Improved determination of natural emissions in Africa from a joint inversion of TROPOMI NO_2 and HCHO in the MAGRITTE CTM

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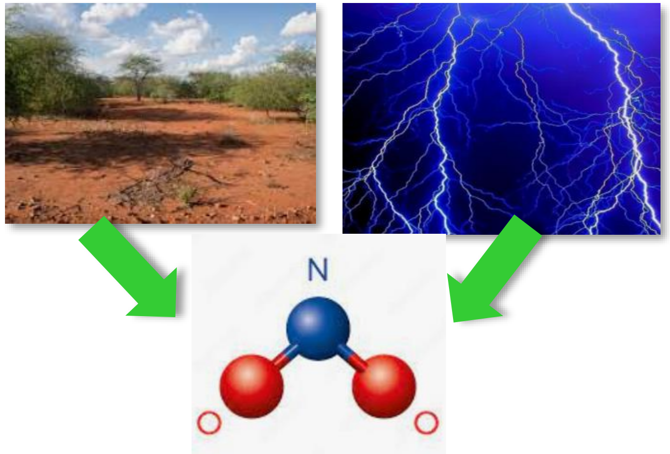
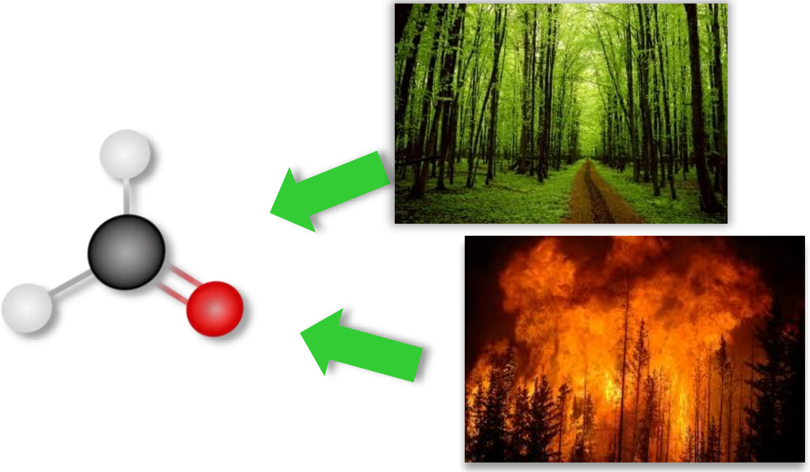
Thanks to data providers

Context and objectives

- Africa is a largely understudied continent; data scarcity leads to large uncertainties in emission estimates
- Natural NOx sources are significant in remote environments; Africa is major contributor for soil/lightning fluxes
- Satellite NO_2 provides valuable information on the spatial distribution and magnitude of natural NO sources

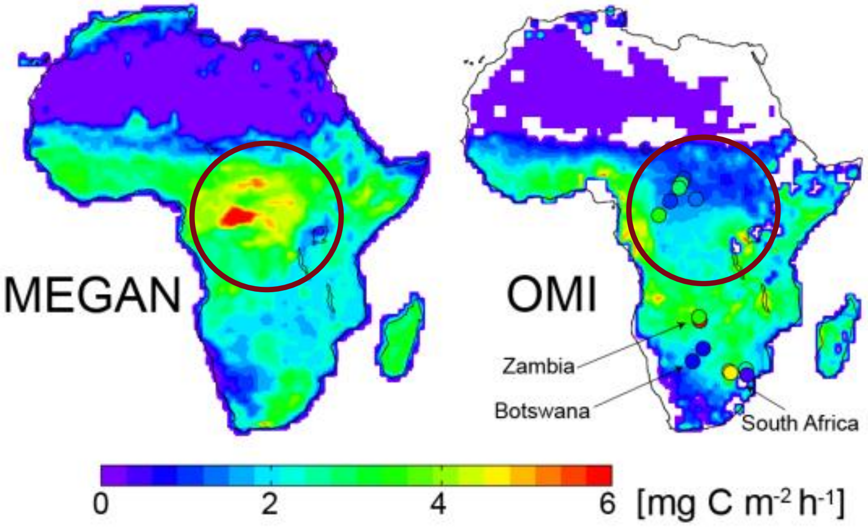


- African forests are a major source of biogenic VOCs, most importantly isoprene
- The oxidation of short-lived biogenic and pyrogenic VOCs dominate HCHO total production in Africa → satellite HCHO is an excellent proxy
- Thanks to high resolution of TROPOMI data, they can be combined with models to derive improved top-down NOx/VOC flux estimates



