# Oxygenated Compounds in the Tropical Atmosphere of the ICOS site of La Réunion Island


OCTAVE
Oxygenated Compounds in the Tropical Atmosphere : Variability and Exchanges

BIRA-IASB aeronomie.be

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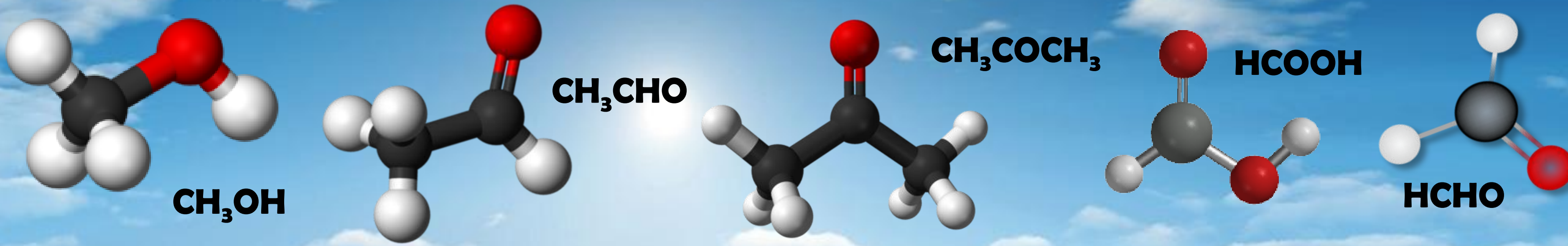
Belgian Science Policy Office belspo

ICOS BELGIUM

ICOS INTEGRATED CARBON OBSERVATION SYSTEM

Gembloux, 20 October 2017

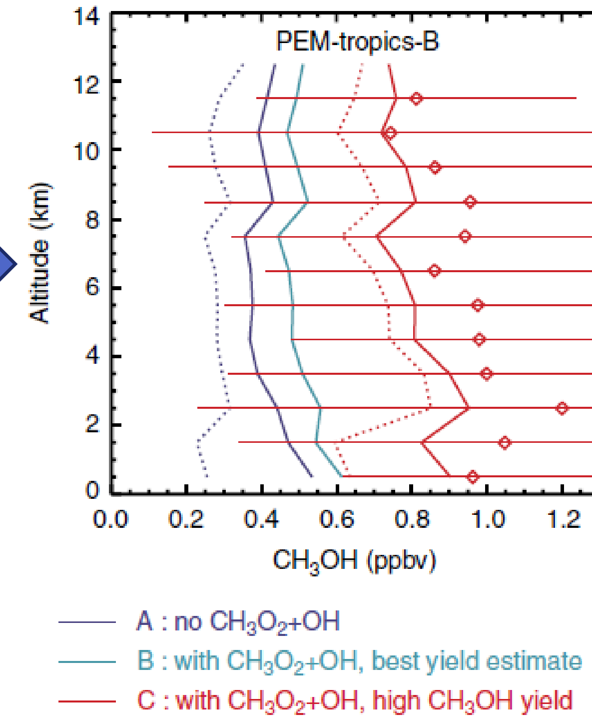
# Why OCTAVE?




