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Improved global distributions of SF6 and mean age of stratospheric air by use of new spectroscopic data

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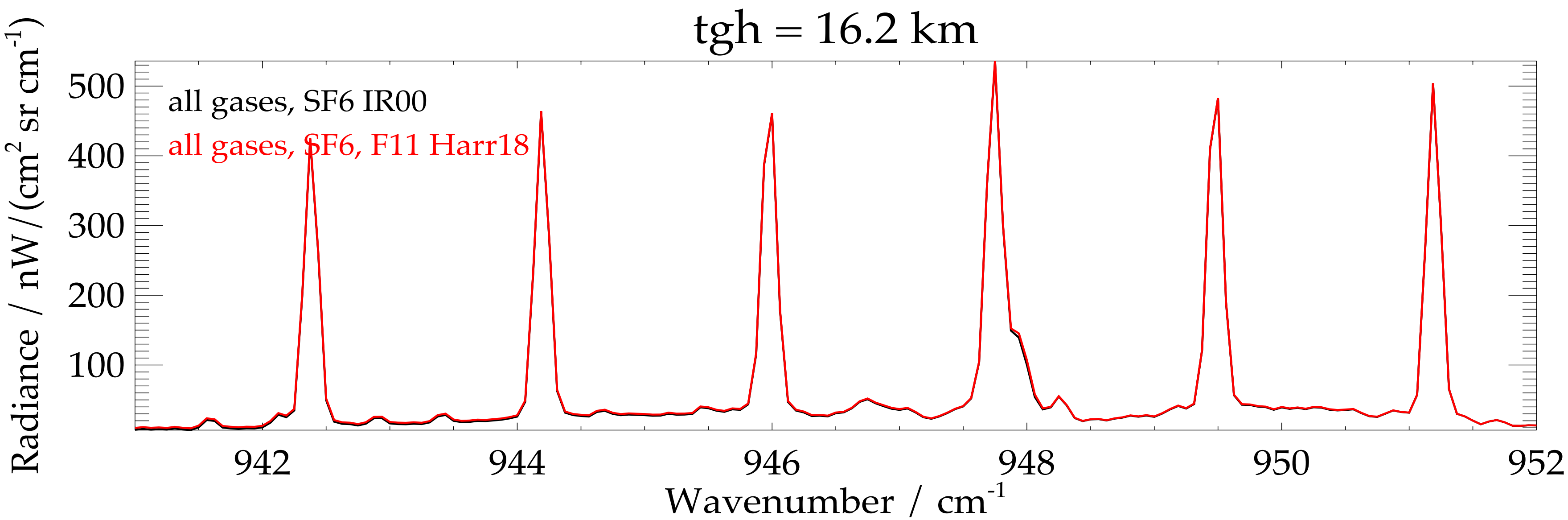
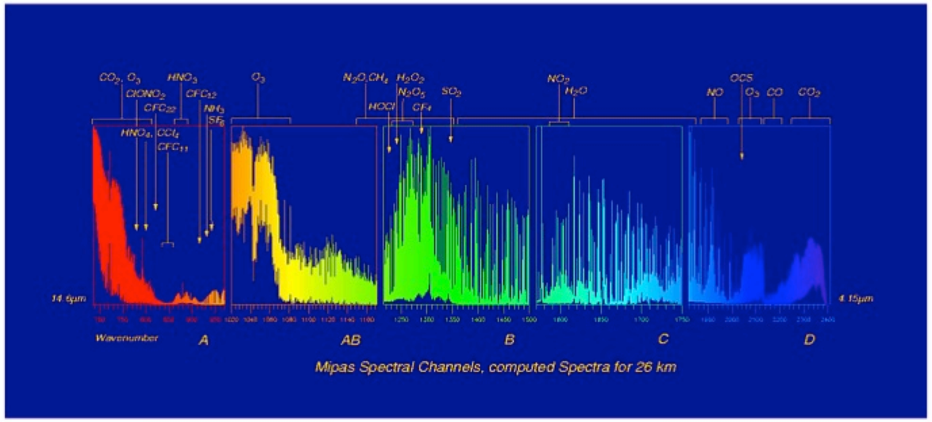
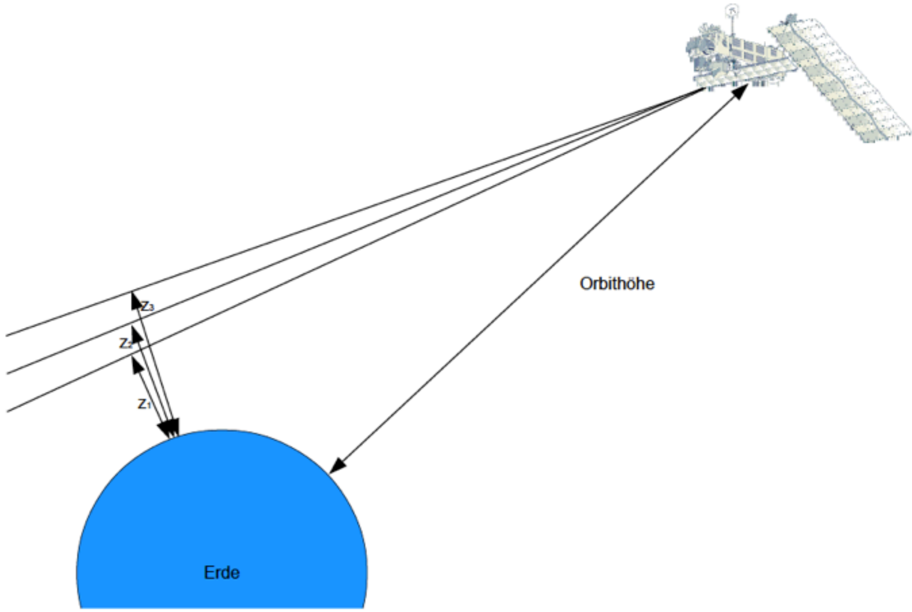


MIPAS observations

- MIPAS was a limb sounder able to detect a wide range of species from the UT to the mesosphere
- Active from July 2002 to April 2012
- At IMK, we have derived 10-years data records of global distributions of ~30 species and isotopologues
- Among them is **SF6** and other tracers and greenhouse gases …
- Due to the limb sounding geometry, the sensitivity to low-abundant species is high.
- The lowest observation altitude is cloud top or ~ 6km, whatever is higher.





