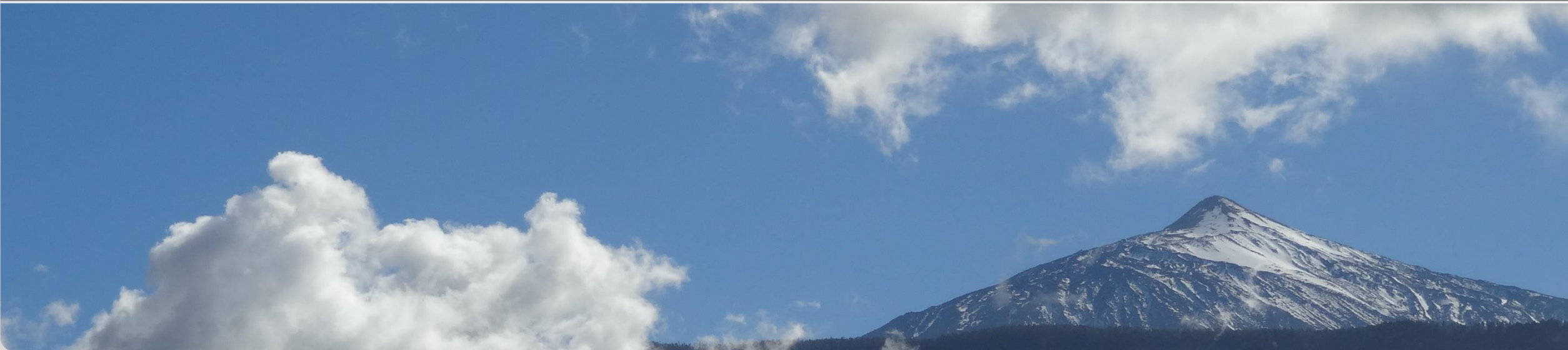


# Measurements of Gas Phase Stratospheric Sulfur

**Michael Höpfner**

**AGU Chapman Conference, Tenerife, 20-Mar-2018**

Institute of Meteorology and Climate Research- ASF



## Acknowledgements:

**Norbert Glatthor**

**Annika Günther**

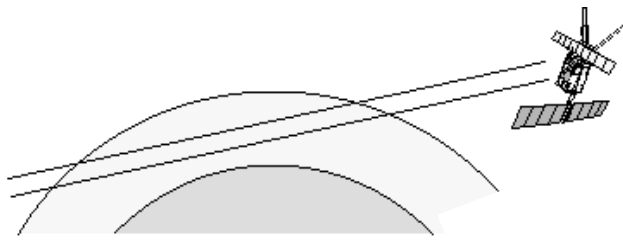
**Björn-Martin Sinnhuber**

**Gabi Stiller**

**Thomas von Clarmann**

# How to measure sulfur gases in the stratosphere?

## Remotely from satellite

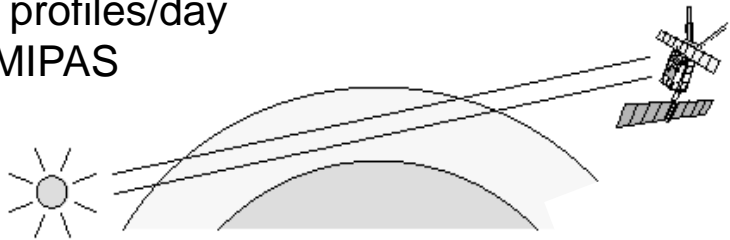


### Limb emission (thermal IR, MW)

- Global coverage, day+night
- >1000 profiles/day
- MLS, MIPAS

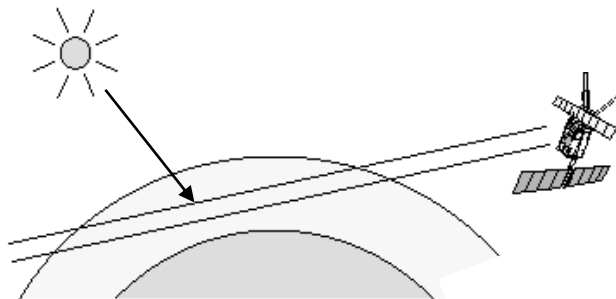
### Limb

- High vertical resolution (< 4 km)
- High sensitivity
- Low horizontal resolution (200-500 km)
- Low across-track horizontal coverage



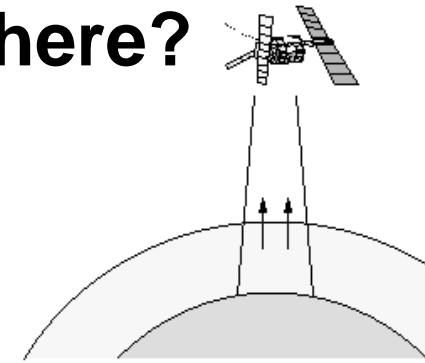
### Solar occultation (IR)

- Very high sensitivity
- ~30 profiles per day
- ACE-FTS, ATMOS



### Solar scatter (UV/VIS)

- OMPS, SCIA, OSIRIS

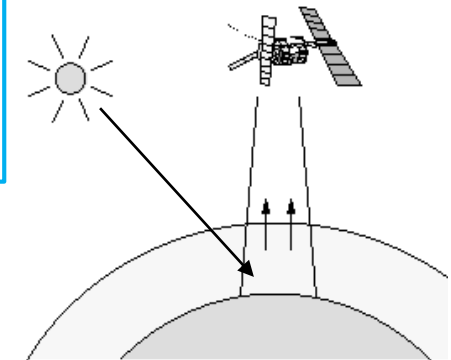


### Nadir (thermal IR)

- Global coverage, day/night+polar
- Low sensitivity in lower troposphere
- IASI, AIRS

### Nadir

- High horizontal resolution
- High coverage
- Low vertical resolution
- Low sensitivity

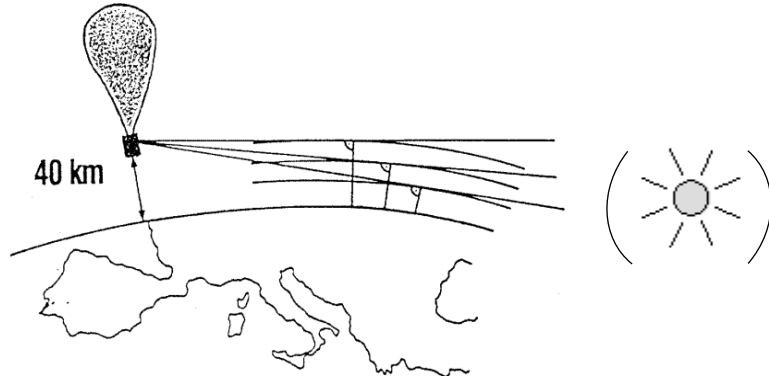


### Nadir (UV/VIS)

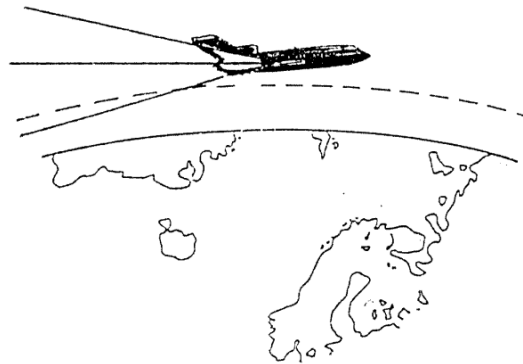
- Sensitive down to ground
- TOMS, OMI

# How to measure sulfur gases in the stratosphere?

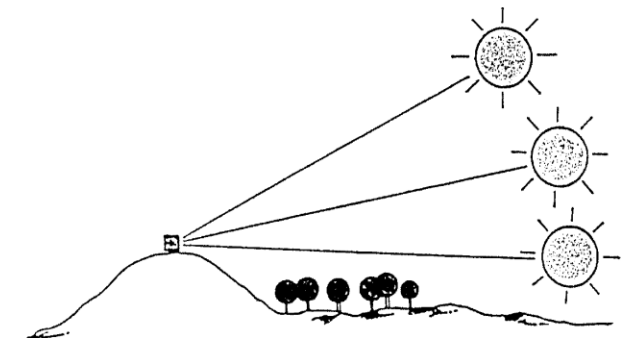
*From ground, ship, aircraft, balloon*



- Limb emission or occultation (MarkIV, MIPAS-B)
- In-situ



- Limb emission
- In-situ



- Solar absorption (IR) (NDACC-FTIRs)

# MIPAS ,sulfur datasets‘

## ■ **SO<sub>2</sub> volume mixing ratio profiles (2 datasets)**

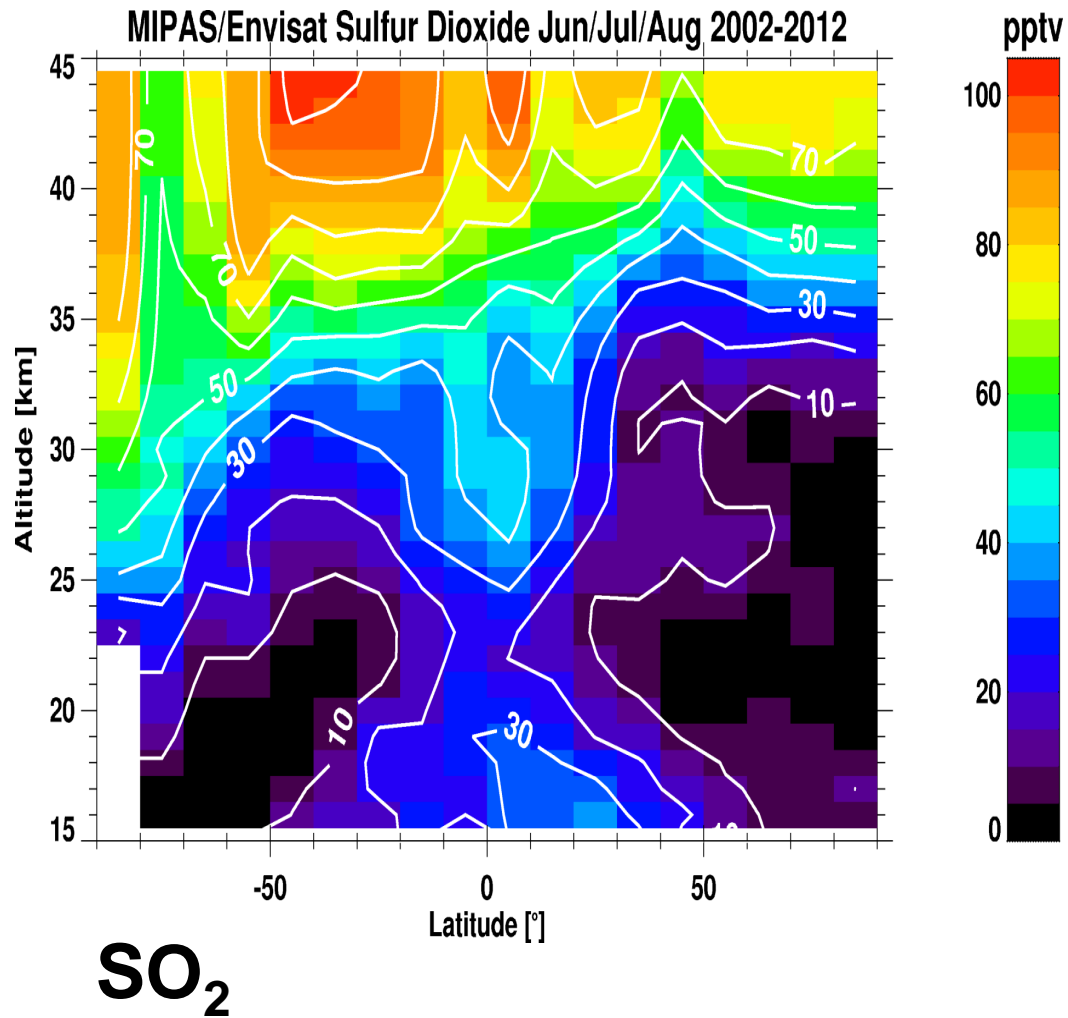
- Retrieval from mean spectra: 15-45 km, monthly+zonal averages, 18 profiles/month (Höpfner et al., ACP, 2013)
- Retrieval from single limb-scans: 8-20 km, high temporal and horizontal resolution, up to 1400 profiles/day (Höpfner et al., ACP, 2015)

## ■ **OCS volume mixing ratio profiles (1 dataset)**

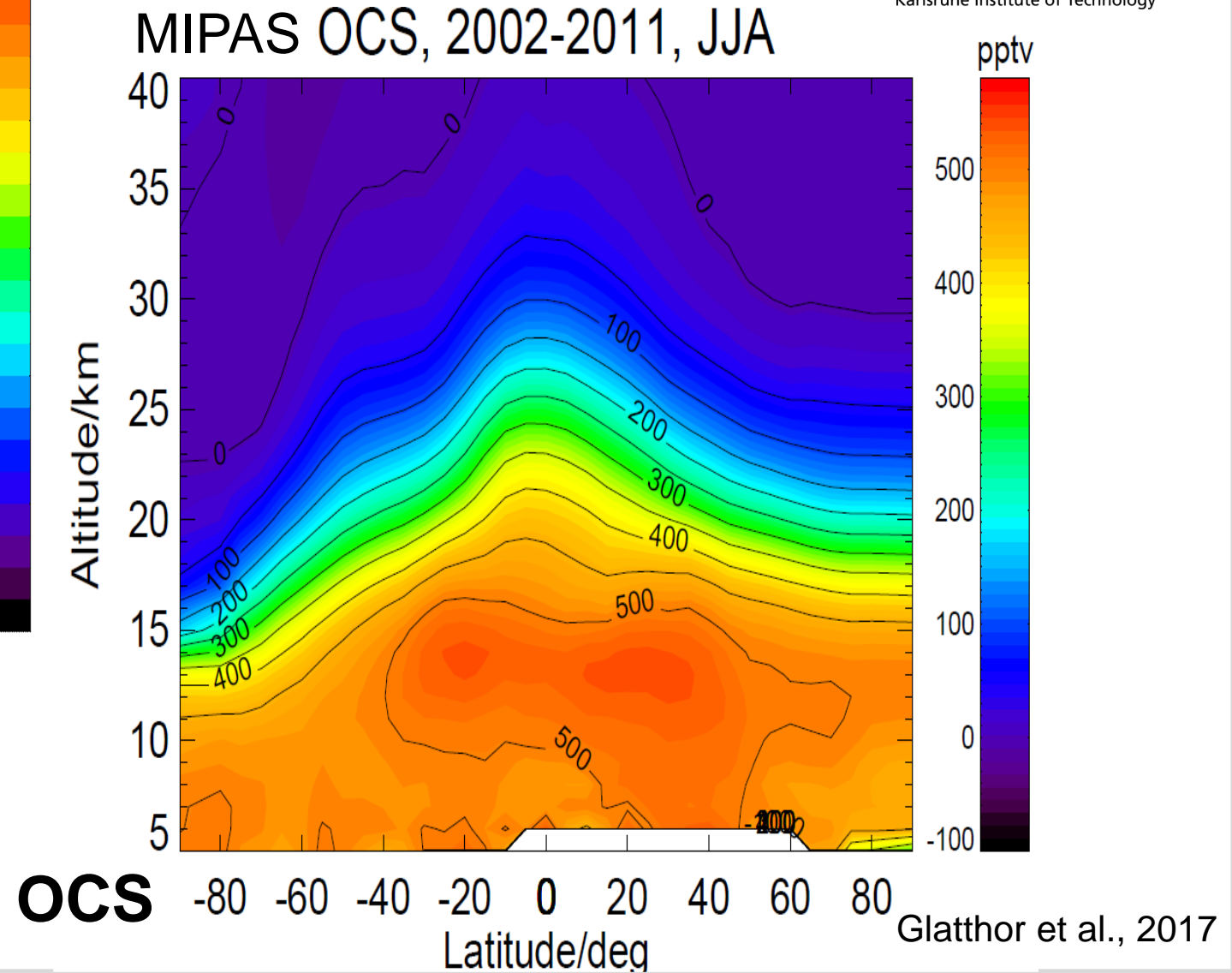
- Retrieval from single limb-scans: 8-35 km, high temporal and horizontal resolution, up to 1400 profiles/day (upper troposphere: Glatthor et al., GRL, 2015, stratosphere: Glatthor et al., ACP, 2016)

## ■ **H<sub>2</sub>SO<sub>4</sub> aerosol volume density profiles (1 dataset)**

- Retrieval from single limb-scans: 8-~33 km, high temporal and horizontal resolution, up to 1400 profiles/day (Günther et al., ACP, 2018)

