Oxygenated volatile organic compounds (OVOCs) first detected from IASI : understanding the sources

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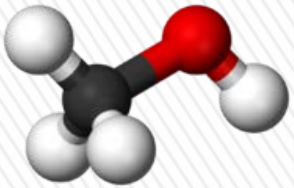
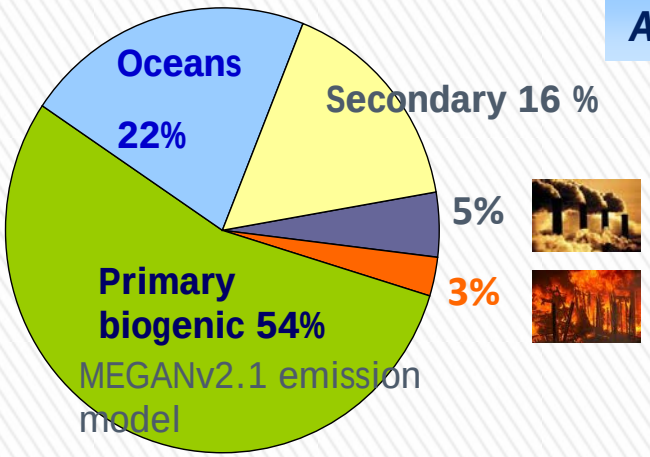
CH_3OH

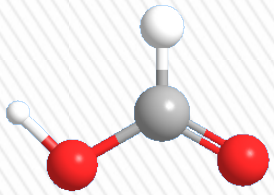


- Abundant over forests in the growing season, impact on OH concentrations in the PBL



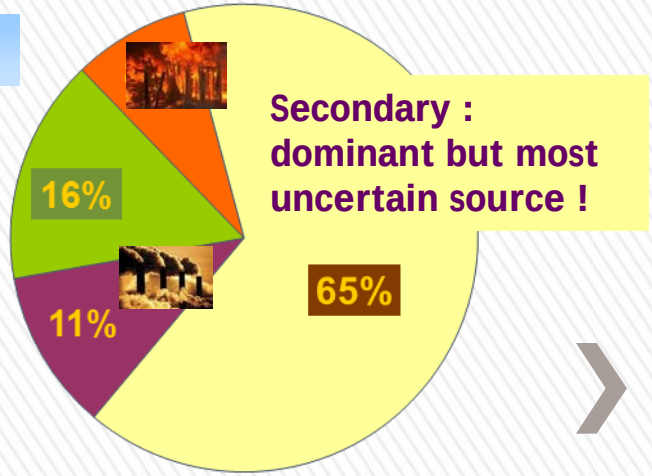
Global source = 193 Tg/yr , T = 6 days

HCOOH

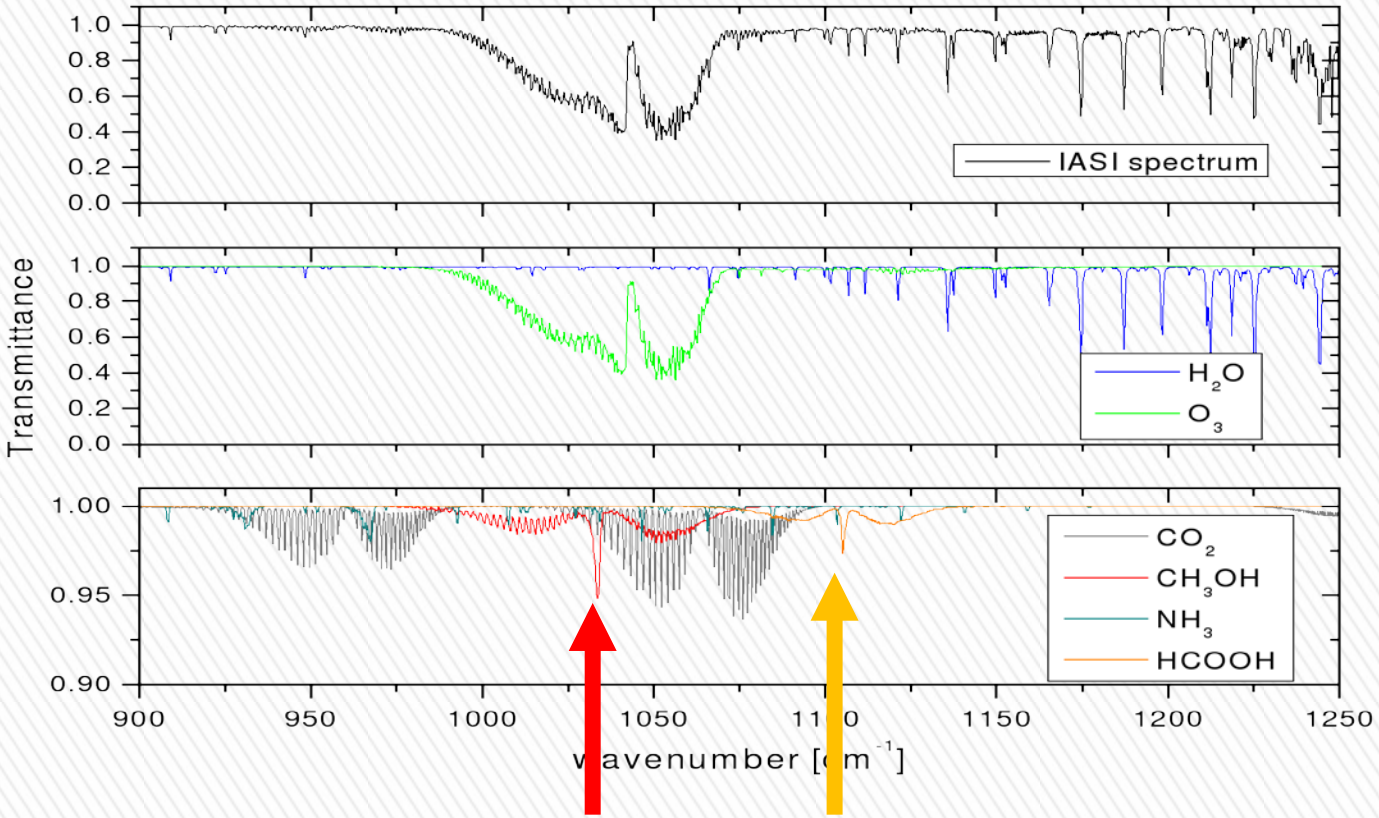


- Major contributor to acidic rain in remote environments, but models underpredict severely the observed HCOOH concentrations → *missing sources*

