

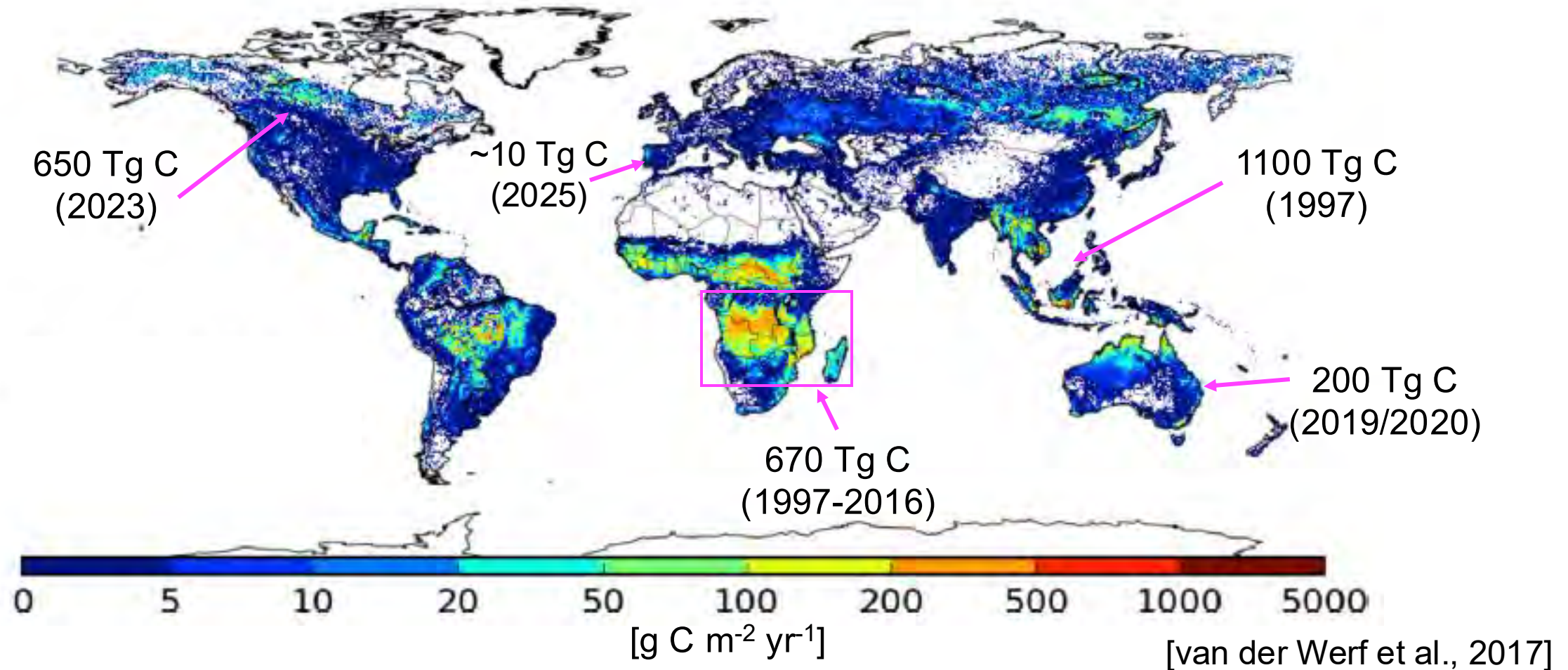
Using satellite observations to identify and address uncertainties in sources of reactive nitrogen



Eloise Marais (UCL) with Martin Van Damme, Lieven Clarisse, Christine Wiedinmyer, Killian Murphy, Guido van der Werf, Nana Wei, Gongda Lu, Mogesh Naidoo, Rebecca Garland, Keita Sekou

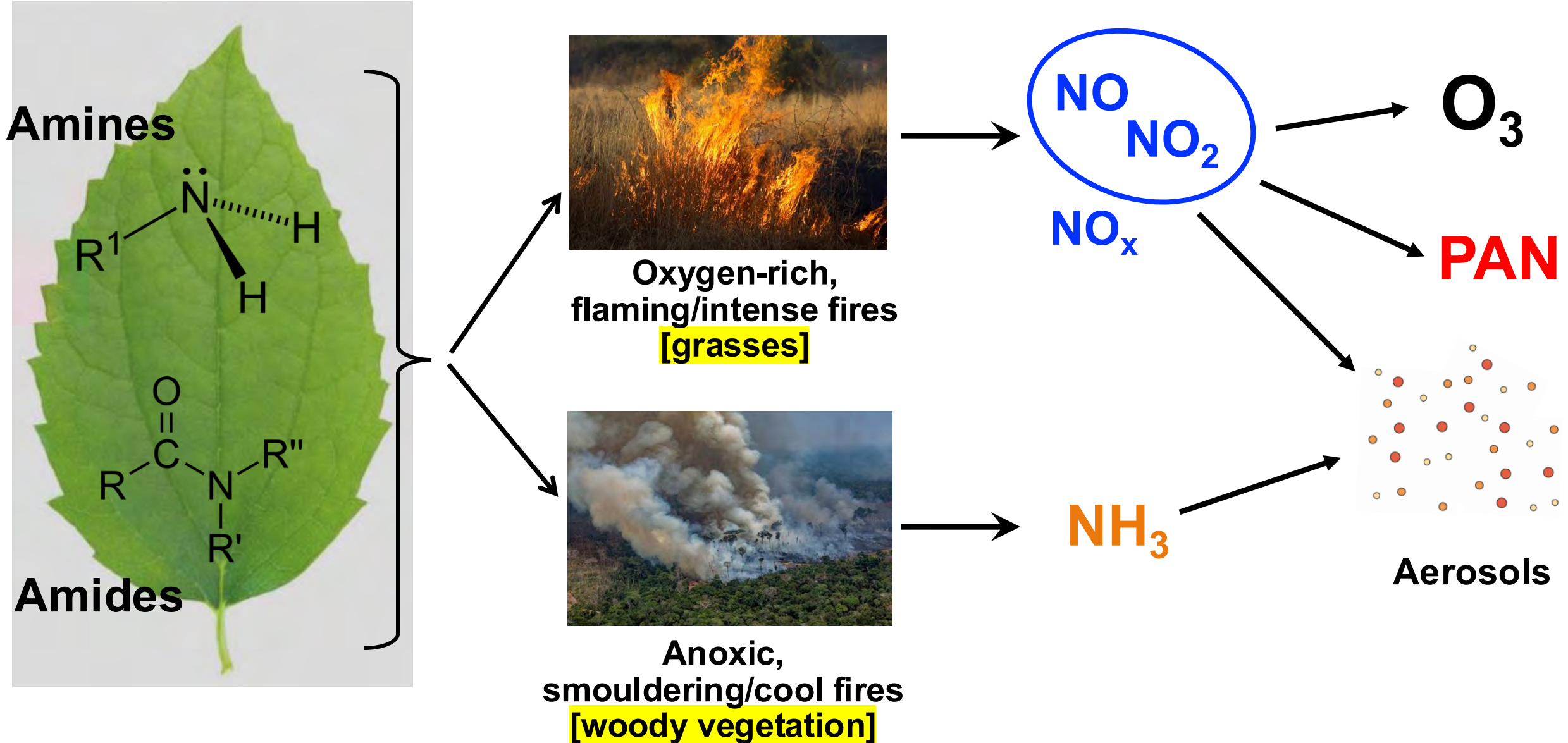
Global Context for Southern Africa Biomass Burning

Biomass burning carbon emissions from the Global Fire Emissions Database (GFED) inventory



Open fire emissions in southern Africa outcompete most anomalous fires in other regions, so is a potentially large global source of reactive nitrogen

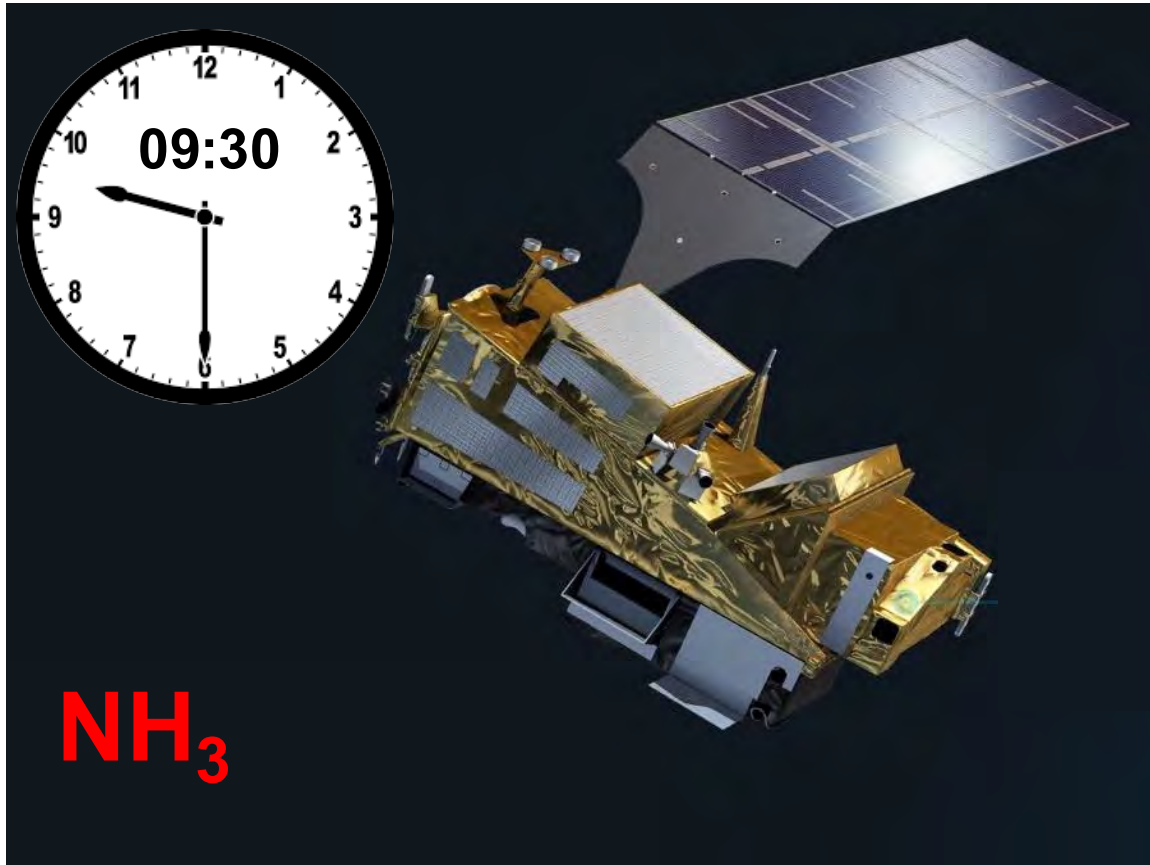
Open Fire Emissions of Reactive Nitrogen



NO_x and NH_3 affect local air quality, regional climate, and global atmospheric composition

