CARBONYL SULFIDE AND ‘AGE OF AIR’

A NOVEL APPROACH TO LOOK AT STRATOSPHERIC DISTRIBUTIONS

28 APR 2021 | CHENXI QIU, FELIX PLOEGER, JENS-UWE GROOß, AND MARC VON HOBE

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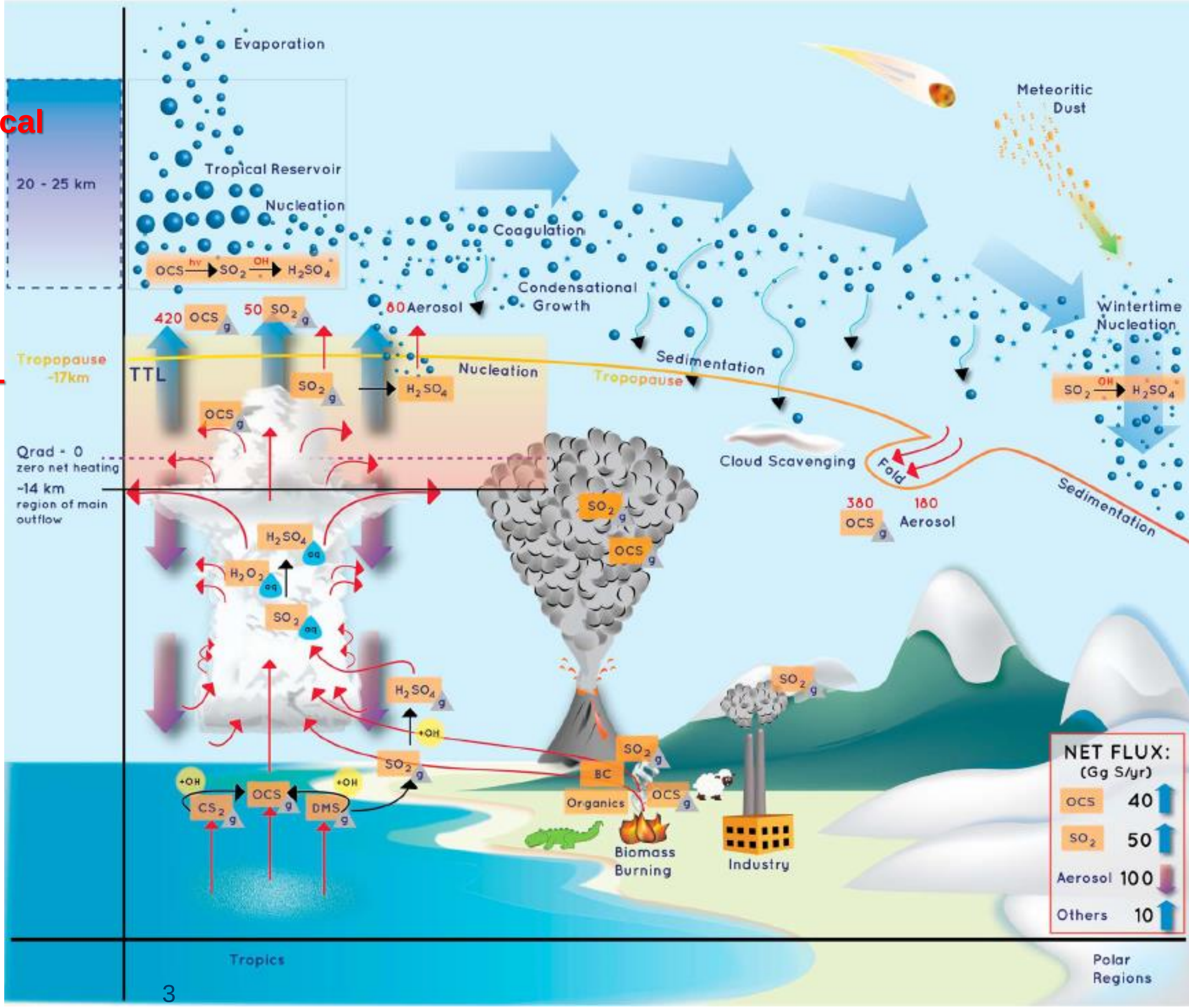
WHY OCS?

- the **longest lived** and the **most abundant** reduced sulfur gas
- very stable in the troposphere, OCS can **reach the stratosphere with little loss**.
- its transport to the stratosphere maintains the **stratospheric aerosol layer** (which has a net cooling effect) in times of volcanic quiescence.
- no significant trend in the atmosphere for the last decades, but **current budget not closed yet**.

Tropical pipe

TTL

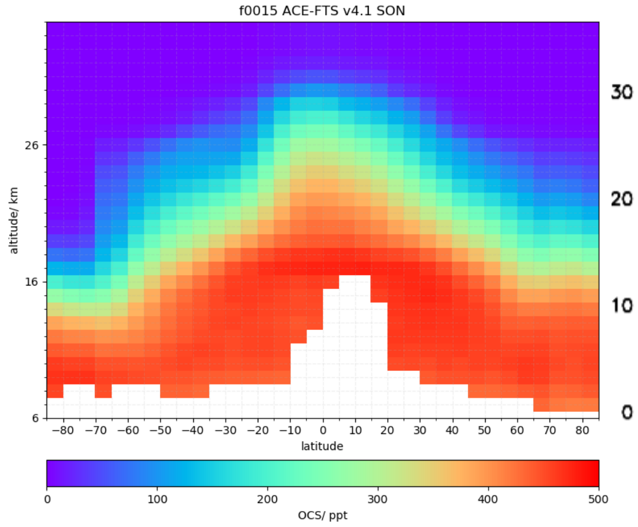


Kremser et al., 2015

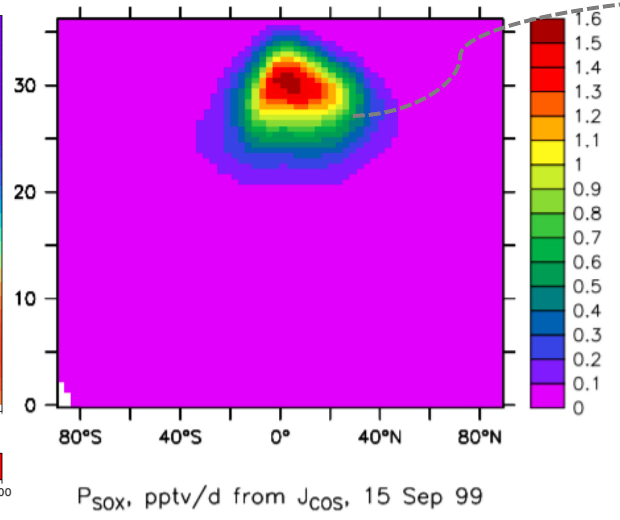
QUESTIONS ABOUT OCS

- How and where is OCS removed in the stratosphere?
- How does the transport of OCS depleted air affect the observed distributions?

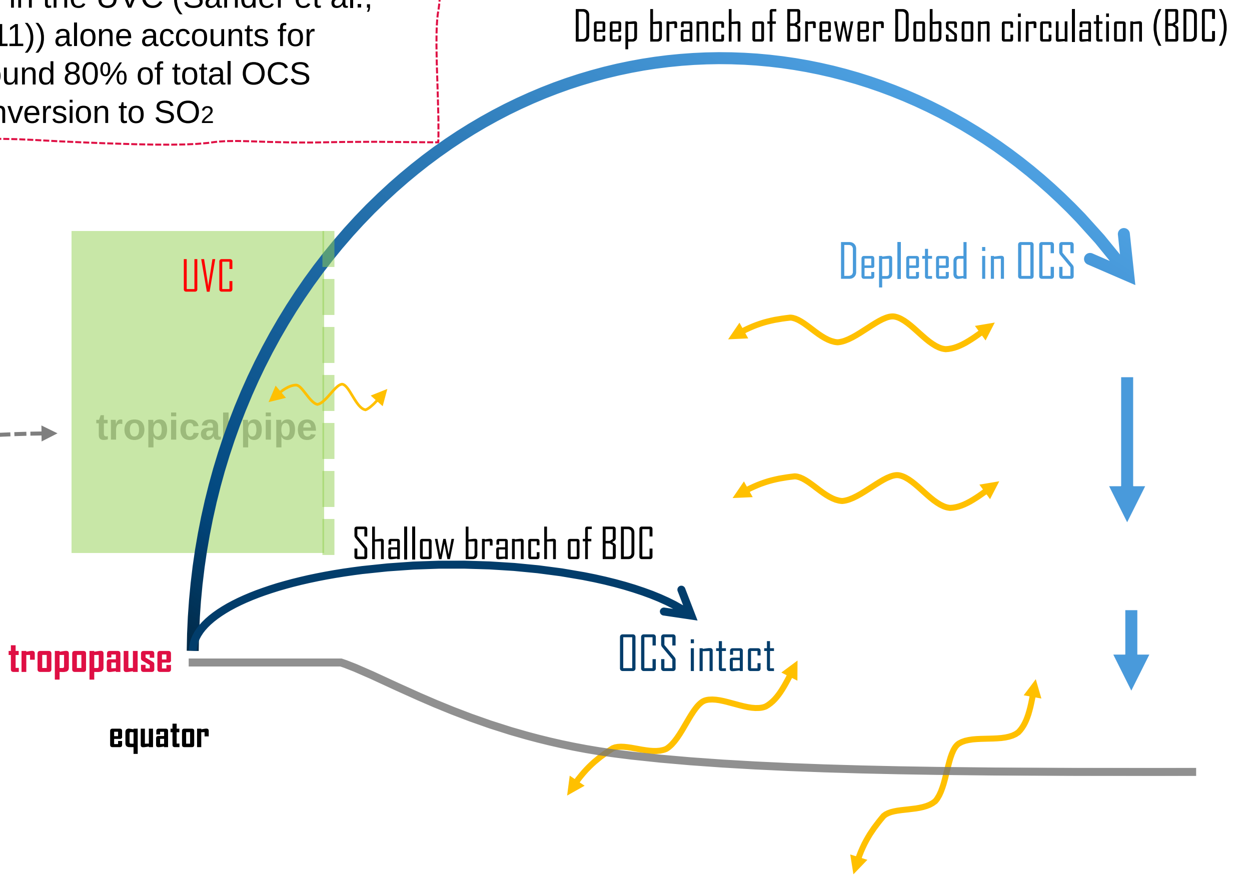
Sheng et al. (2015) reported that photodissociation (peak at 222 nm in the UVC (Sander et al., 2011)) alone accounts for around 80% of total OCS conversion to SO_2