OVOCs : Oxygenated volatile organic compounds

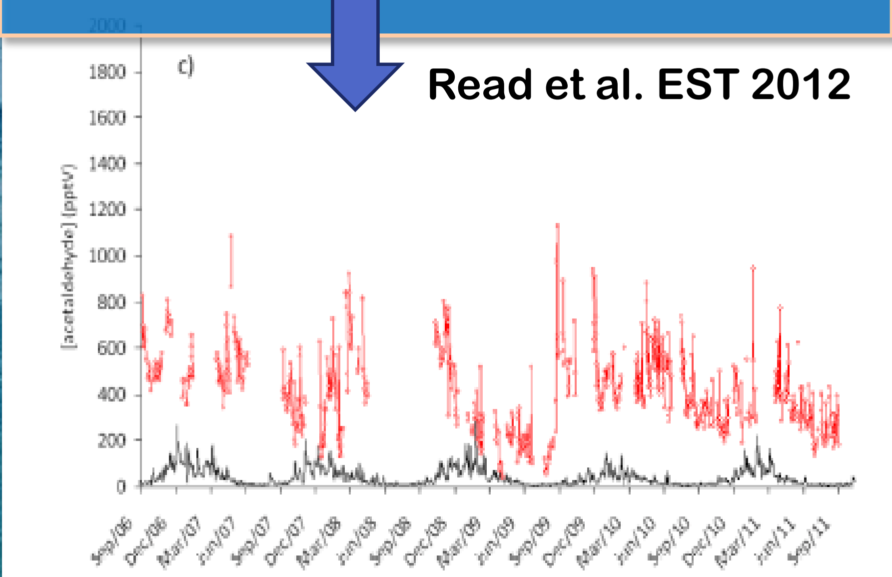
- **Significant impact on the atmospheric oxidative capacity & climate**
- **$CH_3OH$, $CH_3CHO$, $CH_3COCH_3$ : most abundant OVOCs, esp. in marine atmosphere**
- **Poor knowledge of sources due to paucity of observations**
- **$CH_3OH$ and HCOOH are exceptions thanks to satellite observations (IASI/MetOp), used to tie down estimates of their emissions over land** (Stavrakou et al. ACP, 2011, Wells et al. ACP, 2014, Stavrakou et al. 2012)

- New photochemical source of $CH_3OH$ through $CH_3O_2$+OH : explanation for persistent model underestimations in remote tropical oceans?




**Müller et al. Nature Comm. 2016**

- Very large, unexplained model understimation of observed $CH_3CHO$ in remote Tropics at surface and FT (Millet et al ACP 2010)




**Read et al. EST 2012**

- Observed $CH_3CHO$ incompatible with observed PAN (Millet et al. ACP 2010)

- Key difficulty: ocean/atmosphere exchanges of OVOCs (and precursors)

# Objective of OCTAVE

**Improve our appraisal of global budget of key OVOCs and their role in tropical regions, relying on an integrated approach combining in situ measurements, satellite retrievals and modelling**


**@ 2100 m a.s.l**

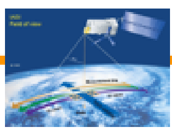
# OCTAVE *in the blink of an eye*

**WP1 (BIRA-IASB, CNRS)**
**Local (O)VOC measurements at Reunion Island**

- *T1.1* PTR-MS
- *T1.2* FTIR
- *T1.3* Campaign-based HCHO and GC-MS

**WP2 (BIRA-IASB, CNRS)**
**Analysis of local measurements**

- *T2.1* (O)VOC source apportionment
- *T2.2* Interpretation with Meso-NH model
- *T2.3* Intercomparison of local measurements



**WP3 (ULB, BIRA-IASB)**
**IASI retrievals of (O)VOCs**

- *T3.1* Neural network retrieval methodology
- *T3.2* Evaluation of retrieved (O)VOC total columns
- *T3.3* Decadal IASI record for methanol and other (O)VOCs

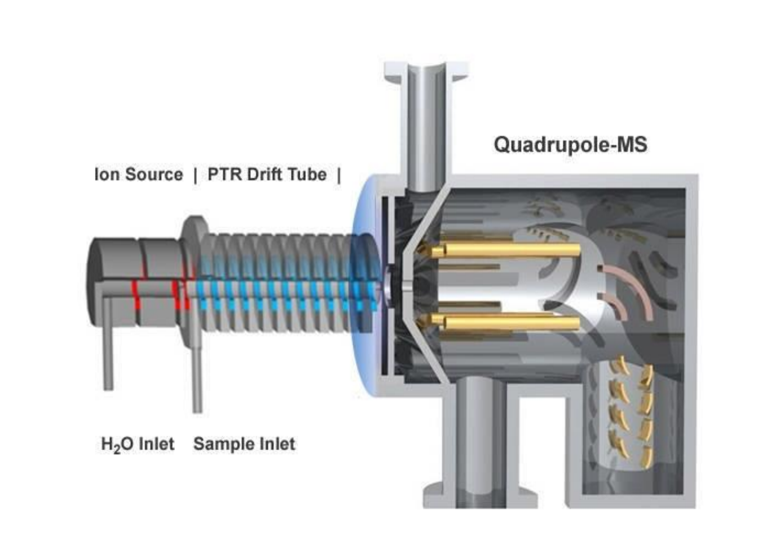
**WP4 (BIRA-IASB,CNRS,ULB)**
**Global modelling evaluation of OVOC budget and impacts**

