The South Asian monsoon— pollution pump and purifier

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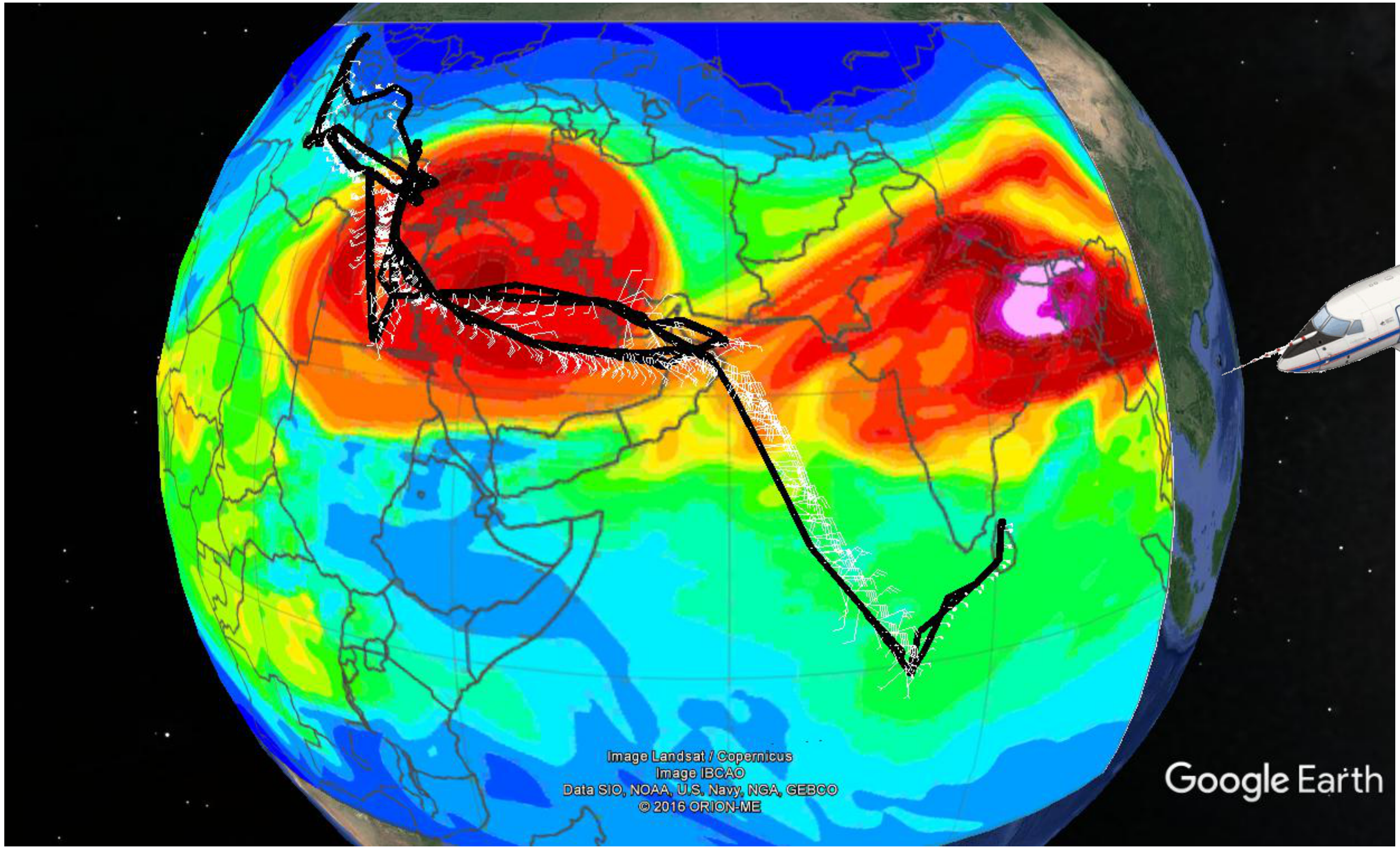
OMO – Oxidation Mechanism Observation

KIT Karlsruher Institut für Technologie
DLR
JÜLICH FORSCHUNGSZENTRUM
DFG Deutsche Forschungsgemeinschaft
MAX-PLANCK-INSTITUT FÜR CHEMIE



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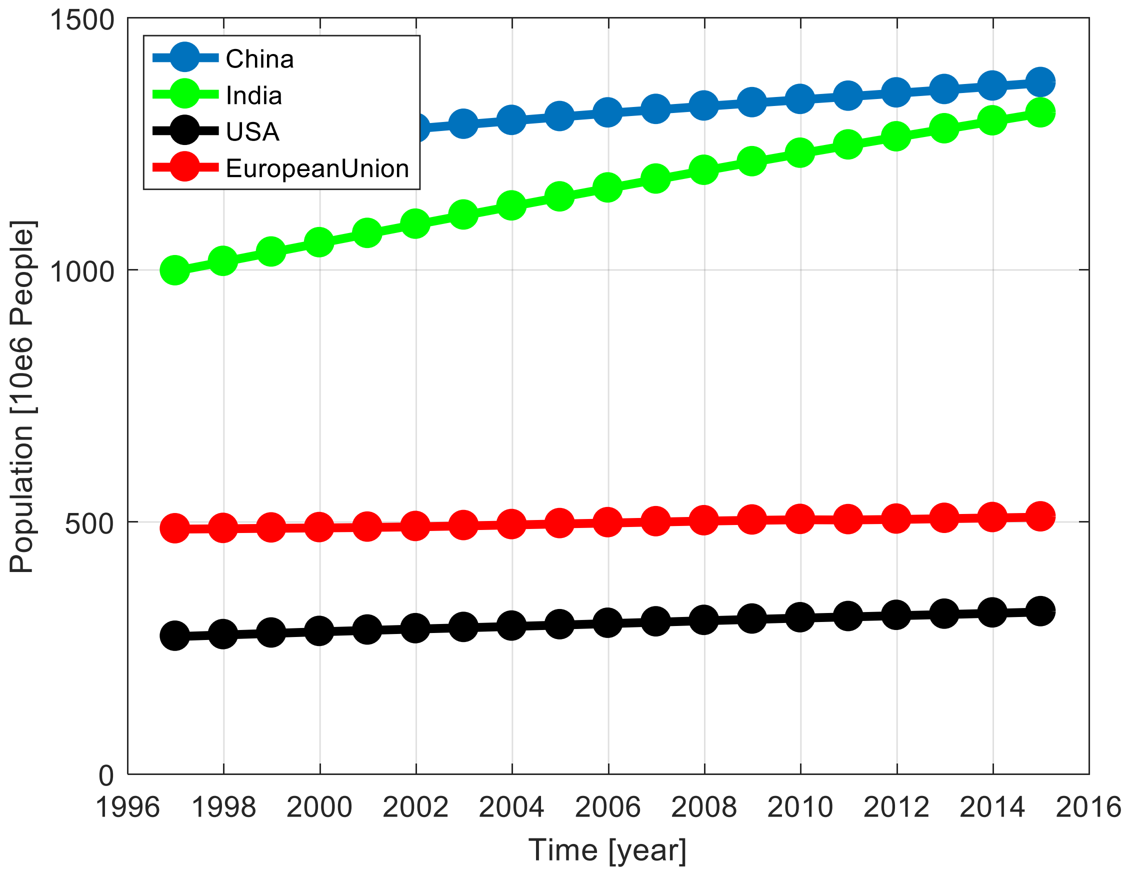
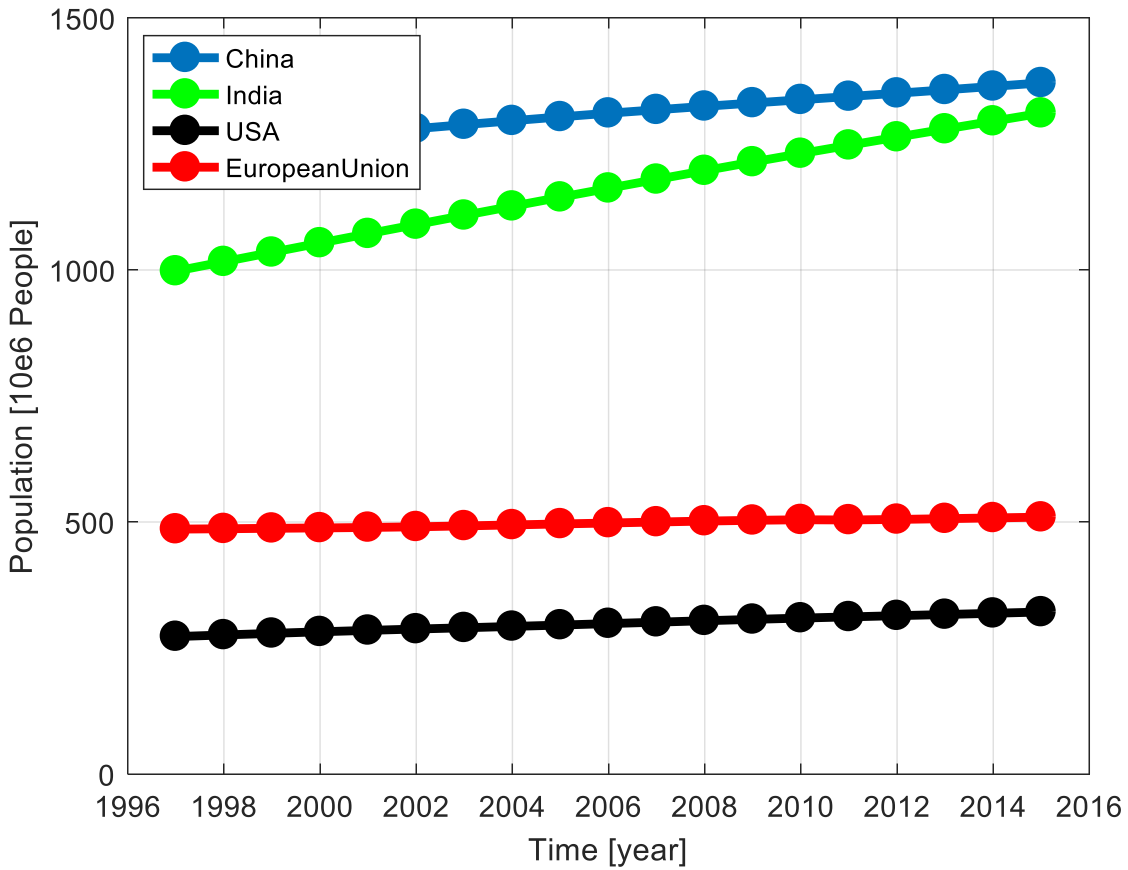
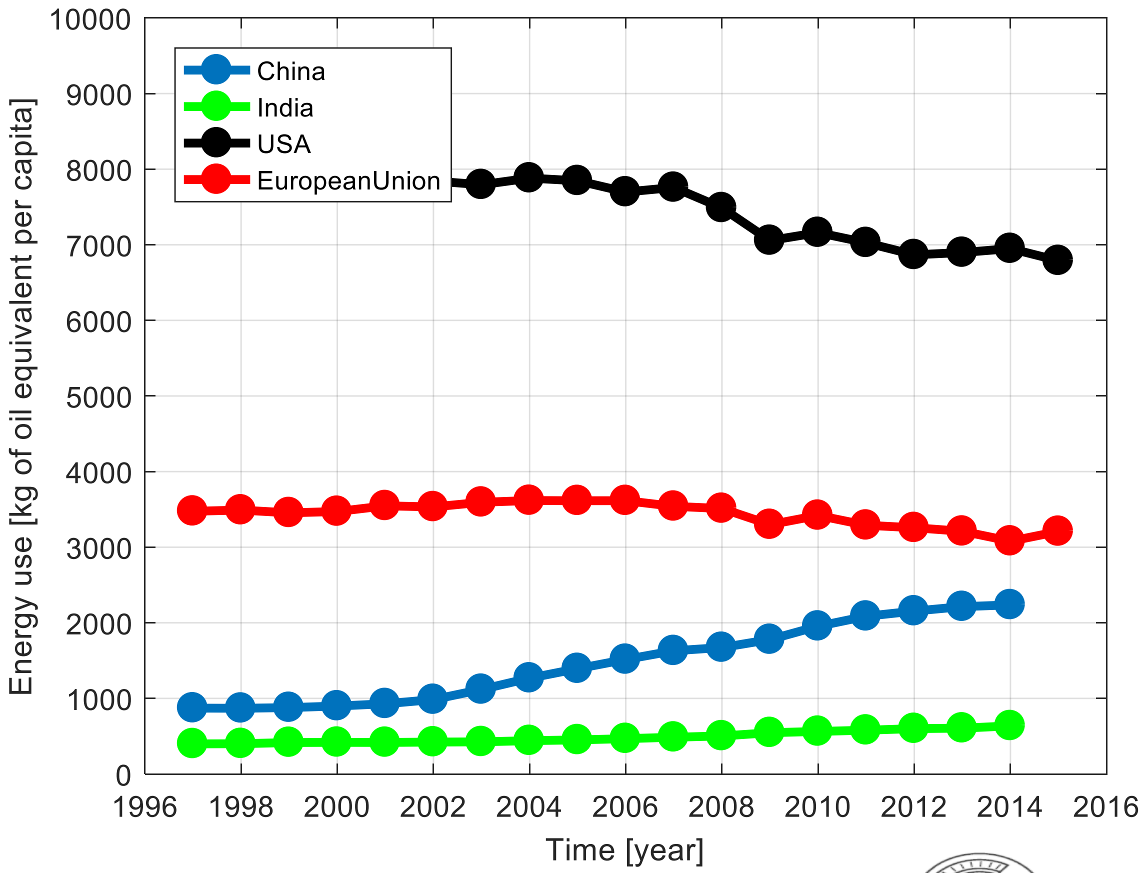
Region of growth



International Energy Agency & World Bank

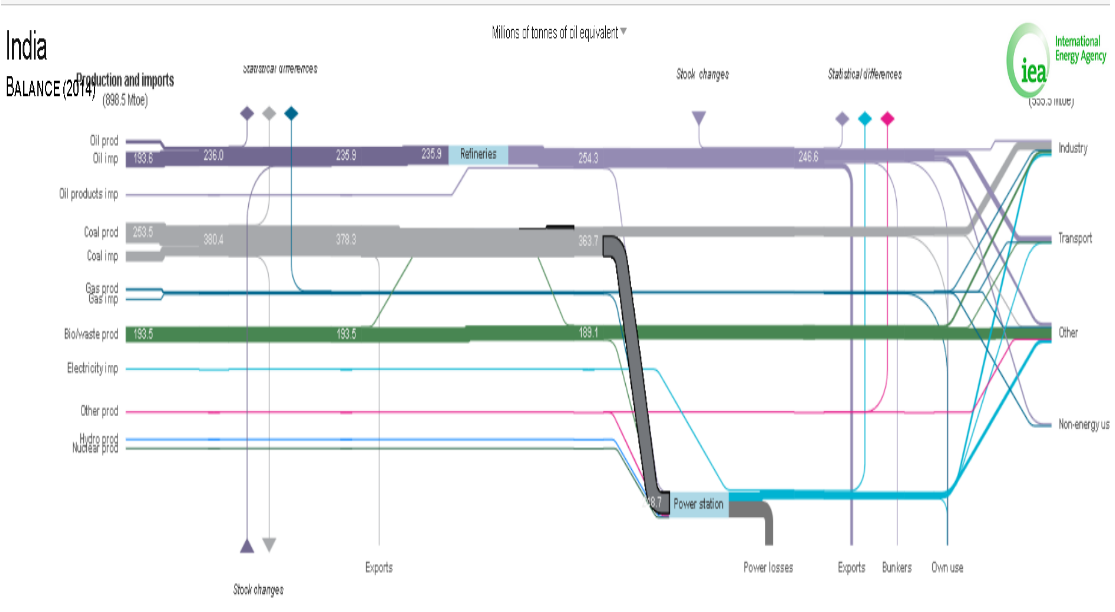
Region of growth

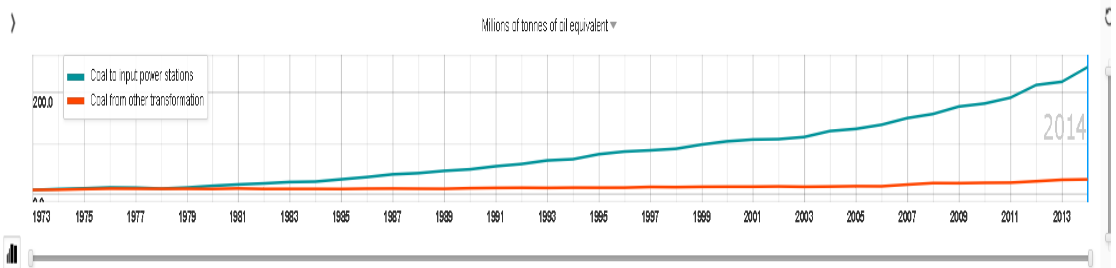




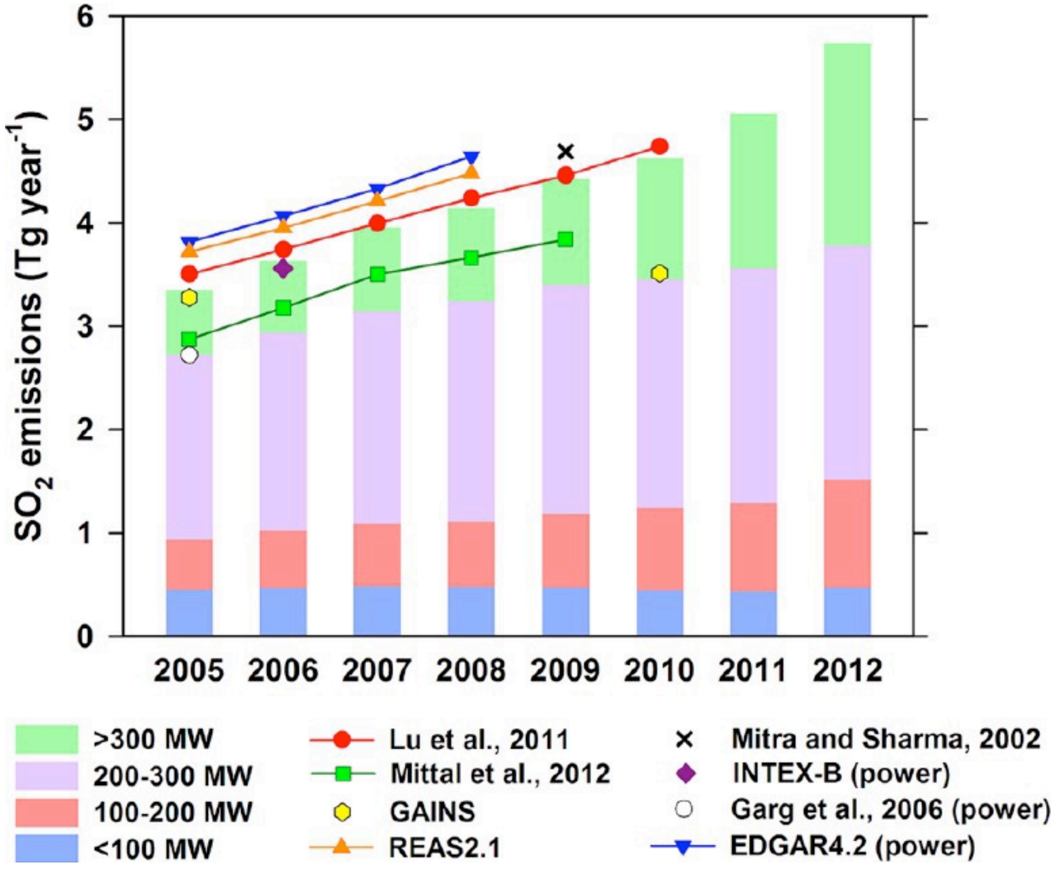
International Energy Agency & World Bank

Coal consumption



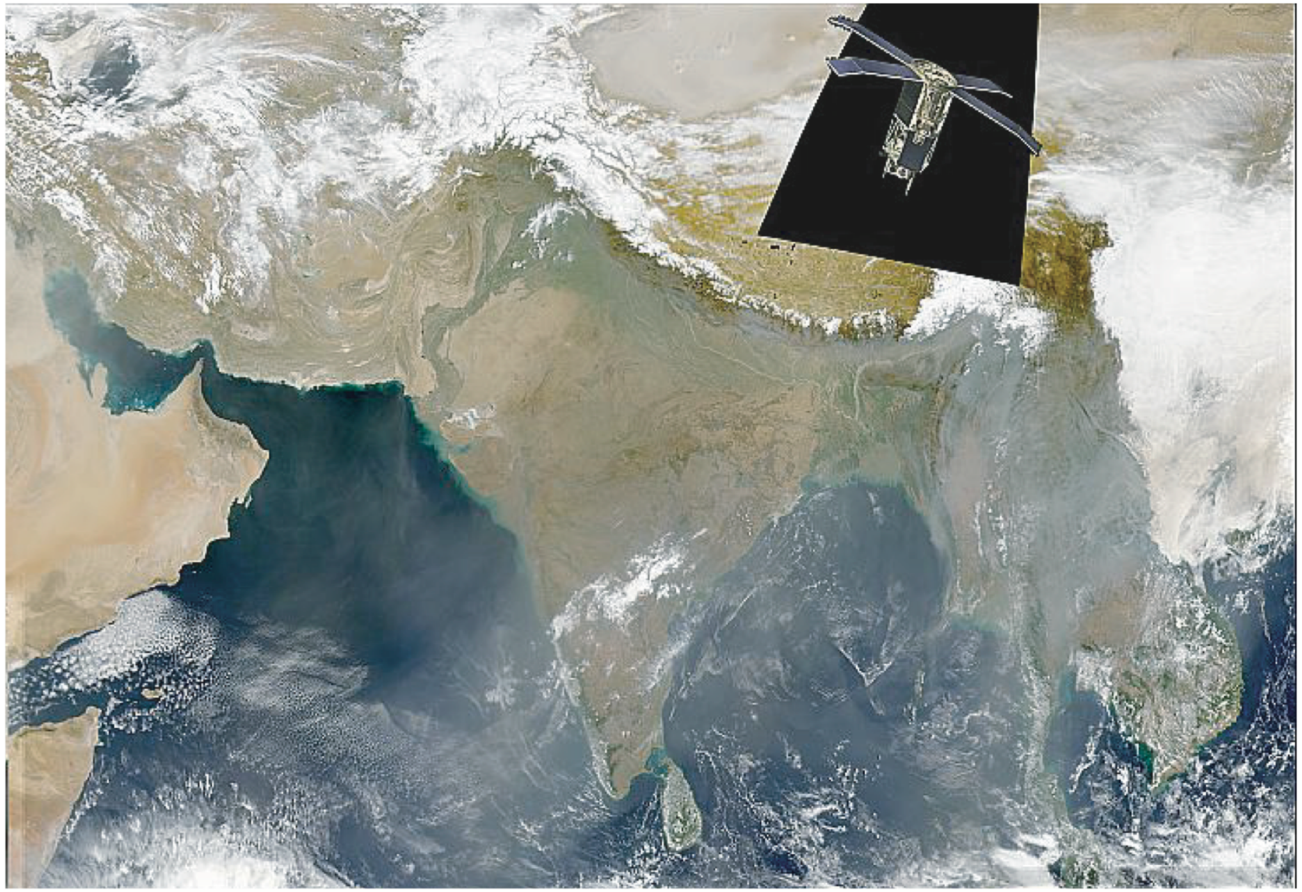


http://www.iea.org/sankey/#?c=India&s=Balance

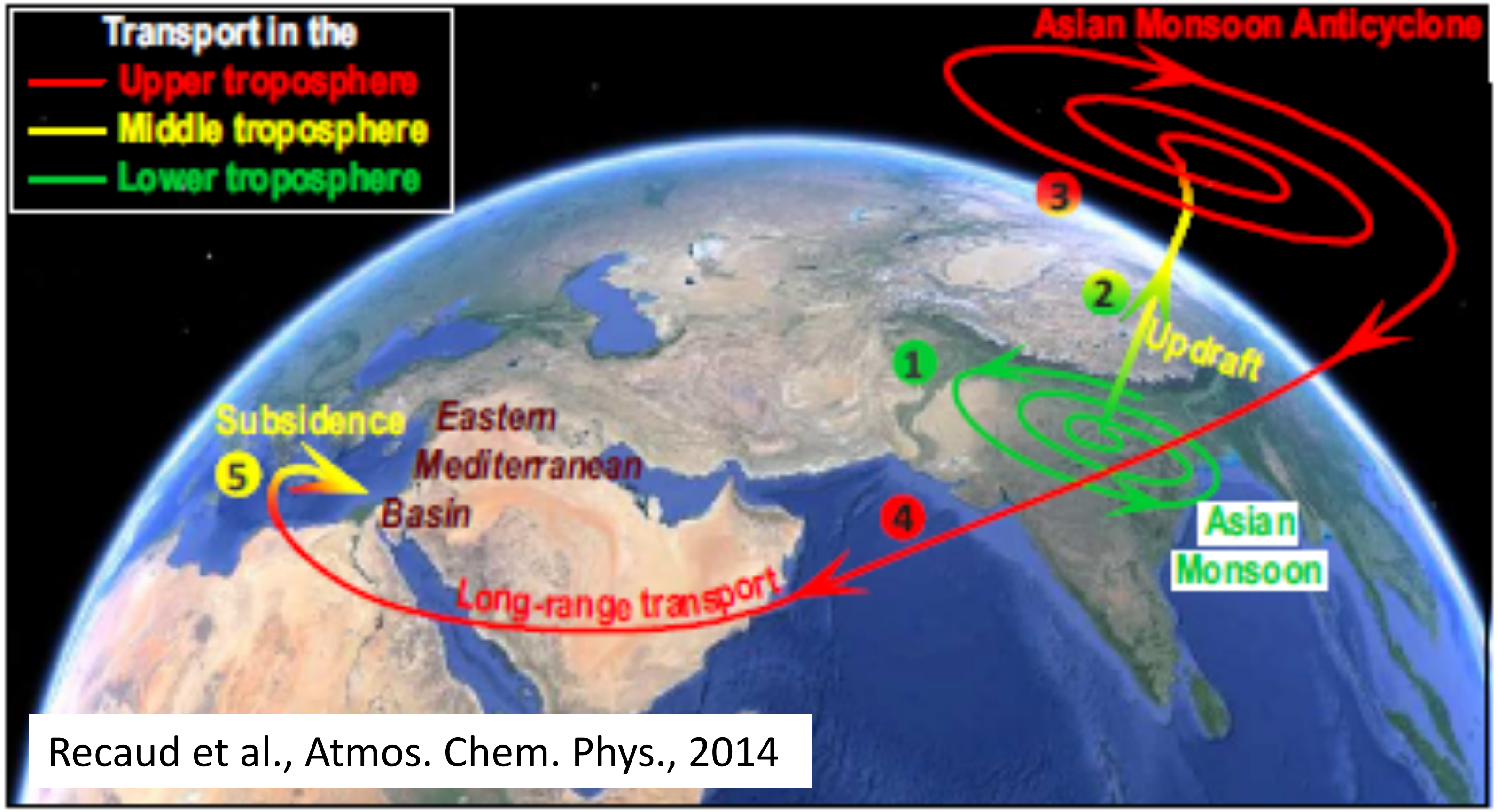


Lu et al. 2013

S-Asian brown cloud in dry winter



South Asian Monsoon



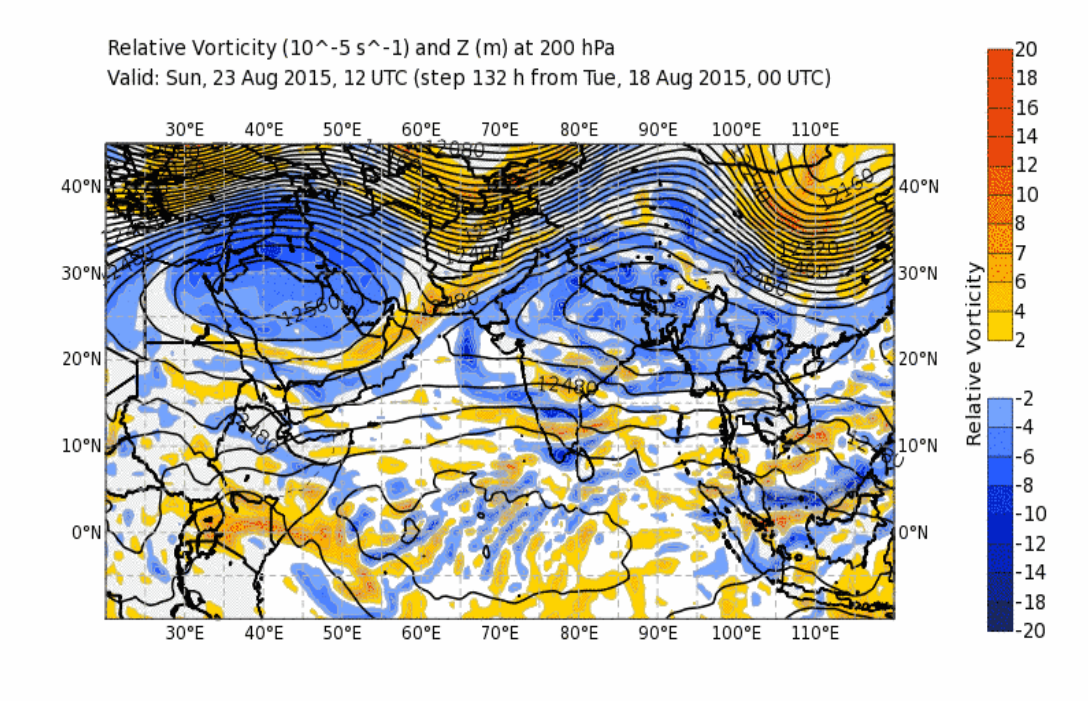
Recaud et al., Atmos. Chem. Phys., 2014

OMO
Instrumentation on HALO

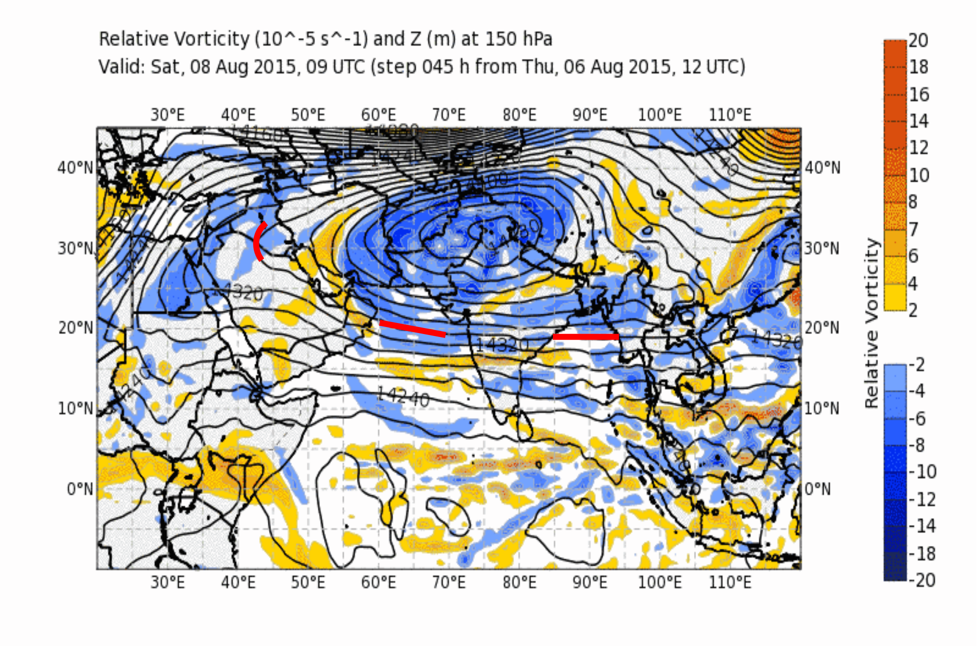
- Actinic Flux
- OH/HO2/RO2
- O3/CO/H2O
- NO/NO2/NOy
- VOC/OVOC/HCHO
- H2O2/org. Peroxides
- SO2/Particle

HALO Aircraft

Meteorological situation during OMO



Cyprus 13.8.-27.8.

