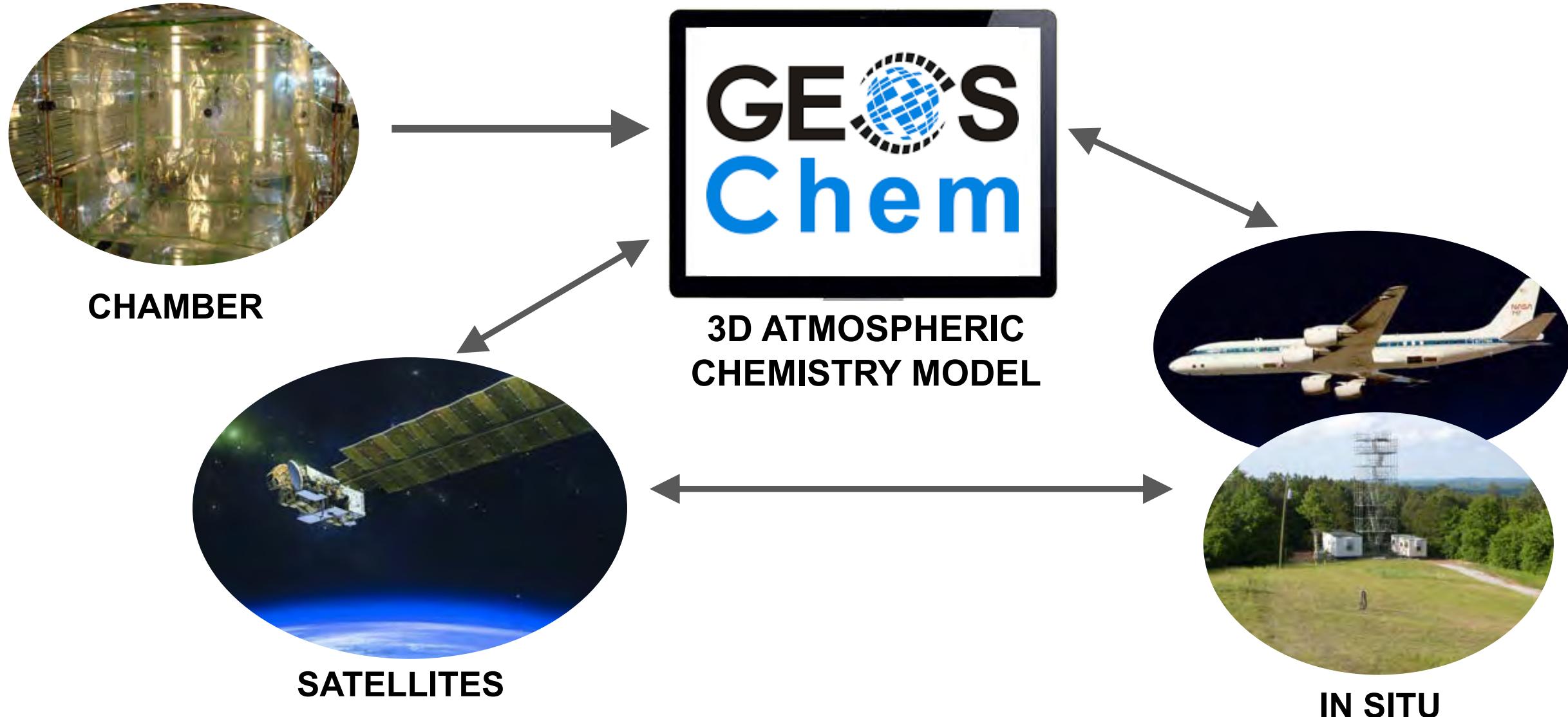


Using satellites to determine pollutant emissions on the ground and atmospheric composition up above



Multiplatform Approach to Solve Pressing Environmental Issues



My group uses and develops state-of-science observations and tools to inform policy.

Research Group Members



Alfred (PhD), Alok (postdoc), Chloe (visiting UG), Gongda (PhD), Karn (PhD), Nana (PhD), Rob (postdoc)

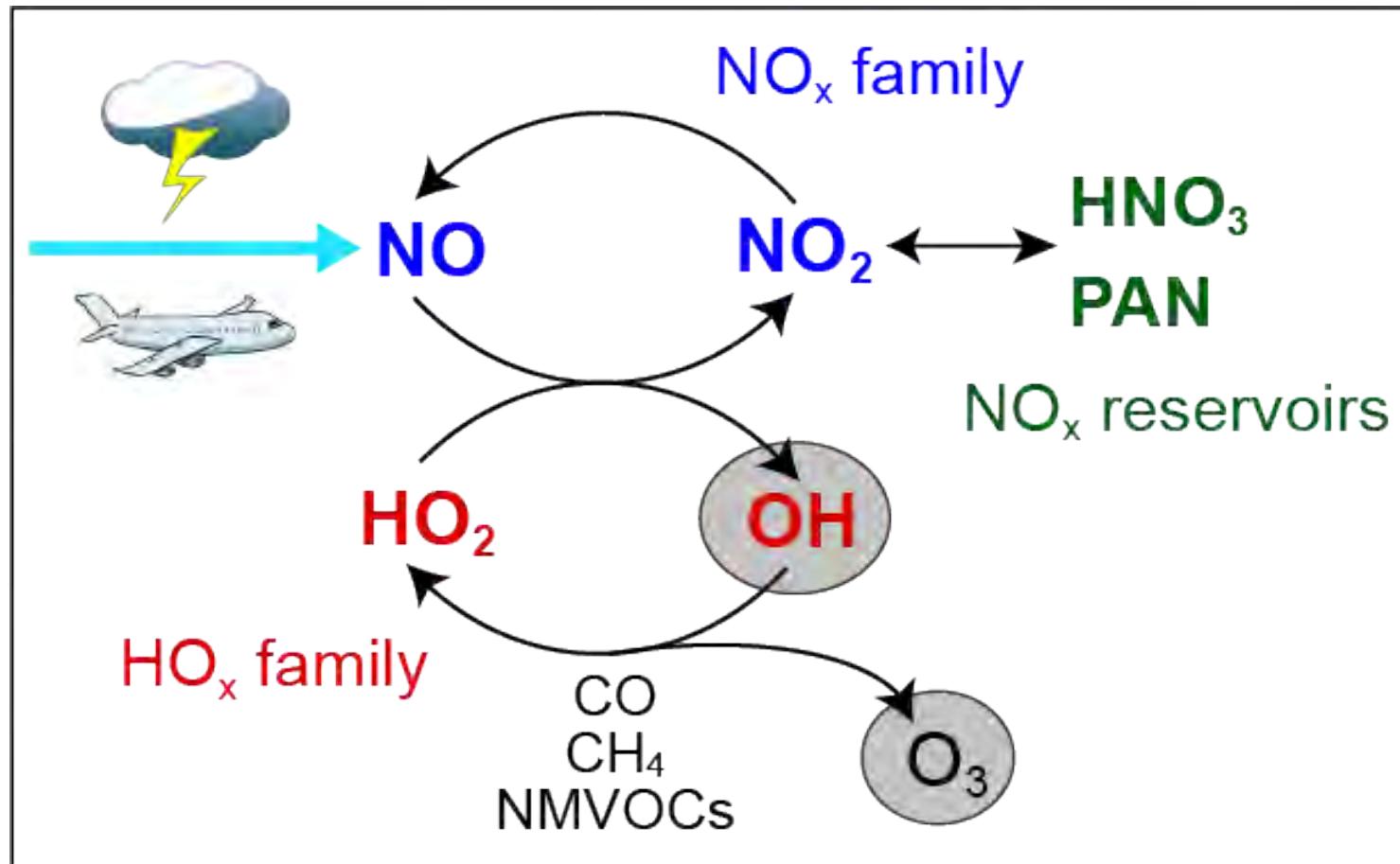
First Estimate of Upper Tropospheric NO₂ from TROPOMI



In review: <https://amt.copernicus.org/preprints/amt-2020-399/>

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)



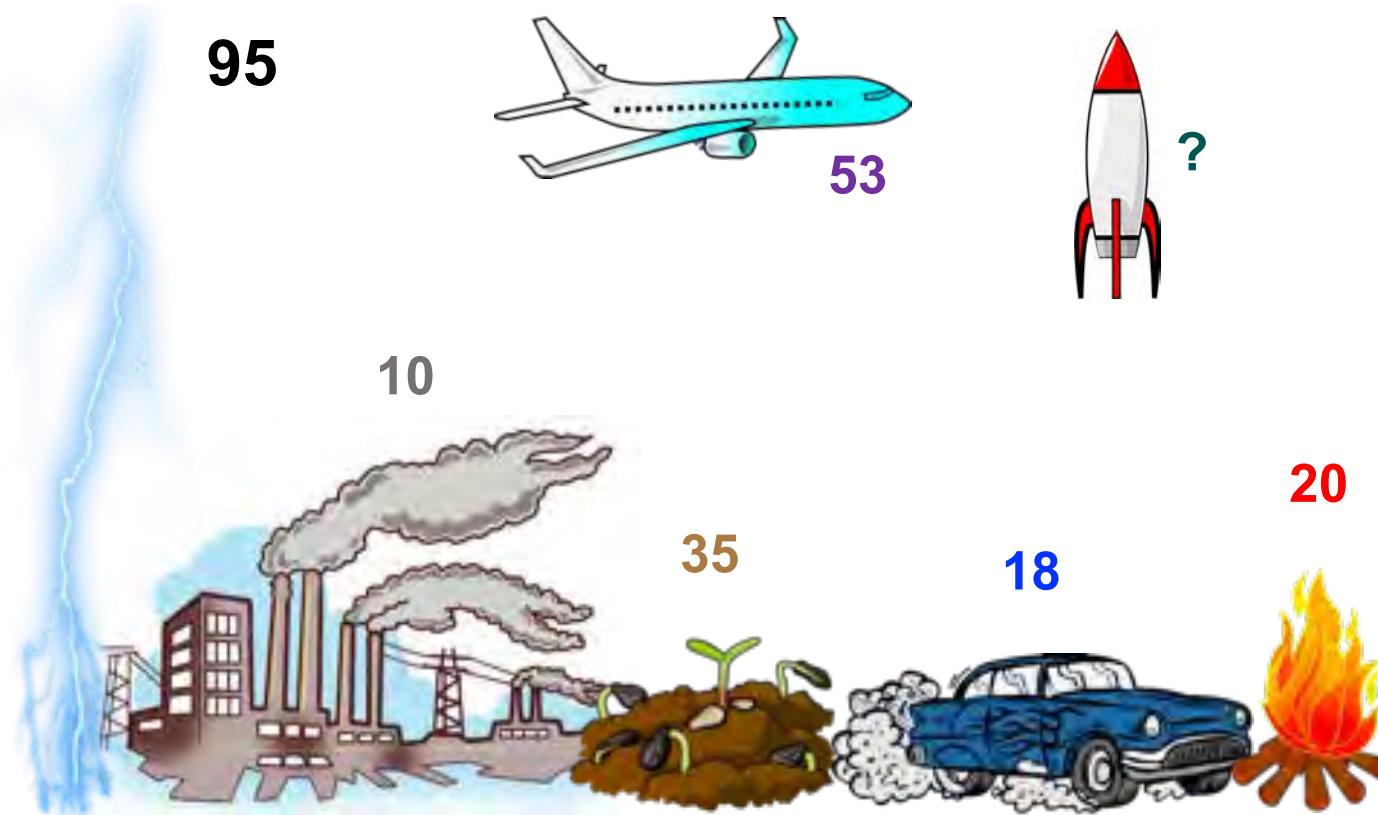
Influence atmospheric oxidants (OH, O₃) and climate (O₃ formation, methane persistence)

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

Influence of upper tropospheric NO_x

NO_x in the upper troposphere is very efficient at producing the greenhouse gas ozone

OPE (molecules O₃/molecule NO_x) for individual NO_x sources



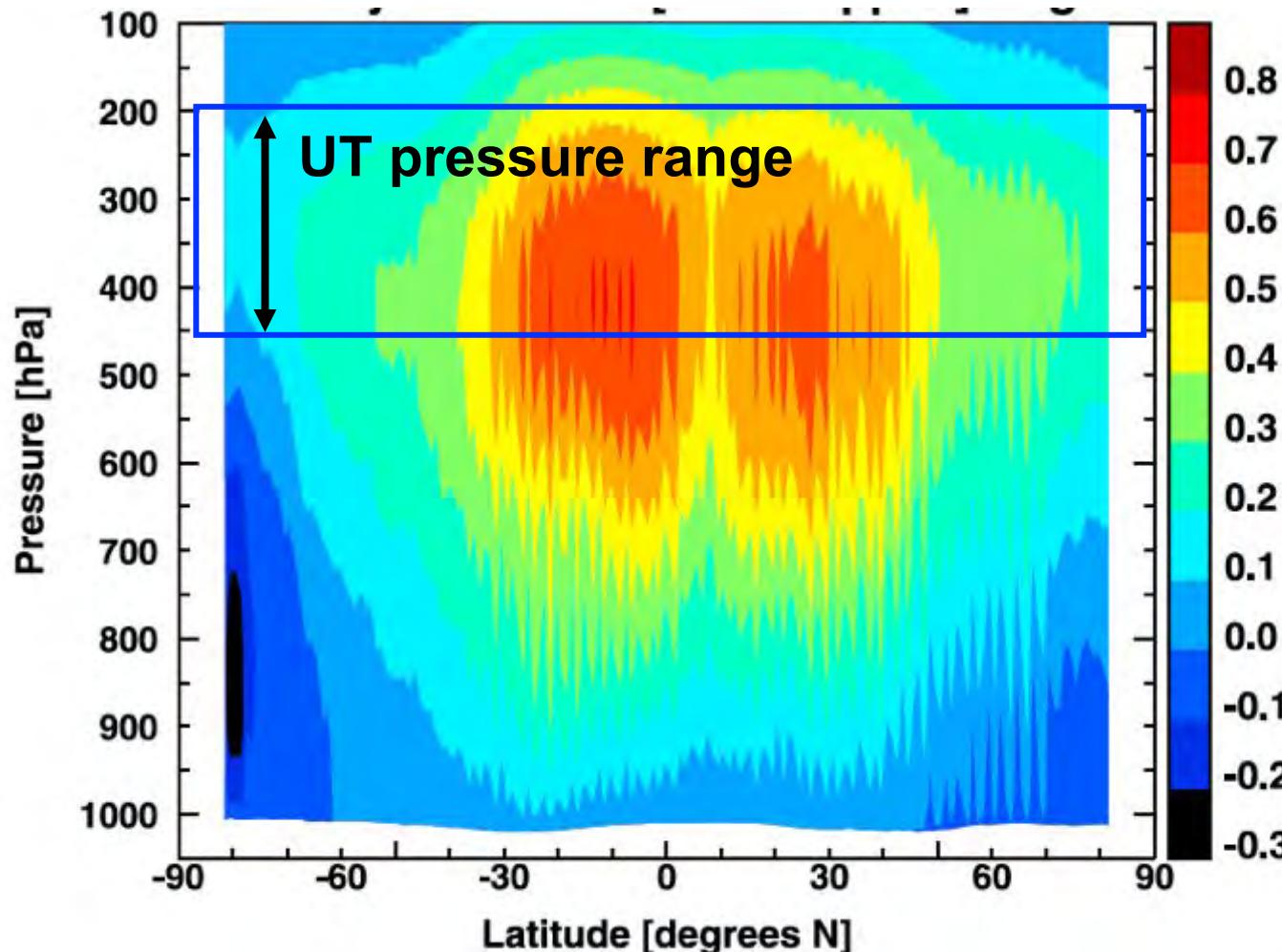
[adapted from Dahlmann et al., 2011]

Longer NO_x lifetime at higher altitude → greater OPE

Influence of upper tropospheric NO_x

The warming potential of ozone peaks the upper troposphere

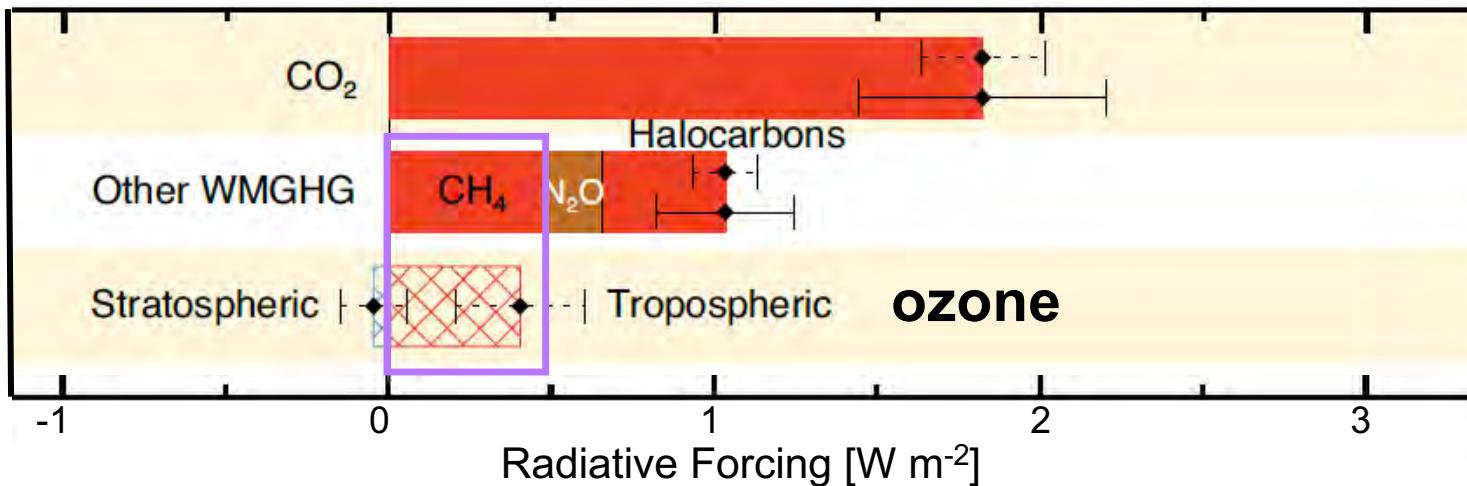
Sensitivity of outgoing longwave radiation to variations in ozone concentrations



Why uncertainties in the upper troposphere matter

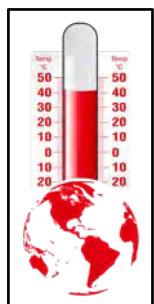
Warming due to tropospheric ozone is similar to that of methane (CH_4)

