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# Continuous monitoring of biogenic VOC fluxes over South America by inversion of TROPOMI HCHO, 2018-2021


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Belgian Science Policy Office
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EQUATOR
Emission QUantification of Atmospheric tracers in the Tropics using ObseRvations from satellites

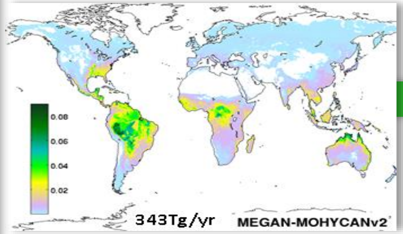
# Context and objective

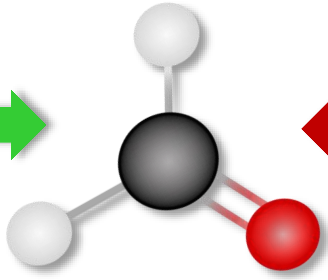


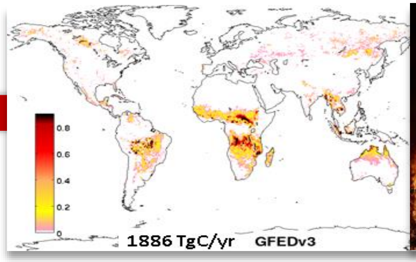
- South America hosts the Amazon forest, single largest source of *biogenic hydrocarbon (BVOC)* fluxes, also a region with extreme wildfires
- Data scarcity leads to large uncertainties in emission estimates and in their changes

✓ The photo-oxidation of most hydrocarbons leads to HCHO formation

✓ Satellite HCHO can inform us on the emitted hydrocarbons of biogenic and pyrogenic origin (Millet et al. 2008, Stavrakou et al. 2009, Barkley et al. 2013, Bauwens et al. 2016,…)





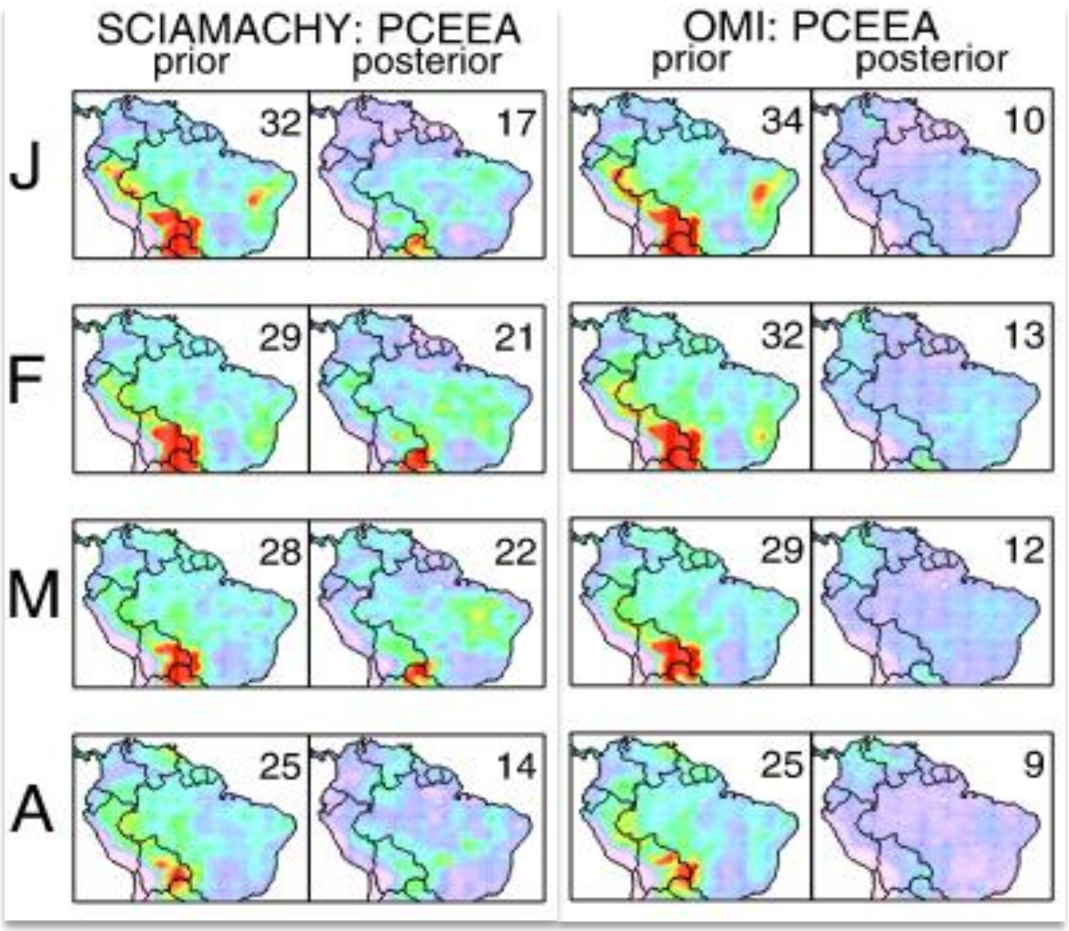

✓ Thanks to TROPOMI, HCHO is retrieved at 3.5×5.5 $km^2$ and high signal-to-noise

✓ Combined with CTMs enhanced with inverse modelling capabilities, these observations allow to infer <u>improved, space-based estimates of BVOC emissions</u>