- *T4.1* Model developments
- *T4.2* Global budget evaluation of acetaldehyde and acetone
- *T4.3* Revisiting the global methanol budget

**WP5 (BIRA-IASB, ULB, CNRS)** Management, outreach to the scientific community and to a broader public, interaction with follow-up committee

# Local measurements at Reunion

# PTR-MS measurements




- *in situ* technique, fast & sensitive, online
- well suited for long-term VOC monitoring
- no unambiguous detection of isomeric and isobaric VOCs
- 2-year continuous measurements → <u>**seasonal evolution of the contribution of VOC sources to the local air masses**</u>
- Simultaneous GC-MS (BIOMAIDO 2018)

| *Compound* | *m/z* | *Compound* | *m/z* |
|---|---|---|---|
| methanol | 33 | MVK/MACR | 71 |
| acetonitrile | 42 | MEK | 73 |
| acetaldehyde | 45 | benzene | 79 |
| acetone | 59 | toluene | 93 |
| DMS | 63 | xylene | 107 |
| isoprene | 69 | monoterpenes | 137 |

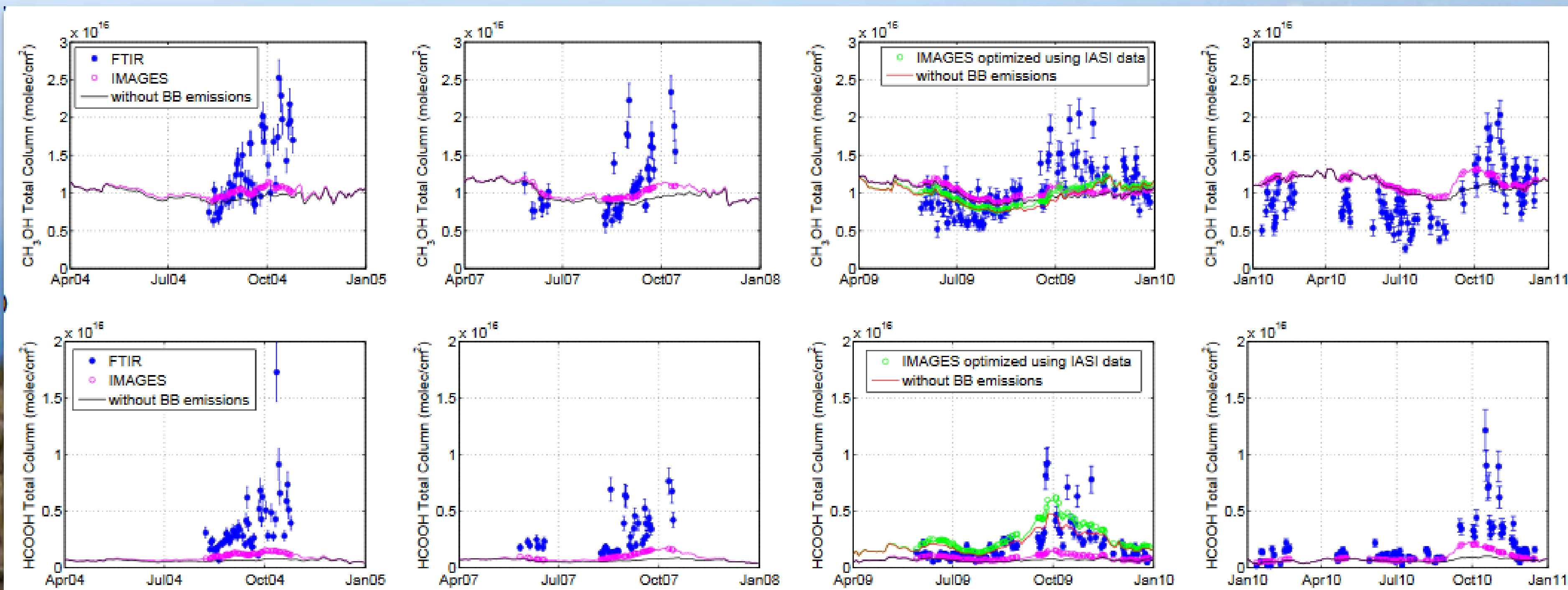



+ HCHO, acetic acid, PAN

# FTIR measurements

**BIRA-IASB operates FTIR instruments at La Réunion**

- St-Denis, since 2002     Maido, since March 2013




Vigouroux et al ACP 2012