Tropospheric ozone and methane have near-equal climate impacts

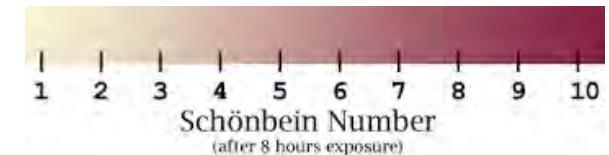
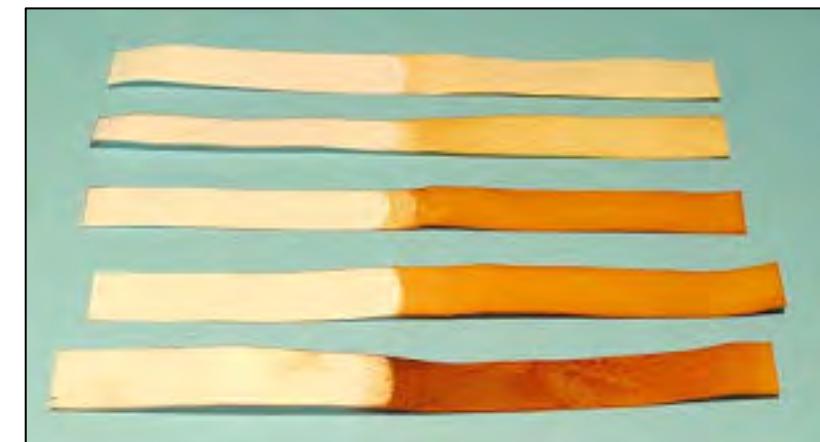


[IPCC AR5, 2013]

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



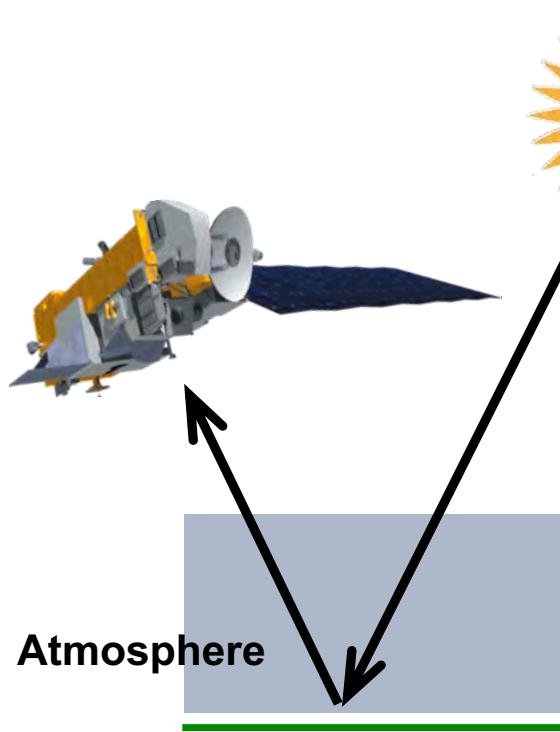
Errors in tropospheric ozone radiative forcing



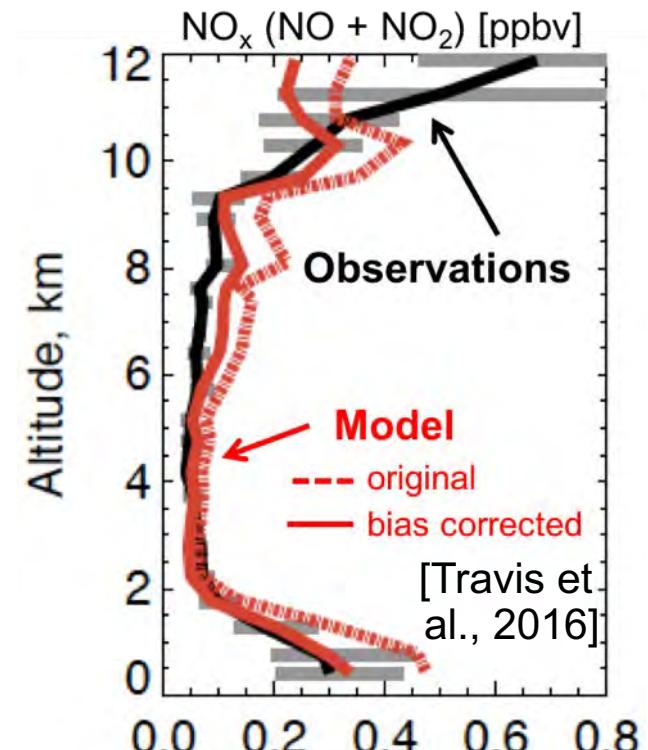
Indicator paper used to measure ozone in the mid to late 19th century

Why uncertainties in the upper troposphere matter

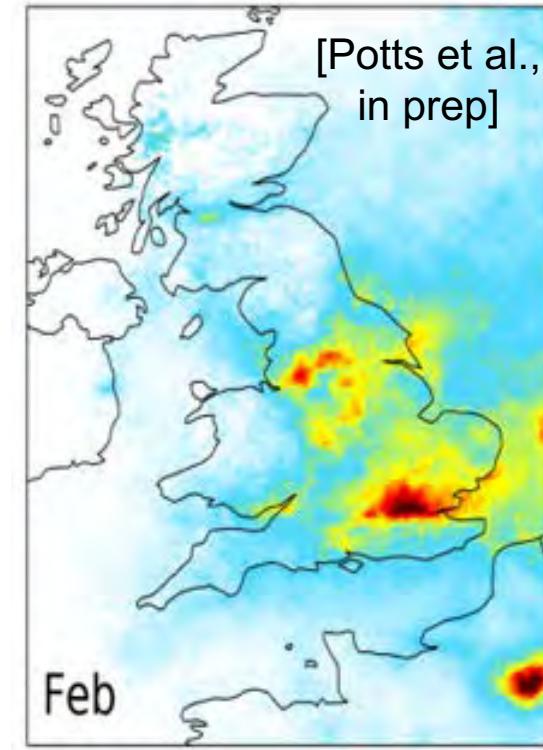
We rely on models to retrieve atmospheric composition from satellite instruments



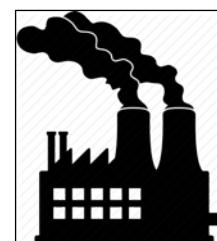
NO₂ along the viewing path



NO₂ vertical profile (model)



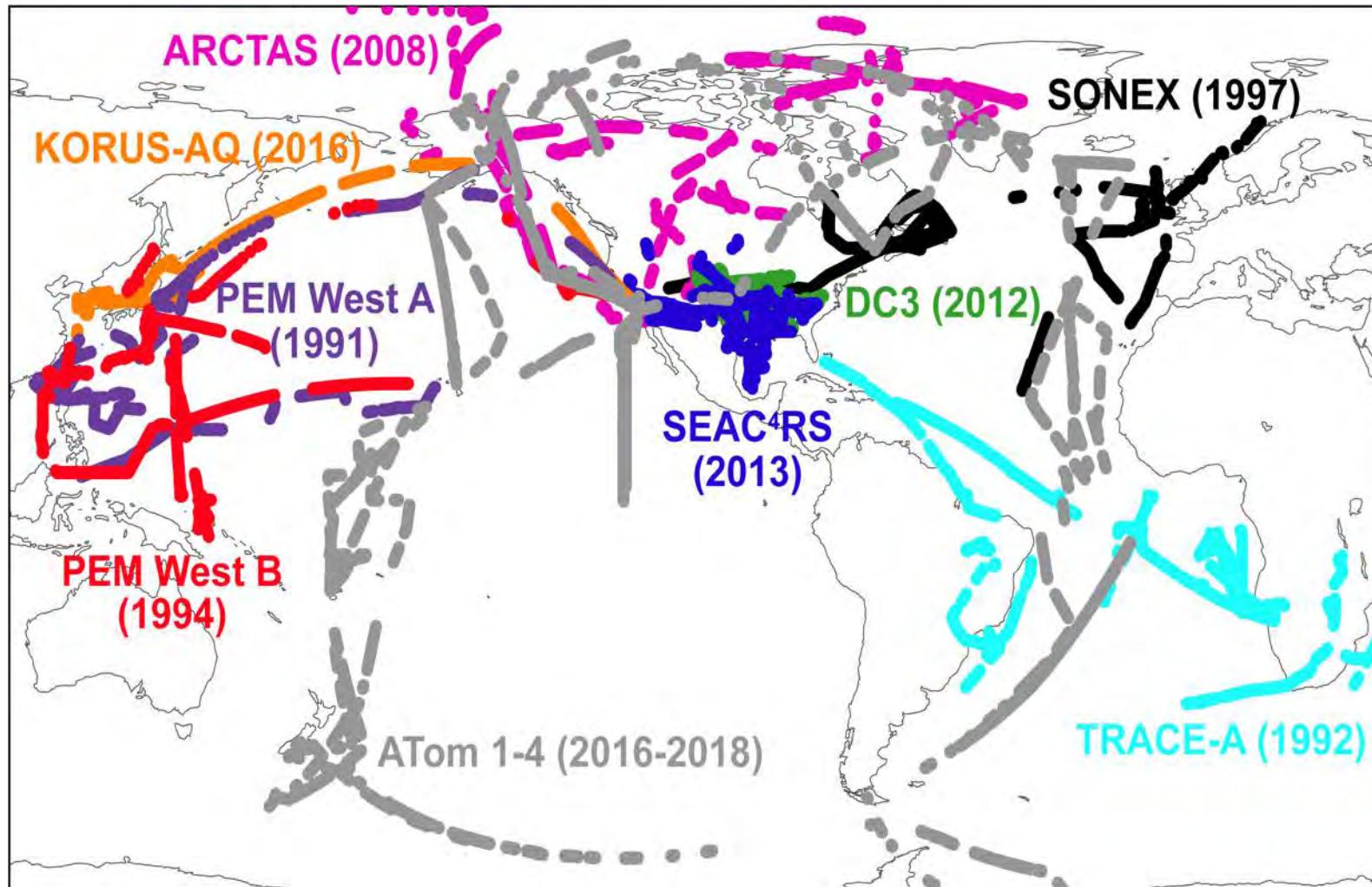
Vertical column densities



Uncertain global air quality
constraints from satellites

Sampling of the upper troposphere is limited

NASA DC8 research aircraft flight tracks in the upper troposphere



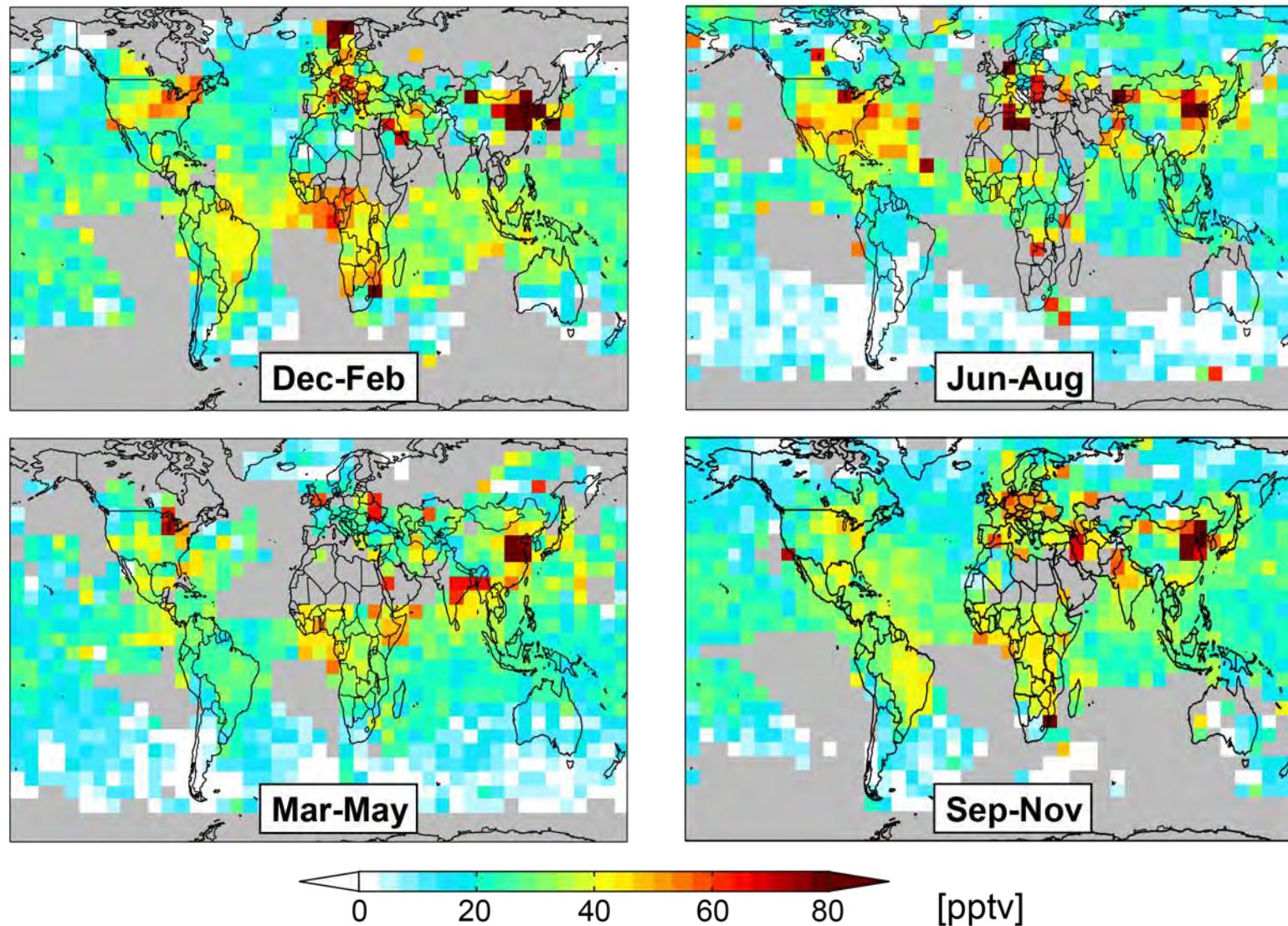
There are also measurements from commercial aircraft

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

Products of upper tropospheric NO₂ from satellite observations

Near global spatial coverage of seasonal mean UT NO₂ at $5^\circ \times 8^\circ$ (50 km \times 80 km)

NASA OMI upper troposphere NO₂ (2005-2007)



OMI:
Ozone Monitoring
Instrument

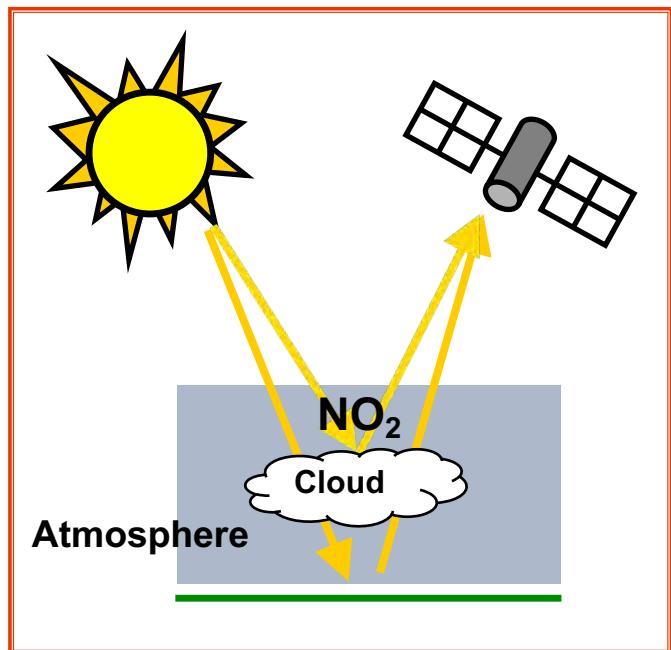
[Marais et al., 2018;
Choi et al., 2014]

Satellite products obtained using the cloud-slicing technique

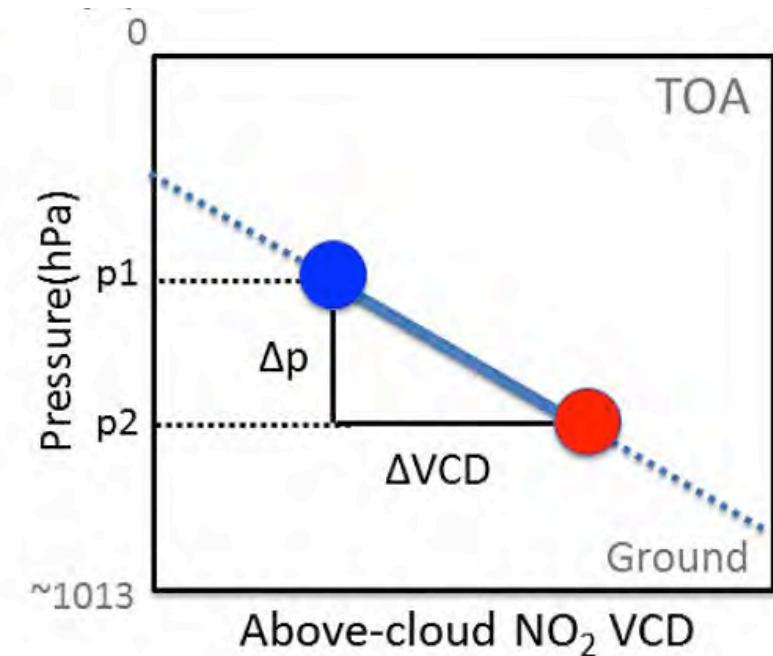
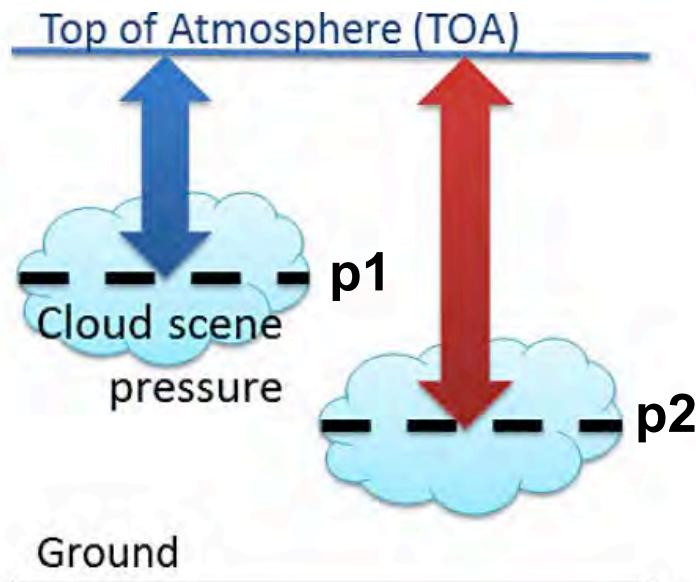
First applied by Ziemke et al. [2001] to TOMS ozone