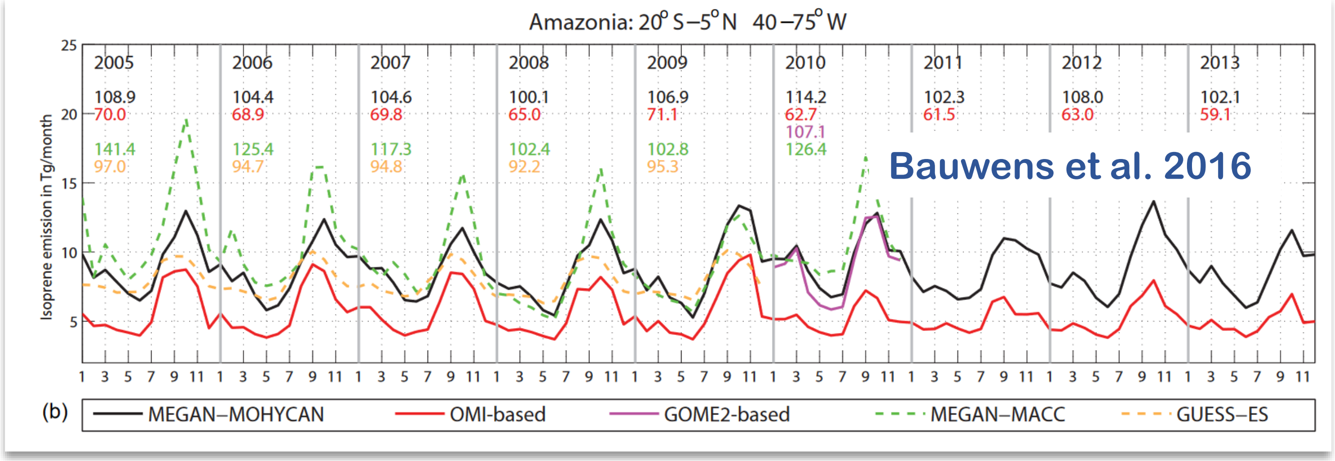
# Need for lower biogenic emissions ?



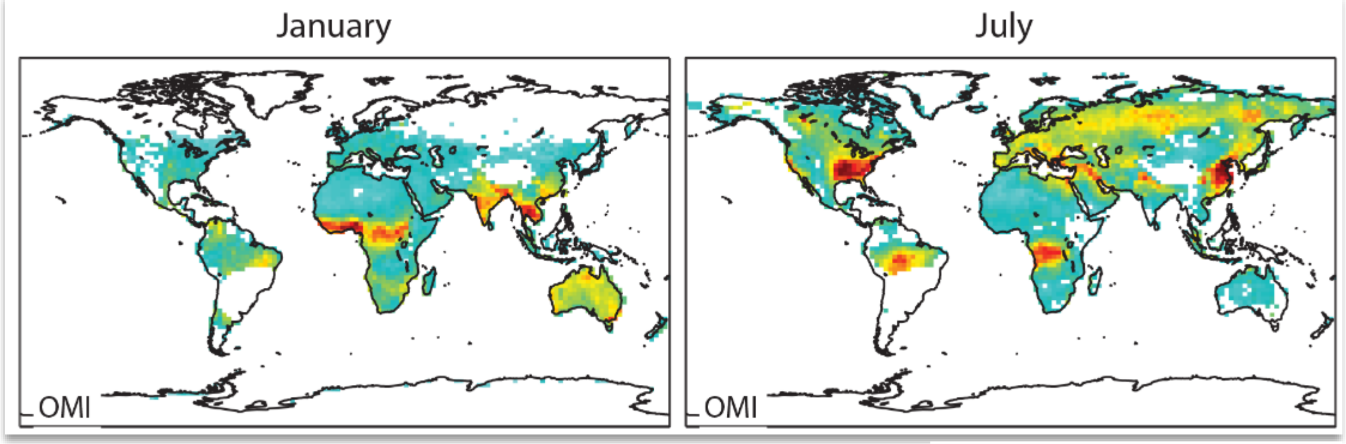
- ✓ A priori fluxes need to be scaled significantly down
- ✓ Large mismatch btw top-down SCIAMACHY & OMI
- ✓ SCIAMACHY badly affected by the SSA




Barkley et al. 2013




## Poor spatial coverage with previous sensors




Bauwens et al. 2016

# VOC and NOx linked through chemistry

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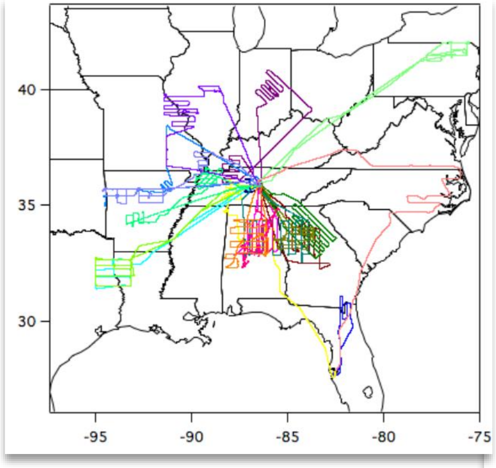
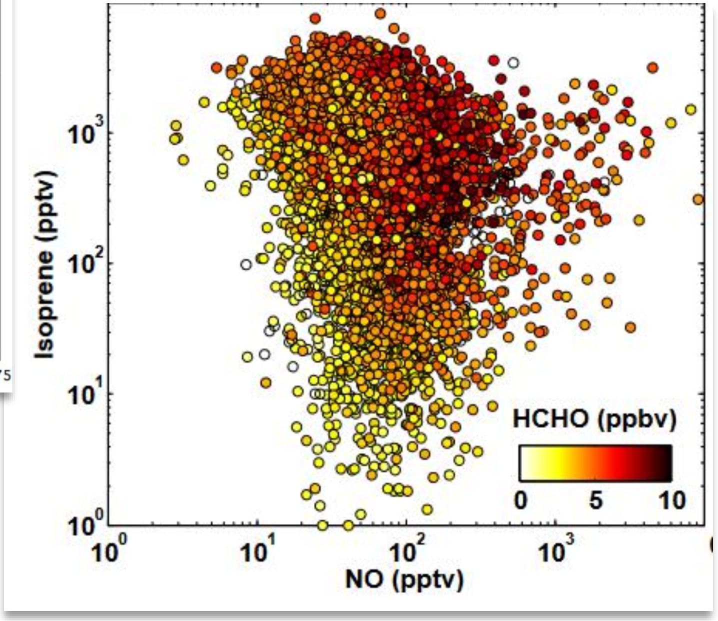
✓ Aircraft data indicate that the NOx levels control the HCHO photochemical production and loss rates




SENEX 2013 mission
Wolfe et al. 2016




✓ [NO]: 5-$10^4$ pptv → Big change in radical chemistry
✓ [HCHO] :0.8-14 ppbv, most abundant when both isoprene and NOx are high

At high [NO]:
✓ ISOPO2 + NO → ISOPO + $NO_2$
✓ ISOPO (+$O_2$) → MVK/MACR + HCHO + $HO_2$
✓ MVK/MACR + OH → … → *m* HCHO

At low [NO]:
✓ ISOPO2 + $HO_2$ → ISOPOOH + $O_2$
✓ ISOPOOH + OH → … → *n* HCHO

How do the NOx levels affect the top-down BVOC estimates over South America?
How do the optimized BVOC levels affect top-down NOx emissions?

