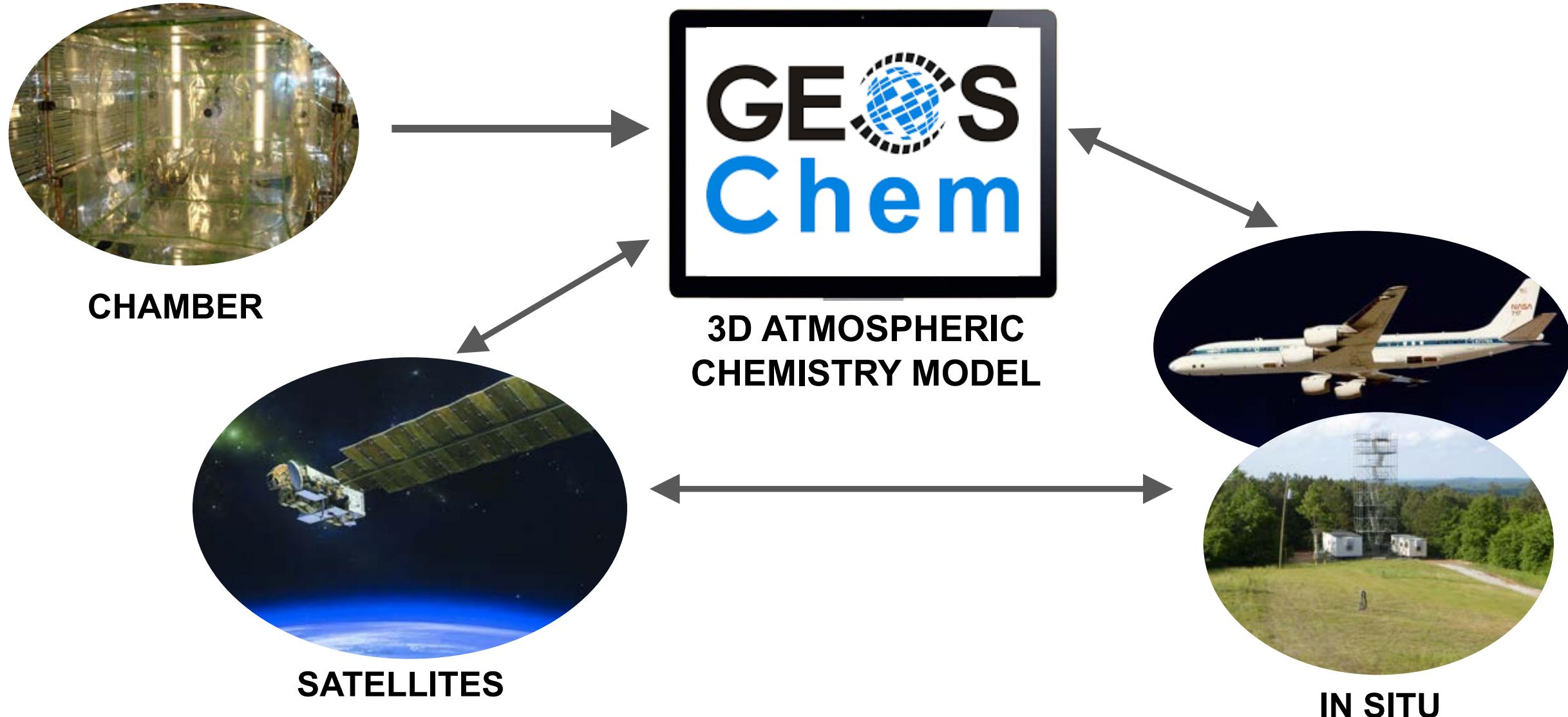


# Using satellites to determine pollutant emissions on the ground and atmospheric composition up above



# Multiplatform Approach to Solve Pressing Environmental Issues



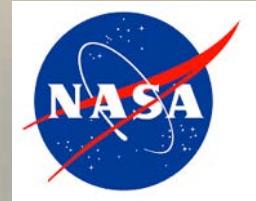
My group uses and develops state-of-science observations and tools to inform policy.

# Research Group Members



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

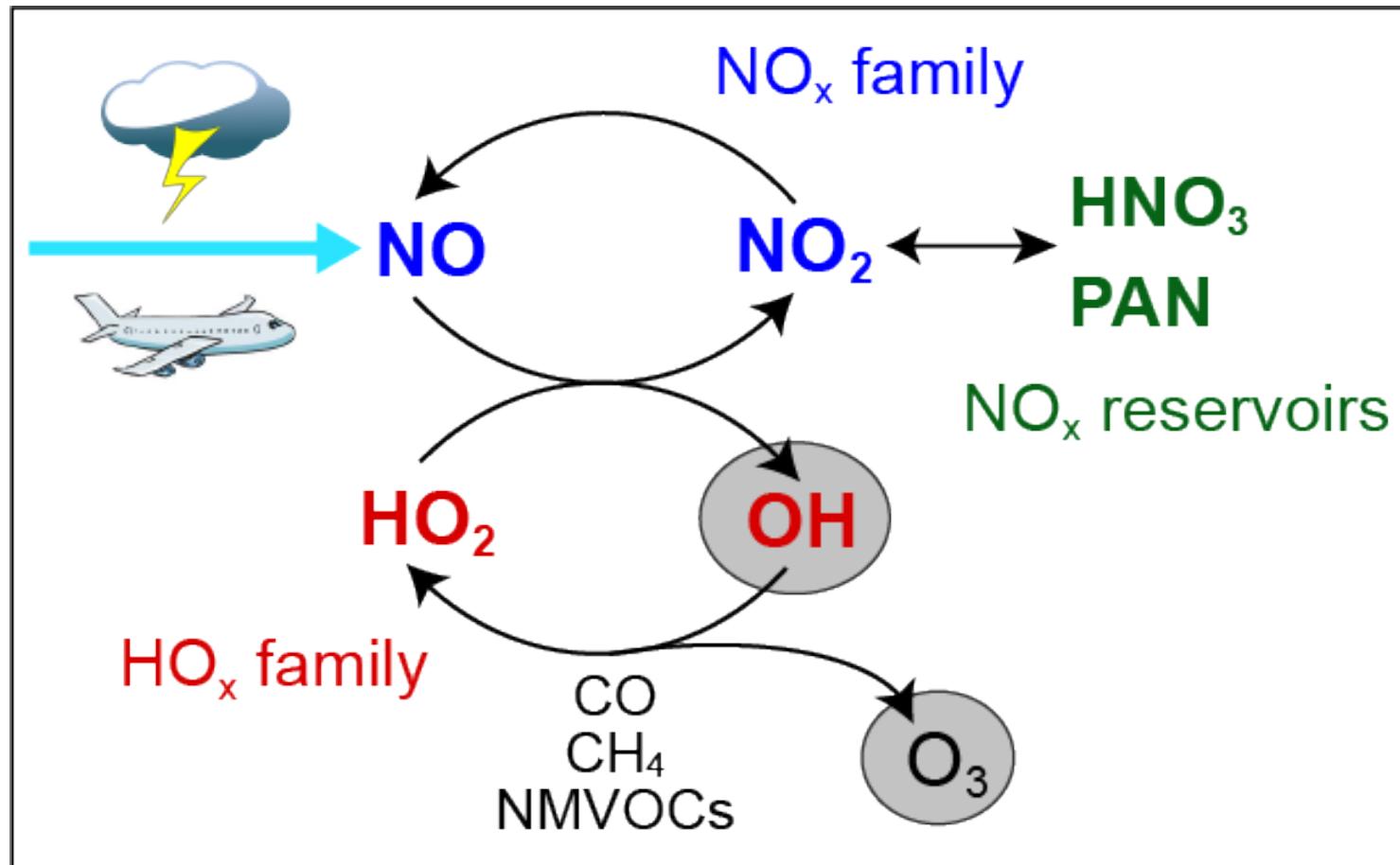
# First Estimate of Upper Tropospheric NO<sub>2</sub> from TROPOMI



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 ( $\text{NO}_x$ ) in the Upper Troposphere (8-12 km)



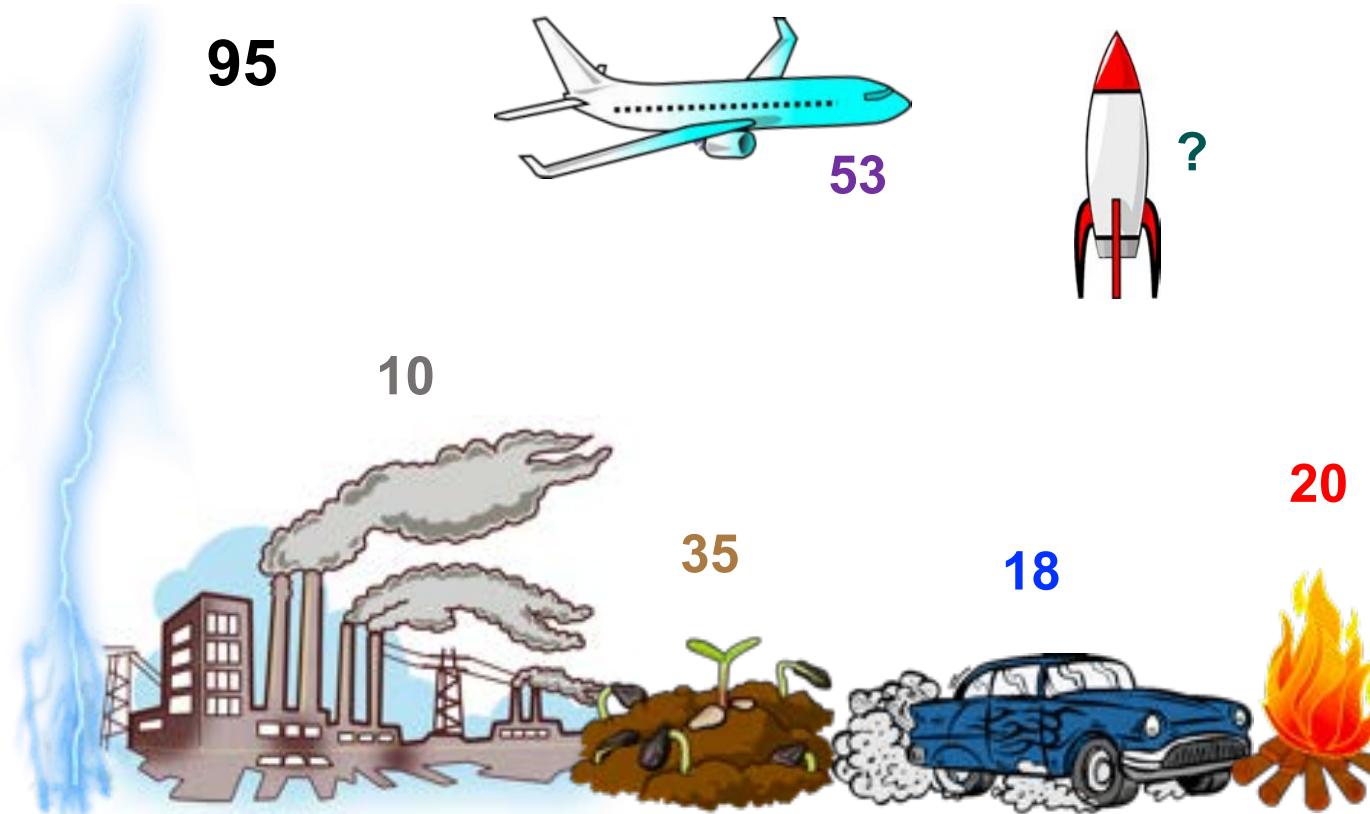
Influence atmospheric oxidants (OH, O<sub>3</sub>) and climate (O<sub>3</sub> formation, methane persistence)

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

# Influence of upper tropospheric NO<sub>x</sub>

NO<sub>x</sub> in the upper troposphere is very efficient at producing the greenhouse gas ozone

## OPE (molecules O<sub>3</sub>/molecule NO<sub>x</sub>) for individual NO<sub>x</sub> sources



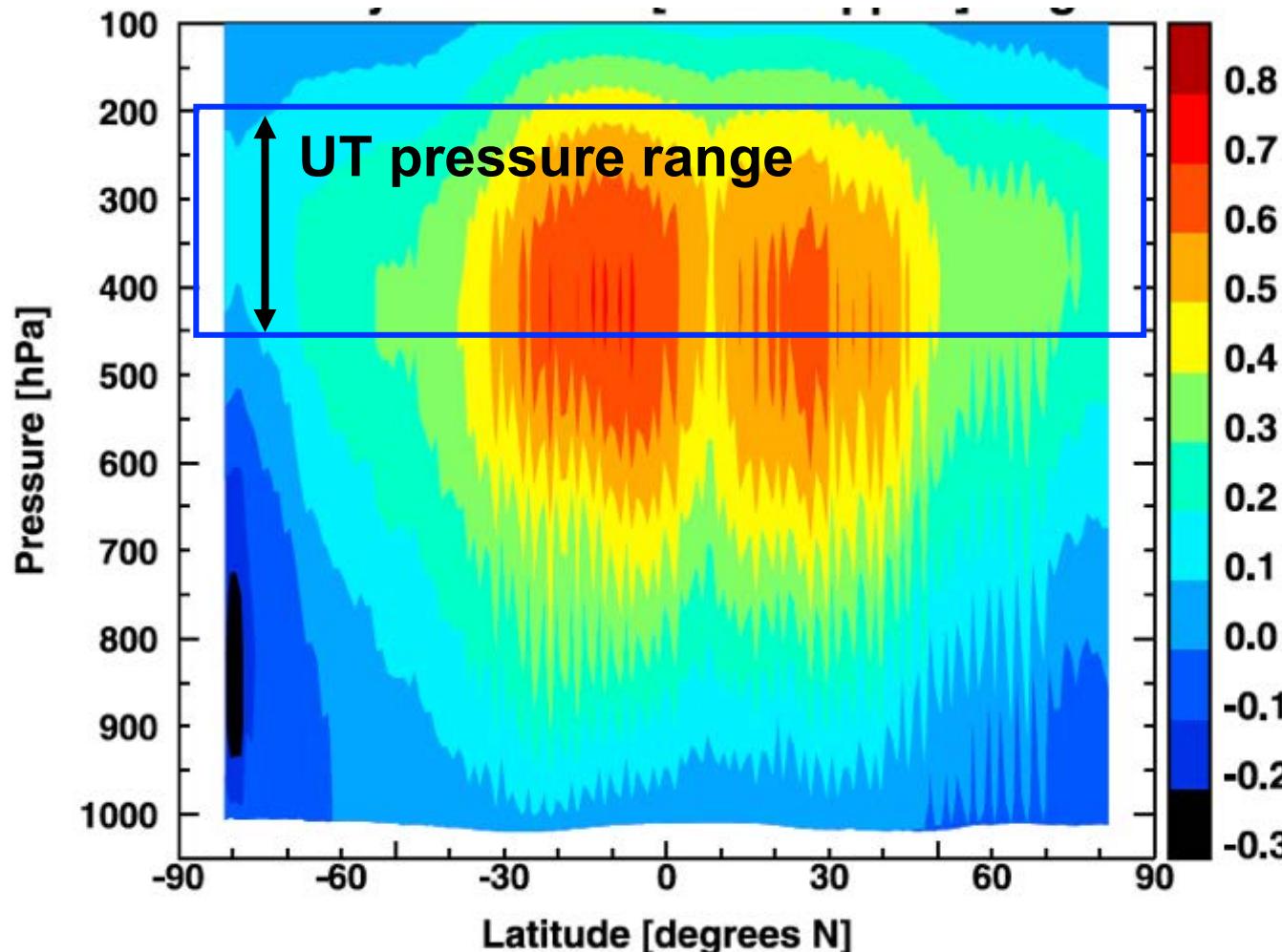
[adapted from Dahlmann et al., 2011]

Longer NO<sub>x</sub> lifetime at higher altitude → greater OPE

# Influence of upper tropospheric NO<sub>x</sub>

The warming potential of ozone peaks the upper troposphere

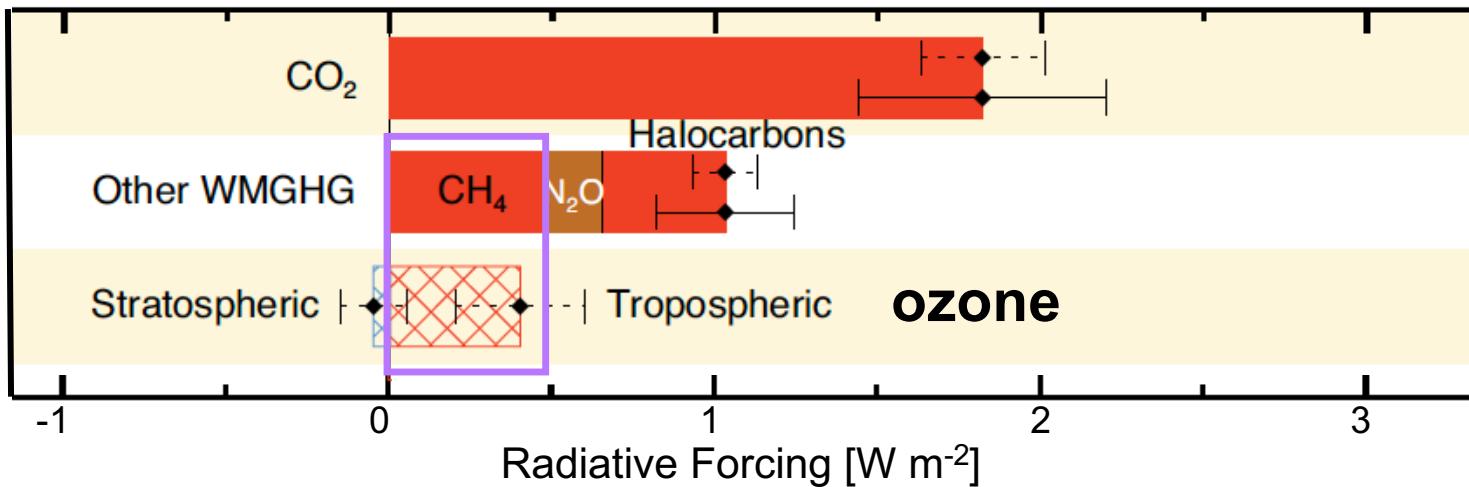
## Sensitivity of outgoing longwave radiation to variations in ozone concentrations