Retrieve partial NO₂ columns over cloudy scenes at different heights

APPROACH



Use cloud height variability to derive partial columns



[adapted from Choi et al., 2014]

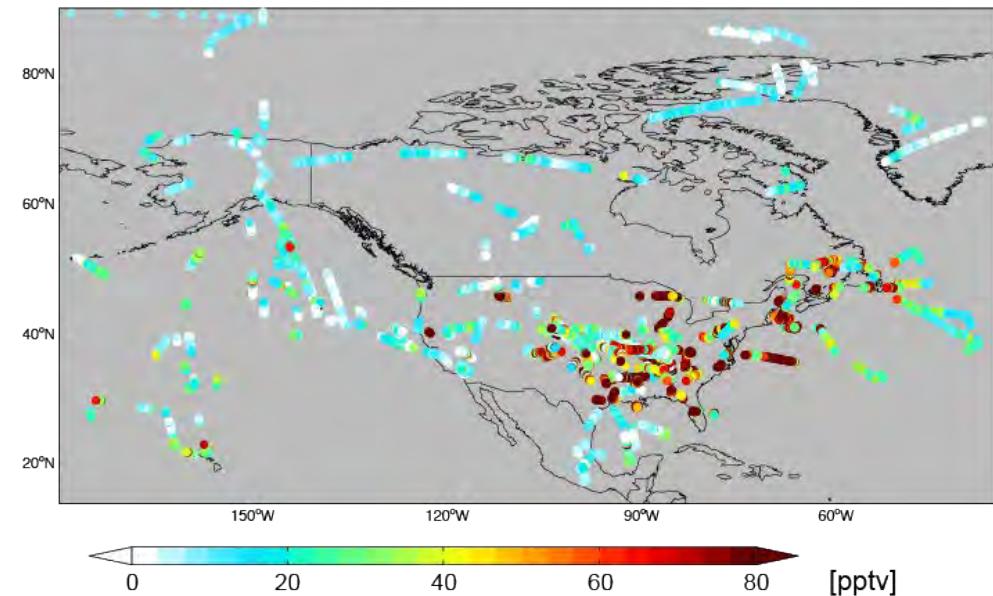
NO₂ volume mixing ratio (VMR) between clouds at p1 and p2

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

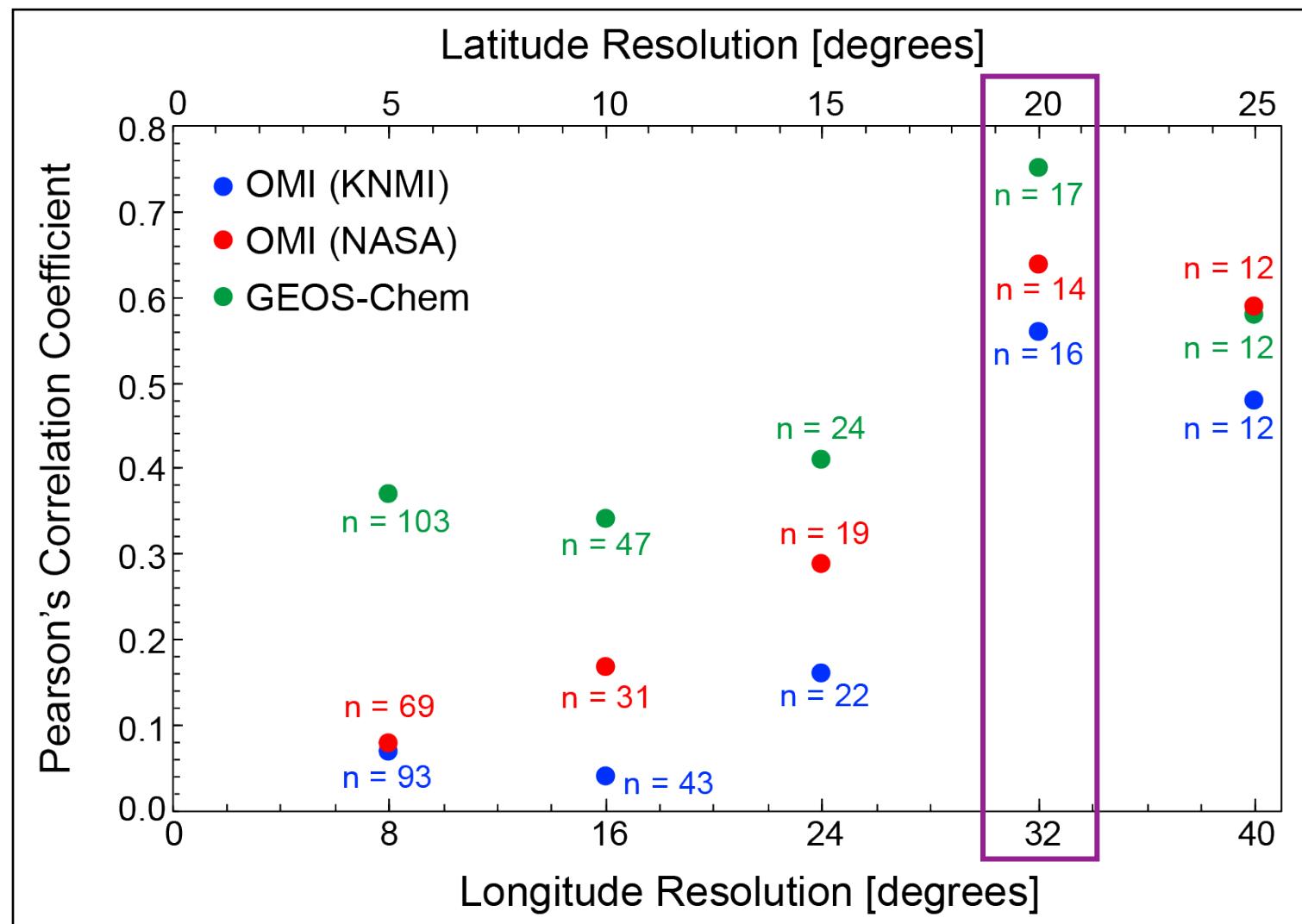
Consistency with aircraft observations at coarse scales

Agreement in spatial distribution between (reliable) aircraft and OMI UT NO₂ observations, but at coarse scales (seasonal, 20° latitude (200 km) × 32° longitude (320 km))

NASA DC8 NO₂ in Spring-Summer

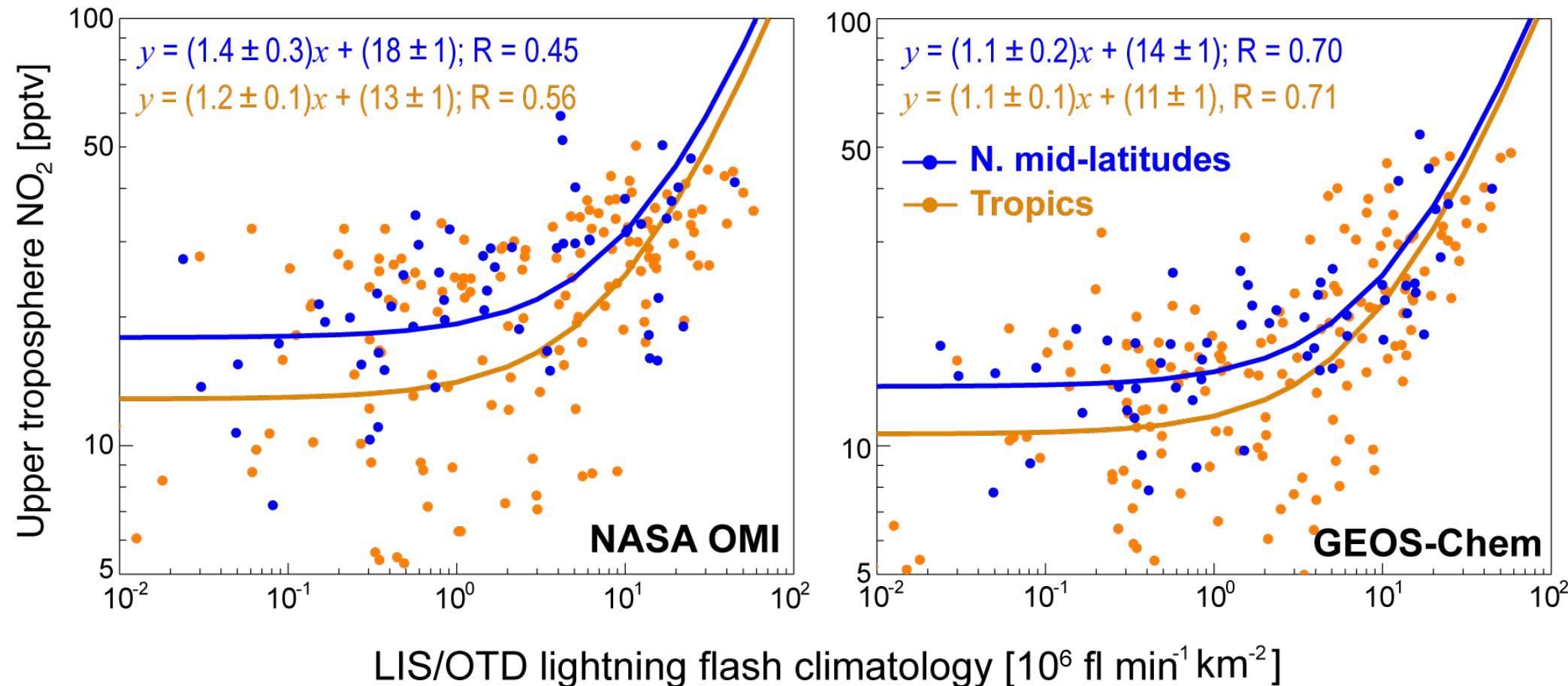


[Marais et al., 2018]



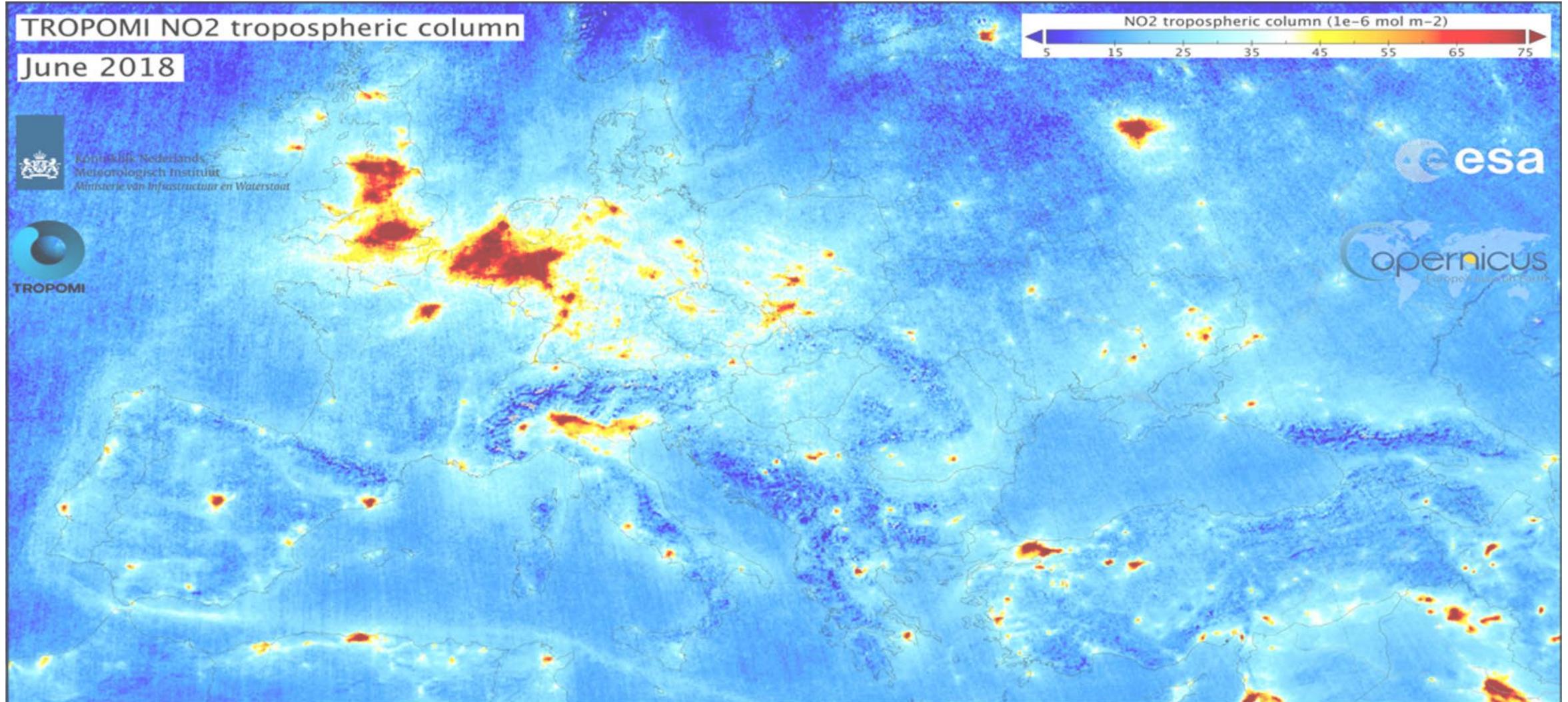
Provide improved constraints on lightning NO_x emissions

Log-log relationship between UT NO₂ 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?

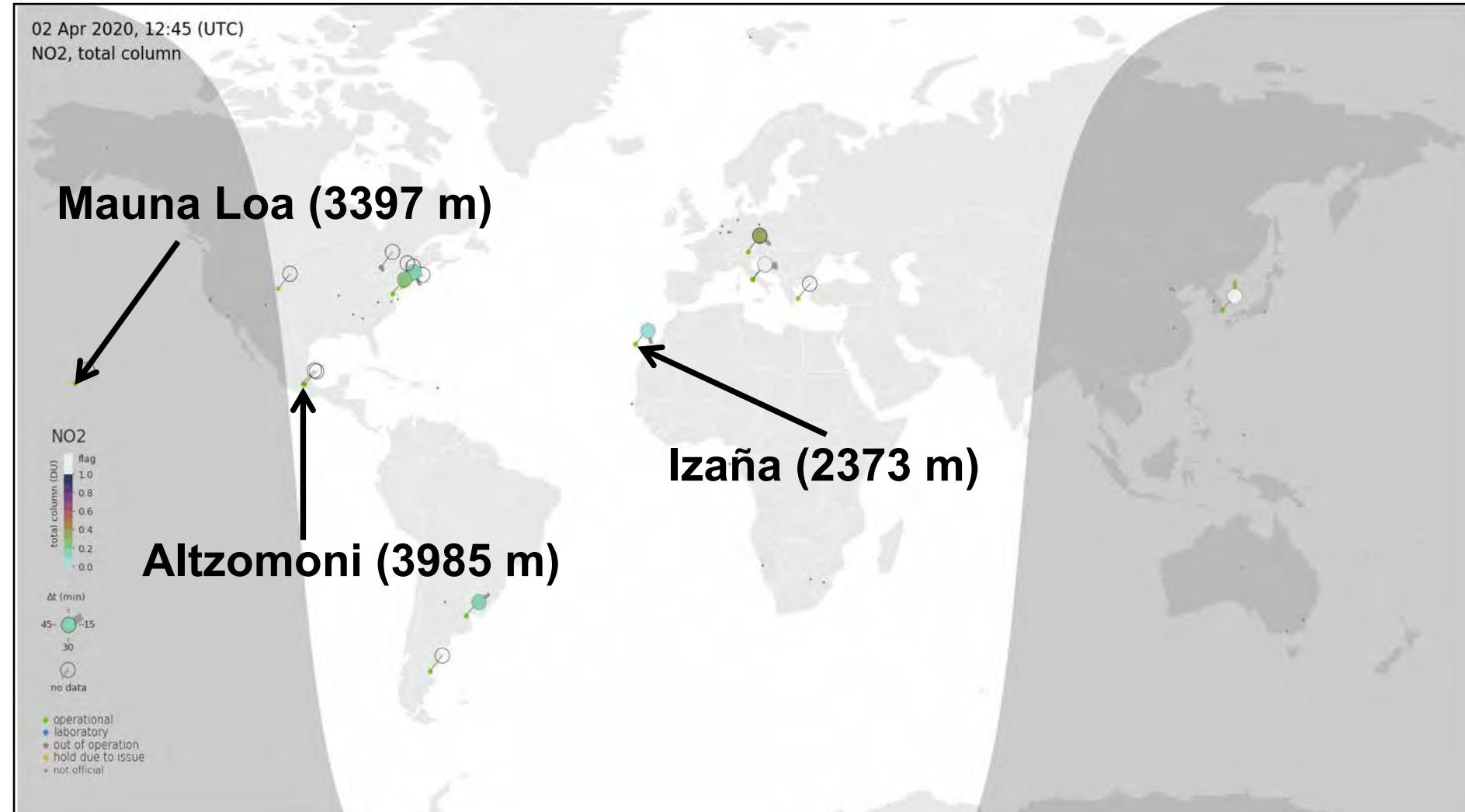
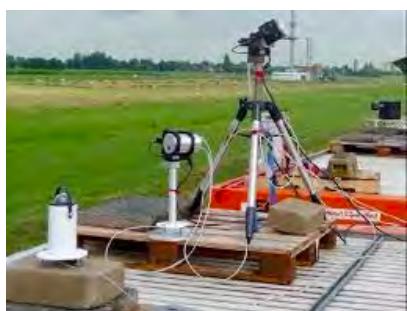


[Source: <http://www.tropomi.eu/data-products/nitrogen-dioxide>]