Design an iterative inversion method which takes into account for the *NOx-VOC-OH feedbacks,* and uses TROPOMI HCHO and $NO_2$ columns as top-down constraints

# TROPOMI HCHO and $NO_2$ data

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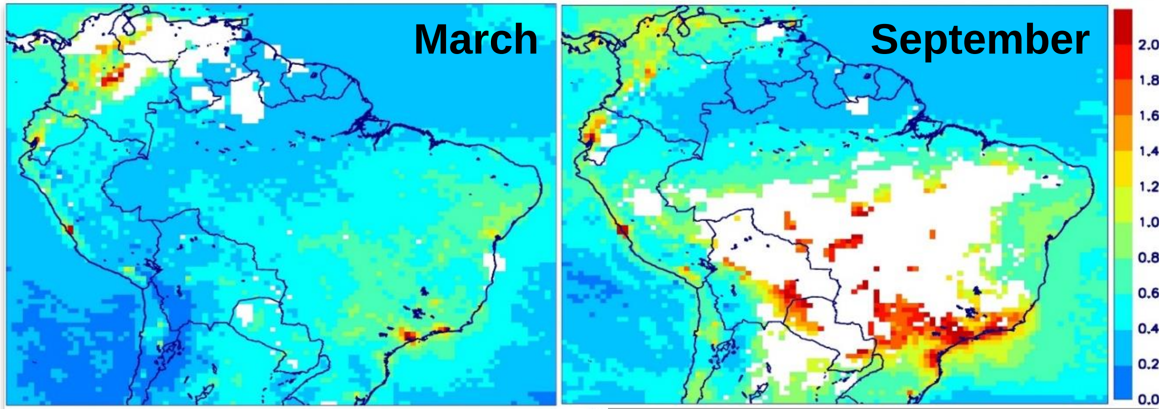
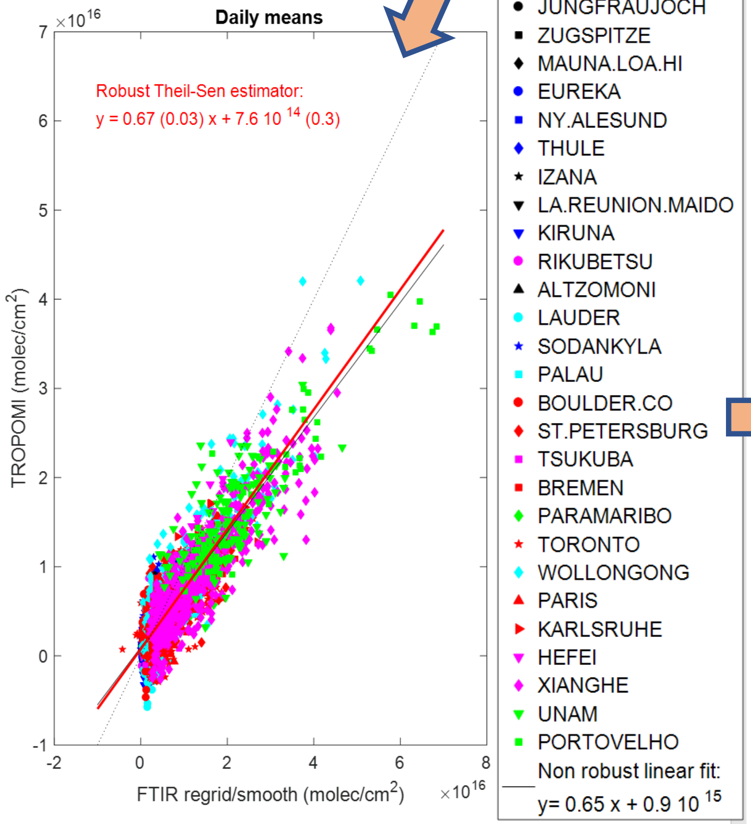
- Use TROPOMI PAL $NO_2$ (van Geffen et al. 2022) : higher tropospheric columns than previous versions & low bias wrt ground-based data
- Filter out $NO_2$ data contaminated by fires based on QFED2.4 and GFEDv4s inventories

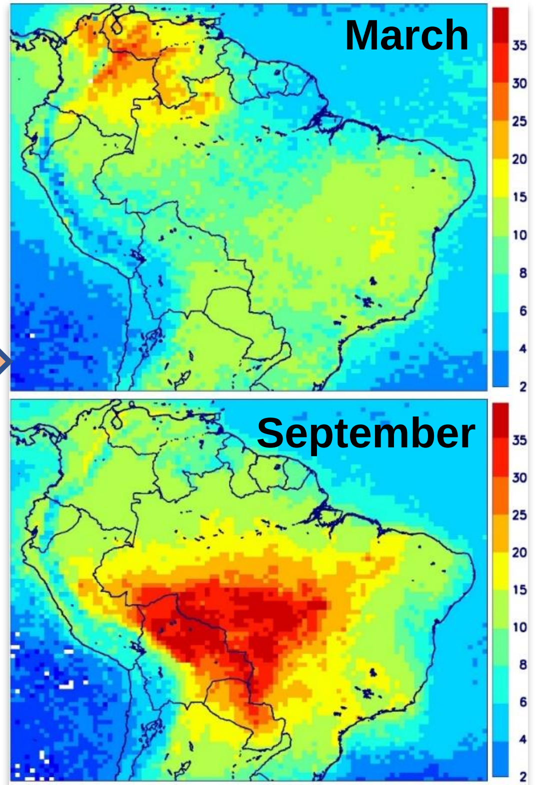
- Use TROPOMI HCHO cloud free (CF<20%), clear sky AMF, QF>0.5 (De Smedt et al. 2021)
- Bias-corrected TROPOMI HCHO (Vigouroux et al. 2020): $1.492 \times \text{TROPOMI} - 1.134\ 10^{15}\ cm^{-2}$

