The impact of SF$_6$ sinks on age of air climatologies and trends

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Abstract.

Mean age of air (AoA) is a common diagnostic for the strength of the stratospheric overturning circulation in both climate models and observations. AoA climatologies and its trends over the recent decades of model simulations and proxies derived from observations of long-lived tracers do not agree. Satellite observations show much older air than climate models and while most models compute a clear decrease of AoA over the last decades, a thirty-year timeseries from measurements shows a statistically non-significant positive trend. Measurement-based AoA derivations are often based on observations of the trace gas SF$_6$, a fairly long-lived gas with a near-linear increase of emissions during the recent decades. However, SF$_6$ has chemical sinks in the mesosphere, which are not considered in most model studies. In this study, we explicitly compute the chemical SF$_6$ sinks based on chemical processes in the global chemistry-climate model EMAC. We show that good agreement of stratospheric AoA in EMAC and MIPAS is reached through the inclusion of chemical SF$_6$ sinks, as those lead to a strong increase of the stratospheric AoA and thereby to a better agreement with MIPAS satellite observations. Remaining larger differences in high latitudes are addressed and possible reasons are discussed. Subsequently, we demonstrate that also the AoA trends are strongly influenced by the chemical SF$_6$ sinks. Under consideration of the SF$_6$ sinks, the AoA trends over the recent decades reverse sign from negative to positive. We conduct sensitivity simulations which reveal that this sign reversal results neither from trends of the stratospheric circulation strength, nor from changes in the strength of the SF$_6$ sinks. We illustrate that even a constant SF$_6$ destruction rate causes a positive trend in the derived AoA, since the amount of depleted SF$_6$ scales with the increasing SF$_6$ abundance itself. In our simulations, this effect overcompensates the impact of the accelerating stratospheric circulation which naturally decreases AoA. Although various sources of uncertainties cannot be quantified in detail in this study, our results suggest that the inclusion of SF$_6$ depletion in models has the potential to reconcile the AoA trends of models and observations.

1 Introduction

The Brewer-Dobson circulation (BDC) describes the stratospheric transport circulation, consisting of the mean overturning circulation of air ascending in the tropical pipe, moving poleward and descending in the extratropics (Brewer, 1949; Dobson and Massey, 1956), as well as isentropic mixing. A good measure to diagnose this transport circulation is the age of stratospheric

air (AoA), which is defined as the mean transport time of an air parcel from its entry into the stratosphere (or from the surface) to any point therein (Hall and Plumb, 1994; Waugh and Hall, 2002). In general circulation models (GCMs), AoA is commonly represented by an inert tracer with a strictly linear temporally increasing surface mixing ratio and is calculated as the corresponding time lag between the local mixing ratio and the mixing ratio at a reference point (Hall and Plumb, 1994). The same method can be applied to real long-lived tracers with a linear trend in tropospheric concentration and AoA has been derived, for example, from balloon-borne in situ measurements of sulphur hexafluoride (SF_6) (Andrews et al., 2001; Engel et al., 2009, 2017). This trace gas is particularly suitable for these studies as it is stable in the troposphere and stratosphere and its tropospheric concentrations increased nearly linearly over the recent decades. Together with observations of other trace gases, these measurements form a long-term dataset of observationally based AoA restricted to the Northern Hemisphere mid-latitudes that covers more than 40 years. A near global dataset of AoA covering ten years from 2002-2012 was derived in Stiller et al. (2012), Haenel et al. (2015) and Stiller et al. (2017), who retrieved SF_6 distributions from MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) satellite observations, but these cover a much shorter time period.

Observations and model simulations of AoA often disagree. AoA derived from observations is mainly older than simulated AoA (see e.g. SPARC, 2010; Dietmüller et al., 2018) and the AoA trend over recent decades even differs in sign between observations and models. While most climate models show a clear decrease of AoA over time (see e.g. Butchart and Scaife, 2001; Garcia et al., 2011; Eichinger et al., 2019), consistent with the simulated acceleration of the BDC in the course of climate change (see e.g. Garcia and Randel, 2008), the time series of the observations presented in the studies by Engel et al. (2009), Ray et al. (2014) and Engel et al. (2017) show a (statistically non-significant) positive trend. This discrepancy has been addressed in numerous studies: Garcia et al. (2011) showed that due to the concave growth rate of tropospheric SF_6 concentrations, the AoA trends derived from an SF_6 tracer are smaller than the trends derived from a synthetic, linearly growing AoA tracer (also after accounting for the nonlinear growth rates of SF_6). They noted that the sparse sampling of in situ observations can be the reason for the above mentioned trend discrepancies. Birner and Bönisch (2011) as well as Bönisch et al. (2011) argued that differences in the changes between the deep and the shallow BDC branch can possibly explain these. Ploeger et al. (2015) showed that the residual circulation transit time cannot explain the AoA trends, and that the integrated effect of mixing (which is coupled to residual circulation changes, see Garny et al., 2014) is crucial. Moreover, Stiller et al. (2017) could explain a hemispheric asymmetry by a shift of subtropical transport barriers. However, a comprehensive explanation for the trend differences between models and observations is still missing.

SF_6 sinks lead to older apparent AoA, as well as shorter lifetimes. Leedham Elvidge et al. (2018) evaluated AoA from several tracers including SF_6 and found clear differences between these, which indicated a shorter SF_6 lifetime than previously assumed. The strongest chemical SF_6 removal reactions take place in the mesosphere, the most important removal processes are electron attachment and UV-photolysis, but these processes have not yet been precisely quantified. Ravishankara et al. (1993) estimated an SF_6 lifetime of 3200 years and Reddmann et al. (2001) found a lifetime between 400 and 10000 years, depending on the assumed loss reactions and electron density. A more recent model study by Kovács et al. (2017) reported a mean SF_6 lifetime of 1278 years and Ray et al. (2017) provided a range between 580 and 1400 years based on in situ measurements in the stratospheric polar vortex. The most recent study of Kouznetsov et al. (2020) shows an SF_6 lifetime ranging between 600

and 2900 years. Due to these uncertainties and the complex computation of the chemical reactions, most model studies do not
60 consider any SF_6 sinks for the calculation of AoA from SF_6 mixing ratios. This can explain why most climate models generally
show younger stratospheric air than observations, in particular within the polar vortices (e.g. Haenel et al., 2015; Ray et al.,
2017).

In the present study, we apply the chemistry climate model EMAC (ECHAM MESSy Atmospheric Chemistry, Jöckel
et al., 2010; Jöckel et al., 2016). It uses the second version of the Modular Earth Submodel System (MESSy2) to link multi-
65 institutional computer codes. In our simulations, we employed the MESSy submodul "SF6" which explicitly calculates SF6
sinks based on physical processes (based on Reddmann et al., 2001), rather than on crude parameterisations. In Sect. 2 we
describe the EMAC model and the SF6 submodel as well as the observational data we use for comparison. Sect. 3 contains a
comparison of the EMAC climatologies with MIPAS data, a comparison of the EMAC trends with MIPAS and balloon borne
measurements and an analysis of the results of two sensitivity simulations. In Sect. 4, we discuss the results and provide some
70 concluding remarks.

2 Atmospheric Model

2.1 EMAC model

We use the EMAC (ECHAM MESSy Atmospheric Chemistry, v2.54.0, Jöckel et al., 2010; Jöckel et al., 2016) model in
a T42 horizontal ($\sim$ 2.8°x2.8°) resolution with 90 layers in the vertical and explicitly resolved middle atmosphere dynamics
75 (T42L90MA). In this setup, the uppermost model layer is centred at 0.01 hPa and the vertical resolution in the upper troposphere
lower stratosphere region (UTLS) is 500-600 m. In the standard reference setup, we use the basic EMAC modules for dynamics,
radiation, clouds, and diagnostics (AEROPT, CLOUD, CLOUDOPT, CVTRANS, E5VDIFF, GWAVE, ORBIT, OROGW,
PTRAC, QBO, RAD, SURFACE, TNUDGE, TROPOP, VAXTRA, refer to Jöckel et al., 2005; Jöckel et al., 2010, for details
on these submodels). Additionally we included the new submodel SF6.