Mean OCS of September, October and November from satellite-based observation



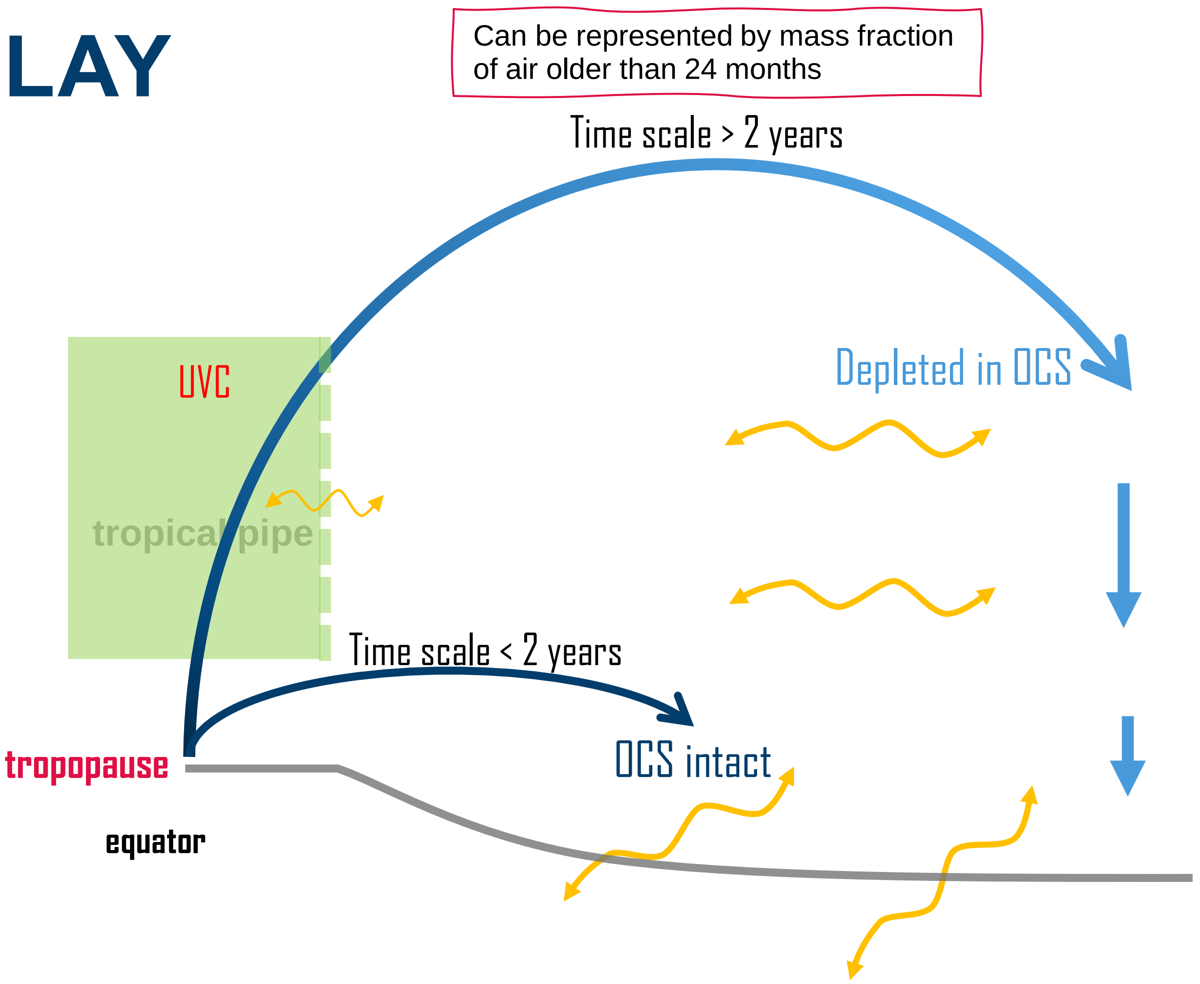
Modeled SOx production from COS, pptv/day (Brühl et al., 2012)



Through the ‘tropical pipe’, OCS is rapidly converted.

‘AGE OF AIR’ COMES INTO PLAY

- **Deep branch of the Brewer Dobson circulation(BDC)** that goes through the ‘tropical pipe’ transports air to the middle atmosphere and the air then descends at high latitudes with time scales of at least **2 years**.
- **Shallow branch of the BDC** transports air to slightly above tropopause and the air descends at mid-latitudes with time scales of **a few months up to 2 years**.
- Therefore, we can evaluate the role of the ‘tropical pipe’ on OCS by studying the relationship between OCS and age spectra.

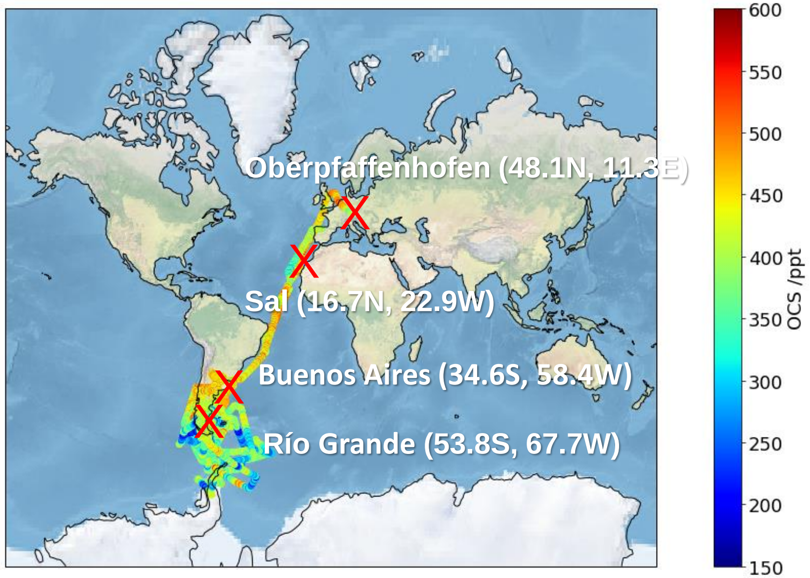


Kremser et al., 2015

IN-SITU OBSERVATIONS: SOUTHTRAC MISSION

Category	Range
Time Period	September – November 2019
# of flights with OCS measurements	22
Latitude	70.3S – 55.7N
Longitude	84W – 11E
OCS	133 – 666 ppt
Polar vortex contribution*	0 – 90%
Mean age of air*	0 – 38 months

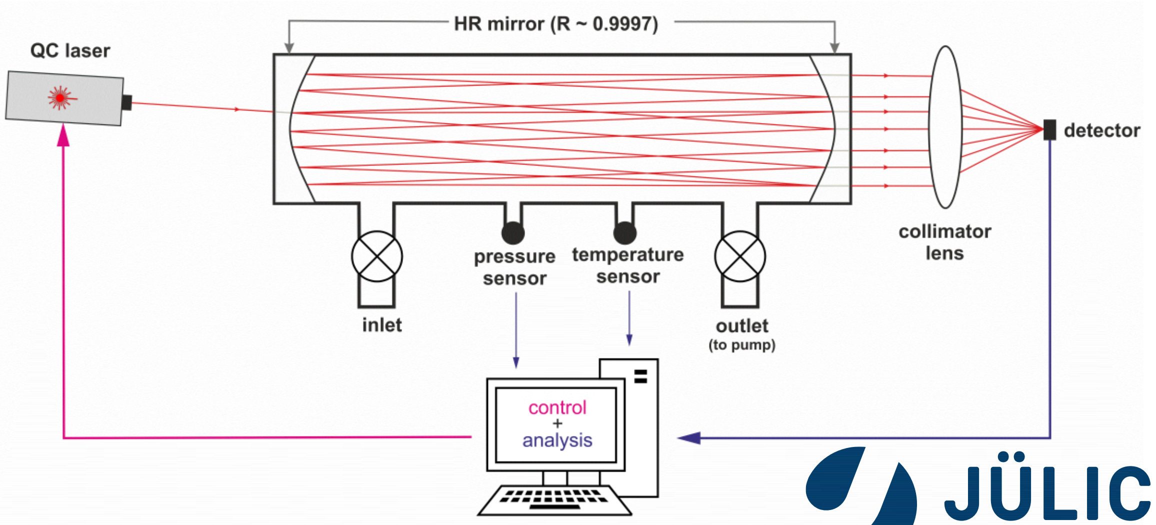
*: From CLaMS model run



Instrument:

