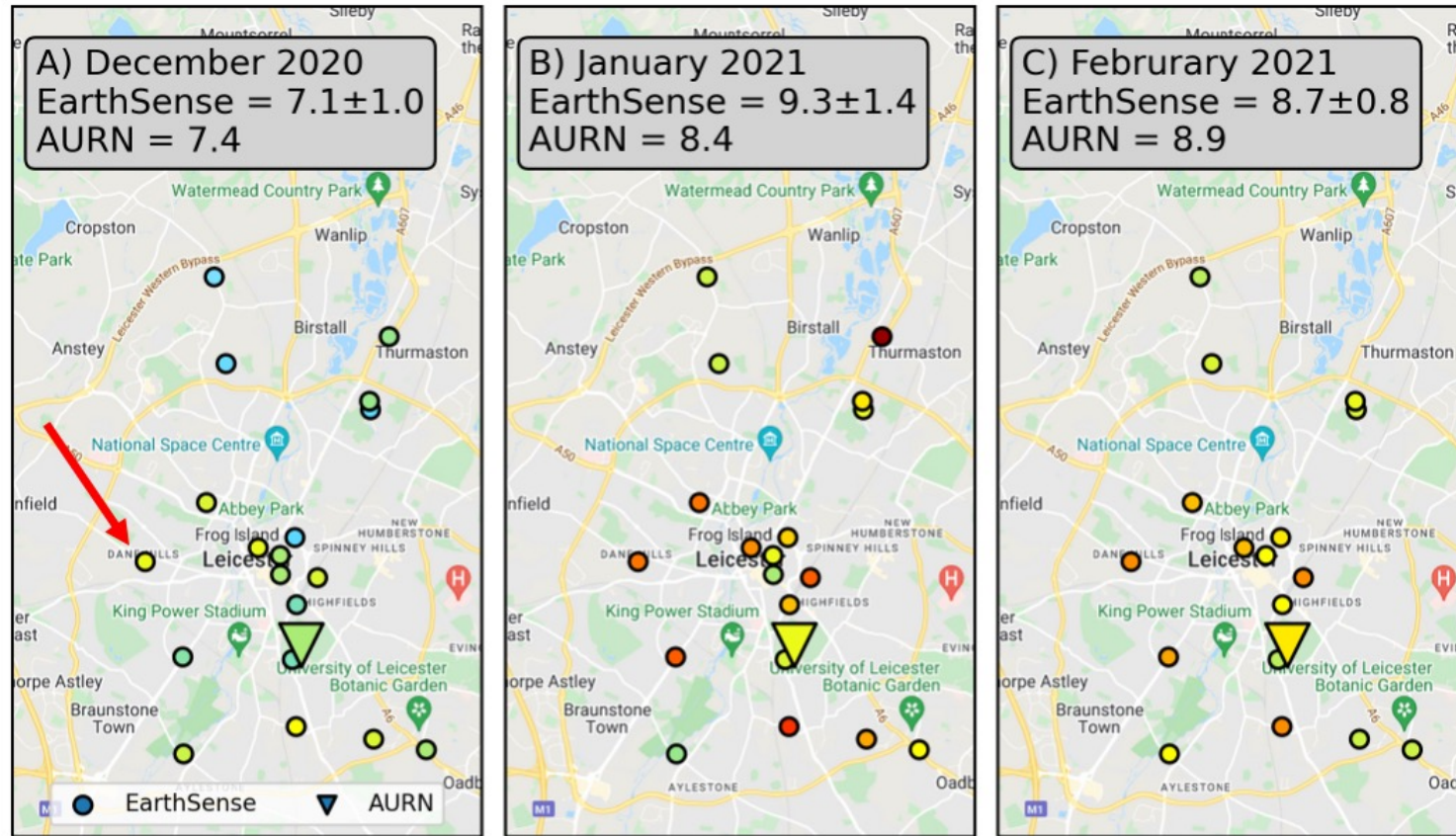


Zephyr Low-Cost Sensor Network Across Leicester

December 2020

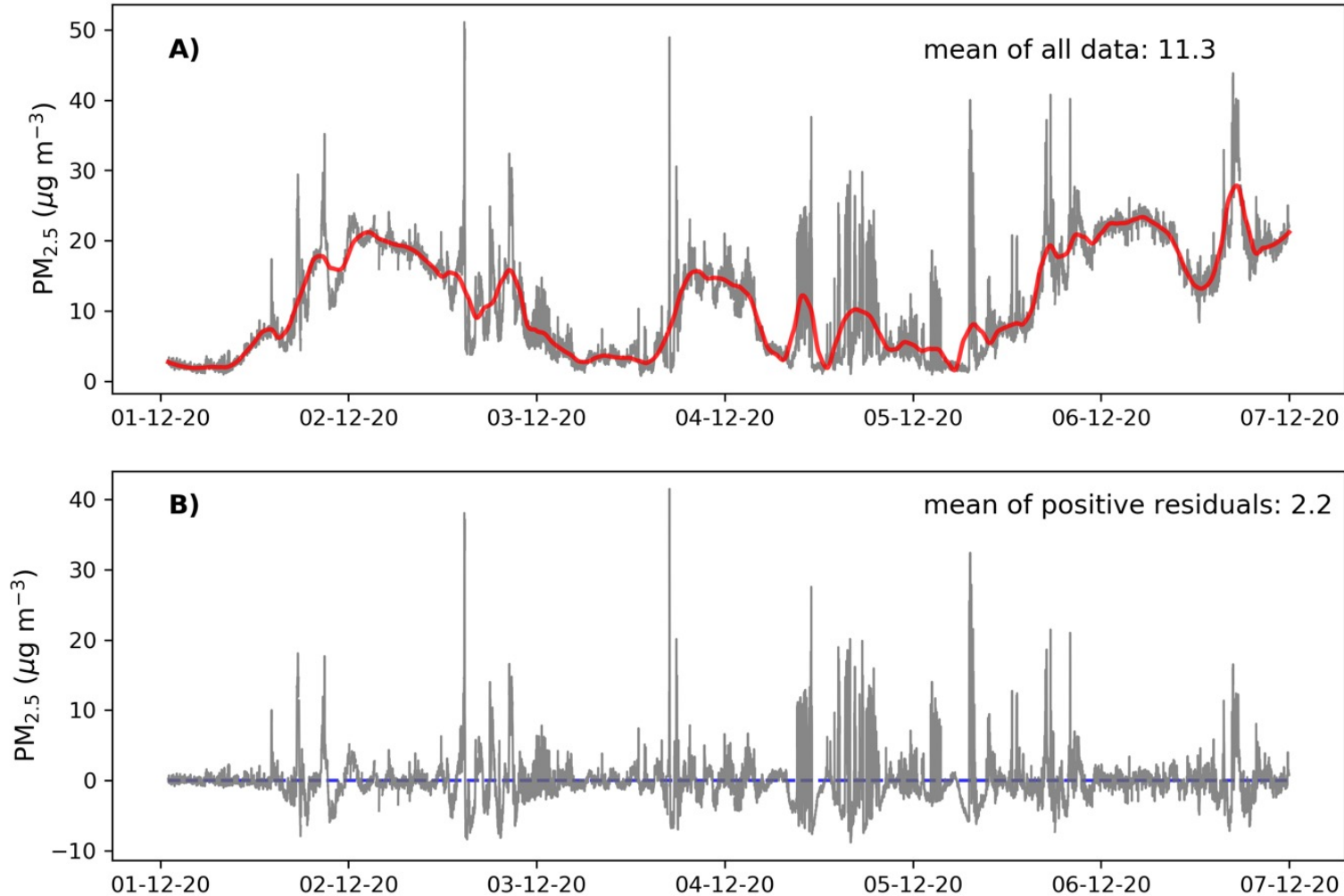
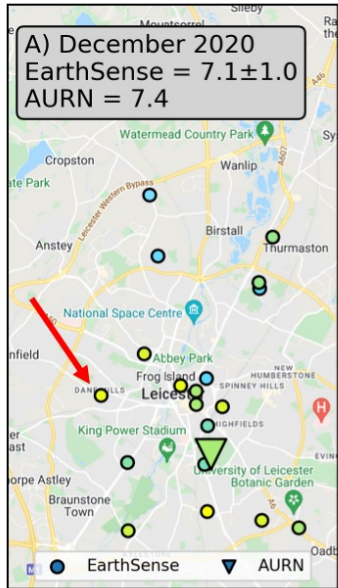
January 2021

