

Responses to the reviewers comments on "Simulating age of air and distribution of SF₆ in the stratosphere with SILAM model"

Rostislav Kouznetsov^{1,2}, Mikhail Sofiev¹, Julius Vira^{1,3}, and Gabriele Stiller⁴

¹Finnish Meteorological Institute, Helsinki, Finland

²Obukhov Institute for Atmospheric Physics, Moscow, Russia

³Currently at Cornell University, Ithaca, NY, USA

⁴Karlsruhe Institute of Technology, Karlsruhe, Germany

Correspondence: Rostislav Kouznetsov (Rostislav.Kouznetsov@fmi.fi)

1 Response to Anonymous Referee #2

Overall the authors addressed the reviewer comments in detail, and they added thorough discussion to the paper. I have no major concerns about the paper and recommend it for publication after a few minor additional comments are addressed (see below).

5 *Minor comments:*

- page 22, line 552: Did you actually check the lifetimes obtained via loss rates, and are they indeed similar? I'm wondering whether differences in lifetime estimates in different models might also be due to methodological differences?

Response: No, we did not. Our period of the stopped emission (2.5 years) is much shorter than the transport time from the surface to the stratosphere. Then these approaches should be equivalent. We fully agree that the differences in the lifetime
10 estimates in different models can be caused by methodological differences. The issue starts from the definition of the lifetime in non-equilibrium system (see forth paragraph of the "Lifetime..." subsection).

- page 25, line 644: The theory for why sinks introduce apparent trends on AoA was also already mentioned by Schoeberl et al. (2000)

Response: Thank you for the reference! The reference is added, the sentence referring to Stiller et al. (2012) rewritten more
15 accurately.

- page 26, line 659: The reason for the difference to the study by Plöger et al. (2015) likely is that they used diabatic heating rates as vertical velocity, and it is known that the diabatic and kinematic vertical transport is inconsistent in the reanalysis (Abalos et al., 2015).

Response: This note and the corresponding reference added.

20 *- page 27, line 710: Did I understand it correctly that the diffusivities in ERA data are refer to parameterized vertical diffusion? In particular in ERA5, likely a considerable amount of vertical mixing by the resolved gravity waves is included, so you might want to restate the judgement of diffusivities in ERA?*

Response: The diffusivities in the ERA5 dataset are given with “mtdh” (mean turbulent diffusion for heat) parameter. It is not clear to us how resolved gravity waves can transport scalars, unless they brake and induce turbulence. Since ERA-interim
25 clearly does not resolve any turbulence, one would expect mtdh to be a measure of mixing due to turbulence and other subgrid processes. Then our judgement holds.

- page 28, line 730: *In general, I wonder what the role of "numerical" diffusion by the advection scheme is?*

Response: The scheme used in SILAM has no diffusion, i.e. it does not create down-gradient transport. It has however a hyper-diffusion, which can be controlled by having thin enough vertical layers.

30 - page 28, line 749: *For a discussion on the correction for the non-linear increase in SF6, you might want to take a look at a new article in ACPD (Fritsch et al, Sensitivity of Age of Air Trends on the derivation method for non-linear increasing tracers). It is important to point out that in principle it is possible to derive "correct" AoA also from non-linear increasing tracers if the parameters are chosen correctly (which is of course very challenging for observational data).*

Response: Thank you! We agree that correct choice of the parameters is important. Besides that, the approach of ? also
35 heavily relies on the assumption that the quadratic fit is a valid model, and that the spectra are steady in time. Essentially, that means that one has to know quite a lot about AoA spectra in order to get AoA properly.

- page 28, line 723: *Did you mean to say "Fig. 14" for a comparison to the vertical velocities?*

Response: Yes. Corrected.

Technical:

40 *In general, please recheck the language in the new sections and paragraphs*

- page 5, line 149: *"transport with of SF6" ? (delete with)?* - page 6, line 165: *"transport" duplicated*

Response: Corrected.

- page 23, line 578: *"likely to cause overstating..." -> "which is likely due to higher depletion rates..." (?)*

Response: The phrase rewritten.

45 - page 24, line 604: *"differences of are" - something missing?* - page 25, line 651: *if AoA -> of AoA* - page 26, line 686: *"in the meteorological driver" -> e.g. "...in the meteorological data used to drive the model.."* - page 27, line 694: *missing ")"* after Fig. 14 - page 27, line 701: *"same-vertical mass-flux" - not sure what the "same" refers to?* - page 27, line 706: *"One can hope": please rephrase, hope is great in life, but not in a scientific paper* - page 28. line 727: *explicit referencing to anonymous reviewers is maybe not appropriate, just remove the first part of the sentence.* - page 28, line 754: *"factor of times" - dont*
50 *understand this phrasing* - page 29, line 761: *"it makes a full sense": please rephrase, e.g. "it would be useful to..."* - page 29, line 768/769: *"comparable" duplicated* - page 30, line 810: *"ithout" (w missing)*

Response: Corrected.

2 Response to Anonymous Referee #3

The authors have improved many parts of the manuscript considerably. However, some issues remain as is detailed in the following. Firstly, it is very hard to find out how and where some of the changes have been implemented. As an example, in response to one of my comments the authors say that “Brief note on the magnitude of vertical advection added. The comparison to the magnitude of the vertical advection in ERA5 added to the discussion section.” However, it is not obvious what has been changed and exactly where, since a) there have been various changes to different parts Section 3.4 and b) the entire Discussion section appears in blue in the version of the manuscript included with the references. Similar problems are encountered with many other responses as the authors do not give the lines corresponding to the changes in the manuscript. I would encourage them to consider this in future responses.

Response: We apologise for lack of clarity in the responses and will be more precise in the future. However, the “Discussion” section has been added during the revision, thus it appears as blue in whole.

Secondly, I would highly recommend proof-reading of the revised document by a native speaker as it still contains numerous language deficiencies (old and new).

Response: We have made another effort to correct language deficiencies in the manuscript. Hopefully, it will become even better after Copernicus language editing.

Specific points (referenced to the pages and lines in the manuscript version added to the responses):

On the response to my first comment of the initial review (which referenced Sofiev et al., 2015): Perhaps the authors have misunderstood this point, so I will rephrase it here: Can you demonstrate that the SILAM model is able to accurately reproduce stratospheric transport patterns including the overturning circulation, transport barriers, the QBO, etc.? Without sufficient evidence towards such a demonstration the frequency distribution underlying the both the SF6 mixing ratios and the mean AoA could be very unrealistic, which would in turn invalidate the SF6-AoA comparison i.e. a main point of this paper.

Response: Apologies for not being sufficiently clear. But SILAM does not compute atmospheric dynamics. Being an offline model, SILAM is only able to reproduce features that are reproduced by a meteorological model that is used to drive it. The Sofiev et al paper showed that SILAM is good at it - this is as much as one say about the model of its class. The ability of ERA-Interim (and SILAM driven with it) to reproduce stratospheric transport patterns including the overturning circulation, transport barriers, the QBO, etc. is certainly important but it is a separate study partly already done by other groups, e.g. Abalos et al. (2015) for Brewer-Dobson circulation.

Imperfection in reproducing stratospheric transport patterns by the model is certainly something to consider when interpreting the comparison results, but it can by no means invalidate the comparison.

On the response to my comment on line 273-275 of the initial review: Given that the absolute bias between MIPAS and SILAM appears to be constant until about 2008 but is increasing subsequently (Figure 7). Such an increasing bias would also affect the SILAM-based AoA (and also AoA trends). I therefore ask the authors to provide actual evidence for their claim that it could not “noticeably affect the results of the present study”.

Response: The overstating of the emissions is a likely reason for the bias. We added corresponding paragraph to the end of the “Evaluation of SF₆ against MIPAS data” subsection. This evaluation is probably the only result affected by the overstating.

On the response to my comment on line 547-548 of the initial review: As these balloon profiles were published with some uncertainties (and I am sure about three of the four), it should be possible to obtain these from the respective manuscripts, or if necessary through a FOIA request.

Response: The uncertainties are given in Fig. 5c. For Fig. 5a they are within 2% (1 σ)(Patra et al., 1997), and for Fig. 5b and Fig. 5d they are “smaller than the size of the symbol” (Ray et al., 2017). Corresponding note added to the figure caption.

Line 116-142: I appreciate the response to my comment on Line 119-113 of the initial manuscript. However, the information given in the response should be added to the revised version. Whether it is acceptable to cite two unpublished papers I leave for the editor to decide.

Response: The part of that response about MIPAS SF₆ retrievals has been added to the “MIPAS observations...” section. We would like to abstain from reviewing unpublished papers.

Line 266-67: Please explain what is meant by “The shooting method was used together with bisection”.

Response: Rephrased to “The shooting method with bisection was used...” The term seems to be quite common. It is explained e.g. here https://en.wikipedia.org/wiki/Shooting_method in details.

Line 348: Should this be “0.001 Kz”?

Response: No. The stated in the paper is correct.

Line 544-579: My count came to 27 mentions of “SF6” in this section, which is a bit excessive given that its title already indicates that it focuses on SF6.

Response: Reduced to 16 (incl. section caption).

Line 576-579: Higher mixing ratios in the stratosphere and lower mesosphere could indeed cause an “overstating” of simulated depletion rates. However, are the mixing ratios between 25 and 50 km from Kovacs et al. actually significantly different (i.e. outside the uncertainties) from the new MIPAS results presented here? This would especially be worth looking into since the new MIPAS SF6 mixing ratios seem to be higher than those in the old data product.

Response: As one can see from comparing Fig. 6 from the manuscript to Fig. 3 from (Kovács et al., 2017) (corresponding panels from the figures are comparable), that with new data the MIPAS values got indeed slightly higher, but the gap between model and measurements is still quite large.

Line 691: “ERA-Interim layers diagnosed from ERA-Interim.”

Response: Corrected.

3 Response to Anonymous Referee #4

accepted as is

Response: Thank you!

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Simulating age of air and the distribution of SF₆ in the stratosphere with the SILAM model

Rostislav Kouznetsov^{1,2}, Mikhail Sofiev¹, Julius Vira¹, and Gabriele Stiller³

¹Finnish Meteorological Institute, Helsinki, Finland

²Obukhov Institute for Atmospheric Physics, Moscow, Russia

³Karlsruhe Institute of Technology, Karlsruhe, Germany

Correspondence: Rostislav Kouznetsov (Rostislav.Kouznetsov@fmi.fi)

Abstract. The paper presents a comparative study of age of air (AoA) derived with several approaches: a widely used passive tracer accumulation method, the SF₆ accumulation, and a direct calculation of an “ideal age” tracer. The simulations were performed with the Eulerian chemistry transport model SILAM driven with the ERA-Interim reanalysis for 1980-2018.

The Eulerian environment allowed for simultaneous application of several approaches within the same simulation – and interpretation of the obtained differences. A series of sensitivity simulations revealed the role of the vertical profile of turbulent diffusion in the stratosphere, destruction of SF₆ in the mesosphere, as well as the effect of gravitational separation of gases with strongly different molar masses.

The simulations reproduced well the main features of the SF₆ distribution in the atmosphere ~~retrieved from~~ observed by the MIPAS satellite instrument. It was shown that the apparent very old air in the upper stratosphere derived from the SF₆ profile observations is a result of destruction and gravitational separation of this gas in the upper stratosphere and the mesosphere. These processes make the apparent SF₆ AoA in the stratosphere several years older than the “ideal-age” AoA, which, according to our calculations, does not exceed 6-6.5 years. The destruction of SF₆ and the varying rate of emission make SF₆ unsuitable for reliably deriving AoA or its trends. However, observations of SF₆ provide a very useful ~~means~~ dataset for validation of the stratospheric circulation in a model with the properly implemented SF₆ loss.

15 1 Introduction

~~AoA~~ The age of air (AoA) is defined as the time spent by an air parcel in the stratosphere since its entry across the tropopause (Li and Waugh, 1999; Waugh and Hall, 2002). The distribution of the ~~age of air (AoA)~~ AoA is controlled by the global atmospheric circulations, first of all, the Brewer-Dobson and the polar circulation. In particular, the temporal variation of AoA has been used as an indicator of the long-term changes in the stratospheric circulation (Engel et al., 2009; Waugh, 2009). AoA has been extensively used ~~to evaluate and compare~~ for evaluation and comparison of general circulation and chemical transport models in the stratosphere (Waugh and Hall, 2002; Engel et al., 2009).

Simulations of the AoA ~~according to the definition as defined~~ above have been performed with Lagrangian transport models. The trajectories are initiated with positions distributed in the stratosphere and integrated backwards in time until they cross the tropopause. The time elapsed since the initialization is attributed as age of air at the point of initialization. Moreover, the

25 distribution of the ages of particles originating from some location can be used to get the age spectrum there. Until recently
Lagrangian simulations of AoA did not explicitly account for turbulent mixing in the stratosphere (Eluszkiewicz et al., 2000;
Waugh and Hall, 2002; Diallo et al., 2012; Monge-Sanz et al., 2012). ~~The account~~ Accounting for mixing adds up to two years
to the mean AoA in the tropical upper stratosphere (Garny et al., 2014). In Lagrangian ~~formulation-models~~, the mixing can be
simulated with random-walk of the particles (Garny et al., 2014), or by inter-parcel mixing (Plöger et al., 2015; Brinkop and
30 Jöckel, 2019).

The Eulerian simulations of AoA can be formulated in several ~~different~~ ways. The approaches with an accumulating tracer,
whose mixing ratio increases linearly in the troposphere, were used in a comprehensive study by Krol et al. (2018) and several
studies before (e.g. Eluszkiewicz et al., 2000; Monge-Sanz et al., 2012). Another approach is to simulate a steady distribution of
a decaying tracer, such as ^{221}Rn , emitted at the surface at a constant rate (Krol et al., 2018). Besides that, a special tracer that is
35 analogous to ~~a~~ the Lagrangian clock has been used. The tracer appears in the literature under names “clock-type tracer” (Monge-
Sanz et al., 2012) or “ideal age” (Waugh and Hall, 2002). The ideal age has a constant rate of increasing of mixing ratio
everywhere, except for the surface where it is continuously forced to zero. Similar tracers have been long used to simulate the
transport times of oceanic water (e.g. England, 1995; Thiele and Sarmiento, 1990).

Direct observations of the age of air, as it is defined above, are not possible; therefore AoA is usually derived from the
40 observed mixing ratios of various tracers ~~-.The tracers belong to one of two types: various tracers-~~ with known tropospheric
mixing ratios and lifetimes (Bhandari et al., 1966; Koch and Rind, 1998; Jacob et al., 1997; Patra et al., 2011), ~~and or from the~~
long-living tracers with known variations in the tropospheric mixing ratios. The studies published to-date used carbon dioxide
 CO_2 (Andrews et al., 2001; Engel et al., 2009), nitrous oxide N_2O (Boering et al., 1996; Andrews et al., 2001), sulphur
hexafluoride SF_6 (Waugh, 2009; Stiller et al., 2012), methane CH_4 (Andrews et al., 2001; Remsberg, 2015), and various
45 fluorocarbons (Leedham Elvidge et al., 2018).

For accumulating tracers, the mean AoA at some point in the stratosphere is calculated as a lag between the times when a
certain mixing ratio is observed near the surface and at that point. The lag time is equivalent to the mean AoA defined above
only in the case of ~~a~~ the strictly linear growth and the uniform distribution of the tracer in the troposphere (Hall and Plumb,
1994).

50 In reality, there is no tracer whose mixing ratio in the troposphere grows strictly linearly. The violation of the assumption
of the linear growth leads to biases in the resulting AoA distribution and its trends. It has been pointed out that the increasing
growth rates of CO_2 and SF_6 lead to a low-bias of AoA and its trends, and make these tracers ambiguous proxies ~~for of the~~
AoA (Garcia et al., 2011). Various corrections have been applied in several studies (Hall and Plumb, 1994; Waugh and Hall,
2002; Engel et al., 2009; Stiller et al., 2012; Leedham Elvidge et al., 2018) to deduce the “true” AoA from observations of
55 tracers with the increasing growth rates. The effect of the correction method on the AoA estimates has not been investigated and
must be considered as a source of uncertainty in ~~resulting estimates (Garcia et al., 2011). Garcia et al. (2011) further conclude~~
the resulting estimates. Thus, Garcia et al. (2011) concluded that accounting for the biases in the trend estimates due to varying
growth rates would likely require uniform and continuous knowledge of the evolution of the trace species, which is not available
from any existing observational dataset. Recently Leedham Elvidge et al. (2018) ~~have shown~~ showed a minor sensitivity of

60 ~~the~~ AoA to the choice of ~~particular~~ ~~the~~ correction method, however without detailed analysis of ~~the~~ assumptions behind these methods. For ~~a~~ similar problem with ~~the~~ ages of oceanic water it has been shown (Waugh et al., 2003) that in case of a ~~non-linearly-varying-inhomogeneously-growing~~ tracer the tracer age is strongly influenced by the shape of ~~the~~ transient time distribution (TTD, also known as ~~an~~ ~~the~~ “age spectrum”) at ~~the~~ particular location and time.