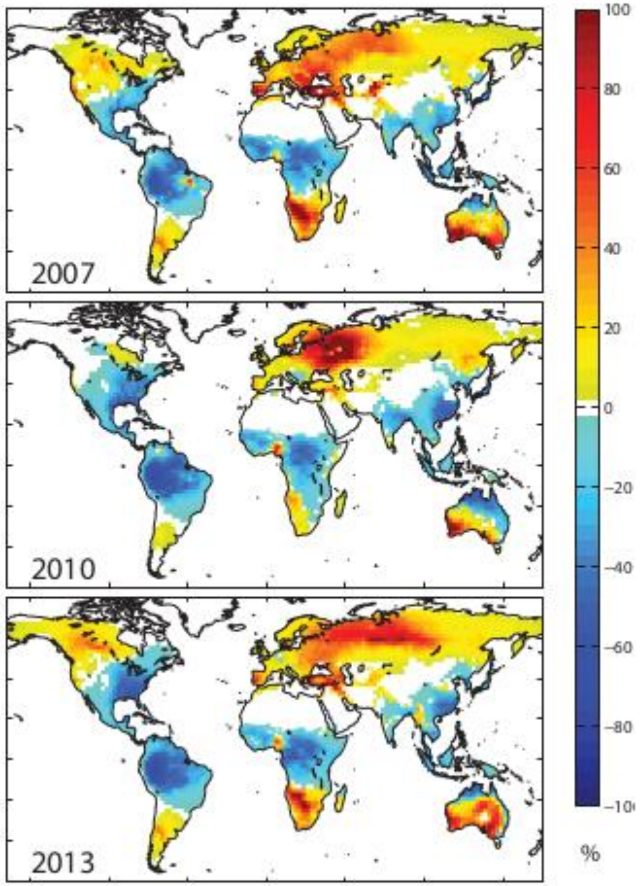
Past inversion studies suggest an overestimation of bottom-up VOC emissions

Models overestimate satellite HCHO over strong biogenic hotspots (Amazonia, Central Africa)



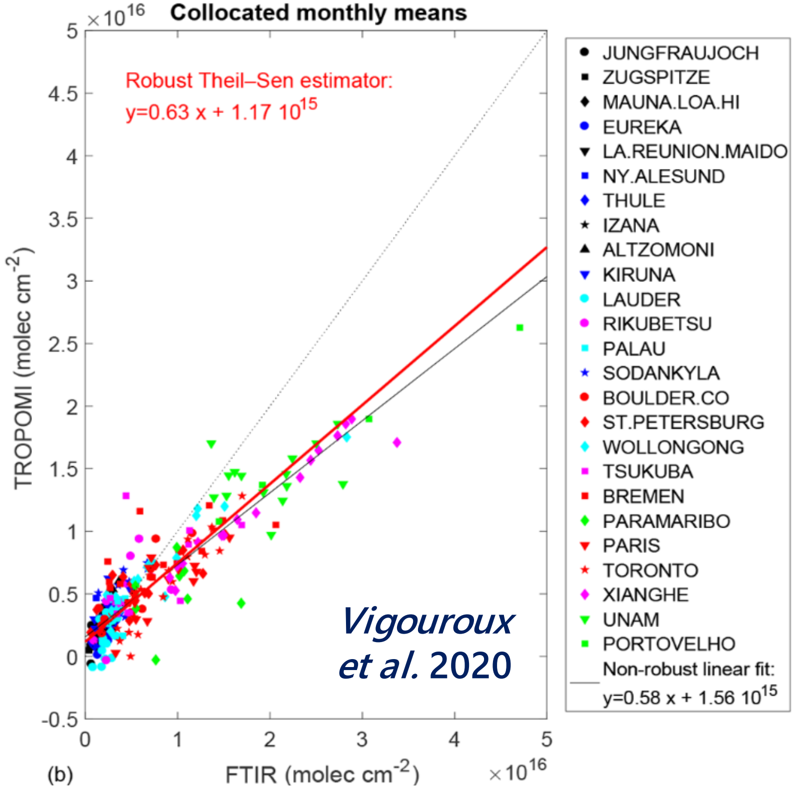
Marais et al. 2014

Isoprene flux update (%)



Bauwens et al. 2016

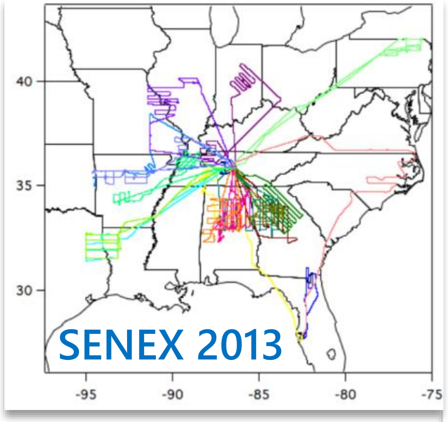
TROPOMI HCHO validation against FTIR data : TROPOMI is biased low

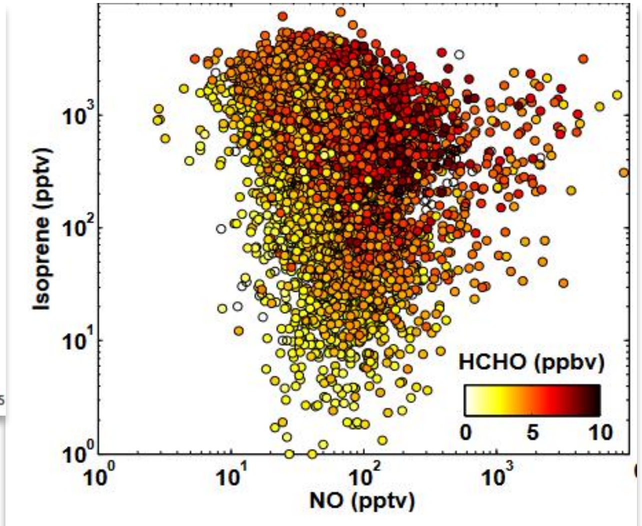


Vigouroux et al. 2020

$\Omega_{BC} = 1.59 \times \Omega - 1.86 \times 10^{15}$ molec. cm^{-2}

VOC and NOx linked through chemistry





Wolfe et al. 2016

- ✓ How do NOx levels affect top-down BVOC estimates over Africa?
- ✓ How do the optimized BVOC levels affect top-down NOx emissions?

