Atmospheric sulfur from the upper troposphere to the upper stratosphere: 10 years of MIPAS observations

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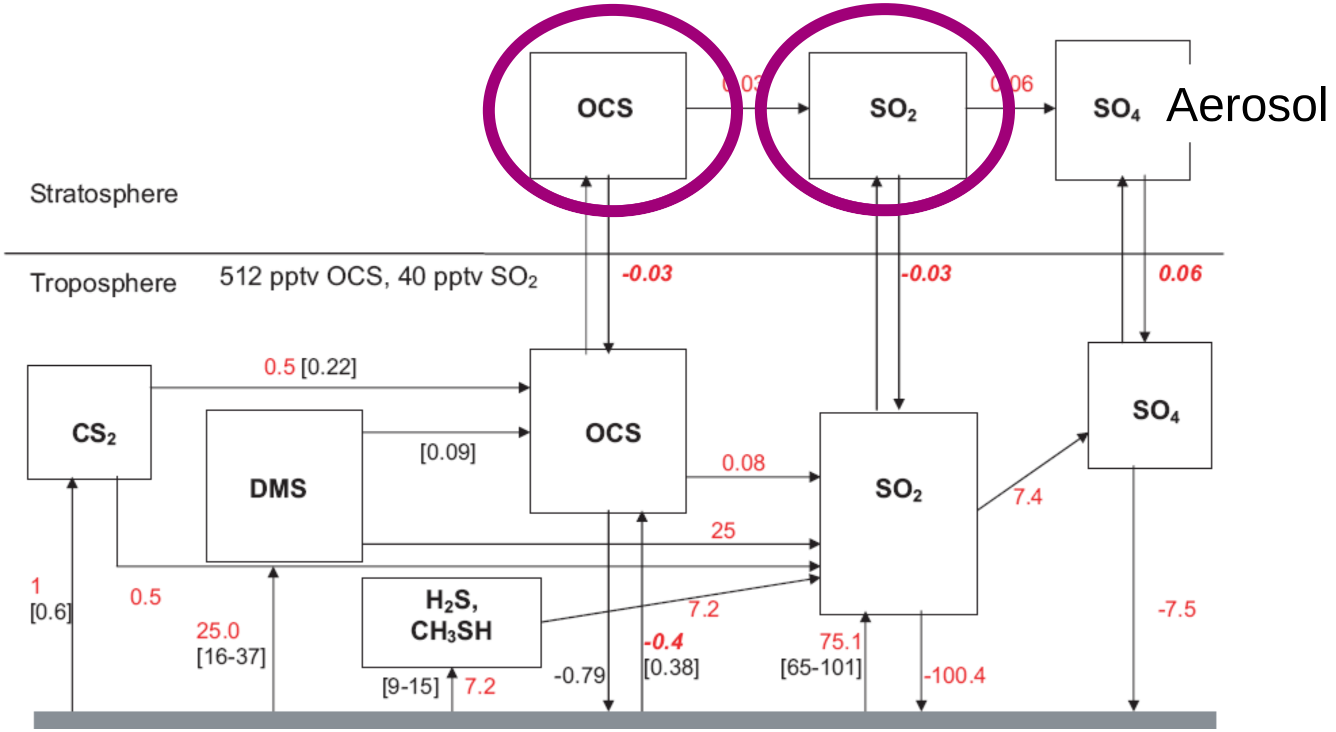
SO2 and OCS in the stratosphere: the aerosol layer



Assessment of Stratospheric Aerosol Properties, SPARC, 2006

The Persistently Variable "Background" Stratospheric Aerosol Layer and Global Climate Change
S. Solomon *et al.*
Science **333**, 866 (2011);
DOI: 10.1126/science.1206027

Sulfur in the troposphere: OCS and the carbon cycle

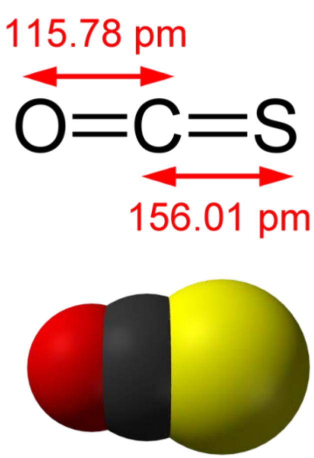
Photosynthetic Control of Atmospheric Carbonyl Sulfide During the Growing Season
J. E. Campbell, *et al.*
Science **322**, 1085 (2008);
DOI: 10.1126/science.1164015

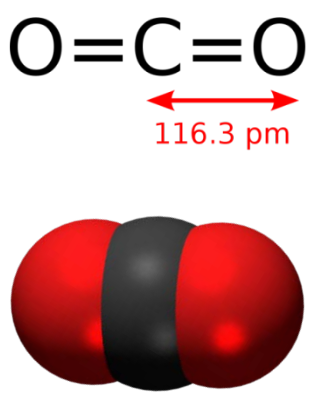
LETTERS
PUBLISHED ONLINE: 17 FEBRUARY 2013 | DOI: 10.1038/NGEO1730
nature geoscience

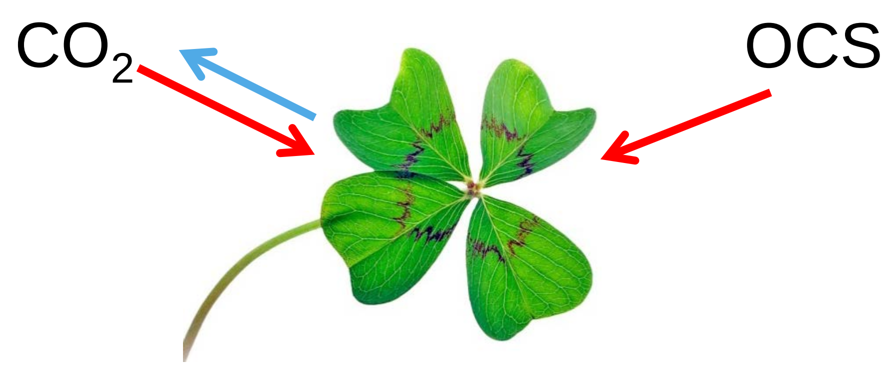
Ecosystem photosynthesis inferred from measurements of carbonyl sulphide flux

David Asaf[1], Eyal Rotenberg[1], Fyodor Tatarinov[1], Uri Dicken[1], Stephen A. Montzka[2] and Dan Yakir[1]*

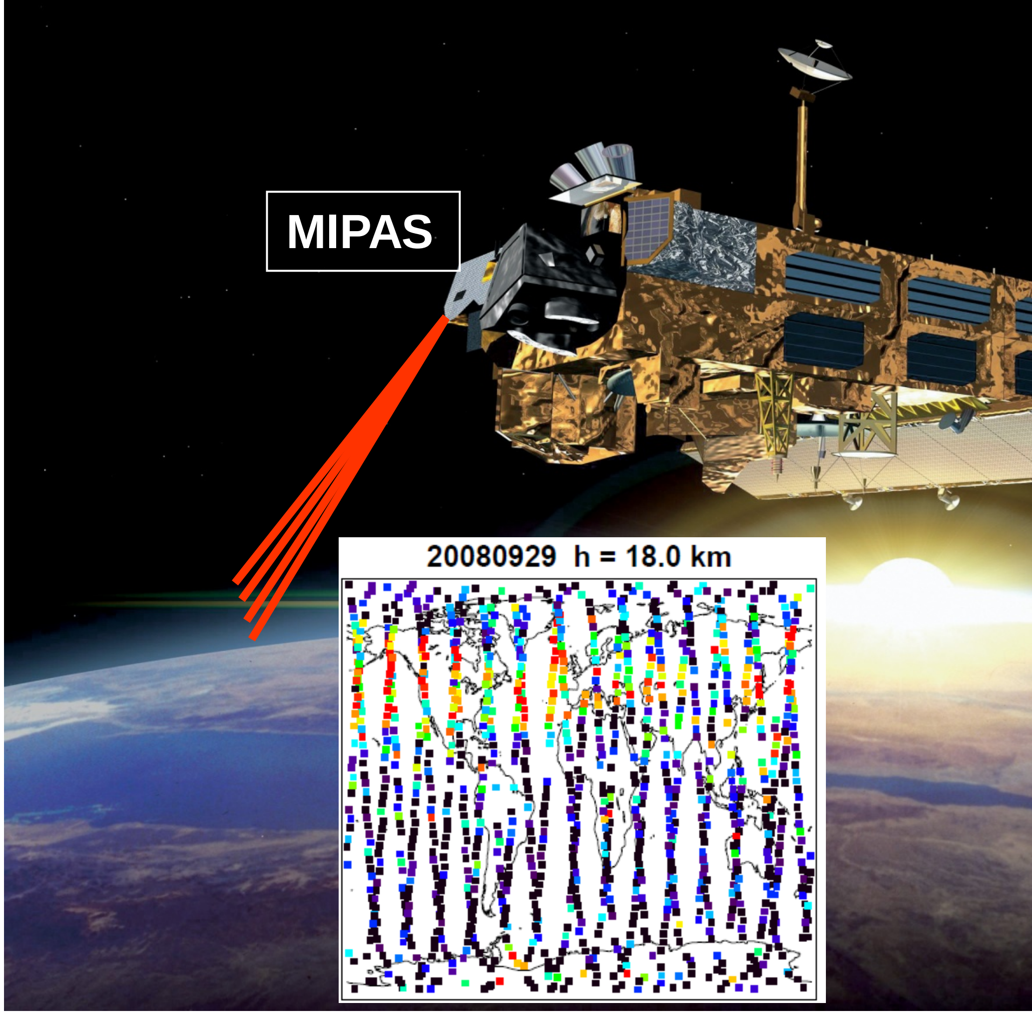
- Limited understanding of CO_2 sinks and sources on land due to difficult separation between the CO_2 taken up by photosynthesis and that released by respiration
- OCS as proxy for photosynthetic CO_2 uptake
- OCS and CO_2 have the same diffusion pathway into leaves
- Hydration reaction is irreversible for OCS: $OCS + H_2O \rightarrow H_2S + CO_2$

