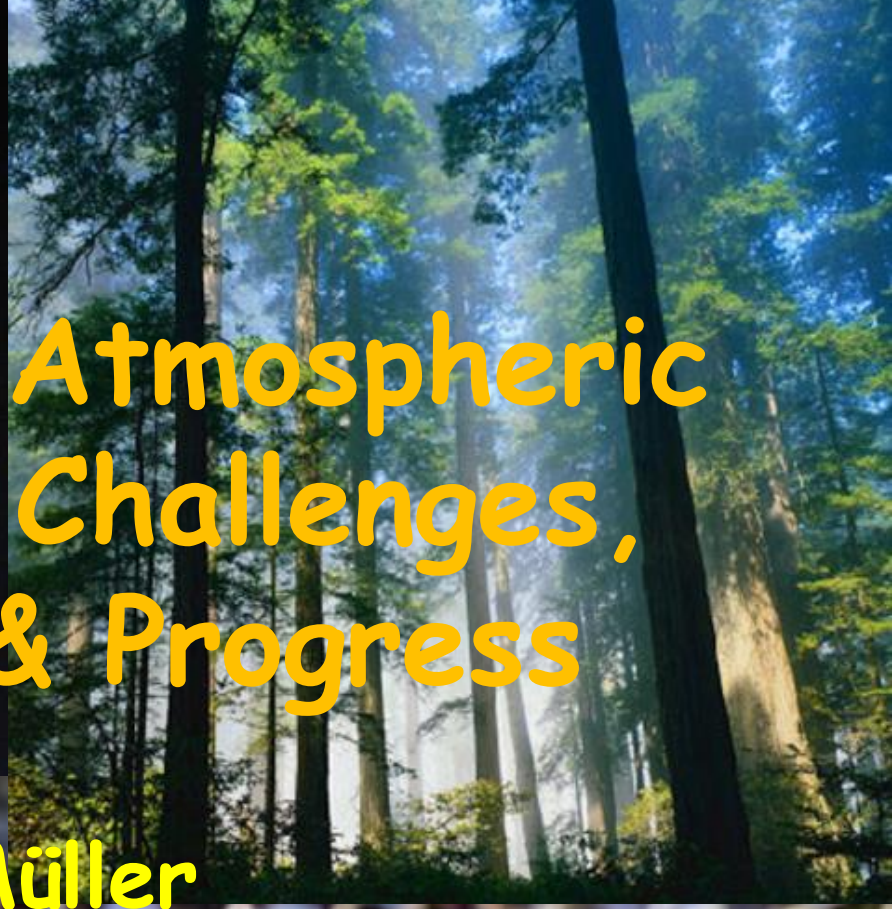
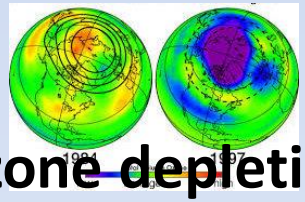
Understanding Atmospheric Composition : Challenges, Importance & Progress



J.-F. Müller
and the D20 Division

"Atmospheric Composition: Sources and Sinks"



ozone depletion

climate

air quality

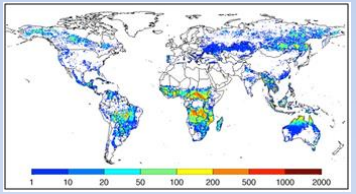
How do human-controlled emissions compare with natural emissions?
Are emission controls effective?
What is the impact of climate change on natural emissions?

APPROACHES TO SOURCE ESTIMATION

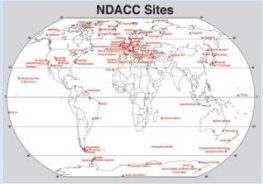
- Experimental (direct)

- Experimental (less direct)

- Emission modelling

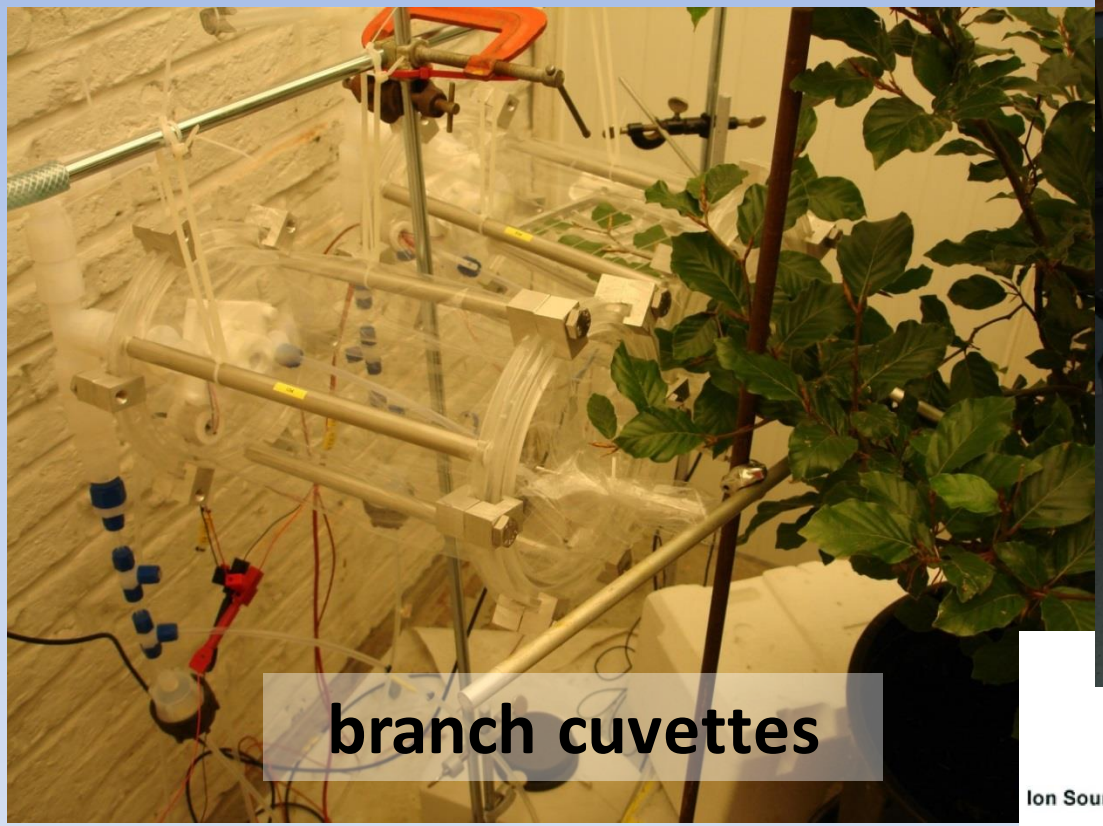


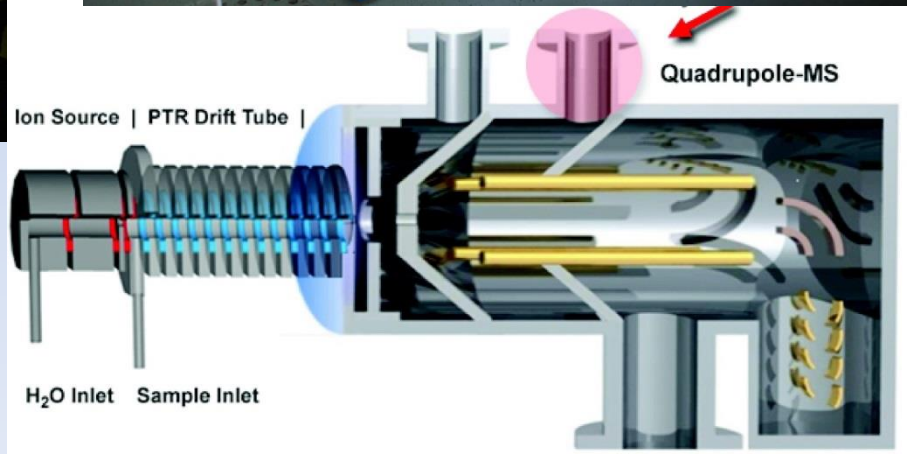
- Estimations based on atmospheric composition measurements (& models)



+

Direct measurement of emissions of volatile organic compounds by trees

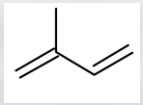


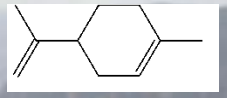


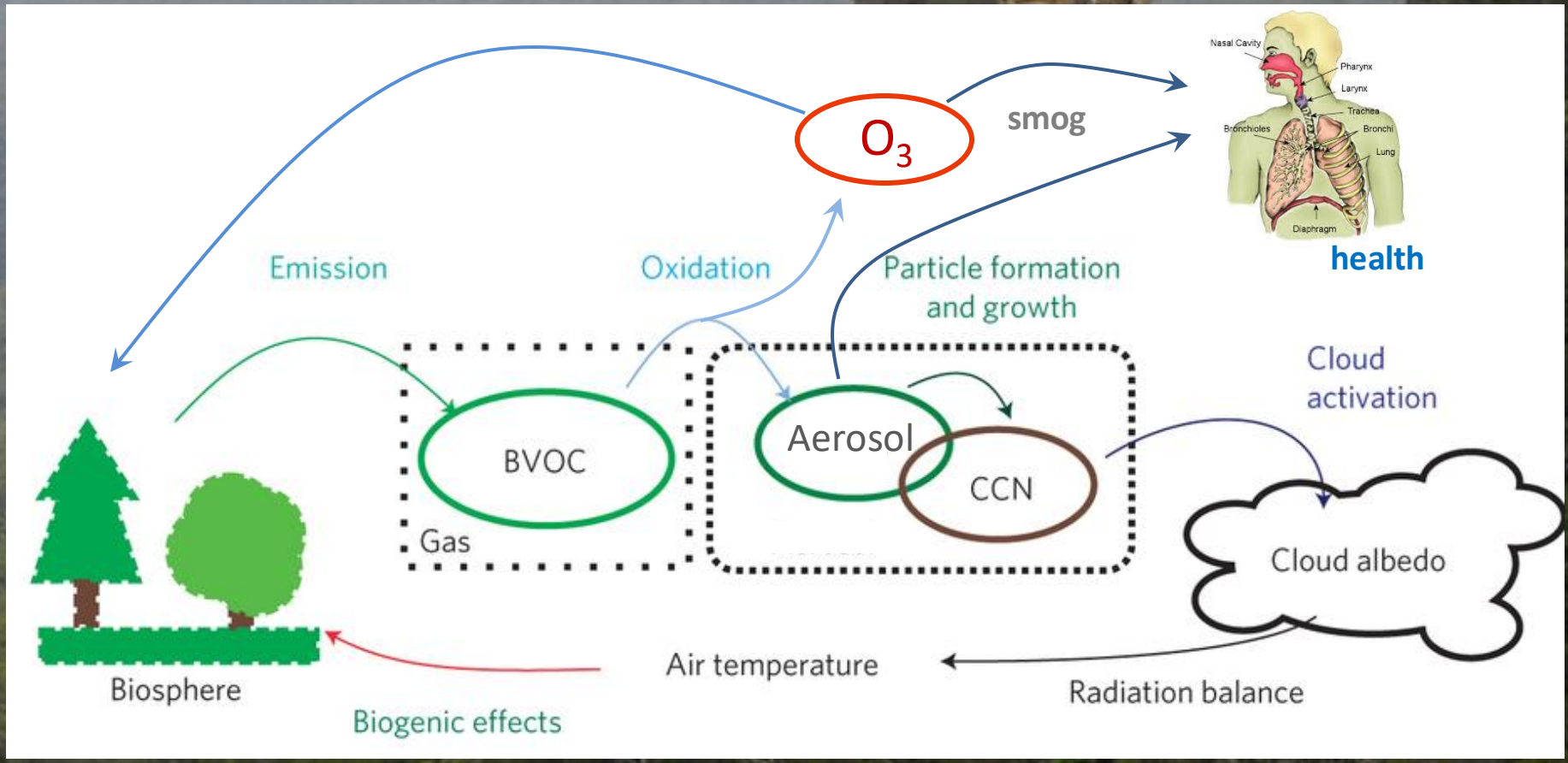
Chemical Ionisation Mass Spectrometry (CIMS)

Amelynck et al., 2005, 2013; Dhooghe et al., 2008, 2009, 2010; Schoon et al., 2008; Demarcke et al., 2010

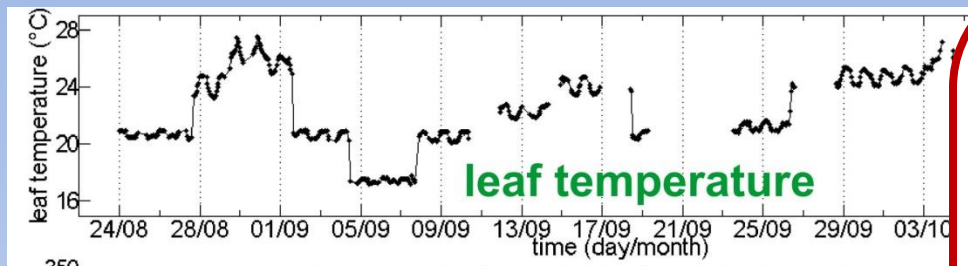
WHY CARE ABOUT BIOGENIC VOLATILE ORGANIC COMPOUNDS (BVOC) ?

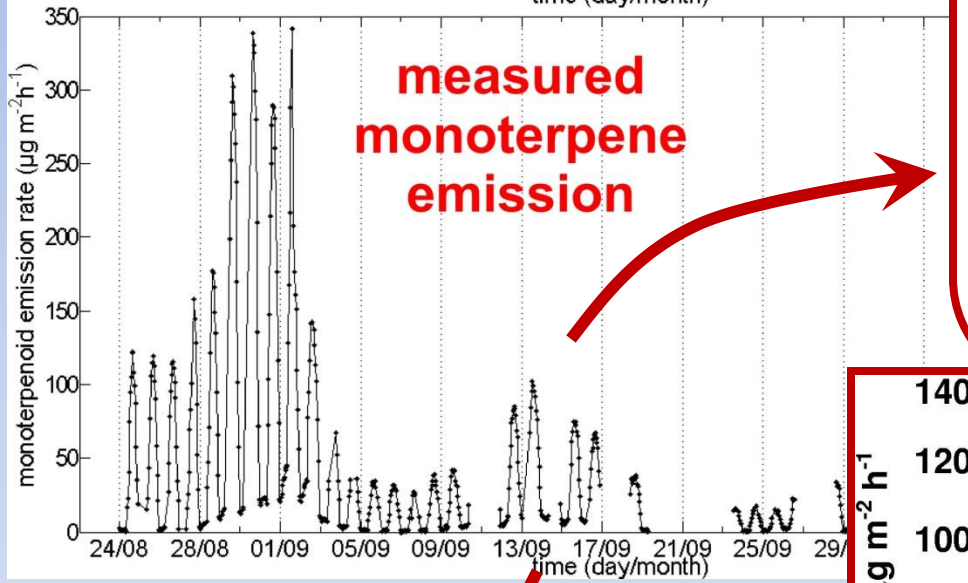


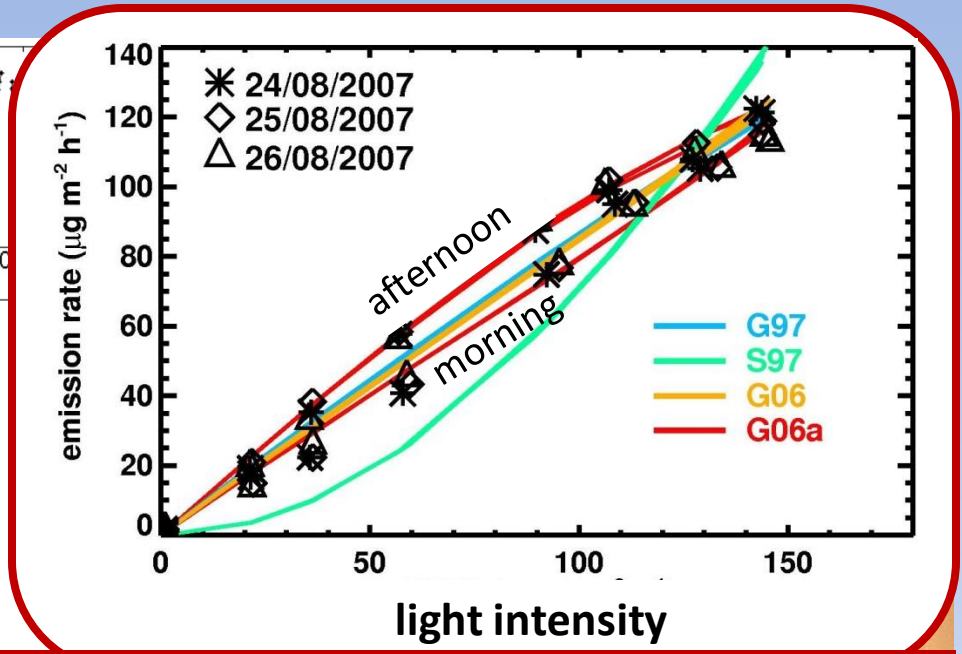


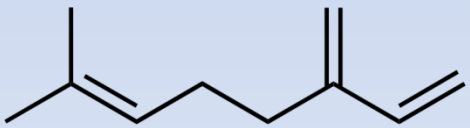


How do hydrocarbon emissions depend on temperature and light?