***A priori* global budget**



Global source = 36 Tg/yr, T = 4 days

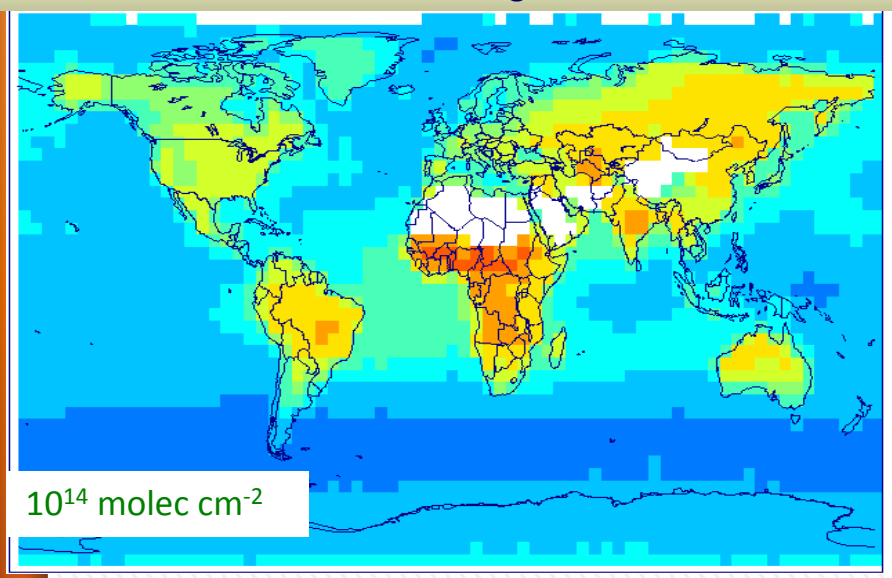


CH_3OH @ 1033 cm^{-1} HCOOH @ 1105 cm^{-1}

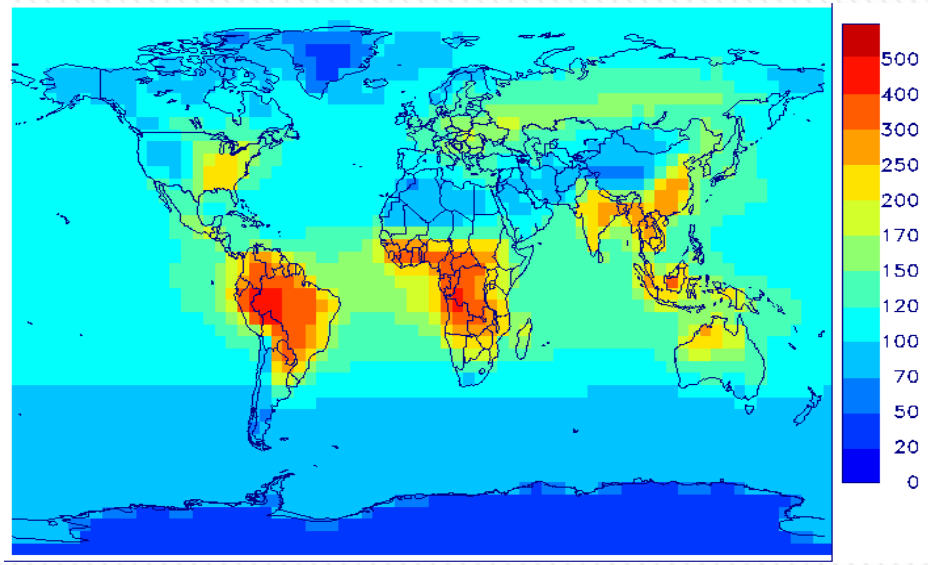
- Very weak absorption interfered by other molecules, especially ozone
- Retrieval based on ΔT_b between the target channel and nearby reference channels in the spectrum baseline

Razavi et al., ACP, 2011

2009 IASI CH_3OH column



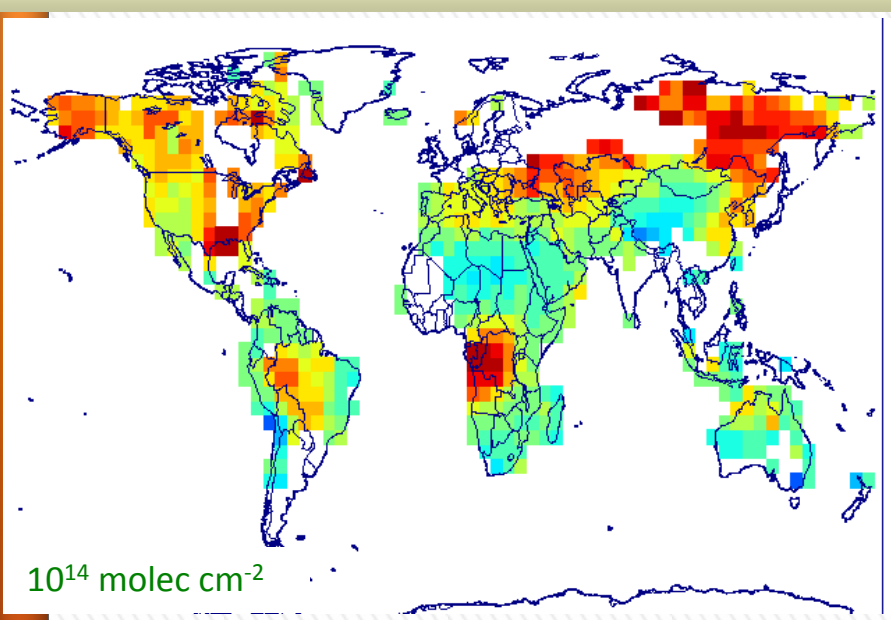
IMAGESv2 CH_3OH column



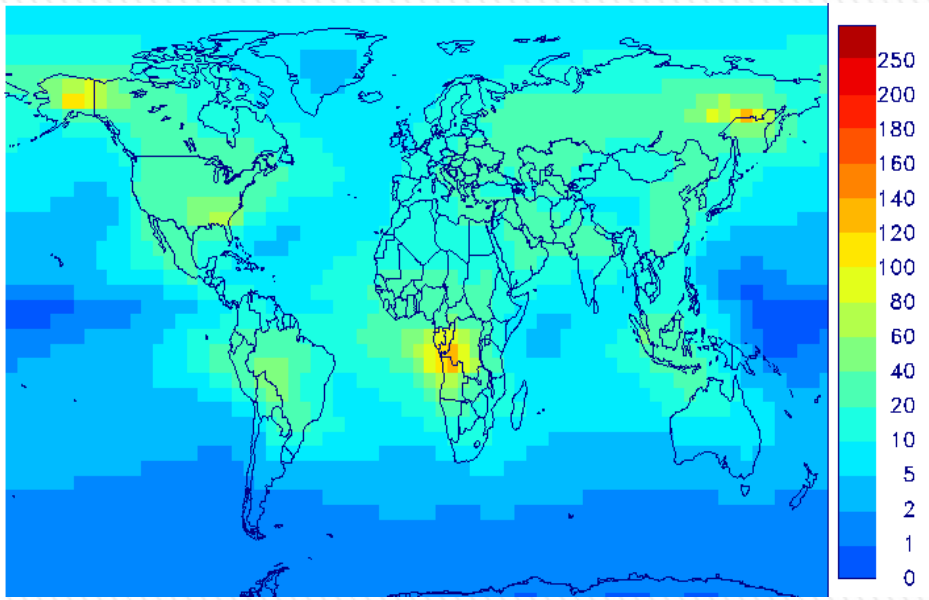
High columns over the majority of continental regions

Large overestimations over Amazonia (factor of 2-3), Africa &Indonesia (1.5-2), moderate over Europe, Eastern US

IASI HCOOH column - July



IMAGESv2 HCOOH column



Strongly enhanced columns over central Africa, S. America, and boreal regions

Important underestimations (factor of 2-3)

Source inversion using the adjoint model

A priori emission distributions

$$G_0(x,t) = \sum_{j=1}^{m} \Phi_j(x,t)$$

Optimized emission distributions

$$G(x,t,f_j) = \sum_{j=1}^{m} \exp(f_j)\Phi_j(x,t)$$

Observation

Emission parameters f

Forward CTM
Transport
Chemistry
Cost function J(f)

Adjoint CTM
Adjoint transport
Adjoint chemistry
Adjoint J(f)

$$J(f) = \frac{1}{2}\left((H(f)-y)^T E^{-1}(H(f)-y) + (f-f_B)^T B^{-1}(f-f_B)\right)$$

Gradient of J(f)
Calculation of new f's
Is J(f) minimum ?