February 2021



Zephyr Low-Cost Sensor Network Across Leicester

Decompose data into local and background contributions

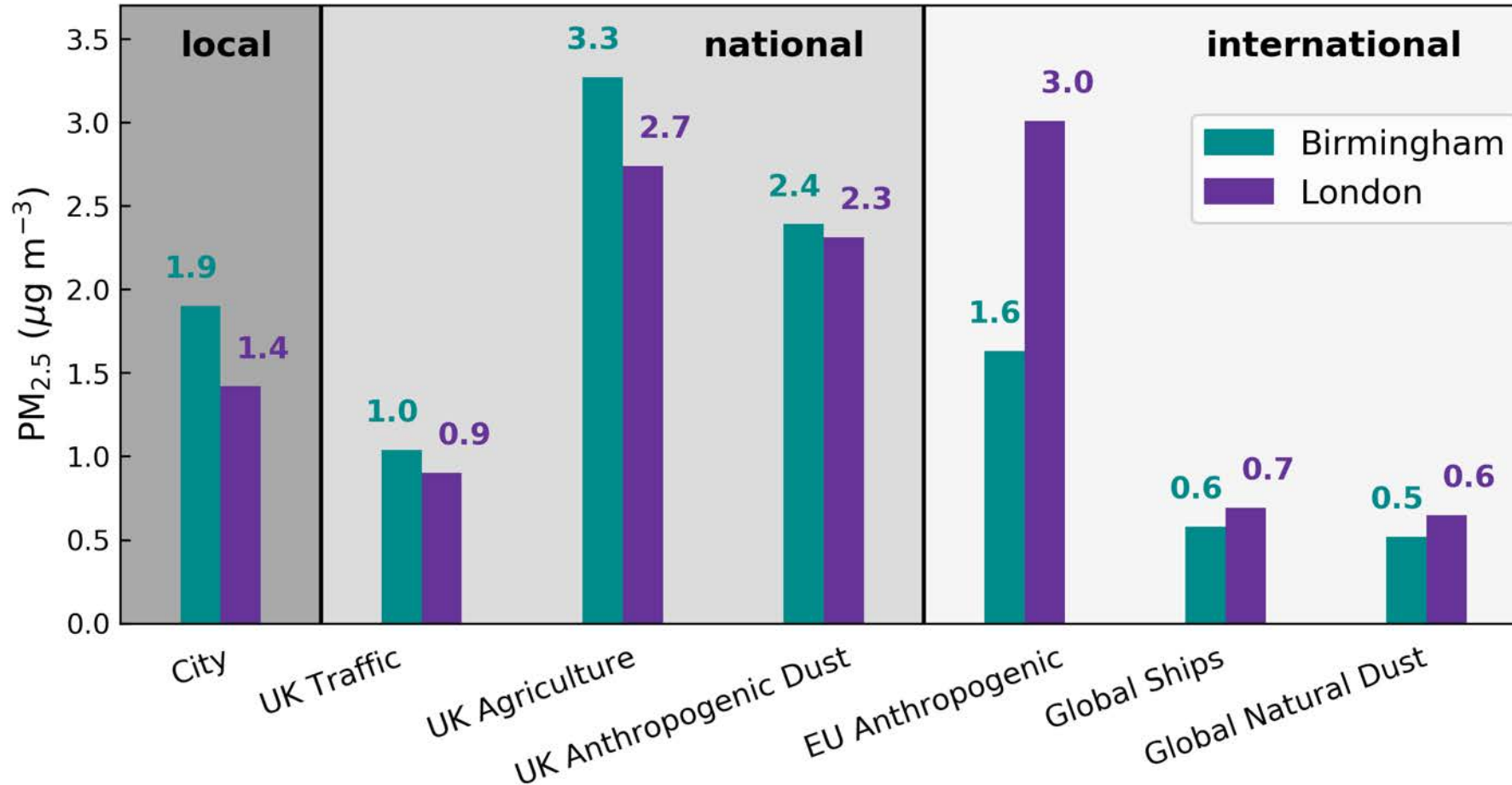


Red: Background

Residuals > 0: Local

According to low-cost sensors, local sources contribute **4-10%**. Small like the model (**2%**)

Results for Large Cities like London and Birmingham



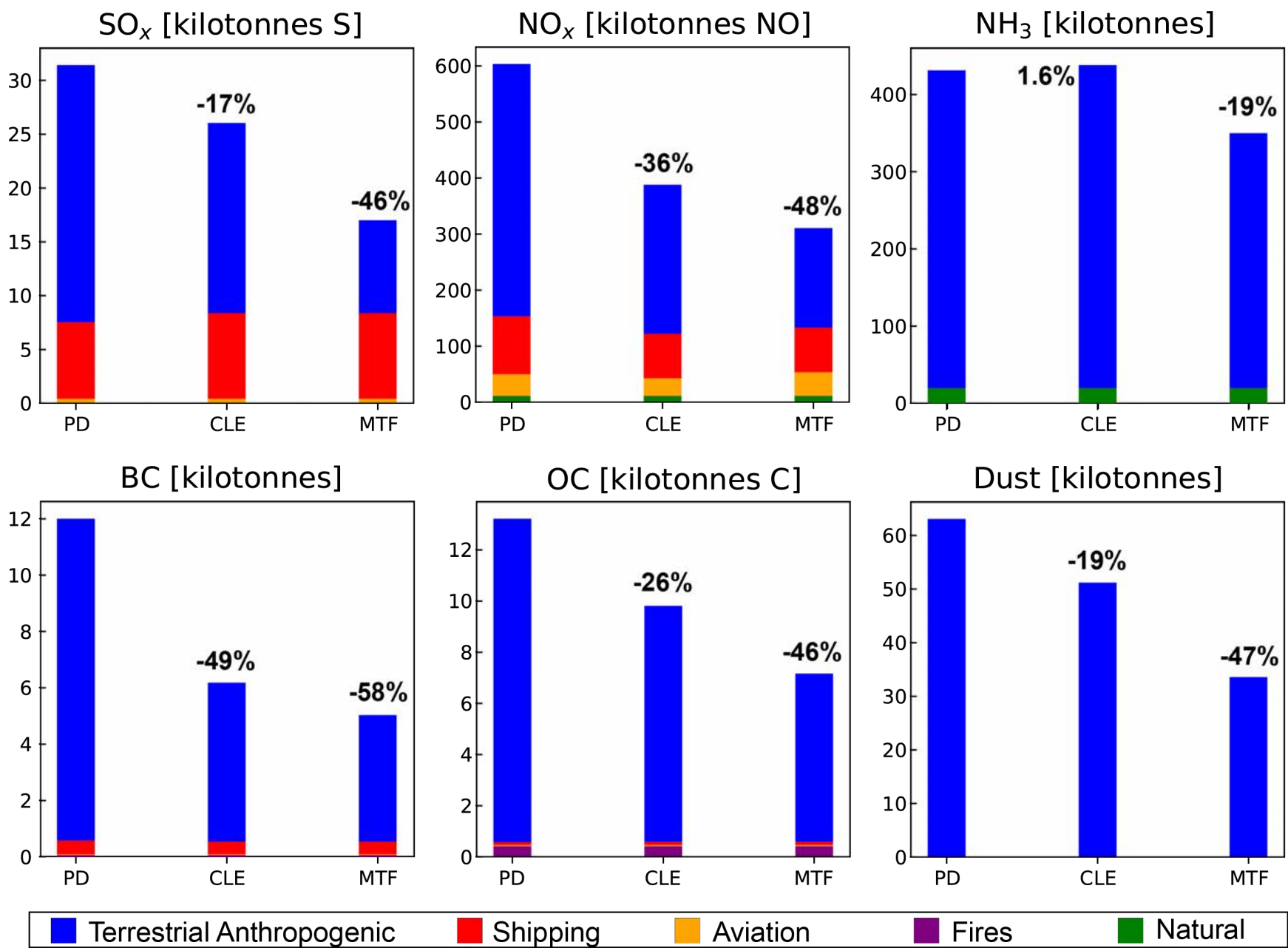
London: 1,600 km²
Birmingham: 270 km²
Leicester: 70 km²

Lower local than rural agricultural ammonia contribution consistent with Leicester

Air quality and health benefits of current legislation vs technically feasible solutions