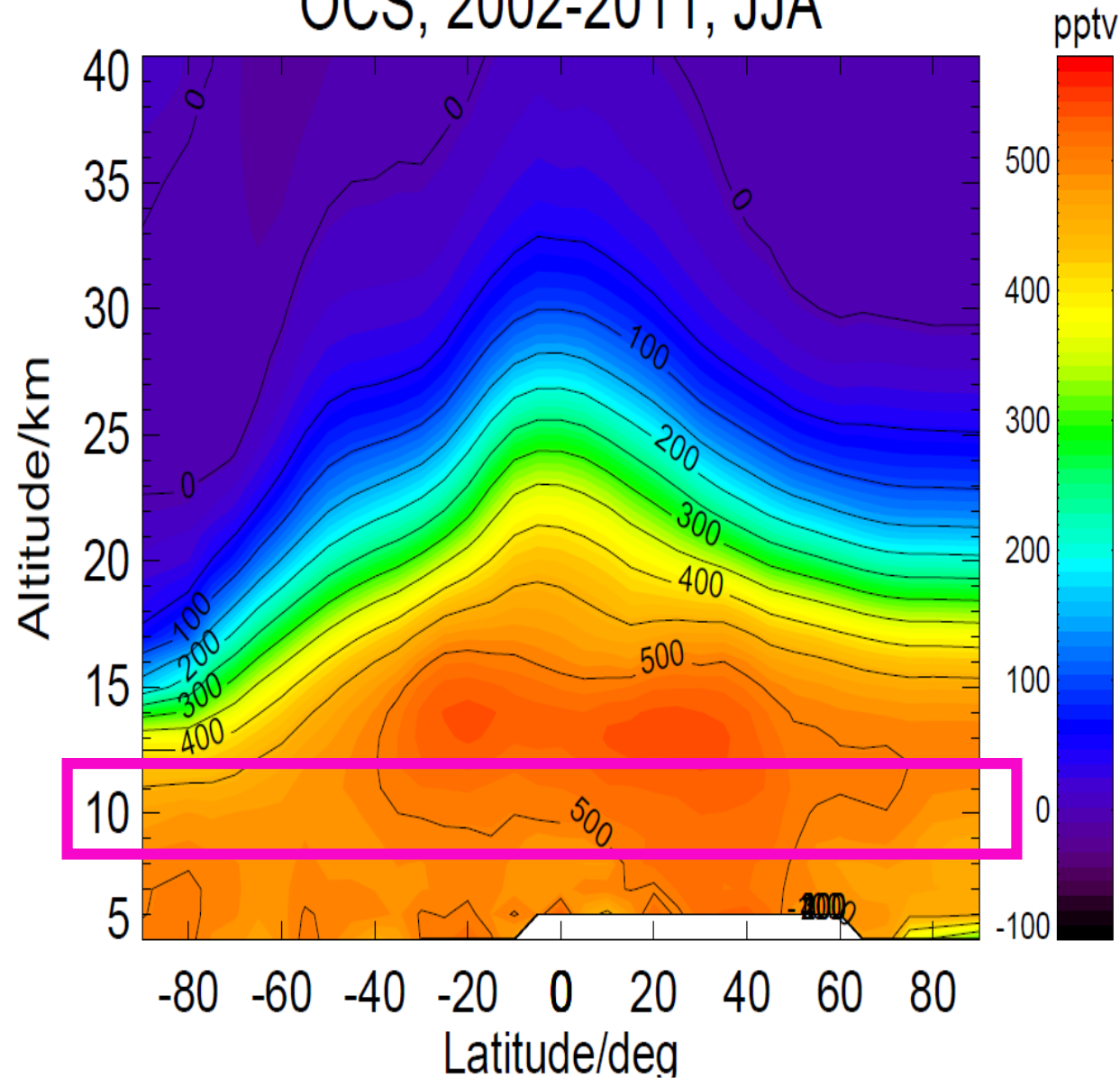


Höpfner et al., 2013





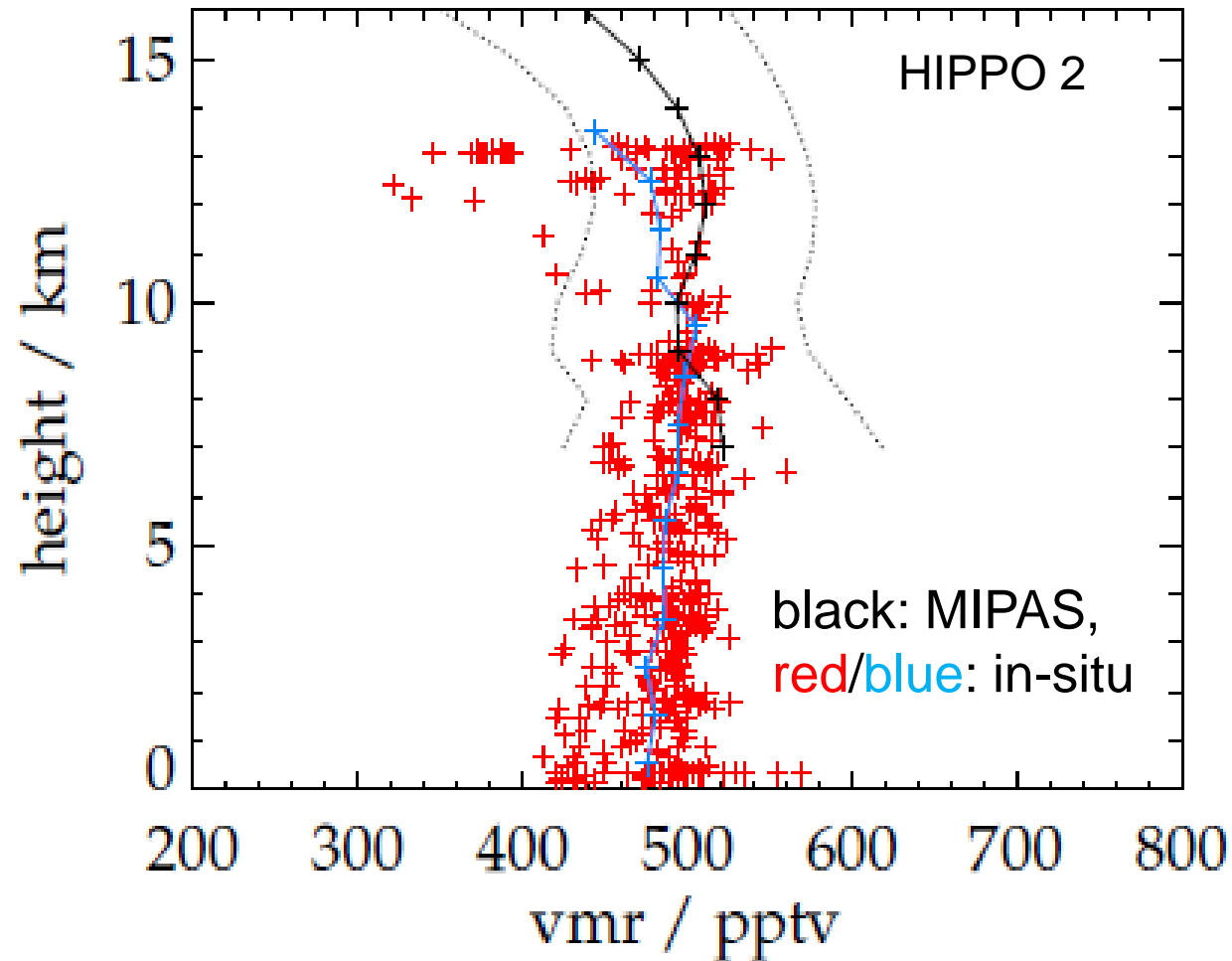
# OCS, 2002-2011, JJA



Glatthor et al., 2017

# Comparison MIPAS OCS vs. airborne in-situ

## MIPAS vs. in-situ aircraft

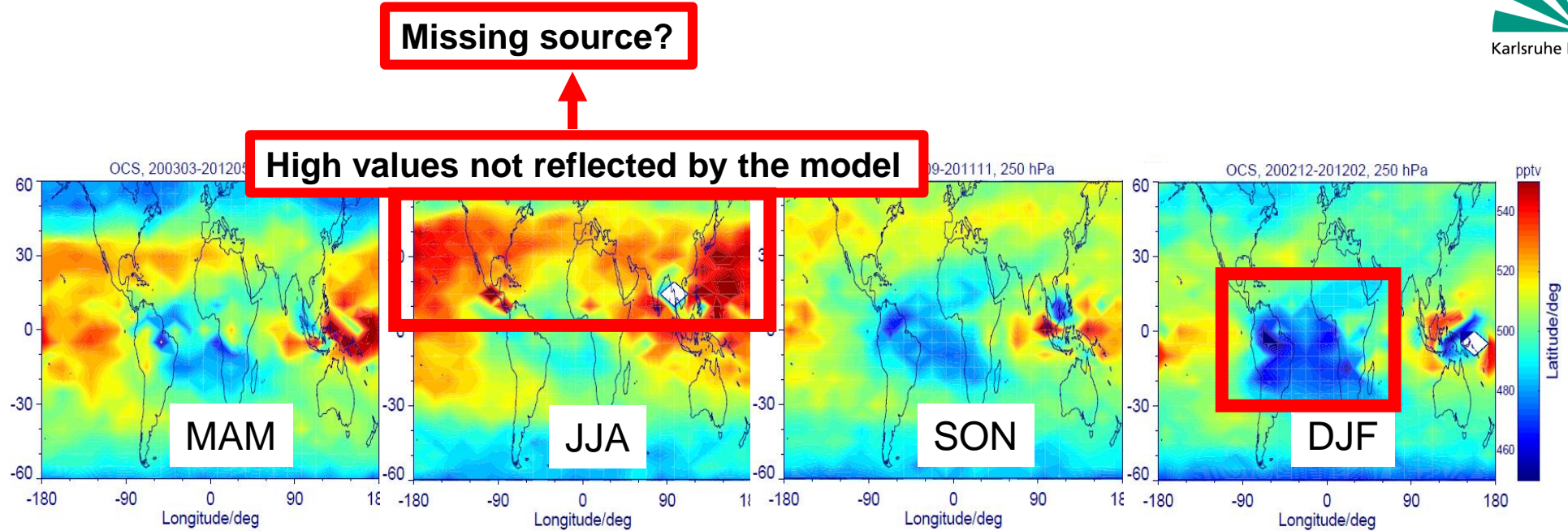




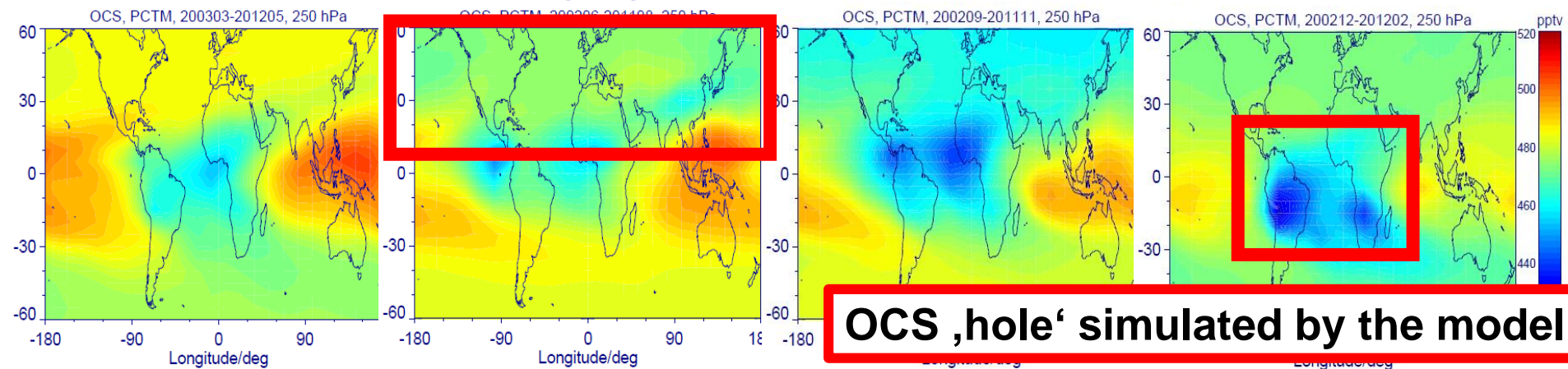
# OCS in the upper troposphere

**MIPAS**

**250 hPa**

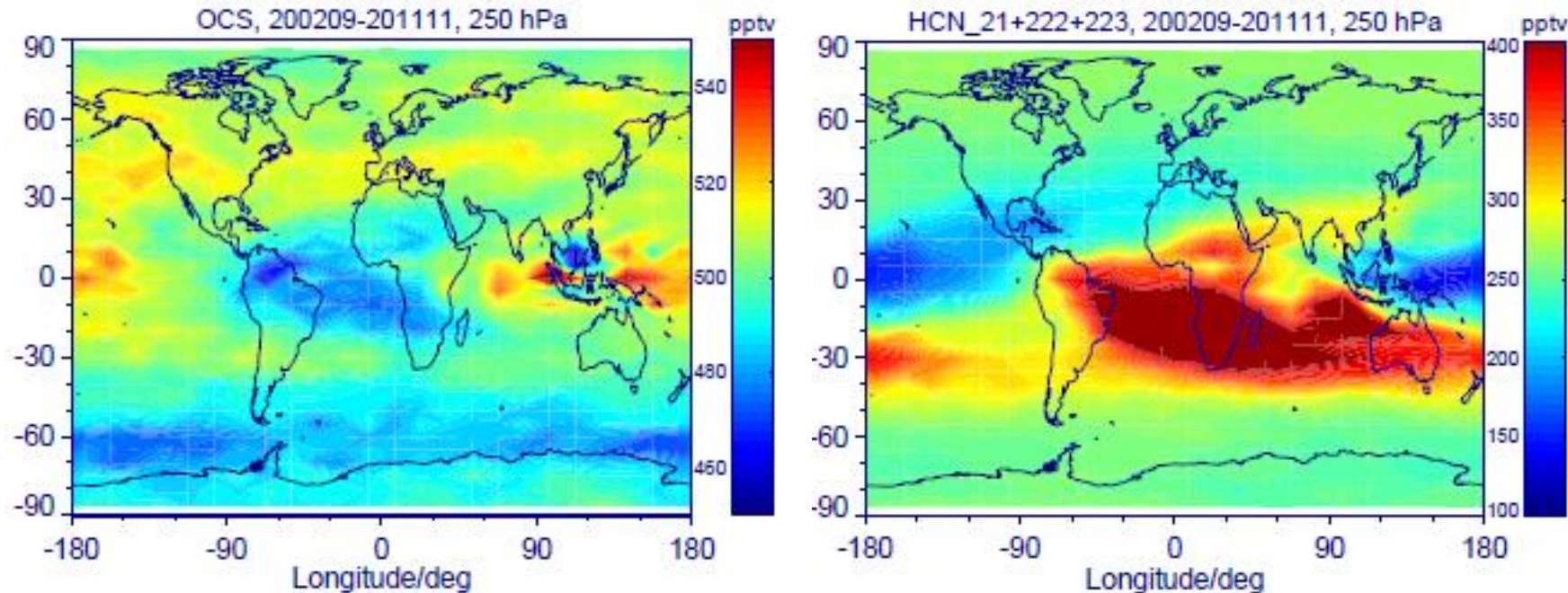


**Model**



# Biomass burning as a source of OCS?

## Biomass burning tracer HCN

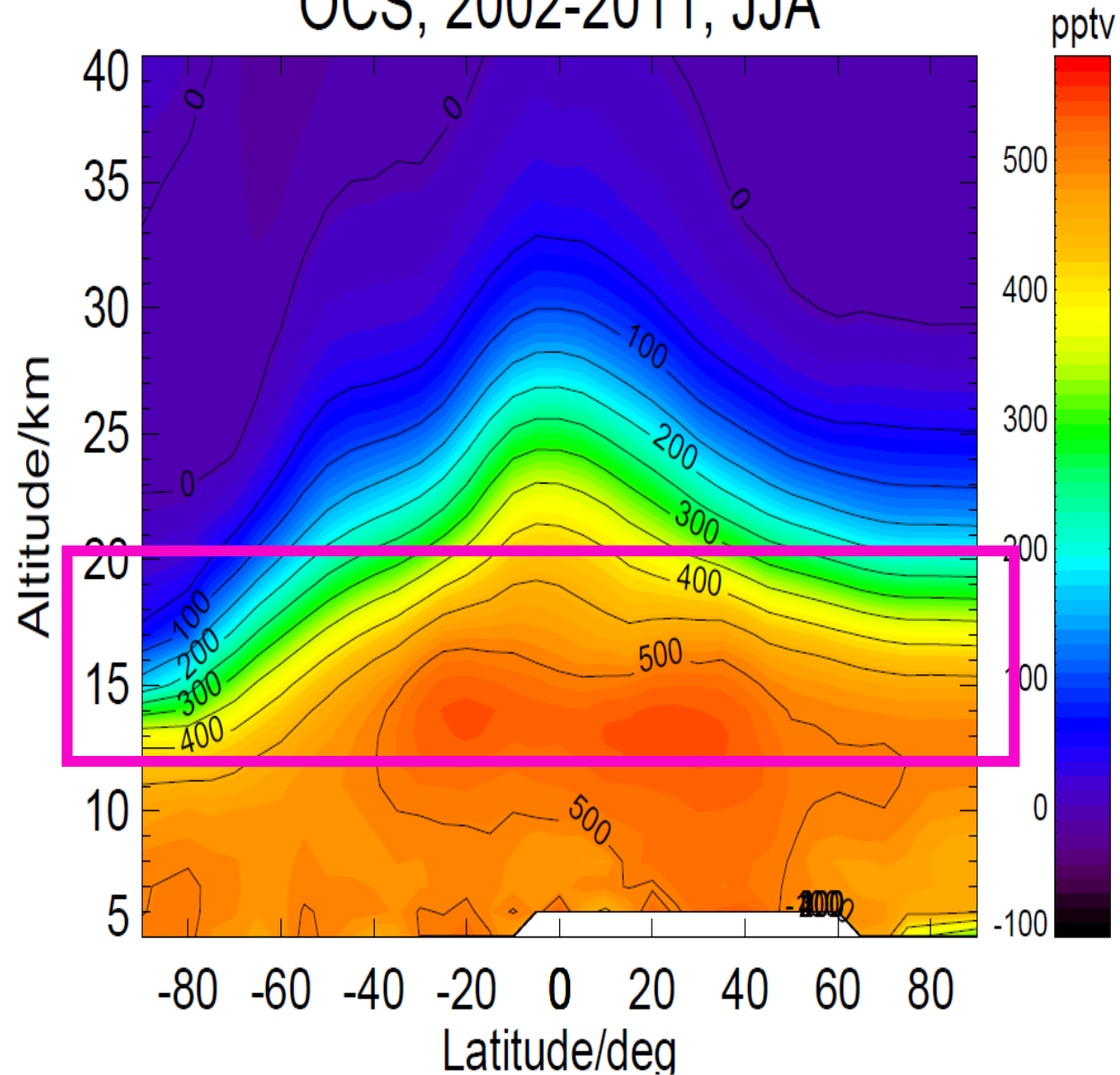