**TROPOMI $NO_2$ ($10^{15}\ cm^{-2}$) - 2019**




**TROPOMI HCHO - 2019**



# Seasonality of HCHO columns

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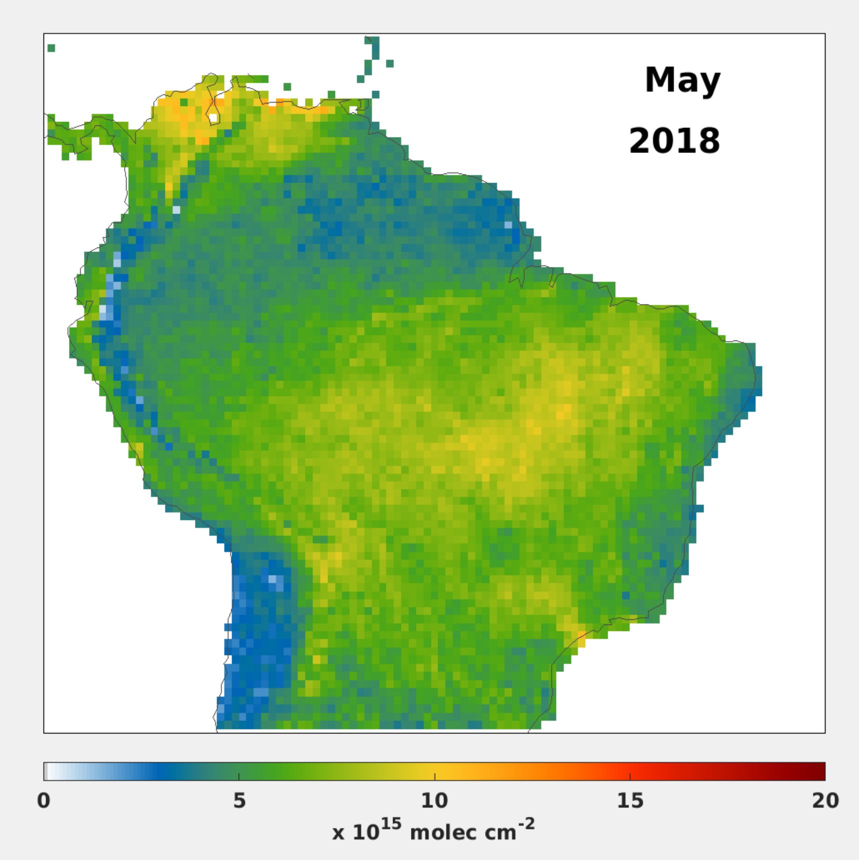
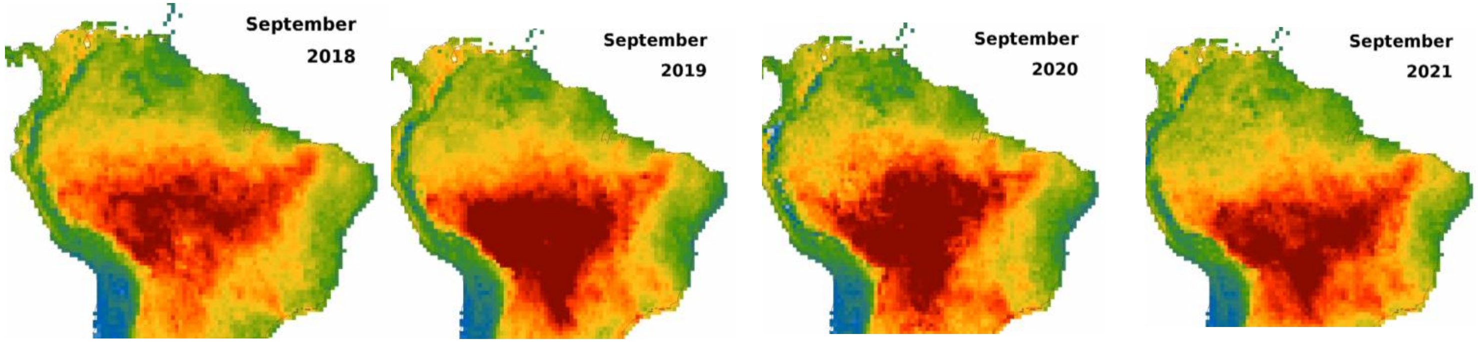



- ✓ Seasonality is driven by biogenic emissions and fire events: low columns in wet season (January-July), enhanced columns in dry season (August-December)
- ✓ Factor of 2-5 column increase between the dry and wet seasons, depending on the region
- ✓ Strongest changes over the tropical rainforest and savanna regions, more prone to fires




- ○ Fire inventories indicate year 2019 emissions are twice as high as in 2018
- ○ Emissions are even higher in 2020 (by 25% wrt 2019)

# The MAGRITTEv1.3 CTM

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Boundary conditions

A priori emission estimates → MAGRITTEv1.3 → Global atmospheric composition

**Biogenic emissions**
MEGAN-MOHYCAN
(Stavrakou et al. 2018, Opacka et al., 2021)

**Biomass burning**
QFED2.4
(Darmenov and da Silva 2015)

**Anthropogenic emissions**
CAMS-GLOB-ANT
(Granier et al. 2019)

## MAGRITTEv1.3

**ERA5 meteorology**
winds, T, $H_2O$, PBL mixing, convective fluxes, clouds, precipitation, downward radiation, etc.
(Hesbach et al. 2020)