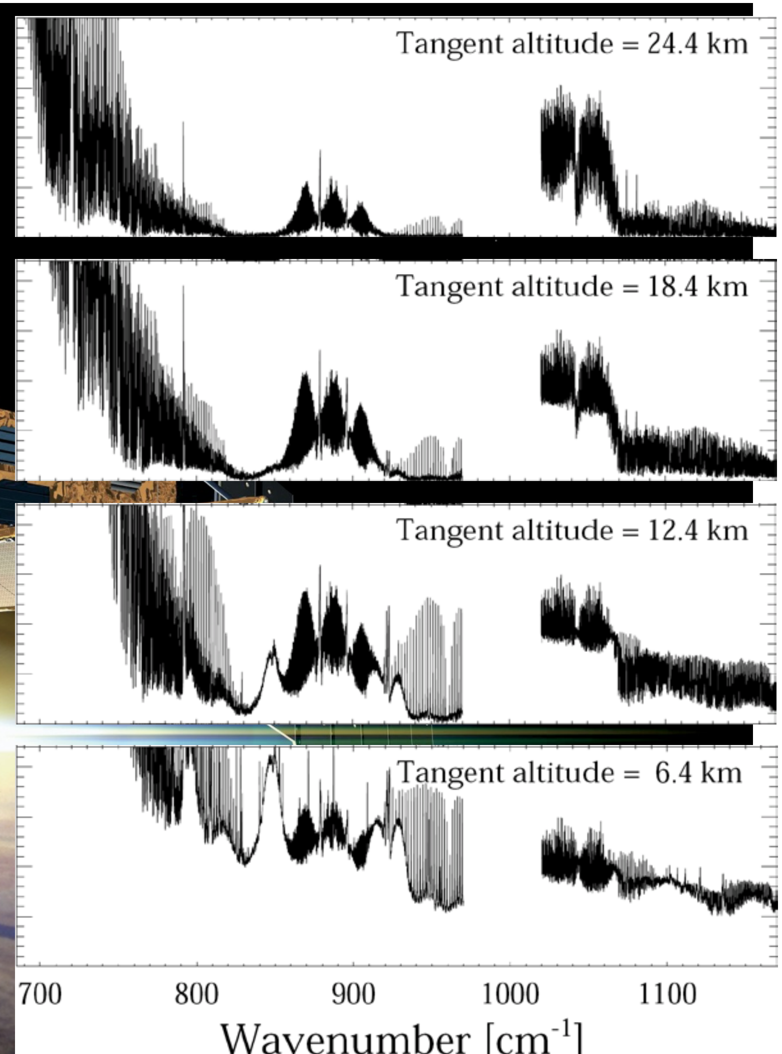






The Michelson Interferometer for Passive Atmospheric Sounding on Envisat





MIPAS products retrieved at KIT

SO_2

- Retrieval from mean spectra: 13-45 km, monthly+zonal averages → Höpfner et al., ACP, 2013
- Retrieval from single limb-scans: 8-20 km, high temporal and horizontal resolution → Höpfner et al., ACPD, 2015

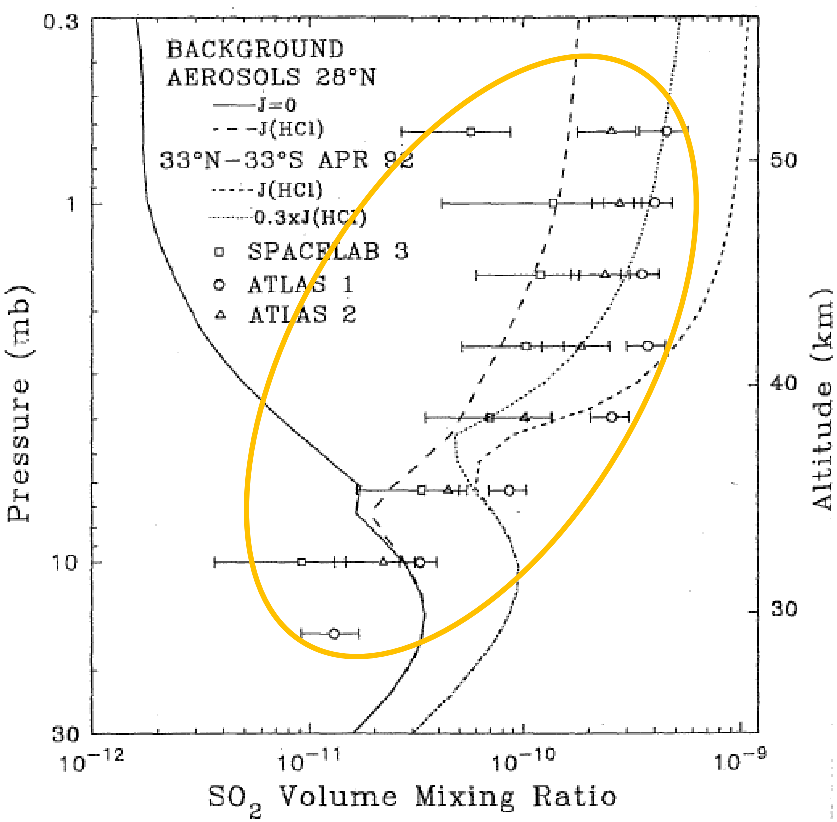
OCS

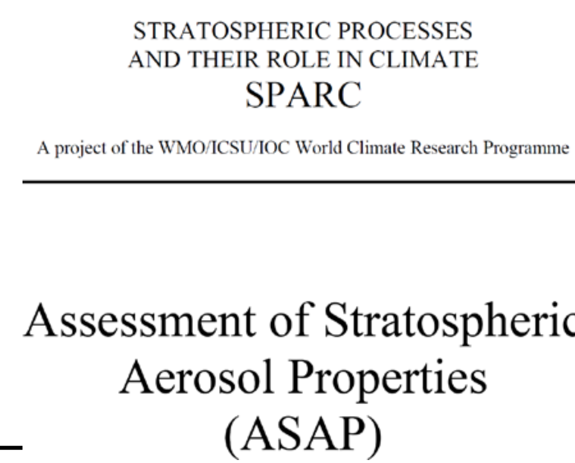
- Retrieval from single limb-scans: 8-35 km, high temporal and horizontal resolution → Glatthor et al., subm. 2015