- ○ Design an adjoint inversion framework based on the MAGRITTE CTM constrained by TROPOMI NO_2 and (bias-corrected) HCHO accounting for chemical interdependencies

- ✓ Change in radical chemistry with increasing NO
- ✓ HCHO most abundant when both isoprene and NOx are high

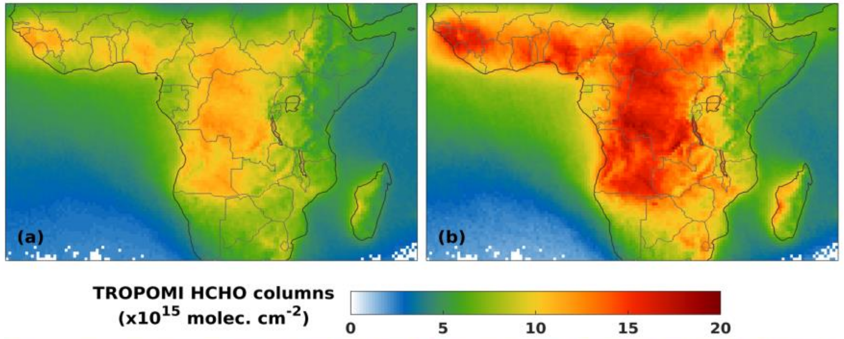
High [NO]:

- ✓ ISOPO2 + NO → ISOPO + NO_2
- ✓ ISOPO (+O_2) → MVK/MACR + HCHO + HO_2
- ✓ MVK/MACR + OH → … → *m* HCHO

Low [NO]:

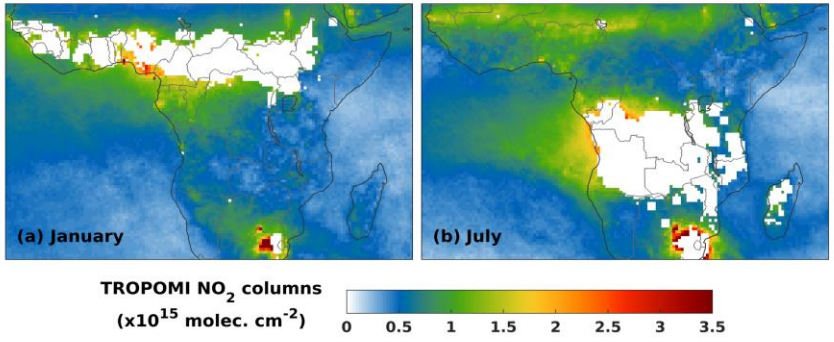
- ✓ ISOPO2 + HO_2 → ISOPOOH + O_2
- ✓ ISOPOOH + OH → … → *n* HCHO