PM_{2.5} precursor emissions for implementation in GEOS-Chem

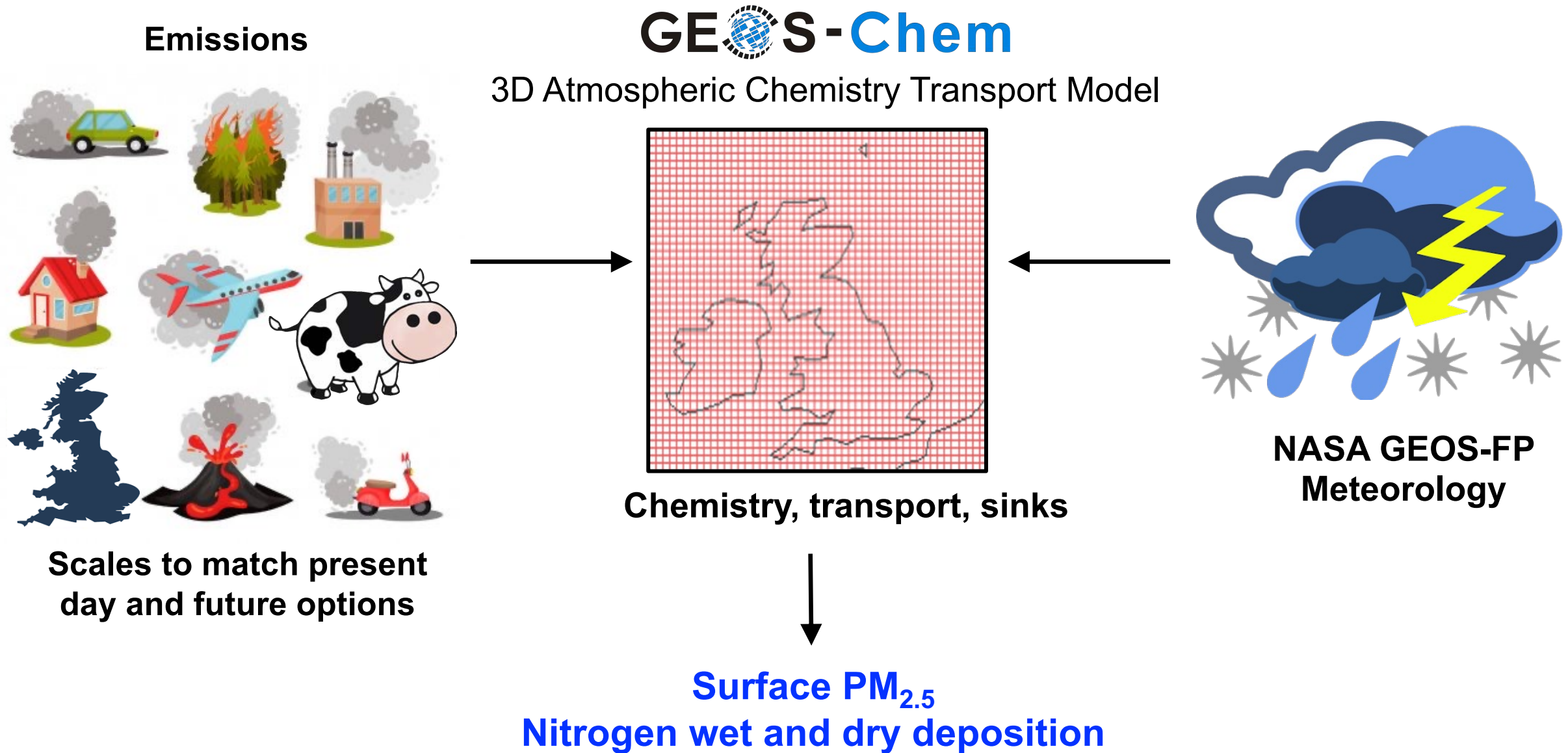


PD: present-day (2019)
Future (2030) emissions:
CLE: current legislation
MTF: max. technically feasible