- **FTIR at Maido (automated, remotely controlled)**
- $CH_3OH$, HCOOH, CO, HCHO, and $C_2H_2$ : total columns (+error+avk)
- **PAN, $C_2H_4$ and $CH_3COCH_3$ : challenging, but less contaminated spectra by water vapour at Maido & signal-to-noise ratio is much improved**

# Analysis of local measurements

✓ *Multivariate statistical analysis*

- EPA Unmix 6.0 : # source types, profiles, relative contributions, used for VOC source apportionment from PTR-MS data

✓ *Backtrajectory calculations*

- FLEXPART : large-scale transport @ 15 km (ECMWF), select air masses of marine origin, infer marine (O)VOC concentration/seasonal cycle

- Calculate concentrations for each set of trajectories → linearize transport btw surface and Maïdo
- 4D least-squares method (Brioude et al JGR 2011) → best estimates of surface fluxes of $CH_3OH$

- AROME forecasts at 2.5x2.5 km
- Meso-NH model at 500 m (case study)

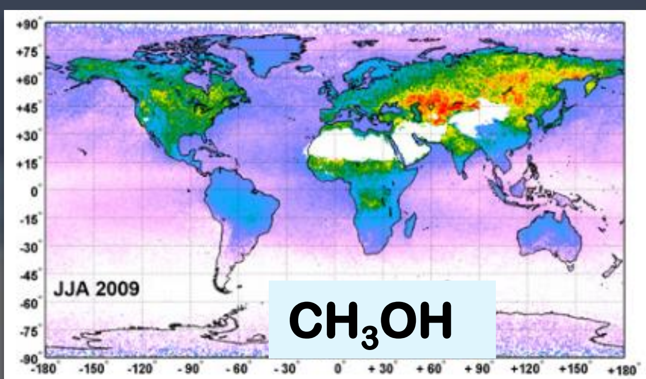
# IASI retrievals of (O)VOCs

**Retrieval algorithms for the (O)VOCs :**

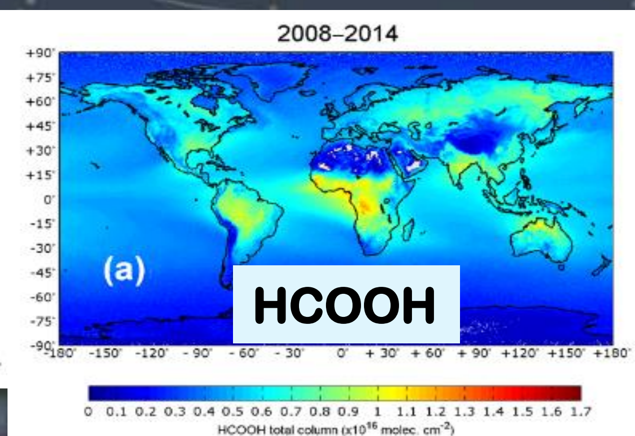
- $CH_3OH$, $C_2H_2$, $C_2H_4$, HCOOH (previously detected) or PAN (challenging)