80 ### 2.2 Submodel SF6

The submodel SF6 is used to calculate the lifetime of SF_6 by explicitly accounting for the sinks of SF_6 in the mesosphere. The
calculation method for this is based on the reaction scheme of Reddmann et al. (2001). The most important reaction involved
in the chemical degradation of SF_6, namely electron attachment, is included in the SF6 submodel. The configuration of the
submodel allows for a simple exponential profile for the electron field and a more complex field based on Brasseur and Solomon
85 (1986), whereas in the present study we use the latter option. It depends on altitude, latitude, solar zenith angle, air density and
day of year (see Fig. S1 in Supplementary Information). In contrast to Reddmann et al. (2001), UV-photolysis of SF_6 is not
included in the submodel. The loss rate by photolysis is several orders of magnitude weaker than that of electron attachment
up to altitudes of about 100 km (see e.g. Fig. 9 in Totterdill et al., 2015) and is therefore not relevant for the focus of our study.
Further reactions considered are photodetachment of SF_6^- (Datskos et al., 1995), destruction of SF_6^- by atomic hydrogen,

Table 1. Chemical reactions of SF_6 used in the SF6 submodel. The labelling of the various reactions mirrors the style used by Reddmann et al. (2001).

Reaction No.	Reaction	Description
(R1)	$SF_6 + h\nu \longrightarrow SF_5 + F$	destructive UV-photolysis
(R2)	$SF_6 + e^- \longrightarrow (SF_6^-)^*$ $SF_6 + O^+ \longrightarrow SF_5^+ + OF$ $SF_6 + N_2^+ \longrightarrow SF_5^+ + NF$ $SF_6 + O_2^- \longrightarrow SF_6^- + O_2$	destructive Electron Attachment and Secondary Reactions
(R3)	$SF_6^- + h\nu \longrightarrow SF_6 + e^-$	photodetachment
(R4)	$SF_6^- + H \longrightarrow SF_5^+ + HF$	destructive
(R5)	$(SF_6^-)^* + M \longrightarrow SF_6^-$	stabilisation against autodetachment
(R6)	$(SF_6^-)^* \longrightarrow SF_6 + e^-$	autodetachment
(R7)	$SF_6^- + HCl \longrightarrow$ products $SF_6^- + HNO_3 \longrightarrow$ products $SF_6^- + SO_2 \longrightarrow$ products	destructive
(R8)	$SF_6^- + O_3 \longrightarrow SF_6 + O_3^-$ $SF_6^- + O \longrightarrow SF_6 + O^-$ $SF_6^- + NO_2 \longrightarrow SF_6 + NO_2^-$	recovery reaction

90   hydrogen chloride and ozone (Huey et al., 1995), stabilisation of excited SF_6^- by collisions and autodetachment of SF_6^-. An overview is provided in Table 1. Reddmann et al. (2001) used climatological profiles for the forementioned gases, while in our submodel channel objects (see Jöckel et al., 2016) are used. Such channel objects can be calculated in other submodules (e.g. interactive chemistry), prescribed as external time series (in this study) or just be simple climatologies. The autodetachment rate can be chosen in the namelist and was set to $10^6 s^{-1}$ (see Reddmann et al., 2001). For the relative importance of the
95   contributions of the various reactions see Fig. S1 in the Supplementary Information.

2.3   Simulation Setup

Other than the SF6 submodel, no interactive chemistry is activated in the simulations for this study. The reactant species involved in the SF_6 chemistry and the radiatively active gases (CO_2, CH_4, N_2O, O_3) are transiently prescribed from the ESCiMo RC1-base-07 simulation (see Jöckel et al., 2016) as monthly and zonal means. Moreover, we prescribe the Hadley
100   Centre Sea Ice and Sea Surface Temperature data set (HADISST), the CCMI-1 volcanic aerosol dataset (for its effect on infrared radiative heating, see Arfeuille et al., 2013; Morgenstern et al., 2017) and QBO nudging (see Jöckel et al., 2016). Our simulations range from 1950 to 2011, whereas at least the first ten years have to be considered as a spin-up period. The projection simulation runs from 1950 to 2100 with the SF_6 reactant species and GHGs prescribed from the ESCiMo RC2-

base-04 simulation (see Jöckel et al., 2016) as monthly and zonal means. In addition to the reference (REF) simulation, we
performed two sensitivity simulations and one specified dynamics simulation. The two sensitivity simulations are as follows:
The CSS (constant reaction partners for SF_6 sinks) sensitivity simulation differs from the REF simulation only by constant
mixing ratios of the reactant species (see above) that influence the SF_6 sinks. For that purpose, we kept the mixing ratios
at the level of the start of the simulation (year 1950 on repeat). The second sensitivity simulation, referred to as TS2000, is
not a transient simulation as is the REF simulation, but instead a time slice simulation with climate conditions (GHGs, SSTs
and SICs) of the year 2000 (climatology of the period 1995-2004). Furthermore, the reactant species for the SF6 sinks have
been averaged over the period 1995-2004 for the TS2000 simulation. In the specified dynamics (SD) simulation, we apply
newtonian relaxation ('nudging') towards ERA-INTERIM (Dee et al., 2011) reanalysis data of potential vorticity, divergence,
temperature and the logarithmic surface pressure up to 1 hPa. This assures that the meteorological situation largely resembles
the ERA-Interim data. The flexible structure of the Modular Earth Submodel System (MESSy, Jöckel et al., 2005) allows us
to use the same executable for all simulations, with the differences between them realised through changes in aforementioned
namelist settings (see Jöckel et al., 2005). A summary of the simulations used in this study can be found in Table 2.

2.4 Satellite and in situ data

The MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) instrument on Envisat (Environmental Satel-
lite) allowed for the retrieval of SF_6 by measuring the thermal emission in the mid-infrared, while orbiting the Earth sun-
synchronously 14 times a day. This high-resolution Fourier transform spectrometer measured at the atmospheric limb and
provided data for SF_6 retrievals in full spectral resolution from 2002 to 2004 and in reduced resolution from 2005 to 2012
between 6 and 40 km of altitude (Stiller et al., 2012; Haenel et al., 2015). In this study, a newer version of the MIPAS dataset
existing as of 2019 will be shown, whereby new SF_6 absorption cross-sections have been used for the SF_6 retrieval (Stiller
et al., 2020; Harrison, 2020). Except for the newer absorption cross-sections for SF_6 and the accounting for a trichlorofluo-
romethane (CFC11) band in the vicinity of the SF_6 signature, the SF_6 retrieval and conversion into AoA was done according
to the description by Haenel et al. (2015). In particular, the Level-1b data version is still V5.

Engel et al. (2009) collected available air samples of SF_6 and CO_2 from balloon-borne cryogenic whole air-samplers flown
during 27 balloon flights, with data up to 43 km, and re-analysed these samples in a self-consistent manner. The derived SF_6
data cover the years 1975 to 2005 (with a gap between 1985 and 1994) and covers the mid-latitudes between 32°N and 51°N.
Since AoA profiles from midlatitudes above approximately 25 km or 30 hPa turned out to be constant over altitude, a mean
mid-latitude middle stratospheric AoA value from each profile was determined by averaging the vertical profile between 30 hPa
and the top balloon flight height. With this procedure, a time series of mid-latitude middle stratospheric AoA values could be
determined back to 1975. It is important to note that part of the Engel et al. (2009) AoA time series is derived from CO_2
measurements. In this paper, only the AoA data points derived from SF_6 are used. Engel et al. (2017) extended the initial
dataset from Engel et al. (2009) to 2016, however the AoA here is derived from CO_2 measurements and thus also excluded.