NH₃ leading to aerosol ammonium (NH₄) increases under current legislation

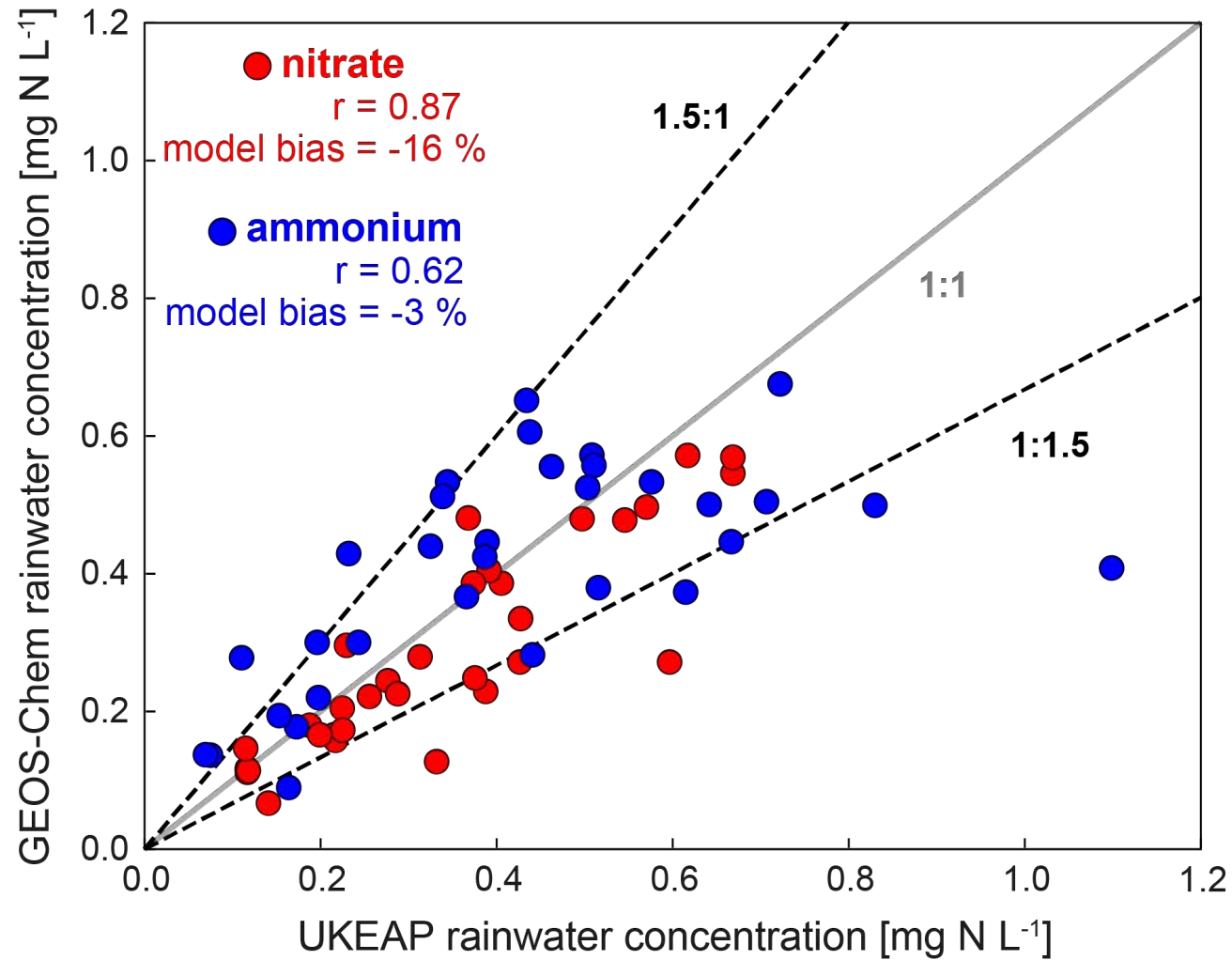
Technically feasible solutions include low nitrogen feed, covered manure storage, improved manure spreading, air filters and scrubbers, and move from urea-based fertilizer