w/o bias-correction



with bias-correction

NO_2 data filtered for fires

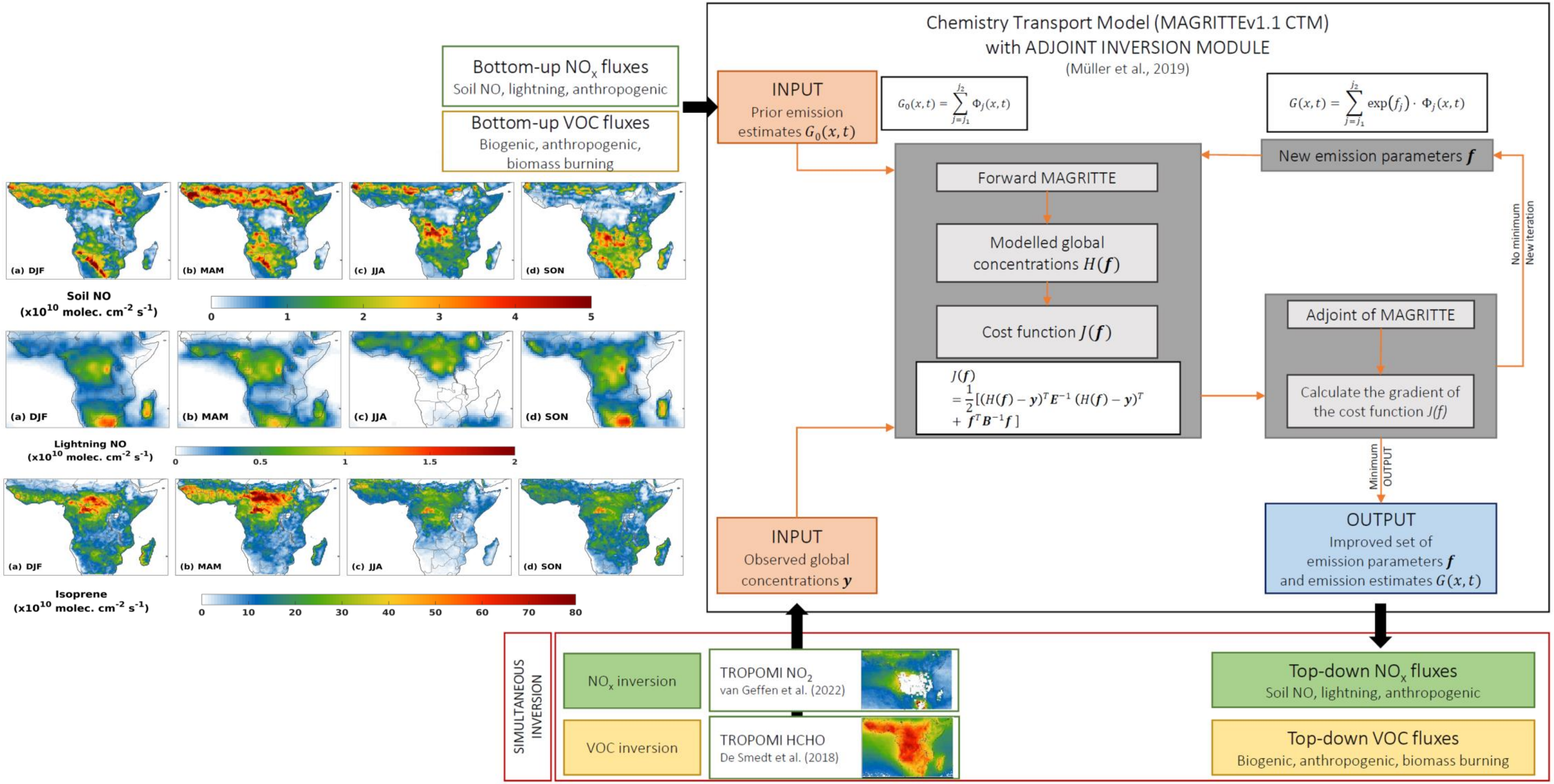


Ingredients of the adjoint model

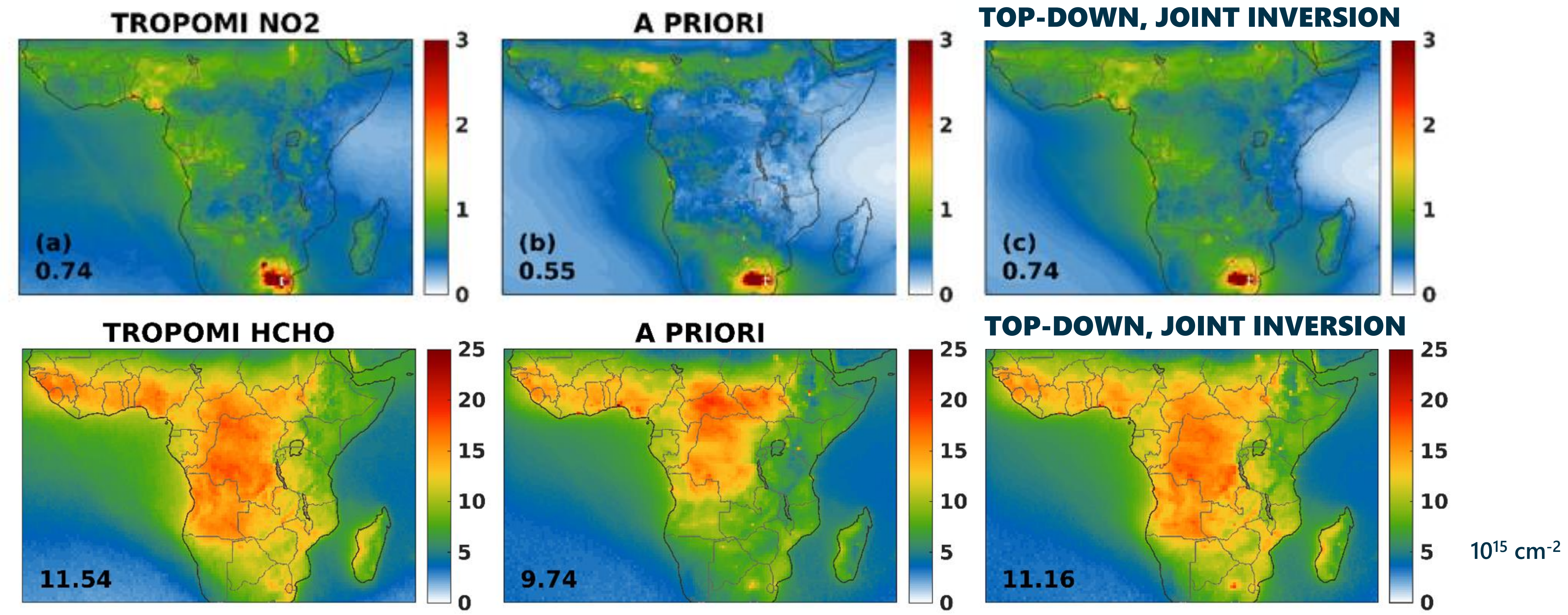
HEMCO (Weng et al. 2020)

