LOng-LIved greenhouse gas PrOducts Performances (LOLIPOP)

WP3400 - CLaMS stratospheric circulation estimates

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Table of Contents

1 Introduction3

2 Approach/Methods4

2.1 CLaMS model simulations4

2.2 Input data description5

2.2.1 Reanalysis data5

2.2.2 Trace gas boundary conditions6

2.3 Compilation of climatologies7

3 Preliminary results10

3.1 The stratospheric circulation in reanalyses10

3.2 Trace gas distributions11

4 Outlook13

List of acronyms and abbreviations13

References15

1 Introduction

The global circulation in the stratosphere (termed the Brewer-Dobson circulation, BDC) is an important element in the climate system as it controls the distributions and changes in concentrations of long-lived trace gas species. Changes in the distributions of these radiatively-active species, in turn, may have substantial effects on the global radiation budget. Hence, profound knowledge on stratospheric circulation changes and the dynamical mechanisms involved is crucial for understanding past and future climate variations.

However, stratospheric circulation velocities are extremely small and cannot be directly measured. Therefore, these velocities must be inferred from the stratospheric distributions of long-lived trace gas species, such as SF6, CFCs, or N2O. However, the relationship between changes in these chemical species and changes in the stratospheric circulation are not fully understood. In particular, changes in observed trace gas mixing ratios are not in agreement with a long-term acceleration of the stratospheric circulation as predicted by global climate models in response to increasing greenhouse gas (GHG) concentrations. Furthermore, meteorological reanalysis data sets, which are designed to provide a best guess of the atmospheric state, show very different changes of the stratospheric circulation over the past decades. Thus, there is an urgent need to improve the representation of the stratospheric circulation in models and reanalyses.

This work package WP3400 (CLaMS stratospheric circulation estimates) will provide improved reanalysis-driven model simulations which will enable enhancing the process understanding of the effects of the stratospheric circulation on the global distributions of long-lived trace gases. In particular, within WP3400 we will carry out model simulations with the Chemical Lagrangian Model of the Stratosphere (CLaMS, for details see Pommrich et al., 2014), driven with meteorological fields from different reanalysis data sets. Trace gas transport in the CLaMS model is formulated in a Lagrangian framework such that numerical diffusion in the model transport scheme is sufficiently reduced. This feature makes the CLaMS model particularly advantageous for simulating trace gas transport in regions of strong transport barriers and of frequent occurrence of small-scale filamentary structures, such as in the stratosphere.

The specific work tasks of WP3400 are:

- Realisation of CLaMS model simulations of long-lived trace gases (N2O, CH4, CFC-11, CFC-12, SF6) over the past 40 years, driven by different meteorological reanalysis datasets.
- Compilation of CLaMS model-based long-lived trace gas climatologies as monthly and zonal mean data.
- Publication of these model-based climatologies in a suitable data archive.
- Comparison between long-lived trace gas climatologies from different model simulations and between model simulations and selected satellite observations.
- Comparison between long-lived trace gas climatologies and suitable stratospheric circulation diagnostics.
- Writing of technical report or journal manuscript.

Overall, the produced model simulations are expected to provide important new information on stratospheric circulation and transport. In particular, comparison to satellite observation data, as provided by other LOLIPOP work packages, will enable understanding of the relevant transport processes and related model deficits. In this way, the project results will be relevant for future improvement of climate models and reanalysis systems regarding the representation of the stratospheric circulation.

2 Approach/Methods

2.1 CLaMS model simulations

In this project, the CLaMS (Chemical Lagrangian Model of the Stratosphere) model was used to simulate the distribution of key trace gases in the stratosphere. CLaMS is a Lagrangian transport model, which represents atmospheric motion by following individual air parcels rather than computing tracer concentrations on a fixed grid. A particular advantage of this Lagrangian framework is its ability to minimize numerical diffusion, a common limitation in Eulerian models. As a result, CLaMS provides a more accurate representation of tracer transport, especially in regions characterized by strong transport barriers, such as around the polar vortex or at the edges of the tropical pipe, and in areas where frequent filamentation of air masses occurs, as is typical in the stratosphere.

Within this study, CLaMS was applied to simulate both the mean age of air, which serves as a proxy for the strength and structure of the Brewer–Dobson Circulation (BDC), and the distributions of several long-lived chemical tracers, including N_2O, CH_4, CFC-11, CFC-12, and SF_6. The analysis of the mean age of air enables an investigation of pure transport characteristics within the stratosphere, independent of chemical processes. In contrast, the simulated distributions of chemical tracers, many of which are also observed by satellite instruments, facilitate direct comparisons between model results and observational data. These comparisons are valuable for model validation, as well as for process-oriented studies aimed at improving the understanding of stratospheric transport and mixing processes.