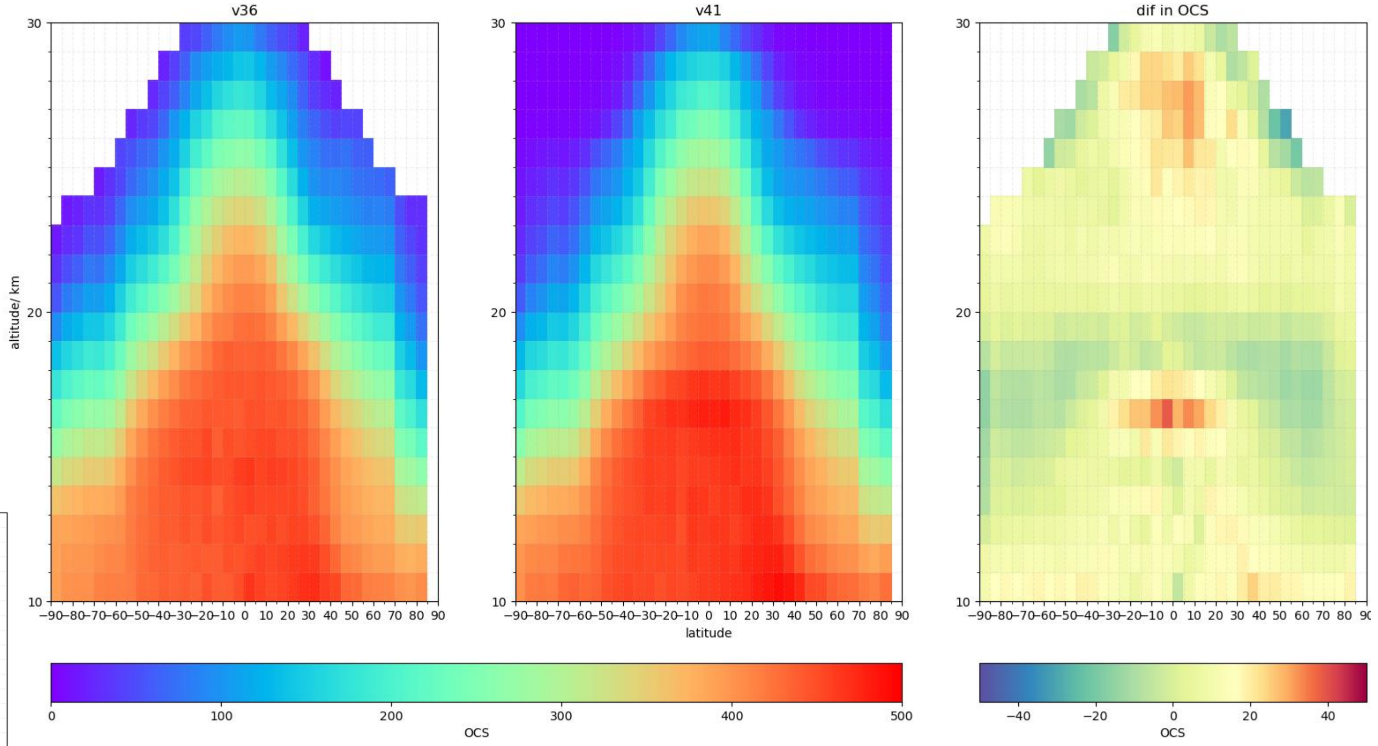
AMICA: Airborne **M**id-**I**nfrared **C**avity enhanced **A**bsorption spectrometer

- Two-cavity design to measure OCS, CO, CO_2, O_3
- AMICA adopts Off-Axis Integrated Cavity Output Spectroscopy (ICOS) →

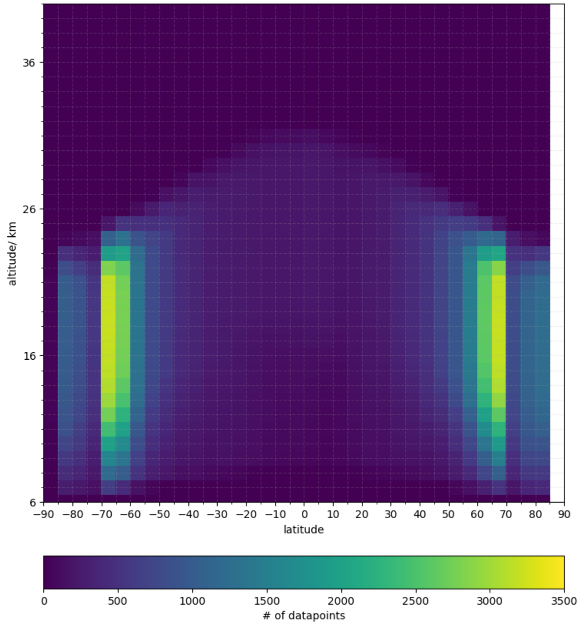


SATELLITE OBSERVATIONS

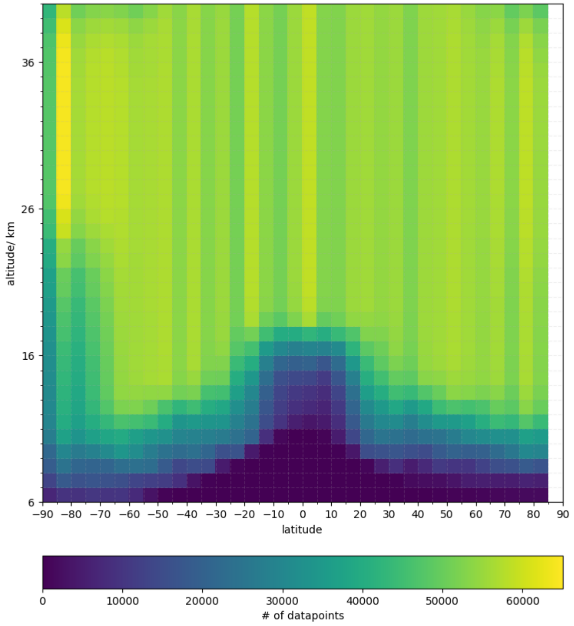
- **MIPAS/Envisat** (Barkley et al., 2008)
- **ACE-FTS** (Glathor et al., 2017)



ACE-FTS: solar occultation



MIPAS: limb emission



Data density between 2004-2012 shows the scarce sampling of ACE-FTS (due to solar occultation viewing geometry)

We've found significant difference between different older and newer versions of ACE-FTS OCS data. Currently data version 3.6 was used for analysis. Later we'll switch to version 4.1.

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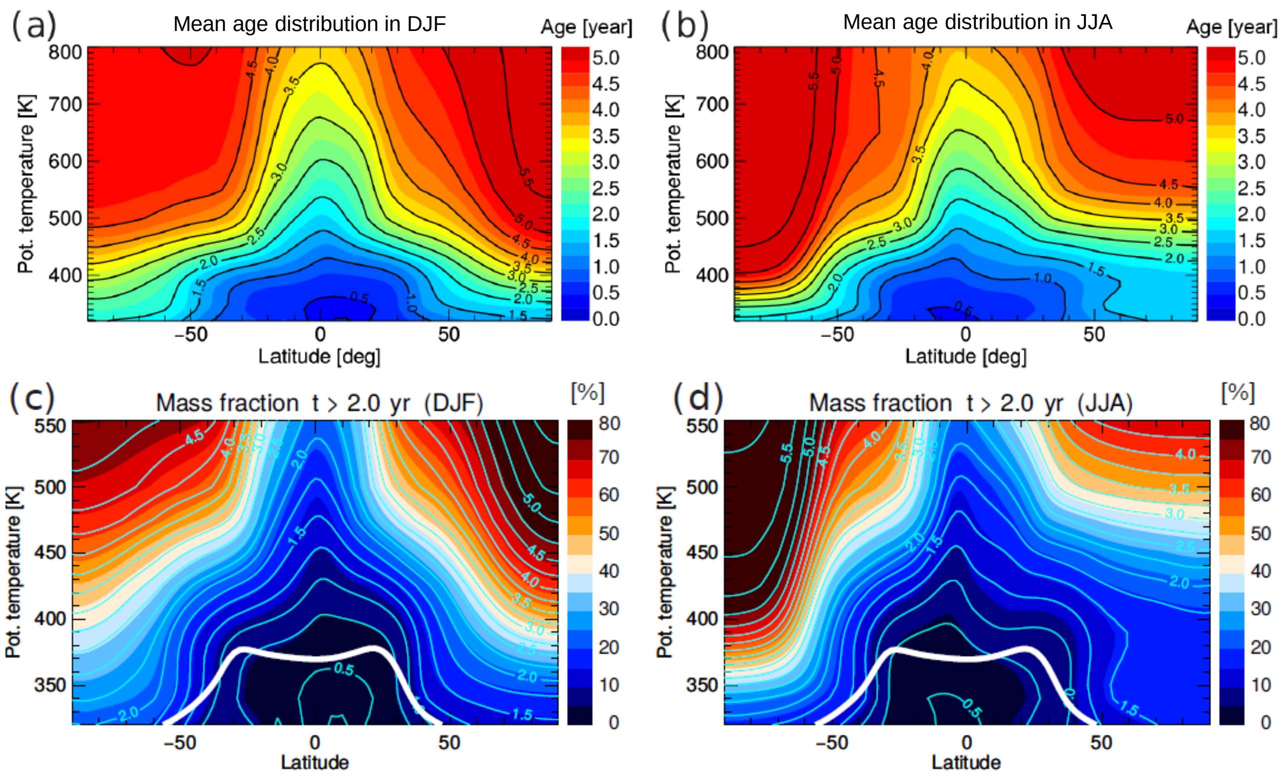
'AGE OF AIR'