Table 2. Overview of simulations undertaken in this study

Simulation	Details
Reference (REF)	Transient 1950 - 2011 Greenhouse gases (CO_2, CH_4, N_2O, O_3) transiently prescribed from ESCiMo RC1-base-07-simulation (Jöckel et al., 2016) as monthly and zonal means
Specified Dynamics (SD)	Transient 1980 - 2011 Newtonian relaxation of dynamics towards ERA-Interim reanalysis data (Dee et al., 2011) up to 1hPa
Sensitivity Simulations	
Constant reaction partners for SF_6 Sinks (CSS)	Transient 1950 - 2011 Same conditions as the REF simulation, but for the SF_6 reactant species the year 1950 concentrations are repeated throughout the model run
Time slice (TS2000)	Time slice 1950 - 2011 Climate conditions (GHGs, SSTs, SICs) of year 2000 Climatology taken as 1995 – 2004 SF_6 sinks reactant species averaged over 1995 – 2004
Projection Simulation	
Climate Projection (PRO)	Transient 1950 - 2099 Greenhouse gases (CO_2, CH_4, N_2O, O_3) transiently prescribed from ESCiMo RC2-base-04-simulation (Jöckel et al., 2016) as monthly and zonal means

2.5 Analysis method

The basic concepts for the calculation of mean AoA are introduced in Hall and Plumb (1994). In the case of a tracer with a linear increasing lower boundary condition, AoA can be determined by the time lag between the mixing ratio at a given point in the atmosphere and the same mixing ratio of the reference time series. As for any realistic tracer, SF_6 does not exhibit perfectly

140   linear growth, for which adjustments in the AoA calculation are needed. This study follows the calculation method employed by e.g. Engel et al. (2009), which was introduced in Volk et al. (1997). The calculation uses a polynomial fit to the reference time series to approximate mean AoA. However, in our study we modified the parameters compared to those used in Engel et al. (2009) to ensure that (passive) SF_6-based AoA agrees with the ideal AoA derived from the linear tracer, following Fritsch et al. (2019). Specifically, in our calculations we used a ratio of moments of 1.0 years and a fraction of input of 95 %. Further,

145   we used the SF_6 mixing ratio averaged over 20°S-20°N at the ground as the reference time series. Due to the availability of data, this reference region is also used in Engel et al. (2009) with balloon-borne oberservations.

For the derivation of AoA from MIPAS SF_6 observations, Stiller et al. (2008, 2012) and Haenel et al. (2015) used a slightly smoothed version of the global mean of SF_6 surface measurements as the reference time series instead of SF_6 at the stratospheric entry point, which is not available from observations (see e.g. Dlugokencky, 2020). The non-linearity of the reference

150   curve was considered by its convolution with an idealized age spectrum parameterized as a function of the mean age within an iterative approach. For more details, see Stiller et al. (2012) and Haenel et al. (2015).

In our simulations the AoA calculations are applied to a total of four tracers, which can be organised into two groups. The first assumes a strict linear growth of SF_6, producing a linear reference curve, while the second considers a realistic growth of SF_6 based on observed emissions, creating a non-linear SF_6 reference curve. Technically, these "emissions" are realised via

155   lower boundary conditions in our simulations. As previously mentioned, SF_6 undergoes chemical degradation predominantly in the mesosphere. Consequently, the absence or presence of mesospheric sinks is additionally considered, resulting in a total of four tracers tr(WS, SF_6), tr(NS, SF_6), tr(WS, lin), and tr(NS, lin). The labelling of these depends on the chemistry involved (with sinks: "WS", without (no) sinks: "NS") and the growth assumed (linear: "lin", non-linear: "SF_6"), and follows the pattern $tr(\text{CHEMISTRY, GROWTH})$. When referring to simulations with a specific tracer, the labelling will follow the

160   notation SIMULATION(CHEMISTRY, GROWTH), and similarily we use the following notation for AoA inferred from the tracer used in a simulation: $\text{AOA(CHEMISTRY, GROWTH)}_{SIM}$. For example: AoA from the reference simulation based on the tracer with mesospheric sinks and non-linear (SF_6 emission based) increase is referred to as $\text{AoA(WS, SF}_6)_{REF}$. AoA inferred from the linear tracer without mesospheric sinks in the reference simulation is denoted as $\text{AoA(NS, lin)}_{REF}$.

3 Results

165   ### 3.1 SF_6 mixing ratios

In order to evaluate the SF_6 mixing ratios simulated of the EMAC model, we first analyse the four tracers in the SD simulation in comparison to observational data. Fig.1a depicts the modelled SF_6 vertical profile climatologies in comparison with MIPAS
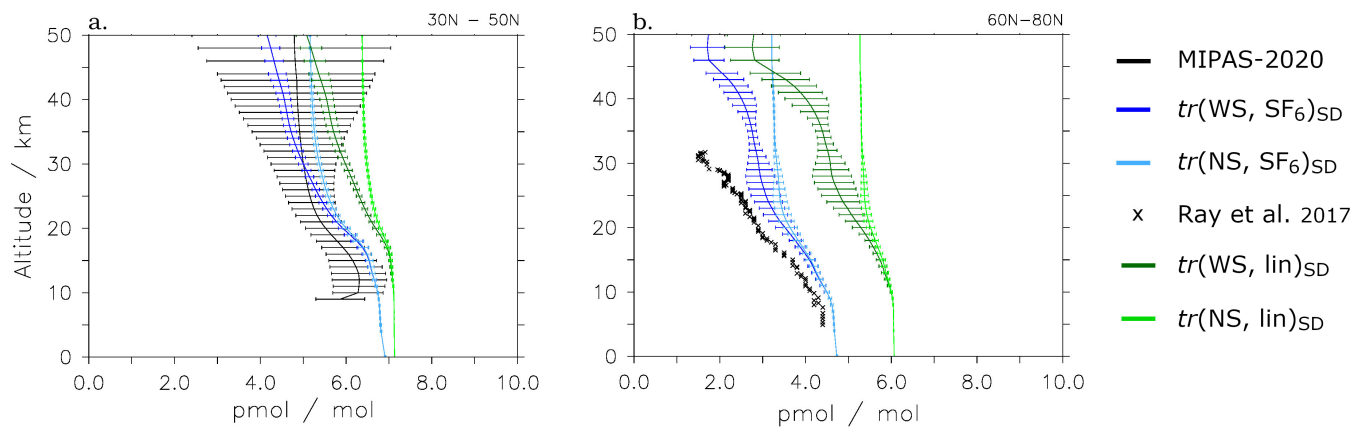
Figure 1. Zonally averaged vertical SF_6 profiles for the four tracers from the SD simulation averaged over (a) 30°N-50°N for 2007-2010 and (b) 60°N- 80°N for 5th March 2000. Horizontal lines show the SF_6 spread over all longitudes. Dark blue: non-linear tracer with sinks $tr(WS, SF_6)$; light blue: non-linear tracer without sinks $tr(NS, SF_6)$; light green: idealised tracer $tr(NS, lin)$; dark green: linear tracer with sinks $tr(WS, lin)$; In panel a), the SF_6 mixing ratios obtained from MIPAS (Stiller et al., 2020, and paper in preparation) are shown in black. Error bars depict the standard deviation of MIPAS SF_6. Black crosses in panel b) represent the balloon-borne measurements (Ray et al., 2017) taken on March 5th, 2000 at Kiruna, Sweden (67°N).

SF_6 (Stiller et al., 2020, paper in preparation). Fig.1b shows the modelled SF_6 vertical profiles and balloon-borne measurements of SF_6 (Ray et al., 2017) on a particular day. The former comparison is for zonal mean SF_6 averaged over 30°N-50°N and 2007-

170 2010. The latter shows modelled SF_6 mixing ratios of the day of the balloon flight. The balloon was launched on March 5th, 2000 at 67°N in Kiruna, Sweden. To account for the relatively coarse model resolution and for possible balloon drifting, the modelled SF_6 values are averaged over all longitudes and 60°N-80°N. In both comparisons, the standard deviation of SF_6 over all longitudes averaged over the respective latitude range is shown as error bars.

