Sulfur hexafluoride (SF_6) simulations with BASCOE for Brewer-Dobson circulation studies

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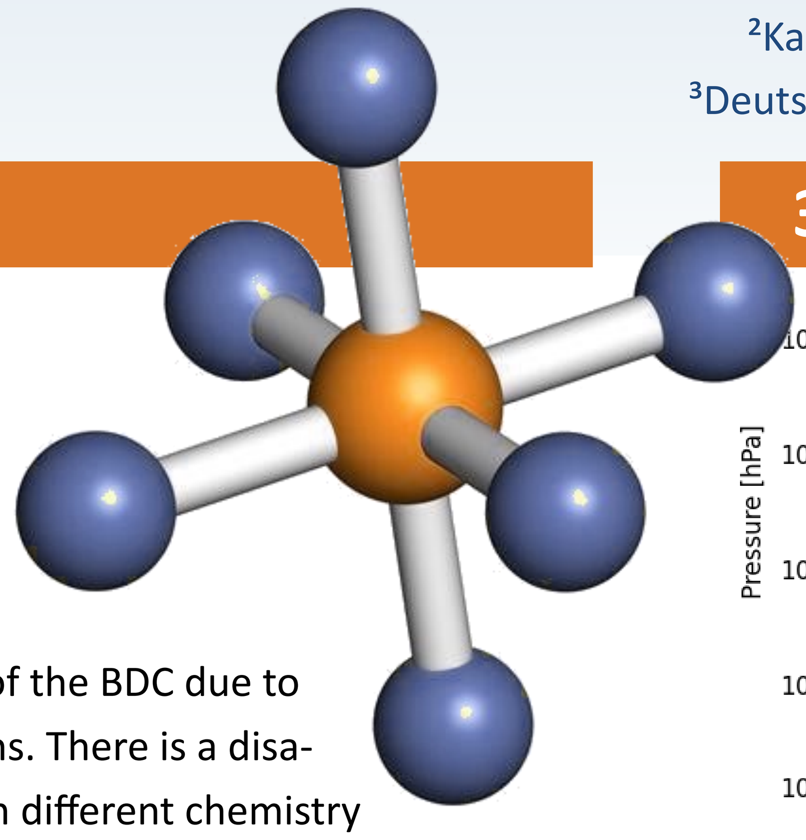
QOS 2024
Session D, 3-17

1. Introduction - circulation studies

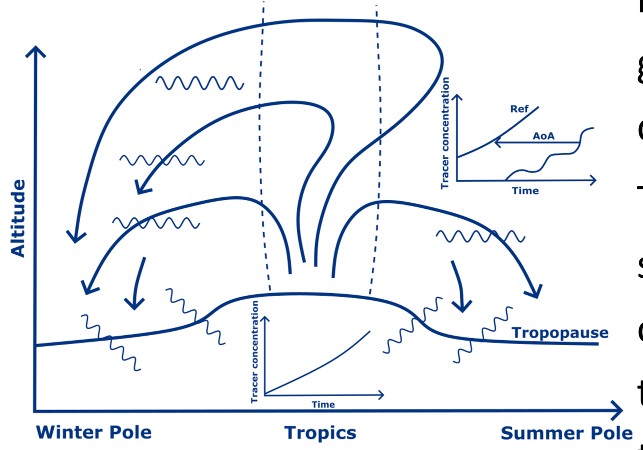


The distribution of stratospheric ozone (O_3) is determined by

- the chemical production and loss mechanisms of O_3
- transport via the Brewer-Dobson Circulation (BDC)

Climate change:

General Circulation Models (GCM) predict an increase in the strength of the BDC due to rising greenhouse gas concentrations. There is a disagreement between BDC trends from different chemistry climate models, observations and reanalysis data sets.



The CAIRT mission, a candidate for ESA's Earth Explorer 11, aims to resolve this by measuring trace gases to compute Age of Air (AoA). Age of Air is the time it takes for an air parcel to move from the troposphere to the stratosphere. AoA can be computed from linearly increasing trace gases with a long lifetime in the stratosphere such as CO_2 and SF_6.

This work:

The chemistry of SF_6 has recently been implemented in the chemistry transport model (CTM) of the Belgian Assimilation System for Chemical ObsErvations (BASCOE) to support CAIRT.

We discuss two BASCOE simulations spanning 10 years each (2002-2012), driven by the meteorological reanalyses ERA5 and MERRA2 respectively. The implementation of the SF_6 chemistry is validated via a computation of the global atmospheric lifetime of SF_6 using BASCOE output. Both model simulations are also evaluated using MIPAS and ACE-FTS satellite observations.