→ no obvious enhancement of OCS due to biomass burning

Glatthor et al., 2017

M. Höpfner, IMK-ASF

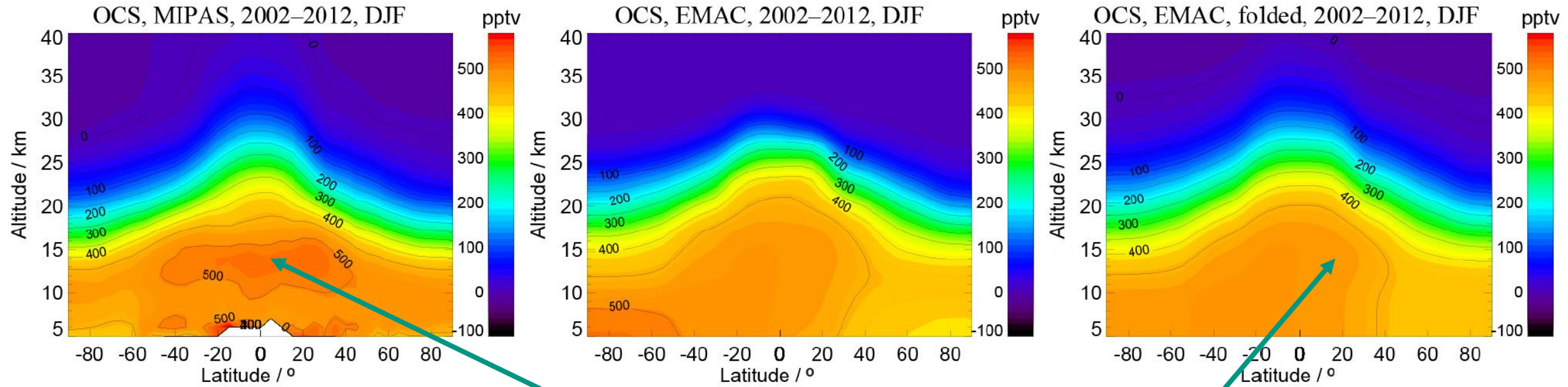
# OCS, 2002-2011, JJA



Glatthor et al., 2017



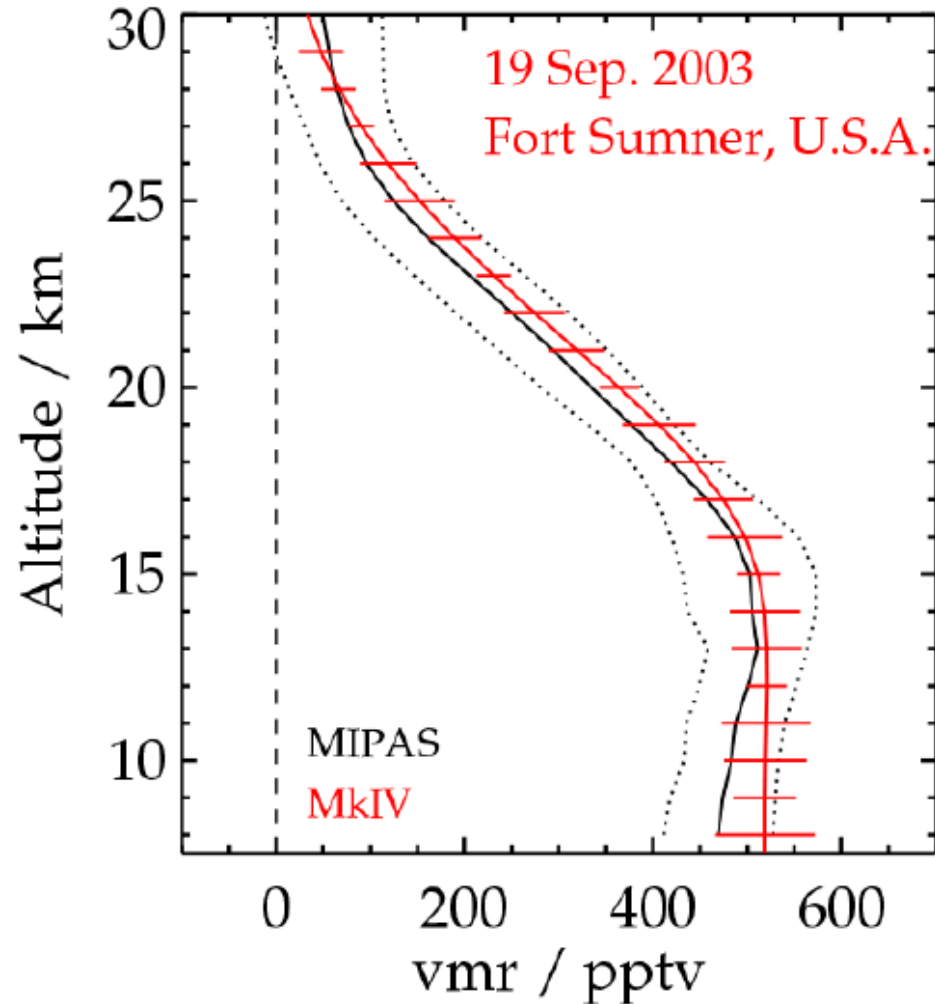
# Upper tropospheric enhancement?



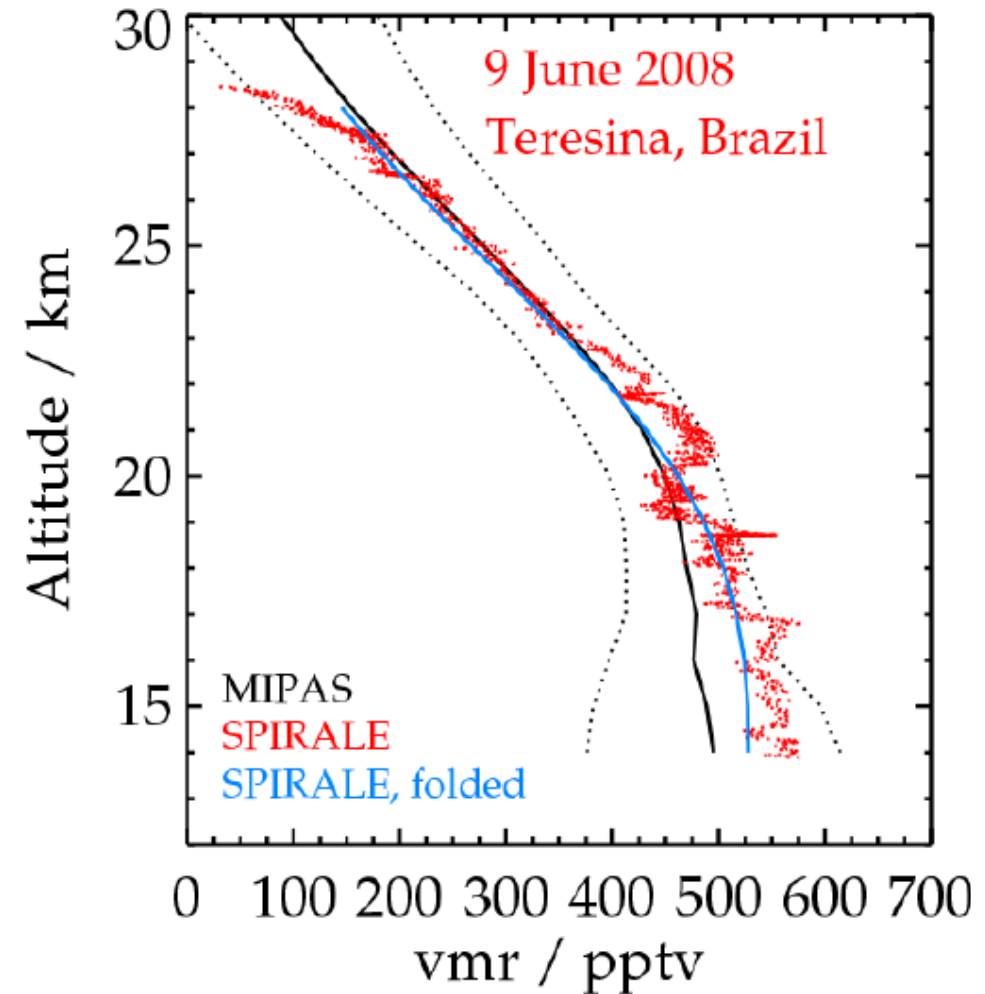
- The UT enhancement is visible in MIPAS but not (or much less) in model calculations

# Comparison MIPAS OCS vs. balloon

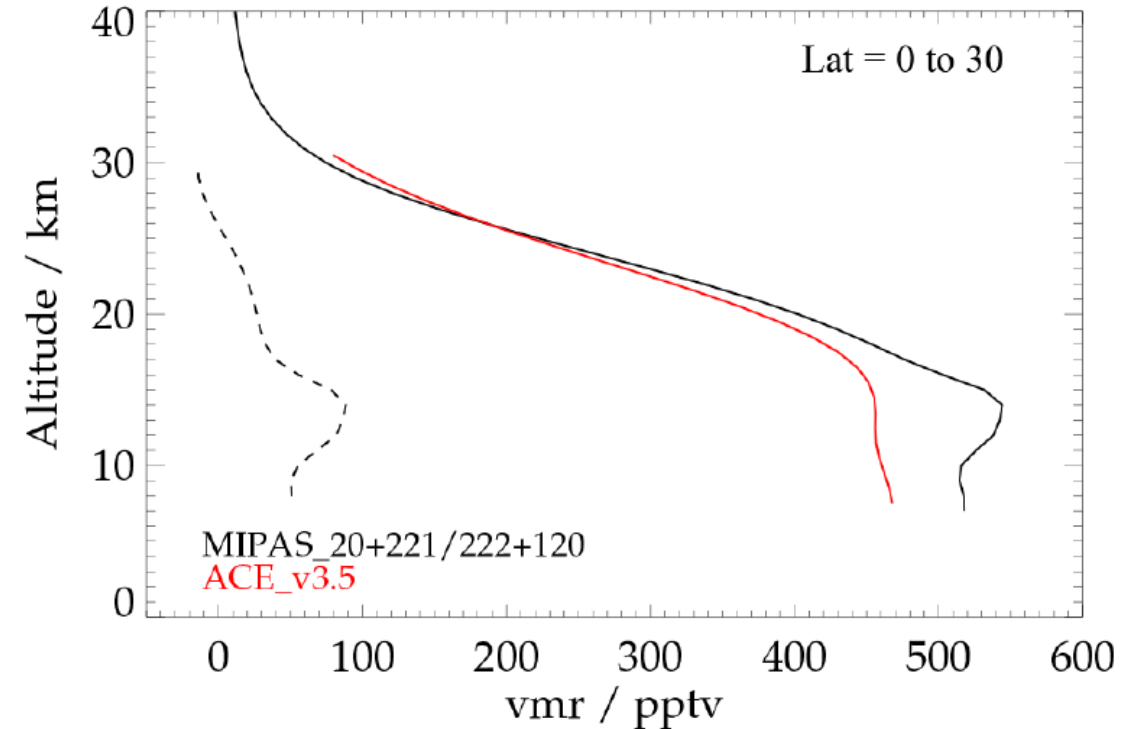
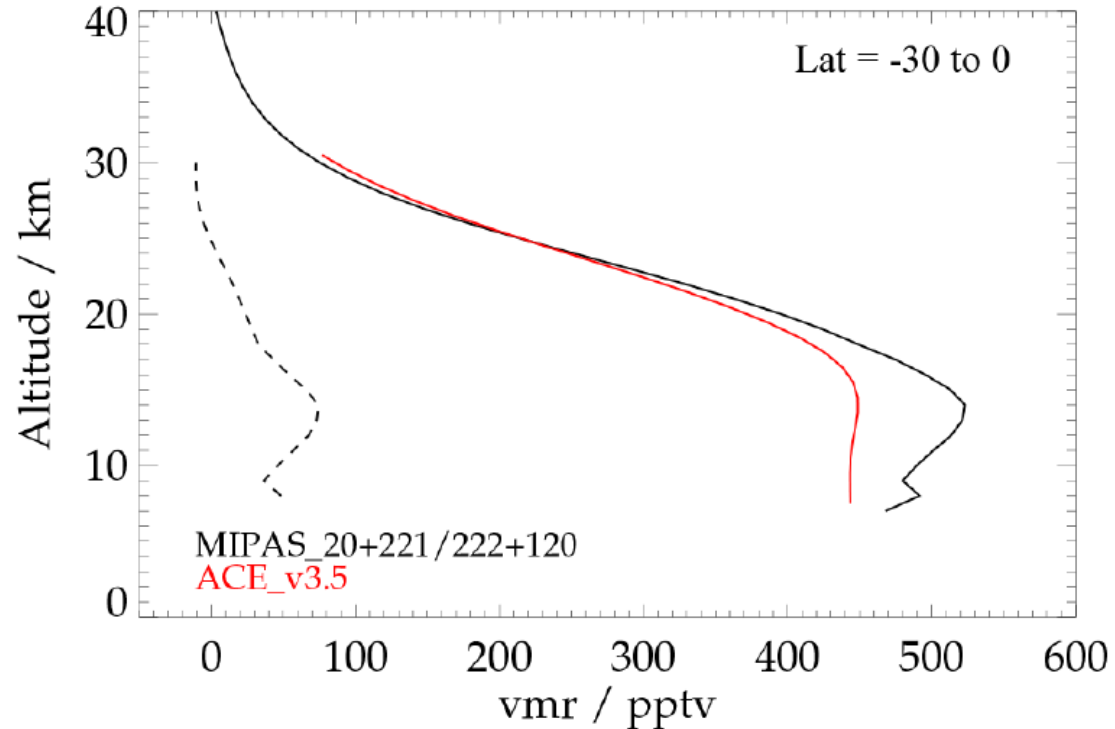
## MIPAS vs. MkIV balloon