Model emissions impacts on PM_{2.5} and nitrogen deposition



Assessment of deposition

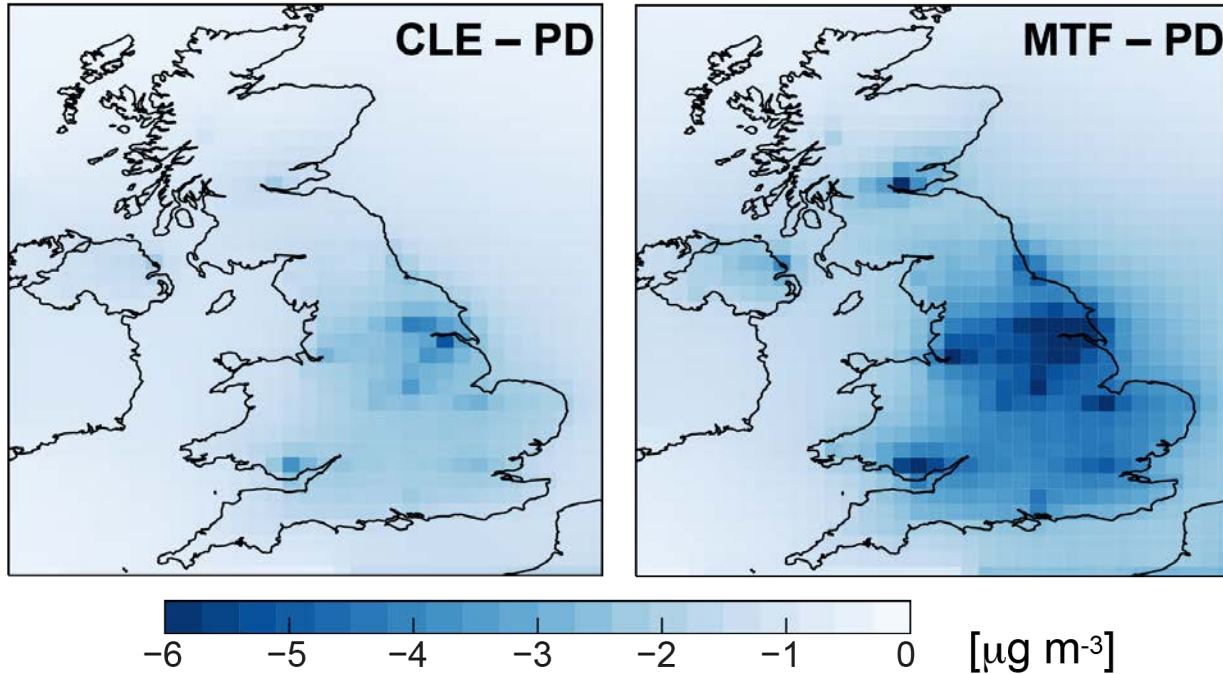
Rainwater nitrate and ammonium



Includes correction to model (GEOS-FP) underestimate in rainwater volume

Air Quality Impacts of Future Mitigation Measures

Change in $\text{PM}_{2.5}$ relative to the present-day due to emission controls



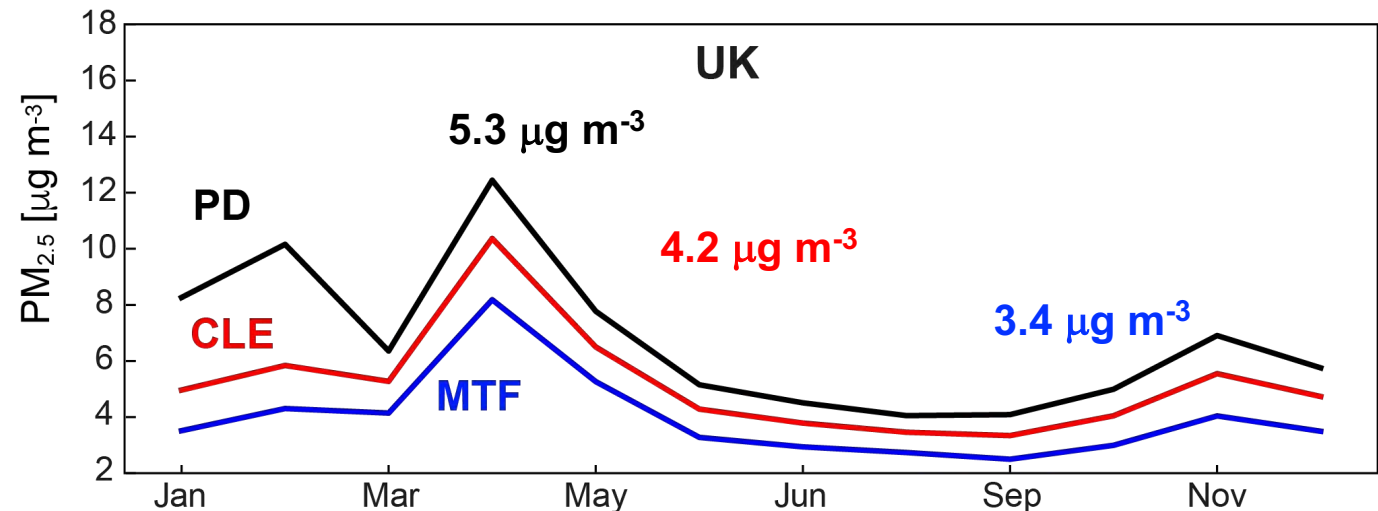