Spectral signal of SF6

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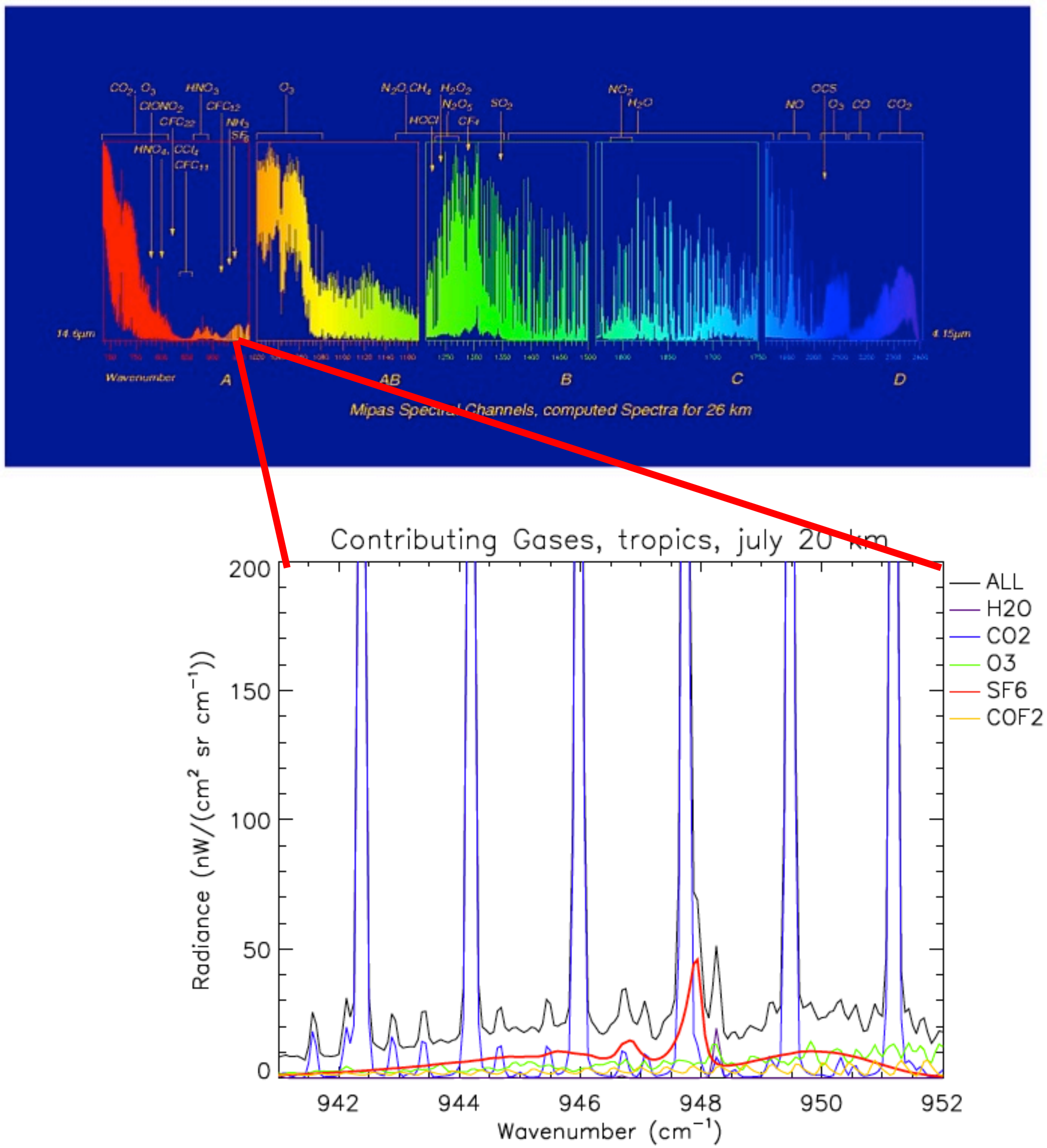
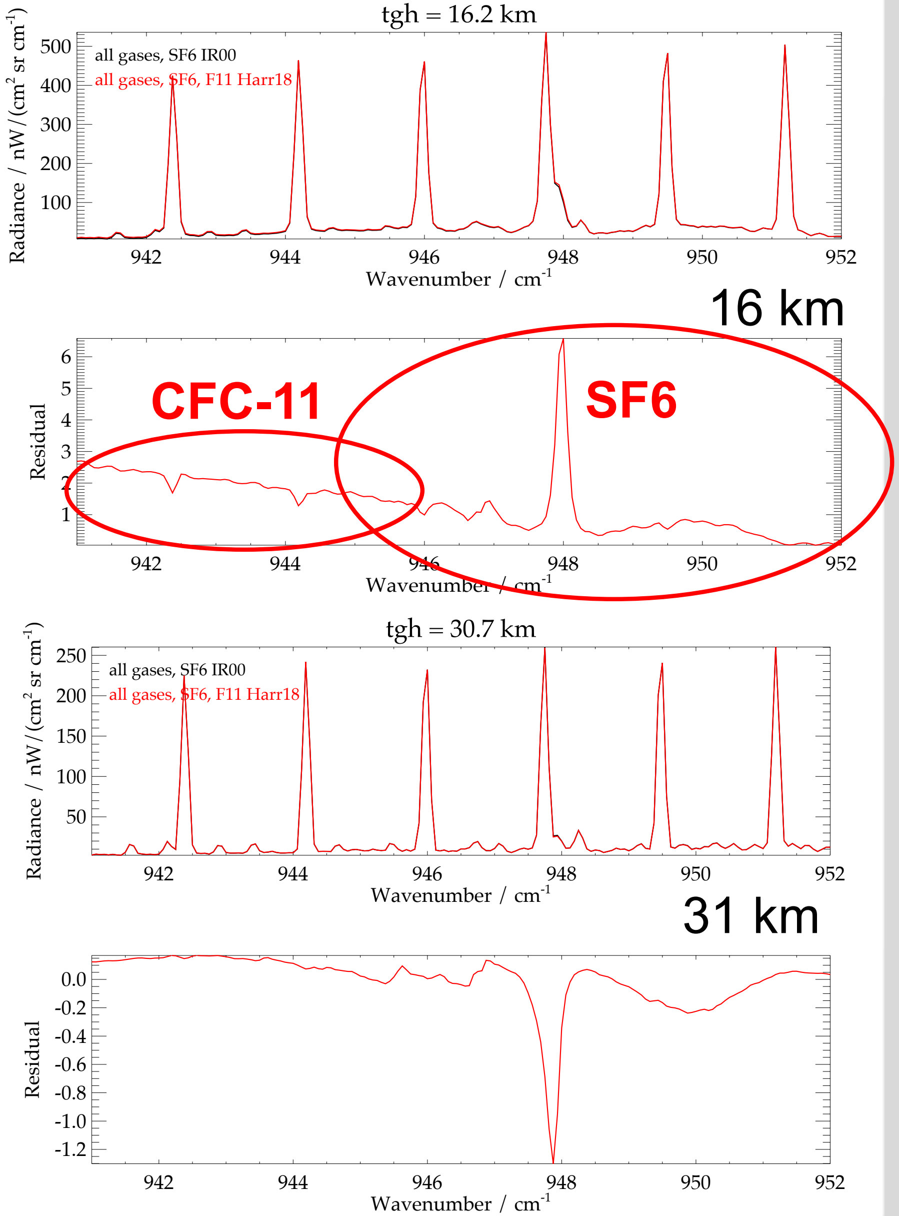


- The spectral signature of SF6 is shown by the red curve (lower figure on the left).
- It is a rather small signature between strong CO2 lines (shown in blue).
- CO2 is in Non-LTE at higher altitudes; this affects the CO2 line shapes and must be modelled carefully.
- Other species with signatures in the relevant spectral regions are also shown in the figure; CFC-11 is missing.
- It is important to model all contributions correctly in order to get the signal of SF6 right.

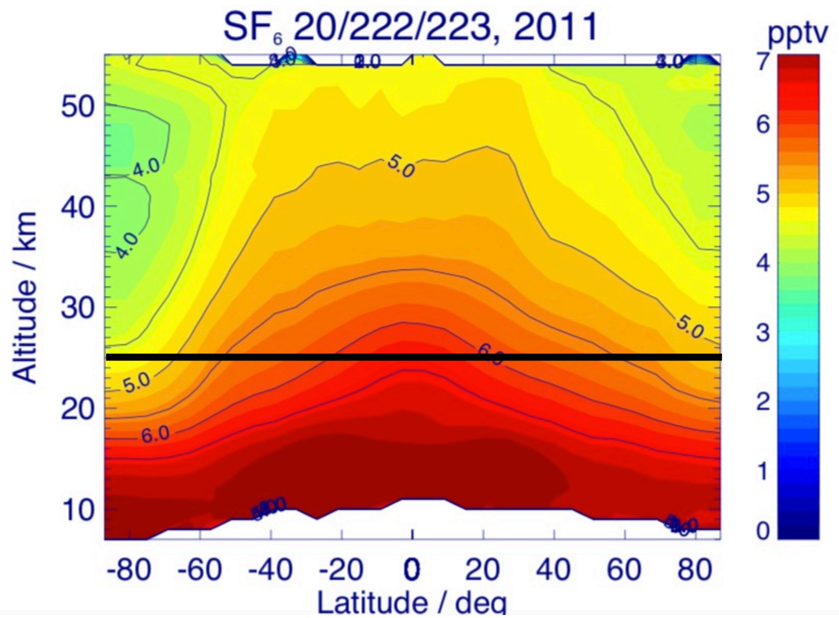
Impact of new absorption cross sections of SF6 and CFC-11

New spectroscopic improvements (J. Harrison, to be published):

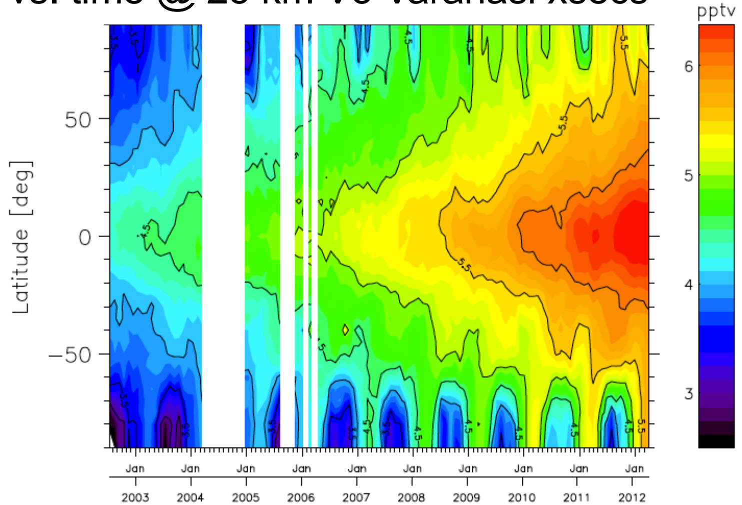
- Newly measured SF6 absorption cross sections (37 data sets for pressures between 0 and 750 torr and temperatures between 189 and 294 K)
- Completely new data on CFC-11 in this spectral range (not available before)
- The figures on the right show calculations of spectra for two MIPAS tangent heights (16 and 31 km) with the new (red) and the old (black) spectroscopic data for SF6 and CFC-11 (no CFC-11 included in the old ones). The lower panel of each figure shows the difference new - old.
- In the lower atmosphere (top panels) the SF6 signal becomes stronger for the same SF6 vmr profile => for a given signal in the measurement, the retrieved vmr will decrease with the new spectroscopic data.
- In the upper atmosphere (bottom panels), the situation is the contrary: the signal becomes weaker, i.e. the retrieved SF6 vmr will increase.