Instruments in space measuring NH_3 and NO_2 column densities

IASI: Infrared Atmospheric Sounding Interferometer



Resolution: 12 km (elliptical pixels) at nadir

Swath width: 2200 km

Launch date: 2012

Year used: 2019

TROPOMI: TROPospheric Monitoring Instrument



Resolution: 5.5 km x 3.5 km at nadir

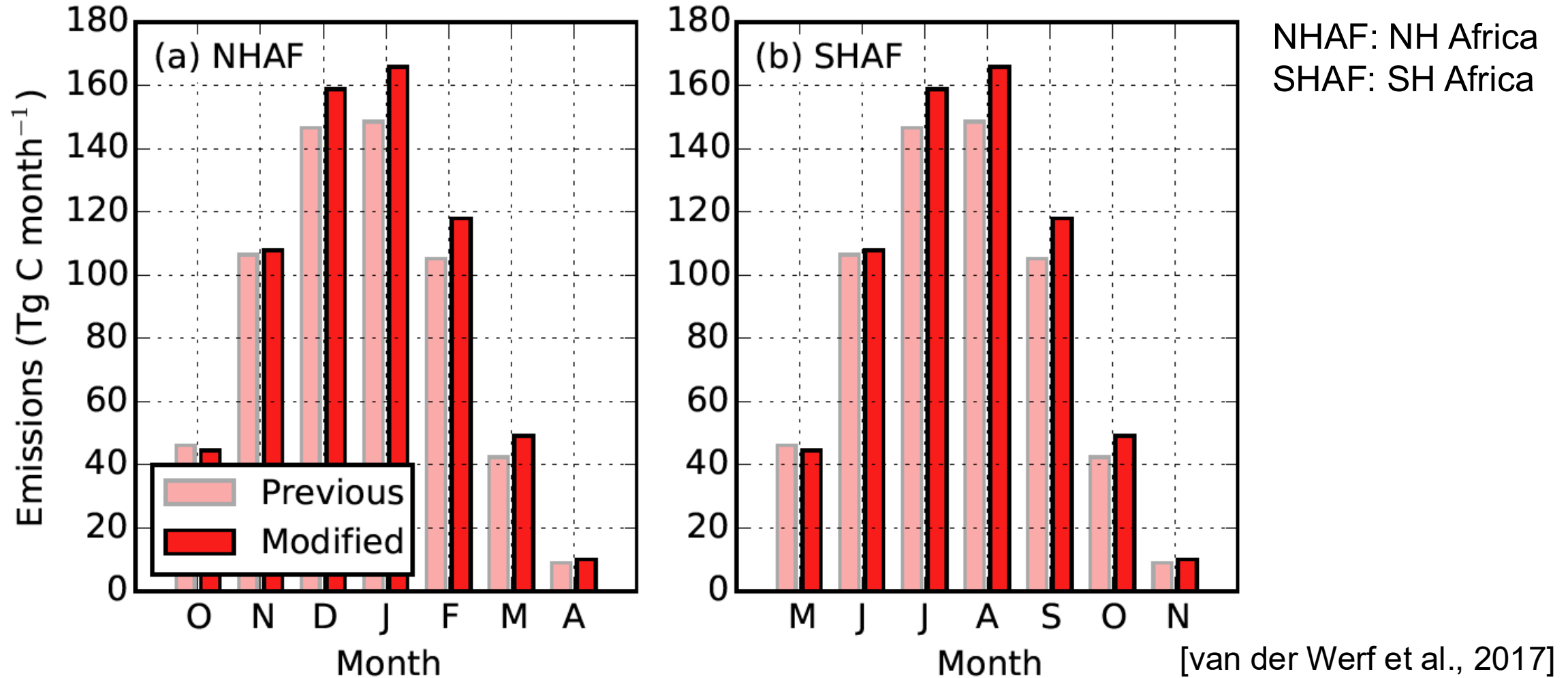
Swath width: 2600 km

Launch date: 2017

Year used: 2019

Seasonality in Southern Africa Biomass Burning

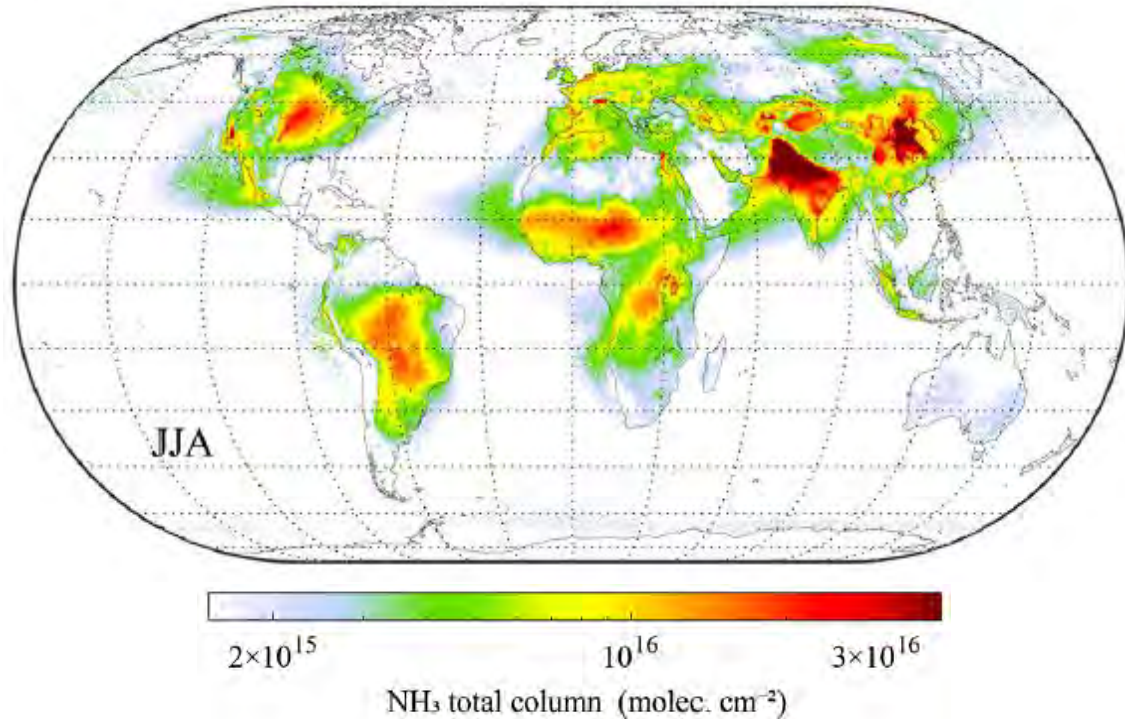
Multiyear mean monthly carbon emissions from biomass burning [Tg C]



Southern Africa fire season persists for all 6 months of the dry season, peaking in July and August

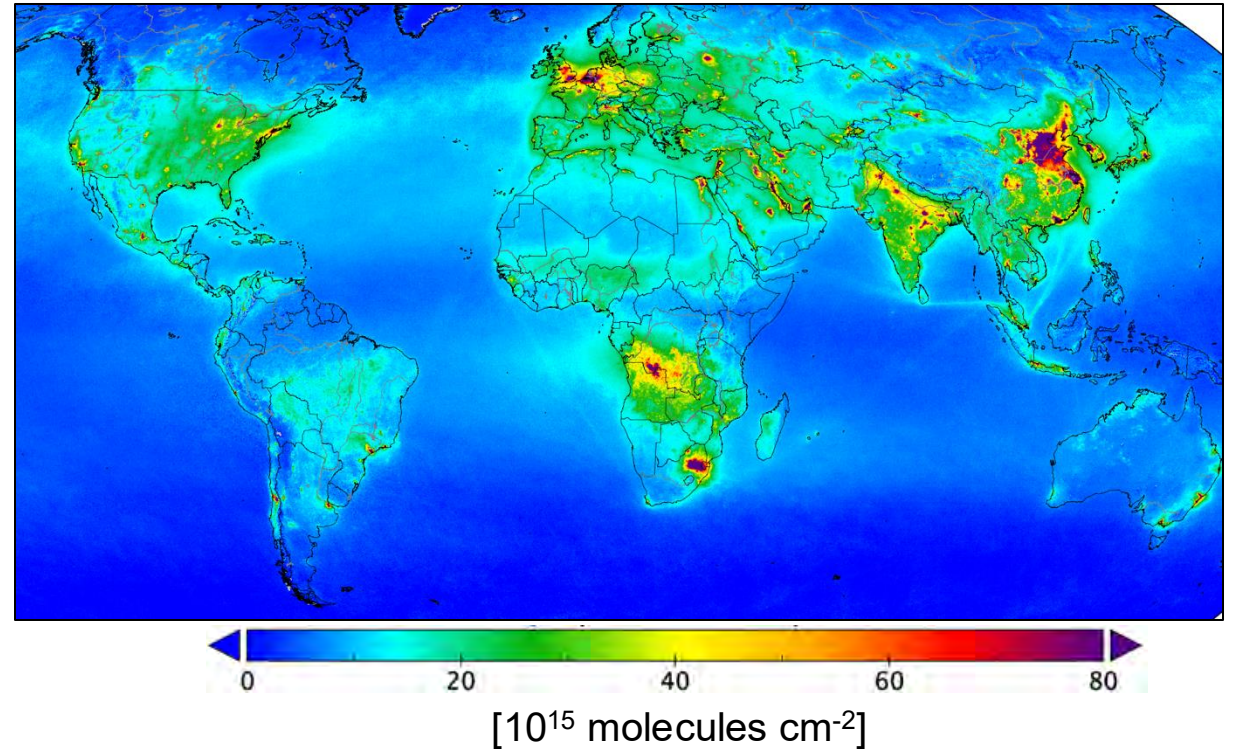
Fires Detected with Both Instruments

IASI multiyear seasonal mean NH_3



[Clarisse et al., 2023]

TROPOMI annual mean NO_2 (2018-2020)



[https://www.esa.int/Applications/Observing_the_Earth/Copernicus/Sentinel-5P/Nitrogen_dioxide_pollution_mapped]

Instruments provide constraints on NH_3 for IASI and NO_x for TROPOMI NO_2

NO_x and NH_3 account for most reactive nitrogen from open fires

Bottom-Up Biomass Burning Emissions

$$\text{Emission} = \text{DMB} \times \text{EF}$$

DMB: dry matter burned

EF: emission factor

DMB = Area burned x above-ground biomass x combustion completeness

3 prominent inventories:

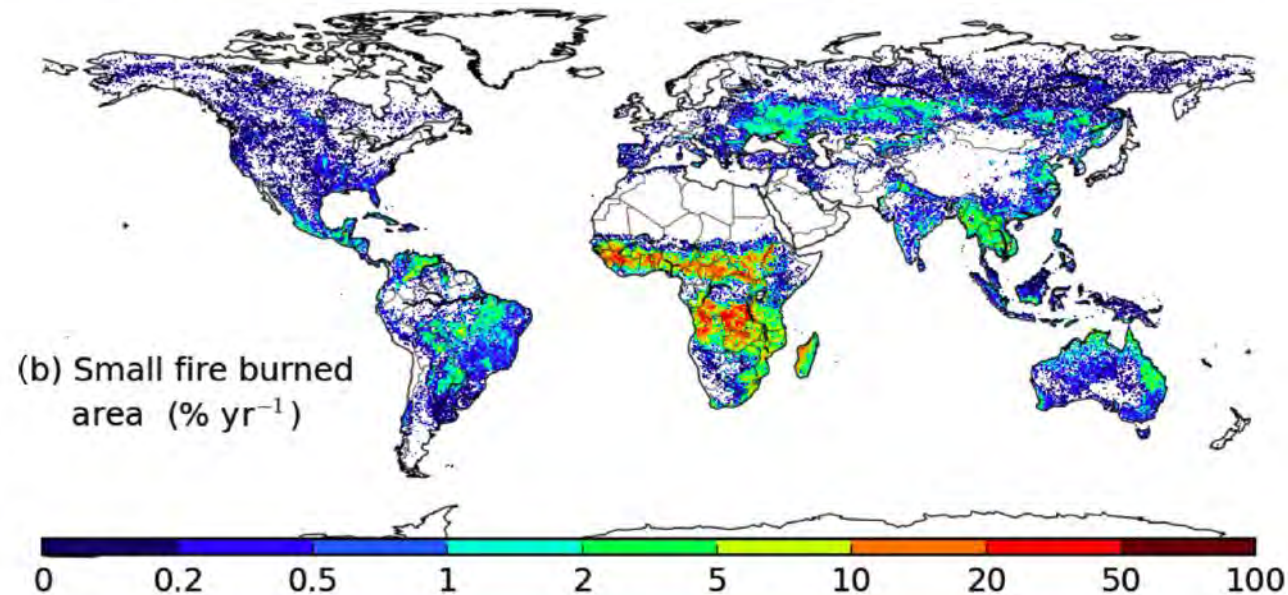
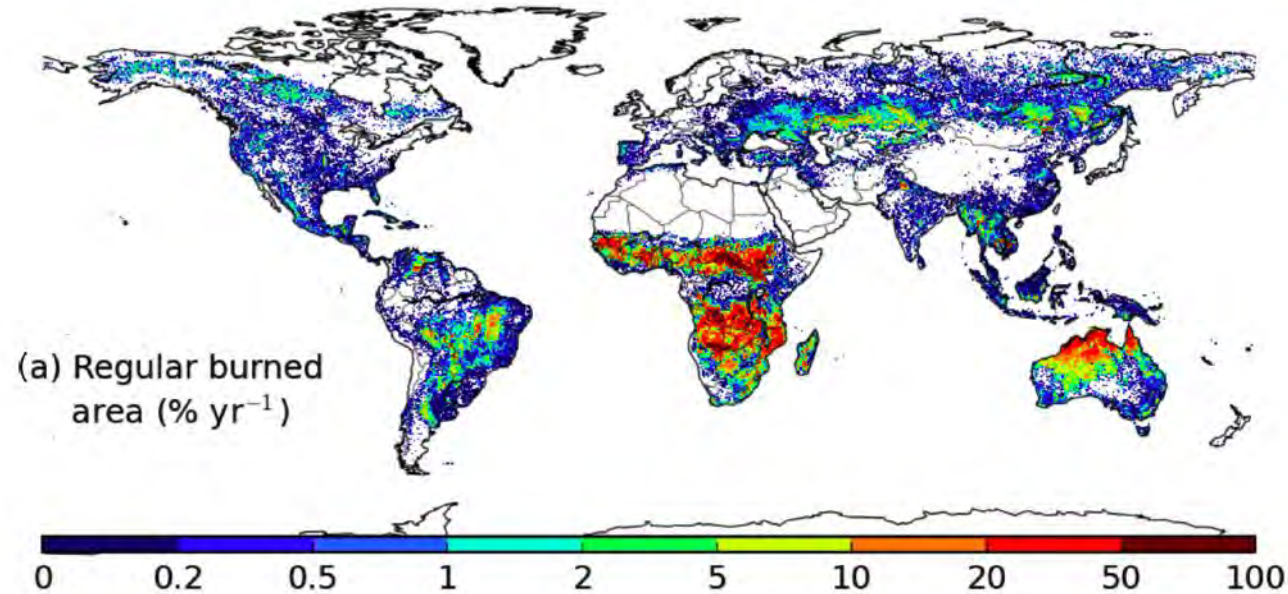
GFED: Global Fire Emissions Database

FINN: Fire INventory for NCAR

GFAS: Global Fire Assimilation System (CAMS)