# Why uncertainties in the upper troposphere matter

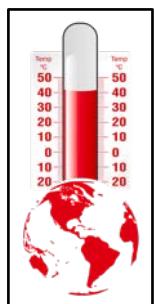
Warming due to tropospheric ozone is similar to that of methane ( $\text{CH}_4$ )

Tropospheric ozone and methane have near-equal climate impacts

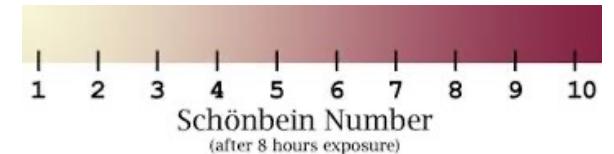


[IPCC AR5, 2013]

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



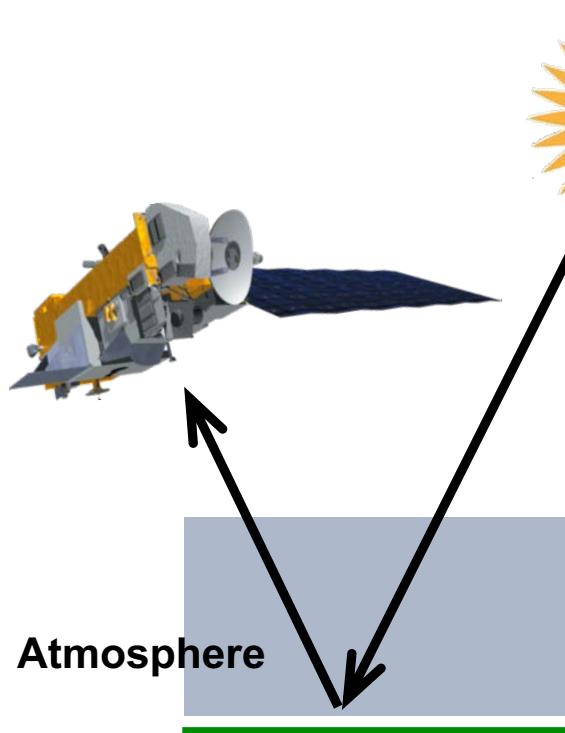
Errors in tropospheric ozone radiative forcing



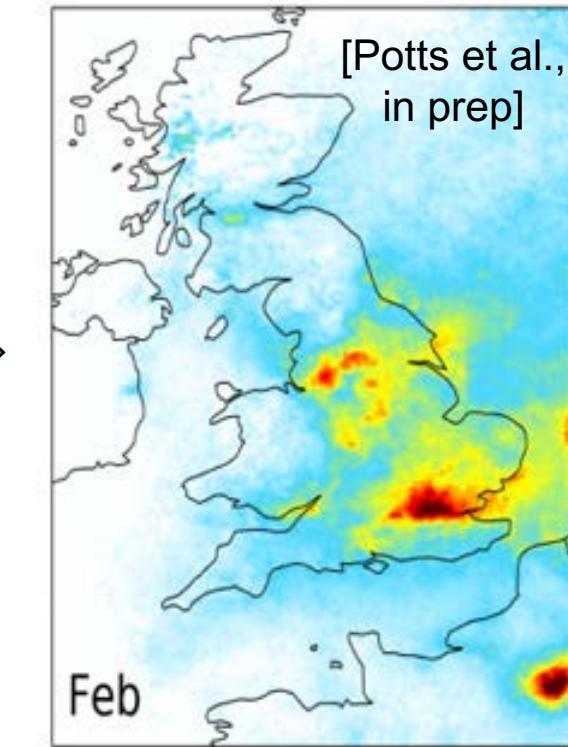
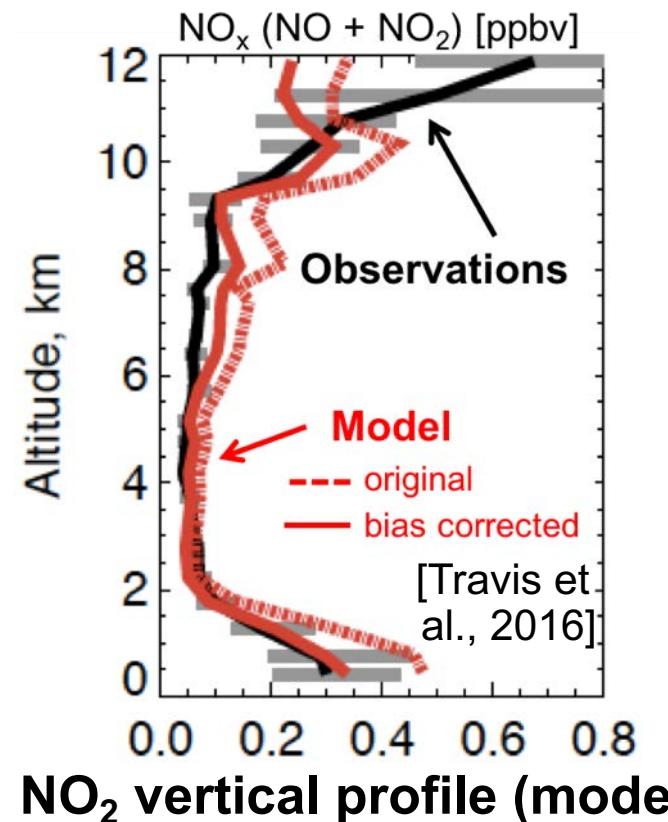
Indicator paper used to measure ozone in the mid to late 19<sup>th</sup> century

# Why uncertainties in the upper troposphere matter

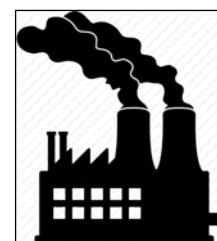
We rely on models to retrieve atmospheric composition from satellite instruments



**NO<sub>2</sub> along the viewing path**



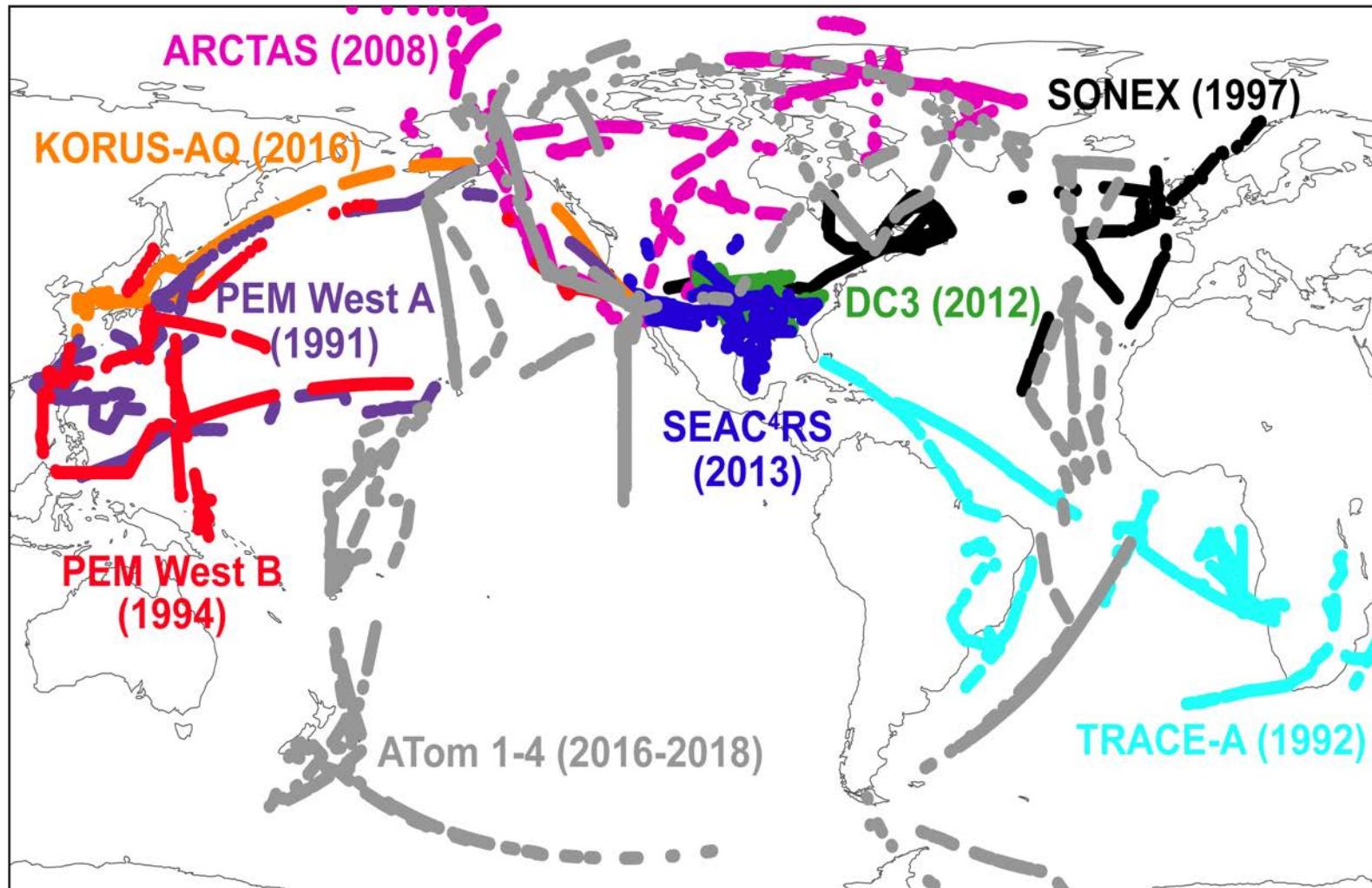
**Vertical column densities**



**Uncertain global air quality constraints from satellites**

# Sampling of the upper troposphere is limited

NASA DC8 research aircraft flight tracks in the upper troposphere



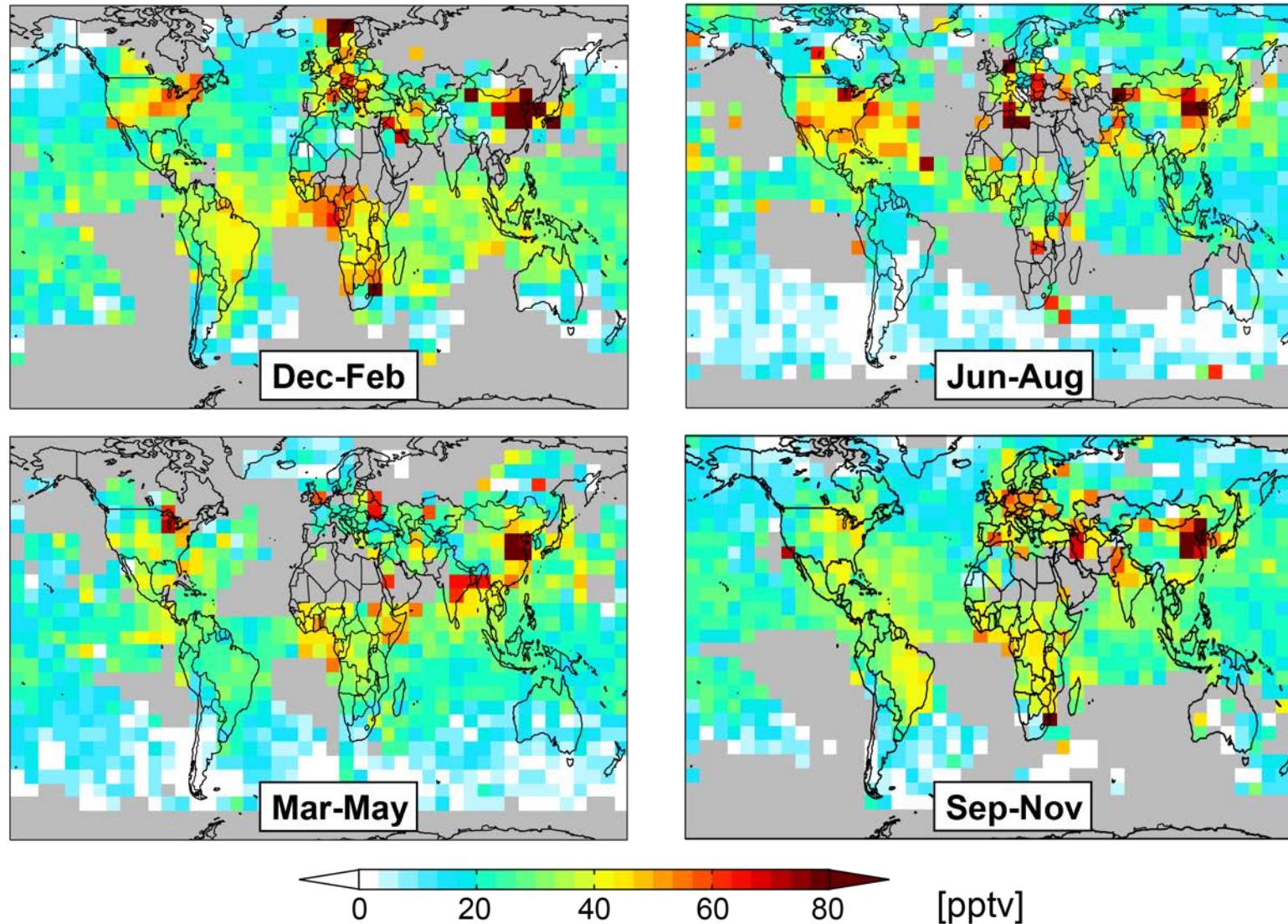
There are also measurements from commercial aircraft

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

# Products of upper tropospheric NO<sub>2</sub> from satellite observations

Near global spatial coverage of seasonal mean UT NO<sub>2</sub> at  $5^\circ \times 8^\circ$  (50 km  $\times$  80 km)

NASA OMI upper troposphere NO<sub>2</sub> (2005-2007)



**OMI:**  
Ozone Monitoring  
Instrument

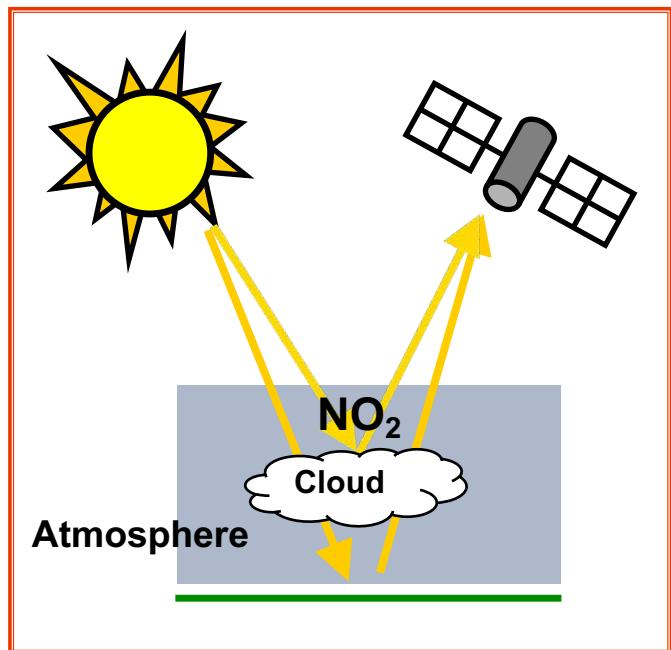
[Marais et al., 2018;  
Choi et al., 2014]

# Satellite products obtained using the cloud-slicing technique

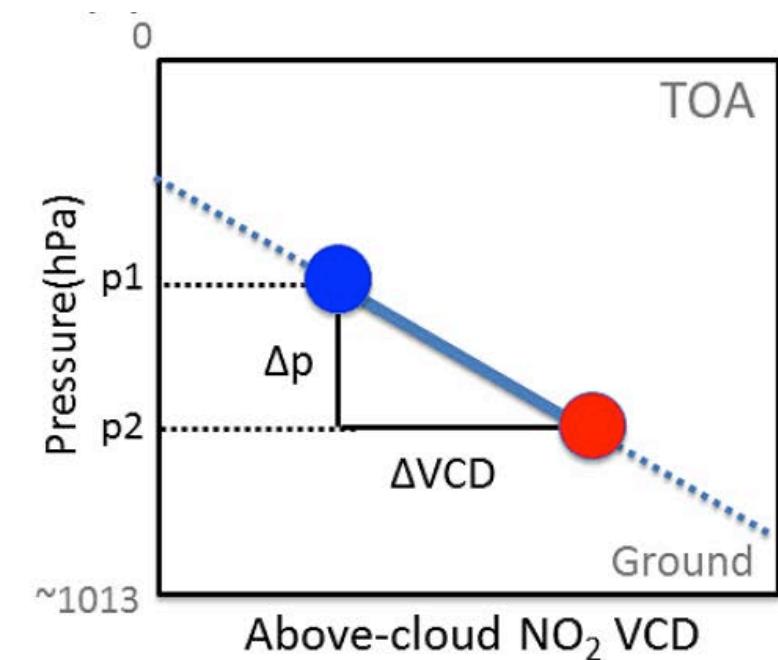
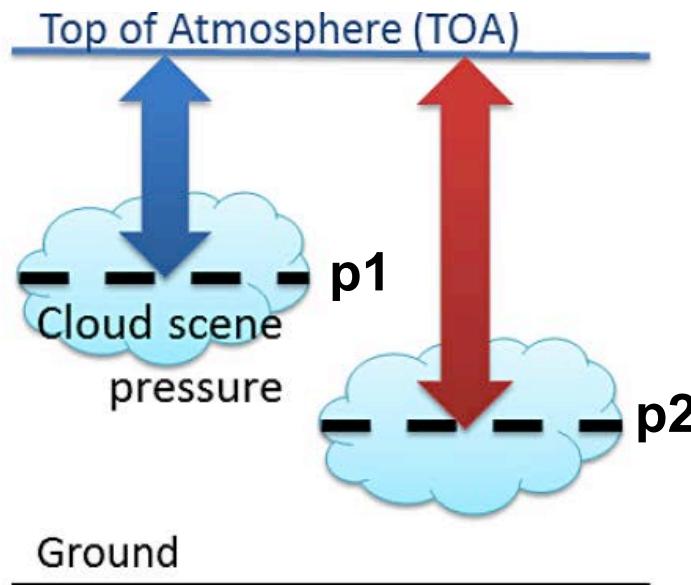
First applied by Ziemke et al. [2001] to TOMS ozone

Retrieve partial NO<sub>2</sub> columns over cloudy scenes at different heights

## APPROACH



## Use cloud height variability to derive partial columns



[adapted from Choi et al., 2014]