Another major source of uncertainty in ~~the~~ observational AoA is ~~the~~ violation of conservation of ~~a~~ ~~the~~ tracer due to sources
65 and sinks, such as oxidation of carbon monoxide and methane for CO₂ ~~;~~ or mesospheric destruction for SF₆. The mesospheric sink of SF₆ leads to ~~an~~ ~~the~~ “over-aging” ~~;~~ especially pronounced in the area of ~~the~~ polar vortices. The magnitude of the over-aging was estimated as ~~2-or-more-at least two~~ years Waugh and Hall (2002). Besides being visible in many evaluations, e.g. Stiller et al. (2012, Fig. 4), Kovács et al. (2017, Fig. 8), ~~a-dedicated-study-on~~ the over-aging of ~~the~~ polar winter stratospheric air was ~~performed-by-Ray-et-al.~~ (2017, Fig. 4) ~~studied by Ray et al. (2017, Fig. 4) within the dedicated exercise.~~

70 The simulations of SF₆ and ~~the~~ AoA in the atmosphere with WACCM model (Kovács et al., 2017) have ~~also~~ reproduced the effect of over-aging, ~~but-of-much-smaller-magnitude-than-if-inferred-from.~~ However, ~~its magnitude was much smaller than that inferred from the~~ SF₆ retrievals ~~from-of~~ the limb-viewing ~~MIPAS MIPAS~~ instrument operated on-board of the Envisat satellite in 2002-2012 (Stiller et al., 2012), and ~~from the in-situ~~ observations ~~from-of~~ the ER-2 aircraft (Hall et al., 1999). Kovács et al. (2017) offered two possible ~~scenarios-reasons~~ for the discrepancy: either SF₆ loss is still underestimated in WACCM,
75 or MIPAS SF₆ ~~is-observations are~~ low biased above ~ 20 km. Neither of the ~~scenarios-cases~~ have been analysed in depth ~~so far~~, which leaves the status of MIPAS, ~~the-richest-to-date-currently the richest~~ observational dataset for the stratospheric SF₆, unclear.

The aim of the present study is to provide self-consistent simulations of spatio-temporal distribution of ~~the~~ AoA and of the SF₆ mixing ratio in the troposphere and ~~the~~ stratosphere during last 39 years. The main modelling tool is the Eulerian chemistry
80 transport model SILAM (~~baekronym-for~~ System for Integrated modelLling of Atmospheric coMposition). The stratospheric balloon observations and retrievals ~~from-of~~ the limb-viewing ~~MIPAS instrument-operated-on-board-of-the-Envisat-satellite-in 2002-2012 are used as a validation for~~ ~~MIPAS instrument said above are used for validation of~~ the simulated distribution.

With ~~the-results-of~~ these simulations we

- compare different methods of estimating the AoA and quantify ~~inconsistencies-in~~ ~~the inconsistencies in the~~ AoA and its
85 trends arising from violations of the underlying assumptions behind each method
- analyze the causes of the discrepancies in the upper stratosphere between different methods of deriving the AoA
- provide a solid basis for further studies of stratospheric circulation with observations of various trace gases and for studies of climate effects of SF₆

The paper is organized as follows. Sec. 2 gives an overview of the modelling tools ~~;~~ and the modelling and observational
90 data used for the study. Sec. 3 describes the developments made for SILAM in order to perform the simulations: vertical eddy diffusivity parametrisation in the stratosphere and the lower mesosphere, and ~~the~~ SF₆ destruction parametrization, as well as the modelling setup. The sensitivity tests and evaluation of the simulations against ~~MIPAS-satellite-retrievals~~ ~~;~~ ~~the MIPAS~~

retrievals and stratospheric-balloon measurements of SF₆ mixing ratios are given in Sec. 4. Sensitivity of the AoA and its trends to the ~~choice-of-the~~ simulation setup and the AoA proxy is studied in Sec. 5. The ~~findings-of-the-whole-study-are~~ study is summarised in Sec. 7.

2 Methods and input data

2.1 SILAM model

SILAM (System for Integrated modeLLing of Atmospheric coMposition, <http://silam.fmi.fi>) is an ~~off-line~~ offline chemical-transport model. SILAM features a mass-conservative and positive-definite advection scheme that makes the model suitable for long-term runs (Sofiev et al., 2015). The model can be run at a range of resolutions starting from a kilometer scale in limited-area ~~or-in-global-mode~~ and up to a global coverage. The vertical structure of the modelling domain consist of stacked layers starting from the surface. The layers can be defined either in z- or hybrid sigma-pressure coordinates. The model can be driven with a variety of NWP- (numerical weather prediction) or climate models.

In order to accurately model the AoA and the needed tracers, the vertical diffusion part of the transport scheme of SILAM has been refined to account for gravitational separation. In addition, several tracers with corresponding transformation ~~and transport~~ routines have been implemented into the model. The model setup used for the present study is described in Sec. 3.5.

2.2 ECMWF ERA-interim reanalysis

The ERA-Interim reanalysis ~~from-of~~ the European Centre for Medium-range Weather Forecasts (ECMWF) had been used as a meteorological driver for our simulations. The ~~data-set~~ dataset has T255 spectral resolution and covers the whole atmosphere with 60 hybrid sigma-pressure levels (~~Dee et al., 2011~~), having the uppermost layer from 0.2 to 0 hPa with nominal pressure of 0.2 hPa (Dee et al., 2011). The reanalysis uses a 12-h data assimilation cycle, and the forecasts are stored with a 3-hour time step. We used the fields retrieved ~~form-from~~ the ECMWF's MARS archive on a lat-lon grid 500x250 points with the step of 0.72 degrees. The four forecast times (+3h, +6h, +9 h and +12h) were used from every assimilation cycle to obtain a continuous dataset with 3-hour time step. To drive the dispersion model, the data on horizontal winds, temperature and humidity for 1980-2018 were used. The procedure for diagnosing the vertical transport is ~~described~~ described in Sec. 3.5.

The ERA-interim reanalysis has been used earlier for Lagrangian simulations of AoA (Diallo et al., 2012) and found to provide ages that agree with those inferred from ~~in-situ~~ in-situ observations in the lower stratosphere.

2.3 MIPAS observations of SF₆

To evaluate the results of the SF₆ modelling we used the data from the MIPAS instrument operated on-board of Envisat satellite in 2002-2012. MIPAS was a limb-sounding Fourier transform spectrometer with a high spectral resolution measuring in the infrared part of spectrum. Due to its limb geometry, ~~a-the instrument provided~~ good vertical resolution of the derived trace gas profiles and ~~a-showed~~ high sensitivity to low-abundant species around the tangent point ~~has-been-achieved~~. Along the orbit

path, MIPAS measured a profile of atmospheric radiances about every 400 km with an altitude coverage, in its nominal mode, of about 6–70 km from 6 to 70 km. The vertical sampling was 1.5 km in the lower part of the stratosphere (up to 32 km) and 3 km above, with a vertical field of view covering 3 km at the tangent point. Over a day, about 1300 profiles along 14.4 orbits were measured, covering all latitudes up to the poles at sunlit and dark conditions. The vertical distributions of trace gases were derived from the radiance profiles by an inversion procedure, fitting simulated spectra to the measured ones while varying the atmospheric state parameters.

The retrieval of SF₆ is based on the spectral signature of this species in the vicinity of 10.55 μm wavelength and is in principle described in Stiller et al. (2008, 2012); Haenel et al. (2015). In this study here the current study, we use an updated version of the SF₆ data (compared to the one described in Haenel et al. (2015)) called V5H/R_SF6_21/224/225; the 225. The new algorithm uses the new absorption cross-section data on the SF₆ and a new CFC-11 band in the vicinity of the SF₆ signature by Harrison (2018) has been used instead of the older cross section data by Varanasi et al. (1994). The updated version provides considerably up to 0.6 pptv higher SF₆ mixing ratios in the upper part of the stratosphere (above 30 km) than the versions before old versions and is closer to independent reference data.

The retrieved profiles are sampled on an altitude grid spaced at 1 km, where as whereas the actual resolution of the profiles is between 4 and 10 km for altitudes below 30 km. The retrievals are supplemented with averaging kernels and error covariance matrices describing the uncertainties due to random noise in the radiance measurements, called hereinafter referred as measurement noise error or target noise error or retrieval noise error in the following. This error component, which is normally in the order of 10% of the retrieved value, is fully uncorrelated from profile to profile, and therefore virtually cancels out when averaging averaged over a large number of profiles. In contrast, there exist systematic error components that are fully correlated between the profiles. Their assessment is difficult and depends on the knowledge about sources of systematic errors. Stiller et al. (2008) has assessed them to be in the order of 10% at 60 km, and 4% at 30 km. These error components have to be considered when comparisons of larger datasets (monthly or seasonal means) with other data are performed.

3 SILAM developments

The destruction of Destruction of the atmospheric SF₆ occurs at altitudes above 60 km (Totterdill et al., 2015) that fall within the topmost layer of the ERA-Interim. The exchange processes in the upper stratosphere and lower mesosphere have to be adequately parameterized together with the destruction process. In our simulations we have suppressed the transport with of SF₆ mean wind above with mean wind through the modelling domain top (0.1 hPa, 65 km) and parameterized the SF₆ loss due to the eddy and molecular diffusion towards the altitudes where the destruction occurs. In this section we introduce the set of parametrizations that were implemented in SILAM for this study.

3.1 SF₆ destruction

Estimates of AoA from the SF₆ tracer rely on the assumption of it being a passive tracer. SF₆ is indeed essentially stable in the troposphere and the stratosphere. IPCC (2013, Sec 8.2.3.5) mentions that photolysis in the stratosphere as the main

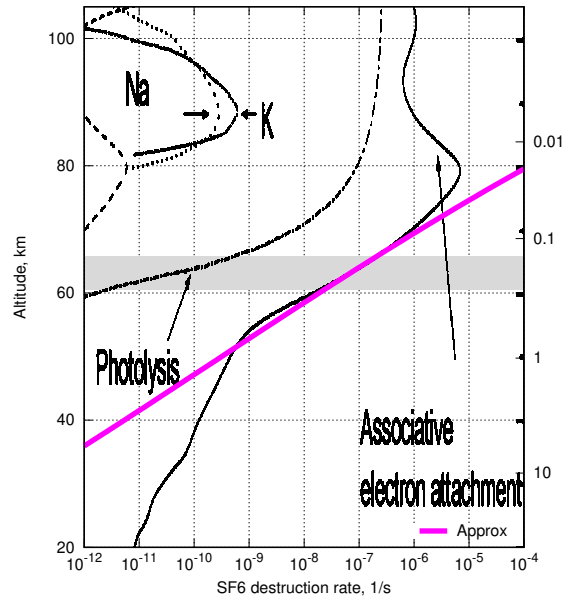


Figure 1. The vertical profiles of SF_6 destruction rate (after Totterdill et al., 2015) and its approximation in range of 55-75 km, given by Eq. (1).

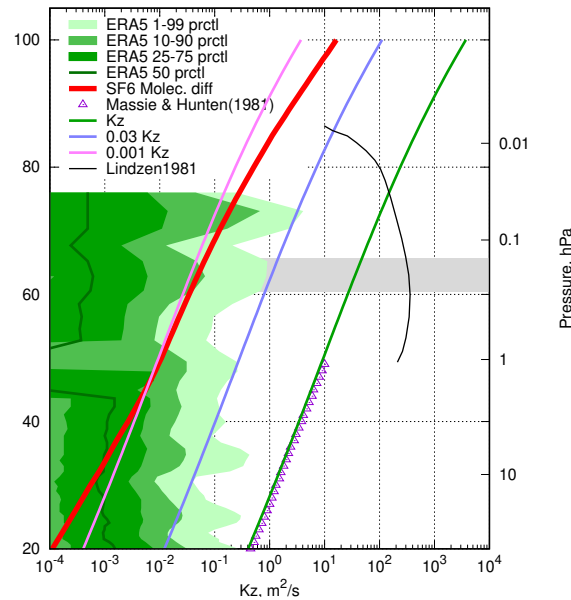


Figure 2. Vertical profiles of diffusion coefficients. The distribution of [the](#) ERA5 profiles of the “mean turbulent diffusion coefficient for heat” parameter, molecular diffusivity for SF_6 in [the](#) standard atmosphere, and [the](#) three prescribed K_z profiles. The eddy diffusion profile due to breaking gravity waves (after Lindzen, 1981) is given for [a](#) [the](#) reference.

155 mechanism of SF₆ loss, however without any reference to original studies. The statement is probably taken from Ravishankara et al. (1993). Reddmann et al. (2001) pointed at associative electron attachment in the upper stratosphere and mesosphere as the main destruction mechanism for SF₆ below 80 km. The recent study of Totterdill et al. (2015) gives some 1-2 order of magnitude slower rates of electron attachment, however keeping it the dominant mechanism of the SF₆ destruction in the altitude range up to 100 km. The highest destruction rate of $1 \times 10^{-5} \text{ s}^{-1}$ occurs at the altitude of 80 km (Fig. 1). An important
 160 feature of this profile is that the destruction rate becomes significant above the top of our modelling domain (0.1 hPa, 65 km). The ERA-Interim meteorological fields have the uppermost level at 0.1 hPa and do not resolve ~~a~~the vertical structure of the atmosphere above that level. In order to assess the loss of SF₆ ~~due to destruction~~ we have to parameterise the combined effect of ~~transport of~~the SF₆ transport through the 0.1 hPa and its destruction. Then the resulting fluxes can be applied as the upper boundary condition for our simulations.

165 As an approximation to the vertical profile of the destruction rate in an altitude range of 50–80 km we have fitted ~~a~~the corresponding part of the curve in Fig. 9a of Totterdill et al. (2015) with a power function of pressure (magenta line in Fig. 1):

$$\frac{1}{\tau} = 3 \times 10^{-8} \text{ s}^{-1} \left(\frac{0.2 \text{ hPa}}{p} \right)^3, \quad (1)$$

where τ is the lifetime of SF₆ at the altitude corresponding to pressure p .

3.2 Eddy diffusivity

170 Large variety of vertical profiles for eddy diffusivity in the stratosphere and the lower mesosphere can be found in literature. In many studies in 1970-s – 1980-s the vertical profiles were derived from observed tracer concentrations neglecting the mean transport. Most studies suggested that the vertical eddy diffusion has a minimum of 0.2-0.5 m²/s (Pisso and Legras, 2008) at 15-20 km agreeing quite well to the ones derived from the radar measurements in the range of 15-20 km Wilson (2004). Above that altitude K_z was suggested to gradually ~~increases~~increase by about 1.5 orders of magnitude ~~at~~towards 50 km due
 175 to breaking gravity waves (Lindzen, 1981).

The theoretical estimates of the effective exchange coefficients considering the layered and patchy structure of stratospheric turbulence suggest 0.5–2.5 m²/s for the upper troposphere and 0.015–0.02 m²/s for the lower stratosphere (Osman et al., 2016), which is about an order of magnitude lower than the estimates above.

The values of the eddy exchange coefficient at heights of 10-20km estimated from the high-resolution balloon temperature
 180 measurements (Gavrilov et al., 2005) are $\sim 0.01 \text{ m}^2/\text{s}$ with no noticeable vertical variation. It is not clear, however, how representative the derived values are for UTLS in general. We could not find any reliable observations of vertical diffusion in a range of 30-50 km.

The parameterisation for vertical eddy diffusivity above the boundary layer used in SILAM has been adapted from the IFS model of the European Centre for Medium-range Weather Forecasts (ECMWF, 2015). However, in the upper troposphere
 185 the predicted eddy diffusivity is nearly zero. For numerical reasons a lower limit of $0.01 \text{ m}^2/\text{s}$ is set for K_z in SILAM. Our sensitivity tests have shown that long-term simulations are insensitive to this limit as long as it is low enough (see results and discussion). The K_z in the stratosphere is routinely set to the limiting value with relatively rare peaks, mostly in UTLS.