The tracers with non-linear growth in the SD simulation show smaller tropospheric SF_6 mixing ratios than the linear tracers

175 (Fig.1a and b). This can be explained by the two different growth scenarios of SF_6 and the prescribed lower boundary conditions (see Fig. S2 in Supplementary Information). The sinks do not have a considerable effect in the troposphere, hence the effect of the SF_6 sinks becomes only noticeable higher up. Furthermore, the effect of the SF_6 depletion becomes increasingly evident with altitude. This is portrayed in the growing differences with altitude between $tr(WS, lin)$ and $tr(NS, lin)$ and the non-linear equivalent. The differences particularly increase for the tracers with linear emissions, as these exhibit higher SF_6 mixing ratios

180 and hence experience greater SF_6 depletion than those with non-linear boundary conditions. Due to the small turnaround times for air in the middle atmosphere, the tracers without sinks exhibit a very low decrease of the SF_6 mixing ratios with altitude.

Fig.1a shows that the EMAC simulated non-linear SF_6 is within the observed range of MIPAS SF_6. Below 30 km, MIPAS SF_6 mixing ratios are smaller, with a near-constant offset of approximately 0.5 pmol/mol up to 20 km. Above 30 km, MIPAS SF_6 shows larger mixing ratios than EMAC. This means that EMAC SF_6 (SD(WS, SF_6)) shows a larger decrease with altitude

185 than MIPAS SF_6 , suggesting that the sinks in EMAC are too strong. Another explanation could be too strong vertical mixing in

EMAC. However, the EMAC SF_6 lies within the MIPAS uncertainty range throughout the atmosphere. The standard deviation increase with height in MIPAS SF_6 can be attributed to the decrease of the SF_6 signal with height, which leads to an increase in noise-error of SF_6. Additionally, the natural variability of SF_6 itself, as well as the evolution of SF_6 over time, contribute to the increasing standard deviation in the MIPAS SF_6 profile. The increase in standard deviation with height can also be seen in the

190 EMAC SF_6 profiles, particularly in the tracers $tr(WS, SF_6)$ and $tr(WS, lin)$. However, it is by far not as large as in the MIPAS data because the simulations have no measurement error and possibly show a smaller natural variabilty than the observations.

The balloon flight SF_6 profile (Ray et al., 2014) in Fig.1b largely resembles the profile of the realistically modelled tracer $tr(WS, SF_6)$. Below 25 km, the modelled SF_6 profile shows a constant high bias of around 0.3 pmol/mol, presumably due to the lower boundary conditions used. Larger discrepancies can be seen above 25 km altitude, with higher mixing ratios of the

195 modelled SF_6. As the data presented in Fig.1b are only for a specific day and region, the particular meteorological situation can be crucial for the comparison.

3.2 SF_6 lifetimes

The atmospheric lifetime of SF_6 can be used as an indicator for the accuracy of the SF_6 degradation scheme. We calculate the lifetime according to the definition Braesicke et al. (see e.g. 2019). Only the realistic tracer $tr(WS, SF_6)$ is considered.

200 The reference simulation yields an average lifetime of 2101 years. Reddmann et al. (2001) carried out sensitivity experiments with this scheme, using various options for the chemical mechanisms. In this way, the lifetime could be varied between 400 and 10000 years. Our value is below the value 3200 years calculated by Ravishankara et al. (1993), but above the numbers 1278 years and 850 years of the more recent studies by Kovács et al. (2017) and Ray et al. (2017), respectively. In another new modelling study, Kouznetsov et al. (2020) presented a range of 600-2900 years. Our value of around 2100 years is therefore

205 within, but rather at the upper range of the uncertainties. In contrast to the comparison of the model results with SF_6 observations shown in the previous section, our lifetime value points towards rather weak SF_6 sinks in our scheme. To assess the variability of the atmospheric SF_6 lifetime, we show in Fig. 2 the timeseries of the SF_6 lifetimes for the four simulations that were described in Sect. 2. The lifetime of the REF simulation lies at about 2500 years in 1965 and decreases by approximately 25% to 1900 years in 2011. The lifetime of the projection simulation behaves similarly, and increases to about 2400 years by

210 the year 2100. This shape of the lifetime resembles that of projected O_3 (see e.g. Eyring et al., 2007), which is reflected in the ESCiMo simulations (Jöckel et al., 2016) from which the O_3 and the other SF_6-reactant species are prescribed here. However, our SF_6 degradation scheme includes a number of simplifications, which can modify the lifetimes. For example, a constant profile is used to prescribe the sinks through NO. This can simplify the long term variability of the SF_6 lifetimes by making it overly dependent on the species that are transiently prescribed in the simulations. Apart from seasonal variations, the lifetimes

215 of the CSS and the TS2000 simulations are fairly constant, their lifetime values lie around 2600 and 1900 years, respectively. The fact that the lifetimes of the CSS simulations are fairly constant implies that the long-term trends of the SF_6 lifetimes can mostly be attributed to the abundance of the species involved in the SF_6 degradation. Variations of stratospheric temperatures or the circulation strength seem to only play a minor role.
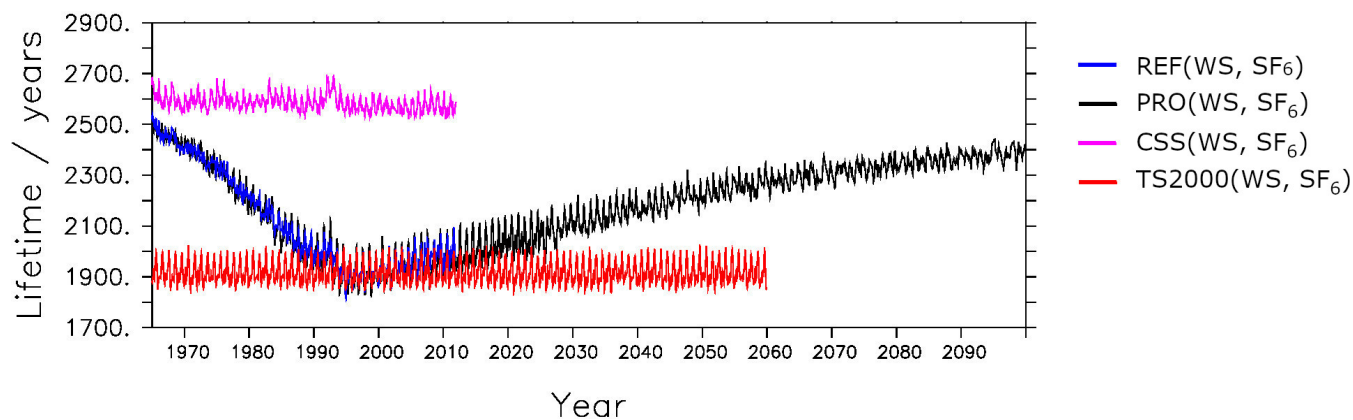
Figure 2. Global stratospheric and mesospheric lifetimes of SF_6 calculated from the tracer with realistic lower boundary conditions and SF_6 sinks (tr(WS, SF_6)). Blue: reference simulation (REF); black: projection simulation (PRO); pink: constant reactant species simulation (CSS); red: time slice 2000 simulation (TS2000).

3.3 Age of air climatologies

220 AoA climatologies averaged over 2007 to 2010 from the reference simulation are shown in Fig. 3a-d (REF(NS, lin), REF(NS, SF_6), REF(WS, lin), and REF(WS, SF_6), from the specified dynamics simulation (SD(WS, SF_6)) in Fig. 3e and from MIPAS SF_6 observations in Fig. 3f (Stiller et al., 2020, paper in preparation). The years 2007 to 2010 were chosen as these are the only years with complete MIPAS data.