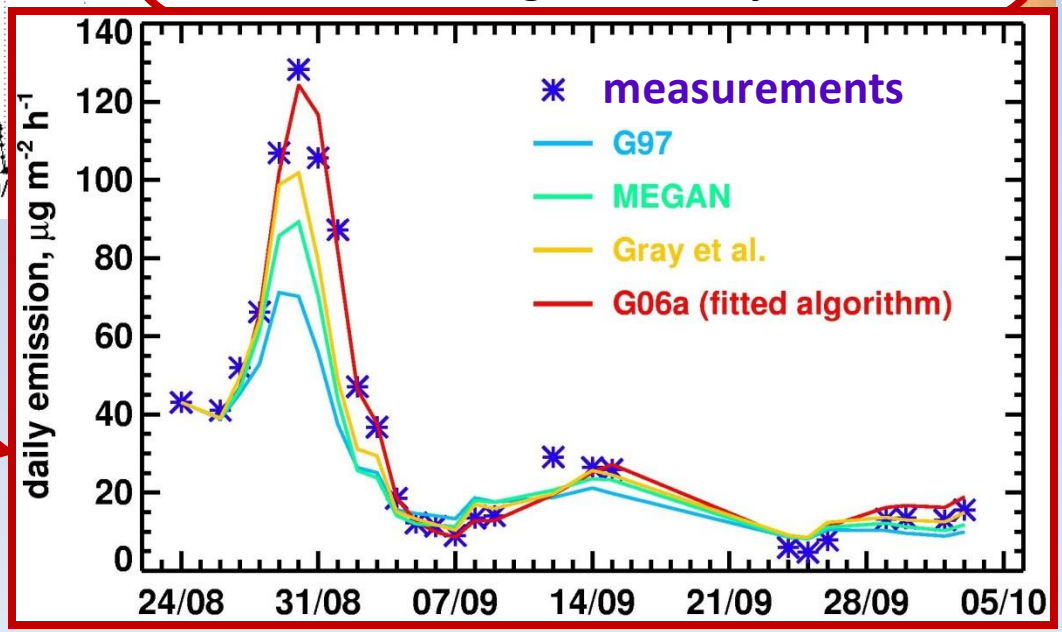




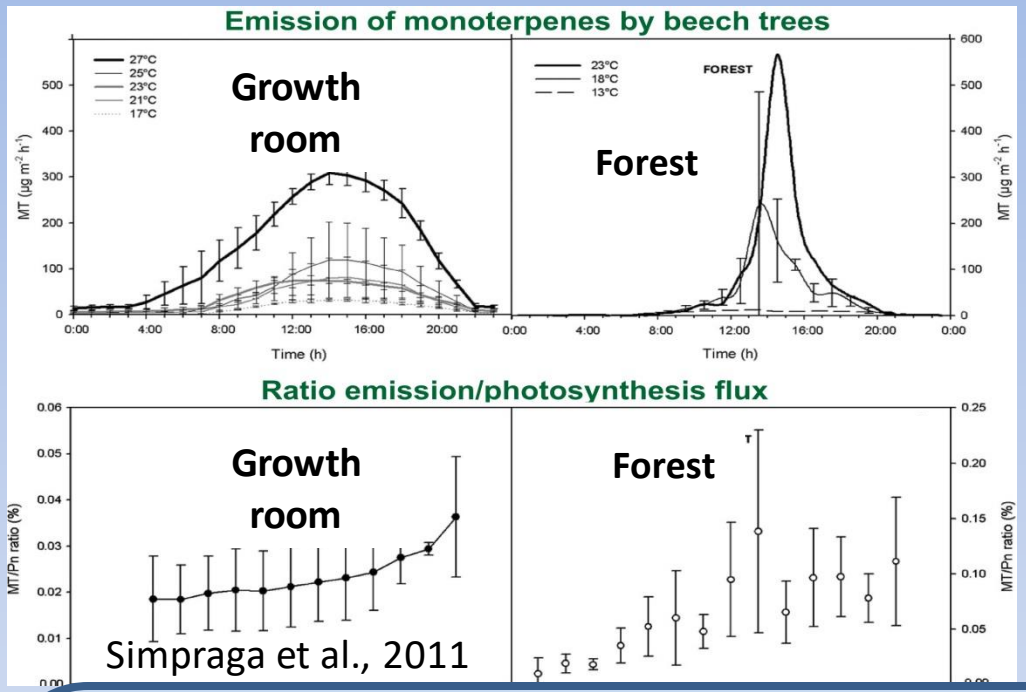




Demarcke et al., AE, 2010

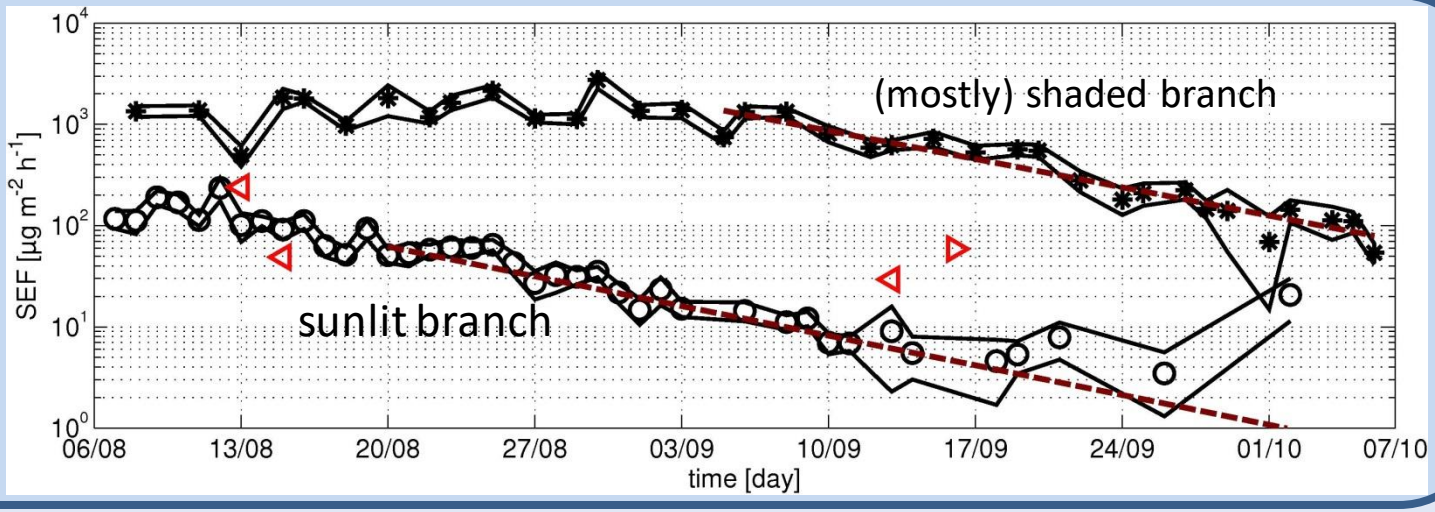


From the growth room to a real forest (Aelmoeseneie near Ghent)



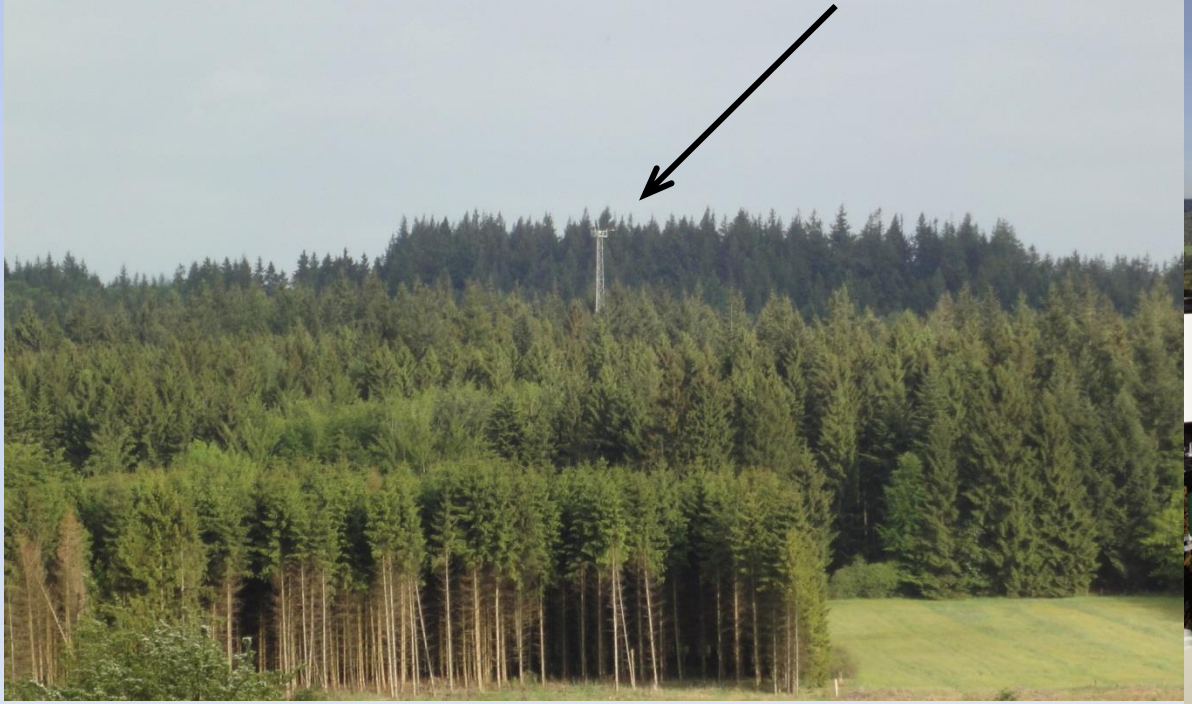
Leaf senescence induces a decline in the (normalized) emission factors

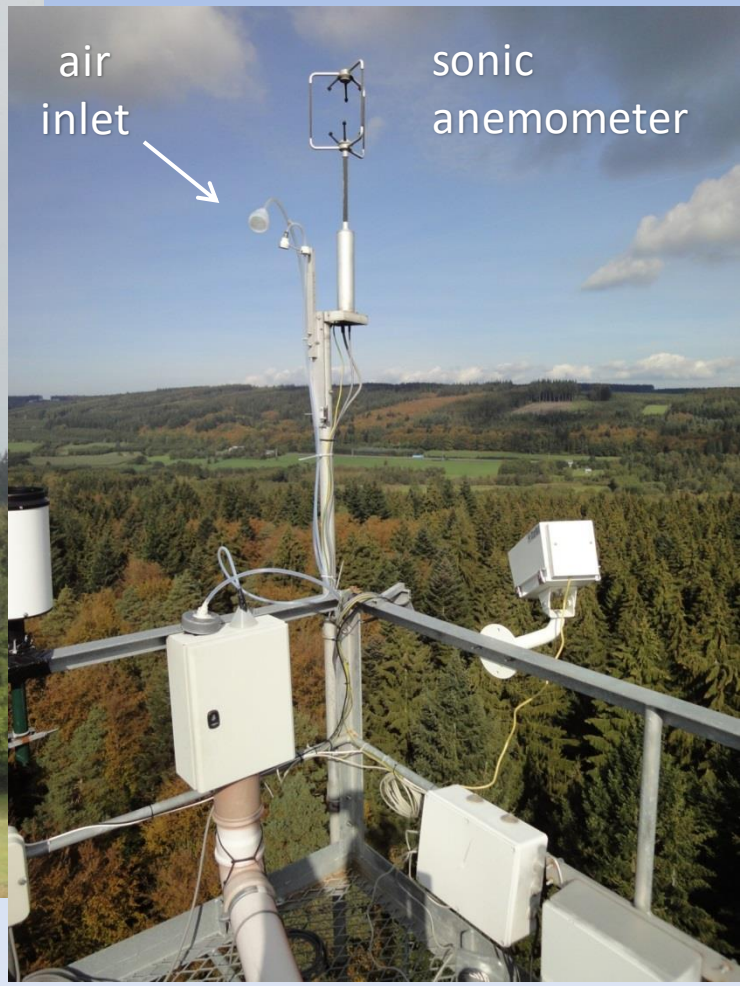
(M. Demarcke, PhD Thesis, 2011)



Measuring the emissions from the entire forest canopy

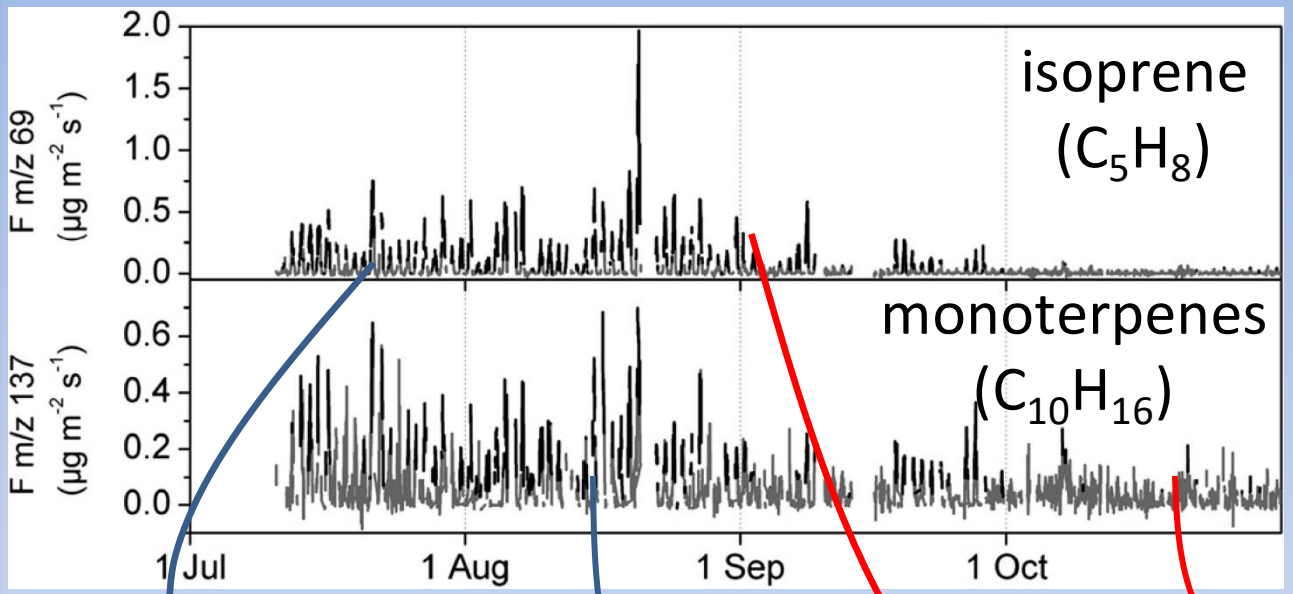
Eddy covariance technique : use FAST on-line measurements of VOC concentration and air velocity above a canopy, i.e. from a tower

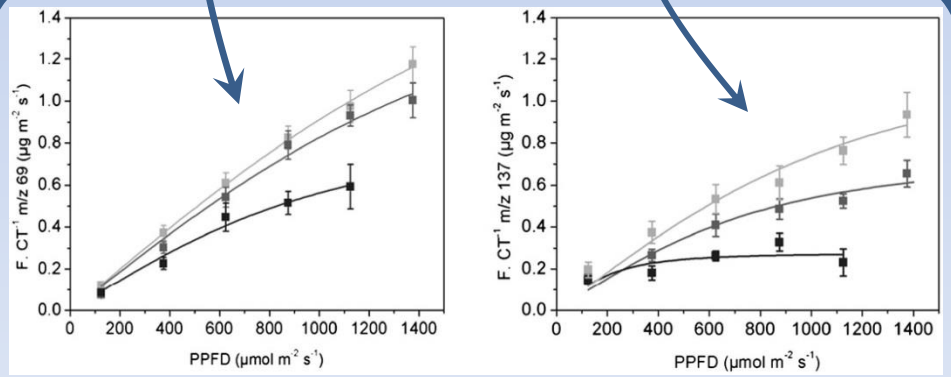




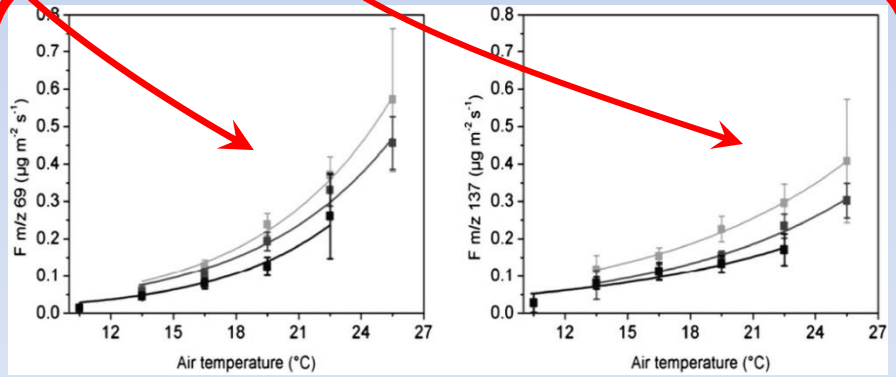
gembloux
agro bio tech

Measured fluxes from a forest near Vielsalm





dependence on light intensity

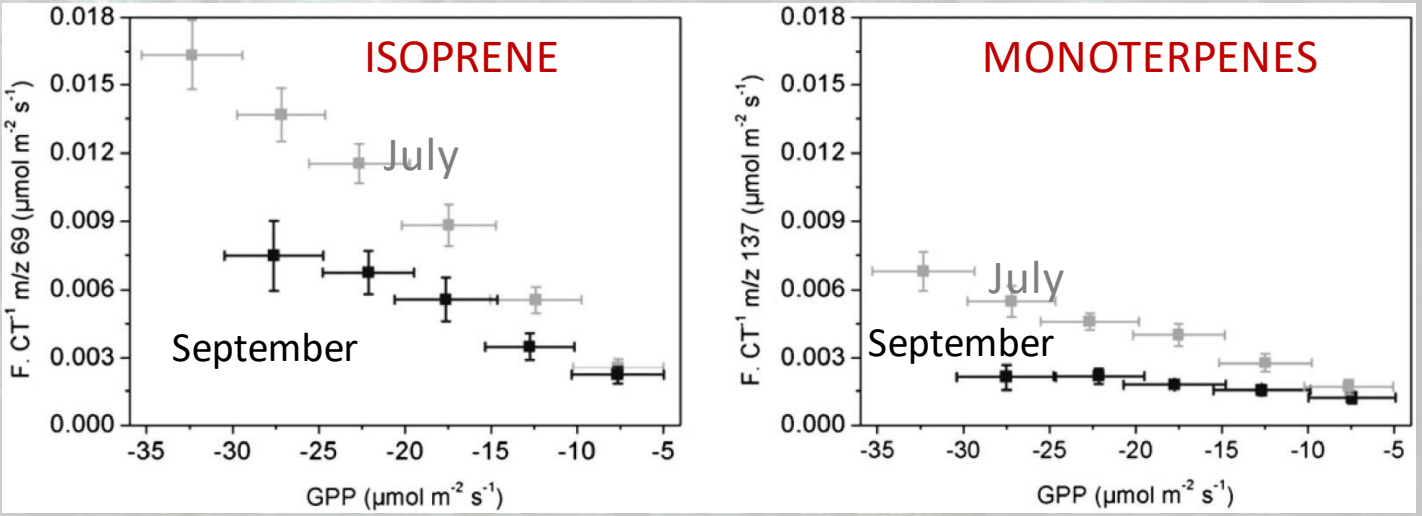


dependence on temperature

(Laffineur et al., AE 2011)

What really drives the emissions?