SO_2 from mean MIPAS spectra

Pre-MIPAS observations of SO_2 in the stratosphere

ATMOS: Rinsland et al., 1995



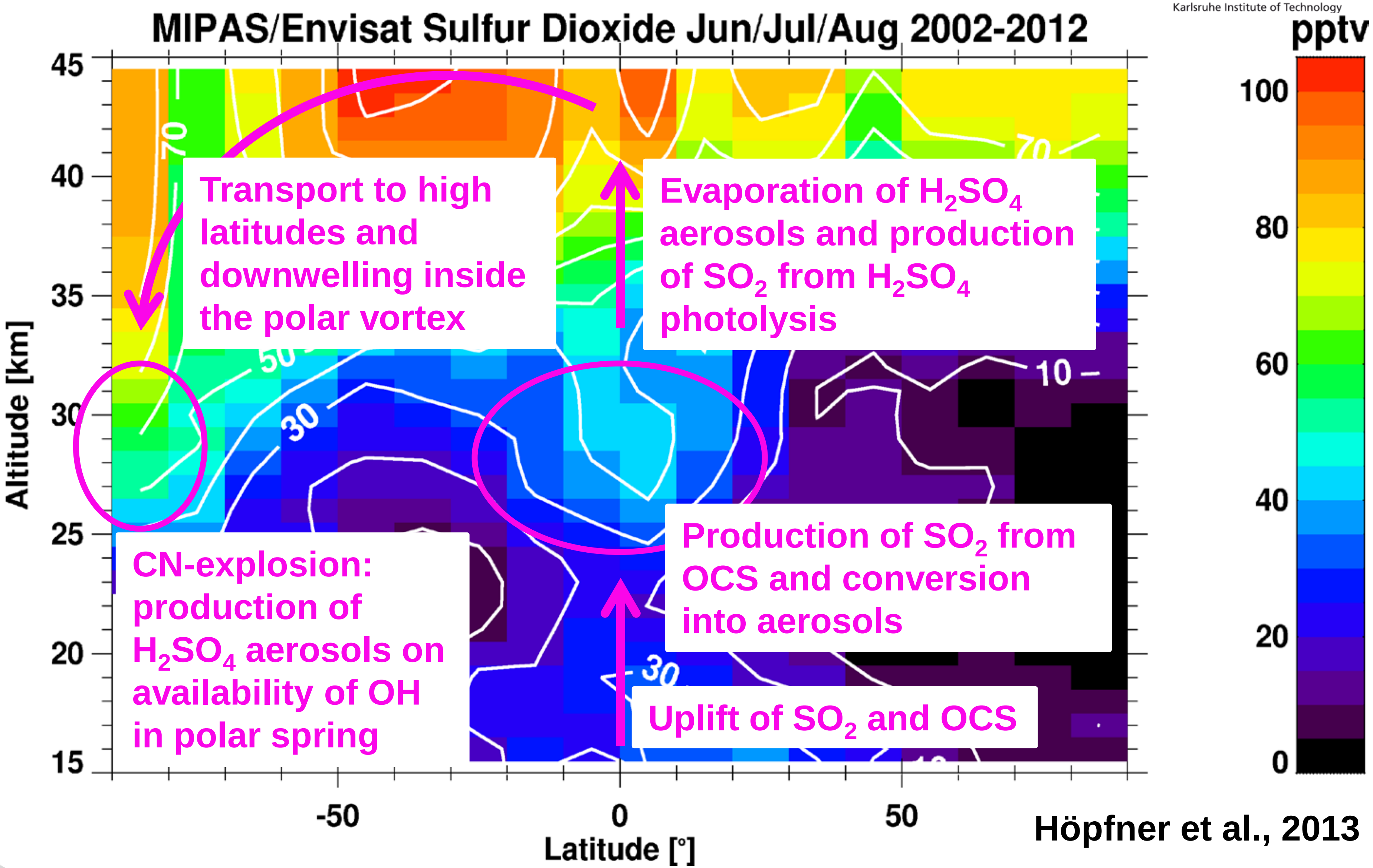

STRATOSPHERIC PROCESSES
AND THEIR ROLE IN CLIMATE
SPARC

A project of the WMO/ICSU/IOC World Climate Research Programme

Assessment of Stratospheric Aerosol Properties (ASAP)

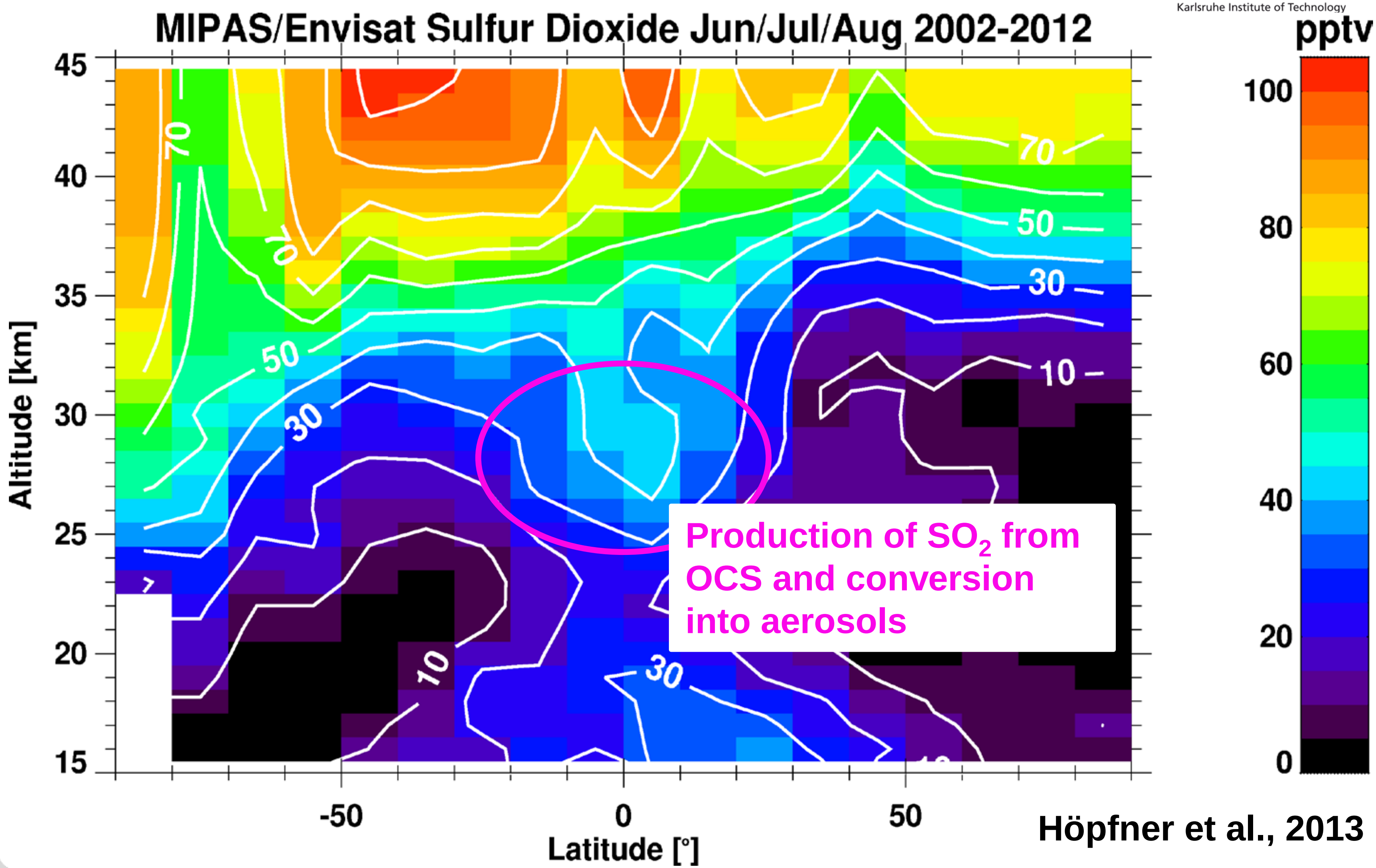
- **Observations of SO_2 in the upper troposphere and lower stratosphere and of H_2SO_4 and SO_2 in the middle and upper stratosphere would be extremely valuable to improve our modeling and predictive capabilities of stratospheric aerosol.** Currently, there is a general scarcity of measurements of key sulfur-bearing gases during their transport from the upper troposphere into the upper stratosphere.

First global measurements of SO_2 throughout the stratosphere



Höpfner et al., 2013

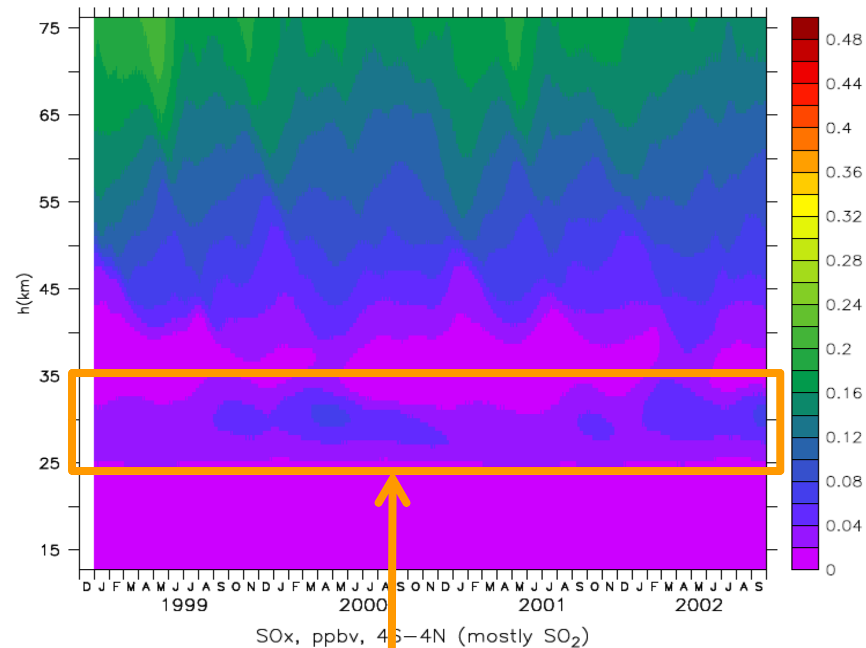
First global measurements of SO_2 throughout the stratosphere