- **CLaMS model**

Chemical **La**grangian **M**odel of the **S**tratosphere



Ploeger & Birner, 2016

- the climatological 'age of air' was found to significantly depends on the reanalysis (Ploeger et al., 2019)
- Our simulation is from CLaMS driven by **ERA-interim** reanalysis (future comparison with that of ERA-5 possible)

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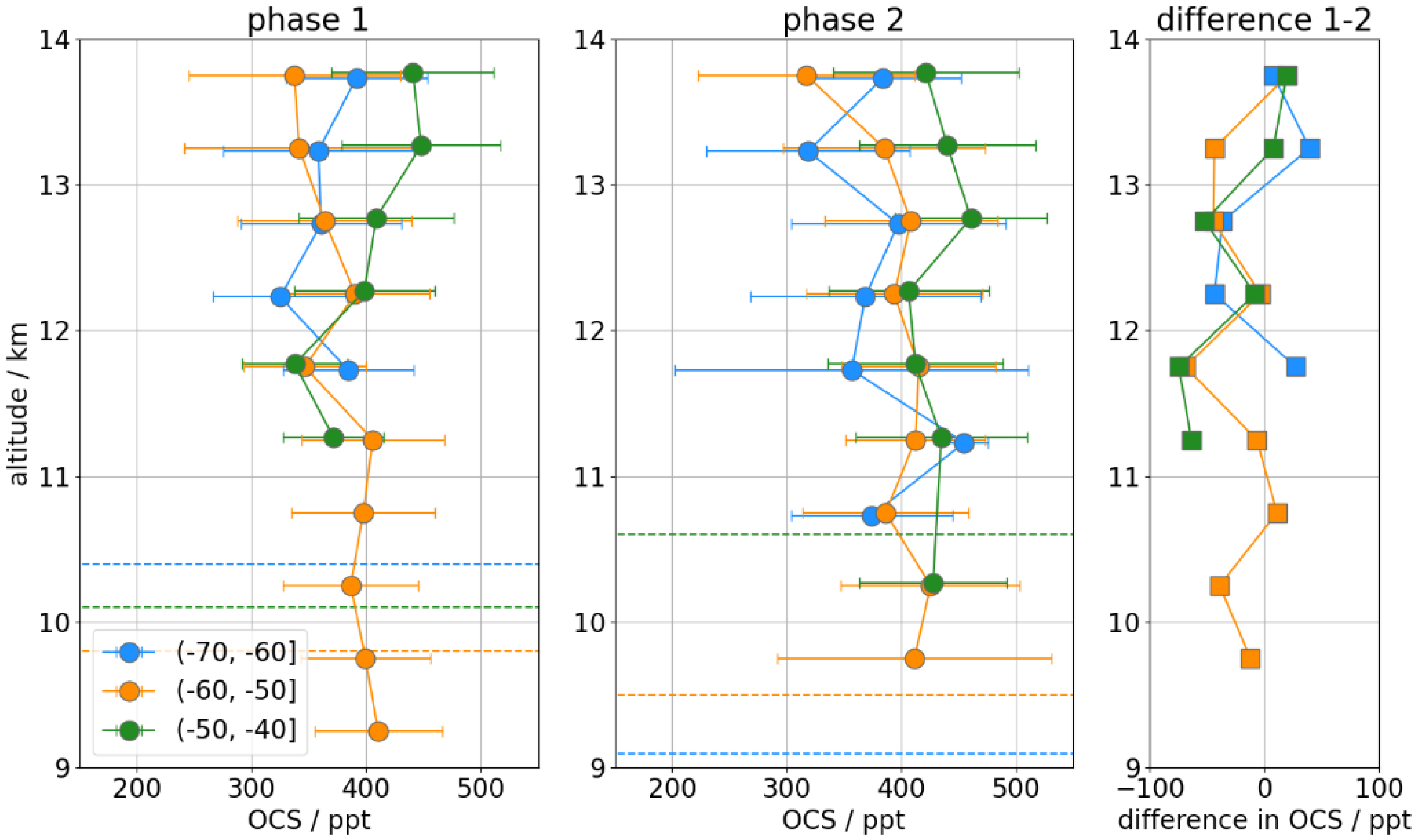
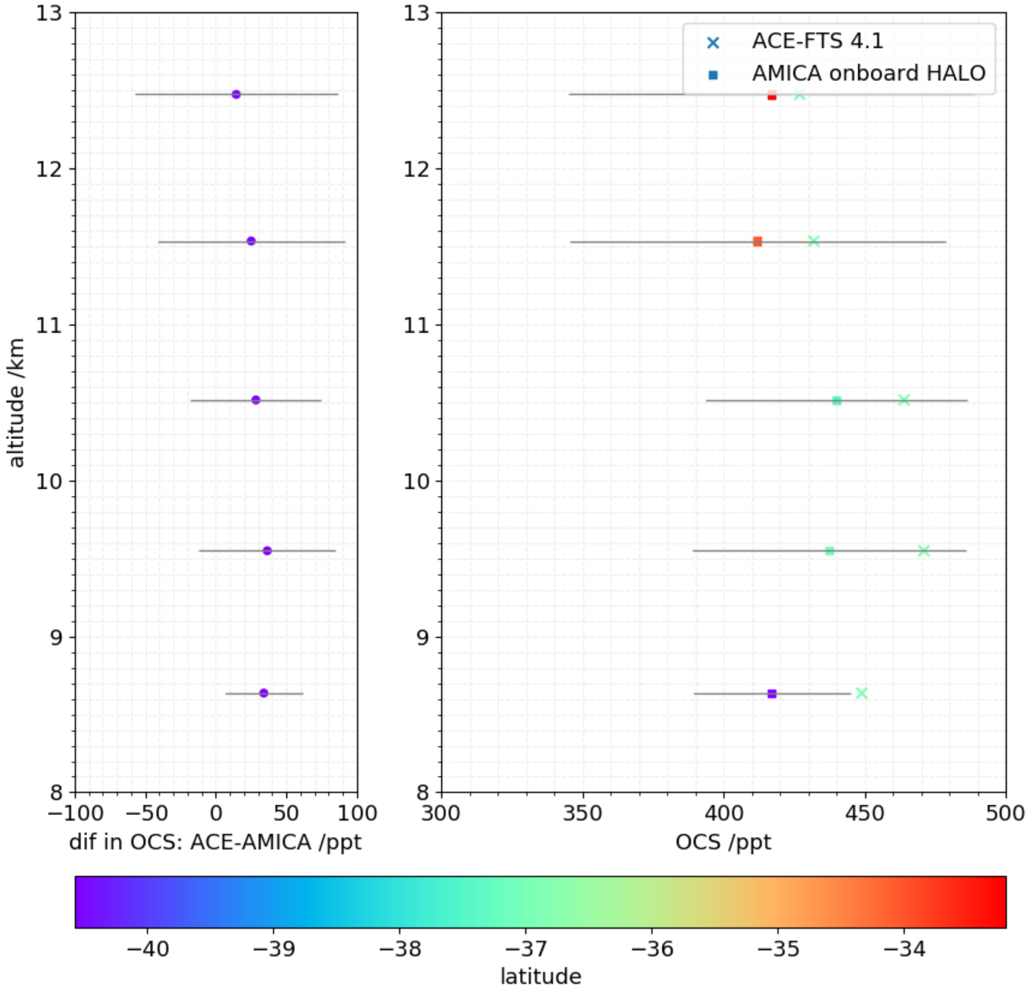
CHARACTERIZATION OF OCS FROM SOUTHTRAC



- Comparison of in-situ SouthTRAC OCS data obtained from local flights between phase 1 (September) and phase 2 (November)
- OCS in phase 2 tends to be at least as high than as that of phase 1, with OCS in phase 2 up to around 75 ppt higher between 11.5 km and 12 km in altitude and 40S and 50S.