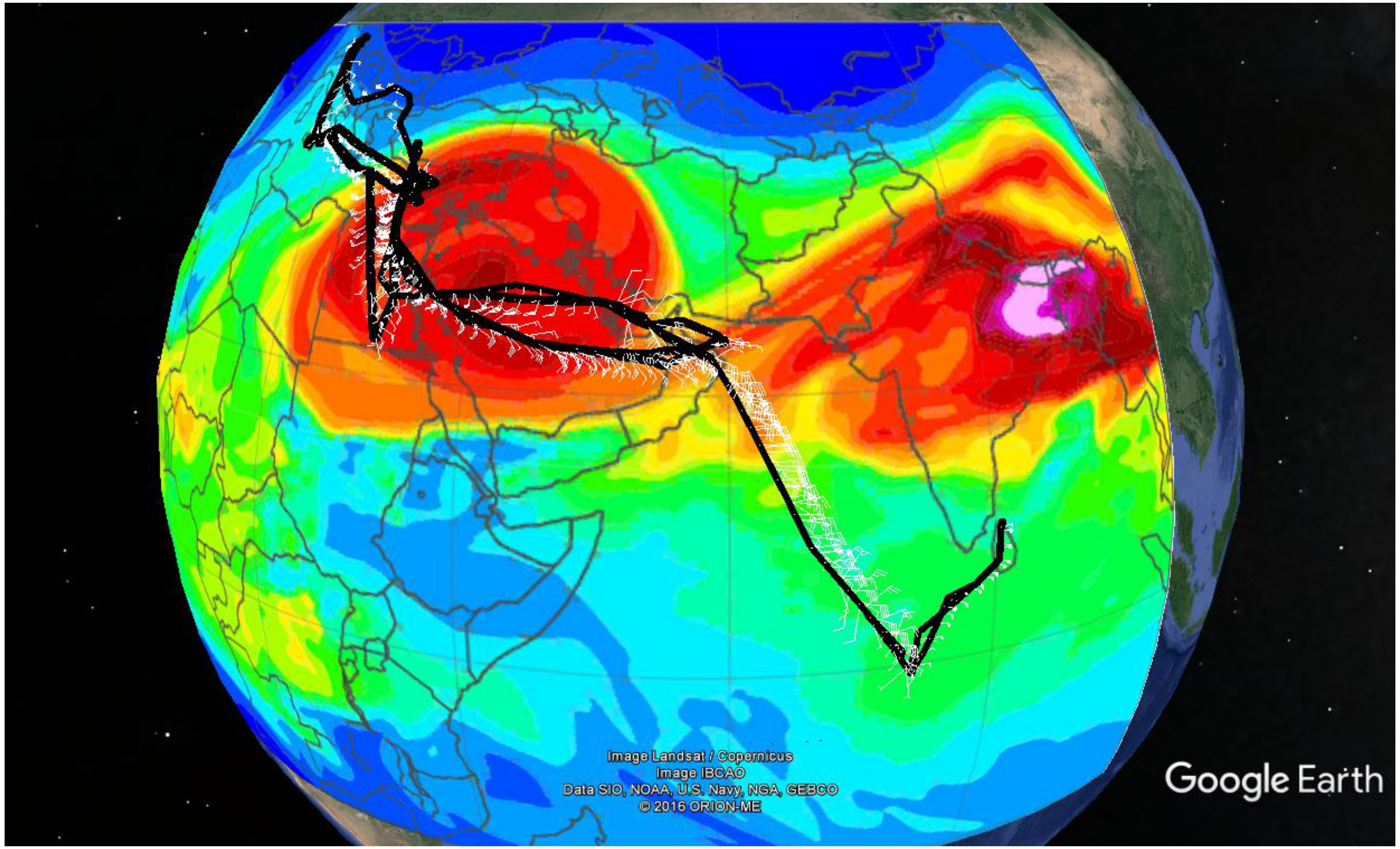


Gan 6.8. – 9.8.

Hans Schlager

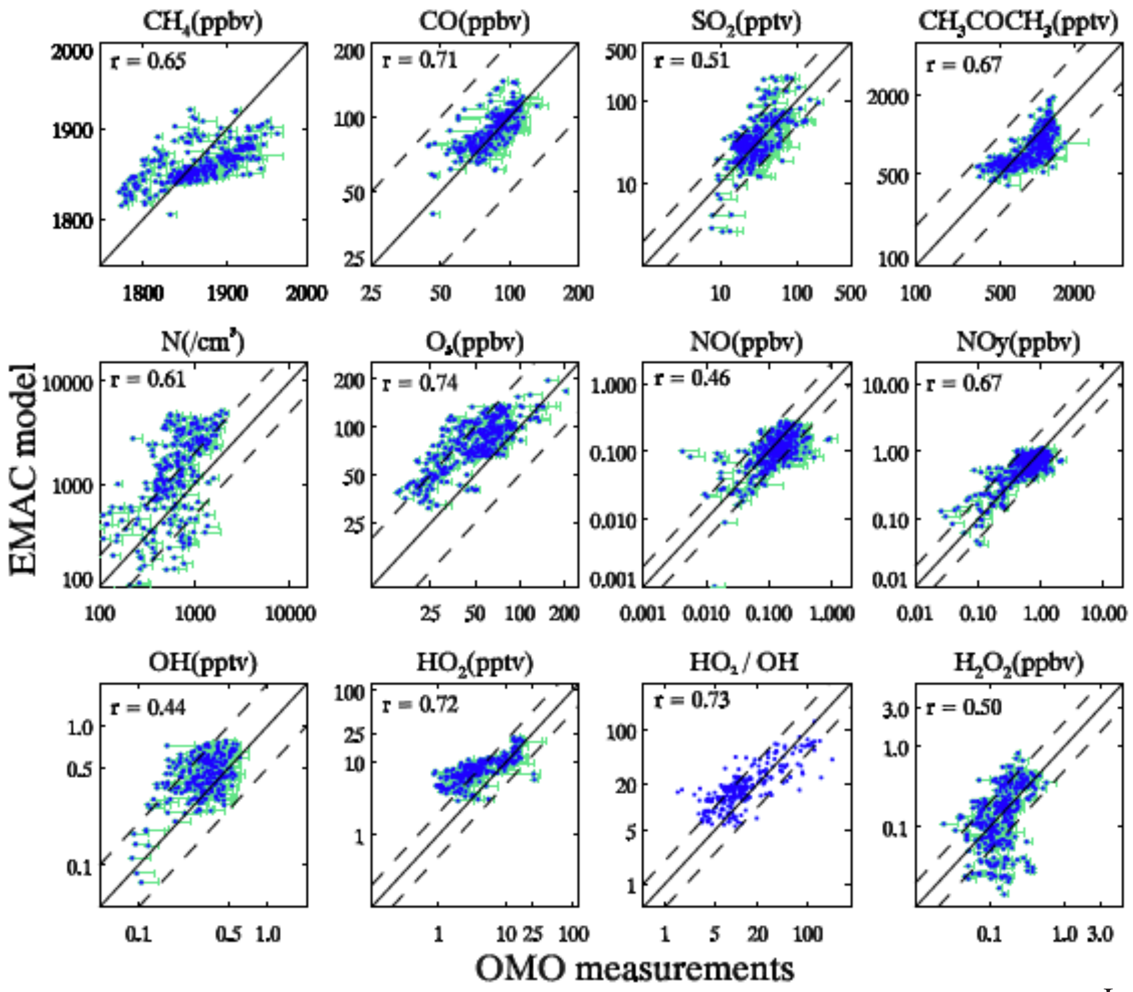
Flight tracks

- Around 120 flight hours
- More then 80% overall instrument data coverage



MACC model result
South Asian CO tracer

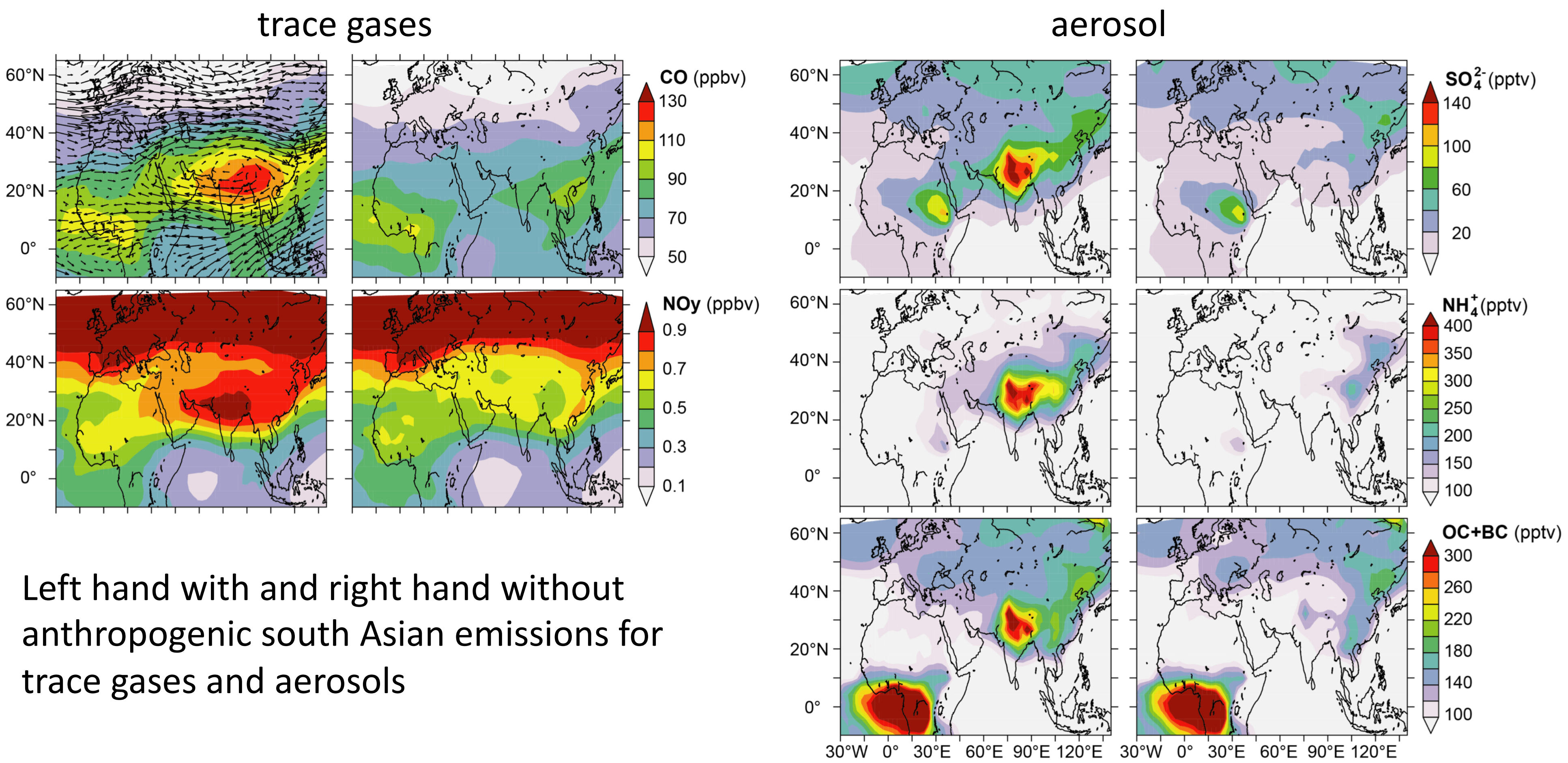
EMAC model vs Measurements



SO2 emissions in S Asia increased to match observation

Lelieveld et al., 2018

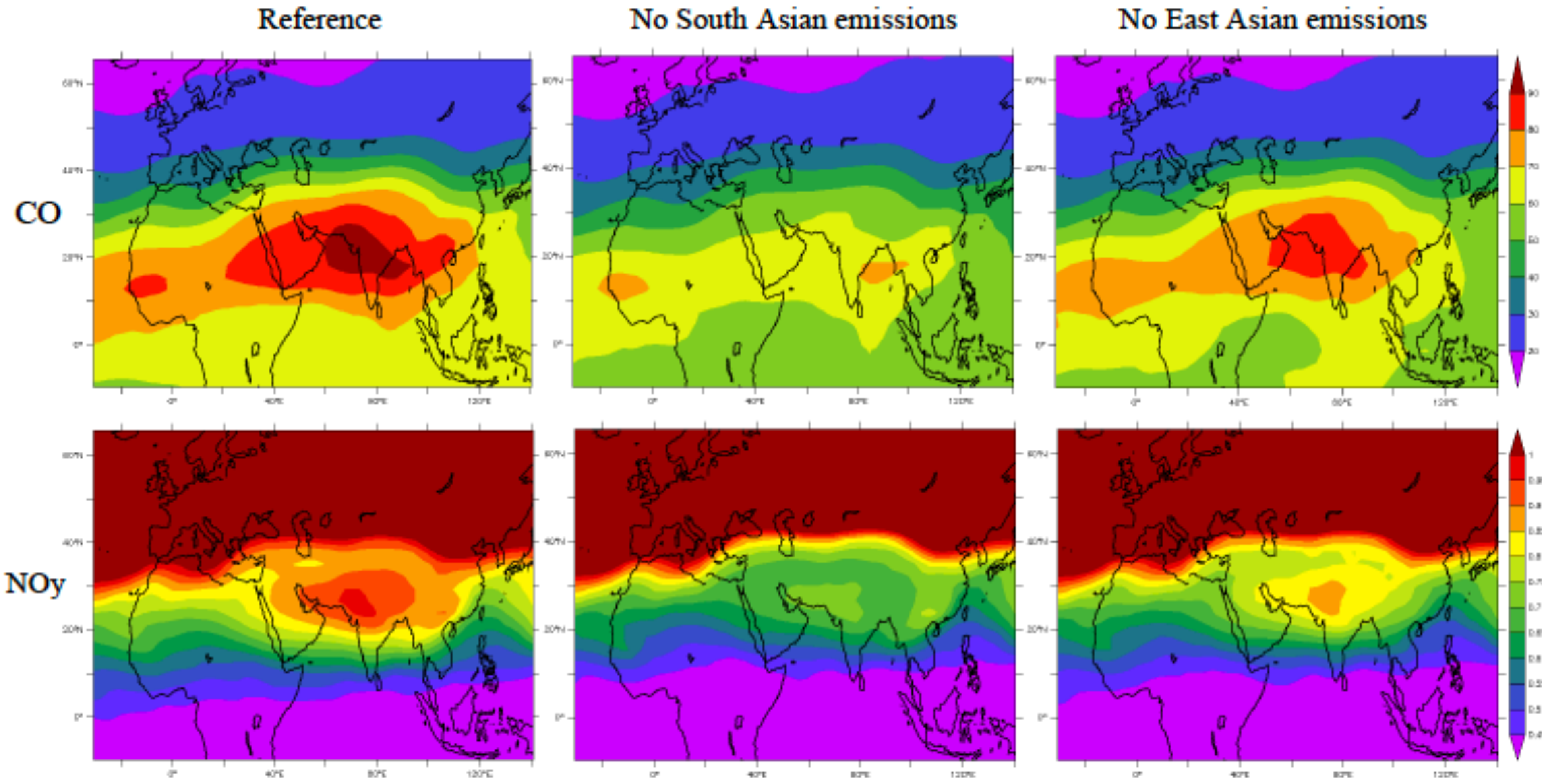
Influence of South Asian anthropogenic Emission



Left hand with and right hand without anthropogenic south Asian emissions for trace gases and aerosols

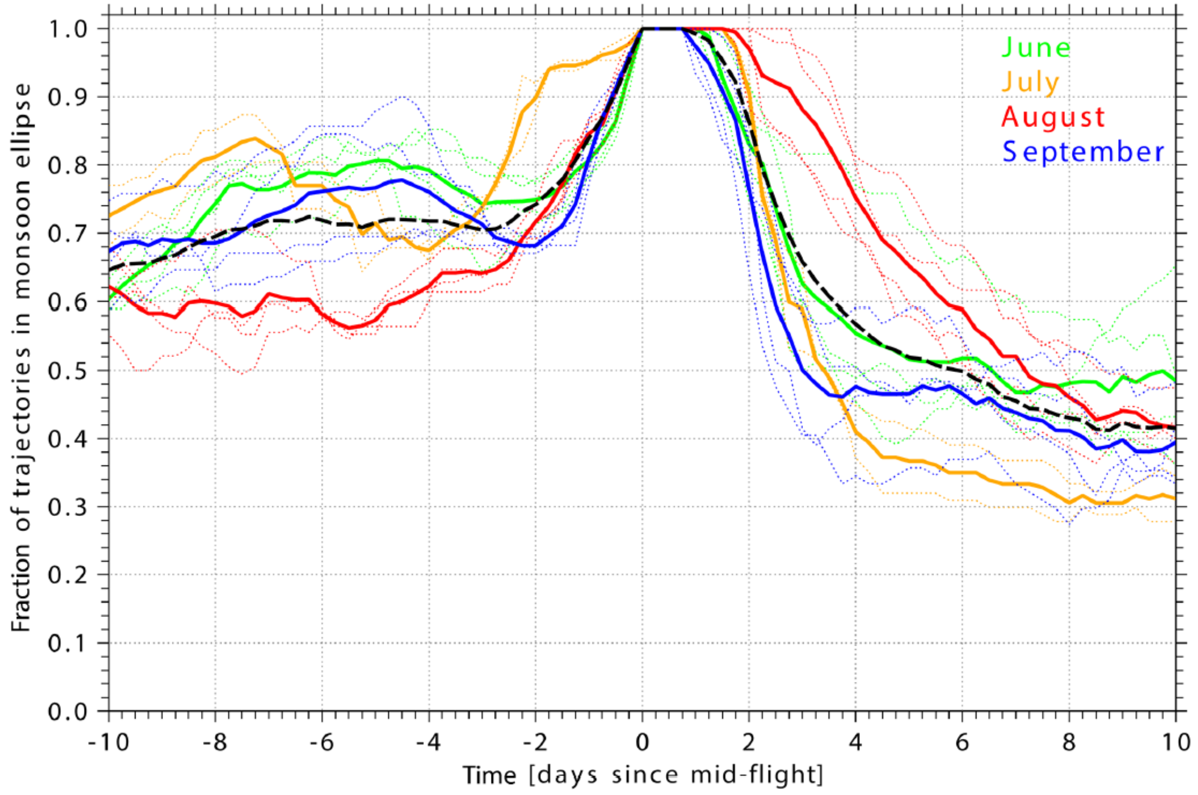
Lelieveld et al., 2018

Contribution of South and East Asian Emissions

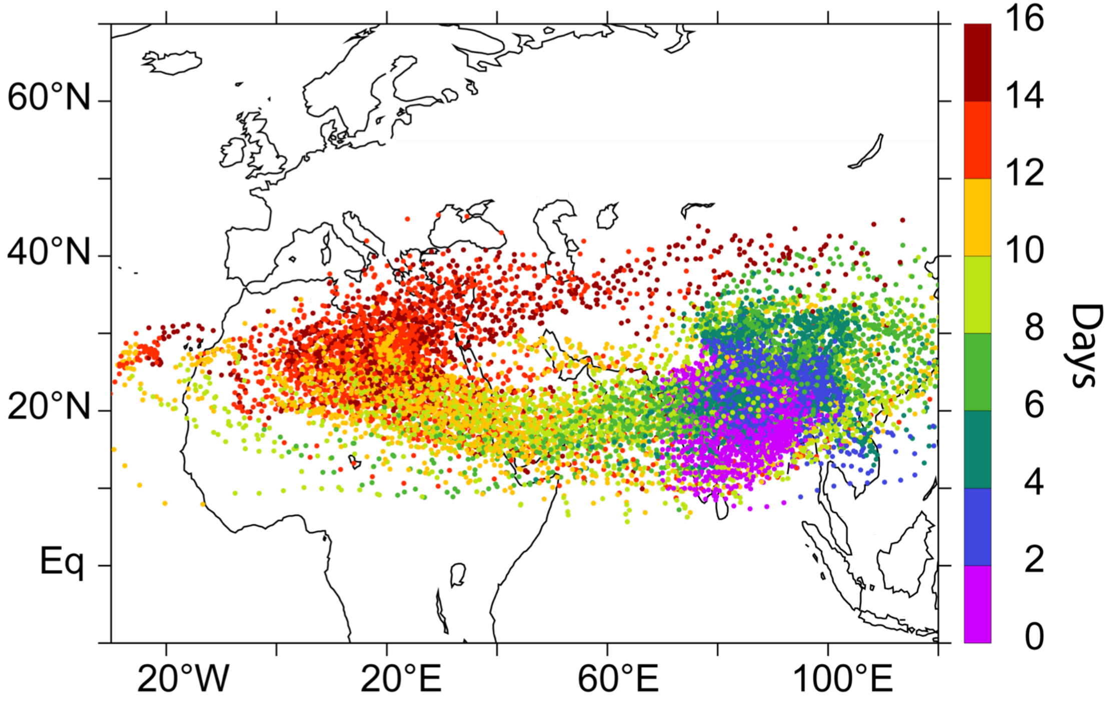


Lelieveld et al., 2018

Residence time inside the AMA



Rauthe-Schöch et al 2016 for CARIBIC JJA 2008

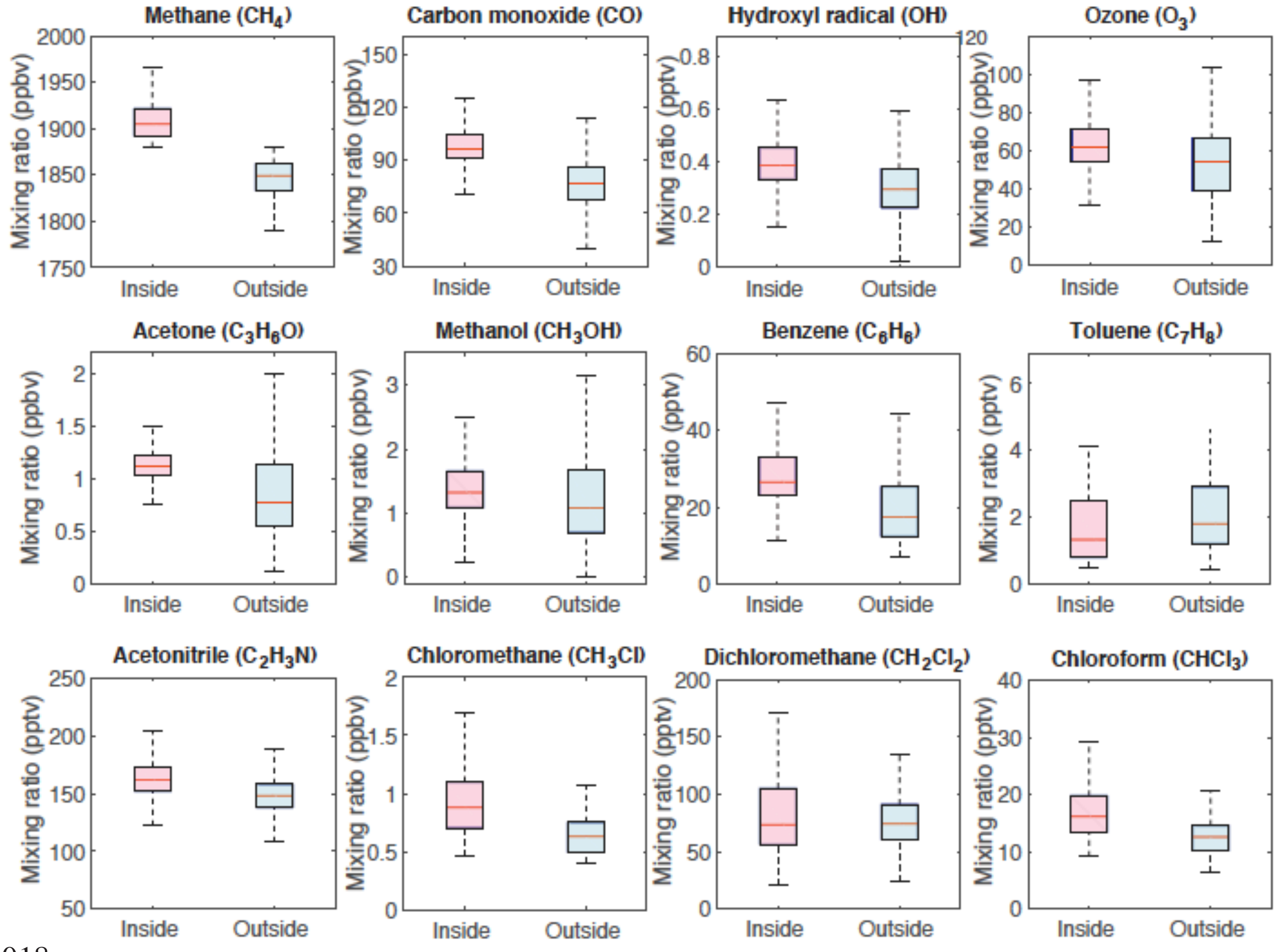


Schlager; Tomsche et al. 2016
FLEXPART/HYSPLIT 5-10 days

Chemical aging of the airmass confined in AMA

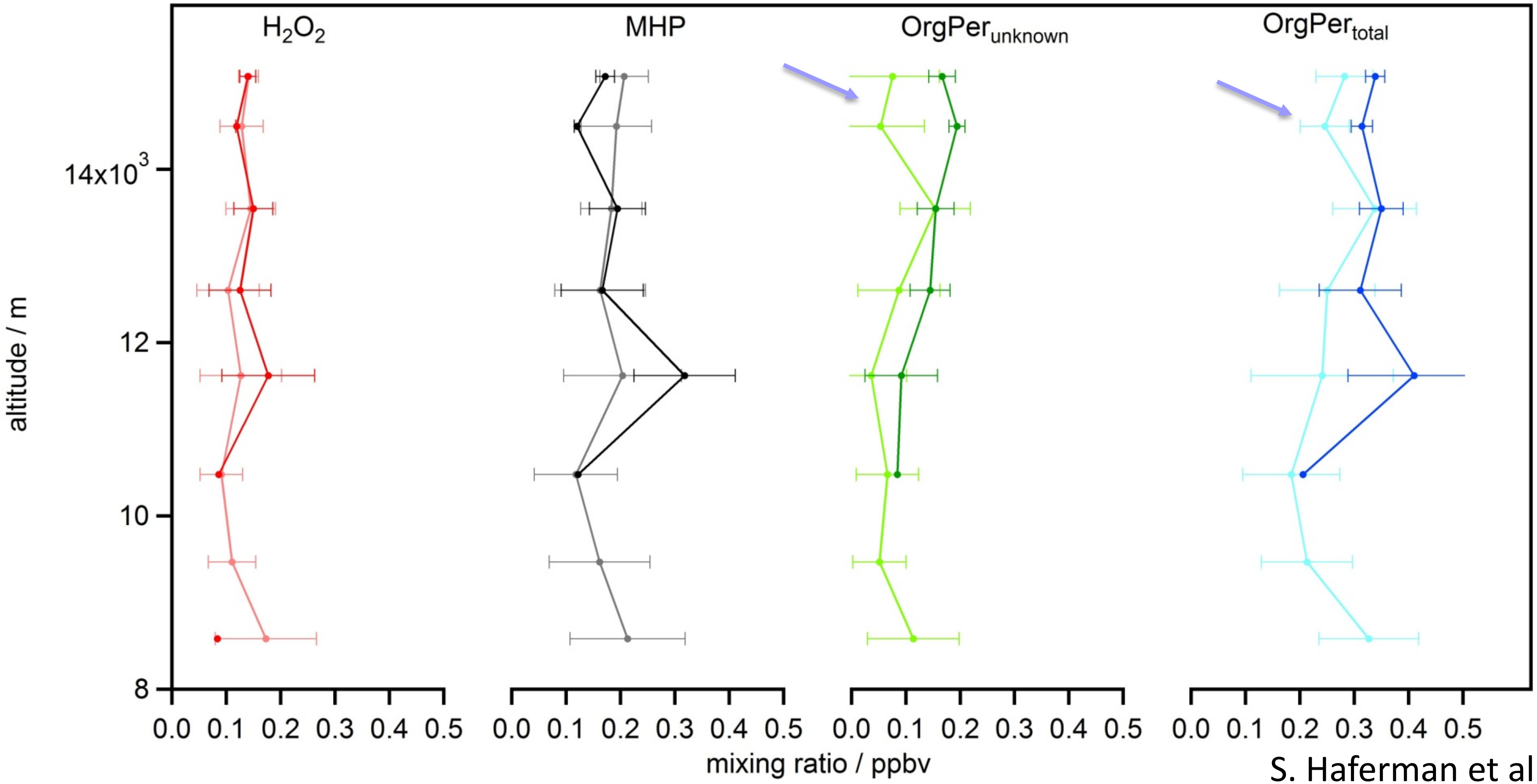
Benzene/Toluene ratio
Indicates chemical age
4-12 days;
mean 9+-2 days