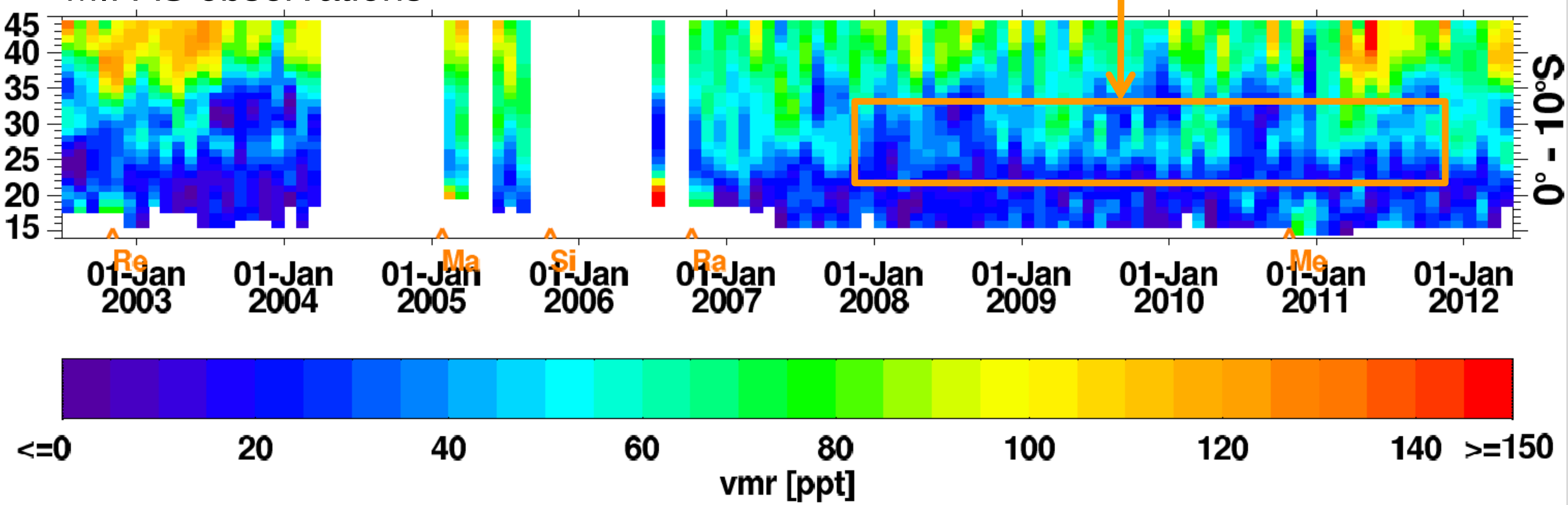
Höpfner et al., 2013

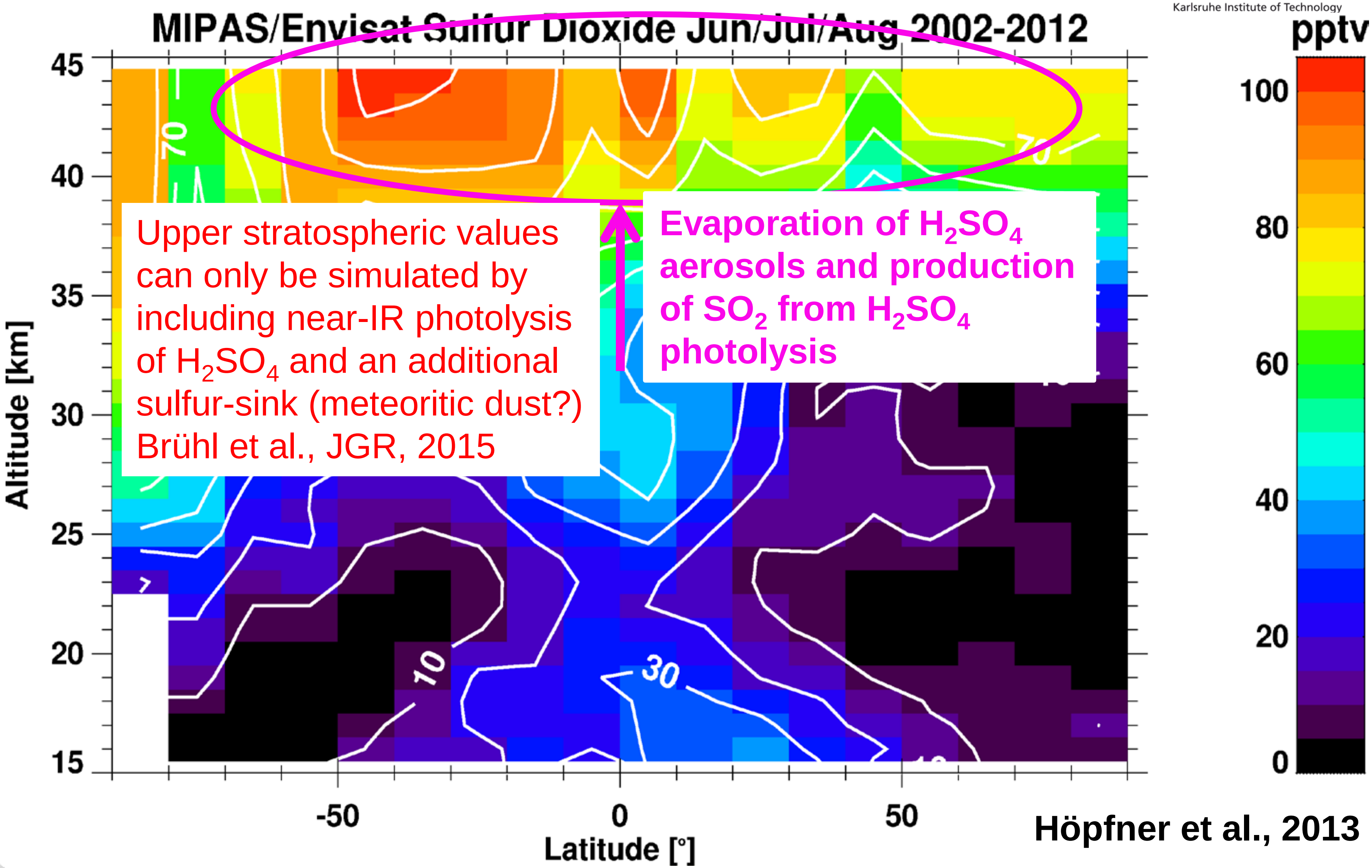
The background aerosol layer: production from OCS at 25-30 km