## MIPAS vs. SPIRALE



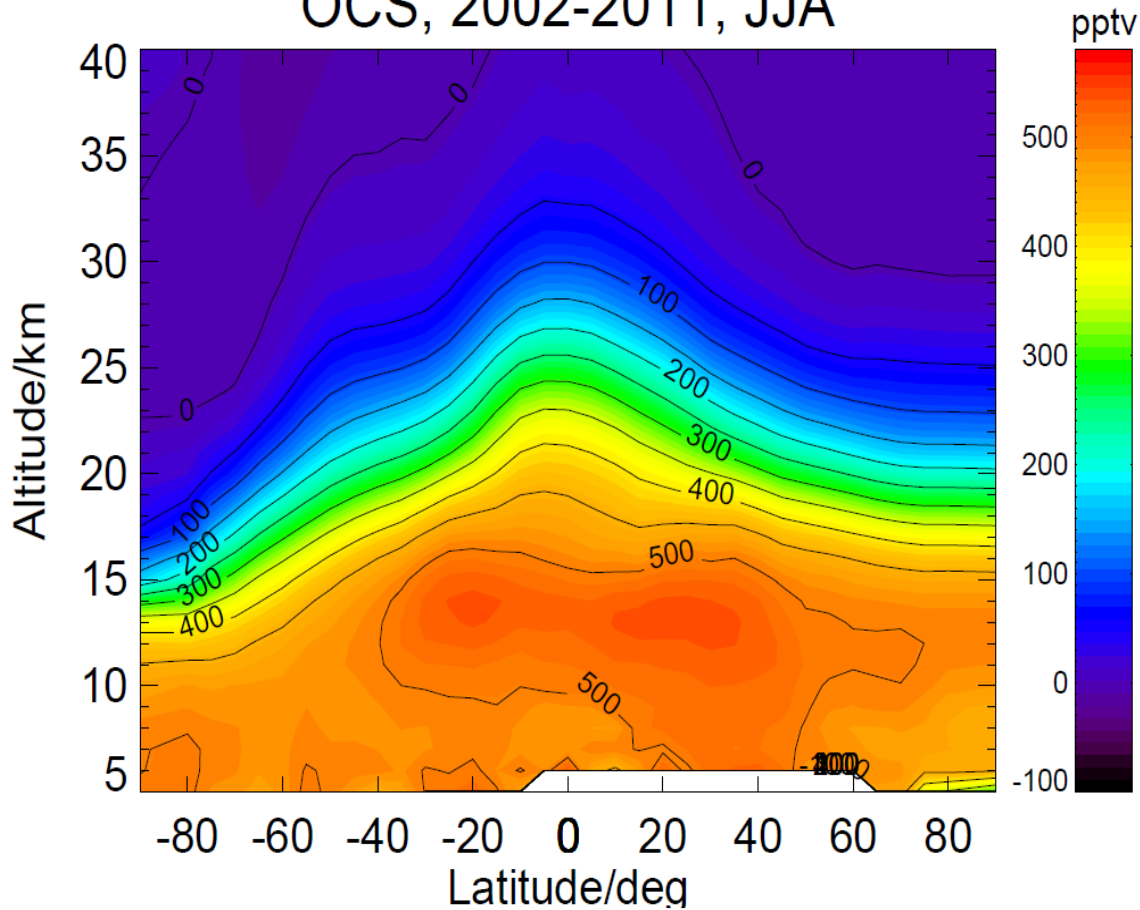
# Co-incident comparison: MIPAS vs. ACE-FTS



- Up to 20 km MIPAS values are ~50-70 pptv higher than ACE-FTS
- UT-"nose" not or only slightly visible in ACE-FTS

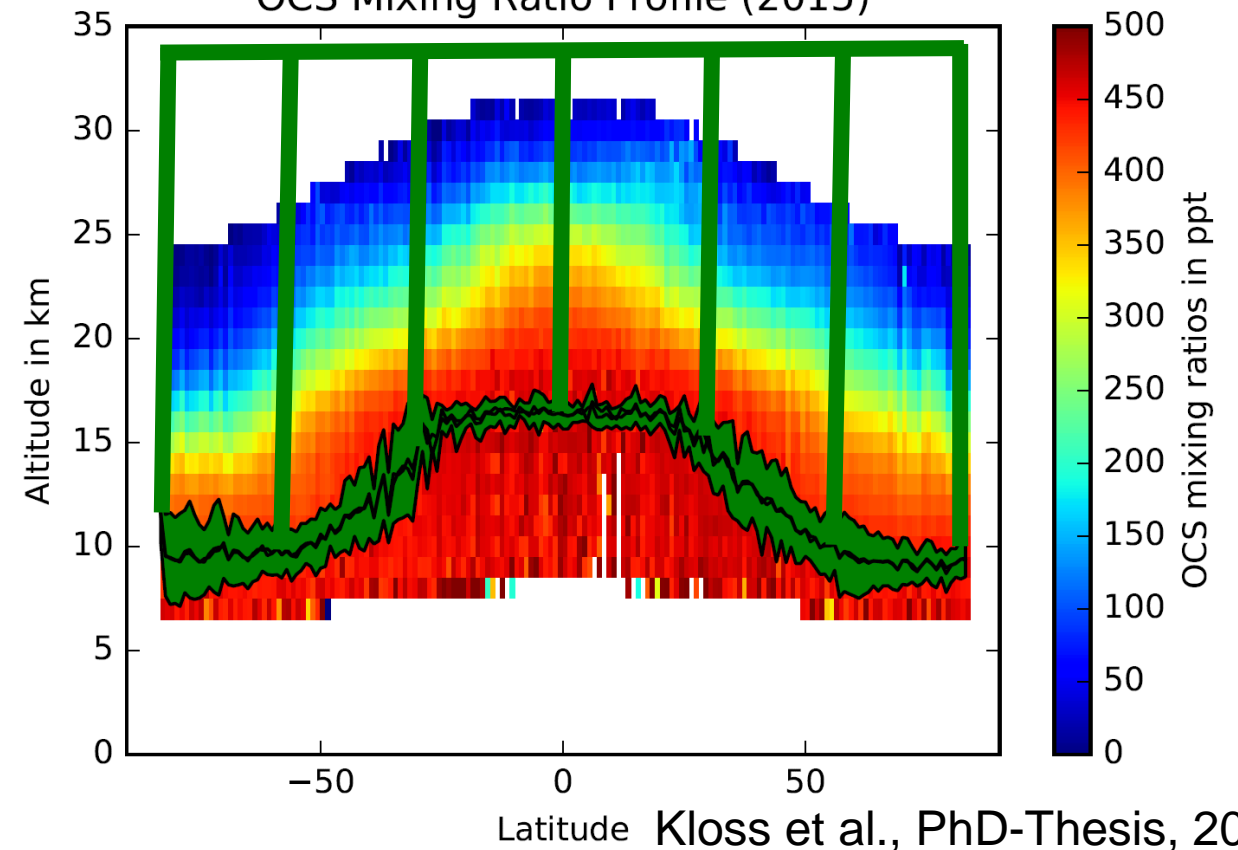
# Global distributions: MIPAS vs. ACE-FTS

OCS, 2002-2011, JJA



Glatthor et al., 2017

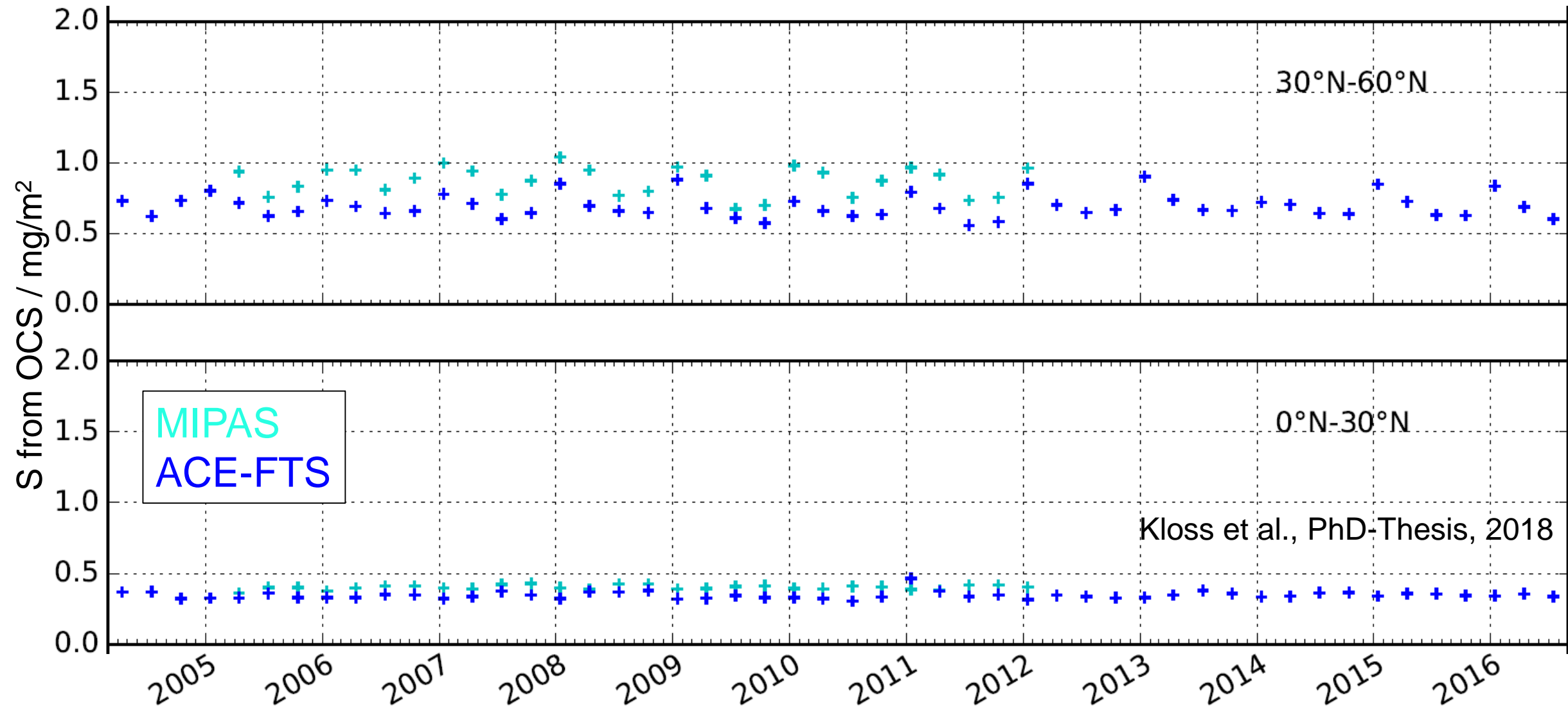
OCS Mixing Ratio Profile (2015)