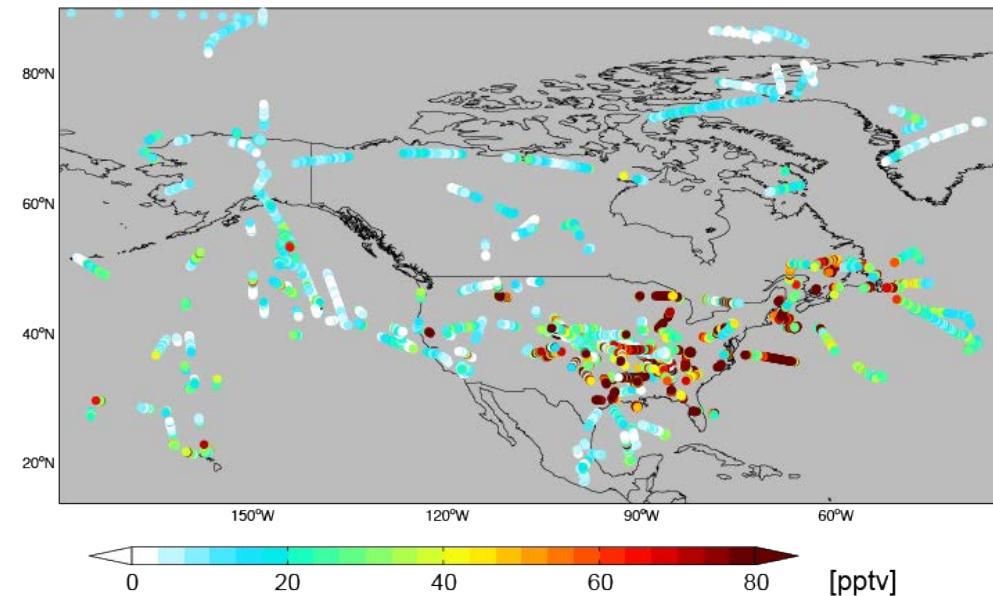
NO<sub>2</sub> volume mixing ratio (VMR) between clouds at p1 and p2

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

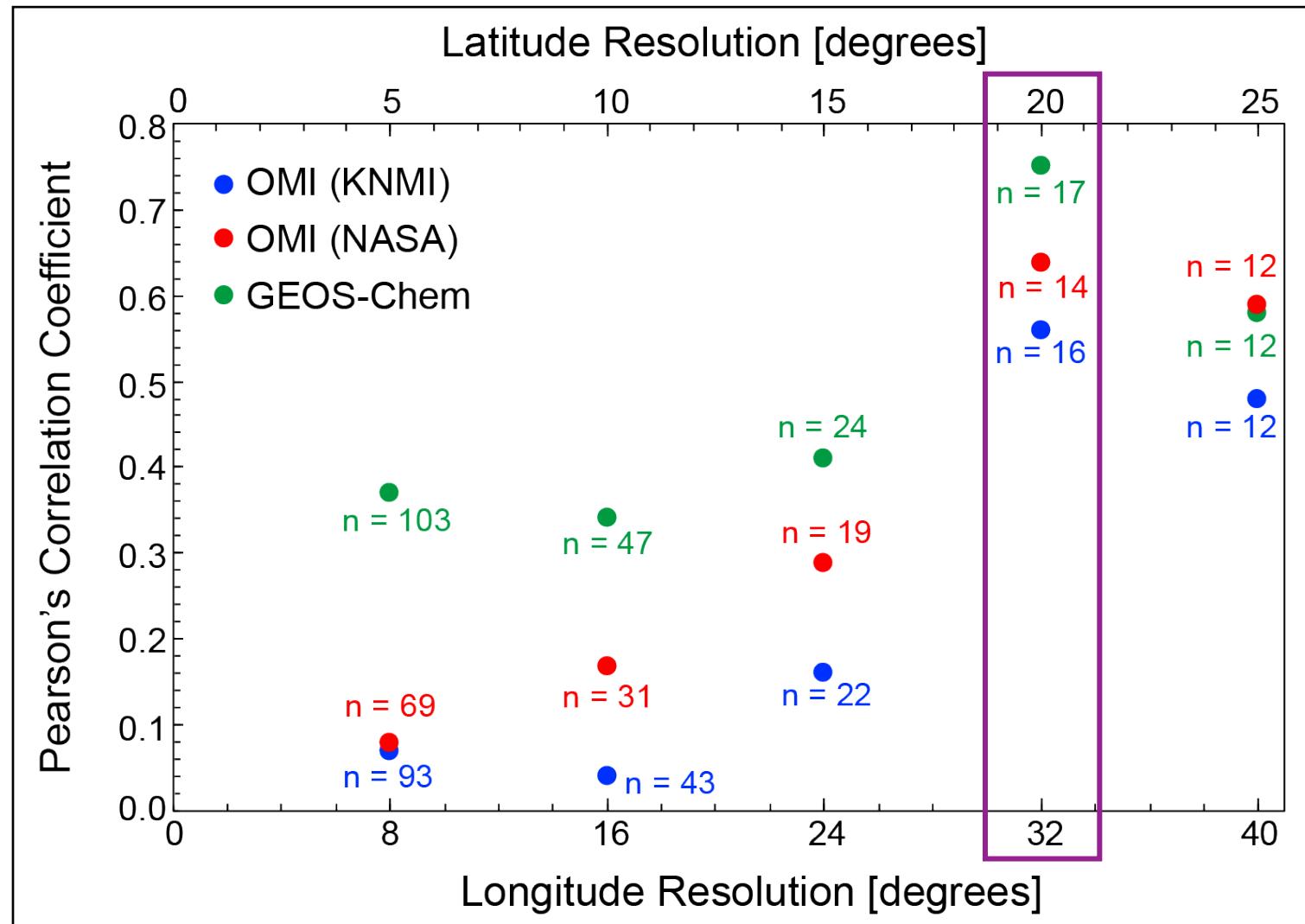
# Consistency with aircraft observations at coarse scales

Agreement in spatial distribution between (reliable) aircraft and OMI UT NO<sub>2</sub> observations, but at coarse scales (seasonal, 20° latitude (200 km) × 32° longitude (320 km))

NASA DC8 NO<sub>2</sub> in Spring-Summer

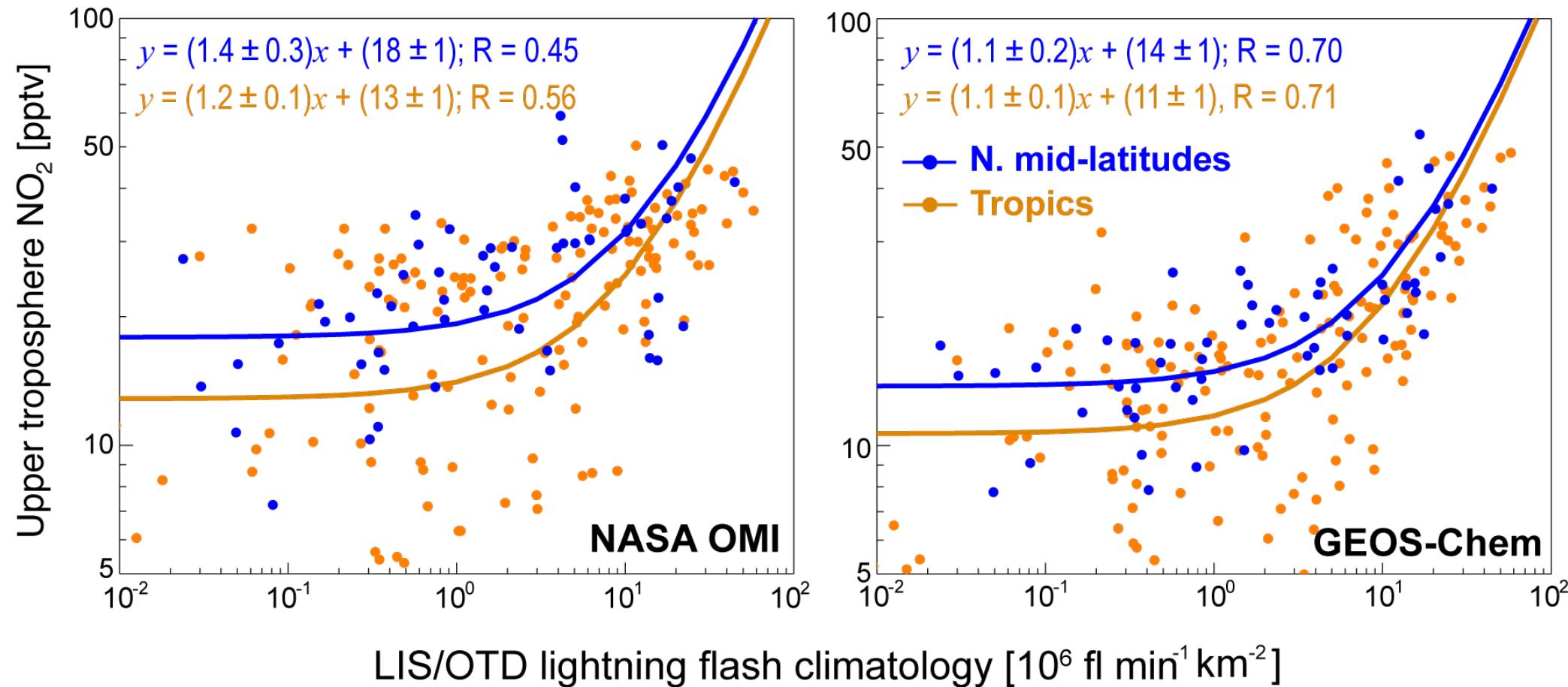


[Marais et al., 2018]



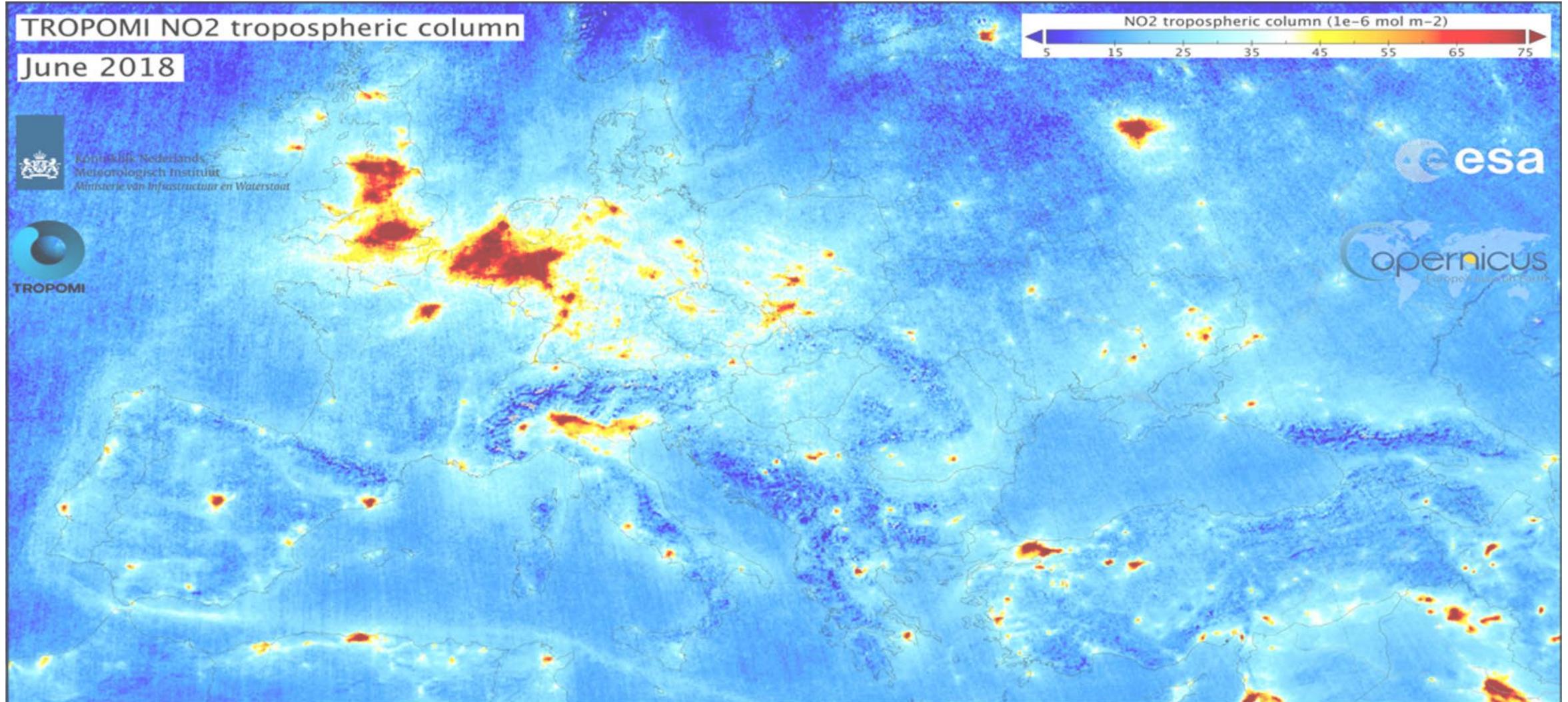
# Provide improved constraints on lightning NO<sub>x</sub> emissions

Log-log relationship between UT NO<sub>2</sub> 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<sub>x</sub> production rates

# Can we do better with high-resolution TROPOMI measurements?

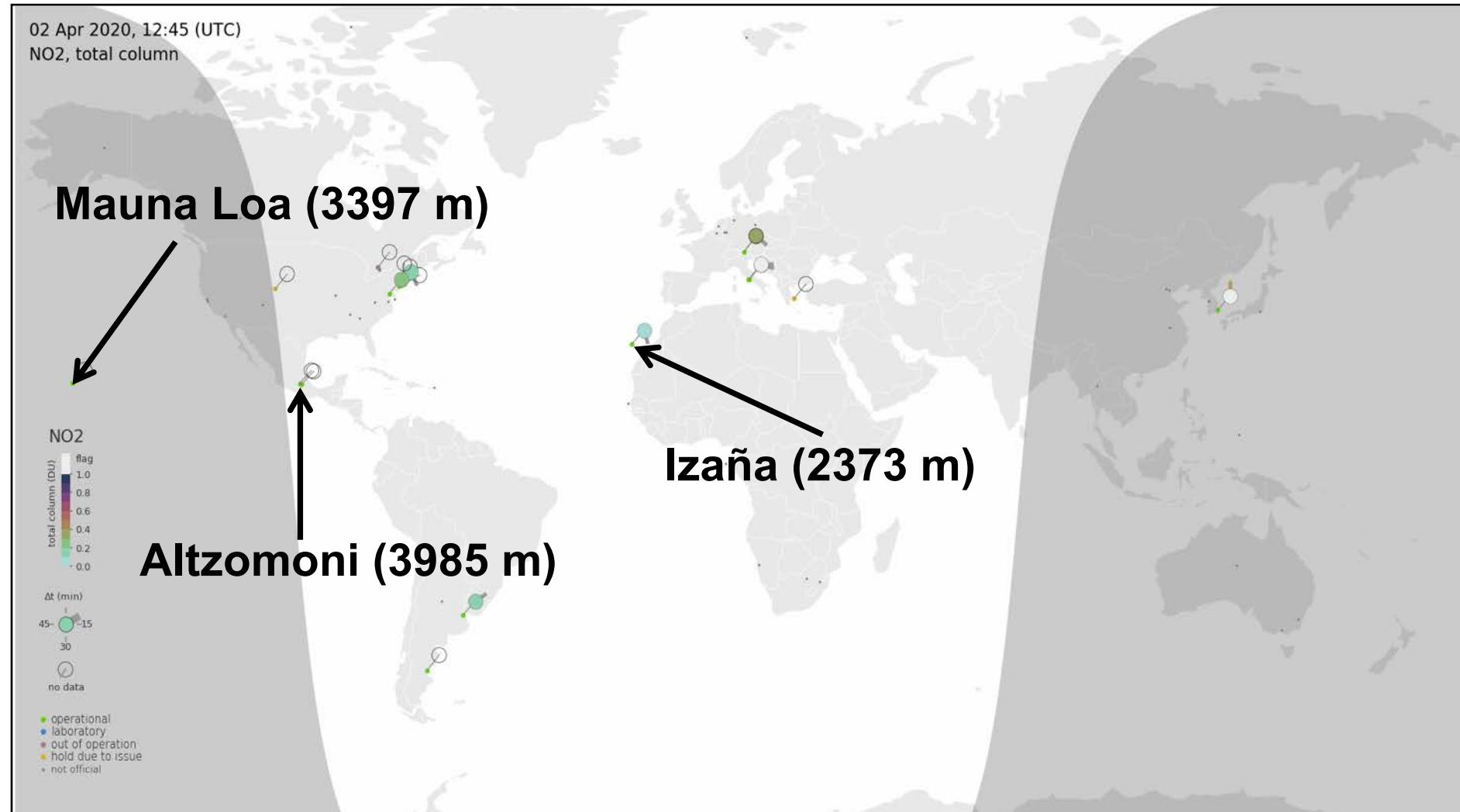


[Source: <http://www.tropomi.eu/data-products/nitrogen-dioxide>]

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

# Evaluate TROPOMI with ground-based measurements

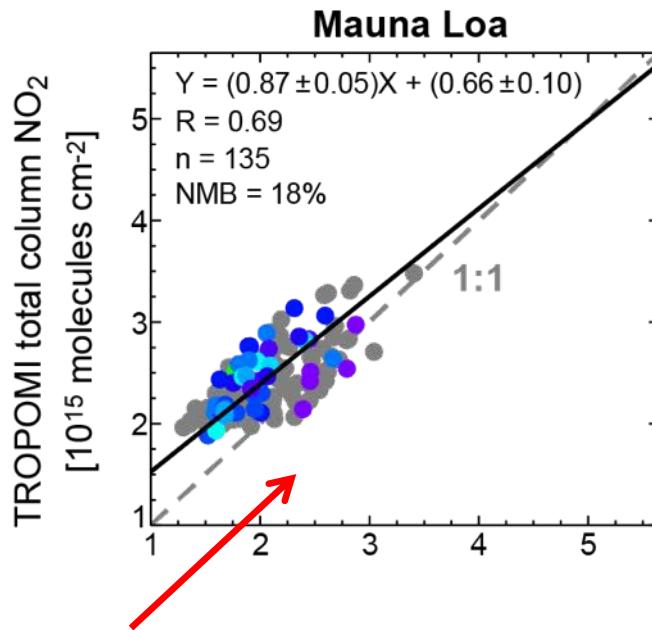
Global Pandora network, indicating locations of high-altitude sites (large relative contribution from the UT) used to evaluate TROPOMI