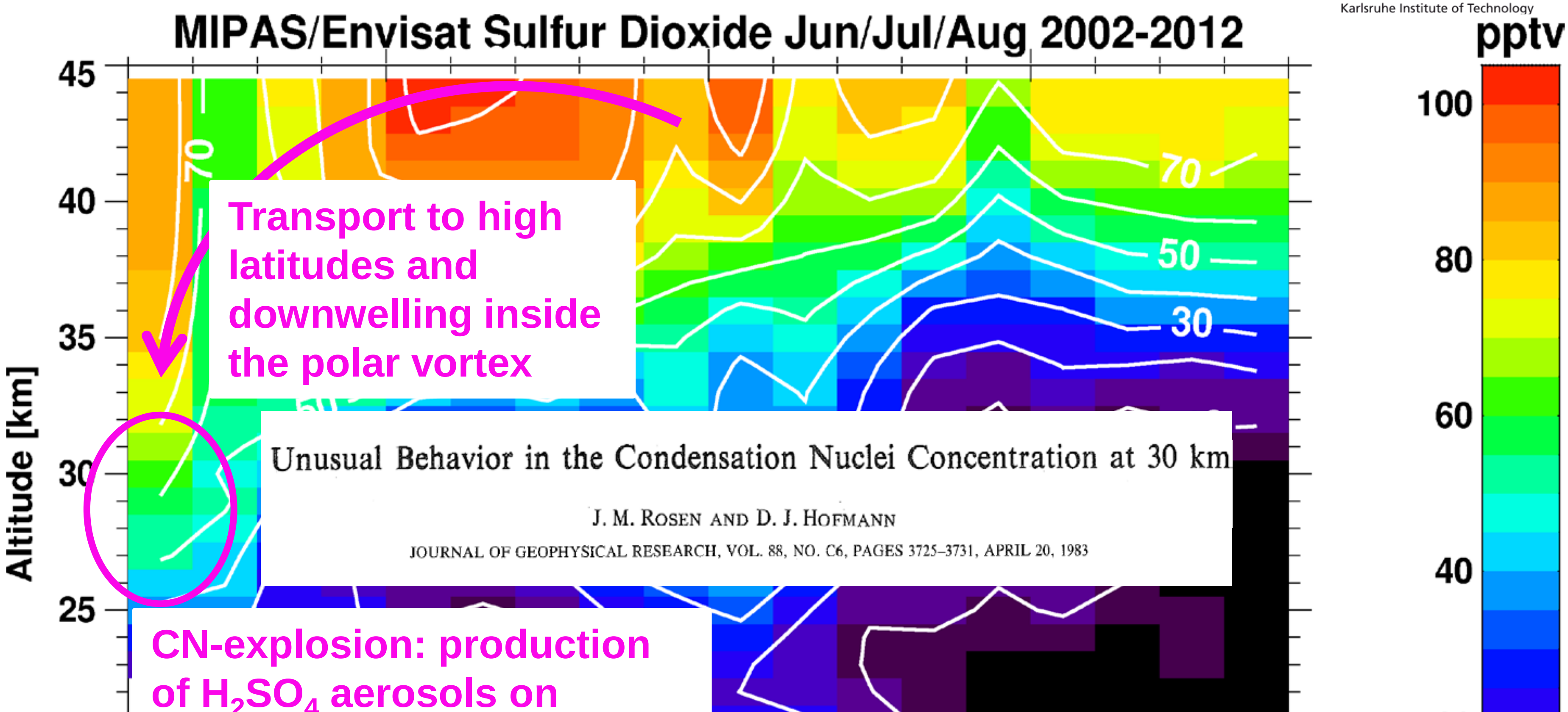
Model SO2:
Brühl et al., ACP, 2012



MIPAS observations



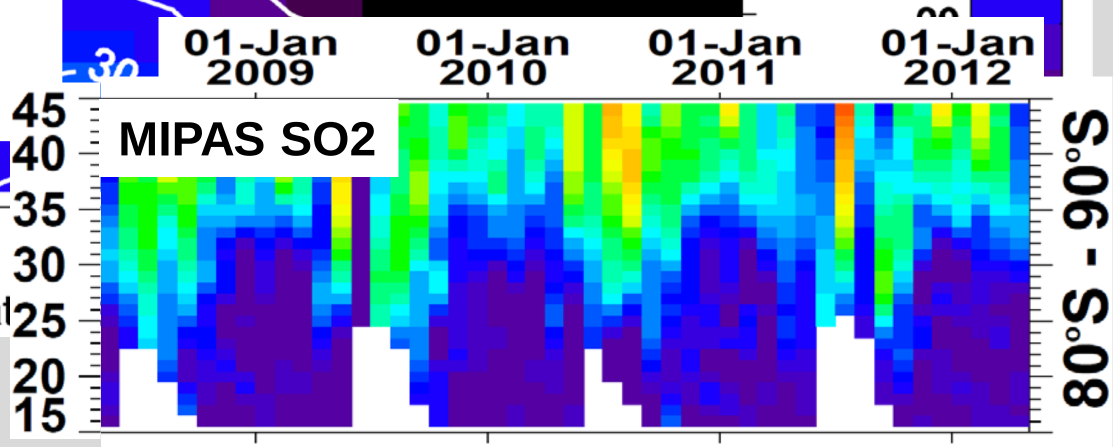

MIPAS/Envisat Sulfur Dioxide Jun/Jul/Aug 2002-2012
Altitude [km]
45
40
35
30
25
20
15
-50
0
50
Latitude [°]
pptv
100
80
60
40
20
0
70
10
30
Upper stratospheric values can only be simulated by including near-IR photolysis of H_2SO_4 and an additional sulfur-sink (meteoritic dust?) Brühl et al., JGR, 2015
Evaporation of H_2SO_4 aerosols and production of SO_2 from H_2SO_4 photolysis
Höpfner et al., 2013



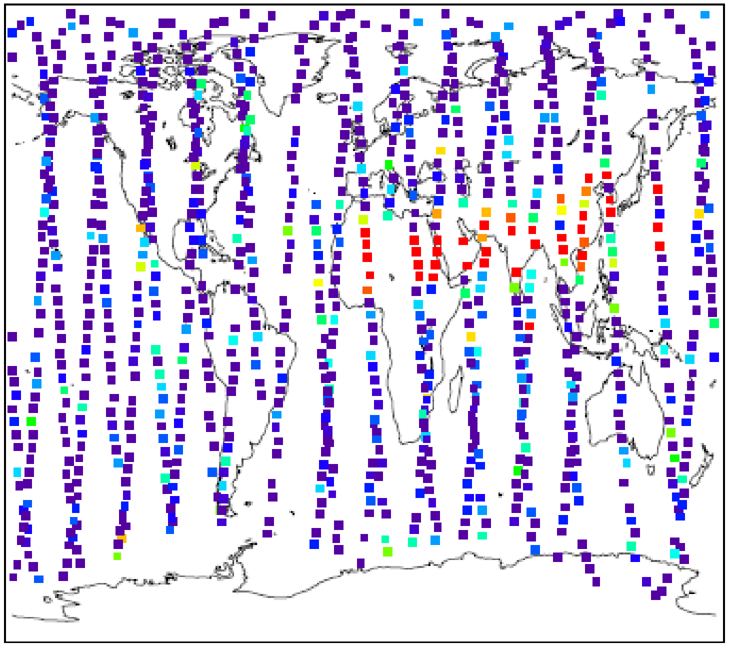
Unusual Behavior in the Condensation Nuclei Concentration at 30 km

J. M. ROSEN AND D. J. HOFMANN

JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 88, NO. C6, PAGES 3725–3731, APRIL 20, 1983



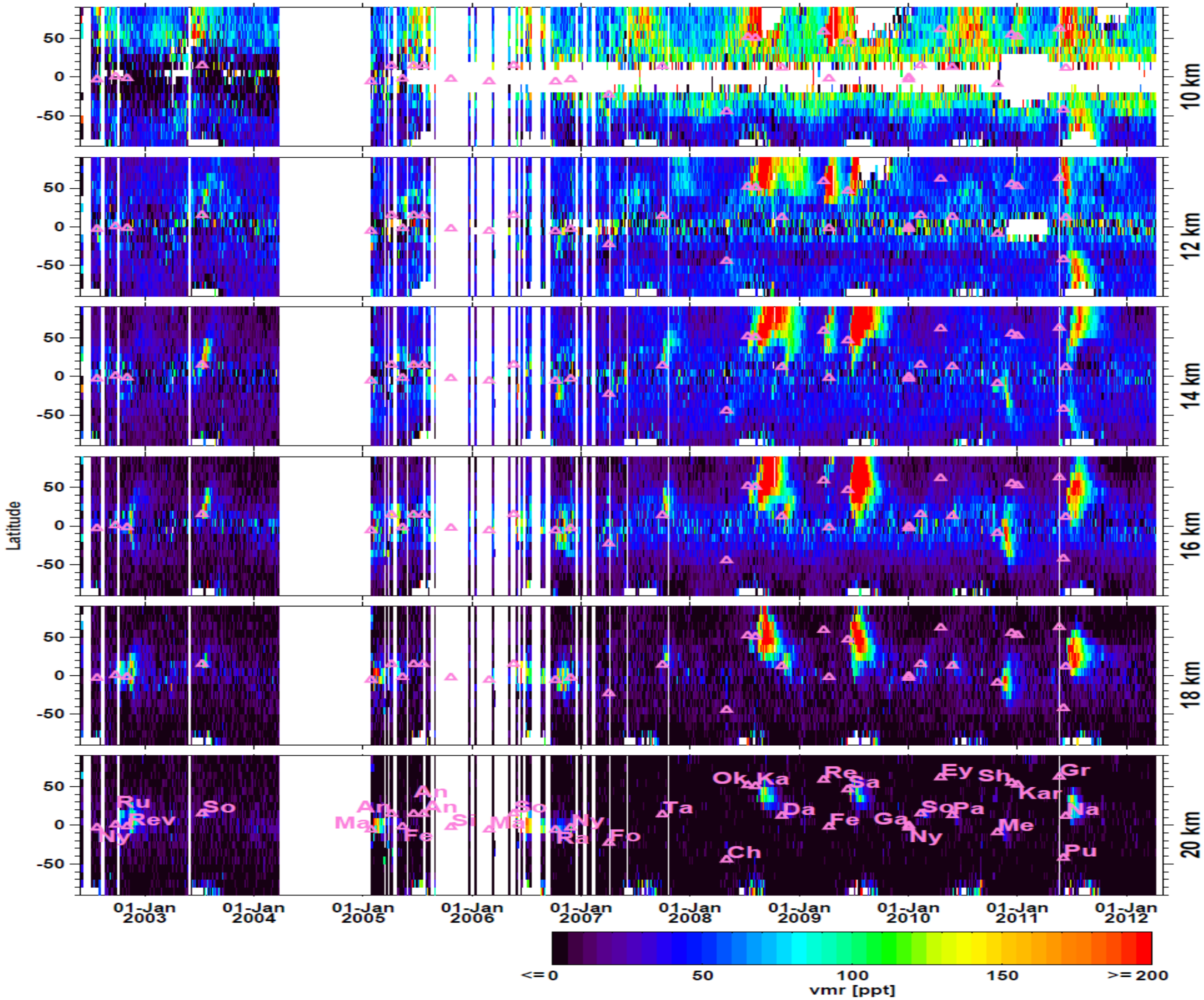
SO_2 from single MIPAS limb-scans



Höpfner et al., ACPD, 2015

Global measurements of vertically resolved volcanic plumes → Injection mass of SO_2 for climate-chemistry models