**New method based on NN : gain in sensitivity, uncertainty estimates, computationally efficient, optimized retrievals over land/oceans**

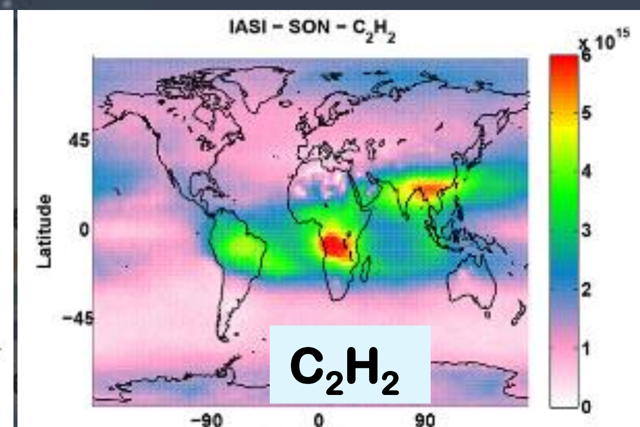
**Evaluation against ground-based observations & previous satellite retrievals**




Razavi et al ACP 2011




Pommier et al ACP 2016

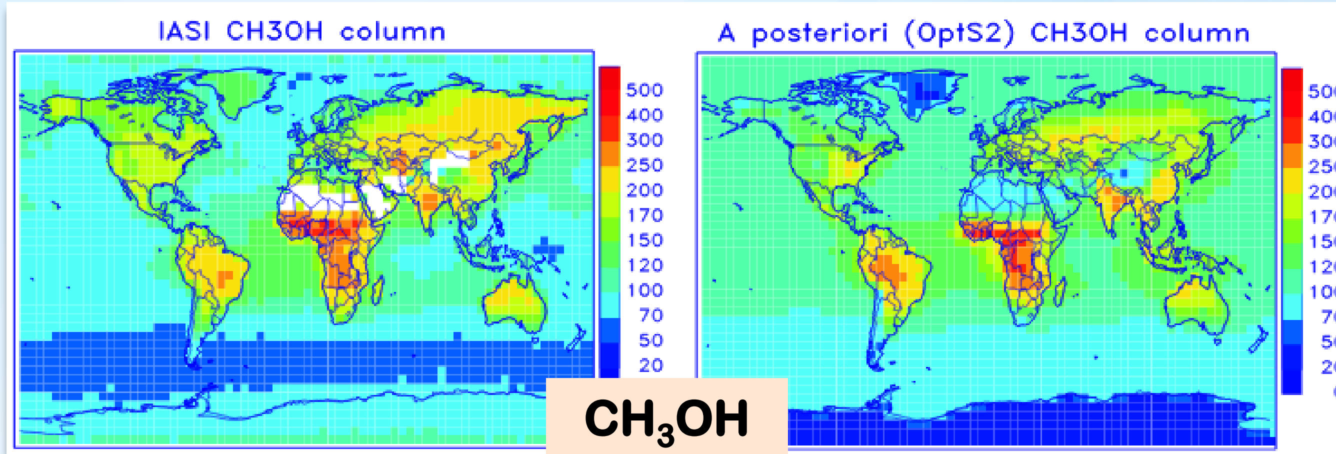



Duflot et al ACP 2015

**Decadal record (2008-2018) for $CH_3OH$, HCOOH, $C_2H_2$, $C_2H_4$ (& PAN)**

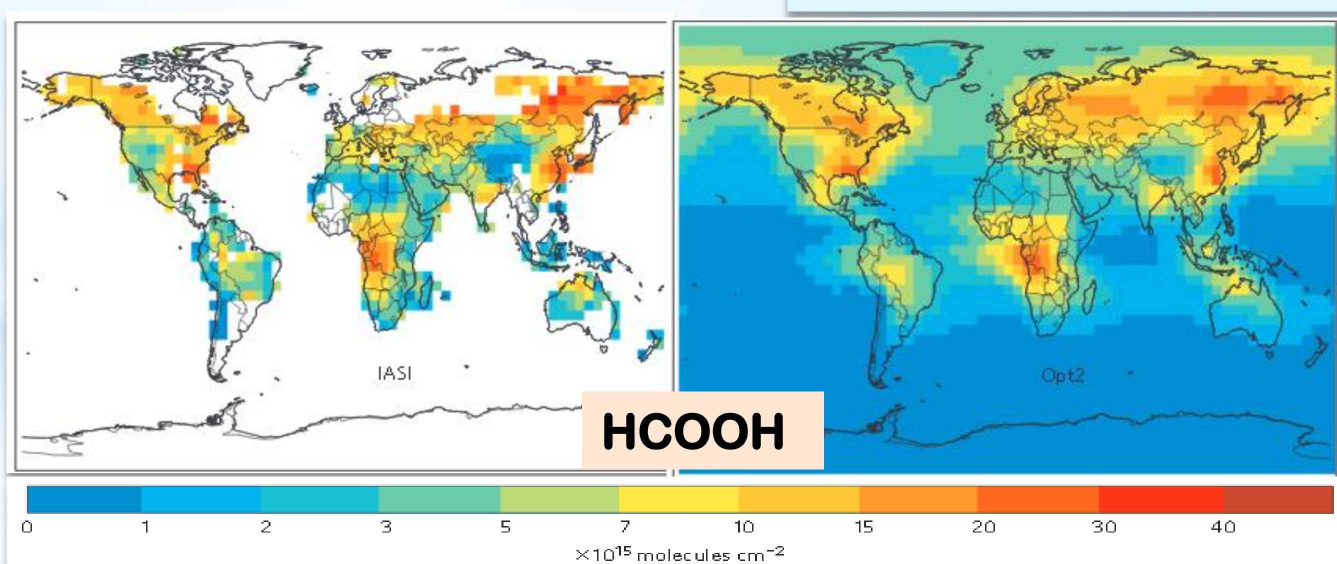
# Global modelling evaluation of OVOC budget and impacts

- IMAGESv2 global CTM
- Adjoint-based inversion scheme
- Used to infer $CH_3OH$ and HCOOH fluxes over land based on IASI
- Method will be improved and extended