In all cases, AoA increases with increasing altitude and latitude. The cases considering sinks show older apparent AoA
225 than those without, especially with increasing altitude and latitude. This apparent ageing can be explained by the fact that the inclusion of mesospheric sinks results in smaller SF_6 mixing ratios. The reduced mixing ratios lead to seemingly older AoA as the corresponding reference value lies further in the past. Downwelling within the polar vortices transports old air from the mesosphere to the stratosphere. With the breakdown of the polar vortex at the end of the winter season, the old air is then mixed into lower latitudes.

230 The patterns of modelled AoA from the linear tracers are similar to those of the SF_6 emissions-based tracers (compare Fig. 3a with Fig. 3b, and Fig. 3c with Fig. 3d). However, the similarities are weaker in the polar regions, especially when the SF_6 sinks are considered (Fig. 3c and Fig. 3d). In these regions, REF(WS, lin) reaches AoA values spanning 10–15+ years, and REF(WS, SF_6) 9–14 years. This difference can be attributed to the greater initial growth of the tracer with linear emissions than that with non-linear emissions (Fig. S2 in Supplementary Information). This leads to enhanced SF_6 mixing ratios in the
235 REF(WS, lin) case and in turn strengthens the influence of SF_6 sinks on AoA. This is particularly relevant in the winter months when SF_6-depleted mesospheric air is transported downward into the polar stratosphere.

AoA derived from SF_6 emissions including chemical SF_6 sinks (REF(WS, SF_6), Fig. 3d) agrees best with MIPAS AoA (Fig. 3f). Overall good agreement between EMAC and MIPAS AoA is found in the tropics, but there is a large high bias in
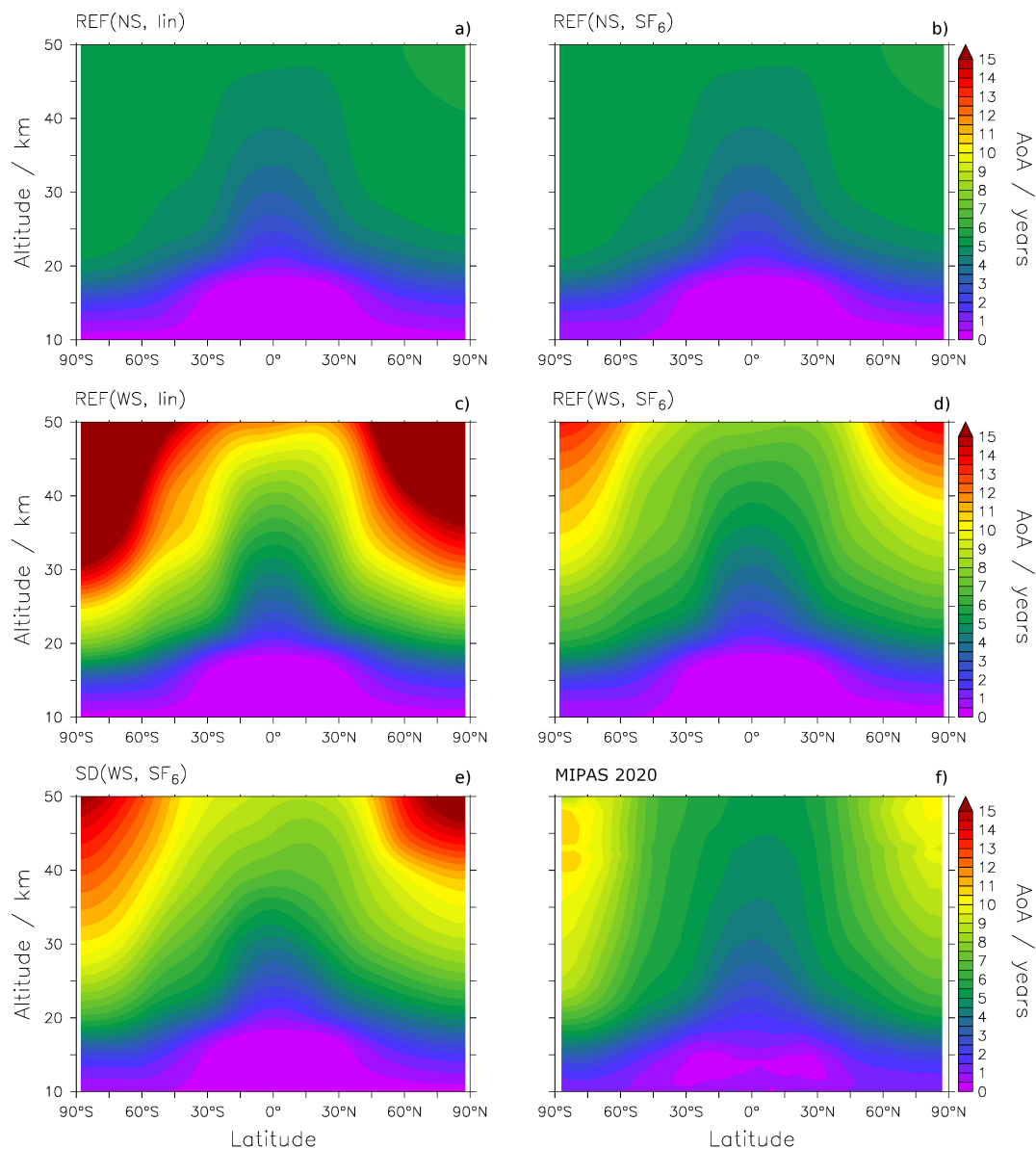
Figure 3. AoA climatologies of annual means over 2007-2010. Model AoA from the reference simulation for the different tracers tr(NS, lin), tr(NS, SF_6), tr(WS, lin), and tr(WS, SF_6) is shown in panels a) through d), respectively, while AoA (tr(WS, SF_6)) from the specified dynamics simulation is shown in panel e). MIPAS (Stiller et al., 2020, paper in preparation) AoA can be seen in panel f).

the high latitudes in EMAC: between 40°N-50°N, modelled AoA is up to 2 years older than MIPAS AoA in the stratosphere,
240 and up to 3 years older in the polar upper stratosphere (see Fig. S5 in Supplementary Information). In comparison to the MIPAS observations, the SF_6 sinks therefore seem to be too strong in the model, as already mentioned above. However, in

comparison with the previously published MIPAS data (Stiller et al., 2012; Haenel et al., 2015), the EMAC AoA was actually too young, i.e. the MIPAS AoA was much older in the previous versions in the polar regions. The spectroscopic data used for the SF_6 retrieval in MIPAS cause a rather large bias (that has now been corrected by improved spectroscopy). The new

245 spectroscopic data lead to considerably younger AoA in the middle to upper stratosphere. There are, however, good reasons to believe that the most recent MIPAS data are improved compared to the previous ones: the spectroscopic data used are far better characterized than the previous ones (Harrison, 2020), and the new AoA data from MIPAS agree significantly better with independent measurements than the previous version, in particular at higher altitudes (Stiller et al., 2020). A detailed analysis of this will be provided in an upcoming paper on the new MIPAS retrieval. On the other hand, free-running EMAC simulations

250 generally have a too weak Antarctic polar vortex (see Jöckel et al., 2016), which is, however, stronger than that of the reference simulation. Therefore, the more stable vortex in the SD simulation leads to enhanced isolation and ageing of polar stratospheric air, especially in the Southern Hemisphere during austral spring (see Supplement, Fig. S4, for details). This could somewhat resolve the issue for the comparison with the previous MIPAS AoA version (see Stiller et al., 2012; Haenel et al., 2015, for further details); for the present MIPAS version, however, the discrepancies in the high altitudes and latitudes with the EMAC

255 SD simulation are even larger than with the REF simulation. The comparison of Fig. 3e) with Fig. 3f) illustrates that the model cannot reproduce the tropical pipe and exhibits too much horizontal mixing, or too slow upwelling. This could explain why the model does not reproduce the constant AoA with height in the mid-latitudes. A detailed assessment of both the satellite data and the model simulations is necessary to resolve these discrepancies. In the model, dynamical effects like the strength of the polar vortex or the gravity wave parameterisation can play important roles for the downwelling strength. Moreover, various

260 processes of the chemical SF_6 removal can be revised and/or parameterised differently. Here, we showed that SF_6 sinks have the potential to resolve the differences between simulated and observed climatologies of AoA and that EMAC AoA lies within the uncertainties of MIPAS AoA throughout the atmosphere. We therefore consider our simulations suitable for studying the temporal evolution of AoA.