Average decline in $\text{PM}_{2.5}$ in the UK of 21% with current legislation and 36% with maximum technically feasible options

Decline greatest over densely populated urban areas and North Yorkshire (coal-fired power plants)

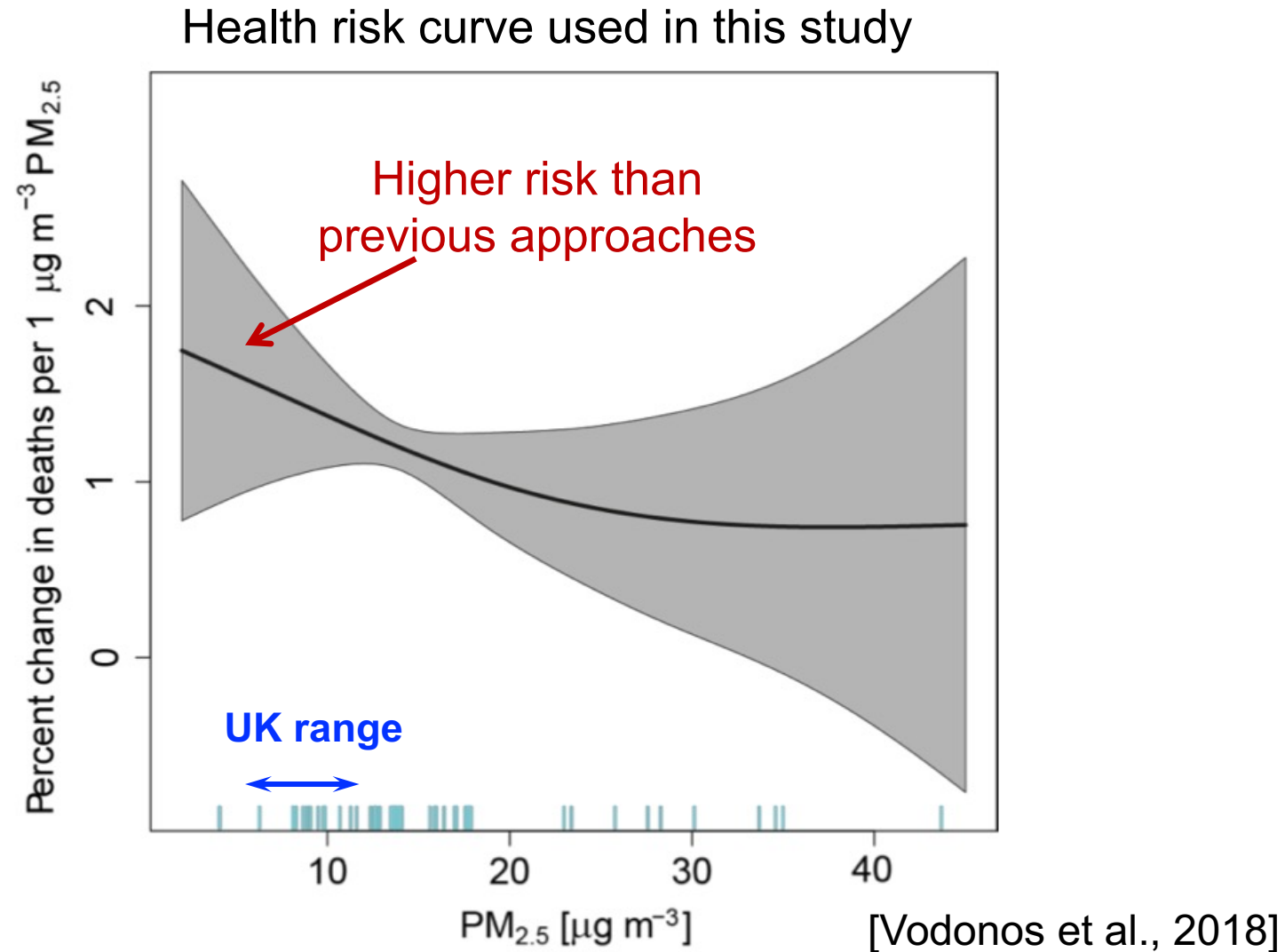
Percent UK grids above the updated WHO guideline ($5 \mu\text{g m}^{-3}$) decreases from 48% in PD to 32% for CLE, and 2% for MTF

Similar decline in $\text{PM}_{2.5}$ in all months, except Jan-Feb.