$CH_3OH$

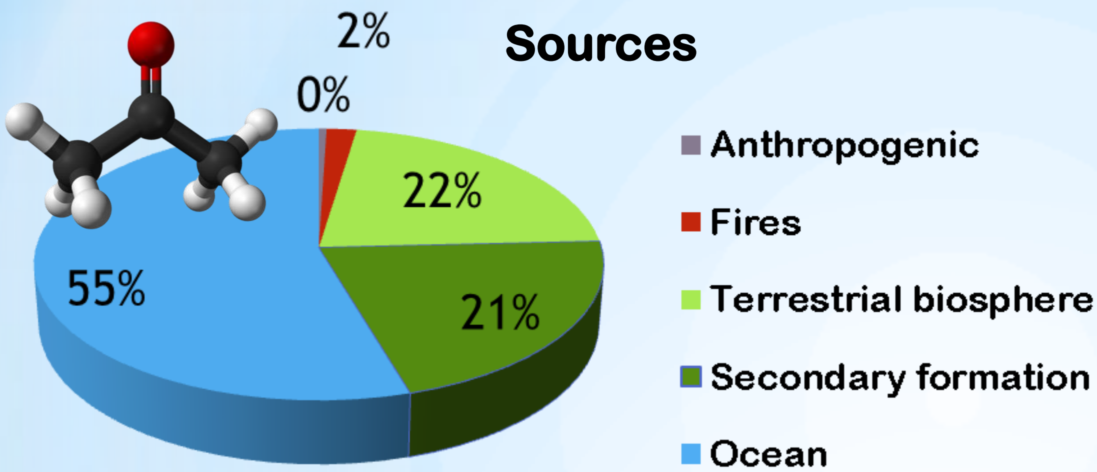
Stavrakou et al ACP 2011

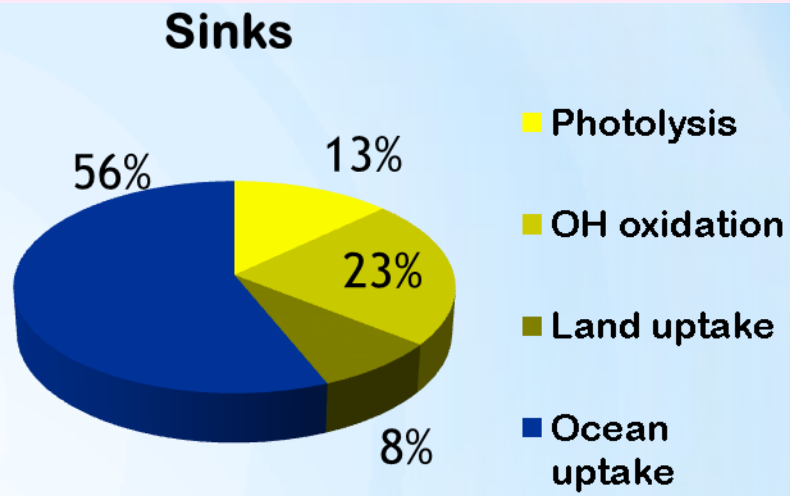



HCOOH

Stavrakou et al NGeo 2012

# Our « *best* » knowledge on OVOC budget




Fischer et al GRL 2012

**Global source : 147 Tg/yr**
**Lifetime = 1 