3.4 Apparent age of air trends

265 In this section, we analyse the EMAC AoA trends and compare them with observation-based AoA trends. Fig. 4 shows the AoA time series and linear regressions from the REF and the SD simulations as well as from MIPAS observations (Stiller et al., 2020, and paper in preparation) and from the SF_6 measurements by Engel et al. (2009). As the latter were collected from balloon flights in the Northern Hemisphere mid-latitudes at around 30 km altitude, the EMAC and MIPAS data are also taken from that height and averaged over 30°N to 50°N for consistency.

270 For better quantification of the trends, Tab. 3 provides the AoA trend values of the EMAC simulations for two periods. The trend of the entire simulation period 1965-2011 is taken for long-term trend assessment and comparison with the measurements by Engel et al. (2009). For the comparison with MIPAS data, the EMAC AoA trends are shown for the period 2002-2011 between 30°N and 50°N, for the realistic tracer tr(WS, SF_6). The trend calculation follows that of Haenel et al. (2015).

The tracers without SF_6 sinks lead to negative AoA trends, which are consistent with the simulated acceleration of the BDC

275 in the course of climate change (e.g. Garcia and Randel, 2008). Positive AoA trends are obtained for all tracers that take SF_6
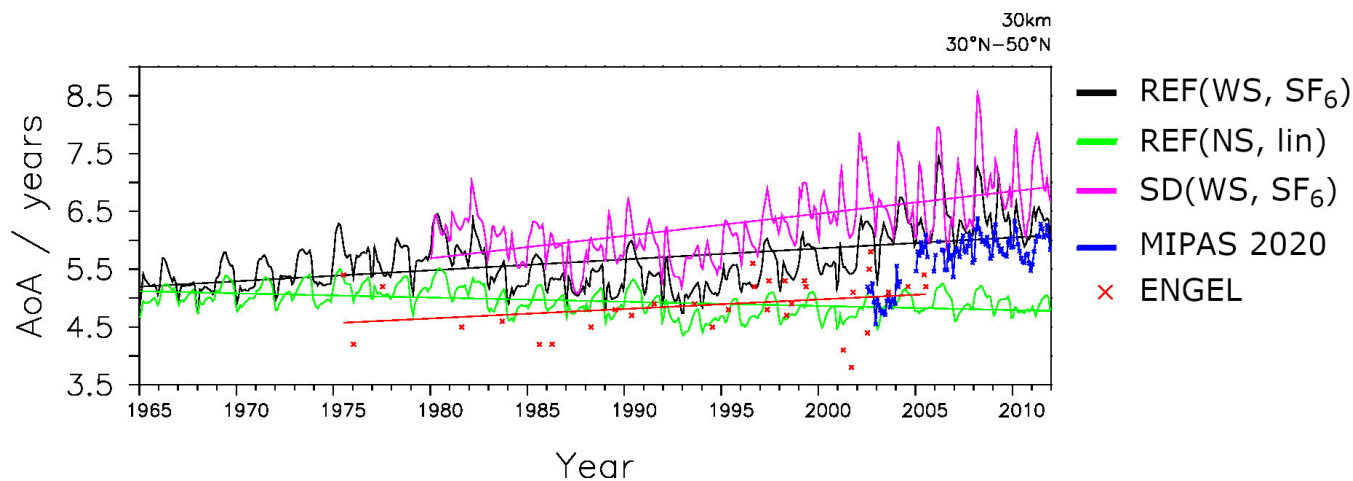
Figure 4. AoA time series and linear regressions calculated at 30 km averaged over 30°N-50°N. EMAC AoA from SF_6 with non-linear emissions from the reference simulations is shown in black (REF(WS, SF_6)); AoA from the tracer with linear emissions without sinks in green (REF(NS, lin)). AoA from SF_6 with non-linear emissions with sinks in the specified dynamics simulation SD(WS, SF_6) is shown in pink. AoA from balloon-borne measurements (Engel et al., 2009) and from MIPAS observations (Stiller et al., 2020, and paper in preparation) are shown in red and dark-blue, respectively.

Table 3. EMAC AoA trends at 30 km averaged over 30°N-50°N. The calculation of the 2002-2011 trends is provided for the two relevant simulations, REF(WS, SF_6) and SD(WS, SF_6), and follows the methods used in Haenel et al. (2015). The 2002-2011 trends are also provided for the remaining simulations.

Simulation	1965-2011 Trend (years/decade)	2002-2011 Trend (years/decade)[a]
REF(WS, SF_6)	0.19 ± 0.26	0.22 ± 0.12
REF(NS, SF_6)	-0.06 ± 0.09	-0.07 ± 0.06
REF(WS, lin)	0.70 ± 0.96	0.23 ± 0.22
REF(NS, lin)	-0.07 ± 0.10	-0.06 ± 0.05
SD(WS, SF_6)[b]	0.39 ± 0.36	0.50 ± 0.13
CSS(WS, SF_6)	0.11 ± 0.16	0.19 ± 0.08
TS2000(WS, SF_6)[b]	0.23 ± 0.32	0.07 ± 0.12
TS2000(NS, lin)[b]	-0.00 ± 0.01	-0.21 ± 0.05
MIPAS[c]		0.34 ± 0.13
Engel et al. (2009)[d]	0.24 ± 0.22	

[a] Trend calculated following methods of Haenel et al. (2015)

[b] Trend calculated over 1980 - 2011 at 30 km altitude.

[c] MIPAS trend calculated over the 2002-2012

[d] Trend calculated over 1975 - 2005 between 32°N-51°N and 24 km-35 km

chemistry into account. The trend of 0.19 ± 0.26 years per decade in the REF simulation (WS, SF_6) is within the limits of the uncertainties of the trend obtained by Engel et al. (2009), who calculated an AoA trend of 0.24 ± 0.22 years per decade. This means that in our simulations, the sinks help to reconcile the modelled and the measured AoA trends over the recent decades. Note that Engel et al. (2009) and Engel et al. (2017) also obtained a positive trend for AoA derived from CO_2 measurements.

280   Haenel et al. (2015) calculated MIPAS AoA trends for the period 2002-2012 of 0.25 ± 0.11 years per decade for 30°N-40°N at 30 km and 0.24 ± 0.11 years per decade for 40°N-50°N at 30 km. For the new MIPAS dataset, the AoA trend is 0.34 ± 0.13 years per decade. Note that these trends are calculated by applying a bias correction for the discontinuity between the two different observational periods of MIPAS (see Fig. 4), a description of the method can be found in von Clarmann et al. (2010).

Following the trend calculation used in Haenel et al. (2015), the REF(WS, SF_6) and SD(WS, SF_6) time series show that
285   EMAC AoA bears a good resemblence to that of the new MIPAS retrieval, with an AoA trend of 0.22 ± 0.12 and 0.50 ± 0.13 years per decade, respectively. Consistent with the trend calculation of the MIPAS data, the variability due to the QBO is considered by a respective term in the multivariate linear analysis. However, this measure induces only small differences in the EMAC trend calculations (see Table S1 in Supplementary Information for further details). Note also that the rather short period of the MIPAS observations implies rather large uncertainties in the trends because interannual variability can have a large
290   effect on the trend calculations. For sake of completion, the 2002-2011 trends of the remaining simulations are also provided. The 2002-2011 AoA trend for the timeslice simulation TS2000(NS, lin) is strongly negative at -0.21 ± 0.05 years per decade, as opposed to the 1965-2011 AoA trend of -0.00 ± 0.01 years per decade, due to the sharp decrease towards the end of the 2002-2011 time period. As 2002-2011 is a rather short time period, the AoA trend of the TS2000(WS, SF_6) simulation is less positive at 0.07 ± 0.12 years per decade compared to the AoA trend from the simulations REF(WS, SF_6) (0.22 ± 0.12 years
295   per decade) and CSS(WS, SF_6) (0.19 ± 0.08 years per decade).