Reduces vulnerability to interannual variability in meteorology (dry, stable conditions)

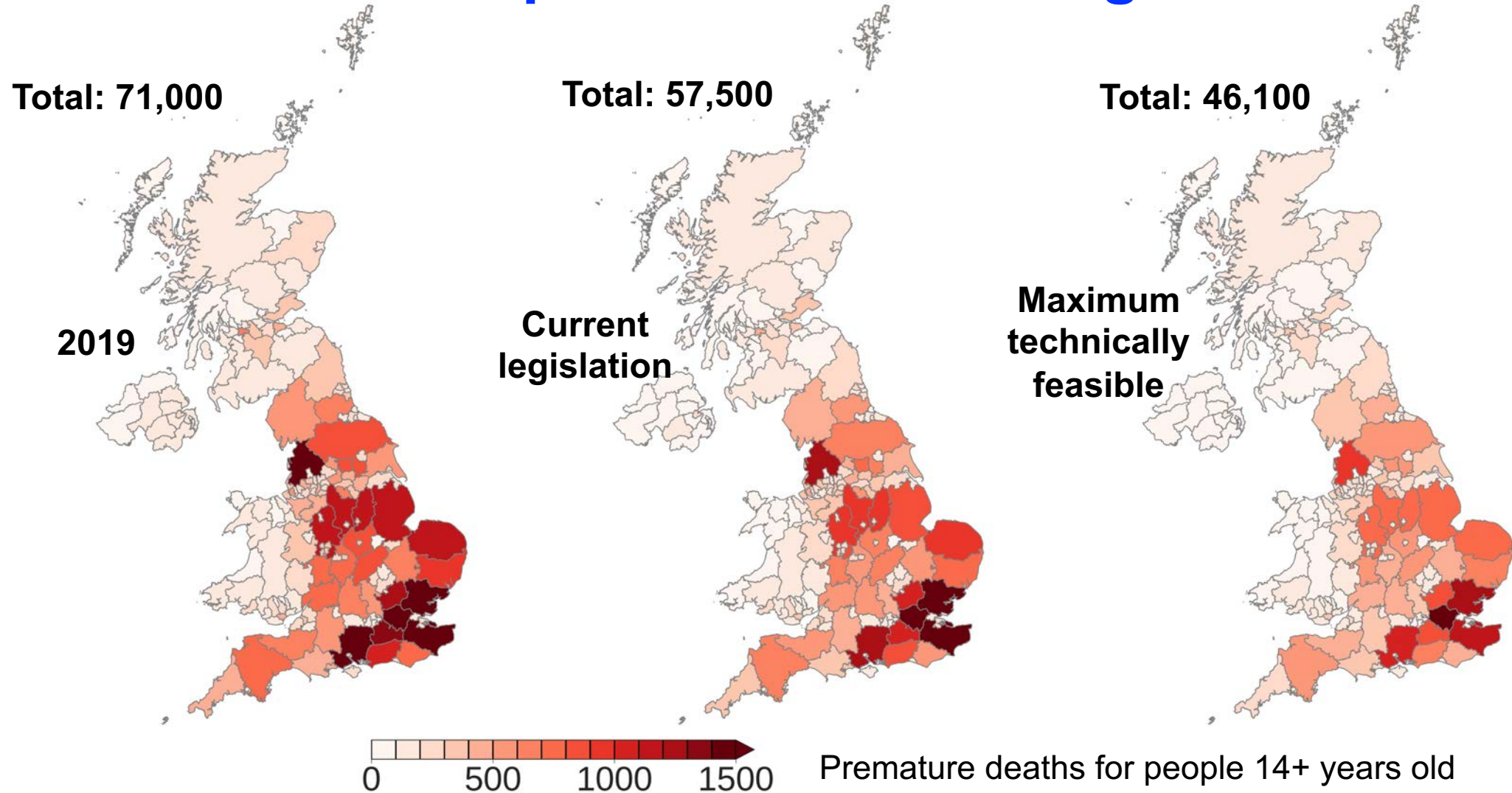


Public Health Impacts of Future Mitigation Measures



Yields more excess deaths than Public Health England 2014 study due to differences in relative risks