DMB determined using distinct satellite data products for each inventory

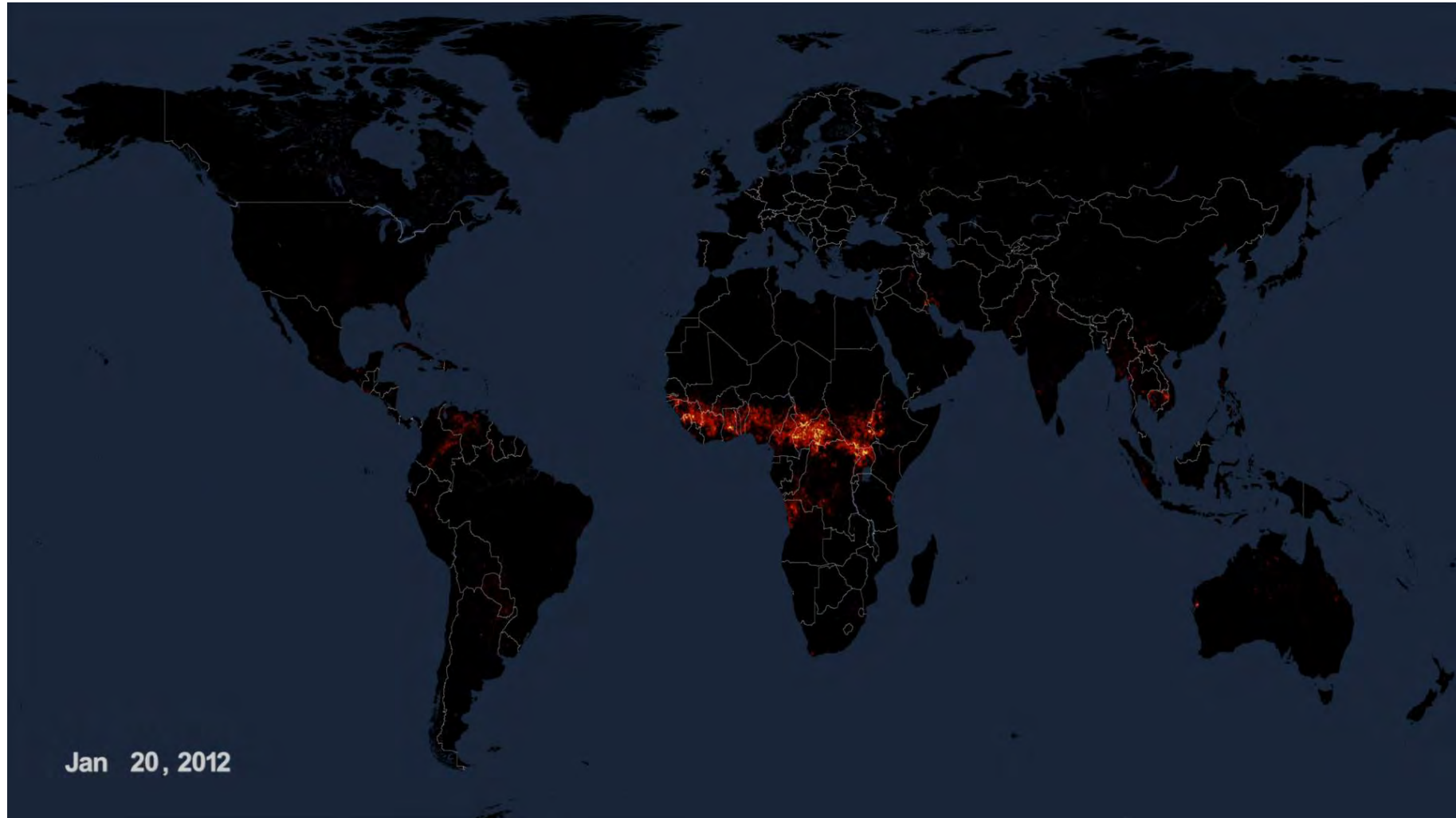
GFED Uses Burned Area



Small fires:
Parameterization that
uses MODIS fire counts

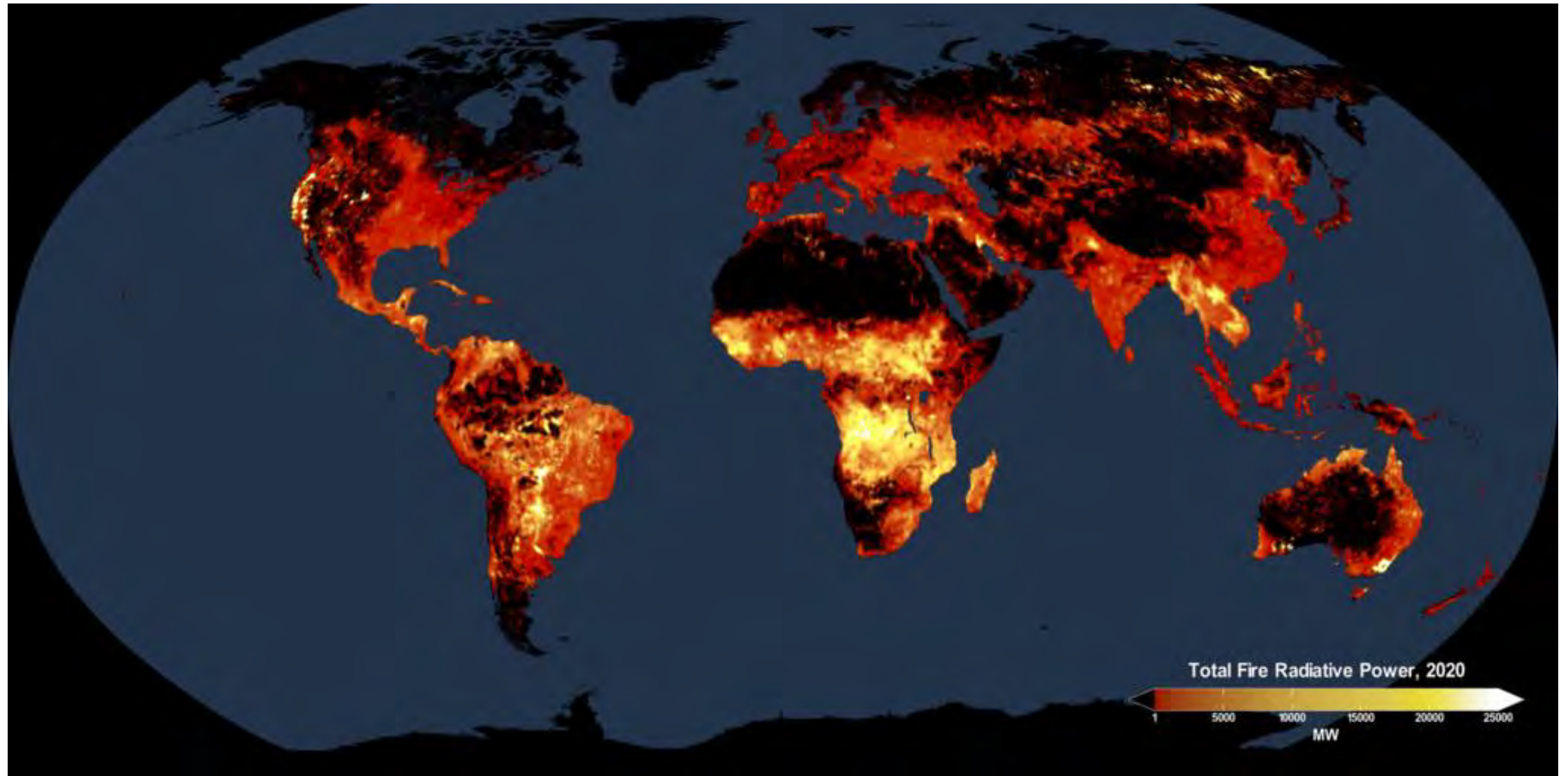
[van der Werf, 2017]

FINN Uses Fire Counts



Data from MODIS (1 km) and more recent finer resolution VIIRS (375 m)

GFAS Uses Fire Radiative Power



Landcover Specific Emission Factors (EF)

$$\text{Emission} = \text{DMB} \times \text{EF}$$

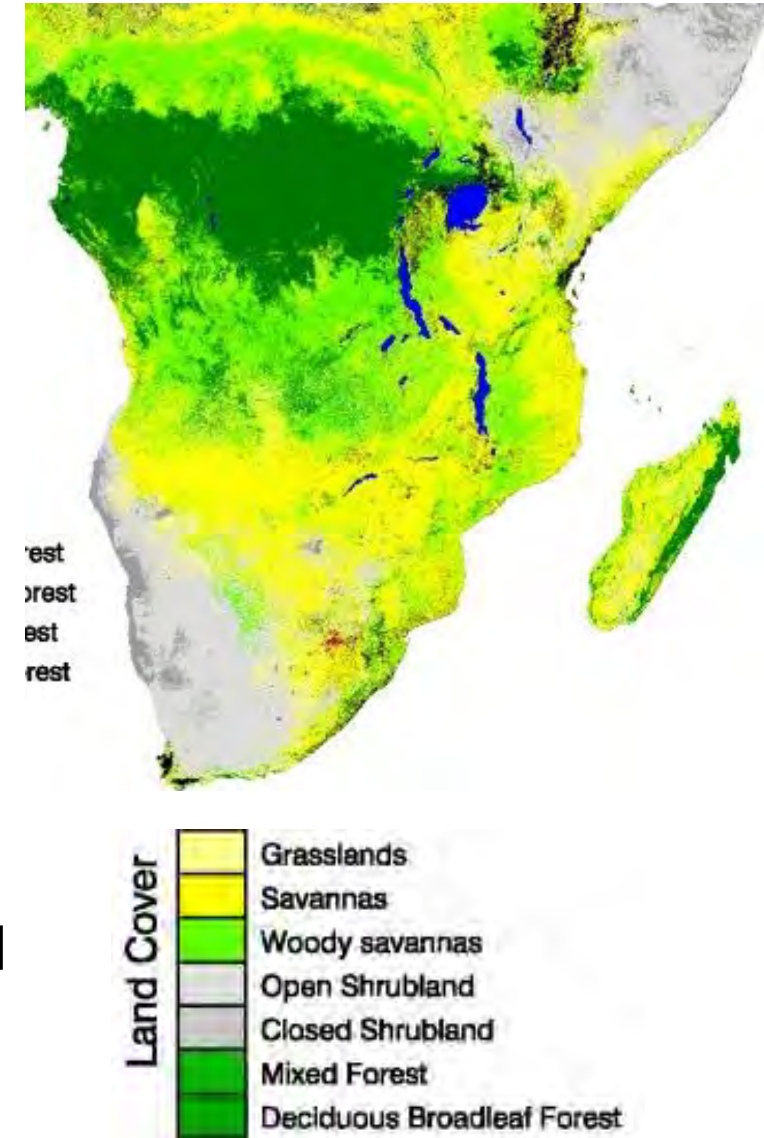
Vegetation type	Emission factor [g kg ⁻¹] ^a		
	GFEDv4s	FINNv2.5 ^b	GFASv1.2
NO _x as NO			
Tropical forest	2.55	2.6	2.3
Savanna	3.9	3.9	2.1
Woody savanna ^c	—	3.65	—
NH ₃			
Tropical forest	1.33	1.3	0.93
Savanna	0.52	0.56	0.74
Woody savanna ^c	—	1.2	—

[Marais et al., 2025]

Biggest difference for NO_x applied to savannas

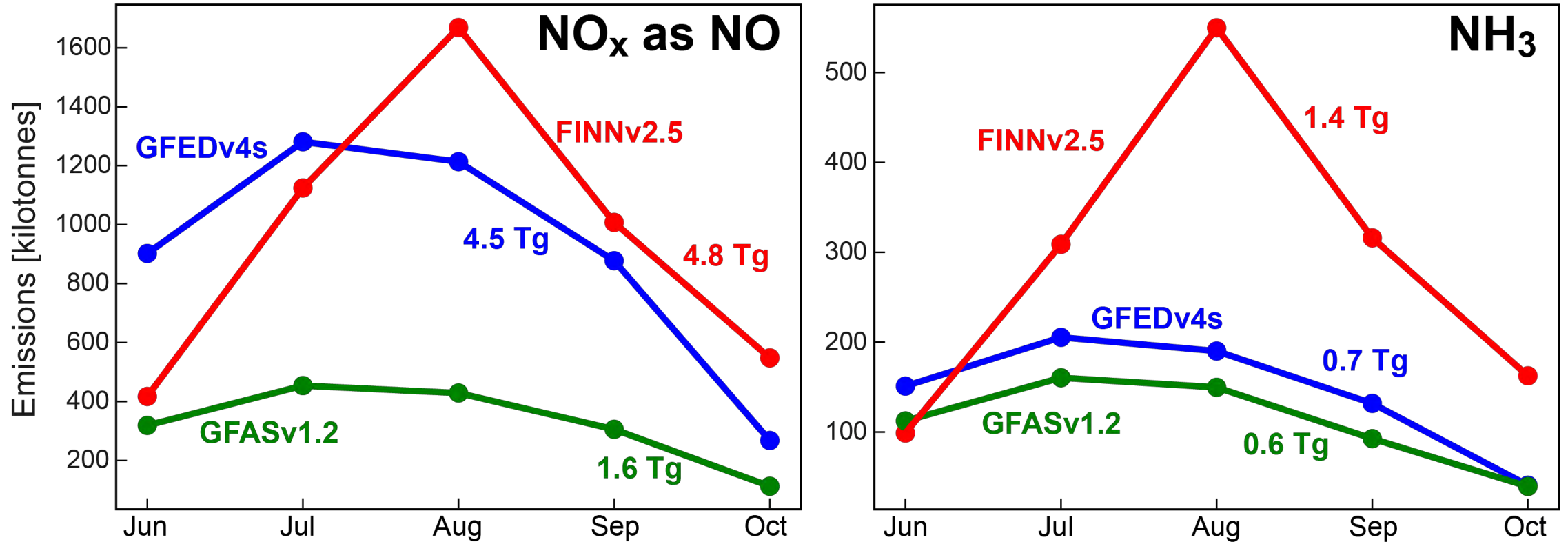
FINN emits NO_x as NO and NO₂

MODIS Landcover



Reactive Nitrogen Emissions in Southern Africa

Monthly bottom-up June-October 2019 emissions

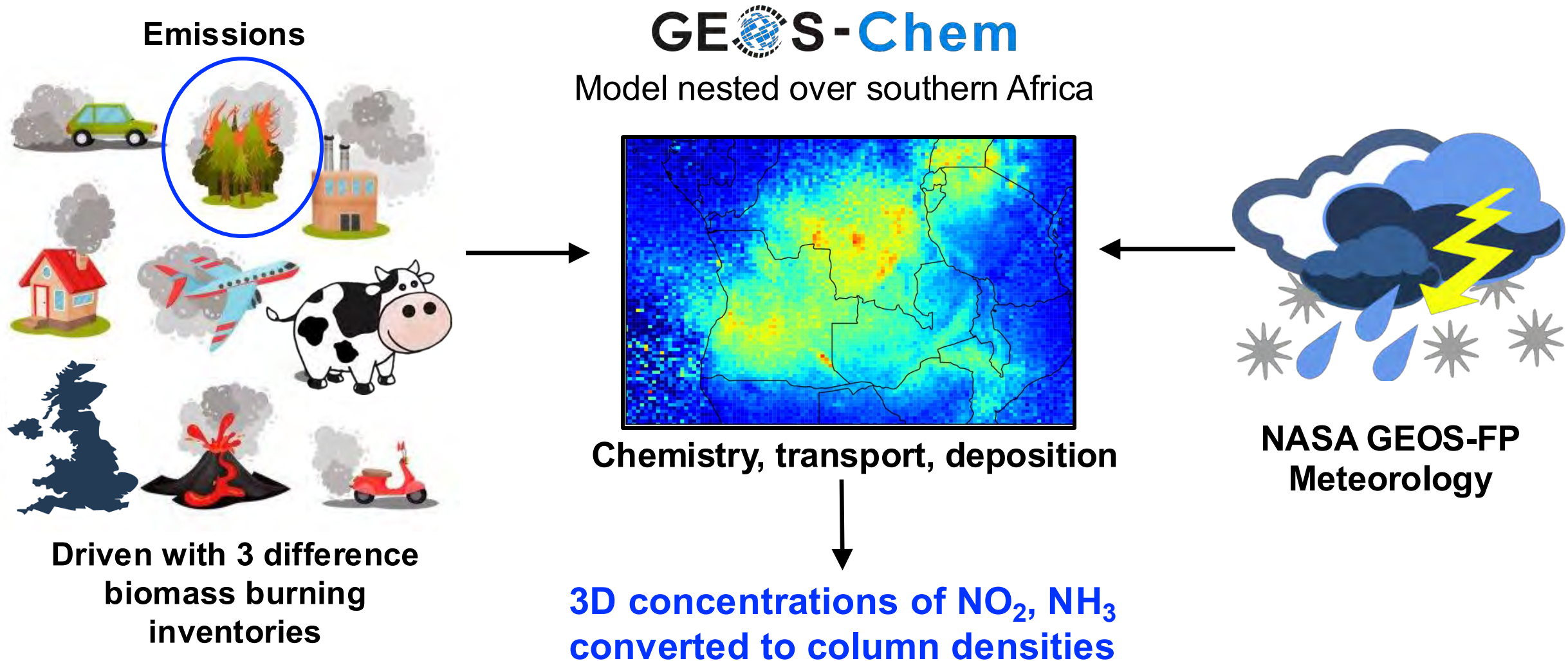


Other differences: Time and spatial resolution, injection height, landcover classes