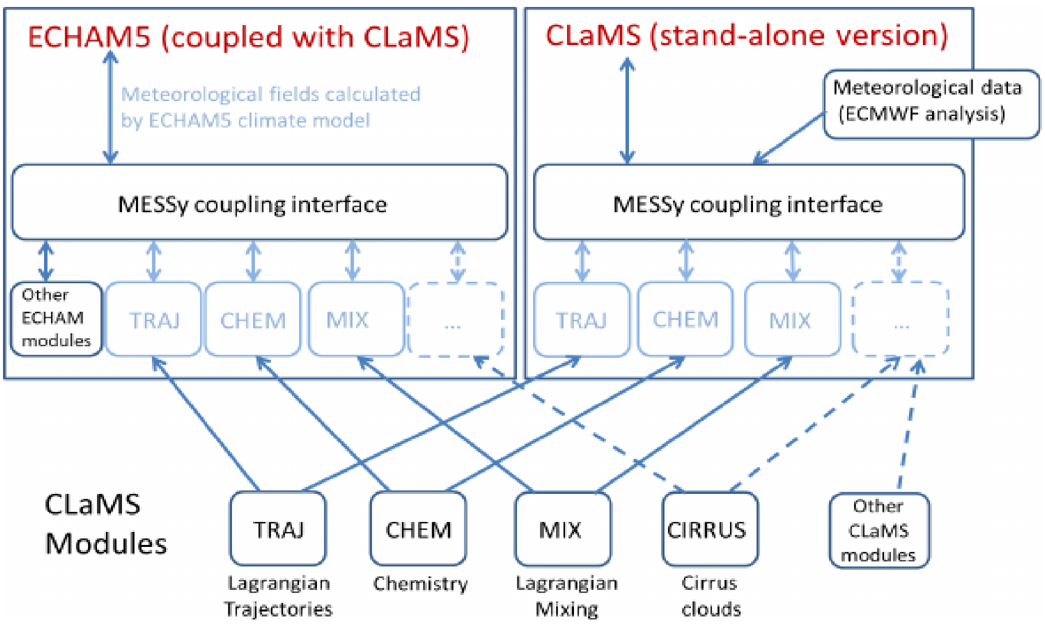

Figure 1: Schematic of the CLaMS modules integrated in the MESSy interface. (From Hoppe et al., 2014).

For the simulations conducted in this project, the CLaMS model was applied using the same configuration as detailed by Voet et al. (2025). The Lagrangian framework of CLaMS allows the model grid to adapt dynamically in space and time, enabling an accurate representation of small-scale mixing and transport processes. Figure 1 presents an overview of the different CLaMS modules for calculating advective transport, mixing, chemistry and microphysics.

In particular, the model was applied in “stand-alone” version (Fig. 1, right) driven by meteorological fields from several global reanalysis products (further details on the reanalyses used are given below). Simulations begin on 1 January 1979 (1 January 1980 for MERRA-2 reanalysis), preceded by a 20-year spin-up using repeating meteorology and boundary conditions from the first year. This procedure ensures dynamically and chemically equilibrated initial conditions for the transient simulations.

CLaMS employs a hybrid vertical coordinate system, transitioning from an orography-following coordinate in the troposphere to an isentropic coordinate in the stratosphere (e.g., Pommrich et al., 2014). Vertically, the model domain extends from the surface up to approximately the stratopause (~55 km or 2650 K).

A suite of long-lived trace gases is simulated, with particular relevance for this project given to SF6, CFC-11, CFC-12, CH4, and N2O. Observation-based, zonal-mean surface time series provide the lower boundary conditions for these species. Additionally, an idealized “clock tracer”, i.e., a passive tracer with a linearly increasing surface mixing ratio, is included to determine the mean age of air within the model environment.

Vertical cross-isentropic transport is derived from ERA5 diabatic heating rates following Ploeger et al. (2021). Convection is parameterized using convective available potential energy (CAPE) as a trigger for unresolved convective processes (Konopka et al., 2022). Further details on the model setup, chemical loss schemes, and validation against observations can be found in Voet et al. (2025).

2.2 Input data description

2.2.1 Reanalysis data

Atmospheric reanalysis is a method for reconstructing past atmospheric conditions by assimilating historical observations into a consistent numerical weather prediction model framework. This approach produces comprehensive, gridded datasets of atmospheric variables over extended time periods, providing a valuable resource for climate research, process studies, and model validation. Reanalysis datasets emerged in the 1990s in response to the growing need for long-term, homogeneous datasets that capture the state of the atmosphere consistently over decades. As such, they represent some of the most complete and reliable sources of information on past climate.