Such scheme essentially turns off turbulent diffusion in the stratosphere. Same is true for [the](#) recent ERA5 reanalysis dataset (Copernicus Climate Change Service (C3S), 2017) that provides the values of K_z among other model-level fields: the eddy
190 diffusion routinely falls below the molecular diffusivity above 40 km (Fig. 2).

As a reference for this study, we took a tabulated profile of Hunten (1975), as it was quoted by Massie and Hunten (1981). The original profile covers the range up to 50 km, and the extrapolation up to 80 km matches the theoretical estimates by (Lindzen, 1981) and by Allen et al. (1981). We approximate the profile as a function of pressure in the range of 100 – 0.01 hPa (15 – 60 km):

$$195 \quad K_z(p) = 8 \text{ m}^2/\text{s} \left(\frac{1 \text{ hPa}}{p} \right)^{0.75}. \quad (2)$$

The approximated profile was stitched with the default SILAM profile with a gradual transition within an altitude range of 10 – 15 km to keep the tropospheric dispersion intact. This profile gives values of K_z ~~is~~ 3 – 6 orders of magnitude higher than [the](#) ones provided by [the](#) ERA5 reanalysis (Fig. 2), and 1-2 orders of magnitude higher than ~~more recent estimates~~ [the estimates of](#) (Legras et al., 2005).

200 In order to cover the range of ~~vertical profiles of~~ K_z between [the](#) ERA5 profiles and the reference one (2) we used two intermediate profiles obtained by scaling the reference one with factors 0.03 and 0.001. The three prescribed eddy-diffusivity profiles are hereinafter referred as “1Kz”, “0.03Kz”, and “0.001Kz”, respectively. The dynamic eddy-diffusivity profile adopted from the ECMWF IFS ~~model~~ is referred to as “ECMWF Kz”. In all simulations, [the](#) parameterization of K_z in the troposphere is the same ~~;~~ and linear transition from the SILAM K_z to the prescribed one occurs in the altitude range of 10 – 15 km.

205 3.3 Molecular diffusivity and gravitational separation

In tropospheric and stratospheric CTMs, [gaseous admixtures](#) are transported as tracers, i.e. advection and turbulent mixing do not depend on ~~a~~ [the](#) species properties, whereas the molecular diffusion is negligible. Models that cover the mesosphere, such as WACCM (Smith et al., 2011), account for molecular diffusion explicitly. Since some of the K_z parametrizations ~~above of~~ [the previous section](#) often result in values below the molecular diffusivity, the parametrization of molecular diffusion has been
210 implemented in SILAM.

The molecular diffusivity of SF₆ in the air at temperature $T_0 = 300$ K and pressure $p_0 = 1000$ hPa, is $D_0 = 1 \times 10^{-5} \text{ m}^2 \text{ s}^{-2}$ (Marrero and Mason, 1972, Table 22). The diffusivity at ~~a~~ different temperature T and pressure p is given by:

$$D = D_0 \frac{p_0}{p} \left(\frac{T}{T_0} \right)^{3/2}, \quad (3)$$

see e.g. Cussler (1997). The vertical profile of molecular diffusivity in the US standard atmosphere (NOAA et al., 1976) is
215 shown in (Fig. 2). Note that the value for the reference diffusivity of SF₆ used in this paper is about a half of the one used in simulations with WACCM by Kovács et al. (2017). The reason is that WACCM uses a universal parametrization (Smith et al., 2011, Eq. 7 there) for all compounds. That parametrization relies solely on molecular mass of a tracer and does not account for e.g. the molecule collision radius. The latter is about twice larger for SF₆ ~~molecule~~ than for most of stratospheric tracers.

Thus, for this study we use the value from Marrero and Mason (1972), which results from fitting laboratory data for diffusion of SF₆ in the air.

The vertical diffusion transport velocity of admixture with number concentration \tilde{n} and molecular mass $\tilde{\mu}$ in neutrally-stratified media is given by (Mange, 1957):

$$w = -D \left[\frac{1}{\tilde{\mu}} \frac{\partial \tilde{\mu}}{\partial z} + \left(\frac{\tilde{\mu}}{\mu} - 1 \right) \frac{\mu g}{kT} \right], \quad (4)$$

where μ is molecular masses of air, g – acceleration due to gravity, k is the Boltzmann constant, and T is temperature. With ideal gas law $p = nkT$, in which p is pressure, and n is number concentration, and static law $dp/dz = -g\rho$, where $\rho = \mu n$ is the-air density, the equation (4) can be reformulated in terms of the-admixture mixing ratio $\xi = \tilde{n}/n$ and pressure. Then the vertical gradient of the equilibrium mixing ratio will be:

$$\frac{\partial \xi}{\partial p} = \left(\frac{\tilde{\mu}}{\mu} - 1 \right) \frac{\xi}{p}. \quad (5)$$

It is non-zero for an admixture of a molecular mass different from one of the air. Integrating the gradient (5) over vertical, one can obtain that the equilibrium mixing ratios ξ_1 and ξ_2 at two levels with corresponding pressures p_1 and p_2 are related as:

$$\frac{\xi_1}{\xi_2} = \left(\frac{p_1}{p_2} \right)^{\tilde{\mu}/\mu - 1}. \quad (6)$$

For heavy admixtures, such as SF₆ ($\tilde{\mu} = 0.146$ kg/mole) the equilibrium gradient of a mixing ratio is substantial. For example, the difference of the equilibrium mixing ratio of SF₆ in-the-atmosphere between 0.1 and 0.2 hPa is a factor of 16.

In most of the atmosphere, the effect of gravitational separation is insignificant due to the overwhelming effect of other mixing mechanisms, whereas in the upper stratosphere the molecular diffusivity may become significant. Therefore, in the upper stratosphere heavy gases can no longer be considered as tracers and the molecular diffusion should be treated explicitly. The effect of gravitational separation of nitrogen and oxygen isotopes in the stratosphere has been observed (Ishidoya et al., 2008, 2013; Sugawara et al., 2018), however for isotopes the ratio of masses is relatively small, so the observed differences were also small (up to 10^{-5}). For SF₆ the molecular mass difference is much larger.

In order to enable the gravitational separation in SILAM we have introduced a-the molecular diffusion mechanism, which can be enabled along with the turbulent diffusion scheme. The exchange coefficients due to molecular diffusion between the model layers are pre-calculated according to Eq. (4) discretized for the given layer structure for each species according to its diffusivity and molar mass. The US standard atmosphere (NOAA et al., 1976) was assumed for the vertical profiles of temperature and air density during pre-calculation of the exchange coefficients. The exchange has been applied throughout the domain at every model time step with a simple explicit scheme.

3.4 Parametrization for destruction of SF₆ in the mesosphere

As it-has-been-mentioned above, the topmost level of the ERA-Interim meteorological data-set-dataset is located at 0.1 hPa, which is below the layer where the destruction of SF₆ occurs. Therefore we have to put a boundary condition to our simulations

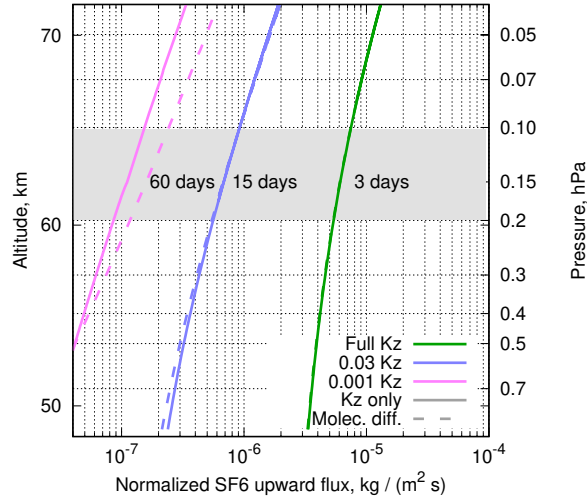


Figure 3. Vertical profiles of steady-state upward flux of SF₆ normalized with mass mixing ratio $F(p)/\xi(p)$, for eddy diffusivity and lifetime profiles given by (2) and (1). The upper model layer of SILAM and effective lifetimes of SF₆ there due to the destruction in the mesosphere for different K_z profiles are given.

to account for the upward flux of SF₆ through the upper boundary of the simulation domain. For that we assume that SF₆ distribution above the computational domain [top](#) is in equilibrium with ~~destruction-and-the destruction~~ [the destruction and the](#) vertical flux.

Assuming the profiles for $K_z(p)$ and the SF₆ lifetime $\tau(p)$ are given by (2) and (1), one can obtain a steady-state distribution of [the](#) mass-mixing ratio ξ of SF₆ due to destruction in the mesosphere at any point where both (2) and (1) are valid and vertical advection is negligible. The latter assumption implies that the diffusive vertical flux overwhelms the advective one. The validity and implications of neglecting the regular vertical transport are discussed below. The steady-state profile of ξ can be obtained from a solution of ~~a-the~~ [the](#) steady-state diffusion equation with a sink:

$$\frac{\partial \xi}{\partial t} = g \frac{\partial}{\partial p} (F) - \frac{\xi}{\tau(p)} = 0, \quad (7)$$

where $\rho(p)$ is air density, and g is acceleration due to gravity, and the upward flux of SF₆ is given by

$$F(p) = g \rho^2 K_z(p) \frac{\partial \xi}{\partial p} \quad (8)$$

The above equation was solved numerically as a boundary value problem with unit mixing ratio at a height of 1 hPa and vanishing flux $F(p)$ at $p = 0$ for the set of K_z profiles. The shooting method ~~was-used-together-with-bisection~~ [with bisection](#) [was used](#) to get the steady-state profiles of $\xi(p)$ and $F(p)$, corresponding to $\xi(1 \text{ hPa}) = 1$. For all considered cases the flux $F(p)$ decreased by several orders of magnitude already at the level of a few Pa, i.e. below the maximum of the depletion profile of Totterdill et al. (2015), indicating that particular shape of $\tau(p)$ above that level does not influence the fluxes at the domain top (0.1 hPa). The steady-state upward flux of SF₆ $F(p)$ normalized with [the](#) corresponding mixing ratio at each pressure $F(p)/\xi(p)$, for the three test profiles of K_z is shown in Fig. 3 with solid lines.

The gravitational separation can be accounted for by introducing ~~into the vertical flux Eq. (8)~~ a term responsible for molecular diffusion and its equilibrium state (5) into the vertical flux Eq. (8) :

$$F(p) = g\rho^2 K_z(p) \frac{\partial \xi}{\partial p} + g\rho^2 D(p) \left(\frac{\partial \xi}{\partial p} - \frac{\tilde{\mu} - \mu}{\mu} \frac{\xi}{p} \right) \quad (9)$$

The profiles of $F(p)/\xi(p)$ resulting from ~~this term~~ $F(p)$ in ~~the~~ equation (7) are given in Fig. 3 with dashed lines. The magnitude
 270 of $F(p)/\xi(p)$ gives an equivalent regular vertical air-mass flux that would result in the same vertical flux of SF_6 if it were passive and non-diffusive. The equivalent regular vertical velocity ω_{eq} (in units of the Lagrangian tendency of a parcel pressure due to vertical advection) can be expressed as:

$$\omega_{eq} = -gF(p)/\xi(p). \quad (10)$$

Accounting for molecular diffusion may either enhance or reduce the upward flux of SF_6 in the model. Along with setting
 275 the equilibrium state with the bulk of a heavy admixture being in the lower layers, molecular diffusion provides additional means for transport to the upper layers where the destruction occurs. For very low eddy diffusivities, the molecular diffusion is a sole mechanism of the upward transport of SF_6 towards depletion layers. For higher eddy diffusivity the effect of molecular diffusion and gravitational separation becomes negligible.

For ~~a the~~ model consisting of stacked well-mixed finite layers, the loss of SF_6 from the topmost layer due to the steady
 280 upward flux would be proportional to the SF_6 mixing ratio in the layer. This loss of mass is equivalent to a linear decay of SF_6 in the layer at a rate

$$\tau^{-1} = g \frac{F(p)}{\xi(p) \Delta p}, \quad (11)$$

where Δp is ~~a~~ pressure drop in the layer.

~~For In~~ the upper layer of our simulations (between 0.1 hPa and 0.2 hPa, grey rectangle in Fig. 3), ~~and $K_z(p)$ given with~~
 285 ~~Eq. (2)~~, the SF_6 lifetime τ due to turbulent diffusion is about 3 days for K_z of Eq.(2). After scaling the $K_z(p)$ profile with factors of 0.03, and 0.001 one gets the lifetimes of 15 and 60 days, respectively. Noteworthy, the molecular diffusion sets the upper limit to the SF_6 lifetime in the topmost model layer: it can not be longer than 60 days for the 0.1 - 0.2 hPa layer. Close to this regime, the system becomes insensitive to the actual profile and values of the turbulent diffusion coefficient. The loss of SF_6 through the domain top was implemented as a linear decay of SF_6 in the topmost model layer, at a rate corresponding to
 290 the $K_z(p)$ profile used in each simulation.

3.5 Simulation setup

The ~~simulations of atmospheric transport were performed with the SHAM model~~ atmospheric transport simulations were performed for 1980-2018 years on a 1.44x1.44 degree global grid with 60 hybrid sigma-pressure layers starting from surface, with the uppermost layer between pressures of 0.1 and 0.2 hPa. The model time step ~~of was~~ 15 minutes ~~was used~~ and the output
 295 consisted of daily mean concentrations of tracers together with air density ~~was arranged~~.

The simulations were driven with the ERA-interim meteorology at 0.72-degree resolution, so the meteorological input for both cell-interface for winds, and cell mid-points for other parameters (surface pressure, temperature and humidity) was available without ~~further~~-interpolation. The gridded ERA-interim fields are, however, a result of reprojection of the original meteorological fields from spherical harmonics. Moreover, differences in the ~~representation of~~ model vertical structure ~~between~~
 300 ~~of~~ IFS and SILAM ~~make-a-made~~ vertical reprojection necessary. These reprojections together with a limited precision of the gridded fields and inevitable small differences in physical parametrizations between IFS and SILAM result in inconsistency between the surface-pressure tendencies and the vertically-integrated air-mass fluxes calculated from the meteorological fields in SILAM. Such inconsistencies cause spurious variations in wind-field divergence that on long-term run result in accumulation of errors in tracer mixing ratios, and consequently, in the simulated AoA. Therefore, horizontal wind fields were adjusted
 305 by distributing the residuals of pressure tendency and vertically-integrated horizontal air-mass fluxes as a correction to the horizontal winds following the procedure suggested by Heimann and Keeling (1989). The correction is of the order of centimeters per second, which is comparable to the precision of the input wind fields. The vertical wind component was then re-diagnosed from ~~a divergence of the~~ divergence of the horizontal air-mass fluxes for the individual SILAM layers as described in Sofiev et al. (2015).

310 SILAM performs the 3D transport by means of a dimension split: transport along each dimension is performed separately as 1D transport. To minimize the inconsistency between the tracer transport and air-mass fluxes ~~;~~ caused by the dimension split at finite time step, the splitting sequence has been inverted at each time step ~~to reduce the accumulation of errors~~. The residual inconsistency was resolved by using a separate unity tracer, which was initialized to the constant mass mixing ratio of 1 at the beginning of a simulation. ~~If advection was~~ Should advection be perfect, the concentration of ~~unity~~ the unity tracer would be
 315 equivalent to air density (mixing ratio would stay equal to 1). The mixing ratios of the simulated tracers were then evaluated as a ratio of ~~a~~ the tracer mass in a cell to the mass of ~~unity~~ the unity tracer.

In order to assess the effects of gravitational separation and destruction on the atmospheric distribution of SF₆, we ~~have~~ used four tracers: SF₆ as a passive tracer “sf6pass”, SF₆ with gravitational separation but no destruction “sf6nochem” (no chemistry), SF₆ with destruction but no gravitational separation “sf6nograv”, and SF₆ with both gravitational separation and
 320 destruction in the upper model level “sf6”.

All SF₆ tracers had the same emission according to the SF₆ emission inventory (Rigby et al., 2010). The inventory covers 1970-2008, and was extrapolated with a linearly growing trend of 0.294 Gg/y/y until July 2016. The last 2.5 years were run without the SF₆ emissions to evaluate its destruction rate. Note ~~;~~ that the emission extrapolation gives for 2016 9.4 Gg/y, which is somewhat higher than later estimate 8.8 Gg/y (Engel et al., 2018).