Mostly savanna fires

In FINN, ~20-times more fuel consumed for tropical forest fires (smouldering) than the other inventories

Drive GEOS-Chem with all Three Inventories

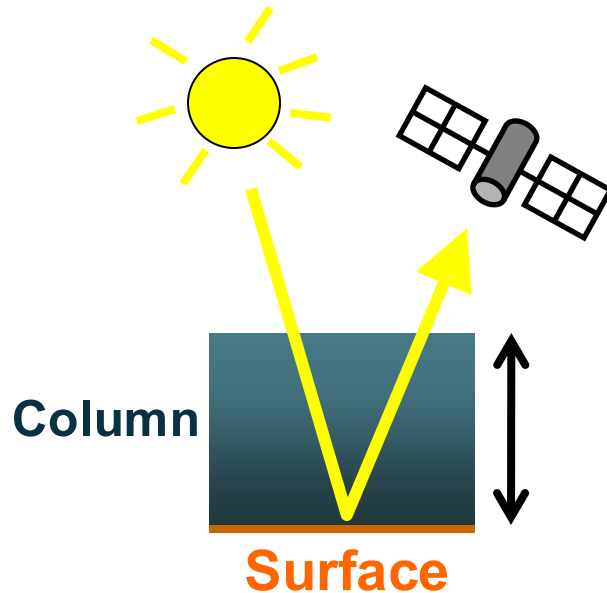


Simulate model with each inventory turned on to compare the model to IASI and TROPOMI

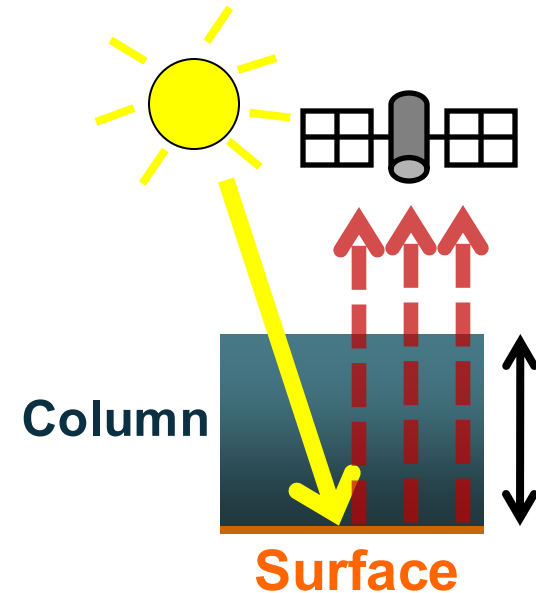
Sample the model at the same overpass time as the instruments

Account for Instrument Vertical Sensitivities

UV-visible instrument (TROPOMI)



Infrared instrument (IASI)



Sensitivity peaks in
mid troposphere

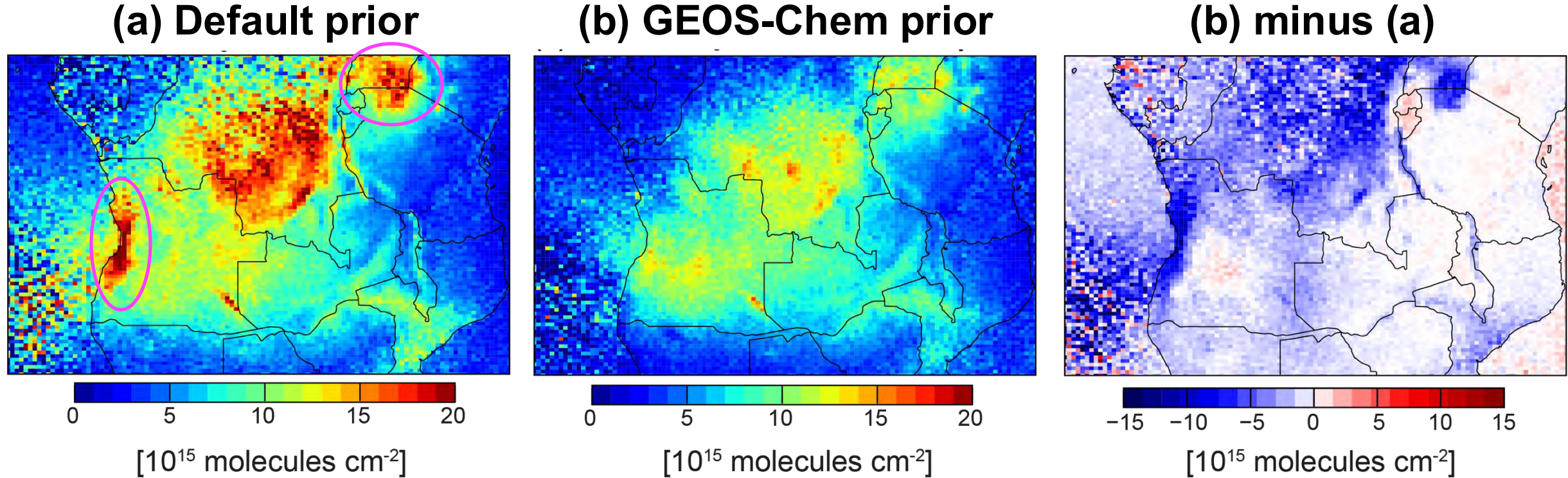
Different approach for each instrument:

TROPOMI: apply averaging kernels (quantifies vertical sensitivity) to GEOS-Chem

IASI: reprocess (re-retrieve) IASI NH_3 with local GEOS-Chem a priori profiles

Reprocess IASI with Local GEOS-Chem Priors

IASI columns for July-October 2019. Prior from GEOS-Chem using FINN



Overall decline in columns with local a priori, as more NH₃ placed higher up

Less noisy over Atlantic Ocean east of Angola and northern Namibia

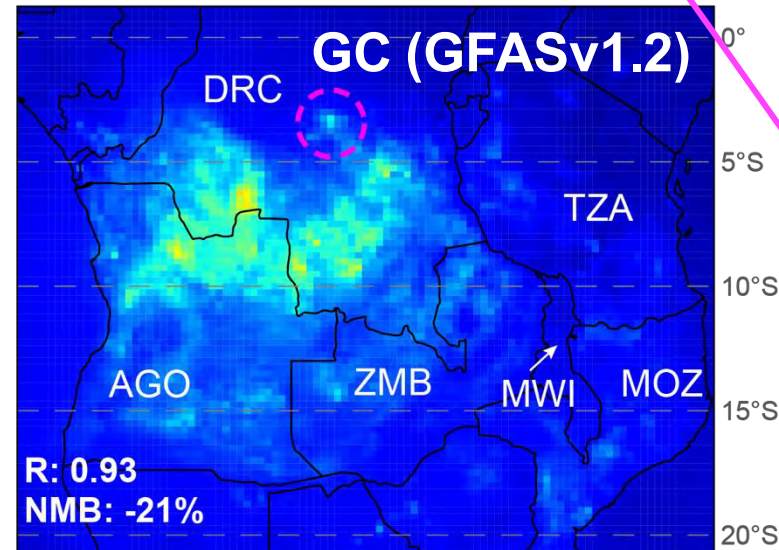
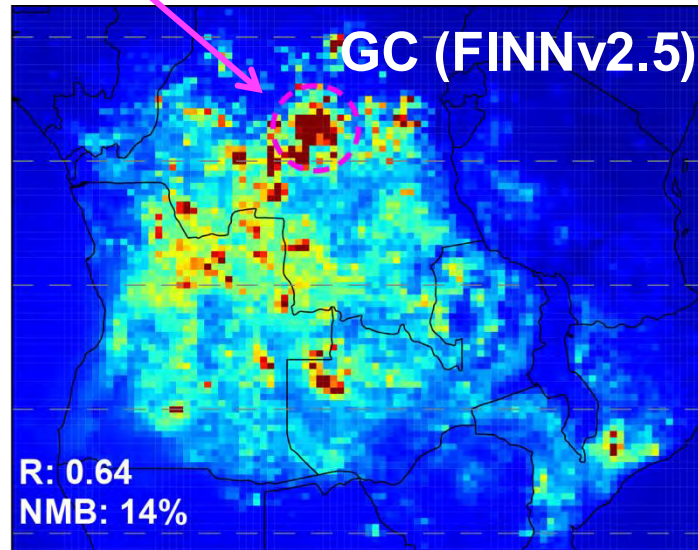
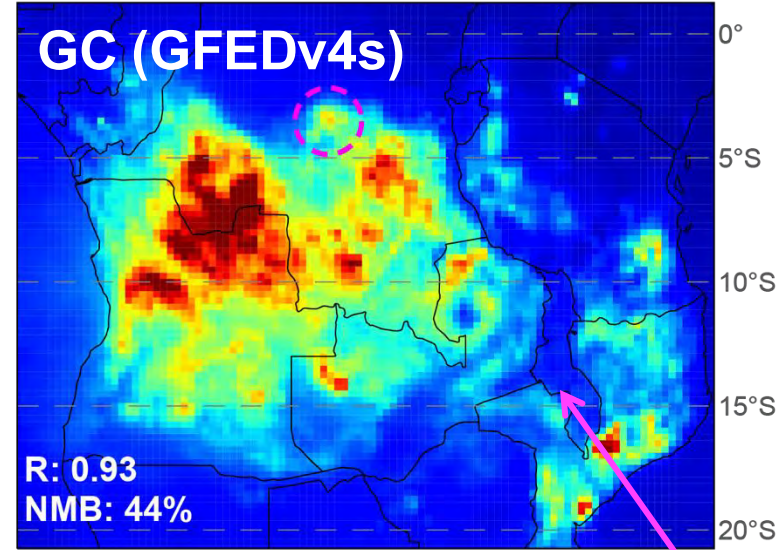
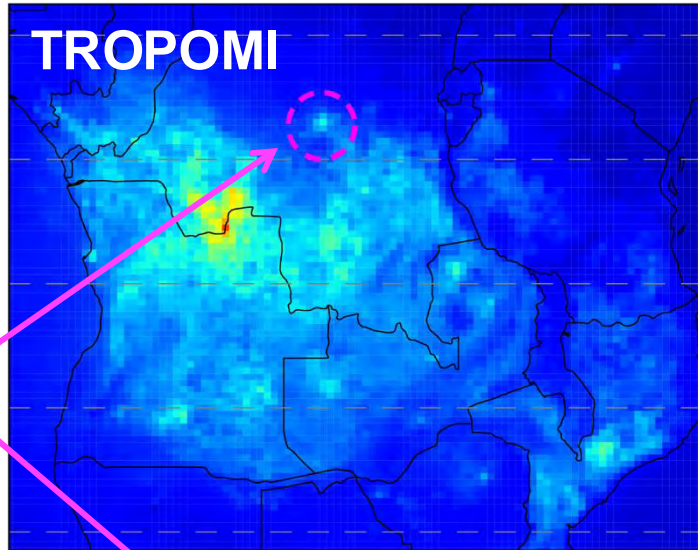
More retrievals pass quality checks

Evaluate Inventory NO_x Emissions with TROPOMI NO₂

NO₂ vertical column densities for Jun-Oct 2019

GC: GEOS-Chem

Far more NO_x
from tropical
forests in FINN
(fuel load)



GFED and GFAS NO₂
spatially similar, but
>50% difference due
to emission factors

Low emissions in
Malawi, as spread of
fire suppressed by
dense population



Impact of Different NO_x Emissions on Ozone Formation

Ozone production efficiency (**OPE**) = ozone produced per mass unit NO_x emitted

GFAS: 13 Tg O₃ (Tg NO)⁻¹

FINN: 9.6 Tg O₃ (Tg NO)⁻¹

GFED: 6.9 Tg O₃ (Tg NO)⁻¹

FINN OPE > GFED OPE, as far more VOCs and CO in FINN:

FINN: 108 Tg CO and 13 Tg C for 21 NMVOCs

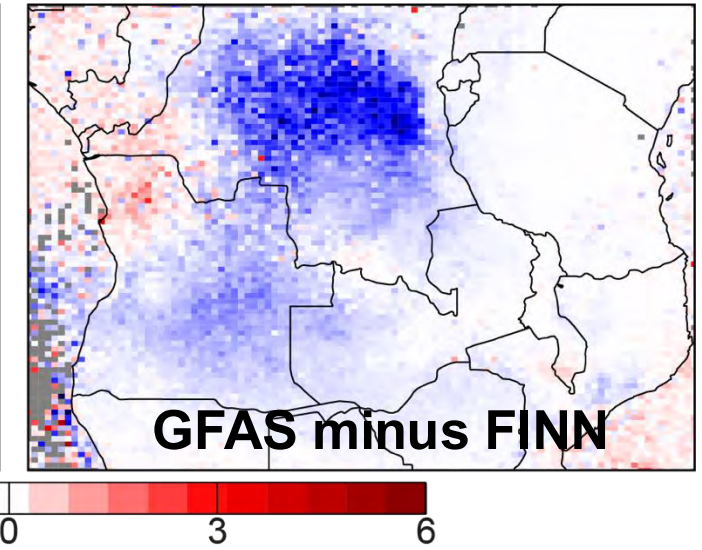
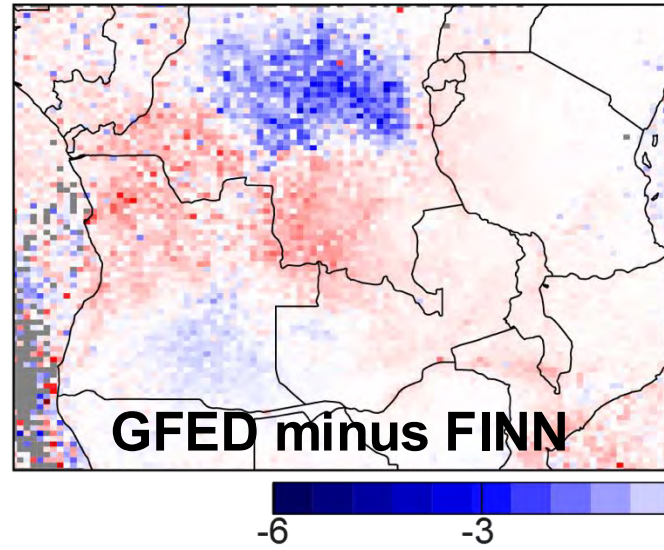
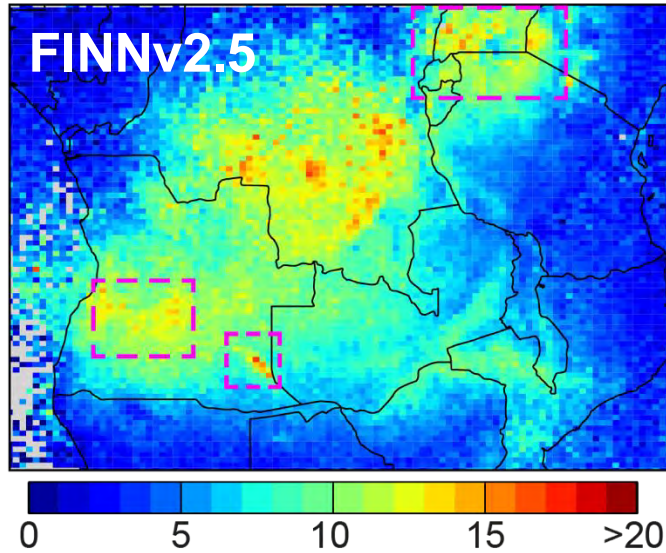
GFED: 82 Tg CO and 2 Tg C for 13 NMVOCs

Less NO_x from GFAS increases OPE, as O₃ more sensitive to NO_x

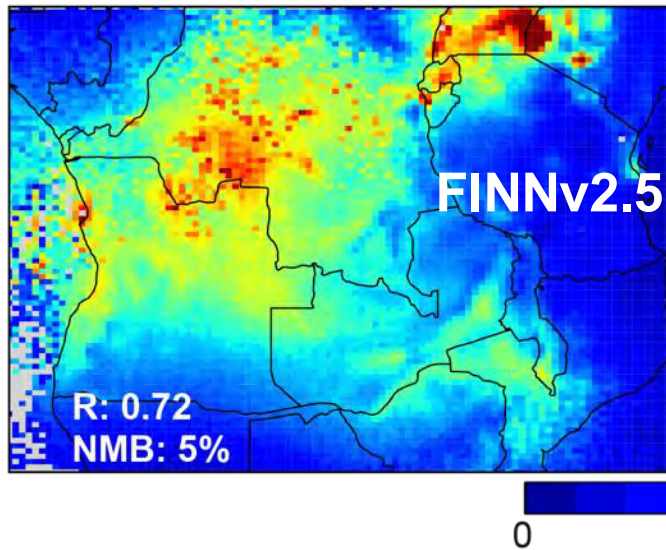
Evaluation of Inventories with Satellite Observations

NH₃ vertical column densities for Jul-Oct 2019 [10^{15} molecules cm⁻²]

IASI with
GEOS-Chem
prior:



GEOS-Chem:



June excluded, as no inventories consistent with IASI observations ($R < 0.5$)

Top-down emissions estimate

Convert atmospheric **column concentrations** to surface **emissions** using a **model**

COLUMNS

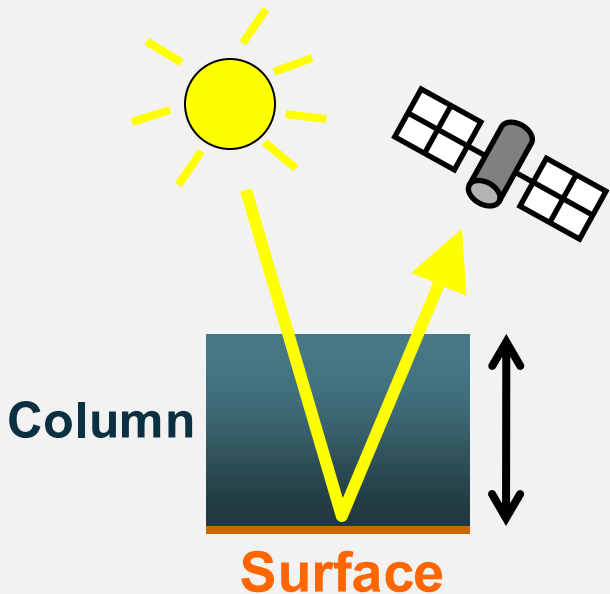


Conversion Factor



EMISSIONS

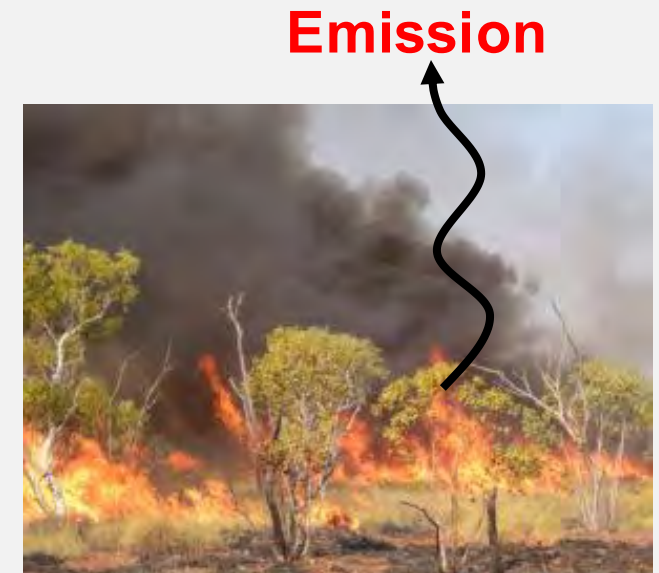
Satellite columns



**Column-to-Emission ratio
(model)**



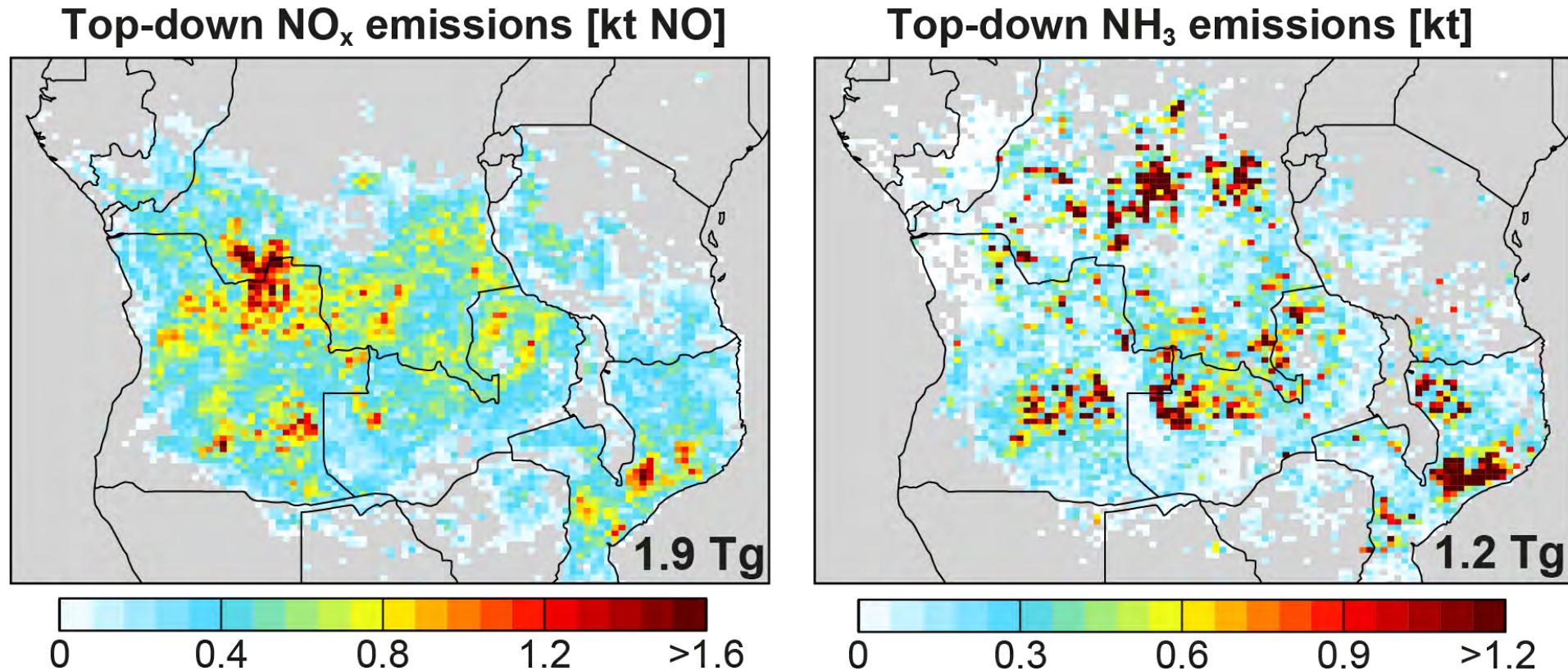
**Satellite-derived
surface emissions**



Simple mass balance approach, as it's a first order problem (very large errors)

Use best-performing inventory (**GFAS** for NO_x , **FINN** for NH_3) for gridsquares where open fires > 50% total emissions

Top-down Emissions with Best Performing Inventories



Distribution normal for NO_x , long-tailed for NH_3

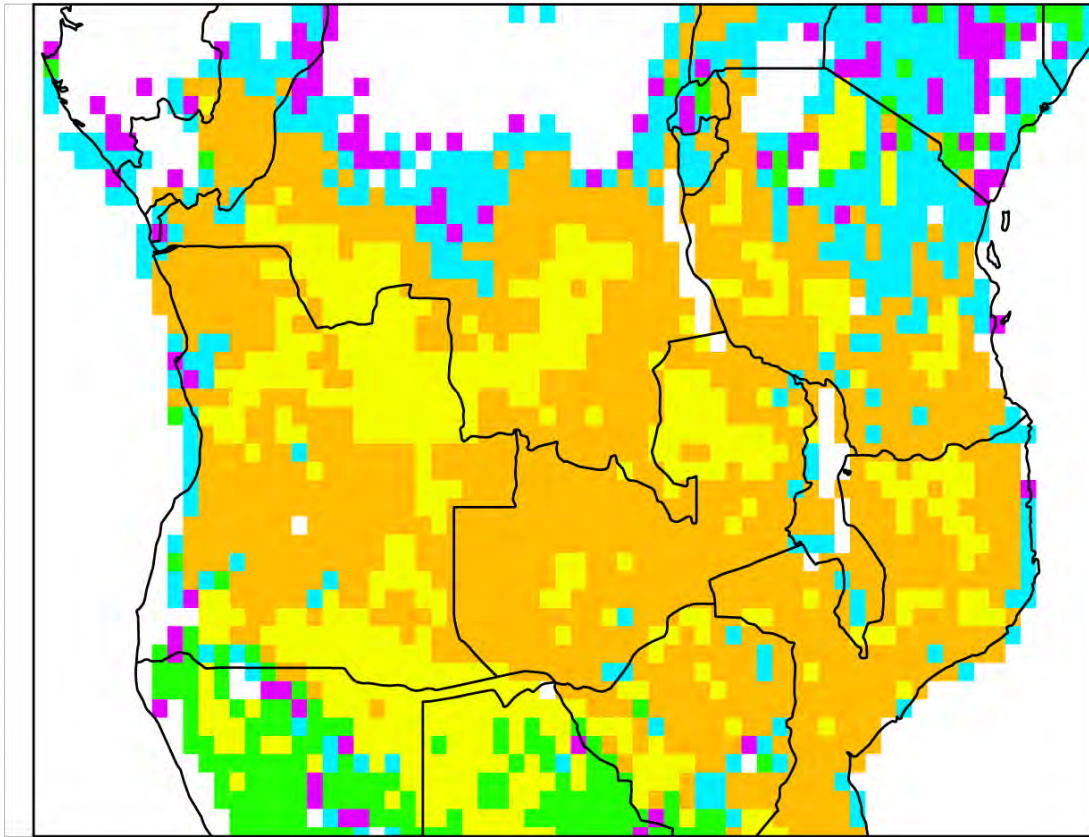
Correlation between top-down NO_x and NH_3 weak ($R < 0.4$), but strong in inventories ($R > 0.8$), as none account for the impact of combustion efficiency/pyrolysis regime on emission factors