In this study, we make use of data from four global reanalyses: the Modern-Era Retrospective Analysis for Research and Applications, Version 2 (MERRA2), the Japanese 55-year Reanalysis (JRA55), and the ERA-Interim and ERA5 reanalyses from the European Centre for Medium-Range Weather Forecasts (ECMWF).

ERA5 is the fifth-generation ECMWF atmospheric reanalysis, covering the period from 1940 to the present. It features 137 vertical levels extending from the surface to 0.01 hPa, with roughly 45 levels representing the stratosphere. Vertical resolution gradually decreases with altitude, ranging from approximately 350 m in the lowermost stratosphere to about 1500 m at the top. ERA5 provides high-resolution spectral data with triangular truncation T639, corresponding to a horizontal resolution of roughly 0.28°, and is available at hourly intervals.

ERA-Interim, the predecessor of ERA5, was discontinued in 2019 and covers the period from 1 January 1979 to 31 August 2019. This older reanalysis was included in the inter-comparison to provide the basis for evaluating potential improvements in the newer ERA5 reanalysis. ERA-Interim has 60 vertical levels up to 0.1 hPa, with about 20 levels in the stratosphere. Vertical resolution ranges from ~1000 m in the lowermost stratosphere to ~3000 m at the uppermost levels. Its spectral resolution is T255, corresponding to a horizontal resolution of ~0.70°, and data are available at 6-hour intervals.

JRA55 spans the period from 1958 until its discontinuation in January 2024. Similar to ERA-Interim, it has 60 vertical levels up to 0.1 hPa, with roughly 20 levels in the stratosphere and a comparable vertical resolution. Its spectral resolution is T319, giving a horizontal resolution of ~0.56°, and the data are available at 6-hour intervals.

MERRA2 covers 1980 to the present and features 72 vertical levels from the surface to 0.01 hPa, about 25 of which are in the stratosphere. Vertical resolution ranges from ~1000 m in the lowermost stratosphere to ~2000 m at the top. Its native horizontal resolution is 0.5° × 0.625°, and three-dimensional products are available at 3-hour intervals.

Among these four reanalyses, ERA5 is the most recent and offers the highest spatial and temporal resolution, making it particularly suitable for studies requiring fine-scale representation of atmospheric processes.

2.2.2 Trace gas boundary conditions

The lower boundary conditions for all trace gases in the CLaMS model simulations are specified using observation-based, zonal-mean surface time series derived primarily from long-term, ground-based in-situ measurements conducted by the Global Monitoring Laboratory (GML) of the National Oceanic and Atmospheric Administration (NOAA). These datasets capture the temporal evolution of surface mixing ratios across different latitudes, providing a consistent and realistic representation of tropospheric sources for the model tracers (SF6, N2O, CFC-11, CFC-12, CH4).

Upper boundary conditions are defined based on the individual chemical and physical properties of each tracer. For SF6, the upper boundary is constrained by monthly zonal-mean time series derived from measurements by the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) aboard the ESA Environmental Satellite (Envisat) (Fischer et al., 2008). These observations capture the pronounced seasonal and latitudinal variability of SF6 mixing ratios near the stratopause, where mesospheric loss processes begin to be significant. For periods beyond the MIPAS observation window, the upper boundary is extrapolated using a sinusoidal least-squares fit that reproduces the observed seasonal cycle and latitudinal gradients, adjusted according to zonal-mean age-of-air estimates at the 2500K potential temperature level.

For CH4, the upper boundary is based on the climatological mean seasonal cycle derived from the Halogen Occultation Experiment (HALOE) (Grooss and Russel, 2005), representing the long-term distribution of methane in the upper stratosphere and lower mesosphere. N2O follows the upper boundary conditions provided by the Mainz photochemical two-dimensional (2-D) model, ensuring a physically consistent transition to the overlying region where N2O undergoes rapid photochemical destruction. In contrast, CFC-11 and CFC-12 are assumed to be fully decomposed well below the model top; their upper boundary mixing ratios are therefore set to zero throughout the simulation, consistent with previous CLaMS configurations and laboratory-derived lifetime estimates (for further details on boundary conditions see Pommrich et al., 2014).

At the start of each simulation time step, the values from both the lower and upper boundary conditions are interpolated onto the Lagrangian air parcels within the respective boundary layers of CLaMS, ensuring a spatially smooth and temporally consistent representation of tropospheric and stratospheric tracer input throughout the model domain.