325 Besides the four SF₆ tracers we ~~have~~-used a “passive” tracer emitted uniformly at the surface at constant rate during the whole simulation time and an “ideal age” tracer. The “ideal age” tracer is defined as a tracer whose mixing ratio ξ_{ia} obeys continuity equation (Waugh and Hall, 2002):

$$\frac{\partial \xi_{ia}}{\partial t} + \mathcal{L}(\xi_{ia}) = 1, \quad (12)$$

(where \mathcal{L} is ~~an~~-advection-diffusion operator), and boundary condition $\xi_{ia} = 0$ at the surface. The “ideal age” tracer is trans-
 330 ported as a regular gaseous tracer ~~-, and to maintain consistency with other tracer mixing ratios, the ideal age is~~ and updated at
 every model time step Δt ~~using~~ with the unity tracer correction:

$$M_{ia} \mapsto \begin{cases} 0, & \text{at lowest layer,} \\ M_{ia} + M_{\text{unity}} \Delta t, & \text{otherwise,} \end{cases} \quad (13)$$

where M_{ia} and M_{unity} are masses of the “ideal age” tracer and of the unity tracer in ~~a~~ the grid cell. The mixing ratio of the
 “ideal age” tracer is a direct measure of the mean age of air in a cell, so the tracer is a direct Eulerian analog of the time-tagged
 335 Lagrangian particles with clock reset at the surface. Note that the AoA derived from the “ideal age” tracer and AoA from a
 passive tracer with a linearly-growing near-surface mixing ratio are equivalent (Waugh and Hall, 2002), and implementation
 of both provides a redundancy needed to ensure self-consistency of our results.

~~A set of the simulations was~~ The simulations were performed with four ~~settings for the eddy diffusivity profile within the~~
~~model domain, eddy diffusivity profiles~~ described in Sec. 3.2 and the corresponding destruction rates of “sf6” and “sf6nograv”
 340 tracers in the uppermost model layer. All runs were initialized with the mixing ratios from the final state of a special initial-
 ization run. The initialization simulation with “0.1Kz” eddy diffusivity was started from 1970 with zero fields for all tracers,
 except for unity tracer that was set to unity mixing ratio. The simulation ~~was run with~~ used 1970-1989 emissions for SF₆
 species from the same inventory as for the main runs (Rigby et al., 2010), and driven with the twice repeated ERA-Interim
 meteorological fields for 1980-1989. The mixing ratios of all SF₆ tracers at the end of the initialization run were scaled to
 345 match the total SF₆ burden of 20.17 Gg in 1980 (Levin et al., 2010).

4 Sensitivity and validation of SF₆ simulations

4.1 Gravitational separation and mesospheric depletion

To evaluate the relative importance of gravitational separation and mesospheric depletion and their effect on the SF₆ concen-
 trations we ~~have~~ compared the simulations for ~~various~~ the SF₆ tracers and evaluated the relative reduction of the SF₆ content
 350 in the stratosphere due to these processes. As a conservative estimate of the reduction, we evaluated the relative differences
 between the tracers in the latitude belt of 70-85S, since both processes have the most pronounced effect in the southern polar
 vortex, where the downwelling of Brewer-Dobson circulation is the strongest.

Hereafter we quantify the effect of a relative difference between atmospheric contents of two SF₆ tracers “X” and “Y”
 defined as:

$$355 \quad \Delta(\text{“X”}, \text{“Y”}) = 2 \frac{\xi_X - \xi_Y}{\xi_X + \xi_Y} \cdot 100\% \quad (14)$$

The relative differences for the SF₆ tracers in the Southern polar region (70-85S) simulated with two extreme ~~models for~~ K_z
profiles is given in Fig. 4 as a function of time and altitude. Noteworthy, every 5% of the decrease of SF₆ with respect to its
 passive counterpart correspond to about one year of a positive bias in AoA derived from the SF₆ mixing ratios.

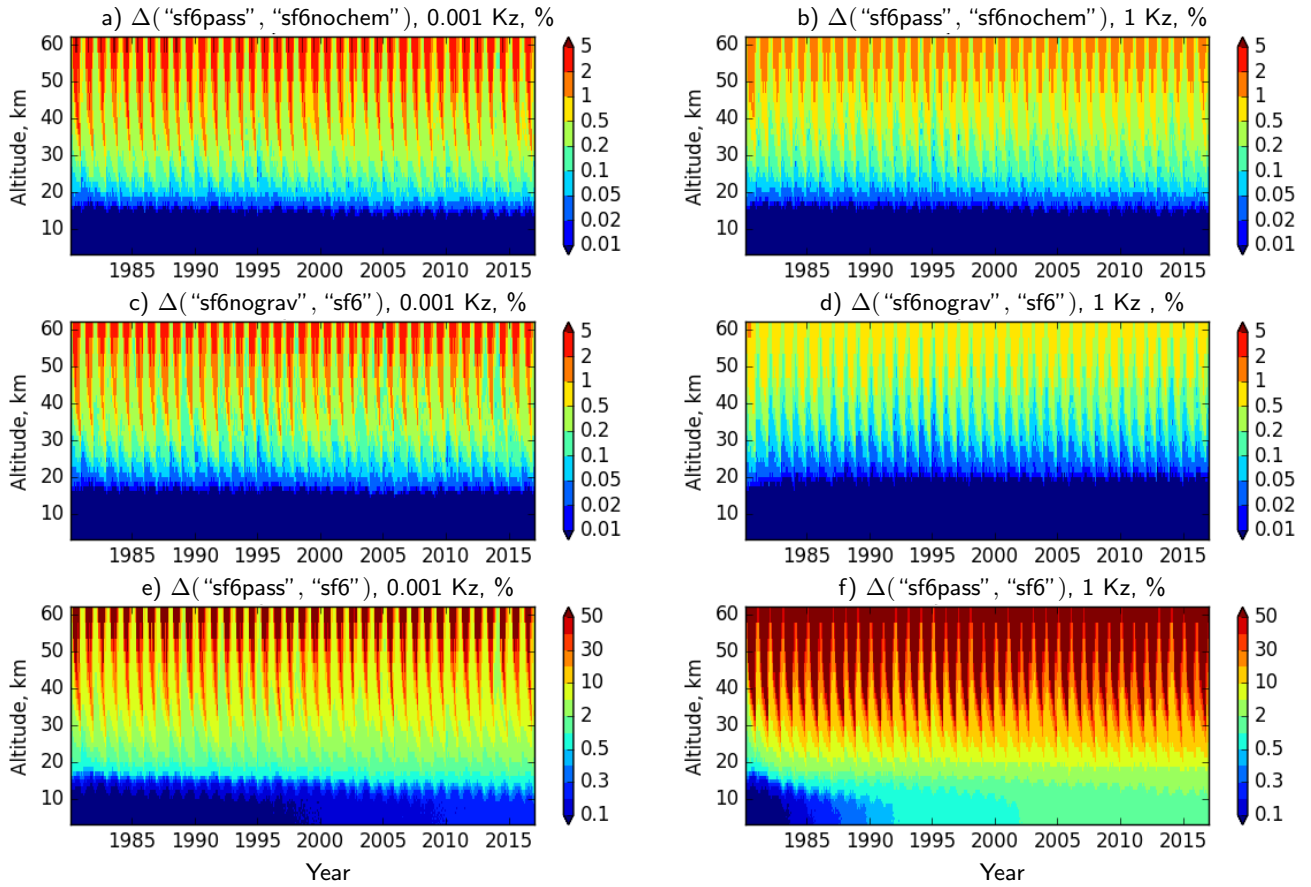


Figure 4. The relative reduction of [the](#) SF_6 content (in %) at 70-85S due to gravitational separation with (a, b) and without (c, d) depletion, and due to combined effect of depletion and separation (e, f) at two extreme K_z cases. Note different color scales for e) and f).

The reduction of [the](#) SF_6 content due to gravitational separation if the mesospheric depletion is disabled is given by the relative difference of “sf6nochem” and “sf6pass” (Fig. 4ab). Expectedly, the effect of gravitational separation is most pronounced for the case of low eddy diffusivity (“0.001 K_z ”), and the reduction of SF_6 in the altitude range of 30–50 km reaches 2 – 5 %. In the case of strong mixing, the effect of separation is about 1 %.

The reduction of [the](#) SF_6 content due to gravitational separation in presence of stratospheric depletion [is](#) given by the relative difference of “sf6nograv” and “sf6” tracers. The effect of the separation for low K_z is very similar between [the](#) depletion and no-depletion [case-cases](#) (Fig. 4c vs. Fig. 4a). Depletion reduces the effect of the gravitational separation for high K_z (Fig. 4b vs Fig. 4d). Regardless depletion, stronger K_z reduces the effect of the gravitational separation, however the latter is still non-negligible if precisions of order of a month for AoA are required.

The combined effect of depletion and gravitational separation is seen in the relative difference of “sf6pass” and “sf6” tracers (Fig. 4e and 4f). For both K_z cases the effect of depletion is stronger than the diffusive separation by more than one order of magnitude. Regardless ~~of~~ the K_z profiles ~~used~~, the reduction exceeds 50 %, which roughly corresponds to 10 years of an offset in the apparent AoA.

In all cases the reduction of the SF₆ content has strong annual cycle associated with the cycle of the downwelling in winter and the upwelling in summer. Besides that the reduction has a noticeable inter-annual variability that poses substantial difficulties ~~on-for~~ applying a consistent correction to the apparent AoA. Contrary to the former two comparisons, strong eddy mixing leads to strong reduction of SF₆ since it intensifies the transport to the depletion layers ~~;~~ and thus enhances the depletion rate.

The simulations for different K_z have been initialized with the same state obtained from a separate spin-up simulation with “0.01 Kz”, which was scaled to match total burden of SF₆ in 1980. Thus a relaxation of the SF₆ vertical distribution during the first few years of the simulations is clearly seen in Fig. 4. For “1 Kz” case (4f) the gradual increase of the difference between SF₆ and its passive version in the troposphere can be seen as well. The rate of this increase is about 0.5% per 39 years of the simulations. This rate should not be confused with the depletion rate of SF₆ in the atmosphere since the difference is a combined effect of depletion and growth of emission rate, despite the latter is exactly the same for both tracers.

The above comparison indicates that ~~the~~ depletion has the stronger effect on the ~~distribution-of-the~~ SF₆ mixing ratio in the upper stratosphere than gravitational separation and molecular diffusion. However, the important role of ~~the~~ molecular diffusion in the model is that it maintains the upward flux towards the mesosphere in the simulations even if the eddy diffusivity ceases.

Further in this paper only the “sf6pass” and “sf6” tracers will be used.

4.2 Evaluation against balloon profiles

The tropospheric concentrations of SF₆ in our simulations have been insensitive to the SF₆ destruction or to the ~~choice-of-the~~ ~~eddy-diffusivity-profile~~ eddy diffusivity profiles in the stratosphere. The difference in the modelled profiles can however be seen above the tropopause. For comparison we took the simulations with prescribed eddy diffusivity in the stratosphere (1Kz, 0.03Kz, and 0.001Kz, see Sec. 3.2), and with dynamic eddy diffusivity “ECMWF Kz”. The simulations were matched with the stratospheric balloon observations (Fig. 5) published by Patra et al. (1997); Engel et al. (2006); Ray et al. (2014, 2017).

Two balloon profiles observed at Hyderabad (17.5N,78.6E) in 1987 and 1994 by Patra et al. (1997) indicate an increase of the SF₆ content during the time between the soundings (Fig. 5a). Both profiles have a clear transition layer from tropopause at ~ 17 km to the undisturbed upper stratosphere above ~ 25 km. The simulated profiles agree quite well ~~to-with~~ the observed profiles, except for the most diffusive case that gave notably smoother profiles and somewhat overstated SF₆ mixing ratios due to too strong upward transport by ~~the~~ diffusion through the tropopause and in the lower stratosphere.

The profile in Fig. 5b has been obtained from Kiruna (68N, 21E) in early spring 2000 during the SAGE III Ozone Loss and Validation Experiment, SOLVE, (Ray et al., 2002) with the Lightweight Airborne Chromatograph (Moore et al., 2003). The profile is affected by the polar vortex and clearly indicates a strong reduction of SF₆ with height with a pronounced local minimum at 32 km. The corresponding SILAM profiles tend to overestimate the SF₆ vmr. The SF₆ profiles for “ECMWF Kz”

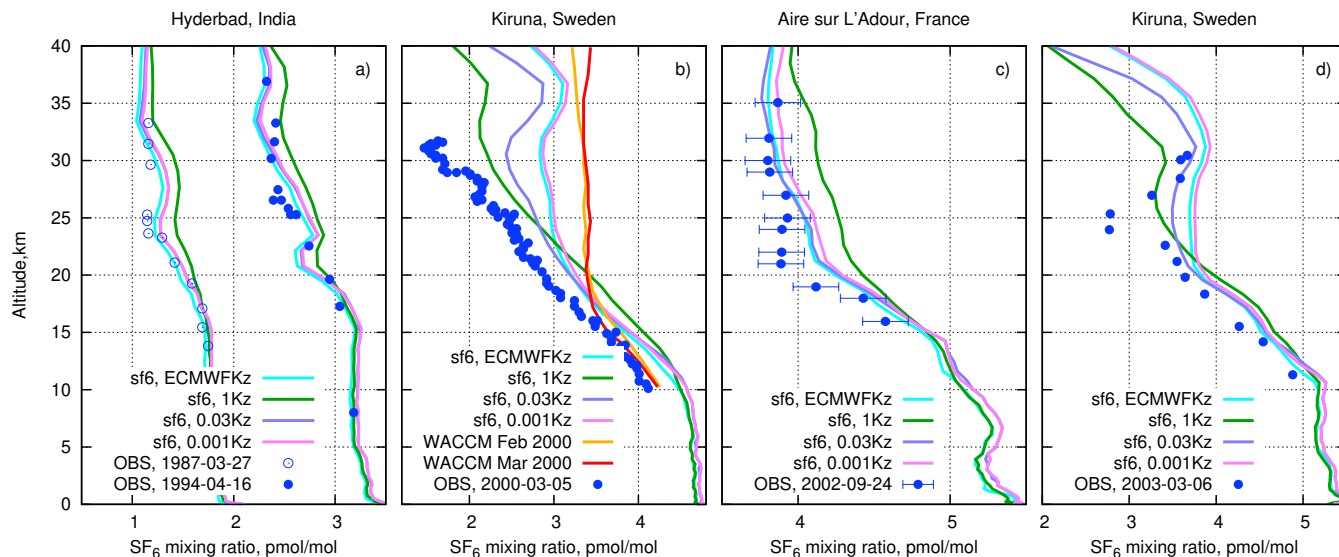


Figure 5. Observed SF_6 balloon profiles and corresponding daily-mean SILAM profiles for the date of observations. The observational data obtained from Patra et al. (1997), Ray et al. (2017), Ray et al. (2014), and Engel et al. (2006) for panels a–d correspondingly. The observation uncertainties are about 2% (1σ) for Hyderabad profiles (a), and smaller than the size of the symbol for Kiruna profiles (b,d). The model profiles from WACCM model are from Ray et al. (2017).

and “0.001Kz” match each other, since vertical mixing is negligible in both cases. The most diffusive profile “1Kz” has the strongest depletion in the upper part, but the largest deviation from the observations below 20 km. The intermediate-diffusion
 405 profile (“0.03Kz”) is almost as close to the observations as the non-diffusive profile. Moreover, the “0.03Kz” profile has a minimum at the same altitude as the observed one, albeit the modelled minimum is substantially less deep.

For comparison, Fig. 5b also contains monthly-mean profiles from the WACCM simulations of Ray et al. (2017)~~along with the observation data.~~ The WACCM profiles match very well the observations below 17 km, but turn nearly constant above, thus under-representing the depletion of SF_6 inside the polar vortex. Monthly-mean SILAM profiles (not shown) were much
 410 closer to the plotted daily profiles than to ~~monthly WACCM ones.~~ Note, that the version of WACCM, used for the the ones of WACCM. However, the WACCM simulations did not include the electron attachment mechanism.

For the mid-latitude profile in Fig. 5c from Aire-sur-l’ Adour, France (43.7N,0.3W), all SILAM profiles except for “1Kz” fall within the observational error bars provided together with the data by Ray et al. (2017). Similar to the Kiruna case in Fig. 5b, the SILAM profiles are ~~much~~ smoother than the observed ones and are unable to reproduce the sharp transition at 20 km.