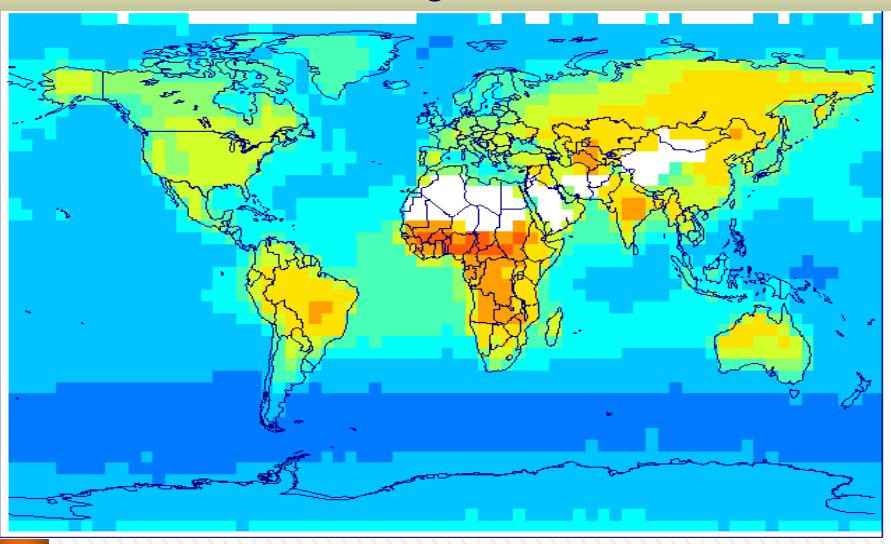
no!

yes!

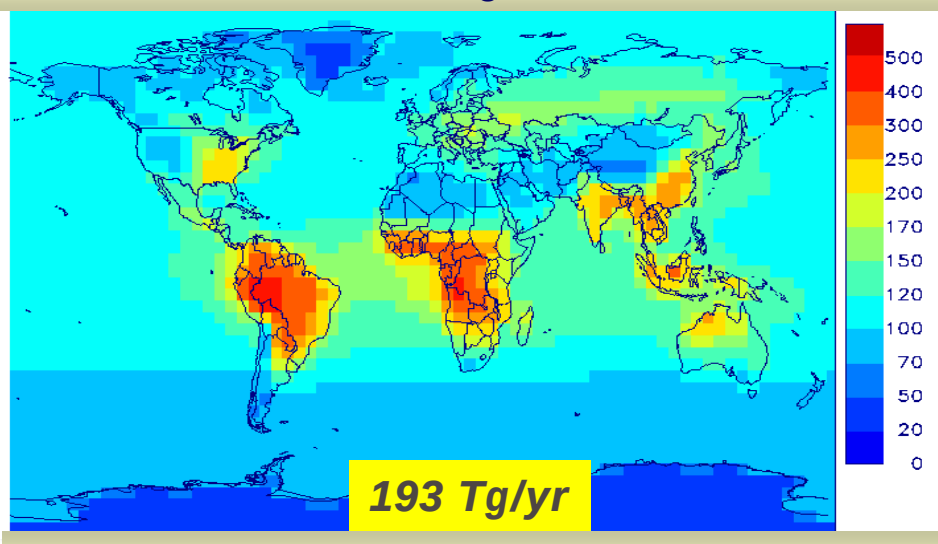
Updated emission parameters

Handle very large numbers of control variables
Address non-linearities
Distinguish between emission categories

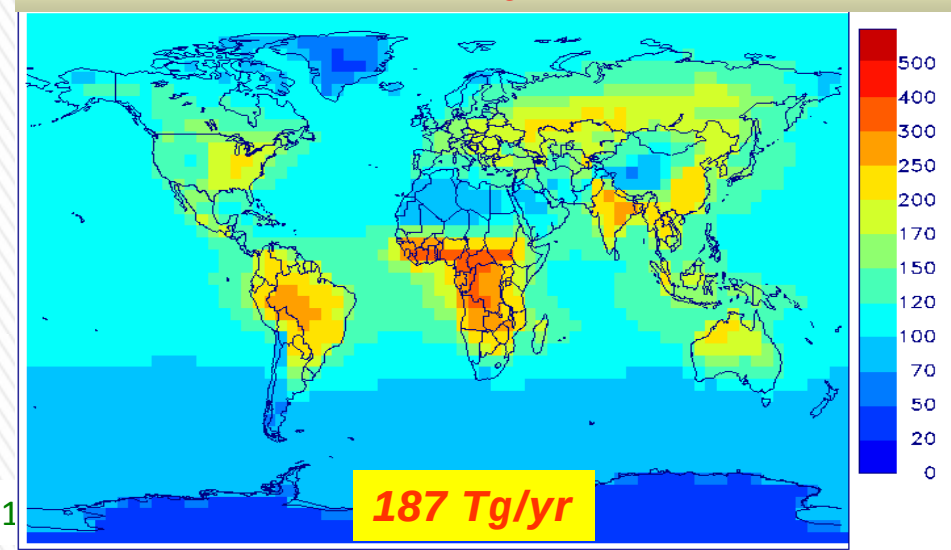
IASI CH_3OH column



IMAGESv2 CH_3OH column



Optimized CH_3OH column



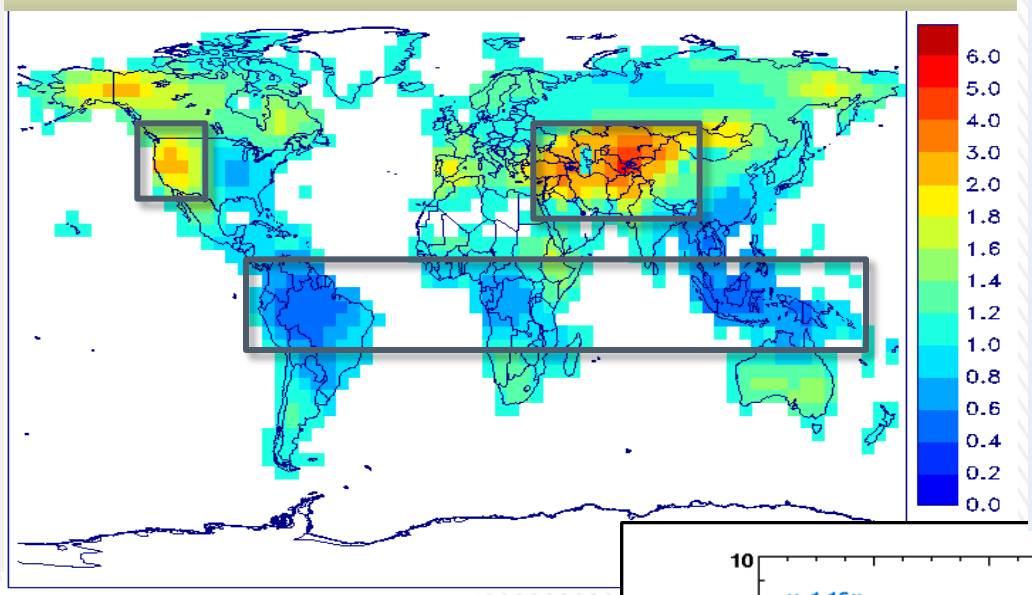
- Global *a posteriori* biogenic CH_3OH source very close to the *a priori* MEGANv2.1
- BUT…