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

Evaluate TROPOMI with ground-based measurements

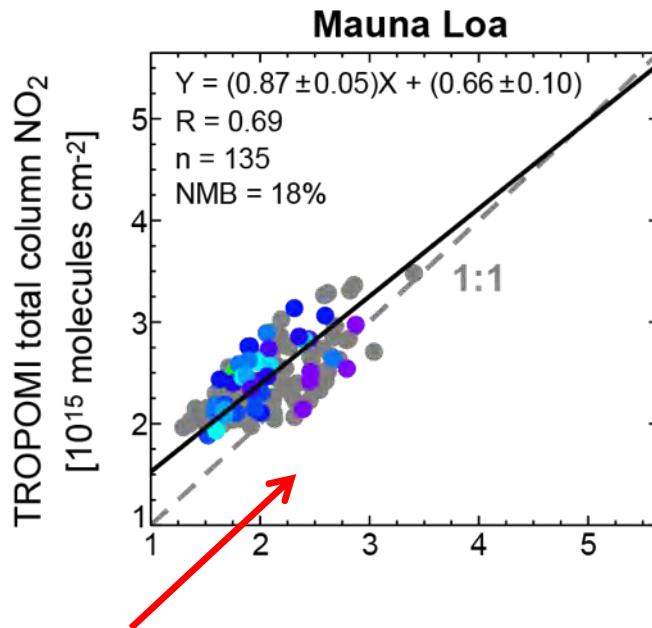
Global Pandora network, indicating locations of high-altitude sites (large relative contribution from the UT) used to evaluate TROPOMI



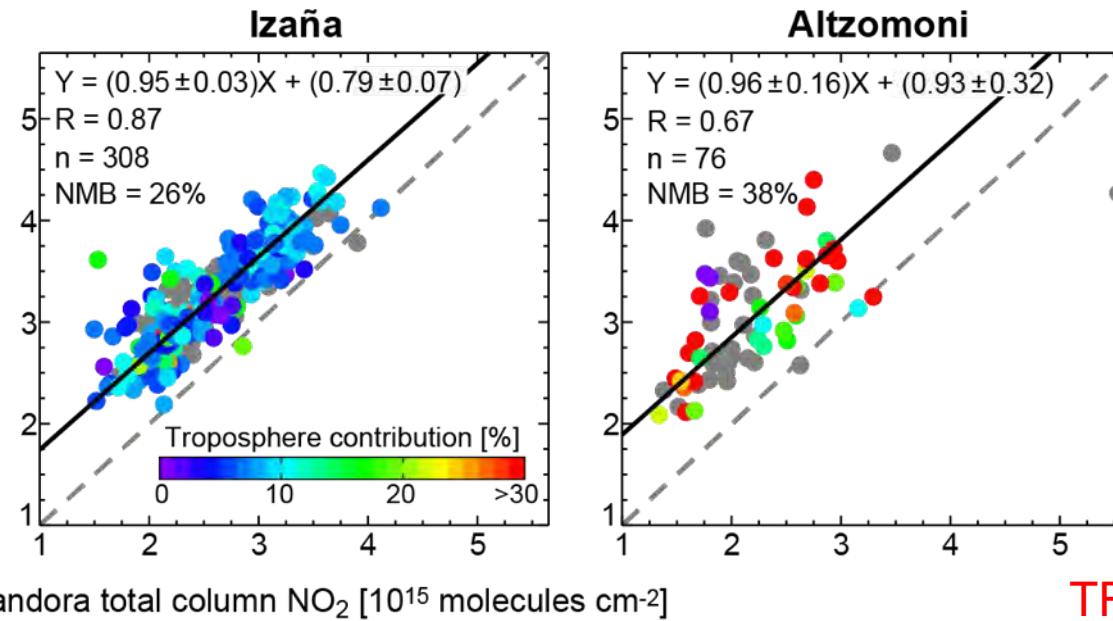
[Source: <https://www.pandoria-global-network.org/>]

Evaluate TROPOMI with ground-based measurements

Stratosphere:



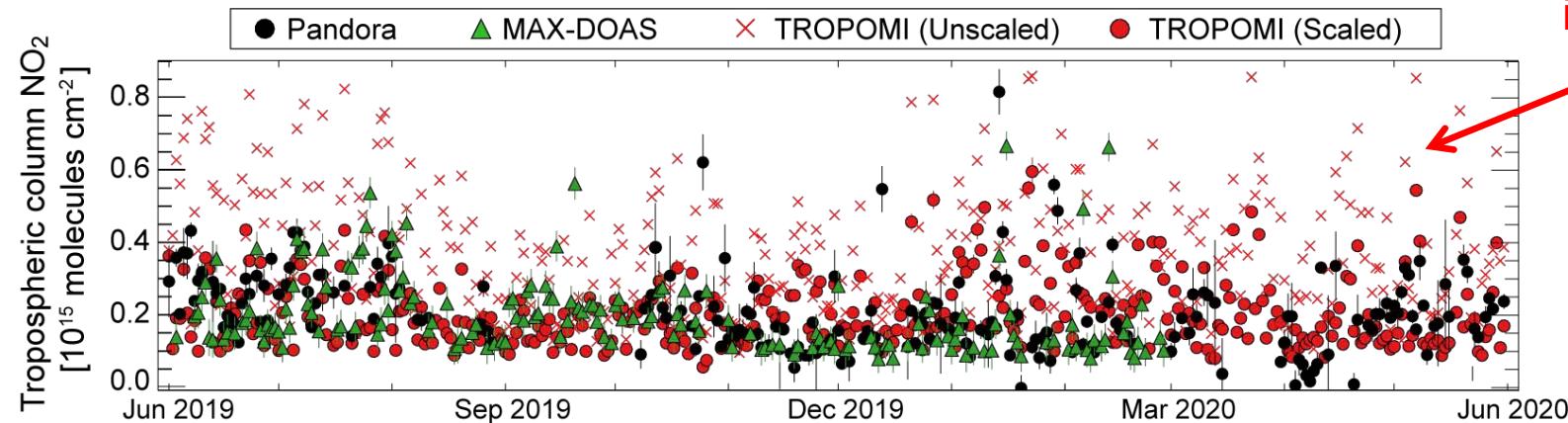
Stratosphere + free troposphere:



TROPOMI
stratospheric column
variance is
underestimated

TROPOMI free
tropospheric column
is overestimated

Free troposphere:



TROPOMI cloud-sliced upper tropospheric NO₂

Data are at $1^\circ \times 1^\circ$

Range: 30-80 pptv

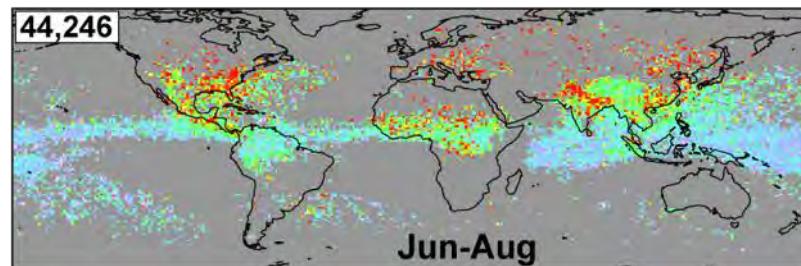
Background: ~30 pptv

Cloud products give similar UT NO₂ in the tropics

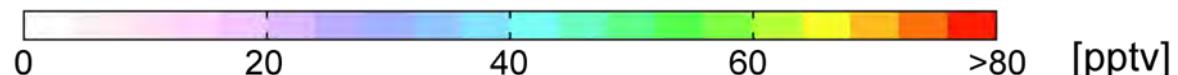
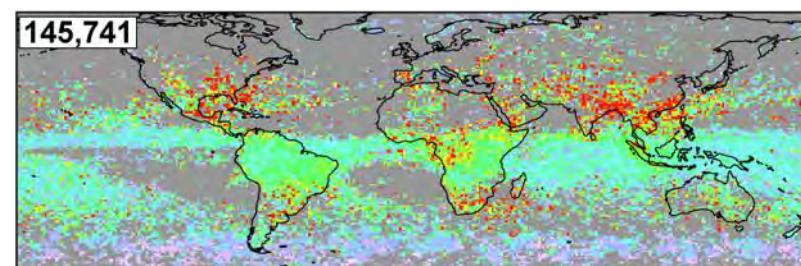
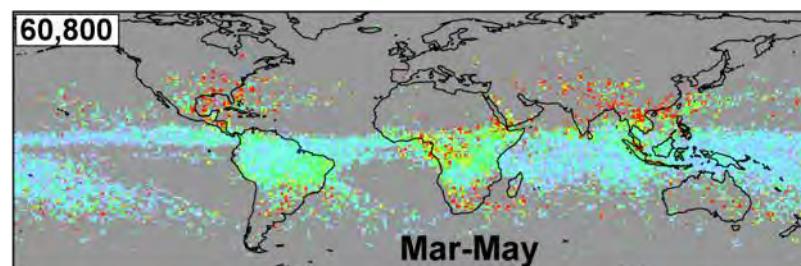
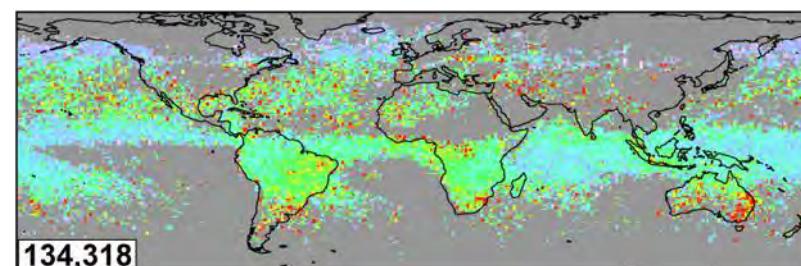
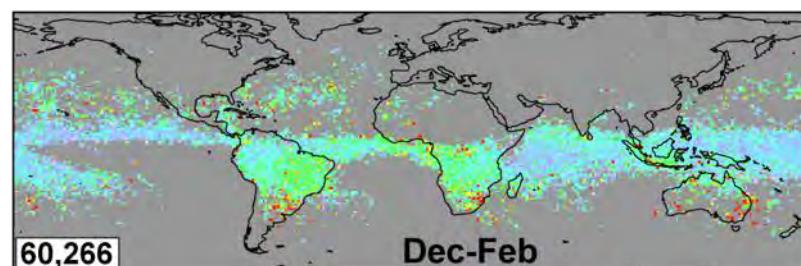
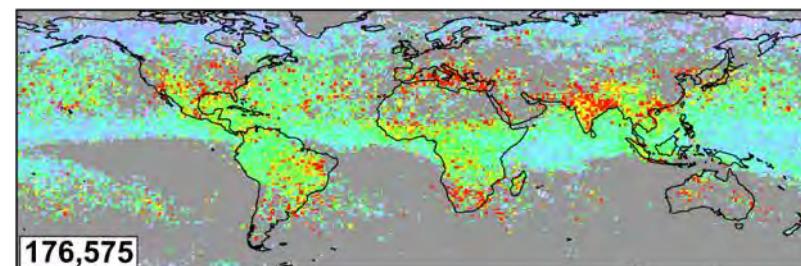
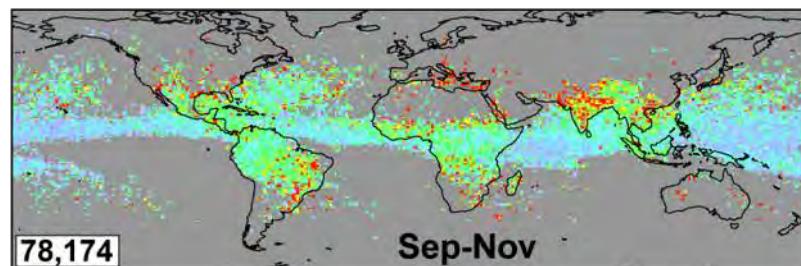
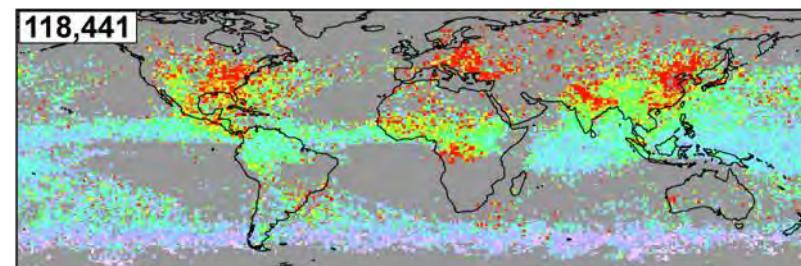
Cloud product two gives greater global coverage

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

Cloud Product One

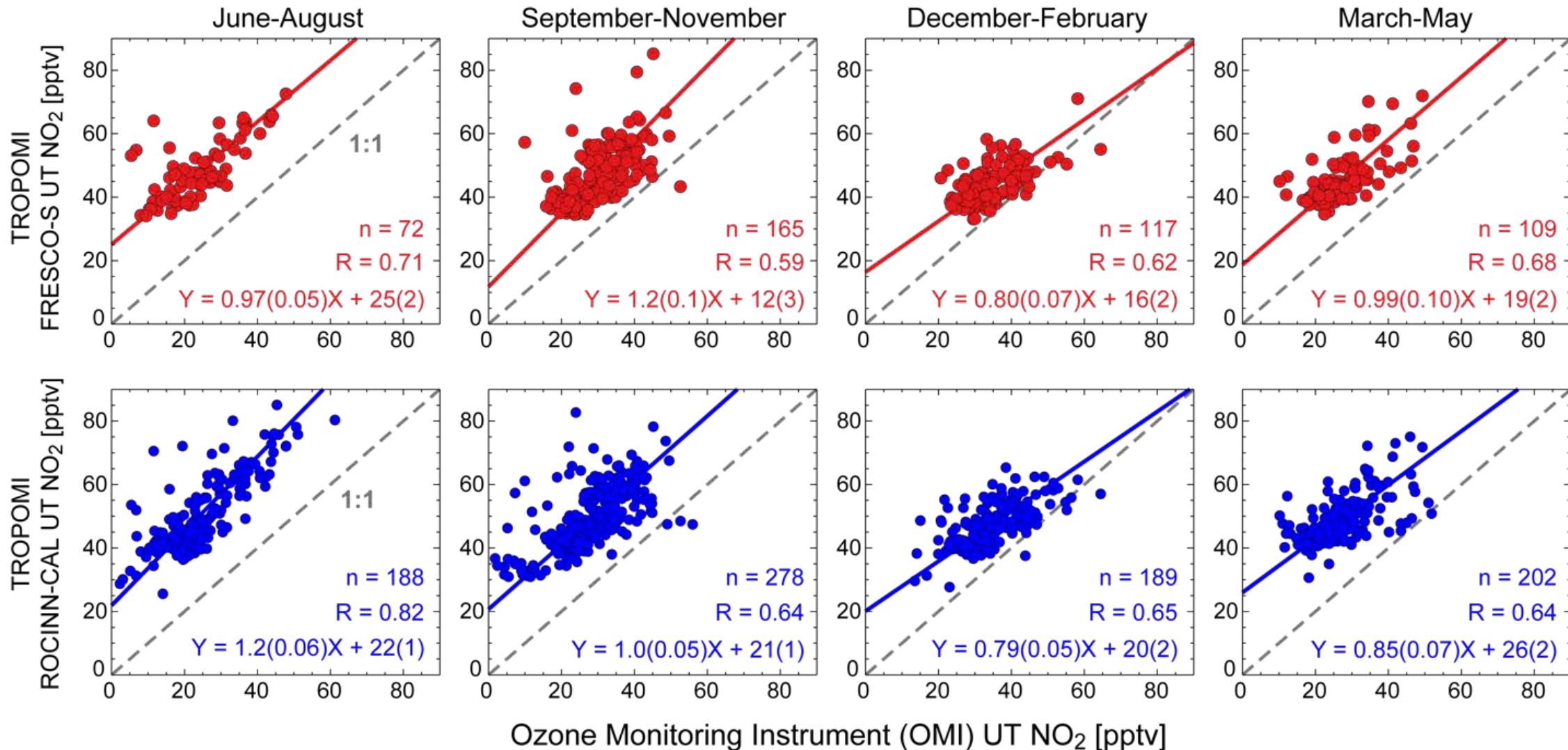


Cloud Product Two



Comparison to the NASA OMI product

TROPOMI UT NO₂ obtained at 1° × 1° and gridded to the NASA product resolution (8° × 5°)



Spatial consistency. TROPOMI is 12-26 pptv higher than OMI (clouds and vertical NO₂ profile)

Back to Earth...