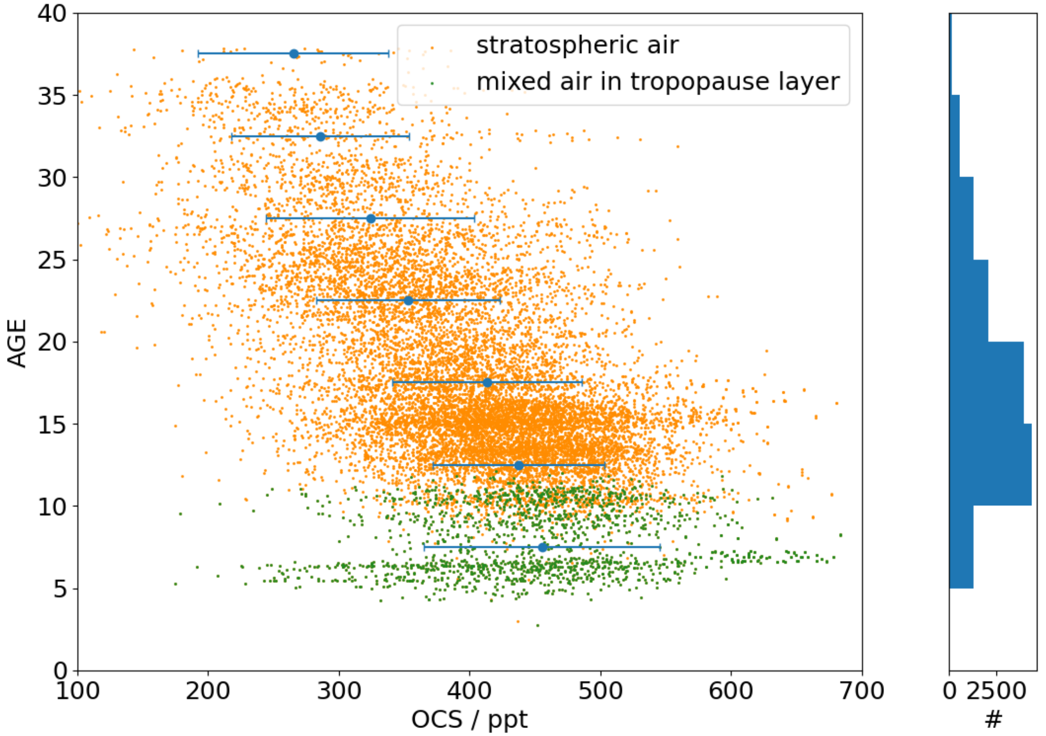
As the data are spread out in latitudes, we assemble zonal mean OCS for three different latitude ranges that cover the whole dataset . The right plot shows the difference in OCS between phase 1 and phase 2, with positive value meaning that phase 1 has higher OCS than phase 2 and vice versa. The dotted lines show the typical tropopause heights in the given latitudinal ranges and time of year (Rieckh, 2014). One standard deviation uncertainties are shown for the profiles. Bins with fewer than 3 measurements are excluded. Data with latitudes between 60S to 70S (blue) and between 35S to 50S (green) are shifted vertically by ± 0.02 km for readability.

COMPARISON OF ACE-FTS AND AMICA OBSERVATIONS



- In-situ observations by AMICA during transfer flight from Buenos Aires to Sal on 7 October 2019 in Southern hemisphere mid-latitude are co-located within 500km horizontal radius and ±6 h time offset with 5 ACE-FTS vertical profiles
- ACE-FTS overestimates OCS compared to AMICA, with magnitude from around 5 ppt to around 40 ppt, but ACE-FTS observations are still in one standard deviation of the AMICA data in the same range

AMICA OCS & MEAN AGE



OCS mixing ratio and mean age measured during SouthTRAC when abs(PV)>2 or theta>380K. The horizontal bar plots on the right of each sub-figure show the total number of datapoints for each 'age of air' bin. Dots represent each measurements and blue error bars represents mean and standard deviation in each bin. 'Stratospheric air' (orange) represents air parcels with absolute value of potential vorticity higher than 4 PVU. 'Mixed air in the tropopause layer' (green) represents air with absolute value of potential vorticity between 2 and 4 PVU. One standard deviation uncertainties are shown for all datapoints at each bin of mean age .

- Individual datapoints of modelled mean age are plotted against OCS observations during SouthTRAC. Measurements with air of absolute value of potential vorticity between 2 to 4 PVU (green in Fig. 23) and higher than 4 PVU (orange in Fig. 23) are presented separately.
- Anti-correlation between OCS and mean age as expected.
- Most datapoints with mean age younger than around one year are mixed air and they show larger variability in OCS.
- The lower OCS mixing ratios observed in the mixed air down to around 200 ppt were mostly measured on the transfer flight from Oberpfaffenhofen (48.1N) to Sal (16.7N) in early November.
- The higher OCS mixing ratios observed in the mixed air up to around 700 ppt were mostly measured during the transfer flight from Sal to Oberpfaffenhofen in early October.

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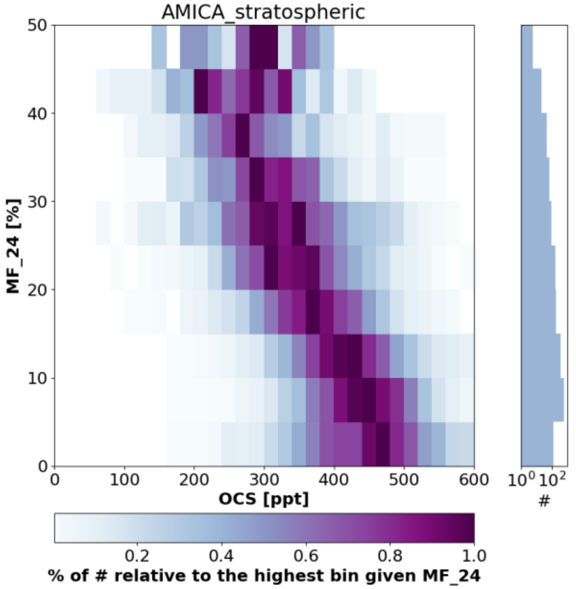
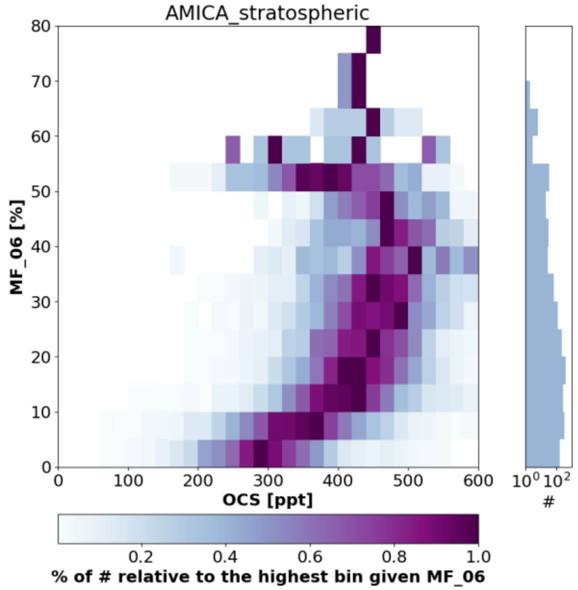
AMICA
OCS & AGE SPECTRA

- Correlation of OCS with MF<6 months and anti-correlation with MF>24 months, medium age and mean age.
- For air with no fraction older than 24 months, OCS are cantered around 450 ppt to 500 ppt. With MF 24m increasing from 0 to around 40%, which is the range where most datapoints are, OCS decreases 'monotonically' to around 200 ppt.
- With MF 06m increasing from 0 to around 40%, which is also the range where most datapoints are, OCS increases 'monotonically' from around 300 ppt to around 500 ppt.
- Medium age is up to 15-20 months, but mean age is up to 40 months due to the small fraction of old air bringing it up.
- For mean age and medium age, OCS decreases from around 450 ppt - 460 ppt to around 300 ppt with mean age being 25 - 30 months or medium age being 15-20 months.

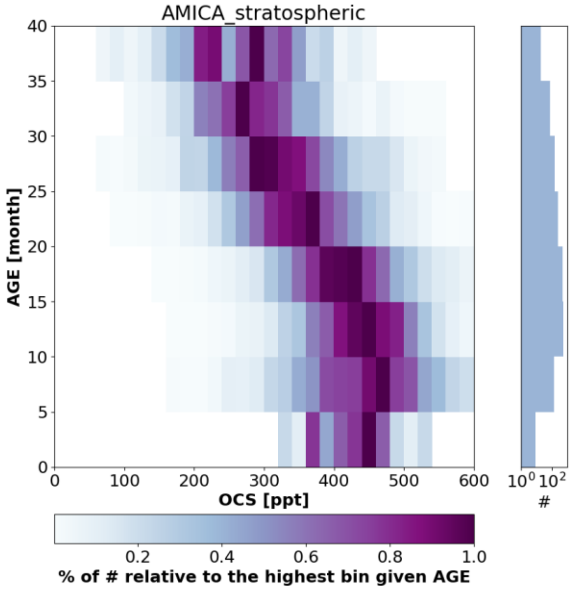
MF=mass fraction of air, **histogram on the left**: the total number of datapoints at each the age parameter level in log scale, **histogram at the bottom**: color bar for the relative number of datapoints in each bin compared to the bin of the same level but with the most datapoints