2. SF_6 chemistry in BASCOE

The Belgian Assimilation System for Chemical ObsErvations is a chemistry transport model driven by winds and temperature from reanalysis data sets (ERA5, MERRA2). We first included the chemistry of SF_6 in BASCOE. SF_6 is destroyed in the mesosphere by electron attachment. The chemical reactions implemented in BASCOE are as follows:

The **electron density** is a climatology that depends on altitude, latitude, scale height, day of the year, solar zenith angle and air density.

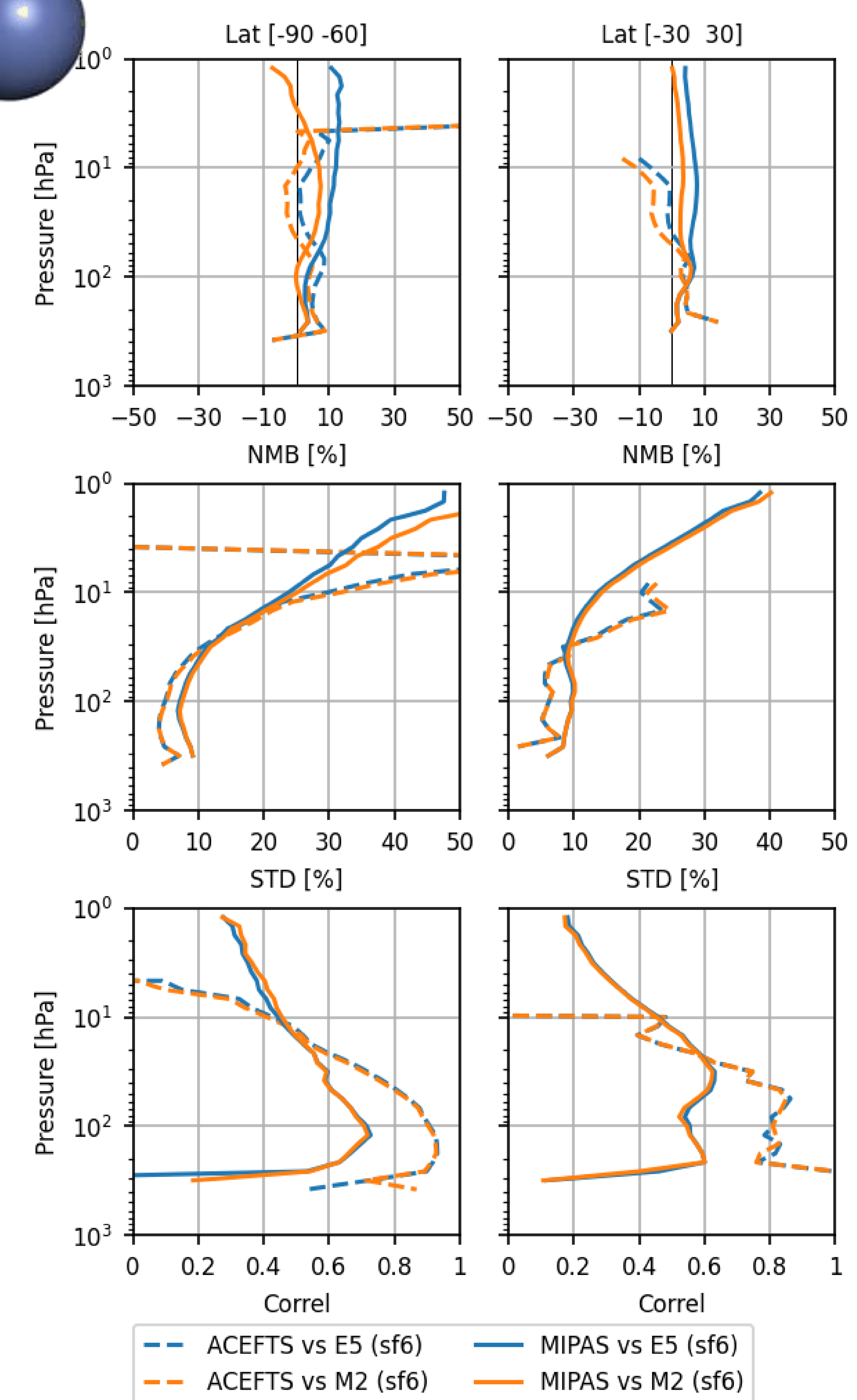
$$\begin{array}{lll}
SF_6 + e^- & \rightarrow (SF_6^-)^* & \text{auto-attachment} \\
(SF_6^-)^* + M & \rightarrow SF_6^- & \text{stabilization} \\
(SF_6^-)^* & \rightarrow SF_6 + e^- & \text{autodetachment} \\
SF_6^- + O_3 & \rightarrow SF_6 + O_3^- & \\
SF_6^- + H & \rightarrow SF_5^+ + HF & \\
SF_6^- + HCl & \rightarrow \text{products} & \\
SF_6^- + h\nu & \rightarrow SF_6 + e^- & \text{photodetachment}
\end{array}$$