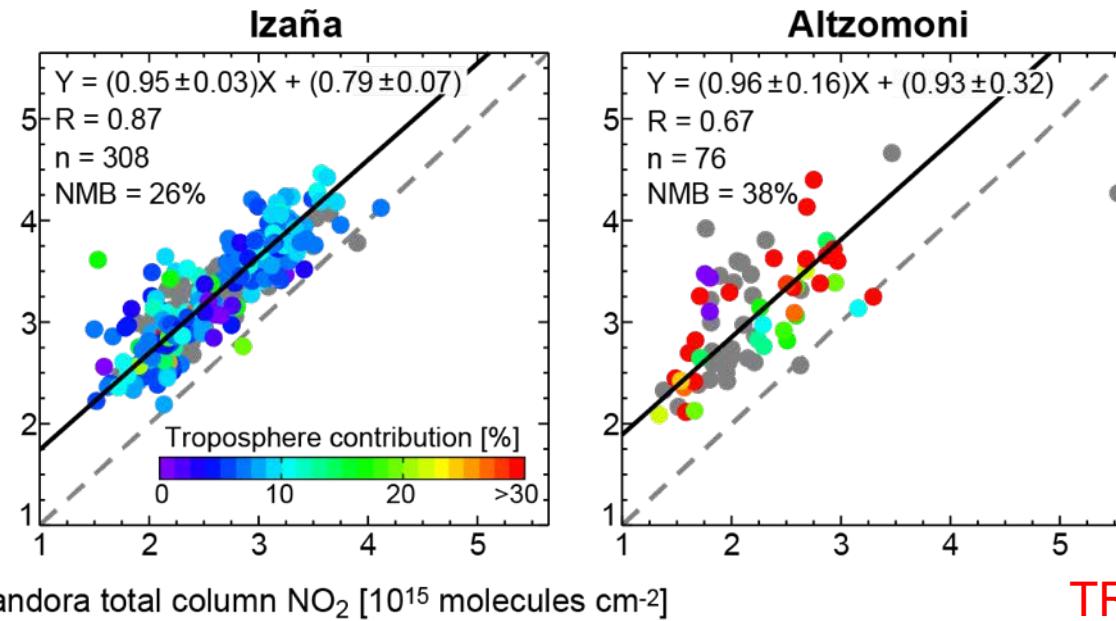
[Source: <https://www.pandoria-global-network.org/>]

# Evaluate TROPOMI with ground-based measurements

## Stratosphere:



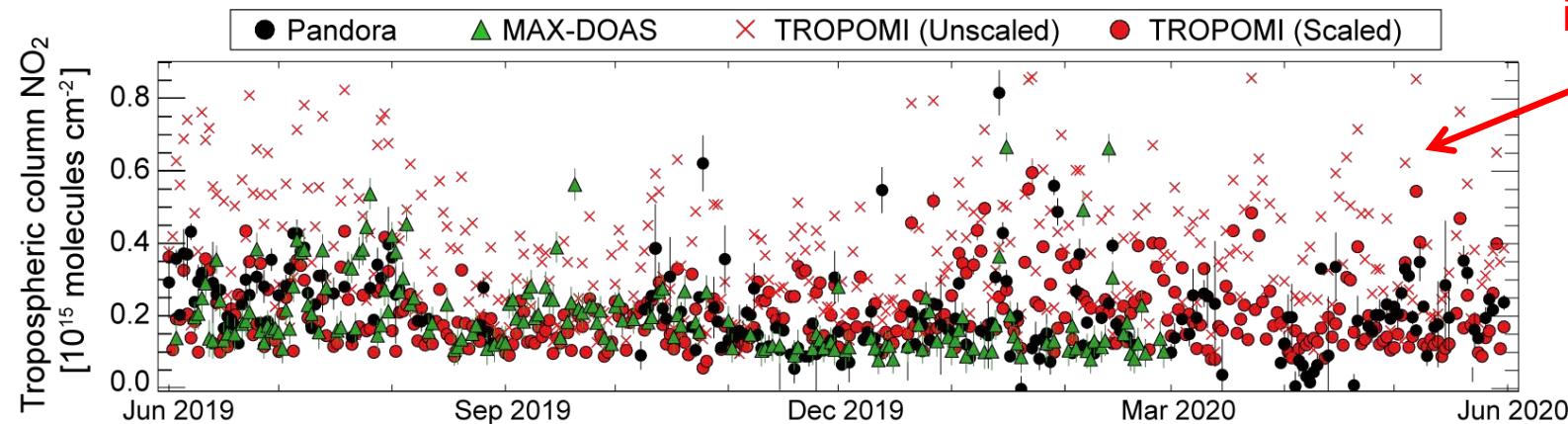
## Stratosphere + free troposphere:



TROPOMI  
stratospheric column  
variance is  
underestimated

TROPOMI free  
tropospheric column  
is overestimated

## Free troposphere:



# TROPOMI cloud-sliced upper tropospheric NO<sub>2</sub>

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

Range: 30-80 pptv

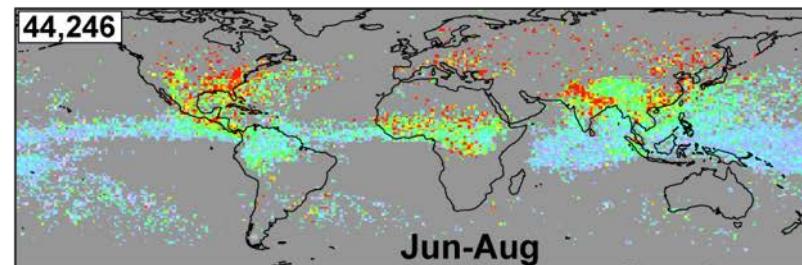
Background: ~30 pptv

Cloud products give similar UT NO<sub>2</sub> in the tropics

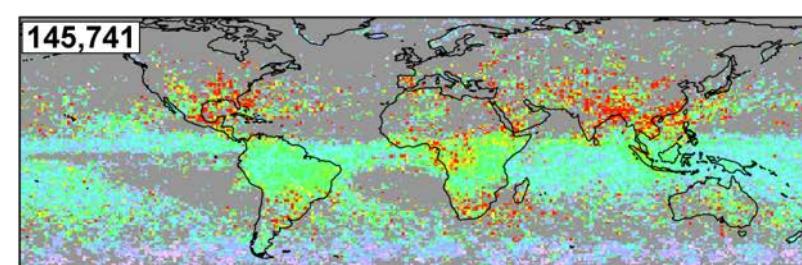
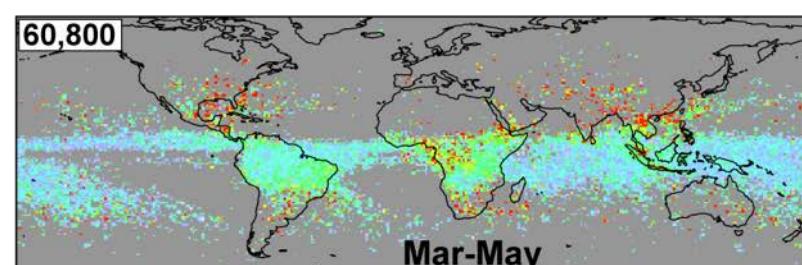
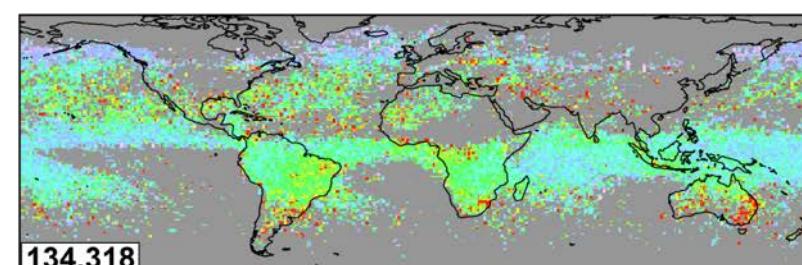
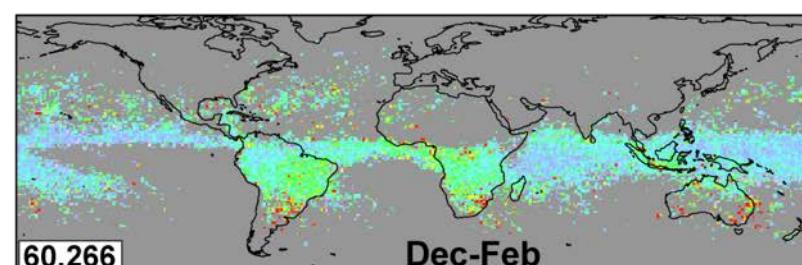
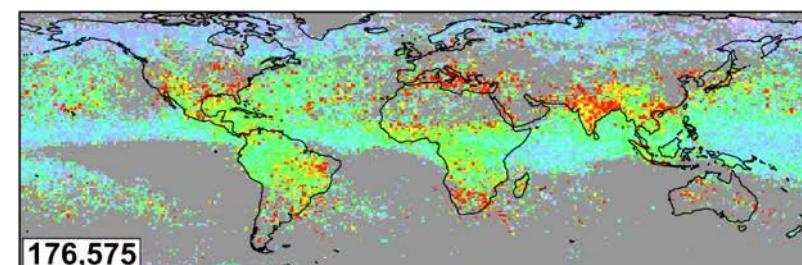
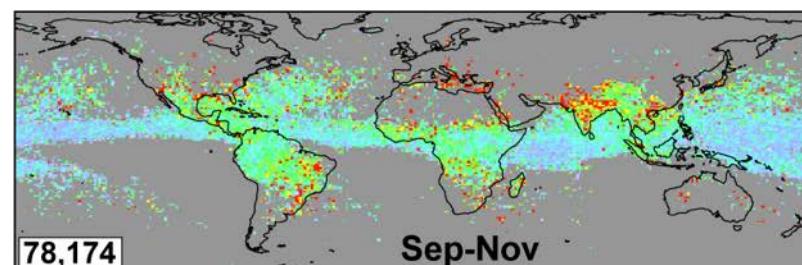
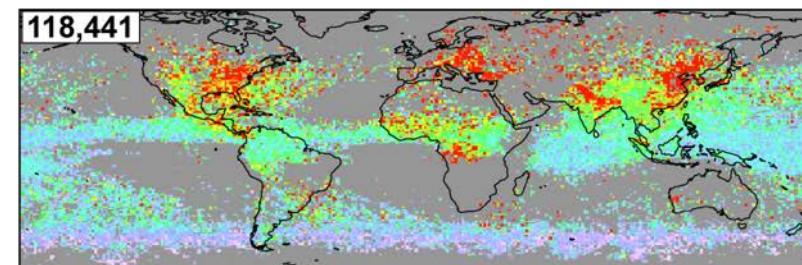
Cloud product two gives greater global coverage

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

Cloud Product One

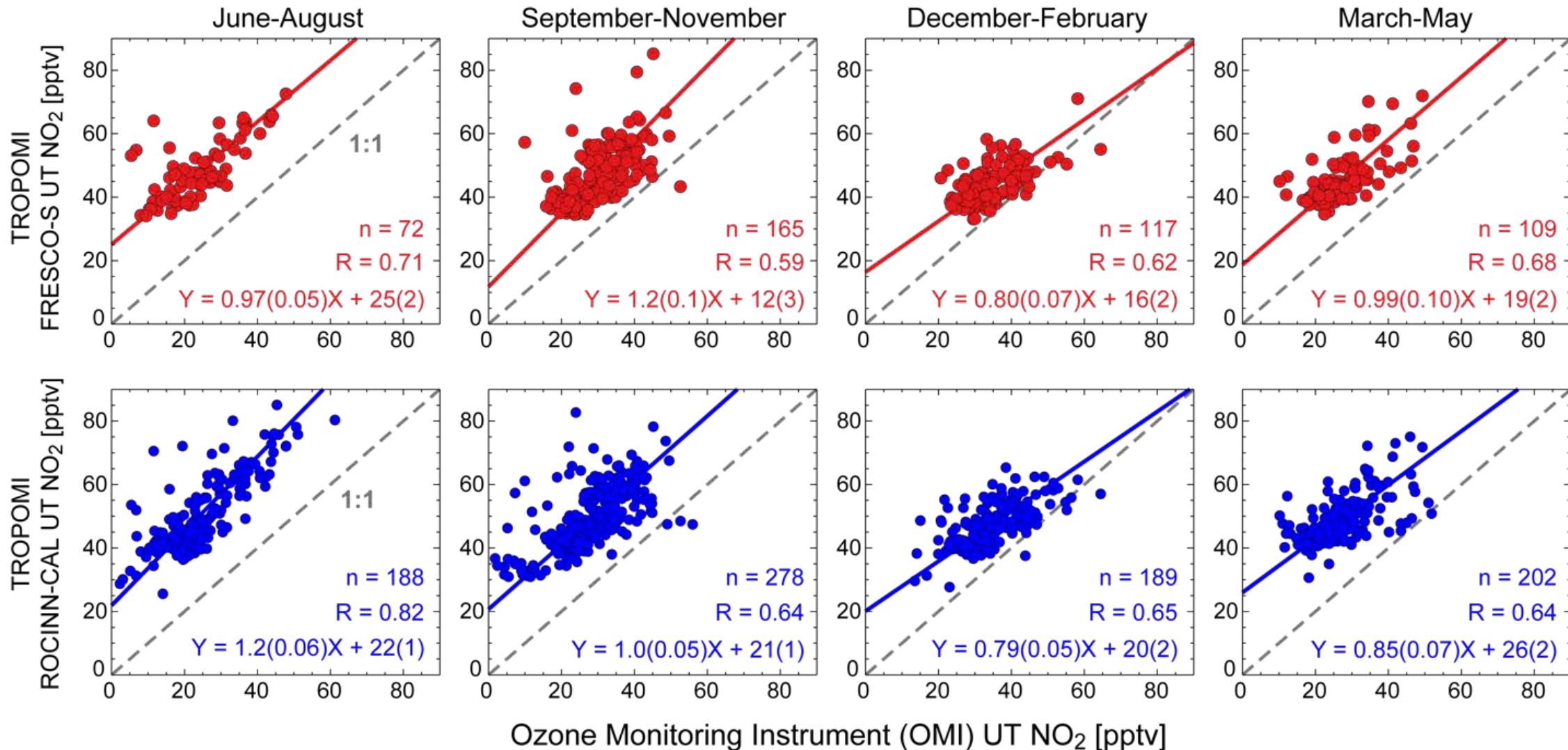


Cloud Product Two



# Comparison to the NASA OMI product

TROPOMI UT NO<sub>2</sub> 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 NO<sub>2</sub> profile)

**Back to Earth...**

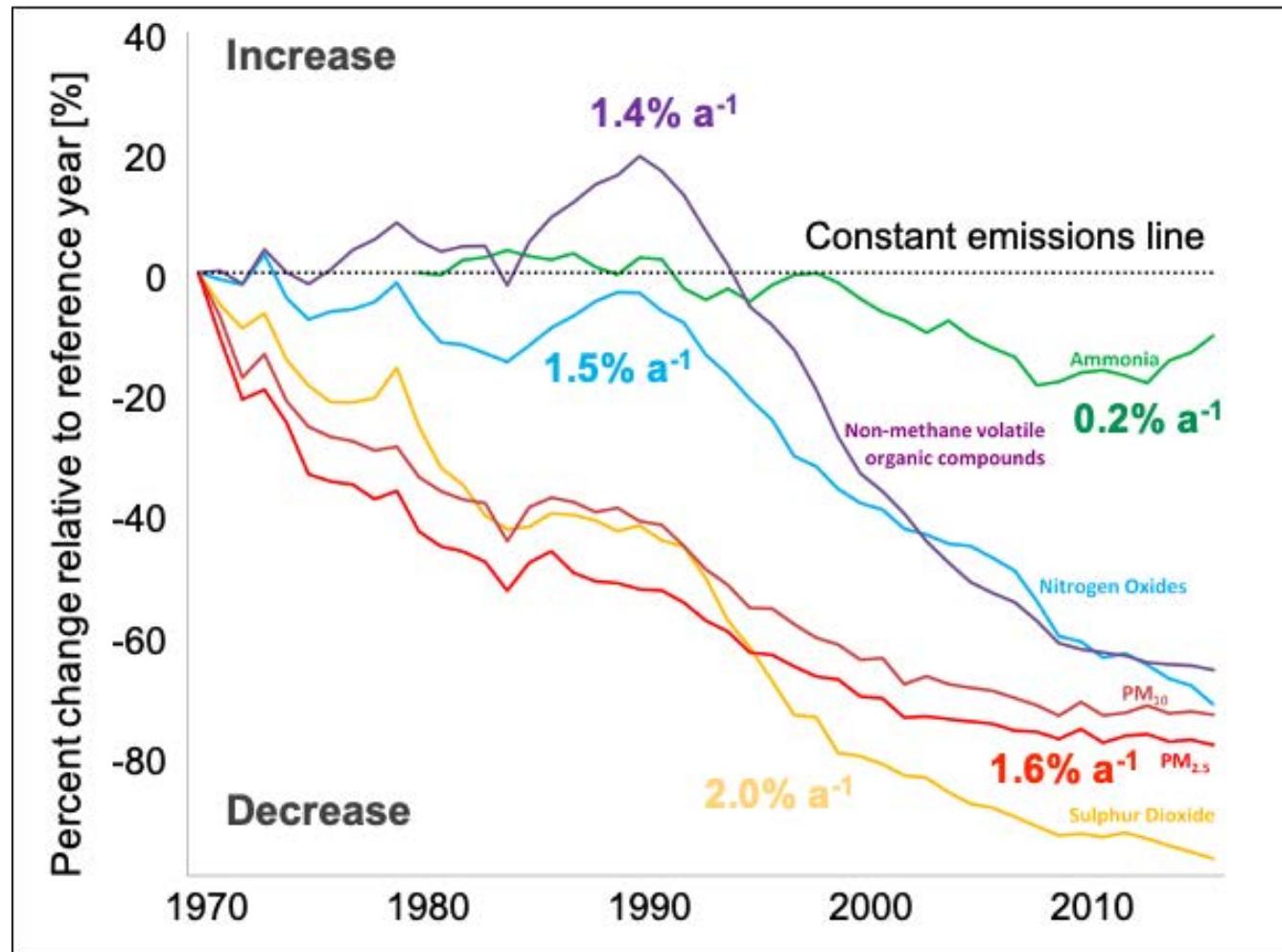
# Addressing uncertainties in UK ammonia emissions