415 Another profile from within the polar vortex (Fig. 5d) was observed at the same Kiruna site as the one in Fig. 5b, but three years later. The observed profile also has a minimum that is much deeper than in the modelled profiles. Similar to the case in Fig. 5b, the “0.03Kz” profile is the only one that has a pronounced minimum at the same altitude as the observed one. The minimum is a result of the spring breakdown of the polar vortex ~~when a regular down draught~~ down draught ceases, and

atmospheric layers decouple from each other. The reduced depth of the modelled minimum is probably caused by insufficient
420 decoupling of the layers in the driving meteorology.

In all above cases, the “1Kz” profile is clearly far too diffusive: in the non-polar cases ~~and is an outlier that is furthest from the observations, whereas for~~ whereas for the Kiruna cases it overstates the lower part of the profiles and smears out the vertical structure of the profiles further ~~away from above~~ the tropopause. The SF₆ profiles simulated with “ECMWF Kz” and “0.001Kz” match each other in all simulations, since vertical mixing is negligible in both cases. The SF₆ resulting from
425 the “0.03Kz” case ~~appear to be~~ appears to be the most realistic out of the four considered simulations: they are close to the observed ones and have the local minima at ~~right the correct~~ altitudes for both Kiruna profiles.

4.3 Evaluation of SF₆ against MIPAS data

The MIPAS observations provide the richest observational dataset for the stratospheric SF₆ profiles. However, each individual observation has a substantial retrieval noise error, which is noticeably larger than the difference between the observation and
430 any of the SILAM simulations. The largest diversity of the modelled SF₆ profiles was observed in polar regions, therefore below we show the mean profiles for each season in ~~southern and the~~ southern and the northern polar areas. Besides that, we consider statistics of the model performance against MIPAS measurements in the lower and upper stratosphere separately. For simplicity, we do not show the statistics for the “ECMWF Kz” runs, since it is very similar to one for “0.001Kz”.

For the comparison, the daily mean model profiles were collocated to the observed ones in space and time, ~~and then, after~~
435 which an averaging kernel of the corresponding MIPAS profile was applied to the SILAM profile. For the comparison we took only the data points with all of the following criteria met:

- MIPAS visibility flag equals 1
- MIPAS Averaging kernel diagonal elements exceed 0.03
- MIPAS retrieval vertical resolution, i.e. the full-width at the half-maximum of the row of the averaging kernel, is better
440 than 20 km
- MIPAS volume mixing ratio noise error of SF₆ is less than 3 pmol/mol

The mean seasonal profiles of the SF₆ mixing ratio for southern and northern polar regions derived from the MIPAS observations and the SILAM simulations for 2007 are given in Fig. 6. In order to facilitate the comparison of our evaluation with
~~earlier study by the earlier study of~~ Kovács et al. (2017) we have chosen the same year and same layout of the panels as Fig. 3
445 there. The main differences between Kovács et al. (2017) and the current evaluation are:

- We used averages of collocated model profiles (bold lines). The non-collocated seasonal- and area-mean model profiles are given ~~with as~~ thin dashed lines for comparison.
- we use a newer version of MIPAS SF₆ data with considerably larger values (up to 0.6 pptv) in the upper stratosphere, compared to the version that was used by Kovács et al. (2017).

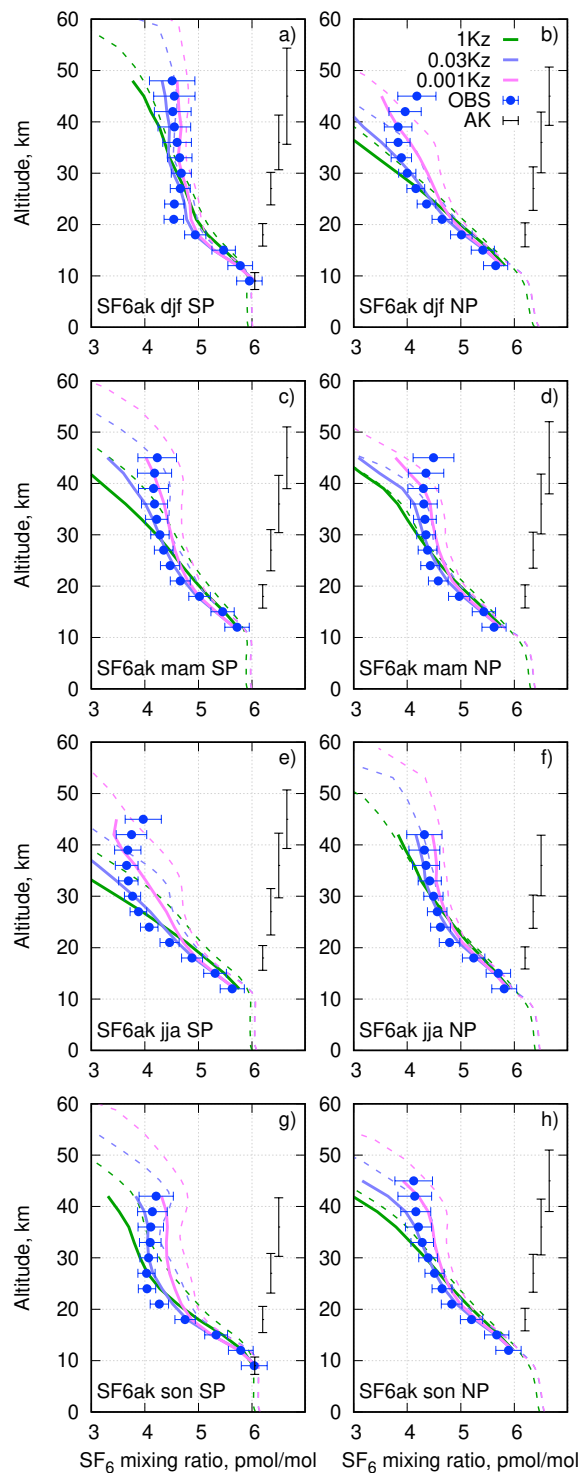


Figure 6. Seasonal mean collocated SILAM SF₆ and MIPAS profiles for 2007, for southern and northern polar regions. Typical ranges covering 75% of the averaging kernel are given with the error bars at the right-hand side of each panel. The horizontal error bars indicate systematic uncertainties of the observations that are fully correlated along profiles and do not cancel out when averaging over a large number of measurements.

- 450 – The horizontal error bars for the observed data indicate the systematic error component that is fully correlated among the profiles and does not cancel out by averaging, or, in other words, the estimate of a possible bias, as analysed by Stiller et al. (2008). These errors are in the order of 4% (below 30 km) up to 10% (at 60 km). The contribution of the retrieval noise error is essentially negligible due to averaging. The error bars shown by Kovács et al. (2017) are noticeably larger, probably indicating that they are for the individual observed values, rather than the uncertainties of the mean.
- 455 – We use 3-km vertical bins for the profiles to make the points in the MIPAS profiles distinguishable
- We also plot the vertical extent of the averaging kernels corresponding to their half-width.

First of all, ~~note there is~~ a substantial difference between the collocated and non-collocated model profiles. The difference is caused by the uneven sampling of the atmosphere by the satellite both in space and in time. In particular, MIPAS, being a polar-orbiting instrument, makes more profiles per unit area closer to the pole than further away. The difference gets somewhat
460 reduced if one uses equal weights for all model grid cells instead of area-weighted averaging, especially for wide latitude belts. The major difference comes probably from the inability of MIPAS to retrieve SF₆ profiles in presence of polar stratospheric clouds that clutter lower layers of the stratosphere and make the sampling of polar regions quite uneven both in time and in vertical. This hypothesis agrees with fact that the difference is most pronounced for the winter pole, especially for the south pole in JJA, and almost invisible at a summer pole.

465 The comparison in Fig. 6 shows that the profiles from the SILAM simulations agree quite well to the observations in the altitude range below 20 – 25 km, with the most diffusive “1Kz” slightly overestimating the SF₆ mixing ratios. In the range above 25 km, the ‘1Kz’ profiles indicate too fast decrease of SF₆ with altitude. The “0.03Kz” profiles give the best results up to ~ 40 km, except for south pole in JJA and North pole in DJF.

An interesting feature of the winter-pole MIPAS profiles is an increase of the SF₆ mixing ratio above 40 km. This increase
470 might ~~have been be~~ caused by issues with retrievals, as the systematic errors of the retrievals increase with altitude. ~~Note, however, that~~ However, non-monotonous profiles can occur due to the mean atmospheric dynamics (see the non-collocated 0.001Kz profile in Fig. 6g).

None of the model setups is capable of reproducing ~~adequately~~ the observations above 40 km. Wintertime poles also pose a problem to the model. The disagreement indicates a deficiency in the model representation of air flows in the upper part of
475 the domain caused by insufficient vertical resolution of ERA-Interim in the upper stratosphere and lower mesosphere, and a lack of pole-to-pole circulation. This discrepancy is in line with the comparisons in Fig. 5 for polar regions. The model tends to overstate the SF₆ content in the lower part of ~~a the~~ polar vortex, and understate it above 40 km.

~~As a more extensive verification of the SILAM simulations we~~ We also computed statistical scores of the simulated SF₆ mixing ratios for each month of the MIPAS mission. The statistics were computed separately for the altitude range of 10 –
480 35 km (Fig. 7) and 30 – 60 km (Fig. 8). As the difference in the statistical scores between the three selected simulations ~~shown in the two figures~~ is quite minor, ~~in addition to the aforementioned selection criteria for MIPAS data, we have selected~~ we used only observations with the retrieval target noise error below 1 pmol mol⁻¹.

The root-mean square error ~~of is turned to be~~ mostly controlled by the bias, and does not allow for clear distinction between the simulated cases. In order to disentangle the effect of bias, we have calculated the standard deviation of the model-measurement difference (STD), absolute bias, and normalised mean bias (NMB):

$$\text{STD(ppt)} = \left\langle (M - \langle M \rangle - O + \langle O \rangle)^2 \right\rangle^{1/2}, \quad (15)$$

$$\text{Bias(ppt)} = \langle M - O \rangle, \quad (16)$$

$$\text{NMB(\%)} = 2 \left\langle \frac{M - O}{M + O} \right\rangle \cdot 100\%, \quad (17)$$

where M and O are modelled and observed values, respectively, and $\langle \cdot \rangle$ denotes averaging over the selected model-observation pairs for the given range of times and altitudes. Along with the STD, we have plotted the RMS error of the observations due to the retrieval noise in the original MIPAS data, labeled as “MIPAS noise” in the top panels of Fig. 7 and Fig. 8.

In the altitude range of 10 – 35 km, the STD of model-measurement difference is uniform in time with minor peaks in August-September (Fig. 7). The level of the noise error constitutes about 85 % of the total model-measurement difference. Application of averaging kernel to the model profiles reduces the STD. The intermediate-diffusivity case “0.03Kz” clearly shows the least STD uniformly over the whole observation period, the same case indicates the least absolute bias.

In the range of 30 – 60 km altitudes (Fig. 8) the level of the retrieval noise is noticeably higher than ~~for the lower stratosphere.~~ ~~Unlike in the lower stratosphere, the,~~ The least biased case is “1Kz”, which however has the largest STD. The STDs of “0.03Kz” and “0.001Kz” are on par, ~~however-but~~ the latter has the strongest biasesbiase. Thus for this altitude range the intermediate-diffusivity case also shows the best performance.

Note the slight increase of the model bias after 2009, which is likely caused by our overstating of the emissions rates since that time (see Sec. 3.5). This increase of the bias does not appear in Fig. 8 due to the delay in the response of the content in the upper layers to the changes in surface emissions.

4.4 Lifetime of SF₆ in the atmosphere

In order to estimate the atmospheric lifetime of SF₆ we turned off the emission of all ~~simulated~~-SF₆ tracers in July 2016 and let the model run until the end of 2018 without emissions (Fig. 9). The decrease of the simulated burden after the emission stop can be used to estimate the removal rate from the atmosphere.

Time series of the total burden of SF₆ in the atmosphere in the simulations are given in Fig. 9. For easier comparison to the observed mixing ratios the burden has been normalised with $1.78 \cdot 10^{20}$ moles – the total amount of air in the atmosphere – to get the mean mixing ratio. The tabulated values for the atmospheric burden of SF₆ from Levin et al. (2010) and Rigby et al. (2010) are given for comparison. Since the removal of SF₆ from the atmosphere is mostly controlled by the transport towards the depletion layer, the vertical exchange is a-the key controlling factor. ~~In all simulated cases, the removal of from the atmosphere is very slow, so the relative difference between the cases is small. Similar rates could have been obtained by averaging the inverse destruction rate mass-weighted over the entire atmosphere.~~

The decrease of the atmospheric SF₆ content after the emission stop, is given at the zoom panel of Fig. 9. As expected, after July 2016 the content of passive SF₆ stays constant, while ~~that undergoes chemical destruction begins~~ the others begin to

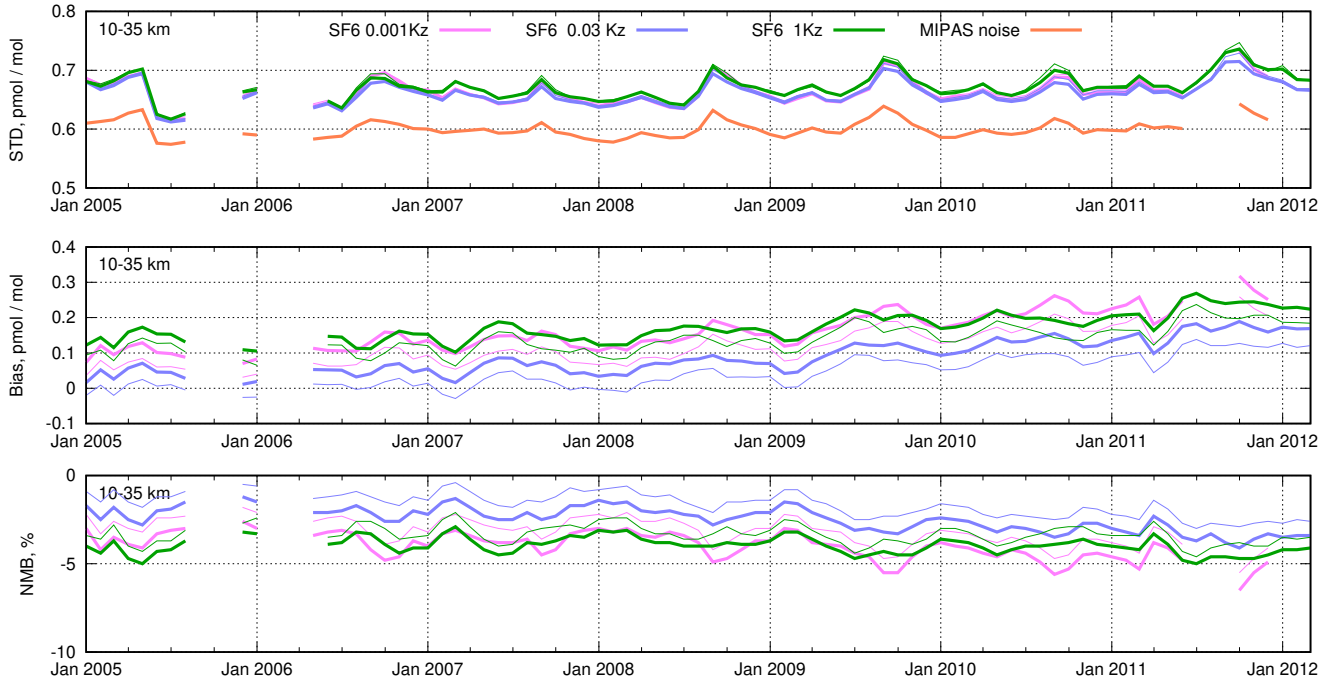


Figure 7. The time series of monthly scores for the SILAM-simulated SILAM SF₆ mixing ratios for the whole period of the MIPAS observations in the altitude range of 10 – 35 km. The statistics are standard deviation of model-measurement difference (STD), absolute bias and normalised mean bias (NMB). The statistics of the model mixing ratios extracted at nominal MIPAS altitudes are given in as thin lines.