**Chemical scheme**
- >170 compounds (45 NMVOCs)
- Inorganic and organic aerosols
- Revised isoprene mechanism (Müller et al. 2019)

Variable model time step : 1 day, 6h, 3h, 20-min (for diurnal cycle simulations)

**MODEL OUTPUT**
Global concentrations of >180 chemical constituents
at 2°x2.5° horizontal resolution and 40 vertical levels

**Zoom into South America** (15°N-35°S, 32-85°W), Spatial resolution : 0.5°x0.5°

# Adjoint-based inversion in 4 steps

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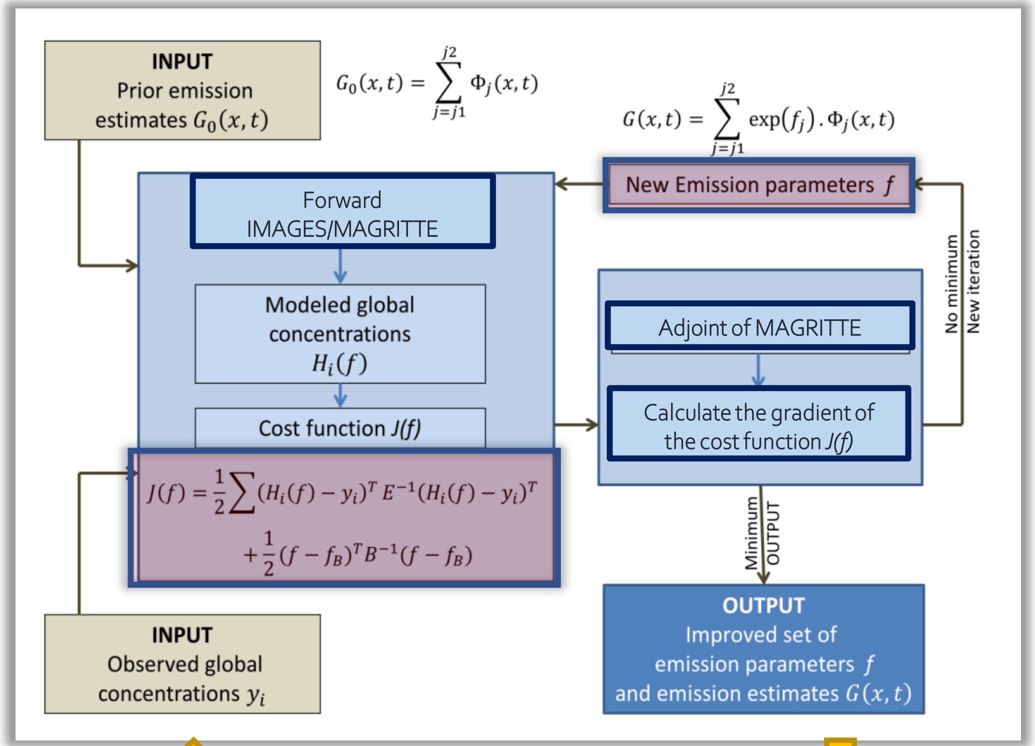
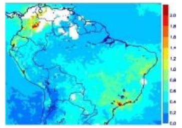