**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

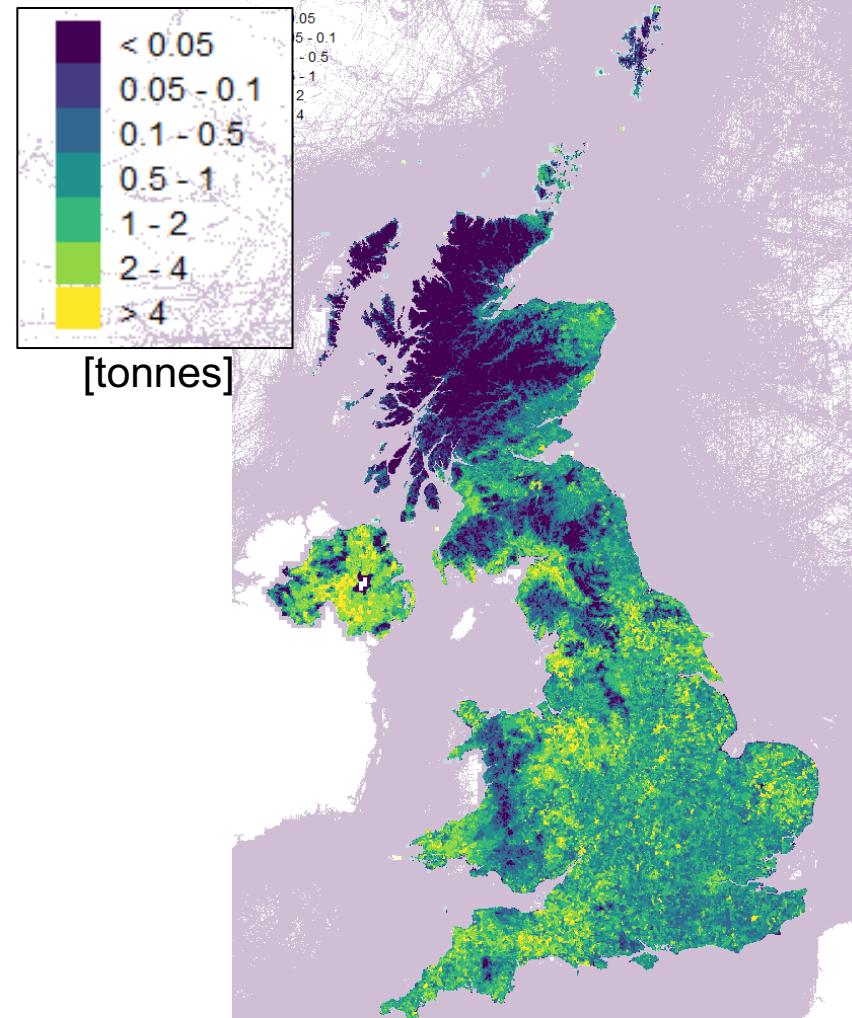


[Adapted from Defra, 2018]

Successful decline in all emissions, except ammonia (NH<sub>3</sub>)

## Spatial Variability in Emissions

### NH<sub>3</sub> emissions for 2018 at 1 km

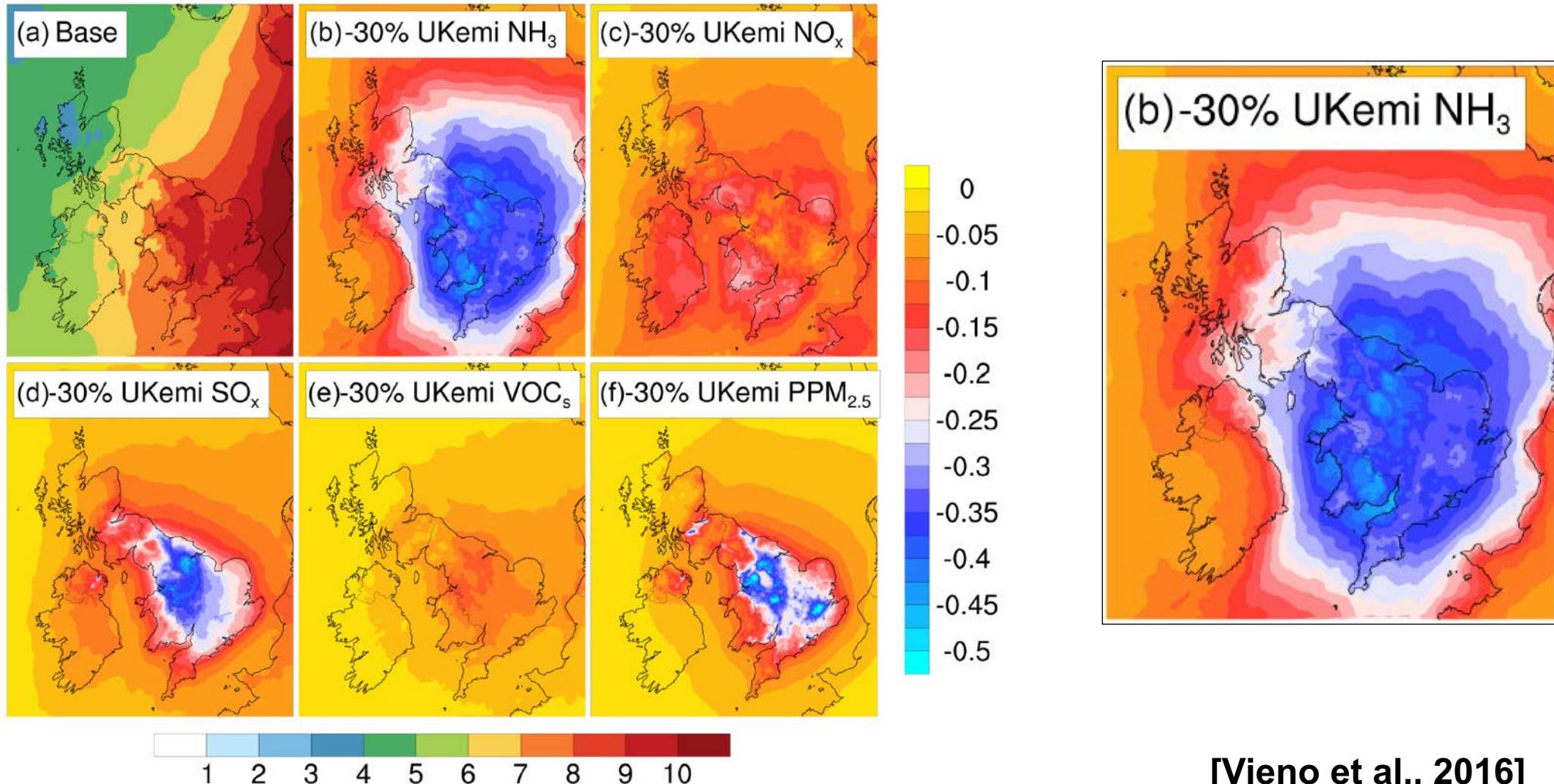


All maps © Crown copyright. All rights reserved Defra, Licence number 100022861 (2020) and BEIS, Licence number 100037028 (2020) LPS © Crown copyright and database right 2020 Licence INSP594

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

# Ammonia impact on air pollutants hazardous to health

## Effect of emission controls on 2010 PM<sub>2.5</sub>



[Vieno et al., 2016]

Largest and most extensive decline in PM<sub>2.5</sub> 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**

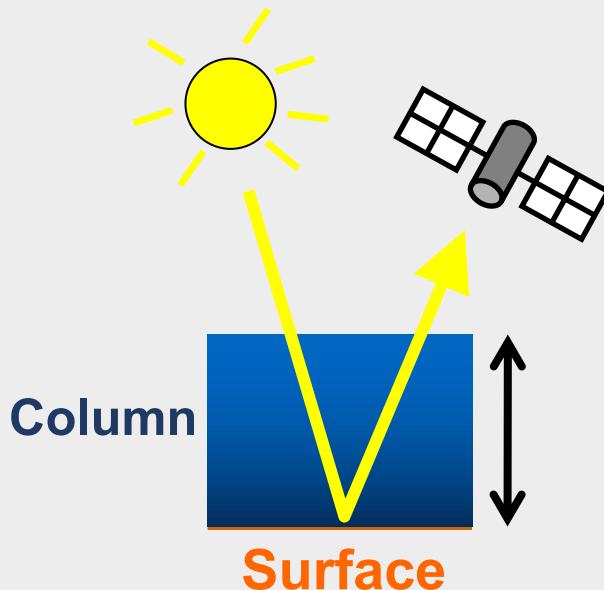


**Conversion Factor**

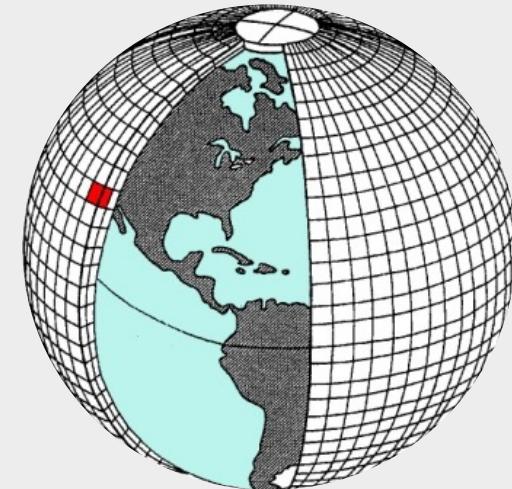


**EMISSIONS**

**Satellite column densities**



**Model Concentration-to-Emission Ratio**



**Satellite-derived Surface Emissions**

**Emission**



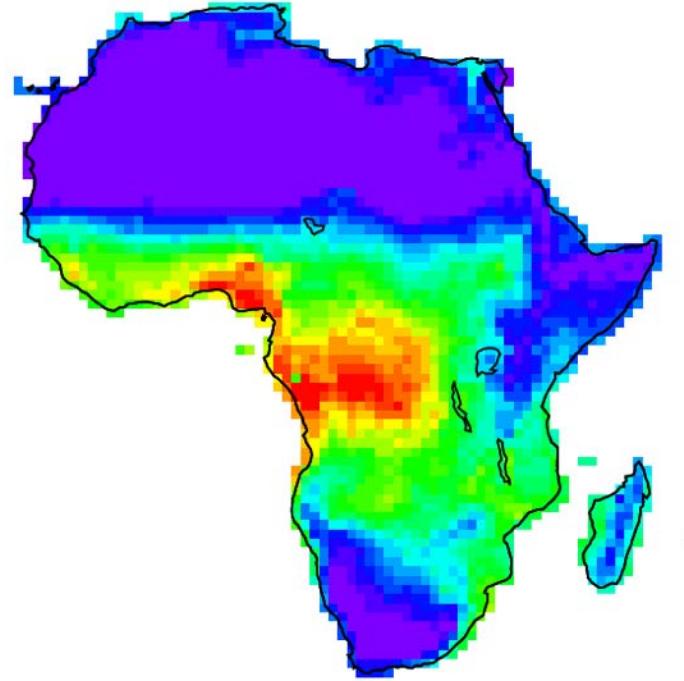
# Widely used to estimate emissions and surface concentrations

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

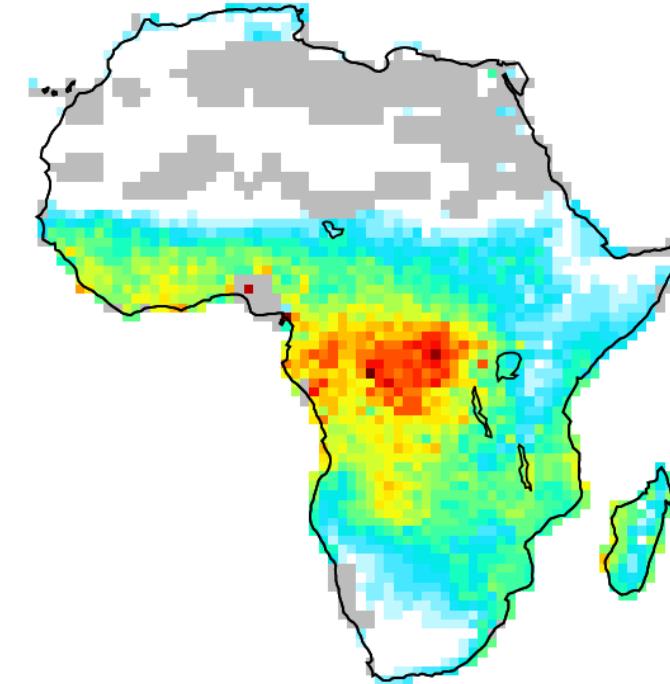


0 2 4 6 8  
[ $10^{15}$  molecules  $\text{cm}^{-2}$ ]



Model effective yields

Isoprene emissions

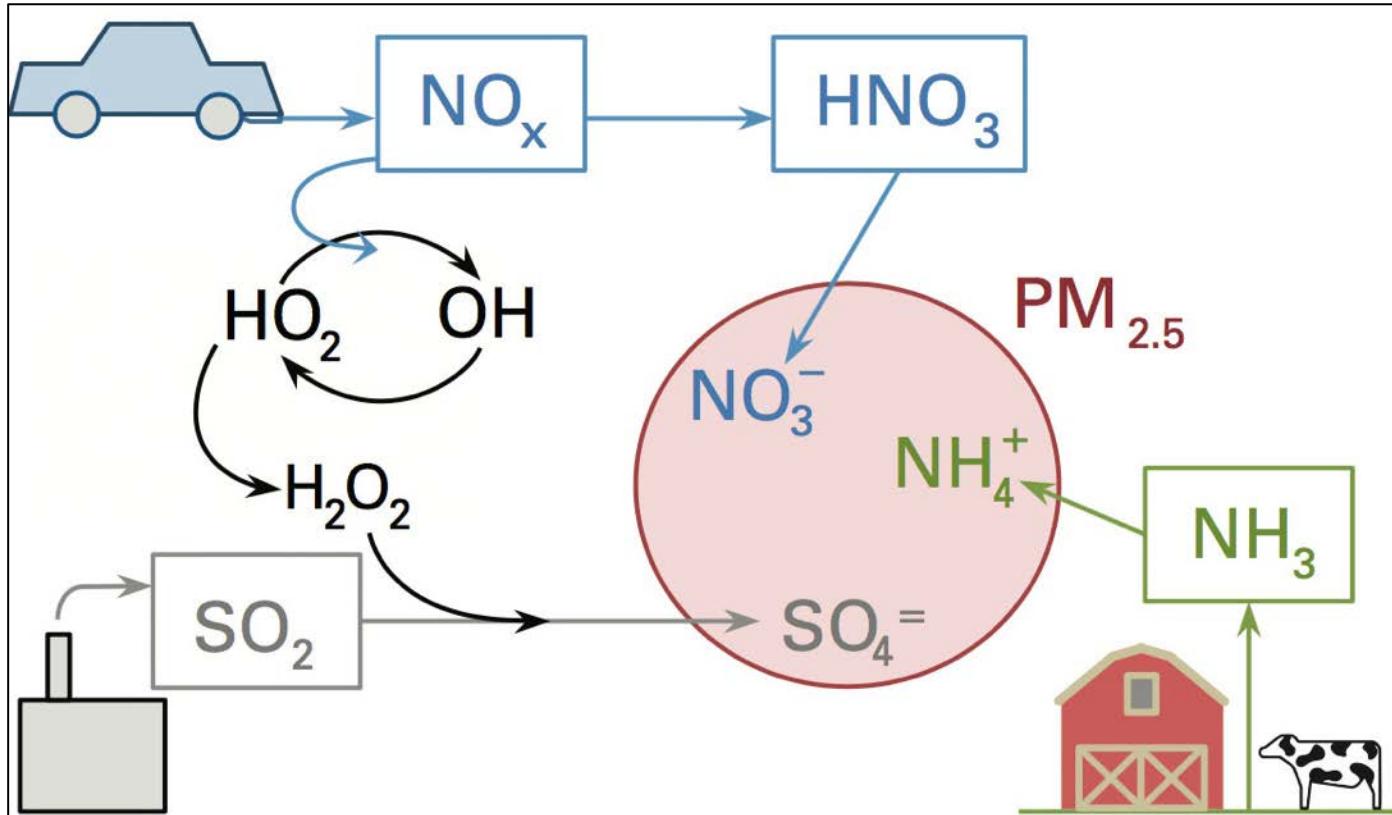


0 1 2 3 4 5 6  
[ $10^{12}$  atoms C  $\text{cm}^{-2} \text{s}^{-1}$ ]

[Marais et al., ACP, 2012]

# Ammonia abundance depends on numerous factors

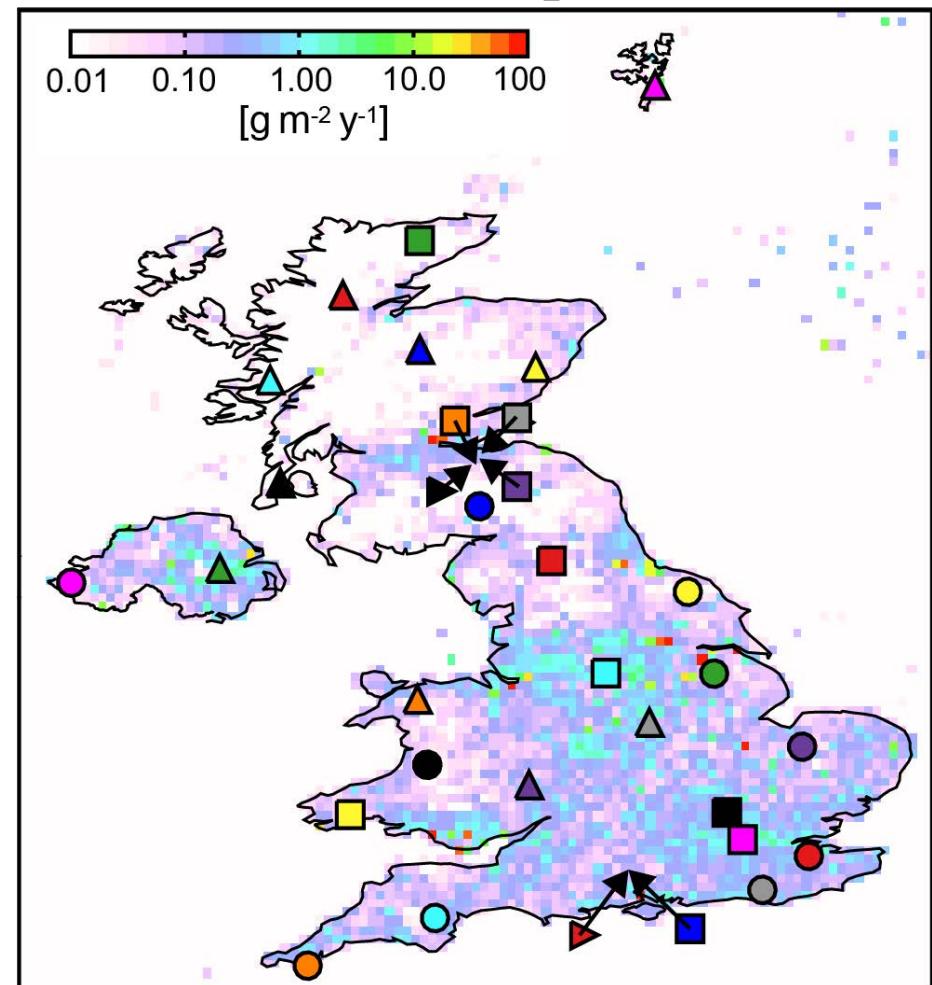
Ammonia partitions to aerosols to form PM<sub>2.5</sub>