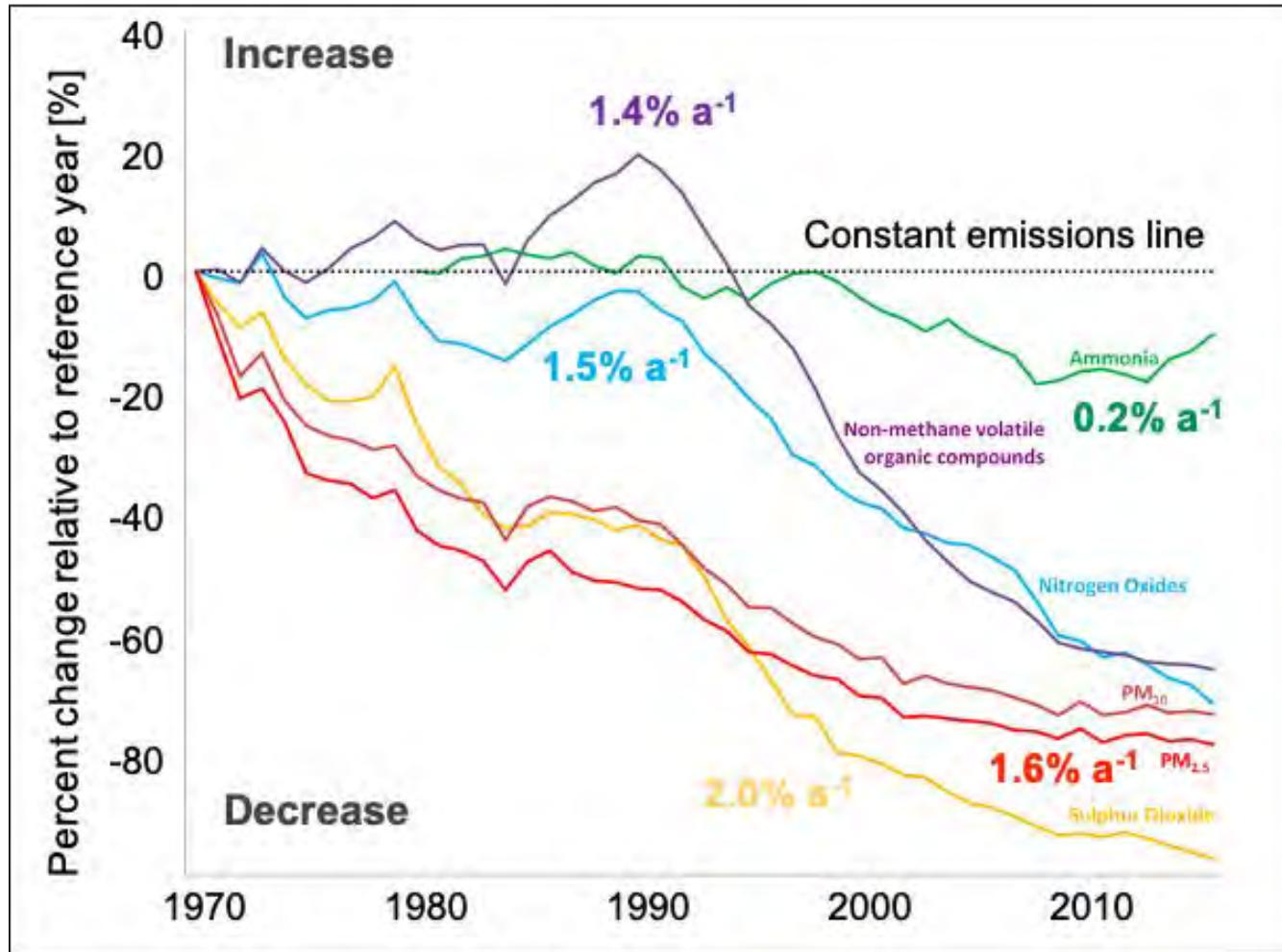
Addressing uncertainties in UK ammonia emissions



Contributors: A. Pandey, M. Van Damme, L. Clarisse, P. F. Coheur, I. Tsagatakis, M. Shephard, K. Cady-Perreira, M. Sitwell, S. Reis, U. Harding, E. Nemitz, M. Veino, G. Hayman, T. Misselbrook, L. Zhu

Ammonia emissions in the UK: the bottom-up perspective

Temporal (Time) Variability in Emissions

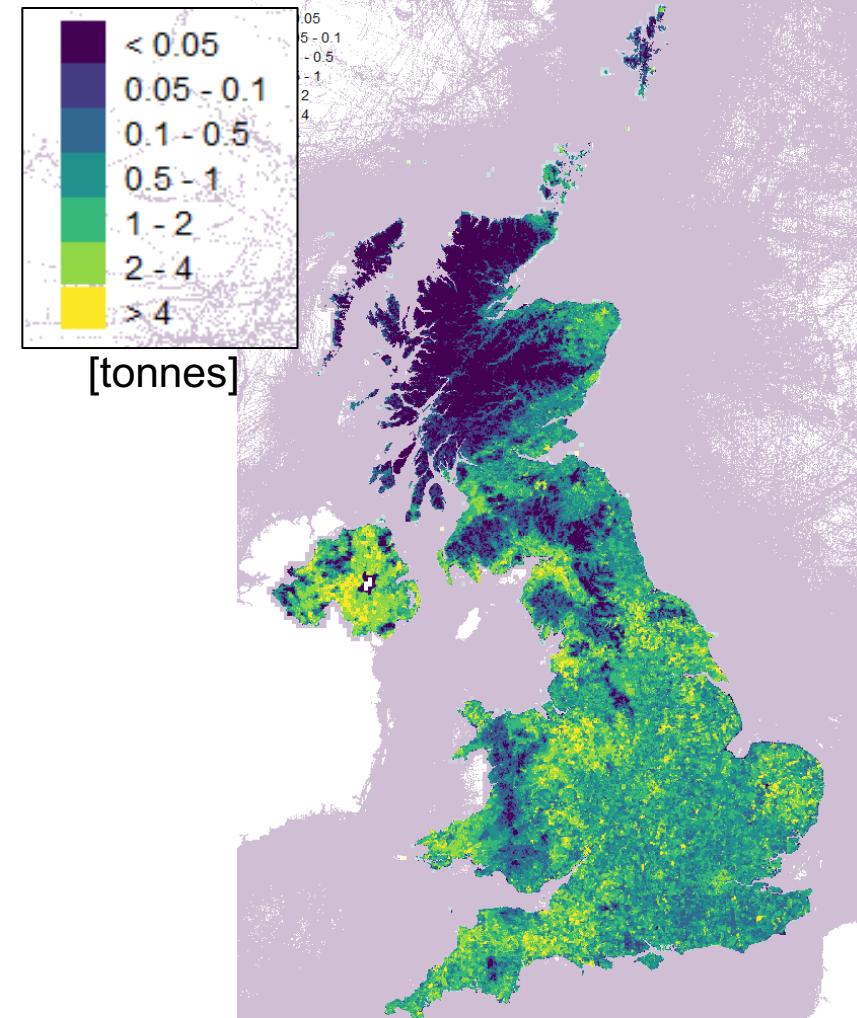


[Adapted from Defra, 2018]

Successful decline in all emissions, except ammonia (NH₃)

Spatial Variability in Emissions

NH₃ emissions for 2018 at 1 km

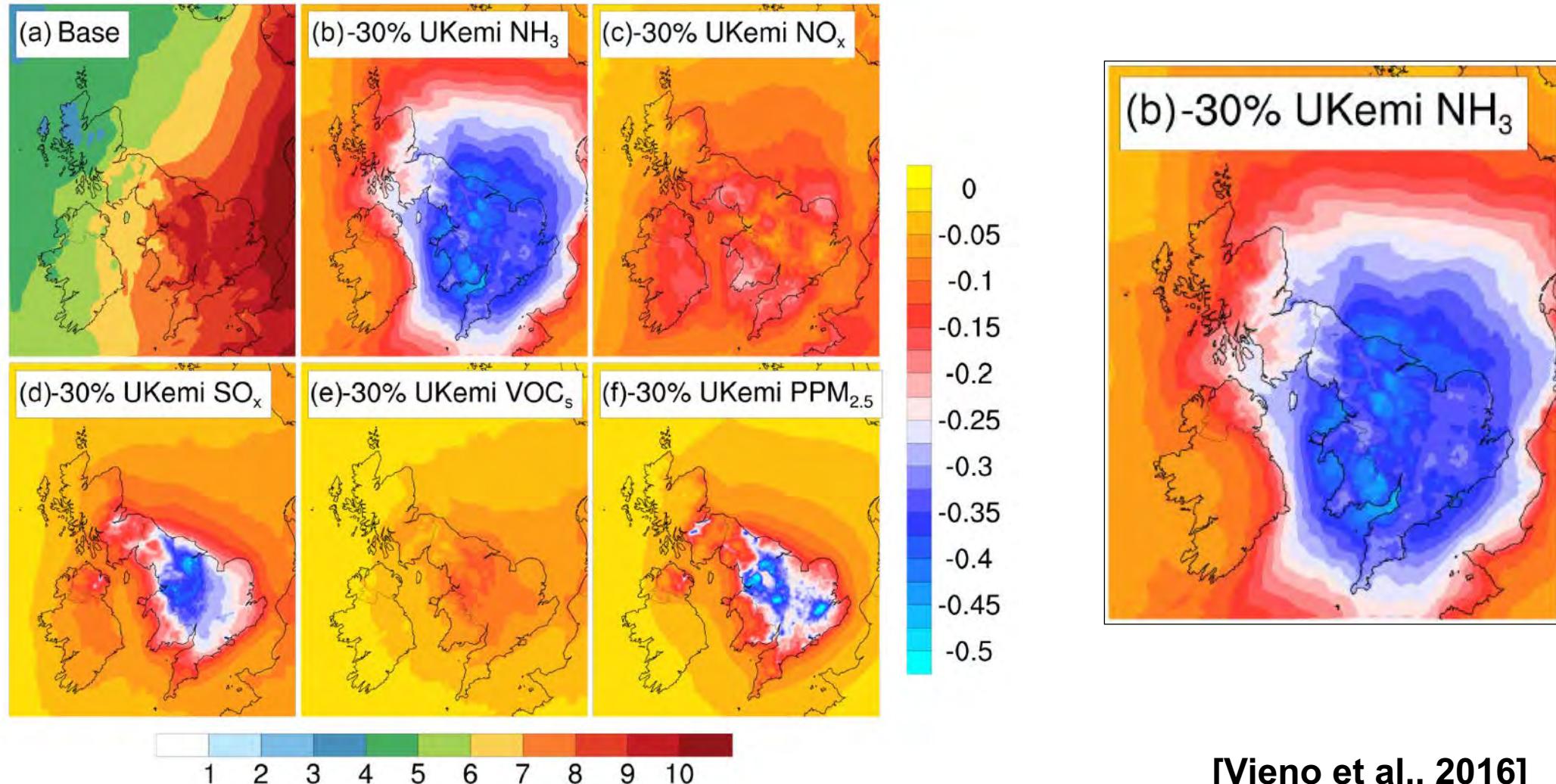


All maps © Crown copyright. All rights reserved Defra, Licence number 100022861 (2020) and BEIS, Licence number 100037028 (2020) LPS © Crown copyright and database right 2020 Licence INSP594

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

Ammonia impact on air pollutants hazardous to health

Effect of emission controls 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

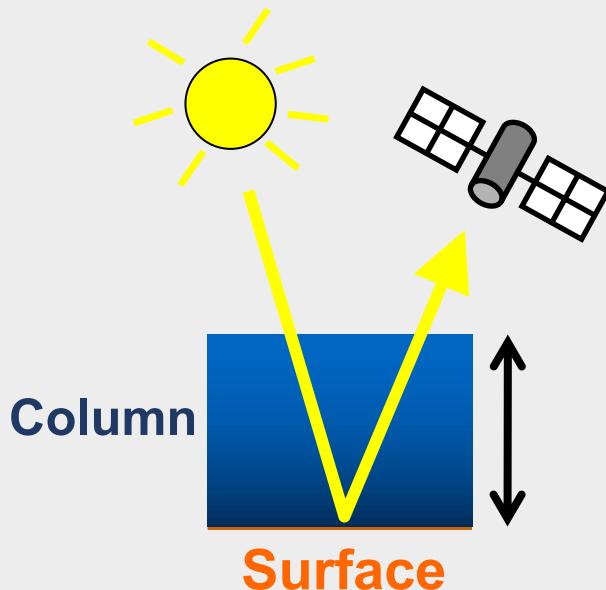


Conversion Factor

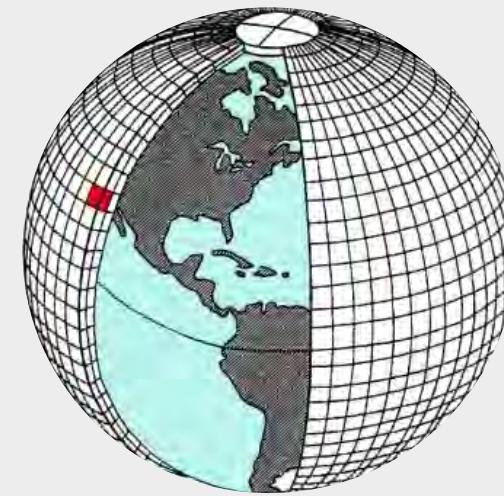


EMISSIONS

Satellite column densities



Model Concentration-to-Emission Ratio



Satellite-derived Surface Emissions

Emission



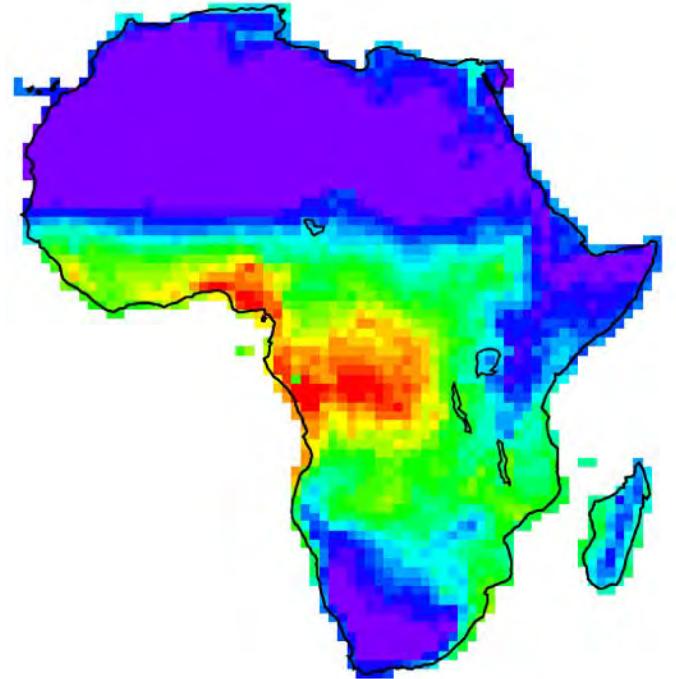
Widely used to estimate emissions and surface concentrations

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

Concentrations → emissions: formaldehyde → isoprene, $\text{NO}_2 \rightarrow \text{NO}_x$

Column → surface: formaldehyde → formaldehyde, $\text{NO}_2 \rightarrow \text{NO}_2$, AOD → $\text{PM}_{2.5}$

Satellite formaldehyde

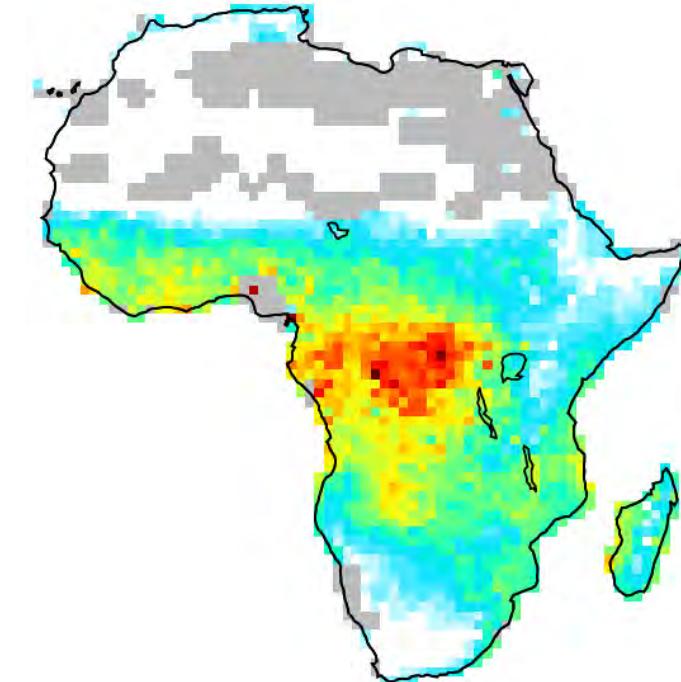


0 2 4 6 8
[10^{15} molecules cm^{-2}]



Model effective
yields

Isoprene emissions

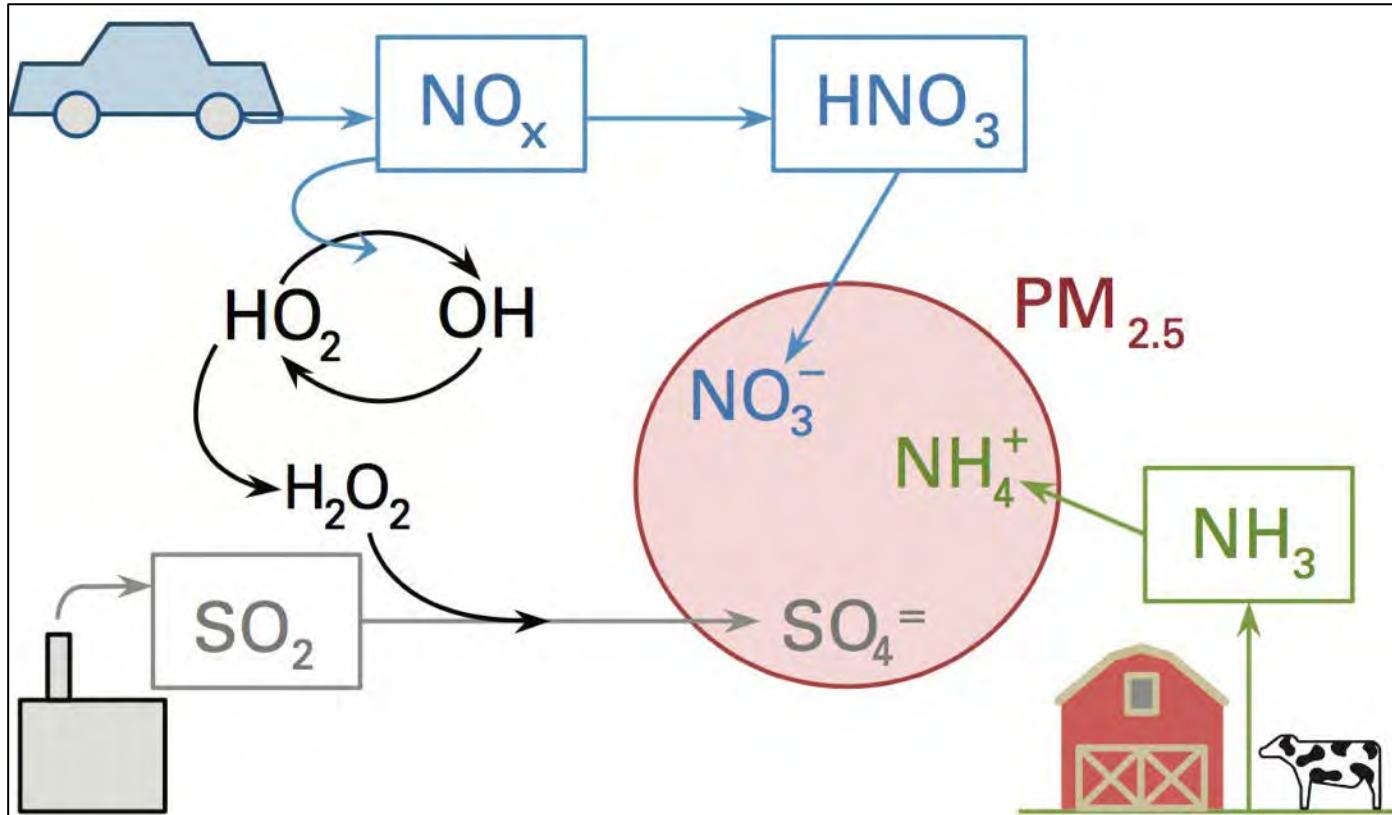


0 1 2 3 4 5 6
[10^{12} atoms C $\text{cm}^{-2} \text{s}^{-1}$]

[Marais et al., ACP, 2012]