TROPOMI $NO_2$

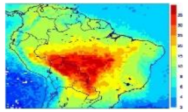


**$NO_x$ inversion**

***Top-down $NO_x$ fluxes***
*Soil NO, lightning, anthropogenic NOx*

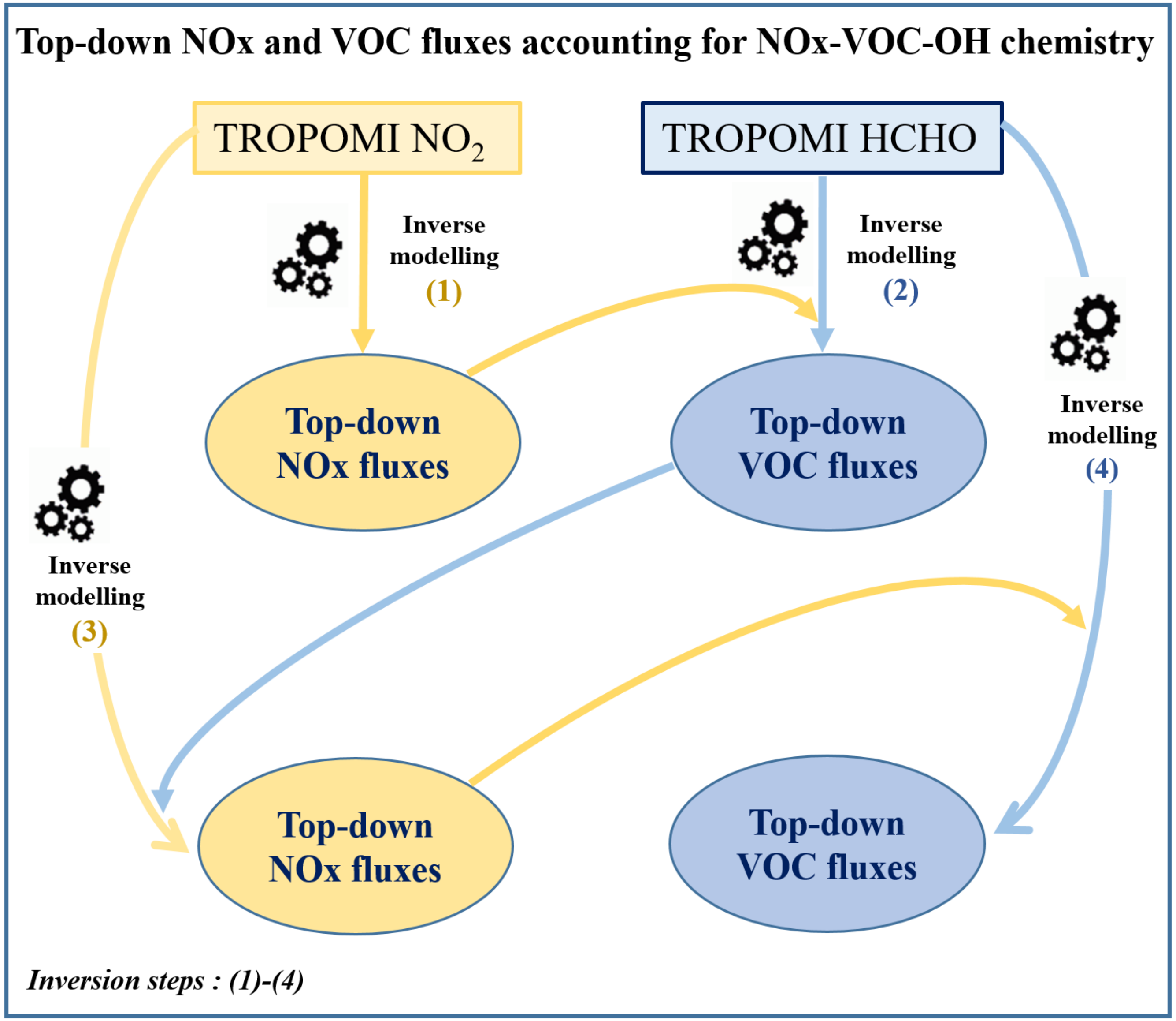
*or*

TROPOMI HCHO



**VOC inversion**

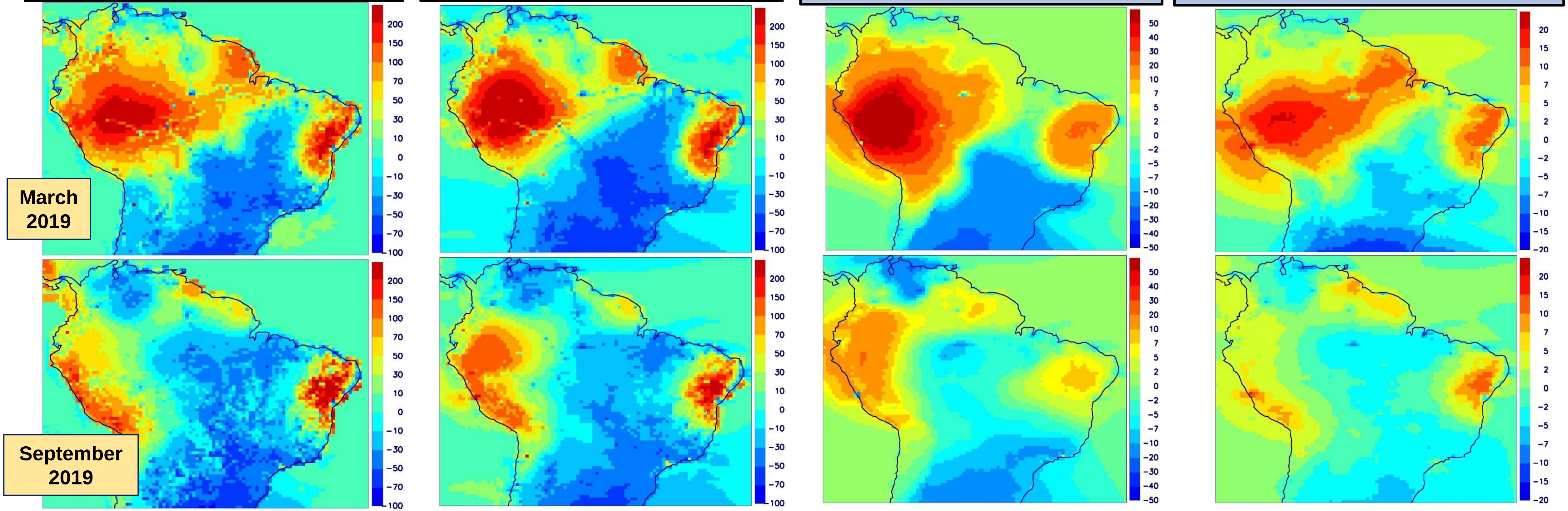
***Top-down VOC fluxes***
*Biogenic, anthropogenic, biomass burning*



# Impact of NOx inversion on HCHO



% Change in total NOx flux | % Change in surface $NO_2$ | % Change in surface $O_3$ | % Change in HCHO column




- ***Significant changes in emission patterns***, esp. in wet season
- ***Increase of soil NO fluxes*** (by 40% over Amazonia)

- Similar but ***slightly stronger*** changes in surface $NO_2$, primarily due to the $O_3$ increase (higher $NO_2$/NO due to NO+$O_3$ reaction)

- $O_3$ levels also ***increase by up to 50%*** in March (ca. 7 ppb), due to higher $NO_x$ levels

- High NOx → ***increased oxidation*** of $CH_4$ and other VOCs into HCHO
- ***Enhanced*** $Y_{HCHO}$ from isoprene due to higher NO