Stavrakou et al. ACP, 2011

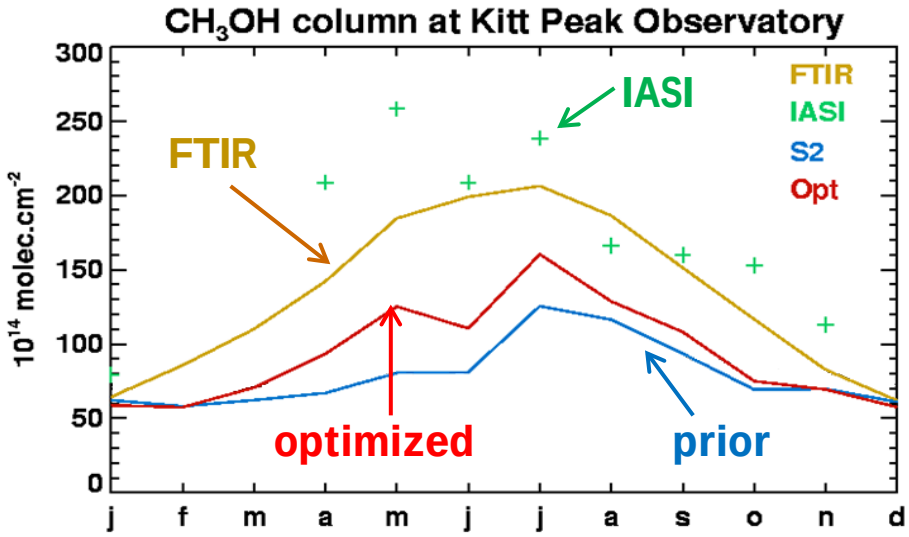
Strong flux increases in arid/semi-arid regions in central Asia (x5), western US (x2)

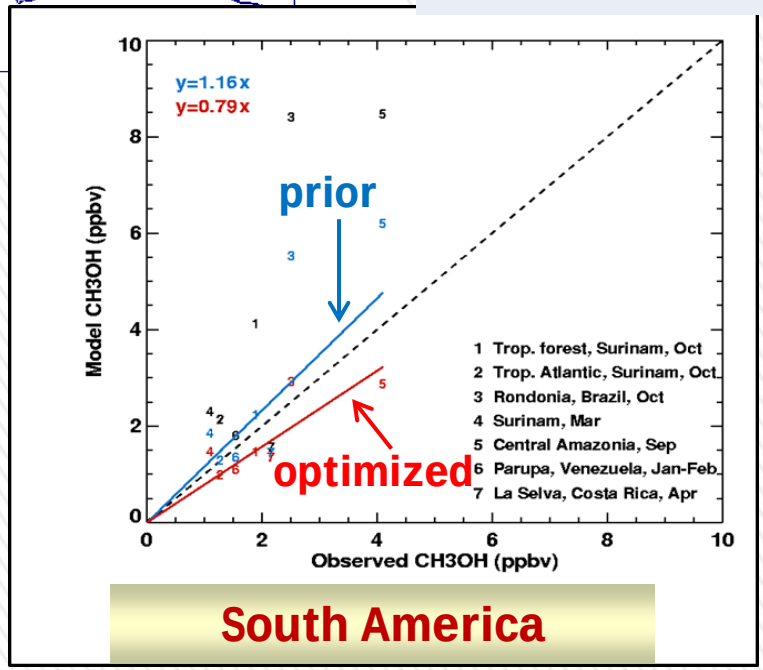
→ underestimated methanol rate from shrub or an unaccounted methanol source in MEGANv2.1 ?

Biogenic emission update



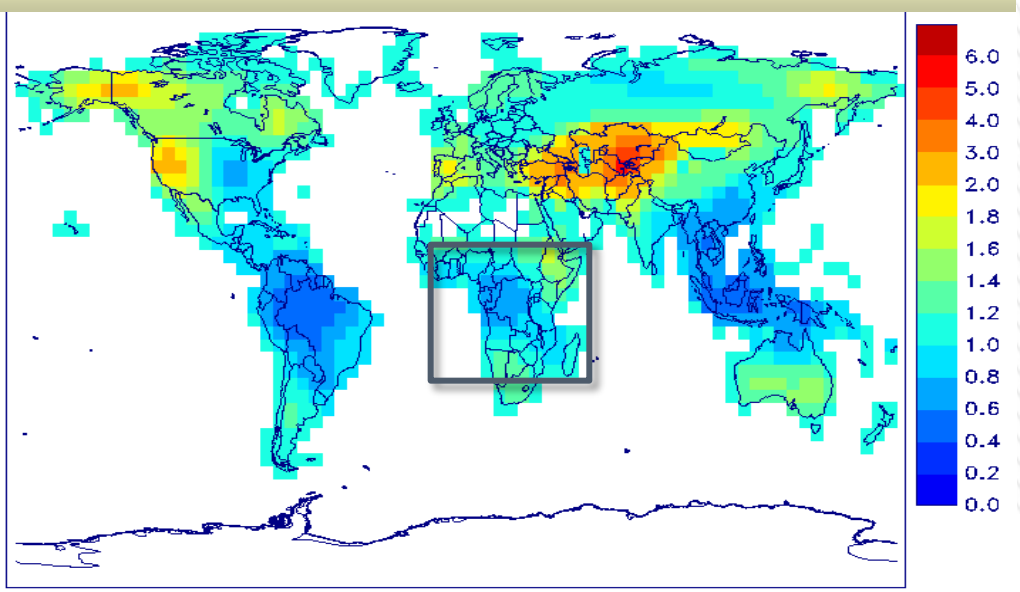
Significant reductions over the tropical forests in the Amazon (40-55%) and Indonesia (45-60%)





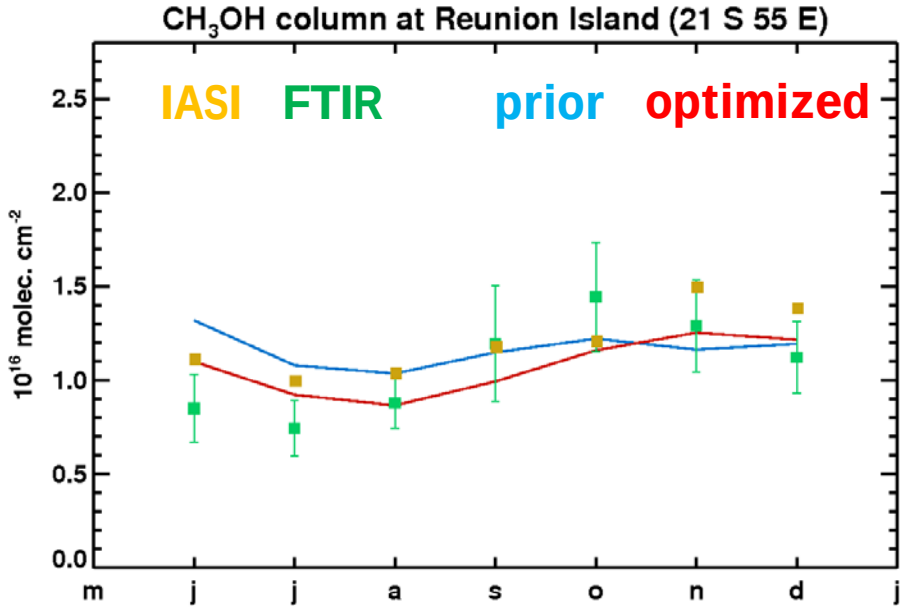
South America

Biogenic emission update



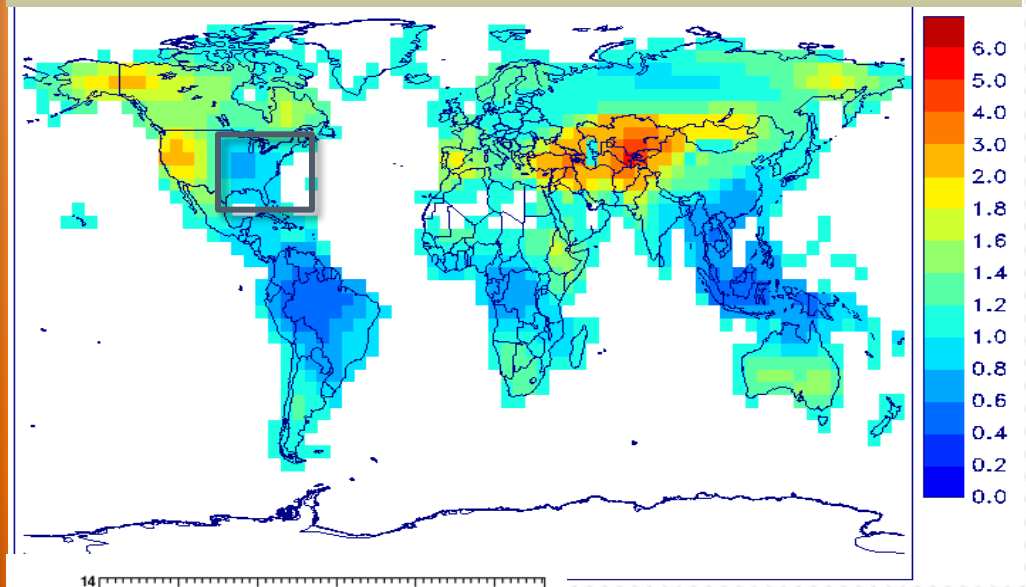
✓ **FTIR supports IASI-derived decrease in biogenic emissions over Central & S. Africa**