Emissions relate to Gross Primary Productivity (GPP)



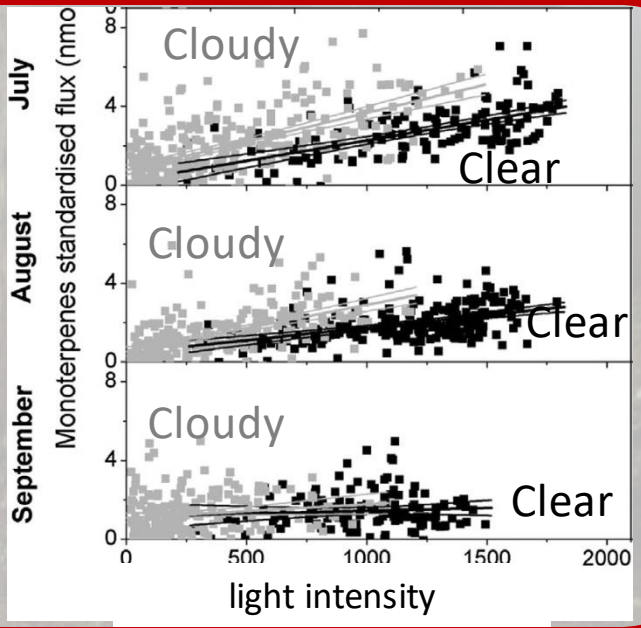
Seasonal changes reflect changes in enzymatic activity

(Laffineur et al., AE 2011)

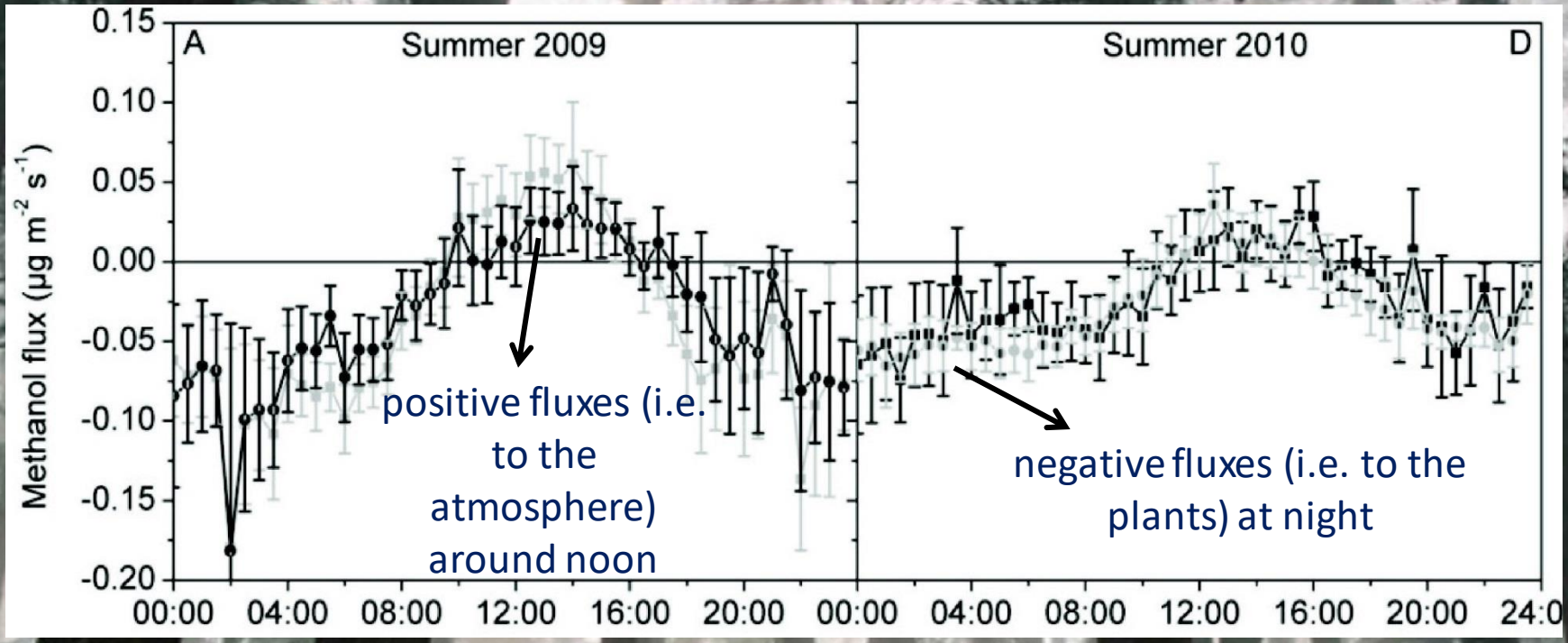
The fluxes are higher under cloudy conditions compared to clear-sky at equivalent temperature and light conditions

Due to better penetration of diffuse light in the canopy

(Laffineur et al., 2013)



The case of methanol



➔ water film on vegetation acting as a reservoir of methanol in moist conditions (Laffineur et al., 2012)

Flux measurements in various landscapes

Grasslands, Dorinne, 2014

To be continued…

Biogenic emission modelling

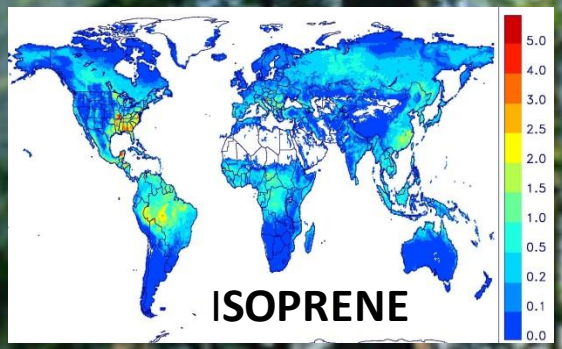
Guenther et al. 2006

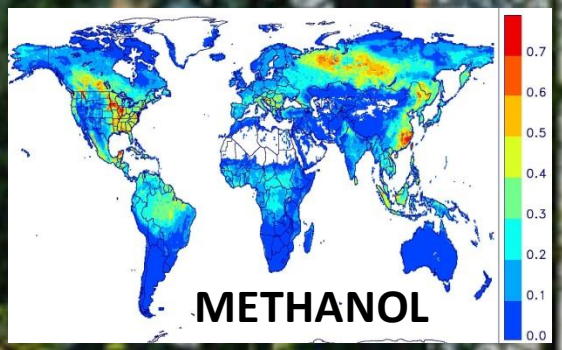
Müller et al., 2008
Stavrakou et al., 2011

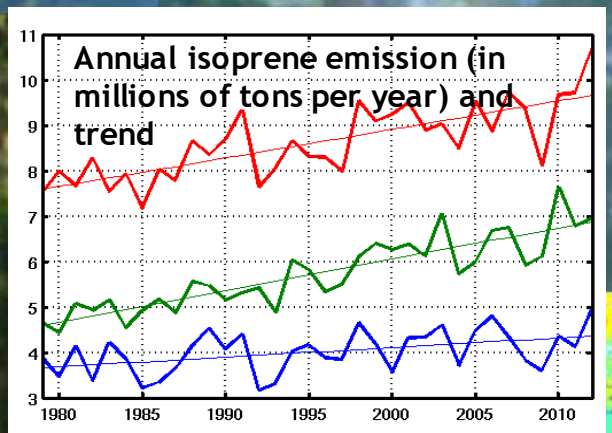
Emission rate in standard conditions

Dependence on temperature, light intensity, leaf age, soil moisture, …

Leaf area index





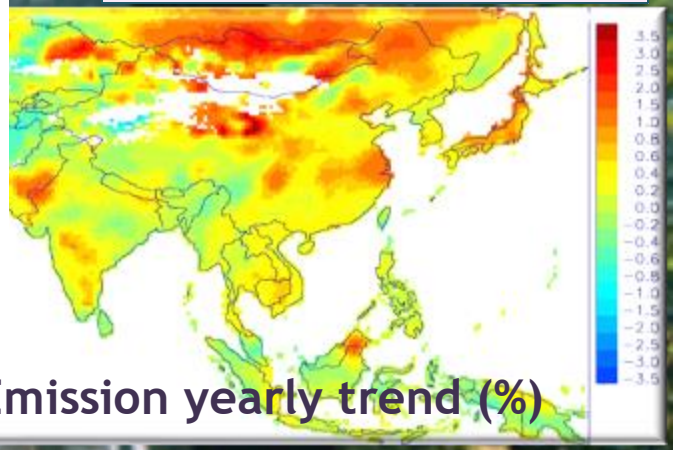


Canada : 5.3%/decade
Europe : 12.1%/decade
Russia : 7.3%/decade

Stavrakou et al., 2014
Sindelarova et al., 2014

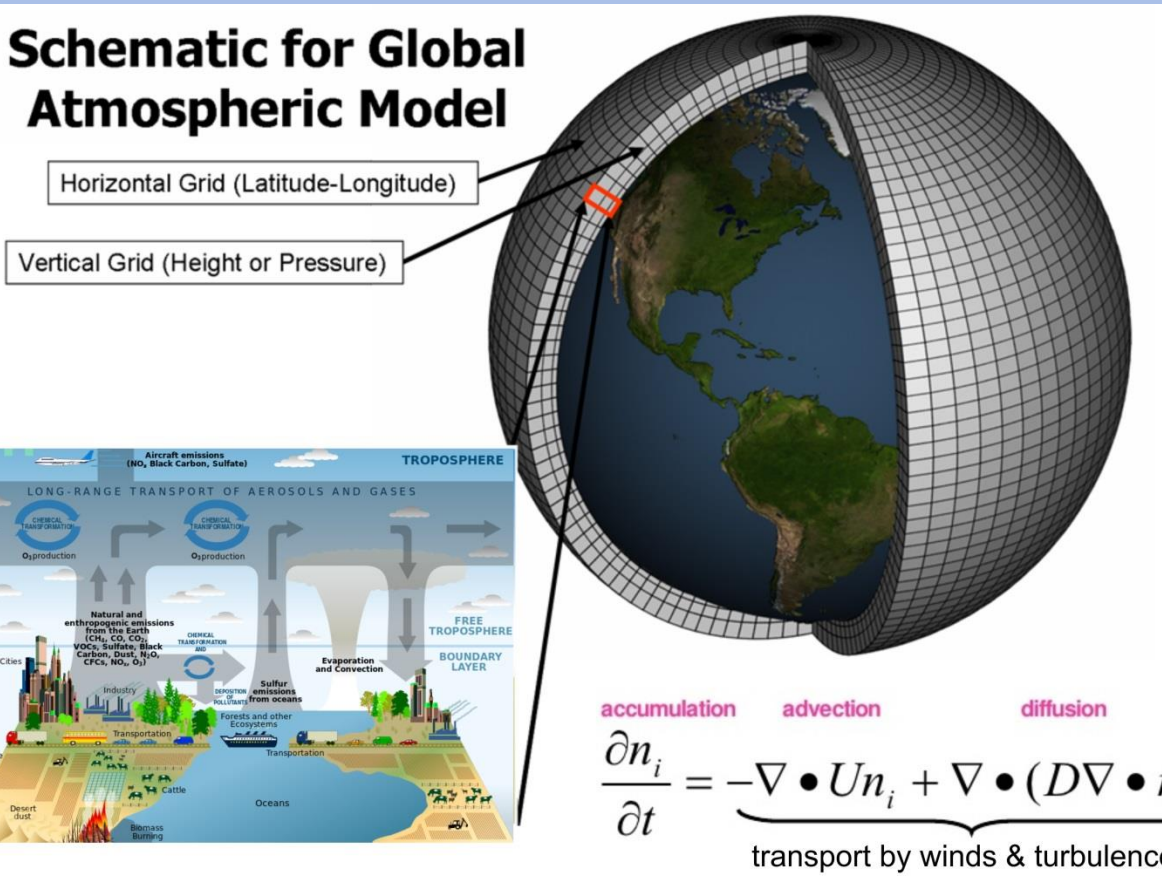
Assess role of

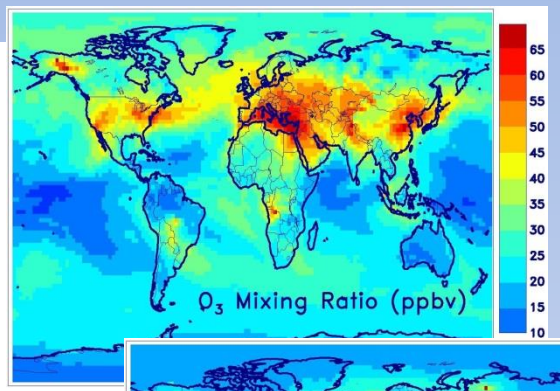
- climate change
- land use change
- solar dimming/brightening

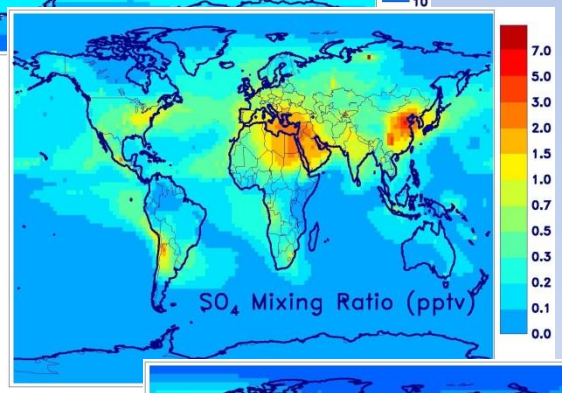


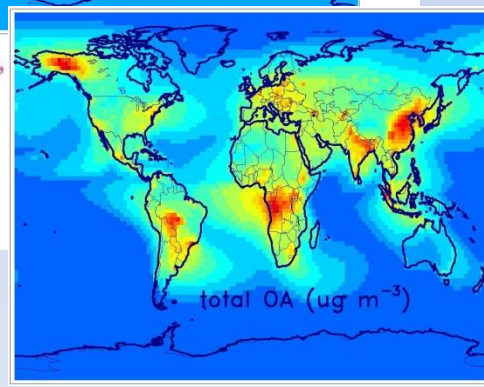
Emission yearly trend (%)

Simulating atmospheric composition – the IMAGES model









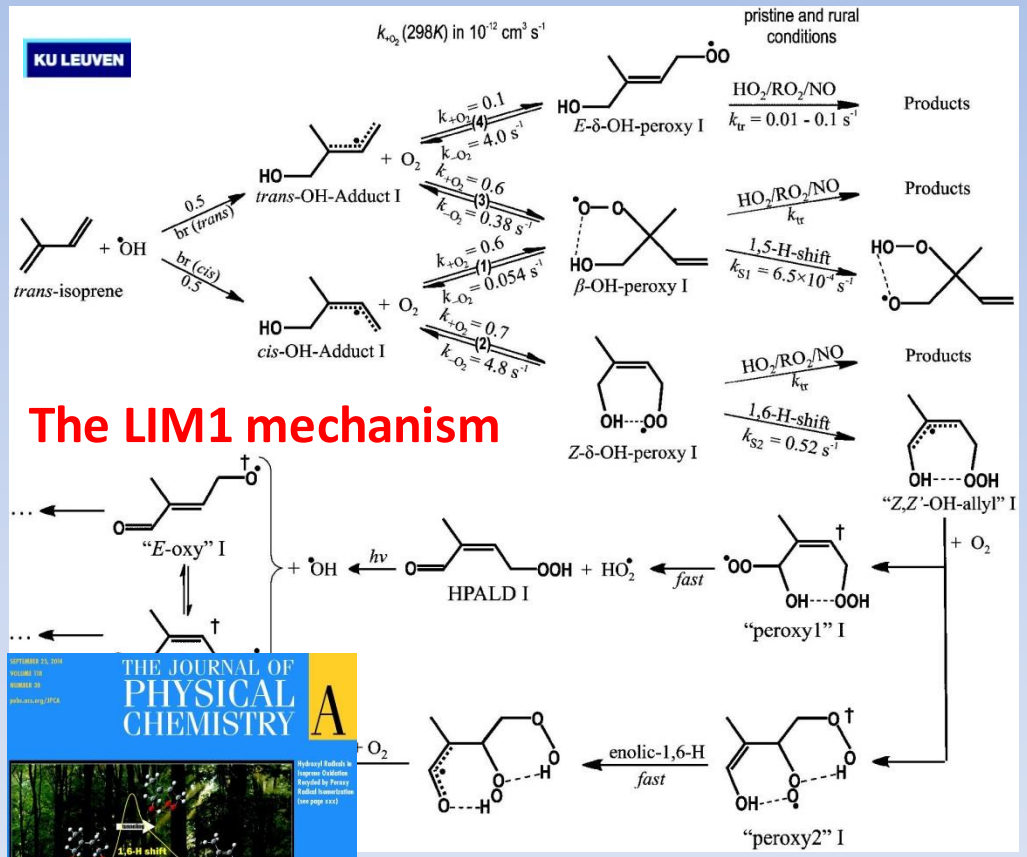
accumulation advection diffusion chemistry, emissions, deposition

$$\frac{\partial n_i}{\partial t} = \underbrace{-\nabla \bullet U n_i + \nabla \bullet (D \nabla \bullet n_i)}_{\text{transport by winds \& turbulence}} + P_i - L_i$$

Müller and Brasseur, 1995, 1999; Pham et al., 1995; Granier et al., 1998, 2000; IPCC Reports, 1999, 2000; Stevenson et al., 2006; Dentener et al., 2006; Shindell et al., 2006; etc.