# Enhanced natural NO fluxes
# Decreased biogenic flux in Amazonia

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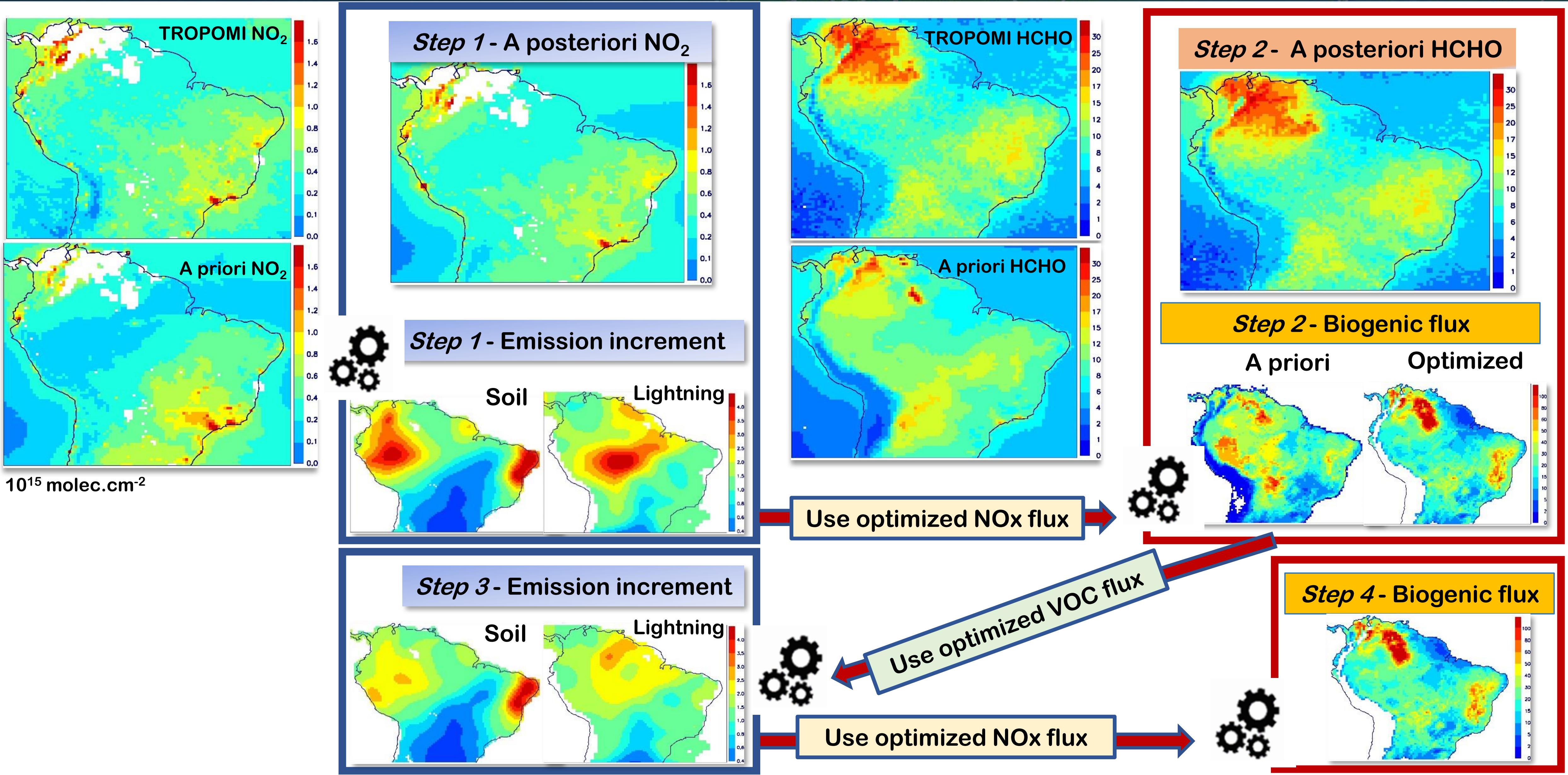
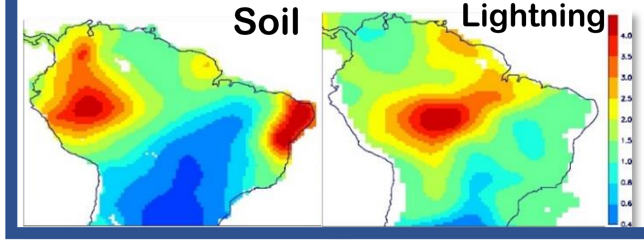


# VOC-NOx chemistry interplay



**Step 1 - Emission increment**

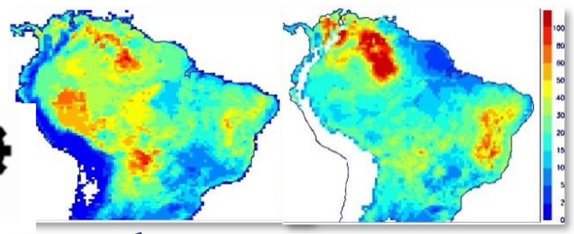
Soil Lightning



Use optimized NOx flux

**Step 2 - Biogenic flux**

A priori Optimized

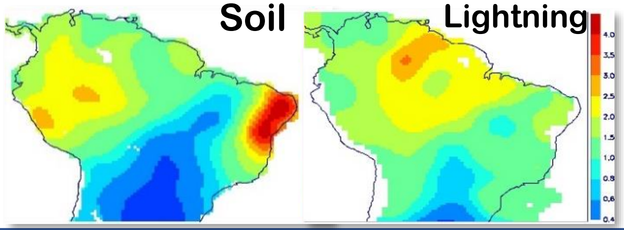


The VOC flux decrease over the Amazon decreases the formation of organic nitrates ($RONO_2$) from VOC + NO reactions

Use optimized VOC flux

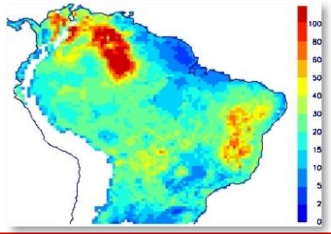
**Step 3 - Emission increment**

Soil Lightning



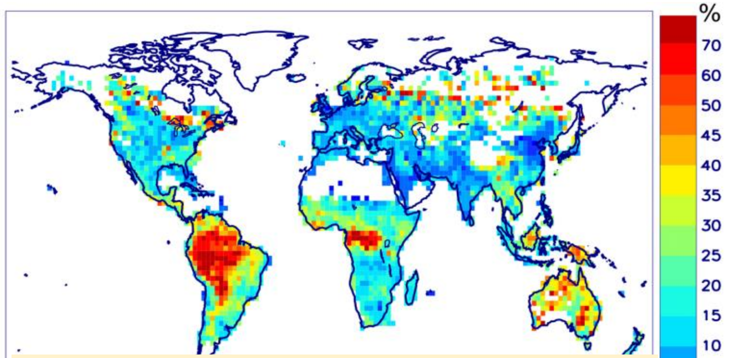
Use optimized NOx flux

**Step 4 - Biogenic flux**



The decreased $RONO_2$ formation in the Amazon leads to increased $NO_2$ columns. Therefore, smaller increase in NOx flux is inferred in *Step 3* compared to *Step 1*.