Impact of new cross-sections on SF6 distributions

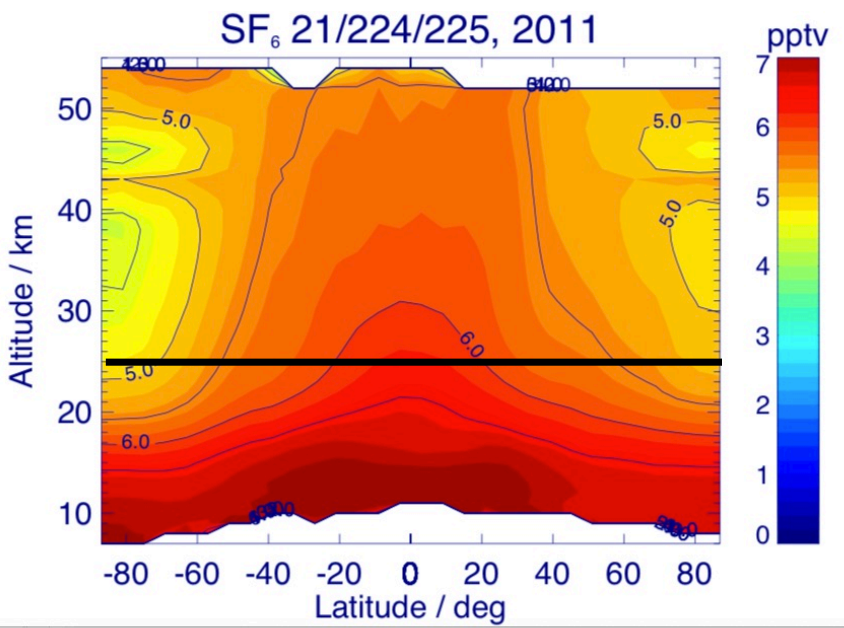


SF6 vs. time @ 25 km V5 Varanasi xsecs



Haenel et al., 2015

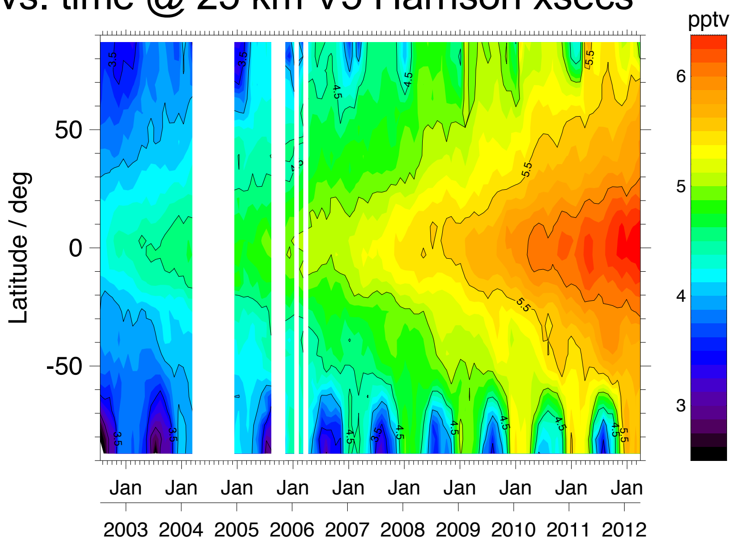
OLD



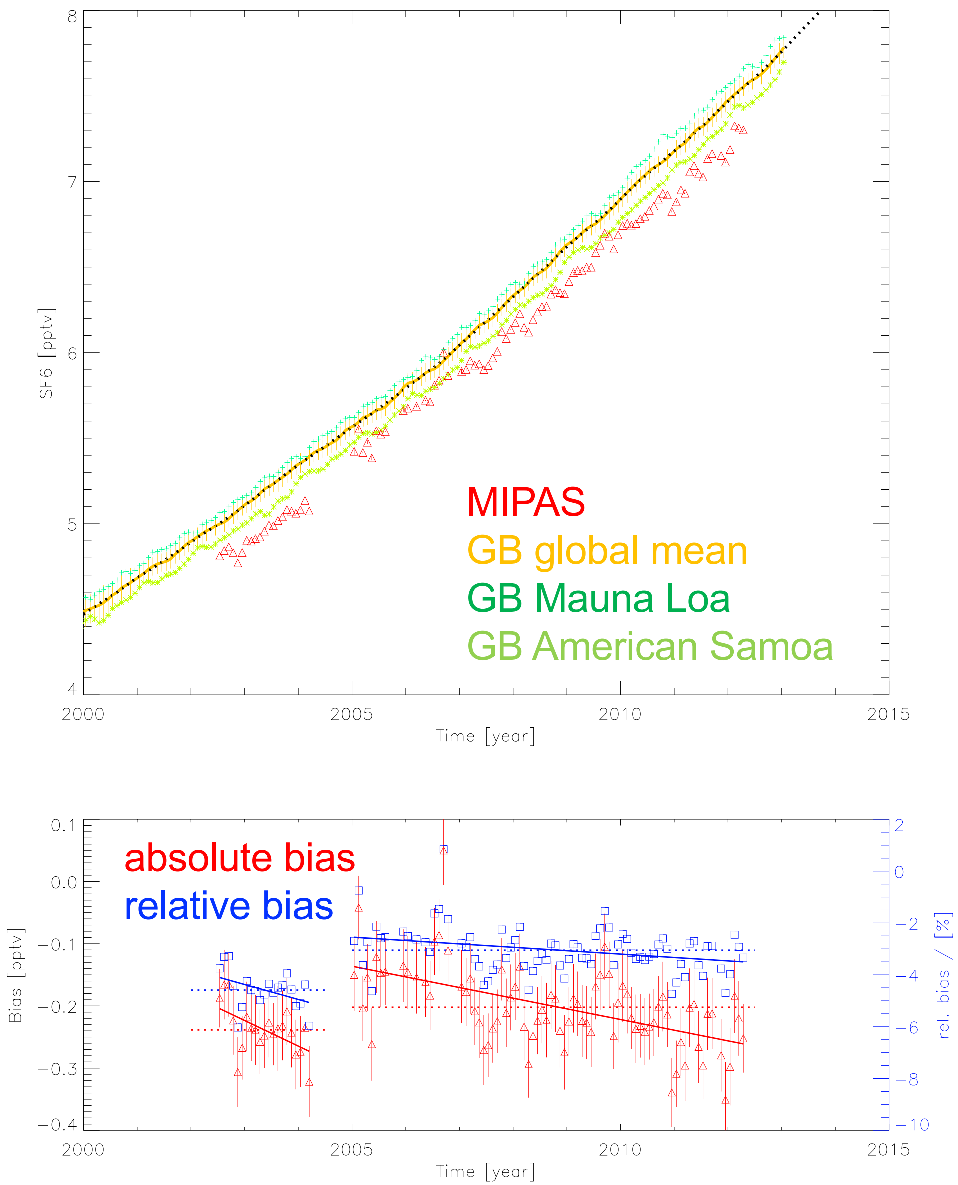
NEW

Small differences in the lower atmosphere (shown for 25 km on the right), strong differences in the upper stratosphere