2.3 Compilation of climatologies

For this project, comprehensive climatologies were compiled from the several CLaMS model simulations driven by the different meteorological reanalyses. Since CLaMS is a Lagrangian transport model, its instantaneous output - originally provided on an irregularly spaced set of air parcels - was interpolated onto a regular latitude–longitude–level grid to enable consistent climatological analysis. Two types of vertical coordinates were used for this gridding: pressure levels and isentropic (potential temperature) levels. The pressure level grid spans from 1000 hPa down to 0.10 hPa, covering 39 levels, while the isentropic grid includes 30 levels from 280 K to 2600 K. The horizontal (latitude-longitude) resolution for all interpolations is 1° × 1°.

Interpolation from the irregular Lagrangian positions to the regular grid was performed using a weighted nearest-neighbor approach based on Delaunay triangulation, ensuring accurate spatial representation of model output. For each reanalysis-driven simulation, the meteorological variables used in the interpolation step were consistently taken from the corresponding reanalysis dataset to maintain internal consistency between dynamics and tracer transport.

Following the interpolation, monthly mean fields were calculated for all variables, which were then further averaged zonally to derive zonal mean climatologies. These monthly mean and zonal mean datasets were compiled into NetCDF files for each simulation year. The time coverage of the simulations varies depending on the availability of the underlying reanalysis data: ERA5-driven simulations cover the period 1979-2023, ERA-Interim covers 1979-2019, MERRA-2 spans 1980-2017, and JRA-55 covers 1979-2017. For the comparison presented here, all climatologies have been calculated over the common period 2004-2017.

The complete set of climatological data products, including pressure-level and isentropic-level climatologies for all reanalysis cases, has been published and is openly available on the Zenodo data repository at https://zenodo.org/records/17357000.