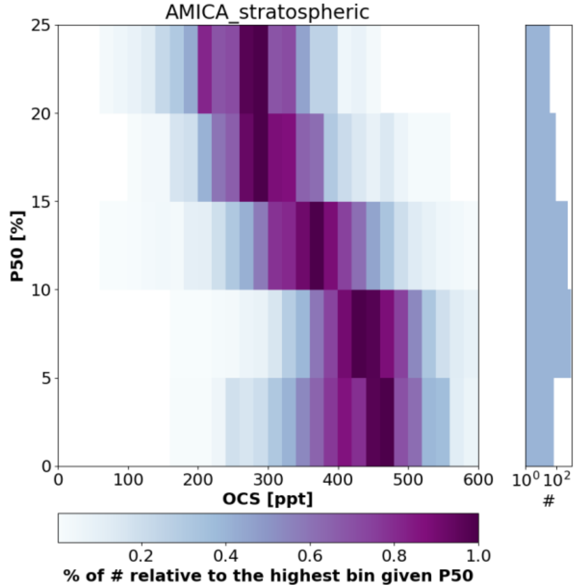
MF>24 months



MF<06 months



Mean age

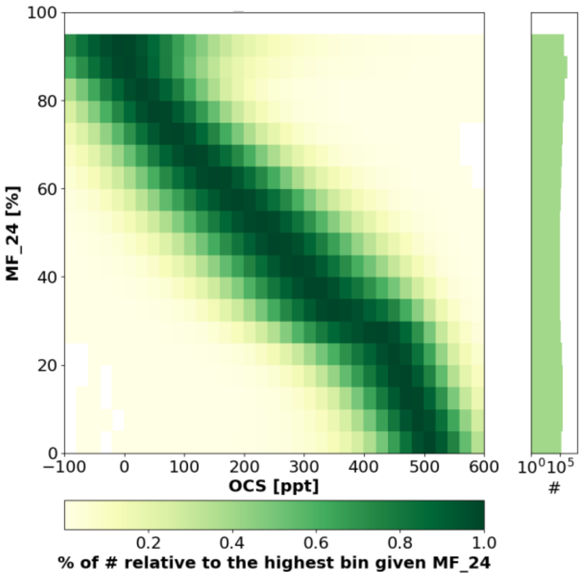


Median age

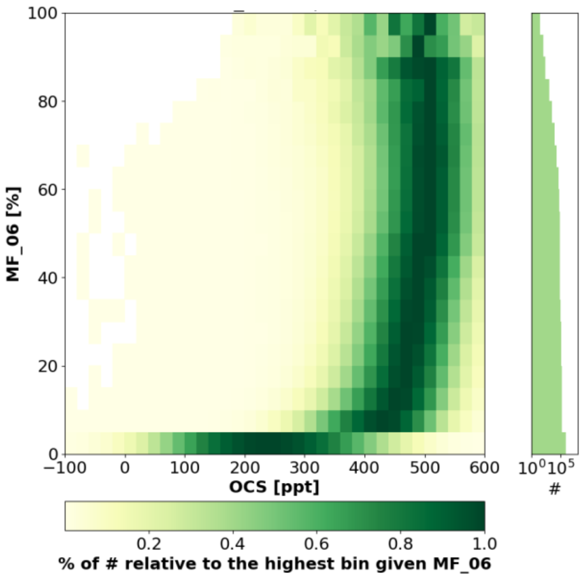
MF=mass fraction of air, **histogram on the left**: the total number of datapoints at each the age parameter level in log scale, **histogram at the bottom**: color bar for the relative number of datapoints in each bin compared to the bin of the same level but with the most datapoints

MIPAS
OCS & AGE SPECTRA

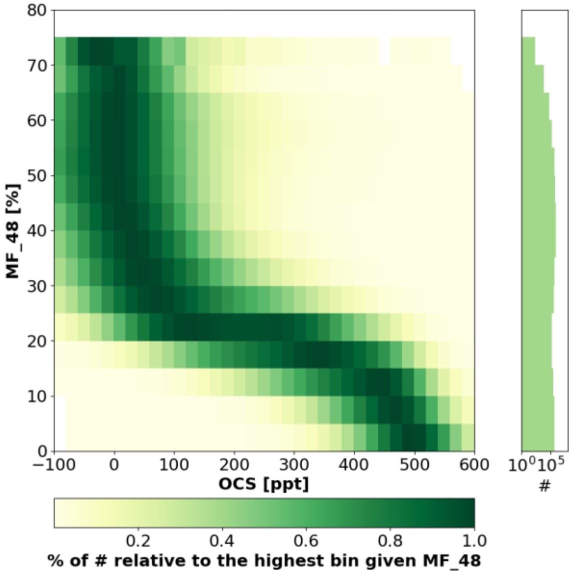
- Correlation of OCS with MF<6 months, MF<3 months and anti-correlation with MF>48 months, MF>24 months, MF>12 months and mean age.
- Strong monotonically decreasing OCS with increasing mass fraction of air older than 2 years
- Curved anti-correlation between OCS and mean age with OCS first decreasing slowly to approx. 20 months and increasingly fast towards below 20 ppt at 50 months.