CH4 level of 1879.8ppb
used as threshold
indicator of AMA



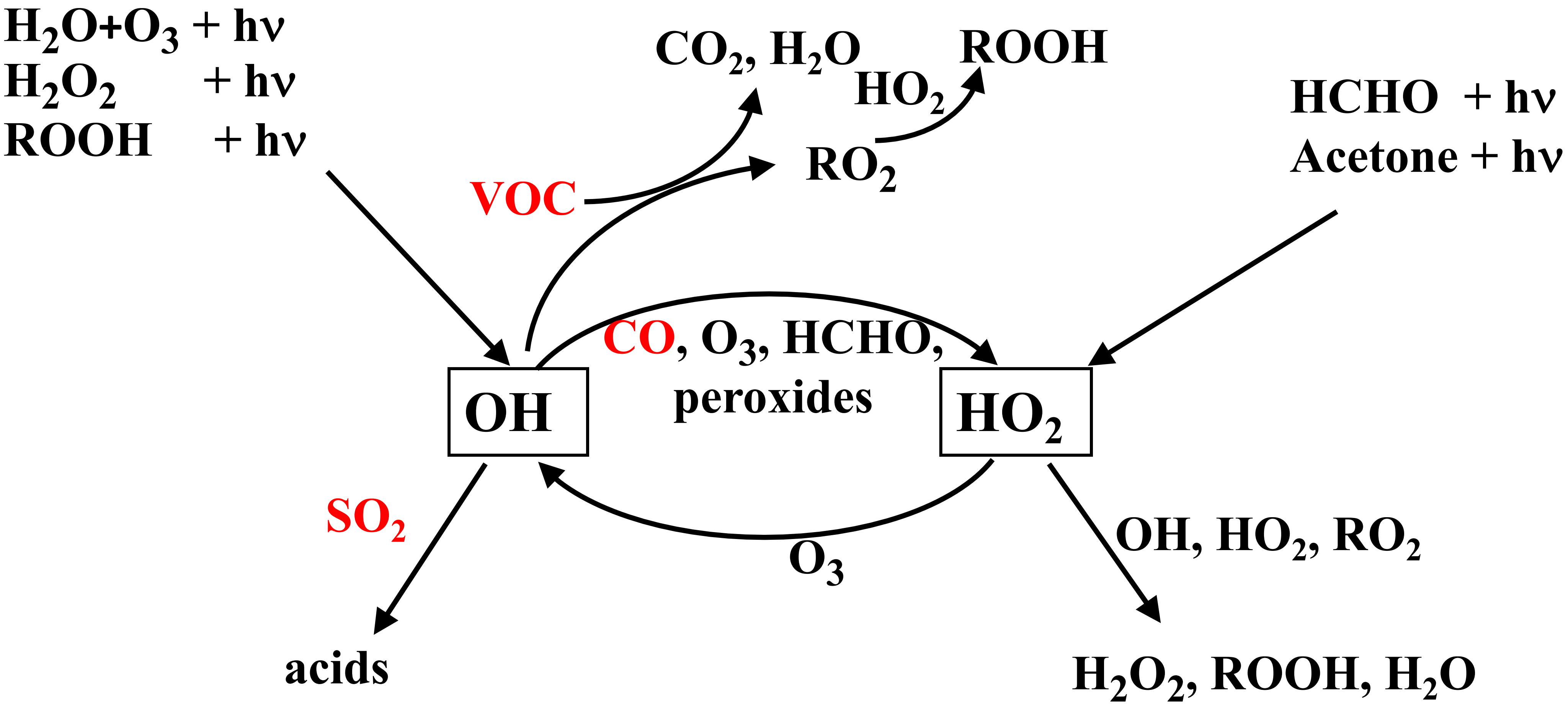
Lelieveld et al., 2018

Peroxides

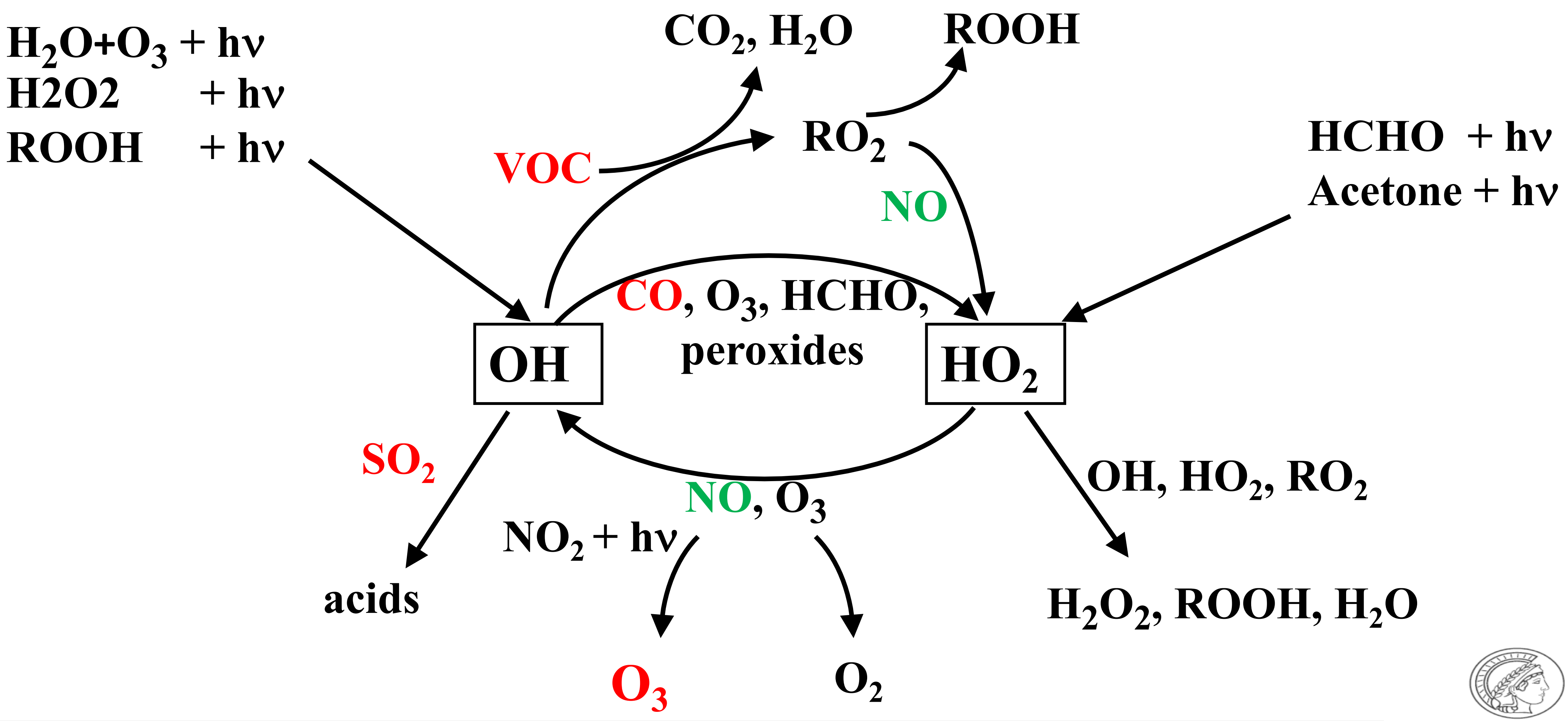


S. Haferman et al. 2016

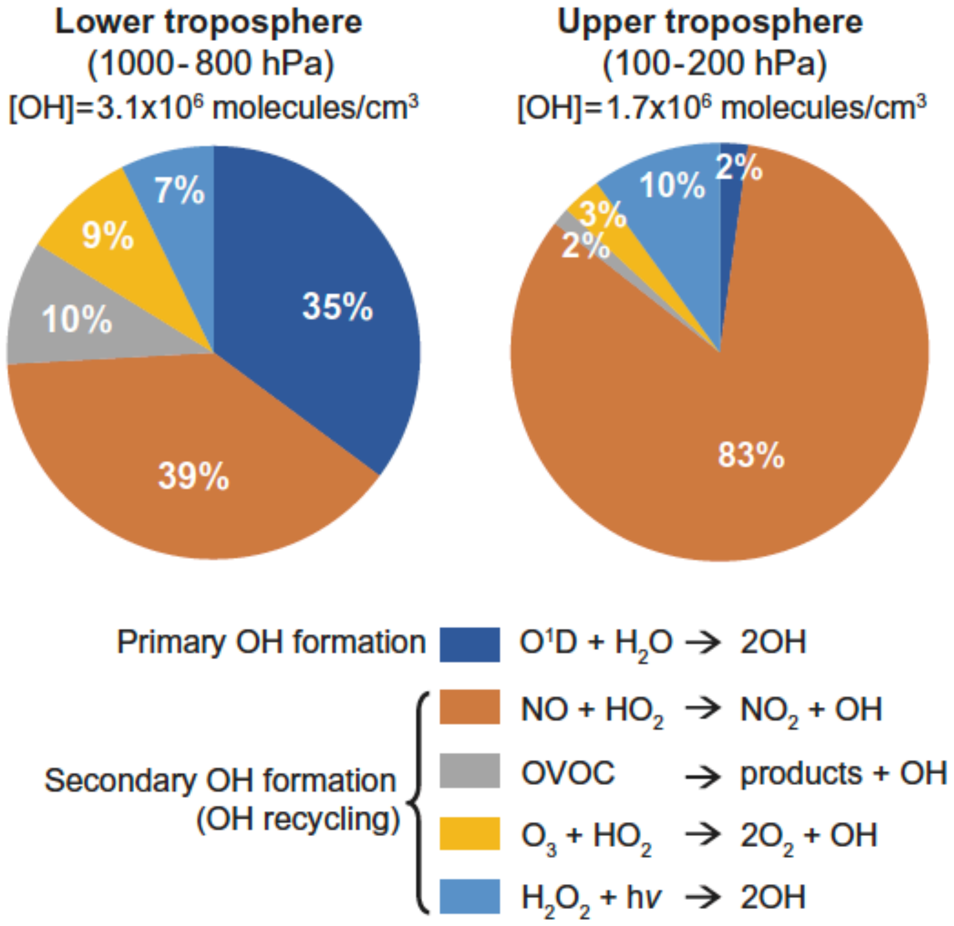
Simplified HOx chemistry, low NOx



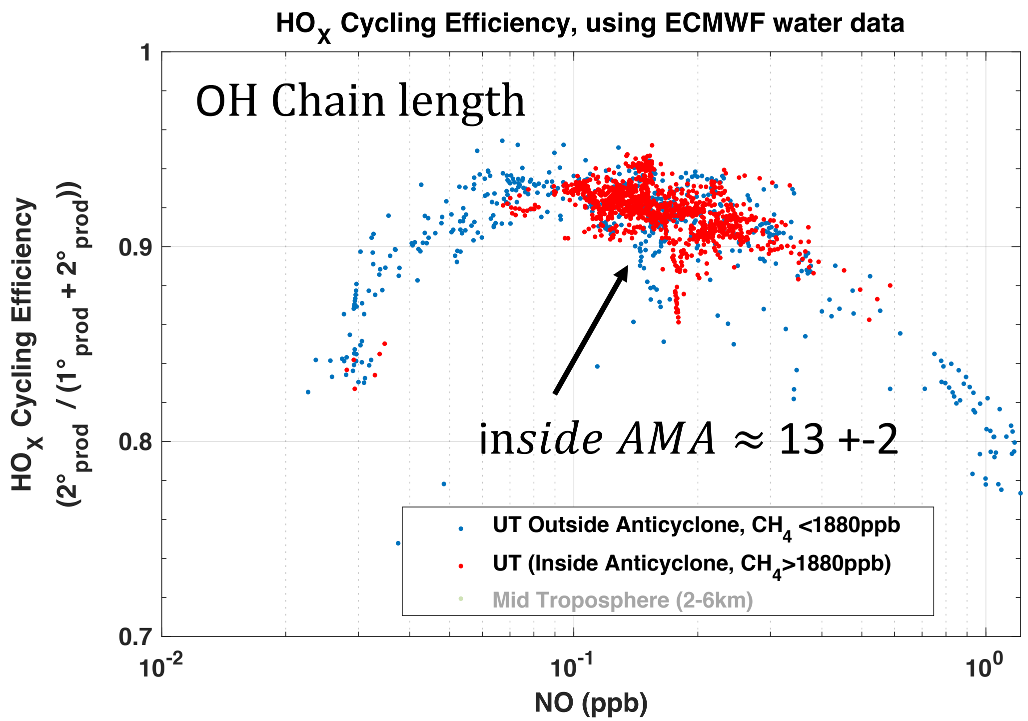
Simplified HOx chemistry



OH recycling

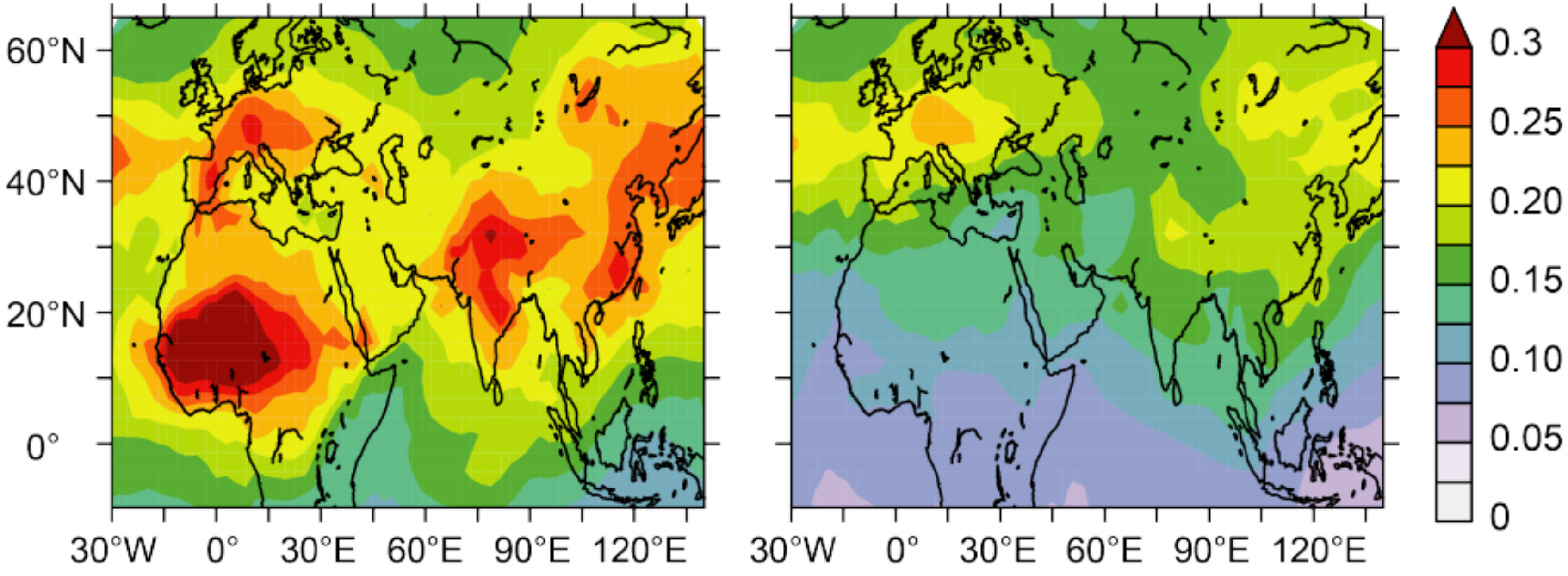


Lelieveld et al., 2018



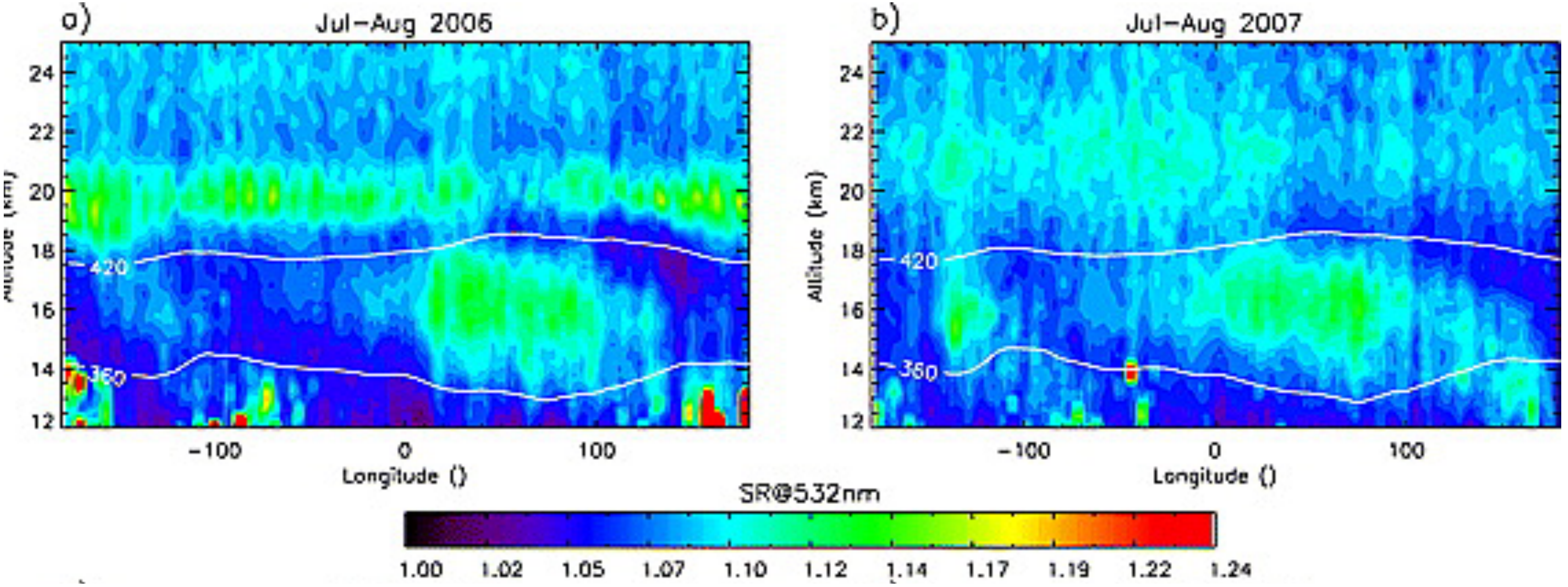
Marno et al., 2019; in prep

Impact of Lightning NO on OH



Lelieveld et al., 2018

ATAL Layer

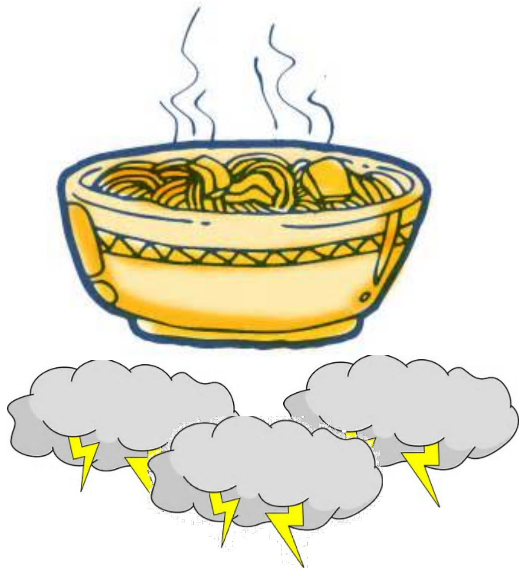


Vernier et al., GRL 2011

Summary

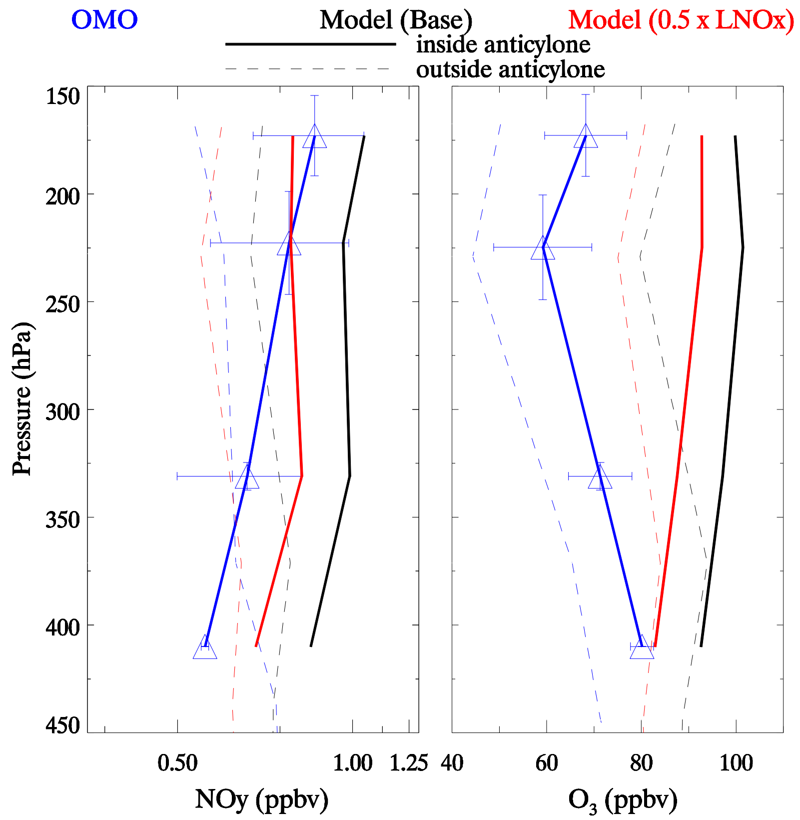
- South Asian emissions dominates as source region
- FLEXPART calculations show a typical residence time inside the anti cyclone of 5-10 days in agreement with chemical clock Benzene/Toluene
- NOx increased due lightning of about 30%
- Ongoing injection of NOx maintains recycling efficiency for OH
- Efficient recycling of OH maintains oxidation of pollutants inside AMA, also forming particles

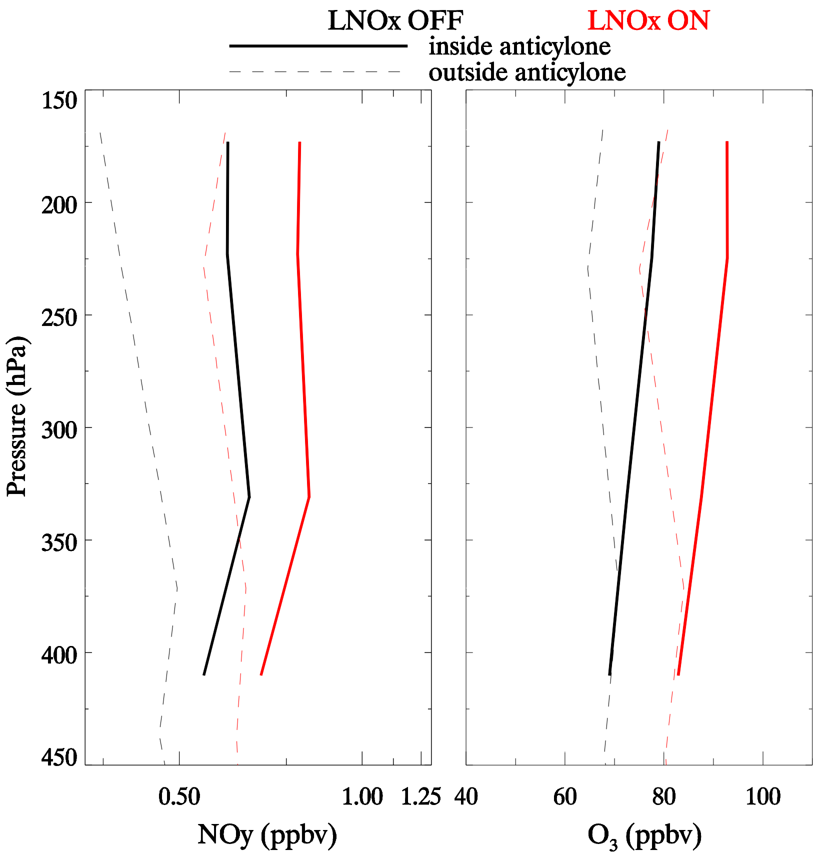
→ Where does the outflow goes next and how much of the emissions are then left ?