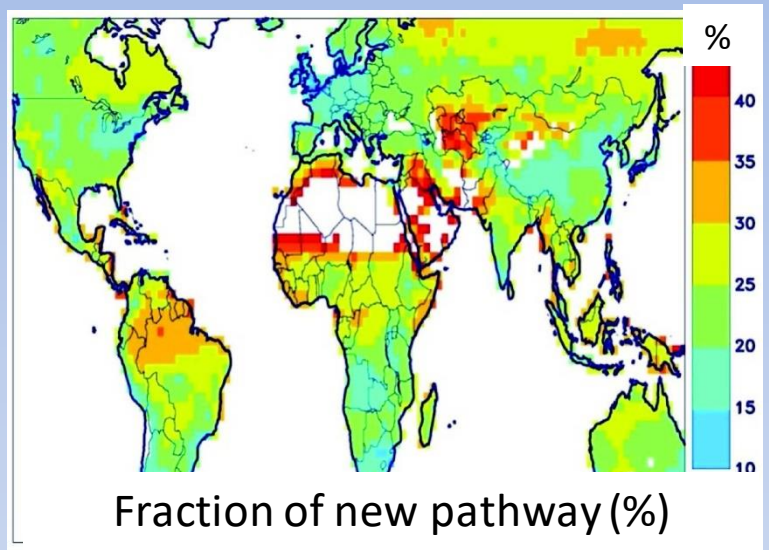
Impact of biogenic hydrocarbons on the oxidizing capacity of the atmosphere

Revisiting the chemistry of isoprene

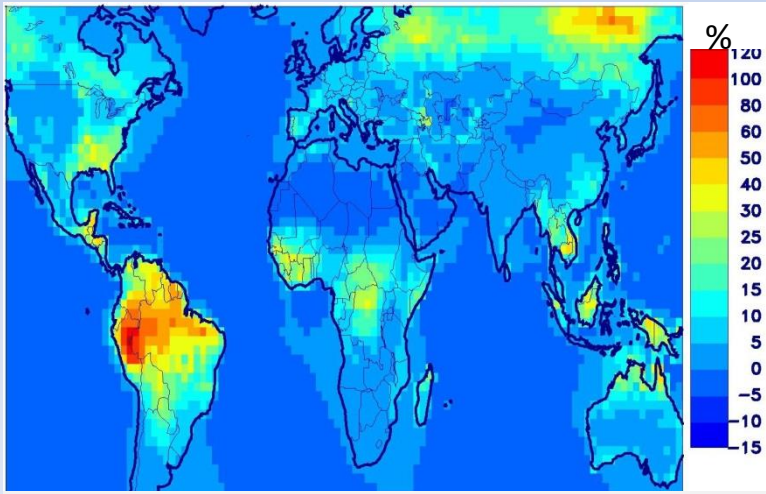


The LIM1 mechanism

Peeters & Müller, 2010
Stavrakou et al., 2010
Peeters et al., 2014



Fraction of new pathway (%)

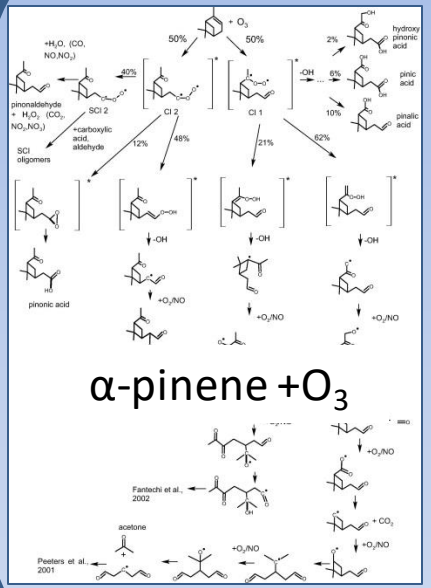


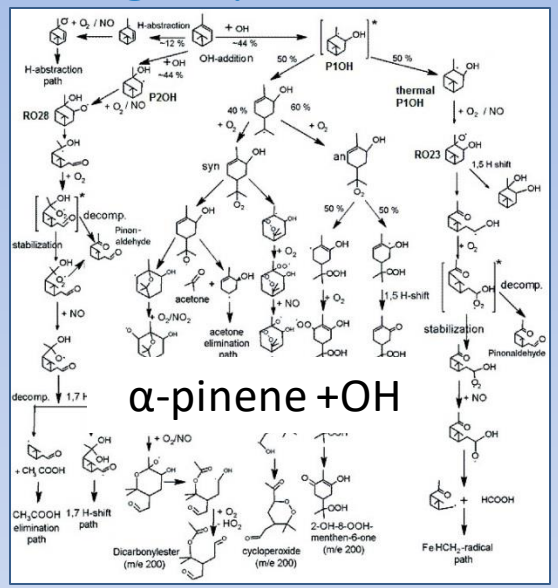
Impact of new chemistry on OH (%)

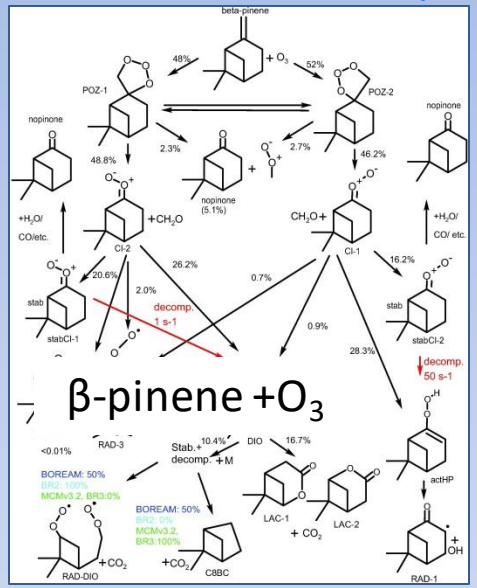
Modelling organic aerosol formation

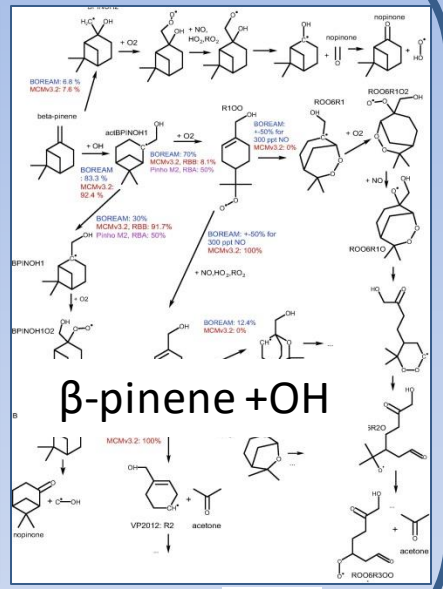
BOREAM model =

detailed gas-phase oxidation mechanism for pinenes





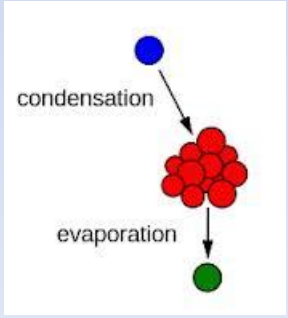




Capouet et al., 2004, 2008; Ceulemans et al., 2010; Ceulemans, 2014

KU LEUVEN

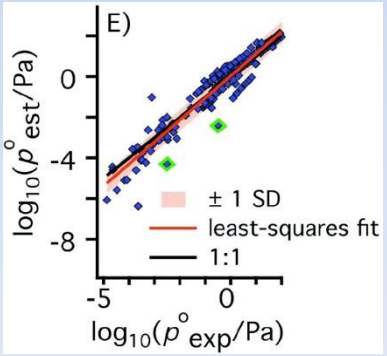
+



Aerosol formation module

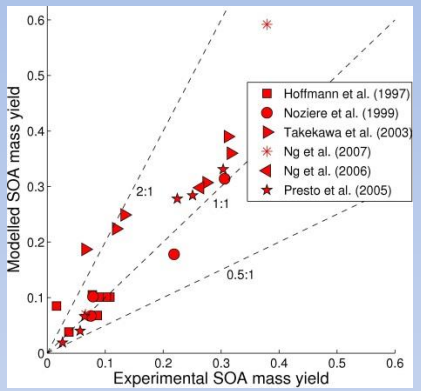
based on state-of-the-art estimation of vapour pressures and activity coefficients

Compernolle et al., 2009, 2010, 2011

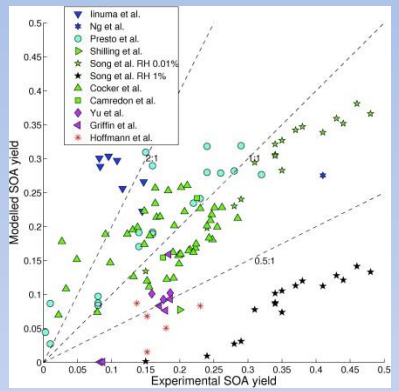


Impact of biogenic hydrocarbons on organic aerosol formation

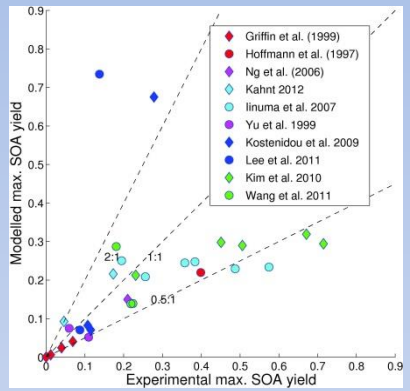
Model evaluation using laboratory data



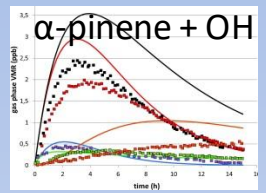
α-pinene + OH, O_3

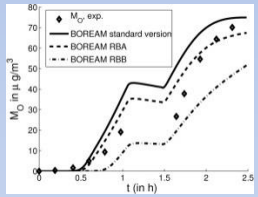


α-pinene + O_3



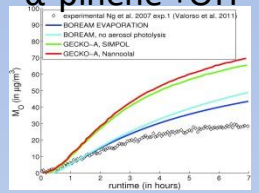
β-pinene + O_3

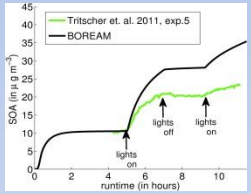




β-pinene + OH

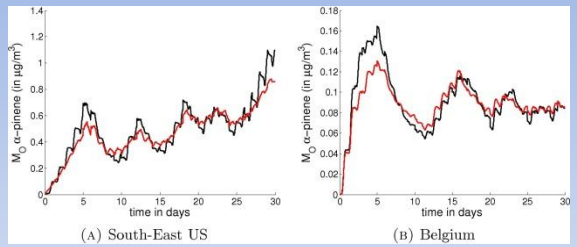
α-pinene +OH

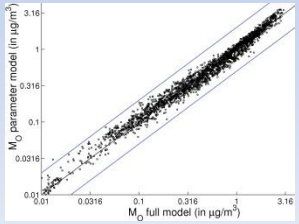




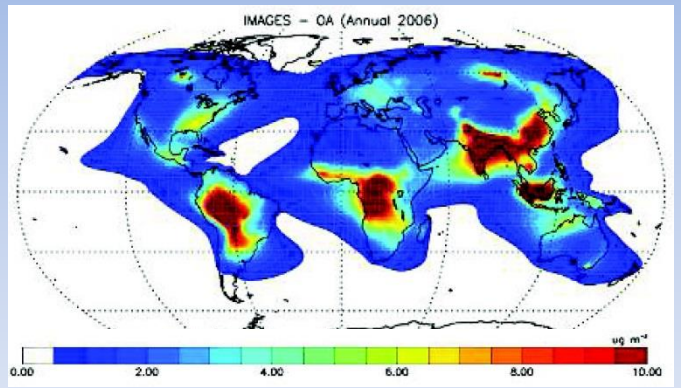
α-pinene+O_3, OH

Simplified module for use in global model

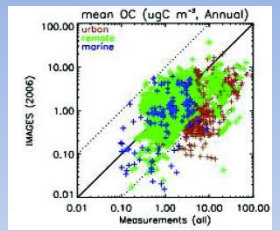


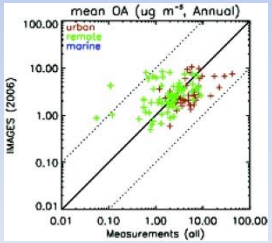


Complete model vs simplified model

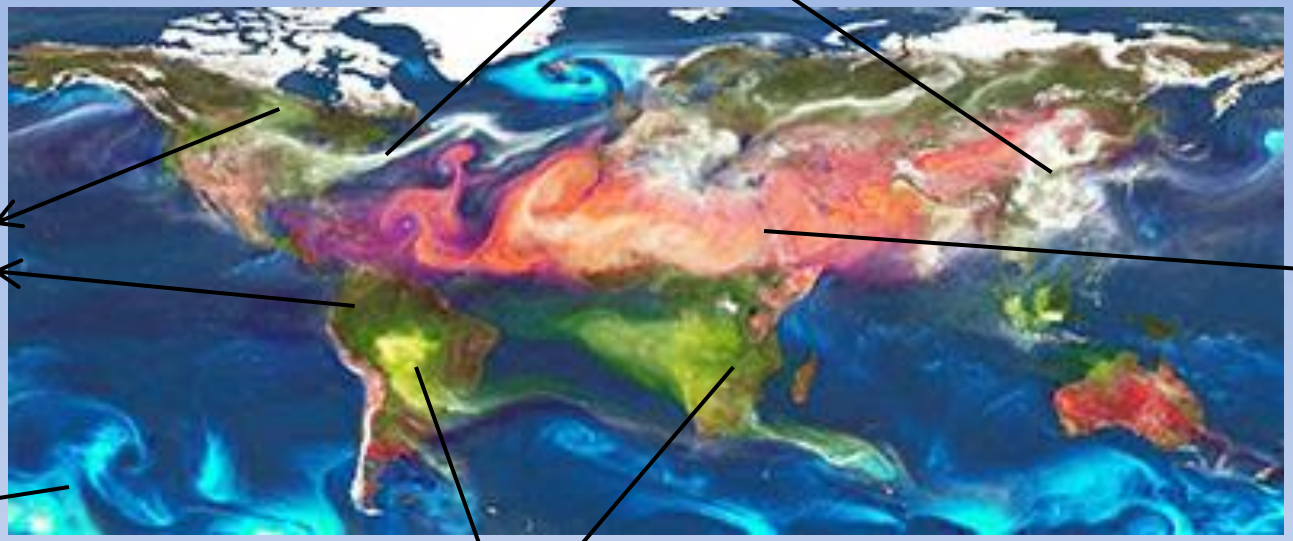


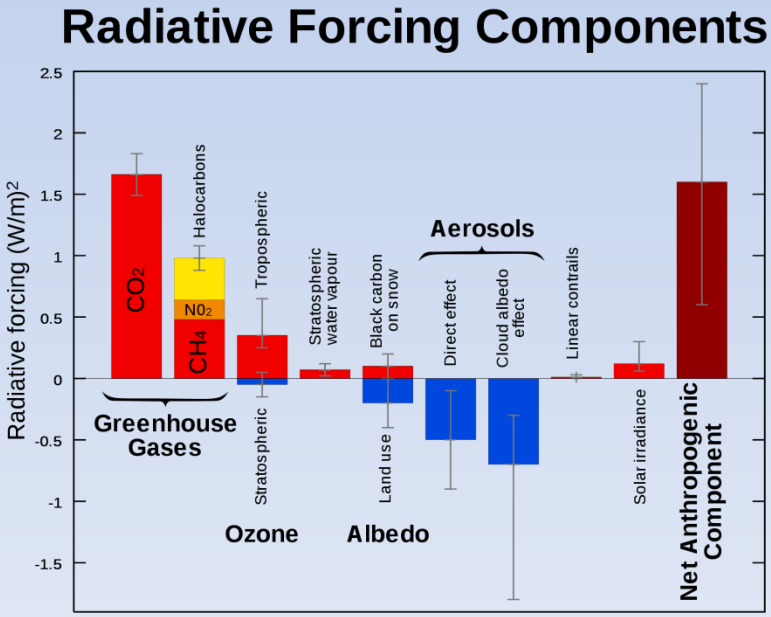
Ceulemans et al., 2012; Tsigaridis et al., 2014





TROPOSPHERIC AEROSOLS





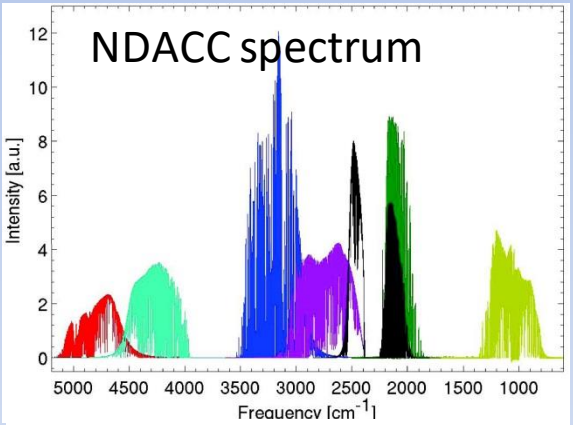
need global scale measurements of atmospheric composition

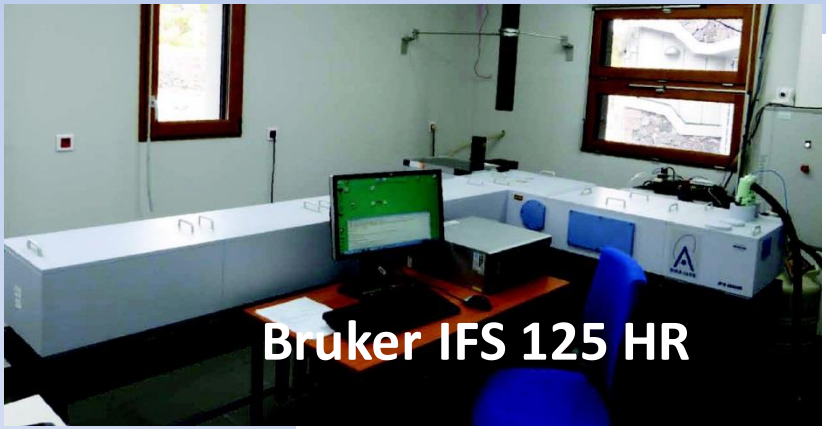
+

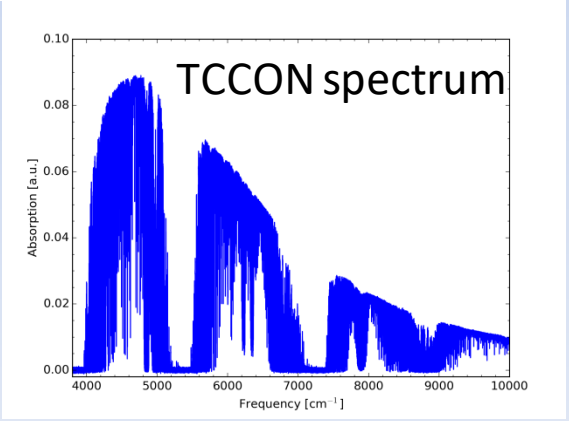
long-term & accurate !