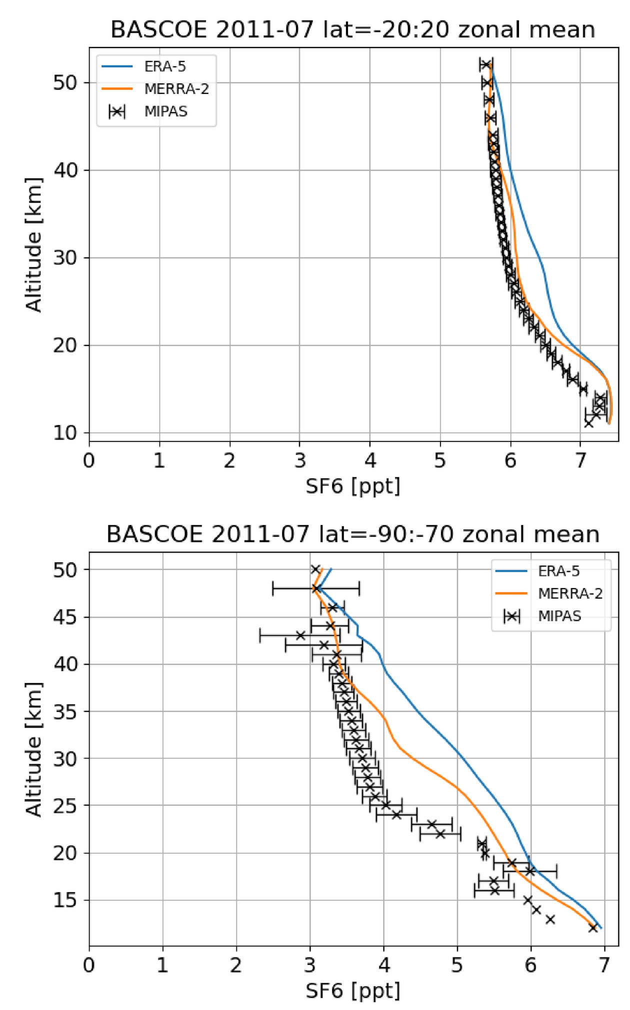
Reference list

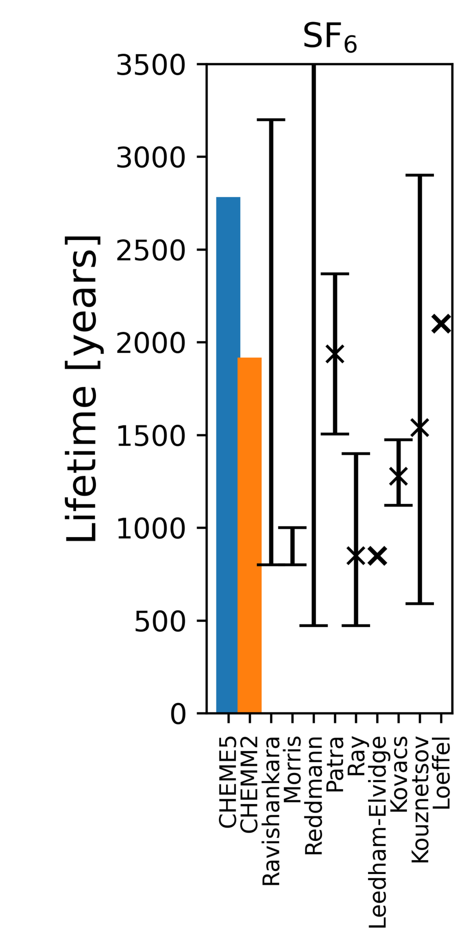
Acknowledgements

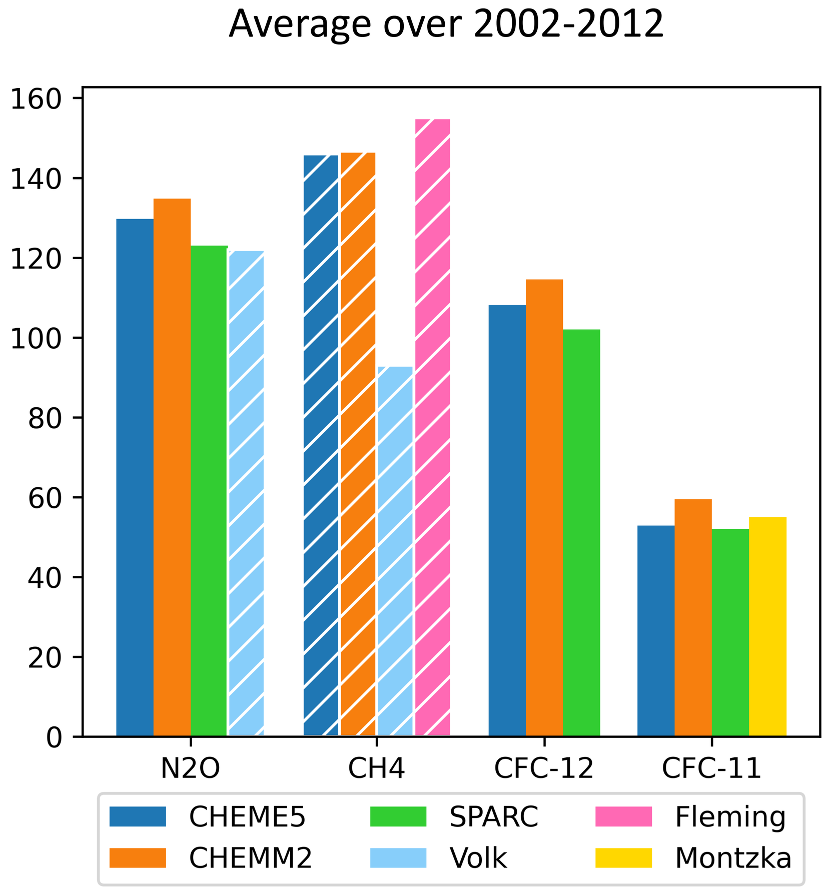
Special thanks to S. Löffel, H. Garny, R. Eichinger and T. Reddmann for sharing their electron climatology and model output and for answering many questions. We also thank G. Stiller for explaining the method to correct age of air for the nonlinearity of the emissions. This work uses the global mean SF_6 flask and in-situ data from NOAA/GML.

3. SF_6 simulation validation - bias and atmospheric lifetime









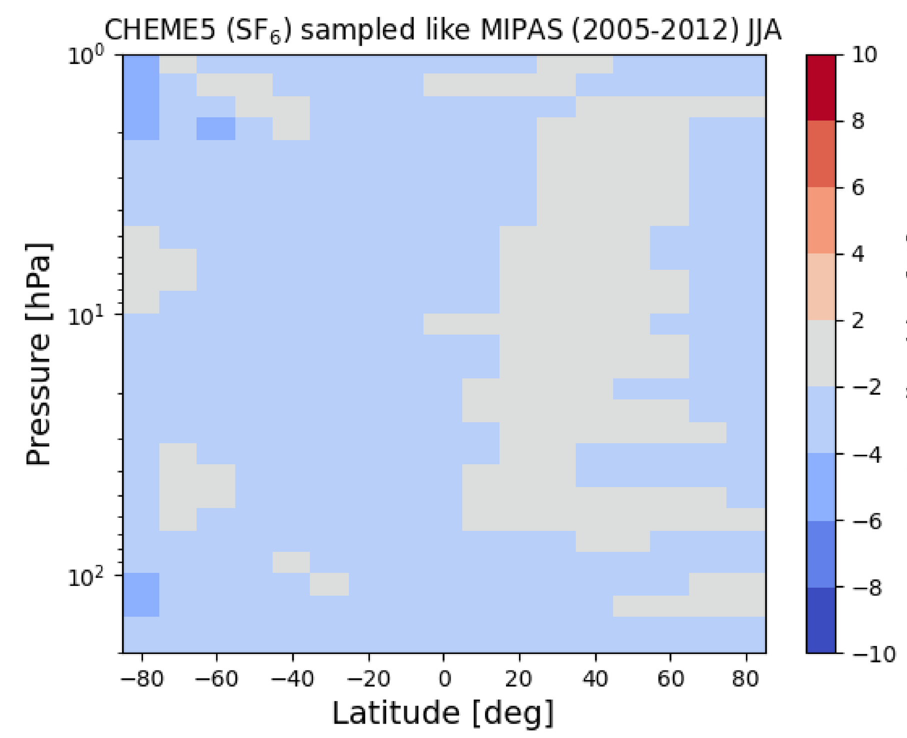
Left plot: bias, standard deviation and correlations of BASCOE output with respect to MIPAS or ACE-FTS measurements of SF_6. The bias with respect to MIPAS is only approximately 10%. Correlations are better (>0.8) with ACE-FTS.
Middle plot: SF_6 volume mixing ratios from BASCOE compared with MIPAS. **Right plot:** the global atmospheric lifetime of SF_6 is computed to test the implementation of the chemistry using **τ=burden/loss**. There is a large difference between the lifetime of SF_6 with the run driven by ERA5 (CHEME5) and the run driven by MERRA2 (CHEMM2). This is due to the differences between the reanalyses at high altitude where the sink of SF_6 becomes important. The lifetimes of other species (N_2O, CH_4, CFC-11 and CFC-12) agree more between the two runs. White hatching indicates a stratospheric lifetime.