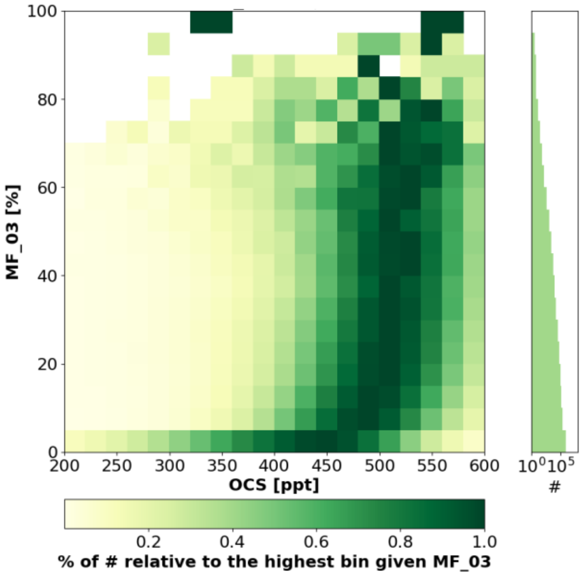
MF>24 months



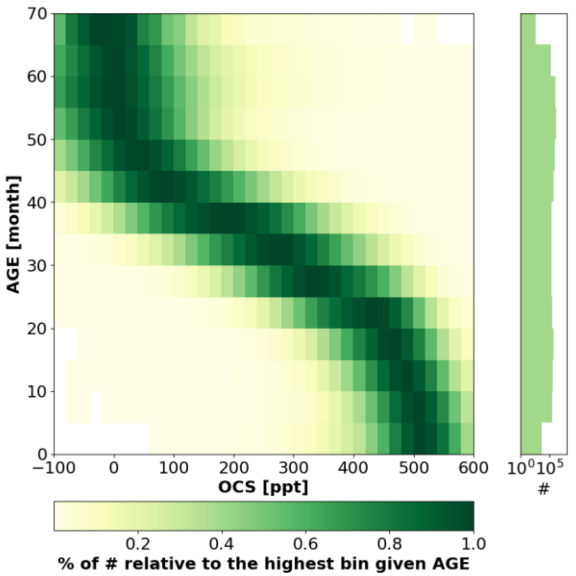
MF<06 months



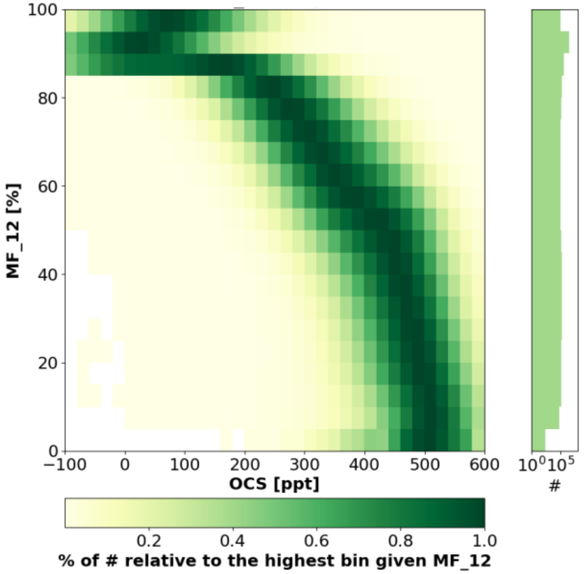
MF>48 months



MF<03 months



Mean age

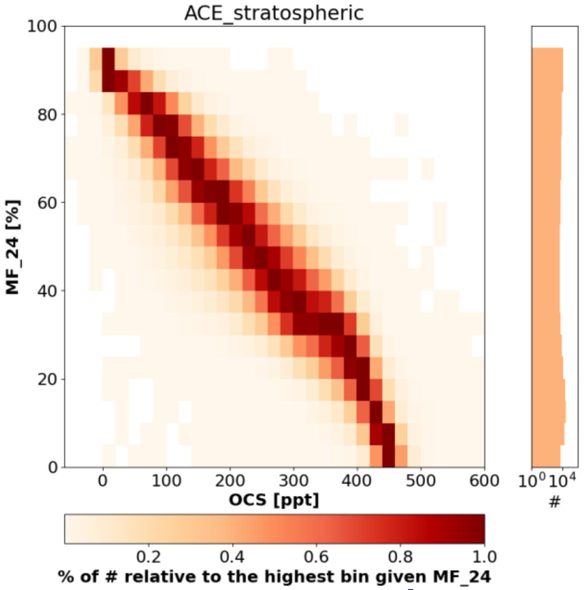


MF>12 months

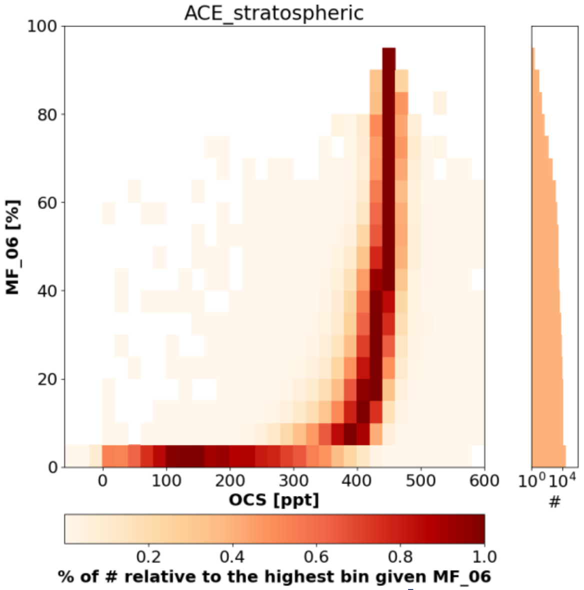
ACE-FTS
OCS & AGE SPECTRA

- Smaller spread of data compared to MIPAS
- Same (anti-) correlation of OCS with the age parameters
- Strong monotonically decreasing OCS with increasing mass fraction of air older than 2 years, as is also seen in MIPAS
- Similarly, OCS first decreases slowly with increasing mean age to approx. 20 months and then decreases increasingly fast towards below 20 ppt at 70 months.

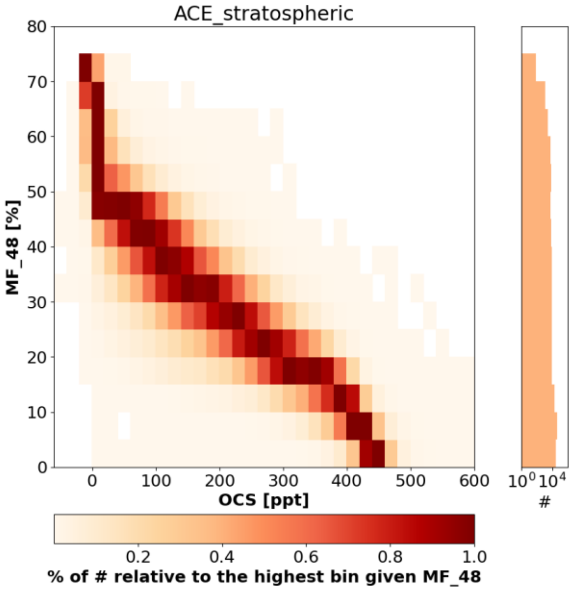
MF=mass fraction of air, **histogram on the left**: the total number of datapoints at each the age parameter level in log scale, **histogram at the bottom**: color bar for the relative number of datapoints in each bin compared to the bin of the same level but with the most datapoints