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

Partitioning of ammonia (NH<sub>3</sub>) to pre-existing aerosols depends on abundance of NO<sub>x</sub> and SO<sub>2</sub>

NAEI Annual SO<sub>2</sub> Emissions



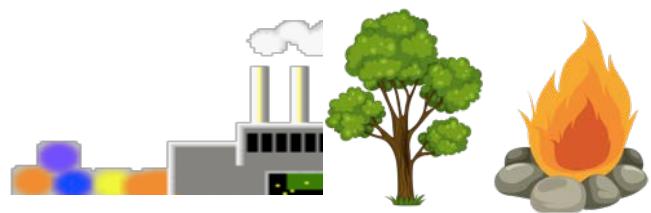
**Symbols:** SO<sub>2</sub> concentration monitors

# Surface SO<sub>2</sub> concentrations calculated with GEOS-Chem



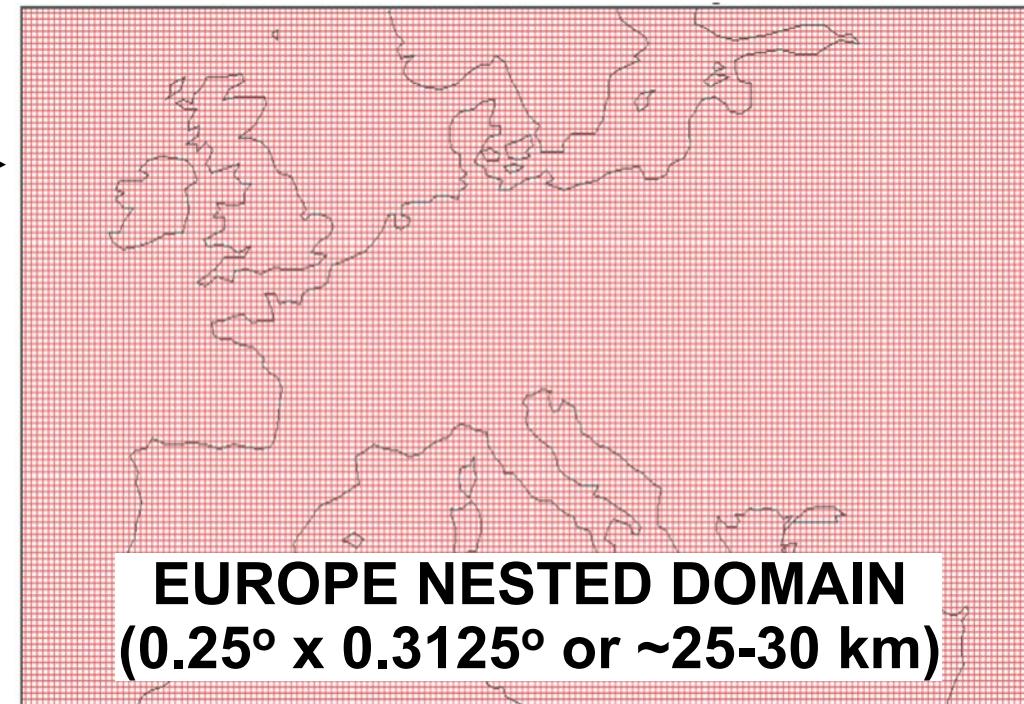
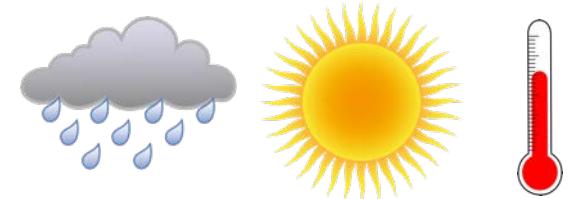
3D Atmospheric Chemistry Transport Model

Emissions  
(natural/human)



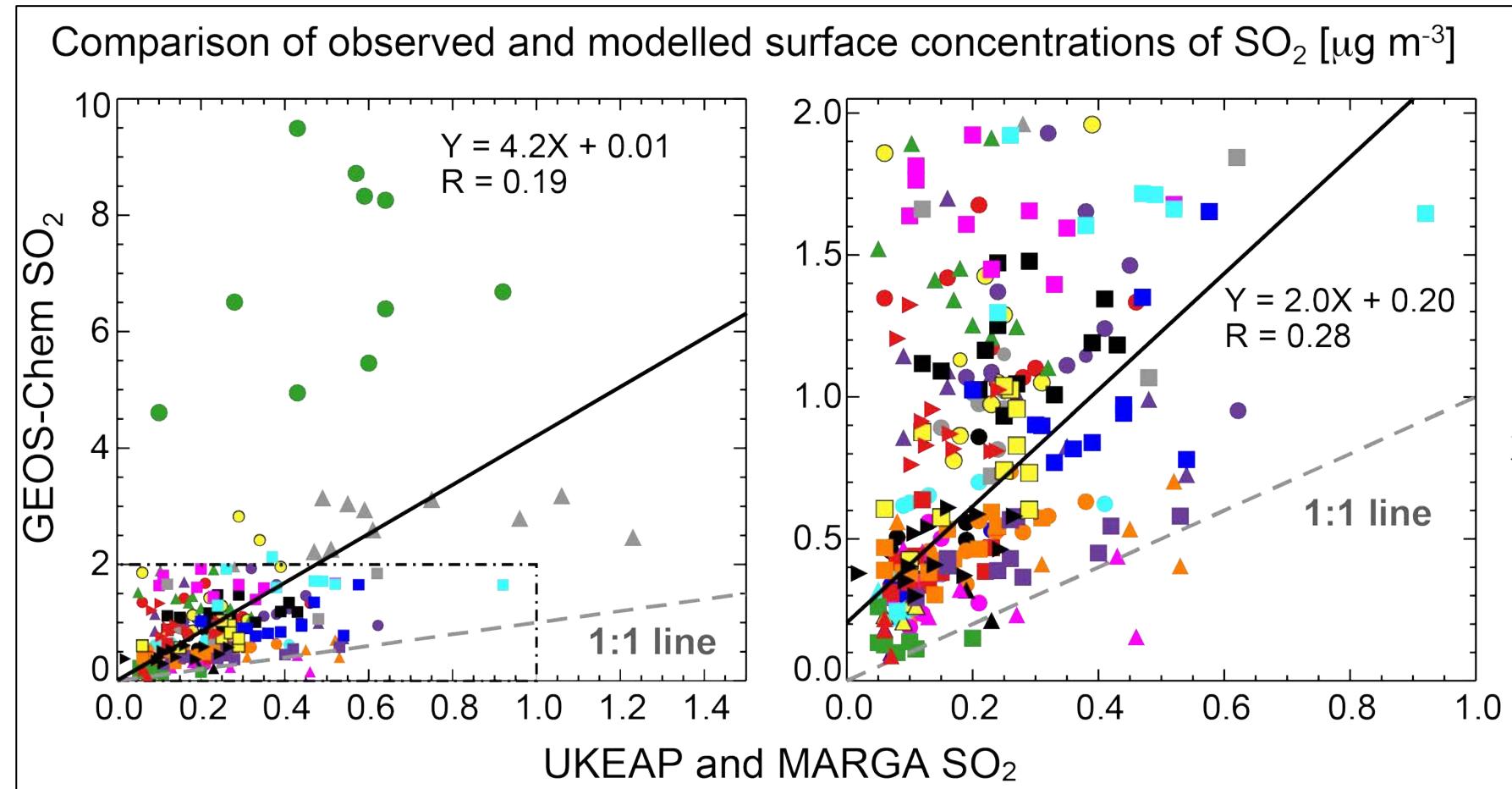
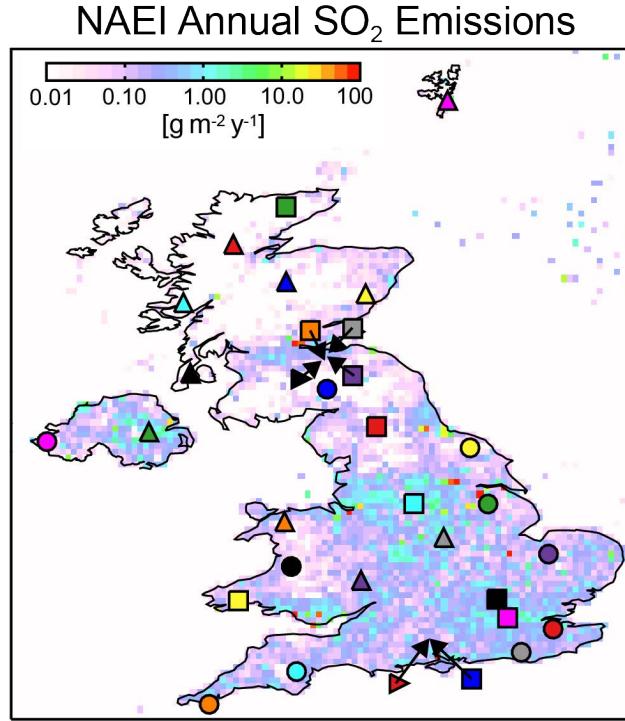
UK NAEI emissions  
(with temporal information)

Offline assimilated  
meteorology



Gas phase and heterogeneous chemistry  
Transport  
Dry/wet deposition

# GEOS-Chem-NAEI (model) versus observed SO<sub>2</sub>



**UKEAP:** offline denuder measurements (0.05 μg m<sup>-3</sup> detection limit)

**MARGA:** semi-continuous denuder measurements (0.04 μg m<sup>-3</sup> detection limit)

Comparison supports large overestimate in NAEI SO<sub>2</sub> emissions (in particular point sources)

Decrease annual SO<sub>2</sub> emissions 161 Gg to 87 Gg

# Infrared Atmospheric Sounding Interferometer (IASI) Instrument

Overpass:

9:30 local solar time

Spatial resolution:

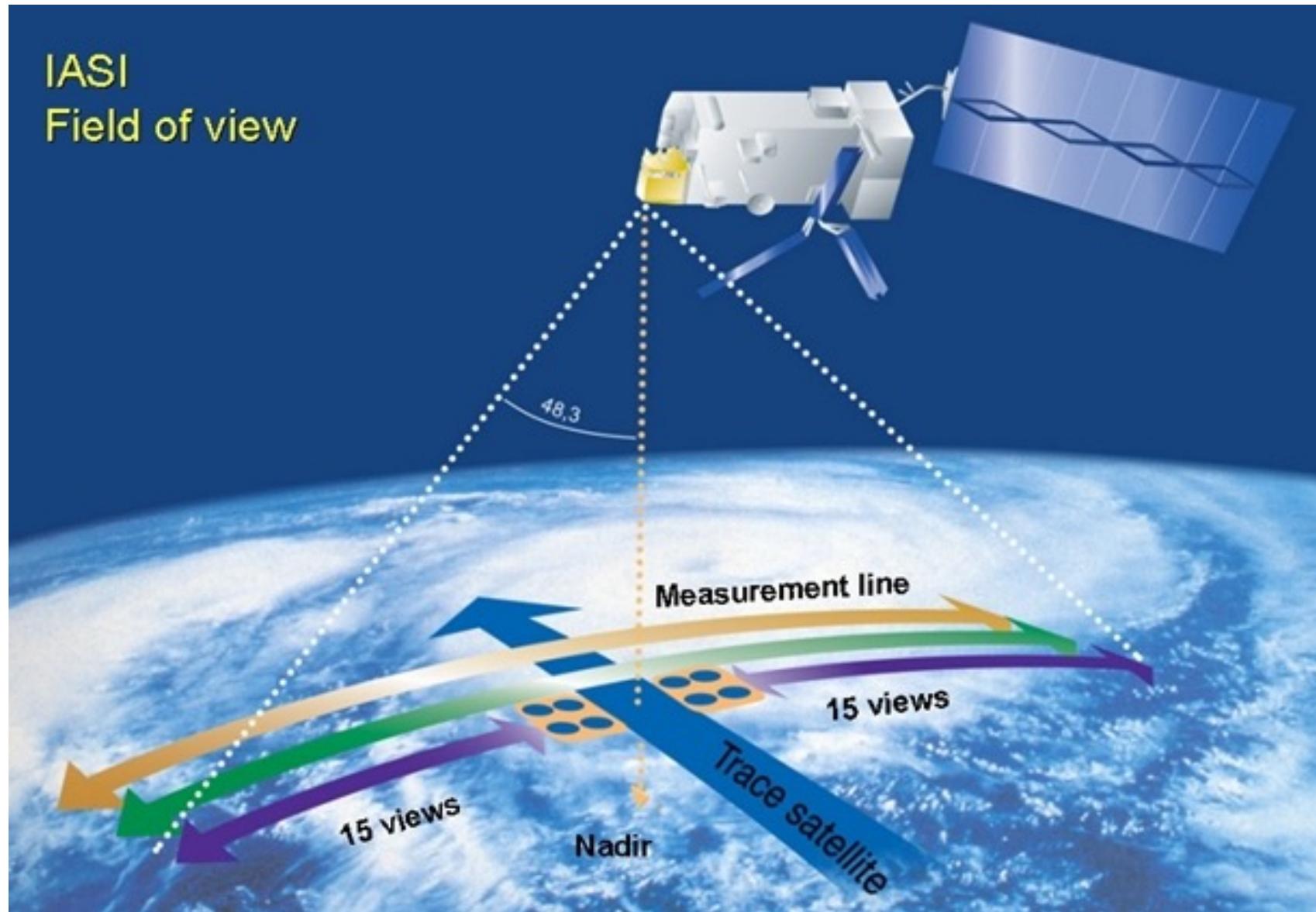
12 km to 39 km

Swath width:

2200 km

Launch date:

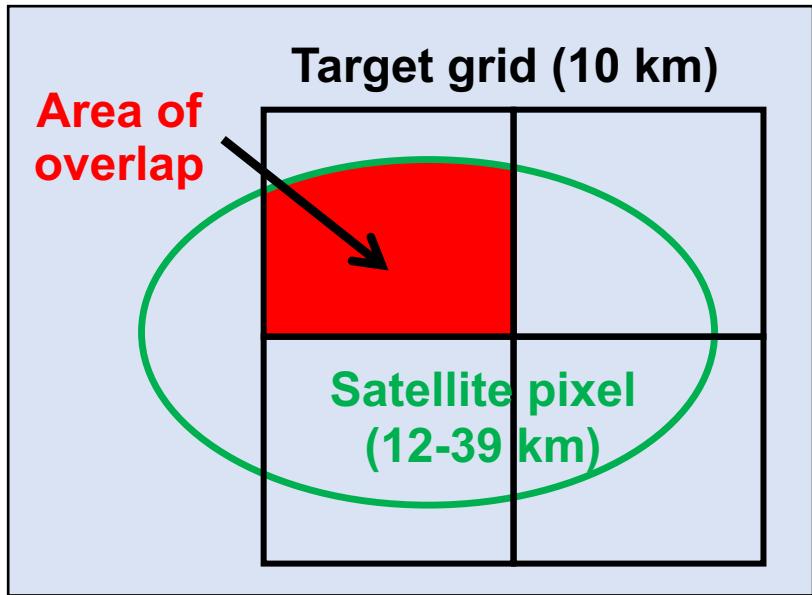
October 2006



# Ammonia emissions in the UK: the top-down perspective

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

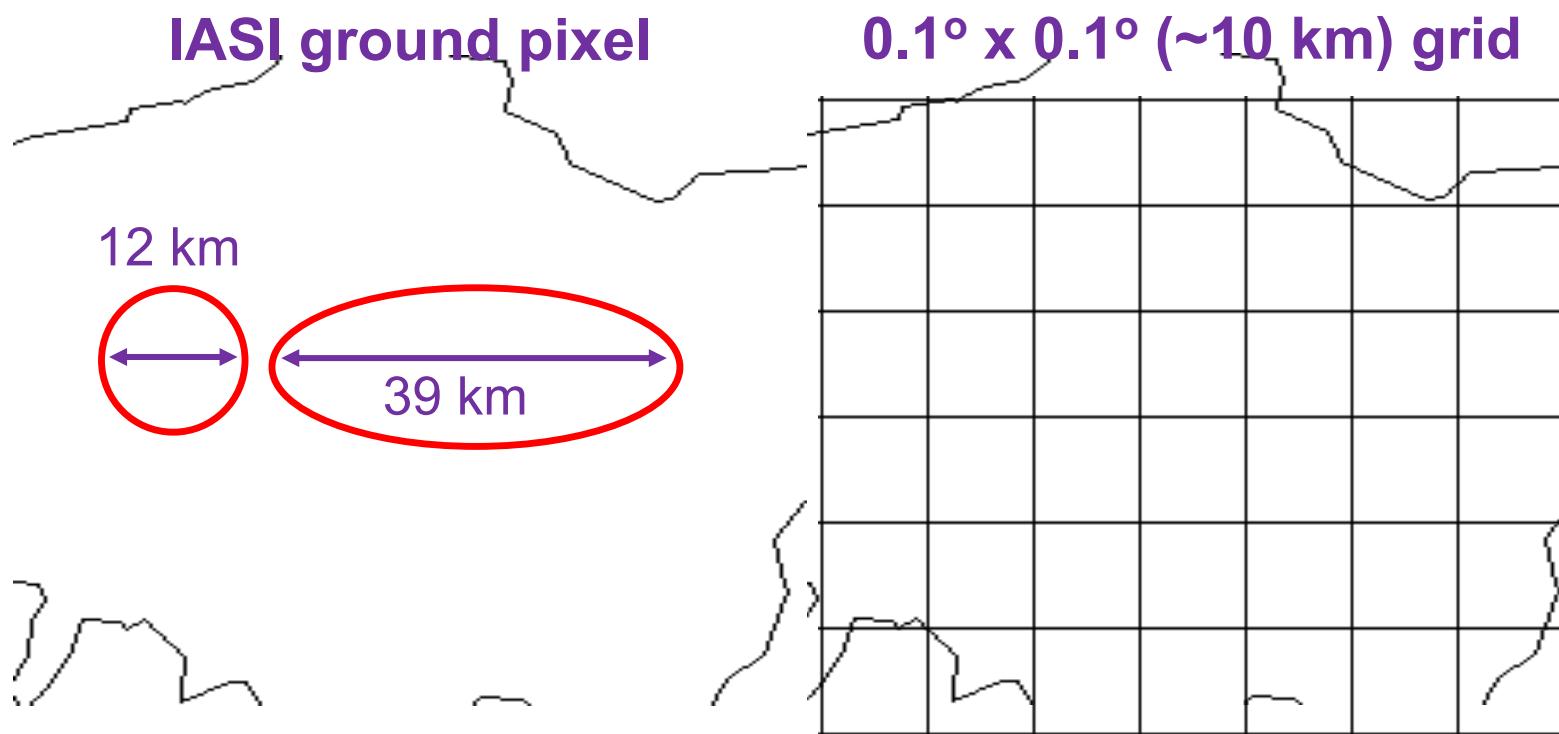
## Oversampling Technique



Weights each IASI NH<sub>3</sub> pixel by area of overlap and the reported uncertainty