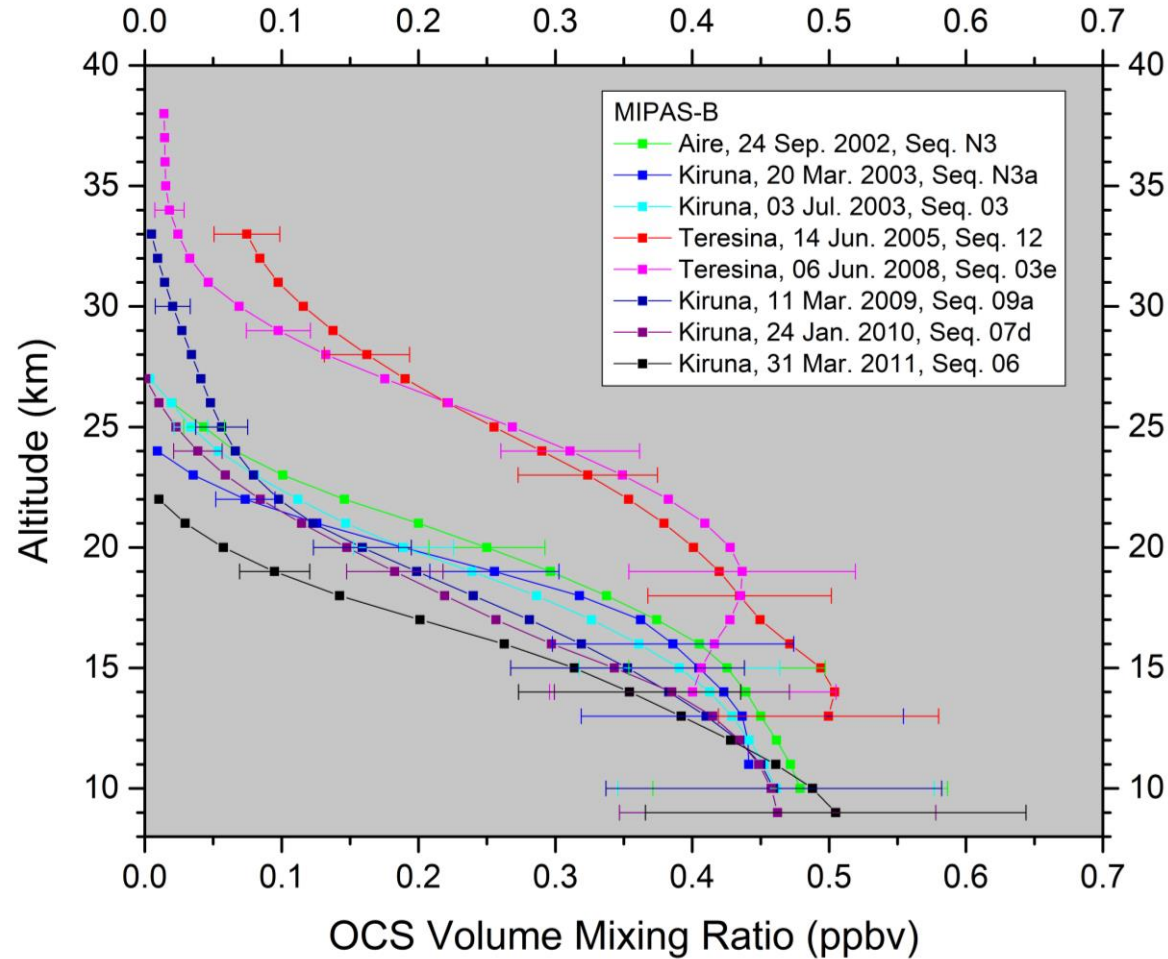
Kloss et al., PhD-Thesis, 2018



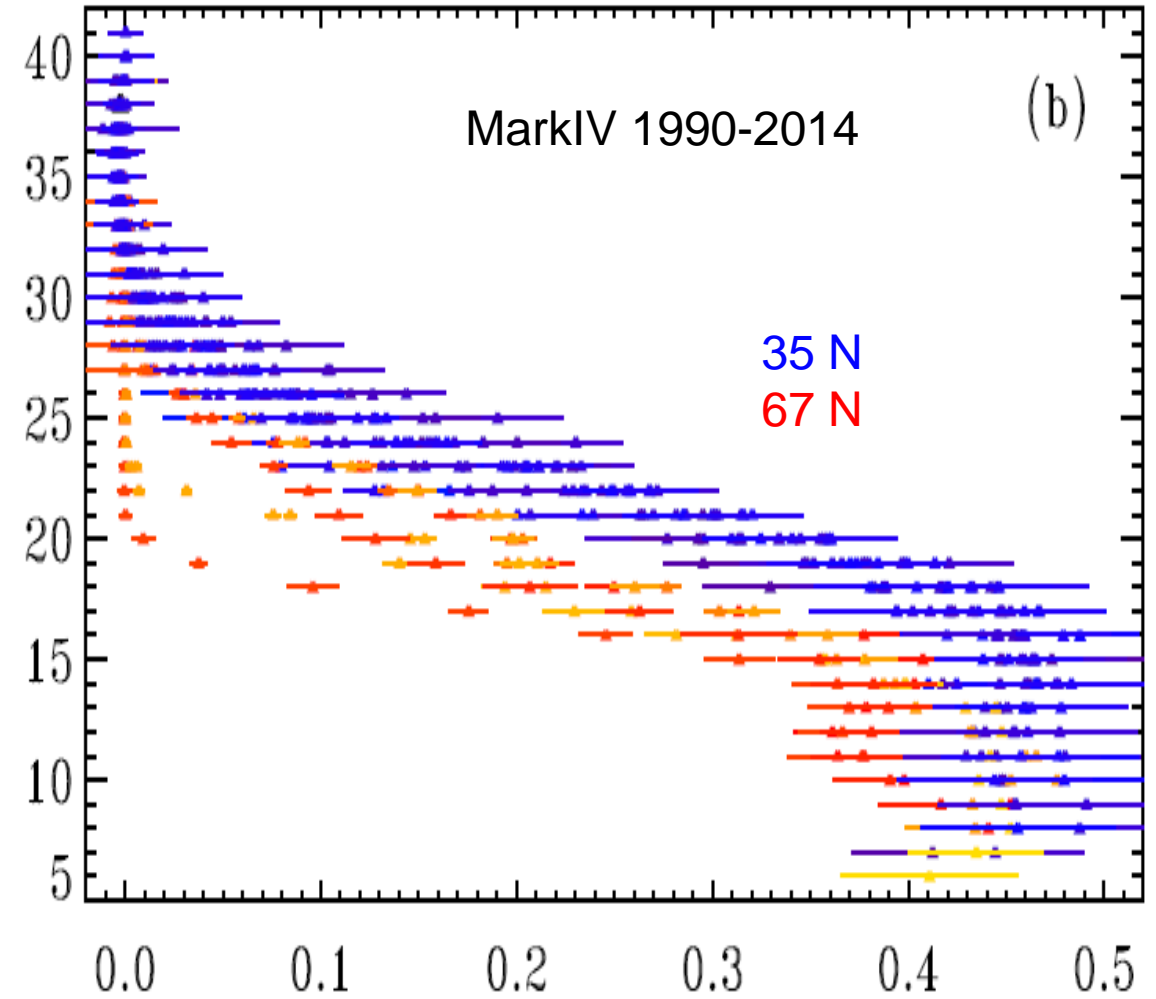
# Stratospheric sulfur column in OCS



# New datasets of OCS

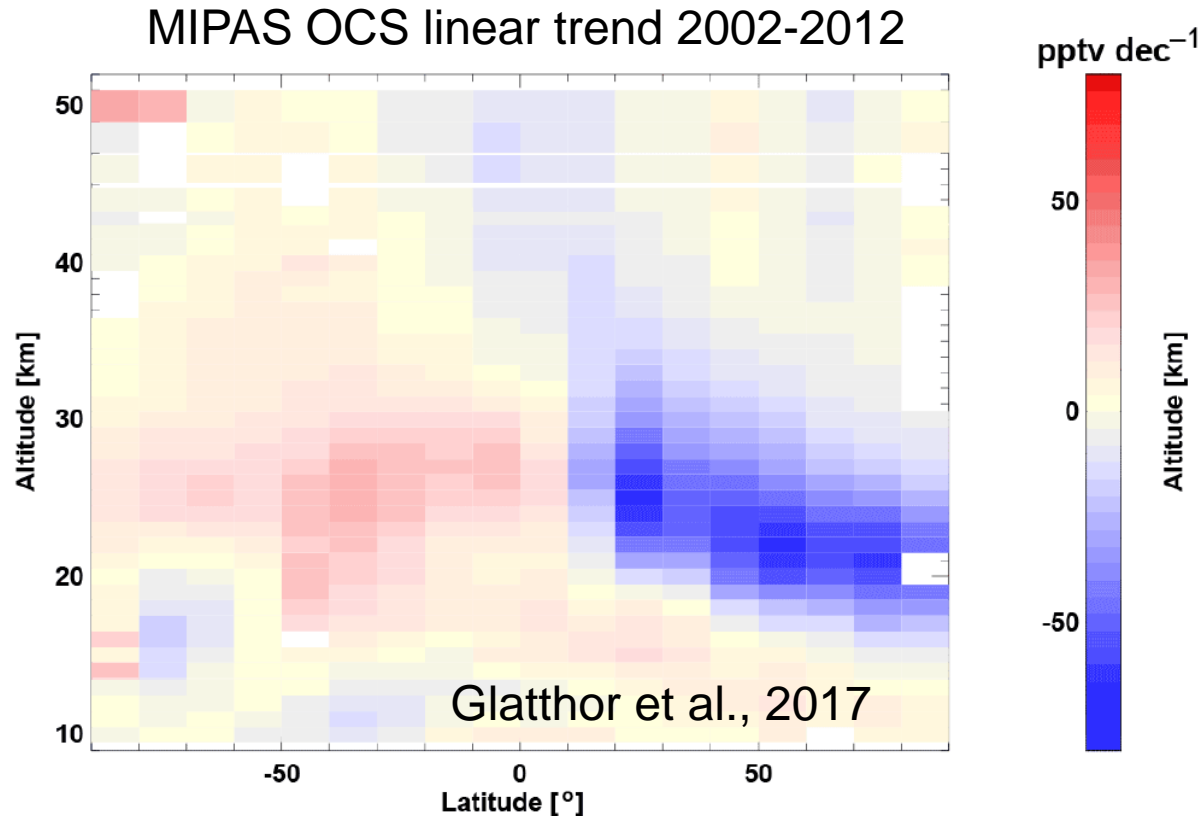


Courtesy: G. Wetzel



Toon et al., 2018

# Trends in OCS



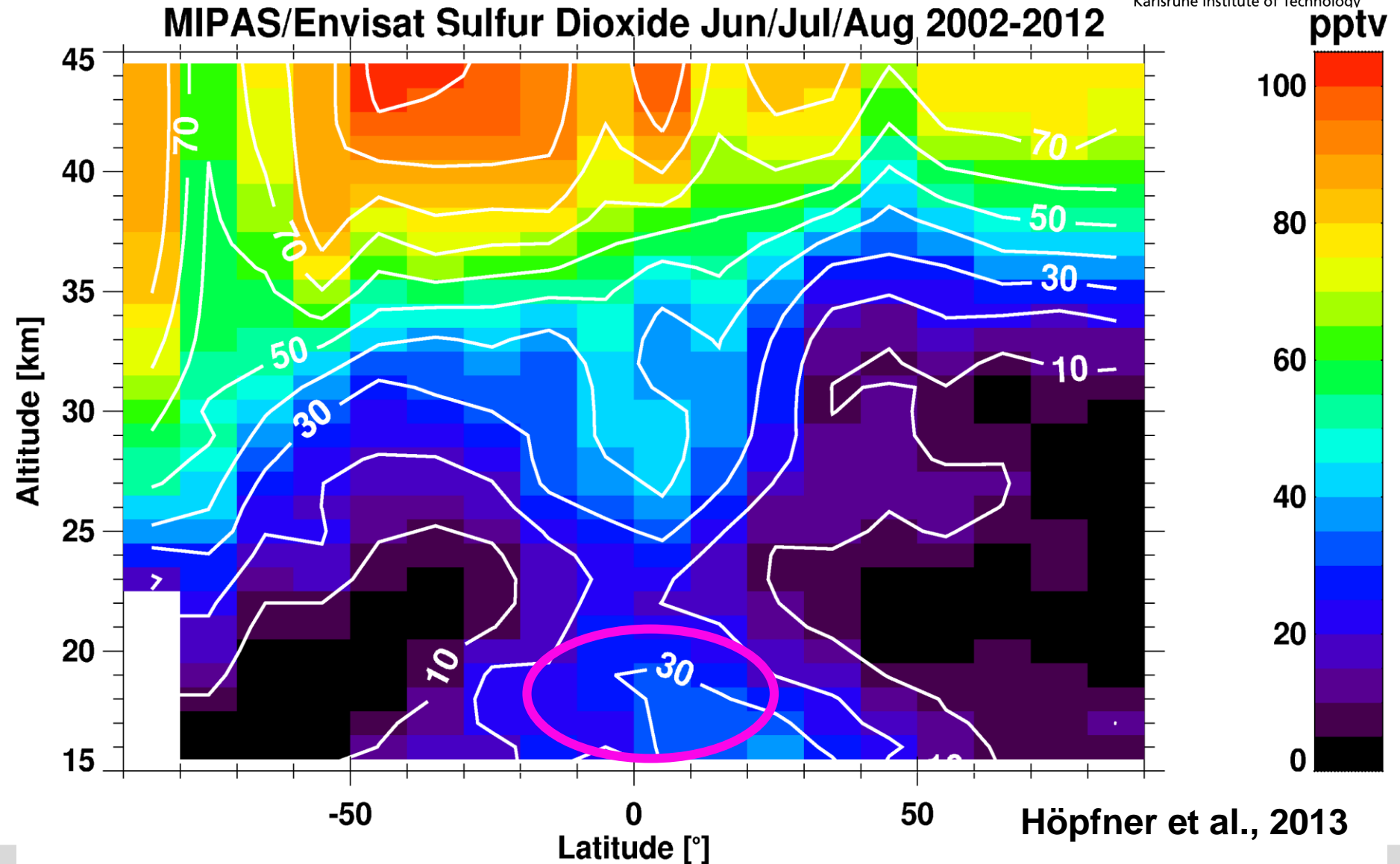
Toon et al., 2018:

- “Balloon results yield no significant stratospheric trend.”
- “Tropospheric OCS, ..., shows a 5% decrease during 1990–2002, followed by a 5% increase from 2003 to 2012. There was no discernible change since 2012.”

- Follows the trend in Age-of-air (from MIPAS SF<sub>6</sub>)

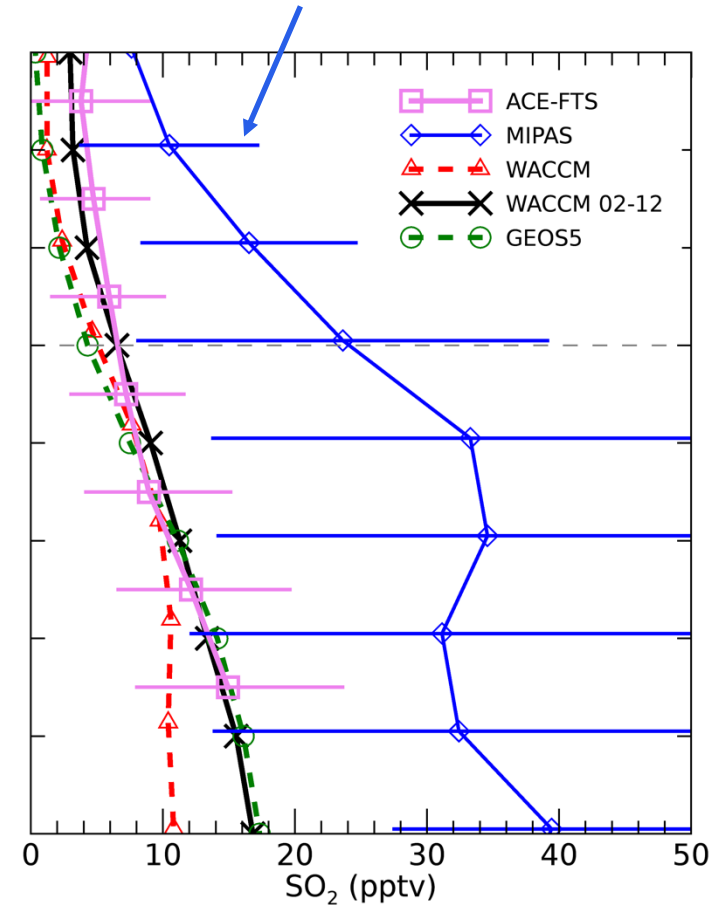
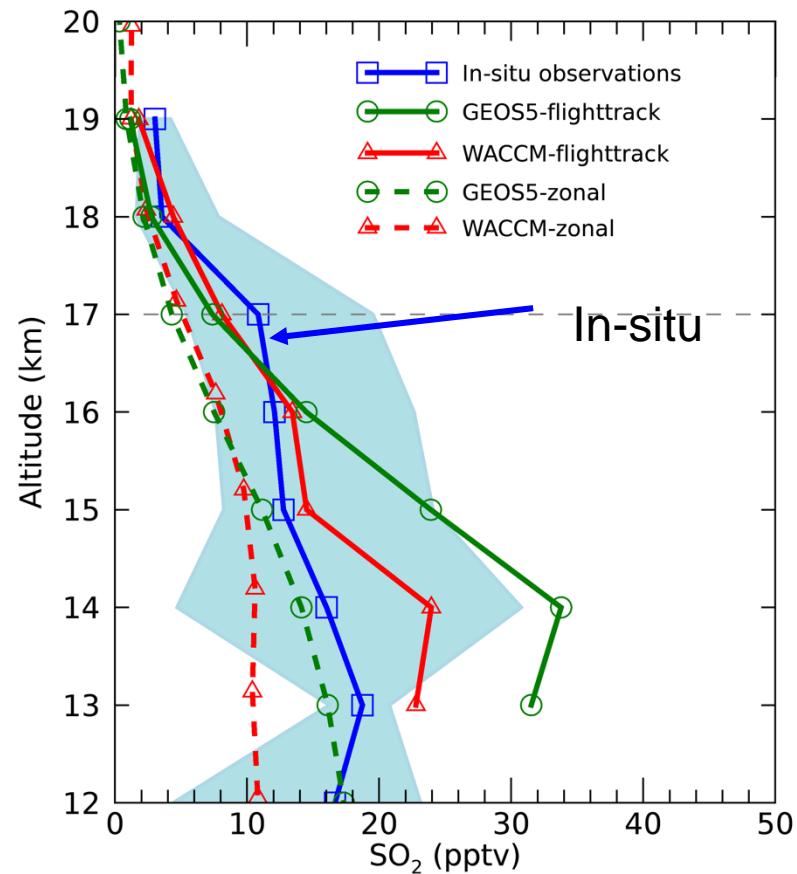
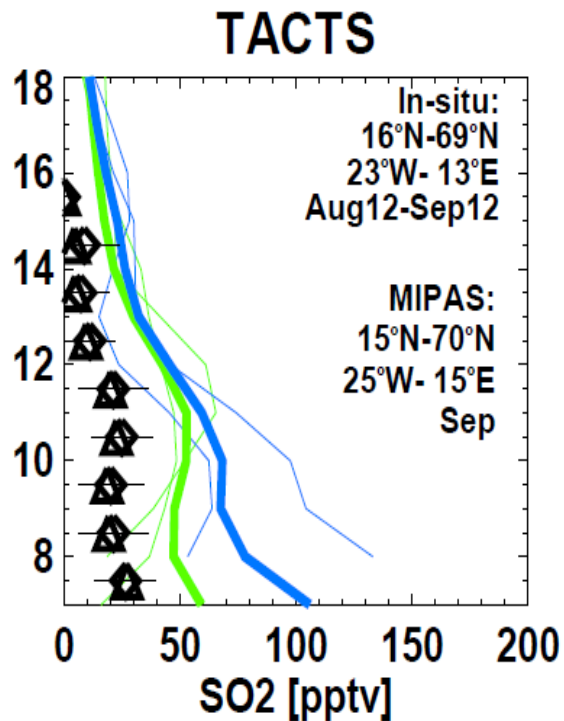
# The tropical upper troposphere SO<sub>2</sub> “background”

- Determines the input of SO<sub>2</sub> into the stratosphere



# How much SO<sub>2</sub> is typically at the tropical tropopause?

MIPAS  $\pm 1\sigma$  Estimated uncertainty of single scan dataset



Höpfner et al., 2015

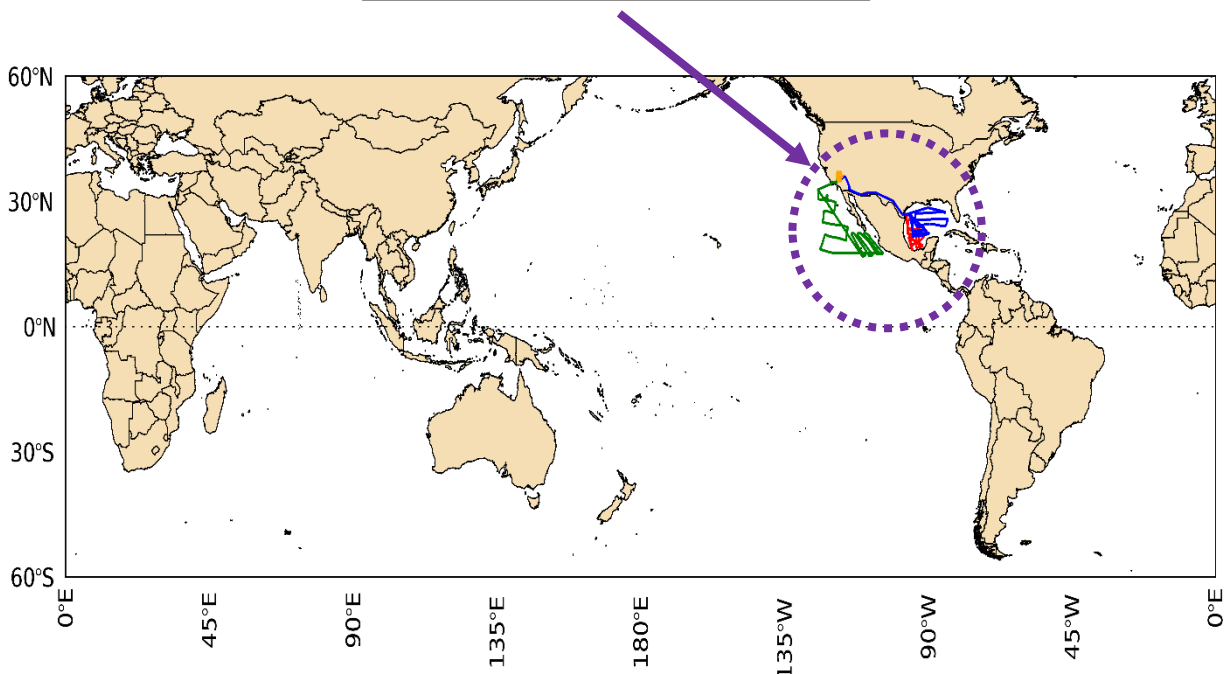
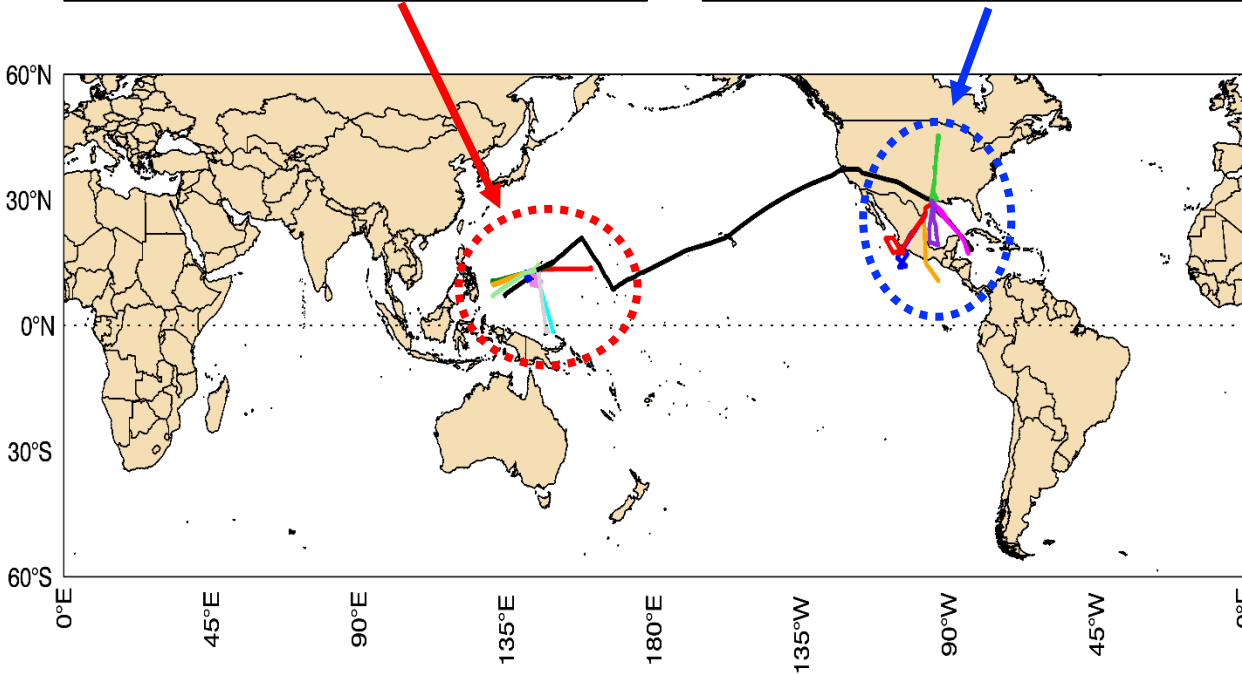
Updated from Rollins et al., 2017