Ammonia abundance depends on numerous factors

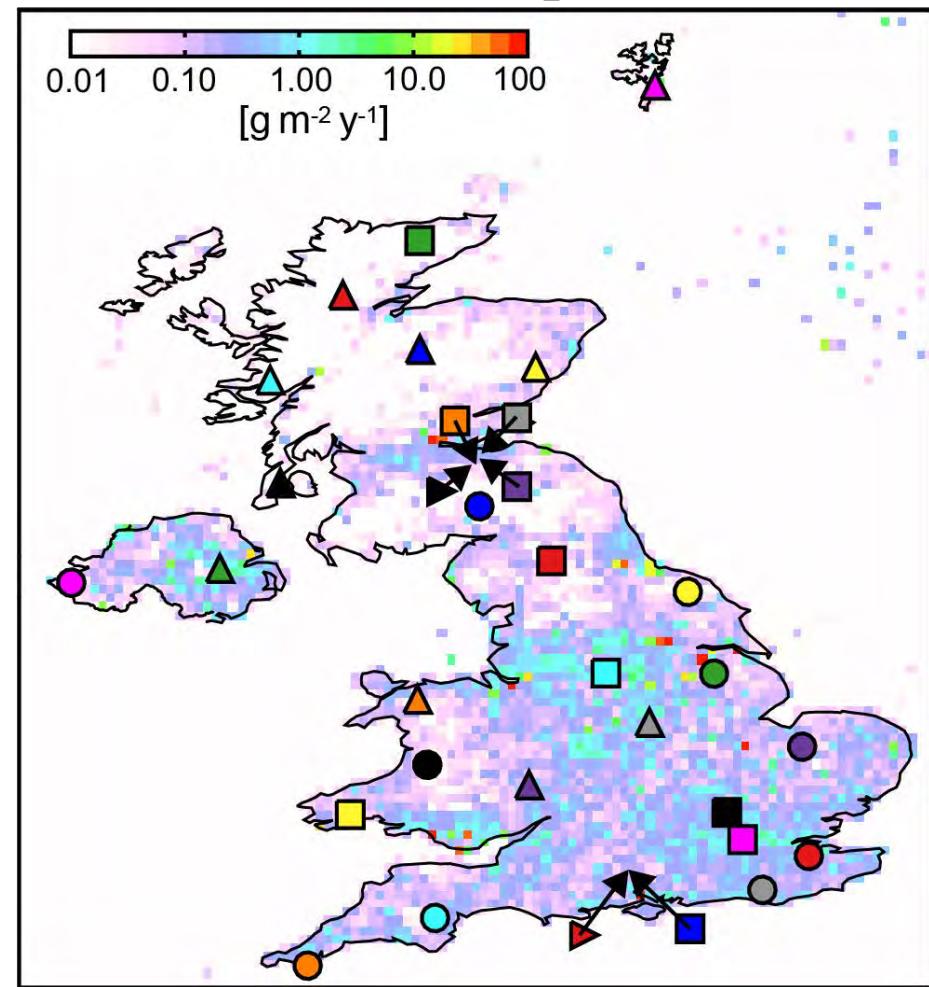
Ammonia partitions to aerosols to form PM_{2.5}



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

Partitioning of ammonia (NH₃) to pre-existing aerosols depends on abundance of NO_x and SO₂

NAEI Annual SO₂ Emissions



Symbols: SO₂ concentration monitors

Surface SO₂ concentrations calculated with GEOS-Chem



3D Atmospheric Chemistry Transport Model

Emissions
(natural/human)

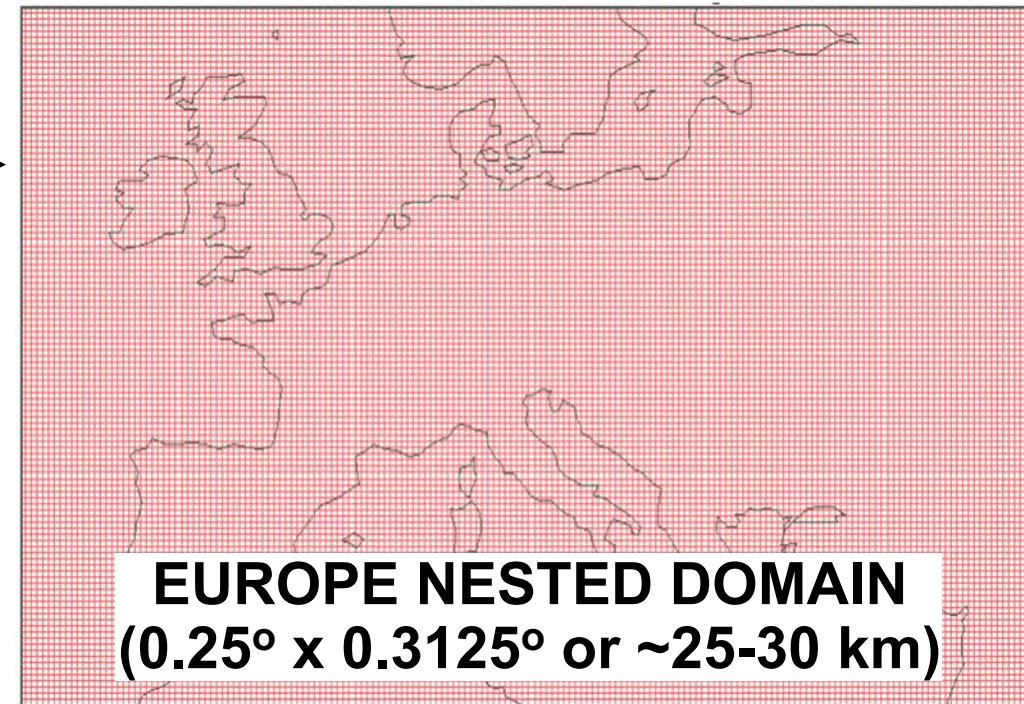


UK NAEI emissions
(with temporal information)

Offline assimilated
meteorology

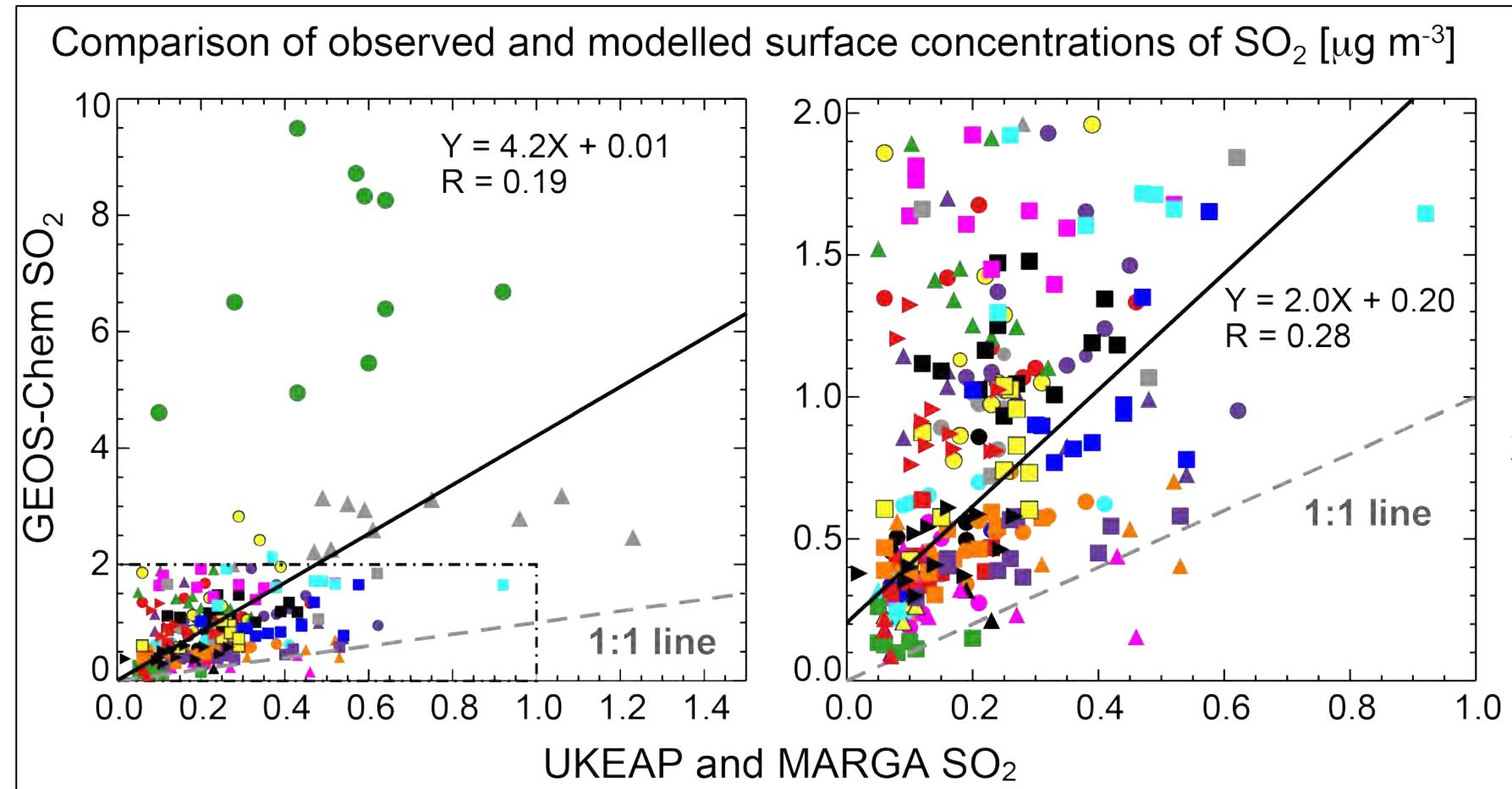
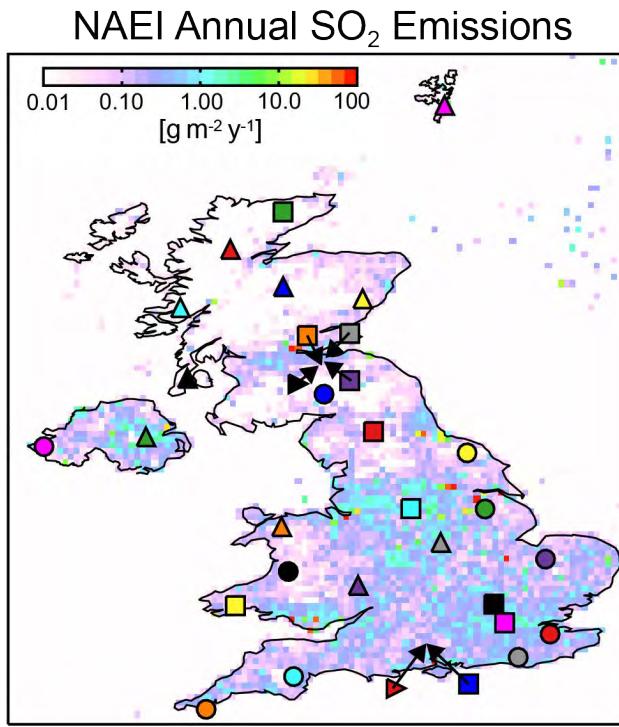


NASA GEOS-FP for 2016



Gas phase and heterogeneous chemistry
Transport
Dry/wet deposition

GEOS-Chem-NAEI (model) versus observed SO₂



UKEAP: offline denuder measurements (0.05 μg m⁻³ detection limit)

MARGA: semi-continuous denuder measurements (0.04 μg m⁻³ detection limit)

Comparison supports large overestimate in NAEI SO₂ emissions (in particular point sources)

Decrease annual SO₂ emissions 161 Gg to 87 Gg

Infrared Atmospheric Sounding Interferometer (IASI) Instrument

Overpass:

9:30 local solar time

Spatial resolution:

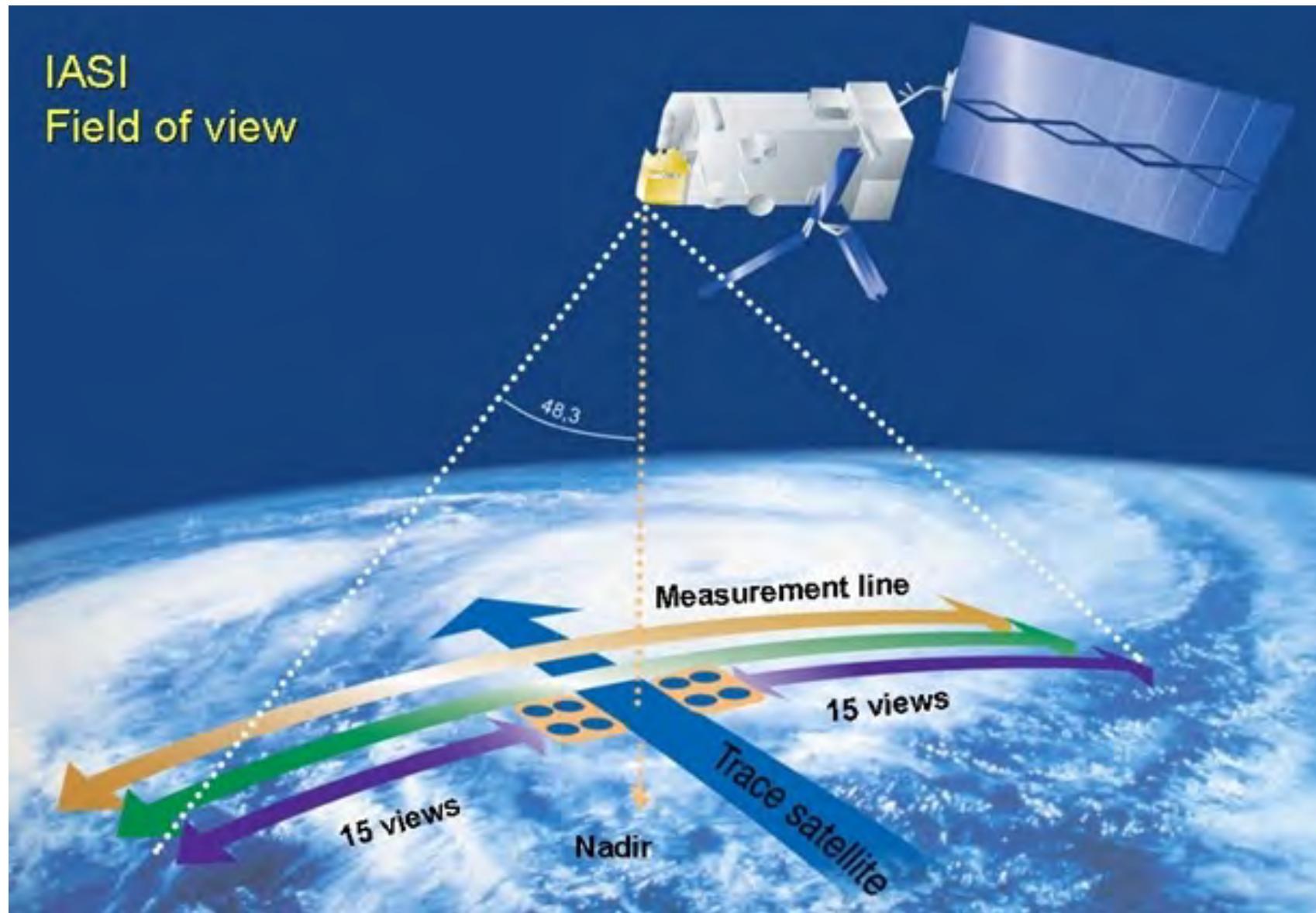
12 km to 39 km

Swath width:

2200 km

Launch date:

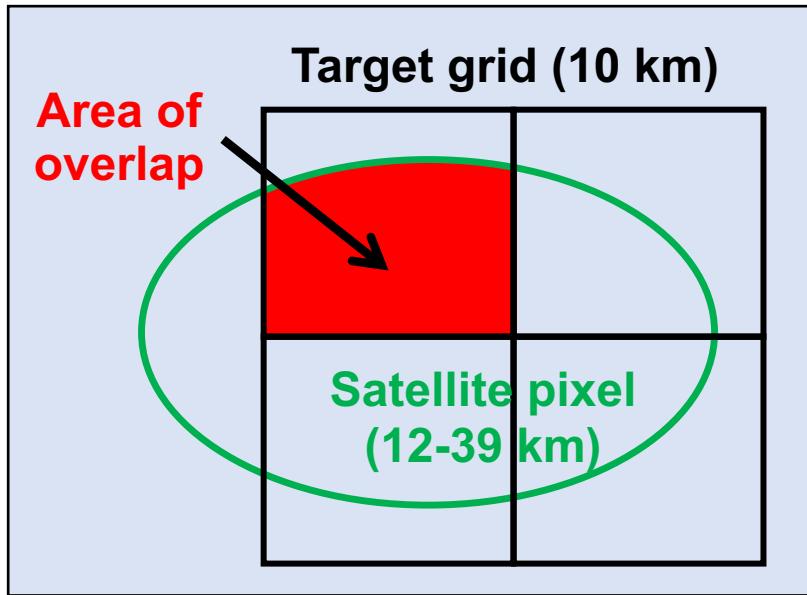
October 2006



Ammonia emissions in the UK: the top-down perspective

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

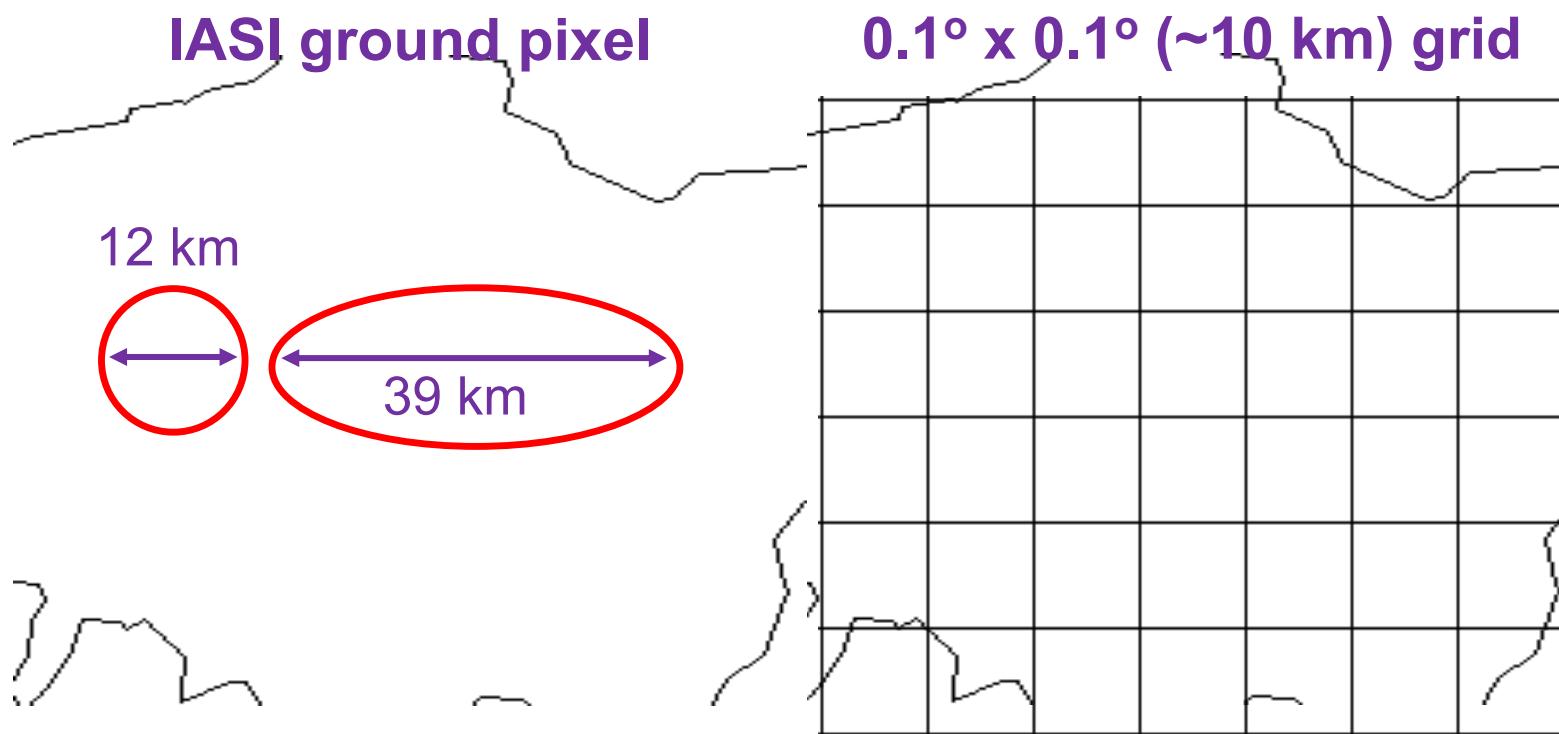
Oversampling Technique



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

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

Oversampling technique over London

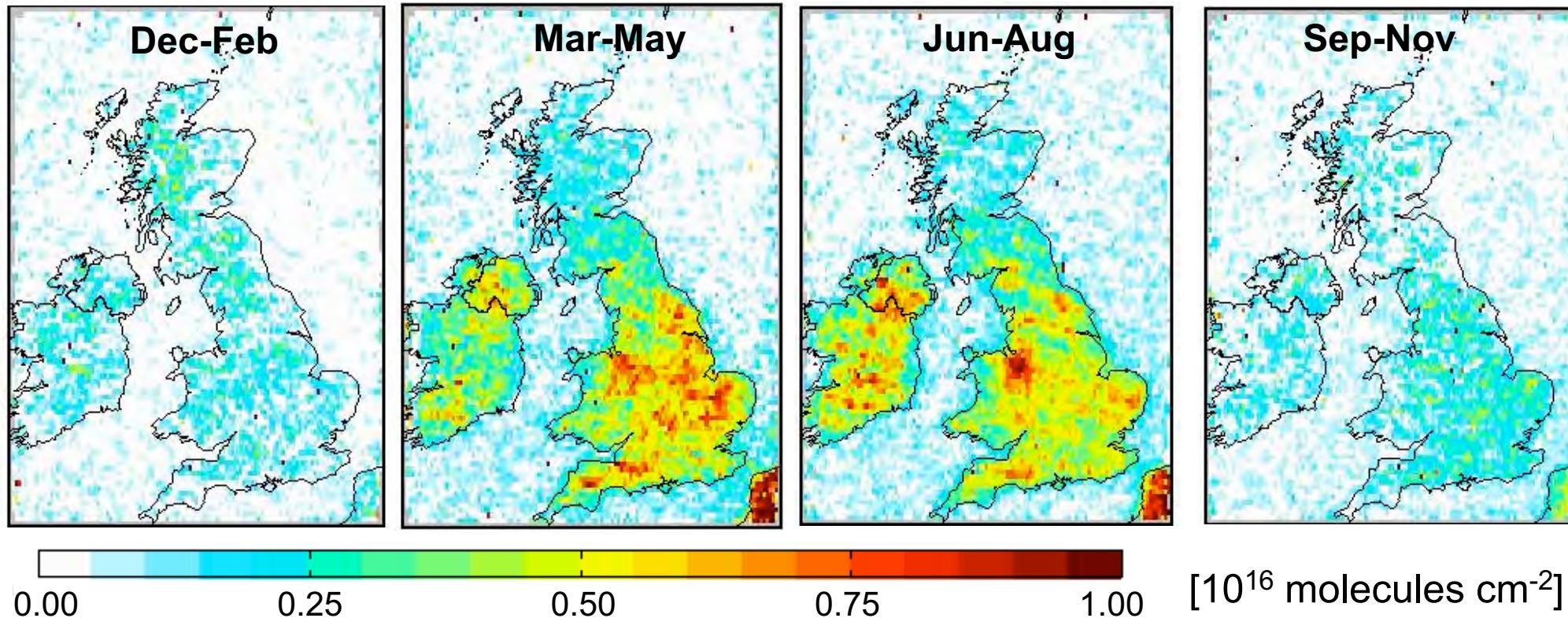


Lose time (temporal) resolution; gain spatial resolution

Ammonia emissions in the UK: the top-down perspective

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

Annual multiyear (2008-2018) mean IASI NH₃ on a 0.1° x 0.1° (~10 km) grid



Units are number of ammonia molecules in a column of air from the surface to the satellite

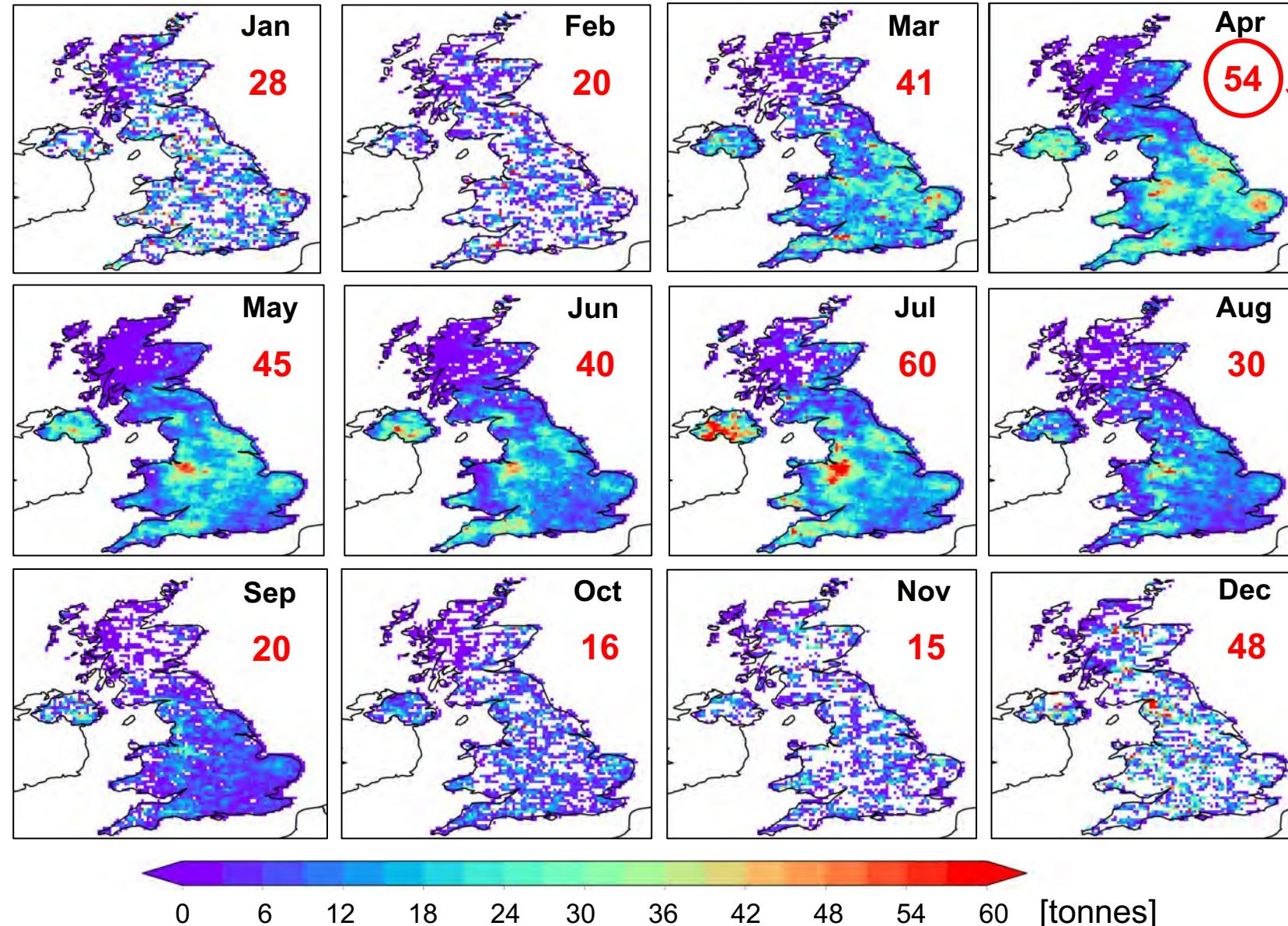
IASI data providers: M. Van Damme, L. Clarisse,
P.-F. Coheur, ULB, Belgium

UK IASI-derived ammonia emissions

Convert IASI NH₃ column concentrations to surface emissions of NH₃

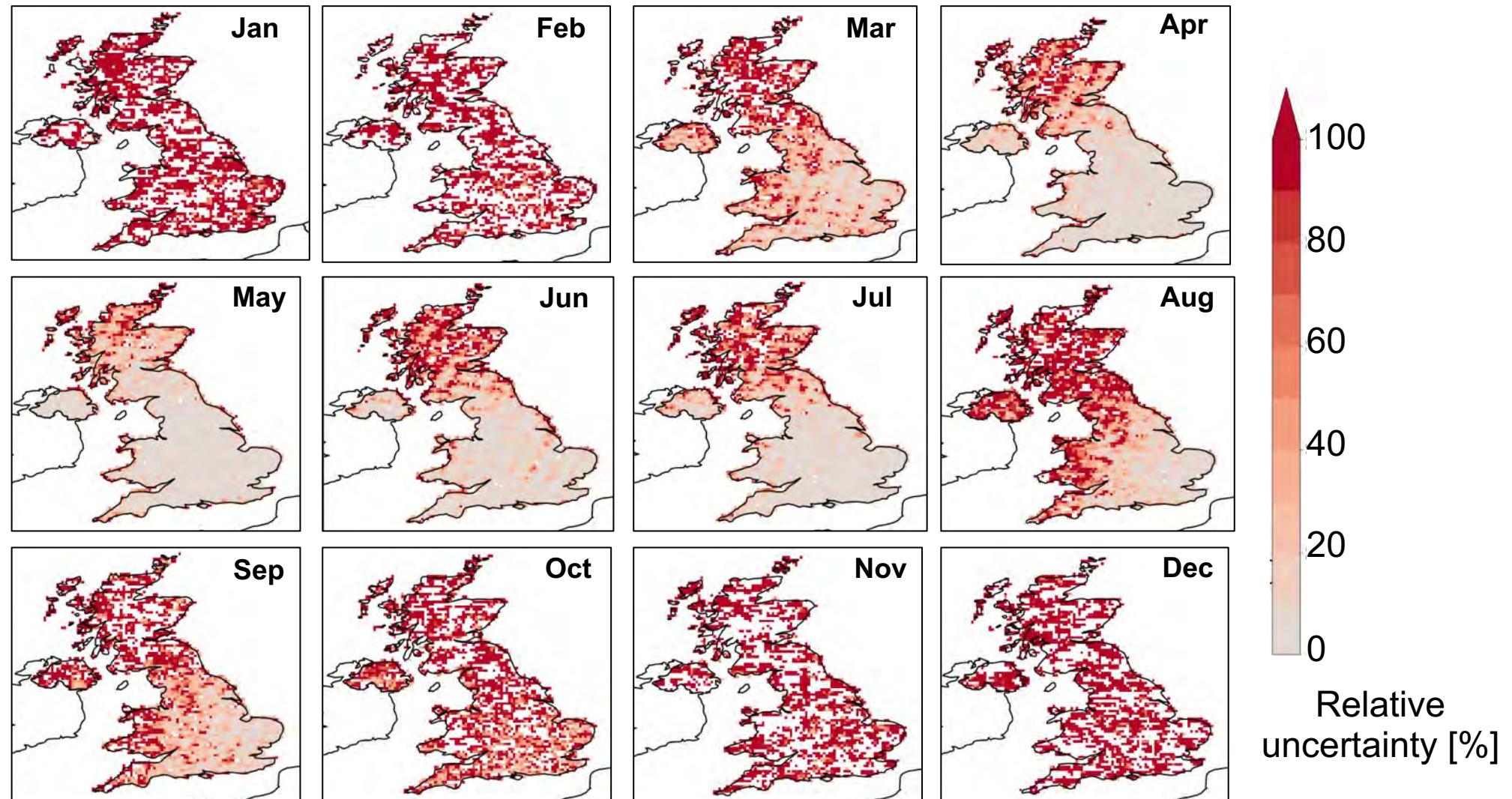
Data noisy in winter, start of spring, and end of autumn

Challenging to retrieve NH₃ in these months



Account for Observation Uncertainties

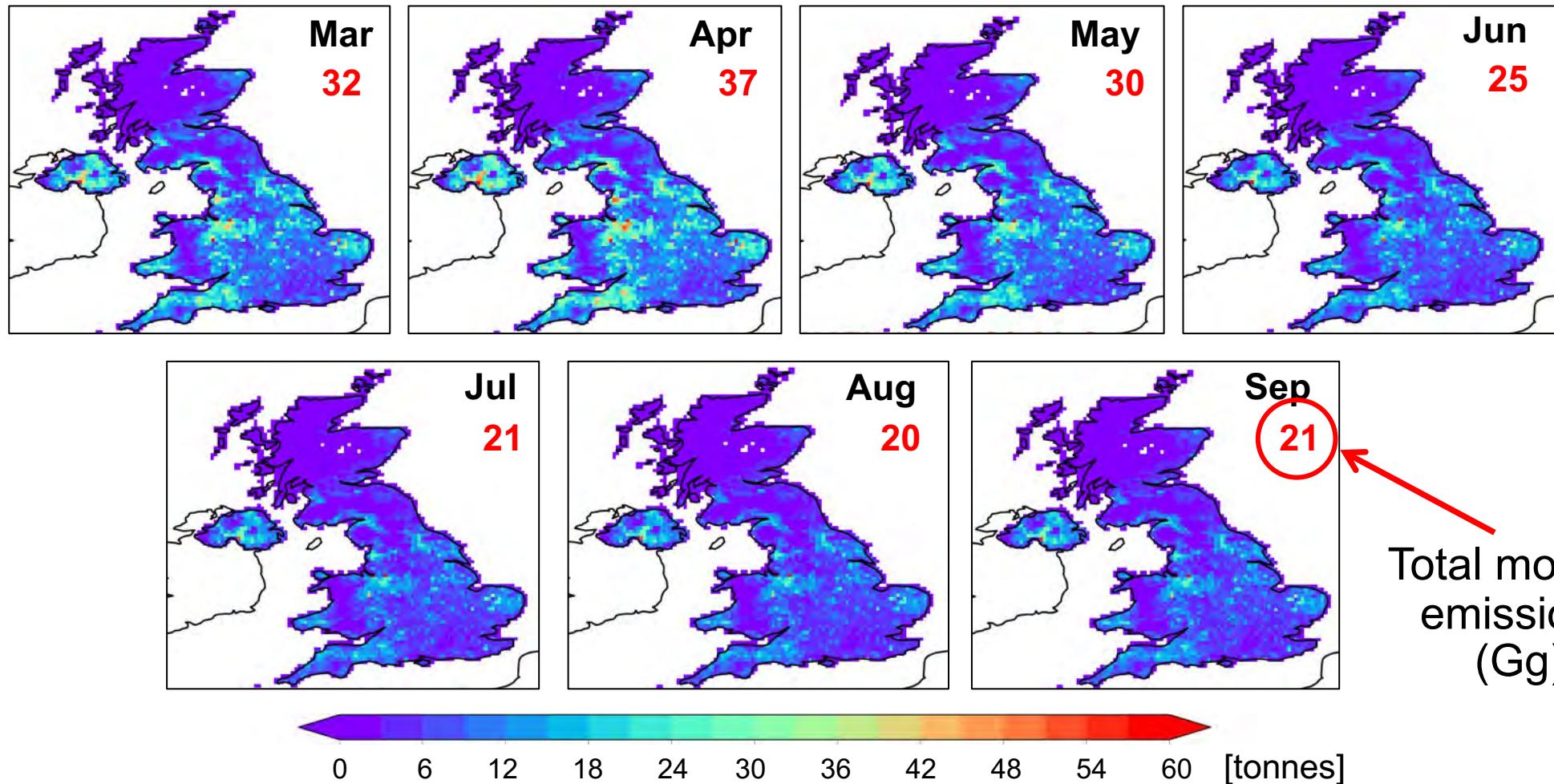
IASI NH₃ column concentrations susceptible to large uncertainties in cold months