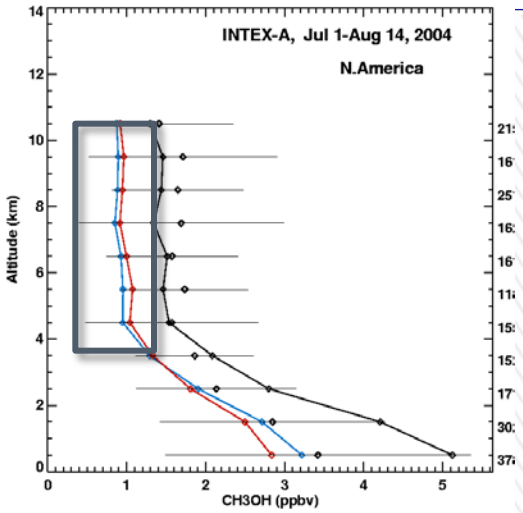
✓ **Observed seasonality well reproduced by the model**



Stavrakou et al. ACP, 2011
Vigouroux et al. ACP, 2012

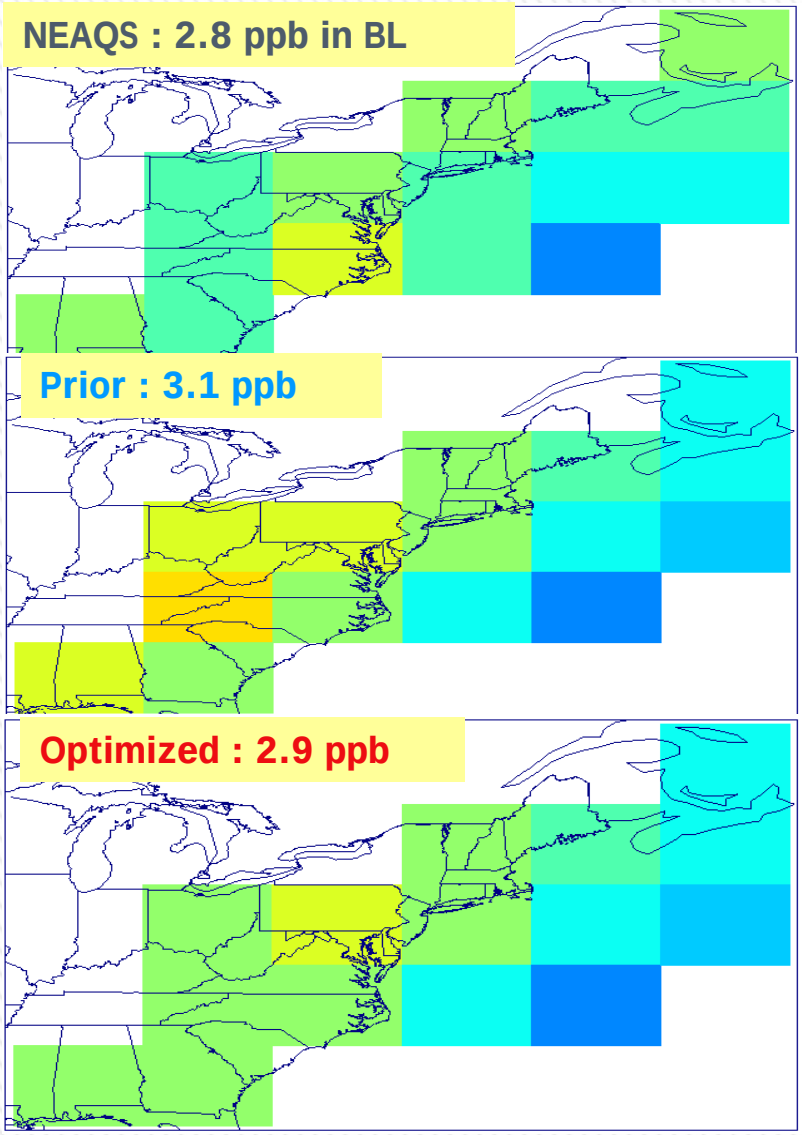
Biogenic emission update





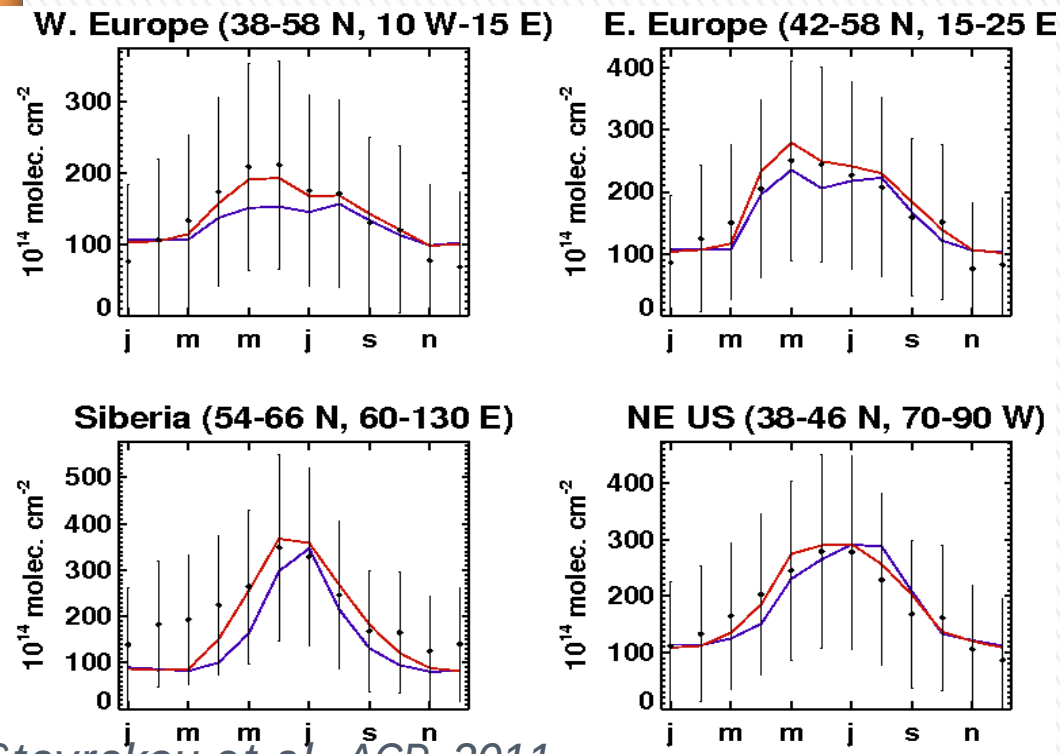
- **Underestimation in the free troposphere : related to model underestimation in the Western US ?**