POSIDON, October 2016

VIRGAS, October 2015

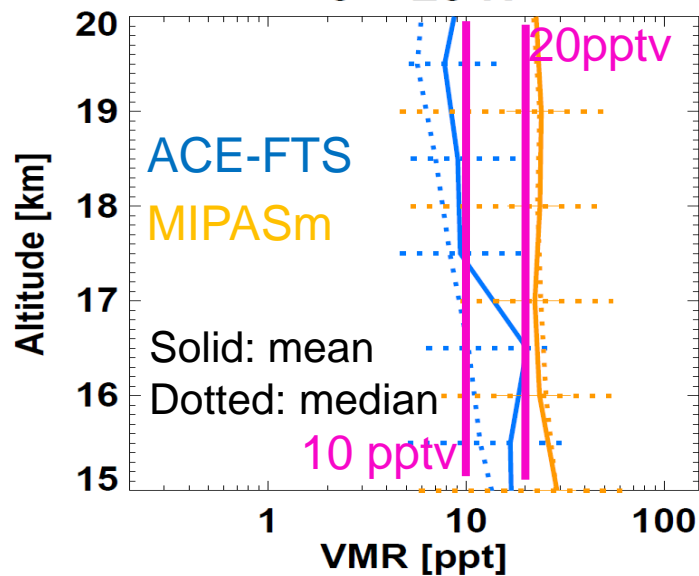
EPOCH, August 2017



Courtesy: **A. Rollins**, T. Thornberry, R.-S. Gao



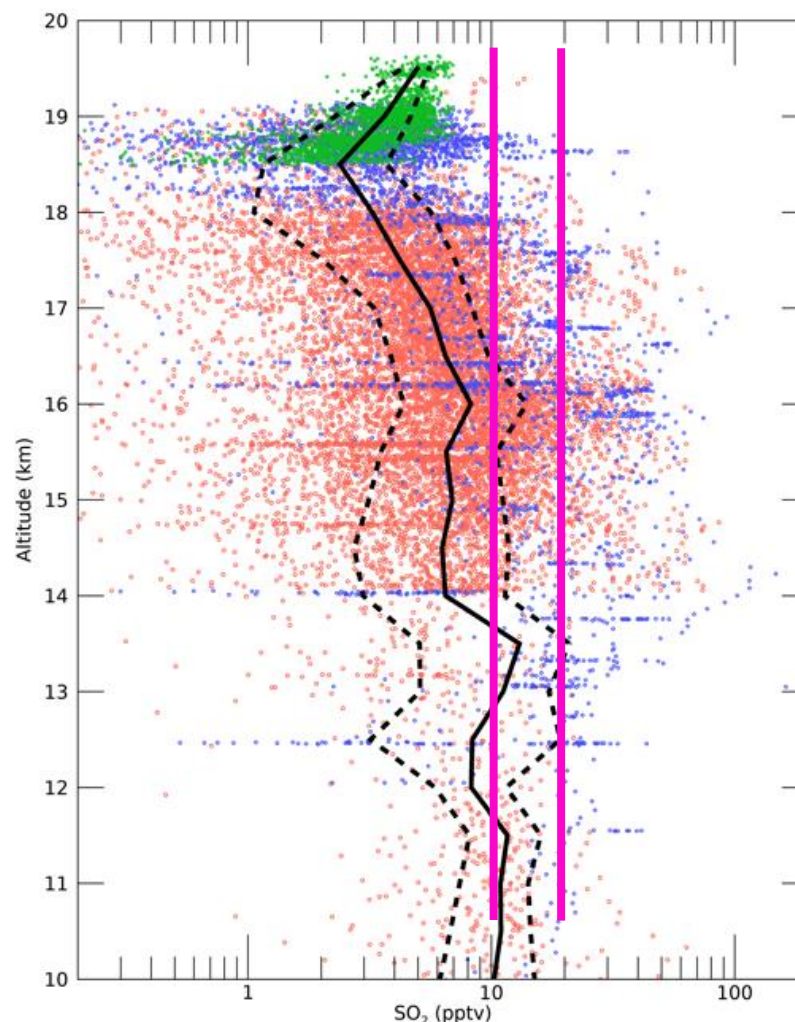
0° - 20°N



Both MIPAS datasets seem to have a positive bias (10-15 ppt) in the tropical UT (tbc)

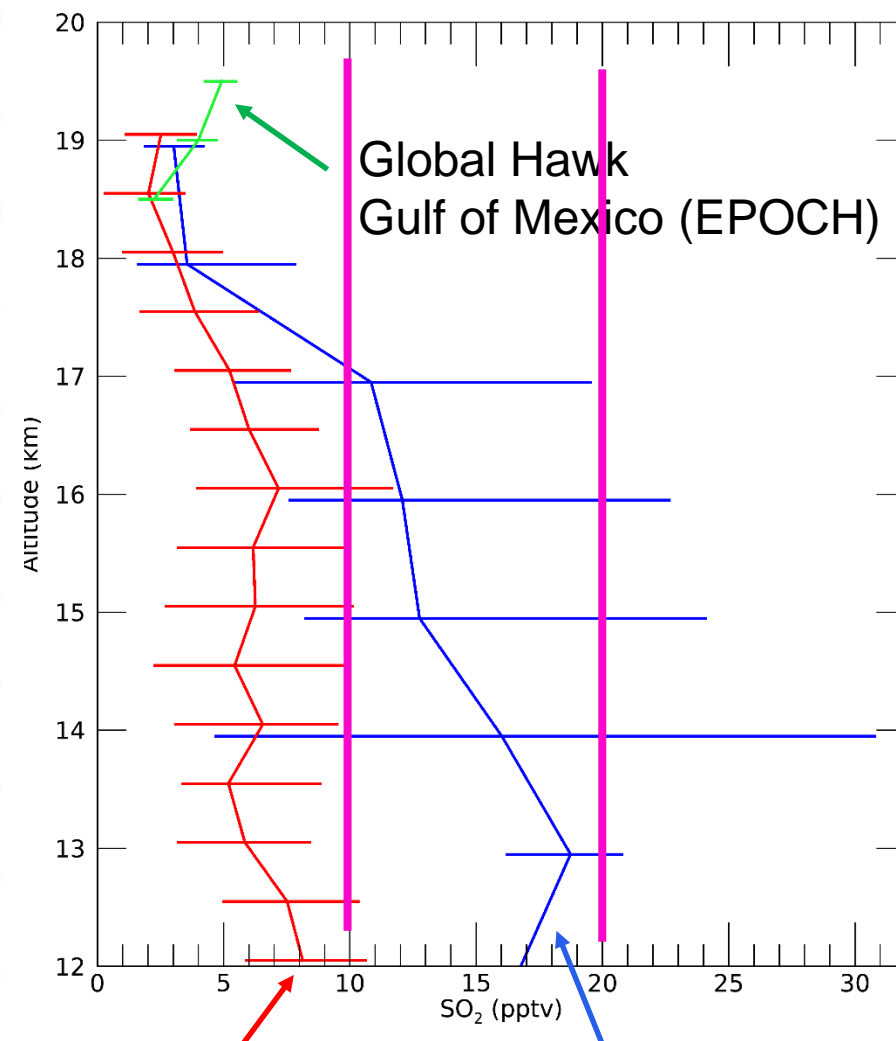
WB-57: 2 S - 20 N

Global Hawk: 20 - 25 N



Lines show median and interquartile range

Statistics by mission, latitude < 25 N



WB-57  
Guam (POSIDON)

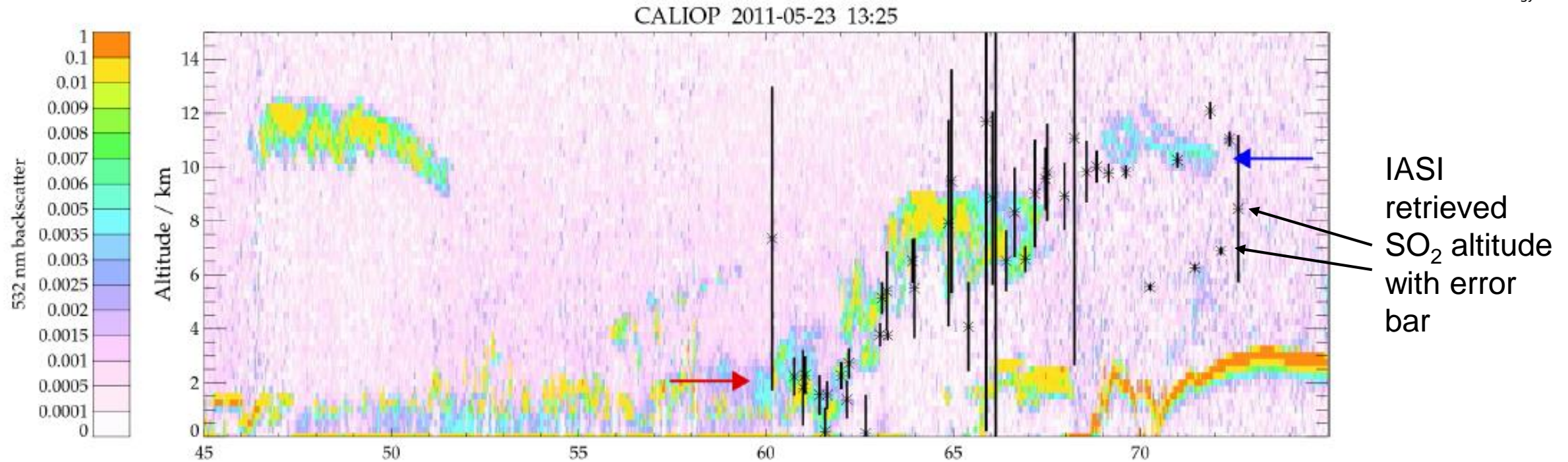
WB-57  
Houston (VIRGAS)

Courtesy: **A. Rollins**, T. Thornberry, R.-S. Gao

M. Höpfner, IMK-ASF



# Altitude resolved SO<sub>2</sub> observations by nadir sounders



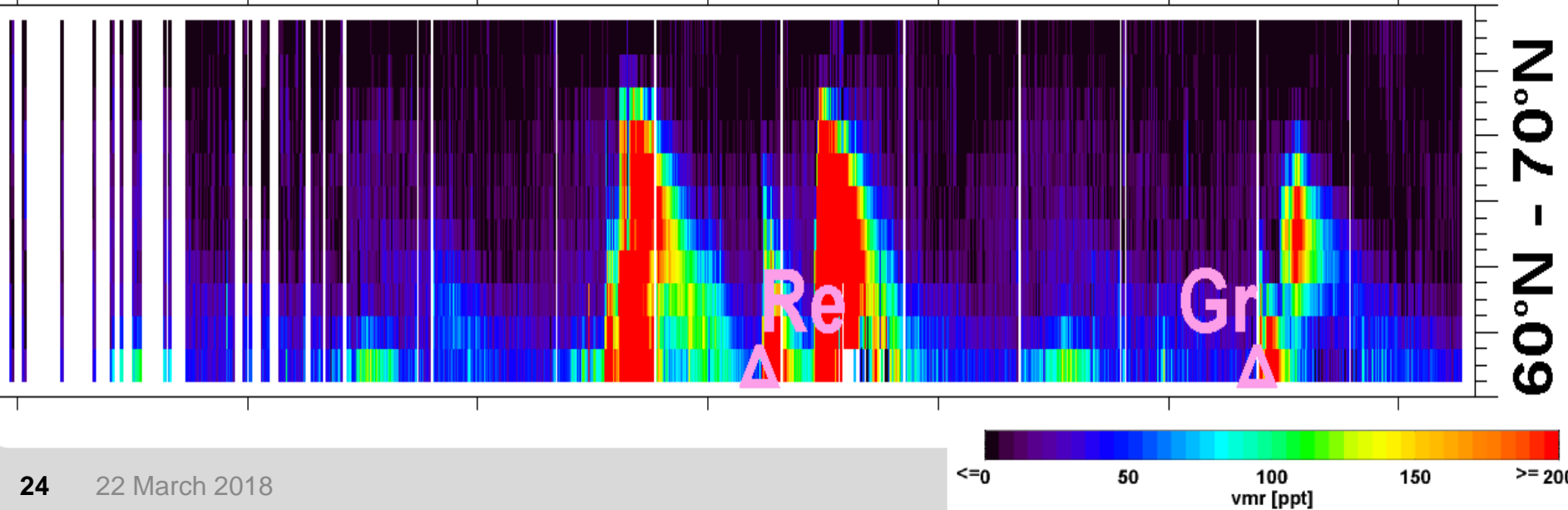
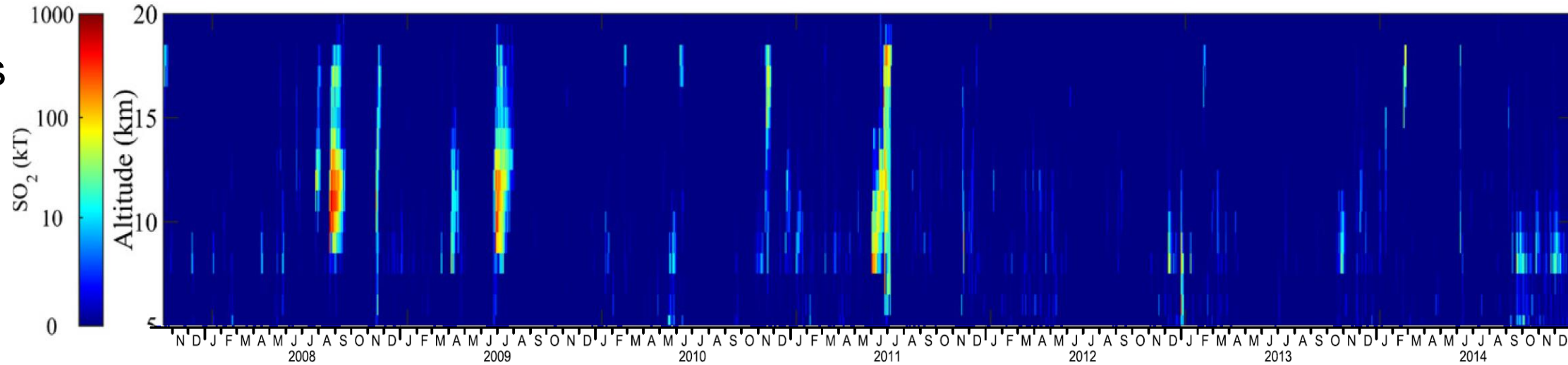
Carboni et al., 2016

- “Despite the ability of IASI to retrieve the plume altitude, it could be difficult to distinguish, with only IASI data, if the eruption injects into the stratosphere or into the high troposphere.”