3.5   Explanations for apparent Age of Air trends

In this section, we will analyse the EMAC apparent AoA trends, in particular the sign change of the trend when SF_6 sinks are switched on. For this, Fig. 5 shows the AoA (WS,SF_6) time series of the sensitivity simulations CSS and TS2000 averaged between 30°N-50°N. For comparison, AoA from the reference simulation (REF(WS, SF_6) and REF(NS, lin)) are also included.
300   In the CSS simulation the mixing ratios of the reactant species involved in the sink reactions of SF_6 are held constant. The CSS simulation with the realistic tracer (CSS(WS, SF_6)) shows an AoA trend of 0.11 ± 0.16 years per decade in AoA over the 1965-2011 period. This is lower than the AoA trend in the reference simulation REF(WS, SF_6) and means that while the changes in the SF_6-depletive substances influence the magnitude of the positive trend, they cannot explain the positive sign of the AoA trend.

305   The TS2000 simulation is a time slice simulation with climate conditions of the year 2000. AoA of that simulation derived from the realistic tracer (TS2000(WS, SF_6)) shows a positive trend of 0.23 ± 0.32 years per decade. This is even stronger than the trend of the REF simulation (0.19 ± 0.26 years per decade). By definition, the TS2000 simulation does not feature any changes in its climatic state or in the composition of the atmosphere. This is reflected by the fact that the idealised tracer tr(NS, lin) does not show a trend in this simulation (see Tab. 3, TS2000(NS, lin): 0.00 ± 0.01). The temporal increase in apparent AoA
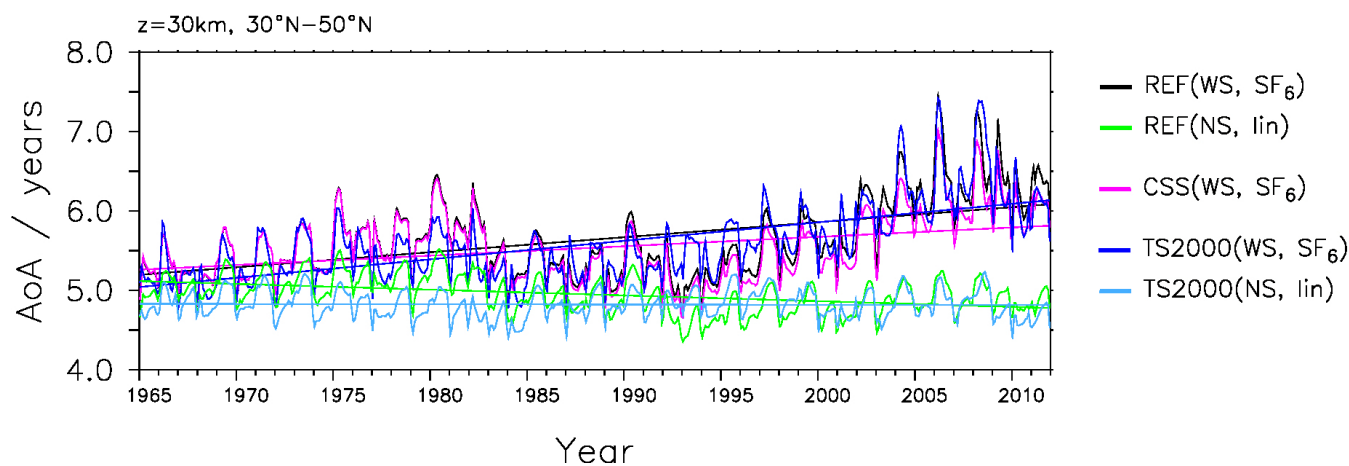
Figure 5. AoA (at 10 hPa, averaged over 30°N-50°N) time series and linear regression of sensitivity experiments TS2000(WS, SF$_6$), TS2000(NS, lin), and CSS(WS, SF$_6$) shown in dark blue, light blue, and pink, respectively. Reference simulations REF(WS, SF$_6$) and REF(NS, lin) shown in black and green, respectively.

310 rise in the TS2000(WS, SF$_6$) simulation despite no climate changes therefore points to the fact that the SF$_6$ sinks themselves lead to the positive trend. The difference between the TS2000 and the REF AoA trends for both tr(WS, SF$_6$) and tr(NS, lin) reflect the negative contribution of the accelerating BDC to the trend, which can be seen in the idealised AoA trend from the REF simulation (NS, lin). Overall, neither changes in the SF$_6$ sinks nor changes in the stratospheric circulation due to climate change are responsible for the positive trend found in AoA with sinks. Instead, the results indicate that the sinks themselves 315 can generate a positive trend.

For a complete discussion of the features that can be seen in Fig.5, we now also describe the sudden decrease in AoA(WS, SF$_6$) shortly after 1982 seen in the REF simulation. It is also visible in the two sensitivity simulations (CSS and TS2000). This means that changes in the SF$_6$-depleting substances as well as climate change and volcanic activity (in particular the volcanic erruption of El Chichón in 1982) can be ruled out as a possible cause for the drop. The solar cycle is not taken into account in 320 the timeslice simulation. The timeseries of the realistic SF$_6$ tracer emissions show a stronger increase in the 1980s (not shown). Fritsch et al. (2019) showed that due to this increase the calculation of AoA based on SF$_6$-like tracers is more sensitive to the chosen parameters in the AoA derivation for this time. The drop in AoA we see is caused by this limitation of the derivation from SF$_6$-like tracers.

The increasing variability and trend of AoA in the last two decades, seen in the simulations with mesospheric SF$_6$ chemistry, 325 can be attributed at first order to the SF$_6$ depletion reactions (see Fig.5). In particular, the effect of mesospheric SF$_6$ sinks is stronger with higher SF$_6$ mixing ratios. Subsequent downward transport of SF$_6$ depleted air into the vortex and in-mixing thereof into lower latitudes after the vortex breakup results in apparent older AoA. This can explain that the annual variability increases over time due to the increase in SF$_6$ mixing ratios.

In the following, we will show that SF_6 sinks with constant destruction rates lead to a positive trend in AoA. The afore-

330   mentioned link between the positive AoA trend and mesospheric SF_6 depletive chemistry, based on Hall and Waugh (1998), is illustrated below and follows the mathematical formulations put forward by Hall and Plumb (1994) and Schoeberl et al. (2000). We consider a tracer $\chi(t)$ experiencing relative loss $e^{-k \cdot t}$ with constant loss rate k over time t. We denote the mixing ratio of reference SF_6 as $\chi_o(t)$ with a constant linear growth rate χ_{oo} and write

$$\chi_o(t) = \chi_{oo} \cdot t. \tag{1}$$

335   At any location we can then write the concentration as

$$\chi(t) = \int_{\tau=0}^{\infty} \chi_o(t-\tau) exp(-k \cdot \tau) G(\tau) d\tau \tag{2}$$

where the Green's function is written as $G(\tau)$ and the concentration lag time as τ. Upon insertion of (1) into (2) we can express the latter as

$$\chi(t) = \chi_{oo} \cdot t \int_{\tau=0}^{\infty} exp(-k \cdot \tau) G(\tau) d\tau - \chi_{oo} \int_{\tau=0}^{\infty} \tau exp(-k \cdot \tau) G(\tau) d\tau \tag{3}$$

340   The first integral corresponds to the Laplace transform of the Green function $G(\tau)$, denoted as $\tilde{G}(k)$, and the second integral the differential thereof, and so

$$\chi(t) = \chi_{oo} \left(t \cdot \tilde{G}(k) + \frac{\partial}{\partial k} \tilde{G}(k) \right) \tag{4}$$