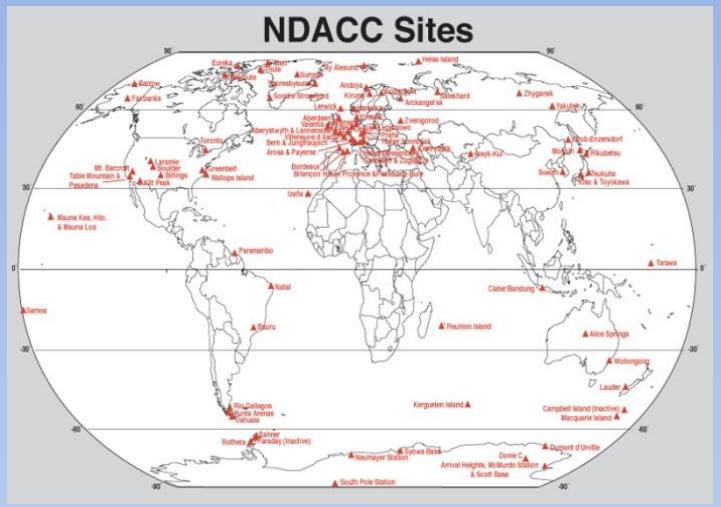
Ground-based measurements using infrared sensors

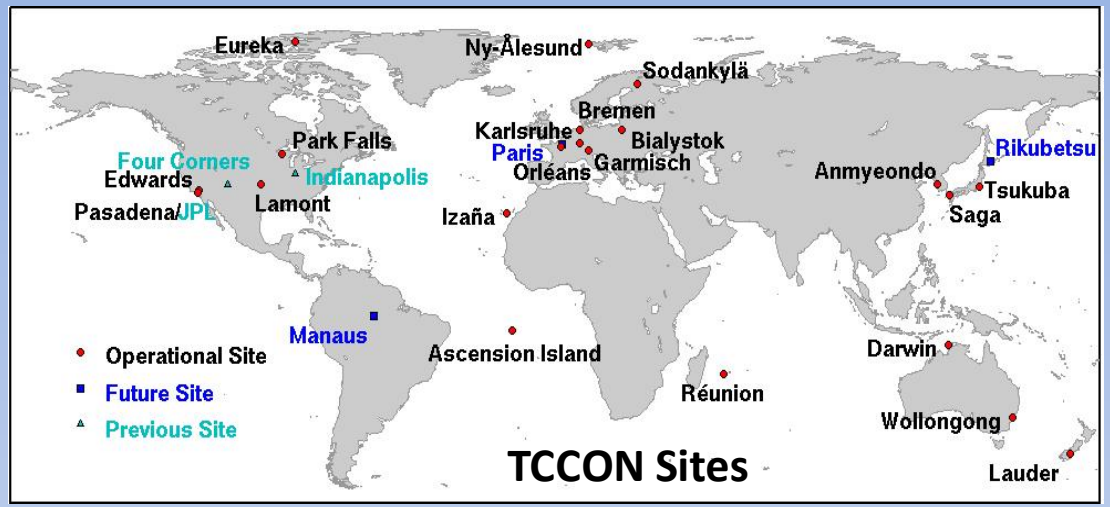
High resolution measurement of absorption spectrum of solar light in the infrared











Jungfraujoch

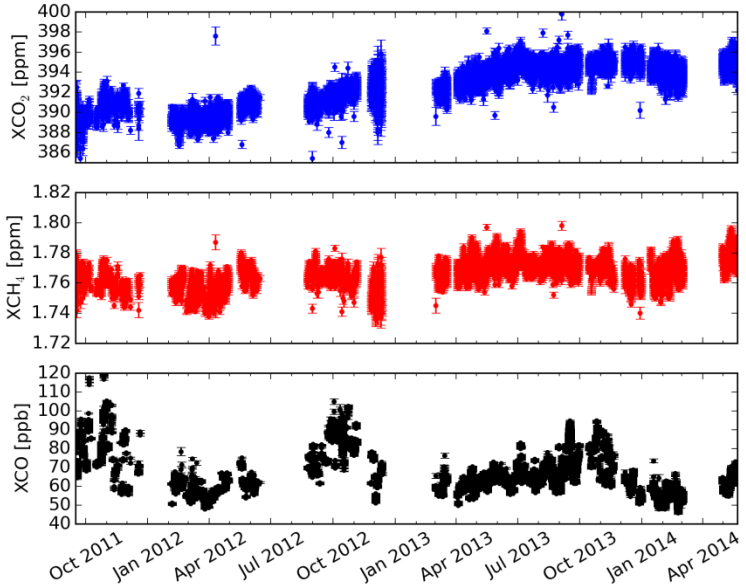
La Réunion

Uccle

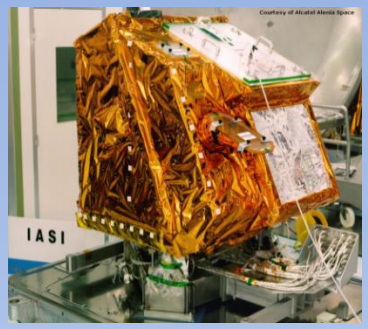
+ Harestua, Haute-Provence

Future : Porto Velho, Brazil

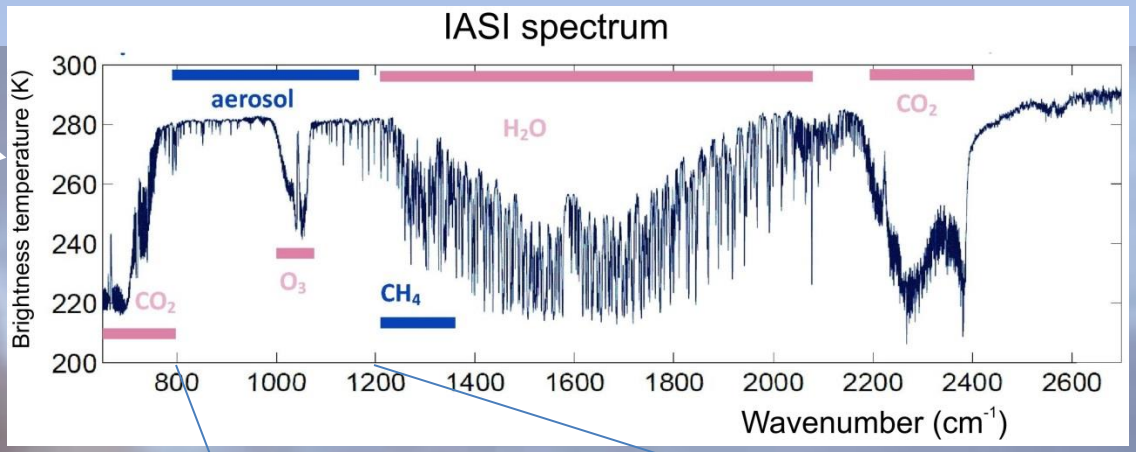
TCCON observations at Reunion Isl.