Tracer/ Kz scheme	loss rate, 10 ³ mol/year	lifetime, years
passive, any Kz	0	∞
SF ₆ , ECMWF Kz	440	2900
SF ₆ , 0.001 Kz	480	2600
SF ₆ , 0.01 Kz	760	1700
SF ₆ , 0.03 Kz	800	1540
SF ₆ , 0.1 Kz	960	1300
SF ₆ , 1 Kz	2160	590

Table 1. SF₆ destruction rate after stopping the emissions and corresponding lifetimes. Mid-2011 atmospheric burden of $1.27 \cdot 10^9$ moles is used as a reference for the lifetime estimate

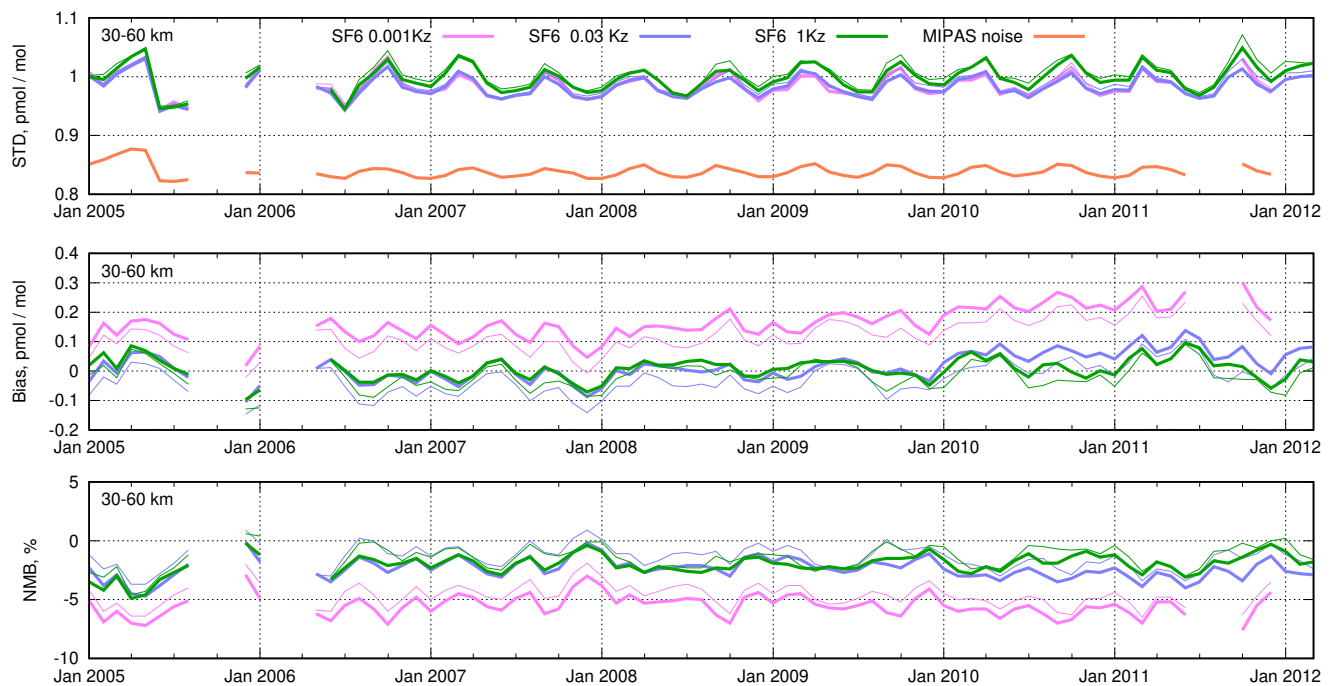


Figure 8. Same as in Fig. 7, but for the MIPAS altitude range of 30 – 60 km.

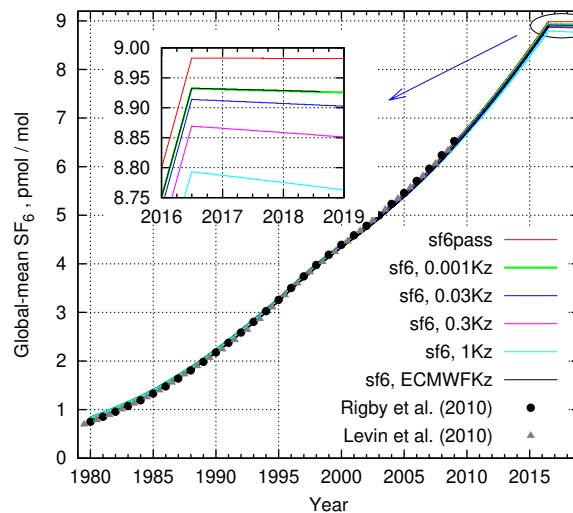


Figure 9. The time series of mean mixing ratio of SF_6 in the atmosphere simulated with emissions stopped in July 2016. The total burdens by Levin et al. (2010) and by Rigby et al. (2010) are shown for comparison.

decrease at a rate that depends on the transport properties ~~of the stratosphere in the simulations, with in the stratosphere with the~~ faster removal for ~~the~~ stronger eddy diffusivity. The removal rate is driven by the SF₆ content in the upper stratosphere, which is not in equilibrium with ~~total-atmospheric~~ ~~the total atmospheric~~ content. A typical delay between ~~the~~ SF₆ mixing ratio in the troposphere, ~~where most of resides,~~ and the upper stratosphere, ~~from where escapes further to the depletion layers,~~ i.e. ~~the~~ AoA
520 in the topmost model layer, is about 5-6 years. ~~Hence, to estimate the lifetimes~~ ~~Therefore, for a reference~~ we used the total amount of atmospheric SF₆ 5 years before the emission stop, i.e. 1.23×10^9 mol, which corresponds to ~~the~~ mean mixing ratio of ~~about~~ 7 pmol/mol. Dividing the destruction rate with the reference amount one gets the range of corresponding simulated SF₆ life times in the atmosphere: 600 to 2900 years. Despite the range of ~~assumed diffusivities is the tested diffusivities of~~ three orders of magnitude, the loss rate ~~of varies varies only~~ within a factor of five (Table 1).

525 The term “life time” implies a linear decay, however, due to emissions the distribution of SF₆ in the atmosphere is far from equilibrium, so the decay is not proportional to the burden. A more accurate way to estimate ~~the~~ life time would be to perform a multi-decade simulation without sources, to get the distribution ~~of~~ into a quasi-equilibrium with the mesospheric sink. In such ~~a~~ quasi-equilibrium ~~a the~~ model of linear decay of SF₆ in the whole atmosphere becomes applicable ~~,~~ and the life time can be estimated as a simple ratio of ~~the~~ burden to the loss rate. The uncertainty in the equilibrium burden corresponding to
530 the modelled loss rates in Table 1 can be estimated as the range of AoA in the upper stratosphere (~ 0.5 years) divided by the growth rate of the burden (0.04 year^{-1}), i.e about 2%. ~~The major~~ ~~A larger~~ uncertainty comes from the over-simplistic parametrization of the loss in the model, which is more difficult to quantify.

The best-performing ~~in terms of simulation~~ ~~simulation “0.03Kz”~~ resulted in 1540 years lifetime. Given the uncertainties above, it meets the ranges suggested by earlier studies. It is in a good agreement with the range of 800 – 3200 years from
535 ~~earlier the~~ model studies (Ravishankara et al., 1993; Morris et al., 1995), and is close to the upper bound of the 580–1400 years range recently obtained by (Ray et al., 2017) from the balloon profile given in Fig. 5b.

Our estimate is also slightly above the range given by Kovács et al. (2017), who obtained 1120 – 1475 years. ~~Note, however,~~ ~~that~~ ~~However,~~ in the simulations of Kovács et al. (2017) the mixing ratios of SF₆ in the stratosphere and ~~the~~ lower mesosphere were noticeably higher than ~~retrieved from MIPAS~~ ~~those retrieved by MIPAS and practically flat in the range of 30 – 50 km.~~
540 ~~Such modelled profiles likely indicate over-stated vertical exchange in the model~~, which is likely to cause overstating the simulated ~~depletion rates~~ ~~loss in the atmosphere~~, and lead to corresponding low bias of the ~~lifetime from those simulations~~ ~~estimated lifetime~~.

5 Simulations of AoA

5.1 Eddy diffusivity and simulated AoA

545 The effect of the vertical eddy diffusivity on AoA in the stratosphere was evaluated with the same set of three prescribed ~~K_z~~ ~~profiles~~ and one dynamic K_z ~~profile~~ ~~profiles~~, as for SF₆ simulations. An example of annual-mean distributions of AoA ~~for the same year~~ is given in Fig. 10. The Hunten (1975) K_z profile (Fig. 10a) gives AoA in the stratosphere of about 3.5 years. It is much shorter than ~~available estimates of~~ ~~the estimates of the~~ stratospheric AoA (e.g. Waugh, 2009; Engel et al., 2009) from the

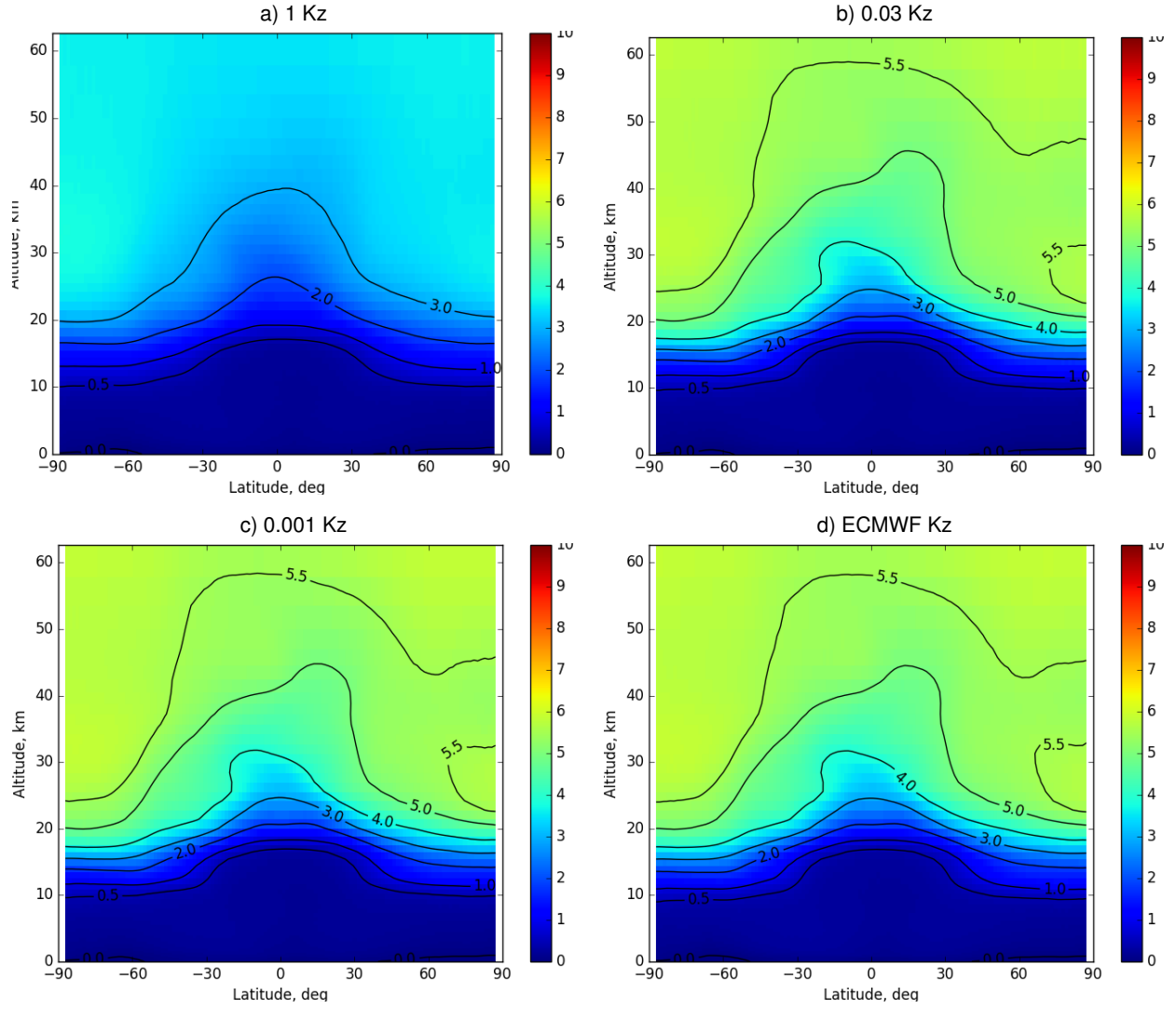


Figure 10. The zonal-mean spatial distribution of the ideal-age AoA for 2011 calculated for different eddy-diffusivity profiles.

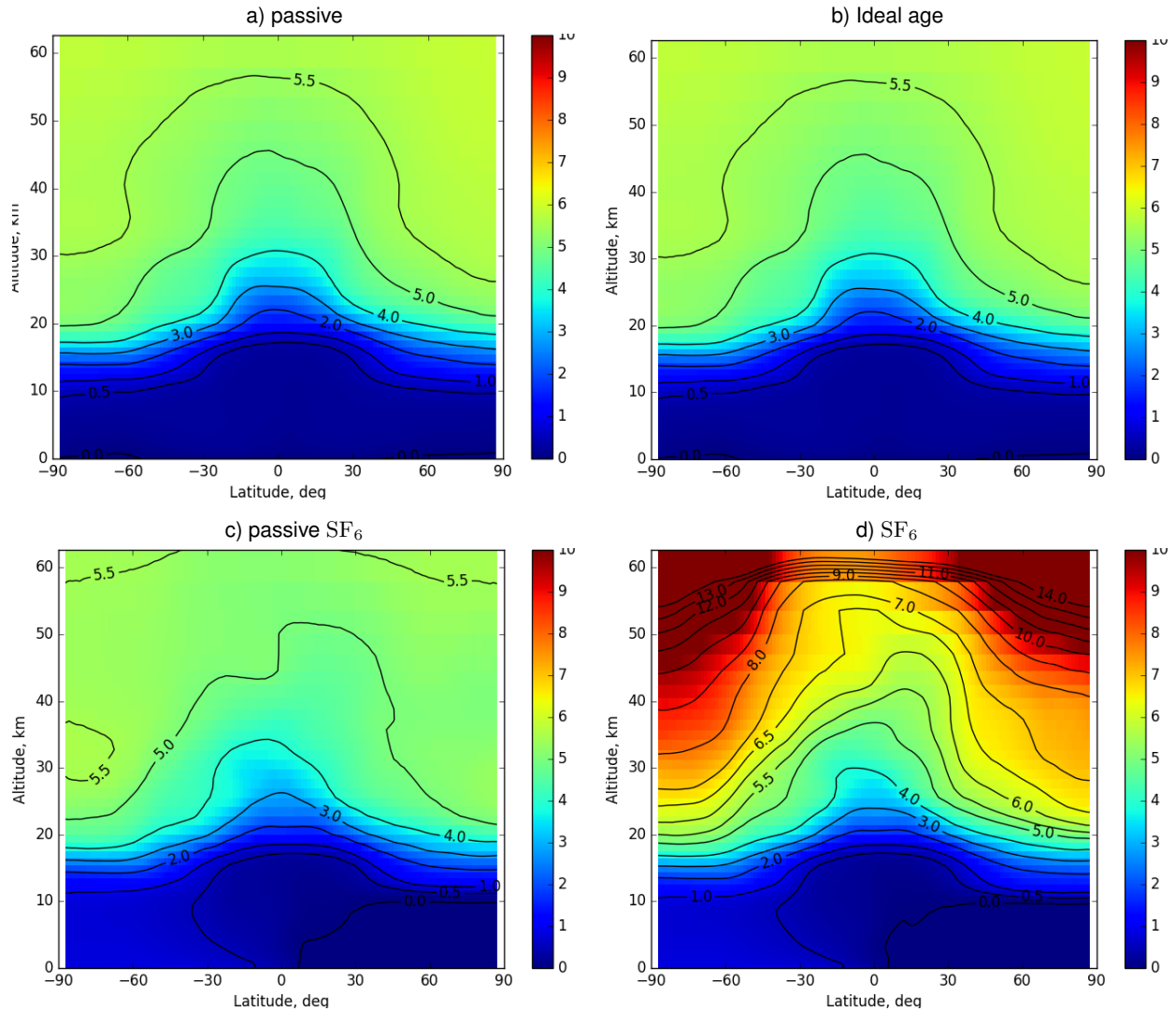


Figure 11. Zonal-mean distributions of atmospheric AoA simulated with “passive”, ideal-age, and two SF₆ tracers, average for 2012.

observations of various tracers. Three other profiles of K_z result in almost identical average ~~distribution~~ distributions of AoA
550 with typical stratospheric AoA of 5.5 years, which agrees quite well with the experimental estimates. In these cases AoA is
controlled by the transport with ~~explicitly-resolved-mean~~ winds. Since “0.03Kz” profiles result in most realistic distribution of
SF₆ in our simulations, in the current section we will use simulated distributions of tracers with ~~“0.03Kz” eddy-diffusivity~~ this
parameterization.