SF6 vs. time @ 25 km V5 Harrison xsecs



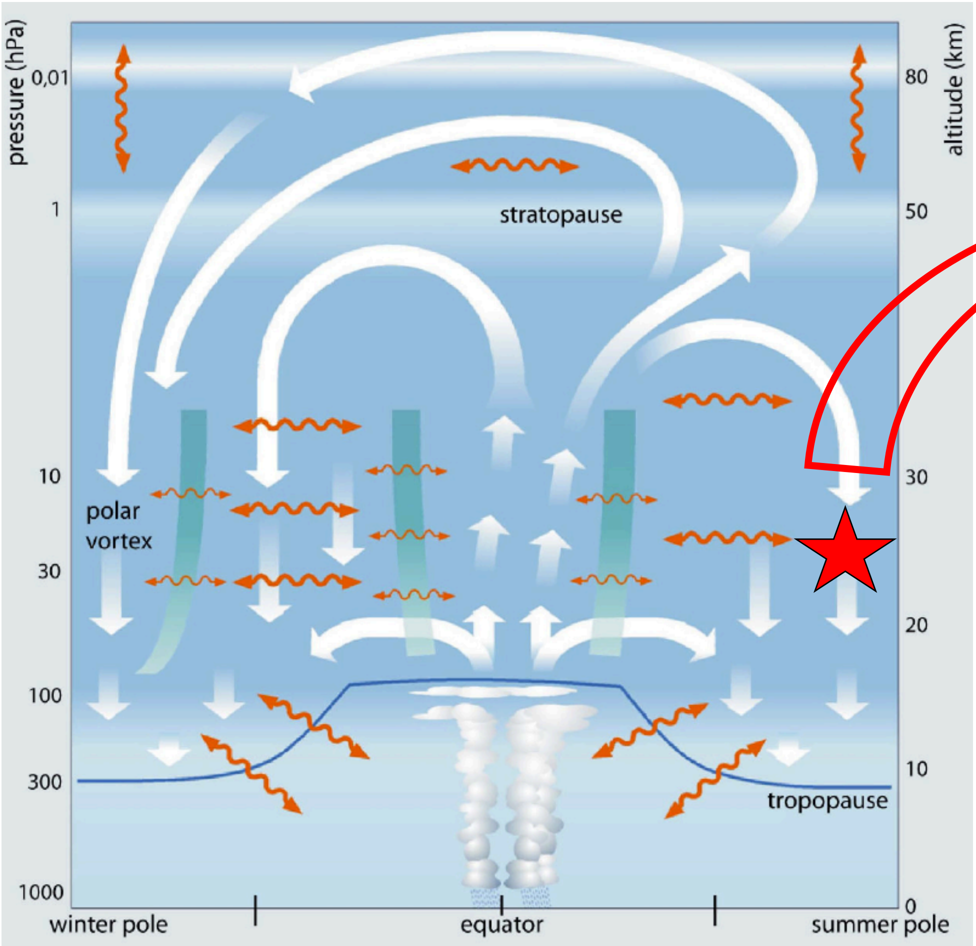
Comparison to ground-based data (NOAA ESRL/GML)



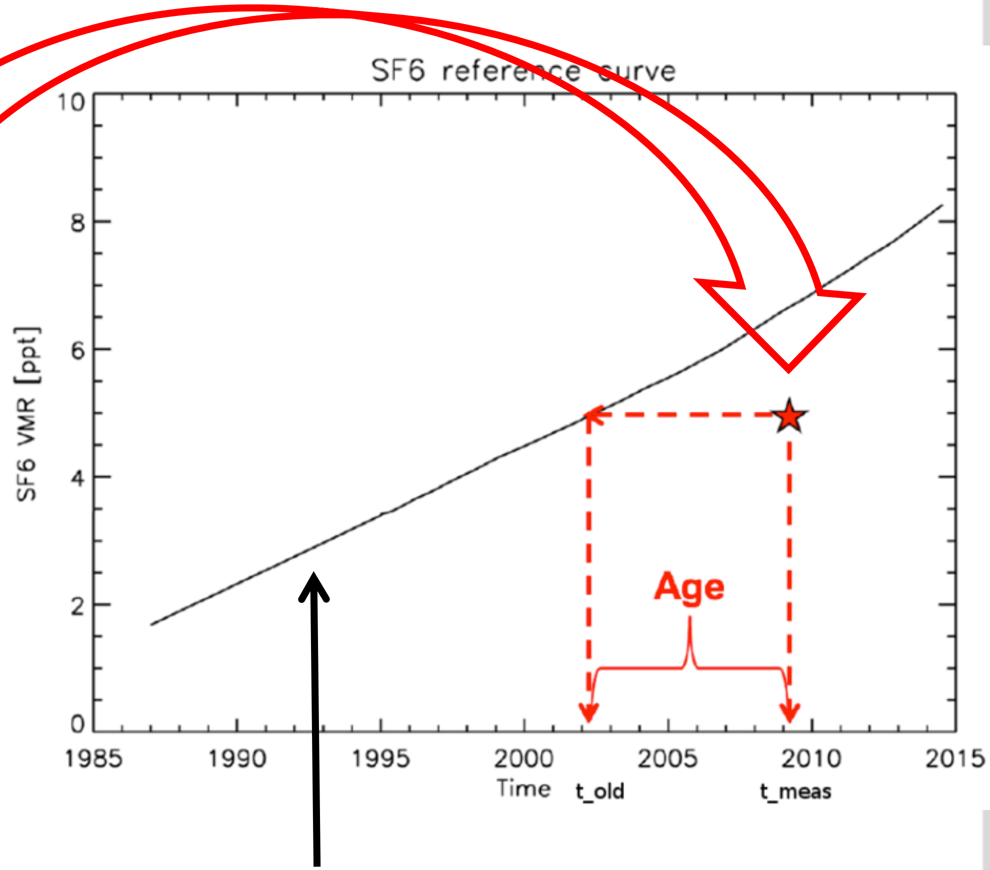
- Top left: comparison of MIPAS tropical free troposphere data (monthly averages over 25S to 25N and 9 to 15 km altitude) with ground-based in situ data from ESRL/GML, two tropical stations and the global mean.
- Tropical tropospheric SF6 data have an almost constant multiplicative (i.e. relative) bias vs. ground-based reference data of ~ -3% (blue in the lower panel); the absolute negative bias (red) increases over time.
- **=> Correction of all MIPAS retrieved SF6 data by 1./0.97**
- Caveat: could be just the cross-section data set for tropical upper troposphere p/T conditions
- Remaining trend vs. GB reference data due to MIPAS instrumental drift? If so, should be improved for V8 data

SF6 and age of air – the Brewer-Dobson Circulation

Climate models show a continuous acceleration of the Brewer-Dobson Circulation since ~ 1960 and predict further acceleration for the future under climate change.