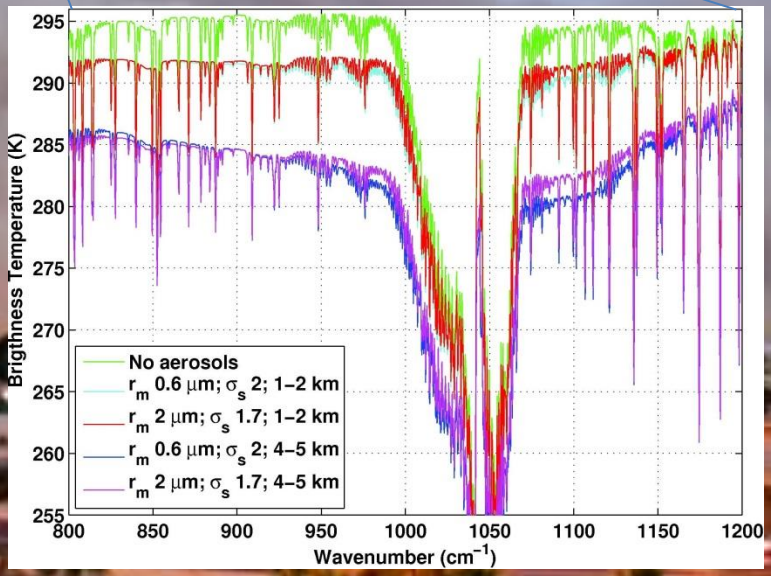
Dust seen by IASI on METOP



Brightness temperature measured by IASI

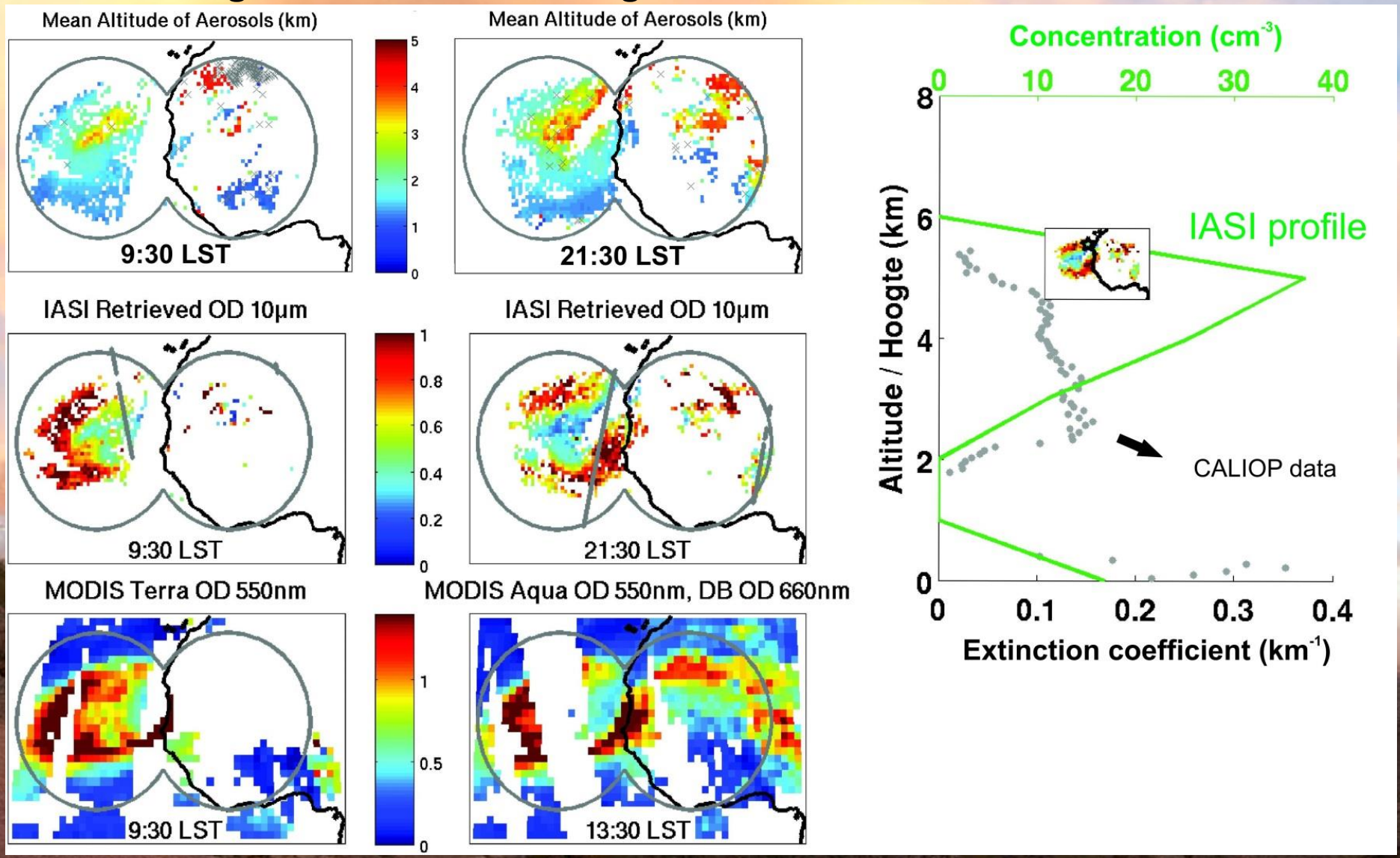


The higher the dust layer, the stronger the impact on brightness temperature



Desert dust seen by IASI

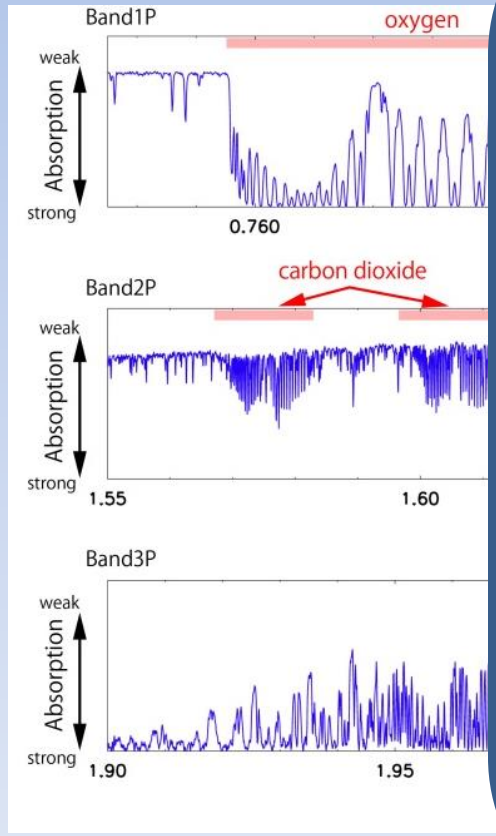
morning **evening**



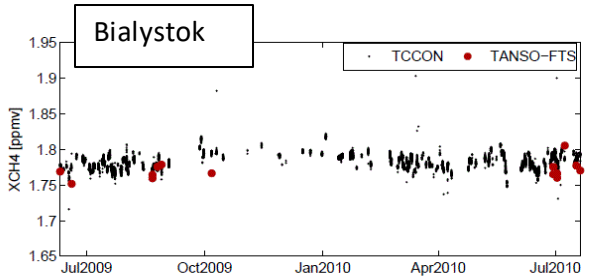
Methane seen by TANSO-FTS on GOSAT

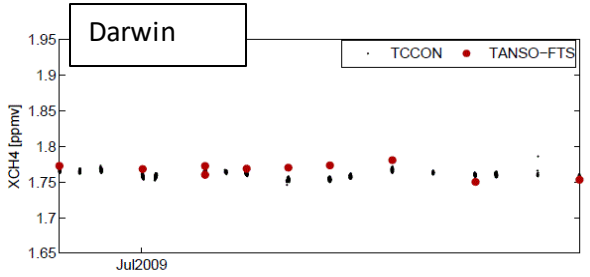
retrieval using ASIMUT-ALVL

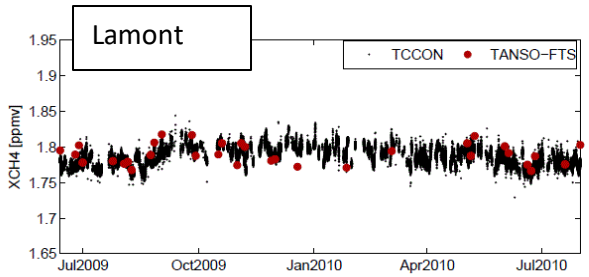
Shor

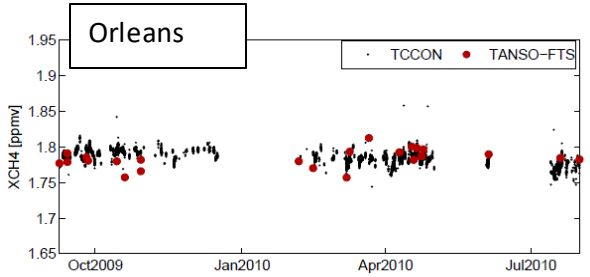


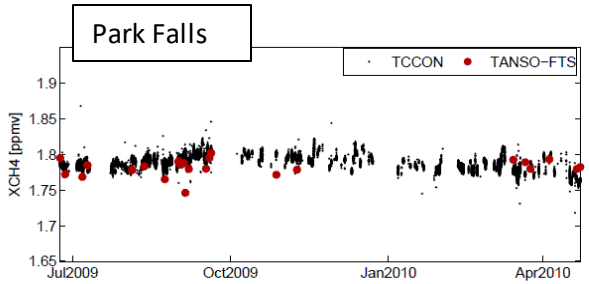
validation with TCCON measurements

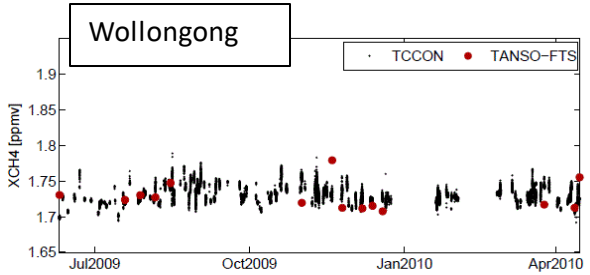


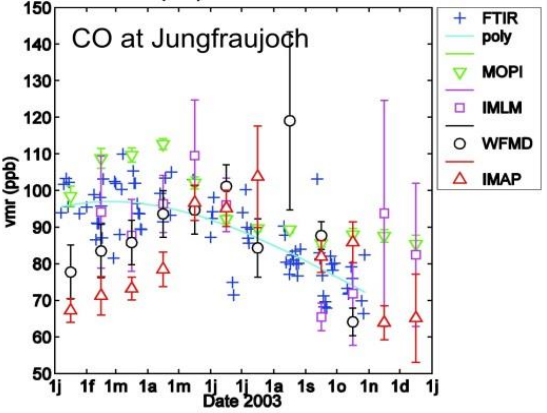


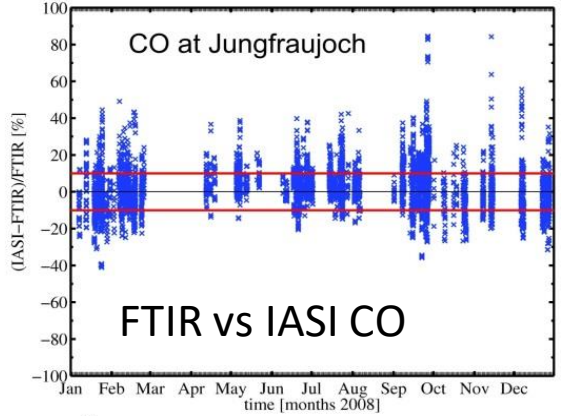


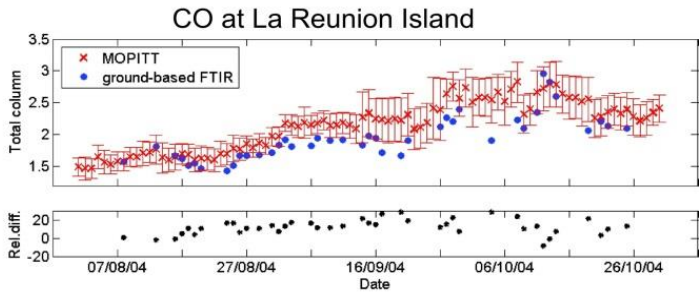




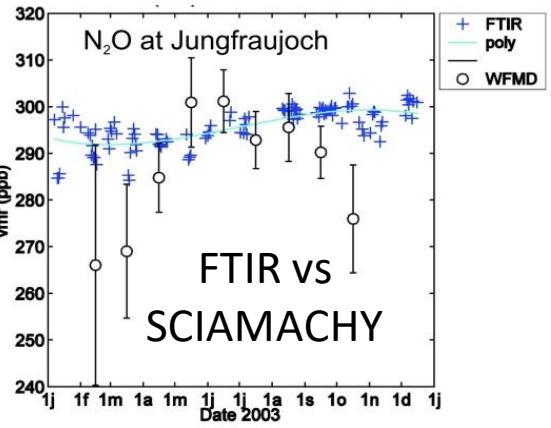


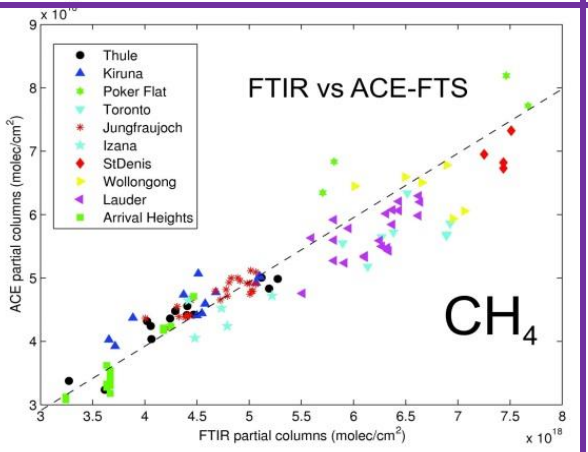


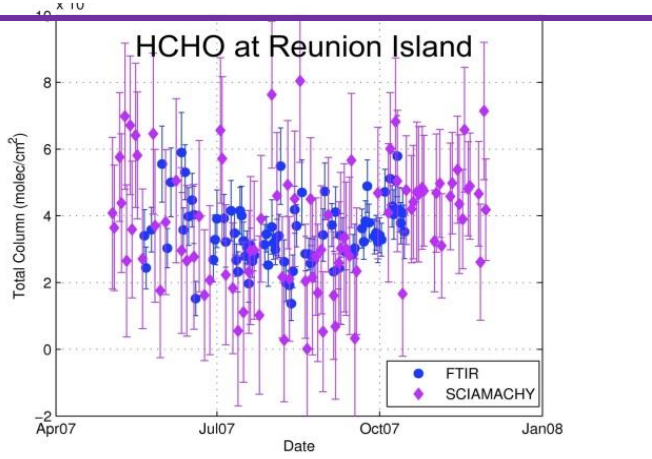


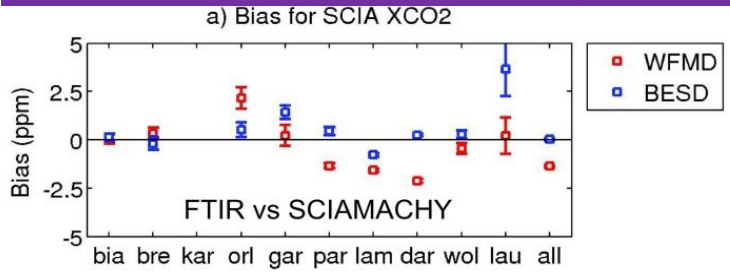


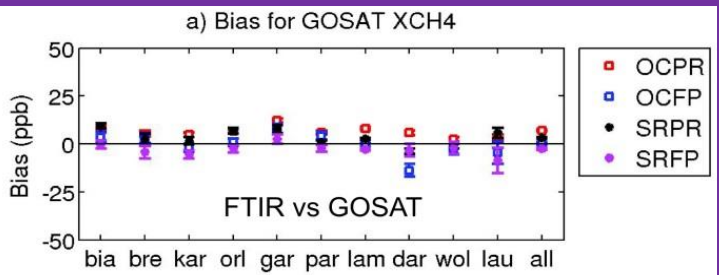
FTIR vs MOPITT

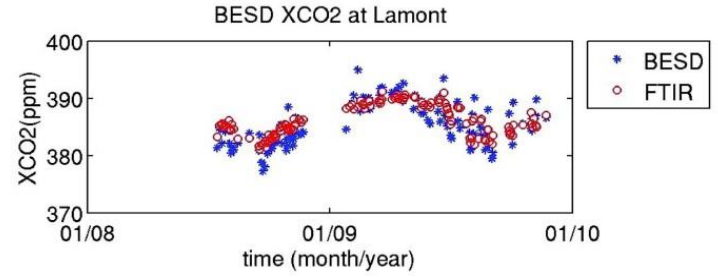


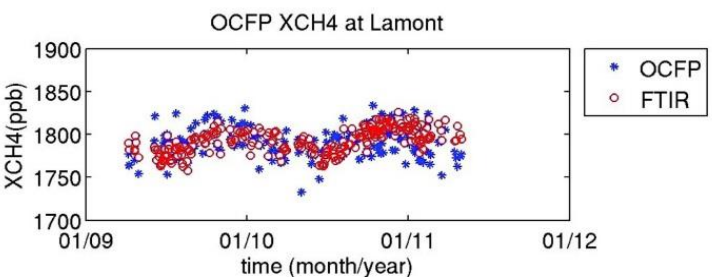






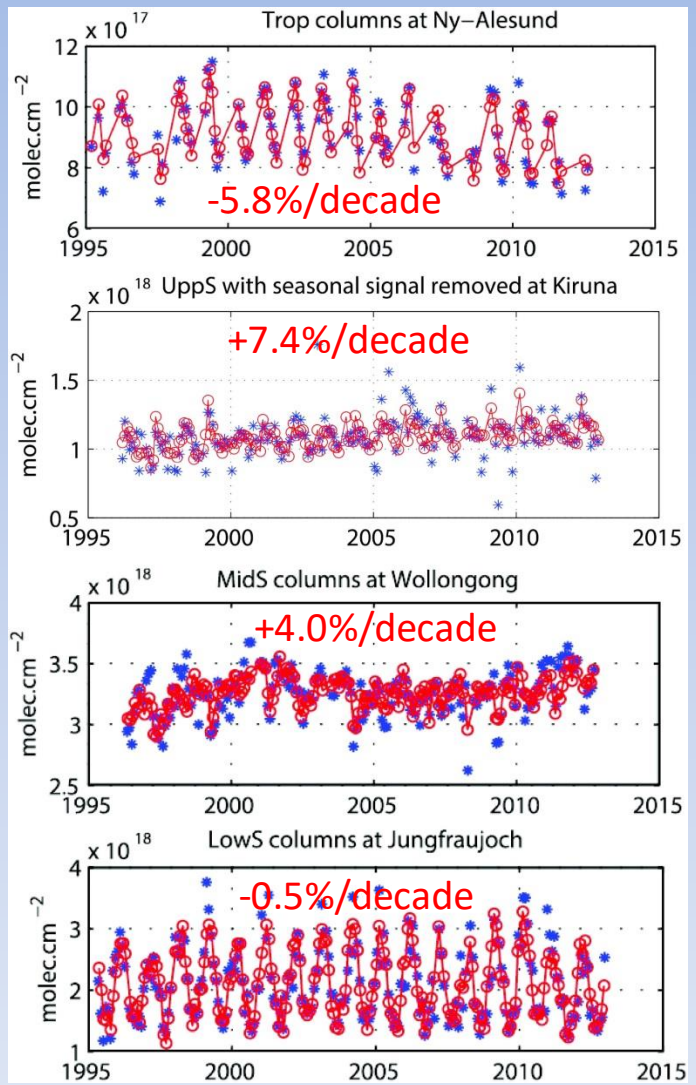






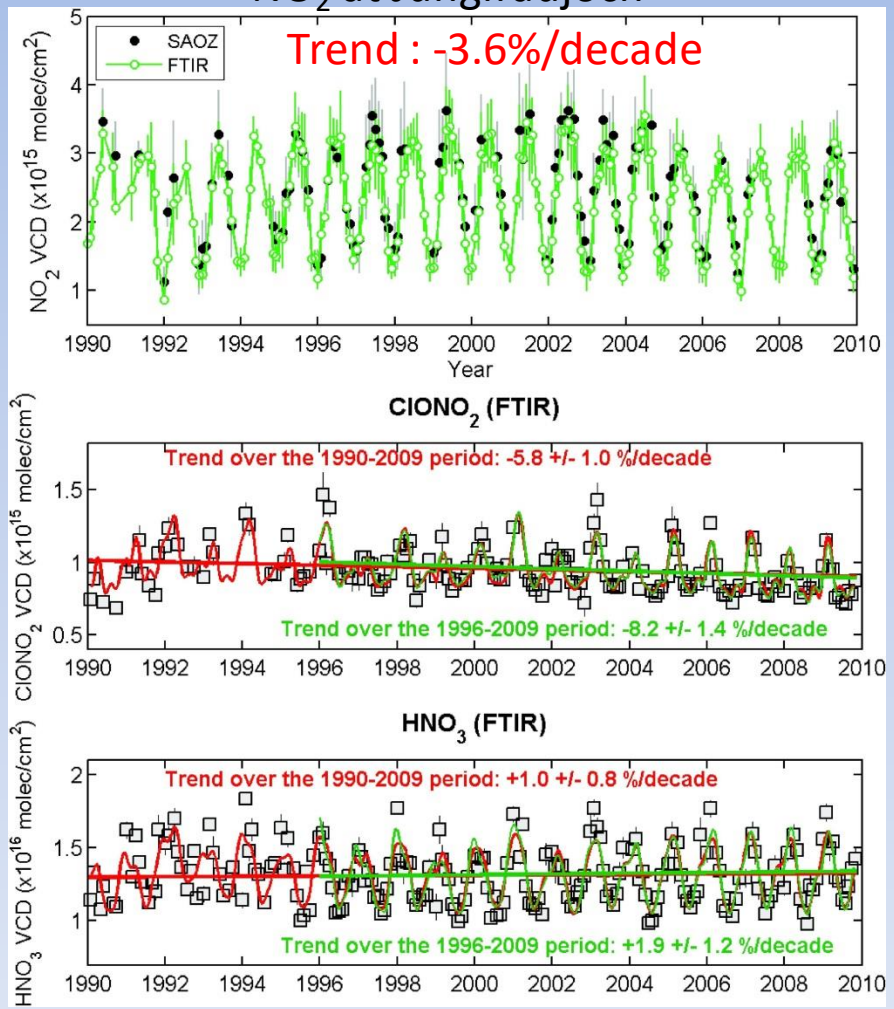
Dils et al., 2006
Kerzenmacher et al., 2012
Senten et al., 2008
De Mazière et al., 2008
Vigouroux et al., 2009
Dils et al., 2014

Long-term evolution of atmospheric composition



Vigouroux et al., ACPD, 2014

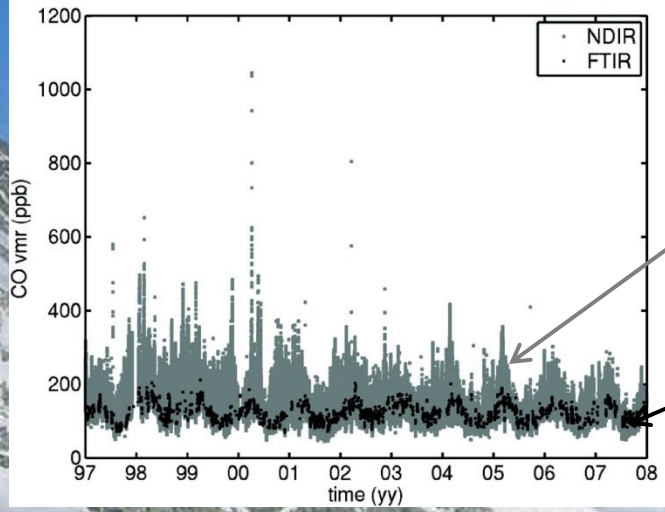
NO_2 at Jungfraujoch



Hendrick et al., ACP, 2012

CO at Jungfraujoch

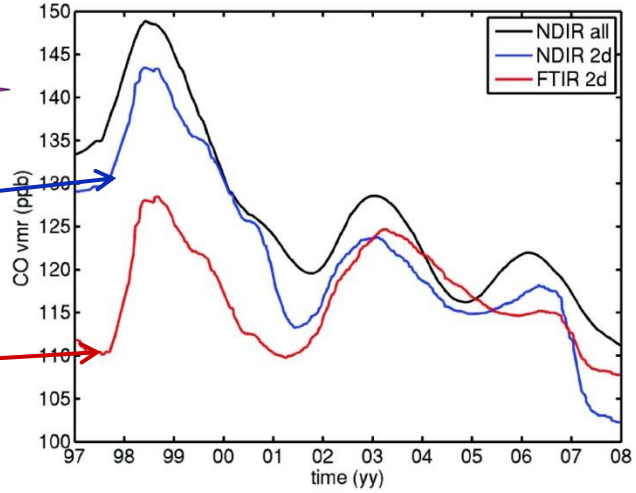
Dils et al., 2011



filtering

In situ (NDIR)

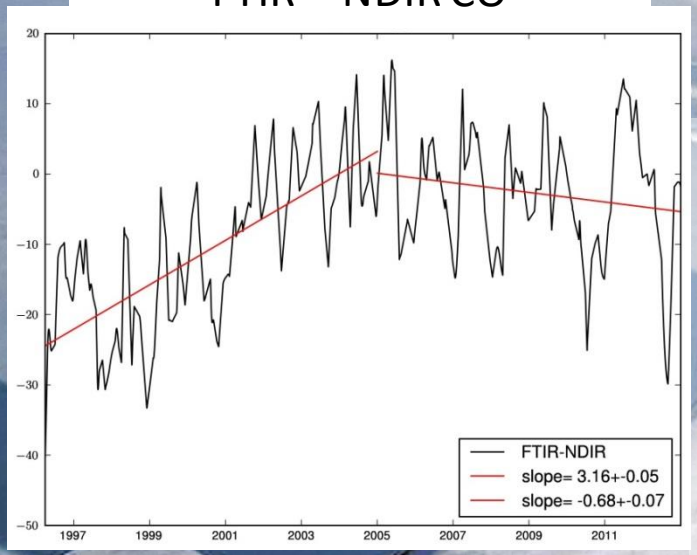
FTIR partial column, 3.6-7.2 km



In situ trend : -3.2 ppbv/year

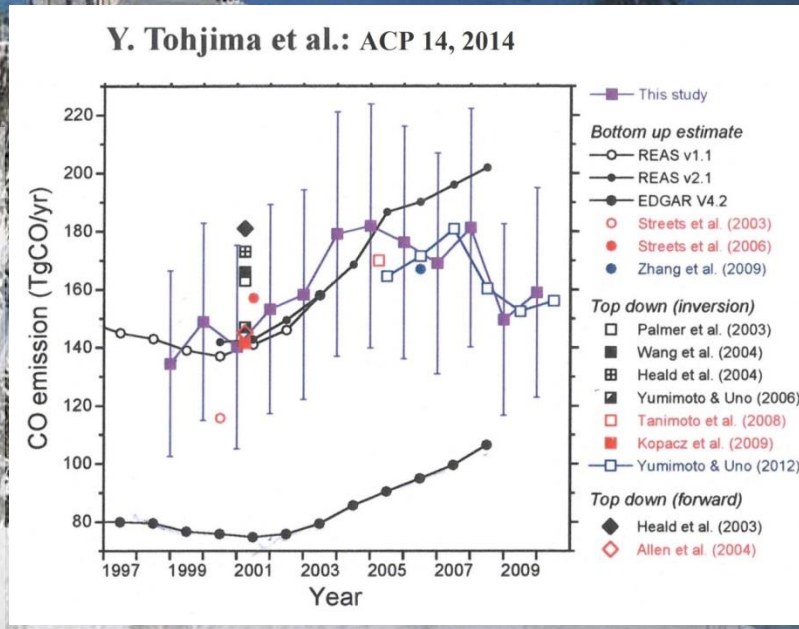
FTIR trend : -0.8 ppbv/year

FTIR – NDIR CO



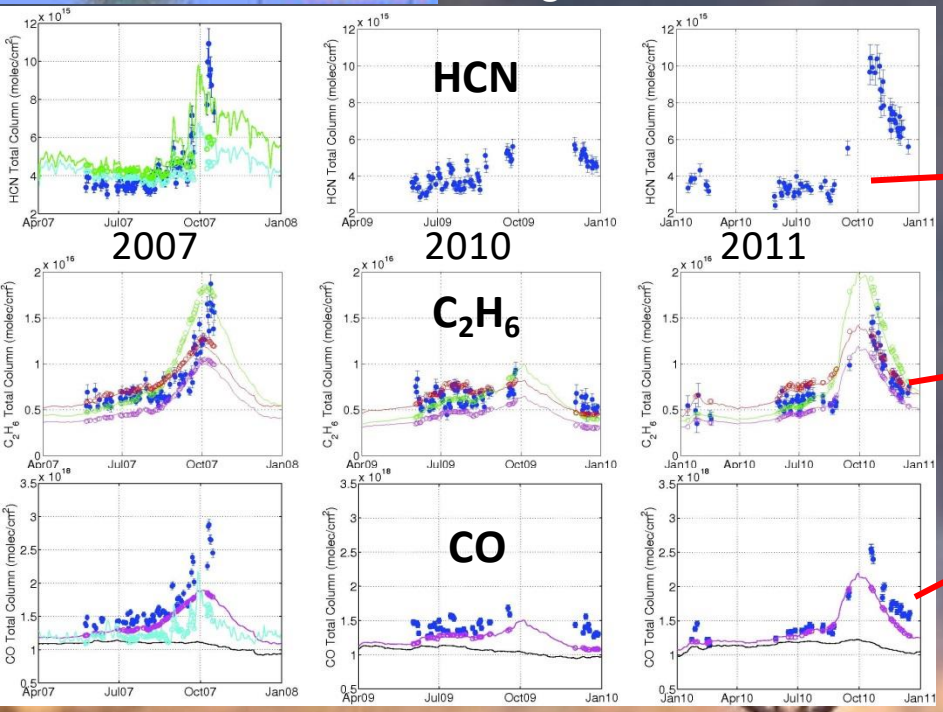
(updated timeseries, 1997-2012)

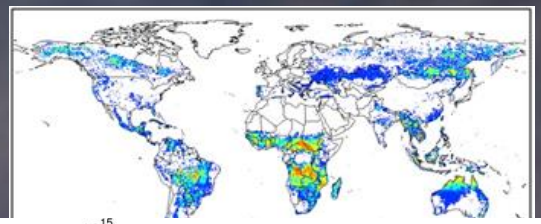
FTIR - NDIR difference : a marker for the evolution of Asian CO emissions ?