INTEX-A (Jul 1-Aug 14 N. America)

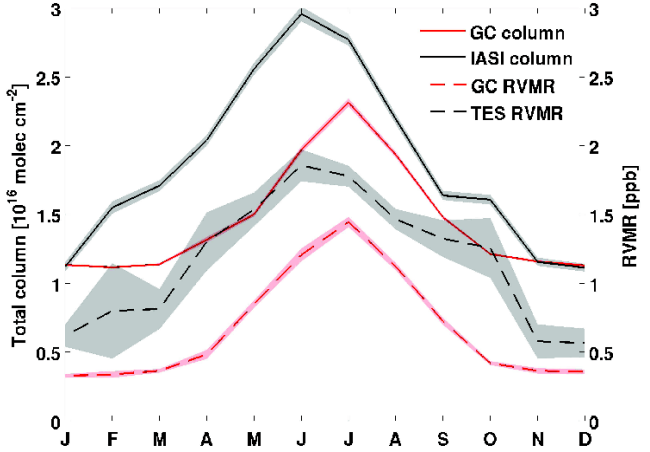


- **Very good agreement with aircraft data in PBL**

Prior **Optimized**



Stavrakou et al. ACP, 2011



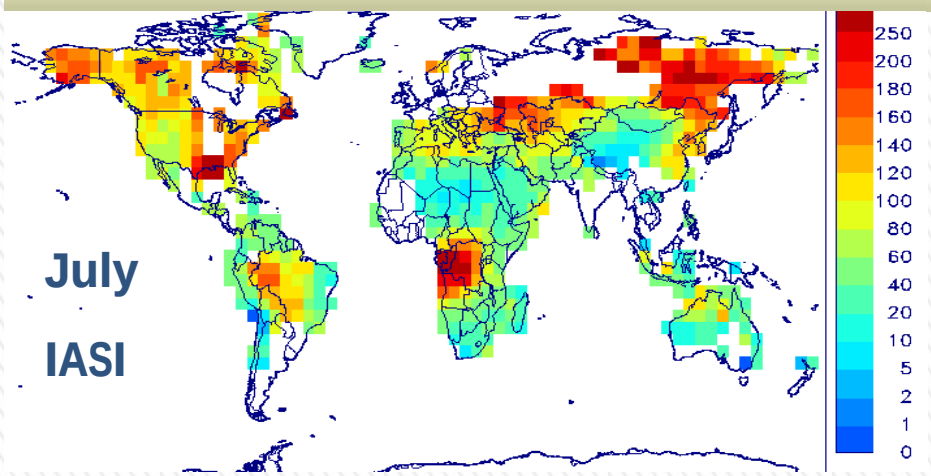
4. Seasonal cycle in atmospheric methanol over midlati- for 2009. Shown are methanol column amounts simulated EOS-Chem (base-case simulation, red solid line) and mea- by IASI (black solid line), and representative volume mixing (RVMR) simulated by GEOS-Chem (base-case simulation,

Wells et al. ACP, 2012

- ✓ **In mid-latitudes IASI is maximum in spring and early summer, i.e. 1-2 months earlier than the model peak, similarly for CH_3OH from TES (Wells et al., 2012) and tower measurements in the US Midwest (Hu et al., 2012)**
- ✓ **Emissions from new leaves are underestimated in MEGAN, and emissions from mature leaves should be decreased → important changes in leaf age classes for broadleaf trees**

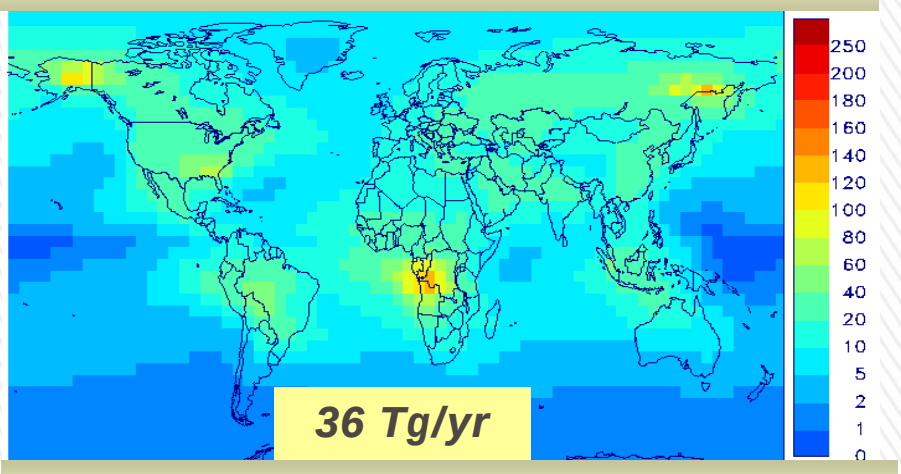
- ***Optimize the global* HCOOH *source:*** assume that missing HCOOH is produced through OH-oxidation of as-yet-unidentified biogenic precursors