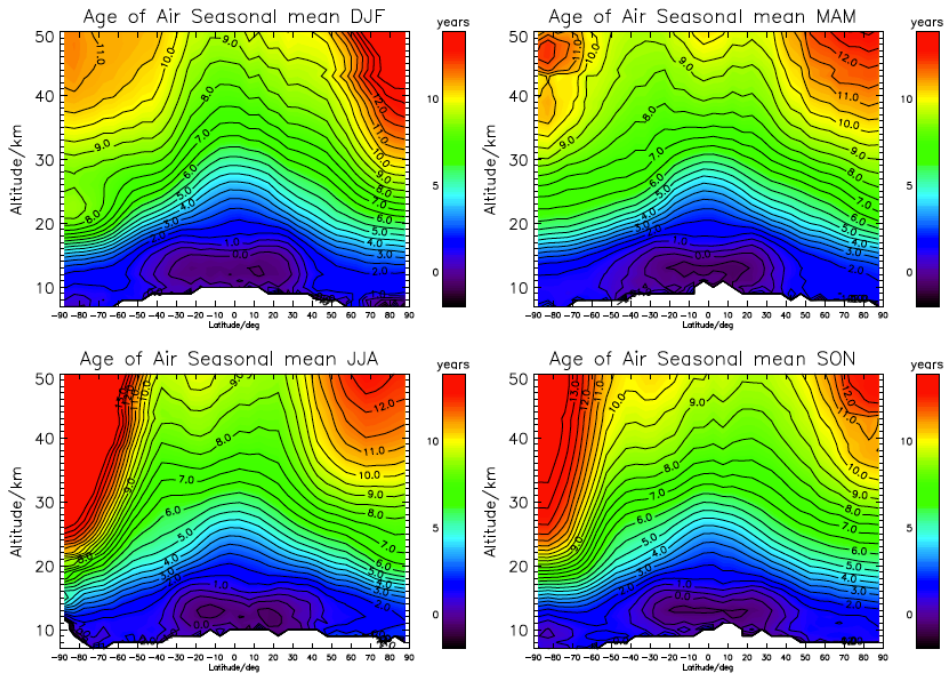
Boenisch et al., 2011

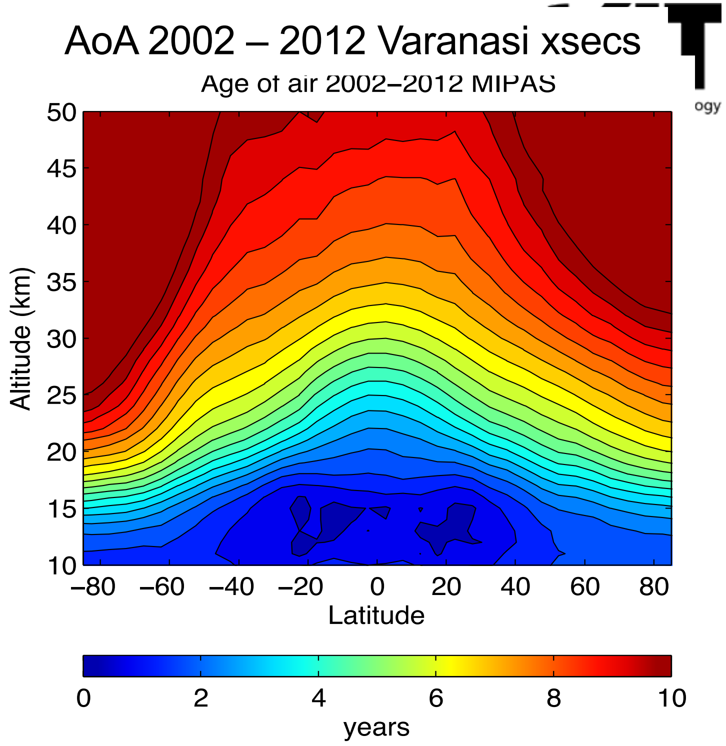


SF6 is an inert tracer and has no sinks in the stratosphere

Reference line – (almost) linear SF6 increase in the troposphere

Impact on AoA distributions

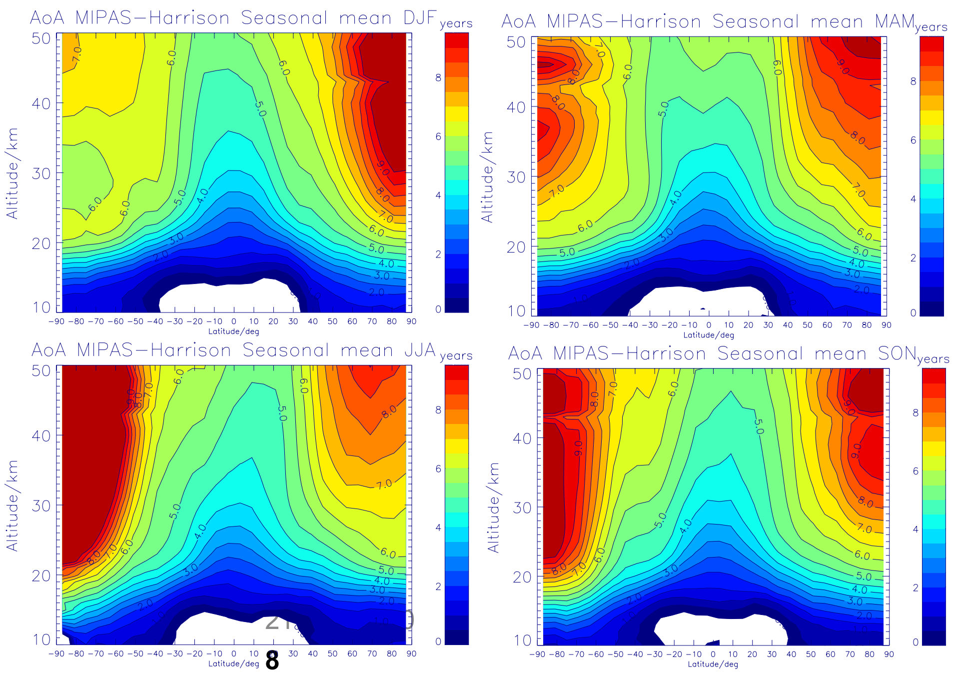




Haenel et al., 2015

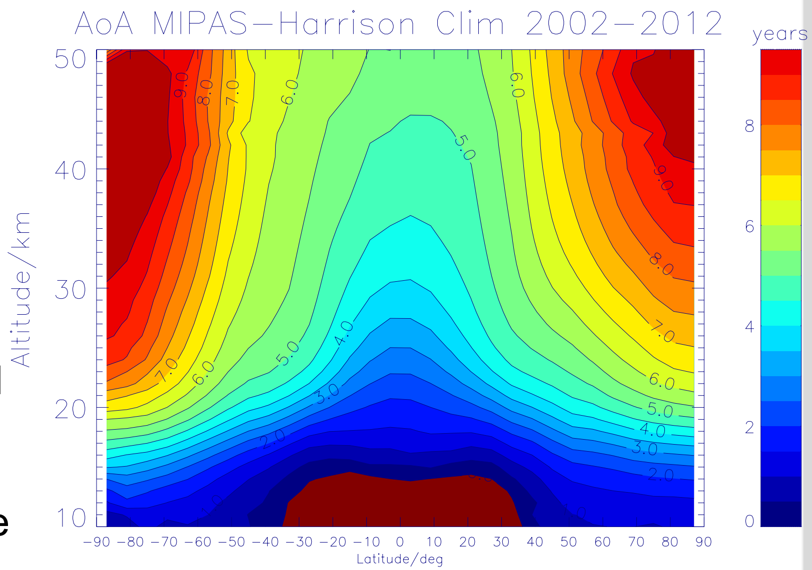
OLD

Seasonal means of AoA (left); mean AoA over complete MIPAS period (right)



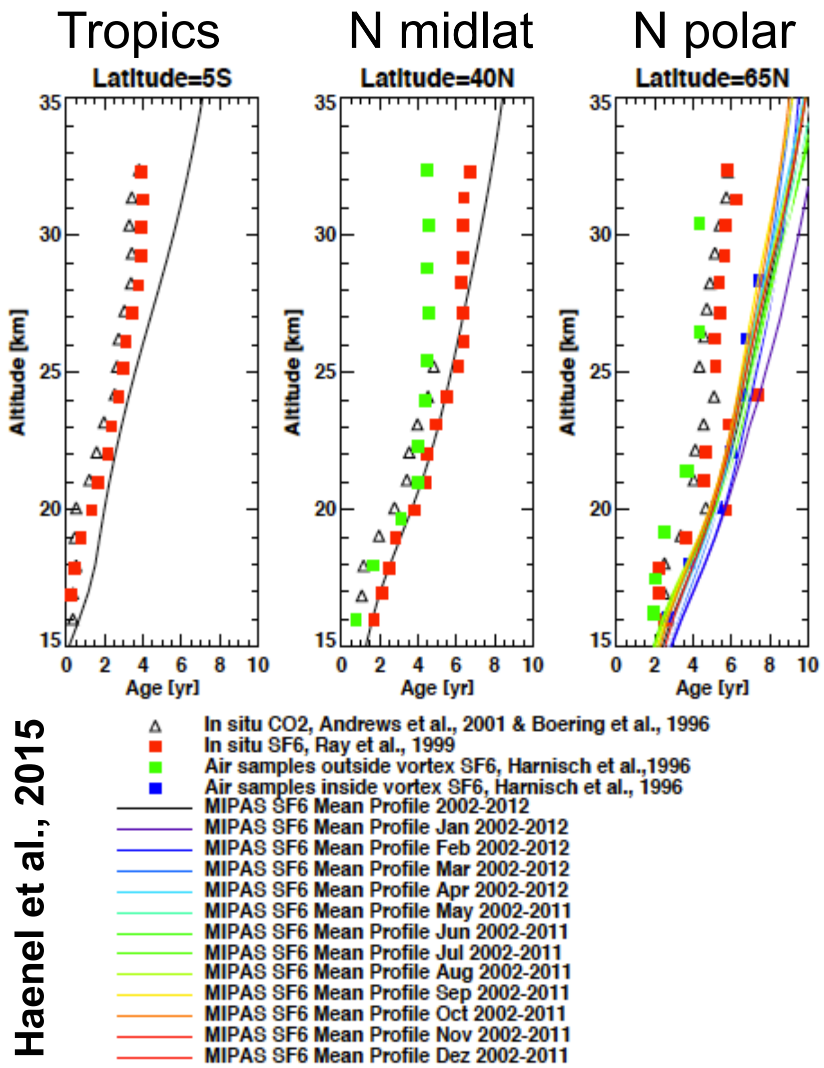
NEW

Similar to SF6, strongest differences between new and old AoA appear in the upper stratosphere