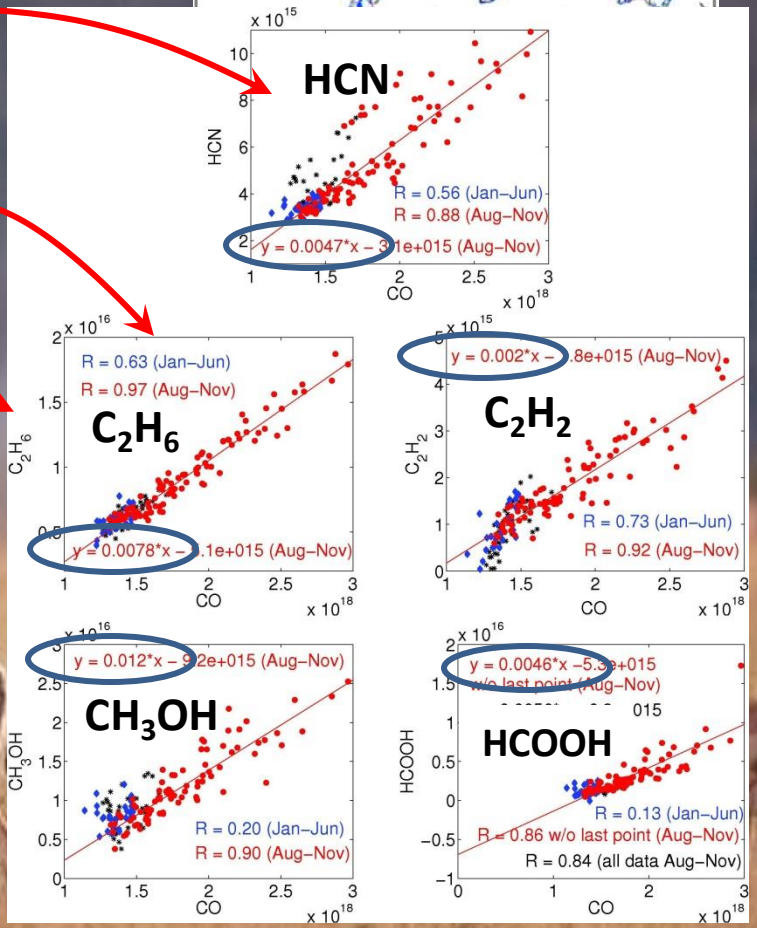


Biomass burning emission factors derived from FTIR measurements

Vigouroux et al., 2012

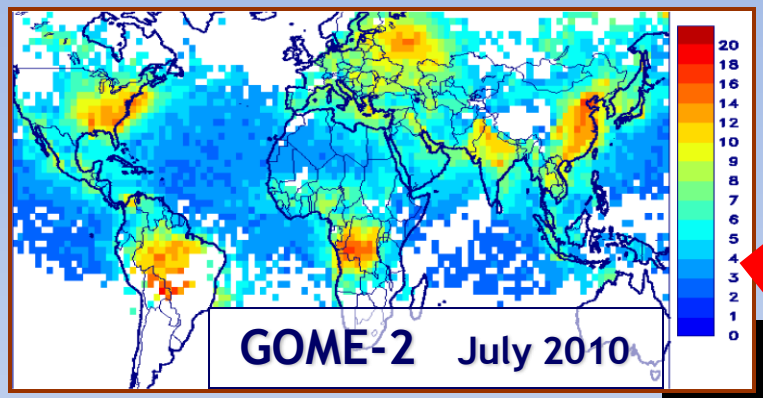


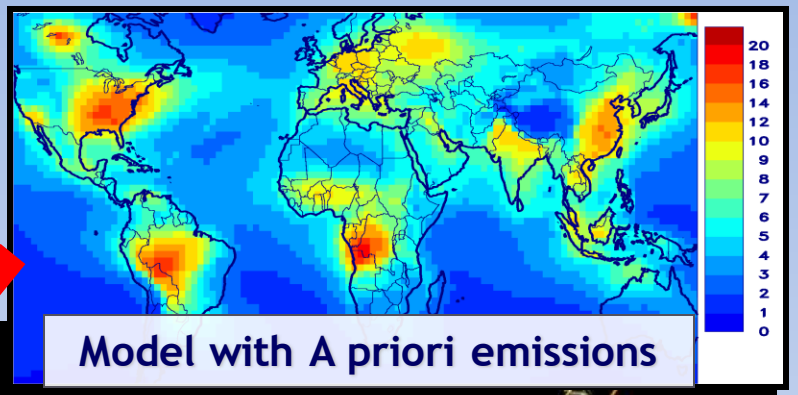


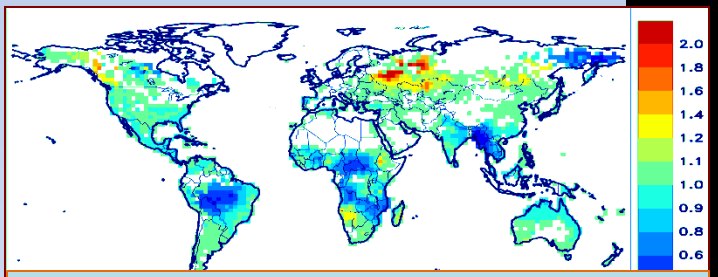


Improved emissions based on satellite observations

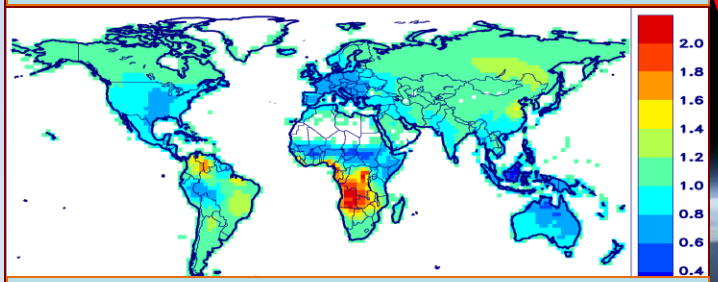
"Inverse modelling" : Example with formaldehyde (HCHO)



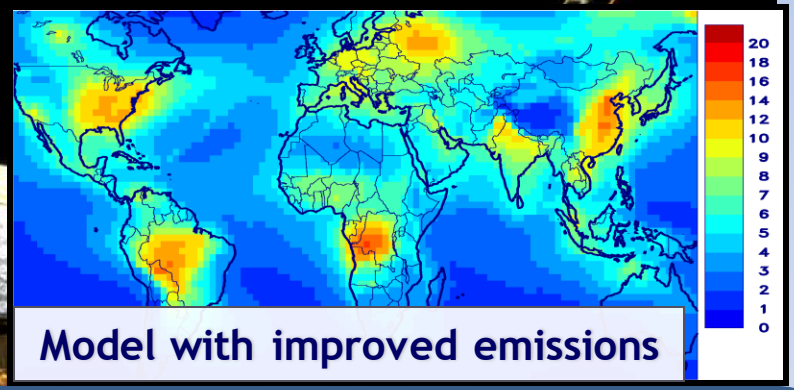


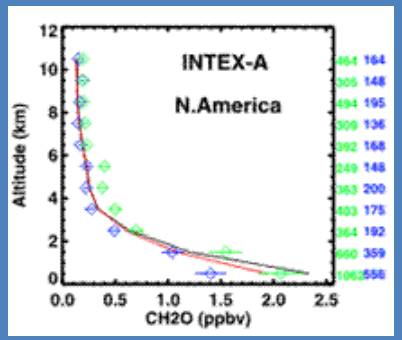


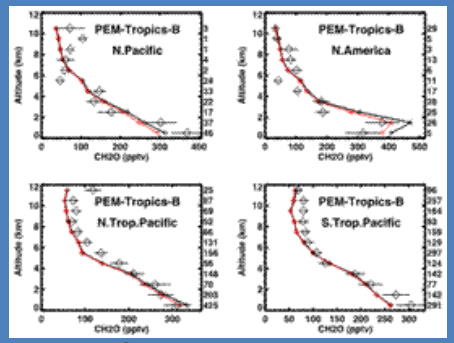
fire emission update



biogenic emission update



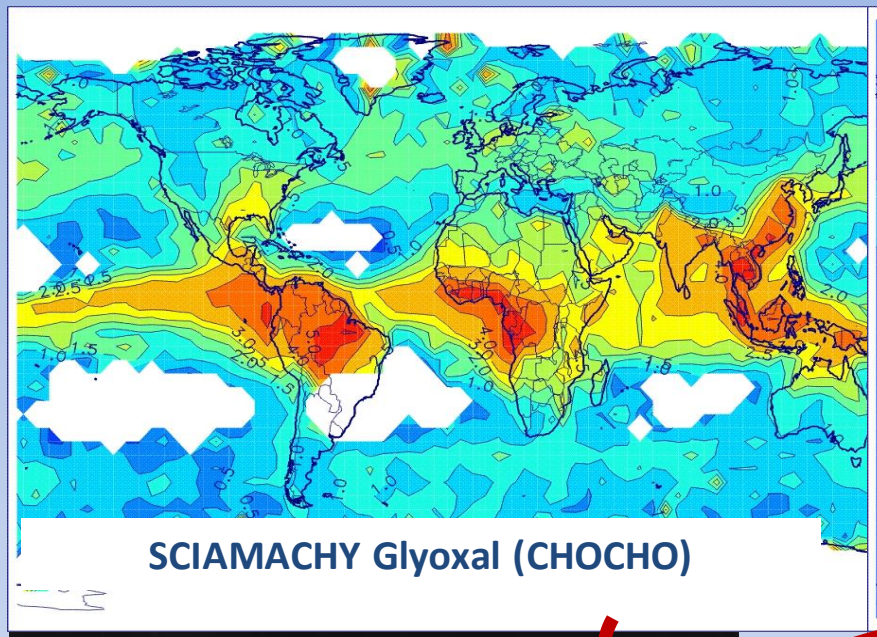


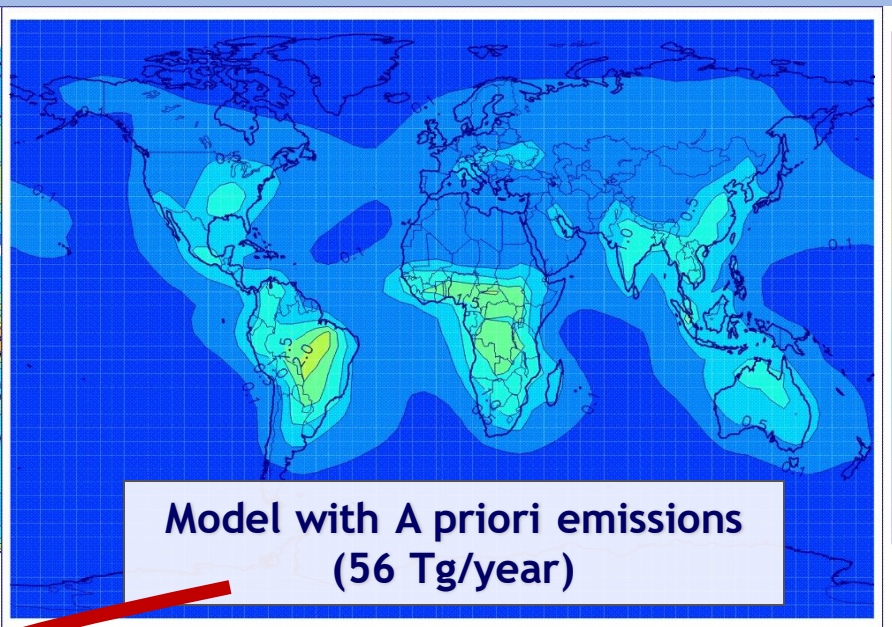


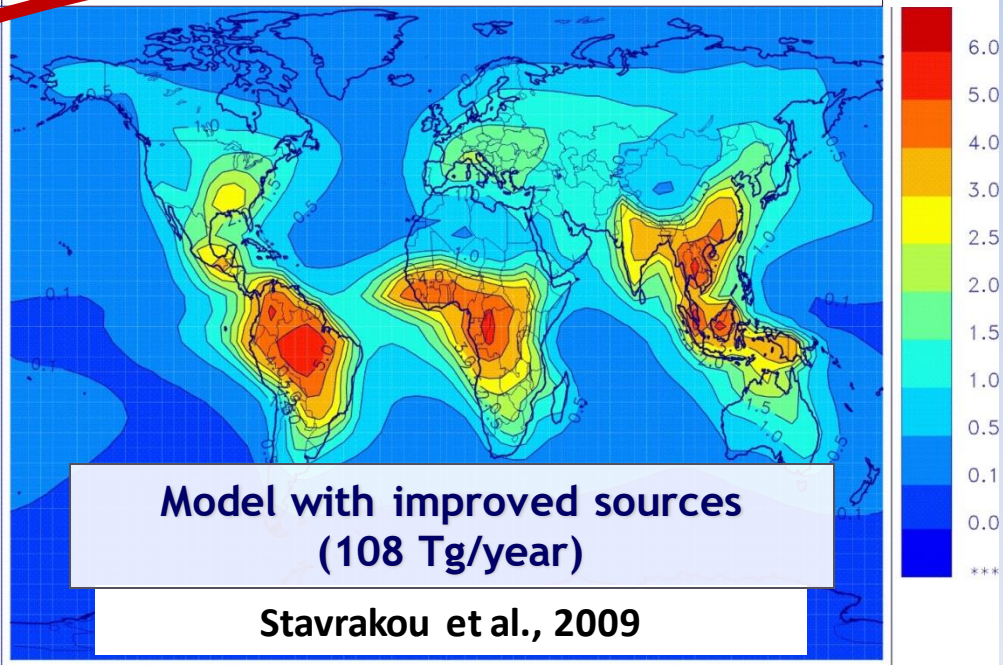
Verification using aircraft observations

Stavrakou et al., 2009, 2014

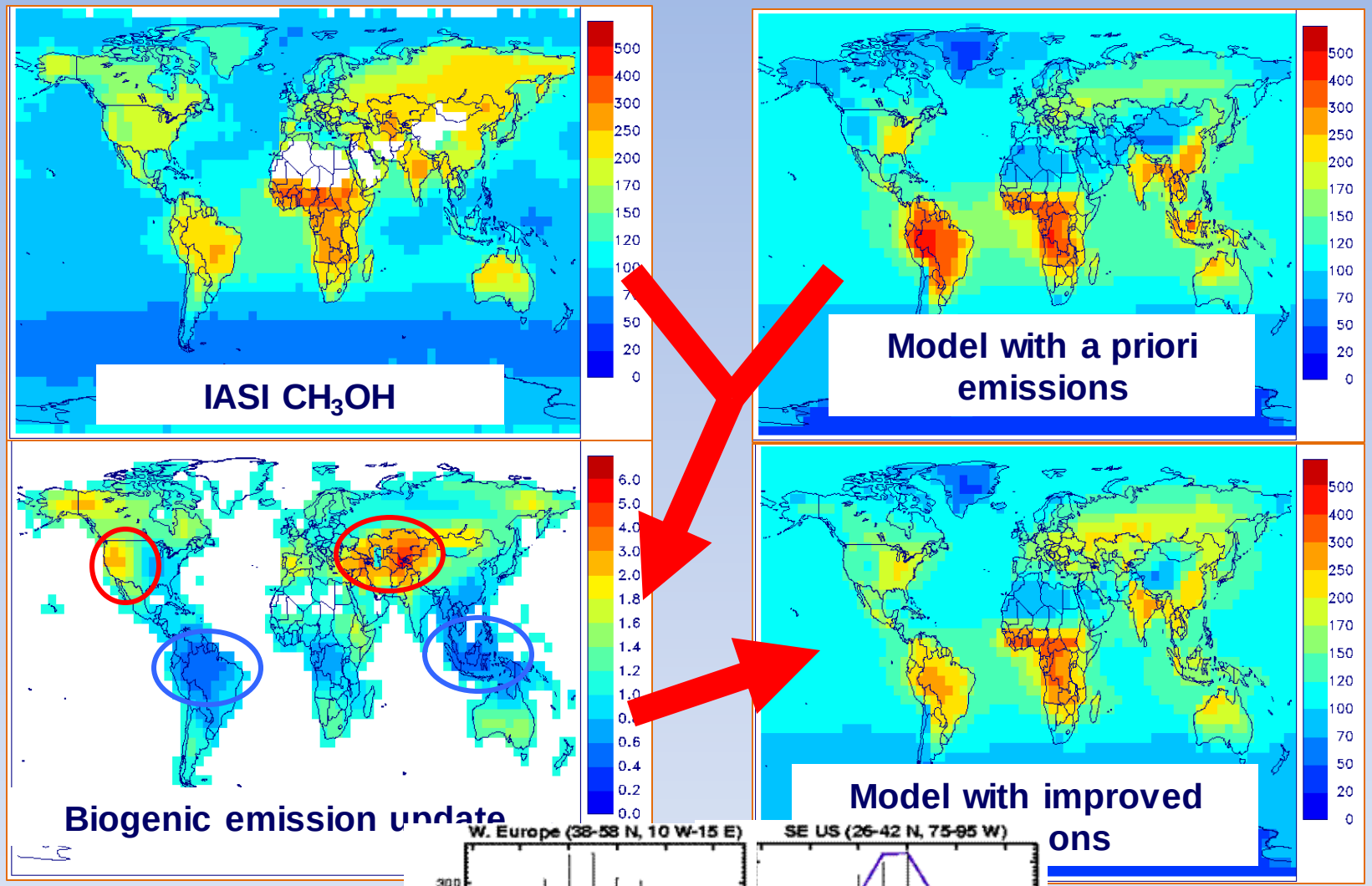
The continental source of glyoxal constrained by satellite