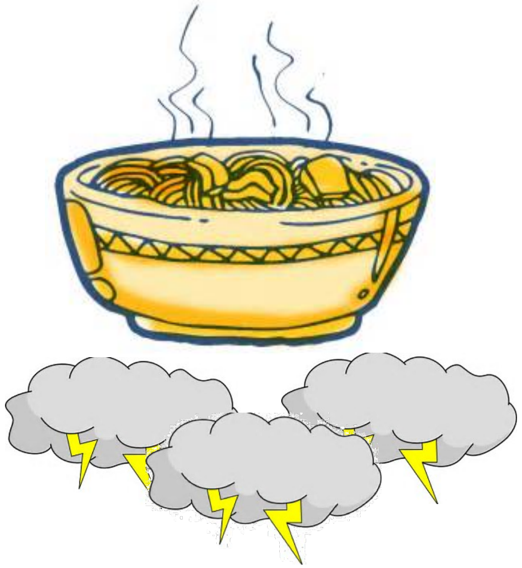


Thank you!!

Impact of Lightning







- Lightning enhances NOy by ~30-40% and O_3 by ~20%.

N.Ohja, 2017

Intercomparison of MPIC and Jülich HO_X

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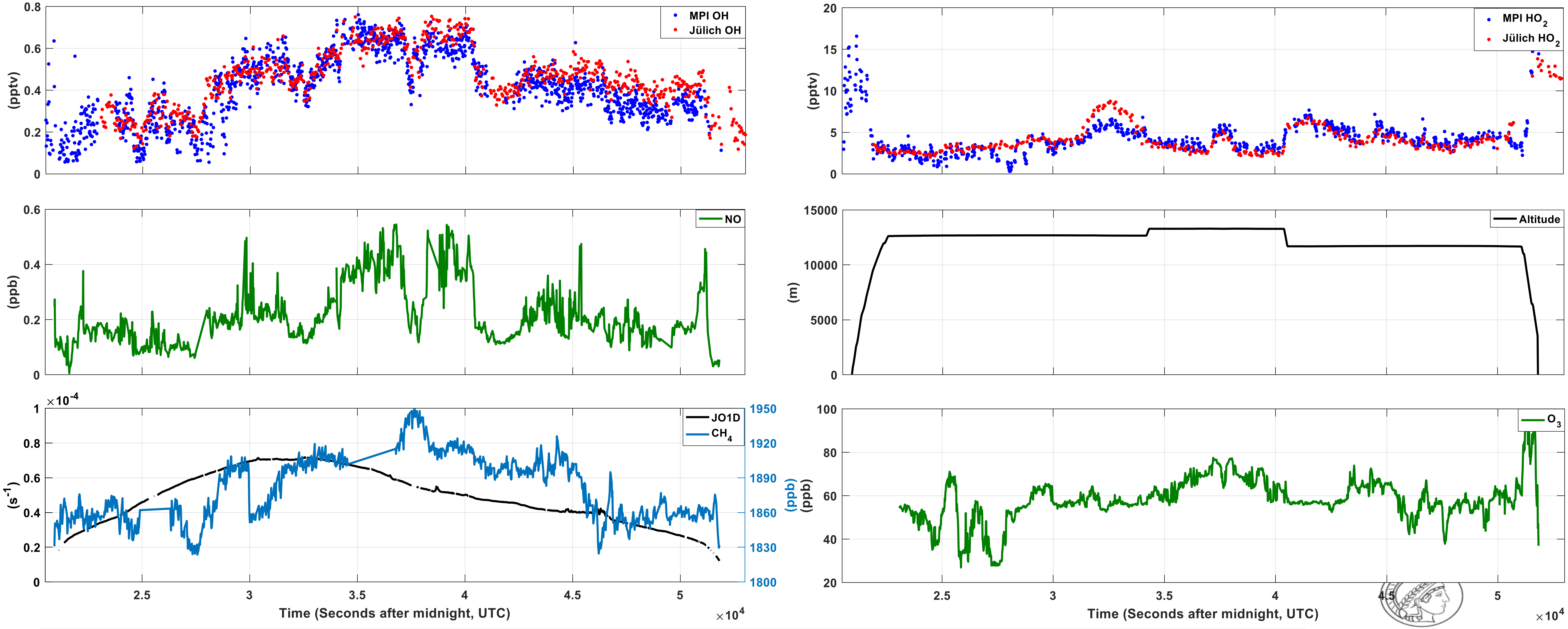
Flight 20



Intercomparison of MPIC and Jülich HO_X

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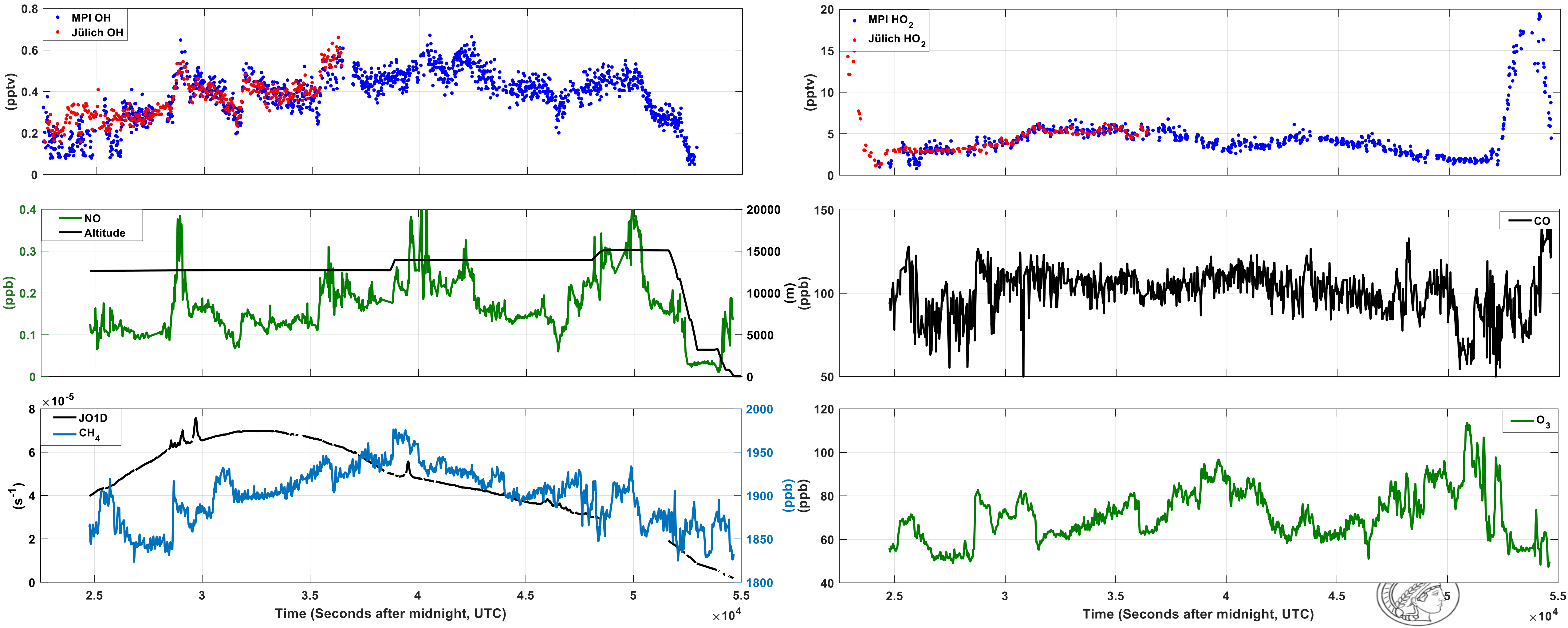
Flight 21



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Intercomparison of MPIC and Jülich HO_X

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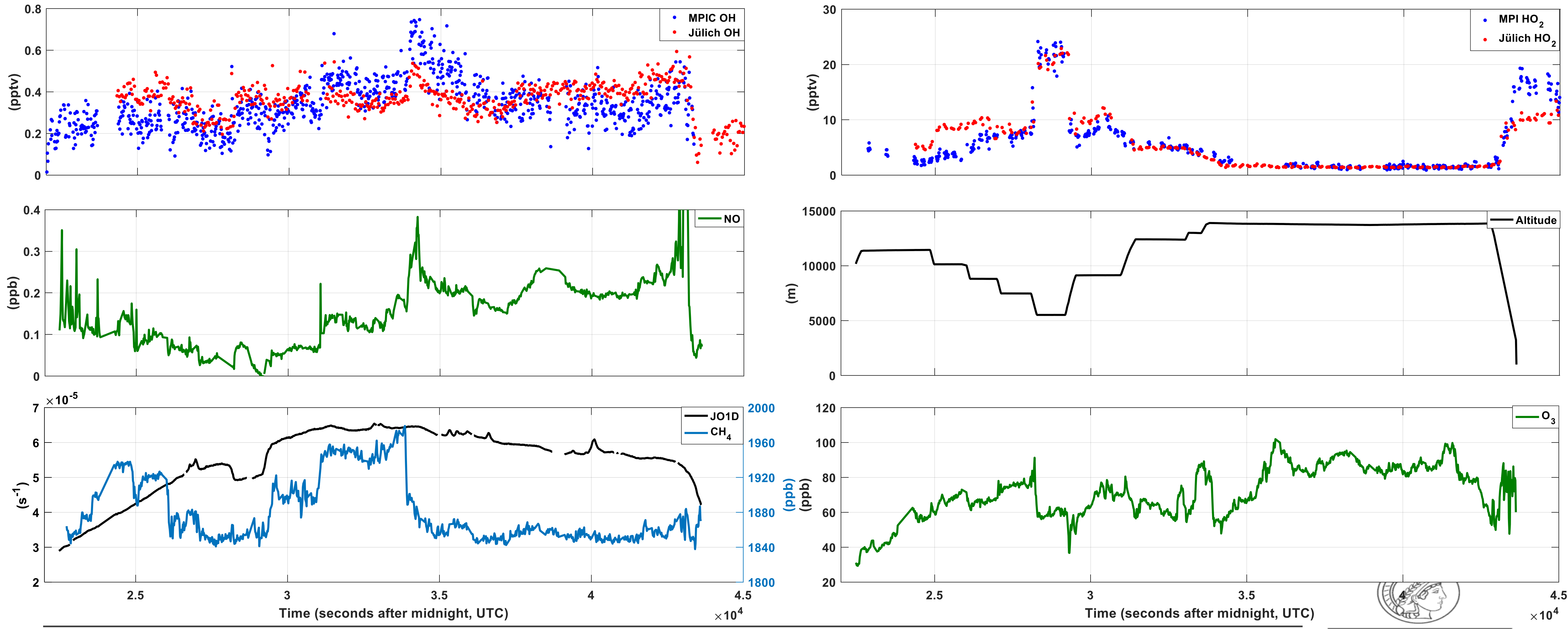
Flight 22



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Intercomparison of MPIC and Jülich HO_X

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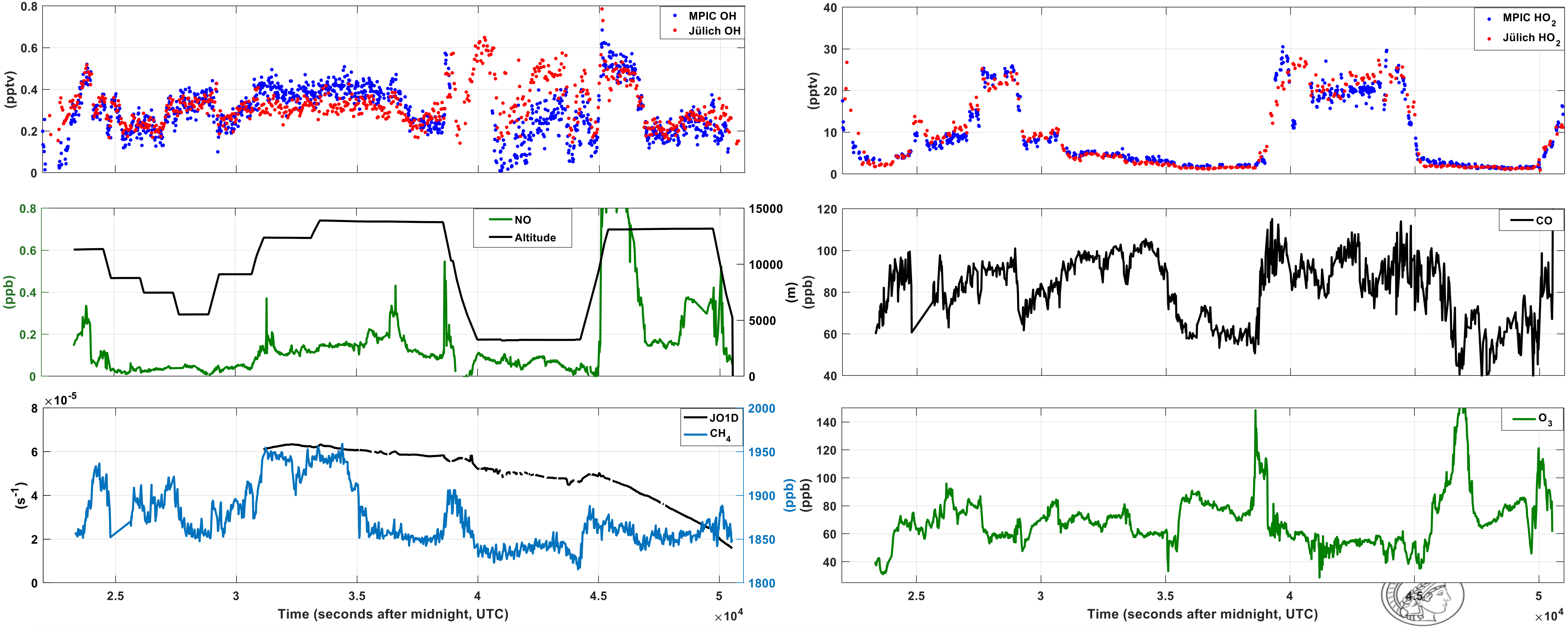
Flight 23



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HO_X cycling efficiency inside outside of AMA

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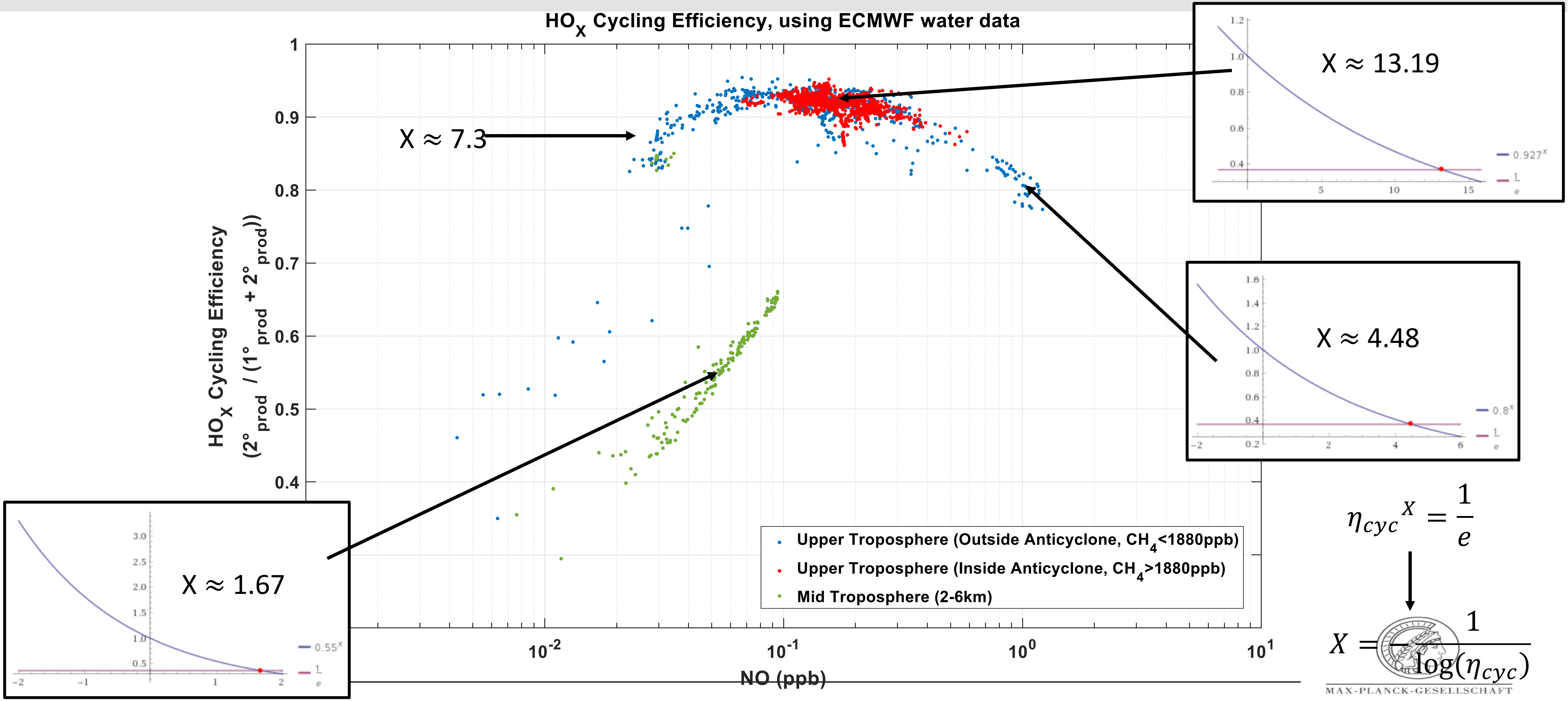
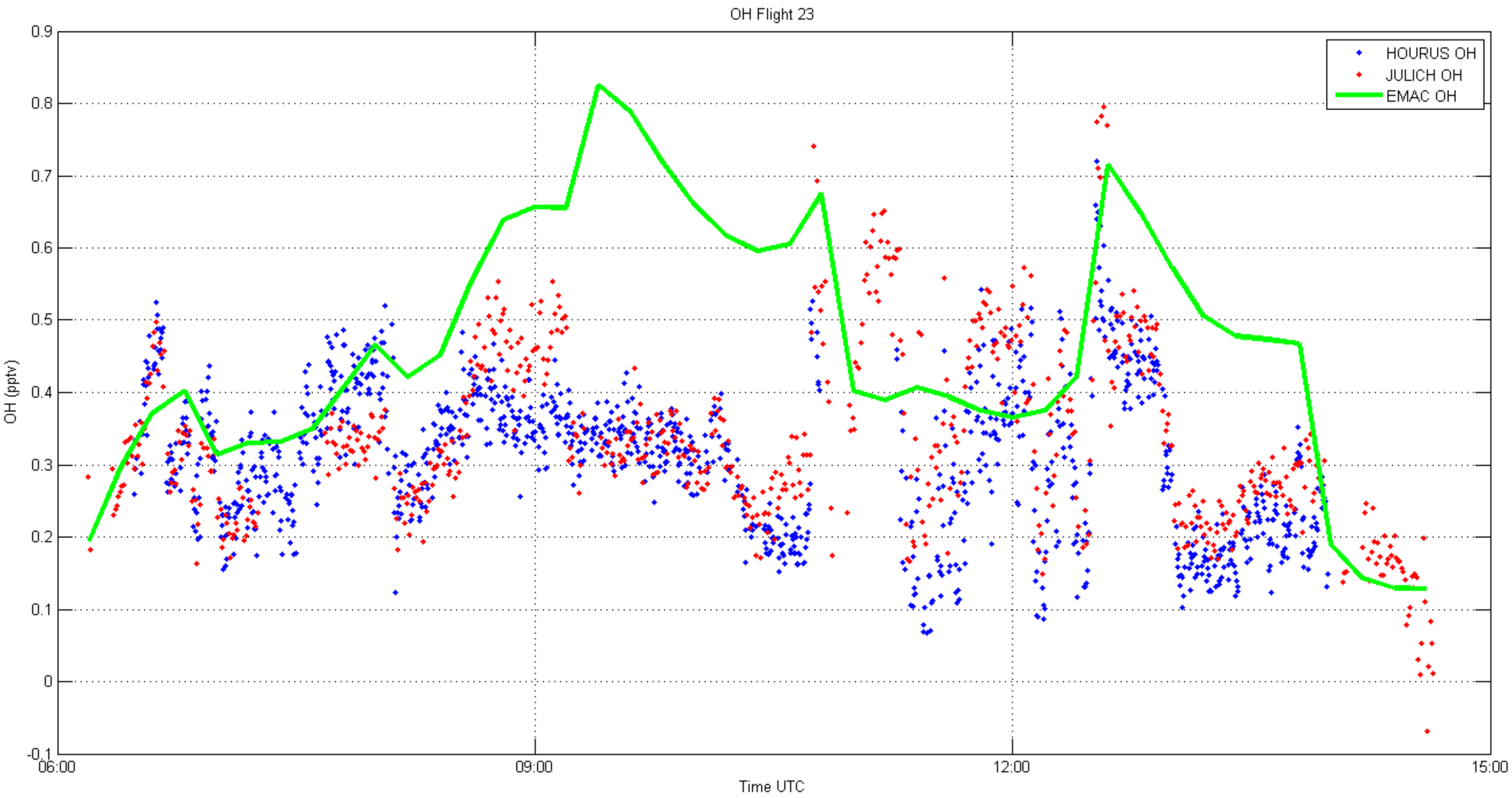


$$\eta_{cyc}{}^{X} = \frac{1}{e}$$

$$\downarrow$$

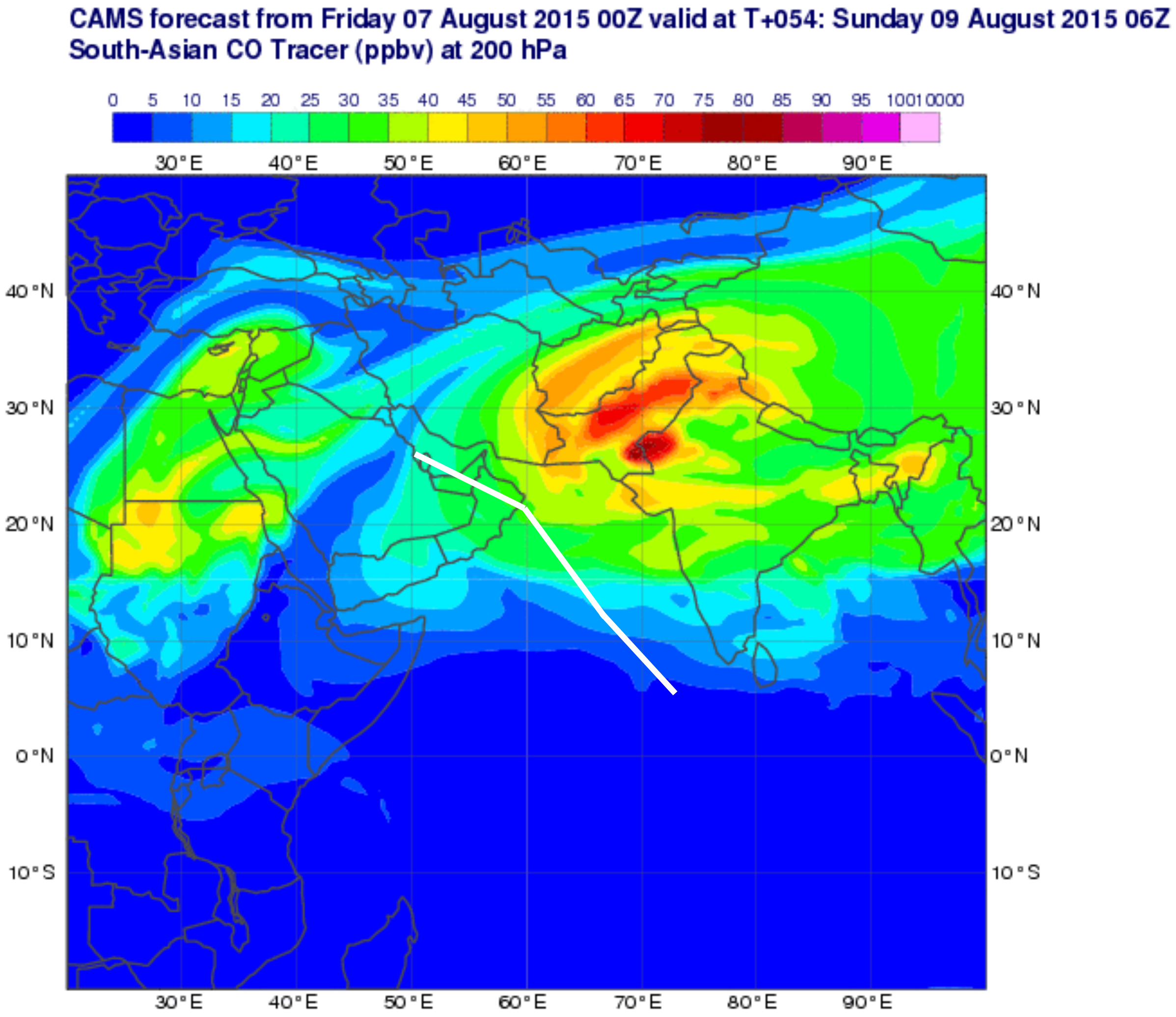
$$X = \frac{1}{\log(\eta_{cyc})}$$

OH comparison



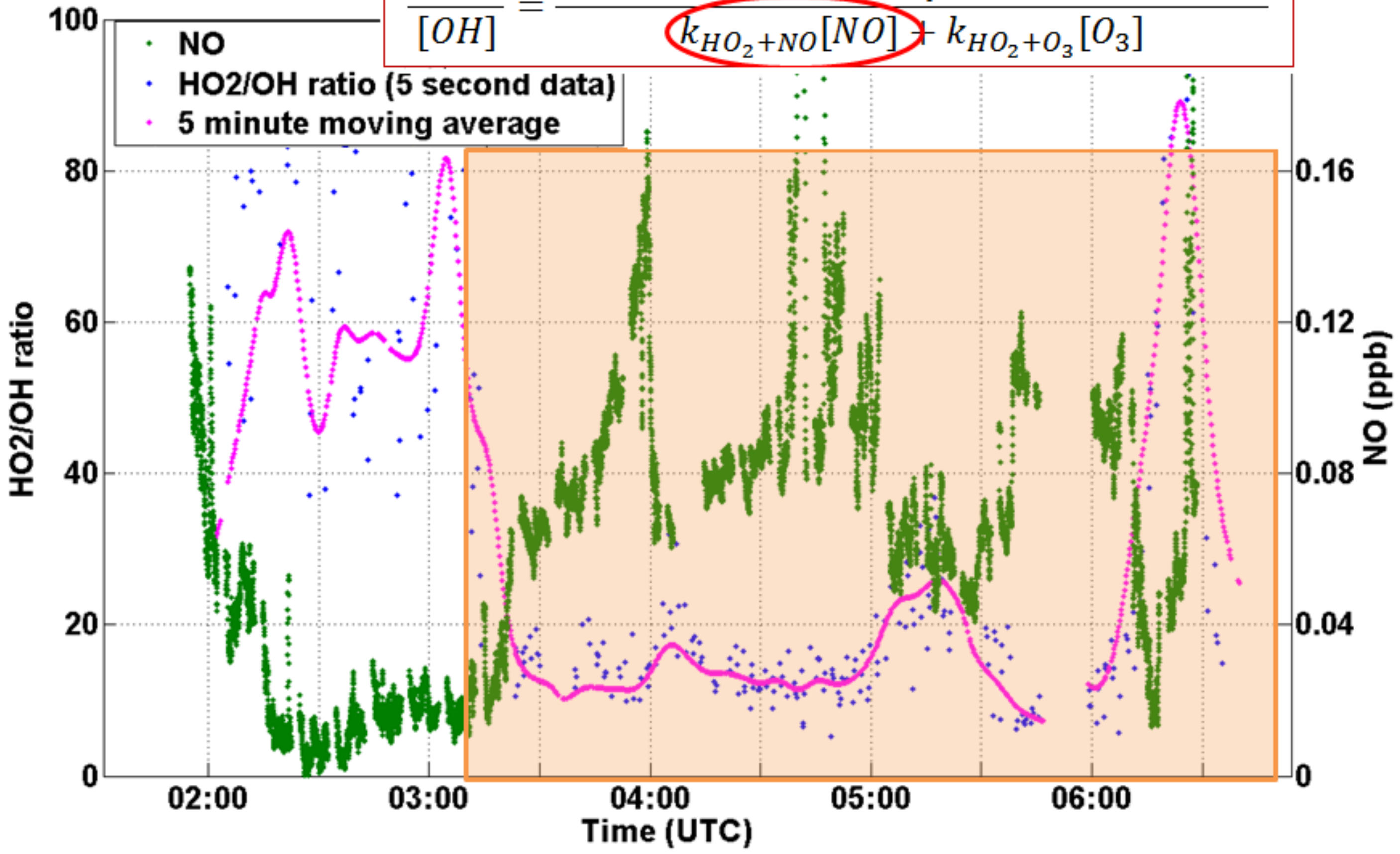
Flight 9.8.2015

- HALO Flight track crossing anti cyclone



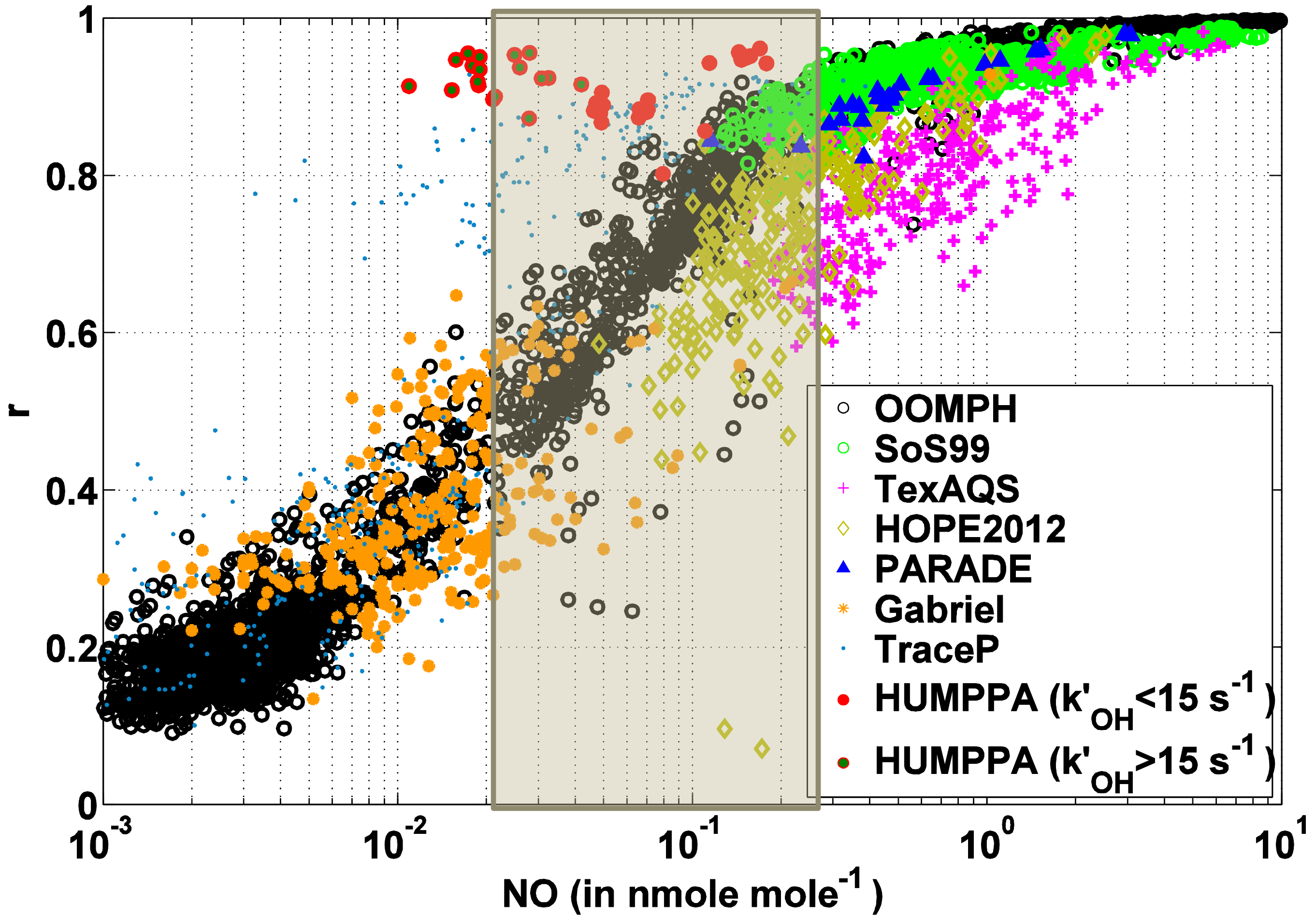
Impact on HOx

$$\frac{[HO_2]}{[OH]} = \frac{k_{OH+CO}[CO] + k_{OH+CH_4}[CH_4] + K_{OH+O3}[O_3]}{k_{HO_2+NO}[NO] + k_{HO_2+O_3}[O_3]}$$