# Lifetime of SO<sub>2</sub> in the UTLS

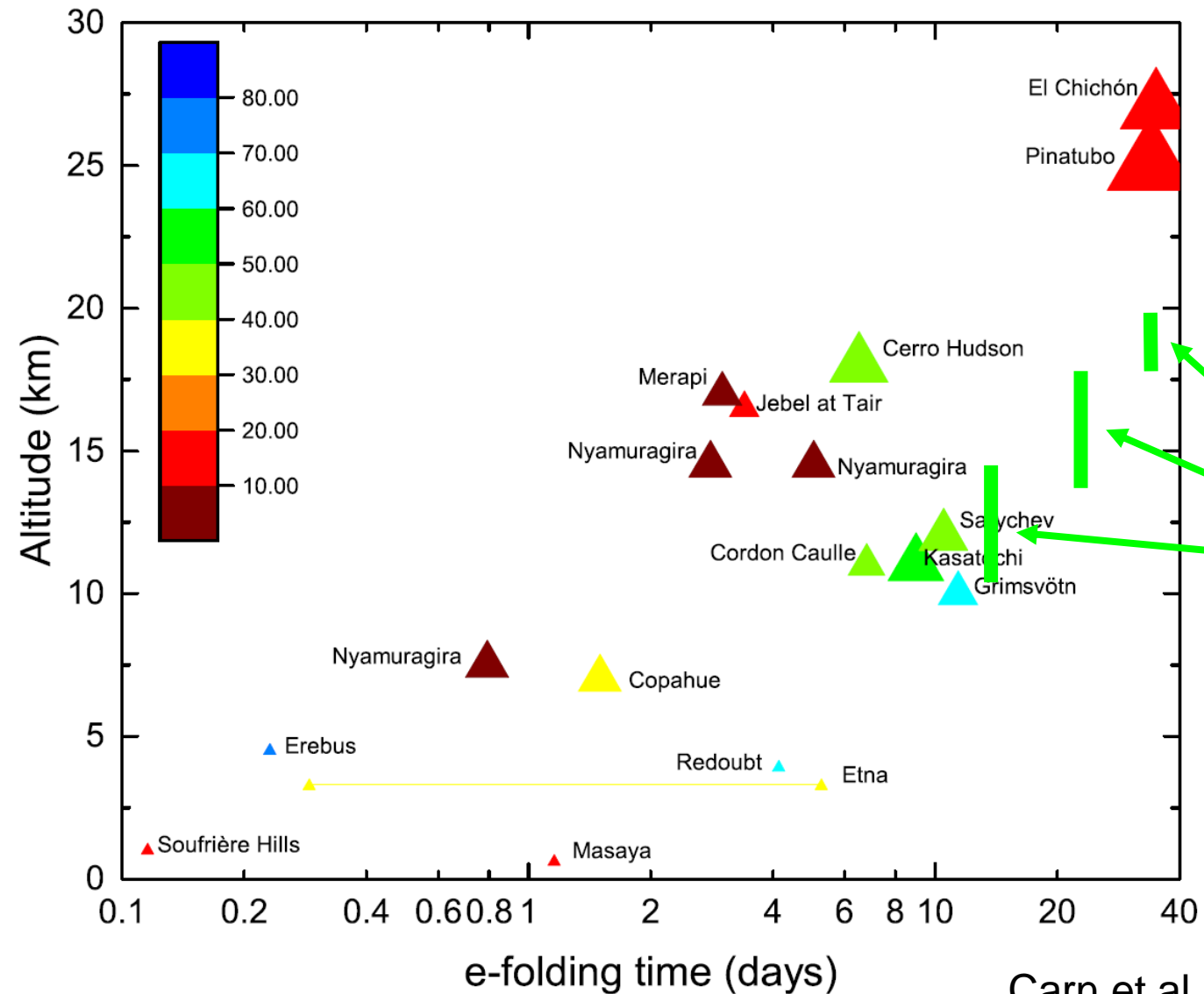
IASI  
total mass

Clarisse et al.,  
2012, 2014  
Carn et al.,  
2016



MIPAS SO<sub>2</sub> vmr

# Lifetime of SO<sub>2</sub> in the UTLS



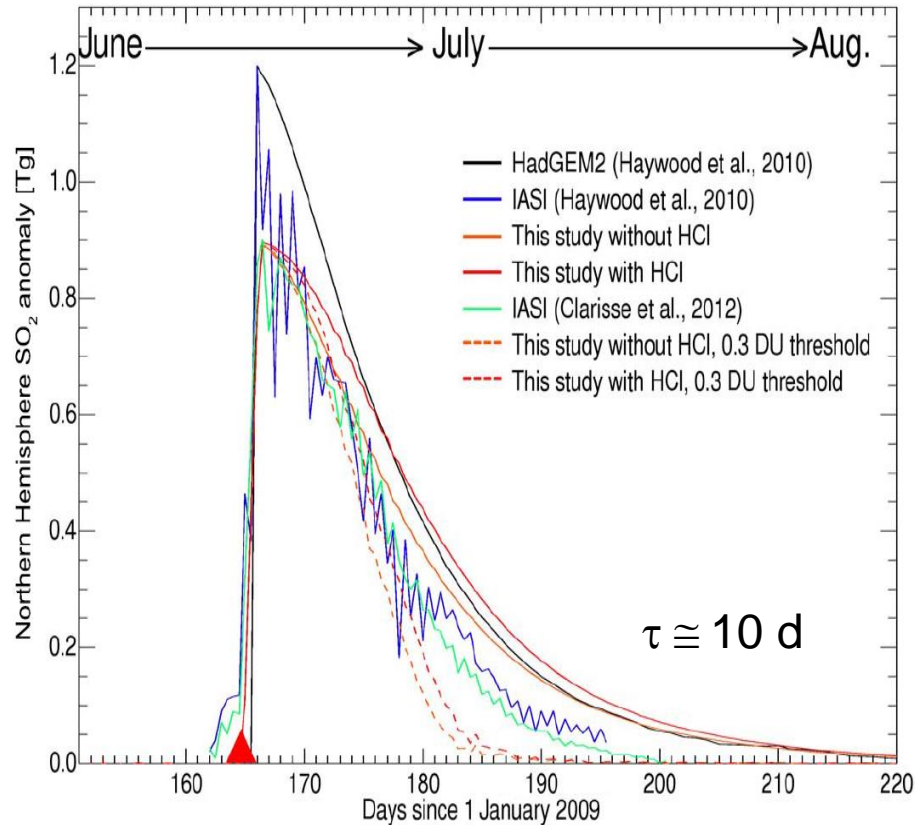
**Sarychev SO<sub>2</sub>  
e-folding times  
from limb-  
sounders**

Carn et al., 2016

# Lifetime of SO<sub>2</sub> in the UTLs

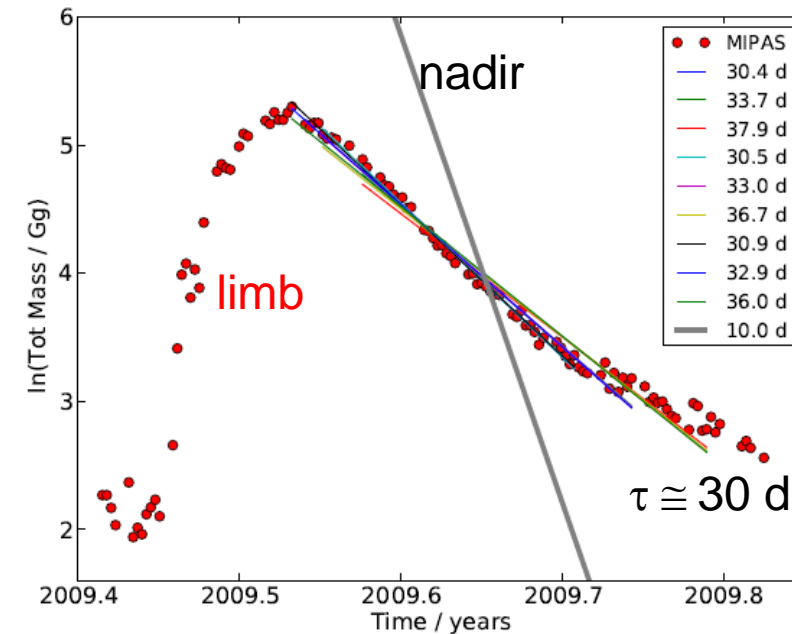
Sarychev

Nadir-Sounder IASI



Lurton et al., 2018

Limb-Sounder MIPAS (and MLS)



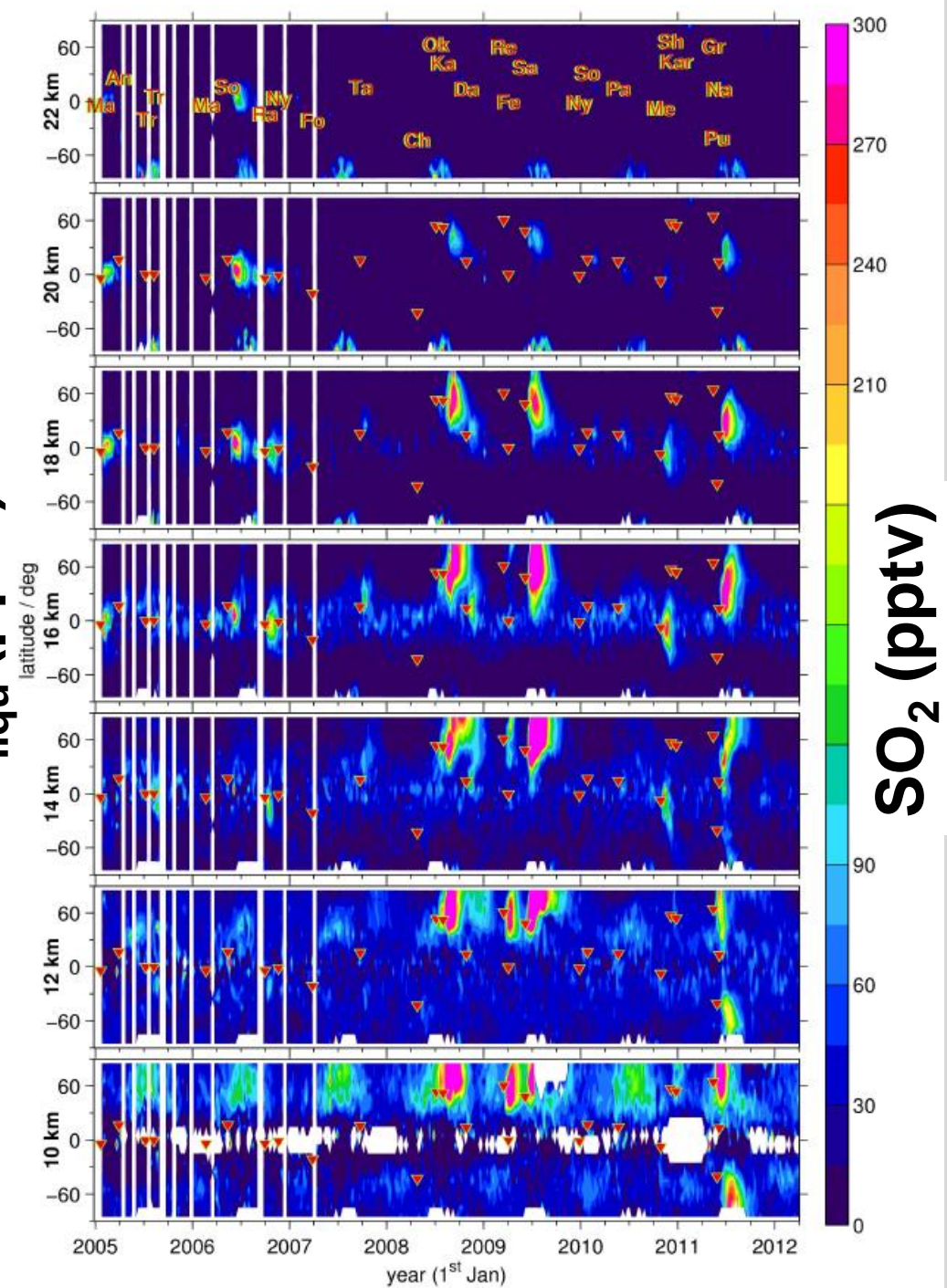
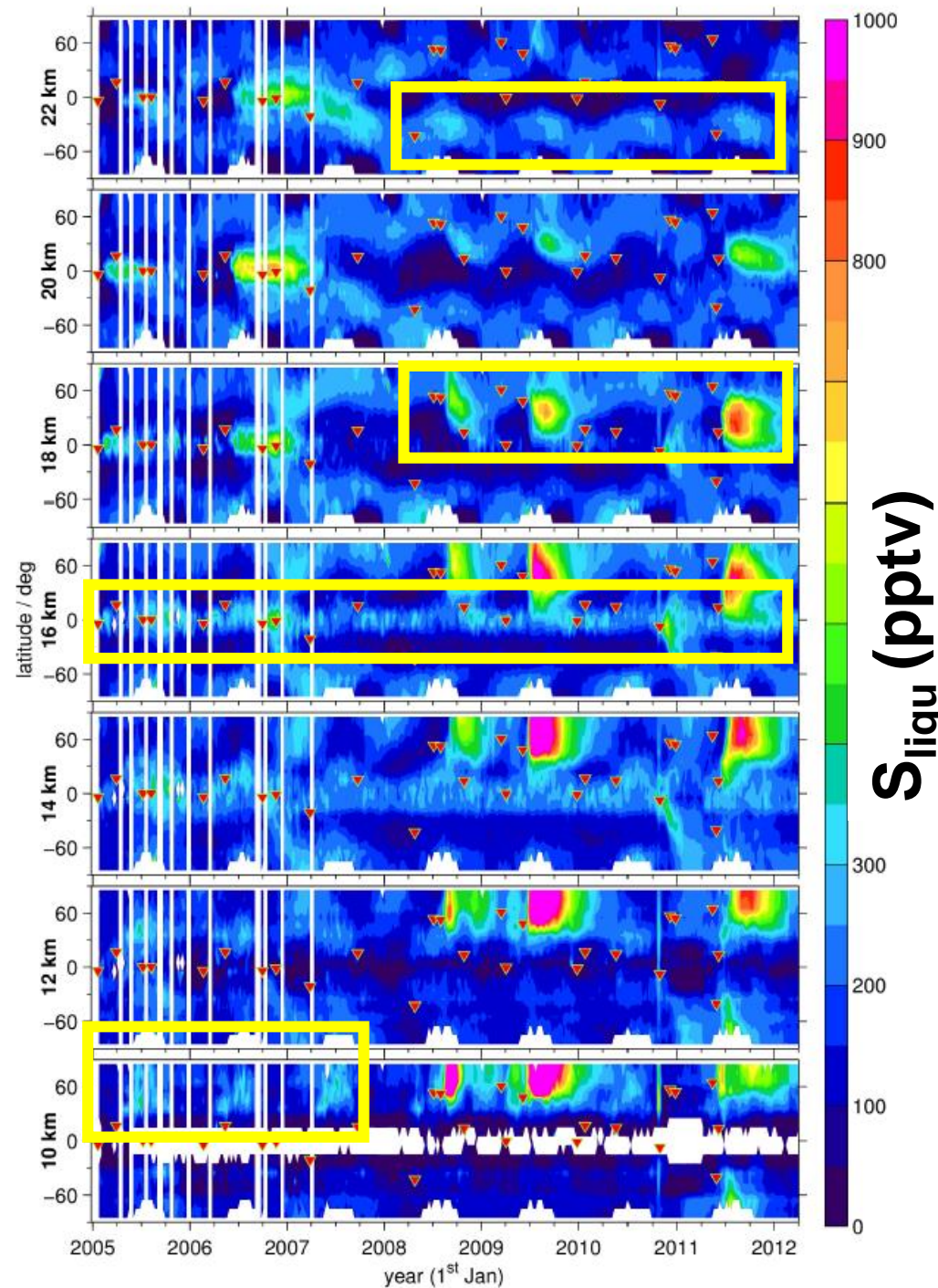
Nadir sounding instruments seem to underestimate the lifetime of volcanic SO<sub>2</sub> plumes in the UTLs:

- Detection-limit of nadir sounders (global dilution of SO<sub>2</sub>, e.g. Haywood et al., 2010)
- Combination of lower SO<sub>2</sub>-lifetime at lower altitudes and nadir averaging kernels