Karlsruhe Institute of Technology



An Anatahan

Ch Chaiten
Da Dalaffilla
Ey Eyjafjallajökull
Fe Fernandina

Fo Fournaise, Piton de la
Ga Galeras
Gr Grímsvötn
Kar Karymsky
Ka Kasatochi
Ma Manam

Me Merapi
Na Nabro
Ny Nyamuragira

Ok Okmok
Pa Pacaya
Pu Puyehue-Cordon Caulle
Ra Rabaul
Re Redoubt
Rev Reventador
Ru Ruang
Sa Sarychev
Sh Shiveluch
Si Sierra Negra
So Soufrière Hills

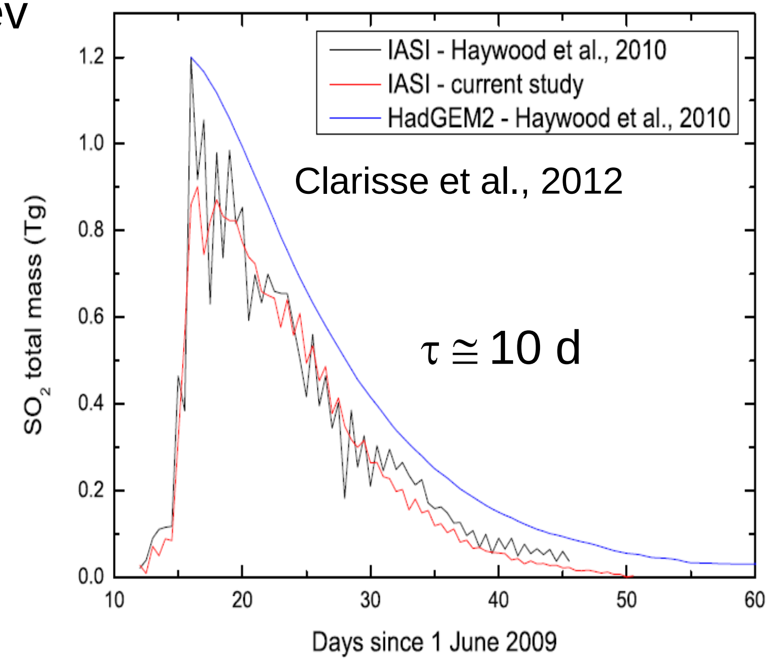
Ta Tair, Jebel al

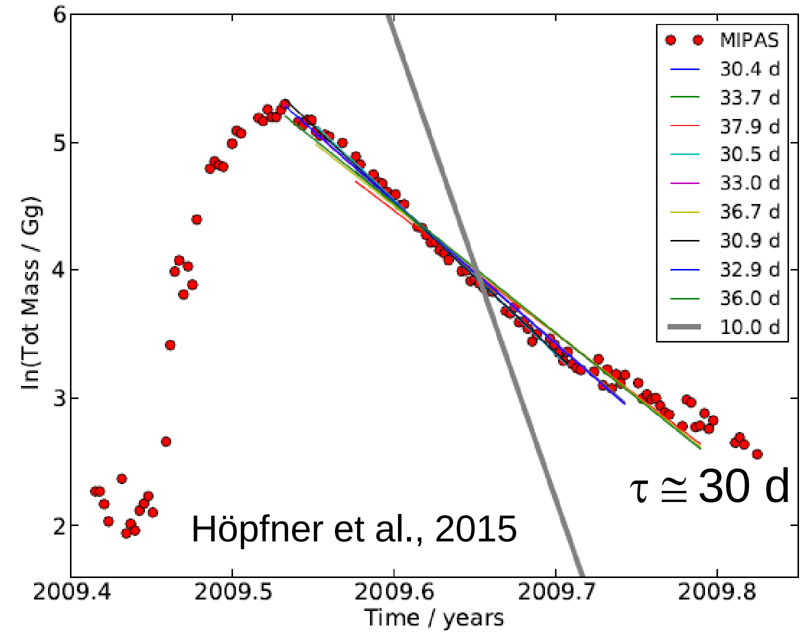
	Name	Eruption date	Location ° N/° E	$M(t_0)$ [Gg] if present: τ [d]	$M(t_0)$ [Gg] if present: τ [d] from other sources
Ny	Nyamuragira	25 Jul 2002	−1.4/29.2	22(1)/12(1)/3(0)/**37(2)**[a]	
Ru	Ruang	25 Sep 2002	2.3/125.4	36(19)/39(9)/15(2)/**90(21)**[b]	74[1]
Rev	Reventador	3 Nov 2002	−0.1 / −77.7	54(47)/29(6)/12(2)/**94(47)**[b]	65–84[1]; 100[2]
So	Soufrière Hills	12 Jul 2003	16.7/−62.2	68(19)/28(7)/2(1)/**98(20)**[b]	100–128[3]; 140[1]
Ma	Manam	27 Jan 2005	−4.1/145.0	79(15)/87(9)/39(3)/**206(17)**[a]	180[1]; $99 \pm 13 (> 68.1\,\text{hPa})$[4]
An	Anatahan	6 Apr 2005	16.4/145.7	34(11)/34(7)/0(0)/**68(13)**[a]	165[1]
Tr	TropVolc	mid-Jul 2005	0.0/0.0	38(17)/21(5)/1(0)/**60(18)**[a]	
Tr	TropVolc	mid-Aug 2005	0.0/0.0	61(26)/23(5)/3(1)/**88(27)**[a]	
Ma	Manam	27 Feb 2006	−4.1/145.0	21(4)/58(8)/1(0)/**80(9)**[a]	
So	Soufrière Hills	20 May 2006	16.7/−62.2	40(29)/38(4)/85(15)/**162(33)**[a]	200[1]; 123–233[5]; $139 \pm 24 (> 68.1\,\text{hPa})$[4]
Ra	Rabaul	7 Oct 2006	−4.3/152.2	75(26)/118(34)/12(4)/**205(43)**[b]	125[1]; 230[2]; $190 \pm 14 (> 100\,\text{hPa})$[4]
Ny	Nyamuragira	27 Nov 2006	−1.4/29.2	49(6)/5(0)/–/**54(6)**[a]	58–216[1]
Fo	Fournaise, Piton de la	4 Apr 2007	−21.2/55.7	57(10)/12(1)/2(1)/**71(10)**[a]	$140 (> 7.5\,\text{km})$[6]
Ta	Tair, Jebel at	30 Sep 2007	15.6/41.8	26(11)/27(5)/3(1)/**56(12)**[b]	46–57[7]
Ch	Chaiten	2 May 2008	−42.8/−72.7	26(7)/2(0)/2(0)/**30(7)**[a]	10[8]; 6[9]
Ok	Okmok	12 Jul 2008	53.4/−168.1	110(41)/31(6)/2(0)/**143(41)**[b]	200–300[5]; 100–200[10]
Ka	Kasatochi	7 Aug 2008	52.2/−175.5	645(127)/210(86)/43(8)/**899(154)**[c]	900–2700[11]; 2200[12]; $1000 (> 10\,\text{km})$[13] 1200[5]; 1700[9]; 1600[14]; $1350 \pm 38 (> 215\,\text{hPa})$[4]
				$\tau = 14(1)/23(5)/32(4)$	$\tau = 8$–9[12]; 18[9]; ≈10[14]; $27 \pm 1 (> 215\,\text{hPa})$[4]
Da	Dalaffilla	3 Nov 2008	13.8/40.5	31(9)/47(10)/1(0)/**79(13)**[b]	100–200[15]
Re	Redoubt	23 Mar 2009	60.5/−152.7	182(10)/18(7)/–/**200(12)**[c]	225–335[16]
				$\tau = 24(1)/22(6)/–$	
Fe	Fernandina	10 Apr 2009	−0.4/−91.6	14(2)/11(3)/2(0)/**27(4)**[a]	
Sa	Sarychev	12 Jun 2009	48.1/153.2	888(293)/542(60)/44(4)/**1473(299)**[c]	1200[17]; 900[14]; $571 \pm 42 (> 147\,\text{hPa})$[4] $1160 \pm 180 (> 215\,\text{hPa})$[4]
				$\tau = 15(2)/25(1)/38(2)$	$\tau = 27 \pm 2 (> 147\,\text{hPa})$[4]; $17 \pm 3 (> 215\,\text{hPa})$[4]; $\tau = 10$–11[17]; ≈10[14]
Ny	Nyamuragira	2 Jan 2010	−1.4/29.2	17(5)/3(1)/2(0)/**22(6)**[b]	
So	Soufrière Hills	11 Feb 2010	16.7/−62.2	11(3)/12(2)/5(1)/**28(4)**[b]	50[18]
Pa	Pacaya	28 May 2010	14.4/−90.6	–/10(2)/4(1)/**14(2)**[b]	20[19]
Me	Merapi	4 Nov 2010	−7.5/110.4	–/253(61)/23(7)/**276(61)**[c]	440[20]
				$\tau = –/15(2)/24(7)$	
Sh	Shiveluch	12 Dec 2010	56.7/161.4	18(4)/1(0)/0(0)/**20(4)**[a]	
Kar	Karymsky	1 Jan 2011	54.0/159.4	–/–/1(0)/**1(0)**[a]	
Gr	Grímsvötn	21 May 2011	64.4/−17.3	273(101)/2(0)/–/**276(101)**[a]	350–400[14]; $108 \pm 11 (> 215\,\text{hPa})$[4]
Pu	Puyehue-Cordón Caulle	4 Jun 2011	−40.6/−72.1	185(33)/–/–/**185(33)**[c]	250[14]
				$\tau = 32(3)/–/–$	$\tau = 6.8$[22]
Na	Nabro	12 Jun 2011	13.4/41.7	131(86)/343(79)/65(5)/**539(117)**[c]	1500[14]; $650 (> 10\,\text{km})$[21] $543 \pm 45 (> 147\,\text{hPa})$[4]
				$\tau = 11(3)/23(2)/ 27(1)$	$\tau = 20 \pm 2 (> 147\,\text{hPa})$[4]