D. Marno, 2016

OH recycling probability as a function of NO in different environments



Ongoing injection of NO by lightning
into the anticyclone
maintains OH recycling efficiency
And therefore enhances the oxidation capacity
Inside the anti-cyclone

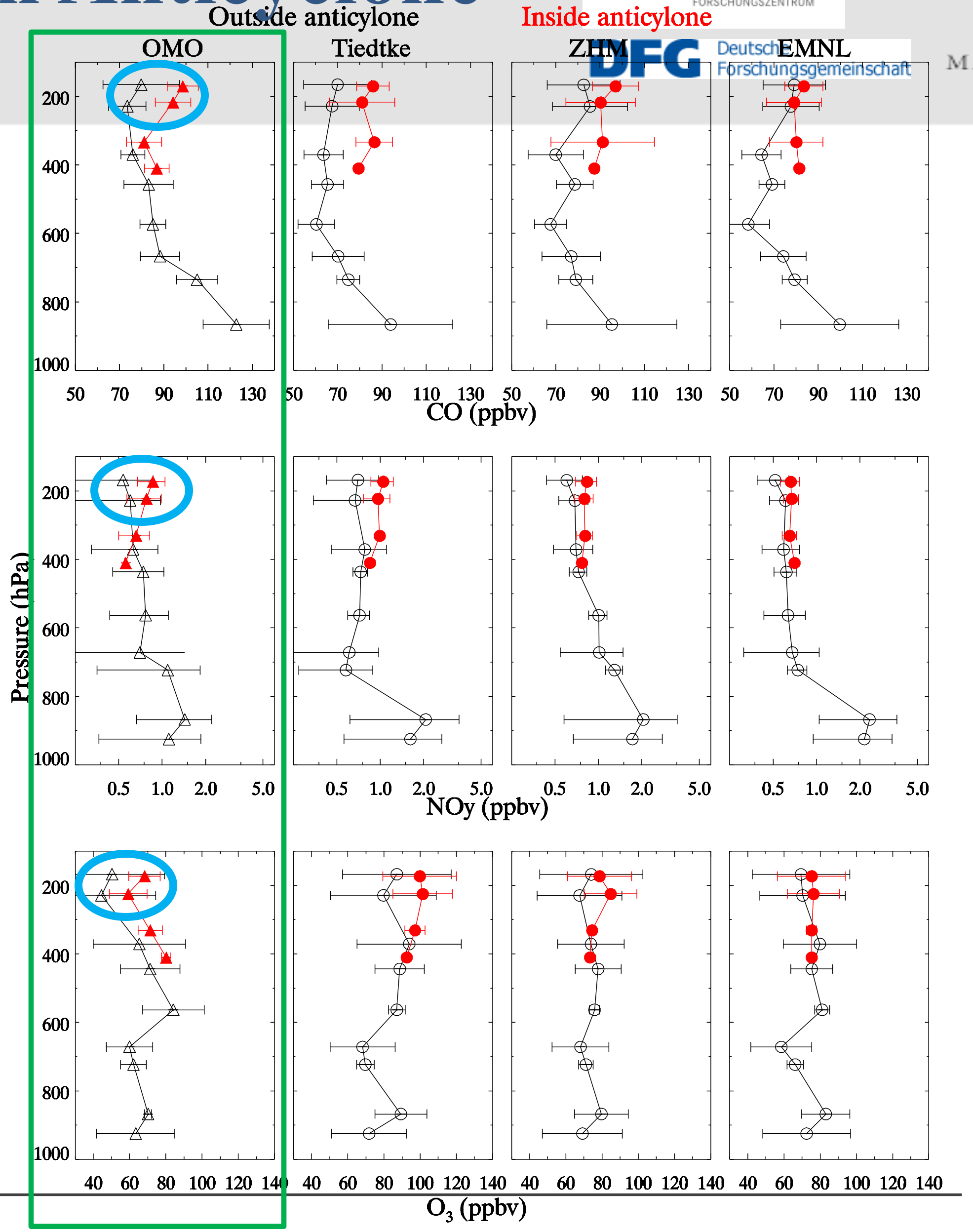
$$r = \frac{S(\mathrm{OH})}{P(\mathrm{OH}) + S(\mathrm{OH})}$$

Impact of Monsoon Anticyclone

- CH_4 used as a tracer to segregate the observations. **Inside anticyclone**

  $CH_4 > 1879.8$ ppbv

- Tiedtke shows better agreement with OMO, especially near 200 hPa (altitude of stronger impact).



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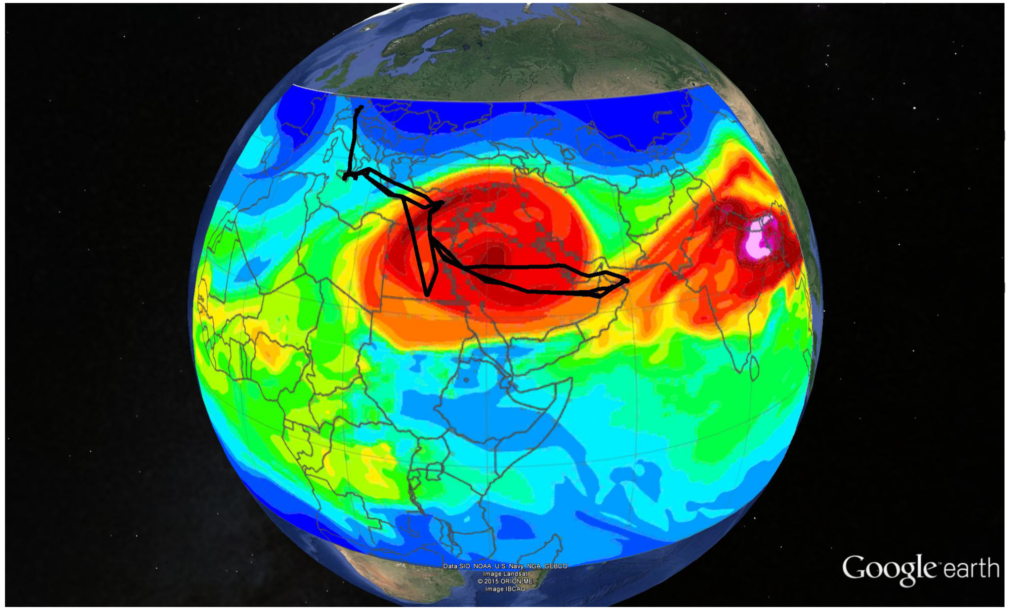
Cyprus 13.8.-27.8.

Strong western anticylone over the Arabian peninsula

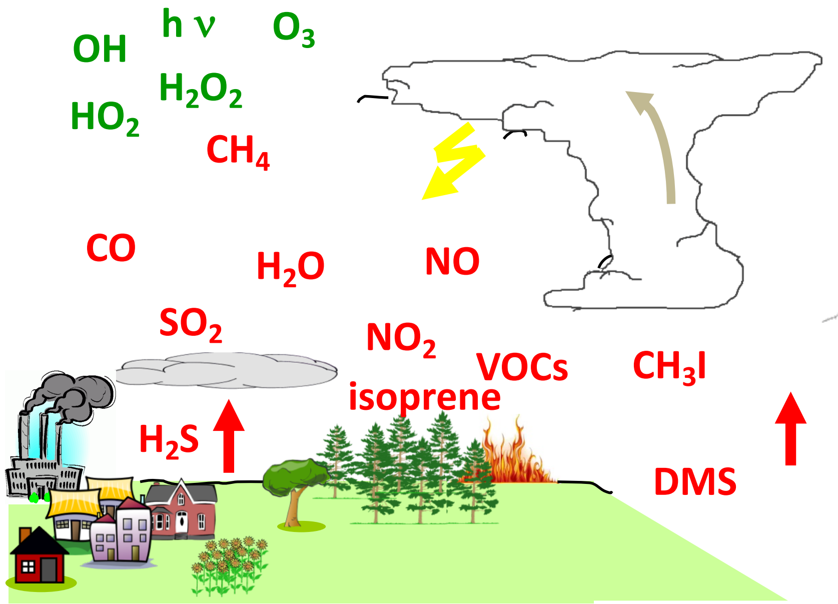
Transect flights through the photochemical aged airmass

→ Enhanced CO, enhanced particles (CN), oxygenated VOC,

CAMS forecast from Sunday 23 August 2015 00Z valid at T+054: Tuesday 25 August 2015 06Z
Global CO Tracer (ppbv) at 150 hPa



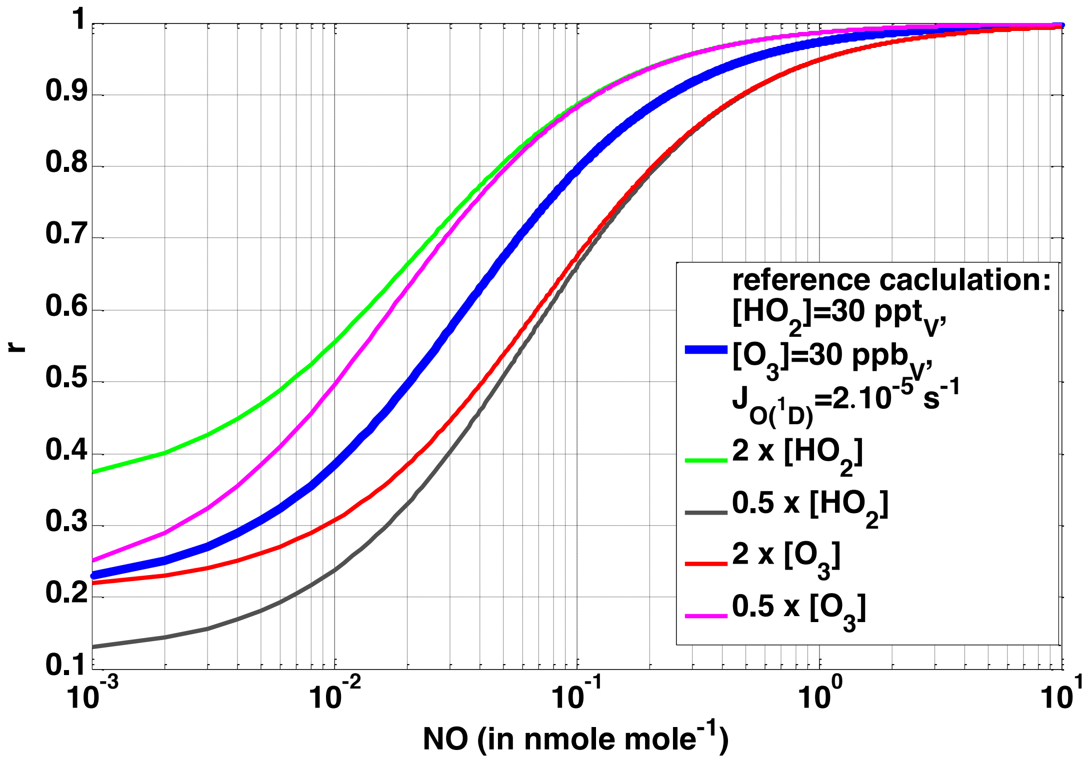
Oxidation capacity influenced by Convection

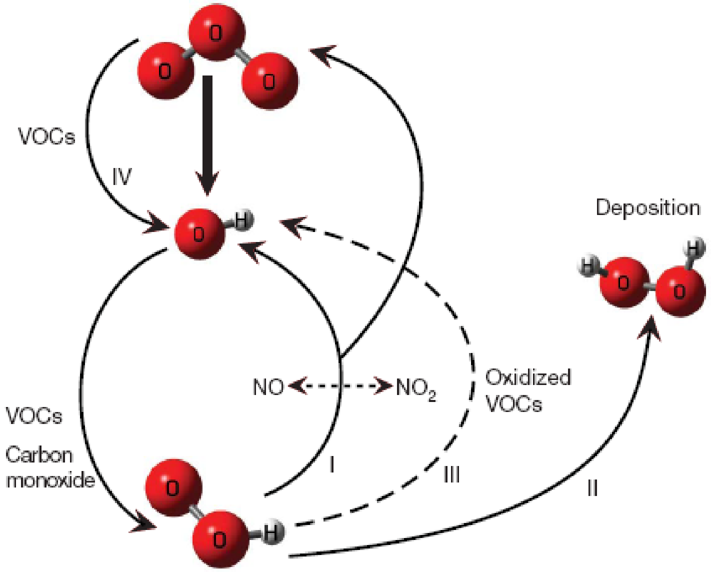


OH recycling probability and cycling-lifetime

$$r = \frac{S(\mathrm{OH})}{P(\mathrm{OH}) + S(\mathrm{OH})}$$

$$n = -\frac{1}{\ln(r)}$$





$$r_{HUMPPA}^{median} = (0.86 \pm 0.05)$$

$$n_{HUMPPA}^{median} = (6.6 + 4.0 / -1.9)$$

Lelieveld et al. (2002)

- Lifetime of NOx about xx days
- Continous injection of NO keeps up the oxidation capacity

Cyprus 13.8.-27.8.

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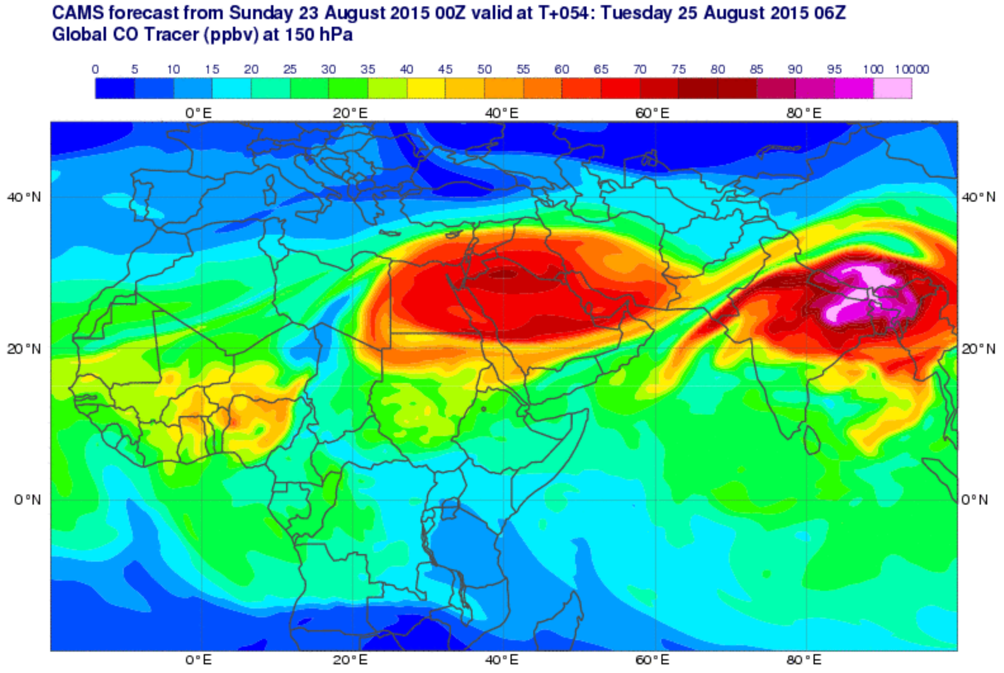
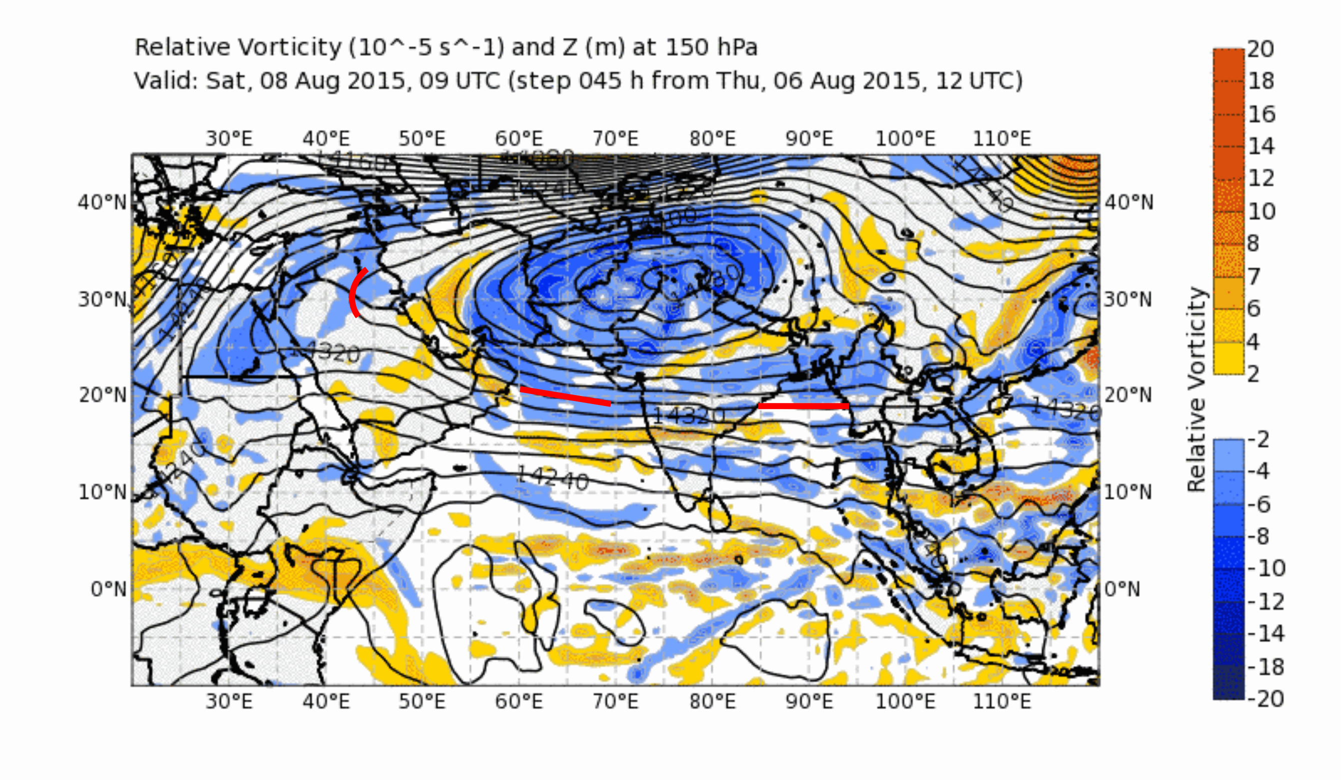
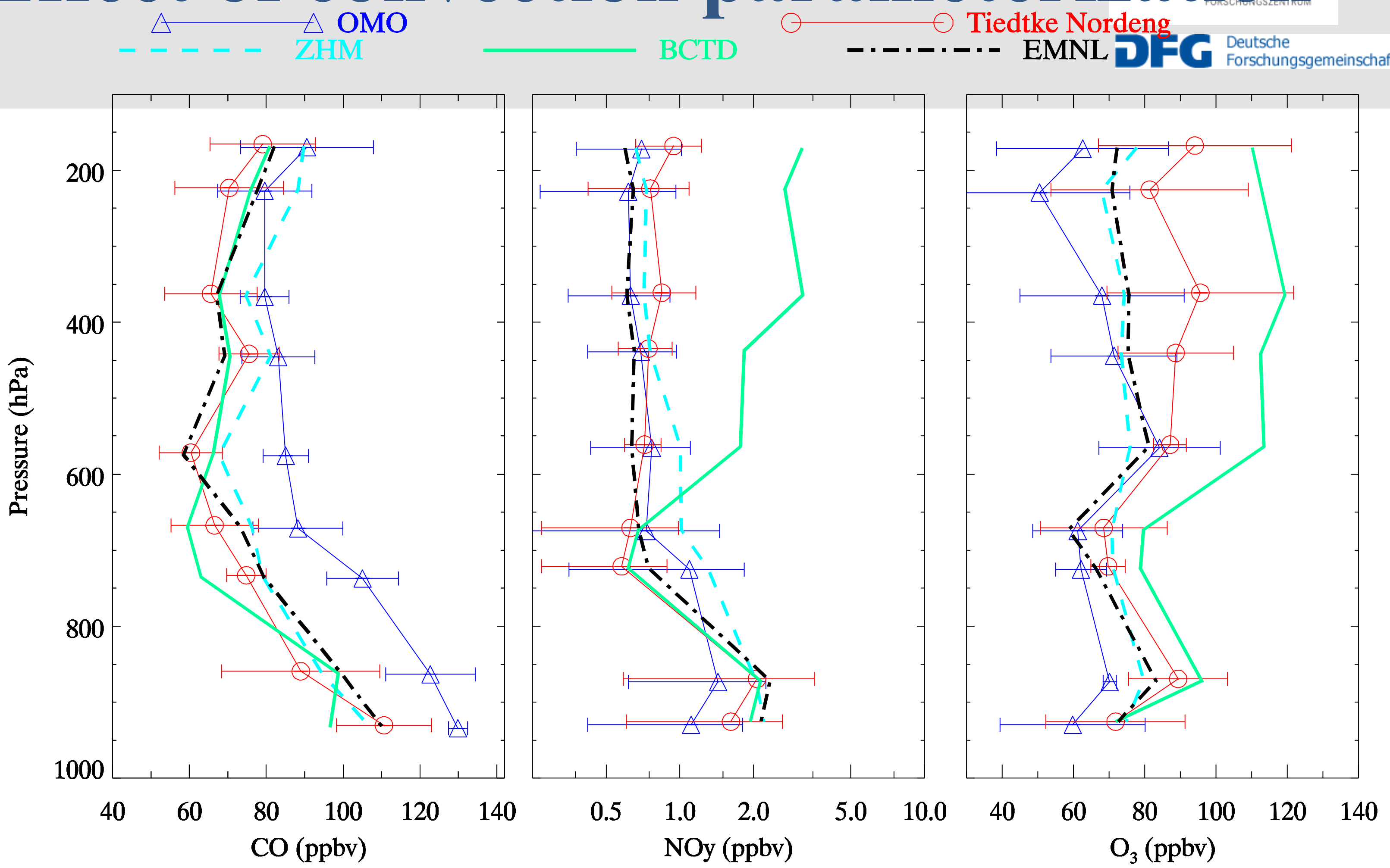


Gan 6.8. – 9.8.



Hans Schlager

Effect of convection parameterization

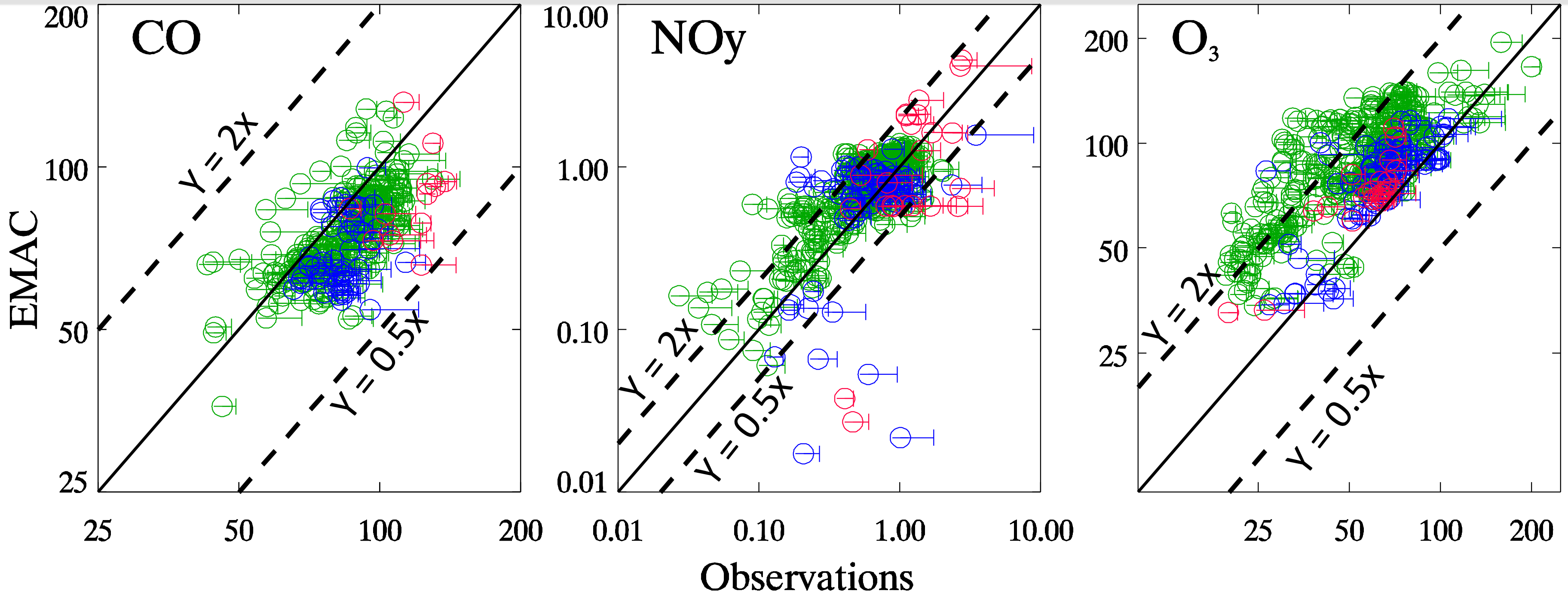


- Choice of convection schemes considerably influence modelled distributions.
- All the schemes capture the typical C-Shape structure of CO distribution.
- BCTD convection predicts too high NOy and O_3.