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

## Oversampling technique over London

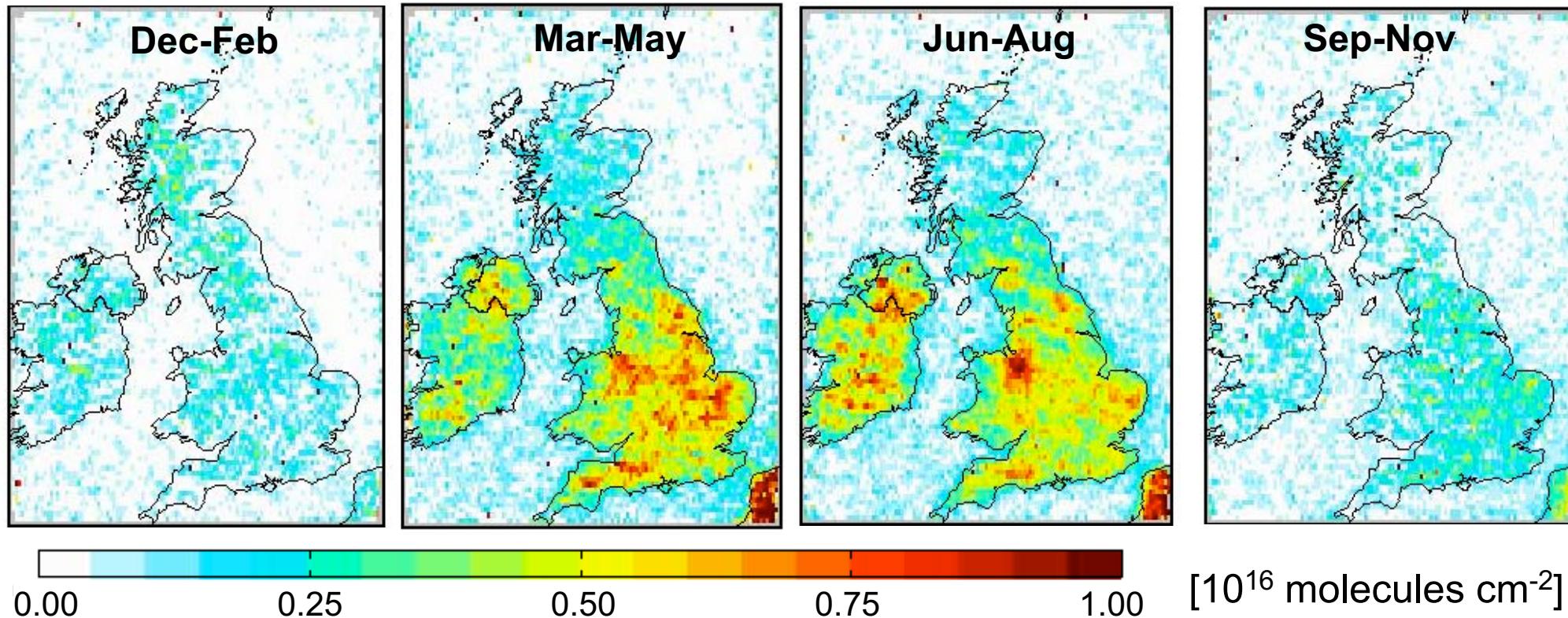


Lose time (temporal) resolution; gain spatial resolution

# 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 NH<sub>3</sub> 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

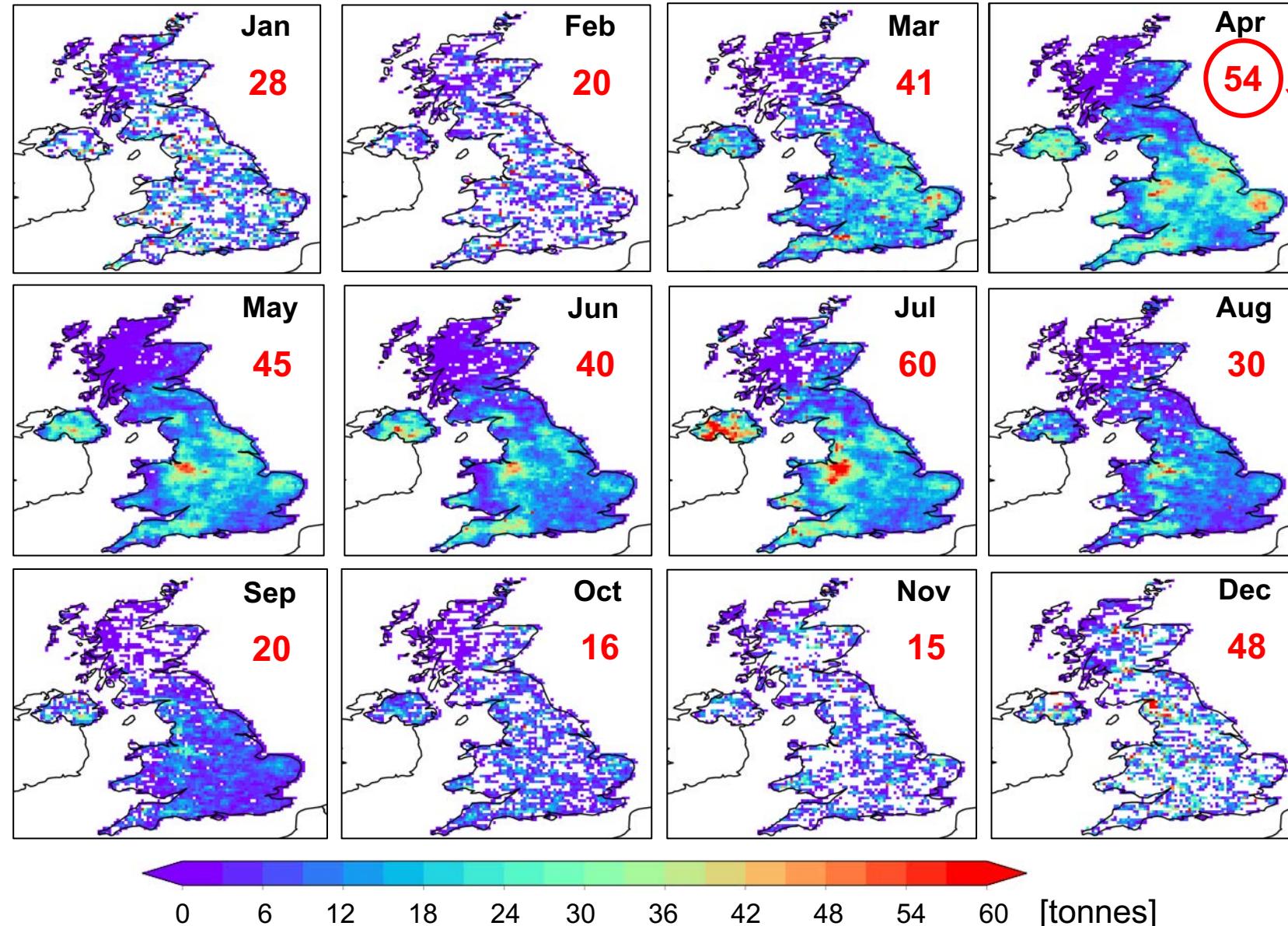
**IASI data providers:** M. Van Damme, L. Clarisse,  
P.-F. Coheur, ULB, Belgium

# UK IASI-derived ammonia emissions

Convert IASI NH<sub>3</sub> column concentrations to surface emissions of NH<sub>3</sub>

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

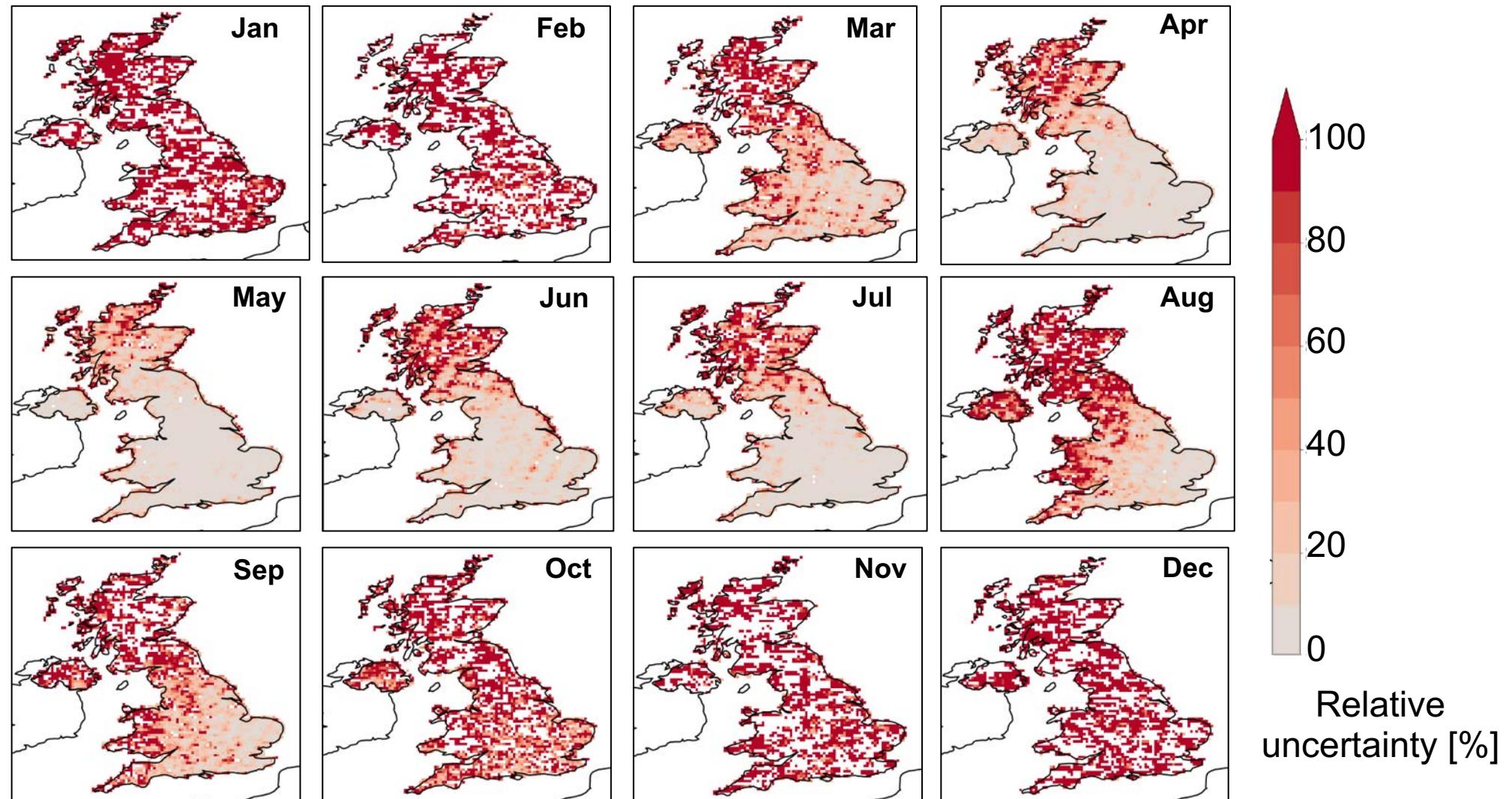
Challenging to retrieve NH<sub>3</sub> in these months



Total monthly emissions (Gg)

# Account for Observation Uncertainties

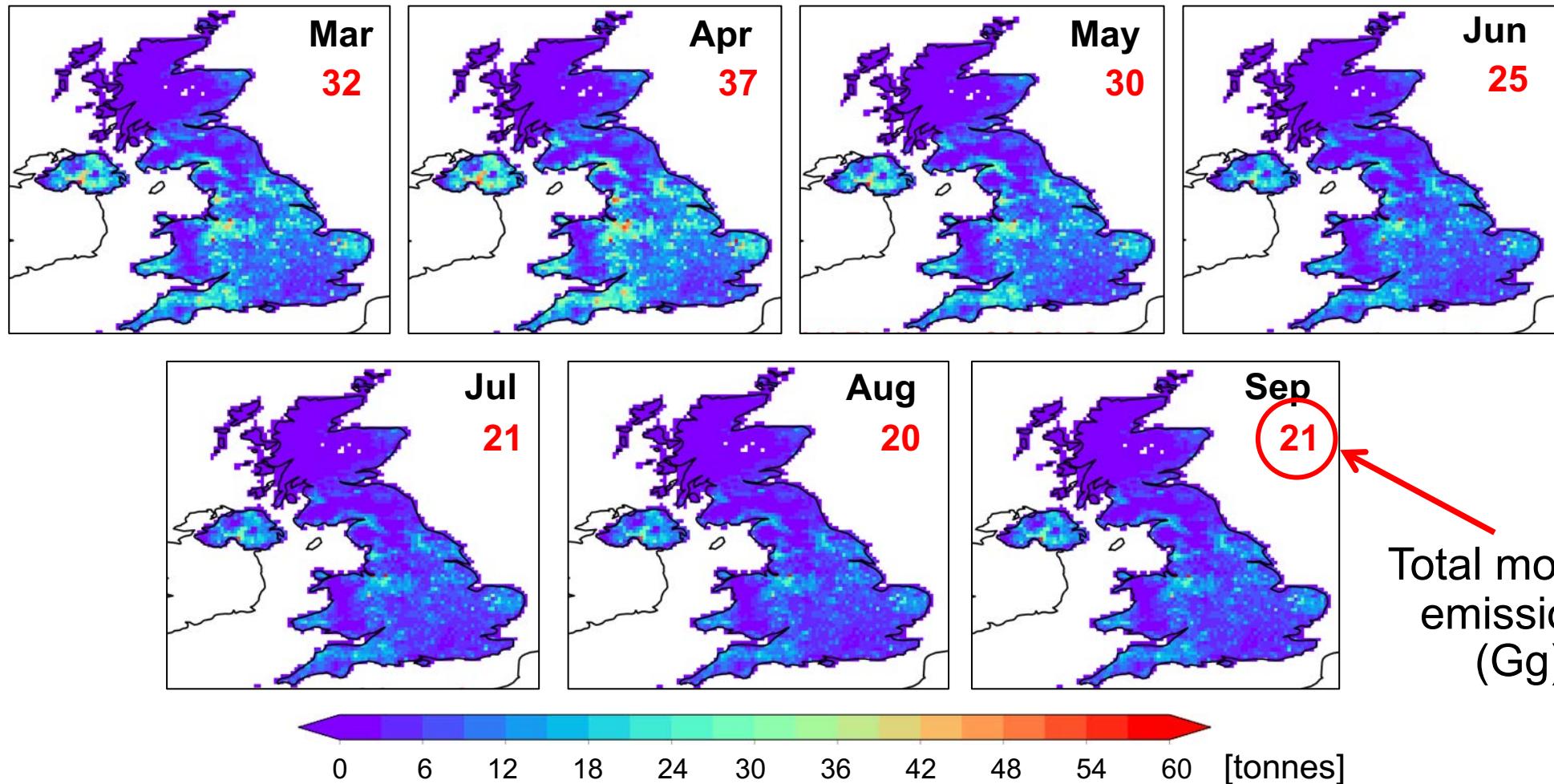
IASI NH<sub>3</sub> column concentrations susceptible to large uncertainties in cold months