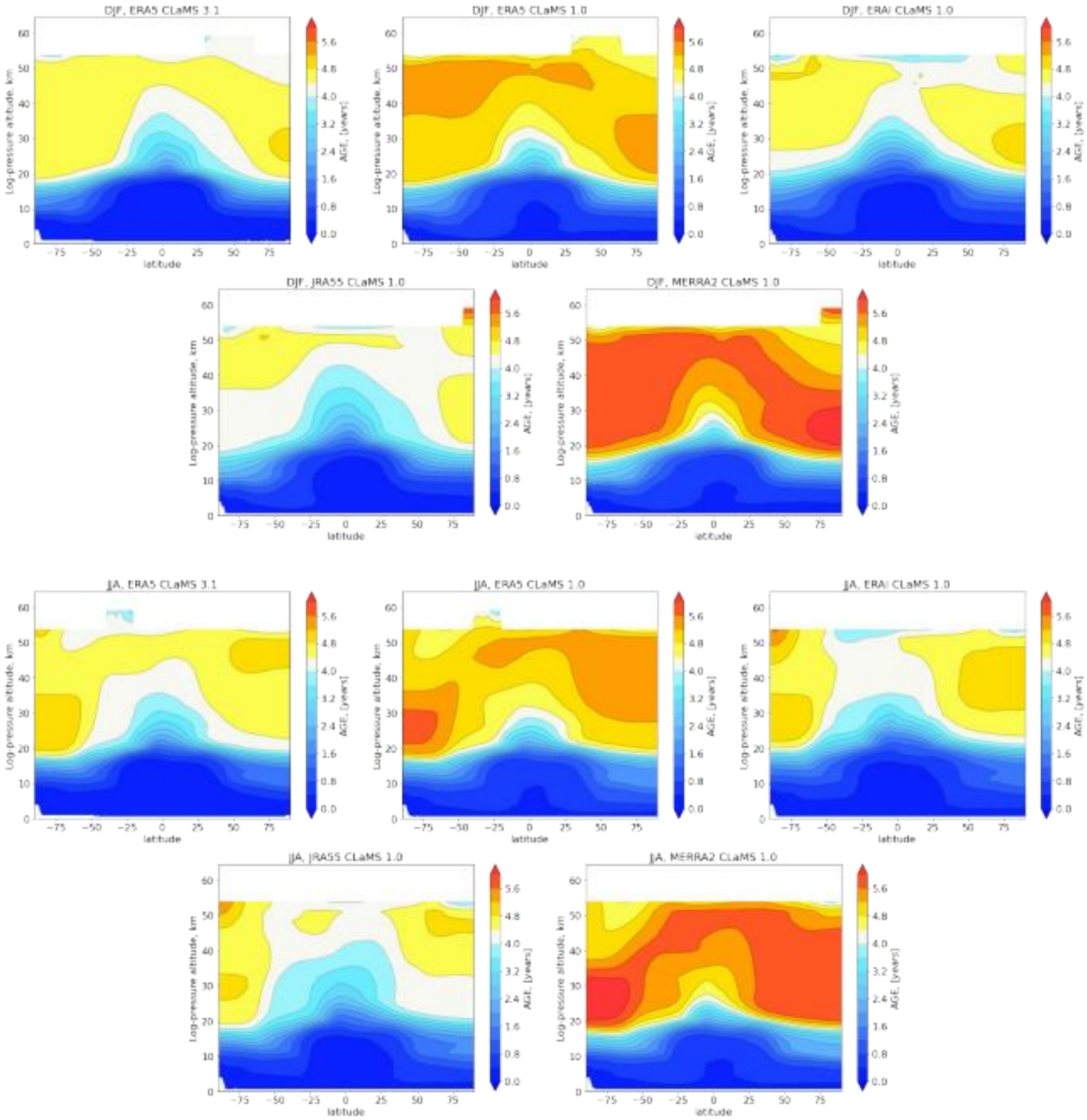

Figure 2: Climatological zonal mean distributions of mean age of air from the different CLaMS model simulations for boreal winter (December-February, DJF, top panel) and boreal summer (June-August, JJA, bottom panel). White areas above about 50km are above the model top (which is approximately at the stratopause).

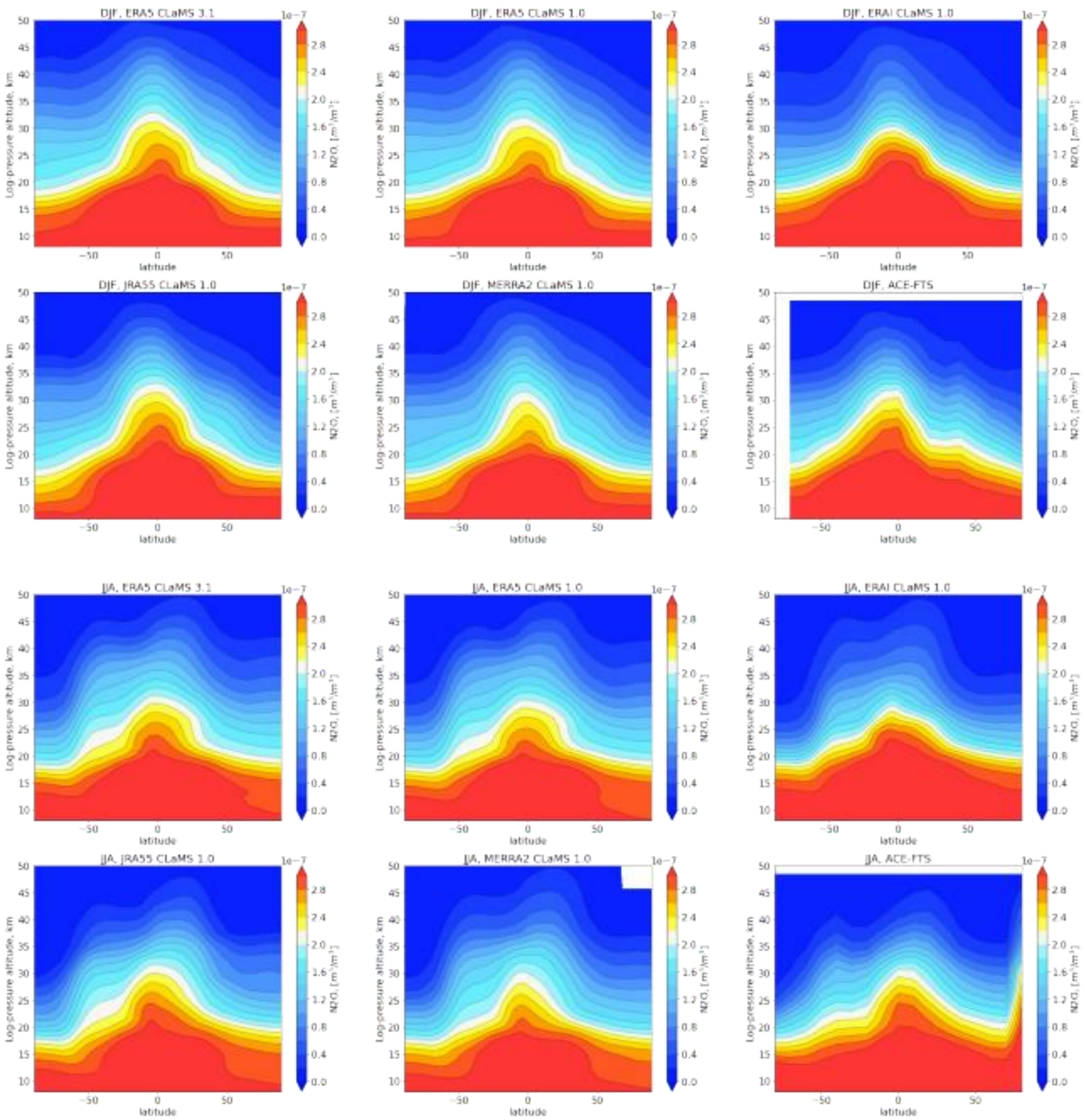

Figure 3: Climatological zonal mean distributions of nitrous oxide (N2O) from the different CLaMS model simulations and ACE-FTS satellite observations for boreal winter (December-February, DJF, top panel) and boreal summer (June-August, JJA, bottom panel). The last subplot in the top and bottom panel shows the satellite observation data.