month**

Biogenic flux : 32 Tg/yr (range : 20-172), first study accounting for oceanic flux

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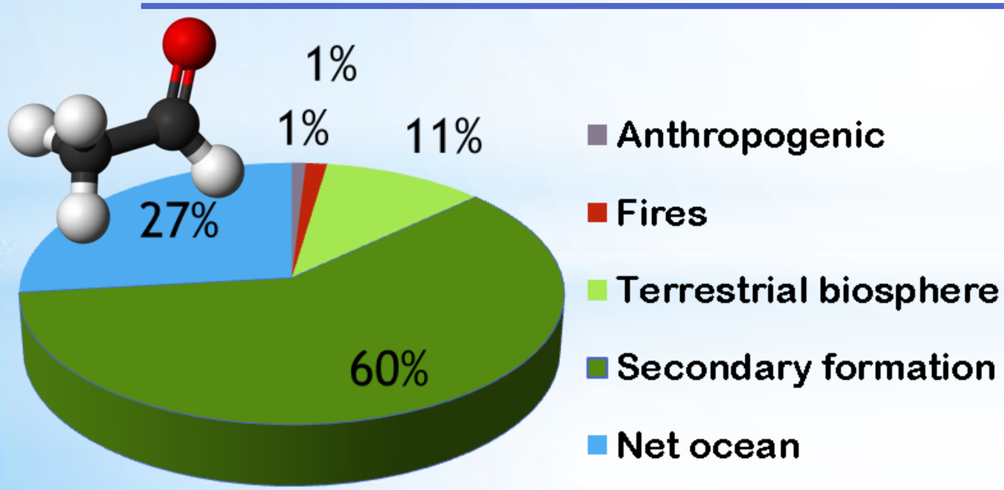
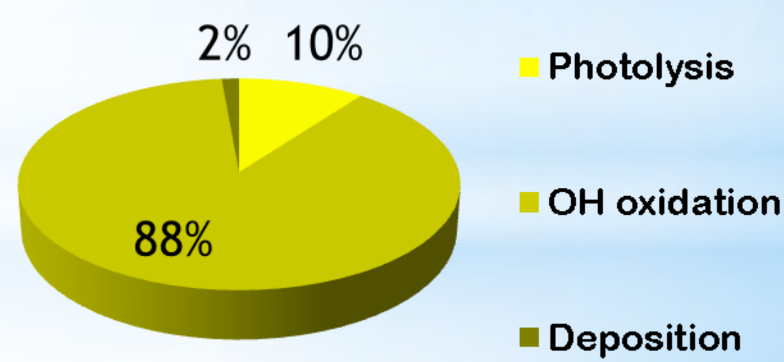
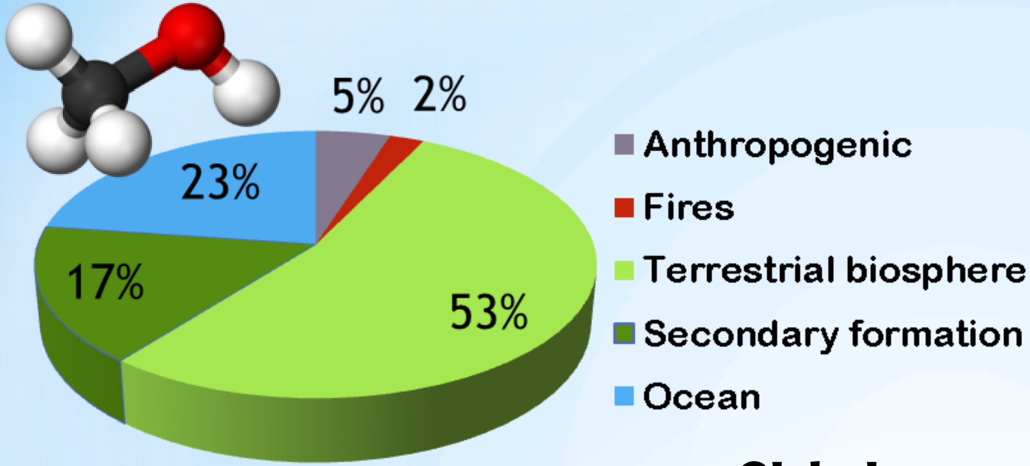