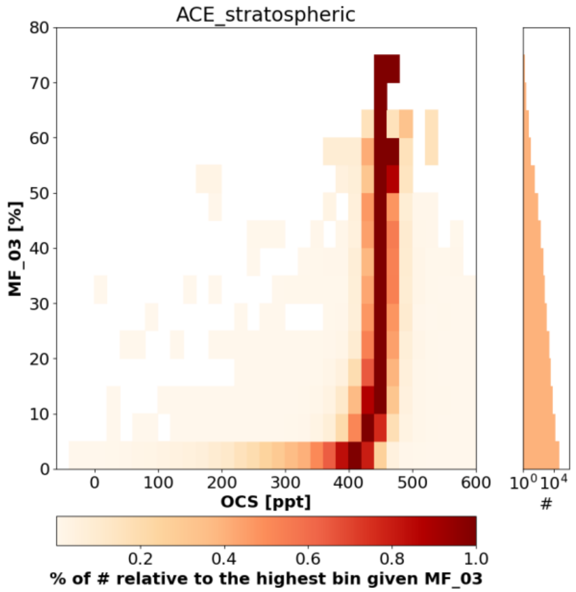
MF>24 months



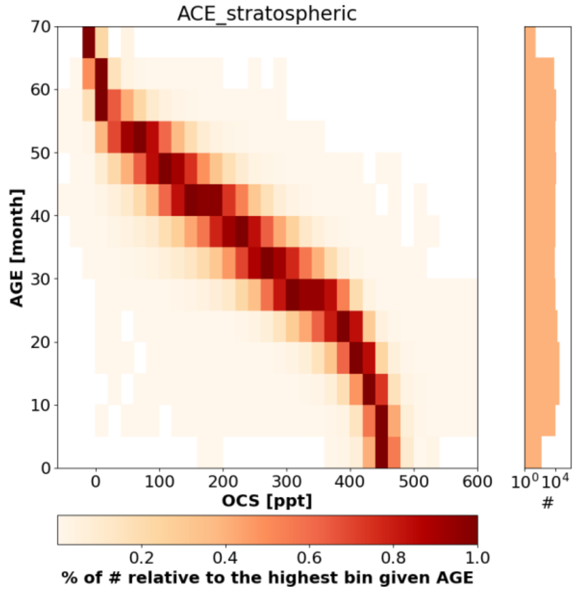
MF<06 months



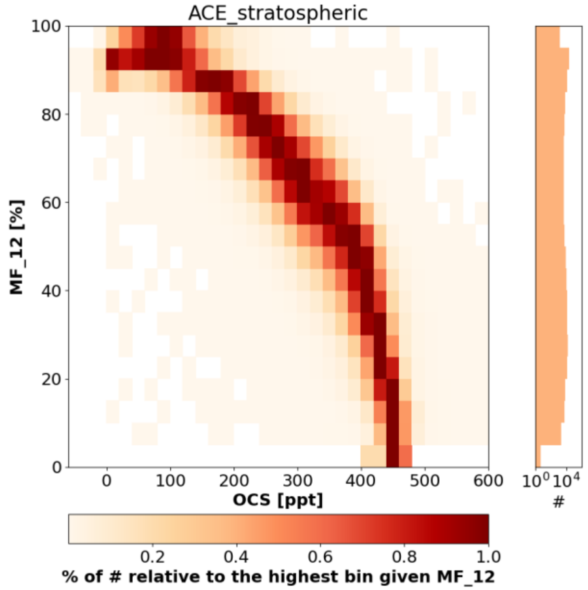
MF>48 months



MF<03 months



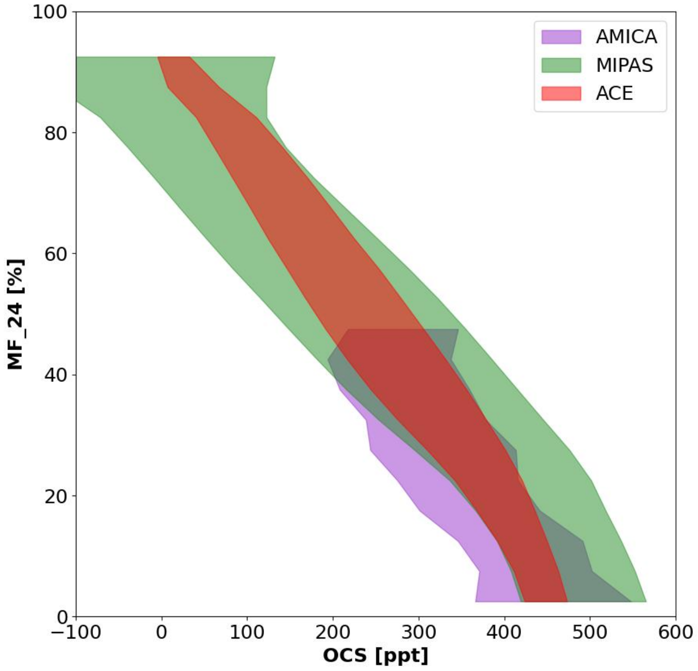
Mean age



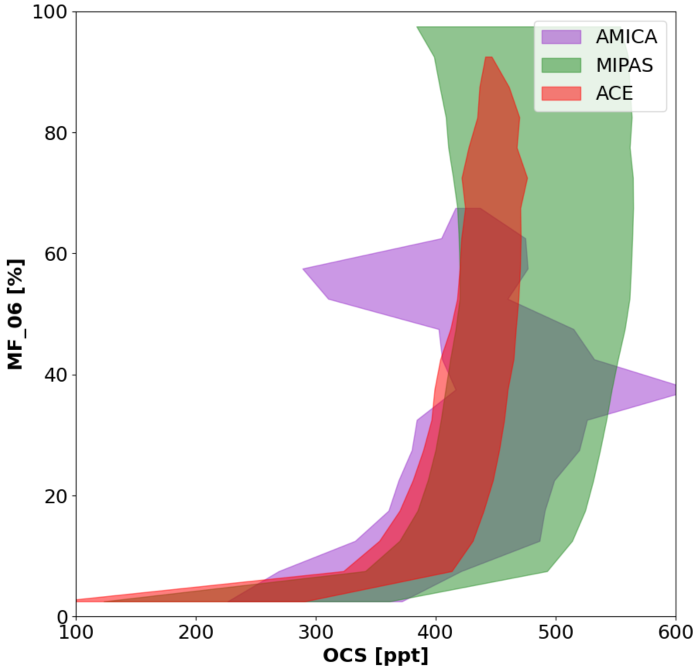
MF>12 months

INTERCOMPARISON

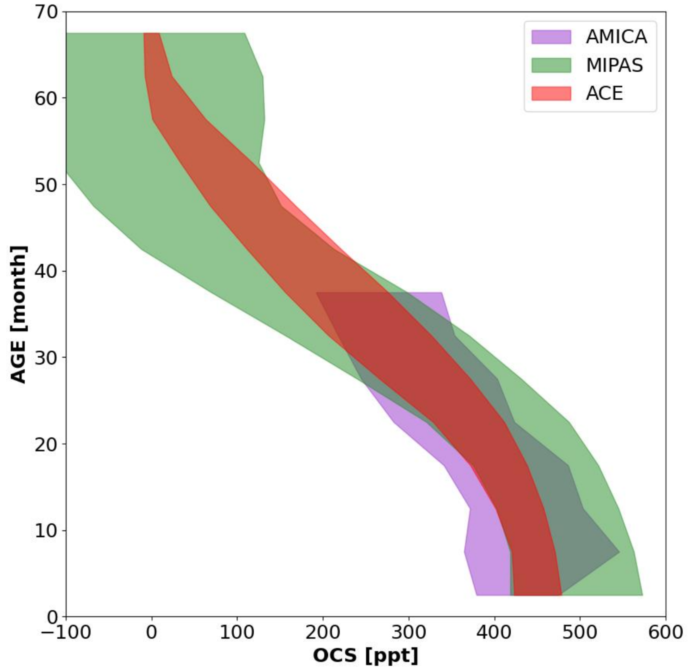
- Robust 'Linear' relationship between mass fraction of air older than 24 months (MF 24m) and OCS.
- With MF 24m increasing from 0 to approx. 93%, OCS decreases from around 450 ppt to approx. 20 ppt.
- The observed relationship is robust among AMICA, ACE-FTS and MIPAS observations.



MF>24 months



MF<06 months



AGE

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CONCLUSION

- A robust anti-correlation between OCS and the mass fraction of air older than 24 months (MF>24m) found, with a quasi-linear relationship that can be represented by:

  [OCS] in stratospheric/mixed air = (1 - MF_24) x [OCS] in tropospheric air

- The observed OCS distribution and its correlation with MF>24m clearly supports our picture that OCS is depleted mainly in the ‘tropical pipe’ region and the OCS depleted air transported through the deep branch of the Brewer Dobson circulation.

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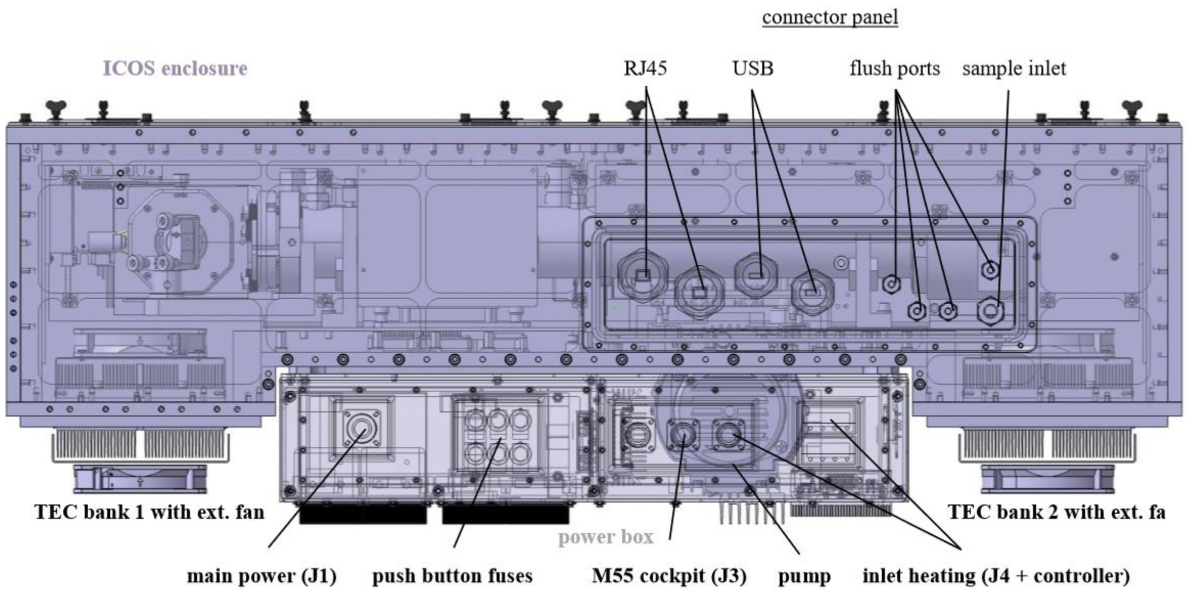
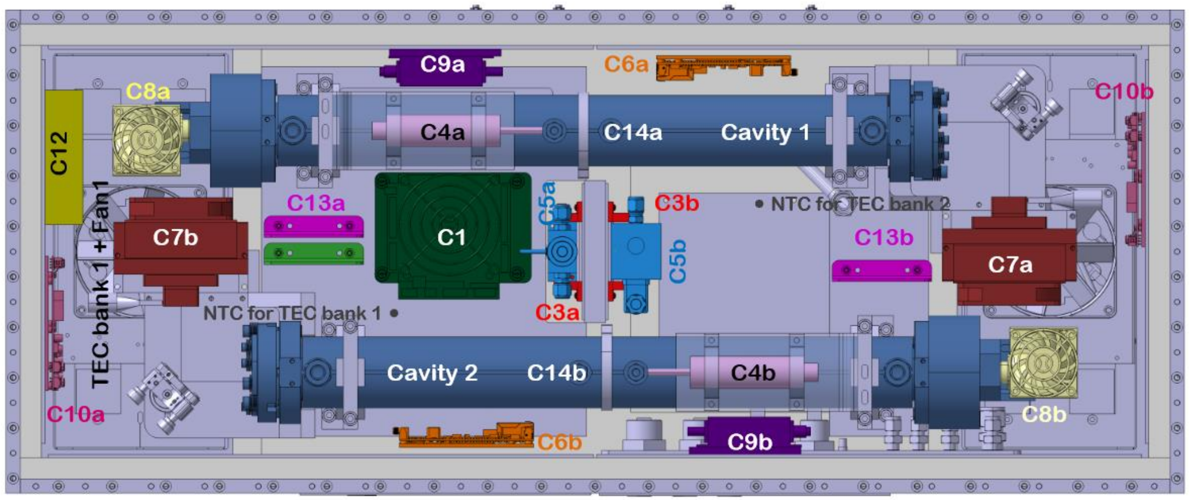
SUPPLEMENTARY MATERIAL

AMICA PERFORMANCE

Flight #	date	Channel 1 OCS,CO,CO_2,H_2O	Channel 2 O_3	remarks
ST1	19 Aug	X	X	netCDF4 files not synchronized to disk
ST2	28 Aug	X	X	File handling issue
ST3	2 Sep	✓	✓	nominal
ST4	6 Sep	✓	✓	nominal
ST5	6 Sep	(✓)	✓	issue with laser driver board temp after > 8 h, ~ 20 % data loss CH1 at beginning
ST6	8 Sep	(✓)	✓	issue with laser driver board temp after > 8 h, ~ 5 % data loss CH1 in last phase
ST7	9 Sep	(✓)	✓	issue with laser driver board temp after > 5 h, ~ 5 % data loss CH1 in mid flight
ST8	11 Sep	✓	(✓)	issue with laser driver board temp, ~ 60 % data loss CH2
ST9	13 Sep	✓	✓	nominal
ST10	16 Sep	✓	✓	nominal
ST11	18 Sep	✓	X	channel 2 did not power up (reason unidentified)
ST12	20 Sep	✓	✓	nominal
ST13	24 Sep	✓	✓	nominal
ST14	26 Sep	✓	✓	nominal
ST15	29 Sep	✓	X	channel 2 netCDF file corrupted
ST16	30 Sep	✓	✓	nominal
ST17	2 Oct	✓	✓	nominal
ST18	6 Oct	✓	✓	nominal
ST19	7 Oct	✓	✓	nominal
ST20	9 Oct	✓	✓	nominal
ST21	2 Nov	(✓)	(✓)	Software hang-up; started working after power cycle ~ 2 hours into flight.
ST22	4 Nov	✓	✓	nominal
ST23	6 Nov	✓	✓	nominal
ST24	9 Nov	✓	✓	nominal
ST25	12 Nov	✓	✓	nominal
ST26	15 Nov	✓	✓	nominal
ST27/8	16 Nov	✓	✓	nominal

AMICA ICOS ARRANGEMENT





Kloss et al., 2021, AMTD