3 Preliminary results

3.1 The stratospheric circulation in reanalyses

The mean age of air serves as a widely used proxy for assessing the strength and speed of the stratospheric circulation, the BDC. Figure 2 shows the mean age from the different CLaMS model simulations driven with different reanalyses (ERA5, ERA-Interim, MERRA2, JRA55) and using different parameterizations for convective and mixing transport (v1.0 vs. v3.1). Despite differences among model parameterizations and reanalysis products, the overall spatial pattern of the age of air distribution is remarkably robust, reflecting the overall pattern of the stratospheric BDC. In all datasets, relatively young air masses are found in the troposphere and within the tropical pipe region of the lower stratosphere, consistent with strong upwelling from the troposphere into the lower stratosphere in the tropics. In contrast, the oldest air is typically observed within the wintertime polar vortex, where isolation and weak mixing allow air parcels to age considerably, most prominently in the Southern Hemisphere, where the vortex is stronger and more persistent.

While this large-scale structure is consistent, substantial variations exist in the absolute magnitude of mean age, reflecting differences in the simulated speed of the BDC. Among commonly used reanalyses, JRA-55 exhibits the fastest circulation, whereas MERRA-2 shows the slowest, leading to discrepancies in mean age of more than one year, consistent with recent findings (Ploeger et al., 2021).

In addition to these differences in large-scale dynamics, also the parameterization of unresolved transport, here small-scale mixing and convection, affects the mean age distribution. Improved representations of these processes can substantially alter the simulated circulation speed and tracer transport efficiency. Comparison of the new and improved parameterization scheme in CLaMS v3.1 (see Konopka et al., 2021) with the control set-up in CLaMS v1.0 shows clear differences. In particular, the improved convection and mixing parameterizations make the stratospheric air masses younger of up to about 0.5 years. These differences underline the importance of both resolved dynamics and subgrid-scale processes in determining the simulated age of air and, consequently, the inferred strength of the BDC.