Lower NOx flux in *Step 3* results in lower HCHO production yield in isoprene oxidation → lower HCHO columns → higher BVOC emission in *Step 4* compared to *Step 2* is needed to match TROPOMI




Annual NOx loss due to $RONO_2$ formation (Müller et al. 2019)

# Summary of top-down estimates

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| $NO_x$ fluxes (Tg N) | A priori | *Step 1* | *Step 3* |
|---|---|---|---|
| Soil | 1.67 | 2.14 | 2.04 |
| Lightning | 1.14 | 1.34 | 1.25 |
| Total | 2.81 | 3.48 (+24%) | 3.29 (+17%) |
| Amazonia | | | |
| Soil | 0.88 | 1.24 (+40%) | 1.10 (+25%) |
| Lightning | 0.57 | 0.75 (+31%) | 0.68 (+20%) |

| Biogenic VOC fluxes (Tg) | A priori | *Step 2* | *Step 4* |
|---|---|---|---|
| | 142 | 132 (-7%) | 134 (-6%) |
| | Amazonia | | |
| | 93 | 73 (-22%) | 80 (-14%) |

The satellite data suggest

- Increased natural NOx fluxes over Amazonia; Strongly enhanced fluxes in Nordeste in Brazil
- Slight decrease of BVOC fluxes over Amazonia

✓ The use of bias-corrected TROPOMI HCHO data results in top-down estimates close to the prior (on average)

# Evaluation against Porto Velho data

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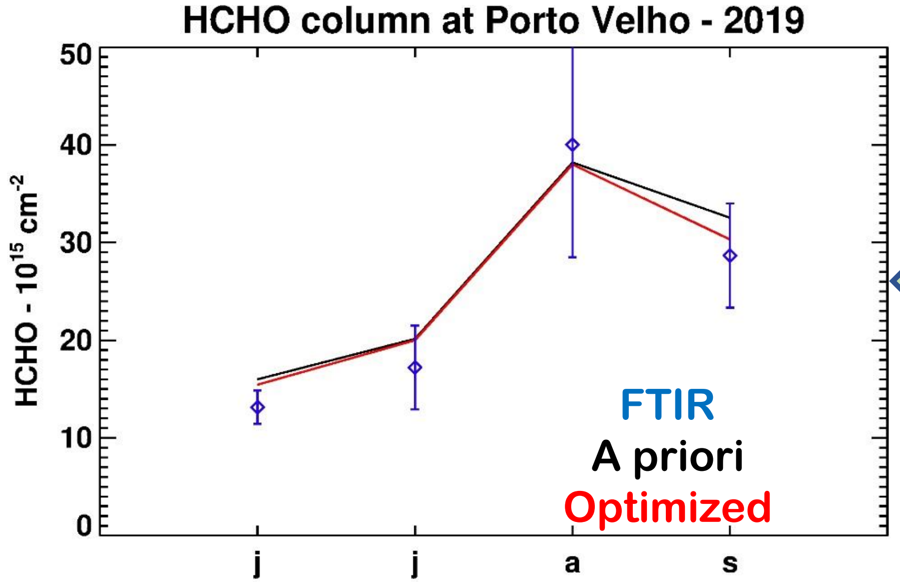
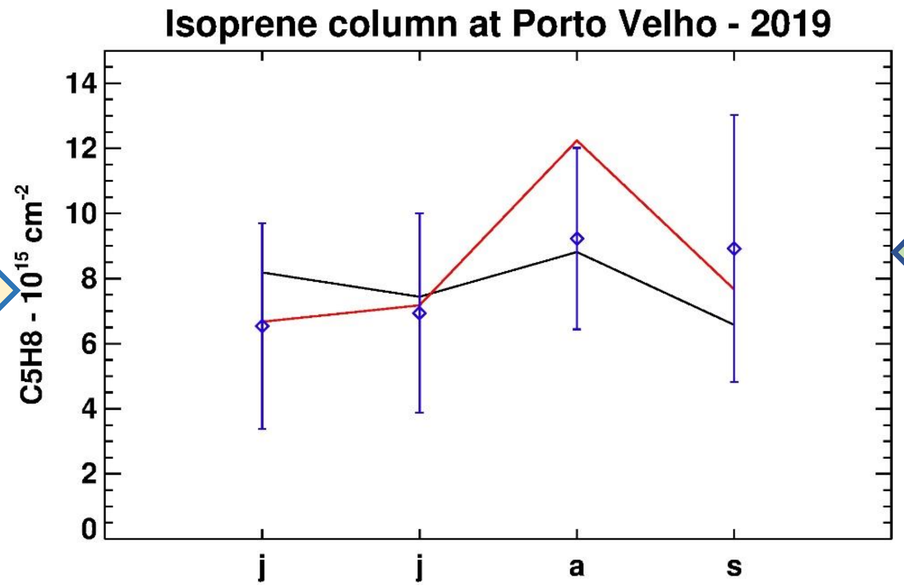
- FTIR HCHO and isoprene column measurements at Porto Velho (8.77°S, 63.87°W), on the border between Rondônia and Amazonas Brazilian states
- June-September 2019




Excellent agreement for HCHO in the a priori model, little change inferred from space

First retrievals of isoprene from ground-based FTIR spectra (Wells et al., 2022)




Both a priori and optimized columns offer a fairly good agreement with the observations.

# Take-home message

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- ✓ Due to the VOC-NOx chemistry, combining satellite $NO_x$ and HCHO columns leads to an *improved top-down determination of NOx and VOC fluxes*, especially in tropical regions where $NO_x$ fluxes bear large uncertainties

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

- ✓ Factor of 2 higher top-down natural $NO_x$ flux over Amazon, Northern South America and eastern Brazil - *cf. poster of Beata Opacka et al.*
- ✓ Strong reduction in biogenic fluxes over western Amazonia, and increase to the north of the Amazon river, in line with preliminary comparisons with CrIS isoprene

- ✓ Fairly good comparison against ground-based HCHO and isoprene column data at Porto Velho

  - Extend to more years to study the *interannual variability* of top-down sources
  - Evaluate against more data (e.g. aircraft missions)
  - Design similar setups and study other tropical regions