Model Evaluation

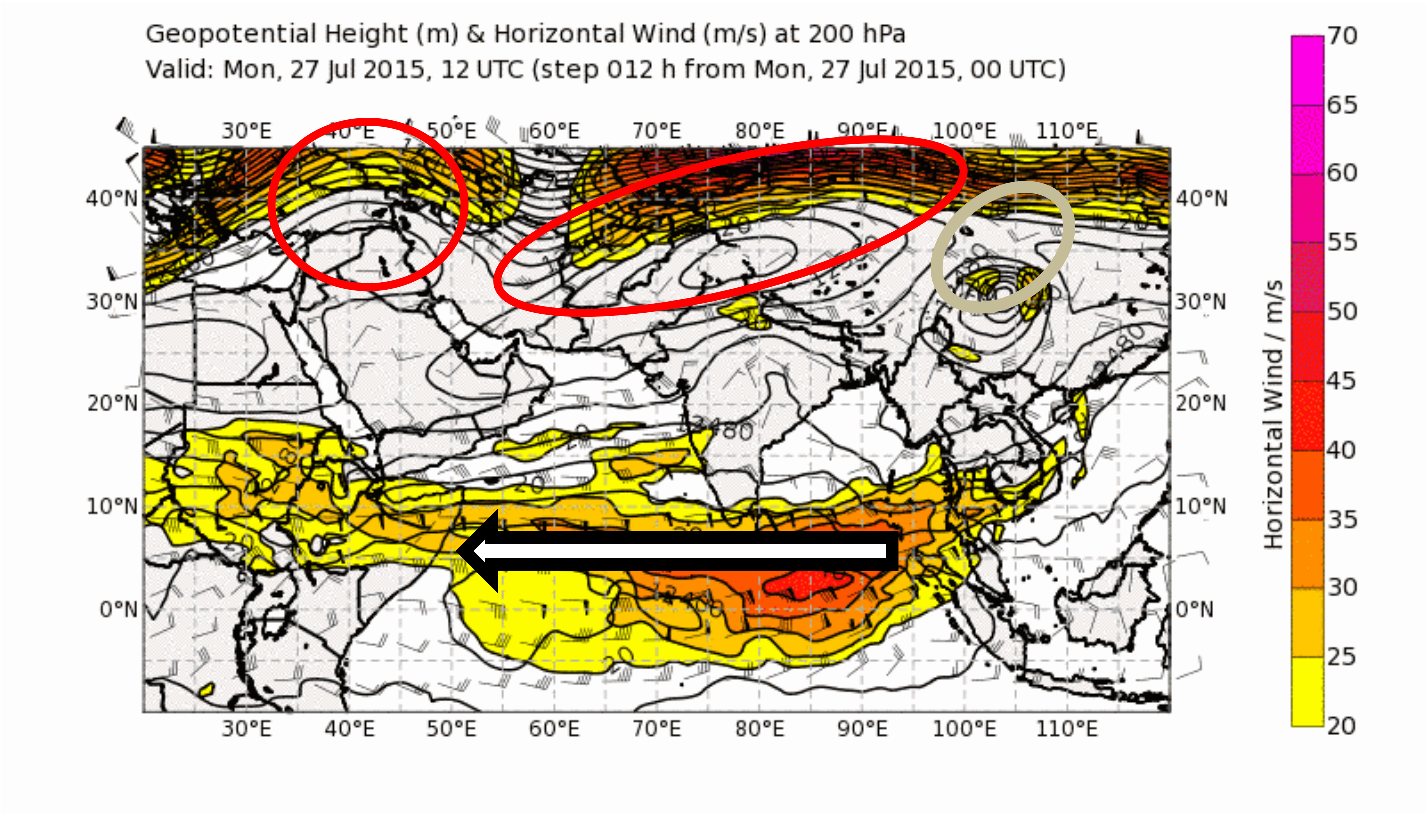
p > 700 hPa | p = 700-300 hPa | p < 300 hPa | All data

r =	0.30	0.29	0.69	0.64	0.57	0.43	0.68	0.62	0.80	0.74	0.72	0.68



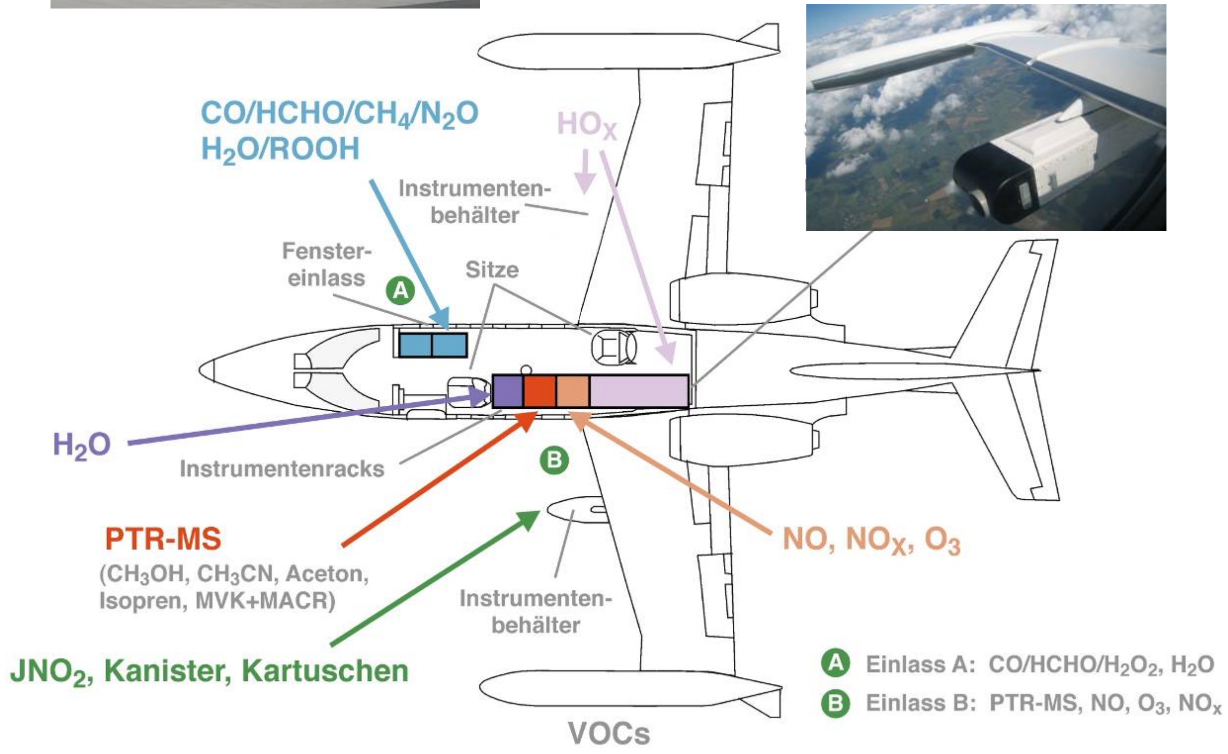
- ➢ The reference simulation reproduces the spatio-temporal variations to some extent (r= 0.62 to 0.68).
- ➢ Model performance is better in the Upper Troposphere (UT).

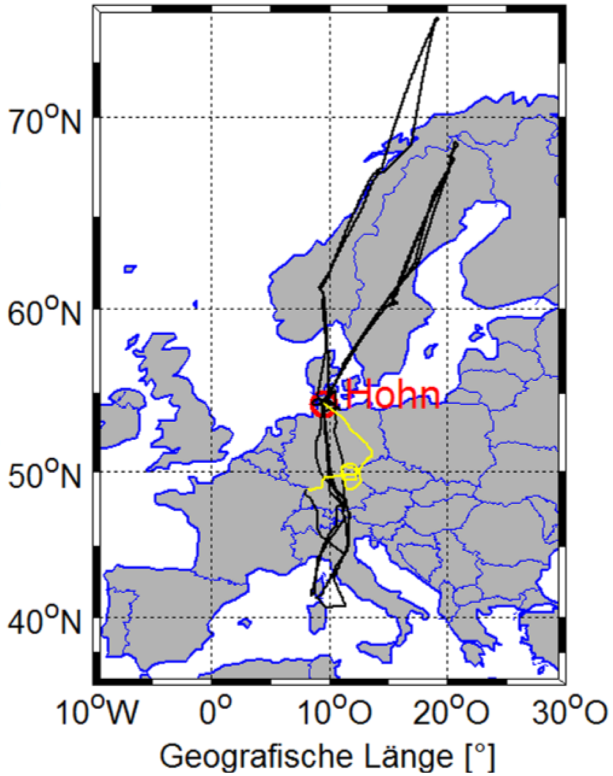
Cyprus 25.7. & 28.7.



Hans Schlager

Small aircraft instrumentation





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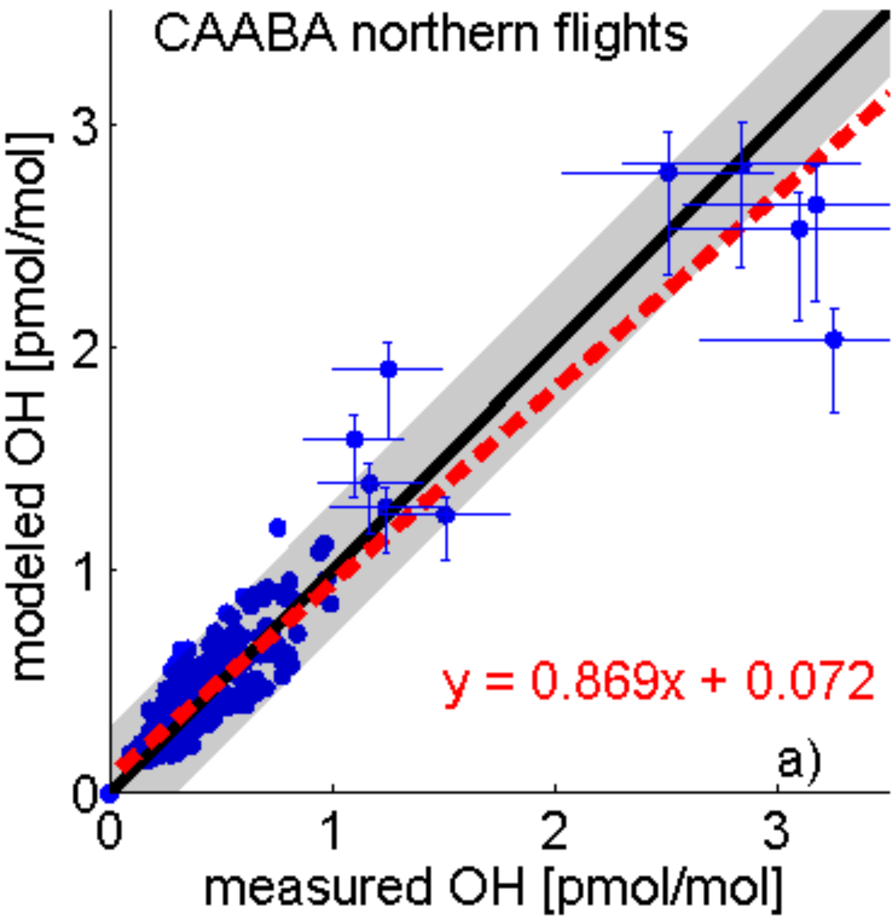
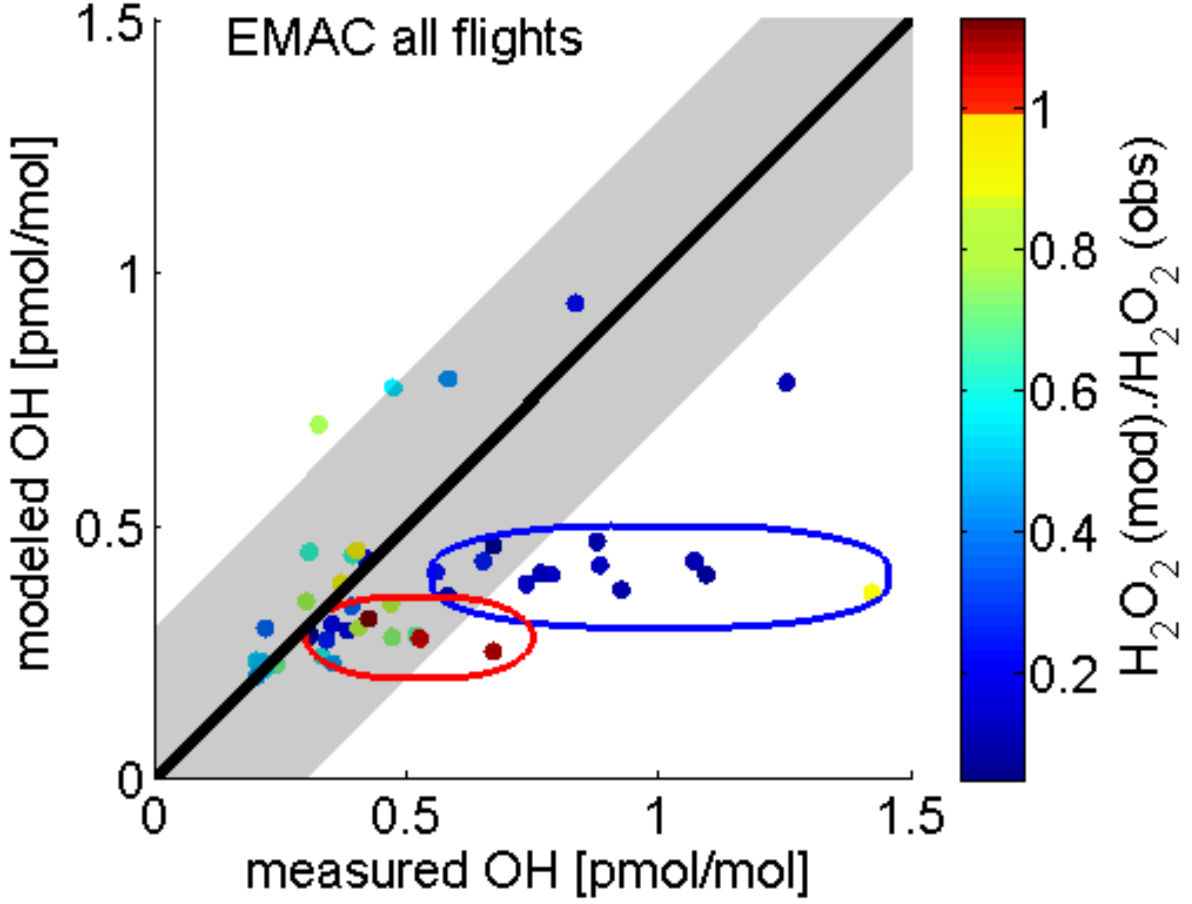
Measurement vs. Box model

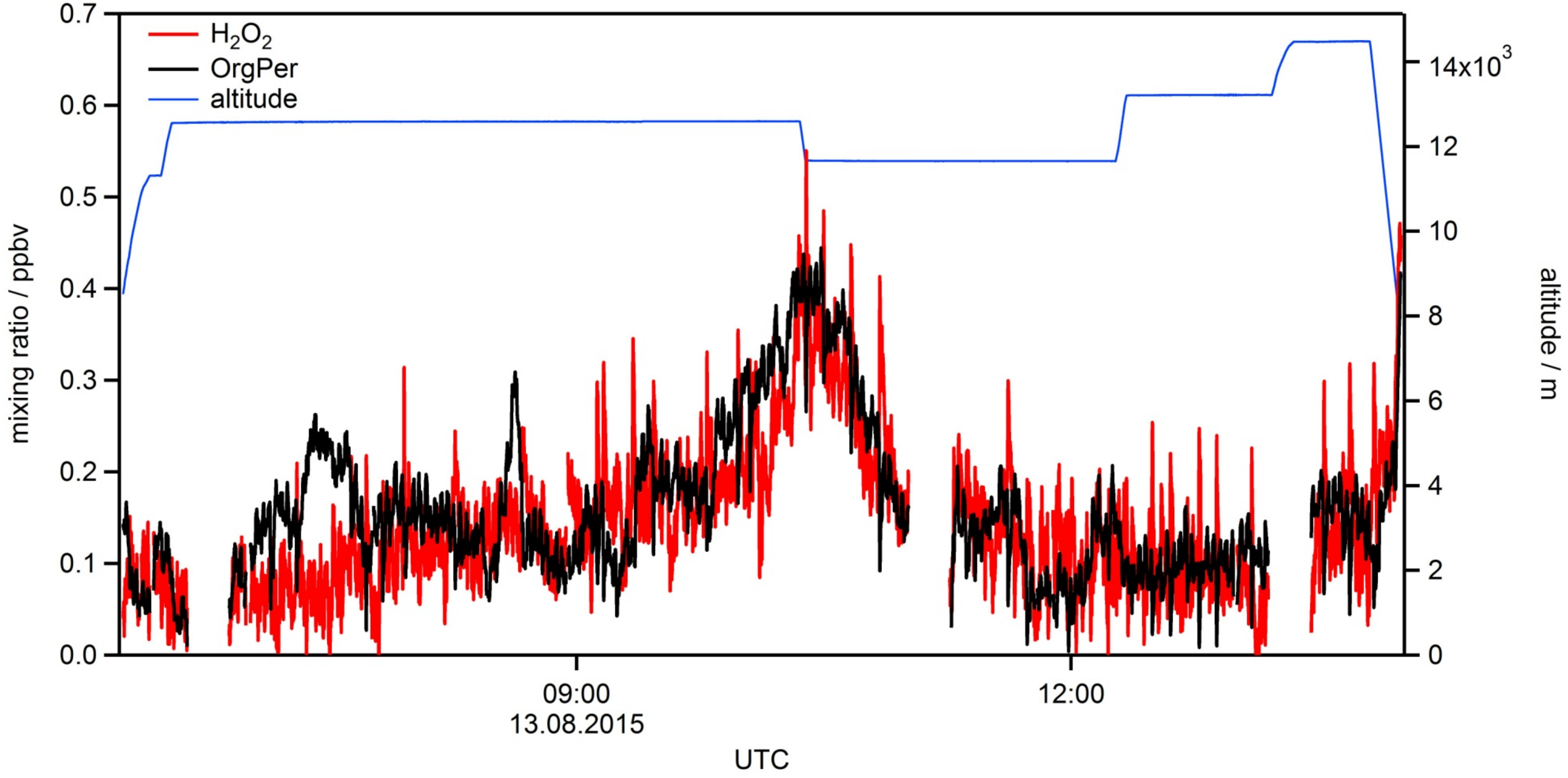


Regelin et. al

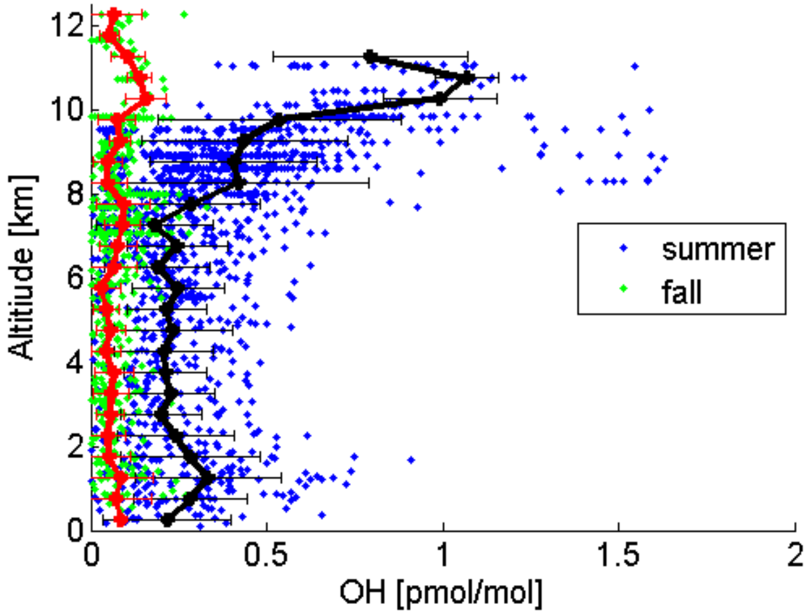
Measurement vs. global model



Flugnummer #19

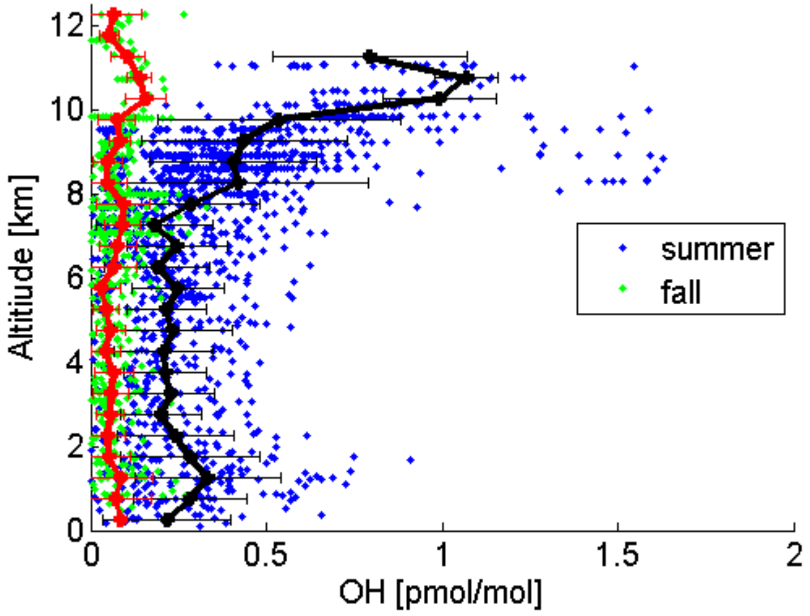


Oxidation capacity



- Larger OH- and HO_2 mixing ratio in summer
- C-shaped distribution of OH in summer
- Strong elevated OH mixing ratio above 10 km

Oxidation capacity

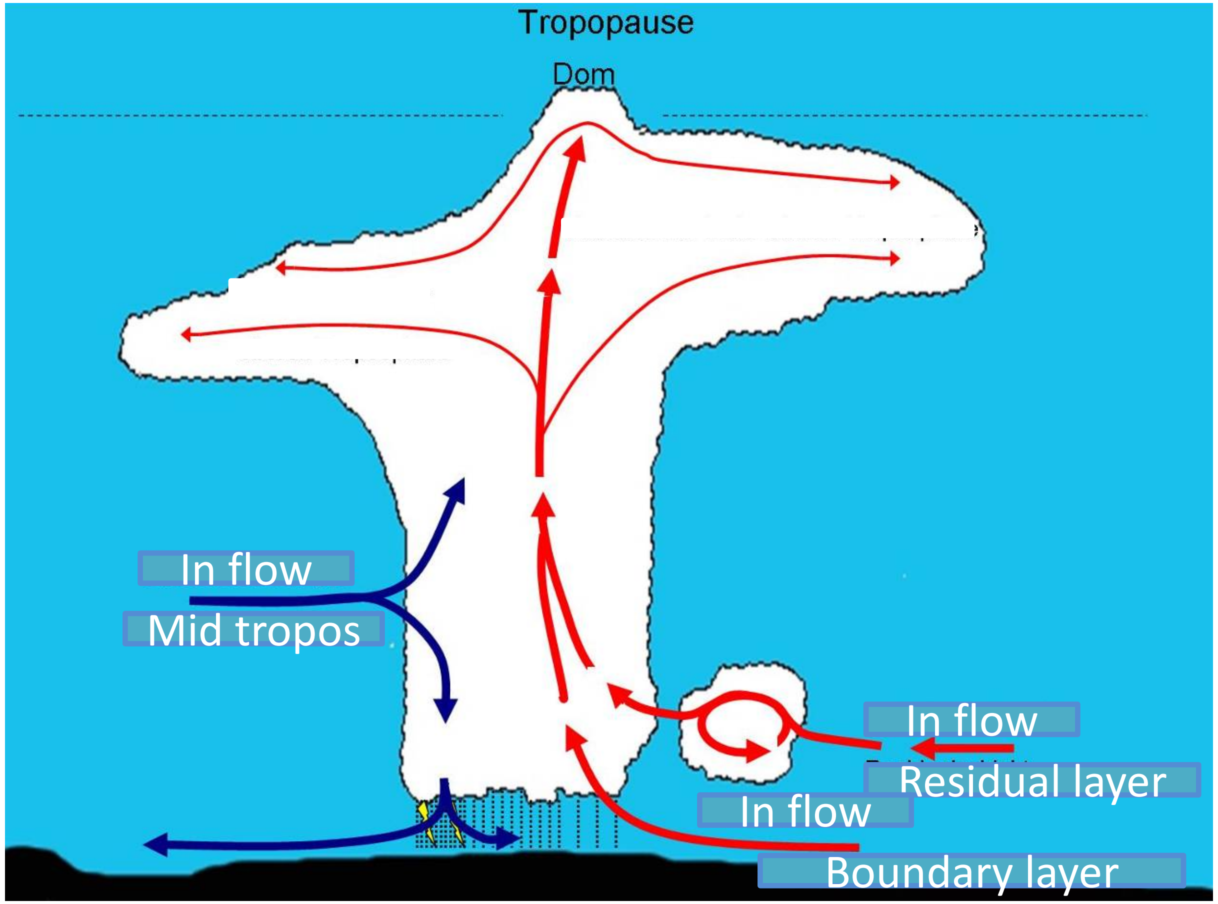


- Larger OH- and HO_2 mixing ratio in summer
- C-shaped distribution of OH in summer
- Strong elevated OH mixing ratio above 10 km

CH_4-life time (days) in fall and summer in the upper troposphere		
altitude [km]	7 - 7.5	10 - 10.5
Fall	6906	11372
Summer	2947	1349