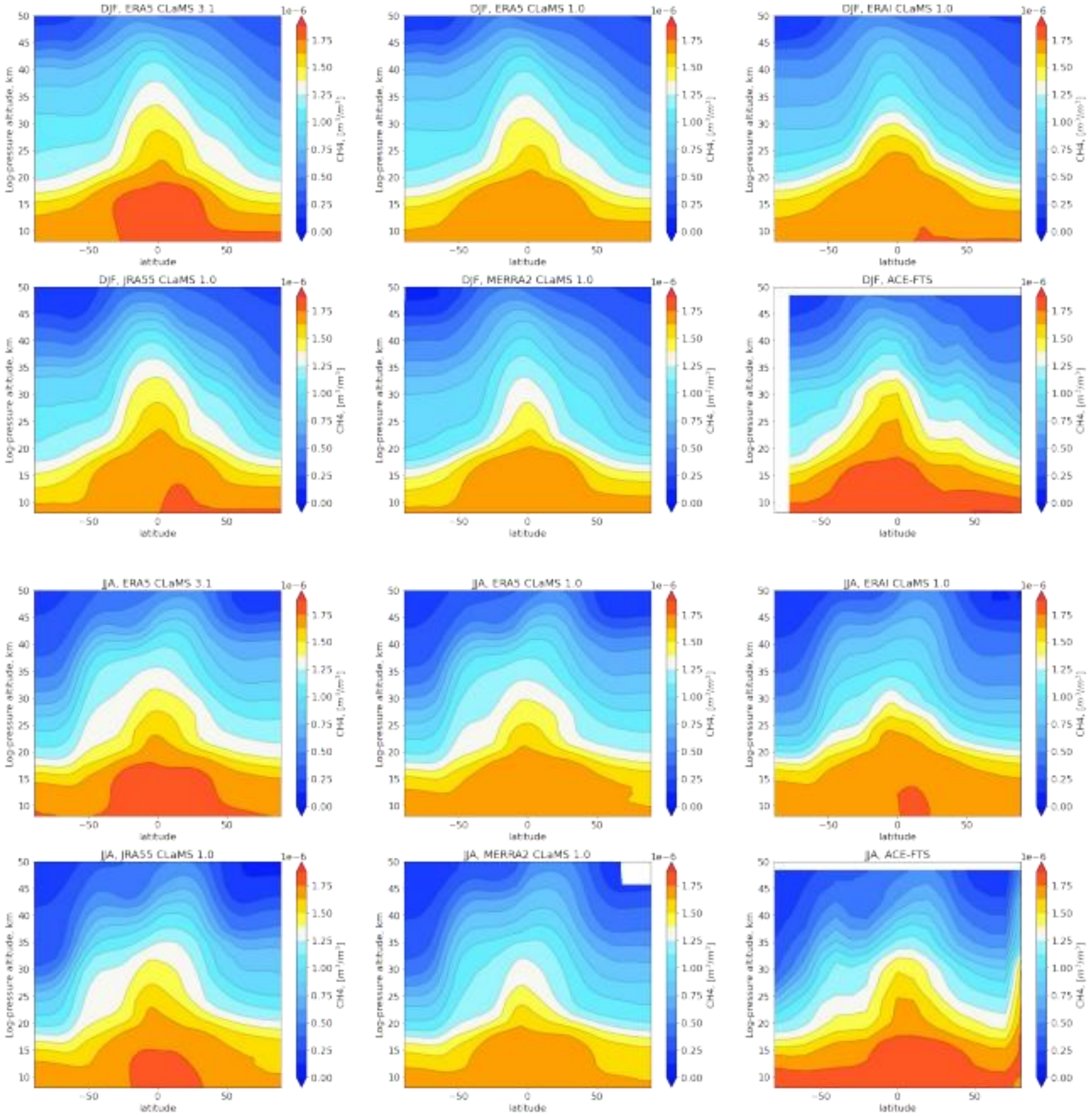

Figure 4: Climatological zonal mean distributions of methane (CH4) from the different CLaMS model simulations and ACE-FTS satellite observations for boreal winter (December-February, DJF, top panel) and boreal summer (June-August, JJA, bottom panel). The last subplot in the top and bottom panel shows the satellite observation data.

3.2 Trace gas distributions

The stratospheric distributions of the long-lived trace gas species (here N2O, CH4, CFC-11, CFC-12, SF6) show the influence of the BDC. Therefore, characteristics of the BDC can be inferred from distributions of long-lived tracers. Stratospheric distributions for N2O are shown in Figure 3, for CH4 in Figure 4 and for SF6 in Figure 5. Note that the distributions for CFC-11 and CFC-12 are not shown here explicitly for conciseness, but are largely consistent with the distributions for the species shown. Comparison of the N2O distribution (Figure 3) to mean age (Figure 2) reveals the effects of stratospheric transport on the tracer distribution. In particular, high mixing ratios in the troposphere and in the tropical pipe coincide with low values of mean age, indicating the upwelling of young, tropospheric air into the tropical lower stratosphere. Old air masses in the polar lower stratosphere (high age of air values), in turn, are characterized by low mixing ratios of N2O. A very similar picture emerges for CH4 (Figure 4) and SF6 (Figure 5), and also for CFC-11 and CFC-12 (not shown).

Comparison between the different simulations driven with different reanalyses and including different parameterizations shows similar differences as for mean age. Hence, simulation of stratospheric trace gas distributions critically depends on the representation of both large-scale and small-scale (parameterized) stratospheric transport processes in the models. Differences in large-scale transport (here reanalysis differences) appear to cause larger differences in trace gas distributions throughout the stratosphere, while the effects of differences in small-scale transport (here between CLaMS 3.1 and 1.0) are largely limited to the upper troposphere and lower stratosphere region.

Furthermore, comparison between the model simulated trace gas distributions to ACE-FTS satellite observations allows assessing the reliability of the specific model simulation. Comparison between simulated and observed CH4 distributions in Figure 4 (and likewise for the other species) shows that the slow BDC in MERRA2 reanalysis (high mean age values) results in too low mixing ratios of tracers with tropospheric source. Hence, MERRA2 seems to have a too slow BDC. Among all considered cases, the model version driven with ERA5 reanalysis and including the improved small-scale mixing and convection parameterizations simulates trace gas distributions in closest agreement with the observations and therefore appears to have the most reliable representation of stratospheric transport