IASI HCOOH column

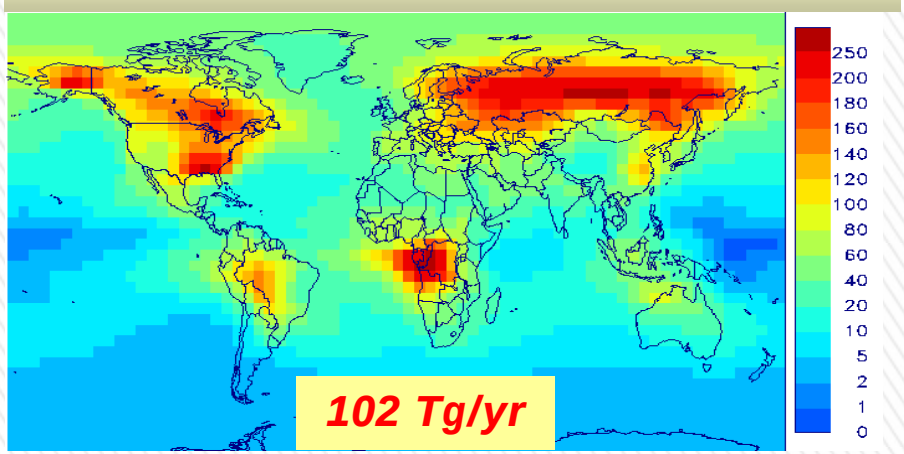


10^{14} molec cm^{-2}

IMAGESv2 HCOOH column

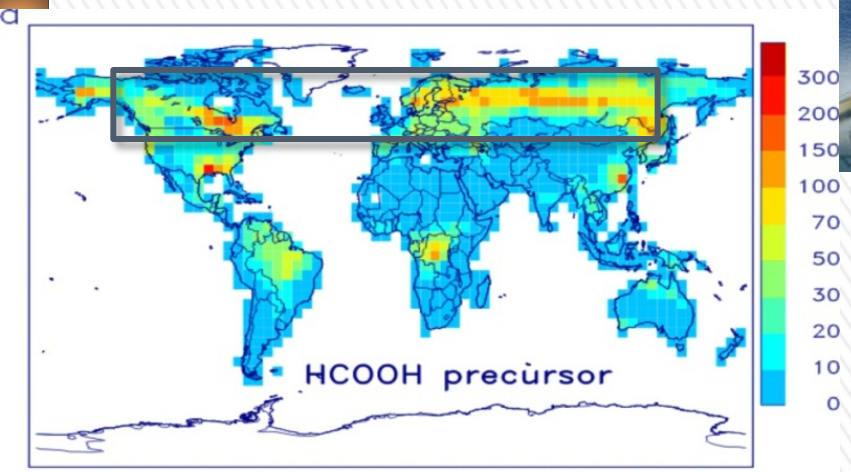


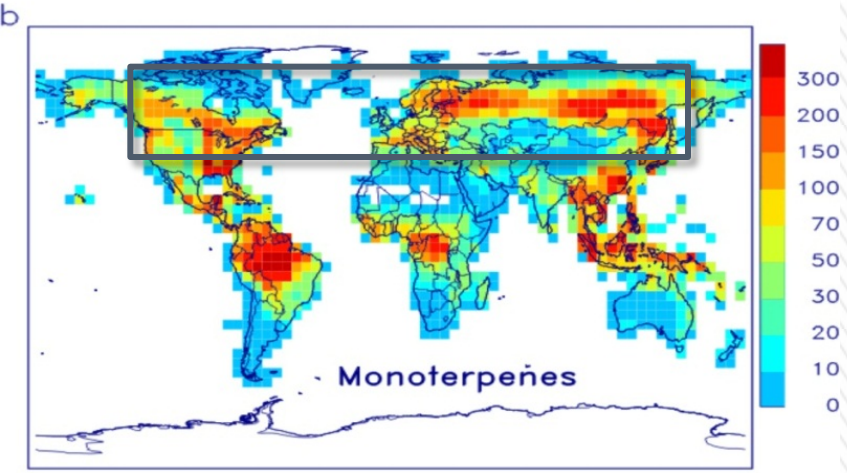
Optimized HCOOH column

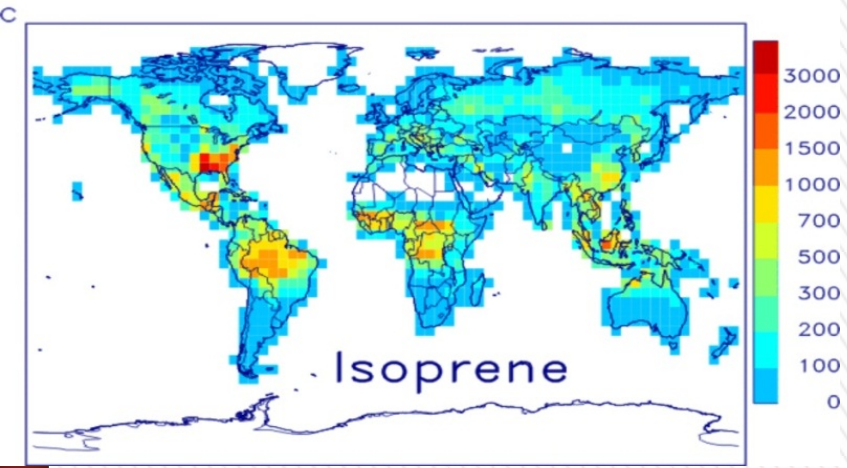


- **Strong biogenic HCOOH source : ~ 3 x larger than in the *a priori*, ~90% of the global source**

- **Enhanced emission over boreal forests likely reflecting oxidation of BVOC from conifers**





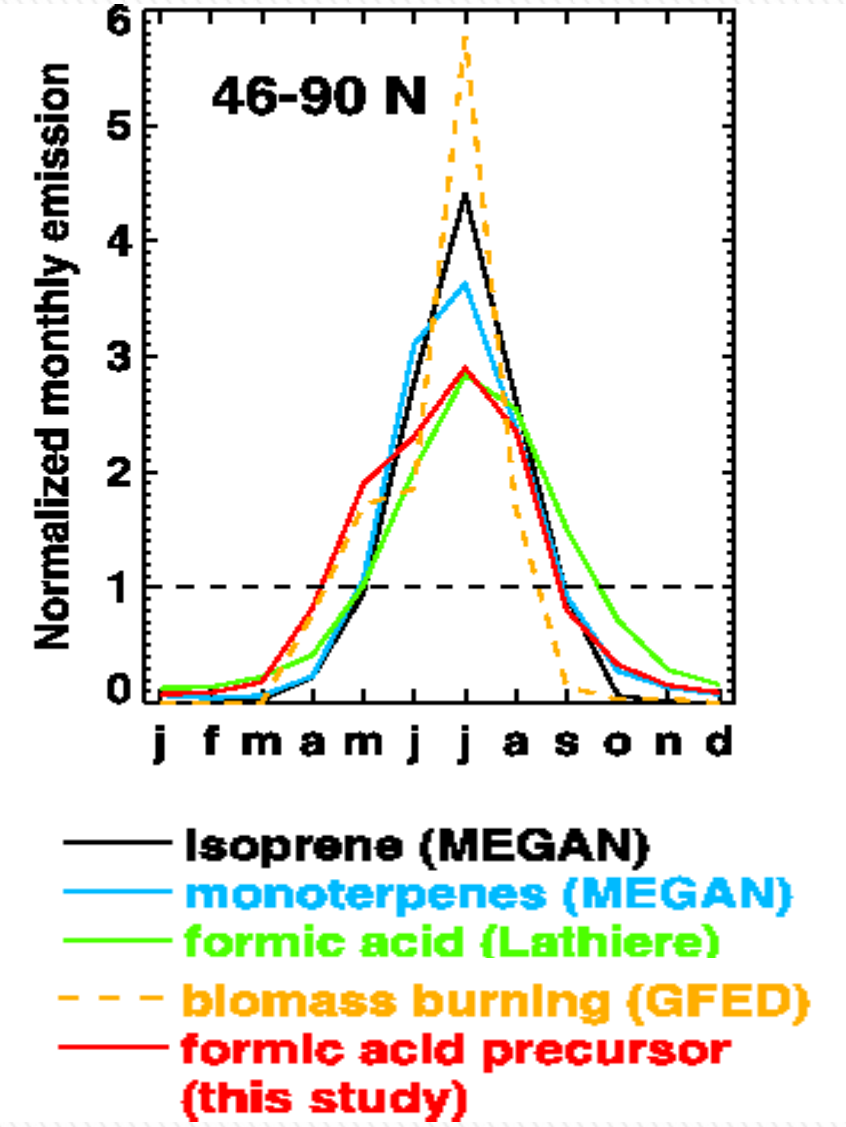


The distribution of the extra biogenic source suggests that oxidation of terpenoids leads to substantial amounts of HCOOH

Should the entire missing source of HCOOH be due to monoterpenes only, a molar yield of 200% HCOOH would be required