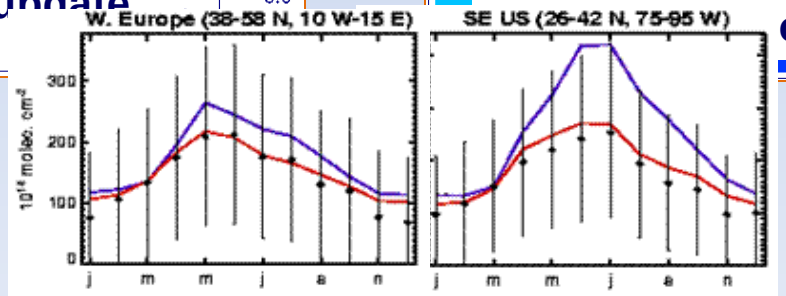






Methanol emissions constrained by satellite

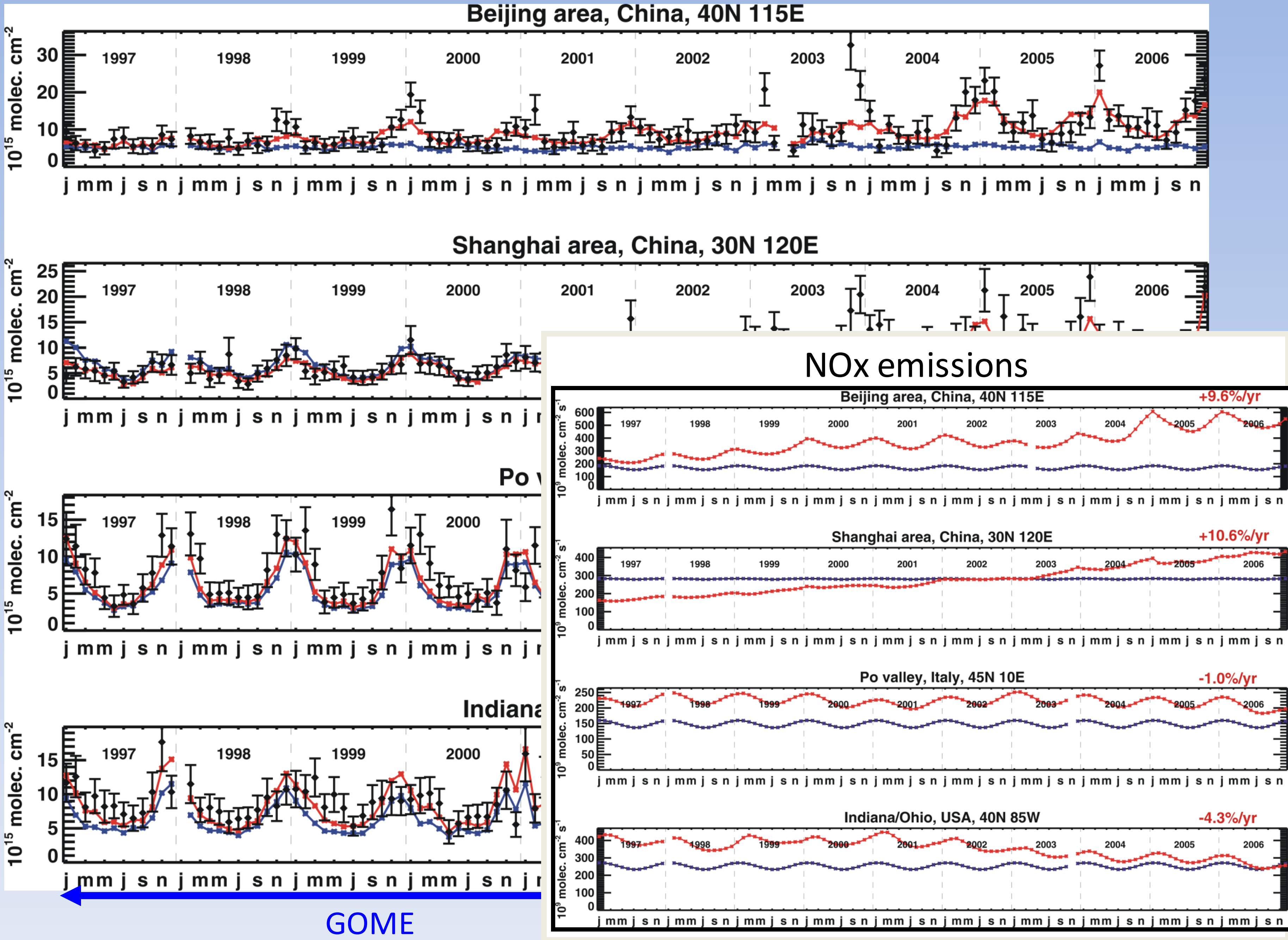




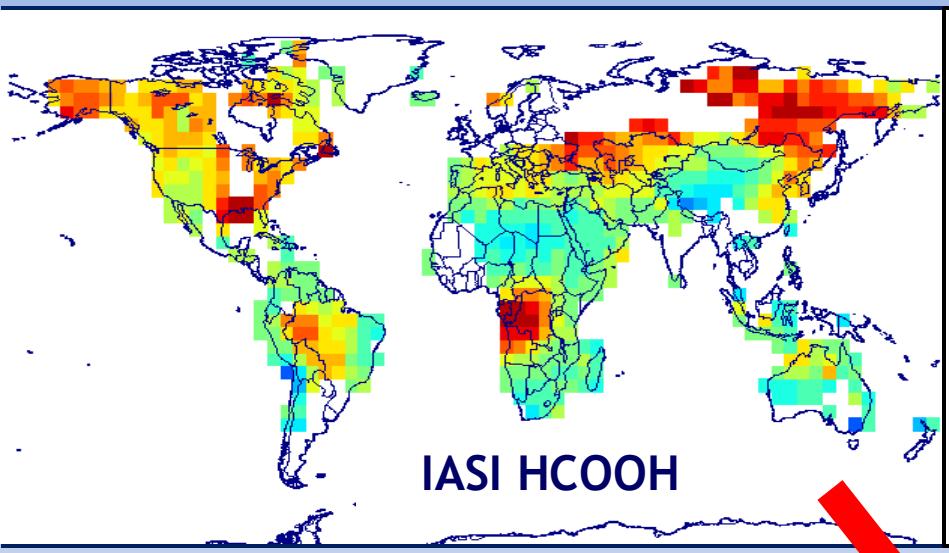
Biogenic emissions : springtime maximum

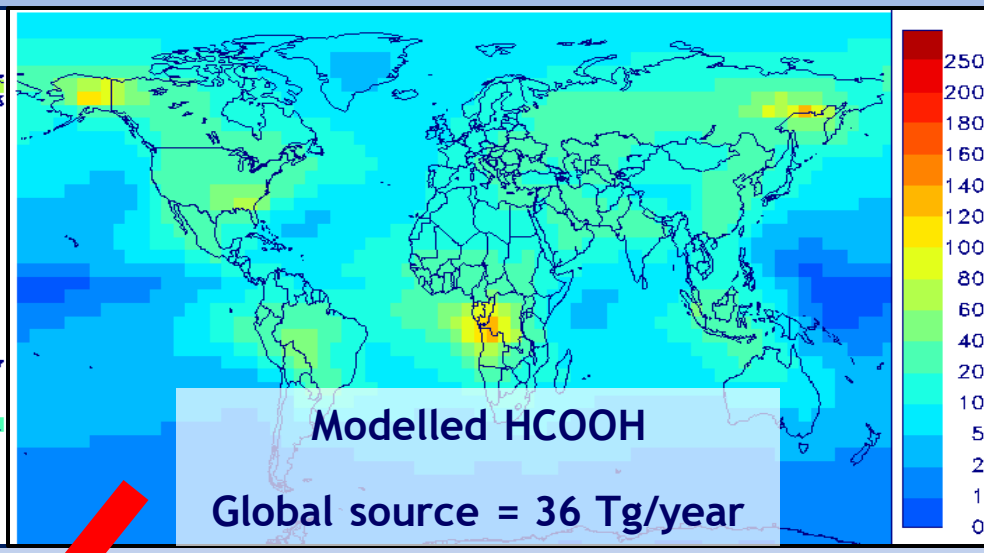
Stavrakou et al., 2011

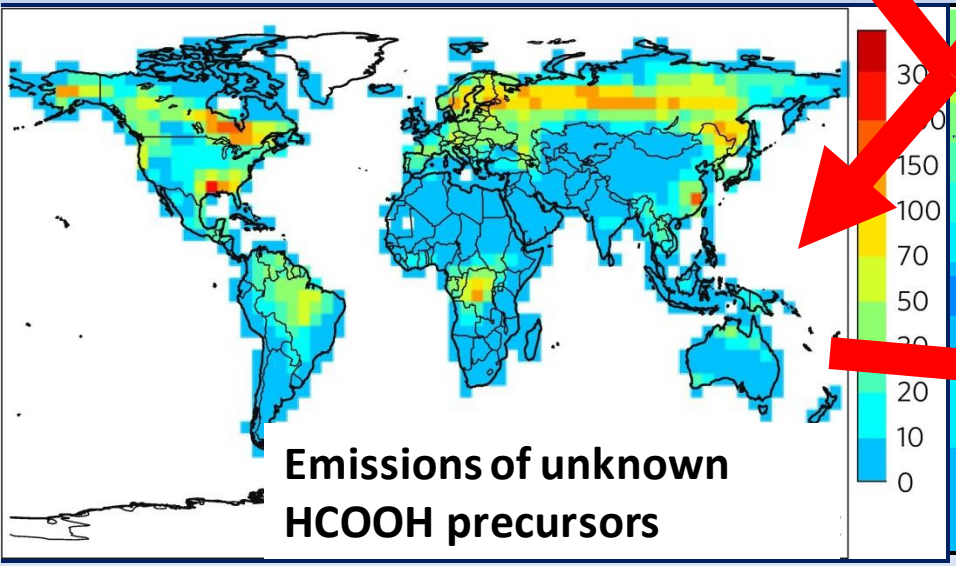
Observed vs. modelled NO_2 over heavily polluted areas

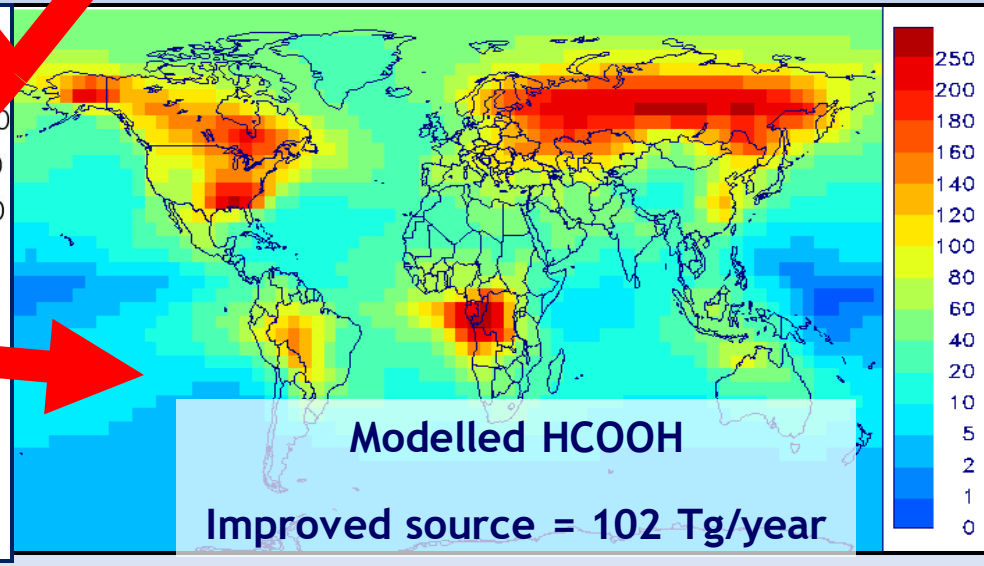


Formic acid : quantifying the missing source



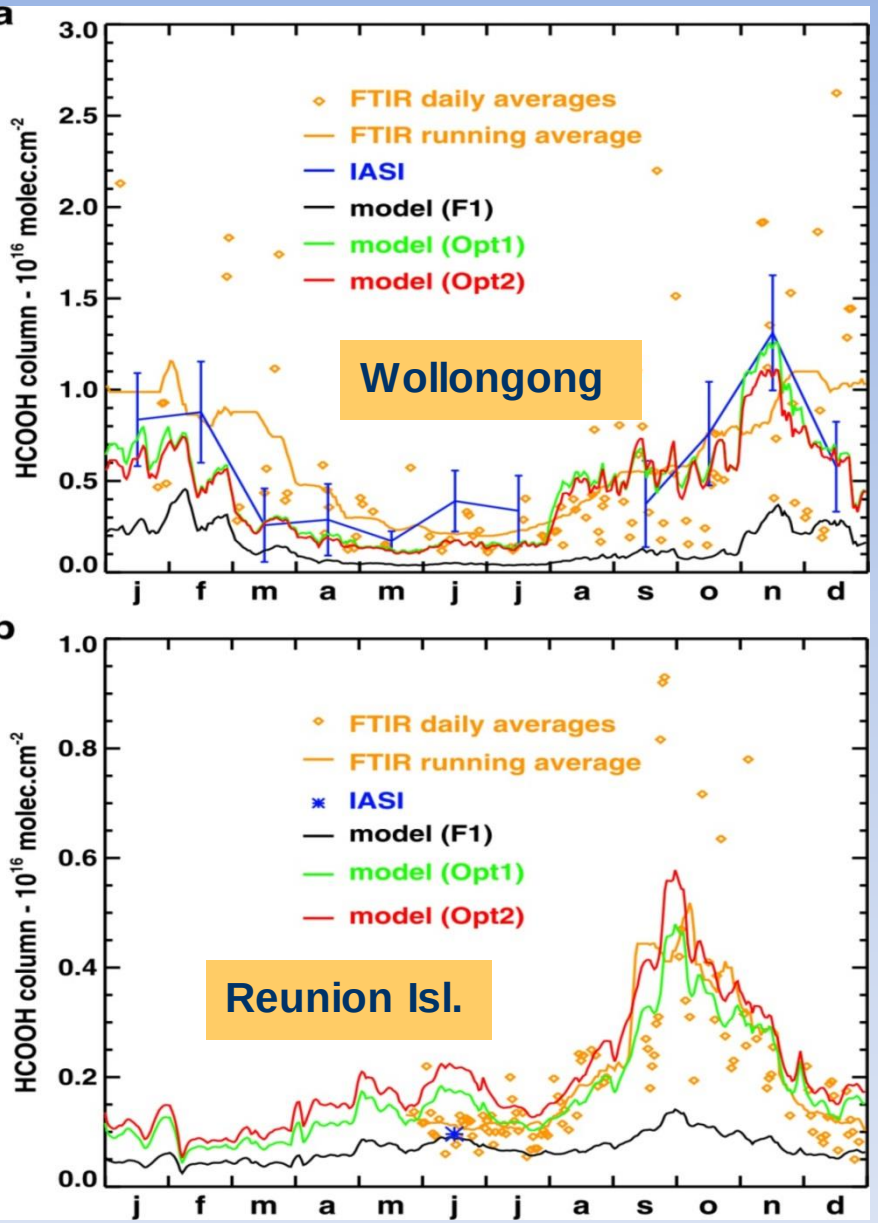






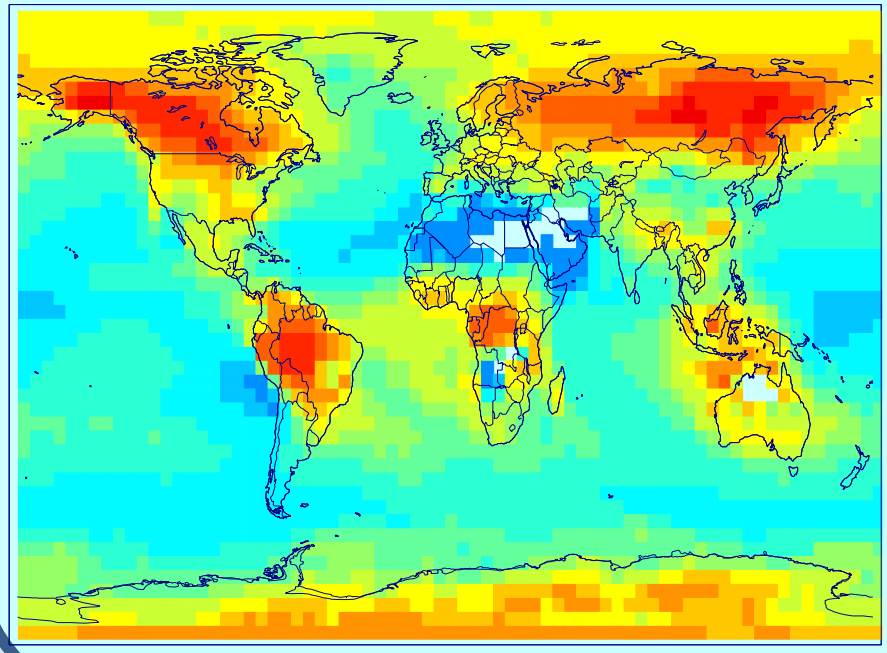
Large source over boreal forests likely reflects oxidation of BVOC from conifers

Stavrakou et al., Nature Geosci., 2012



Comparison with independent observations (e.g. FTIR) confirm the magnitude of the additional source

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




THANK YOU
FOR YOUR ATTENTION