SO_2-lifetime: differences between nadir and limb

Karlsruhe Institute of Technology

Sarychev

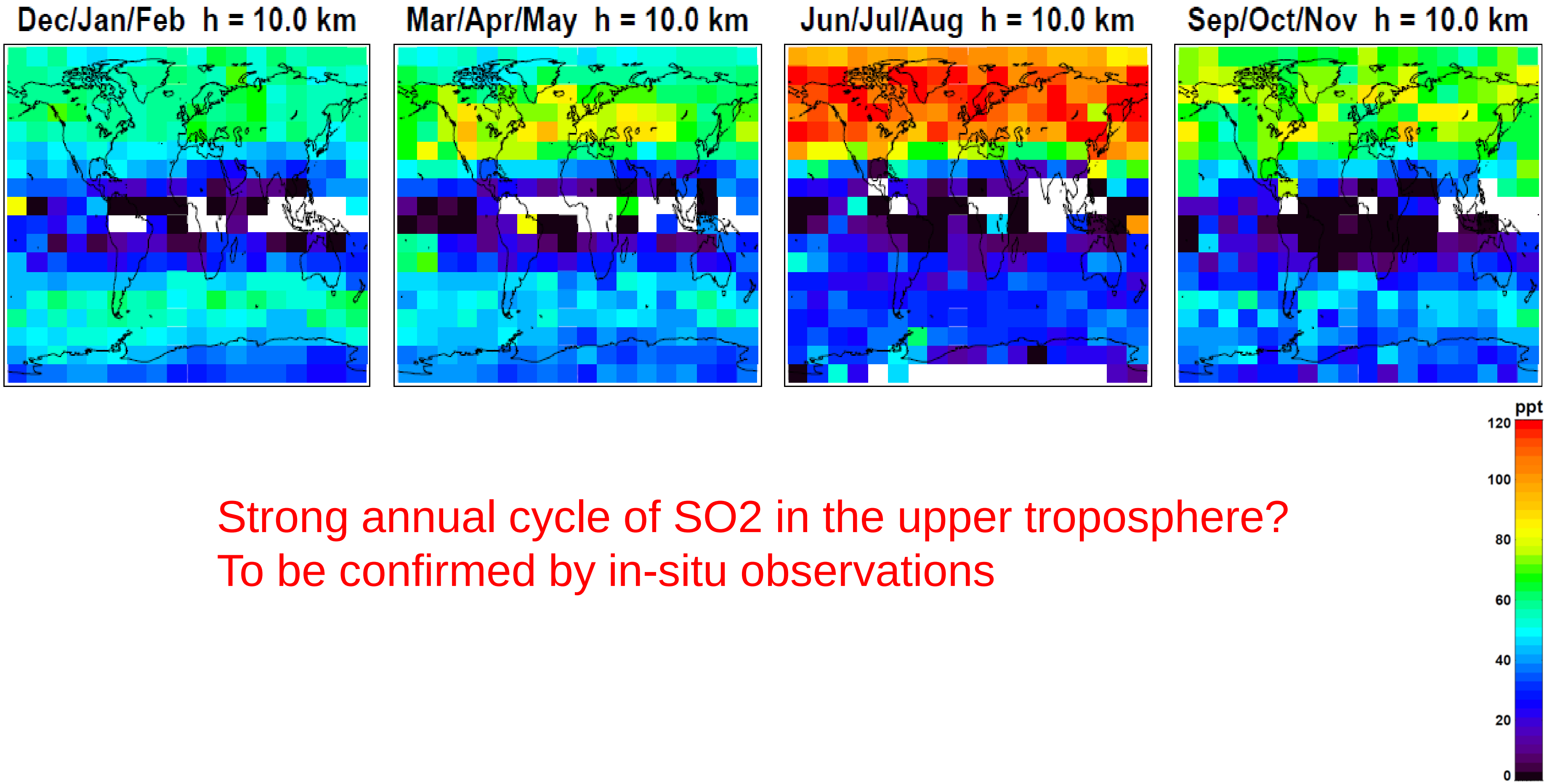




Nadir sounding instruments seem to strongly underestimate the lifetime of SO_2 in the UTLS:

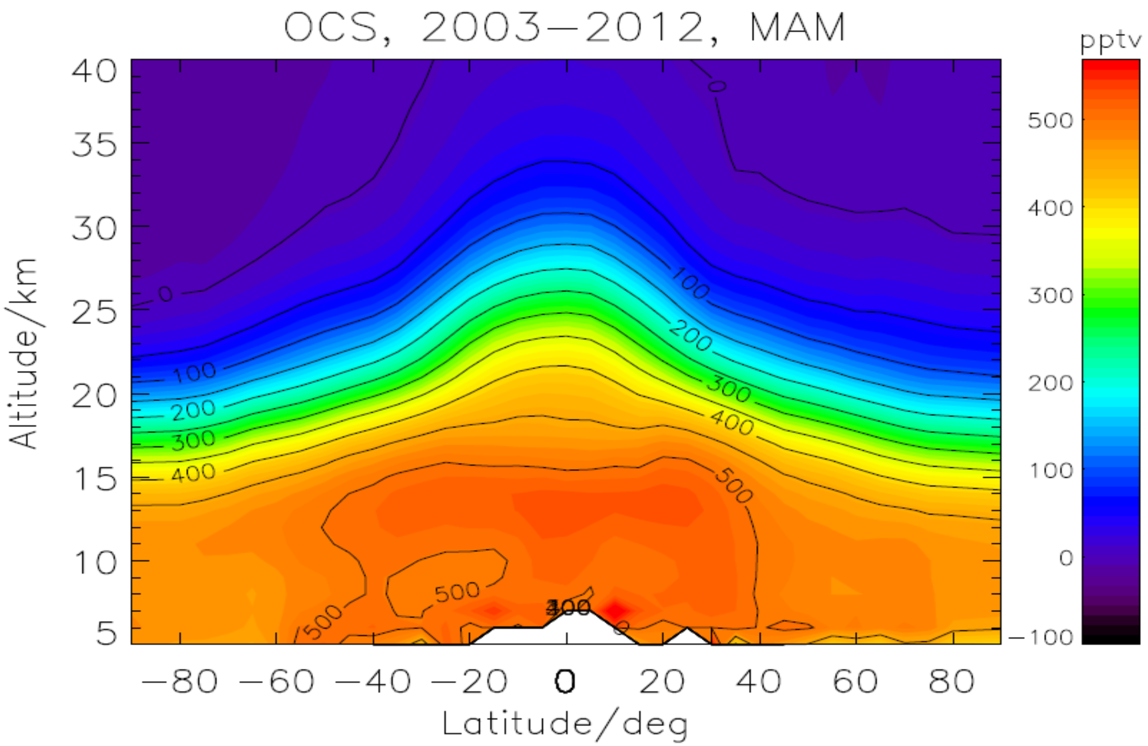
- **Detection-limit of nadir sounders (global dillution of SO_2, Haywood et al., 2010) ?**
- **Combination of lower SO_2-lifetime at lower altitudes and nadir averaging kernels?**