5.2 AoA and apparent SF₆ AoA

555 The AoA for all tracers (except for the “ideal age”) was calculated as a simple time lag between ~~a mixing ratio in a given the~~
mixing ratio at each point of the domain and the mean near-surface mixing ratio. As it has been pointed out by (Vaugh and
Hall, 2002), this lag equals to AoA only in case of a fully passive tracer with linearly growing (or decreasing) near-surface
mixing ratio. Corrections have been applied to the AoA derived from SF₆ in many studies ~~studies~~ (Volk et al., 1997; Stiller
et al., 2008, 2012; Engel et al., 2009) to account for non-linear growth of the near-surface SF₆ mixing ratio and for ~~mesospheric~~
560 ~~sink of its mesospheric sink~~. The corrections rely heavily on various assumptions that can hardly be rigorously verified ~~for the~~
~~atmospheric circulation~~. Therefore in this study ~~for the sake of simplicity~~ we do not apply any corrections to the AoA derived
from ~~the~~ time lags of tracers. The corrections and assumptions behind them are discussed in Sec. 6.

The constant-rate emission of the “passive” tracer ~~in our simulations resulted in nearly~~ resulted in almost linear growth of ~~the~~
its near-surface mixing ratio ~~of the tracer after a decade of~~ after the spin-up. The latter makes the age derived from the “passive”
565 tracer equivalent to the age derived from the ideal-age tracer. The resulting distributions ~~of “passive” and ideal-age AoA~~ are
indeed very close to each other (Fig. 11 a and b). The agreement confirms the self-consistency of the transport procedure since
the tracers have opposite sensitivity to the advection errors: higher mixing ratios correspond to younger air for the accumulating
tracers, while for the ideal-age tracer higher mixing ratios correspond to older air. The remaining differences ~~of~~ are caused by
spatial inhomogeneities of near-surface mixing ratio of “passive” due to variations in the near-surface air density.

570 The distribution of the AoA derived from “sf6pass” (Fig. 11c) is qualitatively similar to the ideal-age one, however one can
see substantial differences. The negative AoA in the northern troposphere for the “sf6pass” tracer is caused by the predominant
location of the sources in the northern hemisphere, so the concentrations there exceed the global-mean levels. The growing rate
of the SF₆ emissions leads to the ~~greater-than-linear~~ faster-than-linear increase of near-surface mixing ratios, which leads to
an old bias up to 3-5 months of the “sf6pass” AoA. This old bias has been one of the drawbacks of the SF₆ AoA pointed by
575 Garcia et al. (2011).

The ages shown in Fig. 11a – c agree well with the ages derived from the *in-situ* observations of SF₆ and CO₂ at the
25 km altitude by Vaugh and Hall (2002). They also agree quite well with the earlier simulations with five climate models that
give annual mean ages in the upper stratosphere between 4.5 and 5.5 years (Butchart et al., 2010), and with the Lagrangian
simulations of (Diallo et al., 2012) driven by the same ERA-Interim meteorological fields as used for the present study. A
580 substantial disagreement, however, exists with the ages derived from the MIPAS satellite observations (Stiller et al., 2012;
Haenel et al., 2015), who calculated ages exceeding 10 years in the polar areas and in the upper stratosphere. The reason
for the disagreement follows from the above analysis: SF₆ can neither be considered as a passive tracer nor does its mixing
ratio in the troposphere ~~grows~~ grow linearly with time. Denoting the AoA derived from the SF₆ profiles as “apparent AoA”
(Vaugh and Hall, 2002), we calculated it from the SILAM-predicted SF₆ profiles, which, as shown above, agree well with
585 AoA derived from MIPAS. The resulting model-based apparent AoA (Fig. 11d) is indeed much older than the “ideal-age”
AoA. The distribution of the apparent SF₆ AoA agrees to the AoA retrived from MIPAS SF₆ profiles by Haenel et al. (2015):
well over 5 years AoA around equator with well over 10 years AoA in the polar regions.

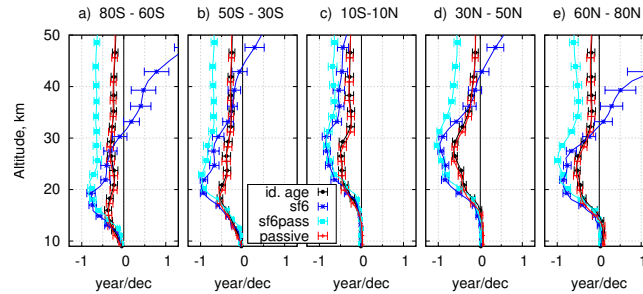


Figure 12. Vertical profiles of the simulated age of air linear trends over 11 years 2002-2012 for example latitude belts

The effect of the apparent over-aging in the stratosphere due to the subsidence of the mesospheric air was estimated by Stiller et al. (2012) to be a fraction of a year in the upper stratosphere. Earlier experimental balloon studies (Strunk et al., 2000) indicated up to 3.5-year difference between CO₂ and SF₆ ages. In our simulations, the over-aging due to the SF₆ depletion and other factors discussed in the previous sections is much stronger and affects the whole stratosphere.

5.3 Trends in apparent AoA

Changes in the AoA have been used in many studies as an indicator of changes in the atmospheric circulation. In order to evaluate the effect of the way AoA is evaluated on trends in AoA the AoA is computed on its trend we have calculated trends in-of the apparent AoA at different altitudes and latitudes for 11 years 2002-2012. This period roughly covers the MIPAS mission time and allows for comparison with trends reported by Haenel et al. (2015).

The zonal-mean vertical profiles of the AoA trends during 2002-2012 are shown in Fig. 12 for five latitudinal belts. The presented variable is a slope of the linear fit of the deseasonalized monthly-mean time series for each tracer, averaged over the corresponding latitudinal belt and the model layer. The fit was made with the ordinary least-squares method for each tracer. The error-bars show 95-% confidence intervals calculated as if a model of linear trend with uncorrelated Gaussian noise was applicable to the time series.

The trends of the apparent AoA for the non-passive SF₆ species have a clear increase with height in the upper part of the profiles. The increase is the largest at high latitudes. Such a behaviour of trends Such behaviour agrees well with the AoA trends of Haenel et al. (2015, Fig. 7) obtained from the MIPAS observations. The over-aging due to the mesospheric depletion of SF₆ has been discussed and estimated by Haenel et al. (2015); Kovács et al. (2017). However, Fig. 12 shows that the mesospheric depletion of SF₆ also affects its trend: the over-aging increases with time. The reason is that depletion is proportional to the SF₆ load, which grows with time. This effect has been pointed out earlier by Stiller et al. (2012) and evaluated earlier for N₂O by Schoeberl et al. (2000). For SF₆ the effect of its loss on the AoA was evaluated by Stiller et al. (2012), who concluded that “in-mixing of mesospheric SF₆-depleted air plays a minor role for the assessment of AoA trends”, at least within the framework of their approach (2002 – 2010, up to 35 km altitude).

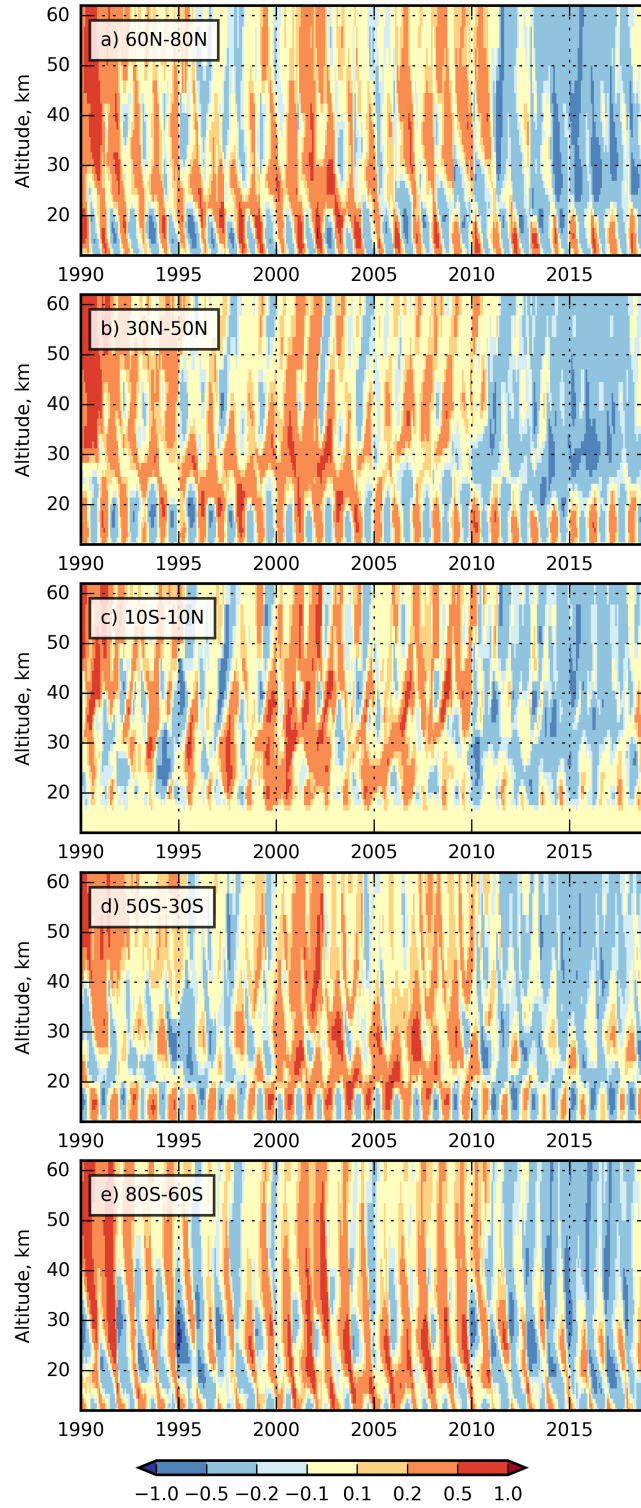


Figure 13. Anomaly of the ideal-age AoA (years) for the period of 1990-2018 with respect ~~of to~~ the mean AoA ~~over the same period~~

The apparent AoA derived with the passive SF₆ tracer “sf6pass” indicates a negative trend of about 0.5 years/decade. The trend is caused by the temporal variation of SF₆ emissions. In order to get unbiased AoA estimate from a~~the~~ passive tracer, one needs the mixing ratio at the surface increasing linearly with time. A steady growth of emission rate ~~at the surface leads to~~ leads to the faster-than-linear increase of the near-surface mixing ratio and, thus, low-bias of ~~AoA since younger (i.e. more rich~~ the AoA. According to the inventory (Levin et al., 2010) used in this study, the SF₆ emission rate was growing in 1997–2000 about twice slower than after 2005. Consequently, the negative bias of the apparent AoA has increased resulting in ~~negative trend if the negative trend of the~~ negative trend of the AoA in the stratosphere.

The AoA trends derived from the “ideal age” and “passive” tracers agree through the whole range of altitudes and latitudes indicating internal consistency of our simulations. The main common feature of the profiles is the negative tendency of about –0.5 year/decade in the altitude range of 15–30 km with a profile that varies across altitudes. Similar-magnitude trends for the same period were reported by Plöger et al. (2015), who used the same ERA-Interim to simulate AoA. The major difference between the obtained trends is that we have consistently negative trends for both hemispheres, whereas Plöger et al. (2015) indicate a positive trend of a fraction of year per decade in the altitude range of 20 – 30 km in the Northern hemisphere, and a similar-magnitude negative trend in the Southern hemisphere. The reason for the discrepancy ~~despite the~~ despite the same input dataset deserves further investigation. Possible reason for the discrepancy likely is that Plöger et al. (2015) used diabatic heating rates as vertical velocity, and it is known that the diabatic and kinematic vertical transport is inconsistent in the reanalysis (Abalos et al., 2015).

The trends might be a feature of the non-uniformity of the ERA-Interim dataset, which was produced with assimilation of an inhomogeneous set of the observations. During 2002–2012, the amount of the assimilated data ~~on of~~ on of the upper-air temperatures was by an order of magnitude higher than before 2000 and two orders of magnitude higher than after 2010 (Dee et al., 2011). It had a clear impact on the patterns of the analysis increments in ERA-Interim and, consequently, on the predicted stratospheric circulation. Due to such inhomogeneities, the quality of trends derived from reanalysis data needs to be verified for each geophysical quantity (Dee et al., 2011). Deducing reliable trends for atmospheric temperature, a quantity that is measurable and extensively assimilated, took a major effort (Simmons et al., 2014). The fact that the AoA is not a directly observable quantity makes the verification of the AoA trends ~~in ERA-Interim hardly possible~~ difficult.

To get more insight on the nature of the simulated long-term AoA variability at different altitudes and latitudes we have plotted the time series of the monthly zonal-mean ideal-age AoA for the same latitude belts as in Fig. 12 over 1990–2018 (Fig. 13). To make the temporal variations more visible, the mean AoA profile for each latitude averaged over the same period was subtracted ~~form from~~ from the profiles. One can see a clear seasonal variation of the AoA outside ~~of~~ of the equatorial zone. The variation has opposite ~~phase between phases in~~ phase between phases in the upper and the lower stratosphere. In the altitude range of 20 – 30 km, where the trends are most pronounced, the temporal variation of the AoA has a ramp structure with more-or-less steady intervals and relatively quick changes. Such structure is similar to the one shown for the ~~ERA=Interim~~ ERA-Interim analysis increments (Dee et al., 2011) and is likely to be caused by temporal inhomogeneities in the assimilated dataset. Therefore we do not draw any conclusion here on the actual trends of AoA but highlight that trends of the apparent AoA are strongly influenced by the selected time interval ~~;~~ ; and by the method of the trends calculation.

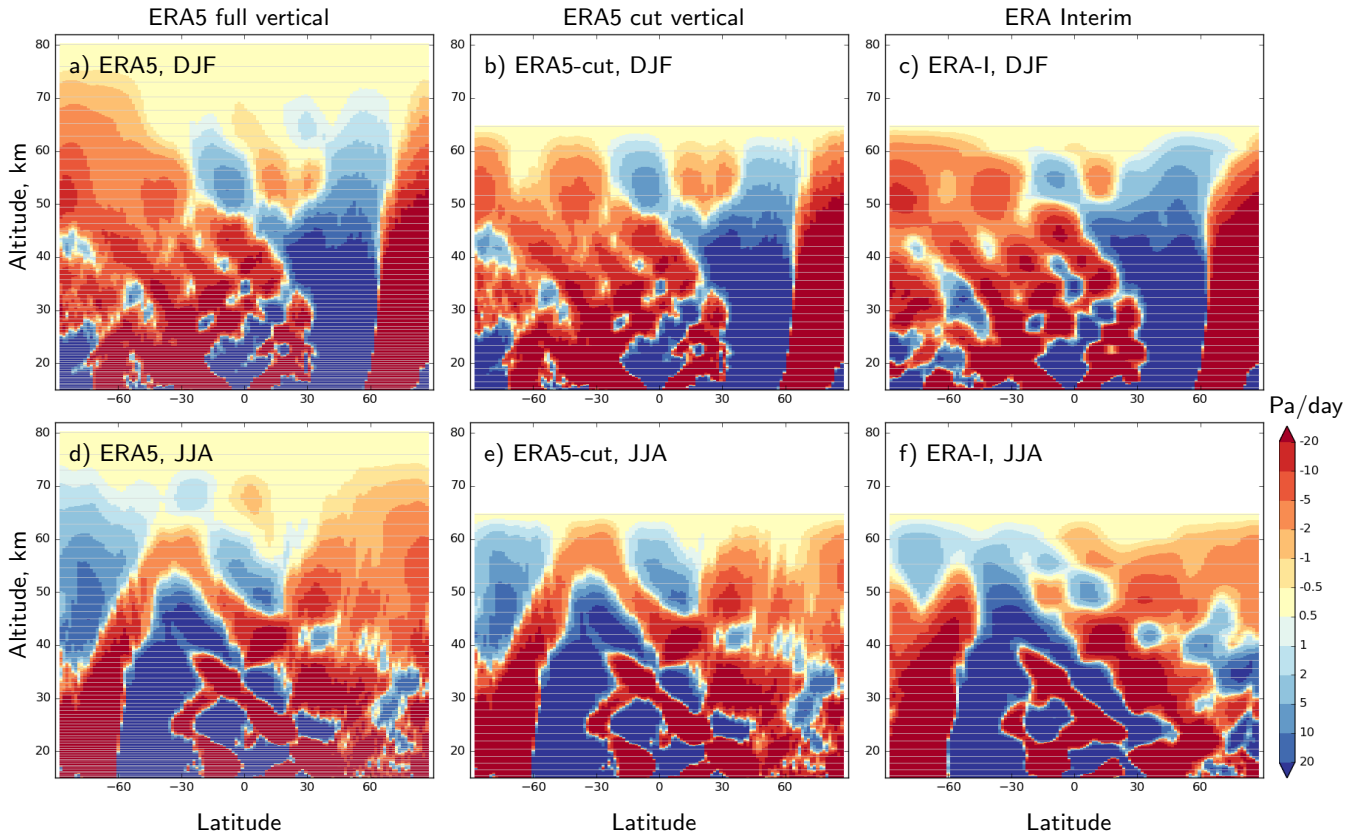


Figure 14. The seasonal and zonal-mean vertical air-mass fluxes diagnosed by SILAM from ERA5 and ERA-Interim fields for 2017 solstice seasons, expressed in terms of vertical velocity ω . Updrafts are red. The vertical-layers boundaries are shown with grey lines.