Emissions peak in similar month to bottom-up: July and August for NO_x and August in NH_3

Total anthropogenic emissions in Europe in 2019: ~1.4 Tg N for NO_x and ~8 Tg for NH_3

Consistency of Top-down Emissions and Pyrome Regimes

Archibald et al. (2013) pyrome regimes



NO_x emissions coincide with intense fires that would tend to flame (efficient)

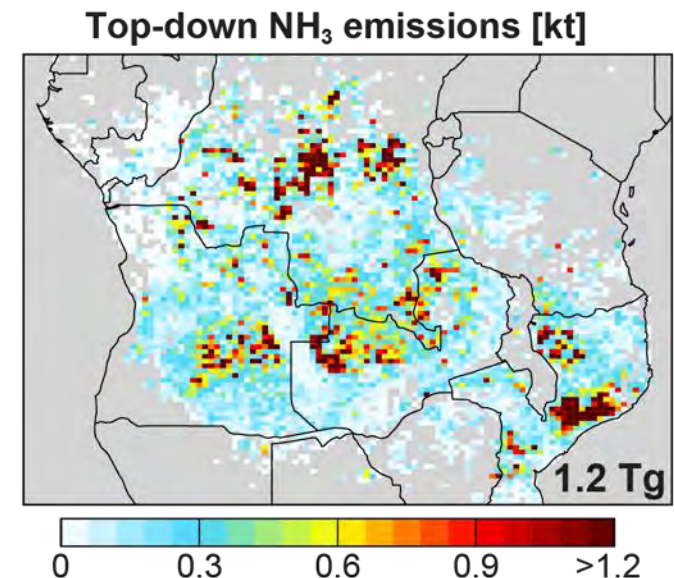
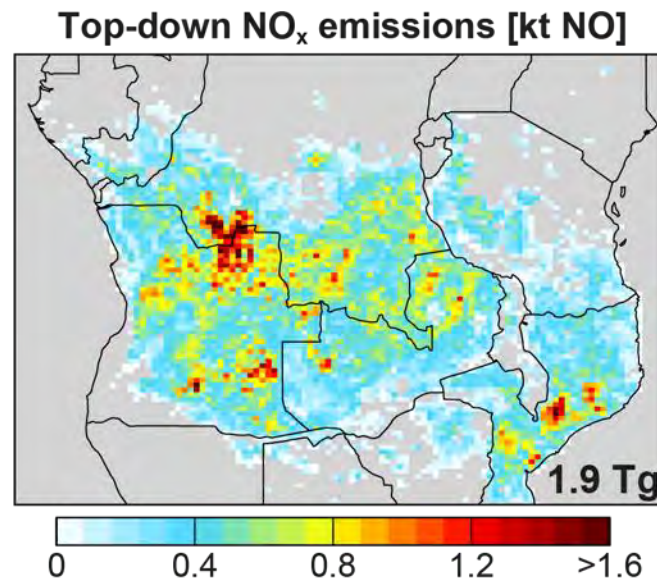
Pyrome type

(frequency, intensity, size)

- rare, cool, small (RCS)
- intermediate, cool, small (ICS)
- frequent, cool, small (FCS)
- frequent, intense, large (FIL)
- rare, intense, large (RIL)

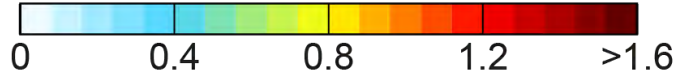
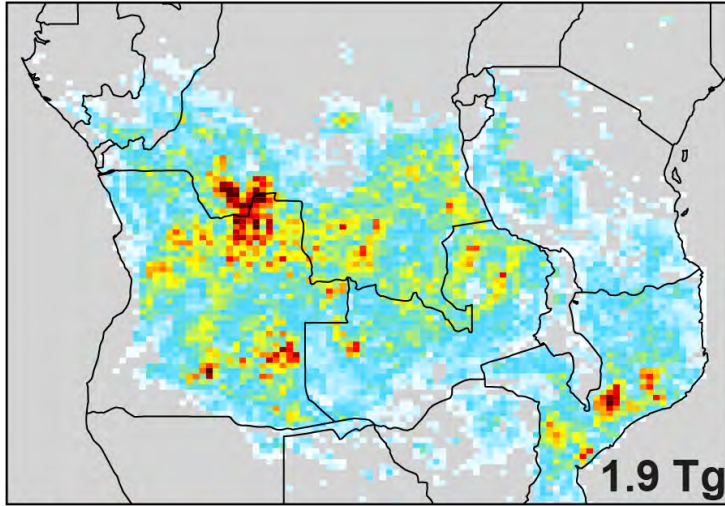
NH₃ emissions

coincide with cool fires that would tend to smoulder (inefficient)

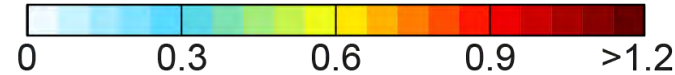
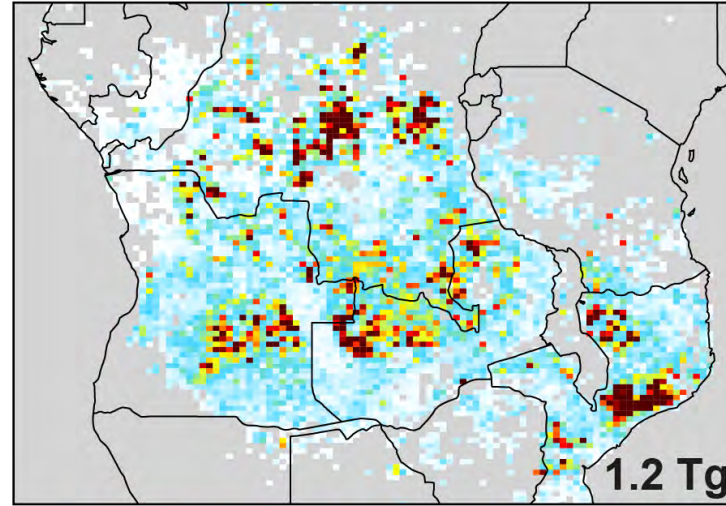


Top-down vs Best Performing Inventory Emissions

Top-down NO_x emissions [kt NO]



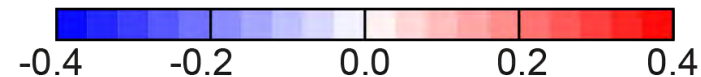
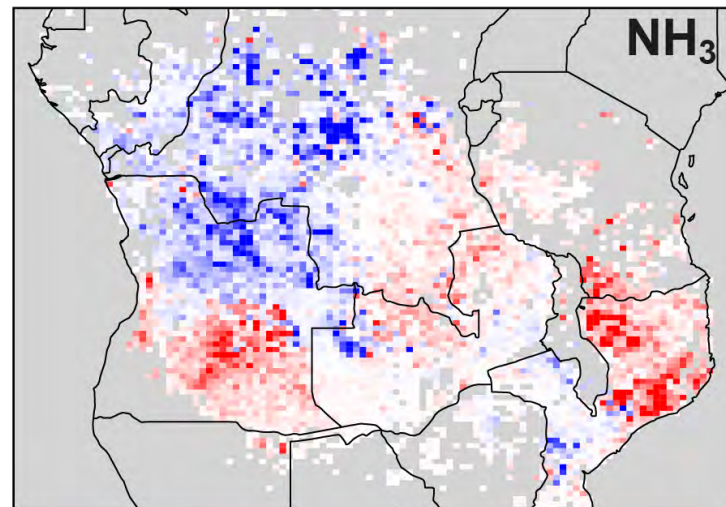
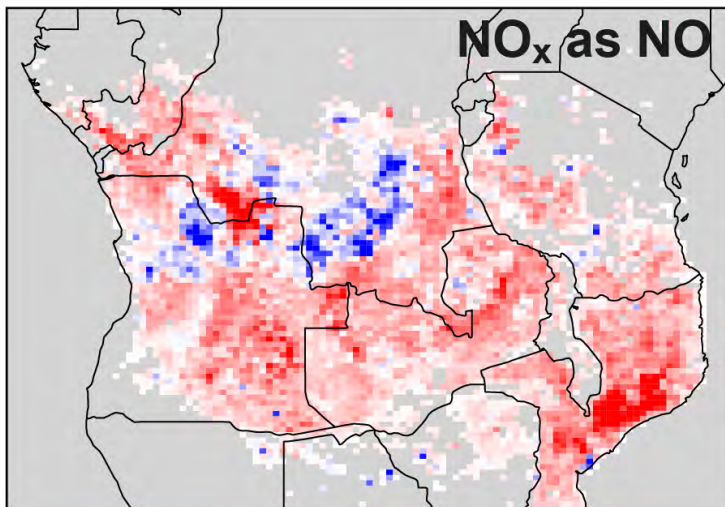
Top-down NH_3 emissions [kt]



Observationally constrained
ozone production from
biomass burning NO_x
emissions in southern Africa:

25 Tg O_3

Top-down minus bottom-up emissions [kt]



Emissions uncertainty estimate
(mostly due to instruments):

31% for NO_x , 33% for NH_3

NO_x : 1.9 ± 0.6 Tg NO

NH_3 : 1.2 ± 0.4 Tg

First Part Summary

Top-down approach could be further refined with more complex inverse modelling methods or with iteration. Regardless, highlights the large disparities between top-down and bottom-up emissions.

Inventories collocate NH_3 and NO_x emissions (smouldering and flaming fires), but these are mostly separate in the top-down estimates

Could adopt hybrid approach: FINN for smouldering fire emissions of NH_3 , VOCs, CO, organic aerosols and methane and GFAS or GFED for flaming fire emissions of NO_x , black carbon and CO_2

Choice of emission factors remains an issue without observations to constrain these

Critical need for observations to validate satellite observations and top-down estimates. Ideally in National Parks and in anticipation of geostationary Sentinel-4 IR instrument measuring NH_3 and CO (both markers of smouldering fires)

Invited contribution in Royal Society of Chemistry *Environmental Science: Atmospheres* journal (<https://pubs.rsc.org/en/content/articlelanding/2025/ea/d5ea00041f>)

Hotspot NO_x Emissions in Sub-Saharan Africa

The largest anthropogenic point source emissions of NO_x are in Sub-Saharan Africa (South Africa)

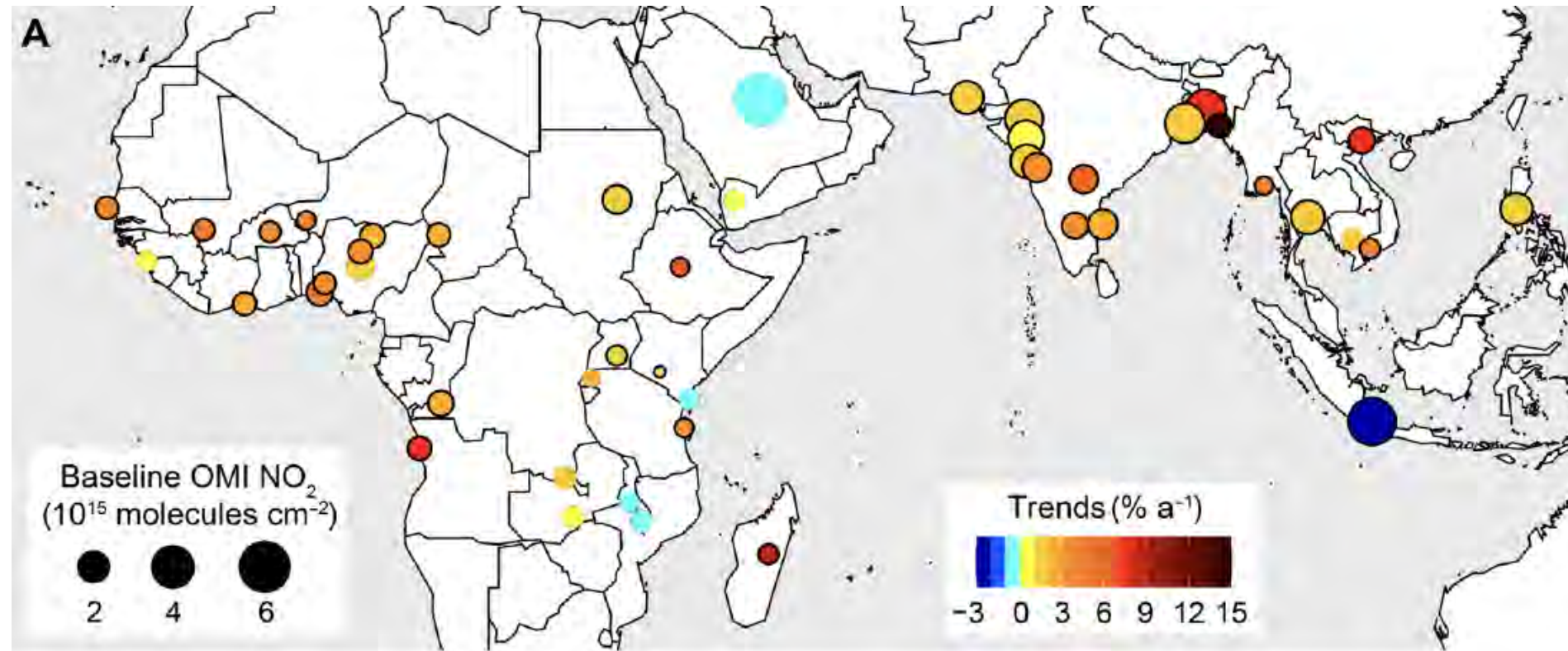
Rank	Lat [° N]	Long [° E]	Emissions [kg s ⁻¹]	Error [kg s ⁻¹]	Power plants (GPPD) ¹	Cities (WCD) ¹	Comment ²
1	−26.2875	29.1625	2.76	0.47	Matla; Kriel		
2	−26.5625	29.1625	2.47	0.39			Secunda CTL ³
3	−23.6875	27.5875	2.47	0.56	Matimba		also Medupi (not listed in GPPD)
4	−26.7375	27.9875	2.03	0.44	Lethabo	Vereeniging	
5	−27.1125	29.7875	2.03	0.31	Majuba		
6	22.3875	82.6875	2.01	0.59	Korba		
7	40.6375	109.7375	1.81	0.57	Baotou	Baotou	
8	21.0125	107.1375	1.80	0.42	Quang Ninh	Ha Long; Cam Pha	
9	−26.0875	28.9875	1.74	0.32	Kendal		
10	−32.4125	151.0125	1.73	0.30	Bayswater; Liddell		