Stavrakou et al. Nature Geosci., 2012

Seasonal variation of the normalized monthly emission

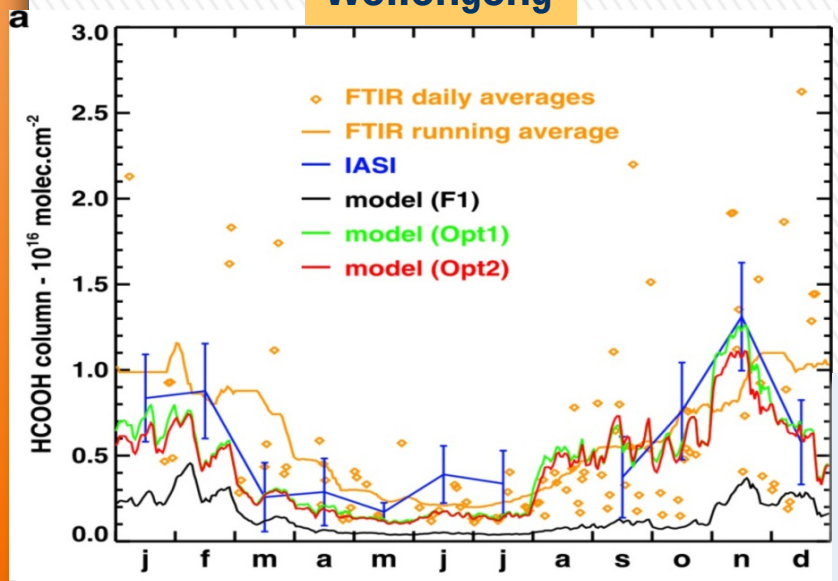


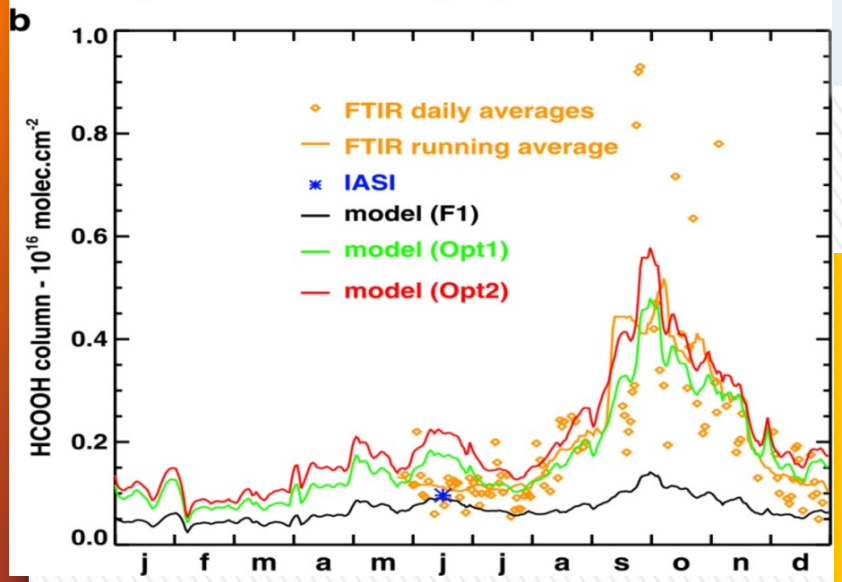
- Summertime maximum
- Relatively high emissions also in spring at high latitudes, in agreement with field studies on monoterpene emissions
- Field measurements over boreal forests point towards the existence of large emission of undetected short-lived organic compounds *(Di Carlo et al. Science, 2004)*

Comparison with FTIR data

cnes IASI Infrared Accuracy from Space In a fragile world EUMETSAT
4 - 8 February 2013 Presqu'île de Giens - Hyères les Palmiers - France

Wollongong





Reunion Isl.

✓ ***A priori*** **mean underestimation by factor of 3-4 → excellent agreement after inversion**

✓ **Comparison with airborne data (INTEX-B)**

Region	Observed	*A priori*	Optimized
Western US	490 ppt	66	230
North Pacific	140 ppt	28	120

✓ **Comparison with ground-based data**

HCOOH	Observed	*A priori*	Optimized
Gas-phase (ppbv)	1.33	0.29	0.78-1.17
Rain (µmol/L)	6.7	1.32	4-4.2

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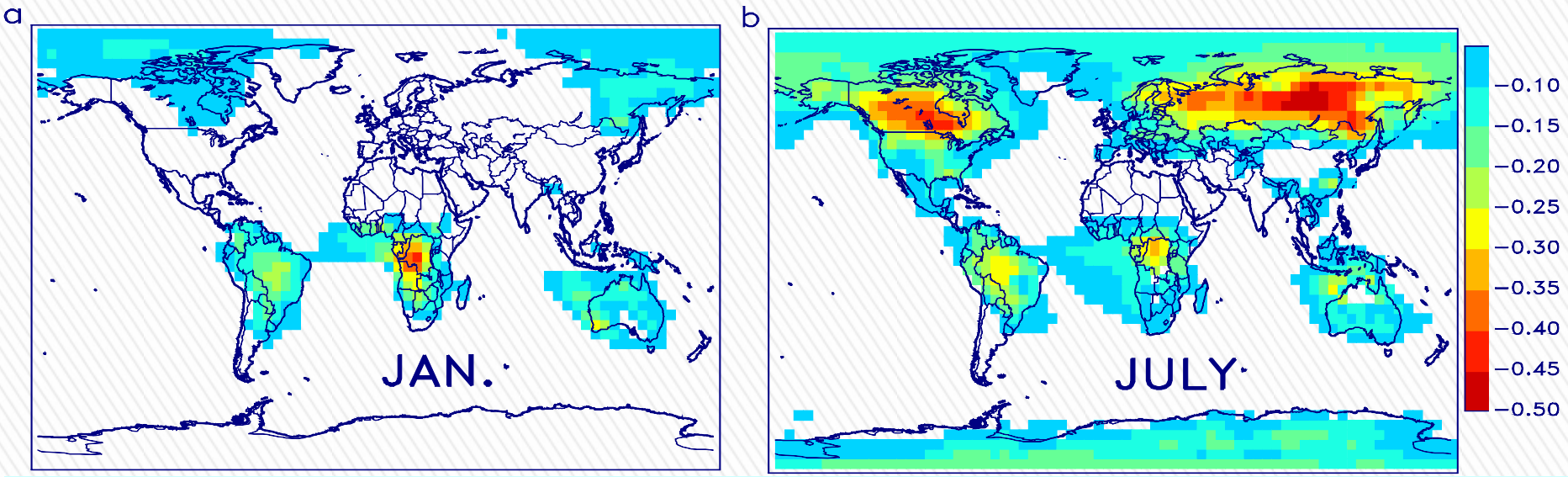
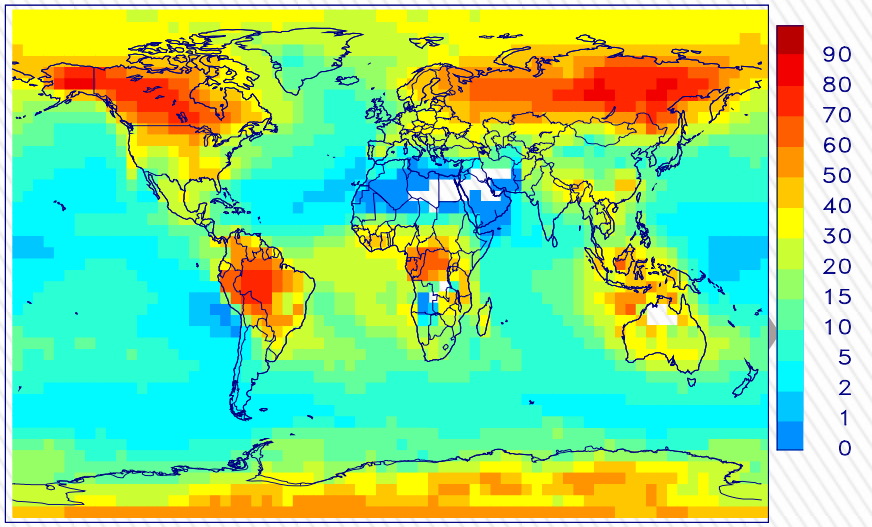
Impact of the additional biogenic HCOOH on precipitation acidity



pH reduced by 0.25-0.5 over boreal forests in summer, 0.15-0.4 in the Tropics

Calculated contribution of HCOOH to the total concentration of $[H^+]$



HCOOH accounts for as much as 60-80% of the rainwater acidity over Amazonia and boreal forests in summertime

Conclusions

- ✓ Revisited the CH_3OH & HCOOH global distribution and budget
- ✓ CH_3OH : globally inferred biogenic source : ca. 100 Tg/yr, IASI suggests higher emissions over arid regions, and lower over tropical forests, supported by independent data, issues with seasonality in the mid-latitudes
- ✓ HCOOH : biogenic secondary source ~ 90% of the total source, originates mostly in boreal and tropical forests → high-yield product in the oxidation of organic compounds, such as monoterpenes and other terpenoids
- ✓ IASI measurements pave the way to a better understanding of the origin and fate of oxygenated compounds