Lightning NO based on LIS/OTD

MEGAN-MOHYCAN



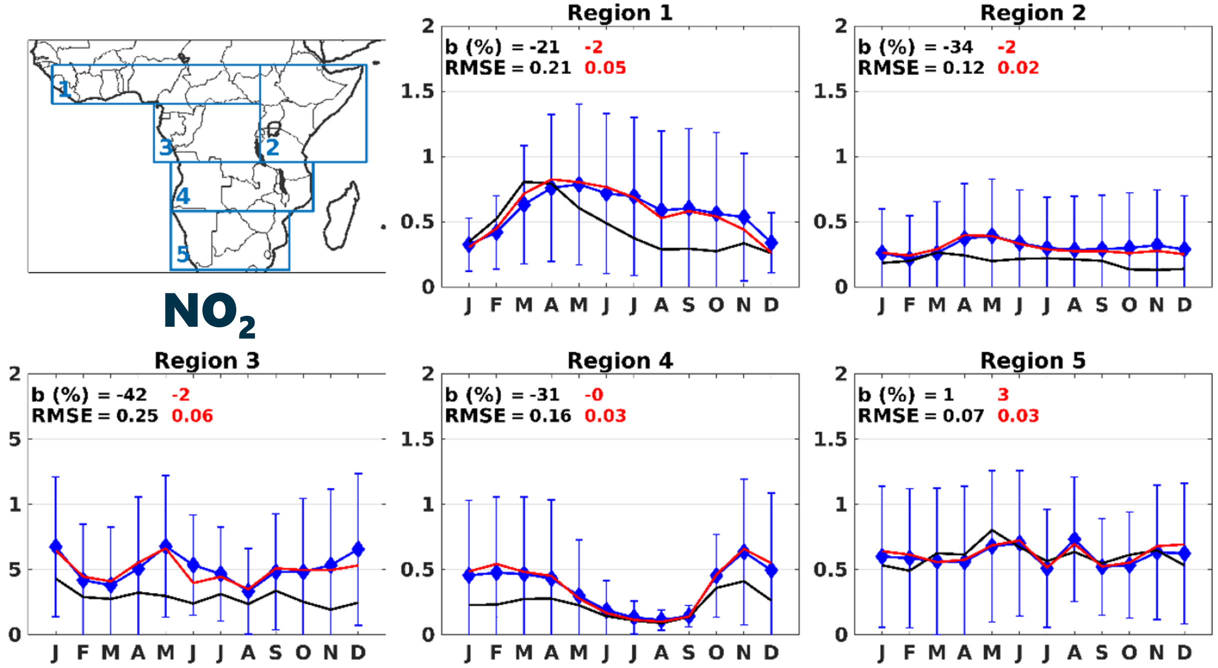
Annual results : observed vs. modelled columns



- ✓ Very good a posteriori agreement in terms of spatial distribution and magnitude
- ✓ NO_2: strong bias reduction from -26% to almost zero; HCHO: -12% to -2%
- ✓ Discrepancies still persist after inversion, e.g. Highveld plateau
- ✓ Although oceanic data are not used in the inversion, the bias over ocean is also reduced (from -32% to -16% for NO_2)

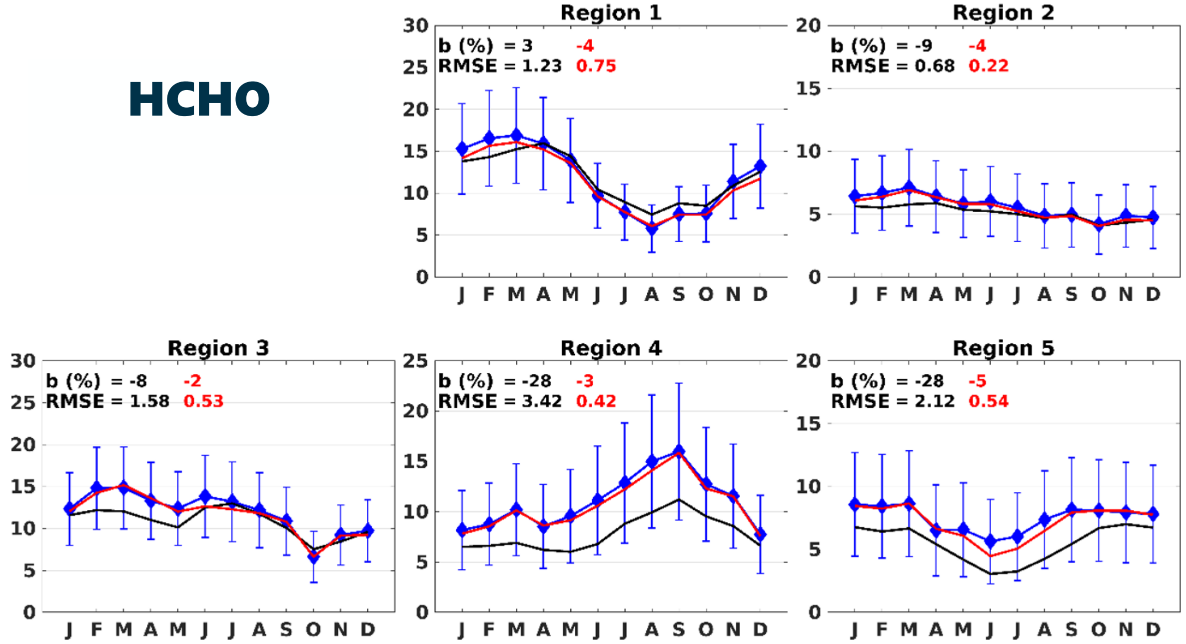
Seasonal variability of top-down columns