[Beirle et al., 2023]

Unregulated coal-fired power plants (Kriel, Matimba, Lethabo, Majuba, Kendal)
and a synthetic fuels plant (Secunda)

Unprecedented Increases in NO_x in Cities in Africa

2005-2018 trends in Ozone Monitoring Instrument (OMI) NO₂ over fast-growing cities in the tropics

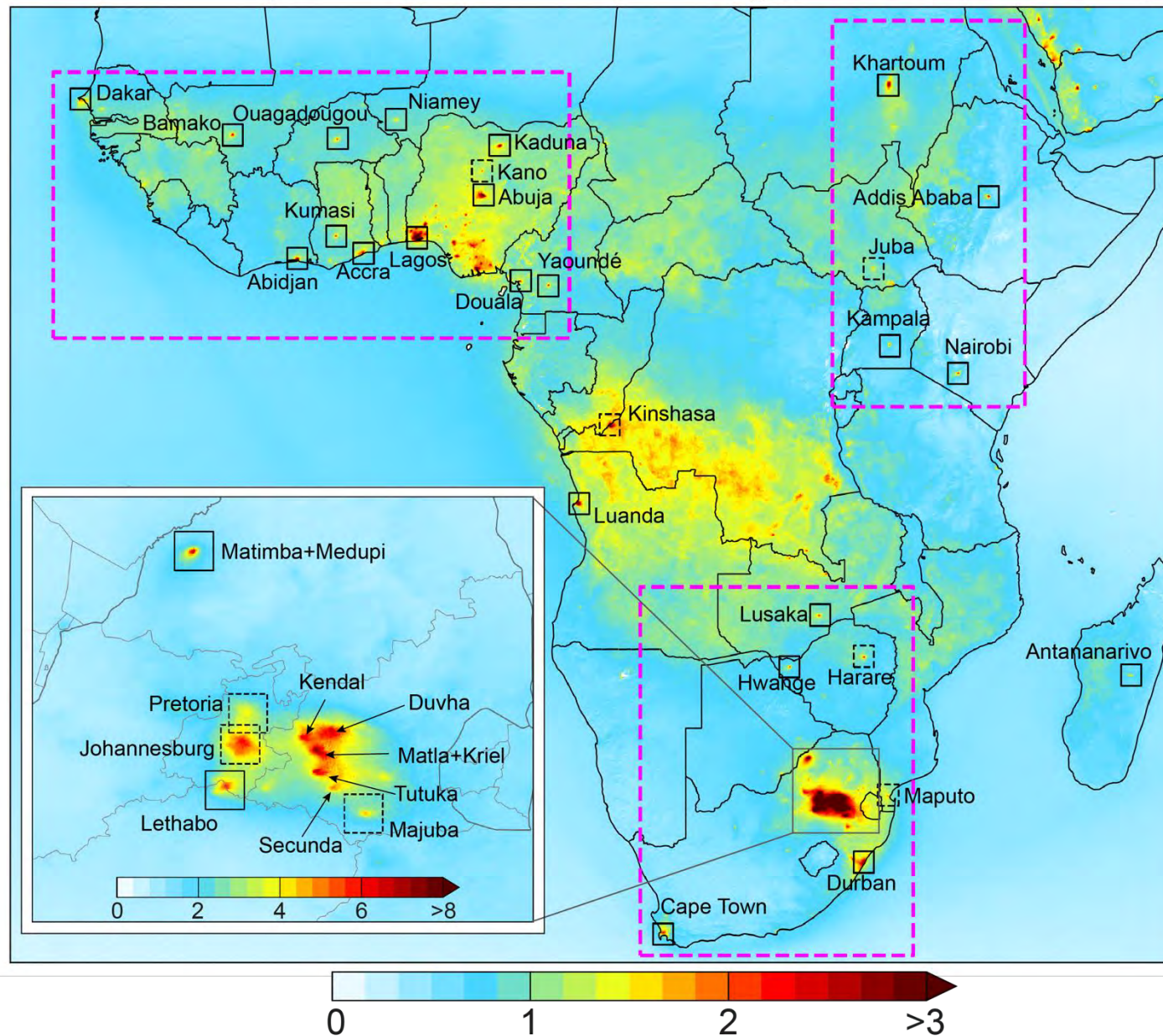


[Vohra et al., 2022]

OMI: earlier generation version of TROPOMI with coarser resolution, but longer record

Urban and Point Sources Resolved with TROPOMI

Annual multiyear mean TROPOMI NO₂ [10^{15} molecules cm⁻²]



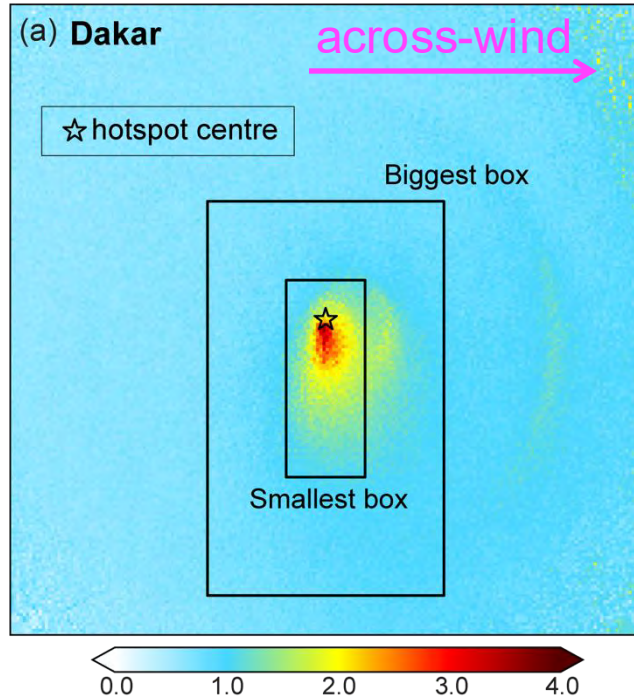
Oversample 4 years of TROPOMI data to finer scale (~2 km) than nadir resolution

Identify 32 isolated hotspots: most urban, 4 power plants

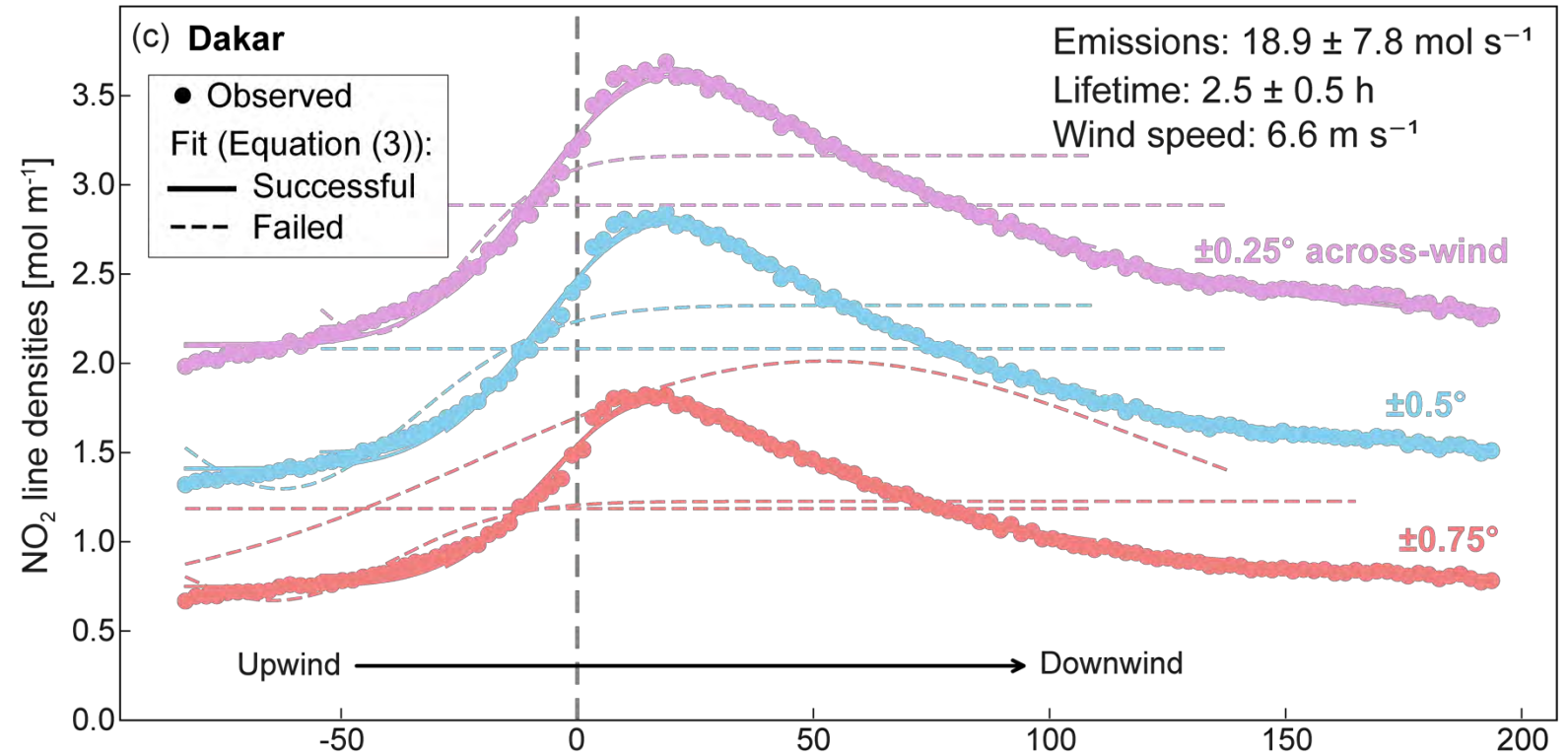
Boxes: dashed if attempt to calculate emissions fails; solid if succeeds

Hotspot NO_x Emissions Inversion Method

Wind rotate TROPOMI NO₂
about the hotspot centre



Sum across-wind NO₂ to yield 1D line densities and apply an
Exponentially Modified Gaussian (EMG) fit

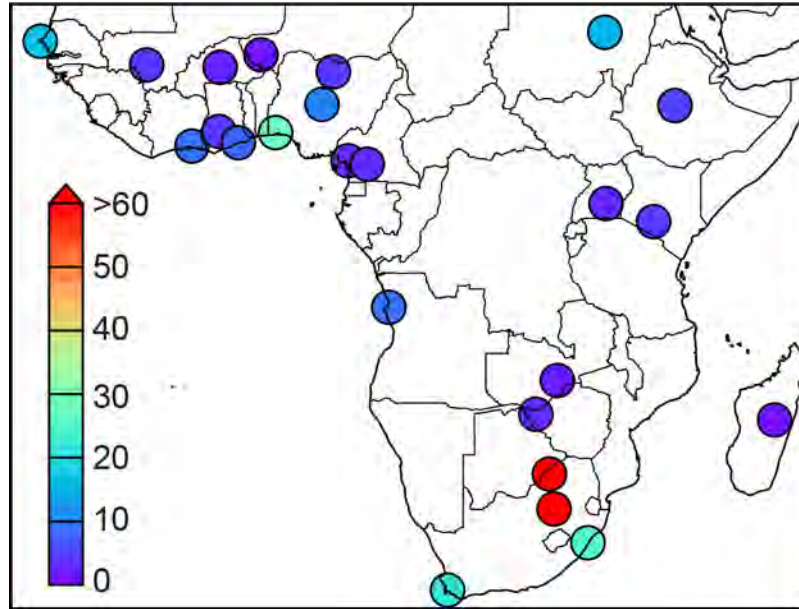


29 out of 36 successful fits for Dakar (Senegal) yielding the following quantities:

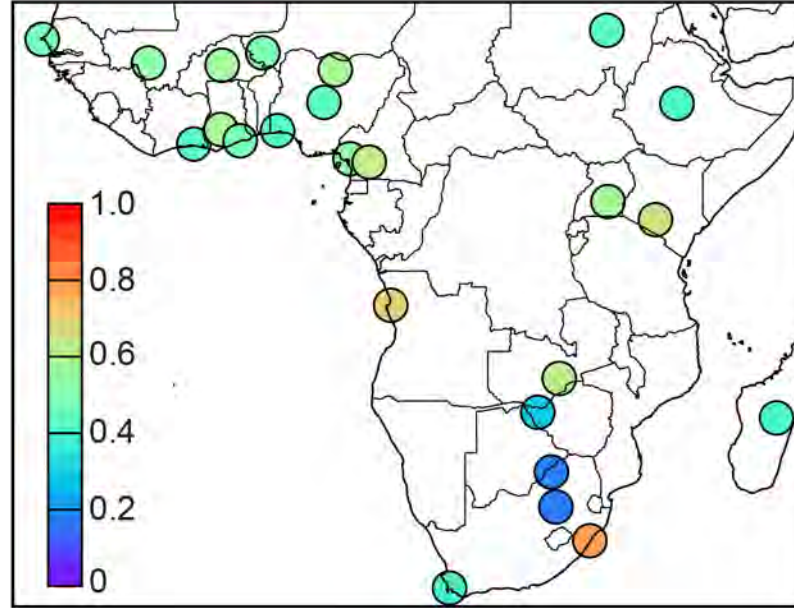
$18.9 \pm 7.8 \text{ mol NO}_x \text{ emitted s}^{-1}$, $2.5 \pm 0.5 \text{ h}$ effective lifetime, 6.6 m s^{-1} wind speed

NO_x Emissions for All Successful Hotspots

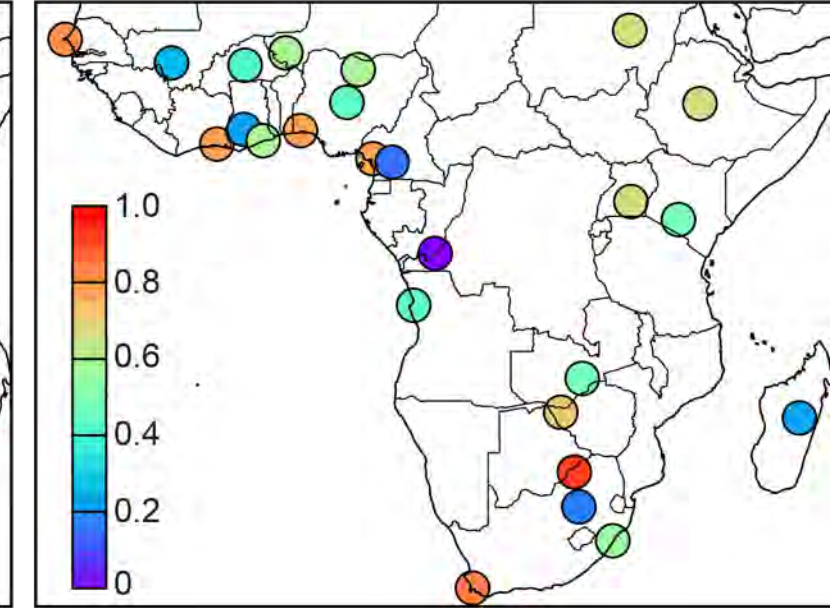
(a) Top-down NO_x emissions [mol s⁻¹]



(b) Relative error (error / emissions)



(c) Relative success (successful / total fits)



Derived emissions for 24 hotspots compared to at most 5 Sub-Saharan hotspots in past studies

Emissions total 207.3 kilotonnes NO

Most hotspots very small (<10 mol s⁻¹) sources of NO_x compared to urban hotspots in Southeast and Southeast Asia (> 60 mol s⁻¹ for Delhi and Dhaka [Lu et al., 2025])

Are Power Plant Hotspot NO_x Emissions Accurate?