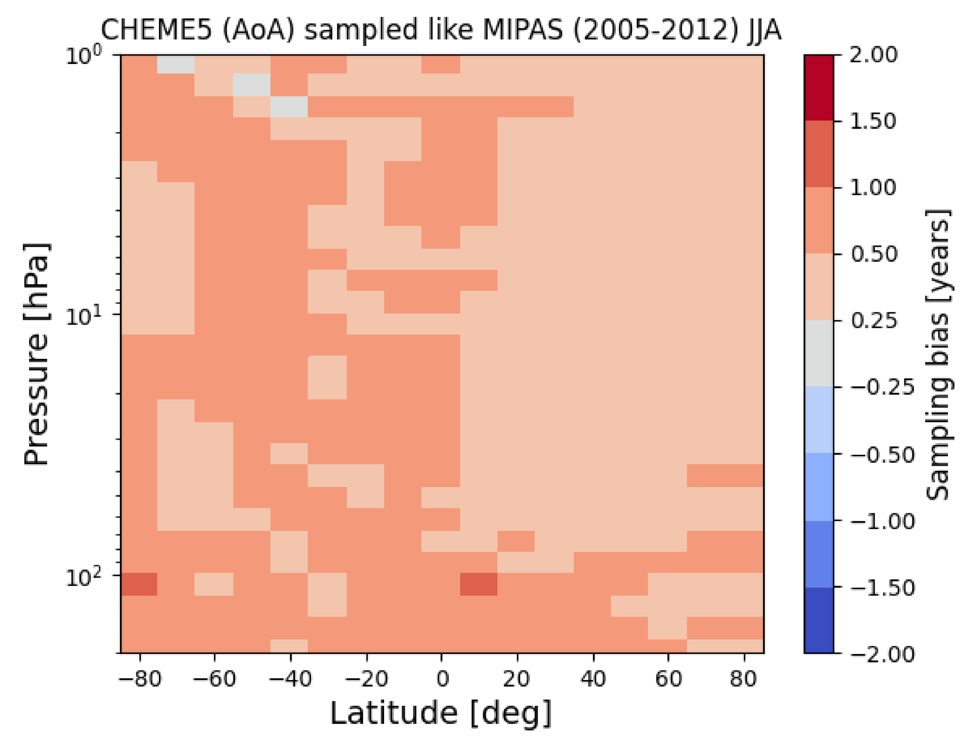
4. Comparing models and observations - sampling bias

The atmosphere is non-uniformly sampled by satellite instruments. This can create a sampling bias when climatologies are computed from observations. The bias is computed by taking the difference between monthly zonal means of the model sampled like the instrument (interpolated at the locations of the observations) and monthly zonal means of the model on its own grid.

$$\text{sampling bias} = \frac{\text{mod at obs} - \text{mod}}{\text{mod at obs}}$$

We only look at the effects of horizontal and temporal resolution, but not at the vertical resolution. We find a sampling bias of 2-4% for SF_6 with a higher bias in the southern hemisphere for all seasons (left plot). The figure shows a seasonal mean of June-July-August, averaged over the period 2005-2012. This translates into an age of air sampling bias (right plot) of around 0.25 years. Note that the AoA sampling bias is not normalized. Results are similar with ERA5 and with MERRA2.





Future work:

- Do an observing system simulation experiment for the CAIRT mission
- Try other reanalyses (ERA-Interim, JRA-3Q)