OBS A PRIORI TOP-DOWN



NO_2

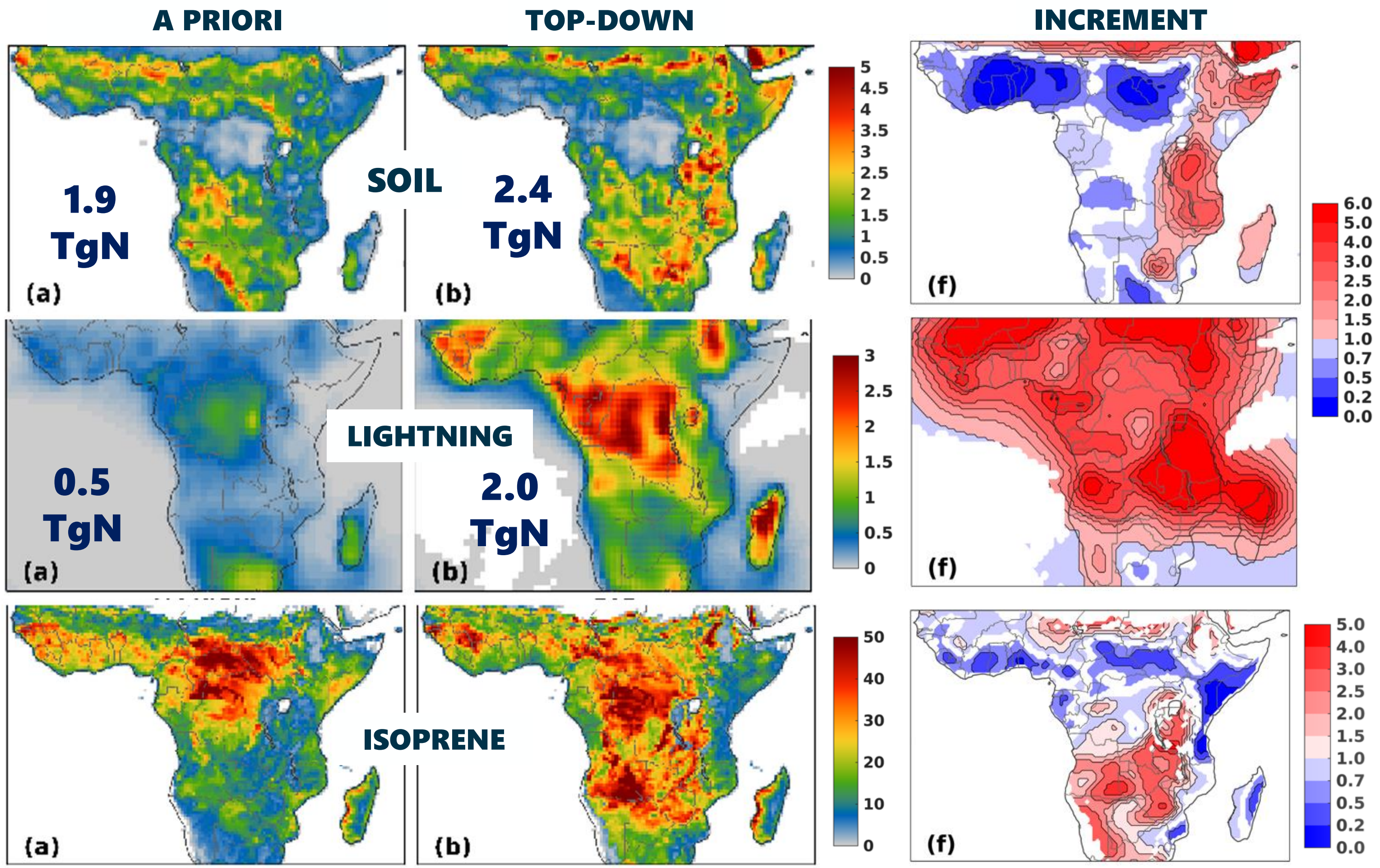
HCHO

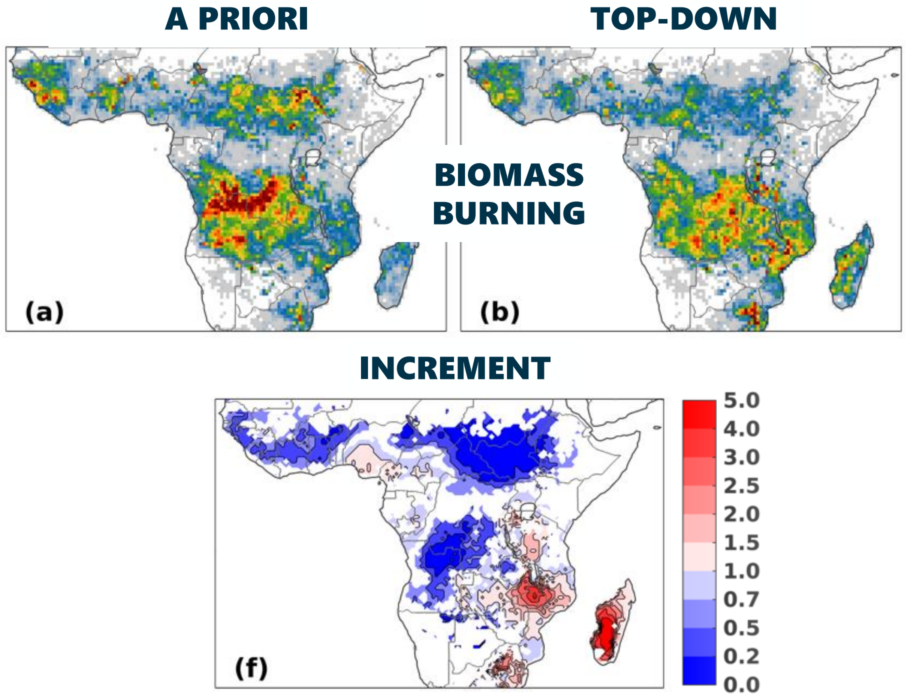


The a priori columns capture relatively well the observed seasonality and the agreement greatly improves after inversion