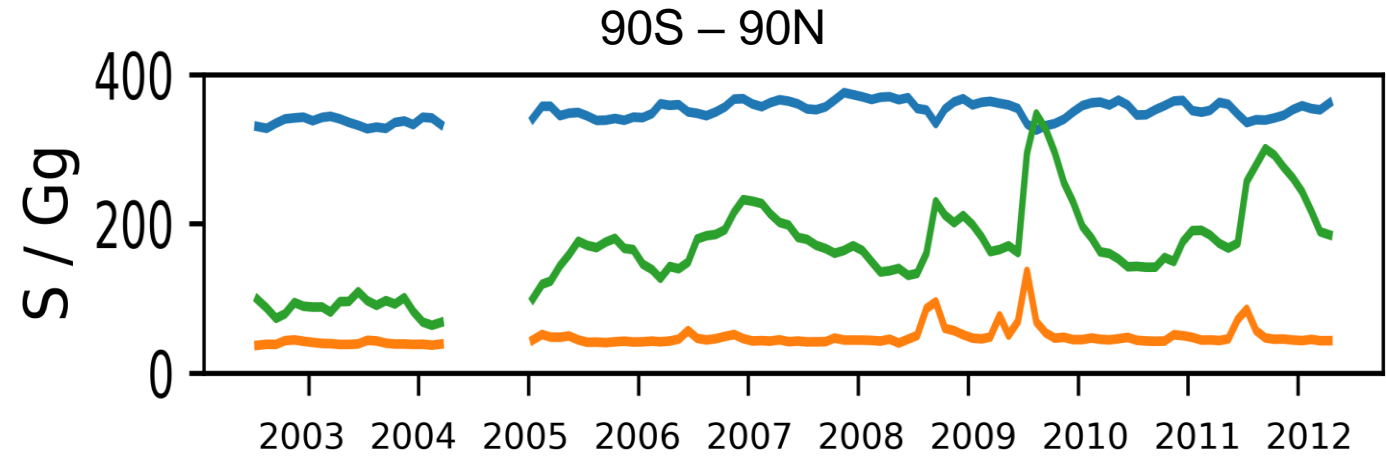
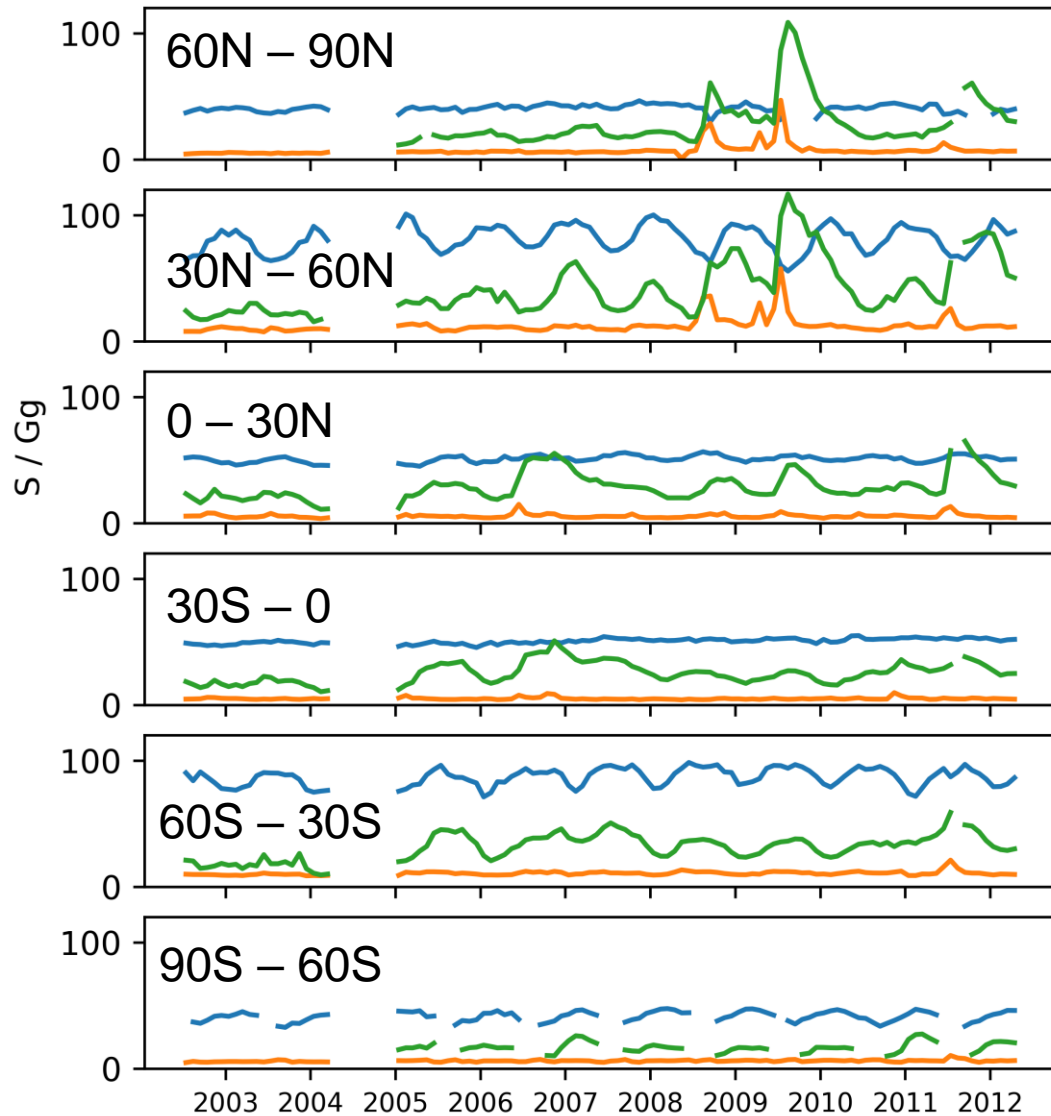


# New MIPAS dataset on aerosol volume

Günther et al.,  
ACP, 2018



# MIPAS: total stratospheric sulfur burden (monthly means)

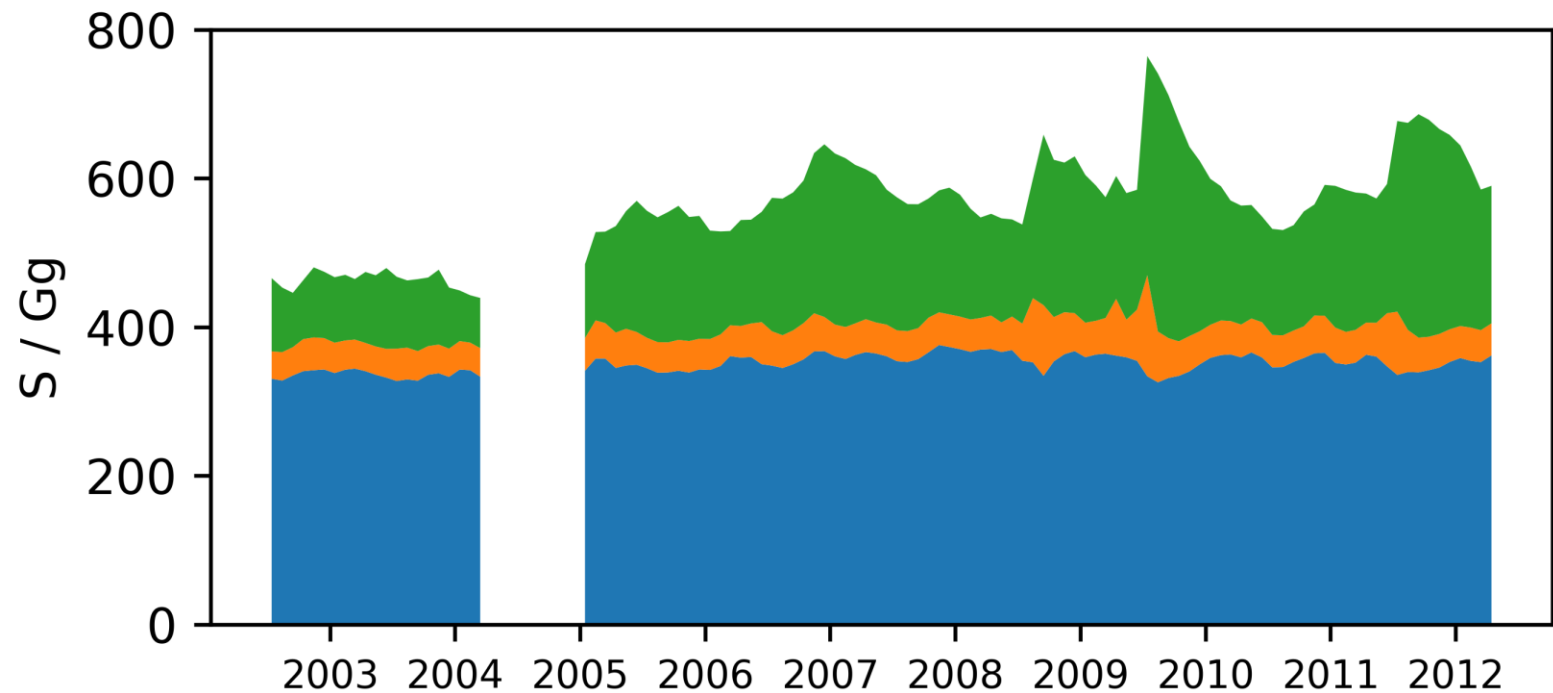


$S_{\text{OCS}}$   
 $S_{\text{SO}_2}$   
 $S_{\text{liqu}}$

Estimated errors:

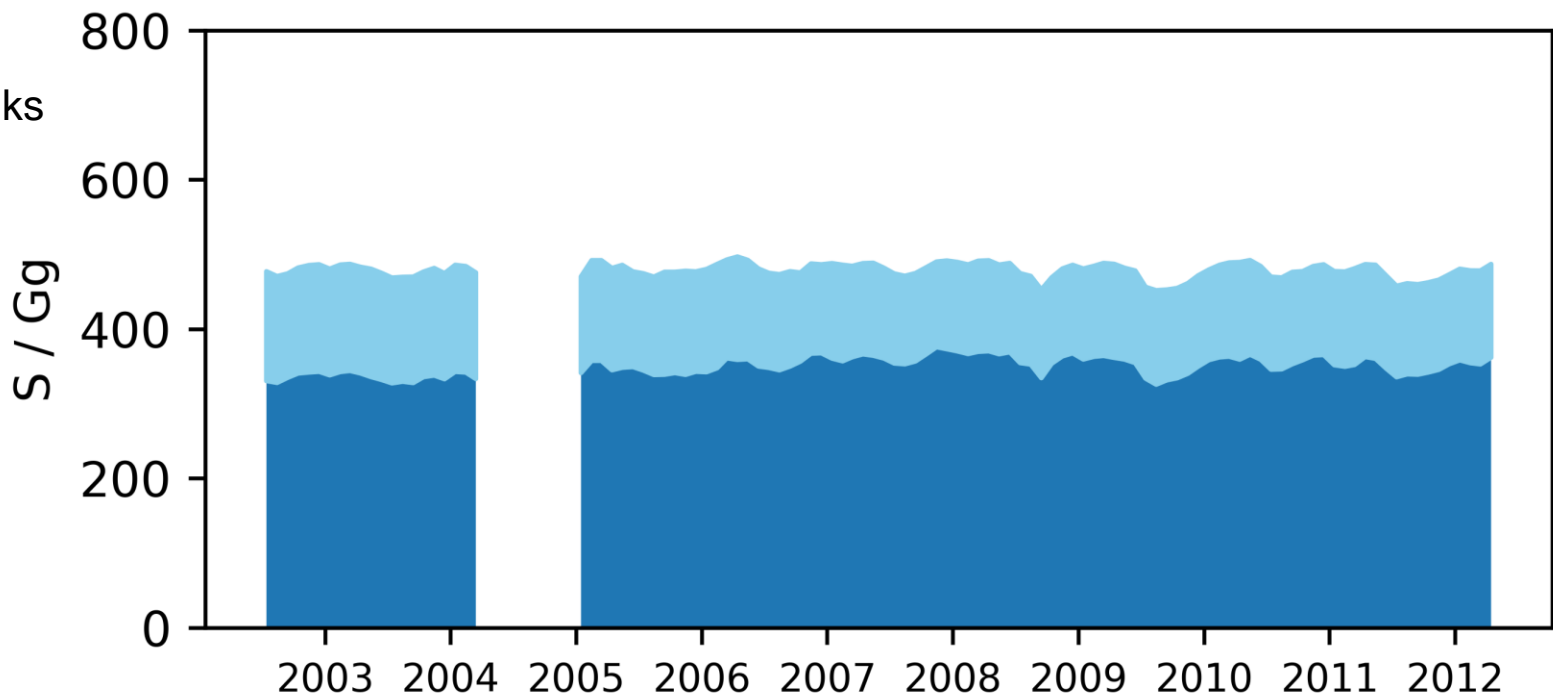
- OCS ~10-15% (positive bias wrt ACE?)
- $\text{SO}_2$  ~20% (+large underestimation until 2-4 weeks after larger eruption)
- Aerosol ~20%

# MIPAS: total cumulative stratospheric sulfur burden (monthly means)



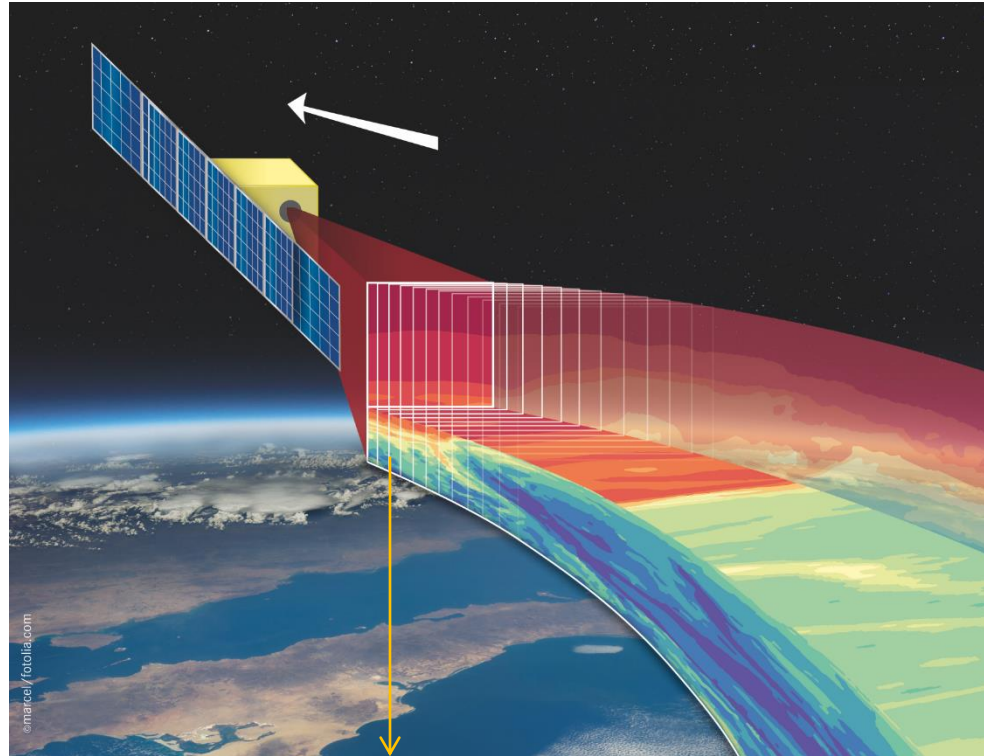
Estimated errors:

- OCS ~10-15% (positive bias wrt ACE?)
- $\text{SO}_2$  ~20% (+large underestimation until 2-4 weeks after larger eruption)
- Aerosol ~20%



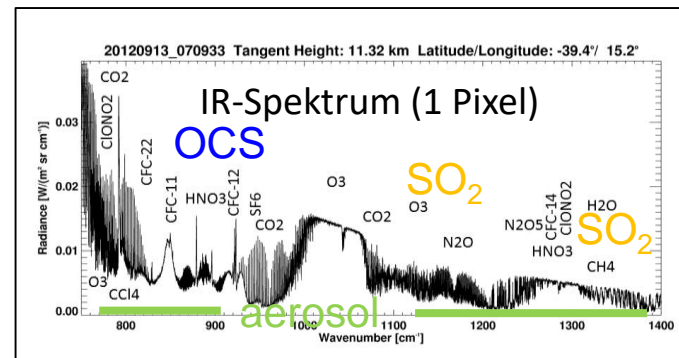


# German proposal for an infrared imaging limb-sounder



MIPAS+++ with:

- Across-track sounding
- Strongly enhanced vertical and horizontal resolution



WR

WISSENSCHAFTSRAT

ATMO-SAT

2017

Bericht zur  
wissenschaftsgeleiteten  
Bewertung umfangreicher  
Forschungsinfrastruktur-  
vorhaben für die Nationale  
Roadmap

# Summary

## OCS

- What is the effect of convection on the UT-OCS “hole”?
- How to explain the large UT OCS values over the W-Pacific in spring+summer?
- Does biomass burning really emit any OCS? If yes, where is it?
- Is there a tropical/subtropical upper tropospheric OCS-nose?
- Which instrument is right, MIPAS or ACE-FTS (OCS differ up to 20 km by about 50 pptv)
- Is there any stratospheric OCS trend not connected to changes in Age-of-air?

## SO<sub>2</sub>

- Are the new aircraft in-situ observations of < 10 ppt the “real” background values at the tropical upper troposphere? (supported by ACE-FTS)
- Do nadir instruments underestimate SO<sub>2</sub> lifetimes in the UTLS? If yes, is it an effect of sensitivity or of the lower peaking averaging kernel?

## H<sub>2</sub>SO<sub>4</sub>(liqu)

- Is the summertime enhancement seen in the new MIPAS aerosol volume dataset real?
- What is the reason for the annual cycle of downwelling aerosols in the S-hemisphere?
- How well can the stratospheric total aerosol burden be constrained?