Only consider months with relatively low uncertainty: March-September

# NAEI ammonia emissions with assumed seasonality

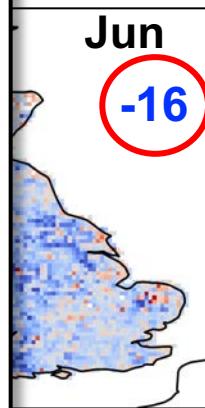
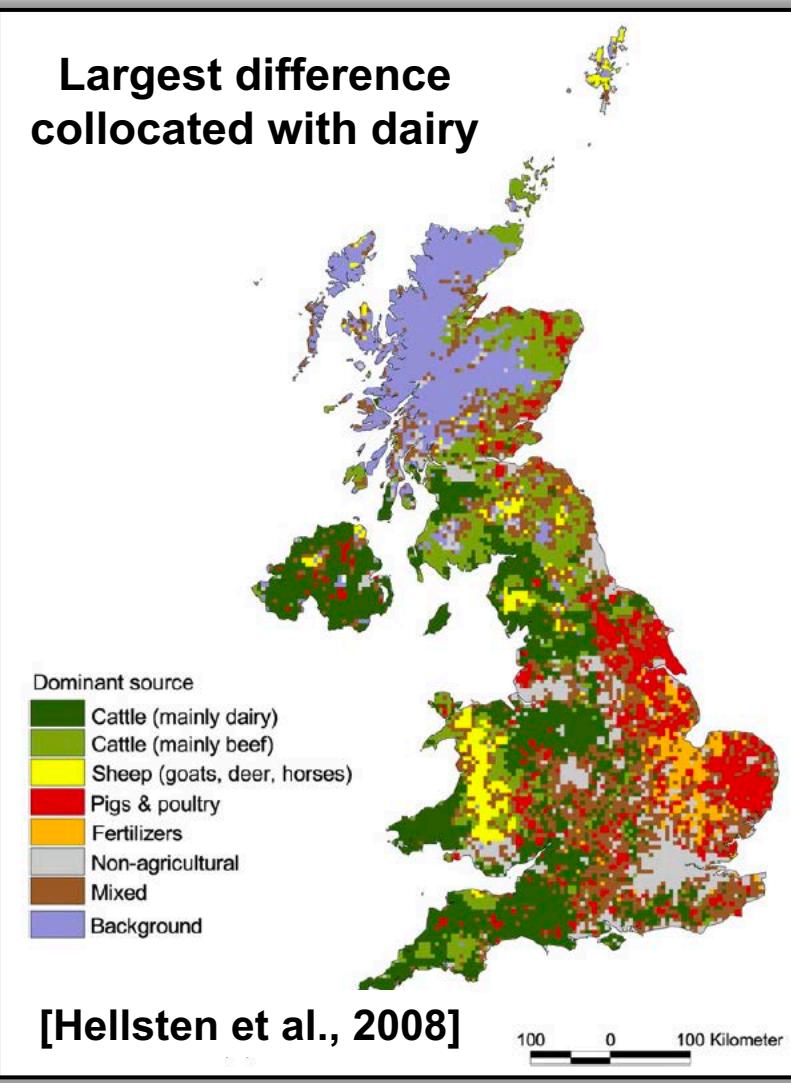
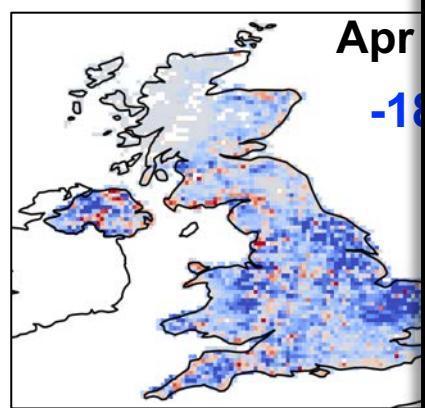
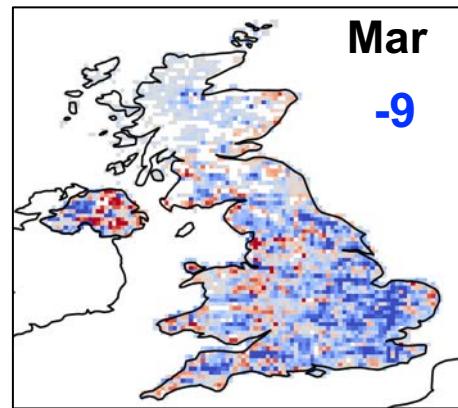
NAEI NH<sub>3</sub> emissions with monthly scaling factors used in GEOS-Chem applied



NAEI NH<sub>3</sub> emissions in March-September are 67% of annual NAEI NH<sub>3</sub> emissions

# Assessment of the UK National Emission Inventory

Compare IASI-derived and NAEI  $\text{NH}_3$  emissions with representative scaling factors applied to the NAEI

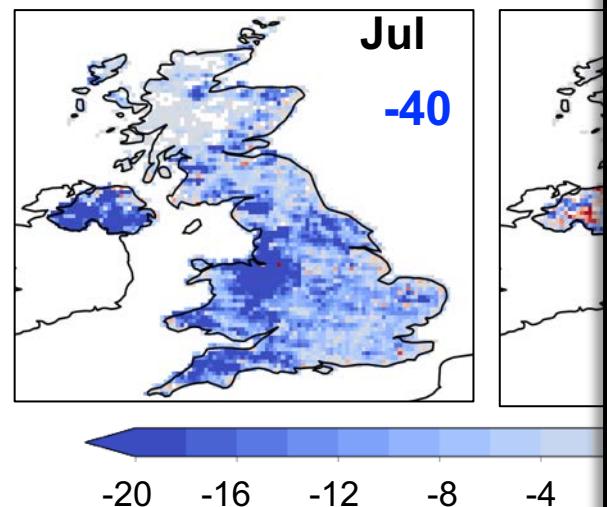


Difference in monthly emissions (Gg)

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

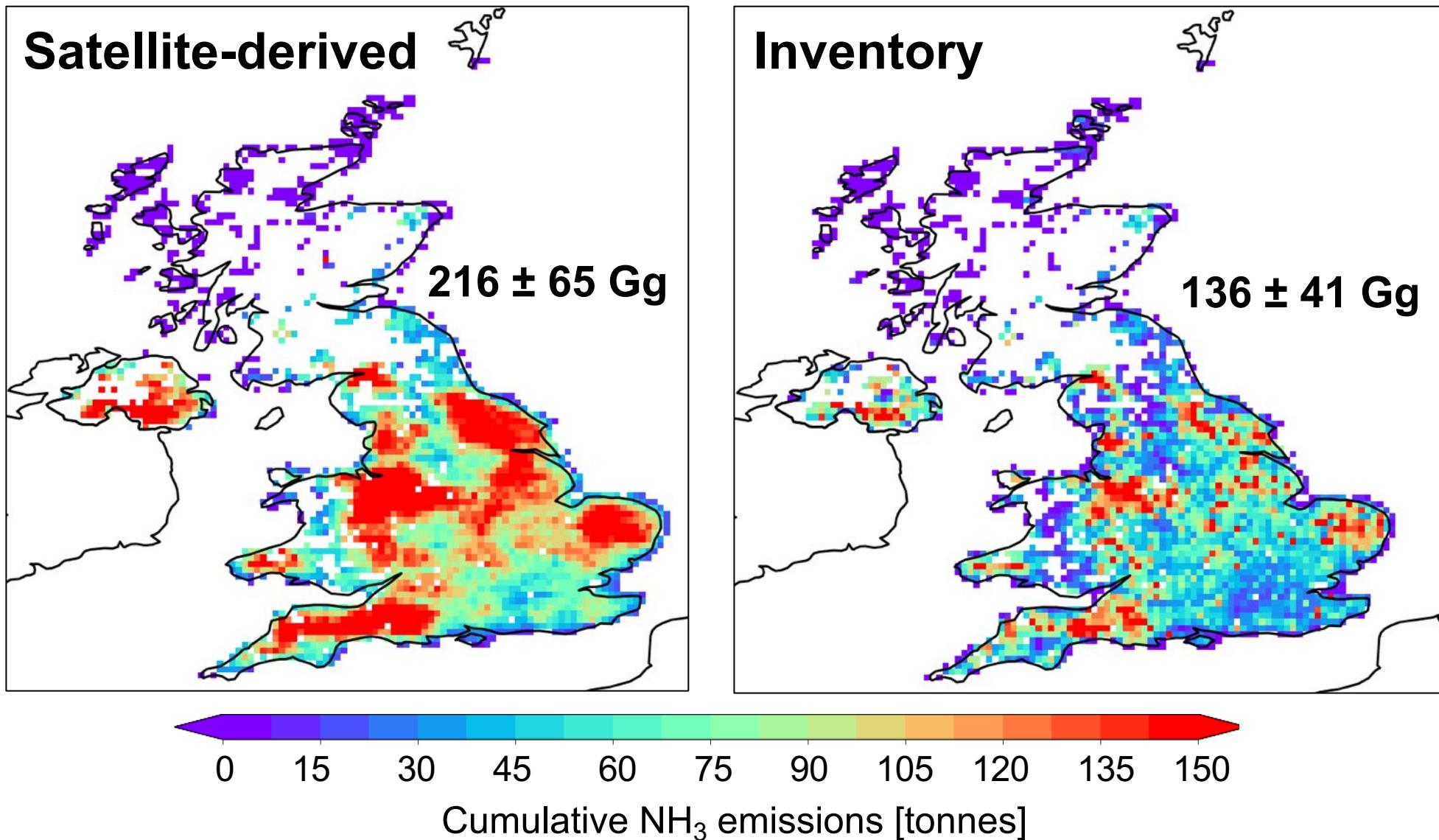
Largest discrepancy over locations dominated by **dairy farms**

# Comparison of total March-September emissions

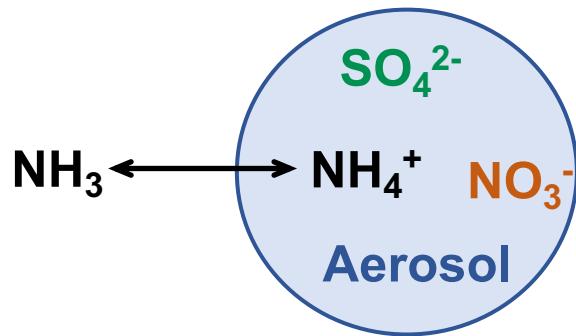
Comparison suggests satellite-derived estimate suggests NAEI underestimates emissions.

Similar relative errors

Satellite-derived emissions 60% more than inventory emissions



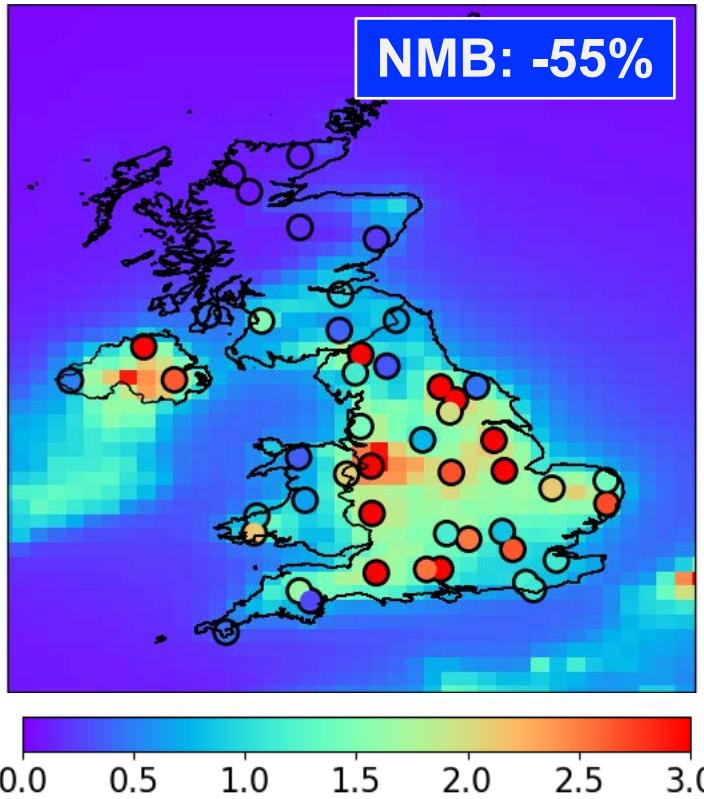
# But, the results we obtain may be erroneous ...



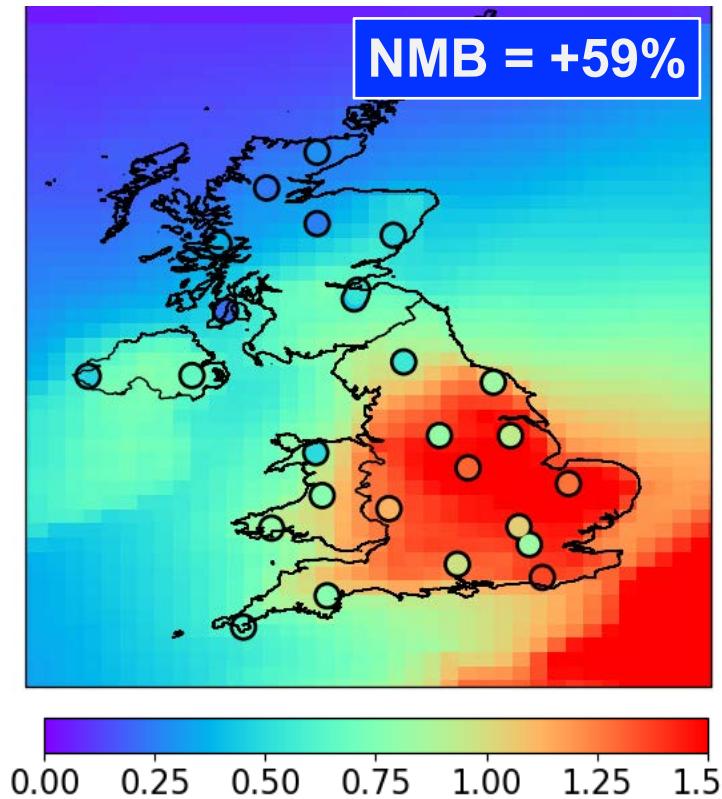
Gas-phase abundance of  $\text{NH}_3$  depends on sulfate and nitrate

Does the model get this balance right?

Annual mean  $\text{NH}_3$  [ $\mu\text{g m}^{-3}$ ]



Annual mean  $\text{NH}_4^+$  [ $\mu\text{g m}^{-3}$ ]

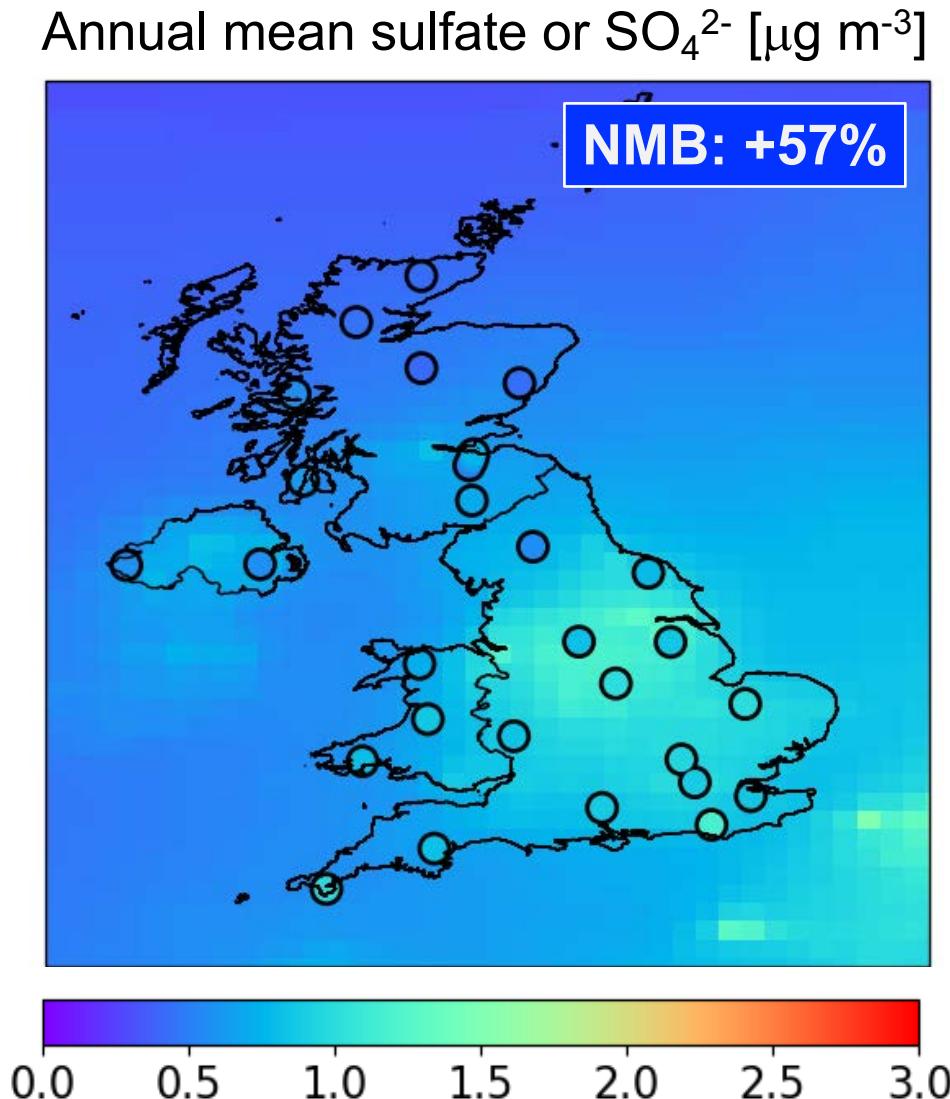
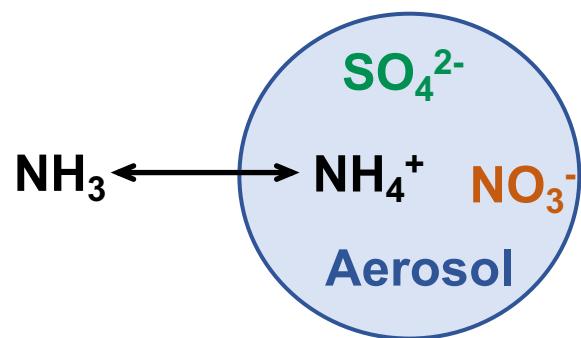


**NMB:** Model  
normalized  
mean bias

Model underestimates  
 $\text{NH}_3$ , but overestimates  
ammonium ( $\text{NH}_4^+$ )

# Model overestimates sulfate, despite decreasing SO<sub>2</sub> emissions

Suggests that reduction in NAEI SO<sub>2</sub> emissions in the model is too conservative



NMB: Model  
mean bias

Also emphasizes importance  
of sources and sinks in  
deriving emissions from  
satellite observations of NH<sub>3</sub>

# Concluding Remarks

- Developed fine spatial and temporal resolution products of NO<sub>2</sub> in the upper troposphere (UT) using TROPOMI
- TROPOMI UT NO<sub>2</sub> reproduces spatial variability of peer-reviewed OMI product
- Potential to address data sparsity in a profoundly important portion of the atmosphere
- UK NAEI overestimates SO<sub>2</sub> emissions by at least a factor of 2
- Satellite-derived NH<sub>3</sub> 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 NH<sub>3</sub> to aerosols due to remaining positive biases in NAEI SO<sub>2</sub> emissions