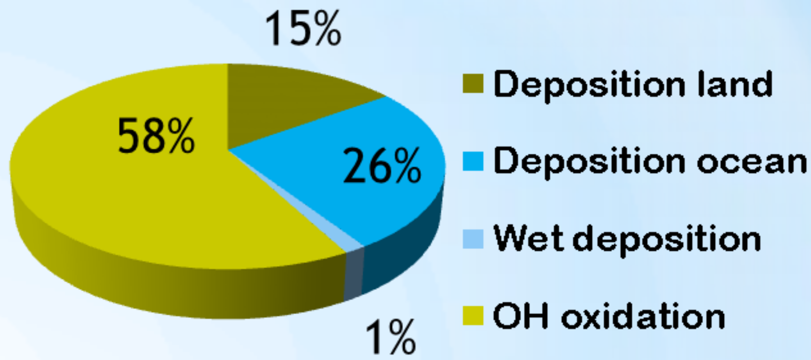




Millet et al ACP 2010

**Global source : 213 Tg/yr**
**Lifetime ~1 day**

Compared to Singh et al JGR 2004:
- x2.5 lower ocean flux
- x4 higher secondary production




Stavrakou et al ACP 2011

**Global source : 187 Tg/yr**
**Lifetime ~6 days**

# Key questions

- ✓ Can we explain the elevated $CH_3CHO$ levels over remote tropical ocean? Are there unknown oceanic or biogenic $CH_3CHO$ precursors?
- ✓ What are the contributions of $CH_3CHO$, $CH_3COCH_3$ and other OVOCs to PAN formation?
- ✓ Can we confirm and better constrain the importance of the $CH_3O_2$+OH reaction as major remote source of $CH_3OH$?
- ✓ Can we better constrain the biosphere/atmosphere and ocean/atmosphere exchanges of $CH_3CHO$, $CH_3OH$, $CH_3COCH_3$?
- ✓ What is the overall impact of OVOCs on atmospheric oxidants (esp. OH)?

http://octave.aeronomie.be

OCTAVE

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Thanks for your attention