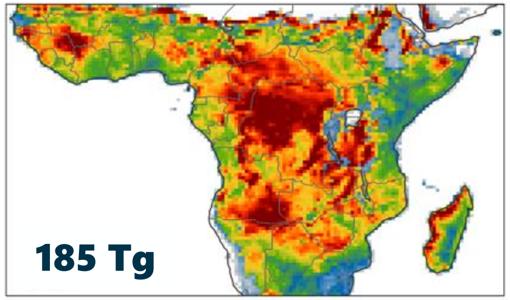
The large negative biases of a priori modelled HCHO columns in regions 4, 5 are strongly reduced after inversion due to the strong enhancement of isoprene fluxes

TROPOMI suggests increased NOx and isoprene emissions





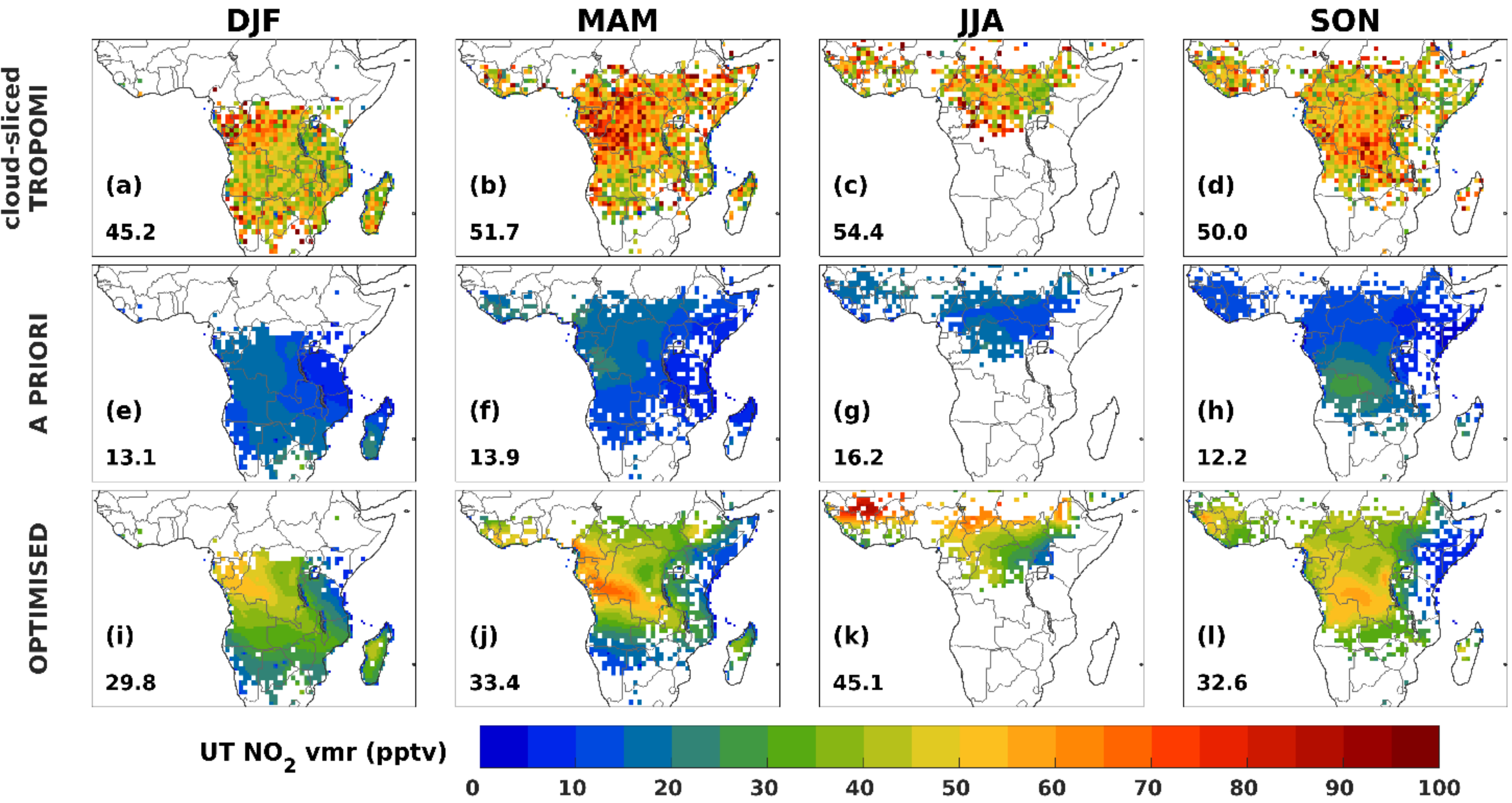
TOP-DOWN ISOPRENE, CONSTRAINED BY HCHO ONLY



Category	A PRIORI	TOP-DOWN
Soil, TgN/yr	1.9	2.4
Lightning, TgN/yr	0.5	2.0
Isoprene, Tg/yr	125	162*