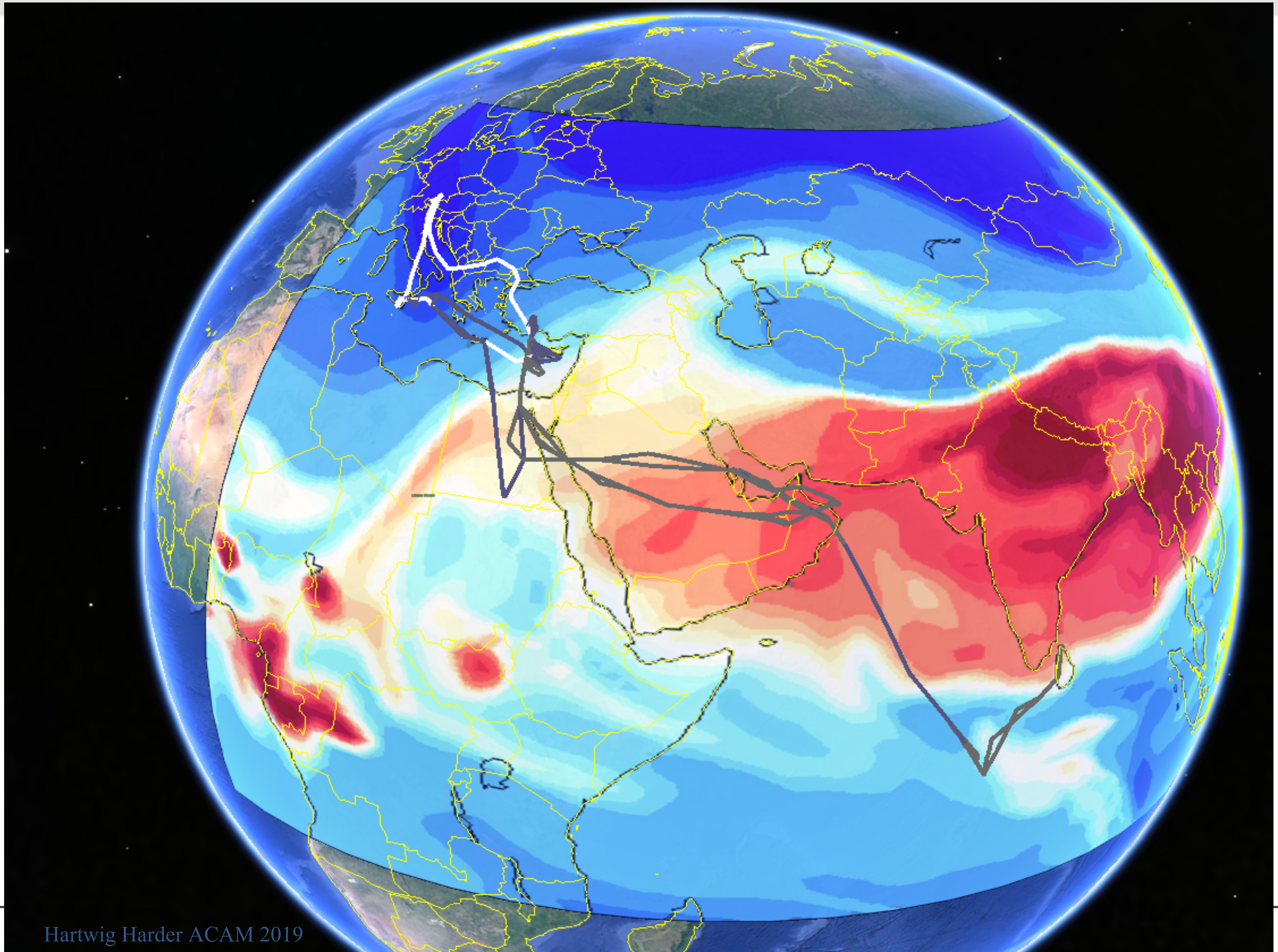
Possible reason for larger oxidation capacity in summer			
Flughöhe [km]	OH [Molecule/cm^3/s]	HO_2 [Molecule/cm^3]	NO [Molecule/cm^3]
7 - 7.5	$0.25 \cdot 10^6$	$1.37 \cdot 10^8$	$0.19 \cdot 10^9$
10 - 10.5	$3.4 \cdot 10^6$	$0.80 \cdot 10^8$	$4.07 \cdot 10^9$

Convection



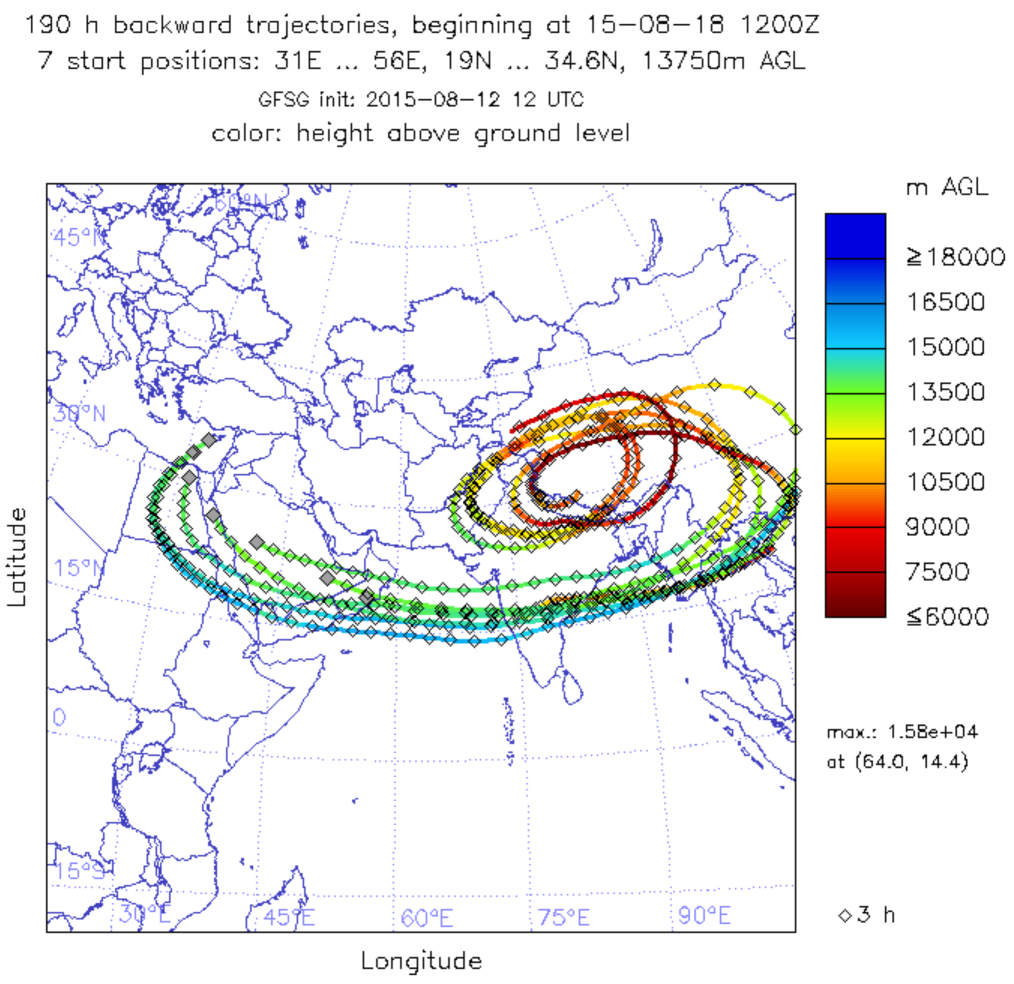
Tropopause
Dom
In flow
Mid tropos
In flow
Residual layer
In flow
Boundary layer

Where Did We Fly?



MAX-PLANCK-INSTITUT FÜR CHEMIE

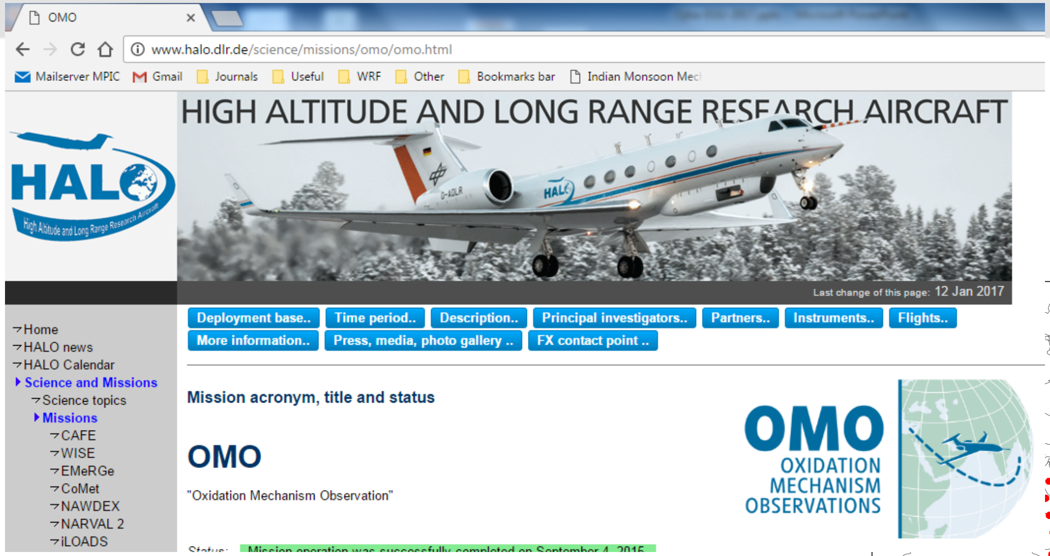
MAX-PLANCK-GESELLSCHAFT

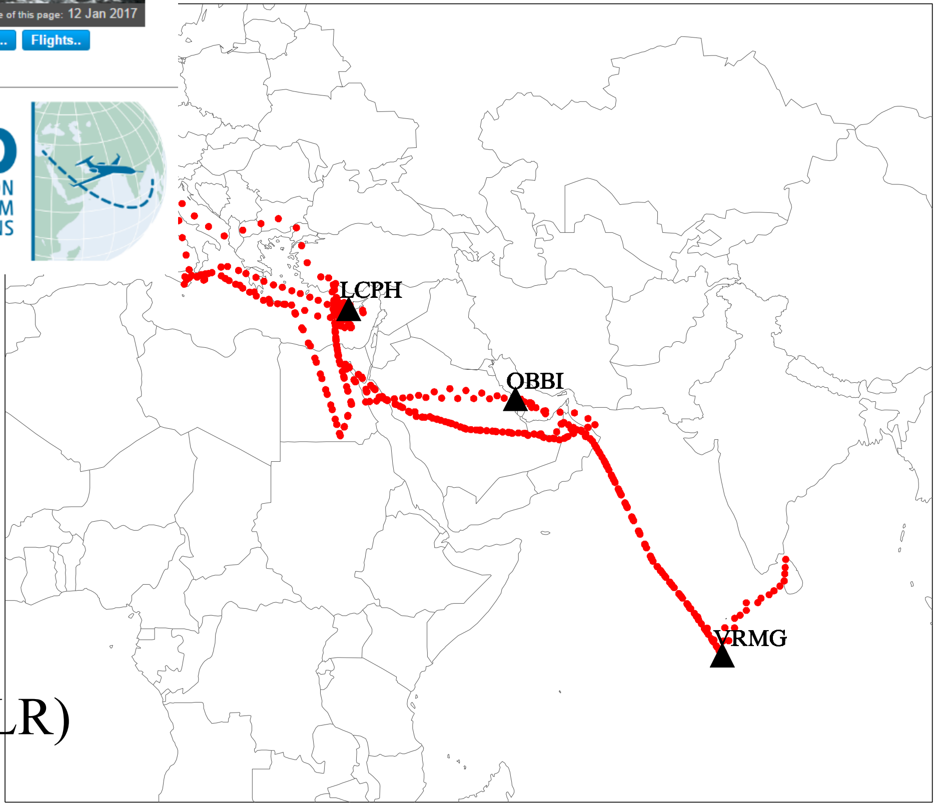


Hans Schlager

Oxidation Mechanism Observations (…015)

http://www.halo.dlr.de/science/missions/omo/omo.html





- OMO period: July-August 2015
- Here we analyze
  - O_3 measured using FAIRO (KIT)
  - CO, CH_4 (MPIC)
  - NOy measured using IPA-NOy (DLR)

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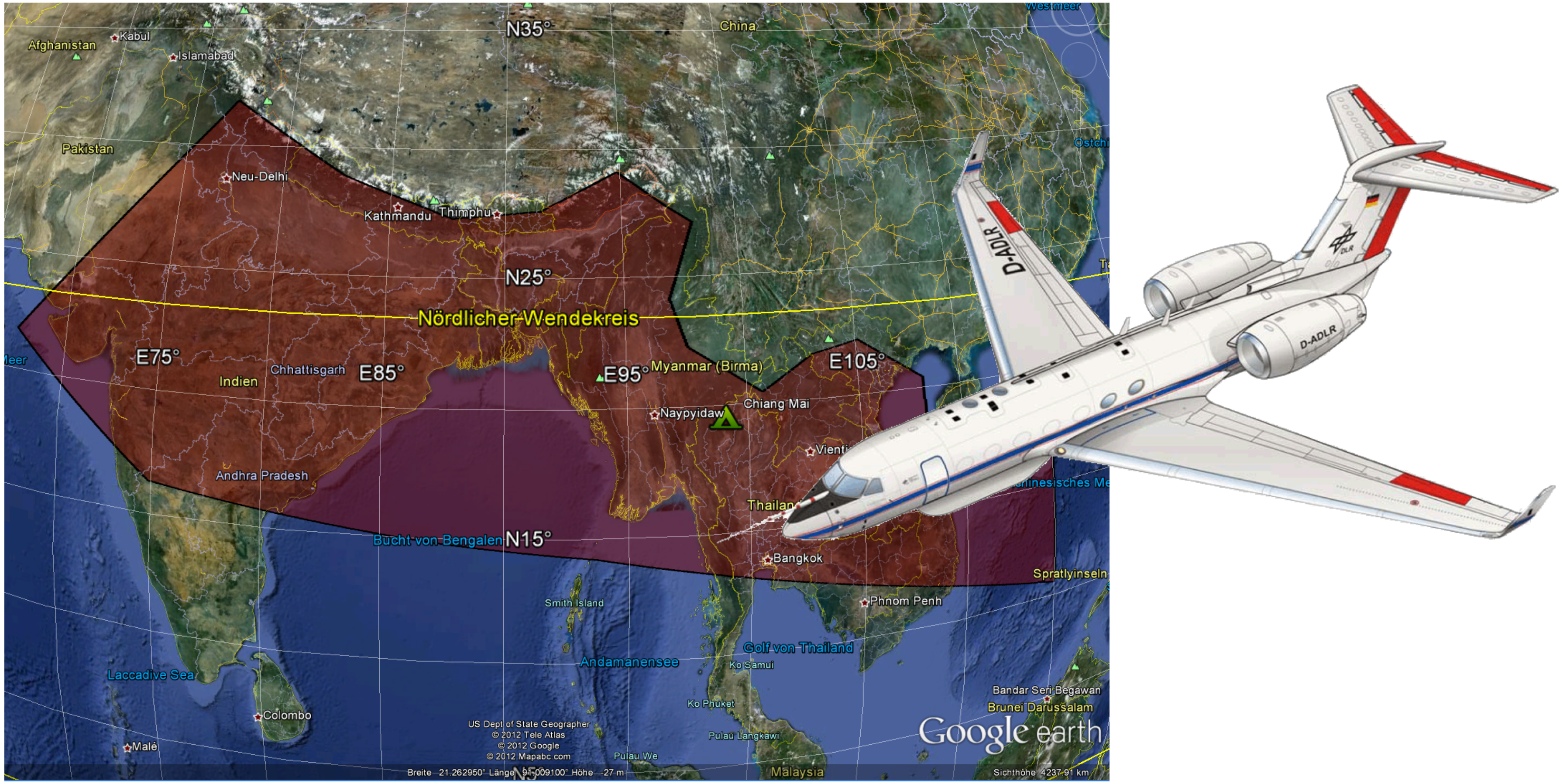
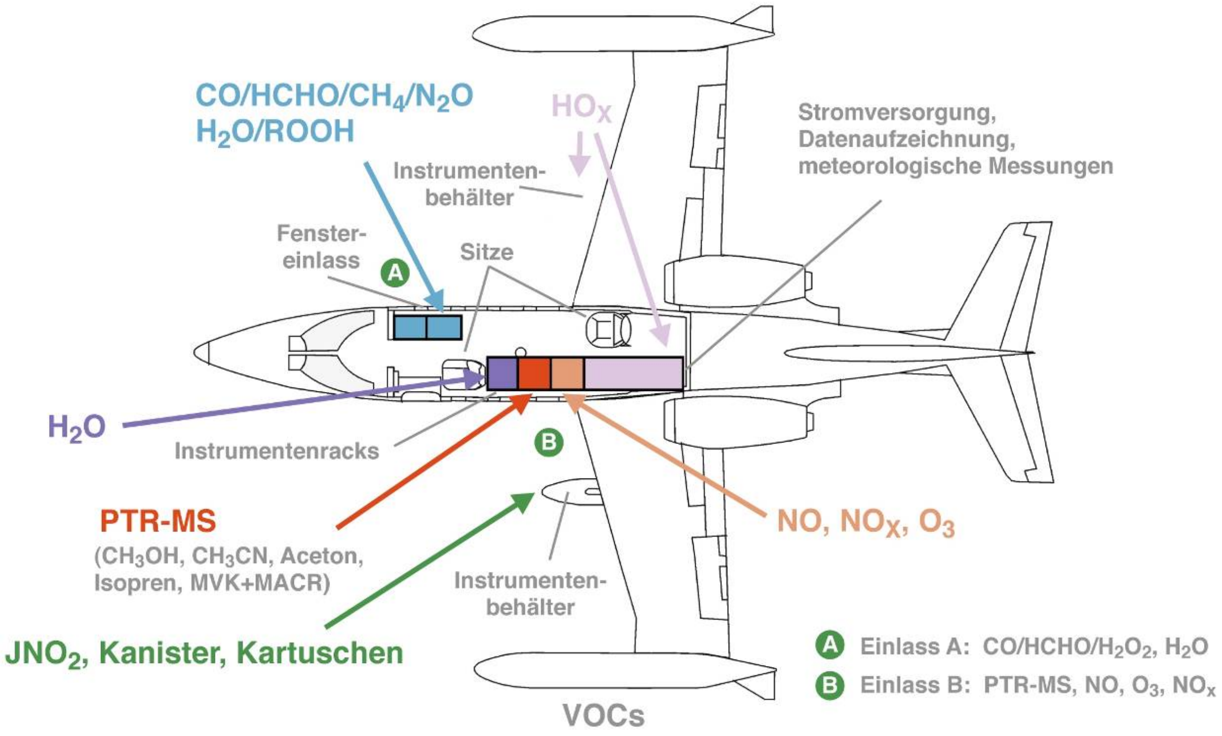
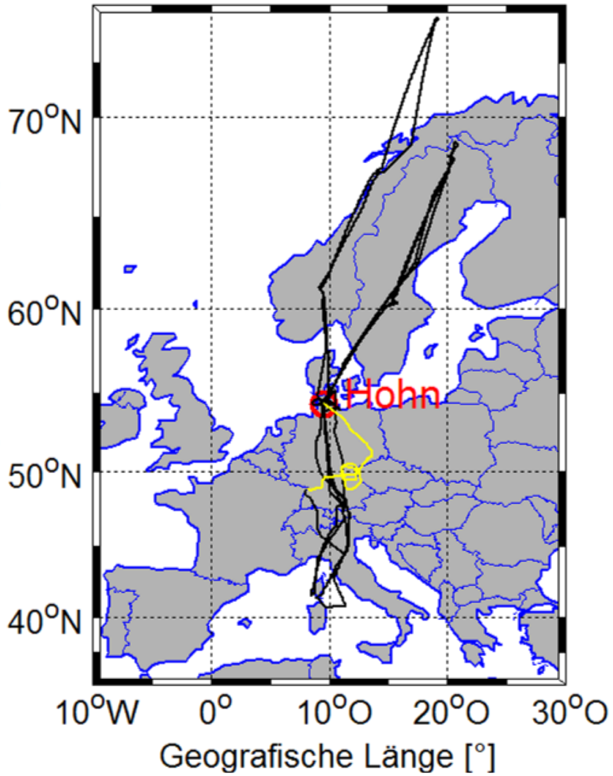
OMO
Oxidation capacity influenced by Monsoon Outflow



Small aircraft instrumentation





Convection and OH & HO2

JÜLICH FORSCHUNGSZENTRUM
DLR
KIT Karlsruher Institut für Technologie
DFG Deutsche Forschungsgemeinschaft
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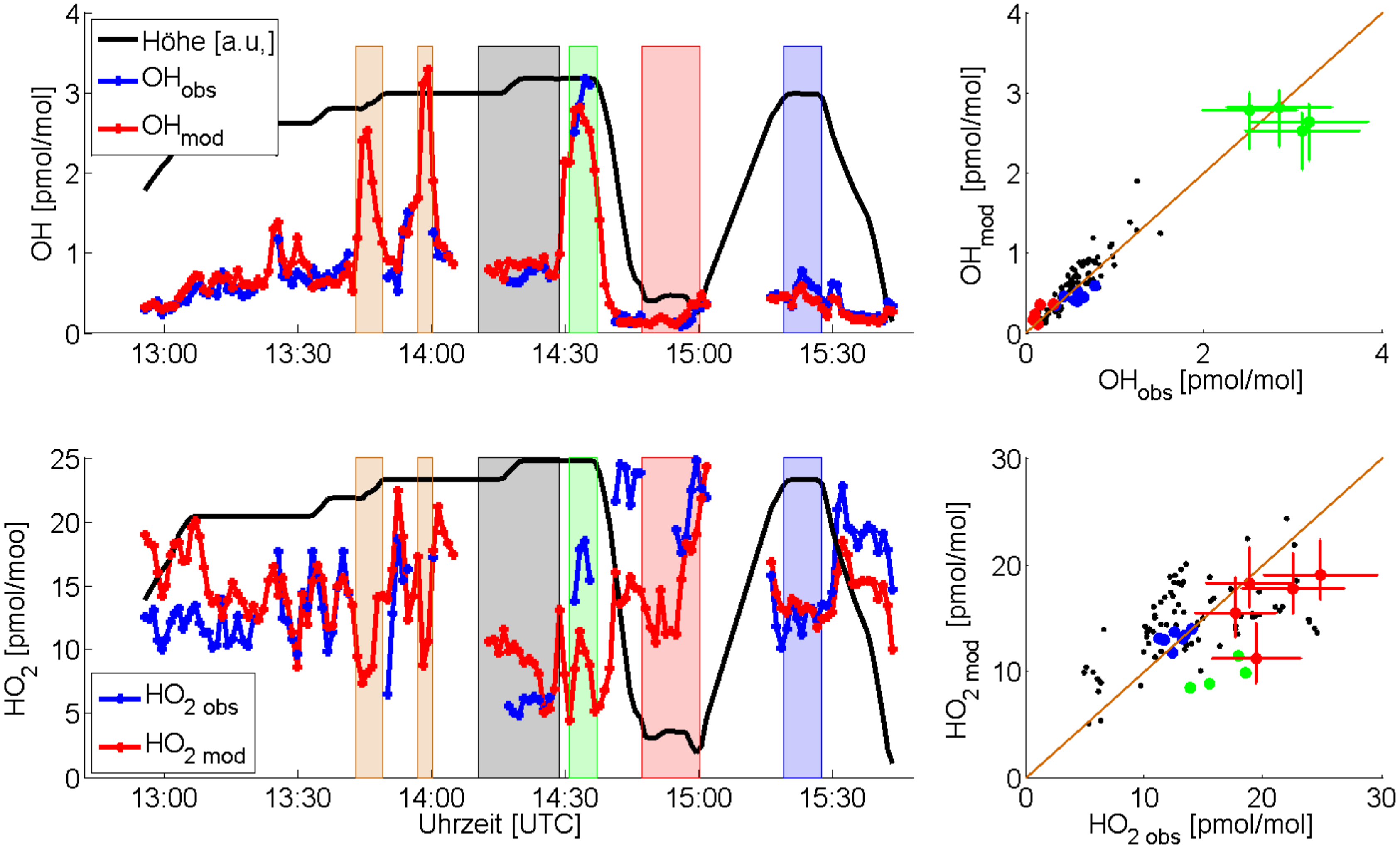
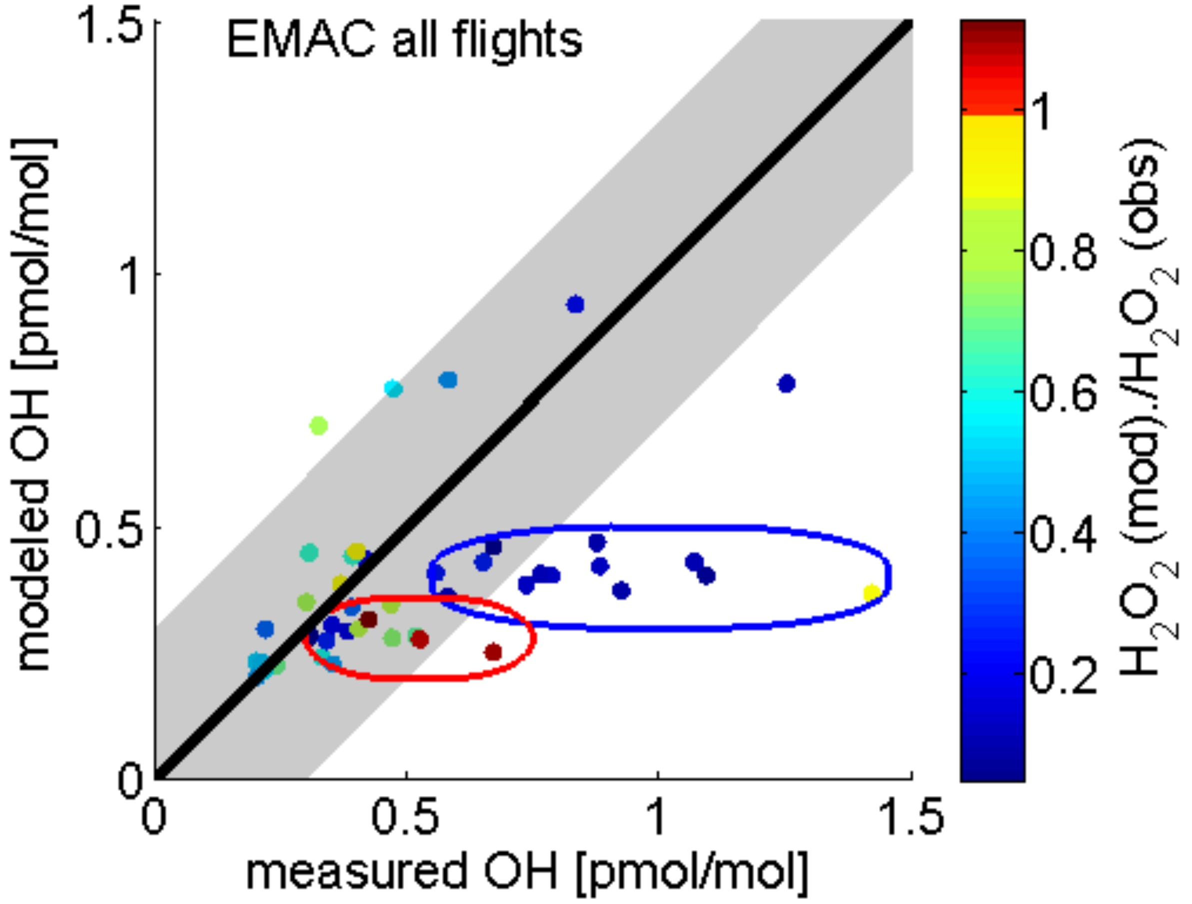
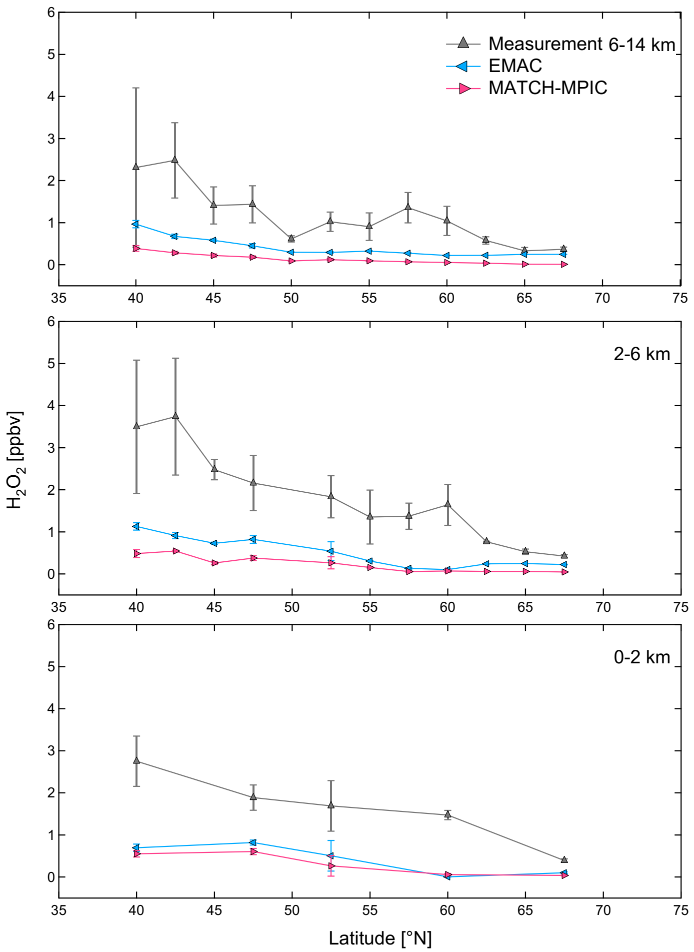