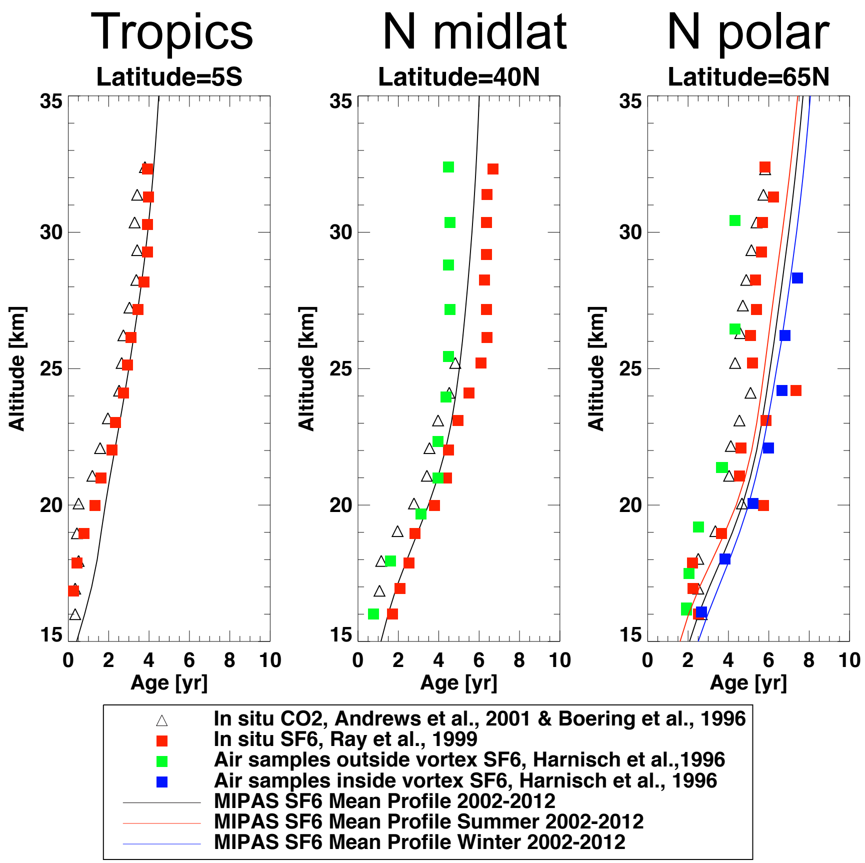
Comparison to independent measurements

OLD



Haenel et al., 2015

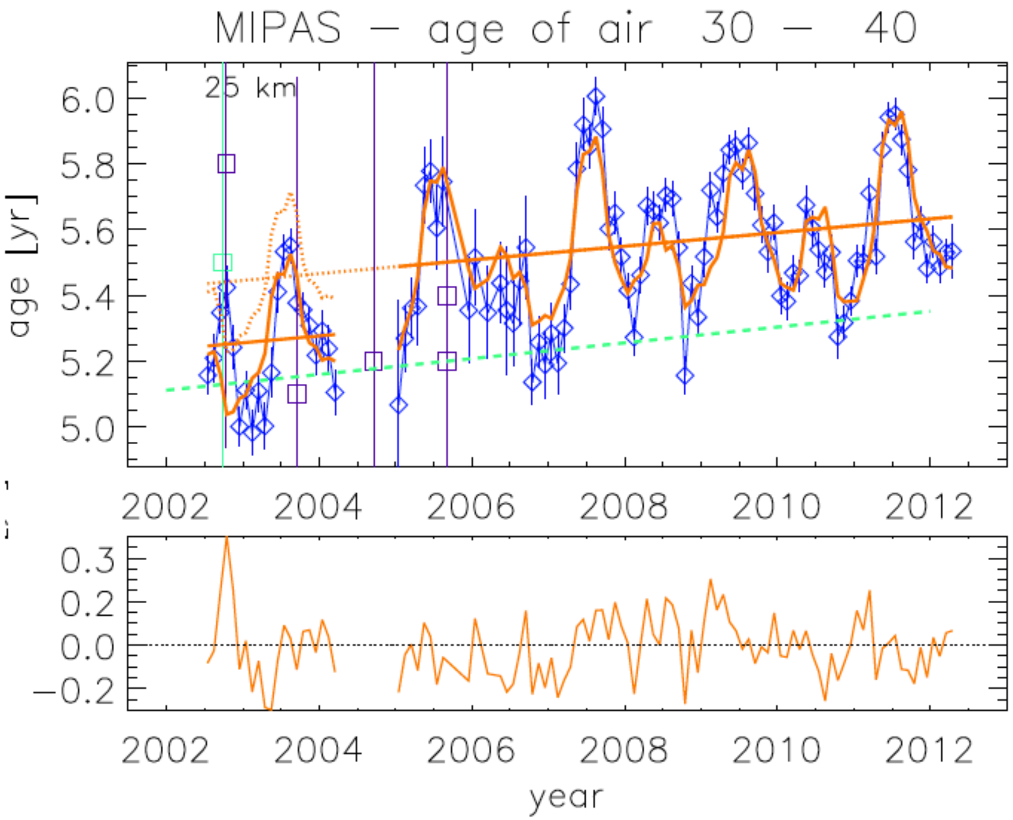
NEW



Better agreement with balloon-borne reference data, in particular in the upper part of the profiles

Determination of trends

- Multivariate linear regression of the time series of monthly zonal means of volume mixing ratios in a latitude/altitude bin (typically 10 deg, 1 km)
- Parameters fitted: seasonal variation (sin/cos) and higher harmonics, 2 QBO terms, linear term, offset
- The linear term is interpreted as the trend over time
- Example:



Haenel et al., 2015

Zonal AoA trend distribution (with idealized AoA spectra, autocorrelation and modelling errors considered in the trend fit)