6 Discussion

The present study has several limitations that deserve specific attention. Forced zero air flux through the domain top at 0.1 hPa caused distortion of the mean transport within the domain, and left diffusive transport as the only means for the upper-boundary fluxes of SF_6 . Moreover, we used prescribed profiles of the eddy diffusivity within the domain, which also affects the results of the simulations. In this section we evaluate the role of these distortions.

6.1 Distortions of air-flows

The transport procedure used in this study is done with a “hardtop” diagnostics forcing zero mass-fluxes at the domain top and forced air-mass conservation everywhere within the domain. Since the upper boundary of the domain is at 0.1 hPa, the divergence of the air flow above that level in the meteorological driver data used to drive the model is compensated by adjusting the divergences within the domain. To evaluate the effect of this adjustment on the mean circulations we used the new ERA-5 data-set, which has the topmost level at 1×10^{-3} hPa, as a reference. The diagnostic procedure was applied to ERA5

for two sets of vertical layers: the 61 ERA-Interim layers, same as used in the SF₆ simulations (hereafter ERA5-cut), and a refined vertical matching the 137 native ERA5 vertical layers (hereafter ERA5). The resulting vertical winds were compared to the ones used in the SF₆ simulations: 61 ~~ERA-Interim~~ layers diagnosed from ERA-Interim. The seasonal and zonal-mean
660 vertical air-mass fluxes, expressed in units of Pa/day for the three cases and two solstice seasons of 2017 are shown in Fig. 14 together with the corresponding layer boundaries.

The wind patterns in ERA5 (Fig. 14abde) have finer features than in ERA-Interim due to the higher horizontal resolution ~~of the former~~. The difference between the ERA5 and ERA5-cut vertical winds is the strongest at the ~~cut-domain-cut-domain~~ top (0.1 hPa, 65 km), where the zero vertical air-mass flux is forced. For both seasons the disturbances introduced by the cut
665 vertical ~~to the ERA5 dataset below 55 km~~ are minor, except for the summertime poles (South pole in Fig. 14ab, and North pole in Fig. 14de), where a noticeable disturbance is visible down to 35-40 km altitude. Such systematic disturbances influence the performance of the AoA and the SF₆ simulations in the polar stratosphere, and they are a probable reason for the failure of the model to reproduce the SF₆ profiles there (see Fig. 6).

The comparison of the ~~same-vertical~~ mass-fluxes for the same vertical levels (panels b vs. c, or e vs. f in Fig. 14) shows
670 that the difference between ERA-Interim and ERA5 is noticeably larger than between cut- and full vertical of ERA5. Thus we conclude that the distortions introduced by our diagnostic procedure are within the uncertainty of the input meteorological data.

6.2 Top-boundary mass fluxes and eddy diffusion profiles

The used modelling approach replaces the vertical transport through the domain top with the diffusive fluxes for the depleting
675 SF₆ and a hard lid for other species. ~~One can hope that the approach does not introduce any~~ This approach is unlikely to introduce major disturbances into the AoA fields ~~since~~ since the AoA is quite uniform close to the domain top. The uncertainty introduced with this approach into the SF₆ fields is not straightforward to evaluate due to a major uncertainty in the vertical diffusivity profiles.

As mentioned in Sec. 3.2, the eddy diffusivity profiles of the C-IFS model ~~form from~~ the ERA5 reanalysis (Fig. 2) are clearly
680 unrealistic within and above the stratosphere. They do not exhibit any growth of the eddy diffusivity in the mesosphere ~~due to breaking gravity waves~~ either. According to Lindzen (1981) the mean diffusivity due to the breaking gravity waves has an order of magnitude of $1 \times 10^2 \text{ m}^2/\text{s}$, whereas the eddy diffusion in ERA5 for that region is below the molecular diffusivity (Fig. 2). On the other hand, if we assume that the mesospheric turbulence ~~due to the breaking gravity waves~~ results in a diffusivity profile as predicted by Lindzen (1981) (Fig. 2) ~~then such a~~ then such turbulence provides quite rapid exchange of SF₆ towards the
685 depletion layers making the advective vertical transport above ~ 50 km negligible. The profiles of (Lindzen, 1981), however, do not allow for a simple extrapolation ~~to~~ below 50 km ~~and therefore the vertical profiles by Massie and Hunten (1981) (“1Kz”) were involved as the ones that are simple to implement and smooth enough to be easily approximated and extrapolated. The sealing of the “1Kz” profile allowed for the sensitivity tests.~~

The normalized diffusive SF₆ mass-fluxes above the domain top for the scaled profiles of the eddy diffusivity (Fig. 3) allow
690 for evaluation of the validity of the assumption of neglected regular vertical transport above the domain top. The equivalent

vertical air-mass flux due to diffusion at the level of 0.1 hPa (domain top) is 6×10^{-6} , 9×10^{-7} , and 2.5×10^{-7} kg/m²/s for “1Kz”, “0.03Kz”, and “0.001Kz” ~~correspondingly~~respectively. These mass fluxes, divided by g give the vertical velocities of -5 , -0.8 , and -0.4 Pa/day. Comparing these values to those shown in Fig. 2-14 for the level of 65 km, one can see that the diffusive limit is valid for the “1Kz” profile except for the very vicinities of the poles. For lower values of the eddy diffusivity
695 the regular circulation becomes comparable with the diffusion or even exceed it.

Although the “0.03Kz” profiles gave ~~somewhat~~ better agreement with the observations of SF₆, this does not indicate that “0.03Kz” profiles are more realistic. ~~As suggested by one of the anonymous reviewers, this~~This profile is likely to over-mix the lower stratosphere and under-mix the upper stratosphere and the mesosphere. Thus the vertical structure of the eddy diffusivity remains a major source of uncertainty in the modelling approach. Using more realistic vertical diffusion profiles and high-top
700 ERA5 reanalysis is planned for the future studies.

6.3 Notes on the observed SF₆-age

There are three main factors ~~that are responsible for~~responsible for the SF₆ age being different ~~form~~from the “ideal age”: the non-linear growth of tropospheric burden, the gravitational separation, and the mesospheric sink. Here we consider the effects of these factors and corrections to the SF₆ observations that can be applied to compensate for the effect of these factors on the
705 resulting AoA.

The correction for the non-linear growth rate introduced by Volk et al. (1997) and used in many subsequent studies is based on a simple analytical model of 1D diffusion with constant diffusivity and exponential distribution of air density. The model was suggested by (Hall and Plumb, 1994) as an illustration for the concept of the age spectrum. The model spectrum has two parameters: the mean age Γ and the width parameter Δ . In order to use the spectrum for the correction one has to involve an
710 additional constraint connecting these parameters. Based on a 3D simulation with a general circulation model Hall and Plumb (1994) suggested that a constant ratio $\Delta^2/\Gamma = 0.7$ year can be used throughout the stratosphere. Note that this dimensional parameter, while having proper units originally, appears without units in several subsequent papers (Engel et al., 2002; Stiller et al., 2012). Volk et al. (1997) used the value $\Delta^2/\Gamma = (1.25 \pm 0.50)$ year for the lower stratosphere based on the results of a more advanced GCM than the one used by (Hall and Plumb, 1994). With this approach Volk et al. (1997) obtained the
715 difference between the mean age and the lag time (apparent SF₆ age). The difference becomes significant for the air older than 3-4 years and approaches (0.50 ± 0.25) years for the oldest (6 years) air~~measured~~, which agrees quite well with the difference between the ideal age and the passive SF₆ in our simulations ~~shown in~~ (Fig. 11bc). The correction for this difference derived from the 1D has been used to reduce the systematic biases from the SF₆-based AoA, though “the global stratosphere is poorly represented by a 1-D model” (Waugh and Hall, 2002). The uncertainty of the correction of up to ± 0.5 years is systematic, ~~and~~
720 is not guaranteed to be uniform in space or in time, and is likely to affect the trend estimates.

As shown in Sec. 4.1, the biases introduced to the SF₆-based AoA by gravitational separation reach a fraction of a year in the upper stratosphere. One could in principle elaborate a correction for gravitational separation; however, the correction would be well within the uncertainty of the correction for the non-linear growth rate, and thus probably not worth considering.

The mesospheric sink ~~clearly~~ has the largest impact on the SF₆-derived AoA. The effect of the mesospheric sink is clearly visible above 15-20 km at all latitudes (Fig. 11) ~~;~~ and leads to a ~~factor-of-times-apparent-strong~~ over-aging in the upper layers, especially in the polar areas. The effect of the sink alone can explain the discrepancy between the AoA derived from the MIPAS observations (Haenel et al., 2015) and the AoA from the modelling studies (e.g. Diallo et al., 2012; Brinkop and Jöckel, 2019). Compensating for such over-aging is hardly possible without detailed modelling of the physical processes including depletion, diffusion and mean transport, ~~which cause the over-aging~~. Since the AoA is derived as a *difference* of the SF₆ mixing ratios, whereas depletion introduces *multiplicative* change to the SF₆ abundance, the effect of the sink on apparent SF₆ AoA is unsteady in time. ~~This effect is clearly seen in~~ (Fig. 12).

Once one has a model that is capable of reproducing the processes behind the SF₆ depletion, it ~~makes a full sense is natural~~ to validate such a model directly against the available SF₆ observations, rather than deriving ~~AoA from the AoA from the~~ SF₆ observations and comparing it against the modelled one. In any case the AoA derived from the SF₆ tracer observations with all the ~~needed corrections applied~~ corrections cannot be considered as a purely observed one.

7 Conclusions

Eulerian simulations of the tropospheric and stratospheric transport of several tracers were performed with the SILAM model driven by the ERA-Interim reanalysis for 1980-2018. The simulations included ~~several~~ species representing SF₆ under different assumptions, a passive tracer emitted uniformly at the surface, and an “ideal age” tracer directly comparable ~~are comparable~~ to other state-of-the-art CTM simulations of the AoA. To our best knowledge this is the first systematic evaluation of AoA derived from several different tracers within the same ~~simulation over several decades~~ multi-decadal simulation, combined with the extensive evaluation against MIPAS and balloon SF₆ observations.

Due to the limited vertical coverage and resolution of ERA-Interim in the upper stratosphere, the SILAM simulation domain had a lid at 0.1 hPa, which is below the altitude of the SF₆ destruction. In order to perform realistic simulations of SF₆ in our setup, the eddy diffusion in the upper stratosphere and lower mesosphere had to be parameterised, along with the mesospheric sink of SF₆.

A set of simulations with different parameterisations for the vertical eddy diffusion showed that published profiles derived with no account for advection (see e.g. Massie and Hunten, 1981, and references therein) overestimate the eddy diffusivity. On other hand, the eddy-diffusivity profiles for scalars calculated from the ERA-Interim fields according to the IFS procedures ECMWF (2015)), or readily available from the ERA5 reanalysis, appear to be of no relevance for the upper stratosphere, since they fall below the molecular diffusivity. Evaluation of our simulations against satellite and balloon observations indicated that the best agreement between the simulated and observed SF₆ mixing ratios within the model domain is achieved for the tabulated eddy-diffusivity profile of Hunten (1975) scaled down with a factor of 30. ~~Note, however, that~~ However, this conclusion is likely to be a feature the specific model setup. Thus, the question of the importance and magnitude of the eddy diffusivity in the upper stratosphere and lower mesosphere remains open ~~;~~ and the SF₆ observations is a good means ~~to~~ validate more sophisticated parametrizations of it of its evaluation.

The mesospheric sink of SF₆ has a major impact on the mixing ratios above 20 km. The depletion impact is especially strong in the wintertime polar areas due to the downdraft within a polar vortex. A set of sensitivity tests showed that molecular diffusion and gravitational separation of SF₆ are responsible for up to a few percent of further reduction in SF₆ mixing ratios in the upper stratosphere.

A good agreement of the simulated SF₆ distribution ~~to~~ with the MIPAS observations up to the altitudes of 30-35 km and ~~to~~ with the available balloon profiles was shown. The standard deviation between ~~MIPAS and the~~ MIPAS and the modelled SF₆ mixing ratios is ~~up to~~ mainly 80 % controlled by the noise error of the satellite retrievals, i.e. the standard deviation between model and MIPAS is about as large as the error on the satellite data. The results of the comparison also underline the importance of accurate collocation of ~~model and the~~ modeled and the observed data in terms of space, time and vertical averaging of the observed data.

The lifetime of SF₆ in the atmosphere estimated from the best-performing setup is about 1500 years, which is at the high side of the range of other recent estimates. Our estimate is likely to be biased high due to underrepresented vertical exchange to at the domain top due to missing advective transport and the missing effect of braking gravity waves.

Our simulations were able to reproduce both AoA obtained in other model studies, and the apparent SF₆ AoA derived from the MIPAS observations. This highlights the role of fast mesospheric destruction of SF₆ due to the electron attachment mechanism. Having all tracers within the same simulations we were able to trace the differences in the estimated AoA to the peculiarities of each tracer. A good agreement of the passive-tracer and “ideal-age” AoA indicates a consistency of the simulations, since these two methods have opposite sign of sensitivity to errors of the transport scheme.

The mesospheric sink has severe implications on the AoA derived ~~form~~ from the SF₆. The apparent over-aging introduced by the sink is large and variable in space and ~~seasentime~~ time. Moreover, the over-aging due to the sink increases as the atmospheric burden of SF₆ grows. All this makes SF₆ unsuitable to infer AoA above ~ 20 km. For a fully-passive SF₆ tracer, the variable rate of emissions causes deviations from the “ideal age”, and these deviations can be compensated to some extent. However, ~~the correcting for~~ correcting the deviations due to the mesospheric sink of SF₆ is hardly possible ~~without detailed modeling~~. These deviations appear as long-term trends in the apparent AoA. These trends differ from the trends in the “ideal-age AoA”, and have no direct correspondence to the actual trends in the atmospheric circulation.

Procedures used to derive the AoA from observations of various tracers in the atmosphere are inevitably based on assumptions and idealisations that have limited and often unknown area of applicability. The resulting uncertainties in the AoA are large enough to preclude the use of apparent AoA and its trends for evaluation of changes in atmospheric circulation or for validation of atmospheric models. Observations of the tracers themselves, however, have ~~quite~~ well quantified uncertainties, so direct comparisons of simulated tracers to the observed ones are a very promising means for the atmospheric model evaluation. AoA in turn is a convenient means for model inter-comparison if a protocol of the AoA derivation is well specified.

Code and data availability. The SILAM source code is freely available from GitHub (<https://github.com/fmidev/silam-model>). The simulation results are available from MS or RK on request. The MIPAS observational data available from GS on request. ERA-Interim and ERA5 reanalyses data sets are available from the European Center for Medium-range weather forecast <http://www.ecmwf.int>.

Author contributions. RK performed the simulations and data analyses, prepared text and illustrations. MS and JV inspired the study and helped with discussions on content and structure of the study, and participated in editing the text. GS provided MIPAS data and wrote sections about MIPAS observations. All authors participated in the final preparation of the text.

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