$G(\tau)$ is assumed to be constant, i.e. this is for a constant circulation strength. The tracer without sinks is referred to as the passive tracer, and (using the subscript p for passive) the concentration $\chi_p(t)$ can be expressed as a function of AoA, shown

345   here as Γ, namely:

$$\chi_p(t) = \int_{\tau=0}^{\infty} \chi_o(t-\tau) G(\tau) d\tau = \chi_{oo} \cdot (t - \Gamma). \tag{5}$$

When rearranging (5), we can write the AoA inferred from the passive tracer as

$$\Gamma = t - \frac{\chi_p(t)}{\chi_{oo}} \tag{6}$$

Differentiating this with respect to time shows that no trend is found when considering the passive tracer, since $\chi_p(t) = \chi_{oo} \cdot t + a$

350   where a is constant, and so

$$\frac{\partial \Gamma}{\partial t} = 1 - \frac{1}{\chi_{oo}} \cdot \frac{\partial \chi_p}{\partial t} = 1 - \frac{1}{\chi_{oo}} \cdot \chi_{oo} = 0 \tag{7}$$

When considering an active tracer (i.e. with mesospheric sinks), the apparent age Γ_s can be expressed as

$$\Gamma_s = t - \frac{\chi_s(t)}{\chi_{oo}} \tag{8}$$

The subscript s denotes the active tracer. In combination with (4) one can thus write

355
$$\Gamma_s = t \cdot \left(1 - \tilde{G}(k)\right) - \frac{\partial}{\partial k}\tilde{G}(k) \tag{9}$$

with

$$\frac{\partial \Gamma_s}{\partial t} = 1 - \tilde{G}(k) \tag{10}$$

where $\tilde{G}(k=0) = 1$ whereas $\tilde{G}(k \to \infty) = 0$. The former case yields no trend in the case excluding mesospheric SF$_6$ sinks, as seen in the time slice simulation with the idealised tracer (see TS2000(NS, lin) in Table 3).

360 In the case of a passive tracer (i.e. a tracer without sinks), $k = 0$ and $\tilde{G}(k=0) = 1$. In other words, the AoA trend is zero in the absence of a trend in transport (constant G). This is the case in the TS2000 time slice simulation for the passive tracers (see TS2000(NS, lin) in Table 3). In the reference simulation this translates to a negative trend, consistent with the modelled acceleration of the BDC (e.g. Eichinger et al., 2019; Butchart et al., 2011). For sinks of finite strength, $G(k)$ lies between 0 and 1, so that with consideration of the mesospheric SF$_6$ degradation scheme in the simulations, a positive trend in apparent

365 AoA is found. In our simulations, this effect overcompensates the impact of the BDC acceleration. It is important to note that the above is valid for a linearly increasing tracer, and that variable growth rates will modify the influence of SF$_6$ on apparent AoA.

4 Summary and conclusions

Disagreements with regards to stratospheric AoA and particularly its trends between model simulations and observations have

370 been raising many questions by scientists for more than a decade now. AoA from observations is mostly older than AoA from model simulations and models simulate a decrease in AoA over recent decades, whereas trend estimates from observational data report a non-significant positive trend. To make another step towards understanding the reasons for these discrepancies, we here study the impact of mesospheric SF$_6$ sinks on AoA climatologies and trends using the EMAC (Jöckel et al., 2010; Jöckel et al., 2016) model with the SF6 submodel (Reddmann et al., 2001) allowing for explicit calculation of SF$_6$ sinks.

375 The EMAC SF$_6$ mixing ratio profiles show good agreement with balloon-borne measurements as well as with satellite observations. Some of the differences between the model and observations are within the uncertainty range of the observations. However, reasons for the quantitative differences in the high latitudes and altitudes can also be found in deficiencies in the representation of the SF$_6$ sinks in the model or in the dynamics simulated by the model. The EMAC reference simulation yields a global stratospheric SF$_6$ lifetime of 2100 years, varying between 2500 years and 1900 years for the simulation period. This

380 value lies within the range of 600-2900 years provided by the model study of Kouznetsov et al. (2020) and below the value 3200 years calculated by Ravishankara et al. (1993). Kovács et al. (2017) and Ray et al. (2017) recently found somewhat lower SF$_6$ lifetimes of 1278 years and 850 years, respectively. Although this shows that large uncertainties still exist in determining the SF$_6$ lifetime, these results also confirm that the EMAC SF$_6$ depletion mechanisms are reasonable. In our transient simulations (REF and PRO), the SF$_6$ lifetimes vary by about 25 % following the abundances of the reactant species, basically resembling

385 the pattern of the stratospheric ozone concentration. This behaviour, however, may be a result of the fact that several effects that potentially influence SF_6 lifetime variability are not implemented in full detail.

The inclusion of SF_6 sinks translates into apparent older stratospheric air. When SF_6 sinks are enabled, EMAC AoA therefore compares better with MIPAS satellite observations. In the tropics, overall good agreement can be found, but the results also indicate that EMAC does simulate a too broad, less isolated tropical pipe. In polar regions, however, EMAC AoA is higher than
390 MIPAS AoA. In comparison with the previously published MIPAS data (Stiller et al., 2012; Haenel et al., 2015), the EMAC AoA in the polar regions was actually too low. More research on both models and observations is necessary to resolve these remaining discrepancies.

Without SF_6 sinks, EMAC shows a negative AoA trend over 1965-2011. This is consistent with the simulated acceleration of the Brewer-Dobson circulation resulting from climate change (see e.g. Garcia and Randel, 2008; Butchart et al., 2011;
395 Eichinger et al., 2019). The inclusion of chemical SF_6 sinks leads to positive AoA trends in our simulations, which in turn is consistent with the positive AoA trend derived from MIPAS in the Northern Hemisphere (Haenel et al., 2015). Moreover, the SF_6 sinks help to improve the agreement of our model results with the AoA derived from the balloon-borne in situ measurements by Engel et al. (2009), from which a (non-significant) positive AoA trend was obtained. However, this only accounts for SF_6 -derived AoA, our results cannot help explain the positive trend of the CO_2-derived AoA in Engel et al. (2009, 2017).
400 Furthermore, it has recently been shown that the AoA trend derived in Engel et al. (2009, 2017) was likely overestimated due to non-ideal parameter choices in the calculation of AoA (Fritsch et al., 2019). Our sensitivity studies show that the positive AoA trends are neither a result of climate change, nor of changes in the substances involved in SF_6 depletion. The SF_6 sinks themselves are the reason for the increase in apparent AoA. The reason for that is the temporally increasing influence of the chemical SF_6 sinks on AoA. In our simulations, this effect overcompensates the effect of the simulated acceleration of the
405 stratospheric circulation leading to a net increase of AoA. Due to various sources of uncertainties, this result bears quantitative leeway and has to be assessed in finer detail. But nevertheless, for now we can conclude that SF_6 sinks have the potential to explain the long-lasting AoA trend discrepancies between models and observations.

Code availability. The Modular Earth Submodel System (MESSy) is continuously developed and applied by a consortium of institutions. Use of MESSy and access to the source code is licensed to all affiliates of institutions that are members of the MESSy Consortium. Institutions
410 can become a member of the MESSy Consortium by signing the MESSy Memorandum of Understanding. More information can be found on the MESSy Consortium website (http://www.messy-interface.org, last access: 27 March 2020). The exact code version used to produce the simulation results is archived at the German Climate Computing Center (DKRZ) and can be made available to members of the MESSy community upon request.

Data availability. The simulation results are archived at DKRZ and are available upon request. MIPAS SF_6 and AoA data are available from
415 G. Stiller on request.

Author contributions. SL, RE and HG designed the study. SL analysed the data with support from RE, HG and FF. RE performed the simulations. TR and SV implemented the SF6 submodel in MESSy. GS and FH provided the MIPAS data analyses. SL and RE drafted the paper and all authors helped with discussions and with finalising the manuscript.

Competing interests. The authors declare no competing interests.

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