Public Health Impacts of Future Mitigation Measures



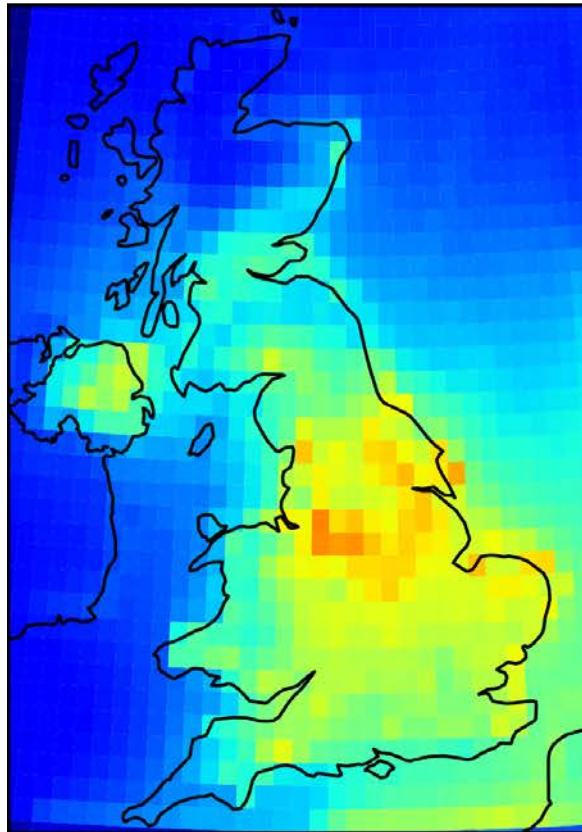
Excess deaths from $PM_{2.5}$ in 2019 of ~71,000 (PHE report for 2010 was ~29,000).

Avoided deaths of ~13,000 for current legislation and of ~25,000 if adopt best solutions available

Ecosystem Impacts of Future Mitigation Measures

Reduced (ammonium) and oxidized (nitrate) nitrogen deposition today and likely in the future

Total N deposition in 2019



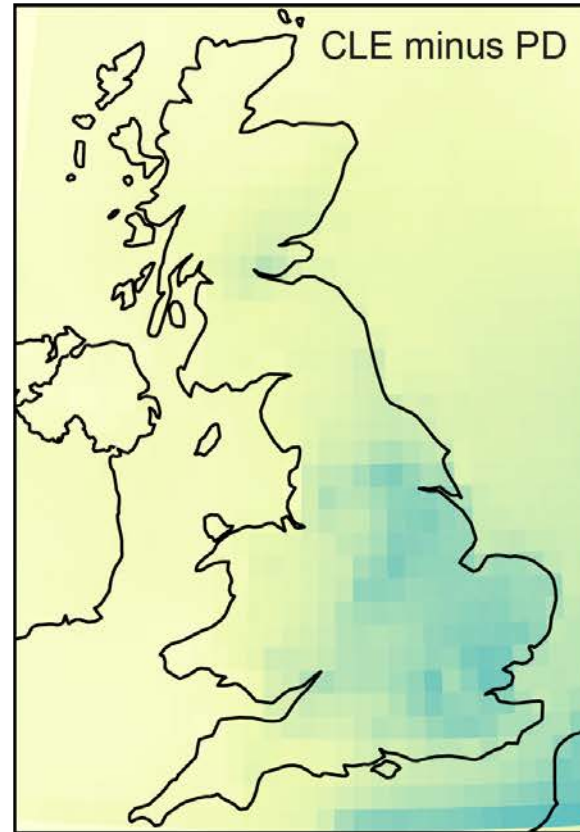
0 4 8 12 16 [kg N ha⁻¹]

Total: **429 Gg N**

Ammonium (NH_x): **252 Gg**

Nitrate (NIT): **177 Gg**

Decline in total annual N deposition due to emission controls

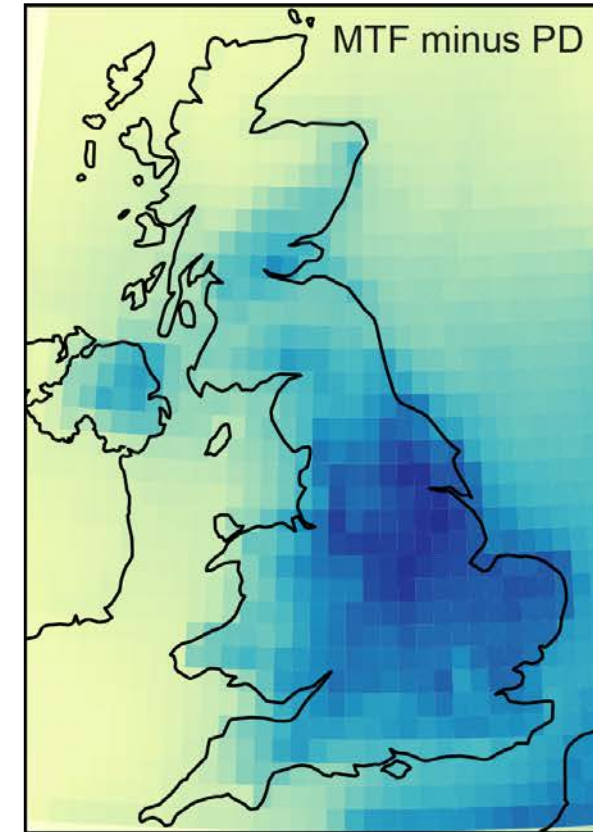


-4 -3 -2 -1 0 [kg N ha⁻¹]

48 Gg decrease in total

5 Gg increase in NH_x

53 Gg decrease in NIT



116 Gg decrease in total

42 Gg decrease in NH_x

74 Gg decrease in NIT

Concluding Remarks

- Satellite columns effective for estimating top-down ammonia emissions
- Suggest underestimate in dairy cattle ammonia emissions in summer; support springtime peak from fertilizer application
- Discrepancy in dairy cattle ammonia emissions needs to be resolved
- Rural ammonia emissions dominant contributor to $\text{PM}_{2.5}$ in all UK cities
- City sources make modest contribution to urban $\text{PM}_{2.5}$, supported by network of low-cost sensors, so local solutions alone insufficient
- Current legislation only marginally effective at reducing $\text{PM}_{2.5}$
- Adoption of maximum technically feasible solutions yields health burden reductions 2-times greater than current legislation
- Potentially large ecosystem benefits too, though still to quantify exceedances of nitrogen critical load