Only consider months with relatively low uncertainty: March-September

NAEI ammonia emissions with assumed seasonality

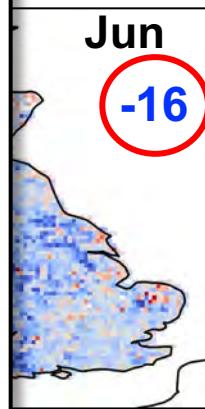
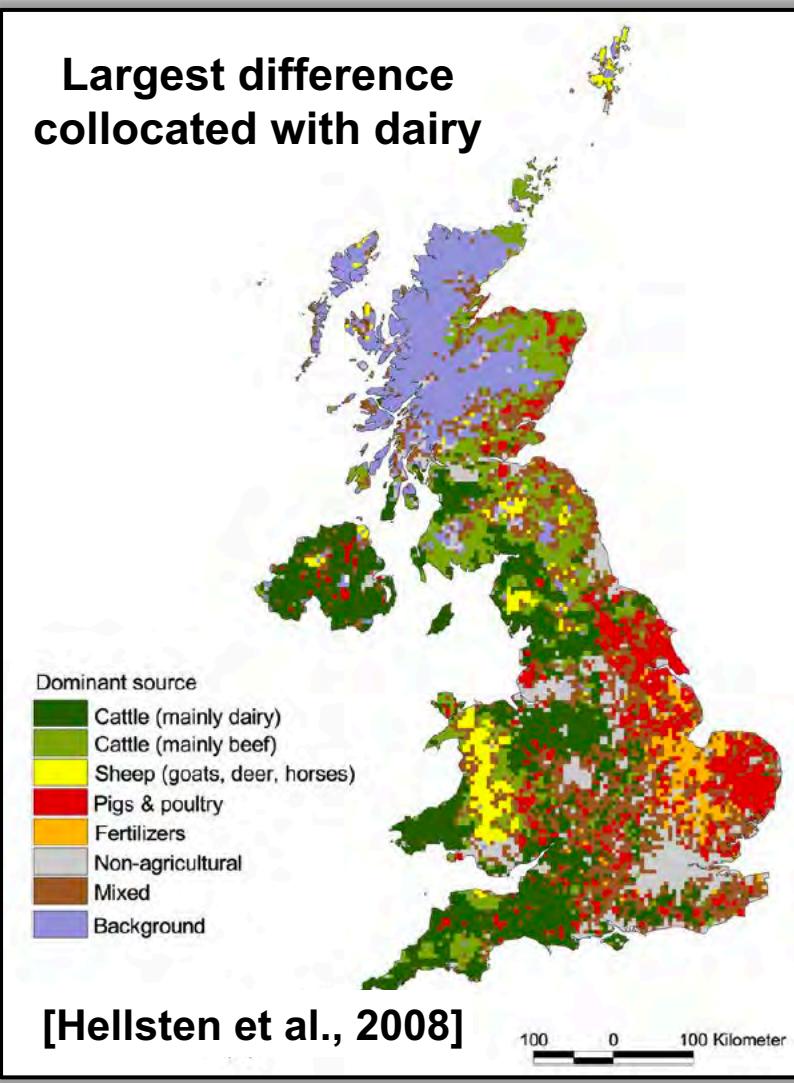
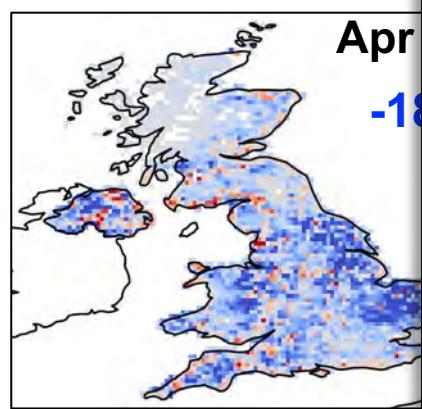
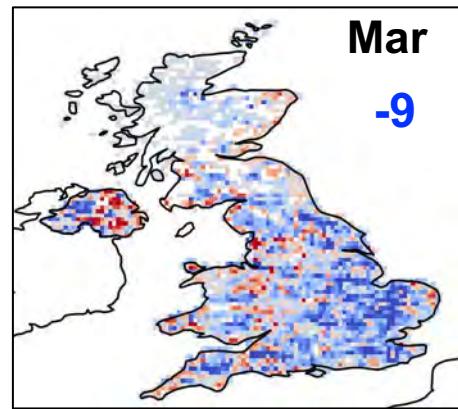
NAEI NH₃ emissions with monthly scaling factors used in GEOS-Chem applied



NAEI NH₃ emissions in March-September are 67% of annual NAEI NH₃ emissions

Assessment of the UK National Emission Inventory

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

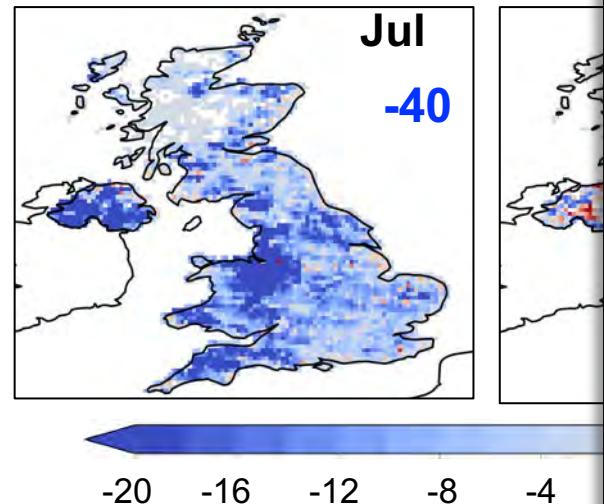


Difference in monthly emissions (Gg)

Blue: NAEI < IASI

Red: NAEI > IASI

Red mostly large uncertainties



Coincident grids:

Total NAEI: 174 Gg
(60% of 2016 total)

Total IASI: 290 Gg
NAEI 1.6 times less than IASI

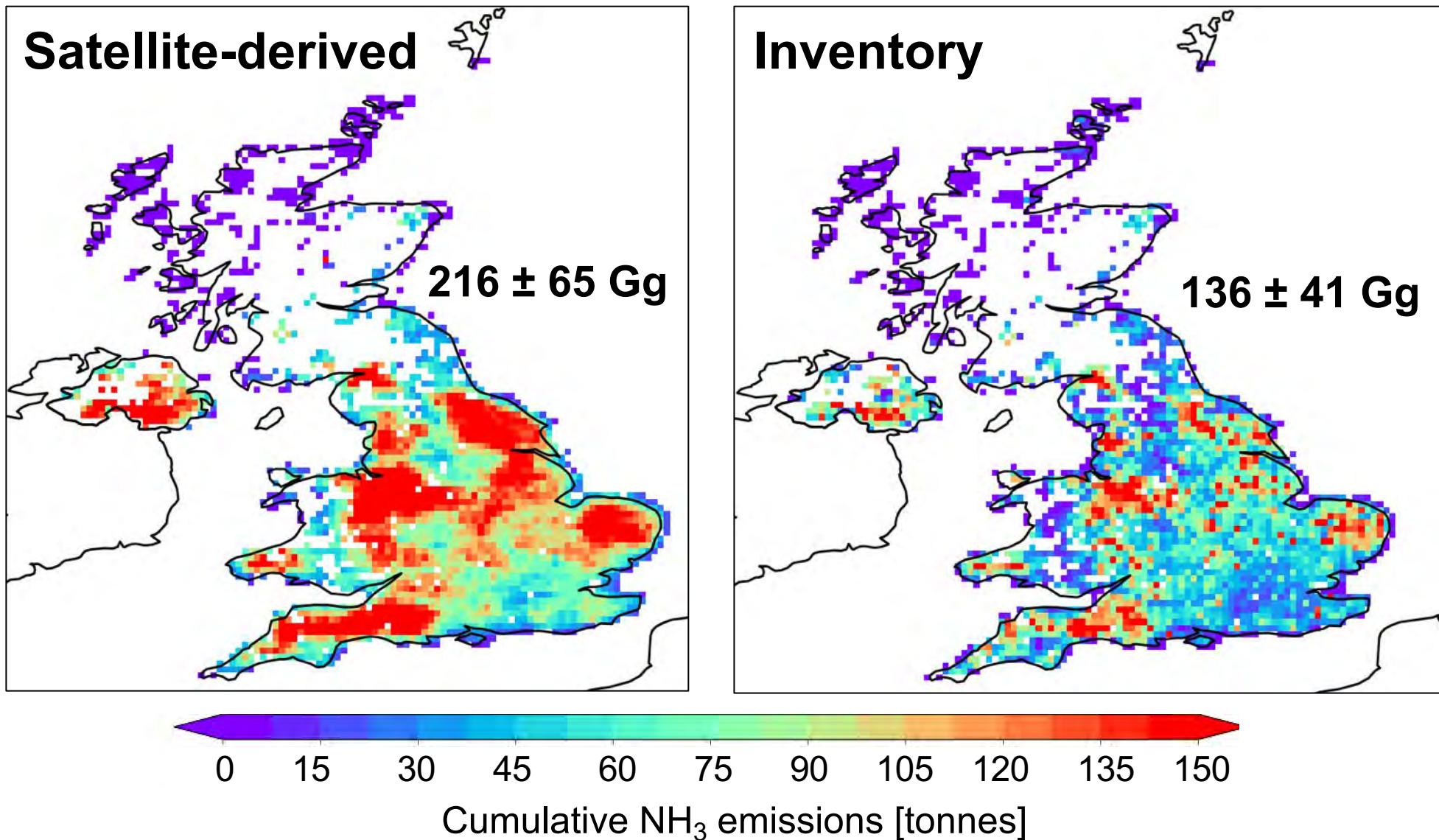
Largest discrepancy over locations dominated by **dairy farms**

Comparison of total March-September emissions

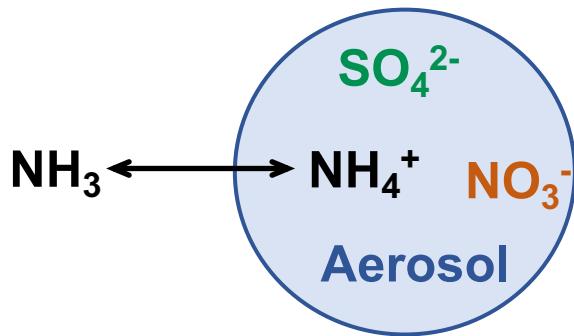
Comparison suggests satellite-derived estimate suggests NAEI underestimates emissions.

Similar relative errors

Satellite-derived emissions 60% more than inventory emissions



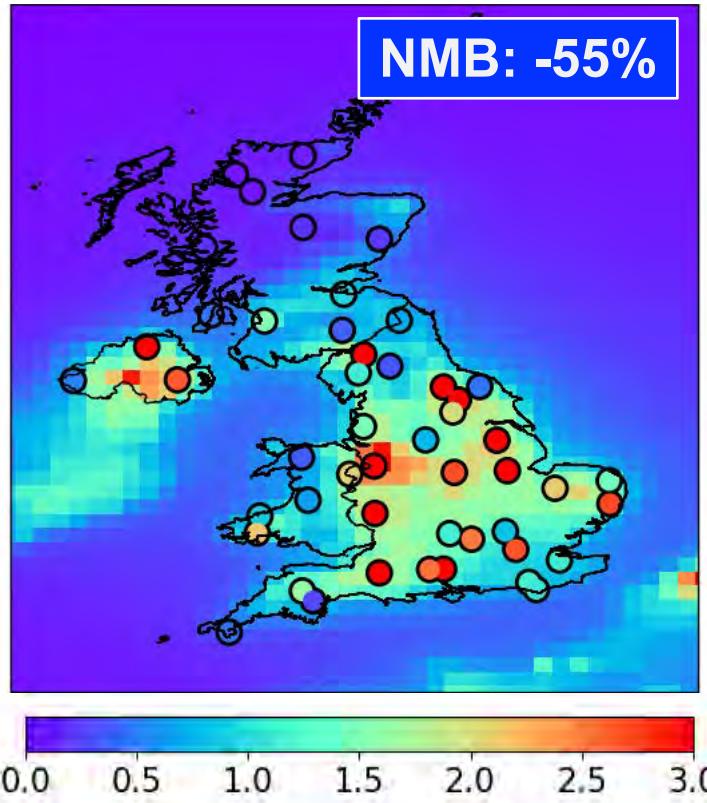
But, the results we obtain may be erroneous ...



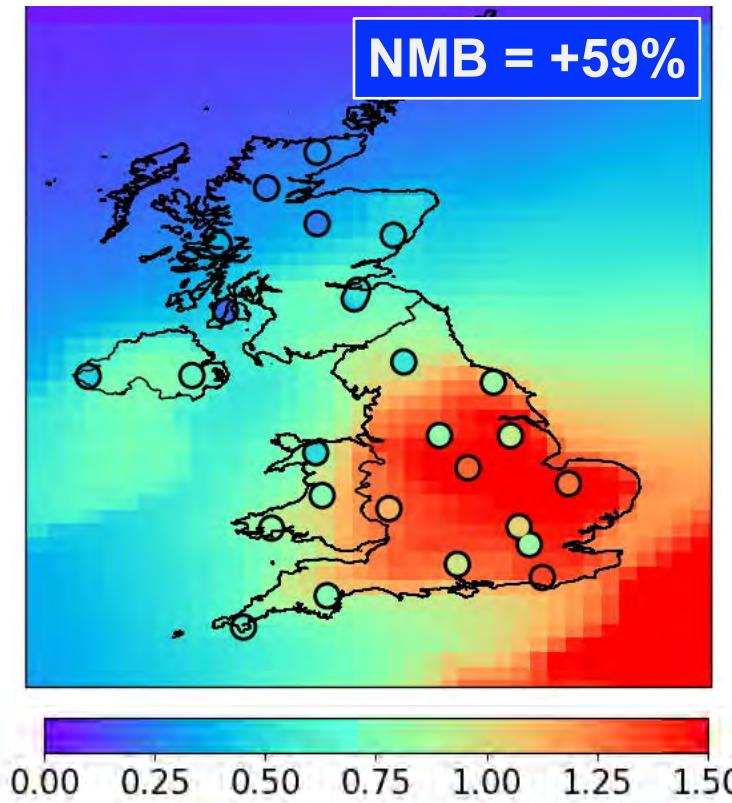
Gas-phase abundance of NH_3 depends on sulfate and nitrate

Does the model get this balance right?

Annual mean NH_3 [$\mu\text{g m}^{-3}$]



Annual mean NH_4^+ [$\mu\text{g m}^{-3}$]

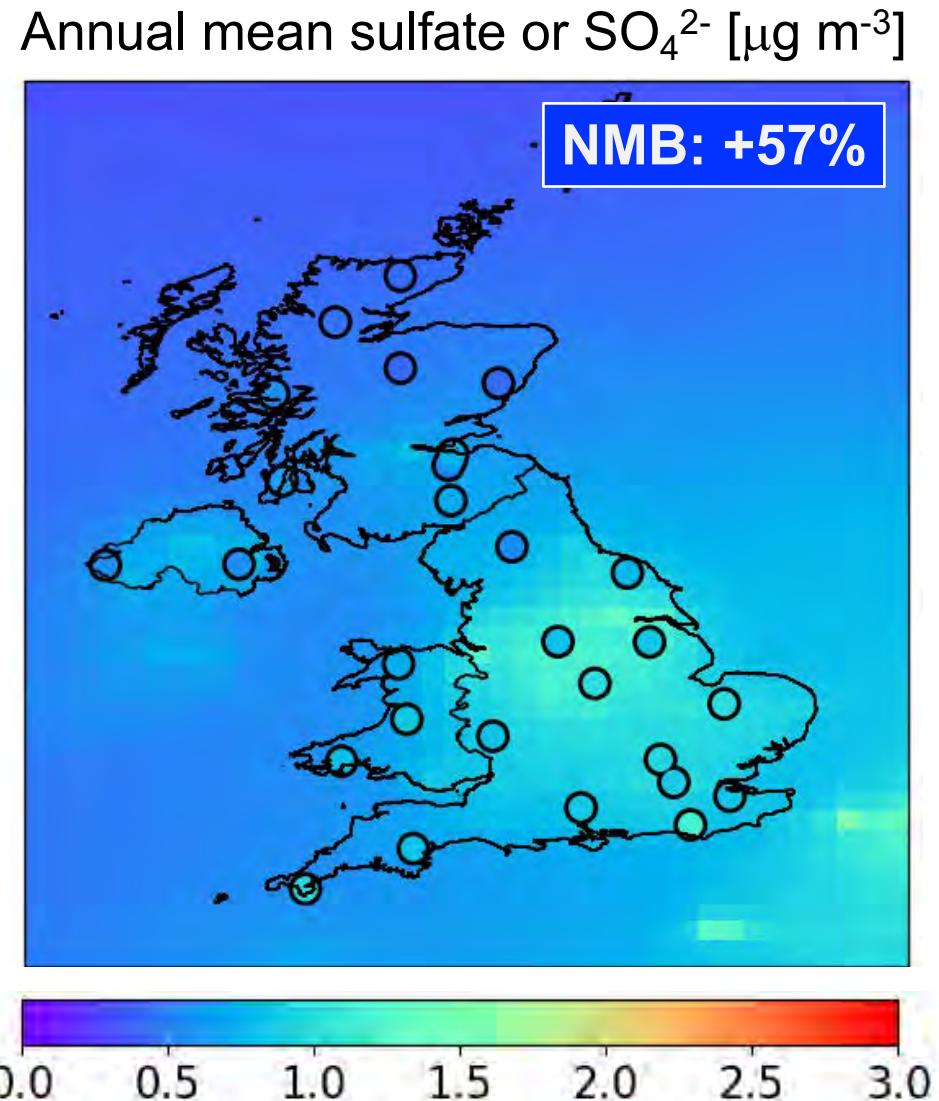
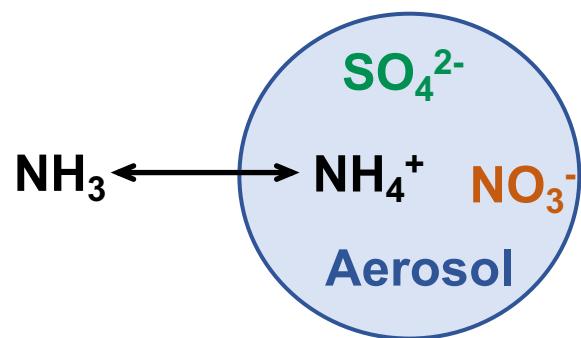


NMB: Model
normalized
mean bias

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

Model overestimates sulfate, despite decreasing SO₂ emissions

Suggests that reduction in NAEI SO₂ emissions in the model is too conservative



Also emphasizes importance
of sources and sinks in
deriving emissions from
satellite observations of NH_3

Concluding Remarks

- Developed fine spatial and temporal resolution products of NO₂ in the upper troposphere (UT) using TROPOMI
 - TROPOMI UT NO₂ reproduces spatial variability of peer-reviewed OMI product
 - Potential to address data sparsity in a profoundly important portion of the atmosphere
-
- UK NAEI overestimates SO₂ emissions by at least a factor of 2
 - Satellite-derived NH₃ emissions from IASI and GEOS-Chem are 60% more than the UK NAEI estimate
 - Largest underestimate in NAEI is over dairy farms in July
 - But, the model overestimates partitioning of NH₃ to aerosols due to remaining positive biases in NAEI SO₂ emissions