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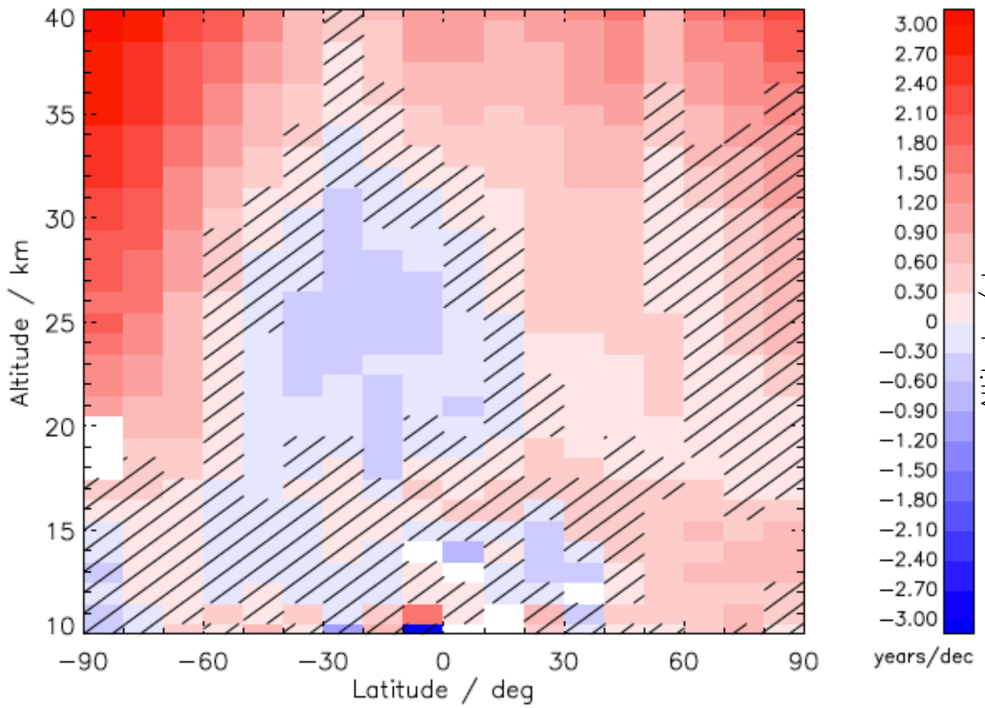
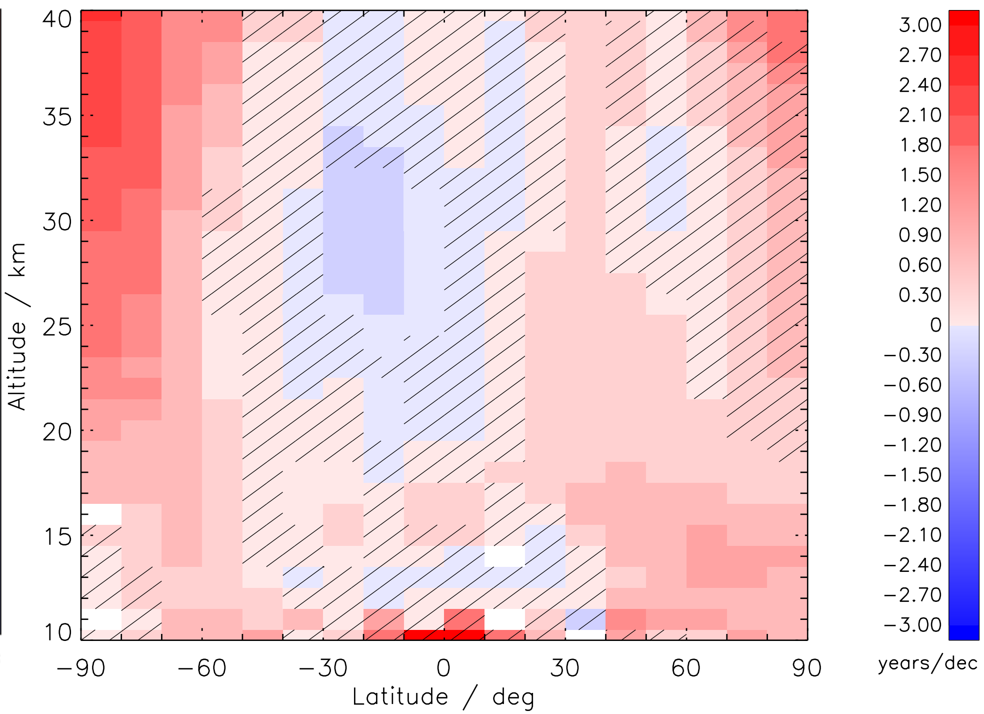
From SF6 V5 Varanasi Xsecs



From SF6 V5 Harrison Xsecs



Haenel et al., 2015 Hatched areas are insignificant on the 2-sigma level (this means the trend is consistent with zero within its 2-sigma uncertainty)

Morphology of the trend distribution remains the same; i.e.
positive trend in the NH and negative trend in the SH and the tropics
Differences are present in details of the distribution

How to explain the different trends in NH and SH?

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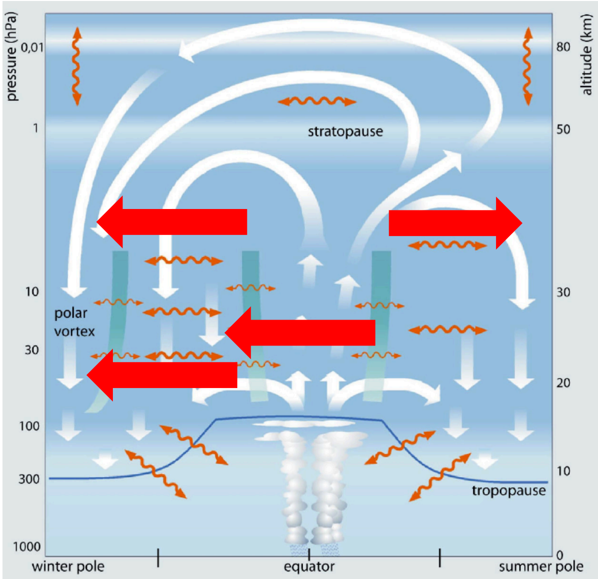
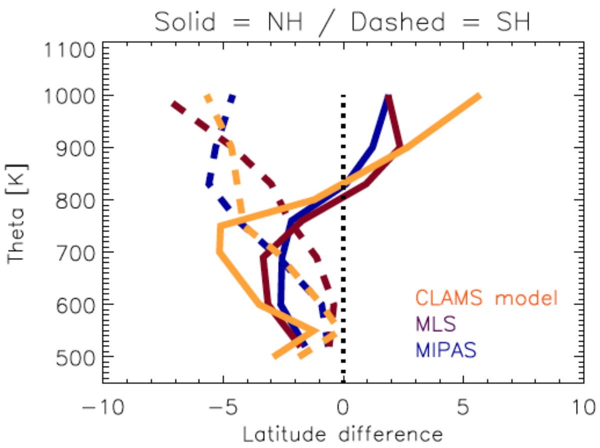
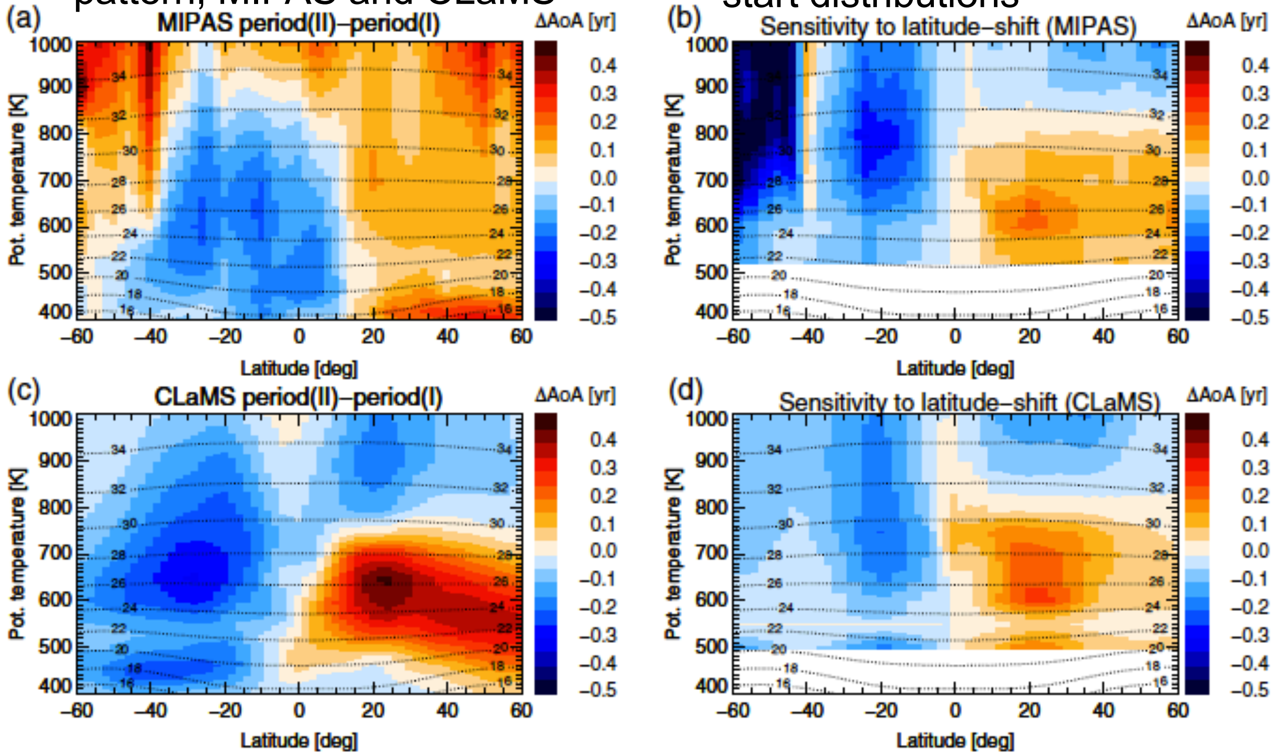




Stiller et al., 2017

"Observed" AoA trend pattern, MIPAS and CLaMS

Detected shift applied to the start distributions



- Hypothesis: Shift of the circulation pattern to the South and widening of the tropical pipe above ~30 km (top left) can explain AoA and tracer trends
- Such a shift has been detected by analysing the positions of the subtropical mixing barriers (bottom left)
- Applying the detected shift on AoA and N2O distributions reproduces well the detected trend patterns (top)

Do the new trends make (more) sense? Yes!