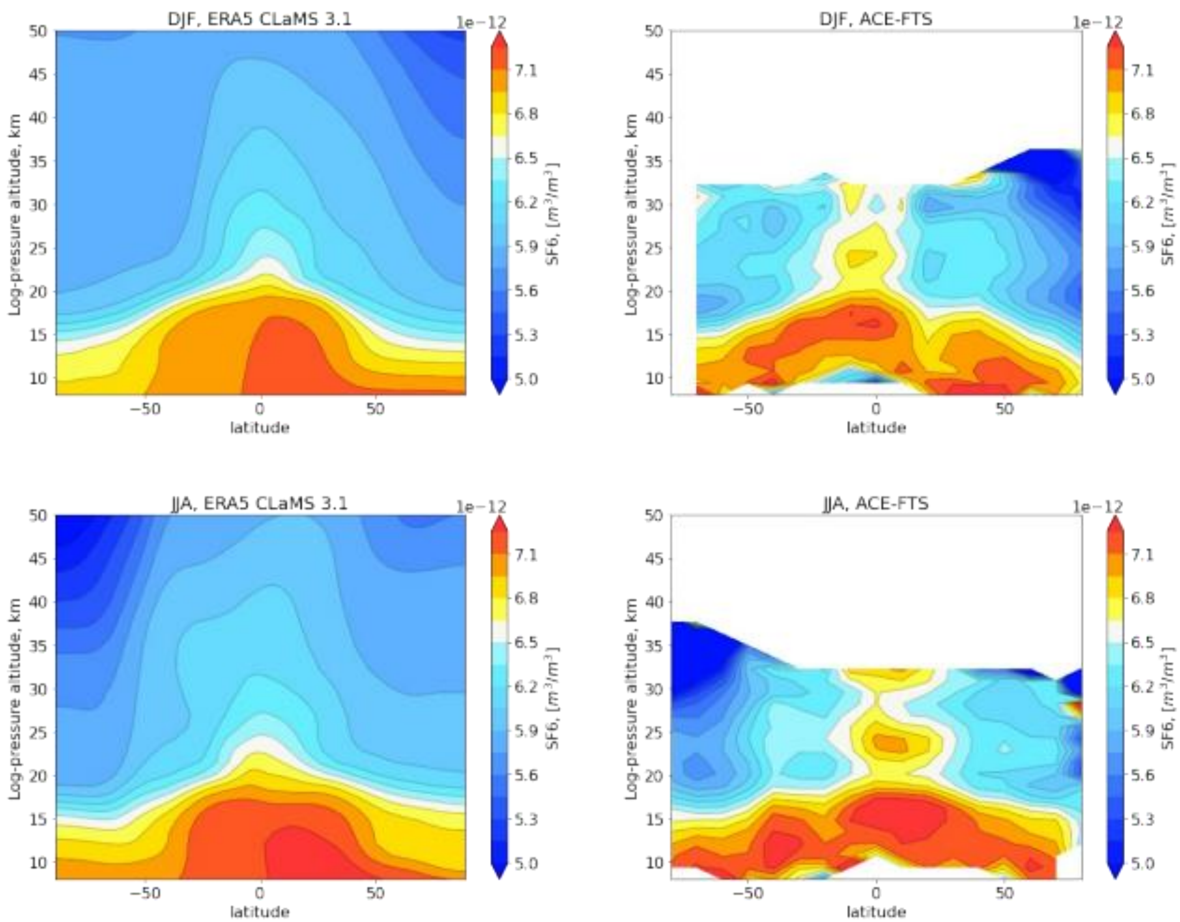

Figure 5: Climatological zonal mean distributions of sulfur hexafluoride (SF6) from the CLaMS v3.1 model simulation and from ACE-FTS satellite observations for boreal winter (December-February, DJF, top panel) and boreal summer (June-August, JJA, bottom panel). The right subplot in the top and bottom panel shows the satellite observation data.

4 Outlook

- Usage of CLaMS simulation data for proof of concept for new method to calculate mean age from trace gas distributions (Voet et al., 2025).
- Application of new mean age calculation method to ACE-FTS satellite observation data.
- Investigation of stratospheric circulation changes based on the comparison of this new CLaMS and ACE-FTS mean age product.
- Summarizing results and writing peer-reviewed publication and/or update of summary report.

List of acronyms and abbreviations

ACE/FTS	Atmospheric Chemistry Experiment – Fourier Transform Spectrometer

BDC	Brewer-Dobson Circulation
CLaMS	Chemical Lagrangian Model of the Stratosphere
ECMWF	European Centre for Medium-Range Weather Forecasts
ERA-Interim	European Centre for Medium-Range Weather Forecasts Interim Reanalysis
ERA5	European Centre for Medium-Range Weather Forecasts Reanalysis version 5
ESA	European Space Agency
GHG	GreenHouse Gas
GML	Global Monitoring Laboratory
HALOE	Halogen Occultation Experiment
JRA55	Japanese 55-year Reanalysis
LOLIPOP	Long Lived greenhouse gas PrOducts Performances
MERRA-2	Modern-Era Retrospective analysis for Research and Applications, Version 2
MIPAS	Michelson Interferometer for Passive Atmospheric Sounding
NOAA	National Oceanic and Atmospheric Administration

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