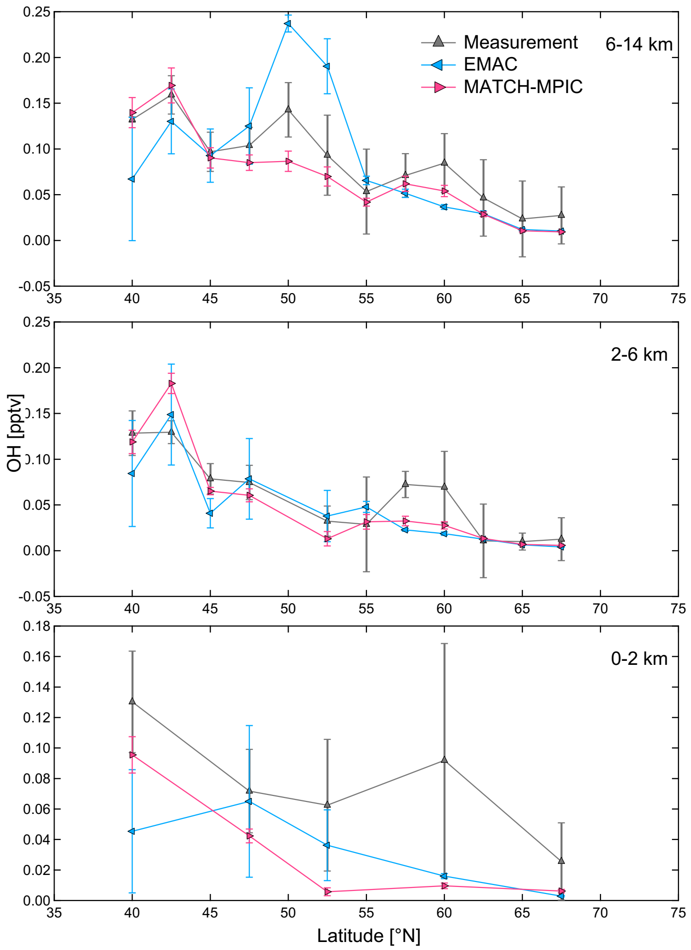
MAX-PLANCK-GESELLSCHAFT

Measurement vs. global model



Measurement vs. global model



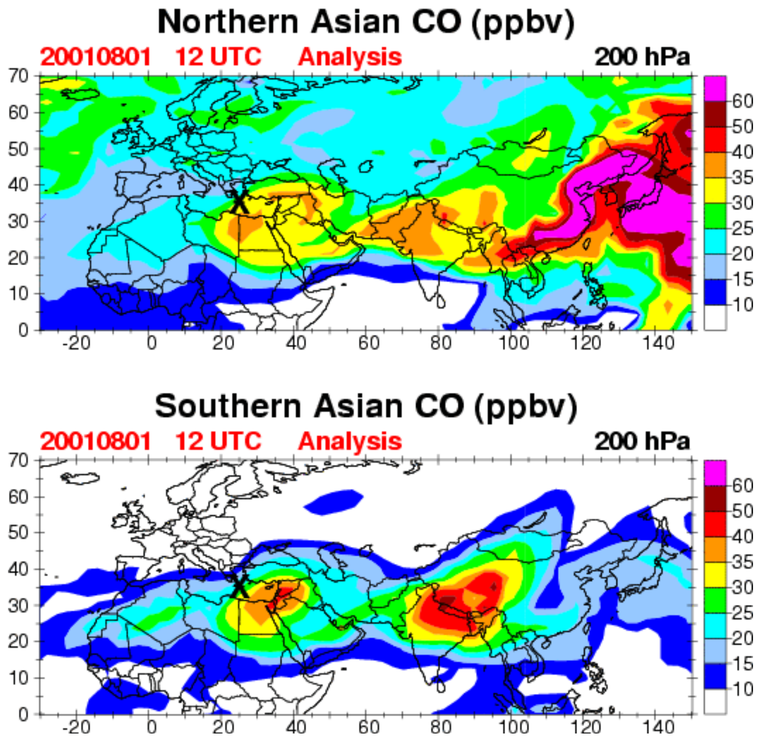


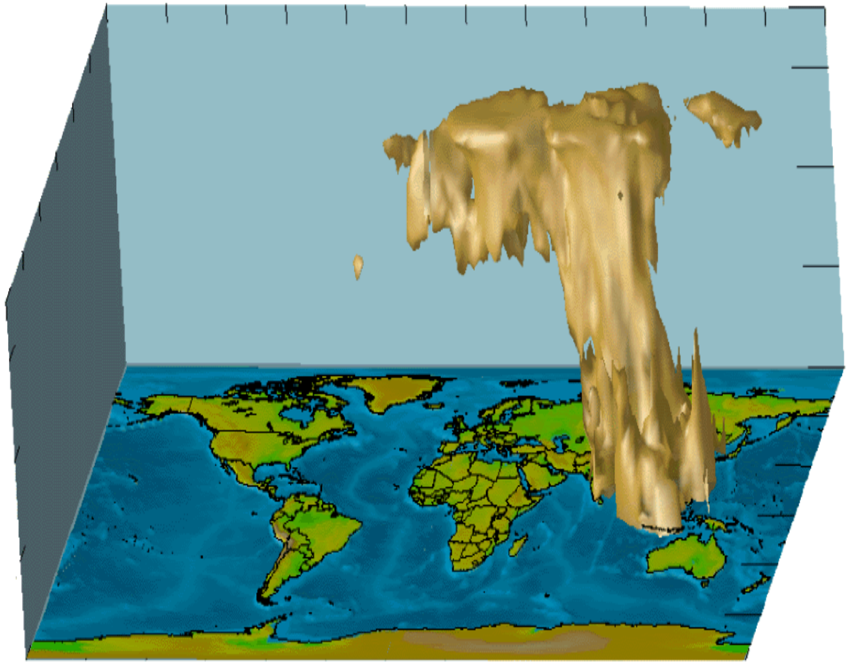
Summary

- Convection is an important process changing the trace gases and the oxidation capacity in the upper troposphere
- Prescribed box model calculates OH concentration acurately
- Global model underestimates OH mainly due to underestimating peroxides

- Vertical transport in global model well described ?
- Uptake of peroxides on ice/water droplets in these well described ?

Monsoon & convection

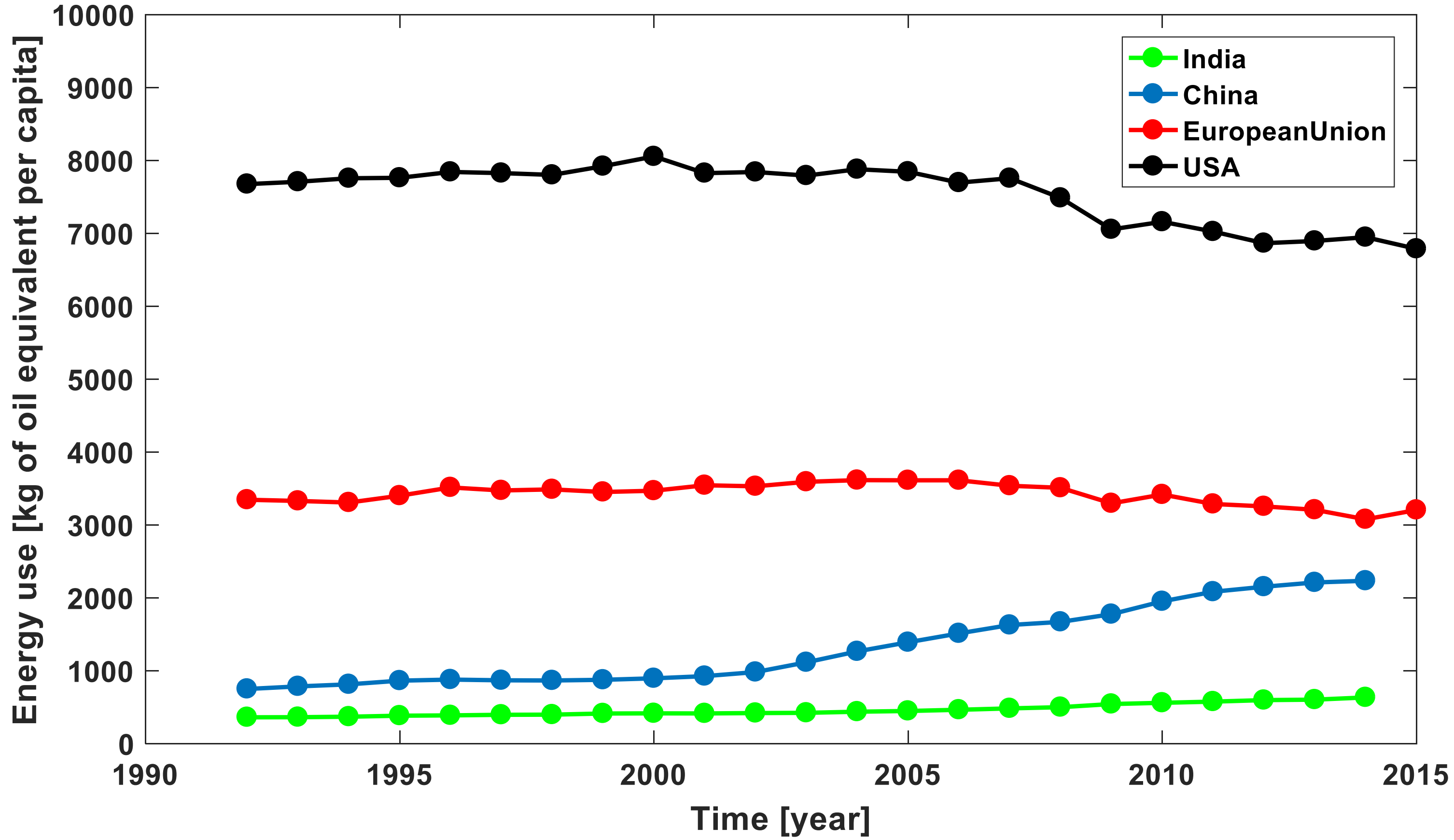




OMO Instrumentation

- Actinic Flux
- OH/HO2/RO2
- O3/CO/H2O
- NO/NOy/PAN
- VOC/OVOC/HCHO
- H2O2/org. Peroxides

Energy use per capita

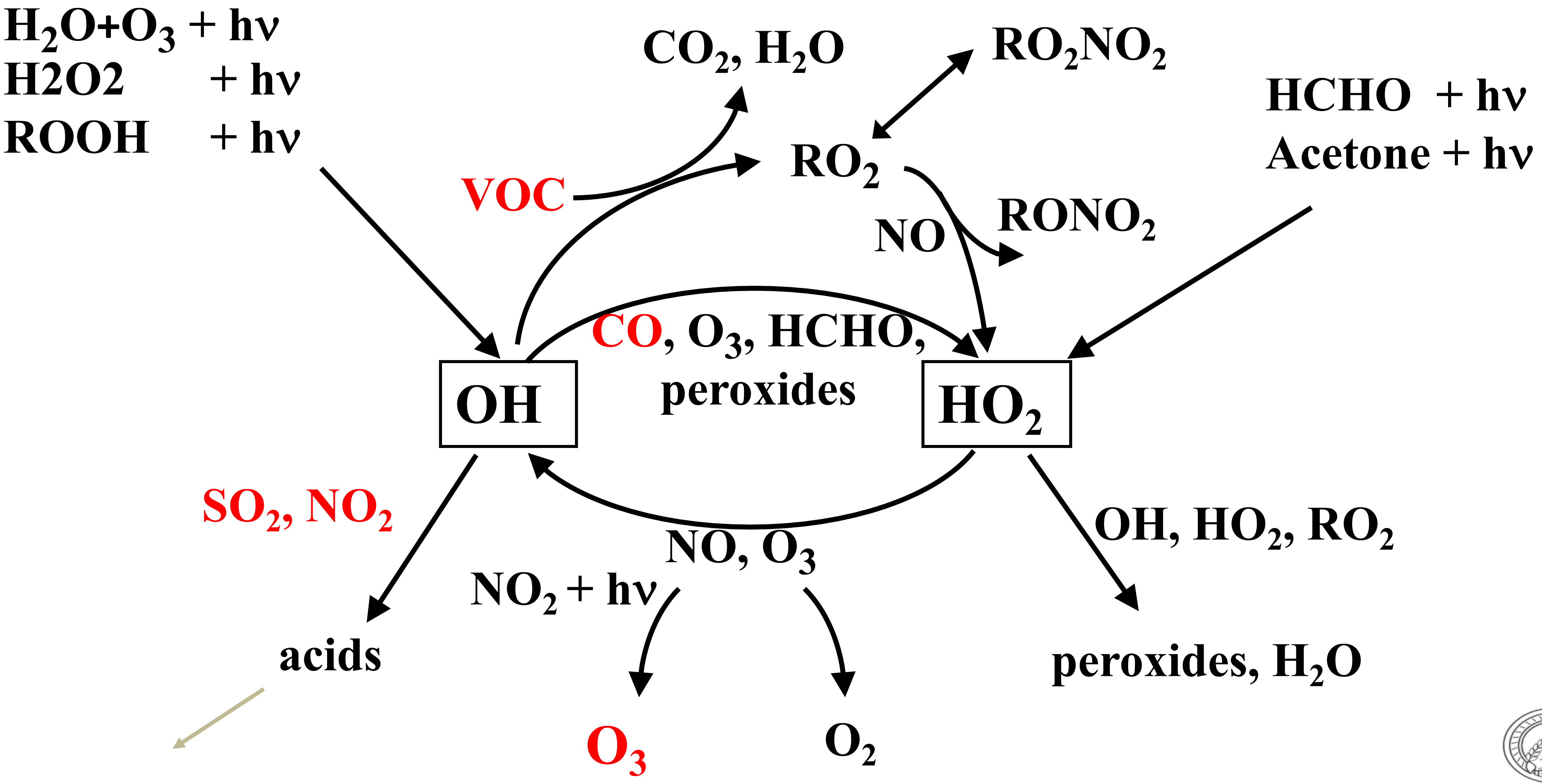


International Energy Agency

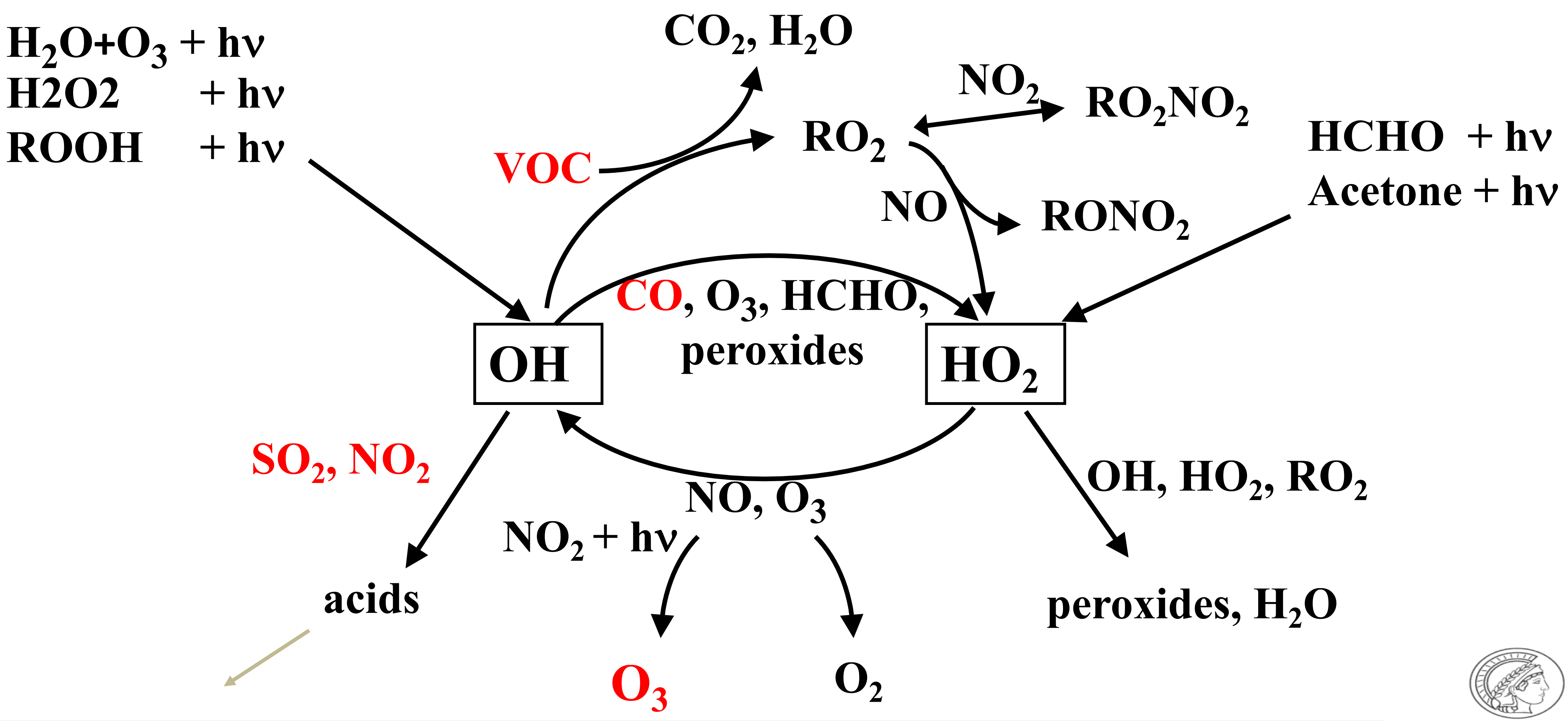
Region of growth

- South Asia, booming economy and solid growth in population
- Increasing standard of living -> needs on energy per capita

Simplified HOx chemistry

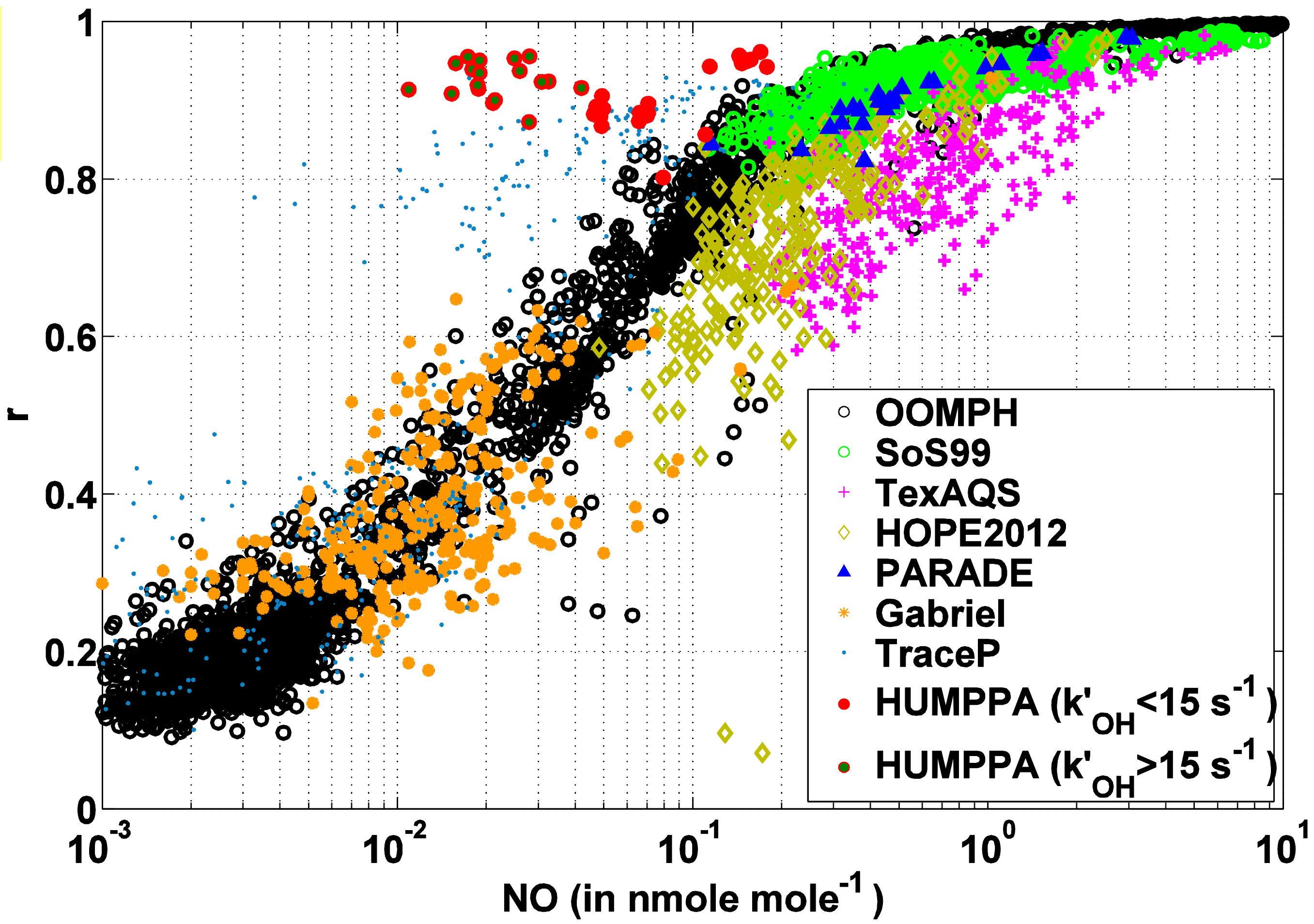


Simplified HOx chemistry



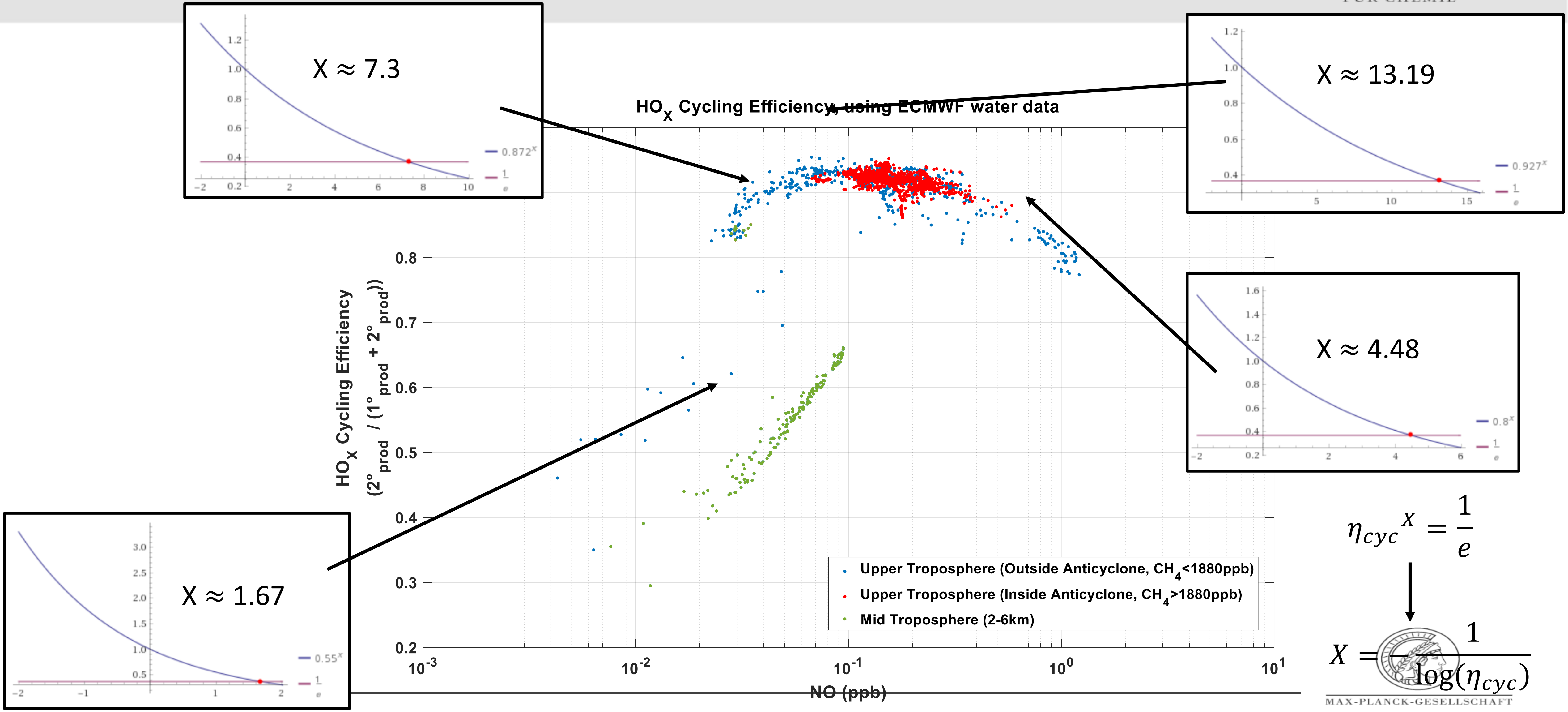
OH recycling probability as a function of NO in different environments

$$r = \frac{S(\mathrm{OH})}{P(\mathrm{OH}) + S(\mathrm{OH})}$$



Hens & Harder, 2014

HO_X cycling efficiency inside outside of A



$$\eta_{cyc}{}^{X} = \frac{1}{e}$$

$$X = \frac{1}{\log(\eta_{cyc})}$$

Contribution of South and East Asian Emissions

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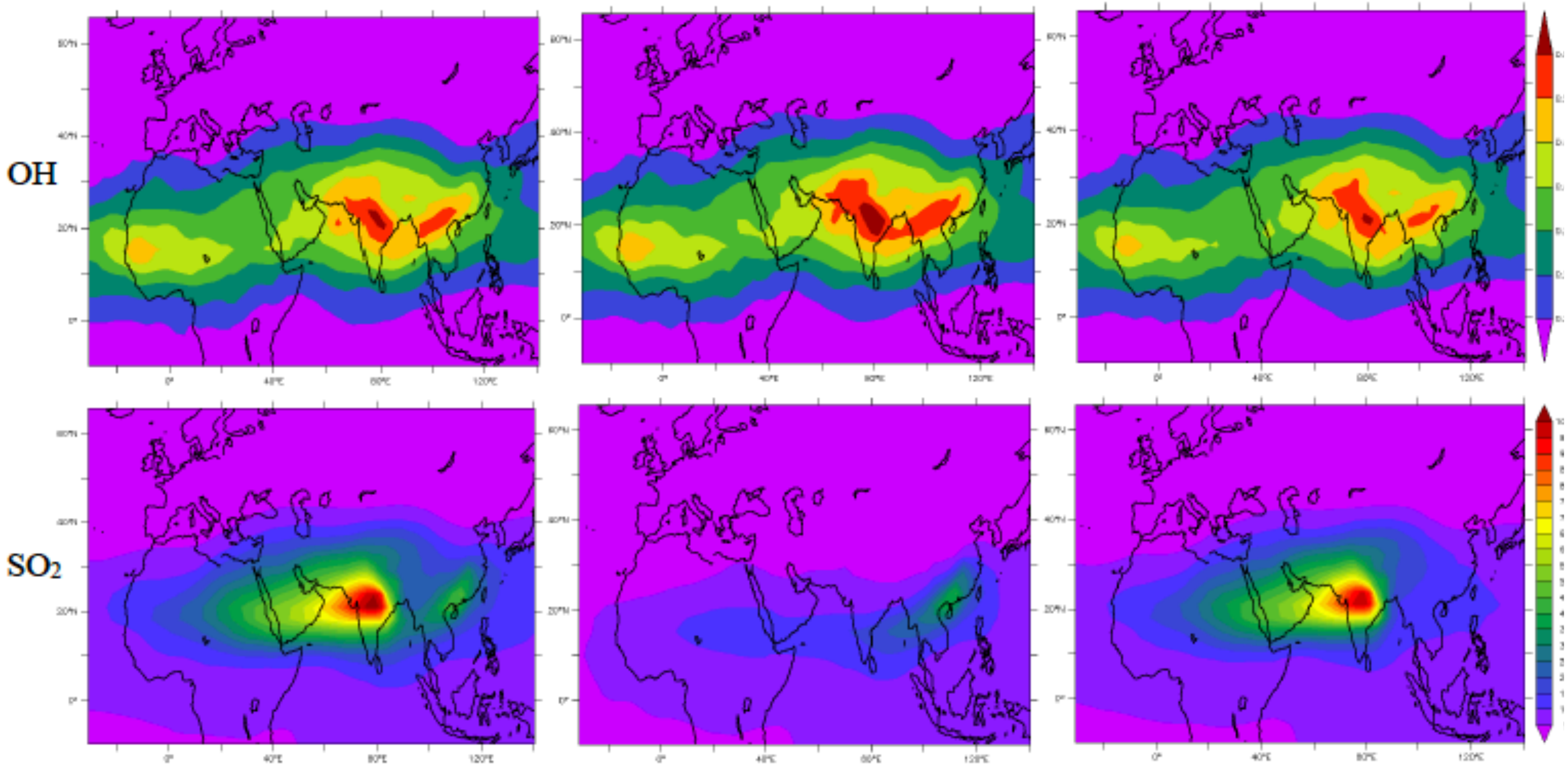

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