Non-volcanic aerosol background



Strong annual cycle of SO2 in the upper troposphere?
To be confirmed by in-situ observations

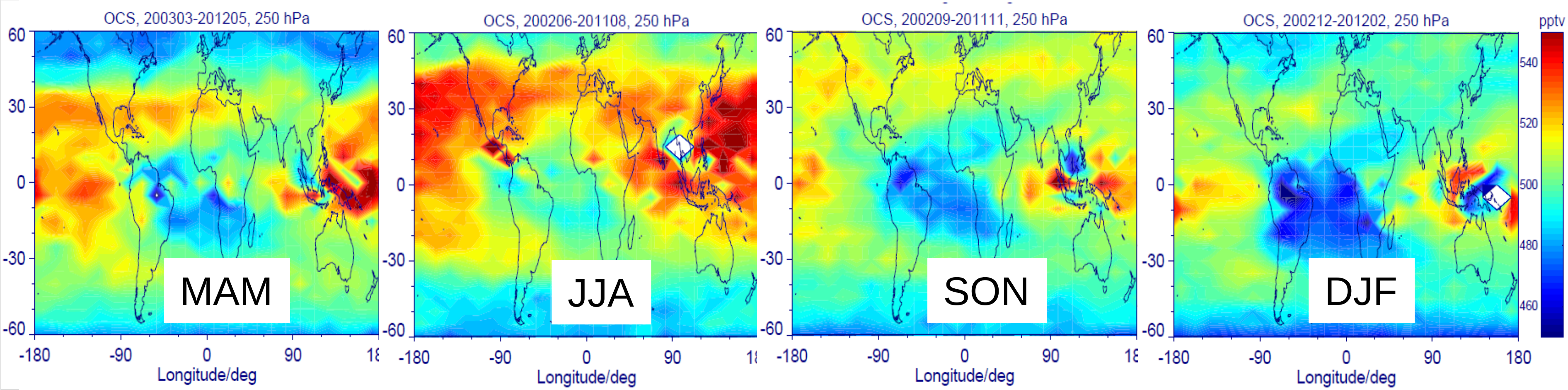
OCS from single MIPAS limb-scans



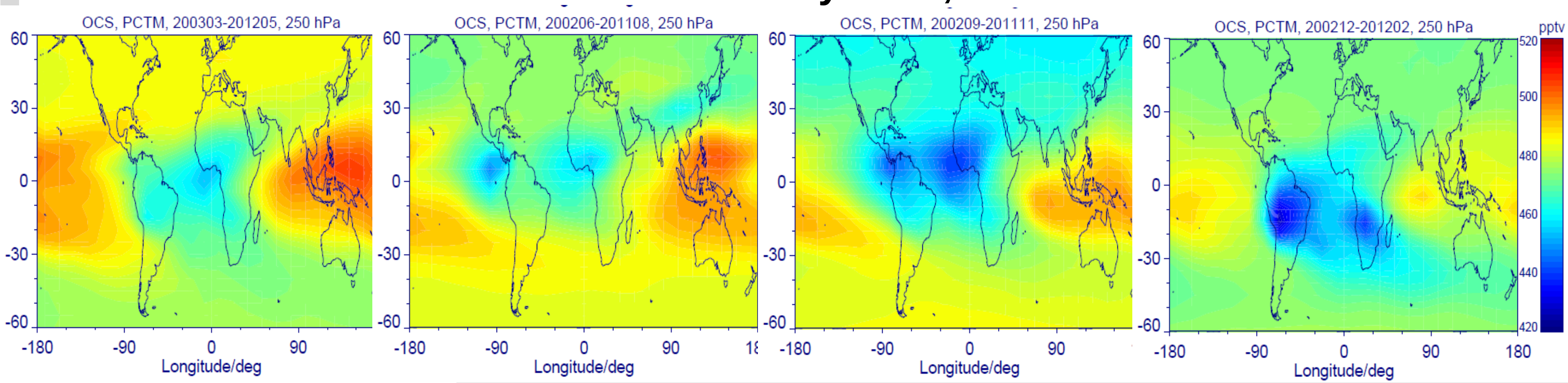
Glatthor et al., subm. 2015

First global distributions of OCS: tropical sink

MIPAS measurements

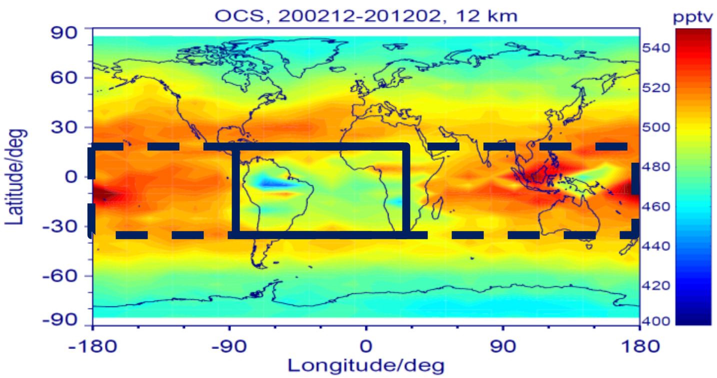


Model: Berry et al., 2013

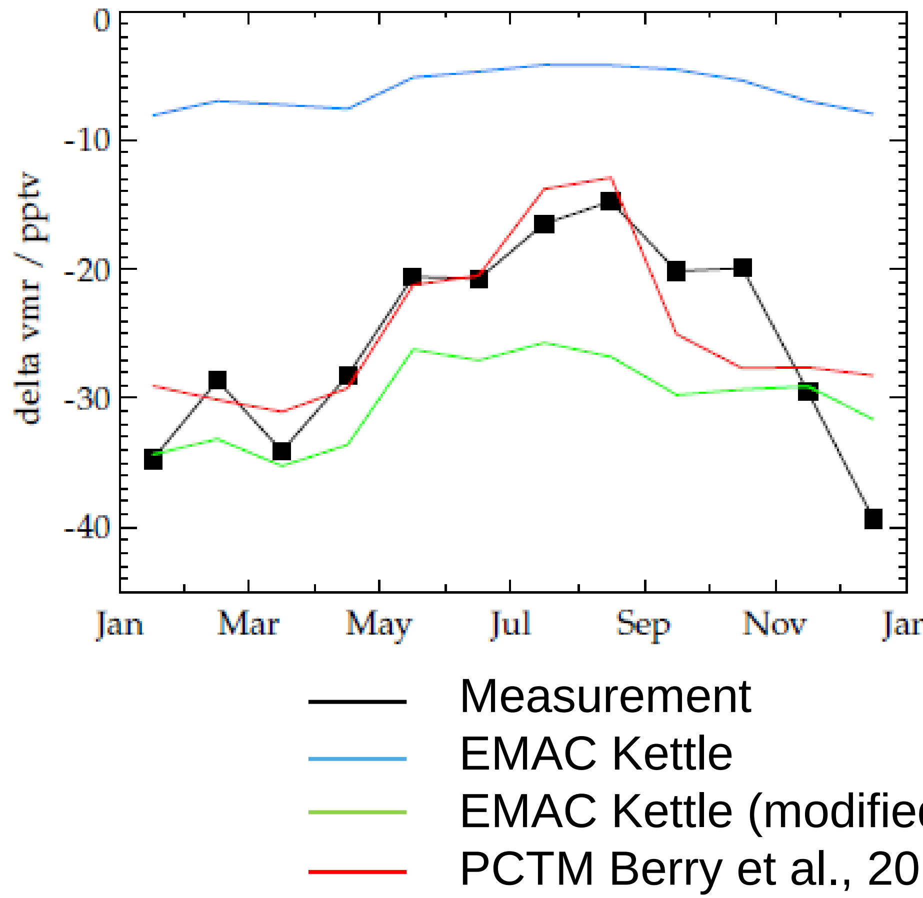


Glatthor et al., subm. 2015

OCS-hole annual cycle: model comparison



Difference: Box – Whole lat. Band



Signal of biomass uptake over S-America: observations can only be reproduced by global models, when a much larger vegetation uptake and a corresponding increase in oceanic emissions than in earlier estimates is considered

Glatthor et al., subm. 2015

Summary

MIPAS SO_2

- Stratospheric maximum @26-30 km
- Downwelling during polar winter and springtime depletion of SO_2 as explanation for the polar aerosol bursts
- Visible and near-IR photolysis of H_2SO_4 and irreversible sink of sulfur
- Height-resolved SO_2 masses and lifetimes for ~30 volcanic eruptions reaching stratospheric levels
- Nadir instruments seem to underestimate SO_2 lifetimes in the UTLS
- Strong seasonal cycle in the northern hemispheric UT
- Enhanced values in monsoon regions

MIPAS OCS

- First global distributions of OCS including observation of tropical sink: need of much larger vegetation uptake in models
- Strong source over W-Pacific in spring: not well captured by models
- Biomass burning cannot be identified as a strong source of OCS