South Africa power plant emissions measured with Continuous Emissions Monitoring Systems (CEMS)
(<https://www.eskom.co.za/dataportal/emissions/ael/>)

Matimba and Medupi:

CEMS: 74.1 mol s⁻¹

Top-down (this work): 69.8 ± 25.7 mol s⁻¹

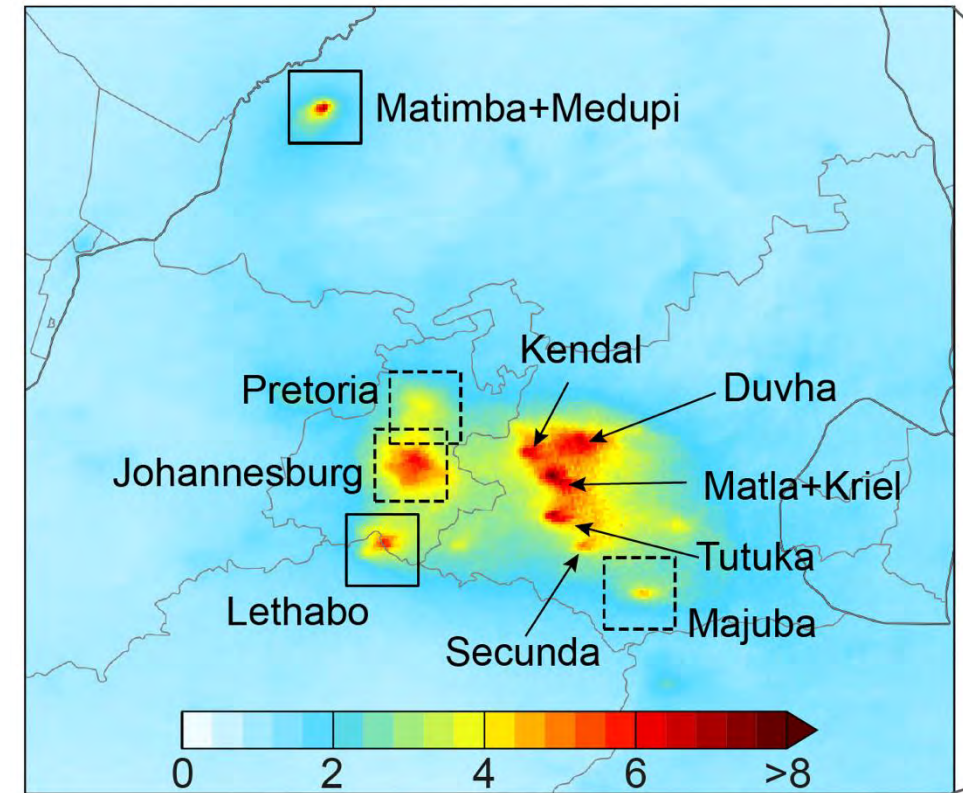
Top-down (Beirle et al., 2023): 82.3 ± 18.7 mol s⁻¹

Lethabo:

CEMS: 65.2 mol s⁻¹

Top-down (this work): 70.4 ± 23.8 mol s⁻¹

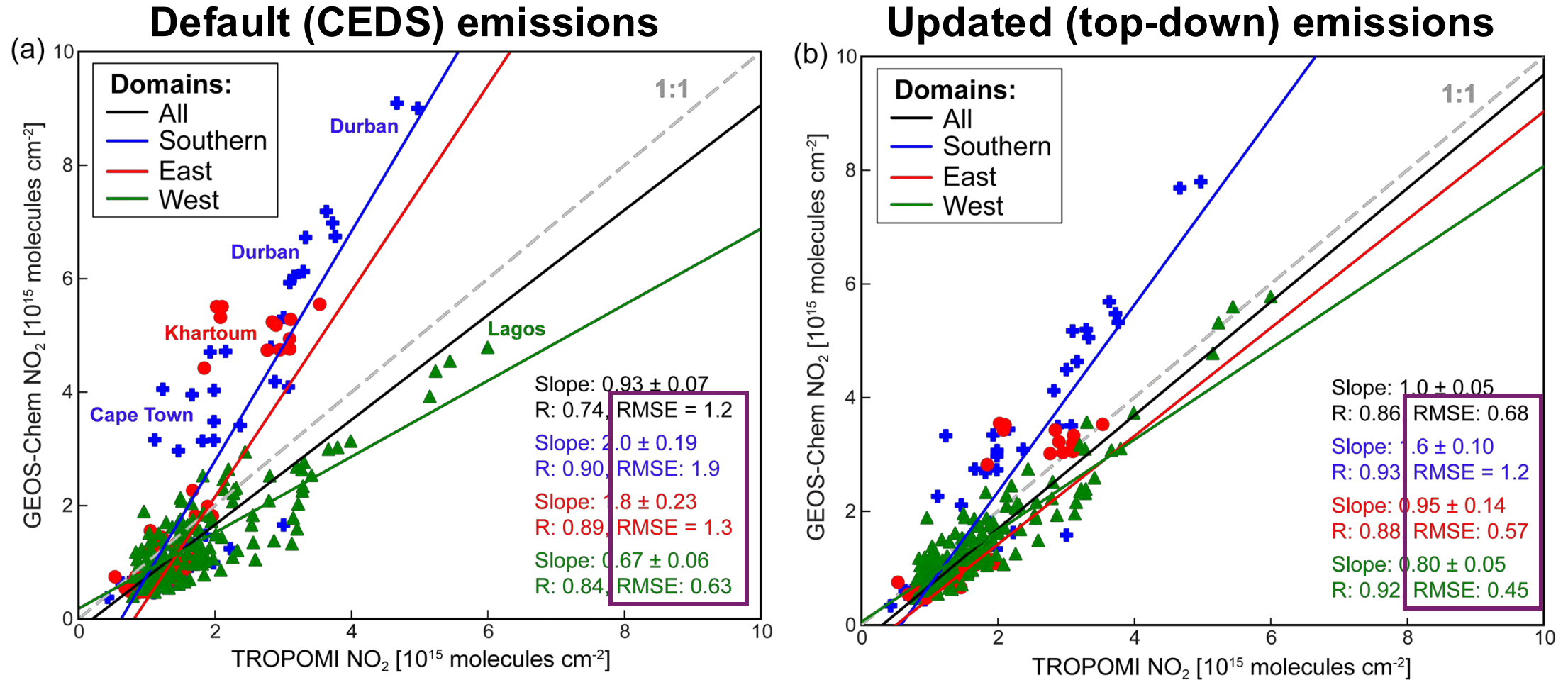
Top-down (Beirle et al., 2023): 67.7 ± 14.7 mol s⁻¹



Our values are within 18-20% of CEMS and within 4-15% of an alternate top-down approach

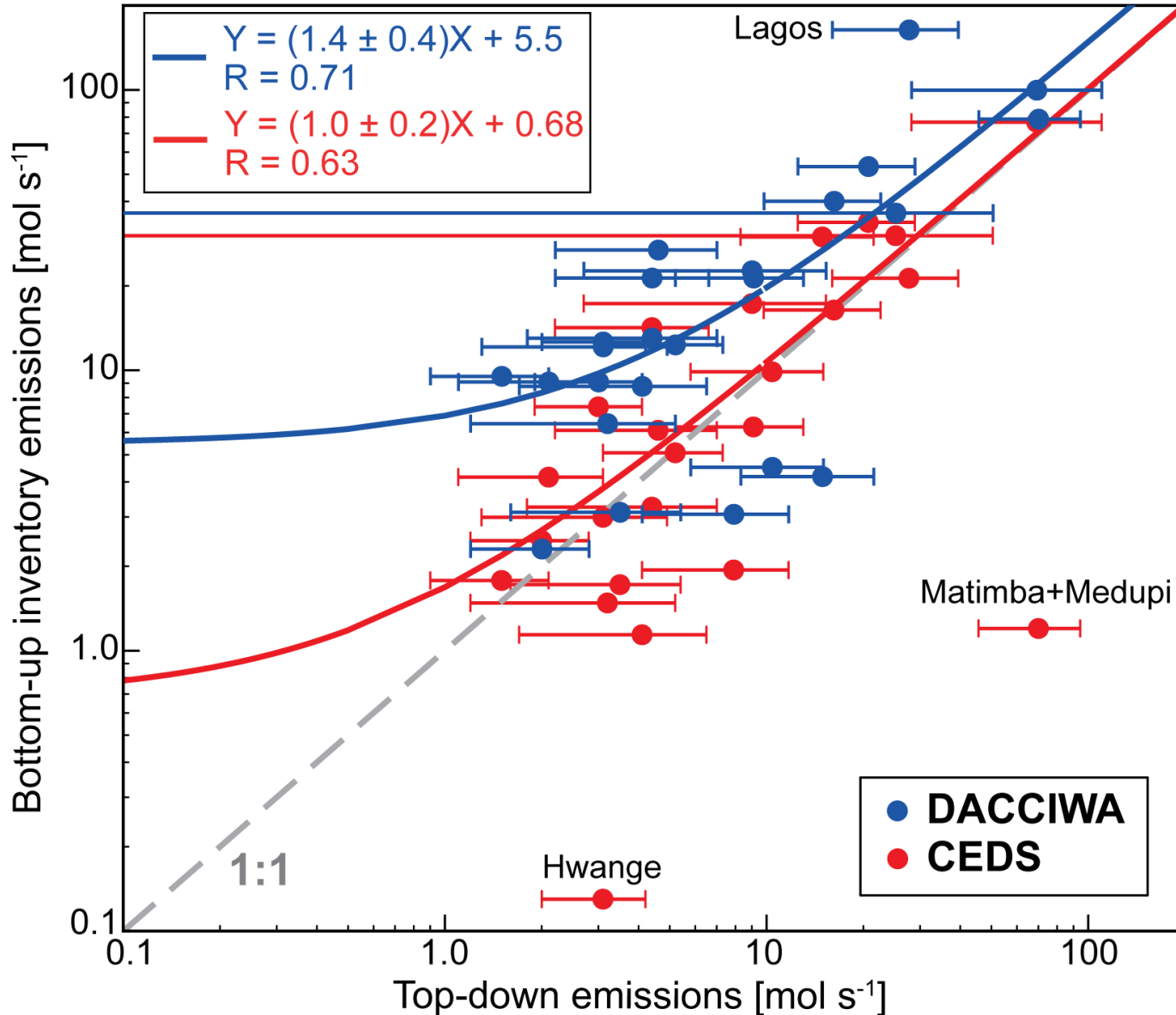
Are Urban Hotspot NO_x Emissions Accurate?

Emissions \longrightarrow **GEOS-Chem** \longrightarrow NO₂ column densities



Urban CEDS emissions total 159 kilotonnes NO, whereas top-down total 135 kilotonnes NO

Top-down versus inventory Hotspot NO_x Emissions



Comparison on log-log scale

DACCIWA: regional emissions inventory for Africa

CEDS: global emissions inventory widely used in models

CEDS on average consistent (slope = 1.0), but missing Medupi and Matimba power plants and underestimate Hwange power plant

DACCIWA on average 40% more than top-down emissions. Almost factor of 6 more for Lagos!

Second Part Summary

Derived annual NO_x emissions for 24 isolated hotspots (21 urban, 3 power plants)

Annual hotspot emissions total 207.3 kilotonnes NO

Urban hotspot emissions range from $< 2 \text{ mol s}^{-1}$ for Antananarivo in Madagascar to $27.7 \pm 11.7 \text{ mol s}^{-1}$ for the megacity Lagos in Nigeria

Coal-fired power plant emissions are $2.7 \pm 0.9 \text{ mol s}^{-1}$ for Hwange in Zimbabwe and similar ($\sim 70 \text{ mol s}^{-1}$) for Lethabo and combined plumes of Medupi and Matimba in South Africa

Top-down estimates are within $\pm 20\%$ of measured CEMS

CEDS inventory urban hotspot emissions decline from 159 to 135 kt NO and model root mean squared error declines from 1.2×10^{15} to 0.71×10^{15} molecules cm^{-2}