**185 Tg when NOx is not used to constrain the inversion*

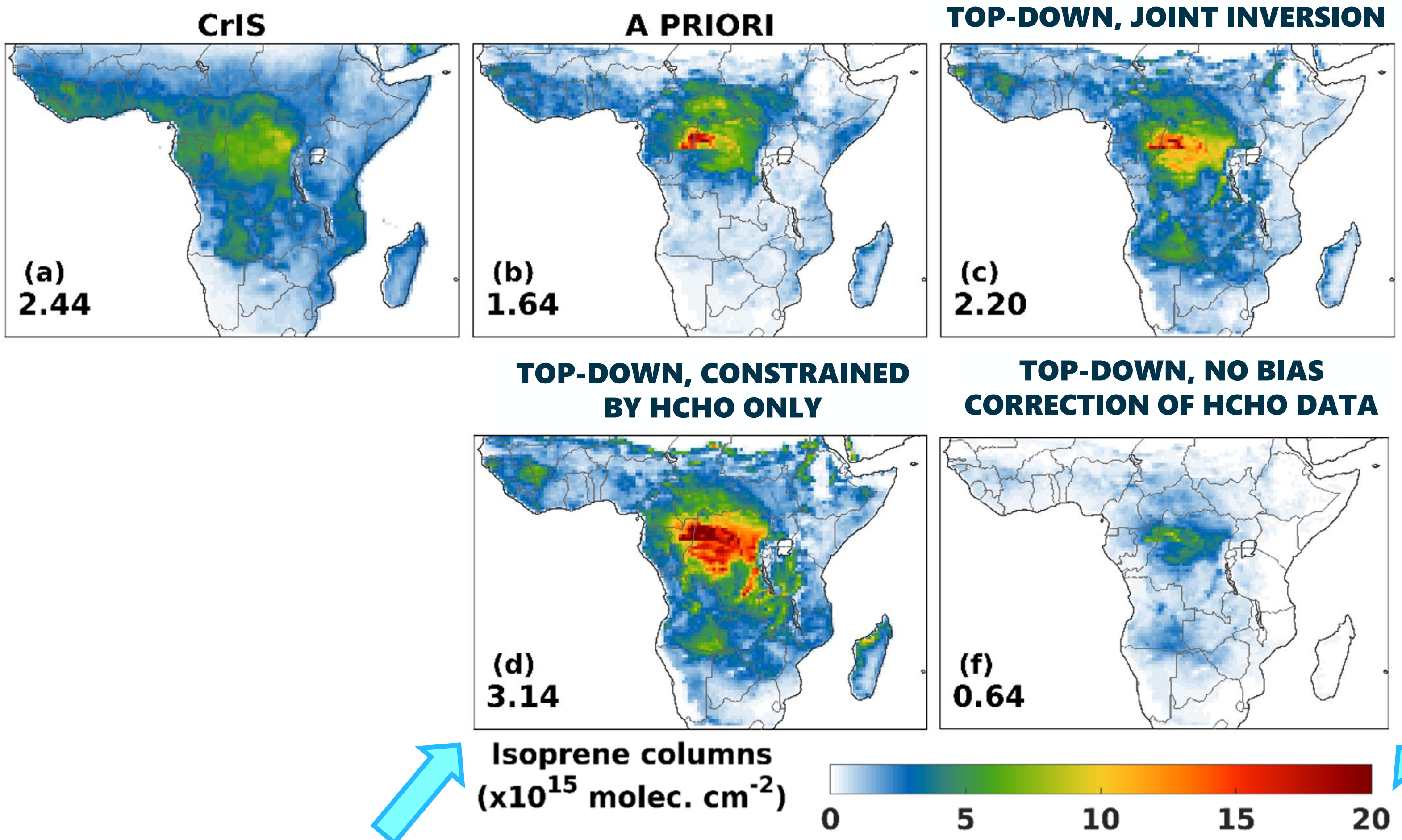
Evaluation of the top-down model : UT NO_2



- ✓ UT NO_2 vmr (180-450 hPa) from TROPOMI NO_2 for 2018-2022 (Horner et al. 2024; Marais et al. 2021)
- ✓ Observation: ~50 pptv, relatively constant across seasons; A priori model: 14 pptv; Optimized : 35 pptv
- ✓ Stronger W-E gradient in the model than in observations → underestimation of lightning NO in the vicinity of the Horn of Africa or long-range transport (longer NOx lifetime?)

UT NO_2 data supports the substantial increase of lightning NO suggested by TROPOMI.
An even larger source might be required to match UT NO_2 levels.

Evaluation of the top-down model : CrIS isoprene columns



- ✓ CrIS : highest over Congo, Angola and along coast of the Guinea Gulf & Indian Ocean south of Equator, low values over grassland and shrubs (Namibia, Horn of Africa)
- ✓ A priori spatial patterns are more contrasted than the observations
- ✓ Joint inversion leads to a much improved agreement with CrIS (-33% → -10%), improved spatial distribution
- ✓ TROPOMI HCHO suggests a southward displacement of the isoprene hotspot and an increase of the overall column levels

Compared to the joint inversion, increased columns by 40%. This is due to lower NOx fluxes, lower OH levels and longer isoprene lifetimes in HCHO-only inversion

Ignoring the bias correction results in very low isoprene, x4 lower than CrIS → strong evidence for the need to account for bias correction in the inversions.

Conclusions and outlook

- ✓ **Due to the chemical feedbacks, the simultaneous inversion of VOC and NOx emissions leads to an *improved top-down determination*, especially in Africa where NO_x fluxes are highly uncertain**

- ✓ **TROPOMI data suggest *substantial spatial changes* in emissions**

- ✓ **Top-down fluxes are much higher than bottom-up. Largest increase found for lightning emissions (x4), supported by UT NO_2 levels from cloud-sliced TROPOMI**

- ✓ **Tricky to discriminate between soil and lightning NOx flux → need for additional constraints, e.g. *in situ* INDAAF data**

- ✓ **Bias correction is crucial to the VOC optimization, leads to higher top-down emissions over source areas than in past studies, supported by CrIS isoprene**

- ✓ **Two-species inversion affects the inversion of isoprene fluxes due to chemical feedbacks**