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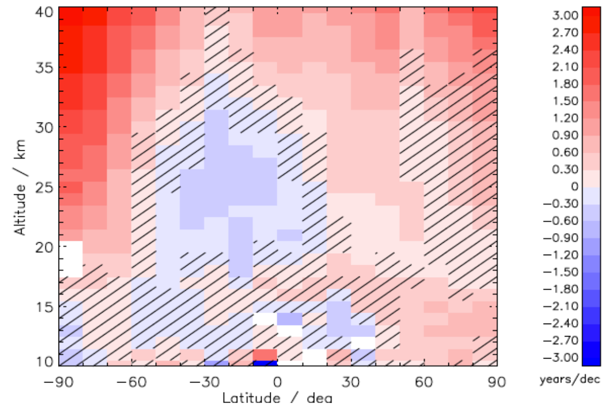
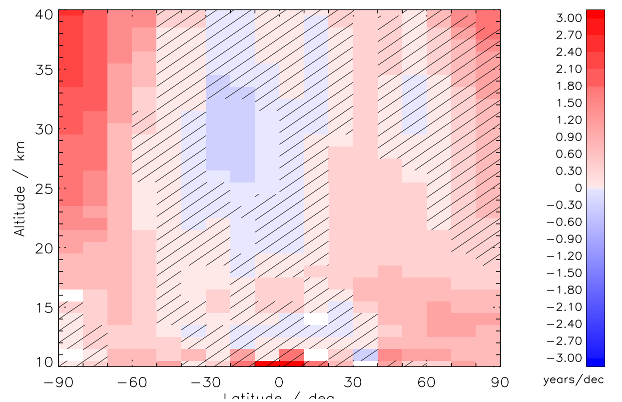
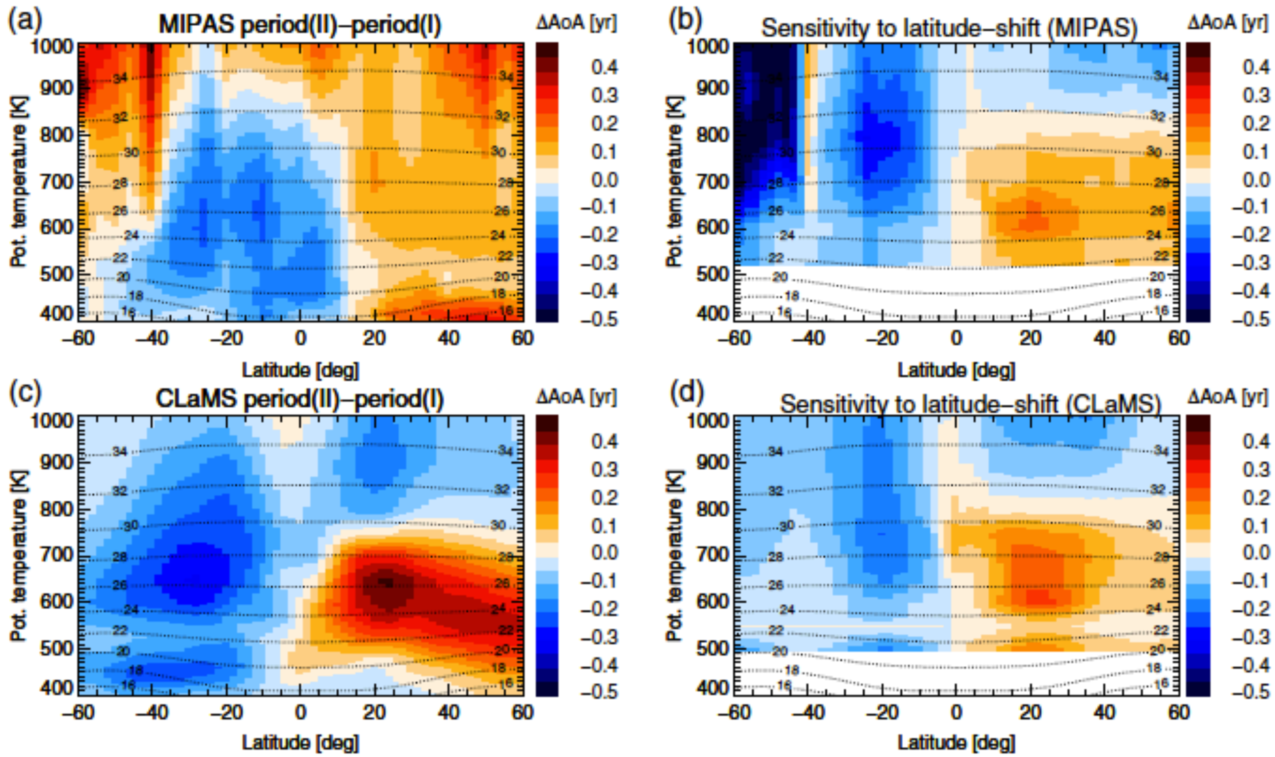
The **new AoA trend** pattern (top right) reproduces the **trend pattern expected from the shift of the mixing barriers** (middle right) even better than the **previous AoA trend pattern** (top and middle left; note the different representations: altitude vs. potential temperature as vertical axes)



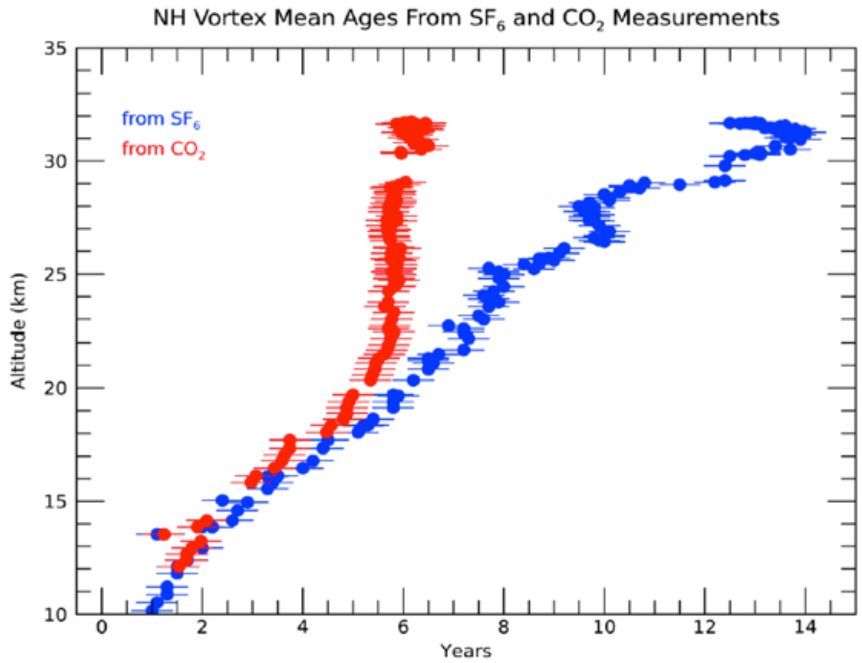




Stiller et al., 2017

Problems with SF6 as AoA tracer

- SF6 has a sink in the mesosphere that is much stronger than previously thought (Ray et al., 2017)
- Newly estimated lifetime is ~850 years instead of 3200 years
- SF6-depleted air coming down from the mesosphere (in polar winters) makes the air looking far older than it is in reality



Ray et al., 2017

this is observed in MIPAS AoA data as well

➔ search for new (better) AoA tracers

➔ alternative approaches, e.g. inversion of the continuity equation (see von Clarmann et al., 2016; 2019; 2020)

Summary and conclusions

- New spectroscopic data of SF6 and CFC-11 provided by J. Harrison were used to retrieve SF6 from MIPAS data
- SF6 distributions change considerably: much higher vmrs in the upper stratosphere
- MIPAS SF6 in the tropical free troposphere has a low bias of ~ -3%; as a consequence, all SF6 data have been scaled accordingly
- AoA distributions reflect the changes in SF6: much younger air in the upper stratosphere, in particular in the tropics
- New AoA data agree better with independent balloon measurements and model results, and support better the finding of a shift of the mixing barriers/ the tropical pipe
- Take home message: remote sensing measurements depend critically on high quality laboratory spectroscopy measurements!

Thank you!